All-optical Control of Magnetism I (including pump-probe techniques) Theo Rasing

Radboud University Nijmegen Institute for Molecules and Materials



: ship to Boston





Controlling magnetism by lightning

Controlling magnetism by light!

How does it work? Can we control magnitude? Can we control direction? Can we switch? What about the nanoscale?

Lecture topics:

- 1. Time scales and stimuli in magnetism
- 2. Laser induced effects
 - a. Thermal effects
 - **b.** Nonthermal opto-magnetic effects
- 3. Experiments
 - a. AOS of Ferrimagnets
 - **b. AOS of Ferromagnets**
 - c. AOS of Dielectrics
- 4. Towards applications
 - a. AOS at the nanoscale
 - **b.** Neuromorphic applications
- 5. Outlook









Lecture topics:



- 1. Introduction: stochastic/deterministic dynamics
- 2. Stroboscopic imaging.
- 3. Magneto-optical setupsa. Faraday/Kerr effectsb.XMCD
- 4. Examples
- 5. Outlook





How much is 1 fs in magnetism?



How much is 1 fs in technology?



stochastic/deterministic dynamics



NUCLEATION OF MAGNETIC ORDER COHERENT ROTATION OF MAGNETIZATION

Leland Stanford



Edward Muybridge: The Horse in Motion



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Experimental fs-pump-probe technique

All-optical pump-probe technique

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Ti:Sapphire laser 100fs pulses & amplifier system 20µJ/pulse (DANGER) 1kHz rep. rate Magnet Pump Probe Sample O.T.H.M. O.67 15 Detector

Revealing ultrafast dynamics with fs flashes





SHG studies of laser-induced surface melting



1. Time-scales in magnetism vs switching mechanisms



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Oersted/Faraday: electrical current creates magnetic field



Current creates a magnetic field

optical switch for short current pulses:



10 ps rise time 400 ps decay time **short magnetic field pulses!**







Switching: pulse length=half a precessional period!

Switching by controlling pulse width!

Switching within 200 ps !



Th.Gerrits et al, Nature 418, 2002

Magnetism and light

Magnetism and light







they rendered by their researches into phenomena" (together with Lorentz)5

Magnetism and light





Zeeman effect



The Nobel Prize in Physics 1902

in recognition of the extraordinary service they rendered by their researches into the influence of magnetism upon radiation phenomena" (together with Lorentz)



Magnetism and light inverse?





Faraday effect

2. Ultrafast "demagnetization" by light







FIG. 2. Transient remanent longitudinal MOKE signal of a Ni(20 nm)/MgF₂(100 nm) film for 7 mJ cm⁻² pump fluence. The signal is normalized to the signal measured in the absence of pump beam. The line is a guide to the eye.

VOLUME 76, NUMBER 22 PHYSICAL REVIEW LETTERS 27 MAY 1996

Ultrafast Spin Dynamics in Ferromagnetic Nickel

E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot Institut de Physique et Chimie des Matériaux de Strasbourg, Unité Mixte 380046 CNRS-ULP-EHICS, 23, rue du Loess, 67037 Strasbourg Cedex, France (Received 17 October 1995)

2-Temperature model



2-Temperature model



$$\begin{split} C_{\rm el}(T_{\rm el}) \, \frac{\partial T_{\rm el}}{\partial t} &= \nabla(\kappa \nabla T_{\rm el}) - g(T_{\rm el} - T_{\rm ph}) + S(z, t) \\ C_{\rm ph}(T_{\rm ph}) \, \frac{\partial T_{\rm ph}}{\partial t} &= g(T_{\rm el} - T_{\rm ph}) \end{split}$$



Coupled diffusion equations for T_{el} and T_{ph} note that $C_{el} << C_{ph}$ – this is why electrons get so hot!

2-Temperature model



$$C_{\rm el}(T_{\rm el}) \frac{\partial T_{\rm el}}{\partial t} = \nabla(\kappa \nabla T_{\rm el}) - g(T_{\rm el} - T_{\rm ph}) + S(z, t)$$
$$C_{\rm ph}(T_{\rm ph}) \frac{\partial T_{\rm ph}}{\partial t} = g(T_{\rm el} - T_{\rm ph})$$



FIG. 2. Transient remanent longitudinal MOKE signal of a Ni(20 nm)/MgF₂(100 nm) film for 7 mJ cm⁻² pump fluence. The signal is normalized to the signal measured in the absence of pump beam. The line is a guide to the eye.

