



BOOK OF ABSTRACTS 7-11 DECEMBER | Virtual

The Joint European Magnetic Symposia 2020

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Special Technical Session: building bridges with magnetic technology

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Plenary Sessions

2D Magnetic Materials

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The first 2D magnetic material in which ferromagnetism has been shown experimentally to persist down to individual monolayers has been reported less than three years ago. Since then a number of different experiments have been performed on atomically thin magnets of different types, and led to interesting observations (giant tunneling magneto resistance, gate-tuning of the magnetic state, strong exchange bias at van der Waals interfaces, etc.). In my talk I will give a short introduction to this rapidly evolving domain of research, and discuss results obtained in my group on atomically thin multilayers of materials such as CrI₃, CrCl₃ (layered antiferromagnets) and MnPS₃ (antiferromagnetic within individual layers). Most of our experiments focus on investigations of the magnetic properties of atomically thin crystals of these materials by using them to form tunnel barriers and by measuring their tunneling magnetoresistance. As I will show, tunneling magnetoresistance allows tracing the boundaries between different magnetic phases, and it can therefore be used to establish magnetic phase diagrams. Specific phenomena that I will discuss include the observation of a giant tunneling magnetoresistance in CrI₃, a complete analysis of the magnetic phases of multilayers of CrCl₃ as a function of thickness, magnetic field and temperature, and the observation of a spin-flop transition in MnPS₃ persisting to the ultimate thickness of an individual monolayer.

STT-MRAM adoption in microelectronics: what can we expect next?

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The adoption of the Spin-transfer Torque Magnetic Random Access Memory (STT-MRAM) technology by the main microelectronics industrial actors constitutes a major achievement of spintronics RD. Thanks to its unit combination of assets, MRAM can be used for applications that alternative technologies of non-volatile memory cannot address, particularly CMOS voltage compatibility and its write endurance (much larger than in Phase-change RAM or resistive-RAM). In fact, STT-MRAM are nowadays introduced in chips as replacement of embedded FLASH since MRAM fabrication is much simpler at advanced technology node. Facing speed and power restriction of STT mechanism, Spin-orbit torque (SOT) MRAM has emerged as a credible next-generation MRAM technology targeting replacement of SRAM and offering a better footprint than CMOS-based SRAM. Beyond, more advanced MRAM family concepts, based on voltage control of anisotropy (VCMA), and interconversion between spin and charge current may open the route towards very low power applications.

To fully benefit from the unique properties brought by spintronic components, and considering the increasing amount of data that have to be processed in modern applications, the architecture of electronic circuits should be deeply revisited with a much finer interpenetration between memory and logic blocks. Complete new approaches can be envisioned such as in-memory computing or massively parallel approach as in neuromorphic architecture. On the route towards these new concepts and applications, I will discuss the many challenges that must be overcome, involving innovative materials (providing larger tunnel magnetoresistance, larger spin Hall angle, larger VCMA coefficient, enhanced thermal stability, reduced thermal variation), improved processes (reduced dot to dot variability, patterning at sub-20nm feature size and small pitch, process compatibility of innovative materials) and new architectural development (revised design tools and concepts) [1].



[1] B.Dieny et al, Opportunities and challenges for spintronics in the microelectronics industry, Nature electronics, 3, 446 (2020)

All-optical manipulation of magnetism: from fundamentals to brain-inspired computing

Theo Rasing

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The explosive growth of digital data use and storage has led to an enormous rise in the energy consumption of Information and Communication Technology (ICT), which already stands at 7% of the global electricity consumption¹. New ICT technologies, such as Artificial Intelligence and the Internet of Things push this exponentially increasing energy requirement even more, though the underlying hardware paradigm is utterly inefficient: tasks like pattern recognition can be performed by the human brain with only 20W, while conventional (super)computers require 10 MW. Therefore, the development of radically new physical principles that combine energy-efficiency with high speeds and high densities is crucial for a sustainable future. One of those is the use of non-thermodynamic routes that promises orders of magnitude faster and more energy efficient manipulation of bits². Another one is neuromorphic computing, that is inspired by the notion that our brain uses a million times less energy than a supercomputer while, at least for some tasks, it even outperforms the latter. In this talk, I will discuss the state of the art in ultrafast manipulation of magnetic bits and present some first results³ to implement brain-inspired computing concepts in magnetic materials that operate close to these ultimate limits.



Figure 1: Using circularly polarized femtosecond laser pulses, artificial synapses can be constructed from magnetic materials, the magnetization of which can be optically controlled.

- [1] Lannoo, B. Energy consumption of ICT Networks. TREND Final Workshop Brussels (2013)
- [2] A. Kirilyuk, A. V. Kimel and Th. Rasing, Ultrafast optical manipulation of magnetic order, Rev. Mod. Phys. 82, 2731-2784 (2010)
- [3] A. Chakravarty, J.H. Mentink, C. S. Davies, K. Yamada, A.V. Kimel and Th. Rasing, Supervised learning of an optomagnetic neural network with ultrashort laser pulses, Appl. Phys. Lett. 114, 192407 (2019)

Magnetic Weyl Semimetals!

Claudia Felser

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Topology a mathematical concept became recently a hot topic in condensed matter physics and materials science. One important criteria for the identification of the topological material is in the language of chemistry the inert pair effect of the s-electrons in heavy elements and the symmetry of the crystal structure [1]. Beside of Weyl and Dirac new fermions can be identified compounds via linear and quadratic 3-, 6- and 8- band crossings stabilized by space group symmetries [2]. In magnetic materials the Berry curvature and the classical Anomalous Hall effect helps to identify interesting candidates. Magnetic Heusler compounds were already identified as Weyl semimetals such as Co_2YZ [3,4], Mn_3Sn [5,6,7] and $Co_3Sn_2S_2$ [8-10].

The Anomalous Hall angle helps to identify even materials in which a Quantum Anomalous Hall Eeffectshould be possible in thin films. First evidence for a QAH in $Co_3Sn_2S_2$ will be discussed. Besides this k-space Berry curvature, Heusler compounds with non-collinear magnetic structures also possess real-space topological states in the form of magnetic antiskyrmions, which have not yet been observed in other materials [11].

- [1] Bradlyn et al., Nature 547 298, (2017) arXiv:1703.02050
- [2] Bradlyn, et al., Science 353, aaf5037A (2016).
- [3] Kübler and Felser, Europhys. Lett. 114, 47005 (2016)
- [4] Belopolski, et al., Science 365, 1278 (2019), arXiv:1712.09992
- [5] Kübler and Felser, EPL 108 (2014) 67001 (2014)
- [6] Nayak, et al., Science Advances 2 e1501870 (2016)
- [7] Nakatsuji, Kiyohara and Higo, Nature 527 212 (2015)
- [8] Liu, et al. Nature Physics 14, 1125 (2018)
- [9] Liu, et al., Science 365, 1282 (2019)
- [10] Morali, et al., Science 365, 1286 (2019) arXiv:1903.00509
- [11] Nayak, et al., Nature 548, 561 (2017).

Magnetic Particle Imaging: Physical Principles and Medical Applications

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Magnetic Particle Imaging (MPI) is a versatile tomographic medical imaging technique that uses iron-oxide based tracers in order to determine the spatial distribution of the particle concentration. In this talk the basic principles of MPI are introduced and the most important milestones of MPI since its invention in 2001 are reviewed. Starting with the physical basics and the used static and dynamic magnetic fields, which are responsible for spatial encoding, an overview of the MPI signal chain is given. The resulting model of the imaging process can be expressed as a linear system of equations that relates the particle concentration and the signal detected in the receiving coils. For the determination of the particle concentration, an ill-conditioned inverse problem has to be solved, for which different reconstruction methods are presented. New reconstruction methods are also discussed, which enable the simultaneous reconstruction of several quantitative parameters like temperature, viscosity and binding states in addition to the particle concentration. Finally, numerous medical applications are presented in which MPI has already demonstrated its potential in preclinical studies. In addition to diagnostic applications such as hyperthermia will be presented.

Semi-Plenary Sessions

Advanced Multi-degrees of freedom Electromagnetic Actuators

Elena Lomonova TU Eindhoven

In high-precision and robotics systems, advanced actuator topologies and novel actuator systems with multiple degrees-of-freedom (e.g. motion and magnetic levitation) which combine an ultra-high position accuracy with very demanding motion profiles is the main research subject. Strong contributions are in the electromagnetic modeling, design optimization, and experimental validation of advanced and unique motors and actuators are discussed. The modeling is strongly oriented on fast harmonic modeling techniques and include mode-matching techniques and methods derived from rigorous coupled-wave analysis to consider complex shaped iron structures and position dependent material properties (e.g. conductivity and permeability). These methods are applied in both 2-D and 3-D geometries in Cartesian, cylindrical and spherical coordinate systems. Besides the harmonic methods also fast hysteretic actuator models and surface charge models have been further developed. These models are of large importance as they offer significantly shorter simulation times than FEM and allow to analyze, optimize and design motors with three-dimensional structures and multiple degrees-of-freedom such as magnetically levitated and elevated planar motors, linear-rotary systems and spherical actuators with integrated energy transfer sand gravity compensation. The short-simulation time, not only allows to effectively optimize these novel machines, but also to have fast multi-physical models which include thermodynamics, mechanics (deformation) and electric fields.

2D mutually synchronized spin Hall nano-oscillator arrays for highly coherent microwave signal generation and neuromorphic computing

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Mutually synchronized spin torque nano-oscillators (STNOs) are one of the promising platforms for bioinspired computing and microwave signal generation [1, 2]. Using STNOs one can achieve 90% recognition rate in spoken vowels [3]. However, in order to do more complex tasks, larger scale synchronized oscillators are needed, something that is not easily done with the STNOs demonstrated so far.

In my talk, I will describe a different type of spin current driven device called spin Hall nano-oscillators (SHNOs), which can generate microwave frequencies over a very wide frequency range [4]. The SHNOs are based on 50 - 120 nm wide nano-constrictions in Pt(5)/Hf(0.5)/NiFe(3) trilayers (all numbers in nm). When multiple nano-constrictions are fabricated close to each other (300 - 1200 nm separation) they can mutually synchronize and chains of up to nine nano-constrictions have been demonstrated to exhibit complete synchronization [5]. For the first time, we can now also synchronize two-dimensional SHNO arrays with as many as 8 x 8 = 64 SHNOs [6]. The mutual synchronization is observed both electrically and using scanning micro-BLS microscopy. Both the output power and linewidth of the microwave signal improves substantially with increasing number of mutually synchronized SHNOs, such that quality factors of about 170,000 can be reached. Following the approach of Romera et al [3], we also demonstrate neuromorphic computing using a 4 x 4 SHNO array with two injected microwave signals as inputs.

Given their high operating frequency (~10 GHz), easy fabrication, and highly robust synchronization properties, nano-constriction SHNO arrays are likely the most promising candidates for neuromorphic computing based on oscillator networks.

- [1] J. Grollier, D. Querlioz, and M. D. Stiles, Proc. IEEE 104, 2024 (2016).
- [2] J. Torrejon et al, Nature 547, 428 (2017)
- [3] M. Romera et al, Nature 563, 230–234 (2018)
- [4] T. Chen *et al*, Proc. IEEE 104, 1919 (2016)
- [5] A. A. Awad *et al*, Nature Physics 13, 292–299 (2017)
- [6] M. Zahedinejad, et al. Nature Nanotechnology 15, 47 (2020)

Antiferromagnetic Insulatronics: Spintronics without magnetic fields and moving electrons

Mathias Kläui

Johannes Gutenberg University Mainz

While known for a long time, antiferromagnetically ordered systems have previously been considered, as expressed by Louis Néel in his Nobel Prize Lecture, to be "interesting but useless". However, since antiferromagnets potentially promises faster operation, enhanced stability with respect to interfering magnetic fields and higher integration due to the absence of dipolar coupling, they could potentially become a game changer for new spintronic devices. The zero net moment makes manipulation using conventional magnetic fields challenging. However recently, these materials have received renewed attention due to possible manipulation based on new approaches such as photons [1] or spin-orbit torques [2].

We recently realized switching in the metallic antiferromagnet Mn₂Au [3] by intrinsic staggered spin-orbit torques [4] and characterize the switching properties by direct imaging.

While switching by staggered intrinsic spin-orbit torques in metallic AFMs requires special structural asymmetry, interfacial non-staggered spin-orbit torques can switch multilayers of many insulating AFMs capped with heavy metal layers.

We probe switching and spin transport in selected collinear insulating antiferromagnets, such as NiO [5,6], CoO [7] and hematite [8,9]. In NiO we find that there are multiple switching mechanisms that result in the reorientation of the Néel vector and additionally effects related to electromigration of the heavy metal layer can obscure the magnetic switching signal [5].

For the spin transport, spin currents are generated by heating as resulting from the spin Seebeck effect and by spin pumping measurements and we find in vertical transport short (few nm) spin diffusion lengths [6,7].

For hematite, however, we find in a non-local geometry that spin transport of tens of micrometers is possible [8,9]. We detect a first harmonic signal, related to the spin conductance, that exhibits a maximum at the spin-flop reorientation, while the second harmonic signal, related to the Spin Seebeck conductance, is linear in the amplitude of the applied magnetic field [8]. The first signal is dependent on the direction of the Néel vector and the second one depends on the induced magnetic moment due to the field. We identify the domain structure as the limiting factor for the spin transport. From the power and distance dependence, we unambiguously distinguish long-distance transport based on diffusion [8,9] from predicted spin superfluidity that can potentially be used for logic [10].

[1] A. Kimel et al., Nature 429, 850 (2004).

- [2] J. Zelezny et al., Phys. Rev. Lett. 113, 157201 (2014); P. Wadley et al., Science 351, 587 (2016).
- [3] M. Jourdan et al., J. Phys. D: Appl. Phys. 48, 385001 (2015).
- [4] S. Bodnar et al., Nature Commun. 9, 348 (2018)
- [5] L. Baldrati et al., Phys. Rev. Lett. 123, 177201 (2019)
- [6] L. Baldrati et al., Phys. Rev. B 98, 024422 (2018); L. Baldrati et al. Phys. Rev. B 98, 014409 (2018)
- [7] J. Cramer et al., Nature Commun. 9, 1089 (2018)
- [8] R. Lebrun et al., Nature 561, 222 (2018).
- [9] A. Ross et al., Nano Lett. 20, 306 (2020).
- [10] Y. Tserkovnyak et al., Phys. Rev. Lett. 119, 187705 (2017).

CrI3, a wonder 2D ferromagnet

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The observation in 2017 [1] of ferromagnetic order in a standalone monolayer obtained from exfoliation of CrI3, a layered bulk ferromagnetic insulator, marked the beginning of the new research field of Van der Waals ferromagnetism 2D crystals. In this talk I will discuss, from the theory stand-point, the outstanding properties that make CrI₃, and the related compounds CrBr₃ and CrCl₃, a wonder family of 2D Ferromagnets [2]. First, the magnetic anisotropy of CrI₃ is large enough to make magnetic order robust in the 2D limit [3]. Second, the interlayer exchange in CrI₃ thin films becomes antiferromagnetic [1,3]. This makes CrI₃ bilayers natural spin valves [4] that have demonstrated very large tunnel magnetoresistance in spin-filter magnetic tunnel junctions. CrI₃ can produce magnetic proximity effect in other materials, which can be used for new device concepts [5]. Our first-principles calculations show that exciton binding energy is particularly strong in CrX₃ (X=I, Br, Cl) and the Kerr effect is dominated by the spin orbit of the ligand. Finally, the honeycomb lattice formed by Cr in CrI₃ results in Dirac magnons with a topological gap and chiral magnon edge states².



Figure 1: Taken from [2]: Left: Spin filter Magnetic tunnel junction. Center: Van der Waals spin valve. Right: Quantized anomalous Hall induced by spin proximity.

[1] Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit, B Huang, G Clark, E Navarro-Moratalla, DR Klein, R Cheng, KL Seyler, et al., Nature 546 (7657), 270-273

[2] Magnetic Two-Dimensional Chromium Trihalides: A Theoretical Perspective, D. Soriano, M. I. Katsnelson, and J. Fernández-Rossier, Nano Letters 2020, 20, 9, 6225–6234

[3] Interplay between interlayer exchange and stacking in Crl₃, D. Soriano, C. Cardoso, J. Fernández-Rossier, Solid State Communications 299, 113662 (2019)

[4] *Probing magnetism in 2D van der Waals crystalline insulators via electron tunneling*, D. R. Klein, D. MacNeill, J. L. Lado, D. Soriano, E. Navarro-Moratalla, K. Watanabe, T.Taniguchi, S. Manni, P. Canfield, J. Fernández- Rossier, P. Jarillo-Herrero, Science 360, 1218 (2018)

[5] Van der Waals spin valves, C. Cardoso, D. Soriano, N. A. García-Martínez, J. Fernández- Rossier, Phys. Rev. Lett. 121, 067701 (2018)

[6] Magneto-optical response of Chromium trihalide monolayers: chemical trends, A. Molina-Sánchez, G. Catarina, D. Sangalli, J. Fernández-Rossier, J. of Mat. Chemi. C, 2020,8, 8856-8863

[7] Topological Magnons in CrI3 monolayers: an itinerant fermion description, A. Costa, D. Santos, N. M. R. Peres, J. Fernández-Rossier, 2D Materials (July 2020)

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Exploring antiferromagnetic order at the nanoscale with a single spin microscope

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Experimental methods allowing for the detection of single spins in the solid-state, which were initially developed for quantum information science, open new avenues for the development of highly sensitive quantum sensors. In that context, the electronic spin of a single nitrogen-vacancy (NV) defect in diamond can be used as an atomic-sized magnetometer, providing an unprecedented combination of spatial resolution and magnetic sensitivity under ambient conditions. In this talk, I will illustrate how scanning-NV magnetometry can be used as a powerful tool for exploring condensed-matter physics, focusing on chiral spin textures in antiferromagnetic materials.

Magnetic Spin Textures in Thin-Film Heterostructures

Suzanne G.E. te Velthuis

Materials Science Division, Argonne National Laboratory

Topologically protected chiral spin textures, such as magnetic skyrmions, that exhibit quasi-particle like behavior have been envisioned to enable low-power information technologies, and consequently have engaged considerable the interest of the scientific community in recent years [1]. To this end, inversion asymmetric multilayers consisting of ferromagnetic and heavy metal layers have been explored and have been shown to host chiral domain walls and stabilize magnetic skyrmions due to interfacial Dzyaloshinskii-Moriya interactions. The controlled generation and manipulation of these spin structures is key to harnessing their potential in applications. We have shown that diverging electric charge currents can locally generate skyrmions. The diverging currents can be created by making use of geometric constrictions [2,3], and from nonmagnetic conducting point contacts [4]. Due to spin orbit torques at the interfaces, the spin textures can also be manipulated by charge currents. Magnetic skyrmions have been shown to exhibit a transverse motion relative to the current direction, i.e., the skyrmion Hall effect [5]. This effect arises due to the non-trivial topological charge of the skyrmions and is the analogue of the ordinary Hall effect for electrical charges in the presence of a magnetic field. The skyrmion Hall effect should vanish for antiferromagnetic skyrmions because of the cancelation of opposite topological charges and likewise for ferrimagnets at the compensation temperature. To explore this idea, we have investigated the current driven motion in artificially ferrimagnetic multilayers consisting of Co and Gd layers. We have studied the temperature dependence of the motion from room temperature down to temperatures below the compensation point of around 100 K and find a dependency of the skyrmion Hall angle and domain wall speed on the applied temperature.

[1] W. Jiang, et al., Physics Reports **704**, 1 (2017).

- [2] W. Jiang, et al., Science 349, 283 (2015).
- [3] O. Heinonen, et al., Phys. Rev. B 93, 094407 (2016).
- [4] Z. Wang, et al., Phys. Rev. B 100, 184426 (2019).
- [5] W. Jiang, et al., Nature Phys. 13, 162 (2017).

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Operating quantum states in individual magnetic molecules

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The endeavour of quantum electronics is driven by one of the most ambitious technological goals of today's scientists: the realization of an operational quantum computer. We start to address this goal by the new research field of molecular quantum spintronics, which combines the concepts of spintronics, molecular electronics and quantum computing. The building blocks are magnetic molecules, i.e. well-defined spin qubits. Various research groups are currently developing low-temperature scanning tunneling microscopes to manipulate spins in single molecules, while others are working on molecular devices (such as molecular spin-transistors and carbon-nanotube based devices) to read and manipulate the spin state and perform basic quantum operations. We will present our recent measurements of geometric phases, the iSWAP quantum gate. the coherence time of a multi-state superposition, and the application to Grover's algorithm [1-5].



Figure 1: A molecular spin transistor based on a single TbPc₂ molecular magnet.

[1] S. Thiele, F. Balestro, R. Ballou, S. Klyatskaya, M. Ruben, W. Wernsdorfer, Science, 2014, 344, 1135.

[2] M. Ganzhorn, S. Klyatskaya, M. Ruben, W. Wernsdorfer, Nature Nanotechnol., 2013, 8, 165; Nature Comm., 2016, 7, 11443.

[3] M. N. Leuenberger, D. Loss, Quantum computing in molecular magnets. Nature 410, 789-793(2001).

[4] C. Godfrin, A. Ferhat, R. Ballou, S. Klyatskaya, M. Ruben, W. Wernsdorfer, F. Balestro, Phys. Rev. Lett. 119, 187702 (2017).

[5] C. Godfrin, et al.,. npj Quant. inf. 4, 53 (2018).

Artificial neural networks with analog or probabilistic spintronics devices

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There have been increasing demands on realizing computing hardware capable of addressing complex tasks that classical von-Neumann computers cannot readily execute. Here I give an overview of the hardware technology for computationally hard problems and discuss the potential and current status of the spintronics technology in this field [1,2]. I then describe our studies to create artificial neural networks using analog or probabilistic spintronics devices [2-7].

I show material and device technologies to realize artificial synapses and neurons constructed from antiferromagnet/ferromagnet structures operated by the spin-orbit torque [2-5] and a proof-of-concept demonstration of associative memory operation based on the Hopfield model [5,6]. I also present a rudimentary probabilistic computer with binary stochastic neurons made of stochastic magnetic tunnel junction, which has a similar functionality with quantum annealing machines and is capable of addressing computationally hard problems such as combinatorial optimization [7].

The works have been carried out in collaboration with H. Ohno, W. A. Borders, A. Kurenkov, team members of S. Sato, Y. Horio, S. Datta, P. Gambardella, and J. Åkerman. The works are partly supported by RD Project for ICT Key Technology of MEXT, ImPACT Program of CSTI, JSPS KAKENHI No. 17H06093, 19H05622, and JST-CREST No. JPMJCR19K3.

- [1] J. Grollier, D. Querlioz, K. Y. Camsari, K. Everschor-Sitte, S. Fukami, and M. D. Stiles, *Nature Electronics*, **3**, 360 (2020).
- [2] A. Kurenkov, S. Fukami, and H. Ohno, Journal of Applied Physics, 128, 010902 (2020).
- [3] S. Fukami, S. DuttaGupta, C. Zhang, A. Kurenkov, and H. Ohno, Nature Materials, 5, 535 (2016).
- [4] A. Kurenkov, S. DuttaGupta, C. Zhang, S. Fukami, Y. Horio, and H. Ohno, Advanced Materials, **31**, 1900636 (2019).
- [5] S. Fukami and H. Ohno, Journal of Applied Physics, 124, 151904 (2018).
- [6] W. A. Borders, H. Akima, S. Fukami, S. Moriya, S. Kurihara, S. Sato, and H. Ohno, *Applied Physics Express* **10**, 013007 (2017).
- [7] W. A. Borders, A. Z. Pervaiz, S. Fukami, K. Y. Camsari, H. Ohno, and S. Datta, Nature, 73, 390 (2019).

Coherent nanomagnonics based on thin yttrium iron garnet

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Thin ferrimagnetic yttrium iron garnet (YIG) films have generated large interest in the field of nanomagnonics. Their long relaxation times and large decay lengths (1,2) put a new spin on grating couplers, artificial crystals and quasicrystals. For them, coherent scattering of spin waves (magnons) is essential for the design of efficient microwave-to-magnon transducers and nanoscale microwave filters based on tailored magnon band structures. We have created and studied grating couplers (3), magnonic crystals and quasicrystals (4) based on insulating YIG thin films. Using broadband microwave spectroscopy, inelastic light scattering, micromagnetic simulations and magnetic imaging techniques based on magnetic force microscopy and x-ray magnetic circular dichroism we investigated one-dimensional and two-dimensional periodic lattices and aperiodic tilings (such as Penrose and Ammann tilings) prepared from YIG combined with different nanostructured ferromagnets. In contrast to recently explored all-metal based quasicrystals (5) the YIG-based structures allow us to resolve clear signatures of magnon coherency such as (partial) forbidden frequency gaps.

The work has been performed in cooperation with K. Baumgaertl, M. Bechtel, V.S. Bhat, P. Che, C. Dubs, J. Förster, J. Gräfe, A. Mucchietto, G. Schuetz, N. Träger, S. Watanabe, and M. Weigand. It is funded via SNF under grant numbers 163016 and 171003.

References

- [1] H. Yu et al., Sci. Rep. 4, 6848 (2014).
- [2] S. Maendl, I. Stasinopoulos, and D. Grundler, Appl. Phys. Lett. 111, 012403 (2017).
- [3] K. Baumgaertl et al., unpublished.
- [4] S. Watanabe et al., unpublished.
- [5] S. Watanabe, V.S. Bhat, K. Baumgaertl, and D. Grundler, Adv. Funct. Mater. (2020),

https://doi.org/10.1002/adfm.202001388.

Imaging phase transitions with scanning SQUID

Beena Kalisky

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We use a local magnetic imaging technique, scanning SQUID microscopy, to map the spatial distribution of electronic states near surfaces and interfaces. We track conductivity, superconductivity and magnetism in systems undergoing phase transitions, where the local picture is particularly meaningful.

I will describe two recent findings: (1) Conduction landscape that changes dramatically near the metal to insulator transition at the 2D $LaAlO_3/SrTiO_3$ interface. (2) Non-trivial behavior of superconducting fluctuations near the superconductor to insulator transition in NbTiN. The local view allows us to investigate the way local features control the behavior of each system.



Figure 1: Scanning SQUID (a, illustration) maps of localized regions of reduced conductivity (b), fluctuating superfluid density (c), modulated current flow over channels (d), electric response to stress (e), gate tunable conductivity (f), and static magnetic landscape - here vortices in superconductors (g). Scale bars are 3µm for c, and 30µm otherwise.

The long way to magnetic materials design run it fast

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The development of novel materials is a strong enabler for any technology, to the point that often technology and materials innovation cannot be separated. Unfortunately the process of finding new materials, optimal for a given application, is lengthy, often unpredictable and has a low throughput. Here I will describe a systematic pathway to the discovery of novel compounds, which demonstrates an unprecedented throughput and discovery speed. The method can be applied to any materials class and any potential application. I will use the example of magnetism to introduce the main features of the method, and I will demonstrate the discovery of several new high-performance magnets. Furthermore, I will highlight how such high-throughout schemes can be combined with machine-learning methods for data-mining to extract novel materials designing rules and for identifying new prototypes for further investigation. Based on an extensive electronic structures library of Heusler alloys containing 236,115 prototypical compounds, we have filtered those alloys displaying magnetic order and established whether they can be fabricated at thermodynamical equilibrium [1]. Specifically, we have carried out a full stability analysis for in- termetallic Heuslers made only of transition metals. Among the possible 36,540 prototypes, 248 are found thermodynamically stable but only 20 are magnetic. The magnetic ordering temperature, T_c , has then been estimated by a regression calibrated on the experimental T_c of about 60 known compounds. As a final validation we have attempted the synthesis of a few of the predicted compounds and produced two new magnets. One, Co2MnTi, displays a remarkably high T_c in perfect agreement with the predictions, while the other, Mn2PtPd, is a complex antiferromagnet. In the second part of my talk I will discuss the use of machine-learning methods for predicting the Curie temperature of ferromagnets, based solely on their chemical composition (see figure) [2], and for sorting magnets into hard and soft. In particular I will discuss how to develop meaningful feature attributes for magnetism and how these can be informed by experimental and theoretical results. I will also show a potential promising scheme for extracting information from published literature. All these together can pave the way for the large-scale design of novel magnetic materials at unprecedented speed.



Figure 1: T_c prediction as a function of composition for the ternary system Al-Co-Fe. Data are presented as a function of the atomic fraction of the three species and the T_c is expressed as a heat map. The figure also introduces a detailed analysis of the three relevant binary phase diagrams, where the blue line traces the ML prediction, black crosses (green dots) are experimental points included (not included) in the training set. The light-blue shadowed area in the binary plots corresponds to the range of predicted T_c 's, namely it indicates the uncertainty of the machine-learning model. The solid square (circles) included in the ternary T_c diagram are for experimental data included (not included) in the training set, with the colour code describing the T_c . Numbers correspond to four known stoichiometric phases: 1) Co₂FeAl, $T_c = 1,000$ K, 2) Fe₂CoAl, $T_c > 873$ K, 3) Fe₄CoAl, $T_c = 420$ K, 4) Fe₃Al, $T_c = 573$ K.

[1] S Sanvito, C Oses, J Xue, Anurag, Tiwari, M Zic, T Archer, P Tozman, M Venkatesan, J. M D. Coey and S Curtarolo, *Accelerated discovery of new magnets in the Heusler alloy family*, Science Advances **3**, e1602241 (2017).

[2] J Nelson and S Sanvito. *Predicting the Curie temperature of ferromagnets using machine learning*. Phys. Rev. Mat. **3**, 104405 (2019).

Special Technical Session: Building bridges with magnetic technology

Spintronic-photonic integrated circuit platform for memory applications

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There is a growing gap between memory and processor performance in computing, which is expected to increase even further when new computing paradigms are considered [1]. Relevant metrics are density, latency, energy consumption and bandwidth. The target values for these parameters depend on the position in the memory hierarchy. A promising approach is to use magnetic memory integrated on a chip, thereby combining non-volatility, i.e., low static power consumption, with relatively high bandwidths and integration density [2]. One implementation is magnetic random-access memory (MRAM), where bits are stored as the magnetization direction of a free layer. By stacking this free layer on a fixed magnetized layer, the memory state is defined by parallel or anti-parallel magnetization, i.e., 'low' or 'high' resistance, respectively. In spin transfer-torque (STT) MRAM, such states can be written by a current. An issue is that the write current is still relatively high, though.

An alternative way of switching a magnetic layer is by optical means, i.e., the use of a short optical pulse [3]. It was first observed that a layer could be deterministically switched by changing the polarization of a sub-picosecond optical pulse, i.e., depending on the chirality of its circular polarization. This is a subset of a wider class of switching phenomena, where magnetic layers can be switched by pulses with arbitrary polarization and durations up to a few picoseconds, and with a large enough energy, i.e., switching fluence. However, this approach is not compatible with mature CMOS-compatible processing of current memory technologies.

The drive towards integration of CMOS-based electronics and photonics is an ongoing effort, driven by the need for bandwidth and bandwidth density, and reduced energy consumption of interconnects [4]. Theoretical calculations have shown that energy consumption can be in the tens of femtojoule-per-bit range for such optical networks-on-chip (NoCs). The first commercial implementation for off-chip interconnects is currently being developed by Ayar Labs.

The European FET-open project SPICE – Spintronic-Photonic Integrated Circuit platform for future Electronics – aims for the convergence of MRAM technology with photonics in a fully integrated technology, thereby enabling all-optical writing of MRAM at unprecedented bandwidths and energy-efficiencies. To achieve this, we are addressing four key challenges. First we need to develop optically-switchable magnetic layers that can be used in a magnetic tunnel-junction (MTJ). Secondly, this layer stack needs to be embedded in MTJ elements that are optically accessible, i.e., have a transparent contact. Thirdly, an optical NoC has to be designed that can switch the optical pulses to the MTJ elements to be written. And finally, the coupling between the optical NoC and the MTJ elements needs to be optimized.

In this talk I will give an update on this SPICE technology, showing our latest results, and the feasibility of using fully integrated optically-switched MRAM technology.



Figure 1.

[1] https://www.synopsys.com/designware-ip/technical-bulletin/building-efficient-deep-learning-dwtb_q318.html

[2] R. L. Stamps et al., J. of Phys. D: Appl. Phys. 47, 33, 333001 (2014)

[3] C. D. Stanciu et al., Phys. Rev. Lett. 99, 047601 (2007)

[4] D. A. B. Miller, Proceedings of the IEEE 97, 7, 1166-1185 (2009)

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Magnetostrictive thin film based strain and magnetic field sensors

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Magnetostrictive thin films offer attractive options as they transduce magnetic energy into mechanical energy or vice versa. Exploiting this property opens up new device concepts and applications. A prerequisite for the technical implementation, e.g. in highly sensitive sensors, is the compatibility of the different materials, often also the possibility of microsystem fabrication and the transfer of known concepts for magnetic thin-film systems into layer systems in the micrometer thickness range.

As a first example, magnetoelectric (ME) composites are presented. They consist of an amorphous magnetostrictive phase and a piezoelectric phase, which are fabricated by means of thin-film technology. Their strain-mediated ME coupling coefficient far exceeds the coupling strength in natural magnetoelectrics. Designed as simple bending beams, the ME coupling coefficient alpha_ME reaches up to 19 kV/cmOe by taking advantage of mechanical resonance effects [1]. Based on this high coupling strength, highly sensitive magnetic field sensors can be realized which typically reach a detection limit ranging from 1 pT to several 100 pT [2]. Magnetoelectric surface acoustic wave sensors [3] offer an alternative to bending beams, the latter being limited in bandwidth. Here, a 200 nm FeCoSiB layer functions as magnetostrictive phase. It is deposited on a quartz substrate inbetween two pairs of interdigital electrodes. Utilizing the Δ E effect, the phase change between input and output signal serves as a measure of the applied magnetic field. Depending on the ME sensor concept, specific magnetic multi-layer systems are discussed in the view of magnetic noise suppression [4] and control of sensor operating point [5].

A second interesting example are tunnel magnetoresistance junctions. In the context of magnetoresistive device fabrication magnetostrictive effects are often considered as undesired side-effects, because processing related mechanical stress can alter the magnetic characteristic of the finally packaged storage or sensing unit. Though, the targeted utilization of inverse magnetostriction opens the path to highly sensitive strain gauges with gauge factors exceeding GF = 2000 [6]. They have application potential as miniaturized pressure sensors or deflection sensors in atomic force microscopes (AFMs) [7]. In AFMs they would replace the classical optical detection method, enable the investigation of light sensitive samples, facilitate the use in vacuum systems and allow multi-frequency mode AFM.

[1] R. Jahns, et al., Am. Ceram. Soc. 96 (6), 1673–1681 (2013)

- [2] V. Röbisch, et al., J. Mater. Res. 32 (06), 1009–1019 (2017)
- [3] A. Kittmann, et al., Sci. Rep. 8 (1), 278 (2018)
- [4] M. Jovičević Klug, et al., Appl. Phys. Lett. 114 (19), 192410 (2019)
- [5] E. Lage, et al., Nat. Mater. **11** (6), 523–529 (2012)
- [6] A. Tavassolizadeh, et al., J. Magn. Magn. Mat. 384, 308-313 (2015)
- [7] A. Tavassolizadeh, et al., Appl. Phys. Lett. 102 (15), 153104 (2013)

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Bringing institute-level research to volume production

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It is no secret that many corporations like to look towards research institutes and universities to supercharge innovation, gaining access to some of the best minds on very advanced technologies. As is common with the 80/20 rule (80% of the effort is required to make the last 20% of progress), moving some of these concepts into volume production (millions of units), has its own unique challenges. This session will review several unique challenges that we face in validating the technical concept to fully meet the commercial need, the over operating envelope conditions performance, lifetime stability, manufacturability with high yield and cost effective, high-volume production. Perhaps unsurprisingly the earlier one starts to work on these challenges the more effective the institute / corporate collaboration is and the more rapid the results. We will review development & validation challenges and potential solutions with respect to metrology, DoE (Design of Experiments), process sensitivity / cornering analyses, and performing accelerated life testing on prototype level technology. Finally, we will cover challenges in pilot line and volume production and how early intervention can smooth this transition.

Symposium 1. Biomagnetism and Biomedical Applications

3993 Liquid flow and control without solid walls

Peter Dunne CNRS-IPCMS

Solid walls become increasingly important when miniaturizing fluidic circuitry [1]. They limit flow-rates achievable for a given pressure drop, and are plagued by fouling [2]. Approaches to reduce the wall interactions include hydrophobic coatings [3], liquid-infused porous surfaces [4], nanoparticle surfactant jamming [5], changing the surface electronic structure [6], electrowetting [7], surface tension pinning [8,9], and atomically flat channels [10]. A better solution may be to avoid the solid walls altogether.

Here we demonstrate a new approach¹¹, where wall-less aqueous liquid channels are surrounded by an immiscible magnetic liquid, both being stabilised by a quadrupolar magnetic field. This creates self-healing, uncloggable, antifouling, and near-frictionless liquid-in-liquid fluidic channels with millimetre effective slip lengths. Pumping is achieved by moving permanent magnets that have no physical contact with the liquid channel. We show that this magnetostaltic pumping method can be used to transport whole human blood with very little damage due to shear forces; haemolysis is reduced by an order of magnitude compared to traditional peristaltic pumping. Our liquid-inliquid approach provides new avenues to transport delicate liquids, particularly when scaling channels down to the micron scale with no need for high pressures, while retaining basic microfluidic circuitry functionalities.





Figure 1.: Right: exploded view, of permanent magnets (red, blue) in an in-plane quadrupolar configuration create a low-field zone at the centre **a**, where an antitube of water (yellow) is stabilized inside an immiscible magnetic liquid; **b** contour plot of the magnetic field; **c** X-ray transmission radiograph of a water antitube (yellow) surrounded by ferrofluid; **d** optical end-view of a water antitube in ferrofluid; Scale bars (black/white) are 2 mm.

Figure 2. : **a** Top-view X-ray images illustrating the mechanical rupture of a static tube using a spatula, followed by self-healing, returning to equilibrium, without being under flow, within minutes; **b** 0.6 mm glass beads inserted into a 1.5 mm antitube that can be expelled with a slight increase in applied pressure of 20 mbar; **c** a 2 mm diameter bead larger than the antitube diameter (d = 0.5 mm) does not cause clogging; d pressure and flow rate during the expulsion of the bead – there is only a small decrease in flow rate from 30 to 23 µl min⁻¹ when the bead is present.

- [1] Tabeling, P. Introduction to Microfluidics. (OUP Oxford, 2005).
- [2] Mukhopadhyay, R. Anal. Chem. 77, 429 A-432 A (2005).
- [3] Zhao, B., Moore, J. S. Beebe, D. J. Science 291, 1023–1026 (2001).
- [4] Wong, T.-S. et al. *Nature* **477**, 443–447 (2011).
- [5] Forth, J. et al. Adv. Mater. 1707603 (2018).
- [6] Secchi, E. et al. Nature 537, 210–213 (2016).
- [7] Banerjee, A., Kreit, E., Liu, Y., Heikenfeld, J. Papautsky, I. Lab. Chip 12, 758 (2012).
- [8] Lee, W. C., Heo, Y. J. Takeuchi, S. Appl. Phys. Lett. 101, 114108 (2012).
- [9] Walsh, E. J. et al. Nat. Commun. 8, 816 (2017).
- [10] Keerthi, A. et al. Nature 558, 420–424 (2018).
- [11] Dunne, P. et al., Nature accepted (2020). https://doi.org/10.26434/chemrxiv.7207001.v2

Vacancy-induced correlated disorder in colloidal nanocrystals: a favorable tweaking path for magnetism-engineered functionalities

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Colloidal magnetic nanoparticles designed for important biomedical applications, [1] such as magnetic hyperthermia treatment of tumors, occasionally require defect-free, large entities (>20 nm) due to their high capacity to dissipate heat in their near vicinity. However, this attribute may come with some drawbacks, including patient discomfort. Here, we combine nanochemistry, detailed characterization and theoretical Monte Carlo considerations to explore the relation of structural defects on the size and shape of iron-oxide nanocrystals (NCs) and how these couple to magnetic properties relevant to nanobiotechnology [2]. Connecting the compositional complexity with the underlying magnetism (Fig. 1a-b) during the oxidative conversion of rock-salt NCs to spinel type, allows insights on how controlled cation vacancy-induced disorder tailors physical properties, including exploitable energy transfer for small-size magnetic nanocarriers. Synchrotron X-ray total scattering (Fig. 1c) corroborates our idea that the size-dependent evolution of the metal-cation valence state, produces pinning defects which promote favorable magnetic exchange interactions at subcritical sizes (10 nm), and beyond the limitations of finite-size effects alone. In such small particles, optimal magnetic anisotropy is offered by imperfections that impede the coherent reversal and easy relaxation of magnetic moments, allowing a remarkable ten-fold rise of the nanomaterial's thermoresponsive capability as compared to that obtained by defect-free entities (Fig. 1d). We offer a description of atomic interaction effects and knowledge of the different length-scale mechanisms required to facilitate the performance of single-crystal nanoscale particles as functional nanoheaters (cf. theranostics).



Figure 1.: Oxidative conversion of 8.1 nm wüstite spherical NCs: **a** Fourier synthesis of HRTEM diffraction, points to defected (top) and defect-free (bottom) lattice planes; **b** Hysteresis loop and its derivative, reflect two switching field distributions (5 K); **c** Synchrotron X-ray pair-distribution function analysis suggests vacant tetrahedral Fe-sites; **d** Enhancement of Specific Absorption Rate (SAR) for defected vs. defect-free NCs (Monte Carlo calculation).

[1] D. Yoo et al., Acc. Chem. Res. 44, 863 (2011).

[2] A. Lappas et al., Phys. Rev. X 9, 041044 (2019).

MRI relaxivity properties of magnetoferritin as a pathological model system of native ferritin

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Various pathological processes, including neurodegenerative disorders, are associated with the accumulation of iron, while it is believed that a precursor of iron accumulation is ferritin. Physiological ferritin is due to low relaxivity only weakly detected by magnetic resonance imaging (MRI) techniques. On the other hand, pathological ferritin that is associated with disrupted iron homeostasis and structural changes in the mineral core causes the hypointensive artifacts in MRI. On the basis of recent regarding the pathological ferritin structure, we prepared the magnetoferritin particles as a suitable pathological ferritin model system. We studied the relaxivity properties of magnetoferritin with different loading factor (LF) in order to find the MRI methodology able to distinguish native ferritin form magnetoferritin as a model system of pathological ferritin. It is believed that pathological ferritin is associated with the unphysiological iron accumulation and mineralization that is connected with various disorders. Unfortunately, we still do not know whether the iron accumulation is the initial cause or consequence of pathology. MRI measurements were performed at 7 T BRUKER system using longitudinal (T₁) and transversal (T₂) relaxation time mapping protocols. Relative contrast, relaxation time, relaxation rate, and relaxivity of native ferritin and magnetoferritin with different loading factors were analyzed and compared. The results clearly show a significant difference between native ferritin and magnetoferritin in T₂-weighted protocols. The difference in the T₁-weighted protocol is also obvious but not as considerable. Finally, we compared the transversal and longitudinal relaxivity ratios that provide information about the dominant relaxation mechanism in the sample. The results show that this ratio provides a powerful tool in discrimination of native ferritin (physiological ferritin) from magnetoferritin, as a model system of pathological ferritin. We are convinced that these findings could significantly contribute to the exploitation of biogenic iron oxides nanoparticles accumulation for a noninvasive diagnostics of pathological processes related to disrupted iron homeostasis (e.g., neurodegenerative disorders, neuroinflammation, cirrhosis, etc.).

Viral RNA Detection of Zika, Dengue and Chikungunya using a portable magnetoresistive platform

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This work describes the use of magnetoresistive (MR) sensors coupled with a portable platform for the detection of neglected tropical diseases: Zika (ZIKV) and Dengue (DENV) viruses from the genus *Flavivirus* (FLAV) and Chikungunya virus (CHIKV) from the genus *Alphavirus*. These mosquito-borne infections have had an increase in geographic range over the last decades, following the geographic spread of their vector species, namely the invase mosquitos *Aedes albopictus* and *Ae. aegypti*. These infections share common symptoms that may lead to misleading clinical diagnoses and delay in the implementation of control and preventive measures.

Currently, the diagnosis relies either on molecular tests of viral RNA for an acute infection phase (<5-8 days) or on the host humoral immune response for an advanced infection phase (>5 days). The present work focus on the virus detection at the acute phase. Usually, a reverse transcription-polymerase chain reaction (RT–PCR) of patient blood or urine sample is performed [1]. However, it tends to be complex and expensive, requiring expert staff and fully equipped laboratories. The magnetic detection system used in this work was developed in the past years by INESC-MN and INESC-ID and is a portable and user-friendly device capable of detecting analytes in a fast and reliable manner [2,3]. A spin-valve based MR biochip [3] was functionalized with specific DNA oligo probes designed using Sequencer software. The target samples were provided by the reference laboratory (INSA-CEVDI) and were amplified by asymmetric PCR. Immobilization of the probes over the 6 sensing areas of the biochip was done by automatic spotting using a micro-array non-contact imprinting machine. The functionalized biochip was inserted in the MR platform and biotinylated target DNA was transported over the sensing area by a PDMS microfluidic channel. A proprietary strategy of on-chip current lines was employed to promote hybridization between the target and probes. Finally, streptavidin coated magnetic nanoparticles (MNPs) with a size of 250 nm were incubated on the chip during signal acquisition. These MNPs present a magnetic moment of 1.6 x 10-16 A.m² for a 1.2 kA/m magnetizing field and a susceptibility of $\chi \sim 5$. The measured output corresponds to the voltage variation (ΔV) before and after MNP addition. This value is then normalized by the sensor output ($\Delta V/V$) and is directly proportional to the number of target molecules hybridized to its complementary probe. With this MR sensor platform, tests were carried for detection of 3 different viral targets: ZIKV, DENV serotype 2 (DENV-2) and CHIKV. The ZIKV, DENV and CHIKV targets successfully hybridized with their complementary probes with signals of 2.5 %, 0.8% and 3.6%, respectively (Fig.1). The FLAV probe detected both ZIKV and DENV targets. The specificity of the signals obtained allows us to establish a threshold approach with a YES or NO result for the presence of the viruses in a sample. Further tests are being made in the optimization of the biochip functionalization to increase assay specificity. Testing of other arboviruses co-circulating in the same areas of endemicity are to be carried out to assess probe



Figure 1. : a Signal acquired in the MR platform for ZIKV, DENV-2 and CHIKV targets; b working principle for the magnetoresisitve detection.

[1] L. Zé-Zé et al., *IDCases* **4** (2016).

specificity.

- [2] J. Germano et al., Sensors 9 (2009).
- [3] V.C. Martins et al., Biosens Bioelectron 24 (2009).

Oral Presentation

Magnetoelastic immunosensors for the multiple detection of pathogens in foods

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Recently, in order to guarantee health and safety to an ever increasing number of people, the interest in the development of portable biosensing devices capable of detecting biological entities such as biomolecules and pathogens has grown exponentially, driven by the miniaturization of the electronic circuits that allows the concentration of laboratory equipment with high performance in chips of a few millimeters [1]. In this scenario, the spread of biosensing, which promises to be cheap, portable, fast and easy to use, it's gaining more and more attention [2].

In recent years, magnetoelastic materials (ME) have proven to be an interesting platform for the development of wireless sensing with high performance [3]. The main advantages of the resulting device are its cheapness, given the low production costs, and the possibility of creating a multiplexing scheme, considering the small size of the sensor [see Fig. 1(a)]. The operating principle is based on the Joule magnetostriction of the magnetic material, which can vibrate longitudinally at a characteristic frequency when subjected to a variable magnetic field. The adhesion of a small mass to the surface of the material involves a change of the resonance frequency, which can therefore be used as a sensing parameter.

In this work, we propose a ME-based biosensor with a measurement protocol that addresses the issue of the signal amplification by using gold nanoparticles (Au-NPs) functionalized with the appropriate antibody. In this way, in a sandwich configuration [see Fig. 1(a)], we are able to significantly amplify the effect of the antigen recognition. As shown in Fig. 1(a), the ME-based ribbon is coated with gold so that its surface as well as the Au-NPs can be functionalized by using a recently introduced Photochemical Immobilization Technique (PIT) [6]. PIT has proven to be a cheap, reliable and effective procedure for binding Abs upright on a gold surface making the ME-based technology flexible and easy to use for any type of analyte. Fig. 1(b) reports a sensorgram showing the entire measurement process: (i) sensor functionalization by antibodies, activated via PIT (green area), (ii) water rinsing for cleaning the fluidic circuit (blue area), (iii) bovine serum albumin (BSA), for blocking (violet area), (iv) target antigen flowing (human IgG) at a concentration of 5 μ g·ml⁻¹ (yellow area). (v) water rinsing (blue area), (vi) introduction of Au-NP, functionalized with human IgG produced in goat (pink area). Although preliminary, this result shows the possibility of detecting the target antigen at different concentrations, building a dose-response curve at the level of the state-of-the-art in biosensing, paving the way to the application of ME-based technology to food analysis in several contexts such as industrial food chain.



- [1] I. Giouroudi and G. Kokkinis. *Nanomaterials* **7**, 171 (2017).
- [2] B. Srinivasan and S. Tung., Journal of Laboratory Automation 20, 1 (2015).
- [3] C. Menti et al. Appl. Microbiol. Biotechnol. 100, 6149 (2016).
- [4] B. Della Ventura et al. Analyst 144, 6871 (2019).

Magnetic detection of neural activity in spinal cord slices

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ByAxon is devoted to the development of a new generation of sensors and electrodes based on nanotechnology materials for neural interfacing. We aim to design and build a prototype of an active implant that could work directly at the spinal cord (SC) level. This implant will be primary focused on restoring the transmission of electrical signals in the injured SC, acting as an active local bypass, something not possible with current technology. Further stimulation applications might include deep brain or retinal implants, among others. Current neural interfacing approaches are based on detecting electric potentials at the brain level, and/or triggering functional electrical stimulation (FES) through electrodes at muscular or SC levels [1]. However, these approaches present drawbacks such as the large number of cables and electrodes they require and, specially, the lack of sensory feedback. The ultimate non-contact sensing devices (magnetoencephalography) detect magnetic-field pulses generated by potentials at the brain, but require cryogenic temperatures, and, hence, are not portable. We will exploit here the enhanced properties of nanostructured materials to develop improved room temperature magnetoresistance-based high-resolution magnetic sensors [2]. Here we present the detection of neural signals in spinal cord slices (SCS) recording in real time the activity with no magnetic shielding, no time-series averaging, reference free, at room temperature and having a distance between sensor and tissue big enough to be noninvasive.

[1] Megan, L. Gill et al., Nature Medicine 24, 1677–1682 (2018).

[2] Caruso, L. et al., Neuron, 95, 1283–1291 (2017).

Oral Presentation

Micro and nanomagnets for biomedicine: from fabrication to cellular uptake

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Magnetic particles have attracted much attention in biomedicine by their physical properties to manipulate cells, release drugs at specific targets, develop novel diagnostics techniques, and envision promising therapies. A new kind of particle with great potential in all these fields is a disk-shaped nanomagnet. These nanomagnets are fabricated by top-down lithography techniques and present remarkable properties for biomedical applications, e.g., large magnetic moments and anisotropic shapes appropriate for mechanical stimulations [1]. This work presents micro and nano lithography processes to fabricate disks over large areas, covering sizes from few hundreds of nm to few microns. The structures are made of magnetic material in a vortex state, a flux closed spin configuration that shows zero remanence in magnetization curves. Magnets are released in water and purified for biomedical applications. The process was optimized to avoid the chemical degradation of particles and the consequent loss of magnetic properties [2]. The viability of micro and nanomagnets was evaluated by time-lapse microscopy and XTT assays of skin cancer cells. *In-vitro* cultures of tumoral cells and macrophages reveal the cellular uptake of nano and microdisks, even with feature sizes as large as four microns. Optical confocal microscopy and scanning electron microscopy images uncover details of the internalization process.

[1] L. Peixoto et al., Appl. Phys. Rev. 7, 011310 (2020).

[2] B. Mora et al., ACS Appl. Mater. Interfaces 10, 8165 (2018).

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Mixed sensors for medical imaging

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Since the initial proposal of mixed sensors, combining a superconducting loop and a spin electronics sensor, Giant Magneto-Resistance (GMR) or Tunnel Magneto-Resistance (TMR) devices [1], performance has been steadily improved by playing on the design of the superconducting loop to reduce the inductance for a given surface trapping and by improving the spin electronics sensors. Mixed sensors are now produced at wafer level.

The use of the manipulation of supercurrents by toggling them through different ways has allowed to partly suppress the 1/f noise of the devices.

The current performances reach a sub femtoTesla noise per sqrt(Hz) at frequencies above 10kHz, interesting for very low field Magnetic Resonance Imaging (MRI) devices and level of noise below 100fT/sqrt(Hz) noise at 1Hz, useful for magnetocardiography or magnetoencephalography measurements.

For MRI, we are coupling mixed sensors to flux-flux transformers with a room temperature part and a cold secondary coil which allows to optimize both the filling factor and the high sensitivity of that approach [2]. In that case, mixed sensors can be seen as cold amplifiers with less than 100pV/sqrt(Hz) of noise level.

We present here these recent improvements and the practical implementation of these devices in high performance very low field MRI for clinical applications.

[1] M. Pannetier et al., Science 304, 1648-1650 (2004).

[2] Q. Herreros et al., Review of Scientific Instruments 84, 095116 (2013).

Cantilever torque magnetometry on individual spherical Janus particles

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Magnetic nanoparticles are widely used in different fields of research and have (potential) applications in biomedicine, biomedical research as well as magnetic data storage and spintronics. To reliably control their motion, as for example in micro-fluidic experiments, the particles should ideally be in a well-known magnetic state to guarantee uniform behavior if exposed to static or dynamic external magnetic fields. A means to fabricate such particles in a controlled manner is to cover one half of silica spheres with a magnetic material. These so called Janus particles (JP) can show different magnetic configurations, such as vortex or onion states.

Due to their small size and thin magnetic capings, until now, magnetic measurements have been performed on large ensembles of JPs rather than on individual specimens. Interpretation of such measurements can be challenging, since the data represents an average over thousands of particles, and is further by excess magnetic material from the fabrication process, which can show different anisotropic behavior due to the difference in geometry.

Here, we present magnetic measurements of individual CoFe and IrMn/CoFe Janus particles, respectively, using sensitive dynamic cantilever magnetometry (DCM). For the measurements, each JP has been removed from its substrate and attached to the apex of an ultra-soft cantilever via a mechanical micromanipulator. The cantilever's response to the forces arising from the magnetic moment of the JP in an externally applied field allows to extract information on magnetic properties such as anisotropic behavior or the presence of exchange bias. Additionally, we performed micro magnetic simulations using the software package MUMAX³ in order to gain insight into the magnetic state of the JPs and to determine numeric values for the saturation magnetization Ms = 1.69×10^6 A/mand the demagnetization factor *D*= *-0.334*.

Optimizing Size Nanoparticle and Agglomeration on Magnetic Nanotags for Lateral Flow Immunoassays

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Lateral Flow Immunoassay (LFIA) is a paper-based architecture whose most well-known and popular use is the pregnancy test. They are being increasingly used for determination of biomarkers, allergenic pathogens, drugs and metabolites in biomedical, food safety and environmental settings [1]. Their sensitivity, selectivity, quickness and ease of use make them ideal for *Point-of-Use* (PoU) testing. One of the key points of the LFIAs is the labelling of the biomarker. This task has been done traditionally by latex or gold nanoparticles that provide a visible signal, perfect for a qualitative (presence/absence) analyses. To add quantification capacities to LFIAs, the use of Magnetic Nanoparticles (NPs) has been lately proposed [2].

The magnetic LFIAs must be associated to a magnetic reader that should be itself fast and portable. A radiofrequency inductive sensor has been developed for this purpose which takes advantage of the superparamagnetic character of the NPs [3,4]. The clue parameter of this technique is the initial magnetic permeability of the particles at the frequency of detection. The particle size has a crucial influence on the magnetic permeability, while the agglomeration degree determines the number of particles per molecule of interest. Thus, both size and agglomeration have a great importance in this type of test. The purpose of this work has been to elucidate the effects of the magnetic core size and particle agglomeration before and after the biofunctionalization process in the magnetic LFIA. To do so, three different iron oxide particles with core sizes of 8, 12 and 23 nm and different initial agglomeration have been evaluated as labels in LFIAs functionalized with neutravidin and immobilized by a biotin test line. We showed that agglomeration of the particles before running the LFA has an enormous positive influence on the inductive measurement because it increases the magnetic mass captured at the test line. We conclude that 12 nm particles agglomerated in clusters of 100 to 300 nm give the best results in inductively-read magnetic LFA.

[1] X. Huang et al., Biosens. Bioelectron. 75 (2016).

- [2] Lago-Cachón, D. et al., *IEEE Magn. Lett.* 8 (2017).
- [3] Lago-Cachón, D. et al., Nanotechnol. 24 (2013).
- [4] Rivas, M. et al., Sensor Actuar A-Phys. 216 (2014).

This work was supported by the Spanish Ministry of Economy and Competitiveness under projects MAT2017-84959-C2-1-R and EUIN2019-103338, the Council of Gijón-IUTA under grant SV-18-GIJON-1-27, and the Principality of Asturias under project IDI/2018/000185.

TMek: a lab on chip diagnostic test for selective capture and detection of magnetic fingerprints of malaria

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The most relevant methodologies for malaria diagnosis are the direct parasite investigation by microscopy examination and the indirect antigen detection implemented by rapid diagnostic tests (RDTs). Nevertheless, microscopy examination takes about 60 minutes and requires trained personnel, while RDTs suffer from a sizable number of false positive/negative results. Recently, we have developed a compact, low cost and easy to use diagnostic system, called TMek, which allows a pan-plasmodic and rapid malaria detection, as sensitive and accurate as microscopy.

The physical concept of the test exploits the paramagnetic property of infected erythrocytes and hemozoin crystals [1], the magnetic fingerprints of malaria common to all species, which allows them to undergo a selective magnetophoretic separation driven by a magnetic field gradient in competition with gravity. Upon separation, infected red blood cells (RBCs) and hemozoin crystals (HC) concentrate at the surface of a silicon microchip, where interdigitated electrodes are placed in close proximity to magnetic concentrators. Then, a change in resistivity, proportional to the amount of attracted particles, can be detected as an impedance variation by an electronic circuit, following a lab-on-chip approach.

In this paper we discuss the capability of our test to disentangle different corpuscles, tuning the competition between gravity and magnetic force by varying the experimental configuration and exploiting the peculiar dynamics of impedimetric signals in response to the application/removal of external magnetic fields.

The capability of TMek to perform the selective detection of infected RBCs and HCs has been tested by means of capture experiments on bovine RBCs treated with NaNO₂ to induce the full transformation of hemoglobin in methaemoglobin [2] and mimicking the behavior of malaria infected ones, and on suspensions of synthetic HCs. Thanks to the higher magnetic susceptibility of HC ($4.1\cdot10^{-4}$) with respect to infected RBCs ($1.8\cdot10^{-6}$) when the chip is operated in the horizontal configuration and the gravity force opposes the magnetic one (perpendicular to the chip surface), only HC are detected. In the vertical configuration, instead, there is no threshold: the magnetic and gravity force are perpendicular and both HC and infected RBCs can be captured. Different configuration angles of the chip with respect to gravity force and different thicknesses of the microfluidic chamber containing the blood sample have been investigated experimentally and by multiphysics simulations. The optimum conditions leading to maximum sensitivity and specificity of the test are described.

Finally, we present a study on cultered RBCs infected by Plasmodium falciparum showing the capability of TMek to distinguish the different infection stages of Plasmodium (ring, trophozoite, gametocyte) by analyzing the waveform of the impedimetric signal in response to the application/removal of the external magnetic field which activates the on-chip magnetic concentrators.

These features are fundamental in order to develop diagnostic tests providing additional information on the sickness state, beyond the sole detection/quantification of parasites.

[1] Giacometti et al., Appl. Phys. Lett. 113, 203703 (2018).

[2] Nam, Jeonghun et al. Analytical chemistry 85.15 7316-7323 (2013).
Key role of the Co2+ cations on the destabilization of the collinear ferrimagnetism in cobalt ferrite nanoparticles by selective incorporation of structural defects

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Cobalt ferrite nanoparticles (NP) are competitive candidates in nanomedicine and biomedical applications, especially in the fields of magnetic drug delivery, magnetic resonance imaging, and separation and detection of biomolecules [1]. The reason behind is a suitable combination of excellent chemical and magnetic characteristics, such as a high chemical stability, surface active sites, and ease of synthesis and functionalization, together with a high anisotropy constant, a high coercivity, and a moderate saturation magnetization. However, in order to control the functional response of cobalt ferrite NP, one of the biggest challenges is to quantitatively disentangle the dependence of the composition, structure, or surface chemistry onto the overall magnetic response.

In this respect, the main finding of our study is to unravel the key role played by the Co^{2+} cations on the destabilization of the collinear ferrimagnetism in cobalt ferrite NP through the selective incorporation of structural defects, by combining an advanced synthesis approach with a broad set of world-class complementary local probes. A set of samples of monodispersed, 8 nm cobalt ferrite NP of identical stoichiometry but with a progressive inclusion of structural defects was prepared [2,3]. As local characterization probes, synchrotron-based, element-, valenceand site- specific X-ray spectroscopy and magnetometry on ensembles of NP was combined with high resolution transmission microscopy of selected, individual NP [3]. The analysis of element-specific X-Ray Magnetic Circular Dichroism (XMCD) spectra and hysteresis loops for all cationic sites reveals that the collinear alignment of the Co²⁺ cations in octahedral sites is significantly more affected by the structural disorder than in any other cation. This is due to the local anisotropy axes induced by the strain associated with the structural defects, giving rise to Co²⁺ spin canting via spin-orbit coupling due to the relatively large Co²⁺ orbital momentum. As the number of structural defects increases, the rest of the cations are progressively dragged off the ferrimagnetic alignment, being the Fe³⁺ cations in tetrahedral sites the last ones to be affected by the disorder [3]. Our results may help clarify the much debated, large variability of magnetic properties in the literature of cobalt ferrite NP with slightly different structural features. Our work highlights the importance of combining a great control over the NP microstructural features, by employing suitable synthesis methods, with the use of advanced complementary local probes in order to disentangle and quantify the effect of every cation and the local environment into the final magnetic response of cobalt ferrite NP, which is essential to tailor and boost their performance for targeted biomedical applications.



Figure 1. : (left) HRTEM images for nanoparticles with increasing crystalline quality, S1 to S4. (right) XMCD hysteresis loops measured at the four cationic sites, at 2 K within \pm 69 kOe, for the four samples.

[1] S. Y. Srinivasan et al., Future Medicine 13, 1221 (2018).

[2] C. Moya et al., Phys. Chem. Chem. Phys. 17, 13143 (2015); J. Mater. Chem. C 3, 4522 (2015).

[3] C. Moya et al., in review.

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Heating efficiency of magnetic nanoparticles for magnetic hyperthermia: effects of temperature and driving-field waveform

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The power released by magnetic nanoparticles (MNPs) submitted to an alternating driving field in the typical frequency range used in therapeutic treatments based on magnetic hyperthermia is studied using a rate-equation approach in which MNPs are modelled as double-well systems with randomly distributed easy axes.

Taking into account the temperature dependency of MNP magnetization and anisotropy, a detailed picture of the their hysteretic properties and of the resulting heating efficiency. The temperature variation in a host medium is then studied by solving a simple, radial Fourier equation with two typical boundary conditions. The time evolution of the temperature increment of the medium turns out to be strongly affected by the non-monotonic temperature dependence of the power released by NPs (see Figure 1a); the direct consequences on the heating efficiency make the determination of the specific loss power (SLP) and the resulting maximum temperature from the initial slope of the heating curve misleading. An alternative, sounder method to measure the average power released by magnetic nanoparticles and the SLP is proposed [1].

Another way of optimizing the performance of MNPs as heat generators consists in applying non-harmonic drivingfield waveforms, with notable advantages with respect to the effects of harmonic magnetizing fields of same frequency and amplitude [2]. This improvement is related to the presence of fast, quasi-adiabatic transformations where the occupancy numbers in the two potential wells are virtually frozen, resulting in largely off-equilibrium magnetization states and in hysteresis loops with a greater area. In particular, changes of the inclination of the trapezoidal waveform's lateral sides are also shown to induce controlled changes in the specific loss power generated by the activated nanoparticles (Figure 1b). Specific issues typical of the therapeutic practice of hyperthermia, such as the need for fine tuning of the optimal treatment temperature in real time, the possibility of combining sequential treatments at different temperatures, the ability to substantially reduce the heating transient in a hyperthermia treatment are properly addressed and overcome by making use of versatile nonharmonic driving fields of trapezoidal shape [3].

In the light of our results, features, methods, advantages and dangers of MNP-assisted precision nanomedicine are assessed in a more accurate way, and the efficiency of magnetic hyperthermia treatments is improved.



Figure 1. : **a** time profile of the temperature increment and corresponding temperature behaviour of the power released curves for two NP diameters using convective boundary condition; **b** effect of the inclination of the trapezoidal waveform's lateral sides (parameter y) on the time evolution of the temperature in a sample containing a volume fraction of $f_v = 0.5\%$ of polydisperse magnetite nanoparticles distributed in size according to a lognormal probability density function with D_c = 13.25 nm and $\sigma = 0.06$.

- [1] G. Barrera et al., Nanoscale 12, 6360-6377 (2020).
- [2] P. Allia et al., Phys. Rev. Applied 12, 034041 (2019).
- [3] G. Barrera et al., Nanoscale Advances, in review.

Observation of Cellular Ion Transport by Magnetoencephalography (MEG) of Cells in Culture

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Metastatic cancer cells are more depolarized than normal cells and there is evidence in literature to support the hypothesis that ion channel/transporter activation and related ionic flux are linked to cancer. We report a non-invasive magnetoencephalography (MEG) technique to measure the magnetic field emanating due to the cellular level process of ion transport through the cell membranes. The technique is used to measure weak and intrinsic magnetic field emanating from HeLa, HEK293 and H9c2(2-1) rat cardiac cells in culture. Addition of a non-lethal dose of ionomycin to HeLa and capsaicin to HEK293 cells expressing TRPV1 channels, respectively, resulted in a sudden change in the magnetic signal consistent with the confocal fluorescence experiments. In contrast, addition of capsaicin to HEK293 cells expressing TRPV1 channels or fluorescent signals. These results unequivocally confirmed that the MEG detected signals are due to cellular ion transport through the cell membrane. Non-differentiated H9c2(2-1) rat cardiac myoblasts and the differentiated myocytes showed a remarkable difference in their magnetic signatures. Myoblasts, a cancer cell line, showed only the characteristic frequency of 27.8 Hz that is similar to that of the HeLa cells. The differentiated cell lines, in addition to 27.8 Hz, also showed three additional, highly resolved characteristic frequencies (triplets) at 194.4, 222.2, and 250.0 Hz.

Tailored Magnetic Nanoparticles for Hyperthermia

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Magnetic nanoparticles (MNP) have a very wide range of applications, namely in Biomedicine and Environment. Their use for imaging purposes or as ultrasensitive sensors has pushed forward related technologies. In fact, the magnetic behavior of MNP is also the basis of magnetic fluid hyperthermia (MFH). MFH was primarily envisaged for cancer therapy, standalone or as an adjuvant, but it may well extend to other health-related issues, namely viral infections. In 2012, MFH was approved for clinical use in Europe, in combination with radiotherapy for the treatment of recurrent glioblastoma. MFH uses the energy dissipation of MNP under an alternate magnetic field to heat the tumor cells, leading to their apoptosis or necrosis. Results have shown that it has the ability to destroy cancer stem cells [1,2], thus impacting cancer progression.

The MagNano group (BiolSI/FCUL) focus its research on the optimization of the heating efficiency of MNP, with emphasis in iron oxide nanoparticles (IONP). The strategy is to control Néel frequency through the tailoring of magnetic anisotropy. This can be achieved by acting on the surface layer and/or the shape anisotropy of the MNP. Environmentally friendlier and up-scalable synthesis methods were chosen, aiming to reduce both toxicity and cost. In this work, we present our latest results concerning the production of stable aqueous suspensions of MMNP with mean sizes ca. 10 nm. Magnetite/maghemite NP were either directly synthesised by a standard co-precipitation method, suitably modified by the addition of various dispersion/coatings agents, or indirectly via another compound NP.

The IONP were characterized for composition, structure and magnetic properties, and finally for their heating efficiency under bio-accepted alternate magnetic fields (270 kHz, 14 kA/m). The results are discussed in relation to the physical properties of the MNPs, and their comparison with reported results is also carried out.

[1] H. Park et al., Int J Hyperthermia 24(8), 638-48 (2008).

[2] H. Huang et al., J Stem Cell Transplant Biol. 2(2), 113 (2017).

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2D magnetrode for local magnetic neuronal recording

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lonic flows circulating inside and outside neurons are creating magnetic signals with amplitudes estimated from pT to nT, according to the number of neurons involved and to the distance to the current source. We have fabricated probes to record local magnetic signals *in-vivo* in close vicinity of neurons for direct neuronal imaging.

The probes contain two Giant Magneto-Resistive (GMR) sensors based on spintronic principles that provide very sensitive and miniaturized detection of the magnetic field. These probes, called magnetrodes, has been used for recording evoked response fields associated with the electrical response of population of neurons in the visual cortex [1].

We are targeting the current mapping of individual neurons by designing a probe with a reduced invasiveness on one hand, and with a sensitivity along two directions in the plane of the probe on the other hand.

After the processing of the sensing elements and contacts, we have used deep Reactive Ion Etching (RIE) on Silicon On Insulator (SOI) substrates for thinning the magnetrode's tip to decrease damage during probe insertion in the brain and to be as close as possible of neurons to avoid the fast magnetic field decay with distance. We achieved a thinning of our magnetrode's tip from 300µm down to 25µm without loss of sensitivity or increase of sensor's noise for a resulting limit of detection around 1nT at 1kHz.

Then we implemented two GMR sensors with orthogonal axis of sensitivity on the same magnetrode to allow the mapping of neuronal currents in two dimensions. The reference layer magnetization direction determines GMR axis of sensitivity and in order to set both reference layer magnetization perpendicularly, two steps are required. First, a global annealing sets both sensors magnetization in the same direction, and then a local repining sets the reference layer magnetization of a single sensor at 90°.

In this work, we present the thinning and the repining process of a 2D magnetrode, the characterization of both sensors at 0° and 90° with a phantom and a path towards *in-vivo* application.

[1] L. Caruso et al., Neuron 95, 6, 1283-1291.e4 (2017).

Biocompatibility and magnetic hyperthermia effect of NiFe2O4 nanoparticles

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We have produced magnetic nanoparticles (MNPs) and tried to use these particles for some medical applications. For example, an agent of magnetic resonance imaging (MRI), magnetic particle imaging (MPI) and magnetic hyperthermia treatment (MHT). In this study, we tried MNPs to apply for MHT. Hyperthermia is one of a cancer therapies using heat effect. In order to realize MHT, dispersibility and biocompatibility in blood vessel and heating efficiency of MNPs were important factors. So, we have devised a method to coat MNPs with polyethylene glycol (PEG), which had highly hydrophilic agent. We prepared various NiFe₂O₄ nanoparticle samples coated by PEG with this method. These single phase spinel structure and particle size of 5, 8, 10, 14 and 21 nm were examined by X-ray diffraction (XRD) and X-ray fluorescence (XRF) measurements. Modification of PEG was confirmed by Fourier transform-infrared spectrometer (FT-IR) and mass spectrometry (MS).

To estimate the amount of PEG on the particle surface quantitatively, we measured TG-DTA (Thermogravimeter-Differential Thermal Analyzer) of 5 nm and 21 nm samples. These results showed 21 nm sample had larger amount of PEG ratio per a particle and we measured hydro dynamic diameter in water to analyze dispersibility of MNPs. As a result, hydro dynamic diameter of 21 nm sample showed higher dispersibility than that of 5 nm sample.

The magnetization curves (M-H) of all samples were measured at 300 K in ±10 kOe field. None of the samples exhibited a coercive force, which is characteristic of superparamagnetic behavior. The imaginary part of the magnetic susceptibility χ'' related to heat release depending on the particle size is analyzed at room temperature in AC magnetic field. The maximum value of χ'' was 21 nm sample and this sample would be expected to generate the highest heat efficiency following magnetic relaxations of superparamagnetic system.

We measured temperature rise effect in AC magnetic field on 15 kHz, 110 Oe for all samples. The temperature of the sample was measured by an optical fiber thermometer. The temperature rise of 44 K was observed for 21 nm sample as the highest temperature rising effect. This temperature increase would be high enough to kill the cancer cells. In vitro experiment using cultured human breast cancer cells (KPL-4) with 21 nm sample were carried out and cell survival rate of the half of cancer cells was confirmed. As a result, the sample was expected as magnetic hyperthermia agent.

Synthesis Optimization of Mn-Zn Ferrites for Self-Regulated Heating Applications

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Manganese-Zinc ferrite nanoparticles have been the subject of increasing research due to their desired properties for a wide range of applications. These properties include adjustable nanometer particle size, tunable magnetic behavior, and high saturation magnetization, providing these ferrites with the necessary requirements for cancer treatment via magnetic hyperthermia [1].

During this research, we have synthetized and characterized Mn-Zn ferrite powders, aiming to optimize their structural and magnetic properties for further application in a ferrofluid.

In this work, samples of $Mn_{1-x}Zn_xFe_2O_4$ (x=0; 0.5; 0.8; 1) were synthetized via the sol-gel auto-combustion and hydrothermal methods. Synthetized powders were characterized by XRD, SQUID, SEM, TEM and induction heating setup. The XRD diffractograms of hydrothermally produced samples presented low percentage of secondary phases, such as hematite (α -Fe2O3) (<10%) for all samples. The Williamson-Hall analysis revealed the crystallite size increasing with
br />the increase of Mn content. TEM images of hydrothermal samples reveal agglomerates composed of few nanoparticles, with spherical shape and mean particle size varying from 7 to 41 nm with increasing Mn content. SQUID of hydrothermally prepared nanoparticles showed that with the increase of Zn, saturation magnetization varied from 79 to 19 emu/g, coercive field from 41 to 11 Oe and remnant magnetization from 5 to ~0 emu/g. More noticeably the M(T) curves, figure 1.a), revealed a shift in the samples Curie temperature towards lower temperatures with the increase of Zn content, from ~610 (estimated) to ~250 K.





The nanocrystals of Mn-Zn ferrite produced by the hydrothermal method present better crystallinity and magnetic properties than the sol-gel auto-combustion samples. The hydrothermally synthetized samples revealed dependence of its structural and magnetic properties with Mn/Zn ratio. The Curie temperature of these ferrites can be used as a self-controlled mechanism of heating, rising these ferrites to a class of intelligent materials.

- [1] N. Zhu et al., IEEE Trans. on Applied Superconductivity ASEMD, 18, 8265.
- [2] P. H. Nam et al., Physica B: Condensed Matter 550, 428-435 (2018).
- [3] Y. Xuan et al., Jour. of Magnetism and Magnetic Materials 312, 464-469 (2007).

Magnetoresistive immuno-microarrays: towards relevant clinical limit of detection using proprietary antibody immobilization solutions

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Immunoassays based on magnetoresistive biochips [1] and a portable platform [2] are currently under development for the detection of protein biomarkers. Each biochip comprises an array of 30 spin-valve sensors, arranged in 6 sensing sites surrounded by a gold frame for micro-spotting purposes of probe solutions. The sensors are covered by thin gold pads and functionalized with a crosslinker (sulfo-SPDP), which mediates the covalent binding between the substrate and the probe molecules. Labelling of target molecules and transduction of recognition events is based on the use of superparamagnetic nanoparticles. In the presence of an external field, these particles exhibit a magnetic fringe field detectable by the spin-valve in its proximity, causing a change in its electrical resistance. The sensor resistance variation is proportional to the number of magnetic particles bound to the captured target molecules [1]. In previous work, the carcinoembryonic antigen (CEA) was used as a model protein for validation of the MR immunoassay with a limit of detection at the 4.69 ng/ml (Fig. 1a) [3]. However, improvement of the limit of detection is still necessary in order to reach lower detection limits (CEA clinical relevant lower range down to 3.5 ng/ml) [4]. Since antibodies serve as recognition elements, assay sensitivity is highly dependent on the effective immobilization of the capture antibodies onto the sensor surface. Therefore, in this work antibody immobilization was studied and optimized using IMG Pharma's proprietary antibody immobilization solutions. The immunoassay used a sandwich strategy consisting of, from bottom to top, capture antibody, antigen (CEA), detection antibody and secondary labelled antibody (fluorophore or magnetic particle) (Fig.1a - inset). The spotting of the reagents was achieved by automatic spotting using a micro-array capillarity imprinting machine (Arrayit, CA, USA). Antibody immobilization efficiency on gold surfaces was compared between SIVG 0.05% (IMG Pharma patented) and a standard buffer solution (PBS). Antibodies labelled with a fluorescent dye (alexa 555) were spotted onto sulfo-SPDP functionalized gold substrates. Three washing steps were performed with spot intensity being quantified between each step through volume analysis (Fig. 1b). The results show that SIVG allows a stronger immobilization since spot intensity tends to increase after washing. Optical assays have been performed on gold substrates employing SIVG and the limit of detection was improved to 1 ng/ml (Fig. 1c). Further tests on the MR platform will be conducted to validate the improvement on the limit of detection of the immunoassay.



Figure 1.: **a** MR platform measurements for CEA concentrations of $1 \mu g/ml$ and 10 ng/ml and for negative control (anti-E.coli antibody). A sandwich strategy represented in the inset was employed in these immunoassays; **b** Evolution of intensity of spotted antibodies labelled with a fluorescent dye prepared in two different immobilization solutions (SIVG 0.05% and PBS), after three washing steps; **c** Optical detection assays performed on gold substrates for CEA concentrations of 1 ng/ml, 10 ng/ml and 100 ng/ml, and for negative control (PBS solution).

- [1] V. C. Martins et al., Biosens. Bioeletronics 24 (2009).
- [2] V. Martins et al., J. Magn. Magn. Mater. 322 (2010).
- [3] D. C. Albuquerque et al., IEEE Magn. Lett. 10 (2019).
- [4] Ballesta A.M. et al., Tumor Biol. 16 (1995).

Poster

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Distributed heat production in clusters of magnetic nanoparticles

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We address the issue of quantifying the heat produced by a single magnetic nanoparticle (MP) embedded within an interacting MP cluster. This is relevant for MP hyperthermia considered as a modality for enhancing cancer therapies, where recent experiments raised questions whether it is the net heating effect of the entire MP aggregate that leads to tumor death, or the local heat distribution across it [1]. Thus, it becomes necessary to understand the distribution of heat production across a MP aggregate inside a living cell.

In this work we apply the non-equilibrium stochastic thermodynamics combined with the Néel-Arrhenius transition state theory to systems of interacting Stoner-Wohlfarth MPs [2]. We show that it is possible to associate the expressions for entropy production with the individual MPs within an interacting assembly, which in turn allows computing the single particle heat across the assembly.

Our calculations suggests that dipolar interactions lead to a significant heat distribution across a particle system, often varying by as much as 50% - 100% of the mean heat practically measurable from the full hysteresis loop area. This could lead to considerable local heating produced inside cancer cells while the net temperature effect remains small, corroborating the earlier experimental findings. We also show that due to the presence of dipolar interactions between the MPs, the area of the hysteresis sub-loops corresponding to individual MPs does not represent the particle heat, and performing explicit calculations based on evaluating the entropy, such as developed in this work, is necessary.

M. Creixell et al., ACS Nano 5, 7124 (2011).
U. Seifert, Rep. Prog. Phys. 75, 126001 (2012).

Controlling the morphology and the oxidation state of iron oxide nanoparticles by thermal decomposition method

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Iron oxide nanoparticles (NPs) have attracted a great attention for biomedical and environmental applications due to their good biocompatibility and magnetic performance. Nevertheless, the structural and magnetic properties of these NPs are strongly dependent on the synthesis conditions [1,2].

The overall aim of this work is to improve the reproducibility and to optimize the synthesis of iron oxide NPs by a thermal decomposition method in order to tune each sample to its specific application. On the one hand, we found that greater amounts of either 1,2-hexadecanediol or the solvent 1-octadecene yielded monocrystalline NPs, but both reagents hampered the growth of the NPs and diminished the reaction yield. On the other hand, small amounts of one of them increased the reaction yield but induced the formation of wüstite as a parasitic phase. Consequently, the samples exhibited two distinct magnetic behaviors. At room temperature, small NPs were superparamagnetic while the bigger ones showed ferrimagnetic behavior as well as shifted hysteresis loops at 5 K after field cooling at 1 T due to exchange bias. Zero-field cooling-field cooling (ZFC-FC) curves below 200 K for the small NPs showed one peak at low temperatures while those for the bigger particles displayed two peaks at higher temperatures which can be associated with a Verwey transition of magnetite and the Neel transition of wüstite.



Figure 1. : TEM images of the samples synthetized with increasing amounts of 1,2-hexadecanediol in 5 mL of 1-octadecence. **a** 0 mmol, **b** 2.5 mmol, **c** 6 mmol and **d** 12 mmol. Bottom panels show samples with increasing volumes of 1-octadecene using 6 mmol of 1,2-hexadecanediol. **e** 0 mL, **f** 2.5 mL, **g** 5 mL, **h** 20 mL.

[1] A. F. Rodríguez et al., J. Mater. Chem. C 6, 4, 875–882 (2018).

[2] C. Moya et al., Phys. Chem. Chem. Phys. 17, 19, 13143–13149 (2015).

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In silico modelling of magnetic nanodisks for hyperthermia applications

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Several kinds of magnetic nanostructures have been recently investigated for applications in nanomedicine therapeutics, as an alternative to superparamagnetic iron oxide nanoparticles (SPIONs). As an example, magnetic nanodisks have been proposed as magneto-mechanical actuators to destroy cancer cells or deliver drugs, exploiting their ability to rotate under the action of low frequency magnetic fields [1].

Another application is magnetic hyperthermia for cancer cure, where disk-shaped nanostructures of different magnetic materials have been explored as heating agents for generating strongly localized increments of temperature [1-3]. Under the action of ac magnetic fields, they are able to produce heat via magnetic hysteresis, showing values of specific loss power (SLP) higher than those of clinically approved SPIONs. Moreover, they can be characterized by vortex state at remanence, feature that leads to small magnetostatic interactions and, advantageously, to low probability of aggregation. However, their design needs a careful selection of the source excitation parameters (magnetic field frequency and amplitude) to not exceed the upper limitation of 5×10^9 Am⁻¹s⁻¹ (Hergt-Dutz limit), conventionally used to avoid non-selective heating due to eddy currents.

Despite the heating efficacy of magnetic nanodisks, a systematic analysis is required to identify dimensional and physical properties as well as ac source conditions that maximize SLP, guaranteeing at the same time the fulfillment of biophysical constraints. Another important aspect is the determination of the temperature increase produced in biological tissues as a function of the exposure time, to select operative conditions to be used in possible therapeutic sessions.

In this framework, we focus on permalloy nanodisks, by performing a parametric analysis aimed at finding the optimal geometrical features for magnetic hyperthermia, considering the Hergt-Dutz limit for the selection of field parameters. Specifically, we vary the disk diameter from 100 to 800 nm and the thickness from 15 to 30 nm; SLP is derived from hysteresis losses, being the predominant heating contribution. The analysis is carried out via numerical modelling, calculating the hysteresis loops with a GPU-parallelized micromagnetic solver for the spatial-time integration of the Landau-Lifshitz-Gilbert equation [3]. The influence of nanodisk concentration is also investigated, focusing on the role of magnetostatic interactions and relative orientation with the applied field. We also analyse the mechanical behaviour in media with decreasing viscosity, to verify the occurrence of oscillatory motion driven by ac fields.

After selecting field parameters in accordance with Hergt-Dutz limit, we calculate the temperature increase in test tubes filled with ferrofluids containing permalloy nanodisks with variable concentration, under both adiabatic and non-adiabatic conditions. The analysis, carried out with a finite element model for the heat transfer equation, is focused on nanodisks with optimized hyperthermia properties, addressed by the previous micromagnetic modelling study. Then, we determine, by solving the Pennes' bioheat transfer equation [4], the temperature increase induced in biological tissues within mouse anatomical models, also analysing the influence of field exposure.

- [1] Applied Physics Reviews 7, 011306 (2020).
- [2] Adv. Funct. Mater. 25, 812–820 (2015).
- [3] *Sci. Rep.* **9**, 6591 (2019).
- [4] Int. J. Mol. Sci. 20, 4644 (2019).

Nanoimprint lithography based fabrication of synthetic antiferromagnetic nanoplatelets for bio-applications

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Magnetic nanoparticles are widely used in bio-application due to their ability to remotely manipulate their surroundings. Here, we show novel magnetic nanoplatelets based on synthetic antiferromagnetic (SAF) stacks consisting of multiple repeats of [Pt/CoB/Ru/CoB/Pt], where the two magnetic layers are antiferromagnetic coupled through a Ru spacer layer [1]. The basic stack of the nanoplatelets is shown in Figure 1. These particles exhibit zero magnetic moment in zero magnetic field preventing agglomeration in fluids. Moreover, the response to magnetic fields can be fully customized. These nanoplatelets also exhibit a strong perpendicular magnetic anisotropy, which makes them ideal candidates for applications that rely on the application of local mechanical forces or torques, such as magnetic tweezer, separation and cancer therapy based on mechanical cell disruption [2].

In this contribution, we will report on a new top-down method to fabricate monodisperse SAF nanoplatelets ranging from hundreds of nanometers to several micrometers in size through substrate conformal nanoimprint lithography (SCIL). Figure 2 shows the AFM graph of nanoplatelets after fabrication. Compared to other fabrication methods, SCIL can easily be used to produce particles with size down to tens of nanometers at a large scale.

In this presentation, we will show the fabrication process of the SAF nanoplatelets and the change of magnetic properties: (i) we will show that the SAF nanoplatelets maintain their perpendicular anisotropy and antiferromagnetic properties as compared to blanket thin film stack. (ii) upon patterning, the coercivity and its distribution increases which we will elaborate on and discuss methods to circumvent this. Finally, we will show the magnetic response of the nanoplatelets in solution.



Figure 1. : Basic unit of the SAF stack.



Figure 2. : AFM of the nanoplatelets after fabrication.

[1] R. Mansell et al., *Scientific reports* **7**.1, 1-7 (2017).

[2] I. Vlaminck et al., Annual review of biophysics 41, 453-472 (2012).

Fe-Cr-Nb-B magnetic particles and STEM cells, triggers for cancer cells apoptosis by magneto-mechanical actuation

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Recently, we have introduced a new type of magnetic particles (MPs), prepared by wet milling of superferromagnetic Fe-Cr-Nb-B precursor glassy ribbons, for cancer treatment by magneto-mechanical actuation in low magnetic fields (1÷20 Oe) [1]. The rectangular shapes of newly developed MPs and the superferromagnetism of the glassy alloys of which they are made induce important magnetic shape anisotropies which, in association with a large saturation magnetization, generate an improved torque in a rotating magnetic field, producing important damages on the cellular viability of MG-63 human osteosarcoma (HOS) cells. The specific behavior of the Fe-Cr-Nb-B MPs offers them destructive effect even in low magnetic fields, and this characteristic allows the use of coils systems which provide large experimental spaces. In this work we studied the possibility of using adipose derived stem cells (ADSCs) for the transport of Fe-Cr- Nb-B MPs to areas with HOS, and the effect of particles incorporated into ADSCs on the viability of HOS following the magneto-mechanical actuation of MPs. Fe Cr Nb B MPs with dimensions between 5 and 100 nm were obtained by high-energy wet milling of rapidly solidified meltspun ribbons precursors in oleic acid. We studied the effect of magneto-mechanical actuation of the MPS on ADSCs and HOS, respectively. For experiments we used a rotating magnetic field of 10 Oe at the frequency of 2 Hz, provided by a system of perpendicular coils. After incubating the cells with nanoparticles for 24 h, they were exposed to a rotating magnetic field. The viability tests performed 24 hours after the magneto-mechanical actuation by MTT assay (3-(4,5-dim ethylthiazolyl-2)-2,5-dyphenyltetrazolium bromide) indicate values of 24÷25% for HOS and up to 10% for ADSCs. We investigated also the capacity of ADSCs to incorporate Fe-Cr-Nb-B MPs, and we found that large amounts of MPs were ingested as confirmed by using a colorimetric ferrozine-based assay. All these characteristics make ADSCs an ideal candidate for the internalization of a large quantity of Fe-Cr-Nb-B MPs with the purpose of cell targeting. Therefore, considering the advantages of ADSCs, we have used them as carriers of MPs to target and destroy cancer cells through magneto-mechanical actuation. Hence, we used a time-lapse cell imaging method to observe and record the cellular migration of ADSCs loaded with MPs towards tumor cells in vitro. The recorded films demonstrated that MPs-loaded ADSCs were able to target osteosarcoma cells. We performed in vitro tests, where ADSCs were incubated with Fe-Cr-Nb-B MPs then transferred onto an osteosarcoma culture and exposed to the rotating magnetic field. Magneto-mechanical actuation led to the destruction of ADSCs and to the release of MPs on HOS culture, which incorporated the released MPs. By magneto-mechanic actuation, the HOS viability dropped down to 21÷22%. TEM images demonstrate that MPs incorporated in the cells reached the cells structure, except the nucleus. The irregular shape of the MPs combined with their rotational motion in the rotating magnetic field produce irreversible damages on the cell's membranes and organelles, and, subsequently, the cell's death. Work supported by the Nucleu Programme (Project PN 19 28 01 01) and Contract No. 11PFE/16.10.2018 financed by the Romanian Ministry of Education and Research.

[1] H. Chiriac et al., Sci. Rep. 8, 11538 (2018).

Microfluidics mixing with rotational supramolecular structures of magnetic nanoparticles

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Developing integrated multifunction microfluidic systems combining magnetic stirring and magnetic nanoparticles is very promising for low power, automated, contactless fluid control and bioanalysis even in low-resource environments. Yet, their optimal performance demands deep understanding of combined effects of complex 3D magnetic field gradients, stream fluid properties and biosystems. The balance between hydrodynamic and magnetophoretic forces as well as fluid-particle dynamics remain unclear, particularly with micro-to-femto mass flow rates in the presence of strong external fields as rotation-based.

An experimental setup was assembled to perform microfluidic tests on mixing with supramolecular structures of magnetic particles. It consists in a mixing platform, whose 15 mm radius chamber was filled with isopropyl alcohol and NdFeB magnetic particles at 10% V/V. The chamber was placed above an automated rotating platform with block-shaped magnets (NdFeB, Q-05-1.5-01-N, Supermagnete) and a Levenhuk DTX 30 Digital Microscope connected to the computer allowed to register the mixing phenomena. The automated rotating platform consists of the PDMS platform for magnets (Fig. 1) placed on a stepper motor (Vexta Model PK245-01AA, Oriental Motor Co.), which is actuated by the 2H Microstep driver (DM420, ACT Motor GmbH) and controlled by the microcontroller (Arduino Uno).

Preliminary results show several stable nanoparticle structures that can be generated and rotated inside the mixing chambers dependent on the rotating speed, number and position of magnets and distance between rotating magnets and fluidic chamber. The nanoparticle structures demonstrated having considerable friction with the PMMA surface and even tend to adhere irreversibly to it for lower rotation speeds (< 0.1rpm). The nanoparticles supramolecular structures are seen to result mainly from the balance between magnetic forces, friction forces and surface tension forces. Homogeneous mixing performance was achieved for 4 external magnet configuration (Fig 1*aiii*) and was found to be reduced for lower rotation speeds or increased number of magnets (8 magnets with 4 internal + 4 external configuration, instead of only 4 external).



Figure 1 : a) 4 external magnet configuration. **b)** 4 internal + 4 external magnet configuration. **i)** schematics of magnet positioning on the automated rotating platform. Frame of flow conformation resulting from **ii)** 10 rpm and **iii)** 20 rpm rotation speeds. The black residue corresponds to the nanoparticle structures, the silver rectangles are the magnets.

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Symposium 2. Magnetic Information

Storage and MRAM

Quench-switching high resistivity states in antiferromagnetic CuMnAs

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Research of antiferromagnetic spintronic memories is stimulated by promising features of the antiferromagnetic materials such as their ultrafast dynamics (several orders of magnitude faster compared to ferromagnets [1,2]), and the rich properties of this abundant class of material. The understanding of the processes in antiferromagnets is, however, very challenging since the tools for the observation of antiferromagnetic order are limited. This triggers broad efforts to develop new techniques suitable for antiferromagnets, often utilizing recent discoveries in spintronics.

Here, we will focus on a newly observed mechanism of storing information in epitaxial films of antiferromagnetic CuMnAs which is based on quench-switching of the magnetic order resulting in large changes of the electrical resistivity [3]. The multilevel functionality, electrical and laser pulse writing and erasing, and the temperature dependence of the relaxation processes will be discussed in detail.

Furthermore, possible scenarios of the microscopic origin of the observed resistivity changes will be discussed in the context of the domain fragmentation observed by several experimental techniques [4,5].



Figure 1: Highly reproducible resistivity switching and relaxation in a simple CuMnAs bar device.

[1] Wadley, P. et al., Science 351, 587-590 (2016)

- [2] Olejník, K. et al., Science Advances 4, eaar3566 (2018)
- [3] Kašpar, Z. et al., arXiv:1909.09071 (2020)

[4] Wörnle, M. S. et al., arXiv:1912.05287 (2020)

[5] T. Janda et al. Phys. Rev. Materials 4, 094413 (2020)

Magnetization reversal driven by Spin-Transfer-Torque in Perpendicular Shape Anisotropy Magnetic Tunnel Junctions

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The perpendicular Spin-Transfer-Torque Magnetic Random Access Memory (p-STT-MRAM) is one of the most promising emerging non-volatile memory technologies. However, these devices are limited by their thermal stability factor at technological nodes smaller than 20 nm. Indeed, in conventional p-STT-MRAM, the interfacial perpendicular magnetic anisotropy (iPMA) at the FeCoB/MgO interfaces is partially balanced by the easy-plane magnetostatic energy, reducing significantly the retention time of the memory [1-3]. A promising solution to this problem relies on inducing, in the storage layer, a perpendicular shape anisotropy by increasing its thickness. In this case, the magnetostatic energy adds up to the iPMA, resulting in a much larger and easily tunable effective perpendicular anisotropy than in a conventional p-STT-MRAM. Combining these sources of anisotropy allows to extend the downsize scalability of the STT-MRAM towards sub-10 nm technological nodes [4, 5] thanks to a significant reinforcement of the thermal stability factor. As the storage layer thickness is affecting both the thermal stability factor and the writing operation of the cell, thereby the knowledge of the physical phenomena acting during the STT-driven magnetization reversal is mandatory for the engineering of optimized devices. In this work, a systematic study of the magnetization reversal mechanism of the PSA-STT-MRAM induced by STT was conducted. Micromagnetic simulations were carried out, enabling the identification of different regimes of the magnetization reversal in such memory cells. We focused firstly on pillars of 20 nm in diameter and thicknesses varying between 30 and 60 nm subject to spin transfer torque and thermal fluctuations. It was found that, below a threshold thickness of 50 nm, the mechanism of reversal is similar to a collective curling-like reversal with sharp variations of the magnetization. Above this threshold, a transverse domain wall is nucleated at the MgO interface (facing the spin-polarized layer), propagates and the reversal takes longer time (Fig. 1a). The inverse of the switching time follows a linear relation with the applied bias voltage (Fig. 1b). Moreover, this dependency remains linear when considering thermal fluctuations, which demonstrates a robust reversal mechanism. For a relevant comparison between PSA-STT-MRAM and conventional p-STT-MRAM more simulations were performed for selected pillars having similar thermal stability factors. The studies were done both in the thermally activated regime (limit of long current pulses, i.e. much longer than 10 ns) and in the ballistic regime (limit of short pulses).



Figure 1: a) Time evolution of the mean reduced magnetization for different pillar thicknesses with an applied bias voltage of -1.25 V. b) Dependency of the applied voltage bias on the inverse of the switching time for pillars of different thicknesses ranging from 30 nm to 60 nm.

- [1] C. Yoshida et al, Jpn. J. Appl. Phys. 58, SBBB05 (2019).
- [2] H. Sato et al, Jpn. J. Appl. Phys. 58, 0802A6 (2017).
- [3] L. Thomas et al, J. Appl. Phys. 115, 172615 (2014).
- [4] N. Perrissin et al, Nanoscale 10, 12187-12195 (2018).
- [5] K. Watanabe et al, Nat. Com. 9, 663 (2018).

The influence of finite-size effects on the Curie temperature of L10-FePt

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Iron Platinum in L1₀-phase (L1₀-FePt) has been widely studied [1] for application in Heat-Assisted Magnetic Recording (HAMR). An important aspect for successful HAMR media is controlling the Curie temperature dispersion, which for L1₀-FePt a strong dependence on finite-size effects is reported [2, 3]. This poses a serious problem as the inevitable grain size distribution causes a Curie temperature (T_c) distribution which potentially limits recording density.

We employed an atomistic model based on the VAMPIRE code [4] to study finite-size effects in L1₀-FePt grains using nearest-neighbour Heisenberg Hamiltonian exchange. Our grains had variable surface geometries (cylindrical, parallelepiped, and randomly-generated shape) and sizes (from 1 to 10nm). The magnetisation distribution M(T) was calculated from 0K to 1000K in a 5K step using a Monte-Carlo integrator. The Curie temperature of the whole grain and of each of its atomistic layers were computed from the normalised mean magnetisation length and mean susceptibility distribution of the grain as functions of temperature. A finite-size scaling fit to the Curie temperature distribution $T_c(D)$, with D being the characteristic grain size, was used to determine critical exponents and the bulk Curie temperature $T_c(bulk)$. In addition, a novel feature of our model gave access to the magnetisation properties of each atomic layer of the grains which provided insights into the magnetisation profile along a specific dimension. We hypothesised a correlation between the T_c distribution of L1₀-FePt and the percentage of atomistic bond loss on the grain surface as a function of grain size.

Our results establish a size threshold at 2.5nm below which the impact of finite-size effects starts to permeate into the centre of the grain and contribute to the overall drop of the grain T_c (Fig. 1). We observe that above this threshold the magnetisation loss due to surface effects is proportional to the percentage of atomistic bonds loss on the grain surface. Contrarily, below 2.5nm the Curie temperature decreases much faster as a function of bond loss, suggesting that in this limit finite size effects have propagated into the entire grain. This phenomenon is further illustrated by the grain magnetisation profile (Fig. 2) in which for larger grains surface disorder (lower magnetisation) is seen to penetrate only a few atomistic layers, whilst for smaller grains surface effects begin to dominate. Consequently, the grain T_c profile is also found to follow a similar trend. Our findings are consistent with semi-analytical mean-field calculations and have been extended to incorporate different crystal structures, which strongly suggests that if using a suitable correlation factor the T_c distribution of a generic material can be correlated to the percentage of atomistic bond loss on the surface as a universal parameter.



Figure 1: At 2.5 nm ΔT_c is seen to transit between the 2 limits of atomistic bond loss where surface effects fully confine to grain surface (correlation factor = 1) or fully permeate into the grain centre (correlation factor = 2).



Figure 2: The magnetisation profile at 550K shows the dominance of surface disorder in smaller grains.

- [1] D. Weller et al., Journal of Vacuum Science Technology B, 34, 060801 (2016)
- [2] O. Hovorka et al., Applied Physics Letters, 101, 052406 (2012)
- [3] A. Lyberatos et al., Journal of Applied Physics 114, 233904 (2013)
- [4] R. Evans et al., Journal of Physics: Condensed Matter, 26, 103202 (2014)

A novel design of a racetrack memory based on functional segments

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The huge amount of new data generated every day has caused the need to search for new systems to store all this information in the smallest possible areas. S. Parkin proposed a new storage device called racetrack memory, in which magnetic domains are used to store the information in ferromagnetic stripes along its length [1]. In this novel system, the application of spin current pulses would ease the movement of the bits along the stripe reaching the read/write heads, increasing the speed of reading/writing data and the information density stored on a device. However, this potential new storage device still has some limitations that prevent its use. One of the biggest drawbacks is the unwanted overheating of the system caused by the current pulses in the physical notches in the stripe, which are required to pin the domain walls (DWs). This issue could be solved using chemical constraints, which would allow to pin the DWs maintaining the diameter constant along the wire. Considering this idea, a novel configuration based on cylindrical nanowires (NWs) with chemical constraints is proposed in this work. The choice of cylindrical NWs instead of stripes with rectangular section allows, in addition to the use of chemical constraints, to avoid the Walker breakdown thus enabling a faster propagation of information along the device [2]. Furthermore, the manufacturing method of NWs is cheaper than the stripes fabrication method, making the NWs more scalable [3].

In this work, we propose a novel design of a racetrack memory using two functional segments along the NW: a "writing section" and a "recording section". The great novelty in this design is the introduction of a "writing section" along the NW made of two magnetic segments with different coercive fields, isolated from the "recording section" by a non-magnetic constraint. The magnetic simulations performed have proved how one can switch the magnetization (bit information) of the "writing section" without modifying the bit recorded in the rest of the NW. The information written in this first section can then be recorded into the racetrack memory wire by applying a spin-polarized current through the device. The magnetic simulations show a successful depinning of the DWs from the chemical constraints achieved by the application of a spin current pulse through the NW, thus moving the magnetic bits in the whole NW without any loss of information (Fig. 1).

Three different configurations of "writing sections" were simulated, obtaining in all three cases a successful recording and bit movement [4]. Arrays of 3 and 7 NWs (hexagonal array), were also simulated to investigate the effect of neighboring magnetic wires in the bit movement of the central wire. The improvements achieved in the use of cylindrical magnetic NWs as racetrack memories will help to develop a faster magnetic storage device with greater capacity and reliability, fabricated with low-cost and scalable methods.

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Figure 1: DW movement through the NW under the application of a spin-polarized current.

- [1] S. P. Parkin et al., Science 320 (2008) 190
- [2] N. Biziere et al., Nano Lett. 13 (2013) 2053
- [3] C. T. Sousa et al., Appl. Phys. Rev. 1 (2014) 031102
- [4] J. Rial and M. P. Proenca, in preparation.

Synthesis and studies of magnetic and magneto-resonance properties of epitaxial Pd_{0.96}Fe_{0.04}/VN/Pd_{0.92}Fe_{0.08} superconducting spin-valve heterostructure

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Thin ferromagnet/superconductor/ferromagnet (FSF) layered heterostructure is one of the possible logic elements in superconducting spintronics [1]. We synthesized a fully epitaxial FSF heterostructure, which consists of two layers of Pd_{1-x}Fe_x as a ferromagnet and a VN layer as a superconductor. The MgO/Pd_{0.96}Fe_{0.04}(20 nm)/ VN(30 nm)/ Pd_{0.92}Fe_{0.08}(12 nm) heterostructure was deposited by molecular-beam epitaxy technique (MBE system by SPECS, Germany) and reactive magnetron sputtering (BESTEC, Germany) onto the (001)-oriented, epi-polished single-crystal MgO substrate [2-4]. The epitaxial growth mode of the layers was verified by low-energy electron diffraction (LEED) and X-ray diffraction (XRD) techniques. Magnetic properties were studied using VSM magnetometry (QD PPMS-9) and ferromagnetic resonance (FMR, X-band Bruker ESP300 spectrometer).

The specific magnetic moment versus temperature dependence M(T) is presented in Fig. 1 (left). It exhibits a clear kink at a temperature of about 125 K, which can be well reproduced by decomposing the M(T) dependence on the magnetic responses of the constituent Pd_{0.92}Fe_{0.08} and Pd_{0.96}Fe_{0.04} layers (shown by dash lines) using M(T) data for single-layered films. At a temperature of 5.4 K, an additional diamagnetic contribution arises due to the superconducting transition in the VN layer. The FMR technique was used to study magnetic anisotropies in the Pd_{0.96}Fe_{0.04}/VN bilayer (Fig. 1 (right)). The magnetic anisotropy of the bilayer is the same as for single Pd_{0.96}Fe_{0.04} layer – cubic with tetragonal distortion [2, 4].

Different concentration of iron in the ferromagnetic layers leads to different coercive fields [2], which make it possible to utilize them like "free" and "fixed" layers (soft-hard pseudo spin-valve). On the minor hysteresis loop we obtained parallel (P) and antiparallel (AP) mutual alignment of the ferromagnetic layers magnetizations. Resistive measurements showed the inverse superconductive spin-valve ($T_c(P) > T_c(AP)$) effect.



Figure 1. (Left) Temperature dependences of the saturation specific magnetic moment for the trilayer film and the single films; (Right) Angular dependence of the FMR field of the epitaxial Pd_{0.96}Fe_{0.04}/VN film.

[1] Linder J., Robinson J. W. A. Nat. Phys. 11, 307 (2015).

[2] Esmaeili A., Yanilkin I. V., Gumarov A. I. et al., Thin Solid Films 669, 338 (2019).

[3] Esmaeili A., Mohammed W. M., Yanilkin I. V. et al., Magnetic Resonance in Solids. Electronic Journal 21, 4 (2019).

[4] Mohammed W. M., Yanilkin I. V., Gumarov A. I. et al., Beilstein J. Nanotechnol 11, 807 (2020).

[5] Esmaeili A., Yanilkin I. V., Gumarov A. I. et al., https://arxiv.org/pdf/1912.04852

Poster

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Cross-sectional TEM imaging of NiCrMnSi and CoFe:N alloys for magnetic tunnel junctions

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Heusler alloy is one of the mainstream research studies in materials science due to their possible applications in spintronic devices. Such a half-metallic ferromagnetic material has a unique property for exhibiting only one spin channel at the Fermi level [1]. In this study, we investigated structural and magnetic properties of CoFe:N and NiCrMnSi (NCMS) alloys to reveal a key structural parameter to increase a tunneling magnetoresistance (TMR) ratio.

The CoFe:N and NCMS samples were sputtered using ultrahigh vacuum (UHV) magnetron sputtering. The structures of the samples are as follows: (i) Si sub.//Ta (5)/Ru (10)/Co₇₅Fe₂₅:N (15)/Ta (5) and (ii) MgO(001) sub.//NCMS (100)/Ta (3) (thickness in nm). The NCMS samples were *in situ* annealed at $T_{anneal} = 500^{\circ}$ C and 700°C before the Ta layer was deposited. The samples were thinned down as transmission electron microscopy (TEM) specimens using focused ion beam (FIB) for TEM observation.

The CoFe:N samples show the epitaxial growth of the $Co_{0.75}Fe_{0.25}$:N layer on the Ru seed layer without phase segregation and interfacial mixing. The lattice constants of these layers are estimated to be 0.416 and 0.297 nm, respectively, confirming that the $Co_{0.75}Fe_{0.25}$:N layer is grown as a face centred cubic on the hexagonal Ru layer. Partial N₂ gas introduction during the sputtering is found to be the optimum and the optimised $Co_{0.75}Fe_{0.25}$ N layer will be implemented into a magnetic tunnel junction (MTJ) for magnetic transport measurements.

High resolution (HR)-TEM and cross-sectional energy dispersive X-ray spectroscopy (EDX) mapping are obtained for the NCMS samples. The NCMS sample which grown at $T_{anneal} = 500^{\circ}$ C shows atomic mixing at the MgO(001)/NiCrMnSi interface up to 5 MLs, confirming polycrystalline nature as shown in Fig. 1a), even though the corresponding EDX mapping proves all the constituent elements are distributed homogeneously within the NiCrMnSi layer. These results indicate that the growth temperature of 500°C is not enough for the NiCrMnSi crystallisation. The sample grown at $T_{anneal} = 700^{\circ}$ C is found to be epitaxially grown on the MgO(001) substrate with the grain size of 5~10 nm with interfacial mixing of about 20 monolayers, confirming the NiCrMnSi crystallisation (see Fig. 1b)). This agrees with the X-ray diffraction (XRD) signals measured on these devices. However, the corresponding EDX map shows Ni and Cr segregations from the NiCrMnSi matrix, which also agrees with XRD signals.

Resolution of Non-Destructive Imaging for Buried Interfaces

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Non-destructive imaging for buried interfaces has been recently developed by decelerating electron-beam energy in scanning electron microscope (SEM) [1]. This imaging technique has proven to be very useful to identify the contaminations formed in a magnetic tunnel junctions (MTJs), which reduce their tunnelling magnetoresistance (TMR) ratios. For example, in a Heusler-alloy MTJ, residual resist has been found to react with Al in Al-O used to isolate the top and bottom electrodes during sputtering. By reducing the Al-O sputtering power, the yield of the MTJs has been reported to be improved over 70% [2]. In order to further evaluate the resolution of our technique, we prepared a new set of samples with elevating capping layers in this study.

The samples, consisting of Ta (5)/(Ru (5)/)W or Pt (0.5)/Ta (5~60) (thickness in nm), were deposited on Si substrates using a high target utilisation sputtering system. Here, the heavy metal layer of W or Pt were dispersed as nanoparticles, of which diameters can be imaged using the non-destructive technique to confirm the resolution. The penetration depth of electron beam with different energy was estimated using CASINO programme [3]. The electron beam at 4.7 keV is found to reach the interface between Pt and Ta for the case of the 60 nm thick Ta capping layer and to generate back scattered electrons (BSE). The electron beam at 5.0 keV, on the other hand, is found to reach the bottom Pt (or W) interface and to generate BSE. By subtracting these images, the distribution of the Pt nano-particles can be assessed. The corresponding SEM image taken by the subtraction of these acceleration voltages can resolve the Pt nano-particles with 10 nm diameter. For the case of the 5 nm thick Ta capping layer, 2 nm resolution is achieved. Such resolution can be useful for the evaluation of buried MTJs with a few 10 nm diameter. We will further improve the resolution by optimising the imaging conditions, so that we will be able to image smaller MTJs.

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Symposium 3. Magnetic sensors and hybrid integration of magnetic technologies

Non-collinear Anisotropy Arrangements of Exchange Biased Multilayer System for Enhanced Magnetoelectric Sensor Sensitivity

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With the latest breakthroughs in magnetoelectric (ME) sensor development and research, ME sensors have become good candidates for integration in biomagnetic sensing applications such as magnetocardiography and magnetoencelography [1]. One type of ME sensors are the micromachined composite ME sensors that have the advantage of miniaturized design and easy integration into an array and chipsets [2]. These sensors, incorporating complex magnetic multilayer stacks such as antiparallel exchange bias (APEB) [3], have shown low to none magnetically induced noise limitations, even with large total ferromagnetic thicknesses as high as 4 μ m [4]. However, the reduced sensitivity, due to the additional exchange bias anisotropies, still poses challenges for the integration into biomagnetic applications.

In order to improve the sensitivity of such sensors, we present a novel magnetic heating scheme that allows manipulation of the uniaxial anisotropy of the multilayer stack with APEB configuration. The result of such procedure is the formation of an in-plane canted uniaxial anisotropy in each ferromagnetic layer, which results in a softer magnetic response and improved magnetostrictive response along the sensitivity axis. Furthermore, the manipulation allows two possible outcomes that relate to the achieved coherent in-plane rotations of the individual anisotropies. As a result, the two states either have the uniaxial anisotropies of all layers rotated in the same direction or in opposite direction to each other in a sequential manner. Two possible magnetic states with non-collinear anisotropy arrangements were theoretically confirmed using a macrospin model and experimentally determined using in-situ magneto-optical Kerr effect microscopy and inductive magnetometry. The impact of the new multilayer states incorporated within a composite ME sensor is investigated by characterizing the electrical output of the sensors. It is shown, that compared to previous sensors, without modified uniaxial anisotropy of APEB, the newly developed sensors show a 1.5 times larger sensitivity and up to 5-fold improved limit of detection (from 55 pT/VHz to 11 pT/VHz) during magnetic excitation at their electromechanical resonances. The newly modified anisotropy APEB multilayers provide the means to improve further the sensitivity of the APEB sensors, whilst withholding the low magnetic noise properties of the exchanged biased magnetic multilayer.

[1] D. Viehland, M. Wuttig, J. McCord, E. Quandt, Magnetoelectric magnetic field sensors, MRS Bull. 43 (2018) 834–840. https://doi.org/10.1557/mrs.2018.261.

[2] C.M. Leung, J. Li, D. Viehland, X. Zhuang, A review on applications of magnetoelectric composites: from heterostructural uncooled magnetic sensors, energy harvesters to highly efficient power converters, J. Phys. D. Appl. Phys. 51 (2018) 263002. https://doi.org/10.1088/1361-6463/aac60b.

[3] M. Jovičević Klug, L. Thormählen, V. Röbisch, S.D. Toxværd, M. Höft, R. Knöchel, E. Quandt, D. Meyners, J. McCord, Antiparallel exchange biased multilayers for low magnetic noise magnetic field sensors, Appl. Phys. Lett. 114 (2019) 192410. https://doi.org/10.1063/1.5092942.

[4] S. Salzer, V. Robisch, M. Klug, P. Durdaut, J. McCord, D. Meyners, J. Reermann, M. Hoft, R. Knochel, Noise limits in thinfilm magnetoelectric sensors with magnetic frequency conversion, IEEE Sens. J. 18 (2018) 596–604. https://doi.org/10.1109/JSEN.2017.2776039.

High sensitive symmetric response MR sensor with AC field modulation

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High-sensitive Magneto-resistance (MR) sensors have attracted much attention for detecting fine magnetic field from human body or from defects in the Li-ion battery. MR sensors that have detectivity of about tens to hundreds of pT have been reported [1]. They employed arrays of many MR units and/or using averaging procedure for reducing its 1/f noise. AC modulation of a response of the MR sensor is a possible solution to suppress the influence of 1/f noise [2]. The frequency range of the detected signal is converted to a higher frequency where the 1/f noise was low enough. However, distinguishing a fine signal from a large modulation signal is difficult within a restricted dynamic range of a system.

In this paper, a novel AC field modulation method using even-function MR elements (Symmetric Response MR (SRMR) sensor) with a new bridge circuit is proposed.

Basic idea of SRMR system is shown in Fig. 1. A current-in-plane Giant MR (GMR) element is used. Magnetization in the pinned layer and the free layer are aligned to the long axis direction in order to give the even function MR response. A metal line (AC line) is placed on the GMR element to apply an AC magnetic field H_{ac} (frequency: f) to the GMR line. As shown in Fig 1 (b), the frequency of the output signal from the AC carrier field is 2f while that of the detected field (H_m) is f [2]. Then, small H_m can be distinguished to large AC field by their frequency. Figure 1(c) shows a full-bridge circuit for the proposed method. All the GMR units have the same pin/free configuration but the phase of the AC current of the GMR A is opposite to that of the GMR B. The 2f component is suppressed significantly in the differential output signal.

Dual-free layer type GMR sensor was fabricated on a Si substrate. The film stack was NiFeCr(7)/ IrMn(7)/ CoFe(2)/ Ru(0.9)/CoFe(2)/Cu(3.4)/CoFe(3)/Cu(3.4)/CoFe(2)/Ru(0.9)/CoFe(2)/IrMn(7) (in nm). The GMR unit was 1 mm in length and 5 μ m in width. An Au layer was placed on the GMR line as an AC line via 300 nm-thick SiO₂ layer. The MR ratio was about 5% and the sensitivity was 0.06 %/Oe. A sinusoidal AC voltage (V_{ac}) with a frequency of 10 kHz was applied to the AC line. Two output lines shown in Fig. 1 (c) were connected to differential input terminals of a lockin amplifier. A reference signal from the AC power supply was used for detecting the 10 kHz component of the output signal. Figure 2 shows the output signal as a function of an external magnetic field. V_{DC} was 5V and V_{ac} was 2V_{rms}. The minus sign means a signal with opposite phase. The offset in the H axis is an ambient DC magnetic field mainly from the geomagnetic field. Linear response against the external field is shown. The linearity is about 3%. The output level reached 0.08 V/Oe when V_{DC}=12V and V_{ac}=2V. Regarding the noise level at 10 kHz, this output corresponds to the sensitivity of sub-nT range. The sensitivity can go down to the range of pT with a magnetic field concentrator with the gain of more than 100.

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[1] K. Fujiwara et al: Appl. Phys. Express, 11, 023001 (2018)

[2] K. Tsukada et al.: AIP Advances, 7, 056670 (2017)

Multi-GMR sensors tuned by dipolar coupling

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Spin electronics is widely used for magnetic field sensing thanks to a high sensitivity, over a large range of temperature and frequencies. For low frequencies applications requiring a high spatial resolution or limited footprint, an interesting strategy for noise reduction is to use vertical packaging of multiple Giant Magnetoresistance (multi-GMR) [1].

In this work we report on multiple GMR sensors (up to 12 repetitions) packaging. We observe a crossover in the magnetoresistance responses from step-like behavior to a linear response. This behavior can be explained through micromagnetic simulations as a competition between Neel coupling and additive dipolar coupling which occurs between the GMR's free layers [2].

We evaluate the performances of the sensor (sensitivity, noise and limit of detection) as a function of the number of repetitions. We demonstrate that this multiple stack structure leads to reduced noise and show that we can obtain sensors which are very robust against Random Telegraphic Noise (RTN) when operating at high voltages. Furthermore, we can design sensors with an extended range of sensitivity without reduction of GMR ratio. These results open the path towards spintronics sensors connected and coupled in 3D with reduced noise, compact footprint, using dipolar coupling as a tuning parameter.

M. Silva, D. C. Leitao, S. Cardoso, and P. P. Freitas, IEEE Trans. Magn. 758 53, 1 (2017).
J. Torrejon *et al.*, Phys. Rev. Applied 13, 034031 (2020).

Enhanced coupling factor of integrated on-chip microtransformer for data transmission application

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The miniaturization and integration are important market requirements for emerging electronic devices. To follow these requirements, all components need to be miniaturized. Magnetic passive components are one of the most bulky components on the electronic main board. Therefore, a significant challenge for magnetic devices in the last decade is miniaturization and integration towards system in package (SiP) and system on chip (SoC) [1]. Increasing the switching frequency in the applications is a way for miniaturization and integration of magnetic components. Inductors and transformers with integrated magnetic materials on silicon are potential candidates for high switching frequency applications [2-4]. Developments of thin-film magnetic components in the last years show significant improvement in integration technology and magnetic material research. Recent works show the implementation of microinductors and microtransformers in different electrical applications [5-10].

This paper presents the improved microtransformer with a bar magnetic core fabricated on silicon using thin-film technology [11]. The microtransformer device can be applied for higher frequencies data transfer applications up to 30 MHz. The microtransformer consists of a bar magnetic core and interleaved primary and secondary windings with a minimum distance between primary and secondary turns of 10 μ m. Both windings feature 12 turns each. The dimensions of a single turn are 620 μ m x 40 μ m x 1 μ m (L x W x T). The winding material is copper fabricated using physical vapor deposition (PVD) process. As magnetic core consists of two layers with dimensions of 1400 μ m x 500 μ m with a thickness of 0.4 μ m and oxide gap in-between of 0.8 μ m. The insulation material is a combination of SiO₂ and Si₃N₄. The chip size of microtransformer device is only 1.6 mm x 0.8 mm. The inductance of the device is about 40 nH and the breakdown voltage is about 1.5 kV. Compared to the first design, the new design has a significantly higher coupling factor with a value of about 60%.

Using PVD deposition a copper layer with only 1 μ m is fabricated, therefore the microtransformer device shows high electrical resistance of about 7 Ω . For this reason, the microtransformer is suitable for using in signal applications. In the full paper, the design, fabrication and evaluation of the device and comparison with other devices will be presented.

Analysis of the Low-frequency Noise in Vortex-based Devices for Sensing Applications

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Spin-torque nano-oscillators (STNOs) are spintronic devices based on the excitation of precession modes whose frequencies range from less than 100 MHz up to tens of GHz[1]. A particular configuration for these devices is one exhibiting a vortex magnetization distribution of the free layer in its equilibrium state. The precession of the vortex core induced by the spin-transfer torque in STNOs is especially relevant due to being a model system for the study of the corresponding magnetization dynamics and because it provides excellent signal properties. Furthermore, they are promising candidates for future-generation devices and applications, such as rf generation[2], detection[3] or bio-inspired computing[4]. A less explored application for vortex-based STNOs is as magnetic field sensors. Magnetic field sensors are widely used for a range of applications, such as industrial or biomedical uses. Vortexbased devices provide a large linear range for magnetic field detection[5] and the size of STNOs in the range of 100 nm shall allow achieving a better spatial resolution of the detected magnetic field than typical TMR based sensors. These advantages motivate the study of such vortex devices for magnetic sensor applications. One of the main limitations of magnetic sensors, notably the vortex based ones, is however their intrinsic 1/f noise, which can hinder the device's detection capacity at low-frequencies. Here, we study the low-frequency noise of vortex-based STNOs, comparing the measured noise level in the different possible magnetic states, and extracting the Hooge parameter[6] - used to characterize and compare sensor's noise properties. The one measured in the uniform states is in the same order of magnitude as the ones measured in state-of-the-art TMR sensors with the same RA product, $\alpha = 10^{-11} - 10^{-10} \mu m^2$. In the vortex state, the measured noise level is around one order of magnitude greater than in the parallel (and antiparallel) state, see Fig. 1, due to the high probability of pinning of the vortex core related to material defects and/or inhomogeneities of the magnetic properties of the free layer. Additionally, we study the influence of the vortex dynamics in the low-frequency noise. For a current density above a certain threshold, the spin transfer torques lead to the full compensation of the intrinsic damping of the free layer resulting in the precessional motion of the vortex core (gyrotropic motion) around its equilibrium position[7]. We find that these self-sustained dynamics strongly modify the main characteristics of the STNOs and, in consequence, the achievable performances of magnetic field sensors relying on magnetic vortex dynamics. We present a detailed analysis of these characteristics and how they may relate to the device's noise properties.

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Fig.1 a) Hooge parameter and b) device resistance as an in-plane magnetic field is swept from the parallel to the anti-parallel state and reverse.

[1] S. Kiselev et al., Nature Vol. 425, 380 (2003)

[2] S. Wittrock et al., Phys. Rev. B Vol. 99, 235135 (2019)

[3] A. S. Jenkins et al., Nat. Nanotech., Vol. 11, 360 (2016)

- [4] M. Romera et al., Nature Vol. 563, 230–234 (2018); J. Torrejon et al., Nature, Vol. 547, 428-431 (2017)
- [5] D. Suess et al., Nature Electronics volume 1, pages 362-370 (2018)
- [6] F. Hooge and A. Hoppenbrouwers, Physica 45, 386-392 (1969)

[7] A. Dussaux et al., Nat. Comm., Vol. 1, 8 (2010)

Effect of the deposition temperature on the performance of AMR Sensors Based on La2/3Sr1/3MnO3 thin films

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The final goal of our project is to develop a device that acts as a bypass for restoring neuronal signals in regions with spinal cord injuries^[1]. To detect such signals, a magnetic field sensor can be employed, placed near the spinal cord. This in turn sets several constraints for the sensor itself. It must be of reduced size, operate at zero or very small external biasing field and achieve low detectivity at low frequency, while operating at body temperature. The detectivity of a magnetic sensor is determined both by its sensitivity and its intrinsic noise. If the target signal is in the low frequency region, which is the case for biomedical applications, Anisotropic Magnetoresistance (AMR) sensors become competitive when compared to Tunnelling Magnetoresistance and Giant Magnetoresistance technologies, due to its low 1/f noise, even if they do not reach same sensitivity values. Additionally, AMR sensors can be of much simple fabrication, not requiring the stacking of several thin films.

We will present results obtained with AMR sensors made of 60 nm thick epitaxial $La_{2/3}Sr_{1/3}MnO_3$ thin films, which present low intrinsic noise^[2], deposited on vicinal 8° SrTiO₃ substrates and etched in a Wheatstone Bridge design (Figure 1). Pulsed Laser Deposition (PLD), sputtering, photolithography and Ion Beam Etching were used for in-lab samples fabrication. The use of a vicinal substrate, which presents a surface miscut angle regarding the crystallographic plane, was selected so as to induce uniaxial anisotropy and an easy magnetic axis along the step edges^[3]. The measured AMR curves can be compared to the expected behavior when considering the Stoner-Wohlfarth model for coherent magnetization reversal. In order to achieve a higher sensitivity, thus lower detectivity, PLD deposition was performed at different temperatures, which leads to a change in thin film T_c and therefore affects the anisotropy field H_a. A clear reduction in H_a can be seen when changing the deposition temperature from 730 °C to 680 °C, achieving detectivity values at 310 K as low as 2.7 nT·Hz^{-1/2} at 1 Hz and 200 pT·Hz^{-1/2} at 1 kHz in a single-layer sensor without using performance enhancing techniques such as flux focusers.



[1] http://www.byaxon-project.eu/ FET Open 2017-2020

[2] L. Méchin et al 2013 J. Phys. D: Appl. Phys.46 202001

[3] P. Perna et al Adv. Funct. Mater. 2017, 27, 1700664

Peculiarities of measuring local magnetic fields using a scanning magnetic microscope based on a magnetoimpedance sensor

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The glass coated Co-rich amorphous ferromagnetic microwires with small metallic core diameters $d = 5 - 30 \mu m$ are promising for various technological applications. In particular, Co-rich microwires with a negative nearly zero magnetostriction show giant magnetoimpedance (GMI) effect with a high GMI ratio [1]. Such microwires are used to develop a new generation of sensitive miniature GMI magnetometers. As has been shown recently [2], an offdiagonal magnetoimpedance sensor based on a microwire was used as a measuring sensor of a scanning magnetic microscope. The important characteristics of magnetic microscope are magnetic sensitivity and spatial resolution. These parameters are determined by the measuring sensor.

In this work the scanning magnetic microscope with spatial resolution of order of 20 micrometers has been developed and tested. The off-diagonal GMI-sensor was based on a 4 mm microwire segment and a measuring coil wound around the microwire. The GMI-sensor was placed perpendicular to the sample surface. The minimal distance *h* between the microwire's tip and the sample surface could be up to 20 micrometers. Relative movement of the sample and the GMI-sensor was performed by means of X-Y-Z positioner. Measurements were carried out within a magnetic shield to reduce the effects of external magnetic fields.

To test the capabilities of the developed scanning magnetic microscope, we used tested current carrying samples and segments of Co-rich amorphous microwires. The current-carrying samples were made from thin copper wire with a diameter of 0.01 mm. We used straight lines and square samples of different sizes as samples. During measurements, a dc currents from 1 mA to 10 mA were passed through the samples. During measurements of Corich amorphous ferromagnetic microwires, we used segments with a length of 4 mm, as samples. We measured the stray magnetic fields of microwire samples in zero magnetic field and in a longitudinal applied magnetic field with an amplitude of up to 60 microTesla. Obtained data indicated that the GMI-sensor can detect small and highly localized magnetic fields. Features and characteristics of the GMI-microscope developed are discussed in the presented work.

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[1] V. Zhukova, A. Chizhik et al., IEEE Trans. Magn. 38, 3090 (2002)

[2] S. Gudoshnikov, V.Terasov et al., J. Magn. Magn. Mater., 510, 166938 (2020).

Detection limit normalization and study of TMR magnetic field sensors

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Magnetic tunnel junction (MTJ) it is a promising candidate for the development of ultrasensitive magnetic field sensor (TMR sensor) capable to detect extremely weak magnetic fields (0.5) opening the possibilities for biomedical applications . The performance of these type of sensors is limited by the high low frequency noise (1/f) which appears due to the presence of magnetic domain fluctuations or by defects inside the insulator barrier. In this work, we study the normalization and voltage bias effect on the detection limit in TMR sensors based in MTJs. The multilayer stack is Ta(5)/SyF /MgO/SyF /Ta(10), where the synthetic ferrimagnet structures are SyF: PtMn(25)/ CoFe(2.3)/Ru(0.83)/CoFe(1)/Ta(0.1)/CoFeB(1.5) and SyF: CoFeB(1.5)/Ta(0.1)/CoFe(1)/Ru(2.6)/CoFe(2)/PtMn(16) (nanometers). Samples were processes using optical UV lithography to define the diameter (D) and number of MTJs circular pillars in serie (N) inside the sensor. The RA product was typically around 13-20 kΩµm at 0 mT and the TMR 200 % at room temperature. In order to provide a good interpretation of the results we propose a theoretical model to extract the real output voltage noise from standard electronic circuits. This model was used on a ¼ Wheatstone bridge setup, where the resistive elements were modelled as fluctuating resistances. Our TMR sensors show a sensitivity of 4 %/mT (<0.1 %/mT) at 0mT (P state at -100 mT). The 1/f noise was quantified by the Hooge-like parameter, which allow to compare the 1/f noise of different types of TMR sensors. We observed that well known equation works only below a threshold voltage, due to the non-linearity of the I-V curve related maybe to different symmetry conduction bands. At low bias voltage, devices show higher values at 0 mT (3.10⁻⁸ μm) than in the P state (2–4.10⁻⁹ µm). An interesting feature for applications is the almost constant detectivity (100 nT/Hz^{0.5} at 10Hz) as a function of the bias voltage. This is a result of the compromise between the output noise voltage and the sensitivity. The subsequent output power and detection limit normalization allow us to design new TMR sensors with optimal conditions.

High accuracy of absolute magnetic position measurement systems based on TMR-Sensors

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In recent years, magnetic position systems are getting more and more attractive as the accuracy of these systems becomes comparable with optical position measurement systems while offering significant price advantages. In contrast to optical systems, magnetic systems are regarded as very robust and able to operate in dusty and oily environments. One approach to improve accuracy is to replace hall sensors with magnetoresistive sensor (MR) technology like AMR, GMR and TMR¹. Recent developments on MR technology provide a competitive and reliable solution for position control systems based on magnetic encoders. MR sensors and especially TMR sensors offer a high spatial resolution and work as high sensitivity devices, thus surpassing other magnetic field detection techniques.

Magnetic position systems for linear and angular applications consist of a multipole ferromagnetic scale as well as a magnetic sensor. The reading distance (RD) between sensor and scale is determined by the magnetic material and by the length of the magnetic poles (PP) as well as the reading technology. Motion control applications, for example in industrial robotics and machining tools, require high accuracy up to single digit micrometer range combined with a large reading distance range. There are different approaches for absolute and incremental position measurement with various technologies. The advantages and disadvantages of these methods are described. To achieve a positioning system with a high accuracy, a magnetic scale is just as important as the sensor. We demonstrate how the pattern affects the system accuracy.

Up to now, measurements in high accuracy were only possible in a narrow distance range to the scale in short vicinity of the scale due to technology limitations. There is a strong need for magnetic sensors and scales which address this issue. Moreover, it is required that the same TMR sensor can read the strong fields from the media and the small fields equally well; this is quite challenging.

The optimization of magnetic positioning systems entails the improvement of the magnetic pattern for the scales as well as the sensor. As part of the EU-funded "MASMA"³ project, we developed TMR-sensors with tunable linear range, as to enable a large RD excursion during the scale measurements in real situations. We will present the fundamentals of this sensor: The S/N ratio, the magnetic and temperature stability, the thermal and field stress response and the sensor bandwidth. Afterwards we will show a first application for this new sensor design and the reached accuracy.

[1] C. Zheng *et al., IEEE Transactions on Magnetics*, vol. 55, no. 4, pp. 1-30, April 2019, Art no. 0800130, doi: 10.1109/TMAG.2019.2896036.

[2] J. Paul et al., proceedings of MikroSystemTechnik Kongress2017, 23.-25.10. 2017, München/Unterschleißheim, pp. 223-226
[3] https://cordis.europa.eu/project/id/858934/de

Application of magnonic crystals in magnetic particles detection

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Recently, it was shown that magnetic patterned films based on antidot arrays can be employed as high-sensitivity magnetic field detectors, exploiting their dynamic behavior in the Gigahertz range. The sensing mechanism was demonstrated for the case of magnetic nano/microparticles adsorbed on the surface of permalloy nanostructured films [1-3]. In particular, the magnetic stray field generated by the particles interact with the magnetization state of the antidot array, producing a measurable shift in its ferromagnetic resonance (FMR) frequencies. The geometrical properties of the patterned film microstructure can be tuned to maximize the generated signal.

In this framework, we aim at investigating, from a modeling point of view, the dynamic response of permalloy antidot arrays with hexagonal lattice, in presence of different types of magnetic particles with variable size and saturation magnetic moment. Particular attention is given to the influence of particle concentration on the FMR signal, in order to determine a relationship useful for particle quantification. The calculation of the sensing element response is performed by means of a parallelized micromagnetic code, which was engineered to run on Graphics Processing Units to efficiently solve the Landau-Lifshitz-Gilbert equation [4].

The final spatial distribution of magnetic particles on the antidot array surface is predicted by means of a Newtonian dynamics model. This enables us to describe particle transport considering the effects of viscous drag force, the inter-particle magnetostatic interactions and the action of the localized magnetic force due to the antidot array [5]. The final objective is a parametric analysis, aimed at determining the effects of particle concentration on the sensing element FMR spectra, in order to define a quantification relationship associated with the shifts in the FMR frequencies of edge, extended and localized modes.

- [1] P. J. Metaxas et al., Applied Physics Letters 106, 232406 (2015).
- [2] M. Sushruth et al., Physical Review Applied 6, 044005 (2016).
- [3] A. Manzin et al., Scientific Reports 6, 22004 (2016).
- [4] O. Bottauscio, A. Manzin, J. Appl. Phys. 115, 17D122 (2014).
- [5] M. Vicentini et al., JMMM, 167234 (2020).

Electrical detection of spin resonance in DPPH free radicals and manganites

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Detection of electron paramagnetic resonance (EPR) using a microwave cavity resonating at a fixed frequency (between 9 and 10 GHz) remains the most popular method until now. Here, we report a cavity-less technique which makes use of only an impedance analyzer and a copper strip coil to detect L-band EPR (f = 1-3 GHz) in the standard EPR marker 2,2-diphenyl-1-picrylhydrazyl (DPPH) and also in manganite series La1-xCaxMnO3 (x = 0.1-0.6). Our method relies on measuring the magnetoimpedance (MI) response of DPPH through a copper stripcoil that encloses DPPH and/or passing high frequency current directly through manganites. In contrast to commercial EPR which measures only the field derivative of power absorption, our method enables us to deduce both absorption and dispersion. Changes in resistance (R) and reactance (X) of the copper strip while sweeping an external dc magnetic field, were measured for different frequencies (f = 0.9 to 2.5 GHz) of radio frequency current in the coil. R exhibits a sharp peak at a critical value of the dc magnetic field, which is identified as the resonance field and X shows a dispersion at the same frequency. The data were analyzed to obtain line width and resonance field parameters. The resonance field increased linearly with frequency and the obtained Landé factor of 1.999 is close to the accepted value of 2.0036, measured in X-band. The composition dependence of the electrically detected ESR signal intesnsity and potential mechanisms in manganites will be discussed. We believe that this new and simple technique can be exploited to study spin dynamics in spintronic materials.

