# Magnetization dynamics revealed by time resolved X-ray techniques

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**Lecture topics:**

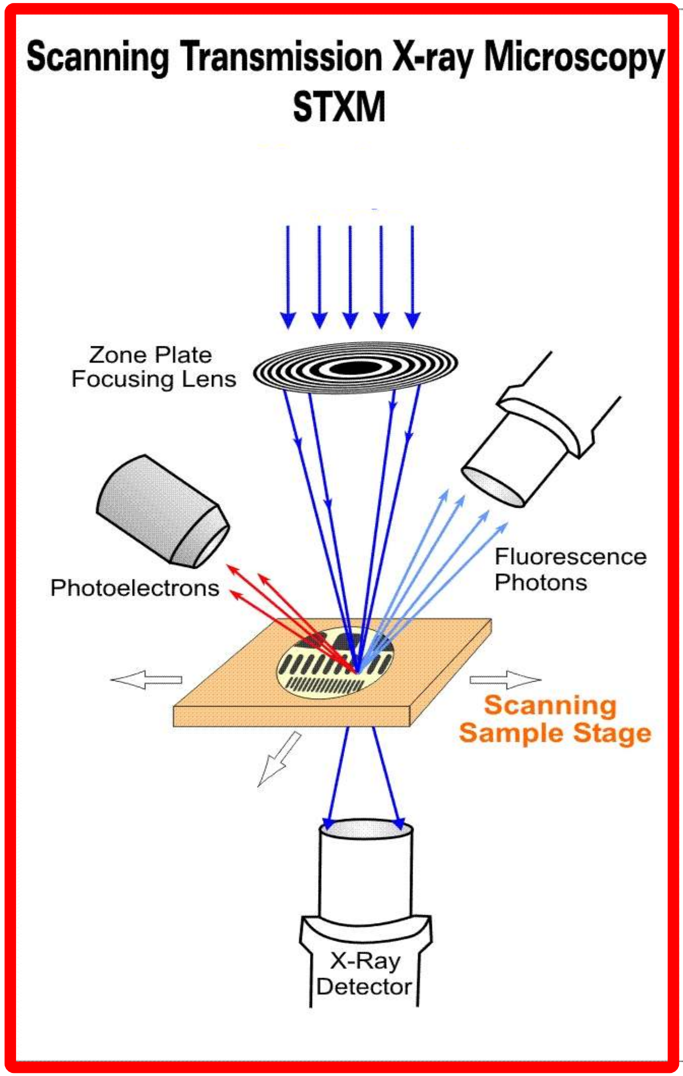
1) X-ray sources and their time structure

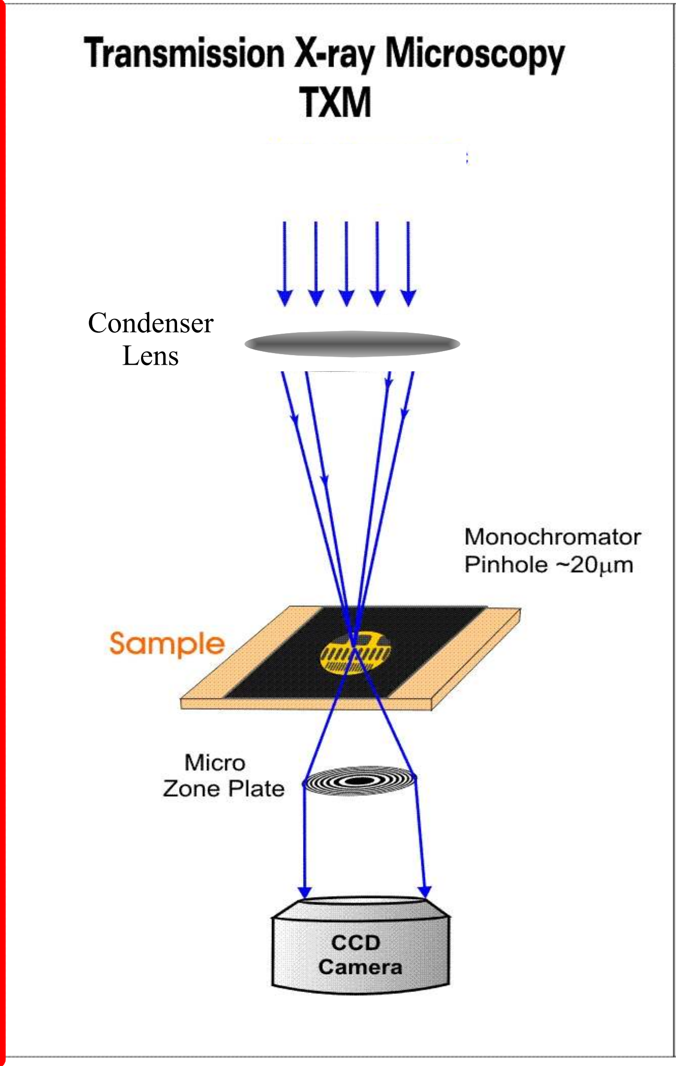
2) Collective magnetization dynamics

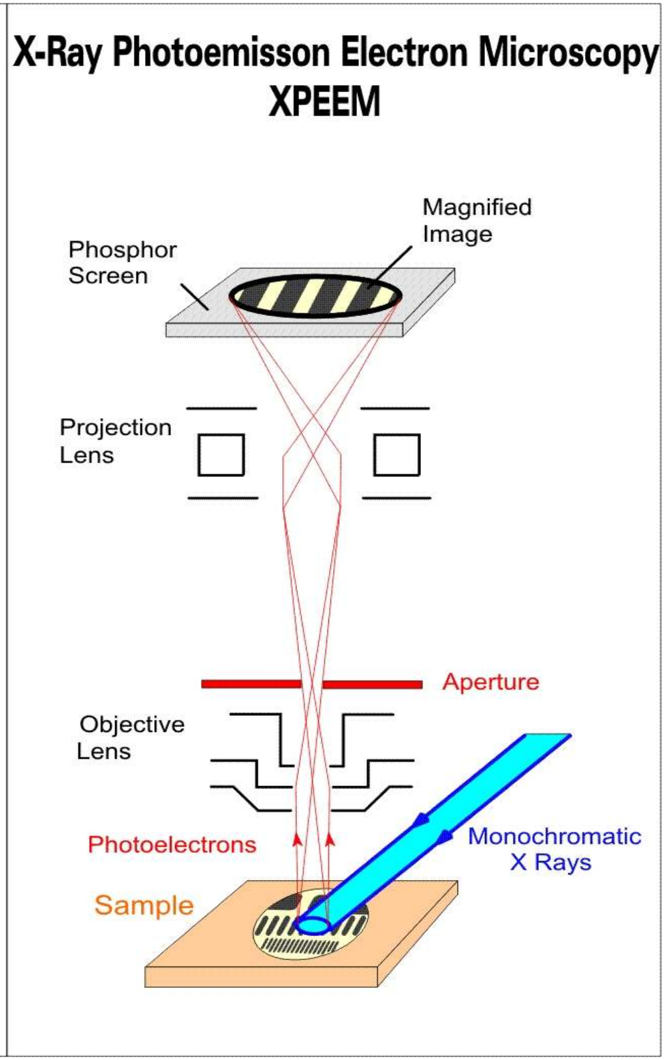
3) Ultrafast magnetization dynamics

# X-ray spectromicroscopy techniques

Quantitative imaging with sensitivity to
elemental and chemical distribution and charge/spin ordering

