

The zoo of all-optical magnetic switching *mechanisms & time-scales*

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Ultrafast Spin Dynamics in Ferromagnetic Nickel

E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot

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23, rue du Loess, 67037 Strasbourg Cedex, France*
(Received 17 October 1995)



Éric Beaurepaire
28.10.1959 – 24.04.2018

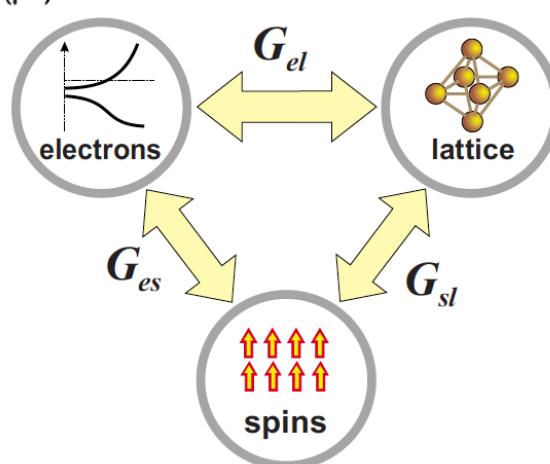
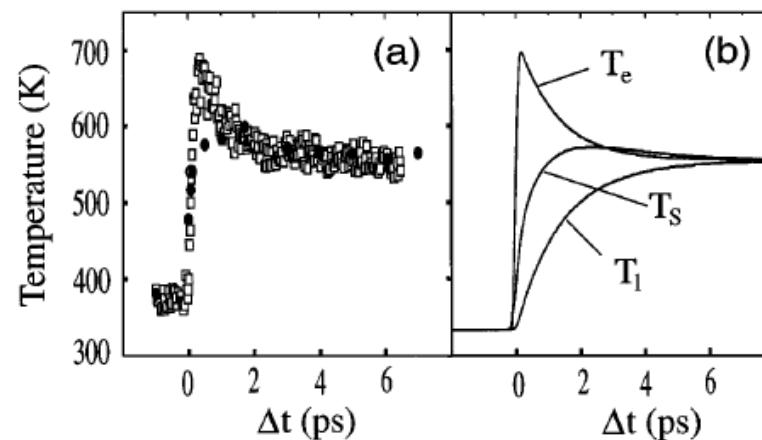
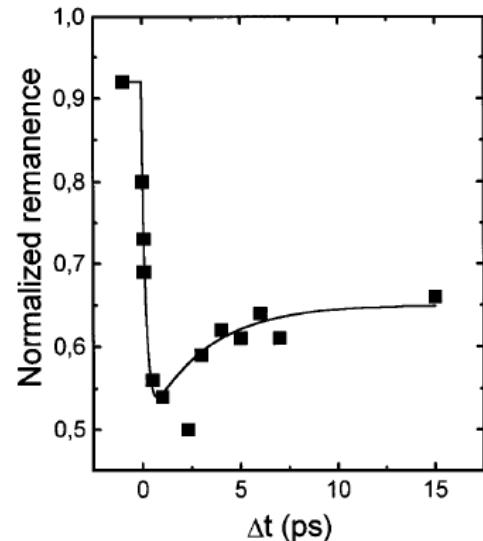


Jean-Yves Bigot
29.02.1956 – 02.05.2018

Ultrafast Spin Dynamics in Ferromagnetic Nickel

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$$C_e d(T_e)/dt = -G_{el}(T_e - T_l) - G_{es}(T_e - T_s) + P(t)$$

$$C_s d(T_s)/dt = -G_{es}(T_s - T_e) - G_{sl}(T_s - T_l),$$

$$C_l d(T_l)/dt = -G_{el}(T_l - T_e) - G_{sl}(T_l - T_s),$$

Coherent terahertz emission from ferromagnetic films excited by femtosecond laser pulses

E. Beaurepaire^{a)}

IPCMS (UMR 7504 CNRS-ULP), 23 rue du Loess, BP43, F-67034 Strasbourg Cedex 2, France

G. M. Turner, S. M. Harrel, and M. C. Beard

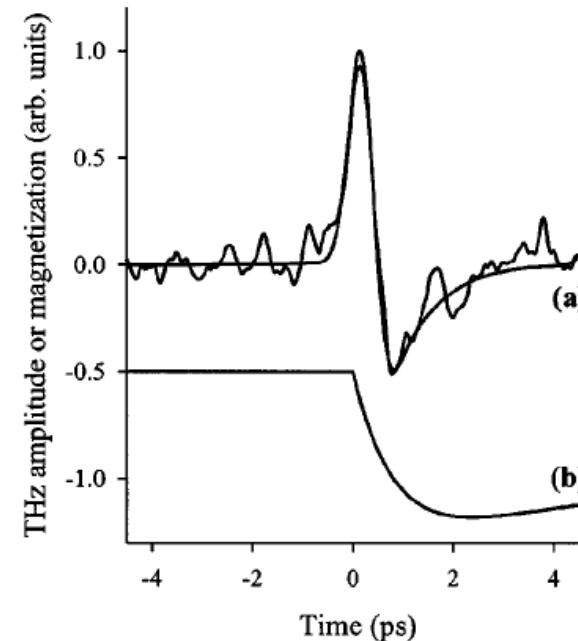
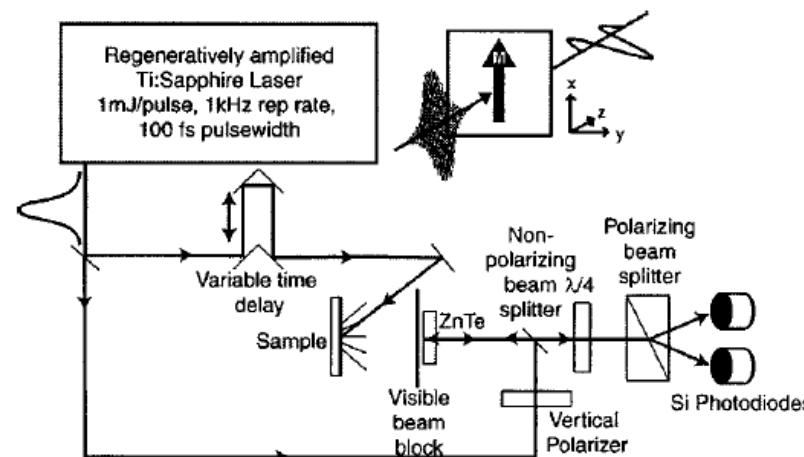
Yale University, Chemistry Department 225 Prospect Street, P.O. Box 208107, New Haven, Connecticut 06520-8107

J.-Y. Bigot

IPCMS (UMR 7504 CNRS-ULP), 23 rue du Loess, BP43, F-67034 Strasbourg Cedex 2, France

C. A. Schmuttenmaer^{a)}

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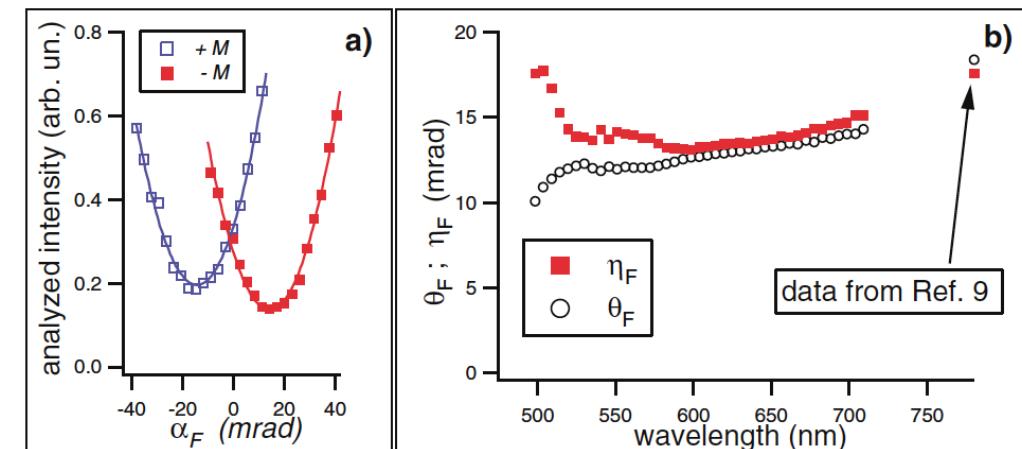
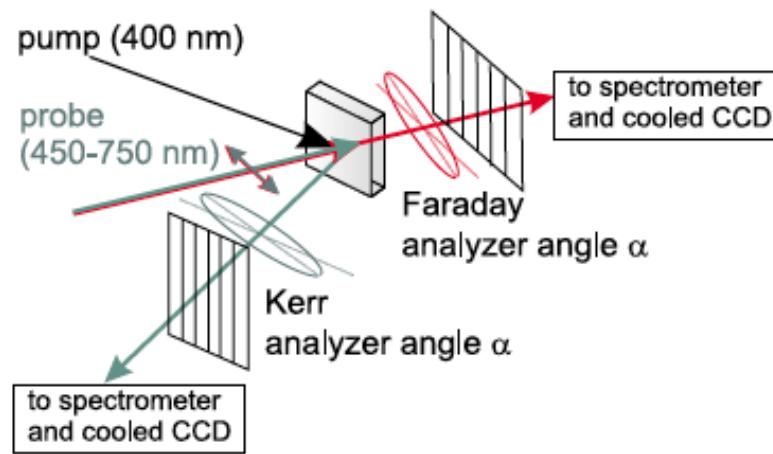


Femtosecond Spectrot temporal Magneto-optics

J.-Y. Bigot,* L. Guidoni, E. Beaurepaire, and P. N. Saeta†

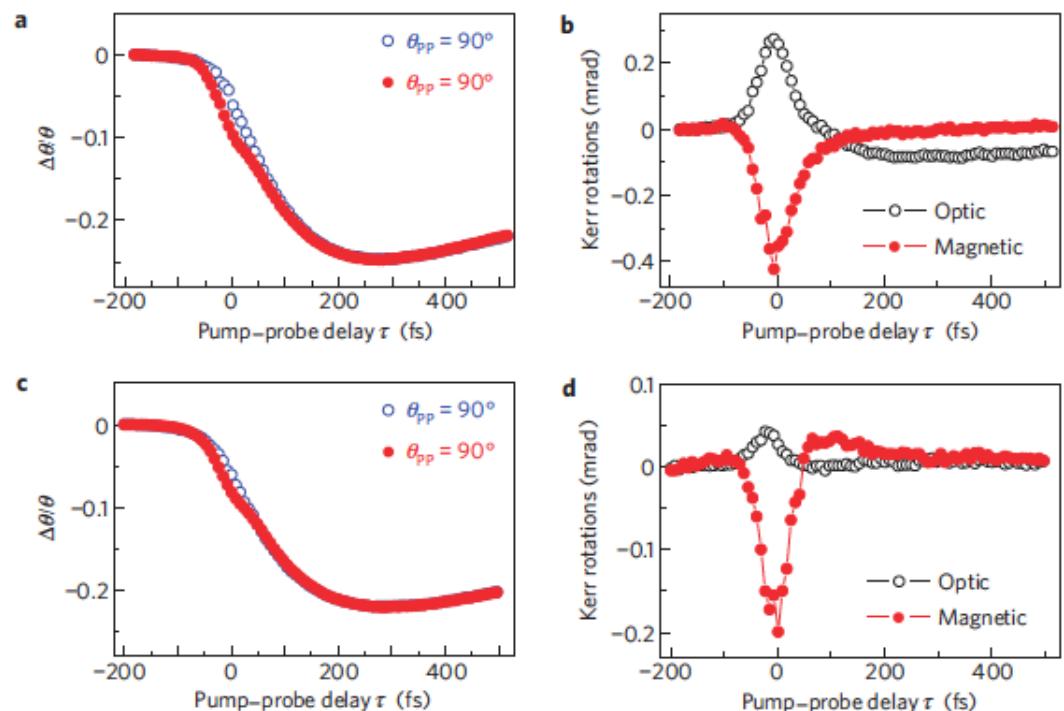
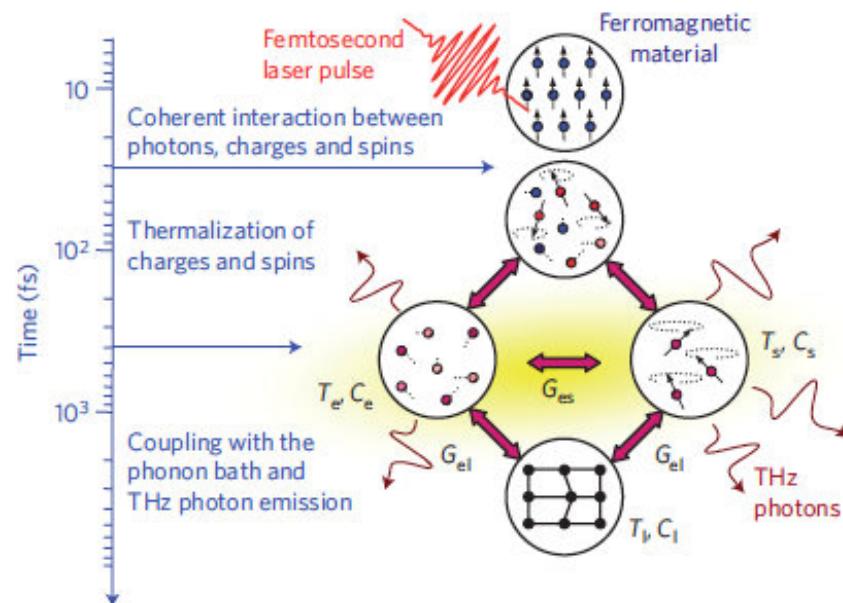
*Institut de Physique et Chimie des Matériaux de Strasbourg, Unité Mixte CNRS-ULP-ECPM,
23 rue du Loess, B.P. 43, 67034 Strasbourg Cedex, France*
(Received 31 October 2003; published 13 August 2004)

A new method to measure and analyze the time and spectrally resolved polarimetric response of magnetic materials is presented. It allows us to study the ultrafast magnetization dynamics of a CoPt₃ ferromagnetic film. The analysis of the pump-induced rotation and ellipticity detected by a broad spectrum probe beam shows that magneto-optical signals predominantly reflect the spin dynamics in ferromagnets.



Coherent ultrafast magnetism induced by femtosecond laser pulses

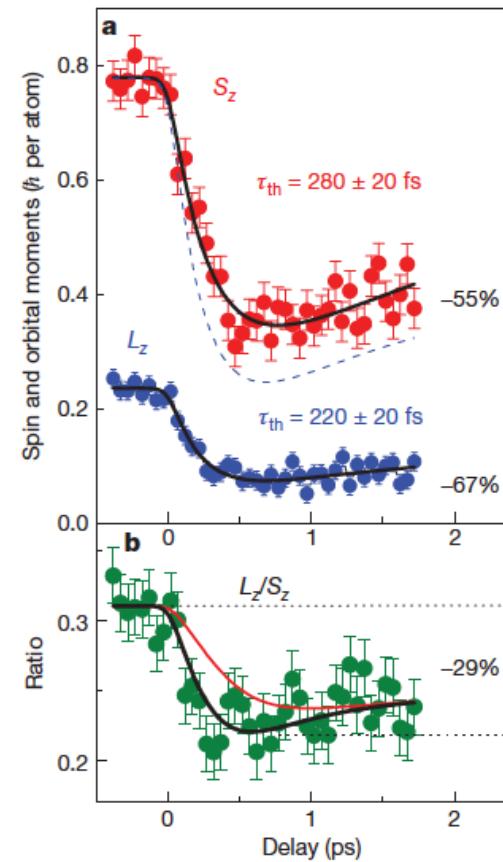
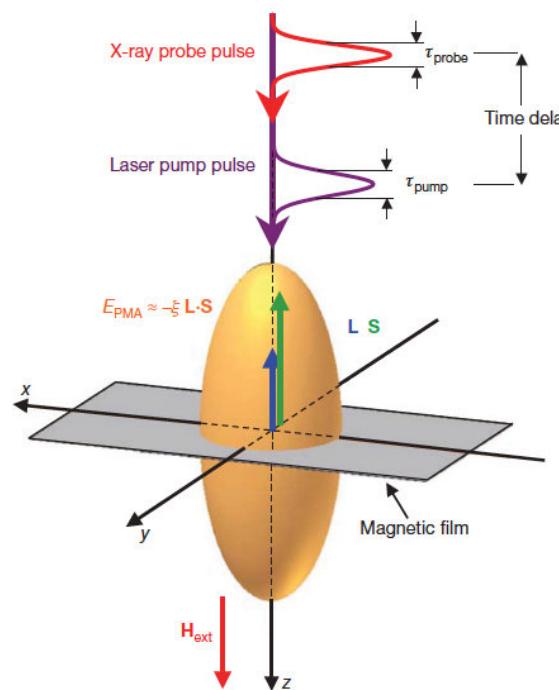
Jean-Yves Bigot*, Mircea Vomir and Eric Beaurepaire



