Magnetization dynamics
and
fs pulsed X-ray sources

Jan Lüning

Laboratoire de Chimie Physique – Matière et Rayonnement
Université Pierre et Marie Curie, Paris

and

Synchrotron SOLEIL

Studying fs-laser pulse driven ultrafast magnetization dynamics in real time with fs-short X-ray pulses

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Synchrotron SOLEIL

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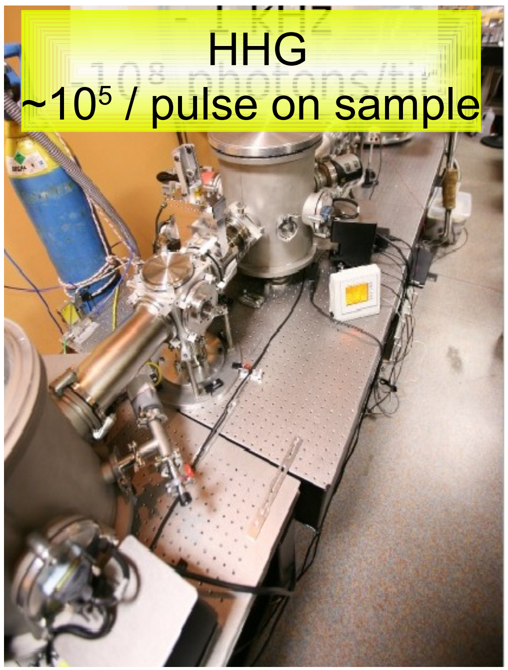
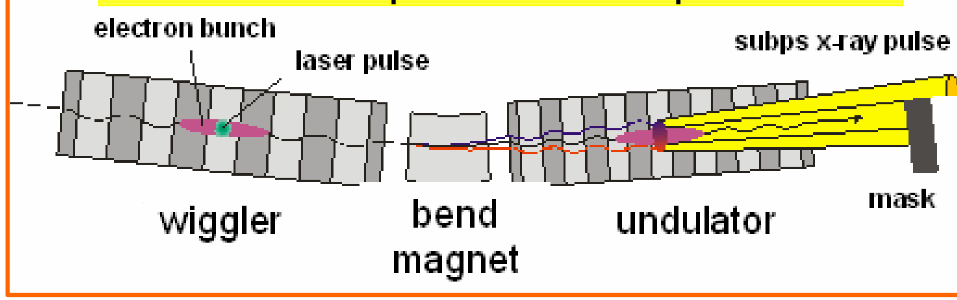
Combine *nanometer spatial* resolution with *femtosecond temporal* resolution



HHG
~10^5 / pulse on sample

Femtoslicing (BESSY, SLS, ***SOLEIL***)
~10^6 /pulse on sample



FLASH / LCLS / FERMI / SACLA
~10^{12} /pulse on sample

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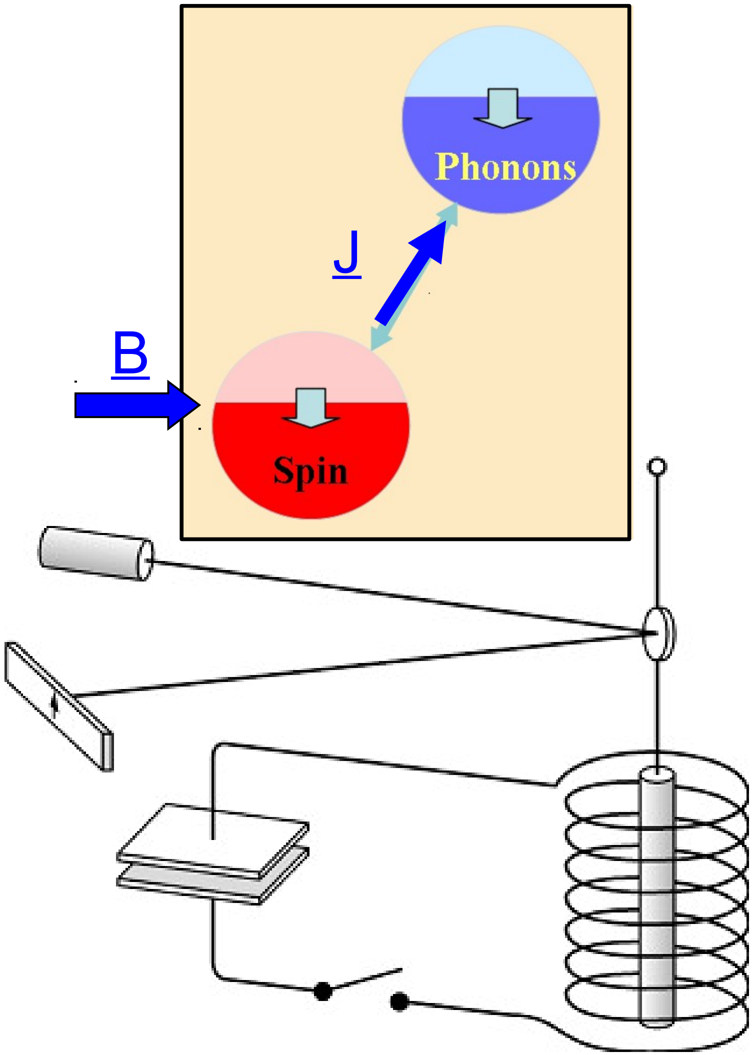
Einstein – de Haas effect



Experimenteller Nachweis der
Ampereschen Molekularströme
A. Einstein, W. J. de Haas,
DPG Verhandlungen 17, 152 (1915)

Magnetization reversal implies change of (orbital and spin) angular momentum!

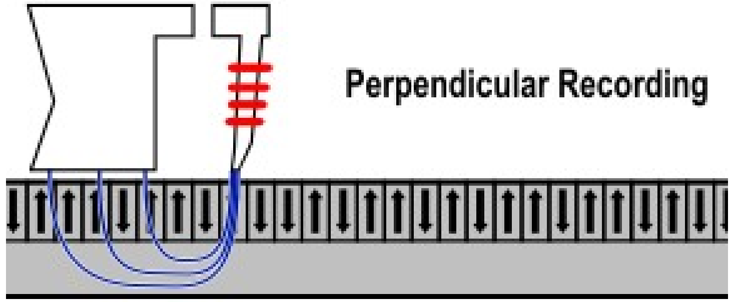
Picture from Wikipedia

Transfer to lattice via magnon-phonon scattering!

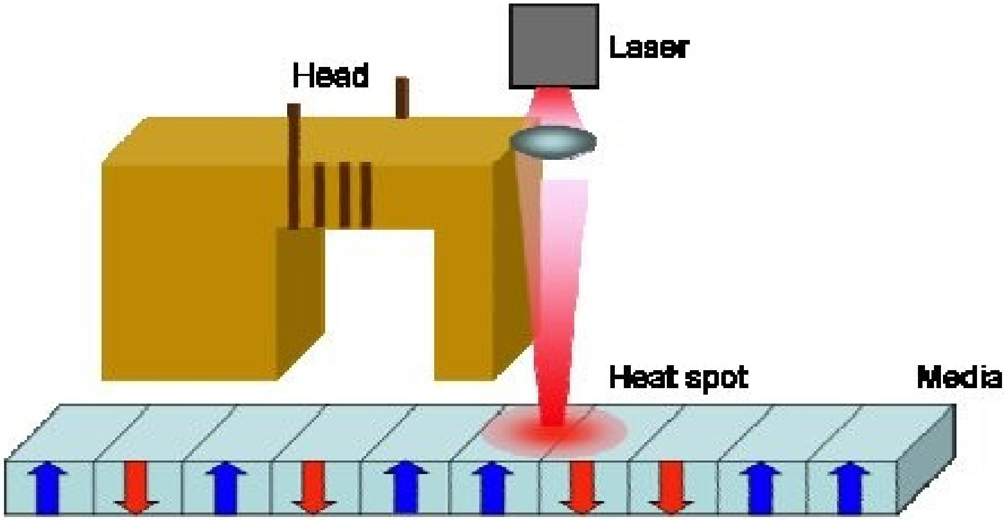
The next generation: Heat Assisted Magnetic Recording

To further decrease bit size, but keep long-term stability:

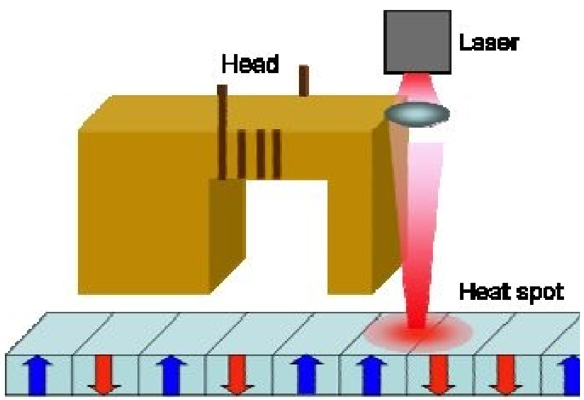
Use magnetically *harder* materials



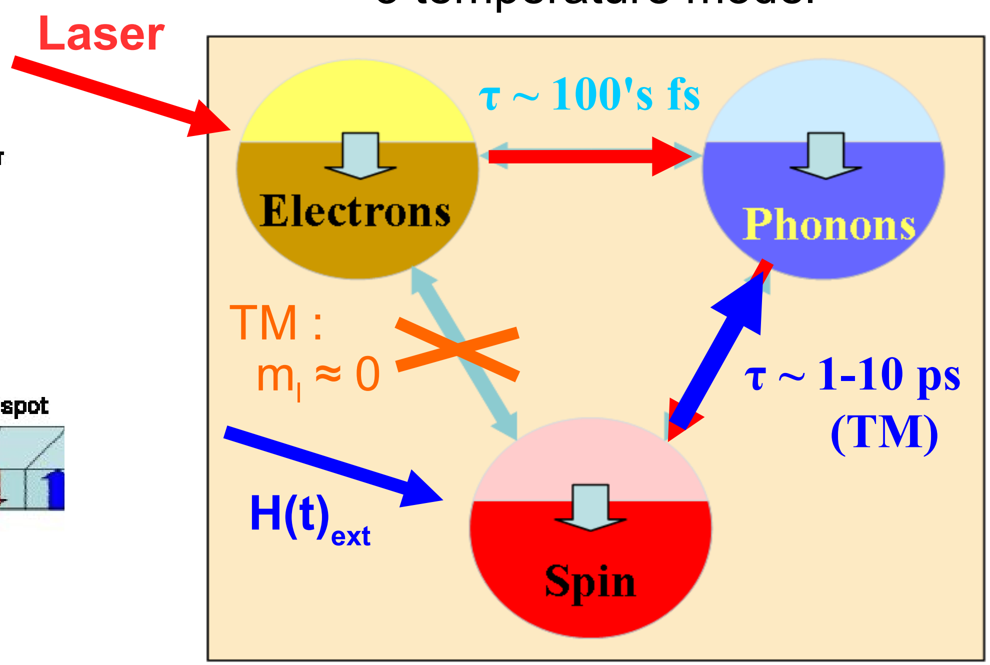
Heat sample to *temporally* lower *locally* the magnetic 'hardness'



Magnetization reversal by magnetic field pulse



3 temperature model



NOTE: Magnetization switching caused by external ***magnetic*** field pulse acting on spin system!

QUESTION: Could we manipulate magnetization with ***only*** a laser pulse?

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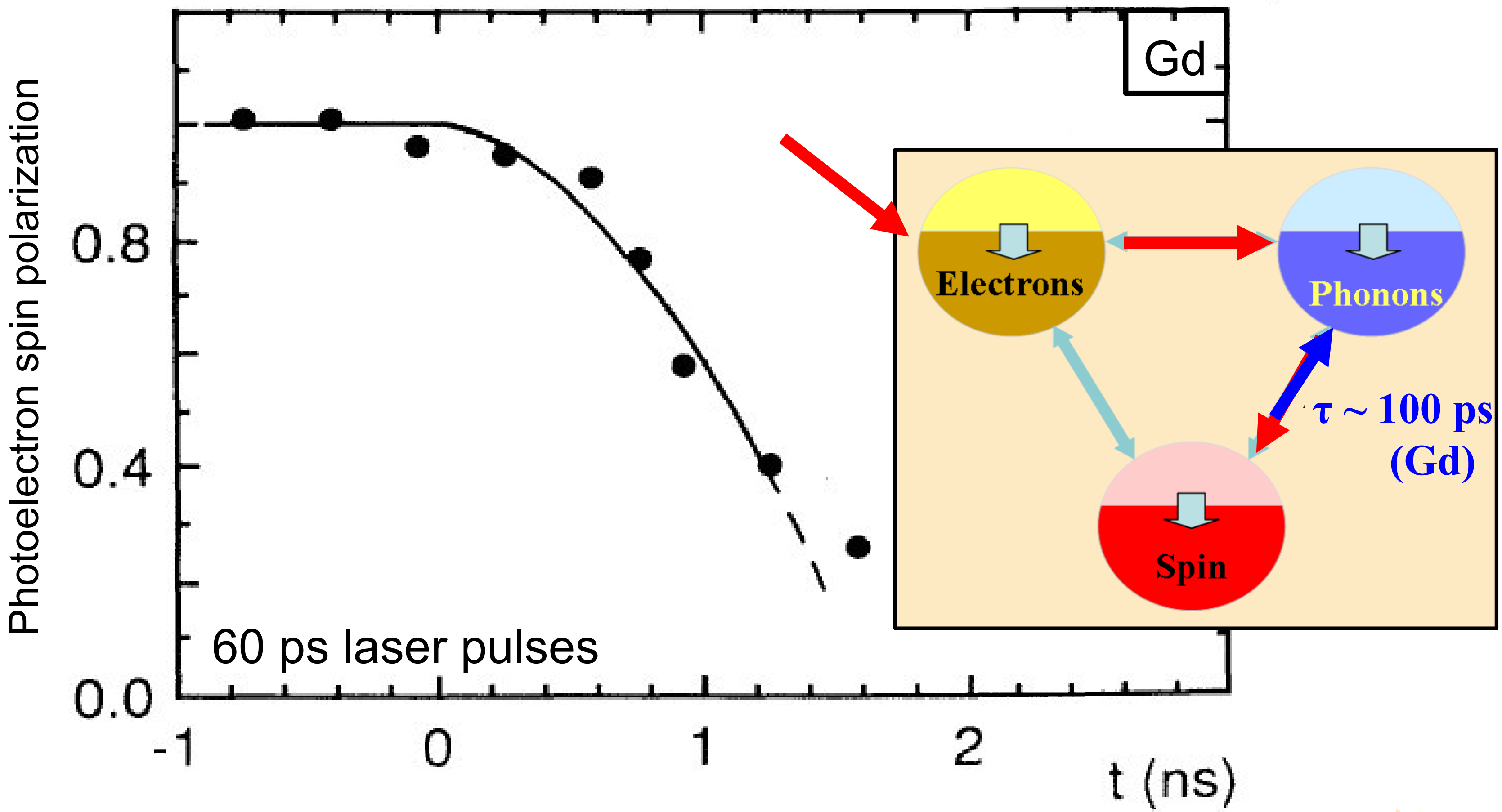
1992: Spin-lattice coupling in Gd ~100 ps

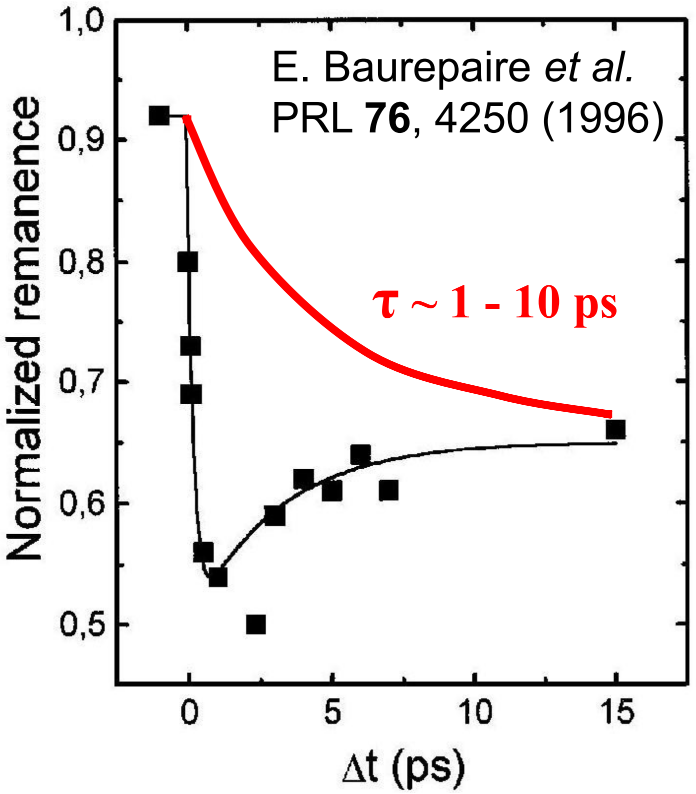
VOLUME 67, NUMBER 23 PHYSICAL REVIEW LETTERS 2 DECEMBER 1991

Spin-Lattice Relaxation Time of Ferromagnetic Gadolinium Determined with Time-Resolved Spin-Polarized Photoemission