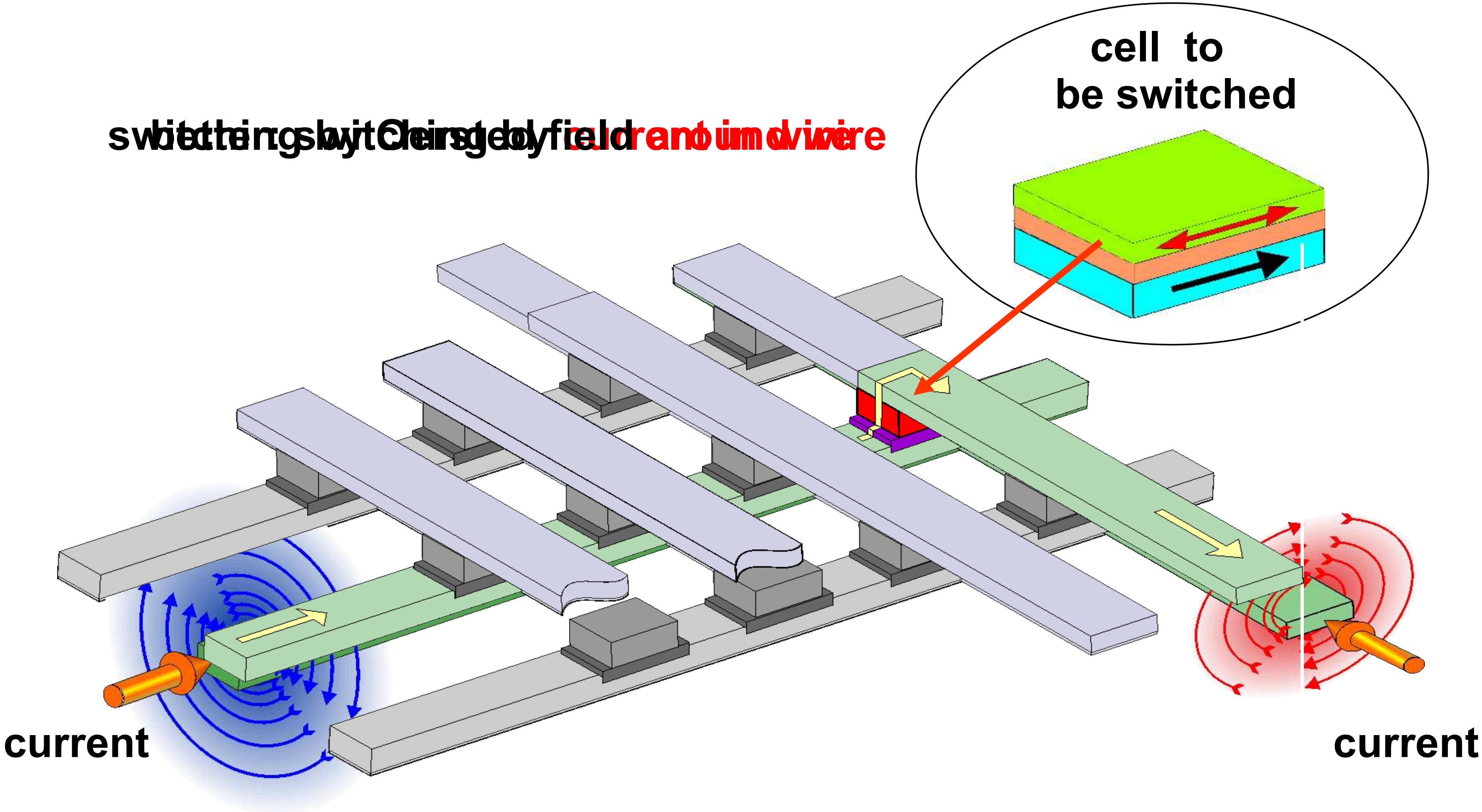


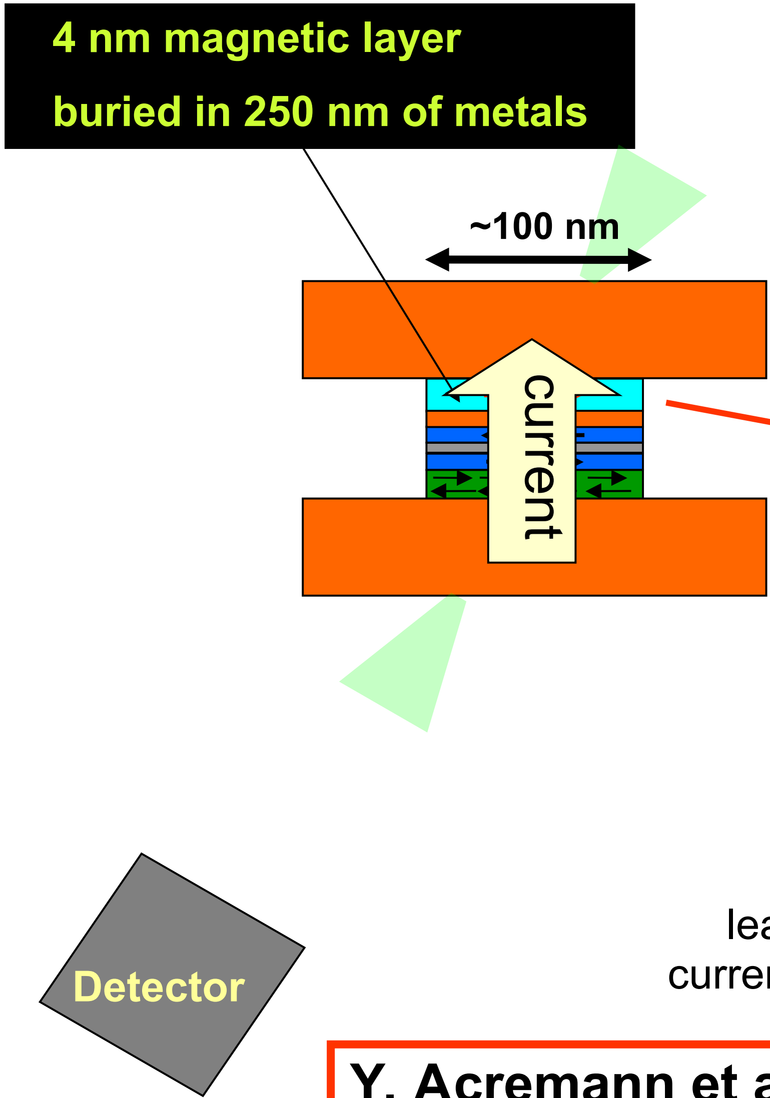



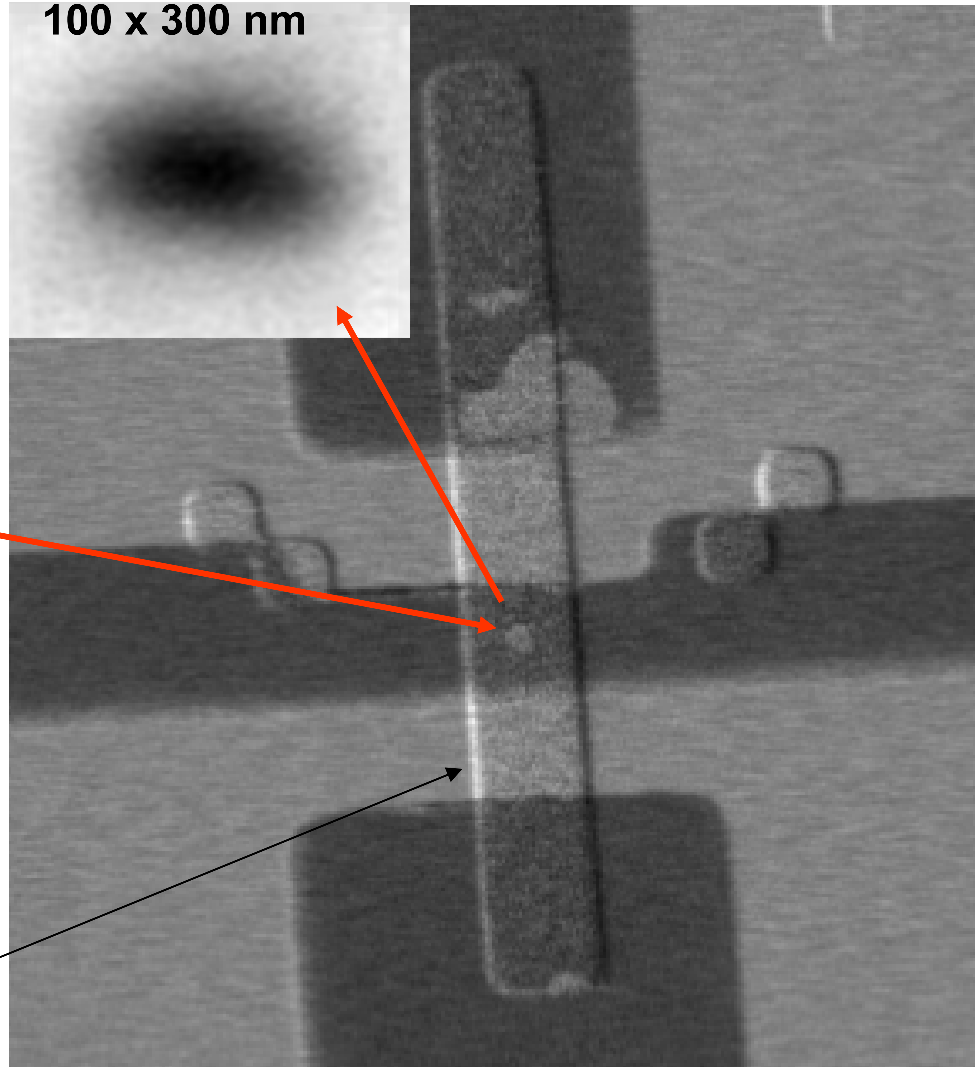
# Motivation: Switching of magnetic memory cells (MRAM)

