XFEL investigation of light pulse induced magnetization dynamics

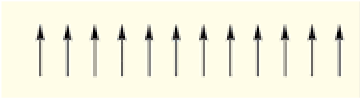
Jan Lüning

Laboratoire de Chimie Physique – Matière et Rayonnement
Sorbonne Université, Paris

XFEL investigation of light pulse induced magnetization dynamics

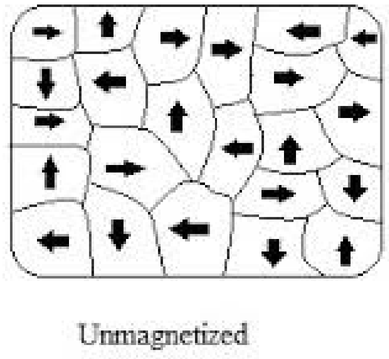
Outline

- Magnetic data recording
  An amazing story of need driven technology development
- Laser driven magnetization dynamics
  The discovery of sub-picosecond magnetization dynamics
- Resonant magnetic small angle X-ray scattering
  - evidence for relevance of transport phenomena
- Towards more decisive X-ray based experiments

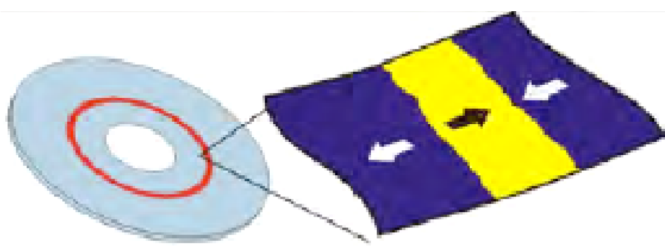


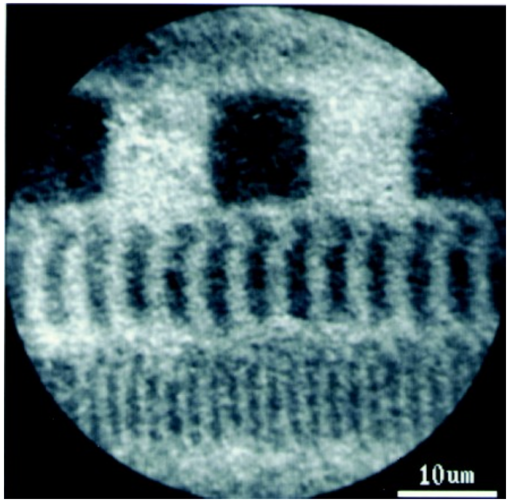
Ferromagnetic

Magnetic domains



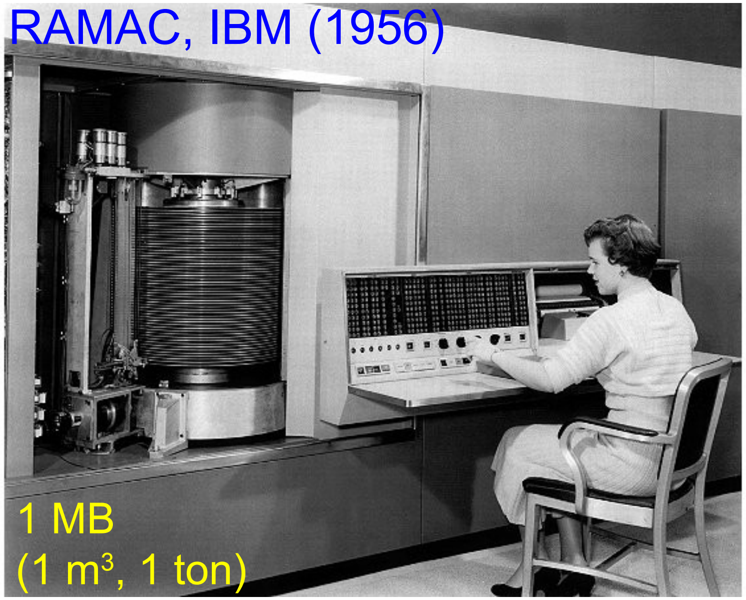
Magnetic data storage
as magnetization direction





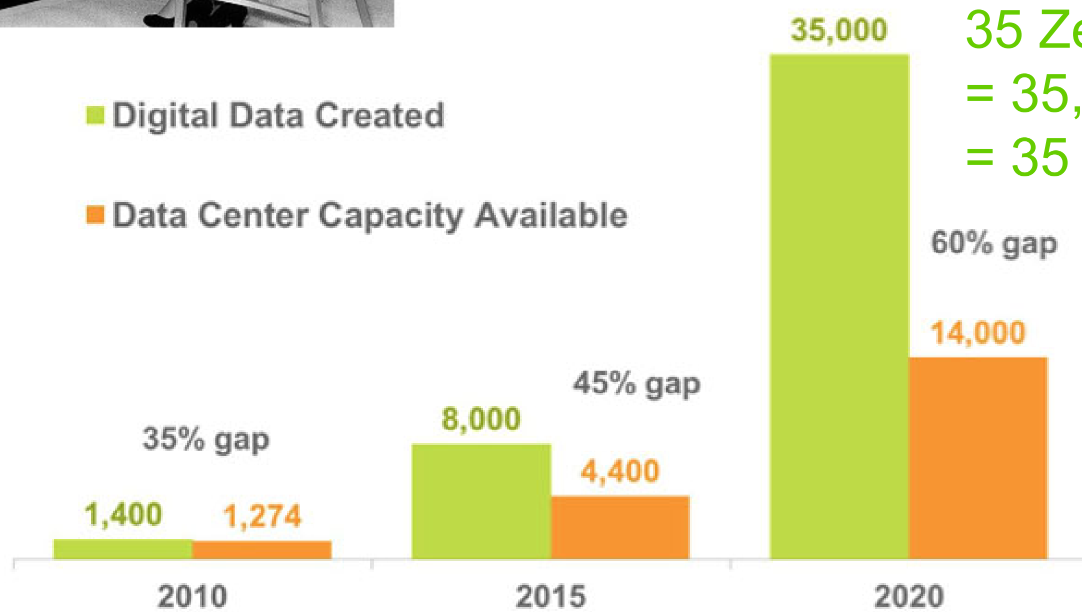
Stöhr et al, Science 259, 858 (1993)

Context: Magnetic data storage



$1\ \text{TB} = 10^6\ \text{MB}$

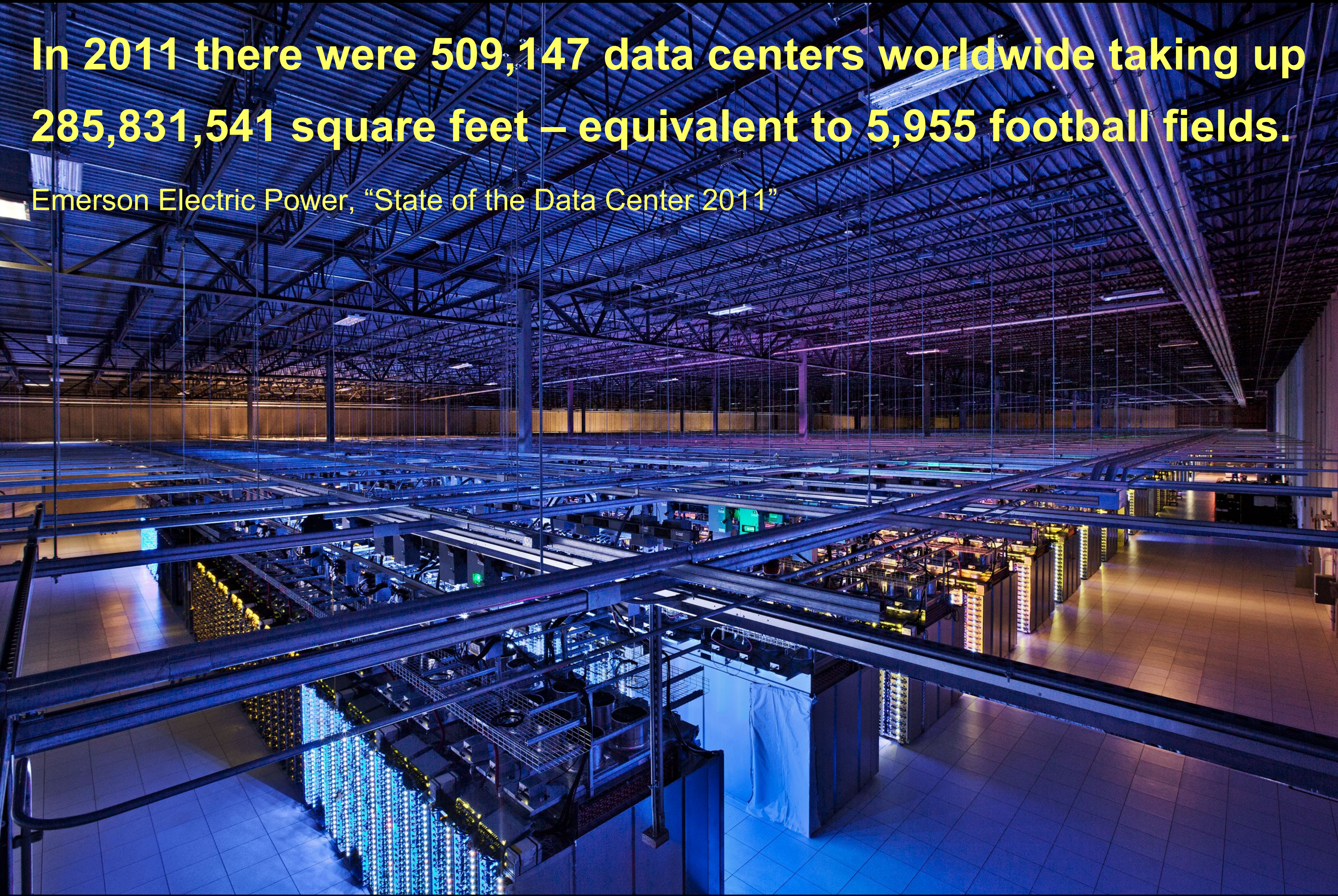
2 TB hard disk (2014)



35 Zetta Byte
= 35,000 Exa Byte
= $35 * 10^9$ TB

Source: IDC Digital Universe Study, 2011, Numbers in Exa-Byte

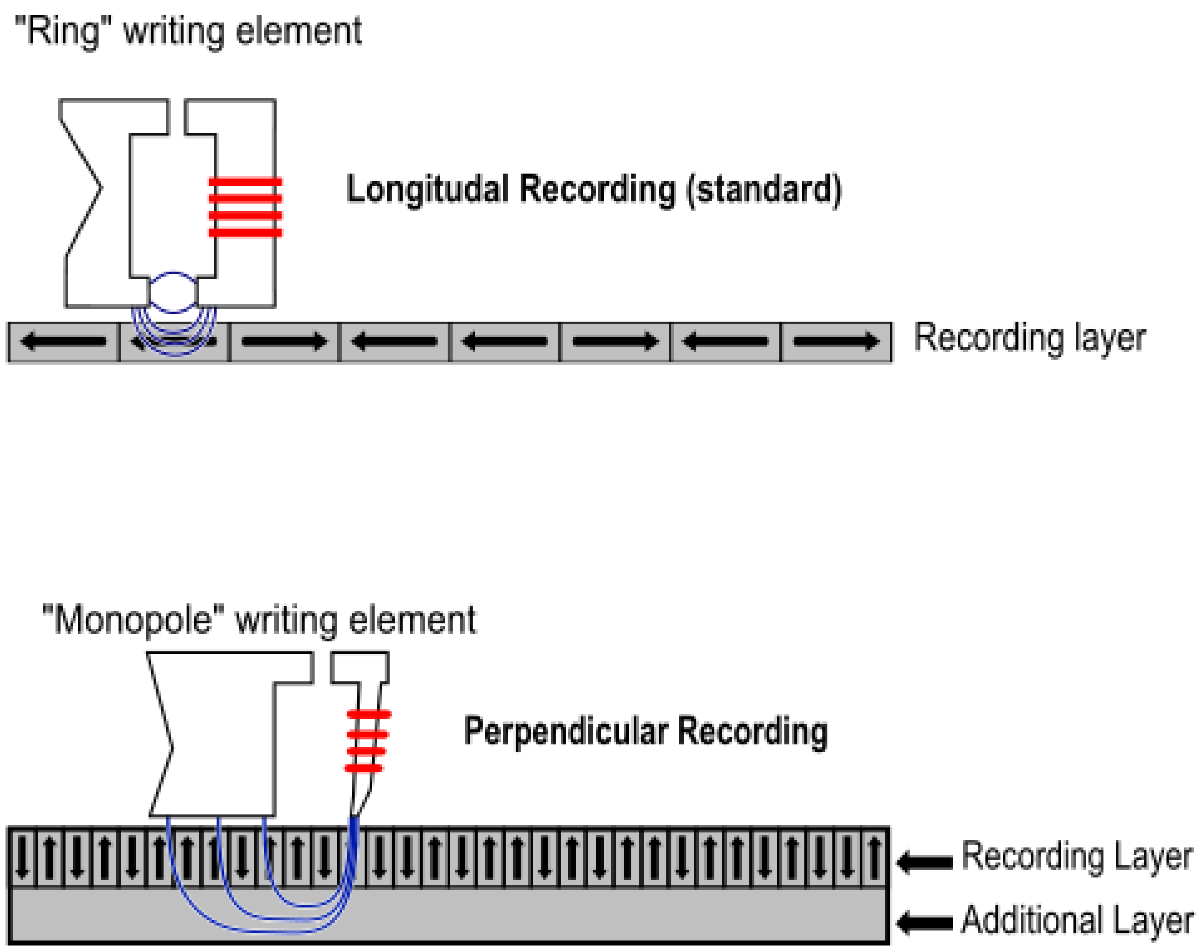
You can't continuously increase the number of storage centers ...



Google data center

You can't fabricate 1000x more disks

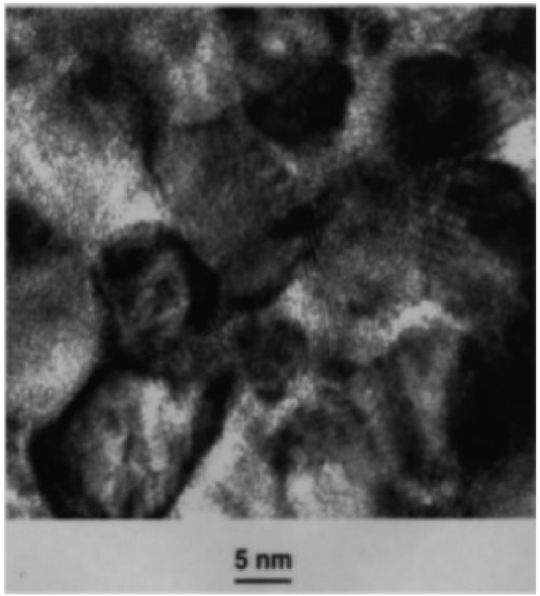
You have to increase the storage density:



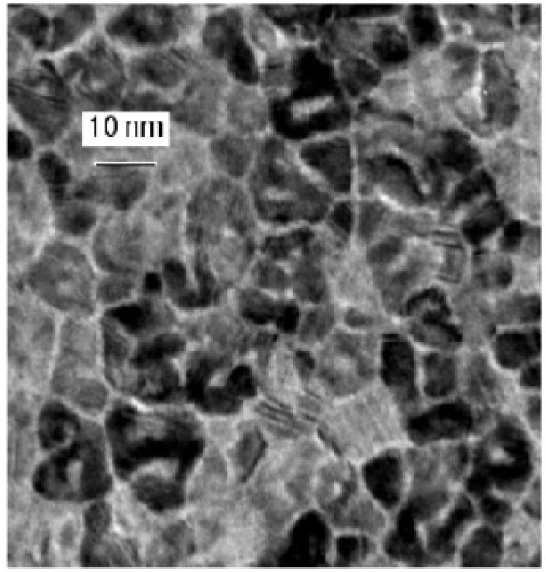
(Wikipedia)

Magnetic media based on CoCrPt

1990 LMR	2000 LMR	2012 PMR

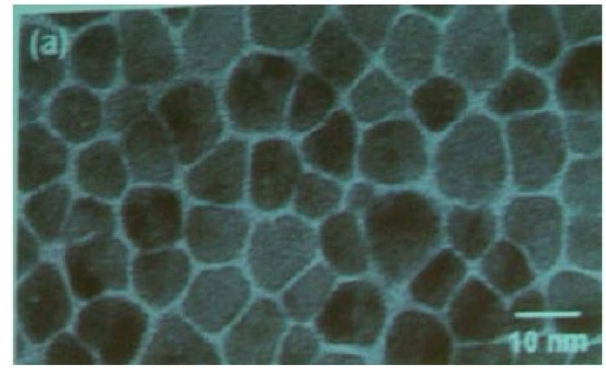


10 Gb/in² product media
12 nm grains, σ_{area} ~ 0.9



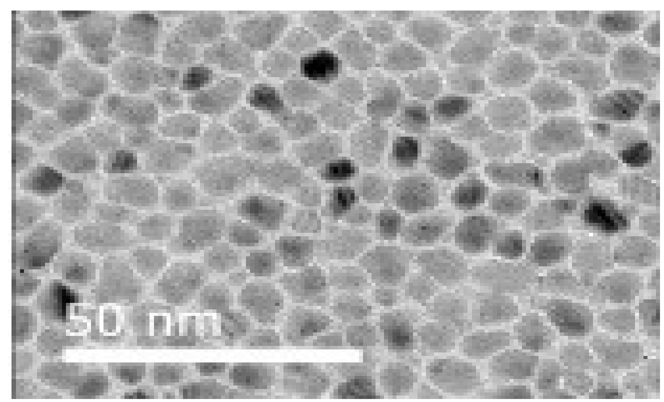
35 Gb/in² prototype media
8.5 nm grains, σ_{area} ~ 0.6

Production:
~750 Gb/in²



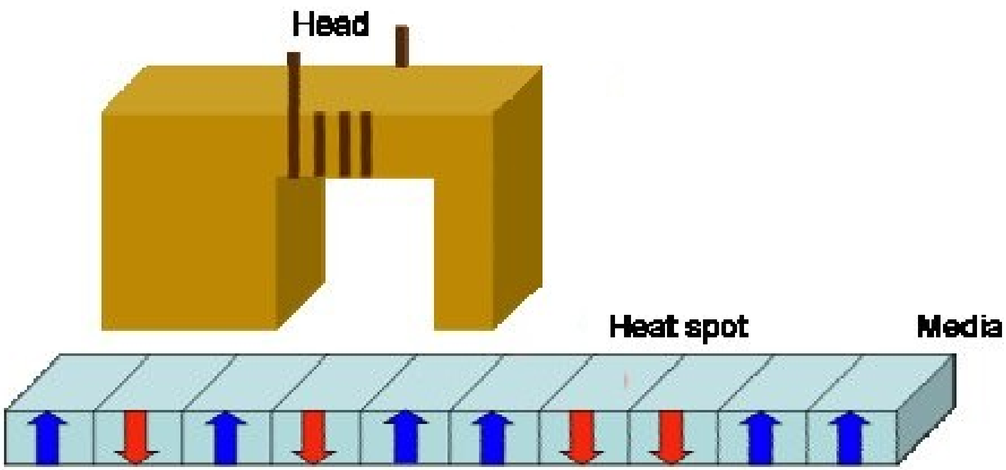
750 Gb/in² prototype media
8.5 nm grains, σ_{area} ~ 0.36

Next generation: FePtX-Y media
- smaller, more uniform grain size
- higher magnetic anisotropy