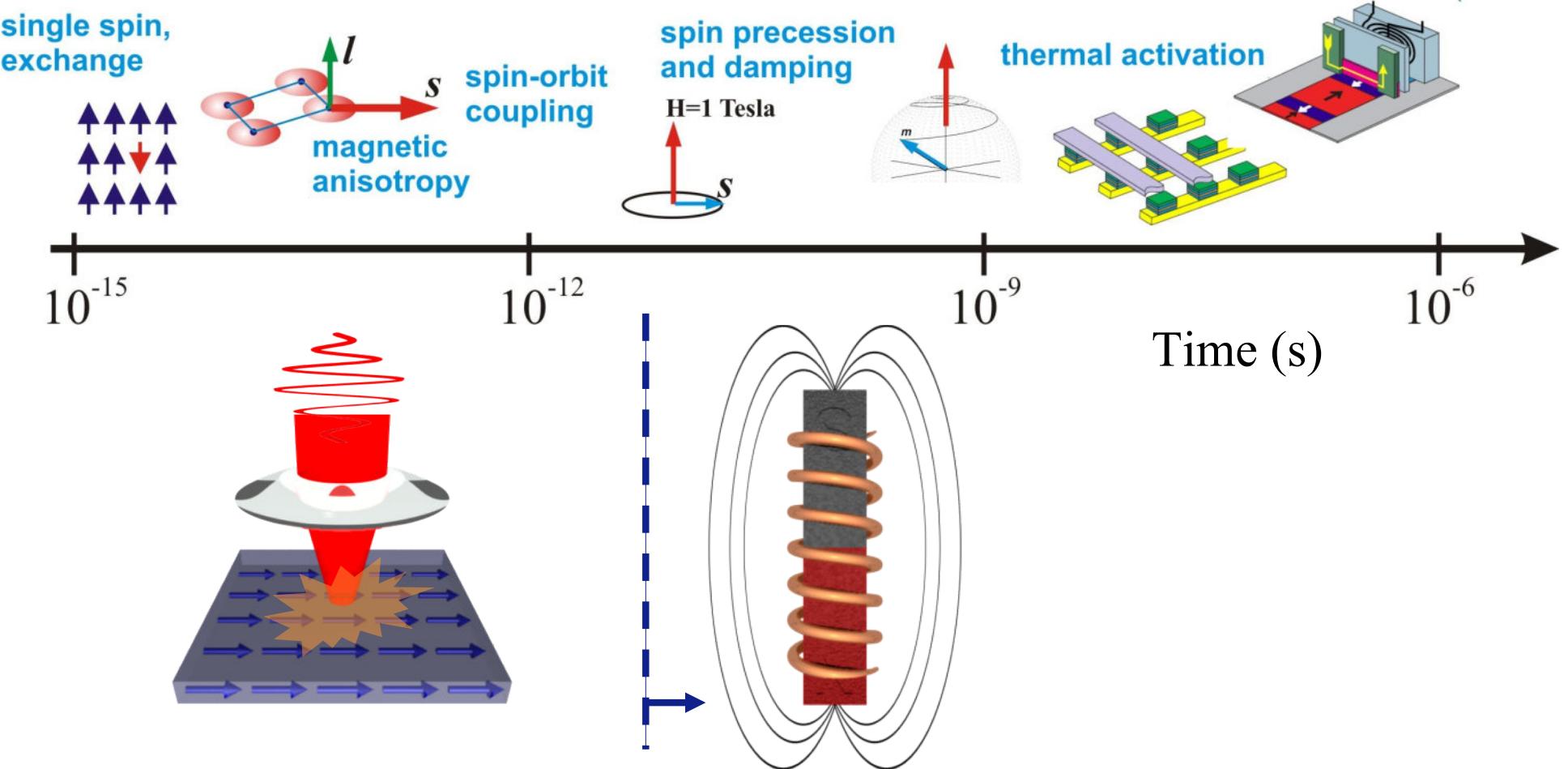
LETTERS

Distinguishing the ultrafast dynamics of spin and orbital moments in solids

C. Boeglin¹, E. Beaurepaire¹, V. Halté¹, V. López-Flores¹, C. Stamm², N. Pontius², H. A. Dürr^{2†} & J.-Y. Bigot¹



Only laser pulses can be fast enough!



**Benchmark: 180° (or 90°) switching
reverse in $<10^{-10}$ s, keep stable for 10^8 s**

Did it switch? Interpretation of the data...

picture by Bert Koopmans, TUe



Part 1: classification of laser-induced effects

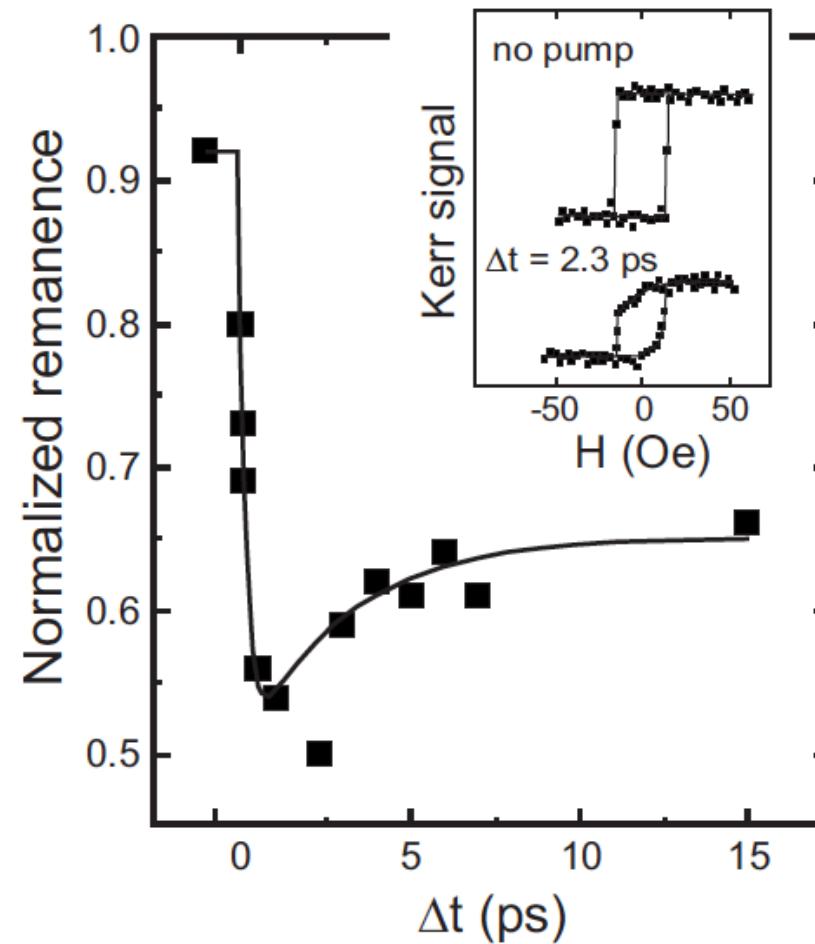
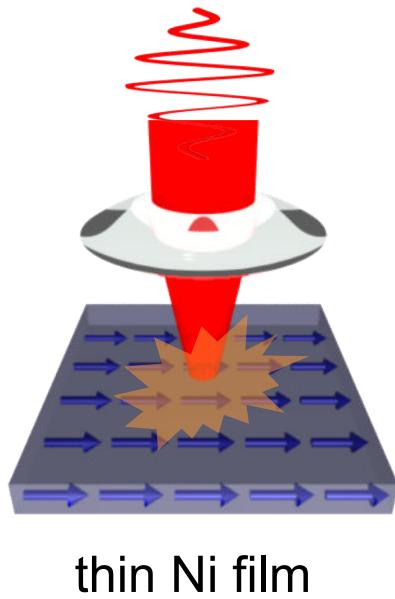
Part 2: the switching as such

Effects of the laser pulse: classification

I. Thermal effects:

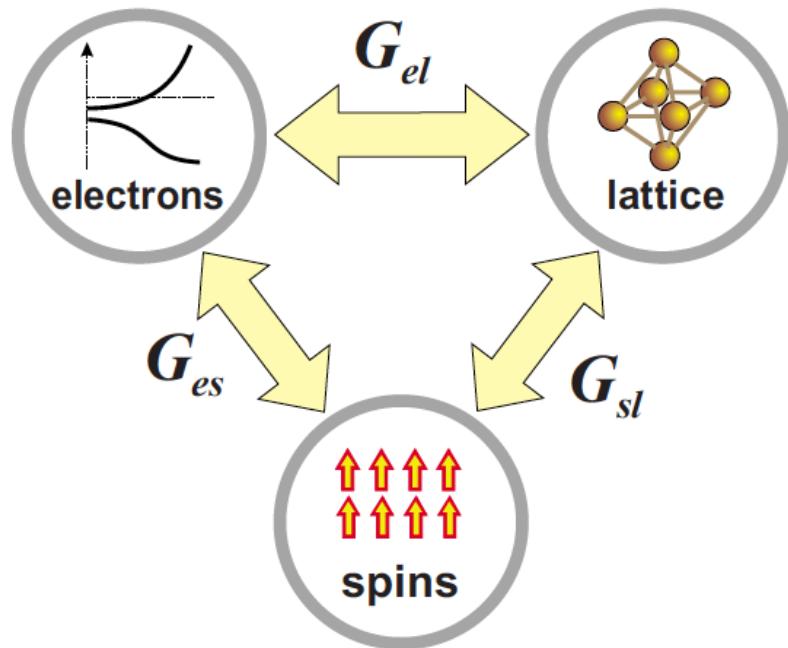
change of M is a result of change of T

Laser-induced collapse of magnetization



Beaurepaire et al, PRL 76, 4250 (1996)

3T model and derivatives

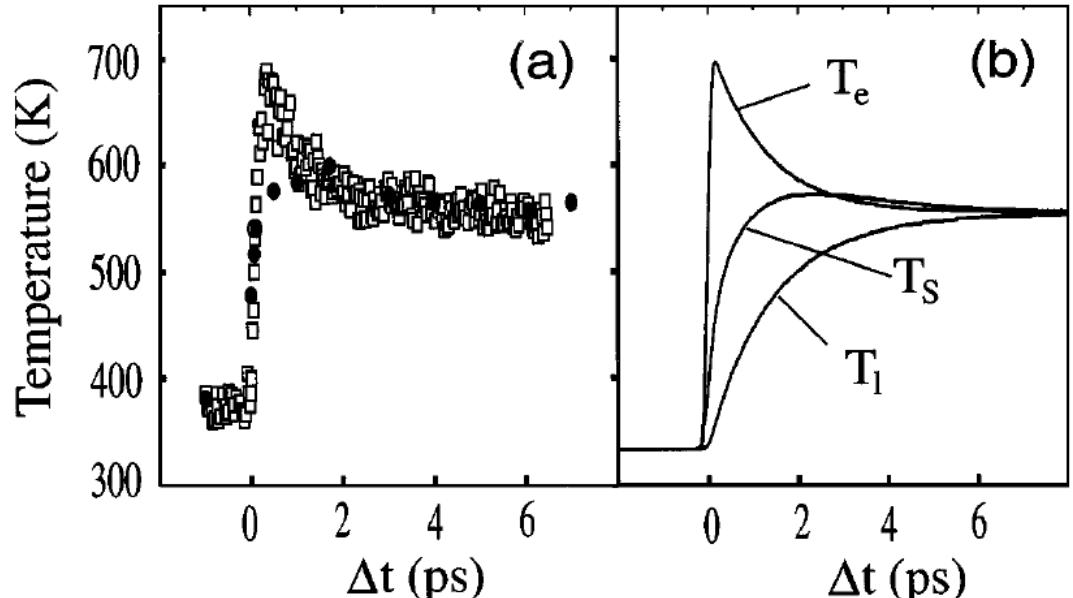


$$C_e d(T_e)/dt = -G_{el}(T_e - T_l) - G_{es}(T_e - T_s) + P(t)$$

$$C_s d(T_s)/dt = -G_{es}(T_s - T_e) - G_{sl}(T_s - T_l),$$

$$C_l d(T_l)/dt = -G_{el}(T_l - T_e) - G_{sl}(T_l - T_s),$$

Beaurepaire et al, PRL 76, 4250 (1996)



$$\frac{dm}{dt} = Rm \frac{T_p}{T_C} \left(1 - m \coth \left(\frac{mT_C}{T_e} \right) \right)$$

$$R = a_{sf} \frac{8g_{ep}kT_C^2}{E_D^2 \mu_{at}}$$

M3TM

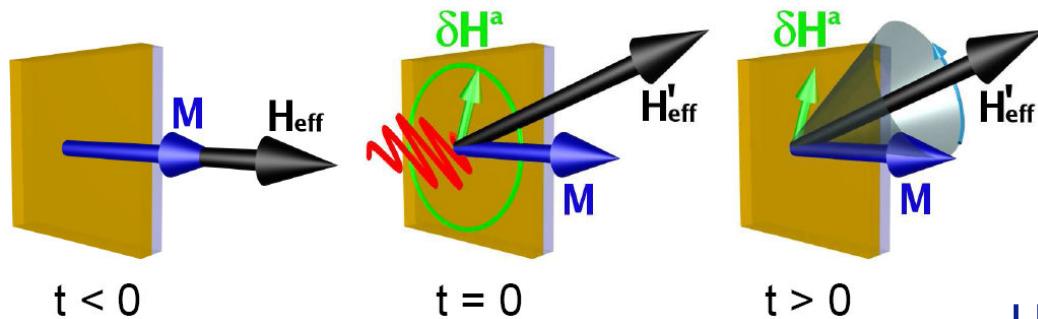
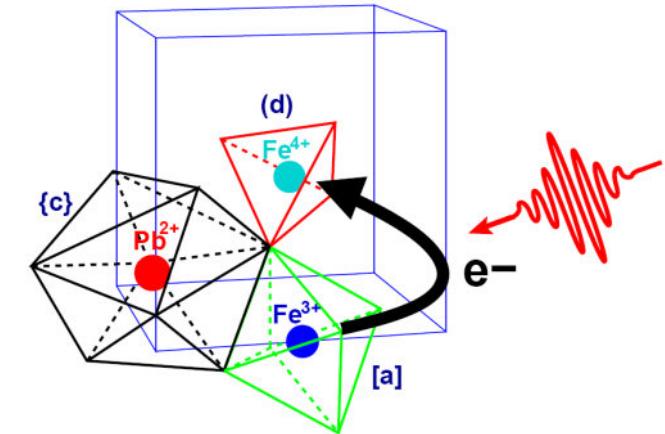
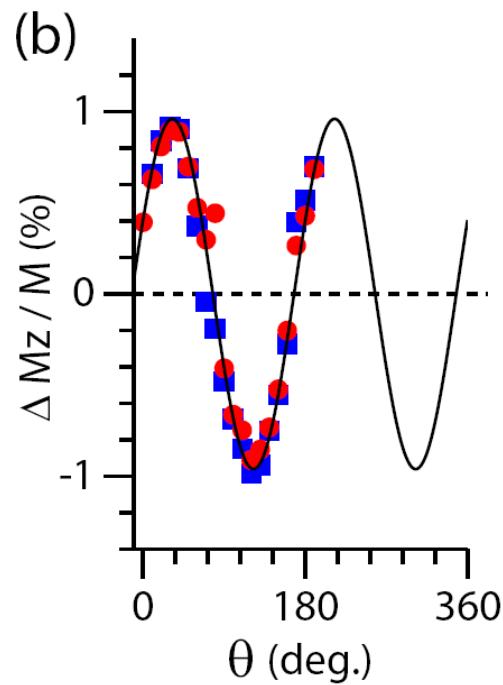
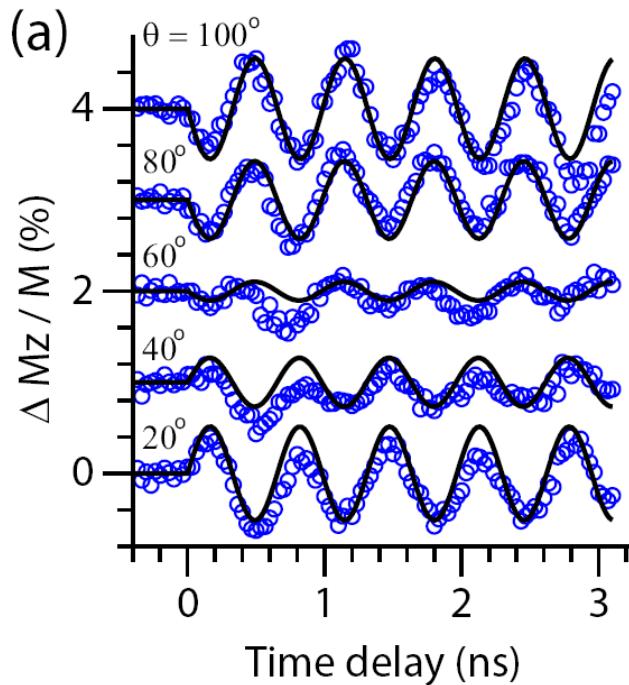
Koopmans et al, Nature Mater. 9, 259 (2010)

$a_{sf} \frac{T_C}{\mu_{at}}$

Effects of the laser pulse: classification

II. Nonthermal photo-magnetic effects: based on photon absorption

Photo-magnetic anisotropy in garnets



pump polarization
dependence!

Hansteen *et al.*, PRL 95, 047402 (2005);
Phys. Rev. B 73, 014421 (2006).

