Calculating ultrafast laser-induced magnetization dynamics

K. Carva, D. Legut, P. M. Oppeneer

IT4Innovations
national01$#&0
supercomputing
center@#01%101

Charles University in Prague

IT4Innovations Center, VSB TU Ostrava

Uppsala University

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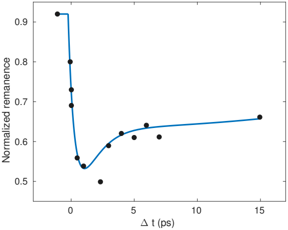
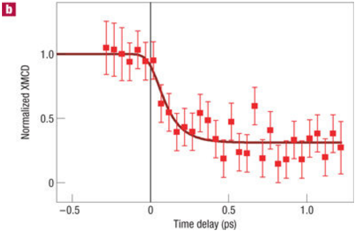
Femtosecond magnetization dynamics

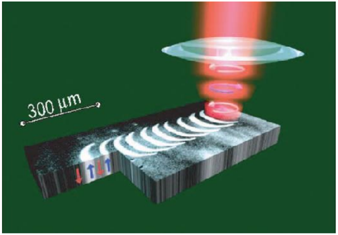
- Magnetization change without the external magnetic field, caused by an intense laser pulse
- *Beaurepaire, 1996*: A significant decrease of magnetization in Ni after an intense laser pulse. (Later observation by Stamm et al.)

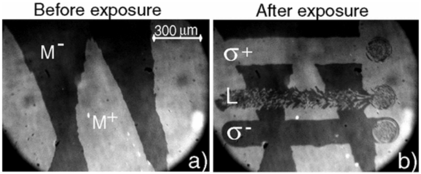




- Previously unexplored timescale for magnetism: below 1 ps

Optically controlled magnetization switching

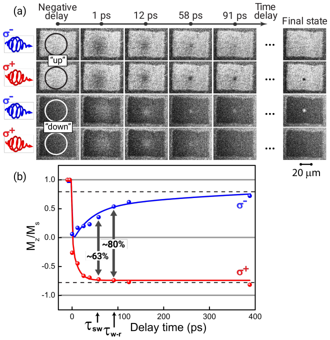




- Stanciu et al., PRL 99 (2007)
- Vahaplar et al., PRL 103 (2009)
- Applications with high impact in data storage - permanently stored information at the speed of RAM
- Origin: Inverse Faraday effect?

Outline

Evolution after the pulse disappears



- $Gd_{24}Fe_{66}Co_{10}$: chemically disordered, amorphous. Close to $GdFe_2$.
- After 1ps: no information about the direction of evolution
- No precession as in field-driven switching

A narrow window with controlled reversal

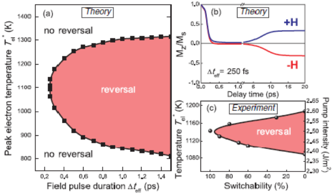
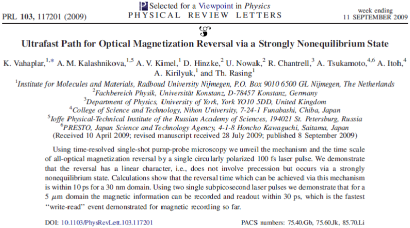


FIG. 2 (color). (a) Phase diagram showing the magnetic state of the $(30\ \mathrm{nm})^3$ volume achieved within 10 ps after the action of the optomagnetic pulse with parameters $H_{\mathrm{eff}} = 20$ T, Δt_{eff}, and T_{el}^{*}. (b) The averaged z component of the magnetization versus delay time as calculated for 250 fs magnetic field pulses $H_{\mathrm{eff}} = \pm 20$ T and $T_{\mathrm{el}}^{*} = 1130$ K. (c) Switchability versus the pump


Selected for a Viewpoint in *Physics*

PRL **103,** 117201 (2009) PHYSICAL REVIEW LETTERS week ending 11 SEPTEMBER 2009

Ultrafast Path for Optical Magnetization Reversal via a Strongly Nonequilibrium State