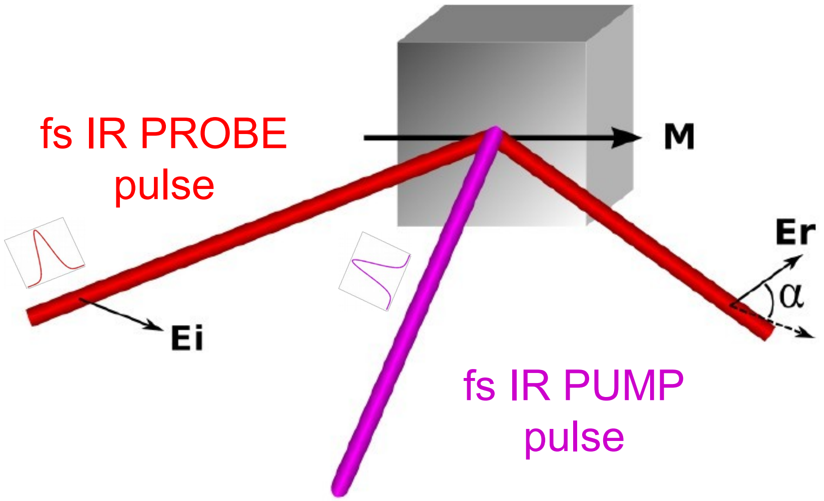
A. Vaterlaus, T. Beutler, and F. Meier



1996: Discovery of ultrafast magnetization dynamics



All-optical fs time resolved
IR pump – MOKE-probe experiment



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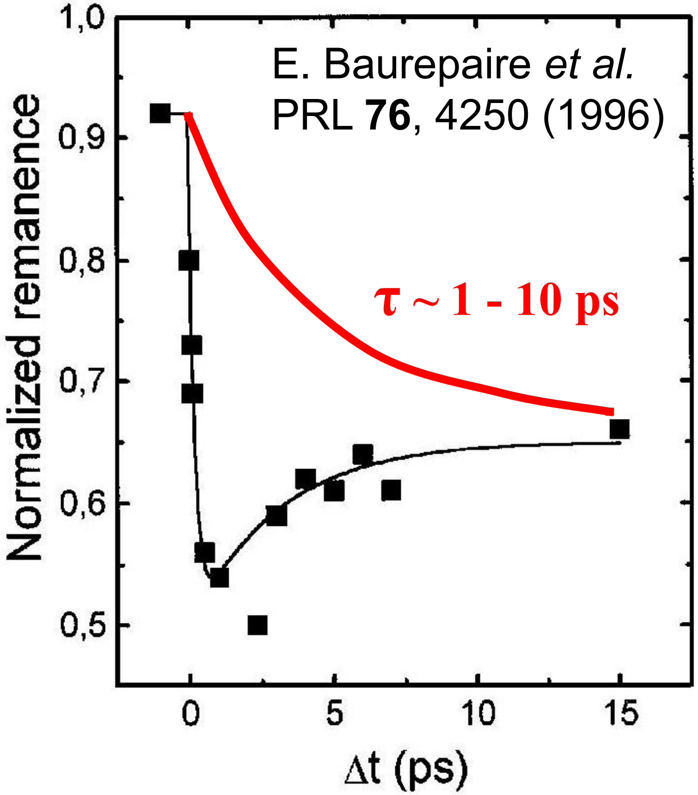
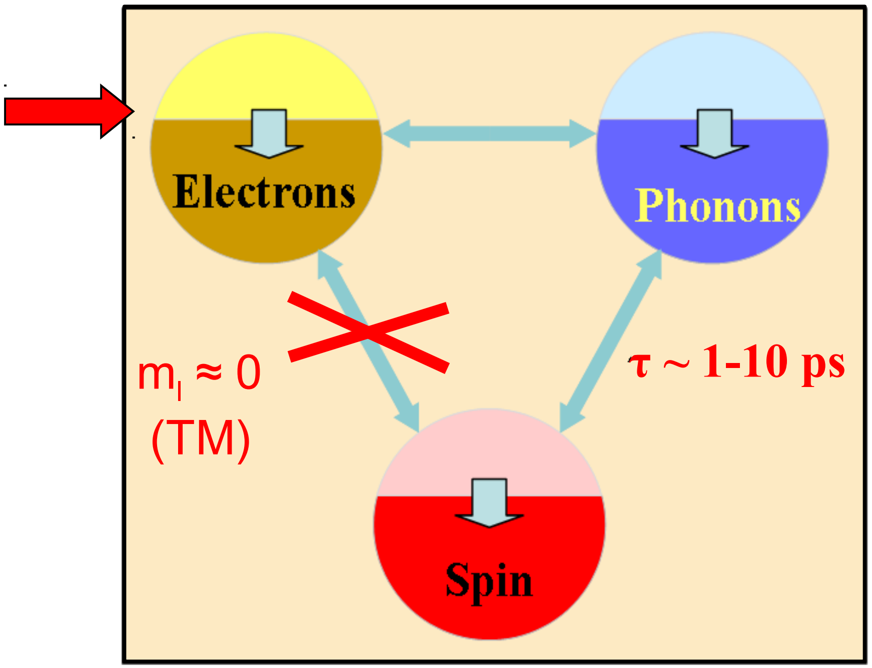


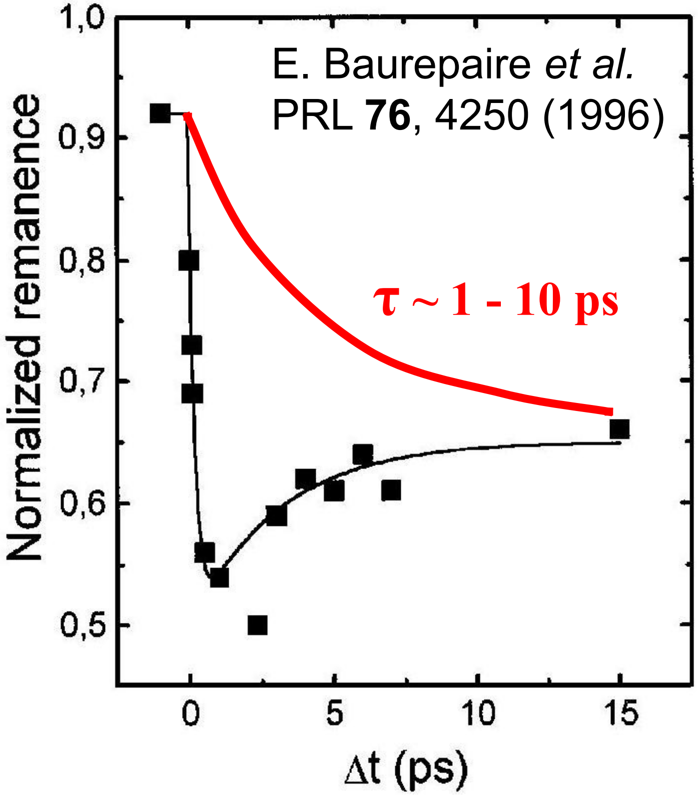


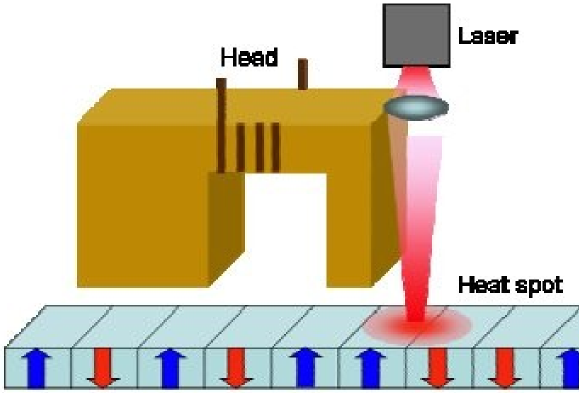
Questions still discussed since 1996:

- How does energy flow into the spin system?
- What happens to the angular momentum on femtosecond time scale?

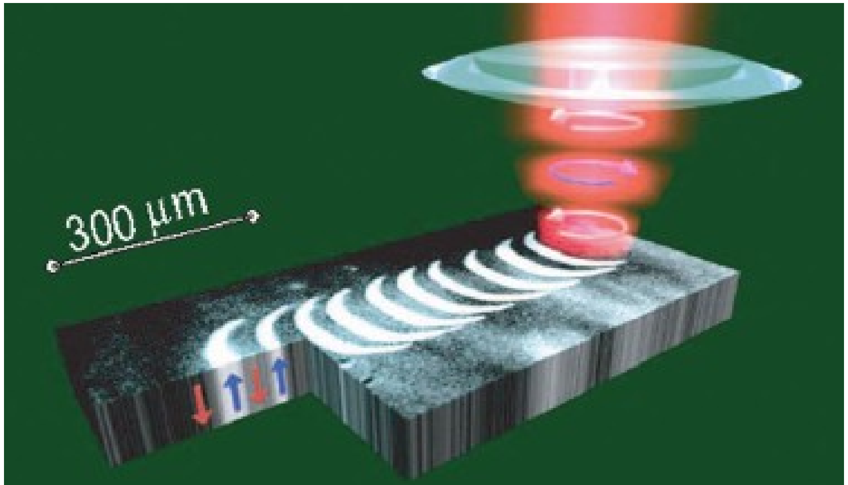
2007: Discovery of ultrafast magnetization dynamics

From ultrafast demagnetization ….





… to ultrafast magnetization *CONTROL*



C.D. Stanciu *et al.*, PRL 99, 047601 (2007)

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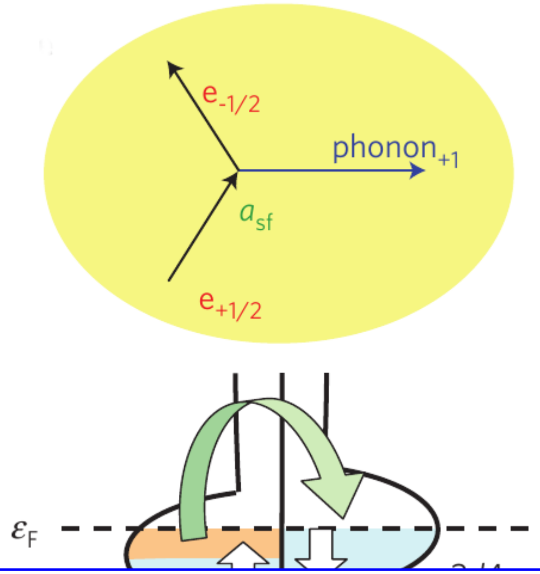
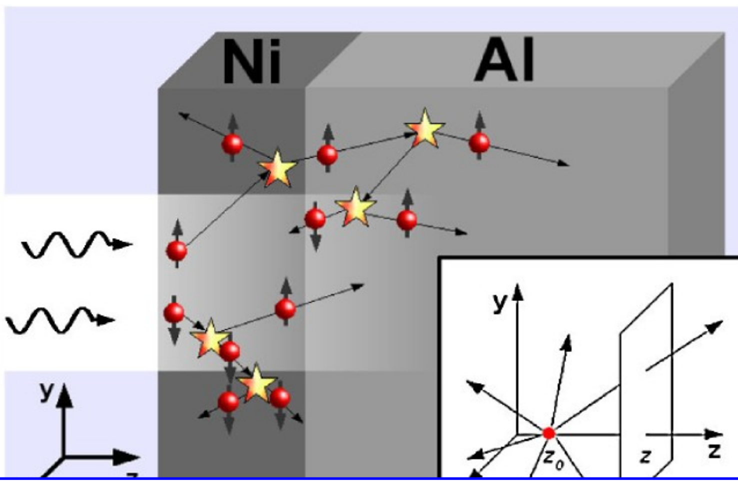
Most discussed potential mechanisms

Elliott - Yafet like spin-flip electron - phonon scattering (local mechanism)

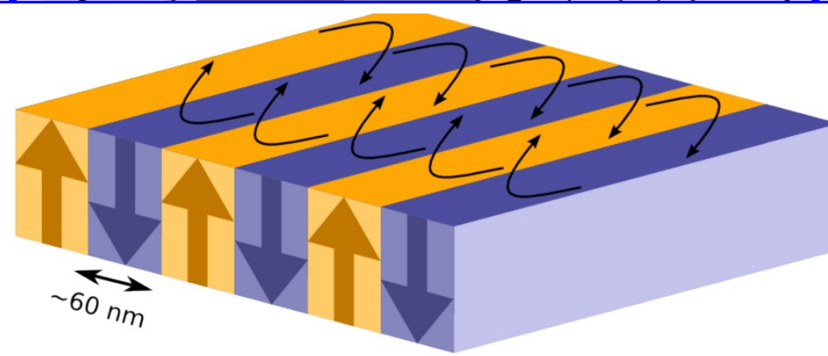


Angular momentum transport by hot, spin-polarized electrons (non-local mechanism)

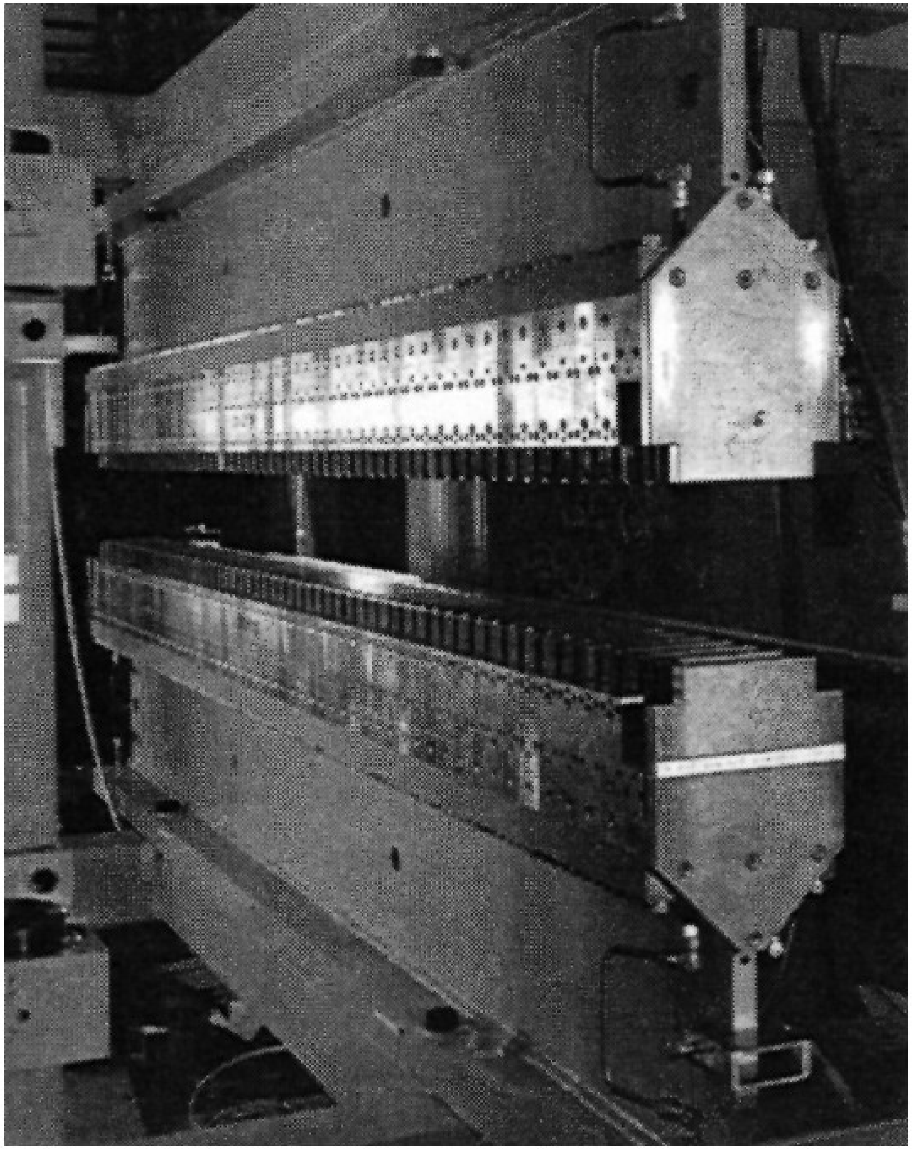


- Requires ~10 nm spatial resolution
- Element sensitivity
- Strong dichroism signal

→ properties of X-ray based techniques



Synchrotron Radiation



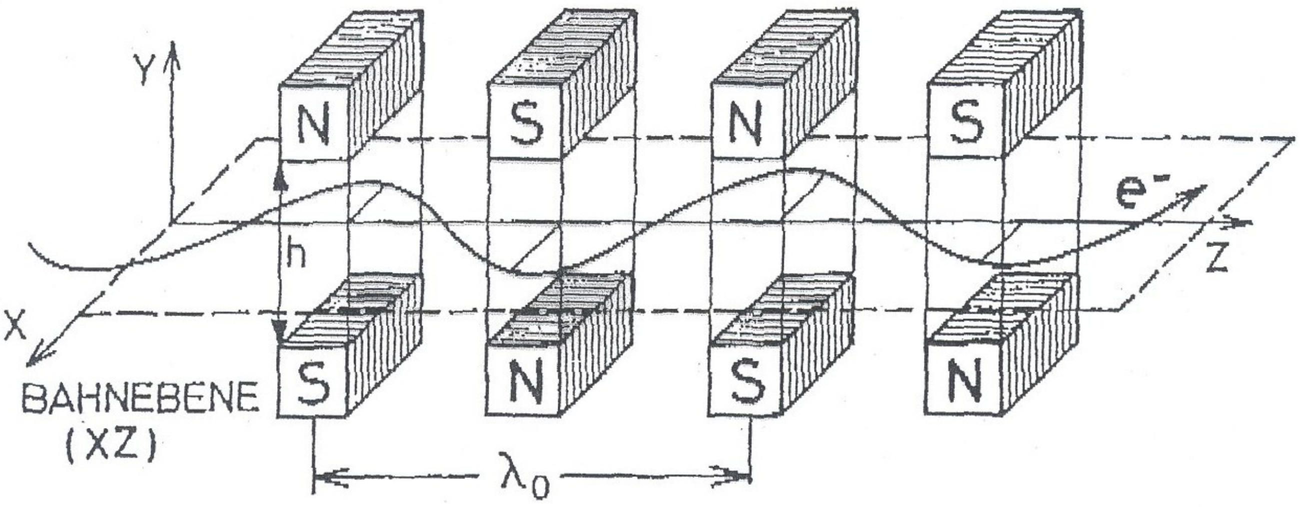
LBNL/EXXON/SSRL (1982), SSRL Beamline VI
55 pole (N = 27.5), λ_w = 7 cm

Insertion devices of 3rd generation sources provide X-ray beams with:

- Flux: 10^{14} ph / (sec·0.1% BW)
  → $10^6 – 10^8$ pulses / sec
- Brilliance:
  10^{22} ph / (sec·0.1%·BW·mrad²·mm²)
  → low coherence degree (deg. < 1)
- Polarization control
- Time structure:
  ~50 ps X-ray flashes, ns-μs spacing
  with few photons:
  - few ps in low-alpha
  - ~150 fs in femtoslicing

→ inadequate for fs dynamics

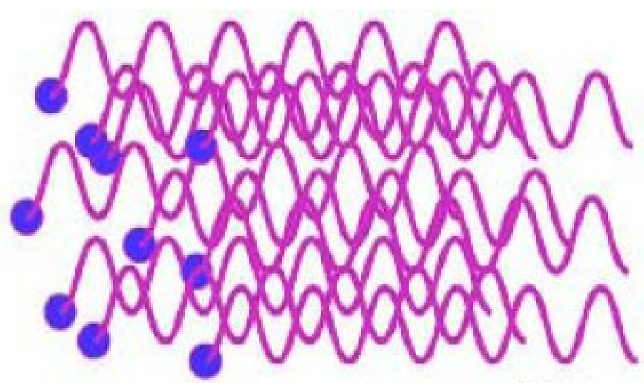
Synchrotron radiation of an undulator

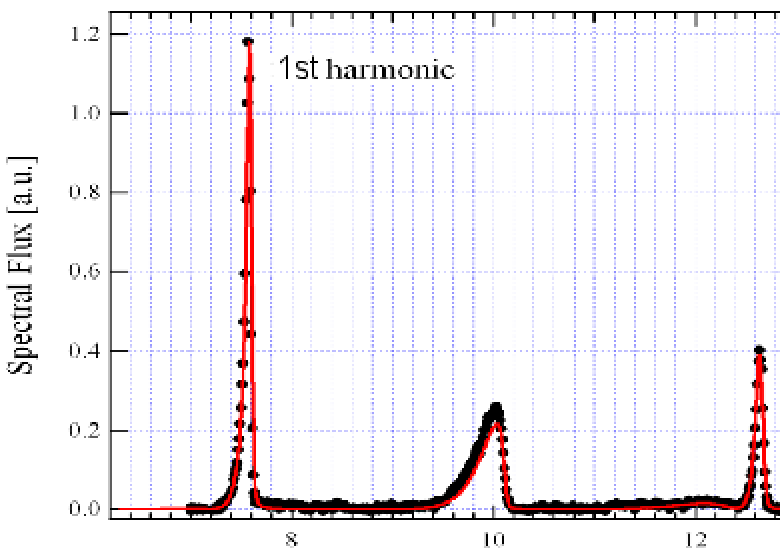


Spontaneous emission

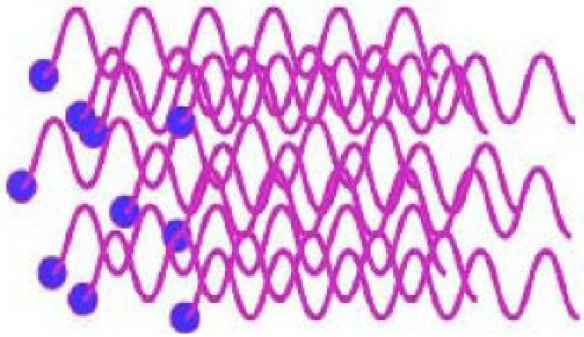
Note: each electron interferes within undulator with radiation emitted by itself!

