

Methoden moderner Röntgenphysik II: Streuung und Abbildung

Lecture 19	Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2018 G. Grübel, <u>A. Philippi-Kobs</u> , F. Lehmkühler, L. Frenzel, M. Martins, W. Wurth		
Location	Lecture hall AP, Physics, Jungiusstraße		
Date	Tuesday	13:00 - 14:30	(starting 3.4.)
	Thursday	8:30 - 10:00	(until 12.7.)



Outline

Part II/2:

Studies on Magnetic Nanostructures

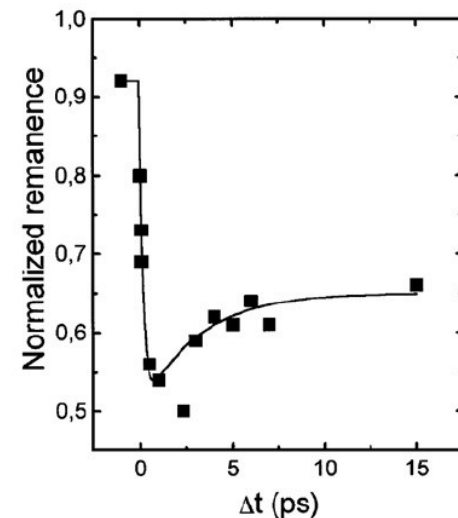
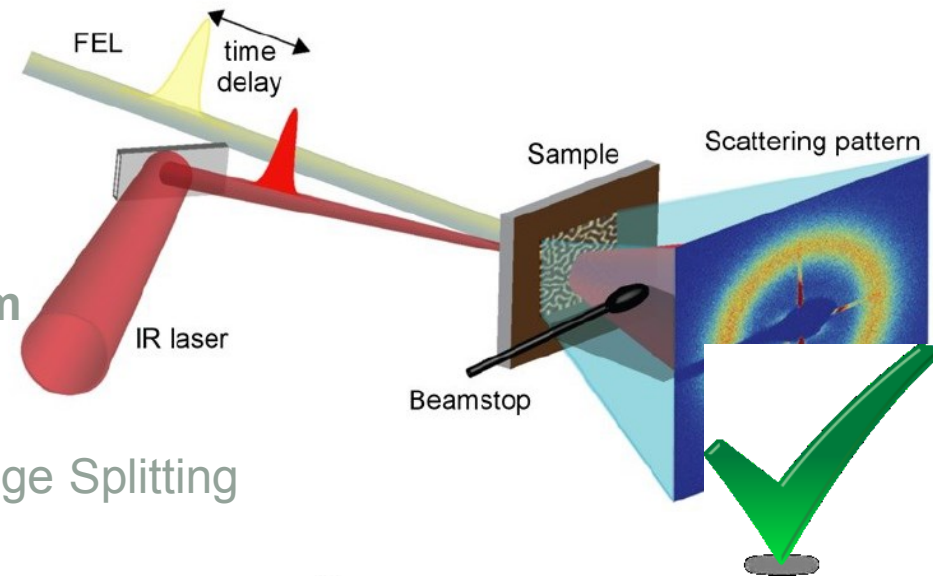
by André Philippi-Kobs (AP)

[19.6.] X-ray Magnetic Circular Dichroism (XMCD) & Resonant Magnetic Small Angle X-ray Scattering (mSAXS)

- Role of Spin-Orbit Coupling and Exchange Splitting
- Sum Rules
- XMLD and Natural Dichroisms
- mSAXS of Magnetic Domain Patterns

[21.6.] Femtomagnetism

- Introduction to Ultrafast Magnetization Dynamics Induced by Femtosecond Infrared Pulses
- Pump-Probe Experiments of Nano-Scale Magnetic Domain Patterns
- All-Optical Switching
- Manipulating Magnetism by XUV and THz Pulses



Time-resolved MOKE (Ni 120 fs)

Femtomagnetism

1.) Ultrafast demagnetization (IR pump, $\lambda = 800 \text{ nm}$)

- Discovery and typical parameters
- Three-temperature model
- Ultrafast demagnetization of nano-scale domain patterns
 - TR Fourier transform holography

2.) All-optical switching (IR pump, $\lambda = 800 \text{ nm}$)

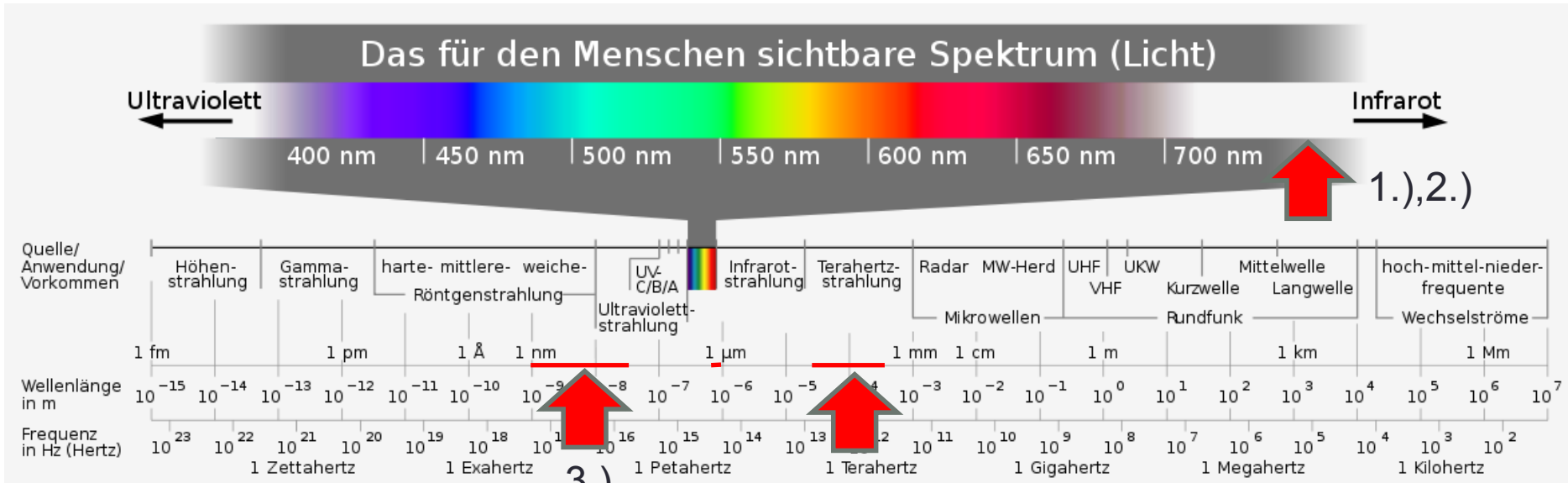
- Ferrimagnetic systems
- Is there AOS in ferromagnetic systems?

3.) High X-ray fluences (XUV pump, $\lambda = 20.8 \text{ nm}$)

4.) THz dynamics (THz pump, $\lambda = 100 \text{ }\mu\text{m}$)

Femtomagnetismus

> Electromagnetic spectrum



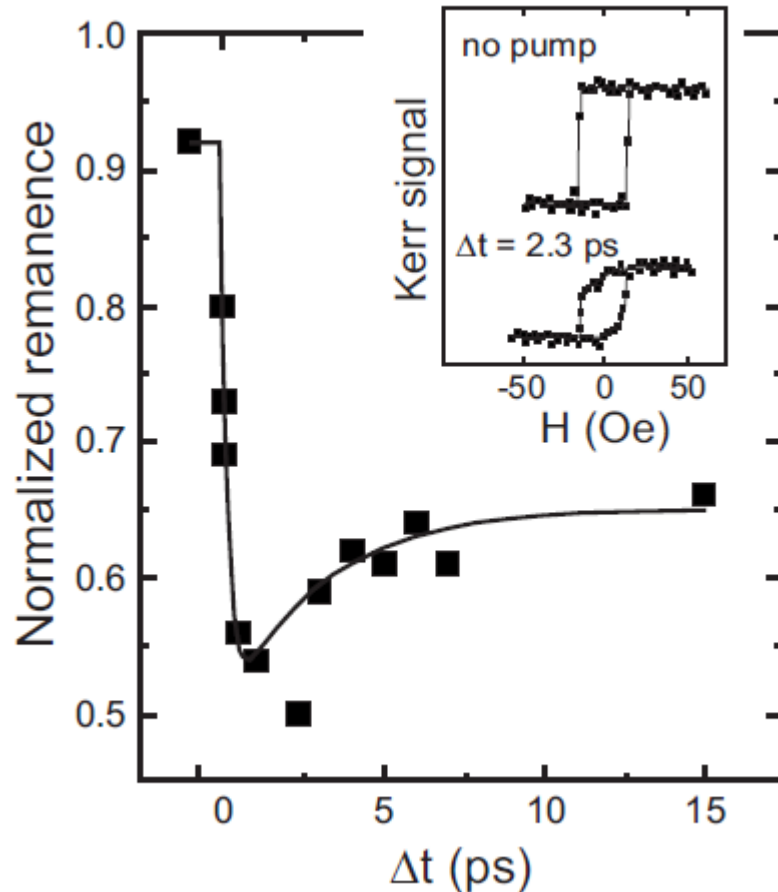
L-edges (soft X-rays)

M-edges (extreme UV, i.e., XUV)

Femtomagnetism

> Ultrafast demagnetization (IR pump, $\lambda = 800$ nm)

- Discovery and typical parameters



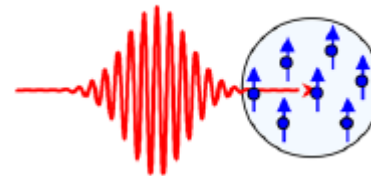
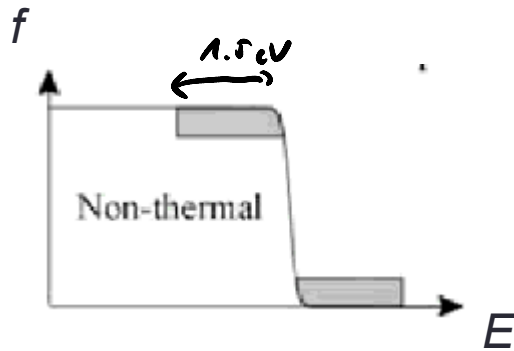
- Pulse duration 70 fs
- In-plane magnetized Ni film
- Characteristic time of demagnetization of 260 fs
- Recovery time of magnetization > 100 ps
- Fluence of 7 mJ/cm^2
($2.8 \cdot 10^{16}$ Photonen/ cm^2 ,
 $4 \cdot 10^{29}$ Photonen/ (cm^2s))