Effects of the laser pulse: classification

III. Nonthermal opto-magnetic effects: do not require absorption

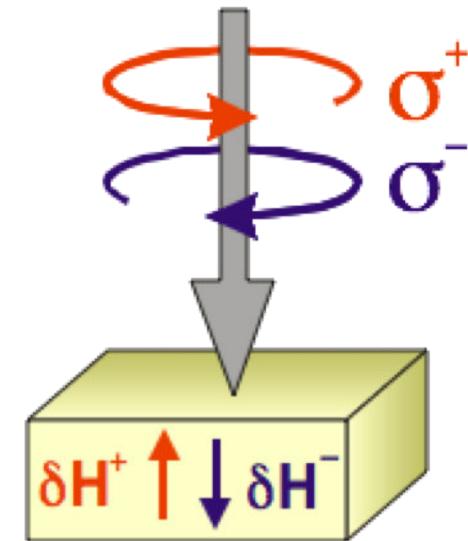
Inverse Faraday effect

$$\Phi = \epsilon \epsilon_0 E(\omega) E^*(\omega)$$

$$H(0) = -\frac{1}{\mu_0} \frac{\partial \Phi}{\partial M(0)} = -\frac{\epsilon_0}{\mu_0} E(\omega) E^*(\omega) \frac{\partial \epsilon}{\partial M}$$

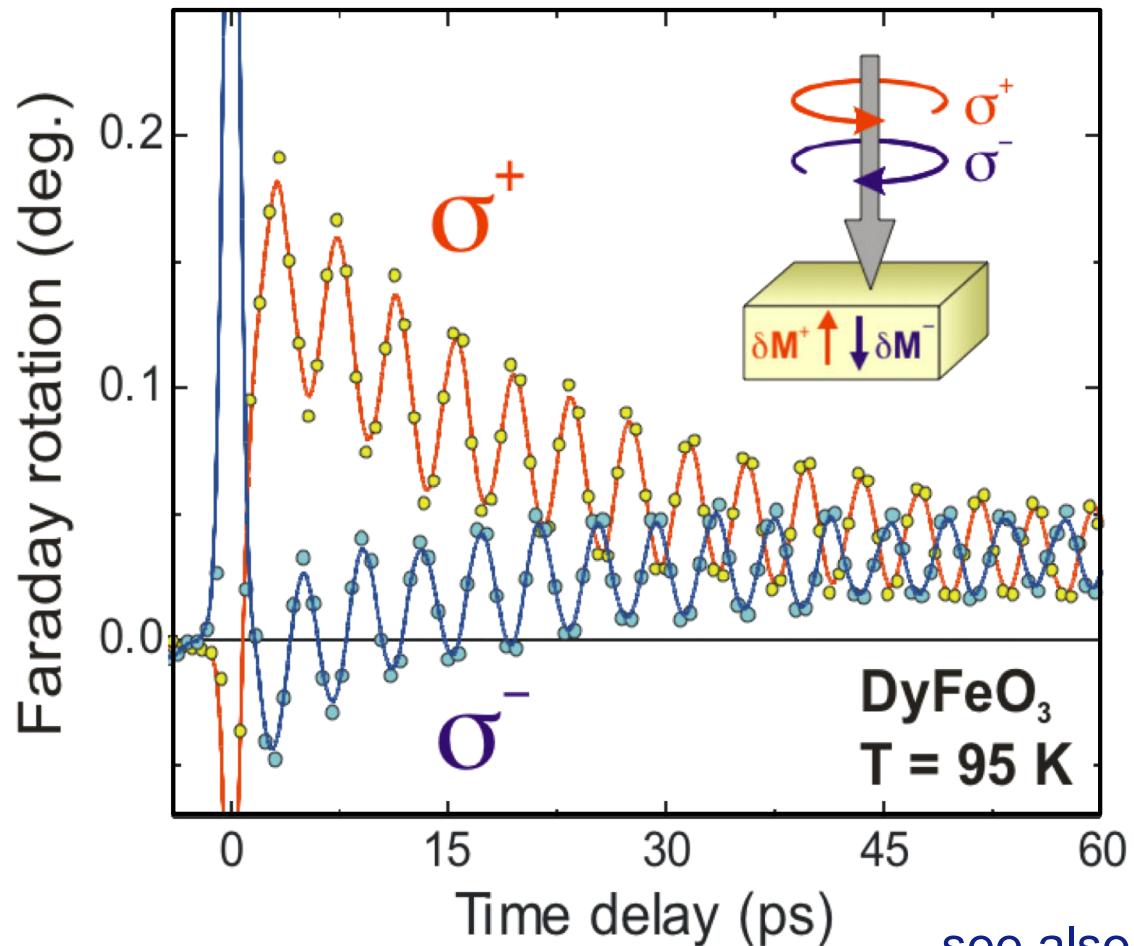
$$\hat{\epsilon} = \begin{pmatrix} \epsilon_{xx} & -i\alpha M & 0 \\ +i\alpha M & \epsilon_{yy} & 0 \\ 0 & 0 & \epsilon_{zz} + O(M^2) \end{pmatrix}$$

$$\boxed{H(0) = \frac{\epsilon_0}{\mu_0} \alpha [E(\omega) \times E^*(\omega)]}$$



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

Inverse Faraday effect to excite spin dynamics



Kimel et al., Nature **435**, 655 (2005)

equivalent to a 100 fs
magnetic field pulse of
some 0.5–1 Tesla!

again, the
dependence on
pump polarization!

see also:

Hansteen et al., PRB **73**, 014421 (2006);
Kalashnikova et al., PRL **99**, 167205 (2007)

Effects of the laser pulse: summary



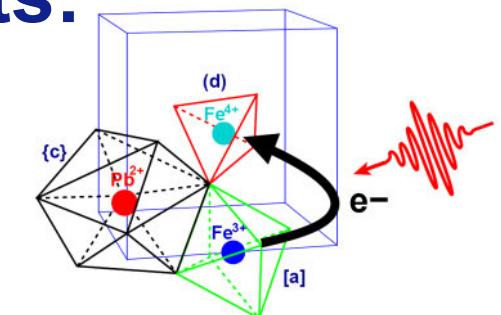
I. Thermal effects:

change of M is a result of change of T

II. Nonthermal photo-magnetic effects:

based on photon absorption

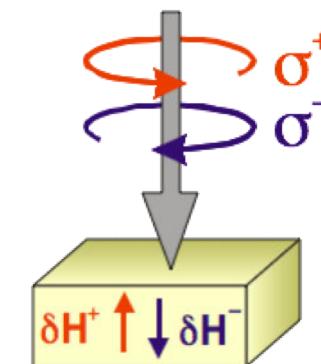
displacive effect



III. Nonthermal opto-magnetic effects:

do not require absorption

impulsive effect



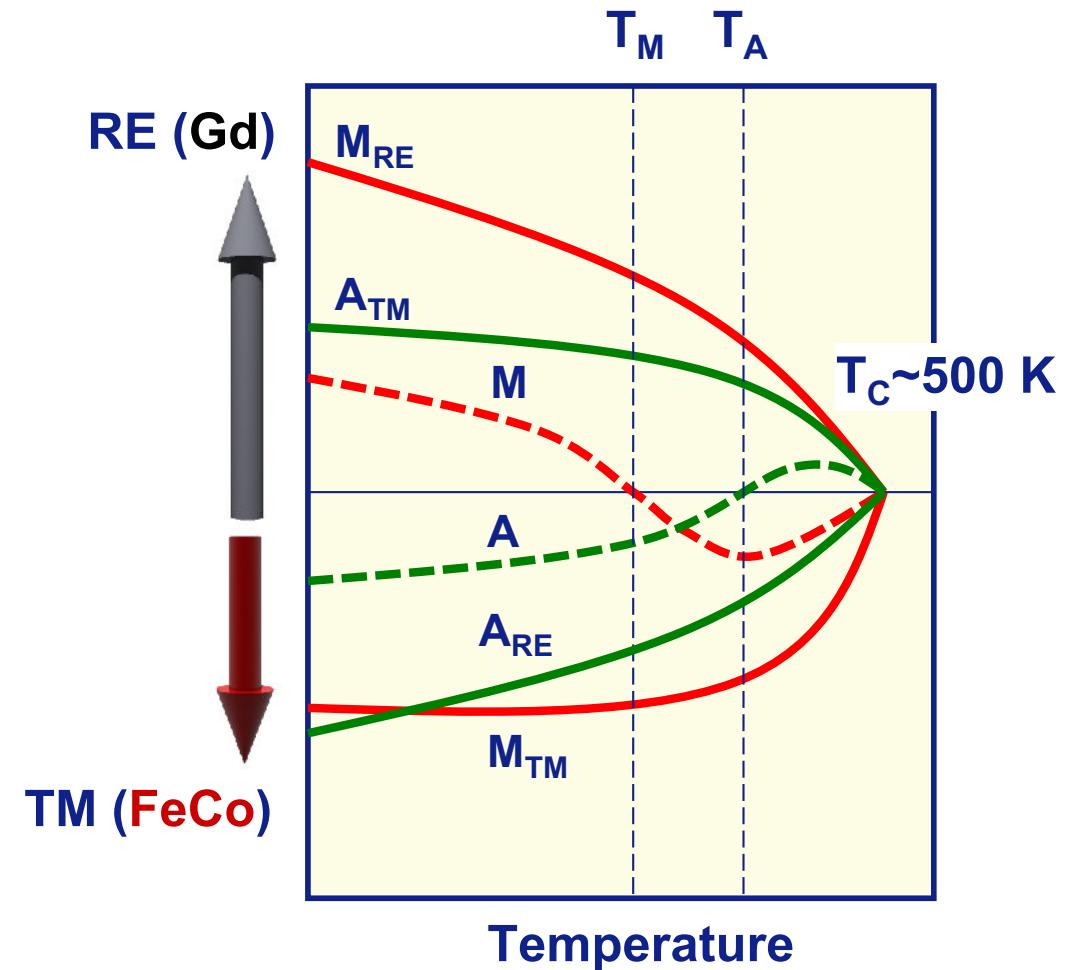
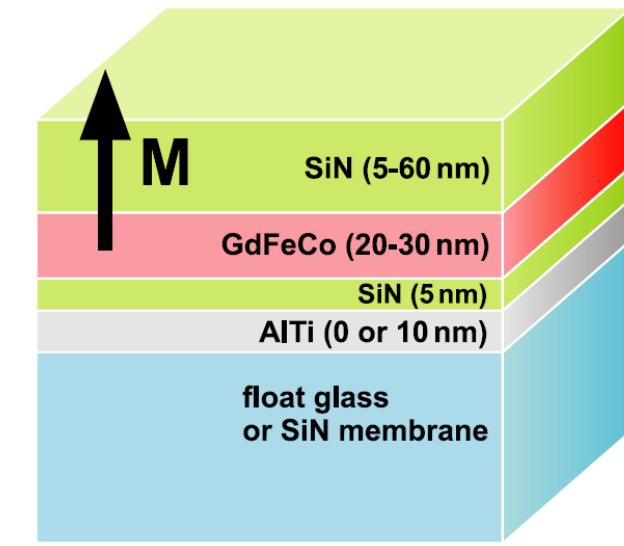
Part 1: classification of laser-induced effects

Part 2: the switching as such

1. Switching based on thermal effects



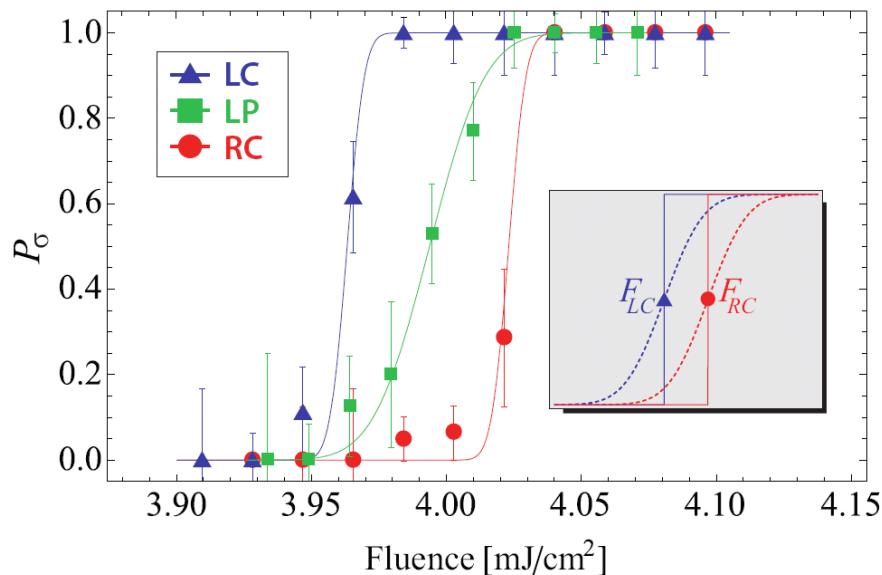
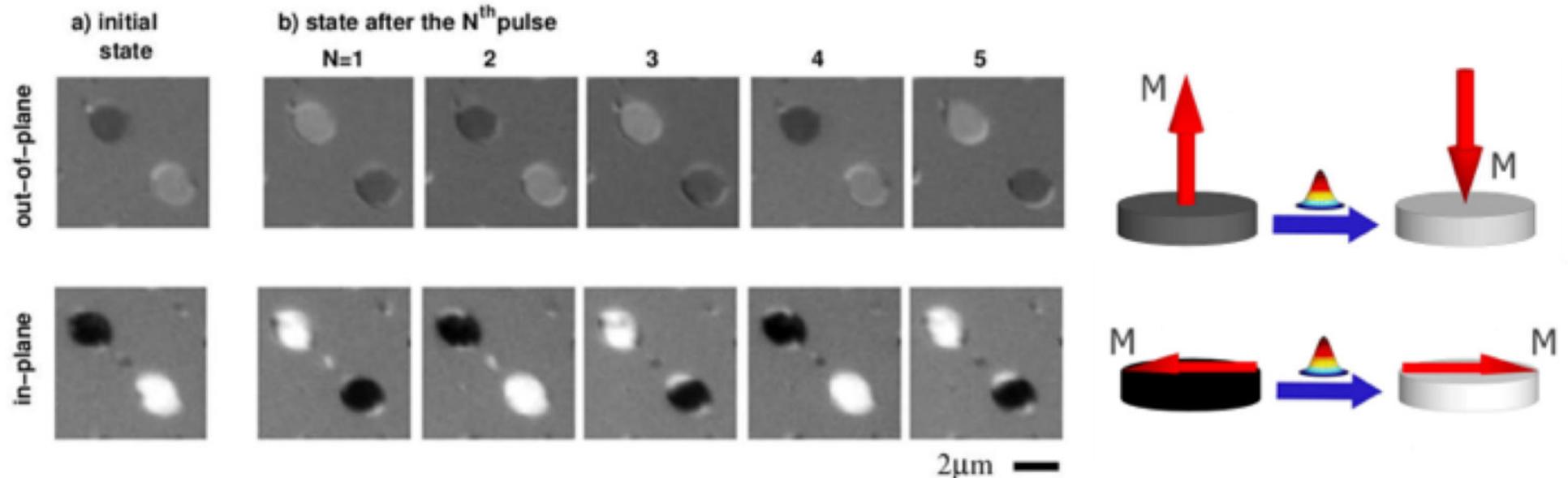
Ferrimagnetic RE-TM alloys & multilayers (e.g. GdFeCo)



$$g_{\text{Gd}} < g_{\text{FeCo}}$$

Toggle switching in GdFeCo

Each next image - a single unpolarized laser pulse

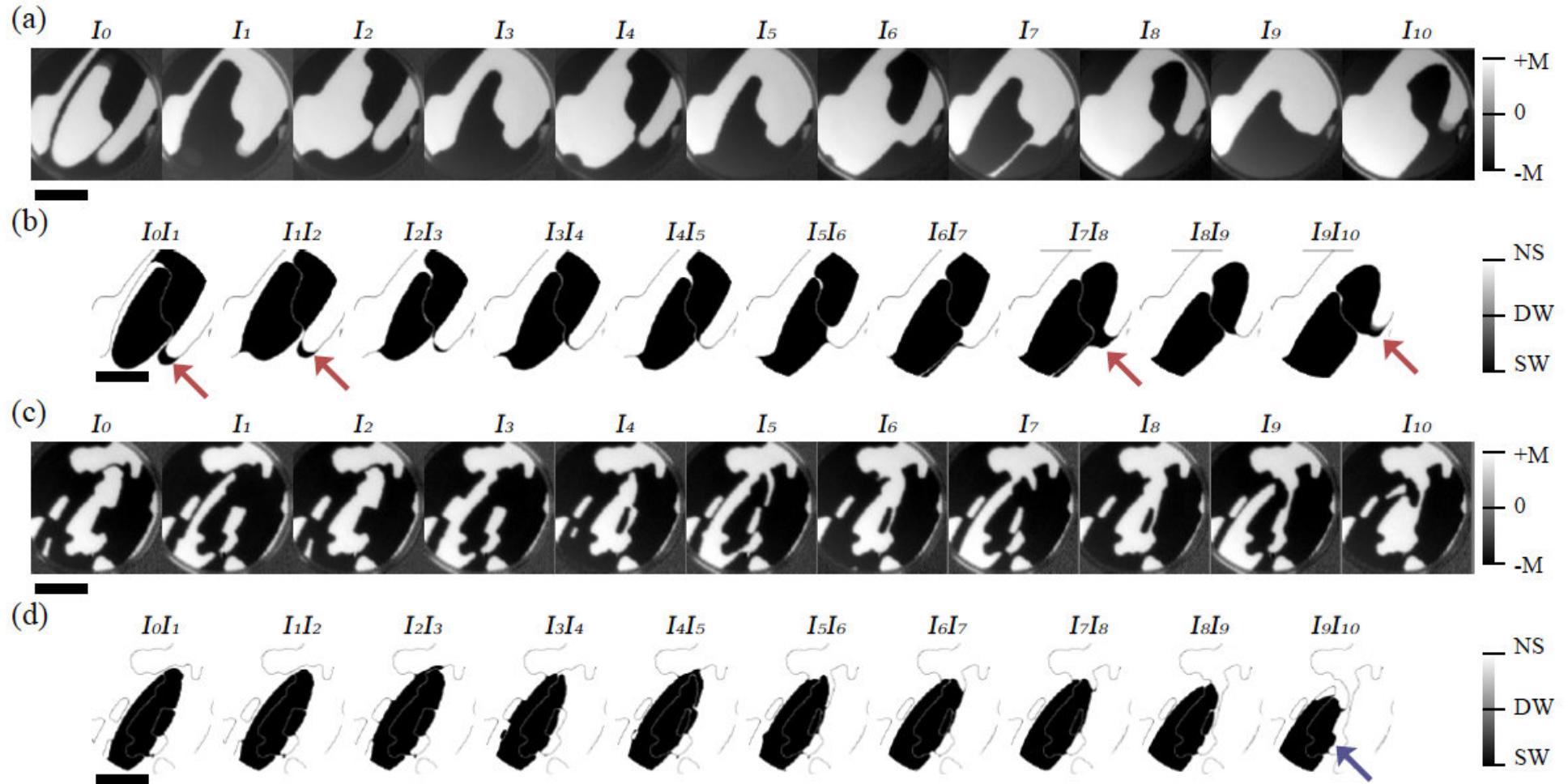


Ostler et al., Nature Commun. 3, 666 (2012)

amount of energy absorbed in the sample per pulse stays constant

Khorsand et al,
Phys. Rev. Lett. 108, 127205 (2012)