K. Vahaplar,[1,*] A. M. Kalashnikova,[1,5] A. V. Kimel,[1] D. Hinzke,[2] U. Nowak,[2] R. Chantrell,[3] A. Tsukamoto,[4,6] A. Itoh,[4]
A. Kirilyuk,[1] and Th. Rasing[1]
[1]*Institute for Molecules and Materials, Radboud University Nijmegen, P.O. Box 9010 6500 GL Nijmegen, The Netherlands*
[2]*Fachbereich Physik, Universität Konstanz, D-78457 Konstanz, Germany*
[3]*Department of Physics, University of York, York YO10 5DD, United Kingdom*
[4]*College of Science and Technology, Nihon University, 7-24-1 Funabashi, Chiba, Japan*
[5]*Ioffe Physical-Technical Institute of the Russian Academy of Sciences, 194021 St. Petersburg, Russia*
[6]*PRESTO, Japan Science and Technology Agency, 4-1-8 Honcho Kawaguchi, Saitama, Japan*
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Using time-resolved single-shot pump-probe microscopy we unveil the mechanism and the time scale of all-optical magnetization reversal by a single circularly polarized 100 fs laser pulse. We demonstrate that the reversal has a linear character, i.e., does not involve precession but occurs via a strongly nonequilibrium state. Calculations show that the reversal time which can be achieved via this mechanism is within 10 ps for a 30 nm domain. Using two single subpicosecond laser pulses we demonstrate that for a 5 μm domain the magnetic information can be recorded and readout within 30 ps, which is the fastest "write-read" event demonstrated for magnetic recording so far.

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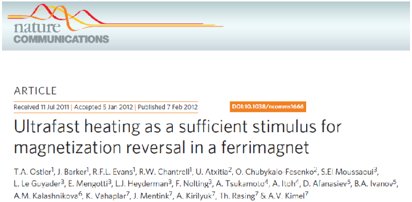
Reversal induced by each pulse



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ARTICLE

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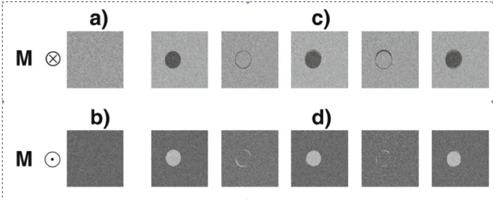
Ultrafast heating as a sufficient stimulus for magnetization reversal in a ferrimagnet

T.A. Ostler[1], J. Barker[1], R.F.L. Evans[1], R.W. Chantrell[1], U. Atxitia[2], O. Chubykalo-Fesenko[2], S.El Moussaoui[3], L. Le Guyader[3], E. Mengotti[3], L.J. Heyderman[3], F. Nolting[3], A. Tsukamoto[4], A. Itoh[4], D. Afanasiev[5], B.A. Ivanov[5], A.M. Kalashnikova[6], K. Vahaplar[7], J. Mentink[7], A. Kirilyuk[7], Th. Rasing[7] & A.V. Kimel[7]

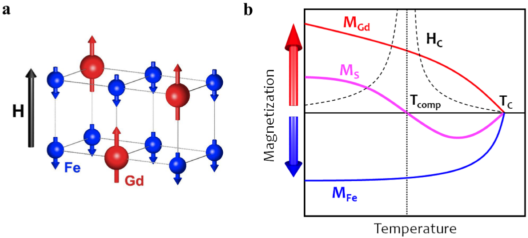


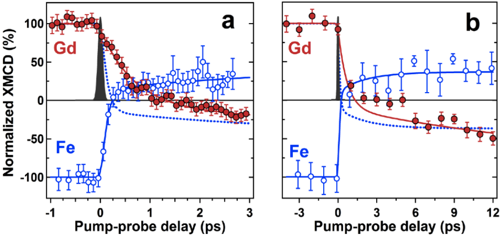
Discrepancy in experiments?

