Methoden moderner Röntgenphysik II: Streuung und Abbildung

Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2019

G. Grübel, F. Lehmkühler, L. Müller, O. Seeck

Location Lecture hall INF, Physics, Jungiusstraße 11

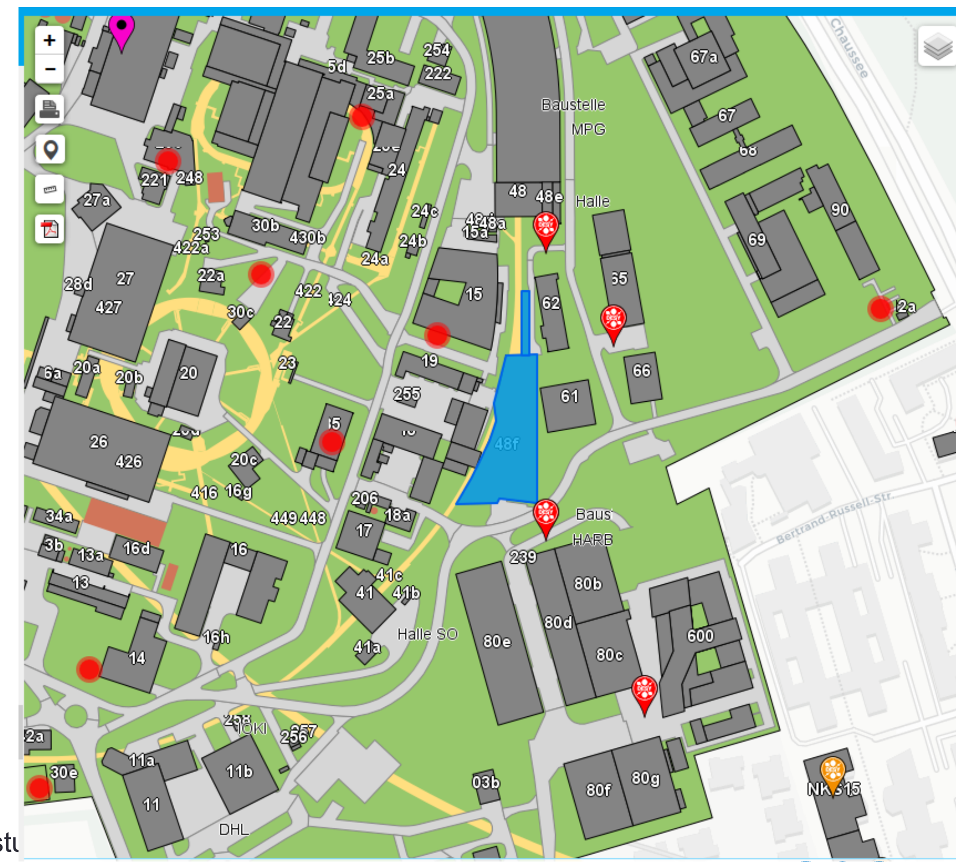
Time Tuesday 12:30 - 14:30
Thursday 8:30 - 10:00

Exkursion zu DESY am 25.6.

Beginn: 13:00

Treffpunkt: PR Point in Gebäude 48f – Ada Yonath Halle (Petra III extension)

Inhalte: DESY FS, Petra III Halle, FLASH Halle, Liquid-Jet Labor

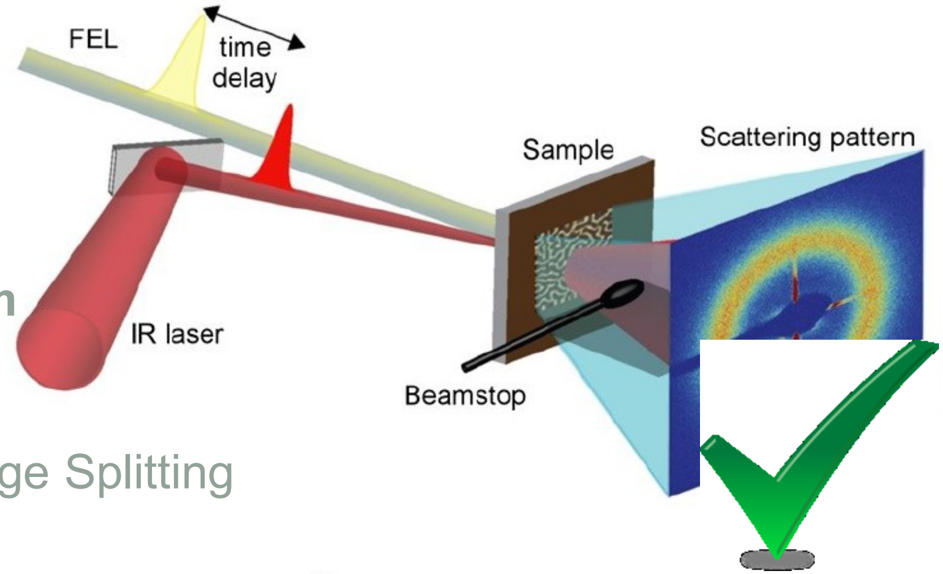


Outline

Part II/2:

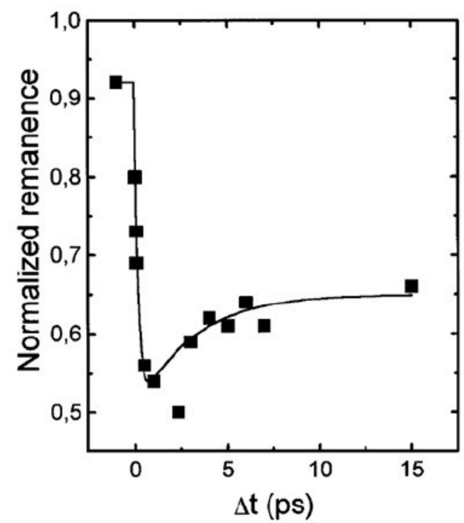
Studies on Magnetic Nanostructures

by André Philippi-Kobs (AP)



[18.6.] **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 Dicroisms
- mSAXS of Magnetic Domain Patterns



Time-resolved MOKE (Ni 120 fs)

[20.6.] 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

Femtomagnetism

1.) Ultrafast demagnetization (IR pump, **λ = 800 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, **λ = 800 nm**)

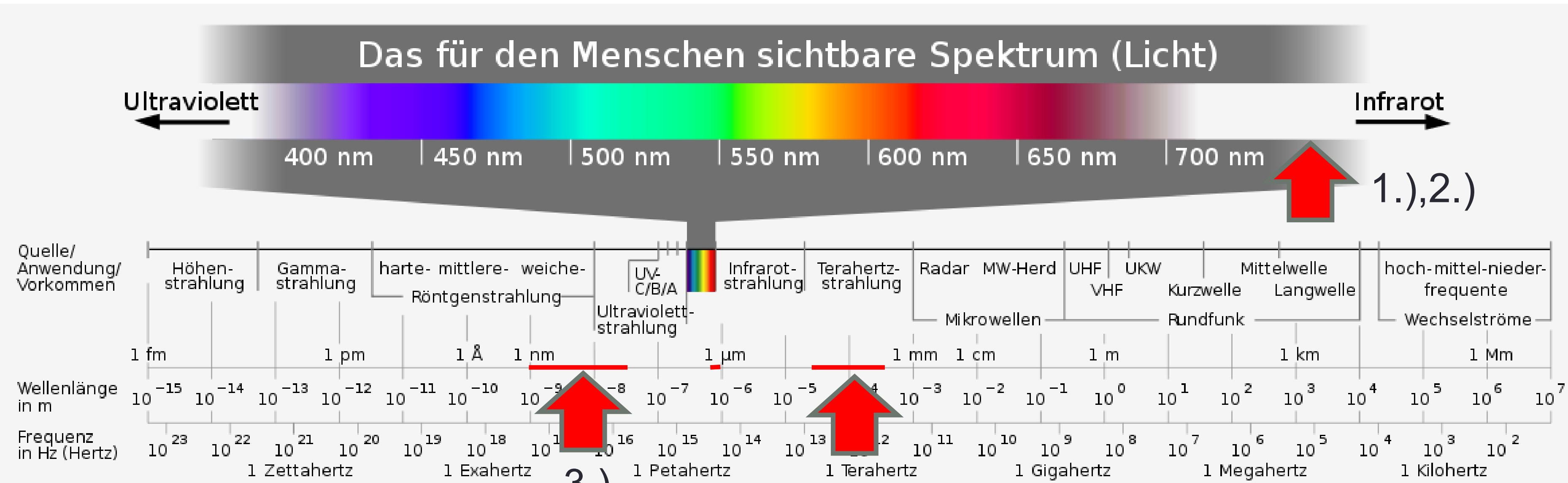
- Ferrimagnetic systems
- Is there AOS in ferromagnetic systems?

3.) High X-ray fluences (XUV pump, **λ = 20.8 nm**)

4.) THz dynamics (THz pump, **λ = 100 μm**)

Femtomagnetism

> Electromagnetic spectrum



3.)

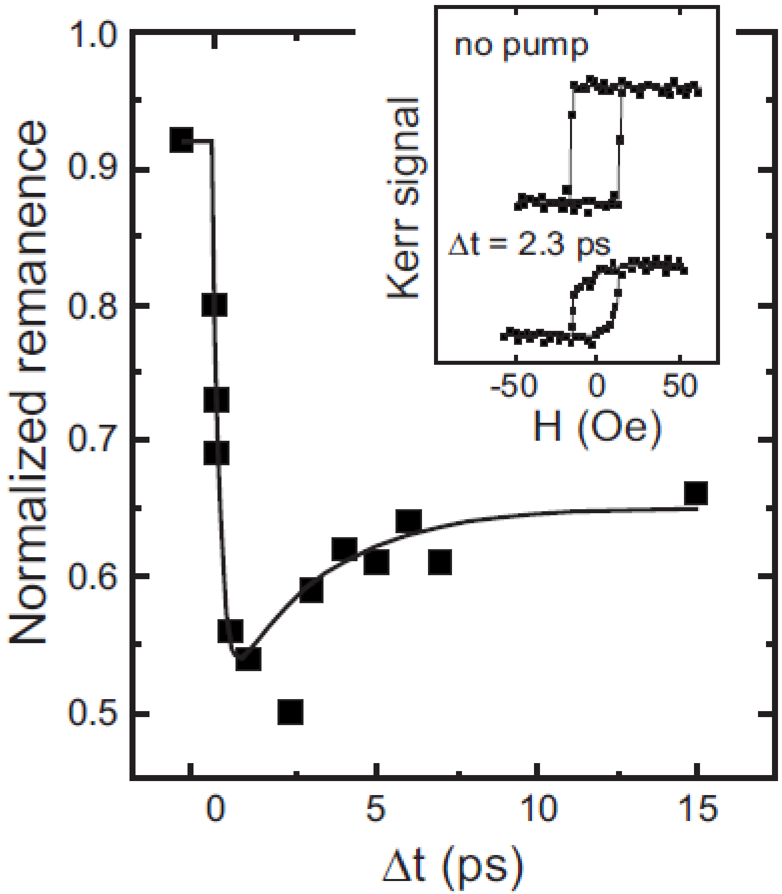
4.) THz gap (no good sources until recently)

L-edges (soft X-rays)
M-edges (extreme UV, i.e., VUV/XUV)

Femtomagnetism

> Ultrafast demagnetization (IR pump, λ = 800 nm)

- Discovery and typical parameters



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

- 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/cm² (=100 GW) ($2.8*10^{16}$ Photons/cm², $4*10^{29}$ Photons/(cm²s))

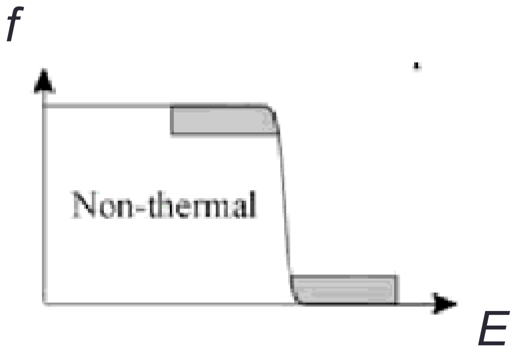
Note:
- non-destructive!
- one can easily reach multi TW
- Largest nuclear Power station 4.5 GW (thermal)

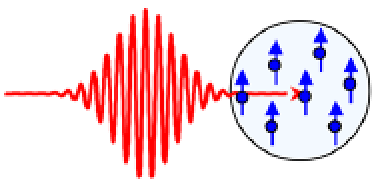
1163 citations (13.06.2019)
~100 in the last 12 Month(!)

Femtomagnetism

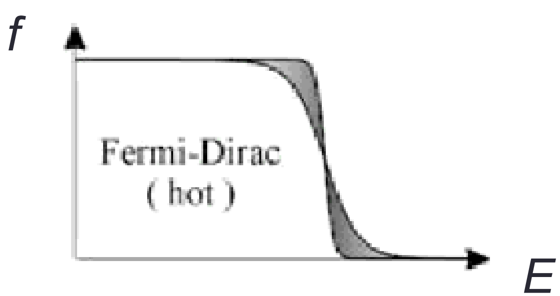
> ## Ultrafast demagnetization

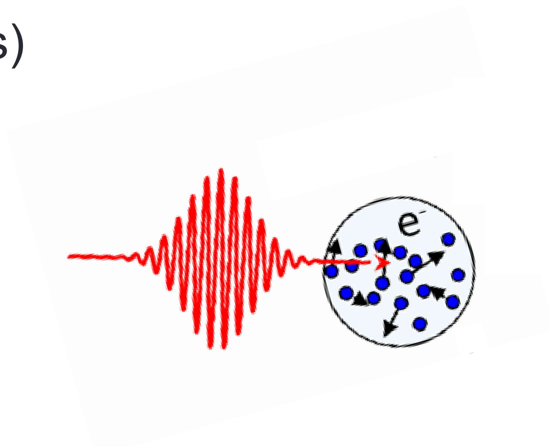
1.) Creation of highly excited electrons
during the pulse duration (< 20 fs)





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

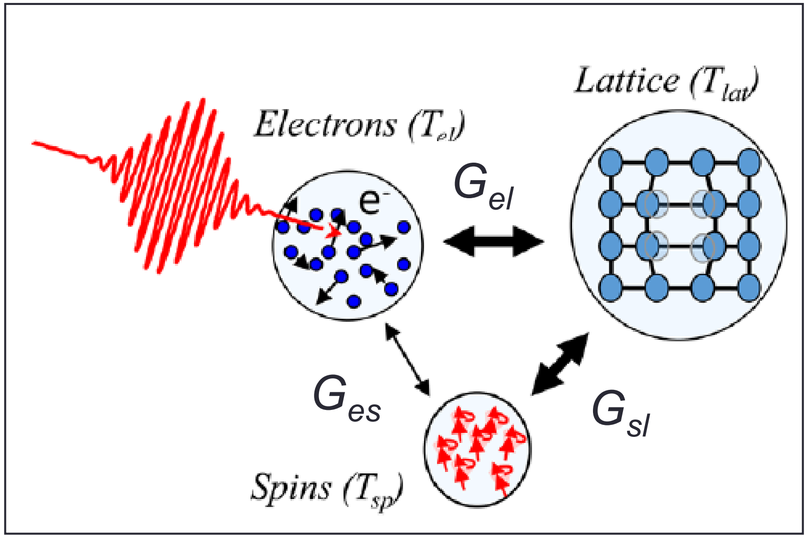


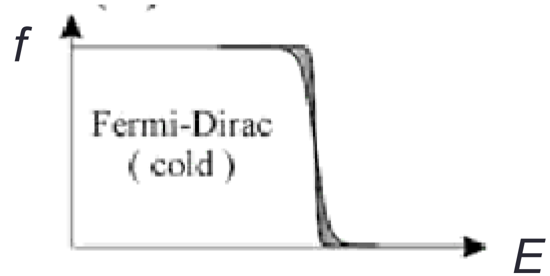


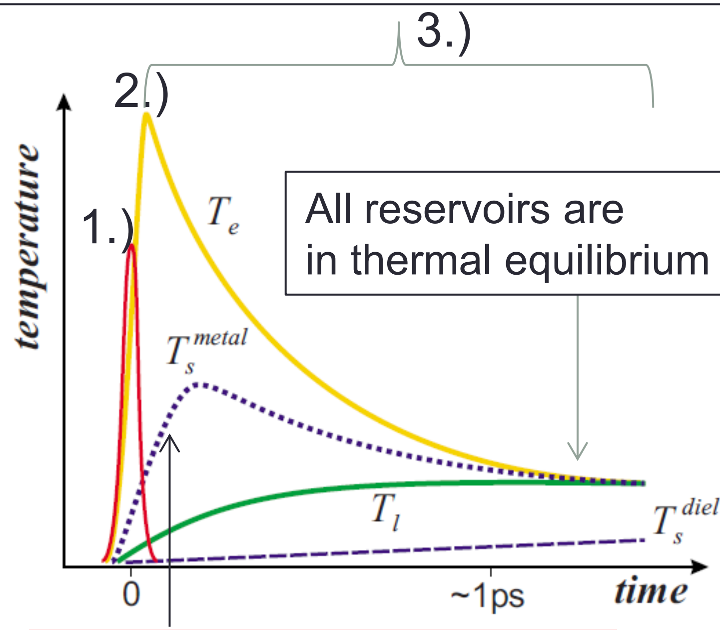
Femtomagnetism

> Ultrafast demagnetization

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







Ultrafast demagnetization

➔ Differential equations (Rate equations)

