Multifunctional (magnetic) nanostructures

Denys Makarov

Manipulation of the magnetic order parameter using external means

Multifunctional nanostructures with a focus on functions, devices, MEMS, sensors and actuators, energy harvesting etc.

Duration: 1.5h



Dresden, Germany





Dresden (~800 years old & ~500.000 inhabitants):

Capital of the Free State of Saxony

Scientific landscape:

Technical University of Dresden: about 40.000 students

Max Planck Institutes: 3 | Fraunhofer Institutes: 8 |

Leibniz Institutes: 3

Helmholtz Center: 1

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Helmholtz-Zentrum Dresden-Rossendorf (HZDR)

Research for the World of Tomorrow

RESEARCH AREAS

之4?	Established	1992 (1955)
	Member of Helmholtz Association	2011
	Base Budget ~ 12	20 Mio. €/a
	Employees	~ 1400
	10 Institutes 11 Junior Research Gr	oups
	Sites: Dresden, Leipzig Grenoble (FR), Hambu	g, Freiberg, ırg, Görlitz



NERGY

$\mathsf{HEALTH} (\rightarrow \mathsf{Oncology})$



MATTER (\rightarrow Materials)



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HZDR Facilities



Center for High-Power Radiation Sources







Ion Beam Center

Industry Services via



User Facilities



High-Magnetic Field Laboratory





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Activities on (magnetoelectric) antiferromagnets



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Nature Physics & Nature Electronics & Nature Communications & Phys. Rev. Lett. & Nano Letters & Advanced Materials <u>Review:</u> DM et al., Advanced Materials **34**, 2101758 (2022)





Nature Physics & Nature Electronics & Nature Communications & Phys. Rev. Lett. & Nano Letters & Advanced Materials <u>Review:</u> DM et al., Advanced Materials **34**, 2101758 (2022)

II. Flexible sensors & Flexible actuators









Science Advances & Nature Electronics & Nano Letters & Advanced Materials & npj Flexible Electronics & Nature Commun.

 Review:
 Santiago Canon et al., Adv. Funct. Mater. 31, 2007788 (2021)

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What is multifunctional material?

Multifunctional material is defined to be any material or material-based system which integrally combines two [or possibly more] properties, one of which is normally structural and the other functional, e.g. optical, electrical, magnetic, thermal etc...





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Multifunctional materials are the materials that perform multiple functions in a system due to their specific properties

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The applications of such new "smart" materials include energy, medicine, nanoelectronics, aerospace, defence, semiconductor, and other industries

Multifunctional materials can be both naturally existing and specially engineered

https://www.e-education.psu.edu/eme807/node/698

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What is the most well known natural multifunctional material?

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https://www.e-education.psu.edu/eme807/node/698

https://courses.lumenlearning.com/boundless-ap/chapter/the-skin/

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Do you know <u>not</u> multifunctional materials?





Semiconductor => electronics Suitable band gap => optics (detectors), communication

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Multifunctional materials



Smart materials

Multifunctional materials



Smart materials Intelligent materials Advanced Materials

Multifunctional materials



Smart materials Intelligent materials Advanced Materials

Multifunctional materials

Advanced functional materials

Multiferroic materials

Magnetoelectric materials

Magnetic composites



Smart materials Intelligent materials Advanced Materials

Multifunctional materials

Advanced functional materials

Multiferroic materials

Magnetoelectric materials

Magnetic composites

Most probably, these are (in some sense) synonyms



Multifunctional magnetic nanoparticles



Medical applications: hyperthermia (pre-clinical)

J. Gao et al., Acc. Chem. Res. 42, 1097 (2009)



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Nanosized magnetic objects

Electron beam lithography



Hu et al., J. Appl. Phys. 95 (2004) 7013

Challenges

- i. Expensive
- ii. Time consuming
- iii. Size limited

X-ray/nanoimprint lithography

Ross, *An. Rev. Mat. Res.* **31** ('01) 203; Chou et al., *J. Appl. Phys.* **76** ('94) 6673

As grown FePt $\emptyset = 5 \text{ nm}$ -10 Tbit/inch^2

Magnetic particles self-assembly

Sun et al., Science 287 (2000) 1989



!!! Agglomeration !!!