Switching of a multi-domains structure: reproducibility

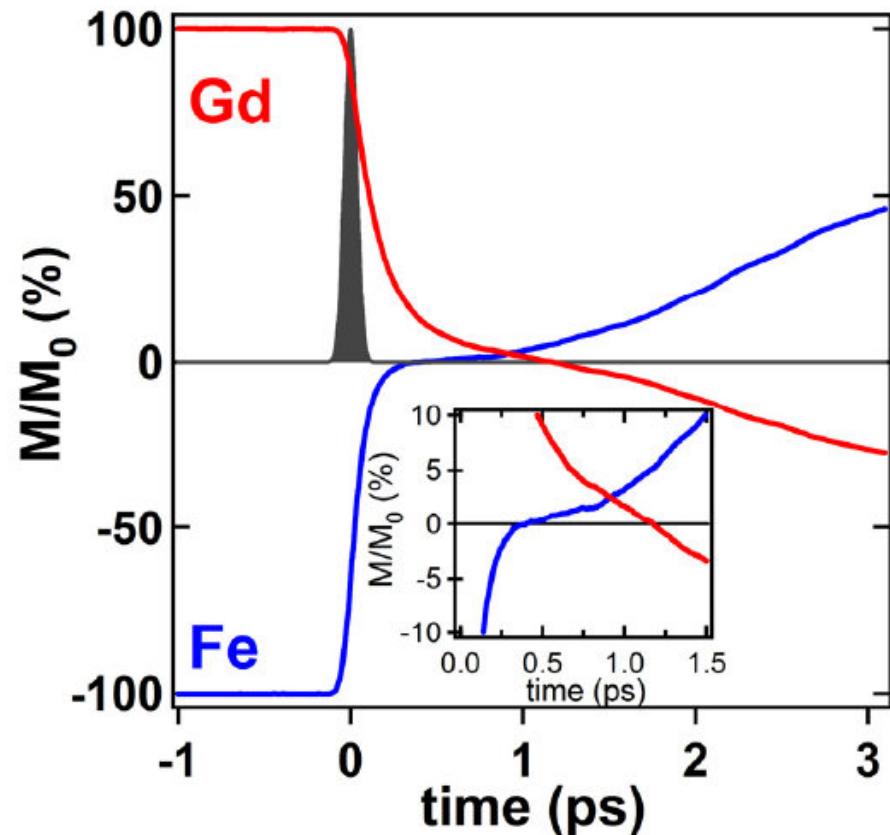
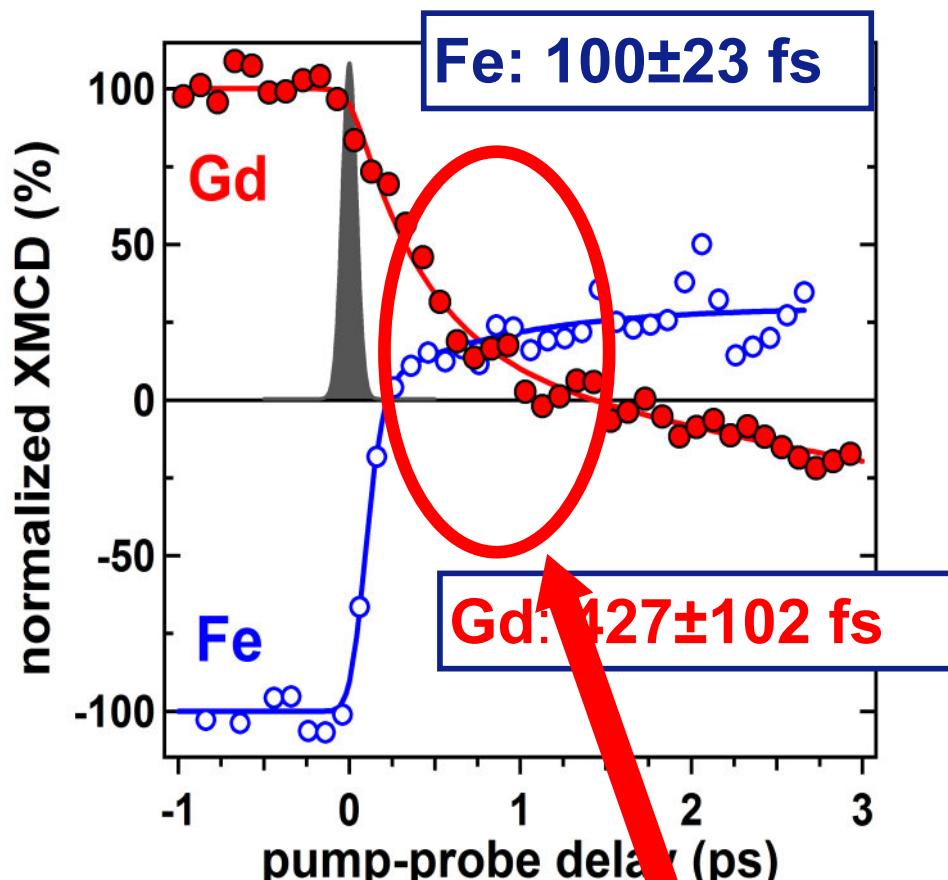


no domain wall motion, just reversal of the whole pattern

Le Guyader et al., Phys. Rev. B **93**, 134402 (2016)

Dynamics of sublattices

Radu et al., Nature 472, 205 (2011)



ferri-magnet turns ferro!

Longitudinal relaxation in multi-sublattice magnets

Mentink et al., PRL **108**, 057202 (2012);

$$\frac{dS_1}{dt} = \lambda_e (H_1 - H_2) + \lambda_1 H_1$$
$$\frac{dS_2}{dt} = -\underbrace{\lambda_e (H_1 - H_2)}_{\text{exchange}} + \underbrace{\lambda_2 H_2}_{\text{relativistic (usual damping)}}$$

where $S_i = M_i/\gamma_i$

and $H_i = -\delta W/\delta S_i$

$$\lambda_e(T) = \lambda_e(J_{12}(T)) \quad \lambda_i(T) \sim T/T_C$$

conservation S_{tot}

$$\frac{dS_1}{dt} = -\frac{dS_2}{dt}$$

Bloch relaxation

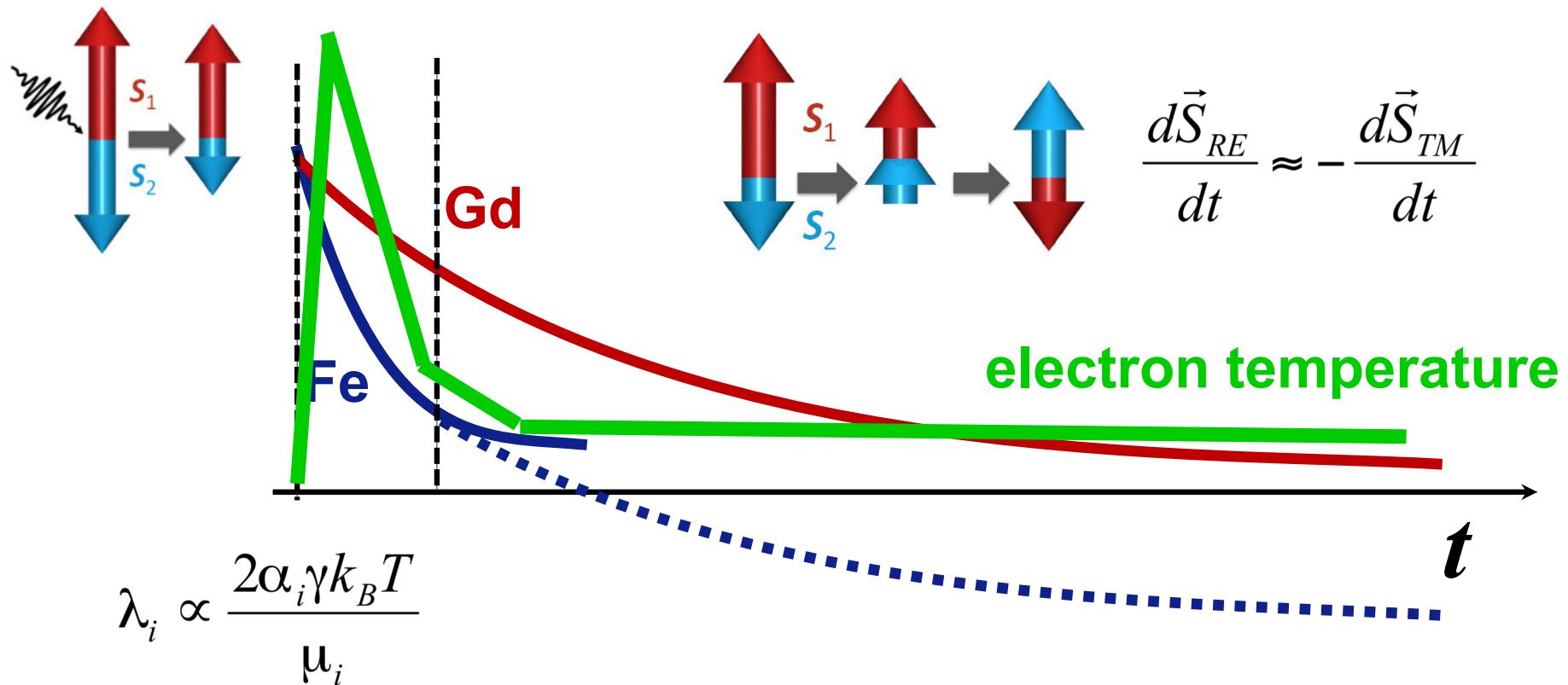
$$dS_i/dt = -S_i/\tau_i$$

$$\tau_i = \chi_i/\lambda_i$$

$$\lambda_i \propto \frac{2\alpha_i \gamma k_B T}{\mu_i}$$

Crossover from temperature- to exchange-dominated

$\vec{S}_{RE}, \vec{S}_{TM}$ independent

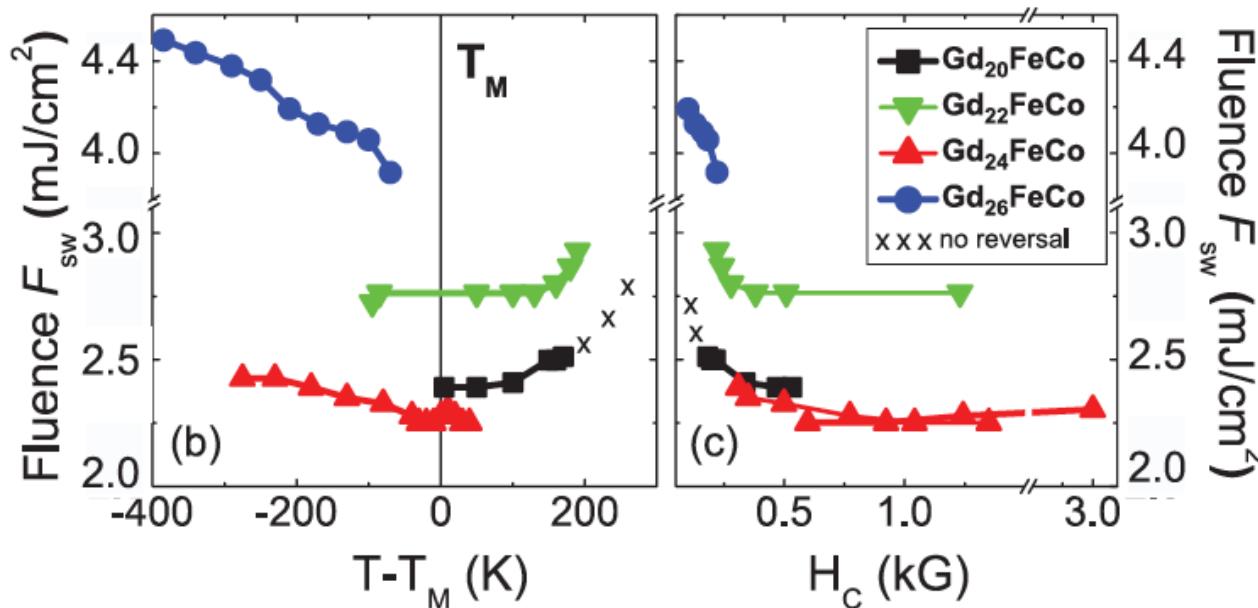


derived in Mentink et al., PRL 108, 057202 (2012);

see Kirilyuk et al., Rep. Prog. Phys. 76, 026501 (2013) for summary

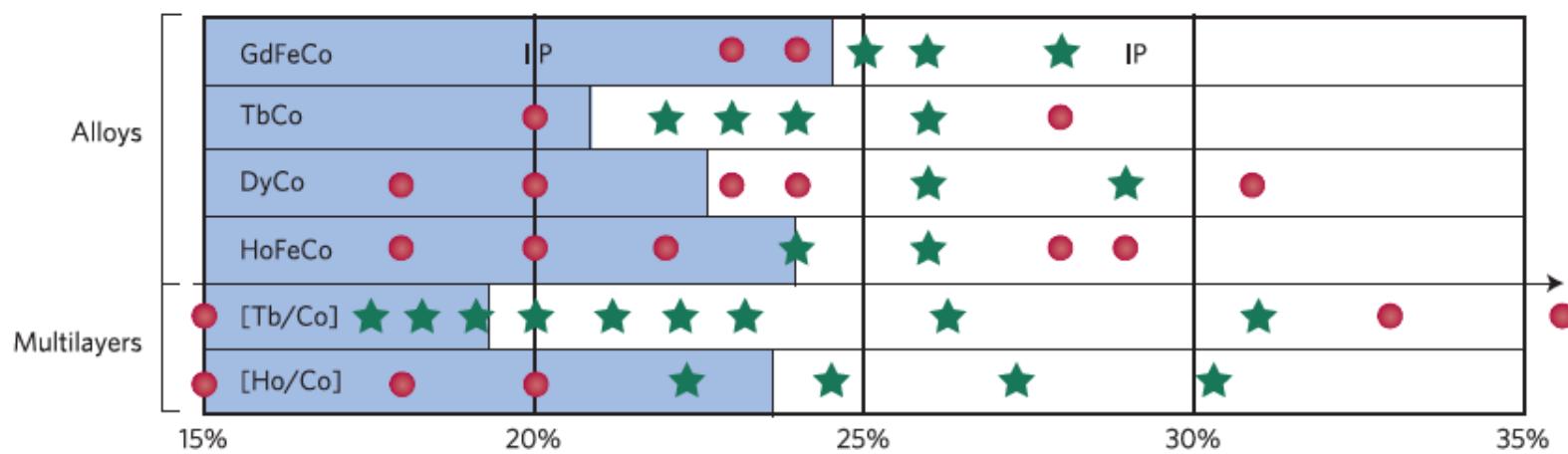
The range of switching

Vahaplar et al, PRB 85, 104402 (2012)



It works in
the broad vicinity
of the compensation
temperature

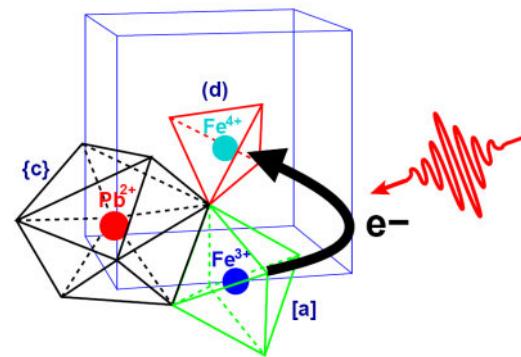
Mangin et al, Nature Materials 13, 286 (2014)



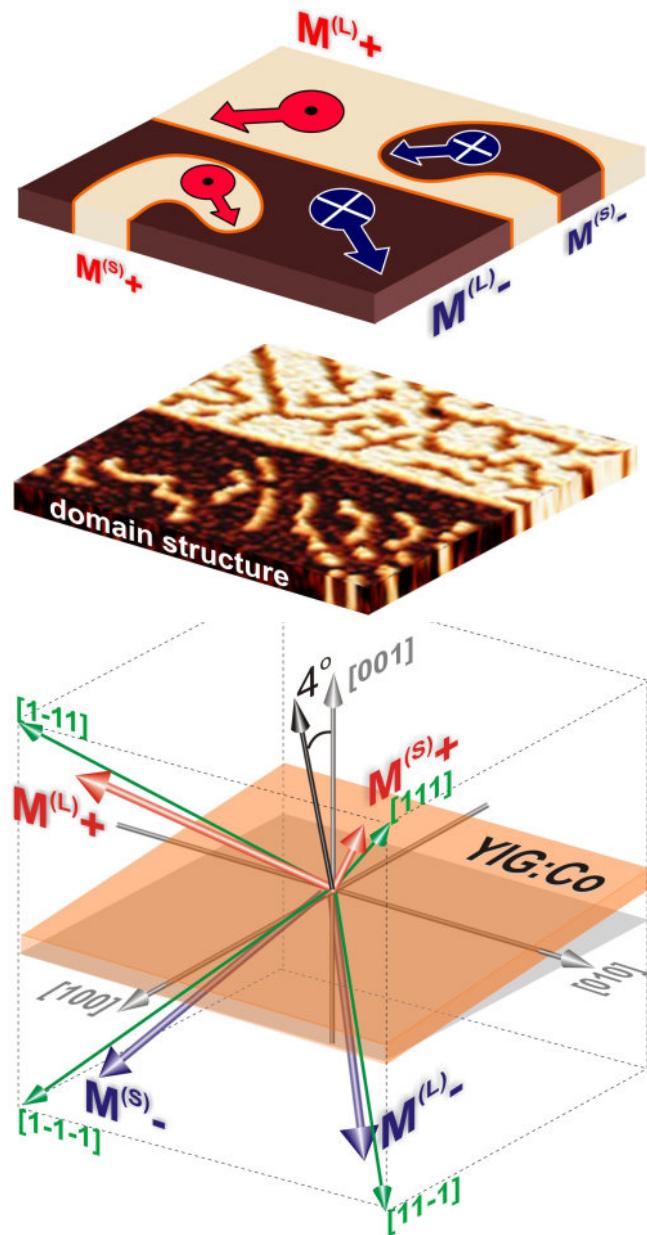
Mechanism: thermal, fast sublattice-selective demagnetization + exchange-driven reversal