Coupled diffusion equations for T_{el} and T_{ph} The signal is normal of pump beam. The note that $C_{el} << C_{ph}$ – this is why electrons get so hot!



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 $\mathbf{H}_{\rm eff} = \mathbf{H}_{\rm ext} + \mathbf{H}_{\rm A} + \mathbf{H}_{\rm D}.$



 $\mathbf{H'}_{\rm eff} = \mathbf{H}_{\rm ext} + \mathbf{H'}_{\rm A} + \mathbf{H'}_{\rm D}.$

 $\omega = \gamma \left(H_A + H_0 \right)$ 250 Oe 4 500 Oe 750 Oe 1000 Oe Frequency (GHz) 3-1250 Oe 2 Hz 300 600 900 1200 0 Magnetic field (Oe) anisotropy field 400 Ó 200 600 800 1000 1200 Delay (ps)

thin film of magnetic garnet ($\sim Y_3 Fe_5 O_{12}$)

Time-resolved Magneto-optical Imaging


Propagation Dynamics of Optically-excited Spin Waves



YIG sample





Y. Hashimoto et al., Nature Communications 8, 15859 (2017)





Optical charge transfer transition near 400 nm (3.1 eV)



TmFeO₃: antiferromagnetic resonance



and R. V. Mikhaylovskiy6*

All Optical Switching (AOS) by femtosecond laser pulses:





All-optical Control of Magnetism II (including pump-probe techniques) Theo Rasing

Radboud University Nijmegen Institute for Molecules and Materials



Changes in society

Happening April 2005

Same Happening March 2013





Changes in society

Connected Devices

March 2013







increase FUNCTIONALITY

Lots of data = Lots of energy GOOGLE You Hotel Palacky, Brno, Czech Republic Solution Hotel Palacky, Brno, Czech Republic Solution Hotel Palacky, Brno, Czech Republic

7% of electricity produced in the world



Google (The Netherlands)



Facebook (Sweden)



AOS by femtosecond laser pulses: *counterintuitive?*

Simple single spin problem



$$\frac{\partial S_i}{\partial t} = -\gamma S_i \times \mathbf{B}_{eff} + \frac{\lambda}{(S_i)^2} S_i \times (S_i \times \mathbf{B}_{eff})$$

Intuitive estimate:

If 100 fs pulse reverses the magnetization, it should act as an effective magnetic field of about 90 Tesla (γ =28 GHz/T)!



Light acts as a magnetic field, which is either strong (>>1 Tesla) or stays long (>>100 fs).

Why?

Thermodynamics of laser-matter interaction $W = \varepsilon \varepsilon_0 E(\omega) E^*(\omega)$

$$H(0) = -\frac{1}{\mu_0} \frac{\partial W}{\partial M(0)} = -\frac{\varepsilon_0}{\mu_0} E(\omega) E^*(\omega) \frac{\partial \varepsilon}{\partial M}$$
$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_{xx} & -i\alpha M & 0\\ +i\alpha M & \varepsilon_{yy} & 0\\ 0 & 0 & \varepsilon_{zz} + o(M^2) \end{pmatrix}$$



$$\vec{H}(0) = \frac{\varepsilon_0}{\mu_0} \alpha \left[\vec{E}(\omega) \times \vec{E}^*(\omega) \right]$$

Inverse Faraday effect

Pitaevskii, *Sov. Phys. JETP* **12**, 1008 (1961). van der Ziel *Phys. Rev. Lett.* **15**, 190 (1965).