Dai et al., *NanoLett.* **1** (2001) 443

Magnetic films on nanospheres





Magnetic properties

- Perpendicular anisotropy
- Single-domain state
- Exchange isolated caps

Albrecht et al., Nature Materials 4 (2005) 203



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Driving force: magnetic recording and spintronics

L1₀ chemically ordered FePt alloy allows thermal stability for 3-nm-large grains

Single grain
Single bit => Patterned Magnetic Media



Manipulation of the (magnetic) order parameter



Effects of topology



wikipedia

pngkit.com

- Geometry
 - Symmetric boundary conditions (tubes and spheres)
 - Antisymmetric boundary conditions (Möbius rings and Klein bottles)
 - Shells with perforations...



https://tallbloke.wordpress.com/2014/03/21/p-a-semi-topology-of-the-curved-space-time-universe/



- Texture
 - Domain walls, skyrmions, vortices
 - Dislocations and disklinations (antiferromagnetic textures)

- Band structure
 - Topological insulators, Weyls...
 - 2D materials and heterostructures





Smeikal et al., Nat. Phys. (2018)

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Flexomagnetism, Magnetostriction...

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wikipedia

pngkit.com

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 - Antisymmetric boundary conditions (Möbius rings and Klein bottles)

Pylypovskyi et al., PRL (2015)

Curvilinear magnetism

Shells with perforations...

Topology of the curved space-time universe



https://tallbloke.wordpress.com/2014/03/21/p-a-semi-topology-of-the-curved-space-time-universe/



Texture

- Domain walls, skyrmions, vortices
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Smeikal et al., Nat. Phys. (2018)

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Planar vs 3D magnetic nanostructures



Information bits in perpendicular recording media



Higher areal density <=> Reduce the bit size



Bit size: 50 nm A: 10⁻¹¹ J/m M_S: 0.6 T

*K*_U: 1 MJ/m³

Piramanayagam, *J. Appl. Phys.* **102** (2007) 011301 Eisenmenger et al., *Nature Mater.* **2** (2003) 437 Criteria: Long time stability (~10 years)

[Anisotropy Energy] = 60 x [Thermal Energy]

$$K_U \cdot V = 60 \cdot k_B \cdot T$$



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3D curved magnetic shell structures

Cylindrical surfaces



Streubel et al., Nano Lett. (2012) & (2014) & Adv. Mat. (2014)

Curvature induced skyrmions on a sphere



Kravchuk et al., PRB (2016); PRL (2018)





Spherical surfaces

Albrecht et al., *Nat. Mater.* **4**, 203 (2005) Ulbrich et al., *PRL* (2006); DM et al., *APL* (2007)... Kravchuk et al., *PRB* **85**, 144433 (2012)





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Impact of curvature on a magnetic system

Magnetic interactions in the anisotropic Heisenberg ferromagnet:
Magnetic interactions in the anisotropic Heisenberg ferromagnet:

$$E = L \int_{S} \left[A \sum_{i=x,y,z} (\nabla m_i)^2 + K(\boldsymbol{m} \cdot \boldsymbol{n})^2 \right] dS$$
Exchange energy Anisotropy energy
In a curvilinear basis, micromagnetic energy can be rewritten:

$$\mathcal{E} = \left[\nabla \theta - \Gamma(\varphi) \right]^2 + \left[\sin \theta \left(\nabla \varphi - \Omega \right) - \cos \theta \frac{\partial \Gamma(\varphi)}{\partial \Gamma(\varphi)} \right]^2$$

$$\mathcal{E}_{ex} = \left[\mathbf{\nabla} \mathbf{\Theta} - \mathbf{\Gamma} (\mathbf{\varphi}) \right] + \left[\operatorname{sin} \mathbf{\sigma} (\mathbf{\nabla} \boldsymbol{\varphi} - \mathbf{\Omega}) - \cos \mathbf{\sigma} \right]$$
$$\mathcal{E}_{ex} = \mathcal{E}_{ex}^{0} + \mathcal{E}_{ex}^{A} + \mathcal{E}_{ex}^{D} \qquad \qquad \mathcal{E}_{ex}^{0} = (\mathbf{\nabla} \theta)^{2} + \sin^{2} \theta (\mathbf{\nabla} \varphi)^{2}$$

Induced anisotropy responses:

$$\mathscr{E}_{ex}^{A} = \mathbf{\Gamma}^{2} + \sin^{2}\theta\mathbf{\Omega}^{2} + \cos^{2}\theta(\partial_{\varphi}\mathbf{\Gamma})^{2}$$

Induced chiral responses:

$$\mathscr{E}_{ex}^{D} = D_{\alpha\beta\gamma}m_{\beta}\nabla_{\gamma}m_{\alpha}, \quad D_{\alpha\beta\gamma} = -D_{\beta\alpha\gamma}$$

Linear in curvature

Quadratic in

curvature

Gaididei et al., PRL ('14); Pylypovskyi, DM et al., PRL ('15); Kravchuk, DM et al., PRL ('18); Volkov, DM et al., PRL ('19)...

