Methoden moderner Röntgenphysik II: Streuung und Abbildung

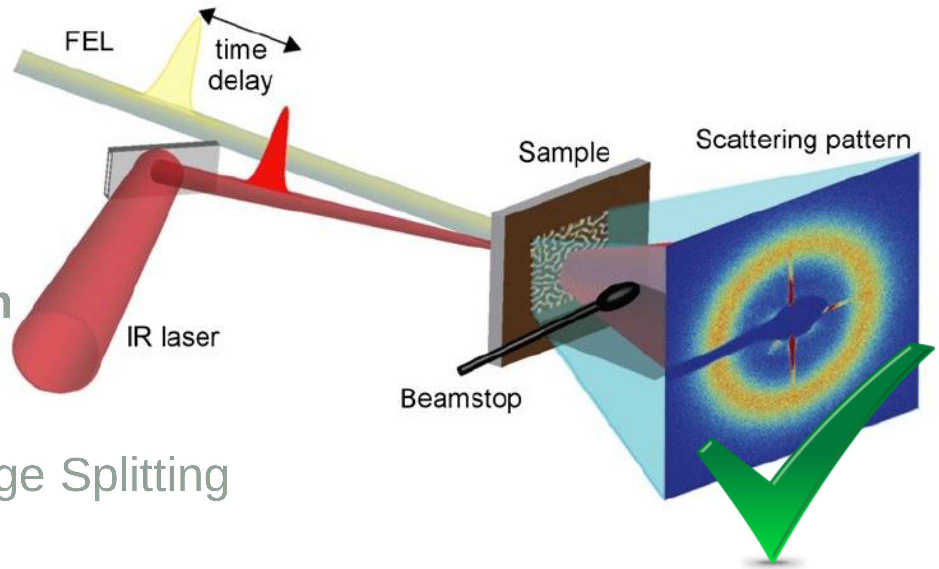
Lecture 25	Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2021		
	G. Grübel, O. Seeck, V. Markmann, F. Lehmkühler, A. Philippi-Kobs, M. Martins		
Location	online		
Date	Tuesdays	12:30 - 14:00	(starting 6.4.)
	Thursdays	8:30 - 10:00	(until 8.7.)

Outline

Part III/2:

Studies on Magnetic Nanostructures

by André Philippi-Kobs (AP)

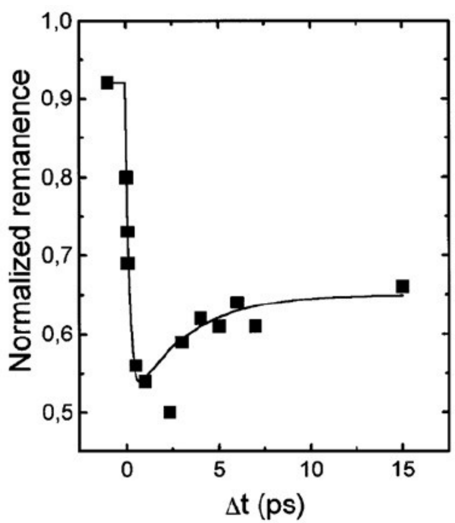


[29.6.] **X-ray Magnetic Circular Dichroism (XMCD) & Resonant Magnetic Small Angle X-ray Scattering (mSAXS)**

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

[1.7.] **Femtomagnetism**

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

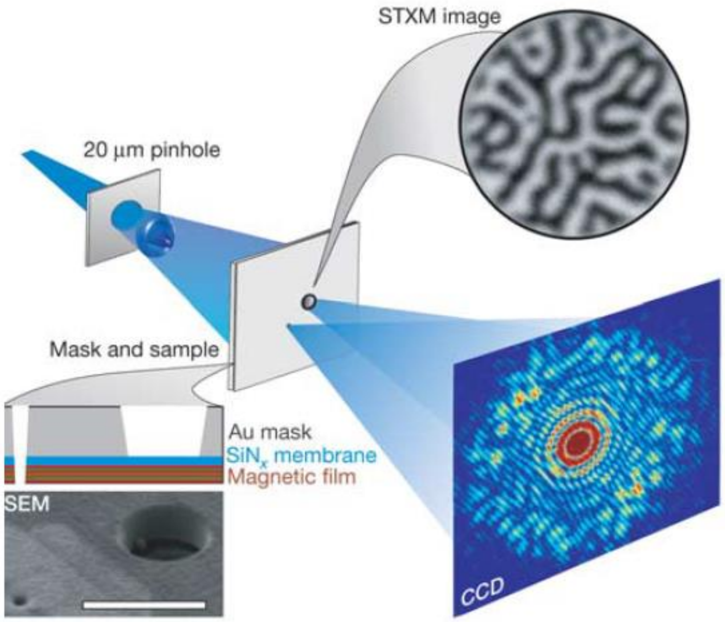


Time-resolved MOKE (Ni 120 fs)

Part III/3:

Studies on Magnetic Nanostructures

by André Philippi-Kobs



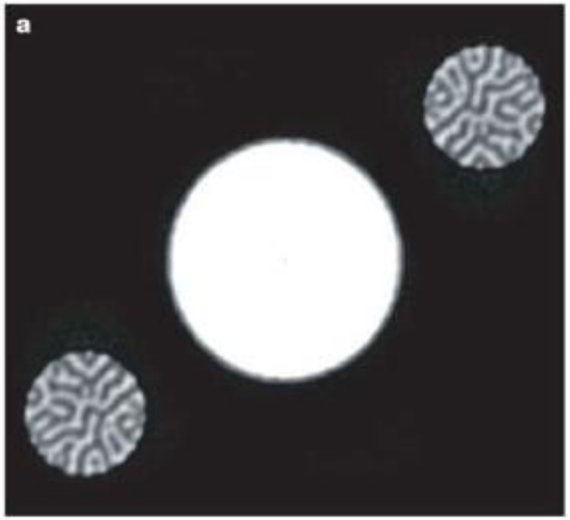
[6.7.] Imaging of Magnetic Domains (briefly)

- **Fourier Transform Holography (FTH)**
- Scanning Transmission X-ray Microscopy (STXM)
- Coherent Diffraction Imaging (CDI)

Lensless imaging of magnetic nanostructures by X-ray spectro-holography

S. Eisebitt[1], J. Lüning[2], W. F. Schlotter[2,3], M. Lörgen[1], O. Hellwig[1,4], W. Eberhardt[1] & J. Stöhr[2]

NATURE | VOL 432 | 16 DECEMBER 2004 |



FTH

Femtomagnetism

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

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

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

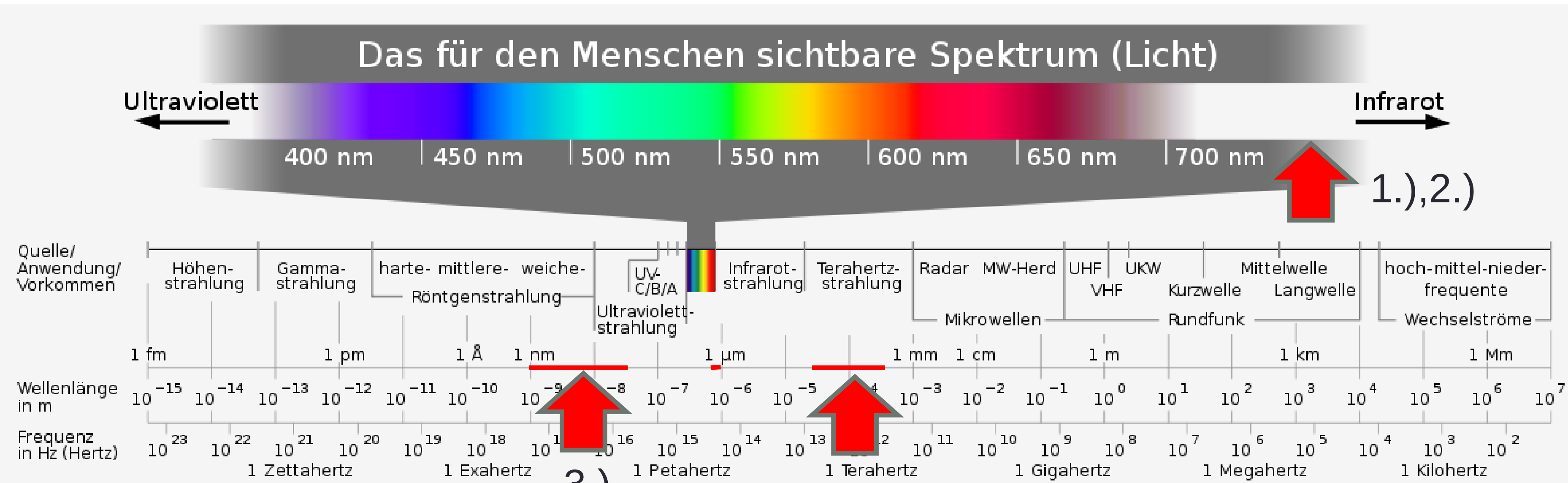
3.) THz dynamics (THz pump, **λ = 100 µm**)

4.) All-optical switching (IR pump, **λ = 800 nm**)

- Ferrimagnetic systems
- Is there AOS in ferromagnetic systems?

Femtomagnetism

> [Electromagnetic spectrum]



3.)

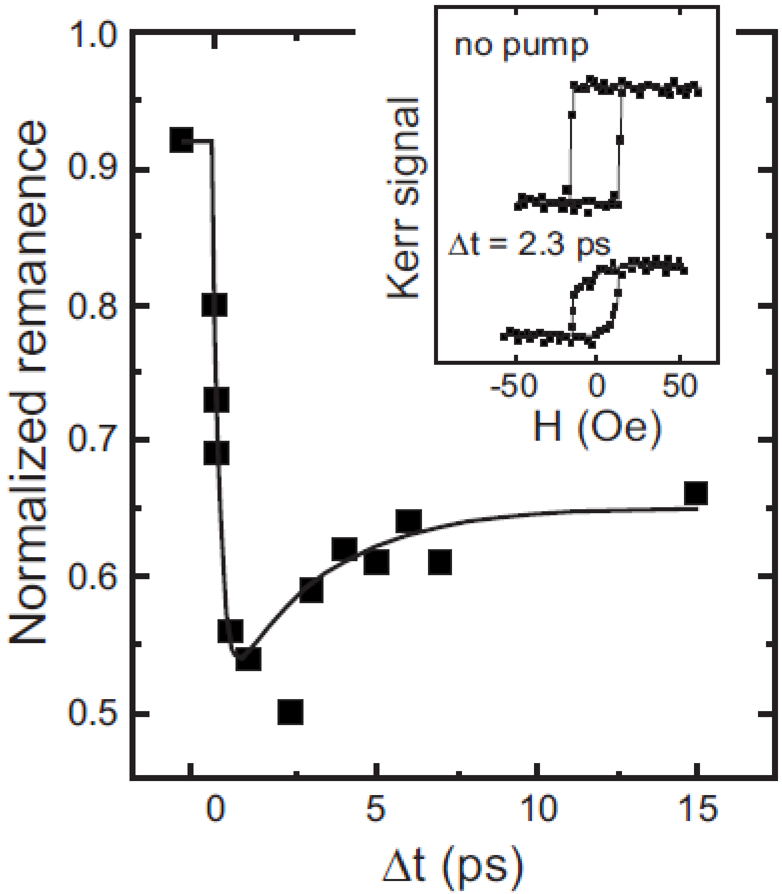
4.) THz gap (no good sources until recently)

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))

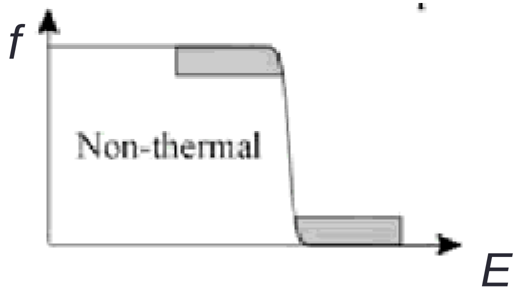
Note:
- non-destructive!
- one can easily reach multi TW
- Largest nuclear power station 4.5 GW (thermal)

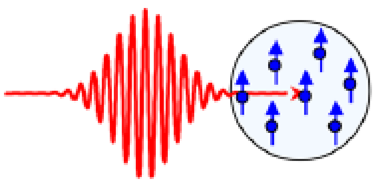
1283 citations (22.06.2020)
~100 in the last 12 months(!)

Femtomagnetism

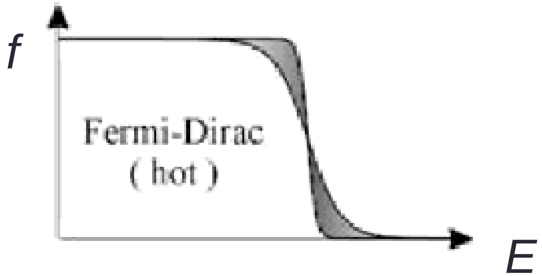
> Ultrafast demagnetization

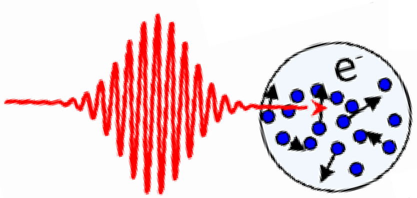
1.) Creation of highly excited electrons during the pulse duration (‘instantaneous‘)





2.) Thermalization of electron system (<100 fs) due to electron-electron scattering

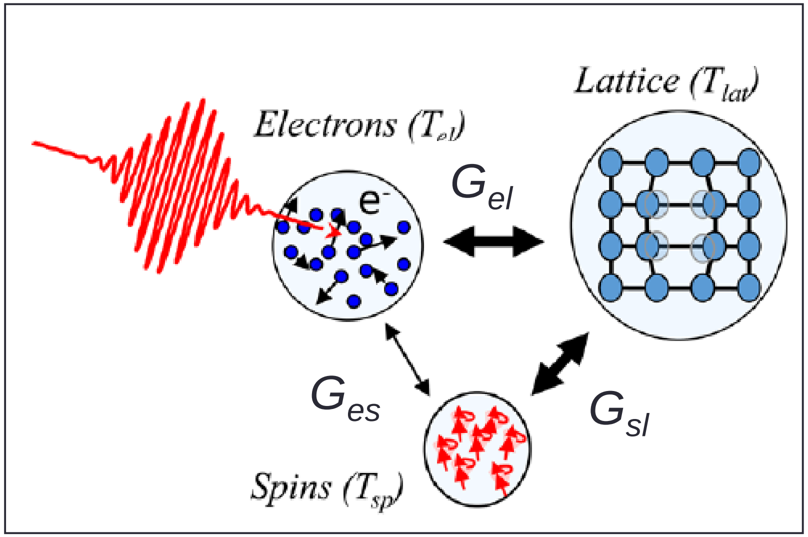


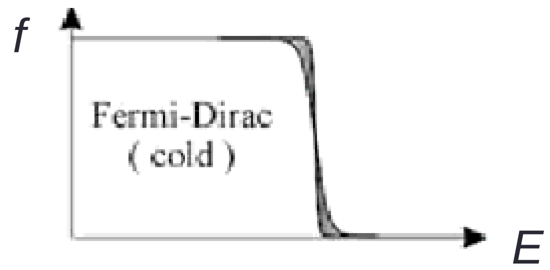


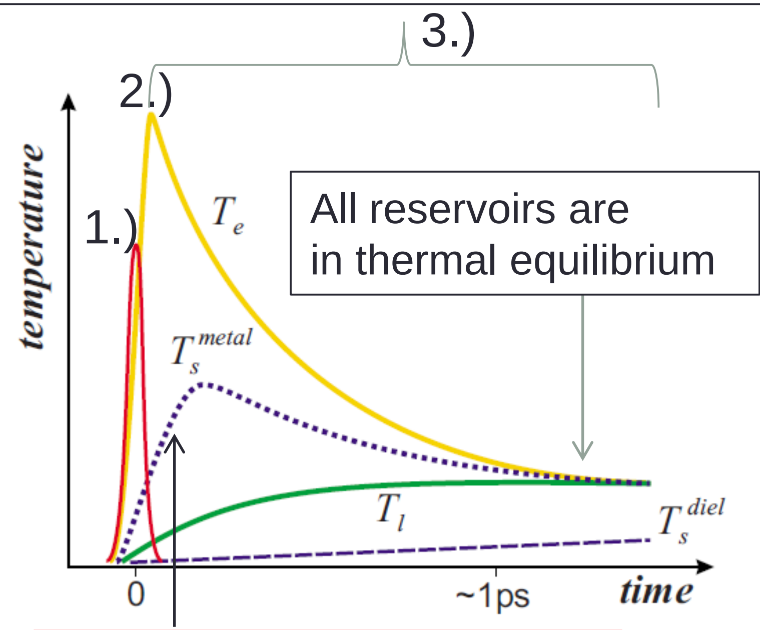
Femtomagnetism

> 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)

