

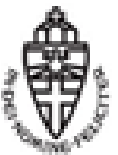
Femtosecond magnetism: the role of the exchange spin-spin interaction

Alexey V. Kimel

**Radboud University Nijmegen,
Institute for Molecules and Materials,
Nijmegen, The Netherlands**



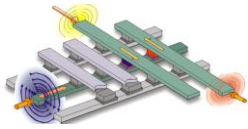
Radboud University Nijmegen



Ultrafast magnetism: *terra incognita* of modern science

Technology

MRAM



Computing time
per bit

Hard Drive



Recording time
per bit

Science

Equilibrium
Thermodynamics

Macrospin

Adiabatic
approximations



1 ns

100 ps

10 ps

1 ps

100 fs

10 fs

1 fs



Stimulus

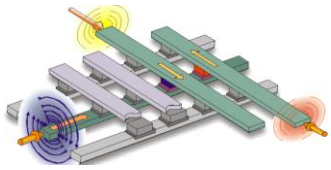
Challenge:

To control magnetic state
of a medium with the best
possible temporal resolutions

Magnetism in Technology and Science

Technology

MRAM

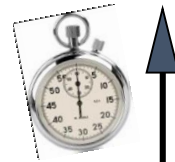


Hard Drive



Computing time
per bit

Recording time
per bit



1 ns

100 ps

10 ps

1 ps

100 fs

10 fs

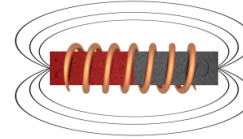
1 fs

Challenge:

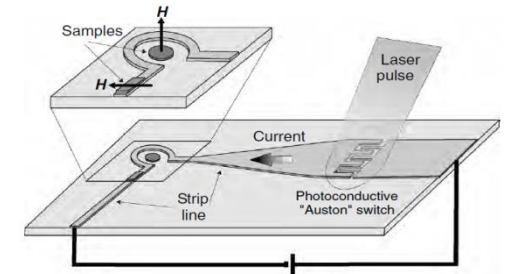
To control magnetic state
of a medium with the best possible
temporal and spatial resolutions

Stimuli

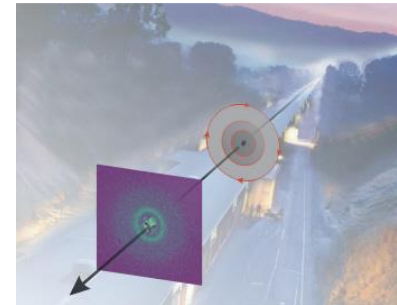
Electromagnets



Photoswitch



Stanford Linear Accelerator

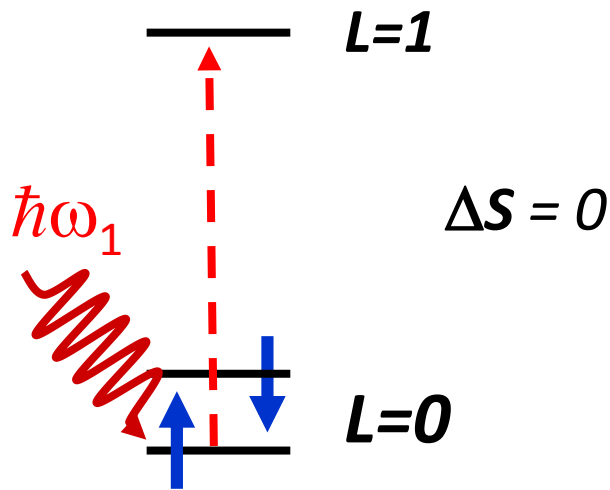


Laser Pulse



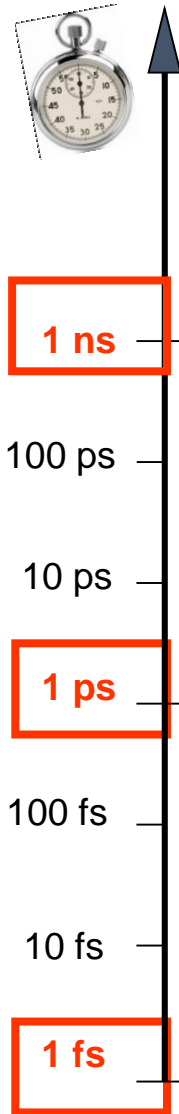
Magnetism in Technology and Science

Can light act as a magnetic field?

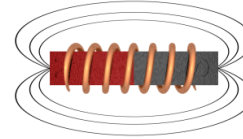


Challenge:

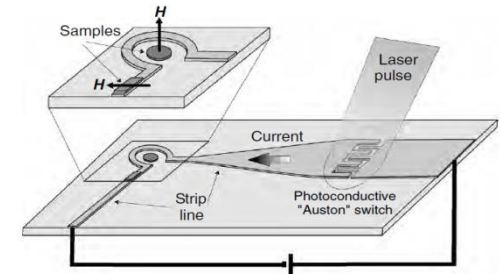
To control magnetic state of a medium with the best possible temporal and spatial resolutions



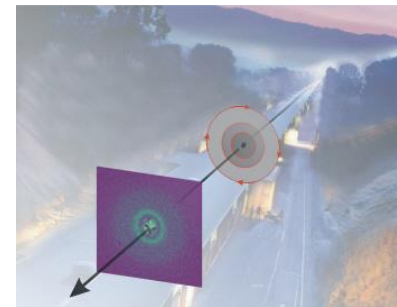
Electromagnets



Photoswitch



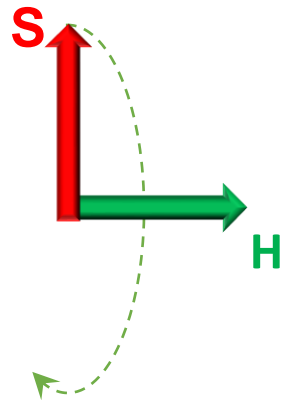
Stanford Linear Accelerator



Laser Pulse

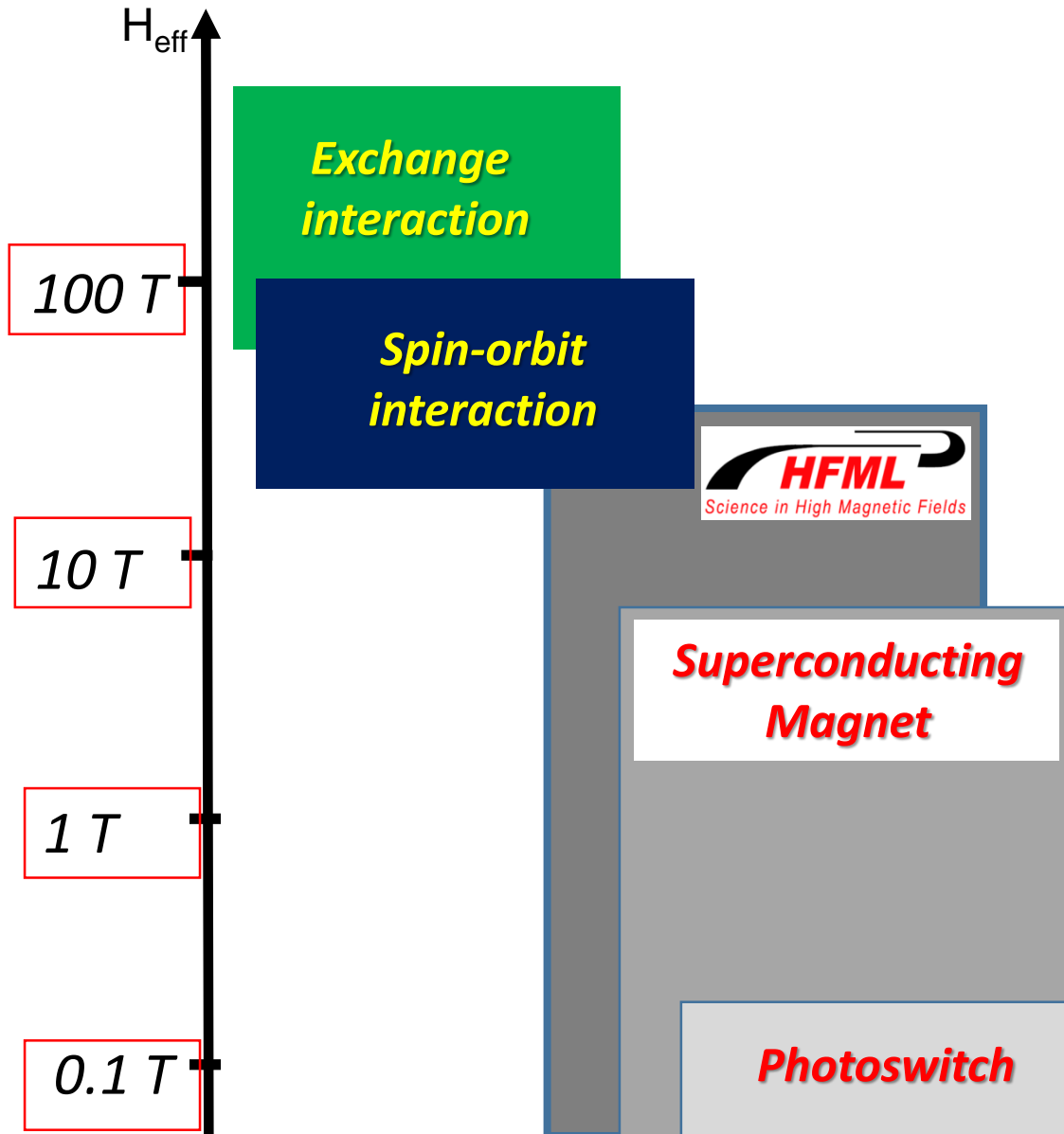
What can spins “feel”?

$$\mathcal{H}_i = \mathbf{H} \mathbf{S}_i + \alpha \mathbf{L}_i \mathbf{S}_i + J_{ij} \mathbf{S}_j \mathbf{S}_i$$



$$\frac{\partial \mathbf{S}_i}{\partial t} = -\gamma \mathbf{S}_i \times \mathbf{H}_{\text{eff}}$$

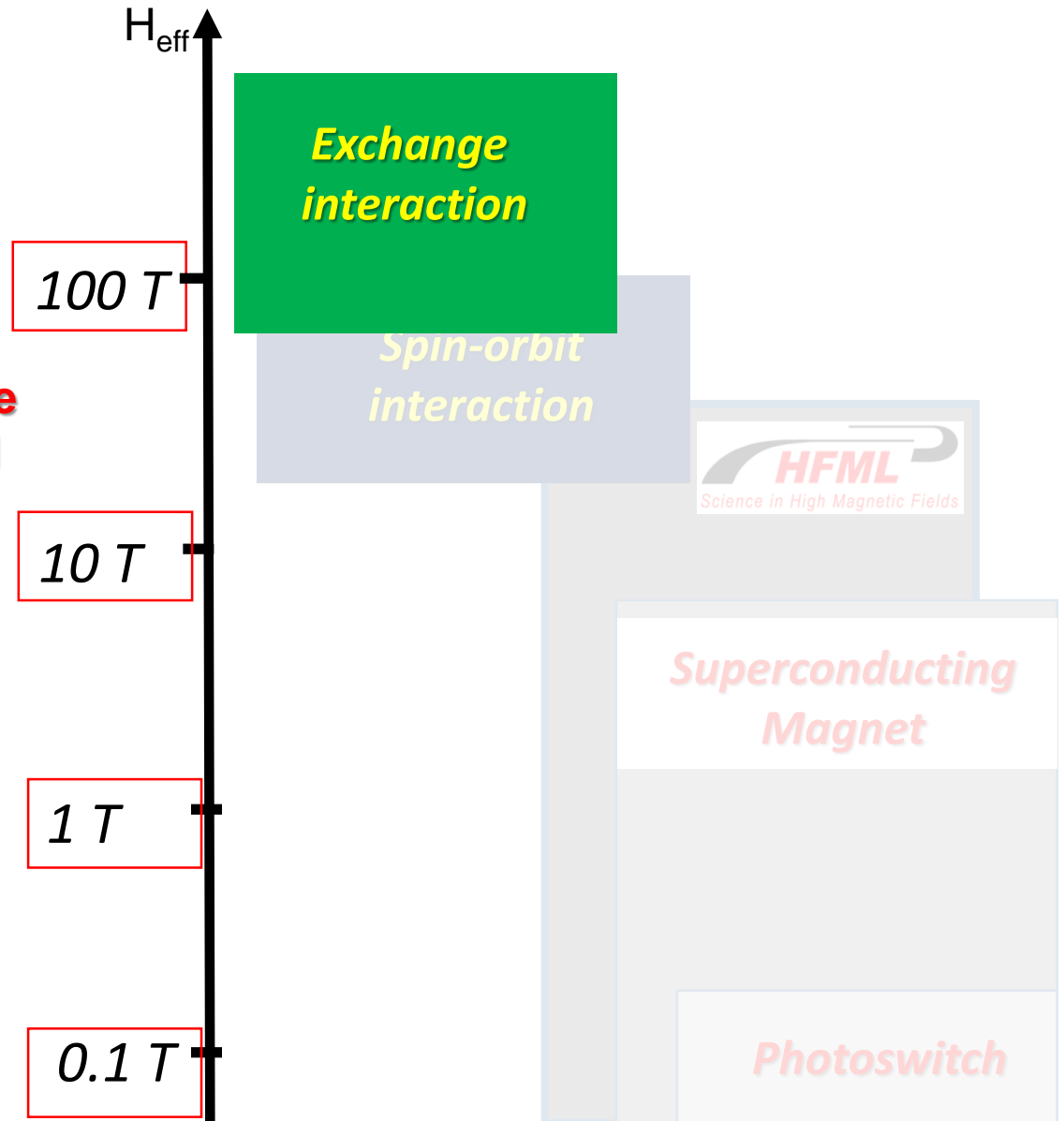
$$\mathbf{H}_{\text{eff}} = \frac{\partial \mathcal{H}_i}{\partial \mathbf{S}_i} = \mathbf{H} + \alpha \mathbf{L}_i + J_{ij} \mathbf{S}_j$$



Questions

Can we harness the exchange interaction for optical control of spins?