- Inverse Faraday effect ruled out, the original polarization dependence was only due to different absorption coefficients for different polarizations (happening in a very narrow fluence window)
- No external field, circular polarization of the laser pulse or any other direct source of angular momentum, but: magnetization acquires specific direction different from the original one

Time-resolved XMCD study

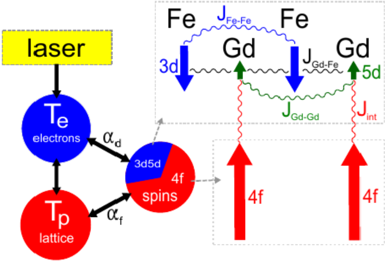
Radu et al., Nature 472 (2011) :





Model (co-operation with Konstanz)

- $GdFe_2$: cubic Laves phase (C_{15}), T_C=560K, T_M=300K
- Gd: 90% of mag. moment in 4f states, but localized more than 4eV below E_F
- J_{TM-TM}= 32.5 meV, J_{R-R}= 7.8 meV, J_{TM-R}= -3.2 meV, J_{int}= 130 meV



Simulations based on LLG equation

$$\mathcal{H} = -\sum_{\langle ij \rangle} \frac{J_{ij}}{2} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_{i \in \mathrm{Gd}} J_{int} \mathbf{S}_i \cdot \mathbf{S}'_i - d_z \sum_i (S_i^z)^2. \qquad (1)$$

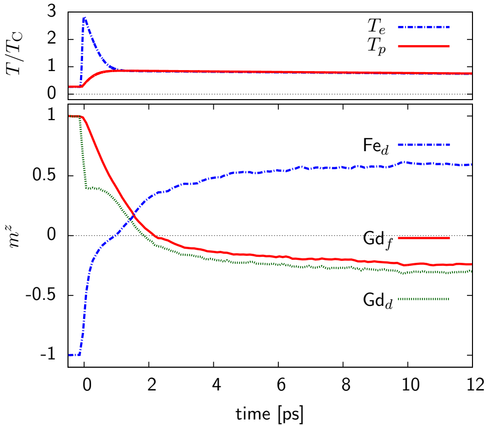
Landau-Lifschitz-Gilbert equation:

$$\frac{\partial \mathbf{S}_i}{\partial t} = \; - \frac{\gamma_i}{(1+\alpha_i^2)\mu_S^i} \mathbf{S}_i \times \mathbf{H}_i(t) \quad - \frac{\alpha_i \gamma_i}{(1+\alpha_i^2)\mu_S^i} \mathbf{S}_i \times \big(\mathbf{S}_i \times \mathbf{H}_i(t) \big).$$

γ_i ... the gyromagnetic ratio. For the $4f$ electrons of Gd: the same equation with the primed quantities.
Thermal fluctuations: additional white-noise term - the effective field $\mathbf{H}_i(t) = -\frac{\partial \mathcal{H}}{\partial \mathbf{S}_i} + \boldsymbol{\zeta}_i(t)$, the thermal noise term $\boldsymbol{\zeta}_i$

$$\langle \boldsymbol{\zeta}_i(t) \rangle = 0, \quad \langle \zeta_{i\eta}(0) \zeta_{j\theta}(t) \rangle = \delta_{ij} \delta_{\eta\theta} \delta(t) 2\alpha_i k_B T_i \mu_S^i / \gamma_i. \qquad (2)$$

Simulated magnetization evolution



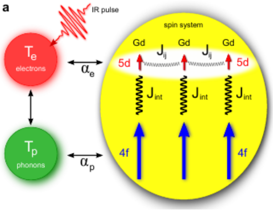
S. Wienholdt, D. Hinzke, K. Carva, P. M. Oppeneer, and U. Nowak, Phys. Rev. B 88, 020406(R) (2013)

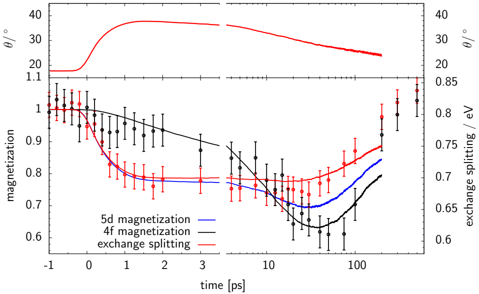
Gd: LLG with 5*d* momentum calculation

- Ab initio exchange constants employed in LLG simulation *(B. Frietsch, KC, PMO et al. , Nat Commun 6, 2015):*

$$\frac{\partial \mathbf{S}_i}{\partial t} = -\frac{\gamma_i}{(1+\alpha_i^2)\mu_S^i}\,\mathbf{S}_i \times \mathbf{H}_i(t) \quad -\frac{\alpha_i\gamma_i}{(1+\alpha_i^2)\mu_S^i}\,\mathbf{S}_i \times (\mathbf{S}_i \times \mathbf{H}_i(t)).$$