E. Beaurepaire *et al.*, Phys. Rev. Lett. **76**, 4250 (1996).

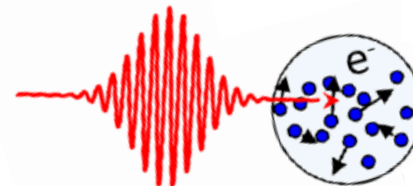
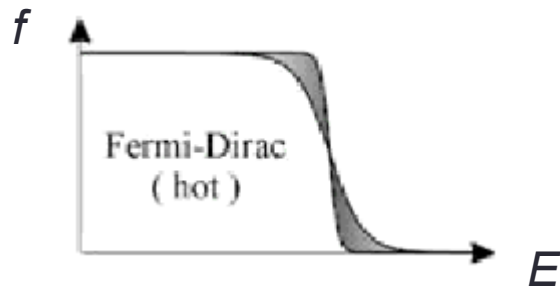
Femtomagnetism

> Ultrafast demagnetization

1.) Creation of highly excited electrons during the pulse duration (< 20 fs)



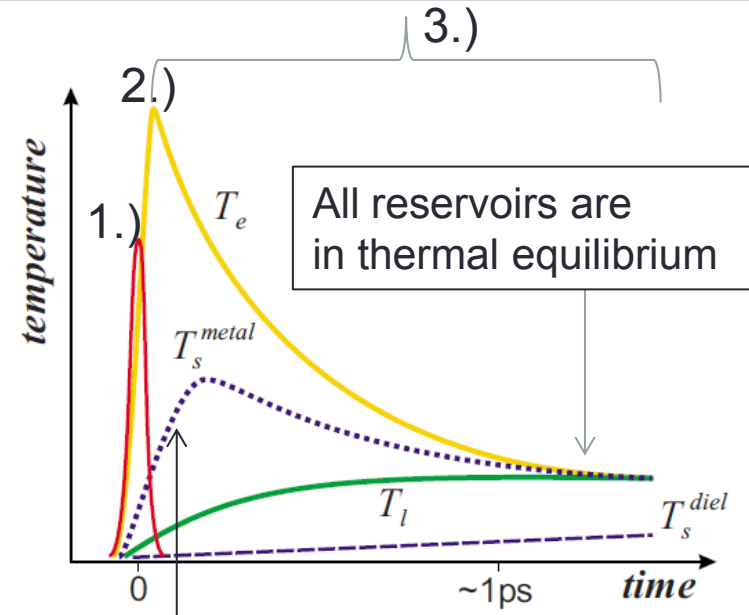
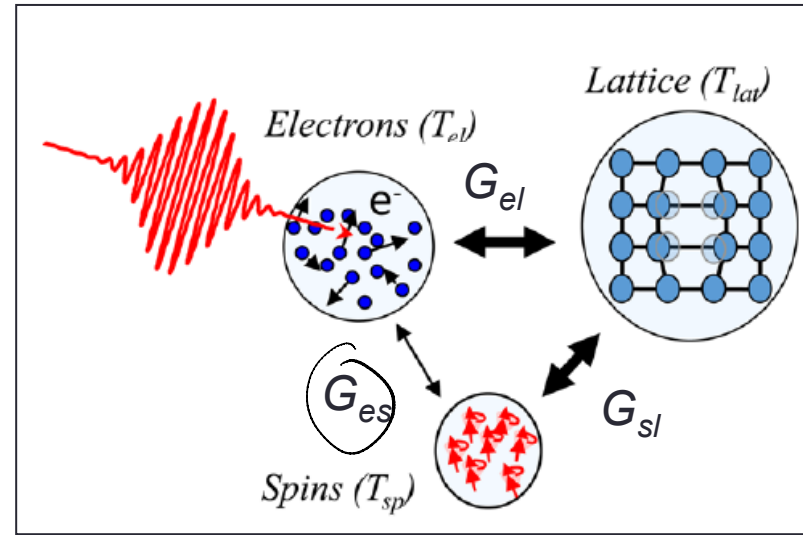
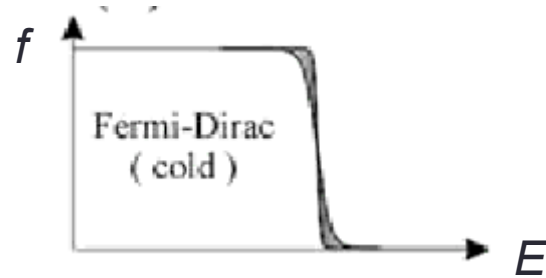
2.) Thermalization of electron system (< 200 fs) due to electron-electron-scattering



Femtomagnetism

> Ultrafast demagnetization

- 3.) Thermalization of electron, phonon, and spin reservoirs (<1 ps) due to electron-phonon-, electron-spin-, and phonon-spin-scattering



→ Differential equations (Rate equations)

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

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

Energy density of pulse

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

C_i : specific heat, T_i : temperature of electrons, phonons, spins
 G_i : strength of interaction between electrons, phonons, spins

Ultrafast demagnetization



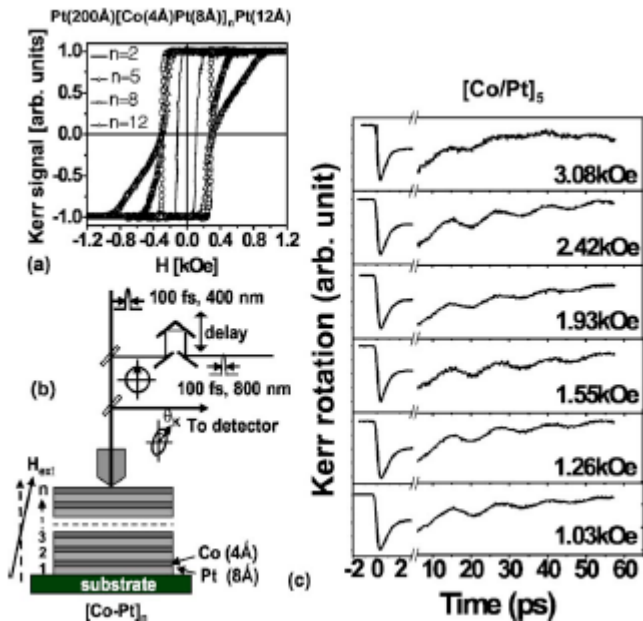
Femtomagnetism

> Ultrafast demagnetization

4.) Cooling of sample due to interaction with environment (~100 ps – 100 ms)

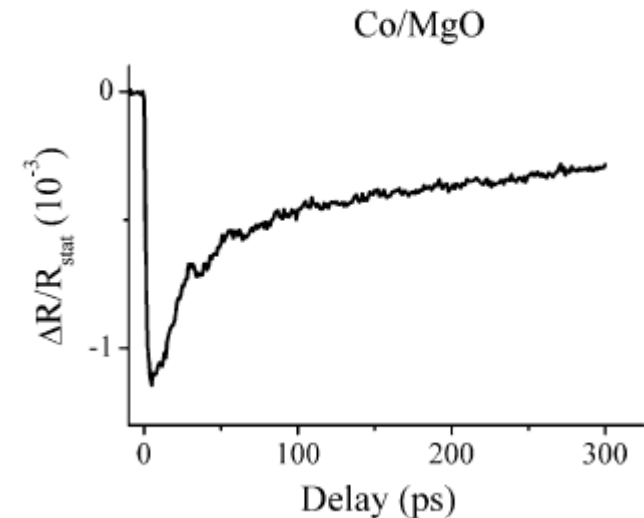
JOURNAL OF APPLIED PHYSICS 101, 09D102 (2007)

Ultrafast magnetization dynamics in high perpendicular anisotropy [Co/Pt]_n multilayers



Chemical Physics 318 (2005) 137–146

Ultrafast magnetization dynamics in ferromagnetic cobalt: The role of the anisotropy



Femtomagnetism

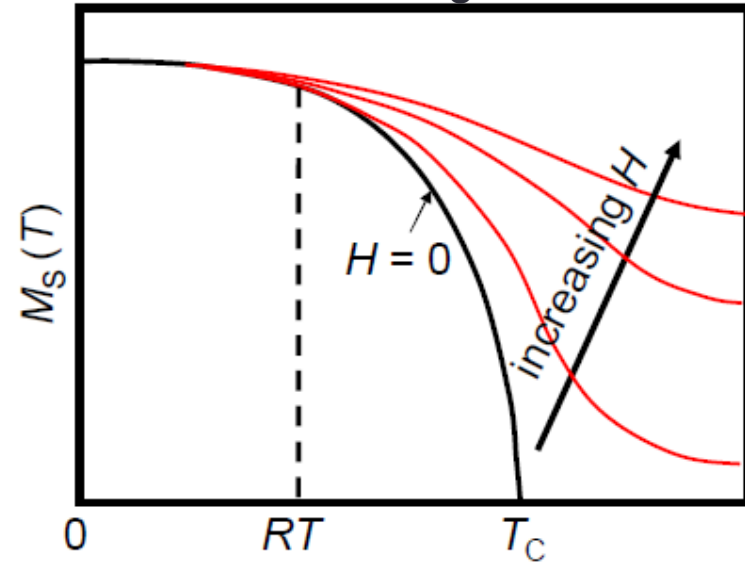
> Ultrafast demagnetization

- T -dependence of saturation magnetization (low T)