 $\mathscr{E}_{er}^{D} = -2\left[\left(\boldsymbol{\nabla}\boldsymbol{\theta}\cdot\boldsymbol{\Gamma}\right) + \sin\boldsymbol{\theta}\boldsymbol{\nabla}\boldsymbol{\varphi}\cdot\left(\boldsymbol{\Omega} + \cos\boldsymbol{\theta}\partial_{\boldsymbol{\varphi}}\boldsymbol{\Gamma}\right)\right]$

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Impact of curvature on a magnetic system



Spherical

surfaces

Cylindrical

surfaces

Magnetic interactions in the anisotropic Heisenberg ferromagnet:

$$E = L \int_{\mathcal{S}} \left[A \sum_{i=x,y,z} (\nabla m_i)^2 + K(\boldsymbol{m} \cdot \boldsymbol{n})^2 \right] d\mathcal{S}$$

Exchange energy Anisotropy energy

New approach to material science

designing magnetic responses by tailoring the geometry of thin films

Induced anisotropy responses:
$$\mathscr{E}_{ex}^{A} = \Gamma^{2} + \sin^{2}\theta\Omega^{2} + \cos^{2}\theta(\partial_{\varphi}\Gamma)^{2}$$
Quadratic in
curvatureInduced chiral responses: $\mathscr{E}_{ex}^{D} = D_{\alpha\beta\gamma}m_{\beta}\nabla_{\gamma}m_{\alpha}, \quad D_{\alpha\beta\gamma} = -D_{\beta\alpha\gamma}$ Linear in
curvature $\mathscr{E}_{ex}^{D} = -2\left[(\nabla\theta\cdot\Gamma) + \sin\theta\nabla\varphi\cdot(\Omega + \cos\theta\partial_{\varphi}\Gamma)\right]$ Linear in
curvature

Gaididei et al., PRL ('14); Pylypovskyi, DM et al., PRL ('15); Kravchuk, DM et al., PRL ('18); Volkov, DM et al., PRL ('19)...

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Experimental confirmation of curvature effects



Volkov et al., Phys. Rev. Lett. 123, 077201 (2019)

Magnetic Skyrmion states on a curvilinear defect



Kravchuk et al., *Phys. Rev. Lett.* **120**, 067201 (2018) Pylypovskyi et al., *Phys. Rev. Appl.* **10**, 064057 (2018)

Discovery of a non-local chiral effect in curvilinear ferromagnetic shells



Sheka et al., Communications Physics 3, 128 (2020)
Multiferroic materials



The behavior of any physical system from cosmological objects in field theories through living organisms in biology up to synthetic objects in condensed and soft matter is determined by the order parameter that lives in spacetime



The behavior of any physical system from cosmological objects in field theories through living organisms in biology up to synthetic objects in condensed and soft matter is determined by the order parameter that lives in spacetime

Order parameter of magnetic materials: magnetization

In ferromagnets, order parameter brakes the time reversal symmetry



The behavior of any physical system from cosmological objects in field theories through living organisms in biology up to synthetic objects in condensed and soft matter is determined by the order parameter that lives in spacetime

Order parameter of magnetic materials: magnetization

In ferromagnets, order parameter brakes the time reversal symmetry

There are 4 ferroic order parameters



time	invariant	change
invariant	ferroelastic	ferroelectric ⊕⊕ ⊕⊕
change	ferromagnetic	ferrotoroidic

Туре	Micro. property	Macro. property	Force field
Ferroelastic	Deformation	Strain	Stress
Ferromagnetic	Magnetic moment	Magnetization	Magnetic field
Ferroelectric	El. dipole moment	Polarization	Electric field
Ferrotoroidic	Magnetic vortex	Toroidization	Toroidal field

N. A. Spaldin, M. Fiebig & M. Mostovoy

The toroidal moment in condensed-matter physics and its relation to the magnetoelectric effect J. Phys.: Condens. Matter **20**, 434203 (2008)

Manfred Fiebig: https://www.hikari.uni-bonn.de/research/multiferroics/new-forms-of-ferroic-order



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A material is termed as ferroic if there is a spontaneous property, called the order parameter, that arises in uniform alignment of some microscopic property of the unit cell throughout macroscopic regions of a crystal

Manfred Fiebig: https://www.hikari.uni-bonn.de/research/multiferroics/new-forms-of-ferroic-order

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The order parameter O can be switched into at least two different directions by some external field A



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The order parameter O can be switched into at least two different directions by some external field A

This works because a ferroic contributes a term F = -OA

to the free energy so that an alignment of O parallel to A minimizes this energy



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Regions with different orientation of the order parameter can coexist. They are called domains

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Long-range ordering is present below a critical temperature only



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Ferroelectric barium titanate: BaTiO₃

parent phase: ideal cubic ABO₃ perovskite structure => no electric polarisation

ferroelectric phase: the Ti⁴⁺ ion is shifted away from the center of the octahedron causing a polarization (only one ferroic order; no magnetism; empty *d* shell)



https://blogs.hrz.tu-freiberg.de/pyro/pyroelektrizitaet/



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Mutiferroic BiFeO₃ (BFO)