Electrical characterization of the azimuthal anisotropy of NiCo-based ferromagnetic nanotubes

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Ferromagnetic one-dimensional conduits are subject to intensive research. These are model systems for domainwall motion and spin waves, and have also been proposed as a platform for 3D magnetic storage or field sensor [1]. While wires have been largely addressed, nanotubes are only emerging especially on the experimental side, while they promise improved suitability for spintronics as multilayered core-shell structures can be synthesized. We recently reported the unexpected azimuthal curling of magnetization in electroless Co-rich CoNi nanotubes [2], attributed to curvature-induced magneto-elastic anisotropy. Here, we measure precisely their associated azimuthal anisotropy through anisotropic magnetoresistance. NiCo-based ferromagnetic nanotubes were fabricated by electroless plating in polycarbonate templates. Tubes have diameter in the range 160 to 450 nm, wall thickness from 20 to 65 nm, and length several tens of µm. Following template dissolution and dispersion on highlyconductive Si wafers, nanotubes were contacted using laser lithography (figure 1a). Transport measurements were carried out at varying temperature, applied field magnitude and direction. At 300K the material resistivity is 2*10⁻ 6 Ω m, one order of magnitude higher than in the bulk, which may be related to the nanocrystalline nature of the tubes. The AMR is 0.12% at 300K and 0.3% at 10K. From R(H) loops with magnetic field applied along the tube axis, we infer the magnetization process and thereby extract the anisotropy field, of the order of K_{eff} ~7.5*10³ J/m³. We are currently extending these measurements to various diameters and tube wall thickness to draw a general picture of the curvature-induced anisotropy.

[1] Magnetic nanowires and nanotubes, M. Stano, O. Fruchart, in: Handbook of magnetic materials vol.27, Ed. Ekkes Brück, North Holland (2018).

[2] Michal Stano, Olivier Fruchart et al., SciPost Phys. 5, 038 (2018)

Changes in property of thin film magnetoimpedance element by Joule heating

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The thin film is effective for miniaturization of magnetoimpedance (MI) element which is contributed to magnetic field sensor with higher sensitivity. Controlling magnetic anisotropy is quite important to obtain large impedance changes subjected to external magnetic field. When using Co-based amorphous material, the magnetic anisotropy can be controlled by field annealing in vacuum. The furnace for field annealing is generally large-scale and is impossible to control localized anisotropy as well as consume a lot electric power for heating and applied large field. On the other hand, Joule heating, which a current is directly applied to an element, achieves compact equipment with low power consumption. The Joule heating is utilized for wire and ribbon configuration to improve MI characteristics: relaxation of internal stress induced during material production. However, there are few reports that Joule heating applies to thin film. Also, no reports try to control magnetic property itself exists, including wire and ribbon case. Therefore, we tried to change or improve the magnetic properties of thin-film MI element using Joule heating as a feasibility study.

The thin-film elements fabricated by amorphous CoZrNb with 20 or 40 micrometers width, 2 micrometers thick and 1 mm long were used for investigation. For Joule heating, a direct-current of 50-200mA was applied to elements using a wafer probe in the atmosphere with ranged from 30 seconds to 8 minutes. The effect of applied field during heating is also investigated. In the case of element with 40 micrometers width, the field intensity where the impedance has a peak shifts to lower field, the annealing up to dc-current of 130 mA with applied field parallel to length direction. Additionally, the MI property drastically was deteriorated with 200 mA and 1 minute annealing. As to simulation result, the temperature of element rises up to around 720 K with 200 mA, this temperature is over the crystallization temperature of CoZrNb. Thus, the degradation of MI property is caused by crystallization: it is well known that soft magnetic property is lost by crystallization. The large reduction of resistivity of the element after heating is also an evidence of crystallization. On the other hand, the MI property has no variation after Joule heating with lower current intensity and without applied field, which means the changes in MI property are not attributed to relaxation of internal stress. The heating time is very short time, therefore, the influence of oxidation is also not dominant in this study: at least we did not confirm remarkable increases in resistivity of the element. The obtained results show the potential of controlling MI property of thin-film element using Joule heating with applied field.
New lab-made feedstock for magnetic composite filaments for additive manufacturing

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Successful 3D printing via material extrusion for additive manufacturing of parts with functional properties requires filaments with tight dimensional tolerances and homogenous dispersion of magnetic fillers, which are usually fabricated using feedstocks produced by industrial techniques.

Here we present an original way of producing the initial magnetic composite feedstock for filament extrusion, creating printable filaments with good homogeneity of well-dispersed magnetic particles throughout the matrix by using single screw extrusion. The proposed technique avoids the typical industrial reliance for which initial feedstock for making filaments is usually fabricated by roll mill kneaders, mixers, injection molding and/or twinscrew compounding. Such specific machinery is not accessible in many laboratories and studies are also highly dependent on industrial specifications and restrictions. An alternative, commonly used procedure, implies multicycles of extrusion (at least six runs) at the expense of less predictability in filament composition and more extended work time. In contrast, results from the magnetic composite filaments with good homogeneity and printability using a desktop extruder. Good particle dispersion in the composite filament is further demonstrated by X-ray tomography characterization, enabling a construction of the 3D projection of the embedded magnetic particles. Moreover, this non-chemical method is not restricted to polymer compositions or size / characteristics of the particles, not being limited to working with magnetic or metallic particles. Furthermore, our magnetic filaments are functionally characterized by magnetometry and Kerr microscopy. The level of detail of the printed parts demonstrates the good quality of the filaments produced by the method presented in this talk.

[1] Á. Díaz-García et al., Materials Today Communications 24 (2020) 101049.

Franco¹

Ultra-low magnetic field AMR magnetometers study based on LSMO for biomedical applications

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Brain-machine interfaces record the neural activity at the brain and send those recordings to a computer to control a robot, or to transfer signals to electrodes placed over the limbs, allowing spinal-cord-injury patients to gain some functions. Being able to address directly the spinal cord will give us the change of a more compact and simplified solution. We propose the use of magnetoresistive materials to build sensors of the magnetic field created by the neural activity that do not need an intimate contact with the neural tissue. Magnetic fields created by biological electric currents in the nervous system have specific characteristics such as very low amplitude, rapid decrease with distance, low frequency pulse duration, etc [1]. Historically, magnetometers used for detecting neural signals were superconducting quantum interference devices (SQUID) or optically pumped magnetometers (OPM) because of their low detectivity but they have difficult working conditions, such as low operating temperatures, magnetically shielded environment, etc. Recent studies have started using magnetoresistive sensors that, although having worse detectivity than the magnetometers previously mentioned, can be scaled and shaped to the convenience of the experiment and work at room temperature [2]. The aim of the project is to develop an implantable device, therefore the magnetic sensor's size has to be no bigger than a few millimeters in addition to work at room temperature. In order to address these specifications, the project has developed a novel magnetoresistive sensor based on the anisotropic magnetoresistive effect (AMR) made of $La_{1-x}Sr_xMnO_3$ (LSMO) which can be easily scaled, shaped and has the best performance at temperatures close to the human body temperature. In this work, we present a full characterization of the sensitivity and detectivity of the sensors at different temperatures and different bias voltages to determine the optimum working conditions and stablish their maximum detectivity. We test the sensors against the signals produced by an specifically-build artificial neuron.



Figure 1: Left image presents the LSMO sensor batch. Right graph shows a sensor's AMR and sensitivity dependence with temperature highlighting its best performance at the human body temperature.

[1] Wijesinghe, R. S.; Journal of Electrical Electronic Systems 2014, 3, 2332-0796

[2] Caruso, L., et al.; Neuron 2017, 95, 1283-1291

Modelling of compliant magnetoresistive sensors behaviour using a generalized macrospin approach

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Compliant magnetoresistive sensors for micro/nano-scale magnetic field sensing technology open up bright perspectives in high precision instrumentation¹, skin-like human-machine interfaces or smart wearables ^{1,2} applications. By incorporating magnetic functionalities within flexible substrates, a low-cost and lighter alternative to its rigid Si-based counterparts can be attained, with intrinsic shaping capability. Still the microfabrication of flexible sensors poses some challenges, as the presence of non-negligible magnetostriction effects and increased substrate surface roughness may limit sensor performance. We present a modified Stoner-Wohlfarth model as a tool to closely predict, control and better understand the role of magnetoelastic anisotropy on compliant magnetoresistive sensor's performance. By providing experimental input parameters of the sensor's magnetic and mechanical properties, our analytical model is able to numerically calculate the elastic deformation of the thin-film multilayer composite due to residual stresses and applied external bending, and the expected sensor magnetic and electrical response considering the sensor's micrometric geometry. Fig.1(A) demonstrates the modelled magnetic field dependence of the normalized electrical resistance for different levels of applied tensile stress or applied bending radii, R_b. The system is a 2x100 µm² top-pinned spin-valve sensor [Ta 2/Ni₈₀Fe₂₀ 2.5/Co₈₀Fe₂₀ 2.8/Cu 2.8/Co₈₀Fe₂₀ 2.6/Mn₇₄Ir₂₆ 7/Ta 5 (nm)], fabricated on 25µm-thick PET foil. In the presence of mechanical stress, the magnetoelastic field ($\mu_0 H_\sigma$) comes into play, mostly arising from the sensor's free-layer, namely, Co80Fe20 which is known for having a non-negligible magnetostriction constant $(\lambda s=32 \times 10^{-6})^3$. By applying tensile stress along the sensor longitudinal direction (decreasing the curvature radii, R_b), the free-layer magnetization stabilizes along the stress axis, creating an extra source of magnetic anisotropy in the overall energy function. It reinforces the energy barrier mainly induced by shape anisotropy while competing with the uniaxial induced anisotropy, directly affecting the saturation field. In Fig.1(B) the experimental curve and calculations considering our model are presented. As the saturation field proves to be dependent on the magnetoelastic anisotropy, $\mu_0 H_\sigma$ (0.15mT) is estimated by considering the difference between the sensor experimental saturation field (3.20mT) and the calculated value neglecting magnetostrictive effects (3.05mT). The presence of a dipolar magnetostrictive term due to the pinned layer is also present, affecting the offset field. Although simplistic in its formulation, our model was validated to qualitatively control and predict linearization conditions in micrometric magnetoresistive sensors fabricated both on conventional and non-conventional substrates. It should also be noted that the system neutral axis and the stress-strain distribution across the sensors layers can be calculated by our model, a particularly useful advantage for optimal encapsulation strategies.



Fig.1(A) μ_0 H dependence of normalised electrical resistance for different radii of curvature R_b and **(B)** comparison between the experimental data and the calculations obtained with the energy minimization model of 2x100 μ m² sensors on PET.

- [1] Ch. Zheng et al., IEEE Trans. Magn 55,4, 2019
- [2] C. Bermúdez et al., Sci. Adv., 4,1, 2018
- [3] R. M. Bozorth, Ferromagnetism. USA: IEEE Press, 1993

Study of the free layer influence on electrical/magnetic noise in magnetic tunnel junction-based Magnetoresistive sensors

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When targeting the detection of small magnetic fields, sensors should have large sensitivity and low noise, for enhanced signal to noise ratio/detectivity. In the case of micrometric/nanometric magnetic tunnel junction (MTJ) MR sensors, low frequency applications such as biomedical signal detection[1] are still limited by 1/f noise, especially as the sensor size decreases. Although packed sensors[2] and magnetic flux guides[3] have been researched, a systematic study of the thin film materials impact on the electrical noise characteristics of MTJ MR sensors is still lacking and could hold the answer for the next generation of low field detection sensors. In this work, we evaluate electrical and magnetic noise in MgO MTJ stacks, with varying CoFeB/NiFe ratio in the free layer (FL). The MgO stacks were deposited in a TIMARIS machine by PVD, with structure Si (substrate)/Ta 50/[Ru 180/Ta 30]x3/Ru 50/MnPt 200/CoFe 22/Ru 7/CoFeB 30/MgO/FL/MgO/Ta 50/Ru 50 (thicknesses in Å), and were microfabricated into pillars with dimensions $20x2 \ \mu m^2$. Magnetotransport characterization is done in a 4 probe configuration, with applied field from -140 Oe to 140 Oe. Low frequency noise characterization is done systematically at a bias voltage of 50 mV, with electrical signal amplification of 100x through an amplifier with DC-100 kHz bandwidth. Focusing on the magnetotransport results, the different stacks show TMR ranging from 140% to 175%, with the exception of purely NiFe 60 FL, with a smaller TMR of 49%, owing to the absence of CoFeB near the MgO, required for high TMR[4]. The normalized R(H) curves show increasing slope with decreasing CoFeB/NiFe ratio, owing to a larger component of NiFe. However, the purely NiFe FL shows a non-linear R(H) curve, with Barkhausen jumps. Concerning the electrical noise characterization at saturation in the parallel state, the Hooge parameter α_{H} , extracted from the 1/f noise at frequencies under 10kHz, is of the order of (2-3) x 10⁻⁹ μ m², and generally decreases when the CoFeB/NiFe ratio increases. However, the NiFe only FL shows an exception, where $\alpha_{\rm H}$ decreases significantly, although the 1/f slope (measured by the y parameter) is also significantly different. The noise in the linear region increases for the characterized stacks when compared to saturation, owing to magnetic contributions to noise[5]. Noise characterization on MgO MTJs gave evidence of reduced low frequency noise at saturation for pure NiFe FL. The results show promise for pure NiFe FL stacks, where TMR is reduced but electrical noise is also significantly reduced. However, the linearization of such a stack, as well as the noise in the linear region, must be addressed for sensing applications.



Figure 1: a) Normalized R(H) curves of rectangular 20x2 μ m² MTJ sensors with varying FL. b) Noise spectra of the sensors, measured at the parallel state. c) α_{H} comparison for the measured sensors at the parallel state (saturation) and in the linear region, with other values from reference (adapted from [6]). d) α_{H} and γ parameter for the measured sensors, as well as α_{H} normalized by MR sensitivity for linear region noise measurements.

- [1] M. Pannetier-Lecoeur et al., APL, 98(15), 153705 (2011)
- [2] J. Valadeiro et al., IEEE Trans. Magn., 51(1), 1-4 (2015)
- [3] M. Silva et al., IEEE Trans. Magn., 55(7), 1-5 (2019)
- [4] S. Yuasa et al., J. Phys. D: Appl. Phys., 40(21), R337-R354 (2007)
- [5] C. Ren et al., PRB, 69(10) (2004)
- [6] A. Silva et al., Eur. Phys. J. Appl. Phys., 72(1) (2015)

Smart Fingertip biosensor for food quality control

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Automated technologies for manipulation and quality inspection of fruits has attracted great interest from the food industry [1]. The development of non-destructive processes to handle and assess fruit quality along the food chain may lead to the reduction of food waste, with major impact for high perishable products [2].

Herein, a texture sensor based on highly sensitive hair-like cilia receptors [3-5] to allow a quick fruit quality evaluation is proposed. The operating principle of the device is based on detecting changes of the stray magnetic field created by an elastic magnetized cilia array when deflected according to the fruit skin texture, hence changing the sensors' signal.

The texture sensor combines a permanently magnetized and highly elastic nanocomposite cilia array realized on top of magnetic sensing elements (spin-valve sensors). The nanocomposite is made of polydimethylsiloxane (PDMS) and NdFeB magnetic microparticles. Three cilia arrays of fixed height (150μ m) but varying in cilia diameter/space between them: 150/100, 100/150 and 100/100 (in μ m) were fabricated. The cilia arrays were implanted over giant magnetoresistive (GMR) sensors in a Wheatstone bridge configuration, with a sensitive area of 3 mm², and fully characterized. All devices shown to be capable of detecting cilia deformation along the GMR sensors sensitive direction with signals up to 9 mV.

The sensing device with 150µm diameter cilia was used to assess the capability of detecting blueberry fruit skin texture (Fig. 1a). The experiments were performed by holding the fruit with tweezers and passing it over the sensor, parallel to the sensitive direction. Figure 1b shows that the sensor is capable of differentiating fruits with smoother skin from rougher ones, as the red curve (unripe fruit) has lower signal variations than the others. Moreover, Figure 1c indicates that signal amplitude and ripeness stage are proportional.

Further improvements include optimization of the sensor architecture, by increasing the array dimensions, number of cilia and size of sensing area. Also, the integration of a gas sensing layer on the ciliary structures, to enable ethylene detection, will add a complementary non-destructive capability to this device. Ultimately, this multiparametric biosensor has potential to be integrated in a robot fingertip enabling automated fruit quality analysis.



Figure 1: (a) Photograph of one developed device. b) Skin roughness measurements of three blueberry fruits in different ripeness states. c) Amplitude of four measured signals for each maturation state, corresponding to the fruit image bellow (from left to right: unnripe, ripe and overripe).

[1] Z. Hussein, O. A. Fawole, and U. L. Opara, Hortic. Plant J., pp. 1–28, 2019

- [2] M. C. N. Nunes, J. P. Emond, M. Rauth, S. Dea, and K. V. Chau, Postharvest Biol. Technol., vol. 51, pp. 232–241, 2009
- [3] A. Alfadhel and J. Kosel, Adv. Mater., vol. 27, no. 47, pp. 7888–7892, 2015
- [4] P. Ribeiro et al., IEEE Robot. Autom. Lett., vol. 2, no. 2, pp. 971–976, 2017
- [5] A. Alfadhel, M. A. Khan, S. Cardoso De Freitas, and J. Kosel, IEEE Sens. J., vol. 16, pp. 8700–8705, 2016

Controlled operation point on highly sensitive spin valves coupled with magnetic flux concentrators

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Magnetic sensing technology are widely used in several applications wherein magnetoresistive (MR) sensors are stated due to its high performance in harsh industrial environments or highly sensitive biomedical experiments¹. They work at room temperature with an established nTesla minimum field detectable at low frequencies. However, an improvement on detectivity can push forward the MR technology as a high precision tool. The key parameters of a MR sensor performance are given by sensitivity, noise, power consumption and mechanical, thermal and magnetic robustness. The improvement on sensitivity can be achieved by increasing the MR ratio or by reducing the linear range through the incorporation of magnetic flux concentrators (MFC). Previous works report the optimization of the MFC geometry² and gap size to improve the sensitivity achieving values of pT at low frequencies. However, at zero field the sensor is saturated requiring a constant field along the sensitive direction of the sensor to shift its response to the most sensitive point. Monolithically integrated permanent magnets and external coils based strategies impose a constant magnetic field and increase the size of the device, respectively. Here the strategy is to implement a current line on the device to apply a controlled magnetic field which can be easily tuned by the current without compromise the spatial resolution. Our sensor transfer curve shows a coercive field around 1 Oe and a linear range of 0.1 Oe. A precise control is needed to configure the sensor within its linear range. Simulations were carried out in order to study the position and distribution of the lines and the bias current magnitude. The simulation results through a finite element method show that if the line is designed close to the yoke of the MFC (fig. 1(a)), creates a local magnetic field which tends to close around the line even near a high magnetic permeability medium (MFC). Consequently, the solution is to design the line in the vicinity of the MFC pole (d= 5 μ m) to promote a magnetic field flow through the gap to the other MFC. Different number of lines were tested (i) on a single MFC side or (ii) symmetrically distributed over both MFC. A higher number of current lines under the same bias equally distributed on both sides is more beneficial enhancing the field at the gap. Finally, a balance between the number of lines, the power consumption and heating density results into a design of 10 lines even distributed over both MFC with less than 7 mA per line in order to promote the desired shift (Figure 1 (b)).



Fig. 1: (a) Schematics of the magnetic device composed of two MFC and a MR sensor between the poles. (b) Magnetic flux density at the MFC pole created by 10 current lines distributed over both MFC, enhancing the magnetic field on the sensor area.

[1] K. Fujiwara, M. Oogane, A. Kanno, M. Imada, J. Jono, T. Terauchi, T. Okuno, Y. Aritomi, M. Morikawa, M. Tsuchida, N. Nakasato, and Y. Ando, Appl. Phys. Express **11**, 23001 (2018).

[2] J. Valadeiro, D.C. Leitao, S. Cardoso, and P.P. Freitas, IEEE Trans. Magn. 1, 1 (2017).

Optimization of Asymmetric Reference Structures Through non-evenly Layered Synthetic Antiferromagnet for Full Bridge Magnetic Sensors based on CoFeB/MgO/CoFeB

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Magnetoresistive (MR) Wheatstone bridges are widely implemented in industrial applications due to their performance. Anisotropic and giant MR sensors are maturated technologies that provide a reliable solution as positioning sensors where a symmetric resistance variation in full bridge topologies can be obtained through a post-process discrete assembly or a combination of multi pinning directions. The former is a straightforward technique counterweighted by alignment errors and large footprint devices while the latter is associated to high production cost. Magnetic tunnel junctions with CoFeB/MgO/CoFeB interfaces require high annealing temperatures being more suitable a multi deposition method compatible with a single annealing step. A double deposition process of non-evenly layered synthetic antiferromagnet (SAF) structures is a successfully demonstrated concept that provides asymmetric references that fully preserves the sensor linearity and enhances the performance as a MR Wheatstone bridge. Nevertheless, industry provides MR sensors for angular positioning and motion applications that operate upon a saturated the free layer requiring magnetically stable references to enhance the device accuracy and extend its operating range. Therefore, a theoretical model of MR structures composed by a double and a triple magnetic layered SAF is proposed and numerically solved in order to understand the influence of the thickness of each magnetic layer and the coupling strength between them. The theoretical model of triple magnetic layered SAF structure pinned through exchange coupling with an antiferromagnetic (AF) layer follows the formulation:

 $E = -M_{P}t_{P}H\cos\theta_{P} - A_{ex}\cos\theta_{P} - M_{R_{1}}t_{R_{1}}H\cos\theta_{R_{1}} + A_{R_{1}P}\cos(\theta_{R_{1}} - \theta_{P}) - M_{R_{2}}t_{R_{2}}H\cos\theta_{R_{2}} + A_{R_{2}R_{1}}\cos(\theta_{R_{2}} - \theta_{R_{1}})$

where $M_P t_P$ and $M_{R1}t_{R1}$ are the magnetization thickness product of the pinned layer (PL) and reference layers (RL), A_{ex} is the exchange coupling energy per unit area between the pinned layer and the AF layer, while A_{R1P} and A_{R2R1} are the indirect exchange coupling energy per unit area between RL1-PL and RL2-RL1. Consequently, the net moment of the structure is obtained through the angle between the magnetic field H and each layer magnetization that minimizes the system energy. The expected behavior of a double (AF/CoFe/Ru/CoFeB) and triple (AF/CoFe/Ru/CoFe/Ru/CoFeB) magnetic layered SAF structure is illustrated in Figure 1 (a) and (b), respectively. The double system is characterized by an antiparallel state between the PL and the RL1 where its negative saturation magnetic field is enhanced through a lower t_P while a symmetric antiparallel state around zero is tuned by tR1. However, the triple magnetic layered system provides up to two antiparallel states at a negative external field and around zero magnetic field. The former emerges as the indirect exchange coupling A_{R2R1} increases or t_{R1} decreases promoting the full rotation of RL1. Furthermore, as the negative plateau is enhanced the antiparallel state around zero field degrades since its negative saturation field is pushed towards zero which decreases the magnetic stability of the device when compared with a standard double system. Nevertheless, a uniform plateau and balanced SAF can be tuned by a compromise between a higher t_{R1} and lower t_{R2} .



Fig. 1: Theoretical behavior of double (a) and triple (b) magnetic layered SAF structures.

New applications for magnetic ink – how to revive an old technology

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Enabling Fe-based magnetostrictive materials for composite damage detection

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Magnetostrictive sensors are one technology that is being considered for use in structural health monitoring (SHM) of aerospace composite components. SHM uses non-destructive monitoring that is integrated with the material, to essentially functional them, to create smart materials. As the carbon fibre reinforced polymer composites (CFRPs) used experience non-trivial failure due to their complex structure, functionalizing them can: 1) Save manpower time and costs from manual inspection, 2) Reduce the potential of human error, 3) Reduce costs associated with automated visual systems, 4) Enable detection of sub-surface and barely-visible damage, and 5) Allow *in-situ* inspection during service. Previous studies have found that magnetostrictive sensors can be comparable to existing technologies such as fibre optics and piezoelectric sensors [1] but are inexpensive alternatives. Three characteristics must be exhibited in order to demonstrate SHM capability: 1) Detection of damage location, 2) Determination of damage intensity, 3) Quantification of damage type.

The magnetostrictive SHM consists of FeSiB ribbons embedded within the CFRP, with an array of coil sensors located along the ribbon length to measure the change in induction. Comparing the output of the coil sensors will provide information on the damage within the composite. In this work, we constructed a data acquisition system that is capable of recording data from 4 inductive sensors concomitantly. The inductive sensors were found to have a strain sensitivity resolution of 1.9 μ Strain when paired with a single FeSiB ribbon; a previous prototype utilizing 3 ribbons and a single sensor showed a strain sensitivity resolution of ~20 μ Strain [1].

The experimental design utilizing 70-turn inductive pancake sensors, FeSiB magnetostrictive actuators, and an Arduino microcontroller-based data acquisition system was setup onto a VTC401 composite coupon. Fabricated coupons were tested for three different failure modes following ASTM standards (D5528, D7905, E399); results were validated with video data of the crack propagation. The results for Mode 1 demonstrate a decrease in inductance as the crack propagates past each sensor, showing good validation of crack propagation in the sample with magnetostrictive actuator readings. The change in inductance across each sensor position was shown to be correlated to the failure mode; thus, the sparse sensor array was capable of distinguishing the damage type. The study demonstrates that the current setup can satisfy all three of the SHM requirements at the lab scale. Further post-processing has been demonstrated utilizing algorithms to determine similarities between each sensor. Doing so allows damage (as a function of inductance) to be characterized independent of the background noise, increasing sensitivity despite a lower strain sensitivity resolution. This strategy can also be used to determine damage location for impact damage by triangulating the inductive sensor response. The triangulated data was found to be in good agreement with the actual damage location.

[1] Z. Leong, W. Holmes, J. Clarke, A. Padki, S. Hayes, and N. A. Morley,', IEEE Trans. Magn., 55, 7, 1–6, 2019

Macrospin modelling of magnetoresistive angular sensors in high temperature environments

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Nowadays, several applications rely on angular positioners and encoders (e.g. robotics or automotive industries), where precision in the detection of the magnetic field orientation is necessary [1]. Tunnelling magnetoresistance (TMR) sensors are good candidates for angular sensors as they provide high output signals, high sensitivity and small footprint having an in-built directional reference that allows the field direction to be obtained [2]. However, when operating in high temperature regimes, this reference weakens and a clear reduction of performance is observed, with a final angular sensor response distancing from the ideal sinusoidal behaviour. In the high field regime, the magnetic anisotropy ratios also play a key role [3]. This work shows an analytical model for the magnetic behaviour of a full TMR stack including its dependence on temperature and applied field angle. The modelled system includes an exchange biased pinned layer (PL) and a synthetic antiferromagnetic (SAF) reference layer (RL) – fig 1A. The free Layer (FL)-RL coupling fields and all intrinsic magnetic anisotropies are included. Figure 1B shows the final magnetization and resistance dependence on the magnetic field at arbitrary angles for a standard buffer/MnIr/CoFe/Ru/CoFeB/MgO/CoFeB/cap stack. The calculations are coupled to temperature dependent models for exchange bias field (Hex), saturation magnetization (Msat), SAF coupling field (Hsaf) and magnetic anisotropy (Ku) [3,4]. Several stack properties (e.g. Ku, Hex, Hsaf) and its impact on the final device electrical output are explored. Input values (fig 1A) are obtained from experimental data. Figure 1C displays the temperature dependent stack M(H,T) loops. The RL plays a major role when operating at high fields that may compete with HSaf. With increasing temperature, the spin flop field decreases due to the loss of Hex and HSaf leading to a less stable reference (e.g. from ~1000 Oe at 22°C to 630 Oe at 200 °C). This competition induces deviations from the ideal behaviour [3] but is less observable when the ratio interlayer coupling/external field is high. The FL saturation field changes as function of temperature, towards smaller values as temperature increases. We observed that the FL intrinsic anisotropies induce second harmonic type deviations in a pure sinusoidal function. An optimal field value for operation is obtained when a balance between the free and reference system anisotropies is achieved; this needs to be considered when choosing the operation range. In $R(\theta)$ (not shown), negligible deviations from the sinusoidal behaviour (<2^e) can be achieved when operating at a saturation field that is 10 times higher than the FL anisotropy and 10 times lower than the SAF spin flop field. The presented model allows to evaluate each magnetic parameter contribution for the sensor performance aiming at identifying the source of deviations from the ideal response. This tool can be applied to design TMR sensor stacks in order to select the best combination for top performance. The optimal range of operation of the angular sensor can also be extracted based on material properties/interlayer couplings. This model will be applied to experimental M(H,T), R(θ) and R(T) for TMR microfabricated devices.



[1] Becker C et al., Sci Adv. 2019;5(12):eaay7459

[2] Freitas P et al., Proceedings of the IEEE. 2016;104(10):1894-1918

[3] Jaffrès H et al., Physical Review B. 2001;64(6)

[4] Lv H et al., J Magn Magn Mater. 2019;477:68-73

Influence of plastic deformation on the piezomagnetic behavior of a dual-phase steel

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The application of a mechanical stress to a demagnetized ferromagnetic material can modify the magnetic microstructure of the material without macroscopic magnetization [1]. When the material is initially magnetized, even weakly (which is the general rule in industrial parts), a stress can lead to a variation of the magnetization. This is the "Villari effect", or "piezomagnetic effect" [2][3]. The magnetization shows thus a high sensitivity to the prior metallurgical-mechanical state of a ferromagnetic material and to the applied mechanical loading, and its evolution will be different depending on the level of loading, its multiaxiality and its nature. The measurement of the variations of magnetization can be done via a secondary coiling (by measuring the induced voltage) or via external magnetic flux gates (so-called leakage method employed in previous studies [4]). The piezomagnetic response can thus be used for the purpose of passive monitoring of structures under their in-service cyclic loading. The principle is based on measuring Villari's signal evolution in use. Different authors have dealt with the study and the modeling of the effect of an elastic-plastic mechanical loading on the magnetic behavior of a ferromagnetic material. Several models have been developed in order to simulate the evolution of the magnetic response according to type of loading. The development of innovative experiments and innovative modeling methods is however required for identification of mechanisms, comprehension of principles and validation of modeling strategies.

In this study, we evaluate the influence of a homogeneous plastic strain on the piezomagnetic behavior of a dualphase steel in a magnetic field-controlled framework. Indeed, a plastic deformation is known to have a significant influence on the magnetic behavior of ferromagnetic materials [5]. Its influence on the piezomagnetic behavior is presented and discussed. A modeling is next proposed, as a combination of a coupled magneto-elastic multiscale model [6], a thermodynamically admissible irreversible stress expression and an evaluation of the second order residual stresses associated with plasticity by a two-phase description of the material [5].

[1] R. M. Bozorth, "Ferromagnetism", New York: Ed. Van Nostrand, 1951.

[2] E. Villari, "Change of magnetization by tension and by electric current", Ann. Phys. Chem, 126 (1865), 87-122.

[3] L. Lollioz, S. Pattofatto and O. Hubert, "Application of piezomagnetism for the measurement of stress during an impact", J. of Electrical Engineering, 57 8 (2006) 15-20.

[4] S. Bao, T. Erber, S.A. Guralnick, and W.L. Jin, "Fatigue, Magnetic and Mechanical Hysteresis", Strain 47(4): 372–381 (2011).
[5] O.Hubert., S.Lazreg, "Two phase modeling of the influence of plastic strain on the magnetic and magnetostrictive behaviors of ferromagnetic materials", J. of Magnetism and Magnetic Materials, 424 2 (2017) 421-442.

[6] O. Hubert, "Multiscale magneto-elastic modeling of magnetic materials including isotropic second order stress effect", J. of Magnetism and Magnetic Materials, 491, (2019), 1-16, 165564.

Frequency-modulated MEMS magnetometer using magnetic flux concentrator and permanent magnets

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Integrated magnetometers for large scale electronic products rely on Magneto Resistance technology due to their compact size, low cost and good performance. These sensors are usually coupled with inertial sensors (MEMS accelerometers and gyroscope) into inertial measurement unit (IMU) to provide motion information (inertial navigation, virtual reality, etc.). A MEMS magnetometer could enable a fully-MEMS IMU reducing chip size and cost and improving performance *e.g.* sensors alignment. For this reason, several example of MEMS magnetometers have been proposed [1]–[3], yet the leading technology remains the MR one. A new approach for a frequency modulated MEMS magnetometer is proposed. The novel device is inspired by the Magnetic Force Microscopy (MFM) technique where the resonance frequency of a magnetized cantilever is shifted by an external magnetic force gradient. In this work, the MFM concept is translated into a MEMS magnetometer compatible with industrial MEMS process. The device is composed of a resonator integrating permanent magnets coupled with magnetic flux concentrators (MFC) (figure 1). The MFC are used to shape the incoming magnetic field and generate a magnetic field gradient i.e. a force gradient acting on the permanently magnetized element of the resonator (figure 1). Due to the force gradient, a frequency shift of the device resonance frequency ($\Delta\omega$) is induced, according to the following formula:

$$\Delta\omega \propto \frac{\partial^2 B_x}{\partial x^2} \propto G_m B_0$$

where G_m is a controllable parameter which links the external field (B_0) with the second derivative of the field in the constriction. With respect to Lorentz force magnetometers, to first order, the scale factor does not depend on the current consumption, making the novel device a good candidate for a fully MEMS, low power, high performance IMU. At the same time, when compared to other resonant MEMS magnetometers[3], [4], the device shows a frequency shift which is proportional to the incoming field and not to its absolute value, a bias magnetic field is therefore unnecessary. The new device could take the advantage of the mature MEMS clocks technology and combines it with established magnetic elements such as the MFC to deliver a low power, high-performance magnetometer. Indeed, the expected power consumption and resolution of this sensors are in the order of few uW and few nT/rtHz respectively, with large margin for process optimization. In this work, the analytical and numerical modelling of the magnetometer is presented together with preliminary experimental results. This work is part of the European FET project OXINEMS.



Figure 1 - a) schematic representation of the device; b) schematic cross-section of the sensor; c) Analytical study of the second derivative produced by the MFC

G. Laghi, C. R. Marra, P. Minotti, A. Tocchio, G. Langfelder, Journal of Microelectromechanical Systems, 25, pag. 637, 2016.
 D. DiLella et al., Sensors and Actuators A: Physical, vol. 86, n. 1, pagg. 8–20, 2000.

[3] Y. Hui, T. Nan, N. X. Sun, e M. Rinaldi, Journal of Microelectromechanical Systems, vol. 24, n. 1, pagg. 134–143, 2015.

[4] S. Brugger e O. Paul, Journal of Microelectromechanical Systems, vol. 18, n. 6, pagg. 1432–1443, 2009

Proof-of-Concept of Mobile Platform with Tunable Magnetic Force

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Mobile platforms have been developing in various industrial fields for inspecting defects as a crack of buildings or structures which need safety inspection for maintenance due to aging or environmental influences. The structures which are required defect inspection could have constraint as stepped/cascaded or high location that workers cannot approach. Large numbers of the structure had been built up with ferromagnetic materials. This is the reason the most of mobile platform has magnetic attachment systems. The magnetic attachment systems make the mobile platform conduct reliable defect inspection by having strong magnetic force on the surface. The mobile platform carries numerous devices for performing germane investigation. It makes the weight of mobile platform heavy. Then magnetic attachment systems would be designed taking big or strong magnets for having enough force to cling on the magnetic wall with safety. However, moving on the magnetic surface with strong magnetic force could induce restriction of accessing the constraint area or applying a load to a drive motor. It can be overcome the limitation by applying a system or wheel that can adjust the magnetic force. In this paper, a mobile platform with applicable magnetic wheels is proposed. A magnetic force method applied on the applicable magnetic wheel can have additional magnetic forces to provide a system capable of variety of ferrous metal construction by using two magnets. The magnetic wheel includes two permanent magnets to change the magnetic flux between them by rotating one of the magnets. A magnet between of two-wheel rims let the wheel maintain attachment force from the surface, and the other magnet located on center of the housing which is connected with the wheel rims can control the magnetic force by being rotated. The magnetic wheel for stable movement in complex paths is applied, and the mobile platform is designed that facilitates attachment and detachment by increasing or reducing the adhesive force when necessary. The states are termed adhered-state and free-state, when the two poles of magnets are arranged with parallel or anti-parallel. And the magnet rotation causes the effective area of magnetic field among two magnets at each state. The magnetic flux density from the magnets would be the highest when the poles are exact parallel or anti-parallel, and the magnetic field would be effective by the flux density. Within parallel and anti-parallel by the angle change of the magnet, the magnetic flux density is reduced than those statuses. Using these conditions, the area affected by magnetic flux is decided as an effective area and analysis is executed. After the analysis, the types of permanent magnets are able to be chosen. The finite element simulation is performed to verify the adhesion force and magnetic flux change of the magnetic wheels by MAXWELL-3D. The mobile platform with the four applicable magnetic wheels is designed and fabricated by applying the applicable magnetic wheels which are decided by analysis and designed. In addition to the size considerations for application to various paths, the feasibility of the adhesion force is evaluated through finite element analysis.

Use of MFAM Technology in Diagnostics of Steel Wire Ropes

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Microelectromechanical systems (MEMS) have allowed to develop the magnetic field sensors with potential applications such as automotive industry, navigation systems telecommunications and non-destructive testing. Micro-Fabricated Atomic Magnetometer (MFAM) is a magnetometer system which can be used for land, airborne or even marine survey applications with the proper housing. The paper presents one of the first applications of MFAM sensors for diagnostics of steel wire ropes.

The work aims at, through a review of currently known magnetic sensors, to indicate the criteria of their selection in the assessment of the technical condition of steel wire ropes. Classification determining the magnetometers degree of sensitivity was presented. The list of advantages and disadvantages of each sensor allowed the development of characteristics enabling precise selection of measurement technology for the desired metrological parameters. In the case of steel wire ropes, these parameters can be classified into two groups: defects resulting from wires discontinuities, and weakness of the material due to stress appearing in the rope during its work. The most common stresses in ropes are tensile, bending and torsion, but almost always their interaction is more or less interrelated.

Differences of the magnetic field strength around the rope material are used to identify imperfections of steel wire ropes such as broken wires, deformations, wear and corrosion. The most common method of non-destructive testing of steel wire ropes is the active magnetic method, which uses imperfections leaked by a strong, formatted by an external source, magnetic field. Unfortunately, this method is ineffective in case of flaw detection of modern compacted ropes.

Research has been undertaken to analyze the possibilities of using of MFAM technology, as a result of local changes in the magnetic field strength around the rope material, for identification of steel wire rope technical condition. Under the influence of cyclically changing work loads, due to the effects of magnetomechanics, changes in electromagnetic properties occur both in the wires and in the entire rope.

Magnetoresistance and its relaxation of nanostructured la-sr-mn-co-o films: application for magnetic sensors

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It has been demonstrated that colossal magnetoresistance (CMR) phenomenon in nanostructured lanthanum manganite films can be successfully used for the development of CMR-B-scalar sensors, which are capable to measure the magnitude of pulsed magnetic fields in very small volumes [1,2]. Such sensors have been used at room temperatures to measure the distribution of magnetic fields in railguns and non-destructive pulsed-field magnets. However, for the scientific or industrial applications, sensors operating at wide range of temperatures and magnetic fields are required. It was found that B-site doping of lanthanum manganite $La_{1-x}Sr_xB_zO_3$ (B=Mn) with 3d ions (Co) would destroy the long-range ferromagnetic ordering of the Mn network, resulting in changes of magnetic and electrical properties of the material [3]. The substitution of Co for Mn in La-Sr-Mn-O films decreases the paramagnetic-ferromagnetic phase transition temperature and increases the resistivity of the films. Therefore, such doping could result in increase of the magnetoresistance MR. For nanostructured films consisting of high-quality crystallites separated by disordered grain boundaries, the origin of these effects is not enough understood, thus their investigations are of great importance. In this study, the magnetoresistance of column-shaped nanostructured La_{1-x}Sr_x(Mn_{1-v}Co_v)_zO₃ films deposited using the pulsed injection metal–organic chemical vapor deposition technique onto a polycrystalline Al₂O₃ substrate by changing Co content y (0; 0.05; 0.10; 0.12), while keeping the same ratio z=(Mn+Co)/(La+Sr)=1.17, was investigated in pulsed magnetic fields up to 25 T in the temperature range of (80-290) K. It was found that the Co substitution for Mn increases the low-field magnetoresistance (20% at 80 K at magnetic flux density 0.5 T) as well as high-field magnetoresistance values (68% at 80 K and 64% at 290 K at 25 T). It was demonstrated that the MR dependence on magnetic field for nanostructured films can be well described by modified Mott's hopping model taking into account contribution of both crystallites and grain boundaries. Resistance relaxation of the films after the action of 200 2s duration 10 T magnetic field pulse was investigated at 80 K. It was found that in the resistance dynamics one can distinguish relaxation processes occurring at different time scales: "ultrafast" (<1 μ s), which follows the magnetic field pulse; "fast" (~100 μ s) and slow (>1 ms) which were observed after the magnetic field was switched-off. The relaxation processes were analyzed using Kolmogorov–Avrami–Fatuzzo and Kohlrausch–Williams–Watts models. Influence of remnant resistivity and magnetic memory effects on the operation of B-scalar magnetic field sensors based on nanostructured manganitecobaltite films is discussed.

[1] T. Stankevic et al., Rev. Sci. Instrum., vol. 85, no. 4, Apr. 2014, Art. no. 044704.

[2] T. L. Haran, R. B. Hoffman, and S. E. Lane, *IEEE Trans. Plasma Sci.*, vol. 41, no. 5, pp. 1526–1532, 2013.

[3] X. G. Chen et al., J. Appl. Phys., vol. 116, no. 10, Sep. 2014, Art. no. 103907.

Symposium 4. Magnetization dynamics, spin waves and magnonics

Direct-write nano-architectures for 3D magnonics

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The increasing demand for integrated spin-wave microcircuits requires extension of magnonics to the third dimension [1]. In this regard, focused electron beam induced deposition (FEBID) is a technique of choice for direct-writing of 3D nano-architectures [2]. Though its capabilities for magnonics have recently been demonstrated in a few proof-of-concept experiments [3,4], it is not widely known in the magnonics community yet. At the same time, FEBID can offer unique features which go beyond the reach of traditional fabrication techniques employed in magnonics [5], as will be detailed in the talk.

As a foundational block of magnonic circuits, a direct-write phase-controlling element with broad tunability will be presented upon a Co-Fe-FEBID conduit containing a single nanodefect [2]. This device demonstrates, in particular, the required functionalities for spin-wave logic gates and magnonic lenses in spin-wave nano-optics. Next, a recently developed method of magnetic characterization of individual nanoelements will be introduced [3]. It allows for microwave inductive sensing of individual circular magnetic elements with radii down to 100 nm and the determination of their magnetization and exchange constant from the standing spin-wave resonances with high precision. The deduced magnetic parameters, in return, can be used in micromagnetic simulations for more complex-shaped nanomagnets such as "nanovolcanoes" (Fig. 1a) whose magneto-dynamic response not only contains signatures of its individual constituents (disk and ring) but exhibits further spin-wave resonances because of the mode hybridization. After that, bi-component magnonic crystals will be presented. They can readily be fabricated by FEBID alone or in combination with focused ion beam (FIB) milling, with a unique possibility to gradually modify the spin-wave transmission characteristics by stopping the deposition/milling process at the desired stage, and/or continuing it after a magneto-dynamic measurement. Finally, capabilities of FEBID for the fabrication of graded-index [6] magnonic conduits (Fig. 1(b)) will be demonstrated.



Figure 1 : Atomic force microscope images of a "nano-volcano" direct-write nano-architecture **a** and a magnonic waveguide with gradually decreasing thickness that induces a graded refractive index for spin waves via the internal field gradient **b**.

Research leading to these results was conducted jointly with Roland Sachser, Michael Huth, Sergey A. Bunyaev, Gleb N. Kakazei, Maciej Krawczyk, Konstantin Y. Guslienko and Andrii V. Chumak.

- [1] G. Gubbiotti, ed., Three-Dimensional Magnonics: Layered, Micro- and Nanostructures, Jenny Stanford Publishing, 2019
- [2] A. Fernandez-Pacheco et al., Materials 13, 3774 (2020)
- [3] O. V. Dobrovolskiy et al., ACS Appl. Mater. Interf. 11, 17654 (2019)
- [4] O. V. Dobrovolskiy et al., arXiv:2006.00763
- [5] A. V. Chumak et al., Nat. Phys. 11, 453 (2015)
- [6] C. S. Davies et al., Phys. Rev. B 92, 020408(R) (2015)

Hybrid magnetoelectric systems for spin-wave generation via magnetostriction

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Due to their scalability and their abundant nonlinear effects, spin waves (SW) are proposed as data carriers with ultra-low energies for future information processing systems [1,2]. Magnonic devices may complement CMOS circuits to reduce the overall computing power consumption, which today is a major limitation. However, the excitation, detection and control of spin waves is still a bottleneck since they are usually performed by electric current-based, inductive methods which show a low energy-efficiency due to Ohmic losses and a poor scaling behaviour. In contrast, voltage-driven magnetoelectric systems promise a much higher efficiency due to their capacitive operating principle [3,4]. In this talk, I present our recent results and perspectives of the magnetostrictive approach to realize nano-scaled magnetoelectric SW sources. Here, electric fields are coupled via inverse piezoelectric effects to mechanical degrees of freedom which, in turn are coupled to the magnetization via inverse magnetostriction.

In the first part, I will discuss the interaction of surface acoustic waves (SAW) with spin waves in ferromagnetic thin films. SAW are excited by RF voltages in interdigital transducer (IDT) on thin piezoelectric substrates and couple to spin waves in adjacent metallic microstructures (e.g. made of CoFeB). Using Brillouin light scattering microscopy (BLS), we show that the vertical confinement leads to a stationary interference pattern of coherent, co-propagating SAW (Fig 1a). Depending on the external magnetic field, the resulting magnetoelastic fields drive different SW modes in the ferromagnetic film (Fig 1b). A strong SW excitation by the SAW is demonstrated by the observation of first and second order SW instabilities.

The IDTs operate in a quite narrow frequency range on single crystalline substrates and are hard to miniaturize. In contrast, an ideal magnetoelectric transducer would combine broadband operation, scalability to nanometredimensions, and the usage of CMOS-compatible materials. Thus, in the second part, we use the developed experimental methods to investigate the excitation of propagating SW by acoustic excitations created in single magnetoelectric transducers based on piezoelectric elements with dimensions down to 500 nm which are mechanically coupled to SW waveguides made of Ni/NiFe bilayers (Fig 1c). BLS reveals the generation of both propagating phonons as well as SW with a large frequency bandwidth. Calculations of the strain profiles of the different modes indicate that shear stresses play a significant role in the excitation of the SW and that, in contrast to inductive antennas, higher order SW modes can be efficiently generated. Our results indicate that magnetoelectric systems are very promising as scalable and energy efficient sources for spin waves.



Figure 1 : a) Schematic of the BLS setup and SAW interference pattern. **b)** SAW and SW signal (magnons) in the ferromagnetic microstructure as a function of magnetic field. **c)** SEM cross section of the magnetoelectric transducer.

- [1]. A. Khitun and K.L. Wang, J. Appl. Phys., 110, 034306 (2011)
- [2] Q. Wang et al., Nat. Electronics, https://www.nature.com/articles/s41928-020-00485-6, (2020)
- [3] S. Manipatruni, D. E. Nikonov, I. A. Young, Nature Phys., 14, 338 (2018)
- [4] M. Weiler, L. Dreher, C. Heeg, et al., Phys. Rev. Lett., 106, 117601 (2011)

Oral Presentation

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Deterministic and time resolved thermo-magnetic switching in a nickel nanowire

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Thermally activated processes are ruled by Boltzmann statistics and they are stochastic by nature, occurring with a probability proportional to the Boltzmann exponential $\exp\{-E_A/k_BT\}$. In everyday life, increasing the temperature makes things happen. Thermally activated processes are often a blessing in our macroscopic world but they are certainly a curse in modern nanomagnetism. Temperature makes any process stochastic and, at the nanoscale, it makes magnetic experiments and devices unpredictable.

In this context, our work has the potential to make an impact in the field. We describe a thermally induced magnetic process in the nanoscale (a nucleation and subsequent propagation of magnetic domain walls in a Ni nanowire) that is completely deterministic [1]. A 700 nm section of an 80 nm diameter cylindrical nanowire is heated close (but below) to its Curie temperature. The local heat flow modifies the reversal mechanism from localized nucleation (critically dependent on disorder and stochastic by nature) towards almost coherent rotation of a well-defined volume in the heated area (hardly dependent on disorder and completely deterministic). The nucleation of the magnetic domain walls that triggers the switching can be achieved at a field considerably smaller than the nucleation field and, importantly, the exact moment of the magnetic switching area. Using micromagnetic simulations, we provide a very graphic example (and videos) of how this mechanism is sensitive to the nanosecond: heating during 51 ns does not induce nucleation but heating during 53 ns does. Combining micromagnetic simulations with a macrospin model, we show how this deterministic thermo-magnetic switching operates. Beyond the fundamental interest of describing a mechanism that leads to a deterministic thermal switching, we believe our results can help the future development of new concepts and magnetic nano-devices.

[1] M. P. Proenca et al., Scientific Reports 9, 17339 (2019)

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Domain wall dynamics in circular microwires under transversal magnetic field

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Domain wall dynamics in thin magnetic wire is used in modern spintronic devices to transfer and store information. One of the most crucial parameter governing the domain wall dynamics is a domain wall velocity, which is limited by the Walker limit. Cylindrical wire has advantage in hindering the Walker limit thus fast domain wall velocities can be observed.

Bistable glass-coated microwires are ideal material for domain wall dynamics study. They offer fast domain wall dynamics and also many other interesting effects for theory and application.

Within this contribution, we would like to refer on the directional effect of transversal magnetic field applied on cylindrical wire during the domain wall propagation. We show, that properly selected direction of transversal field can be used to manipulate the domain wall velocity by enhancing or hindering it. The effect is solved in terms of distribution of transversal anisotropy induced into the cylindrical wire during its production.

Oral Presentation

3545

Real-time Hall detection of spin-orbit torque-induced switching of GdFeCo dots

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We propose a new all-electrical technique for the real-time detection of the magnetization dynamics based on the measurement of the anomalous Hall resistance. Our approach allows for minimizing the non-magnetic background and revealing the dynamics with temporal resolution < 100 ps. The sensitivity is large enough to detect single-shot events and access their stochasticity. We applied the technique to study the magnetization switching induced by spin-orbit torques in ferrimagnetic GdFeCo, which is known for its fast dynamics [1,2].

Our investigation with nanosecond electric pulses reveals two important novel aspects of the ferrimagnet dynamics. First, contrary to expectations [3], the response of the magnetization to the torques is not immediate but is characterized by a finite waiting time. The time scale of this initial latency, which we ascribe to thermal activation [4], is comparable to the pulse duration. Consequently, despite the ensuing fast reversal typical of ferrimagnets [1,2], the waiting time represents a most significant bottleneck of the dynamics. Second, both the waiting and reversal times are characterized by large statistical distributions, influenced by the magnetic field assisting the switching and the pulse amplitude. This variability is smoothed and concealed by after-pulse switching-probability measurements and pump-probe measurements [5,6]. Our detection scheme unravels the stochastic switching dynamics of ferrimagnets and demonstrates a simple table-top solution for performing real-time Hall effect measurements with sub-nanosecond time resolution [7]. Beyond our specific investigation of spin-orbit torques, we envisage a broader range of applications, such as the detection of skyrmions and domain walls in racetrack devices and the dynamics of antiferromagnets.

- [1] Caretta, L. et al., Nat. Nanotechnol. 13, 1154 (2018)
- [2] Cai, K. et al., Nat. Electron.3, 37-42 (2020)
- [3] Garello, K. et al., Appl. Phys. Lett. 105, 212402 (2014)
- [4] Grimaldi, E. et al., Nat. Nanotechnol. 15, 111 (2020)
- [5] Decker, M. M. et al., Phys. Rev. Lett. 118, 257201 (2017)
- [6] Baumgartner, M. et al., Nat. Nanotechnol. 12, 980 (2017)
- [7] Sala, G. et al., Real-time Hall detection of current-induced dynamics, submitted for publication

Deeply Nonlinear Ferromagnetic Resonance in a YIG nanodisk

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The Landau-Lifshitz equation which governs the motion of magnetization in ferromagnetic bodies is highly nonlinear, yielding a series of interesting phenomena [1]. It is well-known that in ferromagnetic resonance (FMR) experiments, spin-wave (SW) instabilities quickly develop as the excitation power is increased, preventing to achieve large angles of coherent precession [2]. However, this limitation could be overcome in nanostructures, where the geometric confinement drastically reduces the density of normal modes and suppresses the nonlinear SW interactions present in bulk ferromagnets [3].

Here we show that the magnetization of a nanomagnet can be driven to unprecedented large steady state motion by a time-harmonic magnetic field. For this, we probe the out-of-plane magnetization dynamics of a low damping ($\alpha = 0.0005$) YIG nano-disc using a magnetic resonance force microscope [4]. Ultra-large amplitude precession with a nearly complete suppression of the longitudinal component of magnetization is achieved by pumping the sample with a strong uniform microwave field [5]. It is accompanied with an extremely hysteretic foldover of the resonance line. The measured foldover shift starts to deviate from the macrospin model only when the angle of the uniform precession increases above 30°. Sticking to a macrospin description of the magnetization dynamics, one can describe this behavior by introducing a nonlinear damping parameter [6]. Using micromagnetic simulations [7], we are able to quantitatively reproduce the experimental behavior over the full range of excitation amplitude, without any fitting parameter nor additional phenomenological nonlinearity. The simulations reveal a progressive spatial extension of the quasi-uniform mode, followed by the onset of instabilities of quantized non-uniform SW modes at large pumping power and turbulent dynamics.

[1] I. D. Mayergoyz, G. Bertotti C. Serpico, Nonlinear Magnetization Dynamics in Nanosystems (Elsevier, New York, 2009)

- [2] H. Suhl, J. Phys. Chem. Solids 1, 209 (1957)
- [3] G. Melkov et al. IEEE Magn. Lett. 4, 4000504 (2013)
- [4] C. Hahn et al. Appl. Phys. Lett. 104, 152410 (2014)

[5] Y. Li et al. Phys. Rev. X 9, 041036 (2019)

- [6] Y. S. Gui et al. Phys. Rev. B 80, 060402(R) (2009)
- [7] https://github.com/MicroMagnum/MicroMagnum

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Control of non-linear spin-wave generation at low magnetic bias field

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Recently it was shown that the prediction of the non-linear spin-wave excitation in the framework of Suhl instability processes is not adequate at low magnetic bias fields. In particular, it was shown by spatially averaged and time-resolved x-ray ferromagnetic resonance spectroscopy that in the low field regime non-linear spin waves are excited parametrically at 3/2 of the excitation frequency [1].

Here we demonstrate the 3/2 (Ω) non-linear spin-wave (NLSW) generation in Ni₈₀Fe₂₀ microstructures using a novel variant of scanning magneto-optical microscopy which we term super-Nyquist sampling microscopy (SNS-MOKE) [2]. This technique allows for phase-resolved imaging of the sample at multiple arbitrary frequencies. In this way we detect parametrically excited NLSWs at 3/2 of the excitation frequency in space and time directly. The corresponding wave vectors obtained from the 2D-FFT of the observed spatially resolved spin-wave pattern at 3/2 (Ω) above the threshold rf-field are in agreement with the theoretical predictions from Bauer et al. [1]. For this type of non-linearities we determine the threshold rf-field for different sample geometries and investigate the phase stability of the NLSW generation as a function of rf-field and bias field. By seeding the dynamics with an additional low power frequency component at 3/2 (Ω) we can achieve phase control of this novel type of spin-waves and increase the phase stability over time.

[1] H. G. Bauer et al., Nat. Commun. 6, 8274 (2015)
[2] R. Dreyer et al., arXiv:1803.04943 (2018)

Frequency multiplication effects in thin ferromagnetic layers detected by diamond nitrogen-vacancy center microscopy

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In thin ferromagnetic layers inhomogeneous magnetic properties can lead to frequency multiplication effects generating high harmonics of the rf driving field close to ferromagnetic resonance. Scanning time-resolved Kerr microscopy is employed to spatially resolve those high harmonics as well as parametric excitations [1]. Since the spatial frequency of the magnetic response increases at higher harmonics, the diffraction limited resolution of the microscope leads to an averaging of the Kerr response within the laser spot. Hence, a more local probing technique is required to resolve magnetization dynamics. Because of their extremely small size and strong response to magnetic fields, nitrogen-vacancy defect centers in diamond offer an ideal method to detect fields locally. The optical detection of magnetic resonance (ODMR) is a double-resonant technique to locally probe magnetic fields with the help of those defect centers [2]. It allows for the detection of up to the 25th harmonic of the rf excitation frequency generated by the precessing magnetic moments in a Permalloy film at low magnetic bias fields as well as parametric excitations at large driving amplitudes.

[1] R. Dreyer et al., ArXiv:1803.04943 (2018)

[2] C. S. Wolfe et al., ArXiv 1512.05418v2 (2016)

Inelastic spin-wave scattering by Bloch domain wall flexure oscillations

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We calculated the inelastic spin wave scattering by flexure vibrations of the Bloch domain wall (Winter's magnons) in thin magnetic films due to cubic nonlinearity in the magnetic subsystems [1]. The approach is based on the interaction of the propagating spin waves with the dynamical emergent electromagnetic field generated by the moving inhomogeneous magnetization texture (domain wall). The particular forms of the spin wave spectra of the magnetic film and domain wall (the equilibrium magnetization direction in the domains, magnetic anisotropy and dipolar interaction) result in specific angular dependences of the spin wave scattering probability. These angular dependences are different for the emission and absorption of the Winter's magnons by propagating spin waves. The probability of the spin wave scattering for the Winter's magnon emission and absorption processes essentially rises with the spin wave scattering angle increase up to 90 degrees. The angular dependence of the scattering probability is essentially stronger for the magnon absorption processes [1] that allow distinguishing these elementary emission/absorption processes experimentally.

The obtained results are quite general. They are also valid for the Neel domain walls in thin magnetic films with an uniaxial magnetic anisotropy. The scattered spin waves modulated by the domain wall oscillations in ferromagnetic materials can be registered with the methods of microwave or micro-BLS spectroscopy. We believe that the considered phenomenon can serve as an additional tool to study the spin wave and domain wall dynamical properties in thin magnetic films and other systems, for example, in magnetic stripes and multilayers with an uniaxial (transverse or longitudinal) magnetic anisotropy. Exploiting the flexure domain wall oscillations opens a new way to manipulate the high-frequency spin wave propagation on the nanoscale redirecting them by the large angles up to 90 degrees.

[1] N.N. Dadoenkova et al., pss- Rapid Res. Lett. 13, 1800589 (2019)

3696 **Slow-Wave Based Magnonic Diode**

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Spin waves, the collective excitations of the magnetic order parameter, and magnons, the associated quasiparticles, are envisioned as possible data carriers in future wave-based computing architectures. On the road towards spinwave computing, the development of a diode-like device capable of transmitting spin waves in only one direction, thus allowing controlled signal routing, is an essential step.

In most wave physics systems, dispersion relations are reciprocal, i.e., f(k)=f(-k). The magnetic systems have the particularity of breaking the time-inversion symmetry and could present, inversely, strong nonreciprocities [1]. It is the case of spin waves that propagate perpendicularly to the direction of the magnetization (Damon-Eshbach configuration) in ferromagnetic asymmetric thin films [2]. We will expose how we have exploited this nonreciprocity in the case of a ferromagnetic bilayer to fabricate a slow-wave based magnonic diode, where the spin-wave group velocity is reduced to very low values for one direction of propagation, and not for the other, thus producing unidirectional slow spin waves [3]. From the theoretical point of view, we will explain how this nonreciprocity arises from the chiral character of the dipolar interaction and how the exchange interaction plays a key role in its control. Propagating spin-wave spectroscopy was used to experimentally demonstrate the diode-like behavior of the bilayer, the composition of which was previously optimized through micromagnetic simulations. To perform this experiment, two antennas have been deposited on a CoFeB/Py bilayer thin film, as schematically shown in Fig. 1a. The dispersion relation (Fig. 1b) has been extracted from the change of mutual inductance associated with the spin wave propagation (symbols) and compared to MuMax3 micromagnetic simulations (colormap) and to Brillouin Light Scattering (BLS) measurements. It is possible to identify a plateau only for the waves traveling to the right (k>0). Consequently, at higher frequencies the wave propagation is possible only to the left (k<0), as expected from the dispersion relation.



Figure 1

[1] R. E. Camley, Surface Science Reports 7, 103 (1987)

[2] O. Gladii et al., Phys. Rev. B 93, 054430 (2016)

[3] M. Grassi et al., arXiv:1912.09735 (2019)

Bandgap engineering in nanometer-thick YIG-based magnonic crystals

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Magnonic crystals made of yttrium iron garnet (YIG) with ultralow damping offer promising prospects for spin-wave manipulation [1]. However, the frequency bandgap attained in YIG-based magnonic crystals has thus far been limited to a few tens of MHz. The realization of large tunable bandgaps in YIG-based magnonic crystals is essential for low-power magnonics.

Here, we report on one-dimensional magnonic crystals comprising 2 to 4 discrete nanometer-thick YIG stripes and experimentally demonstrate robust bandgaps up to 200 MHz for Damon-Eshbach-type spin waves [2]. Using broadband spin-wave spectroscopy and micromagnetic simulations, we show that the bandgaps are formed by strong Bragg reflection of spin waves with $k = n_p/a$. Spin wave transmission within the bandgap is almost completely suppressed. By changing external magnetic field, lattice constant, or stripe width, we establish strong tuning of the bandgap size from 50 MHz up to 200 MHz. Next, we compare results on discrete YIG stripes separated by airgaps and gaps filled by CoFeB. After the insertion of CoFeB, the transmission of spin waves in the allowed minibands is enhanced substantially while the size of the bandgap is reduced only slightly. We attribute low-loss spin-wave propagation in YIG/CoFeB magnonic crystals to enhanced dynamic dipolar coupling between the YIG stripes. This result illustrates that strong ferromagnets can act as mediator of magnetic interactions in YIG-based magnonic crystals. We also show that Bragg scattering on two airgaps or CoFeB stripes i.e., only 1 YIG stripe separated from a continuous YIG film, already produces clear frequency gaps in spin-wave transmission spectra. Finally, we show robust bandgaps with a size of 30 – 200 MHz in magnonic crystals provides effective spin-wave manipulation and low-loss propagation, a vital parameter combination for magnonic technologies.

[1] A.V. Chumak et al., Nat. Phys. 11, 453 (2015)

[2] H.J. Qin et al., Nat. Commun. 9, 5445 (2018)

Single shot acquisition of spatially resolved spin wave dispersion relations using x-ray microscopy

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Over the last two decades spin wave generation in various materials and geometries has been intensively studied revealing, for example, spin wave wavelengths in the nanometer regime or spin wave interference effects realizing magnonic logic devices [1, 2]. Experiments and theory have shown that magnonic structures exhibit highly anisotropic dispersion relations [1, 3]. Therefore, knowing the exact dispersion is a crucial factor in designing and characterizing nanoscaled devices and their prospective capabilities in magnonics applications.

We address the challenge of determining the full dispersion relation for a large range of frequencies and wavevectors combined with real space imaging. This is achieved by transitioning to time resolved scanning transmission x-ray microscopy (STXM) with high spatial (< 20 nm) and temporal (< 35 ps) resolution for detection of spin waves. Additionally, we introduce a modified Sinc function in time by an arbitrary waveform generator as excitation signal. This signal defines a variable rectangular shape in the frequency domain. Consequently, an arbitrary frequency range can be excited simultaneously with a uniform power distribution, revealing the full dispersion relation. Our subsequent evaluation procedure for time resolved STXM movies relies on fast Fourier transformation algorithms in space and time and results in frequency dependent maps of amplitude, phase, and *k*-space information. In doing so, we present an approach for measuring two dimensional spin wave dispersion relations *f*(*k*) within one single measurement and a measurement time below 40 minutes.

This is demonstrated using a sample with a coplanar waveguide on top of a 100 nm thin yttrium-iron-garnet film as illustrated in Fig. 1(a). The resulting dispersion relation is shown in Fig. 1(b) and the measurement includes phase and amplitude maps in real space. Furthermore, isofrequency curves as indicated in Fig. 2 can be directly measured which has not been achieved so far. In principle, time resolved STXM with modified Sinc excitations can detect and directly image $f(k_x, k_y)$, including all excited magnons, in all directions, and hence, reveal full isofrequency curves as schematically shown in Fig. 2. At the same time spin precession amplitudes and relative phases are extracted and non-reciprocity is evaluated. By combining signal theory and x-ray microscopy the investigation of spin wave excitation and propagation properties reveals outstanding possibilities for future magnonic ultra-thin structures.





Figure 1. | a Snapshot of a time resolved STXM movie revealing the mz component. The orange overlay illustrates ground (G) and signal (S) lines; b Experimental dispersion relation f(kx). The emerging mode branch perfectly fits the theoretical prediction (blue dotted line).

- [1] Dieterle, G. et al., Phys. Rev. Lett., 122, 117202 (2019)
- [2] Kithun, A. et al., J. Phys. D Appl. Phys., **43**, 26 (2010)
- [3] Lenk, B. et al., Phys. Rep., 507, 107 (2011)

Figure 2. | Three dimensional dispersion relation f(kx,ky) of the YIG sample. The white dashed lines indicate theoretical predictions. Additionally, isofrequency curves are shown.

Spin Wave Based Spectrum Analysis Implemented with FIB-Created Gratings in YIG

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In this work, we demonstrate the fabrication of concave gratings in YIG (yttrium iron garnet) by means of 50 keV Ga⁺ focused ion beam (FIB) irradiation. One of the promising applications of this technique is a spin-wave-based microwave spectrum analyzer proposed in [1]. This device requires a concave grating, which can act as a source of spin waves driven by a nearby microwave antenna. In the original design, the grating is expected to be accomplished by local etching (e.g. ion milling) of the spin wave medium (YIG). In contrast to this approach, we realize spin wave based spectrum analysis at full performance with a binary magnetization landscape achieved by FIB without the need of physically removing material.

In order to use YIG for magnonic computing applications, the energy landscape for spin waves has to be modified to influence their propagation characteristics, i.e. an alternation of the magnonic refractive index. Ion irradiation is a well known method to modify magnetic properties in various materials, and the effect of FIB on the magnetic properties in YIG has been investigated on film level in [2]. We deploy this technique with a significantly higher ion dose (1015ions/cm²) to locally switch off the magnetization through a strong increase of magnetic damping. This way, we are able to create binary magnetization patterns in YIG analogously to locally etching away material with nanoscale precision. The Ga⁺ irradiation effect has been studied via Transmission Electron Microscopy, revealing the formation of an amorphous top layer of about 25 nm in the 100 nm thick YIG film. Though the ion implantation depth is in good agreement with TRIM simulations, we suppose damage in the underlying YIG layer to some degree, since SW transmission is inhibited in the irradiated areas.

The measured spin wave propagation pattern of a fabricated spectrum analyzer is presented in Fig. 1, whereby the FIB irradiated pattern is highlighted in semi-transparent red. The diffraction of the excited spin waves (wavelength \approx 3 µm) is clearly visible and the diffraction angle (zero order and first order peaks) matches the expectations for this particular grating design. We observe a deviation of the diffraction peaks from the expected location (green vs. red circle) that is most likely a result of a tilted bias field or anisotropies in the system.



Figure 1: Measured spin wave interference pattern in the FIB created concave grating. Time-resolved MOKE microscopy has been used to image the spatial distribution and the resulting diffraction of propagating spin waves in forward volume configuration (isotropic propagation). A coplanar microwave antenna (gray stripe indicates the ground line) is placed behind the FIB grating (semi-transparent red pattern) for SW excitation.

[1] Á. Papp, W. Porod, Á. I. Csurgay, and G. Csaba, Scientific reports, 7, 1 (2017)

[2] W. Ruane, S. White, J. Brangham, K. Meng, D. Pelekhov, F. Yang, and P. Hammel, AIP Advances, 8, 056007 (2018)

Bose-Einstein Condensation of nonequilibrium magnons in laterally confined systems

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*The author has chosen not to make public additional content

Domain wall dynamics in bent magnetic cylindrical nanowires

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Magnetic cylindrical nanowires (NWs) hold important promises for future 3D information nanotechnologies [1]. Their use demands the proper description of magnetic domain wall (DW) dynamics which are strongly influenced by cylindrical geometry. Importantly, it has been predicted that DWs in straight cylindrical NWs do not suffer from the Walker breakdown phenomenon [2]. Here we performed a detailed micromagnetic analysis of the DW motion along a bent NW under the action of axial and tangential magnetic fields. We showed that the interplay between curvature and the external magnetic field plays a role in the DW velocity, precession frequency, and DW oscillation motion. At quite low applied fields, DW performs an oscillatory motion with large amplitude along the nanowire, together with the precession around its axis. The presence of bends in cylindrical NWs introduces back the Walker breakdown for the DW motion with the consequent decrease of DW velocity. A detailed analysis of the influence of curvature on the DW velocity is numerically performed, showing that the maximum DW average velocity can be controlled by external fields and decreases as a function of the curvature. The effect of the Walker breakdown is larger in the case of tangential fields in comparison to that of the axial one due to the similarity to the DW dynamics in magnetic stripes. The oscillatory dynamics of DWs in bent cylindrical NWs makes them a potential candidate to be used as nano-oscillator in prominent concepts regarding 3D nanotechnologies.

[1] A. Fernandez-Pacheco et al., Nature Comm. 8, 15756 (2017)

[2] M. Yan, A. Kakay, S. Gliga, and R. Herttel, Phys. Rev. Lett. 104, 057201 (2010)

Dynamical behaviour in Vortex-based Magnetic Tunnel Junctions for Emerging ICT applications

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Spin torque nano-oscillators (STNOs) are nanoscale tunable multifunctional radio-frequency devices which have been proposed for a diverse variety of applications, ranging from novel wireless communications paradigms [1], energy harvesting [2], random number generators [3] more recently the building blocks of novel bio-inspired computing architectures for neuromorphic computing [4]. The competitiveness of STNOs as an emerging technology lies in several key characteristics relative to conventional electronics, including tunability, multi-functionaility, scalability and radiation hardness. Vortex-based STNOs have been shown to demonstrate superior performance parameters in terms of output power emission and signal linewidth.

By harnessing the rich dynamics associated with non-homogeneous magnetisation configurations in confined nanostructures, the free layer of a magnetic tunnel junction (MTJ) can be forced, via a localised magnetic field, to transition between two magnetic states: the quasi-uniform and vortex states. In this report we demonstrate that such transitions can be driven back and forth to a dynamic equilibrium by exciting an MTJ with alternating magnetic fields close to the resonant modes of the magnetic vortex. The frequency dependence of the dynamic state transitions leads to a proposal for a novel type of broadband analogue frequency to resistance converter (FRC) with sub-MHz resolution [1] and a random number generator with a data rate of 38.7 Mbits/s [2].

- [1] A. S. Jenkins, et al., Phys. Rev. Appl., 13, 014046 (2020)
- [2] M. Tarequzzaman et al., Appl. Phys. Lett., 112, 252401 (2018)
- [3] A. S. Jenkins, et al Sci. Rep., 9, 15661 (2019)
- [4] H. Farkhani et al., Front. Neurosci., 13, (2020)

Spin wave propagation through domain wall

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Spin wave propagation through magnetic textures is recently subject of big interest. It has been shown that domain walls can serve as a spin wave filter [1], or as a phase shifter. It is possible to manipulate the domain wall position through spin torque generated from magnon a spin current [2]. However, in all conducted studies spin waves were propagated in either forward volume geometry or with an angle between propagation and effective field direction equal to 45 degrees. The decay lengths in both of these geometries are rather short, which would make designing of logic spin wave circuit challenging.

To overcome this drawback, we used a system of thin layer metastable iron, where the magnetic micro-structures can be imprinted by focused ion beam irradiation [3]. Depending on the irradiation parameters and focused ion beam scanning strategy different directions of uniaxial magnetic anisotropy within magnetic structures can be induced. By this technique, narrow waveguides with in-plane magnetic anisotropy pointing perpendicular to the waveguide long axis can be prepared and spin wave propagation in the Damon-Eshbach geometry (which results in longer propagation distances) can be achieved without the presence of external magnetic field [4].

Remanent two domain state of the waveguide was prepared first in the Kerr microscope. We set the distance between excitation microstrip antenna and domain wall to 3 µm. The sample was then transferred to the magnetic force microscope to probe the internal structure of the domain wall. We observed a cross tie domain wall, which is combination of in-plane magnetization and out-of-plane core. Spin wave propagation through the domain wall was studied by phase resolved Brillouin Light Scattering microscopy. Thermal spin wave spectra show that in the vicinity of the domain wall the effective magnetic field is increased. To observe the impact of the domain wall on the spin wave propagation we excited coherent spin waves by the microstrip antenna. Two regimes of propagation were observed. At frequencies around 7 GHz two spin wave beams are emitted from the sides of the domain wall and the propagation through the central part with the vortex core is suppressed. On the other hand, for the frequencies around 9 GHz, the spin waves are re-emitted only from the center. To get better insight into the process behind this behavior we performed micromagnetic simulations. Based on this we could connect these two propagation regimes to different regimes of the domain wall oscillation. In the lower frequency regime we excite the tails of the domain wall, thus two spin wave beams are emitted. In case of the higher frequency the core oscillates resonantly and emits spin waves.

[1] S.J. Hämäläinen, M. Madami, H. Qin, G. Gubbiotti, S. van Dijken, Nature Communications 9, 1 (2018)

[2] J. Han, P. Zhang, J.T. Hou, S.A. Siddiqui, L. Liu, Science 366, 1121 (2019)

[3] M. Urbánek, L. Flajšman, V. Křižáková, J. Gloss, M. Horký, M. Schmid, P. Varga, APL Materials 6, 060701 (2018)

[4] L. Flajšman, et al., Physical Review B 101, 014436 (2020)

Magnetization dynamics of Bi-doped YIG nanodisks

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One current goal of spintronics is the development of a sustainable information technology based on the transport of pure spin currents. One promising way to do this is to use spin waves (SWs), the elementary excitations of magnetically ordered materials: by collective precession of magnetic moments, they can transport angular momentum. SW information processing also offers interesting alternatives to conventional microelectronics [1]. For this, yttrium iron garnet (YIG), an insulating ferrimagnet, is the ideal material because it has the lowest magnetic damping. Furthermore, thanks to the spin transfer torque, it is possible to control the relaxation time of SWs in YIG by an electric current injected into an adjacent platinum layer [2]. However, such metal/insulator hybrid devices presently have certain limits, as a sudden drop in the amplitude of the main mode beyond a certain excitation threshold, which originates from non-linear coupling with other SW modes [3].

Different strategies can be considered to overcome these issues. In nanopatterned samples the available SW modes become quantized [4], which strongly affects the nonlinear regime. The perpendicular magnetic anisotropy can also be used to control the sign of the nonlinear frequency shift [5]. Recently, the growth of ultra-thin films of Bismuth doped YIG (BiYIG) with tunable perpendicular magnetic anisotropy has been achieved while preserving a high dynamic quality [6]. Here, we study the magnetization dynamics in individual nanodisks patterned from such a 30 nm thick BiYIG film, with diameters ranging from 1 μ m down to 200 nm. We employ a magnetic resonance force microscope (MRFM) [7] in which a cobalt nanosphere grown at the tip of a soft cantilever [8] dipolarly couples to the magnetic nanodisk to detect SWs.

To extract their magnetic parameters, we first measure the SW spectra of these BiYIG nanodisks in the linear regime. Similarly to the extended film, they all display a weak, still out-of-plane effective anisotropy ranging from 10 mT to 30 mT. Due to the geometrical confinement, the splitting between SW harmonics increases as the diameter decreases. However, only a weak dependence on the diameter of the main resonance field is observed, which contrasts with the expected behavior [4]. Also, the Gilbert damping of the largest disks, from 0.0008 to 0.001, is similar to the thin film, but it substantially increases as the diameter decreases: a threefold increase is witnessed for the smallest disks. These observations indicate that the BiYIG properties might be affected at the edge of the nanostructures during nanofabrication. Micromagnetic simulations and analytical calculations are in progress to quantitatively evaluate those effects. We will also discuss MRFM measurements performed at large microwave power on the same BiYIG nanodisks, which highlight the influence of the perpendicular anisotropy on the nonlinear regime of magnetization dynamics.

- [1] A. Chumak et al. Nat. Phys. 11, 453 (2015)
- [2] A. Hamadeh et al. Phys. Rev. Lett. 113, 197203 (2014)
- [3] M. Evelt et al. Appl. Phys. Lett. 108, 172406 (2016)
- [4] C. Hahn et al. Appl. Phys. Lett. 104, 152410 (2014)
- [5] B. Lührmann et al. J. Magn. Magn. Mater. 96, 237 (1991)
- [6] L. Soumah et al. Nat. Commun. 9, 3355 (2018)
- [7] O. Klein et al. Phys. Rev. B 78, 144410 (2008)
- [8] S. Sangiao et al. Beilstein J. Nanotechnol. 8, 2106 (2017)

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Oral Presentation

Zero-field propagation of spin waves in Damon-Eschbach mode in Permalloy

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Recently a strong interest powered by the need of going beyond-CMOS and being energy efficient has grown for the use of spin waves for data transfer and wave-based computation [1, 2]. Functional spin-wave devices allowing for control of the spin waves are essential for future spin-wave based information processing. However, the fabrication and the technical realization using conventional approaches relying only on planar magnonic structures is extremely difficult. To face this challenge we exploit the effect of the curvature of the system on the spin-wave properties by utilizing the topography of the magnetic material, hence inducing the effective magnetic interaction to the system [3].

Current innovations in nanofabrication technologies make available the fabrication of complex three-dimensional structures with properties unobtainable by classical planar approaches, e.g. electron beam lithography [4]. The possibility of inducing effective magnetic interaction by using the curvature of the system is one of them. The corrugation of the system is created by the use of focused electron beam induced deposition (FEBID). It is a direct-write approach that is utilized to create sinusoidal modulation of the substrate surface and therefore changes the surface morphology before the deposition of the magnetic material. The magnetic material grown on such a modified substrate shows a dominant induced effective uniaxial anisotropy.

For the study of the magnetization reversal processes and related quantification of the induced uniaxial magnetic anisotropy, a Kerr magnetometry with an external magnetic field applied in the film plane was employed. To exclude any shape-related effects, magnetization reversal processes were studied on the magnetic discs with a diameter of 7 μ m. From the shape of magnetization loops, the forming of the easy and hard axis can be observed depending on the direction of the external magnetic field with respect to the ripples. When the external magnetic field was perpendicular (parallel) to the ripples we speak about the hard (easy) axis, respectively. Also, depending on the amplitude of modulation, the strength of the induced uniaxial anisotropy changes.

For spin-wave propagation studies in corrugated structures, we put the excitation antenna on top of a magnonic waveguide and measure using micro-focused Brillouin light scattering (μ -BLS). By optimizing the curvature in the structure it is possible to achieve a minimal influence of the amplitude of modulation on spin-wave decay. Also, the induced magnetic anisotropy is strong enough to overcome the shape anisotropy of the waveguide leading to the propagation of spin waves in the Damon-Eschbach geometry without the use of the external magnetic field.

[1] A. V. Chumak, V. I. Vasyuchka, A, A, Serga B. Hillebrands. Magnon spintronics. Nat. Phys. 11, 453 (2015)

- [2] A. Haldar, D. Kumar A. O. Adeyeye. Nat. Nanotechnol. 11, 437 (2016)
- [3] K. Chen, R. Frömter, S. Rössler, N. Mikuszeit H. P. Oepen. Phys. Rev. B, 86, 064432 (2012)
- [4] M. Huth, F. Porrati, O. V. Dobrovolskiy, Microelectron. Eng. 185, 9 (2018)

Direct Observation of Magnon Modes in Kagome Artificial Spin Ice with Topological Defects

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Kagome artificial spin ice (KASI) is a network of Ising type nanobars on a kagome lattice [1,2]. Great progress has been made in understanding defects including disordered states in KASI via quasistatic imaging techniques [1,3]. However, the magnetodynamic study of disordered states has been confined to global magnetodynamic (e.g. broadband spin wave spectroscopy) and micromagnetic simulations [4]. Furthermore, the simulation study on KASI has shown the presence of novel magnetodynmic microstates that offer excellent insights into the KASI's disordered regime [4]. From the fundamental physics side dynamically controlled microstates may offer a way to create Dirac strings via microwave assisted switching interior to the KASI lattice in a controlled manner and study the disordered regime systematically. From the magnonic application perspective experimental studies of microstates in KASIs are key towards their usage in as a new type of microwave filter [5] and reprogrammable magnonic crystal [6].

We investigate spin dynamics of a KASI consisting of Ni₈₁Fe₁₉ nanomagnets arranged on an interconnected kagome lattice using broadband all electrical spin wave spectroscopy (AESWS) (Fig. a), magnetic force microscopy (MFM) (Fig. b), and micro-focus Brillouin light scattering (BLS) microscopy (Fig. e-f). Micro-focus BLS performed on magnetically disordered states exhibit a series of magnon resonances which depend on topological defect configurations that we image by magnetic force microscopy. Nanomagnets on a Dirac string and between a monopole-antimonopole pair show pronounced modifications in magnon frequencies both in experiments and simulations. Our work is key for the creation and annihilation of Dirac strings via microwave assisted switching and reprogrammable magnonics based on ASIs.



Figure 1: Gray scale AESWS spectra map of KASI for a field protocol of **a** -90 mT \rightarrow +90 mT and **b** -90 mT \rightarrow 45 mT \rightarrow 0 mT. To enhance the contrast, we show the difference between neighboring spectra (derivative). White arrows represent sweep direction of H. The additional colored dots and dotted arrows highlight positions explored by micro-BLS; c Magnetic force microscopy images of KASI after a field protocol of -100 mT \rightarrow +45 mT \rightarrow 0 mT. Stray fields detected by MFM are colored in blue and red consistent with charges Q = +q and Q = -q, respectively; **d** Sketches of charge configurations (blue and red spheres) and magnetization vectors M (arrows) for configurations C1 to C5; e BLS intensities measured at the central position on horizontal nanobars belonging to the configurations displayed above the spectra at $\mu_0 H = 25$ mT; **f** Spatially resolved BLS intensity maps at 25 mT for fixed frequencies in configurations C1 to C5 displayed above. Here red (blue) color corresponds to maximum (minimum) BLS intensity.

- [1] S. Ladak, et al., Nature Physics 6, 359 (2010)
- [2]R. Wang, et al., Nature **439**, 303 (2006)
- [3]E. Mengotti, et al., Nat. Phys. 7, 68 (2011)
- [4] V. Bhat, et al., Physical Review B 93, 140401 (2016)
- [5] X. Zhou, et al., Advanced Functional Materials 26, 1437 (2016)
- [6] M. Krawczyk and D. Grundler, J. Phys.: Condens. Matter 26, 123202 (2014)

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Spin wave propagation in orthogonal MTJ stacks

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Spintronic devices are promising candidates to complement CMOS technology with improved throughput per energy consumption. MRAM technology based on magnetic tunnel junctions (MTJs) has already demonstrated its potential and compatibility with CMOS. Based on such memory technology, interconnected MTJs via their free layer have been proposed for logic application using domain walls [1], but they have not yet been explored for spin wave applications.

In this work, we report on the magnetization dynamics in the free layer (FL) of orthogonal MTJ stacks formed by an in-plane magnetized CoFeB FL separated by an MgO tunnel barrier from a CoFeB layer with perpendicular magnetic anisotropy (PMA). The PMA CoFeB layer serves as reference layer (RL) for the MTJ structure and coupled to Co/Pt synthetic antiferromagnet (SAF) with PMA (see Fig. 1a for schematic of the structure). The MTJ stacks were patterned into structures ranging from single (μ m-wide) to multiple waveguides (down to 250 nm width). Three FL thicknesses have been considered: 2.5, 5, and 10 nm.

The spin waves were excited by the Oersted field produced by RF currents flowing through single wire inductive antennas. A second antenna was used for detection and the magnetic structure was electrically isolated from the antennas by 60 nm of SiO₂. Different propagating distances were studied in order to assess the decay length of the spin waves. Using all-electrical spin-wave spectroscopy [2], we have investigated the propagation characteristics of spin waves in the different MTJ structures in the Damon-Eschbach geometry. Using this approach, spin waves propagation could be efficiently measured even in 2.5-nm-thick FLs. The shift of the FMR frequency, the changes in the spin wave band, and the decay characteristics were studied in devices with different waveguide width and propagation distance (see Fig. 1). The results can be explained by modifications of the internal effective field, coupling between adjacent layers, and the spin wave dispersion relations. The results are crucial for the understanding of propagating spin wave modes in ultra-scaled structures and open paths for magnetic interconnects between MTJs. Moreover, they may allow for the exploration of spin-wave-based logic devices using the shared FL between different MTJs.



Figure 1 : a Sketch of two antennas experiment for spin wave propagation showing the whole orthogonal MTJ stack under investigation composed by a CoFeB FL of different thicknesses, CoFeB RL separated by MgO and a PMA CoPt SAF; **b** Field derivative of the transmitted power for a 20 μ m wide waveguide and 2.5 nm FL after 2 μ m propagation and **c** spin wave transmission at 100 mT in a 10 nm FL for different waveguide widths: single 5 μ m line, and parallel lines of 2.5 μ m (2) and 250 nm (20).

The funding from the European Union's Horizon 2020 research and innovation program within the FET-OPEN project CHIRON under grant agreement No. 801055 is acknowledged.

- [1] E. Raymenants et al, 2018 IEEE International Electron Devices Meeting (IEDM), 36.4, 1 (2018)
- [2] F. Ciubotaru et al., APL 109, 012403 (2016)

Oral Presentation

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Of the importance of the substrate properties in the spin pumping in Co/Pt bilayers

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Spintronics research concentrated its efforts on handling spin polarization current leading to the success-story of giant magnetic resistance spin valves [1-2]. More recently, pure spin currents, i.e. a spin flux without charge flux, are at the centre of interest since they do not produce Joule heating, reducing power dissipation in spintronics devices, as in STT-MRAM [3]. These applications require an increase in the emission and detection efficiencies via a full understanding of the complex physics behind pure spin currents. Spin current can be detected by inverse spin Hall effect (ISHE) that corresponds to the conversion of a spin current into a detectable charge current by measuring the voltage on a normal metal presenting a strong spin-orbit interaction. Pure spin current emission can be obtained by temperature gradients, by charge currents and by magnetization dynamics. This last mechanism permits to generate pure spin current via ferromagnetic resonance in a ferromagnetic material [4].

We will present acoustic spin pumping experiments in Co/Pt bilayers, using surface acoustic waves (SAW) to induce the magnetization precession in the Co layer taking advantage of resonant magnetoelastic coupling.

We will report a peculiar behaviour of SAW-FMR assisted spin pumping in a multiferroic system where Co/Pt bilayer is in contact with a ferroelectric substrate LiNbO₃. We will show that the substrate on which the bilayer is deposited plays a role in the spin current excitation processes. We will show that the electrical polarization vector affects the ISHE voltage: the inversion of the magnetization and of the SAW propagation vector modifies the spin pumping efficiency. We suggest that this polarization is an important factor in the spin pumping excitation processes. The nature of the coupling with the magnetic polarization may be due to a magnetoelectric coupling at the interface between LiNbO₃ and Co, inducing non equilibrium spin density across the whole thin film [5] and affecting spin pumping efficiency.

[1] M. N. Baibich, et al., Phys. Rev. Lett., 61, 2472 (1988)

- [2] J. M. George, et a., Phys. Rev. Lett., 72, 408 (1994)
- [3] S. Maekawa, S. O. Valenzuela, E. Saitoh, and T. Kimura, Spin Current (Oxford University Press, 2017) pp. 1-520
- [4] M. Weiler, H. Huebl, F. S. Goerg, F. D. Czeschka, R. Gross, and S. T. B. Goennenwein, Phys. Rev. Lett., 108, 176601 (2012)
- [5] C.-L. Jia, T.-L. Wei, C.-J. Jiang, D.-S. Xue, A. Sukhov, and J. Berakdar, Phys. Rev. B, 90, 054423 (2014)

Enhanced Gilbert damping parameter in Co thin films in contact with the epitaxial and large-area Sb₂Te₃ topological insulator

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Spin-based electronic devices constitute an intriguing area in the development of future nanoelectronics [1,2]. Recently, 3D topological insulators (TI), when in contact with ferromagnets (FM), play a central role in the context of enhancing the spin-to-charge conversion efficiency [1]. Here, we report on the chemical, structural, and magnetic properties of evaporated polycrystalline Co thin films, with thickness between 2 and 15 nm, grown on top of a 30 nm thick Sb₂Te₃ TI grown by Metal-Organic Chemical Vapor Deposition (MOCVD). The Co layers are capped in situ with~4 nm of thermally evaporated Au. The MOCVD-grown Sb₂Te₃ layers here employed are characterized by a nearly epitaxial quality, as recently communicated by some of us [3]. X-ray reflectivity also revealed a chemicallysharp Co/Sb₂Te₃ interface, as we previously observed in Co layers deposited by atomic layer deposition on top of granular Sb₂Te₃ [4]. All-electrical broadband ferromagnetic resonance spectroscopy is employed to investigate the magnetization dynamics of the developed Au/Co/Sb₂Te₃ heterostructures. The results are compared to those obtained in reference Au/Co/SiO₂ and Au/Co/Pt samples, thus allowing to extract the role played by the 3D-TI Sb₂Te₃ when compared to Pt and SiO₂. Studies on the presented systems focused on the determination and analysis of magneto-structural inhomogeneities (DH₀), magnetic anisotropies (H_k) and the damping parameter (α). In Figure 1 we show the resonant frequency of the Au/Co (X nm)/Sb₂Te₃ heterostructure series of samples (X in the range of 2-15 nm) as a function of the magnetic field. The main result is that, when Co is grown in contact with Sb₂Te₃, a significant enhancement of the α value is measured respect to the reference Pt and SiO₂-based systems. The enhanced α value can be attributed to an increased spin-pumping effect taking place at the Co/Sb₂Te₃ interface, possibly due to the topologically protected surface states of the 3D-TI Sb₂Te₃. The high structural, morphological and magnetic quality achieved in the Au/Co/Sb₂Te₃ heterostructure is very promising for the optimization of efficient spin-to-charge conversion processes across the Co/Sb₂Te₃ interface.

- [1] Y. Wang, R. Ramaswamy, and H. Yang, J. Phys. D: Appl. Phys., 51, 273002 (2018)
- [2] F. Hellman et al., Reviews of Modern Physics, 89 2 (2017)
- [3] M. Rimoldi et al., RSC Adv., 10, 19936 (2020)
- [4] E. Longo et al., Journal of Magnetism and Magnetic Materials, 509, 166885 (2020)

Optically Inspired Nanomagnonics with Patterned Spin Textures in Synthetic Antiferromagnets

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Spin waves represent a promising avenue for implementing unconventional wave-based computing platforms. However, combining controlled generation, manipulation and long-distance propagation of submicrometric wavelength spin waves is an outstanding challenge.

At the same time, spin textures in magnetic materials, such as magnetic domains, domain walls or vortices, are gaining attention for their potential as active components in data storage and processing devices. Recently, the possibility of using spin textures for controlling spin waves have become extremely appealing, due to their strong influence on a wide range of different aspects, ranging from the generation, propagation, to the spatial confinement of spin waves down to the nanoscale.

In this framework, we demonstrated that nanopatterning spin textures via thermally assisted magnetic scanning probe lithography (tam-SPL) [1] allows the stabilization, in exchange bias systems, of 2D domains with arbitrary shape and spin configuration, 1D domain walls and 0D magnetic solitons such as vortices with tailored topology and position [2].

Here, first we show the channeling and steering of spin waves in arbitrarily shaped nanomagnonic waveguides based on straight and curved domain walls, and a prototypic nanomagnonic circuit comprising two converging waveguides, allowing for the tunable spatial superposition and interference of confined spin-wave modes [3]. Then, we present an optically inspired platform for controlling the generation, propagation, and interference of short-wavelength spin waves, using nanopatterned spin textures in synthetic antiferromagnets (SAF), at remanence [4]. By coupling radiofrequency magnetic fields with engineered magnonic nanoantennas consisting of patterned domain walls and vortices, we generate and shape spropagating pin waves.

We demonstrate the spatial engineering of spin-wave wavefronts, the directional emission and focusing of spinwave beams, and the generation of robust interference patterns, which span multiple times the wavelength. Furthermore, we show that SAF allows to combine concepts borrowed from optics, with phenomena naturally arising from the nonreciprocal spin wave dispersion, such as resilience to spurious back reflections. The ability to control magnons via nanopatterned spin textures at remanence opens up a range of new possibilities for developing energy-efficient unconventional computing concepts.

- [1] E. Albisetti, D. Petti, M. Pancaldi et al., Nat. Nanotechnol., 11, 545 (2016)
- [2] E. Albisetti, A. Calò, M. Spieser et al., Appl. Phys. Lett. 113, 162401 (2018)
- [3] E. Albisetti, D. Petti, G. Sala et al., Commun. Phys. 1, 56 (2018)
- [4] E. Albisetti, S. Tacchi, R. Silvani et al., Adv. Mater. 32, 1906439 (2020)

Generation and tuning of a spin wave frequency comb

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Optical frequency combs are powerful tools used for synchronization, stabilization and frequency conversion in both fundamental science and technical applications.

In this work, we present experimental observations on the generation of a spin wave frequency comb in a low damping Co₂₅Fe₇₅ conduit measured using Brillouin light scattering microscopy. By driving the magnetization to large precession angles, nonlinear interactions such as four magnon scattering can be observed. When applying two RF signals with independently tunable frequencies and amplitudes to our microstructure, we can actively control the final states that will be populated by these scattering processes. Our results show the generation of a frequency comb consisting of several spin waves with adjustable frequency spacing and amplitude.

We demonstrate that the known effect of frequency mixing for k=0 modes can be extended towards propagating spin waves. This enables simultaneous information transport and processing. This behaviour is studied for different sample geometries which allow mixing of co-propagating as well as counter-propagating spin waves. Our observations are in qualitative agreement with micromagnetic simulations.

The presented data encourage a deeper understanding of the interaction of propagating spin waves in the nonlinear regime and propose utilization of spin wave frequency combs as broadband tunable clocks for information processing.

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Observation of spin-motive force in ferrimagnetic GdFeCo alloy films

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Non-uniform magnetic structures produce emergent electromagnetic phenomena such as the topological Hall effect and the spin-motive force (SMF). The experimental reports on the SMF, however, are very few and the relationship between the SMF and material parameters is still unclear. Furthermore, the detected SMFs are almost 0.1 - 1 mV and we have no guidelines for the material development to enhance the SMF. In this study, we investigated the SMF in GdFeCo alloy films using the spin-torque-induced ferromagnetic resonance method and clarified the relationship. The GdFeCo alloy is a ferrimagnetic material, where the magnetic moment of FeCo is antiparallel to that of Gd. The saturation magnetization, the magnetic anisotropy, and the damping constant in the GdFeCo alloys strongly depend on the Gd composition and the composition of the alloy can be changed by the cosputtering. Therefore, GdFeCo is one of the suitable materials in studies on the relationship between the SMF and some material parameters such as the saturation magnetization, the demagnetizing field, the composition, the magnetic anisotropy, and the damping constant.

The Si₃N₄(10 nm)/Gd_x(Fe₈₂Co₂₈)_{1-x}(16 nm)/Pt(10 nm) strips were fabricated by a magnetron sputtering, electronbeam lithography, and a lift-off process. The widths of the strips are changed along a longitudinal direction of the strips and the structures are not rectangular, but trapezoidal. When the current is injected into the trapezoidal strip, the spin torque and the rf Ampère field are applied to the GdFeCo layer and the magnetic resonance is excited. In this case, since the spin torque and the rf Ampère field is inversely proportional to the width of the strip, the non-uniform magnetic dynamics are excited by the rf current and it induces the dc SMF voltage along the longitudinal direction of the strip. The dc voltage was detected by using the lock-in amplifier and the amplitude modulation method.

We investigated the Gd composition dependence of the SMF. The amplitude of the detected SMF becomes larger than that of the transition metal alloy FeCo by the Gd doping and reaches the maximum near a Gd composition of the boundary between in-plane and perpendicularly magnetized films. According to the analytical calculation, the enhancement is related to the trajectory of the magnetization precession. Moreover, we find that the SMF is inversely proportional to the square of the damping constant. Therefore, we should take into account the trajectory of the magnetization precession and select ferromagnetic materials with lower damping constants to enhance the SMF.

Spin Transport across epitaxially grown magnetically ordered SrMnO3 ultrathin films

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Spin transport across insulating antiferromagnetic materials have gained significant attention in recent years due to their fast magnetization dynamics and absence of stray field effects thus enhancing prospects for potential applications. Exploiting the (inverse) Spin Hall Effect, various spin-based phenomena have been studied via the Spin Hall Magnetoresistance (SMR) and Spin Seebeck effect. The highly correlated orbital, charge and spin degrees of freedom in manganite based oxide materials offer high tunability of magnetic properties through strain, temperature and magnetic field application. The versatility of magnetic interactions in SrMnO₃ due to its codependence with structural properties increases the broad range of possibilities that can be studied using local and non-local spin transport in SrMnO₃. By varying the strain and Pulsed Laser Deposition growth parameters such as oxygen pressure, SrMnO₃ films are tuned to show variable bulk magnetization features indicating different magnetic ordering. Both uncapped and capped SrMnO₃ thin films have been studied in this work. The surface magnetic ordering of insulating SrMnO₃ films, grown under different conditions, is characterized using SMR as a probe, allowing for a distinction between antiferromagnetic and ferromagnetic ordering. Additionally, by studying the second harmonic response using Pt Hall-Bar structures, we realize spin current propagation in this magnetic insulator by detecting Spin Seebeck response in Pt. Besides these local techniques, magnon spin transport using non-local approach is investigated for either electrical and thermal injection. A comprehensive overview of bulk and interface characterization establishes SrMnO₃ thin films as an interesting material choice in spintronics based condensed matter research.

A micromagnetic study of the spin waves eigenmodes of magnetic wires and dots in presence of Dzyaloshinskii-Moriya interaction

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Comparison of the magnon diffusion length for electrically and thermally excited magnons in 2-μm-thick Y₃Fe₅O₁₂

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Recent demonstration of efficient transport and manipulation of spin information by magnon currents have opened exciting prospects for processing information in devices. Magnon currents can be excited both electrically and thermally, even in magnetic insulators, by applying charge currents in an adjacent metal layer [1]. Earlier reports in thin yttrium iron garnet (YIG) films suggested that the diffusion length of magnons is independent of the excitation method [2,3], but different values were obtained in thicker films [4-7]. Here, we study the magnon diffusion length for electrically and thermally excited magnons in a 2-µm-thick yttrium iron garnet (YIG) film as a function of temperature and magnetic field [8]. Our results evidence that the diffusion length depends on the generation mechanism, suggesting that magnons of different energies are excited -sub-thermal and thermal magnons for electrically and thermally driven magnon currents, respectively- and, consequently, indicating that the magnon diffusion length the magnetic field is weaker than for those excited electrically. Finally, we demonstrate that the magnon diffusion length for thermally excited magnons is independent of the YIG thickness and material growth conditions, confirming that this quantity is an intrinsic parameter of YIG.

- [1] Y. Kajiwara et al., Nature 464, 08876 (2010)
- [2] L. J. Cornelissen et al., Nat. Phys. 11, 1022 (2015)
- [3] L. J. Cornelissen et al., Phys. Rev. B 94, 180402(R) (2016)
- [4] S.T.B. Goennenwein et al., Appl. Phys. Lett. 107, 172405 (2015)
- [5] B. L. Giles et al., Phys. Rev. B 92, 224415 (2015)
- [6] B. L. Giles et al., Phys. Rev. B 96, 180412(R) (2017)
- [7] J. Shan et al., Phys. Rev. B 94, 174437 (2016)
- [8] J. M. Gomez-Perez et al., arXiv:1912:00490 (2019)

Spin-wave dispersion of magnetic materials measured by the propagating spin-wave spectroscopy technique using variable gap antennas

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Propagating spin-wave spectroscopy (PSWS) is a technique often used for the characterization of spin-wave propagation in magnetic layers and structures (e.g. [1–3]). In our work, we show how this approach can be used in order to reconstruct the dispersion relation of a studied material with a high number of experimental points. We use a vector network analyzer (VNA) that is contacted onto a pair of lithographically fabricated stripline antennas on the sample through microwave probes. The stripline antennas are separated by a defined distance. The spin-waves are propagating across this distance (gap width) and their characteristics are measured in the form of the transmission parameters S21 and S12 by the VNA. Using an array of antennas with variable gap width between them, we measure the change of the spin-wave phase as a function of the gap width. From this data, we can directly calculate the k vector of the spin-wave for the given frequency. By measuring over a range of frequencies we can reconstruct the dispersion relation. The maximum k vector is limited by the geometry of the excitation antenna. For example, by using a 500 nm wide stripline and the gap width ranging from 1 um to 4 um we were able to reconstruct the spin-wave dispersion of a Permalloy thin film in for k vectors up to 7 rad/um with approx. 1000 data points.

This approach is versatile with regard to the measured materials. The samples can be either full layers (e.g. evaporated, sputtered or epitaxial) or the material can be patterned as well forming magnetic waveguides with the only limiting factor being the possibility of the excitation antenna fabrication. It can also be used for systems with complex dispersion relation e.g. for magnonic crystals [4]. This was previously studied only by the means of magnitude measurements by showing a drop in VNA transmission (no dispersion was measured) [5] or by measuring k-resolved Brillouin light scattering where the time limits the number of measured points [6–8]. The approach using VNA is simpler and quicker than BLS while it would offer and extra valuable insight (compared to the magnitude observation only) into the studied crystals by measuring the dispersion relation showing the bandgaps directly.

- [1] K. Yamanoi, S. Yakata, T. Kimura, and T. Manago, Jpn. J. Appl. Phys. 52, 083001 (2013)
- [2] F. Ciubotaru, T. Devolder, M. Manfrini, C. Adelmann, I. P. Radu, Appl. Phys. Lett. 109, 012403 (2016)
- [3] H. Qin, S. J. Hämäläinen, K. Arjas, J. Witteveen, S. van Dijken, Phys. Rev. B 98, 224422 (2018)
- [4] A. V Chumak, A. A. Serga, B. Hillebrands, J. Phys. D. Appl. Phys. 50, 244001 (2017)
- [5] A. V. Chumak, A. A. Serga, B. Hillebrands, M. P. Kostylev, Appl. Phys. Lett. 93, 022508 (2008)
- [6] A. V. Chumak, et al., Appl. Phys. Lett. 95, 262508 (2009)
- [7] G. Gubbiotti, et al., J. Phys. D Appl. Phys. J. Phys. D Appl. Phys 43, 13 (2010)
- [8] B. Obry, et al., Appl. Phys. Lett. 102, 202403 (2013)

Spin Wave Radiation by a Topological Charge Dipole

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The use of spin waves (SWs) as data carriers in spintronic and magnonic logic devices offers operation at low power consumption, free of Joule heating. Nevertheless, the controlled emission and propagation of SWs in magnetic materials remains a significant challenge. Here, we propose that skyrmion-antiskyrmion bilayers form topological charge dipoles and act as efficient sub-100 nm SW emitters when excited by in-plane ac magnetic fields. The propagating SWs have a preferred radiation direction, with clear dipole signatures in their radiation pattern, suggesting that the bilayer forms a SW antenna. Bilayers with the same topological charge radiate SWs with spiral and anti-spiral spatial profiles, enlarging the class of SW patterns. We demonstrate that the characteristics of the emitted SWs are linked to the topology of the source, allowing for full control of the SW features, including their amplitude, preferred direction of propagation, and wavelength.

[1] S. A. Díaz, T. Hirosawa, D. Loss, and C. Psaroudaki, arXiv:2002.12282

Micromagnetic study of frustrations and high-frequency excitations in artificial buckyball nanostructures

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Spin transport through insulating multilayers

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Spin transport in magnetic insulators allows for transport without Joule heating. Furthermore, many magnetic insulators are oxides with exceptionally low damping, giving rise to energy efficient transport of angular momentum for future spin-wave based technology.

The design of spin-transport based devices such as transistors or spin valves will require multilayer systems composed of different magnetic materials [1]. Thus, understanding the behaviour of spin currents propagating across interfaces is crucial.

We study equilibrium and transport properties of both, homogenous and heterogeneous two and three layer systems composed of insulating ferro (FM)- and antiferromagnets (AFM). Our calculations are performed within a classical atomistic spin model, treated numerically.

The materials are characterized by different exchange coupling constants leading to a significant variation of the critical temperatures across the multilayer systems. Temperature dependent studies then can reveal an interesting behaviour such as the steady-state magnonic proximity-effect shown in the figure which depicts the spatial dependent order parameters comparing FM|FM|FM and FM|AFM|FM systems. One can see how the order parameter penetrates the central layer in the vicinity of the interfaces to the ordered outer layers, especially at the critical temperature.

Based on the equilibrium properties, we perform transmission studies of a spin current across the interfaces depending on the magnon frequency and on the interface and bulk properties. This magnonic dynamics is especially interesting close to the critical temperature of the central layer, as investigated in recent experiments [2].



[1] Cramer et al., Nat. Commun., 9, 1089 (2018)

[2] Schlitz et al., Appl. Phys. Lett. 112, 132401 (2018)

3935 Quantum magnonics at the edge of space and time

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*The author has chosen not to make public additional content

3951 Excitation of spin superfluids in easy-plane magnets

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It has been known for a long time that easy-plane magnets exhibit an order parameter with SO(2) symmetry, which is equivalent to the U(1) gauge symmetry of the macroscopic wave function of a Bose condensate. For small outof-plane components the magnetic equations take then a form similar to the Gross-Pitaevski equation, describing the time evolution of a Bose condensate and, hence, superfluidity. Because of this very resemblance, there is a specific type of transport in such magnets called "spin superfluidity" [1-2]. It is characterized by a well defined precession frequency (Ω_0) for all spins and a spin accumulation that spans under ideal conditions over the entire magnet. However, there is a major difference to conventional superfluidity: a spin superfluid always exhibits dissipation because of finite Gilbert damping.

A spin superfluid can be excited, for instance, by inducing a spin accumulation at one end of the magnet (induced by the spin-Hall effect in experimental setups). In this contribution we analytically estimate the resulting precession frequency (Ω_0) of spin superfluids in easy-plane ferromagnets in a first step. The next step is to use atomistic spin simulations to investigate the excitation with respect to changes in the driving strength, the geometry and disorder not covered by the analytical theory. We find that in particular the exact geometry of the excited magnet does play a major role on the resulting frequency (Ω_0). Furthermore, there are two limiting cases: a minimal excitation strength required for a finite spin-superfluid response and a maximum spin-superfluid frequency, which are both also studied numerically. In addition, the atomistic framework readily allows to test also antiferromagnets, which turn out to show almost the same behavior as ferromagnets, except for their lower resulting spin accumulation [3].

- [1] B. I. Halperin et al., Phys. Rev., 188, 898 (1969)
- [2] S. Takei et al., Phys. Rev. Lett., 112, 227201 (2014)
- [3] M. Evers et al., arXiv: 1911.12786 (2019)

Systematic micromagnetic study of the band structure of one-dimensional artificial magnonic crystals in presence of interfacial Dzyaloshinskii-Moriya interaction

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Artificial magnetic materials with periodically varied properties, known as magnonic crystals (MCs), are interesting to modify and control spin waves propagation. These materials are characterized by a symmetric band structure for spin waves propagating in opposite direction. In presence of a heavy metal underlayer, the spin-orbit coupling may induce the so-called interfacial Dzyaloshinskii–Moriya interaction (DMI) in the magnetic material, causing a marked non-reciprocity of the dispersion relation, i.e. removing the frequency degeneracy for counterpropagating spin waves.

In this work we systematically analyse the effect of DMI, whose strength *D* is varied between (±2) mJ/m^2 , on the band structure of periodical magnetic nanostructures, by numerical simulations based on the MuMax3 software. Two different one-dimensional MCs are considered, both with the same periodicity *p=300 nm*, saturation magnetization $Ms = 730 \ kA/m$ and exchange stiffness $A = 10 \ pJ/m$, but different implementation of the DMI modulation. In the first case, the sample is a thin ferromagnetic film (thickness $t = 2 \ nm$) supporting a regular array of parallel heavy metal nanostripes (width $w = 150 \ nm$) equally separated with an edge-to-edge distance *d=150 nm*. In the second case, a continuous heavy metal substrate supports an array of magnetic alternated parallel nanostripes (width $w = 150 \ nm$) with variable thickness ($t_1 = 2 \ nm$ and $t_2 = 4 \ nm$). Note that the above geometrical parameters have been shown to match the characteristics of samples suitable for experimental investigation by Brillouin light scattering (BLS), where the accessible range of wavenumbers is limited (by the used probe, i.e. light) to about $2 \times 10^7 \ rad/m$.

We show that in both model systems, the presence of folded branches of the dispersion curves, caused by the artificial periodicity, permits to access a larger wavevector range if compared to that usually accessible by BLS in plane films. In fact, the periodicity in real space manifest itself in the availability of wavevectors $\left(k_G = \pm \frac{2\pi}{n}\right)$ in

the reciprocal space, that may be summed to the exchanged wavevector between light and spin waves. Consequently, since the frequency non-reciprocity increases linearly with the modulus of the *k*-vector of the spin waves, these results pave the way to extend the sensitivity of BLS experiments to values of the DMI constant *D* smaller than those usually measurable by BLS in plane films. Finally, we analyse how the forbidden band gaps, induced by the artificial periodicity, evolve with the intensity of the DMI. We find that for sufficiently large values of *D*, the lower bands flatten and the gap-width increases, reflecting a spatial localization of the corresponding spin wave modes.

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Understanding magnetization dynamics of a magnetic nanoparticle, with disordered shell, using micromagnetic simulations.

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Research of magnetic nanoparticles (NPs) continues to be of great interest due to its wide range of possible applications, like magnetic fluid hyperthermia [1]. However, magnetization dynamics of real magnetic NPs are affected by both their structural and spin disorder. In order to better understand such effects, micromagnetic simulations using mumax3 [2] were performed, looking in particular to the blocked state magnetization dynamics of a nanoparticle with a "real" spin structure.

For the simulations, a prototype NP with structurally and magnetically coherent core and somewhat disordered shell, was considered. The size of the core was fixed at 5.5 nm in diameter, whereas the shell thickness was varied from 0.5 nm to 3 nm. This prototype NP was constructed based on a cobalt ferrite NP sample, whose structural and magnetic properties were previously characterized in our laboratory [3].

The results obtained from the simulation study allow for a better insight into the interplay between the magnetization dynamics of the core and shell as a function of their anisotropies. The role of the phenomenological damping parameter, thermal field interaction and computational time step was also evaluated. In summary, the performed micromagnetic study showed to be a very useful toolbox for better understanding the magnetization processes within a single domain magnetic NP, with a realistic "magnetic core-shell structure".



Figure 1 : (a) Experimental magnetization isotherms at different temperatures for a real sample of $CoFe_2O_4$ NPs, as measured on a SQUID magnetometer; (b) Simulated internal spin structure for the NP, showing both the ordered core and the disordered shell; (c) Simulated magnetic isotherms when applying the external magnetic field (H_{ext}) along the y-axis, which is perpendicular to the ordered core anisotropy direction (H_k along the x-axis) of the NP.

- [1] Z. Shaterabadi, G. Nabiyouni, and M. Soleymani, Progress in Biophysics and Molecular Biology, 133, 9 (2018)
- [2] A. Vansteenkiste, et al., AIP Advances, 4, 107133 (2014)
- [3] J. Miksatko, et al., Nanoscale 11, 16773 (2019)

Static and dynamic magnetoelectric coupling in TbMnO₃ modified by Fe³⁺ substitution

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Optimal control of magnetization switching in nanowires

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Bistable magnetic nanowires hold great promise as a basis of novel devices for data transmission, storage and processing where information can be encoded by domains with opposite magnetization [1]. Development of this technology depends critically on the knowledge about energy-efficient manipulation of the magnetization.

In this study, we apply optimal control theory to predict energy-efficient protocols of external magnetic field pulses for the magnetization reversal in Co nanowires [2]. We calculate optimal control paths (OCPs) of the magnetization reversal, i.e. reversal trajectories minimizing the energy cost of magnetization switching, and explore how the OCPs depend on various parameters such as the switching time, nanowire length and Gilbert damping. We obtain timeand space-resolved shape and amplitude of optimal switching pulses from calculated OCPs by solving the inverse problem in the Landau-Lifshitz-Gilbert equation.

The OCP calculations demonstrate that short nanowires reverse their magnetization via coherent rotation which can be induced by applying uniform external magnetic field with frequency defined by a collective in-phase precession of the magnetization [3]. If the length of the wire exceeds a certain critical length, standing spin wave emerges and effectively assists the magnetization reversal (see Fig. 1). We find that the critical length at which the crossover between the coherent rotation of magnetization and spin wave assisted switching occurs depends on the switching time and damping parameter. Spin wave assisted magnetization switching has recently attracted much attention as a promising technique to reduce the switching field for magnetic recording [4-5]. Our results provide a valuable insight into this phenomenon, contributing to the development of low-power technologies.

The calculated OCPs are very different from minimum energy paths (MEPs) of the magnetization reversal (see Fig. 1). In contrast to MEPs, OCPs demonstrate dependence on dynamical parameters such as switching time and damping. Moreover, analysis of the energy variation along the OCPs and MEPs demonstrates that the highest energy point along an OCP is typically higher than the energy barrier derived from an MEP (see Fig. 1), suggesting that energy-efficient control of magnetization switching does not necessarily translates into the minimization of the energy barrier between the target states.

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Figure 1: Variation of the energy of a Co nanowire along the minimum energy path (dashed line) and the optimal control path. Reaction coordinate is defined as a normalized displacement along the path. The letters label the states for which magnetic configurations are shown in the upper panel.

- [1] S.S.P. Parkin, M. Hayashi, L. Thomas, Science 320, 190 (2008)
- [2] M.H.A. Badameh, G.J. Kwiatkowski, P.F. Bessarab, Nanosyst.: Phys. Chem. Math. 11, 294 (2020)
- [3] G.J. Kwiatkowski, et al., under review at: Phys. Rev. Lett. Manuscript available online: https://arxiv.org/abs/2004.02146
- [4] T. Seki et al., Nat. Commun. 4, 1726 (2013)
- [5] T. Seki et al., Appl. Phys. Lett. 103, 122403 (2013)

4184 Spin-wave Talbot effect in thin ferromagnetic film

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The Talbot effect has been known in linear optics since XIX century and found various technological applications. With the help of micromagnetic simulations, we demonstrate this self-imaging phenomenon for spin waves (SW) in three basic configurations: (i) out-of-plane magnetized thin film (forward volume) and in-plane magnetized thin film (ii) parallel (Damon-Eschbach) and (iii) perpendicularly (backward volume) to the grating. The first scenario is the closest to the textbook Talbot effect for electromagnetic waves due to isotropic dispersion. However, for the second and third scenario, SWs dynamics is strongly anisotropic what is especially prominent for lower frequencies where dipolar interaction are of great significance. We demonstrate that the properties of SW self-imaging are consistent with the theoretical predictions based on the general formalism of wave optics. This compliance cannot be introduced in advance, rather, Landau-Lifshitz nonlinear equation describing SW propagation must be solved for this purpose. This has systematically been done in our study. By performing micromagnetic simulations in permalloy film with characteristic damping constant (α = 0.005) (Fig. 1), we show that the observation of the first Talbot images shall be feasible with standard micro-focused BLS. We expect, that in Yttrium Iron Garnet (YIG) thin films, the (first) primary images can be reached due to 10 times smaller damping. In addition to out-of-plane magnetization, we have also analyzed the Damon-Eshbach and the backward volume wave geometry to show that the Talbot effect is obtainable for smaller, in-plane aligned external magnetic fields and with anisotropy, for which SW wavelengths depends on direction of propagation. It gives a lot of application possibilities as well as ways of manipulating and adapting the effect to specific needs thanks to additional anisotropic and caustic effects. To prove that, we demonstrate logical functions performed with SWs in finite width ferromagnetic waveguides. The obtained results helps to better understand SWs interference and scattering at the diffraction gratings; moreover, our findings open an avenue to practical application of the Talbot effect in future magnonic devices.



Figure 1 : SW Talbot effect in thin permalloy film with damping constant (α = 0.005) for 3 GHz as 2D intensity plot.

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Subwavelength control of the spin waves phase by ferromagnetic resonators

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The application of arrays of nano-resonators constituting metasurfaces was a ground-breaking concept in modern optics, as it made it possible to design a new class of optical systems with properties exceeding classical possibilities. Metasurfaces enable manipulation of amplitude and phase of electromagnetic waves at ultra-short distances. This idea has not yet found widespread applications in magnonics, although the ability to process waves at very short distances may be of importance especially in the case of spin waves (SWs), whose propagation length is severely limited by damping.

We present here an extensive theoretical research on the influence of the position and geometry of a ferromagnetic stripe (resonator) on SWs reflection from the edge of a permalloy film (Fig. 1). The interactions between the film and stripe affect the phase of reflected SWs, which is found to vary with the stripe's dimensions and its separation from the permalloy film. We study this problem utilizing numerical simulations, employing three different approaches, i.e., the finite element method in the frequency domain, finite element modal method, and micromagnetic simulations, which enable cross-checking of methods. All three employed approaches agree very well with each other. We have found that the dependence of phase shift on stripe width (Fig. 2) is characterized by areas of slow but steady growth, separated by sharp resonances accompanied by a significant increase of the spin wave's amplitude. Our analytical model shows that the reflection coefficient consists of two terms of different origins. The first produces a slow but steady increase of the phase shift with increasing stripe width and is dominated by the phase accumulated by a long-wavelength mode of the stripe-film bilayer during a single round-trip. The second term has an appreciable effect on the reflection coefficient only when conditions are favourable for Fabry-Perot resonances of short-wavelength modes propagating in the bilayer, and it is this term that is responsible for the observed rapid changes of the phase shift with stripe width.

Overall, this research shows that SW Fabry-Perot resonators could be used to modify the phase shift of reflected SWs in a wide range, thanks to the sensitivity of the reflection coefficient to tiny changes of the resonator's width and stripe-film distance. This is significant for the further development of magnonic applications where SW phase control is of key importance, in particular in integrated systems with components smaller than the SW wavelength.

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Spin-wave diode and circulator based on vertically coupled ferromagnetic films

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Low-energy consumption of the spin waves makes the growth of interest of magnonics in computing. Among the magnonic logic devices, the spin-wave diode was already proposed [1,2], while the spin-wave circulator has not been proposed yet. Basing on numerical simulations, we design dipolarly-coupled ultra-thin ferromagnetic multilayers which work as a diode or a circulator for spin waves, which utilizes the interfacial Dzyaloshinskii-Moriya interaction [3]. Our devices base on the effect of unidirectional coupling which allows to transfer the spin waves between the ferromagnetic layers in only one direction of their propagation, while in the opposite direction, the spin wave can propagate in one layer only. A diode is made from narrow Co/Pt stripe as a coupler laying above the low-damping magnetic layer like permalloy. The difference between the signal power measured in forward and reverse direction exceeds 20 dB. A circulator is made from two Co/Pt multilayers in the opposite arrangement and permalloy stripe in between working as a coupler. With this composition, we design the three- and four-port circulators that complete the list of magnonic signal processing devices. Our work contributes to further development and miniaturization of the spin-wave logic devices, making them competitive with the electronic logic components.

[1] J. Lan et al., Spin-wave diode, (2015), Phys. Rev. X 5.4 041049

- [2] M. Grassi et al., Slow-wave based magnonic diode, (2019), arXiv:1912.09735
- [3] K. Szulc et al., Spin-wave diode and circulator based on unidirectional coupling, (2020), arXiv: 2002.06096

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4312 Quantum effects in magnetic transition rates

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Most widely used data storage technologies are based on nanoscale magnetic structures [1]. Due to the ongoing miniaturization of electronic systems, long-term stability of stored information has be-come a pressing issue. Most commonly, mean lifetime (τ) of a given state is described by Arrhenius law:

$$\left(\tau = f^{-1} e^{\frac{\Delta E}{k_B T}}\right)$$

Where (ΔE) is the energy barrier, (k_B) is the Boltzmann constant, (T) is the temperature and (f) is a prefactor dependent on the internal dynamics of a given sample. Energy barrier can be resolved using geodesic nudged elastic band method [2], while the prefactor is often calculated using classical rate theories [3], which assume all internal modes of freedom to be thermally activated.

In this work we show that classical models based on Boltzmann statistics are often not applicable to spin systems as a significant part of the internal modes is not activated thermally even in high temperatures, which is due to the extremely high frequencies of magnon modes. As we present, the difference between classic theory and one with quantum corrections is most significant for 3D systems and can reach over 10 orders of magnitude, while for 1D systems the prefactor changes by less than 50%.

As the internal degrees of freedom have such a strong impact on mean lifetime calculations, it is important to carefully establish the numerical model - especially in the case of micromagnetics. We show, how the chosen coarse graining affects the results (see Fig. 1) and how to estimate the minimum resolution required for accurate calculations.



Figure 1: Prefactor for uniform rotation switching mechanism of a cubic ferromagnetic grain with an edge 30 spins long calculated using classic and quantum statistics at ($T = T_c/10$) as a function of coarse graining resolution, where (N) is the number of nodes along an edge of the cube and (f/f_0) is the prefactor normalised to classic macrospin solution [4]. Inset depicts the increase in coarse graining resolution.

[1] W. A. Challener et al. Nat. Photon. 3, 220 (2009)

[2] P. F. Bessarab et al. Computer Physics Communications **196**, 335 (2015)

[3] P. F. Bessarab et al. Phys. Rev. Lett. 110.2, 020604 (2013)

[4] W.F. Brown, Phys. Rev. 130, 1677 (1963)

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Symposium 5. Ultrafast laser induced magnetization dynamics and terahertz spintronics (including Magneto Optical phenomena)

Ultrafast Magnetization Dynamics in Antiferromagnets

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Antiferromagnets represent the largest class of magnetically ordered materials. It is also believed that antiferromagnets can dramatically improve spintronics in terms of density and speed. However, experimental studies of antiferromagnets and the development of antiferromagnetic spintronics are considerably hampered by the lack of the net magnetization in antiferromagnets in unperturbed state. It makes conventional magnetometry impossible and requires exceedingly high magnetic field to control spins.

The goal of my talk is to demonstrate that ultrashort electromagnetic pulses is a game changer in the field. Starting from the simplest example showing that a weak, but rapidly varying magnetic field is an efficient mean to control antiferromagnetically coupled spins, I will discuss the fundamental limits on the speed with which an antiferromagnet can acquire the net magnetization. I will pay a special attention to the role of the lattice in this process and describe coupled magneto-structural dynamics in ultrafast optical control of antiferromagnetism.

Ultrafast spin dynamics: ab-initio description

Sangeeta Sharma¹, John Kay Dewhurst² ¹Max Born Institute ²Max Planck Institute of Microstructure Physics;

I will talk about all-optical switching of long-range magnetic order. The type of coupling between the constituent atoms of a magnetic solid, usually ferromagnetic (FM) or anti-ferromagnetic (AFM), is a fundamental property of any magnetic material. This coupling is governed by the exchange interaction, for which the time scale of a typical magnetic material is of the order of a few 100s of femtoseconds. In our work, using time-dependent density functional theory (TDDFT), we demonstrate that a rich control over magnetization at sub-exchange time scales (of the order of few tens of femtoseconds) is possible [1,2,3,4]. This even includes changing the magnetic order from AFM to FM [5]. By investigating a wide range of multi-sublattice magnetic materials we are able to formulate three simple rules that predict the qualitative dynamics of magnetization for ferromagnetic, anti-ferromagnetic, and ferrimagnetic materials on sub-exchange time scales.

[1] Dewhurst et al., Nano Lett., 18, 1842 (2018)

- [2] Elliott et al., Scientific Reports, 6, 38911 (2016)
- [3] Shokeen et al., Phys. Rev. Lett., 119, 107203 (2017)
- [4] Chen et al., Phys. Rev. Lett., 122, 067202 (2019)

[5] Siegrist et al., Nature, 571, 240 (2019)

Deterministic All-Optical Magnetization Writing Driven by Spin Currents

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Single pulse all-optical switching (AOS) of magnetization by fs laser pulses has been proposed as a candidate for writing data in future ultrafast memory applications. However, in this regard it is limited by being a toggle mechanism, where the final state is always the opposite of the initial state.

In this contribution, we show that optically induced spin currents generated in a magnetic reference layer can significantly impact single pulse AOS in an all-optically switchable free layer. Using this mechanism, we experimentally demonstrate deterministic all-optical magnetization writing, where the final state of the free layer depends purely on the laser pulse sequence and the magnetization direction of the reference layer [1]. The system under investigation (sketched in Fig. 1) consists of a Co/Ni multilayer, which serves as a reference layer, separated by a conductive Cu spacer from a free Co/Gd bilayer. This latter has been shown to exhibit single pulse AOS [2]. Upon excitation with a fs laser pulse the reference layer demagnetizes, generating a spin current which flows through the spacer to the free layer. The angular momentum carried by the mobile spins is then absorbed in the free layer. We conjecture that formation of the temporary ferromagnetic state which is essential for single pulse AOS [3] is affected by this additional angular momentum.

We find that the threshold laser fluence for single pulse AOS is significantly lower when the moments of the reference layer and the top Co layer are initially antiparallel, compared to the parallel alignment (as sketched in Fig. 1). We demonstrate that the gap between the two fluences can be as large as 25%, and prove the nature of the effect by showing that this gap vanishes when inserting a Pt spin sink layer between the free and reference layers. We show that our mechanism allows for deterministic writing of both states of the free layer, independent of the initial state. This is realized by using either a single, or a sequence of two laser pulses, with carefully selected energies. Time-resolved optical pump-probe measurements are underway to provide more insight into the effect of the spin current on the (sub-) ps timescale.

Besides offering a novel platform for investigating the nature of optically induced spin currents, the deterministic magnetization writing in these structures could lead to more straightforward integration or even new applications of AOS in data storage and logic devices.



Figure 1 : Sketch of the magnetic multilayer stack under investigation.

^[1] van Hees, Y.L.W. et al., Under review, arXiv:2001.09033 (2020)

^[2] Lalieu, M.L.M. et al., Physical Review B, 96, 220411 (2017)

^[3] Radu, I. et al., Nature, 472, 205 (2011)

Optically induced spin transfer phenomena in magnetic alloys

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Magneto-refractive SHG from plasmonic multilayer antennas

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Recently, a breakthrough in infrared magneto-plasmonics has been achieved [1], utilizing the magneto-refractive effect (MRE) in magnetic multilayers and opening the door for a novel branch of infrared nanophotonics. The intrinsic reliance of MRE on the diagonal components of the dielectric tensor, in contrast to the off-diagonal ones responsible for magneto-optical effects, has shown its strong potential for magneto-plasmonics. In particular, sizeable (up to 5%) modulation of the magneto-refractive response in plasmonic systems in the mid-infrared spectral range has been observed [1]. This is a significant improvement upon the usual values on the order of 1% which are found in a number of conventional magneto-plasmonic systems.

In our work, we show that MRE systems can constitute an important step towards solving the main problem of nonlinear magneto-plasmonics, combining large nonlinear-optical yield and strong magneto-optical effects. We utilized the infrared radiation from a free electron laser for performing second harmonic generation (SHG) spectroscopy measurements on resonant magneto-refractive Au/Py antennas with interlayer exchange coupling [2]. This coupling is responsible for the antiferromagnetic alignment of the magnetizations in adjacent Py layers in ground state. By means of an external in-plane magnetic field, this alignment can be forced into a parallel state with different optical properties. The antennas exhibit a pronounced plasmon resonance in the mid-infrared range, responsible for an enhancement of SHG output observed in the experiments. Moreover, upon applying the magnetic field the modulation of the SHG output at the resonance was as high as 20%. In contrast to conventional magneto-plasmonic systems, we found that the strongest SHG intensity from these MRE antennas coincides (spectrally) with the largest magnetic contrast.

We further note that the 20 % modulation cannot be simply explained by the modification of the local field enhancement due to the different dielectric functions in the parallel and antiparallel states. Instead, we discuss a model where the nonlinear susceptibility of the entire multilayer antenna depends on its magnetic state. In the antiparallel state of the antenna, the effective scattering rate of the electrons obtains a non-zero spin-dependent contribution (the spin valve is closed) which is absent in the parallel state. As such, the effective nonlinear susceptibility of the free electrons gets a second-order (in magnetizations) correction term which results in unequal SHG yield in the two states. Becoming characteristic of infrared magneto-plasmonics, this nonlinear-optical mechanism of the magnetic SHG modulation demonstrates increased efficiency into the infrared spectral range. As the first demonstration of magneto-refractive infrared SHG, our results stimulate the expansion of nonlinear magneto-plasmonics into previously unexplored spectral domains where strong nonlinear MRE effects can be expected.

[1] G. Armelles et al., ACS Photonics, 5, 3956 (2018)

[2] S. S. P. Parkin et al., Phys. Rev. Lett., 72, 3718 (1994)

Light induced magnetization dynamics in ultrathin Bismuth doped garnet: pumping from the absorption edge to the transparency

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Conventional electronics relies on charge transport to operate, which is intrinsically dissipative. To improve both energy efficiency and speed of future devices, the spin degree of freedom can be exploited. Therefore, the understanding and the control of spin dynamics is of paramount importance for upcoming technological devices. Collective excitation of spins, so-called spin waves, can travel over micrometer distances in materials with low magnetic damping such as Yttrium Iron Garnet (YIG)¹. Nanometer-thick YIG films with low magnetic losses enables now the fabrication of spin waves devices at the micro- and nanometer scale with standardized lithography techniques^{2,3}. In those ultrathin films, RF antennas and DC current induced spin transfer torques⁴, are the most common ways to drive spin dynamics. Here, we demonstrate an alternative approach by using optical light pulses to trigger magnetization dynamics in an ultrathin film with low magnetic losses.

Using a femtosecond pump-probe approach to excite and observe magnetization dynamics in a 17 nm thick bismuth-doped YIG film, we pumped the material above and below bandgap using two different pump wavelengths: 400 nm (3.1 eV) and, respectively, 800 nm (1.55 eV). By performing fluence-dependent, time-domain measurements of both transmissivity and Faraday effect, we could determine how the energy is absorbed and dissipated in the garnet film in the two cases. For both pump wavelengths, we could trigger magnetic precession via ultrafast lattice heating, although we could evidence that the absorption processes were quantitatively different in the two cases. The amplitude of the magnetic oscillations showed instead a qualitatively different trend depending on the pump energy. For energy below bandgap, the magnetic oscillations scaled proportionally with the pump fluence as seen in Fig.1(a), while above bandgap a more complex behavior was observed, as shown in Fig.1(b). In the latter case, the pump-deposited energy caused reversible modifications of absorption coefficient, magnetic moment and uniaxial magnetic anisotropy, as seen in Figure 2(a) and (b). Our study shows that ultrafast optical pulses are an efficient way to trigger magnetization dynamics in ultrathin Bismuth YIG films.



Figure 1 : Fluence dependency measurement of the Faraday effect response for 800 nm (a) and 400 nm (b) pump.

- [1] Evelt, M. et al., Phys. Rev. Appl., 10, 041002 (2018)
- [2] Collet, M. et al., Nat. Commun., 7, 10377 (2016)
- [3] Demidov, V. E. et al., Sci. Rep., 6, 1-8 (2016)
- [4] Kajiwara, Y. et al., Nature, 464, 262–266 (2010)



Figure 2 : Fluence dependency of the resonant frequency (a) and the corresponding evaluated effective field (b) for 400 nm pump.

Ultrafast Light-Induced Nucleation of Skyrmion Lattices

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The use of magnetic skyrmions in technological applications is constrained by the ability to nucleate, stabilize and manipulate them[1]. Small skyrmions are frequently metastable states, so that at remanence either the saturated or the stripe domain states are present and the skyrmion nucleation is not easily accessible. Recent experiments have shown the feasibility of laser-induced nucleation of skyrmions by means of non-equilibrium ultrafast magnetisation dynamics processes [2]. This technique also opens the possibility of developing less-consuming and ultrafast skyrmionic memory or logic-gate devices.

In the present work we explore the nucleation of magnetic skyrmions in realistically parametrised Pt/Co/Heavy-Metal magnetic trilayers via atomistic simulations using the software package Vampire [3]. Quasi-static simulations by increasing temperature above the Curie one and cooling the system down produce a stable state in the form of stripe domains. Next, we model the dynamics under ultrafast non-polarised femtosecond laser pulse varying the pulse duration and intensity. We present a state diagram of the magnetic states after the laser pulse action which shows the existence of small region of parameters where the final state is the skyrmion lattice. This region can be increased by application of static external field parallel or antiparallel to the initial magnetic state. Our results unambiguously demonstrate the necessity of highly non-equilibrium dynamical path which could lead to the skyrmion lattice.



Figure 1: Sketch of the modelled system and simulated processes: **a)** Modelled Pt/Co/HM magnetic trilayer. The texture on the top of the system represents the out-of-plane component of the magnetisation before applying the laser-pulse; **b)-c)** Final states obtained after following a zero-field slow-cooling process and after applying a laser pulse with a fluence of 56 mJ/cm² and a pulse width of 10 fs, respectively; **d-e** Temperature profile of the processes leading to the spin configurations at panels **b** and **c**.