Time-scale: ~1 ps reversal, 30-1000 ps recovery

2. Photo-magnetic switching in dielectrics



Co-substituted YIG film



$\text{Y}_2\text{CaFe}_{3.9}\text{Co}_{\textcolor{red}{0.1}}\text{GeO}_{12}$ on GGG (001)

thickness $d=7.5 \mu\text{m}$ (grown by LPE)

magnetic anisotropy:

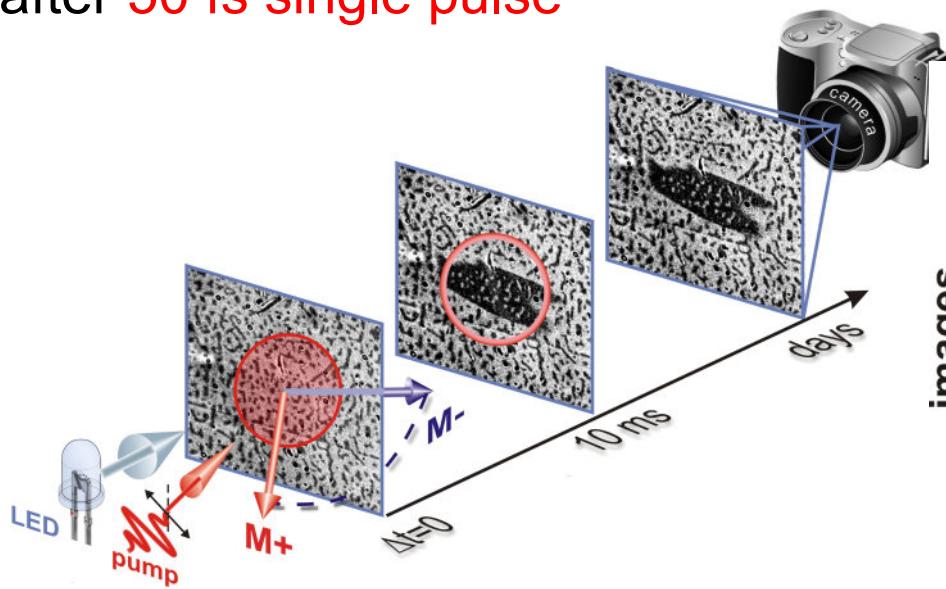
$$K_1 = -10^4 \text{ erg/cm}^3$$

$$K_U = 10^3 \text{ erg/cm}^3$$

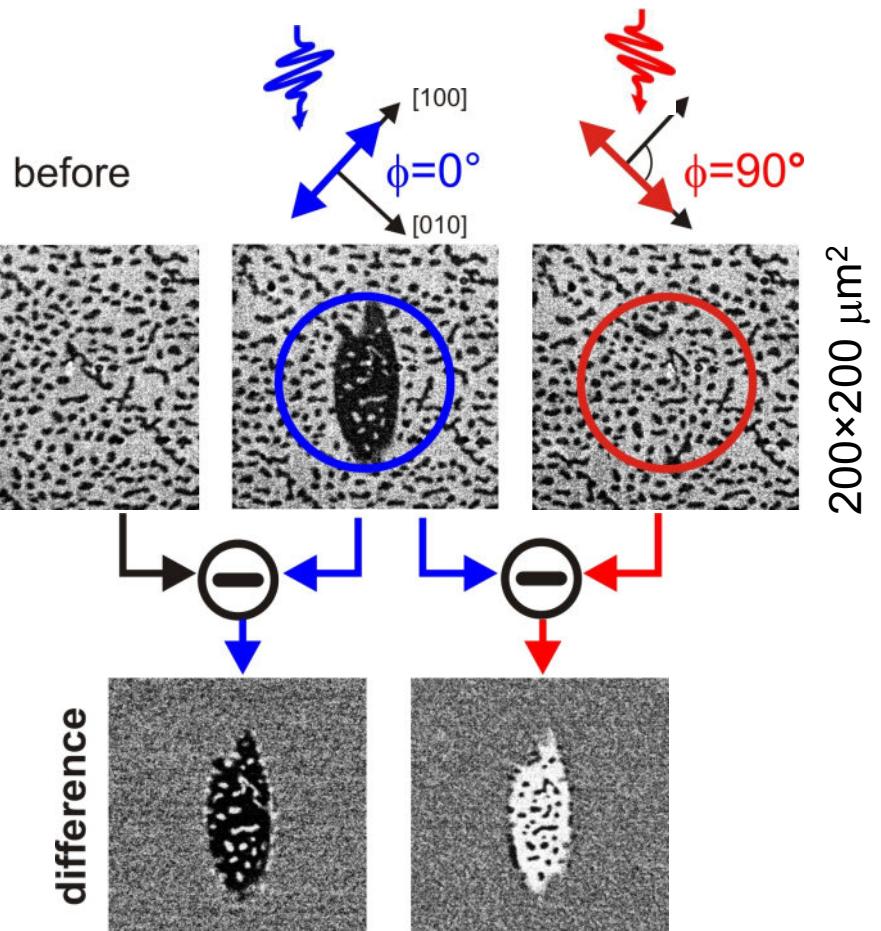
domain structure: metastable states

Single-pulse switching

Image of the final state >10 ms
after **50 fs single pulse**



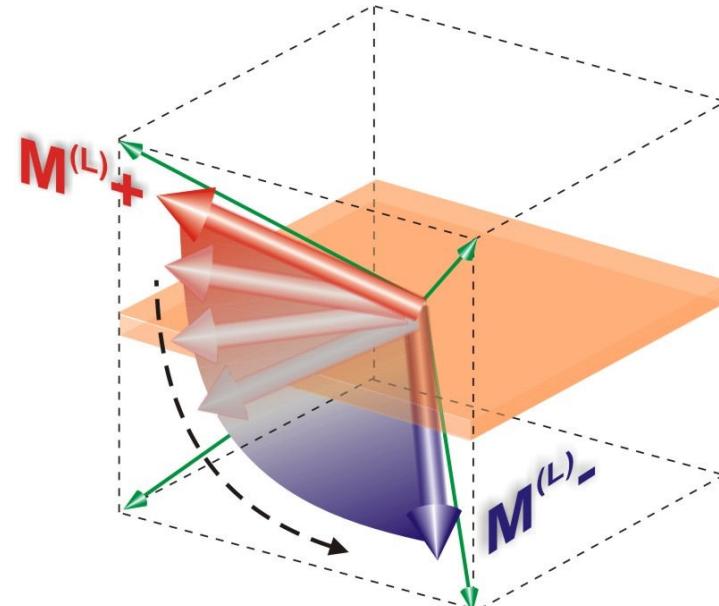
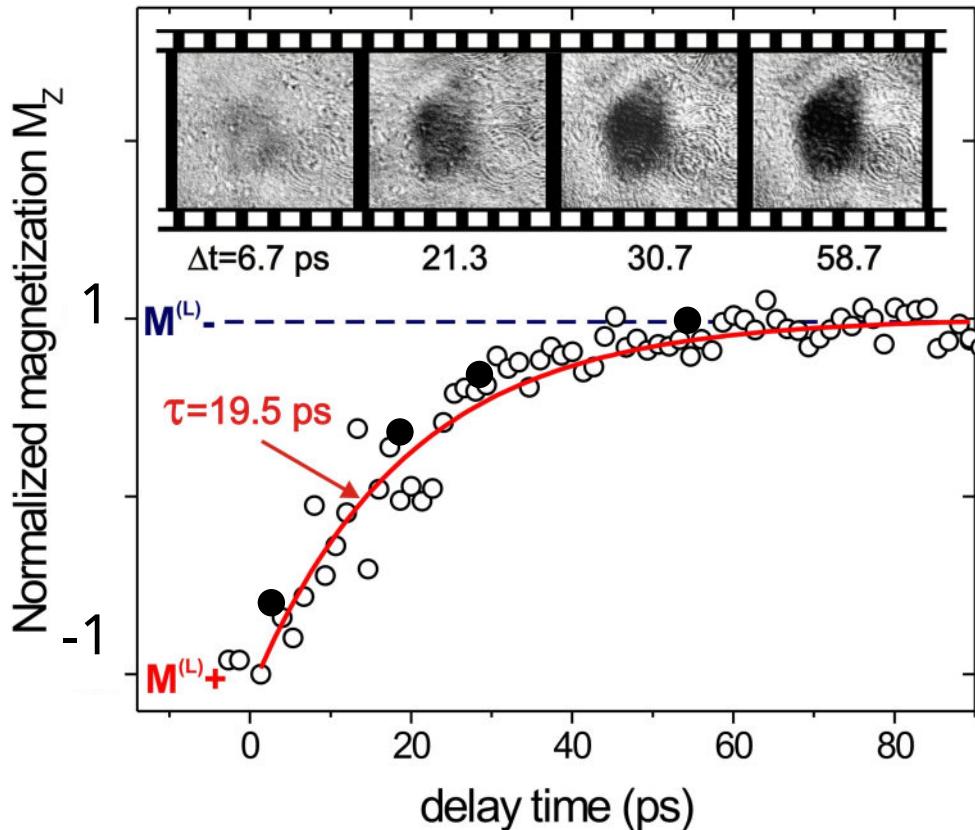
A. Stupakiewicz et al., Nature **542**, 71 (2017)



- repeatable
- zero applied field
- room temperature
- tiny absorbed energy (<1 K)

Time resolved observation of switching

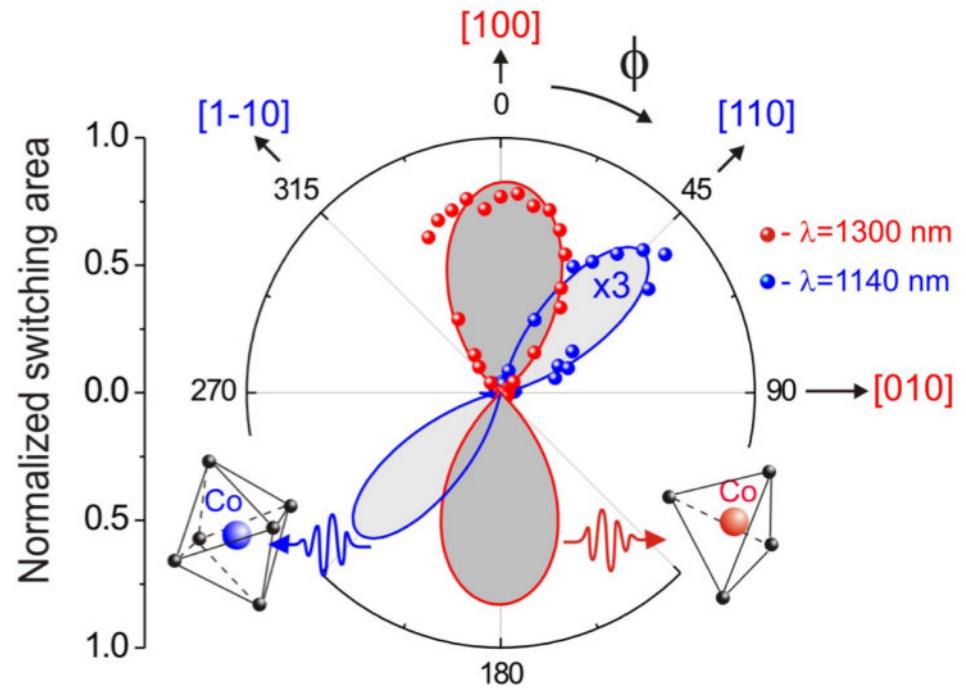
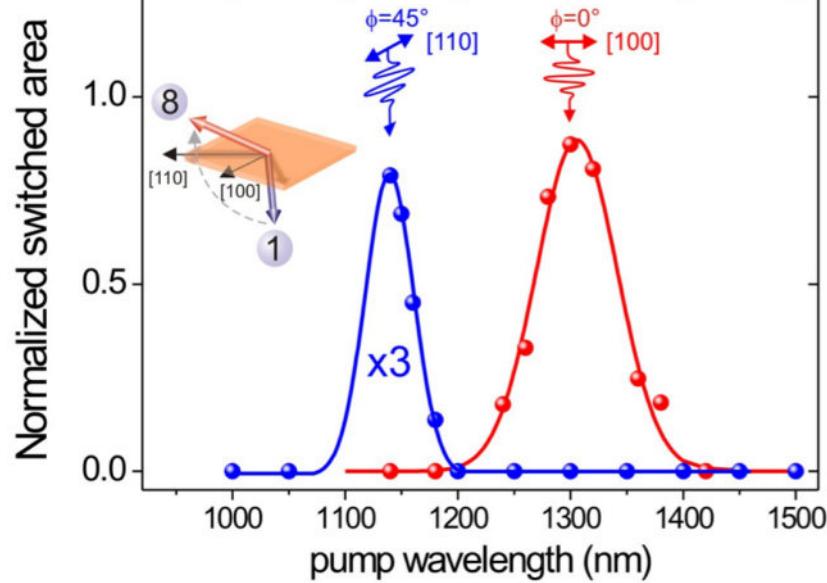
switched after first quarter-period



precessional switching!

A. Stupakiewicz et al., Nature 542, 71 (2017)

Precise atomic-scale control of anisotropy?

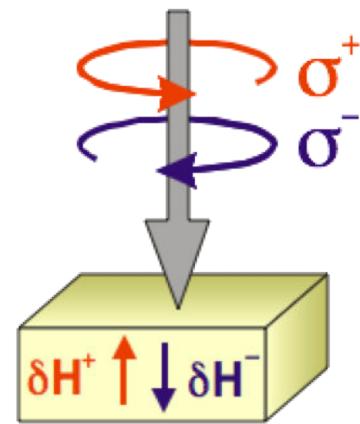


A. Stupakiewicz et al., to be published

**Mechanism: photo-magnetic anisotropy driving
the precessional reversal (nonthermal I)**

**Time-scale: precessional motion in the
anisotropy field: 20-60 ps**

3. Opto-magnetic effect (but not only...)



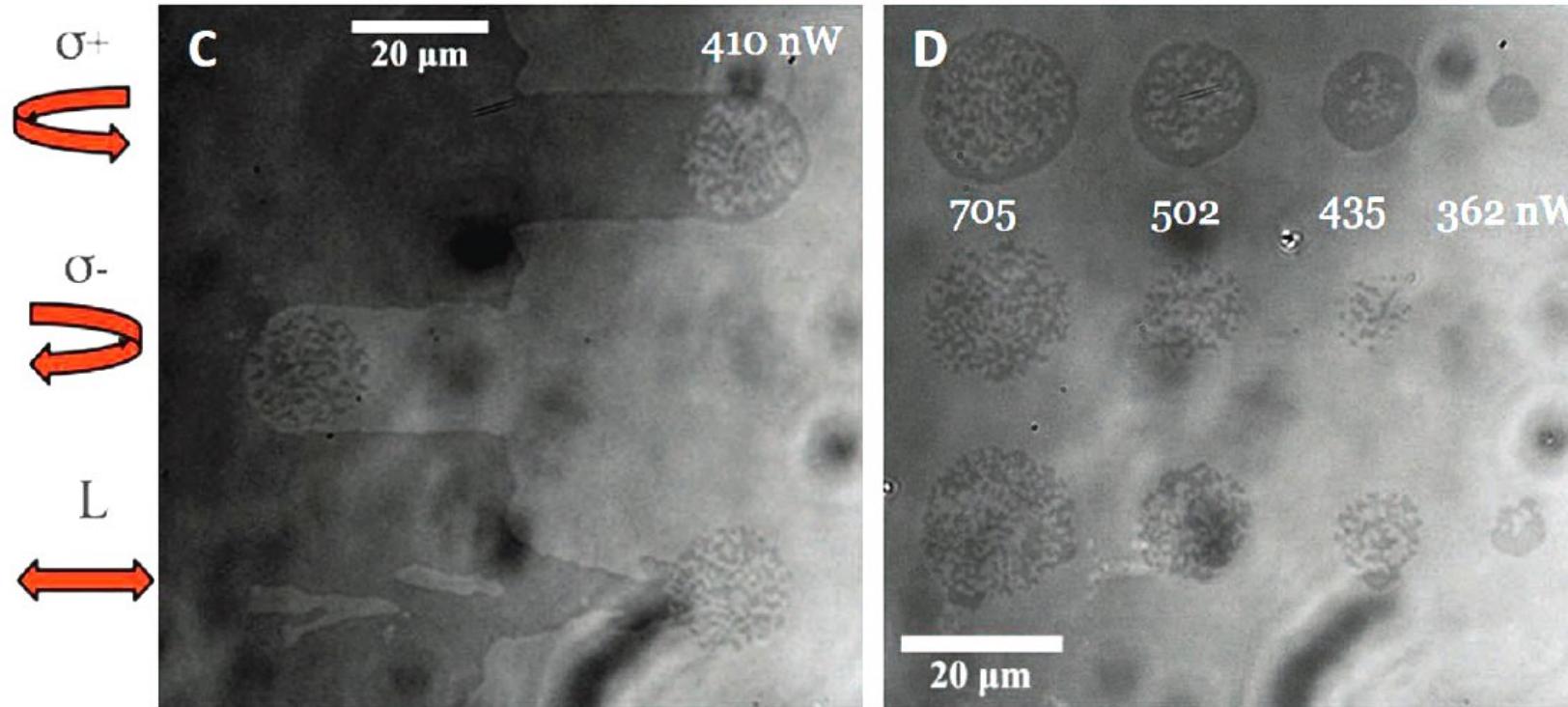
More universal?