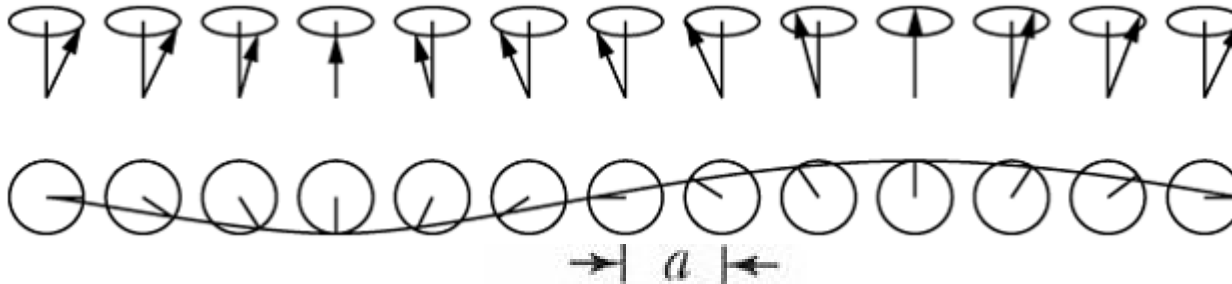
$$M_S(T) = M_S(0)(1 - BT^{3/2})$$

$$B = 3.3 \cdot 10^{-6} \text{ K}^{-3/2} \text{ for bulk Co}$$

“thermal” demagnetization

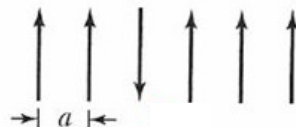


- Collective spin excitations = “magnons” or spin wave (low T)



Magnon reduces saturation magnetization by \hbar

- Single spin excitations (high T)



Single spin exc. reduces saturation magnetization by \hbar

Femtomagnetism

- Ultrafast demagnetization
... of magnetic domain pattern

ARTICLE

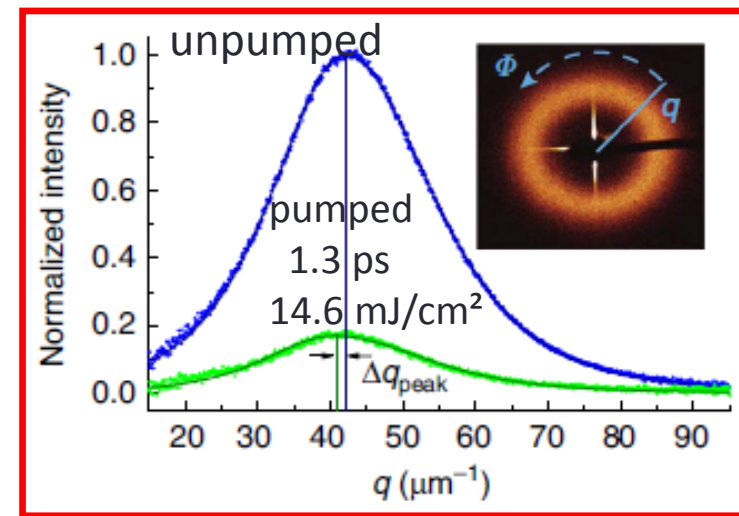
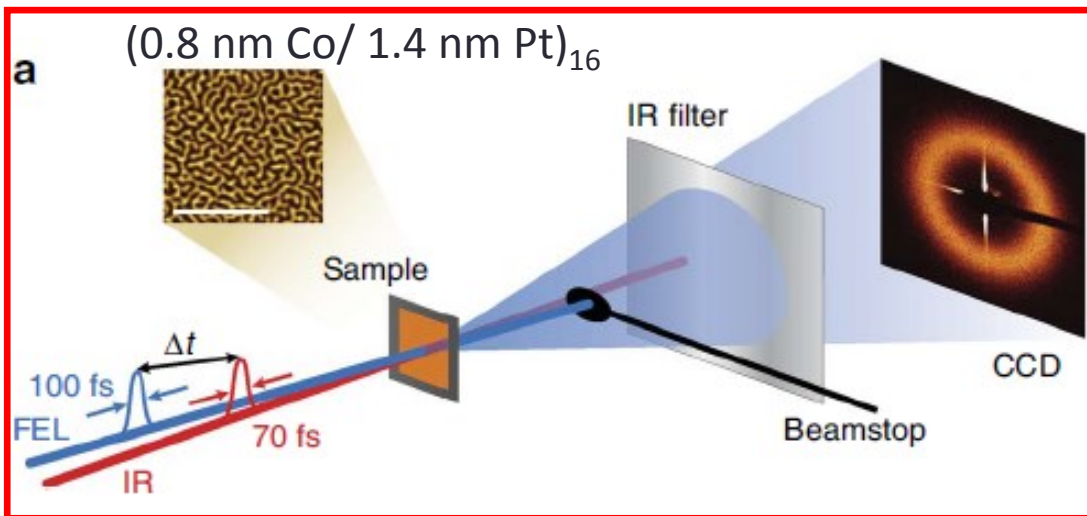
Received 18 Apr 2012 | Accepted 3 Sep 2012 | Published 2 Oct 2012

DOI: 10.1038/ncomms2108

Ultrafast optical demagnetization manipulates nanoscale spin structure in domain walls

B. Pfau¹, S. Schaffert¹, L. Müller², C. Gutt², A. Al-Shemmary², F. Büttner^{1,3,4,5}, R. Delaunay⁶, S. Düsterer²,
 S. Flewett^{1,4}, R. Frömter⁷, J. Geilhufe⁸, E. Guehrs¹, C.M. Günther¹, R. Hawaldar⁶, M. Hille⁷, N. Jaouen⁹,
 A. Kobs⁷, K. Li⁶, J. Mohanty¹, H. Redlin², W.F. Schlotter¹⁰, D. Stickler⁷, R. Treusch², B. Vodungbo^{6,11},
 M. Kläui^{3,4,5}, H.P. Oepen⁷, J. Lüning⁶, G. Grübel² & S. Eisebitt^{1,8}

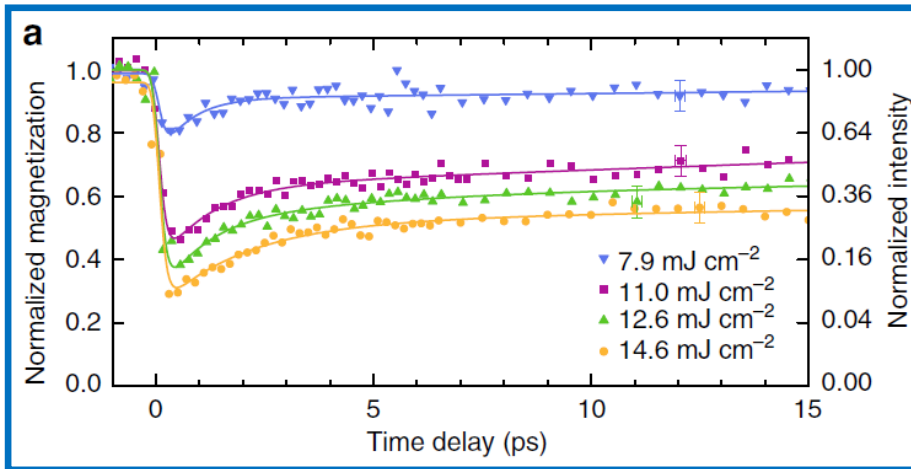
IR-pump–XUV-probe experiment



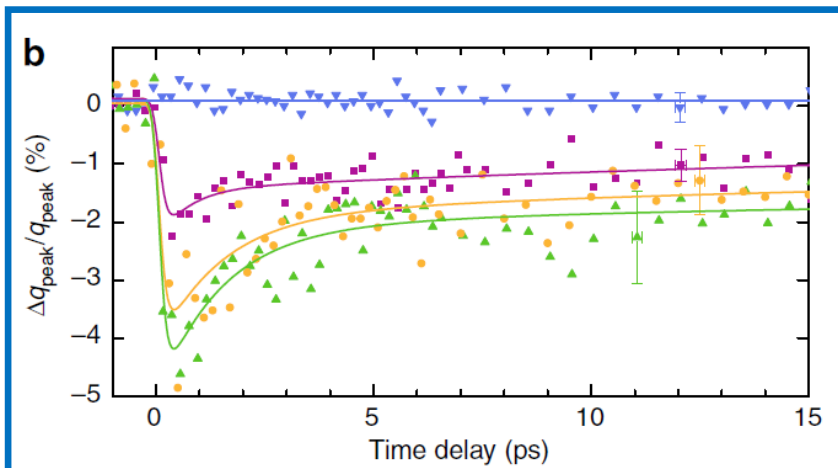
Femtomagnetism

Ultrafast demagnetization

... of magnetic domain pattern



Ultrafast demagnetization (see above)

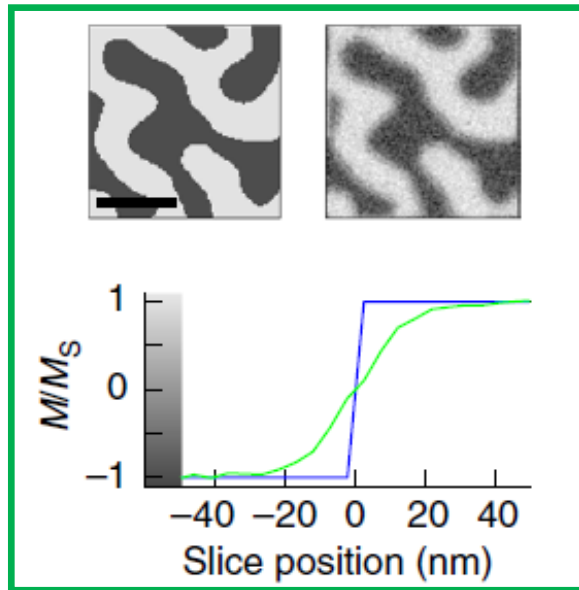


Decrease in radius of scattering ring

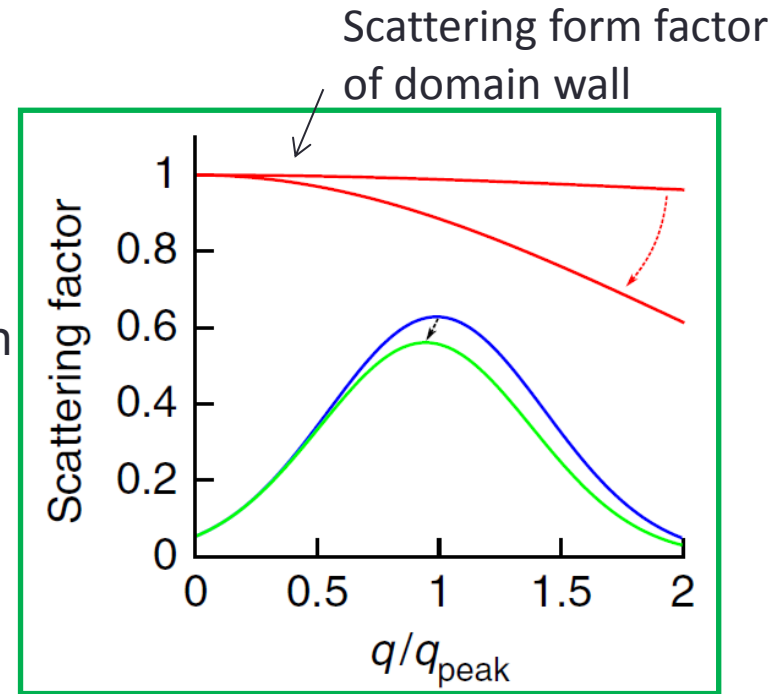
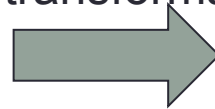
Femtomagnetism