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

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

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

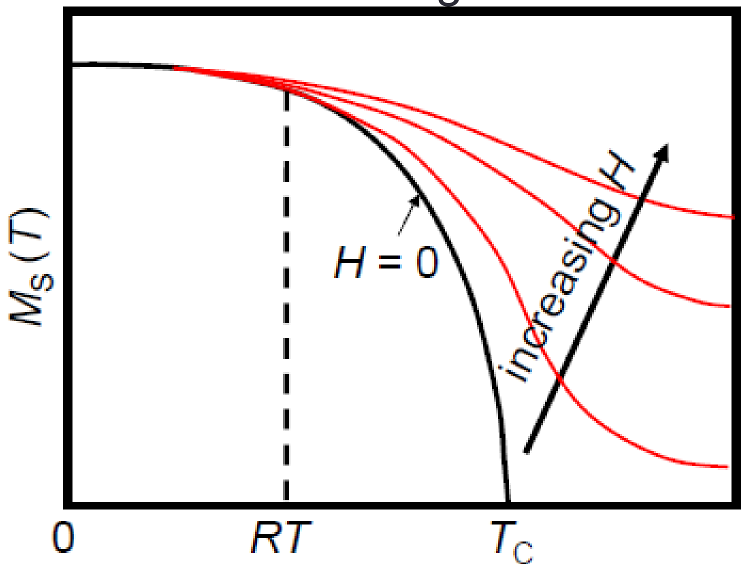
Energy density of pulse ($P(t)$)

Ultrafast demagnetization

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

Femtomagnetism

"thermal" demagnetization



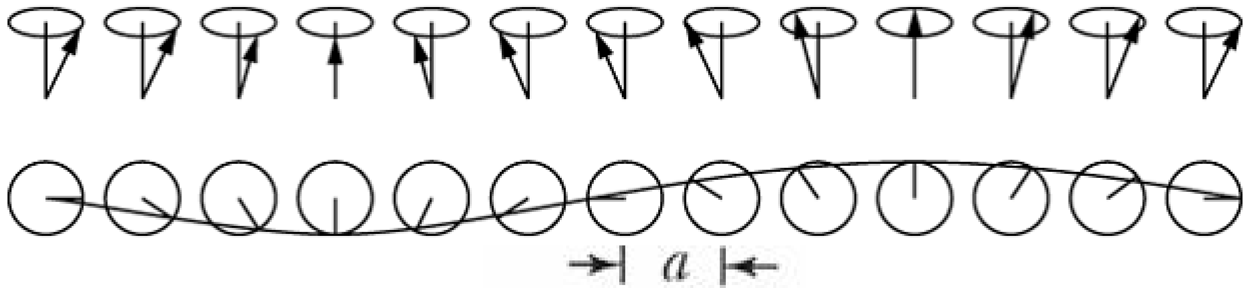
> Ultrafast demagnetization

- T-dependence of saturation magnetization (low T)

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

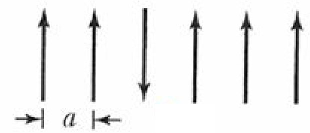
$B = 3.3 \cdot 10^{-6}\ \mathrm{K}^{-3/2}$ for bulk Co

- Collective spin excitations = "magnons" or spin wave (low T, $T \ll T_C$)



Magnon reduces saturation magnetization by $\hbar$

- Single spin excitations (high T, near T_C)



Single spin exc. reduces saturation magnetization by $\hbar$

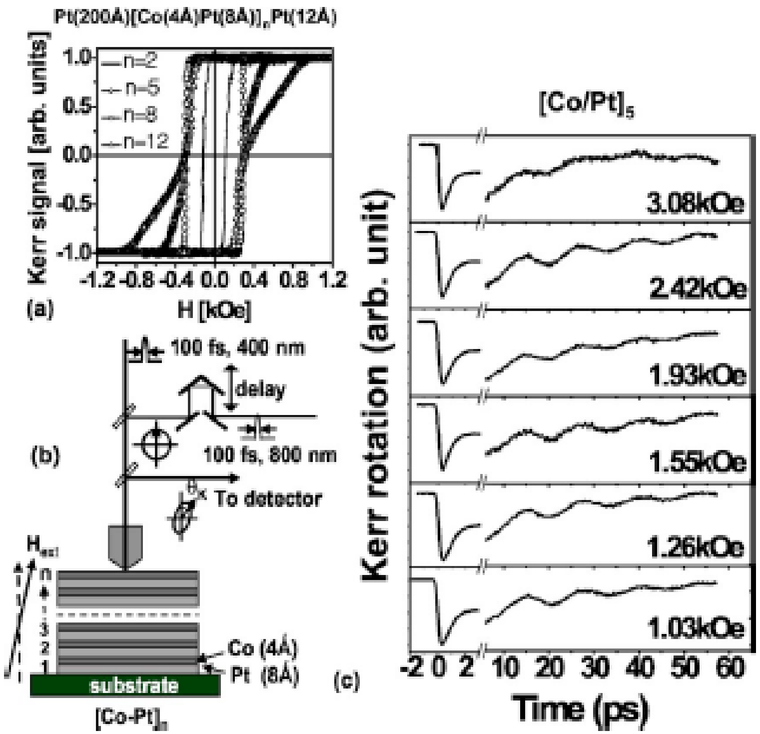
Femtomagnetism

> Ultrafast demagnetization

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

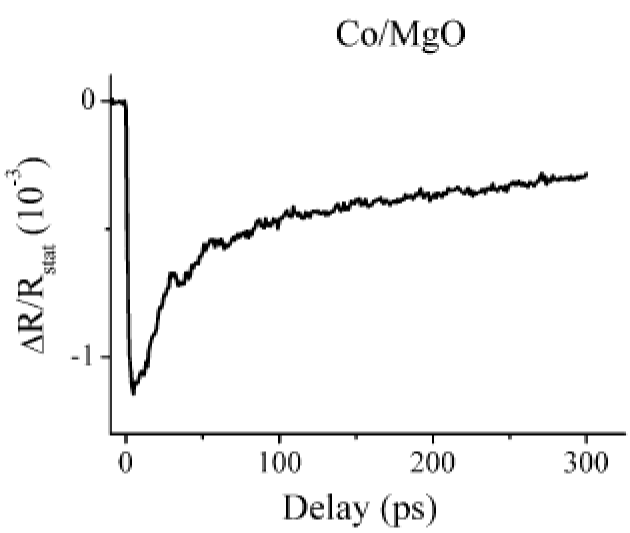
JOURNAL OF APPLIED PHYSICS 101, 09D102 (2007)

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

… of magnetic domain pattern

First pump-probe data (Co M-edge) taken at FLASH

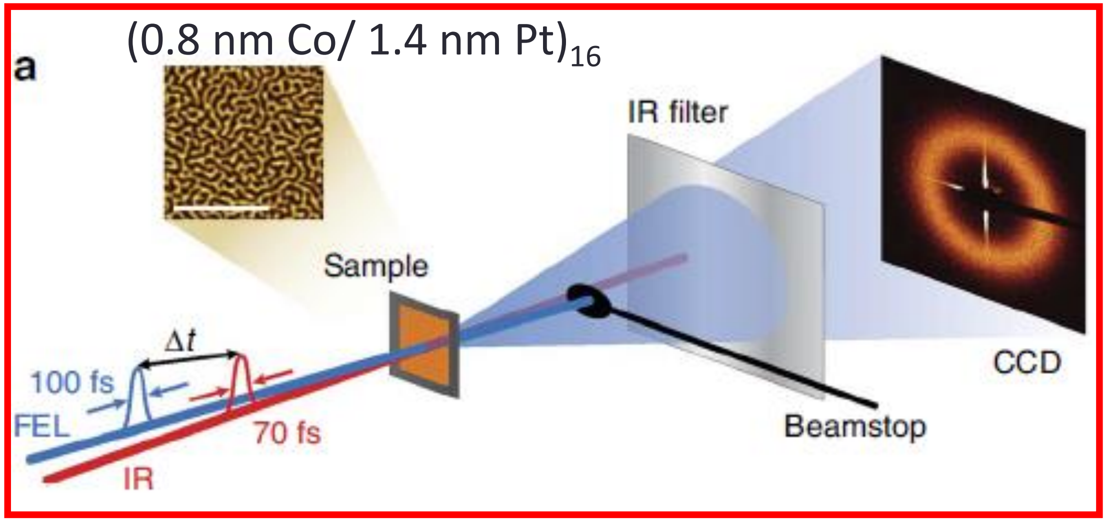
ARTICLE

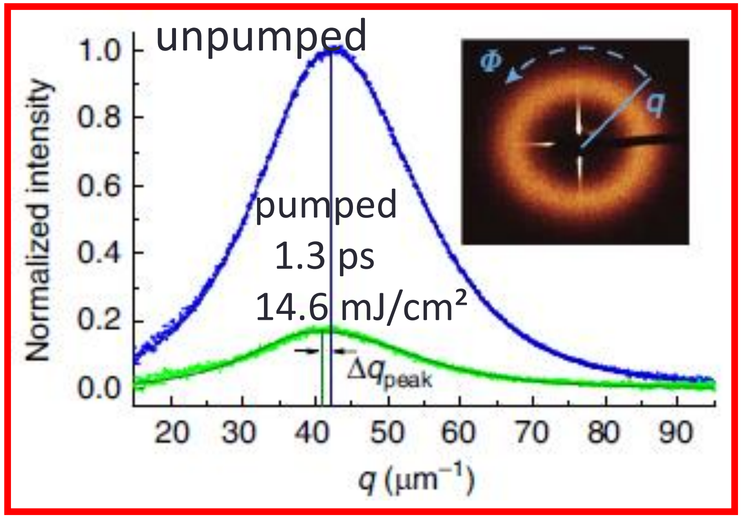
Received 18 Apr 2012 | Accepted 3 Sep 2012 | Published 2 Oct 2012 DOI: 10.1038/ncomms2108

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]