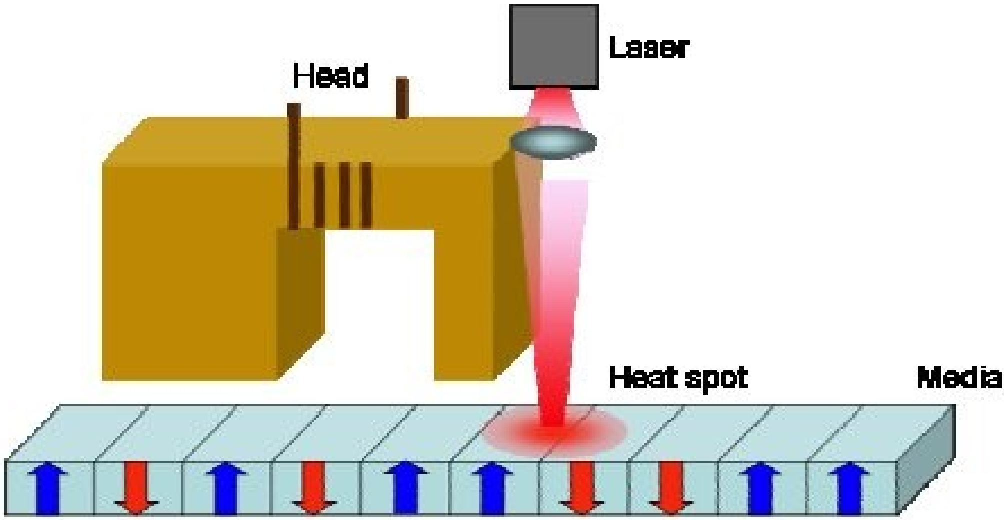
Problem

Field of write head not sufficient to switch magnetization



Idea

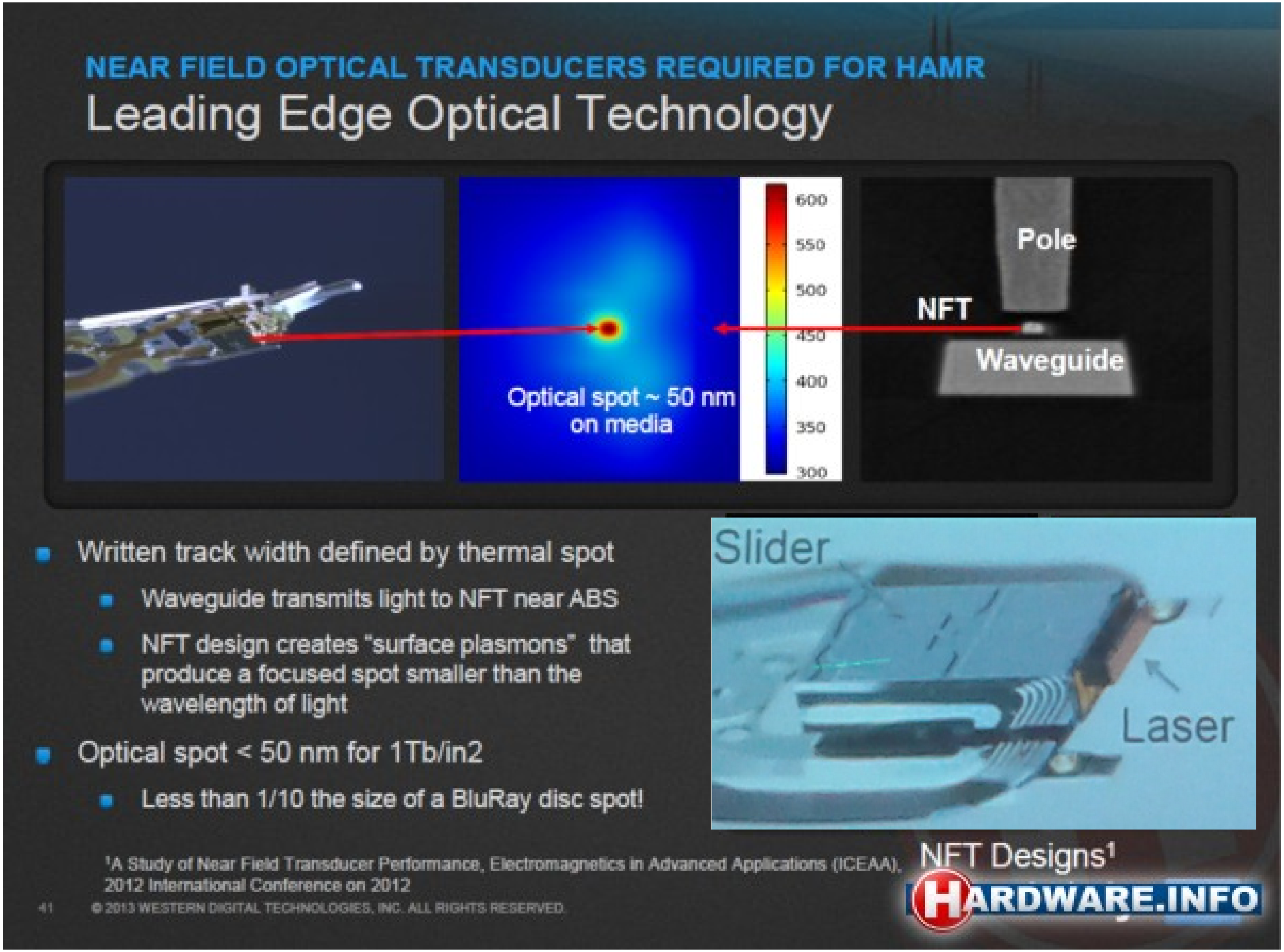
Heat sample to lower locally, temporary the magnetic 'hardness'



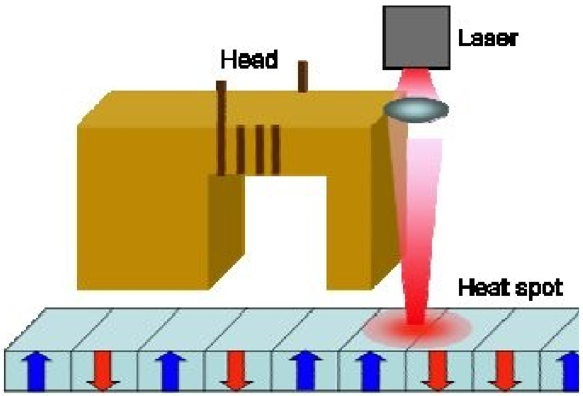
Many problems, but one very fundamental:
Diffraction limited focal spot size > 200 nm

The next generation: Heat Assisted Magnetic Recording

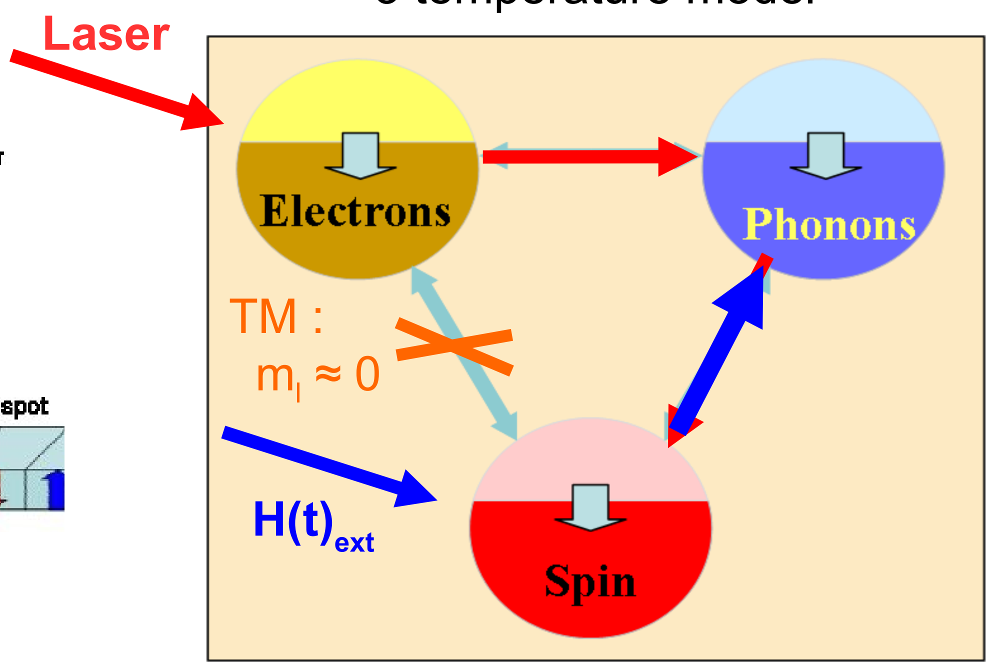
Focusing achieved using surface plasmons on near field antennas



Magnetization reversal by magnetic field pulse



3 temperature model

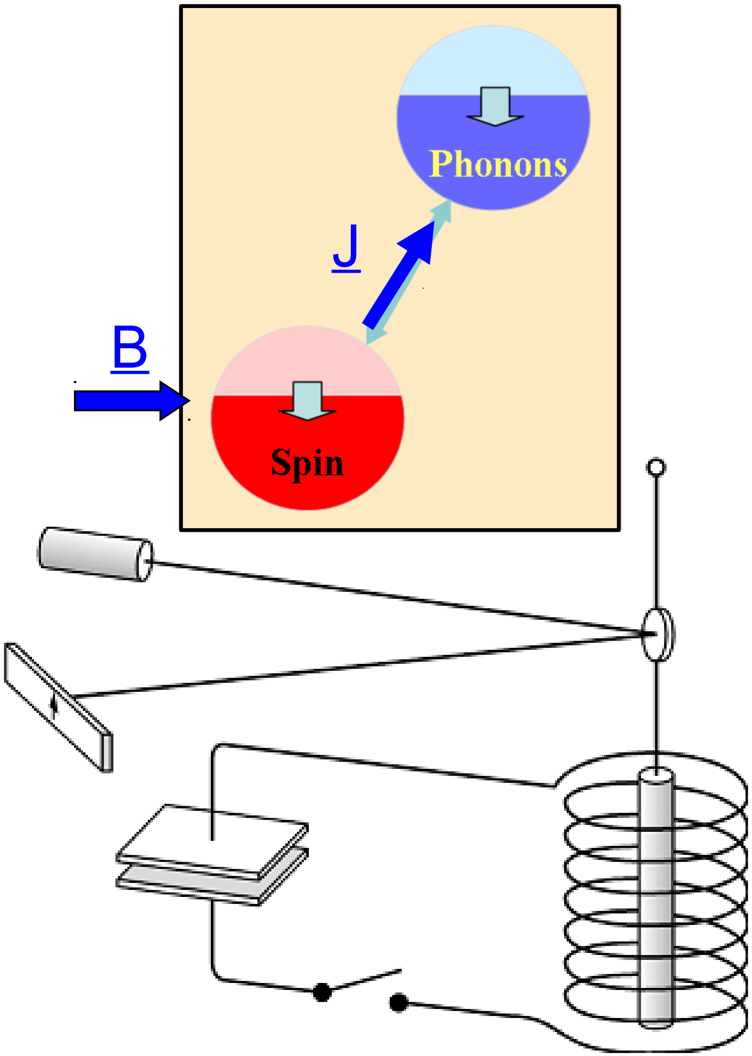


NOTE:

Magnetization is an angular momentum!

When changing the magnetization, this angular momentum needs to be transferred out of the spin system!

Einstein – de Haas effect



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

Picture from Wikipedia

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

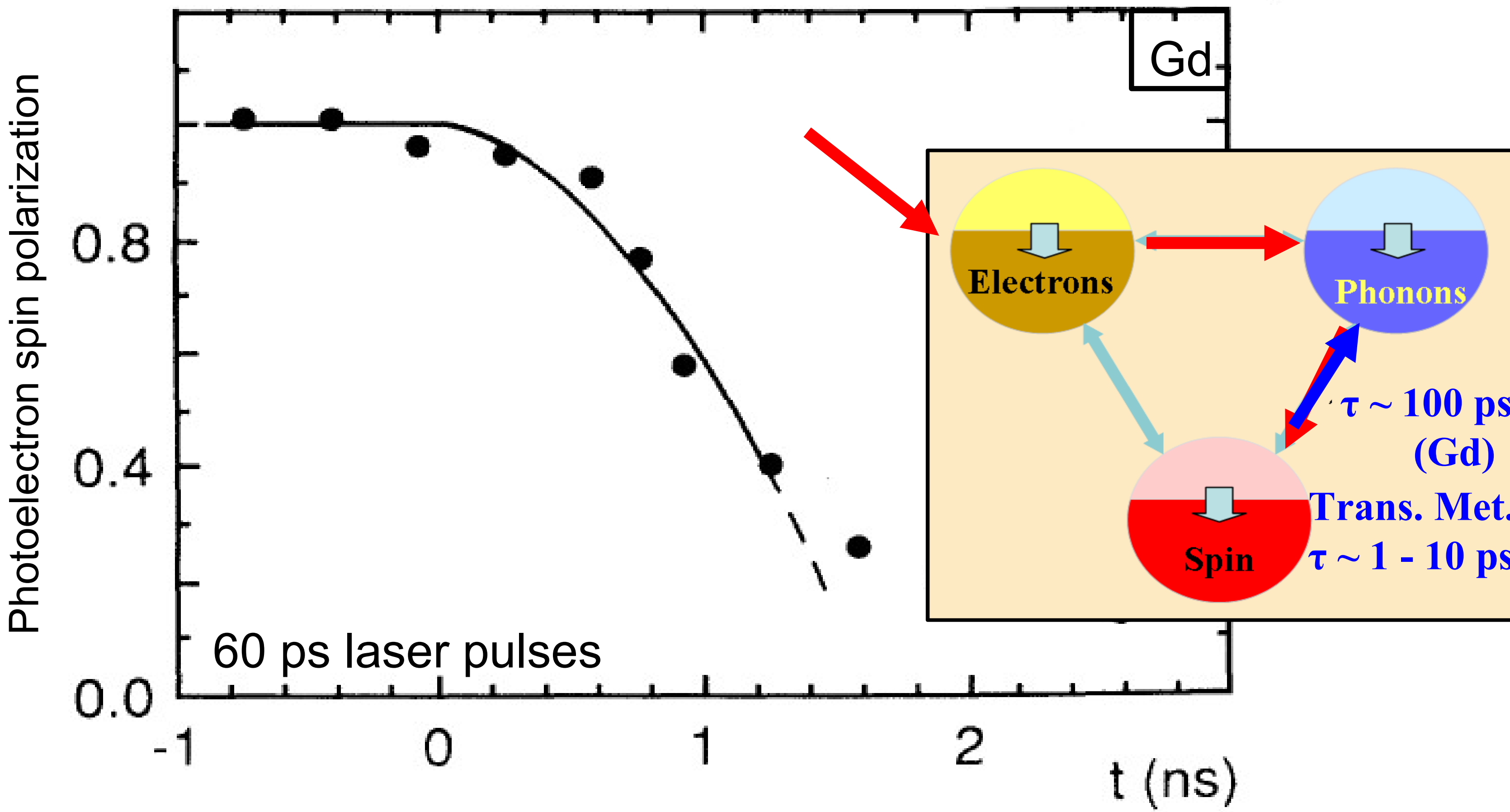
Transfer to lattice via magnon-phonon scattering!

1991: Spin-lattice coupling in Gd ~100 ps

VOLUME 67, NUMBER 23 PHYSICAL REVIEW LETTERS 2 DECEMBER 1991

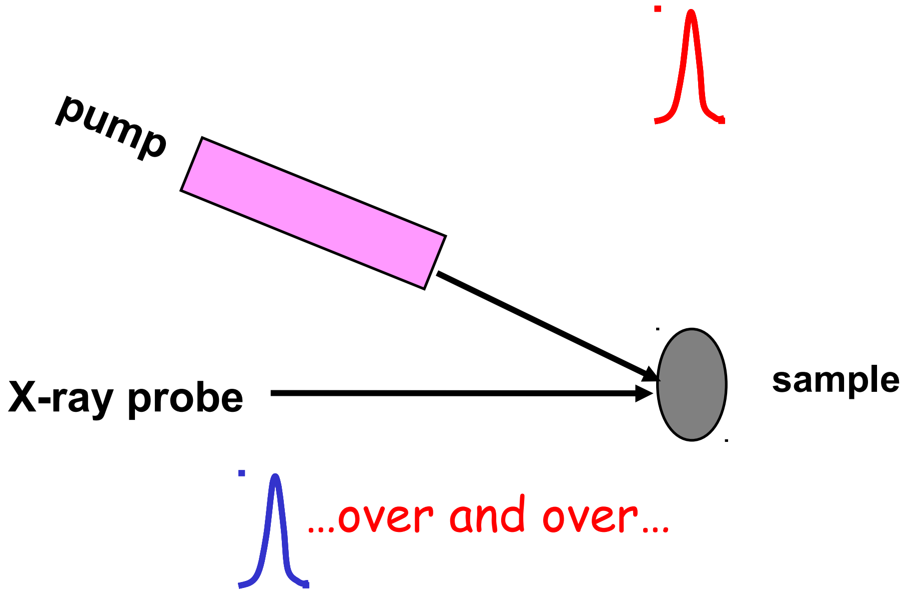
Spin-Lattice Relaxation Time of Ferromagnetic Gadolinium Determined with Time-Resolved Spin-Polarized Photoemission

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



Studying dynamics by pump – probe cycles

Problem: Today not enough intensity for single shot experiments with nanometer spatial and picosecond time resolution

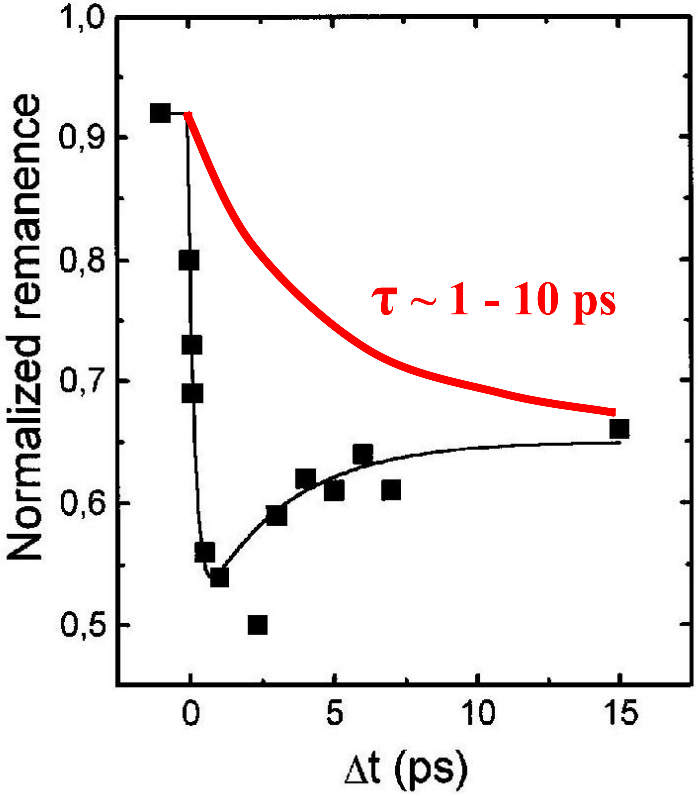


Limitation:

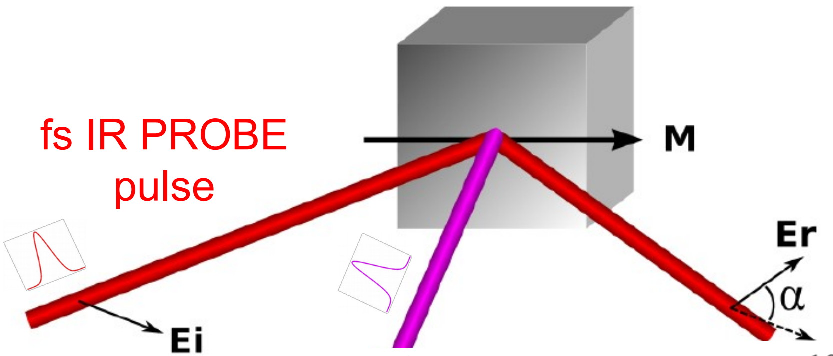
Process has to be repeatable:

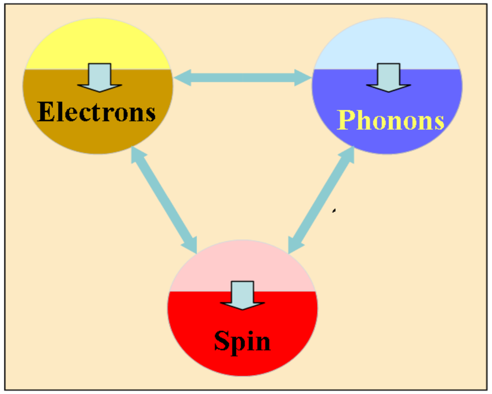
1996: Discovery of ultrafast magnetization dynamics

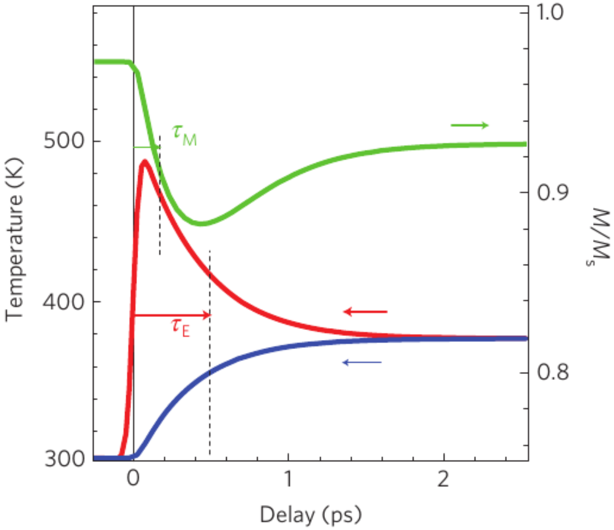
E. Baurepaire *et al.*, PRL **76**, 4250 (1996)



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





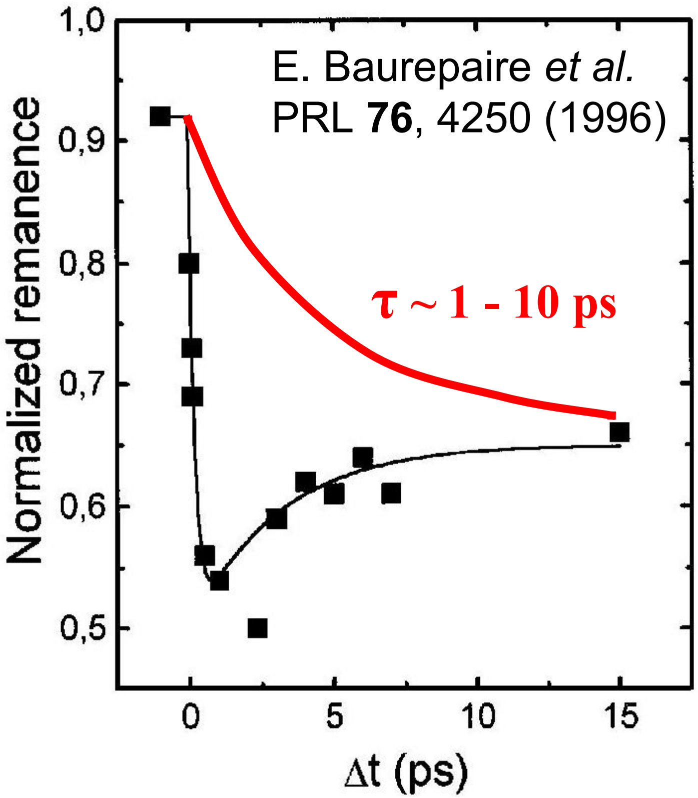


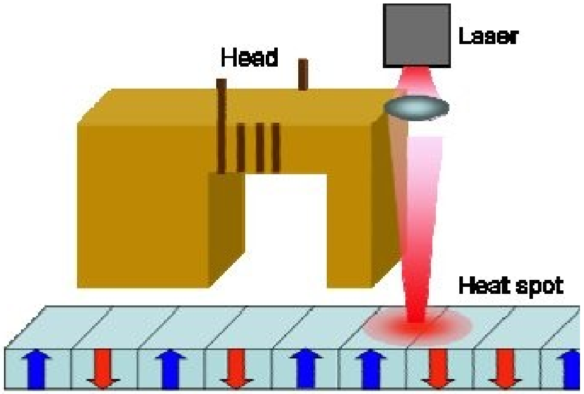
Questions still discussed since 1996:

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

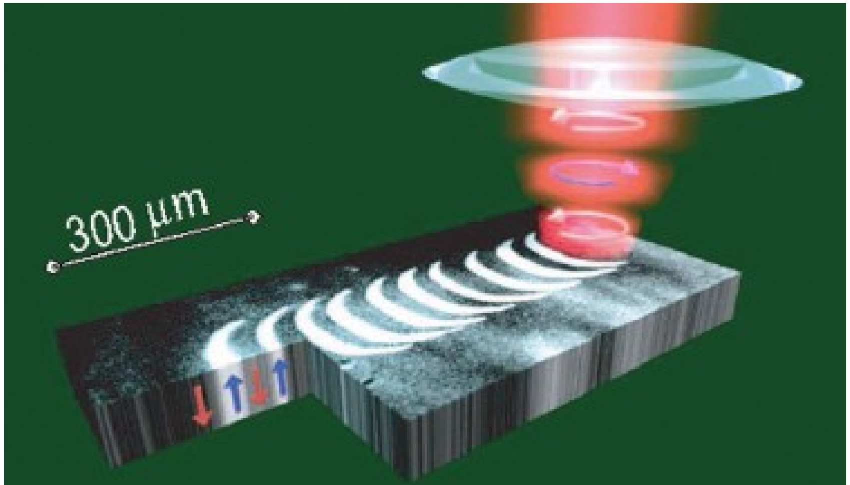
Discovery of ultrafast magnetization dynamics

From ultrafast demagnetization ….