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

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

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

Energy density of pulse

C_i: specific heat, T_i: temperature of electrons, phonons, spins
G_{ij}: strength of interaction between electrons, phonons, spins

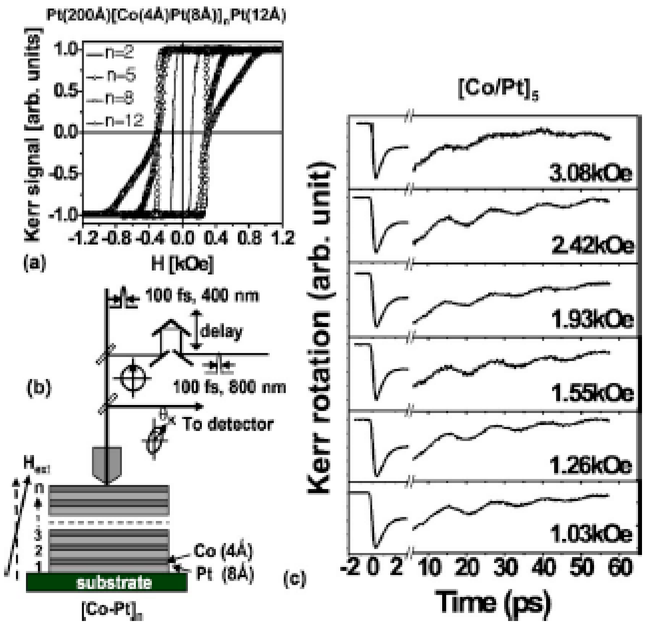
Femtomagnetism

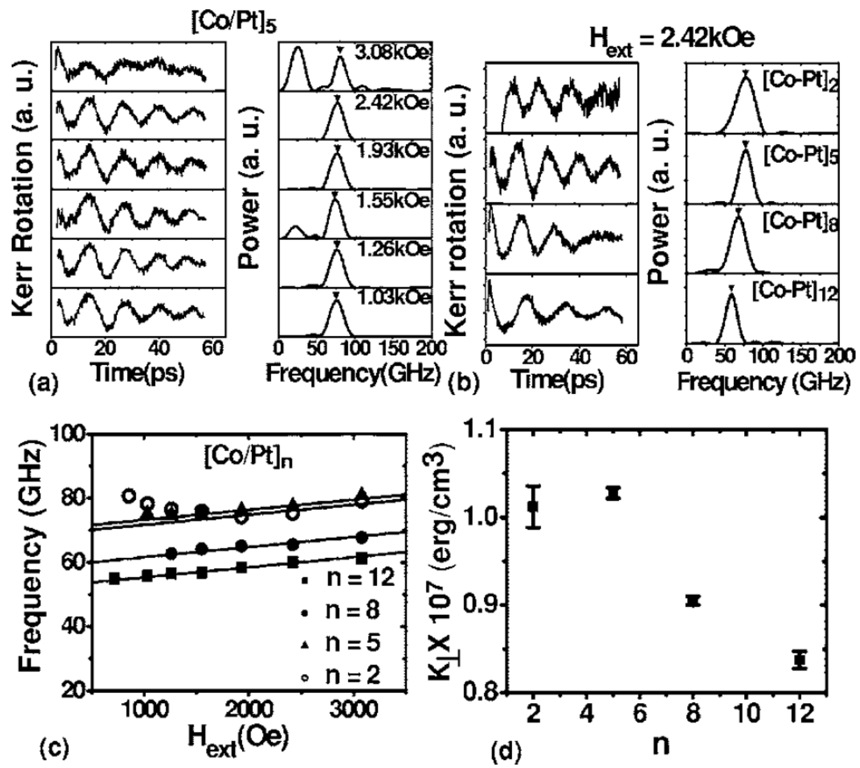
> Ultrafast demagnetization

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





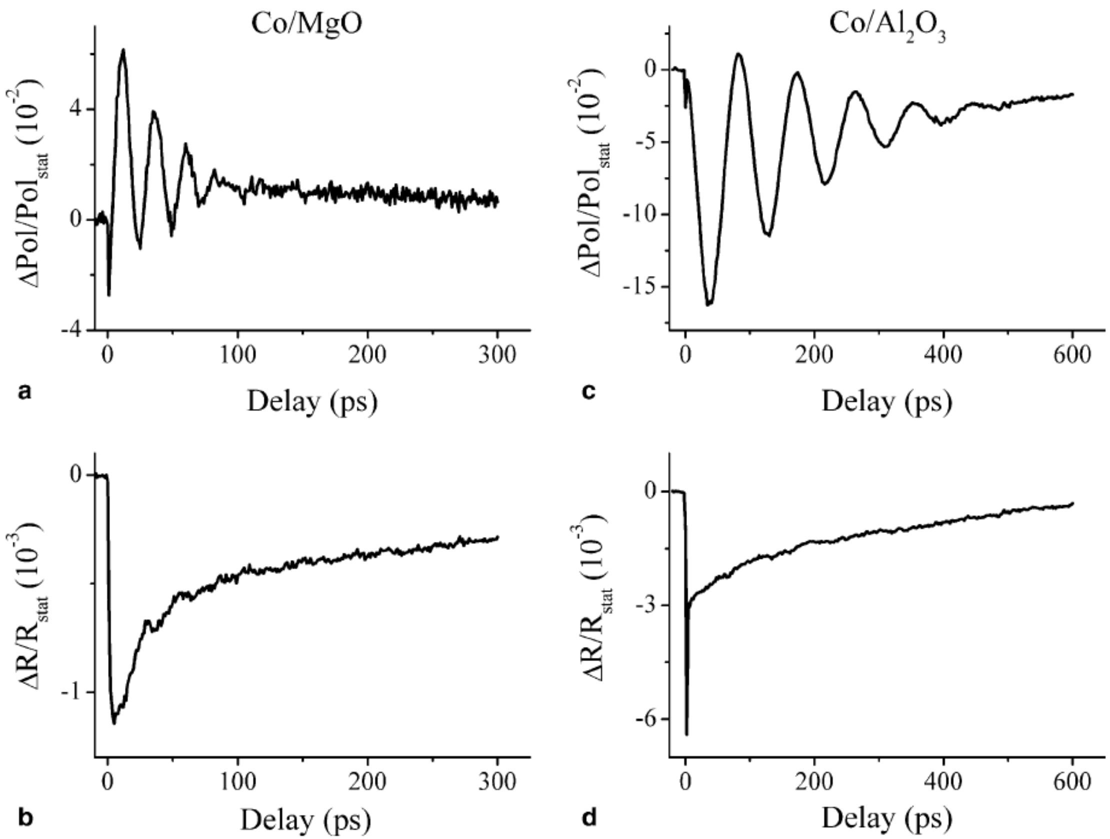
Femtomagnetism

> <u>Ultrafast demagnetization</u>

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

Chemical Physics 318 (2005) 137–146

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



Co/MgO: in-plane magnetic anisotropy

Co/Al_2O_3: out-of-plane anisotropy

Femtomagnetism

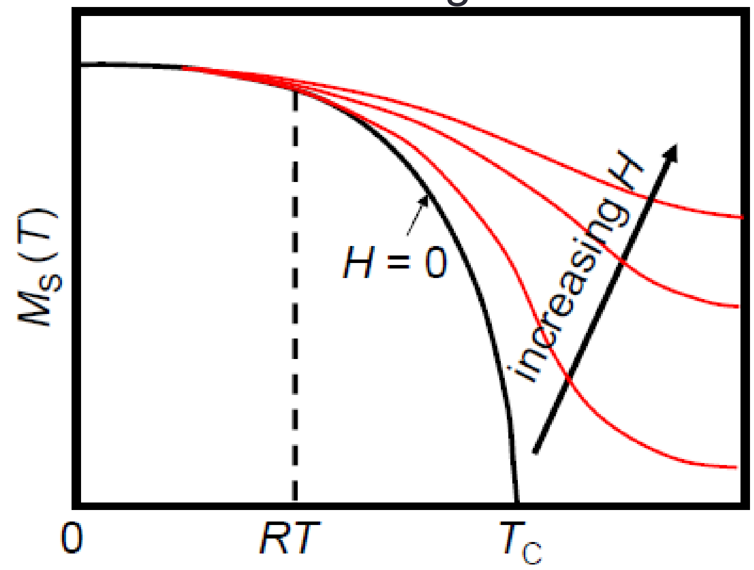
> Ultrafast demagnetization

- T-dependence of saturation magnetization (low T)

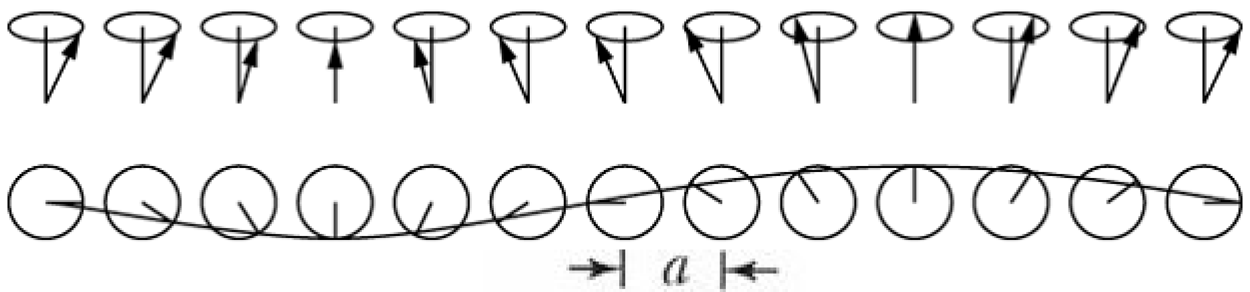
$$M_S(T) = M_S(0)(1 - BT^{3/2})$$

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

"thermal" demagnetization

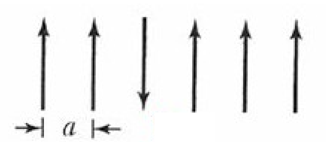


- Collective spin excitations = "magnons" or spin wave (low T)



Magnon reduces saturation magnetization by $\hbar$

- Single spin excitations (high T)



Single spin exc. reduces saturation magnetization by $\hbar$ - can be neglected

Femtomagnetism

> Ultrafast demagnetization

… of magnetic domain pattern

ARTICLE

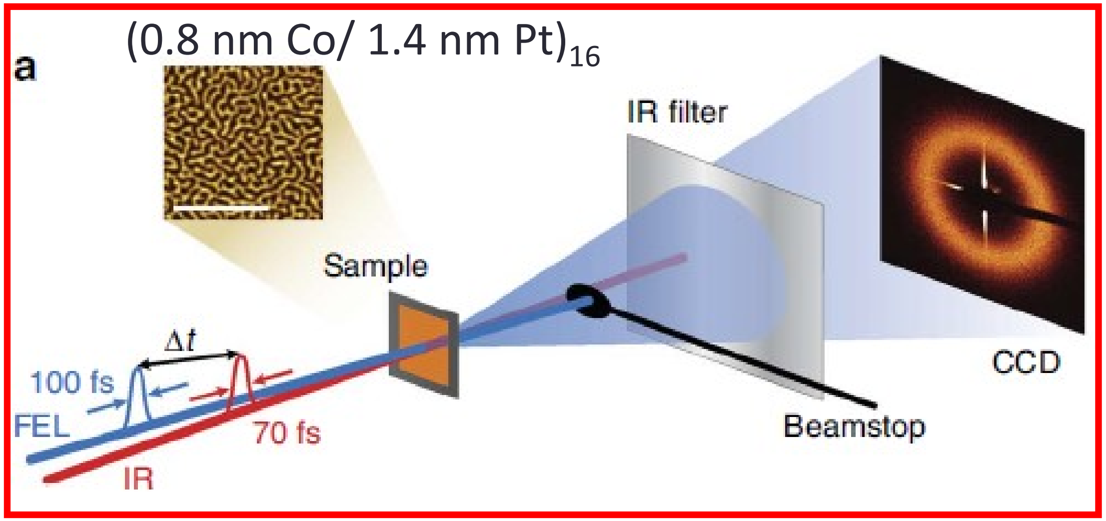
Received 18 Apr 2012 | Accepted 3 Sep 2012 | Published 2 Oct 2012

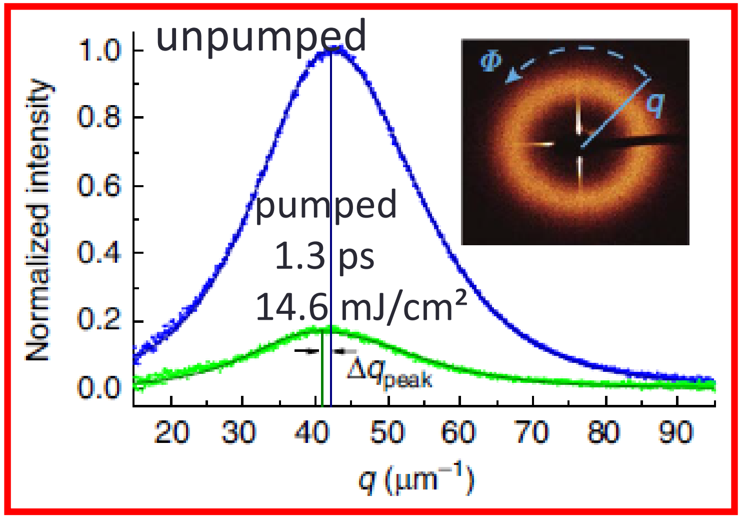
DOI: 10.1038/ncomms2108

Ultrafast optical demagnetization manipulates nanoscale spin structure in domain walls

B. Pfau[1], S. Schaffert[1], L. Müller[2], C. Gutt[2], A. Al-Shemmary[2], F. Büttner[1,3,4,5], R. Delaunay[6], S. Düsterer[2], S. Flewett[1,4], R. Frömter[7], J. Geilhufe[8], E. Guehrs[1], C.M. Günther[1], R. Hawaldar[6], M. Hille[7], N. Jaouen[9], A. Kobs[7], K. Li[6], J. Mohanty[1], H. Redlin[2], W.F. Schlotter[10], D. Stickler[7], R. Treusch[2], B. Vodungbo[6,11], M. Kläui[3,4,5], H.P. Oepen[7], J. Lüning[6], G. Grübel[2] & S. Eisebitt[1,8]

IR-pump–XUV-probe experiment

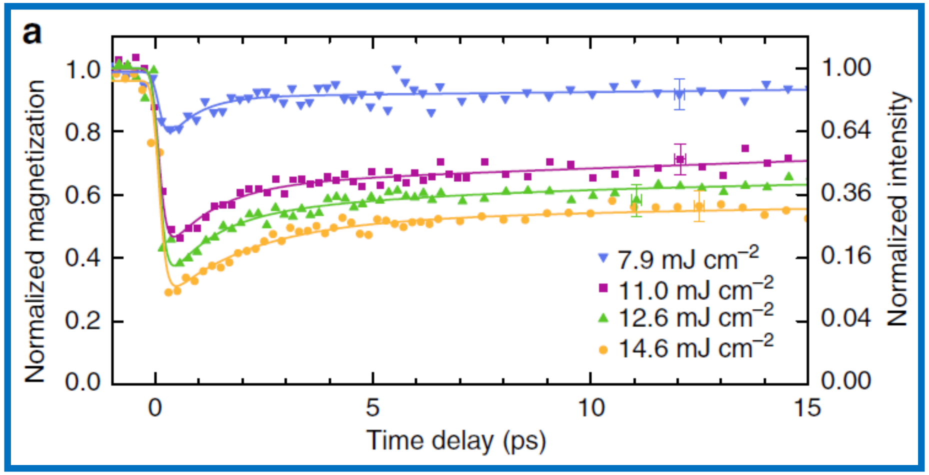




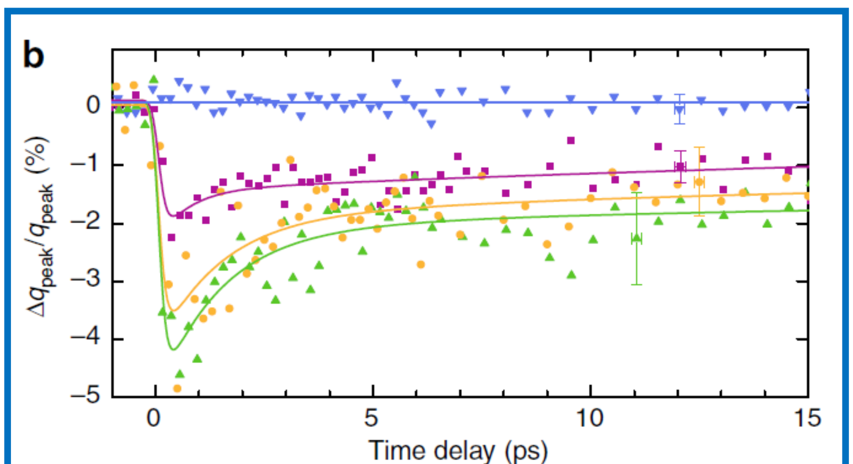
Femtomagnetism

> Ultrafast demagnetization

… of magnetic domain pattern



Ultrafast demagnetization (see above)



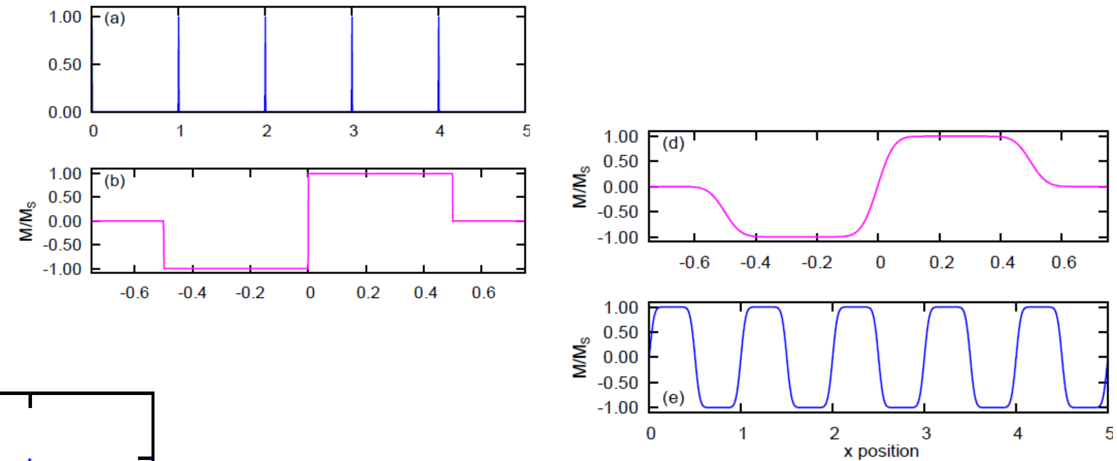
Decrease in radius of scattering ring

- Effective probed length scale seems to increase on 100fs time scale
- Necessary domain-wall speed of 10^6 to 10^7 m/s is unphysical
- Alternative explanation?