All-optical control of ferromagnetic thin films and nanostructures

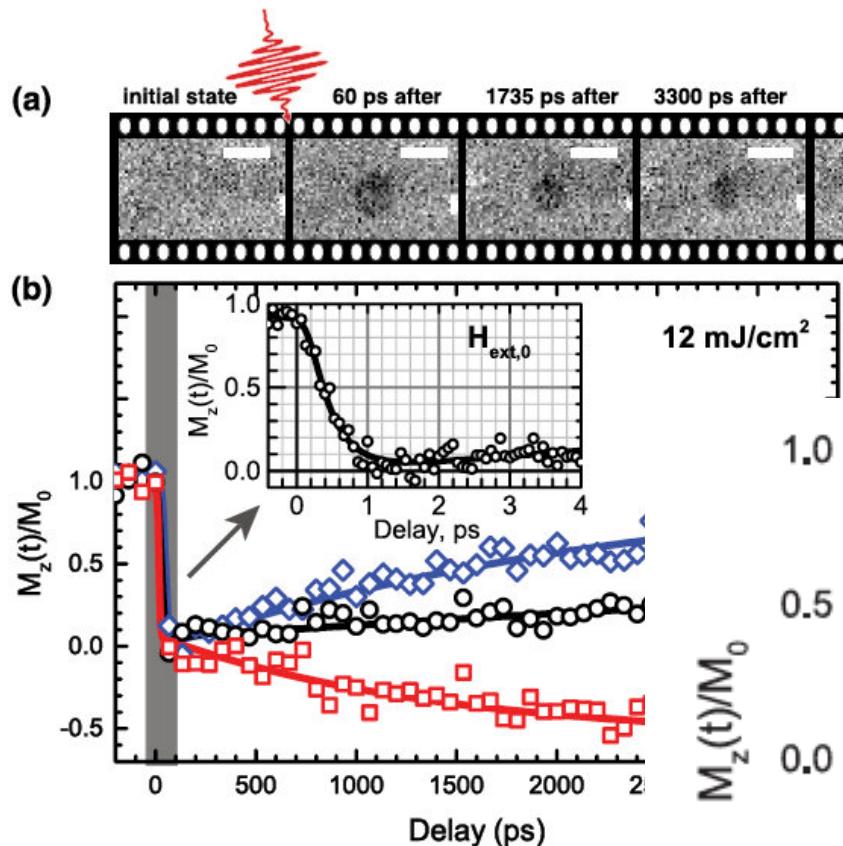
Co/Pt, FePt

C-H. Lambert,^{1,2} S. Mangin,^{1,2*} B. S. D. Ch. S. Varaprasad,³ Y. K. Takahashi,³
M. Hehn,² M. Cinchetti,⁴ G. Malinowski,² K. Hono,³ Y. Fainman,⁵
M. Aeschlimann,⁴ E. E. Fullerton^{1,5*}

Science 345, 1337 (2014)

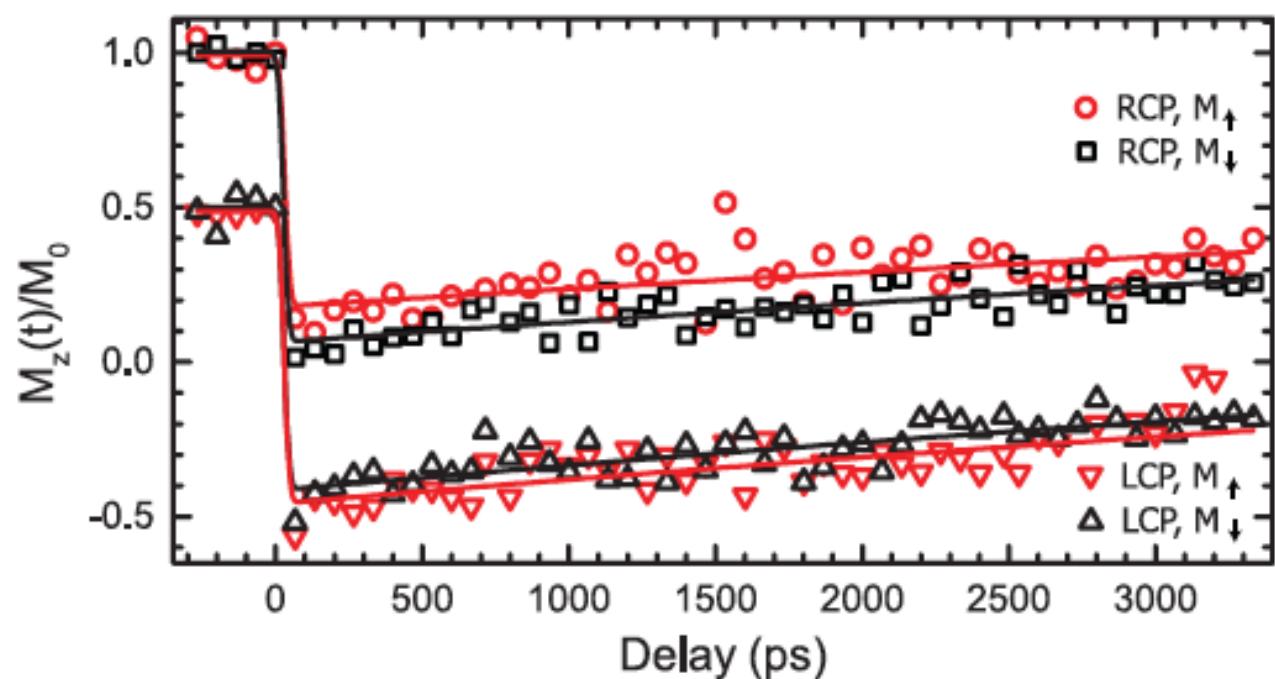


Helicity-effect in the ultrafast demagnetization



[Co(0.4 nm)/Pt(0.7 nm)]₃ multilayers

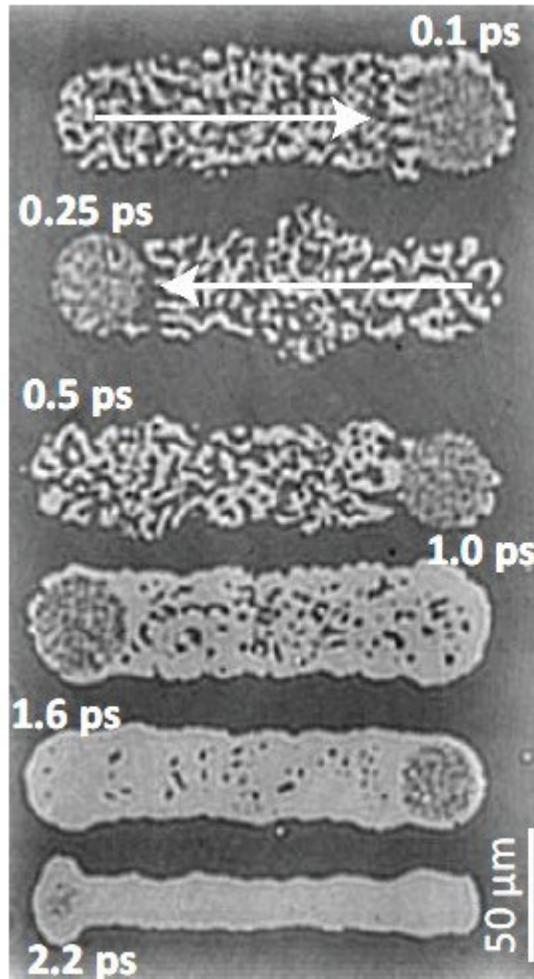
magnetic circular dichroism?



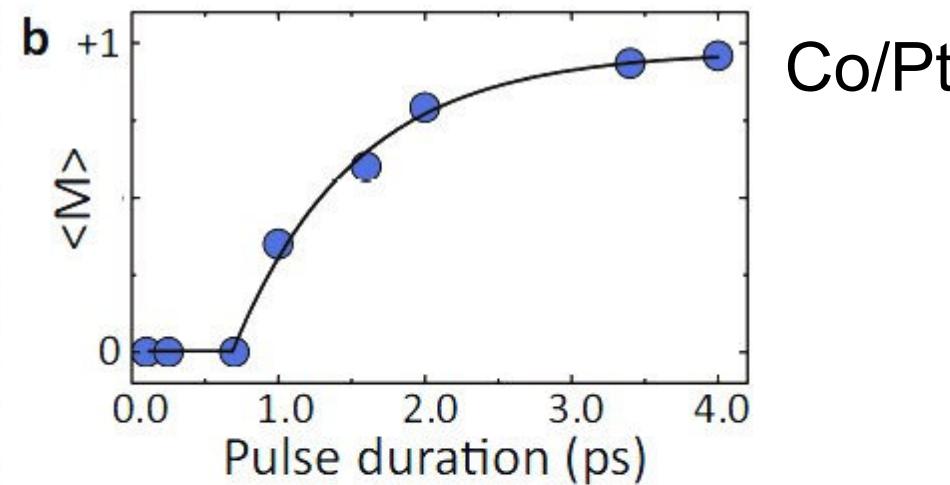
Tsema et al, Appl. Phys. Lett. **109**, 072405 (2016)

Pulse width dependence

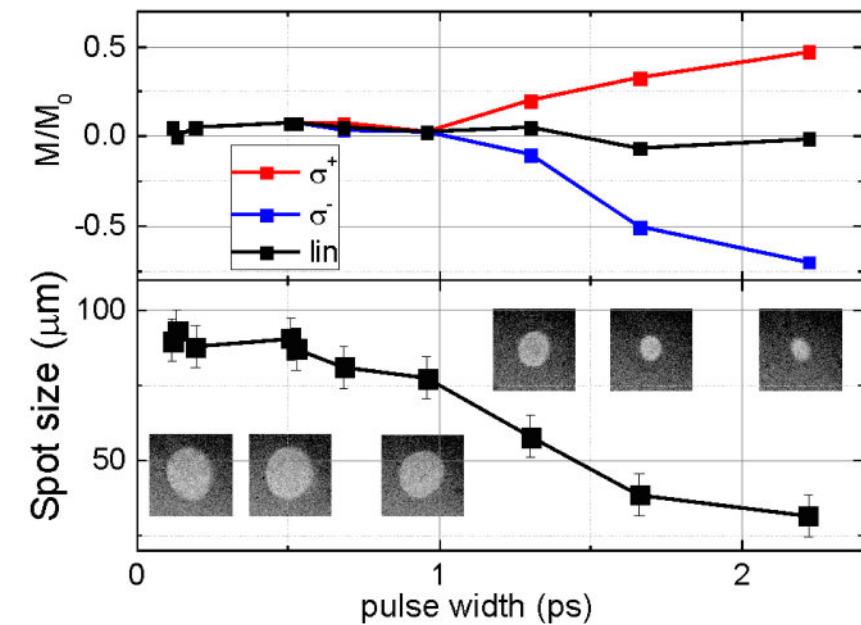
R. Medapalli et al, PRB **96**, 224421 (2017)



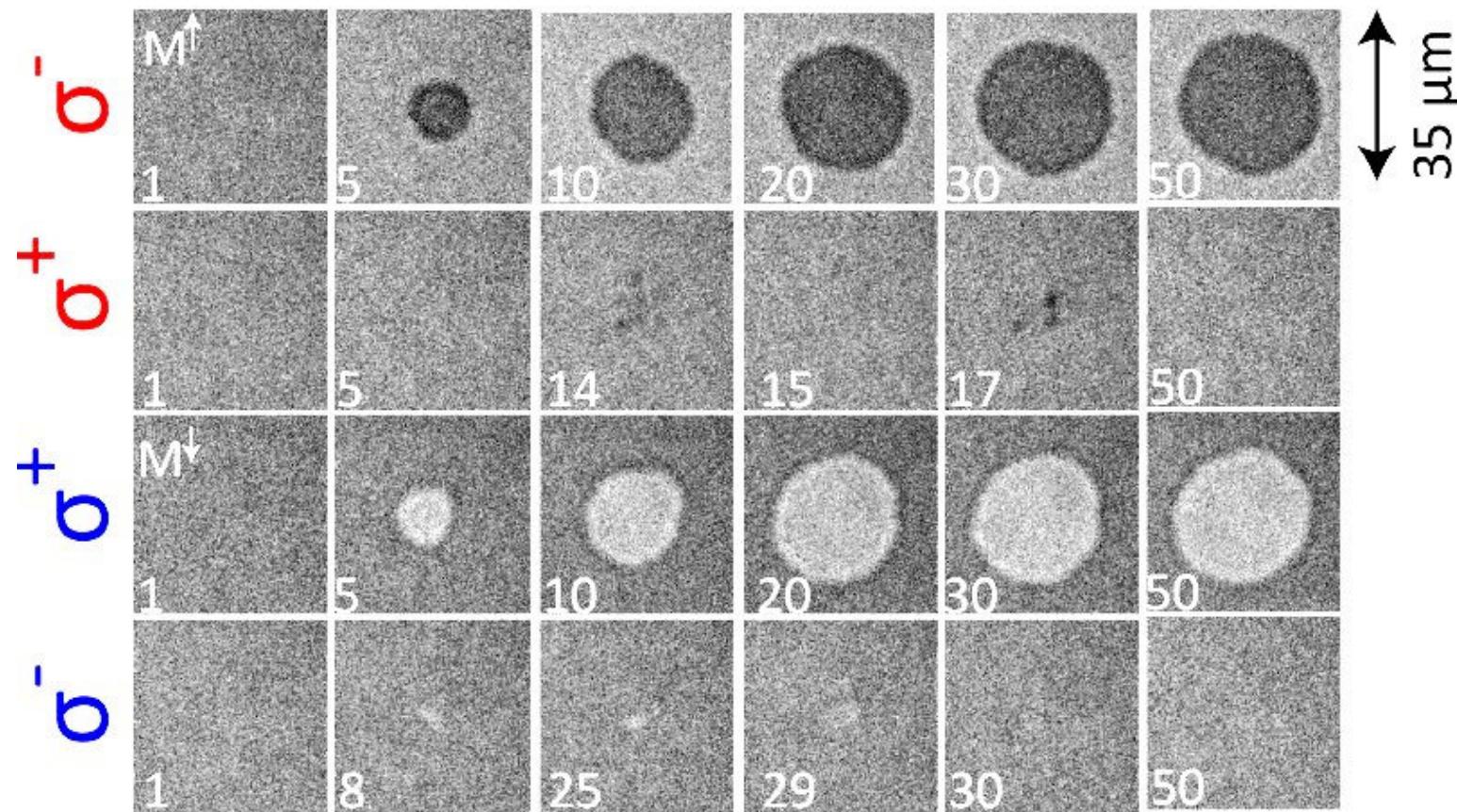
in collaboration with
R. Medapalli and E. Fullerton