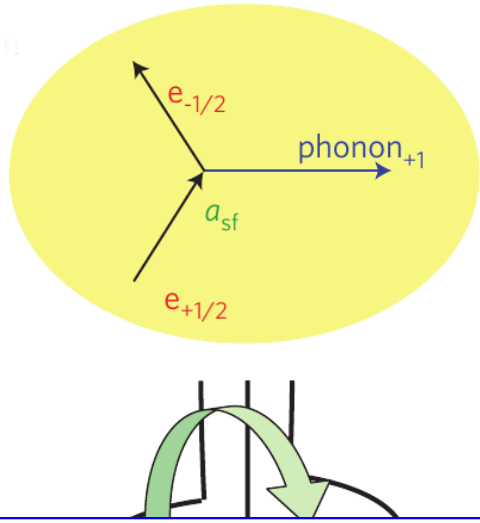
… to ultrafast magnetization *CONTROL*



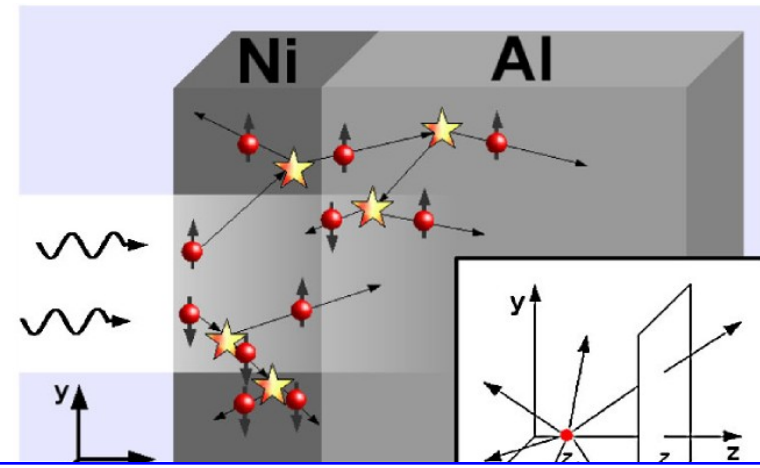
C.D. Stanciu *et al.*, PRL 99, 047601 (2007)

Most discussed potential mechanisms

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

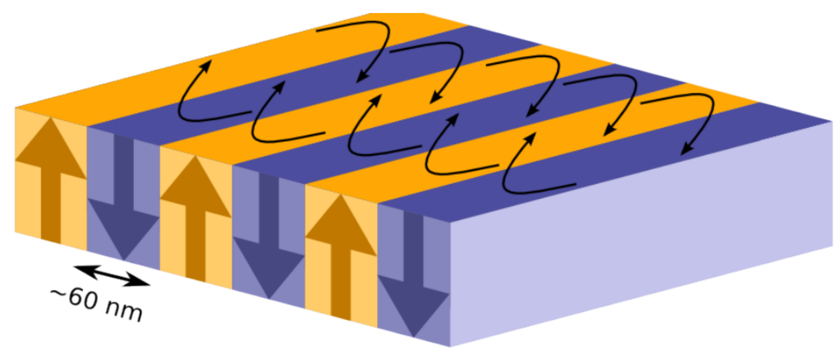


Angular momentum transport by hot, spin-polarized electrons (non-local mechanism)



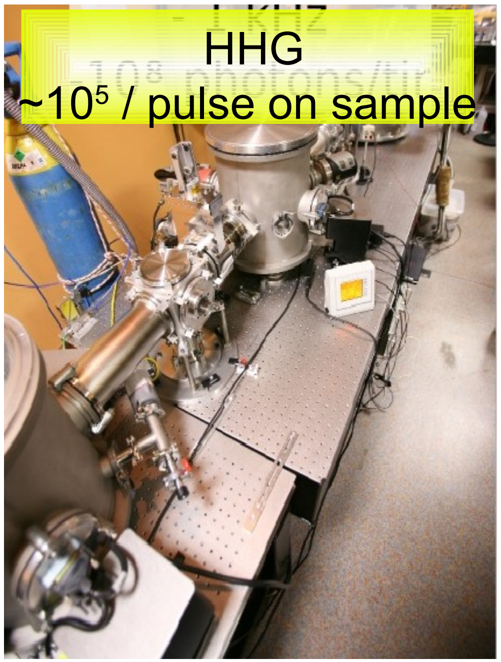
- Requires ~10 nm spatial resolution
- Element sensitivity
- Access to buried layers
- Strong dichroism signal

→ properties of X-ray based techniques

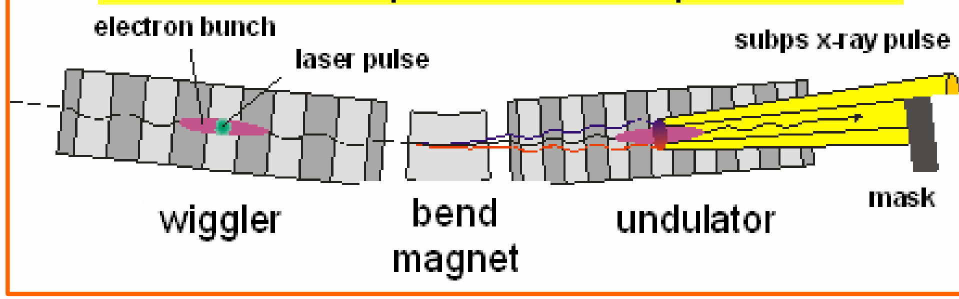


fs pulsed X-ray sources

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



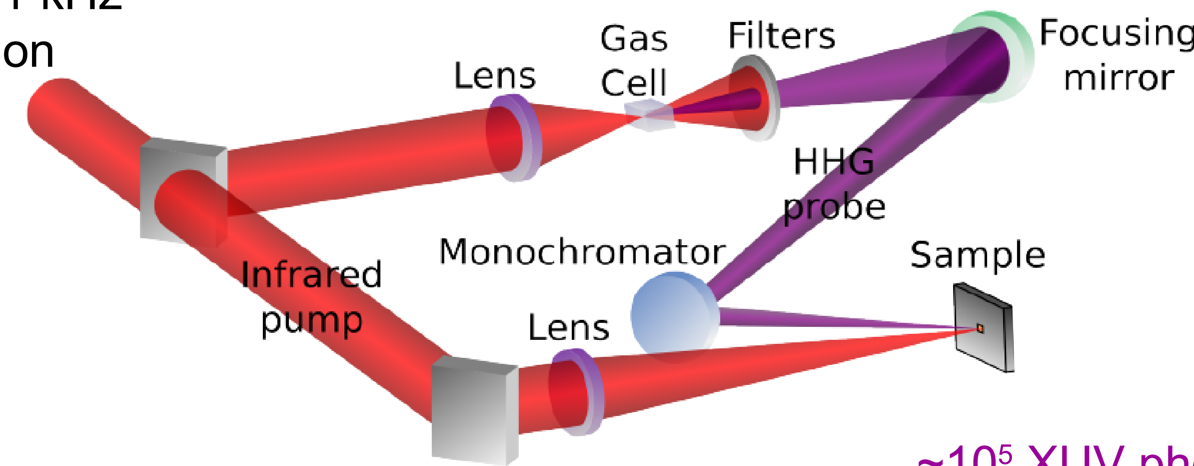
Femtoslicing (BESSY, SLS, ***SOLEIL***)
~10^6 /pulse on sample



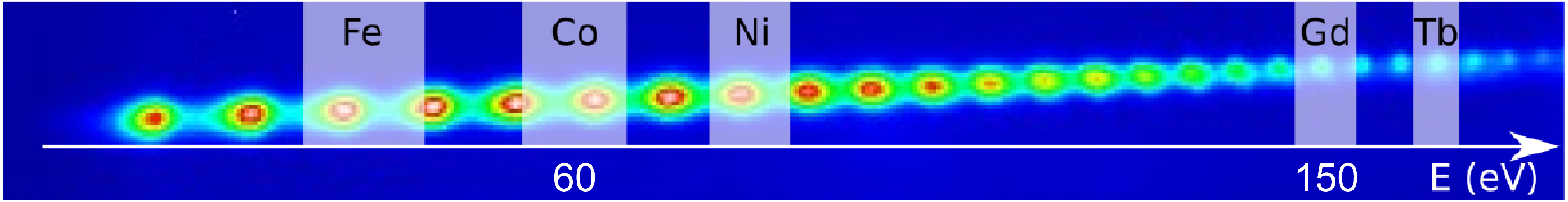
Higher Harmonic Generation of XUV radiation

Boris Vodungbo (LOA/LCPMR)

800 nm, 35 fs, 1 kHz
10 mJ / impulsion



~10^5 XUV photons per pulse
on sample (E/ΔE ~ 10^2)



Key advantages:
- 'easier' accessible, since small scale light source
- jitter free pump-probe setup (IR limited total time resolution)

Acknowledgement

LCPMR	- *B. Vodungbo*, S. Chiuzbaian, R. Delaunay, ...
Synch. SOLEIL	- N. Jaouen, F. Sirotti, M. Sacchi…
IPCMS Strasbourg	- C. Boeglin, E. Beaurepaire, …
LOA Palaiseau	- J. Gautier, P. Zeitoun, ...
Thales/CNRS	- R. Mattana, V. Cros, …

TU Berlin	- S. Eisebitt, *C. von Korff Schmising*, B. Pfau, ...
DESY / U.Hamburg	- G. Grübel, L. Müller, C. Gutt, H.P. Oepen, ...

LCLS	- B. Schlotter, J. Turner, ...
SLAC / Stanford U.	- A. Scherz (→ XFEL), J. Stohr, H. Dürr, A. Ried, …

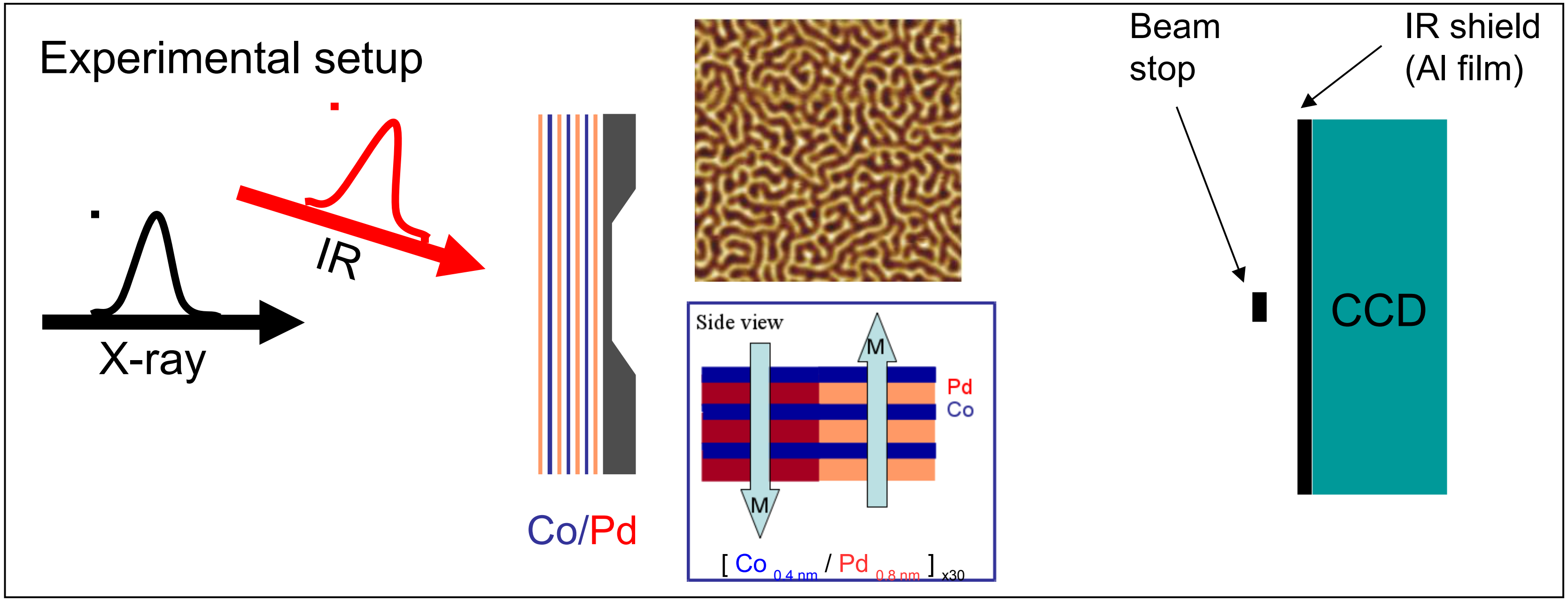
SLS / PSI	- M. Buzzi, J. Raabe, F. Nolting, …
LMN / PSI	- M. Makita, C. David, ...

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***

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



Integrated intensity → measure of the local magnetization

XMCD spectroscopy

Circular polarization

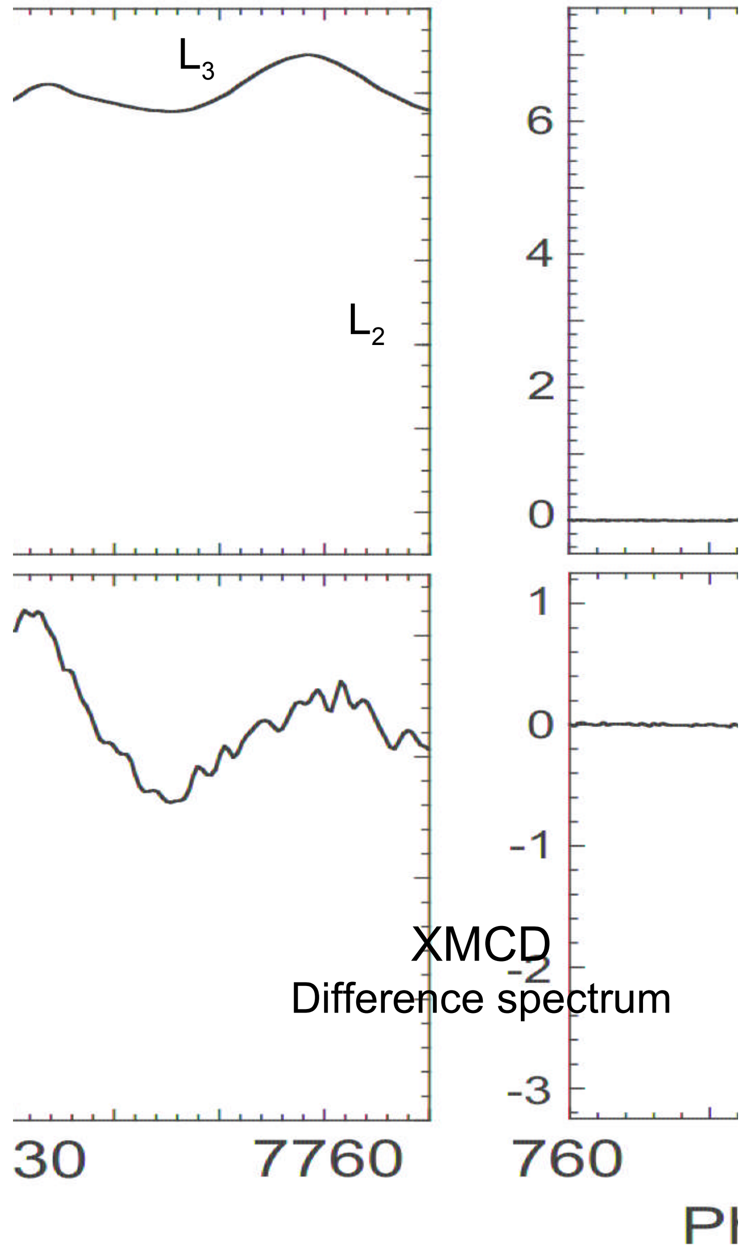
Right (solid)

Left (dashed)

Magnetization reversal

Right (solid)

Left (dashed)

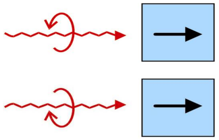


XMCD spectroscopy

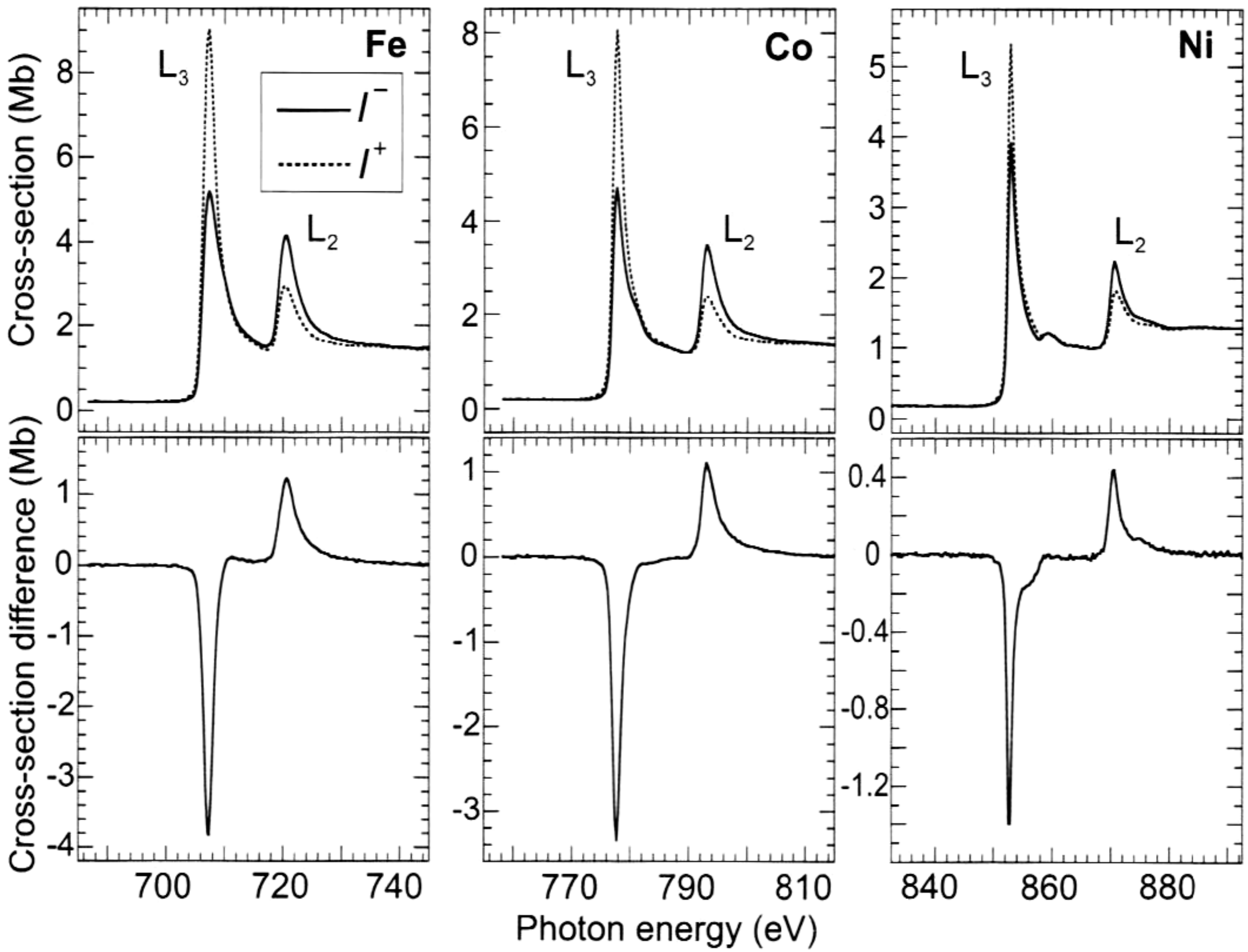
Circular polarization

Right (solid)

Left (dashed)