[1] Fert, A., Reyren, N., Cros, V., Nature Reviews Materials, 2, 17031 (2017)

- [2] Je, S-G. et al., Nano Lett., 18, 7362 (2018)
- [3] Evans, R.F.L. et al., J. Phys. Condens. Matter, 26, 103202 (2014)

Probing the antiferromagnetic to ferromagnetic phase transition of FeRh with femtosecond time-resolved small angle X-ray scattering (tr-SAXS) and X-ray absorption spectroscopy (tr-XAS)

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The observation of ultrafast manipulation and coherent control of spins in magnetic materials at room temperature [1-2] has prompted the intense experimental and theoretical efforts to understand the underlying microscopic mechanisms driving such magnetic phenomena at sub-picosecond timescales. In order to understand the driving mechanisms, in this non-equilibrium regime, it is necessary to disentangle the relevant interactions (exchange, spinlattice, electron-phonon, coulomb etc.). Stoichiometric B2 ordered epitaxial (001) FeRh undergoes a first order magnetic phase transition from antiferromagnetic (AFM) to ferromagnetic (FM) at \approx 380K. In the AFM phase of FeRh, only Fe has net magnetic moment, whereas in the FM phase both Fe and Rh carry net moments. The phase transition is also accompanied by a \approx 1% isotropic lattice expansion in the bulk BCC structure, and changes in electronic structure, see [3-5] and refs. therein. Since magnetic and structural dynamics occur at different timescales, it makes FeRh an ideal candidate for disentangling relevant interaction mechanisms at different timescales. The phase transition has been extensively studied theoretically and experimentally [6-10], in thermal equilibrium and in the time domain, but a precise understanding of the roles of the electronic, phononic and spin sub-systems remains elusive. To disentangle relevant driving mechanisms for this AFM-FM phase transition, we have studied the laser-driven AFM to FM phase transition in FeRh with small angle X-ray scattering (tr-SAXS) and Xray absorption spectroscopy (tr-XAS) around the Fe L3 edge. The experiments were performed at the SCS Instrument of the European XFEL. We also performed the time-dependent density functional theory (TD-DFT) calculations to simulate the XAS spectra at different time delays. In this contribution, we will discuss the changes of the electronic structure of FeRh on the femto- and picosecond timescales, derived from the XAS and scattering measurements at the Fe L3 absorption edge performed at European XFEL, and compare them with the theoretical simulations, in order to show the relation between them and the first order AFM-FM phase transition in FeRh.



Figure 1 : a X-ray absorption (XAS) spectrum measured in transmission geometry near the Fe L3 edge before and after laser excitation at a fixed time delay of 300 fs; **b** TDDFT simulation for XAS scan in AFM and FM state of FeRh; **c-e** The difference between the excited and equilibrium XAS specta @ 300 fs and 300 ps respectively; **d-f** TDDFT simulations for difference in XAS spectra for transient states (excited - eqb AFM) and equilibrium states (eqb FM - eqb AFM).

- [1] E. Beaurepaire et al., Phys. Rev. Lett., 76, 4250 (1996)
 [2] J.-Y. Bigot et al., Nature Physics, 5, 515 (2009)
 [3] J.-U. Thiele et al. Appl. Phys. Lett., 82, 2859 (2003)
 [4] A.X. Gray et al., Phys. Rev. Lett., 108, 257208 (2012)
 [5] C. Baldasseroni et al., Appl. Phys. Lett., 100, 262401 (2012)
- [6] J.-U. Thiele, et. al., Appl. Phys.Lett, 85, 2857 (2004) [7] G. Ju et al., Phys. Rev. Lett., 93, 197403 (2004)
- [8] I. Radu et al., PRB, 81, 104415 (2010)
- [9] S. Mariager et al., Phys. Rev. Lett., 108, 87201 (2012)
- [10] F. Quirin et al., Phys. Rev. B, 85, 020103(R) (2012)

Ultrafast laser induced magnetization dynamics in Fe₃GeTe₂

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Van der Waals materials are characterized by strong in-plane covalent bonds and weak out-of-plane Van der Waals bonds. As a result, crystalline single layers with near-perfect surfaces can easily be removed from the bulk, providing a unique platform for the study of 2D physics. Furthermore, the weak inter-plane bonds allow for stacking of different Van der Waals materials on top of each other regardless of lattice matching and relative orientation, making these materials ideal building blocks for (complex) multilayer structures. Van der Waals materials have garnered a lot of attention from the physics community since the first synthetization of graphene in 2004. However, magnetic order in Van der Waals materials such as Cr₂Ge₂Te₆ and Fe₃GeTe₂ (FGT) was only discovered recently, and magnetic characterization has mostly been limited to studies of their static magnetic properties [1,2].

In this work, we report the first observation of ultrafast laser induced demagnetization dynamics of a 2D Van der Waals magnet, more specifically, the itinerant ferromagnet FGT. In figure 1b and c, the Magneto-optical (M.O) contrast of the studied flake after saturation with a positive and negative field respectively is visualized. At the top of the flake, a relatively large single-domain area is observed. In this region, pump-prove experiments are employed to map the laser induced dynamics on a sub-picosecond timescale. In figure 1a, the demagnetization trace is visualized. After laser excitation, the FGT magnetization is quenched on two distinct timescales. This is reminiscent of type II magnetization dynamics, where electron-phonon equilibration (τ_e) is faster than the spin system relaxation (τ_m) [3]. This behavior persists in a wide temperature range (80K – 150K), and we observe a critically slowing down of the demagnetization while approaching the Curie temperature. From these measurements, demagnetization timescales of 10s of picoseconds have been extracted, which is one to two orders of magnitude larger than typically observed in transition magnets such as Co and Ni [3, 4].

Our results thus give insights on the microscopic interactions between electrons, phonons and spins in FGT. In order to increase our microscopic understanding of the magnetization dynamics, we plan a systematic layer-dependent study of the demagnetization timescale for a wide range of temperatures in the near future. Furthermore, more detailed analysis in terms of the Microscopic 3 temperature model is underway [3].



Figure 1 : a Demagnetization trace of thin FGT at 90K, where the red dashed line is a fit with the 3-temperature model. Two distinct timescales can be observed. The inset depicts the studied FGT flake, where the red dots indicate the large pump beam and smaller probe beam and the light-blue circle denotes the studied area. The flake is approximately 10 monolayers thick; **b** and **c** Scanning magneto-optical microscopy images after saturation with a positive and negative field, respectively.

[1] C. Gong, et al., Nature, 546, 265 (2017)

- [2] Y. Deng, et al., Nature, 563, 94 (2018)
- [3] B. Koopmans et al., Nature Materials, 9, 259 (2010)
- [4] E. Beaurepaire, Physical review letters, 76, 4250 (1996)

Designing energy-efficient layered structures for magneto-optical recording in the infrared spectral range

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Learning how to optically address magnetization with maximum efficiency represents a key goal of ultrafast magnetism, with the ultimate target of improving the speed and efficiency of data-storage technologies. Here, we perform experiments at the Free Electron Laser for Infrared eXperiments (FELIX) facility to investigate how the efficiency of all-optical switching of magnetization in multi-layered structures can be improved in the infrared spectral range [1].

In our first experiments, we surprisingly demonstrate that helicity-dependent all-optical switching of magnetization can be achieved using pulses with a central wavelength of 9 μ m [Fig. 1]. By sweeping circularly-polarized pulses across the GdFeCo alloys, we all-optically switch the magnetic polarity of one domain but not the other. We attribute the effect to the 60 nm-thick SiN layer that caps our magnetic alloy, which enhances the magnetic circular dichroism of our structure [2].

In our second experiments, we discover a peculiar effect associated with silicon-mounted GdFeCo samples that allows one to deliver up to 50% more energy to adjacent magnetic structures. Upon focusing the infrared optical pulse through the Silicon substrate, we clearly identify that the region of addressed magnetization is larger compared to the result of focussing directly on the metallic layer [Fig. 2]. We discuss possible origins of this effect, which may include layer-resolved absorption profiles or non-linear self-focussing effects [3].

Our results may have profound implications for the design of efficient all-optical data-writing technologies, opening up new avenues for engineers seeking to minimize energy costs.



Figure 1: Typical magneto-optical images (of height 100 µm) showing helicity-dependent (σ ±) all-optical switching of magnetization (M \uparrow ↓) in different Gdx(FeCo)100-x alloys, using pulses of wavelength 9 µm.

C. S. Davies *et al.*, Phys. Rev. Appl., **13**, 024064 (2020)
 A. R. Khorsand *et al.*, Phys. Rev. Lett., **108**, 127205 (2012)
 G. Fibich, A. L. Gaeta, Opt. Lett., **25**, 335 (2000)

Figure 2: Size of the optically-addressed spot as a function of the incident macropulse energy, with the optical pulse (of wavelength indicated) incident either on the metallic GdFeCo layer (unfilled symbols) or on the silicon substrate (filled symbols).

Anisotropic ultrafast demagnetization and spin relaxation in epitaxial cobalt thin films

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Ultrafast photo-magnetic back-switching in garnet films

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Detailed investigation of the key material parameters in examined structures and its contribution to the ultrafast all-optical magnetization switching mechanism is crucial for the development of the underlying switching model, which may provide next step to the invention of applicable devices. Recently, it was discovered that without any external magnetic field only by a single laser pulse reversible photo-magnetic switching in Co-doped yttrium iron garnet film (YIG: Co) can be obtained [1]. Such switching involves a single, linearly polarized pulse for recording the 'up' magnetic bit. The subsequent laser pulse with orthogonal polarization reverses the magnetization to the initial state, recording 'down' magnetic bit. The substrate (GGG) in this garnet was tilted from the surface normal by roughly 4°. It introduces an additional in-plane anisotropy, acting on the magnetic domain structure.

Here, we demonstrate the back-switching mechanism, which was observed in the YIG: Co with a similar composition without miscut of GGG substrate. For the investigation of photo-magnetic switching, we employed single-shot magneto-optical microscopy at Faraday geometry. The magnetization in YIG: Co thin films was switched by the single linearly polarized pump beam at 1300 nm wavelength with 50 fs pulse duration. Imaging was ensured by the CCD camera illuminated by a polarized LED light. In contrast to the sample with miscut, the static magneto-optical visualization of the symmetric sample gives a characteristic labyrinth-like structure. The pump pulse locally switches the magnetization at the domains but does not change the shape of the labyrinth structure. The final magnetization state depends on the initial magnetization state and the number of incident pulses. Therefore, the even number of incident pulses leaves the same magnetic state, and the odd number of pulses reverses it. This back-switching mechanism may seem to be qualitatively similar to the toggle heat-induced all-optical switching in metallic ferrimagnets [3]. However, in our case, the physical mechanism based on the non-thermal photo-magnetic modification of anisotropy drives precessional reversal of magnetization in a dielectric. Obtained results highlight the possibilities for the engineering of devices for the cold all-optical recording by tuning of the laser photon energy, polarization and pulse selection.

[1] A. Stupakiewicz, et al., Nature, 542, 71 (2017)

- [2] A. Stupakiewicz, et al., Nat Commun., 10, 612 (2019)
- [3] T. A. Ostler et al., Nature Commun., 3, 666 (2012)
Ultrafast Nonequilibrium Phonon Dynamics in Ferromagnetic Metals

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Magnetization can be modified on unprecedently short timescale employing femtosecond lasers. The associated magnetization and lattice dynamics has commonly been described employing the three temperature model. However, its limits should be more thoroughly examined. Here we study out-of-equilibrium phonon population dynamics within femtoseconds after the laser pump, and show its difference from the thermalized one. We have calculated electron-phonon scattering rates for systems with high electronic temperatures [1], and phonon lifetimes due to phonon-phonon scattering. From these we obtain phonon populations that differ sharply from the thermal ones within picoseconds after the pump [2]. This allows us to understand recent experimental observations and disproves the applicability of the model which assumes a transient thermal behaviour for the lattice [3]. Advanced experimental methods have made it possible to observe phonon population dynamics throughout the Brillouin zone and compare it to theoretical predictions for Ni [4].

In FePt nanoparticles a significant anisotropy of size change was observed change for the in- and out-plane directions upon the laser pump, in fact the lattice shinks along the c-axis, while it expands along a, b axis. After about 3ps the lattice parameter c starts to revert back to its original value, while a, b remains expanded for longer times. This unexpected behavior reveals that on such short timescales the relation between lattice dimensions and magnetization cannot be described just by the standard magnetostriction theory [5]. Ab initio calculations of the induced stress including the non-equilibrium phonon population recover this behavior when coupled together with a magnetic stress, which corresponds to the tendency to modify lattice parameters to those of a paramagnetic state. Our analysis allows us to suggest that the effect of magnetoelastic stress is seen on a sub-ps timescale, while later within ps the increasing occupation of phononic modes leads the system to a different type of deformation.

- [1] K. Carva, M. Battiato, P. M. Oppeneer, Phys. Rev. Lett., 107, 207201 (2011)
- [2] P. Maldonado, K. Carva, M. Flammer, P. M. Oppeneer, Phys. Rev. B, 96, 174439 (2017)
- [3] T. Henighan et al., Phys. Rev. B, 93, 22030 (2016)
- [4] P. Maldonado et al., Phys. Rev. B, 101, 100302 (2020)
- [5] Reid et al., Nat. Comm., 9, 388 (2018)

Oral Presentation

Ultrafast demagnetization following excitation with terahertz and optical fields

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Studied for more than 20 years after the first discovery [1], the fundamental process of ultrafast demagnetization has reached a deeper understanding, but some questions still remain unanswered [2,3]. Among the most important ones is how the spin angular-momentum transfer from the excited electrons to the crystal lattice depends on the nature of the initial distribution of the excited electrons (thermal vs nonthermal). A promising approach is to use different photon energies for the excitation and to, thus, directly tune the initial electron distribution. Low photon energies of 4 meV (which is smaller than the thermal energy of 25 meV at 300 K) are expected to induce a transient quasi-thermal distribution induced with three orders of magnitude higher optical photon energies of 3 eV. By comparing the magnetization dynamics following such different excitations, one can draw important conclusions about the role of nonthermal carriers in the ultrafast demagnetization process.

In this, work we perform a direct comparison of magnetization dynamics induced in a thin iron film by ultrashort optical (3 eV) and terahertz (THz, 4 meV) pump pulses [4-6]. The dynamics are monitored by a subsequently arriving probe pulse (1.5 eV) that detects both the polarization rotation and ellipticity due to the magnetooptical Kerr effect [7]. By varying the the sign of the magnetic field and the polarity of the THz pump pulse, we are able to extract the signals related solely to the demagnetization process in the small-perturbation linear regime. By deconvoluting the temporal profiles of the pump pulse and the measured signals, we are to obtain demagnetization dynamics following pump pulses of different photon energy but with the same duration. A central result of this analysis is shown in Fig. 1. Here, demagnetization traces for rotation and ellipticity variation are normalized and directly compared for two pump photon energies. The grey area indicates the pump-pulse shape which, after the deconvolution procedure, is the same for THz and optical excitation. Although the photon energies differ by three orders of magnitude, variations of both ellipticity and rotation remain the same up to the experimental uncertainties and within time resolution of about 100 fs. We conclude that on time scales of 100 fs and slower, ultrafast magnetization quenching is independent of the pump photon energy and dictated by thermal electrons. This observation justifies a greatly simplified thermal theoretical description of this phenomenon and, potentially, more complex processes such as all-optical magnetization switching.



Figure 1 : Temporal evolution of the probe rotation and ellipticity following excitation with 3.1 eV (400 nm) and 1 THz pump pulses. The grey area shows the pump pulse shape for both cases.

- [1] E. Beaurepaire, J.-C. Merle, A. Daunois, J.-Y. Bigot, PRL 76, 4250 (1996)
- [2] B. Koopmans et al., Nat. Materials, 9, 259 (2010)
- [3] K. Carva, M. Battiato, P. M. Oppeneer, PRL 107, 207201 (2011)
- [4] S. Bonetti et al., PRL 117, 087205 (2016)
- [5] R.B. Wilson et al., PRB 96, 045105 (2017)
- [5] M. Shalaby et al., PRB 98, 014405 (2018)
- [6] I. Razdolski et al., J. Phys.: Condens. Matter 29, 174002 (2017)

Oral Presentation

Linear and Nonlinear Mechanisms of Terahertz Excitation of Antiferromagnetic Spins

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It is believed that using high-intensity ultrashort terahertz (THz) pulses it must be possible to control spins in antiferromagnets in the fastest and the least dissipative way and the mechanisms of excitation of antiferromagnetically coupled spins is presently a subject of intense research. We have explored these mechanisms in ferrimagnets (Tm₃Fe₅O₁₂), compensated antiferromagnets (CoF₂) and antiferromagnets with canted spins (alpha-Fe₂O₃ and FeBO₃ [1]) using nearly single cycle THz pulses with the peak electric and magnetic fields of 1 MV/cm and 0.33 T, respectively. It is shown that the pulses can efficiently excite spins via linear and nonlinear coupling of the THz electric and magnetic fields. Using both the experimental results and theoretical analysis based on the Lagrangian formalism we derive selection rules for these excitations and define the experimental geometries allowing optical detection of the oscillating antiferromagnetically coupled spins in the most efficient way.

[1] Phys. Rev. Lett., 123, 157202 (2019)

An s-d model for local and non-local spin dynamics in laser-excited magnetic heterostructures

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In the nineties, it was shown that a femtosecond laser pulse can quench the magnetization of a ferromagnetic material on a subpicosecond timescale [1]. Later, it was demonstrated that the laser pulse generates a spin current [2]. The microscopic origin of these ultrafast phenomena is still heavily debated. Recent experimental [3] and theoretical [4] studies imply that the s-d interaction, which describes the coupling of the localised magnetic moments and itinerant spins, could be the dominant mechanism driving both ultrafast demagnetization and spin current generation in magnetic heterostructures.

In this work [5], we discuss a joint microscopic model for the laser-induced magnetization dynamics and spin transport in magnetic heterostructures, based on the s-d interaction. The model describes the coupled dynamics of the local magnetization and out-of-equilibrium spin accumulation, and includes diffusive spin transport. We relate this description to the microscopic three-temperature model and show that the models converge in the limit of a strong s-d coupling. The calculations for magnetic heterostructures show that during laser-induced demagnetization in the ferromagnetic layer, a short-lived spin accumulation is created that acts as a bottleneck for the demagnetization process. Secondly, the spin accumulation leads to the generation of a spin current at the interface of a ferromagnetic and non-magnetic metal. Depending on the specific system, both local spin dissipation and interfacial spin transport are able to enhance the demagnetization rate by providing relaxation channels for the spin accumulation in the ferromagnetic material.

Figure a) shows the magnetization in a typical transition metal ferromagnet as a function of time after laser-pulse excitation at t=0, for different values of the (spin-flip) relaxation time of the spin accumulation. It clearly shows that the demagnetization is enhanced when the spin accumulation relaxes efficiently. Figure b) shows the demagnetization as a function of the thickness of the ferromagnetic layer. The demagnetization is enhanced for thin ferromagnetic layers due to the injection of spins into the non-magnetic layer. Finally, we will discuss how the model can be extended to describe the laser-induced magnetization dynamics in non-collinear magnetic heterostructures and the generation of coherent spin precession in a secondary layer.



- [1] E. Beaurepaire, J.-C. Merle, A. Daunois, Y.-I. Bigot, PRL, 76, 4250 (1996)
- [2] G. Malinowski, et al., Nat. Phys., 4, 855, (2008)
- [3] G.-M. Choi, B.-C. Min, K.-J. Lee, D.G. Cahill, Nat. Comm., 5, 4334 (2014)
- [4] E.G. Tveten, A. Brataas, and Y. Tserkovnyak, PRB, 92, 180412(R) (2015)
- [5] M. Beens, R.A. Duine, B. Koopmans, arXiv: 2005.03905

Ultrafast spin dynamics of a multidomain antiferromagnet with strong magnetoelastic coupling

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Searching for efficient manipulations of antiferromagnetic-based devices, we consider spin dynamics of a multidomain antiferromagnet (AF) induced by femptosecond optical pulses or spin currents. We focus on an antiferromagnet with a pronounced magnetoelastic effect, which can play a significant role in the magnetic response. Using NiO as a model system, we calculate the magnon spectra of a multidomain sample and compare them with the single-domain case. The spectra of a multidomain sample includes two localised modes, that correspond to oscillations of the wall separating two T domains. The eigen-frequencies of these modes scale with the magnetoelastic constants and are close (but not equal) to the frequencies of coherent magnetic oscillations in a single-domain sample. We also demonstrate coupling between the different magnon modes propagating in a multidomain sample and resulting in mixing of magnon frequencies and magnon polarizations. This effect results from the symmetry breaking of the magnetic anisotropy landscape induced by the multidomain structure. Studying the nonlinear regime we further demonstrate the possibility of efficient and controllable frequencies.

What is the interplay of ultrafast charge and spin dynamics in ferromagnetic thin films?

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In order to integrate ultrafast magnetism in applied devices, more and more technologically relevant systems are nowadays composed of multiple elements. In such composite systems, the interplay of charge and spin between the different elements is not yet fully understood. Since the advance of free electron laser and high harmonic generation sources, the investigation of ultrafast magnetic dynamics has taken advantage of shorter probe wavelengths in the EUV and soft X-ray range with the benefits of nanometer spatial resolution, element specificity or few femtosecond time resolution. Despite a lot of studies, none have probed directly and simultaneously the electronic and magnetic dynamics of two distinct ferromagnetic (FM) layer in one snapshot.

We developed recently a novel optical element that allows us to follow with a single X-ray pulse the individual evolution of the absorption cross section of two elements (Fe and Ni) of a tri-layer material over a time window of a few picoseconds. This new experimental geometry intrinsically avoids any timing uncertainty between the two elements and unambiguously reveals an approximately 100 fs delay of the magnetic response with respect to the electronic excitation for both Fe and Ni. While the electronic dynamics can be explained by a red shift of the absorption spectra under the infrared excitation [1,2], the meaning of the delayed onset between the electronic and magnetic response is still unclear [3,4]. In the soft X-ray wavelength range, especially at photon energies resonant with element absorption edges used for element selectivity, it has been observed that ultrafast electronic effects could modify the absorption spectra [1]. Although these modifications are small, they could have an impact on the magnetic dynamic and could explain the experimentally observed delayed onset between the electrons and spins response [2]. However a systematic investigation of the relationship between electronic and magnetic dynamics for different probing energies remains to be done. In this communication, I will first present the electronic and magnetic dynamics of a Ni/Cu/Fe tri-layer measured by time resolved X-ray absorption spectroscopy and X-ray magnetic circular dichroism, showing a clear delay between the magnetic and the electronic response (Figure 1). Then I will present the electronic and magnetic dynamics of simple ferromagnetic layer for different probing energies, to bring new information about this delayed onset as well as the interplay between the electronic and magnetic spectra dynamics.



C. Stamm, et al., Nat. Mater., 6, 740 (2007)
 K. Carva, et al., Europhys. Lett., 86, 57002 (2009)

Figure 1 : a Layer structure of the investigated trilayer film; **b**, Relative XUV absorption $\Delta XAS/XAS_0$ showing a charge dynamic (solid symbols, left panel), and the $\Delta XMCD/XMCD_0$ signal that represents the spin dynamics (open symbols, right panel) for probing energies of 65.8 eV (Ni M edge, red and pink curves) and 55.7 eV (Fe M edge, blue curves); **c** zoom of Figure **b** where the grey area represents the 100 fs onset difference between charge and spin signal. Note that the $\Delta XAS/XAS_0$ curve for Ni has been scaled down for better comparability.

[3] B. Rösner, et al., arXiv:2005.10611[4] K. Yao, et al., arXiv 2005.02872 (2020)

4306 Terahertz Spin Seebeck Effect in Antiferromagnets

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*The author has chosen not to make public additional content

Ultrafast all-optical Switching of Magnetization in Mn₂RuxGa

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Ultrafast control of Dzyaloshinskii-Moria interaction and magnon excitation in metallic Rashba antiferromagnets

Alireza Qaiumzadeh QuSpin, NTNU

In this talk, I propose a novel way for ultrafast excitation of magnons in antiferromagnets metallic thin films by optical manipulation of Dzyaloshinskii-Moriya (DM) interaction.

In antiferromagnetic thin films, broken inversion symmetry or coupling to adjacent heavy metals can induce DM interactions. Knowledge of the DM parameters is essential for understanding, designing and control of exotic spin structures [1]. We introduce a framework for computing the nonequilibrium DM interaction in metallic antiferromagnet thin films by the electric field of ultrafast laser pulses [2]. We use this formalism to calculate laser-induced renormalization of DM interaction in a 2D Rashba antiferromagnet as a prototype toy model for metallic antiferromagnets with spin-orbit interactions. This formalism, in principle, might be implemented in ab initio calculatenate nonequilibrium DM interaction for realistic band structures.

[1] A. Qaiumzadeh, et al., Phys. Rev. Lett., 120, 197202 (2018)

[2] S. Ø. Hanslin, A. Brataas, A. Qaiumzadeh, to be submitted (2020)

A real-space tight-binding approach to model ultrafast spin dynamics in heterostructures

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In the process of laser-induced ultrafast demagnetization hot spin-polarized electrons are excited. In a system with a magnetic/nonmagnetic interface they generate a spin current crossing this interface. Various origins of this spin current generation are discussed in literature, for example the spin-dependent Seebeck effect [1] or energy- and spin-dependent electron transmittance of the interface [2].

To better understand the underlying microscopic processes and the role of the interface we implemented a realspace tight-binding model. We follow the temporal evolution of occupation numbers by solving the corresponding equations of motion. We include the excitation by a laser pulse and the coupling with phonons. Currents, defined as intersite occupation flow, provide access to charge and spin currents. Our results for Co/Cu heterostructures reveal the details of demagnetisation process at interfaces.

[1] Seifert et al., J. Phys. D: Appl. Phys., 51 364003 (2018)

[2] Alekhin et al., J. Phys.: Condens. Matter 31 124002 (2019)

Ultrafast Control of Spin Density waves and Periodic Lattice Displacements in Chromium

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Experimental advances in ultrafast physics have allowed to monitor structural and electronic processes and even phase transitions on their natural timescales. Here we model recent experiments on ultrafast control of spin density wave phase in elemental Chromium with a sequence of optical pulses. The strain wave and CDW, induced by the spin density modulation via exchange striction, are monitored using X-ray diffraction. Results show order parameter oscillations and a partial melting of the SDW in response to optical pulses. Interestingly, depending on the exact delay between two sequential optical pulses, one can increase or decrease the oscillation amplitude, allowing for optimal control. We use Landau theory and heat transfer equations to describe the dynamics of the interacting charge and spin density waves. All details of the experiment are replicated to a high degree by the model.

Picosecond magnetization dynamics of spin modes revealed by diffractive ferromagnetic resonance

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The understanding and ability to manipulate collective spin excitation modes in topologically ordered magnetic structures offers great potential for the development of novel spintronic devices. At present, understanding of these dynamic magnetization modes is based on ferromagnetic resonance (FMR) studies, probing the bulk of the material system, which is analysed alongside micromagnetic models. Experimental tools to directly probe complex magnetisation dynamics in such spin systems are lacking.

Our novel diffractive ferromagnetic resonance (DFMR) technique combines the power of two measurement techniques. Firstly, resonant elastic X-ray scattering (REXS) provides sensitivity to the detailed periodic spin structure within a magnetic system [1]. In combination with X-ray detected ferromagnetic resonance (XFMR) the technique is also sensitive to time-resolved and element-selective magnetization dynamics [2,3].

Here we demonstrate the capabilities of DFMR measurements and reveal the complex magnetisation dynamics in Y-type hexaferrite [4,5]. Two dynamic modes are observed and coupling of the dynamic processional cones onto the chiral spin structure is characterised.

- [1] G. van der Laan. C. R. Phys., 9, 570 (2008)
- [2] G. van der Laan, J. of Electron Spectrosc. Relat. Phenom., 220, 137 (2017)
- [3] G. Boero et.al. New J. Phys., 10, 013011 (2008)
- [4] F.P. Chmiel et.al. Phys. Rev. B, 100, 104411 (2019)
- [5] D.M. Burn et.al. Nano Lett., 20, 345 (2020)

Control of THz emission from artificial antiferromagnetic spin emitters

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Artificial antiferromagnets have a long tradition in the study of spin transport in magnetic materials. Here we use multilayer spin emitters prepared by oblique incidence deposition (OID) [1] that sets a uniaxial anisotropy in the magnetic materials such that the magnetic static interlayer coupling is not needed to stabilize the antiferromagnetic state. The ferromagnetic and antiferromagnetic states can be prepared without a stabilizing external field. Thus, in single layers OID spin emitters the external magnetic field is obsolete. In multilayers the antiferromagnetic alignment can be set without any magnetic interlayer coupling by tuning the anisotropy in each layer. We study the THz emission in both states and find a strong increase in the antiferromagnetic state.

The MOKE hysteresis curve of the W/Fe/Pt/Fe/W multilayer stack reveals antiferromagnetic and ferromagnetic states. The spin emitters are excited via ~60 fs long near-infrared pulses. The electric field of the single-cycle THz pulses emitted was measured via electro-optical sampling and corresponds to a spectrum centered at 1.5 THz, extending up to 3 THz.

In the antiferromagnetic state, the electron currents in the non-magnetic layers are all in the same direction, inducing an increase of the THz radiation. In the ferromagnetic state, the electron currents ideally cancel each other, cancelling the THz emission.

The OID technique applied to the engineering of multilayers of spin emitters enables new degrees of freedom to control the THz waveform, improve the conversion efficiency from input laser power to THz radiation.



Figure 1 : a The principles of the normal and oblique incidence deposition techniques, **b** the magnetization direction of the FM layer (black arrow) and the electron currents in the multilayer sample (gelb), **c** the increase of one order of magnitude of the measured THz electric field in the antiferromagnetic over the ferromagnetic states.

[1] K. Schlage, L. et al., Adv. Func. Mater., 26 7423 (2016)

Analysis of microscopic origin of magnetooptical Kerr effect in bcc Fe

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We present analysis of microscopic origin of magnetooptic spectra from the electronic structure of bcc Fe. The magnetooptic (MO) permittivity spectra are obtained by the WIEN2k code and compared with the corresponding experimental MO Kerr effect spectra. The ab-initio spectra are given by Kubo formula, i.e. as a result of summation over all pairs of electronic bands and integration over the Brillouin zone that all sum up to single MO spectra. We investigate what features of electronic structure contribute to outgoing final MO spectra. We have also developed several novel ways of visualizing MO-related phenomena in the Brillouin zone.

It turns out that strong MO signal comes from several isolated k-points in the Brillouin zone where the band degeneracy is avoided by the spin-orbit interaction. There are two major types of MO contributions determined by the nature of the avoided degeneracy (given by topology of band approaching), that contribute differently to the total MO spectra. Both types are visualized and their contributions to the total spectra are demonstrated in local MO spectra.

Chiral versus collinear magnetic order dynamics: faster chiral recovery after optical excitation revealed by femtosecond XUV scattering

Nico Kerber

Johannes Gutenberg University of Mainz

While chiral spin structures stabilized by Dzyaloshinskii-Moriya interaction (DMI) are candidates as novel information carriers, their dynamics on the fs-ps timescale is little known. Since with the bulk Heisenberg exchange and the interfacial DMI two distinct exchange mechanisms are at play, the ultrafast dynamics of the chiral order needs to be ascertained and compared to the dynamics of the conventional collinear order. Using an XUV free-electron laser we determine the fs-ps temporal evolution of the chiral order in domain walls in a magnetic thin film sample by an IR pump - X-ray magnetic scattering probe experiment. Upon demagnetisation we observe that the dichroic (CL-CR) signal from the chiral order in the domain walls recovers significantly faster than the (CL+CR) sum signal from the average collinear domain magnetisation. We explore possible explanations based on spin structure dynamics and reduced transversal magnetisation fluctuations inside the domain walls and find that the latter can explain the experimental data leading to distinctly different dynamics for collinear magnetic order and chiral magnetic order.

Terahertz spin dynamics in antiferromagnetic Mn₂Au

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In antiferromagnets, the intrinsic terahertz (THz) magnon resonances [1] are expected to enable pathways to highspeed spin information processing. In certain non-centrosymmetric antiferromagnetic materials such as CuMnAs and Mn₂Au, switching of the antiferromagnetic Néel vector has recently been demonstrated by using pulsed electrical currents in both materials [2-4] and by free-space THz pulses in the case of CuMnAs [3]. The switching was attributed to the so-called Néel spin-orbit torque (NSOT) [5], which is proportional to the current inside the sample. However, the mechanisms and governing timescales of such magnetic-order control are still debated. It is, therefore, important to understand the spin dynamics in these materials on ultrafast time scales.

Here, we investigate spin dynamics in the perturbative regime of Mn₂Au thin films. The direction of the Néel vector was prealigned via application of a pulsed magnetic field that exceeded the spin-flop transition field (60 T) [6]. A THz single-cycle pulse is used to trigger dynamics, whose evolution is monitored by magneto-optical probing with femtosecond laser pulses. Our measurement scheme allows us to separate effects which are odd and even with respect to the driving THz field. Figure 1 shows the component of the magnet-optical signal (yellow curve) which is odd with respect to the driving THz field (blue dotted curve). Its amplitude grows linearly with the THz field strength and thus can arise from either the antiferromagnetic Zeeman torque (time derivative of the magnetic field) or the Néel spin-orbit torque (electric field) [7]. A symmetry analysis of the signal along with measurements as a function of sample orientation are found to be consistent with a magnetic signal origin. By analyzing the dependence on the incident probe polarization, magnetic linear birefringence is identified as the probe mechanism [8]. We model the spin response upon THz excitation as a damped sinusoidal oscillation (red curve) convoluted with the THz electric field. A fit (purple curve) to the measured signal (yellow) yields very good agreement, a resonance frequency of about 0.6 THz and very strong damping with a time constant of 0.62 ps. The resonance frequency approximately agrees with that of a recently predicted NSOT-driven in-plane precession of the Néel vector [7]. In conclusion, we observe THz spin dynamics in Mn₂Au whose characteristics are in good agreement with those of a predicted NSOTdriven antiferromagnetic mode. The observed THz frequency response of the spins suggests that NSOT allows ultrafast control over magnetic order of Mn₂Au.



Figure 1: Polarization rotation of the probe pulse (yellow curve) versus delay since arrival of the driving terahertz pulse (dotted blue curve). The purple curve is a fit that is given by the convolution of the pump field with a strongly damped harmonic oscillation (orange curve).

- [1] T. Kampfrath et al., Nat. Photonics 5, 31 (2011)
- [2] P. Wadley et al., Science 351, 587-590 (2016)
- [3] K. Olejnik et al., Sci. Adv. 4, eaar3566 (2018)
- [4] S. Yu. Bodnar et al., Nat. Comm. 9, 348 (2018)
- [5] J. Železný et al, Phys. Rev. Lett. 113, 157201 (2014)
- [6] A. Sapohnik at al., Phys. Rev. B 97, 134429 (2018)
- [7] O. Gomonay, at al. Phys. Rev. B 98, 104430 (2018)
- [8] V. Saidl et al., Nat. Photon. 11, 91-96 (2017)

Effect of Optical Nanoantennas on Ultrafast Magnetization Dynamics in TbCo

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The increasing amount of digital information generated globally has created the requirement for a storage technology that is fast, dense and energy efficient. One phenomenon that can potentially be applied for this purpose is All-Optical Switching (AOS) of magnetization [1], wherein the magnetization of a material can be reversed following excitation by an ultrashort laser pulse. AOS has been observed in several classes of materials such as rare-earth transition metal (RE-TM) alloys and multilayers, and ferromagnets and even dielectrics [2, 3], and is very promising with regard to writing speed and energy efficiency [3]. However, the bit size attainable is constrained by the diffraction limit to be of the order of micrometers. This is much larger than the bit size attainable by current technologies, like HAMR [4].

The use of plasmonic nanostructures can help to reduce the size of the switched spot to the nanometer scale due to the collective electronic oscillations called localized surface plasmon resonances that can be excited in these structures by incident light at a characteristic resonance wavelength [5]. The enhancement of the incident electric field at resonance could also result in more energy efficient switching of magnetization. Such a system could also help us to investigate AOS at the nanoscale.

Here we study the effect of plasmonic antennas on the demagnetization dynamics of the RE-TM alloy TbCo in a hybrid magneto-plasmonic system, with the plasmonic elements either as nanocones or nanorings, using an all optical pump-probe technique. The system is excited with the pump wavelength at and away from resonance, and the resulting dynamics is compared for the two cases. The field enhancement in the near field of the nanostructures at resonance appears to lead to increased demagnetization of TbCo for the case of resonant excitation as compared to the off-resonant excitation.

- [1] C. D. Stanciu et al., Phys. Rev. Lett., 99, 1 (2007)
- [2] S. Mangin et al., Nat. Mater., 13, 286 (2014)
- [3] A. Stupakiewicz, et al., Nature, 542, 71 (2017)
- [4] W. A. Challener et al., Nat. Photonics, 3, 220 (2009)
- [5] Tian-Min Liu et al., Nano Letters, 15, 6862 (2015)

Strong impact of the interface on ultrafast spin-to-charge conversion in metallic bilayers

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The efficient conversion of spin to charge transport by spin-orbit interaction (SOI) is of great relevance for the detection and generation of spin currents in future spin-based electronics [1] and the development of efficient emitters of terahertz electromagnetic pulses [2,3]. Thus, understanding and optimization of spin-to-charge conversion (S2C) is highly desirable. Current research has started taking the role of interfaces into consideration. For example, recent works demonstrated ultrafast S2C by the inverse Rashba-Edelstein effect at interfaces of metallic heterostructures [4,5]. Here, we analyze S2C in an important model system: F|N bilayers consisting of ferromagnetic (F) and nonmagnetic (N) metal thin films. To measure S2C, we employ femtosecond optical pulses to trigger ultrafast spin transport from the F into the N layer (see figure 1) [2-5]. Through S2C, the spin current is partially converted into a transverse charge current that is monitored by detecting the concomitantly emitted THz electromagnetic radiation. To simplify the separation of interfacial S2C, we minimize S2C in the N bulk by using the common low-SOI materials AI and Cu. To exclude a dominant S2C in the F bulk, we compare several ferromagnetic materials. Our measurements indicate that S2C at Co|Al and Py|Cu interfaces can, respectively, become comparable to the overall S2C in Co|Pt and Py|Pt reference bilayers. Furthermore, we show that the sign of S2C is drastically affected by modification of the interface by changing the sequence of F and N layers or by dusting the interface region with oxygen atoms. We discuss our results in terms of a possible extrinsic S2C mechanism: skew scattering of spin-polarized electrons at interfacial imperfections. This view is consistently supported by model calculations of the skew-scattering strength of Al(F) and Cu(F) interface layers. We suggest that S2C is further enhanced by the relatively long relaxation length ($\lambda_N \sim 4$ nm) of the ballistically propagating electrons in the Cu layer (see figure 1). This remarkable nonlocal scenario is a promising route to enhance ultrafast S2C.

- [1] F. Hellman et al., Rev. Mod. Phys., 89, 025006 (2017)
- [2] T. Kampfrath et al., Nature Nanotech., 8, 256 (2013)
- [3] T. Seifert et al., Nature Phot., 10, 483 (2016)
- [4] M.B. Jungfleisch et al., Phys. Rev. Lett., 120, 207207 (2018)
- [5] C. Zhou et al., Phys. Rev. Lett., 121, 086801 (2018)

Accelerating the laser-induced demagnetization of a ferromagnetic film by antiferromagnetic order in an adjacent layer

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Magnetic recording and storage media will face major challenges in the near future regarding energy efficiency and data transfer speed due to increased demand and continuing growth of data storage volume. Much could be gained by switching from field-induced manipulation of magnetic order, where speed is limited by spin precession dynamics and energy is limited by the impedance of coils, to manipulation by ultra short light pulses.

We study the ultrafast demagnetization of Ni/NiMn and Co/NiMn ferromagnetic/antiferromagnetic bilayer systems after excitation by a laser pulse using magnetic circular dichroism in time-resolved pump--probe resonant X-ray reflectivity. Tuning the sample temperature relative to the ordering temperature of the antiferromagnetic NiMn layer allows to isolate effects induced by the magnetic order of the latter. In the Co/NiMn system, the presence of antiferromagnetic order in NiMn speeds up the demagnetization of the ferromagnetic Co layer, which is attributed to bidirectional laser-induced superdiffusive spin currents between the ferromagnetic and the antiferromagnetic layer.

Influence of transport on the time-resolved MOKE signal during ultrafast demagnetization

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Ultrafast demagnetization of itinerant ferromagnets is mediated by a variety of local and non-local processes such as spin-flip scattering and optically induced spin transport that both reduce the spin polarization in the probed area [1,2]. This microscopic picture is based on a number of theoretical and experimental investigations over the past two decades.

The majority of experimental studies so far followed the optically induced demagnetization dynamics in magnetic materials using the time-dependent magneto-optical Kerr response of the magnetic and non-magnetic materials. Using thin magnetic films with film thickness smaller than penetration depth of the laser light leads to a homogeneous optical excitation of the material which is reflected in a single Kerr response of the material.

In contrast, the situation becomes much more complicated when using materials with larger film thickness where the optically excited volume of the materials is no longer homogeneously heated by the laser pulse. The heat as well as the spin-dependent charge transport will affect the depth dependent magnetization dynamics of the material and, hence, the time-resolved MOKE signal. In particular, optically generated spin currents will propagate through the magnetic materials and will hence contribute to the magnetization dynamics at different depths in the material at different timescales.

In this contribution, we theoretically and experimentally study the role of transport in the time-resolved MOKE signal of a 100nm thick Ni film. Starting with simulation based on the Thermodynamic μ T-model [3] we determine the spatial and temporal evolution of magnetization.

We take into account that the MOKE signal of such a 100nm magnetic film is determined by the sum of the Kerr responses from different material depths in the complex Kerr plane [4]. By integrating the depth sensitivity function [5] weighted by spatially resolved magnetization at various time instances we obtain the changes in the MOKE signal on a femtosecond timescale. To demonstrate the capability of this approach, we compare our theoretical predictions with the changes in Kerr-rotation and Kerr-ellipticity measured using the Complex-MOKE technique [6,7].

- [1] B. Y. Mueller et al., Phys. Rev. Lett., 111, 167204 (2013)
- [2] S. Kaltenborn, Y-H Zhu and H. C. Schneider, Phys. Rev. B, 85, 235101 (2012)
- [3] B. Y. Mueller and B. Rethfeld, Phys. Rev. B, 90, 144420 (2014)
- [4] G. Traeger, L. Wenzel and A. Hubert, Phys. Status Solidi A, 131, 1 (1992)
- [5] W. Kuch, R. Schaefer, P. Fischer and F. Hillebrecht, ISBN: 978-3-662-44532-7 (2015)
- [6] M. Hofherr et al., Phys. Rev. B., 96, 100403 (2017)
- [7] Y. Liu et al., J. Magn. Magn. Mat., 502, 166477 (2020)

Multiscale dynamics at the antiferromagnetic-ferromagnetic phase transition in FeRh

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Using a double-pump pulse approach and laser-induced THz emission as an ultrafast probe of the magnetization, we show that a femtosecond laser pulse generates ferromagnetic nuclei in a FeRh/Pt bilayer, *i.e.* these nuclei acquire a net magnetization and a susceptibility to a magnetic field, but only 20 ps after the initial laser excitation. We argue that this latency is intrinsic to the phase transitions from antiferromagnetic to ferromagnetic states and must be present even in the case when the sign of the exchange interaction changes instantaneously.

Photon energy dependent fs-demagnetization dynamics of thin Nickel films

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In a laser-induced fs demagnetization process, it is expected that the specific optical transitions in the band structure of magnetic materials can essentially affect the overall dynamics of the optically induced loss of spin order. However, almost all experimental fs-demagnetization studies so far employed fs light pulses of 1.55 eV to trigger the magnetization-dynamics. Hence, the role of the photon energy of the exciting light pulse has been largely neglected so far. We have implemented an all-optical time-resolved MOKE setup with variable pump photon energy in the range of 1.55 to 3.10 eV. As prototypical system, we investigated the ultrafast demagnetization dynamics of thin Ni films on insulating and conducting substrates. The characteristic parameters of the demagnetization process, i.e., the demagnetization time for various excitation photon energies are compared with simulations based on the density of states of Nickel and interface effects to describe the non-equilibrium dynamics of the spin-carrying excited electrons. For metallic substrates, we further discuss the magnitude and timescale of the optically injected spin carriers across the magnetic/non-magnetic interface.

M. Hofherr, et al., Physical Review B 96, 100403 (2017)
U. Bierbrauer, et al, J. Phys.: Condens.Matter 29, 244002 (2017)
B.Y. Mueller and B. Rethfeld, Phys. Rev. B 90, 144420 (2014)

Direct experimental evidence of inertial dynamics in ferromagnetic thin films

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It is known that the dynamics of magnetisation described by the Landau-Lifshits-Gilbert (LLG) equation correctly models the reversal of a magnetic bit and hence the writing of digital information at nanosecond rates. Recently, the LLG equation was reformulated by including a physically correct inertial response, which predicts the appearance of spin nutations at a frequency much higher (in the terahertz range, i.e. at sub-picosecond timescales) than the spin precession described by the conventional LLG equation, typically at gigahertz frequencies. In this presentation we show the first direct experimental evidence of inertial spin dynamics in ferromagnetic thin films in the sub terahertz frequency regime. We observed that when a ferromagnetically ordered spin system is driven by intense narrowband terahertz magnetic field pulses of tunable centre frequency, its behavior is similar to the one of a forced oscillator. By analyzing both amplitude and phase of the response, probed with femtosecond magneto-optical Kerr effect, we detect a broad resonance at approximately 0.6 THz, which we ascribe to nutation spin dynamics being driven by the terahertz magnetic field. From the experimental data, we were able to extract the angular momentum relaxation time, which we found to be on the order of 10 ps for three distinct samples with different Gilbert damping constant. Our experimental results are in good agreement with the numerical simulation of LLG equation with the inertial term. This work deepens our understanding of ultrafast magnetism and paves a new way towards controlling magnetism exploiting the nutation resonance.

Monte Carlo simulation of ultrafast nonequilibrium spin and charge transport in iron

Johan Briones, Hans Christian Schneider, Baerbel Rethfeld TU Kaiserslautern

Ultrashort laser pulses in magnetic materials and ferromagnet-metal heterostructures drive complicated dynamics that may result in ultrafast demagnetization as well as ballistic to diffusive transport of highly excited electrons [1-3].

We focus here exclusively on the spin-dependent transport of electrons excited to states far above the Fermi energy. We have adapted the asymptotic-trajectory Monte Carlo technique [4] to trace the spatio-temporal dynamics of electrons excited by an ultrashort laser pulse in ferromagnetic metals, and apply our approach to the case of iron. In particular, we resolve the change of electronic energy induced by various scattering processes. Our model takes into account electron-ion scattering, electron impact ionization and, in a phenomenological way, the contribution of spin flips.

This approach directly yields a space-time picture of electron cascade processes and allows us analyze the influence of secondary-electron generation on the transport characteristics of high-energy electrons in the linear excitation regime. The Figure shows as an example the time evolution of the parameter that characterizes the particle transport [2,3]. The hot electron dynamics is initially ballistic (= 2), then superdiffusive (> 1) and finally diffusive (= 1). The secondary electrons keep the system noticeably longer in the superdiffusive regime.



Figure 1

[1] D. Rudolf, et al, Nat. Commun., **3**, 1037 (2012)

[2] M. Battiato, K. Carva, P. M. Oppeneer, Phys. Rev. Lett., 105, 027203 (2010)

[3] D. M. Nenno, B. Rethfeld, H. C. Schneider, Phys. Rev. B, 98, 224416 (2018)

[4] K. Huthmacher et al., Physica A, 429, 241 (2015)

Ultrafast spin momentum transfer in noncollinear spin valves

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Ultrafast demagnetization of metallic thin films induced by a femtosecond laser pulse is a well-known phenomenon studied since 1996 [1]. One of the key mechanisms which contributes to such phenomena is superdiffusive spin-dependent transport [2] of hot electrons excited by the pump laser from the localized *d* band to the *sp* one above the Fermi level. It has been shown that superdiffusive spin-dependent transport can sufficiently explain ultrafast demagnetization in metallic thin films and multilayer with collinear magnetizations. Moreover, the excitation of hot electrons in magnetic materials can lead to generation of spin current inducing spin transfer torque [3]. As a result one can observe terahertz magnetization dynamics in noncollinear magnetic multilayers [4] as well as ultrafast domain wall motion [5].

Here, we generalize the model of spin-dependent transport for noncollinear magnetic multilayers with arbitrary magnetic configurations. In our calculations we focus on Al(3nm)/Ni(5nm)/Ru(2nm)/Fe(4nm)/Ru(5nm) multilayer, with Ni and Fe magnetic layers separated by thin nonmagnetic Ru. While the spin-dependent transport inside the layers is treated via nonlocal equations of motion for the electron densities, transport through the interfaces between the layers is described by energy-dependent reflections and transmissions calculated *ab initio* using the wave function matching method [6,7]. Similarly, the nonmagnetic Ru spacer is treated as an effective interface between Ni and Fe layers described by its energy and spin-dependent reflections and transmissions.

The model allows us to calculate position and time resolved spin densities as well as spin currents. From the transverse spin current density we calculate the time dependent spin transfer torque acting on the Ni and Fe magnetizations. A substantial effect of spin filtering and of non-collinear magnetic configuration on ultrafast demagnetization and spin transfer torque is predicted for the studied spin valve.

[1] K. Carva, et al., Laser-Induced Ultrafast Magnetic Phenomena, Handbook of Magnetic Materials, Elsevier (2017)

[2] M. Battiato, K. Carva, P. M. Oppeneer, Phys. Rev. Lett., 105, 027203 (2010)

[3] P. Baláž, M. Žonda, K. Carva, P. Maldonado, P. M. Oppeneer, J. Phys.: Cond. Matter, 30, 115801 (2018)

[4] U. Ritzmann, P. Baláž, P. Maldonado, K. Carva, P. M. Oppeneer, Phys. Rev. B, 101, 174427 (2020)

[5] P. Baláž, K. Carva, U. Ritzmann, P. Maldonado, P. M. Oppeneer, Phys. Rev., B 101, 174418 (2020)

[6] K. Xia, M. Zwierzycki, M. Talanana, P. J. Kelly, G. E. W. Bauer, Phys. Rev. B, 73, 064420 (2006)

[7] M. Zwierzycki, et al., phys. stat. sol. B, 245, 623 (2008)

Features of laser-induced magnetization precession in ferromagnetic films with timedependent magnetic anisotropy

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One of the promising directions of modern magnetism is the study of laser-induced precession of magnetization and propagating spin waves for implementation in spintronics and magnonic devices.

Recent works [1,2] demonstrate efficient all-optical excitation of magnetization dynamics (precession and spin waves) due to laser-induced ultrafast changes of the anisotropy field. Owing to ultrafast laser-induced heating, a local change in the material parameters (anisotropy and magnetization) occurs, which leads to a reorientation of the effective magnetic field in the sample and thus triggers the magnetization dynamics [3].

When describing thermal excitation of the laser-induced precession, an instantaneous step-like change in the anisotropy field, regardless of the nature of the anisotropy (magnetocrystalline, growth, etc), while the further evolution is often of this field is often neglected. This allows one to simplify the mathematical description of the experiment data. The approach is substantiated by the relatively long cooling time of the excited material as compared to short precession life-times. It was experimentally demonstrated for metal films during pulsed laser heating [2], that anisotropy relaxation to its equilibrium value is observed at times of the order of hundreds of picoseconds. Therefore, in the case of long life-times of the excited precession it becomes important to account for the temporal evolution of the anisotropy field after excitation, since it may yield a number of important physical phenomena including magnetization switching [4].

In this work, we pave the way to deriving the temporal evolution of the anisotropy parameters from the combined experiments on laser-induced ultrafast demagnetization, magnetization precession and time-dependent hysteresis loops. Epitaxial films of Galfenol with the composition Fe_{0.81}Ga_{0.19} and with thicknesses of 5 and 10 nm on (001)-GaAs substrates were used as the films demonstrating long precession life-times [5]. The films possess strong magnetocrystalline cubic anisotropy with an additional growth-induced anisotropy easy axis in the film plane. In experiments, the parameters of laser-induced precession of magnetization were obtained from the time-resolved polar magneto-optical Kerr effect measurements (pTRMOKE) obtained for various directions of the external magnetic field in the film plane. The evolution of magnetization and effective anisotropy fields, in turn, were obtained from longitudinal TRMOKE. A theoretical description of the precession parameters was obtained using the Smit-Beljers approach for the case of instantaneous changes in material parameters.

As a result, evolution of the anisotropy parameters over the temporal range of 0-3 ns after ultrafast heating were obtained. It is shown that evolution leads to a change in the precession frequency with time, which is seen as a nonsymmetrical shape of the frequency spectrum of the observed precession.

[1] N. E. Khokhlov et al., Phys. Rev. Appl., 12, 044044 (2019)

[2] E. Carpene et al., Phys. Rev. B, 81, 060415 (2010)

[3] A. Kirilyuk et al., Rev. Mod. Phys., 82, 2731 (2010)

[4] E. Carpene et al., Phys. Rev. B, 84, 134425 (2011)

[5] A. V. Scherbakov et al., Phys. Rev. Appl., 11, 031003 (2019)

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Time-resolved magnetooptical study of the magnetic inhomogeneity in epitaxial palladium-iron alloy films

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Palladium-iron $Pd_{1-x}Fe_x$ alloys with cubic fcc crystal structure at low Fe-content (x < 0.10) are soft low-temperature ferromagnets promising for fast and energy-efficient superconductor (S) / ferromagnet (F) heterostructure-based superconducting spintronic devices [1], both logical elements and memory cells. Iron content x in the alloy determines its Curie temperature, magnetization as well as the magnetic anisotropy [2]. F-layer in S/F/S heterostructures serves as a phase shifter of the superconducting wavefunction between two S-layers, with the phase shift value related to the magnetization and layer thickness [3]. For this reason, one of the key features of the F-layer is its magnetic homogeneity, as inhomogeneous magnetization over the contact area destroys the coherence and smears out Josephson effect associated with it.

One should note that palladium-rich $Pd_{1-x}Fe_x$ alloys are in fact solid solutions and therefore are intrinsically inhomogeneous systems. Magnetic inhomogeneity in these materials had been studied in the bulk crystalline samples by neutron diffraction [4]. This method however is not suitable for ultrathin weakly ferromagnetic films, and complementary approaches are required.

Here we present the results of a systematic study of a series of 20-nm epitaxial high-quality $Pd_{1-x}Fe_x$ thin films (x = 0; 0.012; 0.034; 0.062 and 0.080) on MgO (001) substrate with ultrafast optical and magnetooptical spectroscopy methods. Details on the film synthesis can be found elsewhere [2]. Chemical composition, crystalline structure and magnetic properties of all the samples have been thoroughly investigated before [5].

For studies of both the reflectivity transients and time-resolved magnetooptical Kerr effect a sensitive pump-probe setup was used. In the reflectivity transients of the Pd film the relaxation after an instantaneous (with our temporal resolution) rise can well be described by a sum of the fast (~0.24 ps) and the slow (~400 ps) components. No any qualitative or quantitative modification of the transient took place on cooling the Pd film. Results for the Pd_{0.94}Fe_{0.06} film are similar in the temperature range above the Curie temperature ($T_c \sim 190$ K), only the lifetime of the slow component is somewhat shorter, ~240 ps. On further cooling below T_c the transient experiences a drastic, both quantitative and qualitative, evolution. Namely, while the fast relaxation component is left essentially untouched, the amplitude of the slow component decreases by more than 80%, and its lifetime shortens down to ~40 ps below 100 K (Fig. 1(b)). Moreover, additional relaxation components of an opposite sign develop on cooling: an intermediate with the lifetime of ~0.8 ps and a very long one ~1 ns.

We will argue basing on the data on a whole series of our samples that the complex multi-component character of the reflectivity and TR-MOKE transients originates from the magnetic inhomogeneity of the films. Nature of the developing and vanishing components will be discussed.

[1] V.V. Ryazanov, V.V. Bol'ginov, D.S. Sobanin et al., Phys. Procedia, 36, 35 (2012)

[2] A. Esmaeili, I.V. Yanilkin, A.I. Gumarov et al., Thin Solid Films, 669, 338 (2019).

[3] I.V. Vernik, V.V. Bol'ginov, S.V. Bakurskiy et al., IEEE Trans. Appl. Supercond., 23, 1701208 (2013)

[4] G.G. Low and T.M. Holden, Proc. Phys. Soc. 89, 119 (1966)

[5] A. Esmaeili, I.V. Yanilkin, A.I. Gumarov et al., https://arxiv.org/pdf/1912.04852

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4309 Probing THz emission from Fe/Ta-multilayers

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The THz emission from metallic spintronic ferromagnetic (FM) and non-magnetic (NM) bilayers and multilayers has opened new routes in the direction of ultrafast spintronics. It combines the field of THz optics with the field of spintronics since the creation of the THz pulse is based on the generation of spin currents. The spin current, excited by a fs laser pulse in the FM layer, diffuses into the NM metal through a super diffusive process. Spin-orbit coupling transforms the pure spin current into a transient charge current due to the inverse spin Hall effect (ISHE). The transformations' proportionality constant is the spin Hall angle, a material parameter. The accelerated electrons of the transient current emit radiation in the THz range.

Initially, Fe/Pt bilayers were optimized with respect to layer thickness, geometrical arrangement and the crystal structure [1,2]. Recently we investigated the influence of the multilayer structure on the emitted radiation, concentrating on Ta as an NM material. The spin Hall angle of Ta is opposite to the spin Hall angle of Pt which leads to interesting possibilities in the emitters designs: Namely we investigated bilayers where Ta replaced Pt and multilayers where we combined Ta and Pt.

[1] G. Torosyan et al., Scientific Reports, 8, 1311 (2018)

[2] D. Nenno et al., Scientific Reports, 9, 13348 (2019)

Symposium 6. Spin-based Quantum Technologies

Quantum Microscopy of Antiferromagnetic Domains Walls

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Diamond nanoprobes with single engineered NV centers allow enhancing scanning probe microscopy with quantum metrology. In this talk, I will introduce the basic concepts and technology of diamond nanoprobes, and give illustrative examples of imaging applications to antiferromagnetic domains and domain walls in Cr_2O_3 [1] and CuMnAs [2].

M. S. Wörnle, P. Welter, M. Giraldo, T. Lottermoser, M. Fiebig, P. Gambardella, and C. L. Degen arXiv:2009.09015 (2020).
 M. S. Wörnle, P. Welter, Z. Kašpar, K. Olejník, V. Novák, R. P. Campion, P. Wadley, T. Jungwirth, C. L. Degen, and P. Gambardella, arXiv:1912.05287 (2019).

Oral Presentation

3708

Spintronic harvesting of thermal fluctuations on paramagnetic molecular centers

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In order to secure sustainable energy sources, several measures aim to harvest energy, in our environment, from natural (solar, wind, water) and human-made (WiFi, GSM, human motion, vibrations) sources. Recent experiments have explored the operation of nanoscale thermoelectric generators [1,2], while a few reports suggest that spintronics [3] can enhance the harvesting of thermal fluctuations on spin-split discrete electronic states [4-6] (www.spinengine.tech). We have grown stacks containing this spin engine's two key ingredients (see panel a): Fe/C_{60} interfaces with a high spin polarization as the spintronic selector [7], and CoPc molecules with a Co S=1/2 paramagnetic (PM) center [8,9] as the thermal fluctuator. To maintain the PM fluctuation against possible magnetic coupling [10] to the electrodes, the ultrathin CoPc layer is sandwiched between C_{60} spacer layers. Several nanoscale organic junctions [8] reveal, at constant bias voltage, a current whose sign depends on the device's magnetic state (panel b). This power generation, whose origin is thus clearly spintronic, reaches 370nW at 40K and 23nW at 360K, i.e. a 270x improvement over previous results [6] (panel c). Its temperature dependence indicates a T=120K transition between two activation energy regimes that could correspond to the magnetic exchange energy between PM centers [9]. The offset-corrected magnetoresistance reaches 770% at 40K, a record for molecular spintronics.



Figure 1: (a) Schematic of the spin engine's potential landscape, comprising spintronic selectors and a PM center as the thermal fluctuator [6]. Results on Fe/C₆₀/CoPc/C₆₀/Fe nanojunctions. (b) I(H) and (c) In(R, V_{O1t}, Max. P) vs 1/T.

[1] Thierschmann, H. et al. Nature Nanotechnology 10, 854–858 (2015).

[2] Strasberg, P., Schaller, G., Brandes, T. Esposito, M. Physical Review X 7, 1-33, (2017).

- [3] Kent, A. D. Worledge, D. C. Nature Nanotechnology 10, 187–191 (2015).
- [4] Hai, P. N., Ohya, S., Tanaka, M., Barnes, S. E. Maekawa, S. Nature 458, 489-492 (2009).
- [5] Miao, G.-X., Chang, J., Assaf, B. A., Heiman, D. Moodera, J. S. Nature Communications 5, 3682-3687, (2014).
- [6] Katcko, K. et al. Communications Physics 2, 116-123, (2019)
- [7] Djeghloul, F. et al. American Chemical Society 7, 13, 2310-2315 (2016)
- [8] Katcko, K. et al. arXiv:1910.10578.
- [9] Serri, M. et al. Nature Communications 5, 3079-3087, (2014).
- [10] Gruber, M. et al. Nature Materials 14, 981–984 (2015)

Storage and Retrieval of Microwave Pulses with Molecular Spin Ensembles

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Hybrid architectures combining complementary quantum systems will be largely used in quantum technologies and the integration of different components is one of the key issues. Here quantum memories are fundamental components and, to perform Storage/Retrieval operations, the physical system must fulfil two conditions: i) having a phase memory time long enough to allow the survival of the information during operation and ii) allow to the possibility of codifying a pulse sequence into the memory through a suitable protocol. Thanks to their long coherence times and the relatively easy manipulation by microwave pulses, electronic spin degrees of freedom provided by electron spin ensembles have been efficiently exploited into circuit quantum electrodynamics architectures based on microwave devices and resonators at low temperature. Here collective excitations of the spin ensemble are exploited to preserve and coherently exchange information within the different components of the architecture.

Molecular spins have recently emerged as a new class of quantum systems whose electronic and nuclear spin states and their relative quantum features (including g-factor, coherence time, atomic-clock transitions) can be extensively tailored synthetically. Different strategies for encoding quantum information with molecular ensembles or single molecules have been developed and experimentally proved. Moreover, the coherent coupling with microwave photons have been recently successfully achieved with transition metal-based oxovanadium(IV) complexes [1,2] as well as with organic radicals [2,3] embedded into planar resonant geometries, paving the way for the integration of molecular spin ensembles into microwave quantum architectures. Here however optimal experimental conditions and protocols (i.e. pulse sequences) to efficiently address the spins and to eventually correct intrinsic (related to the ensemble) or extrinsic factors (such as inhomogeneities of resonant geometry) that may limit the memory time still need to be found.

In this work we test single crystals and solid dispersions of diluted oxovanadium tetraphenyl porphyrin (VO(TPP)) as prototypical molecular spin ensembles for the Storage/Retrieval of microwave pulses when embedded into planar superconducting microwave resonators at low temperature (2K). We first measure the memory time by means of the Hahn-echo sequence and the Rabi Oscillations of the samples. We then test two Dynamical Decoupling sequences: the Carr-Purcell-Meiboom-Gill and the Uhrig Dynamical Decoupling. Both the sequences are found to enhance the memory time of the crystal sample up to three times after the application of a low number (3,4) of π pulses, reaching up to 3 μ s. We then successfully store and retrieve into the ensembles trains of up to 5 small pulses by using a single recovery π pulse and we show that individual control on such excitations can be achived. Our proof-of-principle results demonstrate the memory capabilities of molecular spin ensembles when embedded into quantum circuits [4].

- [1] Bonizzoni et al. Sci. Rep. 7, 13096 (2017)
- [2] Bonizzoni et al. Adv. on Phys. X 3, 1435305 (2018)
- [3] Ghirri et al. Phys. Rev. A 93, 063855 (2016)
- [4] C. Bonizzoni et al. NPJ Quantum Inf. in press (DOI: 10.1038/s41534-020-00296-9)

Magnetic and spin-transfer properties of amorphous and polycrystalline iron silicides thin films

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Over the last decades, iron silicides attract a lot of attention due to a variety of useful physical properties inherent to them. At the same time, the low cost and the high accessibility of iron and silicon make this group of materials a very promising candidate for the replacement of some rare, toxic, and expensive materials. For this reason, the new approaches of iron silicides thin films formation and characterization should be found.

We report the magnetic and spin-transfer properties of amorphous and polycrystalline iron silicide thin films, grown by pulsed laser deposition (PLD) and ion beam deposition (IBD). Samples with the thicknesses from 5 to 60 nm oxidized silicon substrates were investigated. To prevent oxidation of thinner films different capping layers were used. The growth pressure varied from 10^{-7} Pa for PLD to 10^{-2} Pa of argon for IBD. Stoichiometric Fe₃Si or pure iron and silicon targets were used to grow samples.

It was shown, that all amorphous (room temperature grown) samples show high in-plane anisotropy, which strongly depends on growth technique, formation parameters, and the type of sublayer and capping layer.

For polycrystalline samples, vacuum annealing temperatures varied from 150 to 550°C. Magnetic and structural properties depending on growth parameters, element ratio, and thickness were investigated. The investigations of spin-transfer properties by point-contact Andreev reflection for 50 nm thin films with near Fe₃Si compositions show a high level of spin polarization with the values up to 54%.

Majorana-Kondo interplay in T-shaped double quantum dots

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In this contribution, the electrical transport properties of a T-shaped double quantum dot side-coupled to a topological superconducting nanowire hosting Majorana zero-energy modes are discussed. The system is studied theoretically using the numerical renormalization group method. The spectral functions and linear response conductance are analyzed. In the absence of coupling with superconducting wire, the double dot system exhibits a two-stage Kondo effect, where a total decrease in conductance at low temperatures is observed. Coupling with Majorana wire has a significant impact on the second stage of the Kondo effect, both for different values of hopping t between two dots, as well as different strength of the coupling V_m between dot and wire. We show that the second stage screening is suppressed for one of the spin channels. It results in the low-temperature conductance equal $0.5 e^2/h$ for the case of long wire (there is no overlap of the Majorana modes, $\varepsilon_M = 0$). The finite Majorana wire case with $\varepsilon_M \neq 0$ is also discussed, where the quantum interference, as well as low-temperature conductance, are suppressed, restoring the two-stage Kondo effect.

Superconducting proximity effect in In/NbP junction

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Recently, an idea of introducing superconductivity in topological materials has been developed owing to the possibility of non-zero momentum Cooper pairing. In particular, introducing nonzero superconducting order parameter into topological materials by inducing superconductivity through the proximity effect enables to employ Weyl semimetals (WSMs) and conventional superconductors (S). Experimental studies of conductance across S-WSM junctions prepared in the conventional way, i.e. by deposition of In on NbP single crystal were performed. Single crystal of NbP, grown by chemical vapor transport method, was carefully characterized by XRD, EDX, SEM, ARPES techniques and by electron transport measurements. For the studies of interface transmission, the (001) surface of the crystal was covered by several 300 nm thick metallic layers of In. DC current-voltage characteristics and AC differential conductance through the interfaces as a function of the DC bias were investigated. Upon cooling of the device during which In becomes superconducting, the junction shows conductance increase, pointing out the Andreev reflection as a prevalent contribution to the subgap conductance and very high and narrow peak at zero bias was observed. The conductance at the peak reaches the bulk value indicating that almost whole contact area is transmitting and, additionally, a superconducting proximity phase is formed in the material. This as a result of In diffusion into NbP, where the metal atoms penetrate the surface barrier and form very transparent superconductor-Weyl semimetal contact inside. However, further diffusion occurring already at room temperature leads to degradation of the effect, so it is observed only in the pristine structures. Despite of this, our observation directly demonstrates possibility of inducing superconductivity in a type-I Weyl semimetal.

Symposium 7. Domain walls, Skyrmions and Spin-orbit Related Phenomena
Elliptical Bloch skyrmion chiral twins in an antiskyrmion system

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*The author has chosen not to make public additional content.

Spin-orbit magnetic state readout in scaled ferromagnetic/heavy metal nanostructures

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The efficient detection of a magnetic state at nanoscale dimensions is important for the development of spin logic devices. Magnetoresistance effects can be used to detect magnetic states, but they cannot produce sufficiently large signals when the dimensions of the devices are reduced to the nanoscale, nor do they generate an electromotive force that can be used to drive a circuit element for logic device applications.¹ In this work², we report a favorable scaling law for the detection of an in-plane magnetic state of a magnet by using the inverse spin Hall effect in cobalt–iron/platinum ($Co_{50}Fe_{50}/Pt$) nanostructure devices. By reducing the dimensions of the device, we obtain a large spin Hall signal of 0.3 Ω at room temperature and quantify an effective spin-to-charge conversion rate for the ferromagnetic/heavy metal nanostructure system. Finally, we predict that this spin-orbit detection of magnetic states could be used to drive spin logic circuits.

[1] S. Manipatruni, et al., Nature 565, 35–42 (2019).

[2] V.T. Pham*, I. Groen*, et al., arXiv:2002.10581 (2020) (accepted in Nature Electronics).

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Fast Chiral Domain Wall Motion in Magnetic Insulating Garnet Films

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Chiral exchange interactions manifest from broken spatial inversion symmetry. In metallic ferromagnet systems, engineered interfaces can give rise to the chiral Dzyaloshinskii-Moriya interaction (DMI) in otherwise symmetric systems [1-4]. Research has primarily focused on the study of chiral spin textures in chiral metallic systems, such as chiral domain walls and skyrmions, where a heavy metal underlayer such as Pt provides both a large spin-orbit torque and an interfacial DMI [5-7]. However, damping in metals is generally high, which limits the domain wall mobility. Magnetic insulating garnets are a broader, ubiquitous class of materials that have been extensively studied for their technologically desired spintronic properties, such as low damping and high magneto-optic constants [8-10]. Their low damping and ferrimagnetic dynamics also make them ripe candidates for ultrafast spin textures. Moreover, spin-orbit effects and spin-transport phenomenon from adjacent heavy metal layers have been used to manipulate the magnetisation in these materials [11-13]. However, the Dzyaloshinskii-Moriya interaction (DMI) and chiral spin textures have only recently been observed in rare-earth iron garnet (REIG) films, and the origins of the DMI are still not clear [14]. Here, we use a pure spin current from an adjacent Pt overlayer to characterise chiral domain wall motion in epitaxial iron garnet films with perpendicular magnetic anisotropy. By studying both the rare-earth dependence and thickness dependence of the DMI, we find that the DMI is a general feature of REIG films, and that the DMI manifests from the substrate oxide – magnetic oxide interface. Furthermore, we find that the interfacial DMI necessitates the presence of a rare-earth element in the magnetic insulator. We use the significant interfacial DMI and a large spin-orbit torque to drive chiral domain walls to unprecidented record velocities of a few thousand meters per second.

- [1] Bode, M. et al. Nature 447, 190–193 (2007).
- [2] Thiaville, A., Rohart, S., Jue, E., Cros, V. Fert, A. Eur. Lett. 100, 57002 (2012).
- [3] Fert, A., Cros, V. Sampaio, J. Nat. Nanotech. 8, 152–156 (2013).
- [4] Yang, H. et al. Nat. Mater. 17, 605–609 (2018).
- [5] Woo, S. et al. Nat. Mater. 15, 501–506 (2016).
- [6] Boulle, O. et al. Nat. Nanotech. 11, 449–454 (2016).
- [7] Jiang, W. et al. Science 349, 283–286 (2015).
- [8] Bibes, M. Barthélémy, A. IEEE Trans. Electron. Dev. 54, 1003–1023 (2007).
- [9] Kajiwara, Y. et al. Nature 464, 262–266 (2010).
- [10] Chumak, A. V, Vasyuchka, V. I., Serga, A. A. Hillebrands, B. Nat. Phys. 11, 453 (2015).
- [11] Nakayama, H. et al. Phys. Rev. Lett. 110, 206601 (2013).
- [12] Cornelissen, L. J., Liu, J., Duine, R. A., Youssef, J. Ben Van Wees, B. J. Nat. Phys. 11, 1022–1026 (2015).
- [13] Avci, C. O. et al. Nat. Mater. 16, 309–314 (2017).
- [14] Avci, C. O. et al. Nature Nano Adv Online (2019)

4512 Unravelling 3D magnetic textures with X-rays

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Three dimensional magnetic systems promise significant opportunities for new physics, ranging from ultra-high domain wall velocities and geometry-induced magnetochirality effects, to 3D topological structures as well as 3D technological devices [1,2]. Experimentally, appropriate techniques are required to map both complex three-dimensional magnetic configurations, and the response to external excitations.

For three-dimensional magnetic imaging, we have developed X-ray magnetic nanotomography [3], combining a new iterative reconstruction algorithm [4] with a dual rotation axis experimental setup, therefore providing access to the three-dimensional magnetic configuration at the nanoscale. In a first demonstration, we have determined the complex three-dimensional magnetic structure within the bulk of a micrometre-sized soft magnetic pillar and observed a magnetic configuration that consists of vortices and antivortices, as well as Bloch point singularities [3].

In addition to the static magnetic structure, the dynamic response of the 3D magnetic configuration to excitations is key to our understanding of both fundamental physics, and applications. With our recent development of X-ray magnetic laminography [5,6], it is now possible to determine the magnetisation dynamics of a three-dimensional magnetic system [5] with spatial and temporal resolutions of 50 nm and 70 ps, respectively.

A final challenge concerns the identification of nanoscale topological objects within the complex reconstructed magnetic configurations. To address this, we have recently implemented calculations of the magnetic vorticity [7,8], that make possible the location and identification of 3D magnetic solitons, leading to the first observation of magnetic vortex rings [8].

These new experimental capabilities of X-ray magnetic imaging open the door to the elucidation of complex three-dimensional magnetic structures, and their dynamic behaviour.

- [1] Fernández-Pacheco et al., Nat. Comm. 8, 15756 (2017)
- [2] Donnelly and V. Scagnoli, J. Phys. D: Cond. Matt. (2019).
- [3] Donnelly et al., Nature 547, 328 (2017).
- [4] Donnelly et al., New Journal of Physics 20, 083009 (2018).
- [5] Donnelly et al., Nature Nanotechnology 15, 356 (2020).
- [6] Witte, et al., Nano Lett. 20, 1305 (2020).
- [7] Cooper, PRL. 82, 1554 (1999).
- [8] Donnelly et al., Nat. Phys. Accepted (2020)

Threshold magnetic thickness for full emergence of interfacial Dzyaloshinskii-Moriya interaction

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Interfacial phenomena play decisive roles in modern science and technology as the scale of nanostructures shrinks down to a few atomic layers. Such minute nanostructures require more comprehensive understanding beyond the commonly accepted concept that interfacial phenomena mainly arise from the interface itself, requiring no minimum ferromagnet thickness. Contrary to the common notion, we experimentally demonstrate that interfacial phenomena require finite thickness for their full emergence. The interfacial Dzyaloshinskii-Moriya interaction (DMI) begins to be generated as the ferromagnet thickness increases and fully emerged at the thickness of 2-2.5 atomic layers, where the magnitude of DMI is maximized. This result indicates the need to refine conventional perspectives on interfacial phenomena and imposes the lowest structural bound and optimum thickness to maximize interfacial effects for technological applications.

Spin-orbitronics at a topological insulator-semiconductor interface

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Topological insulators are receiving an increasing attention since they are expected to have a high spin-charge interconversion efficiency. This is mainly due to the fact that these materials show spin-momentum locked surface states with a linear dispersion. Here we study the spin-to-charge conversion properties of a Bi₂Se₃ heterojunction (see Fig. 1): a contacted Bi₂Se₃ stripe is evaporated on top of a Ge(111) substrate and exploited as a spin detector, whereas a series of parallel Pt stripes are used as spin injectors. We use a Ge substrate since Ge is an efficient platform where spins can be injected [1] and transported thanks to the large spin diffusion length [2,3]. Optical spin injection is performed inside Ge by exploiting circularly polarized light impinging on a Pt stripe edge at normal incidence [1,3]: in this case, a spin population is generated in the Ge conduction band with a spin polarization lying in the plane of the sample. If the light beam impinges on the opposite edge of the Pt stripe, the spin polarization is reversed. Then, the spins diffuse inside Ge and are absorbed into the Bi₂Se₃ detector, where spin-to-charge conversion occurs at the Bi₂Se₃/Ge(111) interface states by means of the inverse Rashba-Edelstein effect (IREE), giving rise to a 2D charge current which can be probed as a voltage difference between the two ohmic contacts. By rastering the sample surface at normal incidence with a focused light beam, we generate spin populations at different distances with respect to the Bi₂Se₃ detector and we record an IREE map as a function of the spin generation point, as shown in Fig. 2a. From the profiles of the IREE map along the x-axis of the sample (see Fig.2b), we also estimate the electron spin diffusion length in Ge, by employing a simple unidimensional diffusion model [3], obtaining L_s = 6 µm. At this point, we calculate the conversion efficiency of the IREE process for the Bi₂Se₃ detector obtaining $\chi_{Bi2Se_3,2D}$ = -30 pm, in line with previously reported values [5]. The IREE mechanism at the Bi₂Se₃ interface is further investigated by first principle relativistic calculations, which indicate a Rashba-like helical spin texture exhibiting a counter-clockwise chirality of the hybridized Bi₂Se₃/Ge(111) states. The Bi₂Se₃ efficiency is also compared to the one of a Pt detector, where the spin-to-charge conversion occurs by means of spin-dependent scattering with Pt nuclei and is referred to as inverse spin-Hall effect (ISHE). To this purpose, we investigate a sample with the same structure of the Bi₂Se₃ one, but with a Pt stripe replacing the topological insulator detector. We find that spin-to-charge conversion of Bi₂Se₃ and Pt detectors is comparable, highlighting the importance of interface hybridization with topological insulators in spin-charge interconversion.



Figure 1: Sample Geometry.

- [1] F. Bottegoni et al., Nat. Mater. 13, 790 (2014)
- [2] C. Zucchetti et al., APL Mater. 7, 101122 (2019)
- [3] C. Zucchetti et al., Phys. Rev. B 96, 014403 (2017)
- [4] E. Sagasta et al., Phys. Rev. B 94, 060412(R) (2018)
- [5] H. Wang et al., Phys. Rev. Lett. 117, 076601 (2016)



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Robust skyrmion-bubble textures in SrRuO₃ thin films stabilized by magnetic anisotropy

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Topological spin textures in an itinerant ferromagnet, SrRuO₃, has been studied using Anomalous Hall transport. We report unconventional features in Hall transport in single SrRuO₃ films, akin to recent studies on THE in SrIrO₃/SrRuO₃ bilayers and ascribed to skyrmions. A relatively large thickness of our films and absence of heavy metal layers make the interfacial Dzyaloshinskii-Moriya interaction an unlikely source of these topological spin textures. Additionally, the transport anomalies exhibit an unprecedented robustness to magnetic field tilting and temperature. Our films are tailored to be ferromagnetic and multidomain, thus expected to display complex magnetocrystalline anisotropy dependence both with the crystalline axes and temperature below the phase transition temperature of 120 K.

Combined with numerical simulations, we infer the origin of the unconventional features in $SrRuO_3$ films results from magnetic bubbles with skyrmion topology stabilized by multiaxial anisotropies and magneto dipolar interactions in an unexpected region of parameter space. The robustness of the bubble domains against the rotation of the magnetic field vector is significantly influenced by the complex angular dependence of the multiaxial anisotropy energy in our $SrRuO_3$ films.

This work presents a first comprehensive understanding of the origin of THE in $SrRuO_3$ thin films and enhances new prospects for their manipulation with electric fields [1].

[1] P. Zhang, et al. arXiv preprint arXiv_2001.07039.

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Observation of a Tricritical point and a Lifshitz point in the skyrmion host Cu₂OSeO₃

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In recent years, topologically stable vortex-like spin structures, known as skyrmions, have been observed in various magnetic materials with different crystal symmetries and interactions. These skyrmionic materials have attracted great interest due to their potential applications in spintronics and ultra-high dense magnetic storage devices. We report the complete phase diagram of Cu_2OSeO_3 in the vicinity of Curie temperature, T_c , by analyzing field-dependent magnetization data recorded at various temperatures. Cu₂OSeO₃ crystallizes in the cubic phase in space group P2₁3, which is the same group of MnSi, FeGe, and other B20 skyrmionic materials. Existence of strong spin-orbit coupling and non-centrosymmetry give rise to Dzyloshinskii-Moria interaction, which competes with spin-spin exchange interaction resulting in rich physics, including the observation of the skyrmion phase. We present, here, the detailed analysis of Arrott plot and field-dependent entropy to identify the different phases and order of phase transitions. The order of phase transition between all the phases, the existence of a tricritical point, and a Lifshitz point were estimated. The tricritical point was observed at a finite magnetic field and temperature, where the first-order magnetic phase transition line changes into second order. Another critical point, having properties of a Lifshitz point, was observed at field-dependent T_c line at the finite magnetic field, where a field-induced phase transition line meets tangentially. The existence of Lifshitz point also gives the evidence of the existence of an incommensurate or inhomogeneous precursor phase, which is known as fluctuation disordered regime, in between the helimagnetic and paramagnetic phase. It appears that the precursor phase is essential for the formation of the skyrmion phase, at least in the cubic B20 skyrmionic materials.

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Bloch-type spin helix in bilayer Fe islands on Ir(110) by spin-polarized STM

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Magnetic films that come into contact with heavy metals have a wealth of interesting interfacial phenomena that include Dzyaloshinskii-Moriya interaction (DMI), which may lead to chiral magnetic structures. Most work on interfacial DMI focused on films exhibiting C_{3v} symmetry, known to only support Néel type spin helices and skyrmions [1]. Here, the DMI is isotropic and can be described by exactly one micromagnetic parameter. In contrast, C_{2v} symmetry systems such as the (110) surface of an fcc crystal, are predicted to show anisotropic DMI leading to various scenarios of topological pattern formation [2].

Our spin-polarized STM/STS study reveals a magnetic stripe phase with a period of 1.27 nm along the closepacked direction, [-110], in bilayer Fe islands on unreconstructed Ir(110). The spin texture behind the stripe pattern is an incommensurate spin helix. Based on detailed field dependent measurements with a vector magnet, we conclude that the spin helix is of Bloch-type, where the rotation axis of the helix is parallel to the propagation direction. We also found that the rotational sense is degenerated as left and right rotations are found in different Fe islands. These results contradict the assumption of the spin helix being driven by an interface in-plane DMI vector. Combined with theoretical insights, we discuss our findings in terms of the formation energy in systems with C_{2v} symmetry.

We acknowledge funding from Deutsche Forschungsgemeinschaft (DFG) through CRC 1238 (project number 277146847, subprojects A01 and C01) and the Jülich Supercomputing Center.

[1] S. Heinze et al. Nat. Phys. Vol.7, p.713 (2011).

[2] M. Hoffmann et al. Nat. Commun. Vol.8, p.308 (2017).

Deterministic approach for skyrmionic dynamics at non-zero temperature: pinning sites and racetracks

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The finding of room temperature skyrmions has boosted the potentiality of skyrmionic devices. From the theoretical point of view, in micromagnetic systems the temperature has been taken into account using stochastic spin dynamics, the stochastic Landau-Lifshitz equation or the Landau-Lifshitz-Block equation. Also, Thiele's rigid model can be used to describe the skyrmion dynamics taking into account thermal effects (stochastic Thiele equation). All these methods require a statistical study of the obtained data (they rely in stochastic simulations), which results in large computation time. In addition, analytical tendencies or limits can hardly be obtained.

We use a deterministic method to study the skyrmion dynamics, in which the temporal motion of skyrmions can be studied without relying on repetition of stochastic simulations, but on solving once the associated deterministic Fokker-Planck equation. By finding and solving a single partial differential equation, one could obtain all the relevant probabilistic information of the system. Indeed, within the rigid approximation, the probability density for the presence of skyrmions under any external potential can be found at any time and position, and as a function of the temperature. This allows one to evaluate the probability of presence (or of survival, or of trapping, or of escaping, etc.) of skyrmions in many practical situations, which is key information to ensure the viability of some of the skyrmion applications. In particular, we show a detailed study of the probability of trapping/escaping of a skyrmion that encounters a pinning site and of the probability of survival of a skyrmion along a racetrack.

Spin waves in skyrmions with various topological charges

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Magnetic skyrmions continue to attract considerable research interest due to their high stability, unique transport properties and characteristic magnon excitations. The most widely studied mechanism capable of stabilizing skyrmions is the Dzyaloshinsky–Moriya interaction. This leads to cylindrically symmetric isolated skyrmions on a collinear magnetic background, the spin wave modes of which may be characterized by angular momentum quantum numbers [1]. Although a high variety of differently shaped spin wave modes has been predicted theoretically, their experimental observation has been restricted by the angular momentum selection rules to breathing and gyrational modes so far, which can be excited by out-of-plane and in-plane magnetic fields, respectively.

An alternative mechanism for skyrmion stabilization is the competition between ferromagnetic and antiferromagnetic Heisenberg exchange interactions in frustrated magnets, which enables the formation of skyrmions with different topological charges simultaneously [2]. The Dzyaloshinsky-Moriya interaction distorts the initially symmetric shape of the various types of skyrmions stabilized by the frustrated Heisenberg interactions [3], and this distortion considerably influences their dynamics [4]. Here, the spin wave excitation modes of skyrmions with different topological charges are compared. It is discussed how the angular momentum selection rules depend on the topological charge, and how magnon modes belonging to different angular momentum values hybridize in distorted spin structures. Predictions on the possible experimental observation of the magnon modes in different types of skyrmions are provided.

- [1] M. Garst, J. Waizner, and D. Grundler, J. Phys. D: Appl. Phys. 50, 293002 (2017).
- [2] A. O. Leonov and M. Mostovoy, Nat. Commun. 6, 8275 (2015).
- [3] L. Rózsa, K. Palotás, A. Deák, E. Simon, R. Yanes, L. Udvardi, L. Szunyogh, and U. Nowak, Phys. Rev. B 95, 094423 (2017).
- [4] M. Weißenhofer and U. Nowak, Phys. Rev. B 99, 224430 (2019).

Interfacial exchange field in a heavy metal/ferromagnetic insulator and heavy metal/paramagnetic insulator systems by spin Hall magnetoresistance

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Spin-dependent scattering transport at heavy metal (HM)/magnetic insulator (MI) interface can be described in terms of three parameters: the so-called spin-sink conductance G_s and the real and imaginary part of the spin-mixing conductance, $G_{\uparrow\downarrow} = G_r + iG_i$ [1,2]. Each parameter is relevant for different spin-dependent phenomena. For instance, G_s originates from spin-flip processes and therefore is the leading parameter in electrical and thermal excitation of magnons [3], whereas G_r represent a spin-transfer (Slonczewski) torque to the magnetization and plays a fundamental role in spin-pumping experiments [4]. On the other hand, G_i quantifies the interfacial exchange field, which induces a field-like torque in the conduction electrons of the HM. the quantification of G_i is important for example in spin-splitting field experiments in superconductivity [5,6]. These conductances are broadly studied in ferrimagnetic insulators, where usually the contribution of G_r is much larger than that of G_i [Jia2011], leading to only few reports on the exchange field at HM/MI interfaces [7, 9].

In this work, we study the three spin conductance terms by spin Hall magnetoresistance (SMR) in two new systems: a paramagnetic insulator (PMI) such as $Gd_3Ga_5O_{12}$ (GGG) and a ferromagnetic insulator (FMI) such as EuS. By SMR measurements as a function of magnetic field in the case of the PMI and as a function of temperature in the case of the FMI, and taking advantage of the newly developed microscopic theory for SMR [2], we can extract relevant microscopic parameters such as the exchange interaction between the 1*s* electrons in Pt and the 4*f* electrons of Gd in the GGG ($J_{sf} \sim 10 \text{ meV}$) or Eu in EuS ($J_{sf} \sim 18 \text{ meV}$). Our results demonstrate that, in HM/PMI interfaces at large magnetic field, the field-like torque contribution (G_i) is as important as the spintransfer torque contribution (G_r). Furthermore, for the HM/FMI case, we demonstrate for the first time experimentally a G_i which is larger than G_r , as suggested to be the case for europium chalcogenides [10]. Unlike in ferrimagnets, where the local magnetic moments at the interfaces are partially compensated, all local moments contribute equally to the interfacial exchange field in these two studied examples, giving rise to such a large G_i .

- [1] T.-Y. Chen et al., Phys Rev. B 87, 144411 (2013)
- [2] X.-P. Zhang et al., Nano Lett. 19, 6330 (2019)
- [3] L. J. Cornelissen et al., Nat. Phys. 11, 1022 (2015).
- [4] M. Weiler et. al., Phys. Rev. Lett., 111, 176601 (2013)
- [5] B. Li et al., Phys. Rev. Lett. 110, 097001 (2013).
- [6] M. Rouco et al., Phys. Rev. B 100, 104523 (2019).
- [7] N. Vlietstra et al., Appl. Phys. Lett. 103, 032401 (2013).
- [8] X. Jia et al., Europhysics Lett. 96, 17005 (2011)
- [9] T. Kosub et al., Appl. Phys. Lett. 113, 222409 (2018)
- [10] A. Brataas et al., Phys, Rep. 427, 157 (2006)

Rashba-like spin-orbit coupling in PbTiO₃

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Understanding and manipulating the spin degree of freedom in materials is a hot topic due to the potential applications to develop new computing systems or low-consumption-operating electronic devices. First well-known in semiconductor heterostructures, and now in many other materials such as transition-metal oxides, the Rashba spin-orbit effect, which appears as a result of the surface inversion asymmetry, has recently motivated lots of research due to its role in transport processes and its ability to be tuned by an external electric field. In 2013, the Rashba effect has been predicted theoretically¹, and later confirmed experimentally², in ferroelectric materials. The link between the reversible spin textures resulting from the spin-orbit interaction and the electric polarization is very promising to design devices based on non-volatile magneto-electric states.

We will present our recent results based on *ab initio* calculations applied to the Rashba spin-orbit effect in the well-known ferroelectric perovskite $PbTiO_3^3$. We propose to detail the spin-texture differences linked to the Ti-d and Pb-p bands of $PbTiO_3$, which exhibit respectively a *cubic* and *linear* Rashba spin-splitting (RSS) variation as a function of the wave vector \mathbf{k} . The effect of strain on the most stable phase as a route to modify the RSS and the spin textures will be discussed.

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- [1] D. Di Sante, et al., Adv. Mater. 25, 509 (2013).
- [2] C. Rinaldi, et al., Nano Lett. 18, 2751 (2018).
- [3] R. Arras, et al., Phys. Rev. B 100, 174415 (2019).

Single magnetic skyrmions in restricted geometries

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Non-linear localized excitations (solitons) are attracting the attention of physicists for a long time. The solitons can be often classified by integer numbers (topological charges), which are preserved in their dynamics. Topologically non-trivial magnetization configurations in ferromagnets, such as domain walls and skyrmions are currently the focus of activity of researchers working in the area of solid-state magnetism. In this talk I consider 2D magnetic topological solitons – skyrmions in restricted geometries: magnetic films and dots. The magnetic skyrmions possess intriguing and novel properties due to their topologically non-trivial magnetization configurations.

The talk is organized as follows. First, I present a definition of the topological charges of the magnetic skyrmions and conduct a classification of the magnetic skyrmions. Then, stabilization of the individual skyrmions by the Dzyaloshinskii-Moriya exchange interaction (DMI) and magnetostatic interaction is considered. The stability and sizes of chiral skyrmions in ultrathin magnetic films and dots are calculated accounting for the isotropic exchange, Dzyaloshinskii–Moriya exchange interaction, and out-of-plane magnetic anisotropy within micromagnetic approach. The Bloch skyrmions in magnetic films with B20 cubic crystal structure and Neel skyrmions in ultrathin films and multilayers Co/X (X = Ir, Pd, Pt) are considered. The skyrmions can be stabilized due to two different mechanisms, primary DMI or primary magnetostatic interaction, leading to small and large size skyrmions, respectively. The single skyrmion stability in circular magnetic nano dots is considered in detail.

The emergent electromagnetic field generated by the skyrmion magnetization texture, its consequences, low and high frequency spin excitation modes on the skyrmion magnetization background are considered. It is shown that the topological Hall effect and skyrmion Hall effect are direct consequences of the skyrmion non-zero topological charge.

The main features of the dynamics of magnetic skyrmions in nanodots including the spectra of spin waves and gyrotropic modes of an isolated skyrmion are calculated by using the Landau-Lifshitz equation of the magnetization motion. The classification of the excitation modes of skyrmion state dots is performed similar to that developed for the spin excitations in the magnetic vortex state dots based on the azimuthal and radial mode indices (m, n). The gyrotropic mode has the lowest frequency in the excitation spectrum and is localized in the central area of nanodot. The gyrotropic frequencies of the Bloch- and Neel-type magnetic skyrmions are calculated in the GHz range as a function of the skyrmion equilibrium radius, dot radius, and the dot magnetic parameters. The critical currents and nonlinear gyrotropic frequencies of the skyrmion spin torque oscillator are calculated.

Engineering chiral magnetic textures in W/Co/AuPt thin films

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Topological magnetic textures such as skyrmions have been extensively studied during the last decade due to their potential application in spintronics devices. In ultrathin films systems, a key parameter to stabilize chiral topological magnetic textures is the Dzyaloshinskii-Moriya interaction (DMI), obtained at the interfaces between ferromagnetic layers and heavy-metals, promoting a unique chirality of the magnetic texture.

In this work, we focus on an ultra-thin epitaxial $W/Co(0.6-1nm)/Au_{1-x}Pt_x$ trilayer, a model system exhibiting perpendicular magnetic anisotropy and interface DMI. Moreover, due to the epitaxial growth of Co(0001) on W(110) following the Nishiyama Wasserman relationships, the circular symmetry is broken, giving rise into a C_{2v} crystal symmetry at the W/Co interface. This symmetry results into a biaxial magnetic anisotropy, with an in-plane easy axis along the W[1-10] direction, and into an anisotropic DMI [1]. The balance between the in-plane anisotropy and the DMI leads to an anisotropic domain wall configuration along the easy- and hard axis. When the domain wall is oriented along the W[1-10] direction, the in-plane easy-axis, the DMI is promoting a Néel-like rotation while the in-plane anisotropy is promoting a Bloch-like rotation. On the other hand, when the domain wall is oriented along the W[001] direction, the in-plane hard-axis, the DMI and the in-plane anisotropy promote a Néel-like rotation. Consequently, the competition between both interactions lead to an anisotropic domain wall configuration as well as domain wall energy. This is confirmed experimentally by our X-ray magnetic circular dichroism photoemission electron microscopy (XMCD-PEEM) experiments [2]. Figure 1a shows chiral stripedomains mostly oriented along the in-plane hard-axis as a consequence of the lower energy for domain walls oriented along this axis, with respect to the in-plane easy axis. Another example is shown in Figure 1b, where the stripe-domains are transformed into elliptical skyrmions when a weak out-of-plane magnetic field is applied which is confirmed by our micromagnetic simulations (Figure 1c).

In this presentation, we will show the effect of different Pt-to-Au composition on the magnetic properties such as the perpendicular magnetic anisotropy and the DMI by Brillouin light spectroscopy. Finally, we will show the observation by XMCD-PEEM of different chiral magnetic textures in sub-micron circular dots, and discuss under which conditions these textures can be stabilized.



Figure 1: a) Chiral stripe domains aligned along the W bcc[001] observed with XMCD-PEEM. b) Elliptical skyrmion obtained after the application of a weak out-of plane field. c) Micromagnetic simulated elliptical Skyrmion stabilized in a 400-nm circular dot.

[1] Camosi, L. et al. Physical Review B 95, 214422 (2017).

[2] Camosi, L. Pena Garcia, J., et al. to be submitted.

Observation of coexistence of different skyrmion phases in hybrid ferromagnetic/ferrimagnetic multilayers

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Thin films with interfacial Dzyloshinskii-Moriya interaction (DMI) was shown to support skyrmions at room temperature. Skyrmions are promising candidates for information carriers in memory devices [1]. In a racetrack memory, a chain of skyrmions needs to be moved between pinning centers defined by notches. In this device, skyrmions has to be stable with fixed inter-skyrmion distances. However, distribution of the pinning energies and intrinsic pinning cites in the racetrack material lead to variations in the depinning currents, times and velocities causing bit errors [2]. To reduce the error rates, a racetrack device consisting of a double layer structure was proposed [3]. The first layer supports linear skyrmion lattice and coupled to the second, while the second layer may contain a skyrmion or not at given position, representing 1 or 0 bit, respectively.

Here we report a thin film system consisting of three main layers (Fig. 1A) that supports two different skyrmion phases at room temperature (Fig. 1B and C). The top and bottom ferromagnetic (FM) layers consist of [Ir/Fe/Co/Pt]_{x5} sublayers which supports skyrmions. In between FM layers is a ferrimagnetic (Fi) layer consisting of TbGd/Co-multilayers. Additionally, individual main layers (FM or Fi) and different combinations these (e.g., FM/FM, Fi/FM, FM/Fi, FM/Ta/FM) have been fabricated for comparison. In one of the systems, Fi layers was replaced with a nonmagnetic Ta layer. All systems have been studied by in-vacuum quantitative magnetic force microscopy (qMFM). qMFM image of the heterostructure under 133 mT magnetic field reveals a dense pattern of skyrmions with two distinct contrast levels (Fig. 1B and C). All other samples show dense patterns of skyrmions as well. However, their contrast is comparable either to the strong contrast skyrmion of the FM/Fi/FM heterostructure (e.g., FM/FM and FM/Ta/FM), or weak contrast skyrmions (e.g., FM and FM/Fi). From micromagnetic simulations we have found that two different types of skyrmion spin textures can be stable depending on the initially set spin-texture. First, if a skyrmion spin texture is initially imposed only into the top and bottom FM layers and not in the Fi layer, the relaxed structure reveals skyrmion spin textures with clock-wise (CW) Néel walls in all sublayers of the top FM layer and in the bottom four sublayers of the bottom FM layer, while the magnetization of the Fi layer is nearly homogeneous. This results in smaller-diameter skyrmions. On the other hand, if a skyrmion spin texture is also imposed into the Fi layer in the initial state, the final state shows a larger skyrmions going through all sublayers. While the spin texture in the top FM layer again shows CW Néel walls, the texture is more complex in Fi and bottom FM layer and governed by the competition of DMI, sublayer exchange coupling, and demagnetization fields. Note that the larger diameter of this skyrmion explains the higher contrast of the larger skyrmions observed by MFM. The micromagnetic calculations confirm the experimentally observed co-existence of two skyrmion phases. The observed co-existence of two distinct skyrmion states at room temperature in the heterostructure is governed by a balance of the different magnetic energy density terms arising from the different multilayers and their exchange and magnetostatic coupling. Our results suggest that transition between the skyrmion states can be exploited in a 3-dimensional data storage device.



Tuning the Skyrmion Hall effect via Engineering of Spin-Orbit Interaction

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Ferromagnetic skyrmions are promising information carriers for nonvolatile, energy-efficient, ultradense memory and logic devices of the future. However, their integration in these applications is hampered by the undesirable skyrmion Hall effect (SkHE), a motion transverse to the direction of current flow. In this study we demonstrate that the Magnus force acting on magnetic skyrmions can be efficiently tuned via modulation of the strength of spin-orbit interactions. We show that the skyrmion Hall effect, which is a direct consequence of the nonvanishing Magnus force on the magnetic structure, can be suppressed in certain limits. Our calculations show that the emergent magnetic fields in the presence of spin-orbit coupling (SOC) renormalize the Lorentz force on itinerant electrons, and thus, influence topological transport. In particular, we show that, for a Néel-type skyrmion and Bloch-type antiskyrmion, the skyrmion Hall effect (SkHE) can vanish by tuning appropriately the strength of Rashba and Dresselhaus SOCs, respectively. Our results open up alternative directions to explore in a bid to overcome the parasitic and undesirable SkHE for spintronic applications.

Material engineering and fast domain wall motion governed by topology in cylindrical nanowires

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Current-induced magnetic domain wall (DW) motion has attracted a broad interest, not only in the context of futuristic 3D data storage concepts, but also from a fundamental point of view. A textbook case for both fundamental and applied focus are cylindrical nanowires (NW). In contrast to flat nanostrips, simulations and theory state that NWs should not experience intrinsic DW instabilities responsible for the Walker breakdown, and simultaneously give rise to fascinating new physics [1]. Key to these predictions is the novel Bloch-point wall (BPW), which is unique to NWs [2]. It has a distinct topology and exhibits azimuthal curling of magnetic moments around a Bloch-point [3], a local vanishing of magnetization at the DW core. This feature and its rotational symmetry are at the origin of the predicted steady-state dynamics of this DW type, allowing theoretically speeds of over 1000 m/s and the controlled emission of spin waves (spin-Cherenkov effect) [4].

Here we report unprecedented experimental results on current-induced DW motion in NWs [5]. We use magnetic force microscopy and shadow X-Ray Magnetic Circular Dichroism coupled to PhotoEmission Electron Microscopy (XMCD PEEM) to image DWs in <100 nm diameter ferromagnetic CoNi NWs before and after the application of nanosecond current pulses. We evidence that the specific topology of the BPW together with the stabilizing effect of the previously overlooked Œrsted field allow wall velocities >600 m/s (FIG), quantitatively consistent with predictions and providing a lower bound for the non-adiabatic STT contribution. We also screened material composition and microstructure to minimize extrinsic DW pinning, correlated with advanced structural analysis. Present work aims to 1/ improve the accuracy of the velocity measurements by reducing DW pinning, thereby also approaching the conditions where the spin-Cherenkov effect is expected. 2/ Further increase DW speed by tuning damping and non-adiabatic terms through material engineering.



Figure 1: BPW speed as a function of applied current density, monitored with MFM (open circles) and XMCD PEEM (full circles) [5].

- [1] Fernandez-Pacheco A., Streubel R., Cowburn R.. Nat. Commun., p.15756 (2017).
- [2] Da Col S., Jamet S., Fruchart O.. Phys. Rev. B, p. 180405 (2014).
- [3] Feldtkeller R., Z. Angew. Physik, p. 530 (1965).
- [4] Hertel R.. J. Phys.: Condens. Matter, p. 483002 (2016).
- [5] Schöbitz M., De Riz A., Fruchart O.. Phys. Rev. Lett., p. 217201 (2019).

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Prospecting chiral multi-site interactions in prototypical magnetic systems

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Atomistic spin models have found enormous success in addressing the properties of magnetic materials, grounded on the identification of the relevant underlying magnetic interactions. The huge development in the field of magnetic skyrmions and other noncollinear magnetic structures is largely due to our understanding of the chiral Dzyaloshinskii-Moriya interaction. Recently, various works have proposed new types of chiral interactions [1,2,3], with seemingly different forms, but the big picture is still missing.

Here, we present a systematic construction of a generalized spin model containing isotropic and chiral multi-site interactions. These are motivated by a microscopic model that incorporates local spin moments and the spin-orbit interaction, and their symmetry properties are established.

We show that the chiral interactions arise solely from the spin-orbit interaction and that the multi-site interactions do not have to follow Moriya's rules, unlike the Dzyaloshinskii-Moriya and chiral biquadratic interactions [1]. We then illustrate our theoretical considerations with density functional theory calculations for prototypical magnetic systems. These are triangular trimers built out of Cr, Mn, Fe and Co adatoms on the Re(0001), Pt(111) and Au(111) surfaces, for which C_{3v} symmetry applies, and Cr and Fe square tetramers on Pt(001) with C_{4v} symmetry. The multi-site interactions are substantial in magnitude and cannot be neglected when comparing the energy of different magnetic structures. Finally, we discuss the recent literature in light of our findings, and clarify several unclear or confusing points.

- [1] S. Brinker et al., New. J. Phys. 21, 083015 (2019)
- [2] A. Lászlóffy et al., Phys. Rev. B 99, 184430 (2019)
- [3] S. Grytsiuk et al., Nat. Commun. 11, 511 (2020)

Anisotropic skyrmion diffusion controlled by magnetic field directions

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Thermally activated processes are intrinsic effects in every physical system and their understanding key to the underlaying dynamics of such systems. Using skyrmions, magnetic, topologically stabilized spin structures, we investigate the thermal diffusion dynamics in specifically tailored metal-multilayer. We find that for low pinning materials stacks, the thermal diffusion dominates the dynamics and allows for stable skyrmions at room temperature that move by thermal activation [1].

These stable Skyrmions allow for a wide range of possible applications in logic, data storage or Brownian token computing devices [2]. In Brownian circuits as well as other applications, the control of the skyrmion diffusion and the implementation of a certain control is necessary [3]. We show that by applying an in-plane field, the skyrmion diffusion becomes anisotropic with faster diffusion parallel to the field direction than perpendicular to it. We furthermore show that the absolute value of the applied field also controls the absolute values of the diffusion coefficients so that one can tune both the orientation of the diffusion and its strength. We can analytically and numerically explain the anisotropic diffusion due to an elliptical deformation of the skyrmions by the application of the in-plane field, which leads to a preferential diffusion axis.

- [1] J. Zázvorka et al., Nat. Nanotech. 14, 658–661(2019).
- [2] F. Peper et al., ACM J. Emerg. Technol. Comput. Syst. 9, 3 (2013).
- [3] T. Nozaki et al., Appl. Phys. Lett. 114, 012402 (2019).

Integration of synthetic antiferromagnetic-based free layer in domain wall devices with magnetic tunnel junction read and write components

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The experimental demonstration of spin-orbit torque -driven chiral Neel domain wall (DW) motion at high speed, has fueled the development of fast and energy-efficient logic and memory applications such as racetrack memory [1] and the spin-torque majority gate [2]. Information is encoded in magnetic DWs in a track shared by multiple perpendicular magnetic tunnel junction (pMTJ) pillars which act as inputs and outputs of the device. For practical applications, many significant challenges exist. These are particularly related to the integration of the reading and writing components with MTJs and to the fabrication of such devices using an industrial integration platform [3].

We propose to integrate a synthetic antiferromagnetic (SAF) material as the free layer (FL) of a pMTJ in fully integrated DW-based devices, aiming for high DW velocities and low pinning. Our SAF-based DW conduit is compatible with CoFeB-MgO based pMTJs allowing for efficient spin-transfer torque (STT) write and high tunneling magnetoresistance (TMR) read at the inputs and outputs of the device. Fig. 1 compares pMTJ pillars with a dual MgO-based FL and a SAF-based FL. We demonstrate no compromise of TMR readout signal in our SAF-based FL (a) and efficient STT reversal (b). Additionally, antiferromagnets are less sensitive to external perturbations as can be seen from the reduced offset field in our SAF-based sample (c).

Lastly, the SAF-based FL design can be fully integrated in DW-based devices in imec's 300 mm CMOS fab with a BEOL-compatible flow. In a fully integrated DW-based device, the perpendicular magnetic anisotropy of our SAF-based FL can be decoupled from the MgO-induced interfacial anisotropy of CoFeB, widening the process window significantly. We demonstrate full operational capability of a DW-based device where a DW can be locally generated by STT below the first pillar and the motion of the DW, driven by external field, is monitored and read using TMR at the second pillar, Fig. 2.

Moreover, driven by spin-orbit torque, high DW velocities could be reached. This demonstration leads the way towards the future development of ultra-fast and more efficient spintronic devices.



Figure 1: (a) Comparison of tunnel magnetoresistance, measured from resistance vs field loops in single MTJs. (b) TMR as a function of voltage amplitude with 50ns pulse width showing STT-driven magnetization reversal (c) offset field measured from resitance cersus field loops.



Figure 2: TEM of 2 MTJs sharing the same FL, a DW is nucleated by STT at pillar 1 and arrives after some depinning time at pillar 2 by a small external magnetic field.

[1] S. S. P. Parkin, U.S. Patent 6834005 (2004)

[2] Nikonov, D. et al., IEEE Electron Device Letters 32.8 (2011): 1128-1130.

[3] Raymenants, E., et al. 2018 IEEE International Electron Devices Meeting (IEDM), 2018.

Ferromagnetic Resonance of skyrmions in thin film multilayers

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The observation of magnetic skyrmions at room temperature has triggered extensive research to decipher their various static and dynamic properties for potential applications in memory and logic devices [1]. In the case of ultra-thin multilayer films consisting of heavy metal_(HM)/ferromagnet_(FM)/insulator_(I), skyrmions are primarily stabilized by interfacial Dzyaloshinskii-Moriya interaction in combination with: perpendicular anisotropy, dipolar, exchange and Zeeman energies, where each of these contributions can be finely tuned [2]. Owing to their non-trivial topology, skyrmions are theoretically shown to exhibit unique spectral signatures when excited by an RF magnetic field [3], opening up new prospects for skyrmion-based microwave detectors [4] and nano-oscillators [5]. However, the experimental observation of their excitation modes is challenging due to usually elevated damping parameters and material inhomogeneities in sputtered HM/FM/I systems.

Here we study the magnetization dynamics in a [Pt/CoFeB/AlOx]×20 multilayer deposited by sputtering. First, the system is optimized by tuning the Pt and CoFeB thicknesses to host magnetic skyrmions at room temperature, along with a minimized damping parameter $\alpha \sim 0.02$, measured by ferromagnetic resonance (FMR). Next, we observe the static magnetic domain configuration using magnetic force microscopy (MFM). While sweeping the field from saturation (0.4 T) to zero, the MFM images reveal random skyrmion nucleation, followed by an increase in their density, forming a lattice structure. This lattice phase then breaks into a mixture of skyrmions and stripes at lower fields, finally transforming into labyrinthine domains. The dynamic response of the system is then measured by broadband FMR over a frequency range of 0.5-20 GHz, with an out-of-plane magnetic field swept over ± 0.6 T. The frequency-field dispersions show several resonant modes corresponding to the domain configurations observed by MFM at the respective applied fields. At fields above the saturation, the well-known Kittel modes are observed, pertaining to uniform precession. Below saturation, distinct modes arise in the resonance spectrum with both positive and negative dispersions, indicating the spectroscopic signatures of the following phases: randomly distributed skyrmions, skyrmion lattice, skyrmion-stripe mix, labyrinthine domains. Finally, we present preliminary results on the study of skyrmion dynamics in patterned nanodisks with an integrated microwave antenna, using magnetic resonance force microscopy (MRFM) [6]. This technique shall allow probing locally magnetic skyrmion dynamics in confined geometries.

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- [1] A. Fert et al., Nat. Rev. Mater. 2, 17031 (2017)
- [2] W. Legrand et al., Sci Adv 4, 7, (2018)
- [3] J.-V. Kim, et al., Phys. Rev. B 90, 064410 (2014)
- [4] G. Finocchio et al., Appl. Phys. Lett. 107, 262401 (2015)
- [5] F. Garcia-Sanchez et al., New J. Phys. 18, 075011 (2016)
- [6] V. Castel et al., Phys. Rev. B 85, 184419 (2012)

Topological Hall effect in thin films of the tetragonal inverse Heusler compound Mn_xPtSn

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Topological spin textures in quantum materials are of great interest, along with the associate transport signatures, for next-generation spintronic applications. Recently, the tetragonal Heusler compounds [1] show to host antiskyrmions, in addition to other topological spin textures of interest. Spin chirality in metallic materials with noncoplanar spin structure gives rise to a Berry phase induced topological Hall effect (THE). The THE is a transverse response to an applied current that can be used to distinguish magnetic textures for device applications, such as the racetrack memory.

Here, we present the structural, magnetic, and transport properties in epitaxial thin films of the tetragonal Mn_xPtSn Heusler compound [2]. We tune the Mn content (x) by magnetron sputtering, which allows for microscopic control of the magnetic exchange parameters. With our thin film method, we can access a novel and fundamental understanding of this compound not possible with other methods. We show a microscopic control of the exchange parameters that influence the size of the magnetic textures and thereby the transport signatures. We focus on the anomalous and chiral-type Hall effects and the behavior of the dc-magnetization, in relation to chiral spin textures.

A. K. Nayak et al., Nature 548, 561-566 (2017).
 P.Swekis et al., Phys. Rev. Materials 3, 013001(R) (2019).

Tuning the properties of zero-field room temperature ferromagnetic skyrmions by interlayer exchange coupling

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Magnetic skyrmions [1], topologically protected magnetic quasi-particles, are at the core of many of the recently proposed spintronic devices. Many different materials systems have been shown hosting magnetic skyrmions at room temperature [2,3]. However, usually an external magnetic field is required to stabilize skyrmions in unpatterned magnetic thin-films, and this is not desirable for real applications due to scalability issues. Accordingly, it is highly desirable to have a robust and tunable protocol to build materials systems hosting magnetic skyrmions in zero external magnetic fields.

Here we report the observation of isolated ferromagnetic skyrmions at room temperature and zero magnetic field [4], stabilized through interlayer exchange coupling [5,6] between the reference magnet and the free magnet of an epitaxial magnetic multilayer. By carefully choosing the material and the thickness of the non-magnetic spacer in between the two magnets, we are able to tune the handedness and the size of the observed skyrmions, whose 3D spin-texture is directly imaged by spin-polarized low energy electron microscopy. Our findings open up possibilities to the development of skyrmion-based spintronic devices suitable for general-use applications which go beyond modern nanoelectronics.

- [1] Romming N. et al., Science 341, 636- 639 (2013).
- [2] Jiang W. et al., Science 349, 283-286 (2015).
- [3] Moreau-Luchaire C. et al., Nature Nanotechnol. 11, 444-448 (2016).
- [4] Lo Conte R. et al., Nano Lett. 20, 4739-4747 (2020).
- [5] Nandy A. K. et al., Phys. Rev. Lett. 116, 177202 (2016).
- [6] Chen G. et al., Appl. Phys. Lett. 106, 242404 (2015).

Magnetic skyrmions of hybrid chirality in ferromagnetic and synthetic antiferromagnetic multilayers: design and measurements

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Magnetic skyrmions are localized magnetic structures in planar systems, behaving as single particles and topologically different from the uniform state, which have been identified to be extremely promising for applications, as well as of fundamental interest [1]. The magnetism community has provided a great effort in the last couple of years to succeed to stabilize them at room temperature, most often by designing magnetic and heavy-metal multilayers combining perpendicular magnetic anisotropy and Dzyaloshinskii-Moriya interaction (DMI) [2]. The DMI promotes a unique chirality of the skyrmionic spin textures and, combined with spin-orbit torques generated in the heavy-metal layer(s), allows efficient current-induced motion of skyrmions [3]. However, recent observations have revealed that multilayered skyrmions are prone to present a hybrid chirality, due to the competition between dipolar interaction and DMI, which can result in a dramatic annihilation of the currentinduced motion [4]. Therefore, we have built an extensive model for describing hybrid chirality in multilayered magnetic skyrmions [5]. We will discuss the impact of such hybrid chirality in technologically relevant stacks, depending on number of layers, interfacial anisotropy, DMI, etc. We will show that the multilayered nature of these skyrmions has a large influence on their size and stability, which has been neglected so far [6]. A good match between the spin-orbit torque injection geometry and hybrid chirality is required to achieve efficient motion [4,6]. Under some conditions, the skyrmion Hall effect of hybrid chiral skyrmions can be compensated, to reach a regime with zero deflection angle [6]. We will also describe how these results are to be revisited for antiferromagnetic skyrmions, which allow to cancel the skyrmion Hall effect and stabilize smaller skyrmions [7]. Detecting the SAF textures is challenging, and we will demonstrate how the electronic magnetotransport can be used to detect spin spirals and other textures in SAF multilayers [8].

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- [1] A. Fert, N. Reyren, V. Cros, Nat. Rev. Mat. 2 (2017), 17031.
- [2] C. Moreau-Luchaire et al, Nat. Nanotech, 11 (2016), 444 ; S. Woo et al, Nat. Mater. 15 (2016), 501 ; A. Soumyanarayanan et al, Nat. Mater. 16 (2017), 898 ; A. Hrabec et al, Nat. Comm. 8 (2017), 15765.
- [3] K. Litzius et al, Nat. Phys. 13 (2017), 170 ; S. Woo et al, Nat. Comm. 8 (2017), 15573.
- [4] W. Legrand et al, Sci. Adv. 4 (2018), eaat0415 ; Y. Dovzhenko et al, Nat. Comm. 9 (2018), 2712.
- [5] F. Büttner et al, Sci. Rep., 8 (2018), 4464 ; A. Bernand-Mantel et al, SciPost Phys. 4 (2018), 027.
- [6] W. Legrand et al, Phys. Rev. Applied 10 (2018), 064042.
- [7] W. Legrand et al, Nat. Mater. 19 (2020), 34-42
- [8] D. Maccariello et al, to be published.

Field Dependance of Hybrid Chiral Textures in Ferromagnetic Multilayers

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Chiral magnetic structures induced by Dzyaloshinskii-Moriya interaction (DMI) have been pro-posed as the cornerstone of new technology applications such as high-density data storage devices or neuromorphic computing [1], due their room temperature stability and efficient current induced motion. In this work, we used soft X-ray Magnetic Scattering (XRMS) to reveal directly the chiral properties of FM multilayers with tailored magnetic chiralities driven by spin-orbit-related effects at interfaces [2,3]. We show that it can straightforwardly and unambiguously determine the main characteristics of chiral magnetic distributions in perpendicularly magnetized multilayers [4]: its chiral nature, the quantitative winding sense (clockwise or counterclockwise), and its type, i.e. Néel (cycloidal) or Bloch (helical). Moreover, we prove that this approach combined with micromagnetic simulations reveals hybrid chiral spin texture in multilayers [5]. Finally, we study the in-plane field dependence intensity of the dichroism and the appearance of second order diffraction peaks, usually forbidden on these systems. A hysteretic behavior of the magnetic asymmetry ratio with the external field suggest an interplay between the Bloch and Néel components of the hybrid chiral domain texture. XRMS simulations on the simulated magnetic states of the multilayer at different fields shows the same experimentally observed behavior of the magnetic asymmetry ratio for the second order peak which is produced by the in-plane magnetization pattern formed when the field is applied on plane.

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Figure 1: (a) Magnetic asymmetry for 0.175 T in-plane external field (b) Magnetic asymmetry peaks intensity in function of each field step (positive to negative direction), after being saturated at 1 T.

[1] A. Fert, N. Reyren and V. Cros, Nat. Rev. Mat. 2, 17031 (2017)
[2] J.-Y. Chauleau et al., Phys. Rev. Lett. 120, 037202 (2018)

[3] W. Legrand et al., Sci. Adv. 4, eaat45 (2018)

Ab initio calculation of the Dzyaloshinskii-Moriya interaction in correlated magnetic systems

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Magnetic materials with stable skyrmionic behavior attract growing attention due to their promising applications in spintronics. The formation of skyrmions requires, among all, the presence of considerable Dzyaloshinskii-Moriya (DM) interaction, which is induced by spin-orbit coupling effects and broken space-inversion symmetry of the crystal lattice. In this work, we propose an *ab initio* scheme for an accurate calculation of the DM interactions in periodic crystalline systems, based on the electronic structure determined using the RSPt full-potential code [1]. Importantly, the dynamical mean-field theory can be combined with these calculations, which allows to study the impact of electronic correlations on the isotropic Heisenberg [2] and anisotropic DM magnetic exchange parameters. The performance of the proposed calculation scheme is shown for a few well-known systems with skyrmions or other types of non-trivial magnetic order. This includes B20 compounds (MnSi and FeGe) with a helical magnetic ground state and transition metal monolayers on surfaces of heavy metals, such as Co/Pt(111) and Mn/W(001). Our calculations reveal that electronic correlations can change the strength of the DM interaction in these systems by around 10%, which is comparable to the variation of the isotropic Heisenberg exchange parameters. The observed effects are interpreted in terms of the orbital-resolved electronic structure which is renormalized by electronic correlations.

 See e.g. J.M. Wills, M. Alouani, P. Andersson, A. Delin, O. Eriksson, A. Grechnev, "Full-Potential Electronic Structure Method, Energy and Force Calculations with Density Functional and Dynamical Mean Field Theory" (Springer Series in Solid-State Sciences, Volume 167, 2010), DOI 10.1007/978-3-642-15144-6
 Y. O. Kvashnin et al., Physical Review B 91, 125133 (2015).

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Experimental correlation of Interfacial Dzyaloshinskii-Moriya interaction amplitude and Work Function in Magnetic Multilayers

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Magnetic multilayers (MML) with large perpendicular magnetic anisotropy (PMA) and Dzyaloshinskii-Moriya interaction (DMI) have attracted great attention in recent years owing to the possibility of stabilizing non-collinear magnetic textures such as chiral domain walls (DW) [1], spin spirals [2] or skyrmions [3-4] with Néel chiral spin rotation. The latter are promising candidates as carriers of information for next-generation race-track magnetic memories or logic devices. To this aim, the accurate experimental determination of DMI is an important challenge allowing the engineering of future devices with tailored properties. The determination of the effective DMI amplitude D in MML is still a current subject of research, in particular for metallic multilayers with a high number of repetitions. In this study, we have performed a series of measurements to determine DMI values by two different techniques: domain size periodicity (λ) [5] of aligned-stripe domains after in-plane (IP) demagnetization by magnetic force microscopy (MFM), and asymmetric expansion of domains in the presence of an in-plane magnetic field, by Kerr microscopy [6-7]. For this purpose, we selected different materials to build asymmetric trilayers, with the general structure is $[Pt/Co/M]_{xN}$ with M = Ru, Ni, Pd, Al, Al/Ta and N = 2, 3, 4, 5, 6 the number of repetitions. We observe a correlation between $D_s = D/t$ (t is the Co thickness), the interfacial DMI obtained in the previous methods and intrinsic material parameters such as atomic number (Z), Pauling electronegativity (χ) [8] work function (ϕ) [9] of Co/M interfaces. We find a clear linear behaviour between D_s and ϕ . This correlation points to Rashba-like interfacial fields, leading to the modulation of the effective interfacial DMI.

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- [1] A. Thiaville et al. EuroPhys. Lett, 100, 57002 (2012)
- [2] P. Ferriani et al. Phys. Rev. Lett. 101, 27201 (2008).
- [3] A. Fert, N. Reyren, V. Cros, Nat. Rev. Mat. 2, 17031 (2017).
- [4] C. Moreau-Luchaire et al. Nature Nanotechnology 11, 444 (2016)
- [5] I. Lemesh et al. Physical Review B 95, 174423 (2017)
- [6] F. Ajejas et al. Appl. Phys. Lett. 111, 202402 (2017)
- [7] T. Ha Pham et al EPL 113 67001 (2016)
- [8] H. Jia et al. Phys. Rev. M 4, 024405 (2020)
- [9] Y.-K. Park et al. NPG Asia Materials 10, 995 (2018)

Tuning magnetic anisotropy and skyrmion stability with electric field

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Recently, we experimentally demonstrated that the nucleation and annihilation of µm-sized skyrmions in Pt/Co/AlOx trilayers can be controlled by tuning the perpendicular magnetic anisotropy (PMA) with electric field gating [1]. Here, we show our present efforts to extend this concept to smaller skyrmions, down to 50 nm lateral size. Two trilayer systems, in which sub-100 nm skyrmions were observed at room temperature, were studied: Pt/Co/MgO [2] and Pt/Co/TbOx. The stacks were patterned by electron beam lithography and ion beam etching into stripes (1 – 50 μ m width) then covered with a 10 nm thick ZrO₂ dielectric layer and a 6 nm thick Pt top electrode to form capacitor-like structures, Fig (a). Using Magnetic Force Microscopy, Fig (b-c), we show that in both stacks, starting from a stable skyrmion network, a positive electric field (EF) leads to the increase of the PMA and to the annihilation of the skyrmion phase. We attribute this to the EF-induced migration of oxygen towards the top Co interface. This result is confirmed by the variation of the hysteresis loops taken with polar Magneto-Optical Kerr Effect, Fig (d). This new state persists after removing the EF i.e. EF-gating leads to a nonvolatile effect. In the Pt/Co/MgO trilayer, the application of a negative EF shows no effect on the magnetic anisotropy, preventing us to renucleate the skyrmions. This irreversible process is probably due to the stabilization of the stochiometric MgO phase induced by oxygen ion migration towards the Co/MgO interface when a positive gating is applied. On the contrary, for Pt/Co/TbOx trilayers, the application of a negative EF triggered the oxygen ion migration away from the top Co interface. This allows us to tune reversibly the magnetic anisotropy, and therefore to reproducibly nucleate and annihilate 50 nm size skyrmions. This is an easily integrable and energetically efficient solution that might ease the use of magnetic skyrmions as information carriers in future spintronic devices.



Figure 1: (a) Schematic representation of the device. MFM images of Pt/Co/MgO trilayers showing magnetic skyrmions (b) before (c) after applying a positive electric field. (d) PMOKE hysteresis loops of Pt/Co/MgO trilayers before and after EF gating.

Schott et al., Nano Lett 17, 3006 (2017).
 Boulle et al., Nat Nano 11, 449 (2016)

Magnetic skyrmions probed by SP-STM: topology imprinted on the charge current and spin transfer torque

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The controlled creation and annihilation of individual magnetic skyrmions have been demonstrated by using spin-polarized scanning tunneling microscopy (SP-STM) [1], where the spin-polarized current exerts a torque on the spin moments of the sample. This is a promising step towards future technological applications using skyrmions for carrying or storing information. However, the detailed microscopic mechanisms of these processes are presently unknown. Our work contributes to this understanding by a theoretical investigation of the tunneling electron charge and spin transport probing magnetic skyrmions. The spin-polarized charge current (*I*) and tunneling spin transport vector quantities, the longitudinal spin current and the spin transfer torque (*STT*), are consistently calculated within a simple electron transport theory [2]. The electron tunneling model is extended to SP-STM in high spatial resolution, and applied to magnetic skyrmions [3,4]. Besides the vector spin transport characteristics, the relationships between conventional charge current SP-STM images [5], the magnitudes of the spin transport quantities [3], and the topology of various skyrmionic objects [6] are analyzed. It is also shown that at specific STM tip positions the STT efficiency (*STT/I*) can reach large values ~*h/e*. Thus, a theoretical prediction for highly efficient localized magnetic switching is also provided.



Figure 1: Real-space skyrmionic spin structures with various topological charges (Q) [6].

- [1] N. Romming et al., Science 341, 636 (2013).
- [2] K. Palotás et al., Physical Review B 94, 064434 (2016).
- [3] K. Palotás et al., Physical Review B 97, 174402 (2018).
- [4] K. Palotás, Physical Review B 98, 094409 (2018).
- [5] K. Palotás et al., Physical Review B 96, 024410 (2017).
- [6] L. Rózsa et al., Physical Review B 95, 094423 (2017).

The effects of disorder on hysteresis loops in chiral magnets

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We investigate the effect of distribution of pinning sites on the magnetization behavior in systems with Dzyaloshinskii–Moriya interaction (DMI). We consider the standard classical spin Hamiltonian with Heisenberg exchange, DMI, uniaxial anisotropy statistically varying across the lattice sites, and the Zeeman energy. The statistical variation of the anisotropy constants follows from a spatial Gaussian distribution with non-zero mean and standard deviation σ , and serves as a way to model the structural disorder in the sample. First we discuss the reduction of the model to a lattice resolved mean-field theory, and the development of energy minimisation algorithm for solving it. This approach allows to compute systematically and efficiently the magnetisation versus field, M(H), dependence for variable σ and temperature, and can be used for computing qualitative thermodynamic phase diagrams to explore material behaviour and to guide the computationally costly Monte-Carlo simulations.

We show that as the σ increases, relative to the strength of exchange interaction and DMI, the nature of the reversal modes observed along a typical M(H) hysteresis loop changes in a certain temperature window. Namely, in 'clean' systems with small σ , the reversal proceeds first through the appearance of skyrmion lattices at low external fields, while in 'dirty' systems with large σ the reversal is through the nucleation of individual or small groups of skyrmions. We systematically quantify this effect and discuss its broader implications for applications.

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Direct and inverse spin-Hall effects in germanium

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The aim of spintronics is to exploit the spin degree of freedom to add new functionalities to electronic devices and boost their performances. In this sense, spintronic studies in semiconductor-based platforms are both of fundamental and applicative interest. While it is commonly true that materials with high spin-orbit interactions show huge spin-charge interconversion efficiencies, here we demonstrate that it is possible to observe the spincharge interconversion in germanium. This parameter can also be enhanced by increasing the kinetic energy of conduction electrons in Ge.

First, we exploit the spin-Hall effect to generate a uniform pure spin current in an epitaxial n-doped Ge channel, and we detect the electrically induced spin accumulation, transverse to the injected charge current density, with polar magneto-optical Kerr microscopy at a low temperature. We show that a large spin density up to 400 μ m⁻³ can be achieved at the edges of the Ge channel for an applied electric field lower than 5 mV/ μ m. We find that the spin density linearly decreases toward the center of the Ge bar, due to the large spin diffusion length.

Then, we investigate the inverse phenomena by studying the inverse spin-Hall effect in highly doped germanium as a function of the kinetic energy of the carriers. Spin-polarized electrons are generated by the absorption of circularly-polarized light in the Ge conduction band, and their kinetic energy is varied by changing the photon energy in the 0.7–2.2 eV range. The spin detection scheme relies on spin-dependent scattering inside Ge, which yields an inverse spin-Hall electromotive force. The inferred spin-Hall angle increases by about 3 orders of magnitude within the analyzed photon energy range. Since, for increasing photon energies, the phonon contribution to spin scattering exceeds that of impurities, our result indicates that the spin-to-charge conversion mediated by phonons is much more efficient than the one mediated by impurities.

Spin to charge conversion in topological insulator Sb₂Te₃ sputtered thin films towards magnetisation switching devices

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Spintronics has seen remarkable progress in the last decades. By combining well known concepts like Giant and Tunnelling magnetoresistance with spin torques and control of domain walls, developments are foreseen in data storage and logic devices [1, 2]. New materials and structures have also allowed several breakthroughs, from 2D materials to Topological Insulators.

Topological Insulators (TI) are an exotic category of materials. Having been experimentally discovered only in 2007 [3], they have already shown great promise for a wide range of applications. Due to their performance in the control and switch of nanomagnets based on the Spin Orbit Torque (SOT), they could lead the advancements of new memory and logic devices.

One family of materials that has arisen within TIs is the Bi_2Se_3 family of semiconductors. They host a metallic state on their surface while having an insulating bulk. The metallic state is protected by time reversal symmetry and presents a spin texture. Moreover, due to their strong Spin Orbit Coupling, high values of the Spin Hall angle θ_{SH} have been reported [4], leading to outstanding control of nanomagnets. Sb_2Te_3 is one of the members of this family, with the TI surface state being theorised in 2009 [5] and later observed [6]. However, it has not been widely studied due to its smaller band gap. Thus, it is not yet clear if Sb_2Te_3 is a material of interest for SOT applications.

In this paper, the structural, electrical and magnetic properties of ion beam sputtered Sb₂Te₃ - Py thin film bilayers will be reported. We show an exotic behaviour of the Ferromagnetic Resonance (FMR) damping at low temperatures, with the damping increasing approximately 20% with cooling from 10 K to 2 K. The spin pumping experiments performed on the bilayers will be discussed and their results explored. Furthermore, the transport properties of Sb₂Te₃ and of the bilayers will be analysed. The presence of the weak antilocalisation cusps on the magnetoresistance at low temperatures and low magnetic fields is confirmed, which is consistent with the existence of the TI surface state. Considering these properties, we estimate the Sb₂Te₃ θ_{SH} to be 0.36 ± 0.02 at 300 K and evaluate the spin to charge conversion in this bilayer system. As these bilayers were made using a scalable inexpensive method, the results offer promising opportunities for future applications in spintronics.

SFT thanks FCT for PhD Grant PD/BD/141803/2018.

- [1] Grimaldi, E., et al., Nature Nanotechnology, 2020. 15(2): p. 111-117.
- [2] Manipatruni, S., et al., Nature, 2019. 565(7737): p. 35-42.
- [3] König, M., et al., Science, 2007. 318(5851): p. 766.
- [4] Wang, Y., et al., Nature Communications, 2017. 8(1): p. 1364.
- [5] Zhang, H., et al., Nature Physics, 2009. 5(6): p. 438-442.
- [6] Hsieh, D., et al., Physical Review Letters, 2009. 103(14): p. 146401.

The electric field-induced nucleation of 90-degree domain wall-magnetic domains: polarity determines chirality

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Recent reports on electric-field induced nucleation of magnetic skyrmions and bubble domains [1–5] exemplify remarkable charge-asymmetry: the nucleation is possible in the electric field of a certain polarity. The reason for it is that the domain wall surface energy depends on the electric field and this dependence is asymmetric with respect to the electric field.

In this report we will show that unipolar nucleation is not a universal rule: the 90-degrees structures can be generated at both electric polarities, while the general rule remains the same as in the case of magnetic domain bubbles with 180-degree domain walls [5], i.e. the electric polarity is related to the chirality of the domain wall.

The electric field-induced magnetic domain nucleation was observed by the magneto-optical visualization technique in 11.6µm-thick iron garnet film $(BiLu)_3(FeGa)_5O_{12}$ grown on (110) $Gd_3Ga_5O_{12}$ substrate. The high electric field ~ 1 MV/cm was generated by electrically biased scanning probe microscope cantilever tip in contact mode (tip potential V=±500 V). To enhance the magnetoelectric properties of the domain walls the constant inplane magnetic field H_{\parallel} =170 Oe was applied to the sample [1]. Additional magnetic bias field perpendicular to magnetic film H_{\perp} =16 Oe was applied to induce single domain state that is necessary to observe the magnetic bubble domain nucleation at the tip.

The electrically nucleated magnetic domains appeared aside the tip electrode: their position with respect to the tip changed from left to right upon the switching electric polarity of the tip. Thus, the sense of magnetization rotation across the domain wall nearest to the tip switched from clockwise to counterclockwise exemplifying the relation of magnetic domain wall chirality and electric polarity.



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Figure 1: The electric field-induced nucleation of the domains: the schematics of the experiment and the magnetooptical images in red frame for positevely biased tip and in blue one for negatively bias. Magnetization distribution is shown with hollow white arrows. The circle arrows represent the chirality.

- [1] D.P. Kulikova et al, JETP Lett. 104 (2016) 197–200.
- [2] P. Hsu et al, Nat. Nanotechnol. 12 (2016) 123–126.
- [3] M. Schott et al, Nano Lett. 17 (2017) 3006–3012.
- [4] P. Huang, Nano Lett. 18 (2018) 5167-5171
- [5] D.P. Kulikova et al, Phys. Status Solidi Rapid Res. Lett. 12 (2018) 1800066.

Theory of Néel-Bloch transition for compact magnetic skyrmions

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Magnetic skyrmions are a prime example of topologically non-trivial spin textures observed in a variety of magnetic materials. They emerge when the exchange and anisotropy energies promoting parallel alignment of spins in a ferromagnet enter in competition with energies favoring non-collinear alignment of spins such as the Dzyaloshinskii-Moriya interaction (DMI), the long-range dipolar interaction or higher-order exchange interactions. DMI is at the heart of a large number of magnetic skyrmion observations in recent years. This antisymmetric exchange interaction is related to the lack of structural inversion symmetry and is present in a variety of bulk chiral magnets. The basic theoretical understanding of skyrmions in ultrathin ferromagnetic layers with interfacial DMI relies on a model that accounts for the dipolar interaction through an effective anisotropy term, neglecting long-range effects. At the same time, in single ferromagnetic layers with interfacial DMI, large chiral skyrmions, also called skyrmionic bubbles have been observed, suggesting a non-trivial interplay between DMI and longrange dipolar effects [1]. The competition between these two energies also leads to the formation of skyrmions exhibiting spin rotations with intermediate angles between Neel and Bloch, a phenomenon also present in domain walls. Here we use rigorous mathematical analysis to develop a skyrmion theory that takes into account the full dipolar energy in the thin film regime and provides analytical formulas for compact skyrmion radius, rotation angle and energy [2]. We demonstrate that the DMI threshold at which a compact skyrmion loses its Néel character is a factor of 3 higher than that for a 1D domain wall. A reorientation of the skyrmion rotation angle from Néel to intermediate Néel-Bloch angles is predicted as the layer thickness is increased in the low DMI regime, which is confirmed by micromagnetic simulations. The estimation of this reorientation thickness is important for applications as the skyrmion angle affects its current-induced dynamics.