THE EUROPEAN SCHOOL ON MAGNETISM



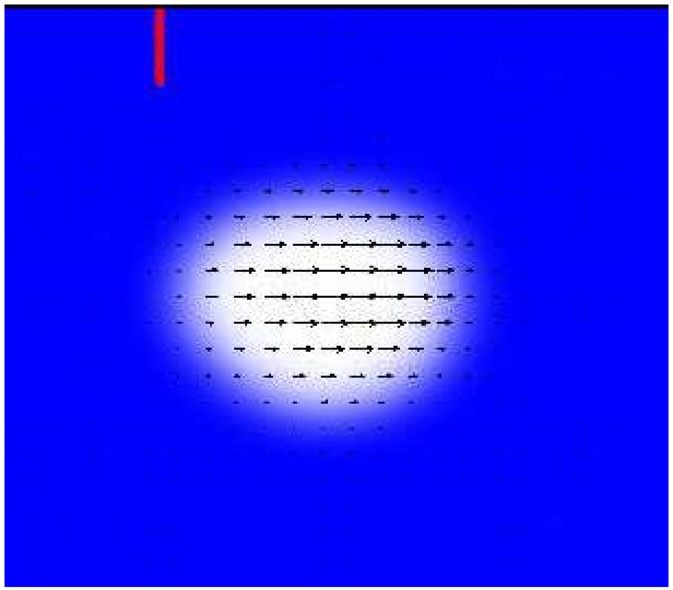
# STXM image of spin injection structure

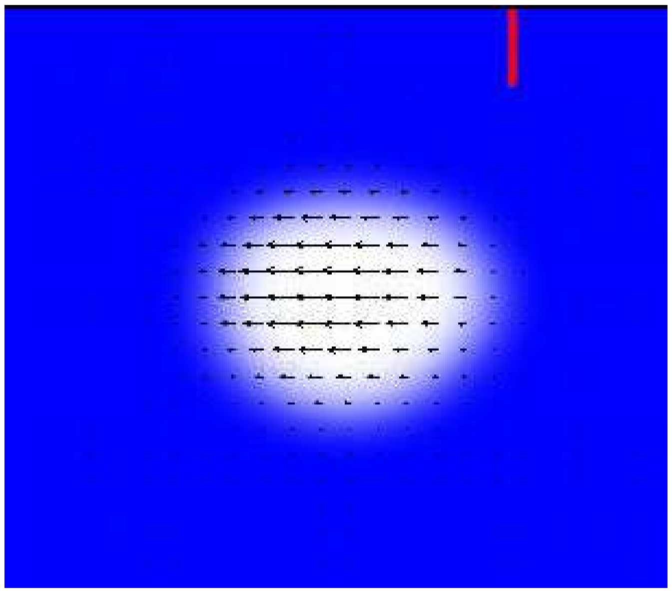



Y. Acremann et al., Phys. Rev. Lett. 96, 217202 (2006)

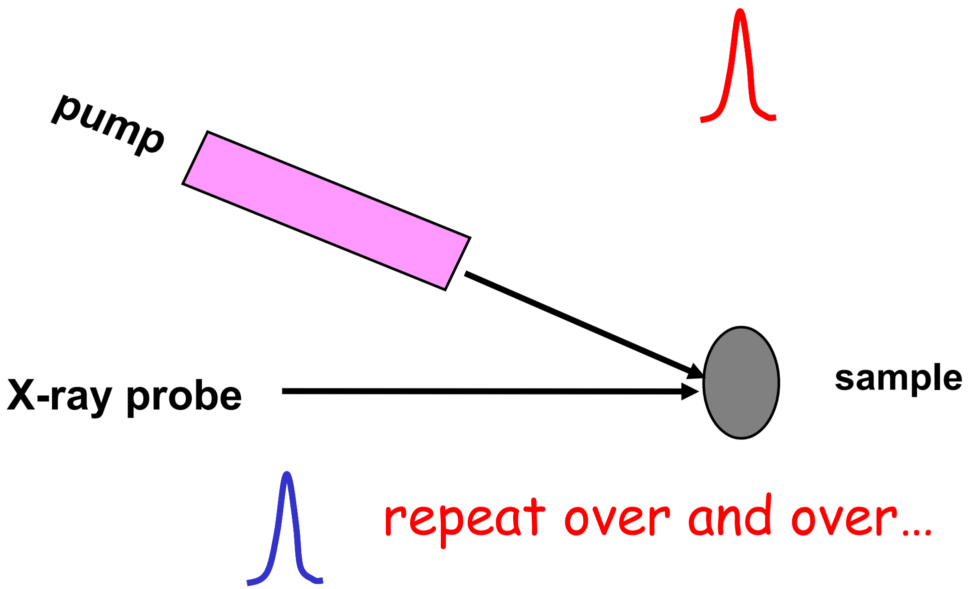
# Static images of the burried layer’s magnetization





# Studying dynamics by pump – probe cycles

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

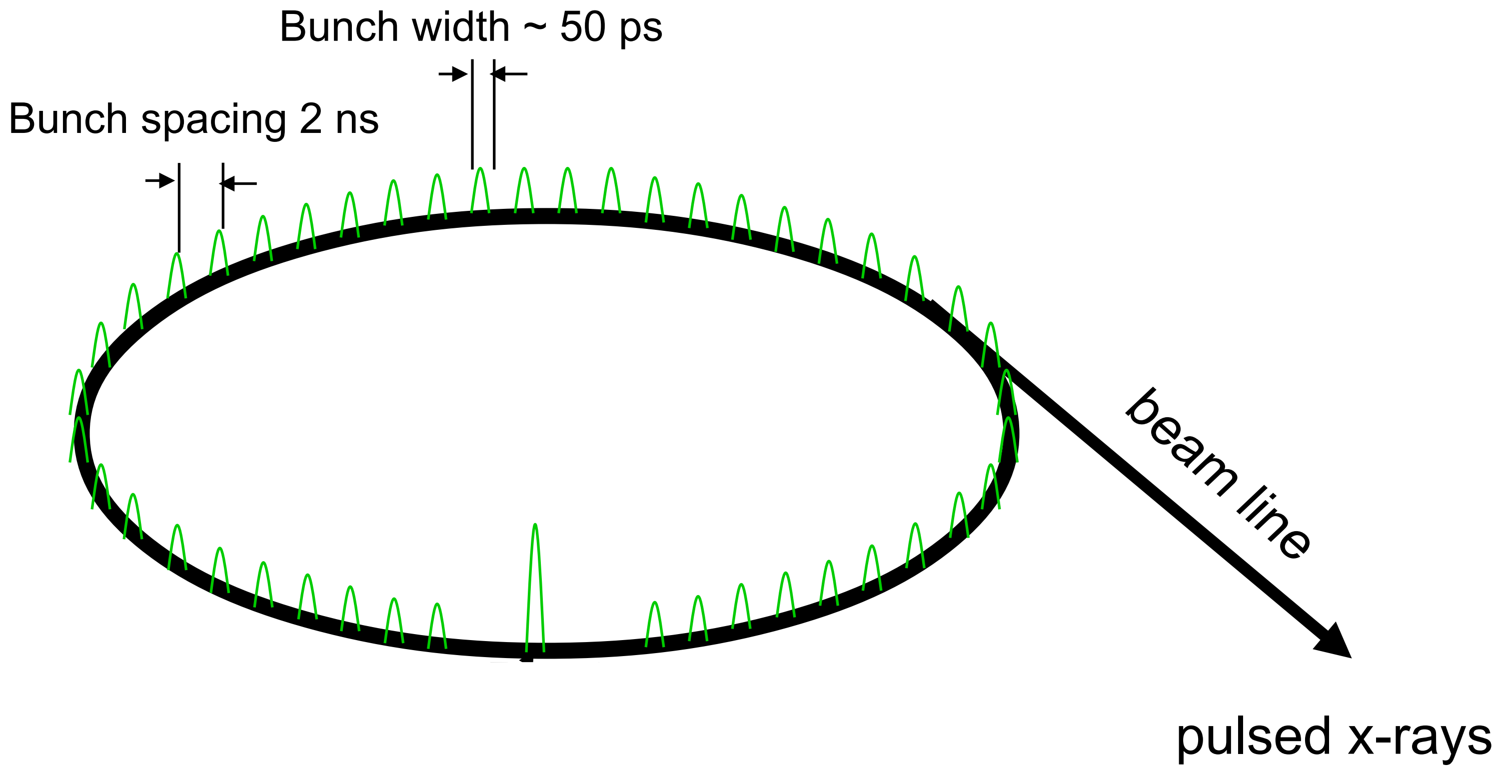



Limitation:

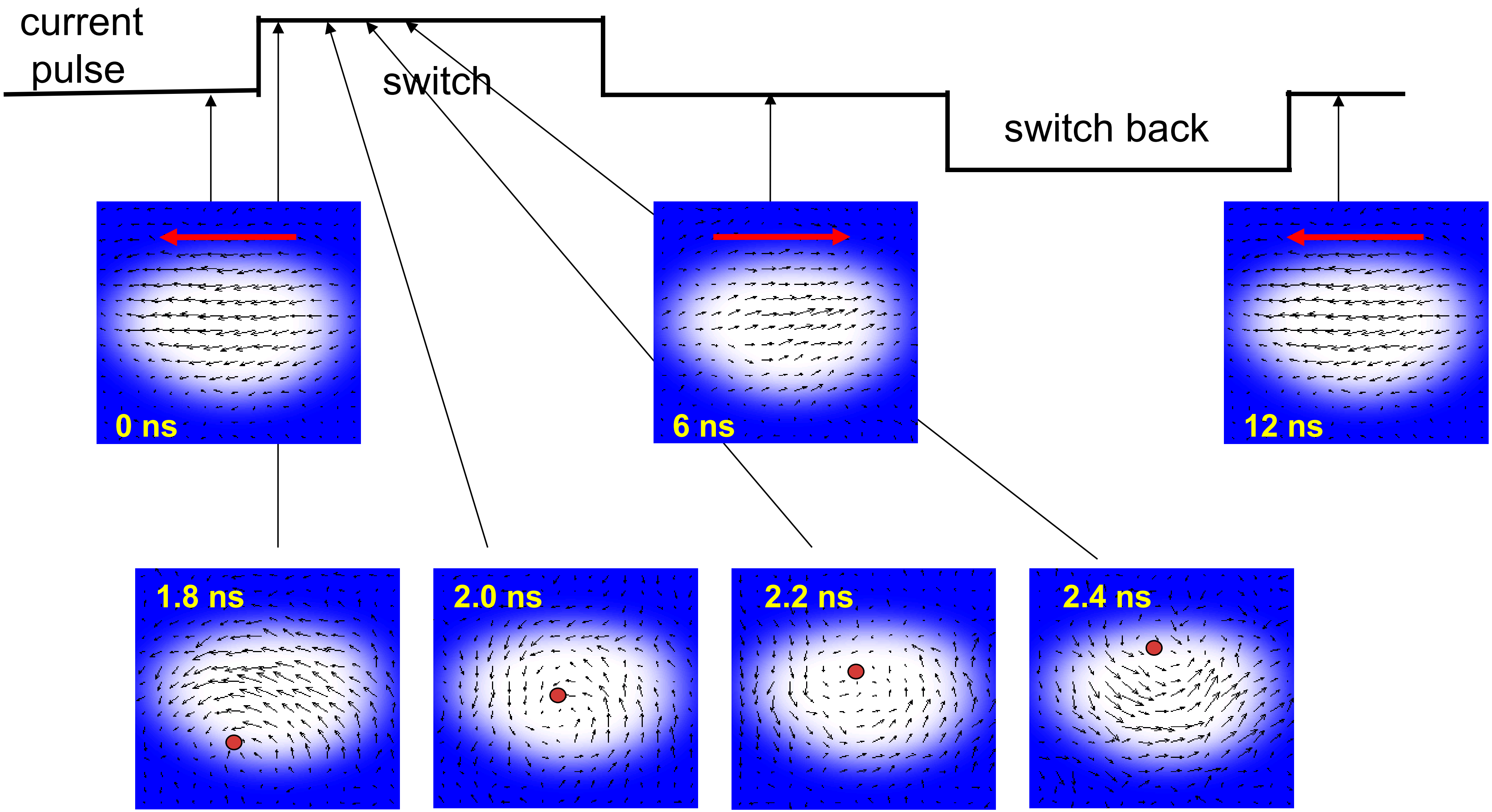
Process has to be repeatable

# Pulse structure of synchrotron radiation

Storage ring is filled with electron bunches → emission of X-ray pulses



# Magnetization reversal dynamics by spin injection




**Switching best described by movement of vortex across the sample!**

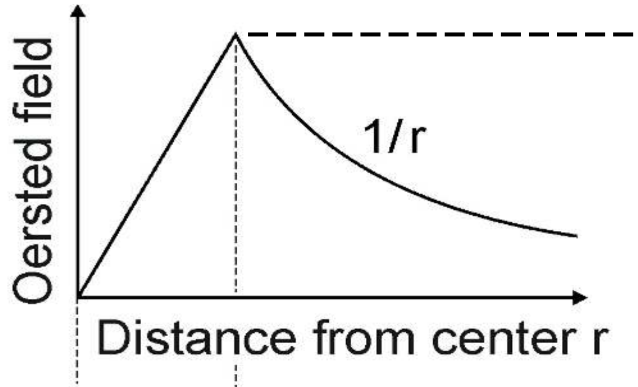
# Magnetic switching by interplay of charge and spin current

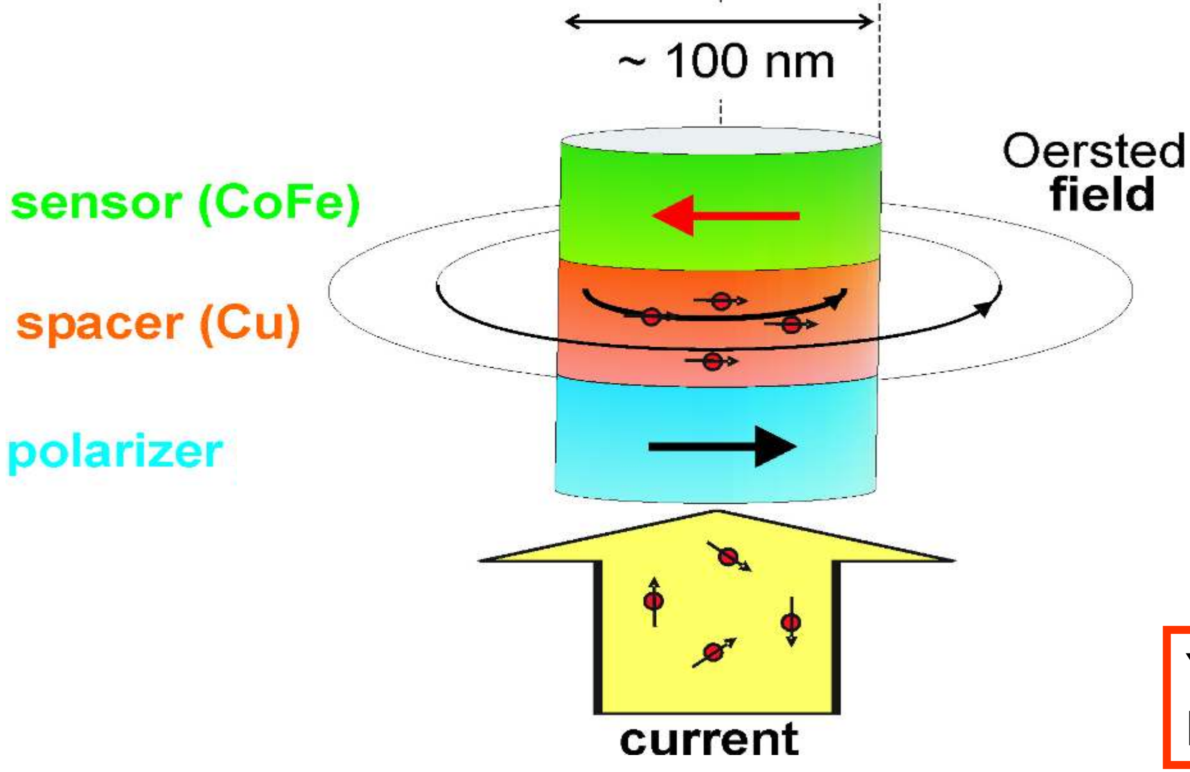
**CHARGE CURRENT:**

creates vortex state

**SPIN CURRENT:**

drives vortex across sample

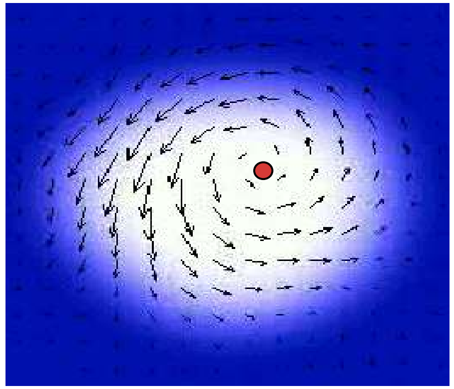



**= 950 Oersted**

**for 150x100nm,**

**$j = 2x10^8$ A/cm$^2$**



**Y. Acremann et al.,**
**Phys. Rev. Lett. 96, 217202 (2006)**

# Soft x-ray spectro-microscopy at its best

- **Sensitivity to buried thin layer (4 nm)**

  **Cross section just right - can see signal from thin layer X-rays can distinguish layers, tune energy to Fe, Co, Ni or Cu L edges**