$N_e \sim 10^9$ $I \sim N_e \cdot N^2$ $N \sim 10^2$

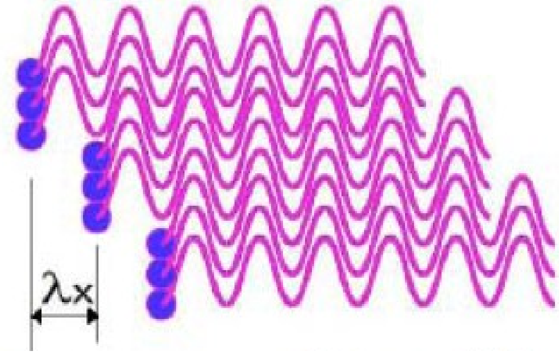




Self Amplified Spontaneous Emission (SASE XFEL)

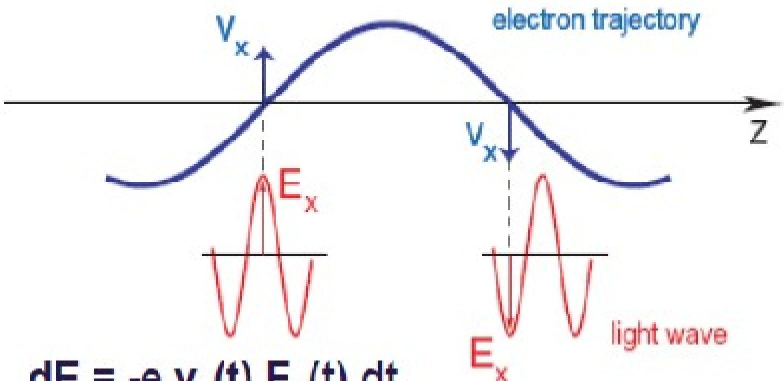


$I \sim (N_e) \cdot (N_u)^2$



$I \sim (N_e)^2 \cdot (N_u)^2$

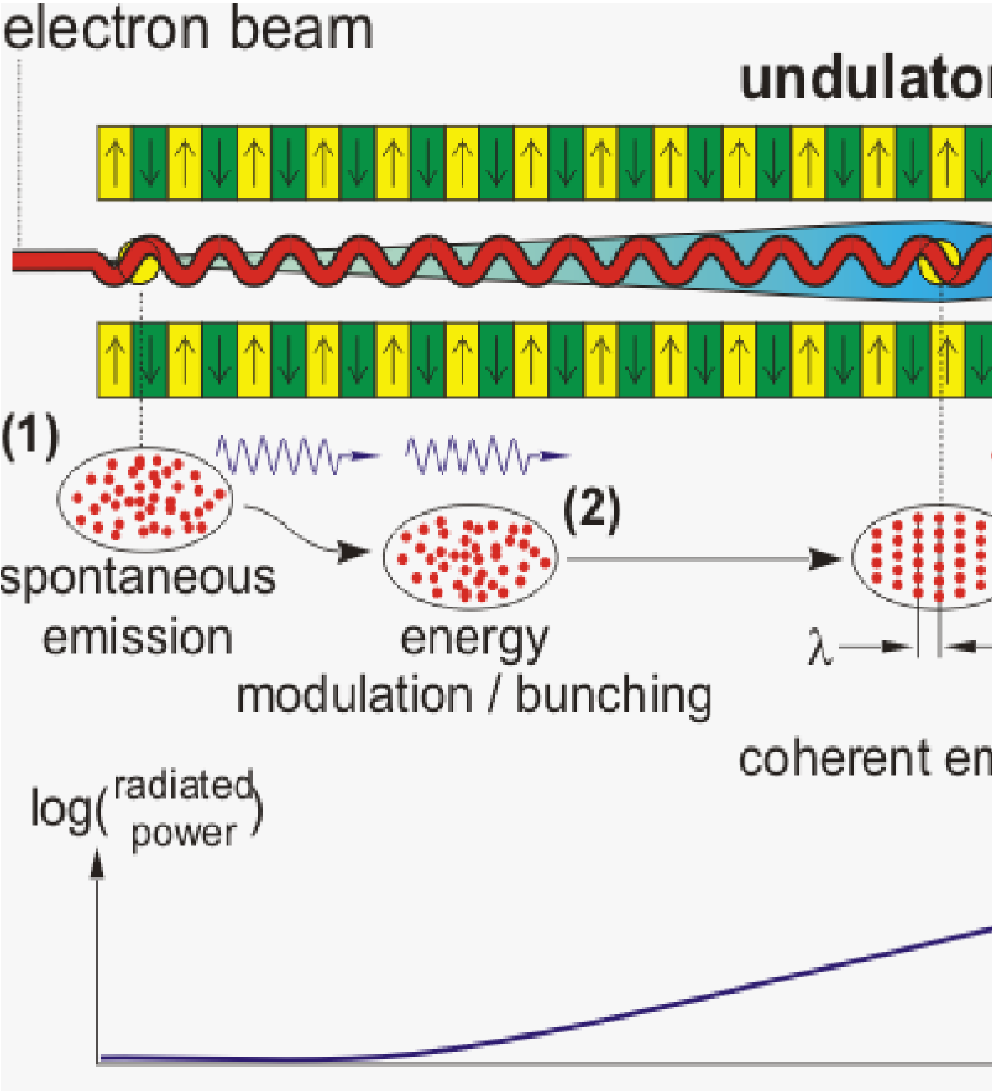
FEL idea: line up electrons by interaction with x-ray field

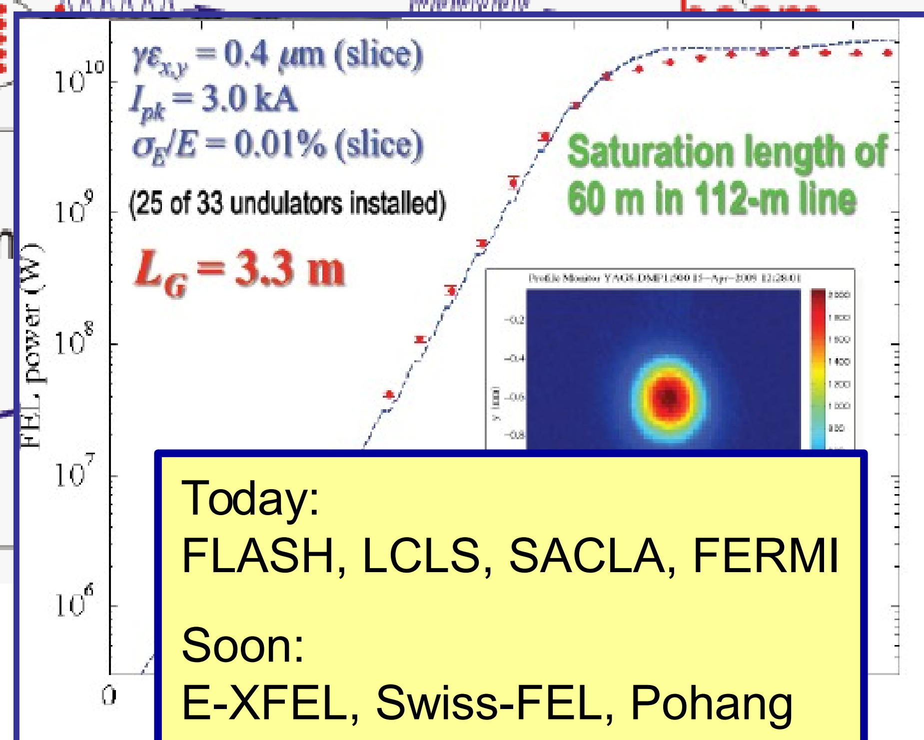


$dE = -e\, v_x(t)\, E_x(t)\, dt$

phase $E_x(t)$, $v_x(t)$ const. ⇒ acceleration & deceleration

SASE-XFEL – a very long undulator





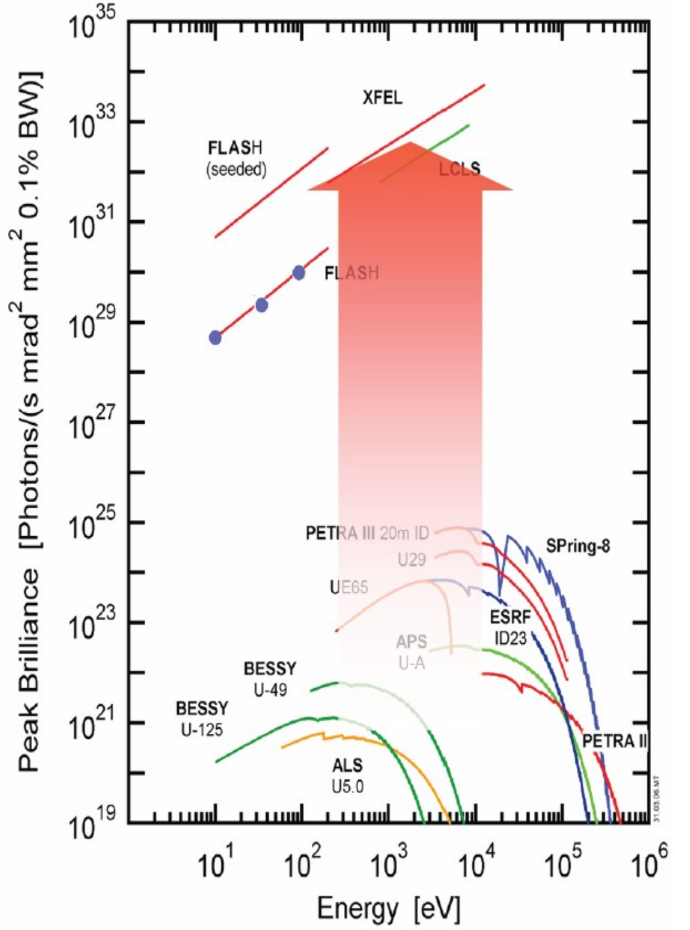
Coherent source → Intensity ~ (# of e$^-$)^2

X-ray Free Electron Lasers

- ~10^{13} photons/pulse
- fsec pulse duration (exp. < 2 fs)
- 100% transverse coherence (exp. 80%)

BUT: XFELs will **NOT** replace synchrotron radiation storage ring sources!

- 'single' user operation
- all parameters fluctuate
- not a gentle probe
- ...



Outline

- Ultrafast Magnetization Dynamics
- X-ray Free Electron Laser
- Combining fs temporal and nm spatial resolution by resonant magnetic (small angle) X-ray scattering
- *Strong* IR pumping
- X-ray streaking
- *Strong* X-ray probing

Acknowledgement

LCPMR	- *B. Vodungbo*, S. Chiuzbaian, R. Delaunay, ...
Synch. SOLEIL	- N. Jaouen, F. Sirotti, M. Sacchi…
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LOA Palaiseau	- J. Gautier, P. Zeitoun, ...
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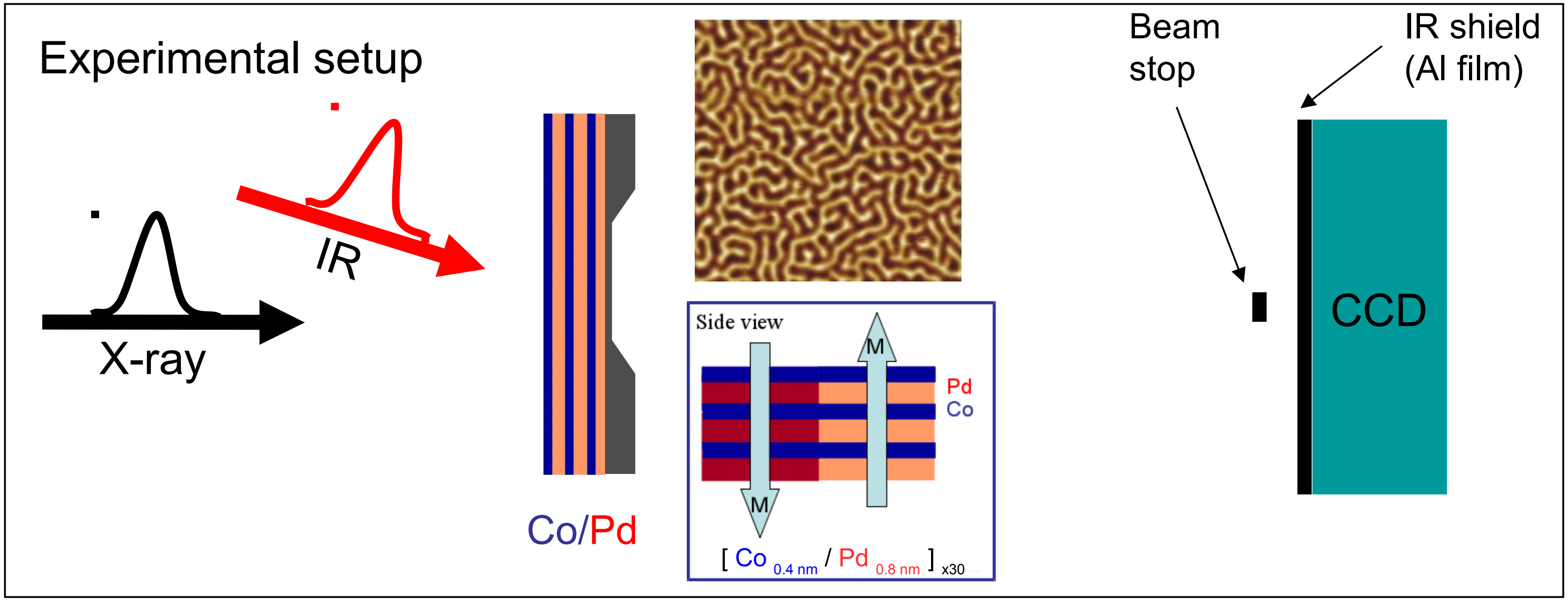
LMN / PSI	- M. Makita, C. David, ...