[1] A. Bernand-Mantel et al., SciPost Physics 4, 027 (2018)

[2] A. Bernand-Mantel, Cyrill B. Muratov, Thilo M. Simon, Physical Review B 101, 045416 (2020)

Quantum Depinning and Curvature effects in Magnetic Skyrmions

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We investigate the role of pinning in isolated magnetic skyrmions. In particular, we study the quantum depinning of a weakly driven skyrmion out of an impurity potential in a mesoscopic magnetic insulator [1]. The rate of tunneling, as well as the crossover temperature between thermal and quantum tunneling, are discussed in terms of macroscopic parameters of the insulator Cu_2OSeO_3 and various skyrmion radii. We demonstrate that small enough magnetic skyrmions, with a radius of ~10 lattice sites, consisting of some thousands of spins, can behave as quantum objects at low temperatures in the millikelvin regime. In addition, we demonstrate that skyrmions experience pinning potentials, generated by the varying curvature of elastically deformable geometries. In particular, we reveal that novel curvature-driven effects emerge in geometries with non-constant curvature, which explicitly break the translational invariance of flat space [2]. For a skyrmion stabilized by a curvilinear defect, an inertia term and a pinning potential are generated by the varying curvature, while both of these terms vanish in the flat-space limit.

[1] C. Psaroudaki and D. Loss, Phys. Rev. Lett. 124, 097202 (2020).

[2] A. Pavlis and C. Psaroudaki, arXiv:2003.01810
Chirality Switch of Magnetic Skyrmions in Ta/FeCoB/TaOx Trilayers

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Magnetic skyrmions [1] are promising candidates for next-generation spintronic devices. Their solitonic and chiral nature allows an efficient current-induced motion up to high velocities [2]. A versatile tuning of their chirality would enable highly-manipulable magnetic skyrmions, which paves the way for new functionalities in spintronic memory, logic and neuromorphic devices [3,4]. This gives great importance to the control of the interfacial Dzyaloshinskii-Moriya interaction (iDMI) [5,6] strength and sign, at the origin of magnetic skyrmions and their chirality.

In this study, we show by using Brillouin-Light-Scattering (BLS) a iDMI sign crossover in Ta/FeCoB/TaOx trilayer when varying either the FeCoB thickness or the TaOx oxidation level. The observed iDMI sign reversal along the oxidation gradient is understood in terms of the change of mechanism, from Fert-Levy [7,8] to Rashba [9,10] when increasing oxidation level, and is consistent with other theoretical or experimental studies [11,12].

For the first time, we show that this iDMI sign change is accompanied by a change of skyrmion chirality, as confirmed by the opposite direction of current-induced motion of magnetic skyrmions under polar-Magneto-optical Kerr effect microscopy. Magnetic skyrmions are either right-handed (iDMI>0), for optimally oxidized FeCoB/TaOx interface, or left-handed (iDMI<0), for underoxidized FeCoB/TaOx interface. This material-dependent chirality switching brings a new degree of freedom to the exciting physics of magnetic skyrmions.

Between the two regions of opposite iDMI sign we have found, as expected, a region where iDMI~0. Interestingly, in this region, skyrmions do not show any motion induced by current, which indicates that either the driving force is very small or that their strong dipolar interaction prevents their motion.

Close to this region with iDMI~0, the control of the iDMI with an electric field [13] may lead to a voltage-induced chirality switching, which hasn't been observed yet. This all-electrical control of magnetic skyrmions represents a cornerstone towards power efficient spintronic devices and multidirectional logic functionnalities.

- [1] A. N. Bogdanov and D. A. Yablonskii, J. Exp. Theor. Phys., 95, 101 (1989)
- [2] W. Jiang, et al., Science, . 349, 283 (2015) ; S. Woo et al., Nat. Mater., 15, 501 (2016)
- [3] X. Zhang, et al., J. Phys.: Condens. Matter, 32, 143001 (2020)
- [4] A. Fert, N. Reyren and V. Cros, Nat. Rev. Mater., 2, 1703 (2017)
- [5] I. E. Dzyaloshinskii, Sov. Phys. JETP, 5, 1259 (1957)
- [6] T. Moriya, Phys. Rev., 120, 91 (1960)
- [7] A. Fert and P.M. Levy, Phys. Rev. Lett., 44, 1538 (1980)
- [8] A. Fert, et al.,. Nat. Nanotechnol., 8, 152(2013)
- [9] K.-W. Kim, et al. Phys. Rev. Lett., 111, 216601 (2013)
- [10] H. Yang, et al., Sci. Rep., 8, 12356 (2018)
- [11] M. Arora, et al., Phys. Rev. B, 101, 054421 (2020)
- [12] A. Belabbes, et al. Sci. Rep., 6, 24634 (2016)
- [13] T. Srivastava, et al., NanoLett., 18, 4871 (2018)

Domains and domain walls in magnetoelastic multilayers

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Domain walls, skyrmions and other nanoscale spin structures are considered as key elements of future 3D nanodevices of information processing, including nonvolatile memory devices [1]. Interest in such structures is also due to the phenomenon of new physical effects associated with nanosize, topology and chirality of magnetic elements. In turn, magnetic nanostructures with controlled magnetoelastic properties are promising for creating information storage and processing devices with record energy consumption [2-4], up to tens of attojoules per one elementary operation to change the state of nanoelement's magnetization.

The possibility to manipulate domain walls in magnetoelastic nanostripes by means of uniform mechanical stresses has been numerically demonstrated [5]. Elastic stresses induced by the piezoelectric layer are used to change the magnetic state of the nanostripe. In this case, the domain-wall velocity of the same order of magnitude as when exposed to magnetic fields or spin-polarized currents is achieved, and energy consumption is significantly reduced. In this report we present the results of modeling and experimental study of the domains and domain-walls in magnetoelastic nanostripes. Multilayered magnetostrictive films of TbCo₂/FeCo composition were deposited onto piezoelectric PMN-PT substrates by RF sputtering under a magnetic field in order to induce a magnetic easy-axis in-plane anisotropy. Then, micro- and nanostructures of various shapes were formed in the films by lithography for experimental study of magnetization distribution, processes of magnetization reversal, formation and movement of domain walls in the films. It is shown that in the initial state in the nanostripe, depending on the prehistory of the magnetization/demagnetization of the nanostripe, there are two variants for the formation of an equilibrium magnetic structure - with asymmetric or symmetrical direction of the magnetization vectors at the edges of the nanostripe and, accordingly, with a state close to the monodomain, or split of the nanostripe into domains. The symmetry breaking of the stable magnetization states in the nanostripe can be realized by a static magnetic field applied perpendicular to the easy axis of the ferromagnetic. Further change of magnetization states allows their manipulation by means of homogeneous mechanical effects induced by the application of an electric field to the piezoelectric substrate.

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- [1] A. Fernandez-Pacheco et al. Nature Comm. 8, 15756 (2017).
- [2] N. Tiercelin et al. Appl. Phys. Lett. 99, 192507 (2011).
- [3] N. D'Souza et al. Nanotechnology 29, 442001 (2018).
- [4] M.V. Logunov et al. Bull. Russ. Acad. Sci.: Phys. 84, 196 (2020).
- [5] T. Mathurin et al. Phys. Rev. B 95, 140405(R) (2017).

Quantum Damping of Skyrmion Crystal Eigenmodes Due To Spontaneous Quasiparticle Decay

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Going Beyond Skyrmions: Alternative Magnetic Nano-Objects for Spintronics

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A novel route towards multi-level magnetic random access memory

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Magnetic random access memory (MRAM) attracts considerable attention for its non-volatility, fast read and write operations, high endurance and long retention. However, in order to remain relevant in coping with the accelerated rate at which humanity generates data, it becomes increasingly important to increase its density. Currently, the basic memory cell of MRAM is based on magnetic tunnel junctions (MTJs) where each MTJ can be stabilized in two states. Here, we show that, by using shape anisotropy, MTJs can be stabilized in many more states, thus paving the way for multi-level MRAM which is expected to be much denser than currently available MRAM.

We fabricate structures consisting of 'N' crossing ellipses, where N = 1, 2, 3 and 4, which exhibit uni-axial, bi-axial, tri-axial and quadro-axial magnetic anisotropy, respectively, in the overlap area [1-2], and we show that by inducing spin-orbit torques (SOTs), generated by flowing current in an adjacent heavy metal layers, up to 2^{2N} discrete remanent states can be stabilized [3]. Considering such large number of states per structures and the ability to switch between them in absence of external magnetic field makes such structures extremely viable for various spintronics applications such as multi-level MRAM and neuromorphic computing etc. To demonstrate the possibility to realize multi-level MRAM based on such structures, we have fabricated MTJs consisting of two-crossing ellipses and a single ellipse as ferromagnetic layers, which shows four distinct states corresponding to four remanent states of two-crossing ellipses and successfully switches between the states by SOTs [4].

- [1] Shubhankar Das el al., Scientific Reports 8, 15160 (2018).
- [2] Shubhankar Das el al., Scientific Reports 9, 20368 (2019).
- [3] Shubhankar Das el al., Manuscript submitted in Appl. Phys. Lett.
- [4] Shubhankar Das et al., Manuscript under preparation.

B20-type MnSi films on Si (111) grown by flash lamp annealing

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B20-type MnSi is one of the noncentrosymmetric materials hosting magnetic skyrmions, which are promising information carriers in spintronic devices. In this work, we report the preparation of (111)-textured MnSi films on Si substrates. The preparation method only includes the deposition of Mn layers at room temperature and a following process by flash lamp at milli-second. By controlling the thickness of Mn layers and flash energy, we can obtain pure MnSi or mixtures of MnSi and MnSi_{1.7}. Surprisingly, all prepared films show Curie temperatures around 41 K, which is much higher than for bulk MnSi or films reported previously. Magnetic skyrmions are stabilized at the whole temperature range below 41 K and under a much wider magnetic-field range. We speculate that the increased Curie temperature is due to the strain in the MnSi films arising from the MnSi_{1.7} phase or from the substrate. Our work calls a re-examination of the structural and magnetic properties of *B20* MnSi films.

Synthesis, preparation and transport analysis of topological insulators $BiSbTeSe_2$ and $BiSbTe_2S$

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Topological insulators exhibit an inverted bulk gap for electronic excitations induced by strong spin-orbit coupling. They represent quantum state of matter which is characterized by special edge or surface states. We have grown single crystals of topological insulator material $BiSbTeSe_2$ and $BiSbTe_2S$. These materials belong to Bi_2Te_3 compound family famous for their thermoelectric properties and selected compositions are chosen for their good insulator behavior. Procedure of growing crystals is shown with details, as is procedure for making 10 μ m thick samples. Structural analysis of grown material has been made and materials don't show signs of unwanted phases.

Prepared samples are characterized by electric and thermoelectric measurements for temperaures below 300 K. Electrical measurements are made with and without strong magnetic field up to 15 T. One can observe temperature at which conductive surface states start playing bigger role in transport from resistivity measurement. Changes at those temperatures can be also observed in Seebeck coefficient. Recorded Shubnikov-de Haas oscilations show quality of samples and interesting behaviour of frequency to temperature dependance is found.

Unveiling the oxygen effect on spin-orbit torque in Pt/Co bilayers via in-situ oxygen exposure and electrical measurement

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Despite oxygen incorporation in heavy metal (HM)/ ferromagnet (FM) heterostructures has been reported to be one of the possible routes to enhance the spin-orbit torque (SOT) efficiency for magnetization manipulation^{1,2}, there has not been a consensus on the exact role of oxygen due to the difficulty in pinpointing the real location of oxygen and studying the oxygen effect on a same sample. In view of this, we systematically investigated the oxygen exposure effect on SOT in Pt/Co bilayer via conducting in-situ Co, Mg deposition, precisely controlled oxygen dose and electrical measurement in ultrahigh vacuum system without exposing the sample to ambience. The second harmonic Hall voltage measurement was performed to characterize the SOTs. The insets of Fig. 1(a) and 1(b) show the second harmonic voltage of Pt (2)/Co (0.8) as a function of an applied field of H_x and H_{y_x} respectively, which display a good linearity. By extracting the SOTs via using the relationship of first and second harmonic voltage, we obtained the damping-like (DL) and field-like (FL) SOT effective field H^{DL} and H^{FL} at varied applied current density. As shown in Fig. 1(a) and 1(b), the SOTs increase linearly as the increase of current density. Based on the extracted H^{DL} and H^{FL} at varied current density, we estimated the DL and FL SOT efficiency ξ^{DL} and ξ^{FL} at different oxygen dose from 0 L to 300 L as plotted in Fig. 1(c). As can be seen, both ξ^{DL} and ξ^{FL} experience initial fast increase from 0 L to 50 L, after which the increase slows down and the values gradually saturate. The observed increase of SOTs after oxygen incorporation is consistent with the reported observations which attribute the increase to enhanced Rashba-Edelstein effect (REE) at FM/oxide interface or more efficient spin current transmission at Pt/Co interface^{1,2}. However, in our case the oxygen is directly added atop the Co layer without any capping layer, and the oxygen atoms are not possible to diffuse to the Pt/Co interface. Therefore, the most possible mechanism behind the enhancement may be the modification of surface electric field on the Co surface, which leads to large Rashba splitting and thus enhanced SOTs due to REE^{3,4}. By performing ab initio calculations, we found that the Rashba splitting is weak in Pt/Co bilayer, while with 1 ML oxygen covering Co surface the Rashba splitting is evidently increased, which indicates the important role of oxygen in modifying the surface state of FM layer. We further deposited a Mg dusting layer (~ 0.2 nm) and subsequently a 1 nm MgO layer on Pt/Co/O to shed more light on the oxygen effect. The SOT efficiencies for all the structures are shown in Fig. 1(d), from which we can find that SOTs decrease after deposition of the Mg dusting layer and increase again after the MgO layer is deposited. The former can be explained by the decreased surface electric field due to charge compensation of Co electrons from Mg atoms, while the latter is attributed to the REE arising from the Co/oxide interface.



Figure 1: (a) H^{DL} and (b) H^{FL} as a function of J at different oxygen dose. Inset: second harmonic Hall voltages with (a) H_x and (b) H_y . (c) ξ^{DL} and ξ^{FL} versus oxygen dose. (d) ξ^{DL} and ξ^{FL} of different structures.

- [1] Hasegawa, K., et al. Phys. Rev. B 98, 020405 (2018).
- [2] Emori, S., et al. Appl. Phys. Lett. 105, 222401 (2014).
- [3] Xie, H., et al. Sci. Rep. 9, 17254 (2019).
- [4] Krupin, O. et al. Phys. Rev. B 71, 201403 (2005).

Observation of elliptical Bloch skyrmion and antiskyrmion in a D2d system

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Magnetic skyrmions and antiskyrmions are topologically protected nanometer-size non-collinear spin textures. They are characterized by the opposite topological charge of ±1 and have distinct chiral magnetic boundaries [1]. Their existence in a material system mostly depends on the underlying symmetry of the crystal structure [1-5]. Chiral cubic B20 compound, for example, MnSi [2], FeGe [3] have the isotropic spin texture of Bloch skyrmion whereas anti-skyrmion have anisotropic spin structure of alternating helicoids and cycloids chiral boundary [4, 5]. Here, using Lorentz transmission electron microscopy (LTEM), we found that both lattices and even spare arrays of individual Bloch skyrmions and antiskyrmions can be stabilized in a tetragonal inverse Heusler compound of $Mn_{1.4}Pt_{0.9}Pd_{0.1}Sn$ with D_{2d} symmetry, whose Dzyaloshinskii-Moriya interaction (DMI) otherwise supports antiskyrmions [6]. Our finding of opposite topological charges of magnetic non-collinear spin textures in a D_{2d} Heusler compound of $Mn_{1.4}Pt_{0.9}Pd_{0.1}Sn$ shows that D_{2d} material has greater functionalities as compared to, for example, B20 materials.

- [1] A. Bogdanov et al., Phys. Rev. B, 66, 214410 (2002)
- [2] S. Mühlbauer et al., Science, 323, 915-919 (2009)
- [3] X. Z. Yu et al., Nat. Mater., 10, 106 (2011)
- [4] A. K. Nayak et al., Nature, 548, 561(2017)
- [5] J. Jena et al., Nano Lett., 20, 59-65 (2019)
- [6] J. Jena et al., Nat. Commun., 11, 1115 (2020)

In-situ Control of Anisotropy for Improved Spin Torque Switching of a Magnetic Thin Film via Helium Ion Irradiation

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Creation of a Skyrmion phase in Ta/CoFeB/MgO Thin Films by FIB Irradiation

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Skyrmions, chiral magnetic spin textures, are proposed for future ultra-high density storage and logic devices. Sputtered Ta/CoFeB/MgO thin films are promising for such applications due to the industry-approved materials that offer ultra-low depinning fields below 1 mT. However, controlling the material parameters by varying the individual layer thicknesses and stack composition is tedious and prone to process variations. Therefore finding the process window for the stabilization of a room temperature Skyrmion phase is a challenging task. Non-destructive Ga+ focused ion beam (FIB) irradiation enables to fine-tune the magnetic properties on a very local scale (potentially sub 10 nm). It leads to short-range intermixing of materials. This makes it possible to easily access effective anisotropy K_{eff} , saturation magnetization M_s , and the DMI constant D, even though not independent of each other. In contrast to annealing, ion irradiation favors an anisotropic diffusion of the materials in the direction of the ion beam, leading to significantly different effects. This precise control, with high lateral resolution, makes it an auspicious alternative to conventional "stack engineering" and annealing.

In this work, we investigate the effects of ion beam irradiation on the magnetic parameters of Ta/CoFeB/MgO thin films. Widefield magneto-optical Kerr-effect (MOKE) measurements are employed to study the effects of different ion irradiation doses on the domain shape, structure and size and the according depinning and nucleation fields. These measurements are performed on film level and on patterned films with varying oop magnetic fields. Furthermore, these measurements are complemented by anomalous Hall effect and polar laser MOKE measurements to get further insight into the coercive fields, on film level and local scale. Additional SQUID measurements are performed to evaluate M_s and K_{eff} of the irradiated samples.

As a second step, we elaborate on the possibility to fine-tune the thin film parameters to host Skyrmions. Using this technique, we present a way to define regions where Skyrmions exist at room temperature in a film that, in its virgin state, is not able to host such spin structures. We show that herefore both, areal as well as local irradiation, creating magnetic potential wells, can be utilized. Beyond that, we will demonstrate that even the bubble density and size can be engineered by using FIB irradiation, as shown in Fig 1. This opens the door for local creation of Skyrmion tracks, nucleation sites, and hopefully logic gates.



0 lons/cm² 1e12 lons/cm²

m² 5e12 lons/cm² 1.2e13 lons/cm² 1.6e13 lons/cm² 2.2e13 lons/cm²

Figure 1: WMOKE images of different areas of the same Ta/CoFeB(1.5nm)/MgO film after irradiation with varying doses. For no irradiation already skyrmion development can be observed in a relaxed state. The number of skyrmions increases up to the dose 5e12 lons/cm² while the size decreases. Beyond that value size of the skyrmions increases again while the density is reduced up to the point where the whole irradiated area shows a uniform magnetization direction.

[1] I.M. Miron, K. Garello, G. Gaudin, P.-J. Zermatten, M.V. Costache, S. Auffret, S. Bandiera, B. Rodmacq, A. Schuhl, and P. Gambardella, Nature 476, 189 (2011).

[2] R. Ramaswamy, J.M. Lee, K. Cai, and H. Yang, Appl. Phys. Rev. 5, 031107 (2018).

[3] S. Woo, M. Mann, A.J. Tan, L. Caretta, and G.S.D. Beach, Appl. Phys. Lett. 105, 212404 (2014).

[4] C. Chappert, H. Bernas, J. Ferré, V. Kottler, J.-P. Jamet, Y. Chen, E. Cambril, T. Devolder, F. Rousseaux, V. Mathet, and H. Launois, Science 280, 1919 (1998).

Antiferromagnetically coupled anti-phase domains under external magnetic fields

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Ni₂MnZ based Heusler compounds have attracted a considerable amount of attention due to their various appealing properties such as the ferromagnetic shape-memory effect or the magnetocaloric effect. The room temperature ground state structure is L2₁ order, while at higher temperature a state of increased entropy is preferred, corresponding to B2 order. Quenching a crystal from the B2 stable regime preserves its disordered structure while going to low temperatures, where atomic diffusion is inactive. During subsequent annealing, L2₁-ordered domains nucleate independently and grow, leading to a division of the crystal into anti-phase domains (APDs) [1].

The magnetic moments in these systems are mainly carried by Mn atoms with neighbouring spins interacting antiferromagnetically and the spins of next-nearest neighbour Mn atoms interacting ferromagnetically. Figure 1 shows the magnetisation curves of a quenched Ni₂MnAl_{0.5}Ga_{0.5} bulk sample after successive annealing steps. As the annealing time increases the magnetisation rises indicating a transition from an antiferromagnet to a ferromagnet. In small-angle neutron scattering (SANS) measurements on Ni₂MnAl_{0.5}Ga_{0.5} powder samples in distinct ordering states we observed a positive correlation between structural order and magnetic domain sizes with their characteristic length scales being of the same size. Using TEM and Lorentz TEM a reversal of magnetization at structural anti-phase domains has been observed in Ni₂MnAl_{0.5}Ga_{0.5} implying that structural and magnetic domains are identical [1]. The magnetic properties are therefore very sensitive to the degree of structural order in these systems [2].

To clarify the reason of this relation and to study the mechanism of coupling of ferromagnetic domains across APD boundaries, we have investigated Ni₂ MnAl and Ni₂MnAl_{0.5}Ga_{0.5} powder samples in distinct ordering states via temperature-dependent magnetic SANS, giving access to the magnetization microstructure. With an external magnetic field applied we observe a redistribution of intensity along the field direction, corresponding to the ferromagnetic domains coupling antiferromagnetically. Approaching the magnetic transition temperature of our samples with an external magnetic field applied the predominant spin orientation changes, turning them from perpendicular into parallel alignment to the B – field. We observe the transition from antiferromagnetically coupled APDs into a field polarized state.



Figure 1: Temperature-dependent magnetisation curves with a magnetic of H = 10 kOe applied of a Ni₂MnAl_{0.5}Ga_{0.5} bulk sample after successive annealing steps.

^[1] H. Ishikawa et al., Acta Mater. 56, 4789 (2008).

^[2] P. Neibecker et al., Appl. Phys. Lett. 105, 261904 (2014).

Spin-Hall magneto-resistance in Pt/Ga:YIG prepared by the metallic organic decomposition method

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Magneto-optical (MO) spatial light modulators for holography 3D display device [1] have been developed, which magnetization direction is controlled by spin polarized current injection. Magnetic insulators such as Garnet have large magneto-optical effect and are very attractive materials for light modulator application to get better light modulation efficiency, although it had been difficult to control its magnetization by current injection due to its large electrical resistance. Electrical transport properties in the Pt grown on a garnet film was affected by magnetization direction of the garnet, which is known for spin Hall magneto-resistance (SMR) [2]. Recently, spin orbit torque (SOT) induced by current flow in Pt layer succeeded to switch the garnet magnetization [3]. However, those garnet materials are mostly fabricated by vacuum process like pulsed laser deposition or sputtering, which methods are not very appropriate for large area with low cost. On the other hand, metal organic decomposition (MOD) is very attractive method for large area with low cost, and relatively easy to control its composition and control magnetic anisotropy [4, 5]. However, there is not much research about SMR of garnet grown by the MOD method. The SMR evaluation is very important because it is proportional to the SOT to switch the magnetization of the garnet. In this paper, we show SMR properties of Pt/Y₃Fe₄GaO₁₂ (Ga:YIG) bilayer which garnet was grown by the MOD method.

Ga:YIG film was prepared on (111) oriented $Gd_3Ga_5O_{12}$ (GGG) substrate by MOD method with subsequent 800°C annealing to promote crystallization. XRD analysis showed that the Ga:YIG was heteroepitaxially grown on the GGG. Perpendicular magnetic anisotropy was confirmed by a Faraday hysteresis loop, which is attributed to reduction of its magnetization due to a substitution of Fe to Ga and changes in magneto-elastic energy [5].

Pt Hall strips (an inset of fig.1) were fabricated on the Ga:YIG by a lift-off process using optical lithography. The strip was 74 µm long with 4 µm wide having a Hall cross bar at the center. We defined xyz-coordinate as an inset of fig.1, x-direction as current direction. Longitudinal resistance (R_{xx}) was measured by DC current of 50 µA using 4-point probe with magnetic field of 200 mT rotating y-z plane from the plus y-direction (angle ϕ =0) to minus y-direction (ϕ =180). As shown in the fig. 1 the resistance increased with an increase in the angle ϕ up to 90 degree and decreased to the low resistance again. Longitudinal SMR ($\Delta R_{xx}/R_0$) was about 0.037% and followed sin² ϕ very well which was typical SMR loop with perpendicular magnetic anisotropy [3]. The SMR was about 30-50% smaller value compared to the garnet fabricated by the vacuum process [2,3]. The reason for the smaller SMR is unclear, but contamination between Pt and the Ga:YIG could reduce the spin injection from Pt to the Ga:YIG. Since no treatment was applied before the Pt deposition, the Ga:YIG surface is likely to be covered with hydro-carbon contamination in the air. We clearly clarified the spin injection into the MOD grown Ga:YIG from Pt which may have potential to manipulate the Ga:YIG magnetization.



[1] K. Aoshima, et al., J. Disp. Technol., Vol. 12, p. 1212 (2016)

- [4] T. Ishibashi, et al., J. Appl. Phys., 97, 013516 (2005)
- [5] M. Sasaki, et al., Jpn. J. Appl. Phys., 55, 055501 (2016)

^[2] H. Nakayama, et al., Phys. Rev. Lett., 110, 206601 (2013)

^[3] C. Avci, et al., Nature Mat. 16, 309 (2017)

Skyrmion Lattice Phases in Thin Film Multilayers

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Phases of matter are ubiquitous with everyday examples including solids and liquids. In reduced dimensions, particular phases, such as the two-dimensional (2D) hexatic phase and corresponding phase transitions occur [1-4]. A particularly exciting example of 2D ordered systems are skyrmion lattices [5], where in contrast to previously studied 2D colloid systems [6], the skyrmion size and density can be tuned by temperature and magnetic field [7].

In this work we use a skyrmion lattice to study phase transitions in 2D. We first show that we can drive the system from a liquid phase to a hexatic phase as deduced from the analysis of the hexatic order highlighting the 2D nature of the system. Using coarse-grained molecular dynamics simulations of soft disks, we determine the skyrmion interaction potentials and we find that the simulations are able to reproduce the full two-dimensional phase behaviour. This shows that not only the static behaviour of skyrmions is qualitatively well described in terms of a simple two-dimensional model system but skyrmion lattices are versatile two-dimensional model systems that allow for studying phases and phase transitions in reduced dimensions.

- [1] S. C. Kapfer, W. Krauth, Phys. Rev. Lett. 114, 035702 (2015).
- [2] Y. Nishikawa, K. Hukushima, W. Krauth, Phys. Rev. B 99, 064435 (2019).
- [3] M. Engel, J. A. Anderson, S. C. Glotzer, et al., Phys. Rev. E 87, 042134 (2013).
- [4] E. P. Bernard; W. Krauth, Phys. Rev. Lett. 107, 155704 (2011).
- [5] A. Fert, N. Reyren, V. Cros, Nat. Rev. Mater. 2, 17031 (2017).
- [6] K. Zahn, R. Lenke, G. Maret, Phys. Rev. Lett. 82, 2721–2724 (1999).
- [7] K. Everschor-Sitte, J. Masell, R. M. Reeve, M. Kläui, J. Appl. Phys. 124, 240901 (2018).

Spin Orbit driven effects and Thermal Activation of Ferromagnet Intercalated Graphene-Heavy Metal Interfaces

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The development of room temperature magnetic devices exploiting Spin-Orbit effects is at the forefront of actual research. A major challenge for future spintronics is to develop suitable spin transport channels with superior properties such as long spin lifetime and propagation length. Graphene (Gr) can meet these requirements, even at room temperature [1]. However, the development of all-graphene spintronic devices requires that, in addition to its passive capability to transmit spins over long distances, other active properties are incorporated to graphene. The generation of long-range magnetic order and spin filtering in Gr has been recently achieved by molecular functionalization [2,3] as well as by the introduction of giant spin-orbit coupling (SOC) in the electronic bands of Gr [4]. On the other side, taking advantage of the fast motion of perpendicular magnetic anisotropy (PMA) chiral spin textures, i.e., Néel-type domain walls (DWs) and magnetic skyrmions, can satisfy the demands for high-density data storage, low power consumption and high processing speed.

Here, we report on high quality, epitaxial Gr/Co(111)/heavy-metals (HM) (111)-oriented stacks grown on insulating oxide crystals, characterized by XPS, LEED, STEM, Kerr Magnetometrry and Microscopy, XAS-XMCD, and SP-ARPES, which exhibit enhanced PMA for Co layers up to 4 nm thick and left-handed Néel-type chiral DWs stabilized by interfacial Dzyaloshinskii-Moriya interaction (DMI) localized at both Gr/Co and Co/HM interfaces with opposite sign [5]. While the DMI at Co/Pt side is due to the intrinsic SOC [6], the sizeable DMI experimentally found at the Gr/Co interface has Rashba origin [5]. The active magnetic texture is protected by the graphene monolayer and stable at 300 K in air, and since it is grown on an insulating substrate, amenable to transport measurements. In addition, our XPS and STM demonstrate that Co atoms evaporated on top of Gr arrange in 3D clusters and, upon thermal annealing, penetrate through and diffuse below Gr in a 2D fashion [7]. The complete intercalation of the metal occurs at specific temperatures, depending on the type of metallic buffer. The activation energy and the optimum temperature for the intercalation processes are determined. We describe a reliable method to fabricate and characterize in situ high-quality Gr-FM/HM heterostructures, enabling the realization of novel spinorbitronic devices that exploit the extraordinary properties of Gr.



Figure 1: Sketch of the interplay between Spin-Orbit Coupling-induced DMI at the Co/Pt and opposite Rashbatype DMI at the Gr/Co interfaces [5].



Figure 2: The thermally activated Co intercalation underneath Graphene occurs in two steps. The intercalated atoms diffuse then in 2D underneath Gr and form flat, homogeneous and crystalline layers [7].

[1] W. Han, R.K. Kawakami, M. Gmitra, et al. Nat. Nanotech., 9, 794 (2014).

- [2] M. Garnica, D. Stradi, S. Barja, et al. Nature Physics., 9, 368–374 (2013).
- [3] D. Maccariello, M. Garnica, M.A. Niño, et al. Chemistry of Materials., 26 (9), 2883-2890 (2014).
- [4] F. Calleja, H. Ochoa, M. Garnica, et al. Nature Physics., 11, 43–47 (2015).
- [5] F. Ajejas, A. Gudin, R. Guerrero, et al. Nano Letters., 18(9), 5364-5372 (2018).
- [6] F. Ajejas, V. Křižáková, D. de Souza Chaves, et al. Applied. Physics Letters., 111(20), 202402 (2017).
- [7] F. Ajejas, A. Anadon, A. Gudin, et al. ACS Applied Materials Interfaces. (2019).

Collective magnetic behaviour of metallic nanowire arrays

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Nanomagnetism has been traditionally focused on the study of patterned planar structures but, in the last years, it is expanding into three dimensions, triggered by the discovery of new magnetic effects and their potential use in novel applications [1]. Cylindrical electrodeposited nanowires are playing a key role in this expansion. The change from flat structures to cylindrical nanoobjects brings the emergence of novel spin textures directly linked to the cylindrical geometries [2] and overcomes different problems related to applications like the limitations imposed by the Walker Breakdown to the limitation of domain walls by electric currents [3]. In addition, the possibility of nanostructuring the nanowires introduce an additional degree of freedom, producing magnetic tridimensional structures and making new physics to appear and to exploit. Up to now, most of the fundamental studies have been carried out in individual nanowires. However, the most promising applications in data storage, energy storage and biomedical applications are based on compact arrays of long, nanostructured nanowires. Therefore, more work is needed to understand the collective magnetic behaviour of arrays on nanowires to move from fundamental studies to emerging applications.

In a recent work [4] we show that the introduction of local changes in composition (chemical barriers) in permalloy nanowires dramatically affect the domain structure and the magnetization processes of the nanostructures, giving rise to the formation of different types of domain walls, one of them protected under the application of magnetic fields. In this work we study the magnetic behaviour of arrays of $Fe_{20}Ni_{80}$ nanowires with two types of chemical barriers: ferromagnetic iron rich $Fe_{80}Ni_{20}$ barriers and non-magnetic Cu barriers. We have modified different geometrical parameters (diameter, distance between barriers...) keeping fixed the density of nanowires. Using magnetic vibrating sample (VSM) and magneto-optical Kerr effect (MOKE) spectroscopies, we have been able to study the vectorial magnetic behaviour and to correlate it with the geometrical and chemical characteristics of the nanowires. We have found out that the magnetic behaviour of arrays of nanowires depends on the structure of the nanowires and it can be tailored (see Figure 1) increasing the coercivity field and the remanence by the introduction of chemical defects.

These systems are very interesting because, as we will show could be the building blocks for domain-wall based devices – like race-track memories. In addition, these systems can be used as electrodes and magnetrodes for the interaction with neural systems, as we are starting to study in the framework of the ByAxon FETOPEN project.

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Figure 1: Hysteresis loops measured by Magneto-Optical Kerr effect in different arrays of nanowires. In all cases, the magnetic field has been applied perpendicular to the nanowire axis.

- [1] A. Fernandez-Pacheco et al. Nat. Commun 8, 15756 (2017).
- [2] J. A. Fernandez-Roldan et al. Nanoscale 10, 5923 (2018).
- [3] M. Schöbitz et al. Phys. Rev. Lett. 123, 217201 (2019).
- [4] S. Ruiz-Gómez et al. Sci. Rep. (2018) 16695.

Magnetoresistance measurements of the nanostructured thin layers of ferromagnetic semiconductor (Ga,Mn)(Bi,As) in the presence of domain walls

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An interplay between the strain-induced crystalline magnetic anisotropy and a shape-dependent anisotropy, introduced via the lithography process, may lead to a complex domain pattern in magnetic semiconductors [1]. Such a model allows for more in-depth investigations of the magnetization behavior and might be implemented as an alternative concept of magnetic memory [2]. An imparity in the resistances along various areas in the material due to higher scattering in artificially created domain pattern can be treated as an additional degree of freedom to store the information. Writing and accessing the information could be realized with an applied magnetic field or a spin-polarized current. In the configuration, presented at *Fig. 1*, higher values of the resistance are obtained along (3)-(4) contact pairs comparing to (1)-(2) because of the more prominent spin misalignment between the middle section governed by the crystalline anisotropy and the stripe section with constrained spin direction via the pattern-induced anisotropy. Correspondingly, state of low resistance could be associated with "0", while high resistance – with "1" in a binary coding system.

Our material of choice to investigate the magnetoresistance within the proposed model is (Ga,Mn)As – III-V ferromagnetic semiconductor with a well-developed theoretical and experimental background [3]. Promising results were shown when a few percent of Bi atoms were added to the structure, substituting As in a host zincblende lattice. An increase of spin-orbit coupling, as a result of heavy metal doping, enhances the resistance difference between contact pairs, and improves reliability of the information recognition. Thin layers of (Ga,Mn)As/GaAs and (Ga,Mn)(Bi,As)/GaAs (6% Mn, 1% Bi, d = 10...100 nm) were grown by means of low-temperature molecular beam epitaxy (LT-MBE). Series of magnetoresistance analysis were performed on similar quaternary and ternary nanostructured thin films. Magnetization behavior and transition temperatures were evaluated via the superconducting quantum interference device magnetometry, while the homogeneity of the ferromagnetic order was confirmed with a low-energy muon spin-relaxation spectroscopy [4]. Obtained results prove stability and utility of such a magnetic memory unit.



Figure 1: Scheme of the central part of the nanostructure shown under external magnetic field *H*, applied at α_H =25°. Angle α_H defines the orientation of in-plane magnetic field *H* with respect to the easy magnetization direction in the middle section. Thick arrows symbolize the magnetization vectors of individual ferromagnetic domain and boxed numbers correspond to current contacts. Magnetoresistance measurements are performed along two pair of contacts, (1)-(2) and (3)-(4), while sweeping applied magnetic field.

- [1] S. Hümpfner et al., Appl. Phys. Lett. 90, 102102 (2007)
- [2] T. Wosinski et al., Physica E 51, 128 (2013)
- [3] T. Dietl and H. Ohno, Rev. Mod. Phys. 86, 187 (2014)
- [4] K. Levchenko et al., Scientific Reports 9 (2019)

Novel magnetic textures induced by local non homogeneous magnetic field

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Magnetic vortices, domain walls, or a uniform in-plane or out-of-plane 'macrospin' are magnetic configurations that can be found as the naturally forming ground state in magnetic systems. These magnetic configurations can be integrated as the free layer of a magnetic tunnel junction (MTJ) stack, which is the main building block of magnetic memories, magnetic field sensing or high frequency spin-torque oscillators. However, novel magnetic configurations which would not be naturally formed may be explored by control of the magnetic properties and stack composition. The introduction of a heavy metal at an interface can give rise to a new spin-orbit related effect, known as the Dzyaloshinskii-Moriya (DM) interaction. This antisymmetric exchange may induce a spin canting which leads to the generation of new exotic magnetic textures, such as chiral domain walls, spin spirals and skyrmions. Most of the research focuses in maximizing the DM interaction between heavy metal / ferromagnet [1]. Recently, it has been demonstrated that some textures are obtained by a non-homogeneous perpendicular stray field [2]. The perpendicular stray field is generated by a perpendicular magnetic anisotropy (PMA). From micromagnetic simulations, we demonstrate that novel magnetic textures are excited by the nonuniform perpendicular stray field generated by Co/Ni multilayers on a free layer. The same behavior is observed when a thick NiFe layer is used, where a non-homogeneous stray field is provided by the core of a vortex. In the first case, the vortex state can be controlled by changing the thickness of the free layer, while for the second one, by modifying the pillar diameter. The non-homogeneous perpendicular magnetic field can have two important effects, namely the stabilization of magnetic vortices in small diameter (d<100 nm) nanopillars which would not otherwise have a magnetic vortex as the ground state, or the stabilisation of genuinely novel magnetic textures such as a radial vortex or room temperature skyrmions. In order to investigate the dynamic properties of these novel magnetic configurations, we propose a fully electrical characterization of the magnetic properties, by initially integrating our Co /Ni PMA layer into a magnetic tunnel junction.

[1] O. Boulle, J. Vogel, H. Yang et al. Nature Nanotech 2016, 11, 449–454.

[2] R. V. Verba, D. Navas, A. Hierro-Rodriguez et al. Phys. Rev. Applied 2018, 10, 031002.

Angular dependence of the magnetization process in Fe_{77.5}Si_{7.5}B₁₅ amorphous glasscoated nanowires: a comparative analysis between a single nanowire and an array of nanowires

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The aim of this work is to understand the angular dependence of the magnetic properties in case of highly magnetostrictive amorphous glass-coated nanowires by micromagnetic simulations. Previous investigations, such as the study of the angular dependence of magnetic properties in Permalloy nanowires, offered valuable information on the magnetization rotation mechanisms in such materials [1].

We analyzed two different systems: a single nanowire and an array of identical nanowires with cylindrical symmetry. The composition of the investigated nanowire is Fe_{77.5}Si_{7.5}B₁₅. The samples were 30 nm long and 10 nm in diameter. The magnetization process depends on the angle at which the external magnetic field is applied.

Micromagnetic simulations show that both coercivity and remanence decrease monotonously as the angle at which the external field is applied, increases.

Moreover, the comparison between the simulation of a single nanowire and that of an array of nanowires emphasizes that the magnetostatic interactions between the nanowires strongly influences not only the shape of the hysteresis curve but also the coercivity and remanence values.

Micromagnetic simulations of an array of identical nanowires reveal an important fact: nanowires reverse individually or in pairs and not all simultaneously, producing the presence of striking jumps observed in the hysteresis curve (see figure 1). Also, our results show that the nanowires reverse their magnetization through the nucleation and propagation of transverse domain walls.

The higher degree of symmetry of these rapidly solidified nanowires are expected to lead to significant advances in the velocity and mobility of the propagating walls, since these materials allow various optimization routes through their structure, dimensions and composition.

Index Terms - cylindrical amorphous nanowires, coercive field, hysteresis loop, micromagnetic simulations.

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Figure 1: Angular dependence of normalized hysteresis curves of Fe_{77.5}Si_{7.5}B₁₅ nanowires with L=30 nm and d=10 nm for a) an isolated nanowire and b) an array of sixteen nanowires

[1] S. Raviolo, A. Pereira, D. M. Arciniegas Jaimes, J. Escrig, and N. Bajales, JMMM 499 (2020) 166240.

Tuning the domain wall velocity in amorphous glass-coated nanowires and submicron wires

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Metallic amorphous glass-coated nanowires and submicron wires with diameters between 100 and 950 nm have been prepared by means of rapid quenching from the melt using an improved variant of the glass-coated melt spinning method [1]. They are embedded in an insulating glass layer, the entire composite being prepared in a single step. The coating can be employed to facilitate the manipulation of the ultrathin wires, or for biocompatibility reasons when required, but it can also be partially or fully removed, if necessary. When such samples are prepared from ferromagnetic alloys, e.g. $Fe_{77.5}Si_{7.5}B_{15}$ and $(Co_{0.94}Fe_{0.06})_{72.5}Si_{12.5}B_{15}$, they exhibit a bistable magnetic behavior, characterized by a rectangular hysteresis loop. Bistability occurs irrespective of the value of the magnetostriction constant (25 x 10^{-6} for $Fe_{77.5}Si_{7.5}B_{15}$ and $-1 x 10^{-7}$ for $(Co_{0.94}Fe_{0.06})_{72.5}Si_{12.5}B_{15}$).

Magnetization reversal takes place through the displacement of a 180° domain wall along the wire axis when the applied magnetic field reaches a specific threshold value, called switching field. Its velocity typically reaches well over 1000 m/s, being among the highest measured in any type of magnetic wire. The switching field is influenced by the value of the magnetostriction constant.

Here we report on the role of stress relief annealing on domain wall velocity and magnetic hysteresis in rapidly quenched amorphous nanowires and submicron wires with the above-mentioned compositions.

The samples have been furnace annealed in vacuum at low temperatures, i.e. 200°C and 350°C, for 1 hour, to partially relieve internal stresses induced during rapid quenching, but avoiding crystallization. The field dependence of domain wall velocity and the magnetic hysteresis loops have been measured using techniques developed specifically for such thin samples.

For instance, in the case of a nearly zero magnetostrictive $(Co_{0.94}Fe_{0.06})_{72.5}Si_{12.5}B_{15}$ amorphous submicron wire with 770 nm in diameter, the domain wall velocity decreases after annealing, from over 4500 m/s in the as-cast state, down to 2500 m/s after annealing at 350°C. In case of $Fe_{77.5}Si_{7.5}B_{15}$ samples, an opposite effect was observed: domain wall velocity increases with annealing. On the other hand, the switching field takes much lower values in $(Co_{0.94}Fe_{0.06})_{72.5}Si_{12.5}B_{15}$ samples than in $Fe_{77.5}Si_{7.5}B_{15}$ ones.

These results have been explained by considering the dominant type of magnetic anisotropy and the effect of stress relief for each composition. The magnitude of quenched-in mechanical stresses and their dependence on sample diameter are also relevant in the interpretation. Hence, stress relief annealing offers an accurate way of tailoring the domain wall velocity and hysteresis characteristics in amorphous cylindrical nanowires and submicron wires, which is important for their potential applications in domain wall motion-based devices.

Acknowledgements: Work supported by the Nucleu Programme (Project PN 19 28 01 01) and Contract No. 11PFE/16.10.2018 financed by the Romanian Ministry of Education and Research.

[1] T.-A. Óvári et al. (2015), in Magnetic Nano- and Microwires (ed. M. Vázquez), Woodhead Publishing, Cambridge, 199-223.

Synthesis and magnetic studies of low-doped Co₇Zn₇Mn₆, a ß-Mn-type alloy

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The β -Mn type alloys have recently attracted much interest due to the observation of skyrmionic magnetic phases [1]. Skyrmions are topological spin textures observed in certain chiral magnets. A skyrmion is a swirling spin structure carrying a topological quantum number that occurs due to the competition between the ferromagnetic exchange and the Dzyaloshinskii-Moriya interaction.

Skyrmions were first observed in MnSi in 2009 and later observed in a few magnetic compounds belonging to cubic noncentrosymmetric space groups. β -Mn-type alloys based on Co_xZn_yMn_z stoichiometry (x+y+z=20) crystallise into the chiral *P*4₁32 and *P*4₃32 space groups, are one of the few compounds hosting skyrmions. Co₃Zn₈Mn₄, for instance, has a near room-temperature skyrmionic phase and it is described as one of the most promising skyrmionic compounds [1]. On the other hand, Co₇Zn₇Mn₆ may present stabilised disordered skyrmions near the spin-glass transition (T_g~60 K), as well as the typical skyrmion lattice below the ordering magnetic temperature (T_c~180 K) [2]. As skyrmion lattices may find use in nanotechnological devices, such as data storage systems, β -Mn-type alloys are one of the best candidates for those possible applications.

In this work, we prepared three different polycrystalline samples of composition Co₇Zn₇Mn_{6-x}T_x, T=Cr,Ni , x=0,0.25 stoichiometry, to explore how the magnetic properties are affected by low doping by Cr and Ni. The samples were prepared by the Bridgman-Stockbarger method starting from high purity metals and characterised by XRD and EDS to check for purity and composition. Magnetic measurements were performed by VSM magnetometry between 1.8 and 400 K, with fields up to 9 T.

The pure compound shows a ferrimagnetic behaviour with $T_c = 182(2)$ K. The low-field M(T) curves display a irreversible behaviour and below ~30 K the M(H) curves are hysteretic, compatible with a cluster spin-glass state as reported in literature [2,3].

The Co₇Zn₇Mn_{5.75}Cr_{0.25} sample has as a much lower T_c (140(2) K) and similar features compatible with a lowtemperature spin-glass sate are also observed below ~34 K. The value of the high-field magnetisation decreases by ~30% compared to the pure compound. A small anomaly (jump) was observed in the low-temperature M(H)curves occurring at 1800 Oe, and disappearing at ~40 K that is not observed in the pure compound. In the case of Co₇Zn₇Mn_{5.75}Ni_{0.25}, the low-field M(T) curves show the presence of two transitions, one occurring at ~170 K and another one, at a higher temperature (~250 K). In between these temperatures a different magnetic phase appears to exist, likely a helical or skyrmionic phase. At low-temperature, irreversible behaviour was also seen comparing the ZFC and FC curves, with a larger remanence at low temperature.

- [1] Y. Tokunaga et al., Nature Communications 6, 7638, Jul 2015.
- [2] K. Karubee et al., Science Advances 4, eaar7043, September 2018
- [3] Joshua D. Bocarsly et al., Physical Review Materials, 3:014402, January 2019

Domain wall dynamics in nanostripe with perpendicular magnetic anisotropy induced by perpendicular current injection

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Recently, studies of the domain wall (DW) dynamics in ferromagnetic nanowires attract great attention. This is substantially caused by the fact that apart from the purely fundamental interests, the controlled dynamics of the DW underlies promising application for spintronic devices. Up today, the main approach of DW dynamics control is based on current-induced DW motion. It was demonstrated that in case of current perpendicular to the plane (CPP) injection the DW velocities can be rather high even in case of relatively low current densities. However, most of the results in this field correspond to the in-plane or even zero magnetic anisotropy, while the CPP geometry case combined with PMA ferromagnetic nanostripe remain unclear. Up today there is only one experimental demonstration [1] of the DW motion in the MTJ with PMA in CPP geometry, which encourages the detailed numerical and analytical investigation of DW dynamics mechanisms and features.

A numerical and analytical study of the dynamics of domain walls (DWs) in a magnetic tunnel junction (MTJ) nanostripe with perpendicular magnetic anisotropy (PMA) in free layer is presented; such systems are of great applied and fundamental interest. The equilibrium states of the domain wall (Neel, Bloch and hybrid) are obtained for different widths of the structure. The corresponding symmetries of the components of spin transfer torques and the polarizers' magnetization directions, which favour for stable DW motion in the case of perpendicular current injection, are obtained. The DW steady motion with velocities up to hundreds m/s at low current densities are reported. The Walker breakdown is demonstrated and the dynamics of the post-threshold motion of the Neel and Bloch walls for various configurations of torques and polarizers' magnetization directions is investigated. The analytical insight, which demonstrates good agreement with micromagnetic simulations is provided, this model can be useful to analyse the influence of different parameters on the DW dynamics in PMA based systems and devices. Lastly, the design of DW-based logic device is proposed, and the operation of this logic element is simulated by means of micromagnetic modelling.

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[1] S. Lequeux, J. Sampaio, V. Cros, K. Yakushiji, A. Fukushima, R. Matsumoto, H. Kubota, S. Yuasa, and J. Grollier, Scientic Reports 6, 31510 (2016).

Directional motion of domain walls in a low-frequency magnetic field in iron-garnet crystals

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Abstract

The search for the conditions of inducing the directional motion of the stripe domains array is actual in connection with the development of new types of memory devices and spintronic elements [1].

The changes in the direction of domain walls (DW) drift a) with the monotonic change of external harmonic magnetic field amplitude at various field frequencies and b) with the monotonic change of temperature at fixed values of the field amplitude and frequency are established in iron garnet single-crystal plates with different crystallographic orientation.

The investigated samples have stripe domain structure (magnetic anisotropy has an orthorhombic component with a K_p constant in the range (2.5 ÷ 7.5)·10³ erg/cm³). The drift was investigated in a harmonic magnetic field in the frequency range $f = 20 \div 900$ Hz, amplitude H_0 up to 300 Oe, at temperatures $T = 150 \div 350$ K including the magnetic compensation point. Dynamic domain structure of the samples was revealed by means of the Faraday effect and was registered by a high-speed camera with a speed up to 2000 fps.

In case a) the DW drift starts at some threshold amplitude H_{dr} , at $H_0 > H_{dr}$ the DW velocity increases and reaches the maximum value at H_0^{max} . At $H_0 > H_0^{\text{max}}$ the DW velocity decreases to 0 and DW stops at field amplitude $H_0 = H_{\text{st}}$. At $H_0 > H_{\text{st}}$, the DW start moving in the opposite direction. Changing of the drift velocity sign in the frequency range 20 ÷ 900 Hz occurred at $H_0 = 100 \div 110$ Oe. In this case, the velocity changed from 0.1 to -1.5 mm/s. The change in the drift direction was accompanied by a sharp increase in the number of magnetic dislocations nucleating in the sample [2].

The change of the drift direction with temperature (case b)) was established for fixed values of frequency and amplitude of the external magnetic field. For the the frequency f = 800 Hz and amplitude $H_0 = 185$ Oe, when the temperature increases in the range of 293 \div 306 K, DW slow down to a complete stop. When the temperature increases farther the DW begin to drift in the opposite direction. In this case the drift velocity varied from 0.6 to -4.5 mm/s.

The experimental results obtained in the work are discussed within the framework of the proposed theoretical model.

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Moon, K. W., Kim, D. H., Kim, C., Kim, D. Y., Choe, S. B., and Hwang, C., Journal of Physics D: Applied Physics 50(12), 125003 (2017)
 Pamyatnykh, L. A., Filippov, B. N., Agafonov, L. Y., and Lysov, M. S., Scientific reports, 7(1), 1-11 (2017)

Magnetotransport studies on epitaxial Sb₂Te₃

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Chalcogenide thin films have become of interest in energy conversion, as thermoelectric materials, and for spintronic applications. Amongst them, antimony telluride (Sb₂Te₃) has recently gained attention as Topological Insulator (TI), even in its granular form [1]. On the other hand, in order to exploit these materials in devices, it is important to reach epitaxial quality on large areas. We report the development of a room temperature Metal Organic Chemical Vapor Deposition (MOCVD) process to prepare 30 nm Sb₂Te₃ thin films on Si(111) and present the evolution of their surface roughness, morphology, and crystalline orientation upon pre-growth and postgrowth thermal treatments, with a special emphasis on the growth of epitaxial Sb₂Te₃ films [2]. Figure 1 shows a typical atomic force microscopy (AFM) topography. Magnetoresistance (MR) measurements in the Van der Pauw configuration are conducted on a set of samples characterized by a different structural quality as presented in Ref.[2]. In particular, the longitudinal resistance of the Sb₂Te₃ layers is recorded as a function of the perpendicularly applied magnetic field. All the samples show the quantum-originated phenomenon of weak antilocalization (WAL). The MR data are interpreted in the framework of the Hikami-Larkin-Nagaoka model [3] in order to evaluate the dephasing length (α) and phase coherence length (I_{ω}) as a function of the temperature and the different pre and post-growth thermal processing. For all the samples, WAL dominates at temperatures lower than 30 K, with the highest α up to 0.7 (and I_{φ} around 50 nm) being extracted for the Sb₂Te₃ characterized by the best epitaxial quality. We demonstrate a clear connection between the evolution of the MR properties of the 3D-TI Sb₂Te₃ samples and their corresponding crystalline quality. Our results demonstrate that an appropriate material processing is efficient in improving the topologically-protected conduction in such a large-area MOCVDsynthesized Sb₂Te₃ topological insulator.



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Figure 1: AFM image of epitaxial-quality 30 nm Sb₂Te₃ thin film highlighting surface.

- [1] R. Cecchini, Phys. Status Solidi RRL, 12, 1800155 (2018)
- [2] Rimoldi, RSC Advances, 10(34), 19936-19942(2020).
- [3] S. Hikami, A. I. Larkin, Y.Nagaoka, Prog. Theor. Phys. 63,707(1980)

Symposium 8. Spin-transfer based phenomena and devices

Spin-torque switching of perpendicular magnetic tunnel junction nanopillars at cryogenic temperatures

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The semiconductor industry is actively developing high-density, non-volatile random access memories based on perpendicular magnetic tunnel junction nanopillars for consumer electronics and thus operation between room temperature and ~150 C [1]. Such devices are also of interest for cryogenic computing systems (4 K) where a persistent, fast, low-energy consuming, and nanometer scale device is needed. Here we explore the effect of temperature on the nanosecond pulse switching characteristics of state-of-the-art perpendicular magnetic tunnel junction (pMTJ) nanopillar devices (40–60 nm in diameter). Interestingly, and perhaps counterinitiatively, the characteristic switching time decreases with temperature, with the largest reduction occurring between room temperature and 150 K. While the switching energy increases with decreasing temperature, but still compares very favorably with other types of spin transfer devices at 4 K, with <300 fJ required per switch. Write error rate (WER) measurements show highly reliable switching with WER $< 5 \times 10^{-5}$ with 4 ns pulses at 4 K. We also present a systematic study of the temperature dependence of the spin torque switching probability from room temperature down to 4 K, sampling up to a million switching events. The junction temperature at the switching voltage obtained from the thermally assisted spin torque switching model—is found to saturate at temperatures below about 75 K. This shows that junction heating is significant below this temperature and that spin torque switching remains stochastic down to 4 K. This will be discussed along with a thermal model can capture aspects of this behavior.

[1] A. D. Kent and D. C. Worledge, Nature Nanotechnology 10, 187 (2015).

[2] L. Rehm et al., Appl. Phys. Lett. 115, 182404 (2019).

[3] L. Rehm et al., arXiv:2009.01743

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Probing fundamental spin-related properties via photoinduced inverse spin-Hall effect in Pt/semiconductor heterostructures

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The aim of spintronics is to exploit the spin degree of freedom to add new functionalities to electronic devices and boost their performances. In this sense, spintronic studies in semiconductor-based platforms are both of fundamental and applicative interest. Anyway, classical semiconductors lack a net spin population in equilibrium. The optical orientation technique allows generating an out-of-equilibrium spin population in the conduction band of a semiconductor thanks to the absorption of circularly-polarized light. Due to fundamental band structure properties, the spin-polarization of the optically-injected carriers is drastically dependent on the energy of impinging photons and on the investigated semiconductor.

In this work, we studied different Pt/semiconductor heterostructures, in which a spin-polarized electron population is generated via optical orientation in the semiconductor and diffuses into the Pt layer, where it is detected via the inverse spin-Hall effect (ISHE). Since the spin transfer from the semiconductor to the Pt layer is dependent on the spin-diffusion length and on the spin-polarization at the time of generation, the investigation of the ISHE signal allows gaining insight on these fundamental properties of semiconductors. In particular, here we estimate the spin-diffusion length in Ge, Si, GaAs, and Ge/SiGe multiple quantum wells (MQWs). We obtain a long spin-diffusion length (at least of the order of the micrometer) for Ge, Si, and Ge/SiGe MQWs and a shorter one (tens of nanometers) for GaAs. In the case of Ge/SiGe MQWs, we also estimate the spin-polarization at the generation time, which agrees with the one predicted by tight-binding calculations. It is worth noticing that the employed spin injection/detection scheme is completely free from ferromagnetic materials, which simplifies the fabrication and makes the device intrinsically robust against external magnetic fields. Moreover, the technique is a powerful tool for the investigation of fundamental spin-related properties, even in configurations that are otherwise inaccessible to other techniques.

Oral Presentation

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Large voltage-tunability of threshold current and frequency in spin Hall nano-oscillators

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Spin Hall nano-oscillators (SHNOs) utilize dissipation less pure spin currents to excite local nanoscopic regions of magnetic thin films into auto-oscillating precession at microwave frequencies [1-3]. Thanks to ultra-wide frequency tunability [2] and robust mutual synchronization both in long chains [1] and 2D arrays [3], nano-constriction based SHNOs have recently emerged as one of the most promising candidates for high-quality microwave signal generation and ultra-fast neuromorphic computing [4]. While the SHNO frequency can be tuned with both magnetic field and drive current, neither approach is promising for an energy-efficient individual SHNO control in synchronized networks desired to perform cognitive tasks in oscillator-based neuromorphic computing [3,4]. Here we demonstrate electrostatically gated nano-constriction based W(5nm)/CoFeB(1.7nm)/MgO(2nm) SHNOs in which the voltage-induced changes in perpendicular magnetic anisotropy (PMA) not only tunes the auto-oscillation frequency (12 MHz/V) but also introduces a large 22% voltage-control over threshold current (see Figure 1(a-b)). Our detailed analysis based on spin-torque ferromagnetic resonance (ST-FMR) in conjunction with micromagnetic

simulations establishes that the strong tunabilities are caused by an interplay between a moderate voltage control of the PMA and the specific geometry of nano-constriction SHNOs resulting in a large change of the effective damping. Our demonstration provides an energy-efficient approach to critically control the effective damping and is an important step towards the realization of complex neuromorphic tasks [3,4].



[1] A. A. Awad et al., Nat. Phys. 13, 292-299 (2017).

[2] H. Fulara et al., Sci. Adv. 5, eaax8467 (2019).

[3] M. Zahedinejad et al., Nat. Nanotechnol. 15, 47–52 (2020).

[4] M. Romera et al., Nature 563, 230–234 (2018).

Room-temperature, non-volatile electric control of spin currents generation in ferroelectric semiconductors

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The displacive ferroelectrics GeTe and SnTe have been studied for decades but their multifunctional properties based on spin-orbit coupling remained unexplored until few years ago. They are indeed prototypes of ferroelectric Rashba semiconductors, a class of materials in which ferroelectricity is coupled to the Rashba spin texture [1, 2], enabling the nonvolatile electrical control of spins. Moreover, sizable spin Hall effect was recently predicted in these compounds [3, 4], opening the route towards spin transistors free from ferromagnets.

Here, we demonstrate the switchability of the ferroelectric polarization in epitaxial thin films of GeTe through gate electrodes. The switching is obtained through voltage pulses and measured as resistance variation of metal/GeTe heterojunctions. The sizable modulation of resistance is associated to the distribution of ferroelectric domains and related to the polarization-dependent band bending at the metal/semiconductor interface. The ferroelectric switching is obtained at low voltage (3-7 V) and is robust, with endurance up to 10⁵ cycles.

Spin-to-charge conversion in GeTe was studied by spin pumping measurements on Fe/GeTe heterostructures [5-7]. We demonstrate the ferroelectric control of magnitude and sign of spin-to-charge conversion in GeTe, with a conversion efficiency comparable to that of reference heavy metals like Pt. Moreover, first principle calculations point out that spin Hall effect is main mechanism for such conversion.

Finally, we present our recent results on the growth and characterization of ultrathin film of SnTe in view of its exploitation in spintronic devices.

The ferroelectric control of spin-to-charge interconversion in semiconductors open unprecedent opportunities as a tool for the development of all-in-one spin transistors.

- [1] D. Di Sante et al., Adv. Mater. 25, 509 (2013).
- [2] C. Rinaldi et al., Nano Letters 18, 2751 (2018).
- [3] H. Wang et al., npj Comput. Mater. 6, 7 (2020).
- [4] J. Sławińska et al., 2D Mater. 6, 025012 (2019).
- [5] S. Varotto et al., Proc. SPIE 10732, Spintronics XI, 10732 (2018).
- [6] C. Rinaldi et al., APL Mater. 4, 032501 (2016).
- [7] J. Sławińska et al., Phys. Rev. B 99, 075306 (2019).

Spin-orbit torques in Sn/FeCo heterostructures

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One of the less studied materials is the only known single element topological insulator: α -Sn, the diamond allotropic phase of Sn. This material has shown a large spin-to-charge conversion in spin-pumping experiments; however, the reciprocal effect of spin-orbit torques has not yet been measured [1]. Here we present a study on spin-orbit torques of three different Sn/FeCo heterostructures, two made of α -Sn deposited on CdTe, one of which grown with one monolayer of Bi as a surfactant, and one made of β -Sn, the metallic allotropic phase of Sn, deposited on MgO. We characterised the film structure and magnetotransport properties and measured the spin-orbit torques at room temperature. We found that the two α -Sn samples present a larger torque efficiency than the metallic β -Sn, the largest efficiency belonging to the sample with Bi surfactant. This indicates that the topological insulator phase α -Sn is crucially important for generating strong spin-orbit torques. The highest efficiency can be attributed either to a better crystal quality of the α -Sn film or to the role of heavy-metal Bi atoms that segregate at the CoFe interface. Additionally, we found that the torque efficiency of the α -Sn sample with the Bi surfactant decreases with decreasing temperature, suggesting that the carriers contributing to spin-orbit torques are thermally excited.

[1] J-C. Rojas-Sánchez et al, Physical review letters 116.9, 096602 (2016).

Non-equilibrium Spin Injection Into a Helicoidal Ferromagnet

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The phenomena that occur during injection of non-equilibrium spin into a ferromagnet with a noncollinear helicoidal distribution of magnetization are considered theoretically. Spin injection can be carried out in two ways: by spin pumping [1] (pure spin is injected) or by injecting a spin-polarized electric current (spin and charge are injected). In both cases, an additional (uniform) ferromagnet serves as a source of spin.

The exchange interaction couples spatial and spin degrees of freedom in non-collinear magnetic textures such as the magnetic helicoid. If the spin is injected into a helicoidal magnetic system with the helicoid axis parallel to the boundary of the ferromagnet the exchange coupling leads to the flow of electrons (i.e. electric current) along the helicoid axis. If the spin is pumped by exciting the ferromagnetic resonance in spin source this provides an exchange mechanism of conversion of pumped spin current into electric current (see Figure 1 (a)). In contrast to known inverse spin Hall effect [2] and Rashba-Edelstein effect [3], this mechanism gives the electric current along the magnetization of ferromagnetic source. In the case when an electric current is passed in a similar system perpendicular to the boundary between a homogeneous and inhomogeneous ferromagnet, both spin and charge is injected into the magnetization of a homogeneous ferromagnet, which is the source of the spin (see Figure 1 (b)). This current is perpendicular to the electric current initially passed through the system, and thus is the Hall current. This effect is called the topological Hall effect. It is worth noting that the system considered in this paper is indeed non-coplanar, which, as is known from the literature [4], is necessary for the appearance of the topological Hall effect.

We provide microscopic calculations of the mentioned effects based on the solution of Schrödinger equation for conduction electrons in the system. Our results correspond to the symmetry properties of the considered system.



Figure 1

[1] Y. Tserkovnyak et al., Phys. Rev. Lett. 88, 117601 (2002)

[2] J. Sinova et al., Rev. Mod. Phys. 87, 1213 (2015)

[3] J.C. Rojas Sanchez et al., Nature Communications 4, 2944 (2013)

[4] G. Tatara and H. Kawamura, J. Phys. Soc. of Japan 71, 2613 (2002)

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Magnetic dipolar synchronization of vortex-based spin torque nano-oscillators with independent top contacts

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Magnetic tunnel junctions (MTJs) constituted by two ferromagnetic (FM) layers separated by an MgO nanometric insulating barrier are the ultimate Spintronics device. The conjugation of the tunnel magnetoresistance effect and the manipulation of the magnetization of the free FM layer using a local spin-polarized electric current (spintransfer torque effect) makes them candidates to provide a new generation of devices, including the spin torque nano-oscillator (STNO)[1]. Nevertheless, the output power and noise levels of a single STNO fall far below those necessary for industrial implementation. As a possible solution, it was proposed to integrate a large number of synchronized STNOs in an array. It has been shown that the power is increased by N² and the linewidth reduced by N^{-1} , where N is the number of oscillators [2]. Here, one aims to achieve synchronization by dipolar coupling, controlling individually the DC current passing through each STNO. A 5 Ta / 50 CuN / 5 Ta / 50 CuN / 5 Ta / 5 Ru / 6 IrMn / 2.6 CoFe₃₀ / 0.85 Ru / 1.8 CoFe₄₀B₂₀ / MgO (3.7 Ω.μm²) / 2.0 CoFe₄₀B₂₀ / 0.2 Ta / 7 NiFe / 10 Ta / 30 CuN / 7 Ru MTJ stack was deposited by magnetron sputtering (thicknesses in nanometers). Circular pillars (diameter of 350 nm) were defined by e-beam lithography and ion beam milling. The free FM layer (i.e. 2.0 $CoFe_{40}B_{20}$ / 0.2 Ta / 7 NiFe) has a remanent vortex state. Each STNO has an independent top contact (ITC), so that the DC current may be controlled individually. The sustainable oscillation of a single STNO is achieved by applying a sufficiently large DC current and perpendicular field. The oscillation frequency is tuned by DC current, so that for a pair of STNOs the synchronization was attempted by maintaining constant the current of one STNO and sweeping the current of the other [3]. Figure 1 shows the spectral results obtained for a pair of STNOs (STNO₁ and STNO₂) 1µm apart (edge-toedge distance). Starting with I_{STNO2} = -2.5mA, each STNO is oscillating individually at a different frequency. As I_{STNO2} is swept, the frequency of STNO₂ increases until locking with the frequency of STNO₁ (clear for $I_{STNO2} = -3.4$ mA). Nevertheless, the key parameters do not show an effective synchronization, i.e. the power (linewidth) does not increase (decrease) significantly. This implies that a time domain analysis is required to understand the phenomenon, namely measuring the phase difference ($\Delta \phi$) between the oscillations of each STNO. Figure 2 shows time domain results from micromagnetic simulations. Starting from a random $\Delta \phi$, the dynamic behaviour varies depending on the gap between pillars. There is phase locking (i.e. $\Delta \phi = 0$) for a 0.1µm gap, whereas for 1µm the absence of phase-locking explains the experimental results. Via dipolar interaction, a full synchronization between two STNOs (1 µm apart) was attempted. The experimental results show the absence of phase locking between STNOs, corroborated by micromagnetic simulations, where phase locking is achieved by reducing the gap between oscillators.



Figure 1: Spectral results obtained for a pair of STNIs, 1 um apart, with ISTNO=-4, and Hperp= 5.3kOe



 Z. Zeng et al., Nanoscale 5, 2219-2231 (2013).
 S. Tsunegi et al., Sci. Rep. 8, 13475 (2018).
 R. Lebrun et al., Nat. Commun. 8, 15825 (2017).

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Thresholdless non-resonant broadband spin-torque diode rectification

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Growing interest for power autonomous nanodevice networks demands new feasible technologies for energy harvesting from ambient sources, e.g., heat or electromagnetic noise. A spintronic device such as nanopillar magnetic tunnel junction (MTJ), which consists of free and polarizer ferromagnetic layers separated by MgO tunnel barrier, potentially is a viable solution. Recent research has revealed that these devices are capable of converting radio frequency (RF) currents to DC voltage via spin-torque diode (STD) rectification effect [1]. STD Rectification combines tunneling magnetoresistance (TMR) and spin transfer torque effects, and thus, when injecting RF current MTJ generates constant voltage signal which has resonant behavior. State-of-the-art spin torque diodes utilizing nonlinear dynamics significantly outperform conventional Schottky diodes in terms of sensivity for low microwave power below 1 uW [2]. However, for energy harvesting from ambient electromagnetic waves, an essential feature of the rectifier is wide frequency bandwidth. Recently, several works have reported on broadband non-resonant spin-torque diode rectification due to *out-of-plane precession*. From theoretical [3] and experimental [4,5] works, it follows that this mode requires either out-of-plane component of the biased magnetic field, which tilts initially planar magnetization of the free layer, or fine-tuning perpendicular magnetic anisotropy via magnetic layers thickness. Up to date 1 GHz band has been achieved with a critical value 10 uW of microwave power for initialization.

In this work we study thresholdless non-resonant broadband spin-torque diode rectification in a magnetic tunnel junction with *in-plane magnetization of the free layer*. We report on an alternative advantageous approach for wide frequency band rectification which is based on the spatial nonuniformity of the free layer magnetization distribution. We used ST-FMR method to measure rectified voltage as a function of incident signal frequency and external magnetic field.

Next, we investigate various sample geometries to demonstrate how this mode can be induced.

Our spin torque diode rectifier exhibits 3.5 GHz wide frequency band. It does not have power threshold and possess sufficient sensivity below 1 uW. The rectification grows as one approaches to the point of demagnetizing and anisotropy fields compensation. Finally, we present results of micromagnetic modeling which are consistent with our experimental data and provides an insight into magnetization dynamics of the system.

- [1] Tulapurkar et al., Nature 438, 339–342 (2005).
- [2] Miwa et al., Nature Mater 13, 50–56 (2014).
- [3] O. V. Prokopenko et al., J. Appl. Phys. 111, 123904 (2012).
- [4] Fang et al., Phys.Rev.Applied 11, 014022 (2019).
- [5] M. Tarequzzaman et al., Appl. Phys. Lett. 112, 252401 (2018).

Characterization of the RF-to-DC conversion properties of spintronic magnetic tunnel junctions for wireless sensor network applications

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Wireless sensor networks (WSN) are part of our daily lives and their applications are growing, especially with the advent of the Internet of Things. The development of autonomous, adaptive and energy-efficient communicating sensor solutions is a topic of intense research. One way to drastically reduce energy consumption is to wake up the main radio receiver of a WSN system only when it has to be used. For that purpose, a wake-up receiver (WuRx) is used, which itself should operate with ultra-low power consumption and provide immunity to parasitic signals. Magnetic Tunnel Junction (MTJ) devices offer a promising solution since they can serve as RF-DC converters and act as passive, frequency-selective demodulators with high sensitivity to low input power [1,2].

As a preamble to build a full WuRx-system based on MTJs, we have compared the characteristics of two types of MTJs for passive detection (zero DC current biasing) and under zero external magnetic field: uniformly and perpendicularly magnetized MTJs (called pMTJ) for the 1-5 GHz frequency range, [3] and vortex-state MTJs (called V-MTJ) for the 150-400MHz range, [2]. The selection criteria concern the RF-DC conversion efficiency, the RF equivalent circuit as well as the demodulation capabilities.

Overall, we found that V-MTJs show a better RF-DC conversion efficiency than pMTJs, see Fig.1. For the DC equivalent circuit we consider the MTJ as a DC voltage source with an internal resistance R_{DC} . The RF equivalent circuit is derived as illustrated on Fig. 2, after evaluating the RF reflection coefficient through a vector network analysis, and applying de-embedding techniques (to remove any influence from the measurement set-up). It is found that due to their lower RF resistance V-MTJs are the better choice, as compared to pMTJs. For future matching to an antenna (between 50 and 100 Ω), it will be convenient to connect about ten V-MTJs in series. This will improve as well the detector dynamics. To test the demodulation capabilities, the input RF signal frequency was fixed to the maximum of the DC signal and was then modulated with an On-Off Keying (OOK) modulation scheme, increasing the modulation frequency, see Fig. 3. We obtained for V-MTJs data rates up to 20 Mbit/s for an input RF power of -15dBm leading to a signal to noise ratio of more than 14 dB, which guaranties an error free data transmission. In comparison, for pMTJs, the maximum modulation frequency was 100 kbit/s. It has to be noticed that 20 Mbit/s is with a comfortable margin regarding the typical data rates reported for existing WuRx in the literature [4]. Based on this performance, a complete RF wake-up system will be designed and tested.





Fig.2 : RF equivalent electrical model of MTJs.

B. Fang et al., Phys. Rev. Applied 11, 014022 (2019).
 A. S. Jenkins et al., Nature Nanotech. 11, 360 (2016)
 A. Chavent, et al., doi.org/10.1016/j.jmmm.2020.166647
 H. Bello et al., Sensors 19(14), 3078 (2019)

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Manipulating the helical phase of chiral magnets with electric currents

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The competition between the ferromagnetic exchange interaction and antisymmetric Dzyaloshinskii-Moriya interaction can stabilize a helical phase or support the formation of skyrmions. While skyrmions and lattices of such are widely studied and known to be easily manipulable by electric currents, the structurally less complex helical phase is usually stronger pinned by defects. In thin films of chiral magnets, however, the current density can be large enough to unpin the helical phase and reveal its fascinating dynamics.

We theoretically study the dynamics of the helical phase under spin-transfer torques that reveal distinct orientation processes, (i) driven by topological defects in the bulk or (ii) induced by edges. We also study the instability of the helical phase at larger currents and report an intrinsic instability at any finite current in the direction perpendicular to the orientation of the helix. Our experiments confirm the possibility of on-demand switching the helix orientation by current pulses.

We propose that the electrical control over the helical orientation might be the basis for a novel design for memory/computation devices that exploit this orientation as new order parameter.

Influence of the substrate temperature on the build-up of Joule heating in ferromagnetic nanostrips

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Joule heating is an undesired biproduct in spin transfer torque (STT) experiments performed in magnetic nanostrips. These experiments are very common in modern magnetism, due to the technological potential of some proposals such as the Race-Track Memory [1].

Joule heating is generated by the large current density used in spin transfer torque experiments and it plays an important role in the magnetization dynamics even in the nanosecond timescale [2]. There have been few studies evaluating the magnitude of Joule heating in nanostrips [3,4] but, to our knowledge, none of them evaluated how effective is a cryogenic measurement reducing the stripe temperature at the nanosecond timescale. In this work, we solve the heat diffusion equation with COMSOL software [5], to study the temperature reached in Permalloy nanostrips of different dimensions as a function of the current density and the substrate temperature, from cryogenic to room temperature. For instance, when using pulses of 5ns in a nanostripe which seats on top of a substrate at a temperature of 77K, a current density of 2.3 TA/m² can be deployed without a detrimental increase of temperature. The maximum temperature reached is 450K, far from the Curie Temperature of most ferromagnetic materials used in these experiments. If the substrate is at 10K, a 2.6 TA/m² 5 ns pulse would lead to the same 450K temperature. The temperature can be reduced by increasing the width to thickness ratio in the nanostrip. In this work we explore the temperature behaviour for any time scales as shown in Fig 1 for a stripe of cross section $15 \times 300 \text{ nm}^2$ and a length of $6 \mu \text{m}$. As it becomes quite clear in this figure, performing the measurement in a cryostat is beneficial in terms of reducing the final temperature, although Joule heating is still present, increases rapidly, and it can be sizable. The improvement in the cooling capability of the nanostrip, from having a Liquid Nitrogen measurement (77K) to a Helium based cryostat (few Kelvin), may not justify the difference in cost of the measuring setup. Our results may help experimentalists to optimize the design of their experiments and to have a rough estimate of the temperature their stripes may reach during the experiments.



Figure 1: Temperature of the nanostrip in the first 5 n of a current pulse. Dash lines represent COMSOL simulations and continuous lines our model. Grey colours represent the current density which changes due to temperature dependent resistivity of the ferromagnetic strip.

- [1] S.S.P. Parkin et al., Science (80-.) 320, 190–194 (2008)
- [2] S. Moretti et al., Phys. Rev. B. 95, 1–10 (2017)
- [3] C.Y. You et al., Appl. Phys. Lett. 89 (2006)
- [4] K.J. Kim et al., Appl. Phys. Lett. 92, 2005–2008 (2008)

[5] COMSOL, COMSOL Multiphysics® Modeling Software, Comsol. (2016). https://doi.org/10.1007/s11356-015-5536-x.
Optimization of unbiased spin torque diode with perpendicular anisotropy

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Spin diode operating on the basis of the spin transfer torque effect is a new technology in spintronics. This device can be successfully used for detecting radio frequency signals and collecting microwave energy, since its sensitivity can exceed the sensitivity of the now widely used Schottky diodes. One of the most promising designs for a spin torque diode (STD) is a multilayer structure with perpendicular magnetic anisotropy (PMA) in the free layer. Different ratios between magnetic anisotropy and magnetostatic field from the lower magnetic layers lead to different tilts of the free layer magnetization, which in turn strongly affects the properties of the spin diode. Motivated by the work of Fang et al. [1] and Zhang et al. [2], we investigated $Co_{20}Fe_{60}B_{20}/MgO/Co_{40}Fe_{40}B_{20}/Ru/Co_{70}Fe_{30}$ multilayer for different thicknesses of $Co_{20}Fe_{60}B_{20}$.

Using various thicknesses of the free magnetic layer which resulted in the different magnetization tilt we systematically investigated STD sensitivity in the absence of the external magnetic field and DC current. For sensitivity analysis we performed numerical approaches which included macro-, micromagnetic modeling and analytical approach based on Landau-Lifshitz-Gilbert equation with small amplitude oscillation approximation. All theoretical approaches showed strong dependence of the STD sensitivity on the angle of the free layer magnetization and reaches its maximum value within angle region which is not equal to angle regions investigated in earlier works. Our results will bring more insight about spin diodes and will help achieve record sensitivities of such devices.



Figure 1 : a Multilayer structure investigated in the work, top layer has different thicknesses. **b** Relaxed magnetization angle dependence on thickness of the free layer obtained by different theoretical approaches. **c** STD sensitivity dependence on thickness of the free layer (colour indicate input power: red - 2nW, blue - 20nW, green - 100nW, brown - 1000nW).

B. Fang et al., Nature Communications 7(1) (2016). doi:10.1038/ncomms11259
 L. Zhang et al., Applied Physics Letters 113(10), 102401 (2018). doi:10.1063/1.5047547

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Symposium 9. Novel 2D magnetic systems and heterostructures

CrTe₂ thin flake imaging with NV magnetometry

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Even if magnetic van der Waals crystals where known since decades, two-dimensional (2D) magnetism has been obtained experimentally only a few years ago[1]. Since its discovery in Cr₂Ge₂Te₆ and Crl₃, an intense research effort has started, triggered by their high potential in studying 2D magnetic states[2-4]. A wide range of new physical properties emerging from the reduced dimensionality can be explored and exploited like the control of magnetic properties through gating. New devices based on van der Waals magnetic heterostructures have been proposed, such as spin-filter magnetic tunnel junctions based on Crl₃ heterostructures, showing a tunneling magnetoresistance higher than their counterparts based on conventionally grown magnetic thin films. The potential of 2D magnets in topological spintronics is also under investigation, with for example recent works on the stabilization of skyrmions in Fe₃GeTe₂. However, in order to use van der Waals magnets as building blocks for relevant spintronics devices, a magnetic material with a stable Curie temperature (TC) above room temperature is required. Unfortunately, most of the available van der Waals magnets have a low intrinsic TC. Even though a 2D magnetic state at room temperature can be achieved through gating for example, it remains a challenging task and only few materials are reported to have a TC above 300K down to the monolayer limit, like MnSex, VSe2 andmore recently for few-layers CrTe₂. Here, we report our work on CrTe₂ from bulk to 40nm thin mechanically exfoliated flakes using Nitrogen-Vacancy (NV) magnetometry, a well-suited technique for the study of 2D magnets[5]. We find an in-plane ferromagnetic order at room temperature and we extract a value for the saturation magnetization Ms of $24.5 \pm 3.0 \text{ kAm}^{-1}$.



Figure 1.

[1] N. Samarth, Nature 546, 216 (2017)

[2] K. S. Burchet al., Nature 563, 47 (2018)

[3] X. Z. Cheng Gong, Science 363, 6428 (2019)

[4] M. Gibertini et al., Nature Nanotechnology 14, 408 (2019)

[5] L. Thiel et al., Science 364, 973 (2019)

Proximity-induced spin Hall effect in graphene/WSe₂ van der Waals heterostructures with tunable, highly efficient spin-to-charge conversion

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The proximity effect in two-dimensional materials opens ways to achieve important functions for future spintronic devices. In van der Waals heterostructures (vdWHs), transition metal dichalcogenides (TMD) can be used to enhance the spin-orbit coupling of graphene leading to highly efficient spin-to-charge conversion (SCC) by spin Hall effect (SHE) that is predicted to be controllable by a gate voltage. Here, we report for the first time the observation of the SHE in graphene proximitized with WSe₂ in a vdWHs. These kind of vdWHs are a promising platform to study a variety of spin-dependent phenomena [1,2]. By Hanle precession measurements, we quantify the spin transport and SCC parameters from 10 K up to room temperature. Exceptional for graphene/TMD devices, the sole mechanism is the SHE for all measurements and no Rashba-Edelstein effect is observable. Importantly, we are able to amplify and turn off the SCC by applying a back-gate voltage, demonstrating the long-awaited milestone of an electrically-tunable SHE. The amplified SCC shows a high efficiency, measured with an unprecedented SCC length of up to 41 nm (with a lower limit of 20 nm).

W. Yan et al., Nat. Commun. 7, 13372 (2016).
 C. K. Safeer et al., Nano Lett. 19, 1074–1082 (2019).

Reversible control of electric polarization in SrTiO $_3$ -CoFe $_2O_4$ at room temperature by XAS

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Strain engineering of thin-film heterostructures is one of the most widespread and successful approaches to improve the performance of real devices such as transistors, electrochemical energy conversion devices, or multiferroic memories [1,2]. We have engineered local strain in quantum paraelectric SrTiO₃ exploiting CoFe₂O₄ magnetostrictive properties and succeeded in reversibly controlling the electron distribution in the Ti valence band of SrTiO₃-CoFe₂O₄ heterostructures at room temperature. Changes in the energy level scheme of Ti 3d orbitals upon the application of an external magnetic field have been observed by means of soft x-ray absorption spectroscopy. Our results can be summarized as follows: first, samples where Ti-O bonding covalency is enhanced by increasing the Sr non-stoichiometry show a larger strain. Secondly, the derived Ti electronic state supports the existence of a net electric polarization when samples is subjected to moderately large external magnetic fields. Such an induced state disappears upon removing the applied magnetic field. Our approach could also be applied to other 2D heterostructures offering the possibility to induce and reversely control novel properties found at the interface, such as polar vortexes or two-dimensional electron gases. Our approach is expected to lead to the realization of a new class of functional devices for information storage and sensing applications.

[1] N.S. Bingham, A.K. Suszka, C.A.F. Vaz, H. Kim, L.J. Heyderman, Phys. Rev. B 96, 024419 (2017)

[2] R.V. Chopdekar, V.K. Malik, A. Fraile Rodríguez, L. Le Guyader, Y. Takamura, A. Scholl, D. Stender, C.W. Schneider, C. Bernhard, F. Nolting, L.J. Heyderman, Phys. Rev. B 86, 014408 (2012)

Spin to charge conversion at LaAlO₃ // SrTiO₃ interface states

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Creating a dissipation-less pure spin current and being able to detect it, have been one of the biggest challenges that held the spintronics community for decades. In the beginning of 1970's, Dyakonov et al. [1], made a big break through in this aspect by discovering the Spin Hall Effect (SHE), that made it possible to create a pure spin current by sending an electrical current in non-magnetic heavy metals like Platinum (Pt), this effect relies on the Spin-Orbit Coupling (SOC) interaction which links the spin degree of freedom to the orbital degree of freedom resulting in a preferential directional scattering for electrons of different spins.

Recently another type of SOC have been discovered, which relies on the Rashba interaction [2,3], this effect stems for a joint action of the SOC and an electrical built-in potential in two-dimensional electrons systems (2 DES) existing at interfaces of two different materials. A promising system to study the spin to charge conversion is the interface between two band gap insulators like the interface between LaAlO₃/SrTiO₃ (LAO/STO), where a 2DES is created due a polar discontinuity at the interface[4,5]. This 2 DES at the interface of LAO/STO shows a very strong Rashba spin-orbit interaction raising from the breaking of the inversion symmetry. It appears that due to the high dielectric constant in STO below 30K [6], we are able to tune our Rashba splitting using an electric field [7]. In this work we showed that the Rashba splitting in LAO/STO depends on the angular dependence of the injected spins, so we are able to retrieve an important information regarding the symmetry of the orbitals that are participating in the conversion signal[8].



Figure 1. (Colour online) Angular dependence of the DC IEE length for a 4-monolayer-thick LAO. The plot is obtained taking into account the expected efficiency of the IEE voltage due to the prism shape of the Py layer and mirror-symmetrized.

- [1] M. I. DYAKONOV International Journal of Modern Physics BVol. 23, No. 12n13, pp. 2556-2565 (2009).
- [2] E I Rashba 1959 Fiz. Tverd. Tela 1 407 (E I Rashba 1959 Sov. Phys.-Solid State 1 368).
- [3] Linding Yuan et al., Nature Communications volume 10, Article number: 906 (2019).
- [4] Thiel S, Hammerl G, Schmehl A, Schneider C W and Mannhart J Science 313 1942 (2006).
- [5] Cantoni C et al Adv. Mater. 24 3952 (2012)
- [6] H. E. WEAVER7 J. Phys. Chem. Solids Pergamon Press. Vol. 11. pp. 274-277 (1959).
- [7] A. D. Caviglia et al., Phys. Rev. Lett. 104, 126803 Published 26 March (2010).
- [8] J.-Y. Chauleau et al. , EPL, vol 116, num 1 (2016)

Oral Presentation

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Meta-magnetism of Weakly Coupled Antiferromagnetic Topological Insulators

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The magnetic properties of van der Waals magnetic topological insulators MnBi₂Te₄ and MnBi₄Te₇ are investigated by magneto-transport measurements. We evidence that the relative strength of the inter-layer exchange coupling J to the uniaxial anisotropy K controls a transition from an A-type antiferromagnetic order to a ferromagnetic-like metamagnetic state. A bi-layer Stoner-Wohlfarth model allows us to describe this evolution, as well as the typical angular dependence of specific signatures, such as the spin-flop transition of the uniaxial antiferromagnet and the switching field of the metamagnet. In micron-size magnets, the single-domain switching-field astroid are however partly truncated by the nucleation of domain walls along the easy-axis direction.

Oral Presentation

4240

Tunable spin to charge interconversion in van der Waals heterostructures

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Two dimensional materials have provided unique insights in condensed matter physics, yielding a multitude of novel effects [1]. In particular, graphene constitutes a unique material for spintronics owing to its exceptional and tunable electrical properties, which enables propagation of spin information over long distances [2]. Recently, it has also been experimentally demonstrated that spin transport in graphene-based heterostructures can be manipulated by proximity-induced phenomena [3]. These key features provide the building blocks for electric-field generation and manipulation of spin currents by using the spin-orbit phenomena in graphene-based devices.

In this talk, I will show that strong and controllable spin-to-charge (StC) interconversion can be obtained in engineered graphene-WS₂ heterostructures at room temperature. Our findings demonstrate that StC driven by the spin galvanic and the spin Hall effects can be unambiguously detected and quantified [4]. Remarkably, the corresponding StC conversion efficiencies can be tailored by electrostatic gating, with a magnitude that is comparable to the largest efficiencies reported to date. Such unprecedented electric-field tunability provides a novel route for spin generation free from magnetic materials and paves the way for the development of ultracompact and low power consumption magnetic memory devices.

- [1] W. Savero Torres et al. MRS Bulletin 45(5), 357-365, (2020)
- [2] W. Han et al. Nat. Nano. 9, 794-807, (2014)
- [3] L. A. Benítez, J.F. Sierra, W. Savero Torres et al. Nat. Phys. 14, 303-308, (2018)
- [4] L. A. Benítez, W. Savero Torres et al. Nat. Mat. 19, 170-175, (2020)

3481 Introducing CVD WS₂ in Magnetic Tunnel Junctions

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Spintronics has opened a new paradigm through the use of the spin variable as the vector of information and has been largely applied from hard drives read-heads to the STT-MRAMs. While very recent, the introduction of 2D materials in Magnetic Tunnel Junctions (MTJs) has already shown some promising properties[1]. Graphene and the 2D insulator h-BN have been the first 2D materials to show strong impact on spin transport in MTJs. The recent advent of the wide TMDC family of 2D semiconductors opened new opportunities for further tailoring of spintronics properties. We will present results on the scarcely studied WS₂. We will detail a protocol using laser lithography technology to fabricate spin valves based on CVD grown WS₂, with step by step characterizations in support (Raman spectroscopy, photoluminescence, and AFM measurements). We will finally show our first spin transport measurements obtained in a CVD WS₂ based MTJ. Our measured MR signals, above state of the art for 2D semiconductor based MTJs, validates our integration approach. We observe that the spin signal extracted from a ferromagnetic electrode can be tuned by placing atomically thin WS₂ on top of it. Furthermore the thickness of WS₂ significantly affects the extracted spin polarization of the 2D/FM interface. We discuss that trend using the peculiar band structure of WS₂, supported by DFT calculations, leading to thickness dependent spin filtering. Our work opens the way to the integration of different members of the very large TMDCs family, in order to reveal their spin transport properties in MTJs[3].

- [1] M. Piquemal-Banci et al., J. Phys. D: Appl. Phys., 50(2017) 203002
- [2] M-B. Martin et al.Appl. Phys. Lett. 107 (2015) 012408 ; M. Piquemal-Banci et al.ACS Nano, 12 (2018) 4712
- [3] V.Zatko et al., ACS Nano, 13 (2019) 14468

Study on the magnetic characteristics induced in Ni based layers on LiNbO3, Si, and SiO2/Si substrates by X-ray photoemission spectroscopy

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An artificial formation of ferromagnetic/ferroelectric heterojunction can provide the novel properties to the system, resulting in paving a way to fabricate or develop the novel functional materials. Well, what is the origin to induce the uniaxial magnetic anisotropy in Ni layer deposited on LiNbO₃ substrate? The magnetostriction and magnetoelastic effect paly a significant role in modulating magnetic propeties. In this study, we investigate the magnetic characteristics of Ni-based films on various substrates using the X-ray photoelectron spectroscopy and X-ray circular dichroism photoelectron emission microscopy (XMCD-PEEM).

In this study, XPS and XMCD-PEEM were carried out at the laboratory and SPring-8 BL17SU, respectively. Ni-base materials such as Ni, Ni₈₁Fe₁₉, NiCu alloys thin films were prepared onto Si, SiO₂/Si, and LiNbO₃ substrate by means of magnetron sputtering process. First, depth profile of XPS spectra for each system was measured. Figure 1 shows a typical experimental result: a system comprising 12.5-nm-thick Ni layer deposited onto a Si substrate covered with naturally oxidized silicon layer. Figure 1 presents a typical depth profile of XPS for the Aucap/Ni/naturally oxidized SiO₂/Si system. We found that the NiO XPS peak appears near the interface between Ni and Si substrate. This result indicates that the Ni layer is oxidized near the interface. On the other hand, we confirmed that the XPS peak of Ni shifted near the interface between Ni layer and LiNbO₃ substrate and NiO peak cannot be detected. By comparison of some XPS depth profiles and spectra, these XPS peak shifts are attributed to the modulation of electronic state distribution near the interface through magnetoelastic, magnetostriction, and interference effect. In addition, combining the XMCD-PEEM observation and XPS measurements, we found that the heterostructure can induce the uniaxial magnetic anisotropy in the Ni layer on the LiNbO₃ substrate and control the magnetic domain structure. The same measurement was performed for the other systems, and the electronic state analysis of the interface was performed. As a result, the modulation of electronic state at the interface was induced in several systems.

Thus, the material design by the introduction of heterojunction opens a door to develop and create novel artificial materials and to investigate the new magnetic properties.



Figure 1. (a) Typical XPS depth profile of Au-cap/Ni/naturally oxidized Si/Si substrate system. (b) Etching time dependence of XPS Ni 2p spectra.

Disorder-induced time effect in the antiferromagnetic domain state of Fe_{1+x}Te

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Iron-based chalcogenides (IBCs) have a quasi-two-dimensional crystal structure, which is the simplest one among iron-based superconductors. This makes IBCs ideal candidates for both experimental and theoretical investigations. FeSe is superconducting below a critical temperature $T_c = 8$ K [1], which is tunable by various parameters and can be increased up to 100 K when a monolayer of FeSe is grown on SrTiO₃ [2]. The analog tellurium-based IBC compound, Fe_{1+x}Te, is non-superconducting and, in contrast to FeSe, has an antiferromagnetic (AFM) ground state, which depends on the amount of excess Fe concentration *x*. Explaining the large difference between FeSe and FeTe ground states, despite their almost identical crystal structures, could help to understand the origin of unconventional superconductivity. Recently, 100 nm-sized AFM domains have been reported to form on the surface of FeTe as a result of a strong magneto-elastic coupling [3].

Here, we report on temperature dependent X-ray absorption spectroscopy (XAS) measurements using linear and circular light polarization as to probe magnetic phase transitions in Fe_{1+x} Te with x = 0.073. X-ray magnetic circular dichroism (XMCD) signals in magnetic fields of 7 T scale linearly with the bulk magnetization $\langle M(T, B = 7 T) \rangle$ (measured by a SQUID instrument) and exhibit a characteristic drop at the Néel temperature $T_{\rm N}$ = 57 K, when Fe1+xTe enters the AFM state. The magnetic phase transition is confirmed by our bulk sensitive specific heat measurements. On the contrary, X-ray magnetic linear dichroism (XMLD) signals (~ $<M^2>$) start to increase monotonically at temperatures below T_N , which proves an in-plane AFM alignment of Fe spins along the [100] crystallographic direction of the monoclinic FeTe parent phase. Hereby it is important to note that the collinear AFM alignment on a macroscopic scale is achieved by a [100]-oriented biasing field, which favors a single-domain state during cooling across $T_{\rm N}$. For the sample under study, however, we find that the field-cooled AFM state is highly unstable even at lowest temperatures. After switching off the biasing field at T = 1.6 K, the XMLD signal decays exponentially with a relaxation time of 1500 s. The initial XMLD signal is restored only upon repeating a cycle consisting of heating and field-cooling through $T_{\rm N}$. We explain the time effect by a gradual formation of a multi-domain state with 90 deg rotated AFM domains, similar to that reported by local scanning probe techniques [3]. In the chosen experimental geometry, the 90 deg rotated AFM domains contribute to the XMLD with an opposite sign and thus compensate the total signal, when averaging over many domains within a 100 µm wide XAS probing area. We attribute the instability of the magneto-elastic domain formation to a critical structural disorder in our sample, which is evident from significantly broadened peaks in its powder x-ray diffraction pattern. Fe1+xTe reference samples with similar stoichiometry but less disorder show stable XMLD signals in time. XMLD data at lowest temperatures are compared with results of our density functional theory calculations. The stability of magnetic phases can be an important parameter in IBC, since $Fe(Te_1-xSe_x)$ phase diagrams show that magnetism is usually intimately connected with superconductivity.

^[1] F. C. Hsu et al., PNAS 105, 14262 (2008)

^[2] S. L. He et al, Nat. Mater. 12, 605 (2013)

^[3] J. Warmuth et al., Quantum Materials 3, 21 (2018)

Strain-Induced Effects of Topological Deformed Graphene

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The dynamics of electrons in graphene can be described by a two-dimensional Dirac equation. Graphene is a two dimensional material embedded in three dimensions and is assumed to define a 2D flat surface. However, one expects deviations from flatland. Structural corrugations and ripples, have been observed in suspended graphene, and furthermore, atomistic simulations have shown that ripples appear spontaneously owing to thermal fluctuations [1].

Geometric curvature and strain in graphene can give raise to pseudo-magnetic fields, which can lead to observable phenomena. Furthermore, it is possible to connect strain with the possibility of opening energy gaps in the graphene electronic spectrum as already experimentally observed [1].

We consider the out-of-plane deformation of the form:

$$z(x,y) = \frac{1}{2}A\cos[k(x+y)]\cos[\sqrt{3}k(-x+y) + k(x+y)]\cos[\sqrt{3}k(-x+y) - k(x+y)]\sin[k(x+y)],$$

where A is the deformation amplitude and k refers to the modulation frequency of the deformation. For a small deformation parameter $\eta = Ak \ll 1$, we obtain at the approximation $O(\eta^4)$

$$\Gamma^1(x, y) = \gamma^1 \qquad \Gamma^2(x, y) = \gamma^2$$

with the spin connection expression written as $\omega_1^{12}(x, y) = -A^2 k^4 x$ and $\omega_2^{12}(x, y) = A^2 k^4 y$.

After rearranging the terms we obtain the Dirac oscillator expression

$$(i\{\gamma^0 \ \partial_0 + \gamma^x(\partial_x + yB) + \gamma^y(\partial_y - xB)\}\Psi = 0)$$

With *B* being a constant. For this out-of-plane deformation it is possible to obtain the Dirac harmonic oscillator equation, where Landau levels can here be addressed.

The main motivation to search for a deformation of this nature is because graphene-type materials inevitably deform (out-of-plane) as described by the function presented above. The physics described by the Dirac oscillator equation allows to interpret many experimental results carried out in graphene-type materials. As a consequence of the Dirac harmonic oscillator equation, it is possible to address the physics of the Landau levels and also consider the theoretical framework of Jaynes-Cummings. [2,3]

^[1] A. J. Chaves et al., J. Phys. Condens. Matter 26, 18 (2014), and references therein.

^[2] A. Bermudez et al., Phys. Rev. A 76, 041801(R) (2007).

^[3] P. Rozmej, R. Arvieu, J. Phys. A 32, 5367 (1999).

Interplay of magnetic disorder and layer native defects within topological insulators

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Material can host several kinds of native defects, which includes chemical defects as well as structural defects. In general, it leads to the modification of their physical properties including magnetic and transport ones. Therefore, in the present work based on ab-initio calculations we focus on the influence of the layer defects, namely the experimentally evidenced twinning planes, on the properties of the well known bismuth chalcogenide topological insulators Bi₂Se₃, where we are concerned especially with their interplay with magnetic dopands. We employ the TB-LMTO-ASA method based on the layered Green's functions, which allows us to treat the chemical disorder efficiently in the framework of the CPA. The distribution of the twinning planes within multilayer sample is discussed together with the dependence of their formation energy in relation to the type of the point defects and their concentration. The interplay between layer defects and magnetic dopants and their magnetism is emphasized. Besides, the impact of the presence of the structure defects on the electron structure, particularly on the surface states and on the surface gap, is estimated.

Exfoliation of hematite: Morphology, structural and magnetic investigations

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Exfoliation of Hematite (Fe₂O₃) which is an ionic compound has recently been demonstrated with potential magnetic and photo-catalytic applications.¹ Magnetism in hematite is explained by anti-ferromagnetic Fe³⁺–O– Fe³⁺ superexchange and a weak Dzyaloshinski–Moriya interaction results in slight canting of sublattice moments when they lie in the basal plane of the hexagonal corundum structure. The canted moment is 0.005 μ_B per Fe³⁺ ion above the spin reorientation Morin transition temperature (T_M = 265 K), below which the sublattice moments rotate to lie along the c axis. Our aim was to study the spin rearrangement in quasi-two-dimensional 'hematene' using magnetic and Mössbauer analysis.

From a preliminary study of seven well-crystallised natural samples, nanoscale sheets were obtained from hematites from Cumbria, England and Iron County, Utah using ultrasonic exfoliation in DMF and centrifugation. TEM analysis showed some micron-sized hematite sheets with composition (O = 61.59 at %, Fe = 34.08 at %) determined by XRF analysis, while most of the grid was covered by nano-sized particles ~ 100 nm and dust ~ 5 nm (Fig 1a and 1b). AFM shows nano-structured sheets with thickness ranging from ~ 2nm-12nm. The nano-sized sheets were analyzed structurally using XRD, Raman and IR spectroscopy. Magnetic analysis reveals a drop in the room-temperature magnetic moment from 0.18 Am²kg⁻¹ for hematite to 0.03 Am²kg⁻¹ for the nano-structured sheets, which showed with no evidence of a Morin transition in the thermomagnetic scan (Fig. 1c). An additional paramagnetic phase was observed in addition to the ferromagnetic sextet of hematite (IS = 0.42 mm/s, QS ($\Delta/6$) = -0.022 mm/s, HF = 51.36T) in the Mössbauer spectrum (Fig. 1d). Further experiments at temperature below the Morin transition are underway to monitor the magnetic transformation in bulk and nano-structured sheets.

The absence of the Morin transition in hematine can be explained by additional easy-plane anisotropy arising from the surface of the quasi 2D-sheets. The extent of exfoliation as a function of the deposited ultrasonic energy with aim of enhancing the yield of clean 2D-nanostructured sheets.



Figure 1. Representative TEM image of micron-sized sheet for Cumbria, England (b) TEM image of micro-sized sheet covered with nano-sized particles. (c) Representative magnetic analysis of bulk hematite and exfoliated sheets, inset shows M vs. T plot for both materials (d) Mossbauer spectra shows characteristic ferromagnetic sextet for hematite with paramagnetic components.

[1] (a) Balan, A. P. et. al., Nat. Nanotechnol. 13, 602–609 (2018), (b) Zhiwei Fang et. al., ACS Nano., 13, 14368–14376 (2019),
(c) A. C. M. Padilha et. al., J. Phys. Chem. C, 123, 16359-16365(2019).

Symposium 10. Magnetism in molecular/ionic/organic based systems

Is exchange bias at the hybrid Organic/Ferromagnet interface an intrinsic effect?

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Exchange bias (EB) plays an important role in spin electronics since it allows pinning the magnetization direction of one of the ferromagnetic (FM) layers involved in devices. It has been thoroughly studied between inorganic anti-ferromagnets and metallic ferromagnetic layers but only recently and rather surprisingly similar exchange bias effects have been reported between metallic ferromagnets and organic layers. This is considered a "spinterface" induced magnetic effect, that may rise when organic molecules (OM) are in proximity to inorganic ferromagnetic (FM) thin films. Molecular exchange bias effect has been first observed in 2015 [1] between cobalt films and manganese phthalocyanine (Co/MnPc), then expanded and reproduced with different metal (Zn, Fe, Mn, Co) phthalocyanine molecules [2]. However, fundamental understanding of this FM/OM spinterfaces and their accompanied effects are not fully achieved, and research activities are strongly ongoing in this direction. In our work, we investigate the possibility of molecular exchange bias in cobalt/metal tetra phenyl porphyrin (MTPP; M=Zn and Ni) hybrid structures. Careful SQUID magnetometer measurements show the absence of exchange bias in "freshly deposited" hybrid samples. Surprisingly, exchange bias appears within few hours and gradually increases in the time span of days and weeks. In addition, new and similar "freshly deposited" samples kept under Ultra High Vacuum (UHV) for the same time span did not show exchange bias when measured directly after removal from the UHV chamber. This shows that EB effect has its origin from the exposure of samples to air. In the light of these unexpected results, further investigations have been undertaken, Atomic Force microscopy (AFM) and X-ray Photoelectron spectroscopy (XPS) have been performed to identify and understand the reasons of our observations. Our results strongly suggest that EB in FM/OM systems is not an intrinsic effect and originates from air-driven oxidation of the cobalt films transforming a part of the Co metallic film into CoO_x oxide that is well known to produce EB effects.



Figure 1. | Magnetization loop of (a) 50 nm gold capped "freshly deposited" Co (6nm)/ZnTPP (10nm) hybrid sample (black loop) and of the same sample measured after 2 weeks (red loop). (b) 50 nm capped "freshly deposited" Co (6nm)/ NiTPP (10nm) hybrid sample (black loop) and of the same sample measured after 1 week (red loop). Loops acquired at 2K after field cooling in +5T in-plane magnetic field.

- [1] M. Gruber et al. doi.org/10.1038/nmat4361
- [2] S. Boukari et al. doi.org/10.1021/acs.nanolett.8b00570

Synthesis of Gold @ Spin Crossover Nano-Composites - Highlighting the Synergy between Surface Plasmon Resonance and Switching Properties

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Spin crossover (SCO) materials are based on coordination complexes of switchable electronic configuration upon external stimuli (temperature, light, pressure...), associated with memory effects (hysteresis loops). Photoswitching of such materials at room temperature remains a challenge, especially when ultrafast and weakly energetic switching is targeted. Based on the pulsed irradiation inside thermal hysteresis loops, photothermal effects can be achieved [1]: this intense laser pulse induces a short and huge local heating allowing the materials to experience the spin crossover. Strategies to reduce the energy and time needed for this switching focus on hybrid nanoparticles [2]. These hybrid architectures are based on a strong interplay between surface plasmon resonance of metallic nanoparticles that act as local nanoheaters [3] and optical thermo-induced switching of spin state.

We present here the original synthetic approach we followed, based on a direct grafting [4] of SCO on Au NPs, to elaborate in a controlled way, such core-shell architectures, with gold nanorods embedded in a SCO nanoparticle, Au@SCO [5]. The use of such morphologies targets an efficient diffusion of the heat produced by the gold nanoparticles to the whole SCO component.

In this presentation, we will show the strong synergy between the surface plasmon resonance of metallic nanoparticle and the spin crossover behavior through a strong modulation of the SPR and an efficient photoswitching of spin state in a nanocomposite material of Au@SCO[5]. We will also report a study on ns-laser pulse excitation of the nanocomposite material.

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- [1] (a) G. Gallé et al., Appl. Phys. Lett. 102, 063302 (2013); (b) F. Guillaume et al., Chem. Phys. Lett. 604, 105 (2014).
- [2] (a) I. Suleimanov et al., Chem. Commun. 50, 13015 (2014); (b) D. Qiu et al., R. Soc. Chem. Adv. 4, 61313 (2014).
- [3] G. Baffou et al., Laser Photonics Rev. 7, 171 (2013).
- [4] L. Moulet et al., Chem. Commun. 52, 13213 (2016).
- [5] M.Palluel et al., Adv. Funct. Mater., accepted (2020).

Spin dynamics of Fe4 Single Molecule Magnet monolayer on a type I superconductor

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Coupling magnetic materials with superconductors has recently disclosed novel phenomena attractive for spintronics and quantum technologies [1]. In particular, molecular spins deposited on superconductive surfaces led to the observation of localized magnetic states, known as Yu–Shiba–Rusinov bound state [2], or enhanced molecular spin lifetimes [3]. Single molecule magnets (SMMs), e.g. tetrairon(III) propeller-shaped complexes (Fe₄), represent an interesting class of magnetic molecules for their magnetic bistability properties that can be retained at the single molecule level even when deposited on a substrate [4,5]. Here we show a detailed microscopic and spectroscopic study of a sub-monolayer of Fe₄ SMMs deposited on a Pb(111) surface. We reveal that the transition of lead to the superconducting state affects the magnetization of the SMMs that locally switch from a blocked magnetization state to a resonant quantum tunneling regime. These findings open intriguing perspectives for controlling single molecule magnets in devices as well as for their use as local probes for superconducting materials.

[1] J. Linder et al., Nat. Phys. 11, 307 (2015).

- [2] L. Malavolti et al., Nano Lett. 18, 7955 (2018).
- [3] B. W. Heinrich et al., Nat. Phys. 9, 765 (2013).
- [4] A. Cornia et al., Eur. J. Inorg. Chem. 2019, 552 (2019).
- [5] M. Mannini et al., Nat. Mater. 8, 194 (2009).

Hybrid materials: how the surface can play an important role in functional device based on Spin-Crossover compounds

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A fundamental step toward the practical use of functional molecular materials as active units in molecule-based devices corresponds to their deposition as thin films assembled on solid surfaces. Obviously, the molecular properties observed in bulk must be retained or their alteration must be minimized and eventually controlled. These mandatory requirements are rarely satisfied in coordination compounds: the deposition process often influences the molecular structure, and in some cases interactions between the molecule and the surface affect the electronic nature of the molecular system, drastically altering its properties [1-4]. Herein, we report the spectroscopic characterization of different Fe(II) spin-crossover complexes (SCO) at the nanoscale, that can retain their SCO behavior when are in contact with the surfaces. X-ray Photoelectron Spectroscopy (XPS), Ultraviolet Photoelectron Spectroscopy (UPS) and X-ray Absorption Spectroscopy (XAS) were used to investigate the chemical composition, the temperature and light-induced spin-crossover of the deposited material and flanked by DTF simulations we proved the full evidence of these molecular systems to be efficiently employed into nanometric films. We also demonstrate the possibility to use in-house spectroscopic setups to check both the integrity of the system and its switching capability after nanostructuration [5-8]. Vertical tunnel junction, for which the spin-state switching of SCO compounds films can induce up to two orders of magnitude change in the tunneling current density flowing through the junction, will be presented [6,8,9]. Large-area junctions-based transport measurements suggest a feasible approach for the integration of these systems in novel multifunctional spintronic devices.

- [1] Halcrow, Spin-Crossover Materials: Properties and Applications (John Wiley Sons Ltd, Oxford, UK, 2013).
- [2] Harzmann, Frisenda et al., Angew. Chemie Int. Ed. 54, 13425–13430 (2015).
- [3] Devid, Martinho et al., ACS Nano 9, 4496–4507 (2015).
- [4] Senthil Kumar et al., Coord. Chem. Rev. 346, 176–205 (2017).
- [5] Poggini et al., Mater. Horizons 5, 506–513 (2018).
- [6] Poggini et al., J. Mater. Chem. C 7, 5343–5347 (2019).
- [7] Atzori et al., J. Mater. Chem. C 6, 8885-8889 (2018).
- [8] Poggini et al., Adv. Electron. Mater., 1800204, 1800204 (2018).
- [9] Cucinotta et al., ACS Appl. Mater. Interfaces 12, 28, 31696–31705 (2020).

Aging effect on the magnetic properties of Mn12 – Stearate SMMs anchored on spherical silica surface

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Single-molecule magnets (SMMs) engage a great research interest due to outstanding magnetic properties and prospective applications at quantum computing and molecule-based spintronics [1]. However, addressing and manipulating of individual SMMs on the molecular level require their organization on the surface [2]. The main challenge here is to examine how their magnetic properties retain after surface deposition and how long such characteristics can be preserved with time.

Here we investigate the aging effect on the structural and magnetic properties of Mn_{12} -stearate SMMs anchored on the surface of 300 nm spherical silica nanoparticles. Using propyl carbonic acid groups gives a possibility of deposition of SMMs on the silica surface while triethoxy silane spacer units provide the opportunity to control the distribution of such molecules. Raman spectroscopy measurements confirm of silica network functionalization by Mn_{12} -stearate molecules. Preservation of basic magnetic characteristics of SMMs after anchoring on the surface is shown by the DC magnetic susceptibility and relaxation measurements, which reveal that such material possess visible hysteresis in M(H) loops and slow magnetic relaxation. To investigate the aging effect on the sample we carry out a series of isothermal magnetization measurements during the 220 days period. Aging studies show exponential decrease of coercivity and remanence magnetization and also change in magnetic relaxation behavior. Raman measurements of aged sample reveal partial decomposition of stearate groups and possible formation of manganese oxide phase over time, which explain observed magnetic changes.

[1] G. Christou et al., MRS Bulletin 25, 66 (2000).

[2] A. Cornia and M. Mannini, Single-Molecule Magnets on Surfaces (Springer-Verlag, Berlin Heidelberg, 2014).

Crystal structure and magnetic properties of metal pyrophosphate materials $Fe_4(P_2O_7)_3$ and $Cu_4(P_2O_9)_2$

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The phosphates are incredibly versatile materials that have variety of applications as inorganic pigments, catalysts, biomaterials, proton conductors, lasing materials and host materials for nuclear waste. Metal containing pyrophosphates present an increasing interest due to its chemical and physical (magnetic, non-linear optical) properties [1]. In our investigation we concentrate attention on the two metal containing compounds: $Fe_4(P_2O_7)_3$ (Iron(III) pyrophosphate) and $Cu_4(P_2O_9)_2$ (Copper pyrophosphate). Both materials show field-induced metamagnetic behavior from an antiferromagnetic (anti-aligned spins) state to a ferromagnetic (aligned spins). The mean feature of the structure of $Fe_4(P_2O_7)_3$ is the occurrence of Fe_2O_9 dimers made of face-sharing FeO_6 octahedra.

The structural model of ferric pyrophosphate was studied theoretically starting from the crystallographic data [2]. The monoclinic unit cell, space group P2₁|n, and lattice parameters a=7.389 Å, b=21.337 Å, c=9.517 Å and β =90° were used as input parameters. Iron atoms were slightly off-centered in each octahedron in order to minimize the repulsive interactions between the common faces. For the second structure Cu₄(P₂O₉)₂ a similar monoclinic unit cell, with space group P2₁|c, and unit cell parameters: a=6.081 Å, b=10.428 Å, c=11.310 Å, β =90° were taken. The crystal parameters and atomic positions were optimized within the density functional theory using the VASP program [3]. After relaxation, the electronic band structures and density of states were obtained for both crystals. These studies revealed that electronic and magnetic properties of the Fe-based compound are significantly different from those of the copper pyrophosphates. The optimized crystal structures are used to calculated the interatomic force constants and frequencies of the infrared and Raman phonon modes.

- [1] M. Laskowska et. al., Microporous and Mesoporous Materials, submitted (2020).
- [2] L. K. Elbouaanani et. al., Journal of Solid State Chemistry 163, 412 (2002).
- [3] G. Kresse and J. Furthmuller, Phys. Rev. B 54, 11169 (1996).

Poster

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Magnetic measurements on Bimetallic MIL-100(Al/Cr) metal-organic frameworks

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Probing the continuity of Tetra phenyl porphyrin organic layers embedded between cobalt and iron films by Ferromagnetic Nuclear Resonance

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Magnetic behaviour of novel metal-organic low dimensional magnetic coordination polymers containing Mn, Cr, Cu and Ca metals

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Coordination polymers with metal-organic frameworks and organic or inorganic functional cations offer many possibilities in constructing interesting multifunctional materials such as ferromagnetic conductors, photo and piezo switching magnets, chiral magnets and multiferroics. Especially interesting are oxalate-based compounds, where the C_2O_4 ²⁻ acts as a linker between metal centres and provides various possibilities of coordination to metal centres resulting in structures with different dimensionalities. The other useful property of oxalate ions is their ability to mediate super-exchange interaction.

In this work we will present magnetic behaviour of three novel metal-organic coordination polymers synthetized by a layering technique. The investigated compounds consist of single or multi metal cations bridged by an oxalate ion.

The first compound, {Mn(bpy)(C₂O₄) \cdot 1.5H₂O }_n, has a 1D chain structure, where the chains are connected by π interactions between 2,2' bipyridyl molecules coordinated on the manganese ions of neighbouring chains. The second compound, {[CrCu(bpy)(C₂O₄)₃][CrCu₃(bpy)₃(CH₃OH)(H₂O)(C₂O₄)₄]·CH₂Cl₂·CH₃OH·H₂O}_n, consists of two kinds of chains in which two metal ions, chromium and copper ions, are bridged by the oxalate ion. The 2,2'-bipyridyl molecules are coordinated on the copper ions, enabling the existence of π interaction between the chains. One chain consists of only Cu and Cr ions bridged by oxalate, while the other chain has two more Cu atoms connected to the Cr ion. The third compound, {[CaCr₂Cu₂(phen)(C₂O₄)₆]_n·4CH₃CN·2H₂O}_n, is a two dimensional coordination polymer which consists of copper, chromium and calcium ions bridged by the oxalate bridge in the 2D network. The molecule coordinated on the copper is a 1,10-phenanthroline.

The magnetic properties of polycrystalline samples in powder form were investigated using a MPMS5 SQUID magnetometer. Temperature dependence of magnetization was measured in the temperature range 2 - 400 K, measuring both zero field cooled (ZFC) and field cooled (FC) magnetization curves. The MT curve of a single metal chain compound showed a broad peak at 15K, characteristic of antiferromagnetic chains, while the other two compounds show curves similar to the paramagnetic ones, with no maximum, or saturation at low temperatures, indicating a more complex magnetic coupling. The field dependence of magnetization was measured up to 5 T. The MH curve for the first compound is almost linear with no saturation, characteristic of antiferromagnetic chains, and the value of 1.1 $\mu_B/f.u.$ at 5T and 2K. The second and third compound show saturation of the magnetization at the value of 8 and 7 $\mu_B/f.u.$ respectively, indicating ferromagnetic or ferrimagnetic coupling of Cu and Cr ions.

Tailoring the magnetic anisotropy of lanthanides on surface-supported metal-organic networks

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The capacity of investigating and tailoring the properties of materials down to nanoscale has created new perspectives for the development of functional devices. In this context, the stabilization of magnetic remanence in single atoms represents the ultimate limit on the size reduction of storage devices.

In 2016, it was experimentally shown by X-ray Magnetic Dichroism that single bistable Ho atoms adsorbed on MgO(100)/Ag(100) [2] exhibit magnetic remanence, which is considered a landmark for the engineering of single atom magnets. Immediately after, in 2017, it was demonstrated for the first time that it is possible to read and write single magnetic atoms by means of electron spin resonance (ESR) [1], confirming that their use for magnetic storage is possible.

Recently, another system based on single Dy atoms adsorbed on graphene grown on Ir(111) [3] displayed magnetic remanence, thus revealing that lanthanides are promising candidates for atomic magnets. However, single standing atoms adsorbed on surfaces are not suitable for practical applications due to their high diffusion, i.e., low thermal stability. The next step towards more realistic systems is the coordination of these atoms in metal-organic networks.

We have prepared coordinated networks of Dy atoms on Cu(111) with *p*-terphenyl-4,4-dicarboxylic acid (TDA) and benzene-1,4-dicarboxylic acid (TPA) molecules, exploiting the high affinity of lanthanide elements for carboxylate moieties. The electronic and magnetic properties were investigated by scanning probe microscopy, X-ray natural linear dichroism (XNLD) and X-ray magnetic circular dichroism (XMCD). The experimental results were complemented by density functional theory (DFT) calculations.

Dy atoms coordinated in TDA and TPA metal-organic networks show a significant increase of the anisotropy when compared to isolated atoms or Dy clusters. Surprisingly, the effective crystal field of the surface-supported TDA and TPA networks induces a change in the orientation of the easy magnetization axis. Dy coordinative centers display an in-plane magnetic anisotropy even though the symmetry axis of the molecular lattice field is out-of-plane.

[1] F. D. Natterer et al., Nature 543 226 (2017).

[2] F. Donati et al., *Science* **352** 318 (2016).

[3] R. Baltic et al., Nano Lett. 16 7610 (2016).

Theoretical Predictions of single-molecule magnetism in rigid lanthanide organometallics

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Single molecule magnets (SMM) are a class of materials with incredible potential – these molecules can act as little magnets on their own, displaying unique effects such as magnetic hysteresis, slow relaxation of magnetization or quantum tunneling of magnetization on molecular level. These properties can potentially be exploited in applications like high-density memory media, quantum bits or spintronic devices, therefore it is crucial to find a way, how to make the most effective materials, that can fit for practical applications.

In this field, very successful approach was utilized in developing lanthanide double-decker systems, such as $[Dy(Cp^{ttt})_2]$ ($Cp^{ttt} = 1,2,4$ -tris-tert-butylcyclopentadienyl)[1] and $[Dy(Cp^{iPr5})(Cp^*)]$ ($Cp^{iPr5} = penta-iso-propylcyclopentadienyl, Cp^* = pentamethylcyclopentadienyl)[2], which had shown its key property, the blocking temperature, to rise up to 60 and 80 K respectively, which is a lot higher, than any value seen before in SMM systems. However, such systems have many drawbacks, which prevents them from wide applications, the biggest ones being instability and a very complicated preparation.$

In our work, we attempt to use theoretical methods to predict structures and properties of a series of dysprosium (III) double-deckers, with cyclopentadienyl rings connected by various number of aliphatic carbon chains (1-5), with aim to find systems with more axial geometry and better encapsulation of dysprosium ion of [DyL]⁺ system, which might lead towards increased stability of a compound. Structures were optimized with DFT computational method, which was followed by detailed study of ground state electronic structure done by CASSCF, including recently developed methods of analyzing magnetization reversal energy barrier [3] and a prediction of its blocking temperature [4]. Furthermore, we attempted to find the most probable relaxation path for reversal of magnetization through excitations within ground state Kramers doublets and performed simplified analysis of influence of vibrations on relaxation processes [5].



- [1] C. A. P. Goodwin et al., *Nature* **548**, 439–442 (2017).
- [2] F. Guo et al., Science **362**, 1400–1403 (2018).
- [3] D. Aravena, J. Phys. Chem. Lett. 9, 5327–5333 (2018).
- [4] A. Castro-Alvarez et al., Inorg. Chem. Front. 7, 2478–2486 (2020).
- [5] L. Escalera-Moreno et al., J. Phys. Chem. Lett. 8, 1695–1700 (2017).

4220 Molecular coupling effects interface on a heavy metal

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5d metals are used in electronic architectures because of their high spin-orbit coupling (SOC) leading to efficient spin \leftrightarrow electric conversion and strong magnetic interactions. When C₆₀ is grown on a metal, the electronic structure is altered due to hybridisation and charge transfer. Previously, it has been shown that this can lead to the emergence of spin ordering [1]. This interfacial effect is also critical in spin filtering and spin transport effects [2]. The spin Hall magnetoresistance for Pt/C₆₀ and Ta/C₆₀ at room temperature are up to a factor 6 higher than for the pristine metals, with the spin Hall angle increased by 20-60%. At low fields of 1-30 mT, there is an anisotropic magnetoresistance, increased up to 700% at room temperature by C₆₀. This is correlated with noncollinear Density Functional Theory simulations showing changes in the acquired magnetic moment of transport electrons via SOC. Given the dielectric properties of molecules, this opens the possibility of gating the effective SOC of metals, with applications for spin transfer torque memories and pure spin current dynamic circuits.

By fitting the SHMR data, we can extract the spin Hall angle (Fig. a) which, for wires ≤ 5 nm is significantly higher with the molecular overlayer. This effect disappears for thick wires (> 10nm), where the molecular interface does not significantly change the spin Hall angle. Inset: Top view of the optimized C60/Pt(111)-(2v3x2v3)R30° interface DFT model. The C60 molecules are adsorbed on top of one Pt-vacancy. The continuous black polygon marks the in-plane periodicity of the system. Pt: silver, C: cyan. The SHMR saturates once the applied field saturates the magnetisation out-of-plane, at 0.1-0.15 T for a YIG film 170 nm thick at 290 K, and no higher than 0.5 T for any measured condition. However, above this field range, other contributions such as Koehler MR, localisation and the Hanle effect can result in significant linear and parabolic contributions to the MR that would artificially enhance the SHMR ratio and \mathbb{D}_{SH} (Fig. c)[3]. In YIG/Ta(4nm), where C₆₀ increases the resistance rather than reducing it, both the SHMR up to 0.15 T and the polynomial MR at higher fields are enhanced (Fig. d).





[1] F. Al ma'mari et al., *Nature* **524**, 69-73 (2015).

[2] M. Cinchetti et al., Nature materials 16, 507 (2017).

[3] S. Velez et al., Physical Review Letters 116, 016603 (2016).

Real-time MOKE measurements of CoTMPP on magnetic Ni/Cu(110)-(2x1)O

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Understanding the coupling of organic molecules on ferromagnetic surfaces is of high relevance due to the fact that the spinterface dictates the behavior of the overall organic/inorganic material. Since the magnetic signal of a single molecular layer is rather small, a high sensitivity is required to monitor the changes of the magnetic properties caused during the formation of the organic/ferromagnetic interface.

Recently, we have demonstrated the switching of the magnetization of a 10.5 ML thin Ni film deposited on a Cu(110)-(2x1)O surface from out-of-plane to in-plane through the deposition of 2.7 ML of CoPc by employing a reflectance difference spectroscopic MOKE (RD-MOKE) system [1]. Nickel thin films deposited on Cu(110)-(2x1)O show an extremely sharp spin reorientation transition (SRT) at a critical thickness of 9 ML [2,3]. As a result, the orientation of the easy magnetic axis for Ni films with thicknesses slightly above or below this critical thickness can be influenced by small changes of the magnetic anisotropy, such as the adsorption of ultrathin layers of organic molecules.

In this work, we report the extension of our setup by combining a sinusoidal modulation of the magnetic field with the synchronous detection of the RD-MOKE signal. Besides improving the detection limit to variations of the Kerr rotation angle below 1 µrad/mT, the present setup allows recording hysteresis loops *continuously* as a function of coverage, time or temperature. We illustrate the capabilities of our improved setup for Ni thin films grown on a Cu(110)-(2x1)O surface and the subsequent deposition of cobalt tetramethoxyphenylporphyrin (CoTMPP) thin layers. The adsorption of the molecules induces characteristic changes in the magnetic properties (magnetization amplitude, remanence and coercive field) that are monitored as a function of the coverage with sub-monolayer resolution and as a function of temperature, revealing the decrease of the Curie temperature upon CoTMPP deposition on Ni films with different thicknesses.

- [1] M. Denk et al., J. Appl. Phys. 125, 142902 (2019).
- [2] R. Denk et al., Phys. Rev. B 79, 073407 (2009).
- [3] Th. Herrmann et al., Phys. Rev. B 73, 134408 (2006).

Symposium 11. Magnetic based metrology tools and techniques

Experimental determination of the interfacial Dzyaloshinskii-Moriya interaction: a Round Robin Comparison

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Over the recent years a lot of attention was paid to the Dzyaloshinskii-Moriya Interaction (DMI) [1,2], an anisotropic exchange interaction, which favours chiral structures with outstanding dynamic characteristics, as chiral domain walls or skyrmions. Especially DMI originating from the interface between a heavy metal (HM) layer and an ultrathin ferromagnetic (FM) film with perpendicular magnetic anisotropy (PMA) was studied intensively, due to the wide range of possible applications in the field of spintronics [3]. In these bilayers the DMI occurs due to inversion symmetry breaking and indirect exchange interaction at the interface. The strength of the DMI is expressed by the energy coefficient D (DMI constant) [2], a measure for the stability of the chiral magnetic structures. Although DMIbased phenomena have created an extremely active research field, a classification of materials according to the DMI constant and clear dependencies of D on the interface roughness, the preparation method, the layer thickness, etc. are still missing. One problem is the lack of an established measurement protocol for D and currently disagreements are present in the literature regarding different techniques, especially for small DMI values (D<0.5mJ/m<sup>2). The many different measuring techniques are found to provide contradictory values for D, and controversies are even present when utilizing the same method on similar stacks. Since literature data are difficult to compare due to differences in sample preparation, measurement details and data analysis, a systematic comparison of different techniques and samples is indispensable. We therefore performed a quantitative round robin comparison of the interfacial DMI constant, employing two different measurement techniques and investigating different compositions of sample stacks (e.g. FM: Co, CoFeB; HM: Pt, Ta, Ir, W). These techniques, currently the most widely spread to measure DMI, are based on: a) asymmetric bubble domain expansion [4] in creep regime in bi- or multilayers with PMA when an in-plane magnetic field is applied and b) asymmetric dispersion relation of thermal spinwaves [5] (frequency non-reciprocity). The measurements were performed using magnetooptic Kerr effect and Brillouin light scattering, respectively, in five different laboratories. Our aim is to try to clarify the differences of the techniques, the role of the stack materials, considering the layer thickness and the preparation methodology, and to define measurement protocols in order to be able to compare DMI values, obtained by different techniques. Furthermore, we evaluate different models [6] for analysing the experimental data of the bubble domain expansion method, which can influence strongly the resulting value of D.

- [1] I. Dzyaloshinskii, Sov. Phys. JETP 5, 1259 (1957)
- [2] T. Moriya, Phys. Rev. 120, 91 (1960)
- [3] S. Parkin and S.-H. Yang, Nat. Nanotechnol. 10, 195 (2015)
- [4] S.G. Je et al., Phys. Rev. B 88, 214401 (2013)
- [5] J.H. Moon, Phys. Rev. B 88, 184404 (2013)
- [6] D. Lau, et al., Phys. Rev. B 98, 184410 (2018)

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New Approach for Dynamic Imaging of Competing Magnetic States and Stochastic Switching Pathways

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For real world applications, spintronic devices based e.g. on domain wall motion need to be highly reliable. The magnetization states have to be stable and the switching pathways transforming one state into another must be highly reproducible. A single failure can be a definitive obstacle for device functionality, yet scientifically it is a difficult task to understand stochastic processes and detect rare events that might lead to errors and hence this is rarely assessed. With direct imaging there are pump-probe magnetic imaging techniques which are based on billions of dynamic repetitions. However, the resulting image is then only an average of all events that occurred and in general, no quantitative information is yielded concerning the reliability of the observed dynamic pathways and if there are rare events during such measurements.

In this work we demonstrate a novel approach to determine details of competing magnetic states and dynamic switching pathways based on a newly developed scanning electron microscopy with polarization analysis system which offers pump-probe imaging with ns temporal resolution [1]. We apply this to the investigation of domain wall based switching [2] and chirality control in asymmetric Py half-ring pairs via temperature dependent dynamic imaging. In particular such curved structures have received significant interest as attractive geometries for a range of applications such as data storage, logic and sensors [3-5]. Understanding and controlling the switching behaviour of such elements, including the domain wall (DW) motion in the systems, is vital for reliable device operation. Furthermore, control over the DW chirality is required for chirality-encoded DW logic and devices based on DW motion due to chirality-dependent pinning [6-7].

In this study we detect the DW switching pathways of the structures and compare them to micromagnetic simulations. We show how we are able to extract details of stochastic pathways and determine the probabilities of the various processes occurring, as well as the effectiveness of the DW chirality protection in the system. The results reveal that the control of chirality by curvature works with very high reliability but also reveal details of the rare thermally activated processes that lead to different dynamic switching events. Furthermore, the attraction of vortex DWs in adjacent wires is found to play a key role in stabilizing the dynamics and affects the prevalent switching pathways. Through our approach we are able to overcome some of the limitations of conventional pump-probe imaging.

- [1] D. Schönke et al., Rev. Sci. Instrum. 89, 083703 (2018)
- [2] M.-A. Mawass et al., Phys. Rev. Applied 7, 044009 (2017)
- [3] M. Diegel et al., Sens. Lett. 5, 118 (2007)
- [4] D. Allwood et al., Science 309, 1688 (2005)
- [5] X. Han et al., IEEE Trans. Magn. 47, 2957 (2011)
- [6] K. Omari et al., Appl. Phys. Lett. 107, 222403 (2015)
- [7] E.-S. Wilhelm et al., Appl. Phys. Lett. 95, 252501 (2009)

Oral Presentation

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Validation of Quantitative Magnetic Force Microscopy by NV-Magnetometry

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Magnetic Force Microscopy (MFM) is a magnetic imaging technique that allows to image magnetic structures with nanometer resolution. However, per se it only delivers qualitative information since the magnetic properties of the tip are not known. In 1998, Hug et al. proposed a calibration approach allowing *quantitative* MFM based magnetic field measurements (qMFM) [1]. In qMFM, the quantitative output is achieved by calibrating the tip with a reference measurement on a sample with calculable stray field resulting in a tip transfer function (TTF). The TTF represents the tip stray field or its gradient depending on the calibration inputs [2]. However, the validity of the approach and the thereof resulting TTF has never been investigated with an independent measurement technique. Nitrogen-vacancy centers (NV) are color centers formed by a defect in diamond crystals where a carbon site adjacent to a vacancy was replaced by a nitrogen atom. Due to their spin splitting, they can be used to locally measure the magnetic field with high precision using optically detected magnetic resonance (ODMR) [3]. Here, we used NV-center magnetometry to investigate for the first time the lateral and spatial magnetic field profile of a calibrated qMFM tip. The observed discrepancies between the measured stray fields and the stray fields as determined by qMFM are investigated by forward simulations of qMFM calibrations in the presence of artificial noise and for different filter parameters of the calibration deconvolution algorithm in the Fourier spectrum. Based on this, the differences are explained by noise filtering occurring in the calibration algorithm in combination with the limited size of the calibration sample. Taking account of these results, the validity range of qMFM measurements is discussed.

[1] H.J. Hug, B. Stiefel, P. J. A. van Schendel, et al. Quantitative magnetic force microscopy on perpendicularly magnetized samples. J. Appl. Phys, 83, 5609 (1998).

[2] D. Nečas, P. Klapetek, V. Neu, et al. Determination of tip transfer function for quantitative MFM using frequency domain filtering and least squares method. Sci Rep 9, 3880 (2019).

[3] A. Gruber et al., Scanning confocal optical microscopy and magnetic resonance on single defect centers, Science 276, 2012 (1997).

Metrology and standardization for magnetic hyperthermia: an interlaboratory study

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To ensure delivery of optimal safety and performance, the medical technology industry is subject to compliance with relevant standards and regulations. High quality measurement science is therefore vital for the construction of reliable product testing procedures, and provision of adequate decision-making tools to regulatory bodies. It was previously noted that the development of magnetic nanoparticles for biomedicine has so far proceeded without significant industry standardisation, and a roadmap was proposed to help to meet this need [1]. In addition, work on a new series of international standards for magnetic nanomaterials has begun at ISO, with the first document in the series having been published last year [2]. It is against this backdrop that the authors report the findings of the first interlaboratory comparison of measurements for the characterisation of the heating power produced by magnetic nanoparticles for magnetic hyperthermia. With clinical trials already underway at multiple locations, a major world-wide research effort has recently focused on the development and characterisation of nanomaterials for this application. The heating efficiency of new nanoparticles is often reported by different laboratories in terms of either the specific loss power (SLP) in W/g, or the intrinsic loss power (ILP) in (Hm2/kg). Competition between researchers to report the most effective hyperthermia agents is growing, however the measurement and analysis techniques in use at different locations vary greatly. Here we report the findings of an interlaboratory study (n=21) comprising members of the European RADIOMAG project [3]. A survey of the equipment used at each location, and measurements and analysis results for different hyperthermia agents were collected from the consortium in multiple rounds. An initially startling variation in the first-round results was systematically probed via careful analysis of the equipment, analysis techniques and procedures in place at each location. By the final measurement round, the emergence of rank correlation between samples revealed that the stochastic uncertainty component had been greatly reduced, albeit with large site-specific systematic effects remaining. Despite these improvements, even the most rigid iteration of the study failed to produce quantitative results with acceptable uncertainties. The results provide a powerful demonstration of the present lack of methodological homogeneity in this field, a topic which must surely be addressed for magnetic hyperthermia therapy and other biomedical applications of nanoparticles to gain regulatory and market acceptance.