Co/Pt
Co/Pd
in collaboration with O. Hellwig, HGST

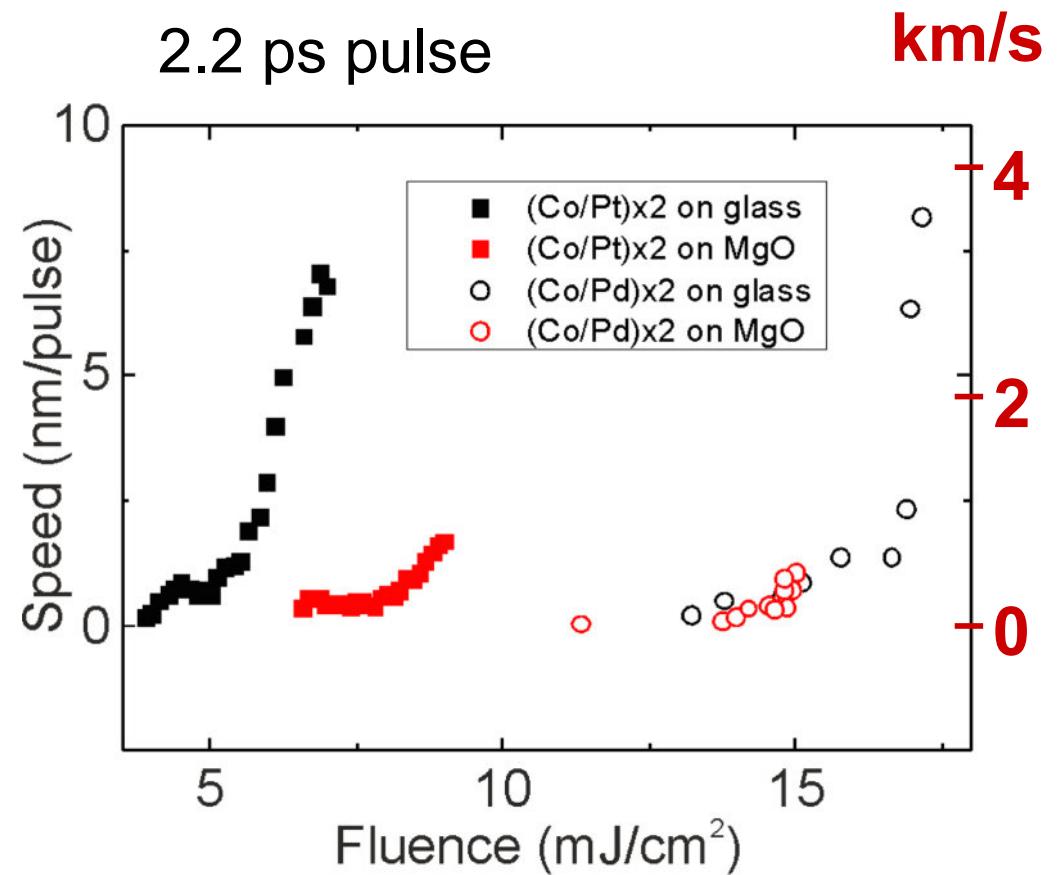
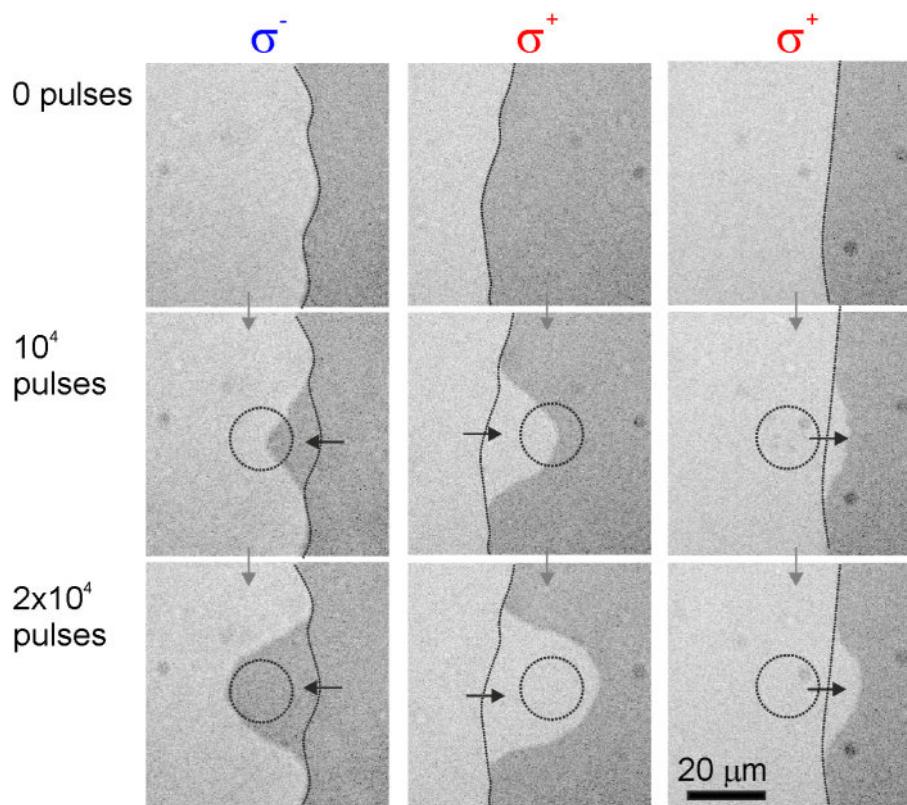


Number-of-pulses dependence in Co/Pt, Co/Pd



The initial nucleation is due to randomized demagnetization,
and is followed by helicity-dependent growth

Domain wall motion (CoPd sample from HGST)

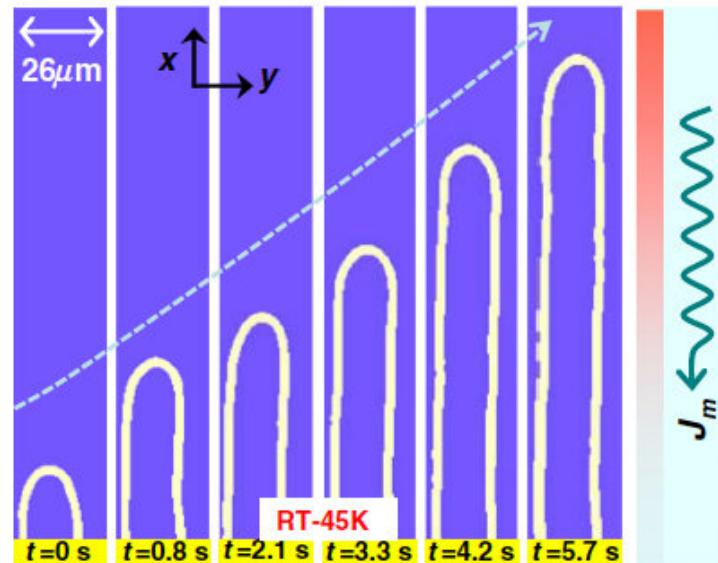
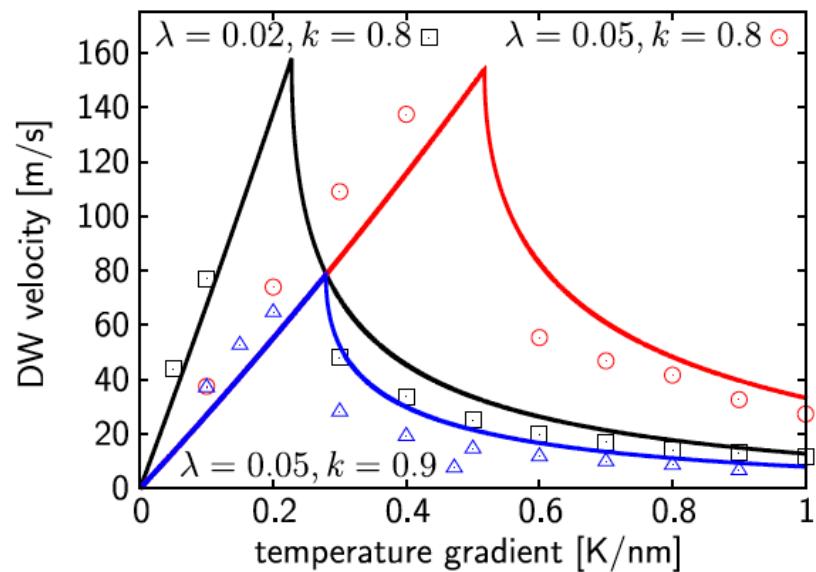


The (too) high speed probably implies after-pulse motion
thermal (MCD) or opto-magnetic?

Entropy, thermal magnons?

Magnon flow

W.Jiang et al, PRL 110, 177202 (2013)



Role of entropy

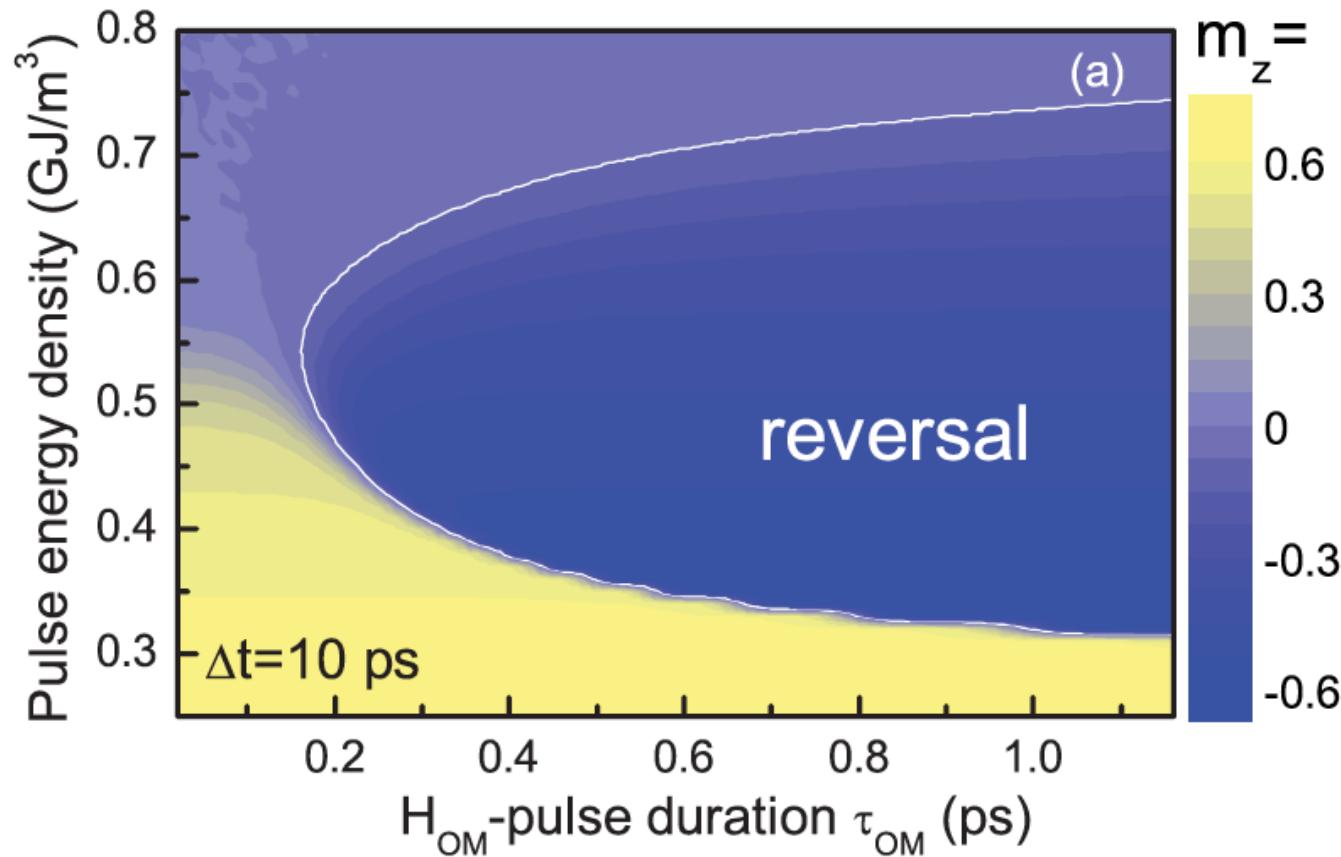
F. Schlickeiser et al, PRL 113, 097201 (2014)

Both thermal, based on MCD

why would they be so sensitive to the pulse width??

inverse Faraday effect

Vahaplar et al, PRB **85**, 104402 (2012)



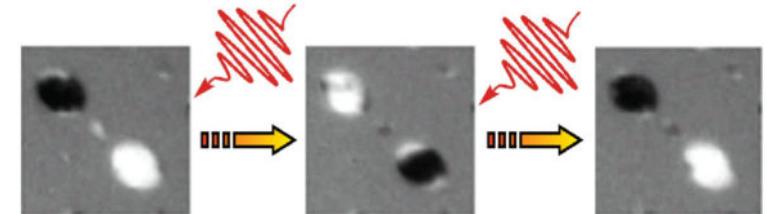
**Combination of thermal + opto-magnetic, better with longer pulses
difficult to estimate the effective field!**

Mechanism: demagnetization-driven nucleation followed by domain-wall motion (magnons, entropy, iFE?) - i.e. thermal + nonthermal

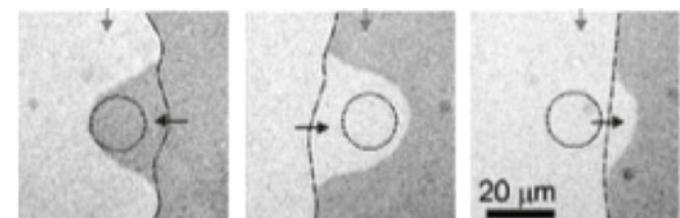
Time-scale: DW motion of few nm/pulse

Summary:

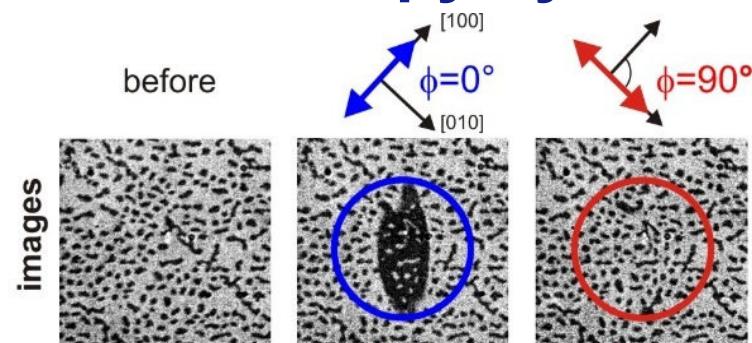
- **Metallic ferrimagnets: thermally-induced, exchange driven toggle switching**



- **Multilayers with strong spin-orbit: domain wall motion by inverse Faraday effect**



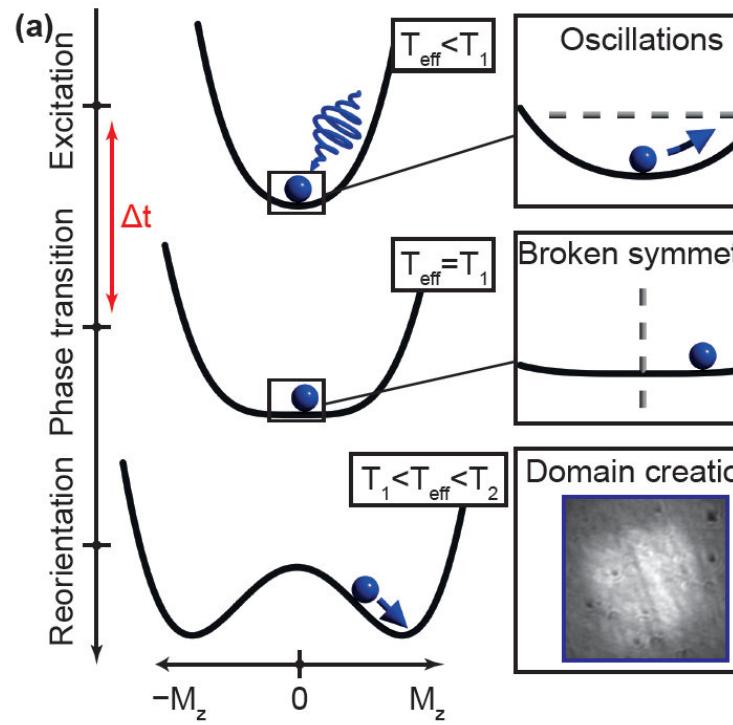
- **Dielectrics: non-thermal, change of anisotropy by photo-magnetic effects**



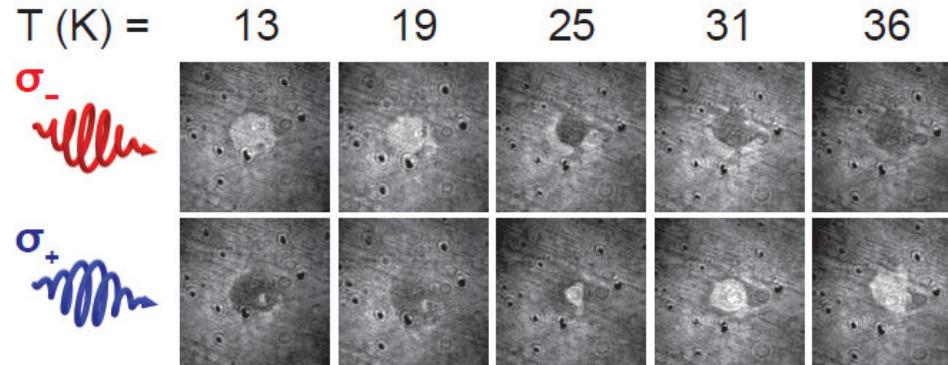
Spare slides

Controlling the route of the phase transition

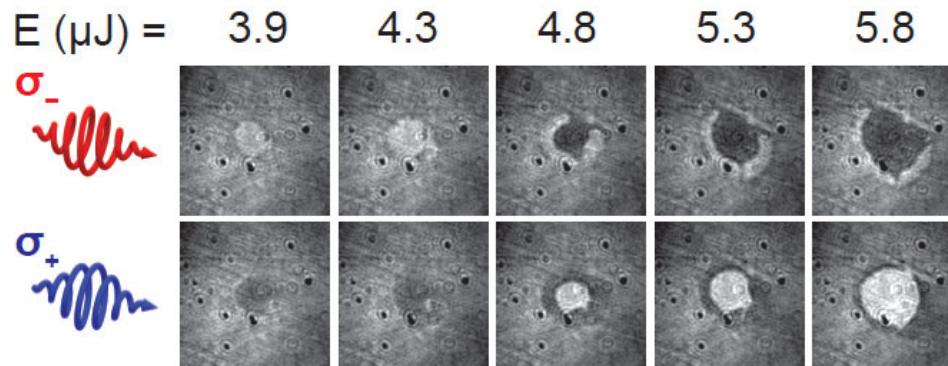
90° reorientation phase transition



$t = 3.7 \text{ ns}, E = 4.3 \mu\text{J}$



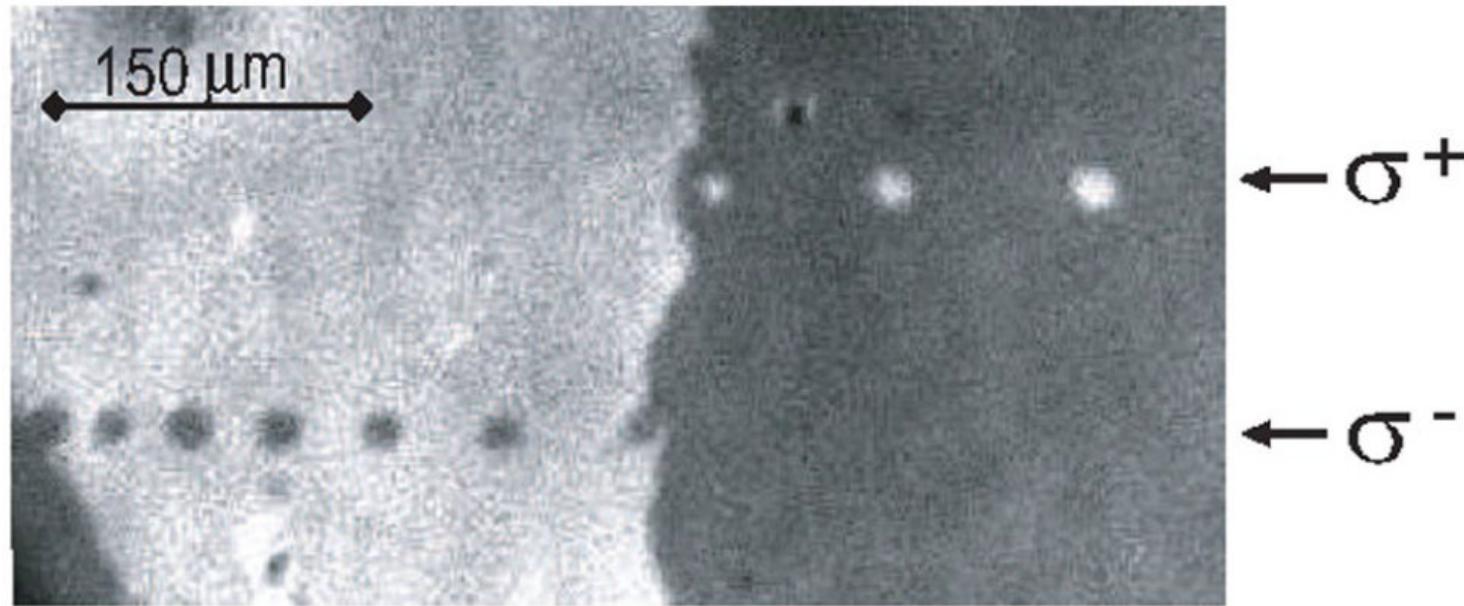
$t = 3.7 \text{ ns}, T = 19 \text{ K}$



de Jong et al, PRL 108, 157601 (2012)