SXR / LCLS	- B. Schlotter, J. Turner, …
DiProI / FERMI	- F. Capotondi, E. Principi, …
FLASH / DESY	- N. Stojanovic, K. Tiedtke, ...
	+ colleagues from the accelerator, laser, … groups

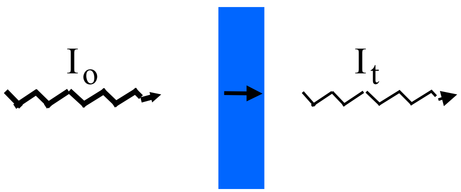
Resonant scattering for local probing of magnetization

IR (EUV/THz) ***pump*** – Resonant (magnetic) **X-ray** (small angle) scattering ***probe***



X-ray Magnetic Circular Dichroism in Absorption / Scattering

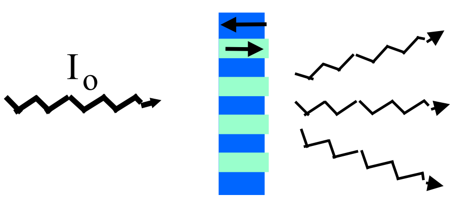
Absorption



Sample density ρ

$$I_t = I_o e^{-\sigma_a \rho t}$$

Small Angle Scattering

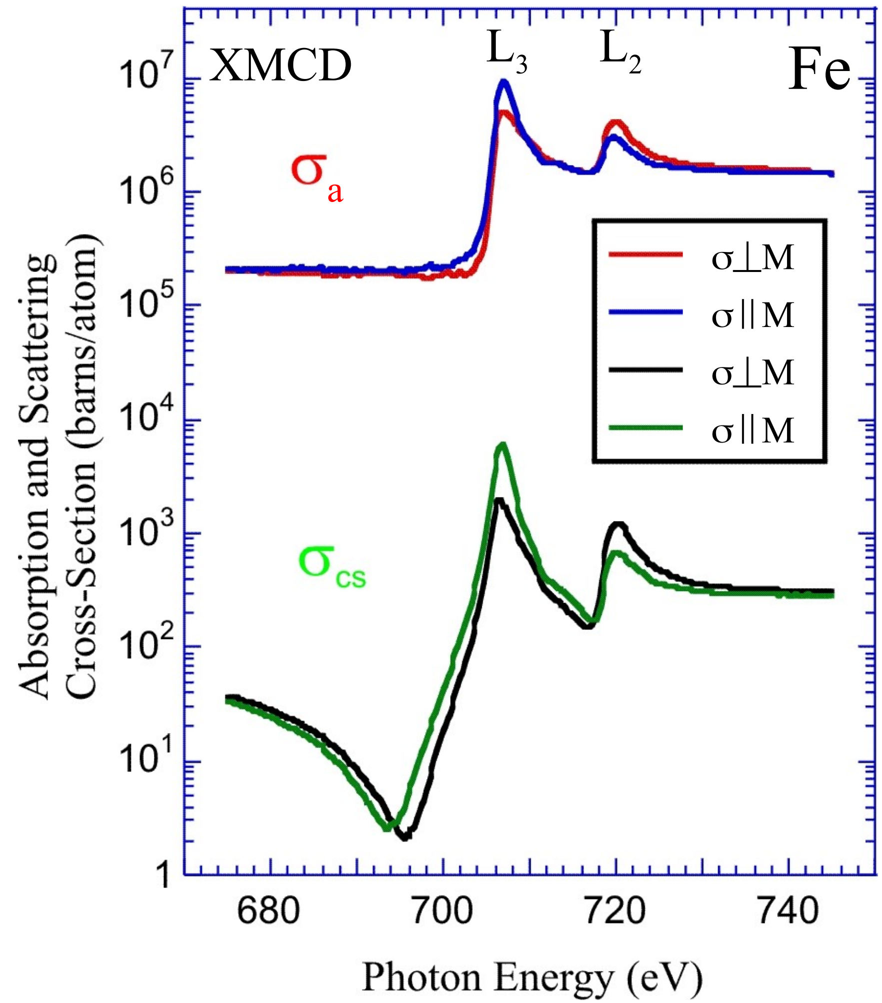


Sample density ρ

$$I_{cs} = I_o \sigma_{cs} / \sigma_a$$

$$\sigma_{cs} = c\,|f_1 + i f_2|$$

Data from Jeff Kortright (LBNL)



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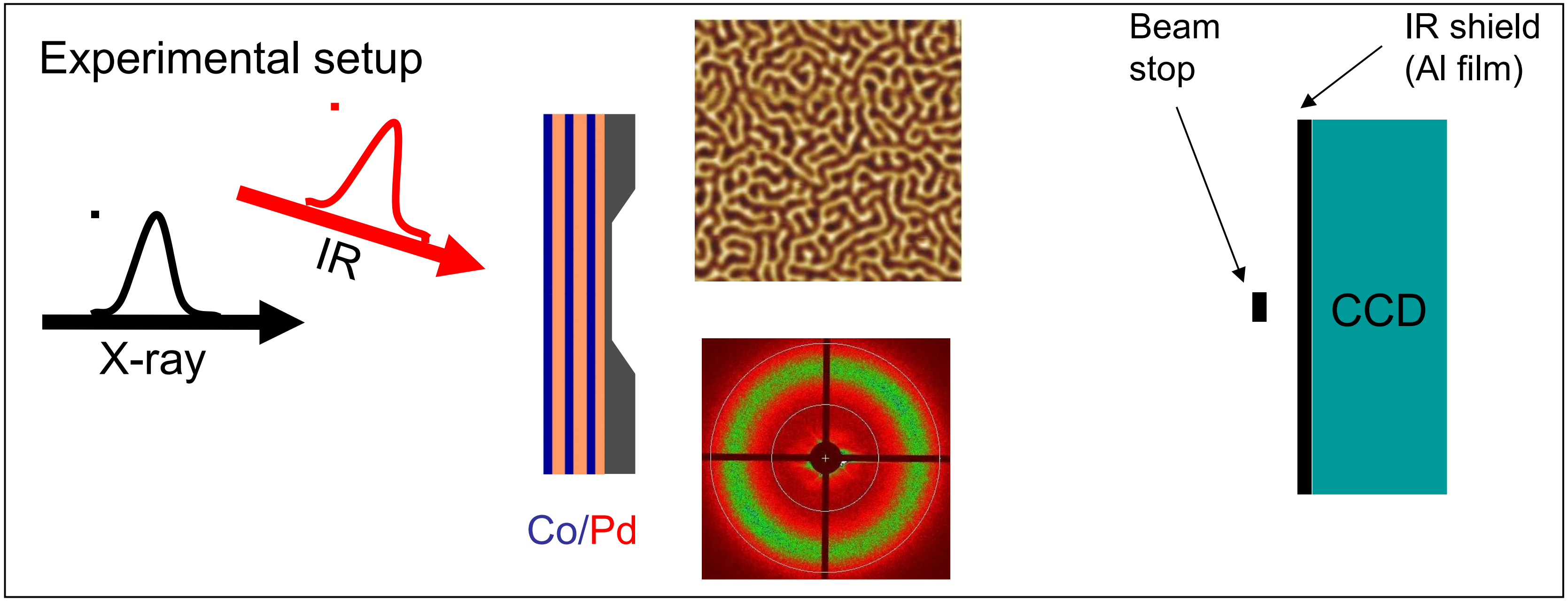
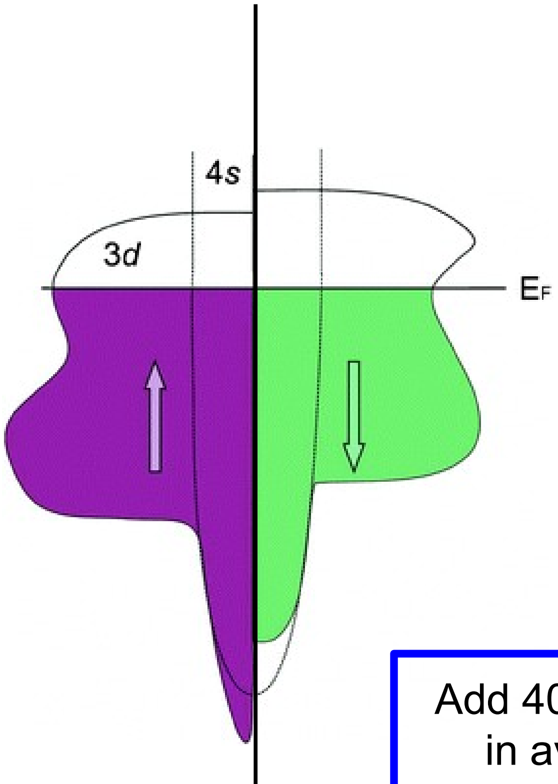
Magnetically dichroic absorption edges of transition metals:
- LCLS: **$L_{2,3}$** (700 – 850 eV)
- FLASH, FERMI (HHG): **$M_{2,3}$** (55 - 65 eV ↔ **37th – 41st** harmonic)



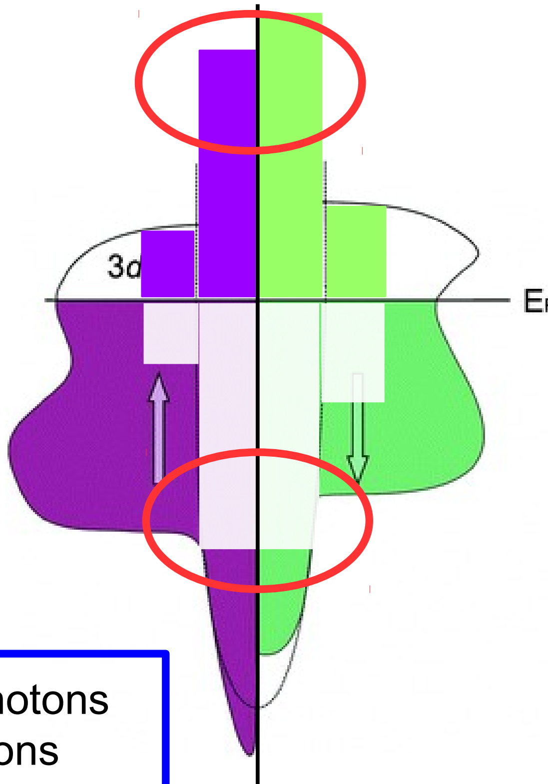
Integrated intensity → measure of the local magnetization

Is direct photon excitation and presence of hot electrons needed?

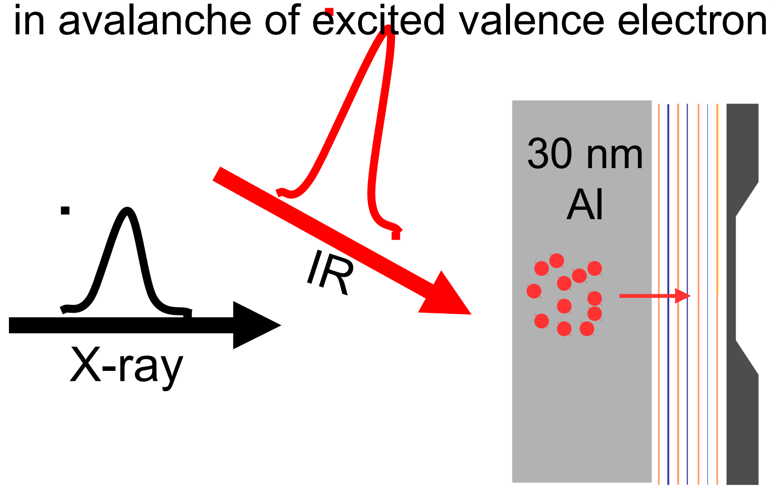


1.5 eV laser

excitation



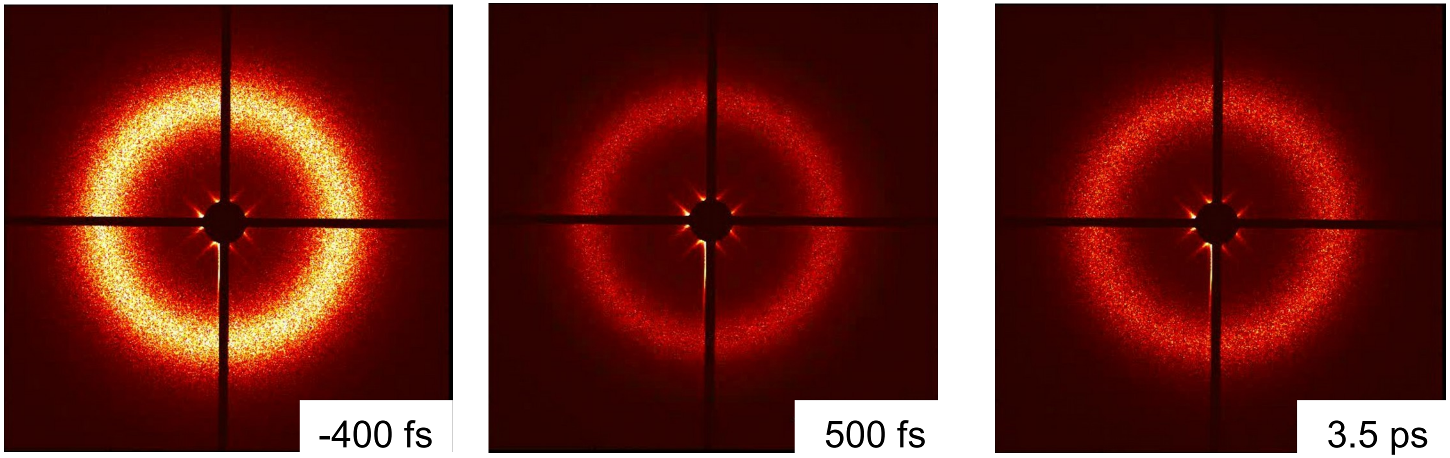
Add 40 nm Alu cap layer to convert IR photons in avalanche of excited valence electrons

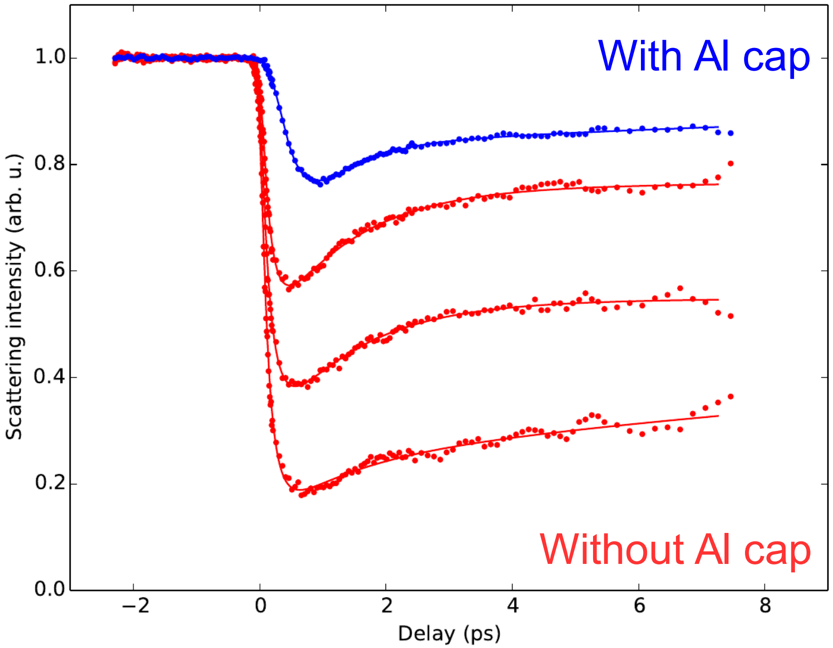


Hot electron excited ultrafast magnetization dynamics

SXR @ LCLS

B. Vodungbo et al. (submitted, 2015)





Stimulation of ultrafast demagnetization dynamics does not require direct interaction with photon pulse

Presence of very hot electrons not necessary for excitation of ultrafast demagnetization dynamics

See also from BESSY Slicing-Source:
A. Eschenlohr et al., Nat. Mater 12, 332 (2013)

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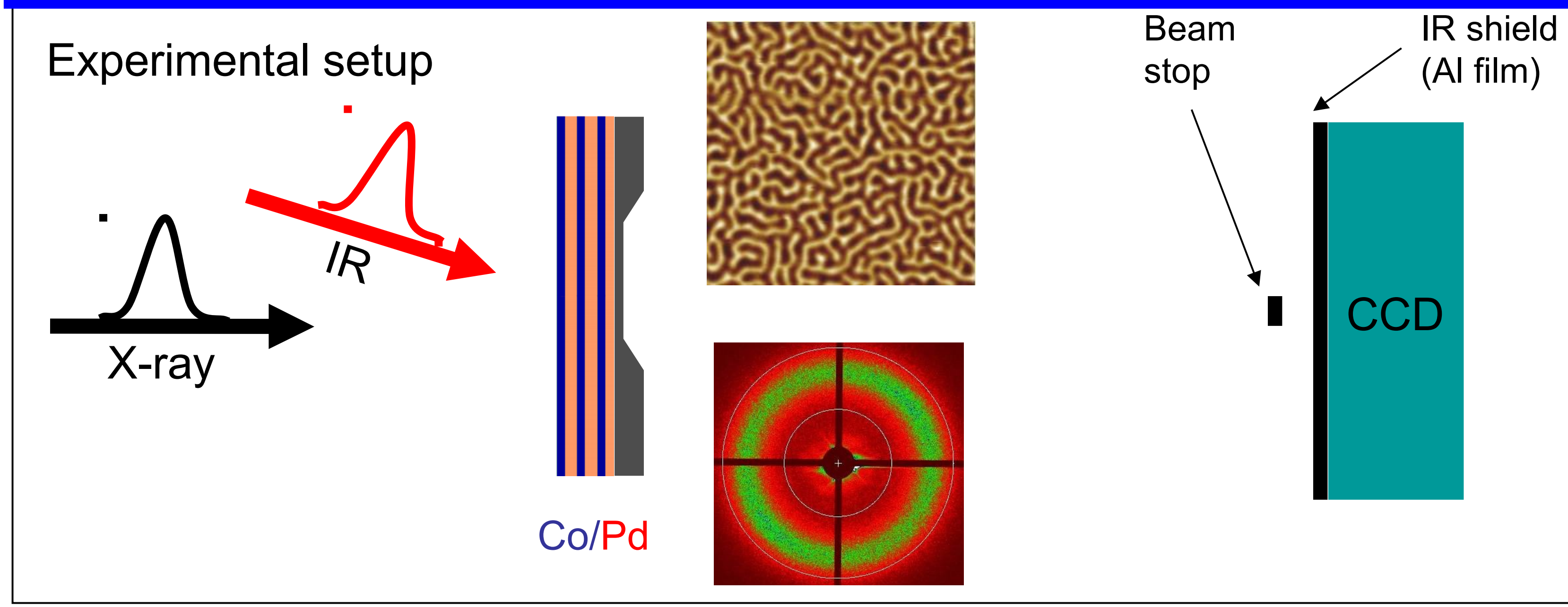
→ combines femtosecond temporal with nanometer spatial resolution



Integrated intensity → measure of the local magnetization

Form of scattering pattern → spatial information

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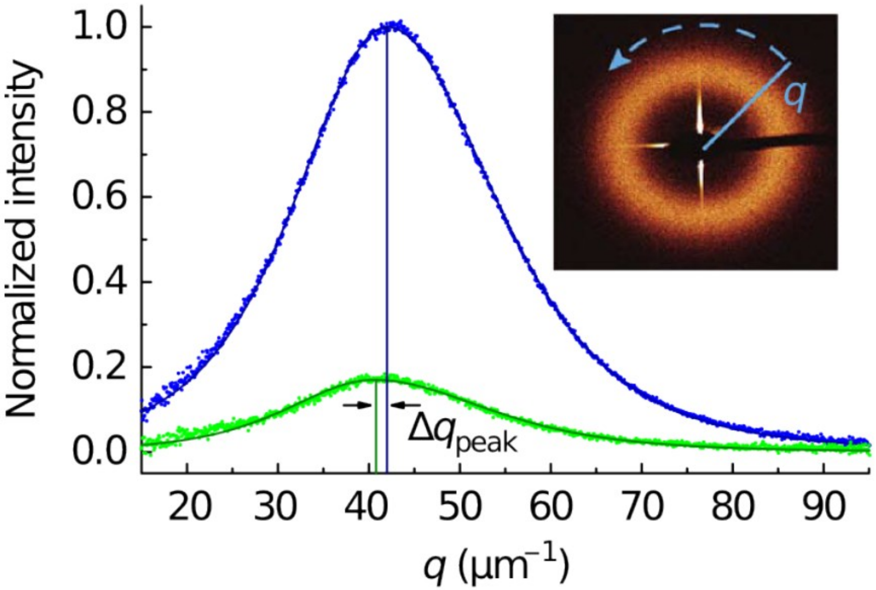
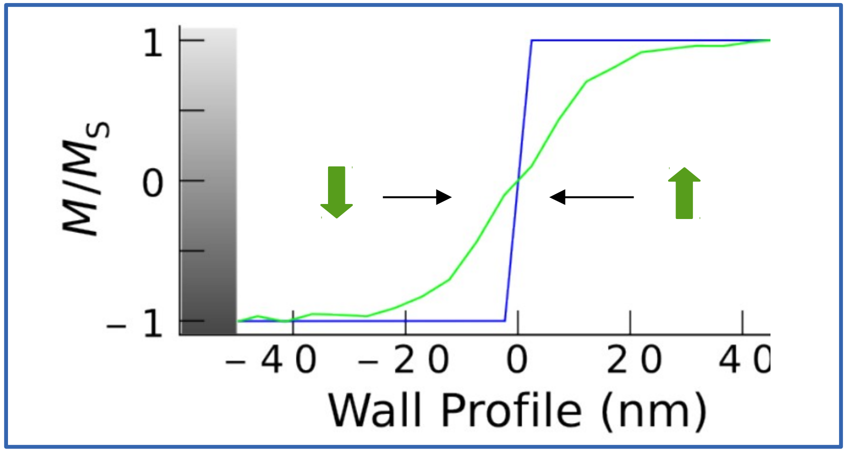
UF demagnetization in the high IR pump power limit