[1] J. Wells et al., J. Phys. D: Appl. Phys. 50 (2017) 383003

[2] ISO 19807-1 Nanotechnologies – Magnetic nanomaterials – Part 1: Specification of characteristics and measurements for magnetic nanosuspensions

[3] http://cost-radiomag.eu/

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Magnetic-field imaging using novel Vortex-Core MFM tip

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Nanomagnetism and spintronic will play crucial role in the near future. The manipulation of magnetic domains in ferromagnetic nanostructures opens new roads for unique electronic schemes including memory architectures with higher storage density and fast performance, new logic devices, sensors, etc. Together with designing new logic schemes, it is important to develop also novel tools for the observation of magnetic states at nanoscale. Imaging systems should offer quantitative and reproducible picture of the states at ambient conditions.

One of the well-established method for investigation of the magnetic nanostructures is magnetic force microscopy (MFM) [1]. It has big advantage over other imaging systems thanks to a simple sample preparation, simple operation, and relatively good spatial resolution (10-50 nm). The method is powerful and shows interesting applications in several scientific fields. MFM uses magnetized tip as a probe for measuring of the tip-sample magnetic interaction and the magnetic contrast corresponds to the frequency or phase shift of the vibrating cantilever. The task of the day in MFM methods is to provide correct quantitative interpretation of the magnetic signal obtained [2]. To do that one has to know the distribution of the tip magnetization and its interacting volume with the magnetic sample. Actual MFM methods show also other drawbacks which prevent [3] quantitative scanning - limited spatial resolution and low durability of the tips. When the spatial resolution is improved by sharp tips, one loses their durability and lowers the chance to calibrate such tips for longer period – the shape and magnetic moment of such sharp tip is changing during scanning.

Here we present how to solve mentioned challenging problems. Instead of a sharp magnetic tip, we propose to use physically thick and blunt tip with ferromagnetic disk placed at its very end. Such disk gives us narrow magnet (diameter ~ 15 nm), represented by the vortex core (VC) located in the centre of the disk and its stray field has out of plane orientation represented by single magnetization polarity vector. The vortex core acts like a magnetically sharp nanoscale probe. Figure shows the sketch of such probe and the detail of the tip with the ferromagnetic disk of diameter ~ 300 nm, placed at its apex. The VC tip provide constant magnetic moment defined only by the disk thickness and the material used – it is independent on disk diameter and small imperfections at its edges, so it can be considered as magnetic normal at nanoscale. As the tip is blunt, it is mechanically more resistant to wear and thus provide higher durability.

In this work we describe basic properties of the VC tip, the technology of preparation, sensitivity to magnetic field, durability, and we also discuss its scanning limits. First results obtained on hard disk drive are promising. We believe that VC tip could be a suitable sensor for magnetic field imaging in many scientific areas.



Figure 1 a) Sketch of the vortex-core tip placed on a cantilever. The ferromagnetic disk is placed at the tip apex. Vortex core is represented by the out-of-plane brown arrow. b) SEM image of fabricated tip. The fabrication process of the VC tip is based on the modification of commercial AFM tips by focused ion beam trimming.

- [1] Martin Y, et al., 1987 Appl. Phys. Lett. 50 1455
- [2] Kazakova et al., 2019 J. Appl. Phys. 125 060901
- [3] Cambel V et al., 2013 Appl. Phys. Lett. 102 062405

Magnetic scanning probe microscope integrating magnetoresistive sensors

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Giant magnetoresistors (GMR) and magnetic tunnel junctions (MTJ) are spintronic based devices. These sensors are schematically composed of two ferromagnetic layers separated by a non-magnetic spacer. The resistance of the structure depends on the relative orientation of the magnetization of these two layers. Thanks to a typical level of detection of a few nanoteslas, their large frequency range and their compatibility with CMOS technologies, such sensors are widely used for low field measurements, for example in automotive, biological systems or non-invasive testing [1][2][3][4].

We are developing a quantitative and very sensitive magnetic microscope, which combines a MR-based surface stray field sensing technology and a scanning probe microscope. One of the main interests of the microscope is that, by applying a small AC field, the magnetic susceptibility of surface could be measured and used for example for non-destructive testing of steel. Indeed, studies have shown that the magnetic susceptibility of steels is correlated to their mechanical properties [5]. The microscope is composed of a giant magnetoresistive sensor integrated at the apex of the flexible cantilever [6]. The processes of micron-size MR-sensors and micro-cantilevers were developed in the laboratory. Such a probe enables us to get decorrelated information about topography and magnetism of a studied sample. GMR measurement is quantitative, directional but also robust in time and does not require frequent recalibration of the probe comparatively to MFM tips.

In order to improve the spatial resolution, the linear range, and to reduce the magnetic noise by enhanced shape anisotropy [7], we have developed a process to manufacture nanometer-sized GMR sensors (100 to 800 nm) as a first step before integrating them on the scanning probe microscope. The key point of this process is a thinning step of micron-sized GMR sensors performed using electron-beam lithography. We fabricated and characterized several batches of nano-GMR in terms of sensitivity, noise spectral density and detection limit.

We integrated nano-GMR on scanning probe microscopy micro-cantilevers. Tips fabrication is performed using two etching steps respectively on the front and the back side of the wafer. A first batch of functional tips integrating nano-sensors (400 to 600 nm) has been made. We will present performances of the microscope and images on various test samples.

- [1] Freitas, P. P. et al., Proceedings of the IEEE., 2016, vol. 104, n° 10, p. 1894
- [2] Caruso, L. et al., Neuron, 2017, vol. 95, n° 6, p. 1283
- [3] Zheng, C. et al., IEEE Transactions on Magnetics, 2019, vol. 55, n° 4
- [4] Takezaki, T. et al., Jap. J. Appl. Phys., 2006, vol. 45, n° 3B, p. 2251
- [5] Lo, K. H. et al., J. of Nuclear Materials, 2010, vol. 401, p. 143
- [6] Costa, M. et al., IEEE transactions on magnetic materials, 2015, vol. 51, n° 11
- [7] Moulin, J. et al., Appl. Phys. Lett., 2019, vol. 115, n° 12, p. 122406

Analysis of microstructural attributes of bulk martensitic steel surfaces with micrometer-sized grains through magneto-optical Kerr effect

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Within steel industry, there is a tendency to develop novel techniques that provide various measurement information necessary for material analysis within a short time with the same instrument setup. In recent years, development of magnetic characterization techniques arose for investigating microstructural and correlatively mechanical properties of certain steels [1]. However, these techniques provide limited flexibility in terms of surface specific measurements and defect analysis that can become an important part in quality assurance of a produced material.

We present a novel perspective on this matter by utilizing magneto-optical Kerr effect (MOKE) microscopy, which has been proven to be a very powerful and versatile characterization technique [2]. The method allows investigation of magnetic domains and magnetic properties that emerge from the investigated material's surface and allows correlation to the bulk magnetic properties of the material [3]. As such, the method allows high-resolution imaging and investigation of microstructural characteristics such as morphology of both matrix and carbide grains without using etchants that can potentially modify the microstructure and ending results. The method also allows the identification of retained austenite in martensitic steels as this phase in contrast to other matrix phases is non-magnetic. As a result, the method is highly versatile and allows investigation of steels on different aspects of its properties on a microstructural level. In this presentation, we will provide an example of such investigations on conventionally and cryogenically treated high-speed steels [4] with dominating martensitic microstructure. The presented data will provide insight into the correlation of the magnetic information to the microstructure and consequentially to the mechanical properties of such steels.

The novelty of this research lies in the application of MOKE microscopy on steels that are relatively magnetically hard and have a fine grain size (under 10 μ m). The utilization of MOKE microscopy for phase and microstructure investigation on such steels has until now not been performed. Additionally, such an investigation shows a high potential of integrating in-depth magnetic microstructural characterization into industrial applications. Finally, the data shows that MOKE microscopy is applicable to bulk samples of hard magnetic steels with fine microstructure.

[1] J. Liu, J. Wilson, M. Strangwood, C. L. Davis, and A. Peyton, "Magnetic characterisation of microstructural feature distribution in P9 and T22 steels by major and minor BH loop measurements," J. Magn. Magn. Mater., vol. 401, pp. 579–592, Mar. 2016.
[2] J. McCord, "Progress in magnetic domain observation by advanced magneto-optical microscopy," J. Phys. D. Appl. Phys., vol. 48, no. 33, p. 333001, Aug. 2015.

[3] F. Qiu, M. J. Klug, G. Tian, P. Hu, and J. McCord, "Influence of magnetic domain wall orientation on Barkhausen noise and magneto-mechanical behavior in electrical steel," J. Phys. D. Appl. Phys., vol. 52, no. 26, p. 265001, Jun. 2019.

[4] B. Podgornik, V. Leskovsek, and J. Vizintin, "Influence of deep-cryogenic treatment on tribological properties of P/M high-speed steel," Mater. Manuf. Process., vol. 24, no. 7–8, pp. 734–738, Jul. 2009.
The Detection of Higher-Order Multipoles in SQUID-based Magnetometry

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Few areas of measurement are of greater importance for experimental magnetism than the sizing of magnetic moments. It is difficult to imagine modern material magnetism without open-circuit inductive magnetometry. Both of its most popular incarnations the Vibrating Sample Magnetometery (VSM) and the Reciprocating Sample SQUID magnetometery, essentially involve the translation of a sample, subjected to an applied magnetic field, within a set of gradiometer pick-up coils, made of either resistive or superconducting material.

Here we lay out the fundamental principles of gradiometer-based magnetometry focusing on the influence of sample size, shape and magnetic frustration. Our main task is to demonstrate the possibilities for vector measurements, as well as measurements of the higher-than-dipole multipole moments (quadrupole, octupole, etc.) As an illustration, we shall use measurement on a large single crystal of $Mn_3Sn - a$ metallic antiferromagnet with a local octupole arrangement of the sublattice moments, where both strong optical [1] and electronic response has been previously observed [2]. The detection relies on the measurement of coupled flux, while the sample is translated over substantial lengths along the *z*-axis (8 cm), and displaced radially away from the axis, by about 3 mm. A specially-constructed complete orthonormal base-set of point-spread functions is used for fitting.

Indeed, for this system it possible to imprint frustration on the otherwise close to perfect order of octupole moments, by field-cooling in an applied field of 5 T, as shown on fig. 1. This imprinting results in non-zero offset values (at zero applied field) for both dipole components, but rather small imprinting on the main quadrupole component. All imprinting essentially disappears, once the sample is warmed up to RT and zero-field cooled. This set of ZFC/FC measurements of hysteresis loops, clearly demonstrates successful detection of four separate mulipole components dipole-*z*, dipole-*x*, quadrupole-*xx* and quadrupole-*zz*.



Figure 1 : Parallel and perpendicular dipole (a) and quadrupole (b) components for the field-cooled (in 5 T) case, and zero-field-cooled case (c) and (d). The field axis units are T, moment equivalents are in mA.m².

- [1] T. Higo, H. Man, D. B. Gopman, et. al., Nature Photonics, 12, 73-78 (2018)
- [2] S. Nakatsuji, N. Kiyohara and T. Higo, Nature, 527, 212-215 (2015)

Dynamic properties of magnetic Barkhausen noise

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The macroscale magnetization dynamics in conventional ferromagnetic materials has not been fully understood so far. A promising direction of research that should contribute in elucidation of this classical problem is investigation of the magnetization dynamics on a microscale level by measuring Barkhausen noise (BN). BN originates from the abrupt irreversible motion of the magnetic domain walls. Intensity and peak position of the BN signal in soft electrical steels correlate with the corresponding hysteresis parameters: coercive field and losses [1].

We studied the microscale magnetization dynamics for typical soft ferromagnetic materials: thin ribbons, electrical and construction steels. BN was measured simultaneously with the classical hysteresis loops under the controllable magnetization conditions: sinusoidal/triangular waveforms of magnetic polarization/induction J(t)/B(t) or field H(t). The magnetic field H was estimated by a linear extrapolation of two tangential components measured directly by Hall sensors at 1.5 and 2.7 mm above the samples. This method of field determination ensured good measurement accuracy for the magnetically open configuration. Magnetizing frequency was varied in a wide range 0.5-500 Hz fixing the magnetization amplitude on a near-saturation level. Two types of the BN detection coils were used: classical sample-wrapping and an industrial surface-mounted coil.

This report sheds light on a scantily investigated problem of dynamic BN behaviour, which is more complex than that of the hysteresis loops. Recently we have found that total rms intensity of BN in soft magnetic materials (ribbon and electrical non/grain-oriented steels) rises as a square root of the magnetizing frequency, i.e. $\langle U_{rms} \rangle \propto \sqrt{f_{mag}}$.[2]. Dominating square root dependence was explained by stochastic overlapping of independent noise pulses with a partial compensation in the total intensity. Second important finding that rms profile (envelope) of BN is determined by the field rate of change, i.e. $U_{rms}(H) \propto dH/dt$, suggests an idea to reduce the BN envelope by $\sqrt{dH/dt}$. Such normalization fully compensates the envelope differences at low magnetizing frequencies giving a stable quasi-static peak near coercive field, which demonstrate a strong resemblance to the corresponding differential permeability curves [3]. Finally, a Faraday-like normalization principle in the following form $U_{rms}(H) \propto \sqrt{dH/dt}$ has been presented where m is the turn number of the appendice of the appendice of the appendice of the structure permeability curves [3]. Finally, a Faraday-like normalization principle in the following form $U_{rms}(H) \propto m \sqrt{2}$

 $n\sqrt{S}$. dH/dt has been proposed, where *n* is the turn number of the sample-wrapping coil and *S* is a sample cross-section [4].

- [1] O. Stupakov, J. Nondestruct. Eval. 32 (2013) 405-412
- [2] A. Stupakov, A. Perevertov, J. Magn. Magn. Mater. 456 (2018) 390-399
- [3] A. Stupakov, J. Magn. Magn. Mater. 482 (2019) 135-147.
- [4] A. Stupakov, A. Perevertov, J. Magn. Magn. Mater. 498 (2020) 166238 (5 pp)

Magnetic domain observation of unpolished amorphous soft magnetic metal ribbons using MO imaging plates

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To observe domain structures of magnetic metal ribbons with soft magnetic properties by Kerr microscope, those surface have to be polished to obtain mirror surfaces, because polarization properties of the reflected light should deteriorate at the as-prepared surfaces. However, polishing of surfaces could cause of changes of magnetic domain structures. In this study, therefore, we observed magnetic domain structures of unpolished soft magnetic metal ribbons by magneto-optical (MO) imaging technique using MO imaging plates. Amorphous metal ribbon, Metglas 2605SA1, Hitach Metals, Ltd., was used as a sample, which was cut by the metal stamping method. Bi-substituted iron garnet film prepared on glass substrates was used as an MO imaging plate [1]. Magnetic domains were observed by measuring magnetic field distribution quantitatively by the polarization modulation method [2] with an applied magnetic ribbon measured with magnetic fields of 59 - 295 Oe. It was found that the surface of the sample was too rough to measure conventional Kerr images. However, magnetic stripe domain structures evolving with the magnetic field were clearly observed by using the MO imaging plate. In this measurement, it is clearly observed that some of the domains were affected by the magnetic poles appeared at the rough surfaces. Magnetic field distribution quantitatively be appeared at the rough surfaces. Magnetic field distribution glass were affected by the magnetic poles appeared at the rough surfaces.



Figure 1 : An optical image and MO images of amorphous soft magnetic material measured with H of 59 - 295 Oe.

- [1] G. Lou, T. Kato, S. Iwata, and T. Ishibashi, Opt. Mat. Exp. 7 (2017) 2248
- [2] T. Ishibashi et al., J. Appl. Phys., 100 (2006) 093903

Modern FORC data analysis and interpretation approaches

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First-order reversal-curves (FORCs) are a powerful tool, which is increasingly used in material science and nanomagnetism as well as ferroelectricity, geology, archeology, and for spin-transition materials [1-4]. Ideally, it can access microscopic distributions of interaction and coercive fields without the need for lateral resolution [5]. Unfortunately, the reliable data analysis and interpretation poses a major challenge. This is why FORC is often seen as a magnetic fingerprint instead of a measurement method yielding quantitative information. To push past these limitations, we present a fast and user-independent analysis algorithm and possibilities to interpret the resulting FORC diagrams beyond the Preisach model.

We present a new evaluation approach which exploits the diversity of Fourier space to not only speed up the calculation by a factor of 1000 but also move away from the conventional smoothing factor towards real field resolution. By comparing the baseline resolution of the new and the old algorithm we are able to deduce an analytical equation which converts the smoothing factor into field resolution making the old and new algorithm comparable. We find excellent agreement not only for various systems of increasing complexity but also over a large range of smoothing factors. The achieved speed up enables us to calculate a large number of first-order reversal-curve diagrams with increasing smoothing factor allowing for an autocorrelation approach to find a hard criterion for the optimum smoothing factor. This previously computational prohibitive evaluation of first-order reversal-curves solves the problem of over- and undersmoothing by increasing general readability and preventing information destruction.

However, these measured FORC densities are not always straightforward to interpret, especially if the system is interaction dominated and the Preisach-like interpretation of the FORC density breaks down. To understand additional features arising from the interactions in the system, we purposely designed permalloy microstructures which violate the Mayergoyz criteria [7]. These artificial systems allow us to isolate the origin of an additional interaction peak in the FORC density. Modeling the system as a superposition of dipoles allows us to extract interaction strength parameters from this static simulation. Additionally, we suggest a linear relation between integrated interaction peak volume and interaction strength within the system. The presented correlation could be used to investigate the interaction behavior of samples as a function of structural parameters within a series of FORC measurements. This is an important step towards a more quantitative understanding of FORCs which violate the Mayergoyz criteria and away from a fingerprint interpretation.

- [1] J. Gräfe, Phys. Rev. B, 93 (2016) 014406
- [2] F. Groß, Phys. Rev. B, 99 (2019) 064401
- [3] C.-I. Dobrota, J. Appl. Phys., 113 (2013) 043928
- [4] C. R. Pike, Phys. Rev. B, 71 (2005) 134407
- [5] J. Gräfe, Rev. Sci. Instr., 85 (2014) 023901
- [6] D. Cimpoesu, J. Appl. Phys., 125 (2019) 023906
- [7] I. D. Mayergoyz, Phys. Rev. Lett., 56 (1986) 1518

Study of magnetic reversal mechanism in all-oxide-based synthetic antiferromagnets using polarized neutron reflectometry

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Antiferromagnets (AFMs) are playing essential roles in magnetic memory and read-write devices. In the year of 2017, our group reported the antiferromagnetic interlayer exchange coupling (AF-IEC) in La_{2/3}Ca_{1/3}MnO₃/ CaRu_{1/2}Ti_{1/2}O₃(LCMO/CRTO) superlattices (SLs), which is the first time the AF-IEC observed in an all-oxide-based system. The results have attracted wide attention for the delicate step-like magnetic reversal in different layers. However, the microscopic magnetic reversal mechanism is unable to be determined via traditional measurements like MPMS or VSM. Therefore, we use polarized neutron reflectometry (PNR) to detect the detailed reversal procedure and figure out the crucial physical issues.

Consulting the step-like M-H loop of LCMO/CRTO SLs, we performed PNR measurement under various magnetic fields, which is corresponding to different states of magnetic alignment in the sample. We predicted several models and analyzed the data by fitting and calculation. Finally, we found the optimized model and proved the layer-by-layer reversal mechanism in the sample. Our project shields light on the study of micro-scale magnetic structure in the AF-IEC oxide superlattices.

Advanced magnetic microscopy at the ALBA Synchrotron

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One of the main trends in the magnetism roadmap is the study of the magnetic configuration and magnetization dynamics of magnetic nanostructures. The development of magnetic nanostructures requires methods capable of resolving their magnetization. The combination of soft X-ray magnetic imaging with circular magnetic dichroism (XMCD) allows the characterization of nanostructures with chemical sensitivity down to a lateral resolution of 20 nm. The capabilities of the Photo Emission Electron Microscope (PEEM) [1] and transmission X-ray microscopy (TXM) [2] experimental stations of the CIRCE and MISTRAL soft X-ray beamlines at the ALBA Synchrotron (Barcelona, Spain) for magnetic studies will be presented together with representative results from recent experiments.

XMCD-PEEM provides non-invasive element-specific surface magnetic imaging with high spatial resolution and sensitivity (low moment). Thanks to the azimuthal sample rotation and the near grazing X-ray incidence, full 3D magnetization information can be obtained [3], as well as depth information of elevated objects such as cylindrical nanowires [4]. Using X-Ray Magnetic Linear Dichroism (XMLD) contrast, antiferromagnetic domains can also be imaged. A suite of state-of-the-art sample holders and electronics provide a functional sample environment for applying electric and small magnetic fields as well as electrical signals (current pulses) [5]. Dedicated instrumentation has also been developed for time resolved measurements with sub 100 ps time resolution [6].

XMCD-TXM allows to image element specific magnetic domains in full transmission geometry. Element-specific studies of systems with out-of-plane magnetization can be easily performed [7] and by tilting the samples around an axis orthogonal to the photon direction propagation, in-plane magnetized systems can also be addressed [8]. In addition, dual tilt tomography allows reconstructing quantitatively the 3D magnetization vector applying a new iterative algorithm [9]. Sample holders and electronics permit applying high magnetic field pulses (1T) in situ [2]. The ALBA PEEM and TXM are open to external users worldwide through a call for proposals procedure twice a year

[10]

[1] L. Aballe, et al. J. Synchrotron Rad. 22, 745-752 (2015)

- [2] E. Pereiro, et al. J. Synchr. Rad. 16 (2009). A. Sorrentino, et al. Journal of Synchrotron Radiation, 22 (2015)
- [3] S. Ruiz-Gómez, et al. Nanoscale 10, 5566 (2018)
- [4] S. Ruiz-Gomez, et al. Sci. Rep. 8, 16695 (2018)
- [5] M. Foerster, et al; Ultramicroscopy 171, 63 (2016)
- [6] M. Foerster, et al; Nature Communications 8, 407 (2017). L. Aballe, et al; Ultramicroscopy 202, 10 (2019)
- [7] A. Hierro-Rodríguez, et al. Physical Review B 95, 014430 (2017)
- [8] C. Blanco-Roldán, et al. Nature Communications, 6: 8196 (2015)
- [9] A. Hierro-Rodríguez, et al; J. Synchrotron Radiation 25, 1-9 (2018)
- [10] For details and news please visit http://useroffice.cells.es/

A geometry-independent moment correction method for the MPMS3 SQUID-VSM magnetometer

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The sensitivity and automation capabilities of modern superconducting quantum interference device magnetometers are currently unmatched [1]. The measured moment values are, however, prone to deviations from their actual value due to geometric effects, namely sample size, shape, and radial offset [2,3]. A knowledgeable operator will correct measured moment values taking these effects into account. The current procedure for the MPMS3 magnetometer is based on an available simulation tool, valid for both VSM and DC methods [4]. Still, determining the correction factor requires samples with well-defined geometric shapes together with accurate sample dimensions and the usually difficult to determine radial offset [5]. There is no current solution to correct geometry effects of irregular shaped samples. Via an extensive use of the available Sample Geometry Simulator tool [4], in all available geometries, with sample dimensions and radial offset values limited only by the sample chamber volume, we find a systematic relation between the difference of VSM and DC measurements and the corresponding geometric correction factors of the MPMS3 SQUID-VSM device. This relation follows a clear trend, independent of sample size, shape or radial offset, for a given pair of DC scan length and VSM amplitude values. Exploiting this trend, a geometry-independent correction method is here presented and validated in worst-case scenario measurements of metallic Fe powder. In this data set, we observed a maximum deviation to the expected result of 32.5%, which was reduced to just 1.1%, by following the proposed correction method. While the presented methodology requires both DC and VSM data, its ease of use on irregular shaped samples and unknown radial offset values, can be of wide application by the magnetism/ magnetometry community.

- [1] M. Buchner, K. Höfler, B. Henne, V. Ney, A. Ney, J. Appl. Phys. Vol. 124, p. 161101 (2018)
- [2] P. Stamenov, and J. M. D. Coey, Rev. Sci. Instrum. Vol. 77, p. 015106 (2006)
- [3] Quantum Design MPMS Application Note 1500-015 (2010)
- [4] Quantum Design MPMS Application Note 1500-020 (2014)
- [5] G. Morrison, and H-C zur Loye, J. Solid State Chem. Vol. 221, p. 334 (2015)

Influence of the cantilever type on observation of magnetic domains using the magnetic force microscopy in external magnetic fields

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The work is devoted to the development of the magnetic force microscopy (MFM) technique that is currently often applied for observation of magnetic domains on the surfaces of magnetic materials. As a reference sample we used (YSmLuCa)-₃(FeGe)₅O₁₂ garnet film with strong perpendicular anisotropy showing typical crossed stripe magnetic domains with sufficient MFM contrast. Magnetic domain patterns are measured using the commercially available MFM 10 with CoCr 40 nm coating, HA_FM with CoFe 40 nm coating and HA_FM/HC with CoCr 60 nm coating cantilevers.

Our results confirm strong dependence of the cantilever type on the MFM contrast when changing (i) the distance between the cantilever and the sample surface, (ii) the magnitude and sign of the applied external in-plane and out-of-plane magnetic field and (iii) the scanning direction in external magnetic field.

In the first step, we made measurements with all probes at standard conditions and optimal distance in the second MFM pass above the sample surface $d_z = 300$ nm. In the next step, the lift d_z was set up to shorter and longer distances in the 25 nm – 2000 nm range. By comparing MFM images, better magnetic contrast with higher resolution was measured in the case of HA_FM probe. In MFM experiments with external in-plane magnetic field up to $\mathbb{D}1000$ G, the MFM 10 cantilever showed the best results and independence from the scanning direction (iii). Contrary, use of-HA_FM probe resulted in the emergence of areas, where MFM signal was completely lost. These areas were detected in higher in-plane magnetic fields and when scanning direction was chosen to be perpendicular to the magnetic field. Experiments with applied external out-of-plane magnetic field can be performed by all mentioned cantilevers and showed extension of domains with the magnetization parallel to the magnetic field direction at the expense of domains with the magnetization antiparallel.

Due to often difficult interpretation of MFM results the measured magnetic domains were compared with those obtained by other surface-sensitive technique, namely magneto-optical Kerr microscopy (MOKM). Magneto-optical results are further supported by the hysteresis loop captured from the corresponding surface area. Because of dissimilar behaviour of cantilevers in applied external magnetic fields, we also added their bulk hysteresis loops using the vibrating sample magnetometer (VSM) and analyzed basic parameters like coercive field, remnant, and saturation magnetic moment.

In conclusion we can say that selection of suitable probe is crucial for MFM technique. For standard measurements without external magnetic field thin probes like HA_FM (cone radius less than 10 nm) with higher sensitivity to magnetization component perpendicular to the surface and low coercive field (about 0.05 kG) are optimal. On the other hand, they are easily influenced by the external in-plane magnetic field as well as by the direction of scanning. Probes with higher coercive field like MFM 10 (0.3 kG) should be rather used for such types of experiments.

Brillouin scattering investigation of chiral interactions in perpendicularly magnetized epitaxial Ni/Cu(100) films

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The Dzyaloshinskii-Moriya interaction (DMI) has been suggested to be at the origin of the chiral Néel walls that have been surprisingly observed, instead of achiral Bloch walls, in perpendicularly magnetized epitaxial systems, such as Ni/Fe/Cu(001) and Ni/Cu(001) thin films [1,2].

In order to verify the presence of DMI in Ni/Cu(001) epitaxial films, characterized by a thickness-dependent strain, we have exploited the Brillouin light scattering (BLS) technique to investigate thermal spin waves in films of different thickness, namely 5 nm and 2.7 nm, with and without an Au capping layer. Measuring he difference between the frequencies of the Stokes and anti-Stokes peaks in BLS spectra, we were able to confirm the presence of a chiral interaction such as DMI, whose sign is consistent with the right-handed chirality of domain walls observed by spin-polarized scanning electron microscopy. From a quantitative point of view, however, BLS experiments show that the strength of the measured effective DMI is larger for a Ni thickness of 5 nm than for 2.7 nm, contrary to what is expected for interface DMI in ultrathin ferromagnetic films on heavy metal substrates. Moreover, the DMI value is substantially reduced in the absence of the Au capping layer, due to oxidation of the Ni film. The experimental results suggest that the thickness-dependent epitaxial strain might be the symmetry-breaking mechanism that underlies the observed chiral DMI interaction in our, relatively thick, epitaxial Ni/Cu(100) film.

[1] G. Chen et al., Phys. Rev. Lett. 110, 177204 (2013).

[2] B. Boehm, A. Bischof, O. Reich and R. Allenspach, Abstract at JEMS 2019.

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First-Order Reversal Curves Analysis of Fe-based Microwire Arrays

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Magnetically soft FeBSiC microwires (MWs) have a magnetic structure consisting mainly of a longitudinal single domain. The magnetization reversal takes place by fast domain wall motion, producing a single Barkhausen jump and a rectangular hysteresis loop. The MWs' applications are many, with outstanding examples in the fields of sensors and micro electromechanical systems [1]. In practical applications, the MWs are frequently used in arrays, whose magnetic response can be tuned by their separation thanks to the magnetostatic interactions. The First-Order Reversal Curves (FORC) technique has been recognized as a powerful method to study magnetic interactions within and among magnetic materials [2].

In this work FORC analysis has been performed on single and coupled soft microwires with the twofold aim of checking their analogy to ideal magnetic hysterons and getting information on their mutual and internal magnetic interactions.

The task involves several challenges related to the wires' magnetic softness, the fast domain wall propagation, and the Barkhausen jump field dependence on thermal fluctuation. An inductive magnetometer has been adapted for this purpose including software compensation and Fourier differentiation of the magnetization curves [3]. The Fe74B13Si11C2 MWs with core diameter of 18.8 μ m, and glass coating of 4.8 μ m were obtained by Taylor-Ulitovsky method [4].

The single microwire behaves as an ideal magnetic entity whose FORC diagram is identical to the expected Preisach plane. The FORC patterns of the MW arrays cannot be associated to a Preisach patter precisely because, besides the expected magnetostatic interactions, one can clearly identify the fingerprints of the closure domains even when the ends of the MW array are far outside the measurement pickup coil and the hysteresis loop has an ideal rectangular shape. In conclusion, in this research FORC analysis has proved to be an excellent tool to identify and separate the magnetic interactions within systems of bistable or quasi-bistable microwires.

[1] Vázquez, M. (2007). Advanced Magnetic Microwires. In Handbook of Magnetism and Advanced Magnetic Materials (eds H.

- [2] Kronmüller, S. Parkin, M. Coey, A. Inoue and H. Kronmüller).
- [3] F. Béron et al., J. Phys. D. Appl. Phys. 46 (2013).
- [4] V. Kolesnikova et al., J. Magn. Magn. Mater. 508 (2020).
- [5] V.S. Larin et al., J. Magn. Magn. Mater. 249 (2002).

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Rivas²

Symposium 12. Antiferromagnetic spintronics

Colossal spin splitting in a collinear antiferromagnet

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Energy bands of crystals encode quantum, topological and spintronics functionalities. Great interest was devoted to studies of time-reversal symmetry broken energy bands as they are candidates for low dissipation currents [1]. However, further progress is hindered by the lack of realistic systems with robust magnetic symmetry breaking. Here, the common collinear antiferromagnets were anticipated for many decades to be excluded from any macroscopic time-reversal symmetry breaking effects as they exhibit Kramers spin degenerate bands [2]. Our recent prediction of crystal time-reversal symmetry breaking by collinear antiferromagnetism [3] changes this perspective.

In the first part of the talk, we will discuss the basic properties of this new type of antiferromagnetic spin splitting, its local magnetic symmetry origin and symmetry criteria for its emergence. The anisotropic magnetisation densities result from a combination of collinear antiferromagnetism and nonmagnetic atoms. Our antiferromagnetic spin splitting properties starkly contrast those of conventional spin-orbit interaction spin splitting. It can preserve spin quantum number [3], is of exchange origin reaching colossal eV values [3,4] unparalleled in systems without magnetic dipole and can materialize in centrosymmetric potentials [3, 5, 6].

In the second part of our talk, we will discuss the experimental implications of our antiferromagnetic splitting. We will show that this antiferromagnetic spin splitting generates a crystal Hall effect which was recently observed in $RuO_2[3,7]$. We will show that the antiferromagnetic spin splitting effects are surprisingly strong. The spin spitting of more than 1eV predicted in RuO_2 is larger than the record values reported in Rashba systems. Furthermore, we predict spin current angle in RuO_2 to reach 34 degrees, a value three times larger than the largest spin current angles found in a high-throughput scan of 20 000 materials [8].



Figure 1: Colossal spin splitting in Ruthenium dioxide calculated from first principles.

- [1] Šmejkal, L., Mokrousov, Y., Yan, B. MacDonald, A. H., Nat. Phys. 14, 242 (2018)
- [2] Šmejkal, L., Železný, J., Sinova, J. Jungwirth, T., Phys. Rev. Lett. 118, 106402 (2017)
- [3] Šmejkal, L., González-Hernández, R., Jungwirth, T. Sinova, J., Sci. Adv. 6, eaaz8809 (2020)
- [4] Ahn, K.-H., Hariki, A., Lee, K.-W. Kuneš, J. Phys. Rev. B 99, 184432 (2019)
- [5] Hayami, S., Yanagi, Y. Kusunose, H. J. Phys. Soc. Japan 88, 123702 (2019)
- [6] Yuan, L.-D., Wang, Z., Luo, J.-W., Rashba, E. I. Zunger, A. Phys. Rev. B 102, 014422 (2020)
- [7] Feng, Z. et al. Observation of the Crystal Hall Effect in a Collinear Antiferromagnet. arxiv (2020)
- [8] González-Hernández, R. et al. Magnetic Spin Hall Effect in Collinear Antiferromagnets. arxiv (2020)

A journey into strain and electric-field controlled antiferromagnetic spin textures in BiFeO₃

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As antiferromagnetic materials are moving in the focus for low-power spintronic applications [1], it becomes crucial to deterministically control the antiferromagnetic order at a submicron scale to obtain functional devices. A possible route towards this goal is to use multiferroic materials in which antiferromagnetic and ferroelectric orders are intimately connected. BiFeO₃ is the archetypal room-temperature multiferroic with high ordering ferroelectric and antiferromagnetic temperature. In addition to its super-exchange interaction leading to G-type antiferromagnetic order, the antisymmetric magnetoelectric interaction gives rise to a spin cycloid order with a period of 62-64 nm [2,3] in the bulk. BiFeO₃ can be grown epitaxially in thin films heterostructures on various substrates imposing different strains of a few percent both in the compressive and tensile sides [4]. Different types of cycloids are predicted and inferred from Mossbauer spectroscopy [5,6].

Using a non-invasive scanning magnetometer [7,8] based on a single nitrogen-vacancy (NV) defect in diamond in combination with piezoresponse force microscopy (PFM) gives us the possibility to image the antiferromagnetic landscapes that are statically tuned by the epitaxial strain. We find spin textures such as bulk-like and exotic spin cycloids or collinear antiferromagnetic ordering. The observation of the two types of cycloids is confirmed by resonant X-ray scattering. Furthermore, we manipulate the ferroelectric order with the electric field of the PFM tip, and observe the magnetoelectric coupling on the local scale with the NV microscope. We bring to light the interplay between ferroelectric domain structures with the coupled antiferromagnetic order.

- [1] T. Jungwirth et al., Nature Nanotech. 11, 231–241 (2016)
- [2] I. Sosnowska et al., J. Phys. Chem. 15, 4835–4846 (1982)
- [3] D. Lebeugle et al., Phys. Rev. Lett. 100, 227602 (2008)
- [4] I.C. Infante et al., Phys. Rev. Lett. 105, 057601 (2010)
- [5] D. Sando et al., Nature Mater. 12, 641 (2013)
- [6] A. Agbelele et al., Adv. Mater. 29, 160327 (2017)
- [7] L. Rondin et al., Rep. Prog. Phys. 77, 056503 (2014)
- [8] I. Gross et al., Nature 549, 252-256 (2017)

Spin Hall magnetoresistance in antiferromagnetic insulators

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Antiferromagnetic materials promise improved performance for spintronic applications. Compared to ferromagnets, they are robust against external magnetic field perturbations and allow for faster magnetization dynamics. The direct observation of their antiferromagnetic state, however, is challenging due to the lack of a macroscopic magnetization. Here, we present the spin Hall magnetoresistance (SMR) effect [1] as a versatile tool to directly probe the complex spin structure via a simple electrical transport experiment. We investigate the easyplane antiferromagnetic insulators α -Fe₂O₃ (hematite) [2] and NiO [3] in bilayer heterostructures with a top electrode of the heavy metal Pt. We measure the longitudinal and transverse resistivities while rotating an applied magnetic field of up to 17 T in three orthogonal planes and observe characteristic modulations consistent with the SMR effect [2,3,4]. For out-of-plane magnetotransport measurements, we find indications for a multidomain antiferromagnetic configuration in α -Fe₂O₃ whenever the field is aligned along the film normal [2]. For in-plane field rotations, we clearly observe sinusoidal resistivity oscillations characteristic for the SMR effect due to a coherent rotation of the Néel vector in both α -Fe₂O₃ and NiO [2,3]. We analyze their amplitude as well as their phase and compare the data to earlier results from the prototypical collinear ferrimagnet Y₃Fe₅O₁₂ [1,4,5]. We explain the observed magnetic field dependence in a comprehensive model, based on two magnetic sublattices and taking into account magnetic field-induced modifications of the antiferromagnetic domain structure [4]. In α -Fe₂O₃/Pt, moreover, we find an unexpectedly large SMR amplitude of 2.5x10⁻³, twice as high as for prototypical ferrimagnetic Y₃Fe₅O₁₂/Pt heterostructures [1,4]. The SMR effect saturates at much smaller magnetic fields than in antiferromagnetic NiO/Pt [3], making the α -Fe₂O₃/Pt system particularly interesting for room temperature antiferromagnetic spintronic applications [2].

- [1] H. Nakayama et al., Physical Review Letters 110, 206601 (2013)
- [2] J. Fischer et al., Physical Review Applied 13, 014019 (2020)
- [3] J. Fischer et al., Physical Review B 97, 014417 (2018)
- [4] S. Geprägs et al., Journal of Applied Physics 127, 243902 (2020)
- [5] M. Althammer et al., Physical Review B 87, 224401 (2013)

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Efficient magnonic spin transport in insulating antiferromagnetic thin films

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With spin dynamics in the high GHz to THz regime, stability in the presence of external magnetic fields, and a lack of stray fields, antiferromagnetic materials are positioned to become key in future low power spintronics devices [1]. Here, we use develop quality bulk crystals and thin films of hematite (α -Fe₂O₃) (< 500 nm) of different orientations to study spin transport [2-4]. In addition to encoding and reading information stored in the Néel vector, the efficient transfer of information is crucial for integration of antiferromagnets into devices. Recently, we demonstrated that a diffusive magnon current can be carried over micrometres in antiferromagnetic single crystals, but such crystals are not suitable for spintronic devices [4]. Despite theoretical works investigating the mechanisms for the long-distance propagation of pure spin currents carried by the antiferromagnetic order [5, 6], studies on thin film antiferromagnets making use of single-frequency or broadband excitations have failed to achieve efficient transport of angular momentum by magnons [7]. Making use of hematite thin films, a robust magnon current can propagate with intrinsic diffusion lengths of hundreds of nanometres [8]. The efficiency of the transport mechanisms can be tuned by field cycling of the domain structure, the growth orientation, and the relative orientations of the magnetic field and magnetic anisotropies [2,8]. The manner by which the stabilisation of the antiferromagnetic domain structure results in frequency dependent length scales and proves to be critical in the magnon transport will be discussed [8].

- [1] T. Jungwirth et al., Nat. Phys. 14, 200-203 (2018)
- [2] R. Lebrun et al., Comm. Physics 2 50 (2019)
- [3] A. Ross et al., arXiv:2001.03117 (2020)
- [4] R. Lebrun, A. Ross et al., Nature 561, 222-225 (2018)
- [5] S. Takei et al., Phys. Rev. B, 90, 94408 (2014)
- [6] S. Bender et al., Phys. Rev. Lett. 19, 056804 (2017)
- [7] H. Wang et al., Phys. Rev. B 91, 220410 (2015)
- [8] A. Ross, R. Lebrun et al., Nano Letters 20 1, 306-313 (2020)

Current-induced switching in Pt/Mn₂Au bilayers grown by molecular beam epitaxy

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Recent progress in spintronics has been exploited to develop antiferromagnet–based magnetic memory. Demonstration of current-induced switching in antiferromagnets (AFMs) confirmed theoretically predicted magneto-transport properties of AFM materials.¹⁻² Current-induced triangular-shaped Hall resistance change has been recently reported in Pt/Mn₂Au grown by sputtering.³ However, most recent studies show that the origin of the triangular switching may be related to the thermal artifacts of patterned metal structures rather than with current-induced switching of AFM.⁴⁻⁶

In our study we demonstrate electrical switching in Pt/Mn₂Au bilayers grown on MgO(111) by molecular beam epitaxy. Mn₂Au layers with a thickness of 6nm were deposited by co-deposition of Mn and Au and capped with Pt layer with a thickness of 1nm. The eight terminal devices for current-induced switching in Pt/Mn₂Au were fabricated using optical lithography, ion-beam etching and lift-off process. The switching protocol was following. First a set of twenty 1ms-long current pulses were applied along [1-10] MgO direction (Fig. 1(a), green). After a 10s of delay a reading current was applied (I_{H} =1mA) and the transversal Hall voltage (V_{H}) was recorded (Fig 1(a)). Such a series was repeated five times. Then, the series of pulses was applied along the orthogonal direction (Fig. 1(a), blue). To determine the switching dependence on the current density we repeated measurements for different current densities, j ranging from 3.15×10^{11} A/m² to 3.54×10^{11} A/m². For j < 3.3×10^{11} A/m² we noted a step-like change of transverse resistance after the application of the first current pulse in [1-1 0]([11-2]) MgO direction. This step-like change of ΔR_{xy} was followed by a subtle increase (decrease) of the transverse resistance for subsequent pulses applied along the same path. Similar switching characteristics was recently observed for small switching currents in Pt/NiO and was explained in terms of redistribution of AFM domains via domain wall motion.⁴ For $j = 3.3 \times 10^{11} \text{A/m}^2$ we observed a step-like, non-decaying(growing) switching in Pt/Mn₂Au bilayer. This rectangular switching characteristics suggests a saturation of the Néel order after the application of one set of current pulses. Further increase in *j* results in the appearance of the saw-tooth feature in switching characteristics with the reversed sign of switching. This behavior dominates for $j>3.4x10^{11}$ A/m².As it was shown in previous studies sawtooth behavior observed for substantial current densities may be an artifact not related to the AFM switching but describes thermal effects related to the Pt layer.⁴⁻⁶ The present work reveals the feasibility of current switching in Pt/Mn₂Au(110)/MgO(111) grown by molecular beam epitaxy. We demonstrate that a proper tune of current density results in rectangular switching in the bilayer structure.



Figure 1.

- 1. J. Železný, et al. PRL 113, 157201 (2014)
- 2. P. Wadley, et al., Science 351, 587 (2016)
- 3. X.F. Zhou, et al. PRA 11, 054030 (2019)
- 4. L. Baldrati et al., PRL 123, 1772 (2019)
- 5. Y. Cheng et al., arXiv:1906.04694
- 7. C. C. Chiang, PRL 123, 227203 (2019)

This work was supported by the "Antiferromagnetic proximity effect and development of epitaxial bimetallic antiferromagnets – two routes towards next-generation spintronics" project which is carried out within the Homing programme of the Foundation for Polish Science co-financed by the European Union under the European Regional Development Fund.

Towards multistate switching of antiferromagnetic order in NiO

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Antiferromagnetic spintronics has recently gained a lot of interest. The absence of a net magnetization and terahertz-frequency resonances make antiferromagnets good candidates for numerous applications, ranging from data storage devices to terahertz radiation sources. Many of these prospected applications benefit from having electronic control over the orientation of the antiferromagnetic order parameter.

It has been demonstrated that the antiferromagnetic order can be switched electronically in antiferromagnetic insulators (e.g. nickel-oxide, NiO) using an adjacent heavy metal layer (often platinum) [1, 2]. In these experiments, a spin-current is generated from a charge-current using the spin-Hall effect in the heavy metal layer. This spin-current is injected into the antiferromagnet where it reorients the antiferromagnetic order parameter between the anisotropy directions of the antiferromagnet. This way, by using two current paths in the Pt layer that are aligned with the two perpendicular (in-plane) anisotropy directions of a biaxial antiferromagnet, the order parameter can be switched between two directions. However, depending on the crystal orientation and choice of substrate, NiO exhibits either two or three anisotropy directions [1]. Therefore, one could expect that, depending on the exact growth parameter, the antiferromagnetic order can be switched between three directions that are separated by 60 degrees, similar to demonstrations in hematite (Fe- $_2O_3$) [3].

Here we investigate this possibility of switching the antiferromagnetic order in NiO/Pt bilayers between either two 90 degrees or three 60 degrees separated states. Moreover, the impact of the NiO growth parameters (e.g. substrate and crystal-orientation) on the electrical switching is studied. Experiments (see figure 1) show that it seems possible to switch the order-parameter between two 90 degrees rotates states in NiO where three in-plane easy axes are expected [4]. For switching along three directions we use devices similar to the one shown in figure 2. In this contribution we discuss the results of both 90 degrees and 60 degrees experimental geometries. Moreover, we interpret our observations in light of non-magnetic contributions to the observed switching-like behavior, which recently casts doubt on the previously reported observations [5, 6, 7]. Our research adds to the understanding of the interaction between the anisotropy of thin film antiferromagnets and the electrical control of their antiferromagnetic order. Moreover, it contributes to the understanding and development of electronic control of the antiferromagnetic order parameter and builds towards antiferromagnetic-based storage devices.



Figure 1 : Switching experiment between two (90 degrees separated) directions in NiO. Note that this measurement is not taken with the device in figure 2.

- [1] Chen, X. Z., et al. (2018) Phys. Rev. Lett. 120, 207204
- [2] Moriyama, T., et al. (2018) Sci Rep 8, 14167
- [3] Cheng, Y., et al. (2020) Phys. Rev. Lett. 124, 027202
- [4] Wang, Z., et al. (2018) Appl. Phys. Lett. 112, 252404
- [5] Churikova, A., et al. (2020) Appl. Phys. Lett. 116, 022410
- [6] Matalla-Wagner, T., et al. (2019) arXiv 1910.08576
- [7] Chiang, C.C., et al. (2019) Phys. Rev. Lett. 123, 227203



Figure 2 : Micrograph of the device used for switching the antiferromagnet along three (60 degrees separated) directions, indicated by the coloured arrows. V+ and V. indicate the probing contacts.

Spin Transport Experiments in the Easy-plane Phase of the Antiferromagnetic Insulator Hematite

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Electrical control of antiferromagnetic domain walls

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The prospect of using antiferromagnets as the main component in logic devices has gained a lot of interest. Antiferromagnets offer many benefits over their ferromagnetic counterparts: terahertz dynamics, no device crosstalk, and robustness to external fields. However, the latter property makes them difficult to control and has hindered their potential for use in spintronics devices.

Recently, electrical switching of antiferromagnets has been demonstrated using current-induced spin-orbit torques due to bulk crystal symmetry, or spin-transfer torque from a neighbouring heavy metal layer. Changes to the magnetic state are typically detected electrically in the transverse anisotropic magnetoresistance, which depends on the angle between the antiferromagnetic Néel vector and the direction of the probe current. This method is effective for measuring the average Néel vector orientation, but provides limited information on the microscopic magnetic texture.

Here, we study current-induced modification of the antiferromagnetic domain structure in CuMnAs microdevices using x-ray photoemission electron microscopy (XPEEM). Magnetic contrast is obtained due to x-ray magnetic linear dichroism, *i.e.* dependence on the angle between the magnetic moments and the incident x-ray polarisation. Depending on the incidence angle, contrast of the magnetic domains or the domain walls between them can be seen.

Our results show a current-induced motion of both 90° and 180° domain walls, in a direction which depends on the polarity of the current pulse (Fig. 1). This represents an important step towards the development of low power antiferromagnetic domain wall memory devices, analogous to racetrack memory which uses ferromagnetic domain walls. Furthermore, 180° domain walls are topologically protected making them particularly good candidates for non-volatile applications.



Figure 1: XPEEM images of the magnetic domain structure in a Wheatstone bridge device. The incident linearly polarised xrays direction is shown by the blue arrow with polarisation perpendicular. Magnetic domains along the easy axes (white arrows) show maximum contrast. Shown are three stages of domain wall movements; **a)** before applying current pulse, **b)** after 17 MA/cm² current pulse in direction (red arrows), and **c)** after 17 MA/cm² current pulse with opposite polarity.

Magnetic field control of antiferromagnetic domain walls in a thermal gradient

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Theoretical investigations have shown that domain walls (DW) in antiferromagnets can move much faster than their counterparts in ferromagnets due to their low effective mass and the absence of a Walker breakdown limit [1]. Current-driven DW motion in antiferromagnets can be achieved via both spin-transfer and spin-orbit torques, but other approaches that do not require charge currents are also worth exploring, such as using asymmetric field pulses, magnonic currents or thermal gradients.

It was demonstrated theoretically that an antiferromagnetic DW in a thermal gradient moves towards hotter regions [2], as it is the case in ferromagnets. This motion was explained using thermodynamic arguments, namely that it maximizes the entropy of the system or, equivalently, it minimizes its free energy [2]. An analytical equation for the DW motion was derived based on this so-called entropic torque. Another possible mechanism for thermally driven DW motion is the transfer of angular momentum from magnons to the wall when passing through it, leading to an adiabatic torque on the domain wall that drives it against the magnon flow, i.e., towards hotter regions [3]. In compensated antiferromagnets, thermal magnons do not carry angular momentum on average and, therefore, this mechanism has, in principle, no effect on the DW. However, in ferrimagnets, it has been shown recently that, above the Walker breakdown, this magnonic torque changes sign around the compensation temperature, driving the DW towards the hot end above it and towards the cold end below it [4].

In the present work we use atomistic simulations to show that an AF DW in a thermal gradient experiences a force towards the cold region upon the application of a uniform magnetic field along the easy axis. This force increases with the strength of the applied field and it overcomes the entropic force for sufficiently high values. The force is proportional to the thermal gradient and it shows a linear dependence with the net magnetic moment of the domain wall induced by the field. A frequency analysis reveals that the origin of this effect lies on the increase of the DW reflectivity due the field-induced sizable break of antiferromagnetic order inside it, which turns it into an efficient barrier for magnons that transfer linear momentum to the domain wall when they are reflected on it.

- [1] O. Gomonay, T. Jungwirth and J. Sinova. Phys. Rev. Lett. 117, 017202 (2016)
- [2] S. Selzer et al., Phys. Rev. Lett. 117, 107201 (2016)
- [3] S. K. Kim and Y. Tserkovnyak, Phys. Rev. B 92, 020410 (2015)
- [4] A. Donges et al., Phys. Rev. Research 2, 013293 (2020)

Finite bias TMR and STT effects in Mn₃Ga-based ferrimagnet MTJs

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While ubiquitous in bulk resonance devices, ferrimagnets are just beginning to make their way into spintronic devices, as both insulating and metallic components. Here we report on first principles calculations of spin transport in epitaxial magnetic tunnel junctions (MTJs) based on ferrimagnetic Mn₃Ga [1] electrodes and MgO barrier. We consider two types of stacks: one in which the polariser electrode is bcc Fe (Fig.1a) and another fully-ferrimagnetic, with Mn₃Ga polariser as well. Solving self-consistently the ballistic transport problem for the non-equilibrium spin density [2], as implemented in the Smeagol code [3], in a scattering region extended to some 8 nm into the Mn₃Ga electrode, we find a long-range spatial oscillation of the spin-transfer torque (STT) decaying on a length scale of a few nm (Fig.1 c,d), both in linear response and for finite bias. This STT oscillation is understood from the bulk electronic structure of Mn₃Ga and is robust against variations in the stack geometry: we analyse the effect of the barrier thickness, the interface spacing and variations of the *c* lattice constant of the tetragonal Mn₃Ga. We also investigate the TMR effect in these MTJ stacks as a function of the applied bias voltage and find asymmetry and sign-changing in the range ±1V (Fig.1 e,f), in accordance with experimental observations for similar ferrimagnetic junctions [4]. In view of this bias asymmetry, our calculations suggest that further efforts are justified on the Fermi-level engineering of ferrimagnetic electrodes, by means of both composition and substrate-induced strain, for demanding spintronic applications.



Figure 1: (a) Schematic of the Fe/MgO/Mn₃Ga junction studied and **(b)** the unit cell of tetragonal Mn₃Ga depicting also the magnetic moments localised on the Mn₃ and Mn₂ magnetic sublattices. In **(c, d)** are atomically-resolved linear-response STT on Mn₃ and Mn₂, respectively, starting from the MgO interface. In **(e)** are the calculated current voltage characteristics for the two spin alignments of the junction in panel (a) and in **(f)** is the corresponding TMR coefficient.

[1] K. Rode, N. Baadji, D. Betto, et al. Phys. Rev. B, vol. 87, p. 184429 (2013)

[2] M. Stamenova, I. Rungger, S. Sanvito, et al. Phys. Rev. B, vol. 95, p. 060403(R) (2017); I. Rungger, A. Droghetti and M. Stamenova, Non-equilibrium Green's functions methods for spin transport and dynamics. In: Andreoni W., Yip S. (eds) Handbook of Materials Modeling, Springer (2018)

[3] A. R. Rocha and S. Sanvito, Phys. Rev. B, vol. 70, p. 094406 (2004)

[4] K. Borisov, D. Betto, Y.C. Lau, et al. Appl. Phys. Lett., vol. 108, p. 192407 (2016)

Epitaxial growth and electrical control of antiferromagnetic Mn₂Au films

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Having the advantages of immunity to external magnetic fields and fast spin dynamics, antiferromagnetic materials open a new door towards the next generation of high-speed data storage devices. Here, we optimized the growth conditions of epitaxial Mn₂Au thin films. The antiferromagnetic domain configuration of the films is directly mapped exploiting x-ray magnetic linear dichroism. The films are consequently patterned into star-shaped devices for electrical switching measurements. We observe a change of resistance, which implies current-induced switching of Néel vectors. The Néel vector switching offers a platform to investigate the microscopic mechanism of the magnetization dynamics.

Inertial Domain Wall Motion Driven by Staggered Spin-Orbit Fields in Uncompensated Antiferromagnets

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Magnetization switching plays a central role in the field of magnetism-based devices. A concrete example is the case of magnetic textures such as domain walls (DW) which play the role of carriers of information. Therefore, a precise control of their mobility becomes crucial for potential applications such as racetrack device non-volatile memories [1], sensors and logic-devices [2,3]. On the other hand, inertial dynamics for magnetic solitons could potentially lead to a lower amplitude of the driving forces. Both ferromagnets (FM) and antiferromagnets (AFM) can host multiple types of these textures. While the DW dynamics in FM have been investigated in depth, only recently a mechanism to excite its movement in AFM was proposed [4] based on the fact that when magnetic sublattices form inversion partners, it is possible to generate a Néel spin-orbit (SO) field through the injection of an electric current.

In this work, we have evaluated the dynamics of a one-dimensional Néel-like DW in an embedded FM layer of the uncompensated layered AFM Mn₂Au induced through the Néel SO field. For this purpose, we have used atomistic spin dynamics simulations for the real crystalline structure in combination with an analytical model based on the Lagrangian formalism together with the collective coordinates approach [5]. Due to the complexity of the system when theoretically dealing with the problem, a couple of staggered AFM-coupled Néel vectors have been introduced. Taking this into account, we have evaluated three consecutive correlated SO-field based regions: a rest breaking acceleration regime driven by a ramped stimulus, a steady-state process at constant speed achieved after a brief adaptation time, and a friction-driven motion once the field abruptly turns off. Traces of the inertial dynamics of the magnetic texture and a great stability of the DW even at speeds very close to the maximum magnon group velocity of the medium have been observed. The distance travelled by the texture once the excitation stopped is 5-10 times greater than the DW width at rest for fields of the order of 10-60 mT. Simulations and theory show great quantitative correspondence in the entire SO-field based excitation regime. These results open the door to exploit this inertial phenomenon in technological applications such as racetrack memories to decrease power consumption. Moreover, the theoretical approach regarding the construction of the Néel order parameter can be extrapolated to AFM with complex structures to analyze the dynamics of magnetic textures.

^[1] Parkin, S. S., et al. Science, 2008, vol. 320, no 5873, p. 190-194

^[2] Allwood, D. A., et al. Science, 2005, vol. 309, no 5741, p. 1688-1692

^[3] Mattheis, R., et al. J. of Appl. Phys., 2012, vol. 111, no 11, p. 113920

^[4] Železný, J., et al. Phys. Rev. Lett., 2014, vol. 113, no 15, p. 157201

^[5] Schryer, N. L., et al. J. of Appl. Phys., 1974, vol. 45, no 12, p. 5406-5421

Electrically Tunable Detector of THz-Frequency Signals Based on an Antiferromagnet

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Terahertz (THz)-frequency detectors have a great potential for non-destructive testing, security and telecommunications applications [1], since the THz radiation is non-ionizing, and the photons of the THz frequency have relatively low energy. The main difficulty in the use of the THz frequency radiation is the absence of portable solid-state generators and receivers capable of generating narrow-band quasi-monochromatic signals with continuously tunable frequency in that frequency range [2-3].

Spintronics of antiferromagnets (AFMs) has a great potential for the development of practical devices for generation and reception of narrow-band THz-frequency signals [4], because, due to the presence of a very strong internal exchange field, the magnetization dynamics in AFMs is ultra-fast, and there are no stray magnetic fields of a dipolar origin. The output power of a single AFM-based THz-frequency spin-Hall oscillator (SHO), was theoretically calculated in [5], where it was shown that this power increases with the increase of the generation frequency. Although the output power of a single AFM SHO, which was evaluated to be of the order of nanowatts [5], is not sufficient for most practical applications, the reception of quasi-monochromatic THz-frequency signals using an AFM SHO working in a sub-generation- threshold passive regime is quite possible.

In this work, we present both theoretical and numerical data illustrating the possible performance of an electrically tunable resonance receiver of THz-frequency signals based on an antiferromagnetic (AFM) crystal having the frequency of the antiferromagnetic resonance (AFMR) in the THz-frequency range. The receiver is based on a bilayer of a uniaxial AFM crystal and a heavy metal (HM), and the conversion of the received AC signal into an output DC signal is done using the inverse spin Hall effect in the (AFM/HM) bilayer. An additional bias DC current in the HM layer can be used for the tuning of the AFMR frequency of the system, and for a partial regeneration of the system losses. The AFMR eigenfrequency f_{osc} can be continuously tuned in a substantial-frequency interval (of about 0.4 THz) by varying the magnitude of the DC electric current. It is shown, that the AC sensitivity of the proposed AFM/HM-based detector is comparable to the sensitivity of modern sub-millimeter-wave detectors based on the Schottky and Gunn diodes and that the received DC signals are well above the level of the thermal noise for the AC signals having a power of the order of several microwatts.

- [1] A. Y. Pawar, D. D. Sonawane, K. B. Erande, and D. V. Derle, Drug Invent. Today 5, 157 (2013)
- [2] R.A. Lewis, J. Phys. D: Appl. Phys. 47, 374001 (2014)
- [3] B.S. Williams, Nature Photon. 1. 517-25 (2007)
- [4] Baltz V. et al // Rev. of Mod. Phys. vol. 90. 015005 (2018)
- [5] Sulymenko O. et al. // Phys. Rev. Appl. vol. 8. 064007 (2017)

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Current induced switching of the Néel vector in CoO(001)/Pt bilayers

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Antiferromagnets (AFMs) are promising materials for spintronics, thanks to advantageous properties compared to ferromagnets. They promise faster operation, enhanced stability with respect to interfering magnetic fields and higher integration due to the absence of long range magnetic coupling via stray fields. However, the absence of a net magnetic moment makes manipulation using magnetic fields challenging [1]. For the use of AFMs, one requires efficient electrical writing and reading. Recently there have been reports on current-induced switching of the Néel vector orientation and it was shown in the insulating AFM NiO/heavy metal Pt thin film system that there are multiple effects contributing to the electrical signal. These include switching of the magnetic Néel order, whichs mechanism is debated in terms of origin and efficiency [2-5], but also non-magnetic signals [6]. While NiO has been intensively studied, key information such as the torque strength and the identification of non-magnetic contributions to the switching signals are missing. Using other AFMs such as CoO, that we study here [7], entail a number of advantages: We first show that, due to the compressive strain by the MgO substrate, a fourfold in-plane magnetic anisotropy of the CoO layer with two easy axes in the (001) plane is favored and the spin flop field is accessible. Such a system with two orthogonal stable states is ideal for applications where the orientation of n is read by spin Hall magnetoresistance (SMR) [8-10]. Furthermore, we achieve electrical switching and probe its symmetry. By looking at the switching above and below the Neél temperature in the CoO/Pt bilayer we can show that this switching is of magnetic origin, as the signal related to the antiferromagnetism disappears above Néel temperature, which in CoO is easily accessible around room temperature. For the switching 8-arms Hall stars devices with the pulsing arms oriented along the [110] and [-110] easy axes directions are used (Fig. 1). Before the pulses **n** is aligned along [110]. When current pulses are applied along 3-2 (initial state $n \parallel jpulse$), the transverse resistance drops in a step-like fashion, indicating a current-induced 90° **n** rotation (Fig. 1a). Performing a MR scan with field along 4-1 after the current pulses, shown in Fig. 1b, yields a field-induced spin flop transition of *n* back to the initial state (along [110]). The non-magnetic signal, possibly related to a local annealing process and electromigration in the Pt layer, does not disappear above the Néel temperature. Finally, the anisotropies in CoO allow us to quantify the current field equivalence of the current-induced torques, by comparing the effects of the field and the current pulses on the reorientation of *n* in the CoO. With this we show that for the switching of AFMs currents are much more efficient than magnetic fields.



Figure 1 : Symmetry of the current-induced switching. (a) Current-induced spin flop transition. (b) Field-induced spin flop, reseting *n*.

- [1] V. Baltz et al., Rev. Mod. Phys. 90, 015005 (2018).
- [2] L. Baldrati et al., PRL 123, 177201 (2019).
- [3] T. Moriyama et al., Sci. Rep. 8, 14167 (2018).
- [4] X. Z. Chen et al., PRL 120, 207204 (2018).
- [5] P. Zhang et al., PRL 123, 247206 (2019).

- [6] T. Matalla-Wagner et al., arxiv:1910.8576.
- [7] L. Baldrati et al., arXiV:2003.05923.
- [8] X. Z. Chen et al., PRL 120, 207204 (2018).
- [9] H. Nakayama et al., PRL 110, 206601 (2013).
- [10] L. Baldrati et al., PRB 98, 024422 (2018).

Poster

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Current-induced Néel Order Switching in Antiferromagnetic Thin Films

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Antiferromagnetic insulators (AFMI) are promising candidates for spintronics. They have the potential for ultrafast operation due to their resonance frequency in the THz regime, they are insensitive against external magnetic fields and scalable, thanks to the absence of stray fields. At the same time, antiferromagnets with a demonstrated spin transport length of 40 μ m, are of interest for spin-logic based devices [1].

Electrical read-out and writing of information stored in the Néel vector **n** is a key step for applications. While reading of the orientation of **n** can be achieved electrically via the spin Hall magnetoresistance (SMR) [2-4], the mechanism of electrical writing of n is still debated, in terms of both origin and efficiency [5–9]. Here we show, by electrical measurements and direct imaging, that one can electrically switch the antiferromagnetic moments with current pulses. The current is applied to a heavy metal (HM) to achieve spin accumulation by the spin Hall effect at the AFMI-HM interface, exerting anti-damping like torques on the AFMI [5, 6]. The resulting switching was investigated electrically and imaged by direct imagine via x-ray magnetic linear dichroism – photoemission microscopy. In this study we probe the full current-density range that leads from non-deterministic switching below a certain current threshold to deterministic switching above that threshold. Our results can be explained with a theoretical model, which attributes the switching to a motion of domain walls. Domain walls are directly affected by spin torques and a pondermotive force by a variation in the effective magnetic anisotropy. Besides these effects, the magnetoelastic effect due to Joule heating seems to play a role as well in AFM like CoO and NiO [7,8]. High current density pulses can additionally affect the measured resistance by electromigration effects, unrelated to the magnetic order as observed in a control sample with Pt only. This results in a signal consisting of two components, which can be distinguished by their shape ("triangular-like" and "step-like") and these components correspond to non-magnetic electromigration and the magnetic switching contribution respectively [9-12]. This must be considered to correlate the measured transverse resistance to the Néel order orientation.

Based on the reading of antiferromagnetic domains via SMR and the writing of AFM domains via spin torques, these materials can be utilized to realize AFM memory as a future application.



Figure 1: The angles and frequencies of the Néel vector precession for the FiM with a) easy-plane, b) easy-axis.

- [1] R. Lebrun, et al., Nature 561, 222 (2018)
- [2] L. Baldrati, et al., Phys. Rev. B 98, 024422 (2018)
- [3] G. R. Hoogeboom, et al., Appl. Phys. Lett. 111, 052409 (2017)
- [4] J. Fischer, et al., Phys. Rev. B, 97, 014417 (2018)
- [5] T. Moriyama, et al., Sci. Rep. 8, 14167 (2018)
- [6] X. Z. Chen, et al., Phys. Rev. Lett. 120, 207204 (2018)



Figure 2: Phase diagram of instabilities in easy axis ferrimagnet under STT vs spin uncompensation.

- [7] P. Zhang, et al., Phys. Rev. Lett. 123, 247206 (2019)
- [8] L. Baldrati, et al., ArXiv: 2003.05923 1 (2020)
- [9] L. Baldrati, et al., Phys. Rev. Lett. 123, 177201 (2019)
- [10] T. Matalla-Wagner, et al., ArXiv:1910.08576 1 (2019)
- [11] C. C. Chiang, et al., Phys. Rev. Lett. 123, 227203 (2019)
- [12] A. Churikova, et al., Appl. Phys. Lett. 116, 022410 (2020)

Investigation of non-collinearity in compensated ferrimagnetic half-metallic thin films by XMCD

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Mn₂Ru_xGa (MRG) is a half-metallic compensated ferrimagnet crystallising in a cubic inverse Heusler structure (*XA*) with a substrate-induced biaxial strain of about 1% leading to perpendicular magnetic anisotropy (PMA) of the net moment. The Mn atoms occupy two different crystallographic sites, Wyckoff positions 4*a* and 4*c*. From band structure calculations we find that the states originating from the Mn^{4c} sites dominate the transport. Anomalous Hall effect (AHE) resistance should therefore be proportional to the z-component of the magnetisation of the Mn^{4c} sublattice. The AHE measurements show loops with high remanence and only one magnetic phase, for which the coercivity diverges close to compensation. SQUID magnetometry measures the net moment, which is the sum of the antiferromagnetically coupled Mn^{4a} and Mn^{4c} sublattice moments. SQUID measurements in the same geometry as AHE exhibit a combination of an apparent soft magnetic response and in addition to a component with the coercivity observed by AHE. We infer from this that the net magnetic moment is canted due to non-collinearity of the sublattice moments.

The origin of the non-collinearity is thought to be competing symmetric exchange on the Mn^{4c} sites as bulk antisymmetric exchange averages to zero in the -43m point group. Our calculations show that there is an antiferromagnetic Mn^{4c}-Mn^{4c} first nearest neighbour exchange that is larger than the Mn^{4a}-Mn^{4c} antiferromagnetic exchange, while the second nearest neighbour Mn^{4c}-Mn^{4c} exchange is ferromagnetic, which allows for this type of magnetic order to form. The magnetisation of the Mn^{4c} sublattice is about an order of magnitude larger than the net magnetisation, a small canting of the Mn^{4c} moment, not observed in the square AHE loops, can lead to a large change in the direction of the net moment. The canting angle of the net moment increases as compensation is approached.

X-ray magnetic circular dichroism measurements were performed at -20° and +70° grazing incidence angles to map the sublattice magnetisation in the z-x plane. The two sublattices can be distinguished by L-edge absorption due to charge-transfer separation of the Mn absorption edges at the two sites by $\approx 1 \text{ eV}$. Analysis of the m_z-m_x moments shows that they are indeed non-collinear. From balancing of the relevant magnetic torques, the anisotropy fields at the Mn^{4a} and Mn^{4c} sublattices were found to be easy-plane -1.8 T and easy-axis 3 T, respectively. The sum of those fields is the anisotropy field of the net moment, which agrees with those obtained for the net moment by time resolved Faraday effect as well as inferred from magnetometry. We speculate that the non-collinearity can be controlled by composition as well as crystalline order. High crystalline order should favour non-collinearity, whereas defects will suppress the canting, due to real-space averaging.

MRG as a non-collinear half-metallic compensated ferrimagnet is an ideal candidate for spintronic materials and offers many advantages over their ferromagnetic counterparts as well as antiferromagnets. Immunity to external fields, absence of demagnetising fields and high anisotropy fields close to compensation implies high frequency spin dynamics and low Gilbert damping can be achieved. This is accompanied with easy manipulation and detection of the magnetic state, making MRG combine the most desirable properties in spintronics.

Magnetic and resistive effects in electrical switching experiments with antiferromagnets

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Antiferromagnets (AF) have recently attracted a lot of attention due to the possible control of spins arrangement by the current-induced spin orbit torques [1-6,8]. This opens the way to use them as active memory elements robust to external magnetic field and operating at THz frequencies. Different mechanisms of the electrical current control of antiferromagnetic order in AF insulators with adjacent heavy-metal have been proposed: a ponderomotive force or spin torque acting on a domain wall [2], out of plane oscillations of the Néel vector [3], spin torque acting on uncompensated spins [4] or thermo-magnetoelastic effects [5]. Unfortunately, the high current-densities used in numerous electrical experiments gives rise to non-magnetic effects that can mimic the actual behavior of the current-induced switching of an antiferromagnet [6]. Unless combined with a direct magnetic imaging technique [7], it is difficult to distinguish between the different contributions. Here, we present the observation of multiple effects acting in a counteractive way in a device made of NiO and Pt basing on an electrical experiment. The local heating and reversible electromigration are often evoked as a possible explanation for observed apparent switching patterns. However, they cannot fully explain our results obtained for longitudinal resistivity which decreases irreversibly after high current pulses. Thus, consideration of other effects such as recrystallization [8] is also addressed. The conclusions point out the significance of growth parameters for heterostructures dedicated for multi-terminal devices.



Figure 1: Alternating values of transverse resistance measured after pulses of constant high current density in two perpendicular directions (blue and red). The change of sign in the alternations can be seen, which suggest the presence of two competing mechanisms.

[1] P. Wadley et al., Science 351, 587 (2016).

- [2] L. Baldrati et al., Phys. Rev. Lett. 123, 177201 (2019).
- [3] X. Z. Chen et al., Phys. Rev. Lett. 120, 207204 (2018).
- [4] T. Moriyama et al., Sci. Rep. 8, 14167 (2018).
- [5] P. Zhang et al., Phys. Rev. Lett. 123, 247206 (2019).
- [6] C. C. Chiang et al., Phys. Rev. Lett. 123, 227203 (2019).
- [7] M. J. Grzybowski et al., Phys Rev. Lett. 118, 057701 (2017).
- [8] T. Matalla-Wagner et al., arXiv: 1910.08576 (2019).

First-principles theory of the Rashba-Edelstein effect in symmetry-broken magnetic systems

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Efficient manipulation of magnetic order with electric current pulses is desirable for achieving fast spintronic devices. The spin Hall and Rashba-Edelstein effects provide both distinctive means to achieve spin-orbit torques (SOT) and cause magnetization switching. We compute the Rashba-Edelstein effect (REE) from first-principles and show, for the noncentrosymmetric antiferromagnets CuMnAs and Mn₂Au, that the REE not only generates spin polarization but also orbital polarization, which is much larger than the spin counterpart. Investigating the frequency-dependent spin and orbital REE tensors we show that the electrically induced local magnetization can exhibit Rashba-like or Dresselhaus-like symmetries, depending on the magnetic configuration. The generated orbital polarization is however always of Rashba type. We furthermore compute sizable induced magnetizations at optical frequencies, which suggest that electric-field driven switching could be achieved at much higher frequencies, opening a route to petahertz spintronics.

We further compare the sizes of the spin Hall effect (SHE) and spin REE in pure heavy-metal Pt layers and in Pt/3d metal (Co, Ni, Cu) bilayers by quantitative calculations of their layer-resolved magnitudes. We show that the spin REE is not located strictly at the symmetry-broken interface, but is present in the Pt heavy-metal layer, too, and decays into the Pt layer with a decay length of about 9 atomic layers. In addition, we show that the SHE drops with a similar decay length from the interior of the Pt layer to zero at its surfaces. Using a spin diffusion model to compare the spin accumulation due to SHE and spin REE we find that the spin REE only dominates for thin bilayers whereas for thicker bilayers the SHE gives the main contribution to the spin accumulation and thus to the spin-orbit torque.

Current-induced magnetization switching of exchange-biased NiO heterostructures characterized by spin-orbit torque

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Spin-orbit torque (SOT)-induced magnetization switching provides a potentially efficient alternative to spin-transfer torque switching in spin valves or magnetic tunnel junctions. SOT-switching of perpendicular magnetization is observed in an external magnetic field collinear with the current (but non-collinear with the magnetization), which however, is impractical in device applications. Several solutions were proposed for magnetic field-free switching, including interfacial coupling between ferromagnetic and antiferromagnetic layers [1-3] or two coupled ferromagnetic layers exhibiting magnetization easy axes orthogonal to each other [4].

Here, we investigate the SOT generated by heavy metal (HM: W or Pt), which induces switching of Co layer exchange coupled with antiferromagnetic insulator NiO. W, Pt and Co were deposited by magnetron sputtering and NiO in separate chamber by pulse laser deposition (PLD). Investigated systems show the presence of both perpendicular and planar component of exchange bias (EB) at the Co/NiO interface determined by anomalous Hall and anisotropic magnetoresistance effects.

SOT-induced magnetization switching in µm-size Hall bars of W/Co/NiO and Pt/Co/NiO systems with variable thickness of HM characterized by perpendicular magnetic anisotropy of Co was investigated and analyzed in relation to our analytical switching model of critical current density as a function of Pt and W thickness, resulting in estimation of effective spin Hall angle and effective perpendicular magnetic anisotropy. The dependence of the critical current density vs. in-plane magnetic field show maximum at the magnetic field value corresponding to the EB planar component. We found that the field free switching occurs with current density of about 0.8×10^{12} A/m² and 1.5×10^{12} A/m² in HM/Co/NiO system with HM = Pt and HM = W, respectively. The current switching stability and training process are discussed in detail.

- [1] S.Fukami et al., Nature Mater 15 (2016)
- [2] Y.Oh et al., Nature Nanotech 11 (2016)
- [3] Y.Lau et al., Nature Nanotech 11 (2016)
- [4] S.Łazarski et al., Phys. Rev. Applied 12 (2019)

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Direct evidence for Néel order manipulation in Mn₂Au via current induced Néel spinorbit torque

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*The author has chosen not to make public additional content.

Nanoscale mechanics of antiferromagnetic domain walls in single-crystal Cr₂O₃

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Antiferromagnets hold promise as future spintronic devices, where information is encoded in the order parameter of the material [1]. In such applications, the control and understanding of antiferromagnetic spin textures, such as domain walls, is key to developing the technology. Though many studies of the Neel order parameter of antiferromagnets exist [2, 3], investigations of the underlying domain wall physics at the nanoscale remain scarce. This is due in part to a lack of nanoscale techniques as well as sufficiently pure materials, free of defects. Here, we present a study of domain wall structure and mechanics at the nanoscale in single-crystal Cr_2O_3 , a magnetoelectric antiferromagnet of great interest to spintronics due to its room temperature ordering and electrical switching capabilities [4, 5]. In particular, we make use of nitrogen-vacancy (NV) scanning magnetometry, which utilizes the electronic spin state of the atomic NV defect in a scanning diamond tip [6] to obtain quantitative, nanoscale images of the stray fields produced by isolated, nucleated domain walls. We observe the interaction of these domain walls with topographic islands on the crystal surface and are able to draw conclusions regarding the elastic properties of the domain wall. We furthermore present control over the domain wall by shifting it through the fabricated pinning landscape. In this way, we demonstrate the principles necessary to use such domain walls in spintronic applications such as memory devices.

[1] Jungwirth, T., et.al. Nat. Phys. 14, 200 (2018)

- [2] Song, C., et.al. Nanotech. 29, 112001 (2018)
- [3] Fiebig, M., et.al. Appl. Phys. Lett. 66, 2906 (1995)
- [4] Belashchenko, K. D. Phys. Rev. Lett. 105, 147204 (2010)
- [5] Kosub, T., et.al. Phys. Rev.Lett. 115, 097201 (2015)
- [6] Hedrich, N., et.al. arXiv:2003.01733 (2020)

Antiferromagnetic domain structure and magnetostructural kinetics in CuMnAs devices

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The demonstration of multiple means for efficiently controlling electron spins in antiferromagnetic (AF) materials has highlighted the highly attractive properties of AF materials for spintronics, including the lack of magnetic stray fields, robustness against magnetic field perturbations and intrinsic dynamics in the THz regime [1, 2, 3]. Despite major breakthroughs in AF spintronics research the factors dictating their basic domain structure are not well understood. Yet, from the point of view of applications, a clear understanding of the equilibrium domain structure is essential in order to tailor both the configuration of AF domains and the energy barriers separating different states. In fully compensated AFs with no demagnetizing fields, magnetoelastic effects are expected to dominate [4,5].

We report direct imaging and analysis of AF domains in patterned CuMnAs thin films grown epitaxially on a GaP substrate. A pronounced effect of patterned edges on the domain orientation and domain wall width is demonstrated, which extends over several micrometers. We show, how the edge effect leads to different domain configurations depending on the device geometry. Additionally, a local alignment of the Néel vector with microtwin line defects (see also [6]) is observed, which, combined with the edge effect, leads to the formation of biconvex domains oriented perpendicular to the patterned edges. We observe magneto-structural reconfigurations in which defect lines and surrounding AF domains grow together over the timescale of weeks at room temperature and over minutes 30K above room temperature. Our results demonstrate a means to tailor AF domain structures for specific applications and highlight the necessity of considering magneto-structural effects in AF systems.



Figure 1: AF domain structure and correlation to microtwin defects in a patterned 80µm x 10µm sized CuMnAs stripe. Top: AF domain structure imaged in XMLD-PEEM. The double-headed red arrows indicate the local spin axes. Bottom: Scanning X-ray diffraction map of the same bar showing the distribution of microtwin line-defects. The black arrows correspond to the CuMnAs [110] and [1-10] crystalline axes.

P. Wadley, et al., Science, 351 (2016) 587
 X.Z. Chen, et al., Phys Rev Lett 120 (2018) 207204
 P. Nemec, et al., Nature Physics. 14 (2018) 229

[4] O. Gomonay and V. Loktev, J Magn Magn Mater 242(2) (2002) 1418
[5] O. Gomonay, V. Baltz and et al., Nature Physics 14 (2018) 213
[6] F. Krizek, Z. Kaspar, A. Vetushka et al., Phys Rev Mater 4 (2020) 014409

Laser-induced THz magnetism of antiferromagnetic CoF2

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Antiferromagnets represent the largest family of magnetically ordered materials in nature, but despite several unique functionalities, their applications in modern technology are still scarce. In fact, fundamental studies of spin dynamics of antiferromagnets, the developments of means and approaches for manipulation of spins in antiferromagnets are presently among the hottest topics in magnetism [1]. It has been demonstrated that ultrashort laser pulses can be employed to drive THz dynamics of antiferromagnetically ordered spins via excitation of magnons at the edge of the Brillouin zone[2] or via launching non-linear lattice vibrations[3]. In this work, using sub-10 fs linearly polarized laser pulses, pump-probe technique and polarization-sensitive detection, we explore the off-resonant optical excitation of THz coherent lattice and spins dynamics in model antiferromagnetic insulators CoF_2 and MnF_2 . In particular, we demonstrate ultrafast excitation of coherent Raman-active A_{1g} and E_g odd optical phonon modes mediated by impulsive stimulated Raman scattering (ISRS). Analysis of polarization dependences of the coherent lattice dynamics reveals that, unexpectedly, the coherent even E_u phonon mode is also excited via ISRS, though being symmetry forbidden in centrosymmetric CoF_2 and MnF_2 . We ascribe this result to the electric-quadrupole contribution to the ISRS process which enables excitation and detection of the E_u phonons even in these centrosymmetric crystals [4]. Finally, we examine the optimal conditions for ultrafast excitation of the 2-Magnons mode at the edge of the Brillouin zone in the CoF_2 and MnF_2 .

- [1] P. Němec, M. Fiebig, T. Kampfrath, and A. V. Kimel, Nat. Phys., vol. 14, no. 3, pp. 229–241, 2018
- [2] D. Bossini et al., Nat. Commun., vol. 7, no. 1, pp. 1–8, Feb. 2016
- [3] A. S. Disa et al., Nat. Phys., pp. 1–5, Jun. 2020
- [4] J. L. Birman, Solid State Commun., vol. 13, no. 8, pp. 1189–1193, Oct. 1973

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Computationally-Efficient Simulation of The Spin Dynamics of Chiral Magnetic Textures in Compensated Ferrimagnets

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Compensated ferrimagnets are a promising class of materials in the field of spintronics due to their zero net moment and high spin-polarisation at the Fermi level. The first experimentally demonstrated member of this class is Mn₂Ru_xGa wherein two competing Mn sublattices compensate one another at a temperature which can be tuned via chemical doping/thin film growth conditions. This material showcases the expected huge anisotropy and coercive fields near the compensation temperature, while the high-spin polarisation present allows for the electrical control of the magnetisation. In addition, record values of the effective spin-orbit field have been measured in thin films of this material [1], ergo enabling efficient excitation via intrinsic spin-orbit torque.

Due to primarily the large anisotropy and spin-orbit fields of Mn₂Ru_xGa and similar materials, they are predicted to be useful in magnetic oscillators which operate in the THz gap. As of yet, no such devices have been demonstrated due to the sensitive, disorder-dependent, interplay between the crystal, magnetic and electrical structure of these materials. Ab-initio studies of these materials also encounter difficulties due to the complicated dependence of their properties on disorder.

Our work involves the development of a classical dynamic model for the simulation of such materials, capable of encapsulating the real-world physics in a relatively simple 1D spin-Hamiltonian. In the model, two sublattice chains are considered which are individually characterised by their spin-magnitude, anisotropy type/strength, Heisenberg exchange, Dzyaloshinskii-Moriya interaction and Gilbert damping. The other intrinsic effect is that of intersublattice exchange between the two distinct chains. Due to the low/zero net moment, it is justified to neglect the computationally expensive demagnetisation field calculations, which scale as n². Additional Hamiltonian terms for extrinsic effects include a Zeeman term for external applied field and a spin-transfer/spin-orbit torque term to allow for current driven excitation of spin dynamics. The time evolution of the spins is then computed by numerical integration using a framework based on the LLG equation of motion. Both linear multistep algorithms and predictor-corrector algorithms are used for the integration, where appropriate.

Two main goals are pursued in this work; equilibration of topological spin textures to find the ground state as a function of the intrinsic material parameters and investigation of the dynamic motion of the textures as a function of the same intrinsic parameters, as well as, driving external magnetic fields and current densities. We use analytical approximations for the ideal magnetisation profile and allow these to relax in the steady-state to find topological energy minima in the spin-lattice configuration landscape for different values of the intrinsic material parameters, as shown in Fig.1(B). These equilibrium states are then excited into motion via an applied external field or current and the motion of each individual spin is tracked in 3D space, as shown in Fig.1(A). From this data, values for the domain-wall size and velocity can be extracted for example, and in the case of the 1D skyrmionic configurations shown, the breathing mode can be observed and quantified as a function of external field.

[1] Lenne, Simon, et al. arXiv preprint arXiv:1903.04432 (2019)

Symposium 13. Magnetic materials and technologies for energy applications
Advanced characterization of multicaloric materials in pulsed magnetic fields

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The multicaloric effect is described by a temperature or entropy change of a material triggered by external stimuli applied or removed simultaneously or sequentially. The prerequisite for this is a material exhibiting multiple ferroic states. However, direct measurements of the effect are rarely reported. Now, for this reason, we built a measurement device allowing to determine the adiabatic temperature change in pulsed magnetic fields and, simultaneously, under the influence of a uniaxial load [1]. We selected the all-d-metal Heusler alloy Ni-Mn-Ti-Co for our first test because of its enhanced mechanical properties and enormous magneto- and elastocaloric effects. Ni-Mn-Ti-Co was exposed to pulsed magnetic fields up to 10 T and uniaxial stresses up to 80 MPa, and the corresponding adiabatic temperature changes were measured. With our new experimental tool, we are able to better understand multicaloric materials, determine their cross-coupling responses to different stimuli and thus further advance the development of the solid-state cooling process by exploiting the thermal hysteresis [2].

[1] T. Gottschall, E. Bykov, A. Gràcia-Condal, B. Beckmann, A. Taubel, L. Pfeuffer, O. Gutfleisch, Ll. Mañosa, A. Planes, Y. Skourski, and J. Wosnitza, J. Appl. Phys. 127, 185107 (2020)

[2] T. Gottschall, A. Gràcia-Condal, M. Fries, A. Taubel, L. Pfeuffer, Ll. Mañosa, A. Planes, K. P. Skokov, and O. Gutfleisch, Nat. Mater. 17, 929 (2018)

Opportunities for the future production of Nd-Fe-B magnets: Environmental implications and supply constraints

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*The author has chosen not to make public additional content.

Direct and inverse magnetocaloric effects in antiferromagnetic Tb₃Ni

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The magnetocaloric properties of Tb₃Ni have been studied by means of the evaluation of the magnetic entropy change, measuring the isothermal magnetization of a single crystalline sample in a broad temperature range. It presents two important magnetocaloric effects in the temperature range 3-90 K due to the richness and variety of its temperature and magnetic field induced phase transitions. There is a direct magnetocaloric effect with a maximum at 65 K and a value of the maximum of the magnetic entropy change $|DS_M^{pk}| = 16.6 \text{ J/kgK}$, refrigerant capacity RC_{FWHM} = 432 J/kg, with a 50 K span, for $\mu_0 DH = 5$ T, which is due to the transition from a magnetically ordered state to the paramagnetic state with a combined antiferromagnetic to ferromagnetic metamagnetic transition. Besides, it also presents an inverse magnetocaloric effect at very low temperature for which the presence of metamagnetic transitions between AFM and FM states is responsible $|DS_M^{pk}| = 19.9 \text{ J/kgK}$, RC_{FWHM} = 245 J/kg, with a 15 K span, for $\mu_0 DH = 5$ T. At low field (< 2 T), the character of the AFM-PM transition at 61 K has been well established to be second order and influenced by short range order interactions, while the metamagnetic transitions between AFM appear for magnetic fields higher than 2 T, have a first order character. This implies that the use of the Maxwell equation to obtain $|-DS_M^{pk}|$ must be combined with the Clausius-Clapeyron one to obtain reliable results and not spurious peaks.

The properties shown by this material makes it a suitable candidate for magnetic refrigeration applications in the gas liquefaction range 4-77 K.



Figure 1 : Magnetic entropy change $-DS_M$ for $\mu_0 DH$ from 0.5 T to 5.9 T. Insert: $-DS_M$ as a function of $\mu_0 DH$ at 52 K

Oral Presentation

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Magnetocaloric properties and magnetic interactions in Ho6(Fe,Mn)Bi2 intermetallics

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Ho₆MnBi₂, Ho₆FeBi₂, Ho₆(Mn_{0.5}Fe_{0.5})Bi₂, and Ho₆(Mn_{0.75}Fe_{0.25})Bi₂ (which crystalize in the Fe₂Ptype structure) have been studied using magnetic techniques in order to explore the suitability of the intermetallic family Ho₆(Fe,Mn)Bi₂ as a magnetocaloric material. Besides, the critical behavior of the paramagnetic (PM) to ferromagnetic (FM) transitions has also been investigated to obtain a deeper understanding of the range of the magnetic interactions. The obtained critical exponents β , γ and δ point to long-range order interactions, as they are close to those of the Mean Field Universality class. The magnetocaloric effect in the four compounds shares common properties: it spans over a very broad temperature range, limited by the PM-FM transition at high temperature and a spin-reorientation one at low (see Fig. 1), with more than 200 K for Ho₆MnBi₂ at 5T and, on the other limit, 80 K for Ho₆FeBi₂. They present very high values of the refrigerant capacities (from 520 J/kg to 709 J/kg at 5T) and good magnetic entropy changes (from 3.4 to 5.7 J/(kgK) at 5T), as well as a flat and wide temperature span for the working temperature range. The change in properties with composition proves that the magnetocaloric properties can be tuned in this intermetallic family to accommodate different refrigeration applications. The magnetocaloric scaling laws have been successfully tested and universal curves for the magnetic entropy change have also been obtained in the PM-FM transition region.



Figure 1 : Magnetic entropy change $-\Delta S_M$ for $\mu_0 DH$ from 1 T to 6.9 T for **a**) Ho₆MnBi₂, **b**) Ho₆FeBi₂.

Exchange-coupled spinel ferrite-based core-shell nanoparticles for magnetic heat dissipation

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Understanding and governing the complex behavior in magnetic materials at the nanoscale is the key and the challenge not only for fundamental research but also to exploit them in applications ranging from catalysis,[1] to data storage,[2] sorption,[3] biomedicine,[4] and environmental remediation.[5] In this context, spinel ferrites represent ideal magnetic materials for tuning the magnetic properties through chemical manipulations, due to their strong dependence on the cation distribution, spin-canting, interface, size, shape, and interactions. Furthermore, when coupled with other phases (heterostructures), they can display rich and novel physical properties different from the original counterparts (exchange coupling, exchange bias, giant magneto-resistance), allowing them to multiply their potential use.[6] For example, the possibility to tune magnetic anisotropy and saturation magnetization by coupling magnetically hard and soft materials have found usage recently in applications based on magnetic core-shell nanoparticles with homogeneous coating and low size dispersity for uniform magnetic response and to maximize the coupling between the hard and soft phases.[8] Even though some studies have reported interesting results in the field of magnetic heat induction, a systematic study on an appropriate number of samples for a better comprehension of the phenomena to optimize the performance is needed.

In this contribution, the capability of coupled hard-soft bi-ferrimagnetic nanoparticles to improve the heating ability is exploited to understand the influence of the different features on the performances. This systematic study is then based on the correlation between the heating abilities of three magnetically hard cobalt ferrite cores, covered with magnetically soft spinel iron oxide and manganese ferrite having different thickness, with their composition, structure, morphology and magnetic properties. Direct proof of the core-shell structure formation was provided by nanoscale chemical mapping, with identical results obtained through STEM-EELS/EDX, and tomography. ⁵⁷ Fe Mössbauer spectroscopy and DC/AC magnetometry proved the magnetic coupling between the hard and the soft phases, thanks also to the comparison among core-shell NPs, ad-hoc prepared mixed cobalt-manganese ferrites NPs, and cobalt ferrite NPs mechanically mixed with manganese ferrite NPs. The heating abilities of the aqueous colloidal dispersions of the three sets of core-shell samples revealed that, in all cases, core-shell nanoparticles showed better performances in comparison with the respective cores, with particular emphasis on the spinel iron oxide coated systems and the samples featuring thicker shells. This scenario entirely agrees with the hypothesis made based on magnetic parameters (saturation magnetization, Néel relaxation times, effective anisotropy) of the powdered samples, and demonstrated the importance of a sophisticated approach based on the synergy of chemical, structural, and magnetic probes down to a single-particle level.

- [1] Chem. Rev., 2011, 111, 3036-3075
- [2] Nano Lett., 2014, 14, 3395–3399
- [3] J. Mater. Chem. A, 2017, 5, 21688-21698
- [4] Chem. Rev., 2015. 115, 327-394
- [5] Environ. Sci. Nano, 2016, 3, 1241–1253
- [6] Chem. Soc. Rev. 2015, 44, 7540–7590
- [7] Nat. Nanotechnol.,2011, 6, 418-422
- [8] J. Nanosci. Nanotechnol., 2019, 19, 4954–4963

Spherical HoAl₂ magnetocaloric particles for hydrogen magnetic refrigeration fabricated by a gas-atomization process

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Magnetic refrigeration is a cooling technology based on the magnetocaloric effect (MCE) in which the changing magnetic field causes the change in magnetic entropy or temperature of magnetic materials. This technique has the advantages over the conventional vapor-compression refrigeration in that it is environmentally-friendly, does not require a compressor, and is more efficient in principle [1, 2]. Among the potential applications, hydrogen liquefaction has attracted attention [3]. Hydrogen is expected as one of the cleanest energy sources to replace fossil fuels. For practical use, it is efficient and economical to utilize the hydrogen as a liquid state, as is the LNG. In this context, there has been interested in developing unprecedentedly high-efficient cooling systems using magnetic refrigeration.

To realize the hydrogen liquefaction, the hydrogen gas must be precooled from high temperatures to the liquefaction temperature of 20.3 K. For this purpose, the active magnetic refrigerator (AMR) cycle of refrigeration is effective because it allows us to advance magnetic refrigeration over a wide temperature range [4]. An AMR cycle refrigeration system contains magnetic materials with large MCE as magnetic refrigerants, and it operates in a cycle that combines changing the magnetic field with flowing the heat exchange fluid through the magnetic refrigerants. One of the key issues is to gain better heat exchange efficiency between the magnetic refrigerants and the fluid. To improve AMR performance, the spherical magnetic refrigerant is desirable in terms of large surface area.

In this study, we present a way for fabricating spherical magnetocaloric particles of HoAl₂ by a gas-atomization process. HoAl₂ is a Laves phase intermetallic compound with a Curie temperature of 30 K, and it is one of the candidate materials for the hydrogen magnetic refrigeration. The fabricated particles have the size distribution ranging from 100 to 700 μ m in diameter, with a center value of about 212-350 μ m. Compared to the bulk HoAl₂ made by ourselves, the Curie temperature slightly increases while a spin reorientation transition at 20 K becomes broader in the atomized samples. By measuring magnetization and specific heat, we evaluated the magnetic entropy change and the adiabatic temperature change in the atomized samples. These values and temperature dependence are almost the same as those in the bulk sample, indicating that the atomized samples have the potential for use in magnetic refrigeration.

- [1] C. Zimm et al., Adv. Cryog. Eng. 43, 1759 (1998)
- [2] E. Brück, J. Phys. D: Appl. Phys. 38, R381 (2005)
- [3] T. Numazawa et al., Cryogenics **62**, 185 (2014)
- [4] K. A. Gschneidner, Jr. and V. K. Pecharsky, Int. J. Ref. 31, 945 (2008)

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Kinetics of the thermomagnetic transition in La(Fe,Si)₁₃ alloys using the Bean-Rodbell model

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In this work, we propose a combination of the Kolmogorov-Johnson-Mehl-Avrami (KJMA) nucleation and growth theory and the Bean-Rodbell (BR) model to accurately describe the field-induced thermomagnetic transition in La(Fe,Si)₁₃ alloys. The proposed combination has been applied to a highly pure LaFe_{11.6}Si_{1.4} alloy undergoing a first-order thermomagnetic transition presenting associated hysteresis. The kinetic analysis of both magnetization and demagnetization processes reveals nucleation and three dimensional interface controlled growth, corresponding to an Avrami exponent of 4. By introducing the kinetic process between the metastable and stable solutions of the BR model, the field dependence of the magnetization and demagnetization processes, including the magnetic hysteresis, is much better reproduced in comparison to the predictions of the pure model. With this approach, we solved that the BR predicted hysteresis values are larger than the experimentally observed ones and the extremely abrupt drop of the magnetization at the transition predicted by BR, which is also in disagreement with experimental data. Moreover, our approach demonstrates a strong interrelation between the Avrami exponent and the magnetic field that causes the onset of the transformation, allowing to use this onset upon magnetization and demagnetization as an internal validation of the fitting parameters. The combined analysis shows that the main source of hysteresis in the studied sample is the demagnetization process, for which a much slower rate of the transformation process) occurs up to around 40 % of completion.

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Large spin-dependent thermoelectric effects in NiFe-based interconnected nanowire networks

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The coupling between the transport of heat and spin is at the heart of the burgeoning field of spin caloritronics [1]. 3D interconnected multilayered nanowire (NW) networks are promising architectures to determine key spin caloritronic parameters. These overcome the lack of reliability and the limits regarding power generation due to the nanoscale of the structure used so far [2]. Besides, 3D interconnected NW networks allow the observation of giant magnetoresistance (MR) and magneto-thermopower (MTP) effects thanks to the current-perpendicular-to-plane (CPP) geometry and are of particular interest to develop magnetically controllable flexible thermoelectric devices [3,4].

Homogeneous and NWs made of NiFe alloys and multilayered NWs made of NiFe/Cu were fabricated by electrodeposition inside 3D nanoporous templates. NiFe alloys exhibit large thermopowers, especially when the Fe content is increased up to 40% in the alloy [5]. The multilayered structures preserve large thermopower values while being magnetically controlled with relative changes of Seebeck coefficient up to 25% at room temperature, reaching about 60% at 100K. A large spin-dependent Seebeck coefficient S_{\uparrow} - S_{\downarrow} of -12 μ V/K at room temperature has been determined for Ni₈₀Fe₂₀/Cu multilayered NWs. The influence of the alloy composition exhibits a rise of the ratio -MTP/MR when the Nickel content in the alloy increases. This arises from the increasing difference between S_{\uparrow} and S_{\downarrow} when the composition of the alloy tends to pure Nickel [6]. Such large difference of thermopowers paves the way to giant MTP ratio comparable to the giant MR observed in multi-layered structures.

- [1] G. E. W. Bauer, E. Saitoh and B. J. van Wees, Nature Materials 11, 391 EP (2012)
- [2] N. Liebing et al., Phys. Rev. Lett. 107, 177201 (2011)
- [3] T. da Câmara Santa Clara Gomes, F. Abreu Araujo and L. Piraux, Science Advances 5(3), eaav2782(2019)
- [4] F. Abreu Araujo, T. da Câmara Santa Clara Gomes and L. Piraux, Advanced Electronic Materials 5, 1800819 (2019)
- [5] C.Y. Ho, R.H. Bogaard, T.C. Chi, T.N. Havill, H.M. James, Thermochimica Acta 218, (1993)
- [6] M.C.Cadeville, J. Roussel, Journal of Physics F: Metal Physics 1(5), 686 (1971)

Deconvolution of overlapping phase transitions in composites using the universal scaling of the magnetocaloric effect

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It has been shown that the magnetic entropy change, ΔS_M , of second-order phase transition (SOPT) materials collapses onto a single curve when the curves for the different magnetic fields are normalized with respect to their peak values and the temperature axis is rescaled [1]. This universal scaling of the MCE is used to extrapolate the results to experimental conditions that could not be reached in the measurements, to discern between second-order and first-order phase transitions, etc. However, the coexistence of various phases in a material prevents a good collapse of the MCE in SOPT composites. These distortions from universal scaling in multiphase materials are more noticeable when the fractions of the different phases are comparable and their transition temperatures are close to one another [2].

The aim of this work is to present a procedure which allows the deconvolution of the magnetocaloric response of SOPT biphasic composites with arbitrary concentration of phases. In this way, we can calculate the fraction of phases via the reconstruction of the total DS_M of the composite from the addition of the two deconvoluted responses. The procedure has been applied to a Gd-Pd composite with coexisting Gd and Gd₇Pd₃ phases with Curie temperatures separated by ~45 K [3], which is one of the challenging cases.

The procedure consists of obtaining the universal curves of the two phases (*i*=1, 2) from the global response of the composite by scaling the temperature axis with one reference temperature, $T_{r,i}$, as $\theta_i = (T-T_{pk,i})/(T_{r,i}-T_{pk,i})$ and normalizing the DS_M with respect to the corresponding peak value $\Delta S_M^{pk,i}$. Under the assumption of no interactions between phases, the response of the composite has been reconstructed from the addition of the deconvoluted responses after undoing the scaling. For low fields, there is an excellent agreement between the reconstructed response and the experimental curves. However, the power law behavior typical for SOPTs ($\Delta S_M^{pk, \infty} \ll H^n$ and $T_r \propto H^r$) is lost for moderate and large fields due to the overlap of the transitions. Therefore, the reconstruction of the experimental curves from the addition of the deconvoluted contributions requires an iterative procedure. Taking this into account, a very good agreement between the reconstructed and experimental responses of the composite in the entire field range is obtained.

[1] V. Franco et al., Int. J. Refrig. 33 (2010) 465-473

- [2] V. Franco et al., J. Magn. Magn. Mater. 321 (2009) 1115–1120
- [3] P. Gębara et al., J. Magn. Magn. Mater. 500 (2020) 166175

Revisiting T-FORC analysis of Heusler type alloys using a unified driving force approach

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Heusler type alloys are multifunctional materials that are used in a broad variety of applications. In particular, the Ni-Mn-In-Co type alloy has promising magnetocaloric properties [1]. It undergoes a magneto-structural transition from a low magnetization phase to a ferromagnetic phase that can be driven both by temperature and by magnetic field. The low magnetic moment of the low temperature phase facilitates the studies of the phase transition, making Ni_{45.7}Mn_{36.6}In_{13.5}Co_{4.2} the ideal sample for our work.

One of the limitations of Heusler alloys to be applicable as magnetic refrigerant materials is their thermal hysteresis [2]. Therefore, new techniques, like T-FORC (temperature first order reversal curves) are being applied to thoroughly characterize the hysteretic nature of the transition and how this thermal hysteresis is affected by magnetic field [3]. T-FORC is able to fingerprint the details of the transition. However, the fact that the temperatures at which the transition takes place in Ni-Mn-In-Co shift with applied field motivates the choice of a common origin temperature for all the hysteresis loops for making the analysis (usually the chosen origin is the center of the saturation thermal hysteresis loop for each applied magnetic field). This allows for a qualitative comparison of the features of the distribution but prevents actual quantitative analysis of the influence of different applied fields or even comparison between different samples.

Taking into account that the magnetostructural phase transition in Ni-Mn-In-Co can be driven both by temperature and by magnetic field, and following a phenomenological approach similar to that used for describing temperature dependent hysteresis loops in interacting nanoparticles [4], we have proposed an effective temperature T* which accounts for the energy (either thermal or magnetic) supplied to the sample in order to drive the transition [5]. This unified driving force can be used as the variable to study the hysteresis of the transition by defining a T*-FORC, i.e. plotting the magnetization curves (measured either as a function of T or H) vs. T* and performing FORC analysis on those data. In this way, the influence of the magnetic field on the distributions can be quantified because this procedure avoids imposing a reference temperature origin at the center of each loop.

In addition to a more quantitative comparison of the features of the distributions, the use of the unified driving force would allow to perform T*-FORC experiments in a shorter experimental time than the one required for T-FORC.

[1] Gottschall, T; Skokov, K.P.; Frincu, B.; Gutfleisch, O., Appl. Phys. Lett. 106, 021901 (2015)

- [2] Franco, V.; Blázquez, J.S.; Ipus, J.J.; Law, J.Y.; Moreno-Ramírez, L.M.; Conde, A., Prog. Mater. Sci. 93, 112 (2018)
- [3] Franco, V.; Gottschall, T.; Skokov, K.P.; Gutfleisch, O., IEEE Magn. Letters 7, 6602904 (2016)
- [4] Franco, V.; Conde, C.F.; Conde, A.; Kiss, L.F., Phys. Rev. B 72, 174424 (2005)
- [5] Blazquez, J.S.; Franco, V.; Conde, A.; Gottschall, T.; Skokov, K.P.; Gutfleisch, O., Appl. Phys. Lett. 109, 122410 (2016)

First-Principles investigation of the impact of chemical order on the Magnetocrystalline anisotropy in L1₀ FeNi (tetrataenite)

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The volatility in price and uncertainty in supply of the rare earth elements makes it highly desirable to find alternatives to rare-earth based magnets, in order to meet the increasing global demand for permanent magnets. One interesting candidate in this quest is the chemically-ordered $L1_0$ phase of Fe₅₀Ni₅₀ (tetrataenite) found in iron meteorites. While the laboratory synthesis of the ordered phase is very difficult due to the slow diffusion of atoms at the rather low order-disorder transition temperature, several attempts have been made to achieve samples with a high degree of chemical order. Nevertheless, synthesis of a fully ordered system remains challenging.

Using first-principles-based density-functional theory calculations in combination with Monte Carlo (MC) simulations, we investigate the interplay between the degree of chemical order and the magnetic properties of L1₀ FeNi. Our calculations demonstrate a strong effect of the magnetic order on the chemical order-disorder transition temperature, and a strong effect of the chemical order on the magnetic transition temperature, in agreement with previous studies. Furthermore, we investigate the dependence of the magneto-crystalline anisotropy on the chemical long range order. Our results indicate that the anisotropy does not decrease significantly as long as the deviations from perfect order are not too large. Moreover, we also find that a slight disorder in the alloy can in principle result in an even higher anisotropy than for the fully ordered structure.

We further analyze the correlation between the magneto-crystalline anisotropy and the orbital magnetic moment anisotropy. This allows us to study the effect of the local chemical environment on both quantities, potentially enabling further optimization of the magneto-crystalline anisotropy with respect to both long-range order and stoichiometric composition.

Computational design of rare-earth lean hard magnetic phases

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The increasing use of 'green technologies' is interwoven with the increasing demand for high performance permanent magnets. Today most high-performance magnets are based on Nd₂Fe₁₄B which contains about 12% Nd. Thus, new phases with less critical raw materials and a smaller environmental footprint are highly sought after. Computational design based on ab initio methods has been proven to be an efficient and accurate tool to identify new phases [1, 2].

Here, Fe-rich REFe_{12-x}Z_x phases based on the tetragonal ThMn₁₂ structure have been chosen as a starting point for a systematic search. These phases have recently been rediscovered due to the rare earth crisis and the increasing demand of magnets. They are a quite natural choice since they contain significantly less rare earth than commercially used magnets and can be synthesized for various phase stabilizing elements Z and different elements on the RE site.

We have performed a comprehensive ab initio study of $REFe_{12-x}Z_x$ with RE = Nd, Y, Ce, Sm; Z = Ti, V aiming to tune the magnetic performance towards large uniaxial magnetocrystalline anisotropy (MCA) and high Curie temperatures by using a combination of different state of the art first principles methods combined with atomistic simulation methods for temperature dependent properties. To optimize the geometry and test the phase stability the VASP [3] package has been used. The optimized structures were then transferred to the full-potential LMTO code RSPt [4] for magnetic characterization. Atomistic simulations were carried out using UppASD [5] and VAMPIRE [6].

In practice in many cases (except for Sm) light interstitials such as N or H have to be added to e.g. improve the coercive field. Since especially N can alter the orientation of the MCA it has been included in the study. Several promising phases were identified. Here, we focus the discussion on $YFe_{11.5}Ti_{0.5}N_{0.5}$, $Nd_{0.5}Y_{0.5}Fe_{11}TiM_{z}$ (M = N, H), and SmFe₁₁V. The last one has been already synthesized showing a good magnetic performance, i.e. in agreement with the theoretical prediction the MCA as well as the magnetization are increased about 20% compared to the previously known SmFe₁₀V₂ phase [7]. A similar good magnetic performance is achieved for the rare earth free phase $YFe_{11.5}Ti_{0.5}N_{0.5}$. The Curie temperature predicted from atomistic simulations is in the same range as the one of SmFe₁₁V (650 K). Without N the Y-based system has a small planar MCA which turns into a uniaxial one if N is added. It possesses a uniaxial MCA of about 1MJ/m³ which is expected to further increase with higher nitrogenation levels.

- [1] S. Arapan, P. Nieves, H. C. Herper, and D. Legut, Phys. Rev. B 101, 014426 (2020)
- [2] P. Nieves et al., Comput. Mater. Sci. 168, 188 (2019)

- [4] J. M. Wills et al., vol. 167 Springer series in solid state science (Springer, 2010)
- [5] B. Skubic et al. J. Phys.: Condens. Matter 20, 315203 (2008)
- [6] R. F. L. Evan et al., J. Phys.:Condens. Matter 26, 103202 (2014)
- [7] A. M. Schönhöbel et al., JALCOM 786, 969 (2019)

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^[3] G. Kresse and J. Hafner, Phys. Rev. B 48, 13115 (1993); Phys.Rev. B 54, 11169 (1996); Computational Materials Science 6, 15(1996)

High-throughput and data-mining search for rare-earth free permanent magnets

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High performance permanent magnets are needed for a large number of applications, one of which is the 'green' energy conversion, such as electric motors, wind mills, and many more. However most high-performance magnets contain rare-earth (RE) materials which makes them expensive, while some of the RE elements are rapidly decreasing in availability.

We are applying a high-throughput and data-mining approach to the search of rare-earth free permanent magnets. Going through a large number of known structures from ICSD database [1], and using a full-potential linear muffintin orbital method with relativistic formulation as implemented in the RSPt electron structure code [2] to calculate magnetic anisotropy and Curie temperature, we were looking for the materials with high magnetization > 1 T, uniaxial anisotropy with high magnetic anisotropy energy (MAE) > 1 MJ/m³, and T_c > 300 K to identify the suitable replacement for rare-earth containing materials.

To test the method, we started with the materials containing a 3d-, a 5d-, and one extra element of the periodic table to test the method, which makes it more likely for a material to have the large magnetization and MAE. We found several materials that have been known as the strong permanent magnets before as well as some new candidates that have characteristics suitable for a good permanent magnet, such as Pt₂FeCu, Pt₂FeNi, W₂FeB₂, etc. Based on this result we also proposed two new materials, not existing in ISCD database: Pt₂FeCo and Pt₂CoNi [3]. With this 'test' run proving successful we are continuing applying the high-throughput approach to materials with the different initial choice of elements.

[1] http://www2.fiz-karlsruhe.de/icsd home.html

[2] John M. Wills et al., arXiv:cond-mat/9912173 (1999)

[3] Alena Vishina, Olga Yu. Vekilova, Torbjörn Björkman, Anders Bergman, Heike C. Herper, and Olle Eriksson, Phys. Rev. B 101, 094407 (2020)

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Predicting the performance of magnetocaloric systems using machine learning

regressors

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The European Commission has recently proposed a European Green Deal in the important quest for global warming mitigation. In that respect, the use of a significant share of consumed energy is dedicated to refrigeration and heating systems. Hence, innovating the current state of refrigeration and heating systems to improve their efficiencies is a critical challenge to be tackled. Magnetocaloric (MC) systems are one of the most promising innovations, as their theoretical coefficient of performance (COP) is significantly larger than that of vapor-compression systems. MC systems rely on the reversible magnetocaloric effect where applying (or removing) magnetic fields (H) changes the thermodynamic state of the material in such a way that, in adiabatic processes, the temperature increases (or decreases) [1].

Although the development of magnetocaloric systems is now in a relatively mature phase, the geometrical and operating optimization is still a time-consuming task. One way of optimizing a system is by predicting what are the performance values if using a set of geometrical and operating parameters. In that respect, the recent interest of artificial intelligence lead to the development of several statistical learning techniques and respective algorithm implementations [2]. These techniques have recently been used in a wide number of research fields. One specific class of statistical techniques concerns supervised learning, where regressors (or classifiers) are trained based on datasets with continuous (or Boolean) output values [2]. So far, only Aprea et al. have implemented an artificial neural network, i.e. a multilayer perceptron (MLP), to optimize performance values of a particular prototype [3].

In this work, different statistical learning regressors are applied, and critically compared, in predicting the three main performance values for a generic magnetocaloric heat pump: no load temperature span, heating power and COP. Four different regressors were used: ordinary least squares, ridge, lasso, and K-nearest neighbors (KNN). The used dataset was obtained with the recently developed heatrapy framework to compute caloric systems [4]. The minimum average relative error of the temperature span, heating power and COP were 23%, 29% and 31%, respectively. The Lasso regressor shows the best performance with the order of the polynomials. The best order of polynomials range between 3, for the heating power, to 5, for the COP. The worse performance in predicting the three performance values occurs when using the KNN regressor. Furthermore, the application of regressors to the dataset is more adequate to evaluate the temperature span rather than energetic performance values.

[1] A. Kitanovski et al., Magnetocaloric Energy Conversion. Springer (2015)

[2] C. M. Bishop, Pattern recognition and machine learning. Springer (2006)

[3] C. Aprea et al., Int. J. Refrig. 82 (2017) 238

[4] D. J. Silva et al., SoftwareX 7 (2018) 373

Efficient and affordable thermomagnetic materials for harvesting low-temperature waste heat

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Industrial processes release substantial quantities of waste heat, which can be harvested to generate electricity. Nowadays recovering waste heat becomes decisive to use the limited primary energy most effective. At present, the low-temperature waste-to-heat conversion relies solely on thermoelectric materials, but such materials are expensive and have low thermodynamic efficiencies. A promising alternative way for low temperature energy harvesting is the usage of thermomagnetic systems. Within these, heat is first converted into magnetic energy and then into electricity. Most recently a thermomagnetic generator was presented with a new design of the magnetic circuit, which increased the power output and efficiency significantly [1]. Although thermomagnetic materials may offer a promising alternative, their performance remains to be evaluated, thereby hindering their real-world application.

In this talk we evaluate the efficiency and cost effectiveness of thermomagnetic materials that can be used in motors, oscillators and generators for converting waste heat to electricity. We describe the influence of magnetization change and heat capacity on thermodynamic efficiency. In figure 1 the change of magnetization at a temperature difference of 10 K is plotted over the required heat input for different thermomagnetic materials. The ratio of magnetization change and heat input is proportional to the efficiency of the energy conversion from thermal to magnetic energy. This material efficiency is given as grey dashed guidelines in the Ashby type plot, which allows to select materials with highest efficiency. Furthermore, we take a look at the consequences of thermal conductivity on the power density in the materials. The ratio of power density and the raw materials costs give a value for the cost effectiveness, which we can compare with thermoelectric, today's power plants or solar cells. Our analysis reveals that at low temperature differences (10 K) the efficiency of the best thermomagnetic materials is high (45 % of Carnot efficiency) and outperforms the efficiency of thermoelectric materials. Importantly, we find that that the price per watt for some thermomagnetic materials are much lower compared to present-day power plants and thermoelectric generators. In this talk we present a materials library which allows selecting the best available thermomagnetic materials in Ashby plots and gives guidelines for future development [2].



Figure 1 : Evaluating the thermodynamic efficiency η of thermomagnetic materials. To reach high η , a large change of magnetization ΔM is beneficial, as well as a low heat input Q_{in} . The grey dashed lines represent a constant efficiency. Accordingly, the most efficient materials are located in the top left corner, where η approaches 2 %.

[1] Waske et al., Nature Energy 4, 68-74 (2019)[2] D. Dzekan et al., arXiv:2001.03375 (2020)

Systematics in Curie temperature of rare earth permanent magnets materials

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Curie temperatures T_c 's of permanent-magnet materials R_2 (Fe, Co)₁₄B, R_2 (Fe, Co)₁₇, and $RFe_{11.5}Ti_{0.5}$ (R = La, Ce, ..., Lu, Y), are calculated within the meanfield approximation using the exchange coupling constants J_{ij} 's that are obtained by first-principles KKR-Green's function method. While the agreement between the calculations and experiments is rather well for the Co-based systems, there arise some discrepancies in the case of Fe-bases systems. Despite these discrepancies, however, the systematic changes seen in T_c as the rare earth element R changes across the lantanoid are fairly well reproduced by the calculation. In the case of $R_2Fe_{14}B$, the discrepancies can be largely diminished by using J_{ij} that are calculated for local-moment disordered states, which is considered to correspond to the paramagnetic state above T_c . However, this is not the case in general: the discrepancies seen in R_2Fe_{17} cannot be remedied. From the analyses of calculated and experimental data, it is concluded that data assimilations are possible and necessary to make reasonable predictions of T_c for rare earth permanent-magnet materials, in particular, the Fe-based ones.

Oral Presentation

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Intrinsic hard magnetic properties of Sm(Fe,Co,Ti)₁₂ compound with ThMn₁₂ structure

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Iron rich rare-earth intermetallic compounds RFe₁₂ (R is a rare-earth element) with the ThMn₁₂ structure have been considered as promising candidates for high performance permanent magnet material since the 1980's. These compounds have high Curie temperature (T_c), saturation magnetization (μ_0M_s) and magnetocrystalline anisotropy (μ_0H_A) which are comparable with Nd₂Fe₁₄B [1]. Lately, they attract special attention not only due to their superior intrinsic magnetic properties at high temperature but also due to their low rare earth content in terms of being potential for delivering resource-saving and cost-effective technology. Recently, it was founded that Sm(Fe_{0.8}Co_{0.2})₁₂ thin film has excellent intrinsic magnetic properties superior to Nd₂Fe₁₄B ($\mu_0M_s \approx 1.78T$, $\mu_0H_A \approx 12T$, $T_c \approx 860$ K) [2]. But RFe₁₂ compounds are thermodynamically unstable in the bulk state thus there is a need for stabilizing elements, such as Ti. Although, this leads to a reduction in saturation magnetization. For that reason, it is valuable to know the effect of Ti on phase stability and the μ_0M_s . The sole effect of Ti on intrinsic magnetic properties cannot be stabilized for x < 0.9 in RFe_{12-x}Ti_x [3]. Hence, in this study, highly textured single phase (001) oriented epitaxial thin films Sm(Fe,Co)_{12-x}Ti_x are produced by DC magnetron cosputtering in order to investigate systematically the effect of Ti-substitution. The addiction of Ti increases the lattice parameter a, but keeps the c constant which leads to anisotropic volume expansion. The effect of Ti on intrinsic magnetic of Ti on intrinsic magnetic properties and phase stability will be discussed in this talk.

- [1] P. Tozman et al., Acta Mater., 153 (2018), 354-363
- [2] Y.Hirayama et al., Scr.Mater. 138 (2017) 62-65
- [3] Coehoorn, Phys. Rev. B., 41. (1990) 11790

Computational screening of magnetocaloric materials

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The interest in the magnetic cooling devices has led to an intensive search for suitable well-performing magnetocaloric materials. High-throughput studies based on density functional theory calculations can significantly simplify and increase the range of this search. Computational screening makes possible sieving through thousands of known compounds without the need to perform time-consuming measurements and therefore can play an important role to detect new magnetocaloric materials.

We demonstrate results of an effective approach to the screening of magnetocaloric materials with the largest crystallography database provided by AFLOWLIB [1] as the initial source of material properties. To identify systems of interest several screening parameters were developed using properties of various well-known MCE materials as a reference. Along with magnetic properties, other factors important for practical applications are taken into consideration including price, availability and toxicity of candidate materials. Combining these criteria an automated algorithm for the screening process is suggested. It utilizes both information readily available in the database and additional *ab-initio* calculations. A step-by-step application of initial screening parameters to sort out unsuitable materials before performing more computationally heavy assessments allows fast processing of a large number of candidates. This results in a shortlist of promising compounds ranked by their potential to serve as a guide for experimental research.

[1] Richard H. Taylor, Frisco Rose, Cormac Toher, Ohad Levy, Kesong Yang, Marco Buongiorno Nardelli, Stefano Curtarolo, Computational Materials Science 93, 178-192, 2014

Unraveling the interplay between structural and magnetic properties of anodic iron oxide nanotubes for solar photoelectrochemical cells

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Hematite $(\alpha$ -Fe₂O₃) combines the leading vectors to stand as one of the most relevant materials for hydrogen production via photoelectrochemical solar water splitting [1]: low-cost, stability, scalability and efficiency. In particular, ordered arrays of hematite nanotubes have raised as highly promising for photoelectrochemical applications due to their 1D nature. However, the possible existence of multiple iron oxide-crystalline phases, that ultimately decrease the nanostructured photocurrent response, has so far been overlooked. In this work, we synthesized self-ordered iron oxide nanotube arrays using a simple and effective electrochemical anodization route and studied the effect of annealing temperature (400 - 700 °C) and nanotube length (up to 4 μ m) on the resulting crystalline-phase. This allows us to show a clear correlation between the structural and magnetic properties of the produced structures. Both plainly demonstrate that two iron oxide crystallographic phases (hematite and magnetite, Fe₃O₄) are always present, irrespective of the annealing conditions. In particular, temperature dependent magnetization measurements show the presence of two transitions: a magnetic anomaly at 190 K due to the antiferromagnetic spin flop transition of α -Fe₂O₃ (Morin transition) and a breakdown of the magnetization magnitude at 120 K arising from the electronic transition of Fe₃O₄ (Verwey transition). While the Verwey and Morin transitions are often misinterpreted, this study provides a solid foundation to use the magnetic characterization of iron oxide nanostructures as a key route to evaluate the presence of mixtures of magnetic Fe-oxides. Our in-depth study finally solves the contradictions in the literature regarding the magnetic contributions in nanostructured iron oxide.



[1] M. Gratzel, Nature 414(2001) 338-344

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Influence of mechanical deformation on the performance of magnetocaloric composite wires

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Magnetocaloric materials and prototypes have been investigated deeply for the last two decades. Next to Gadolinium, which is broadly seen as benchmark for room-temperature applications, LaFeSi-based alloys are considered as promising alternative [1]. One remaining problem towards application is the shaping of these brittle functional materials into effective heat exchanger geometries [2]. A method to overcome shaping difficulties is presented in [3], by producing composite wires (with 1 mm diameter) with a magnetocaloric material in a steel shell using the powder-in-tube (PiT) process.

However, knowledge of the underlying mechanisms that influence the magnetocaloric properties during the deformation and heat treatment processes is still lacking.

In this work we show the influence of mechanical deformation during the PiT-process on the microstructure and magnetic properties for both core materials, LaFeSi-based and Gadolinium for comparison.

By electron microscopy, micro-hardness testing and computed-tomography the influence of the fabrication process on the microstructure is reported. With these results we optimized the method presented in [3] by adding heat treatment steps, resulting in wires with diameters down to 500 μ m and a wall thickness below 100 μ m. Most importantly, this allowed to increase the mount of core material of above 60 vol. %.

The magnetocaloric effect is assessed by magnetic measurements. We show that by annealing the defects induced by mechanical deformation can be cured and initial magnetocaloric properties can be restored.

[1] A. Waske et al., MRS Bulletin 43 (2018) p. 269

[2] Franco et al., Progress in Materials Science 93 (2018)

[3] F. Funk et al. Mater Today Energy 9, (2018) p. 223

Second order magnetic phase transition and tunable magneto-caloric properties of La0.55CaxSr0.45-xMnO3 near room temperature

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Polycrystalline perovskite La0.55CaxSr0.45-xMnO3 (x = 0.00, 0.05, 0.1, 0.25) has been prepared by the solid-state reaction route using planetary ball milling technique. Room temperature XRD measurements show that all the samples possess Rhombohedral structure. Elemental compositions are determined by energy dispersive X-ray diffractometer (EDX). With increasing x content, lattice parameters are found to decrease resulting a significant change in the magnetic properties which could help tuning the magnetocaloric properties of the samples and bring the Curie temperature (TC) near room temperature (RT). Magnetic measurements reveal that with increased x content up to a maximum of 0.25, the Curie temperature decreased from 354 K to 311 K. Second-order FM-PM phase transition for all the samples is confirmed from Arrott plots using a criterion given by Banerjee. For x = (0.00, 0.05, 0.1 and 0.25), the maximum entropy change ($-\Delta S_m$) max are found (3.088, 2.83, 2.71 and 2.09) J/(Kg.K) respectively for $\Delta H=5$ T due to large variation of magnetization around their Curie temperature. The values of relative cooling power (RCP) are 216, 210, 208, 199 J/Kg for the samples with x = 0.00, 0.05, 0.1, 0.25. The high value of ($-\Delta S_m$) max, tunable TC and large RCP values make La0.55CaxSr0.45-xMnO3 as a competitive material for the magnetic refrigerator near RT.

Magnetic Composition of Thermoelectric PbTe:Cr

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The efficiency of the ever-so-much important thermoelectric energy conversion is described by the material thermoelectric figure of merit (*zT*). One of the most promising ways of *zT* improvement leads via band gap engineering towards the enlargement of the density of states at the Fermi level, e.g. via the band convergence [1], or a co-doping to induce a resonant level in the electric transport relevant band [2]. Cr in PbTe is a particularly interesting example since Cr atoms form a mixed valence $Cr^{2+/3+}$ donor centers resonant with the PbTe conduction band [3]. However, the solubility limit of Cr in IV-VI compounds is rather limited, 0.1 to 0.5%, depending on the exact growth conditions, so a nanocrystalline material is typically obtained: the host dilute magnetic compound (Pb,Cr)Te is enriched by various Cr-rich nanometer-sized crystallites. Therefore, the magnetic response of Cr-doped samples is complex and an extra care is needed to separate out and quantify all the relevant constituencies. In this report, we present results of such magnetic investigations of bulk PbTe:Cr crystals grown by the Bridgman method with nominal Cr content 0.5 < x < 1% [5].

It is shown that up to four different magnetic constituencies can be identified in the studied samples. We detect responses from three magnetically coupled systems and a paramagnetic (PM) one, revealed only at very low *T*. The superparamagnetic (SP) nature of the coupled systems (nanoprecipitates of various Cr_n - Te_m compounds) is clearly shown and the characteristic temperatures established. Low temperature magnetization curves allow to quantify PM contribution, from which the concentration of Cr^{2+} ions is evaluated. The magnitude of the magnetic saturation at *T* = 1.8 K indicates that the total concentration of Cr in the studied samples barely exceeds 0.1% and that about $\frac{3}{4}$ of the magnetic response comes from the substitutional PM Cr ions.

- [1] Y. Pei, X. Shi, A. LaLonde, H. Wang, L. Chen, and G. J. Snyder, Nature 473, 66 (2011)
- [2] J.P. Heremans et al., Science 321, 554 (2008)
- [3] T. Story et al., Acta Phys. Pol. 82, 879 (1992); W. Mac et al., ibid 87, 492 (1995)

Production of Fe-6.9wt%Ti alloy sheets for application in electric machines: Physical properties characterization

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Low magnetostriction values are interesting for using in power transformer's magnetic cores and electric motors' stators. In these machines the A.C. magnetic field created by the coil causes the core contraction and expansion which is due to the ferromagnetic material magnetostriction. Consequently, magnetostriction causes noise and for electric motors there is also the problem of vibrations that can cause the machine long-term structural damage or immediate failure. Nowadays, with the growing of urban areas, the residences are getting closer to the power transformers. Then, health problems due to the noise became an important issue. The Fe-6.5% wt.Si alloy would be a perfect material for this application because it has almost null magnetostriction, 1.8 T saturation magnetic induction and high relative permeability. However, it is a very brittle material. Nevertheless, Fe-Ti alloys with low Ti contents have also low magnetostriction values, lower than 10 ppm and particularly the alloy with ~ 8% at. about 6.9% wt. - of Ti has almost null magnetostriction as well as Fe-6.5% wt.Si alloy. For the present work, the alloy sheet of Fe-6,9% wt.Ti was already produced. The material was easily hot rolled down to the thickness of 0.5 mm, followed by one step at room temperature. After the rolling process, the sample was heat treated at 1250 °C for 12 h. The sheet electric and magnetic properties as electrical resistivity, hysteresis loss, coercive field and permeability will be measured and compared to that of the Fe-6.5% wt.Si alloy. Due to the reasonable ductility of the alloy Fe-6,9%wt. Ti and low magnetostriction, depending on the others properties, Fe-8%at.Ti alloy could be a good candidate to substitute Fe-6.5wt.%Si.

4164 3D printing of high rare-earth content NdFeB powder by LBM

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Since 1996 (first patent acceptation), a new process to create and shape alloys was developed: the laser beam melting (LBM). This technology use a laser to form a 3D shape from a 3D CAD (Computer Aided Design) file by repeatedly and selectively melting matter deposited in the form of thin powder beds. This way of producing parts allows virtually complete freedom of shape. However, it is very complex to understand the solidification process, and thus to get the proper microstructure in the printed parts.

For NdFeB magnets, which are found in nearly all electric device (motors and generators, actuators, sensors), it is essential to correlate the microstructure of the alloys according to the LBM process parameters and the starting powder composition to optimize the magnetic properties. In 2017, Jacimovic and his team [1] demonstrated the feasibility of the LBM technology applied to NdFeB alloys using the industrial powder MQP-S supplied by MagneQuench (19.7%wt of rare-earth). They obtained isotropic magnets and a microstructure showing the presence of a Fe phase, which is known to deteriorate the magnetic properties. The magnetic properties are not optimal with the MQP-S powder composition due to the low amount of rare earth (RE) which limit the formation of the magnetic phase, Nd₂Fe₁₄B, and also because magnets are not anisotropic.

One way to improve the performances of LBM printed magnets may stem from the use of a powder with a composition above the composition of $Nd_2Fe_{14}B$ (26.7%wt RE) and the development of an anisotropic microstructure. We will present the microstructural characterizations and magnetic results obtained using a homemade powder with 32.5%wt of rare earths.



Figure 1 : microstructure of the 3D printed NdFeB sample. In black, the Fe phase in shape of globules or small dendrites, in gray the Nd₂Fe₁₄B phase and in white, the Nd-rich phase. Cracks can also be found on the right side of the picture

[1] J. Jaćimović et al., Advanced Engineering Materials, vol. 19, nº 8, p. 1700098, 2017

4169 Magnetocaloric and barocaloric effect of metal complexes

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Solid state cooling is an alternative for the traditional gas-compression technology due to the environmental friendliness of its materials, energy efficiency, and low noise. Research in this field is focused on the development of advanced prototypes and smart materials; and the present work focuses on the description of a special family of quantum materials: metal complexes. At cryogenic temperatures (close to temperature of liquid He), some metal complexes present a huge value of magnetic entropy change, ranging from c.a. 10 J/kgK to c.a. 70 J/kgK (for 7 T of magnetic field change). These values make some metal complexes appealing as cryogenic coolant materials. We present a comprehensive collection of results from the literature, organised on a chart as a function of time, for different classes of metal complexes; those with 3d-3d magnetic interactions, 3d-4f coupling, and 4f-4f interactions. We observed that those materials that achieved the maximum value of entropy change, i.e., the spin-only value, follow an exponential scaling law with time. This result helps to predict a new class of metal complexes and further outcomes for the field. On the other hand, for a small amount of applied pressure, these materials produce a large barocaloric effect around the spin crossover transition (this transition occurs in a wide range of temperature, even close to room temperature). We introduce the SCO mechanism, along with the recent theoretical models and experimental results. The recent results of barocaloric effect are considered enormously significant (56 J/kgK for 0.9 kbar of pressure change, close to room temperature), even in comparison with traditional metallic barocaloric materials. We also provide perspectives for this subject, with discussions about new mechanisms for the models (as the Jahn-Teller distortion and orbital contribution).

Magnetic Properties of SrFe₁₂O₁₉ nanocrystallites: prospects for bi-magnetic nanocomposites

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Over recent years, magnetic nanoparticles have gained increasing attention due to their potential use as building blocks for next-generation permanent magnets [1]. These are employed in a multitude of energy-related technological applications [2]; hence, the transition from fossil to sustainable energy implies an ever-increasing demand for them. Nowadays, the risk of supply of rare-earth elements has stimulated an intense activity aimed to find novel critical-element-free materials with a $(BH)_{max}$ bridging the gap between hexaferrites and rare-earth-based permanent magnets [3]. In particular, our interest here is mainly focused on SrFe₁₂O₁₉ (SFO) nanoparticles, due to the fact that the inherent anisotropic shape of its crystallites makes them interesting candidates for hard/soft exchange-coupled magnets, thus, leading to the need of a systematic study of the magnetic interaction in such systems.

This work is aimed at investigating the magnetic properties of SFO nanocrystallites obtained by a sol-gel autocombustion approach [4]. The phase evolution during the reaction and subsequent calcination was monitored through X-ray powder diffraction (XRPD). The control of the crystallite size and morphology during these two steps is crucial to optimize the magnetic performance, due to its magnetocrystalline anisotropy. By addition of a surfactant, cetyl trimethylammonium bromide (CTAB), and changing the composition of precursors, it was possible to obtain phase-pure SFO at 800°C, instead of 1000°C for the conventional approach. The morphology of the nanocrystallites was studied using transmission electron microscopy (TEM). The nanoparticles show a hexagonal structure and it is possible to notice highly aggregated crystallites in the form of platelets. The crystallite sizes are estimated to be in good agreement with the average size obtained from the refinement of the XRPD patterns, showing values between 50 and 140 nm. To evaluate the performance of the SFO nanorystallites, their static magnetic properties were investigated at 300 K using a superconducting quantum interference device (SQUID) magnetometer. The magnetic behavior of the different samples is closely related to their morphology; the saturation moment (M_s), remanence (M_r), and coercive field (H_c) vary from 63.6 to 67.3 Am²/kg, 31.4 to 33.7 Am²/kg, and 468 to 480 kA/m, respectively. The highest energy product (BH)_{max} achieved was 8.9kJ/m³. Finally, we discuss the feasibility of using SFO for designing bi-magnetic nanocomposites, by growing SFO around CoFe₂O₄ (CFO) nanocrystallites, through a simultaneous biphasic synthesis route developed previously [5]. The structural and magnetic characterizations confirm that we have obtained a strongly coupled system, paving the way for controlling the size and distribution of hard/soft-phase regions by chemical engineering. We thank the Swedish Energy Agency and Swedish Research Council (VR) for financially supporting this work.