- **Resolving nanoscale details (< 100 nm)**

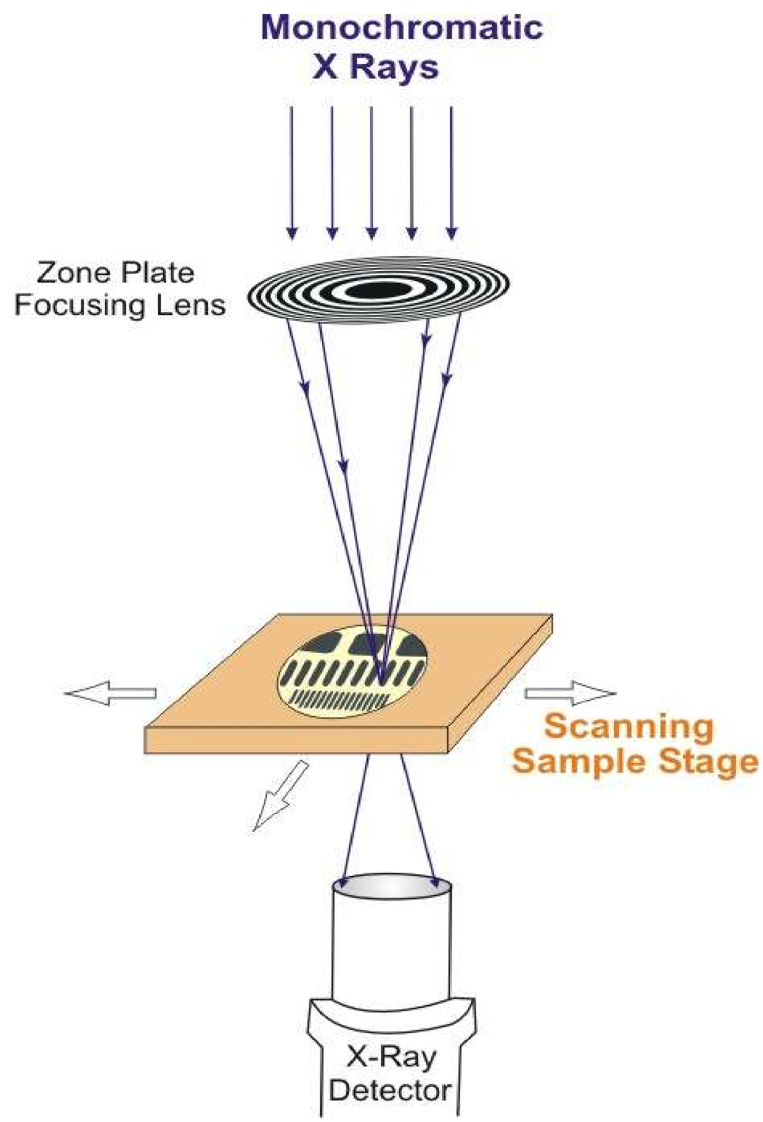
  **Spatial resolution, x-ray spot size ~30 nm**

- **Magnetic contrast**

  **Polarized x-rays provide magnetic contrast (XMCD)**

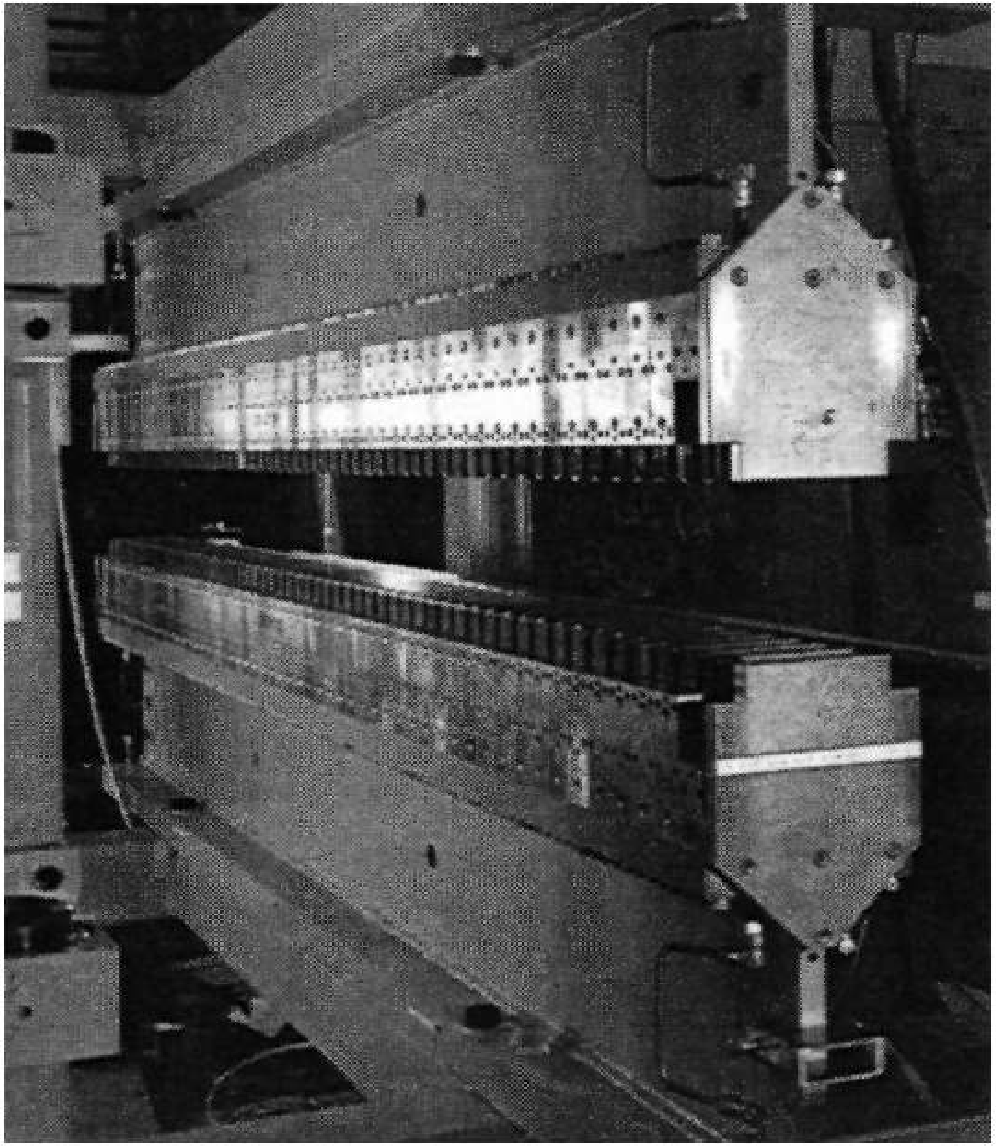
- **Sub-nanosecond timing**

  **Synchronize spin current pulses with ~50 ps x-ray pulses**




Fast detector for X-ray pulse selection

# Synchrotron Radiation



LBNL/EXXON/SSRL (1982), SSRL Beamline VI
55 pole (N = 27.5), $\lambda_w$ = 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·mrad$^2$·mm$^2$)
  → 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