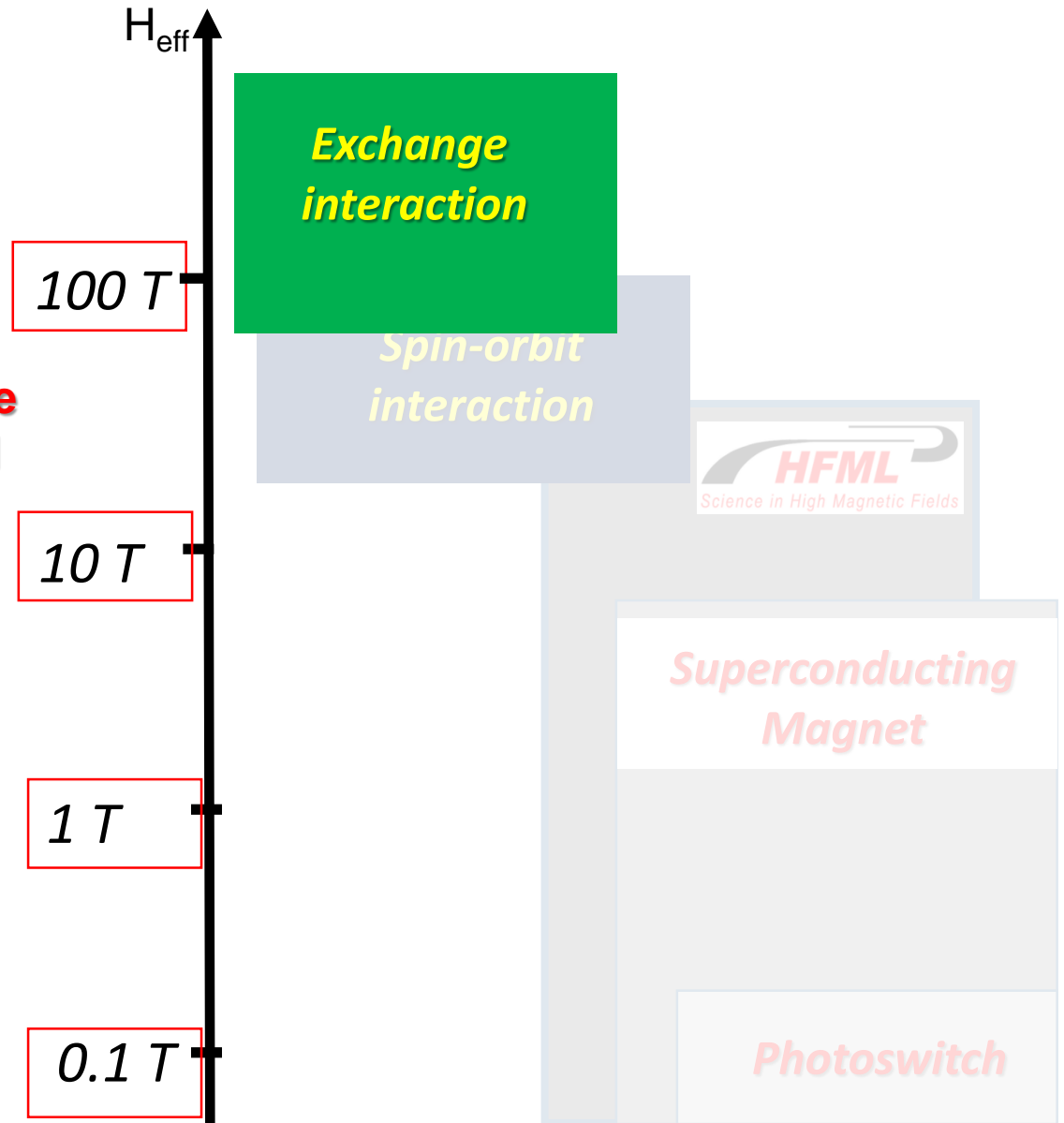
Can light control the exchange interaction?



Questions

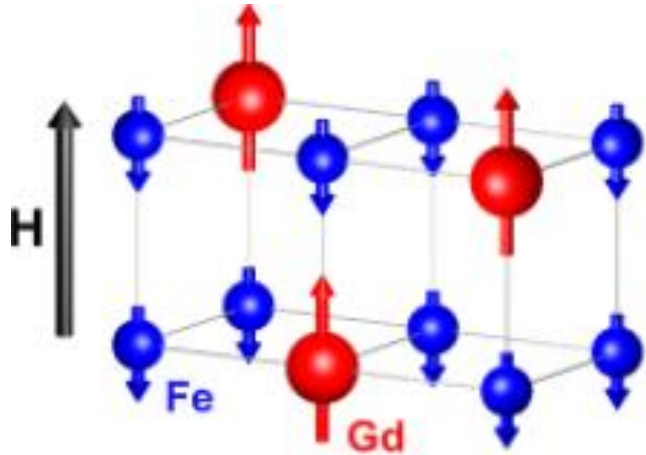
Can we harness the exchange interaction for optical control of spins?

Can light control the exchange interaction?



How do spins respond to a stimulus faster than the exchange interaction?

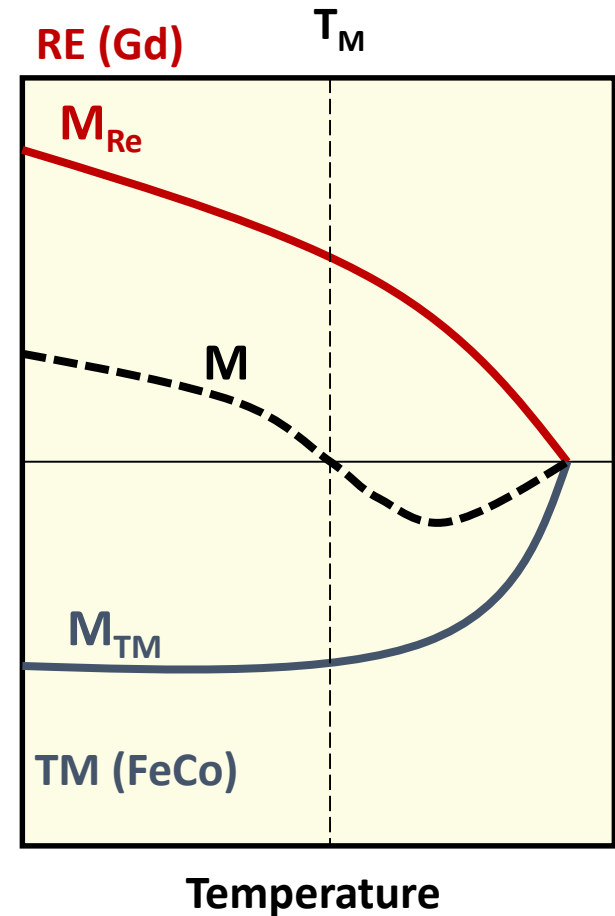
RE-TM ferrimagnetic alloy as a model system



$$J_{\text{Fe-Fe}} = 1.96 \times 10^{-20} \text{ J} \sim 35 \text{ fs}$$

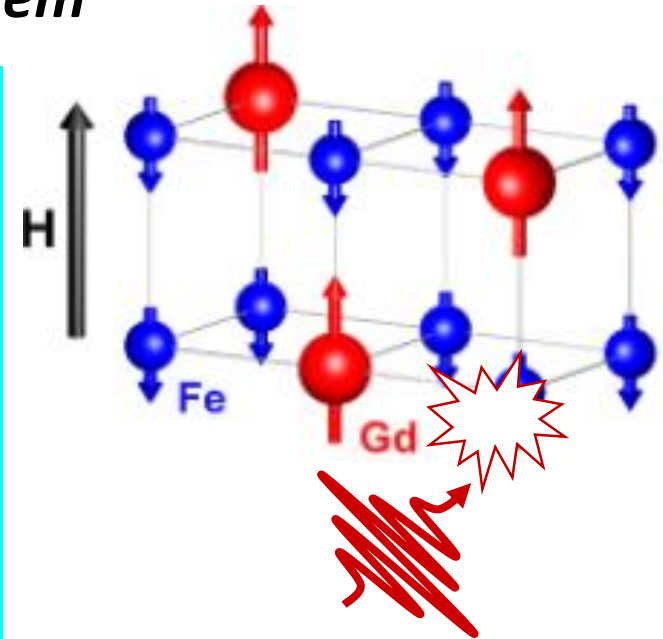
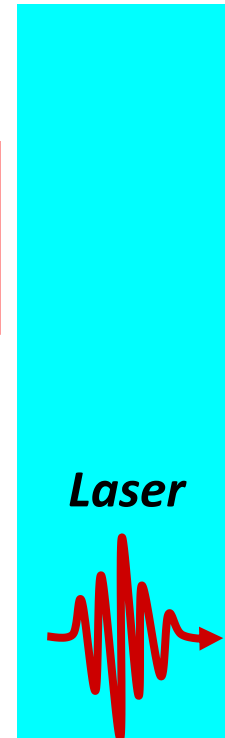
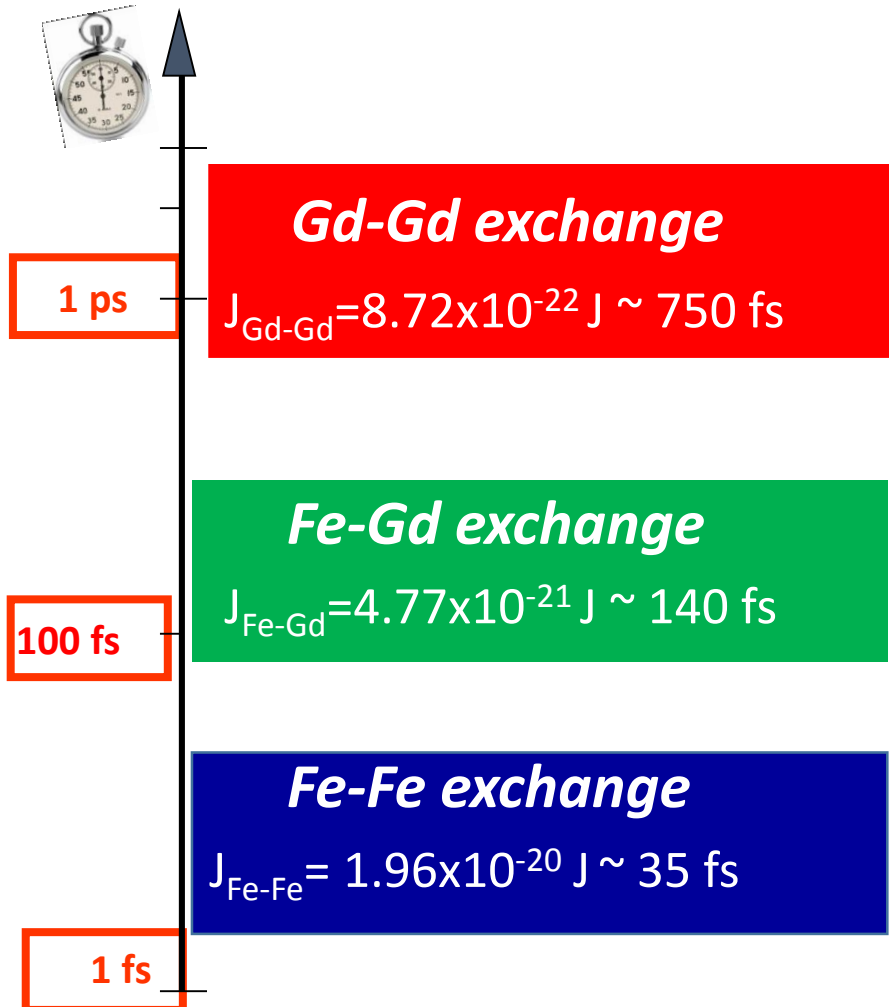
$$J_{\text{Fe-Gd}} = 4.77 \times 10^{-21} \text{ J} \sim 140 \text{ fs}$$

$$J_{\text{Gd-Gd}} = 8.72 \times 10^{-22} \text{ J} \sim 750 \text{ fs}$$

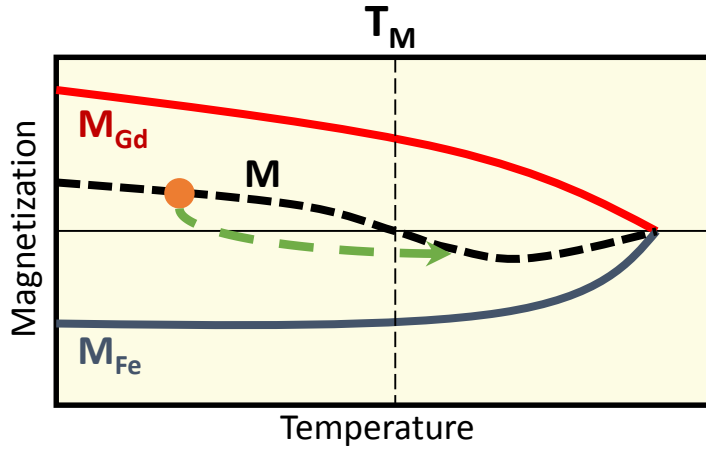


How do spins respond to a stimulus faster than the exchange interaction?

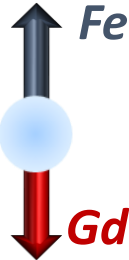
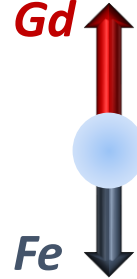
RE-TM ferrimagnetic alloy as a model system



Ultrafast heating of GdFeCo?