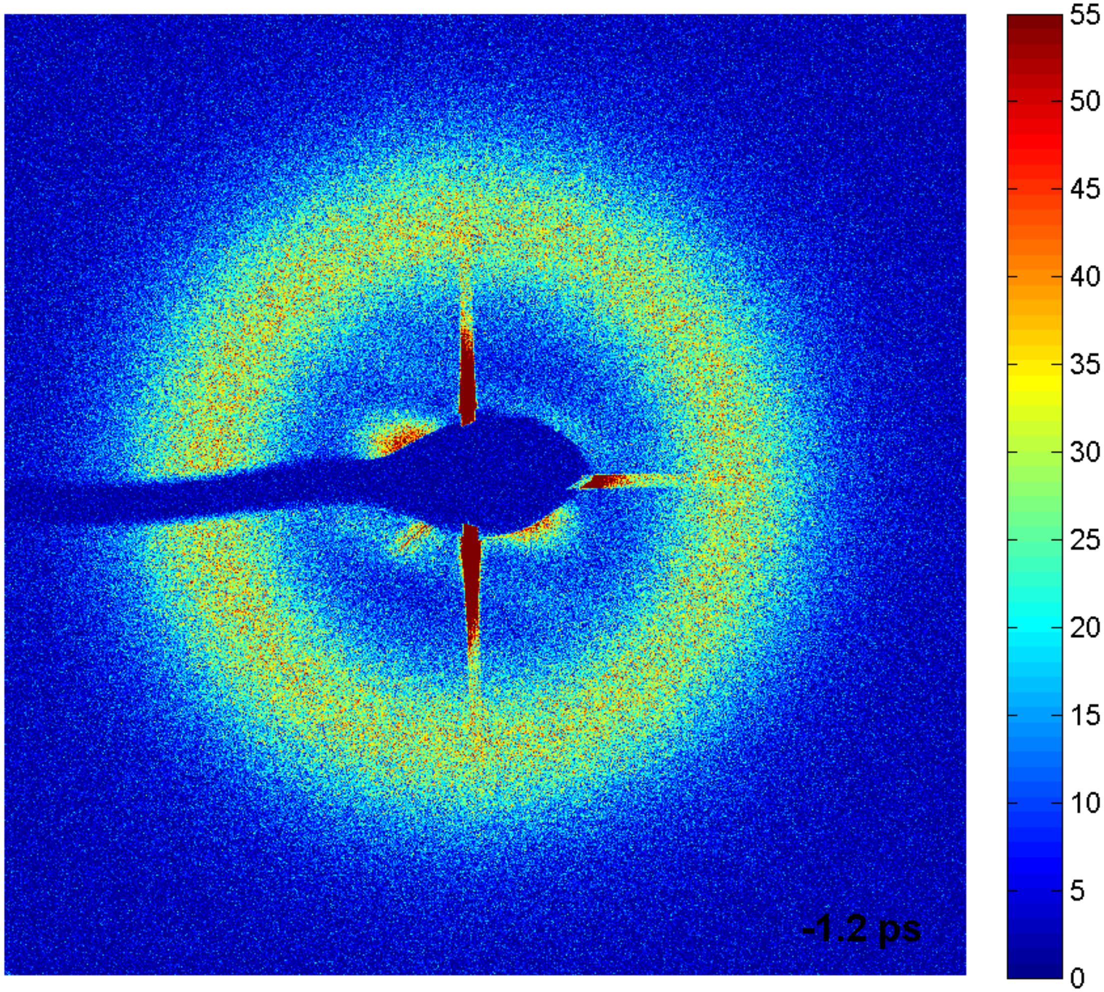
IR-pump−XUV-probe experiment





Femtomagnetism

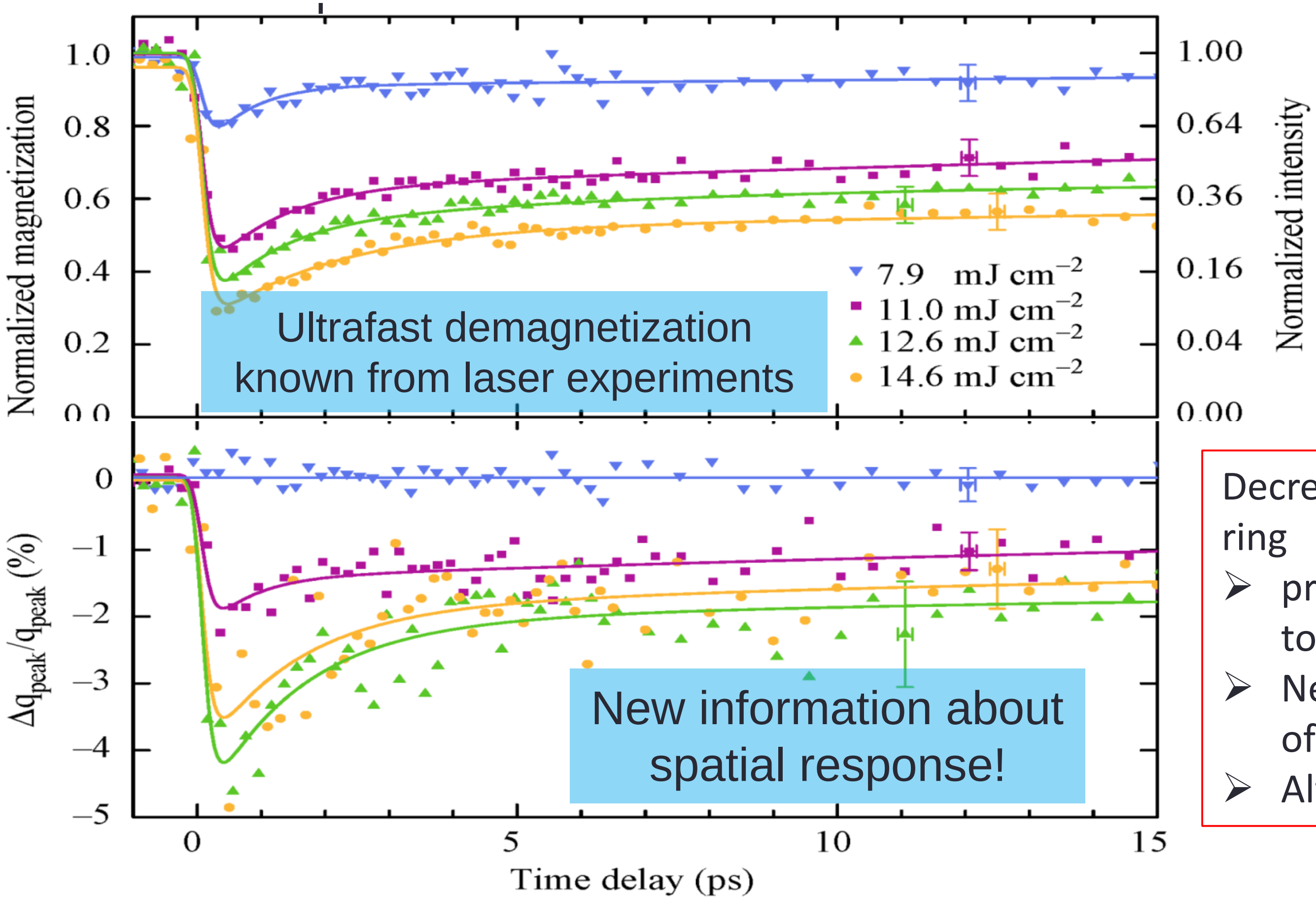
> Ultrafast demagnetization … of magnetic domain pattern



Data used for Nature Commun. **3**, 1100 (2012).

Femtomagnetism

> Ultrafast demagnetization … of magnetic domain pattern



Decrease in radius of scattering ring

- probed eff. 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?

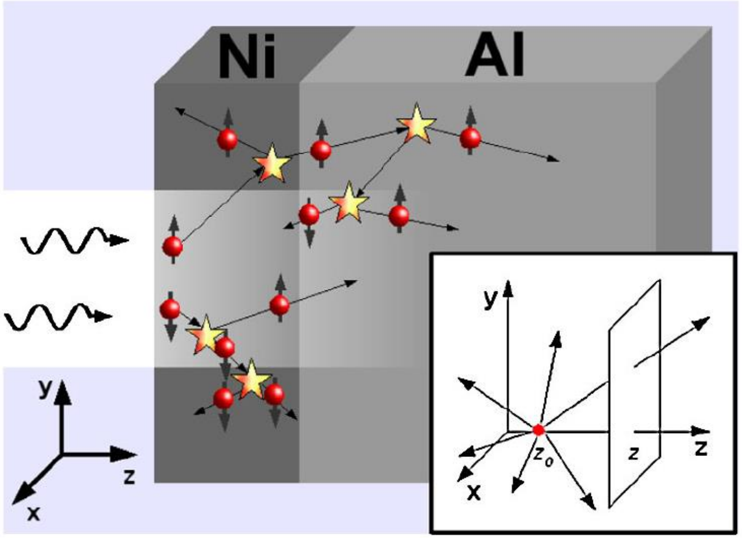
B. Pfau et al., Nat. Commun. **3**, 1100 (2012).

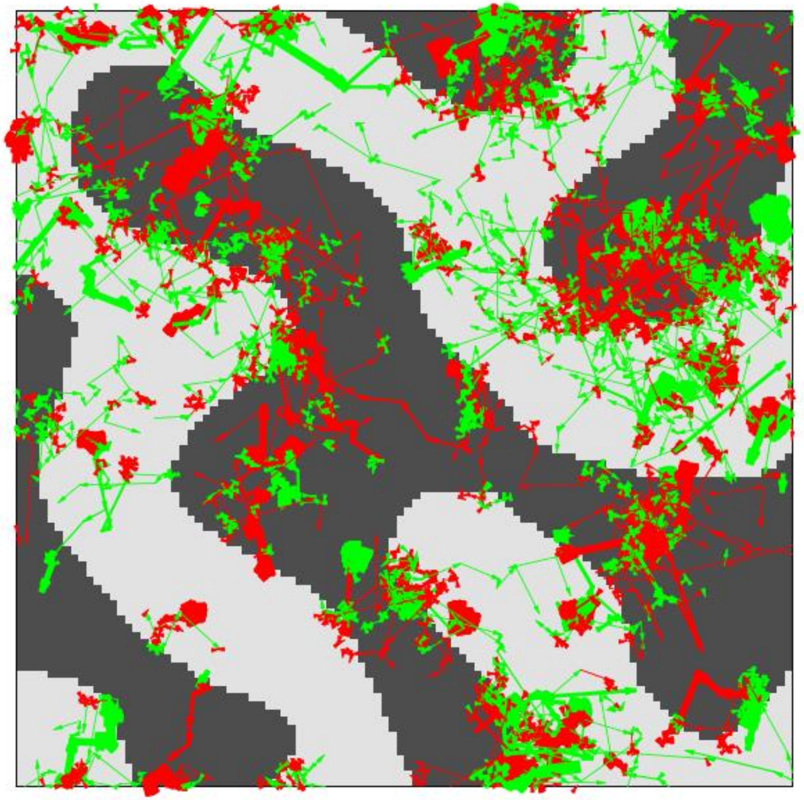
Ultrafast superdiffusive spin transport

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 via superdiffusive currents (spin-dependent electron scattering)

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

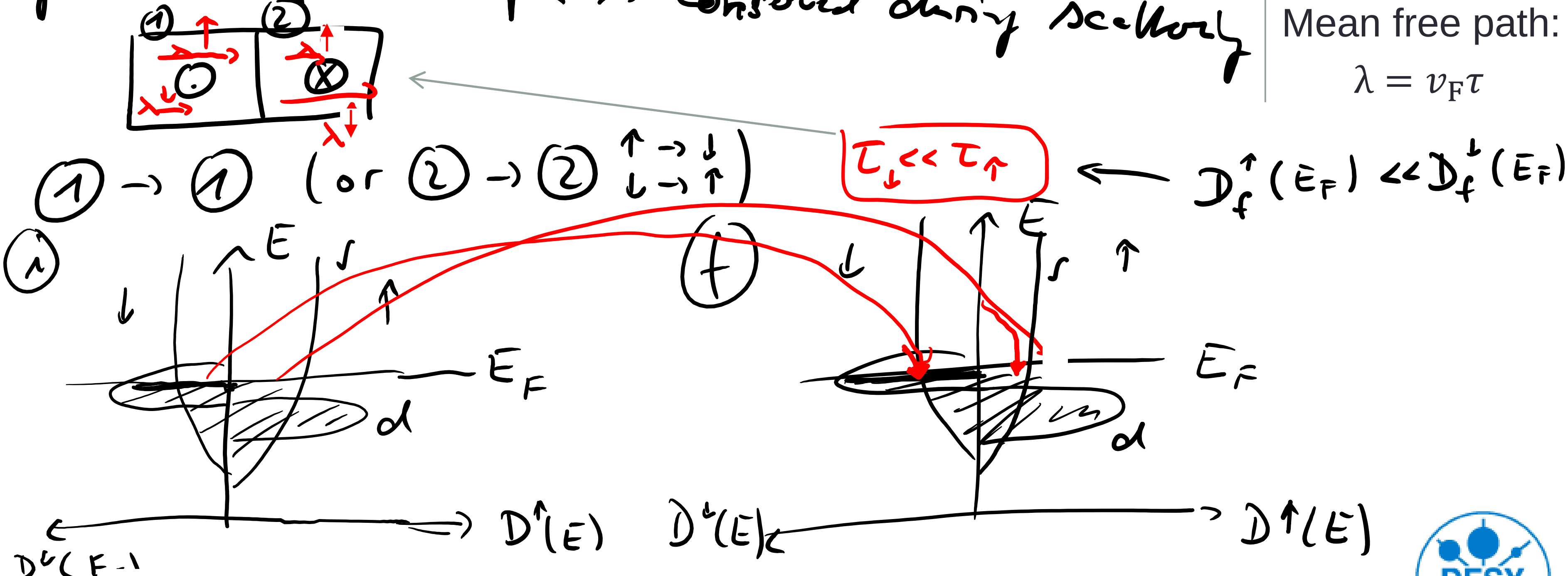
$$\Gamma_{\mathbf{k},\mathbf{k}'} = \frac{2\pi}{\hbar} |< \Phi_f(\mathbf{k}')|V|\Phi_i(\mathbf{k}) >|^2 D_f(E_f')$$

time between 2 scattering events $\tau = \left(\sum_{\vec{k},\vec{k}'} \Gamma_{\vec{k},\vec{k}'}\right)^{-1}$

good assumption: spin is conserved during scattering

Mean free path:

$$\lambda = v_F \tau$$



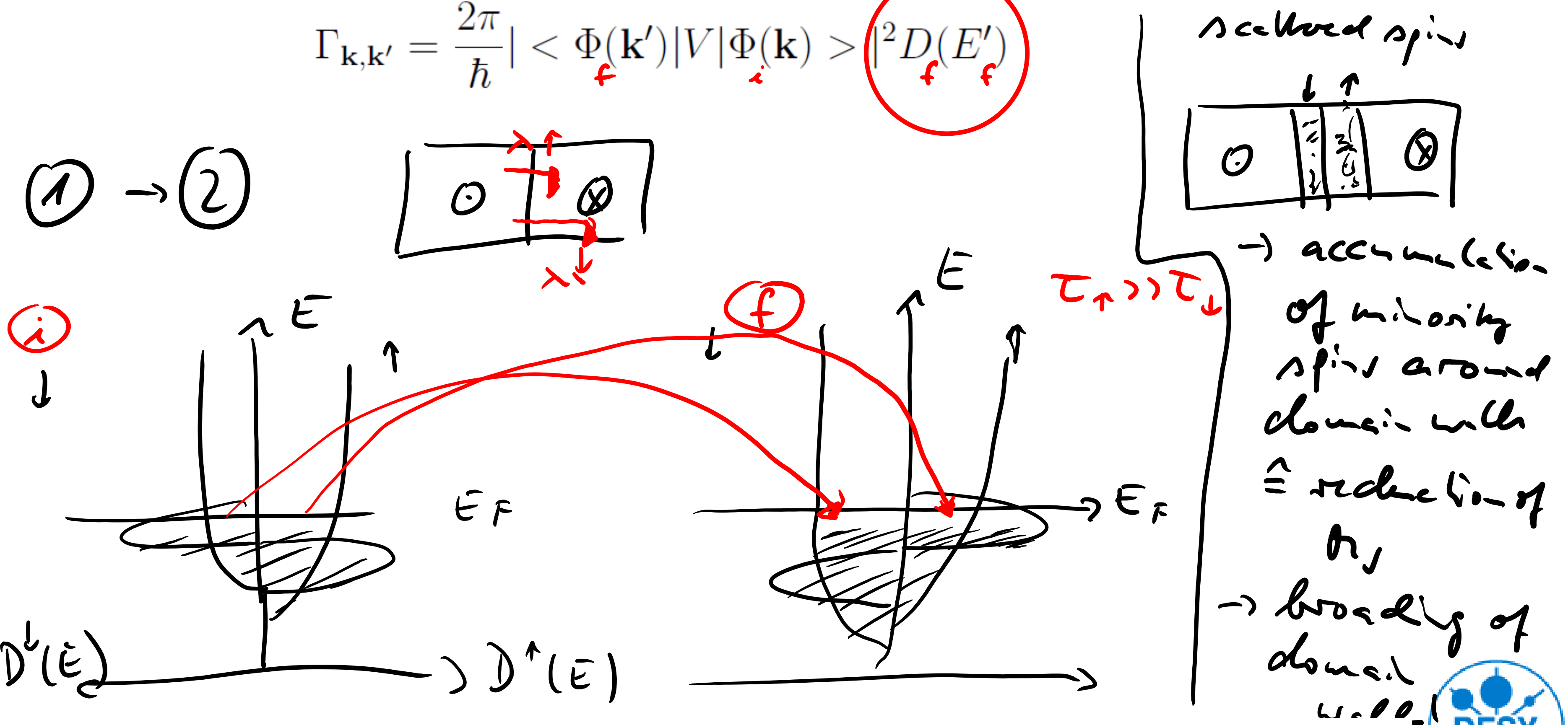
Femtomagnetism

> 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**'

$$\Gamma_{\mathbf{k},\mathbf{k}'} = \frac{2\pi}{\hbar} \left| < \Phi(\mathbf{k}') | V | \Phi(\mathbf{k}) > \right|^2 D(E')$$



mSAXS of magnetic domain patterns

Explanation of transient peak shift via impact of domain wall width on the scattering pattern of a disordered maze domain pattern