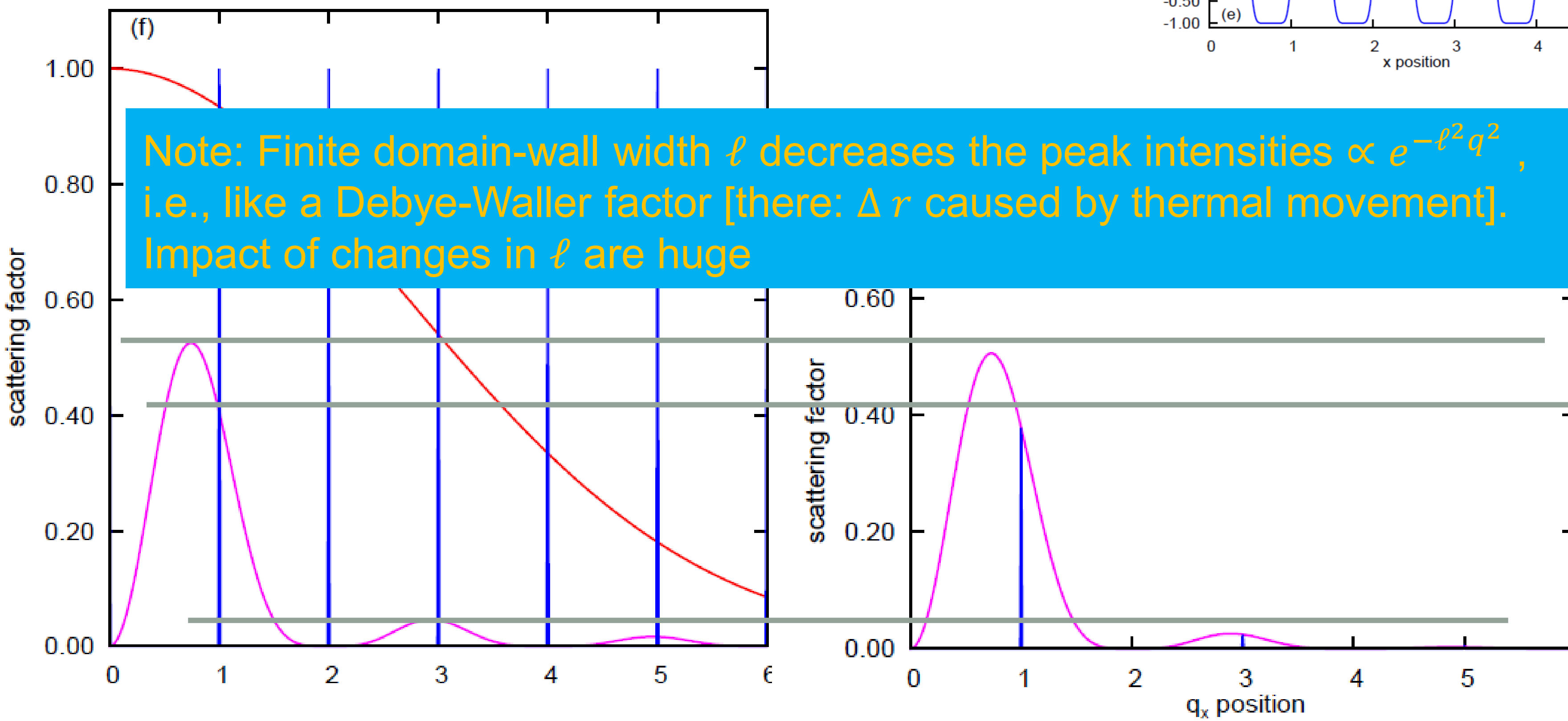
Femtomagnetism

> Ultrafast demagnetization

Recap: 1d regular domains with finte domain wall width



Note: Finite domain-wall width ℓ decreases the peak intensities $\propto e^{-\ell^2 q^2}$, i.e., like a Debye-Waller factor [there: Δr caused by thermal movement]. Impact of changes in ℓ are huge

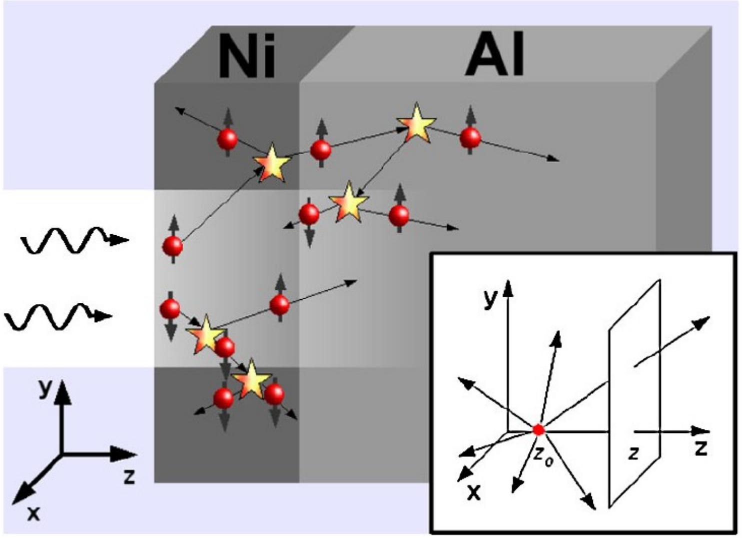


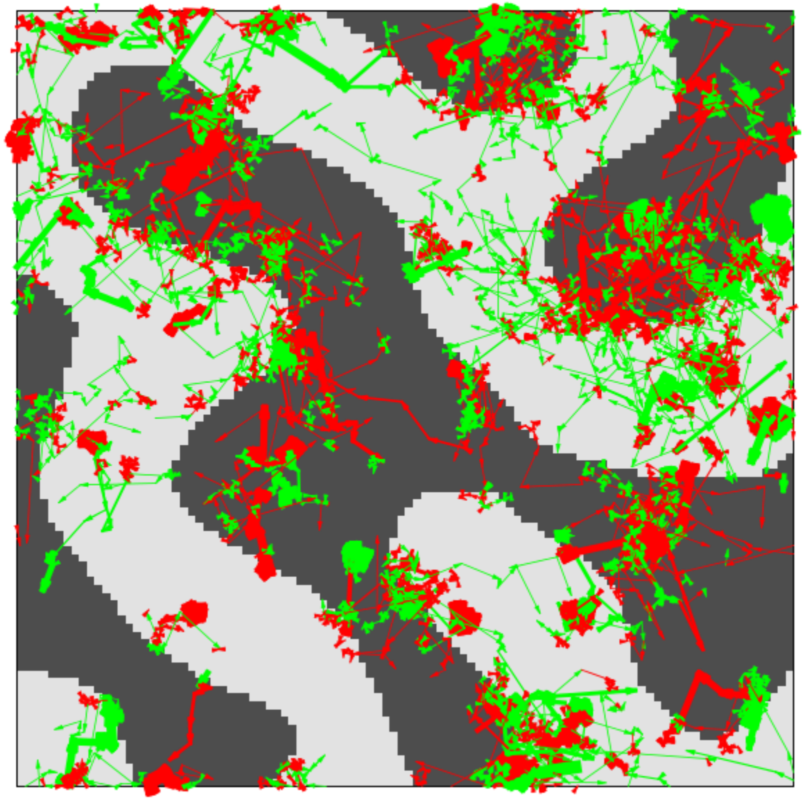
Ultrafast superdiffusive spin transport

PRL 105, 027203 (2010) PHYSICAL REVIEW LETTERS week ending 9 JULY 2010

Superdiffusive Spin Transport as a Mechanism of Ultrafast Demagnetization

M. Battiato,* K. Carva,† and P. M. Oppeneer
Department of Physics and Astronomy, Uppsala University, Box 516, SE-75120 Uppsala, Sweden
(Received 31 March 2010; published 9 July 2010)



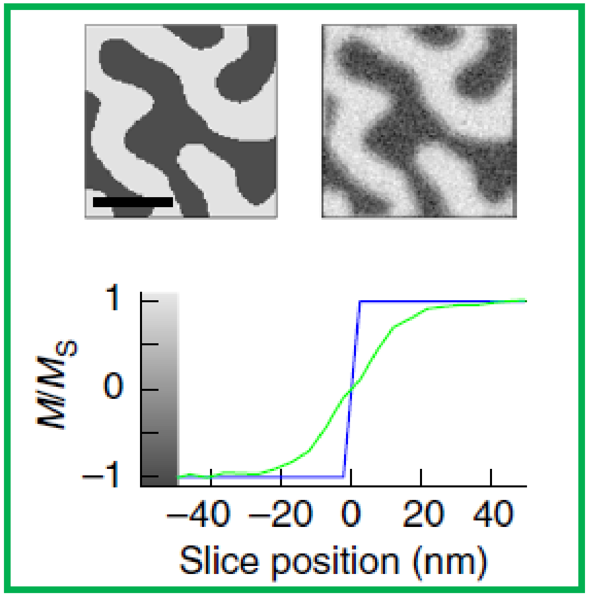


Monte Carlo simulation for a domain system using the different scattering probabilities for spin-up and spin-down electrons. When electron spin and magnetization point into the same direction the electrons are scattered less. → accumulation of „wrong-spin" electrons at the domain boundaries

Femtomagnetism

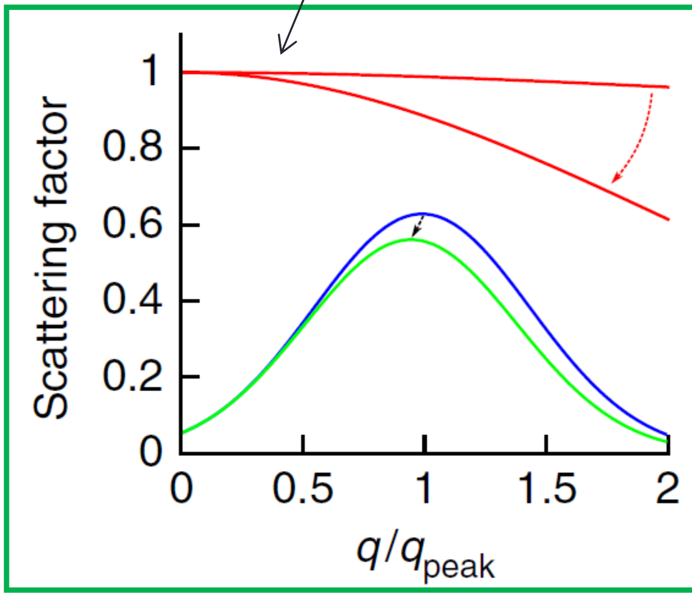
> Ultrafast demagnetization … of magnetic domain pattern

- Explanation of q_{peak} shift



Fourier transformation

Scattering form factor of domain wall



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

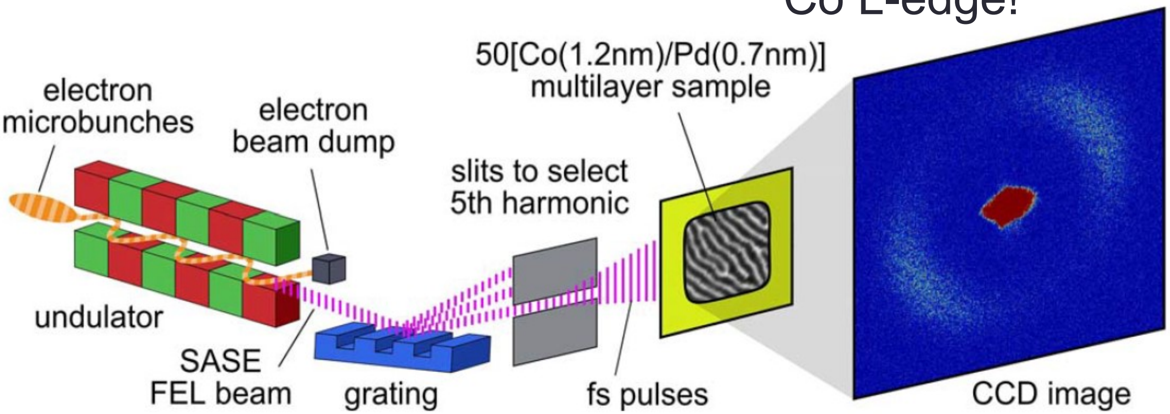
Change of scattering form factor of domain walls and hence q_{peak}

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

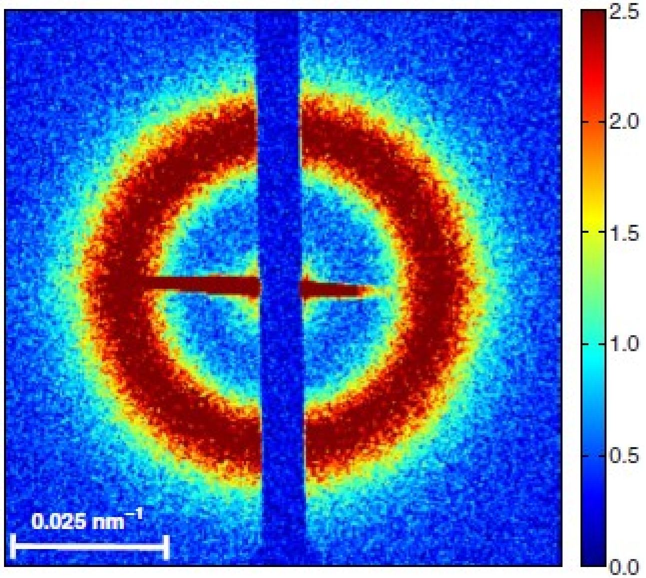
Femtomagnetism

> Ultrafast demagnetization at FEL sources – an incomplete overview

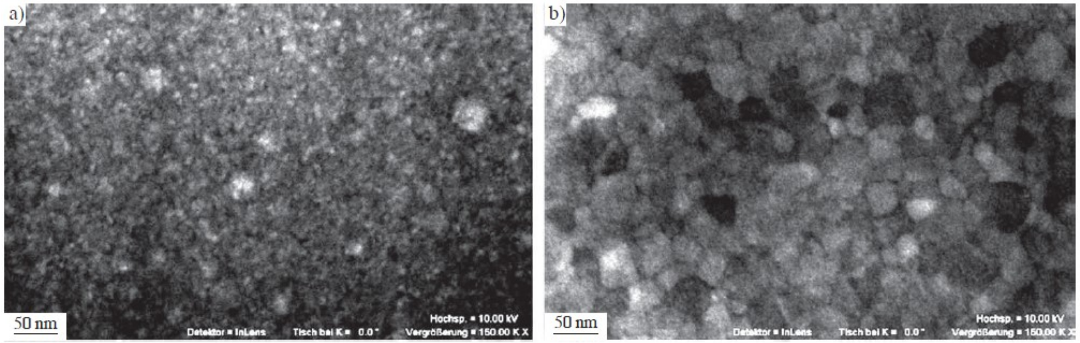
Co L-edge!