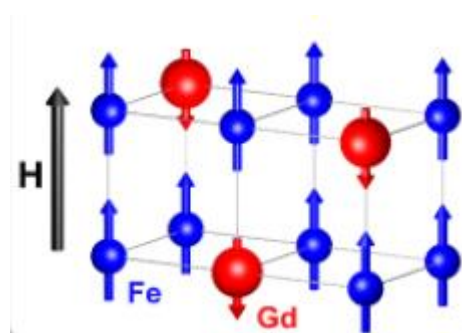
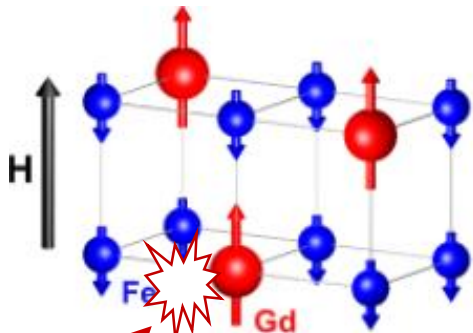


H ↑



*Below
the compensation point*

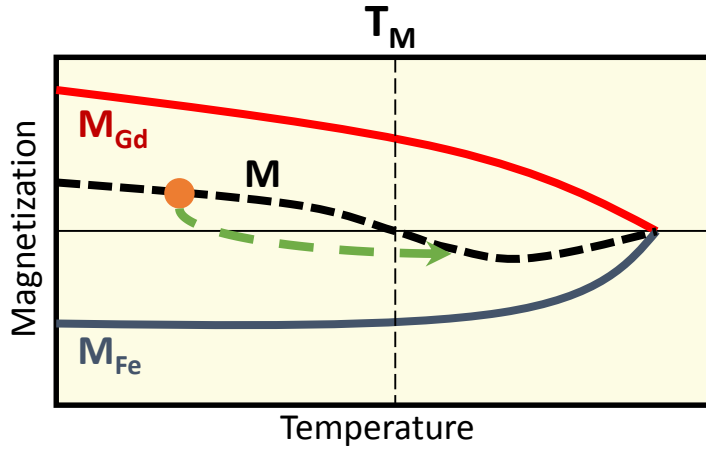
*Above
the compensation point*



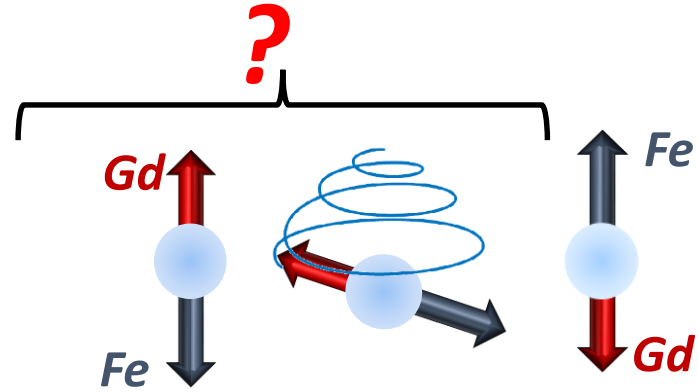
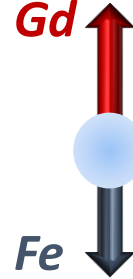
*Ultrafast (60 fs)
heating pulse*

What is the dynamics?

Ultrafast heating of GdFeCo?

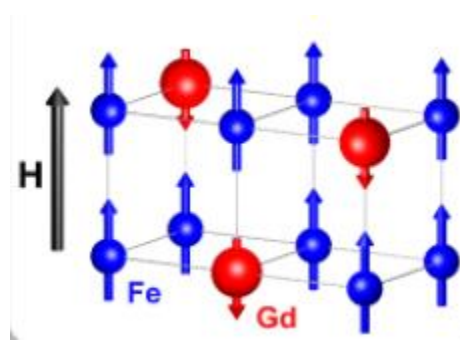
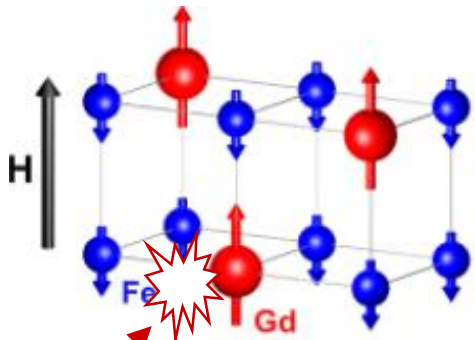


H ↑



*Below
the compensation point*

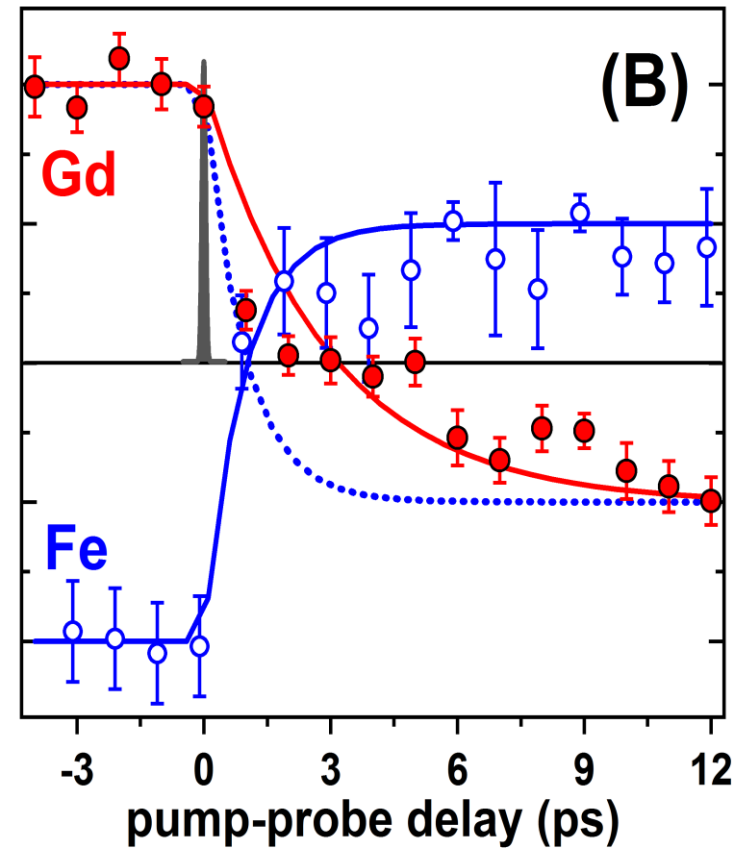
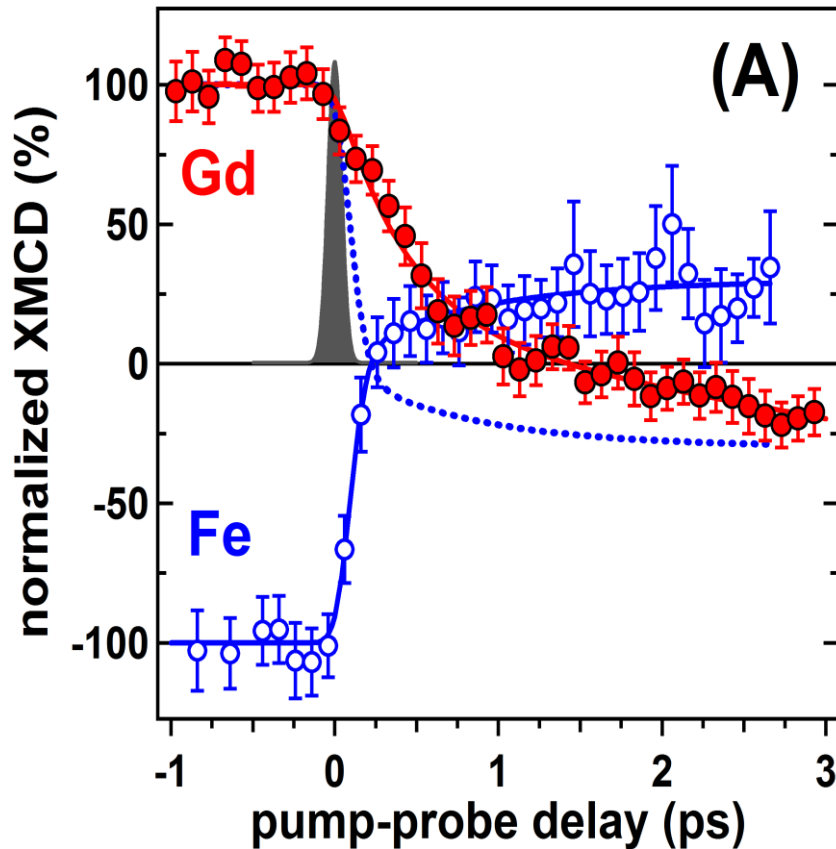
*Above
the compensation point*



*Ultrafast (60 fs)
heating pulse*

What is the dynamics?

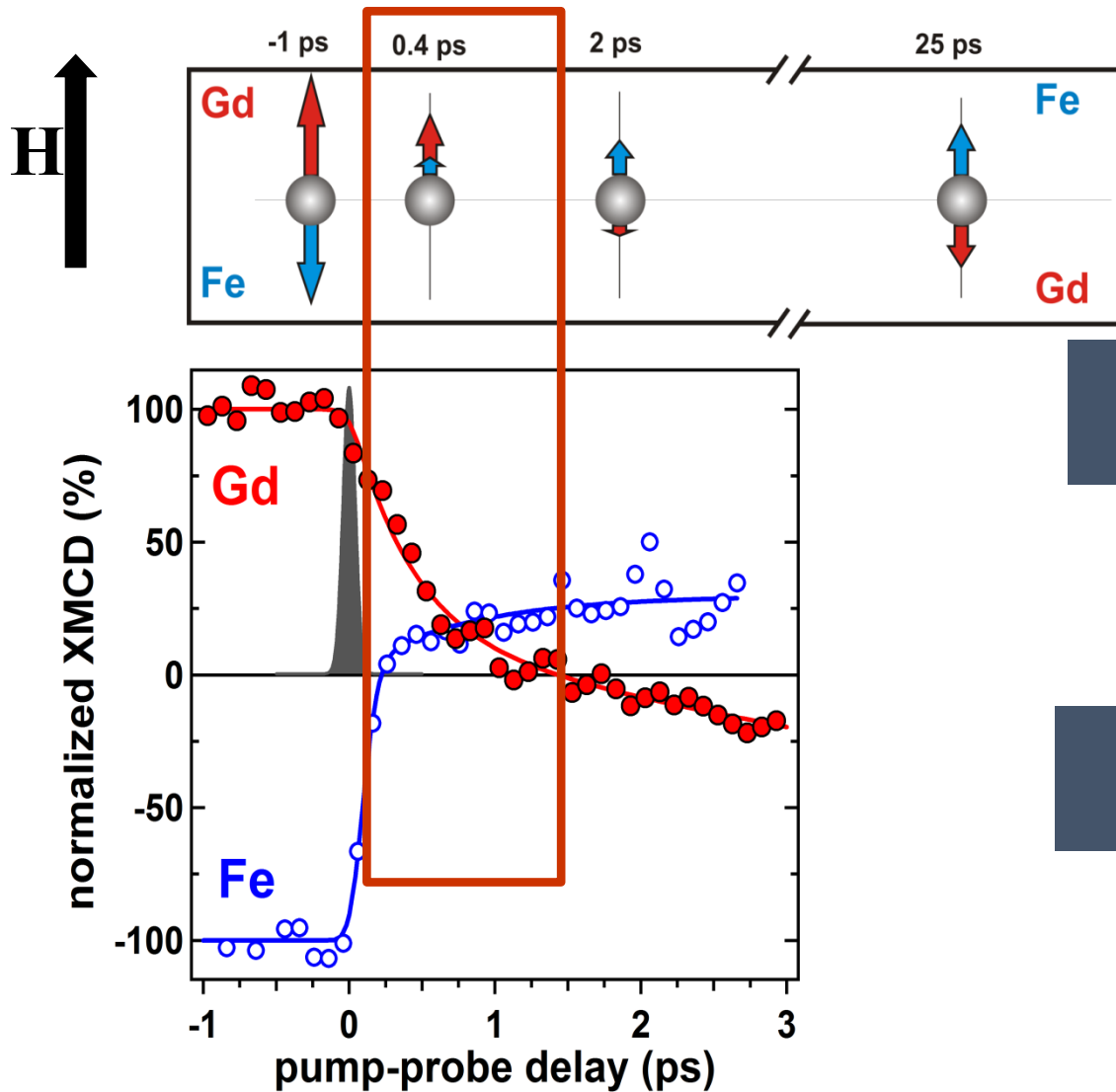
Experiment: Ultrafast dynamics of antiferromagnetically coupled sublattices



Different magnetization switching dynamics at Fe and Gd sites !!!

I. Radu et al, *Nature* **472**, 205-208 (2011).

Ultrafast “ferromagnetism” of antiferromagnetically coupled spins



$$J_{\text{Fe-Gd}} \sim 140 \text{ fs}$$

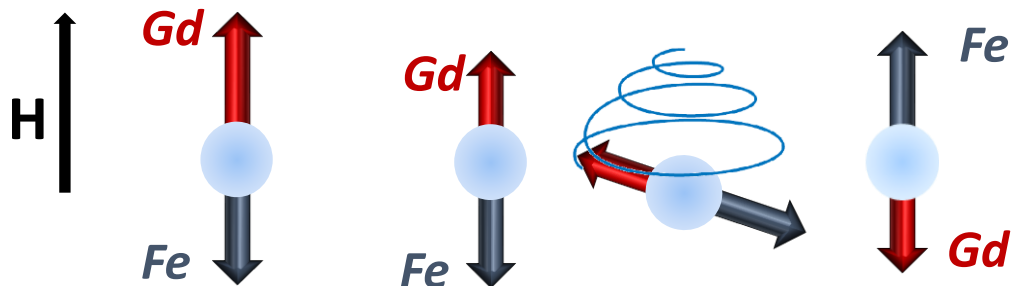
Transient **FERROMAGNETIC**
ALIGNMENT for ~ 1.2 ps

The system evolves against the
exchange interaction !!!