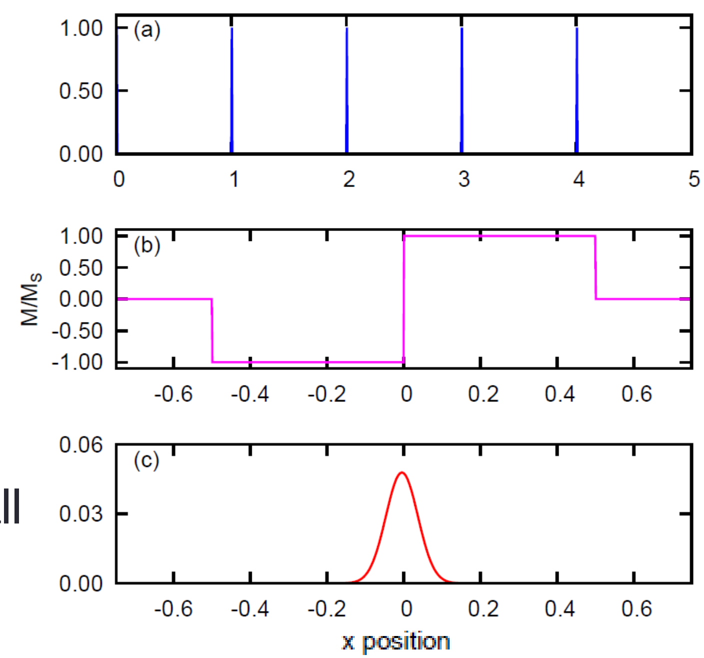
Recap: mSAXS of 1D regular domains with finite domain wall width

Real space

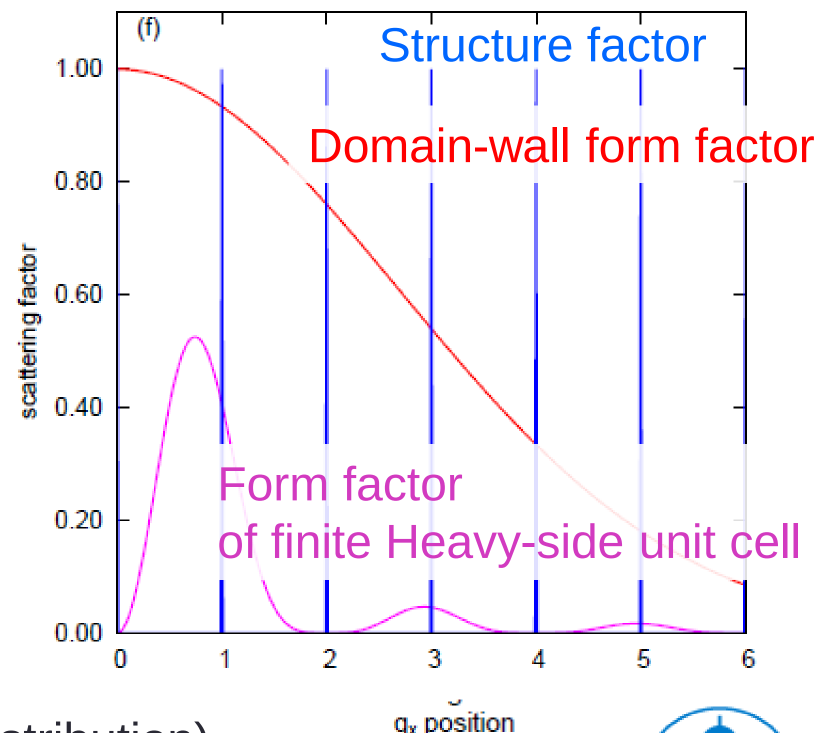
Basic lattic

Unit cell

Domain wall



Q-space

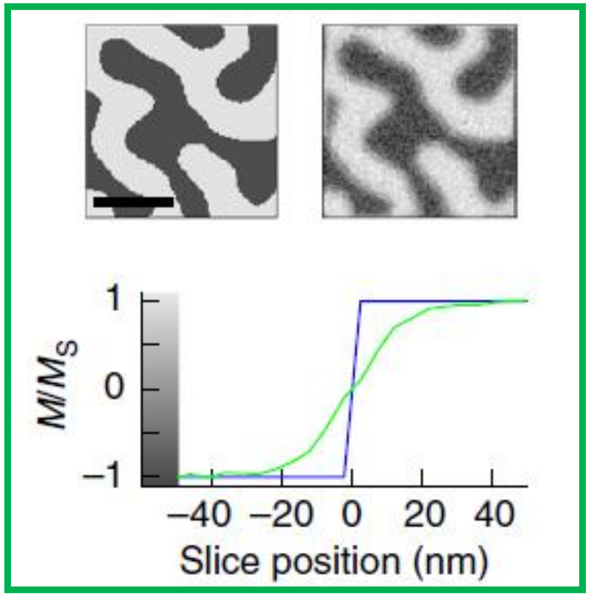


+ consider domain size distribution (Gamma distribution)

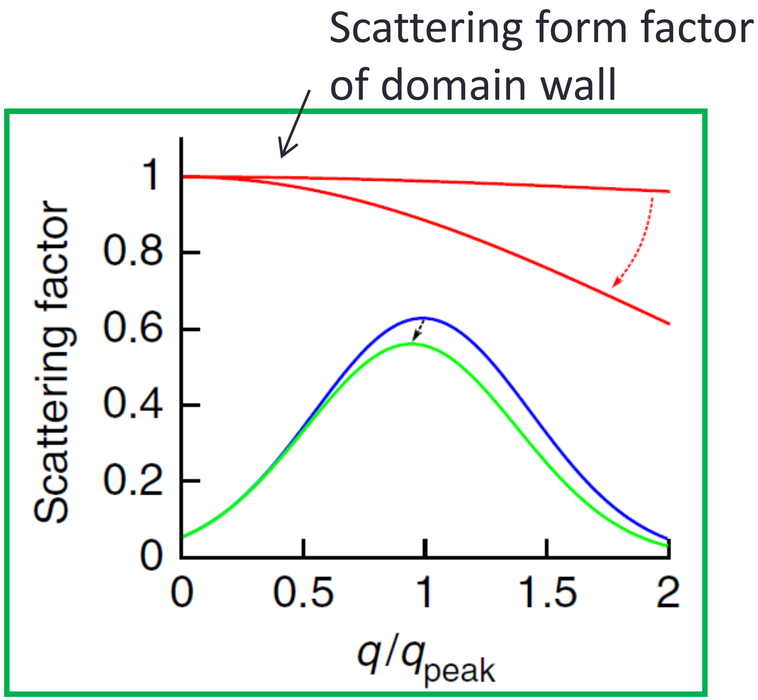
Femtomagnetism

> Ultrafast demagnetization … of magnetic domain pattern

- Explanation of q_{peak} shift



Fourier transformation

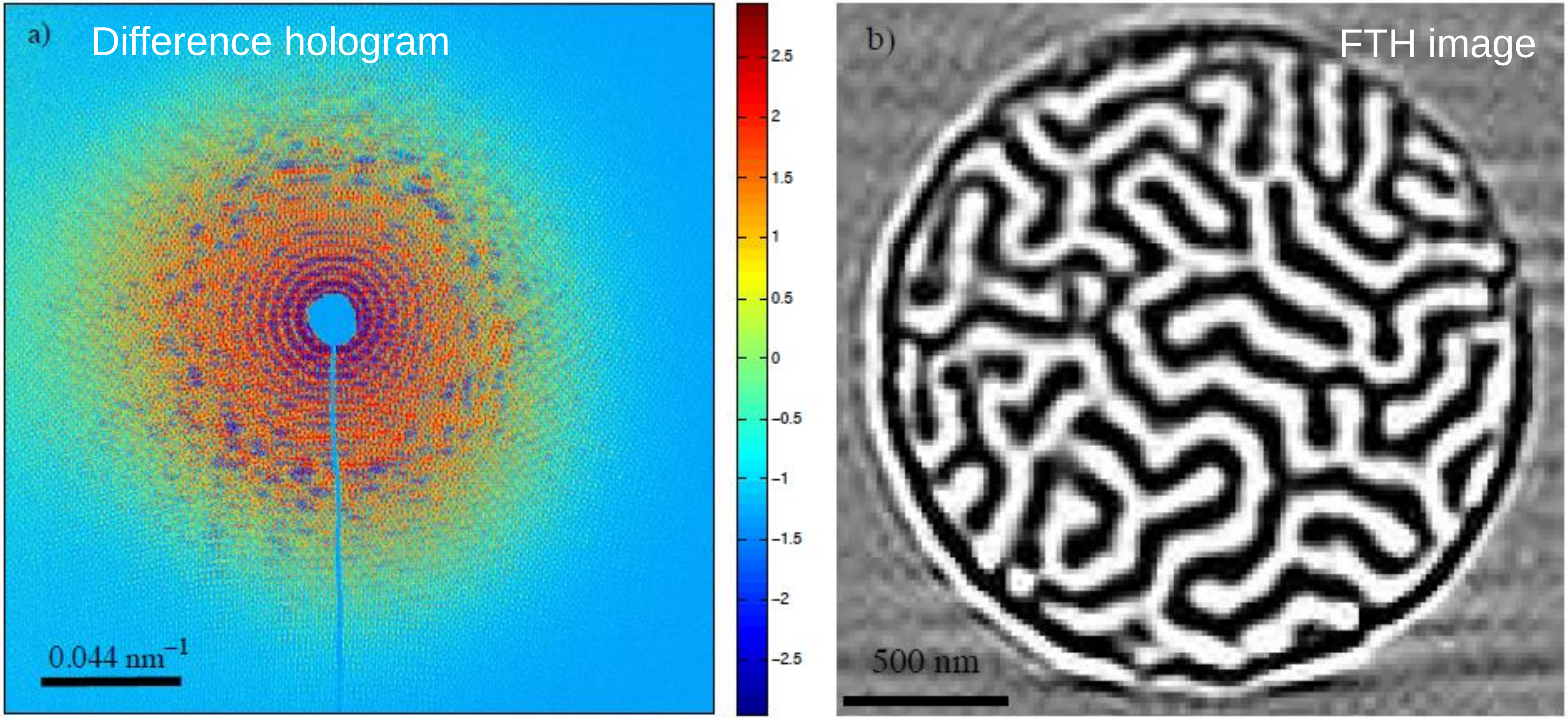


Super-diffusive currents broaden domain walls by 20 nm (FWHM)

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

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

Femtomagnetism



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

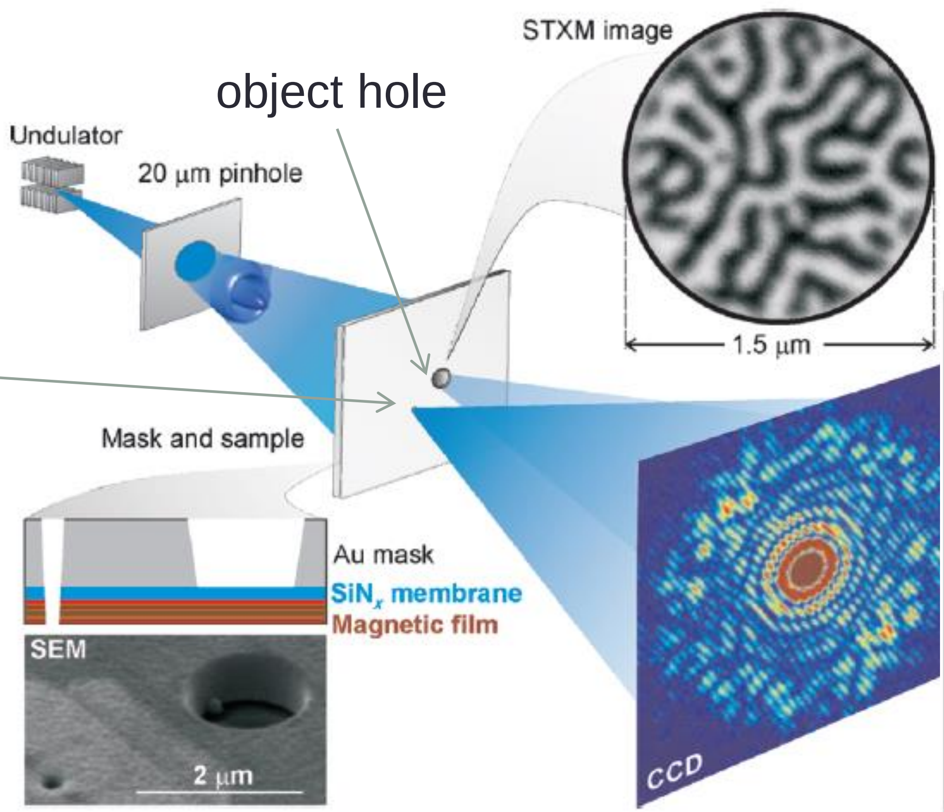
Sync. Rad News **26**, 27 (2013).

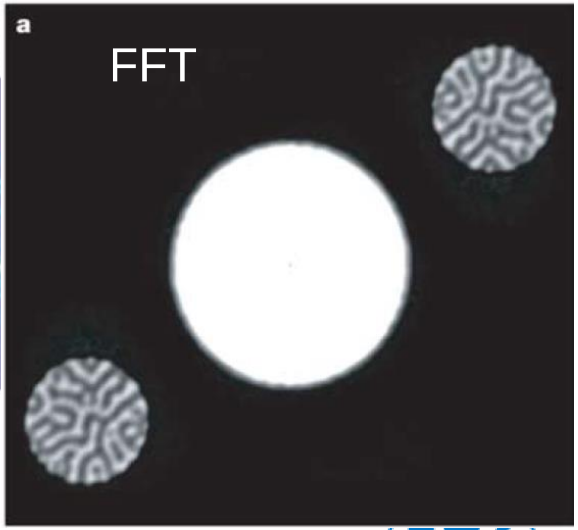
Imaging of magnetic domain patterns with X-rays

> Brief excursion on Lensless Imaging – Fourier transform Holography

	Schlüsselelement-Herstellung		Bild-Rekonstruktion	
TXM	Zonenplatte	XXXXX	-direkt-	X
FTH	Optikmaske	XX	Einfache Fourier-Transformation	XX
CDI	-direkt-	X	Phasen-Rückgewinnung	XXXXX

Requirement: transversal coherence length > largest length scale in sample = mask diameter





Imaging of magnetic domain patterns with X-rays

> Lensless Imaging – Fourier transform Holography (FTH)

Principle:

- Intensity on detector: $$\mathrm{I}(q_x, q_y) = \left| \iint_{-\infty}^{\infty} \Psi(x,y) \exp\left(-i\left(q_x x + q_y y\right)\right)\, \mathrm{d}x\mathrm{d}y \right|^2$$

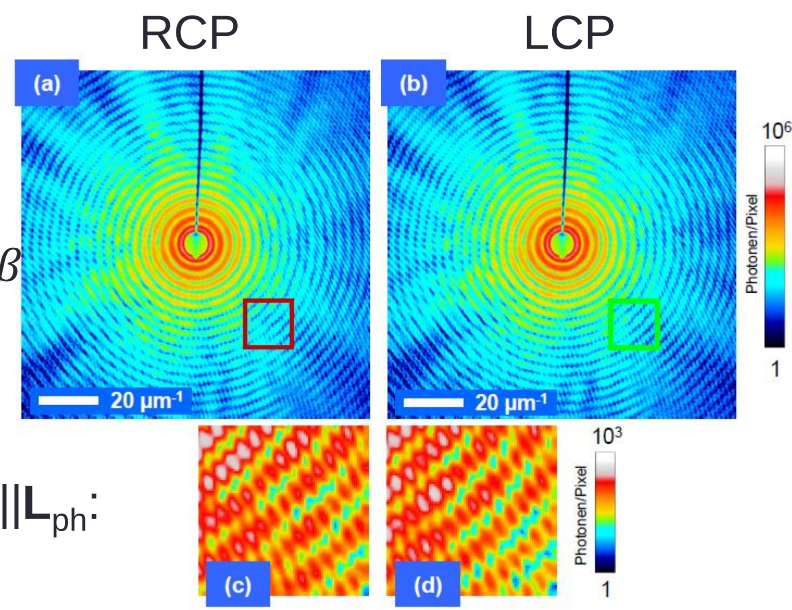
$\Psi(x,y) = t(x,y)\Psi_0, \Psi_0$ =illuminating wave (plane wave)