- [1] B. Balamurugan, D. J. Sellmyer, G. C. Hadjipanayis, R. Skomski, Scripta Materialia 67, (2012) 542
- [2] O. Gutfleisch, M. A. Willard, E. Bruck, C. H. Chen, S. G. Sankar, J. P. Liu, Adv. Mater. 23, (2011) 821
- [3] R. C. Pullar, Prog. Mater. Sci. 57, (2012) 1191
- [4] G. Muscas, P. Anil Kumar, G. Barucca, G. Concas, G. Varvaro, R. Mathieu, D. Peddis, Nanoscale 8, (2016) 2081

[5] F. Sayed, G. Kotnana, G. Muscas, F. Locardi, A. Comite, G. Varvaro, D. Peddis, G. Barucca, R. Mathieu, and T. Sarkar, Nanoscale Advances, 2, (2020) 851

Tuning of microstructure and grain boundary composition for optimized recycled magnets

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Since its discovery in 1983, Nd-Fe-B is the most important magnet material on the market. It combines the highest maximum energy product at room temperature with a satisfactory coercivity and temperature coefficient, so it is the choice of material when it comes to applications offering limited space and/or operating at elevated temperatures. Due to this properties, Nd-Fe-B-magnets are of strategic importance in various fields relevant for society today and in the future. However, mining and conditioning of the rare earth (RE) containing ores do not only have considerable negative effects on the environment but are also limited to a few locations worldwide. The reliability of supply is therefore considered as highly critical [1, 2].

Industrial recycling of RE permanent magnets from end-of-life applications is one measure to reduce criticality. Refeeding scrap magnet material back into the process chain not only lessens the supply risk but magnets made of recycled material are also ecologically favorable due to the elimination of mining and refining of the rare earth elements. Our study focusses on powder metallurgical recycling via hydrogen. This approach has the advantage of directly re-feeding the intermetallic compound Nd-Fe-B into the production chain. It therefore avoids metallurgical and chemical dissolving processes which involve enormous amounts of energy and chemicals. By using hydrogen, it is also possible to separate the magnetic material from the devices without additional processing steps in a simple manner, making it also a feasible technology to be used in countries with high labor costs.

We present innovative hydrogen-based technologies and concepts for functional (magnet-to-magnet) recycling of RE permanent magnets and demonstrate that recycled magnets are a suitable option for various applications [3]. Amongst others, scrap magnets having a determined neodymium content of only 28 wt.-% (and therefore just slightly above the stoichiometric composition of 26.7 wt.-%) and a comparably high carbon-content served as input material. In this case, addition of separately processed Nd-hydride was needed in order to get a dense magnet after sintering.

Magnets exposed to higher temperatures require a high coercivity and often feature an increased content of very critical heavy rare earth (HRE) elements like dysprosium or terbium. In order to optimize the required HRE content, we analyzed in detail the element distribution and mechanism of coercivity increase at atomic level via 3-dimensional atom probe (3DAP) tomography for two different recycling approaches: (i) Blending the recycled magnet powder with separately processed HRE hydrides or by (ii) application of a grain boundary diffusion process (GBDP). The results show how magnets with an optimized HRE distribution after microstructural engineering made of recycled material compare with primary magnets.

[1] K.P. Skokov and O. Gutfleisch, Scripta Materialia View Point Set, 154 (2018) 289-294.

[2] Study on the review of the list of Critical Raw Materials 2017, European Commission.

[3] O. Diehl, M. Schönfeldt, E. Brouwer, A. Dirks, K. Rachut, J. Gassmann, K. Güth, A. Buckow, R. Gauß, R. Stauber, O. Gutfleisch, Journal of Sustainable Metallurgy (2018) 1-13

High-throughput design of MM'X alloys for energy applications

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The MM'X family of compounds where M and M' are metals and X is a main group element crystalize in orthorhombic Pnma and hexagonal $P6_3$ /mmc structures. They have been investigated for various energy applications, namely magnetocaloric [1] and barocaloric [2] cooling, permanent magnets [3] and antiferroelectrics [4].

For specific alloys, such as MnNiGe, a structural transition occurs between the low temperature Pnma and high temperature P6₃/mmc phases [5]. Compositional substitution can be used to induce a first-order magnetostructural transition, leading to a large entropy change, and thus an enhanced magnetocaloric effect. For a structural transition to occur the martensite Pnma phase must be the lowest energy structure and the austenite P6₃/mmc should also be stable [4]. Therefore, the underlying design principle to optimize MCE in MM'X is to tune the structural transition to occur near room temperature and in the Curie temperature window spaned between the martensite and austenite phases [6]. This is has been realized by isostructural substitution, for instance, by alloying between MnNiGe with high temperature transition and FeNiGe where only the P6₃/mmc phase is found, the transition is lowered to room temperature [7].

We performed a high-throughput density functional theory (DFT) computational screening of novel parent compounds, to accelerate the search for novel magnetocaloric MM'X phases. Specifically, we considered M=(V, Ni, Mn, Fe, Co, Cr), M'=(Sc, Ti, Zr, Hf, Nb, Cr, Mo, W, Mn, Fe, Co, Ni, Cu, Zn) and X=(Mg, Ti, Zn, Ti, B, Ga, In, Si, Ge, Sn, Pb, P, As, Sb, Bi). We first assess the synthesizability of the martensite and austenite phases by computing the formation energy (stability in respect to the elemental solids) and distance to convex Hull (decomposition to competing phases). A detailed analysis reveals 79 magnetic compounds with possible structural transitions. Furthermore, we evaluated mechanical (elastic constants) and dynamical stabilities (phonons) and magnetic ground state of these selected compounds. The Curie window is obtained by calculating the Curie temperature for both phases using first-principle exchange constants and Monte Carlo modeling, with further estimation of the structural transition temperature obtained by using the QHA Debye model based on ab-initio elastic constants [8]. In this way, we indentified novel MM'X alloys with structural transition and mapped out possible isostructural substitution routes for experimental optimization.

Lastly, our screening suggests several compounds as potential gap magnets with magnetic anisotropy energy between that of MnAl and SmCo5.

- [1] J. Liu Scientific et al., Reports 6, 23386 (2016)
- [2] P. Lloveras et al., APL Materials 7, 061106 (2019)
- [3] T. N. Lamichhane et al., Appl. Phys. Lett. 109, 092402 (2016)
- [4] J. W. Bennett el al., Phys. Rev. Lett. 110, 017603 (2013)
- [5] W. Bazela el al., Phys. Stat. Sol. A 64, 367 (1981)
- [6] E. Liu et al., Nat. Commun. 3, 873 (2012)
- [7] A. Taubel et al., J. Phys. D: Appl. Phys 51, 464005 (2017)
- [8] O. L. Anderson, J. Phys. Chem. Solids 24, pp. 909-917 (1963)

This work was supported by the European Research Council (ERC) under the 457 European Union's Horizon 2020 research and innovation programme (Grant No. 743116-458 project Cool Innov).

Effect of hydrostatic and chemical pressures on spin-reorientation transition, magnetic and magnetocaloric properties of Fe₇Se₈ single crystals

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The effect of hydrostatic and chemical pressures on structural and magnetic properties of $Fe_{7-x}A_xSe_8$ single crystals (where A = Co, Ni) have been studied. The substitution of Fe with Co or Ni induces a systematic change in the unitcell volume, which is dependent on the average ionic radius of the transition metal. The Fe_7Se_8 single crystal is ferrimagnetic metal with Néel temperature equal to $T_N = 450$ K. The spin reorientation transition (SRT) from easy caxis to easy c-plane has been found at the temperature $T_r \approx 125$ K. It proceeds abruptly, as a first-order phase transition [1]. Thermal hysteresis is unexpectedly small for this transition. The "chemical pressure" affects both the magnetic ordering temperature and the temperature of the SRT. The hydrostatic pressure has been found to have a similar effect on both temperatures. A correlation between hydrostatic and chemical pressure can, therefore, be established from the measurements of phase transition temperature. The dual charge-state model of Fe ions, resulted by the temporary residence time of vacancy at Fe^{2+} site, has been suggested as a mechanism of T_r shifting. Magnetic measurements show the decreasing of Fe_7Se_8 magnetization under applied hydrostatic and chemical pressures. It has been observed, that the rotating entropy ΔS_{rot} and refrigeration capacity RC magnitudes are in strong dependence from T_r and magnetization. Figure 1 presents the effect of chemical (related to the concentration x of Co and Ni ions) and hydrostatic pressures on reorientation temperature T_r . Obtained results confirm a strong correlation between hydrostatic and chemical pressure in Fe_7Se_8 crystals.

Figure 1. Correlation between spin-reorientation temperature and dopant concentration *x* compared with the influence of hydrostatic pressure on T_r for pure Fe₇Se₈.



[1] I. Radelytskyi et al., J. Appl. Phys. 124, 143902 (2018)

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3245 Ab-initio approach for a "simple" reliable entropy estimation

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High-throughput studies based on first-principles calculations represent a potentially powerful approach to detect new magnetocaloric materials for application in magnetic cooling devices and help to estimate trends for material tuning. In order to identify relevant systems in a large body of data, screening parameters are required and must be carefully chosen considering a balance between accuracy and cost of the calculations. A key quantity to characterize magnetocaloric cooling performance is the entropy variation between two magnetic phases. To estimate this quantity in a cost-efficient but accurate way, we test several approaches taking FeRh as a test system. A methodology for a reliable first-principles estimation of the entropy variation between magnetic phases is proposed, considering three distinct and independent entropy contributions: electronic, lattice, and magnetic, and respective results discussed and presented.

Unravelling the mechanism of the one-pot synthesis of exchange coupled Co-based nano-heterostructures with high energy product

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Permanent magnets are key elements for many technological devices that are largely used in everyday life, such as electronic devices, hard disks, automotive, wind turbines, and hybrid electric vehicles. The most powerful magnets currently used in a wide part of industrial necessities, comprise Rare Earth (RE) elements, which are expensive, harmful to the environment and present a high supply risk. [1] At present, a strong research activity is thus focused on finding viable alternatives to RE-based permanent magnets, which can allow their replacement at least in all those applications where high performances are not strictly required. This performance is determined by the magnetic energy density that can be stored within the material, which is represented by the maximum energy product (BH)max.

A significant enhancement of the magnetic performance of traditional ferrite nanoparticles (single phase), can be obtained by reducing the grain size to the nanoscale. Moreover, a further improvement can be achieved producing heteronuclear nanoparticles (NPs), whose magnetic properties are driven by inter or intraparticle exchange – effects, like exchange bias or spring magnet. [2] In this work we present and discuss, for the first time, the formation of $Co/Co_yFe_{3-y}O_4$ nanoparticles by a thermal decomposition method which uses metal oleates as precursors (Iron - cobalt and sodium oleate). The characterization of the obtained nanopowders by High Resolution Transmission Electron Microscopy (HRTEM), Powder X-Ray Diffraction (XRD), Scanning Transmission Electron Microscope (STEM), Electron Energy Loss Spectroscopy (EELS) and X-Ray Photoelectron Spectroscopy (XPS), shows that polyhedral nanoparticles with coexistence of soft Co and non-stoichiometric hard $Co_yFe_{3-y}O_4$ were obtained. Thermogravimetric (TGA) analysis demonstrated that controlling the thermal stability of the reactants in the presence of an excess of Sodium Oleate (NaOL), nanoparticles with different morphology, architecture (core-shell or heterodimers), crystal structure and composition could be obtained. This, in turn allows for the modulation of the heteronuclear nanoparticles's magnetic properties, and particularly for the enhancement of the energy product.

Coercivity and anisotropy measurements on GdCo_(5-x)Cu_x single crystals

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Rare-earth transition-metal permanent-magnets possess remarkably high anisotropy energies and therefore may express high coercivities necessary for permanent-magnet applications. Nonetheless, coercivity is commonly limited to approximately 20-30% of the anisotropy field (Brown's paradox) and even less may be achieved experimentally. Coercivity mechanisms driven by exchange interaction (e.g. exchange bias) recently observed in compensated bulk Heuslers [1] and Heusler segregations [2] suggest a pathway to challenge the Brown's paradox. Though not fully understood, the coexistence of ferromagnetic (FM) and antiferromagnetic (AFM) clusters seems to be advantageous in these systems. Materials with a compensation point (e.g. GdCo_{5-x}Cu_x [3]) present themselves as model objects to study giant coercivity near the FM-AFM transition. Fundamental understanding of these mechanism may lead to tuning defects in permanent magnets to significantly enhance coercivity. Moreover, the magnetic viscosity appears to be small in these samples [4] rendering the system a candidate for high frequency applications.

We investigated a series of ferrimagnetic $GdCo_{5-x}Cu_x$ single crystals ($x \in 0.5, 1, 1.5, 2$), which all show the $CaCu_{5-x}$ type crystal structure and traverse a composition dependent compensation point. We show that the samples abruptly reverse their magnetisation if oriented parallel to the c-axis (Fig. 1(a)) both, above and below the compensation point. At room temperature, the Gadolinium sublattice is dominating the magnetism in $GdCo_3Cu_2$ and the Cobalt sublattice in $GdCo_4Cu$. For the intermediate composition $GdCo_{3.5}Cu_{1.5}$ the compensation point is close to room temperature. In the basal plane however, magnetisation will linearly increase with external magnetic field (Fig. 1(b)). Switching fields of one to two Tesla are realised close to the compensation point, where these do not match the coercive fields. The latter are several times larger, reaching values above six Tesla, whereas magnetisations reach minimum values. Therefore, strong pinning forces are expected when applying such a material design concept to permanent magnets. By adjusting the composition of the ferrimagnetic inclusions, the magnets may be optimized for different operating temperatures.



Figure 1: Field dependent magnetic polarization of $GdCo_{5-x}Cu_x$ single crystals (a) for x = 1, 1.5 and 2 measured parallel to the c-axis at 300 K and (b) for x = 1 measured parallel to the c-axis and a-axis respectively, below (150 K) and above (200 K) the compensation point. The inset shows a Laue-diffraction pattern of the single crystal.

- [1] Nayak et al. Nature Materials, 2015, 14, 679
- [2] Çakir, Scientific Reports, 2016, 6
- [3] Grechishkin et al. Applied Physics Letters, 2006, 89, 122505
- [4] de Oliveira et al. Journal of Magnetism and Magnetic Materials, 2011, 323, 1890

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Fabrication of DyVO₄ nanowires and their magnetocaloric effects

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Conventional vapor-compression refrigeration technology has practical limitations because of its environmental destruction. Recently, new cooling technologies to replace it have been underway, including thermoelectric cooling, thermoacoustic refrigeration, and magnetic refrigeration, etc. Among them, magnetic refrigeration is based on the thermodynamic magnetocaloric effect (MCE). In this presentation, microwave hydrothermal synthesis was employed to fabricate DyVO₄ nanowires as a MCE material for hydrogen liquefaction. During synthesis, EDTA was added as a chelating agent to obtain uniform DyVO₄ nanowires (sample A), and their magnetic properties were compared with those of irregular shaped DyVO₄.nanoparticles (sample B). The structure and morphology were studied by using scanning electron microscope (SEM) and transmission electron microscope (TEM) measurements. The Neel temperature (T_N) of the sample A was higher than that of the sample B due to suppression of antiferromagnetism induced by the size effects of the sample B. The Curie-Weiss fitting showed the weak antiferromagnetism for the both samples. However, the effective magnetic moments for the both samples had lower values than that of bulk DyVO₄ because of the size effects, where uncompensated spins on the surface of the nanostructures induce suppression of antiferromagnetism.

The maximum magnetic entropy change $(-\Delta S_M)$ of the sample A was higher than that of the sample B at H = 50 kOe, and consequently led to an increase in relative cooling power (RCP), a measure of MCE. It is known that DyVO₄ has a structural transition from tetragonal to orthorhombic symmetry around 14 K due to the Jahn-Teller effect induced by the ordering of quadrupole of Dy³⁺ ions [1]. The magnetic entropy changes (- ΔS) with temperature showed an anomaly of - ΔS due to the Jahn-Teller effect for the both samples, which was dependent on the strength of the magnetic field. The quadrupole coupling of DyVO₄ resulted in an enhancement of RCP due to an increase of the full width of the maximum of temperature.

Chiral Anomaly and Anomalous Hall Effect in Hexagonal-Mn_{3+δ}Ge

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Topological quantum materials have attracted enormous attention since their discovery due to the observed anomalous transport properties, which originate from the non-zero Berry curvature. $Mn_{3+\delta}Ge$ has gained special attention because of its large anomalous transport effects that persist starting from Néel temperature (365 K) down to 2 K. Due to the presence of very small in-plane ferromagnetic component, the chirality of magnetic structure can be controlled easily by applying just a few hundred Oersted (Oe) of the magnetic field.

Hexagonal - $Mn_{3+\delta}Ge$ stabilizes in the range of $\delta = 0.2 - 0.55$. In order to understand the involved quantum phenomena - Anomalous Hall effect (AHE) - in such materials, it is also important to check the stability of AHE with the variation of δ . Due to specific mirror symmetry of the triangular antiferromagnetic structure, AHE is expected to be observed when magnetic field (B) is applied along *x* or *y* crystallographic axis. AHE has been reported in the lower range of δ (= 0.22, 0.32 [1]), however the upper range of δ was still unexplored. We have investigated samples with the upper range of δ (~ 0.55) and AHE with very small Hall - hysteresis (<200 Oe) was observed when the magnetic field was applied along x or *y* crystallographic axis. The magnitude of AHE in $Mn_{3+0.55}Ge$ is found to be more than 25% larger than the reported AHE for samples with $\delta = 0.22$, 0.32 [1]. In addition to this, low Fe doping in $Mn_{3.2}Ge$ has also shown AHE of the same magnitude as observed in the case of $Mn_{3+0.55}Ge$.

Despite being considered as a Weyl semimetal, chiral anomaly (signature for the presence of Weyl points) has not been observed in $Mn_{3+\delta}Ge$ yet. To establish the claim for the existence of Weyl points in $Mn_{3+\delta}Ge$, transverse and longitudinal magneto-resistance (MR) measurement was performed with the magnetic field and electric current applied along several combinations of *x*, *y*, *z* crystallographic axes. Angle dependent measurements between the direction of current and applied magnetic field has clearly shown the presence of negative longitudinal MR as long as I||B. Negative longitudinal MR is observed over a long range of magnetic field and temperature. However, the nearly monotonic increase in magneto-resistance with angle (θ) between I and B is observed only for the intermediate magnetic field range (0.5 T- 2 T). This behaviour is the signature of the chiral anomaly, which evidently supports the claim for the presence of Weyl points in $Mn_{3+\delta}Ge$ compounds.

[1] Kiyohara et al. (2015)

High-field magnetic phase transitions and crystal-field interactions in the HoFe₁₁Ti

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In the past decades, the rare-earth (R) – Fe intermetallics with the ThMn₁₂ structure have been closely investigated as potential candidates for rare-earth-lean permanent magnets [1-3]. These 3d-4f compounds are ferrimagnets (the R and Fe magnetic moments are antiparallel to each other) for heavy rare earths. High enough applied magnetic fields force the system to a gradually transition to the ferromagnetic phase. A previously developed phenomenological quantum model based on the interconnection between the crystal-electric-field and the 3d–4f exchange interactions [1,4,5] has been applied to HoFe₁₁Ti and its hydride. In this work we perform the high-field magnetization study of HoFe₁₁Ti and HoFe₁₁TiH both experimentally and theoretically. The magnetization curves were measured on single crystals in fields up to 60 T at 4.2 K along three main crystallographic directions. We evaluate the crystal-field and exchange parameters using high-field magnetization data for HoFe₁₁Ti and HoFe₁₁TiH up to 100 T (see Fig. 1). The developed model allows to study the high-field induced magnetic phase transitions from ferri- to the forced-ferromagnetic state via intermediate transitions. We calculate the H-T magnetic phase diagrams up to 100 T and up to room temperature and investigate all observed features in detail. We demonstrate that the transition from ferrimagnetic to the forced-ferromagnetic state in HoFe₁₁TiH occurs at magnetic fields higher than 60 T.



Figure 1: Theoretical (dashed lines) and experimental (solid lines) magnetization curves obtained for HoFe₁₁Ti and HoFe₁₁TiH single crystals in magnetic fields applied along the [001] axis at 4.2 K.

[1] C. Piquer, F. Grandjean, O. Isnard, G.J. Long. J. Phys. Condens. Matter. 18, 221 (2006)

[2] Y. Skourski, J. Bartolom'e, M. Kuz'min, K. Skokov, M. Bonilla, O. Gutfleisch, J. Wosnitza, Journal of Low Temperature Physics, vol. 170, no. 5-6, pp. 307–312 (2013)

[3] W. Körner, G. Krugel, C. Elsässer, Scientific Reports. 6, 24686 (2016).

[4] I.S. Tereshina, N.V. Kostyuchenko, E.A. Chitrova Tereshina, Y. Skourski, M. Doerr, I.A. Pelevin, A.K. Zvezdin, M. Paukov, L. Havela, H. Drulis. Scientific Reports, 8:3595 (2018)

[5] N.V. Kostyuchenko, A.K. Zvezdin, E.A. Tereshina, Y. Skourski, M. Doerr, H. Drulis, I.A. Pelevin, I.S. Tereshina, Phys. Rev. B, vol. 92, p. 104423 (2015)

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Tuning the Magnetocaloric Effect towards Room Temperature by B-site Doping in the perovskite $La_{0.8}Sr_{0.2}MnO_3$

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Towards development of anisotropic a-Fe/Nd2Fe14B nanocomposite powder using hydrogenation disproportionation desorption recombination process

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Nd-Fe-B based permanent magnets show the largest maximum energy product, $(BH)_{max}$, among all types of permanent magnets and with this are the material of choice for energy conversion and mobility applications such as wind turbines and hybrid/electric vehicles [1,3]. The largest $(BH)_{max}$ reported in the anisotropic Nd-Fe-B sintered magnets is 474 kJ/m³ that is 92% of the theoretical limit for Nd₂Fe₁₄B phase [3]. With a remanence of $\mu_0 M_r$ =1.55 T reaching 96.6% of the saturation magnetization of Nd₂Fe₁₄B phase (1.61 T), significant further improvements here are unlikely.

Nanocomposite microstructure composed of exchange coupled hard and soft magnetic phases enable realizing remanence enhanced permanent magnets based on the so-called exchange spring mechanism [4]. Despite large amount of research, realizing anisotropic magnets with high coercivity has been difficult and no useful top-down approach has been successful so far.

In this work, we demonstrate a novel top-down processing route to synthesize anisotropic nano-composite powders [5]. The route is based on the hydrogenation disproportionation desorption recombination (HDDR) process and allows the microstructure design of nanocomposite magnets in the α -Fe/Nd₂Fe₁₄B system when using appropriate alloy compositions. The influence of HDDR processing conditions as well as detailed microstructural characterization results will be presented. In addition, micromagnetic simulations for optimum microstructure design of a nanocomposite α -Fe/Nd₂Fe₁₄B magnet will be correlated with high resolution electron microscopy as well as atom probe tomography.

[1] O. Gutfleisch, M.A. Willard, E. Brück, C.H. Chen, S.G. Sankar, J.P. Liu, Adv. Mater. 23 (2011) 821-842

[2] H. Sepehri-Amin, S. Hirosawa, K. Hono, Handb. Magn. Mater. 27 (2018) 269-372

[3] Y. Matsuura, J. Magn. Magn. Mater. 303 (2006) 344-347

[4] E.F. Kneller, R. Hawig, IEEE Trans. Magn. 27 (1991) 3588-3600

[5] H. Sepehri-Amin, I. Dirba, X. Tang, T. Ohkubo, T. Schrefl, O. Gutfleisch, K. Hono, Acta Materialia 175 (2019) 276-285

Favoring the LTP phase in MnBi alloys and optimum conditions to obtain the highest magnetization values

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MnBi alloy is known to have 2 crystallization systems, one driven by high temperature treatments i.e. HTP (high temperature phase) with orthorhombic structure [1] and a second one driven by a low temperature treatment i.e. LTP (low temperature phase) with hexagonal structure [2]. The challenge of this type of alloy is to obtain the LTP phase as pure as possible without α -Mn, Bi or MnBi HTP phase in order to reach for the highest magnetization and coercivity values. Currently, the largest coercivity of thermally treated MnBi ingots with a majority of LTP phase is 7.11 kOe measured at room temperature [3]. Until now, various techniques had been used to obtain MnBi alloys most of them including arc-melting in an inert atmosphere. The main issue of this technique is to avoid Bi evaporation during melting and keeping a good atomic ratio of Mn and Bi of 1:1 to favorize the LTP phase formation. In order to limit the previous mentioned risk, several samples with Mn:Bi 51:49 at% were prepared using Mn powder <300 µm and Bi powder <150 µm by compacting and treated them in vacuum at different temperatures in range of 280°C - 300°C. The XRD results indicated a growth of MnBi alloy formation for the samples treated at 280°C, 290°C and 300°C for 24h starting from 35% wt MnBi (in which 13% wt LTP), to 45% MnBi (in which 23% LTP) and 52% wt MnBi (in which 24% wt LTP) respectively. Coercivity and remanent magnetization values were in range of 0.47 – 0.34 kOe and 4.49 – 1.05 emu/g, indicating a partial formation of magnetic phase. Furthermore, the previously treated sample at 290°C was melted by induction melting and treated at 400°C facilitating the formation of MnBi alloy to a 75% in which 37% is LTP. The melted and retreated alloy has a M_s of 40.07 emu/g, M_r of 3.46 emu/g and Hc of 0.19 kOe measured at room temperature. The results improved over successive treatment by rising the MnBi LTP content in the overall alloy ingot, indicating that the current trend facilitate the formation of magnetic phase.

- [1] H. Göbel, E. Wolfgang, H. Harms, Phys. Status Solidi. 35 (1976) 89–95
- [2] A.F. Andersen, J.E. Engebretsen, J. Refsnes, ACTA Chem. Scand. 26 (1972) 175-190
- [3] Y.L. Huang, Z.Q. Shi, Y.H. Hou, J. Cao, J. Magn. Magn. Mater. 485 (2019) 157–164

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Thermal switches in solid state magnetic refrigeration: conductivity change requirements and effects

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The efficient use of energy has become a mainstay in the current world economic growth. The fact that approximately 30% of all energy consumed in the world is used in cooling and heating engines places refrigeration on the top of the priority lists of any modern economy [1]. In that respect, vapour compression is still the most used refrigeration technology, but requires bulky and noisy compressors. Alternative thermoelectric refrigerators are more compact and environmentally friendly, but at the cost of a much lower efficiency and cooling capacity [2]. In a different rank, mainly due to its higher efficiency, is magnetic refrigeration (MR). With the discovery of the giant magnetocaloric effect by Percharsky and Gschneider, it is now possible to induce large temperature variations and develop MR applications near room temperature [3]. MR is hence an excellent candidate to replace the vaporcompression technology. However, there are still obstacles to overcome, such as the use of moving parts and fluids that can easily reduce the working lifetime of the refrigerators. To solve these issues, the use of solid-state thermal switches was proposed [4, 5]. Instead of using fluids to transfer heat from one reservoir to the other, this task can be achieved by externally controlling the thermal conductivity (k) of solid materials that establish contact between the magnetocaloric material (MCM) and each of the reservoirs. Such thermal switches (TSs) are already used in space applications and cryogenics, although involving fluids or mechanical mechanisms. Even though some experimental results have been reported for Peltier based TSs, the performance of a fully solid-state magnetic refrigerator making use of TSs has been addressed mostly by means of numerical simulations [4-6]. However, until now, the numerical studies always considered ideal TSs which behave as perfect insulators (k = 0) in the OFF state. In this work, we numerically searched for the most favourable thermal properties of non-ideal solid TSs. We based the TS properties on real materials, commonly used as thermal conductors, and simulated a percentage change of their conductivity (Δk ; from 0% to 100%) for different operating frequencies (f) and working temperatures (T₀). For each parameters combination, the temperature span between the hot and cold reservoirs (ΔT) was registered and mapped. Our results show that for high performance both thermal switches with near ideal behavior are required. However, for an intermediate performance, only one thermal switch with acceptable behavior is needed. We discuss all the possibilities.

[1] U. N. E. Programme, The importance of energy efficiency in the refrigeration, air-conditioning and heat pump sectors, Briefing note, 2018

- [2] A. Majumdar, Nat. Nanotechnol. 4 (2009) 214
- [3] V. K. Pecharsky et al., Phys. Rev. Lett. 78 (1997) 4494
- [4] D. J. Silva et al., Appl. Energ. 93 (2012) 570
- [5] D. J. Silva et al., Appl. Energ. 113 (2014) 1149
- [6] D. J. Silva, et al., Int. J. Energ. Res. 43 (2018) 742

Fe_{79.7-x}ETM_xNb_{0.3}B₂₀ (ETM = Cr, Mn) glassy materials for energy recovery

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The increasing demand of energy is requiring the continuous diversification of the production sources, and the consideration of alternative potential resources such as the waste heat from industrial processes or residential heating systems. However, new devices and materials must be designed to recover efficiently the waste heat. Among them, the thermomagnetic materials are an interesting alternative. The ideal thermomagnetic materials for energy harvesting devices should have a sharp ferro-paramagnetic phase transition, with minimal hysteresis and minimal latent heat and be accurately tuned to the available waste-heat temperature. Moreover, it should have good thermal diffusivity, withstand repeated cycling, and preferably only contain abundant, low-cost elements.

Therefore, the main objective of this work is to thoroughly investigate the thermomagnetic characteristics and their connection with the microstructural features for $Fe_{79.7-x}ETM_xNb_{0.3}B_{20}$ (ETM = Cr, Mn; x = 11.5÷20 at.%) rapidly quenched alloys, depending on the ETM content. The possibility to tailor T_c around room temperature and their minimal hysteresis recommends these glassy alloys for energy conversion applications. Our aim is to study the complex interrelation among structure, mechanical, magnetic and thermal properties and, subsequently, to design new energy conversion devices with enhanced efficiency.

UHR-TEM studies indicate glassy structures with clusters of 2-3 nm embedded within an amorphous matrix for most of the investigated samples. Mn additions double the saturation magnetization (80 emu/g) compared with Cr compositions (40 emu/g). The saturation magnetization decreases slowly by increasing the amount of Mn and the material behaves as a weak ferromagnet for ETM contents of 20 at.%. The larger the Mn content, the more reduced the Curie temperature is, ranging from ~350 K for x = 12 at.% to ~190 K for x = 16 at.%.

However, the Curie temperature is strongly dependent on the addition element. Mn is antiferromagnetic and behaves in a similar way as Cr does, favoring the formation of Fe-Mn antiferromagnetic clusters surrounded by ferromagnetic Fe-B-based amorphous matrix. Thus, the larger the Mn content the more numerous the antiferromagnetic interactions are, suggesting a transition temperature evolving more towards a Néel temperature for Mn contents over 16 at.%. Moreover, the precipitation of a crystalline phase would alter the yield strength and elasticity of the ribbons significantly and change the elemental composition of the matrix influencing the magnetization. All these aspects will be considered when defining the optimum thermomagnetic materials for our particular energy conversion application.

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Giant magnetocaloric effect in MnAs1-xPx in a cyclic magnetic field: lattice and magnetic contributions and degradation of the effect

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Magnetic materials, which are considered as working bodies for solid-state magnetic refrigerating machines, need to have a specific set of properties. It is evident that the main of them is the manifestation of a giant magnetocaloric effect in moderate magnetic fields. Several families of materials in which an isothermal entropy change or an adiabatic temperature change reaches significant values are currently known. The most studies of the magnetocaloric properties of materials are carried out either in quasi-stationary fields or under a one-time cycle of the magnetic field application, and such results are given as reference data. However, the refrigeration machine, like of the most machines, is a device with periodic operating cycles. Therefore, there is a need to study the magnetocaloric properties of materials under the continuing application of the cyclic magnetic field.

In this report, we present results of the direct measurements of the adiabatic temperature change in $MnAs_{1-x}P_x$ compounds (*x*=0, 0.02, 0.025, 0.03) in cyclic magnetic fields with amplitude up to 8 T. The substitution of As by P results in a slight shift of the Curie temperature and notable change in the MCE value. Estimations of the lattice and magnetic contributions show that in MnAs compound the lattice contribution dominates (about 70 % of the total MCE). Substitution of As with phosphorus leads to a decrease in the total value of the MCE, which is caused by a decrease in the lattice contribution, and the magnetic contribution almost does not change in absolute value. A reversible effect of degradation of the magnetic cooling technology.

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Optical Actuation of Pyrolytic Graphite Diamagnetically Levitating Over Smart Magnets

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Pyrolytic graphite (PyG) passively levitating over permanent magnet arrays can be optically actuated [1, 2]. A new approach is to print magnetic domains into block permanent magnets [3], simplifying construction while allowing more complexity in the arrangement of magnetic domains. The ability to print arbitrary magnetic domain patterns permits rapid development of application-specific actuation tracks for levitated samples. To study the effect of printed magnet domain layouts, PyG dimensions, and optical power on the actuation phenomenon, numerical simulations are required. As a demonstration of the simulation approach, we present a 3D FEM corresponding to an experimental setup where a PyG sample (Panasonic PGS, 252m) levitates above a commercially available printed magnet (Polymagnet 421) and actuated using a 600mW 975-nm laser. The printed magnet features a dot pattern of one magnetic polarity against a background of the opposite polarity as shown in Fig. 1. Modeled PyG magnetic susceptibility temperature dependence follows the existing literature [4]. Laser input is modeled as incident heat flux on the upper surface of the PyG domain. Heat capacity and convective cooling effects are tuned empirically using infrared video captures of a laser-irradiated sample. Simulation results without laser input highlight the magnetic potential barriers that keep a passively levitating PyG sample in static equilibrium in the plane of the magnet array and indicate the locations of static equilibrium as shown in Fig. 2. To achieve translational actuation, these barriers must be overcome by asymmetric, optically induced thermal forces when the laser is applied. This insight concerning barriers to actuation is lost in simplified analytical expressions [5] that are approximately correct only when the PyG sample is significantly larger than the spatial variation in magnetic domains. Fig. 2 shows an increase in y-directed force resulting from laser irradiation at the y-most edge of a circular sample in a timedependent but not motion-coupled simulation.





Figure 1 : (a) Simulation geometry: blue and red indicate zoriented magnetic north and south domains, respectively,(b) corresponding experiment, and (c) magnet with printed domains visible under viewing film

Figure 2: (left) Normalized in-plane forces $(F_x^2 + F_y^2)^{-1/2}$ without laser input relative to displacements of a 10mm diameter PyG sample from the geometric center of a printed magnet at a fixed distance above the magnet (0.58mm). The underlying magnetic domain pattern in this section of the magnet (2mm diameter black circles) is overlaid for clarity. (right) Increase in *y*-directed magnetic force and maximum temperature difference on a 10mm diameter PyG sample versus time as a simulated 600mW laser begins to irradiate the *y*-most edge of the sample at 0s. The inset shows temperature profiles on the PyG surface at 0s, 1s, 2s, and 3s.

- [1] M. Kobayashi et. al., J. Amer. Chem. Soc. 134 20593, 2012.
- [2] M. Ewall-Wice et al., IEEE Trans. Magn. 55, 2501506, 2019.
- [3] G. Küstler, Electronics Letters 50, 1289, 2014.
- [4] C. Siyambalapitiya, Ph.D. dissertation, Univ. South Florida, 2012.
- [5] M. Fujimoto et al., Phys. Rev. B. 100, 045405, 2019.

A miniaturized stepping motor comprising isotropic Pr-Fe-B thin magnets

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Many studies have been conducted on the preparation of miniaturized actuators together with MEMS (Micro Electro Mechanical Systems) comprising a thin permanent magnet. For example, a sputtering-made Nd-Fe-B film was applied to a small motor by Yamashita *et al* in 1991 [1] and Töpfer *et al*. took advantage of the multi-pole magnetization for a small Nd-Fe-B magnet applied to magnetic micro-actuators [2]. Furthermore, various micro-actuators using a Nd-Fe-B film prepared by a sputtering method have been demonstrated by Shinshi *et al*. [3]. Our group reported an axial gap-typed motor and a friction drive motor including a PLD (Pulsed Laser Deposition)-fabricated Nd-Fe-B film [4]. Here, we focused on small stepping motors in order to apply them to a medical application. The characters of the motor include the easiness of position control without a sensor. It is generally known that conventional Nd-Fe-B bonded magnets have been used for the motors. However, the magnetic properties of the bonded ones are inferior to those of conventional sintered magnets because the bonded magnets contain a nonmagnetic phase such as thermosetting resin. Recently, we succeed in the deposition of Pr-Fe-B magnet powders with coercivity (H_{cj}) exceeding 1000 kA/m on a thin stainless shaft by using the PLD method [5].

The multipole magnetization was carried out as follows. In order to adjust the magnetization characteristics, thermal demagnetization of Pr-Fe-B magnets was adopted. After rapidly heating the Pr-Fe-B magnets to temperatures above the Curie point in the air, cooling it in the magnetizing field of the permanent magnets was carried out. Sm-Co sintered magnets were used as magnetizing ones, and a heater in the magnetizing area was prepared. The above-mentioned micro-magnetization equipment enabled us to obtain 10 magnetic poles for a magnet. The thin magnet with multipole magnetization was loaded in a stepping motor with the diameter of 2.9 mm, and the operation of the motor was observed by using a conventional test device (MCH-5B, NPM cooperation). At present, it was difficult to measure the motor performance by concreate physical (mechanical) value such as the value torque because the value was so small. Instead, we confirmed the stable operation of the ultra-small stepping motor. Moreover, magnetic properties as a function of temperature was evaluated from the future application point of view. At room temperature (296 K) and 348 K, the energy product of (BH) at Pc of a PLD-made Pr-Fe-B magnet were 74 and 65 kJ/m³ which were higher than those (57, 47 kJ/m³) of a Nd-Fe-B bonded one. However, the (BH) value at 398 K for the bonded magnet exceeded the value of the Pr-Fe-B one. In the case of a Pr-Fe-B magnet, temperature coefficients of Br and Hcj were calculated at -0.20 and -0.84 % / K. The both values were inferior than those (-0.13 and -0.45 % / K) of Nd-Fe-B bonded one. The result indicates that the use of a PLD-made Pr-Fe-B thin magnet was not suitable under high temperature such as approximately 400 K.

- [1] S. Yamashita et al., J. Appl. Phys., vol. 70, no. 10, pp. 6627-6629, 1991.
- [2] J. Töpfer et al., J. Magn. Magn. Mater., vol. 270, pp. 124-129, 2004
- [3] R. Fujiwara et al., Int. J. of Automation Technology, vol. 7, no. 2, pp. 148-155, 2013
- [4] M. Nakano et al., IEEE Trans. Magn., vol. 39, no. 5, pp. 2863-2865, 2003
- [5] M. Nakano et al., IEEE Trans. Magn, vol. 43, no. 6, pp. 2672-2676, (2007)

Magnetocaloric properties of Ni54Fe19Ga23X4 Heusler glass-coated microwires doped by X = B, Al, Ga, In

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One of the favorable candidates for micro-magnetic cooling applications are Heusler alloys which exhibit properties relevant for various fields in magnetism or technological applications. The most remarkable property consists in the modulation of the valence electron concentration ratio (e/a). It may be used to tune the temperature of the martensitic transformation and take this advantage to improve magnetocaloric properties in presented group of alloys. Taylor-Ulitovsky method affords the opportunity of fast and easy preparation of the alloys in the form of glass-coated microwires. The form of the glass-coated microwires brings improvement of mechanical properties for technological applications, well defined easy magnetization axis, scalable size of sample etc. In this work, magnetocaloric properties of Ni₅₄Fe₁₉Ga₂₃X₄ Heusler glass-coated microwires doped by X = B, Al, Ga, In are presented. These alloys exhibit different chemical composition with constant valence electron concentration (e/a). Magnetic measurements have been performed to proof the structural transformation temperature and to analyze the magnetocaloric properties of doped microwires. Results point to the fact that the partial substitution of Ga, while keeping e/a ratio constant, can change the transformation temperature and significantly improve magnetocaloric properties of the glass-coated Heusler microwires.

[1] M. Acet, L. Mañosa and A. Planes, Handbook of Magnetic Materials, Vol. 19, p. 231–289 (2011)

[2] M. Hennel, L. Galdun and R. Varga, JMMM, Vol. 511, a. 166973 (2020)

[3] C. Craciunescu, Y. Kishi and T. Lograsso, Scripta Materialia, Vol. 47(4), p. 285–288 (2002)

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Effect of Al-doping on the structural and magnetic properties of SrFe₁₂O₁₉ and CoFe₂O₄ nanoparticles

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Among nanoparticles-based materials, ferrites with hexagonal ($SrFe_{12}O_{19}$) and spinel ($CoFe_2O_4$) structures play an important role in the quest for new technological applications; thanks to their chemical and thermal stability, their flexible crystal chemistry can be exploited for an accurate optimization of the magnetic properties [1,2]. Over the last few years, these compounds have gained increasing attention due to their potential use for bi-magnetic nanocomposites; within this context, doping is a widely used strategy for the modification of nanoparticles to tune their magnetic activity.

In this work, we have investigated the magnetic and structural properties of aluminum-doped strontium hexaferrite (SrAl_xFe_{12-x}O₁₉) and cobalt ferrite (CoAl_xFe_{2-x}O₄) nanocrystallites obtained by a sol–gel auto-combustion approach [3,4]; this method has provided a simple route to tune the chemical composition of the particles without affecting their basic structure, and with a good control over the size. The as-obtained nanopowders have undergone various annealing treatments (in a range between 500–1200°C) and the phase evolution was inspected through X-ray powder diffraction (XRPD); it also allowed us to determine the size, obtained from the refinement of the XRPD patterns. The composition of the samples was checked with energy dispersive spectrometry (EDS). To assess the performance of the doped nanocrystallites, their static magnetic properties at 300 K were investigated using a superconducting quantum interference device (SQUID) magnetometer and a physical property measurement system (PPMS) from Quantum Design, equipped with a vibrating sample magnetometer (VSM). The metal doping has led to large changes in the magnetization curves; by increasing the amount of Al in SrFe₁₂O₁₉, the saturation moment (M_S) decreases, but the coercive field (H_c) shows a remarkable increase (up to ~1 T). In contrast, Al-doped CoFe₂O₄ exhibits a decrease in both M_S and H_c. In conclusion, the magnetic behavior of the different samples is strongly related to the amount of dopant, depending on the sub-lattice modification caused by the ions' substitution.

[1] R. C. Pullar, Prog. Mater. Sci. 57, (2012) 1191

[2] D. Peddis, C. Cannas, A. Musinu, G. Piccaluga Chem. Eur. J. 15 (2009), 7822

[3] F. Sayed, G. Kotnana, G. Muscas, F. Locardi, A. Comite, G. Varvaro, D. Peddis, G. Barucca, R. Mathieu, and T. Sarkar, Nanoscale Advances, 2, (2020) 851

[4] G. Muscas, P. Anil Kumar, G. Barucca, G. Concas, G. Varvaro, R. Mathieu, D. Peddis, Nanoscale 8, (2016) 2081

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Investigation of the electronic structure and magnetic properties of GD Fe 1-x Mx Si, M = Cr, Mn compounds

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Compounds of the RTX family (R-rare earth metal, T-transition metal, X-p-metal) are actively studied due to their interesting properties. These compounds are used in magnetic cooling of materials based on the magnetocaloric effect. We study the electronic structure and magnetic properties of $GdFe_{1-x}M_xSi$, M = Cr, Mn, x = 0, 0.5, 1. $GdFe_{1-x}M_xSi$ Compounds crystallize into a CeFeSi-type tetragonal structure with a spatial symmetry group P4/nmm (group number 129) with two formula units (six atoms) in the unit cell [1,2].

Calculations of the electronic structure were performed in quantum ESPRESSO computer package [3] using the exchange-correlation potential in the generalized gradient correction (GGA) approximation of the PBE version [4]. The GGA+U method was used, adjusted for the strong electron-electron interaction. For the parameter of the direct Coulomb interaction U in the 4f shell Gd, the value of 6.7 eV was used, as well as the value of the exchange interaction $J_{H} = 0.7 \text{ eV}$. The first-principle approach is based on the use of experimental data on the crystal structure. As a result of calculations, it was found that the more favorable state for GdFeSi, GdFe_{0.5}Mn_{0.5}Si compounds is the ferromagnetic state in the sublattice of transition metals, and for the GdMnSi, GdCrSi, and GdFe_{0.5}Cr_{0.5}Si compounds is the antiferromagnetic ordering in the transition metal sublattice. In calculations for these compounds, the value of the magnetic (spin) moments of Gd ions was 7.6 2. Fe and Si ions have no magnetic moment in the GdFeSi compound, the total magnetic moment per formula unit is 7.6 📴. In the calculations for GdMnSi, the value of the total magnetic moment per GdMnSi formula unit was equal to 7.7 🛽 due to the value of the magnetic moment of the Gd ion. At the same time, on transition metal ions Mn in this compound, the magnetic moment is 3.1 🗈, but due to the antiferromagnetic ordering in the sublattice Mn, it is mutually compensated for two Mn ions in a cell of 2 formula units and does not contribute to the total moment, Si ions do not have a magnetic moment in this compound. In the calculations for GdFe_{0.5}Cr_{0.5}Si, the value of the magnetic (spin) moments of Fe ions was -2.3 2_B, Cr 3 \mathbb{D}_{B} and Si -0.1 \mathbb{D}_{B} , the total magnetic moment per formula unit 8 \mathbb{D}_{B} . In the calculations for GdFe_{0.5}Mn_{0.5}Si, the value of the magnetic (spin) moments of Fe ions was -1.5 DB, Mn -2.7 DB and Si 0.1 DB, the total magnetic moment per formula unit was 5.6 2.6. In the calculations for GdCrSi, the value of the total magnetic moment per GdCrSi formula unit was equal to 7.4 \mathbb{D}_{B} due to the value of the magnetic moment of the Gd ion. At the same time, the transition metal ions Fe and Cr in this compound have a magnetic moment of -2.9 and 2.7 2_B, respectively. Thus, in the GGA+U calculations, a change in the type of magnetic ordering was obtained when iron ions were replaced by manganese ions and chromium ions.

[1] A.V. Morozkin, J. Alloys. Compd. **292**, 162 (1999)

- [2] H.Kido, T.Hoshikawa, J. Less-Common Met. 99, 151 (1984).
- [3] P. Giannozzi, P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, et al. J. Phys.: Condens. Matter. 21, 395502 (2009)
- [4] J. P. Perdew, J. P. Perdew, K. Burke. Phys. Rev. Lett. 77, 3865 (1996)

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4228 RKKY exchange interactions in L10 FePt

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Machine Learning (ML) captured tremendous attention as massive development of computational algorithms have been made due to its applicability in a wide range of applications such as forecast predictions, speech recognition, and others.

In this work, we propose an alternative method to estimate the exchange interactions between atomic sites of L10 FePt based on RKKY function, similarly to an ML algorithm in which a best-fitting function is determined to predict and classify new events based on the training process. This method confers consistent results with the simulations performed using first-principle calculations for the exchange interactions regarding temperature magnetization dependence and finite-size effects. We found an appropriate rescaling factor of magnetization on the temperature around 2.1 previously reported by the experimental measurements in agreement with first-principle calculations.

Radial dependence of circular magnetic permeability of amorphous magnetic microwires

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Magnetic and electrical properties of amorphous magnetic materials account for the active research in this field both for practical applications and fundamental science. The pronounced soft magnetic properties, high resistivity, and associated magnetic bistability and giant magneto-impedance effect made them extremely prospective for electric machines, binary elements, highly sensitive detectors and so on. Complex micromagnetic structure of amorphous microwires and initial stresses lead to nonuniform permeability distribution over the cross-section of the sample. In this work, evaluations of circular permeability values at different points of the cross-section were carried out. According to quazi-static approach in electrodynamics, impedance of cylindrical ferromagnetic wire in MHz frequency range can be obtained using the following expression: $Z=R_{DC}\frac{ka}{2}\frac{J_{0}(ka)}{J_{s}(ka)}$, where R_{DC} – resistance in DC regime, a – wire radius, $k = \frac{1+i}{\delta}$, $\delta = \frac{1}{\sqrt{\pi f \sigma \mu_0 \mu_{\phi}}}$ – skin depth. With increasing frequency of the current the fraction of the sample volume involved in the current flow decreases due to the skin effect. Thus, frequency dependence of circular magnetic permeability can help in understanding of the main peculiarities of permeability distribution over the cross-section. For the experiments, several samples of amorphous microwires of Fe_{6.1}Co₅₇Ni₁₀B_{15.9}Si₁₁ composition with various metal core diameters and glass thickness were chosen. Experimental investigations of the impedance of amorphous magnetic microwires were conducted using vector network analyzer Agilent FieldFox N9923A in the frequency range from 2MHz to 10MHz. External magnetic field along the wire axis was applied using coil. Field magnitude was varied in the range ±100 Oe. The sample of the amorphous microwire with the length of 2cm was soldered to the substrate and then fixed in a measuring cell, which can be included into the coaxial transmission line. Evaluations of the circular permeability were carried out using Matlab software. Model parameters were chosen in accordance with the experimental data. The program choses the value of permeability, in which the absolute value of the impedance coincides with the experimentally obtained one within the margin of manually given error. Thus, the dependence of average magnetic permeability over the skin layer on its thickness was obtained. Values of circular magnetic permeability (10³-10⁴) is at least one order of magnitude larger than the magnetic permeability of the whole wire obtained in static measurements. Splitting the averaging layer - skin depth - into thin sublayers, the distribution of magnetic permeability over the wire cross-section was obtained. We assume, that the change of permeability is attributed to the boundary between axial and surface domains (see figure 1).



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Magnetic properties and mechanical characteristic of Fe-Pt thin sheet magnets prepared using exfoliation behavior

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Since miniaturization of Fe-Pt magnets with excellent biocompatibility is a hopeful material to develop the performance of a small medical devices [1], some researchers prepared isotropic Fe-Pt thick film magnets with the thickness above several microns by a sputtering, electrodeposition and laser ablation technique [2]-[6]. Moreover, isotropic Fe-Pt thin sheets magnets with the thickness range from 10 to 100 microns were prepared using a cold rolling and a rapidly quenching method [7]-[9]. The use of Fe-Pt thin sheets doesn't need to optimize a substrate for each application. However, it is generally hard to improve the magnetic properties by controlling the microstructure compared with those of Fe-Pt films prepared by various deposition methods. For example, J. P. Liu reported a nano-composite Fe-Pt/ α -Fe film with excellent magnetic properties by using a sputtering method [10]. In the study, a laser ablation technique was adopted to obtain a Fe-Pt thin sheet by peeling a Fe-Pt thick film from a Si substrate. As a future work, we pay attention to the preparation of a nano-composite Fe-Pt/ α -Fe multilayered thin sheets because we have demonstrated rare-earth nano-composite films such as Nd-Fe-B/ α -Fe and Sm-Co/ α -Fe using the laser ablation technique [11]-[12].

In the research, Fe-Pt thin sheets thicker than 10 microns with Fe content range from 50 to 60 at. % were prepared. Isotropic Fe-Pt thin sheets could be obtained by taking advantage of the exfoliation behavior after depositing Fe-Pt films on Si substrates using a laser ablation technique. A post annealing process was used to obtain L_{10} phase, and the coercivity of $Fe_{50}Pt_{50}$ thin sheets showed approximately 400 kA/m. Moreover, the test of a cantilever comprising the obtained Fe-Pt thin sheet showed good mechanical characteristic. Resultantly, it was clarified that the use of a thick film preparation process using a laser ablation technique together with exfoliation behavior is a possible approach to prepare a Fe-Pt thin sheet thicker than 10 μ m. Moreover, improvement in magnetic properties of the obtained Fe-Pt sheets are required as a future work.

- [1] A. Yamazaki et al., J. Magn. Magn. Mater. vol. 272-276, pp. e1741-1742, 2004
- [2] H. Aoyama et al., J. Magn. Soc. Jpn. vol. 20, pp. 237-240, 1996. (in Japanese)
- [3] W. F. Liu et al., J. Magn. Magn. Mater. vol. 302, pp. 201-205, 2006
- [4] M. Nakano et al., J. Appl. Phys. vol. 103, no. 7, #07E110, 2008
- [5] M. Nakano et al., J. Appl. Phys. vol. 111, no.7, #07A737 (2012)
- [6] T. Yanai et al., AIP advances, vol. 10, #015149, 2020
- [7] K. Watanabe et al., J. Japan Inst. Metals, vol. 47, no. 8, pp. 699-703, 1983. (in Japanese
- [8] N.H. Hai et al., vol. 262, pp. 353-360, 2003
- [9] T. Bitoh et al., Scripta Materialia, vol. 53, no. 4, pp. 429–434, 2005
- [10] J. P. Liu et al., Appl. Phys. Lett. vol. 72, pp. 483–485, 1998
- [11] H. Fukunaga et al., IEEE Trans. Magn. vol. 50, no. 1, #2101504, 2014
- [12] H. Fukunaga et al., IEEE Trans. Magn. vol. 48, no. 11, pp. 3154 3157, 2012

Ab-initio informed Monte Carlo simulation of rare-earth based alloys

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Advanced magnetic materials have shown tremendous effect on the design of electronic devices over the years, namely DC motor and loudspeaker. These high-performance permanent magnets usually contain substantial amount of rare-earth elements which through alloying with other transition metals can generate optimal magnetic properties for technological functionality. However, the interaction between the rare earth and the transition metal is still not fully understood in many magnetic systems of crucial importance. To gain new knowledge and to support the design of future permanent magnets with enhanced properties we systematically study from first principles temperature-dependent magnetic properties of unary transition metals (Fe, Co, Gd), CaCu₅-type binary alloys $(GdCo_5, GdNi_5)$ and substitutionally doped disordered pseudobinary compounds $(GdCo_{5-x}Ni_x)$. To accomplish this, we perform Monte Carlo [1-2] calculations that are informed through the density functional theory calculation [3-4] of magnetic interactions upon dilation and tetragonal distortion, thus also investigating the role of magnetostructural effects. Our approach provides a very good description of the non-monotonous ferrimagnetic behavior of GdCo₅ [5-6] in very good agreement with experiment, reproducing complex features such as the compensation point (T_{comp}) and the slope of the magnetization against temperature (see figure). We then extend our study by doping at different chemical compositions in GdCo_{5-x}Ni_x to encapsulate theoretical compositions still unexplored. The high critical transition temperature (T_c) is among one of the characteristics of good permanent magnets, whose change from ~30 K to ~1000 K by substitutional doping at transition metal site of GdTM₅ is also reproduced [7]. Our work is compared with other first-principles calculations [6]. Altogether will be used to discuss the underlying electronic mechanisms giving rise to the ferrimagnetic properties found and to explain further the role of the hybridization between the localized Gd 4f-states with the itinerant electrons.



Figure 1 : Magnetization M(T) from experiment (left) [5] and theory (right)

[1] M. Matsumoto and T. Nishimura, ACM Trans. Model. Comput. Simul. 8, 3 (1998)

[2] M. Lezaic, P. Mavropoulos, G. Bihlmayer, and S. Blugel, Phys. Rev. B 88, 134403 (2013)

[3] H. Akai, J. Phys. Condens. Matter 1 8045 (1993)

[4] H. Akai, P. H. Dedrichs, Phys. Rev. B 47 8739 (1993)

[5] A. L. Tedstone, C. E. Patrick, S. Kumar, R. S. Edwards, M. R. Lees, G. Balakrishnan, and J. B. Staunton, Phys. Rev. Mater. 3, 034409 (2019)

[6] C. E. Patrick, S. Kumar, G. Balakrishnan, R. S. Edwards, M. R. Lees, E. Mendive-Tapia, L. Petit, and J. B. Staunton, Phys. Rev. Mater. 1, 024411 (2017)

[7] Y. C. Chuang, C. H. Wu, and Y. C. Chang, J. Less-Common Met. 84, 201 (1982).

Nanocrystalline magnetic powders prepared by a three jet atomization technique

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High performance core materials for applications in electronic and electrical engineering as modern electronic devices and power systems, where transformation efficiency is required with the ultimate goal of energy saving are increasingly needed. Advanced magnetic materials, such as amorphous and nanocrystalline magnetic materials, have been recently used as the most appropriate materials for the fabrication of low core loss magnetic cores, where it is imperative for the material to exhibit an excellent combination of magnetic properties, namely: highest possible value of the saturation magnetization to achieve the highest magnetic flux density amplitude, high magnetic permeability, and low core loss in the working frequency range. Recently the focus is on the amorphous and nanocrystalline magnetic powders since they offer the possibility to fabricate magnetic cores with desired shapes and reduced core size while increasing efficiency for high frequency applications. Various techniques have been employed to produce the powders, but, among them, gas atomization offers the advantage of the preparation of powders with round smooth shape [1]. The aim of our work is to develop nanocrystalline powders of composition Fe_{73.5}Cu₁Nb₃Si_{13.5}B₉ with various using an improved technique of gas atomization. The technique consists in the use of three jets of fluids (two jets of Ar gas and one jet of water), directed in such a configuration so that the molten alloy jet to be successively fragmented in droplets by the two gas jets and further broken intro droplets and cooled by the liquid jet. Thus, we have obtained powders in the nanocrystalline state with the dimensions of up to 30 µm. Scanning Electron Microscopy (SEM), X-ray diffraction (XRD), thermomagnetic, and magnetic measurements have been performed to assess the structural and magnetic differences between the group of samples after sieving. The magnetic measurements have been performed using the Vibrating Sample Magnetometer technique (VSM). The SEM images revealed the formation of particles with almost spherical shape. The structure of particles, with the size of 0-20 μm, 20-32 μm, have been investigated by X-Ray diffraction and it has been noticed that in the range 0-32 µm there is a mixture of amorphous and nanocrystalline particles so that, we can conclude that approx. 50 % of particles are in nanocrystalline state, which is a quite high percentage. The mixture of amorphous and nonocrystalline phases is also confirmed by the slope of the thermomagnetic curves, the Curie temperature increasing from 350°C to 520°C. The saturation magnetization of powders takes values between 130 to 145 emu/g, with slight variations. The coercive field, Hc, has the lowest value of 4 Oe for the powders with the smallest diameter range, the value increasing continuosly up to 23 Oe for the samples with the larger diameter, for the samples in the as-cast state. Thus, we have obtained powders with a mixture of amorphous and high percentage of nanocrystalline phase for sizes of up to 32µm by using an improved atomization technique, which will be used for the fabrication of magnetic cores for high frequency applications. The research will be taken further by finding the optimum annealing conditions to obtain the full nanocrystalline state in the powders with diameters between under 20 µm up to 48 µm.

[1] K. L. Alvarez, M. M. Ipatov, J. Gonzalez, Journal of Alloys and Compounds, Volume 735, p. 2646 (2018)

Effects of the C interstitial doping on the magnetic properties of the LTP MnBi phase

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Experimental and theoretical investigations on the structural and magnetic properties of the MnBi and MnBiC hard magnetic phases are presented. MnBi and MnBiC ingots have been prepared by arc melting under a purified Ar atmosphere and annealed at 400 °C for 12 hours. X-ray diffraction patterns showed that the resulting samples contain a over 74% LTP MnBi phase. In the study, C atoms were assumed to occupy the *2d* interstitial crystal sites of the hexagonal NiAs structure type, as suggested in earlier studies [1, 2]. The presence of C in the formed alloys was investigated by X-ray photoelectron spectroscopy. The theoretical calculations show that C addition enhances slightly the magnetic moment of the samples compared to pristine LTP MnBi, in agreement with our magnetic measurements. Also, our theoretical and experimental studies show that the magnetocrystalline anisotropy energy (MAE) increases by adding C as interstitial dopant. The increase in anisotropy was reflected in magnetic measurements.

P. Kharel, V. R. Shah, R. Skomski, J. E. Shield and D.J. Sellmayer, IEEE Trans. Magn. 49 3318 (2013)
 J. Park, Y.-K. Hong, J. Lee, W. Lee, S.-G. Kim, C.-J. Choi, Metals 4 455 (2014)

Symposium 14. Multifunctional Magnetic Materials

Tunable magnetoresistance and thermopower in interconnected NiCr and CoCr nanowire networks

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Spin-dependent thermoelectric mechanisms are at the heart of the rapidly emerging field of spin caloritronics and are crucial steps in designing next generation heat management devices [1,2,3]. Highly efficient flexible spin caloritronic devices using macroscopic network films made of interconnected FM/Cu multilayered nanowires (FM = Co, NiCo, NiFe) were recently fabricated by direct electrodeposition into 3D nanoporous polymer host with no sample size limitation (see Fig. 1) [4,5]. Large magneto-thermoelectric effects have been observed and the spindependent Seebeck coefficients were directly obtained, being both negative (n-type) in all those systems. However, significant boost in magneto-thermoelectric performance can be expected in FM/Cu systems where the Seebeck coefficients for the spin-up and spin-down electrons have opposite signs. Such sign asymmetry is possible because the sign and magnitude of the diffusion thermopower depend strongly on the energy derivative of the density of states at the Fermi level, and has already been demonstrated in some FM dilute alloys [6,7]. Moreover, practical thermometric device required both p- and n-type elements. Here, interconnected nanowire networks made of diluted CoCr and NiCr alloys (< 7% of Cr) are studied, revealing large modifications in the magneto-transport and thermoelectric properties [8], as shown in Fig. 2. Negative anisotropic magnetoresistance has been observed, highlighting the large effect of Cr on the density of state (reversal of the resistivity spin asymmetry coefficient) [8-9]. Moreover, positive Seebeck coefficients have been obtained in interconnected nanowire networks made of diluted NiCr alloys, opening the door to magnetically tunable p-type thermoelectric devices [8] and, thus, p-n spin caloritronic junctions.



Figure 1: SEM image of a self-supported interconnected NW network film. The inset highlights the NW branched structure.



Figure 2 : (a-b) MR with respect to the Cr content x_{Cr} for interconnected nanowire networks made of diluted **(a)** NiCr and **(b)** CoCr alloys. **(c-d)** Seebeck coefficient S with respect to the Cr content x_{Cr} for interconnected nanowire networks made of diluted **(c)** NiCr and **(d)** CoCr.

- [1] J. He and T.M. Tritt, Science, 357, 6358 (2017)
- [2] G.E.W. Bauer, E. Saitoh and B.J. van Wees, Nature Materials, 11, 391 (2012)
- [3] S.R. Boona, R.C. Myers and J.P. Heremans, Energy Environ. Sci., 7, 885 (2014)
- [4] T. da Câmara Santa Clara Gomes, F. Abreu Araujo and L. Piraux, Science Advances, 5(3), eaav2782 (2019)
- [5] F. Abreu Araujo, T. da Câmara Santa Clara Gomes and L. Piraux, Advanced Electronic Materials 5, 1800819 (2019)
- [6] T. Farrell and D. Greig, Journal of Physics C: Solid State Physics, 3(1), 138 (1970)
- [7] M.C. Cadeville and J. Roussel, Journal of Physics F: Metal Physics, 1(5), 686 (1971)

[8] Tristan da Câmara Santa Clara Gomes, Nicolas Marchal, Flavio Abreu Araujo and Luc Piraux. Applied Physics 8. 9. 10. Letters, 115, 242402 (2019)

[9] I. A. Campbell, A. Fert and O. Jaoul. Journal of Physics C: Solid State Physics, 3(1S), S95 (1970)

Development of Heusler-type magnetocaloric scree-printing inks

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Solid-state refrigeration is a promising alternative to the less efficient conventional vapour compression cycle refrigeration. Particularly, magnetocaloric (MC) cooling is the most elaborated today method of solid state refrigeration. The Heusler type NiCoMnZ (Z=Sn, In, Ga) metamagnetic shape memory alloys (MetaMSMAs), exhibiting a giant inverse MC effect, are commercially promising as the magnetic refrigerants of the active magnetic regenerators (AMRs). In the refrigeration cycle, AMR should be coupled to a heat transfer liquid. Additive manufacturing can provide an obvious solution to control this coupling. In the present work, for the first time, we have developed inks of several MetaMSMAs and used ink-jet printing to obtain the MC films. First, the alloys exhibiting martensitic transformation near room temperature with a low thermal hysteresis were fabricated by the induction method and homogenized. Then, the ingots were used to prepare melt-spun ribbons which, in turn, served for manufacturing a powder by grinding. The material on each stage was heat treated and characterized magnetically and structurally. MC effect was measured directly by the adiabatic method. The powder of MetaMSMA mixed with the specially designed polymer matrix (80:20) was used for ink-jet printing at room temperature which is a great advantage compared to the other additive manufacturing methods involving melting or high temperature treatment. The printed films exhibited an outstanding cyclic inverse MC effect: Δ Tad equal to 2K in the field of 2T. The technology now can be used, e.g., for 2D printing of cooling microdevices for flexible electronics.

Magnetic and structural properties of Ni-substituted magnetoelectric Co₄Nb₂O₉

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Magnetoelectric materials which provide electric (magnetic) field manipulation of magnetization (polarization) have attracted significant attention due to both their interesting fundamental physics and potential applications [1]. Their potential application in data storage, for instance, lies in the feasibility of controlling the magnetic information by applying an electric field [2]. In a class of magnetoelectric materials such as Cr₂O₃, application of a magnetic field induces electric polarization below their magnetic ordering temperature. This induced electric polarization is zero in the absence of a magnetic field and increases linearly with an applied field [3]. The family of $M_4A_2O_9$ (M = Co, Mn and A = Nb, Tb) compounds was initially reported by Fischer et al. [4] to show a similar magnetoelectric coupling under application of a magnetic field below their magnetic ordering temperature. In recent years, several works have been published considering the magnetic, structural, and magnetoelectric features in this series of compounds [5]. Among these compounds, Co₄Nb₂O₉ has been reported to show high magnetoelectric coupling at the vicinity of its Néel transition temperature. It crystallizes in the trigonal P3c1 space group (No. 165) and shows an antiferromagnetic (AFM) phase transition at around 27 K. Different magnetic structures including collinear AFM structure with moments alignment along the c axis [6], collinear structure with moments lying in ab plane, and canting toward the c axis, and more recently an in-plane noncollinear magnetic configuration [7] have been reported for $Co_4Nb_2O_9$ from neutron diffraction analysis. In order to gain more insights into the magnetic structure of this compound, it is helpful to substitute Co²⁺ by magnetic ions. Recently, it was shown that in the Mn^{2+} doped compound, $Co_{4-x}Mn_xNb_2O9$, the noncollinear AFM structure is stable up to x = 3.9, which is due to the strong easyplane anisotropy of Co²⁺. Here, we will present the effects of substitution of Co²⁺ by Ni²⁺ on magnetic properties of Co_{4-x}Ni_xNb₂O₉ by means of neutron diffraction (ND) (Fig. 2), magnetization and heat capacity measurements (Fig. 1), and density functional theory (DFT) calculations. We will show that for x = 1, the compound crystallizes in the P3c1 space group with an in-plane weakly noncollinear AFM configuration, in agreement with that recently reported for $Co_4Nb_2O_9$. We will also show results for the compound with x = 2, that similarly to Ni₄Nb₂O₉, has the crystal structure of the orthorhombic Pbcn space group. We will present neutron powder diffraction results unraveling a weakly noncollinear ferrimagnetic structure with moments lying along the b axis is revealed for this compound.



Figure 1: Heat capacity measurements and calculations for x = 0, x = 1 and x = 3.

- [1] N. A. Spaldin et al, Science 309, 391 (2005)
- [2] W. Eerenstein et al, Nature 442, 759 (2006)
- [3] A. Iyama and T. Kimura, Phys. Rev. B 87, 180408(R) (2013)



Figure 2: Neutron scattering refinements and corresponding magnetic structures obtained for x = 1 and x = 3.

- [4] E. Fischer, et al, Solid State Commun. 10, 1127 (1972)
- [5] A. Maignan et al, Phys. Rev. B 97, 161106(R) (2018)
- [6] E. Bertaut et al, J. Phys. Chem. Solids 21, 234 (1961)
- [7] G. Deng et al, Phys. Rev. B 97, 085154 (2018)

Search for the origin of room-temperature magnetic spirals in YBaCuFeO₅

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The experimental demonstration that *spiral magnets* can induce ferroelectricity has raised great expectations regarding their use in low-power magnetoelectric devices. Unfortunately, low magnetic ordering temperatures ($T_{spiral} < 100$ K) hinder the way towards everyday technological applications. A notable exception is the layered perovskite YBaCuFeO₅ where, by controlling the degree of Cu/Fe chemical disorder, T_{spiral} can be increased up to 310 K. This unusual behavior has recently been interpreted in terms of a local frustration model [1], whose main premise is the existence of Cu/Fe occupational correlations, leading to the formation of ferromagnetic Cu-Fe dimers. However, to date, conclusive evidence for the existence of such dimers in YBaCuFeO₅ remains elusive. Here, we address this problem by using scanning transmission electron microscopy (STEM) and muon-spin relaxation/rotation (μ SR), two local techniques used in combination with advanced density-functional theory (DFT) calculations. The presence of Cu-Fe dimers is clearly supported by the STEM images and further confirmed by the internal magnetic fields as detected at the muon stopping site, both being in good agreement with the results of DFT calculations.

Our findings provide strong support for the proposed "order-by-disorder" mechanism as the origin of spiral order in YBaCuFeO₅, qualifying it as a promising route to stabilize high-temperature magnetic spirals in other materials featuring chemical disorder.



Figure 1 : (a) Phase diagram of YBaCuFeO5 showing the local internal field vs. temperature. The field values were obtained from the muon-spin precession frequencies. (b) Contour map of the normalized FFT spectra, as determined from ZF-μSR data collected over the entire temperature range. Representative FFT spectra are shown in the inset. Note the very broad frequency distribution in the low-temperature spiral phase.

- [1] A. Scaramucci et al., Phys. Rev. X 8, 011005 (2018)
- [2] T. Shang et al., submitted to Phys. Rev. X (2020)

Optimization of the production of R₅(Si_xGe_{1-x})₄ nanoparticles through Nanosecond and Femtosecond Pulsed Laser Ablation in Liquids

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The $R_5(Si_xGe_{1-x})_4$ alloy family, where R stands for a rare earth element, presents characteristics of great scientific interest, such as the Giant Magnetocaloric Effect and Giant Magnetostrictive Effect, among others [1]. In particular, $Gd_5(Si_xGe_{1-x})_4$ and $Tb_5(Si_xGe_{1-x})_4$ have been extensively studied over the recent years, since both their magnetic transition temperature and their capability to produce the aforementioned effects can be tuned by simply changing the Si concentration, which further increases the range of applications for these materials [1].

In general, physical properties do not always translate from the bulk to the nanoscale, as the interaction between the material with itself and its environment changes, as the material's surface/volume ratio increases drastically. Interestingly, when nanostructured, these alloys seem not only to retain their relevant characteristics, but also new ones emerge, such as the Giant Negative Thermal Expansion [2]. It is of great interest to further explore the new effects arising at the nanoscale. The use of the Pulsed Laser Ablation in Liquids technique to produce nanoparticles of these alloys is viewed as an alternative to the high vacuum methods otherwise needed, because the rare earths oxidize rapidly [3]. Experiments show the possibility of creating 7-11nm particles through sequentially ablating the separated reactants, followed by ablation of the colloidal mixture [4]. However, the ideal parameters to produce the nanoparticles are yet to be fully known and understood, which is a key motivation for the present work.

The bulk materials were produced by Arc-Melting furnace, and post-heat treatments, when deemed necessary, prior to polishing in order to be used as ablation targets. The target's properties were characterized by X-Ray Diffraction, Scanning Electron Microscope, and Superconductive Quantum Interference Device analysis.

For the ablations experiments, two lasers systems were used, a nanosecond laser (KrF Excimer laser) and a femtosecond laser amplifier (Femtopower Compact Pro), so as to study the differences in the nanoparticles created in the different laser-matter interaction regimes. The main parameters studied are the pulse energy and duration, different types and volumes of solution, and rotation of the sample during the process. The nanoparticles were characterized using the same techniques as for the bulk samples. The nanoparticles produced with the ns laser were significantly smaller than the ones produced with the fs laser, with mean sizes and standard deviations of 10-14nm and 3.6-6.5nm, and 53nm and 25nm, respectively. The size difference might be due to the fs pulse duration being shorter than the characteristic time of thermal dissipation of the material [5]. Despite their small size, the nanoparticles still present ferromagnetic ordering, but with reduced transition temperatures and with a paramagnetic component.



Figure 1 : $Tb_5Si_{2,2}Ge_{1,8}$ nanoparticles and their distribution (left) and comparison between $Gd_5Si_2Ge_2$ bulk and nanoparticles' M(T) curves (right).

- [1] V. K. Pecharsky et al. Physical Review Letters, Volume 78, Number 23, 9 June 1997
- [2] João H. Belo et al. Phys. Rev. B 100, 134303 Published 7 October 2019
- [3] David José Pereira Coelho, Master Thesis, FCUP, 2018
- [4] Nathalie Tarasenka et al. J. of Nanoscience and Technology 16, 7451, 2016
- [5] E. Akman et al. Photonics Nanostructures Fundam. Appl., vol. 9, no. 3, pp. 276–286, 2011

Oral Presentation

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Magnetic and self-assembling behavior in Au-Fe₃O₄ dumbbell nanoparticles in solution

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Self-assembly of nanoparticles under external stimuli is the bottom-to-top strategy to develop novel responsive nanomaterials. The most known example is self-assembly of single-phase magnetic nanoparticles in applied magnetic field. A number of experimental and theoretical studies were devoted to the self-assembly of isotropic nanoparticles, however there is a limited understanding of magnetic field induced self-assembly of multicomponent and anisotropic magnetic nanostructures.

We investigate magnetic dumbbell nanoparticles (DBNPs) which consist of epitaxially grown magnetic Fe_3O_4 nanoparticles on the surface of spherical Au nanoparticles. Their structural, optical and magnetic properties are useful for variety of applications such as dual probes for drug delivery [1-2], catalysis[3], sensing[4], optics[5] and electronics [6]. However, fundamental understanding of mechanisms and parameters involved in self-assembly of DBNPs in solution is still missing.

We studied several DBNPs coated with a mixture of oleic acid and oleylamine of different sizes: 13nm/14nm, 15% (A13F14), 11nm/8nm, 16% (A11F8), 10nm/14nm, 8% (A10F14), 10nm/12nm, 16% (A10F12), and 9nm/11nm, 15% (A9F11). The numbers indicate the diameters of Au (A) and Fe3O4 (F), respectively, as well as the polydispersity obtained from TEM studies. TEM images show that they are structurally a spherical particle of Au attached to a truncated sphere of Fe₃O₄. We determine blocking temperatures TB> 250K for every DBNP from zero field and field cooled DC magnetization measurements. Field-dependent DC magnetization reveal different shapes of hysteresis loops for our samples, which is presumably due to a complex magnetic composition resulting from the deviations in their local crystal structure. Exchange bias observed in these samples emerges as a consequence of charge transfer from Au, reducing Fe₃O₄ to antiferromagnetic FeO [7-8]. We also found an increasing exchange bias field at 5K as function of the magnetic surface area-to-volume ratio for the DBNPs.

We studied magnetic field-induced self-assembly of DBNPs dispersed in toluene by means of x-ray and neutron scattering measurements. Small-angle x-ray scattering confirms the results of our TEM studies and shows no self-assembly at zero field. Our results of small-angle neutron scattering (SANS) in applied magnetic field show self-assembly of DBNPs in solutions. We observe the onset of order beginning at low fields of 0.2 T for A10F14 and A9F11 and at 1T for A13F14. Surprisingly, we find no self-assembly for A10F12 and A11F8 even at 3T. We also perform a polarized SANS at 1T and 3T to separate nuclear and weak magnetic scattering. Our results clarify the role of Au and Fe3O4 sizes, concentration and strength of applied magnetic field in self-assembly of magnetic DBNPs.

- [1] Angew. Chem .Int. Ed. ,47, 173 -176 (2008)
- [2] Chem. Soc. Rev, 41, 1911-1928 (2012)
- [4] Chem Commun, 36, 4357-4359 (2008)
- [5] Sens. Actuators, B,220,622-626 (2015)
- [6] Appl. Phy. Lett, 99, 011907 (2011)
- [7] Phys. Rev. B, 92, 054416 (2015)
- [8] ACS Nano 7, 857 (2013)

Assemblies of cobalt and maghemite nanocrystals with tunable supercristallinity, morphology and magnetism

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The assembly of inorganic nanocrystals (NCs) in long-range ordered superlattices is a fascinating field of nanomaterials research. These artificial solids exhibit new and enhanced collective properties (magnetic, mechanical, catalytic, etc.) arising from interactions between NCs, making them very attractive for technological applications. For engineering advanced materials, it is crucial to perfectly control the assembly of NCs at microscopic scale. For this purpose, we investigated the elaboration of supercrystals made of dodecanoic acid-coated 7.0 nm-Co NCs (synthesized by micellar approach), with tunable morphology, supercrystallinity and magnetic properties. The supercrystal growth is controlled using the solvent-mediated ligand-ligand interaction strategy. When the colloidal solvent is pure hexane, the evaporation of the solution induces the formation of supercrystalline films of Co NCs [1]. However, when the solution is evaporated in the presence of ethanol, we show, for the first time, the formation of well-facetted fcc colloidal crystals of Co NCs [2]. The change of morphology is explained by a Flory-type solvation theory using Hansen solubility parameters [3]. Colloidal crystals formation results from the appearance of attractive interactions between the NCs due to the presence of ethanol. Theoretical calculations allow to predict the onset of the new type of supercrystal growth in good agreement with the experiment. Thanks to the use of the same batch of Co NCs (same size polydispersity and ligands), an accurate structural (GISAXS) and magnetic (VSM) comparative study have been performed between the two supercrystalline morphologies. A decrease of the NC distance is observed for the fcc colloidal crystals compared to the fcc supercrystalline film. The results are closer to the limit of ideal packing of ligands predicted by the OCM theory [4]. The enhanced magnetic properties of the colloidal crystals compared to the supercrystalline film composed with the same Co NPs, are evidenced, for the first time [2].

- [1]- Lisiecki, I.; Albouy, P.A.; Pileni, M.P., Adv. Mater. 2003, 15, 712–716
- [2]- Costanzo, S.; Ngo, A-T; Russier, V.; Albouy, P.A.; Simon, G.; Colomban, Ph.; Salzemann, C.; Richardi, J.; Lisiecki I. Control of
- the Supercristallinity, Morphology and Magnetism of Cobalt Nanoparticle Assemblies. Sent to editor
- [3]- Goubet, N.; Richardi, J.; Albouy, P.A.; Pileni M.P., J. Phys. Chem. Lett. 2011, 2, 417-422
- [4]- Landman, U.; Luedtke, W. D., Faraday Discuss. 2004, 125, 1–22

Oral Presentation

Organic ferroelectric based composite thin films for spintronic applications

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We report first ever all in-situ Piezoresponse Force Microscopy (PFM) studies in ultra-high vacuum on 10's of nanometer thick films of Croconic acid (CA), an organic ferroelectric, grown on ferromagnetic cobalt films. Croconic Acid, showing a value of polarization (~30 μ C/cm²) that is the highest among organic ferroelectrics and comparable to some inorganic ferroelectric materials [1,2], could be the most suitable candidate for development of a range of multifunctional spintronic devices. Furthermore, fundamentally intriguing properties may arise at ferromagnet/ferroelectric interface, especially when the ferroelectric component is organic in nature.

On one side, organic multiferroics with high polarization values are scarce and on the other side, detailed studies on ferroelectric properties of CA are lacking. We perform detailed studies on the ferroelectric properties of CA films on a ferromagnetic surface with the intent of utilizing the organic nature and high remnant polarization values of CA to realize an organic-inorganic hybrid composite-multiferroic system. To investigate the ferroelectric properties, we switch the ferroelectric polarization of individual grains of CA on cobalt by applying DC bias with the help of a PFM tip and study the switching properties such as coercive field, its dependence on frequency of probing signal, switching current etc. To explore the multiferroic aspect, we have been successful in electrically writing macroscopic areas ($2 \mu m \times 2 \mu m$) of CA film (figure 1) on cobalt following which we will attempt to perform in-situ characterization of the magnetic properties of the cobalt film underneath to study the magnetoelectric nature of the multiferroic. Such a multiferroic system is expected to have unbounded potential for flexible, non-toxic inexpensive and low power consuming technological applications starting from ultra-energy efficient memory to ultra-sensitive magnetoelectric sensors.

S. Horiuchi, Y. Tokunaga, G. Giovannetti, S. Picozzi, H. Itoh, R. Shimano, R. Kumai, and Y. Tokura, Nature 463, 789 (2010)
 S. Horiuchi, K. Kobayashi, R. Kumai, and S. Ishibashi, Nature Communications 8, 14426 (2017)

Tuning the magnetic properties of ferromagnetic micro-scale wires by amorphization control during preparation

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Ferromagnetic microwires combining anisotropic magnetic properties and miniature dimensions are of interest for developing security control systems, coding devices and high performance sensors of low magnetic fields, mechanical stresses, temperatures for applications in microelectronics and bio-medical field [1–3]. To suit particular application, their magnetic, mechanical and structural properties can be tuned by various post-production annealing treatments and by varying the technological regime of preparation. Thus, the directional crystallization by current annealing is used to reach hard magnetic properties required for μ -magnets applications [4]. In this work, the structural and magnetic properties of microwires produced by drawing and quenching technique (known as Taylor-Ulitovsky manufacturing process) are tuned by reducing the drawing speed in the air environment. This makes it possible to control the extent of crystallization.

The wires of three compositions Fe_{77.5}Si_{7.5}B₁₅, Fe₄₅Co₃₀Si₁₀B₁₅ and Co₆₉Fe₄Cr₄Si₁₂B₁₁ were investigated. Depending on the drawing speed and chemical composition, the microstructure of the inner core is composed of the amorphous matrix with embedded nanocrystals. The structural details (crystalline phase, crystal size, shape and orientation) were studied by XRD and transmission electron microscopy analysis.

In the Co-rich microwires, the crystalline clusters were presented even at the maximum drawing speed (9.1 m/s), while the metallic core of Fe-rich microwires remained amorphous up to the smallest drawing speed. The correlation between the microstructure and magnetic properties was investigated by the FORC-analysis (First Order Reversal Curve) via VSM. This method made it possible to determine two positively interacting magnetic phases for Co-rich microwires. FeCo-based microwires demonstrated a more complex behavior revealing the presence of competing magnetic phases of different compositions. Based on the hysteresis loops analysis, the dependencies of the magnetic characteristics (coercive force, remanent magnetization, squareness coefficient and saturation magnetization) on the drawing velocity were deduced. Using the Sixtus-Tonks method, the influence of the crystallization extent on the domain wall mobility for Fe- and FeCo based microwires was investigated. A new model of "magnetic properties vs. structure properties" was developed.

Thus, the result of this work put forward a novel way of tuning the magnetic properties of micro-scale wires by varying the drawing velocity which could be useful for optimizing the wire-based device characteristics.

- [1] A. Uddin, et al, Compos. Part B. 176 (2019) 107190
- [2] C. Morón, et al, Sensors (Switzerland). 15 (2015) 28340–28366
- [3] S. Gudoshnikov, et al, Phys. Status Solidi Appl. Mater. Sci. 211 (2014) 980-985
- [4] K. Hoshino, et al, Lab Chip. 11 (2011) 3449-3457

On the Dzyaloshinsky-Moria interaction in magnetic multiferroics RMn₂O₅ by the neutron scattering

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Manganite oxides family RMn_2O_5 (R – rare-earth element) represents prominent example of a multiferroics with extremely interesting and close relationship between magnetism and ferroelectricity [1-3]. The most significant issue in the studies of RMn_2O_5 is the understanding of the microscopic mechanisms responsible for spin-driven ferroelectricity in these compounds.

The temperature and electric field dpendences of magnetic ordering in single crystals TbMn₂O₅, NdMn₂O₅, Nd_{0.8}Tb_{0.2}Mn₂O₅ were studied using polarized neutron diffraction methods in order to make a new approach to those mechanisms. Chiral scattering was observed in all compounds, that indicates the presence of the antisymmetric interaction in the system.

The magnetic order in TbMn₂O₅ is characterized by several successive phases - high-temperature incommensurate, at $T_{CP} < T < T_N$ ($T_{CP} = 35 \div 37$ K, $T_N = 45$ K), commensurate at $T_{LTIC} < T < T_{CP}$ ($T_{LTIC} = 20 \div 24$ K) and low-temperature incommensurate, at $T < T_{LTIC}$ [4]. The ferroelectric ordering in TbMn₂O₅ can also be represented as successive phases in the same temperature ranges, with the incommensurate magnetic phases corresponding to weak ferroelectric phases, and the commensurate magnetic phase corresponding to a ferroelectric phase with a stronger polarization [2]. In TbMn₂O₅, chiral scattering was detected in all magnetically ordered phases [4].

In contrast to TbMn₂O₅, chiral scattering was observed in multiferroic NdMn₂O₅ only below the temperature $T \sim 20$ K, i.e., below this temperature, the transition from a incommensurate non-chiral magnetic phase to a chiral one occurs [5]. The temperature of this chiral transition corresponds to the temperature of the ferroelectric transition T_{CE} [6], while the value of the ferroelectric polarization is extremely small. It was possible to change the ratio of "right" and "left" spirals in TbMn₂O₅, NdMn₂O₅, Nd_{0.8}Tb_{0.2}Mn₂O₅ crystals by an external electric field. This can serve as additional confirmation of the "antisymmetric" magnetic origin of ferroelectric polarization. Thus our research indicates the presence of an antisymmetric Dzyaloshinsky-Moria interaction in RMn₂O₅, which not only affects the formation of magnetic ordering, but also indirectly can be a source of ferroelectric order in these compounds.

[1] N. Hur, S. Park, P. A. Sharma, J. S. Ahn, S. Guha, S. W. Cheong. Nature 429, 392 (2004)

[2] Y. Noda, H. Kimura, M. Fukunaga, S. Kobayashi, I. Kagomiya, K. Kohn. J. Phys.: Condens. Matte 20, 434206 (2008).
[3] G. R. Blake, L. C. Chapon, P. G. Radaelli, S. Park, N. Hur, S. W. Cheong, J. Rodriguez-Carvajal. Phys. Rev. B 71, 214402 (2005).

[4] I.A. Zobkalo, S.V. Gavrilov, A. Sazonov, V. Hutanu, J. Phys.: Condens. Matter 30, 205804 (2018)

[5] I.A. Zobkalo, A.N. Matveeva, A. Sazonov, S.N. Barilo, S.V. Shiryaev, B. Pedersen, V. Hutanu. Phys. Rev. B 101, 064425 (2020)

[6] S. Chattopadhyay, V. Baledent, et al., Phys. Rev. B 93, 104406 (2016)

The work was supported by the Russian Foundation for Basic Research grant # 19-52-12047, and DFG grant # SA 3688/1-1.

In-silico Thermodynamic description of Heusler compounds applied to magnetocalorics by Monte Carlo simulations starting from *ab-initio*

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Heusler compounds are intermetallic ternary materials which can display a huge variety of physical phenomena, hence being potential: superconductors, gap tunable semiconductors, topological insulators, magnetoelectrics, magnetocalorics, etc [1].

The unguided search for new and better properties is truly a demanding task, especially if done through the careful, time-consuming and often expensive production of all possible compounds. A popular approach to perform this arduous task is to resort to an *in-silico* approach, namely through DFT calculations considering possible elements and respective stoichiometries [1].

DFT is a powerful tool to search for new Heusler compounds and better understanding their inherent However, DFT calculations give a physical description of the studied system at T = 0K, which is particularly handicapping for magnetocaloric materials whose region of interest is located around their Curie temperature (T_c).

A way to simulate the thermodynamic properties of Heuslers from ab initio results, consists in using them as starting point to perform mean field approximations (MFA), or Monte Carlo simulations, such as the popular Metropolis method [2]. However, MFA tends to be conceptionally too simplistic, whilst the Metropolis method performs a too narrow sampling of the partition function and requires a new set of simulations for each new pair of temperature and magnetic fields.

New Monte Carlo methods, based on the sampling of the energy and magnetization joint density of states (JDOS) to calculate the partition function, show quite promising results in the description of magnetic material using a pure *in-silico* methodology [3-4]. Particularly, the Random Path Sampling (RPS) method samples the partition function through random paths of spin flipping which begin and end in full oriented spins [4].

Using the RPS method to determine the JDOS of a L21 system, we present a full in-silico study of several Heusler compounds, namely the ternary Co2CrGa, Co2FeGa and Co2MnGa compounds, and the quaternary Co2CrX1–xGa (Y=Fe and Mn) families. We show that it is possible to predict relevant thermodynamic quantities, such as the temperature and magnetic field dependence of the magnetization, the TC, and the isothermal magnetic entropy change. This is achieved simply by applying statistical mechanics concepts, after determining the partition function of each studied system from a microscopic classical Hamiltonian, such as the Heisenberg Hamiltonian, whose exchange coupling constants were obtained from DFT calculations [5].

[1] S. Sanvito, et al., Science advances, vol. 3, no. 4, p. e1602241, 2017.

- [2] J. Gonçalves, et al. JMMM, vol. 428, pp. 362–367, 2017.
- [3] D. Landau, et al. American Journal of Physics, vol. 72, no. 10, pp. 1294–1302, 2004.
- [4] J. Amaral, et al. IEEE Transactions on Magnetics, vol. 50, no. 11, pp. 1–4, 2014.

[5] C. O. Amorim, et al. "In-silico Thermodynamic description of Heusler compounds applied to Magnetocalorics by Monte Carlo simulations starting from ab-initio." European Journal of Inorganic Chemistry.

Direct measurement and imaging of magnetocaloric effect inhomogeneities at the microscale in Ni₄₄Co₆Mn₃₀Ga₂₀ with infrared thermography

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The most promising research approach in magnetocaloric materials resides at the micro/nanoscale[1], where the quirks of the magnetocaloric effect (MCE) have not yet been unveiled, preventing the proper MCE control and its full exploitation. Localized micro/nanothermal characterization plays a fundamental role in this context[1-2]: techniques such as Scanning Thermal Probe Microscopy (SThM) and infrared thermography are suitable and powerful assets for gaining a better description and understanding of the magnetocaloric scenario.

A class of materials that has been extensively engineered to enhance the magnetocaloric effect (MCE) are the multifunctional ferromagnetic shape memory alloys (FSMAs), of which the Ni-Mn-Ga (NMG) is the archetypal system. Near the martensitic transformation, the MCE of NMG is very large due to big differences in the magnetization values of the phases involved. Doping NMG with Co attributes higher magnetization to austenite than to martensite, unlike what normally happens in the NMG parent phase[3], leading to the so-called inverse MCE. To study the MCE at the microscale with infrared thermography, we investigated a bulk $Ni_{44}Co_6Mn_{30}Ga_{20}$ sample mounted on a Peltier element. Direct measurements of the MCE were performed for temperatures around the reverse and direct martensitic transformation, by applying and removing several consecutive times a ~1.2T magnetic field to the sample and recording the changes in its surface temperature with an IR camera. This was done for different sample temperatures on heating (273K to 327K) and cooling (327K to 267K). On heating, for 285Ksample<301K, where the reverse martensitic transformation of the sample occurs, the inverse MCE is observed, with ΔT between -0.04K and -0.54K, depending on the sample temperature before applying the field (Fig.1a). The inverse MCE displays strong spatial inhomogeneity on the sample surface (Fig. 1b), and the areas where the effect is more pronounced change considerably with the progress of phase transformation. Also, when the inverse MCE appears in the first magnetic field application (as in Fig.1a), a severe reduction in the magnitude of the effect is seen in the subsequent field applications. Still on heating, the martensitic phase (267Ksample<285K) presents direct MCE (ΔT^{0} ,02K), as does the austenitic phase (301Ksample<327K), where 0.05K< ΔT <0.14K. On cooling, the MCE is, for all considered temperatures, direct and similar in magnitude to the direct MCE range measured on heating. Both on heating and cooling, direct MCE reveals spatial homogeneity on the sample's surface, and manifests consistently for every field application. The study reveals substantial novelty in the manifestation of the MCE at the microscopic scale, associated with phase inhomogeneity dynamics, showing a different behavior on heating and cooling of a FSMA around the martensitic transition. A map of the relative contribution of each microscopic area to the effect during the martensitic transformation will be presented.



Figure 1 : a) MCE vs T_{sample} in heating and cooling; **b)** IR camera images over time of Ni₄₄Co₆Mn₃₀Ga₂₀ sample surface after it was heated to the temperature of 292,8K, before and after the magnetic field was applied.

A.M.Tishin,Y.I.Spichkin, et al, Int J Refrig, 68 (2016) 177
 A.M.Tishin,Y.I.Spichkin, Int J Refrig, 37 (2014) 223

[3] Fabbrici, S., et al., Acta Materialia, 59 412-419 (2011)

3877 Phase transitions in Epsilon-Fe₂O₃ nanoparticles

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The epsilon phase of Fe_2O_3 (ϵ -Fe₂O₃) has gained considerable interest due to its intriguing properties and great application potentials. ε -Fe₂O₃ stands out for its huge coercive field (up to 2 T at room temperature, RT), millimeterwave ferromagnetic resonance, remarkable non-linear magneto-optical effect, photocatalytic activity, magnetoelectric coupling [1], and room temperature ferroelectricity [2]. ϵ -Fe₂O₃ presents a complex noncentrosymmetric structure (Pna21) with four distinct Fe sublattices: two in distorted octahedral (Fe1 and Fe2), one in regular octahedral (Fe3r) environments, and one in distorted tetrahedral sites (Fe4t). The spin frustration provides a rich magnetic phase diagram with at least four different magnetic states upon temperature evolution.

This work investigates ε -Fe₂O₃ nanoparticles (~20 nm) combining neutron and X-ray diffraction (in the range 2-900 K), X-ray absorption spectroscopy (XAS), and angle-dispersive X-ray diffraction under pressure up to 33.7 GPa. The four Fe³⁺ magnetic sublattices are ordered in the collinear ferrimagnetic (FIM) phase below T_{N2}≈480 K (FIM2), where the remarkable net magnetization is consequence of uncompensated Fe3r and Fe4t sublattices due to a reduced ordered moment in tetrahedral Fe4t sites. We show that above the super-hard ferrimagnetic phase there is a different ferrimagnetic order (FIM1, soft) where Fe3r and Fe4t moments are disordered, which allows expanding the working range of this multifunctional iron oxide. The soft FIM1 phase displays a much smaller ferromagnetic component and vanishes above 850 K [3].

We present a thorough description of the structural and magnetic evolution below room temperature, with emphasis on the puzzling changes occurring at the commensurate-incommensurate transition (FIM2-ICM). Notable anomalies in the dielectric permittivity and a sheer reduction of the coercivity (from 20 kOe to 0.8 kOe) have been observed between 150 and 100K under cooling [4,5]. We report the subtle magneto-structural effects in the same temperature region. From data analysis of the FIM2-ICM magnetic transition under cooling, two different but related realistic models, which are compatible with the neutron data taken at low-temperatures, are presented. They both imply the formation and presence of magnetic antiphase boundaries in ϵ -Fe₂O₃ nanoparticles. It is of importance that the observation of abrupt changes in the ordered moment at tetrahedral sites at T_{ICM} =100 K. In this presentation, we will show that the spin frustration at the tetrahedral-site (Fe4t) not only gives rise to the unexpected different FIM1 (soft) and FIM2 (super-hard) ferrimagnetic phases, but also it is at the origin of the singular FIM2-to-ICM magnetic phase transition that disrupts the super-hard ferrimagnetic state of Pna'21' magnetic symmetry.

Finally, we report the precise determination of the crystalline structure of the high-pressure phase associated to the volume collapse of ϵ -Fe₂O₃ above 27 GPa [6]. The symmetry changes during the transformation under pressure are fully described. The implications of the discovered high-pressure structure for the understanding of the ferroelectric switching in ϵ -Fe₂O₃ thin films are also discussed.

- [1] C. Kadlec et al., Phys. Rev. B 88, 104301 (2013)
- [4] M. Gich et al. Nanotechnology, 17, 3 (2006)
- [2] M. Gich et al., Adv. Mater. 26, 4645 (2014)
- [5] J Appl Phys 98(4), 044307 (2005) [3] J. L. García-Muñoz et al., Chem. Mater. 29, 9705 (2017) [6] J.A. Sans et al, Nat Commun 9, 4554 (2018)

Control of Magnetism in Porous Iron Oxide Films and Nanostructures via Electric Field Induced Magneto-Ionic Effect

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Ferromagnetic materials are found in a wide range of applications spanning from electromechanical systems, magnetic data storage to biomedical devices. In all the aforementioned cases, ferromagnetism is manipulated by using an external magnetic field, generated via electromagnets, or by using spin-torque effects. In both cases, a significant fraction of the input power is dissipated via Joule heating. Hence, from a power efficiency viewpoint, conventional magnetic actuation procedures are not optimal. An attractive strategy to reduce heat dissipation is the use of an electric field to control the magnetic properties of the materials while taking advantage of magnetoelectric phenomena.

In this framework, the design of the materials, from composition and morphological viewpoints, is of utmost importance, to both provide a precise control of their magnetic properties and facilitate the integration in miniaturized devices. Here, we address these two issues by producing nanoporous thin films and shaping them as micrometric disks of iron oxide. This particular choice of material is made not only based on its earth-abundancy and non-toxicity, but also driven by its versatility, i.e. the existence of multiple iron oxides stable at room temperature, which show different magnetic properties, whose combination can be controlled and tuned to widen the possible responses of magnetoelectric effects. In particular, we fabricated thin films of iron oxide using sol-gel plus dip-coating techniques, for obtaining a film thickness of around 100 nm and a high degree of porosity (Fig. 1(a) and (b)). Moreover, using photo-lithography plus chemical etching, we patterned nanoporous disks of about 10 μm in diameter. The magnetoelectric effects electrically-induced in both nanoporous oxide films and patterned disks are investigated by measuring the magnetic properties while electrolyte-gating. The results reported in Fig. 1(c) show a considerable increase (around 13-fold) of the magnetic moment at saturation after the application of -50 V. Interestingly, using narrower windows of potentials $(\pm 10V)$, the system shows complete cyclability of the effects, as shown in Fig. 1(d). In order to investigate the mechanism involved in the magnetoelectric effect, Raman and Xray photoelectrons spectroscopy were performed, indicating that the magnetoelectric response is mainly driven by the oxygen ion exchange between the iron oxides and the liquid electrolyte.



Figure 1 : (a) SEM image of the surface of the FeO_x and **(b)** of the cross-section fo the film. Room-temperature magnetic hysteresis loops for the in-plane direction of the field for FeO_x thin films as a function of applied voltage showing the **(c)** change in the magnetic properties upon application of an electric field and **(d)** the reversibility of the effect. The loops are normalized such that the saturation magnetization of the initial state is set to 1.

Magnetic properties of Tb₂MnMO₆ (M=Ni or Co): spin rotation in the polar phase of the M=Ni ferrimagnetic perovskite

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The presence of magnetic atoms at the A-site and the coupling between the A and B spin subsystems in ABO₃ like perovskites and their derived structures are cause for a variety of exciting effects. In particular this coupling is attracting the interest due to its important effects in multiferroic and magnetoelectric materials. This work reports a comparative study of the singularly different properties of the isostructural perovskites Tb₂MnMO₆ with M= Ni or Co, having monoclinic distortion ($P2_1/n$) and perfect ordering of the Mn⁴⁺/M²⁺ metals.

We present a description and comprehensive study on four successive magnetic transitions in ferromagnetic (FM) Tb₂MnNiO₆ double perovskite, investigated by magnetometry and neutron diffraction. These transitions are all driven by zone-center modes of the parent paramagnetic phase. In the ground state ($P2_1'$), the moments of magnetic A and B sites order according to different non-polar magnetic modes of the paramagnetic phase. But the coupling between them generates an overall polar symmetry which makes this oxide potentially multiferroic (and therefore ferromagnetic and ferroelectric) in its ground state due to magnetic trilinear coupling. Its macroscopic magnetization is large (5 m_B/f.u) and not related to a weak ferromagnetic component induced by Dzyaloshinskii-Moriya interaction. In addition, a sharp and severe spin reorientation (D_a≈50^o) of the ferromagnetic transitionmetal moments has been observed in the polar phase which opens the door to the magnetic switching of the ferroelectric state in this perovskite, and conversely to the control of the magnetization direction by electrical fields applied parallel to *b*-axis [2]. We also anticipate that in this material the direction of the magnetization (respectively towards c/a could be used as a lever to switch the polar/non-polar (ferroelectric/antiferroelectric) transformation. The above properties were not found in Tb₂MnCoO₆, which displays a single ferromagnetic ordered phase with $P2_1/c'$ symmetry down to 1.5 K. The large single-ion anisotropy term of divalent cobalt anchors the metal FM spins almost perpendicular to the x-y Tb-layers. This configuration generates an internal exchange field that precludes the long-range ordering of Tb moments, the sharp FM spin rotation at the B-sublattices and the non-polar/polar transition observed when M=Ni.

[1] Zhao et al. Nat. Commun. 8, 1 (2017)

[2] Garcia-Muñoz et al., Phys. Rev. B. 99, 184444 (2019), 9705

Multifunctional continuous Gd₅(Si,Ge)₄ thin films deposited by RF-Sputtering

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In 1997, the discovery of the giant magnetocaloric effect (GMCE) at room temperature on Gd5Si2Ge2 compound by Pecharsky and Gschneidner. completely revolutionized the magnetocaloric community demonstrating the potential of magnetic refrigeration at room temperature [1]. Since then, the intensive effort to find optimal magnetocaloric materials, has highlighted the importance of the strong magnetovolume coupling on the enhancement of MCE [2]; however, with disadvantages such as their magnetic/thermal hysteresis, narrow operational temperature ranges, typically high operational magnetic fields (Hc), and low mechanical stability [2].

In order to overcome these challenges several approaches have been followed such as chemical stoichiometry tuning, multi-stimuli (pressure, magnetic field, and temperature) and micro/nano-structuring. The latter has been the most unexplored yet, despite its promising results on reducing Hc, increasing mechanical stability and enhancing operational temperature range, as reviewed recently [3]. Therewith, in order to foster the fundamental knowledge required to optimize these materials micro/nanostructure, it is required to thoroughly study them at reduced size-scales such as micrometric and nanometric particles and thin films. Our group has been dedicated to synthesis optimization and thorough characterization of such size-reduced structures for the past 10 years following top-down strategies [3]. In addition to deepening the influence of microstructure on the MCE, new size-induced effects have arisen too [4]. There are two major challenges associated with the scale-down of Gd5(Si1-x,Gex)4 materials: 1) the strong reactivity of Gd and Oxygen (Gd2O3 phases) and 2) the complex Gd-Si/Ge phase diagram, where the 5:4 phase occupies a narrow composition range in between the wider 1:1 and 5:3 phases.

Alternatively, in this work we have undertaken a bottom-up strategy by depositing Gd5(Si 1-x,Gex)4 thin films by RF-Sputtering technique. Two approaches were followed (Figure 1): 1) simultaneous co-sputtering of Gd, Si and Ge and 2) individual sputtering of Gd, Si and Ge depositing a multi-layered thin film. For both approaches Si/SiO2 (800nm) substrates were used and the thin film total thickness was ~700nm. In order to prevent O diffusion towards the film, 20nm Ta layers were deposited at the film bottom (buffer) and at the top (capping). Five different Gd5(Si1-x,Gex)4 compositions were tested: x=0, x = 0.375, x= 0.45, x=0.5 and x= 1. In this work we will present the major results on the chemical, morphological, structural and magnetic properties of the above-mentioned thin films deposited at room-temperature followed by post-heat treatments and deposited at different temperatures.

Gd





Ge

 Pecharsky V K and Gschneidner K A 1997 PRL 78 4494–7
 Franco V et al 2018 Prog. Mater. Sci. 93 112–232
 Belo J H, et al 2019 J. Mater. Res. 34 134–57
 Belo J H, et al 2019 PRB 100 1–7



Probing local anomalies in SmCrO₃ a Perovskite-like

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Rare-earth orthochromites of the formula RCrO₃ are currently at the center of great controversy regarding ferroelectricity. While dielectric constant anomalies near 400-500 K in the heavier rare-earth chromites were associated with non-centrosymmetry, others claim that the polarization observed in these systems is due to the combined effect of the electric applied field, that breaks the symmetry, and exchange-field on the R ion from the Cr sub-lattice. Accordingly to these claims, no spontaneous ferroelectric polar-order exists in these systems and the presence of a magnetic R-ion is essential to induce a metastable ferroelectric state. Contrarily, the appearance of ferroelectricity without direct correlation to the magnetic order, arising from polar octahedral rotations and/or cation displacements, was recently claimed. [1,2] Clearly, additional efforts are needed to definitely validate these claims. Since these properties might emerge from local structural landscapes that are not well described by long-range average structural methods, the use of local probe studies, such as Perturbed Angular Correlation (PAC) spectroscopy, provide relevant knowledge.

In this work the temperature dependent of the electric field gradient (EFG) on SmCrO₃ compound was followed, using the ¹¹¹Cd PAC probe, in the 16 KFE≈220 K), the magnetic ordering of Cr atoms sub-lattice (T_N^{Cr} =133 K), the spin reorientation (T_{SR} =34 K) and magnetic ordering of Sm atoms sub-lattice (T_N^{Sm} =20 K).

At high temperatures, T>300 K, a frequency triplet corresponding to a single EFG, i.e., one probe local environment, was observed and in this temperature range no significant changes occur in the spectra when the temperature is lowered. However, below 300 K visible changes can be observed in the perturbation function (R(t)) data and in the corresponding Fourier transforms. In detail, a second EFG emerges and its relative abundance increases with decreasing temperature.

The spectra obtained at high temperatures revealed an EFG characterized by a $V_{ZZ}^{Sm1} \approx 76 \text{ V/m}^2$ and an asymmetry parameter n ≈ 0.2 in good agreement with similar systems, while the EFG, that emerges at low temperatures, is characterized by a similar fundamental frequency but a higher asymmetry parameter n ≈ 0.6 . From our data we observed that a distortion of the high temperature local environment start to develop within the paramagnetic phase. Although our data might be compatible with the most recent reports, our results point to a subtler scenario, where locally an inhomogeneous state emerges. In this new state regular and distorted environments (most probably polar and non polar states) coexist.

- [1] https://doi.org/10.1103/PhysRevB.86.214409.
- [2] https://10.1209/0295-5075/107/47012.

Consequences of polar atomistic model P2 for the interpretation of spin waves in MnWO₄

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The attention attracted on multiferroic properties in crystalline materials is due to their obvious potential of applications, eg. for magnetoelectric sensors and data storages, but also due to the principal interest in the underlying atomic-scale interactions, the role of anisotropy and the competition of both. Different explanations have been proposed early to explain the mechanism of magnetoelectric effects [1]. A noncollinear spin configuration could be of important role, as depicted by the theory associated with Aharonov-Casher effect or the inverse Dzyaloshinskii-Moriya interaction [2].

MnWO₄ is an exemplary prototype of magnetoelectric effects, it has been intensely investigated for several years [3]. At zero field, three phases are observed: the commensurate AF1 below $T_{N1} = 8$ K, an incommensurate elliptical spiral spin structure AF2 at $8K \le T \le 12.3K$, and the incommensurate collinear sinusoidal spin structure AF3 below $T_{N3} = 13.5$ K[4]. Recently, on synthetically grown singles crystals we could confirm the true symmetry P2 of the nuclear structure [5] in contrast to the space group believed to be P2/c before. In consequence, the magnetic moments of Mn²⁺ order in AF1 by a non-collinear, spin-canting structure with two different spin-canting textures and respective angles of magnetic moments at Mn_a and Mn_b. Consequently, the magnetic excitation spectra and the exchange couplings in AF1 had to be re-examined as they are very sensitive to the spin configurations. Spin wave calculations based on the non-collinear magnetic structure have been performed [6], confirmed by the agreement to previous experimental spectra [7]. Interestingly, one of the low-lying excitation modes observed in recent neutron scattering study [8] which cannot be described by the collinear model, is properly described in this work. Finally, by a structure being polar, with broken inversion symmetry, the ground state is intrinsically (but weak) ferroelectric and shows an additional degree of freedom for the Mn atoms. Thermal activation leads directly to a helical structure and increased ferroelectricity [9]. In a next step, the excitation spectra in AF2 are re-audited for further understanding.

- [3] e.g. Sagayama H et al Phys. Rev. B 77 220407(R) 2008; Poole A 2009 et al J. Phys.: Conf. Ser. 145 012074; Finger T et al 2010 Phys.Rev. B 81 054430; Gvozdikova M V et al, Phys Rev. B 94, 020406 (2016) and references therein
- [4] Lautenschläger G et al 1993 Phys. Rev. B 48 6087; Nojiri H et al. Phys. Rev. Lett. 106 237202 2011
- [5] Park S .-H. et al 2018 J. Phys. Condens. Matter 30, 135802
- [6] Liu B-Q et al 2018 J. Phys.: Condens. Matter 30 295401
- [7] Ye F et al 2011 Phys. Rev. B 83, 140401 (R)
- [8] Xiao Y et al 2016 Phys. Rev. B 93, 214428
- [9] Park S.-H. et al 2019 Physica B: Condensed Matter, Vol. 551: S. 118-121

 ^[1] Katsura H, Nagaosa N and Balatsky A V 2005 Phys. Rev. Lett. 95 057205; Mostovoy M 2006 Phys. Rev. Lett. 96 067601;
 Harris A B 2007 Phys. Rev. B 76 054447; Arima T 2007 J. Phys. Soc. Japan 76 073702; Sergienko I A and Dagotto E 2006 Phys.
 Rev. B 73 094434

^[2] Kenzelmann M et al 2005 Phys. Rev. Lett. 95 087206; Arima T, Tokunaga A, Goto T, Kimura H, Noda Y and Tokura Y 2006 Phys. Rev. Lett. 96 097202; Aharonov Y and Casher A 1984, Phys. Rev. Lett. 53 319; Sergienko I A and Dagotto E 2006 Phys. Rev. B 73 094434

Numerical modelling and analysis of FORCs of magnetorheological composites

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We present results of numerical simulations of FORCs [1] of magnetorheological elastomers (MRE). First-Order Reversal Curves (FORCs) is effective tool to characterize hysteretical behaviour of material and analyze its magnetic intrinsic interactions.

MRE are well known for their properties to be strong depended on particles distribution in the elastic matrix. In this work we consider systems of "simple patterns" with particles being arranged in different structures, such as chains, clusters of different morphology and grids. We analyze properties, FORCs and switching field distributions (SFD) of such systems and compare them with "bulk" systems simulating real-life samples.

To calculate particle redistribution in the system molecular dynamic approach was used. To calculate particles positions Verlet integration [2] was used. We considered dipole-dipole model for particle-particle interaction and "springs" [3] model for particle-matrix elastic interaction. We also consider effects of magnetic anisotropy and influence of particles size distribution.

In the model "bulk" magnetorheological system consists of up to 100000 particles. We consider systems varying concentration of magnetic filler, size and space distribution of particles, elastic properties of the matrix.

For simulation of the presented model and visualization of the system tool was designed using C++ and Python programming languages.





 J.C. Martínez-García et al. "FORC analysis of ferro-ferromagnetic exchange bias in nanocrystalline ribbons" Physica B 2016
 Verlet, L. "Computer Experiments" on Classical Fluids. I. Thermodynamical Properties of Lennard-Jones Moleculess" Phys. Rev. 1967

[3] Sánchez, P.A. et. al. "Importance of matrix inelastic deformations in the initial response of magnetic elastomers" Soft Matter 2018

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Tunning physical properties of NiFe₂O₄ and NiFe₂O₄@SiO₂ nanoferrites by thermal treatment

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Nowadays, nickel ferrite NiFe₂O₄ nanoparticles (NFO NPs) as part of broad ferrites family are intensively studied due to their versatile technological applications [1-5]. What is more coating of such NPs by non-magnetic SiO₂--matrix by various chemical methods [5-7] developed in recent years is a step forward on the engineered design multifunctional hybrid nanomaterials. In present work we used the NiFe₂O₄ (NFO) and NiFe₂O₄@SiO₂ (NFO@SiO₂) ferrite nanoparticles. The first one was synthesized via co-precipitation method [1] while the second by microemulsion. Subsequently, the as-synthesized NPs were annealed during a one-step process at 1000°C for 6h at the ambient pressure. The analysis of diffraction patterns reveals almost amorphous form of both studied materials, additionally for NFO@SiO₂ dominated by silica matrix. As we have evidenced the applied heat treatment procedure lead to the gradual crystallisation of NPs with evident impurities of Fe₂O₃ in NFO sample and crystallization of SiO₂ matrix for NFO@SiO₂. The performed TEM studies showed interesting behaviour for NFO (Fig.1a), where crystallization process has been induced by applying electron beam leading to formation of about 3.7 nm sized grains. The microstructural analysis of NFO@SiO₂ composite underlines rather irregular NFO as cores within the silica shell than typical core-shell structure (Fig.1b). In this case the average particle size of was estimated as 60.5 nm. We have demonstrated that applied heat treatment lead to noticeable changes within microstructure which are in good agreement with XRD and XPS data. The hysteresis loops acquired for as-synthesized NFO and NFO@SiO2 at 2K and 300K up to 7T (Fig.1a, d) are typical for spinel ferrites. The existence of evident magnetic hysteresis only at 2K may suggest the emergence of superparamagnetic (SPM) state at higher temperatures confirmed by almost zero coercively. The analysis of DC magnetization exhibits FC-ZFC discrepancy with maximum temperature $T_{\rm B}$, which has been estimated as about 32 ± 5K and 12 ± 3K for NFO and NFO@SiO₂, respectively (Table). One may notice drastic reduction in saturation magnetization an remanence for NFO@SiO₂ nanocomposites as compared to pure NFO caused by domination of amorphous silica matrix. The applied heat treatment procedure lead to possible enhancement of magnetic parameters due to observed crystallization process.



[1] A. Bajorek, C. Berger, M. Dulski, et al., J. Phys. Chem. Solids 129 (2019) 1-21 [2] Z. Cvejić, E. Durdić, G. Ivković Ivandekić, B. Bajac, P. Postolache, L. Mitoseriu, J. Alloys. Compd. 649 (2015) 1231-1238 [3] E. Agouriane, A. Essoumhi, A. Razouk, M. Sahlaoui, M. Sajieddine, J. Mater. Environ. Sci. 7 (2016) 4614-4619 [4] A. Sangeetha, K. Vijaya Kumar, G. Nanda Kumar, Advances in Materials Physics and Chemistry 7 (2017) 19-27 [5] A. Chaudhuri, M. Mandal, K. Mandal, J. Alloys. Compd. 487 (2015) 698-702 [6] N. Shukla, A. Ondeck, J. C. Lee, J. B. Mille, Catal. Lett. 142 (2012) 582-587 [7] L. Wang, J. Li, M. Lu, H. Dong, J. Hua, S. Xu, H. Li, J Supercond. Nov. Magn. 28 (2015) 191-196

Figure 1: (a-b) TEM images (c-d) hysteresis loops and (e) magnetic parameters for as-received NFO and NFO@SiO₂ specimens respectively.

Anisotropy-controlled domain structure in modulated nanowires

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One of the promising nanomagnets studied in the last years are cylindrical magnetic nanowires where novel physical effects comprising topologically non-trivial magnetic configurations are involved. They show great potential to be use as domain wall carriers for memory and sensing applications [1,2]. These applications rely on the understanding and control of the magnetic configurations of the individual magnetic units, determined by the composition through the balance between the geometrical shape and magnetocrystalline anisotropy.

In this work we present a nanowire system of alternating CoNi/Ni segments synthesized via electrochemical route. The magnetic configuration was imaged by X-ray Magnetic Circular Dichroism combined with Photoemission Electron Microscopy (XMCD-PEEM) in the demagnetized state and at remanence after magnetizing axially and perpendicularly. On one hand, Ni segments, with cubic crystal symmetry, show an axial magnetic configuration with a small curling component at the surface. On the other hand, CoNi segments, with hexagonal crystal symmetry and a strong magnetocrystalline anisotropy perpendicular to the nanowires, show a single vortex state in the shorter segments and multi-vortex or multi-transverse magnetic configurations in medium and long segments, respectively. A detailed study by micromagnetic simulations reveals that the magnetic configuration is determined mainly by the coupling between soft Ni and harder CoNi segments. For short CoNi segments, Ni segments are magnetostatically coupled and the chirality of the single vortex formed in CoNi remains the same as that of the curling in neighbouring Ni segments. For longer CoNi segments, the remanent state is either multi-vortex or multi-transverse state depending on whether the previously applied field was parallel or perpendicular to the magnetocrystalline axis. The results show the importance of the cylindrical geometry to promote the occurrence of novel magneto-chiral effects and provide key information for the design of cylindrical magnetic nanowires for multiple applications.

[1] A. Fernández-Pacheco et al., Three-Dimensional Nanomagnetism. Nat. Commun. 2017, 8, 15756.
 [2] C. Bran et al., Magnetization Ratchet in Cylindrical Nanowires. ACS Nano 2018, 12 (6), 5932–5939.

$4204 \\ Hidden \ order \ motivated \ anti-Magnetoelectric \ Effect \ in \ Fe_2O_3 \\$

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*The author has chosen not to make public additional content.

Colossal heating efficiency for nearly zero magnetostrictive amorphous microwires via eddy currents at radiofrequency fields

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This work shows that Co-rich microwires (MWs) with nearly zero magnetostriction have colossal heating efficiency under radiofrequency fields.^[1] MWs of 31 μ m diameter and 5 mm length show a specific loss power (SLP) of 3000 W/g at 36 Oe and 625 kHz. The SLP varies with the MWs length and number. The role of length lies on the magnetic domains that change from radial to axial when the length is reduced, as deduced from the hysteresis curve shapes, and makes SLP to decrease by a factor of 10 when the length switch from 5 to 15 mm, see Fig. 1. The number of MWs sets side by side determine the magnetostatic interactions which affect to the remanence (M_r) and susceptibility: Mr = 80% for 2 to 6 MWs and then decreases to Mr = 50% for 10 MWs, suggesting the formation of closure domains that diminish the magnetostatic energy. This change of the hysteresis loop shapes also influences the SLP.

In this work, it is assumed that this colossal heating efficiency is dominated by the eddy-currents, although a small contribution from hysteresis losses cannot be ruled out. The eddy currents are generated by electromagnetic induction and lead to Joule heating of the material. The eddy–currents in Co-rich MWs are enhanced by the magnetization reversal and, thus, the magnetic susceptibility becomes a key factor for the heating efficiency. This magnetic susceptibility is given by the change of the magnetic domain with the length, with the circumferential domains playing probable the main role at high frequency. Therefore, short MWs with nearly zero magnetostriction are key candidates to show colossal heating efficiency.



Figure 1. SLP as a function of frequency for H=36 Oe and as a function of MWs length.

[1] Morales, D Archilla, P de la Presa, et al., Sc. Report 10, 602 (2020).

Correlation between spontaneous electric polarization and electric field gradient in HIF materials from *ab-initio* calculations

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Ferroelectric and multiferroic materials have received great attention both from the technological point of view and fundamental physical properties. This technological application covers a wide variety of subjects, such as nonvolatile memories, large piezoelectric responses for actuators and sensors, and high-density memories to magnetoelectric sensors [1,2]. Hyperfine interactions are important mechanisms to probe the local order of solids. Because hyperfine interactions are susceptible to the atomic order, experimental measurements of the electric field gradient (EFG) are useful in studying phase transitions associated with ferroelectrics. Thus, the relation between electrical polarization and EFG tensors can help in the analysis of experimental results and also provide a better understanding of these transitions.

It has been suggested that the V_{zz} is either proportional to P^2 in places with inversion symmetry in a paraelectric structure, as in NaNO₂ [3] or proportional to P, as in Rochelle salts [4]. The quadratic relation was also found for PbHfO₃ [5], and BaTiO₃ [6]. Theoretically, there is still a great lack of work on this subject in the current literature. Until now, an ab initio study showed a quadratic dependence in proper perovskite ferroelectrics [7].

Recently, perturbed angular correlation (PAC) spectroscopy combined with density functional theory (DFT) calculations were able to investigate how the EFG changes with the ferroelectric phase transitions. However, in HIF materials this relation is not clear because the trilinear coupling requires a large set of atomic distortions. We advance this research by unraveling the relations between the EFG and the spontaneous polarization in the more complex HIF with the Ruddlesden-Popper (RP) crystal and A-site ordered double perovskites (DP) structures. Here, we performed DFT calculations on RP phases $A_3B_2O_7$ and $AA'_2B_2O_7$ and on DP superlattices $AA'B_2O_6$ with A/A' = Ca, Cd, and B = Ti, Mn. We show that quadratic dependency is found on most sites in these systems, but in some cases, this relation goes up to the fourth power.

- [1] I. B. Bersuker, Phys. Rev. Lett. 108, 137202 (2012)
- [2] M. M. Cheong, S., Nature Mater 6, 6-20 (2007)
- [3] D. Dening and P. Casabella, Journal of Magnetic Resonance (1969) 38, 277-282 (1980)
- [4] M. E. Fitzgerald and P. A. Casabella, Phys.Rev. B7, 2193–2195 (1973)
- [5] Y. Yeshurun, et al., Journal of Physics and Chemistry of Solids 40, 231-237 (1979)
- [6] O. Kanert, H. Schulz, and J. Albers, Solid State Communications 91, 465-469 (1994)
- [7] J. N. Gonçalves, et al., Phys. Rev. B 86, 035145 (2012)

Martensite-enabled magnetic flexibility: the effects of post-growth treatments in magnetic-shape-memory Heusler thin films

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Magnetic-shape-memory Heuslers are an important class of ferroic materials, constantly opening new fields of research and application (e.g. magnetic actuation, energy harvesting, ferroic cooling) arising from the strong interplay between magnetism and structure. The exploitation of thin films opens up barely new perspectives in the realization of micro/nanomachines, memories and smart devices, taking advantage also of their possible integration in microsystems. The control of microstructure is a crucial goal for the full exploitation of their functional properties (e.g. shape memory, magneto-mechanical) since the twin variants configuration plays a major role.

Few recent works have been addressed to microstructure engineering, mainly exploiting the epitaxial growth on different substrates and varying film thickness and growth parameters. In previous papers we have shown the correlation between structure, microstructure and magnetic properties at the different legth-scales of substrate-constrained Ni-Mn-Ga films grown on Cr/MgO(100) [1]. More recently, we have studied the effects of size confinement and reported the possible thermo-magnetic actuation of free-standing Ni-Mn-Ga nano-disks, enabled by the peculiar martensitic microstructure [2]

In the present talk we present an important step forward [3]. By means of a thorough multiscale magnetic and structural study, we demonstrate that a variety of simple post-growth treatments, i.e. post-annealing at low T (350°C), magnetic field cooling and mechanical stress, open up the possibility to tailor the twin variant configuration of epitaxial thin films. In particular, depending on the selected treatment, it is possible to tune the type of variant, i.e. X-type with out-of-plane magnetic easy axis or Y type with in-plane magnetic easy-axis, and their geometrical distribution. The mechanisms underlying the overall and local manipulation of microstructure are discussed by taking into account the role of microstructural defects and disorder together with the influence of external fields on the martensitic transformation path.

Our findings definitely provide a platform to easily control and manipulate microstructural and magnetic patterns in magnetic shape memory thin films, paving the way to their full exploitation in smart applications.

In a broader context, this remarkable "magnetic flexibility", enabled by the peculiarity of martensitic microstructure, makes magnetic shape memory Heusler thin films, a unique class, among magnetic materials, for the easy manipulation of their magnetic configuration by simple after-growth treatments.

Oral Presentation

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Magnetic mesoporous silica nanostructures: Investigation of magnetic properties

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Magnetic mesoporous silica nanocomposites give the possibility of generating multi-functional objects for application in different technological areas [1]. This paper focuses on the magnetic properties of nanocomposites constituted by spinel iron oxide nanoparticles (MNPs, \approx 8-9 nm) embedded in a mesoporous silica (M_S) matrix. The mesoporous structure of the silica matrix and the presence of the nanoparticles inside clearly emerge from Transmission Electron Microscopy (TEM) measurements. Low temperature (5K) field dependent magnetization measurements reveal saturation magnetization (M_S) close to bulk value (M_S Bulk =90 emu/g) for both MNPs particles and MNPs/MS nanocomposite, indicating that the presence of silica does not affect magnetic features of the single MNPs[2]. Moreover the dependence of the remanent magnetization on field (i.e. \square m plots) at low temperature has shown a small but evident decrease of interaction in MNPs/MS sample with respect to MNPs samples, which may prevent aggregation phenomena. Finally, a partial orientation of the easy axis is observed when the MNPs are embedded in the silica matrix.

[1] I. B. Bersuker, Phys. Rev. Lett. 108, 137202 (2012).
[1] N. Knežević, J.O. Durand, Chempluschem. 80, 26 (2015)
[2] A. Talone; L. Ruggiero; S.Slimani; P. Imperatori; G. Barucca; A. Sodo; M.A. Ricci; D. Peddis, Magnetic mesoporous silica nanostructures: Investigation of magnetic properties, Nanotechnology, July 2020 (on the status accepted)

Following the martensitic configuration footprints in the martensitic transition route of shape memory Heusler films

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Magnetic shape memory Heuslers have a great potential for being exploited into the next generation of cooling devices and actuating systems due to their "giant" caloric and thermo/magnetomechanical effects, arising from the combination of magnetic order and a martensitic transition. Thermal hysteresis, broad transition range, and twinning stress are among the major obstacles preventing the full exploitation of these materials.

In order to find possible solutions to overcome these unfavorable obstacles, it is necessary to gain a comprehensive view of the configuration of the twin variants at the different length scales and its evolution upon martensitic phase transition. In the literature, there are a few works focused on the crystallographic structures and the martensitic configurations of epitaxial Ni-Mn-Ga thin films through experiments and models [1-4]. However, the knowledge about the multiscale hierarchical self-accommodation of the twin variants in the martensitic phase and its possible links to the transition route is still limited, mainly due to lack of direct multiscale observations.

In the present study, we directly visualize the crystallography of seven-fold modulated (7M) Ni-Mn-Ga epitaxial films, the symmetry relations between the twin boundaries [5], and the interfaces [6] between colonies of different twin boundaries in the martensitic phase by means of different transmission electron microscopy (TEM) techniques. We combine our direct observations through TEM techniques to atomic force microscopy (AFM) topography imaging vs. temperature [7].

We propose a route for the martensitic forward and reverse transitions, highlighting the major role played by the different martensitic interfaces [7]. The present results represent a step forward in the understanding of the transition processes, and pave the way to the possibility of tuning the characteristics of the transition, e.g. hysteresis and transition width, by microstructure engineering aimed at the full exploitation of martensitic Heuslers for applications requiring cyclic phase transition.

- [1] Ranzieri, Paolo et al., Adv. Mater., 2015
- [2] Yang Bo et al., Acta Mater, 2015
- [3] Takhsha Ghahfarokhi, Milad et al., Acta. Mater., 2020
- [4] Schwabe Stefan et al., arXiv preprint arXiv:2004.09768, 2020
- [5] Straka, Ladislav et al., Acta. Mater., 2010
- [6] Muntiferi, Brittany et al., Acta. Mater., 2014
- [7] Takhsha Ghahfarokhi, Milad et al., Materials, 2020

Hybrid magneto-acoustic metamaterials

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Over the past two decades, magnonics has emerged as a new paradigm for wave-based computing and wave physics research, which advocates the use of spin waves as a promising energy efficient signal or data carrier. However, the difficulty of fabricating low-loss nanoscale magnonic devices and metamaterials proves an ongoing issue. We propose a new class of magnetic metamaterials (dubbed magneto-acoustic metamaterials), formed by periodic arrays of magneto-elastic (ME) elements. In such arrays, the propagation of acoustic waves can still be controlled magnetically, while the effect of the ubiquitous magnetic damping is reduced by keeping the magnetic elements small. We begin by calculating the acoustic reflection and transmission from individual ME elements [1]. We find that acoustic waves at frequencies close to that of the Kittel mode exhibit a Fano resonance in reflection. We show that, despite the typically weak magneto-elastic coupling, the effect of coupling can be enhanced by appropriately selecting the wave polarisation and its incidence angle [1]. For arrays of ME elements, we find that the slower acoustic group velocity at frequencies approaching phononic band gaps leads to a noticeable increase in the ME response, making it observable even at realistic damping values. For frequencies inside phononic band gaps, we reveal a magneto-acoustic version of the Borrmann effect in absorption and the magnetically induced transparency in transmission of acoustic waves. The rich and complex behaviour explored here will prove useful when designing magneto-acoustic sensors, actuators and radio frequency modulators.

[1] O. S. Latcham, Y. I. Gusieva, A. V. Shytov, O. Y. Gorobets, and V. V. Kruglyak, Appl. Phys. Lett. 115, 082403 (2019); ibid. 116, 209902 (2020).

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Prospective materials for a novel caloric cooling approach with a multi-stimuli cycle that exploits thermal hysteresis

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The contribution of the refrigeration sector to climate change is frequently overlooked, however, it contributes approximately 12% of total greenhouse gas emissions as a combined impact of the direct emission of fluorocarbons and the electricity production needed to operate devices. The concept of the magnetic refrigeration is one of the possible options to replace the conventional cooling technology based on vapour compression [1]. Magnetic refrigeration is based on the magnetocaloric effect (MCE) which is known to be the largest in the materials experiencing first order phase transitions. A downside of such materials is the thermal hysteresis that reduces the cyclic MCE and, therefore, the efficiency of the cooling cycle [2].

Instead of attempting to increase the cyclic performance of the first-order materials by reducing the thermal hysteresis, in the alternative multi-stimuli cooling cycle the hysteresis is, on the contrary, exploited by using an additional stimulus, such as mechanical stress, to affect the transformed state of the material [3]. In this novel cooling cycle, the application of magnetic field is used to transform the material to the high-magnetization state, which is maintained after the field removal due to the large thermal hysteresis. Then, the second stimulus (uniaxial stress) is applied to transform the material back to the low-magnetization state. Thus, the heat transfer and the de/magnetization of the magnetocaloric material can be decoupled, which eliminates the need of a large amount of permanent magnets that are required for a magnetocaloric cooling cycle.

The requirement for those multicaloric materials is a large sensitivity towards both stimuli, as well as excellent mechanical stability. Most promising materials for this novel cooling cycle are inverse magnetocaloric Heusler alloys (Ni(Co)-Mn-In) that show a tunable thermal hysteresis of the first-order magnetostructural transformation. Furthermore, the all-*d*-metal Heusler alloys exhibit both large magnetocaloric effect and a large volume change during the martensitic transformation. Compared to other magnetocaloric Heusler alloys, Ni-Co-Mn-Ti shows an enhanced mechanical stability [4].

This work represents a systematic study of magnetocaloric and mechanical properties under uniaxial load for the most promising Heusler alloys of Ni-(Co)-Mn-X composition (X: In, Sn, Ti). It comprehensively demonstrates how magnetic field sensitivity of the phase transition, the thermal hysteresis and the mechanical stability can be designed by tuning the stoichiometric composition, modifying the microstructure by different heat treatment protocols or by introducing secondary phases. The sensitivity towards mechanical load as well as stability have been tested by means of constant-stress thermal cycling experiments, while the magnetocaloric response has been investigated in high magnetic fields at fast field-sweep rates [5]. This study gives a comprehensive overview, evaluating principle design rules for well-performing and durable multicaloric materials by comparing different material systems.

[1] O. Gutfleisch et al., Adv. Mater. 23, 821-842 (2011)

[2] F. Scheibel et al., Energy Technol. 6, 1397-1428 (2018)

[3] T. Gottschall et al., Nature Mat. 17, 929–934 (2018)

- [4] Z- Y. Wei et al., Appl. Phys. Lett. 114, 101903 (2019)
- [5] A. Taubel et al., Acta Materialia, under review

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Oral Presentation

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Pauli paramagnetism of the quaternary cubic Heusler alloy CrVTiAl

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Some remarkable magnetic properties have been attributed to CrVTiAl and similar quaternary Heusler compounds with 18 valence electrons. Based on electronic structure calculations they were predicted to be fully-compensated ferrimagnetic semiconductors with moderate exchange splitting. The calculations, based on the ordered LiMgPdSn quaternary structure with four interpenetrating face-centred cubic sublattices at 4*a*, 4*b*, 4*c* and 4*d* sites in space group F<u>4</u>3*m*, predicted a spin gap in both densities of states, and a magnetic ordering temperature T_c of 2233 K (roughly 800 K higher than Co). There are equal and opposite moments on the Cr and V+Ti sublattices of $\approx 3 \mu_B$, and very little magnetism on Al. [1,2]. Stephen *et al* [3] and Venkateswara [4] later described the material as a magnetically-compensated spin-gapless semiconductor and find similarly large calculated sublattice moments. These authors prepared the material in bulk [3,4] and thin film [3] form. On the basis of the different spin gaps in the two spin sub-bands, the compound has been proposed as a potential spin filter that may be useful for spintronics at ambient temperature. Since three of the constituent atoms are Pauli paramagnets (Ti, V and Al) and the fourth, Cr, is a spin density wave antiferromagnet with $T_N = 312$ K and a largely-orbital ordered moment of 0.43 μ_B [5], it is puzzling that the same elements could have such different magnetic properties when incorporated into the ordered cubic Heusler compound.

Here we prepared the CrVTiZ compounds Z = Al or Ga, by arc melting and annealing. We prepared samples with and without 1 at% ⁵⁷Fe doping to check for magnetic ordering by Mössbauer spectroscopy. The magnetization is strictly proportional to applied field, and practically independent of temperature from 4 - 300 K. Unlike a compensated ferrimagnetic half metal, there is no net magnetization at any temperature. The SI susceptibility of the samples lies in the range $2.1 \pm 0.2 \, 10^{-4}$, a value close to the average of the susceptibilities of the four elements namely $1.91 \, 10^{-9} \, \text{m}^3$ /mole or $2.24 \, 10^{-4}$. This leads us to believe that our alloys are Pauli paramagnets, There is no measured change with 1% iron doping, The iron Mössbauer spectrum is slightly broadened paramagnetic singlet with the isomer shift of iron metal. We attribute the broadening to disordered atomic environments of the iron impurities. The density of states can be deduced from the formula

$\chi_{\text{Pauli}} = \mu_0 \mu_B^2 D$

where *D* is the density of states (both spins) in states/J/m³. Converting the units we find we find D = 4.7 states/ev/atom. We note that the residual resistivity reported in refs 3 and 6 is $\approx 200 \ \mu\Omega$ cm, comparable to the minimum metallic conductivity, as expected for a highly disordered alloy.

In conclusion, we have found no evidence to suggest that our samples of CrVTiAl are anything other than normal *d*-band Pauli paramagnets. This might possibly reflect the lack of LiMgPdSn-type order, which would be very difficult to establish experimentally among three adjacent early 3*d* transition elements.