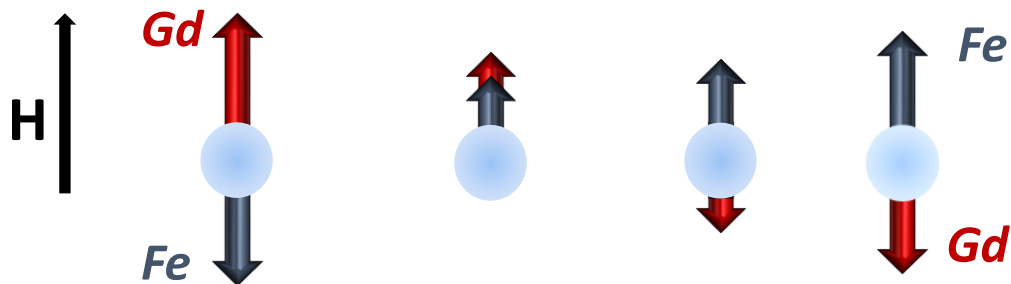
I. Radu et al, *Nature* **472**, 205-208 (2011).

Ultrafast reversal of antiferromagnetically coupled spins

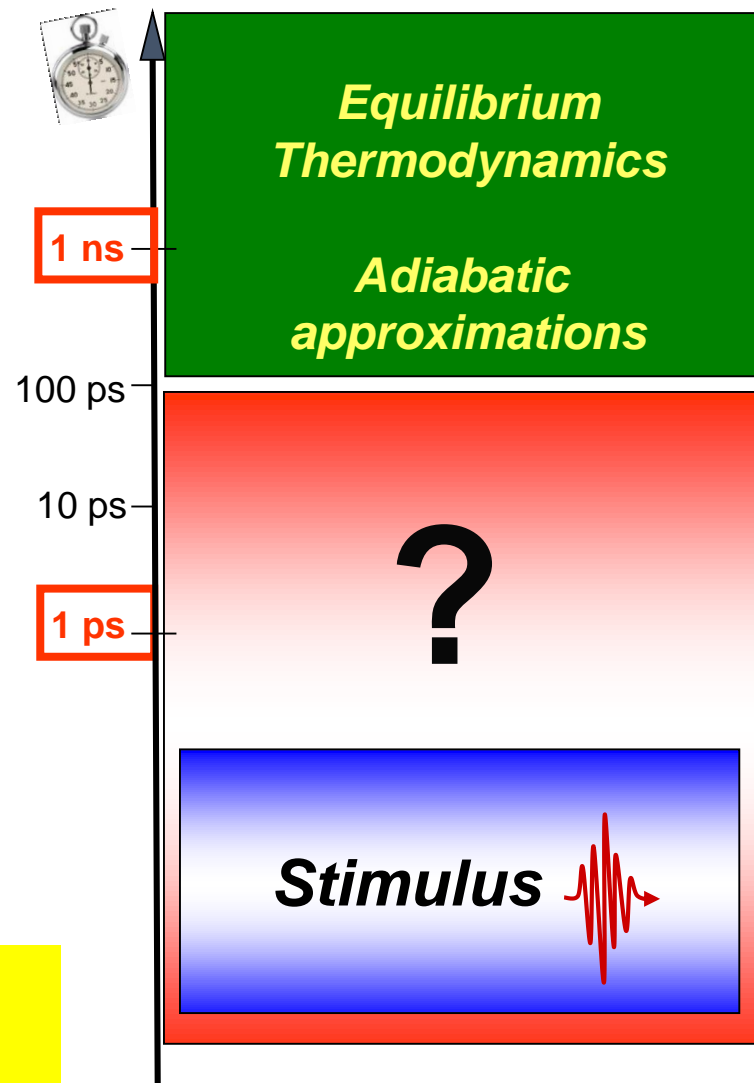
Expected thermodynamically



Observed experimentally

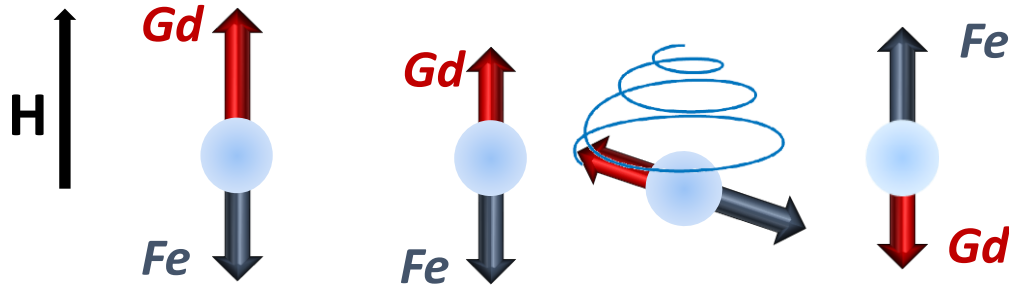


**Fe is faster than Gd! No precession!
Ferromagnetic-like state!**

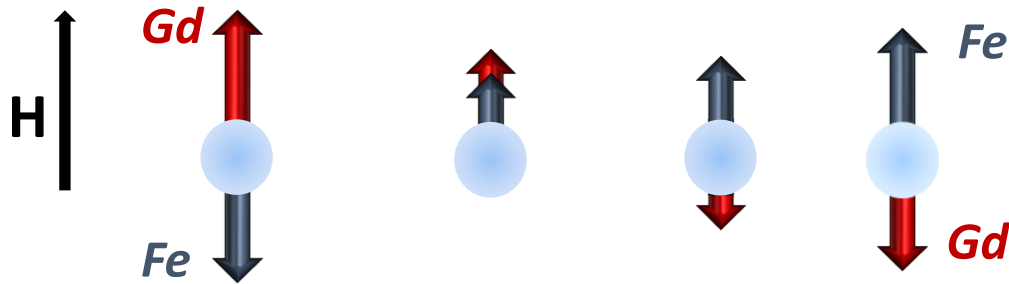


Ultrafast reversal of antiferromagnetically coupled spins

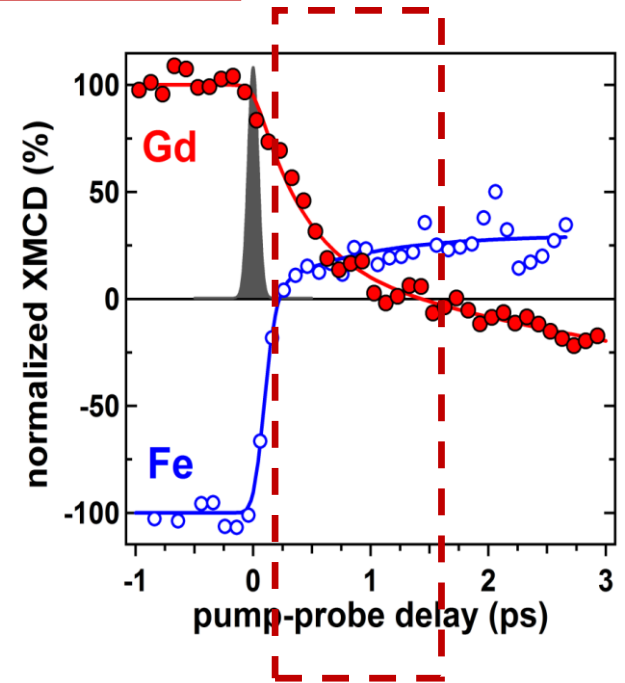
Expected thermodynamically



Observed experimentally



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The system evolves against the exchange interaction !!!

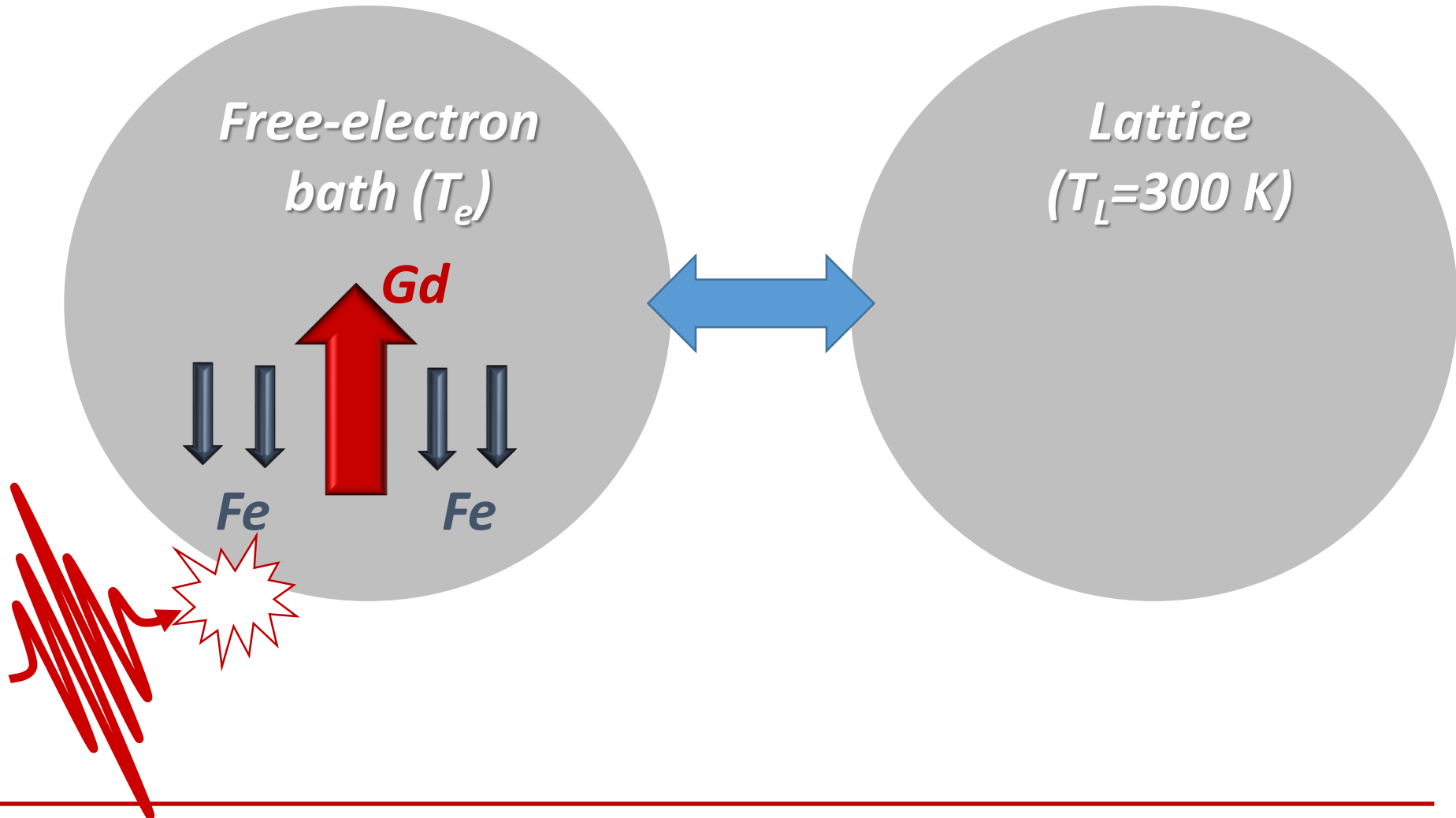
$$H \approx 0.3 \text{ T} \quad H_{\text{ex}} \approx 10 \text{ T}$$

$$H \ll H_{\text{ex}}$$

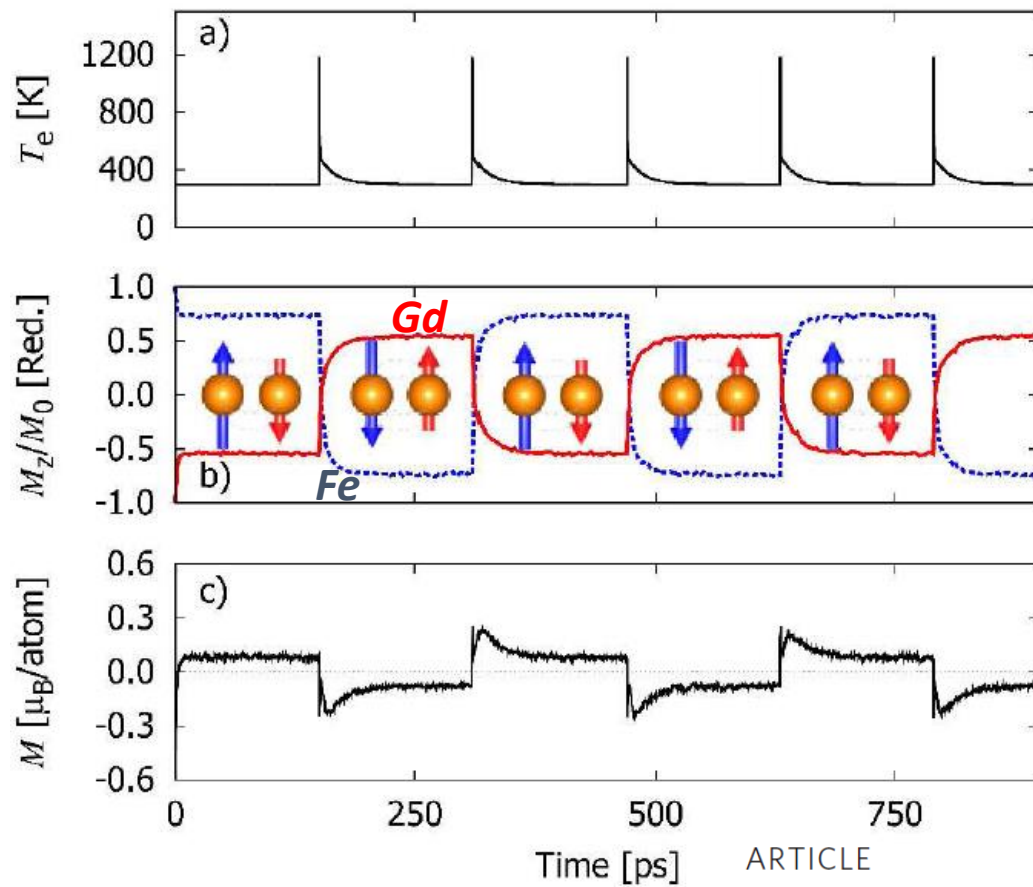
Atomistic simulations of ultrafast spin dynamics

- *Localized atomistic spin model with 10^6 spins coupled with a Heisenberg exchange*
- *Exchange parameters (Fe-Fe, Gd-Gd, and Fe-Gd) obtained by fitting static M_{Fe} , $M_{Gd}(T)$ dependencies*
- *Stochastic term added to the effective field.*
- *No reversing field is present during the process*

Simulated model



Atomistic simulations: Ultrafast heating as a sufficient stimulus to reverse magnetization?