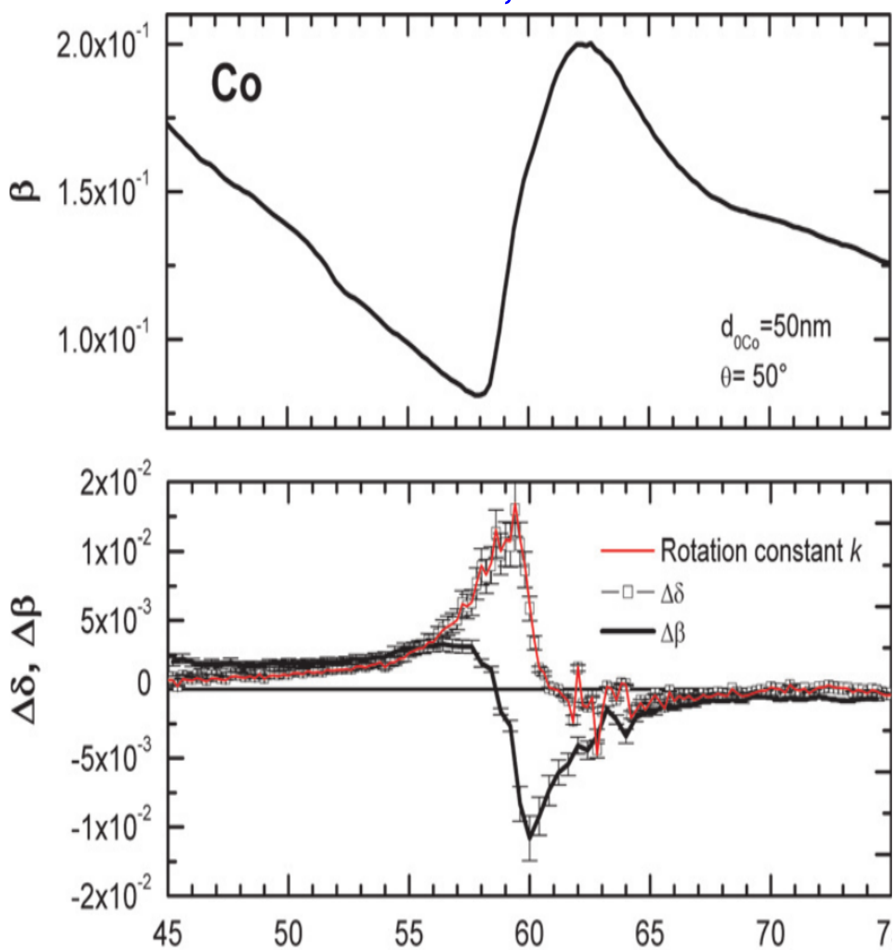


Element specific magnetization probe



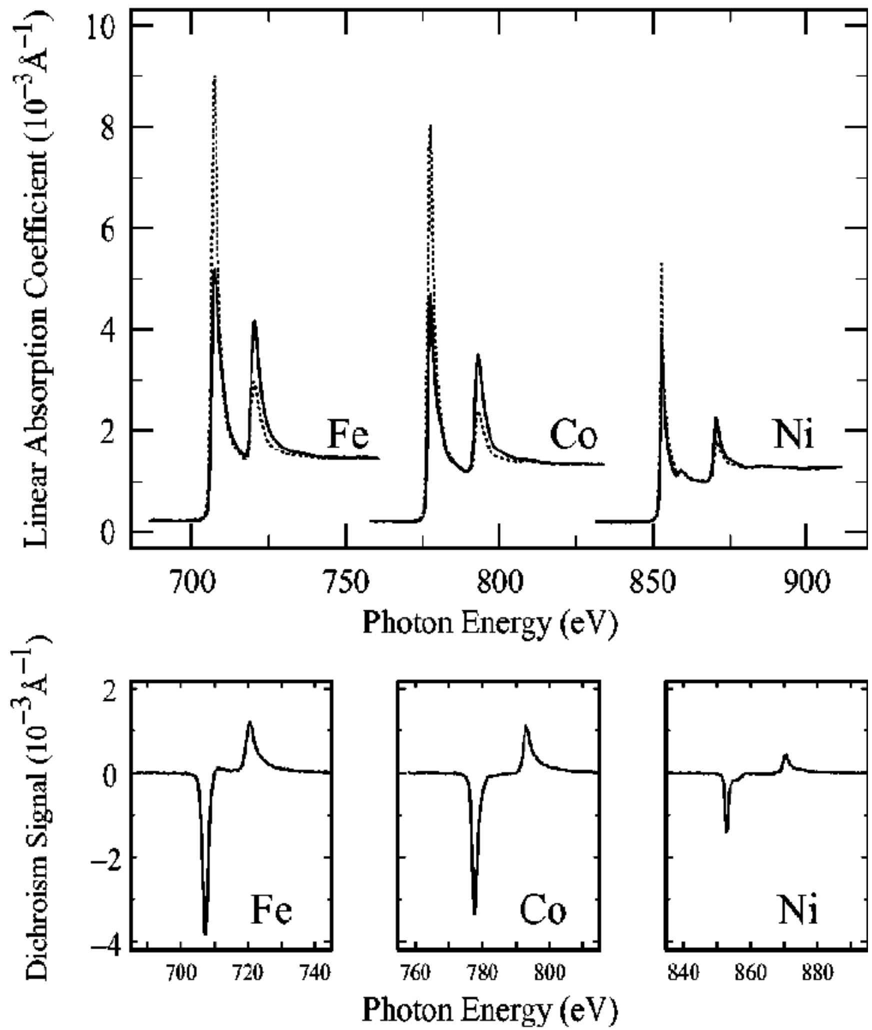
XMCD of transition metal $M_{2,3}$ and $L_{2,3}$ edges

$M_{2,3}$



S. Valencia et al., New J. Phys. 8, 254 (2006)

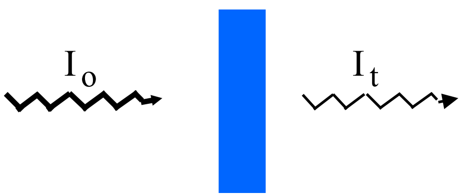
$L_{2,3}$



R. Nakajima et al., Phys. Rev. B 59, 6421 (1999)

XMCD in Absorption and Scattering

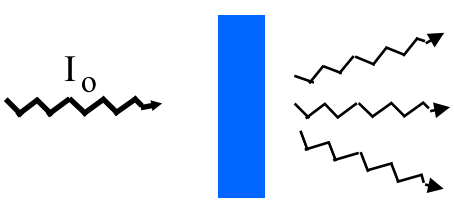
Absorption



Sample density ρ

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

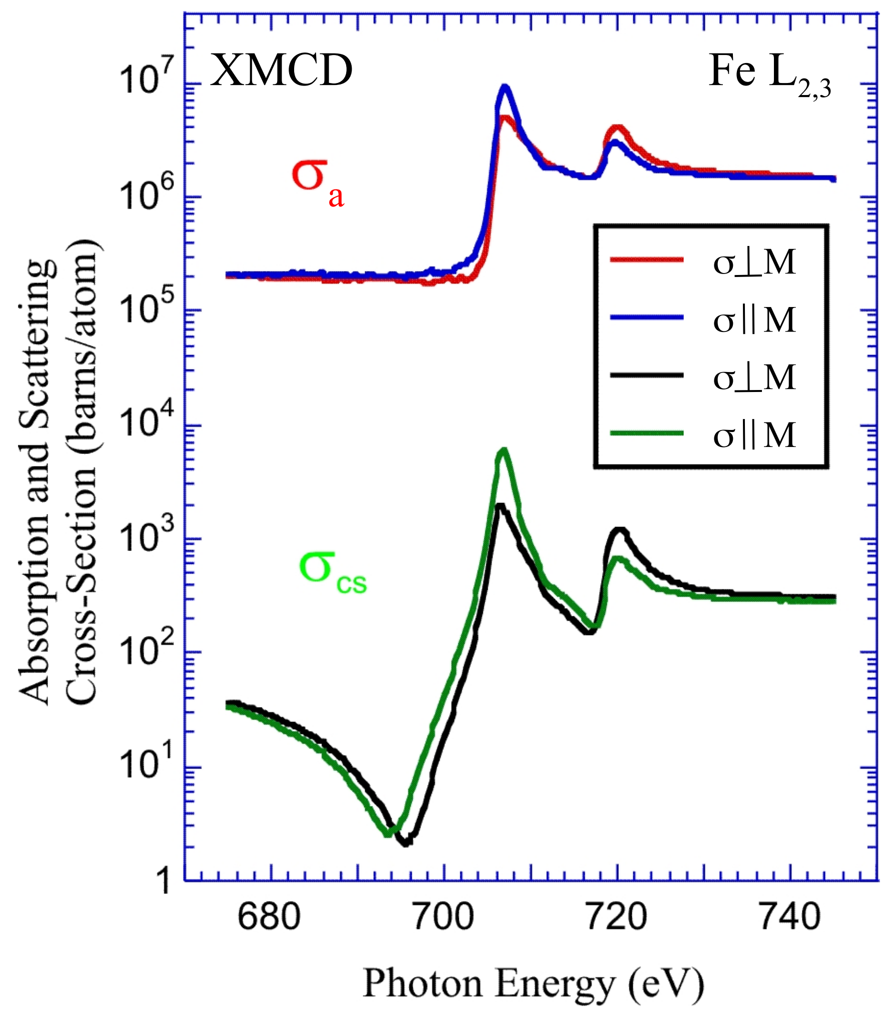
Small Angle Scattering



Sample density ρ

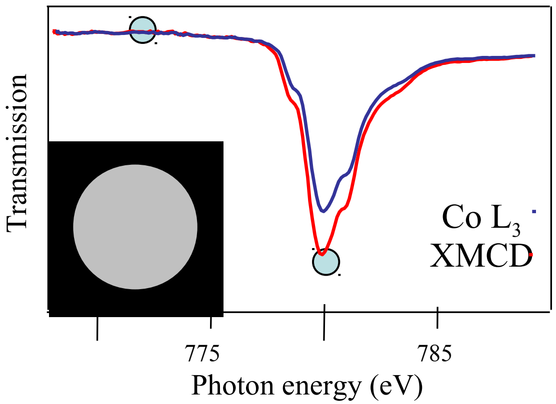
$$I_{cs} \sim I_o \Delta\sigma_{cs} / \sigma_a$$

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

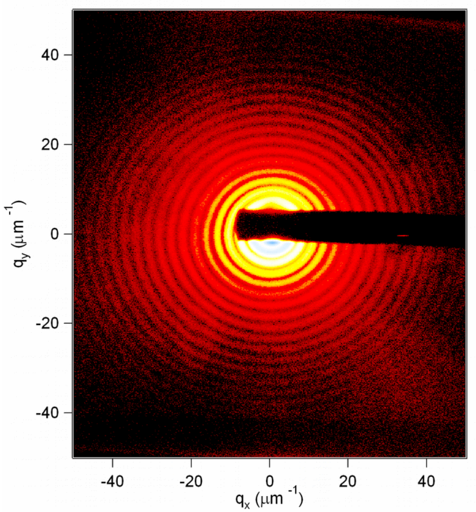


Data from Jeff Kortright (LBNL)

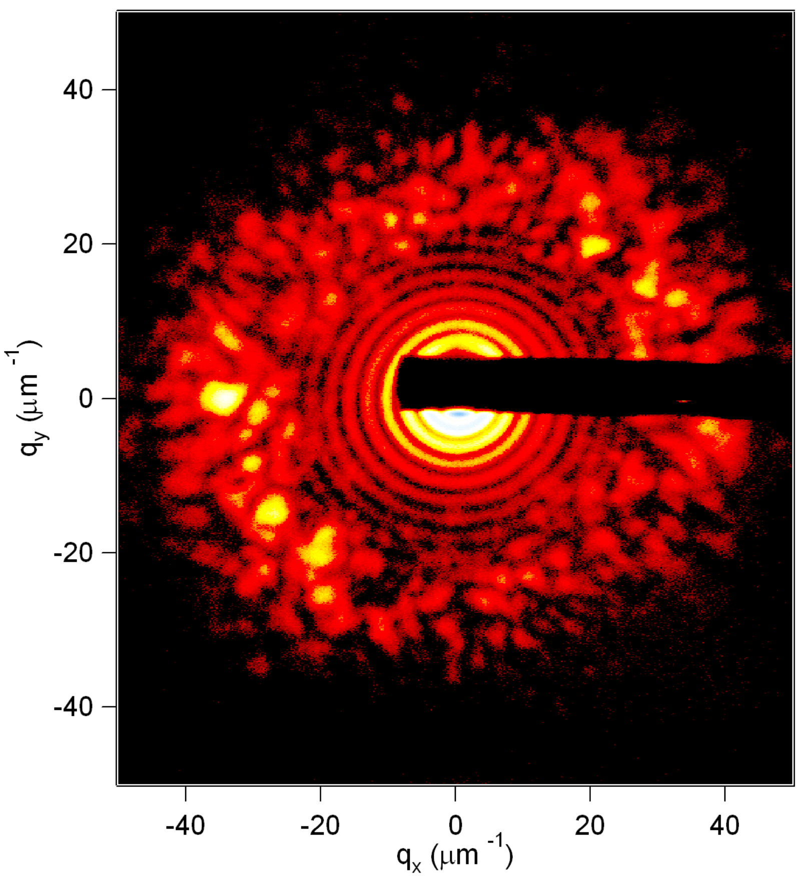
Magnetic scattering contrast



Below Resonance



On Resonance



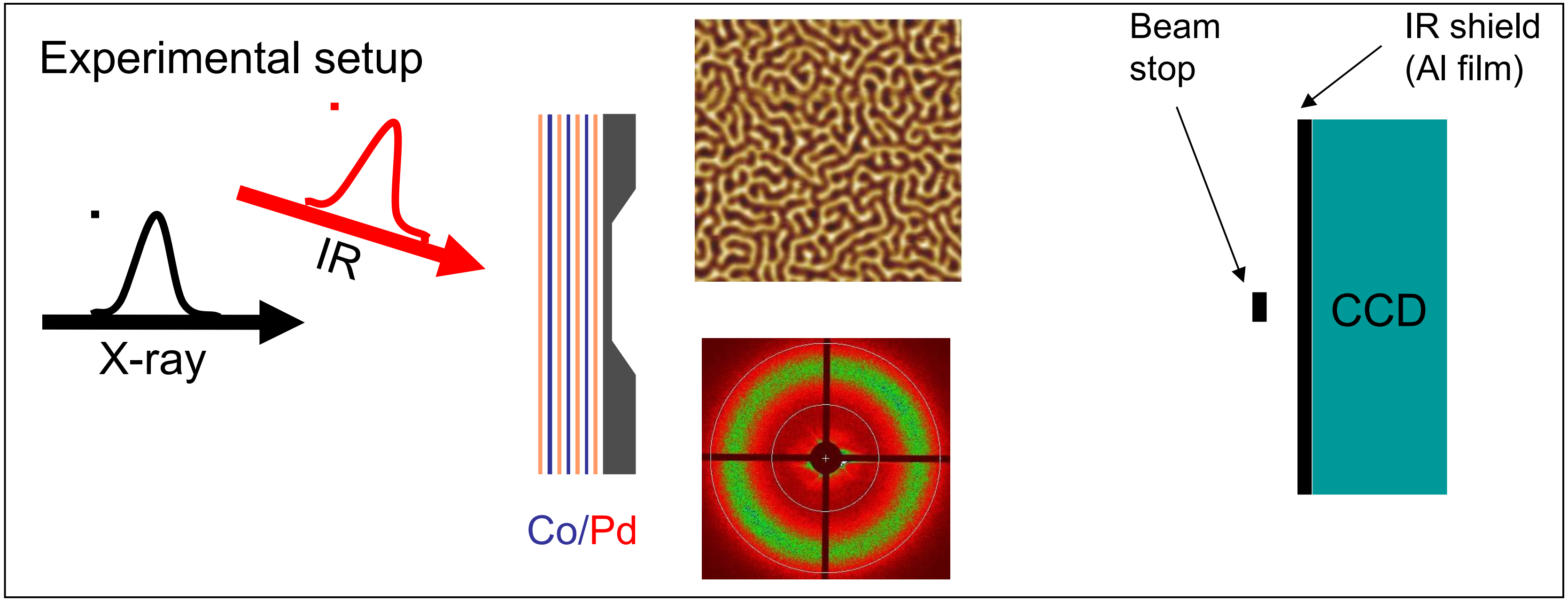
λ = 1.59 nm, 2.5 mm ⊘ Pinhole
fully coherent illumination: visibility = 1, M = 1

Resonant scattering for local probing of magnetization

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

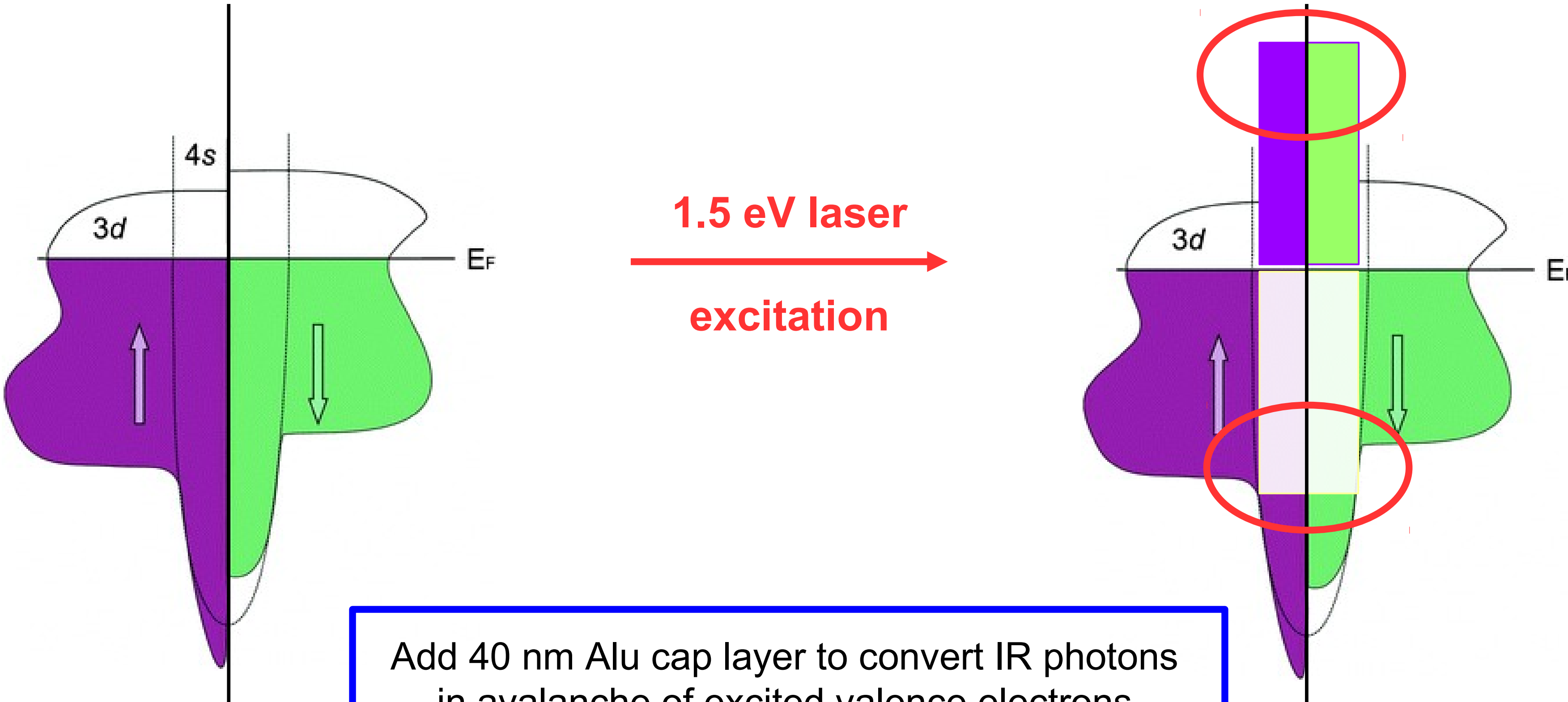
Magnetically dichroic absorption edges of transition metals:

- LCLS: $\mathbf{L_{2,3}}$ (700 – 850 eV)
- FLASH, FERMI (HHG): $\mathbf{M_{2,3}}$ (55 - 65 eV ↔ **37th – 41st** harmonic)

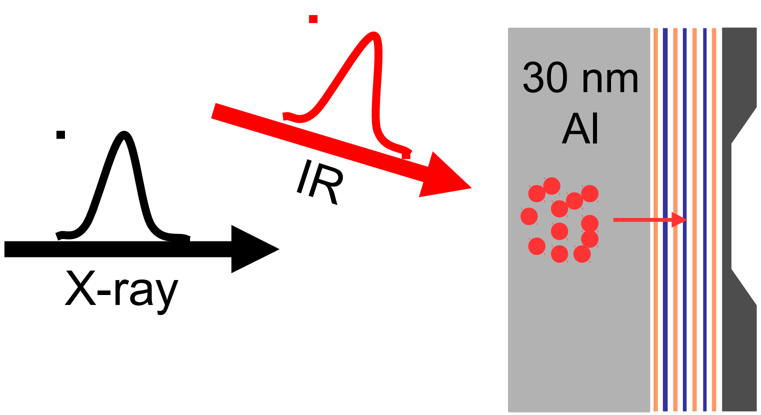


Integrated intensity → measure of the local magnetization

Relevance of hot, directly excited valence electrons



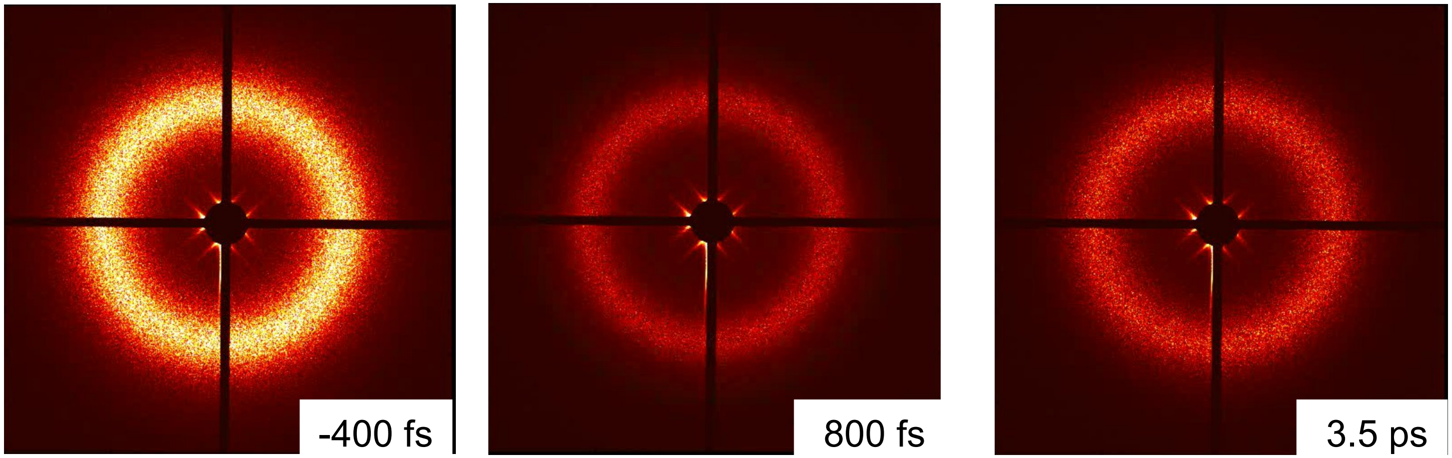
Add 40 nm Alu cap layer to convert IR photons in avalanche of excited valence electrons

