Light-induced (de)magnetization processes and all-optical switching



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Experimental approaches

How one gets a time resolution

How one measures ultrafast magnetization dynamics

Laser-induced demagnetization

Experimental observation

Theories

Laser-induced changes of magnetic anisotropy

Through demagnetization

Through lattice heating

Through strain

Ultrafast opto-magnetic effects

Opto-magnetic effects and Impulsive stimulated Raman scattering

Control of magnetization via opto-magnetic effects

Laser-driven spin transport

Light-induced (de)magnetization processes and all-optical switching



Experimental approaches

How does one get a time resolution?

How does one measure an ultrafast magnetization dynamics?

Experimental approaches

How to realize pump-probe delays? What to measure?



Pump-probe delays



Detecting ultrafast magnetization dynamics

Ferromagnets

Magneto-optical Kerr and Faraday effects

Quadratic MO effects



Tracing all components of magnetization [See the lecture by Paolo VAVASSORI]

Are we sure we are detecting magnetization dynamics?

Time-resolved polar MOKE in Ni films:



MOKE rotation

 $\theta(t) \sim (\chi'_{xyz} + \delta \chi'_{xyz})(M_z + \delta M_z)$ $\delta \theta(t) \sim \chi'_{xyz} \delta M_z + \delta \chi'_{xyz} M_z$

MOKE ellipticily

 $\delta \chi''_{xyz} M_z$ $\delta\epsilon(t) \gamma \chi''_{xvz} \delta M_z$

Magnetization dynamics ©

Electronic, lattice dynamics ⊗

Measuring two MO effects is a good idea!

Detecting ultrafast magnetization dynamics

Antiferromagnets



Light-induced (de)magnetization processes and all-optical switching



Laser-induced demagnetization

Experimental observation Theories

Laser-induced electronic and lattice dynamics in metals



$$C_{\rm el}(T_{\rm el})\frac{dT_{\rm el}}{dt} = -G_{\rm el-ph}(T_{\rm el} - T_{\rm l}) + P(t,r),$$

$$C_{\rm l}(T_{\rm l})\frac{dT_{\rm l}}{dt} = -G_{\rm el-ph}(T_{\rm l} - T_{\rm el}) - C_{\rm l}\frac{T_{\rm l} - T_{\rm 0}}{\tau_{\rm th}},$$

[Kaganov et al., Sov. Phys.-JETP 31, 232 (1956)]

Electrons thermalization times: 350 fs (Ag), 500 fs (Au)

Electron-phonon relaxation times: 950 fs (Ag)

[Del Fatti et al., PRB 61, 16 956 (2000)]

Laser-induced spin dynamics in metals (transition-metal case)



$$\begin{split} &C_{e}d(T_{e})/dt = -\;G_{el}(T_{e}-T_{l}) - G_{es}(T_{e}-T_{s}) + P(t)\,,\\ &C_{s}d(T_{s})/dt = -\;G_{es}(T_{s}-T_{e}) - G_{sl}(T_{s}-T_{l})\,,\\ &C_{l}d(T_{l})/dt = -\;G_{el}(T_{l}-T_{e}) - G_{sl}(T_{l}-T_{s})\,, \end{split}$$



[Beaurepaire et al., PRL 76, 4250 (1996)] 9

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Laser-induced spin dynamics in metals (rare-earth case)



[Koopmans, et al., Nature Mater. 9, 259 (2009) Vaterlaus et al., PRL 67, 3314 (1991)] 10

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Electrons and lattice dynamics: 2T-model

$$C_{\rm el}(T_{\rm el})\frac{dT_{\rm el}}{dt} = -G_{\rm el-ph}(T_{\rm el} - T_{\rm l}) + P(t,r),$$
$$C_{\rm l}(T_{\rm l})\frac{dT_{\rm l}}{dt} = -G_{\rm el-ph}(T_{\rm l} - T_{\rm el}) - C_{\rm l}\frac{T_{\rm l} - T_{\rm 0}}{\tau_{\rm th}},$$

Spin dynamics: via el-ph spin flip scattering with probability α_{sf}



 α_{sf} is related to spin-mixing of electronic states at the Fermi energy due to SO interactions