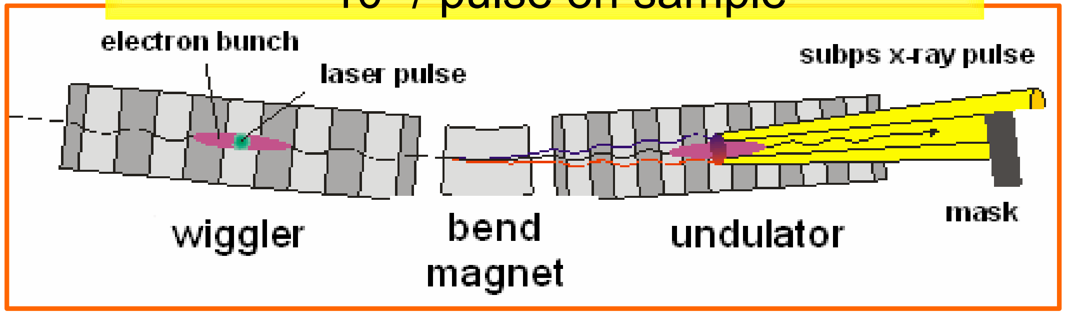
  → inadequate for fs dynamics

# fs pulsed X-ray sources

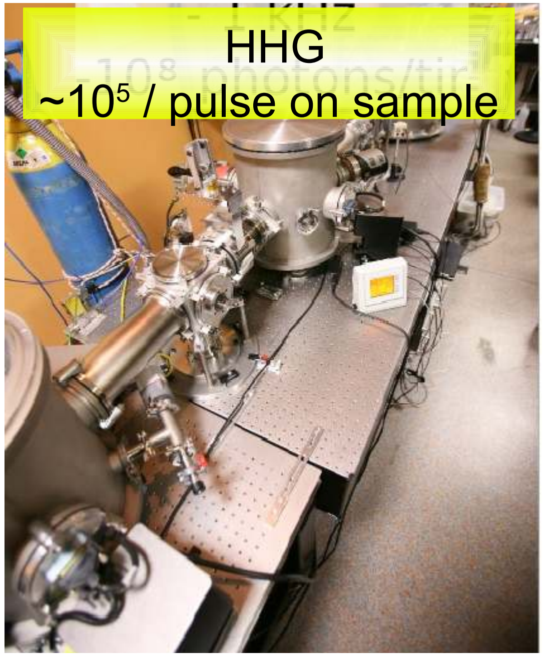


**Combine *nanometer spatial* resolution with *femtosecond temporal* resolution**

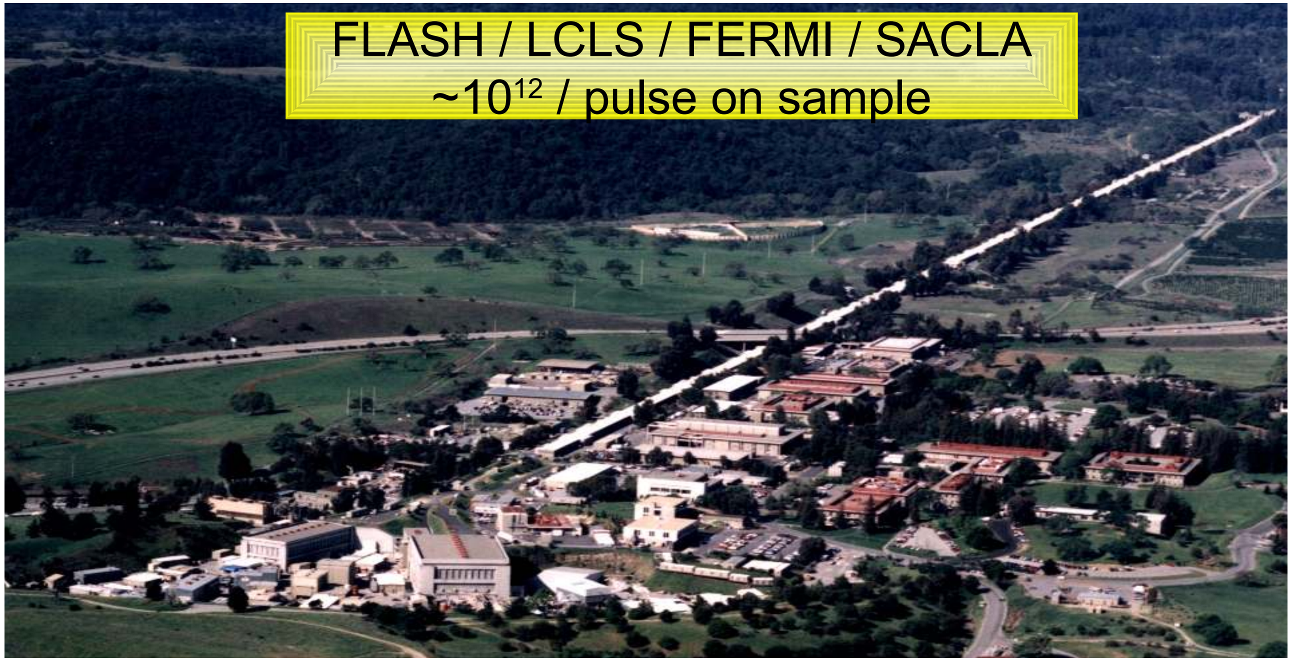
Femtoslicing (BESSY, SLS, ***SOLEIL***)
~$10^3$ / pulse on sample