In BFO, the ferroelectric displacement is driven by the A-site cation, and the magnetism arises from a partially filled *d* shell on the B site

In BFO, the A-site cation (Bi³⁺) has a so-called stereochemically active $6s^2$ lonepair of electrons, and off-centering of the A-site cation is favoured by an energylowering electron sharing between the formally empty A-site 6p orbitals and the filled O 2p orbitals



Spin transfer torque magnetic random access memory (STT-MRAM) is among the most promising spintronic memory for computing

Still, it suffers from resistance scalability (resistance scales inversely proportional to the area and drive current densities) and intrinsically high energy/bit operation (>100 fJ/bit)

https://onlinelibrary.wiley.com/doi/full/10.1002/adma.202001943



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Magneto-electric memory can operate with capacitive displacement charge and potentially reach 1-10 aJ/switching operation

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Magneto-electric memory can operate with capacitive displacement charge and potentially reach 1-10 aJ/switching operation



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Magneto-electric memory can operate with capacitive displacement charge and potentially reach 1-10 aJ/switching operation

1000 (C) La % Energy (µJ/cm² 100 10 BFO 0% 10% 15% 20% 10 100 MF film thickness (nm)

Bi_{1-x}La_xFeO₃ thin films

https://onlinelibrary.wiley.com/doi/full/10.1002/adma.202001943

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>60 years of material science research

Only 1 mutiferroic which is potentially relevant for spintronic applications: BiFeO₃ (BFO)



Spin transfer torque magnetic random access memory (STT-MRAM) ><u>100 fJ</u> per switching



Magnetoelectric memory can operate with capacitive displacement charge and *potentially* reach <u>1-10 aJ</u> per switching operation

S. Manipatruni, D. E. Nikonov, C. C. Lin et al. Scalable energy-efficient magnetoelectric spin–orbit logic Nature 565, 35 (2019)



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Multiferroics based on magnetic materials



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Magnetoelectric composites

BaTiO₃ (BTO) & CoFe₂O₄ (CFO): ferroelectric and ferrimagnetic layers



Magnetoelectric composites

BaTiO₃ (BTO) & CoFe₂O₄ (CFO): ferroelectric and ferrimagnetic layers CFO: large magnetostriction | CFO and BTO: high Curie temperatures BTO/CFO composite gives large strain-mediated ME response



Wireless deep brain stimulation in freely moving mice





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Wireless deep brain stimulation in freely moving mice



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Magnetic stimulation modulates neural activity in mice



Dynamic movement parameters



Static and balance-related movement parameters



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Mutiferroic vs Magnetoelectric materials

Multiferroics are defined as materials that exhibit more than one of the primary ferroic properties in the same phase



Mutiferroic vs Magnetoelectric materials

Multiferroics are defined as materials that exhibit more than one of the primary ferroic properties in the same phase

In magnetoelectric materials electric field modifies the magnetic properties and vice versa



Mutiferroic vs Magnetoelectric materials

Multiferroics are defined as materials that exhibit more than one of the primary ferroic properties in the same phase

In magnetoelectric materials electric field modifies the magnetic properties and vice versa

All ferromagnetic ferroelectric multiferroics are linear magnetoelectrics

Magnetoelectric materials are not necessarily multiferroic



Free energy expansion in powers of E and H according to Landau & Lifshitz (1958):





Free energy expansion in powers of *E* and *H* according to Landau & Lifshitz (1958):



Linear magnetoelectric effect

 Only allowed if time- & space inversion symmetries individually broken

α_{ij}

Magnetoelectric tensor



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I. Dzyaloshinskii, ZETF 37, 881 (1959)





Linear magnetoelectric effect

Only allowed if time- & space inversion symmetries individually broken

 α_{ii}

Magnetoelectric tensor



X. He et al., Nature Mat. 9, 579 (2010)

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I. Dzyaloshinskii, ZETF 37, 881 (1959)





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Magnetoelectric RAM based on Pt/Cr₂O₃



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Magnetostrictive materials



Magnetism and mechanical deformation

Magnetostriction: change of the shape or dimensions of a magnetic material upon magnetisation (in an applied magnetic field)



Magnetism and mechanical deformation

Magnetostriction: change of the shape or dimensions of a magnetic material upon magnetisation (in an applied magnetic field)

Anisotropic magnetostriction (Joule magnetostriction): elongation/contraction (for positive/negative magnetostrsiction coefficient) of the material in an applied field





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Magnetism and mechanical deformation

Magnetostriction: change of the shape or dimensions of a magnetic material upon magnetisation (in an applied magnetic field)