Transmission function

$$t(x,y) = \exp\left(\frac{2\pi}{\lambda}\int_0^d \left(-i\delta(x,y,z) - \beta(x,y,z)\, \mathrm{d}z\right)\right)$$

With δ and β from $\mathrm{n} = 1 - \delta + \beta$ and for magnetic samples $\delta = \delta_0 \pm (\epsilon_k \cdot \vec{m})\Delta\delta$ and $\beta = \beta_0 \pm (\epsilon_k \cdot \vec{m})\Delta\beta$

Identify t(x,y) as $f = f_0^C + f_r^C + f_m$



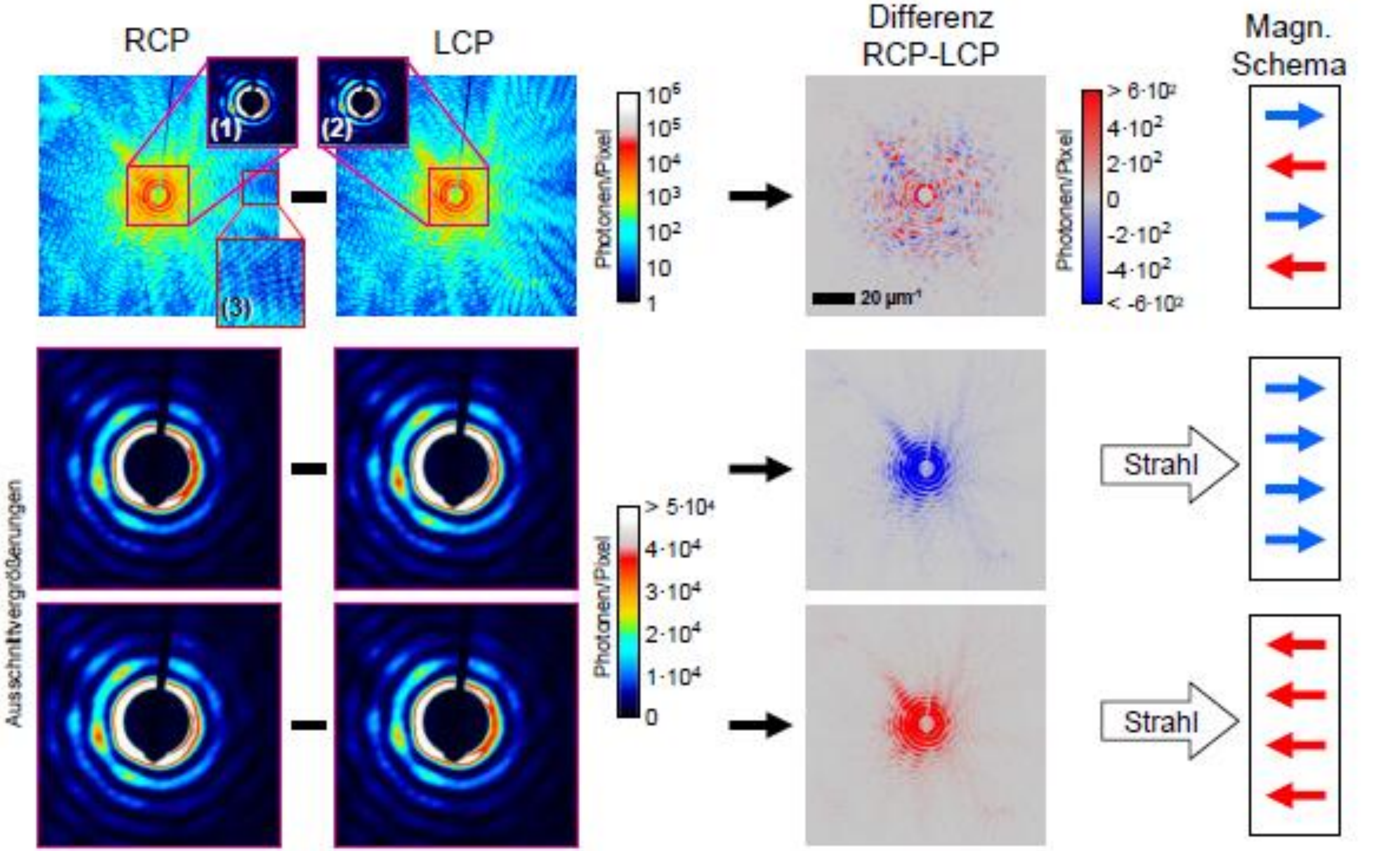
- Scattering factor for circularly polarized light and **M||L**$_{ph}$:

$$I(\vec{Q}) = \left|\tilde{f}_0^C(\vec{Q}) + \tilde{f}_r^C(\vec{Q}) \pm \tilde{f}_m(\vec{Q})\right|^2 = \left|\tilde{f}_C \pm \tilde{f}_m\right|^2$$

$$\Delta I = I(RCP) - I(LCP) = 2 \cdot \left(\tilde{f}_C^* \tilde{f}_m + \tilde{f}_C\ \tilde{f}_m^*\right)$$

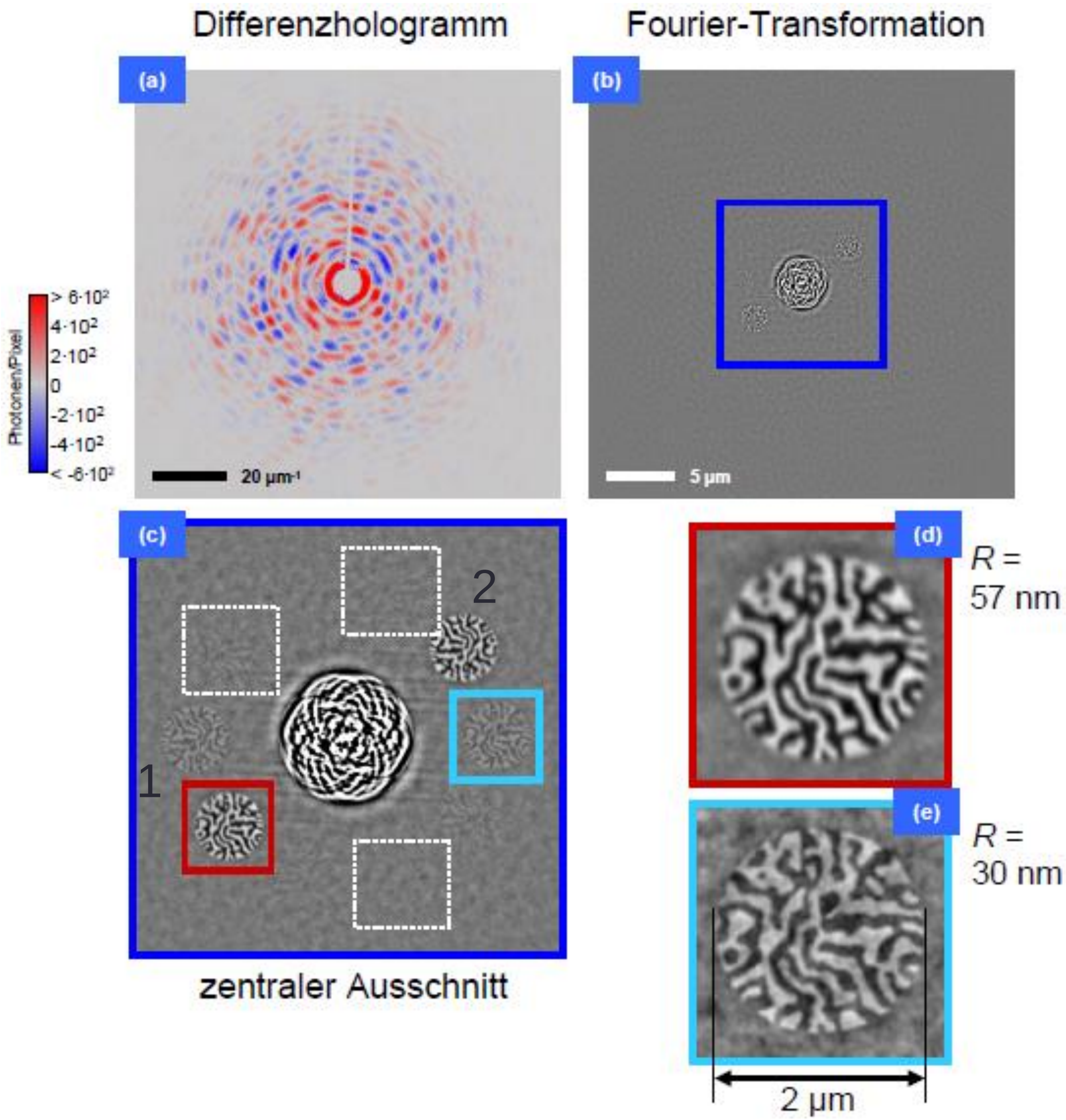
Subtle differences!

Imaging of magnetic domain patterns with X-rays



DESY

Imaging of magnetic domain patterns with X-rays

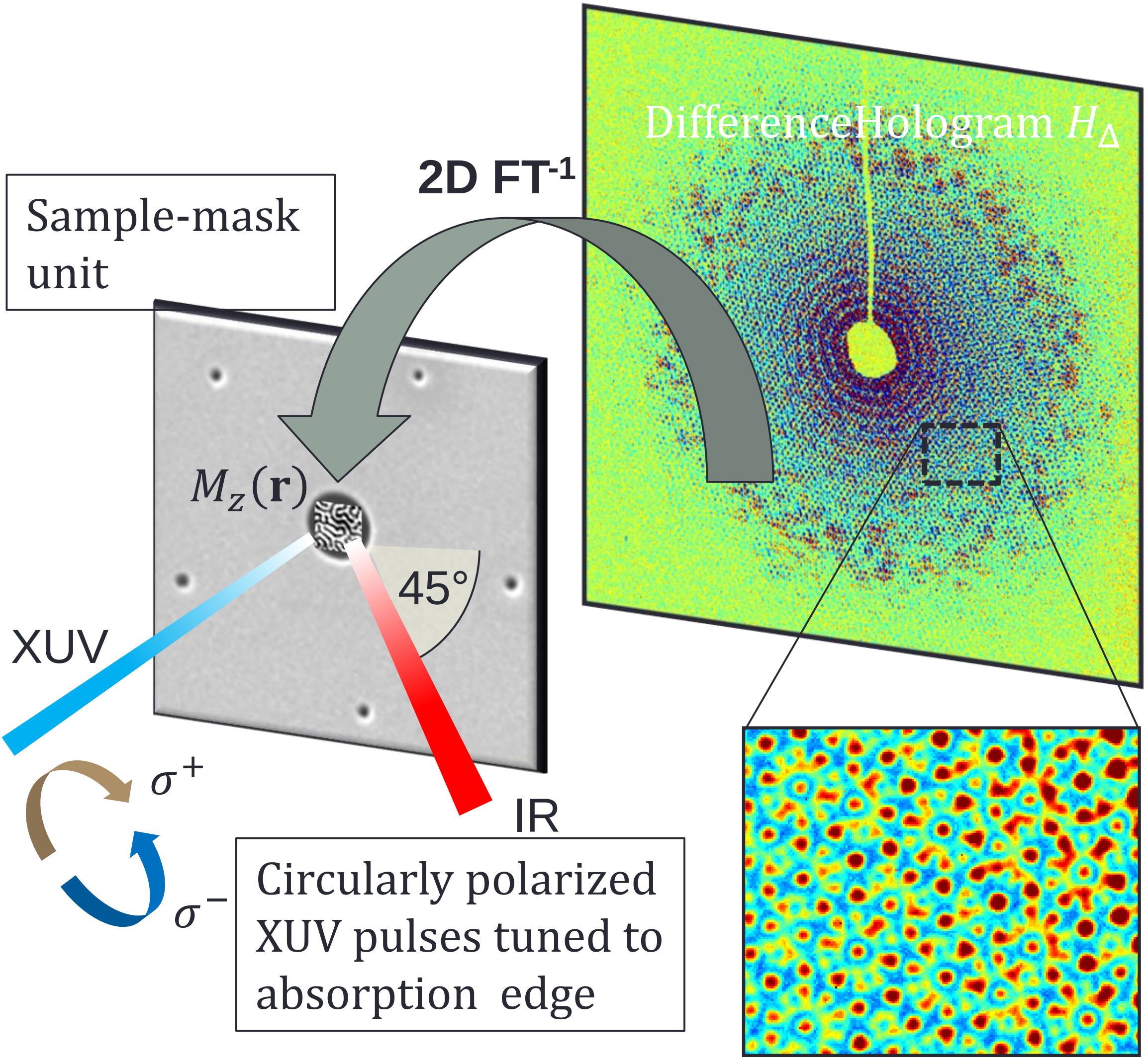


- FT of hologram shows Patterson map of the holographic mask with the magnetic structure
- object-hole autocorrelation in center
- 10 reference-hole-object-hole cross correlations (2 for each reference hole) arranged outside provide an image of magnetic domain pattern in object hole
- Large (small) diameter of reference holes yield high (low) contrast but low (high) resolution