- Mean M, ϑ from previous LLG time step→ 5*d* momentum recalculated

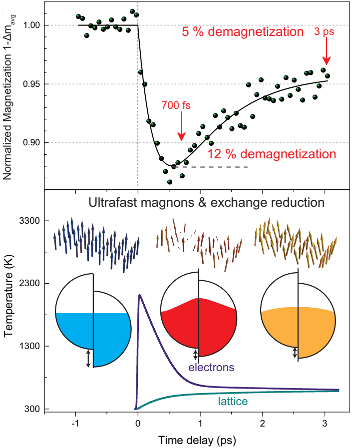




Magnetic excitations in a fs laser pumped system

Three possible effects:

- Disorder of magnetic moments (transversal spin excitation)
- Reduction of exchange splitting (longitudinal spin excitation)
- An increase of electronic temperature

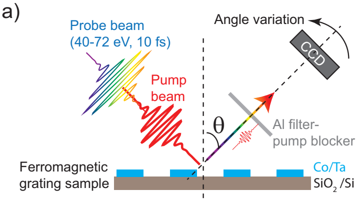


MO response

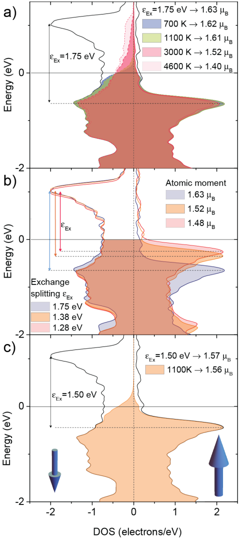
- High harmonics used to generate EUV light and probe the excited system MO response *(Fan, DL, KC, PMO, et al., PNAS 112 (2015) 14206)*
- MO asymmetry

$$A = 2\mathrm{Re}\frac{\epsilon_{xy}\sin 2\vartheta_i}{n^4\cos\vartheta_i^2 - n^2 + \sin\vartheta_i^2}$$

  measured for different energies and angles
- Comparison between the calculated and measured asymmetry spectra *(Turgut, DL, KC, PMO, et al., Phys. Rev. B 94 (2016) 220408)*



Calculated Co DOSes for different excitations



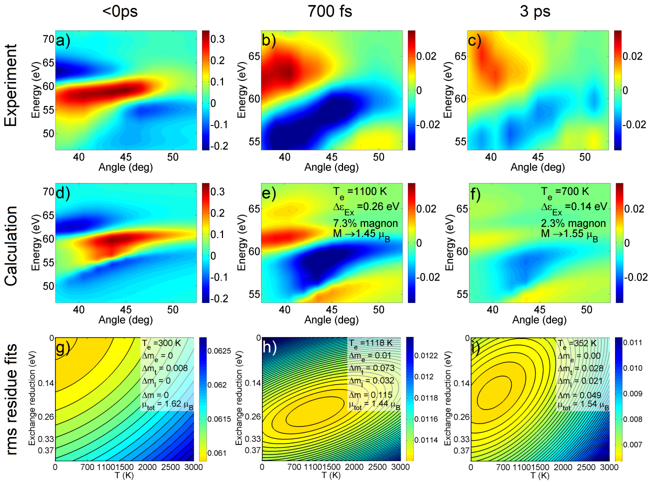
a) Elevated T_e: multiplication with a FD distribution

b) Reduced exchange: fixed spin moment calculation

c) Combination of the above

(Wien2K FP-LAPW code: 1.5mil. corehours, R_{kmax}=8.5 Ry^{-1}, l_{max} = 10, kgrid 44x44x22)

Extraction of the contributions



- Compare *Eich et al., 2017*: only magnons found, a conducting substrate

Conclusions

- Ultrafast heating found to be sufficient stimulus for magnetization reversal - the reversal process is also of thermal origin similar to demagnetization
- Breaking of intra-atomic $4f$-$5d$ coupling in Gd on a ps timescale *(Frietsch et al., Nat Commun, 2015)*
- TR-MO asymmetry measurement allows us to disentangle the different contributions to demagnetization *(Turgut, DL, KC, PMO, et al., PRB 94, 2016)*
- Both transverse and longitudinal excitations found to contribute, at 700fs $\Delta m_t/\Delta m_l > 2$, later the contribution from longitudinal component is growing

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