Anisotropic magnetostriction (Joule magnetostriction): elongation/contraction (for positive/negative magnetostrsiction coefficient) of the material in an applied field

Typically small: relative elongation is in the order of 10×10^{-6}

Terfenol-D (TbDyFe₂ alloy): about 2000 x 10⁻⁶ at 2 kOe and room temperature

FeAI alloy: about 300 x 10⁻⁶ at 0.2 kOe

https://www.tf.uni-kiel.de/servicezentrum/neutral/praktika/anleitungen/m205



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Magnetostiction effects on magnetic textures



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Impact of strain effects on spin-orbit torques



Filianina et al., *Phys. Rev. Lett.* **124**, 217701 (2020) & *APL* **118**, 032401 (2021)

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DRESDEN concept

Magnetostriction: applications



[1] https://www.powertransmissionworld.com/a-new-contacless-magnetostrictive-position-transducer/

[2] https://news.vidyaacademy.ac.in/wp-content/uploads/2019/02/Active-sonar.jpg

[3] https://techblog.ctgclean.com/2012/01/ultrasonics-transducers-magnetostrictive-hardware/

[4] https://aip.scitation.org/doi/10.1063/1.4943770

Magnetostrictive sonar

Magnetic composites for sensors and actuators

Magnetic particles and polymers



Multifunctional magnetic nanoparticles

Lecture by Annelies Coene



J. Gao et al., Acc. Chem. Res. 42, 1097 (2009)



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Magnetic particles and polymers



NdFeB - hard permanent magnet

Primarily microparticles



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PDMS: polydimethylsiloxane



https://pubs.rsc.org/en/content/articlelanding/2017/ta/c7ta04577h



Elongation: 50%; Young's modulus: 1 MPa

www.siliconeall.com/product/html/?637.html



Biocompatible rubber

Cosmetics, fluidics, smart skins, smart textiles...



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Hard magnetic composite: NdFeB in PDMS

NdFeB microparticles (5 um diameter) in PDMS



X. Wang et al., Communications Materials 1, 67 (2020)



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Mechanically soft magnetically hard actuators



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Durability of magnetic soft actuators

X20 slower



Test start



After 2.1 million cycles

X. Wang et al., *Communications Materials* **1**, 67 (2020)



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Interaction with delicate objects

80 times slower



X. Wang et al., *Communications Materials* **1**, 67 (2020)

Magnetic composites for sensor applications



Spintronics



A.Barthélémy et al., JMMM 242-245, 68 (2002)



Spintronics



A.Barthélémy et al., JMMM 242-245, 68 (2002)



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Spintronics



A.Barthélémy et al., JMMM 242-245, 68 (2002)







Giant magnetoresistive (GMR) sensors

20 to 50 doublelayers of ferro- and nonmagnetic conductors

Current-in-plane measurement

Sensitive to in-plane fields

<u>**G**</u>iant <u>**M**</u>agneto-<u>**R**</u>esistance

$$GMR(B_{ext}) = [R(B_{ext}) - R_{sat}] / R_{sat}$$



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M. Ha et al., Adv. Mater. 33, 2005521 (2021) & E. S. Oliveros Mata et al., Appl. Phys. A 127, 280 (2021)

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M. Ha et al., Adv. Mater. 33, 2005521 (2021) & E. S. Oliveros Mata et al., Appl. Phys. A 127, 280 (2021)

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M. Ha et al., Adv. Mater. 33, 2005521 (2021) & E. S. Oliveros Mata et al., Appl. Phys. A 127, 280 (2021)

0

16 µm

Bending

radius

5

Planar

- Planar

1.8

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З

Bending Radius (mm)

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Scrolling documents



(W), while all the orientations being verified by a reference compose and recorded in a video (Fig. 3b and Sapplementary Video 4). From the video we selected several representative frames (N, S and W), which are shown in Fig. 3c- redoctive vide a sugremposed dual indicating the current heading of the person. These results show an on skin device that can reglicate the functionality of a compase and

enable artificial magnetoreception for humans. Geomagnetic virtual reality control

Another application area where we envisage the potential of the e-skin compass is augmented or virtual reality (VR). In this case, the e-skin compass will act as a mechanically compliant interactive input device capable of directly translating the real world magnetoreception into the virtual realm. To evaluate the functionality of the e-skin compass within a virtual reality environment, we set up eriment where we used the output voltage of the compass to an exp control the orientation of a virtual panda inside Panda3D, a Pythonbased game engine". First, the compass was placed on a person's middle finger to define an axis of directionality. Next, the panda was com nded to move forward at a constant speed within a program, while its rotation angle was given by the movement of the person's hand in the geomagnetic field (Supplementary Fig. 8). By noving the axis of directionality, the person could control at will the trajectory of the panda in the virtual environment without the aid of any permanent magnet or optical sensory system, as typically used in VR applications. The entire experiment was recorded in a video (Supplementary Video 5) from which we selected and superimposed three representative frames, as shown in Fig. 4. On the ower right, a compass drawing is included to indicate the physical location of north during the expe

Here, the trajectory of the panda is highlighted as a dotted line and the frames of interest are correspondingly labelled from 1 to 3.