Magnetization dynamics according to the M3T model

► Electrons provide energy for the demagnetization

≻Lattice provides the sink for the angular momentum

$$\frac{dm}{dt} = Rm \frac{T_l}{T_c} \left(1 - \coth\left(\frac{mT_c}{T_{el}}\right) \right)$$

[Koopmans et al., Nature Mater. 9, 259 (2009]





$$\frac{dm}{dt} = Rm \frac{T_l}{T_c} \left(1 - \coth\left(\frac{mT_c}{T_{el}}\right) \right) \qquad R \propto \frac{a_{sf}T_c^2}{\mu_{at}}$$



[Koopmans et al., Nature Mater. 9, 259 (2009)]

Ni: type I dynamics



[Koopmans et al., Nature Mater. 9, 259 (2009)]

Reason for demagnetization: spin-dependent transport from the laser-heated area in the superdiffusive regime:

Electrons are excited from the d-band to sp-bands above the Fermi level: high mobility electrons (~1nm/fs)

➢Spin-conserving scattering events

>Each scattering event is isotropic



Superdiffusive regime:
$$\sigma^2 \propto t^{\gamma(t)}$$
 -variance of particle displacement distribution
γ>1 and decreases with time (γ=1 – diffusion; γ=2 - ballistics)

>Different life times for majority and minority electrons

Majority electrons transfer magnetization away from excited area [Battiato et al., PRL **105**, 027203 (2010)]



Attempts to verify if this mechanism is present

[Rudolf et al., Nature Comm. 3, 1037 (2012)] See also Melnikov et al., PRL **107**, 076601 (2011)

Agrees with the theory predictions but.....



[Schellekens et al., APL 102, 252408 (2013)]

Attempts to verify if this mechanism is present



[Maison et al., Sci. Rep. 4, 4658 (2014)]

Ultrafast (?) demagnetization in dielectrics







Demagnetization in a dielectric is mediated by the phonon-magnon interaction

See lecture by T. Kampfrath

Light-induced (de)magnetization processes and all-optical switching



Laser-induced changes of magnetic anisotropy

- Through demagnetization
- Through lattice heating
- Through strain

Mechanisms of laser-induced changes of magnetic anisotropy



Probing laser-induced changes of magnetic anisotropy



Example: uniaxial easy axis-type anisotropy

$$\mathbf{H}_{a} = -\frac{\partial F_{a}}{\partial \mathbf{M}} = -K_{u}m_{z}\mathbf{Z}$$

$$\frac{d\mathbf{M}}{dt} = -\gamma [\mathbf{M} \times \mathbf{H}_{\text{eff}}]$$
$$\mathbf{T}_{0} = -\gamma [\mathbf{M}_{0} \times \delta \mathbf{H}_{\text{eff}}]$$

Anisotropy of the laser-pulse excited sample:





Magnetization precession serves as a probe of laser-induced anisotropy changes

Ultrafast demagnetization and shape anisotropy (δM-process)



[van Kampen et al., PRL 88, 227201 (2002)]

Laser-induced change of anisotropy constants (&K-process)

Epitaxial metallic film Fe/MgO

Cubic anisotropy:

$$E_{a} = K_{1}(T) \left(\alpha_{1}^{2} \alpha_{2}^{2} + \alpha_{2}^{2} \alpha_{3}^{2} + \alpha_{1}^{2} \alpha_{3}^{2} \right)$$

Dynamics of the anisotropy

 $\mathbf{T}_{0} = -\gamma [\mathbf{M}_{0} \times \delta \mathbf{H}_{\mathrm{eff}}]$



Deviation of **H**_{eff}

[Carpene et al., PRB **81**, 060415(2010); Carpene et al., JAP **108**, 063919 (2010)]

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Laser-induced change of anisotropy constants (&K-process)

Epitaxial dielectric film Y₃Fe₅O₁₂/GGG



Contribution from laser-induced δK dominates at low fields

Laser-induced change of anisotropy δK^{\sim} 1%

[Shelukhin et al., PRB 97, 014422 (2018)]

Laser-induced strain and inverse magnetostriction (δε-process)