Establish first magnetic scattering at an FEL; due to experimental limitations $7 \cdot 10^3$ photons per pulse at the sample
PRB 79, 212406 (2009)



First single-shot magnetic scattering at the Co M-edge at $1.3 \cdot 10^{11}\ \mathrm{W/cm^2}$ leads to a recrystalization of the multilayer sample PRB 81, 100401(R), (2010), Sync. Rad. News 26(6), 27 (2013).



Femtomagnetism

epl A LETTERS JOURNAL EXPLORING THE FRONTIERS OF PHYSICS
January 2015
EPL, 109 (2015) 17001
www.epljournal.org
doi: 10.1209/0295-5075/109/17001

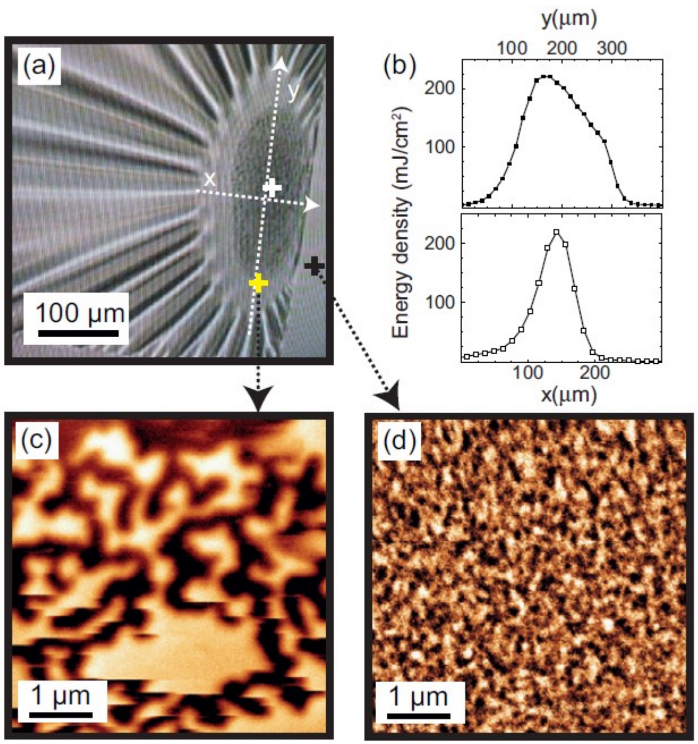
Femtosecond-laser–induced modifications in Co/Pt multilayers studied with tabletop resonant magnetic scattering

C. Weier[1], R. Adam[1], D. Rudolf[1], R. Frömter[2], P. Grychtol[3], G. Winkler[2], A. Kobs[2], H. P. Oepen[2], H. C. Kapteyn[3], M. M. Murnane[3] and C. M. Schneider[1]

> High fluence issues

- Beam damage

Fluence of 200 mJ/cm² for IR pump

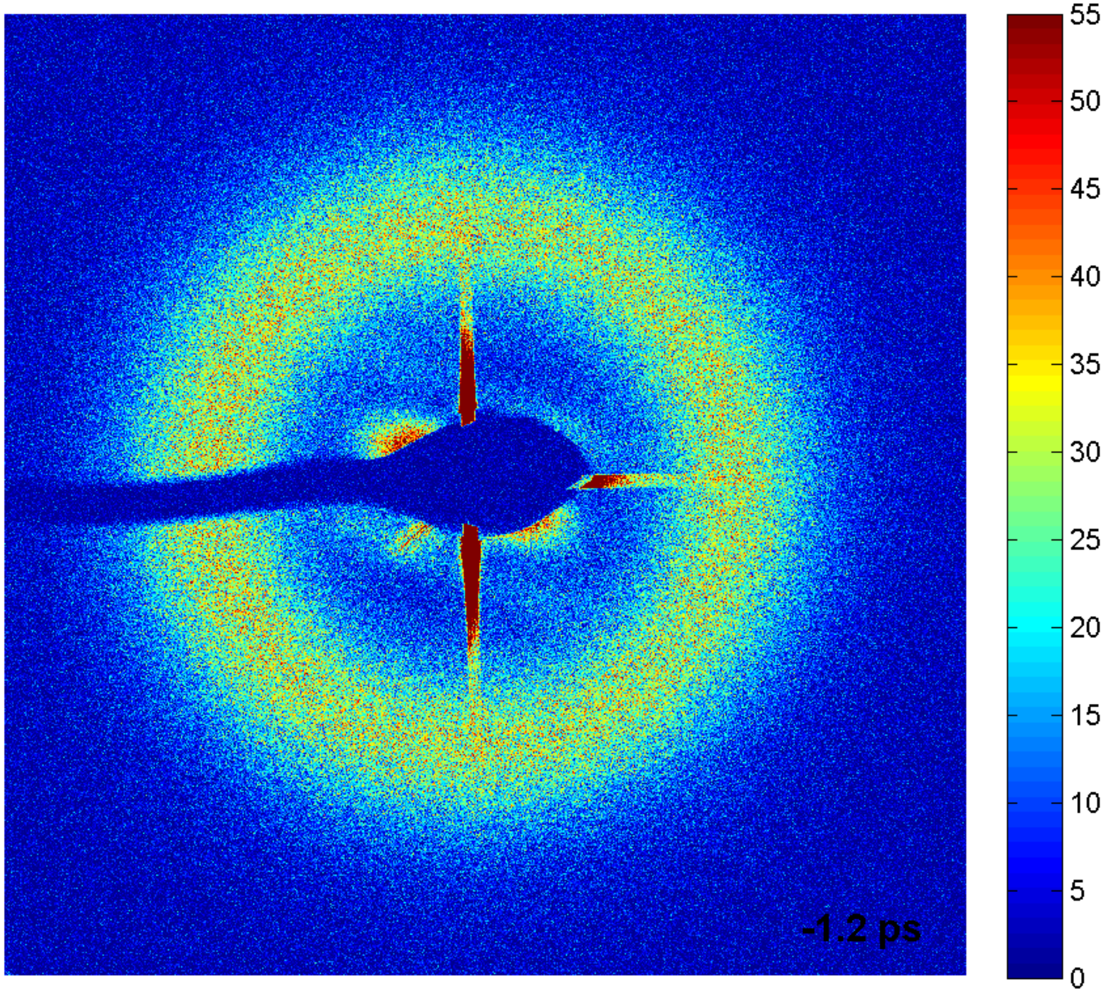


➔ Some experiments can only be done in single-shot mode (sample destroyed after one pulse)

➔ Many equivalent membranes needed

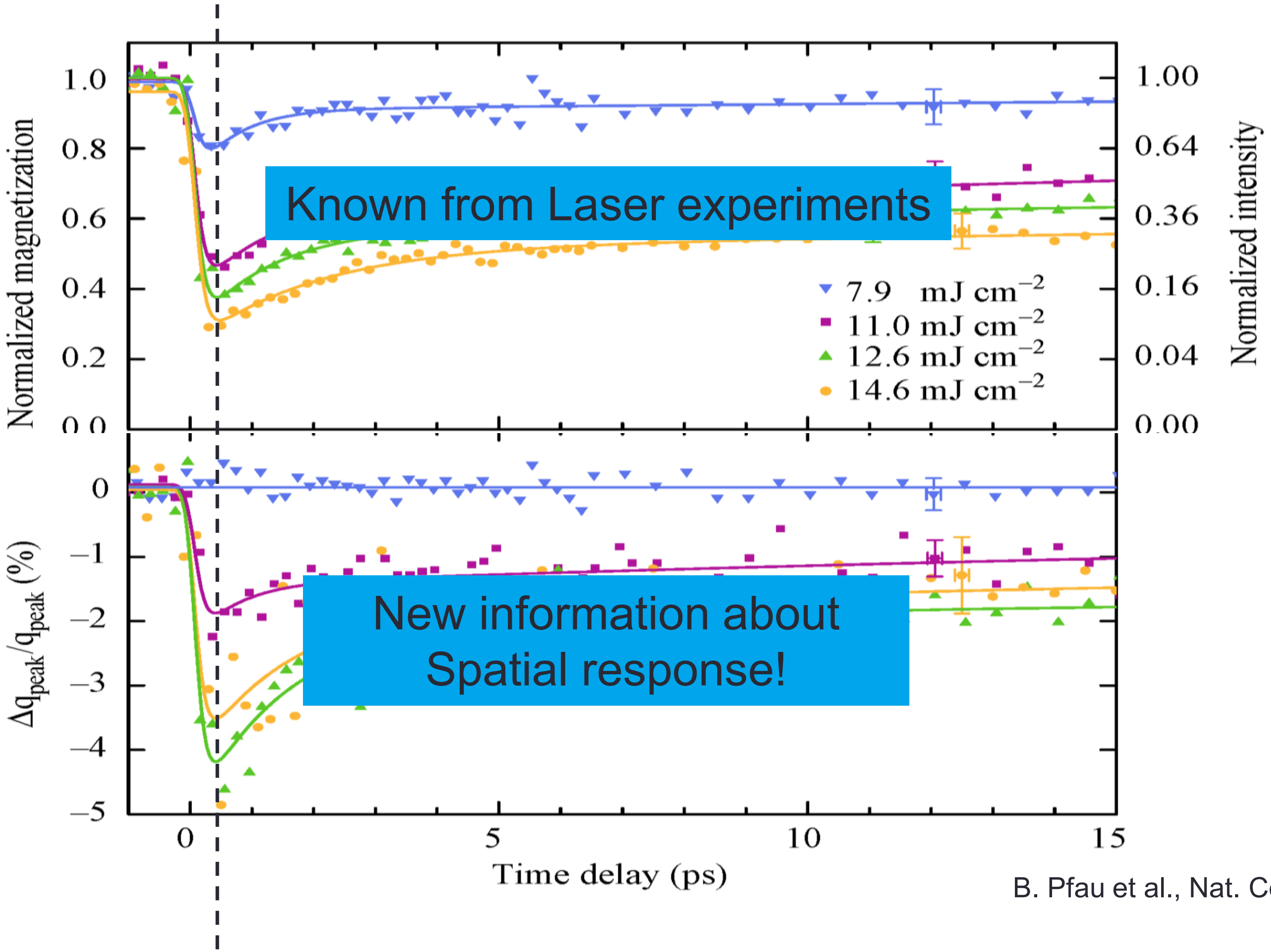
➔ Need to care also about an unintentional impact of the pump

Femtomagnetism



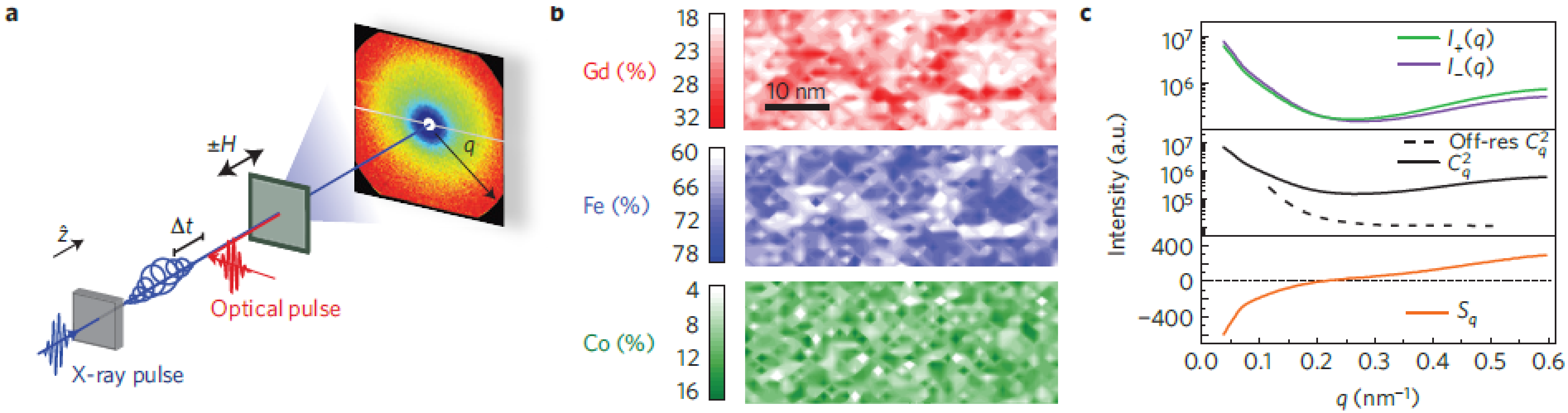
First pump-probe data (Co M-edge) taken at FLASH. Data used for
Nature Commun. 3, 1100 (2012).