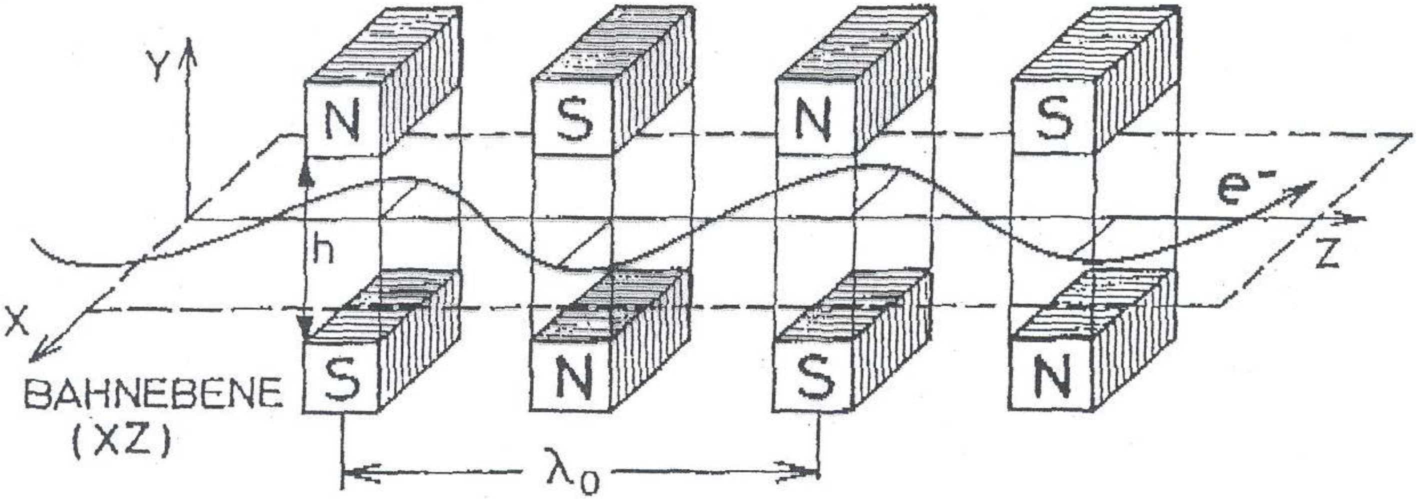

HHG
~$10^5$ / pulse on sample



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



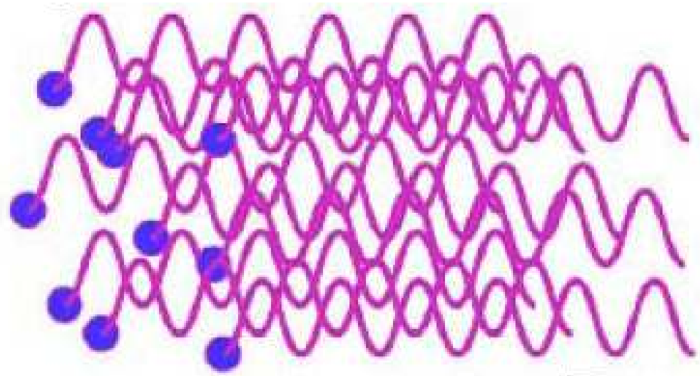
# Synchrotron radiation of an undulator

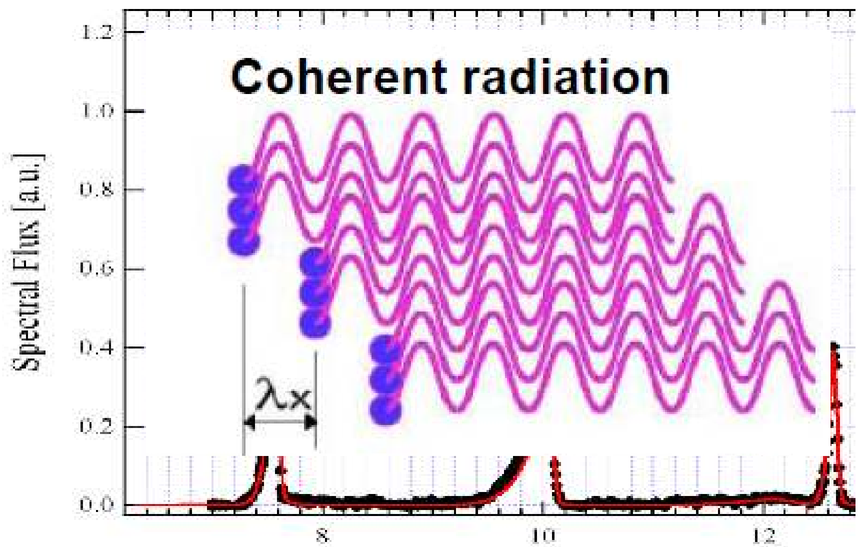



**Spontaneous emission**

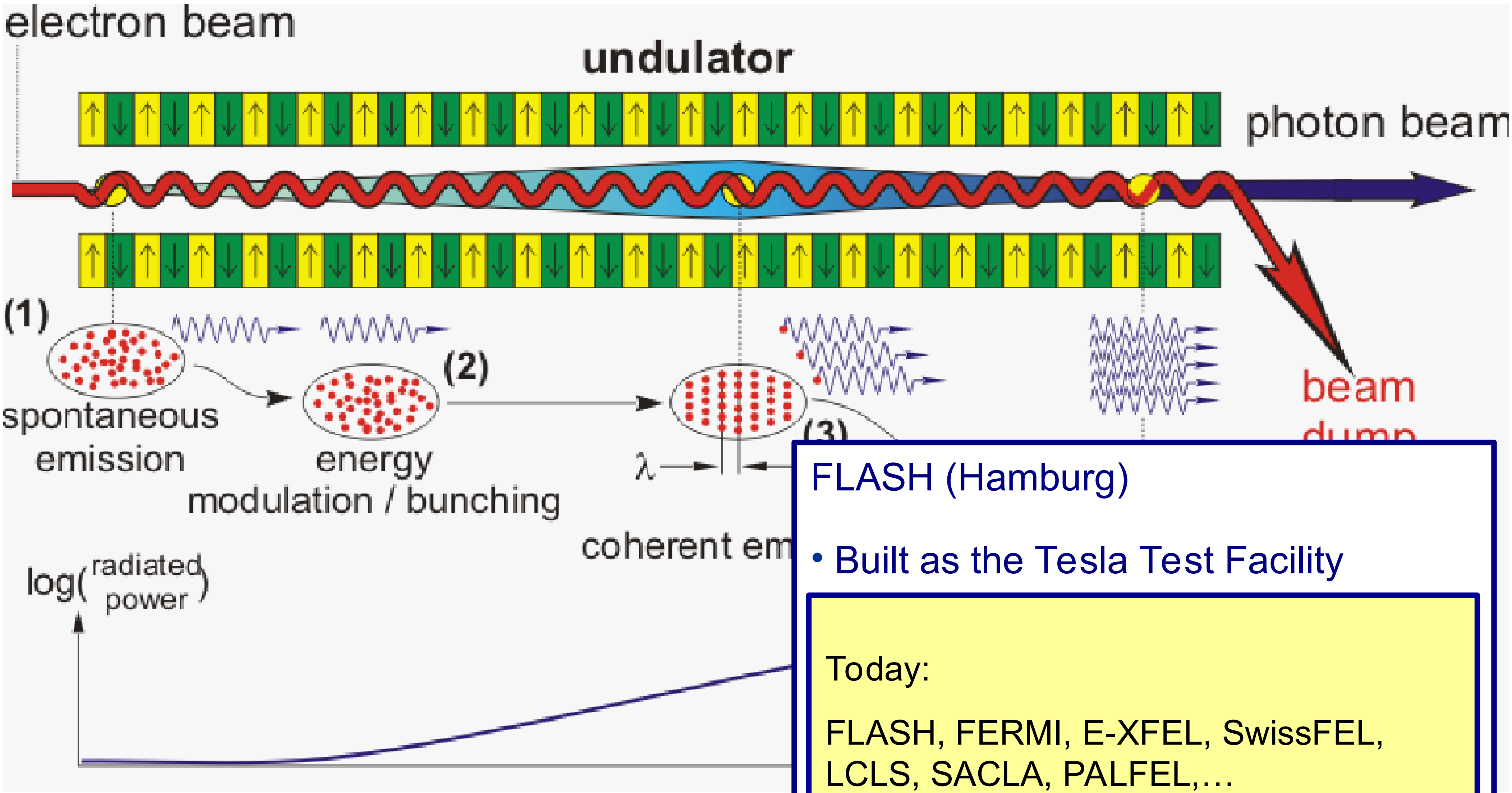
Note: each electron interferes within undulator with radiation emitted by itself!

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





# SASE-XFEL – a very long undulator




FLASH (Hamburg)

- Built as the Tesla Test Facility

Today:

FLASH, FERMI, E-XFEL, SwissFEL, LCLS, SACLA, PALFEL,…

Soon: several FELs in Chine

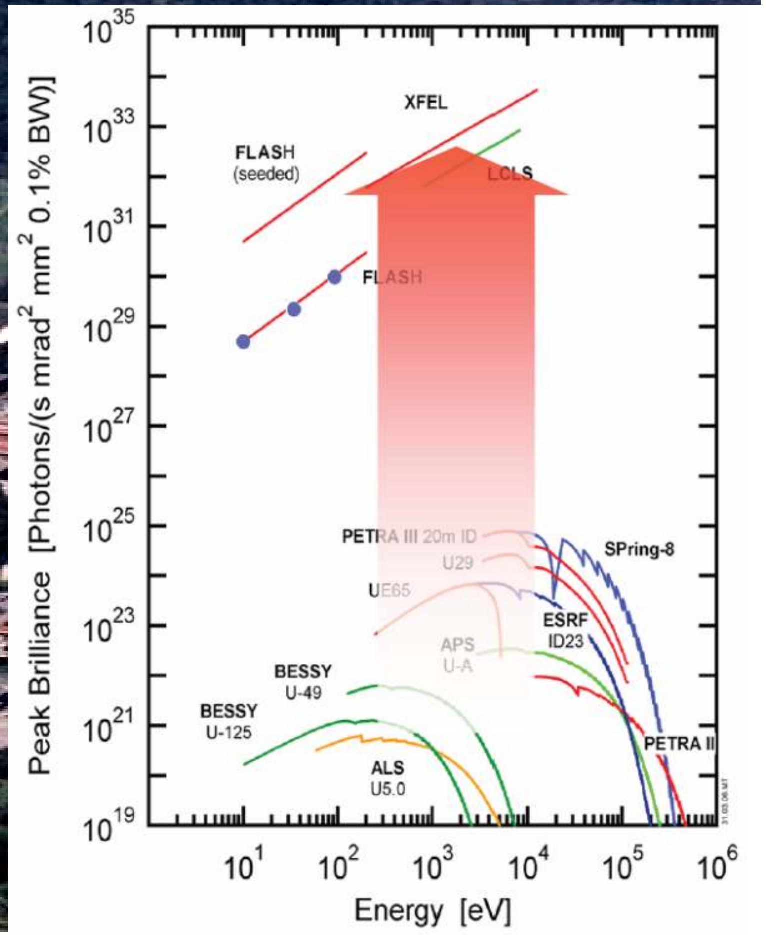
***Coherent source → Intensity ~ (# of $e^-$)$^2$***

# 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
- ...



# Acknowledgement

THE EUROPEAN SCHOOL ON MAGNETISM

| | |
|---|---|
| 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, … |

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|---|---|
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| DESY / U.Hamburg | - G. Grübel, L. Müller, C. Gutt, H.P. Oepen, ... |