(see further lecture on Magneto Optics by Prof. Schaefer)



Ultrafast excitation of spins via IFE in DyFeO₃

(all-optical spin resonance)





A.V. Kimel, A. Kirilyuk, P.A. Usachev, R.V. Pisarev, A.M. Balbashov, and Th. Rasing, **Nature 435**, 655-657 (2005)

Ultrafast excitation of spins via IFE in DyFeO₃

(all-optical spin resonance) σ^{+} 0.2 Faraday rotation (deg) 0.1 ps σ 0.1 photons 0.1 ps **DyFeO**₃ T = 95 K 30 0 15 45 60 Time delay (ps) A.V. Kimel, A. Kirilyuk, P.A. Usachev, R.V. Pisarev,



Amplitude of the laser-induced spin-waves



Inverse Faraday effect

$$H_{eff}(0) = \alpha \frac{\varepsilon_0}{\mu_0} E(\omega) E^*(\omega)$$

Fields up to 5 T! (even up to 20 T!)

Controlling ?



Double pump coherent control

AOS of Ferrimagnetic Metals

Femtosecond laser reversal of magnetization?





Polarization microscope + pulsed laser Circularly polarized 40fs laser pulses 20 nm GdFeCo film **Magneto-Optical** microscope 58



Reversal by 40fs laser pulsen!



switching of magnetization by single pulse!

Sweeping the pulsed laser beam at high speed across the sample



C.D. Stanciu et al., PRL 99,047601 (2007)

Femtosecond Magnetic Recording in GdFeCo!

C.D. Stanciu et al., *patent* #P77323PC00, PRL **99**, 047601 (2007)

AOS: role of light helicity/intensity



PRL 108, 127205 (2012)

PHYSICAL REVIEW LETTERS



Femtosecond laser reversal: role of exchange?



2-Temperature model



Temperature dominated: T >> T_{Curie}~100 fs



Bloch relaxation

$$dS_i/dt = -S_i/\tau_i$$

$$\tau_i = \mu_i / (2\alpha_i \gamma k_B T)$$

Dynamics scales with magnetic moment

$$\mu_2 < \mu_1 \Rightarrow \tau_2 < \tau_1$$

Distinct dynamics of sublattices!

Exchange dominated: $T < T_{Curie}$ $t \sim 1 ps$





Conservation total angular momentum Ground state AFM, transient FM!

How to probe? Element specific view: XMCD!



(for more details: see lecture Prof. Luning)

Femtosecond-XMCD!



Laser heat induced magnetization reversal!





 $dS_1/dt = -dS_2/dt$

T. Ostler et al, Nature Comm.3, 666, 2012 J.H. Mentink et al., PRL 057202, 2012

reversal of magnetization driven by exchange!!!

Ultrafast electrical pulse reverses magnetization!

SCIENCE ADVANCES | RESEARCH ARTICLE

PHYSICS

Ultrafast magnetization reversal by picosecond electrical pulses

Yang Yang,^{1*†} Richard B. Wilson,^{2*†} Jon Gorchon,^{3,4*} Charles-Henri Lambert,³ Sayeef Salahuddin,^{3,4} Jeffrey Bokor^{3,4†}







No hot, spin polarized or spin-orbit coupled electrons!

AOS of Ferromagnets
AOS of ferromagnetic CoPt (FePt)?



What's the mechanism?

HD-AOS in Co/Pt multilayer



HD-AOS in Co/Pt multilayer



HD-AOS in CoPt: fs single-shot imaging

Mechanism of all-optical control of ferromagnetic multilayers with circularly polarized light

R. Medapalli,^{1,*} D. Afanasiev,² D. K. Kim,¹ Y. Quessab,^{1, 3} S. Manna,¹ S. A. Montoya,¹ A. Kirilyuk,² Th. Rasing,² A. V. Kimel,² and E. E. Fullerton¹







No single shot switching
 Stochastic + deterministic
 ~100 pulses

MECHANISM?



Stochastic nucleation & growth!!!

Deterministic displacement of domain walls



2 nm/pulse *a* **0.4mJ/cm² takes many pulses! R. Medapalli** *et. al.*, arXiv: 1607.02505, PRB 96, 224421 (2017).

HD-AOS in CoPt: mechanism

Magnetic recording in Co/Pt requires multiple pulses.

The first pulses form (stochastically) domains with reversed magnetization.

The following pulses cause helicity dependent domain wall motion.

AOS of Dielectrics

Photo-magnetism of Co-substituted iron garnet



Light-induced slow (~ μ m/sec) motion of domain wall

A.Chizhik et al. PRB, 57 (1998).