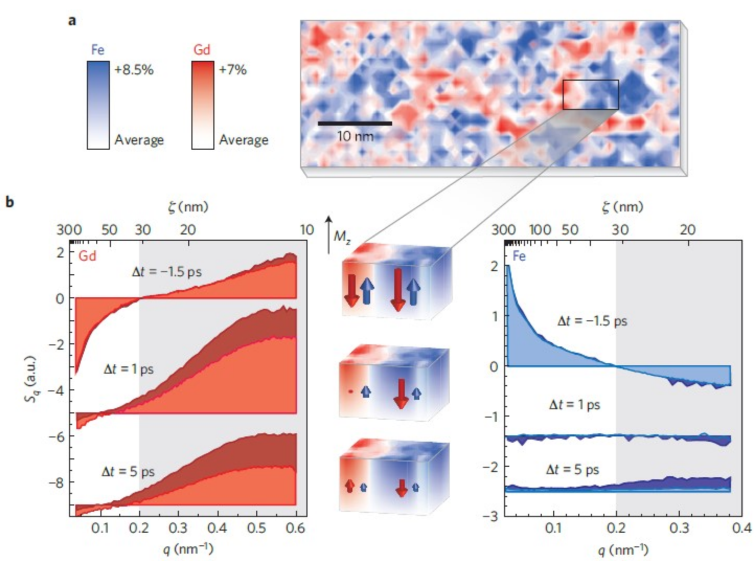
Femtomagnetism



B. Pfau et al., Nat. Commun. **3**, 1100 (2012).

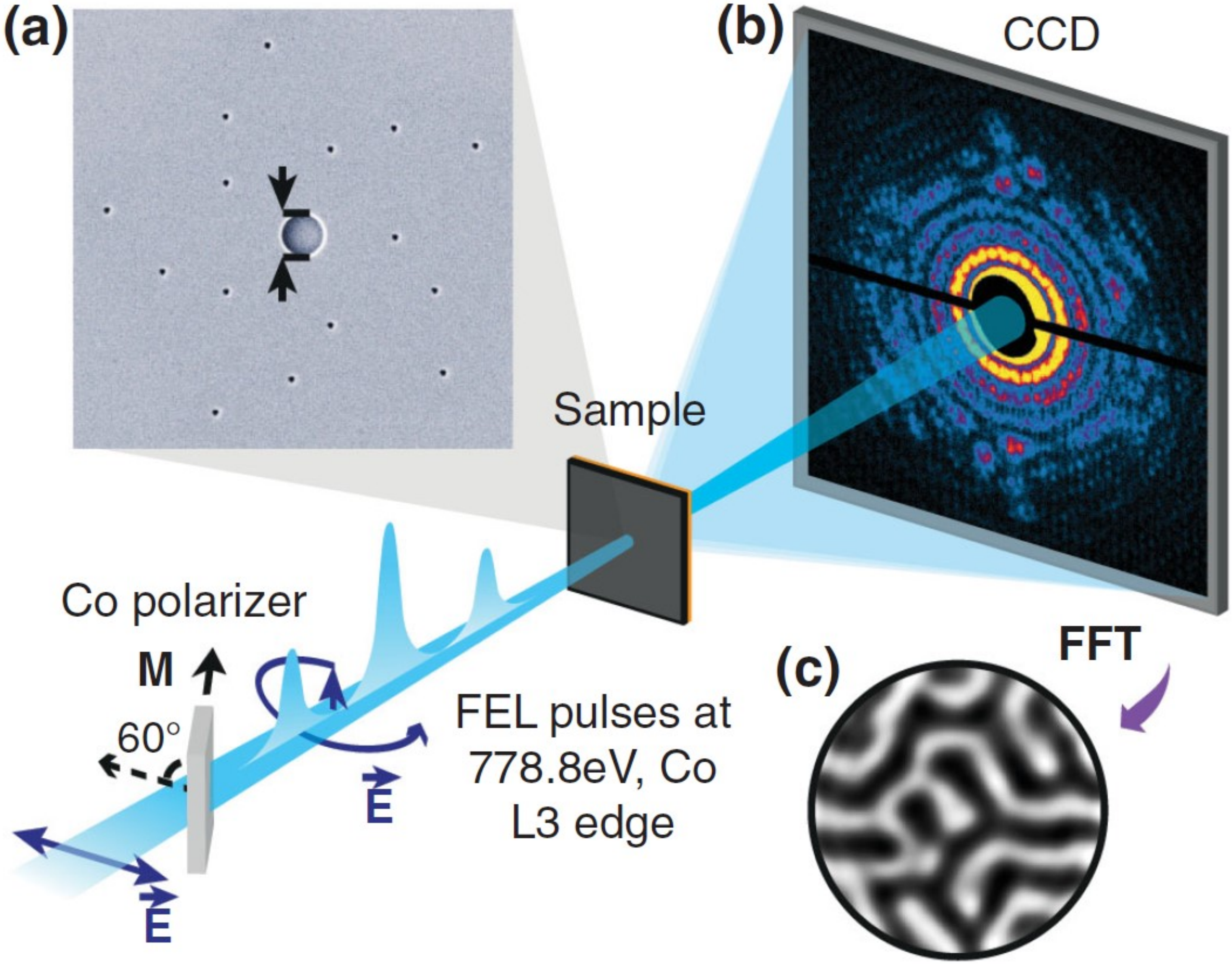
Femtomagnetism



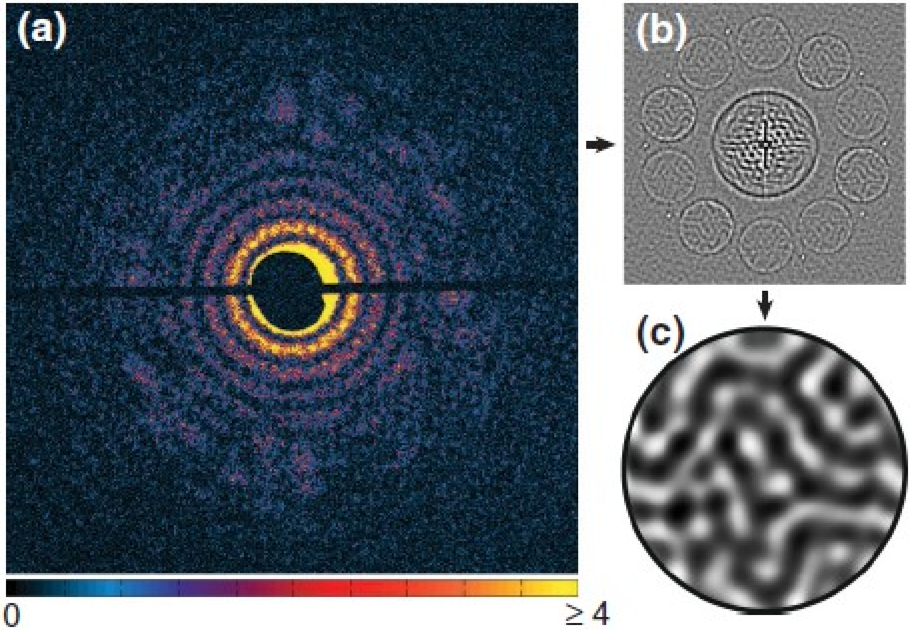


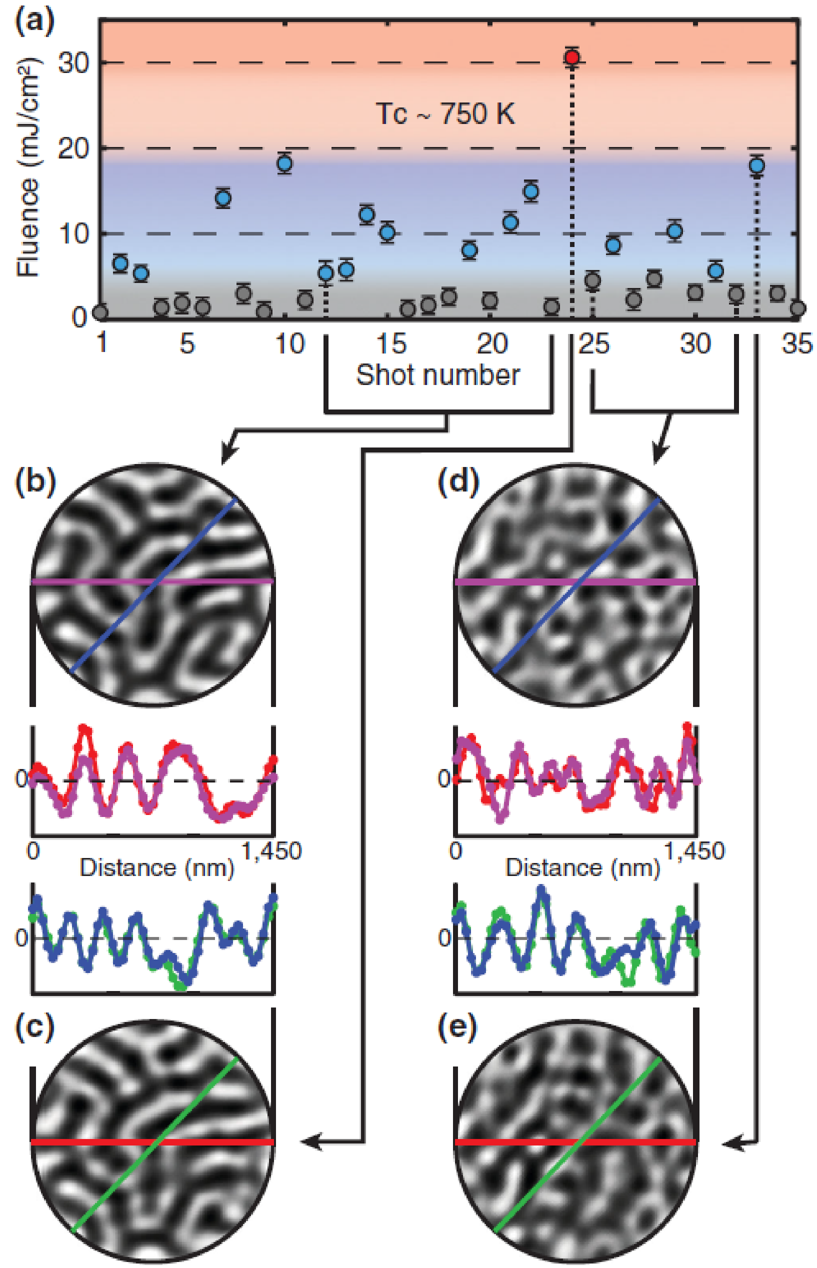
Non-local angular momentum transfere in $Gd_{24}Fe_{66.5}Co_{9.5}$ darker shaded area referes to lower pump fluence!

Nature Materials 12, 293 (2013)



First single-shot FTH holography at the Co Ledge at LCLS, PRL 108, 267403 (2012).

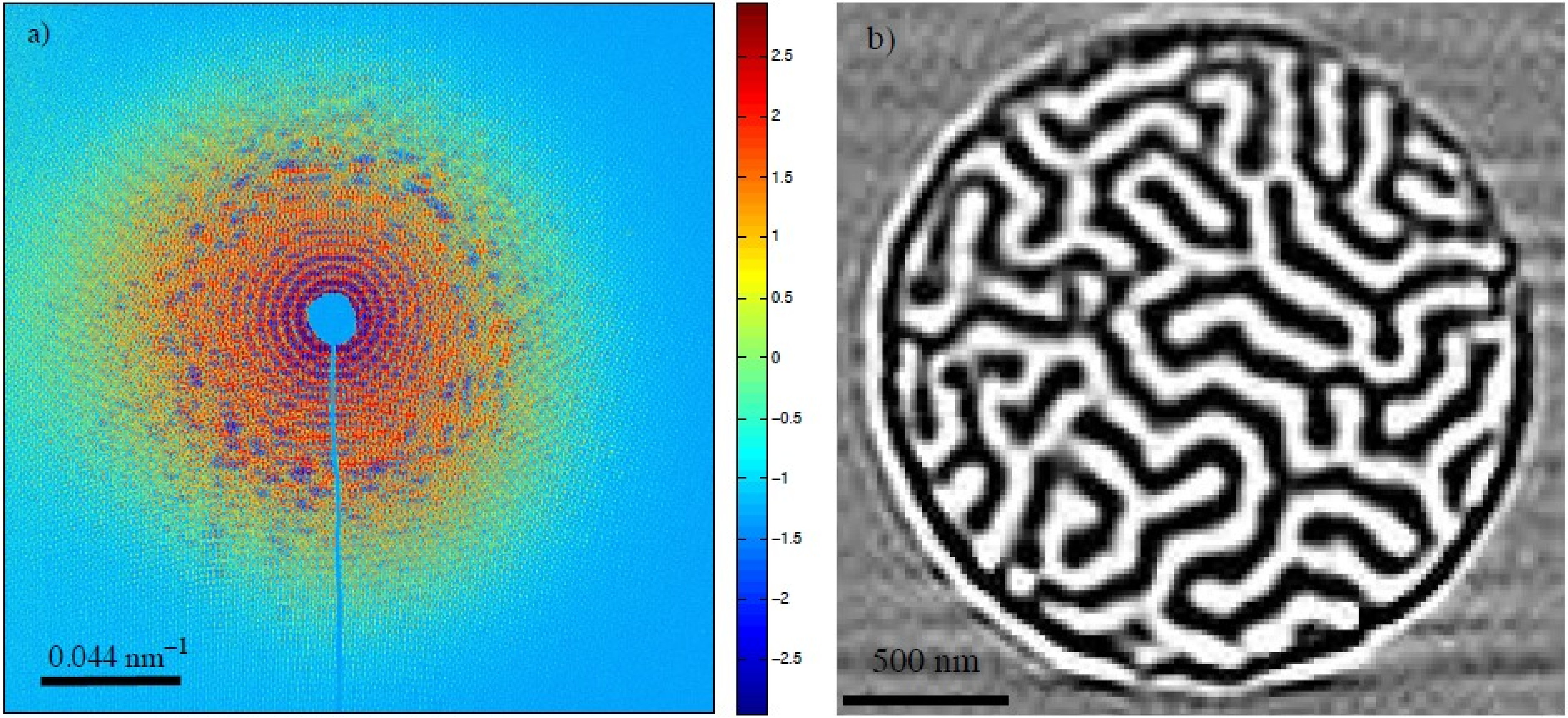




5 references give 10 reconstructions which can be averaged to improve image quality (left, c). The Hologram contains only $1.5 \cdot 10^5$ Photons. Threshold for successful imaging is $5\,\frac{\mathrm{mJ}}{\mathrm{cm}^2}$. Particularly intense shots give good reconstructions but change or destroy the sample

PRL 108, 267403 (2012).

Femtomagnetism

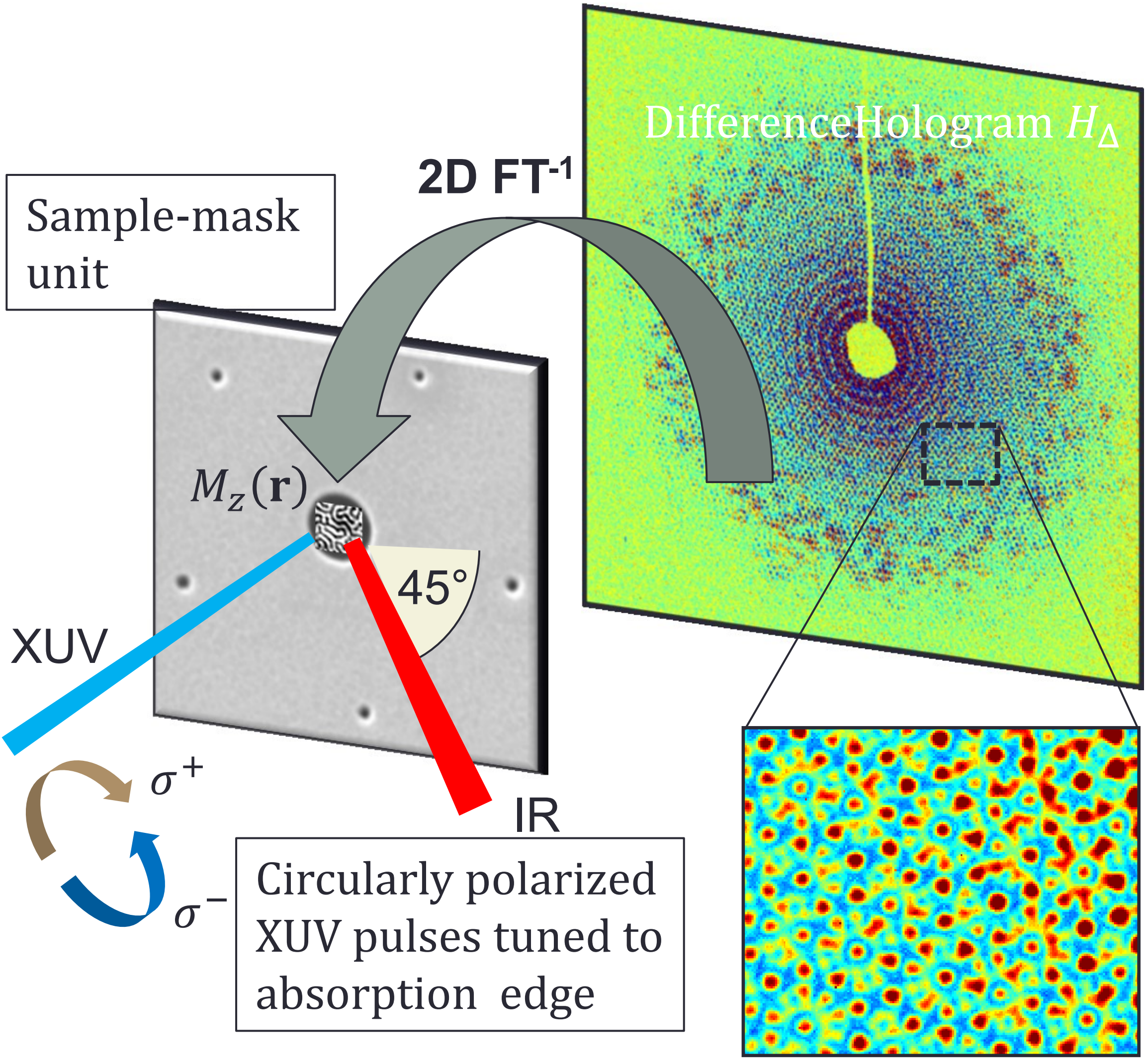


First multi-shot holography at the Co M-edge at FERMI ~13000 shots per helicity, sign preserving logscale for the hologram

Sync. Rad News 26(6), 27 (2013).

Femtomagnetism

> Ultrafast demagnetization … of magnetic domain pattern -



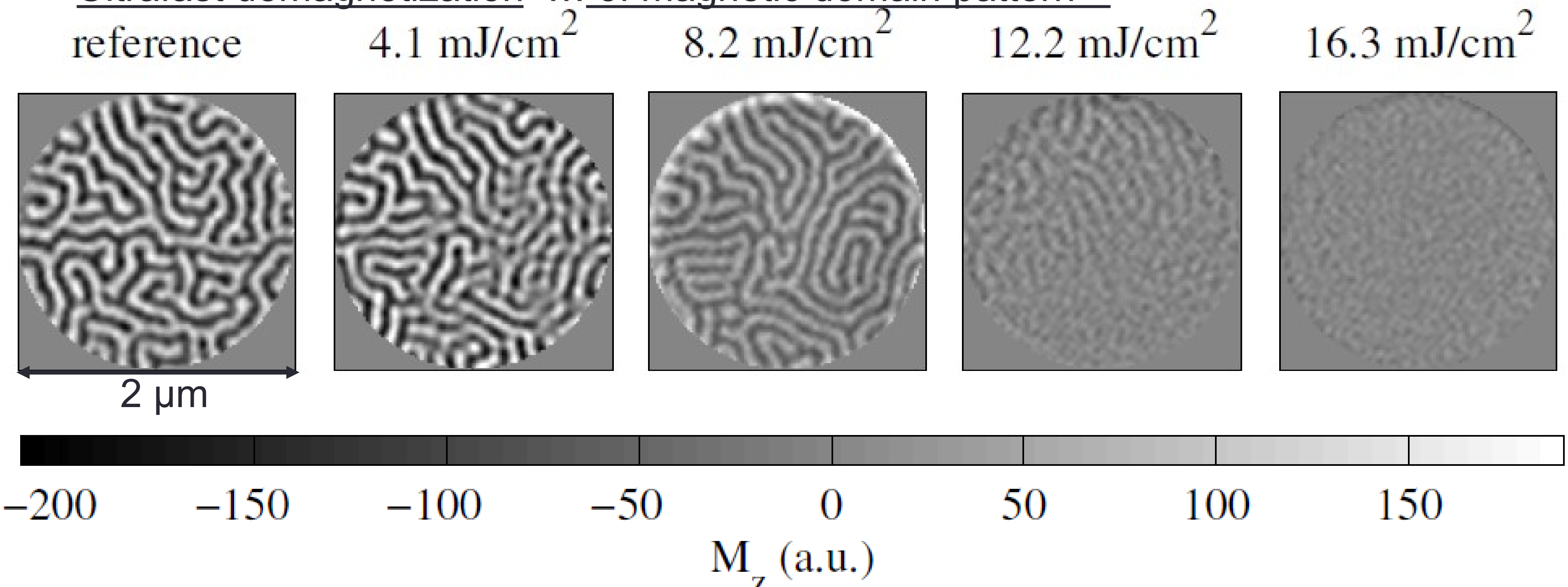
Experimental parameters at FERMI

- ~10x600 shots per helicity (@1 µJ due to extremely instable conditions of the FEL at the time
- 4.1 mJ/cm² ≤ IR fluence ≤ 16.3 mJ/cm²
- Repetition rate 10 Hz
- λ = 20.8 nm (M-edge of Co)
- Fixed time delay of 1 ps
- Pump laser (λ = 780 nm) at 45° with respect to sample surface
- Sample: $(Co(0.4\ nm)/Pd(0.2\ nm))_{30}$

unpublished

Femtomagnetism

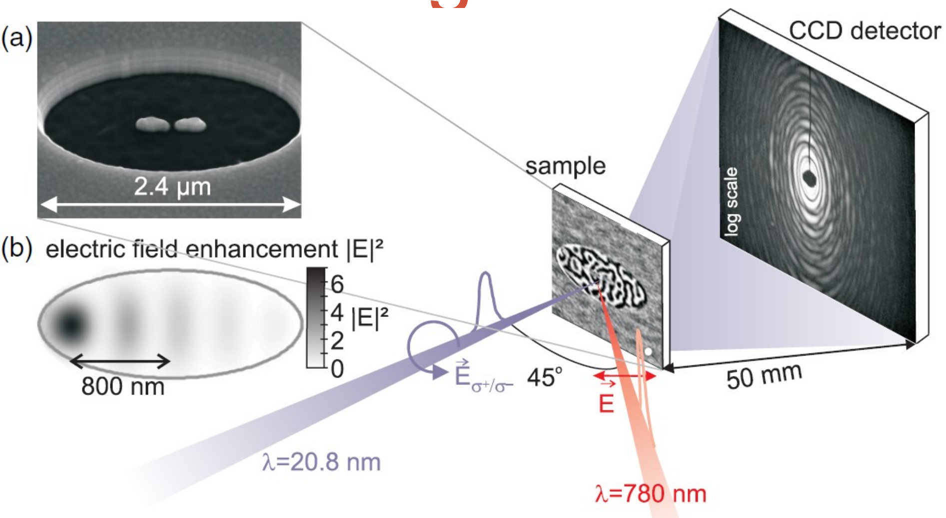
> Ultrafast demagnetization … of magnetic domain pattern -