Picosecond magnetoacoustics



Strain pulse injected to GaAs from the Al film

 $\tau \sim 10 \text{ ps}$ $\varepsilon_{zz} \sim 10^{-3} - 10^{-4}$

Magneto-elastic energy:

$$E_{me} = b \varepsilon_{ij} m_i m_j$$

Torque action on magnetization due to strain:

 $\mathbf{T}_0 = -\gamma \left[\mathbf{M}_0 \times b \varepsilon_{ij} \mathbf{m}_i \right]$





Propagating strain pulse modifies magnetocrystaline anisotropy and launches the precession

[Scherbakov et al., PRL 105,117204 (2010), Kim et al., PRL 109, 166601 (2012)...]

Laser-induced strain and inverse magnetostriction (δε-process)



Low-symmetry magnetostrictive metallic film





Why (311) substrate?

A film on a <001> substrate:



Only compressive/tensile strain

A film on a low-symmetry substrate:



Compressive/tensile & shear strain

Direct optical excitation of precession: magnetic field dependence





Direct optical excitation of precession: magnetic field dependence



Two competing mechanisms of precession excitation!

Laser-induced change of anisotropy in a metallic film: model



Laser-induced change of anisotropy in a metallic film





Laser-induced change of anisotropy in a metallic film



Mechanisms of laser-induced changes of magnetic anisotropy



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Ultrafast opto-magnetic effects

Opto-magnetic effects and Impulsive stimulated Raman scattering Control of magnetization via opto-magnetic effects Interaction of light with a magnetic medium

$$\Phi_{\text{int}} = \varepsilon_{ij} E_i E_j^* + \alpha_{ijk} E_i E_j^* M_k + \beta_{ijkl} E_i E_j^* M_k M_l + \dots$$

Faraday effect



Inverse Faraday effect



[Pitaevskii, Sov. Phys. JETP **12**, 1008 (1961) van der Ziel PRL. **15**, 190 (1965)]

$$\Delta n_{\sigma^+ - \sigma^-} \sim \alpha_{xyz} M_z$$

$$\mathbf{M} \sim \boldsymbol{\alpha}_{xyz} \mathbf{E} \times \mathbf{E}^*$$

What about short laser pulses and magnetic medium?

Ultrafast inverse Faraday effect

DyFeO₃



[A. V. Kimel et al., Nature (2005)]

Laser-induced femtosecond pulse of an effective magnetic field



Ultrafast inverse Faraday effect

Controlling the phase of the coherent spin precession by circularly-polarized light

Ultrafast opto-magnetic effects



Ultrafast inverse Cotton-Mouton effect



Controlling the phase of the coherent spin precession by linearly-polarized light

Spontaneous Raman scattering on magnons



Stimulated Raman scattering with femtosecond pulses



[A. M. Kalashnikova et al., PRL (2007), PRB (2008); V. N. Gridnev, PRB (2008)]

A role of absorption in the magnon excitation?





 $T_N = 246 \text{ K} \qquad H_{ex}/H_A = 10^5$

Approach: tuning the pump wavelength between transparency windows and absorption bands



Excitation of magnons without absorption





How does absorption affects the process of excitation?





Coherent magnons excited via ISRS



decrease of |L| due to laser-induced heating

Mechanism of excitation of coherent and incoherent spin dynamics



Absorption results in excitation of incoherent magnons (demagnetization) mediated by lattice heating

40

 τ_d

120

160

80

Absorption a (cm⁻¹)

(c)

Excitation of spin system: regime of zero-absorption



Coherent magnons excited via ISRS and decrease of |L| due to ?



Extra channel for demagnetization in a transparent dielectric



Criterion: $\tau_r = \tau_d$

A brief detour: Raman scattering and ISRS (on phonons) and the link between them

Coherent optical phonons via ISRS:

Merlin, Solid State Commun. **102**, 207 (1997) Dhar, et al., Chem. Rev. **94**, 157 (1994) Yan and Nelson, J. Chem. Phys. **87**, 6257 (1987)

Crystal structure of CuB₂O₄



123 optical phonon above above 4 THz

P.G.-42m; S.G. I-42d; Z=12; Cu²⁺ ions in 4b and 8d positions.