- [1] K. Ozdogan et al, J. Appl. Phys 113 193903 (2013)
- [2] I. Galanakis et al, J. Phys CM 26 379501 (2014)

[4] Y. Venkateswara et al, Phys. Rev. B 97 054407 (2018)

[5] J. M. D. Coey, Magnetism and Magnetic Materials Cambridge University Press 2010

^[3] G. M. Stephen et al, Appl. Phys.Lett 109 242401 (2016); J. Appl Phys 125 123903 (2019); Phys. Rev. B 99 224207 (2019)

Role of La substitution upon magneto-electric coupling of multiferroic BiFeO³

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Multiferroic BiFeO₃ is a potential system for room temperature microelectronics, optoelectronics, spintronics as well as memory storage device applications. But it faces challenges due to chemical kinetics of formation and limitations due to poor quality of phase fraction in presence of parasitic phases like $Bi_{25}FeO_{40}$, $Bi_2Fe_4O_9$ and Bi_2O_3 possibly having severe effects on the applications. However, doping at Bi site provides enhanced phase fraction as well as improved multiferroic properties. In this work we have prepared $Bi_{1-x}La_xFeO_{3-d}$ (x= 0.1, 0.2) using solid state route. The prepared samples show antiferroelectricity and antiferromagnetism with smaller fraction of La (x=0.1) and ferroelectricity and weak ferromagnetism upon increment of La content (x=0.2). The observed features could be explained in the view of distortion around Bi site due to partial replacement of Bi by La as well as structural transition. An enhanced ferroelectricity has been observed in BLFO (x=0.2) than BLFO (x=0.1) understood to be due to magneto-electric coupling. A detailed Mössbauer studies have been carried out to give an atomic level understanding of the enhanced magnetism in BLFO (x=0.2).

Magnetite transformations analysis in Iron Oxide/Cerium Dioxide Reactive Sorbents by first-order reversal curve diagrams

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Present work deals with the magnetite transformations in Iron Oxide/Cerium Dioxide Reactive Sorbents with two different contents of cerium dioxide (5 vol. % - sample S1 and 50 vol. % - sample S2) depending on the annealing temperature (range from 200°C to 800°C).

Well-known co-precipitation method has been used to prepare the ferromagnetic core of the sorbent from cheap and commercially available raw-materials. Subsequently, the cerous nitrate solution was added. Under extensive agitation, the NH₄HCO₃ solution was added in several portions to precipitate insoluble cerous carbonate as a precursor for the cerium oxide preparation. It was assumed that various functional groups and irregularities on the iron(II,III) oxide core served as nucleation centers facilitating the creation of the core-shell composite precursor of the cerous carbonate/iron(II,III)oxide type.

The morphology of samples was studied by TESCAN LYRA 3XMU FEG/SEM scanning electron microscope at accelerating voltage of 20 kV, equipped with an X-Max80 Oxford Instruments detector for energy-dispersive X-ray (EDX) analysis. X'PERT PRO diffractometer (Panalytical) equipped with Co K α radiation (λ = 0.17902 nm) was used for diffractogram measurements in the range of 2ϑ = 20° - 135°, steps = 0.01°, and time/step 5 s. Rietveld structure refinement method using the HighScore Plus program and the ICSD database were applied to analyze the relative volume of phases and their basic parameters: lattice constant and mean micro-domain size.

Room temperature magnetic measurements were performed using the vibrating sample magnetometer EZ9 (Microsense, Massachusetts, USA). This equipment was used for measurements of major hysteresis loops and well known first-order reversal curves (FORC) with the step 8 kA/m between two closest FORC.

Magnetite transformations in dependence on annealing temperature were investigated by comparing X-ray diffraction (XRD) measurements and FORC diagrams. Samples S1 annealed below 600°C exhibit low coercive field $(H_c \sim 0.2 \text{ kA/m})$ and the highest saturation magnetization $(M_s \sim 53 \text{ Am}^2/\text{kg})$. XRD indicates beginning of transformation from magnetite to maghemite. This fact is supported by weak FORC diagrams without the hematite peak. At samples S1 annealed above 500°C H_c increases from 1 kA/m to 27 kA/m and M_s decreases down to 1 Am²/kg. In this case, transformation from magnetite to hematite is almost finished as confirmed by both XRD and FORC techniques.

Different iron oxide transformation is observed for S2 samples with 50 vol. % of CeO₂. In the range of annealing temperatures 200-500°C XRD and FORC detect only the presence of magnetite and magnetic behaviour is similar to S1 samples, but the values of coercive field and saturation magnetization are lower ($H_c \sim 0.06$ kA/m, $M_s \sim 36$ Am²/kg). On the other hand, sample S2 annealed at 600°C exhibits only partial transformation of magnetite to maghemite, what is confirmed by low decrease of M_s (30.3 Am²/kg) and low increase of H_c (0.25 kA/m). The FORC diagrams and XRD of samples S2 annealed above 600°C indicate the presence of pure hematite and its micro-domain size (tens of nm) grows with increasing annealing temperature.

Based on these results, we can pronounce a hypothesis: a higher content of CeO_2 in the sorbents causes that part of their particles enters into the Fe oxide structure and affect the transformation from magnetite to hematite.

Effect of glass shell on magnetization process of amorphous microwires

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The investigation of glass-coated amorphous microwires is of practical and fundamental interest. Natural ferromagnetic resonance, bistable magnetization reversal, low coercivity, and other properties determine the microwire as a promising material for magnetic electronics. Magnetic structure of amorphous microwires consists of domain of "core" (with magnetic moment directed along the axis of wire) and surface domain layer (here, direction of magnetic moment is depended on a sign of magnetostriction). This type of magnetic structure is associated with the combined influence of the inhomogeneous stress tensor and magnetostriction. Herewith, there are differences in the stress tensor of glass-coated and uncoated microwires.

In this work, we studied the effect of a glass shell on the magnetization reversal process of microwires. All microwires under study were measured by pick-up coil magnetometry under tension in situ. Coercivity versus tension stress dependences were analyzed in terms of the interaction of the glass shell and the amorphous core during deformation. In addition, in this work, a theoretical assessment was made of the value of the average stresses in microwires.

In the framework of this work it was found:

- coercivity dependence on the stress is reversible and consists of an initial "linear" and a square root parts the size of which depends on the initial level and distribution of stress in the material;

- a decrease in the initial average stress leads to a decrease in the rate of coercivity increase;

- an increase in the initial average stress leads to an increase in the "linear" region of tension;

It has been assumed that peculiarities of magnetization reversal process of glass-coated microwires are connected with adhesion between glass and amorphous metallic core. The results of this work are important in terms of application of glass-coated microwires as stress-sensible materials.



Figure 1 : a) Coercivity versus tension stress dependence of microwires; b) change in the diameter of glass and amorphous metallic core depending on tension stress to amorphous metallic core.

- [1] Orlova N. N., Aronin A. S., Bozhko S. I., Kabanov Yu. P., and Gornakov V. S., Journal of Applied Physics 111 (2012) 073906
- [2] Orlova N. N., Gornakov V. S., and Aronin A. S., Journal of Applied Physics 121 (2017) 205108
- [3] Aksenov O.I., Abrosimova G.E., Aronin A.S., Orlova N.N. et al., Journal of Applied Physics 122 (2017) 235103.
- [4] Aksenov O.I., Orlova N.N., Aronin A.S., Journal of magnetism and magnetic materials 465 (2020) 165878
- [5] Churyukanova M., Kaloshkin S., Shuvaeva E., Stepashkin A. et al., Journal of Alloys and Compounds 748 (2018) 199

Magnetic and magnetoelectric effects in multiferroic DyMnO₃

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The manganite DyMnO₃ belongs to magnetic multiferroics, the emergence of ferroelectric polarization in the compound is described by the Dzyaloshinskii-Moriya interaction. The studies of DyMnO₃ single crystal were carried out by neutron scattering, including the diffraction of polarized neutrons, the macroscopic measurements were made as well. Temperature evolution of the magnetic structure parameters demonstrates a significant dependence on the measurement mode whether measurements were performed during heating or cooling of the crystal. The probable origin of this is the strong interaction of manganese and rare earth magnetic subsystems. The hysteresis for DyMnO₃ indicates that in the dysprosium compound rare earth magnetic subsystem has a strong influence on the manganese one.

The DyMnO3 magnetic structure at 4 K is a spin cycloid with configuration of magnetic moments of manganese subsystem of type AyAz and for Dy-subsystem–GxAy. Along with the incommensurate structure, below TNR \approx 8 K Dy³⁺ is also ordered in the commensurate collinear magnetic ordering of type GxAy. The determination of the DyMnO3 magnetic structures has been carried out at 12 K, in order to establish the difference in the magnetic structures realized at heating/cooling modes. Only in the critical region near the transition from the spin wave to the cycloid one can see the similarity of the loop. However, at all temperatures, there is some dependence of the chiral scattering on the "electrical" background. The direct magnetoelectric effect consists of changing the electric polarization in a magnetic field. In the case of orthorhombic DyMnO₃ crystals, the spontaneous polarization vector P, that occurs along the *c* direction, can not only be reversed in the opposite direction by an external electric field, but also can be tilted (flop) in the *a* direction by an external magnetic field applied in the *ab* plane.

An electric field equal to 3.7 kV/cm (100 V at 0.27 mm) was applied to the sample. It was cooled in the field to a temperature of 4.2 K. After that, the electric field was removed and a magnetic field was applied. The polarization under such experimental conditions is small, P_a = 150 μ C/m². The polarization disappears at a temperature of T=20K, which is the temperature of transition from a sinusoidal magnetic structure to a cycloidal one.

A completely different picture is obtained when the sample is cooled under the application of electric and magnetic fields. The measurements were made as follows. At a temperature of 100 K, electric and magnetic fields were applied, the sample was cooled to a temperature of 4.2 K. The electric field was removed and the process of measuring of polarization in the sample heating mode was began. The magnetic field was not changed. In magnetic fields of at least 20 kOe in the low temperature range, a sharp increase in polarization is observed $P_a = 2000 \,\mu\text{C/m}^2$. Temperature dependences of magnetization were measured. In the low temperature region, a maximum corresponding to the temperature T_{NR} of the antiferromagnetic ordering of the rare earth subsystem is observed. The magnetization along the *b* axis is approximately 2 times greater than that along the *a* axis, and 6 times greater than the magnetization measured along the *c* axis. These results correlate with the calculation of the magnetic structure.

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Selective Fe substitution by Mn in the high-Tc YBaCuFeO₅ spiral multiferroic

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The low-magnetic ordering temperatures (typically <100 K) critically restrict the potential uses of magnetoelectric multiferroics for spintronics and low-power magnetoelectric devices. YBaCuFeO₅ (YBCFO) displays magnetism-driven ferroelectricity at unexpectedly high temperatures), being one of the best candidates to switchable, magnetism-driven ferroelectricity at zero field and room temperature [1]. The stability range of its spiral phase can be extended beyond room temperature by manipulating the Cu/Fe chemical disorder in the bipyramids and by chemical pressure [2]. Here we have investigated the effects of the partial selective substitution of Fe by Mn in the Fe³⁺O₅ pyramids.

In this work, a series of YBaCu(Fe_{1-x}Mn_x)O₅ samples have been prepared up to x=20%, in which the onset temperature of the spiral ferroelectric phase has been tuned by different routes: either through the cooling rate during the sample preparation or by compositional variations. The dissimilar cationic distribution in the pyramids produced by both routes has been studied by synchrotron and neutron techniques and the local electronic structure of the metals was characterized by soft XAS. From the structural evolution two distinct effects were found that can favor the stabilization of the FE spiral phase. One is the observation that the inter-bowtie separation (between adjacent bipyramidal units) increases at expenses of the height of the bipyramids formed by corner-sharing Cu²⁺O₅ and Fe³⁺O₅ square pyramids. This evolution has a direct impact on the strength of the main competing magnetic couplings, favoring the FM over the AFM terms and increasing frustration. The second effect, deduced from Rietveld refinement of the diffraction data, confirms that (prepared under equivalent conditions) the chemical disorder in the bipyramids increases with the substitution level, a factor that also fosters the non-collinear order. At variance with these two effects, locally, exchange couplings involving Mn are weaker than those involving Fe ions.

We will present the evolution of the spiral phase and its main features (stability, orientation, incommensurability, ...) deduced from magnetometry and neutron data analysis as a function of the Mn content up to x=20%. The temperature evolution of the collinear and non-collinear magnetic phases is also shown for the different compositions. Finally, the influence of the preparation methods will be discussed to the light of the spectroscopic and diffraction results.

[1] M. Morin et al, Nat. Commun. 7, 13758 (2016)[2] Shang et al., Science Adv. 4, eaau6386 (2018)

Dielectric properties of CoFe₂O₄\LiNbO₃ Bilayers

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Nanostructured multiferroic thin films constructed by combining magnetostrictive and piezoelectric materials have attracted recently much scientific and technological interest [1,2]. In addition to possessing ferroelectricity and ferromagnetism in each phase, they are shown to exhibit stress-mediated coupling between their magnetic and electric properties, called the magnetoelectric effect. This coupling between their magnetic and electric degrees of freedom may then give rise to new physical phenomena and applications.

Lithium niobate (LiNbO₃) is a ferroelectric material with good piezoelectric, pyroelectric, electro-optical, birefringent, photorefractive and photoelastic properties, which are favorable towards applications. LiNbO₃ is currently being widely used on electro-optical, surface acoustic wave, piezoelectric sensors and non-linear optical devices. Cobalt ferrite (CoFe₂O₄) is a ferromagnetic material that presents a high magnetocrystalline anisotropy and magnetostriction, making it suitable for application in magnetoelectric composite thin films.

Here, bilayer composite thin films, composed by a LiNbO₃ layer deposited over a CoFe₂O₄ film have been prepared by laser ablation on platinum covered Si(001) substrates. Their structural, microstructural and dielectric properties were characterized. The X-ray diffraction measurements show a rhombohedral ferroelectric phase in LiNbO₃ and a cubic spinel structure in CoFe₂O₄. The SEM micrographs show dense films, with cobalt ferrite and lithium niobate layer thicknesses in the range 100-200nm and 300-800nm, respectively. Their dielectric properties were characterized by impedance spectroscopy as a function of ferrite content. To obtain the relaxation times and activation energies, the electrical permittivity (figure 1) was fitted, using appropriate models for the behavior of the polarization and including a conductivity contribution. As such, the influence of the synthesis conditions and of the ferrite content, on the dielectric properties of the films and their evolution with individual layer thicknesses will be discussed.



Figure 1: Nyquist plot, with the imaginary component of the electrical permittivity as a function of the real component, for a film with a layer of CoFe₂O₄ with a thickness of 152nm and 376nm for the LiNbO₃.

Spaldin N. A., Ramesh R. (2019), Nature Materials, 18, 203–212
 Barbosa, J.G., Gomes, I.T., Pereira, M.R., Moura, C., Mendes, J.A., Almeida, B.G. (2014), J. Appl. Phys., 116, 164112

Effects of V on crystalline structure and magnetic properties of solid solutions based on MnNiSb

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Semigeisler compounds are of interest not only for fundamental science but also for their practical application, due to the presence of such effects as magnetostriction, magnetoresistance, magnetocaloric effect, magnetic fieldinduced shape memory effect. Such materials include MnNiSb. It has 100% spin polarization at the Fermi level. The high Curie temperature, high magnetization, and spin polarization make it an ideal candidate for spintronic devices. On the other hand, the Curie temperature is too high for *MnNiSb* to be used as a magnetic refrigeration material at room temperature and above the range required for thermomagnetic generation. A review of literature sources has shown that the influence of magnetic field, temperature, pressure, and doping on the physical properties of *MnNiSb* remains poorly investigated. The crystalline structure and magnetic properties of the solids $Mn_{1-x}V_xNiSb$ and $MnNi_{1-x}V_xSb$ (0.00 $\leq x \leq 0.20$) are investigated. Solid solutions are produced by the method of solid-phase reactions in vacuum quartz vials in a single-zone resistance furnace with subsequent water quenching. The crystalline structure is studied in $Cu_{\kappa\alpha}$ -radiation at room temperature. The temperature dependencies of specific magnetization and magnetic susceptibility have been studied by ponderomotor method in a magnetic field with induction B = 0.86 Tl. The specific magnetization of the saturation and the parameters of the hysteresis loop of the specific magnetization of the powder samples have been measured by induction method on a vibrating magnetometer in a magnet field of up to 10 Tl. The parameters of the elementary crystalline cell and magnetic properties (magnetic disruption temperature, average magnetic moments, saturation magnetization, etc.) are defined.

Origin of the magnetic anisotropy in focused ion beam transformed magnetic structures

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In the field of logic circuits, one of the biggest issues is down-scaling and power consumption. In the last decades the promising candidate, which can overcome the limitation of the transistor-based approach appear – spin wave circuits [1]. To tackle with the challenge of the down-scaling is necessary to operate in the range of tens of nm. The technical realization is extremely challenging with conventional approaches (e.g. efficient steering of the spin waves requires local directional control of effective magnetic fields). To achieve this directional control, one of the most effective ways is to locally manipulate the magnetic anisotropy direction. A possible approach to the traditionally used lithography technique is the direct writing with employed focused ion beam (FIB) [2-4].

The system of metastable iron undergoes the paramagnetic to ferromagnetic phase transition upon irradiation by the ion beam [5]. We have already shown that we are able to tune magnetic properties by tuning FIB parameters, such as scanning direction, ion dose [4]. One of the most important discovery is the ability to spatially control magnetic anisotropy which can play a key role in e.g. controlling spin wave propagation in magnetic waveguides [6]. So far it has been shown that the anisotropy is connected to the crystallographic orientation of the bcc structure which can appear in four distinct structural domains. However, the connection between the FIB scanning direction and the orientation of crystallographic domains and resulting magnetic anisotropy is not fully understood.

In order to get better insight into the linkage between the structure and magnetism, we employ spatially resolved electron backscattered diffraction (EBSD) combined with micro-Kerr magnetometry to reveal the origin of the magnetic anisotropy. We observed different crystallography behaviour with respect to the energy of impacted ions. With the higher ion energies (30 keV) we recognize body-centered cubic lattice with the (100) orientation. With the lower ion energies (5 keV) we observe body-centered cubic lattice with the (110) orientation what is in agreement with the previous experiment done in-situ in ultra-high vacuum chamber by broad ion beam and observed by low energy electron diffraction (LEED) [5]. Micro-Kerr magnetometry measurements of transformed patterns show that the strong magnetic anisotropy is present only in patterns transformed with 30 keV ions which points more towards the strain-related origin of the anisotropy rather than the (110) structural-domain-orientation-related origin as was the original hypothesis [URBANEK18].

- [1] A. V. Chumak et al, J. Phys. D: Appl. Phys. 50, 244001 (2017)
- [2] M. Nord et al, Small 15, 1904738 (2019)
- [3] P. Mazalski et al, Journal of Magnetism and Magnetic Materials 477, 317 (2019)
- [4] M. Urbanek et al, APL Materials 6, 060701 (2018)
- [5] J. Gloss et al, Appl. Phys. Lett. 103, 262405 (2013)
- [6] L. Flajšman et al, Physical Review B, 101.1: 014436 (2020)

Hyperfine studies on the structural phase transitions of the naturally layered perovskite Ca₂MnO₄

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Naturally layered perovskites (NLP) such as the Ruddlesden-Popper (R.P.) phases (Ca_{n+1}Mn_nO3_{n+1}) have appeared as a fascinating route to design nonexpensive room temperature multiferroic materials. In these NLP, specifically in the Ca₃Mn₂O₇ compound, distortions of the lattice such MnO₆ octahedral rotation and tilting modes couple to polar cation displacement modes inducing a ferroelectric polarization, in a mechanism known as hybrid improper ferroelectricity. The revived interest in this compound lead us to study the homologous elements belonging to the R.P. series. In particular, Ca₂MnO₄ although non polar presents similar MnO₆ octahedral rotations and, just as Ca₃Mn₂O₇, exhibits a peculiar uniaxial negative thermal expansion.

Perturbed Angular Correlation γ - γ (PAC) hyperfine technique offers a unique opportunity to probe at the local scale the structural, charge and magnetic phase transitions of these NLP systems. The measurement of the electric field gradient (EFG), complemented with neutron and x-ray diffraction studies, allows to accurately probe the MnO₆ octahedral rotations that underlie the material's structural phase transitions and functional properties. At ISOLDE-CERN, by using metastable ^{111m}Cd isotopes as radioactive probes, PAC measurements were performed in an extensive temperature range (10-1200K) on the Ca_{n+1}Mn_nO_{3n+1} series. In the Ca₂MnO₄ compound we have measured for the first time the structural transition that occurs at high temperature from *I4/mmm* to the lower temperature I4₁/acd structure. Contrarily to previously results obtained by transmission electron microscopy, in the temperature range of 10-1000K we only saw evidence for a single local environment.

Combined *ab-initio* electronic structure calculations were also performed to understand and show how the measured EFG at the probing sites is sensitive to the distinct MnO₆ octahedral rotation modes.

Tunning antiferromagnetic properties of crystalline 1-D copper(II) polymers via supramolecular functionality exchange and mechanical effects

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In the field of molecular magnetism, often investigated systems are miscellaneous copper compounds because of its spin simplicity S=1/2 and using copper oxides as catalysts. Because of its dynamic nature, metal-organic systems are not even approximately investigated as oxide compounds, especially compounds with pyrazine and pyridine based ligands in which copper is bridged by halogen element. Latterly, it was shown that these compounds can transform energy into mechanical work, that is, they can mechanically respond (through bending, twisting, fracturing, jumping, creeping) to exposure of external force that is giving the structural feature which is in great interest of today's material science.

So far it is known that pyrazine and pyrazine derivates can be mediators of magnetic exchange within dimers, linear chains and two-dimensional lattices, and they are used in the preparation of low-dimensional magnetic materials. However, some insight into functional group effects on the magnetic exchange of dimers, linear chains or two-dimensional lattices in literature is not observed. Further, the first of a few examples of metal-organic coordination compound which response flexibly on the application of sole external mechanical force is very recently observed (2018.). It has been shown that copper(II) acetylacetonate [Cu(acac)₂] crystals exhibit elasticity enough to be reversibly tied into a knot. Lack of metal-organic crystalline systems responsive to external mechanical stimuli leads to the unexplored insight in the mechanical response effect on the magnetic properties.

To understand the magnetic behavior of crystalline coordination compounds and correlate structural features (in particular, functional groups, chemical linkages, bond length and angles) and mechanical responses to magnetic exchange, we prepared a series of 1-D halide coordination polymers of copper(II) with halogen derivatives of pyridine.

For all obtained coordination compounds ($[CuBr_2(3-Clpy)_2]_n$, $[CuBr_2(3-Brpy)_2]_n$ and $[CuBr_2(3-Ipy)_2]_n$) temperature dependence of magnetization M(T) was measured using SQUID magnetometer in the temperature range 2–300 K. Linear dependence between magnetization and magnetic field allows usage of the linear magnetic susceptibility, χ . In accordance with the crystal structure, we applied approach of Bonner–Fischer and modeled entire M(T) curves for all obtained compounds using a spin chain of antiferromagnetically interacting neighbouring Cu²⁺ ions along structural chains where the impact of the counter ion and Cu–X···Cu angles on superexchange interaction J is observed. Magnetic measurements are performed on powder samples, straight and bent crystals. Especially, magnetic measurements for straight and bent crystals are done in a way that the magnetic field was parallel to the direction of the elongation of the crystal and to the direction in which crystals show plastic mechanical response. These results are compared and discussed within a bending influence on the supramolecular structure.

Combined Density Functional Theory and Perturbed Angular Correlation study of SrMnGe₂O₆, SrCoGe₂O₆ and CaMnGe₂O₆

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Multiferroic materials have been under the spotlight due to their fundamental scientific interest and for potential applications in technology. Among these interesting materials are the group of compounds belonging to the Pyroxene family with general chemical formula AM(Si,Ge)₂O₆. More specifically, SrMnGe₂O₆, SrCoGe₂O₆ and CaMnGe₂O₆ are isostructural, crystallizing with monoclinic C2/c symmetry and are characterized by zigzag chains of MnO₆ octahedra linked by edgesharing, separated by GeO₄ tetrahedra chains along the same axis, linked by cornersharing. Due to this arrangement these systems present a rich diversity of low-dimensional magnetic properties. The existence and possible interplay of low dimensionality and magnetic frustration results in multiferroic and/or magnetoelectric properties. Since these properties might arise from local structural features that are not well described by methods based on long-range average structural models, the use of local probe studies is essential. In this context, hyperfine methods, such as perturbed angular correlation (PAC) spectroscopy where the study of the electric field gradient (EFG) in the vicinity of a probe atom, allows reconstructing of the atomic and electronic environment of the probe in the material, helps to clarify the origin of the properties exhibited in these systems. In this work a temperature dependent EFG study will be presented and discussed, guided by EFG simulation results using ab-initio WIEN2k, attempting to clarify the experimental observations in these compounds.

Electrochemical preparation of ferromagnetic shape memory alloy thin films and nanowires

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Shape-memory effect has gained much attention in the past few years because of possibility to employ this phenomenon into commercially applicable devices. Their applicability ranges from bio medicinal drug-delivery agents to nanoscopic sensors and actuators [1].

The downsizing of well-known shape-memory materials opens new challenges to overcome, before using their remarkable features in practical applications. One of the best candidates for such materials are Heusler alloys, mainly because of their versatility in composition and composition-related properties [2]. Ni₂MnGa is the most remarkable shape-memory Heusler alloy, although it's brittleness makes it unsuitable for longer time applications [3]. The most promising counterpart for Ni₂MnGa are the Ni₂FeX-based Heusler alloys, where X = e.g. Ga. They have been reported to show shape-memory effect at wide temperature ranges around room temperature [4]. They are also simple to prepare using several conventional techniques, which include arc-melting in conjunction with melt-spinning or Taylor-Ulitovsky method [4].

To obtain shape-memory alloy nanowires and thin films, different methods have been developed. Several Heusler alloy thin films have been prepared using pulsed laser deposition technique and employing a focused ion beam yields nanowires with several hundred nm in diameter [5–7]. The most promising method to prepare even thinner nanowires is electrochemical deposition [8]. Even though the resulting nanowires depend on several degrees of freedom (concentration of metal ions in the solution, temperature, pH, current density, etc.), it is possible to prepare billions of homogeneous nanowires within several minutes. The only obstacle in the thin films and nanowires electrochemical preparation is to define the optimal deposition conditions.

In this work we present a simple way to prepare thin films and nanowires with Heusler-like Ni₂FeGa composition using electrochemical deposition. Magnetic measurements of the thin films show the presence of a magneto-structural transformation within the magnetization-temperature dependence. We also present Ni₂FeGa – based nanowires which show the magneto-structural transformation during the measurements of magnetic properties. They also exhibit magnetization fluctuations within the isothermal magnetic measurements, making them possible candidates for ferromagnetic shape memory materials.

- [1] E. Y. Panchenko et al., Materials Science and Engineering: A, vol. 746, pp. 448–455, (2019)
- [2] T. Graf, C. Felser, and S. S. P. Parkin, Progress in Solid State Chemistry, vol. 39, pp. 1–50 (2011)
- [3] S. Singh et al., Physical Review B, vol. 93, pp. 1–11, (2016)
- [4] L. Frolova et al., Journal of Alloys and Compounds, vol. 747, (2018)
- [5] N. Patra et al., Journal of Alloys and Compounds, vol. 804, pp. 470–485, (2019)
- [6] D. Qu et al., Journal of Magnetism and Magnetic Materials, vol. 488, pp. 4–7, (2019)
- [7] E. S. Sadki, S. Ooi, and K. Hirata, Applied Physics Letters, vol. 85, pp. 6206–6208, (2004)
- [8] L. Galdun et al., ACS Applied Nano Materials, vol. 1, pp. 7066–7074 (2018)

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Coexistence of ferromagnetism and superconductivity in the Heusler-type Ni₂NbSn ingot and glass-coated microwire

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The coexistence of ferromagnetism and superconductivity was considered as a non-existing phenomenon, due to their antagonistic properties. This coexistence can be found in new materials such as Heusler alloy Ni₂NbSn and may lead us to new possibilities in technical and medical practice. Superconductivity in connection with ferromagnetism in one material brings us to the new feature, magnetic invisibility. [1]

Heusler alloys show several properties, such as shape memory, spin polarization, and so on. [2] In Heusler alloys one can combine different features by changing the chemical composition and also new characteristics were observed, e. g., topological insulators, skyrmions, [3] and ferromagnetic superconductors. Therefore, we decided to study the coexistence of ferromagnetism and superconductivity in Heusler alloy Ni₂NbSn in the form of ingot and rapidly quenched glass-coated microwire. [4]

From magnetic measurements was shown that the ingot is exhibiting superconductive and ferromagnetic properties. In microwire, the ferromagnetism was not observed. However, resistivity measurements confirmed the presence of superconductivity.

The chemical composition is in good agreement with the expected one. X-ray diffraction analysis has proved that the Ni2NbSn ingot crystallizes in the B2 disorder Heusler phase with two minor phases Nb₃Sn and Ni₃Sn₂. In order to remove minor phases, annealing was performed, which has significantly improved superconductive and ferromagnetic properties.

- [1] Gömöry F., Solovyov M., et. al., Science 335, 1466 (2012)
- [2] Graf T., et. al., Progress in solid state chemistry 39, 1–50 (2011)
- [3] Husain S., Sisodia N., et al., Sci Rep 9, 1085 (2019)
- [4] Kanuch P., Ryba T., et. al., Acta Physica Polonica A Vol. 131, 1057–1059 (2017)

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Electric and magnetic properties of YGdFe₂O₆ from *ab-initio* calculations

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Hybrid improper ferroelectric (HIF) materials opened new routes for multiferroics design [1]. In these materials, the transition to a ferroelectric state is driven by a trilinear coupling of the octahedral rotation, tilt, and polar displacement modes. Such rotation and tilt couple to the magnetic moments, offering alternative magneto-electric coupling mechanisms. The A-site ordered double perovskites (DP) structures can present the HIF mechanism, so they are promising candidates for new ferroelectrics and multiferroics.

We performed Density Functional Theory (DFT) calculations on the double perovskite YGdFe₂O₆, built based on the simple perovskites YFeO₃ and GdFeO₃ of the space group Pnma (#62). GdFeO₃ exhibits ferroelectricity and weak ferromagnetism [2].

Our calculations were performed in the G-type antiferromagnetic (ordering of Fe spins) phase of space group $Pmc2_1$ (# 26). In our first results, this system is a semiconductor, as well as YFeO₃ and GdFeO₃, the gap was calculated with so-called LSDA+U formalism [3]. The ground state of YGdFe₂O₆ is ferroelectric, with a calculated spontaneous polarization of 0.5C/cm², which is bigger than that of GdFeO₃ (0.12C/cm²) [2]. In addition, YGdFe₂O₆ is dynamically stable, its phonon dispersion does not contain soft or imaginary modes.

[1] G. Lawes, Physics 4, 18 (2011), ISSN 1943-2879, URL https://doi.org/10.1103/physics.4.18

[2] P. R. Babu, I. Bhaumik, S. Ganesamoorthy, S. Kalainathan, R. Bhatt, A. K. Karnal and P. K. Gupta, J. Alloys Compd., 2015, 631, 232

[3] A. I. Liechtenstein, V. I. Anisimov and J. Zaanen, Phys. Rev. B 52, R5467 (1995)

PEG-coated copper-zinc ferrite nanoparticles: effect of copper and zinc doping on the structural, physicochemical and magnetic properties

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In this paper, PEG-coated copper-zinc ferrite nanoparticles were synthesized by thermal decomposition of organometallic precursors in the presence of PEG (polyethylene glycol) and characterized in detail in terms of their structural and magnetic properties. The obtained nanoparticles were characterized using dynamic light scattering (DLS) and zeta potential measurements, XPS, TEM and AFM techniques. Magnetic properties of the obtained samples were characterized by vibrating sample magnetometer (VSM) and Mössbauer spectroscopy.

Under transmission electronic microscopy, the obtained copper-zinc ferrite nanoparticles were spherical-shaped and monodispersed with a diameter of ~ 5 nm. The NPs-PEG sample showed a spherical shape and core-shell morphology. The average hydrodynamic radius of the NPs-PEG was 21.1 nm. The ξ -potentials of the nanoparticles was +13 mV. No sedimentation was observed in bottles of NPs-PEG even after 6 months of storage at 4ºC at pH 7.0, indicating that the NPs-PEG nanoparticles were highly stable. Owing to the modified synthesis of thermal decomposition in presence PEG, obtained nanoparticles are colloidal stable in water. Chemical structure was characterized by inductively coupled plasma optical emission spectroscopy (ICP-OES), Energy Dispersive X-ray Spectroscopy (EDX) and X-ray Photoelectron Spectroscopy (XPS) coupled with Argon Gas Cluster Ion Beam (Ar-GCIB). Using Ar-GCIB allowed to study subsequent layers of nanoparticles without altering its chemical structure. It revealed the presence of PEG layer on obtained nanoparticles. In addition, thorough magnetic structure study was implied by Vibrating Sample Magnetometer (VSM) and which proved superparamagnetic behaviour of all samples. Moreover, our novel nanoparticles also proved to be promising candidates for magnetic resonance imaging (MRI) as temperature-dependent sensors. Symposium 15. Strongly correlated systems and frustrated and disordered magnetism

Re-entrant quantum criticality in pressurized Yb2Pd2Sn and Yb2Pd2In1-xSnx

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In many intermetallics with Ce and Yb ions, the exchange interaction between conduction electrons and *f* electrons can be tuned by pressure or doping. The ensuing change in the relative weight of the RKKY and the Kondo coupling in the context of crystalline electric field splitting gives then rise to many remarkable phenomena like superconductivity, heavy fermion behavior, intermediate valence phenomena or zero-temperature quantum phase transitions.

In the heavy-fermion system $Yb_2Pd_2In_{1-x}Sn_x$, the interplay of the above indicated mechanisms leads to complex chemical-, pressure-, and magnetic-field phase diagrams which are still not explored in full detail. By using a series of techniques, we show that even modest changes of parameters other than temperature are sufficient to induce multiple quantum-critical transitions in this highly susceptible heavy-fermion family (compare Fig.1). In particular, we show that above 10 kbar hydrostatic pressure not only induces an antiferromagnetic phase at low temperature, but it likely leads to a reorientation of the Yb magnetic moments and/or the competition among different antiferromagnetic configurations. The influence of frustration of the Yb moments due to the peculiar crystal structure of $Yb_2Pd_2In_{1-x}Sn_x$ is considered as well.



Figure 1 : Pressure-temperature phase diagram of the magnetic ordering temperature (filled circles, resistivity; filled hexagons, μ SR) and the low-temperature resistivity maximum (filled squares) of Yb₂Pd₂Sn. The filled area sketches the phase space where long-range magnetic order exists. The diamonds show the pressure-dependent evolution of the residual resistivity ρ_0 . All lines are guides for the eye.

2524 REXS investigation of the magnetic ordering in Mn doped Ca2RuO4

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Magnetic order on the diamond Kagome lattice: ground state and dynamics

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Low dimensional, geometrically frustrated antiferromagnets are at the forefront of condensed matter research. Materials with antiferromagnetic interactions between spins on a triangle lattice inherently exhibit large frustration between similar energy ground states giving rise to new behavior. Recently, experimental and theoretical results indicate that the kagome lattice can host spin liquid state[1]. The kagome lattice is an enticing example of such frustrated lattices; however, various effects, such as out-of-plane couplings, or anisotropy, hinder its highly degenerate spin-liquid state and instead select a single magnetic ground state. It is therefore worthwhile to study nearly-kagome compounds in an attempt to discern what precisely stops formation of the spin-liquid. We successfully synthesised a novel kagome like single crystal of Cu₂OSO₄, whose structure is shown in Fig. 1, and report here its magnetism, since it has strong antiferromagnetic interactions on a diamond-kagome lattice. In particular the compounds orders at 20 K as shown in Fig 2. Very little was previously published and the only experiment performed were always done on powder samples[2]. We studied the bulk properties of Cu₂OSO₄ in order to understand what kind of magnetic order arises on this Kagome-like lattice. We will present thermodynamic measurements, such as specific heat and magnetisation results, as well as neutron and X-ray diffraction data. We will thus present the latest results analyzed on this compound and their interpretation, in order to explain what are the details of the long-range magnetic order arising in this compound, which is a necessary step on the way to a full understanding of the magnetic excitations of the compound, and ultimately of the physics realized in this diamond Kagomé lattice model compound. We will also present inelastic neutron scattering data Fig. 3, and interpret the results using a model that best describes the compounds in a try to find a model describing the dynamics of Cu₂OSO₄.





Figure 1 : Kagome like lattice formed by copper

Figure. 2 : DC susceptibility measured at $H = 5 \ 10^{-3} \ T$ ions (blue and green) in Cu₂OSO₄. on a single crystal along the three relevant crystallographic directions



Figure. 3: Inelastic neutron scattering spectrum of Cu₂OSO₄. Showing magnetic excitations: spin waves at 1.5 K. The mode presented on the right hand side has been tracked in function of temperature.

[1] P. Mendels and A.S. Wills, in Introduction to Frustrated Magnetism, edited by C. Lacroix et al., Springer Series in Solid-State Sciences 164 (Springer Berlin Heidelberg, 2011), pp. 207-238.

[2] E. Flügel-Kahler, Acta Crystallographica 16 1009 (1963).

[3] N. Takahashi, S. Okubo, H. Ohta, T. Sakurai, M. Fujisawa, K. Masashi, J. Phys. Conf. Ser. 400. 2097 (2012).

Magnetic properties and crystal field scheme of Er2B2O7: the case of B = Zr, Ir

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Cubic A₂B₂O₇ oxides, with A standing for a rare-earth element and B for a transition or main block metal, stand in the foreground of scientific interest for their frequently exotic crystallographic and electronic properties. Diverse ground states, magnetic structures, and predicted exciting electronic, magnetic, and even topological properties originating from a competition between electron-electron correlations and spin-orbit coupling have been reported (see Ref. [1], and references therein). Magnetic moments residing on the A/B crystallographic site are subject to geometric frustration in both types of structure, i.e. defect-fluorite structure (Fm-3m, 225) and pyrochlore structure (Fd-3m, 227). The latter structure consists of a net of interpenetrating cations' corner-sharing tetrahedra, while oxygen form 8/6 anions cages around A/B. The rare-earth/transition-metal octahedra in fluorite structure are edge and not vertex sharing, leading to a different exchange pathway type and thus different magnetic Hamiltonian governing ground-state selection. Moreover, the fluorite lattice is magnetically diluted as A and B share the same Wyckoff position.

We report on the physical properties of two erbium Er₂B₂O₇ oxides: Er₂Zr₂O₇ and Er₂Ir₂O₇. Although the compounds crystallize in different crystallographic structures (defect-fluorite and pyrochlore type, respectively), the magnetization and specific heat data reveal remarkably similar properties (magnetic anomalies) for the two compounds. A bifurcation of ZFC and FC magnetization at low temperature, an anomaly in specific heat around 2 K, and frequency dependent 0.8 K anomaly in ac-susceptibility suggest a short-range magnetic ordering of Er moments in both compounds. Moreover, Er₂Ir₂O₇ exhibit (short-range) magnetic ordering also of Ir sublattice demonstrated by all magnetization, ac-susceptibility and specific heat measurements. Investigating the microscopic properties, nevertheless, significant differences are observed due to different underlying mechanisms. The inelastic neutron scattering (INS) experiment allowed an unambiguous determination of the crystal field (CF) eigenenergies and parameters of Er₂Ir₂O₇ [2]. CF acting on Kramers Er ion splits its multiplet to doublets, while the CF scheme is strongly dependent on the cubic-like oxygen cage around it. The influence of the magnetic iridium cations on the erbium CF scheme is subtle despite the strong spin-orbit coupling expected in a 5d metal. Such conclusion can be tentatively drawn comparing the energy spectra of erbium iridate and other 3d- or 4d- metal containing erbium pyrochlores. In contrast, the cubic point symmetry of the Er site in Er₂Zr₂O₇ (defect-fluorite structure) leads to CF splitting of the ground state multiplet into quadruplets and doublets. Due to Er/Zr mixing on single atomic position, significantly broadened INS peaks are observed in the energy spectrum of Er₂Zr₂O₇. The determined CF parameters were used for calculations of specific heat and magnetization CF contributions, resulting in an excellent agreement with experimental data [2]. The present study is, to our knowledge, the first cogent study on the CF scheme of rareearth pyrochlore iridates and heavy rare-earth A₂Zr₂O₇.

[2] K. Vlášková, P. Proschek, M. Diviš, D. Le, R.H. Colman, and M. Klicpera, submitted to Inorg. Chem.

^[1] W. Witczak-Krempa, G. Chen, Y.B. Kim, and L. Balents, Ann. Rev. Conden. Matter Phys. 5, 57-82 (2014).

Structure and magnetic properties of epitaxial CaFe2O4 thin films

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In magnetic systems, the presence of competing interactions between spins can lead to the development of modulated phases that can be commensurate or incommensurate with respect to the crystal lattice [1]. An interesting case of these type of systems is shown by the antiferromagnetic oxide $CaFe_2O_4$, in which two differently modulated spin arrangements, termed A and B, are found to coexist below 150 K [2,3]. In both structures the Fe^{3+} spins align parallel to the b-axis, giving rise to an Ising-like system with large magnetocrystalline anisotropy. However, A and B differ in the b-axis stacking of such chains: with A having an up-up-down-down configuration; while the periodicity is halved in the up-down-up-down B phase. The relative stability of these magnetic phases is ruled by the competing exchange interactions between Fe^{3+} spins arranged in two distinct crystal environments. In particular, the weak magnetic exchange between Fe^{3+} spins connected through edge-sharing oxygen octahedral varies with temperature, leading to ferromagnetic coupling in the A phase and antiferromagnetic coupling in the B phase. Interestingly, the local structure of the antiphase boundary between two domains of one phase corresponds to a unit cell of the other phase, and the presence of pinned "orphan spins" at the A/B phase boundaries generates an uncompensated magnetic moment along the b-axis.

In this work [4], we have grown for the first time epitaxial $CaFe_2O_4$ thin films on TiO_2 (110) via pulsed laser deposition (PLD). Characterization of our samples via X-ray diffraction (XRD) and transmission electron microscopy (TEM) reveals the coexistence of two out-of-plane crystal orientations and the formation of three in-plane oriented domains, that are clearly visible in atomic force microscopy (AFM) images and confirmed by X-Ray pole figures and electron backscattered diffraction (EBSD).

The magnetic properties of CaFe₂O₄ thin films have been investigated macroscopically, with SQUID magnetometry and Mössbauer spectroscopy. All measurements reveal long-range ordering at below 185 K and a non-zero in-plane magnetization. The largest magnetic hysteresis is found at 130 K, where the coexistence of A and B phases is expected to be maximum. Moreover, a vertical shift of the M-H loops depending on the field-cooling conditions, evidence that this is not standard ferrimagnetic behaviour. Finally, the well defined microstructure of the films allows us to perform local magnetic characterization, yet unreported in this material, by means of magnetiz force microscopy (MFM) and scanning SQUID microscopy. Here, the observation of a spontaneous in-plane magnetization is also in good agreement with an antiferromagnet with orphan spins arising from the coexistence of differently modulated A and B phases, as proposed in bulk samples [3].

- [1] Jensen, M. Bak, P. Spatial Chaos. Phys. Scripta T9, 64–69 (1985).
- [2] Stock, C. et al. Solitary Magnons in the S=5/2 Antiferromagnet CaFe2O4. Phys. Rev. Lett. 117, 1–6
- [3] Stock, C. et al. Orphan Spins in the S=5/2 Antiferromagnet CaFe2O4. Phys. Rev. Lett. 119, 1–6,
- [4] Damerio, S. et al. Structure and magnetic properties of epitaxial CaFe2O4 thin films, arXiv:2001.11446v1 (2020)

Orbital ordering and magnetoelectric coupling in FeCr2O4 Ferrimagnet

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The FeCr₂O₄ compound has attracted heightened attention because of the discovery of a quite pronounced magnetoelectric effect in its ferrimagnetic spin ordered state, which coexists with orbital ordering states of Fe²⁺ [1-2]. The mechanisms of formation of magnetoelectric coupling as well as orbital ordering are not clear. The orbital ordering arises at 135–141 K [2-3]. The electric polarization in non-collinear state arises below ~100 K.

We applied enhanced technic on the calculation of Fe²⁺ crystal field parameters and Jahn-Teller coupling constant utilizing electrostatic Kleiner's correction [4] along with the well-known point charge and the exchange charge models. It takes into account the penetration effect of charge densities and allows to get better estimations of electron-phonon and magnetoelectric coupling parameters.

The energy of the cooperative orbital ordering of the states of iron ions was calculated, taking into account both the interaction through the strain field and the electrostatic interaction of quadrupole moments. We have found that the interaction through the elastic field dominates the quadrupole–quadrupole interaction. The ferro-type orbital ordering has turned out to be the most favorable in agreement with experimental data [2-3]. The estimated critical temperature ~180K is a bit higher than measured one.

The calculation of the magnetoelectric coupling was conducted in two stages. The first stage considered a mixture of electronic configurations of opposite parity ($3d^6$ and $3d^54pL$, $3d^7L^{-1}$) by the strong odd ligand field acting on Fe²⁺. The symbol L stands for electronic configurations of the nearest oxygen ions located at the vertices of the tetrahedron. An effective operator of interaction between orbital moments of 3d electrons and an external electric field H_E was derived in [5]. As the main part of the Hamiltonian was an even ligand field (including covalent bonding effects) and naturally the electric-dipole operator. At the second stage, using the operator's perturbation theory technique with spin-orbit coupling, exchange interaction between iron and chromium ions, the effective Hamiltonian of spin and the electric field coupling was obtained. The final expression for the magnetoelectric bond in crystallographic axes is written as follows

 $\mathsf{H}_{\mathsf{ME}} = \mathsf{A}(\mathbf{SE})\mathsf{U}_{\mathsf{a}} + \mathsf{B} \sum \mathsf{J}_{\mathsf{FeCr}} \{ [\mathbf{S}_{\mathsf{Fe}} \times \mathbf{S}_{\mathsf{Cr}}]_{\mathsf{x}} \mathsf{E}_{\mathsf{x}} - [\mathbf{S}_{\mathsf{Fe}} \times \mathbf{S}_{\mathsf{Cr}}]_{\mathsf{y}} \mathsf{E}_{\mathsf{y}} \} + \mathsf{C} (\mathsf{S}_{\mathsf{x}} \mathsf{S}_{\mathsf{y}} + \mathsf{S}_{\mathsf{y}} \mathsf{S}_{\mathsf{x}}) \mathsf{E}_{\mathsf{z}},$

where $U_a = i (|\theta\rangle \langle \epsilon| - |\epsilon\rangle \langle \theta|) - Pauli like operator in the basis of orbital Fe(⁵E) states.$

Developed microscopic theory provides an analytical expression for coefficients A, B, C and allows us to estimate the value of electric polarization and its direction relative to crystallographic axes of a crystal. Magnetic anisotropy of FeCr₂O₄ crystal is explained by the joint action of linear Jahn-Teller coupling and spin-orbital interaction. Some of our earlier results are published in [6,7]. The orientation of chromium and iron spins is discussed in the context of data on the Mossbauer iron spectrum [8].

- [1] K. Singh, A. Maignan, Ch. Simon, Ch. Martin, Appl. Phys. Lett. 99, 172903 (2011)
- [2] K. Tsuda et al., Phys. Rev. B 81, 180102(R) (2010)
- [3] Q. Zhang et al., Phys. Rev. B 85, 054405 (2012)
- [4] W. H. Kleiner J. Chem. Phys. 20, 1784 (1952)
- [5] M. V. Eremin, Phys. Rev. B. 100, 140404(R) (2019)
- [6] K. V. Vasin, and M. V. Eremin, JETP 2019, 129, 1029 (2019)
- [7] M. V. Eremin, JETP Letters, 109, 249 (2019)
- [8] Sh. Nakamura and A. Fuwa, Phys. Proc. 75, 747 (2015)

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Spin-lattice coupling and quadrupolar effects in the quantum spin ice candidate Tb2Ti2O7 revealed by THz spectroscopy

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In geometrically frustrated magnetism, the very nature of the ground state of $Tb_2Ti_2O_7$, has remained a long standing conundrum. In this pyrochlore material, no conventional spin-ice or long-range magnetic order is stabilized, even at very low temperatures [1]. Quantum fluctuations are suspected of being at the origin of such an exotic quantum phase, yet so far has lacked conclusive evidence.

Using neutrons scattering and high-resolution synchrotron-based terahertz spectroscopy, it has been shown that a double hybridization of crystal-field-phonon modes is present across a broad temperature range from 200 K down to 6 K [2]. This so called vibronic process affects the electronic ground state that can no longer be described solely by electronic wave functions and involve quadrupolar degrees of freedom associated with the rare earth magnetic element in its local environment. Moreover, recent measurements shown that the phase diagram of $Tb_{2+x}Ti_{2-x}O_{7+y}$ is extremely sensitive to off stoichiometry and a quadrupolar order is stabilized for x > 0 instead of the spin liquid state [3].

We present here new results of THz spectroscopy of two $Tb_{2+x}Ti_{2-x}O_{7+y}$ samples, one in the spin liquid state and the second in the quadrupolar ordered phase. The THz measurements are extended to lower temperatures, down to 270 mK, deep into the exotic phase and also under static magnetic field up to 15 T. These data will be confronted to calculations based on a spin-lattice hamiltonian and an electromagnetic approach. Emphasis on the importance of the quadrupolar effects will be made in the unexplained spin liquid behavior of this compound.

- [1] J. S. Gardner et al., PRB 64, 224416 (2001)
- [2] E. Constable et al., PRB 95, 020415(R) (2017)
- [3] T. Taniguchi et al., PRB 87, 060408(R) (2013)

Oral Presentation

Quantum fragmentation in the magnetic frustrated compound family of Nd2Zr2O7

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In pyrochlore compounds, where the magnetic atoms are located on the vertices of a network of corner-sharing tetrahedra, the combination of strong Ising anisotropy along the local <111> axes with ferromagnetic interactions leads to a local organizing principle, the so-called "ice-rule": two spins point in and two spins point out in each tetrahedron. The resulting spin ice state is called a Coulomb phase because the constraint can be interpreted as a divergence free condition of an emergent gauge field. Magnetic excitations above this spin ice ground state can be viewed as magnetic charges, called magnetic monopoles. When the density of the excitations is large enough, they organize as a crystal of alternating magnetic charges resulting in the fragmentation of the magnetic moment into two parts [1]: one associated to the charge order which results in an antiferromagnetic "all-in-all-out" (AIAO) order, and the other which corresponds to a disordered Coulomb phase and is characterized by a spin ice diffuse scattering.

In the Nd2Zr2O7 pyrochlore, a positive Curie temperature indicates the existence of ferromagnetic interactions. Together with the Ising nature of the Nd3+ ion, a spin ice ground state should be stabilized. Experimentally, however, a partial AIAO antiferromagnetic ordering is observed [2]. Furthermore, compared to the scenario described in [1], the spin ice signatures are transferred in the excitation spectrum, taking the form of the flat mode [3]. In addition, dispersive modes develop above the flat mode [3], pointing to a possible interpretation in terms of "quantum fragmentation" [1,4].

To check the robustness of this peculiar state, we have studied the role of defects on the ground state and the excitations, using two types of substitutions: titanium instead of zirconium, to directly affect the magnetic interactions, and lanthanum instead of neodymium to reduce the number of magnetic atoms.

We show that defects have a low impact on the crystal electric field scheme (inelastic neutron scattering 2T @LLB). We establish the (H,T) phase diagram of these doped compounds by measuring the magnetization and the magnetic structure (D23 @ILL, G41 @LLB). It shows that the AIAO phase is reinforced by doping (larger Néel temperature and larger critical fields). Surprisingly, the excitations are still well defined in spite of disorder (IN5 @ILL) and the quantum fragmentation state is preserved, with the same characteristic gap energy. We analyze these results using the theoretical model proposed in ref. [4] which allows to determine the parameters of the effective Hamiltonian. From this theory, a phase diagram is used to determine where the different samples are located, with their exchange parameters, in relation to the U(1) spin liquid phase.

- [1] M. Brooks-Bartlett et al, Physical Review X 4, 011007 (2014)
- [2] E. Lhotel and al, Physical Review Letters 115, 197202 (2015)
- [3] S. Petit and al, Nature Physics 12, 746 (2016)
- [4] O. Benton, Physical Review B 94, 104430 (2016)

Magnetic properties of exchange biased nanocrystalline bulk Fe-Cr alloys produced by severe plastic deformation

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Fulfilling the increasing demand of global renewable energy, magnets with high energy products are essential while simultaneously the amount of rare-earth materials in permanent magnets has to be reduced. A promising approach to enhance magnetic properties is the concept of exchange bias whereby a ferro- and antiferromagnet are coupled. Therefore, a magnetic field is applied during annealing underneath the Curie temperature but above the Neel temperature and subsequent field cooling. The consequence is a horizontal shift in the hysteresis loop and therefore an increased maximum energy product.

Due to its wide range of applications the FeCr system (e.g. stainless steel, giant magnetoresistance, etc.) was extensively studied in the past. Nevertheless, examinations of magnetic properties are mostly measured on thinfilms or fine particles and very rare studies are done on bulk samples exhibiting dimension in the millimeter regime. Applicable hard magnets have to exceed these dimensions, meaning bulk materials are necessary for conventional hard magnets. A large miscibility gap in the thermodynamical equilibrium of FeCr, limits the processing possibilities using metallurgical processing routes, particularly medium compositions are difficult to obtain in bulk form. For this purpose, we use a top-down approach: Severe plastic deformation by high-pressure torsion (HPT) uses powders or bulk materials as starting materials and offers the variation of processing parameters like applied pressure, strain and deformation temperature. The resulting mm-sized bulk samples reveal homogeneous microstructures with nanocrystalline or ultrafine-grained grain sizes, which show magnetic properties as they have been observed for thin-films.

In this study, binary Fe and Cr mixtures are used as starting material, while the chemical composition is varied between 30 to 70 at. % Fe. HPT-deformation directly effects the mixing- and microstructural evolution process. Combined with subsequent annealing treatments this method allows a precise tuning of parameters such as the grain size. The microstructural properties are examined by electron microscopy, X-ray diffraction and atom probe tomography whereby special emphasis is placed on a correlation to the magnetic properties. Results obtained by SQUID magnetometry, such as a thermoremanent magnetization in as-deformed FeCr alloys, are connected to the tunable microstructure, being a promising approach to produce exchange biased rare-earth free permanent magnets.

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3894 Competing Magnetic Interactions in SrTm2O4

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*The author has chosen not to make public additional content.

Oral Presentation

3900

Low-Temperature Phase Transitions in Epitaxial Films of the Spin Ice Dy2Ti2O7

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Low-temperature heat capacity and magnetic measurements have been carried out on epitaxial thin (t=10-30 nm) films of the pyrochlore titanate spin ice material Dy₂Ti₂O₇ (DTO). Previous results [1,2] for bulk DTO suggest that at ~0.5 K the system undergoes spin freezing and falls out of equilibrium, indicated by diverging timescales for magnetic relaxation and the onset of hysteresis in magnetization curves. In contrast, heat capacity measurements on the thin films of DTO show no signs of spin freezing down to 0.35 K and appear to undergo a phase transition at around 0.8 K. We present new magnetization measurements and Monte Carlo simulations that allow us to discuss the phase transition behaviour of the thin films in the context of recent works describing a range of proposed phase transitions in thin films of spin ice, including surface-ordering transitions [3] and transitions associated with the F-model [4].

- [1] Snyder et al., Phys. Rev. B 69, 064414 (2004)
- [2] Bovo et al., Nat. Commun. 4, 1535 (2013)
- [3] Jaubert et al., Phys. Rev. Lett. 118, 207206 (2017)
- [4] Bovo et al., Nat. Commun. 10, 1219 (2019)

Evolution of the electronic structure at the magnetic transition in layered iridates upon doping

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Ruddlesden-Popper layered perovskite iridates (Sr_2IrO_4 and $Sr_3Ir_2O_7$) are insulators, although they have 5 electrons in the Ir 5d band and would then rather be expected to be metallic. The strong spin-orbit coupling splits the three t_{2g} states into doubly degenerate Jeff=3/2 states below EF, containing 4 electrons, and a narrow half-filled band near the Fermi level of Jeff=1/2 character, where correlations could be strong enough to form a Mott insulator [1]. A pending question is to understand the link between the magnetic and insulating states. Indeed, antiferromagnetic order is detected below T_N =240K for Sr_2IrO_4 and T_N =280K for $Sr_3Ir_2O_7$, which should be compared to their insulating gaps of 0.6eV and 0.2eV, respectively. These compounds remain insulating above T_N ; in Sr_2IrO_4 there is even hardly any detectable change in resistivity through T_N , while there is a clear drop in $Sr_3Ir_2O_7$. At first sight, this suggests that the insulating character is not directly connected to the AF order. However, optical measurements [2] have confirmed that the gap does not close abruptly at T_N , but there is a small shift and a lot of spectral weight is transferred inside the gap. This behaviour is unusual and not expected in a Mott scenario. Similar behaviour obtained by STM was interpreted as due to a transition between a Slater insulator and a bad metal above T_N , due to strong 2D fluctuations [3].

A metallic state can be induced in Sr_2IrO_4 through doping with rather different behaviours as a function of the substituent: a pseudogapped bad metal state upon Ir/Rh substitution [4], metallic behaviour with collapsing of the Mott gap upon Sr/La substitution [5], both with low substitution rates (few percents) while an insulating behaviour is conserved until a high substitution rate (above 35%) in Ir/Ru substituted Sr_2IrO_4 . Along with the substitution, a reduction of the magnetic order and T_N which does not seem to be related to the Metal-Insulator transition is observed whatever is the substituent. In $Sr_3Ir_2O_7$ a radically different situation can be observed as in the pure compound a dropped in the resistivity curve occurs at T_N , retaining the insulating behaviour and upon Ir/Ru substitution a metal-insulator transition appears above T_N [6].

I will present the results of a temperature dependent ARPES study to investigate the evolution of the band structure at the magnetic transition upon La and Ru substitution. In Ru doped $Sr_3Ir_2O_7$, we have observed a 0.1eV shift towards Fermi level of the Jeff=1/2 band at the X point just above the Neel temperature. I will discuss the origin of this behaviour and contrast it to what is observed in Sr_2IrO_4 upon La and Ru doping.

- [1] B.J. Kim et al., Phys. Rev. Letters 101, 076402 (2008)
- [2] S.J. Moon et al., Phys. Rev. B 80, 195110 (2009)
- [3] Q. Li et al., Scientific reports 3, 3073 (2013)
- [4] A. Louat et al., Phys. Rev. B 97, 161109 (2018)
- [5] V. Brouet et al., Phys. Rev. B 92, 081117 (2015)
- [6] C. Dhital et al., Nat. Commun. 5:3377, (2014)

MIEZE neutron spin echo spectroscopy of strongly correlated electron systems~

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We report recent progress in the development of longitudinal Modulation of IntEnsity with Zero Effort (MIEZE) neutron spin-echo spectroscopy at the beam-line RESEDA [1] at MLZ. The key technical characteristics are summarized which highlight that the parameter range accessible in momentum and energy, as well as its limitations, are extremely well understood and controlled [2].

Typical experimental data comprising quasi-elastic and inelastic scattering are presented, featuring magneto-elastic coupling and crystal field excitations in $Ho_2Ti_2O_7$ [3], the skyrmion lattice to paramagnetic transition under applied magnetic field in MnSi [4], ferromagnetic criticality [5] and spin waves in Fe [6,7]. Taken together, the advantages of MIEZE spectroscopy in studies at small and intermediate momentum transfers comprise an exceptionally wide dynamic range of over nominally seven orders of magnitude, the capability to perform straight forward studies on depolarizing samples or under depolarizing sample environments, as well as incoherently scattering materials.

- [1] C. Franz, and T. Schröder, JLSRF, 1, A1 (2015)
- [2] C. Franz et al., J. Phys.: Conf. Ser., 1316, 012005
- [3] A. Wendl et al. In preperation
- [4] J. Kindervater et al., Phys. Rev. X, 9, 041059 (2019)
- [5] F. Haslbeck et al., Phys. Rev. B. 99, 014429 (2019)
- [6] J. Kindervater et al., Phys. Rev. B. 95, 014429 (2017)
- [7] S. Säubert et al., Phys. Rev. B., accepted (2019)

Hybridized crystal field-phonon bound state in cerium-113 compounds

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The coupling between elementary excitations in crystalline condensed matter can give rise to novel functional properties and exotic states, such as superconductivity, multiferroicity, thermoelectric properties of various types of polar order. Cerium family of strongly correlated, heavy fermion compounds is of particular interest in that case, as different representatives of the family display overlap, or lack, of various of these properties.

We are particularly interested in the CeTAl₃ (*T* is a transition metal), family of compounds, for which an unusual bound state was reported in CeCuAl₃ [1] and CeAuAl₃ [2]. It arises due to the magnetoelastic coupling between the crystal field excitation (CEF) and phonons. Although it was observed in few other compounds, e.g. $Tb_2Ti_2O_7$ [3] or PrNi₂ [4], it was reported as an interaction of CEF and an optical phonon, while for CeAuAl₃ we have observed an interaction of CEF and strongly dispersive acoustic phonon. This points to a different character of the vibronic bound state in CeAuAl₃, and awaits a microscopic explanation. In order to provide a deeper understanding of that phenomenon we are investigating this effect in other compounds, namely CePtAl₃ and CePdAl₃, and its connection with crystal structure and physical properties. In addition, we want to determine the influence of the magnetoelastic coupling in Ce-113 compounds on their magnetic ordering and dynamics.

We have conducted various neutron diffraction and spectroscopy measurements on Ce-113 compounds. Our measurements show, that CePtAl₃ exhibits a modulated antiferromagnetic ordering below T_N =3.35 K, with a modulation vector q=(2/3 0 0), while CePdAl₃ orders antiferromagnetically at T_N =5.61 K. Magnetic properties, models of magnetic structure and first results on spin and lattice dynamics will be discussed.

[1] D.T. Adroja et al., PRL, vol 108, 216402, 2012

[2] P. Cermak et al, PNAS, vol. 116, pp. 6695-6700, 2019

[3] T. Fennel et al, PRL, vol. 112, 017203, 2014

[4] E. Mühle et al., JMMM, vol. 81, pp. 72-78, 1989

Interplay between multipolar spin interactions, Jahn-Teller effect and electronic correlation in a J_eff = 3/2 insulator

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In our work we study the complex entanglement between spin interactions, electron correlation and Janh-Teller structural instabilities in the 5d1 J_eff= 3/2 spin-orbit coupled double perovskite Ba_2NaOsO_6 using first principles approaches. By combining non-collinear magnetic calculations with multipolar pseudospin Hamiltonian analysis and many-body techniques we elucidate the origin of the observed quadrupolar canted antifferomagnetic state as due to the cooperation of Jahn-Teller distortions with dipolar and multipolar Dzyaloshinskii-Moriya interactions.

We find a strong competition between ferromagnetic and antiferromagnetic canted and collinear quadrupolar magnetic phases. The relative magnetic stability and the degree of spin deflection can be controlled by the strength of the electronic correlation (U) and by the degree of Jahn-Teller distortions.

Nanoscale distortions and ground state selection in geometrically frustrated magnets

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Geometrically frustrated magnets, such as triangular networks of antiferromagnetically coupled spins, can display incredibly rich physical properties that may have potential applications in quantum information science and other technologies. Determining if and how magnetic order emerges from competing magnetic tendencies is an important objective in this field. Here, we discuss the Jahn-Teller active triangular AMnO₂ (A= Na, Cu) antiferromagnets [1] to highlight that the degree of frustration, mediated by residual disorder, contributes to the rather differing pathways towards a single, stable magnetic ground state, albeit with varying ordering temperatures. For these insulating sister compounds, complementary high-resolution synchrotron XRD, local-probe muon-spin relaxation (μ^+SR) studies, corroborate that the layered NaMnO₂ adopts a remarkable magnetostructurally inhomogeneous ground state. [2] In view of this peculiarity, we employ powerful neutron total scattering and magnetic pair distribution function (PDF) analysis to uncover that although CuMnO₂ undergoes a conventional symmetry-lowering lattice distortion driven by Néel order, in the Na-derivative a short-range triclinic distortion (Fig. 1) lifts the degeneracy of the isosceles triangular network on the nanoscale, thereby enabling longrange magnetism to develop with enhanced magnetic correlations above the transition. [3] More generally, the work illuminates the cooperative intertwining of the local atomic and magnetic structures that permits ground state selection when spatial inhomogeneity meets geometrical frustration, a mechanism that may also be operative in other frustrated materials with electronically active transition metal cations.



Figure 1 : Color map of the strength of triclinic distortion, *d*, as a function of temperature in AMnO₂ (A= Na, Cu); the horizontal dashed lines mark the Néel order.

- [1] M. Giot et al., Phys. Rev. Lett. 99, 247211 (2007)
- [2] A. Zorko et al., Sci. Rep. 5, 9272 (2015)
- [3] B. A. Frandsen et al., Phys. Rev. B 101, 024423 (2020)

Coexistence of Kondo correlations with partial magnetization in a frustrated trimer coupled to a ferromagnet

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Triple quantum dots organized in a triangle, called also trimers, are predicted to host exceptionally rich strongly correlated physics, including a quantum phase transition separating conventional and ferromagnetic Kondo phases [1] and a crossover between ferromagnetic and under-screened Kondo regimes [2]. Their phase diagram grows even richer, when they are coupled to a magnetic bath, distinguishing one spin direction [3]. This is caused by the ferromagnetic exchange field induced in the nano-structure by the magnetic leads in the absence of global particle-hole symmetry, which is broken by the frustration even if each quantum dot is tuned to be at its symmetry point.

In this contribution, results of numerical renormalization group calculations concerning the particularly intriguing regime of the trimer strongly coupled to the leads exhibiting itinerant ferromagnetism will be presented. It will be shown that within a wide regime of model parameters the ground state comprises fully or partially Kondo-screened quantum dots, which are quite significantly magnetically polarized – even for extremely weak frustrating coupling. This is in contrast to the case of single quantum dots, where the quantum-dot spin polarization is proportional to the symmetry-breaking detuning. Other physical properties of the state, such as linear-response conductance, tunneling magneto-resistance and current spin polarization, exhibit a high degree of tunability. The generic character of the model allows to hope that similar partially magnetized Kondo states may exist in bulk in frustrated lattices with non-equivalent Kondo sites.

- [1] A.K. Mitchell, T.F. Jarrold, D.E. Logan, Phys. Rev. B 79, 085124 (2009)
- [2] A.K. Mitchell, T.F. Jarrold, M.R. Galpin, D.E. Logan, J. Phys. Chem. B 117, 12777 (2013)
- [3] K.P. Wójcik, I. Weymann, J. Kroha, Phys. Rev. B 102, 035141 (2020)

2645 Electromagnons in GdMn2O5

Balédent Victor

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Electromagnons are unusual elementary excitations that can be defined as electro-active magnetic excitations. This definition arises from its experimental evidence: a magnetic collective mode that you can activate with en electric field (from light for example). There are several propositions to explain such mode (magnon-phonon coupling, crystal field- magnon coupling etc...). These excitations are usually found in multiferroic systems like the famous manganite RMnO3 and RMn2O5, despite this condition has not been proven to be mandatory. In this talk, I will present recent experimental results on multiferroic GdMn2O5 proving the existence of a collective mode satisfying the electromagnon definition. I will compare these results to possible models reported in literature and discuss the necessity for other mechanisms and theory for such an hybrid mode.

3550 Geometrically Frustrated Iridate Ca5Ir3O12

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In a crystal structure of $Ca_5Ir_3O_{12}$ which is a hexagonal with noncentrosymmetric (P6-2m No. 189), 1D chains of edge-sharing IrO_6 octahedra along *c*-axis form triangular lattice in *c*-plane. An average valence of Iridium ions is +4.67; a ratio of $Ir^{4+}:Ir^{5+}$ is 1:2. This situation can lead to the geometrical frustration of charge on both the triangular lattice in *c*-plane and 1D chains along *c*-axis. $Ca_5Ir_3O_{12}$ shows an antiferromagnetic ordering below 7.8 K and a second order phase transition at 105 K [1]. Previous Raman scattering experiments to the single crystal of $Ca_5Ir_3O_{12}$ have shown that the phase transition at 105 K is a structural phase transition [2], however, the origin of the phase transition is not clear at present. Therefore, the crystal and magnetic structure at low temperature have not been yet clarified. The single crystal of $Ca_5Ir_3O_{12}$ shows the semi-conductivity and nonlinear electrical conductivity along *c*-axis [3, 4].

This study was made to investigate the magnetic property of the single crystal of $Ca_5Ir_3O_{12}$. The temperature dependence of magnetic susceptibility was measured under the field up to 7 kOe using a SQUID-type magnetometer (Quantum Design's MPMS 3). The *M/H* indicates anomaly at 7.8 K under a weak magnetic field (~100 Oe) both along *c*-axis and *c*-plane. This transition temperature and the anisotropy are consistent with previous study [5]. We confirmed that the transition temperature increases with increasing the amplitude of the field along *c*-axis. We will present the experimental results in this presentation.

- [1] M. Wakeshima, et al., Solid State Commun., 125, 311 (2003).
- [2] T. Hasegawa, et al., accepted in J. Phys. Soc. Jpn.
- [3] K. Matsuhira, et al., J. Phys. Soc. Jpn. 87, 013703 (2018).
- [4] H. Hanate, et al., J. Mann. Mag. Mater. 498, 166203 (2020).
- [5] G. Cao, et al., Phys. Rev. B 75, 134402 (2007).

Temperature dependence on the hyperfine field of Ce impurities in Gd: an intermediate valence model

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An intermediate valence model for Ce impurities (Ce = [Xe] $4f^1 5d^1 6s^2$) diluted in Gd metal is used to describe the local magnetic moments of this impurity diluted in Gd metal in the whole range of temperatures of the Ce <u>Gd</u> system ($0 \le T \le 293,5K$). The temperature dependence of the local magnetic moments and related hyperfine magnetic interaction are calculated adopting a functional integral approach in the static approximation. Our self-consistent theoretical calculations are in very good agreement with experimental results [1], in particular exhibiting the two regimes of the magnetic hyperfine fields (and local magnetic moments) with temperature. The obtained valence state of Ce impurity ranges from 0.75 at 0K to 0.50 at $T = T_c$.

[1] T.A. Thiel, E. Gerdau, M. Böttcher, G. Netz. Hyperfine Interact. 9 (1981) 459

Structure and Frustrated Magnetism of Layered Alkali Manganites

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Magnetically frustrated compounds are promising for the emergence of various exotic magnetic states. They can demonstrate spin-liquid and spin-ice behaviour, act as valence-bond solids, exhibit an array of helical and cycloidal spirals, or even a variety of periodic skyrmion states with non-trivial topologies composed of skyrmions and antiskyrmions [1-3]. Magnetic frustration results from the competition between ferro- and antiferromagnetic exchange interactions in crystal lattices based on triangles or tetrahedra that share corners, edges or faces [4-5]. One group of interesting compounds from this point of view is the alkali manganites $A_xMnO_2 \cdot yH_2O$ (A = Na, K) with the birnessite structure. These antiferromagnetic compounds have frustrated 2D triangular planes of magnetic $Mn^{3+/4+}$ ions, which form edge-shared MnO_6 octahedral structural units. These layers are well decoupled from each other by non-magnetic A^+ cations and H_2O molecules. This type of structure, with weakly coupled planes of spins pointing typically in the out-of-plane direction, allows tuning of the interlayer distance, the ratio of Mn^{3+} to Mn^{4+} and thus the magnetic exchange and anisotropy along the stacking direction.

The synthesis by different methods and a comprehensive investigation of the magnetic properties of $A_x MnO_2 \cdot yH_2O$ (A = Na, K) will be presented. Determination of the composition and crystal structure was performed by powder Xray diffraction, thermogravimetry, differential scanning calorimetry, X-ray photoelectron spectroscopy, scanning and transmission electron microscopy. The static and dynamic magnetic properties of the compounds have been investigated in detail by dc and ac susceptibility, and magnetization time-decay measurements.

- [1] T. Okubo, S. Chung, H. Kawamura, Phys. Rev. Lett. 108, 017206 (2012)
- [2] S. Hayami, Sh. Z. Lin, C. D. Batista, Phys. Rev. B 93, 184413 (2016)
- [3] A. O. Leonov, M. Mostovoy, Nat. Commun. 6, 8275 (2015)
- [4] G. Toulouse, Commun. Phys. 2, 115 (1977)
- [5] J. E. Greedan, J. Mater. Chem. 11, 37 (2001)

Simultaneous individual and dipolar collective properties in binary assemblies of magnetic nanoparticles

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Applications based on aggregates of magnetic nanoparticles are becoming increasingly widespread, ranging from hyperthermia to magnetic recording. However, although some uses require collective behavior, others need a more individual-like response, the conditions leading to either of these behaviors are still poorly understood. The preparation of binary random compacts (BRCs) with different proportions of low- and high-anisotropy bare oxide nanoparticles (NPs), each with a narrow size distribution, is shown to be an efficient method to tune the magnetic properties of such dense NP assemblies. Here, we have studied a series of BRCs comprising by nanoparticles with a large difference in anisotropy energy (ratio > 8). This series has been achieved mixing equally sized (6.8 nm) pure maghemite (γ -Fe₂O₃) (low-anisotropy) and Co-doped maghemite (high-anisotropy) nanoparticles.

Interestingly, the mixtures display double-loop responses at 5K, indicating that single particle anisotropy dominate in the magnetization reversal process, although the dipolar interactions are strong enough to fully couple the two types of NPs at low fields (e.g., yielding a single collective freezing). The effect of the varying dipolar interactions through the series on the coercive and exchange bias fields has been discussed. This scenario offers new insight (systematically ratified by simulations) into the subtle interplay between dipolar interactions, local anisotropy and sample heterogeneity to determine the behavior of dense assemblies of magnetic nanoparticles.



Figure 1

[1] Elena H. Sánchez et al., Chem. Mater. 32, 3 (2020)

Epitaxial thin films of the quantum spin ice, Yb2Ti2O7

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Spin ices are highly frustrated magnets that have a pyrochlore lattice. The general stoichiometric form of spin ice is $A_2B_2O_7$ where A and B are cations. These cations are generally structured into separate lattices of corner sharing tetrahedra, which are interpenetrating. Spin ice gets its name from the comparison of its magnetic structure and water ice's physical structure. The most notable feature of spin ice materials is their magnetic monopole - antimonopole pairs.

Over the last few years, epitaxial thin films of spin ice and related materials with the stoichiometry of $A_2Ti_2O_7$ have been grown and characterised [1]. Recently we have achieved epitaxial growth of the quantum spin ice candidate Yb₂Ti₂O₇ on an Y₂Ti₂O₇ substrate. Three 66 nm films were grown such that their out-of-plane direction corresponded to the [111], [1-10] and [100] crystallographic directions. The films were characterised using magnetometry, heat capacity and X-ray diffraction experiments. The entropy of both the [1-10] and [100] films are around *R* ln(2), which is in agreement with the bulk. However, the entropy of the [111] film is anomalously high. This anomaly may be due to pyrochlore growth along the [111] direction having a competing fluorite crystallographic phase under the growth conditions employed, with associated disorder providing the additional entropy.

[1] L. Bovo et al., Nat. Commun. 5, 3439 (2014)

Bypassing dynamical freezing and accessing low-energy microstates in artificial kagome ice

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Spin liquids are disordered states of matter that still fluctuate at low temperature. In many cases, the ground state of spin liquids is expected to be ordered, for example, because the dipolar interaction lifts the (quasi)degeneracy between low-energy spin configurations. However, this is not what is observed experimentally, and many systems dynamically freeze in a liquid phase before the ordering takes place. Dynamical freezing is related to the existence of energy barriers between quasidegenerate configurations that are hardly overcome with single spin flip events once the system has been cooled to low temperature.

In spin ice compounds or artificial magnets, ground state configurations are then generally out of reach for practical reasons. In this presentation, we will show how the problem of dynamical freezing can be bypassed in an artificial kagome ice. We will illustrate the efficiency of our method by demonstrating that the a priori dynamically inaccessible ordered ground state of the dipolar kagome ice is imaged reproducibly in real space, and at room temperature.

The central idea of this work is to replace the spin degree of freedom by a micromagnetic knob, which can be adjusted by a proper design of the vertex geometry. Our strategy is to imprint the desired spin configuration, vertex by vertex, in a kagome array made of connected nanomagnets, with a notch at each vertex site. Each notch locally lifts the energy degeneracy between the six states of a given vertex satisfying the ice rule constraint, allowing to reach efficiently any desired spin microstate using a field demagnetization protocol.

This concept will be demonstrated both numerically and experimentally, and we will show how we can access all predicted phases of the dipolar kagome ice.

Spatial Homogeneity and Magnetization Dynamics in FM insulator Ga1-xMnxN Probed by Muon-Spin-Relaxation Spectroscopy

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 $Ga_{1-x}Mn_xN$ belongs to the well-known family of dilute ferromagnetic semiconductors which exhibits low-temperature ferromagnetism driven by ferromagnetic superexchange among the Mn^{3+} ions. However, in this material systematic information about the uniformity and of the volume fraction of the ferromagnetic phase and on the distribution of internal fields is generally lacking.

We report here on the results of the very first studies of spatial homogeneity of ferromagnetic phase in the Ga₁₋ _xMn_xN using muon-spin-relaxation (μ SR) spectroscopy applied to a single phase and high quality molecular beam epitaxy grown Ga_{1-x}Mn_xN layer with x = 9% and Curie temperature T_C = 10 K [1].

Our μ SR measurements revealed that the magnitude of the asymmetry parameter A started to saturate (or the magnetic volume fraction rolled off to zero) at temperatures being about 10 K above the value of the T_c established beforehand for this sample by SQUID magnetometry [1]. These temperature scans were obtained for 50 and 1500 Oe in transverse field (TF) configuration, and are indicative that the magnetic configurations of Mn species are strongly affected by the external field, at least on a time scale relevant to μ SR. Another rather unexpected finding of our μ SR measurements is the observation that the temperature dependence of the TF μ SR depolarization rate λ has truly a λ -like shape, when plotted in a log-log scale, peaking strongly at T = 20 K, that is where A starts to saturate. Above this temperature λ obeys a clear power law dependence on T, I ~ T^{- α}, where $\alpha > 0.2$ ms⁻¹, corresponding to a previously observed similar dependence of magnetic susceptibility in Ga_{1-x}Mn_xN [2]. We assign this nonstandard dependence to a gradual formation of coupled neighbor spin clusters (spin pairs, triplets, etc.) on lowering temperature. Importantly, these clearly non-zero values of λ are seen well above T_c in a very well characterized single phase material [1].

[2] S. Stefanowicz et al., Phys. Rev. B 88, 081201(R) (2013)

This work has been supported by the National Science Centre, Poland through OPUS grant DEC-2018/31/B/ST3/03438.

^[1] K. Gas et al, J. Alloys Compd. 747, 946 (2018)

4197 Magnetic fragmentation within an Arctic circle

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In frustrated spin systems, the ground state manifold, i.e. the family of states of lowest energy, is often completely defined by local constrains inherited from the frustrated nature of spin interactions. These constraints give rise to exotic phases hosting for instance very peculiar dynamics: magnetic monopoles, loop-like excitations, etc. While these systems have been studied widely under open or periodic boundaries conditions, recent interest has been given to а specific kind of boundaries in 2D: Domain Wall Boundary Conditions. By fixing the spin on the borders of the system in a controlled way, the collective organisation unexpectedly propagates at long distance within the bulk of the system. This careful choice of the borders constraints spectacularly results in a spontaneous phase separation of the system: an extensive part freezes while another one fluctuates. This phenomenon is known as the Arctic Circle Phenomenon [1]. We show in this work that a Monte Carlo approach with the appropriate dynamics highlights this phenomenon in several models that are experimentally accessible with the use of meta magnets, like artificial spin ices [2]. More precisely, we demonstrate that we may observe a fragmented spin liquid [3] within a kagome-based Arctic Circle. We also propose a route to generalise these results in the archetypal three-dimensional condensed matter pyrochlore lattice, provided we carefully implement the appropriate spin model.

- [1] L F Cugliandolo, G Gonnella, and A Pelizzola. Journal of Statistical Mechanics: Theory and Experiment (2015)
- [2] Yann Perrin, Benjamin Canals, and Nicolas Rougemaille. Nature (2016)
- [3] Canals, B., Chioar, I., Nguyen, V. et al, Nature Communications (2016)

Surface ferromagnetism in Pr0.5Ca0.5MnO3 nanoparticles as a consequence of local imbalance Mn3+:Mn4+ ratio

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Half-doped praseodymium manganites, Pr_{0.5}Ca_{0.5}MnO₃, nanoparticles synthesized by sol-gel method are annealed at different temperatures in order to obtain nanoparticles sizes ranging from 15 to 100 nm. HRTEM shows that the nanoparticles are very crystalline with no structural disorder at the surface. Rietveld powder diffraction profile determines that the samples have orthorhombic structure with space group Pnma and cell volume decreases with increasing particle size. The thermodiffraction patterns from 50 to 300 K indicate that the charge ordering (CO) is suppressed for the smallest particles, whereas the largest one still resembles the bulk. Hysteresis loops and ZFC-FC curves evince that, as the most of half-doped (Mn³⁺:Mn⁴⁺ = 1) antiferromagnetic (AFM) charge-ordered manganites, ferromagnetism (FM) appears as particle size decreases. Hysteresis loops at FC procedure show a coercivity increase and a shift to negative field, confirming that both AFM and FM phases not only coexist but are also coupled. In addition, а spin glass behaviour is observed with glass temperature around 50 Κ. A possible origin of FM phase development is the lacking oxygen ligands at the surface that increases Mn³⁺ at expenses of Mn^{4+} in order to maintain the electronic neutrality. In this work, we show that $Mn^{3+}:Mn^{4+} = 1$ ratio is preserved in the whole crystalline particle with the exception of the surface, where this ratio changes to Mn³⁺:Mn⁴⁺ >1. This Mn^{3+} excess makes double exchange $Mn^{3+} \rightarrow O^{2-} \rightarrow Mn^{4+}$ prevails over the super-exchange giving place to the FM interactions. We show that a "shell" thickness of only one-unit cell, with a_{cell} = 0.38 nm, is enough to explain the onset of FM at the surface, whereas the volume remains AFM. Furthermore, the FM to AFM ratio fits to the increase of surface to volume ratio with decreasing particle size.[1].



[1] HD Aliyu, JM Alonso, P de la Presa et al., Chem Mater 30, 7138-7145 (2018)

This work was supported by grants from the Spanish Ministry of Science and Innovation MAT2015- 67557-C2-1-P.

4281 Switching barriers in artificial square ice

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The relaxation kinetics and emerging correlations of arrays of interacting Ising-like nanomagnets are governed by the switching rates of the individual magnets. These rates depend, via the Arrhenius law, on the energy barrier for moment reversal in the nanomagnets. In this work, we consider how the switching behaviour of a nanomagnet in archetypical artificial square ice is modified by the interaction with its six nearest neighbours. We find that due to two effects the transition barriers can be significantly lowered compared to an approximation commonly used for kinetic Monte Carlo simulations:

Based on a simplified point-dipole model, we find that the barriers for clockwise and counter-clockwise uniform rotation can differ significantly due to the interactions with neighbours. For a subset of environmental microstates this leads to a significant enhancement of the switching rate, which is especially marked in the limit of rare events.
 Micromagnetic simulations employing the string method to model the moment reversal in an extended nanomagnets reveal that, in addition to the clockwise/counter-clockwise splitting, the environments promote non-uniform reversal leading to further reductions of the switching barriers.

These results highlight that local microstates can significantly enhance moment reversals already in ideal systems, without defects or additional anisotropies, and are particularly pronounced for fully-magnetised configurations. Thus, we expect that the initial stages of demagnetisation will proceed faster than assumed from previous models. These findings are of integral importance to achieve realistic kinetic Monte Carlo simulations of emergent correlations in extended artificial spin systems or the evolution of small-scale nanomagnetic logic circuits.

Symposium 16. Magnetic thin-films, interfaces and multilayers

Spin-dependent Thermoelectric Phenomena in 3D Interconnected Multilayered Nanowire Networks

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The emerging field of spin caloritronics combines heat-driven transport with spintronics [1] and allows promising applications, such as magnetic heat valves or magnetically switchable cooling. However, dimensions in magnetic nanostructures lead to major experimental issues such as insufficient power generation capability and lack of reliable methods to obtain key spin caloritronic parameters [2-3]. In contrast, more efficient macroscopic spin caloritronics devices built from 3D interconnected multi-layered nanowire (NW) networks, as shown in Fig. 1(a-b), allow magnetically-controlled Peltier cooling of macroscopic components and the directly extraction of accurate key spin caloritronics parameters [4-6]. They are fabricated by pulsed electrodeposition into 3D nano-porous polymer host membranes with no sample size limitation. The branched structure provides a reasonable mechanical robustness to the networks, while the membrane provides a certain flexibility. The local removing of the cathode from which the NWs are grown yields a two-probe design suitable for transport measurements. As presented in Fig. 1, we developed experimental set-ups for magneto-Seebeck (c) and magneto-Peltier (d) measurements. We studied FM/Cu (FM = Co, CoNi, Py) multilayered NW networks showing giant magneto-transport (electrical, thermal and thermoelectric) properties, with room-temperature (RT) CPP-GMR up to 40%, and power factors comparable to that of bismuth telluride and that are magnetically modulated with magneto-power factor ratios up to 140% at RT [4-6]. Our interconnected NW networks allow for the direct extraction of large spin-dependent Seebeck and Peltier coefficients from measured parameters and the direct observation of Peltier cooling at the electrode-NW network junction with large Peltier cooling ability. Those observations hold promise for magnetically modulated energy conversion using light and flexible thermoelectric generators and could lead to advances in future spin-caloritronic devices.



Figure 1 : (a) SEM image of a self-supported interconnected NW network film. The inset highlights the NW branched structure. (b) Schematic of the interconnected NW network with alternating magnetic and non-magnetic layers embedded within the 3D nanoporous polycarbonate template. (c) Device configuration for magneto-Seebeck measurements: a temperature difference ΔT between the metallic electrodes generates a thermoelectric voltage ΔV . (d) Device configuration for magneto-Peltier measurements: Peltier heating and cooling are simultaneously recorded at the electrode-NW network junctions when the polarity of the electric current flow *j* is reversed.

[1] G. E. W. Bauer, E. Saitoh and B. J. van Wees, Nature Materials 11, 391 EP (2012)

[2] N. Liebing et al., Phys. Rev. Lett. 107, 177201 (2011).

[3] M. Walter et al., Nature Materials 10, 742 EP (2011)

[4] T. da Câmara Santa Clara Gomes, F. Abreu Araujo and L. Piraux, Science Advances, 5(3), eaav2782 (2019)

[5] F. Abreu Araujo, T. da Câmara Santa Clara Gomes and L. Piraux, Advanced Electronic Materials 5, 1800819 (2019) [6] N. Marchal et al. journal of applied physics (In review)

[6] N. Marchal et al., journal of applied physics (In review)

Tuning the magnetic behaviour of Fe/Cu electrodeposited nanowires with controllable Fe and Cu

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The need for low energy technologies and the recent advances in chemical and self-assembly synthesis techniques have boosted the growth of three-dimensional nano-objects. Nanostructures composed of ferromagnetic materials combined with non-magnetic materials with complex and controlled magnetic behaviour have been extensively studied during the last years [1]. The capacity to control size and shape, not only in the plane but also along the vertical direction, leads to the appearance of new effects in spin configurations, especially when the system has a configuration with different segments. A set of multisegmented Fe/Cu nanowires were synthesized by a two-step anodization process of aluminum substrates and a pulsed electrodeposition technique using a single bath. While the diameter was kept constant (45 ± 5) nm, the Cu length was varied between 15 and 120 nm whereas Fe segment length was varied from 30 to 300 nm. The influence of the non-magnetic layer thickness variation on the nanowires' magnetic properties was investigated through first-order reversal curve (FORC) measurements and micromagnetic simulations. Our analysis confirms that in the multi-segmented Fe/Cu nanowires with shorter Cu segments, the dipolar coupling between Fe segments controls the magnetic behaviour of the nanowires and its performance is like a homogenous Fe nanowire array of similar dimensions. The multi-segmented Fe/Cu nanowires with larger Cu segments act like a collection of non-interacting magnetic entities and their global behaviour is mainly controlled by the neighbour-to-neighbour nanodiscs' dipolar interactions [2]. On the other hand, by tuning the magnetic segment length we were also able to study the influence of Fe aspect ratio in the magnetic properties of the Fe/Cu segmented nanowires. The results show a magnetic anisotropic behaviour for Fe/Cu NWs with the easy magnetization axis lying parallel to the wire axis, arising from the competition between the magnetocrystalline anisotropy and the shape anisotropy factors [3].

[1] L. Peixoto et al., Applied Physics Review 7 (2020) 011310

[2] S. Moraes et al., Nanomaterials 8 (2018) 490

[3] M. Susano and M. P. Proenca et al., Nanotechnology 27 (2016) 335301

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Transmission of high-energy electrons through magnetic and nonmagnetic Schottky junctions

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Using the electron beam of a scanning electron microscope as an external current source with tunable energy, we investigate the transport properties of high-energy electrons injected from vacuum into the metal layer of Pt/Cu/Si and Pt/spin valve/Cu/Si Schottky junctions.

When the injection energy is varied between 1 and 30 keV, the current transmitted into the semiconductor of Pt/Cu/Si junctions increases by several orders of magnitude and reaches values orders of magnitude larger than the current injected from vacuum. Inspecting the energy-dependence of the transmitted current we identify two transport regimes. In the limit of low injection energies and thick metal films, the transport is dominated by the formation and propagation of a secondary electron distribution in the metal layer. However, when the injection energy is sufficiently large and the metal layer sufficiently thin, electrons are transmitted into the semiconductor with negligible energy loss, i.e., the metal layer becomes essentially transparent. The transmitted current is then dominated by impact ionization in the semiconductor. When the metal layer of the Schottky junction is relatively thick and the injection energy of a few keV typically, the transmitted current increases abruptly. The origin of this abrupt change is interpreted as a combined effect of a quasi-ballistic electron transport in the metal layer and a sudden variation of the density of states in the semiconductor substrate.

When transmission experiments are performed in Pt/spin valve/Cu/Si Schottky junctions, the current transmitted into the semiconductor depends on the relative orientation of the magnetizations in the two ferromagnetic layers of the spin valve. Reversing back and forth the magnetization in the soft ferromagnetic layer then leads to a magneto-current, similar to what is observed in spintronic devices but for hot electrons. The origin and amplitude of this magneto-current will be discussed.

Different origins of magnetostriction and magnetic anisotropy in Ga-rich FeGa alloys

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Fe_{100-x}Ga_x alloys are promising materials for magneto-elastic (ME) applications [1-2] since they are rare-earth free systems with large magnetostriction and low coercivity. The magnetostriction as a function of Ga content shows two maxima for both, x close to 20 at.% and 28 at.%, respectively, being higher in the Ga-rich range (28 at.%). The reported strong influence of processing on the magnetostrictive properties for bulk alloys indicate the necessity of a precise control of the growth method when trying to develop FeGa nanosystems [2]. In our recent investigations about Ga-rich Fe-Ga thin films, we have explored how the flow regime of the sputtering technique can be used to tune the in-plane magnetic anisotropy [3]. Furthermore, we have also studied how anisotropy is also dependent on the layer thickness [4-5]. The structural characterization combining x-ray diffractometry (XRD) and x-ray absorption near the edge spectroscopy (XANES) together with the magnetic study done by vibrating sample magnetometry (VSM) indicate that both, growth parameters and thickness, have an influence on the structural phases present in the Ga-rich Fe-Ga films. The increase of the B2 phase together with a preferential alignment of Ga-pairs that tetragonally distort the lattice seems to play a role on the enhancement of the magnetic anisotropy as the films thickness is above 100 nm. When modifying the growth power for the sputtering deposition similar conclusions can be reached [6]. We have used the cantilever beam technique with capacitive detection to also determine the thickness dependence of the effective ME parameter B_{eff} [5]. It is observed a decrease of the effective ME coefficient with the film thickness, being around -7.6 MPa for a 250 nm thick film. Despite the presence of a nonhomogeneous Ga pair distribution, which could suggest an enhancement B_{eff} with respect to the values obtained for films with isotropic distribution of Ga pairs, the thickness dependence of Beff can be explain considering the grain distribution. Thus, the expected value for B_{eff} on films with <110> texture and random distribution of grains in the film plane can be weaken if the proportion of <001> in-plane grains increases and the presence of <111> grains is reduced. Overall, while the magnetic anisotropy seems to be related to the local range order (Ga pairs and tetragonal distortion) in Ga-rich Fe_{100-x}Ga_x alloys, the magnetoelastic behavior can be understood considering the distribution of grains, i.e., medium range order.





[1] A. E. Clark et al., IEEE Trans. Magn. 36, 3238 (2000)

- [2] Q. Xing et al., Acta Materialia 56, 4536 (2008)
- [3] A. Muñoz-Noval et al., Phys. Rev. B 93, 214408 (2016)
- [4] A. Muñoz-Noval et al., J. Alloys Compnd. 745, 413 (2018)
- [5] P. Bartolomé et al., J. Phys. Chem. C 124, 4717 (2020)
- [6] A. Muñoz-Noval et al., J. Phys. Chem. C 123, 13131 (2019)

Ferromagnetic resonance study on Ni-based ferromagnetic films/ferroelectric substrate systems

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Multiferroic materials have attracted much attention for its potential application in spintronic devices. [1] However, the magnetoelectric coupling effect in single-phase multiferroics can be hardly achieved at room temperature. Fabrication of artificial multiferroic heterojunctions consisting of ferromagnetic/ferroelectric layers is expected to pave a way to synthesize or create a novel multiferroic materials which can operate at room temperature. [2] Several studies which fabricated the heterojunctions to create a novel multiferroic material could provide the controllability and multifunctionality to the system. In our previous works, [2] the heterojunction formation of Ni/LiNbO₃ substrate enabled us to control the uniaxial magnetic anisotropy and its domain structure. Then, the magnetization dynamics of the system consisting of Ni/LiNbO₃ substrate is also investigated. As a result, damping constant and rectifying amplitude trended to be enhanced when the stripe domain structure was form in the Ni layer.

In this study, to further investigate the magnetization dynamics in the systems consisting of heterojunctions, ferromagnetic resonance (FMR) [3] was measured for various systems: e. g. Ni/LiNbO₃, Ni/W/LiNbO₃, Ni/SiO₂/Si, Ni/Si, Ni/W/SiO₂/Si, and Ni/W/Si etc. We also fabricated a system comprising various thick Ni and Ni/W multilayers onto a single crystalline Y-cut 128° LiNbO₃ substrate by using magnetron sputtering process. Figure 1(a) shows the typical FMR spectra of 30-nm-thick Ni/LiNbO₃ substrate, Ni/Si substrate, and Ni/SiO₂/Si substrate. Using macro-spin model, [4, 5] we estimated the resonance frequency and damping constant based on the fitting function consisting of the symmetric Lorentz-like and asymmetric dispersive-like functions. Figure 1(b) shows the film thickness dependence of ferromagnetic resonance frequency for the Ni/LiNbO₃ and Ni/Si substrates. As shown in Figs. 1(a) and 1(b), the FMR spectra and magnetization dynamic properties are strongly dependent on the combination of ferromagnetic film and substrate type. These results are attributed to the modulation of electric state distribution near the interface between the ferromagnetic layer and substrate.

Our results manifest that the heterojunction formation plays a significant role in controlling or modulating the ferromagnetic characteristics.



Figure 1: (a) Typical ferromagnetic resonance (FMR) spectra of Ni/LiNbO₃, Ni/Si, and Ni/SiO₂/Si substrates. For clarity, the spectra are vertically shifted. (b) FMR field as a function film thickness.

[1] M. Fiebig, Th. Lottermoser, D. Meier, M. Trassin, Nat. Rev. Mater. 1, 16046 (2016)

- [2] A. Yamaguchi, T. Ohkochi, A. Yasui, T. Kinoshita, K. Yamada, J. Magn. Magn. Mater. 453, 107 (2018)
- [3] H. Suhl, IEEE Trans. Magn. 34, 1834 (1998)
- [4] T. L. Gilbert, IEEE Trans. Magn. 40, 3443 (2004)

[5] A. Yamaguchi, K. Motoi, A. Hirohata, H. Miyajima, Y. Miyashita and Y. Sanada, Phys. Rev. B 78, 104401 (2008)

Wide Band Direct On-chip EMI Shielding Layer with Metallic/Magnetic Multilayer

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For the application of smart phones, IoT devices and electric vehicles, the electromagnetic interference (EMI) between semiconductor chips has become a serious issue [1]. Since many kind of semiconductor chips are densely implemented in a narrow space, an EMI shielding layer that is directly deposited on the semiconductor chips [2] is a promising solution. However, further reduction in its thickness is necessary from a viewpoint of mass production. A multilayer consist of non-magnetic conductive layers and magnetic layers was proposed for thin shielding layer [3]. It uses the multi-reflection of electromagnetic wave at the interface of metallic/magnetic layers and it showed better shielding effect than Cu layer at GHz range [3]. In this paper, EMI shielding effect of a multilayer system with NiFeCuMo magnetic layer and Cu layer is investigated. It is found that the multilayer with total thickness of 1 μ m shows better shielding effect than that of 3 μ m Cu layer at sub-GHz range. It is also found that another magnetic/metallic multilayer system still shows better shielding effect at sub-100 MHz range. This is suitable for the application of smart phones or EVs where EMI shielding at sub MHz range is required. Multilayer samples were deposited on a glass substrate by magnetron sputtering. Ni₇₇-Fe₁₄-Cu₅-Mo₄ (at.%) target was used for the NiFeCuMo magnetic layer. Shielding effect was estimated with the same method as [3]. Magnetic shielding effect (MSE (dB)) is defined as -20log (transmitted power/input power). It was found that [Ta(5nm)/NiFeCuMo(50nm)]n has small coercivity regardless of ite total thickness. Ta layer preventing the crystal growth in the NiFeCuMo layer. Figure 2 shows a hysteresis loop of Cu(400nm)/[Ta(5nm)/NiFeCuMo(50nm)]₄/ Cu(400nm) sample. Coercivity is less than 1 Oe. Figure 3 shows the result of shielding effect of the sample shown in Fig.2. Cu layer with thickness of 3 μ m and 1 μ m were also estimated. The sample (thickness: 1μ m) shows higher MSE than that of Cu layer with the thickness of 3 µm. The peak at around 500 MHz is due to the FMR frequency of the NiFeCuMo layer. Since this peak has relatively long tail to the low frequency range, the MSE gain is shown even at 100 MHz. The FMR frequency is not expected for the frequency range of kHz to MHz. However, the domain wall resonance has its peak frequency at MHz range [4]. Figure 4 shows MSE of a sample with a stack of $[Cu(100nm)/NiFeCuMo(100nm)]_{10}$ (thickness: 2 µm). This sample has higher MSE than Cu layer and has a clear peak at around 50 MHz. This peak is due to the domain wall resonance. Thus, it is shown that a wide band shielding effect can be achieved by using a metallic/magnetic multilayer system. One concern is its processing cost. However, multi-cathode sputtering system such as CCS-2110 [5] may be a solution.



[1] L. Lavagno et al. (Ed.): EDA for IC
Implementation, Circuit Design and
Process Technology, CRC Press, Boca
Raton, (2006)
[2] Y. Shimada et al.IEEE Trans. Magn.
50, 2801704 (2014)
[3] K. Yamada et al., 2011 IEEE
International Symposium on EMC, 432
(2011)
[4] S. Chikazumi: Physics of Magnetism,
Wiley, New York (1995)
[5]
https://www.shibaura.co.jp/e/products
/vacuum/vac_10.html

Magnetic and Structural Properties Of Co2FeGe Thin Films Prepared By Magnetron Sputtering

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Stoichiometric Heusler alloy (HA) Co₂FeGe (CFG) is half-metallic ferromagnet with high magnetization and Curie temperature. These make CFG HA films very attractive for spintronics applications [1-3]. Half-metallic HA with a gap in one spin channel allow low values for another key parameter of spintronic devices - Gilbert damping constant (α) that is proportional to the density of states on the Fermi level [4]. Properties of HA thin films could be tuned by material choice and atomic ordering.

Here we report preparation, structural and magnetic characterization of CFG films in the vicinity of stoichiometric composition. Films of 60 nm thickness were deposited on Corning Glass substrates at T_s = 20°C (RT), 300°C and 500°C by magnetron co – sputtering from Co₂Fe and Ge targets in 5 mTorr of Ar. Deposition rates were fixed at 0.128 nm/sec and 0.085 nm/sec, respectively, assuring desired composition. To evaluate the effect of post-deposition annealing for physical properties tuning the films deposited at RT were annealed in Ar flow at 5 mTorr at temperatures 300°C and 500°C during 1 hour. Structural characterization was done using X-ray diffraction and reflectivity techniques. Static magnetic properties were measured at 300K using SQUID magnetometer. Broadband ferromagnetic resonance (FMR) measurements were carried out at room temperature using a coplanar waveguide connected to a vector network analyzer (VNA) in the frequency range 0.05-20 GHz [5].

Films were found to be polycrystalline with surface roughness ~0.6 nm for T_s=RT and 300°C, while >2 nm for 500°C. The lattice parameter of cubic structure negligibly varies with T_s from 0.573 nm (RT) to 0.752 nm (500°C). These values correspond well to the bulk HA. Average grain size increase from ~10 nm to ~15 nm, respectively. Diffraction line <111> characteristic for ordered L2₁ phase was observed for elevated T_s. Saturation magnetization increase with T_s from ~700 emu/cc (RT) to ~900 emu/cc (500°C) with coercive field showing minimum ~2 Oe for T_s=300°C and peaking at 65 Oe for T_s=500°C. Similar variation was found for α : ~0.004 and ~0.05, respectively. Low α value is consistent with predicted for half-metallic compounds with a gap in minority density of states on Fermi level. Perpendicular standing spin wave mode was detected above the uniform resonance mode for all range of FMR frequencies for T_s=RT and 300°C. Exchange stiffness was estimates as 6.8 pJ/m (RT) and 9.8 pJ/m (300°C). These values a slightly lower than reported in [6]. Annealing cause similar variations of physical properties but is less effective comparing to elevated T_s.

These show the properties of CFG HA films could be effectively tuned in broad range to make them suitable for specific spintronics applications.

- [1] N. V. Uvarov et al., J. Appl. Phys. 112, 063909 (2012)
- [2] S. M. Ryabchenko et al., J. Phys.: Cond. Mater 25, 416003 (2013)
- [3] A. N. Pogorily et at., AIP Adv. 7, 055831 (2017)
- [4] T Kubota et al., Appl. Phys. Lett. 94, 122504 (2009)
- [5] A. S. Sivla et al., AIP Adv. 9, 035163 (2019)
- [6] A. Heggache et al., Appl. Phys. Lett. 104, 252412 (2014)

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Micromagnetism of Co/Cu multi-layered cylinders observed by electron holography

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The study of the new forms of magnetic states in magnetic cylinders has recently attracted much attention [1]. However, multi-layered structures in cylindrical geometry, which are basic blocks for spin torque nano-oscillators for example, have been poorly studied up to now. Among <u>others</u>, one of the main challenge for such studies is the experimental observation of magnetic configurations below 100 nm.

In this work, we used off-axis electron holography to observe the remnant magnetic states of Co/Cu multilayered nano-cylinders with a spatial resolution down to 2 nm. Nanocylinders are grown by electrodeposition in commercial polycarbonate membranes. Their diameters range between 70 and 90 nm. In order to study the effect of the aspect ratio and dipolar coupling on the magnetic states, four series of samples with different nominal Co and Cu thicknesses have been realized, namely $Co_{25 nm}/Cu_{15 nm}$, $Co_{25 nm}/Cu_{45 nm}$, Co_{50} $_{nm}/Cu_{50 nm}$ and $Co_{100 nm}/Cu_{100 nm}$. The magnetic configurations are recovered from direct qualitative and quantitative comparison between the experimental magnetic phase shift image obtained by holography and the one calculated from micromagnetic simulations (OOMMF). These ones are performed including the geometrical properties of each individual layers deduced from EFTEM experiments.

With our method, we have observed in the different wires either uniform magnetic states with antiparallel coupling or vortex states with different orientations of the core with respect to the wire axis [2,3]. In addition, different magnetic states can coexist within the same nanowire. Such a wide variety of magnetic configuration arises from the low values of the Co magnetic constants with respect to bulk, typical of electrodeposition process in single bath. To explain our results, we have calculated the phase diagram of the remnant states in a single disc for different orientations of the crystal anisotropy and saturation field. We show that the occurrence of each configuration depends on the aspect ratio of the layers, the direction of magneto crystalline anisotropy, and, in some cases, the interlayer dipolar coupling. In particular case, we will show that information on the preferential orientation of the crystalline anisotropy can be recovered from the statistics of occurrence of each magnetic configurations in the multilayer.

Finally, we discuss the interest of 3D holographic vector field electron tomography for nanomagnetism studies. Preliminary results performed on the $Co_{25 nm}/Cu_{15 nm}$ cylinders demonstrates that it allows getting the exact shape of each layer and the 3D components of the magnetic induction in each layer. This avoids geometrical uncertainties in micromagnetic simulations and solves the problem of 2D projection in off-axis electron holography experiments, leading to a much better determination of the magnetic constant of the layers.

- [1] A. Fernandez-Pacheco et al., Nat. Commun. 8, 15756, 2017
- [2] D. Reyeset al., Nanoletters 16, 1230, 2016
- [3] N. Biziere, et al., J. Appl. Phys. 126, 163906, 2019

Out-of-plane magnetic anisotropy in GadFeN/AlxGa1-xN heterostructures

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Phase-separated magnetic semiconductors containing magnetic nanostructures are relevant systems for the realization of high-density magnetic recording elements. It has been previously shown, that doping of GaN with Fe above a cation concentration of 0.4% leads to crystallographic phase separation and to the self-assembly of Fe-rich nanocrystals (NCs) with stoichiometry and magnetic characteristics imposed by the growth conditions [1,2]. When employing the α -growth mode during Fe doping of GaN (GadFeN), single-phase face-centered cubic γ' -GayFe_{4-y}N NCs embedded in the GaN matrix are stabilized, and their shape, size and density can be adjusted by fine tuning the growth parameters during a metal-organic vapour phase epitaxy (MOVPE) process [3-5]. Here, in Ga_dFeN/Al_xGa_{1-x}N (0 < x < 0.41) heterostructures grown by MOVPE on *c*-sapphire substrates, the effect of strain - tailored by varying the Al content - on the magnetic and structural properties of the Fe-rich embedded NCs is presented. Already for x < 5% the structural properties - phase, shape, orientation - as well as the spatial distribution of the embedded NCs are modified in comparison to those of the NCs stabilized in Ga_d FeN/GaN. The addition of Al to the buffer, promotes the formation of predominantly prolate hexagonal ε Fe₃N NCs, leading to a change of the magnetic easy axis from in-plane for x = 0 to out-of-plane for all layers grown on the Al_xGa_{1-x}N buffers with x > 5%. This contributes to a sizeable perpendicular magnetic anisotropy that opens wide perspectives for perpendicular recording in these systems.

[4] A. Grois et al. Nanotechnology 25, 395704 (2014)

^[1] A. Bonanni et al., Phys. Rev. B 75, 125210(2007)

^[2] A. Navarro-Quezada et al., Phys. Rev. B 81, 205206 (2010)

^[3] A. Navarro-Quezada et al., Appl. Phys. Lett. 101, 081911 (2012)

^[5] A. Navarro-Quezada et al. Crystals 9, 50 (2019)

Probing the origin of ferromagnetic stability of LSMO/SRO

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Optimally doped manganite (La_{0.67}Sr_{0.33}MnO₃, LSMO) is a perspective ferromagnetic material because of colossal magnetoresistance and T_c around room temperature. The use of LSMO is restricted due to the existence of magnetic dead layer in ulta-thin films. However, in proximity with SrRuO₃ (SRO) layer LSMO remains magnetic down to 1-2 u.c. In this work we investigate the origin of the ferromagnetic stability of LSMO in LSMO/SRO bilayers by using resonant x-ray spectroscopy. We have probed the orbital anisotropy and magnetism of LSMO on bilayers of SRO/LSMO deposited on TiO₂ terminated SrTiO₃ (STO) and compared to STO/LSMO, with varying thickness of LSMO (2/4/8/15 u.c.)/SRO (3/20 u.c.)/STO. Magnetic switching of LSMO at the interface with 20 u.c. SRO was observed even below critical thickness of LSMO. Moreover, 4 u.c. of LSMO shows remanence above SRO T_c. The orbital occupation probed by x-ray linear dichroism (XLD) evidences a preferential d_{322-r2} occupation of Mn at LSMO/SRO interface that confirms theoretical prediction. The factors leading to the magnetic stability of LSMO interfaced with SRO together with simulations of the XLD spectra will be discussed in view of our experimental results.

Ga+ ions irradiation driven Dzyaloshinskii-Moriya interaction and magnetic anisotropy in Pt/Co/Au trilayers

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The aim of this work is to study the evolution of Dzyaloshinskii-Moriya interaction (DMI) and magnetic anisotropy modification by heavy ion irradiation in Pt/Co(d=3nm)/Au(top) structures with asymmetric interfaces and in-plane magnetization in as-deposited state. In contrast to mostly investigated samples grown by sputtering, our systems were fabricated by molecular beam epitaxy, which offers a higher quality of interfaces. Recently, DMI appearing in ultrathin layered structures with asymmetric interfaces have been intensively studied (in samples created mainly by sputtering technique) because of very promising applications connected with skyrmions [1]. We discuss the influence of Ga+ ion irradiation with energy of 30 keV, in a wide range of ion fluences, F. Magnetic anisotropy modifications induced by ion bombardment, with energies of the same order have been reported in numerous papers, e.g. [2, 3]. DMI changes induced by ion irradiation with low energy (few hundred eV) have been recently reported in sputter deposited symmetric Pt/Co/Pt [4] and asymmetric Ta/CoFeB/MgO [5] thin films with perpendicular magnetic anisotropy (PMA). Irradiated films were studied using polar magnetooptical Kerr effect based magnetometry and Brillouin Light Scattering (BLS) techniques. From BLS measurements difference in Stokes and anti-Stokes peaks frequency Δf (connected with DMI) was observed even for non-irradiated sample. As a function of ion irradiation fluence one may observe: (i) increase of magnetic anisotropy with final appearance of PMA for F^{3} *10¹⁵ ions/cm²; (ii) decrease of Δf with final vanishing for $F^{4*10^{15}}$ ions/cm². Fluence dependence of magnetic anisotropy was explained by strain creation in Co layer and later by Co nanodisc formation [6], while DMI modification by Pt/Co interface mixing [5]. The amplitudes of the Stokes and anti-Stokes peaks significantly increase as an ion irradiation fluence increases what is correlated with the fluence induced enhancement of magnetooptical effects [2].

- [1] A. Fert, et al., Nature Mat. 2, 17031 (2017) and references therein
- [2] A. Maziewski, et al., Phys. Rev. B 85, 054427 (2012) and references therein
- [3] P. Mazalski, et al., J. Synch. Rad. 22, 753 (2015)
- [4] A. L. Balk, et al., Phys. Rev. Lett. 119, 077205 (2017)
- [5] L. Herrera Diez, et al., Phys. Rev. B 99, 054431 (2019)
- [6] M. Sakamaki, et al., Phys. Rev. B 94, 174422 (2016)

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Effect of shape and exchange anisotropies, and process parameters on macroscopic magnetization for a quartz MEMS micro-sensor

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Several MEMS-based magnetometers were conceived over the last decade, as they are well suited for small size and low power consumption requirements. Lately, new applications like inertial navigation or micro UAVs (Unmanned Aircraft Systems) ask for even better resolutions than the miniature magnetometer (AMRs and MEMS magnetometers) currently available on the market. With the aim of a 10 nT/vHz resolution and a power consumption of 1 mW, a MEMS based magnetometer has been designed to deal with today's requests at the ONERA lab [1].

ONERA's magnetometer is a resonant MEMS with a quartz-based structure, on which magnetic materials are deposited with a magnetron-sputtering equipment. Its development has two distinct objectives: -getting a high quality factor for the resonant structure and —ensuring the highest stable magnetization around earth magnetic field, meaning that its value and direction is poorly sensitive to magnetic shocks. The paper will focus on this last critical feature because it impacts the bias and the scale factor of the magnetometer [1].

In order to improve the magnetization stability, we decided to take advantage of the exchange bias phenomenon that takes place between ferromagnetic and antiferromagnetic thin layers. Its most well-known effects on magnetic properties in nanostructures are the hysteresis loop shift H_{ex} and the increase of the coercive field H_c after a field cooling from above the Néel temperature [2]. Shifting adequately the hysteresis loop would give at null field a unique magnetization equal to its saturation magnetization, while favouring shape and exchange anisotropies along the same axis would allow to pin its direction.

 $Fe_{65}Co_{35}$ and $Ni_{50}Mn_{50}$ have been used as, respectively, the FM and AFM materials. Their deposition process as a stack of thin layers will be introduced. Furthermore, several parameters have been studied to enhance the exchange field H_{ex} , the stack adherence and the compatibility with MEMS etching processes:

(i) Presence of patterned strips [*figure1*]; (ii) Numbers of bilayers FeCo/NiMn [figure2]; (ii) Type of field cooling procedure; (iv) Presence of an adhesion layer; (v) Thickness of FM AFM layers.





Figure 1 : Influence of number of strips on the magnetization for the Hard and Easy Axis with a "jump"

(Ta(20nm)/[NiMn(50nm)/FeCo(50nm)]₁₀/NiMn - 20mT cooling procedure – same distance between strips)

 $\label{eq:Figure 2: Ratio H_{ex}/H_c as a function of NiMn thickness and number 'n' of bilayers (no strip – 20mT cooling procedure - NiCr(20nm)/[NiMn('x' nm)/FeCo(100 – 'x' nm)]_n/NiMn(50nm))$

R. Levy, T. Perrier, P. Kayser, B. Bourgeteau, J. Moulin, Microsyst. Technol. 23, 3937 (2017)
 J. Nogués, J. Sort, V. Langlais, V. Skumryev, S. Suriñach, J.S. Muñoz, M.D. Baró, Phys. Rep. 422, 65 (2005)

Design of a spin filter device with free-standing graphene-based magnetic membranes

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Ultrathin free-standing magnetic layers are promising candidates to efficiently detect the spin of the electrons by means of the selective transmission of spin parallel or anti-parallel to a magnetization axis [1]. The capability to control the direction of this quantization axis joined to the engineering of a device able to guarantee spatial resolution allow to completely reconstruct the spin configuration of an electron beam. This could represent an outstanding tool integrable in electron spectroscopy experiments, where the spin resolution may be added to the E-k mapping in Angle-Resolved Photoemission Spectroscopy (ARPES) [2]. This task was tackled first by defining a proper fabrication process to obtain a bidimensional matrix of freestanding membranes. Then, in order to guarantee a sufficiently high transmission of the electrons (larger than $3 \cdot 10^{-2}$), the film thickness was chosen not to exceed 10nm. A suitable heterostructure was designed, consisting in a mechanically robust layer, a buffer layer, the active magnetic layer and finally a capping layer. Graphene was chosen for the former aim (see Fig. 1a) and a variable number of monolayers (ML) was tested (from 2 to 8 ML). Also the buffer layer was selected according to its mechanical properties and measured with Atomic Force Microscope (AFM) to infer information about the Young modulus and the film roughness (see Fig. 1b). The desired direction of the magnetization was defined using different magnetic materials (or stacks). Co was chosen for in-plane (IP) configuration, while Ta/CoFeB/MgO for the out-of-plane (OOP) one. The magnetic properties were measured both on micrometric patterned structures on rigid substrates by Vibrating Sample Magnetometer and Kerr microscopy and on ultrathin membranes by Faraday effect. The key features of sufficiently high coercive field and magnetic remanence are obtained (see Fig. 1c). The final aim is the characterization of the spin-filtering efficiency exploiting a spin polarized electron beam at low energy (0-30 eV) produced by a GaAs photocathode with negative electron affinity and recording the transmission of the membranes for different orientations of the beam polarization [3]. In this way we plan to probe the Figure of Merit (FOM) of the device and to infer the Sherman function, i.e. the asymmetry factor,

which should be larger than 0.5.



Figure 1 : a. Scanning Electron Microscopy image of a suspended graphene membrane; b. AFM topography of the free-standing magnetic stack; c. Magnetic hysteresis loop measured probing the Faraday effect for the OOP configuration of the magnetization.

- [1] Oberli D. et al., Phys. Rev. Lett. 81, 4228 (1998)
- [2] Övergaard T. et al., arXiv:1709.03838v3
- [3] Cantoni M. and Bertacco R., Rev. Sci. Instrum. vol. 75(7), 2387 (2004)

Investigation of interfacial Dzyaloshinskii-Moriya interaction and ferromagnetic resonance of MBE grown W/Co/Pt heterostructures

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The interfacial Dzyaloshinskii-Moriya interaction (iDMI) [1,2] is responsible for the chiral magnetic spin structure creating like vortex or skyrmions which are potential candidates for data storage in thin film technology. Here, we show the iDMI at the interface of heavy metal (Pt and W) and magnetic material (Co) with epitaxial growth of W(10A)/Co(6A)/Pt(10A) heterostructure. The iDMI strength has been calculated which approximately equals to 1.97 mJ/m² by K_{eff} method with considering magnetic exchange constant A=11pJ/m [3]. Apart from this, also the strength of iDMI 1.87mJ/m² has been calculated by first principle calculation with fcc lattice structure of W/Co/Pt which agrees to our experimental value of iDMI. Further, the micromagnetic simulation has been performed to calculate the domain wall width. Results of simulations correspond to the MFM image of labyrinth domains at remanence. Estimated from micromagnetic simulation iDMI value agrees with the results obtained by other methods.

The investigations of magnetic dynamics have been performed by VNA-FMR and cavity FMR. We observed different types of resonances by exciting the sample in the presence of GHz frequency and static magnetic field. The gilbert damping has been calculated which is equal to 0.027 for the particular frequency 9.387 GHz. With increasing the magnetic field, from lower to higher, the two types of resonance occur which are interpreted as Kittle resonance and labyrinth domains resonance. Whereas, with the decreasing field the three types of resonance are observed i.e. Kittle and labyrinth domains resonance and we are expecting the third kind of resonance is due to the presence of iDMI which gives the chiral structure e.g. skyrmions which recently have been studied in the reference paper [4,5]. We also observe the dependence of resonance due to labyrinth domains upon the history of the magnetic field, whereas for other resonances such dependence is not seen.



- [1] Dzyaloshinskii I.E. JETP Lett., 5, 259-1262 (1957)
- [2] Moriya T. Phys. Rev., 91,10 (1960)
- [3] Legrand W. et al. Sci. Adv. 4 (2018)
- [4] arxiv.org/pdf/1802.03979.pdf
- [5] T. Schwarze T. et al. Nature Materials 14, 478 (2015)

Figure 1 : Magnetisation reversal and its corresponding magnetic domain at different applied magnetic field.

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The Effect of the Phase Structure on Superconductivity in TiZrHf-Sn-X (X = Fe, Cu, Ni, Nb) Alloys

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We investigated ternary TiZrHf, quaternary TiZrHfSn and pentary TiZrHfSnX (X = Fe, Cu, Ni, Nb) alloys, all containing titanium, zirconium and hafnium. XRD characterization revealed that TiZrHf sample crystallizes in a single-phase hcp crystal structure, while others form multi-phase systems of simple crystal structures, e.g. hcp, bcc, fcc and primitive tetragonal cell, and Mn₅Sn₃, Fe₃Sn, Fe_{1.67}Sn, HfNi₂Sn type intermetallic compounds respectively. All tin-containing alloys segregated in multiple phases, forming inhomogeneous dendritic microstructure. Moreover, inhomogeneity also appeared within individual phases due to the unideal mixing caused by interatomic interaction between different elements.

The focus of our research was the investigation of the phase structure effect on superconducting parameters. Below approximately 1 K in TiZrHf-Sn-Ni samples total drop, while in TiZrHfSnFe only partial drop of resistance value occur. The latter indicate the coexistence of superconducting and normal phase(s). The critical temperature of TiZrHfSnNb sample is the highest of all investigated samples (4.7 K). The heat capacity and the resistivity measurements of TiZrHfSnCu alloy exhibit no superconducting transition. Superconducting alloys had rather broad temperature range of the heat capacity peaks and declination in resistivity, indicating that Cooper pairs do not form instantly. Thus, superconducting phases are not homogeneous and have nanoregions that vary in critical temperatures.

For the 5-component TiZrHfSnNb sample, we performed magnetization measurements, from which we determined temperature-dependent lower critical field H_{c1}. The susceptibility in a low magnetic field reached $\chi = -1$ value below Tc, indicating total Meissner effect. We estimated the zero-temperature value of H_{c1} with linear extrapolation of the experimental data to be 35 mT.

Epitaxial Growth of Compensated Ferrimagnetic Mn2RuxGa Thin Films

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Zero-moment half-metallic materials are immune to magnetic fields and combine high spin-polarisation with high resonance frequency (>200 GHz). They may be the future of spintronics. Mn_2Ru_xGa (MRG) is an easy perpendicular axis ferrimagnet with two anti-parallel Mn magnetic sublattices that compensate at a given temperature T_{comp} due to their different temperature dependecies. It is a hybrid Heusler, falling between a full- X₂YZ and a half- XYZ Heusler structure.

The most promising aspect of this material is its high spin-polarisation even when fully compensated, i.e with zero magnetic moment. Many of the properties of MRG, such as its compensation temperature, are dependent on the strain and tetragonal distortion which can be controlled by changing the concentration x of Ru. Its close lattice matching to $\sqrt{2a}$ of MgO, makes it a prime candidate for spintronic devices, and a tunneling magnetoresistance value of 40% has already been demonstrated.

Typically the material is grown by sputtering, and is characterised by a degree of disorder. This typically results in a more disordered, B2-like structure. Here we demonstrate epitaxial growth of MRG with x=0.4-0.8 with low disorder by sputtering, in our new DCA-built Trifolium Dubium UHV cluster tool, where the in-plane lattice is matched directly by the MgO substrate. Substrate temperature during deposition is important, with a sweet spot in the range of 225-275 °C, likely a preference of Mn₂Ga to grow the C1_b structure rather than DO_{22} at these temperatures. The C1_b is then stabilised into a hybrid structure by the addition of Ru in the crystal lattice.

X-ray crystallography indicates a highly-ordered crystal, characterised by a low intensity ratio $S = [I_{(002)}/I_{(004)}]$; for x= 0.5, S(250 °C) is 0.0106. This compares with films grown at higher temperatures S(350 °C)=0.0190, and S(300 °C)=0.0169. It is also nearly an order of magnitude smaller than samples grown in a HV Shamrock tool, where S=0.0955.

A reciprocal space map of the (206) MRG peak demonstrates heteroepitaxy of the film and substrate, with the MRG in-plane lattice parameter being V2 times that of the substrate to within <0.2%, giving values for a_{MRG}=0.5959-0.5965 nm and c_{MRG} =0.6006-0.6035 nm, with the film relaxing more at higher *x* values. Within the range of *x*, the unit cell volume varies by less than 0.1%, ranging from 0.21405-0.21427 nm³, despite the density measured by XRR increasing from 7.65-8 gcm⁻³. This indicates the presence of vancies within the crystal as expected from the C1_b structure, and that Ru populates these vacancies. As *x* increases, the defect density increases, indicated by the decreasing Scherrer coherence length, L_{c \\x} = 42.92 nm and L_{c \\x} = 17.83 nm for *x* = 0.42 and L_{c \\x} = 14.19 nm and L_{c \\x} = 9.88 nm for *x* = 0.76.

Extraordinary Hall effect can be used to measure magnetotransport properties, which in MRG measures the contribution of primarily the 4_c sublattice. EHE was performed between up to 4 T ranging from 10 K to 300 K. A large Hall resistance, $\sim \pm 2 \Omega$ at 1 mA, and a high Hall angle of order 2%, indicates strong scattering and is typically taken as an indication of high spin-polarisation of the conduction electrons. Saturation is not possible at 4 T for a range of 100K around the compensation temperature.

Epitaxial growth of MRG allows for precise control over strain within the crystal, opening a wide workspace for different applications.
Following stochiometric evolution of high-quality transition metal oxide in real time and space

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In the last decades, transition metal oxides (TMO) are growing in importance in spintronics applications. The physical properties of these materials can be tuned by reducing the material to the nanoscale or incorporating it to a heterostructures. Considering the potential importance of these materials, it is essential to develop growth methods that allow to engineer high quality oxide-based epitaxial nanostructures with customizable functionalities but also tools to study and fully characterize these systems. Extremely high-quality oxide films can be obtained by depositing metals by molecular beam epitaxy on a hot substrate, while the sample is exposed to an oxidizing agent such as molecular oxygen. We have used this method to fabricate highly perfect spinel magnetic nanostructures: ultrathin islands of up to 100 μ m² with atomically flat surfaces and free from antiphase boundaries (APB). Indeed, the spinel islands (CoFe₂O₄ [1], Fe₃O₄ [2] and NiFe₂O₄ [3]) grow from single crystallographic nuclei and are thus APB-free. The extremely low defect concentration leads to a robust magnetic order and exceptionally large magnetic domains. The use of these materials in spintronics devices requires full control of the structure of these systems, which directly affects their magnetic properties. The stoichiometric variability of TMOs can be exploited as high versatility only if the composition is carefully controlled. In particular, at high temperatures, the stoichiometry can change during growth resulting in non homogeneous or non desired composition. Thus the need to acquire chemical maps during growth. In our previous works, mixed cobalt-iron[1], nickel-iron[3], and cobalt-nickel[4] oxides have been studied, combining LEEM observations during growth with the LEED and PEEM information obtained a posteriori. However, a surprising discrepancy has been observed in the first two cases: when measuring the composition of the final films, in all cases the ratio of the cations in the wetting layer and in the spinel islands differ from each other and from the deposition ratio. To measure the evolution of the surface composition in such a multicomponent growth front, we have developed a method to determine the composition at the nanometer scale in real time [5]. We can do this by sequentially recording photoemission microscopy images under illumination of x-rays with photon energies corresponding to the metal absorption edge (Co and Fe L3 in the present case). I will present the growth of mixed cobalt-iron oxides on Ru(0001) by high-temperature oxygenassisted molecular beam epitaxy monitored in real time and real space by x-ray absorption photoemission microscopy. The initial composition is a mixed Fe-Co(II) oxide wetting layer, reflecting the ratio of the deposited materials. However, as subsequent growth of three dimensional spinel islands nucleating on this wetting layer takes place, the composition of the oxide in the wetting layer changes as iron is transferred into the spinel islands (figure1).



Figure 1 : Right panel: estimated composition of different regions of the wetting layer and the spinel islands during growth at 1300 K.

- [1] L. Martín-García et al. Adv. Mater. 27, 5955, (2015)
- [2] S. Ruiz-Gómez, et al. Nanoscale 10, 5566 (2018).
- [3] A. Mandziak et al. Scientific reports 8, 1, (2018).
- [4] A. Mandziak et al. Sci. Rep. 9, 13584 (2019)
- [5] S. Ruiz et al. J. Chem. Phys. 152, 074704 (2020)

Artificial Magnetic Domains Without Domain Walls in Ferrimagnetic Tb/Co Multilayers Patterned by Ion Bombardment

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For many years, light-ion bombardment (IB) has been used for magnetic patterning of ferromagnetic and exchange-biased layered systems. Owing to this post-deposition modification method, it has been possible to fabricate heterostructures with locally modified magnetic properties (e.g. anisotropy, magnetization, coercive field). The possibility of magnetic patterning in layered systems exhibiting perpendicular magnetic anisotropy (PMA) is particularly attractive for applications in a new generation of magnetic hard disks. Here we show an innovative use of IB through photoresist masks on ferrimagnetic (Tb/Co) multilayers

exhibiting PMA and dominance of the Tb sublattice (Tb+): the fabrication of a two-dimensional lattice of Co dominated (Co+) areas with a switching field smaller than that of the protected Tb+ matrix ($H_s^{Co+} < H_s^{Tb+}$). In saturation ($H > H_s^{Tb+} > H_s^{Co+}$) such structure shows a mono-domain state, with a domain wall (DW) on the border between the Co+ and Tb+ areas (Fig. 1 a). After the magnetization reversal of Co+ areas, a multidomain state appears but no DWs are present on the Co+/Tb+ borders (Fig.1b). This reorientation of the magnetic configuration corresponds to the transition between states 1-2 (or 3-4) indicated on the hysteresis loop measured using a magneto-optical Kerr effect (Fig. 1c). Note that for the magnetization reversal from state 2 to 1, a much higher magnetic field is necessary than for the transition from state 1 to 2 (minor loop in Fig.1c). This indicates that the multi-domain state without DW (state2) is energetically more stable than the mono-domain state with DW (state 1) [1].



Figure 1 : Magnetic configurations of: effective magnetization (black symbols) and of Co sublattice (blue arrows) and Tb sublattice (red arrows) in mono-domain (a) and multi-domain (b) states. (c) full and minor hysteresis loops as measured with magneto-optical Kerr effect.

[1] Frąckowiak Ł. et al. Phys. Rev. Lett. 124, 047203 (2020)

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Enhanced exchange bias in FeF2/Ni by inserting non-magnetic atoms at the interface

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The exchange bias (EB) phenomenon appears in antiferromagnetic (AFM)/ ferromagnetic (FM) bilayers due to exchange interactions at the interface. The exchange coupling between pinned AFM spins and FM moments leads to a shift of the magnetization curve known as exchange bias field. In this work, we present an enhancement of the EB field by inserting non-magnetic metals (NM) between AFM and FM layers.[1] FeF₂/NM/Ni films were deposited by electron beam evaporation. A motorized blade was used to cover part of the sample during the NM metal deposition. This part is the reference FeF₂/Ni bilayer for comparison with the NM interface. A continuous movement of the blade controls the thickness of the NM layer on a wedged shape. The EB field was measured by magneto-optical Kerr effect along the wedge direction every two hundred microns. The results show an EB improvement for NM materials as Cu, Au, and Pd. This enhancement is only observed just at the beginning of the wedge, for ultra-thin NM layers around one or two angstroms thick. It is also remarkable that this magnetic system shows positive EB for high cooling fields. The EB enhancement was also seen for the positive EB case. Thus, the negative and positive EB dependences with the NM thickness mirror each other with a significant improvement of the EB field for ultra-thin NM layers.

[1] F. Torres et al. Nanoscale 9 (2017) 17074

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Synthesis of Can+1MnnO3n+1 thin films by Pulsed-Laser Deposition

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Room temperature magnetoelectric (ME) compounds are rare and high-quality artificially multilayered ones are in general difficult and costly to produce. In this respect, magnetic Naturally Layered Perovskite structures (NLP) with improper ferroelectricity [1, 2], such as the Ruddlesden-Popper calcium manganite compound $Ca_3Mn_2O_7$ [2], offer an alternative route to achieve non-expensive and high-performance room temperature MEs for information storage, sensors, and actuators or low power energy-efficient electronics. They allow exploring oxygen octahedra non polar rotations and cation site displacement to attain non-centrosymmetry. Additionally, due to their high sensitivity to lattice-distortions, their preparation in thin film form over crystalline substrates allows the manipulation of acentricity and enables the tuning of lattice, electric and magnetic interactions. However, for achieving this, the preparation conditions to obtain the envisaged $Ca_{n+1}Mn_nO_{3n+1}$ phases, with different n, need to be optimized.

As such, thin films of calcium manganate have been prepared by laser ablation with different oxygen pressures (from 10^{-3} mbar to 10^{-1} mbar), substrate temperatures (from room temperature to 730 °C), laser fluences (1-5 J/cm²) and different substrates (SrTiO₃ and Si). The objective is to study the deposition conditions that lead to phase separated films, CaMnO₃ and Ca_{n+1}Mn_nO_{3n+1} with n = 2, the improper ferroelectric Ca₃Mn₂O₇. Their structure was studied by X-ray diffraction (XRD). Their microstructure and morphology were examined by scanning electron microscopy (SEM), their chemical composition was analyzed with energy-dispersive X-ray spectroscopy (EDS) and their magnetic properties were measured with a SQUID magnetometer.

The X-ray diffraction results show that for oxygen pressures in the range $10^{-2} - 10^{-1}$ mbar, deposition temperatures of 600 °C to 730 °C and laser fluences from 1.7 J/cm² to 2.9 J/cm², the films are polycrystalline, stabilizing in the CaMnO₃ orthorhombic Pnma phase. The SEM measurements show dense films with smooth surfaces. The corresponding EDS measurements indicate a 1:1 atomic proportion in these samples.

On the other hand, by decreasing the oxygen pressure, increasing the temperature and increasing the laser fluence, the Ca/Mn proportion increases, so that for films prepared on SrTiO₃, at 730 $^{\circ}$ C, with 4 J/cm² laser fluence and ~10⁻³ mbar oxygen pressure, the Ca₂Mn₃O₇ (n = 2) phase is stabilized, as confirmed by XRD. The corresponding EDS analysis further gives a Ca/Mn atomic ratio of ~1.4-1.5:1, consistent with the presence of this phase.

The corresponding magnetic properties of the films, that are correlated with their stabilized phases and preparation conditions, will be presented.

- [1] P. Ghosez, J. -M. Triscone, Nature Materials 10, 269 (2011)
- [2] P. Rocha-Rodrigues, S. S. M. Santos, I. P. Miranda, G. N. P. Oliveira, J. G. Correia, L. V. C. Assali, H. M. Petrilli, J. P. Araújo, A. M. L. Lopes, Physical Review B 101, 064103 (2020)

Van der Waals Epitaxy of Multiferroic e-Fe2O3 Thin Films on Flexible Synthetic Mica

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 ϵ -Fe₂O₃ has received considerable attention in the last few years, due to its unique physical properties, including giant coercivity (~2 T at room temperature)^{1,2}, magneto-electric coupling³, room temperature multiferroicity⁴ and millimeter-wave ferromagnetic resonance⁵. To exploit these attractive properties in information technology devices, epitaxial films of ϵ -Fe₂O₃ have to be stabilized on suitable substrates, and, more importantly, a better understanding of the spin-lattice interactions in this oxide is necessary for controlling its magnetic anisotropy. Nevertheless, the progress towards this goal is undermined by the challenging epitaxial stabilization of ϵ -Fe₂O₃, so far only achieved in a limited number of substrates, with the presence of specific buffer layers, and with no real control over the film microstructure.

In this work, we demonstrate the epitaxial growth of ε -Fe₂O₃ thin films by pulsed laser deposition on synthetic mica substrates as a new example of the realization of the growth of 3D crystals on a 2D material via the so called van der Waals epitaxy⁶. The structure of the 50-nm films was characterized by X-ray diffraction and scanning transmission electron microscopy, revealing the (001) axis along the perpendicular direction to the film plane and an unusually large crystallinity (rocking curve FWHM= 0.29° for the (006) peak). The magnetic properties of 2D mica substrates were thoroughly studied in order to unambiguously evaluate the intrinsic magnetic characteristics of the ε -Fe₂O₃ films. To this aim, we quantified and subtracted the different magnetic components of mica. Our results indicate that the magnetic properties of ε -Fe₂O₃ thin films closely resemble those of their nanoparticle counterparts, characterized by an incommensurate to ferro- or ferrimagnetic transition at low temperatures and large coercivity (H_c ~6400 Oe) at room temperature. The flexible, transparent and cleavable nature of mica substrates constitutes a unique platform for understanding the spin-lattice coupling and magnetic anisotropy in this polar ε -Fe₂O₃ thin films.