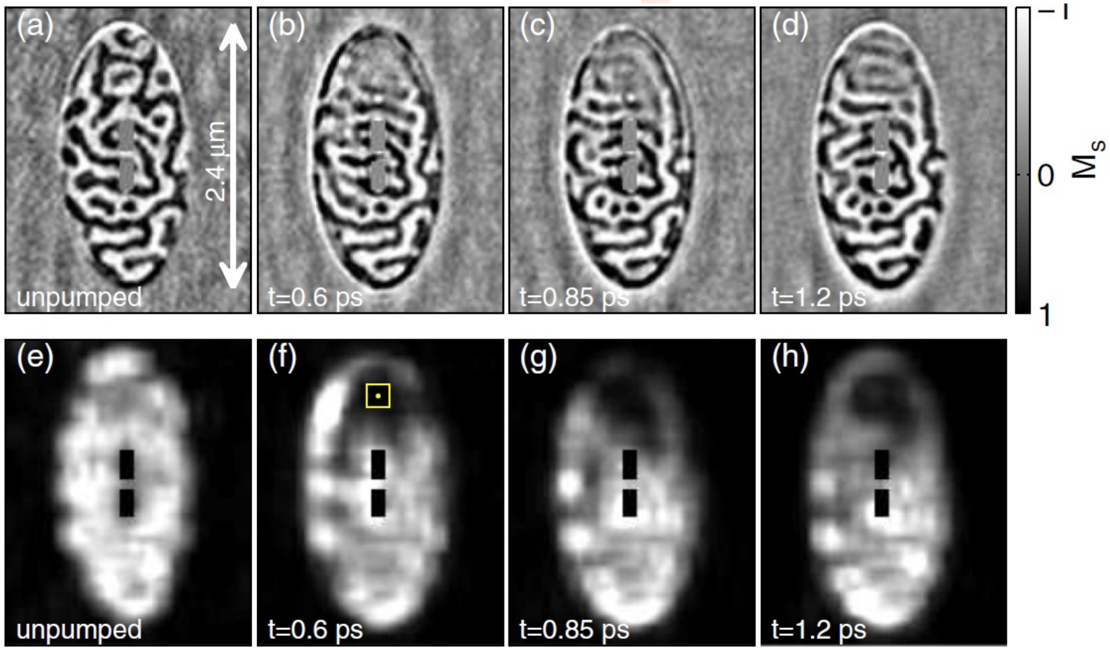
- Blurring due to domain rearrangements initiated by quasi-static heating (thermal demagnetization)
- Global contrast scale $\propto$ saturation magnetization ➔ ultrafast demagnetization on the nanoscale
- Small contrast at high fluences due to high demagnetization and/or domain rearrangements
- Resolution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

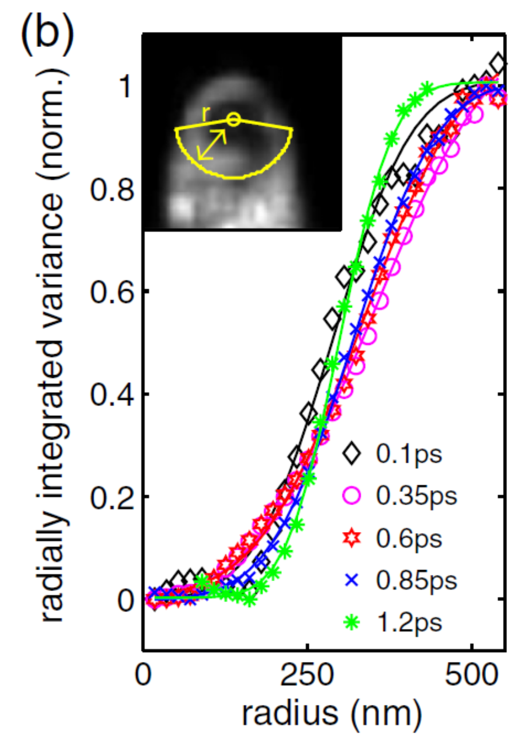
unpublished

Femtomagnetism



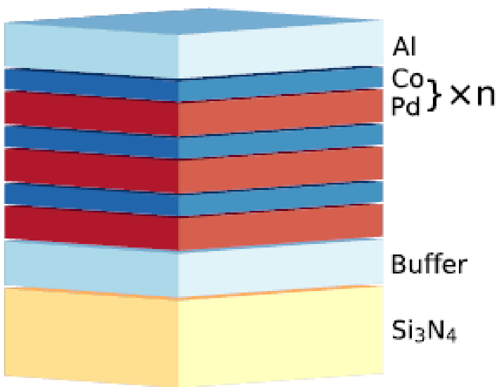
Holography with spatially confined pump via plamonic antennas on the sample surface and object hole geometry



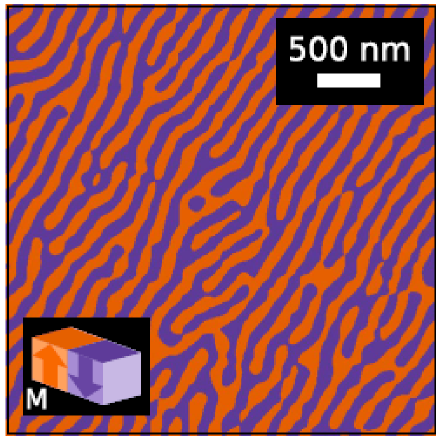


Femtomagnetism

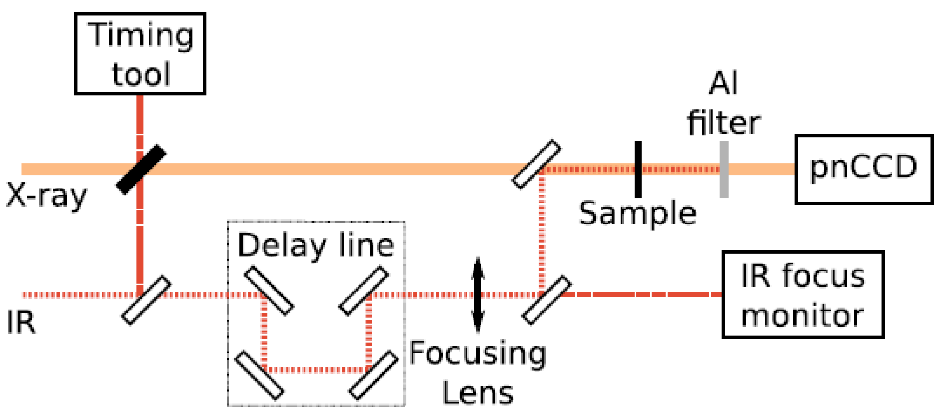
(a) IR opaque top layer



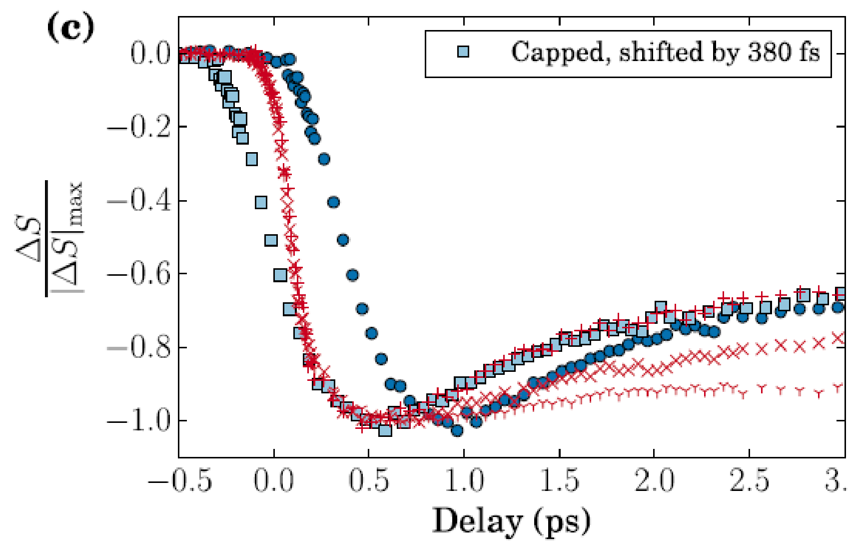
(b)



(c)



Pump probe measurement using IR excited electrons created in a IR opaque top layer to demagnetize the sample



Magnetization drop-off is extended due to pulse broadening as electrons have to difuse into the magnetic layer

Sci. Report 6, 18970 (2016)

Femtomagnetism

PRL 110, 234801 (2013) PHYSICAL REVIEW LETTERS week ending 7 JUNE 2013

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

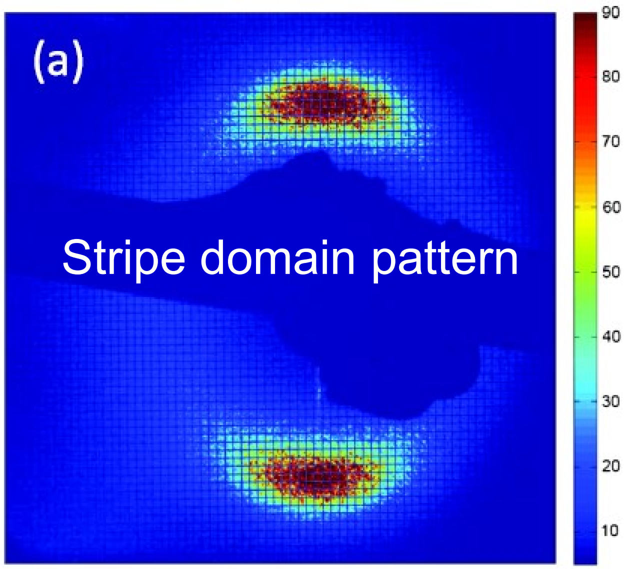
L. Müller,[1,*] C. Gutt,[1,2] B. Pfau,[3] S. Schaffert,[3] J. Geilhufe,[4] F. Büttner,[3] J. Mohanty,[3] S. Flewett,[3] R. Treusch,[1] S. Düsterer,[1] H. Redlin,[1] A. Al-Shemmary,[1] M. Hille,[5] A. Kobs,[5] R. Frömter,[5] H. P. Oepen,[5] B. Ziaja,[1,2,6,7] N. Medvedev,[1,6] S.-K. Son,[1,6] R. Thiele,[1,6]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

> High X-ray fluences

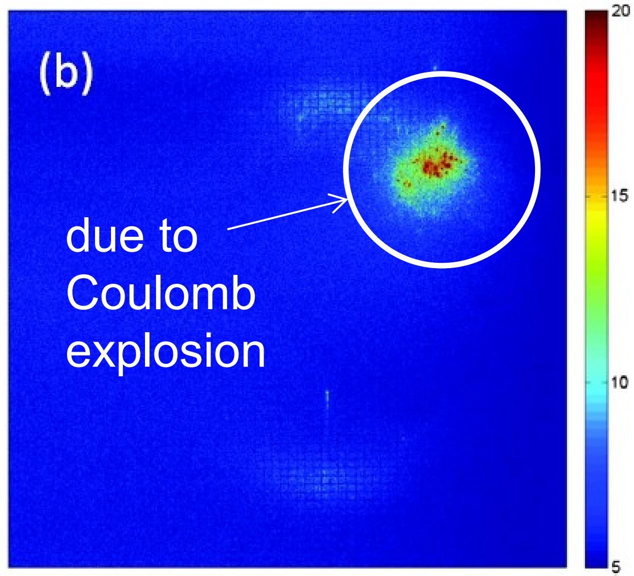
What is happening at high X-ray fluences (λ = 20.8 nm, i.e., M-edge of Co)?

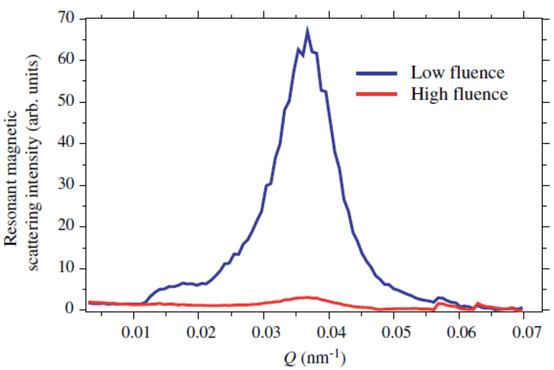
Experiment at FLASH (**F**ree-electron **Las**er in **H**amburg),pulse duration of ~100 fs

1000 shots à 7.5 mJ/cm²



1 shot à 5 J/cm²





➔ Intra-pulse “quenching“/ breakdown of the resonant mSAXS signal

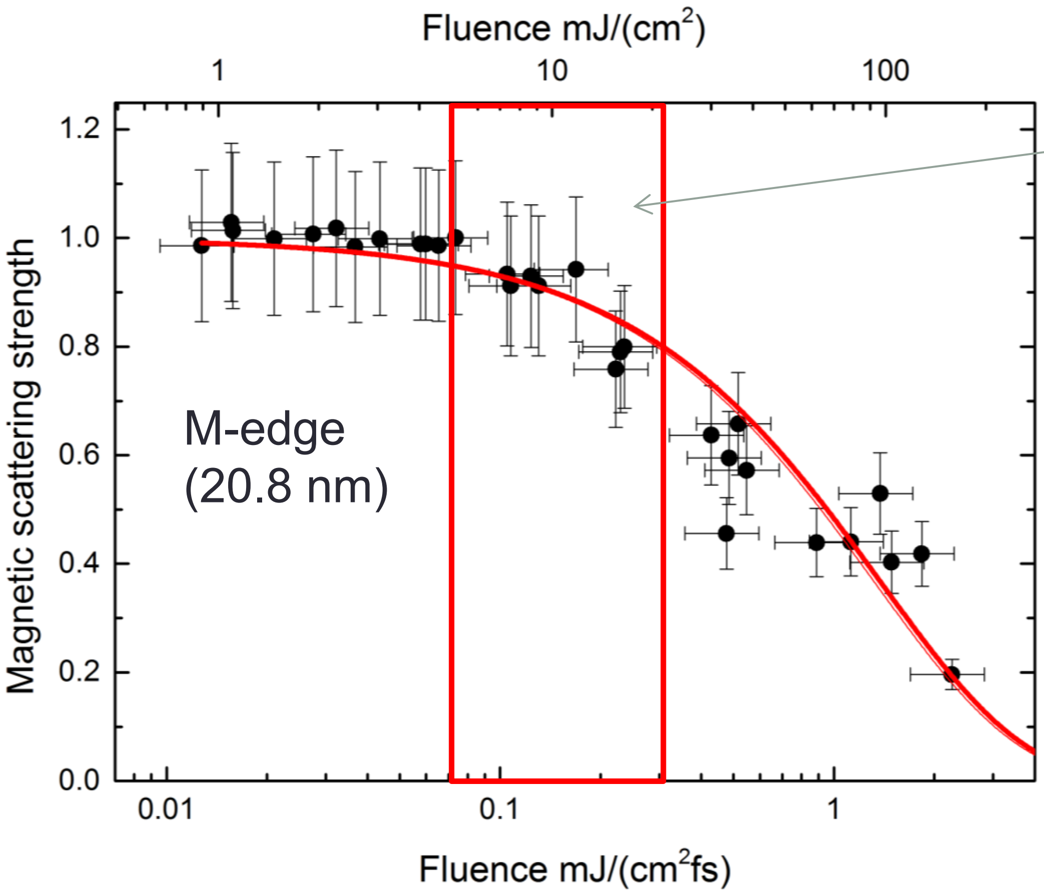
➔ Violation of principle “diffract before destruct“

➔ X-ray pulse <u>does not</u> only act as a non-invasive probe

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!