ARTICLE

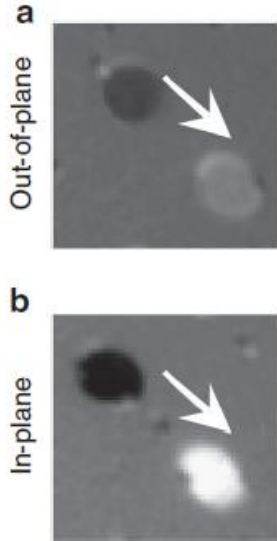
Received 11 Jul 2011 | Accepted 5 Jan 2012 | Published 7 Feb 2012

DOI:10.1038/ncomms1666

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

T.A. Ostler¹, J. Barker¹, R.F.L. Evans¹, R.W. Chantrell¹, U. Atxitia², O. Chubykalo-Fesenko², S.El Moussaoui³, L. Le Guyader³, E. Mengotti³, L.J. Heyderman³, F. Nolting³, A. Tsukamoto⁴, A. Itoh⁴, D. Afanasiev⁵, B.A. Ivanov⁵, A.M. Kalashnikova⁶, K. Vahaplar⁷, J. Mentink⁷, A. Kirilyuk⁷, Th. Rasing⁷ & A.V. Kimel⁷

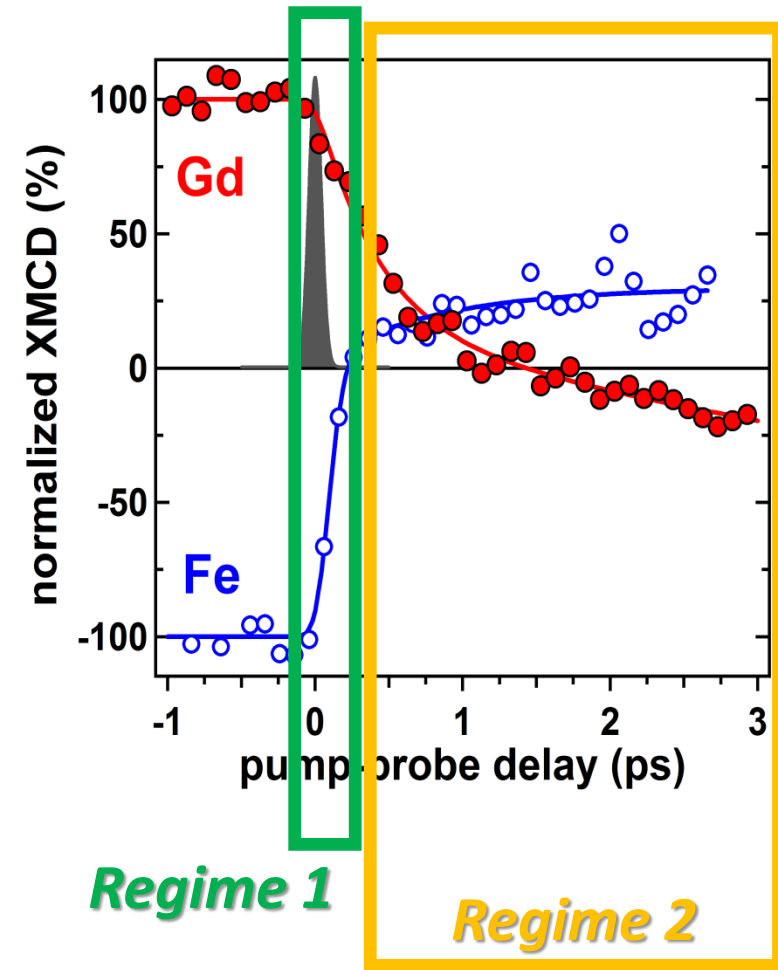
Do we need a magnetic field at all?



Ultrafast heating alone is a sufficient stimulus to reverse magnetization!

T. Ostler et al, *Nature-Communications*
DOI: 10.1038/ncomms1666 (2012).

Ultrafast spin dynamics in multi-sublattice magnets



Regime 1

- Temperature of electron gas is higher than the Curie temperature

$$T_e \gg T_C$$

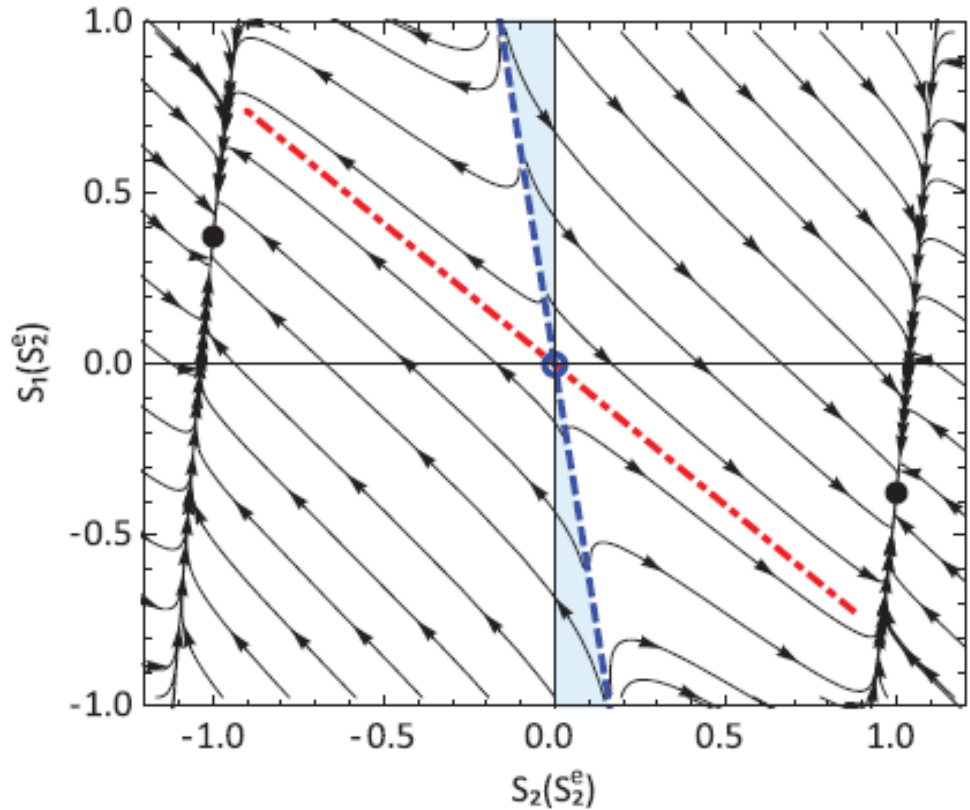
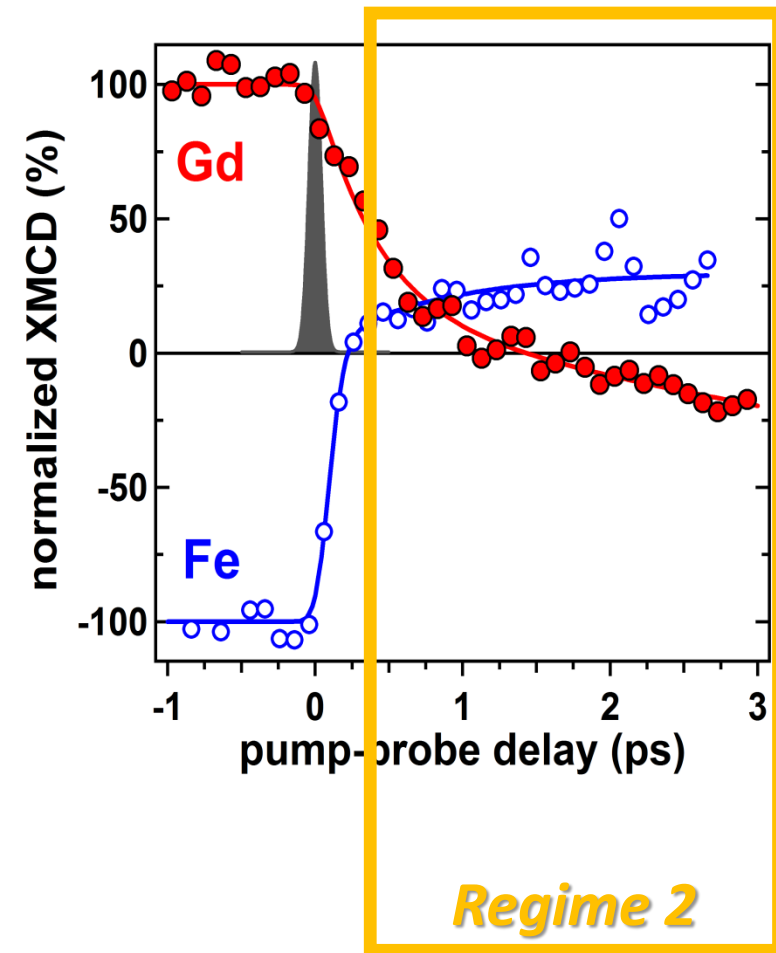
$$\frac{dM_i}{dt} = -\gamma M_i \times H_i + R_i,$$

$$\frac{dM_i}{dt} = -\frac{M_i - M_i^{(0)}}{\tau_i} \quad \tau_i = \frac{\mu_i}{2\alpha\gamma k_B T}$$

$$\mu_{Gd} > \mu_{Fe}$$

$$\tau_{Gd} > \tau_{Fe}$$

Ultrafast spin dynamics in multi-sublattice magnets

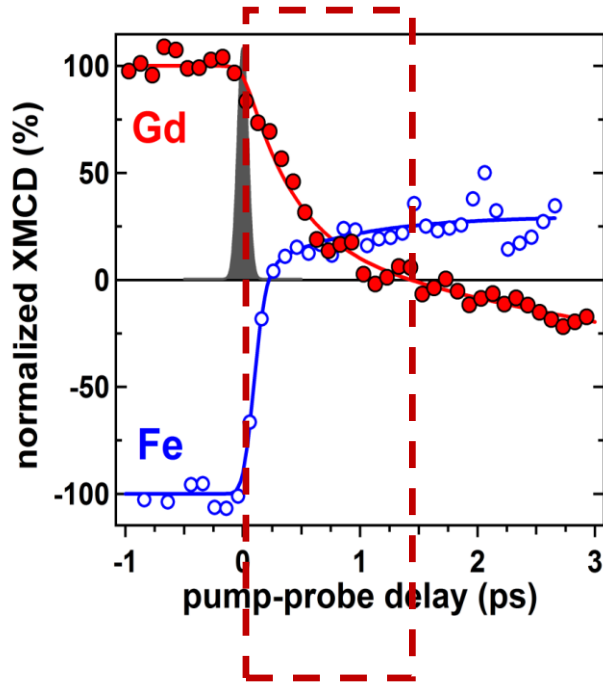


$$\frac{dM_{\text{Fe}}}{dt} = -\frac{dM_{\text{Gd}}}{dt}$$

J. Mentink et al

Physical Review Letters **108**, 057202 (2012).