FLASH

B. Pfau et al., Nature Comm. 3, 1100 (2012)

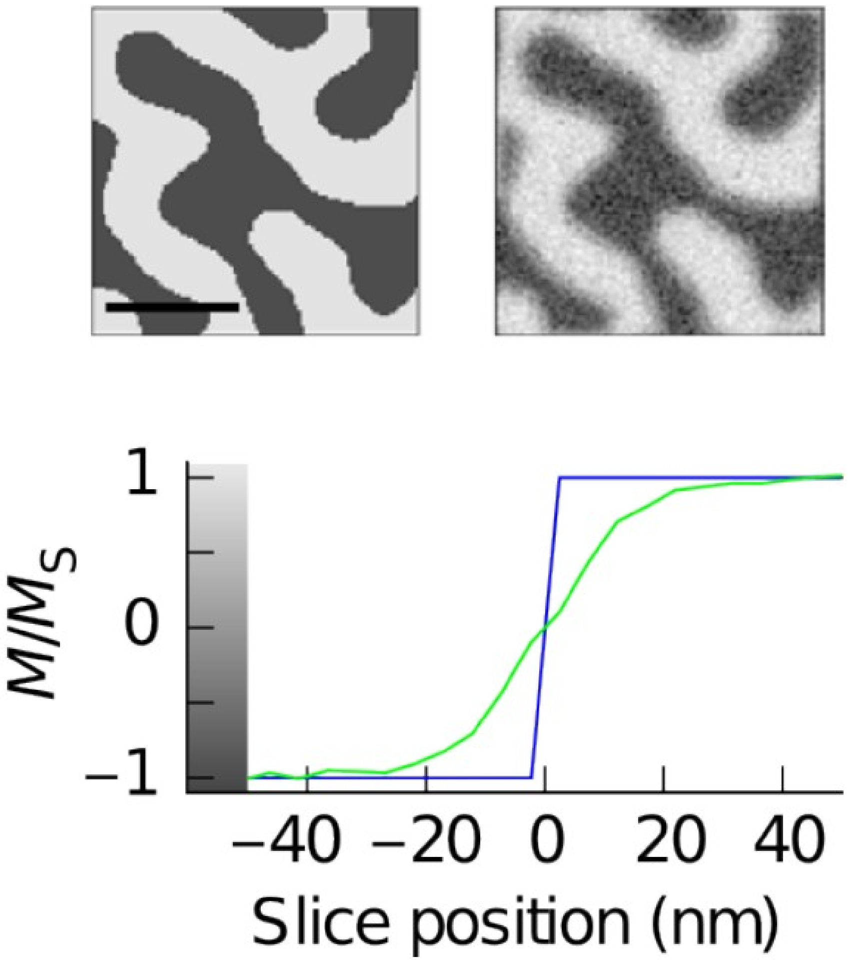




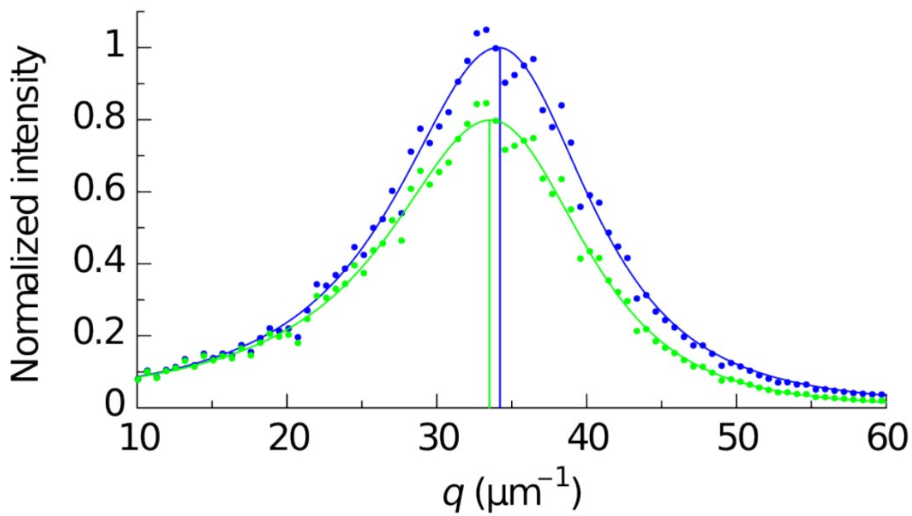
Angular momentum transport by hot polarized electrons

Interpretation based on model of superdiffusive spin transport proposed by
M. Battiato, K. Carva, P.M. Oppeneer, Phys. Rev. Lett. 105, 027203 (2010)

Monte Carlo simulation



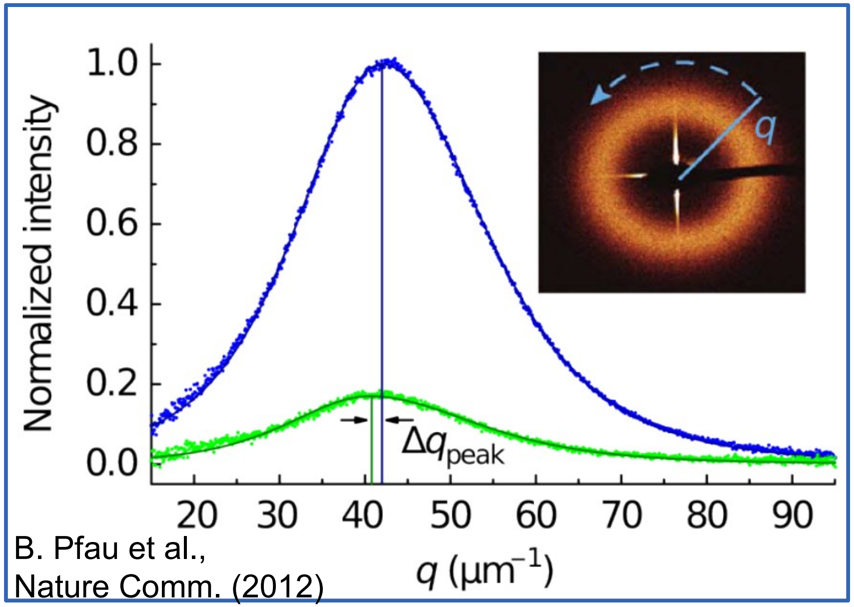
S(Q,t) as calculated from
Monte Carlo simulation

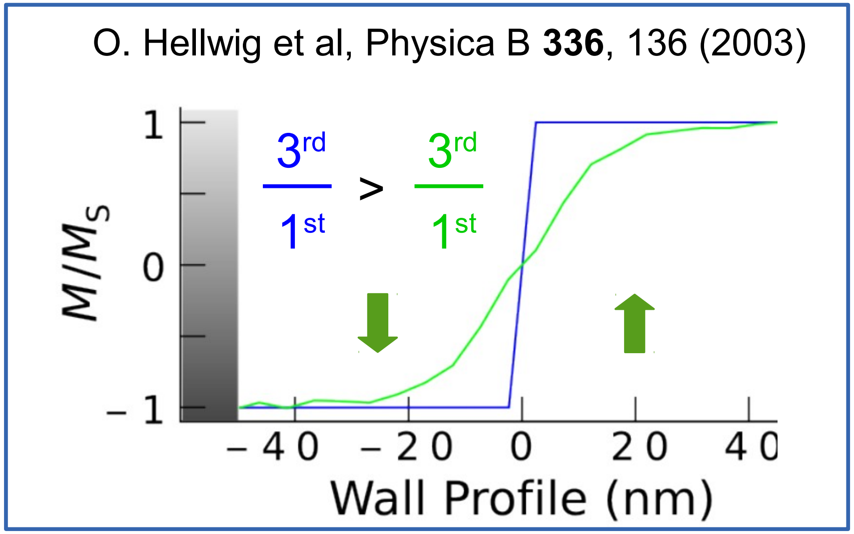


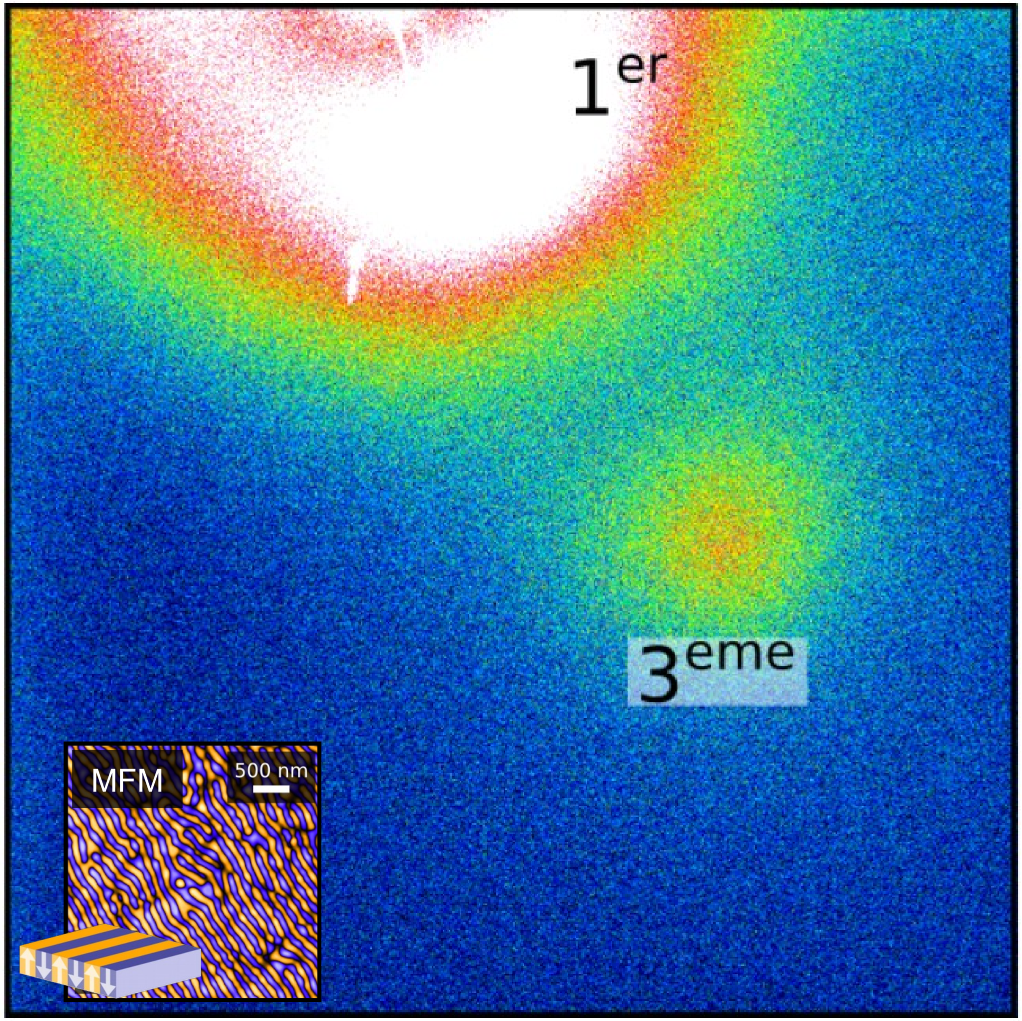
B. Pfau et al., Nat. Comm. 3, 1100 (2012)

Probing domain wall evolution directly

Higher scattering orders → more detailed insight in magnetic domain structure







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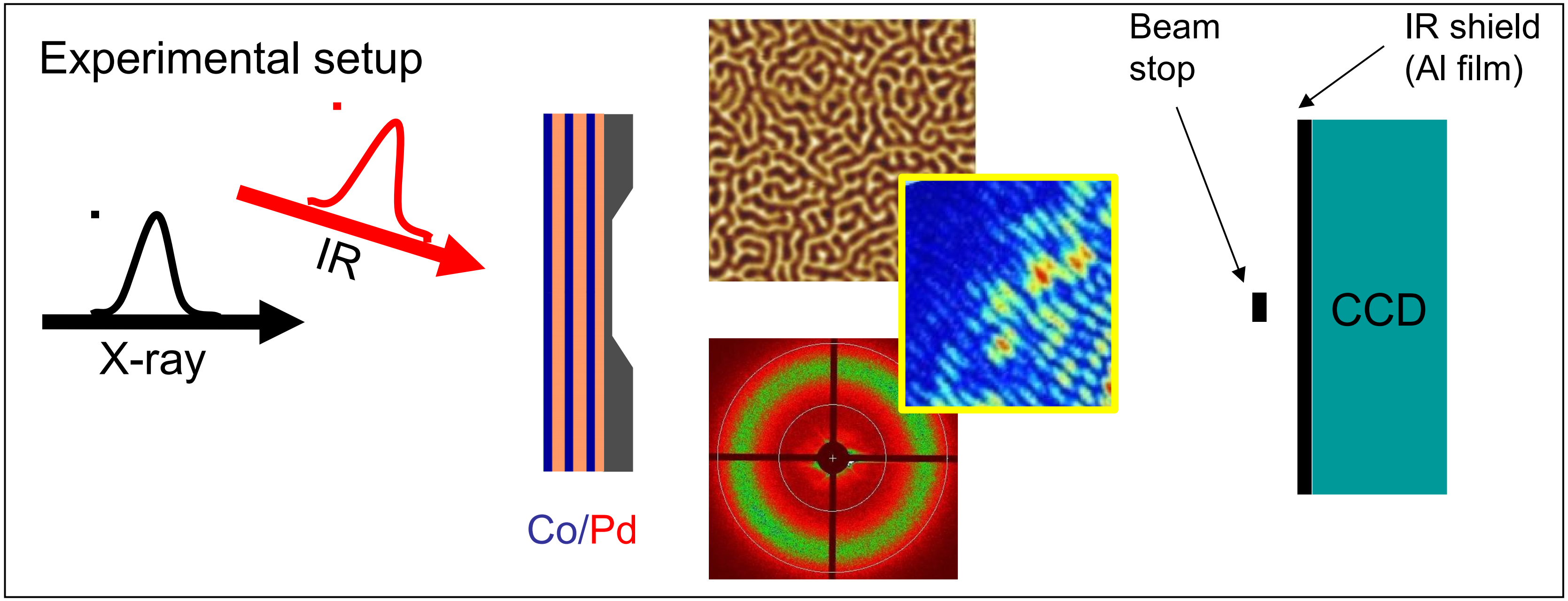
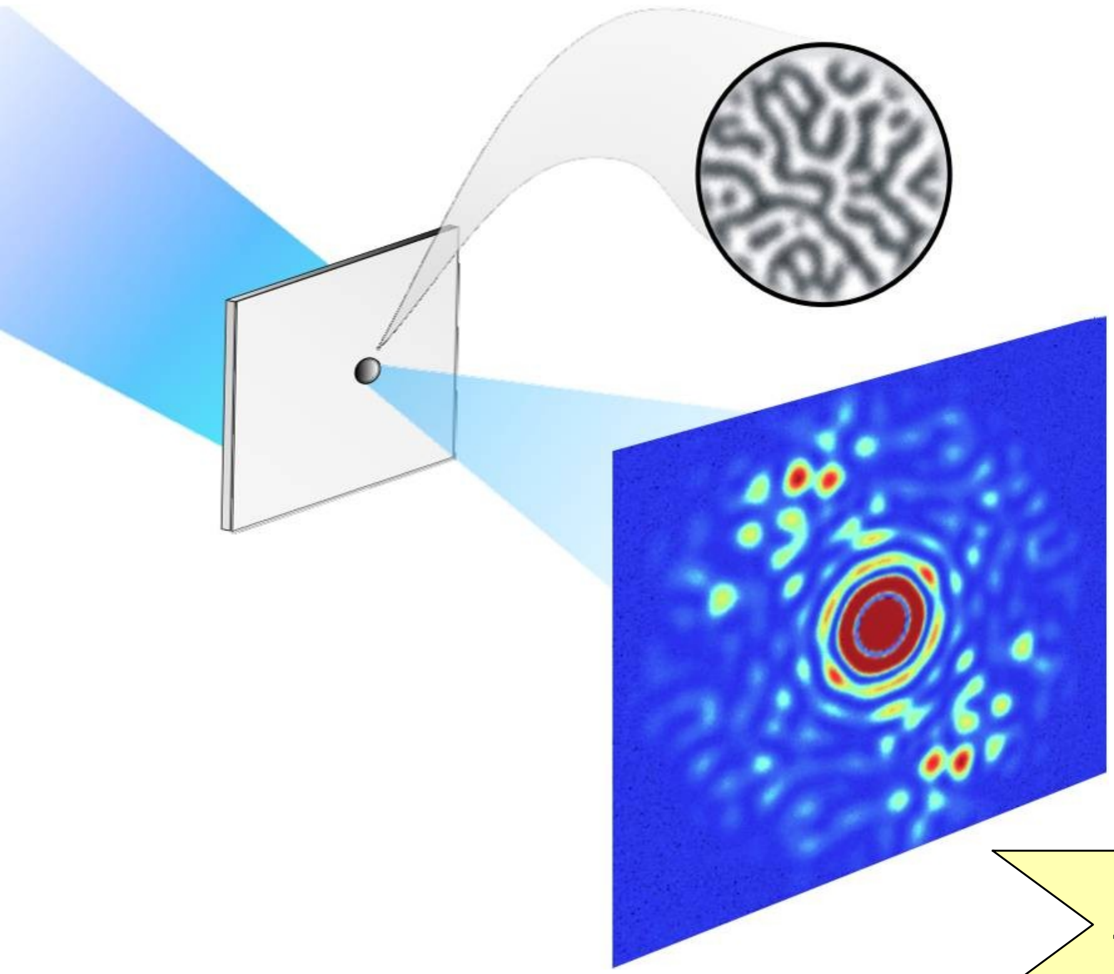