Scrolling up

Methods 15-Max compose tabelication. Gluon dides of 22% 22 mm/ (VWR International) were put cound with Nobilamethyliotane (PDOSS, Sigged 14, mm 110) at CMP rays for No and cound at DVV. for 45 mm. Separathy, a 30 primat mathematical and the second second at DVV. Second at DVV and the Signal mathematical and the composite of the years of defense transmission and here interorings. Also presented mathematical gluons were and a subserving for relation. The resulting Multic convent gluons were used as substration for preparing the e-data compose devects Streetmentum: Fig. 10.

Through you be performed over the Multi rule wind SM13 (Major, UK) the breast one and a set of SM12 (Kajor, UK) the transmission and AMOPT gas. If N van der cort at 110 °C 2 × 2 mm. After caring, the photoresent films were capsued using a direct later white (IW16A) with the distribution of the set of SM13 (Major, SM22) with the distribution of the set of SM13 (Major, SM22) with the distribution of the set of SM12 (Major, SM22) with the distribution of SM12 (Major, SM22) with the distrest distribution of SM12 (Major, SM22) with the distribution

Complete an equal and generating constraining optimizations. It to charge to a final optimization of the strain o

Interacting with a map



M. Ha et al., Adv. Mater. 33, 2005521 (2021) & E. S. Oliveros Mata et al., Appl. Phys. A 127, 280 (2021)

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Magnetic heterostructures





Heterostructures

Heterostructures show different synergistic relations between two or more building blocks that improve functional characteristics

Each component plays a complementary role in producing multifunctionality





Heterostructures

Heterostructures show different synergistic relations between two or more building blocks that improve functional characteristics

Each component plays a complementary role in producing multifunctionality

Heterostructures consist of combinations of different materials, which are in contact through at least one interface

Magnetic heterostructures combine different physical properties which do not exist in nature

Editors: Hartmut Zabel and Samuel D. Bader Magnetic Heterostructures: Advances and Perspectives in Spinstructures and Spintransport Springer Trends in Modern Physics (2008)



Heterostructures



New J. Phys. 16, 043008 (2014)



https://pubs.rsc.org/en/content/articlelanding/2021/mh/d0mh01356k



https://www.eurekalert.org/news-releases/776488



https://www.nature.com/articles/nature12385



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Heterostructures of ferro- and antiferromagnets Exchange bias effect



Magnetic sensors based on magnetoresistive effect

Read heads in magnetic data storage







Magnetic Field

GMR sensor elements:

- Resistance change in small external fields
- Antiparallel orientation of the magnetic moment increases resistance
- Sensing layer is a soft F layer
- Reference layer pinned by exchange bias



S. S. P. Parkin, Annu. Rev. Mater. Sci. 25 (1995) 357

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Exchange bias effect: coupling between F and AF layers



Characteristic features:

- A. AF layer has to be cooled in a magnetic field
- B. Hysteresis loop is shifted and broadened
- C. Asymmetric magnetization reversal processes
- D. Effect is strongly temperature dependent



A. E. Berkowitz and K. Takano, J. Magn. Magn. Mater. **200** (1999) 552 B. K. O'Grady et al., J. Magn. Magn. Mater. **322** (2010) 883

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Integral magnetic investigations of EB effect





Exchange bias measurements:

- (1) Warm sample to 320 K and apply H_{cool} = 70 kOe
- (2) Cool sample to the measurement temperature
- (3) Measure hysteresis loop and acquire coercive fields

 $H_{\rm C} = (H_{\rm C}^{\rm R} - H_{\rm C}^{\rm L}) / 2$ $H_{\rm EB} = (H_{\rm C}^{\rm R} + H_{\rm C}^{\rm L}) / 2$

K. O'Grady et al., J. Magn. Magn. Mater. **322** (2010) 883 M. D. Stiles and R. D. McMichael, Phys. Rev. B **63** (2001) 064405



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Integral magnetic investigations of EB effect



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IrMn/(Co/Pt)₄ exchange bias system

Probe coupling at nm scales Distinct behaviour of bulk and interface in 6 nm IrMn



Field-cooled from 45°C to 7°C



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Heterostructures of ferro- and non-magnetic materials Giant magnetoresistive effect