Open questions

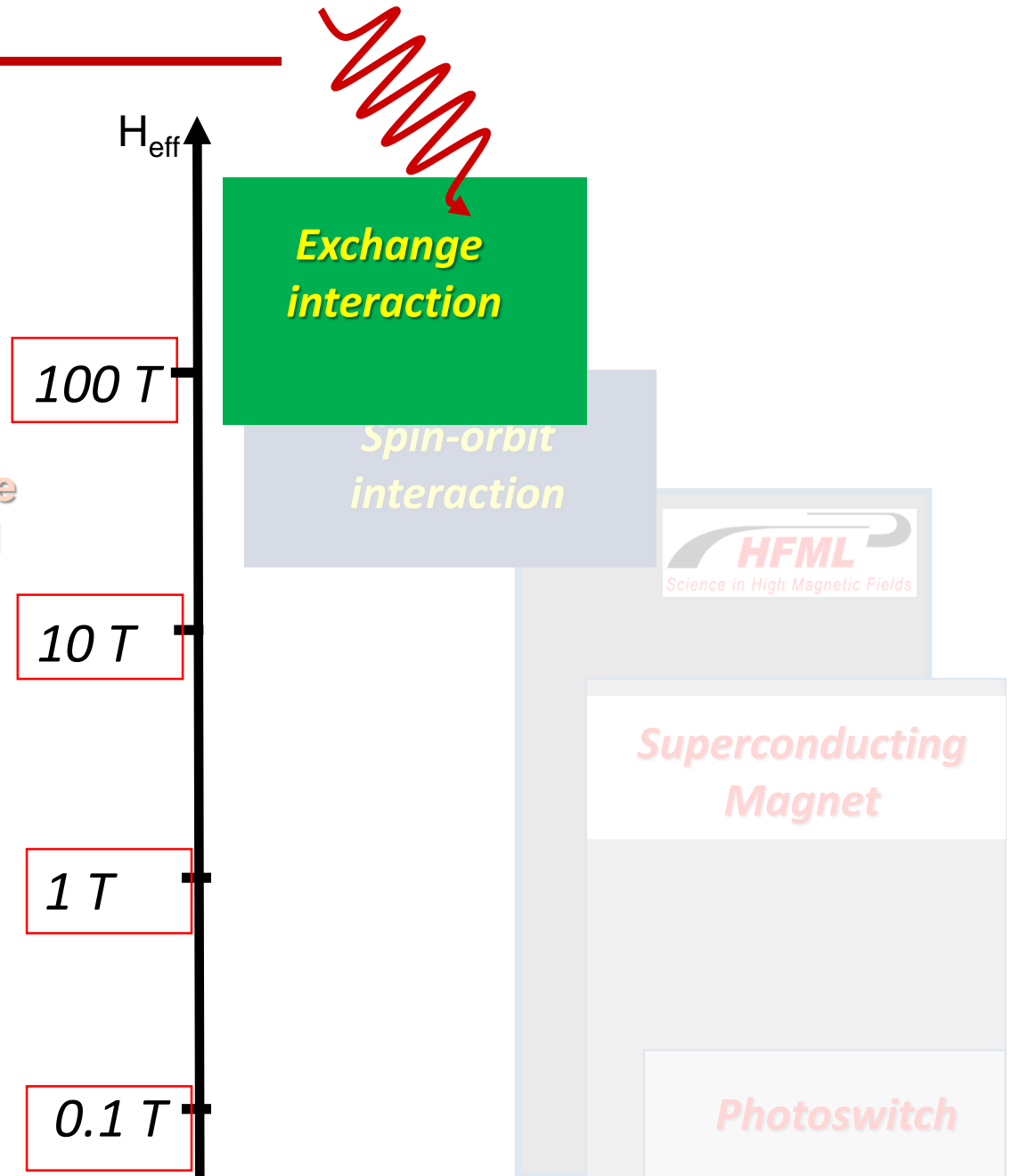


What is the effect of light on the exchange interaction?

Questions

Can we harness the exchange interaction for optical control of spins?

Can light control the exchange interaction?



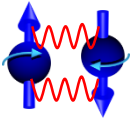
Experimental challenge:

how to demonstrate an ultrafast control of the exchange interaction

Antiferromagnet

Initial state

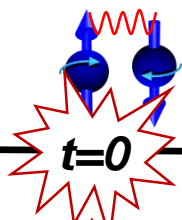
$$E_{ex} = JS_i S_j$$



$t < 0$

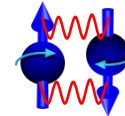
Laser excitation

$$E_{ex} = (J + \Delta U) S_i S_j$$



Final state

$$E_{ex} = JS_i S_j$$



$t \gg \tau$

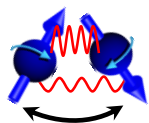
Experimental challenge:

how to demonstrate an ultrafast control of the exchange interaction

Canted Antiferromagnet

Initial state

$$E_{ex} = JS_i S_j + D[S_i \times S_j]$$

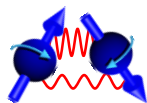


2θ

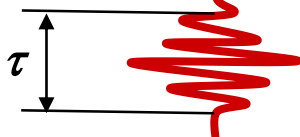
$t < 0$

Laser excitation

$$E_{ex} = (J + \Delta J)S_i S_j + (D + \Delta D)[S_i \times S_j]$$



$t = 0$

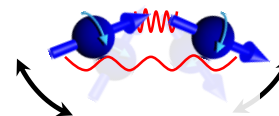


τ

$$D/J \neq (D + \Delta D)/(J + \Delta J)$$

Final state

$$E_{ex} = JS_i S_j + D[S_i \times S_j]$$



$t \gg \tau$

$$\tan(2\theta) = D/J$$

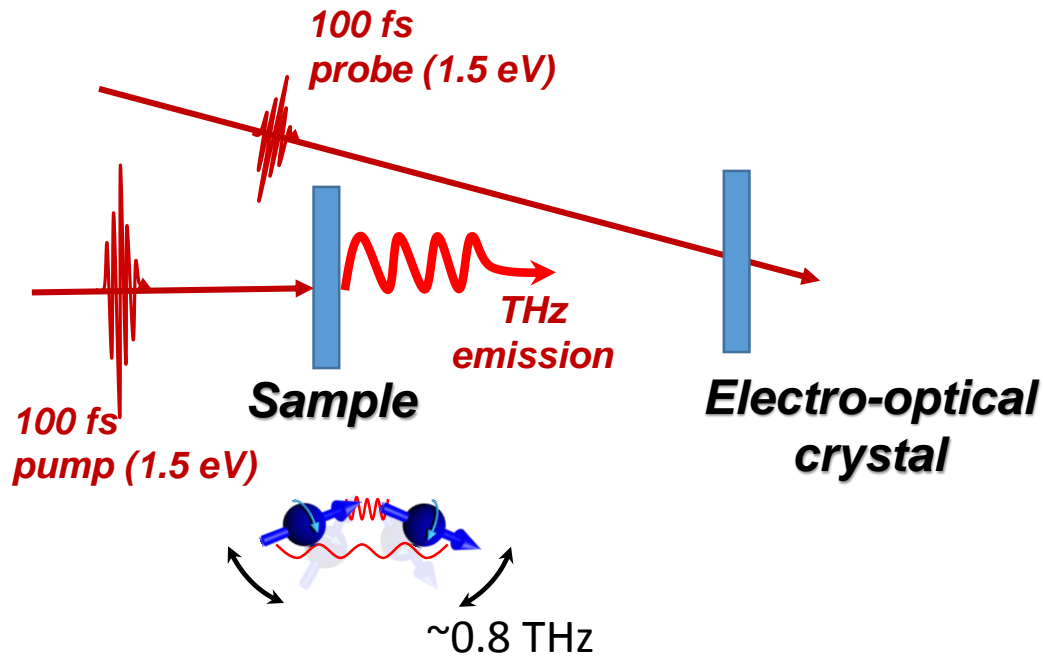
inspired by
S. O. Demokritov et al,
JETP Letters **41**, 38 (1985).

$$\mathbf{T}_{i,j} = -\gamma \left[\mathbf{S}_i \times \frac{\partial \Phi_{\text{IMR}}}{\partial \mathbf{S}_i} \right] = -\gamma \Delta J [\mathbf{S}_i \times \mathbf{S}_j]$$

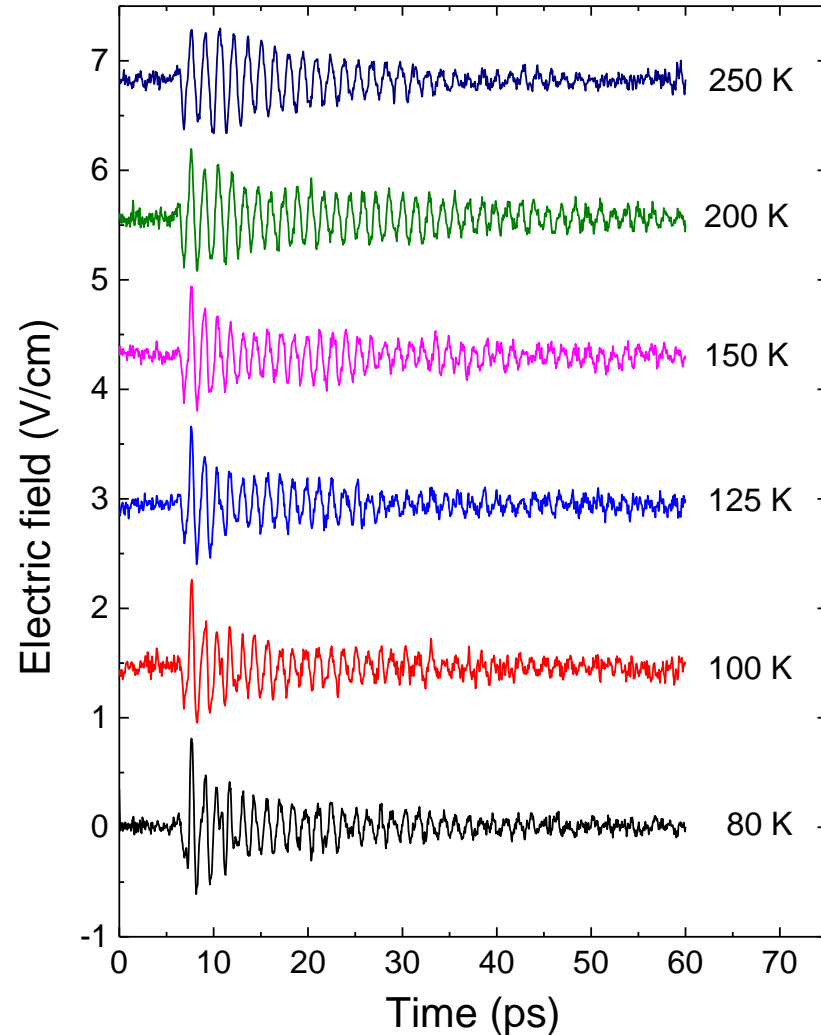
$\alpha\text{-Fe}_2\text{O}_3$, FeBO_3 , RFeO_3 (R=Y, Er, Tm etc)

Solution: Laser-induced excitation of the antiferromagnetic resonance in weak ferromagnets!

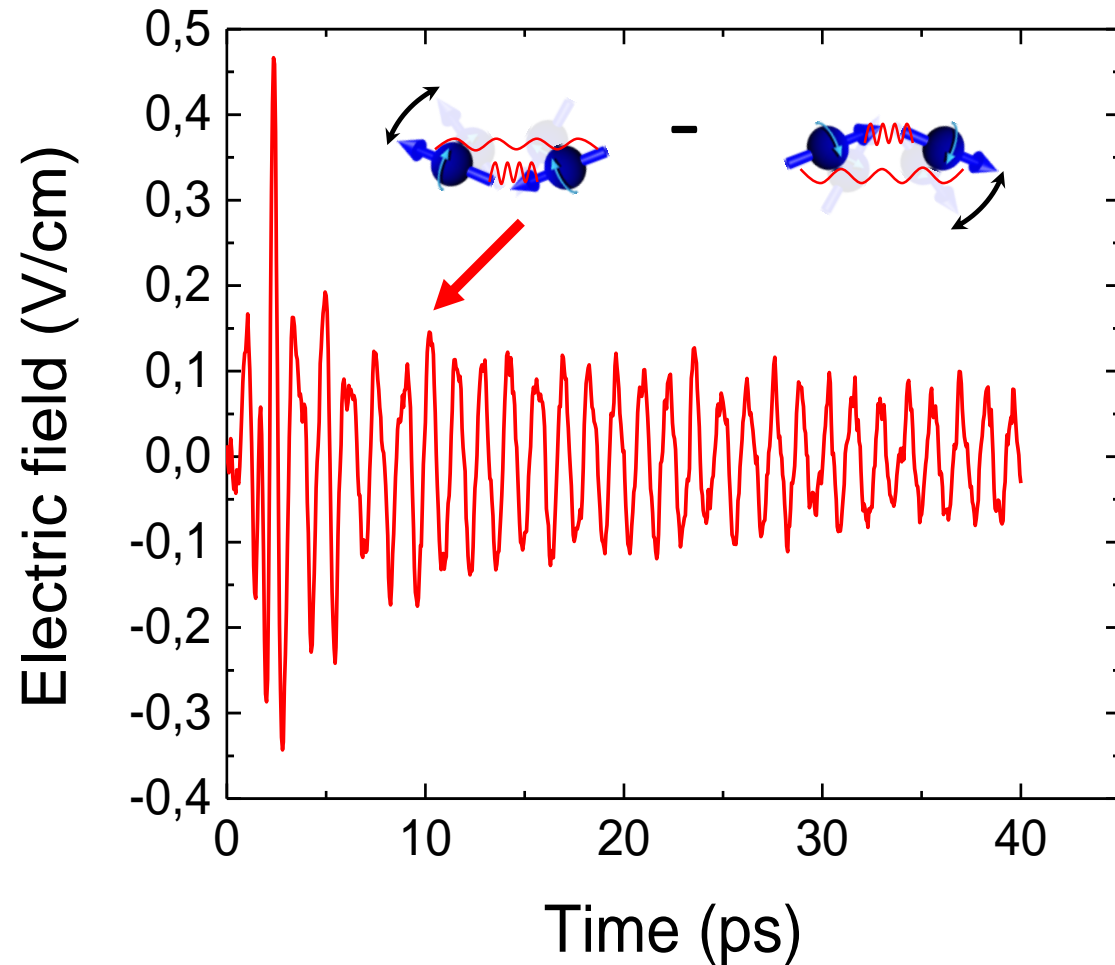
Femtosecond excitation of the antiferromagnetic resonance in ErFeO_3