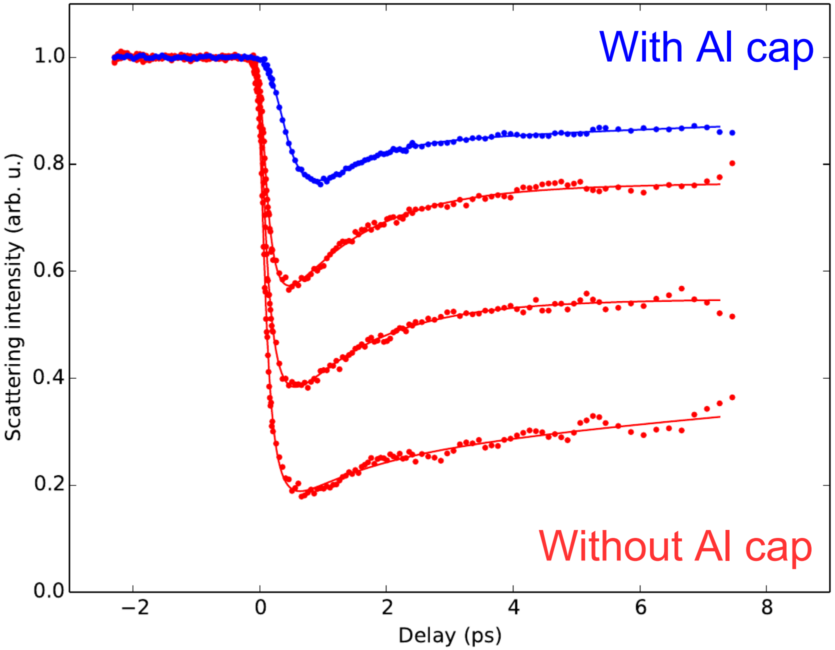


Hot electron excited ultrafast magnetization dynamics

SXR @ LCLS

B. Vodungbo, to be published (2015)





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

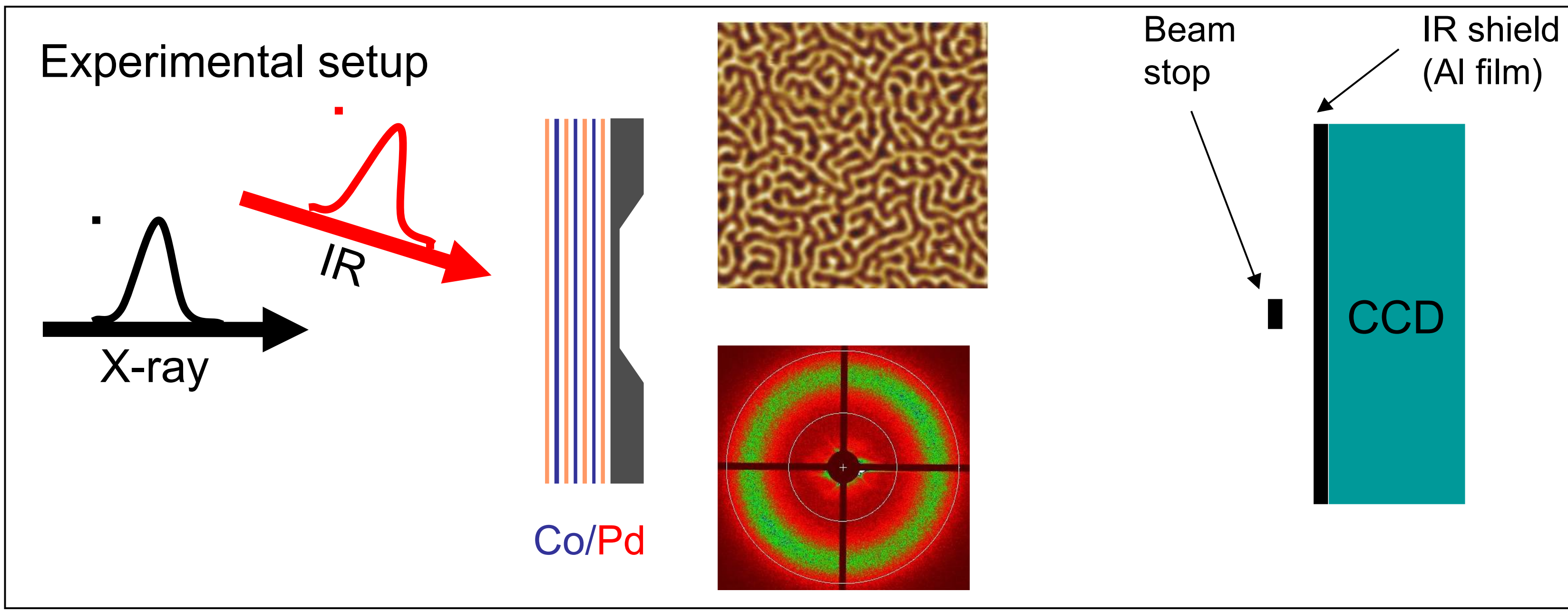
See also from BESSY Slicing-Source:
A. Eschenlohr et al., Nat. Mater 12, 332 (2013)

Resonant scattering for local probing of magnetization

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

Magnetically dichroic absorption edges of transition metals:

- LCLS: **$L_{2,3}$** (700 – 850 eV)
- FLASH, FERMI (HHG): **$M_{2,3}$** (55 - 65 eV ↔ **37^{th} – 41^{st}** harmonic)



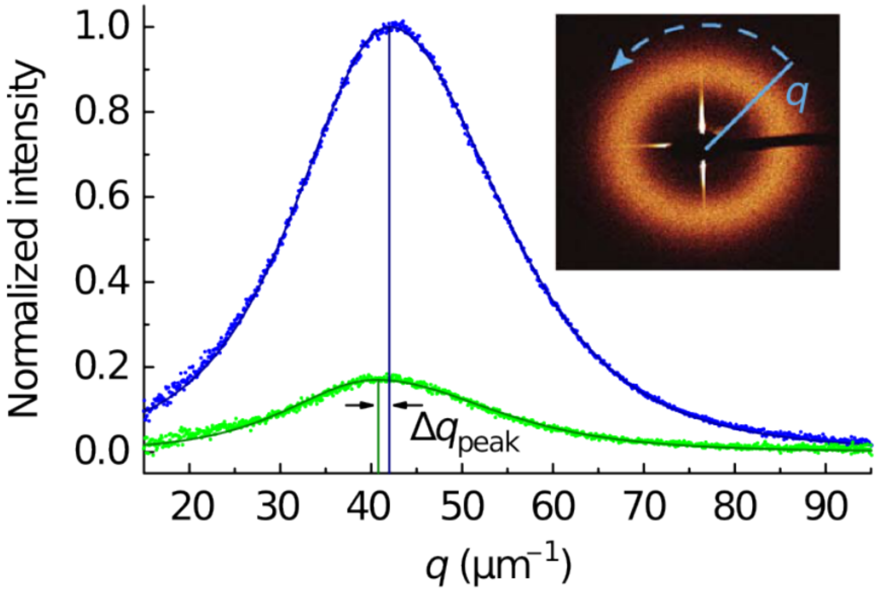
Integrated intensity → measure of the local magnetization

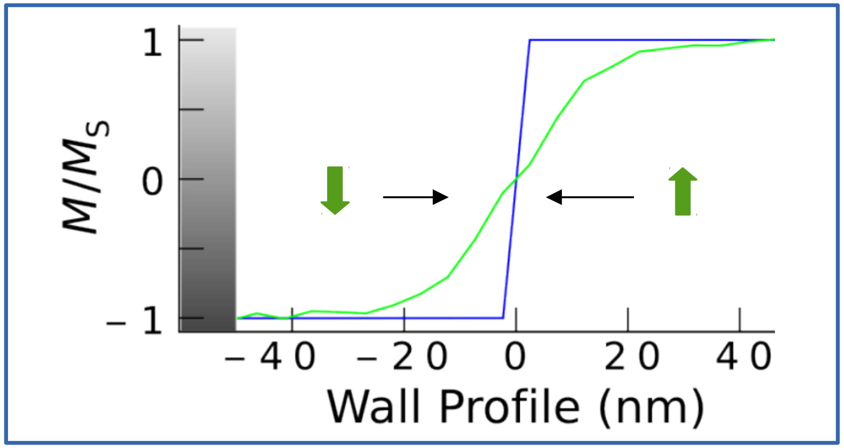
Form of scattering pattern → spatial information

UF demagnetization in the ***strong*** IR pump power limit

FLASH

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

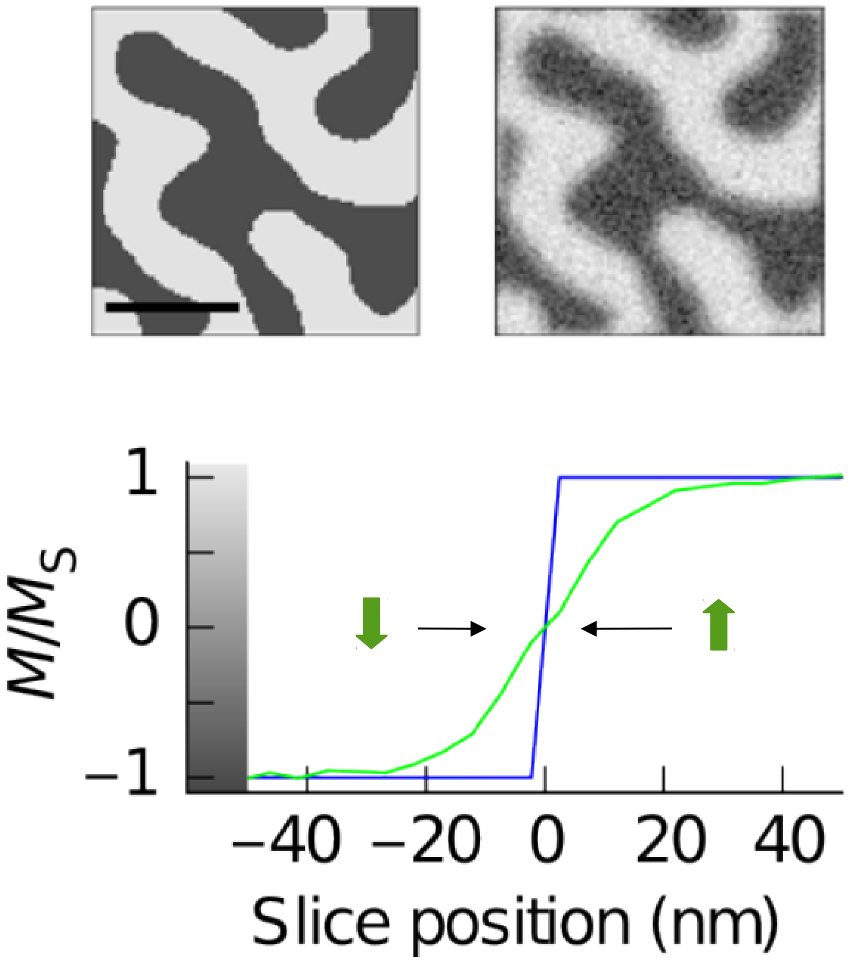




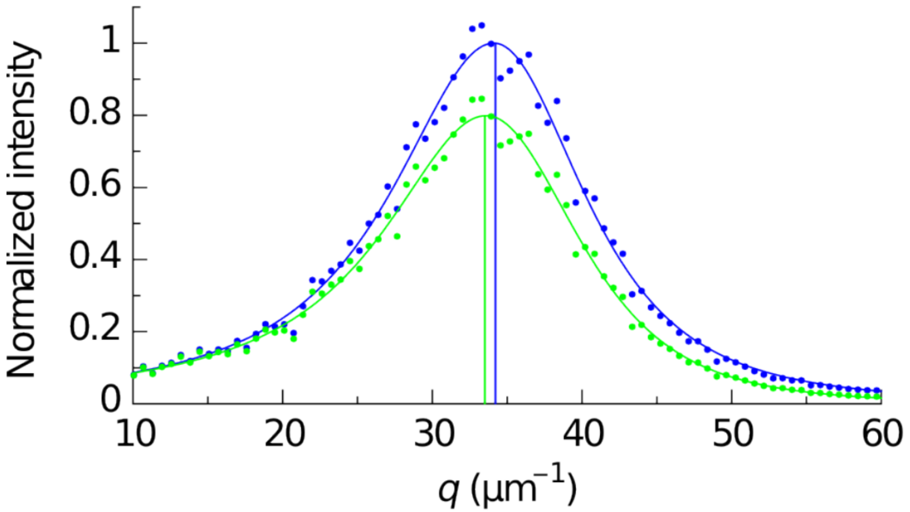
Angular momentum transport by hot polarized electrons

Interpretation based on model of superdiffusive spin transport developed 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

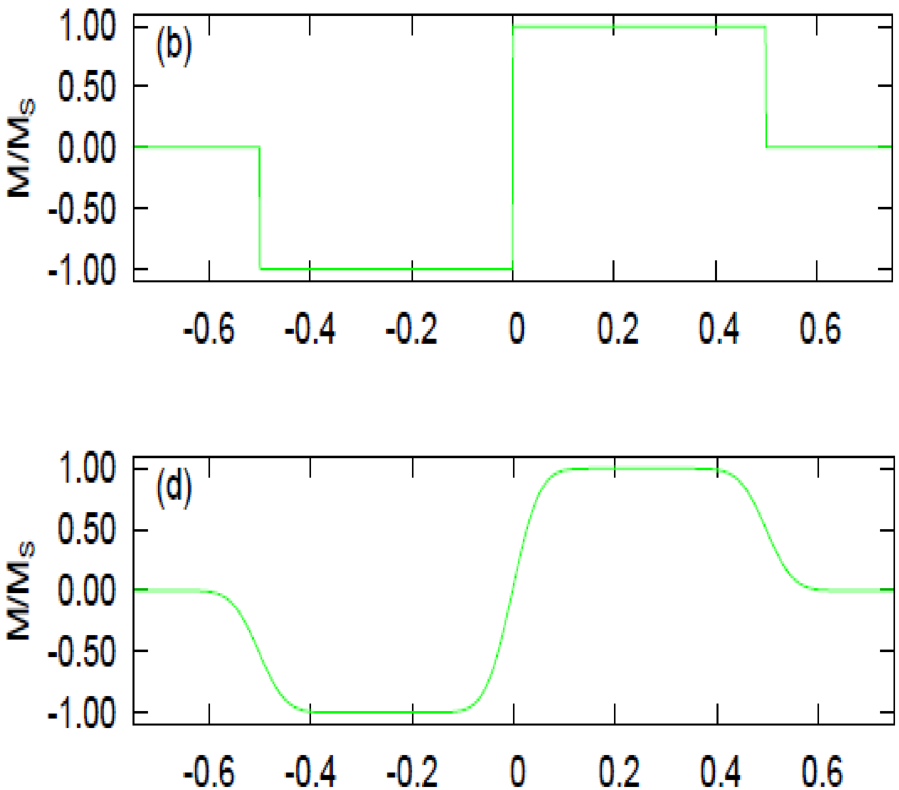


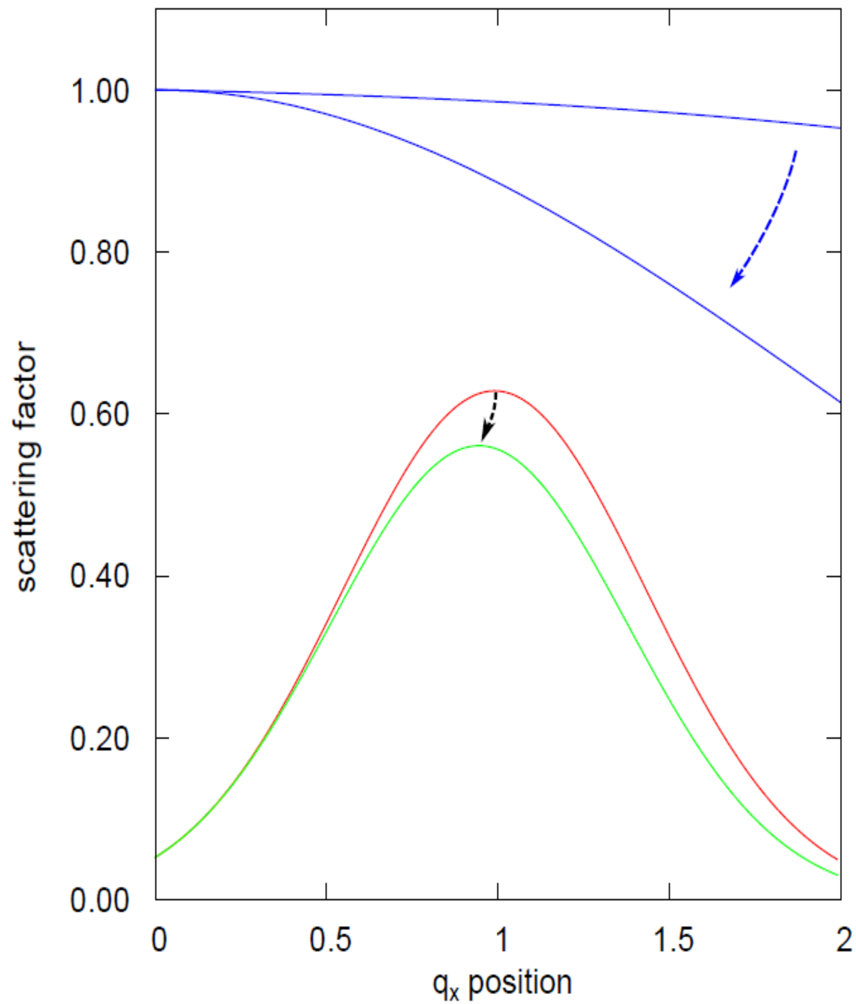
B. Pfau et al., Nat. Comm. 3, 1100 (2012)

UF demagnetization in the high IR pump power limit

SCIENCES SORBONNE UNIVERSITÉ

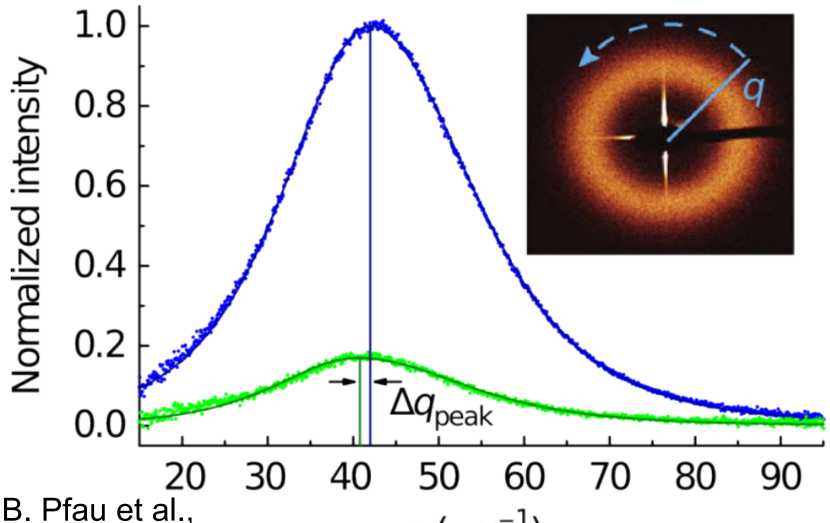
Domain wall broadening introduces shift of center of mass of 1st order scattering peak associated with domain periodicity