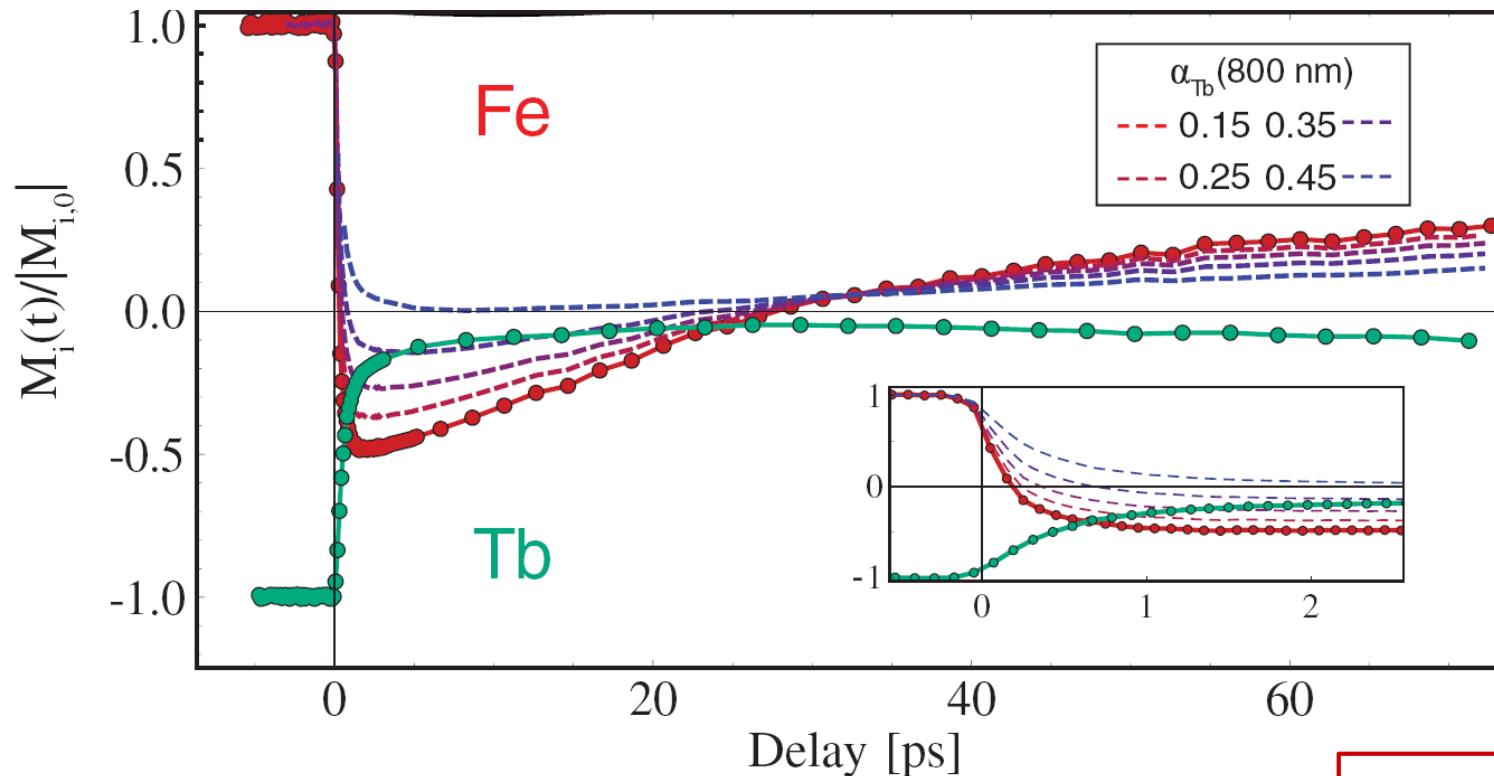
thermal + opto-magnetic

Polarization dependent...



Stanciu et al, Phys. Rev. Lett. **99**, 047601 (2007)

Different ferrimagnets: TbFeCo

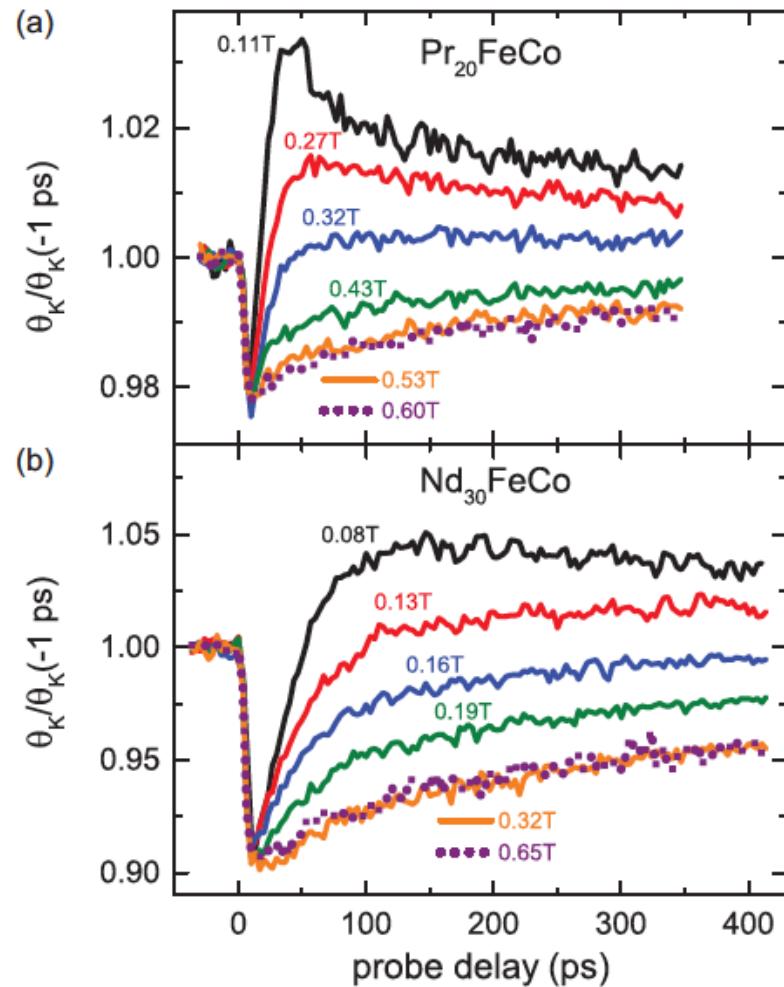


Khorsand et al., Phys. Rev. Lett. **110**, 107205 (2013)

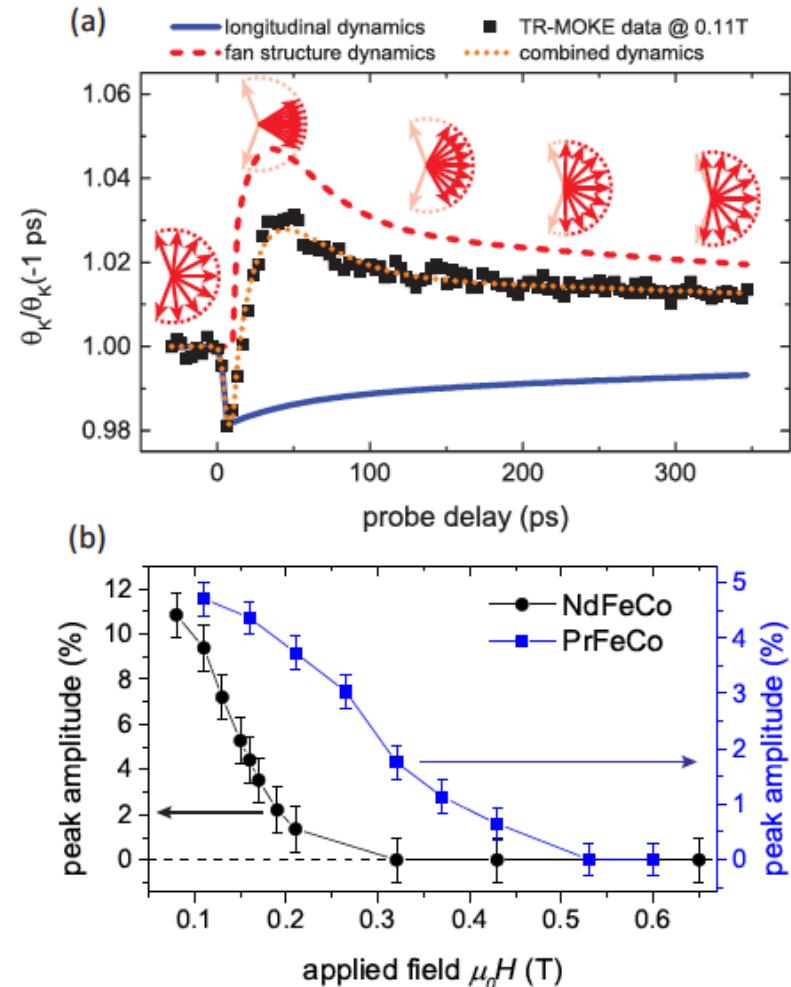
$$g_{\text{Tb}} = \frac{3}{2}$$
$$M_{\text{Tb}} = g_{\text{Tb}} L$$

Different ferrimagnets: NdFeCo and PrFeCo

demagnetization
with an 'overshot'



superimagnetic arrangement

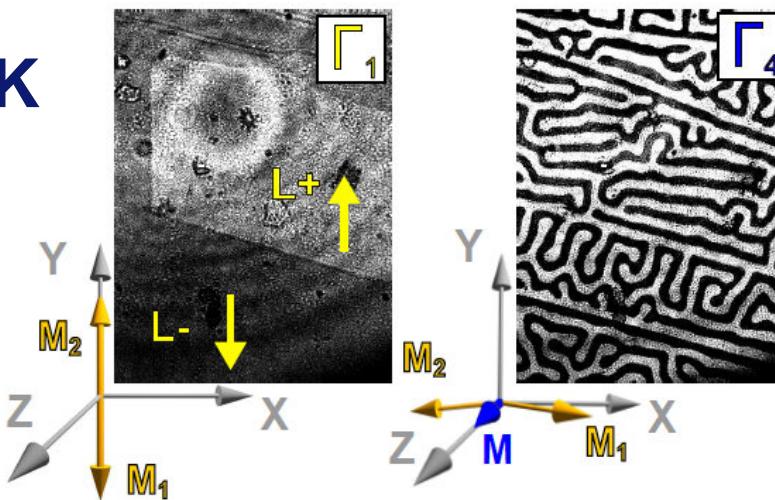


J. Becker et al, Phys. Rev. B **92**, 180407(R) (2015)

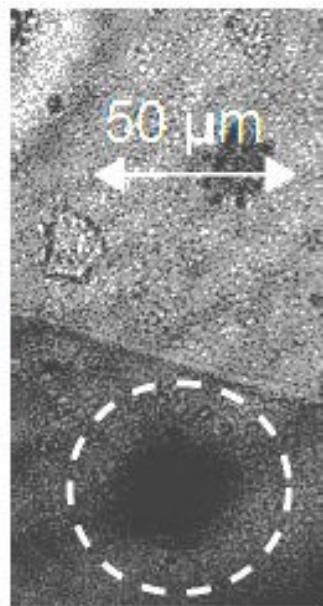
Morin 1st order phase transition in DyFeO₃

$T < T_M = 39 \text{ K}$

AFM



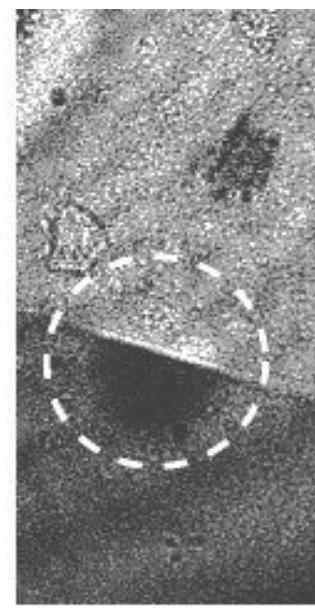
shot 1



shot 2



shot 3



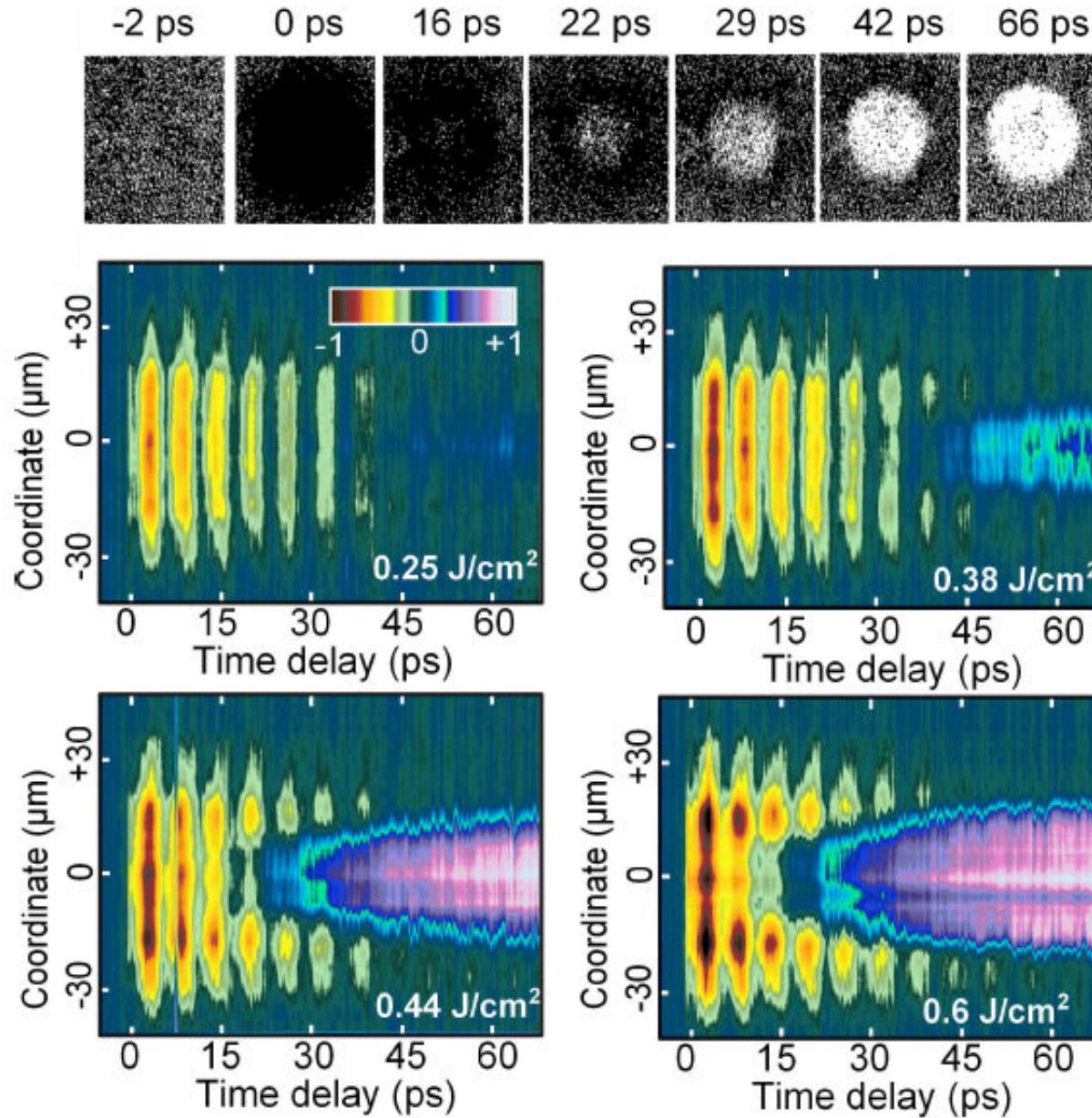
$T > T_M = 39 \text{ K}$

weak FM

D. Afanasiev et al,
PRL 116, 097401 (2016)

same linear polarization

Dynamics: from precession to the new phase



**difference with
2nd order**

D. Afanasiev et al,
PRL 116, 097401 (2016)

**thermal +
opto-magnetic**