DESY

Femtomagnetism

> Ultrafast demagnetization … of magnetic domain pattern -



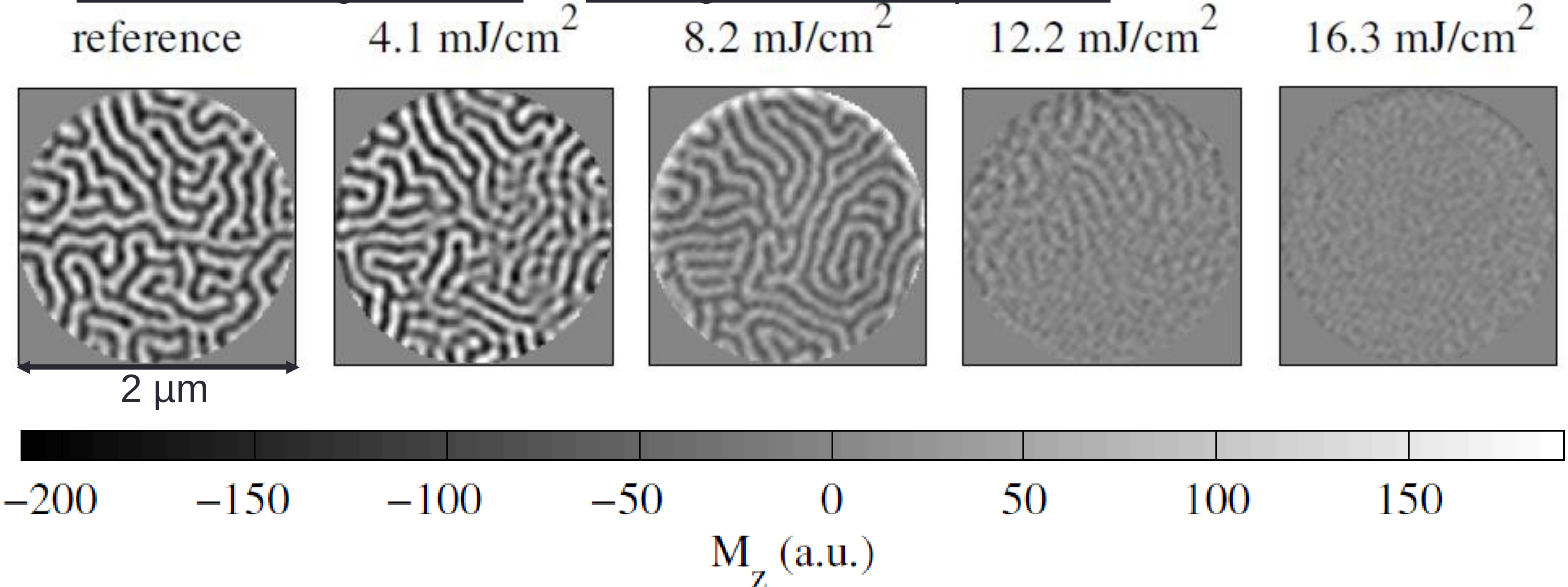
Experimental parameters at FERMI

- ~10x600 shots per helicity (@1 µJ due to extremely instable conditions of the FEL at that 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 -



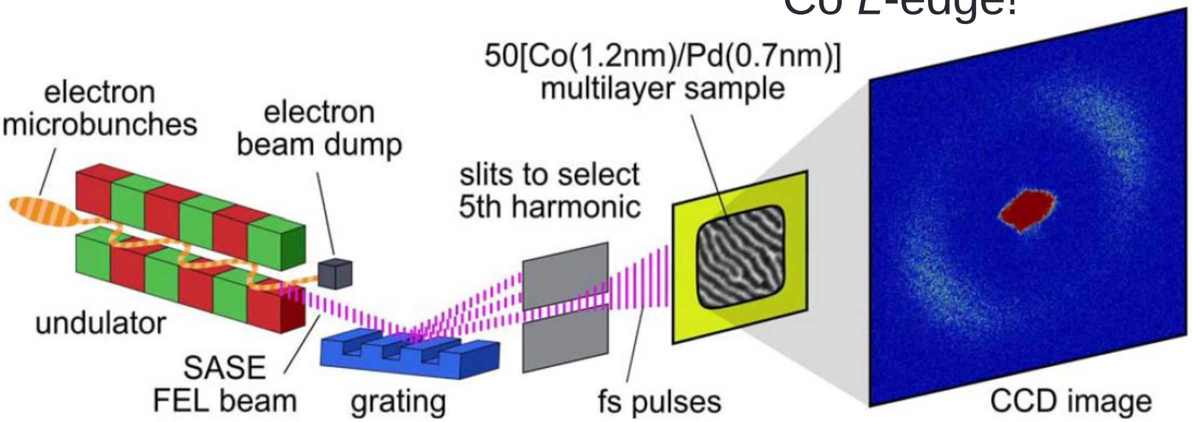
- Blurring due to domain rearrangements initiated by quasi-static heating (thermal demagnetization)
- Global contrast scale ∝ saturation magnetization ➔ ultrafast demagnetization on the nanoscale
- Small contrast at high fluences due to high demagnetization and/or domain rearrangements
- Resolution of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

unpublished

Femtomagnetism

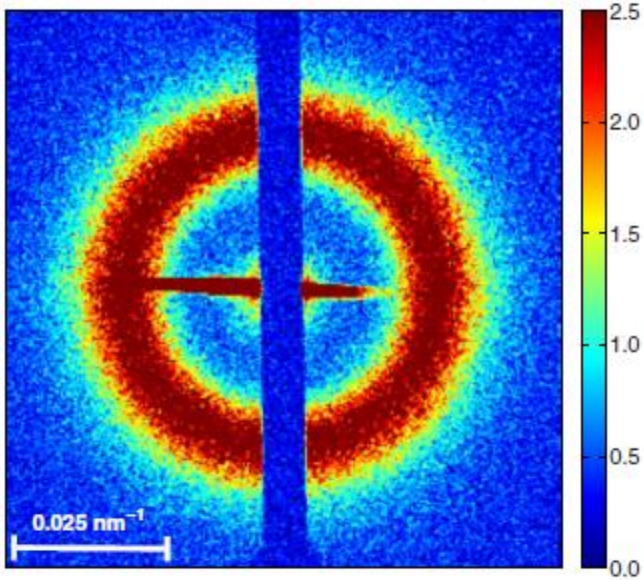
> High X-ray fluences

Co *L*-edge!

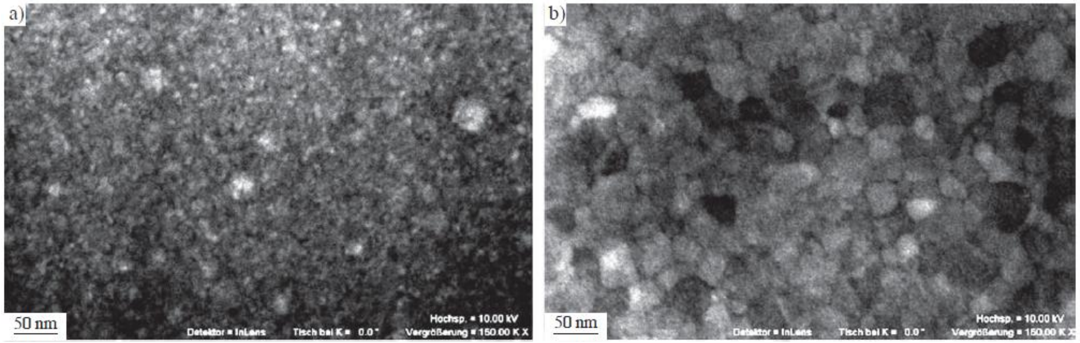


Establish first magnetic scattering at an FEL; due to experimental limitations only $7 \cdot 10^3$ photons per pulse at the sample
PRB **79**, 212406 (2009)

Experiments performed at **F**ree-electron **Las**er in **H**amburg, **FLASH**



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



Femtomagnetism

A Letters Journal Exploring the Frontiers of Physics
January 2015
EPL, 109 (2015) 17001
www.epljournal.org
doi: 10.1209/0295-5075/109/17001

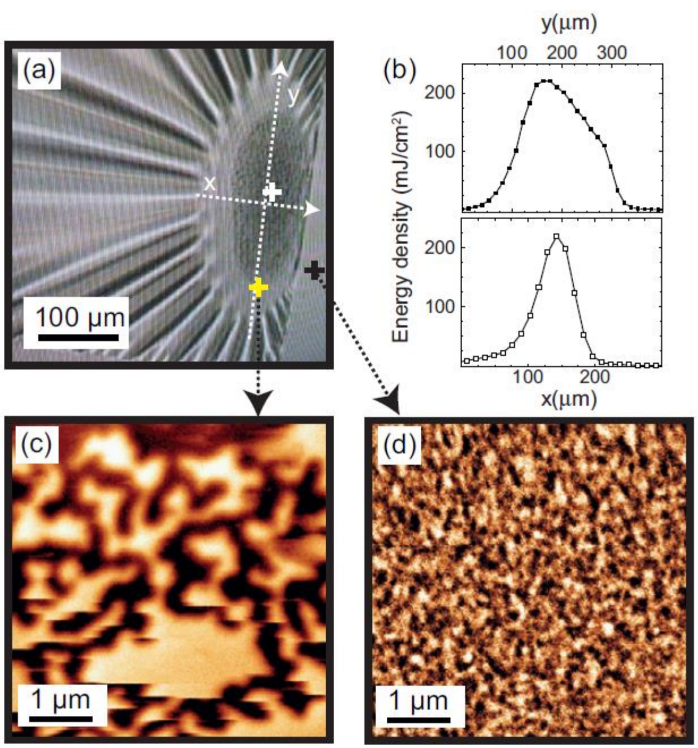
Femtosecond-laser–induced modifications in Co/Pt multilayers studied with tabletop resonant magnetic scattering

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 IR fluences

- Beam damage

Fluence of 200 mJ/cm² (IR pump)



➔ Some experiments can only be done in single-shot mode (sample destroyed after one pulse)
➔ Many membranes for systematic studies needed

➔ Need to care about unintentional impacts of pump AND probe beams

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]

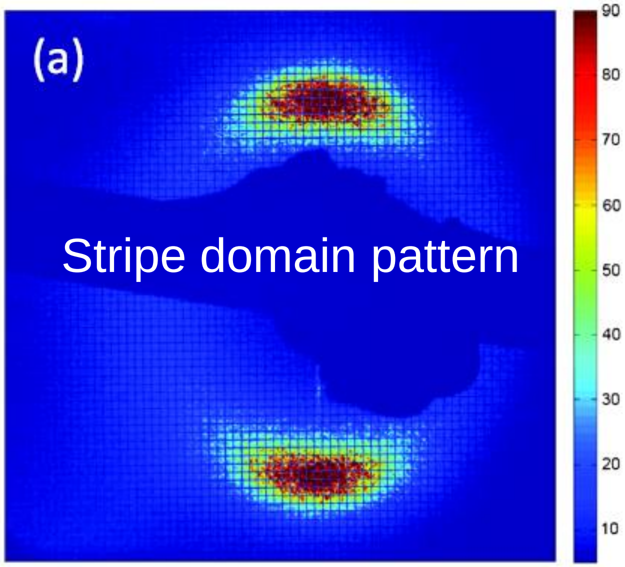
> High X-ray fluences

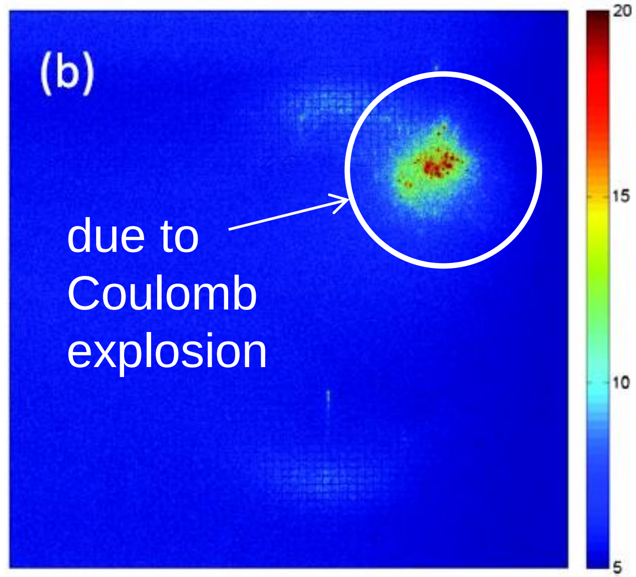
What is happening at high X-ray fluences (λ = 20.8 nm, i.e., M-edge of Co)?

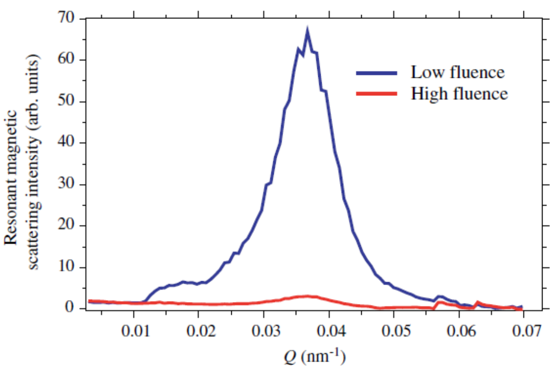
Experiment at FLASH (**Fr**ee-electron **Las**er in **H**amburg),pulse duration of ~100 fs

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 does not only act as a non-invasive probe

Femtomagnetism

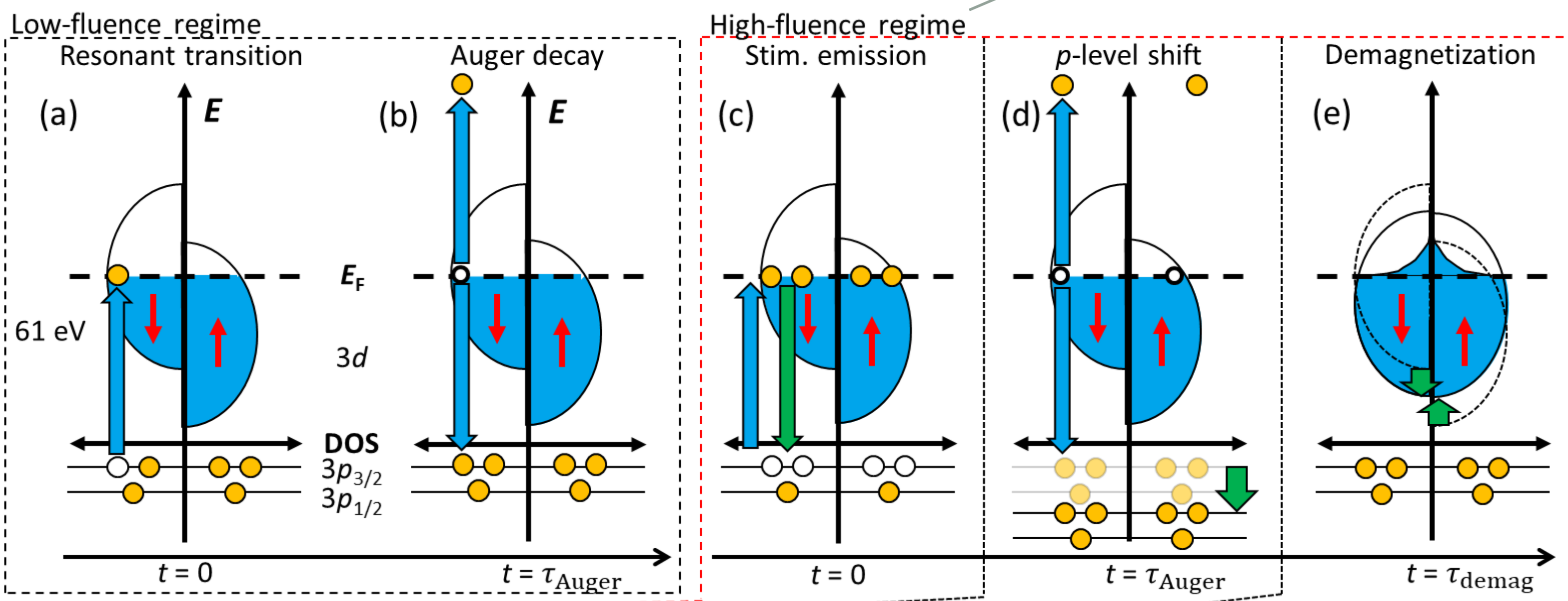
PRL 115, 107402 (2015) PHYSICAL REVIEW LETTERS week ending 4 SEPTEMBER 2015