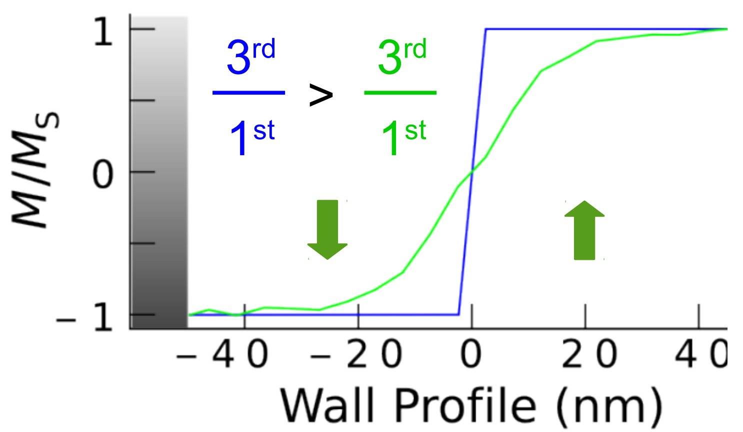
Probing domain wall evolution directly

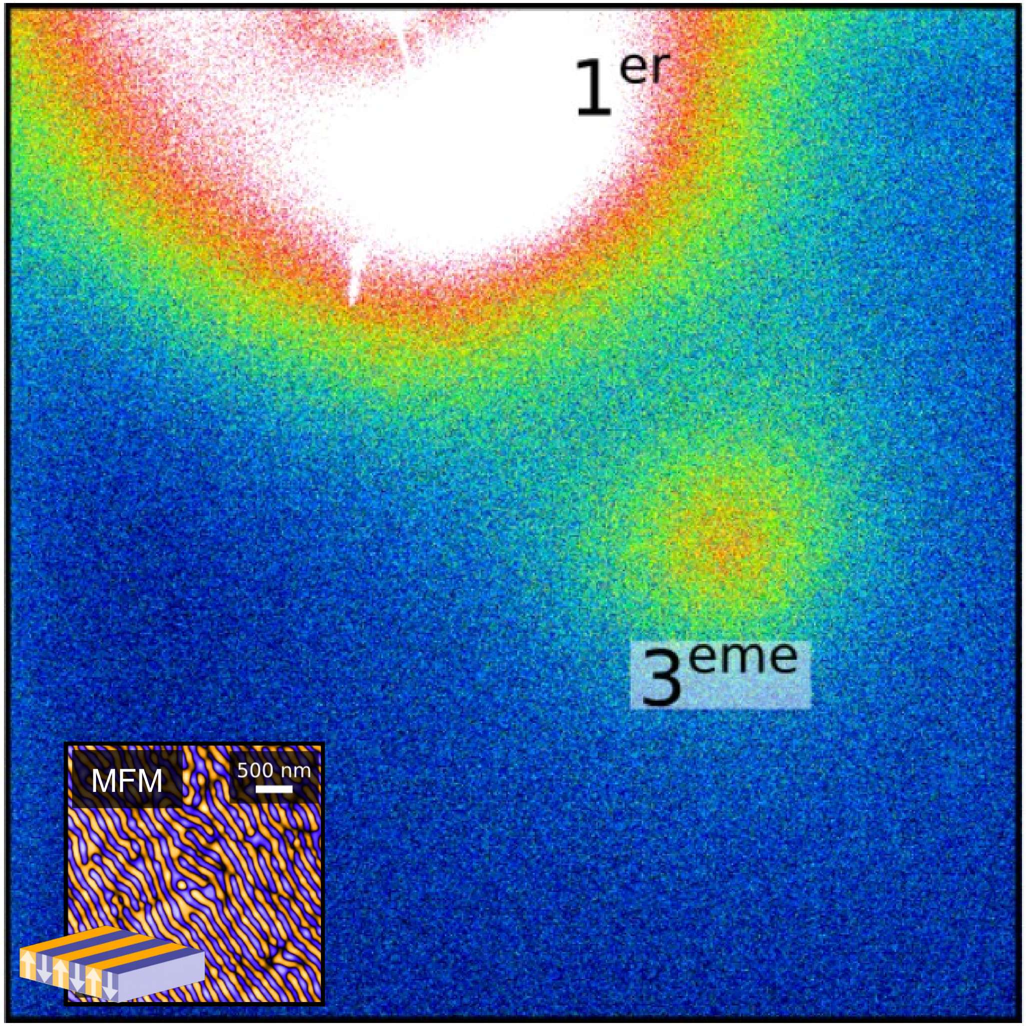
Higher scattering orders → more detailed insight in magnetic domain structure



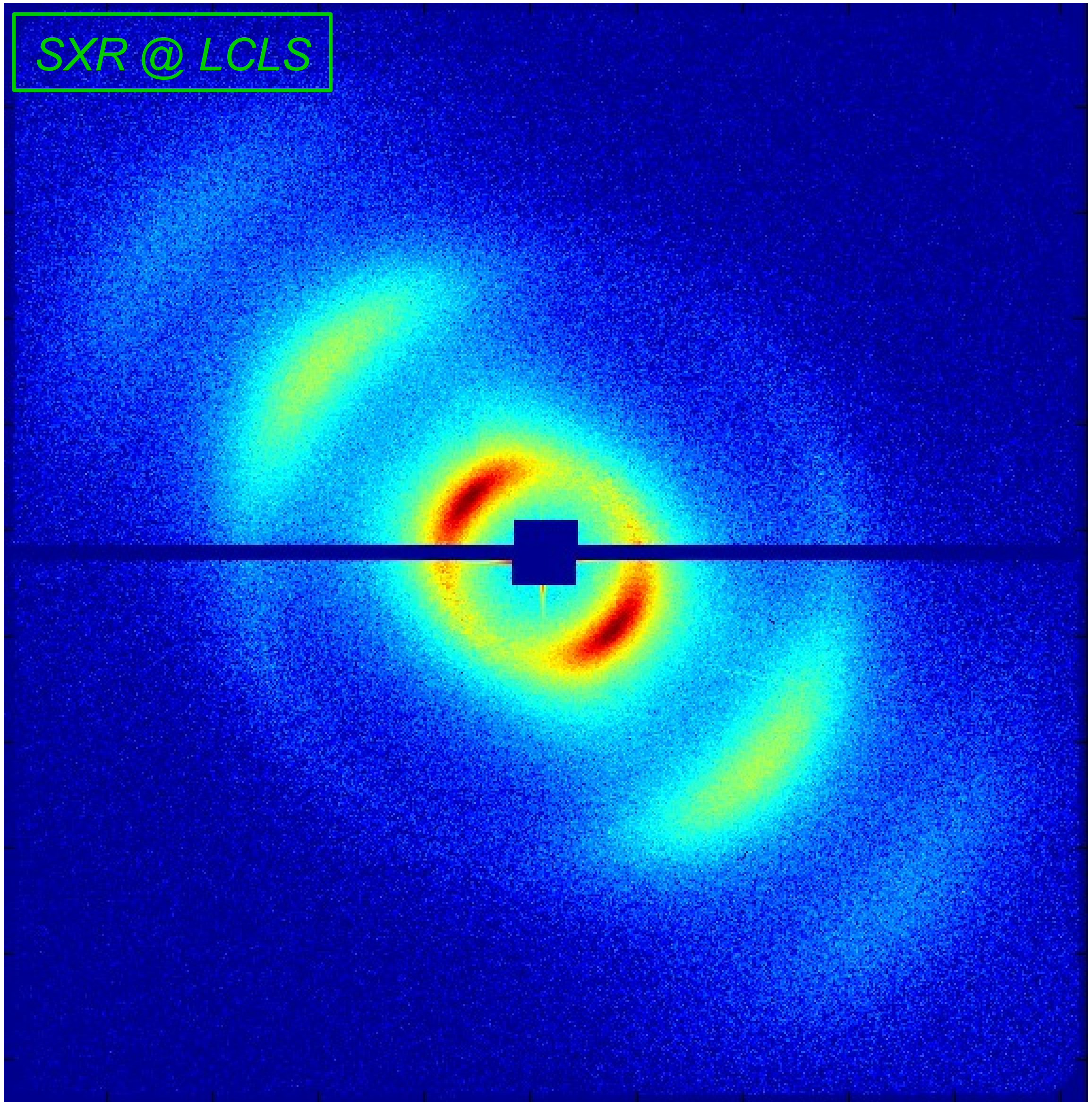
B. Pfau et al.,
Nature Comm. (2012)

O. Hellwig et al, Physica B **336**, 136 (2003)





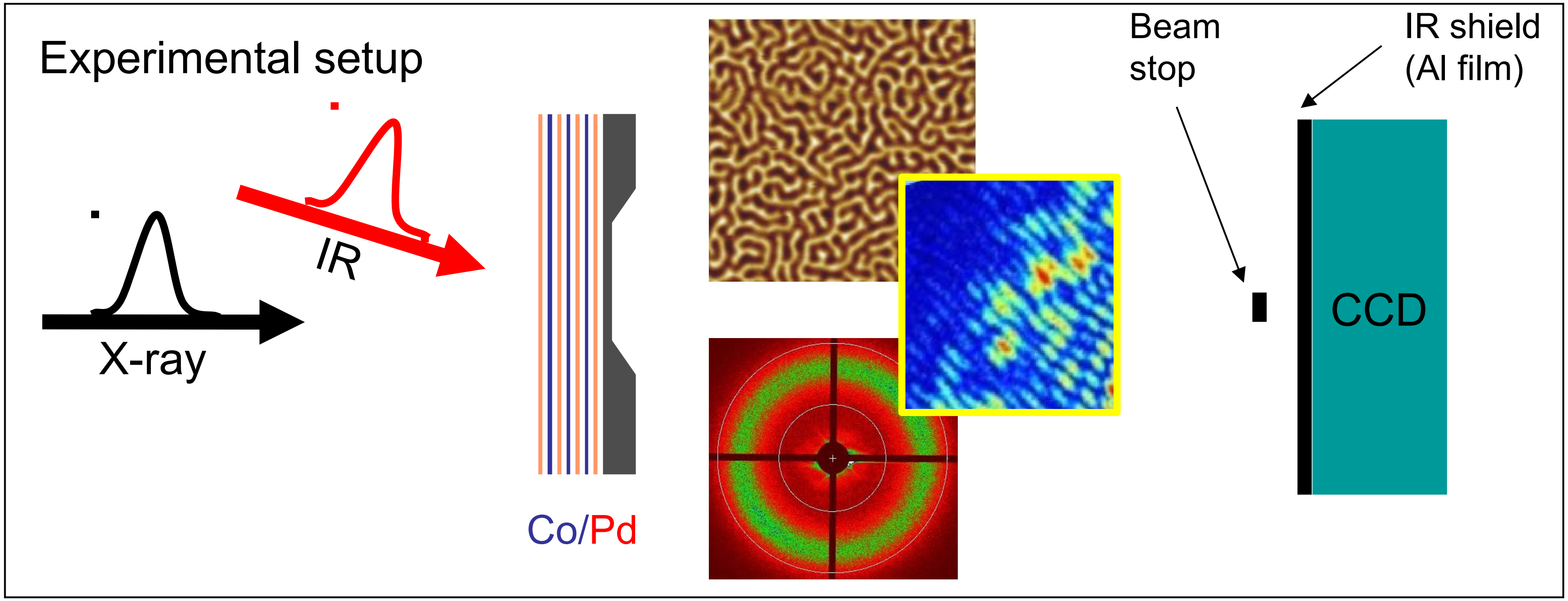
Probing domain wall evolution directly



Resonant scattering for local probing of magnetization

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

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

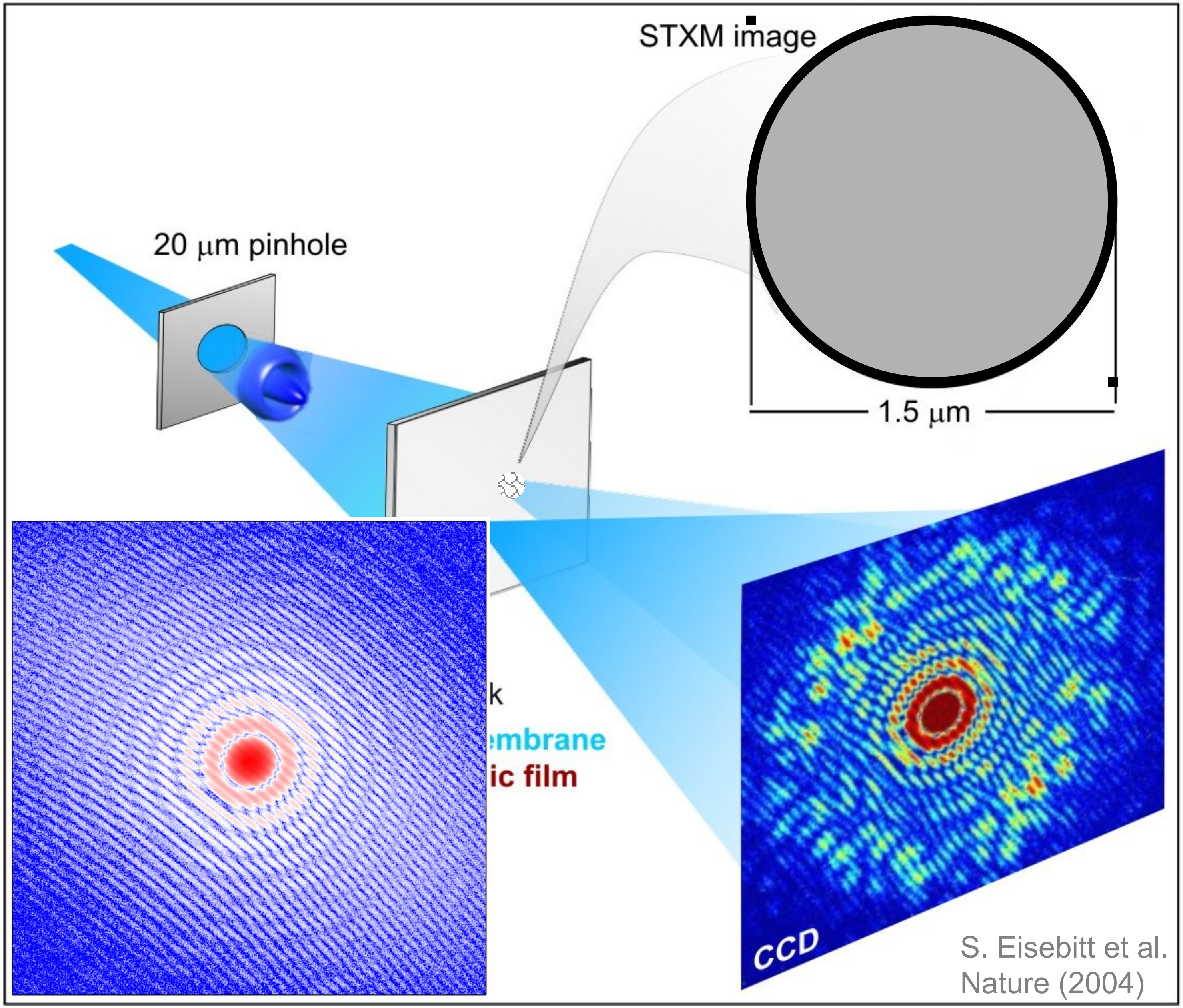


Integrated intensity → measure of the local magnetization

Form of scattering pattern → spatial information

Speckle → imaging

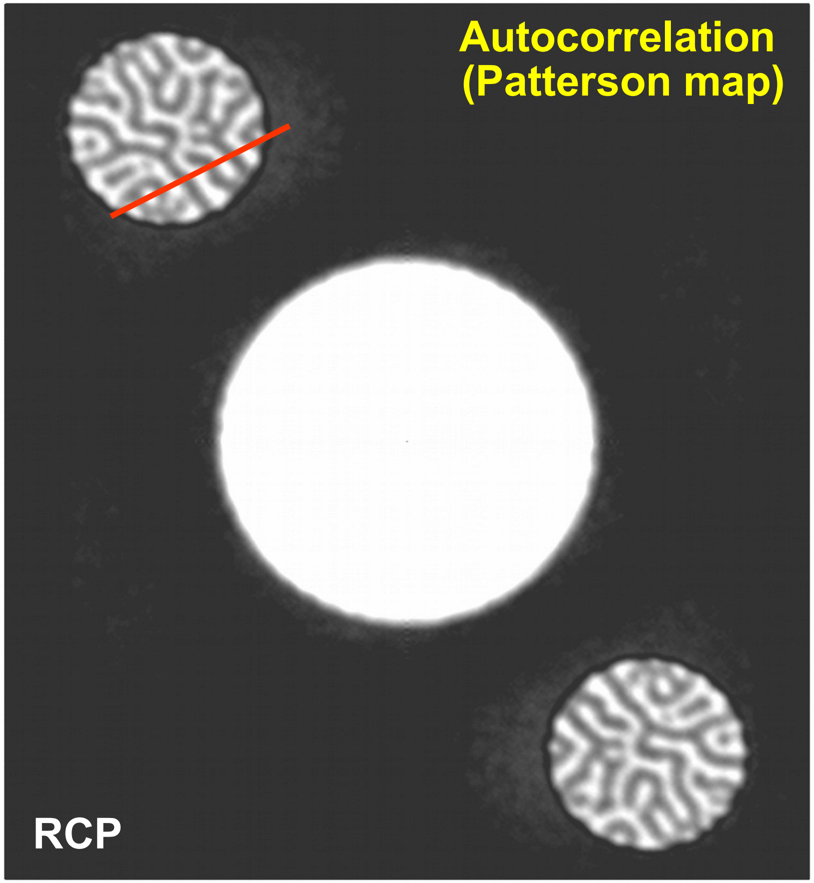
Fourier Transform Holography (FTH)



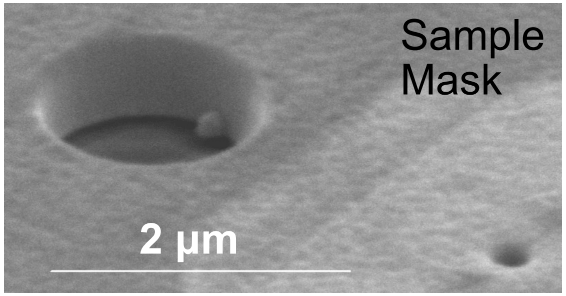
S. Eisebitt et al.
Nature (2004)

Image = FT (scattering intensities)

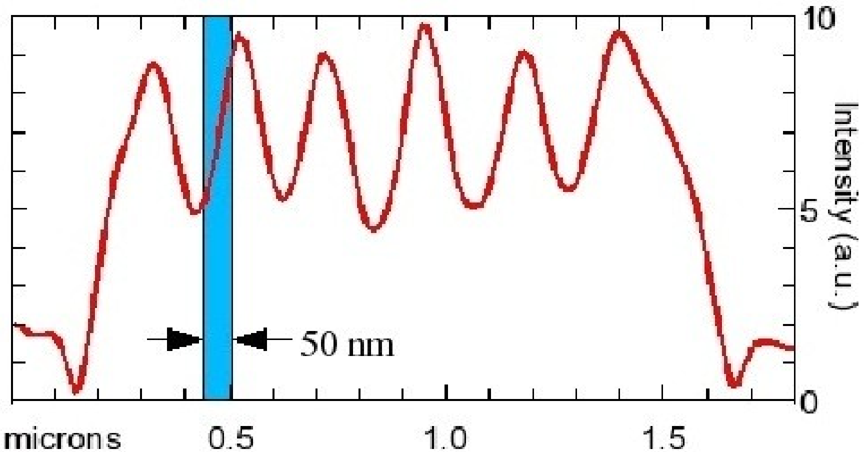
Single Fourier transformation of scattering intensities yields the auto-correlation of sample, which contains image of sample due to the off-axis geometry in FT holography. (convolution theorem)



Intensity in image center, which contains self-correlation of apertures, is truncated.



10% - 90% intensity rise over about 50 nm



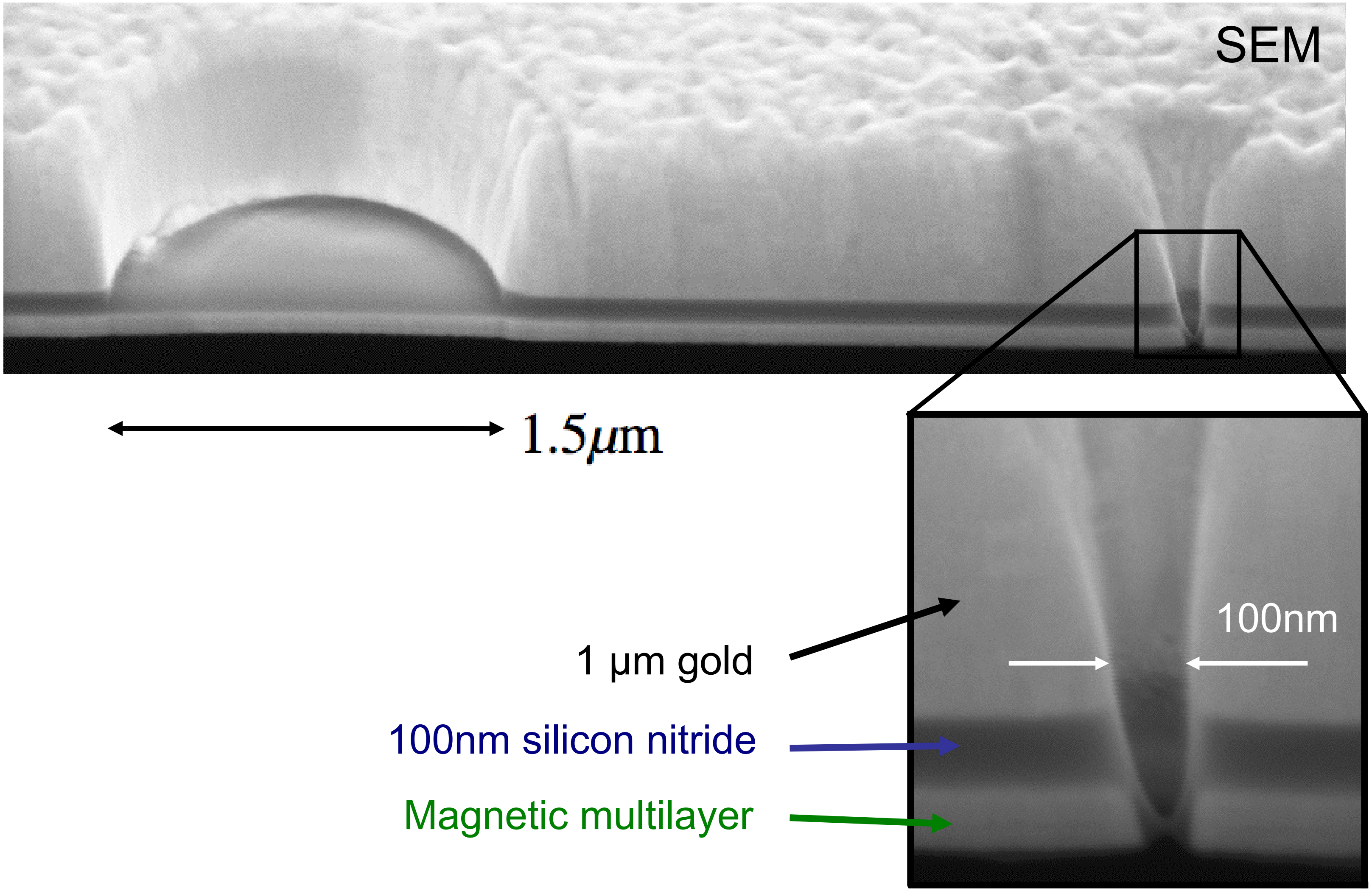
Key properties of Fourier transform X-ray holography

- ***True imaging technique***
- ***Wavelength limited spatial resolution***

  Spatial resolution in FT hologram given by effective size of reference beam source.