> Ultrafast demagnetization ... of magnetic domain pattern

- Explanation of q_{peak} shift



Fourier transformation



Super-diffusive currents
 broaden domain walls
 by 20 nm (FWHM)

Change of scattering form factor of
 domain walls and hence q_{peak}

B. Pfau *et al.*, Nat. Commun. **3** (2012).

Femtomagnetism

> Ultrafast demagnetization ... of magnetic domain pattern

- Explanation of q_{peak} shift via superdiffusive currents (spin-dependent electron scattering)

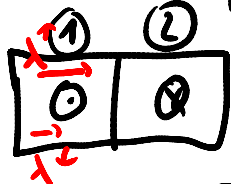
Fermi's Golden rule: Scattering rate of electrons from state k to k'

$$\Gamma_{k,k'} = \frac{2\pi}{\hbar} |\langle \Phi_f(k') | V | \Phi_i(k) \rangle|^2 D(E')$$

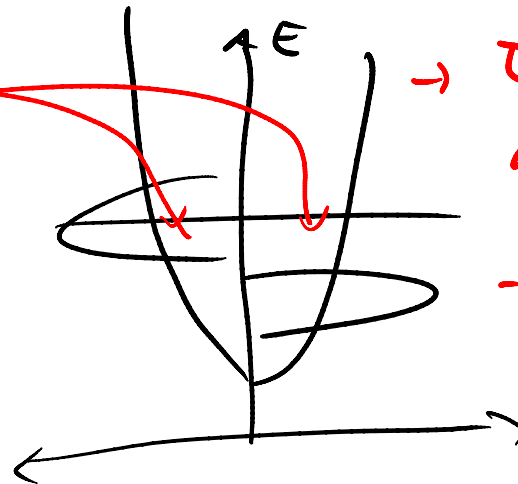
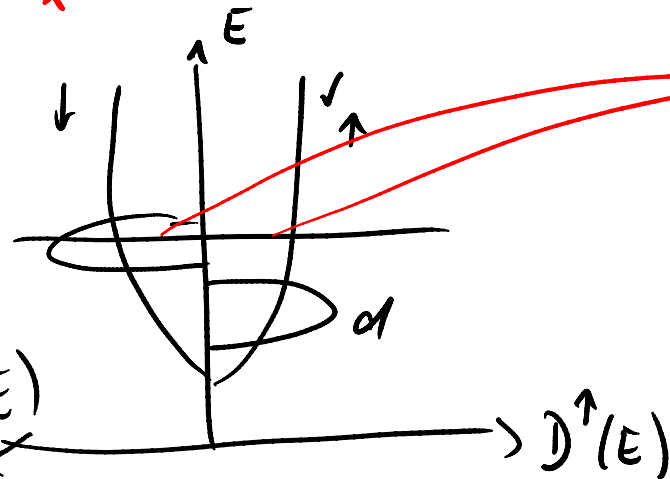
time between scattering events $\tau = 1/\Gamma$

good assumption: spin is conserved during scattering

Mean free path:
 $\lambda = v_F \tau$



$(1) \rightarrow (1)$ (or $(2) \rightarrow (2)$)



$\tau_{\downarrow} \ll \tau_{\uparrow}$
 $\Rightarrow D_f^{\uparrow}(E_F) \ll D_f^{\downarrow}(E_F)$
 $\rightarrow \lambda^{\uparrow} \gg \lambda^{\downarrow}$



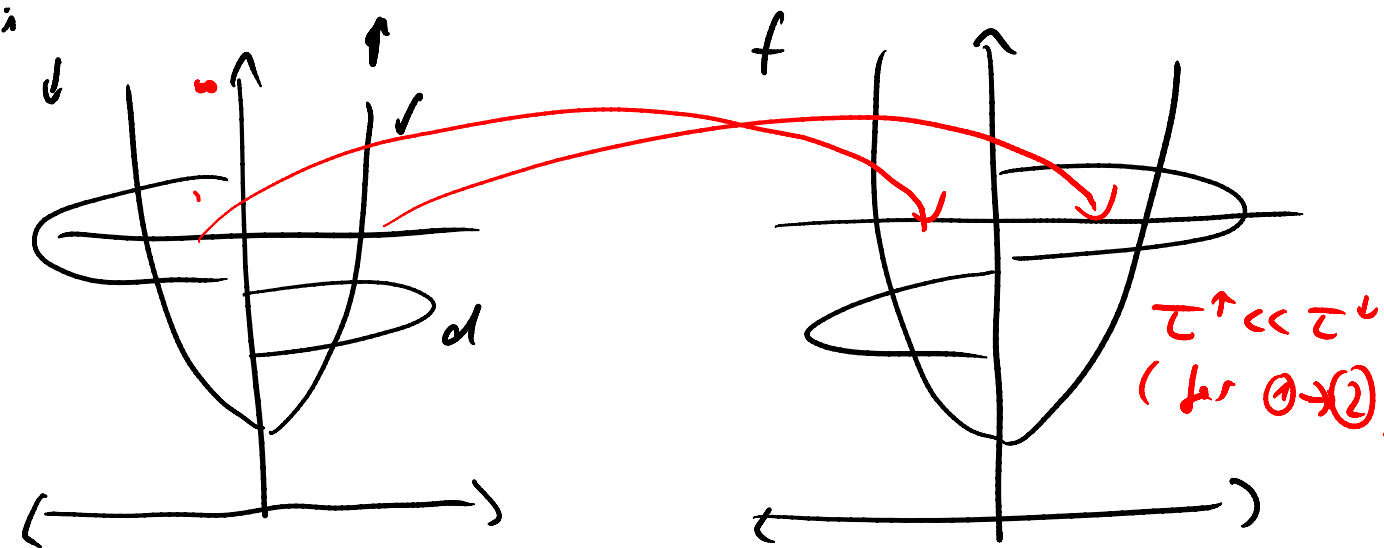
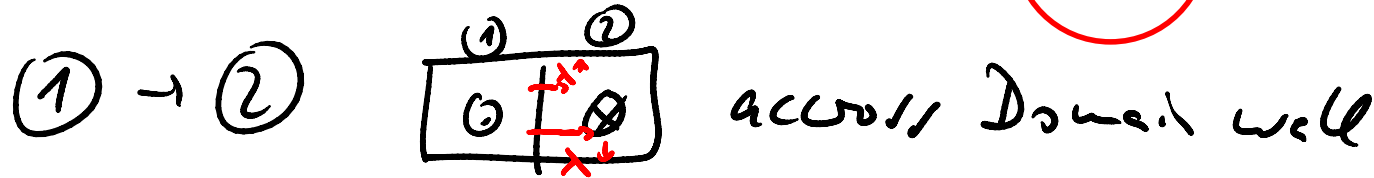
Femtomagnetism

> Ultrafast demagnetization ... of magnetic domain pattern

- Explanation of q_{peak} shift via superdiffusive currents (spin-dependent electron scattering)

Fermi's Golden rule: Scattering rate of electrons from state k to k'

$$\Gamma_{k,k'} = \frac{2\pi}{\hbar} |\langle \Phi_f(k') | V | \Phi_i(k) \rangle|^2 D_f(E')$$



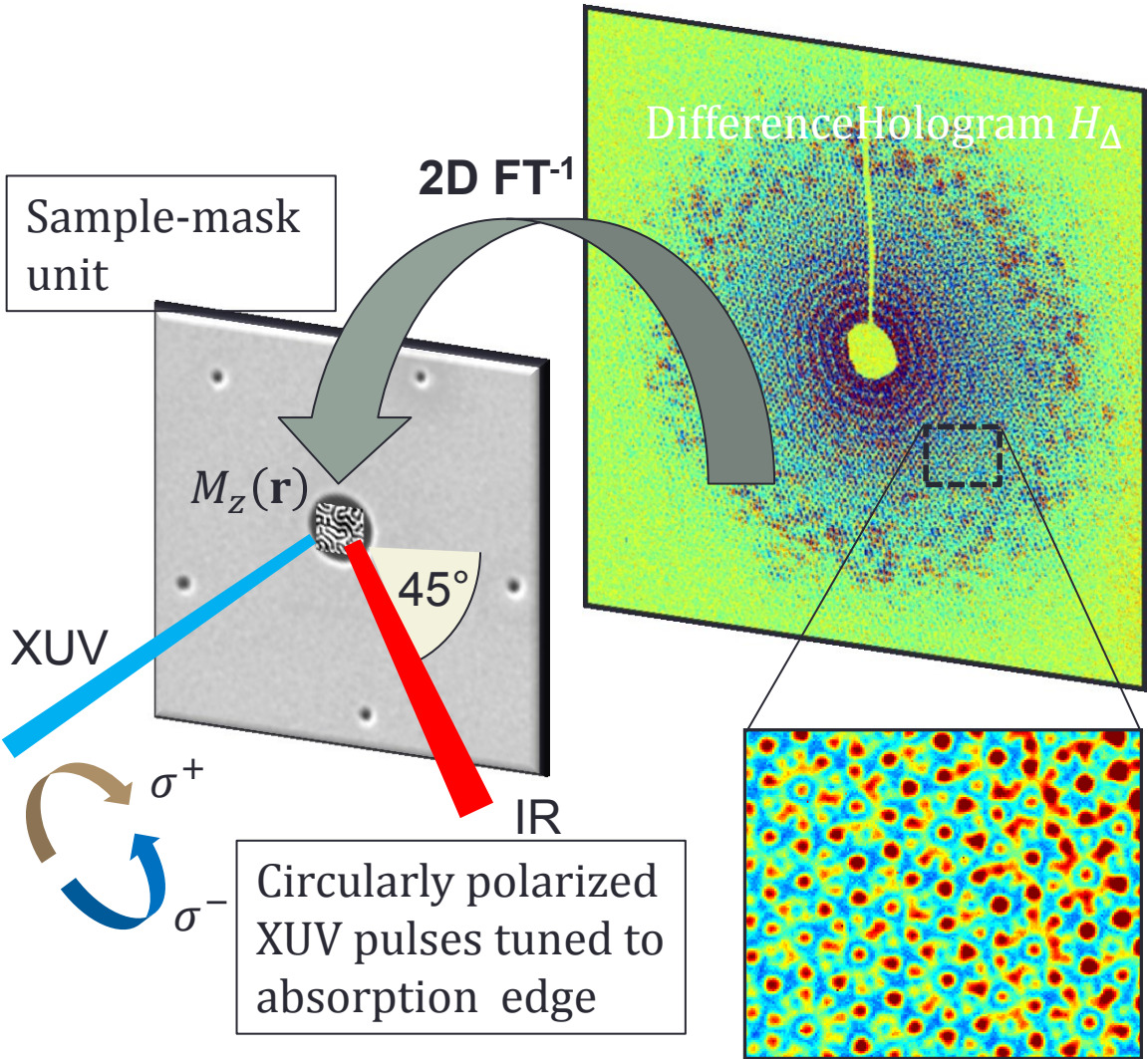
Accumulated spins

→ accumulation of minority spins around domain well
 ≙ reduction of M_s
 ≙ broadening of domain well!