Integrated intensity → measure of the local magnetization

Form of scattering pattern → spatial information

Speckle → imaging

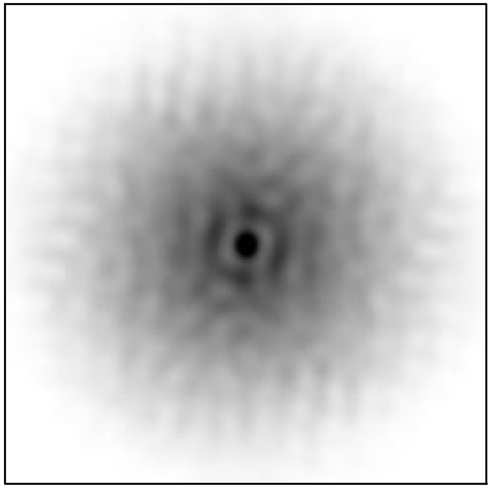
Phase problem in X-ray scattering



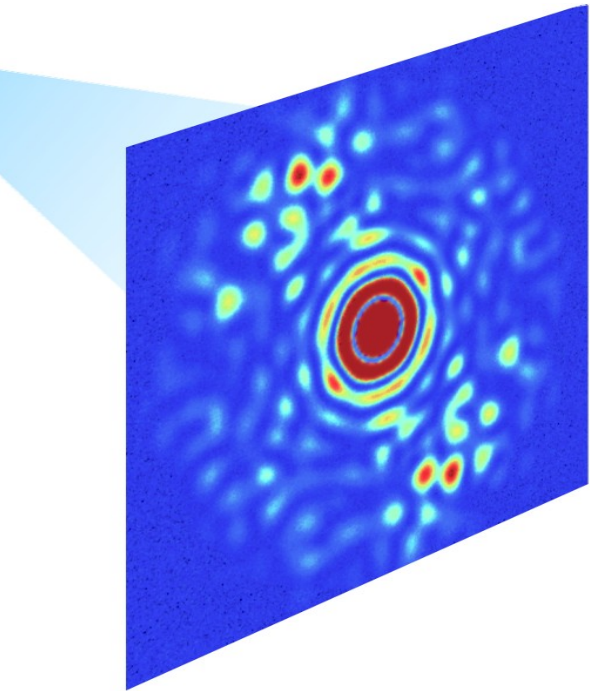
Scattering amplitude is complex, but measurement detects the intensity only:

$$I_{p,q} = \left| M_{p,q} \cdot e^{-i\phi_{p,q}} \right|^2$$

Fourier Transform



Solving the phase problem



Phase problem in X-ray scattering:

Wave on detector is complex, but only intensity is measured, phase information is lost

Two solutions:

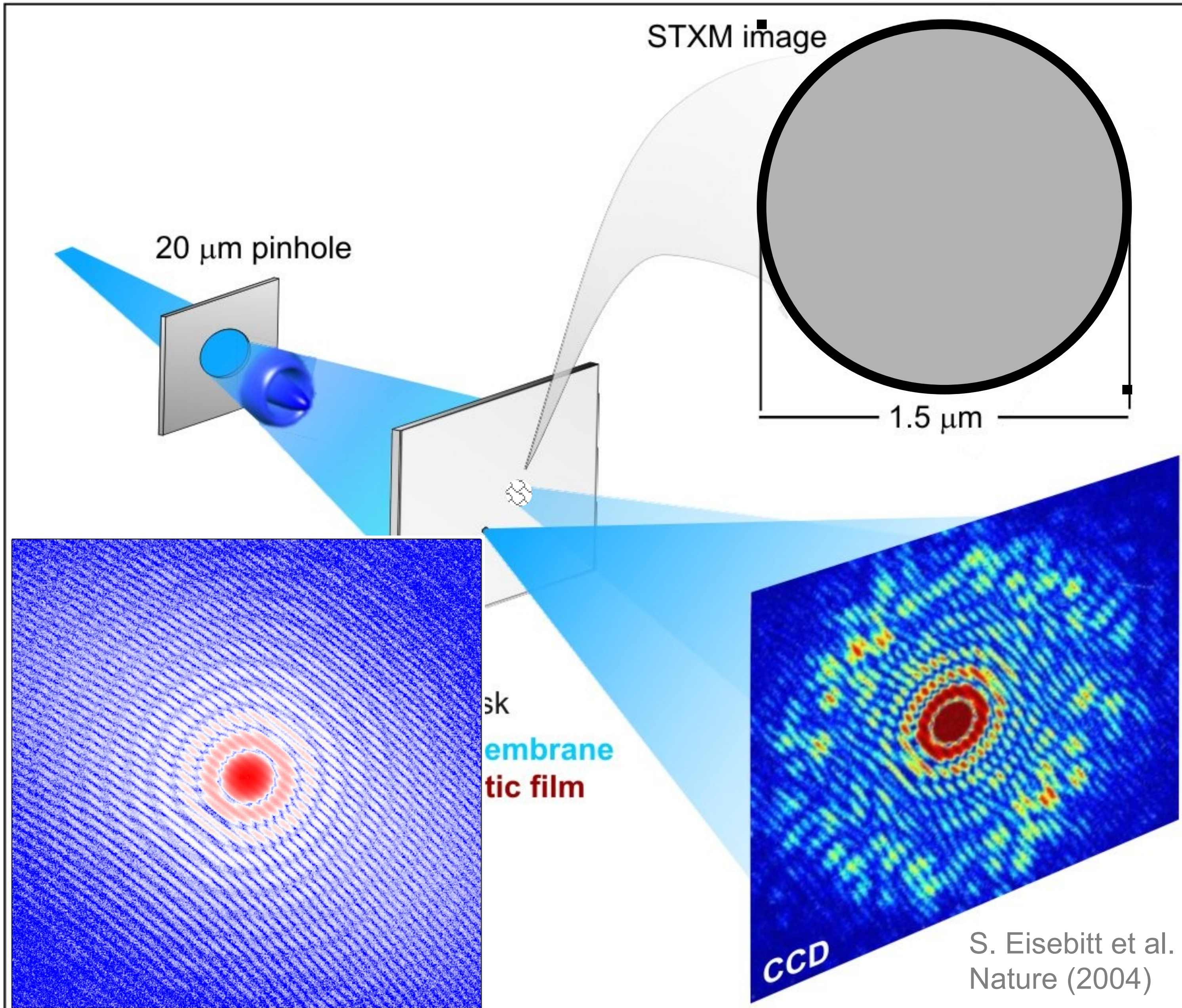
1) X-ray Holography (Gabor 1948, Stroke 1965)

- Phase information is encoded in detectable intensity fluctuations
- True imaging technique

2) Iterative Phase Retrieval (Sayers 1952)

- Surround sample with ‘known’ support
- Measure additional scattering intensities (‘oversampling’)
- Use iterative algorithm to retrieve scattering phases from additional scattering intensities

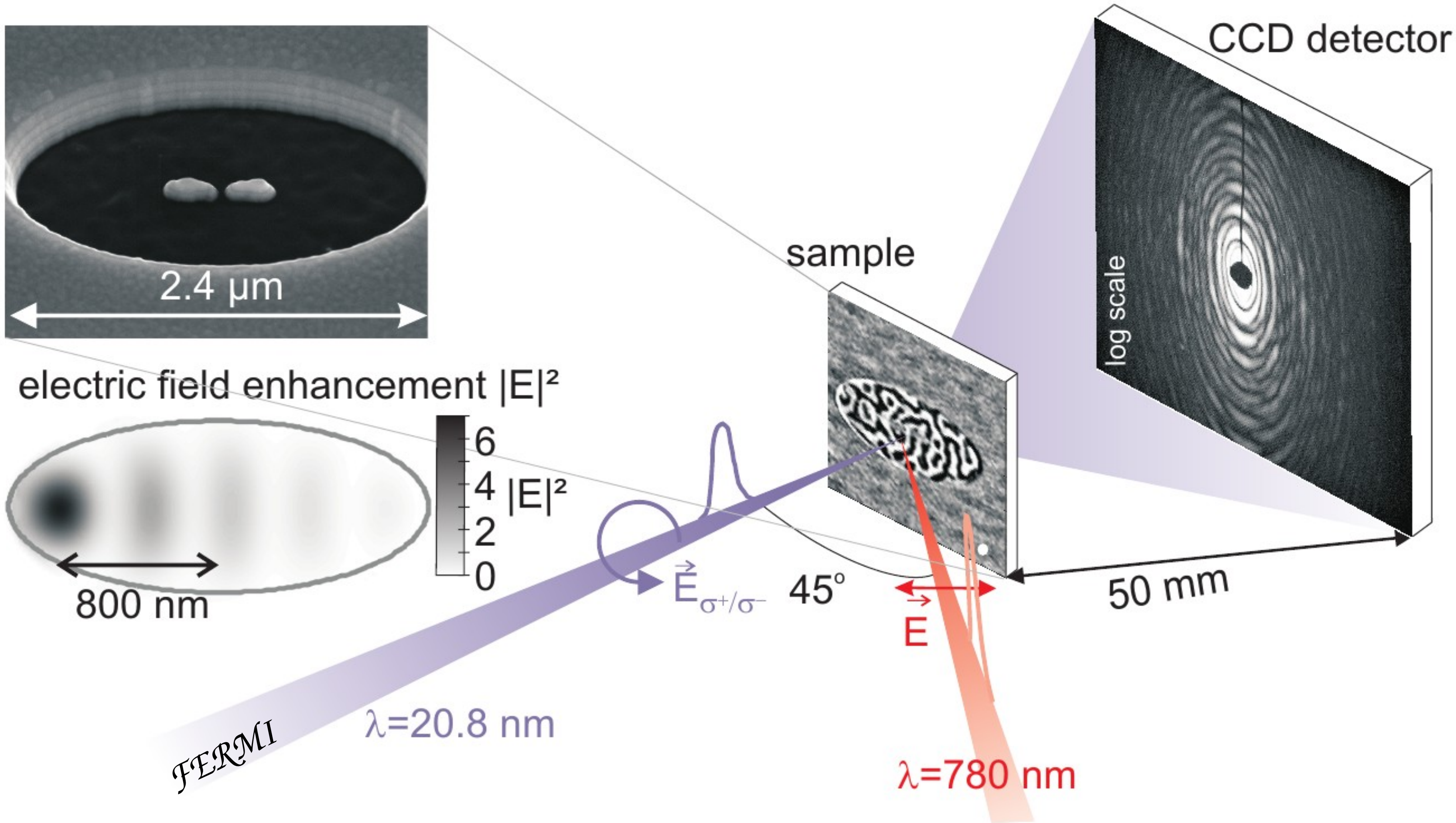
Fourier Transform Holography (FTH)



S. Eisebitt et al.
Nature (2004)

Imaging ultrafast demagnetization dynamics

after a spatially localized optical excitation

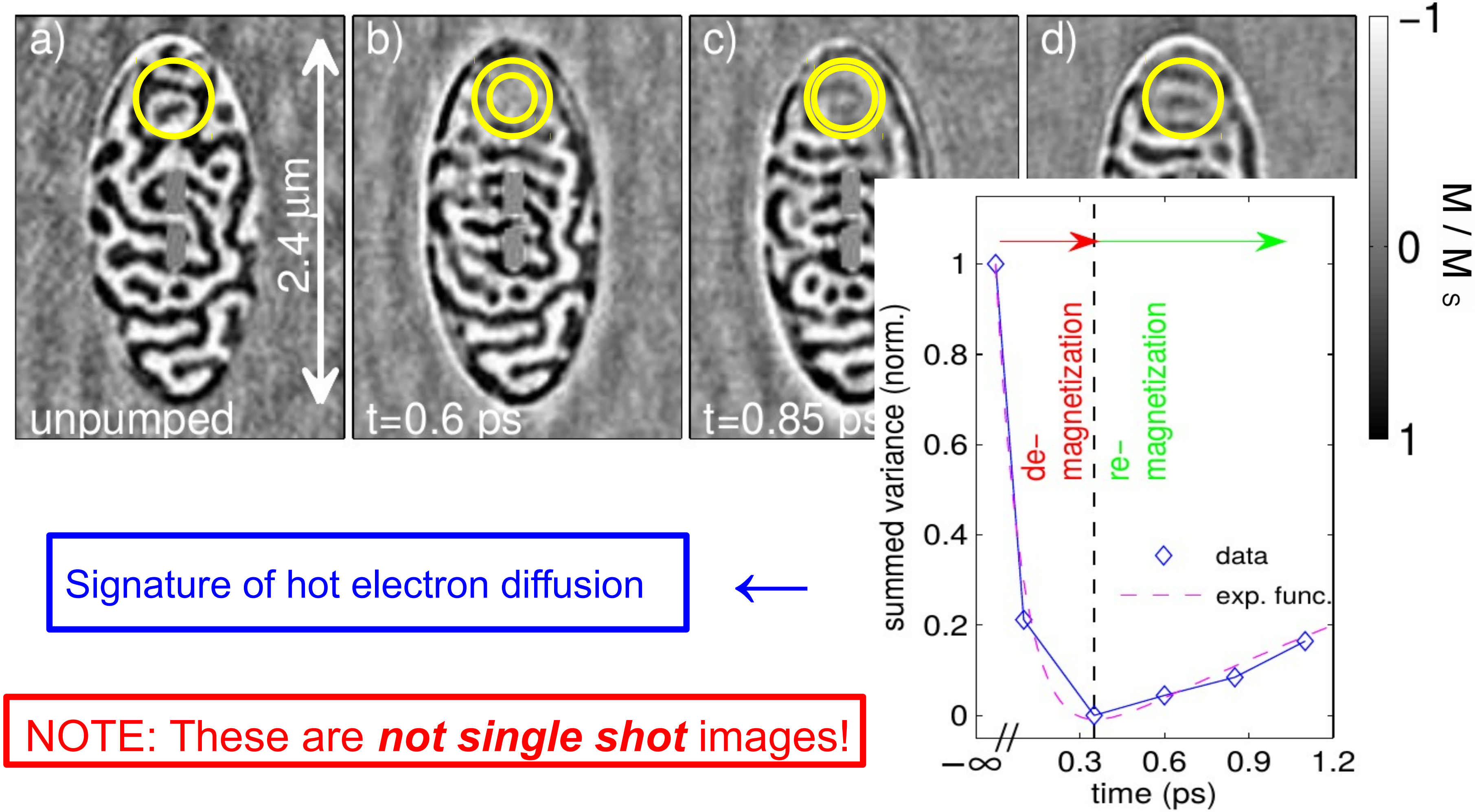


C. von Korff Schmising et al., Phys. Rev. Lett. **112**, 217203 (2014)

Imaging ultrafast demagnetization dynamics

DiProI @ FERMI

after a spatially localized optical excitation

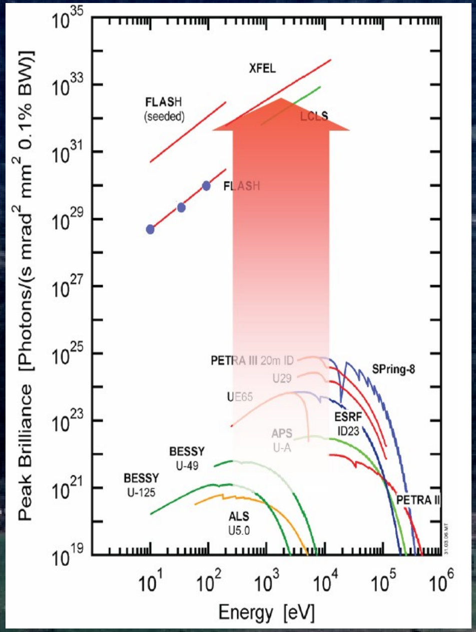


Signature of hot electron diffusion ←

NOTE: These are ***not single shot*** images!