A.Stupakiewicz et al. *PRB*, 64 (2001).





AOS in iron garnet

 $Y_2CaFe_{3.9}Co_{0.1}GeO_{12}$ on GGG (001) thickness d=7.5 µm)





All-Optical Switching (a) the nanoscale!

But....



All-Optical Switching @ the nanoscale?



PEEM experiment SLS; L. Le Guyader et al, APL 2012, Nature Comm. 2015

All-Optical Switching @ the nanoscale?



PEEM experiment SLS; L. Le Guyader et al, APL 2012, Nature Comm. 2015



plasmonic antenna!

Nanoscale switching with plasmonic antennas (with Bert Hecht, Wuerzburg)



Tian-Min Liu et al, Nano Letters, 2015

Outlook: speed and energy consumption in data storage







(10 ns/bit) 0,05 \$/GB (>pJ/bit)

(2 ns/bit) 0,65 \$/GB (>nJ/bit)

(~ ps/bit) ?? \$/GB (~fJ/bit) (20x20nm)

Outlook: Spintronic-Photonic Integrated Circuit



With: Aarhus University, IMEC, CEA SpinTEC, QuantumWise



Donut Switching with L-G beams



Towards stable complex nanostructures:





Skyrmion

Neel skyrmion

Bloch skyrmion

Opto-magnetic generation of Skyrmions

Single pulse illumination (λ = 800 nm) through microscope objective (NA = 0.4)



+

Read-out with Near-field microscopy

(λ = 532 nm, Resolution 80 nm)



Finazzi, et al. Phys. Rev. Lett. 110, 177205 (2013)

Sample: Tb₂₂Fe₆₉Co₉



Opto-magnetic generation of Skyrmions

Low fluence

FeTb

High fluence



Skyrmion generation model

Bloch skyrmion

M. Finazzi et al, Phys. Rev. Lett. 110, 177205, (2013)

End of smaller and faster?

I. End of "Moore": too much heat

II. Higher density = too much energy



III. von Neumann bottleneck: transfer information back and forth

Create a new paradigm, beyond von Neumann

Supercomputer versus Brain:





10 MW ocessing and stor

Processing and storage Separated and serial and 2D

10 W

Processing and storage Integrated and parallel and 3D!

Supercomputer versus Brain:





Processing and storage Separated and serial and 2D



10 W

Processing and storage Integrated and parallel and 3D!



Paradigm shift: *to develop materials that "learn"*



Neuromorphic Computing with Magneto-Optics?

see: A. Chakravarty et al, Supervised learning of an optomagnetic neural network with ultrashort laser pulses, Appl. Phys. Lett. 114, 192407 (2019)



To conclude:

1. Femtosecond optical excitation:

pump/probe magnetism on timescale of exchange interaction

- 2. Laser induced effects:
 - a. Thermal effects
 - **b.** Nonthermal opto-magnetic effects
- **3.** AOS of Ferrimagnets, Ferromagnets antiferromagnets, metals, dielectrics
- 4. AOS at the nanoscale, O-MRAM, Neuromorphic applications









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with many thanks to:

Radboud Nijmegen

A.V. Kimel
A.Kirilyuk
F. Hansteen
D.Stanciu
A.v.Etteger
A. Toonen
K.Vahaplar
M. Savoini (Zurich)
S. Khorsand (ASML)
D. Bossini (Dortmund)
R. Mikhaylovskiy (Lancaster)

University of York R. Chantrell T.A. Ostler (Sheffield) **J.Barker R**.Evans **Ioffe Institute R.V.** Pisarev A. Kalashnikova **Nihon University** Prof. A. Itoh A. Tsukamoto **Kiev** B. Ivanov

UC San Diego

R. Medapalli Yassine Quessab Sheena K.K. Patel Eric E. Fullerton

Bessy

I. Radu C. Stamm (Zurich) T.Kachel N.Pontius L.Le Guyader (Hamburg) **Stanford** Herman Durr (Uppsala) A.Reid C.Graves



STW, NWO, EU, NanoNed, UltraMagnetron, FANTOMAS, IFOX