Femtomagnetism

> Ultrafast demagnetization ... of magnetic domain pattern -



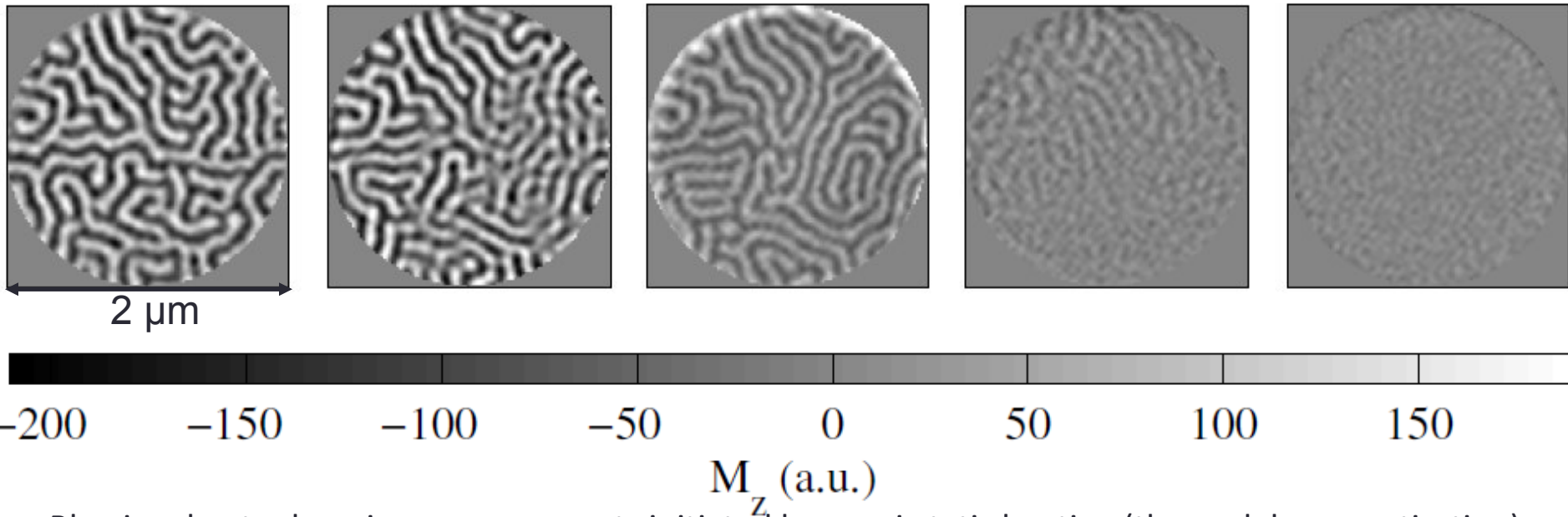
Experimental parameters at FERMI

- 600 shots per helicity
- $4.1 \text{ mJ/cm}^2 \leq \text{fluence} \leq 16.3 \text{ mJ/cm}^2$
- Repetition rate 10 Hz
- $\lambda = 20.8 \text{ nm}$ (M-edge of Co)
- Fixed time delay of 1 ps
- Pump laser ($\lambda = 780 \text{ nm}$) at 45° with respect to sample surface
- Sample: $(\text{Co}(0.4 \text{ nm})/\text{Pd}(0.2 \text{ nm}))_{30}$

Femtomagnetism

➤ Ultrafast demagnetization ... of magnetic domain pattern -

reference 4.1 mJ/cm² 8.2 mJ/cm² 12.2 mJ/cm² 16.3 mJ/cm²



- Blurring due to domain rearrangements initiated by quasi-static heating (thermal demagnetization)
- Global contrast scale \propto saturation magnetization \rightarrow ultrafast demagnetization on the nanoscale
- Small contrast at high fluences due to high demagnetization and/or domain rearrangements
- Resolution of 40 nm limited by exp. geometry is too low to resolve domain wall broadening

Femtomagnetism



All-Optical Magnetic Recording with Circularly Polarized Light

C. D. Stanciu,^{1,*} F. Hansteen,¹ A. V. Kimel,¹ A. Kirilyuk,¹ A. Tsukamoto,² A. Itoh,² and Th. Rasing¹

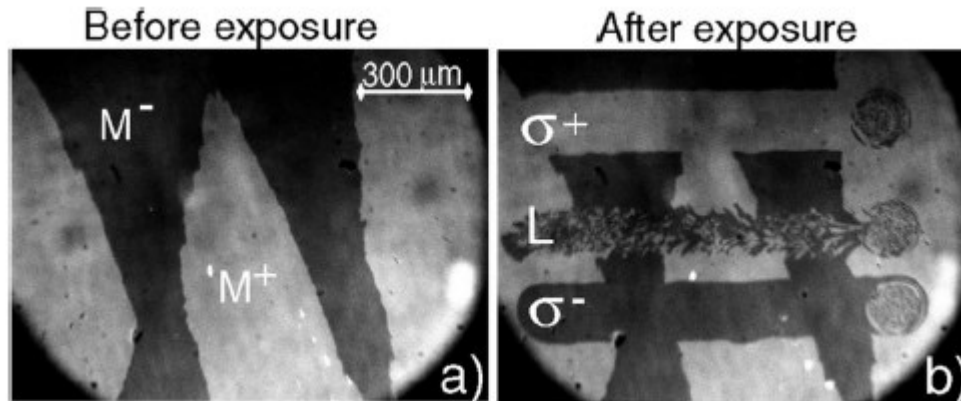
¹Institute for Molecules and Materials, Radboud University Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands

²College of Science and Technology, Nihon University, 7-24-1 Funabashi, Chiba, Japan

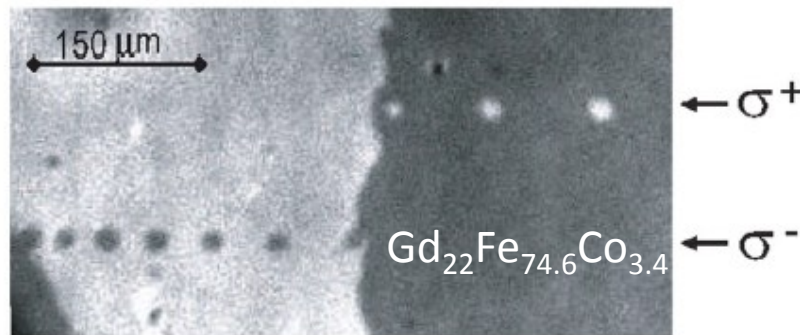
(Received 2 March 2007; published 25 July 2007)