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

J. Stöhr[1,*] and A. Scherz[2,†]

> High X-ray fluences

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



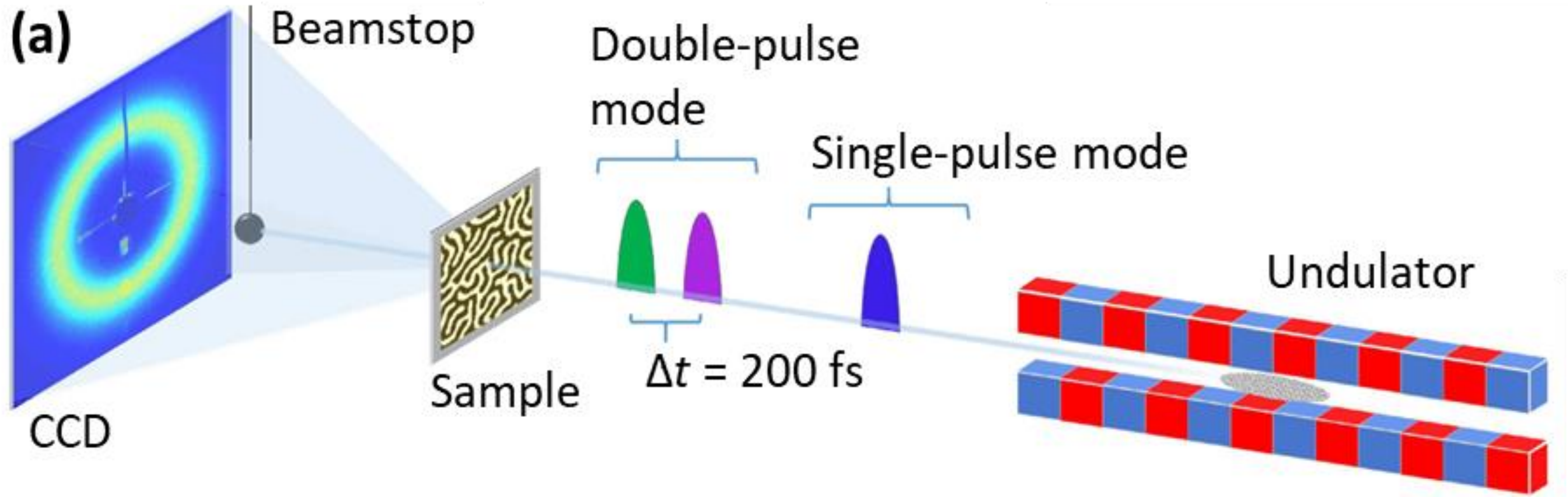
L. Müller et al., Phys. Rev. Lett. **110**, 234801 (2013)

DESY

Femtomagnetism

> High X-ray fluences

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



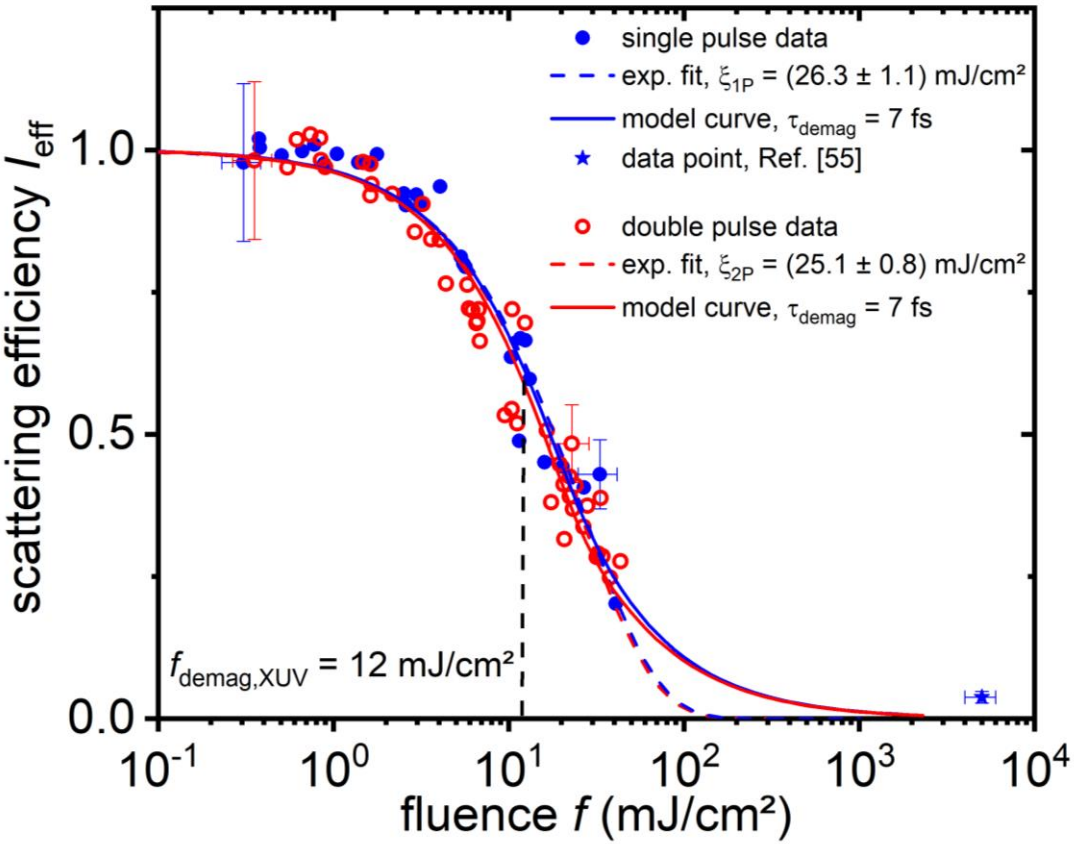
Experimental parameters at FERMI:

- **FEL single pulses & FEL double pulses** with **temporal delay of Δt = 200 fs**
- **Detuned conditions**: Photon energy E_{ph} shifted by ΔE = 1.4 eV with respect to E_{res}
- Fluence f variation: 0.3 mJ/cm² < f < 60 mJ/cm²

Femtomagnetism

> High X-ray fluences

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



- Quenching effect sets in at low fluences typically used in „classical“ pump-probe experiments!

➔ Effect superimposed on data!

- Similarity of both curves

➔ Demagnetization seems to quench the scattering intensity!

- Sub-20-fs demagnetization time

Under revision

Femtomagnetism

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

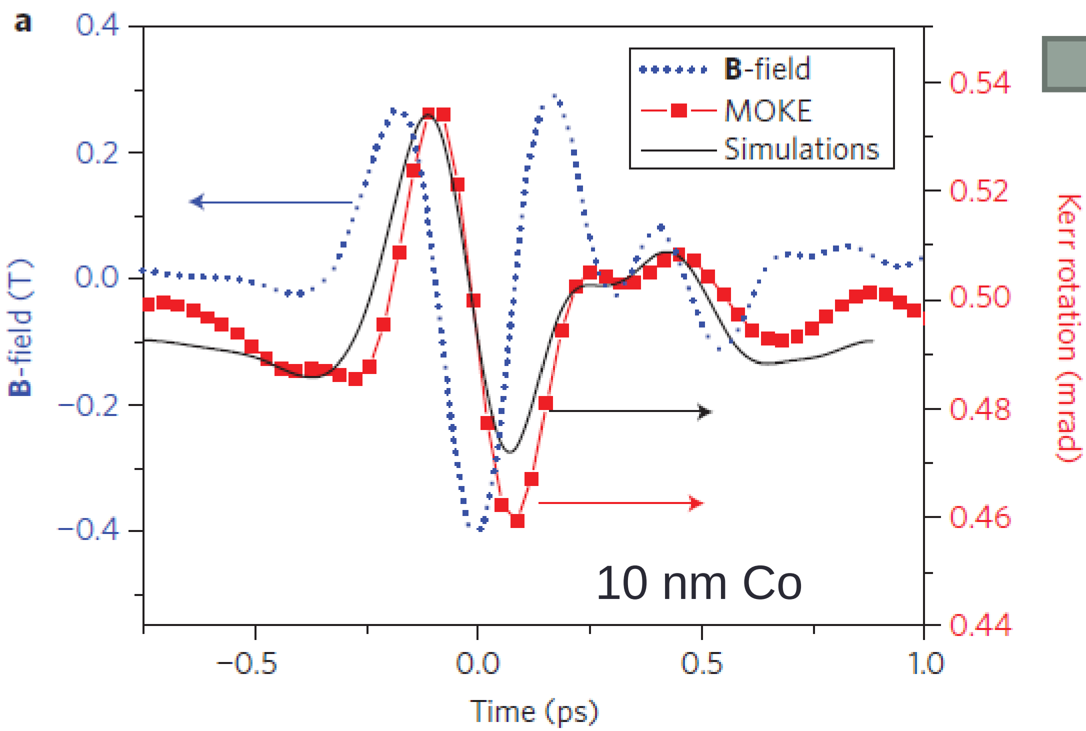
> THZ dynamics

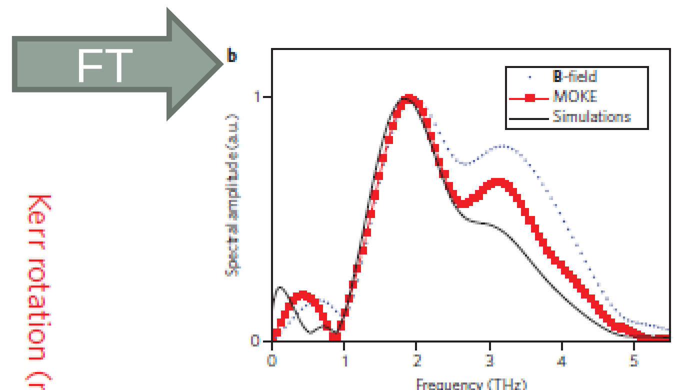
Idea and typical parameters

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

C. 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]*

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



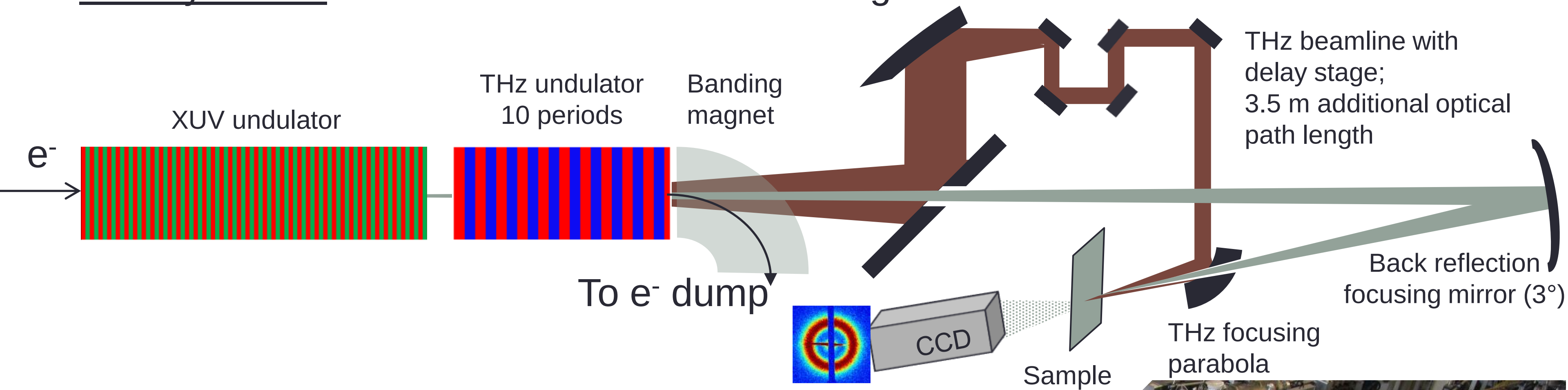


- Fluence of 10 mJ/cm²
➔ |**B**|-field of 200 mT
➔ |**E**|-field of 60 MV/m
➔ role of **E**-field?

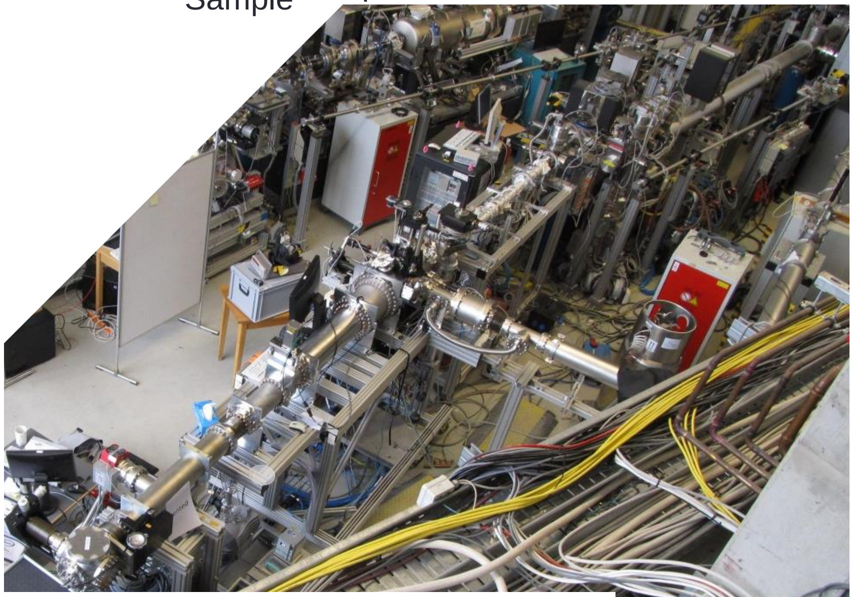
Femtomagnetism

> THz dynamics

What are we doing?