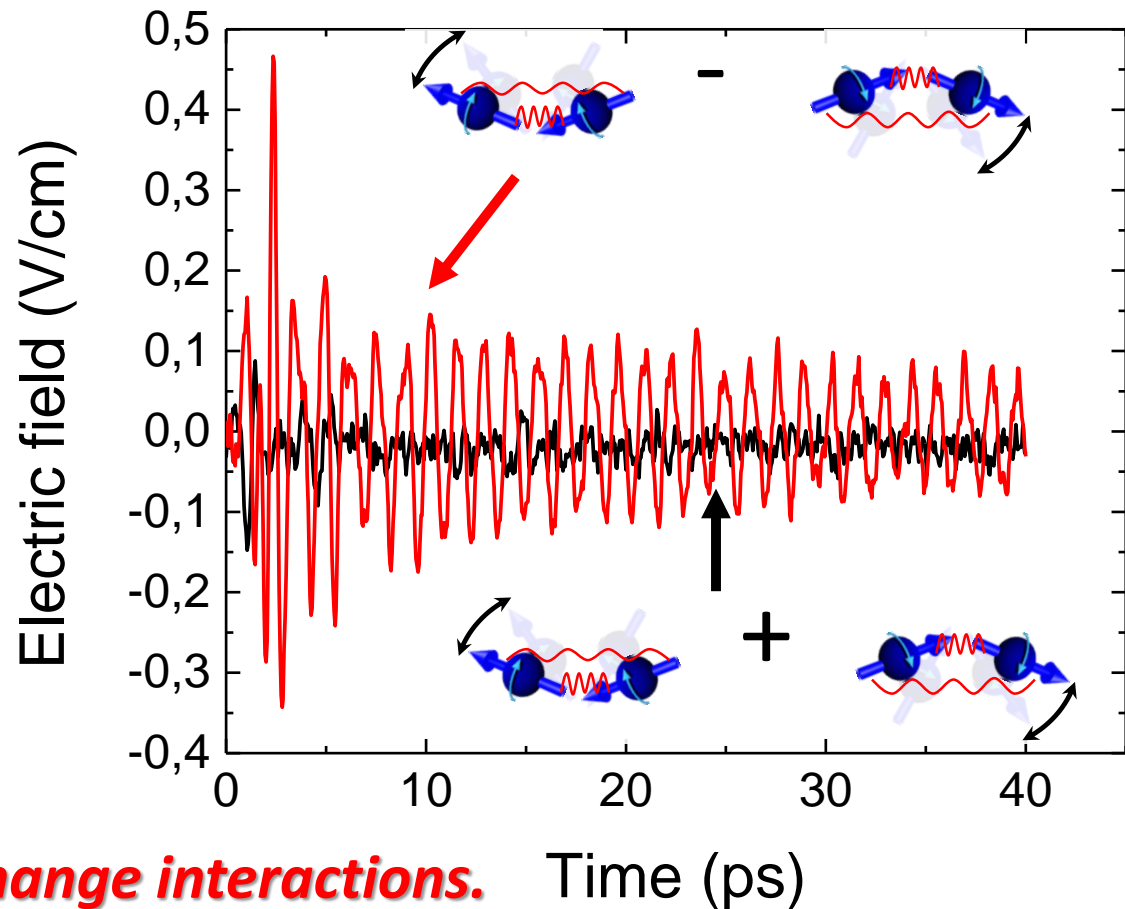
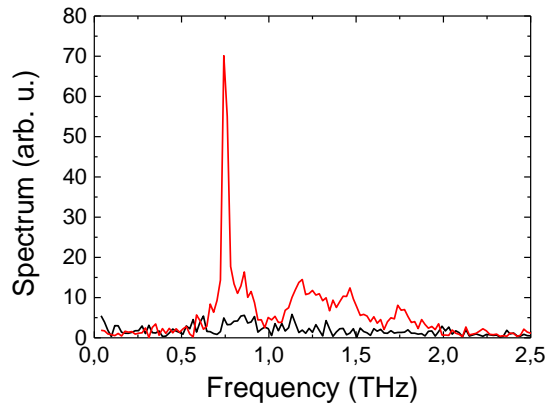
Is it spin resonance?



Femtosecond excitation of the antiferromagnetic resonance in ErFeO₃



Femtosecond excitation of the antiferromagnetic resonance in ErFeO₃



Light affects the exchange interactions. Time (ps)
 $D/J \neq \text{constant}$

How can light change the exchange interaction?

Light OFF

$$\Phi = \Phi_0 + \sum_{i,j} J_{ij} S_i S_j$$

Light ON

$$\Phi = \Phi_0 + \sum_{i,j} (J_{ij} + \Delta J) S_i S_j$$

Is it possible?

If yes $\Delta J = \alpha |E|^2$,

$$\Phi = \Phi_0 + \sum_{i,j} J_{ij} S_i S_j + \alpha |E|^2 \sum_{i,j} S_i S_j$$

$$\Delta J = \alpha |E|^2$$

Dielectric permittivity

$$\varepsilon_{kl} = \frac{\partial^2 \Phi}{\partial E_k \partial E_l^*} = \varepsilon^{(0)} + \alpha \sum_{i,j} S_i S_j$$

$\varepsilon = f(M^2)$ **Isotropic magneto-refraction**

- **Polarization insensitive**
- **Orientation insensitive**
- **Ultrafast**
- **Any material**

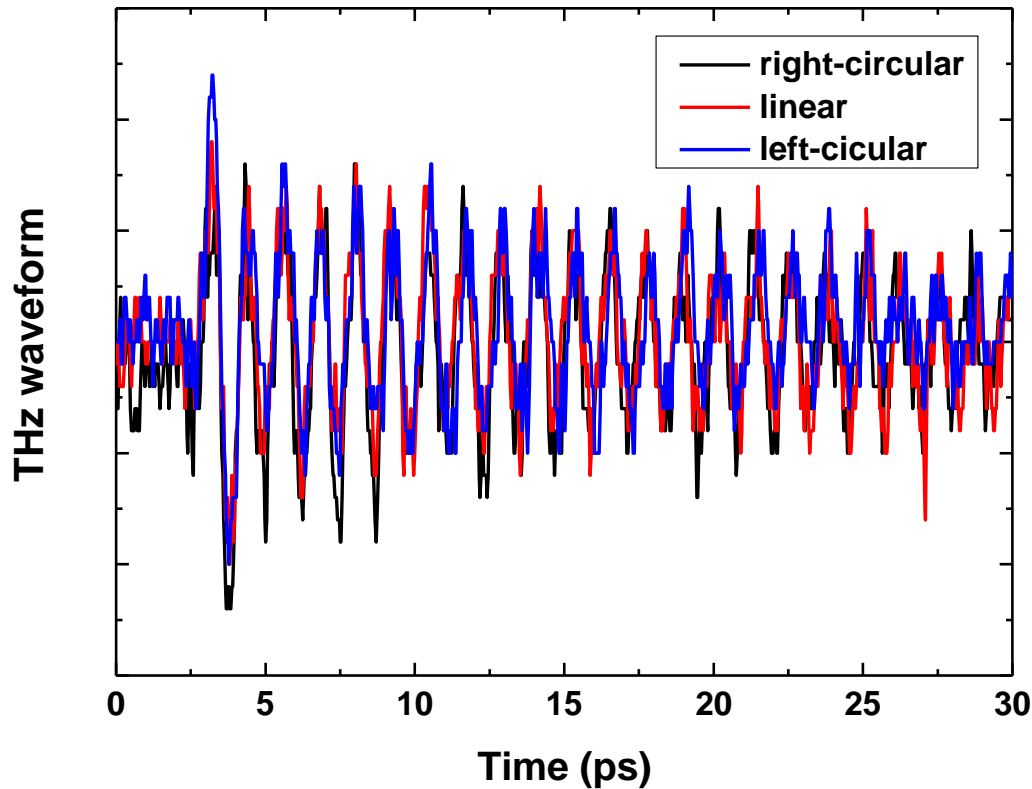
Not impossible!

see also

S. O. Demokritov et al,
JETP Letters **41**, 38 (1985).

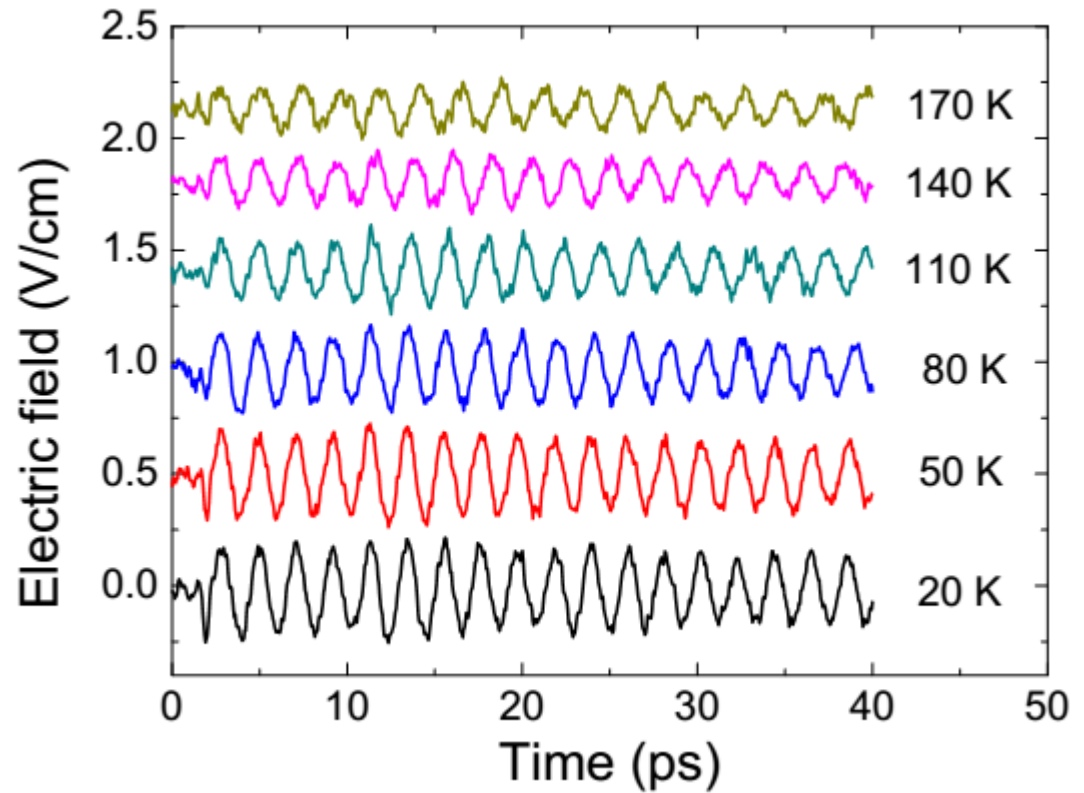
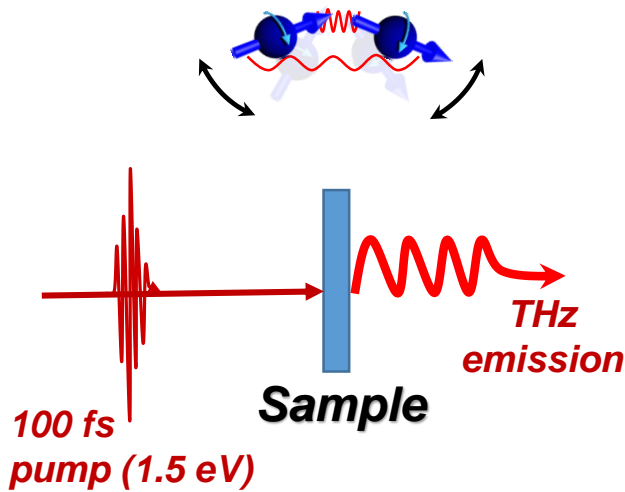
Is the optically controlled exchange polarization insensitive?

TmFeO₃



The effect of light on the exchange interactions D/J is polarization insensitive

Femtosecond excitation of the antiferromagnetic resonance in FeBO_3

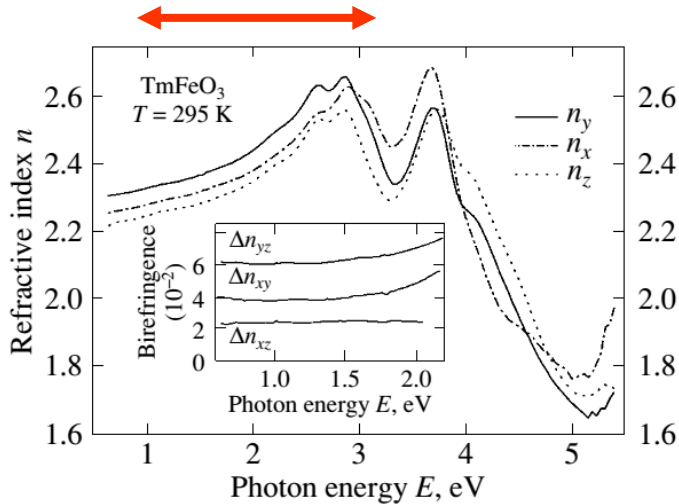


Light affects the exchange interactions.
 $D/J \neq \text{constant}$

How can light change the exchange interaction?

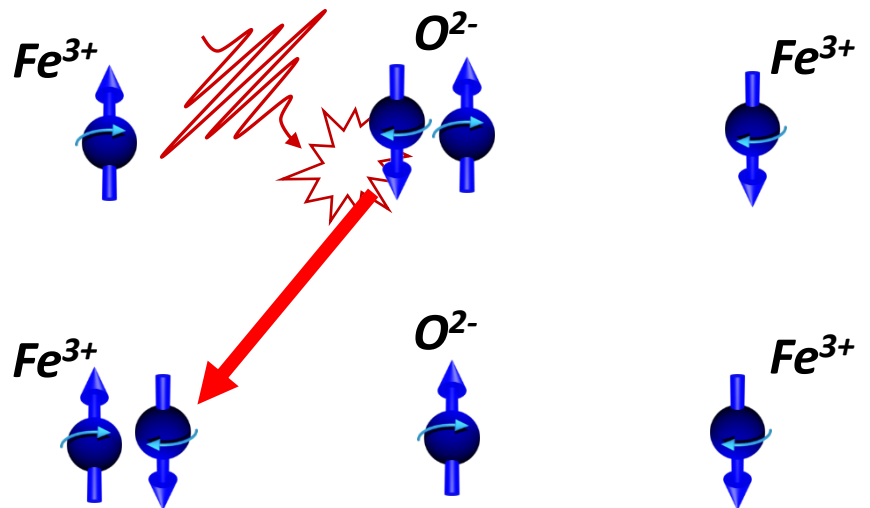
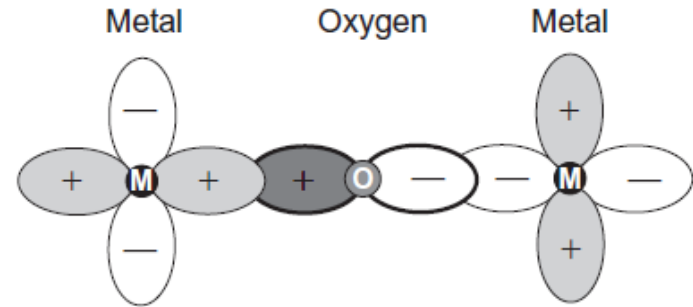
α -Fe₂O₃, FeBO₃, RFeO₃ (R=Y, Er, Tm etc)

Charge transfer transitions



Phys. Sol. State **47** 2292 (2005).