> All-optical switching

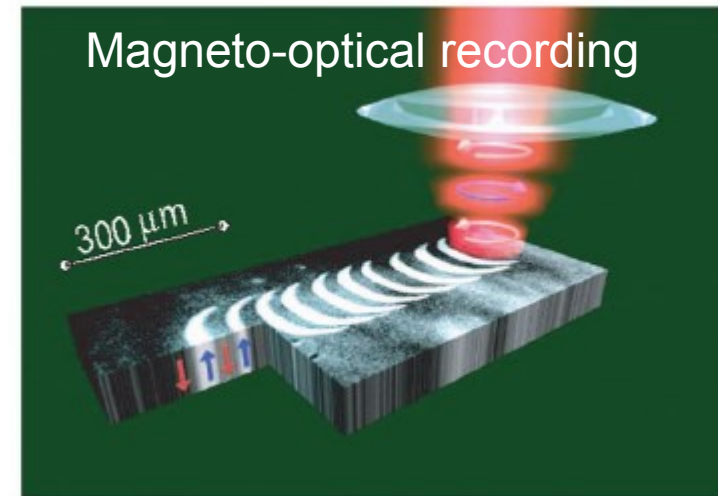
- Discovery for ferrimagnetic materials



Multi-shot



Single-shot



Femtomagnetism

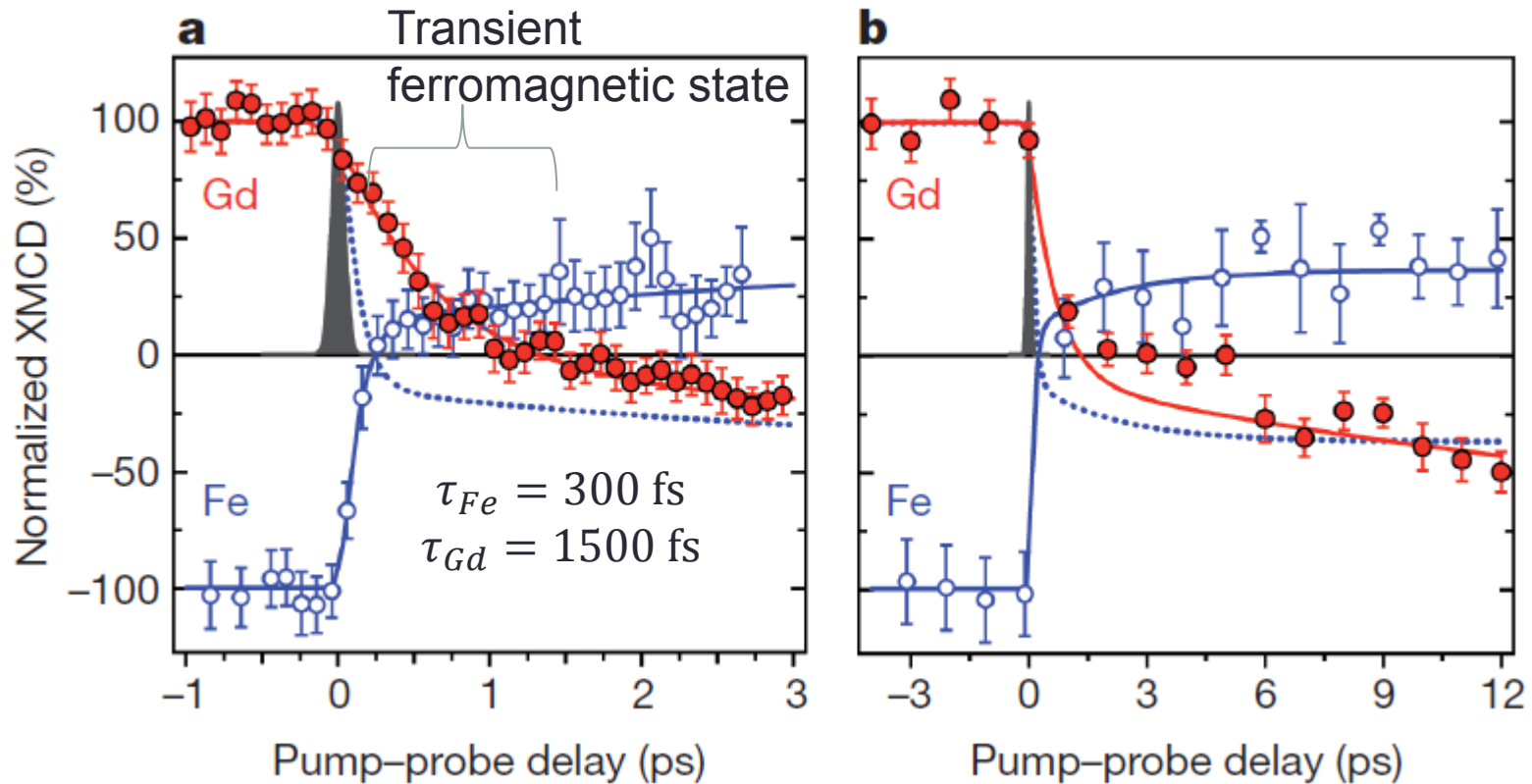
> All-optical switching

- Time-resolved and element-selective studies

Transient ferromagnetic-like state mediating ultrafast reversal of antiferromagnetically coupled spins

2011

I. Radu^{1,2}, K. Vahaplar¹, C. Stamm², T. Kachel², N. Pontius², H. A. Dürr^{2,3}, T. A. Ostler⁴, J. Barker⁴, R. F. L. Evans⁴, R. W. Chantrell⁴, A. Tsukamoto^{5,6}, A. Itoh⁵, A. Kirilyuk¹, Th. Rasing¹ & A. V. Kimel¹

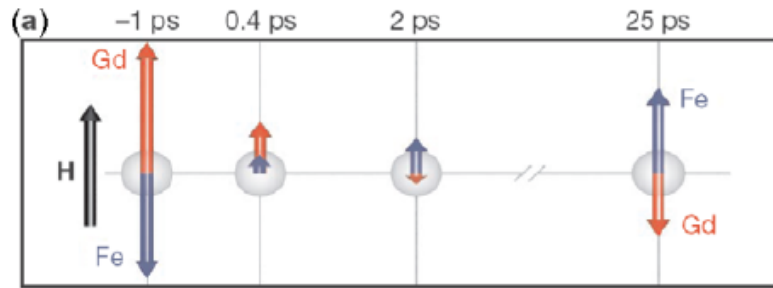


Femtomagnetism

Laser-induced magnetization dynamics and reversal in ferrimagnetic alloys

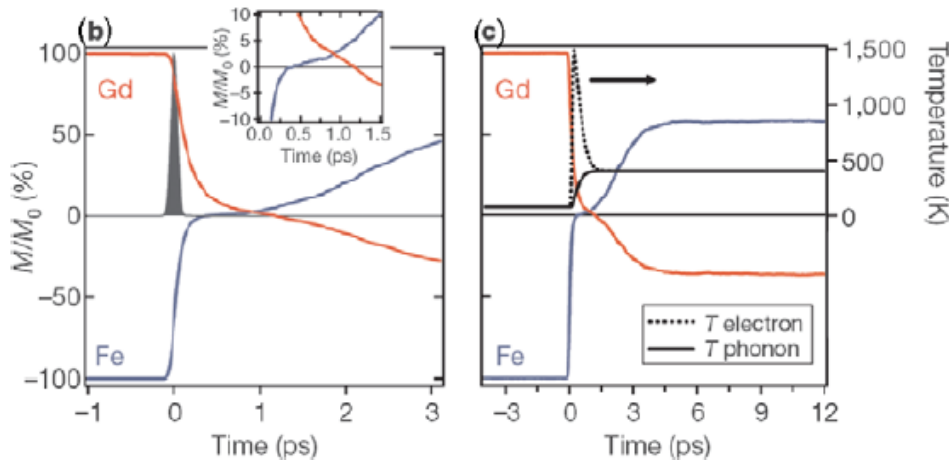
> All-optical switching

- Time-resolved and element-selective studies (theoretical model)



Helicity-independent AOS switching:

- 0-0.3 ps: complete demagnetization of Fe
- 0.4-1.5 ps: transfer of magnetic moment from Gd to Fe to enhance entropy yields to switching of magnetization of Fe
- 1.5-2 ps: antiferromagnetic coupling between Gd and Fe yields to reversing magnetization of Gd
- > 2 ps: Recovery of magnetic moments due to cooling



→ All-optical switching of ferrimagnets!

Helicity-dependent AOS switching due to

- MCD effect (%-effect at off-resonance) + transfer of angular momentum?
- Inverse Faraday effect?

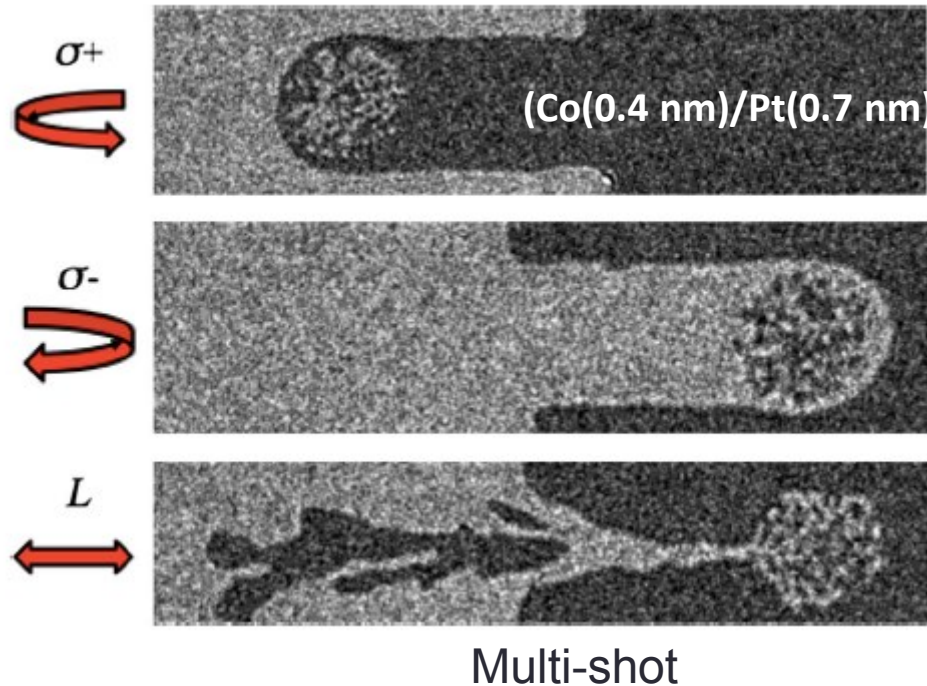
$$\vec{H}_{\text{IFE}} \propto (\vec{E} \times \vec{E}^*)$$



Femtomagnetism

> All-optical switching

- Is there all-optical switching for ferromagnetic materials?



SCIENCE sciencemag.org

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MAGNETISM

All-optical control of ferromagnetic thin films and nanostructures

C.-H. Lambert et al., Science **345**, 1337 (2014).

Many open questions, like e.g.:

- Does deterministic single-pulse HD-AOS work in ferromagnets?
- If yes, for which parameters?
- Underlying mechanisms?
 - Inverse Faraday effect
 - MCD

What we are doing?

We had a beamtime at FLASH in December 2017 to tackle these questions!