Excitation of coherent phonons in CuB₂O₄ via impulsive stimulated Raman scattering



[Imasaka et al., PRB **98**, 054303 (2018)]

Spontaneous and impulsive stimulated Raman scattering

Spontaneous Raman scattering spectra

FFT spectra of the pump-probe data



Why are the amplitudes so different?

Excitation and detection of coherent phonons in the ISRS experiment





Excitation and detection of coherent phonons in the ISRS experiment





Q(t) is the phonon normal coordinate

Dependence on the polarization



Link between spontaneous Raman intensity spectra and ISRS-driven phonon amplitudes



Experiment and calculations



Back to ISRS and magnons

Encoding light polarization state in an antiferromagnet



Stokes parameters

 $I = (E_a^2 + E_b^2)$ $I \cos 2\psi \cos 2\chi$ $I \sin 2\psi \cos 2\chi$ $I \sin 2\chi$

3 Stokes parameters requires three magnon modes **Poincare sphere**



YMnO₃: 3-sublattice antiferromagnet



Magnetic structure





"Writing" light polarization into a magnon mode



"Reading" magnon mode via light polarization



Experimental verification



[Satoh et al., Nature Photonics, 9, 25 (2014)]

We've already discussed: laser-induced changes of anisotropy



Spin reorientation phase transition in *RE*FeO₃



Laser-induced spin-reorientation phase transition



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

How to lift the degeneracy?

Controlling the phase transition by a single laser pulse alone?

Sample: rare-earth orthoferrite (Sm_{0.5}Pr_{0.5})FeO₃

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T=70 K

T=180 K



Spin reorientation phase transition at

98-130 K



T₂

Т

 \mathbf{T}_1

~20°/90 μM

➤Large Faraday rotation

Single-shot femtosecond time-resolved magneto-optical imaging



Magneto-optical images with subpicosecond resolution

Single-shot femtosecond time-resolved magneto-optical imaging



Laser-induced magnetization dynamics in (Sm,Pr)FeO₃



[de Jong et al., PRL 108 157601 (2012)]

Т=90 К

Ultrafast laser-induced SR transition controlled by a laser pulse polarization alone!

Mechanism of the laser-induced SR transition



How and where the information about the laser pulse polarization is stored?

Coherent control of the laser-induced SR transition



Ultrafast inverse Faraday effect

Impulsive excitation of the <u>low amplitude</u> magnetization precession (<10°)



Phase of the precession is helicity-dependent

Degeneracy between two states is lifted dynamically
Control of the SR transition: temperature and fluence



Light-induced (de)magnetization processes and all-optical switching



Laser-driven spin transport

Optically-driven magneto-static waves



Pump and probe spot sizes ~1 μm

Pump-probe scans

BVMSSW driven by ultrafast inverse Faraday effect



MS waves driven by ultrafast opto-magnetic effects

Only in substituted YIG films so far

[Satoh et al. Nature Photon. **6**, 662 (2012) Jackl et al., PRX **2**, 021009 (2017)]

Optically-driven magneto-static waves



MSSW (and BVMSSW) driven by ultrafast change of shape anisotropy (δM-process)





[Au et al., PRL **110**, 097201 (2013) Kamikaki et al., **96**, 014438 (2017)]

Optically-driven spin polarized currents and precession



Spin pumping and dissipative coupling of precessions





 α_i - intrinsic (Gilbert) damping

 β_i - damping due to spin pumping

Two magnetic layers coupled via spin current



Laser-driven spin pumping and dissipative coupling



Top view:



(Fe,Ga)/GaAs

Cubic anisotropy

+ Uniaxial anisotropy



(Fe,Ga)/Cu/GaAs

Cubic anisotropy

Excitation via δK-process

No resonance $f_1 \neq f_2$ -> No coupling



Resonance $f_1 = f_2$ -> dissipative coupling & both modes are excited and observed



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Light-induced (de)magnetization processes and all-optical switching



Laser-induced demagnetization

Is ultrafast in metals but slow in dielectrics

Different theories of ultrafast demagnetization in metals

Laser-induced changes of magnetic anisotropy



No heating is involved

Control of spins by polarized laser pulses

Laser-driven spin transport