Anisotropic magnetoresistance (AMR): about 2% effect

AMR: electrical conductivity of ferromagnetic materials depends on the orientation of the magnetization with respect to the direction of the flowing current

Observed in 1856 by W. Thomson (Lord Kelvin)

W. Thomson, Proc. Roy. Soc. 8, 546 (1856/1857)



Explained by N.F. Mott in 1936: spin-orbit interaction needs to be accounted for

N.F. Mott, Proc. Roy. Soc. (London), Ser. A 153, 699 (1936)



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Giant magnetoresistance (GMR): about 100% effect

GMR: change of the resistance of the magnetic multilayer stack in magnetic field

Application relevant for magnetic sensor devices

Typical GMR values: in the range of 20-100%

Current-in-plane (CIP) measurement scheme



S.S.P. Parkin, Annu. Rev. Mater. Sci. 25, 357 (1995)



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Interlayer exchange coupling

Kerr microscopy image of the Fe/Cr bilayer with continuously varying Cr layer thickness



Lecture notes of the 40th IFF spring school: *Spintronics* (2009)

Dependent on the thickness of the spacer layer:

Ferromagnetic coupling

Antiferromagnetic coupling

 90° coupling

Oscillating F-AF coupling dependent on the thickness of the spacer



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Tuning the sensitivity of the GMR sensor



Microscopic origin of GMR



Following the two-current model by Mott

N.F. Mott, Proc. Roy. Soc., Ser. A 153, 699 (1936)

Spin-dependent scattering at the interfaces

Scattering causes electrical resistance

Scattering inside the interlayer is neglected

Rates of spin-dependent and spin-independent scattering are the same

One scattering event contribute to the total resistance by an amount *r*

 $R_{\rm P} = 2r \ge 4r / (2r + 4r) = 8r/6$

 $R_{\rm AP} = 3r \times 3r / (3r + 3r) = 9r/6$

 $\Delta R/R_{\rm P}$ = 12.5% (max in experiment = 17%)

Lecture notes of the 40th IFF spring school: *Spintronics* (2009)

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Interlayer exchange coupling

Phenomenological description Lecture notes of the 40th IFF spring school: *Spintronics* (2009)

Interlayer exchange coupling energy: $E_{\text{coupl}} = -J_1 \cos(\theta) - J_2 \cos^2(\theta)$

with θ – angle between the magnetizations of the films on both sides of the spacer layer

 J_1 – bilinear coupling constant; J_2 – biquadratic coupling constant

Parameters J_1 and J_2 determines the strength and type of the coupling

If $J_1 >> J_2$ and J_1 is positive (negative) then the coupling will be ferromagnetic (antiferromagnetic) If $J_2 >> J_1$ and is negative, 90° coupling is favorable



Heterostructures of ferro- and non-magnetic materialsDzyaloshinskii–Moriya interactionPhysique 20, 817 (2019)



Heterostructures of ferro- and non-magnetic materials Perpendicular magnetic anisotropy AIP Advances 2, 042182 (2012)



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3D system: a magnetic phase transition occurs at a finite temperature



https://doi.org/10.1007/978-3-319-17897-4_17

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3D system: a magnetic phase transition occurs at a finite temperature

1D system: long-range order is possible only at T = 0 Proc. Camb. Philos. Soc. 32, 477 (1936)

https://www.nature.com/articles/s41565-019-0438-6



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3D system: a magnetic phase transition occurs at a finite temperature

1D system: long-range order is possible only at *T* = 0 *Proc. Camb. Philos. Soc.* **32**, 477 (1936)

2D system: the existence of magnetic long-range order at any finite temperature crucially depends on the number *n* of relevant spin components, usually called spin dimensionality, and determined by the physical parameters of the system (for example, the presence and strength of magnetic anisotropy)

https://www.nature.com/articles/s41565-019-0438-6





Spin dimensionality n = 1: the system has a strong uniaxial anisotropy and the spins point in either of the two possible orientations ('up' or 'down') along a given direction

https://www.nature.com/articles/s41565-019-0438-6





Spin dimensionality n = 1: the system has a strong uniaxial anisotropy and the spins point in either of the two possible orientations ('up' or 'down') along a given direction

Spin dimensionality n = 2 corresponds to an easy-plane anisotropy that favours the spins to lie in a given plane, although the orientation within the plane is completely unconstrained.

https://www.nature.com/articles/s41565-019-0438-6





Spin dimensionality n = 1: the system has a strong uniaxial anisotropy and the spins point in either of the two possible orientations ('up' or 'down') along a given direction

Spin dimensionality n = 2 corresponds to an easy-plane anisotropy that favours the spins to lie in a given plane, although the orientation within the plane is completely unconstrained.