Femtomagnetism

> High X-ray fluences

Breakdown of the X-Ray Resonant Magnetic Scattering Signal during Intense Pulses of Extreme Ultraviolet Free-Electron-Laser Radiation

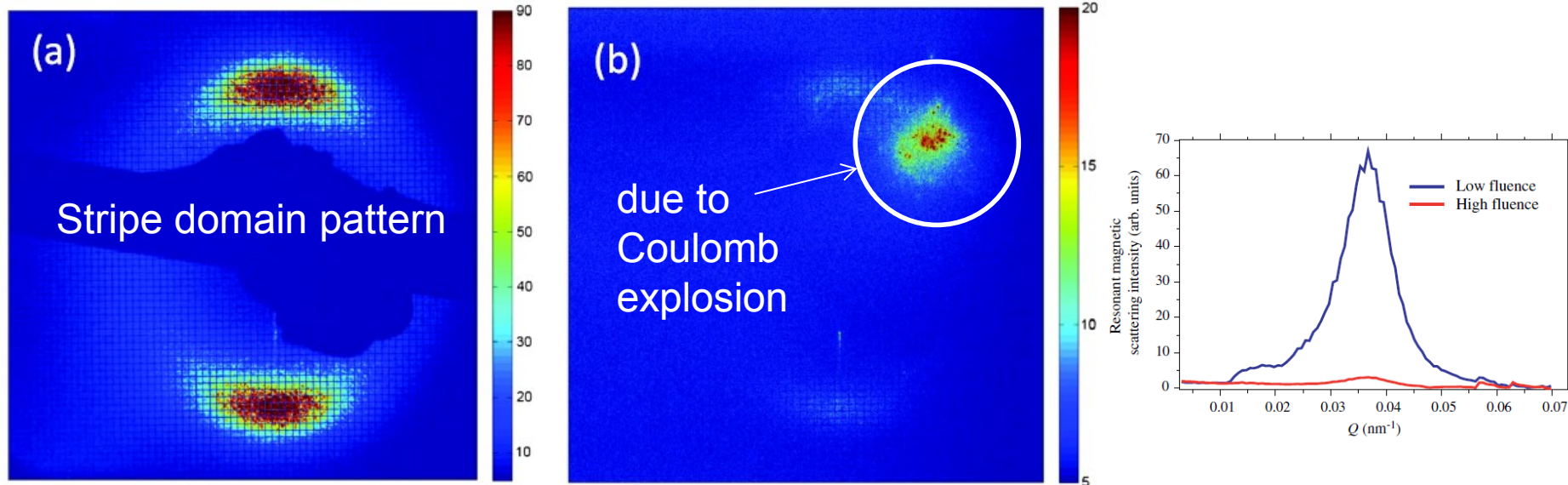
L. Müller,^{1,*} C. Gutt,^{1,2} B. Pfau,³ S. Schaffert,³ J. Geilhufe,⁴ F. Büttner,³ J. Mohanty,³ S. Flewett,³ R. Treusch,¹ S. Düsterer,¹ H. Redlin,¹ A. Al-Shemmary,¹ M. Hille,⁵ A. Kobs,⁵ R. Frömter,⁵ H. P. Oepen,⁵ B. Ziaja,^{1,2,6,7} N. Medvedev,^{1,6} S.-K. Son,^{1,6} R. Thiele,^{1,6} R. Santra,^{1,2,6,8} B. Vodungbo,⁹ J. Lüning,⁹ S. Eisebitt,^{3,4} and G. Grübel^{1,2}

What is happening at high X-ray fluences ($\lambda = 20.8$ nm, i.e., M-edge of Co)?

Experiment at FLASH (Free-electron Laser in Hamburg), pulse duration of ~ 100 fs

1000 shots à 7.5 mJ/cm²

1 shot à 5 J/cm²



- ➔ Intra-pulse “quenching”/ breakdown of the resonant mSAXS signal
- ➔ Violation of principle “diffract before destruct”
- ➔ X-ray pulse does not only act as a non-invasive probe

Femtomagnetism

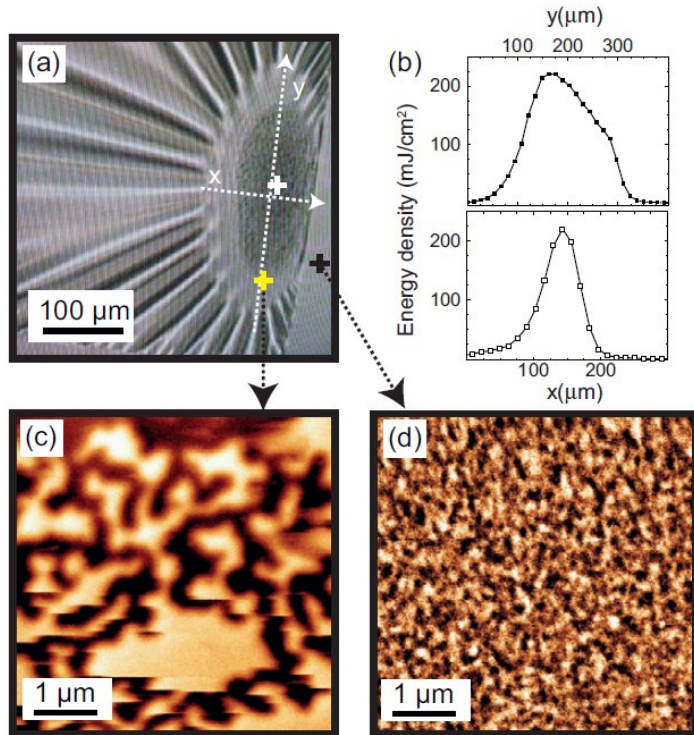
> High X-ray fluences

- Beam damage

Fluence of 200 mJ/cm²

Femtosecond-laser-induced modifications in Co/Pt multilayers studied with tabletop resonant magnetic scattering

C. WEIER¹, R. ADAM¹, D. RUDOLF¹, R. FRÖMTER², P. GRZYCHTOL³, G. WINKLER², A. KOB², H. P. OEPEN², H. C. KAPTEYN³, M. M. MURNANE³ and C. M. SCHNEIDER¹

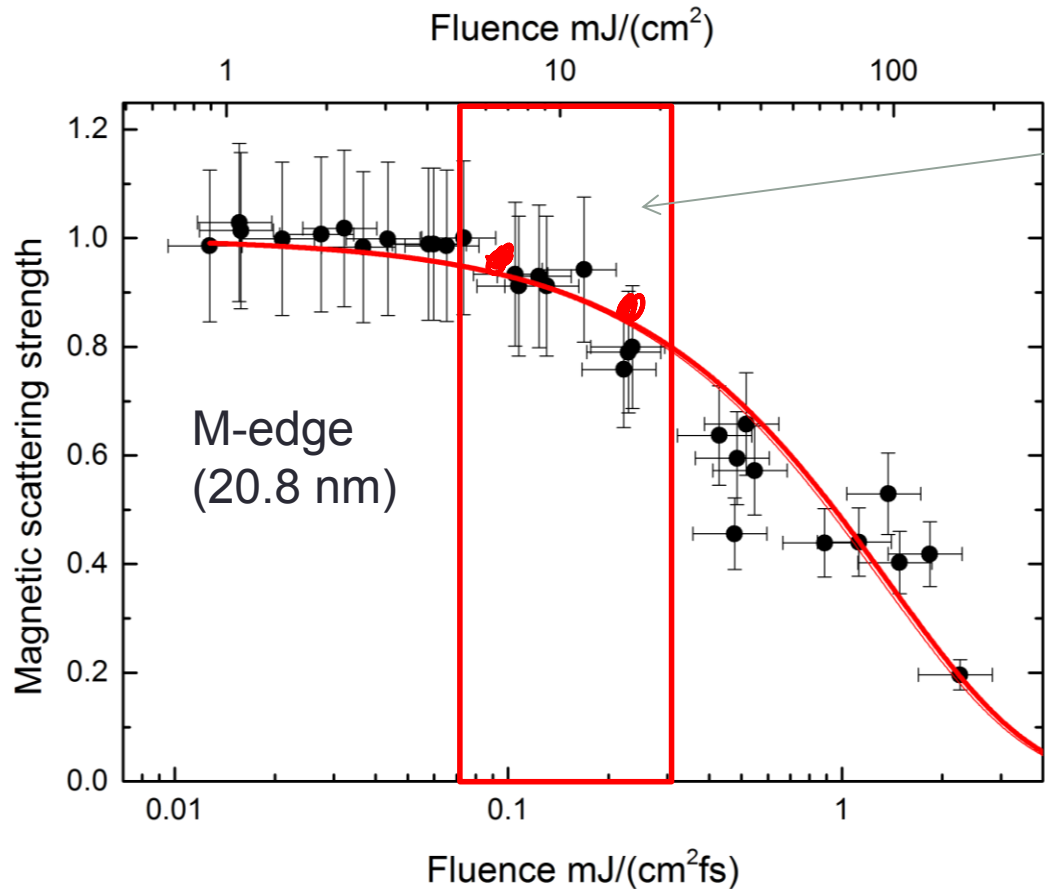


- Single shot experiments
- Many membranes needed

Femtomagnetism

> High X-ray fluences

- Fluence dependence of quenching (measured at FEL FERMI in Trieste, Italy)



- Typically used in „classical“ pump-probe experiments!
- ➔ Variation of scattering strength by ~20%
- ➔ Effect superimposed on data!

Femtomagnetism

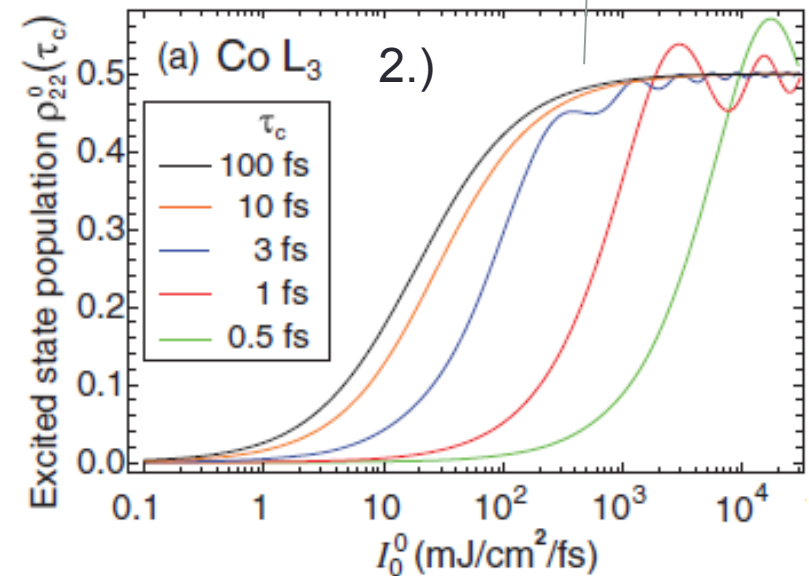
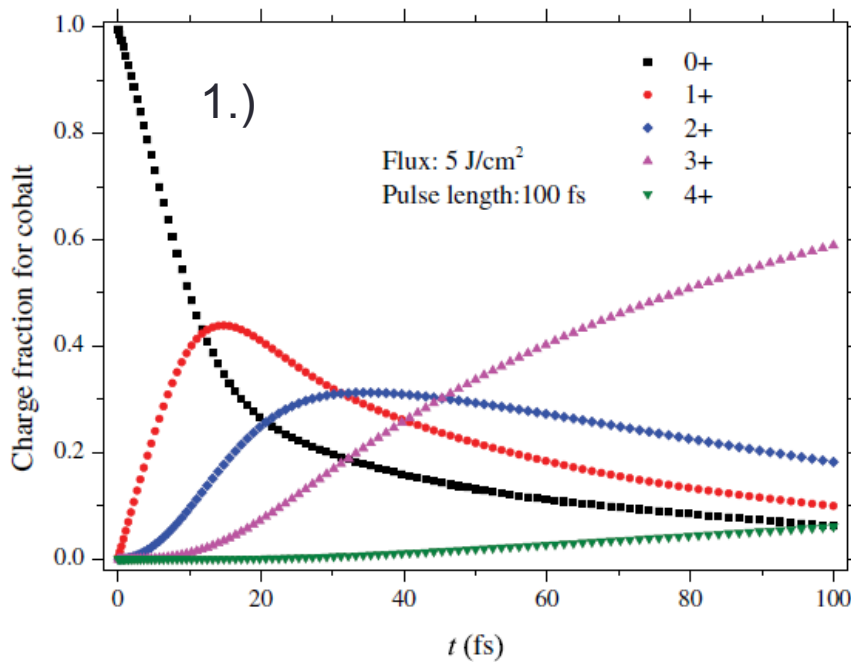
> High X-ray fluences