➔ Quenching efect sets in at unexpectedly low fluences

Femtomagnetism

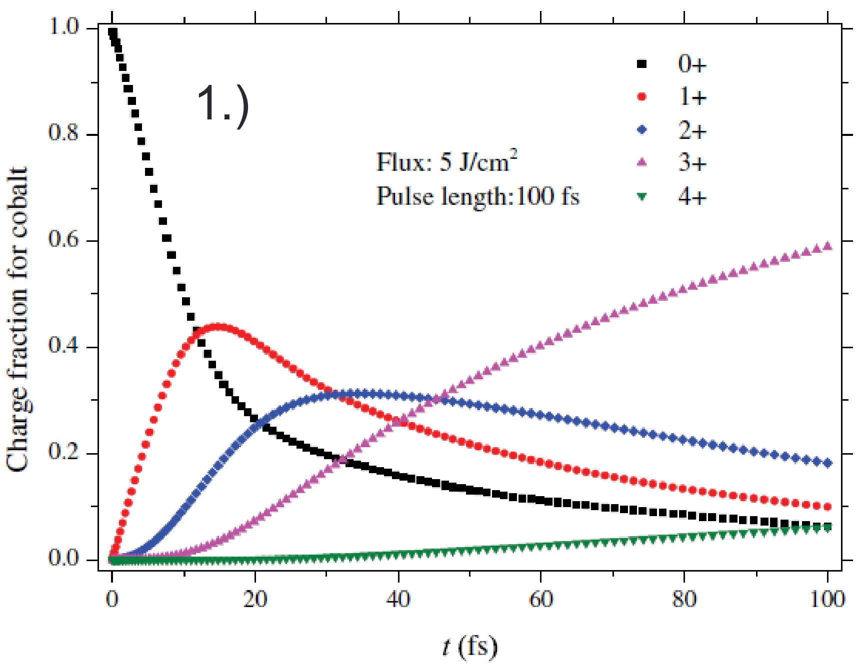
PRL 115, 107402 (2015) PHYSICAL REVIEW LETTERS week ending 4 SEPTEMBER 2015

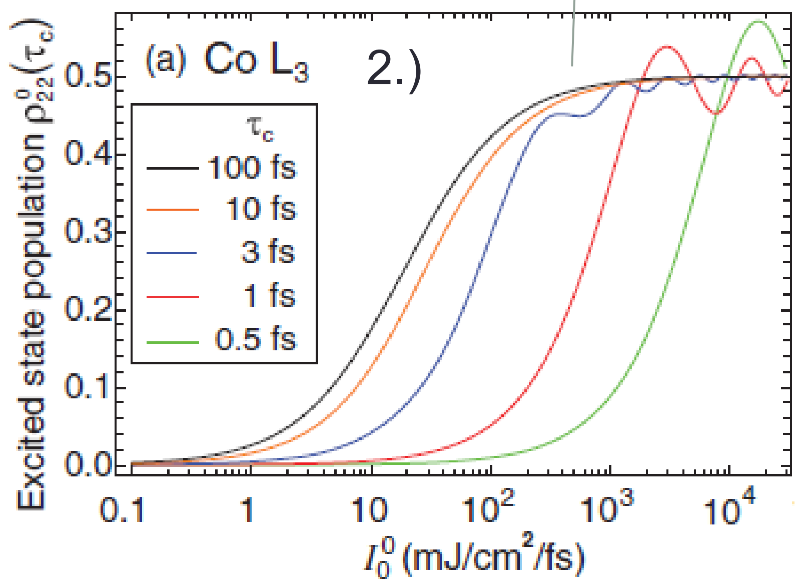
Creation of X-Ray Transparency of Matter by Stimulated Elastic Forward Scattering

J. Stöhr[1,*] and A. Scherz[2,†]

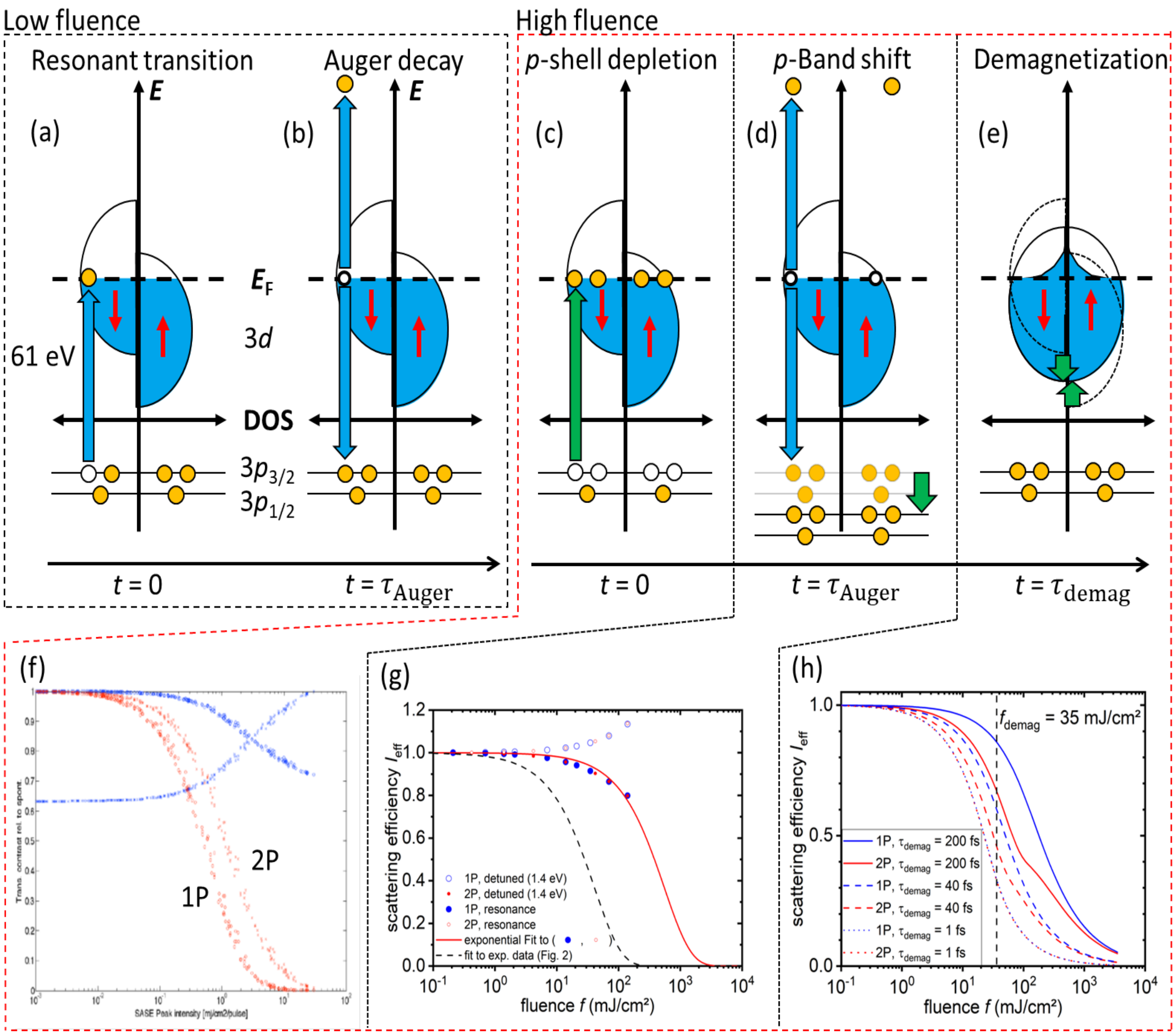
> <u>High X-ray fluences</u>

Possible reasons for quenching (ongoing research)
1.) Creation of highly ionized state and changing band-structure
2.) Stimulated elastic forward scattering
3.) Ultrafast demagnetization





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



tion seems to
cattering intensity!

Femtomagnetism

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

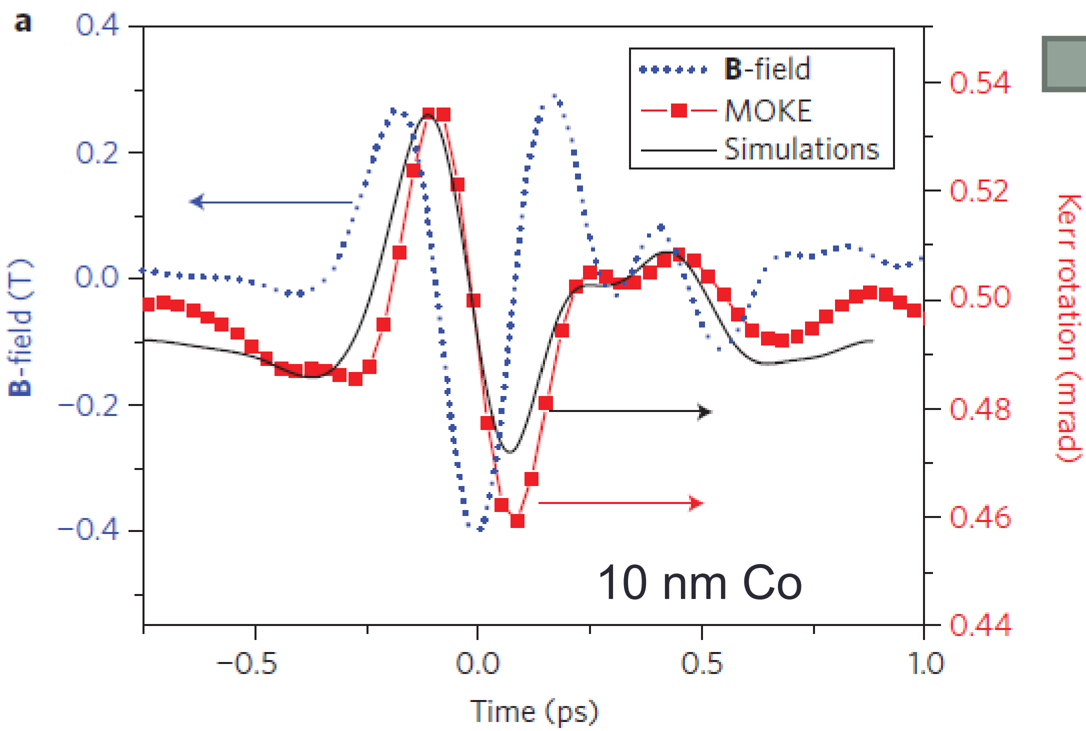
Off-resonant magnetization dynamics phase-locked to an intense phase-stable terahertz transient

C. Vicario[1], C. Ruchert[1], F. Ardana-Lamas[1,2], P. M. Derlet[3], B. Tudu[4], J. Luning[4] and C. P. Hauri[1,2*]

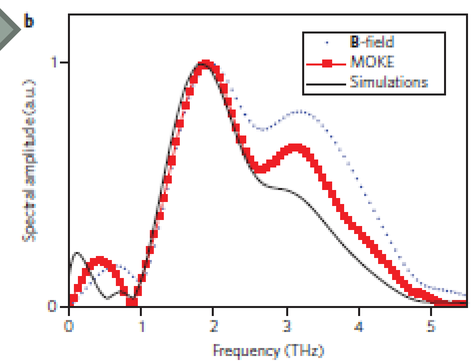
> THz dynamics

Idea and typical parameters

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



FT

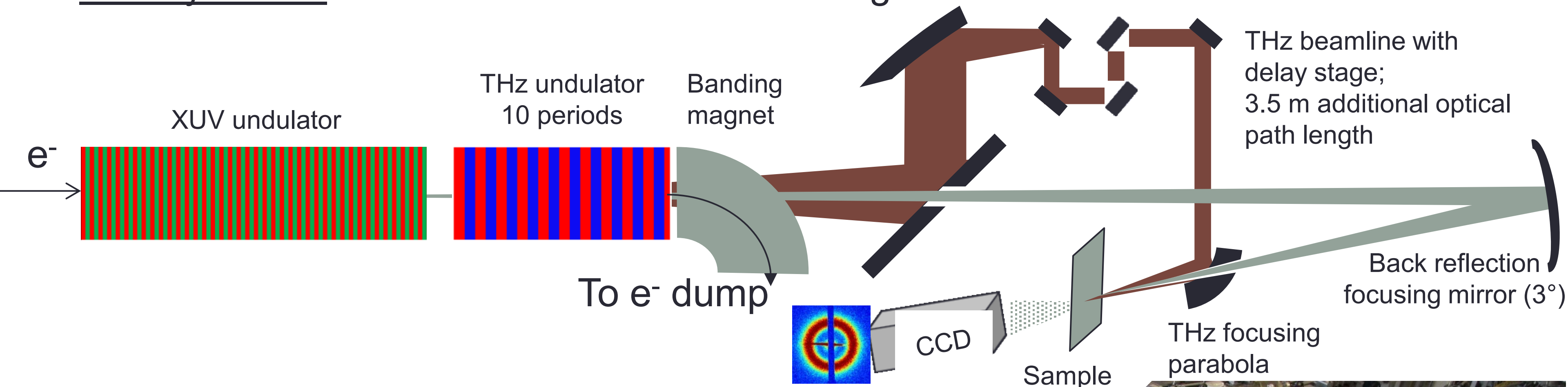


- Fluence of 10 mJ/cm²
➔ |**B**|-field of 200 mT
➔ |**E**|-field of 60 MV/m
➔ role of **E**-field?