C. von Korff Schmising et al., Phys. Rev. Lett. **112**, 217203 (2014)

The Linac Coherent Light Source at SLAC



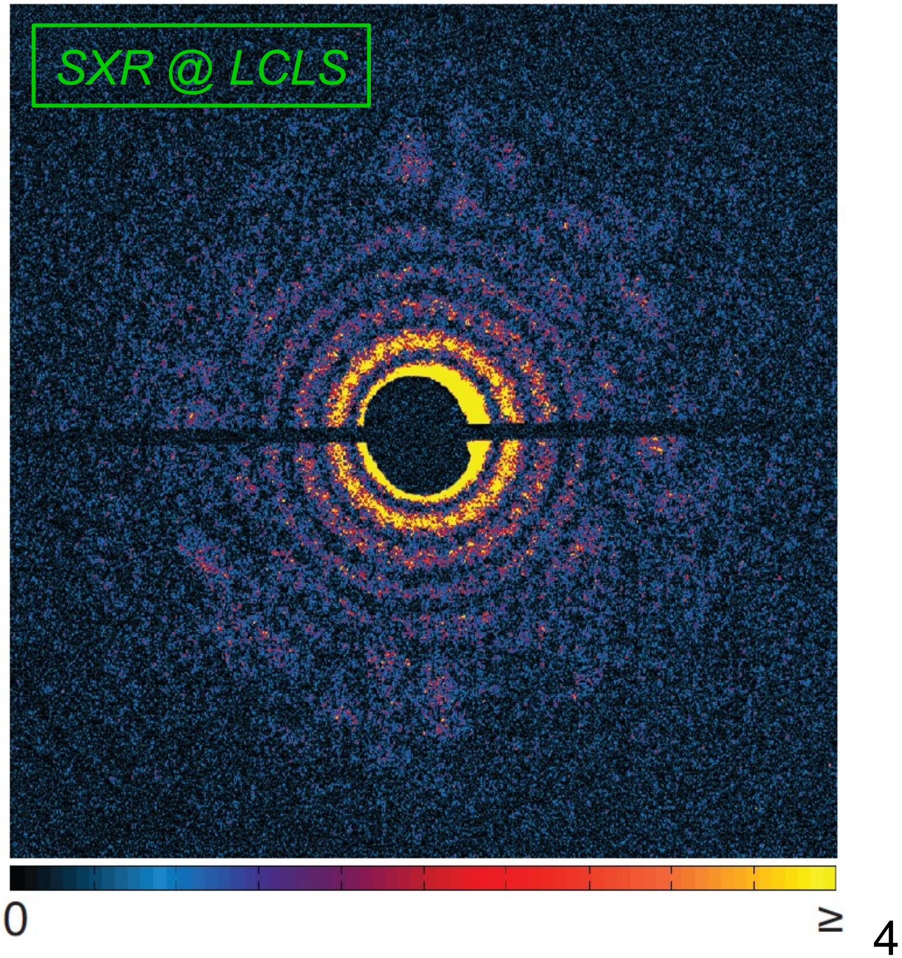
- 10^{13} photons/pulse
- fsec pulse duration
- 100% transverse coherence

Opportunity for
X-ray snapshot studies

- Imaging of non-reproducible 'states'
  - equilibrium fluctuations
  - non – reproducible relaxation dynamics
- Overcomes low rep. rates of extreme conditions
  e.g., pulsed magnetic fields, high pressure

Single x-ray pulse based snapshot imaging

T. Wang et al., PRL **108**, 267403 (2012)



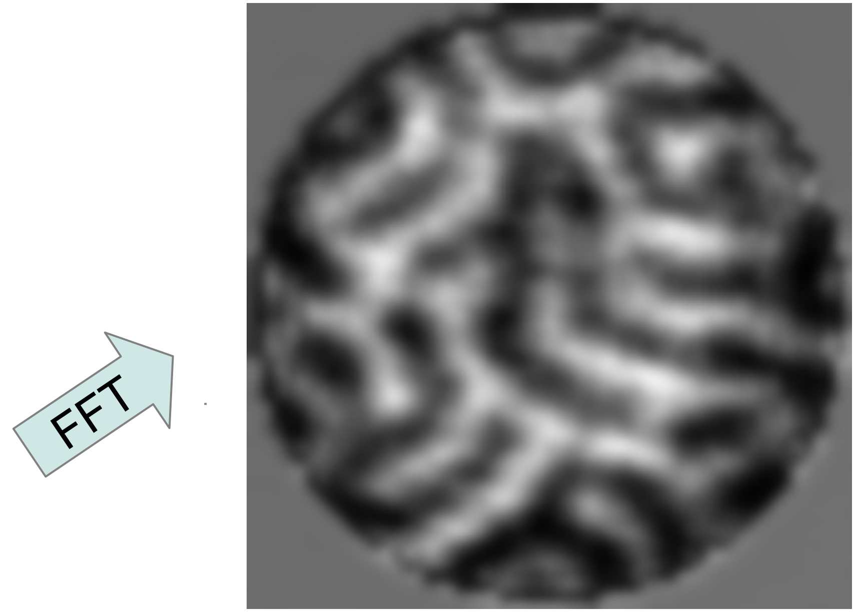


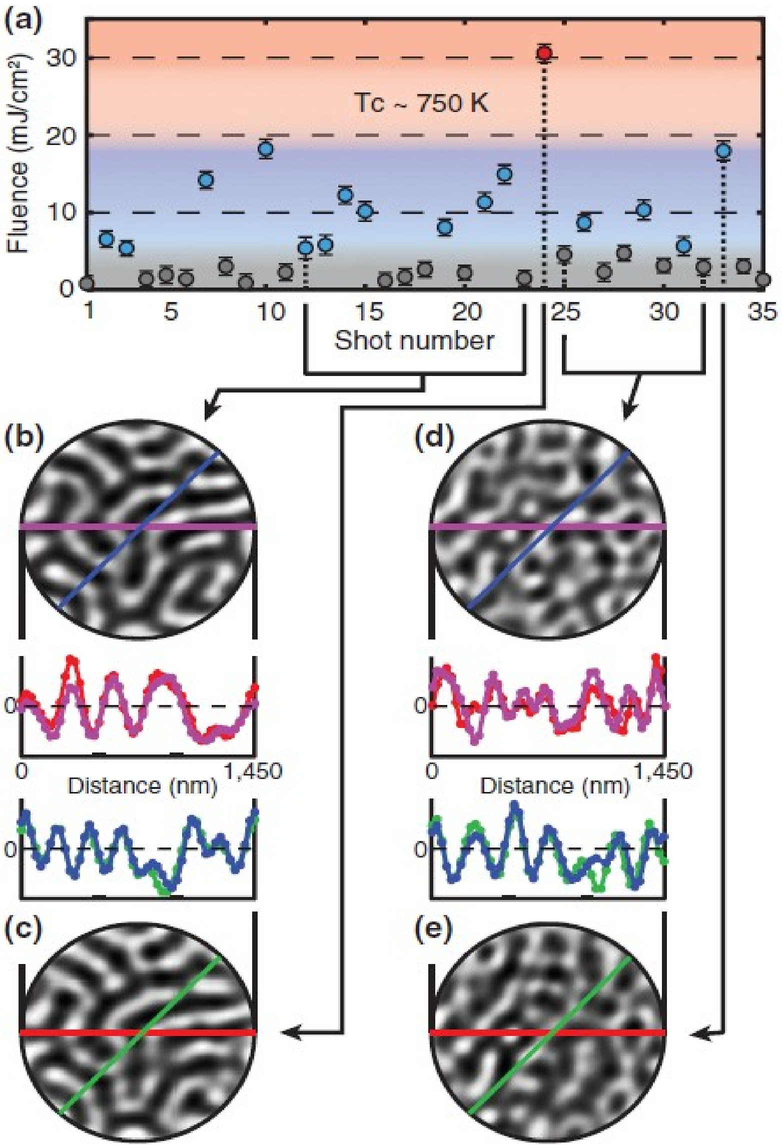
Image of magnetic domain structure obtained from a single X-ray pulse

~ 50 nm spatial resolution
~ < 80 fs temporal resolution
[meanwhile ~ 20 fs shown]

X-ray induced *"modifications"*

T. Wang et al., PRL **108**, 267403 (2012)

- Single shot images can be recorded non-destructively.
- Magnetic domain structure changes *after/due to* intense x-ray pulse.
- Magnetization seems to fade, may indicate inter-diffusion at interfaces of magnetic multilayer.



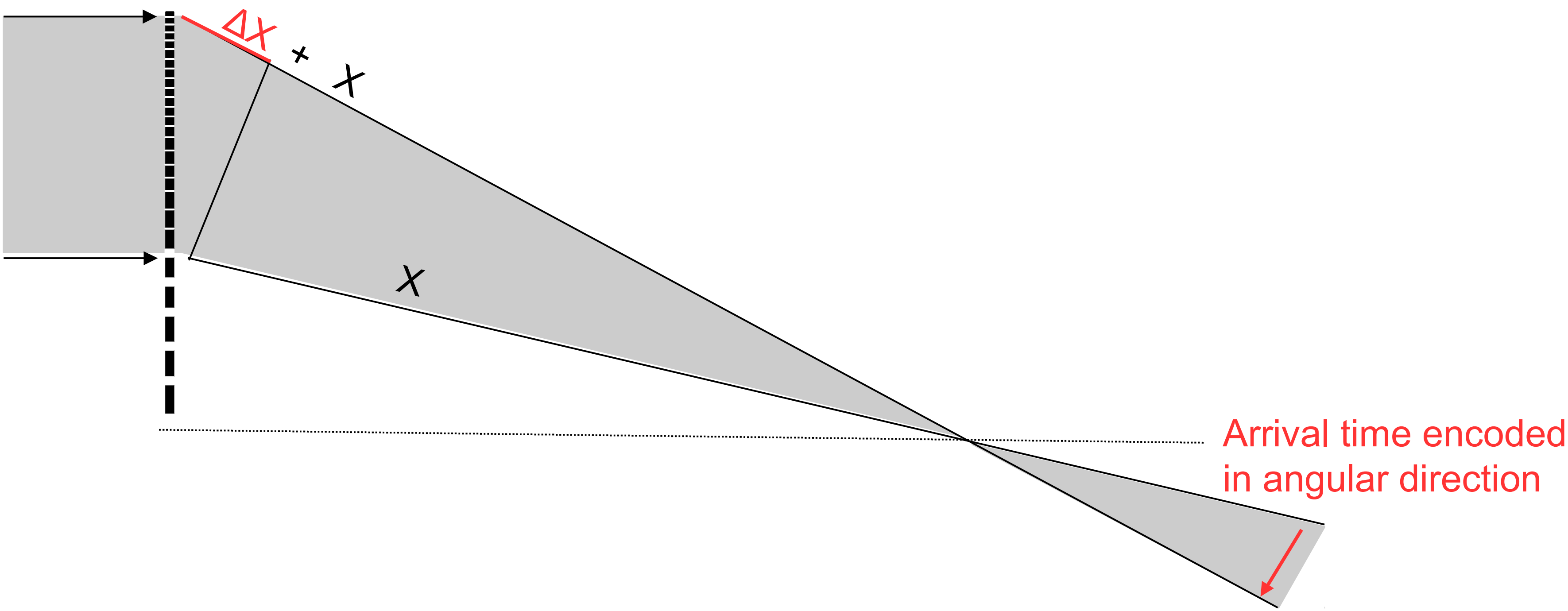
NOTE: This is a single shot image, but ***for one instance*** only!

Snapshot recording of ultrafast dynamics

Michele Buzzi,[a] Mikako Makita,[a] Armin Kleibert,[a] Boris Vodungbo,[b,c] Nicolas Jaouen,[d] Ludovic Howald,[a] Philippe Zeitoun,[c] Jörg Raabe,[a] Kai Tiedtke,[e] Harald Redlin,[e] Christian David,[a] Frithjof Nolting[a], Jan Lüning[b,c,d]

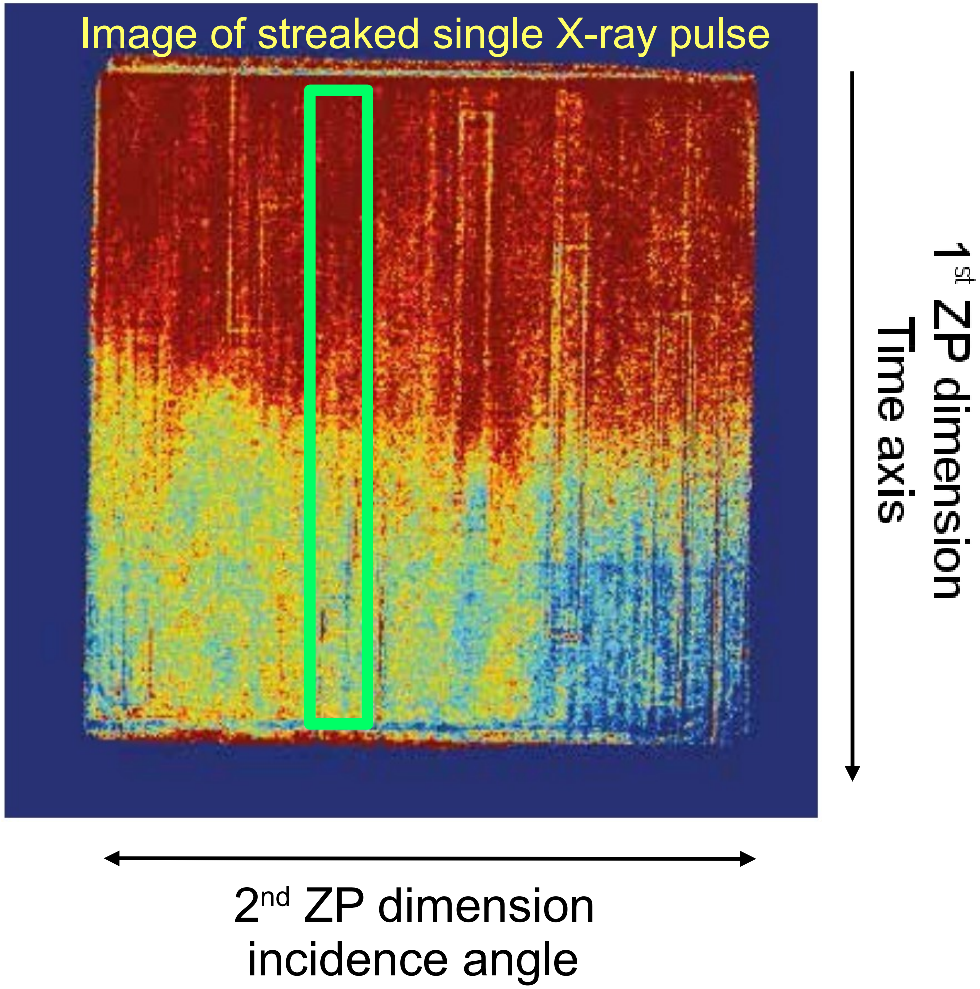
a) Paul Scherrer Institut, Villigen, Switzerland, b) Sorbonne Universités, France
c) ENSTA ParisTech, France, d) Synchrotron SOLEIL, France
e) Deutsches Elektronen-Synchrotron, Germany

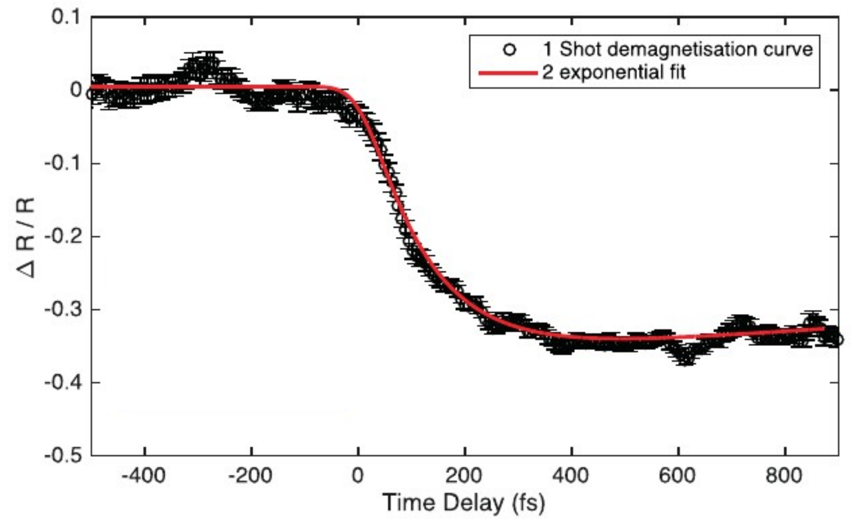
Basic idea:

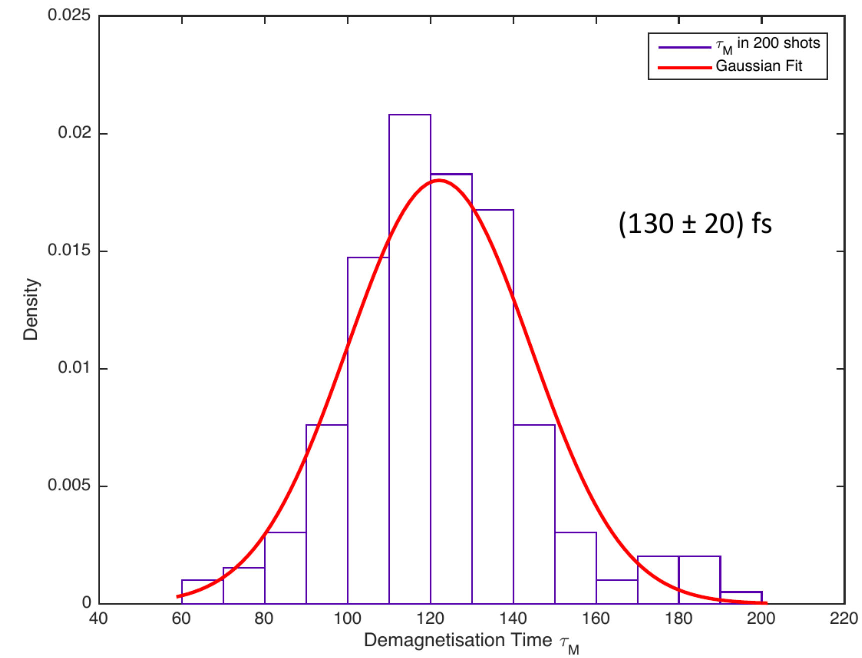


Snapshot streaking of ultrafast demagnetization dynamics

BL3 @ FLASH







Snapshot imaging/microscopy

Hit and Destroy

and measure before the object changes

- Proposed for structural analysis of individual (bio-)nanoparticles
  → *time scale ~ atomic motion*

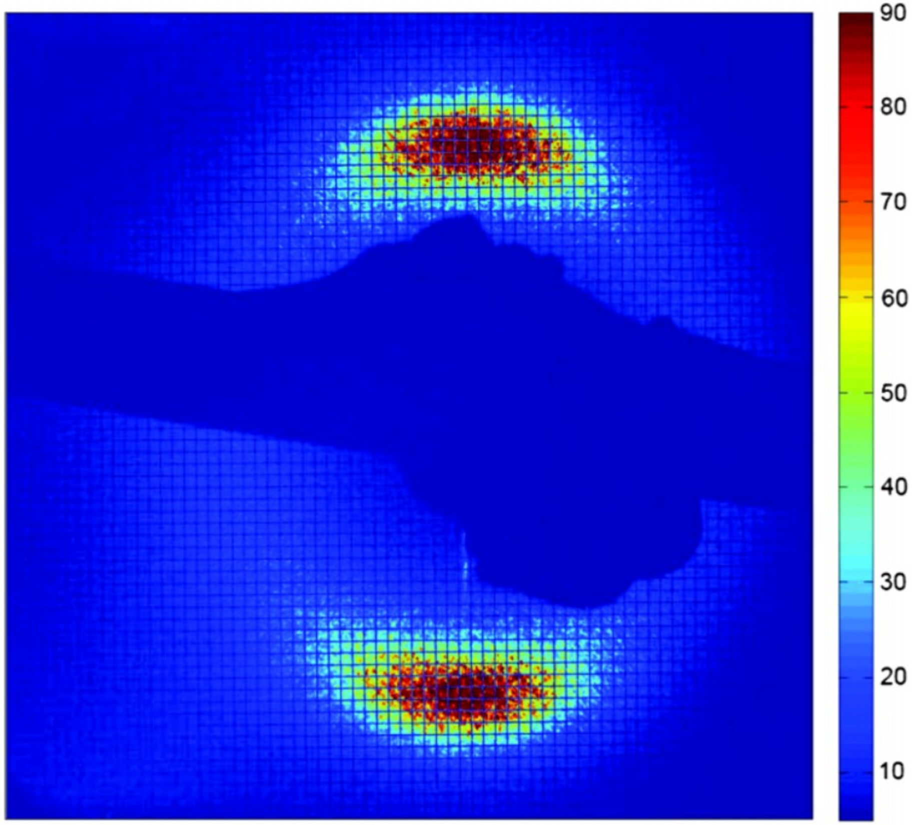
- Imaging of electronic structure ordering (charge, orbital, spin)
  → *time scale ~ electronic motion*

Single X-ray pulse snapshot imaging of magnetic domain structure
T. Wang et al., Physical Review Letters 108 (2012).