Creation of X-Ray Transparency of Matter by Stimulated Elastic Forward Scattering

J. Stöhr^{1,*} and A. Scherz^{2,†}

Possible reasons for quenching (ongoing research)

- 1.) Creation of highly ionized state and changing band-structure
- 2.) Stimulated elastic forward scattering
- 3.) Ultrafast demagnetization



Müller et al., Phys. Rev. Lett. **110**, 234801 (2013)

Femtomagnetism

LETTERS

PUBLISHED ONLINE: 11 AUGUST 2013 | DOI: 10.1038/NPHOTON.2013.209

nature
photonics

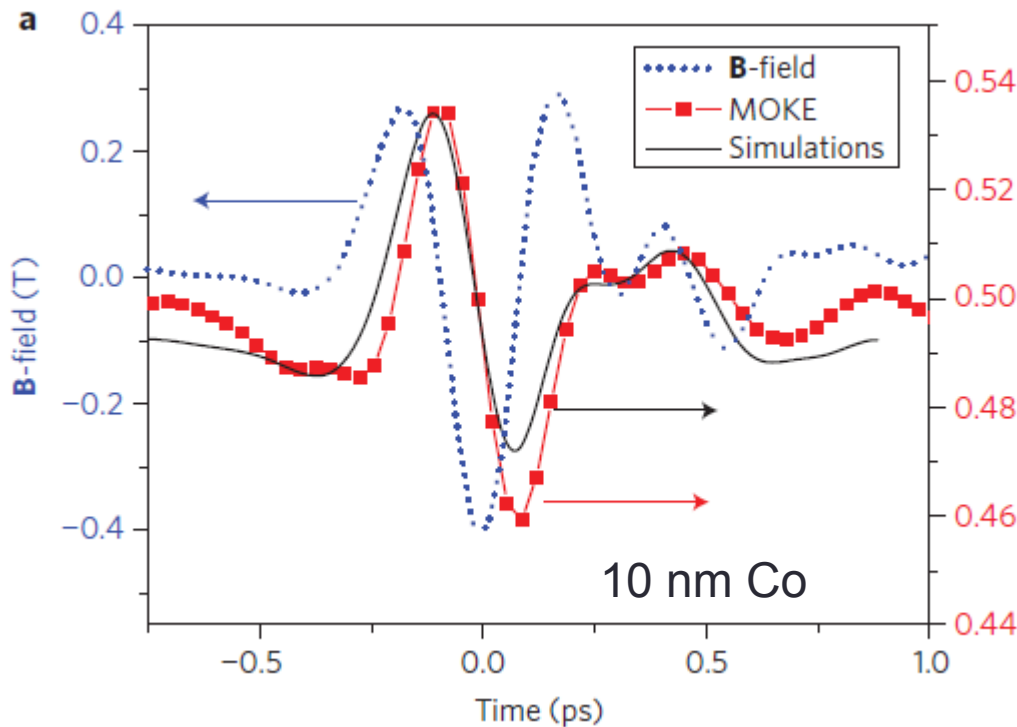
> THz dynamics

Idea and typical parameters

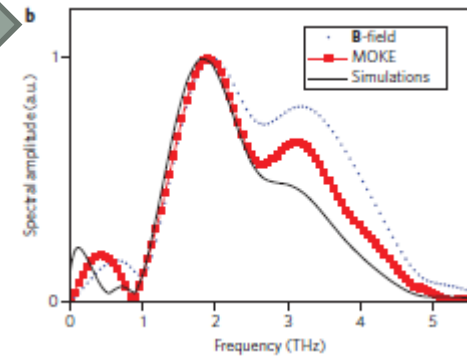
Off-resonant magnetization dynamics phase-locked to an intense phase-stable terahertz transient

C. Vicario¹, C. Ruchert¹, F. Ardana-Lamas^{1,2}, P. M. Derlet³, B. Tudu⁴, J. Luning⁴ and C. P. Hauri^{1,2*}

Coherent control of magnetization (magnetization can follow **B**-field of THz-pulse)



Kerr rotation (mrad)



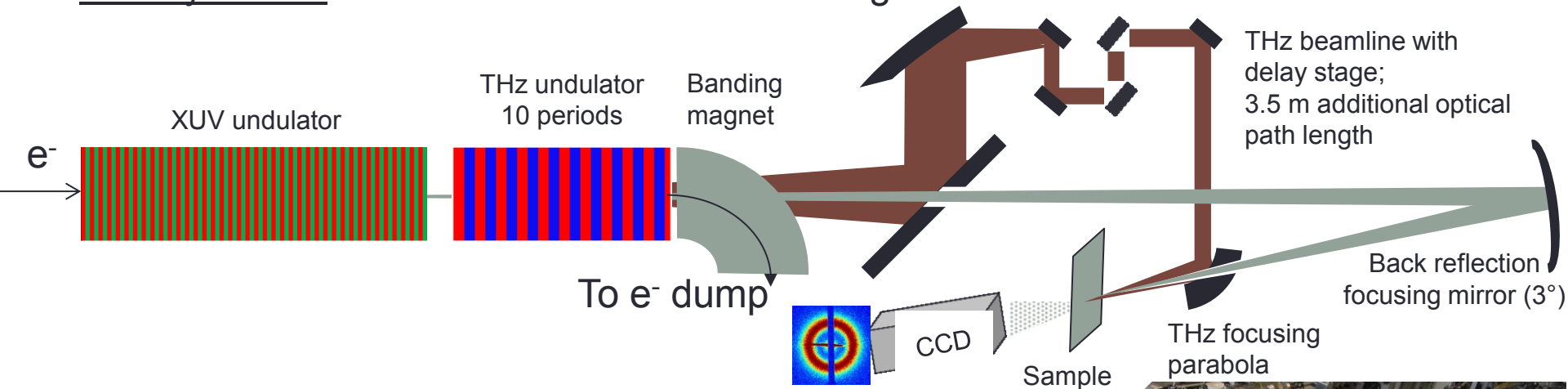
- Fluence of 10 mJ/cm²
- |**B**|-field of 200 mT
- |**E**|-field of 60 MV/m
- role of **E**-field?



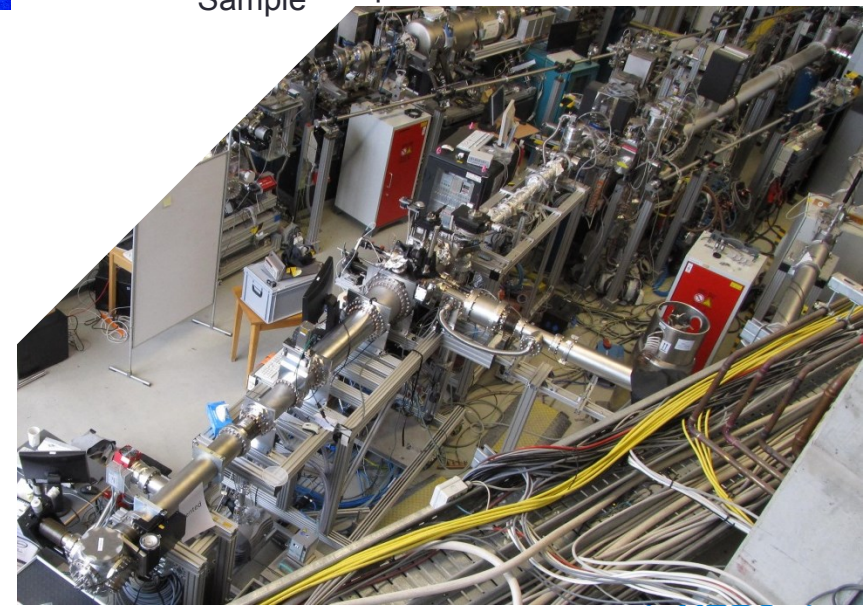
Femtomagnetism

> THz dynamics

What are we doing?



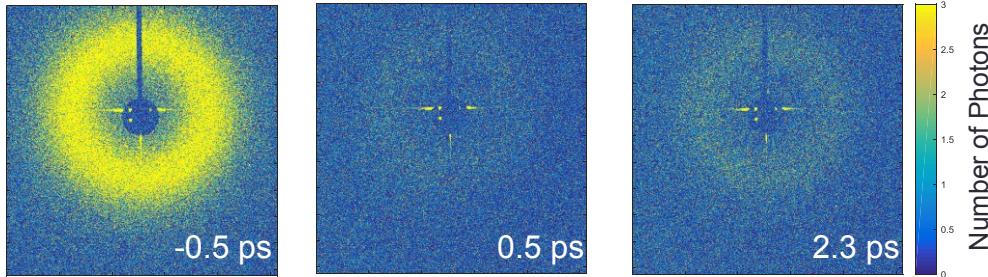
- THz radiation produced from same electron bunch as XUV by a separate electromagnetic undulator with 10 periods
 - Produces *10 full cycles of THz radiation*
- Intrinsic synchronization of XUV and THz pulse; jitter as low as 5 fs (rms)
 - Measurements under phase stable conditions
- THz frequencies from 1.5 – 30 THz



Femtomagnetism

> THz dynamics

What are we doing



- THz radiation (30 μm and longer) from FLASH demagnetizes a nanoscale magnetic domain systems.
- The demagnetization is accompanied by a shift in the $S(q)$ peak position of $\sim 8\%$.
- In contrast to IR demagnetization, the dynamics of both seems to be different