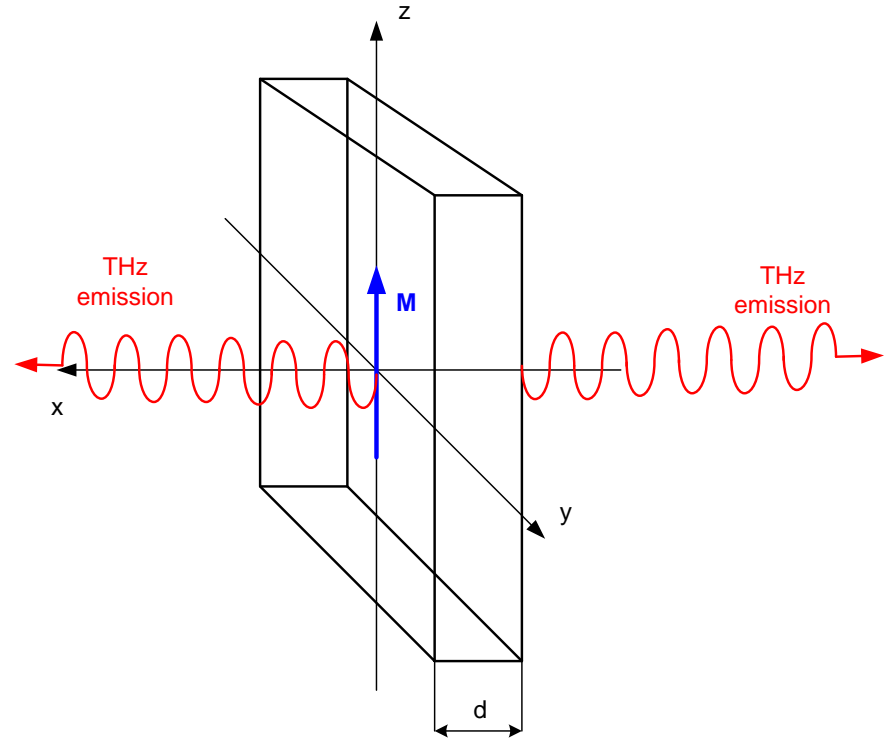
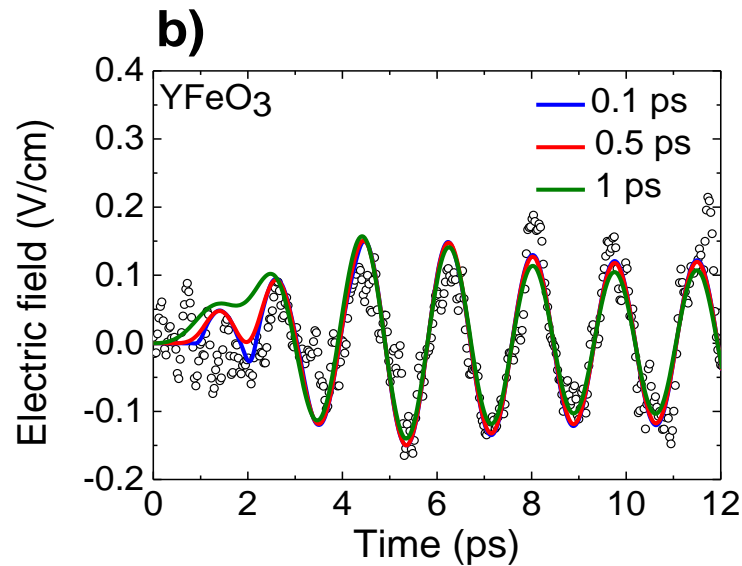
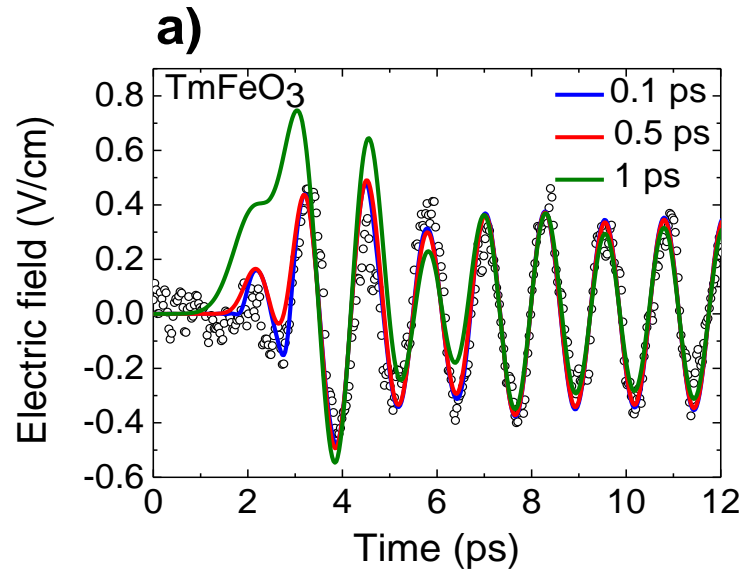
off-resonant pumping!



J. Stöhr and H. C. Siegmann

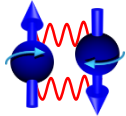
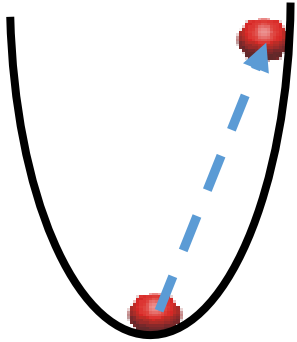
“Magnetism. From Fundamentals to nanoscale dynamics”
Springer (2006)

Is the optically controlled exchange ultrafast?

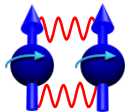
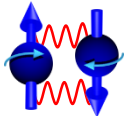


***D/J changes about 0.01%
and faster than 0.5 ps***

$\Delta E/J > 0.01\%$. Is it small?



**Do not look at spins.
Look at the energy!**



G. P. Ju et al., Phys. Rev. Lett. **93**, 197403 (2004).

J. U. Thiele, C. H. Back, Appl. Phys. Lett. **85**, 2857 (2004).

T. Li et al., Nature **496**, 69 (2013).

$$\rho_{\text{Fe}_2\text{O}_3} = 5.25 \cdot 10^3 \text{ kg/m}^3$$

$$m_{\text{Fe}_2\text{O}_3} = 160 \cdot 1.6 \cdot 10^{-27} \text{ kg} \approx 2.5 \cdot 10^{-25} \text{ kg}$$

Pumped volume (1mm*1mm*100 μm)

$$V = 10^{-10} \text{ m}^3$$

$$\text{Pumped spin pairs } N = \rho_{\text{Fe}_2\text{O}_3} V / m_{\text{Fe}_2\text{O}_3} \approx 2 \cdot 10^{18}$$

$$\text{Exchange energy of 1 spin } E_W = 3kT_N \approx 4 \cdot 10^{-20} \text{ J}$$

$$\text{Laser-induced exchange energy of 1 spin } \Delta E_W \approx 4 \cdot 10^{-24} \text{ J}$$

Total laser-induced exchange energy

$$N \Delta E_W \approx 8 \mu\text{J}$$

$$\rho_{\text{Fe}} = 7.8 \cdot 10^3 \text{ kg/m}^3$$

$$m_{\text{Fe}} = 56 \cdot 1.6 \cdot 10^{-27} \text{ kg} \approx 8 \cdot 10^{-26} \text{ kg}$$

Pumped volume (100 μm *100 μm *100 nm)

$$V = 10^{-15} \text{ m}^3$$

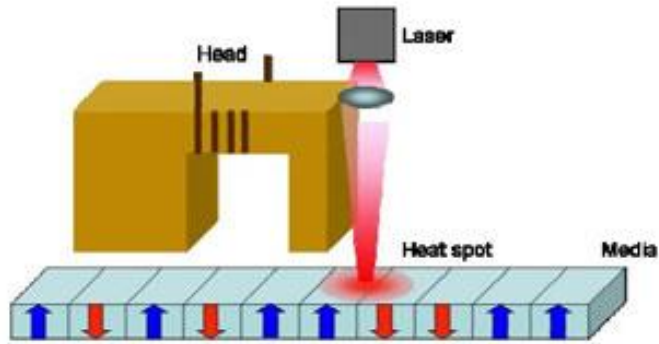
$$\text{Pumped spins } N = \rho_{\text{Fe}} V / m_{\text{Fe}} \approx 10^{14}$$

$$\text{Exchange energy of 1 spin } E_W = 3kT_C \approx 4 \cdot 10^{-20} \text{ J}$$

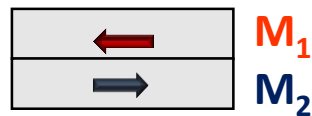
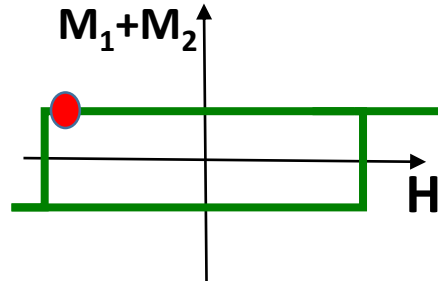
Total laser-induced exchange energy
during 100 % demagnetization

$$E_W N/2 \approx 2 \mu\text{J}$$

Novel concept for magnetic recording: Laser-assisted Recording Without Heat

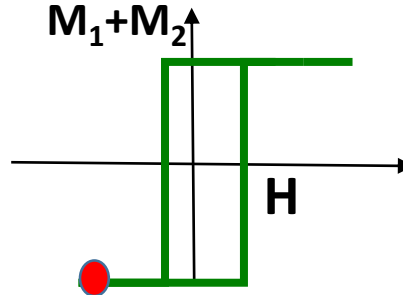


Initial state

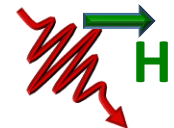
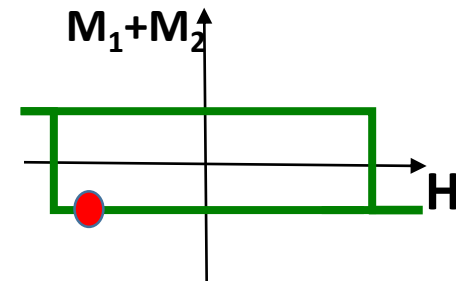


No Heat!

Laser-assist

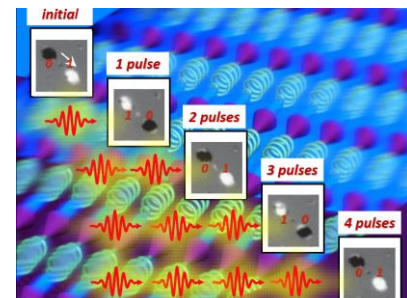
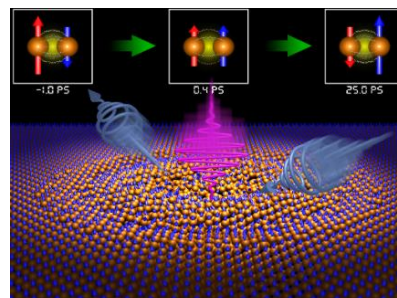
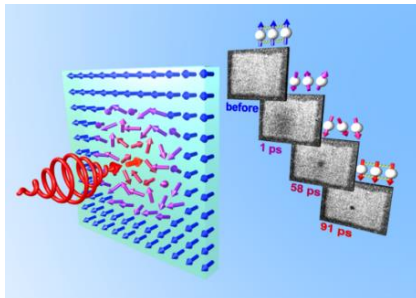
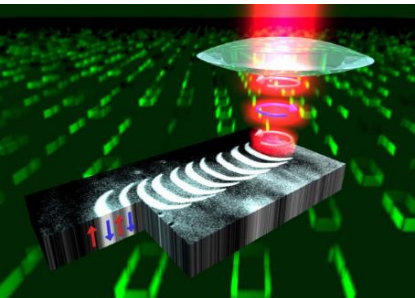


Final state



Conclusions

- *Effective fields of the exchange interaction can be harnessed for optical control of magnetism at sub-1ps time-scale*
- *Exchange interaction can be controlled ultrafast via inverse magneto refractive effect*
- *The mechanism of the control is universal*
- *The dynamics of the exchange interaction can be probed with the help of magneto-refractive effect*
- *Magneto-refraction is a key to magnetism at the time-scale of the exchange interaction*



Acknowledgements

R. Mikhaylovskiy

R. Subkhangulov

Th. Rasing

**Radboud University Nijmegen,
Nijmegen, The Netherlands**



R. V. Pisarev

**Ioffe Institute,
St. Petersburg, Russia**



A. Wu

**Shanghai Institute of Ceramics,
China**



A. B. Henriques

**Universidade de Sao Paulo,
Sao Paulo, Brazil**

P. H. O. Rappl

A. E. Abramof

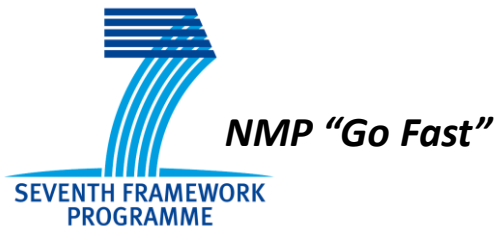
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Radboud University Nijmegen



**Starting Grant
"Femtomagnetism"**

The problem at different scales