Isotropic systems are characterised with n = 3: there is no constraint on the direction of spins

https://www.nature.com/articles/s41565-019-0438-6



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Mermin-Wagner(-Hohenberg) theorem

Phys. Rev. Lett. **17**, 1133 (1966) *Phys. Rev.* **158**, 383 (1967)

Thermal fluctuations destroy long-range magnetic order in 2D systems at any finite temperature when the spin dimensionality is 3 (isotropic Heisenberg model)



There is no isotropic 2D ferromagnet: long-wavelength excitations (spin waves) can be excited at any finite temperature as there is no gap in the spin wave spectrum (no anisotropy)

https://www.nature.com/articles/s41565-019-0438-6



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2D magnet with n = 2

These systems are described by the so-called XY model

They do not possess a conventional transition to long-range order

The susceptibility diverges below a finite temperature



Berezinskii, Kosterlitz and Thouless pointed out that this divergence is associated with the onset of topological order, characterized by an algebraic decay of spin correlations and by the presence of bound pairs of vortex and antivortex arrangements of spins

Sov. Phys. JETP-USSR **32**, 493 (1971) J. Phys. C. **6**, 1181 (1973)

Below the Kosterlitz–Thouless temperature T_{KT} , quasi-long-range magnetic order is established, and the existence of a finite order parameter is suppressed only marginally with the system size

https://www.nature.com/articles/s41565-019-0438-6



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Crl3

- Semiconducting layered vdW material
- Undergoes low-temperature magnetic transitions
- Exhibits different forms of magnetic order

Magnetization and magnetic susceptibility measurements show that bulk Crl₃ is a strongly anisotropic ferromagnet below the Curie temperature ($T_c = 61$ K), with its easy axis pointing perpendicular to the layers, and a saturation magnetization consistent with a spin S = 3/2 state of the Cr atoms



Nature 546, 270-273 (2017)



https://www.nature.com/articles/s41565-019-0438-6

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https://www.nature.com/articles/s41565-019-0438-6

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Heterostructures with semiconducting TMDCs

Heterostructure of a ferromagnetic semiconductor Crl₃ and a monolayer of WSe₂



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2D materials based magnetic tunnel junctions

Fe₃GeTe₂ / hBN / Fe₃GeTe₂ stack

Metallic 2D ferromagnet: Fe₃GeTe₂



Nano Lett. 18, 4303 (2018)



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2D materials based magnetic tunnel junctions

Fe₃GeTe₂ / hBN / Fe₃GeTe₂ stack Metallic 2D ferromagnet: Fe₃GeTe₂



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2D materials based magnetic tunnel junctions



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Magnetic Material – Topological Insulator Heterostructures



Magnetic topological insulators

Pathways for magnetizing topological insulators



 $(Cr_x, Bi_{1-x})_2Te_3$

MnBi₂Te₄

 Bi_2Te_3 - $Cr_2Ge_2Te_6$

https://onlinelibrary.wiley.com/doi/full/10.1002/adma.202007795

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Heterostructure: ferromagnet and topological insulator



Magnetic proximity effect has a relatively short length scale (few Å). Hence, time-reversal symmetry is broken only at the interface of the TI and MM, and not in the bulk of the TI.

Therefore, typically TI must be sandwiched in between two layers of magnetic insulator (MI) with perpendicular magnetic anisotropy

Sci. Adv. 5, eaaw1874 (2019)

https://onlinelibrary.wiley.com/doi/full/10.1002/adma.202007795



Anomalous Hall effect in FM/TI heterostructures



a) p-type 16 quintuple layer (QL) (Bi_{0.25}Sb_{0.75})₂Te₃–MnTe heterostructure and d) n-type 16 QL Bi₂Te₃–MnTe heterostructure

Nano Lett. 20, 1731 (2020)



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[magnetic material] / TI heterostructures

Magnetic met	tal - TI heterostructure					
[118]	AFMM	CrSb	$Cr_x(Bi_{1-\gamma}Sb_{\gamma})_{2-x}Te_3$	-	35 K	Neutron reflectometry, magnetometry, magneto-transport
[121]	AFMM	CrSb	$(Bi_{1-\gamma}Sb_{\gamma})_{2}Te_{3}$	-	90 K	Magneto-transport
[206]	AFMM	CrSe	$(Bi_{1-\gamma}Sb_{\gamma})_{2}Te_{3}$	Ξ.	120 K	Magneto-transport, Magnetometry, Neutron reflectometry
			and many mo	ore		
Magnetic met	tal - TI heterostructure					
[118]	AFMM	CrSb	$Cr_x(Bi_{1-\gamma}Sb_{\gamma})_{2-x}Te_3$	-	35 K	Neutron reflectometry, magnetometry, magneto-transport
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			and many mo	ore		



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Thank you for your attention