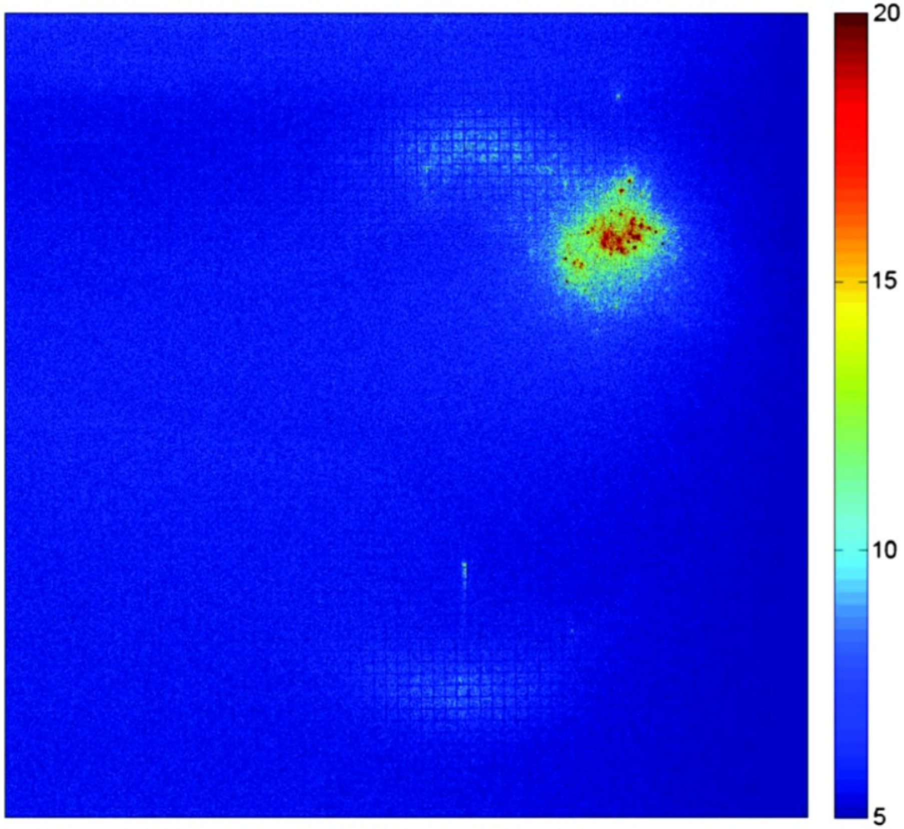
Breakdown of the Resonant Magnetic Scattering Signal

Magnetic scattering pattern recorded with

1000 'weak' X-ray pulses (5 mJ /cm^2)



1 very strong X-ray pulses (5 J /cm^2)



FLASH X-ray pulse: 100 fs, 60 eV (Co M_3)

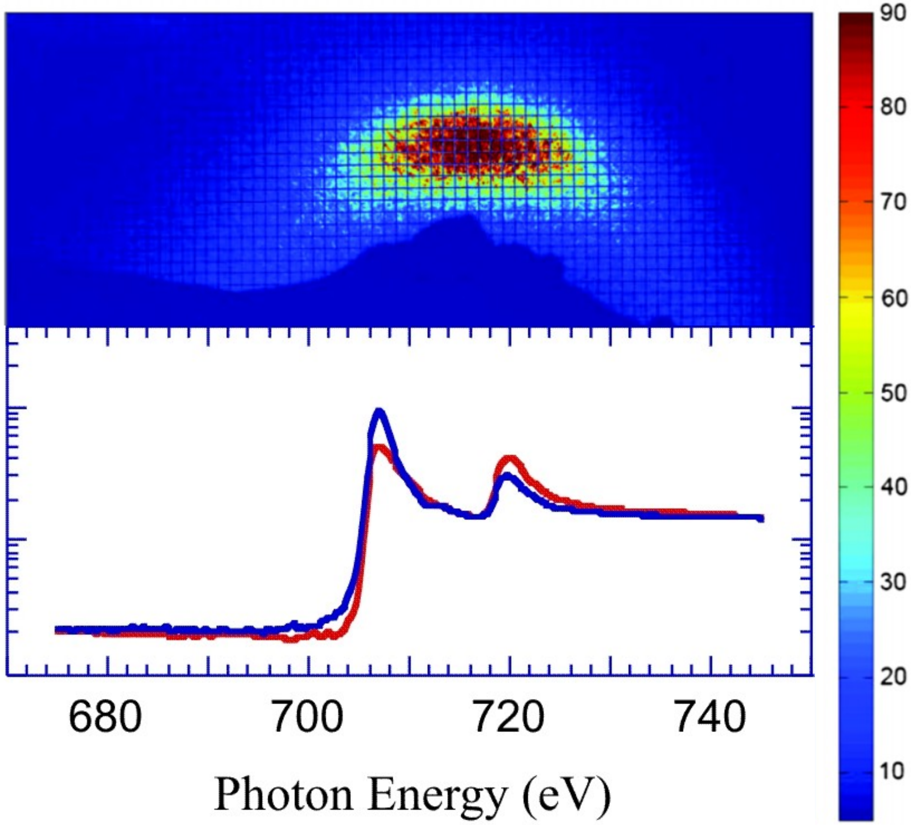
Sample: (Co/Pt) multilayer on SiN membrane

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

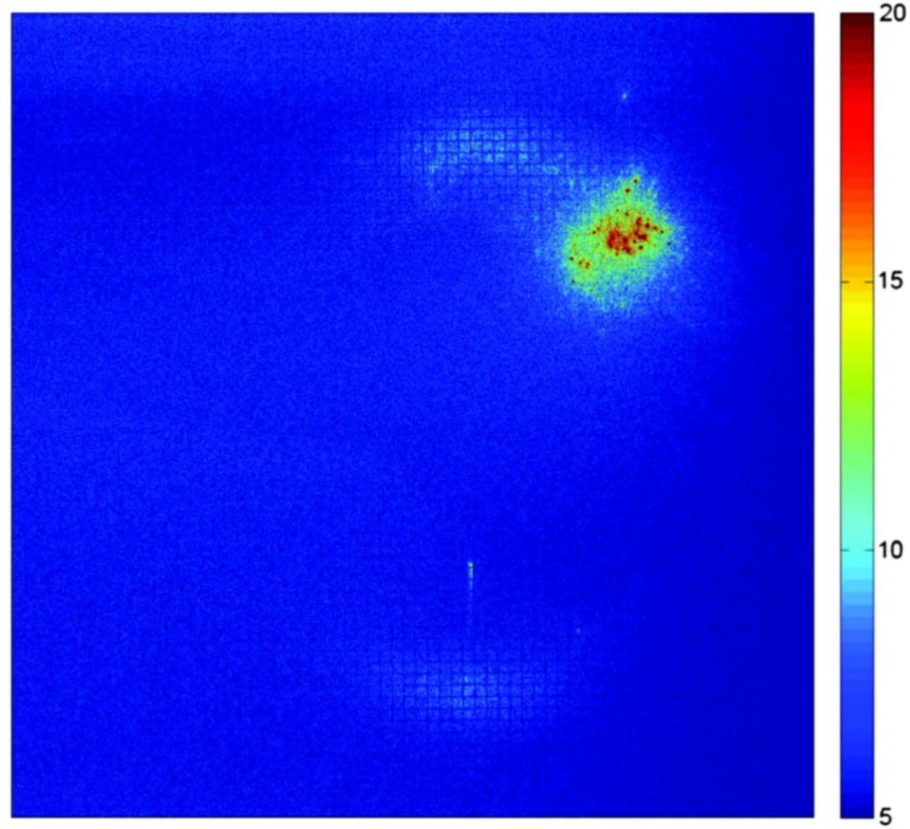
Breakdown of the Resonant Magnetic Scattering Signal

Magnetic scattering pattern recorded with

1000 'weak' X-ray pulses (5 mJ /cm²)



1 very strong X-ray pulses (5 J /cm²)



At least two possible mechanisms:

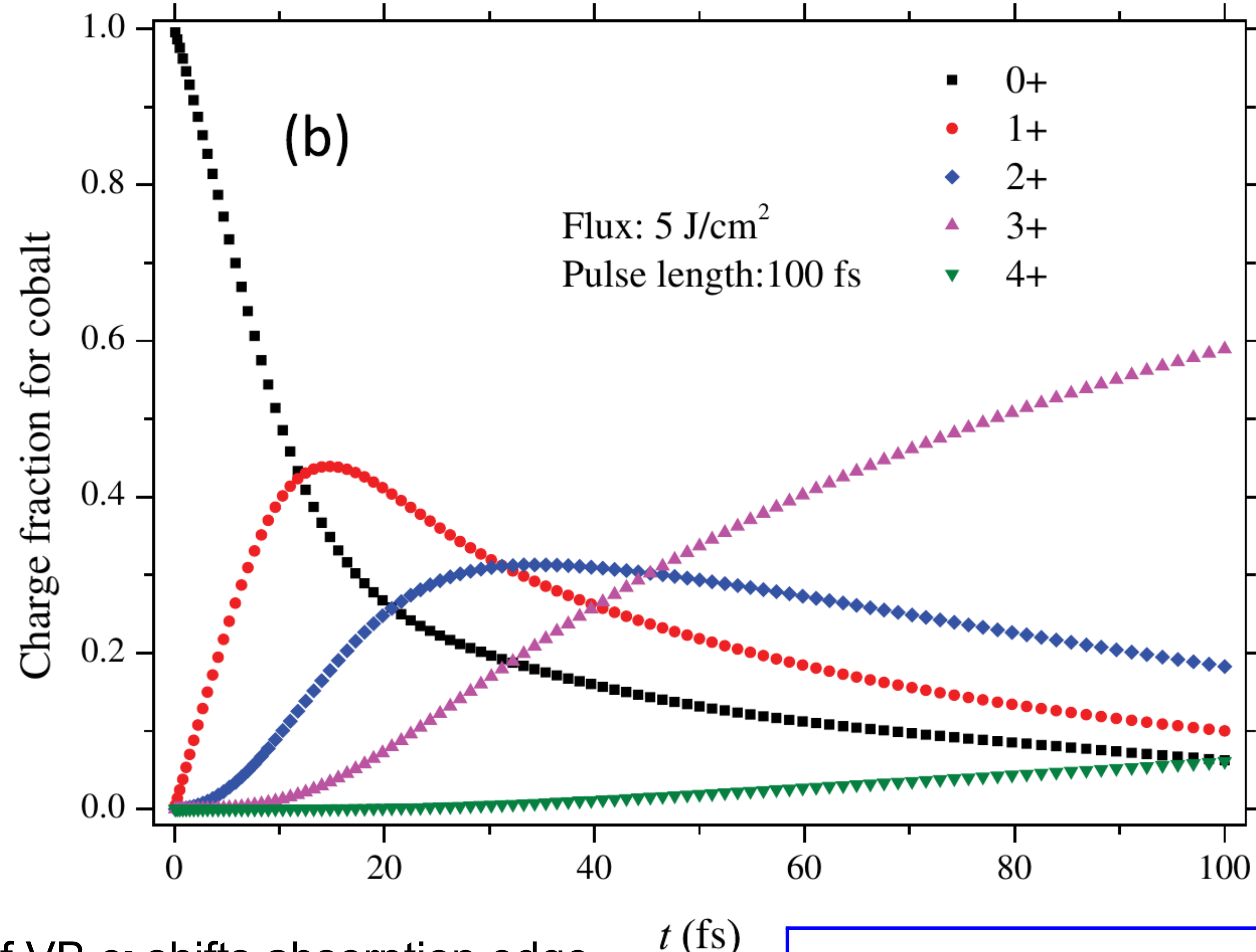
1) ionization of VB e^- → shift in binding energy → turning photon energy off-resonance

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

Ultrafast evolution of the atomic 'oxidation' state

R. Santra (CFEL)

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



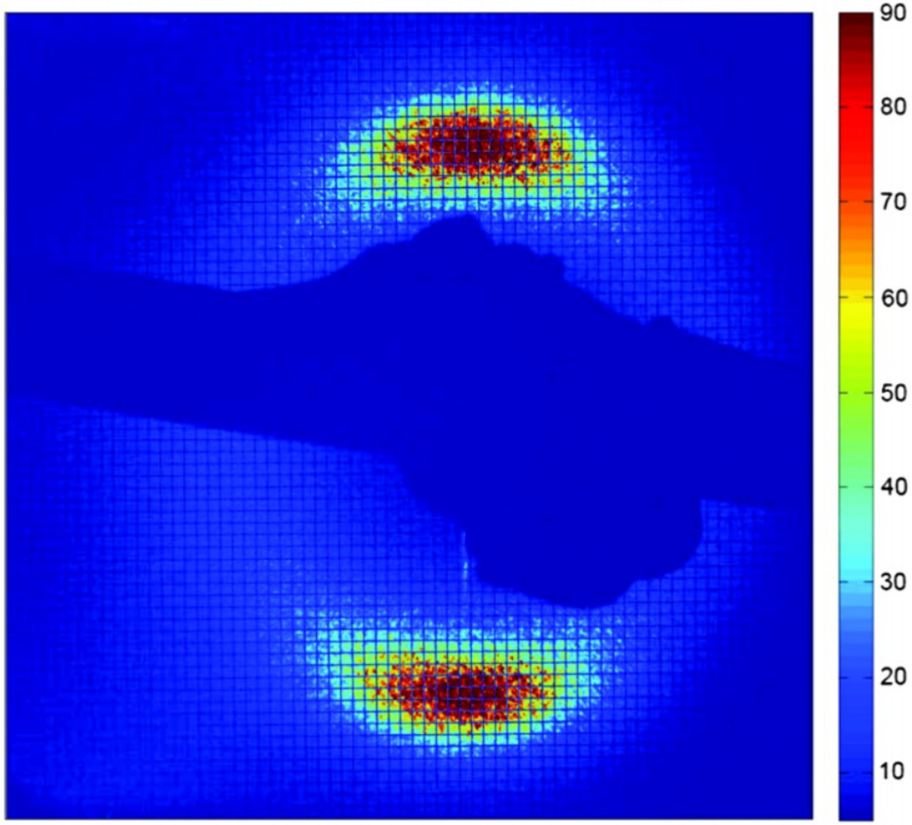
- excitation of VB e^- shifts absorption edge
- photon energy is turned 'off-resonance'
- magnetic scattering is suppressed

In addition, other processes like stimulated scattering may further suppress magnetic scattering!

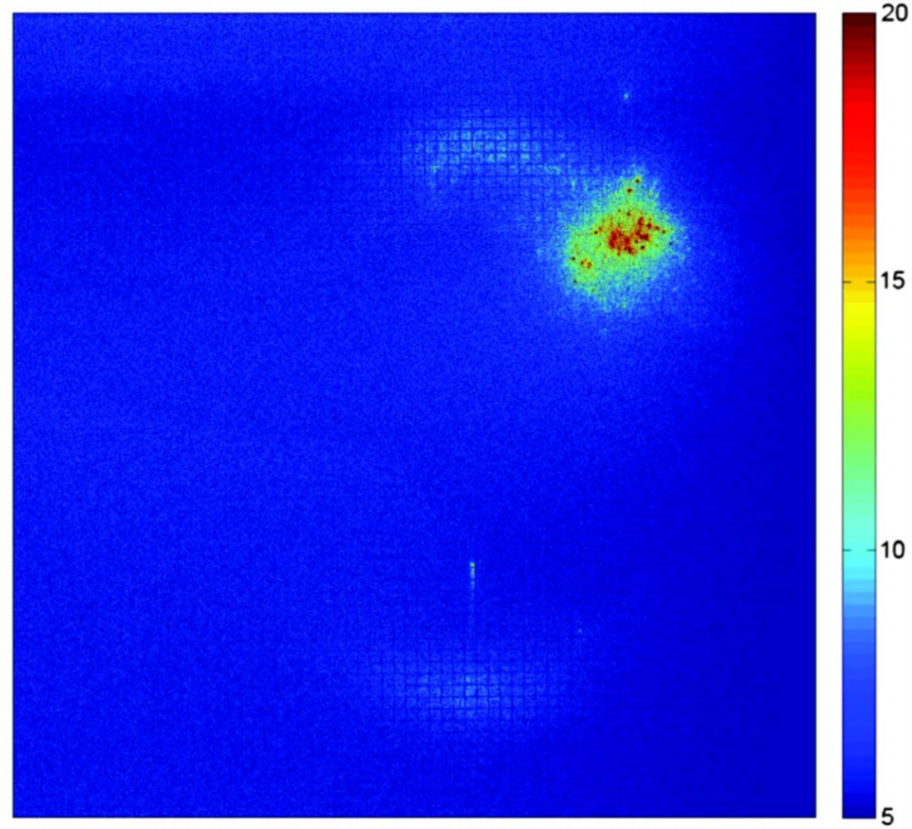
Breakdown of the Resonant Magnetic Scattering Signal

Magnetic scattering pattern recorded with

1000 'weak' X-ray pulses (5 mJ /cm²)



1 very strong X-ray pulses (5 J /cm²)



At least two possible mechanisms:

1) ionization of VB e^- → shift in binding energy → turning photon energy off-resonance
see: L. Müller et al., Phys. Rev. Lett. 110, 234801 (2013)

2) stimulation of competing processes → reduction in yield of resonant magn. scat.
see: J. Stöhr and A. Scherz, Phys. Rev. Lett. 115, 107402 (2015)

Summary

Coherent diffraction techniques ideally suited for XFEL experiments:

- combine nanometer spatial with femtosecond temporal resolution
- probe dynamics of individual components in complex materials
- obtain single shot based view on ultrafast dynamics

Non-local effects contribute to laser driven magnetization dynamics

Next goals :

→ achieve sub 10 fs time resolution

→ obtain ***truly*** single shot based view on ultrafast dynamics

→ multi X-ray color pulses