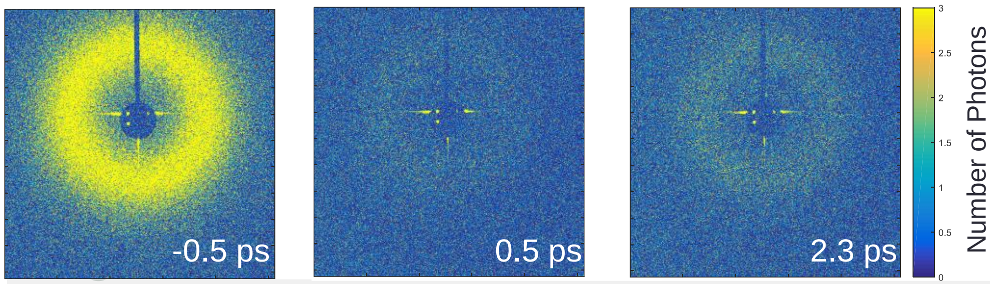


- THz radiation produced from same electron bunch as XUV by a separate electromagnetic undulator with 10 periods
  - Produces *10 full cycles of THz radiation*
- Intrinsic synchronization of XUV and THz pulse; jitter as low as 5 fs (rms)
  - Measurements under phase-stable conditions
- THz frequencies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

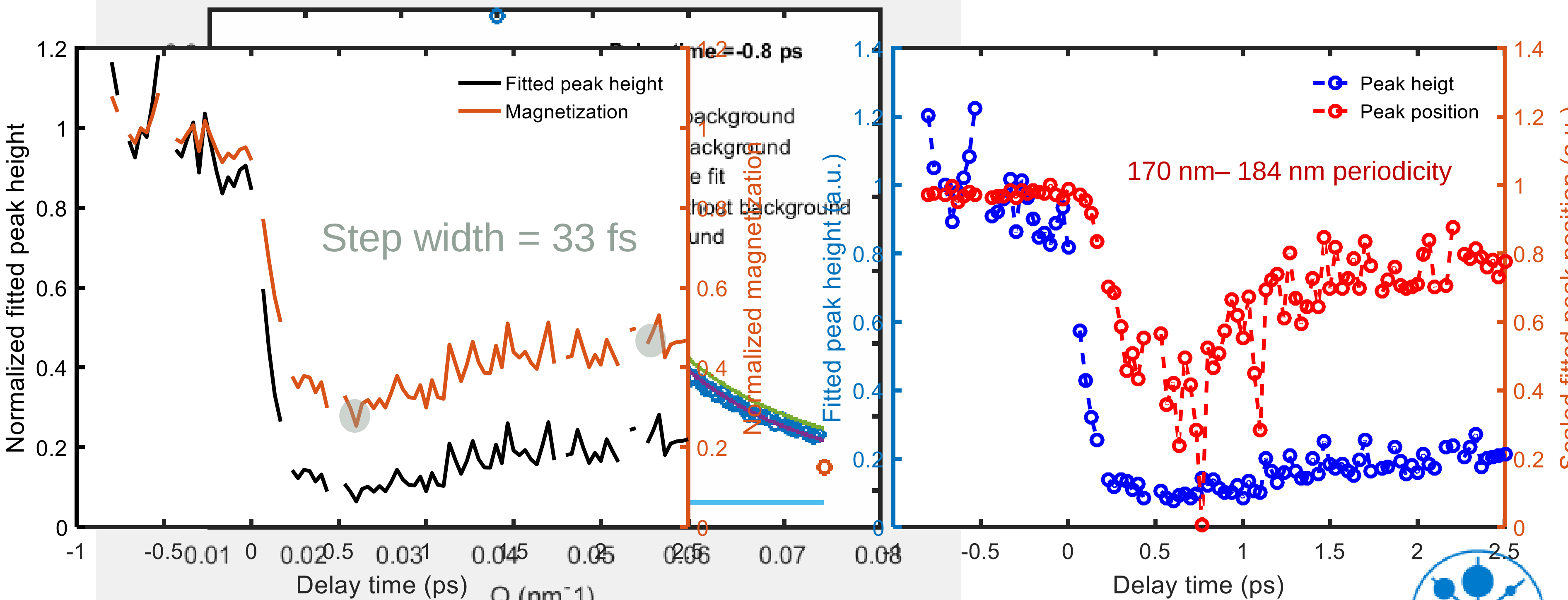


Femtomagnetism

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



Femtomagnetism

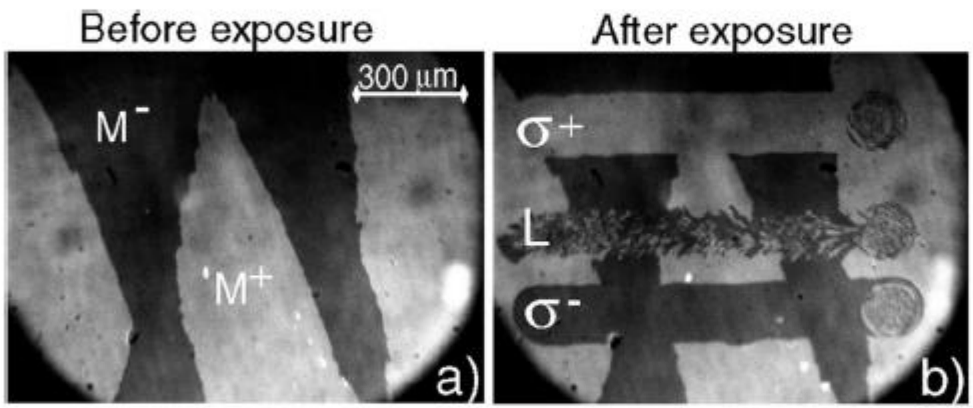
PRL 99, 047601 (2007) PHYSICAL REVIEW LETTERS week ending 27 JULY 2007

All-Optical Magnetic Recording with Circularly Polarized Light

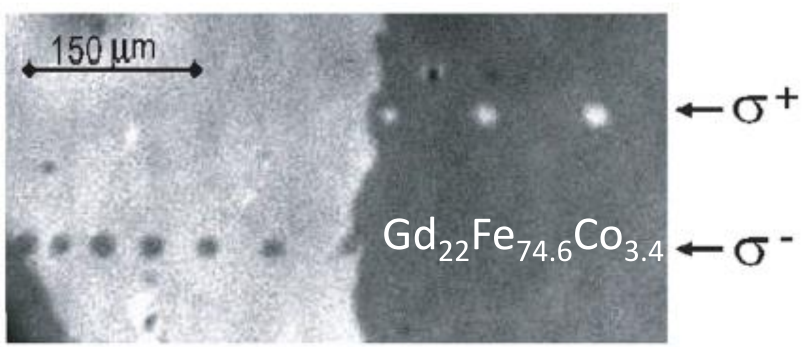
C. D. Stanciu,[1,*] F. Hansteen,[1] A. V. Kimel,[1] A. Kirilyuk,[1] A. Tsukamoto,[2] A. Itoh,[2] and Th. Rasing[1]
[1]Institute for Molecules and Materials, Radboud University Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands
[2]College of Science and Technology, Nihon University, 7-24-1 Funabashi,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

> All-optical (helicity-dependent) switching

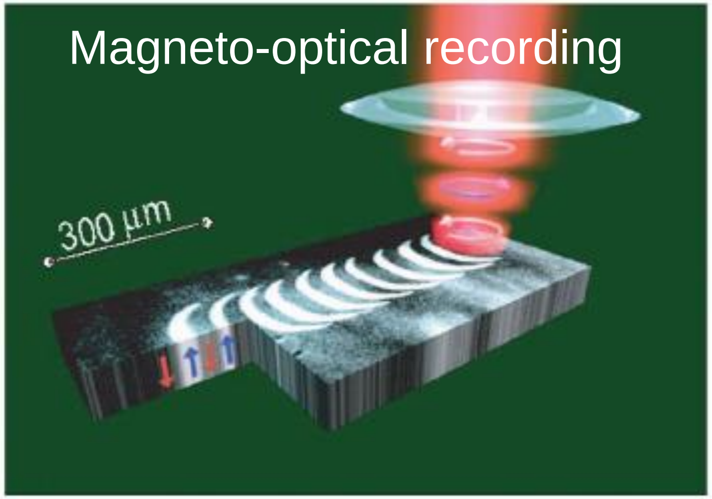
- Discovery for ferrimagnetic materials



Multi-shot



Single-shot



DESY

Femtomagnetism

> All-optical switching

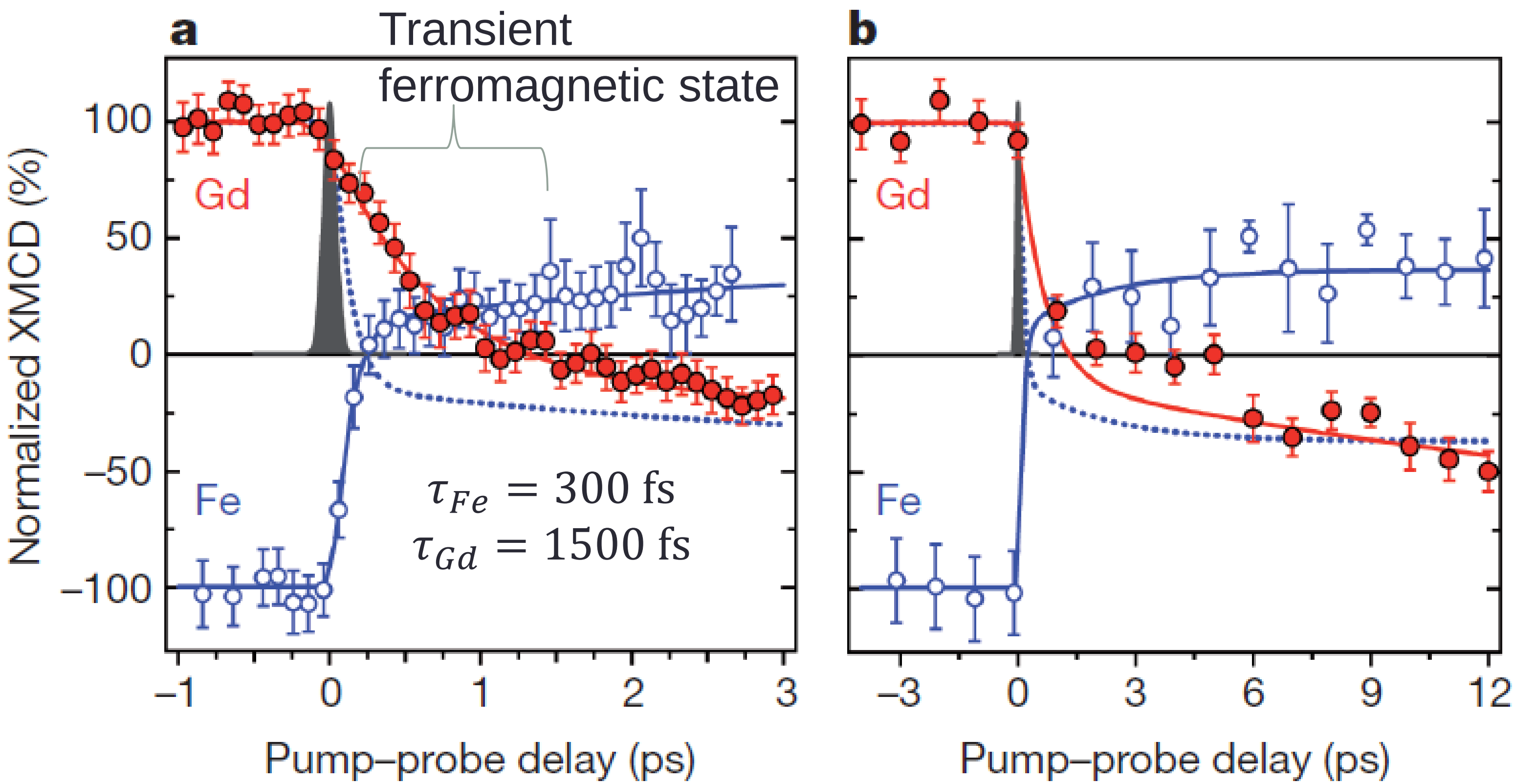
- Time-resolved and element-selective studies

doi:10.1038/nature09901

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]



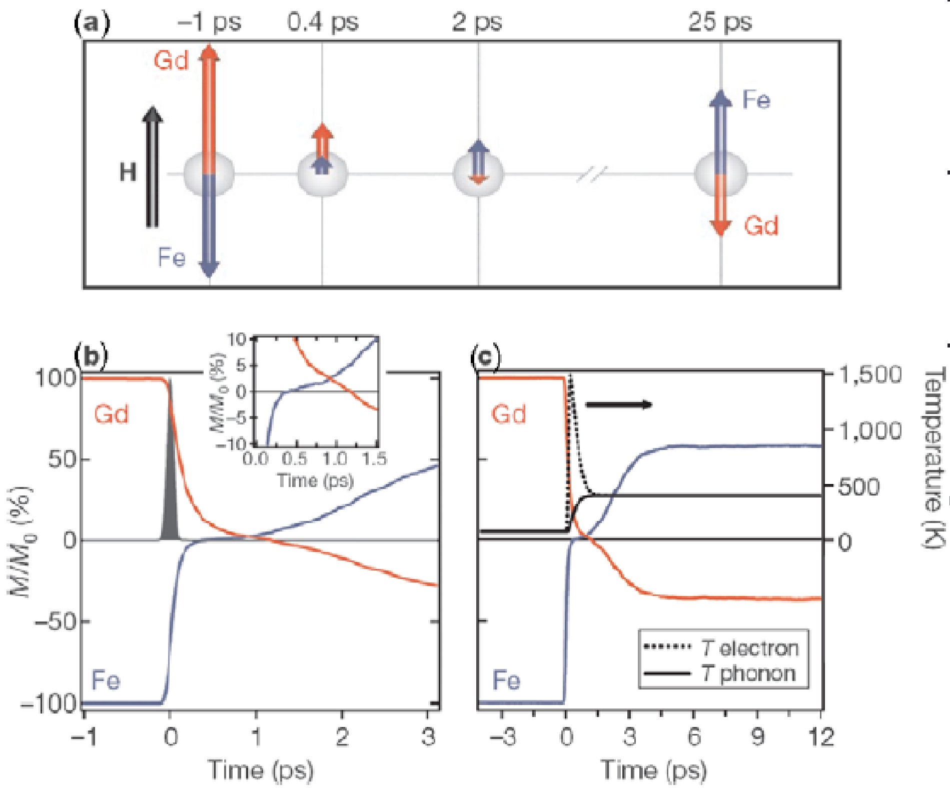
Femtomagnetism

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

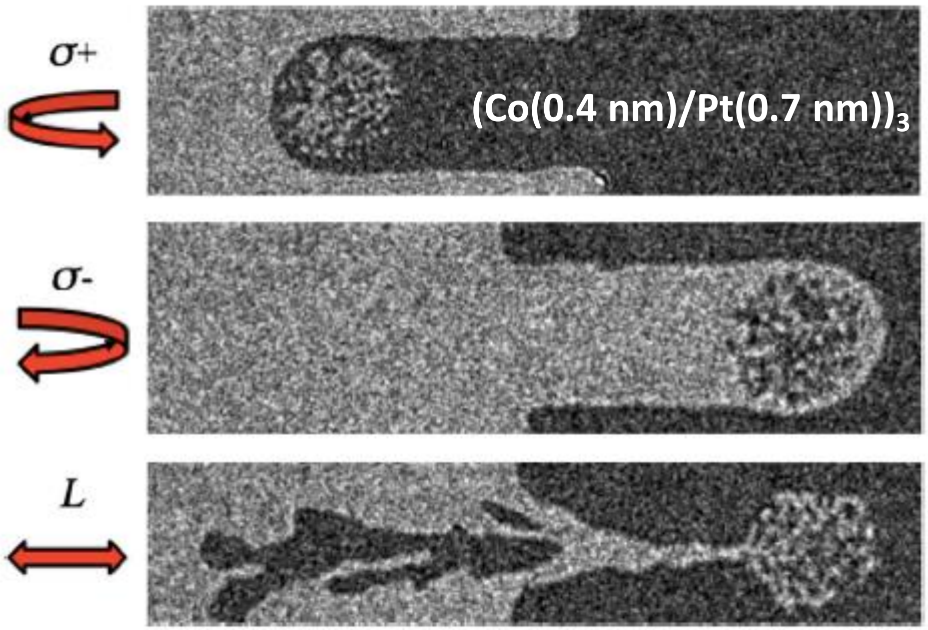
MAGNETISM

All-optical control of ferromagnetic thin films and nanostructures

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

> <u>All-optical switching</u>

- Is there all-optical switching for ferromagnetic materials?



Multi-shot

<u>Many open questions, like e.g.</u>:
- 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… BUT a PhD thesis will be submitted tomorrow.