Integrated mask sample structure

Patterned with focused ion beam



Key properties of Fourier transform X-ray holography

SCIENCES SORBONNE UNIVERSITÉ

- ***True imaging technique***

- ***Wavelength limited spatial resolution***
  Deconvolution and phase retrieval algorithm

- ***Simple and rather 'cheap' setup***

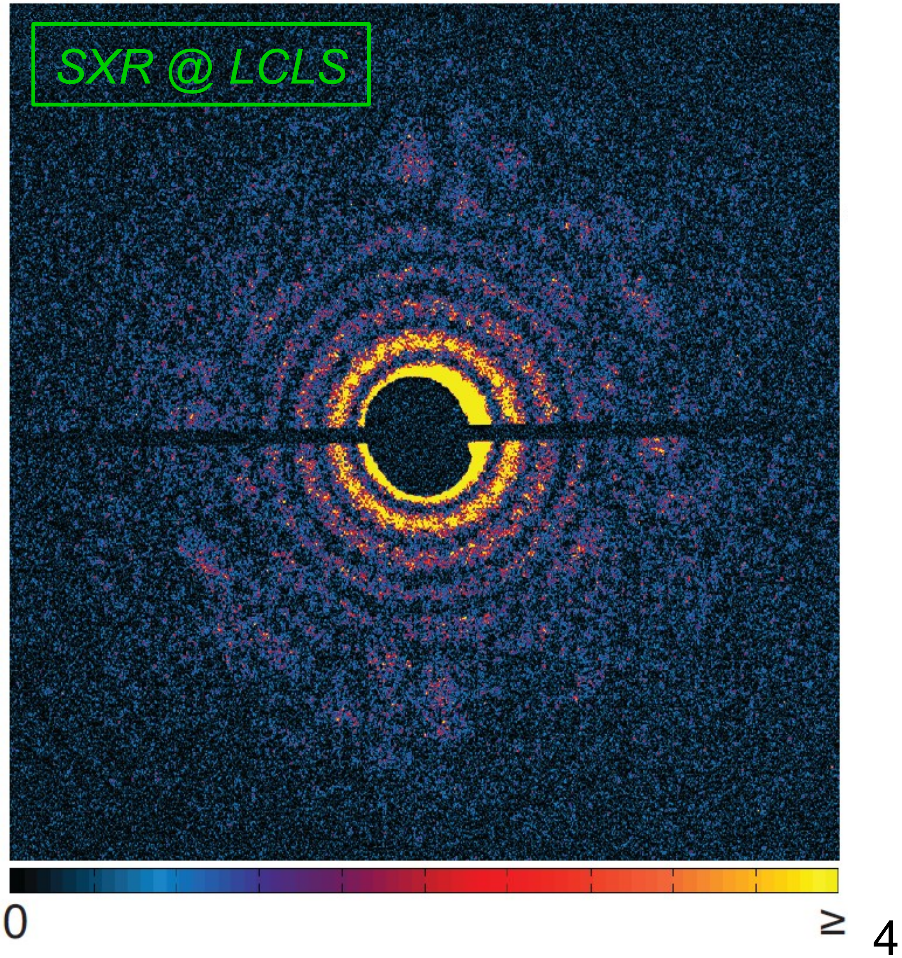
- ***Nanometer resolution with micron stability***
  Setup is basically insensitive to vibrations
  or thermal drifts

- ***Ideally suited for in-situ studies***
  No space constraints around sample

- ***Ideally suited for sample arrays***
  No alignment or focusing required

- ***Wide applicability***
  Samples can be grown or placed in aperture or
  on back of mask or placed separately behind it.
  Reflection geometry may be possible.

Single x-ray pulse based snapshot imaging



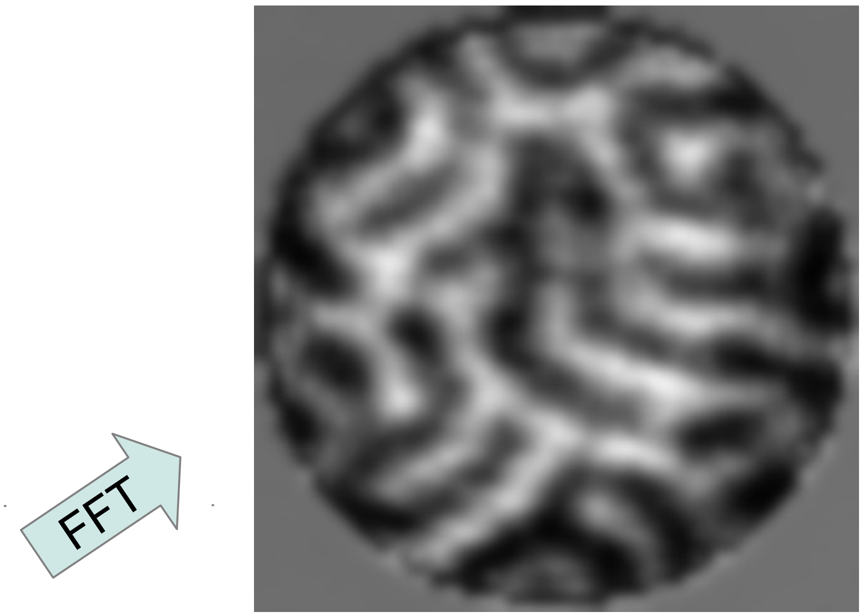


Image of magnetic domain structure obtained from a single X-ray pulse

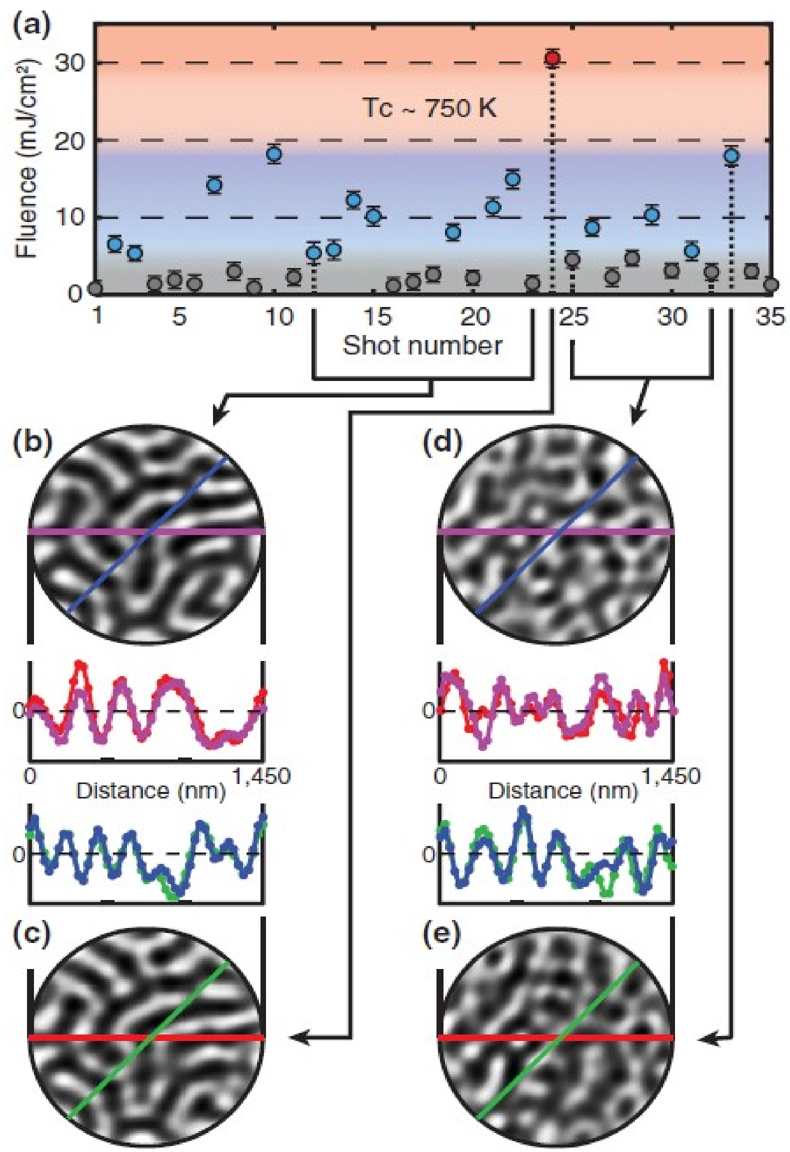
~ 50 nm spatial resolution
~ < 80 fs temporal resolution

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

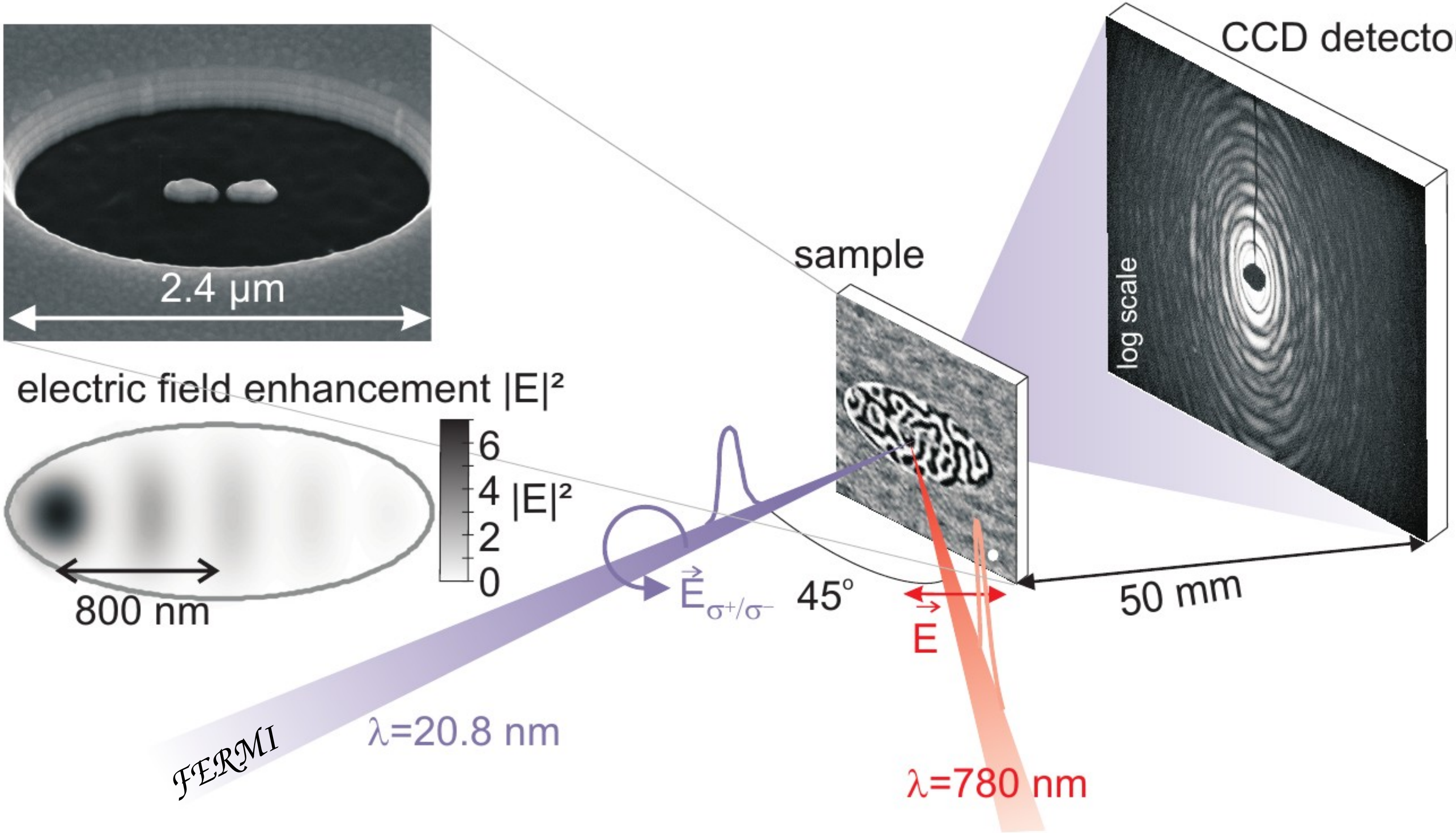
X-ray induced *“modifications”*

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.



after a spatially localized optical excitation



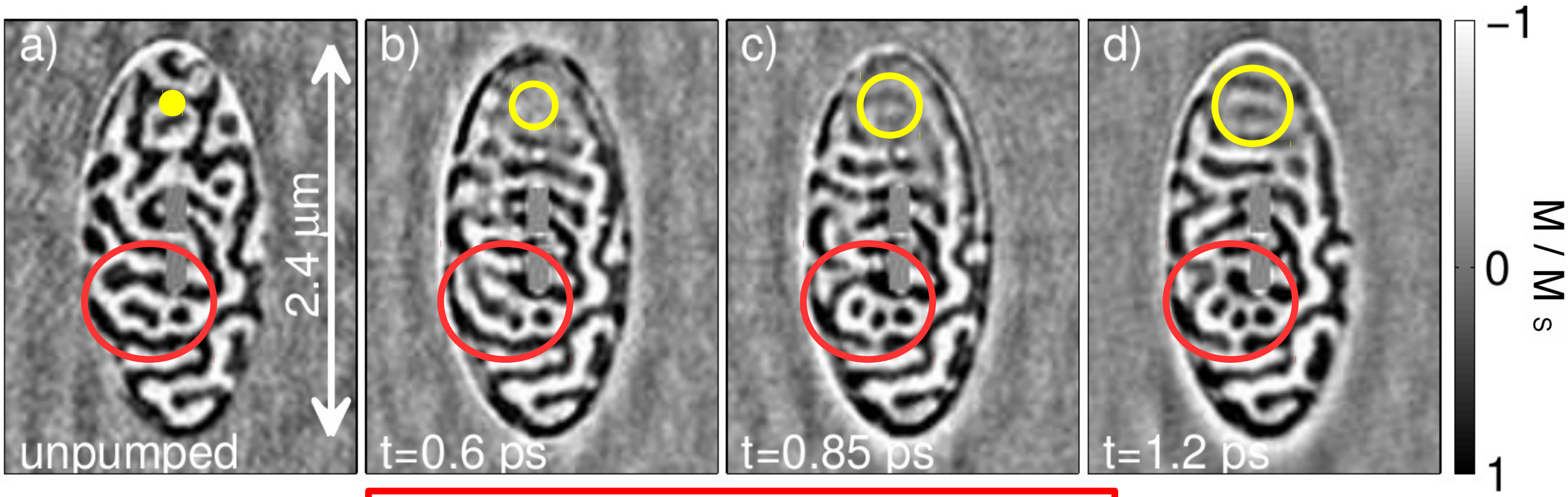
C. von Korff Schmising et al., Phys. Rev. Lett. **112**, 217203 (2014)

Imaging ultrafast demagnetization dynamics

DiProI @ FERMI

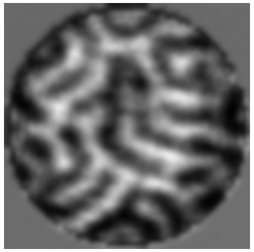
after a spatially localized optical excitation

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



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

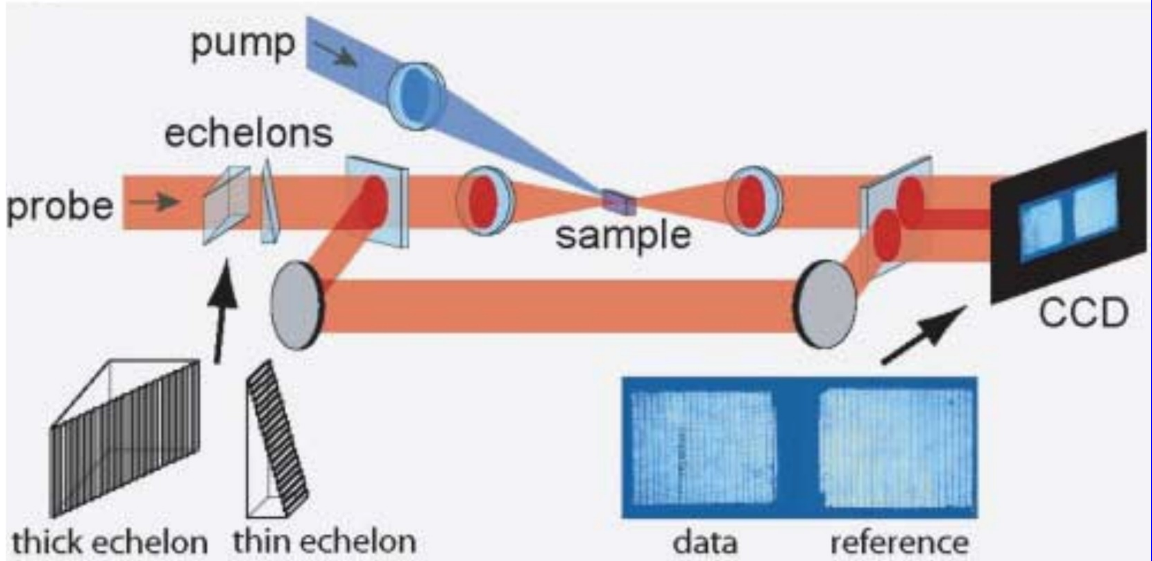
Excellent signal-to-noise due to very high pulse intensity, even for single pulse (snapshot) probing



Can we probe with a single X-ray pulse more than one point in time?

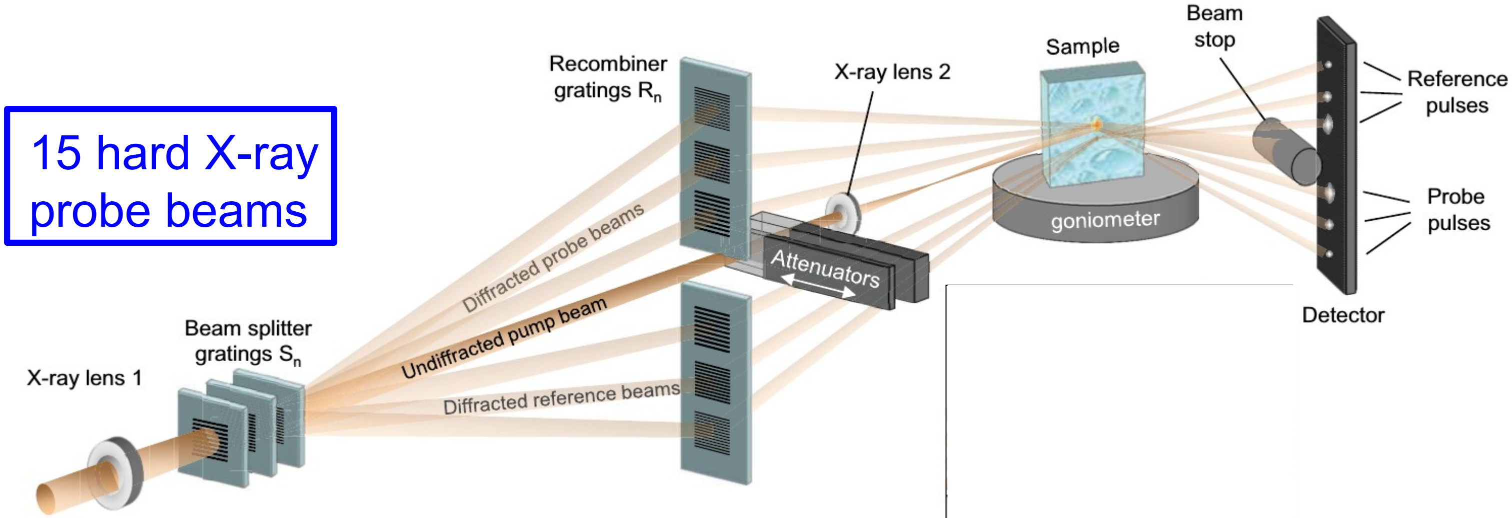
Sampling several pump-probe delays at once

400 optical probe beams



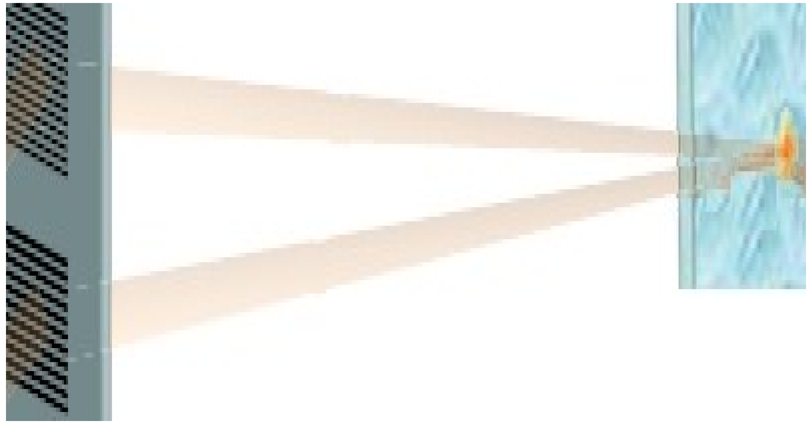
P.R. Poulin & K.A. Nelson, Science 313, 1756 (2006).