- [1] Jin, B., et al., Adv. Mater. 2004, 16 (1), 48–51
- [2] Popovici, M., et al., Chem. Mater. 2004, 16 (25)
- [3] Kadlec, C., et al., Phys. Rev. B 2013, 88 (10)
- [4] Gich, M., et al., Adv. Mater. 2014, 26 (27)
- [5] Namai, A., et al., J. Am. Chem. Soc. 2009, 131 (3), 1170-1173
- [6] Bae, S. H., et al., Nat. Mater. 2019, 18 (6), 550–560

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Controlling interfacial phenomena in hybrid V2O3/Co bilayers

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By engineering artificially the surface/interface of hybrid systems it is possible to tailor their spin-dependent transport characteristics and merge in a single device the functionalities of their individual constituents. To this regard, Co/V_2O_3 bilayers could be considered as an archetypical hybrid system, where a metallic ferromagnetic (FM) film is growth onto a prototype transition metal oxide (TMO) V₂O₃, which exhibits a firstorder metal insulator transition (MIT) from a paramagnetic metallic to an antiferromagnetic (AFM) insulating phase when the temperature falls below 170 K. Besides, the MIT is accompanied by a structural phase transition (SPT) that takes place simultaneously [1], and can be tuned with external stimuli [2]. Thus, we would be provided with an additional degree of freedom in order to control the FM properties at the FM/TMO interface. A series of experiments based on low temperatures measurements have been carried out on FM/V_2O_3 bilayers, proving the existence of interfacial exchange-coupling effects and a peculiar rise of the coercive field [3]. However, there is not a general concordance between the observed interfacial phenomena. To shed light on this issue, by means of high-resolution vectorial-resolved magneto-optic Kerr effect (v-MOKE) experiments, we have studied the magnetic anisotropy landscape of Co/V_2O_3 bilayers, in which the Co layer has been grown on-purpose with a defined uniaxial magnetic anisotropy at room temperature. V-MOKE loops allow investigating the magnetization reversal pathways, the angular analyses give us detailed information about the effective magnetic anisotropy symmetry, while the temperature analyses provide direct information about magnetic phase transitions [4]. We have thus studied the effects of different magnetic anisotropy configurations. The latter are obtained in a controlled way by setting the magnetic orientation of the FM layer across MIT. The strong influence of the magnetic configuration in the heterostructure across the MIT transition become clear when comparing hysteresis loops measured by using different field cooling procedures, as shown in Figure 1. At low temperature (i.e., 50 K), the loops present an enhancement of the coercive field, an exchange bias and an asymmetric magnetization reversals, which are all proof of the interfacial exchange coupling effects that take place in the Co/V_2O_3 heterostructure [5]. Our findings demonstrate that we have an active control of the interfacial exchange phenomena in FM/TMO heterostructures, especially across MIT, which enable the possibility to develop novel magnetic devices with specific functionalities.



Figure 1 : Representative v-MOKE loops of a V203 (100nm)/Co (15nm) heterostructure above (a1,b1) and below (a2,b2) the MIT transitions using different FC conditions: **a**) Negative-FC close to the easy axis and **b**) Positive-FC around 50°C off the easy axis.

[1] Imada, M., et al., Rev. Mod. Phys. 70, 1039 (1998)

[2] Saerbeck, J., et al., J. Mater. Res. 29, 345 (2014).

[3] De La Venta, J., et al., Applied

Physics Letters 102, 122404 (2013).

[4] Jiménez, E. et al., Phys. Rev. B.

80, 014415 (2009)

[5] Camarero, J. et al., Phys. Rev.

Lett. 95, 057204 (2005)

Modulation of the spin structure in NiO by strain and spin-flop interaction

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While in the case of FMs a moderate external magnetic field can be used to align magnetic moments of ferromagnetic layers, a magnetic field is necessary to manipulate spin structure in AFMs are usually very large¹. However, magnetic field can be used to indirectly manipulate the direction of spins in AFM via interface coupling mechanism in FM/AFM bilayer. Besides interaction at FM/AFM interface, manipulation of spin structure in AFMs can be realized with a use of strain. Lattice distortion induced by strain can lead to modification of magneto-crystalline anisotropy and, in a consequence, reorientation of direction of magnetic moments in AFM layer.^{2, 3} In this study we used strain and interface coupling to manipulate spin structure in NiO embedded between Cr and Fe layer. To determine a spin structure in NiO we used the x-ray linear dichroism (XMLD). Figure 1(a) shows the L₂ ratio of NiO, defined as the ratio of the intensity of the lower energy peak to higher energy peak as a function of x-ray incident angle γ . The R_{L2} follows the theoretical angular dependence of the XAS spectrum. In agreement with previous studies at $\gamma=0^{\circ}$ we noted a maximum of L_2 ratio for Fe/NiO/MgO which proves that NiO spins possess a considerable out-of-plane component⁴. A minimum of R_{L2} observed for Fe/NiO/Cr indicates an in-plane NiO spin direction. Thus, we proved that while an out-of-plane spin direction can be observed for NiO layers grown on MgO substrate in Fe/NiO/MgO stack, a strain-induced spin reorientation transition from an out-of-plane to in-plane direction can be observed when NiO layers are deposited on Cr buffer which exerts compression strain on AFM layer. To determine an in-plane distribution of NiO spins in Fe/NiO/(Cr)MgO an XMLD measurements were collected as a function of azimuth angle, ϕ while the γ angle was set to 45° (Fig. 1(b)). Prior to XMLD measurements the sample was magnetized along easy Fe [100] (NiO [110]) direction. For Fe/NiO(14Å)/Cr we noted a well-pronounced extrema in $R_{L2}(\phi)$ dependence for NiO [110] and [1-10] directions, with a maximum for x-ray polarization parallel to the [1-10] NiO crystal axis ($\phi = 90^\circ$) (Fig. 1(b), black). Then, Ni²⁺ spins in Fe/NiO(14Å)/Cr are aligned parallel to NiO[1-10] axis and perpendicular to the spins of Fe. After switching the magnetization of Fe by 90° the NiO spins are rotated by 90°. Together with an increase in NiO thickness the in-plane anisotropy of NiO is weakened. For Fe/NiO(37Å)/Cr we noted almost no change in $R_{L2}(\phi)$ dependence (Fig. 1(b), blue). In the case of Fe/NiO/MgO we registered pronounced in-plane anisotropy (Fig. 3(d), red) which indicates that NiO spins in Fe/NiO/MgO are not aligned perfectly perpendicular to the surface, but possess small in-plane component. Our results demonstrate the feasibility of use strain and coupling with ferromagnet to manipulate spin structure in NiO.





[3] I.J. Park et al., Appl. Phys. Lett. 142403, 142403 (2019)[4] D. Alders et al., Phys. Rev. B 57, 11623 (1998)

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Structural and magnetic properties of FeCoNiAl0.25Mn0.25 high entropy alloy thin films grown by PLD

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High entropy alloys (HEA) are typically defined as solid solutions of 5 elements or more in equimolar ratios [1,2]. These materials have many attractive properties such as slow diffusion, mechanical hardness and excellent resistance to corrosion and oxidisation. Consequently, magnetic HEA in thin film form are of potential interest for technological applications in chemically harsh or high temperature environments. For example, magnetic HEA thin films could open up the possibility to develop spintronic devices such as high temperature spin-Seebeck energy harvesters.

Pulsed laser deposition (PLD) is a high energy deposition technique, which benefits from the relative ease of growth of complex multicomponent thin films via the stoichiometric transfer of material from the target to substrate. Consequently, PLD is an ideal technique for the deposition of HEA films. The energy of deposition can be controlled by introducing a ballast gas, typically argon, and offers a way to optimise film quality with respect to structural and magnetic properties.

It has been suggested that nano crystalline to amorphous HEA's may enhance some of the desirable properties described above [1]. Therefore, being able to control film crystallisation, while maintaining a single-phase homogeneous film, is critical to optimising HEA films for technological applications. Here, we investigate the influence of the deposition energy on the crystallisation and depth dependent magnetisation and density profiles of FeCoNiAl_{0.25}Mn_{0.25} HEA thin films, grown under a range of Ar gas pressures.

The structural and magnetic properties of these HEA films, have been investigated using x-ray diffraction and reflectometry (XRD, XRR), magneto-optical Kerr effect (MOKE), ferromagnetic resonance (FMR) and polarised neutron reflectometry (PNR). XRD results indicate that increasing the Ar pressure decreases the total film crystallisation, while the magnetic and structural depth sensitivity of PNR [3,4], has shown that the film appears to phase separate with respect to depth at high Ar pressures – suggesting that reducing the deposition energy results in multiple regions along the film depth with different magnetisations.

- [1] J.-W. Yeh et al, Adv. Eng. Mater, 6, 299–303 (2004)
- [2] D. B. Miracle and O. N. Senkov, Acta Materialia, 122, 448–511 (2017)
- [3] J.F. Ankner, G.P. Felcher, J. Magn. Magn. Mater, 200, 741-754 (1999)
- [4] S. J. Blundell and J. A. C. Bland, Phys. Rev. B, 46, 3391 (1992)

Design study of the magnetic and structural properties of CoFeNiCrAl thin films

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The search for new soft magnetic thin films has led to the exploration into multi-component alloys (MCAs), which contain four or more elements with percentages between 12.5 and 33%. These alloys when in equimolar ratios are known as high entropy alloys (HEAs), and can be fabricated to have a single phase [1]. The resulting alloys often have interesting functional properties, which are linked to their structure [2, 3]. This work presents a design study of the CoFeNi_{0.5}Cr_{0.5}Al composition in thin films. It investigates how the fabrication parameters (growth power and time) changes the morphology and composition of the films, along with the functional magnetic properties.

The films were grown using RF sputtering, to give the composition CoFeNi_{0.5}Cr_{0.5}Al, as in bulk samples FeCr nanoparticles form in a CoNiAl matrix [3]. The growth parameters were: 0.7 Pa Ar pressure, 150W or 200W power, for 10, 20 or 40 mins. The films thicknesses were in the range 160nm (150W, 10 mins) to 700nm (200W, 40 mins). They were characterised using x-ray diffraction (XRD) to determine the phase and morphology, MOKE magnetometry to determine the anisotropy and magnetostriction constant, SQUID magnetometry to determine the saturation magnetisation and Curie Temperature, ferromagnetic resonance (FMR) to determine the damping constant and polarised neutron reflectivity (PNR) to determine number of magnetic phases within the films.

From the XRD data, it was determined that as the thickness increased the texture and phase changed, from amorphous+intermetallics (10 mins) to a FCC phase (20 minutes) to a BCC phase (40 mins). From the MOKE data, the coercive field (H_c) was strongly influenced by the structure, as the thinner and thicker films all had $H_c < 500$ A/m, while for the 20 mins films H_c was a factor 3 higher which is likely to be due to the FCC phase within these films. The magnetisation increased with increasing film thickness. As the films are thicker than 50nm, it is unlikely that surface/interface effects dominate, rather the different structural phases within the films will influence the total magnetisation. Such that the thinner films with the different non-magnetic intermetallic phases with an amorphous magnetic phase have a lower magnetisation compared to the thicker polycrystalline films with the BCC phase. From the PNR, the 160 nm (150W 10mins) film had one magnetic phase, while the 200nm (200W 10 mins) film had two magnetic phases. This was confirmed using FMR measurements. The damping constant (200 of the films also varied with film thickness and phase, with 20 being between 0.01 (150W 40 mins) and 0.039 (150W 20 mins).

In conclusion, CoFeNi_{0.5}Cr_{0.5}Al thin films are promising candidates for soft magnetic films, with $H_c < 500$ A/m and magnetisations ~ 500kA/m. Also the growth conditions (power, growth time) influence the phase present in the film, which impacts the magnetic properties.

- [1] D. B. Miracle, et al., Acta Mat. (2017), 122, 448
- [2] F. Kormann, et al., Appl. Phys. Lett. (2015), 107
- [3] N. A Morley, et al. accepted by Sci Rep, (2020)

Oxidation effect on the interface magnetoelectric coupling in Co/Pb(Zr,Ti)O3(001)

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Recent research on spin electronics has been strongly influenced by the discovery of alternative means to control the magnetization other than by applying a magnetic field. Rapidly, the electric control of magnetism has appeared as the method of choice for this purpose due to its promising potential for spintronic applications with thrifty energy consumption. In this framework, artificial multiferroic materials such as ferromagnetic/ferroelectric bilayers, represent a natural pathway towards the efficient electric control of the magnetization, owing to the magnetoelectric coupling (MEC). The voltage control of magnetism through electronic processes is particularly promising for spintronic applications due to, *e.g.*, the possibility to electrically control two non-volatile magnetization states, the ultra-low energy consumption of the device, and the intrinsically fast response of the magnetization resulting from changes in the electronic structure. Therefore, electronically-driven interface MEC has attracted much attention lately. In particular, great efforts are being made to improve the functionality of magnetoelectric interfaces by searching for new mechanisms leading to enhanced coupling constants [1,2].

Based on first-principles calculations, we have recently predicted the polarization control of the interfacial magnetic phase and a giant electronically-driven magnetoelectric coupling (MEC) in Co/PbZr_{0.25}Ti_{0.75}O₃(001) (Co/PZT) due to the oxidation of Co at the interface [3]. The magnetic phase of the oxidized Co interface is electrically switched from the ferromagnetic to the antiferromagnetic state by reversing the PZT polarization from upward to downward, respectively. A surface MEC constant $\alpha_s = 2 \times 10^{-10} \text{ G cm}^2 \text{ V}^{-1}$ is derived from our calculations in unoxidized Co/PZT bilayers, in agreement with our previously reported experimental results [4], and a giant surface MEC constant $\alpha_s = 12 \times 10^{-10} \text{ G cm}^2 \text{ V}^{-1}$ is obtained in the case of oxidized Co/PZT interfaces. In this talk, I will discuss in details the changes of the electronic and magnetic properties occurring at the Co/PZT interface as a function of the polarization state, the Co thickness, and its oxidation state.

[1] G. Radaelli, et al., Nature Commun. 5, 3404 (2014)

[2] I. R. Reddy, et al., Phys. Rev. B 98, 140401(R) (2018)

[3] R. Arras and S. Cherifi-Hertel, ACS Appl. Mater. Interfaces 11, 34399 (2019)

[4] O. Vlasin, et al., ACS Appl. Mater. Interfaces 8, 7553 (2016)

Nanoscale manipulation of magnetic domains by strain-induced proximity

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Coupling of lattice and spin degrees of freedom without the use of external magnetic fields allows for energyefficient spintronic devices. In this context, hybrid materials composed of an oxide layer undergoing a firstorder, structural phase transition (e.g. V_2O_3) and a ferromagnetic layer (e.g. Ni) coupled by proximity through the inverse magnetoelastic effect offer a promising alternative to voltage-controlled magnetism or other switching mechanisms without a magnetic field. The ultimate magnetic domain manipulation is at the level of the nanoscale coexisting phases in the V_2O_3 layer.

Here we show by direct imaging of the thermal evolution of the Ni spin structure that the magnetic domains can be tuned in both size and anisotropy axis upon crossing the structural phase transition (SPT) of the proximal V_2O_3 layer [2]. We find a drastic temperature-driven reorientation of the Ni magnetic domains across the SPT which is responsible for the increase in the coercive field. We identify a Ni domain axis rotation across the SPT of V_2O_3 that is well explained by both the angular dependent ferromagnetic resonance and micromagnetic simulations. Direct observations of the lateral correlation length of the Ni domains show an increase of almost an order of magnitude at the SPT compared to room temperature as well as a broad spatial distribution of the local transition temperatures. This corroborates the phase coexistence of Ni anisotropies due to the V_2O_3 SPT. Our data reveal that the reorientation of Ni domains is controlled by the reconfiguration of the structural domains of the oxide layer at the SPT, due to strain induced proximity [2]. Our findings reveal a novel pathway to control magnetic domains without a magnetic field through proximity to a material undergoing a first-order structural phase transition, an approach that may allow for novel device concepts based on "straintronics".



Figure 1 : (top) Representative XMCD-PEEM images recorded at temperatures below (150 K), across (163 K, 170 K) and above (200 K) the SPT of V₂O₃. The insets show their Fourier transforms. **(bottom)** Histograms of the rotation of the Ni magnetization, $\Delta\Phi$, with respect to the initial high-temperature saturated state.

[1] de la Venta, J. et al., Appl. Phys. Lett. 102, 1 (2013); Appl. Phys. Lett. 104, 062410 (2014)

[2] Valmianski, I. et al, submitted.

[3] Mc Leod, A.S., et al., Nat. Phys. 13 80 (2017); Gilbert, D. A., et al., Sci. Rep. 7, 13471 (2017)

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Ferromagnetic insulator (Ga,Mn)N - influence of local substrate temperature on Mn incorporation and magnetic properties

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(Ga,Mn)N is an important follower of (Ga,Mn)As in the diluted magnetic semiconductors family. From one side a ferromagnetic guise of GaN would constitute a major technological advance due to the already dominating role of group III nitrides in photonics and hig power electronics. From the other, the superexchange driven ferromagnetism is realized in an insulating host opening new avenues for device architectures in which dissipation-less information transfer could be realized in a combination with piezoelectricity of this wurtzite structure material. The driving effect is the ability of a direct manipulation of the single-ion magnetic anisotropy specific to Mn^{3+} ions through electric-field-generated uniform strain has been shown possible [1]. Therefore, it remains timely and important to continue the effort to further optimize the growth conditions aiming at a further increase of Mn incorporation and so of the magnitude of the Curie temperature (T_c), while maintaining the single phase, insulating character and macroscopic lateral Mn homogeneity along the wafer.

In this report we summarize the results of studies quantifying the role of the growth temperature T_g on the magnetic properties of (Ga,Mn)N epilayers. Our studies are augmented by a systematic structural and microstructure characterization which do not reveal any crystallographic phase separation, clusters or nanocrystals, even at the lowest T_g [2]. It has been found that a typical 10 °C variation of T_g across 1 inch substrate can lead to 40% dispersion of T_c . The established here strong sensitivity of T_c on T_g turns magnetic measurements into a very efficient tool providing additional information on local T_g , an indispensable piece of information for growth mastering of ternary compounds in which metal species differ in almost every aspect of their growth related parameters determining the kinetics of the growth.

We also employ resonantly enhanced UV Raman scattering [3] to precisely assess Mn incorporation homogeneity within the (Ga,Mn)N wafer. Our Raman scanning clearly confirms substantial lateral distribution of Mn atoms across the layer, which is radial with respect to its center. From the established distributions in two deliberately chosen layers the magnitude of the optimal growth temperature for most efficient Mn atoms incorporation in epitaxial GaN has been confirmed. It is shown that the combination of the 1LO linewidth and its energy provides assessment of the crystalline quality of the investigated layers.

- [1] D. Sztenkiel, et al., Nat. Commun. 7, 13232 (2016)
- [2] K. Gas et al., J. Alloys Compd. 747, 946 (2018)
- [3] K. Gas, D. Hommel, M. Sawicki, J. Alloys Compd., 817, 152789 (2020)

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Metamagnetism in self-assembled FeRh nanomagnets

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The chemically ordered FeRh alloy undergoes a first-order metamagnetic phase transition from antiferromagnetic (AF) to ferromagnetic (FM) order just above room temperature. The transition is accompanied by a volume increase and a reduction in resistivity, resulting from coupled structural, magnetic, and electronic order parameters. The option to drive the transition via multiple external forces and the large changes in magnetization, magnetoresistance, and entropy make this material interesting for magnetic recording, spintronics, and magnetic refrigeration.

While FeRh has been extensively investigated in the bulk and thin-film geometries, the metamagnetic properties of FeRh nanostructures have only been studied recently. For instance, lithographically patterned FeRh stripes show emergent behavior such as a giant phase transition asymmetry and supercooling [1]. On the other hand, self-organization via Volmer-Weber growth has also been utilized to fabricate FeRh nanoisland arrays supported on a substrate. However, it was found that the nanoislands display FM ordering down to very low temperatures and cannot generally support the AF phase, such that metamagnetism is mostly suppressed in these systems [2,3].

Here we show different self-assembly routes for the fabrication of FeRh nanomagnet arrays on single-crystal substrates. The morphology and magnetic properties of the samples are investigated using atomic and magnetic force microscopy, as well as magnetometry. While we confirm that Volmer-Weber nucleation of islands during growth leads to FM-stabilized objects, we have also explored an alternative self-assembly route based on solid-state dewetting of FeRh films, i.e., the thermodynamically driven spontaneous agglomeration of thin films into three-dimensional islands [4]. We control the nanoisland size and shape by tuning the deposition conditions and the epitaxy between the film and substrate. Besides, we find that the experimentally retrieved morphology and crystal faceting of FeRh islands are well reproduced via simple thermodynamic modeling based on surface and interface energy values.

Most importantly, we found that FeRh nanomagnets assembled via solid-state dewetting fully preserve the metamagnetic phase transition even for island sizes in the ~100 nm scale. This striking difference to nanoislands formed via Volmer-Weber nucleation is linked to distinct island morphology and crystallographic faceting obtained under each assembly route. While this strong interplay between morphology and magnetic order has been theoretically suggested [3], our experiments provide clear evidence for this behavior.

Furthermore, we will briefly discuss the way to obtain metamagnetic, free-standing FeRh nanoparticles by releasing the nanoislands from the substrate. Comparing the phase transition characteristics of the separated nanoislands to the supported ones gives further insight into the nanoscale properties of FeRh. We believe that these results can significantly motivate the utilization of nanoscale FeRh objects in several technological domains.

- [1] V. Uhlíř, J. A. Arregi, and E. E. Fullerton, Nat. Commun. 7, 13113 (2016)
- [2] M. Loving et al., J. Phys. D: Appl. Phys. 46, 162002 (2013)
- [3] M. Liu et al., EPL 116, 27006 (2016)
- [4] C. Thompson, Annu. Rev. Mater. Res. 42, 399 (2012)

Experimental Evidences of <100>/<110> in Plane 4-fold Anisotropy Switching in (Ga,Mn)As

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Historically, comprehensive studies of dilute ferromagnetic semiconductors, e.g., p-type (Cd,Mn)Te and (Ga,Mn)As, paved the way for a quantitative theoretical description of effects associated with spin-orbit interactions in solids, such as crystalline magnetic anisotropy. In particular, the theory was successful in explaining uniaxial magnetic anisotropies associated with biaxial strain and non-random formation of magnetic dimers in epitaxial (Ga,Mn)As layers. However, the situation appeared much less settled in the case of the cubic term: the theory [1] predicts switchings of the easy axis between in-plane <100> and <110> directions as a function of the hole concentration and/or temperature, whereas only the <100> orientation has been found experimentally. Here, we report on the first experimental observation of such easy axis switchings - evidenced by extensive magnetization and ferromagnetic resonance studies on a series of highcrystalline quality (Ga,Mn)As films [2]. Our findings are described by the mean-field p-d Zener model [1] augmented with three new ingredients [2]. The first one is a scattering broadening of the hole density of states, which reduces significantly the amplitude of the alternating carrier-induced contribution. This opens the way for the two other ingredients, namely the so-far disregarded single-ion magnetic anisotropy and disorder-driven non-uniformities of the carrier density, both favoring the <100> direction of the apparent easy axis. Accordingly, when the disorder gets reduced a switching to the <110> orientation is possible in a certain temperature and hole concentration range.

[1] T. Dietl, H. Ohno, and F. Matsukura, Phys. Rev. B 63, 195205 (2001)

[2] M. Sawicki, O. Proselkov, C. Sliwa, P. Aleshkevych, J. Z. Domagala, J. Sadowski and T. Dietl, Phys. Rev. B 97, 184403 (2018)

Magnetic properties of wurtzite (Ga,Mn)As

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(Ga,Mn)As, the canonical example of dilute ferromagnetic semiconductor (DFS), has attracted a great deal of attention since the turn of the centuries, becoming the flagship and assuming the role of the main testbed for semiconductor spintronics. To these ends (Ga,Mn)As has been obtained in planar form in the zinc-blende structure. An alternative way, by adopting the core-multishell concept, allowed to obtain (Ga,Mn)As in hexagonal (wurtzite) structure accommodated in 1D nanowires (NW) form [1], where a careful design of the NW layout structure and the associated strain engineering led to a huge, at least 30%, enhancement of the ferromagnetic phase transition temperature in strained MnAs nanocrystals formed in the (Ga,Mn)As shells upon high temperature annealing [2].

Here we report on detailed magnetic studies of the magnetic properties of the wurtzite (Ga,Mn)As cylindrical shells. Ga_{0.94}Mn_{0.06}As shells have been grown by molecular beam epitaxy (MBE) at low temperature as the part of multishell cylinders overgrown on wurtzite (Ga,In)As NWs cores synthetized on GaAs (111)B substrates. HRTEM morphology and structure nano-characterization fully confirmed the designed constitution of the NWs [2]. To obtain a clean signal in the magnetic studies, the NWs have been separated at cryogenic temperatures from their substrate and seeding layer by embedding them in a film of PMMA, an e-beam lithography resist. Our elaborated magnetic studies clearly indicate the presence of a low temperature ferromagnetic coupling, which despite a reasonably high Mn contents of 6% is limited only to below 40 K. It is however clearly shown that this coupling is maintained only in separated mesoscopic volumes resulting in an overall superparamagnetic-like behavior. This superparamagnetism gets blocked below \sim 20 K, and so it mimics deceivingly a uniform low temperature ferromagnetism in this material. It is also shown that about 1/3 of the total magnetic moment exerted form the material comes from noninteracting Mn ions, a fact previously not accounted by the magnetic studies. Both these features are assigned to a relatively low concentration of holes in the (Ga,Mn)As layer and the resulting dominant role of the fluctuations in the local density od states [3]. In a broader view our results constitute an important contribution to the still ongoing dispute on the true and the dominant form(s) of the magnetism in this model DFS material.

[1] A. Siusys, J. Sadowski, M. Sawicki, S. Kret, T. Wojciechowski, K. Gas, W. Szuszkiewicz, A. Kamińska, T. Story, Nano Lett. 14, 4263 (2014)

[2] A. Kaleta, S. Kret, K. Gas, B. Kurowska, S. B. Kryvyi, B. Rutkowski, N. Gonzalez Szwacki, M. Sawicki, and J. Sadowski, Nano Lett. 19, 7324 (2019)

[3] L. Gluba, O. Yastrubchak, J. Z. Domagala, R. Jakiela, T. Andrearczyk, J. Zuk, T. Wosinski, J. Sadowski, and M. Sawicki, Phys. Rev. B 97, 115201 (2018)

Electric field manipulation of magnetization in (Ga,Mn)N layers through piezoelectromagnetic coupling

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An electric field-control of magnetic anisotropy is a promising candidate for building new device functionalities with ultra-low energy consumption. We want to report on this effect in Ga_{1-x}Mn_xN-based structures, in which the magnitude and the sign of magnetic anisotropy can be changed by appropriate material engineering and by an electric field. Recently, we have shown that a strong coupling between piezoelectricity and magnetism exists in paramagnetic state of Ga_{1-x}Mn_xN with x = 0.025 [1]. In these piezoelectric systems a voltage *V* applied across the crystal elongates it and thus deforms the crystal field which surrounds the magnetic ions and so modifies their magnetic anisotropy. Now, by using molecular beam epitaxy grown structures we investigate the magnetoelectric effect in Ga_{1-x}Mn_xN with x = 0.06 at T = 2 K in the ferromagnetic state. We observe an electric field-induced decrease of coercivity and a non-reversible magnetization switching for magnetic fields close to the coercive field. The data is precisely modeled using atomistic spin approach [2] applied to an ensemble of about 10000 Mn spins within the frame of Landau-Lifshitz-Gilbert description of the precessional motion of magnetization. In numerical simulations we allow for the electric field control of the magnitude of uniaxial (along the wurtzite c axis) anisotropy parameter K_{U} , that in turn affects the effective magnetic field H_{eff} acting on Mn ions. Our results open the route for the repetitive magnetization switching in Ga_{1-x}Mn_xN thin films using sub-nanosecond voltage pulses.

D. Sztenkiel et al., Nature Comm., 7, 13232 (2016)
 R. F. L. Evans et al., J. Phys.: Condens. Matter, Vol. 26, p. 103202 (2014)

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Localized magnetic phase transitions in B2-ordered alloy thin films

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Ferromagnetic onsets can be realized within alloy thin films through local lattice disordering, forming locally confined nanoscale magnetic regions embedded within non-ferromagnetic surroundings. Atomic displacements induced by the irradiation of ions can result in the generation of local ferromagnetism, emerging from a paramagnetic initial state in B2 $Fe_{50}Al_{40}$ [1] or an antiferromagnetic state in B2 $Fe_{50}Rh_{50}$ [2]. Lattice disordering through atomic displacements is associated with strain as well as changes to the densities of states, however a detailed characterization of lattice defects correlated with the ferromagnetic onsets is lacking. We deploy light noble gas ion irradiation to sensitively transform B2 $Fe_{50}Al_{40}$ and B2 $Fe_{50}Rh_{50}$ to their ferromagnetic states and directly probe their lattice disorder and corresponding ferromagnetic onsets. Insights into the respective processes have been obtained, for instance, the anisotropic geometry-dependent strain relaxation of locally disordered regions [3], and the crucial role of static disorder in B2 $Fe_{50}Rh_{50}$ resulting in ferromagnetic phase formation [2]. Further characterization of the lattice disorder is achieved by probing the open-volume defect distribution. Recent progress towards a detailed understanding of the mechanisms of lattice disorder leading to ferromagnetic onsets will be discussed.

[1] J. Ehrler et al., New J. Phys. 22, 073004 (2020)

[2] B. Eggert et al., RSC Adv. 10, 14386 (2020)

[3] M. Nord et al., Small 15, 52, 1904738 (2019)

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Study of phonons' and magnons' properties in (Ni80Fe20/Au/Co/Au) multilayers of different number of repetitions

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We have studied the properties of surface acoustic waves' [1] and spin waves' propagation in magnetic $(Ni_{80}Fe_{20}/Au/Co/Au)_N$ (3<=N<=12 - number of repetitions) multilayers. The multilayers were deposited on the naturally oxidized silicon substrate with a 30-nm-thick Au buffer layer. The experimental technique that we used was high-resolution Brillouin spectroscopy, which allows the investigation of both kinds of waves. The behavior of spin waves was studied in two experimental geometries: backward volume geometry (supporting the propagation of backward volume magnetostatic spin waves) and Damon-Eshbach geometry (supporting the propagation on surface magnetostatic spin waves) [2]. The thicknesses of permalloy ($Ni_{80}Fe_{20}$) and gold layers were 2 nm, while that of the cobalt layer was 0.8 nm [3,4]. The measurements were taken in different values of external magnetic field. The crossing of phonon and magnon dispersion relations has been investigated as well. In addition, we have performed simulations using the finite element method in order to obtain the theoretical dispersion dependencies.

A. Trzaskowska, S. Mielcarek, B. Graczykowski, F. Stobiecki, J. Alloys Compd. 517, 132 (2012)
 M. Zdunek, A. Trzaskowska, J. W. Kłos, N. K. P. Babu, S. Mielcarek, J. Magn. Magn. Mater. 500, 166429 (2020)
 M. Urbaniak, F. Stobiecki, B. Szymański, A. Ehresmann, A. Maziewski, M. Tekielak, J. Appl. Phys. 101, 013905 (2007)
 F. Stobiecki, M. Urbaniaka, M. Tekielak, B. Szymański, T. Luciński, M. Schmidt, A. Maziewski, J. Magn. Magn. Mater. 310, 2294 (2007)

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Control of magnetic anisotropy in Co-Ni nanowires self-assembled in SrTiO3

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Self-assembled vertically aligned nanocomposites (VANs), which consist of nanometer-sized nanowires embedded into a surrounding host matrix, have recently emerged as a novel playground for strain engineering of physical properties in thin films. In contrast to classical planar heteroepitaxial systems, VANs consist of two intertwined phases, coupled along their vertical interfaces. While so far, most studies have focused on multifunctional oxide/oxide systems, such as the archetypal perovskite-spinel BiFeO₃-CoFe₂O₄ VANs, metal/oxide systems can be obtained in a similar fashion, as we demonstrate in this contribution. Using $Co_xNi_{1-x}/SrTiO_3$ as a model system, we first present a detailed computational analysis of the PLD-based thin film growth mechanisms giving rise to VANs. We show that a careful choice of the synthesis parameters allows to modify the nanoarchitecture of the composite, which directly impacts the strain-state of the embedded metallic phase. We evidence how this permits to tune the magnetic anisotropy of the system, putting special emphasis not only on the huge mean axial strains and large magnetoelastic effects that can be achieved, but also on the important role played by local strain fluctuations. Finally, we complement our magnetometry measurements with an in depth x-ray magnetic circular dichroism (XMCD) analysis of the nanocomposite performed at the L_{2,3}-edges of Co and Ni and discuss the link between macroscopic straininduced magnetic anisotropy and the anisotropy of the atomic orbital magnetic moment.

Four-spin interactions in magnetic trimers

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Recently, a rapidly growing interest emerged in investigating the role of higher order spin interactions, both the SU(2) invariant and the chiral ones. In this contribution we study the effect of four-spin interactions along selective paths in the spin-configuration space of magnetic trimers. In order to calculate the SU(2) invariant four-spin interactions we use a Green's function perturbation scheme, where the spinless part of the Hamiltonian defines the unperturbed system, while the spin-dependent part of the Hamiltonian is treated as perturbation. Two-spin and four-spin interactions can then be obtained in second and fourth order perturbation of the Green's function. We demonstrate how the four spin interactions enter the spin model parameters obtained from the method of infinitesimal rotations leading to spurious tensorial two-spin interactions. The theory can be straightforwardly implemented in the Korringa-Kohn-Rostoker Green's function technique and in terms of this method we perform calculations for Cr and Mn trimers deposited on heavy metal surfaces. Comparing the energy obtained from the spin model with direct calculations of the band energy proves that the inclusion of the four-spin interactions significantly increases the accuracy of the spin-model description of the magnetic clusters under consideration.

Thickness dependence of magneto-ionic effects in Co3O4 films

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Electric field-induced oxygen motion in magnetic materials (magneto-ionics) has attracted huge interest among magnetoelectric mechanisms due to its potential to largely modulate magnetic properties in a nonvolatile (*i.e.*, permanent) way. Reliable magneto-ionics could become a solution in the search for strategies to enhance energy efficiency in magnetically-actuated devices. However, voltage-driven oxygen motion rates are still rather low at room temperature, hindering magneto-ionics from real applications. Recently, it has been shown that room-temperature magneto-ionic motion in electrolyte-gated Co₃O₄ films can be significantly increased using an electrochemical capacitor configuration (*i.e.*, utilizing an underlying conducting buffer layer as electric contact) instead of a transistor-like configuration in which the electric contacts are placed at the sides of the Co_3O_4 film [1,2]. To further optimize magneto-ionic motion, the role of film thickness (*i.e.*, size-effects) in the magneto-ionic rate of electrolyte-gated paramagnetic Co₃O₄ films in electrochemical capacitor configuration is investigated. Specifically, Co₃O₄ films with thicknesses of 5, 20, 50, 100 and 225 nm have been grown by reactive sputtering on Cu-coated Si substrates. With thickness reduction, magneto-ionic motion strongly enhances, with an increase of two orders of magnitude when comparing the thickest (250 nm) and the thinnest (5 nm) films (20 and 2313 emu·cm⁻³·h⁻¹, respectively). A detailed correlation between magneto-ionic performance and pristine microstructure and electric transport properties has been carried out. Our results approach magneto-ionics towards technological applications which require moderate operation speeds, such as neuromorphic/stochastic computing [2].

[1] A. Quintana et al., Adv. Funct. Mater, 27, 1 (2017)
[2] J. de Rojas et al., Adv. Funct. Mater. 2003704, 1 (2020)

Spin-reorientation transition in an Fe double-layer on W(110) induced by Dzyaloshinsky-Moriya interaction

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A large variety of complex magnetic structures has been proposed for existing and potential spintronics, information technology and biomedical applications. Controlling the preferred direction of the magnetic moments is essential for the design of the relevant devices based on ultrathin films and heterostructures. As the film thickness or the temperature is increased, the easy anisotropy axis is typically reoriented from an out-of-plane direction preferred by surface and interface energy contributions to an in-plane alignment favored by the volume anisotropy terms. The temperature-induced spin-reorientation transition (SRT) is often explained based on the different temperature dependence of the various contributions to the magnetic anisotropy.

In this work, we demonstrate the ultimate role of the Dzyaloshinsky-Moriya interaction (DMI) in the temperature-induced SRT in an Fe double-layer on W(110) in terms of advanced numerical simulations [1]. We perform well-tempered metadynamics simulations using an atomistic spin model parametrized by ab initio calculations to reproduce the experimentally observed SRT in the Fe double layer on W(110). Moreover, we explicitly show that this SRT does not occur if the DMI is absent. The underlying mechanism is revealed to be the interaction between the fluctuating transversal spin components mediated by the in-plane DM vectors present in the system [2]. This finding highlights the role of spin correlations in determining equilibrium magnetic configurations at finite temperature, essential for understanding the properties of room-temperature applications based on chiral domain walls or skyrmions.

[1] B. Nagyfalusi, L. Udvardi, and L. Szunyogh, Phys. Rev. B 100, 174429 (2019)

[2] L. Rózsa, U. Atxitia, and U. Nowak, Phys. Rev. B 96, 094436 (2017)

Magnetic scattering in stripe domain structure of Ni wires deposited on ferroelectric LiNbO3 substrate

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Recently, attempts to fabricate artificial multiferroic materials of which their functionalities worked above room temperature have focused because the magnetic domain structure and their magnetization dynamics can be controlled by the application of electric fields or strain that is electrically induced through piezoelectric effect. The heterojunction leads to a specific texture by formation of domain walls in stripe domain structure. For example, the stabilization of homochiral Néel domain wall is achieved by Dzyaloshinskii-Moriya interaction (DMI) or symmetry breaking at the interface. A domain wall is an unique object to play a physical interpretation of magnetization reversal and magnetoresistance (MR). In this study, we investigate the enlargement of magnetoresistance (MR) of Ni wires with the stripe domain structure. The role of physical origin of the MR enlargement on the interpretation of our experiments is discussed in detail. First, to investigate the magnetization reversal characteristics, 25-nm-thick Ni films are deposited onto SiO₂/Si and LiNbO₃ substrates, respectively by a normal magnetron sputtering. As a result, magnetization hysteresis loops measurements with a vibrating sample magnetometer reveal that the uniaxial magnetic anisotropy is spontaneously induced in the Ni films on LiNbO₃ substrate as shown in Fig. 1. Next, we prepared 30-nm-thick Ni wires placed in the center strip-line of coplanar waveguide (CPW) electrodes. We measured the MR of Ni wires directed perpendicular and parallel to the X-axis of LiNbO₃ substrate. As shown in Fig. 2, the MR behaviors are well known as the anisotropic magnetoresistance (AMR) effect. We found a clear anisotropic behavior that the MR ratio is strongly dependent on the direction of the uniaxial magnetic anisotropy induced by the heterojunction. This is attributed to that the fact that resistance change is significantly influenced by the domain structure and scattering process. To simplify the observed phenomena, we assume that the stripe domain structure consists of series-connected single domains and domain walls. The Δ MR ratio (%) = $n \times \Delta$ MR+(n-1) $\times R_{DW} \sim n \times (\Delta$ MR+ R_{DW}), where n is the number of single domains directed perpendicular to the longitudinal axis of the wire and R_{DW} is the resistance derived from the domain wall. This approximation is the series resistance circuit model. Besides, we approximate that $R_{\rm DW}$ is sufficiently smaller than Δ MR. As a result, here, we approximately calculate *n* as *n* ~ Δ MRstripe/ Δ MRsingle. Here, Δ MRstripe and Δ MRsingle are Δ MR obtained in the case that the stripe domain structure and single domain state are formed in the Ni wires, respectively. According to our previous study, number of stripe domain can be controlled by the wire width because the number is almost proportional to the wire width. Figure 2 shows the wire width dependence of estimated n^{Δ} MRstripe/ Δ MRsingle. The discrepancy of fitting is attributed to the contribution from the domain wall resistivity.



Figure 1 : Magnetization hysteresis loops measured with vibrating sample magnetometer of 25-nm-thick Ni film deposited onto (a) SiO2/Si and (b) LiNbO₃ substrates, respectively.

Figure 2: Magnetic field dependece of MR ratio of Ni wires alined **(a)** perpendicular and parallel to the X-axis of LiNbO₃. **(c)** Domain structures of 0.5 and 2.0 μ m wide Ni wires were calcualted by micromagnetic simulation. **(d)** Wire width depence of estimated $n \sim \Delta MR$ stripe/ ΔMR single

Assessing the effect of RKKY exchange through heavy metal on skyr-mionic devices

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Room temperature skyrmions have been observed in thin film multilayers (MLs) with perpendicular anisotropy (PMA) and interfacial Dzyaloshinskii-Moriya interaction (iDMI)¹. The skyrmions wall type is governed by several competing energies: exchange, anisotropy, magnetostatic, and DMI. It has been shown that the domain wall type and chirality determines the strength of the stray field around the thin film². Moreover, knowing the type and strength of the interlayer coupling is a prerequisite for design of future skyrmionic devices³. Recent studies⁴ showed that the domain structure of the skyrmion in each ferromagnetic (FM) layer depends on its position within the FM/heavy metal ML. The dependence of the skyrmion wall on the vertical position inside the ML was attributed to the minimization of the magnetostatic energy leading to a flux-closure structure. However, the existence of a weak Ruderman–Kittel–Kasuya–Yosida (RKKY) interlayer exchange could significantly alter the scenario because such an interlayer exchange could easily dominate a magnetostatic interaction. Neglecting the RKKY exchange in modelling work stems from the fact that it has not been thoroughly studied in structures supporting skyrmion formation. Therefore, it is important to investigate the role of RKKY coupling in such multilayered structures in combination with the other energies present in these systems.

We report on the ML systems that allow the assessment of interlayer RKKY interaction relevant for systems with iDMI, thus provide the required input for the design of ML systems with well-defined uniform skyrmion spin textures throughout the ML system. Note that a similar concept has been already used⁵, but for systems with a magnetic in-plane anisotropy. Also, because the coercivity of the locked layer was very small, only antiferromagnetic exchange coupling could be measured. Here we are interested in systems with PMA and ability to measure both antiferromagnetic and ferromagnetic exchange, with the latter being particularly important for systems with iDMI supporting skyrmions.

The systems consist of a Co (free) layer exchange coupled through an interlayer of Pt to a Co(0.6nm) layer that is strongly exchange-locked to the magnetically hard rare-earth amorphous ferrimagnet consisting of a Tb_xFe_{1-x} alloy with tunable composition to allow measurement of a ferromagnetic or antiferromagnetic exchange. To measure a ferromagnetic interlayer-RKKY-interaction, the composition of the Tb_xFe_{1-x} alloy is selected such as Tb magnetically dominated. After saturation of the sample in a positive field the Tb moment will align along the positive (up) direction, while the Fe moment and that of the adjacent Co layer remains in the down direction. With sufficiently strong applied positive field, the moment of the free Co layer will also be aligned along the positive field direction. The ferromagnetic interlayer-RKKY-interaction will however flip the magnetization of the latter layer into the down direction if the positive field is reduced below a critical value. Then the RKKY ferromagnetic coupling energy density can be obtained from magnetometry by $E=2M_{free}\cdot H_{ex}$.

The obtained results will serve as a knowledge base for design of complicated ML structures required for future skyrmionic future memory systems.

- [1] Bacani et al., Sci. Rep. 9, 2019
- [2] Marioni et al., Nano Lett. 18, 2263, 2018
- [3] Suess et al., Aip Adv. 8, 2018
- [4] Legrand et al., Sci. Adv. 4, 2018
- [5] Parkin, Phys. Rev. Lett. 67, 3598, 1991

Poster

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In depth domain walls in GdCo/NdCo/GdCo magnetic trilayers studied by transmission X-ray microscopy

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The combination of ferrimagnetic and ferromagnetic alloys leads to a competition between magnetic interactions that can stabilize Bloch points and other singularities at the interfaces [1]. In particular, we have prepared by DC-magnetron co-sputtering $Gd_xCo_{1-x}/NdCo_5/Gd_vCo_{1-y}$ trilayers with 80 nm thickness per layer. By tuning the stoichiometry of the Gd-Co outer layers, the Co magnetic sublattice dominates in one layer, whereas the opposite side is Gd dominated. In this way, in-depth magnetization domain walls can be created due to the balance between exchange and magnetostatic interactions [2]. Moreover, the weak perpendicular magnetic anisotropy of the Nd-Co central layer induces a stripe domain pattern, supporting the formation of Bloch points, meron-like textures and vortex-antivortex pairs [3,4]. Here we present the interaction between both types of structures, in-depth domain walls, and magnetic singularities at the vicinity of bifurcations within the stripe domain pattern. For doing so we have carried out Vibrating Sample Magnetometry measurements, Magnetic Force Microscopy (MFM) imaging and element selective angular dependent Magnetic Transmission Soft X-ray Microscopy (MTXM) at the Gd M_{4.5} edge in the Mistral beamline of the ALBA synchrotron. MFM analysis performed in different series of trilayers shows differences in the stripe domain pattern depending on the Gd-Co composition. With oblique incident angle MTXM measurements we have visualized regions in the domain pattern with partial magnetization reversal where magnetic singularities are formed. Finally, X-ray Transmission vector tomography [5] has been applied to characterize their three-dimensional magnetic structure.

[1] A. Hierro-Rodriguez, C. Quirós, A. Sorrentino, C. Blanco-Roldán, L. M. Alvarez-Prado, J. I. Martín, J. M. Alameda, E. Pereiro, M. Vélez, and S. Ferrer, Phys. Rev. B 95, 014430 (2017)

[2] C. Blanco-Roldán, Y. Choi, C. Quirós, S. M. Valvidares, R. Zarate, M. Vélez, J. M. Alameda, D. Haskel, and J. I. Martín, Phys. Rev. B 92, 224433 (2015)

[3] C. Blanco-Roldán, C. Quirós, A. Sorrentino, A. Hierro-Rodríguez, L.M. Álvarez-Prado, R. Valcárcel, M. Duch, N.

Torras, J. Esteve, J.I. Martín, M. Vélez, J.M. Alameda, E. Pereiro and S. Ferrerk, Nat. Comm. 6, 8196 (2015)

[4] A. Hierro-Rodriguez, C. Quirós, A. Sorrentino, R. Valcárcel, I. Estébanez, L. M. Alvarez-Prado, J. I. Martín, J. M. Alameda, E. Pereiro, M. Vélez, and S. Ferrer, Appl. Phys. Lett. 110, 262402 (2017)

[5] A. Hierro-Rodriguez, D. Gursoy, C. Phatak, C. Quirós, A. Sorrentino, L. M. Alvarez-Prado, M. Velez, J. I. Martin, J. M. Alameda, E. Pereiro and S. Ferrer, J. Synchrotron Rad. 25, 1144 (2018)

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First-order reversal diagrams method as a tool to characterize magnetoplasmonic crystal magnetic properties

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Point-of-care devices and diagnostic platforms for biomedical applications are commonly proposed for capturing, separation and mixing of different biological analytes. The non-magnetic nature of biological tissues often implies the use of magnetic nanoparticles' systems to perform such operations. Quantitative analysis of the particular material can be achieved with the use of a magnetic field sensor [1]. The magnetic field sensor with required locality can be made with the use of magnetoplasmonic crystal (MPIC) [2] due to the magneto-optical response enhancement mediated by excitation of surface plasmon polaritons [3] on the MPIC surface. In this case, measurement of the external magnetic field is achieved through the field dependence of the magneto-optical response.

Working parameters of the MPIC-based magnetic field sensor is determined by its magnetic properties which should be studied in detail to find possible ways of tuning such magnetic field sensor performance. Current work is devoted to the study of the magnetic properties of the MPIC as a thin layer structure of Ag(100 nm)/Fe(5 nm)/Si₃N₄(20 nm) deposited onto the subwavelength trapezoidal diffraction grating with the period and height equal to 740 nm and 100 nm, respectively. Study of the MPIC magnetic properties were performed with the vibrating-sample and magneto-optical magnetometers. Out-of-plane and in-plane hysteresis loops had a complex shape due to the presence of several magnetic phases. To describe the nature of additional phases, the first-order reversal curve diagram and switching field distribution approaches were employed. The use of these methods allowed to determine reversible and irreversible magnetization processes of material deposited onto different parts of the diffraction grating occurring due to the magnetostatic interactions and structural displacements in the sample. These results can be further used to tune the MPIC-based magnetic field sensor response curve.

[1] M. Rivas, Sensors Actuators A Phys. 216 (2014) 123

- [2] V. K. Belyaev, J. Magn. Magn. Mater. 482 (2019) 292
- [3] V. K. Belyaev, J. Magn. Magn. Mater. 480 (2019) 150

Influence of milling damage on magnetic performance of Sm2Fe17N3 fine powder produced by jet-milling

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A process for realizing a sintered magnet of $Sm_2Fe_{17}N_3$ have to meet the following three requirements. The first one is to ensure the sintering temperature is sufficiently low. This is because $Sm_2Fe_{17}N_3$ thermally decomposes at about 620 °C. The second one is to use a raw powder with no surface oxide film. This is because iron oxides contained in the surface oxide film react with the main $Sm_2Fe_{17}N_3$ phase to precipitate α -Fe nanocrystals, resulting in severe coercivity degradation. The third requirement is to ensure a sufficiently high coercivity for the raw powder. It is difficult for $Sm_2Fe_{17}N_3$ to improve coercivity by post-sintering treatments at high temperatures. So, we need to prepare a highly coercive powder in advance of sintering, and to pass the coercivity on to the sintered body without deterioration.

In order to meet the last requirement, it is effective to make the raw powder as fine as possible. As a method for producing very fine and coercive powder of $Sm_2Fe_{17}N_3$, a reduction-diffusion process is well known. However, a thick oxide layer is inevitably formed on the powder surface through this process, and requirement 2 cannot be satisfied. At present, therefore, pulverization of coarse powder in a low oxygen atmosphere seems to be the most effective means for achieving both low-oxygen concentration and fineness. Until now, we have successfully produced low-oxygen $Sm_2Fe_{17}N_3$ fine powders with no decrease in coercivity by sintering, using a swirling-gas-flow type jet-mill in a glove box with an oxygen concentration less than 0.3 ppm.

In the present study, we investigated the relationship between the pulverizing conditions and the properties and characteristics of the obtained powder. N_2 was used as a pulverizing gas. The pulverization was repeated at a constant pulverizing pressure, and changes in the particle size distribution, full width at half maximum (FWHM) of an XRD peak as a measure of crystallinity, and magnetic properties were examined as functions of the number of pulverizations. Similar experiments were performed at different pulverizing pressures (0.3, 0.7, 1.5 MPa) and compared with each other.

The averaged particle diameter (d_{50}) of the pulverized powder depends both on the pulverization pressure and the number of pulverizations. Therefore, powders with almost the same d_{50} can be prepared by different combinations of the pulverizing pressure and the number of pulverizations. As a result, it was found that these powders with similar d_{50} exhibit similar particle size distributions and FWHM of an XRD peak, even though they were prepared at different conditions. In fact, when the FWHM of the Sm₂Fe₁₇N₃ (220) peak was plotted as a function of d_{50} , all the pulverized powders were nearly on a universal curve regardless of the pulverizing pressure. Furthermore, the smaller the particle size, the higher the coercivity, but the lower the magnetization. It has been found that these changes are also apparently independent of the pulverizing pressure and are well scaled by d_{50} of the resulting powder.

Preparation and characterization of Eu0.5Bi2.5Fe5O12 thin films for magnetooptical imaging plates

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A magneto-optical (MO) imaging technique is promising for measuring the alternating magnetic field with a microwave frequency range, and is important for the high-speed telecommunication technology such as 5G. In order to achieve an MO imaging for the microwave, MO imaging plates are required to have a magnetic resonant frequency as high as the microwave range. We have already succeeded to prepare $Y_{3-x}Bi_xFe_5O_{12}$ and Nd_{3-x}Bi_xFe₅O₁₂ thin films for MO imaging plates¹⁻³. Rare-earth elements in R_{3-x}Bi_xFe₅O₁₂ play an important role for the ferromagnetic resonance as well as the saturation magnetization and the magnetic anisotropy. In this study, we prepared and characterized Eu_{0.5}Bi_{2.5}Fe₅O₁₂(EBIG). EBIG thin films prepared on Gd₃Ga₅O₁₂ (GGG) (100) single-crystal substrates by the metal-organic decomposition (MOD) method.

EBIG thin films with a thickness of approximately 600 nm were prepared on GGG (100) substrates by the MOD method using the MOD coating solution (BiFeEu-04 (2.5/5/0.5), Kojundo Chemical Lab.). To prepare EBIG thin films, the MOD coating solution was spin-coated at 3000 rpm on the GGG (100) substrates for 30 s. The resulting films were dried at 100 °C for 10 min, and then pre-annealed at 450 °C for 10 min. This process, spin-coating to pre-annealing, was repeated 5 times. The films were crystallized in a furnace at 770 °C for 3 h in air. To obtain a thickness of approximately 600 nm, the above process was repeated 4 times. For characterizations, Faraday rotation spectra and Faraday rotation hysteresis loops were measured.

Figure 1 shows Faraday rotation spectrum. The spectrum has a similar shape to those of $Y_{3-x}Bi_xFe_5O_{12}$ and $Nd_{3-x}Bi_xFe_5O_{12}$ thin films, indicating that bismuth-substituted rare-earth iron garnet was obtained. The Faraday rotation of EBIG exhibited a maximum at a wavelength of 522 nm. Figure 2 shows Faraday rotation hysteresis loop measured at a wavelength of 522 nm. The Faraday rotation angle was measured to be 16.5 °/µm, which was comparable to 18.1 °/µm of $Nd_{0.5}Bi_{2.5}Fe_5O_{12}^4$ and it was found to have an easy axis of magnetization with a saturation magnetic field of approximately 3 kOe.



Figure 1 : Faraday rotation spectrum of the EBIG thin film.



Figure 2: Faraday rotation hysteresis loop of the EBIG thin film measured at a wavelength of 522 nm

- [1] T. Ishibashi, et al., J. Appl. Phys., 113, 17A926 (2013)
- [2] E. Jesenska, et al., Opt. Mater. Exp., 6, 6 (2016)
- [3] Y. Nagakubo, et al., J. Magn. Soc. Jpn., 41, 29-33 (2017)
- [4] M. Sasaki, et al., Jpn. J. Appl. Phys., 55, 055501 (2016)

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Surface architectures impact on magnetic and magnetooptical properties of NiXFe100 X thin films

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The magnetization reversal process – is a key process for many applications such as magnetic memory devices, read heads, magnetic sensors and others. For thin-films structures, the magnetization process can be tuned by a complex effect of the shape and magnetocrystalline anisotropies [1], by magnetostatic interaction caused by "orange peel", dipole-dipole or domain walls interactions [2]. In this work, we examine the way to tune magnetic properties and magnetooptical response of Ni_xFe_{100-x} thin films and to apply thin films with different types of surface architectures as a magnetic field sensor [3].

The samples of Ni_XFe_{100-X} thin films with the thickness of 5 nm were deposited by magnetron sputtering method onto the Si substrates. The Ni_XFe_{100-X} composition was varied in the range of X from 40 to 80%. With the decrease of Ni percentage, the grain size increases from 6 nm to 20 nm. In this case, disorientation of the local axes of easy magnetization occurs. As a result, magnetization tends to release out of the film plane, coercivity increases from 3 Oe up to 10 Oe.

The obtained features were found to be a reason for magnetooptical peculiarities of the samples prepared onto the substrates with different geometrical parameters. Particularly, it was used the three types of polymer diffraction gratings with different period/height ratios: 320/20 nm, 740/100 nm, and 1400/100 nm. It allowed enhancing the magnetooptical response – the polar Kerr effect that can be used for producing of the magnetic field sensors.

[2] Morini, M., Slastikov, V. (2018). Reduced Models for Ferromagnetic Thin Films with Periodic Surface Roughness. Journal of nonlinear science, 28(2), 513-542

[3] Belyaev, V. K., Kozlov, A. G., Ognev, A. V., Samardak, A. S., Rodionova, V. V. (2019). Magnetic properties and geometry-driven magnetic anisotropy of magnetoplasmonic crystals. Journal of Magnetism and Magnetic Materials, 480, 150-153

^[1] Tretiakov, O. A., Morini, M., Vasylkevych, S., lastikov, V. (2017). Engineering curvature-induced anisotropy in thin ferromagnetic films. Physical review letters, 119(7), 077203

Voltage induced interfacial effects in a LSMO/PMNPT heterostructure

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New developments in oxide heterostructures offer great promise for improvements in magnetic storage, spintronics and high frequency magnetic devices. The combination of ferromagnetic and ferroelectric orders provides an efficient way to manipulate the magnetism by applying voltage. With $La_{0.67}Sr_{0.33}MnO_3 - LSMO$ possessing strong coupling between lattice, charge, spin and orbital degrees of freedom combined with a ferroelectric and piezoelectric $0.7(Pb(Mg_{1/3}Nb_{2/3})O_3)-0.3(PbTiO_3) - PMNPT$, it offers a great prospect of device concepts based on magnetoelectric (ME) coupling. Our main objective is to understand the mechanism driving the magnetoelectric coupling in this system and to investigate any contributions from interface effects due to applied voltage.

Magnetic properties of LSMO/PMNPT (001) heterostructure are studied using SQUID-VSM, which reveals a clear existence of non-volatile ME coupling due to asymmetric butterfly loop obtained on voltage application. After the removal of the voltage, the magnetometry measurements show overall increase of 18% in the magnetization from $0.76\mu_B/Mn$ to $0.9\ \mu_B/Mn$ of the system at 300K. LSMO/PMNPT shows varying behavior of magnetoelectric coupling as function of temperature which is observed clearly in SQUID measurements. Using polarized neutron reflectometry (PNR) magnetic depth profiles are probed as a function of applied voltage which indicate the presence of interlayer with reduced nuclear (NSLD) and magnetic (MSLD) scattering length densities between LSMO and PMNPT. The interfacial morphology of the heterostructure is characterized using transmission electron microscopy, which agrees with PNR measurements confirming the presence of an interlayer. The magnetic depth profile of this system will be studied using electron holography as well in the future.

SAF-based perpendicularly magnetized GMR spin valves on large-area flexible substrates

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Stability of Mn2RuGa Heterostructures

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Modern spintronic materials are being developed for high speed, non-volatile data storage, such as MRAM. An ideal material would have high spin polarisation, yet no net magnetic moment. Mn₂Ru_xGa (MRG) is a ferrimagnetic Heulser alloy, considered to be a zero-moment half-metal (ZHM) at its compensation temperature, and is a potential candidate for these applications owing to its immunity to magnetic fields, and large perpendicular anisotropy. This class of ZHM has garnered interest recently, with the majority of studied materials being Mn-based Heuslers.

Typical MTJ structures use a MgO/CoFeB free layer, which requires annealing at temperatures typically of order 350 °C. A major concern with Mn-based materials is diffusion based defects which affect the crystal structure as well as interfaces within a device. Recently, it was shown that Ta dusting layers can be used to mitigate diffusion between MRG and an MgO barrier, though this impacts the TMR of the device due to the short spin-diffusion length of Ta.

A spin-valve type heterostructure of the form MgO//MRG(30)/y(t)/[Co(0.4)/Pt(0.8)]₈/Co(0.4)/Pt(3) is used to evaluate potential materials to be used as a spacer layer, y, with a spacer thickness t of 1.4-2 nm. The structure uses a Co/Pt multilayer as the free layer to allow perpendicular anisotropy without requiring lattice matching. Crystallographic and magnetotransport characteristics are measured and compared before and after annealing at 350 °C as a standard comparison to industrial practices.

The spacer materials were chosen based on suitability for the task. Candidates are:

- TiN Known diffusion barrier, should grow epitaxially with MRG when deposited at 250 °C
- Hf Ultrathin layers were previously shown to promote coupling between MRG and CoFeB after annealing
- HfO_x Used as high-*k* barrier (gate dielectric) in the semiconductor industry. A known Al getter
- Mo Low affinity for alloying with Mn

Comparison of Extraordinary Hall effect measured on various heterostructures before and after annealing indicates that the anisotropy of the Co/Pt multilayer is highly dependent on interface quality. By contrast, the anisotropy of the MRG layer is related to its crystal structure, with its properties linked to the strain of the crystal in-plane and out of plane.

A comparison of the relative change, in coercivity (H_c), squareness (remanance/saturation), lattice spacing *c*, and perpendicular coherence length L_c (from Scherrer), gives an indication of stability induced by presence of a given spacer layer in the stack. When TiN is used, a larger L_c is observed, but a smaller tetragonal distortion. Annealing appears to remove defects from the MRG film, resulting in a loss of electronic pressure. This suggests that some degree of disorder is benificial for controlling the properties of MRG. Vandium appears to diffuse directly into MRG, filling defects, but also disrupting the crystal structure. The only stacks that maintained perpendicular anisotropy were those with Hf and Mo spacers. This suggests small amounts of Hf and Mo diffusion may help to stabilise the MRG crystal structure by replacing electronic pressure lost from defect removal.

It is important that the properties of MRG be predictable and closely matched to the as-deposited state. Much of the strain in MRG appears to be related to the disorder in the films. Hf or Mo as dusting layers between MRG and barrier are the only ones that help improve performance by reducing diffusion defects during anneal.

Crystallisation of Optically Thick Films of CoxFe(80-x)B20: Evolution of the (Magneto-) optical and Structural Properties

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CoFeB is one of the most extensively studied magnetic material due to its atypical properties, such as structurally smooth growth[1], soft magnetic properties [2], high spin polarization [3], and very low Gilbert damping [4]. This makes CoFeB especially suitable for magnetic tunnel junction devices [5]. Numerous works proclaimed that thermal annealing is a decisive means for achieving the best performance of CoFeB based magnetic tunnel junction devices. Therefore, it is of utmost importance to understand the influence of thermal annealing on this alloy.

Here, the crystallisation of Co-Fe-B alloys triggered by thermal annealing was investigated by X-ray diffraction (XRD) techniques and scanning electron microscopy (SEM), as well as spectroscopic ellipsometry and magneto-optical Kerr effect spectroscopy for annealing temperatures ranging from 300°C to 600°C. The transformation of ~100 nm thick $Co_xFe_{(80-x)}B_{20}$ films prepared by magnetron sputtering on silicon substrates from the amorphous as-deposited to the polycrystalline phase was revealed by the sharpening of spectral features observed in optical and magneto-optical dielectric function spectra. These features are ascribed to direct inter-band transitions due to the hybridization of p- and d orbitals of Co-Fe alloys in the bcc crystalline phase. Cross-sectional SEM of Co-Fe-B layers annealed at 600°C further reveal that in the case of a Pt capping layer, the crystallization starts at the interface of CoFeB and Pt, and expands for a thickness, which is consistent with the vertical coherence length of the crystallites determined from XRD. The influence of B on the dielectric function was assessed both experimentally and by optical modelling. By analysing the Drude component of the optical dielectric function, a consistent trend between the charge carrier scattering time/resistivity and the annealing temperature was observed, in agreement with electrical investigations by means of a four-point-probe method. The result obtained from this study were recently published in ref.[6].

- D. D. Djayaprawira, K. Tsunekawa, M. Nagai, H. Maehara, S. Yamagata, N. Watanabe, S. Yuasa, Y. Suzuki, and K. Ando, Appl. Phys. Lett. 86, 92502 (2005)
- [2] H. Hauser and R. Grössinger, J. Appl. Phys. 85, 5133 (1999)
- [3] S. X. Huang, T. Y. Chen, and C. L. Chien, Appl. Phys. Lett. 92, 242509 (2008)
- [4] C. Bilzer, T. Devolder, J.-V. Von Kim, G. Counil, C. Chappert, S. Cardoso, and P. P. Freitas, J. Appl. Phys. 100, 053903 (2006)
- [5] T. Devolder, J.-V. Kim, L. Nistor, R. Sousa, B. Rodmacq, and B. Diény, J. Appl. Phys. 120, 183902 (2016)
- [6] A. Sharma, M. A. Hoffmann, P. Matthes, O. Hellwig, C. Kowol, S. E. Schulz, D. R. T. Zahn, and G. Salvan, Phys. Rev. B 101, 054438 (2020)

Magneto-ionic control of in-plane and out-of-plane exchange bias

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The power consumption of magnetic devices is a challenge in many technological areas. Low-voltage induced ion migration and electrochemical redox processes is an emerging approach to control ferromagnetic properties at the nanoscale. These magneto-ionic effects are promising for energy-efficient spintronics, actuation and neuromorphic computing [1-3]. Combining a ferromagnet with an antiferromagnet gives rise to the exchange bias effect, which is exploited in a variety of nano-magnetic applications. In a first study, we utilize a FeOx/Fe layer with an underlaying antiferromagnet, making tunable exchange bias properties feasible. Indeed, in an FeO_x/Fe/IrMn film system, we achieve non-volatile (see Fig. 1 (a)) and reversible changes in the in-plane exchange bias. The underlying voltage-triggered oxidation/reduction reaction are confirmed via X-ray photoelectron spectroscopy. Within a simple model, the exchange bias changes can be explained by a change of the ferromagnetic layer thickness when transforming FeO_x into Fe and vice versa[4]. A second study deals with the Co/NiO film system, which exhibits out-of-plane exchange bias. Hydrogen ion pumping [5] was utilized to toggle between out-of-plane and in-plane perpendicular anisotropy of the Co layer. This goes along with an reversible ON/OFF switching of the perpendicular exchange bias field (see Fig. 1 (b)). These results are exciting for designing exchange bias systems and future magneto-electric devices in general.



Figure 1: (a) Non-volatile in-plane EB shift. (b) Off/On switching of perpendicular EB field.

- [1] K. Leistner et al., Phys. Rev. B, 87, p. 224411 (2013)
- [2] C. Navarro-Senent et al., APL Mater. p. 030701 (2019)
- [3] A. Molinari, Adv. Mater. p. 1806662 (2019)
- [4] J. Zehner, R. Huhnstock, S. Oswald et al., Adv. Electr. Mater. 5, p. 1900296 (2019)
- [5] A. Tan et al., Nat. Mat. 18, p. 35-41 (2019)

Magnetotransport properties of deformed Bi nanowires

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The presented investigations of the magnetotransport measurements of Bi wires complement the series of recently published experimental results on bulk Bi in high magnetic field. The design of measurements in magnetic field was diversified by using uniaxial strain directed along the wire axis. By combining high magnetic field and strain, the electronic structure of the bismuth wires was modified; as a result, the quantum limit for light and heavy electrons could be changed in different ways. Measurements of the longitudinal magnetoresistance in magnetic field of up to 35 T oriented along the bisector axis of Bi wires have revealed an anomaly in a magnetic field far above the quantum limit of the electrons: a sharp peak of MR at 33T. Investigation of magnetoresistance under uniaxial strain has revealed that the sharp peak of the magnetoresistance at 33 T is reproduced in lower magnetic fields at 28 T according to a decrease in the light electrons and the Lifshitz Transition has been found. The result is that the critical magnetic field of the Electronic Topological Transition has decreased; thereby, the magnetic field range of the occurrence of magnetic-field-induced instabilities associated with the last Landau level of electrons has been extended.

Anisotropy engineering of soft thin films in the undulated magnetic state

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Advanced materials often involve multi-dependent approaches, merging electrical, optical and magnetic responses in nanostructured systems.¹ To this purpose we have explored a semi-transparent, conductive, soft magnetic thin film (Permalloy) of undulated shape with which uniaxial anisotropy can be induced and tuned. This media has been grown on the ripple surface of flexible, polymeric foil of polyethylene terephthalate (PET) that was previously patterned by a versatile pulsed-laser irradiation technique achieving a linear array with periodicity 220 – 250 nm and large amplitudes up to 58 nm.

Vectorial Kerr (reflection) as well as Voight (transmission) magneto-optical effects confirm a complete uniaxial anisotropy induced with easy axis along the ripple pattern for Permalloy films of thickness 10 - 30 nm. Analysis of magnetization loops and critical fields in comparison with existing models and novel micromagnetic simulations of a quasi-infinite ripple film model (with the same dimensions as the media) indicate and undulated magnetization state with the anisotropy driven by volume-like anisotropy at the ripple crests/valleys. The choice of large pattern dimensions has made possible to realize the undulated magnetic state where the anisotropy strength simply increases with film thickness, without any evidence of surface contributions.



Figure 1 : (A) 5x5 μ m² AFM topography image of the surface pattern of 20 nm thick Permalloy film grown on a nano-undulated PET foil. The inset shows a ripple amplitude histogram from the surface pattern. **(B)-(C)** Normalized longitudinal (black symbols) and transversal (red symbols) magnetization loops obtained from the longitudinal MOKE of the 15 nm thick Permalloy film for the field applied along the easy-magnetic axis (B) and hard-magnetic axis (C). Red solid line is a loop calculated for such direction with Stoner-Wohlfarth model. **(D)** 10 nm thick Permalloy ripple film modelled by micromagnetic simulations. The arrows show the direction of the magnetic moments and the colored regions indicate the normalized z-component (m_z/m_s) of the magnetization at field higher than the anisotropy field (μ_0 H > μ_0 H_K). **(E)** Simulated loops of (D) with the field applied across the ripple pattern (α_H = 89.9^o).

- Miguel A. Arranz, Elena H. Sánchez, Esther Rebollar, Marta Castillejo, and José M. Colino, Opt. Express 27, 21285-21294 (2019)
- [2] Elena H. Sánchez et al., Anisotropy engineering of soft thin films in the undulated magnetic state (submitted to J. Magn. Magn. Mater.)
Pervasive artifacts revealed from magnetometry measurements of rare earthtransition metal thin films

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Amorphous rare earth-transition metal (RE-TM) alloys exhibit a rich spectrum of magnetic properties. They are ferrimagnetic with compensation points that cover a large temperature range, can exhibit perpendicular magnetic anisotropy, and can have very high coercive fields for certain compositions. More recently, RE-TM alloys have been investigated in the vicinity of their magnetization and angular momentum compensation points for high-speed domain wall- and skyrmion racetrack-type memory devices. The high magnetic tunability of RE-TM ferrimagnet-based heterostructures allows them to be tailored to specific applications, provided that their magnetic parameters are determined accurately. These RE-based systems are known to be very sensitive to the growth conditions and the existence of soft magnetic characteristics in the hysteresis loops of an otherwise magnetically-hard film has been previously observed and mainly explained as the result of preferential oxidation of the RE element or chemical inhomogeneities. However, in such complex systems, distinguishing between real magnetic features and artifacts can be challenging and requires some caution. We reveal a class of artifacts manifesting as soft magnetic components from magnetometry measurements of TbFe thin films prepared by magnetron sputtering. They are not inherent to TbFe, but are a direct result of the manner in which the substrates are mounted prior to sample fabrication, with material deposited at the substrate sides giving rise to a significant magnetic moment. The observed artifacts are not caused by preferential oxidation of the RE element, magnetic impurities, or presence of very soft grains within our films. Trying to supress the appearence of this type of artifact has an influence on the coercivity and, in some cases, on the shape of the reversal curves. We find the same artifacts to also be present in rare earth-free [Co/Pt] multilayers. Care needs to be taken during fabrication to ensure reliable and reproducible samples so that sensitive magnetic parameters, such as coercivity and compensation points, can be extracted accurately and that data is not misinterpreted for even more complex systems. This type of artifact is not limited to samples prepared by sputtering, but can extend to other conventional thin-film deposition methods.

Temperature dependence of the magnon diffusion lengths in a spin Seebeck device

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The spin Seebeck effect refers to a magnetic moment flow generation as a consequence of the temperature difference across a magnetic material that usually is an insulating ferrimagnet. A high spin-orbit coupling heavy metal deposited on one surface of the ferrimagnet works as a detector for the magnetic moment current by exploiting the inverse spin Hall effect. In the last decade, these effects on magnetic-insulator/heavy-metal (i.e. Yttrium Iron Garnet (YIG)/Platinum) bi-layer samples have stimulated a wide range of experimental and theoretical research about the interaction between phonons and magnons and the transport properties of magnons. Recently, we have verified the reciprocal relations between the spin Seebeck and the spin Peltier effects through an experiment [2]. By employing a non-equilibrium thermodynamics model [3], we have interpreted the spin Seebeck and spin Peltier coefficients in terms of the product between intrinsic properties of the materials such as the thermomagnetic power coefficient, that relates the gradient of the potential of the magnetization current with the temperature gradient, the magnon diffusion length in YIG and the spin conductivity of each material. From the last two quantities we derived in this work the temperature dependence of the magnon diffusion length, by employing the temperature dependence of the transport coefficients of exchange magnons [4]. This model is in agreement with the experimental results reported so far in the literature [5] and it can be employed for the interpretation of other experiments involving the temperature dependence of magnon properties, or the spin Seebeck effect dependence on the thickness of the Pt and the ferrimagnetic layers. Indeed, a similar approach can be adopted for the Pt layer, by using the temperature dependence of the electric conductivity of Pt, as shown in Fig. 1. From this characteristic, through the relation between the magnetic moment conductivity and the electric conductivity of Pt, it is possible to derive the temperature dependence of spin conductance and therefore the magnetic moment diffusion length.



Figure 1: Temperature dependence of the electric resistivity of Pt and magnetic moment conductance and diffusion length.

- [1] K. Uchida et al., Appl. Phys. Lett. 97, 172505 (2010)
- [2] A. Sola, et al. Sci. Rep. 9.1 (2019)
- [3] V. Basso et al., Phys. Rev. B 93, 184421 (2016)
- [4] V. Basso et al., Phys. Rev. B 94, 144422 (2016)
- [5] E. J. Guo et al. Phys. Rev. X 6 031012 (2016)

Manipulation of magnetism in NdNiO3 by proximity effects

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The possibility to manipulate quantum properties through heterostructure engineering lies on the basis of modern correlated electron physics. By combining materials with different types of ordering new properties appear at their interface. Moreover, this additional tool enables the further understanding of the correlation between different degrees of freedom (charge, magnetism, orbital) by manipulating one property and observing an alteration of the other one.

In this work we have manipulated the magnetic state of NdNiO₃ (NNO) by growing ultra-thin NNO films in proximity to a magnetic layer. NNO undergoes a metal to insulator transition concomitant with an antiferromagnetic ordering in bulk at 200K. We have grown 5 unit cells NNO film in contact with ferromagnetic (La_{0.67}Sr_{0.33}MnO₃, LSMO) or antiferromagnetic (La_{0.33}Ca_{0.67}MnO₃) layers, deposited on NdGaO₃. In order to single out the NNO properties from the manganite we employed x-ray magnetic circular dichroism to measure the Ni and Mn magnetization separately. Our results [1] show that for NNO, in NNO/LSMO system, exhibits ferromagnetism with the same Curie temperature as LSMO. On the other hand, for NNO/LCMO, no ferromagnetism is observed. This clearly shows that the type of magnetic ordering has an influence on the magnetism of NNO. In addition, angular resolved photoemission [1] show clear changes on NNO electronic structure for different proximity layers, showing the correlation between these two degrees of freedom.

[1] M. Caputo, Z. Ristic, R. S. Dhaka, T. Das, Z. Wang, A. Zakharova, C. E. Matt, J. Jandke, M. Naamneh, M. Medarde, N. C. Plumb, M. Shi, J. Mesot, C. Piamonteze and M. Radovic, *in preparation*

Magnetic phase diagram of full-manganite heterostructure unveiled by PNR

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Understanding the interplay between different competing energies at the interface of heterostructures is one of the greatest challenges in both fundamental and applied physics. In particular, interfaces made of strongly correlated oxides have shown unexpected physical properties, such as the exchange bias, proximity effects, charge transfer, spontaneous magnetic reversal, exchange springs and orbital reconstruction [1-5]. Given the complexity of its structural and magnetic phase diagram, LSMO offers a wide range of tunable properties that we can stack into heterostructures. With the atomic precision of the oxide MBE we were able to tune the Sr-concentration x layer by layer (at 0.4 obtaining a ferromagnetic half-metal and at 0.8 an antiferromagnetic insulator), and synthesize superlattices with a large gradient in hole doping. This has proven to be a promising platform to study the competition between diverse exchange interactions, giving rise to different phenomena such as charge transfer, exchange bias, spontaneous magnetization reversal, and long range magnetic interaction.

In this talk, we will discuss the results obtained by polarized neutron reflectometry and SQUID magnetometry of these heterostructures, while trying to shed light on the macroscopic and local magnetic properties and their connection to the Sr-doping depth profile.

- [1] Jason D. Hoffman et al., Phys. Rev. X 6, 041038 (2016)
- [2] J.-H. Kim, I. Vrejoiu, Y. Khaydukov, T. Keller, J. Stahn, A. Rühm, D. K. Satapathy, V. Hinkov, and B. Keimer, Phys. Rev. B 86, 180402(R)
- [3] M. Saghayezhian, S. Kouser, Z. Wang, H. Guo, R. Jin, J. Zhang, Y. Zhu, S. T. Pantelides, and E. W. Plummer; PNAS May 21, 2019 116 (21) 10309-10316
- [4] J. Stahn, et al., Phys. Rev. B 71, 140509 (2005)
- [5] G. Kim, Y. Khaydukov, M. Bluschke, Y. Suyolcu, G. Christiani, K. Son, C.Dietl, T. Keller, E. Weschke, P. A. van Aken, G. Logvenov, B. Keimer, Phys. Rev. Mat.3 (2019)

Oxidized Permalloy Thin Films: Rise of Magneto-optical Response in Infra-red Spectral Range

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Ni-Fe alloys are widespread soft magnetic materials that are often used as a key constituent in a broad range of nowadays investigations–magnetic field sensors [1], spintronic devices [2], magnetooptical metamaterials [3] and spintronic-plasmonic metasurfaces [4].

In the present work, we study permalloy island nanofilms with a thickness of 20 nm. The nanofilms were fabricated on 1mm-thick glass substrates using magnetron sputtering, and then were annealed in air at 300-475 °C for one hour. Properties of the as-deposited and oxidized nanofilms were studied by the optical, XPS, AFM and VSM techniques. We found that, during annealing, the initially smooth amorphous nanofilms (RMS = 1 nm) transformed into a disordered array of nanopillars having characteristic dimensions of order 45 nm×160 nm (height × diameter) and average distance between the pillars was about 1 μ m. For the nanofilms annealed at 300-450 °C, the coercive force and the saturation magnetization in the in-plane direction were significantly increased. For the out-of-plane geometry, magnetization tended to its maximum for the 300-350 °C-oxidized nanofilms and dropped down at temperatures of > 450 °C. These changes can be addressed to the rise of the roughness of the film and, thus, to the increasing number of different crystal phase magnetic domains. Unexpectedly, the magnetooptical figure of merit rose by more than one order of magnitude in the near-infrared range-the transmittance after annealing at 450 °C was three time higher as compared with the initial one (Fig1.(a)) and the values of Faraday rotation at 1.55 µm for the nanofilms under study were 0.02° for as-deposited and 0.51° for 425 °C-oxidized nanofilm (Fig1.(b)). The observed responses can be interpreted by well-known formation of various phases–oxides NiO, Fe₂O₃ and spinel (Ni Fe)₃O₄ [5] that was also supported by our XPS studies. The rise of the Faraday rotation can be explained by the formation of Fe_3O_4 phase [6] (due to Fe_2^{2+} ions in octahedral site) followed by oxidation up to Fe_2O_3 at temperatures > 425 °C. As for the optical properties of the obtained films, it deserves special attention and will be reported at the conference. We do believe that the fabricated nanofilms can be of interest for magnetic [1,2] and magnetooptical applications [3-5,7].



Figure 1 : (a) The transmittance and (b) the Faraday rotation spectra of fabricated nanofilms before (curve 1) and after annealing for one hour at the series of temperatures. Curves 2-7 correspond to 300, 350, 400, 425, 450, 475°C, and curve 8–the substrate. The inset of plot (b) shows the ellipticity difference for as-deposited and annealed at 425°C. Spectra 7 and 8 are divided by two. Data were collected at H = 3.8 kOe.

[1] V.K. Belyaev et al, JMMM 482, 292 (2019).

- [2] S. lihama et al, Phys. Rev. B 94, 020401(R) (2016).
- [3] I.A. Kolmychek et al, Phys. Rev. B 99, 045435 (2019).
- [4] G. Armelles et al, Nanophotonics 8(10), 1847 (2019).
- [5] X. Dong et al, J. Solid State Chem. 246, 309 (2017).
- [6] T. Tepper et al, J. Appl. Phys. 93, 6948 (2003).
- [7] M. Inoue et al., Magnetophotonics: From Theory to applications (Springer, 2013).

Sputtered carbon-doped MnAl thin films on Si substrates

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Magnetic tunnel junction (MTJ) miniaturization results in ever smaller volumes of magnetic material; subsequently, this results in a reduced thermal stability factor, for a given magnetic anisotropy energy of the material. Simultaneously, lowering the critical switching current of MTJs becomes of great importance for reducing the energy consumption of the devices. For satisfying the first requirement, a magnetic material should have high magnetocrystalline anisotropy. The second requirement is fulfilled by lowering the Gilbert damping constant; furthermore a moderate saturation magnetization is required for reducing the switching current, while maintaining sufficiently high tunneling magneto-resistance.

State-of-the-art CoFeB ferromagnetic thin films, which are a typical component of spintronic layer stacks, are reaching their limit, due to the moderate interfacial perpendicular magnetic anisotropy, and the relatively large values of saturation magnetization and Gilbert damping constant.

To overcome these emergent limitations on the material side, one of the promising approaches is to use thin films having high perpendicular magnetic anisotropy, moderate saturation magnetization and low damping. These requirements can be fulfilled relying on $L1_0$ chemically ordered alloy thin films, having a face centered tetragonal phase with ordered alternation of A and B atoms planes along *c* axis of the lattice. Such phase could be formed in numerous binary systems, including those yielding high uniaxial perpendicular magnetocrystalline anisotropy (PMA): e.g. FePt, FePd, CoPt, FeNi.

This work focuses on Mn-based L1₀-ordered thin films. The L1₀ phase of the Mn–Al and Mn–Ga binary alloys is well-known to be ferromagnetic; it exhibits excellent hard magnetic properties although contains neither ferromagnetic elements – such as Fe, Co, and Ni – nor noble metals such as Pt. Application of these alloys in spintronic devices offers several advantages, allowing to skip the limitations for further development caused by the properties of CoFeB alloy. Mn-based alloys are known to poses high PMA, moderate saturation magnetization, and small damping constant; therefore, their use could lead to the reduction of MTJ switching current, while maintaining high thermal stability.

A key limitation of these materials is related to the stringent synthesis conditions that have to be met, including the use of expensive monocrystalline substrates, and non-industrial deposition methods like molecular beam epitaxy. In fact, there is a lack of any evidence that the $L1_0$ crystal structure of Mn-based alloys may be obtained onto oxidized silicon wafers, inhibiting the industrial manufacturing of such MTJ devices.

In this work, we focus on the synthesis of carbon-doped MnAl thin films, magnetron sputtered onto commercial oxidised Si wafers. We explore both Mn-rich and Al-rich films for determining the optimum composition for achieving L1₀ crystal structure. Most importantly, we investigate the effect of carbon-doping on the crystal structure of these films. The deposition and annealing temperature is also studied for ensuring compatibility of the material to standard microelectronics processes. A thorough study of the obtained phases will be presented.

High Entropy Alloys: The next big thing in soft magnetic alloys

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The search for new soft magnetic thin films has led to the exploration into multi-component alloys (MCAs), which contain four or more elements with percentages between 12.5 % and 33 %. These include the subclass of high entropy alloys (HEAs) which often exhibit single phases, thought to be stabilised by the high entropy of mixing of these compounds amongst other competing thermodynamic contributions. The resulting alloys often have interesting functional properties, which are linked to their structure. This work has explored a number of different compositions (CoFeNi_{0.5}Cr_{0.5}Al_x, CoFeNiCrCu_x, CoFeNiMnAl_x) in both bulk form and thin films, to understand how the phases present in the alloys influence the magnetic properties. The work has also used semi-empirical models to predict the different phases within the compositions and machine learning has been used to predict the saturation magnetisation.

For example in bulk CoFeNi_{0.5}Cr_{0.5}Al, it was found that Fe-Cr nanoparticles formed in a weakly magnetic CoNiAl matrix. This formation of nanoparticles increased the magnetisation of the CoFeNi_{0.5}Cr_{0.5} alloy by a factor 4 at room temperature. While the phases present in CoFeNi_{0.5}Cr_{0.5}Al thin films, were strongly influenced by the film thickness, with the thinnest films containing a 2-3 intermetallics, which were both magnetic and non-magnetic, while the thicker films contained a disordered BCC phase, and had a larger saturation magnetisation. For all the CoFeNi_{0.5}Cr_{0.5}Al samples, the coercive field was small and the saturation magnetisation comparable with NiFe.

While for bulk CoFeNiCrCu_x (x = 0.5, 1.0, 1.5) samples, it was found that the addition of Cu could be used to tune the Curie Temperature over a 100 K range, with less than a 30 % reduction in the saturation magnetisation at 10 K, while the addition of Cr to CoFeNiCrCu changed the Curie Temperature by 700 K and reduced the saturation magnetisation by 60 % at 10 K.

The magnetocaloric effect (MCE) was studied in these materials, as the compositions tend to involve cheaper, more widely available elements, plus careful tuning of the composition will allow for the Curie Temperature to be around 300 K. It was found that the refrigeration capacity for the CoFeNiCrCu_x alloys was ~ 1 J/kg and for the CoFeNi_{0.5}Cr_{0.5}Al_x alloys was ~ 17 J/kg. Although this is smaller than existing materials, these HEAs still have advantages due to the presence of second order transitions and having a wider operational temperature range for the MCE.

In conclusion, the MCAs studied have promising soft magnetic properties, with small coercive fields, and reasonable saturation magnetisations. The phases present can be tuned via the composition, allowing for two-magnetic phase systems to be fabricated, that improve the magnetic properties compared to a single solid solution. The research carried out is only the tip of the iceberg, but these alloys are showing encouraging results.

Nanostructured semiconductor – metal magnetic junctions

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Magnetic junctions, typically composed of ferromagnetic layers separated by a nonmagnetic layer of metal or insulator, found their applications in electronics for example as reading heads in hard disc drives or magnetic memories MRAM. The junctions with metallic interlayer show giant magnetoresistance effect (GMR), and in the case of an insulator interlayer the tunnel magnetoresistance effect (TMR) is observed. Magnetic junctions based on those effects are the first spintronic elements used commercially in the electronic industry. Another type of junction is a system in which the spacer layer is a semiconductor. The semiconductor/metal interface forms a junction, called the Schottky diode, characterized by non-linear and asymmetric dependence of current on voltage. The use of a ferromagnetic layer in this type of junction can lead to the formation of diode sensitive to a magnetic field.

Modification of magnetic properties of a junction can be carried out by nanostructurization, a process of spatially ordered structures formation of the size of tens of nanometers. Many methods are used for nanostructurization, like electron lithography, nanoimprinting, or, used in this work, anodization. The anodization is an electrochemical process of formation of homogenous, highly ordered nanoporous or nanotubular oxides of (transition) metals. Additionally, the anodization allows for easy control of dimensions of the nanostructures, such as the diameter and height of the pores/tubes, by changing reaction parameters such as anodization voltage, electrolyte strength, temperature, and time.

In this study, the junctions consist of a thermally deposited ferromagnetic thin iron layer of 50 nm thickness deposited on anodized titanium oxide (ATiO). Fabrication of the ATiO layer was carried out in electrolyte containing ammonium fluoride (NH₄F) at constant voltages of 5 V, 15 V, and 60 V. To protect the iron from oxidation and ensure good electric contact a layer of gold was deposited on top of the junction. The crystal structure of the ATiO was observed with X-Ray Diffraction (XRD). The magnetic properties were investigated with SQUID magnetometry for two different geometries of a magnetic field, perpendicular and parallel to the junction plane. The results showed that an easy magnetization axis for all junctions is in the film plane and the hysteresis curves are a combination of a few magnetic phases including different iron oxides formed at interfaces. The magnetoresistance characteristics conducted at room temperature showed a positive value of magnetoresistance and transition to negative values at 5 K.

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Magnetic and electrical transport properties of double Ti/TiOx/Fe thin film junction with magnetic layer prepared with anodization process

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We present results of magnetic and electrical transport properties of the thin film Ti/TiO_x/Fe magnetic junction. Studies were performed in a temperature range from 5 K to 300 K, and in magnetic fields up to +/- 50 kOe. The obtained results were compared with morphological and structural properties.

The junction was prepared in a few steps. First, a thin Ti layer was formed, and subsequently the top part of the titanium layer was oxidized with anodization process. The junction was finished after covering the oxide layer with 50 nm thick layer of iron. As prepared junction was heated to 750 K.

Magnetic studies showed ferromagnetic properties of the junction for wide range of temperatures with a strong in-plane magnetic anisotropy. On the other hand, resistivity analysis showed greater variation in the obtained results. The magnetoresistance (MR) measured through the double junction at 300 K has a negative value, while at 5 K it is positive. Change in conductivity is observed around 215 K. Detail studies of a single junction, i.e. studies of magnetoresistance between Ti/TiO_x and TiO_x/Fe junctions separately showed that the change in sign is attributed to the TiO_x/Fe interface. Meanwhile, the Ti/TiO_xjunction has a positive value in full range of studied temperatures, however, its value increases with temperature decrease. Values of MR in the Ti/TiO_x/Fe junction and between single junctions were found to be of single percentages.

Except for magnetoresistance, a voltage-driven change in resistivity dR(V) was calculated according to equation R(V)/R(0)-1. The resistivity values were obtained from a derivative of current-voltage characteristics, where dI/dV=1/R(V). Values of such defined resistance variation are small, i.e. tenths of a percentage. Likewise magnetoresistance, the change of dR(V)from negative to positive values was observed with a decrease of temperature. The change in sign happens for similar range of temperatures like for MR case. Interestingly, the change in dR(V) arises from Ti/TiO_x junction while the dR(V) variation in TiO_x/Fe junction is always positive and does not change significantly.

The TiO_2 is well-known semiconductor were at metal/semiconductor interface a voltage-driven transport response can be expected. On the other hand, magnetoresistive properties are found after using in the junction a magnetic metal as a metallic layer. The changes in sign of voltage and field driven dependencies of resistivity are presumably the effect of intermixing and formation of non-stoichiometric oxides at both Ti/TiO_x and TiO_x/Fe interfaces.

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Varying Dzyaloshinskii-Moriya constant with temperature in ultrathin Pt/Co(Fe)B/Ir films

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*The author has chosen not to make public additional content.

Towards electric control of magnetism in thin films of FeRh on BaTiO3

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Controlling and manipulating the magnetic properties of magnetoelectric materials using an electric-field has driven many scientific research studies, as it is considered a promising approach for realising low-power technology [1]. FeRh is a metamagnetic material which exhibits a first-order magnetic phase transition from an antiferromagnetic (AFM) state at low temperature (<360 K) to a ferromagnetic (FM) state at high temperature (>380 K). This transition is accompanied by a change in the resistivity and the out-of-plane lattice constant as a result of the strong coupling between the structural and magnetic properties of FeRh [2,3]. These properties make it an ideal material for heat-assisted magnetic recording and future spintronics applications [1,3]. This transition can be induced by chemical doping, strain, a change in temperature and magnetic field. However, a full understanding of the origin of this phase transition from a non-ferroelectric cubic phase to a ferroelectric tetragonal phase upon cooling from 400 K, in which the latter is divided further into a_1 , a_2 and c-domains in the presence and absence of voltage [3]. Although many studies have reported the electrical control of ferroelectrics, they have been limited to the paramagnetism-FM phase transition within a small range near the Curie temperature [3].

We report the electric control of the magnetic properties of FeRh thin film grown on BTO substrate, as well as the voltage and temperature dependence of the strain induced by the BTO substrate during the FeRh AFM to FM phase transition using soft x-ray. Significant enhancement of the hysteresis of about 200 K with various abrupt transitions was observed in the magnetisation measurements of a FeRh thin film grown on a BTO substrate. We carried out specular and off-specular reflectivity scans using circularly polarised x-ray at the Fe *L*-edge (707.8 eV) at these sharp changes. The specular reflectivity measurements taken as the temperature decreases from 400 K to 250 K show multiple peaks which could correspond to the BTO tetragonal domains. Furthermore, the change in the number of peaks at 280 K is attributed to the first-order phase transition from the tetragonal phase to the orthorhombic phase in the BTO substrate.

We will also present the additional measurements taken with different voltages applied across the thickness of the sample, discuss the possible interpretation of the results supported by complementary magnetic and structural characterisation measurements.

- [1] Z. X. Feng, H. Yan, Z. Q. Liu, Adv. Electron. Mater. 5, 1800466 (2019)
- [2] J. Hong et al., Nanomater. 9(4), 574-574 (2019)
- [3] L. C. Phillips, et al., Sci. Rep. 5, 10026 (2015)

Poster

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Temperature dependence of Gilbert-damping in Permalloy/Topological insulator heterostructures

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Huge spin-orbit coupling makes topological insulators (TI) very attractive for topological spintronics. In our work we present FMR experiment with heterostructure TI/Ferromagnetic layer. As ferromagnetic layer we used Py ($Ni_{80}Fe_{20}$) with 20 nm thickness. Topological insulator was Bi_2Se_3 , due to its record values of spin Hall angle. Thickness of TI layer was 5 nm. In this work S21 parameter was measured as a function of external magnetic field. Then results were approximated with equation (1) to obtain width of resonances peaks. Gilbert-damping parameter was calculated using equation (2).

$$rac{d(S21)}{dH} = y_0 + rac{A[(H-H_r)\Delta H] + B[(H-H_r)^2 - \Delta H^2]}{(H-H_r)^2 + \Delta H^2)^2}$$
, (1) $\Delta H = \Delta H_0 + rac{4\pilpha f}{|\gamma|}$, (2)

We obtained that Gilbert-damping in TI/FM structure is 0.0066 ± 0.0005 and 0.014 ± 0.001 for 295K and 50K, respectively. In pure Py films with 20 nm thickness Gilbert-damping parameter was much lower. Same result was obtained in work [1] but only in pure permalloy samples with thickness 3 nm and mechanism of this phenomenon is in surface and bulk damping. For permalloy sample with same thickness Gilbert parameter was approximately 0.006 in all temperature range. Our hypothesis is that this phenomenon may relay on spin-orbit coupling, or may be spin-orbit coupling can amplify surface and bulk damping.



Figure 1 : Temperature dependence of Gilbert-damping in Py/TI heterostructure.

[1] Yuelei Zhao et al., Scientific Reports (2016)

Unusual manifestation of weak magnetism and superconductivity in bicrystal interfaces of Bi, Sb and Bi1-xSbx (0.07 =x = 0.2) alloys

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The magnetic properties of the high quality nano-width crystallite interfaces (CIs) of Bi, Sb and 3D topological insulator $Bi_{1-x}Sb_x$ (0.06 $\leq x \leq 0.2$) were studied [1] in temperature range 1.6–300K using Quantum Design SQUID magnetometer and a Physical Property Measuring System with a 140kOe induction magnet. These semimetals do not superconduct under normal rhombohedral conditions and are diamagnetic. At the same time, the superconducting phases with $T_c \leq 21K$ and magnetic hysteresis loops on a diamagnetic background typical for strong type II superconductors were identified in interfaces of Bi [2]. In CIs of $Bi_{1-x}Sb_x$ (0.06 $\leq x \leq 0.2$) as well as of Sb with a higher carrier density were observed a superconducting transitions and a ferromagnetic hysteresis loop or a dual loop (superimposed ferromagnetic and superconductivity and weak ferromagnetism, what is specific to 3D topological insulators. The CIs superconductivity with the critical temperature of $T_c \simeq 10K$ and weak ferromagnetism are detected in Sb CIs for the first time. The revealed coexistence of superconductivity and weak magnetism at these CIs is of increased interest for the fundamental physics and future applications in quantum computing and spintronic devices [3].

 Fiodor M. Muntyanu, Andrzej Gilewski, Andrzej J. Zaleski, Vitalie Chistol, Viorel Munteanu, Krzysztof Rogacki and Anatolie Sidorenko, Functional Nanostructures and Metamaterials for Superconducting Spintronics, Springer International Publishing AG, part of Springer Nature., Resp. ed. A. Sidorenko, 2018, Chapter 12, pp.1-15
F. M. Muntyanu, A. Gilewski, K. Nenkov, A. Zaleski, and V. Chistol, Phys. Rev. B. 76 (2007) 014532
F. M. Muntyanu, K. Nenkov, A. J. Zaleski, N. Muntean, V. Chistol, Solide State Comm. 299 (2019) 113660

Structural and magnetic properties of FePd thin film synthesized by electrodeposition method

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Bimetallic nanomaterials in the form of thin film constituted by magnetic and noble elements show promising properties in different applications field such as catalysts and magnetic driven applications. In order to tailor the chemical and physical properties of these alloys to meet the applications requirements, it is of extreme scientific interest to study the interplay between properties and morphology with particular attention on surface properties, microstructure, spatial confinement and magnetic features.

In this work, we deal with this large subject by focusing on bimetallic thin films combining a noble metal, Pd, with a magnetic transition metal, Fe. The former is attracting a wide interest for its catalytic properties, whereas the second, among the magnetic ones, is the least expensive, simultaneously offering among the best magnetic properties. The nanomaterials are prepared in thin films form by electrodeposition, which allows obtaining a Fe-rich composition, and offers the possibility to easily control the film thickness by simply picking the desired deposition time. The surface (e.g. roughness) and magnetic properties depend on the film thickness, as specific microstructure and morphological features develop as a function of deposition time. They have been subsequently refined through chemical etching in hydrochloric acid, which enhanced the roughness on the thickest films and helped decoupling their grains. Such changes resulted in the ability to fine tune the magnetic properties (e.g. coercivity) and increase the surface-to-volume ratio. X-ray diffraction, as well as roughness, contact angle and magnetic measurements have been carried out with the aim of providing a comprehensive characterisation of the fundamental properties of these bimetallic thin films.

Broadband IFE in the PCs with the Magnetic Layer of Gradient Thickness

<u>Olga Borovkova</u>, Mikhail Kozhaev, Andrey Kalish, Vladimir Belotelov; Russian Quantum center

The PC nanostructure composed of two Bragg mirrors and a subwavelength layer between them allows one to support an optical analogue of Tamm state. The sharp resonance peak appears in the bandgap of the transmittance spectrum. The spectral position and amplitude of the peak in the transmittance spectrum depends on the thickness of the microcavity between the Bragg mirrors. The optical Tamm state is also accompanied by the strong localization of light in the microcavity.

If the layer between the Bragg mirrors has magnetic properties, the magneto-optical (MO) effects can be observed there. Among a variety of such effects, we would like to focus on the inverse Faraday effect (IFE). It denotes the effect when the magnetization occurs in a transparent medium exposed to a circularly polarized high-frequency electromagnetic wave. The strong localization of light inside the magnetic layer of the MPC leads to an enhancement of the IFE in the nanostructure.

It is proposed a design of the photonic crystal (PC) nanostructure with the magnetic layer of gradient thickness sandwiched between two non-magnetic Bragg mirrors. The spectral position of the optical Tamm state of the magneto-photonic crystal (MPC) nanostructure depends on the spatial position of the input light spot on the sample surface. Therefore, the resonant enhancement of the IFE associated with the peak in transmittance band gap also changes with the input light spot position. Thus, illuminating the proposed MPC nanostructure by the circularly polarized light at the normal incidence the observation of the broadband inverse Faraday effect can be achieved. The numerical simulations revealed that an increase of the magnetic layer thickness from 120nm to 180nm leads to the resonance spectral position detuning from 0.6um to 0.675um, correspondingly.

Besides the smooth gradient magnetic layer, the PC nanostructure with the magnetic layer with etched discs is proposed. In the magnetic layer with the constant thickness the disks with diameter of dozen microns are etched. The etching depth is different for various disks. As a result, the thickness of the magnetic layer is smaller in the area of such disks. Therefore, the enhanced IFE occurs at the different frequencies for each disk. This idea allows us to achieve spatial localization of the IFE limited just by the disk diameter that is of several microns.

To sum up, two designs of the MPC nanostructures with gradient of the magnetic layer thickness for tunable inverse Faraday effect are proposed. The spectral position and amplitude of the transmittance peak and resonant IFE depend on the thickness of the magnetic layer. The first design of the MPC nanostructure has smooth magnetic layer with thickness gradient that allows to tune the spectral position of peak in transmittance spectra and the IFE gradually. The second design has perforated magnetic layer and provide spatially localized emergence of the IFE in the spots of several microns. The sample fabrication and the experimental verification of the reported results is expected.

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Magneto-optical properties of the spatially asymmetric magnetoplasmonic nanostructures with magnetization modulation by the spin waves

<u>Olga Borovkova</u>, Mikhail Kozhaev, Andrey Kalish, Vladimir Belotelov; Russian Quantum center

Recently, it has been reported the novel concept of the magnetoplasmonic nanostructures with spatial symmetry breaking. In such structures the intensity magneto-optical response in transmittance experiences a non-zero modulation even at the normal incidence of light when the external magnetic field is applied in the transverse direction.

We address the metal plasmonic nanostructure deposited on top of the magnetic layer has a spatial symmetry breaking. The unit cell of the plasmonic grating is spatially non-symmetric along the alternating direction of the grating cells. Thus, the symmetry of the structure is violated with respect to the rotation of the 180 degrees around the normal to the nanostructure. In other words, the asymmetry vector is directed along with the periodicity of the metal plasmonic structure. The surface plasmon polaritons (SPPs) excited at the [metal]/[magnetic dielectric] interface of the addressed structure experience the different conditions for the modes running forward and backward. This nonequivalence of two SPP modes propagating in opposite directions emerges due to the asymmetry of the addressed magnetoplasmonic nanostructure.

The excitation of the spin waves in the magnetic layer of the nanostructure leads to the modulation of its magnetization. As a result, the magneto-optical response of the magnetoplasmonic nanostructure is also dependent on the excitation of the spin waves.

We analyze by numerical simulations the mutual influence of the spin waves dynamics and the asymmetry properties of the magnetoplasmonic nanostructure. The features of the magneto-optical intensity effect in transmittance are discussed.

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Exploring magnetization profile in a all oxide magnetic tunnel junction

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In the last decades the interest in devices utilizing not only the charge but also spin of the electron has grown considerably due to the its smaller energy requirement to manipulate. Such devices including non-volatile magnetic memory, reprogrammable logic and quantum computing exploit spin polarization as the driving force where generation and detection polarized spins is an essential prerequisite for their efficient functioning. Epitaxially grown inverse spinal ferrite films (Fe₃O₄, CoFe₃O₄) have proven to be promising material due to their potential to generate 100% spin-polarized currents by an approach of using the spin selective transport of electrons across a ferro(ferri)-magnetic tunnel barrier called spin filtering [1]. This has motivated us to study an all oxide magnetic tunnel junction Fe₃O₄/MgO/CoFe₂O₄ as a spin filter device where Fe₃O₄(FO) layer serves as a source of spin polarized electrons while the CoFe₂O₄(CFO)/MgO acts as a double tunnel barrier. An essential requirement to achieve successful spin filtering is to decouple FO and CFO magnetizations for which we have grown MgO between the 2 layers to allow their magnetizations to be independently switched [2].

In such heterostructures a high spin filter effect depends critically on the quality of the interfaces which may be limited by degradation of the electronic structure CoFe2O4 film due to spontaneously formed structural disorder. Also, antiphase boundaries (APBs) may significantly reduce the magnetization due to formation of magnetic dead layers at heterointerfaces. We have used soft X ray magnetic reflectivity (SXRMR) technique at SEXTANTS beamline at synchrotron Soleil which is a combination of X ray reflectometry and X ray magnetic dichroism providing a depth resolved magnetic distribution across the film thickness with the advantage of elemental sensitivity and we can probe interfacial magnetic distribution at both Co and Fe L2,3 edge. Thus, we can investigate the coupling state of the two layers as well as the polarization at the bottom and the top interface of MgO sandwiched between CFO and FO.

For an improved picture of the system it is necessary to determine the magnetic distribution across a single purely uncoupled CFO layer grown by molecular beam epitaxy [3]. Through SXRMR measurements we got a homogeneous magnetic distribution at both Co and Fe L2,3 edge where we found a profile with 3 thicknesses of slight structural variations along the growth axis from the Ag interface. This finite Fe magnetic signal gives us an indication of existence of magnetization profile without any contribution from Fe in FO. In the MJT sample we need to study contribution from Fe in both FO and CFO separately for which we collected the magnetic signal by flipping longitudinal applied magnetic field strong enough to only magnetization in the soft FO layer but not strong enough to probe CFO layer and by reversing helicity of the circular polarized light while maintaining the orientation of the applied magnetic field by which we could probe the distribution of Fe in the entire film [4].

For our MJT sample, we have an indication of decoupling and are currently working on obtaining a full magnetic profile at both Fe and Co edges and we wish to present the combination of the results from the two samples films.

[1] S. Matzen et al,APL 101,042409,2012[2] M. Chapline et al,PRB 74,014418,2006

- [3] M. De Santis et al, Acta Cryst 2019
- [4] J.M. Tonnerre et al, PRB 84, 100407 2011

Epitaxial growth, structural and magnetic properties of exchange coupled Pd0.95Fe0.05/Pd0.92Fe0.08 bilayers and Pd0.95Fe0.05/Ag/Pd0.92Fe0.08 trilayers

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The interest to the dilute $Pd_{1-x}Fe_x$ (x = 0.01-0.10) alloy originates from its potential applications in superconducting spintronics [1]. The $Pd_{0.95}Fe_{0.05}/Pd_{0.92}Fe_{0.08}$ heterostructure with the thickness of both layers of 20 nm was synthesized by molecular-beam epitaxy technique (MBE system by SPECS) under ultra-high, $3\square 10^{-10}$ mbar, vacuum conditions on (001)-oriented epi-polished single-crystal MgO substrate. The epitaxial growth mode of the films was verified by the LEED and XRD techniques [2]. Magnetic properties were studied using VSM magnetometry (QD PPMS-9) and ferromagnetic resonance (FMR, X-band Bruker ESP300 spectrometer). The dependence of the magnetic moment on temperature M(T) is shown in the Figure, left panel, open symbols. It reveals a kink at ~ 160 K and can be decomposed into the magnetic responses of the constituent layers (shown by dash lines) using the M(T) data for individual $Pd_{0.95}Fe_{0.05}$ and $Pd_{0.92}Fe_{0.08}$ films from Refs. [2, 3]. The magnetic hysteresis loops (middle panel) measured at T = 20 K demonstrates a consolidated reversal of the magnetic moments of the layers with a unified coercive field. This is most probably a consequence of the strong inter-layer coupling due to the direct contact between the layers.

FMR technique was applied to study the magnetic anisotropies in the Pd_{0.95}Fe_{0.05}/Pd_{0.92}Fe_{0.08} bilayer. The FMR spectra were recorded at *T* = 20K in the in-plane and out-of-plane geometries of the experiment [4]. Two resonance lines were observed, simultaneously reflecting the presence of two oscillators in the system. The angular dependences of the resonance fields (H_{res}) for these lines are presented in the right panel of the Figure. The stronger ferromagnetic Pd_{0.92}Fe_{0.08} layer is expected to have a higher resonance field at $\vartheta_{H} = 0^{\circ}$ (along the film normal) and a lower resonance field at $\vartheta_{H} = 90^{\circ}$ (in-plane alignment) compared with a weaker ferromagnetic Pd_{0.95}Fe_{0.05} layer. Then, the out-of-plane angular dependences of the two independent resonances must intersect at an angle of about $\vartheta_{H} = 20^{\circ}$ between the film normal and the film plane. The experimentally observed angular dependence of H_{res} does not show this crossing (see square and circle symbols in the Figure, right panel). This may occur due to repulsion of resonance lines observed around the expected crossing angle are, in fact, collective in-phase ("acoustic") and anti-phase ("optical") modes [5]. Modelling of the angular dependences of the FMR fields for resonance (black solid lines) provides the magnetic anisotropy parameters of the adjacent Pd_{1-x}Fe_x layers and the strength of the coupling between them.



[1] V.V.Ryazanov, V.V.Bol'ginov, D.S. Sobanin et al., Phys. Procedia 36, 35 (2012)

[2] A. Esmaeili, I.V. Yanilkin, A.I. Gumarov et al., Thin Solid Films 669, 338 (2019)

[3] A.Esmaeili, I.V.Yanilkin, A.I.Gumarov et al., arxiv.org/pdf/1912.04852

[4] M.Farle, Rep. Progr. Phys. 61, 755 (1998)

[5] "Ultrathin Magnetic Structures II", ed. by B. Heinrich and J.A.C. Bland, Springer-Verlag Berlin-Heidelberg (1994) 361

IY, RY and LT were supported by the RSF project No. 18-12-00459, while AG and RK – by the RFBR project No. 20-02-00981. Synthesis and analysis of the films were carried out at the PCR Federal Center of Shared Facilities of KFU.

Symposium 17. Magnetism based post-von-Neumann computation

Neuromorphic computing with radiofrequency spintronic devices

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The need for energy efficient artificial intelligence has motivated research on the implementation of neural networks in hardware, using emerging technology. In particular, spintronic nano-oscillators have emerged as promising candidates to emulate neurons due to their non-linear behavior [1]. For instance, it has been shown that a network of nano-oscillators can learn to recognize spoken vowels, using radiofrequency (RF) signals as communication between neurons and the strength of their coupling as connections to tune [2]. However, in order to scale such systems to deep neural network capable of performing state of the art artificial intelligence tasks, it is necessary to have physical synapses -- which weights can be tuned --connecting the neurons. Here we propose a scalable architecture for neural networks using spintronic RF oscillators as neurons and spintronic RF resonators as synapses.

First, we show how individual spintronic resonators, and in particular magnetic tunnel junctions, can multiply RF signals by a tunable weight, thus emulating synapses. We use the fact that when a magnetic tunnel junction is submitted to an RF input of frequency close to its own resonance frequency, it generates – through the spin-diode effect [3] – a rectified voltage, which is proportional to the input RF power. Then, we show how to assemble these devices into chains performing the multiply and accumulate function, which is at the core of neural network. We use a frequency multiplexing scheme, where the connectivity of the network is defined by the frequency matching between relevant neurons and synapses. Finally, we show how to assemble a full neural network and perform classification tasks.

These results open the path for compact and energy efficient deep neural networks, using spintronic devices connected through RF signals as neurons and synapses.

- [1] J. Grollier et al., Nature Electronics (2020): 1-11.
- [2] M. Romera et al., Nature 563.7730 (2018): 230-234.
- [3] A. Tulapurkar, et al., Nature 438.7066 (2005): 339-342.

Neuromorphic Computation Using Emergent Magnetisation Dynamics

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Emergent behaviour in complex systems arises when the collective behaviour is not represented by discrete individual components, but is rather created from the interactions between the system's individual components [1]. Emergence is found in nature as a mechanism behind intelligent biological processes [2], but thus far, little has been done to exploit the computational power that emergence could offer for machine learning hardware. Here we demonstrate how the emergent properties of a magnetic system can be exploited to perform meaningful computation in materio.

Using a combination of X-ray photoemission electron (X-PEEM) microscopy and polarised neuron reflectivity (PNR), we show that arrays of interconnected magnetic nanowire rings demonstrate emergence as a result of complex domain wall (DW) pinning events when driven by a cyclic magnetic field, with the stochasticity associated with DW pinning producing emergent dynamic equilibria in DW population and array magnetisation [3]. We then present a phenomenological model of these stochastic pinning events, with the DW annihilation and nucleation mechanisms that come as a consequence, and show that the model provides excellent agreement to out experimental data (Fig. 1B).

Finally, we used our model to simulate the response of the ring arrays to signals. The system's nonlinear response and its inherent memory allowed us to implement a machine learning paradigm called 'reservoir computing' (RC). In RC, the traditional neural network is replaced by a time-dependent 'reservoir' which captures the computational advantages of recurrent neural networks (RNN), without the associated computational expense of training and modelling an RNN [4]. This has allowed us to demonstrate the feasibility of the ring arrays performing machine learning tasks, such as spoken digit recognition, speaker identification, and signal classification. We believe the sub-millimetre size, and ability to solve complex tasks, makes the arrays exciting candidates for microprocessors without the need for discrete computational and memory units, enabling more energy efficient data processing tasks due to the reduced computational expense.



Figure 1: A) X-PEEM Images representing domain wall configurations after 30 successive rotations of fields between 36 and 42 Oe. Arrays featured 25x25 permalloy nanorings of 2um diameter, 200nm track width, 5nm thickness. Contrast of blue and red areas show magnetsation in upward and downward directions respectively. [3] B) Comparison between experimental PNR data and modelled arrays of 25x25 rings after 50 rotations of a range of magnetic fields. Error bars represent maximum/minimum values during 100 repetitions, plotted points represent mean values.

- [1] P. Curşeu, J. Inf. Technol., 21, (2006)
- [2] B. Lindsey et al, Comprehensive Physiology, 2, (2012)
- [3] R. Dawidek et al. [In preparation]
- [4] M. Lukoševičius and H. Jaeger, Comput. Sci. Rev., 3, (2009).

Spintronic nano-neuron: solving machine learning tasks with hardware-based artificial neural networks

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The brain displays many signatures of non-linear dynamical behavior, including synchronization and complex transient behavior. These observations have inspired a whole class of neuromorphic concepts based on complex networks of interconnected non-linear nodes. Hardware systems for neuromorphic computing are usually tested using the so-called reservoir computing neural network architecture [1], a machine learning approach. One of the preferred tasks for benchmarking such devices is automatic speech recognition [2-7]. However, this task requires acoustic transformations from sound waveforms with varying amplitudes to frequency domain maps that can be seen as feature extraction techniques (see Fig. 1).

Depending on the conversion method, these may obscure the contribution of the neuromorphic hardware to the overall speech recognition performance. Here, we quantify and separate the contributions of the acoustic transformations and the neuromorphic hardware to the speech recognition success rate [8]. We show that the non-linearity in the acoustic transformation plays a critical role in feature extraction. We compute the gain in word success rate provided by a reservoir computing device compared to the acoustic transformation only (see Fig. 2) and show that it is an appropriate benchmark for comparing different hardware. Finally, we experimentally and numerically quantify the impact of the different acoustic transformations for neuromorphic hardware based on magnetic nano-oscillators.



Figure 1: Principle of spoken digit recognition. (a) Audio waveform. (b) Filtering to frequency channels for acoustic feature extraction. (c) The filtered input is injected into the neural network or directly used to construct the output (no neural network). The neural network is composed of N interconnected filtered inputs. (d) For each digit, the response of the neural network (or directly the filtered output) is constructed from a linear combination of neuron states.



Figure 2: Contributions to the spoken digit cross-validated test recognition rate. Random choice level is shown in grey, the filtering methods in blue, and the neural network under the reservoir computing approach in purple and green for the simulations and experiments, respectively. Here, 9 data subsets (90 % of the database) are used for training our reservoir computing model and the remaining subset (10 % of the database) is used to perform the recognition task.

[1] H. Jaeger, and H. Haas, Science 304, 78-80 (2004)

- [2] L. Appeltant et al., Nat. Commun. 2, 468 (2011)
- [3] Y. Paquot et al., Sci. Rep. 2, 287 (2012)
- [4] D. Brunner et al., Nat. Comm. 4, 1364 (2013)
- [5] K Vandoorne et al., Nat. Comm. 5, 3541 (2014)
- [6] J. Torrejon, M. Riou, F. Abreu Araujo, J. Grollier et al., Nature 547, 428-431 (2017)
- [7] M. Riou, F. Abreu Araujo, J. Torrejon, J. Grollier, IEDM 2017 (2017)
- [8] F. Abreu Araujo, M. Riou, J. Grollier et al., Sci. Rep. 10, 1-11 (2020)

Stochastic domain wall dynamics for machine learning

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Magnetic materials are widely used for long-term data storage but recent advancements have increased their potential as both working memory and computing architectures. In particular, devices based on magnetic domain walls (DWs) have been shown to be able to perform logic operations and can readily store information [1]. However, the stochasticity of DW pinning limits the feasibility of creating technologically viable devices. Here, we demonstrate how stochasticity can be changed from a technologically inhibitive behaviour into a functional property by exploiting it to implement machine learning algorithms that could be used in specialised neuromorphic devices.

We first present experimental measurements that demonstrate the feasibility of actively tuning stochastic processes by applying external stimuli. Focused magneto-optic Kerr effect measurements were used to probe the pinning of DWs at notch-shaped artificial defect sites in 400 nm wide Permalloy nanowires (Fig. 1.(a)). DWs were injected into the nanowires and propagated to the defect sites using a field parallel to the nanowire (Hx). The probability of the DWs being pinned at the defect sites was found to depend sigmoidally on the magnitude of a second field applied transverse to the nanowires (Hy), (Fig. 1.(a)).

The nanowires' stochastic behaviours were then integrated into a series of machine learning models. First, we explored the properties of feedforward artificial neural networks, where the nanowires acted as binary stochastic synapses (BSS). We derived a training rule for the network based upon the mean field probability and find that for a single cycle of the network it can be trained to recognise handwritten digits from the MNIST database with 85% accuracy. Repeated sampling of the network increased the accuracy up to 93% for 128 cycles - close to mean field performance (Fig. 1.(b)). The nanowires have also been modelled as a Restricted Boltzmann Machine (RBM), which exploits their randomness in order to sample a defined, or trained, probability distribution to solve problems such as integer factorisation where solutions are the most probable states [2]. Our work illustrates how the intrinsic stochasticity of DW devices can be harnessed to provide tunable bespoke hardware for machine learning tasks.



Figure 1: (a) Schematic of a notched magnetic nanowire for use as a synapse with a parallel driving field (Hx) and a tunable transverse depinning field (Hy) which gives a sigmoidal pinning probability. (b) The classification accuracy of handwritten digits using a single layer network with binary stochastic synapses (BSS) for varying number of cycles of the network.

- [1] R. L. Stamps et al., J. Phys. D. Appl. Phys. 47, 333001 (2014)
- [2] W. A. Borders et al., Nature 573, 390 (2019)

Neuromorphic computing using spin-wave scatterers

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We investigated the use of spin-wave scatterers as devices for performing linear transformations, or more specifically as fully connected perceptron layers. The device consists of multiple input and output channels, and a spin-wave scatterer in-between (see Fig. 1). In our micromagnetic simulations, this scatterer is realized by an applied magnetic field distribution over a thin YIG film. This field influences the propagation of spin waves from the input channels towards the outputs, and thus it represents a mapping between the input and output channels. Input and output data are represented by spin-wave amplitudes and phases (complex-valued vectors).

In the linear regime, this device performs a linear transformation between the input and output vectors, and thus it can be described by a complex-valued n-by-m matrix (assuming n inputs and m outputs). We performed micromagnetic simulations to demonstrate this functionality. For a given scatterer pattern, n simulations using the orthonormal basis vectors as excitation are sufficient to fully characterize the system, i.e. to determine all the matrix elements.

The more difficult problem to tackle is solving the inverse problem, i.e. for a desired matrix description of the system figuring out an equivalent scatterer pattern. For this we used the approach described in [1], by using machine-learning algorithms with gradient-based training. Using the approach of the Pytorch-based code developed by [1] for the general wave equation, we implemented a training algorithm that applies a custom full 2D micromagnetic solver inside the Pytorch framework with automatic gradient calculation and backpropagation throughout the micromagnetic solver. Using this algorithm, we were able to successfully demonstrate training of the scatterer pattern to achieve a specific output for a given input.

The equivalent perceptron-layer representation of the linear scatterer becomes evident if we consider the perceptron model as a scalar product followed by a nonlinear activation function. The weight vectors of a fully connected perceptron layer (m perceptrons with n input each) constitute an n-by-m matrix with the weight vectors being the columns of the matrix. The nonlinear activation function might be realized in the spin-wave domain by e.g. parametric pumping or by external electronic circuitry.

Finally, although we limited the current investigation to linear operation, we note that allowing nonlinear amplitudes in the spin-wave scatterer has the potential to perform a much wider class of transformations, thus realizing more complex operations in the same device geometry. Considering the neural-network analogy, nonlinear operation would allow the system to solve not-linearly-separable problems. Using linear waves, this is only possible by using multiple layers, with nonlinear activation functions applied in-between. Our current investigations and methods can also be extended to this nonlinear class of devices.

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[1] Hughes, Tyler W., Ian AD Williamson, Momchil Minkov, and Shanhui Fan. Science Advances 5, no. 12 (2019).

Radio-Frequency Synapses for Neural Networks Made of Spin-Torque Nano-Oscillators

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Spin Torque Nano-Oscillators are magnetic devices that use tunnel magnetoresistance to emit microwaves. They hold great promise for neuromorphic computing [1], as we can use their non-linear dynamic to emulate neurons and they can be integrated by millions in CMOS chips. In previous works, it has been shown that their transient or coupled dynamics can be used to perform pattern recognition [2], [3].

However, one of the biggest challenges in Neuromorphic Computing is to include memory units in the computing core: in the brain each neuron receives the weighted sum of 10⁴ neural signals mediated through synapses and the ability to tune these synaptic weights is called learning. Because the huge density of connections in Artificial Neural Network makes hardware implementations complicated, we believe that we could use microwaves signals mediated Neural Network in order to reduce wiring, thus increasing hardware network efficiency. And if we want to build a Deep Neural Network [4] with spintronic radio-frequency artificial neurons, it is mandatory to use radio-frequency synapses that can be tuned in a non-volatile way to weight the radio-frequency encoded neural signals. Here we present an architecture using spintronic resonators to weight the microwave encoded signals of artificial RF spintronic nano-neurons.

These spintronic resonators leverage the spin-diode effect [5,6]. The resistance of the devices oscillates at the same frequency as the signal they receive. The rectified voltages are proportional to the power of the microwaves and depend strongly on the frequency mismatch between the signal and the resonance frequency of the device. Hence to tune these resonance frequencies is analog to tune the synaptic weights. And because of the frequency selectivity of our resonators, each of them rectifies only the signal of its corresponding neuron.

We use an analytical model and dynamical simulations to show that a chain of spintronic resonators wired in series, rectifying different microwave signals, is equivalent to the key operation of neural networks, called Multiply-And-Accumulate (MAC), despite the nonlinearity of these components. We also demonstrate experimentally with Magnetic Tunnel Junctions as resonators and several RF generators, the validity of this MAC operation. Finally, we simulate a full network with spintronic oscillators and resonators to test our architecture. We manage to achieve state-of-the-art recognition on a handwritten digit dataset. These are encouraging results for high density hardware neural network, as it is the first demonstration of a fully-spintronic radio-frequency neural network.





Figure 1: Simulation of the voltage rectified by a 64 spintronic resonators chain with a microwave signal.

Figure 2: Several signals are summed and send to different chains of resonators to implement a MAC operation.

- [1] J. Grollier, D. Querlioz and M. D. Stiles, Proceedings of the IEEE, Vol. 104, p. 2024-2039 (2016)
- [2] J. Torrejon, M. Riou et al, Nature, Vol. 547, p. 428 (2017)
- [3] M. Romera, P. Talatchian, S. Tsunegi et al, Nature, Vol. 563 (7730), p. 230-234 (2018)
- [4] Yann LeCun et al, Nature, Vol. 521, p. 436-44 (2015)
- [5] C. Wang, Y.-T. Cui et al, Phys. Rev. B Vol. 79, p. 224416 (2009)
- [6] D. Marković, N. Leroux, A. Mizrahi, et al, Phys. Rev. Appl, Vol. 13 (4), p. 044050 (2020)

Scalable spin-wave computing

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Spin waves can perform information processing at very low energies and at very high speeds, making them one of the few most promising candidates for future information technologies. The creation and the detection of spin waves, however, is a power-hungry process, due to the large energy mismatch between spin-wave energies (in the micro-nanowatt range) and the energies required to operate microwave input / output circuitry (which consumes power in the milliwatt range). The net energy efficiency of spin-wave based computing can be high only if few inputs and outputs are required for a relatively large and complex operation in the spin-wave domain, i.e. the spin-wave computing medium is scalable to large sizes and magneto-electric interconversions are required only at the input / output peripheries.

Even in low-damping YIG films, however, spin waves can propagate to a distance of few hundred (possibly thousand) times their wavelength, and this may not be sufficient for a truly scalable device. This motivates our work toward an active spin-wave medium, where internal amplification ensures scalability.

The studied structure is shown in Figure 1a): Here spin-wave scatterers (rectangular regions) are connected by narrow spin-wave conduits. Microwave waveguides run atop the spin-wave conduits, these are pumped at two times the frequency of the propagating wave. Due to the ellipticity of the spin-wave orbit in the conduits, the out-of-plane field of the waveguides can parametrically amplify the spin waves, so excitations present in one region can propagate indefinitely to far-away scatterers. Figures 1b-d) show the propagation of such disturbance. Simulations demonstrate that there is a 'global' interference pattern formed, with a wave source felt at far-away locations in the structure.

The functionality of such a device is defined by structuring the scatterers: Patterns or localized magnetic field can be designed to perform certain functions. For example, one may define weighted interconnections between different, far lying magnetic regions and create the magnetic equivalent of a large-scale optical scatterer such as discussed in [2].

Using parametric amplification for long-range interconnections is not without challenges: Parametric pumping can quickly drive the scatterers to the high-amplitude, nonlinear regime, where the (nonlinear) interference patterns become complex and hard to predict. It is also non-trivial to inverse-design the scatterers for a given function. Despite these challenges, this device holds the promise for extremely energy efficient, scalable spin-wave computing, as a single microwave waveguide could supply power to a magnetic film with a size of several millimeters, and the complexity of the resulting interference pattern could rival that of an optical scatterer.

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Figure 1: a) shows a schematic layout of the studied active medium, while b-d) are snapshots of the excitations filling the area upon applying the pumping at t=0.

[1] G. Csaba, A. Papp, W. Porod. Physics Letters A 381, no. 17 (2017): 1471-1476.

[2] Roques-Carmes, Charles, et al. Nature communications 11, no. 1 (2020): 1-8.

Investigation of Anisotropy and Current-Control of Magnetisation in Complex Oxide Heterostructures for Spin-Memristors

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New computing paradigms that take inspiration from the brain have the potential of overcoming some of the limitations with respect to speed and power consumption that are inherent to the von-Neumann computing architecture due to the separation of memory and processing. To realise this, materials are needed which can function as the neurons and synapses of the brain. This requires materials with multistate behavior, meaning that a physical characteristic can be controllably changed by an external stimulus so that the value of a physical property depends on the history and can take on several values for the same control parameter. To emulate artificial synapses, materials are desired that modulate their state in an analog manner and store it nonvolatile way. One of the most widely studied class of devices for this purpose is memristors: materials that undergo a reversible change under the application of a voltage that can be probed as a change in resistance. However, this desired characteristic can also be realised using the magnetisation state of a material, ideally controlled by current. These layers can be integrated in tunnel junctions to create a non-volatile "spin-memristor" in which the resistance can be controlled though an in-plane current. Using magnetic thin films as electrodes, doped SrTiO₃ show resistive multilevel resistive switching with large on/off ratios. A wide range of emergent phenomena have been demonstrated using complex transition metal oxides. We explore new approaches to control the nonvolatile magnetisation state in thin films with tailored magnetic anisotropy. In particular, here complex oxide heterostructures consisting of SrRuO₃ grown on SrTiO₃, where perpendicular magnetic anisotropy is observed, are investigated. The magnetic anisotropy of the SrRuO₃ film is controlled by the substrate. The magnetisation of SrRuO₃ is probed through charge transport measurements. It is shown that a magnetic field can be used to access numerous non-volatile states between two extrema. Through the addition of a heavy metal layer to the system, the effect of spin-orbit torque-induced magnetisation switching is also investigated. Using the spin Hall effect, out-of-plane magnetisation can also be controlled by the application of an in-plane electric current. The possibility of controllable reversal of a portion of the magnetisation through current in an analog fashion makes SrRuO₃/SrTiO₃ a promising building block for low power neuromorphic computing.

Stochastic Computing with Magnetic Nanowire Devices

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Neuromorphic computing at the edge of chaos with a single magnetic domain wall

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Classifying or predicting complex time-dependent signals (e.g. speech, financial data, the weather) is a challenging computational task. Reservoir computing (RC) is an efficient neuromorphic computing approach that is ideally suited to such tasks and is typically implemented in software using a recurrent neural network (RNN) with fixed synaptic weights (the reservoir) connected to a single, trainable readout layer. However, more efficient implementations of RC are possible if the software RNN reservoir is substituted with a physical system with the correct properties, such as non-linear response to input signals and inherent memory, leading to a readily deployable, hardware-based neuromorphic computing platform [1].

In this work, we propose a novel approach to RC where the dynamics of a single magnetic domain wall (DW) trapped between two defect sites in a nanostrip acts as a hardware-based reservoir. We demonstrate how such a device, with dimensions smaller than 1 μ m, is capable of performing complex data analysis tasks, such as speech recognition. We have modeled a Ni nanowire with two anti-notches using both a simple 1D model and micromagnetic simulations. Both models show the DW exhibit complex oscillatory dynamics similar to the Duffing oscillator, thus giving highly non-linear responses to applied magnetic fields. We exploit the DW dynamics for RC by using an applied field to inject time-multiplexed input signals into the reservoir and show how this approach allows the device to perform classification tasks. We have explored how the regime of applied fields affects the accuracy of sine and square wave classification, showing that the best recognition rate is obtained at the edge of a chaotic regime of oscillation. We have, also, demonstrated that the same approach can be used for more complex tasks, such as spoken digit recognition. Our work opens a new perspective for neuromorphic computing in nanomagnetic hardware.

[1] M. Riou et al., Phys. Review Applied, 2019

Easy-plane spin Hall oscillators as spiking neurons for neuromorphic computing

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Spintronic oscillators can be used to emulate neurons at nanoscale. They have recently demonstrated remarkable results in neuromorphic learning tasks [1-3]. However, the existing spintronic oscillators lack the key ingredient of biological neurons, which is to emit spikes when the membrane potential overcomes a threshold. Networks of spiking neurons, called *Spiking Neural Networks*, are more efficient because information can be encoded in the timing between spikes as well as in their rate. In addition, they can implement biologically inspired learning rules, such as the *Spike-Time-Dependant-Plasticity*, that enable unsupervised learning which is more difficult for artificial neural networks.

In this work, we have identified easy-plane spin-Hall oscillators as a novel configuration for spintronic spiking neurons. Voltage spikes are produced through the inverse spin-Hall effect when a spin current induces a single period of the in-plane-oscillation of the magnetic layer. An easy plane perpendicular to the magnetic film can be obtained by compensating the film's perpendicular anisotropy with the demagnetizing field at a set film thickness. Interestingly, such easy-plane spin-Hall oscillators have a particularly small damping and require very weak spin current drives [4].

We perform micromagnetic simulations and address all the necessary ingredients for the experimental implementation of learning with a network of such spintronic spiking neurons. First, we show that easy-plane oscillations can be obtained in a nanoconstriction and that spiking rate can be controlled through the input current. Second, we demonstrate the coupling of several oscillators in series by spin waves and we study their synchronisation properties. We then show that synaptic weights tuning the coupling between neurons can be implemented in two ways - by anisotropy modulation and by damping modulation. Finally, we investigate the spike propagation in such networks and show that brain-inspired learning rules such as *synfire chains* can be implemented with this system. Demonstrating numerically the feasibility of coupling easy-plane spin-Hall oscillators, this work opens a new path towards the experimental realization of spintronic spiking neural networks.

- [1] J.Torrejon et al, Nature 547, 7664 (2017)
- [2] M. Romera et al, Nature 563, 230 (2018)
- [3] D. Marković et al, Appl. Phys. Lett. 114, 012409 (2019)
- [4] B. Divinskiy et al, Nature Comm. 10, 5211 (2019)

Ultra-low power reservoir computing using voltage controlled superparamagnetic ensembles

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Reservoir computing has been gaining traction as a bio-inspired, machine-learning approach that is well-suited to solving technologically relevant problems, such as speech recognition and the prediction of complex time series [1]. Magnetic systems are well positioned to fulfill the requirements of the reservoir, providing the requisite non-linearity, fading memory, and reproducible responses. Furthermore, single magnetic elements subject to temporal input sequences can perform the entire role of the reservoir, replacing the complex networks of transistors that would be required for CMOS-based implementations [2]. This offers the potential for compact devices, with reduced energy costs for computation. Here, we propose superparamagnetic ensembles as ideal candidates for ultra-low-energy reservoirs where thermal noise is utilised to drive ensemble dynamics and control is provided by strain-mediated voltage inputs.

Here, we use an analytical model to describe response of a superparamagnetic ensemble [3], and show how it can be used as a physical reservoir. We also present micromagnetic simulations that validate the physical accuracy of the analytical model. By inputting temporal sequences via voltage controlled anisotropy and training a single layer of synaptic weights at the reservoir's output, we show that the reservoir can perform standard machine learning benchmarks with competitive performance. Figure 1 presents performance for both a spoken digit recognition task (94 % accuracy) and chaotic time series prediction (error comparable to current reservoir approaches). The reservoir can be tuned to operate on timescales from hundreds of nanoseconds up to seconds, which would allow it to be used to tackle a variety of real-time problems, from decision-making in driverless cars (fast) to speech recognition (slow). The low energy consumption expected for these thermally driven devices also makes them ideal candidates for edge computing, where there is a strong emphasis on minimising power consumption.



Figure 1: Reservoir computing with a model of an ensemble of superparamagnetic dots. Standard machine learning benchmarks are performed with competitive performance. **a.** Schema of reservoir computing. **b**. Schematic of the ensemble. **c**. Spoken digit recognition at 94% accuracy. **d**. Reproduction of a tenth order non-linear response to white noise (NARMA10).

[1] L. Appeltant, M. C. Soriano, G. Van der Sande, J. Danckaert, S. Massar, J. Dambre, B. Schrauwen, C. R. Mirasso, and I. Fischer, Nat Commun 2, 468 (2011).

[2] J. Grollier, D. Querlioz, K. Y. Camsari, K. Everschor-Sitte, S. Fukami, and M. D. Stiles, Nature Electronics 1 (2020).

[3] O. Hovorka, R. F. L. Evans, R. W. Chantrell, and A. Berger, Appl. Phys. Lett. 97, 062504 (2010).

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