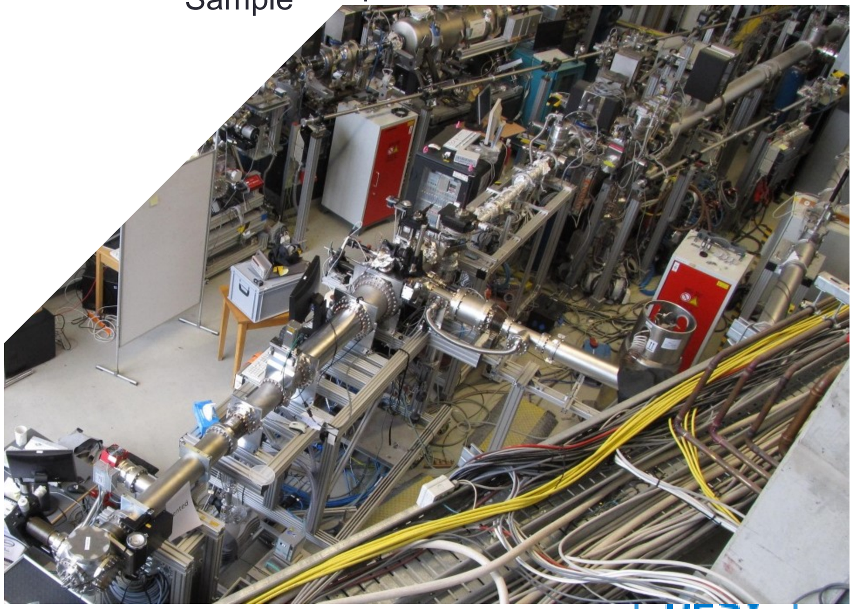
Femtomagnetism

> 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



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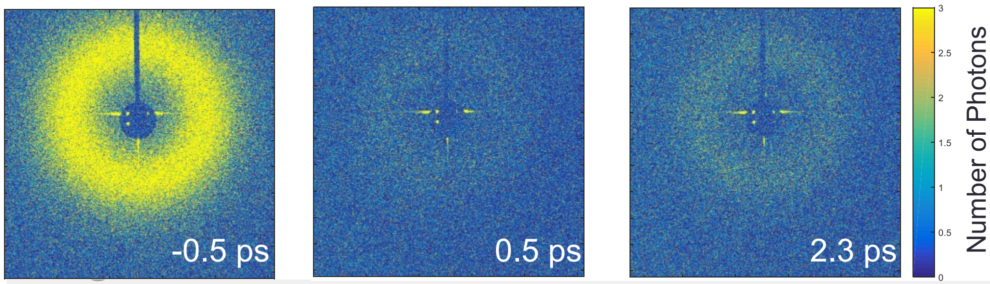
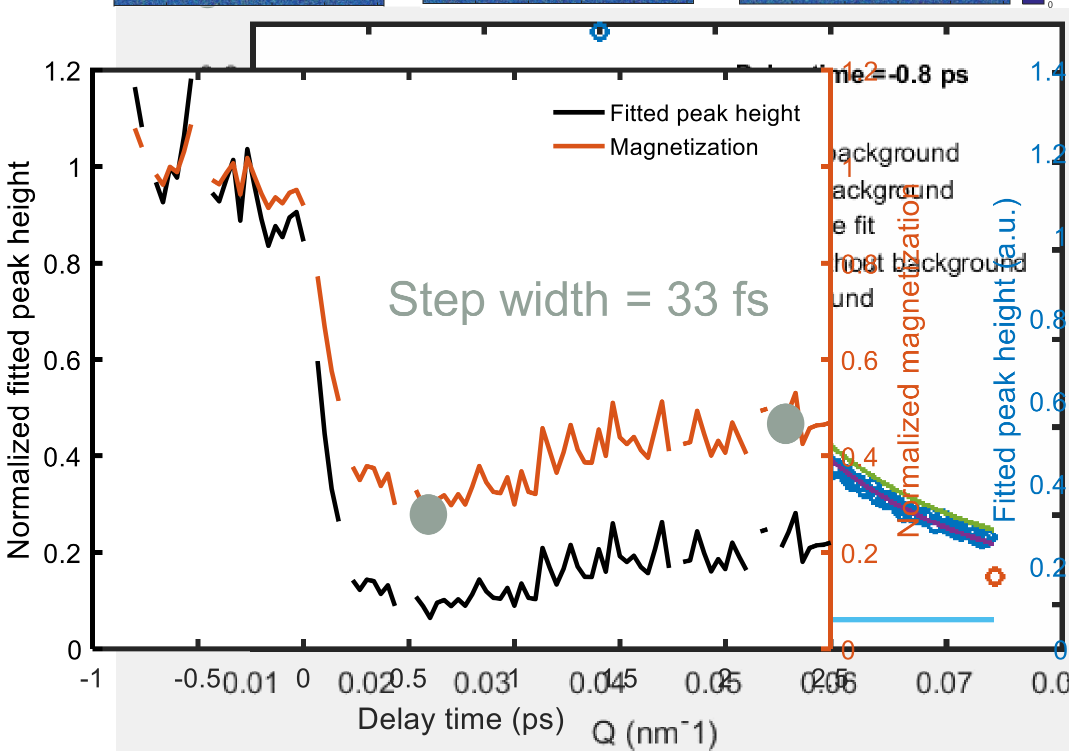
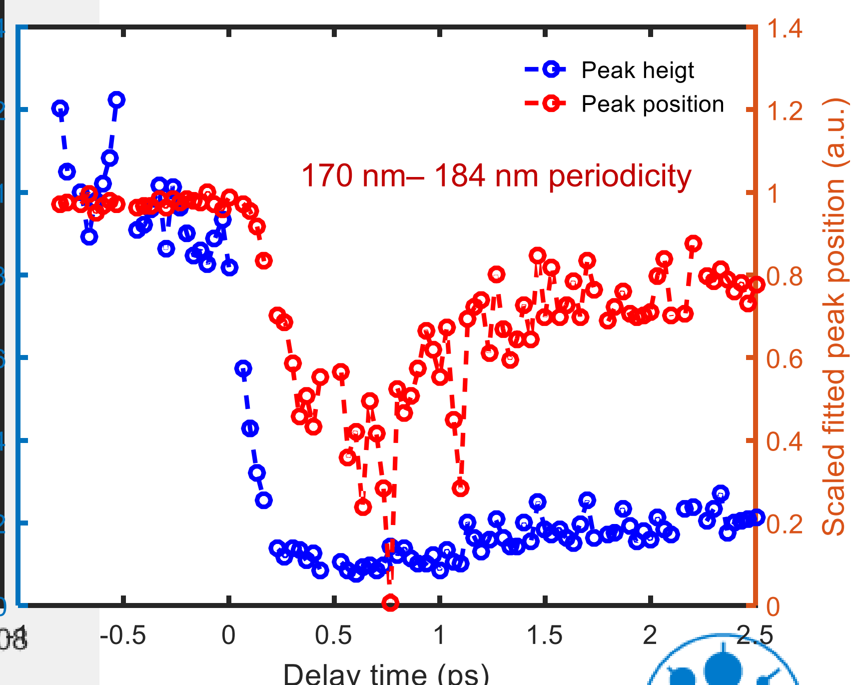
> THz dynamics   What are we doing



- 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 ~ 8%.
- In contrast to IR demagnetization, the dynamics of both seems to be different





Femtomagnetism

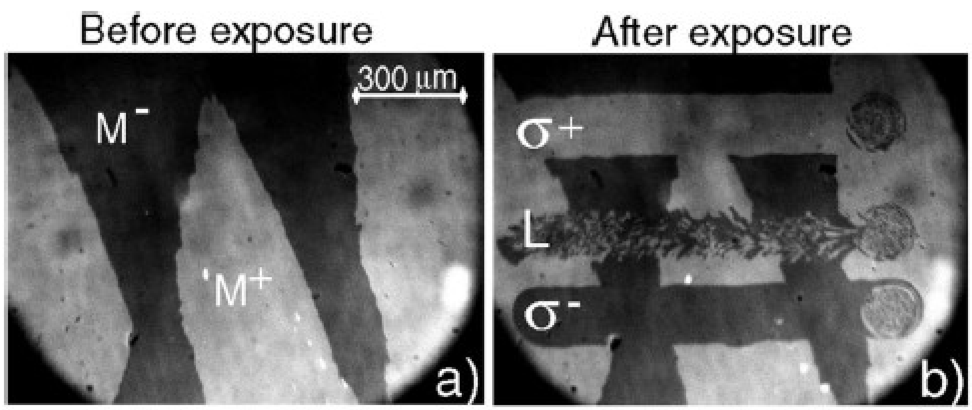
PRL 99, 047601 (2007) PHYSICAL REVIEW LETTERS week ending 27 JULY 2007

All-Optical Magnetic Recording with Circularly Polarized Light

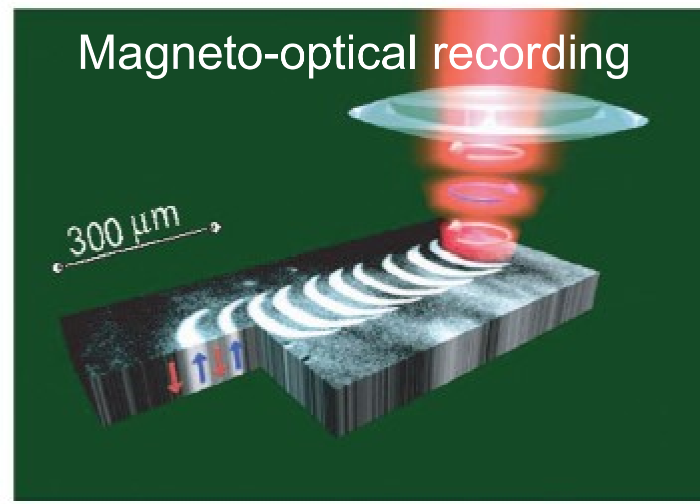
C. D. Stanciu,[1,*] F. Hansteen,[1] A. V. Kimel,[1] A. Kirilyuk,[1] A. Tsukamoto,[2] A. Itoh,[2] and Th. Rasing[1]
[1]*Institute for Molecules and Materials, Radboud University Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands*
[2]*College of Science and Technology, Nihon University, 7-24-1 Funabashi, Chiba, Japan*
(Received 2 March 2007; published 25 July 2007)

> All-optical switching

- Discovery for ferrimagnetic materials



Multi-shot



Single-shot

Femtomagnetism

doi:10.1038/nature09901

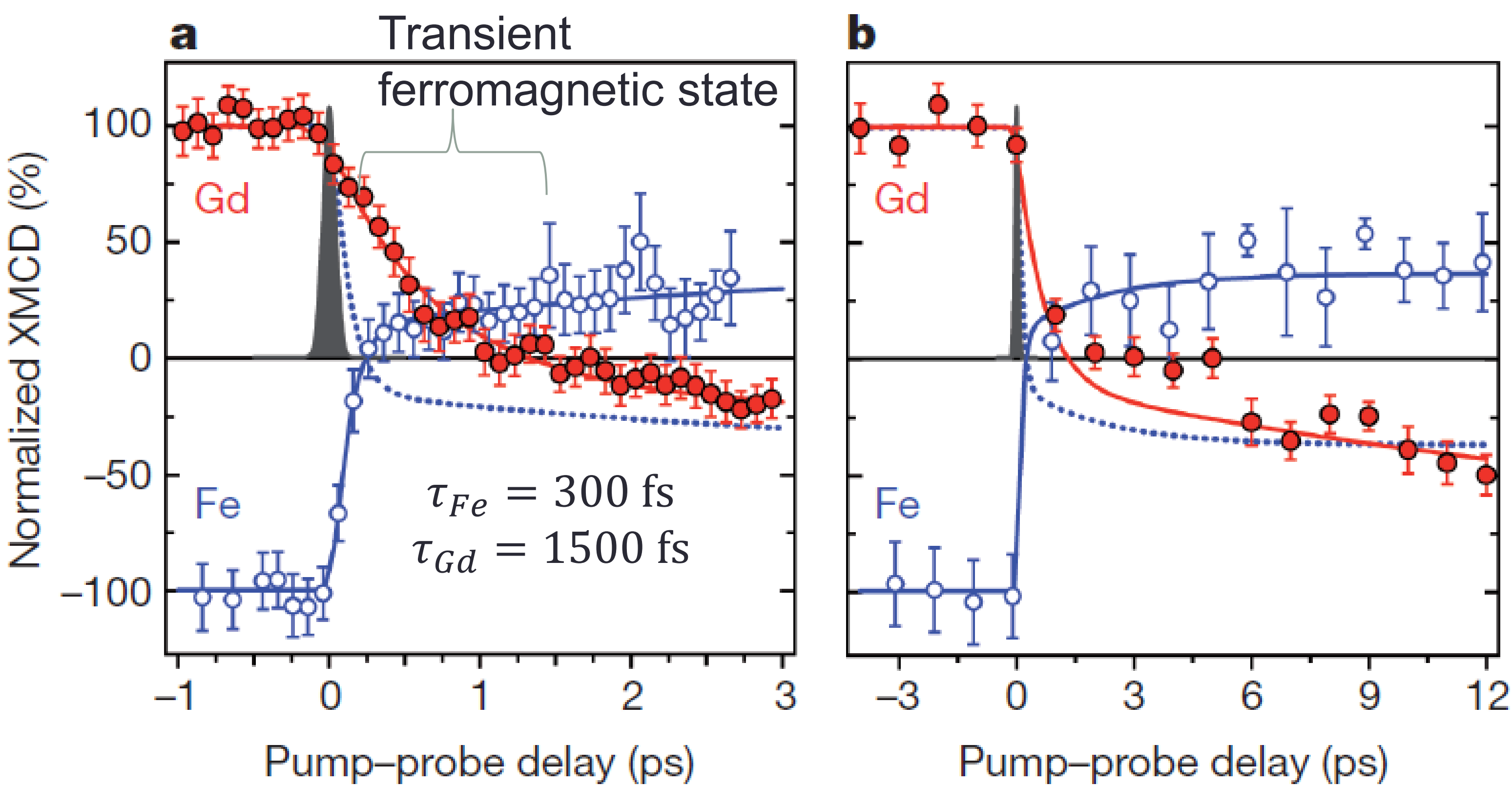
> All-optical switching

- Time-resolved and element-selective studies

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

2011

I. Radu[1,2], K. Vahaplar[1], C. Stamm[2], T. Kachel[2], N. Pontius[2], H. A. Dürr[2,3], T. A. Ostler[4], J. Barker[4], R. F. L. Evans[4], R. W. Chantrell[4], A. Tsukamoto[5,6], A. Itoh[5], A. Kirilyuk[1], Th. Rasing[1] & A. V. Kimel[1]



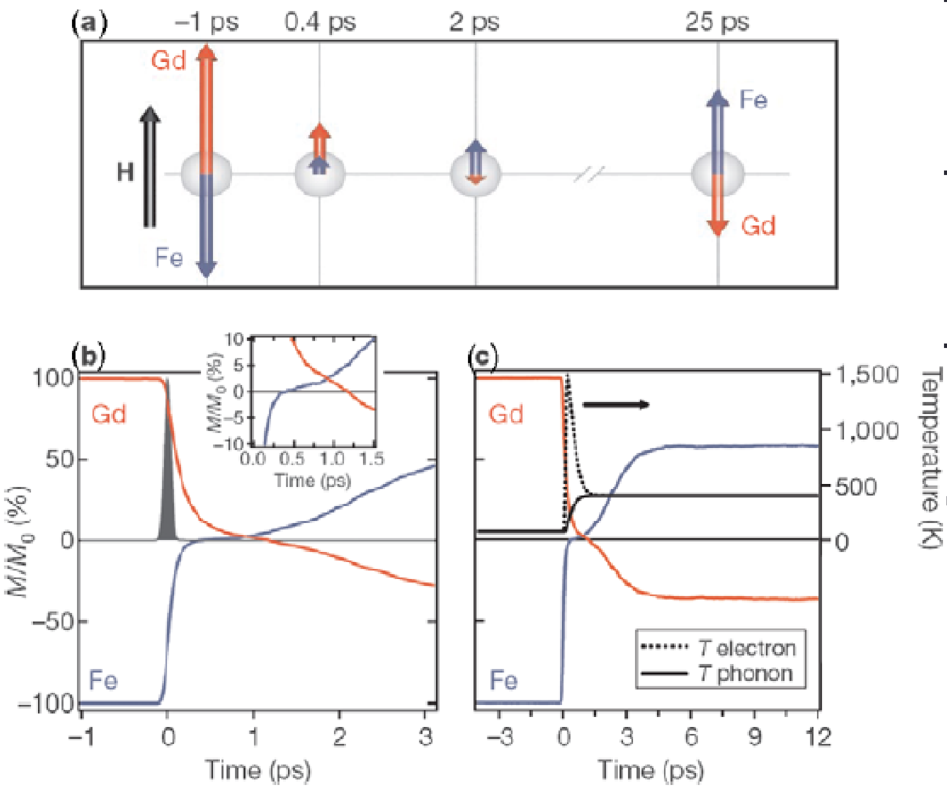
Femtomagnetism

Rep. Prog. Phys. 76 (2013) 026501 (35pp) doi:10.1088/0034-4885/76/2/02650

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 leads to switching of magnetization of Fe
- 1.5-2 ps: antiferromagnetic coupling between Gd and Fe leads 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?

Femtomagnetism

SCIENCE sciencemag.org 12 SEPTEMBER 2014 • VOL 345 ISSUE 6202

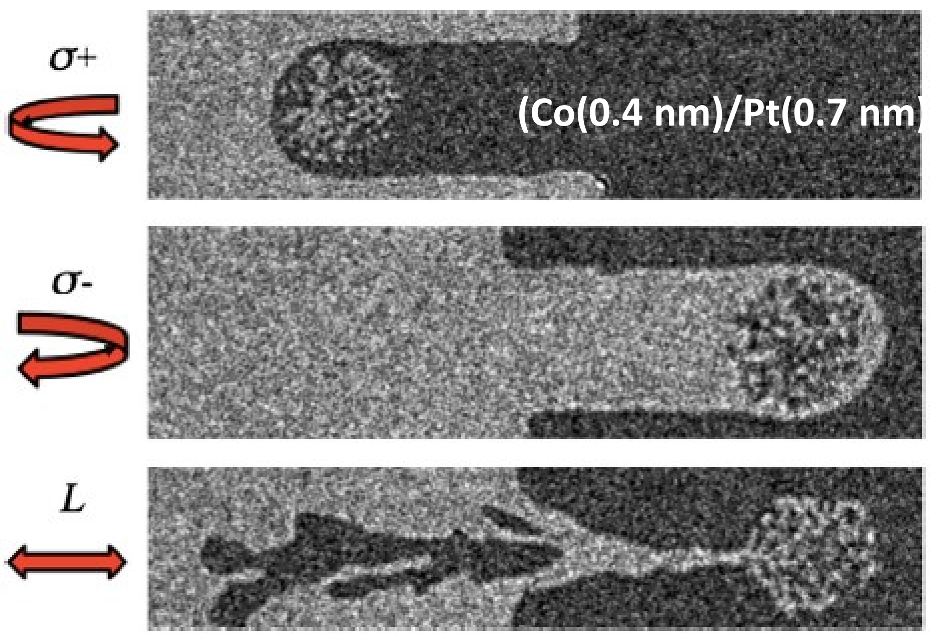
MAGNETISM

All-optical control of ferromagnetic thin films and nanostructures

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

> All-optical switching

- Is there all-optical switching for ferromagnetic materials?



Multi-shot

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

We had a beamtime at FLASH in December 2017 and mid 2018 at FERMI to tackle these questions! Results are difficult to interpret