15 hard X-ray probe beams

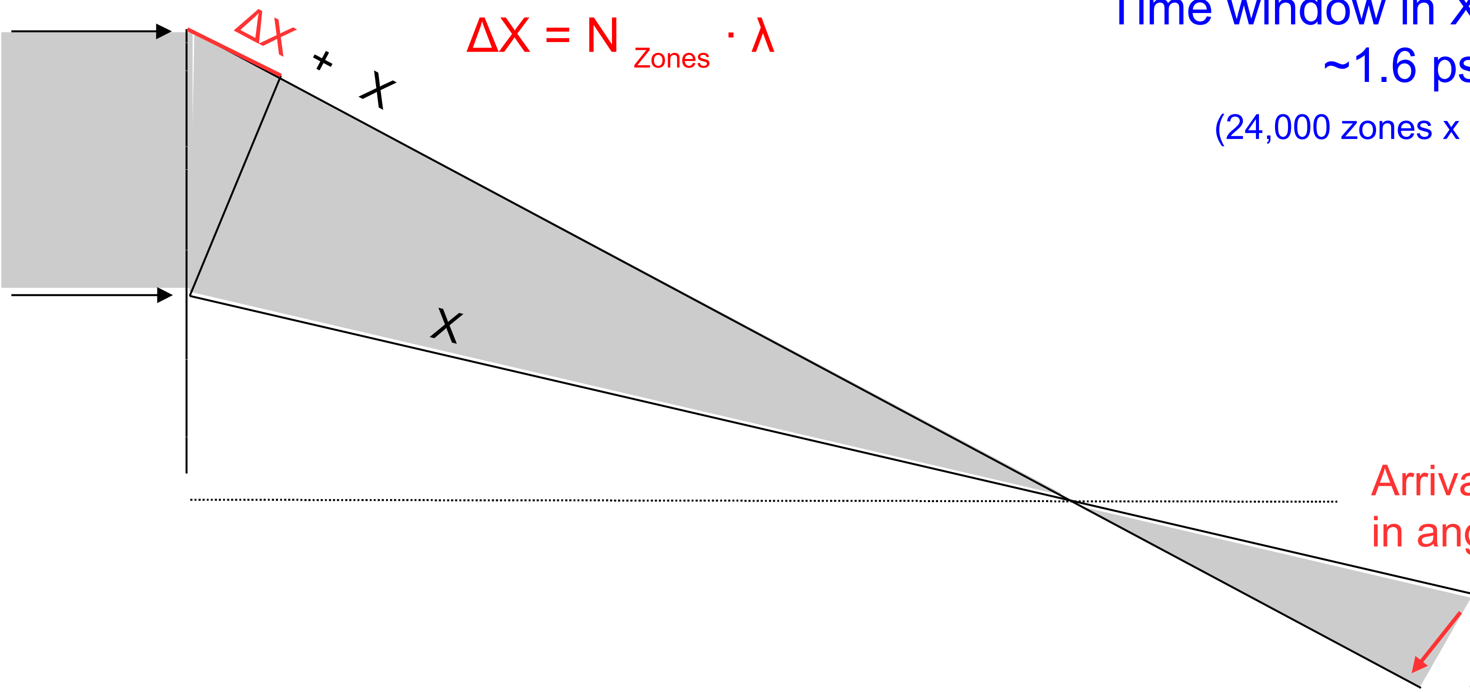


C. David et al., Scientific Reports 5, 7644 (2015)

X-ray streaking to follow dynamics with fs precision



Basic idea:

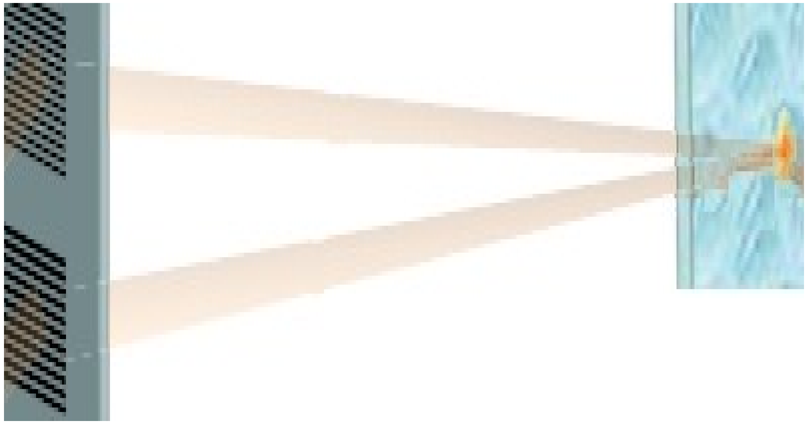


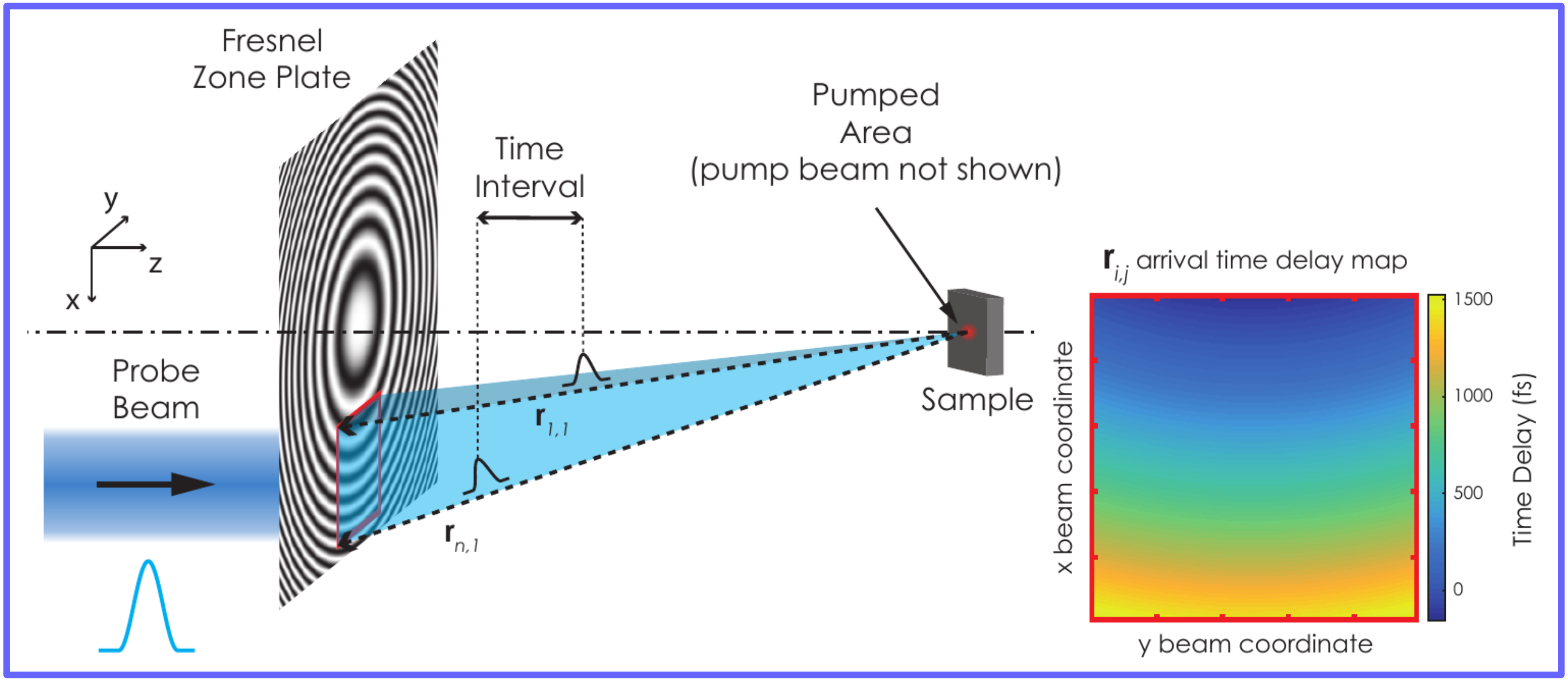
$\Delta X = N_{Zones} \cdot \lambda$

Time window in XUV range
~1.6 ps
(24,000 zones x 20 nm)

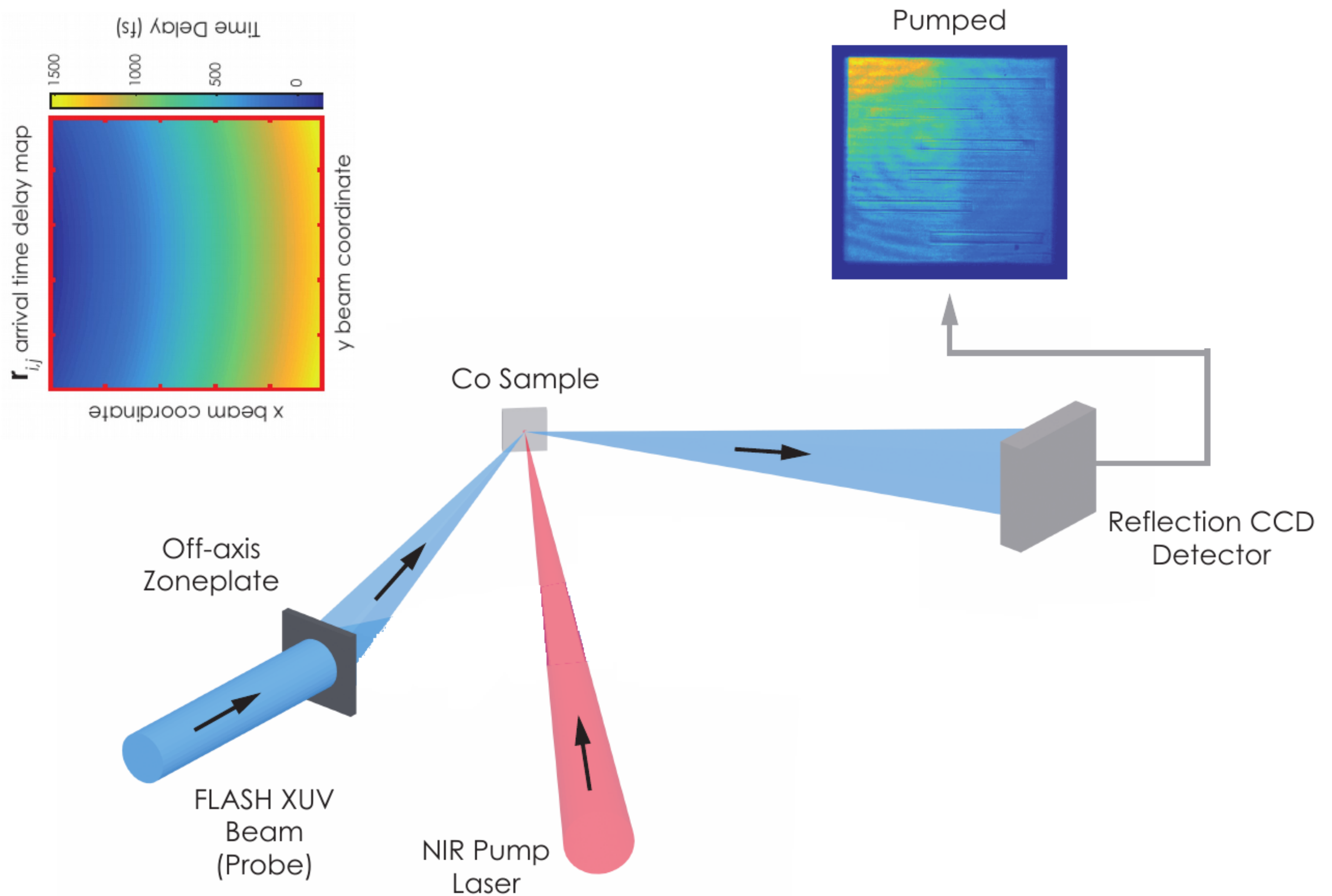
Arrival time encoded
in angular direction

Snapshot recording of ultrafast dynamics

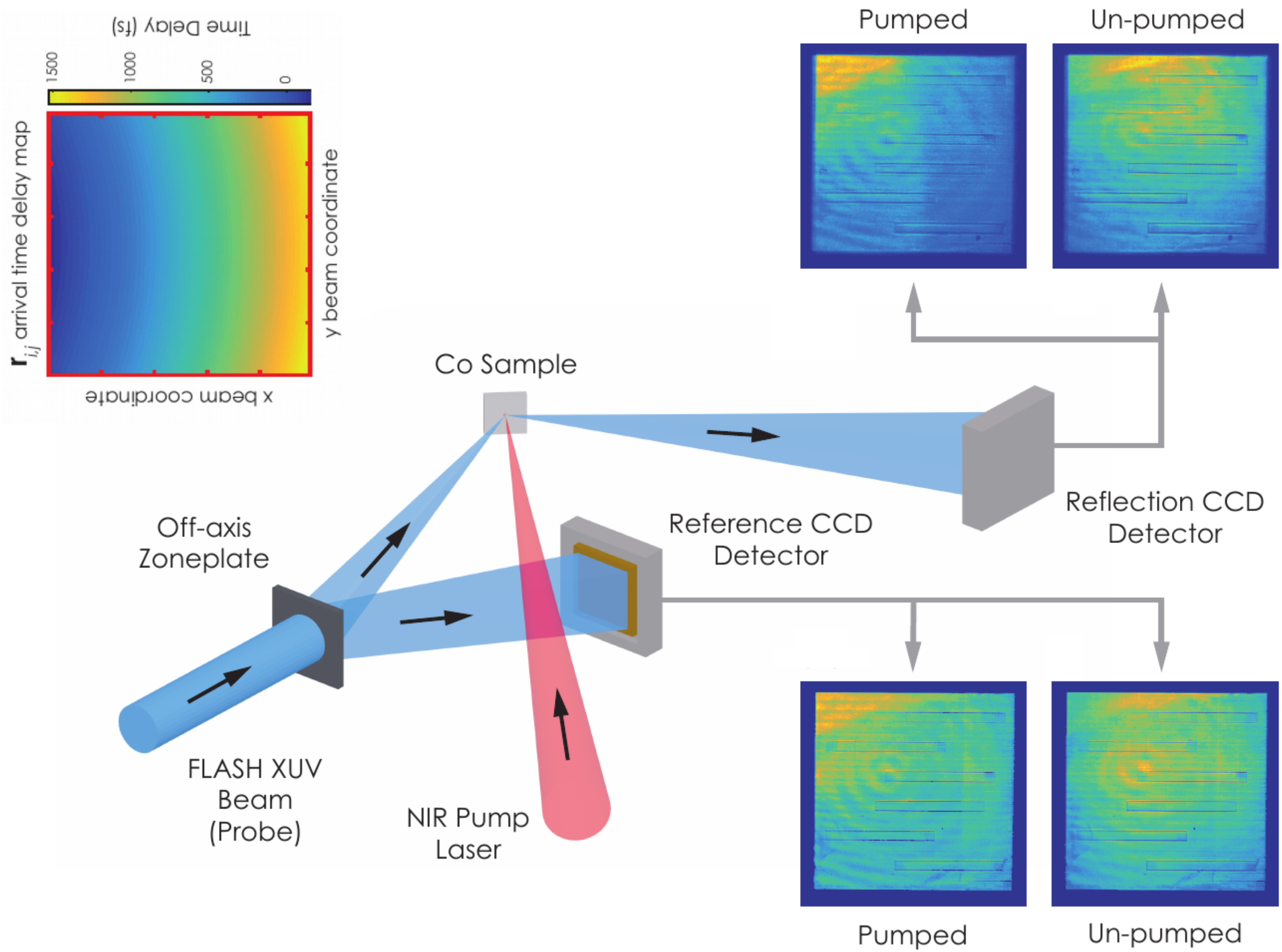




Snapshot streaking of ultrafast demagnetization dynamics

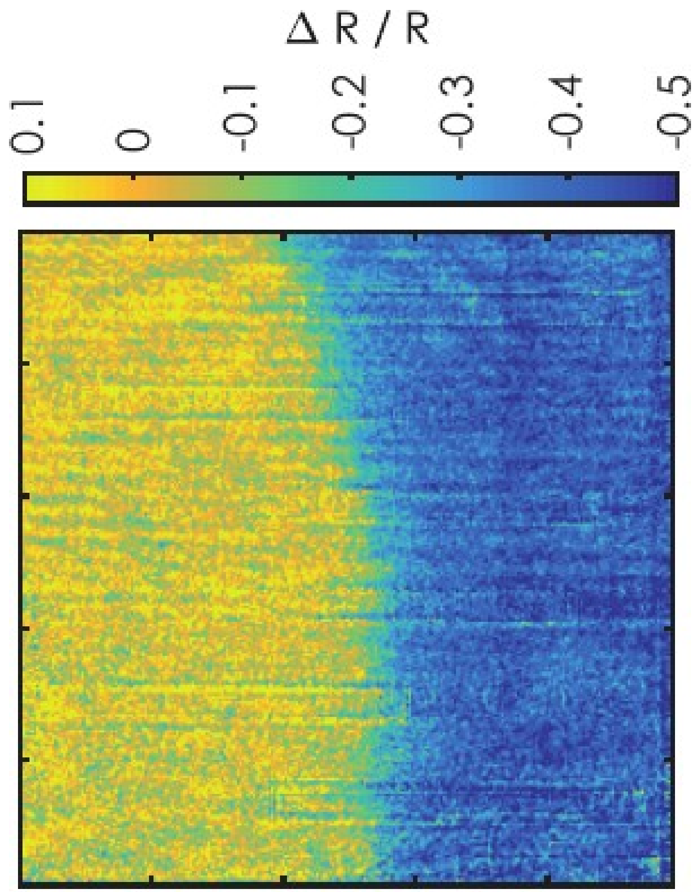


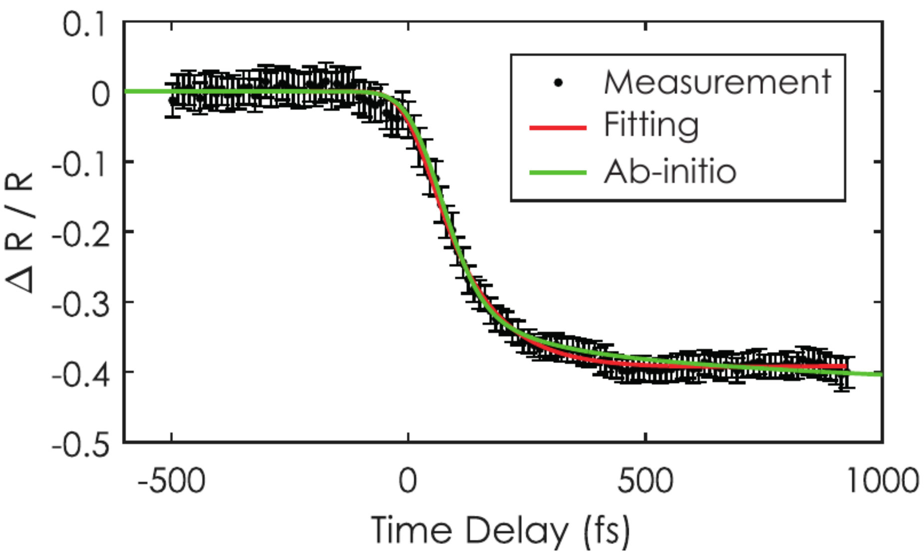
Snapshot streaking of ultrafast demagnetization dynamics

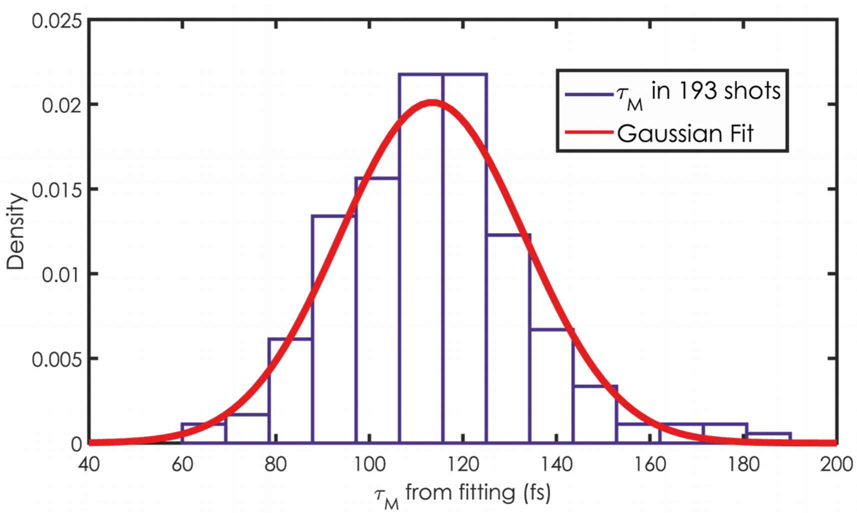


Snapshot streaking of ultrafast demagnetization dynamics

BL3 @ FLASH







$\Rightarrow \tau_M = 113$ fs ± 20 fs

Summary

- Magnetism after thousands of years still at the center of intense research!

- 1996: Discovery of ultrafast magnetization dynamics → femtomagnetism
  2007: Discovery of all-optical magnetization control
  → potential to *disruptively change magnetic data storage technology*.

- *Femtosecond time resolved X-ray techniques provide unique capabilities* for the investigation of ultrafast charge, spin and lattice dynamics.

- Different sources for fs short X-ray pulses with complementary properties:
  – Femtoslicing: - Simplicity to bridge different time scales;
    - SR environment facilitates sample prep/charact.
  – HHG: - jitter-free, simultaneous multi-color probing;
    - potential for very high time resolution;
    - variable pump wavelength 'natural' to implement;
    - several other secondary sources like betatron.
  – XFEL: - snapshot probing using individual X-ray pulses;
    - routine measurements become possible.

- Ultrafast magnetization dynamics:
  – Transport phenomena play a role;
  – Angular momentum transport seems to be not the dominant mechanism.

Thank you for your attention