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

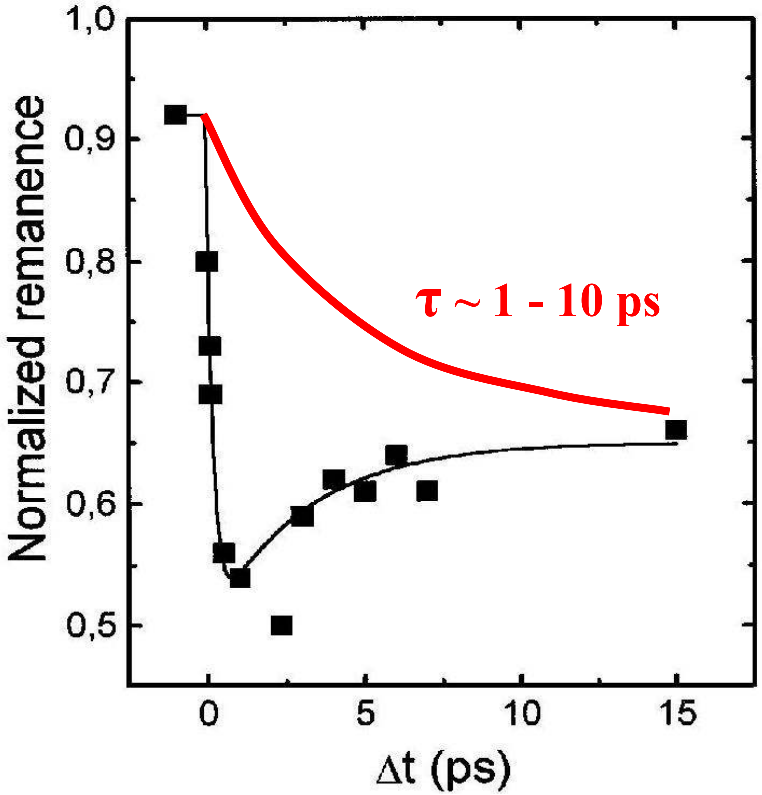
| | |
|---|---|
| SXR / LCLS | - B. Schlotter, J. Turner, … |
| DiProI / FERMI | - F. Capotondi, E. Principi, … |
| FLASH / DESY | - N. Stojanovic, K. Tiedtke, ... |

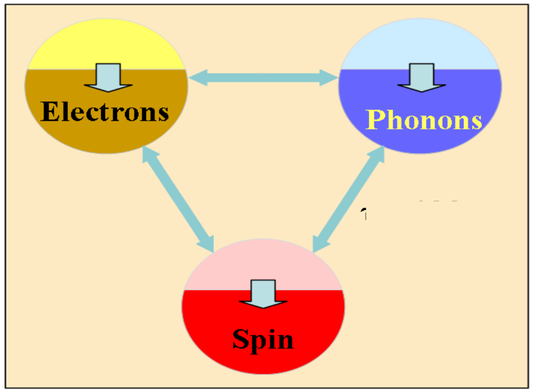
+ colleagues from the accelerator, laser, … groups

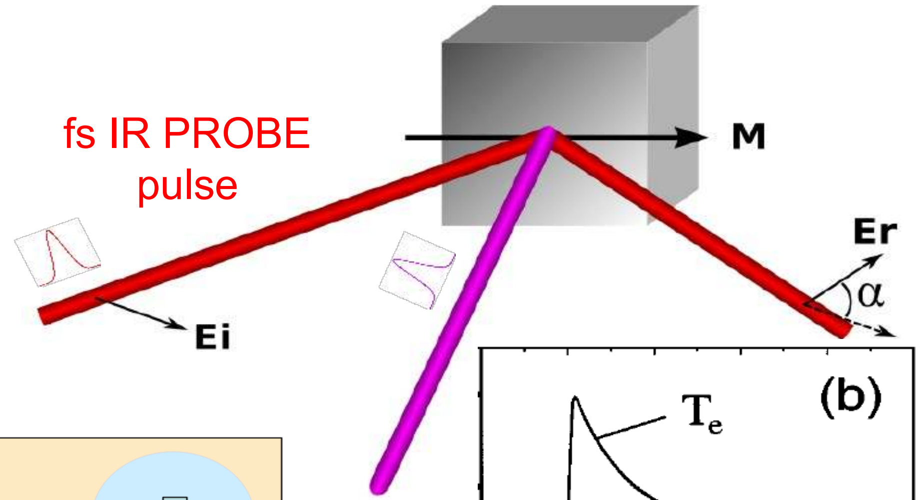
# 1996: Discovery of ultrafast magnetization dynamics

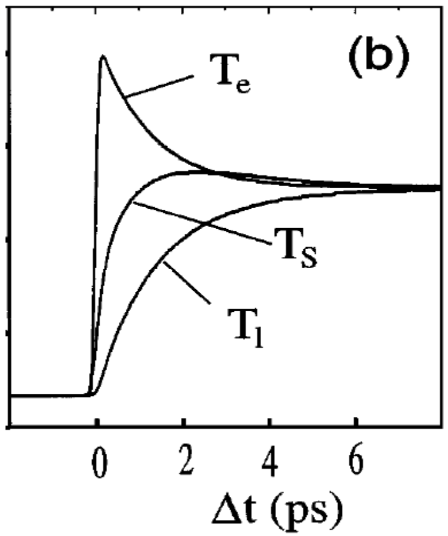
E. Baurepaire *et al.*, PRL **76**, 4250 (1996)

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



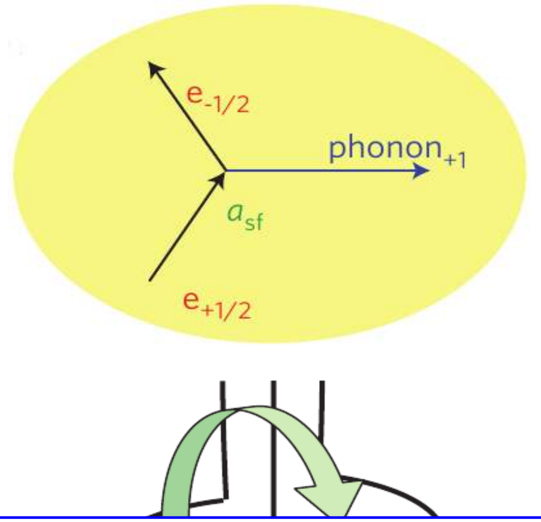




Questions still discussed since 1996:

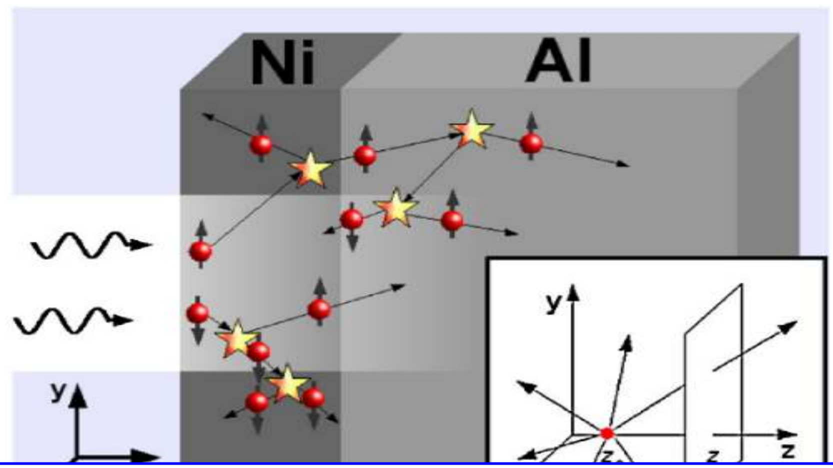
- How does energy flow into the spin system?
- What happens to the angular momentum on femtosecond time scale?

# Most discussed potential mechanisms

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

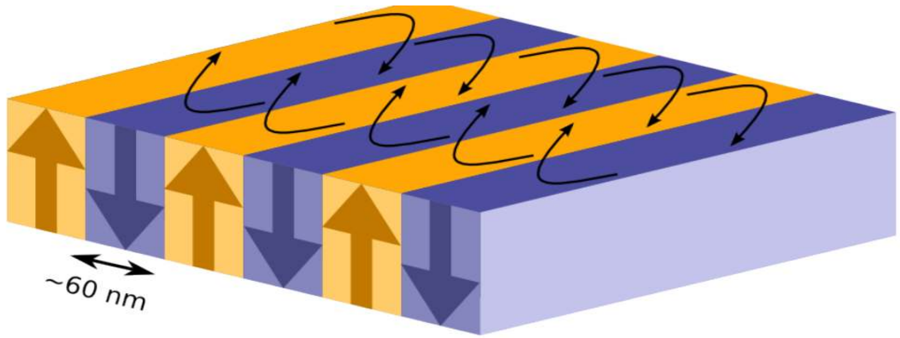



## Angular momentum transport by hot, spin-polarized electrons (non-local mechanism)



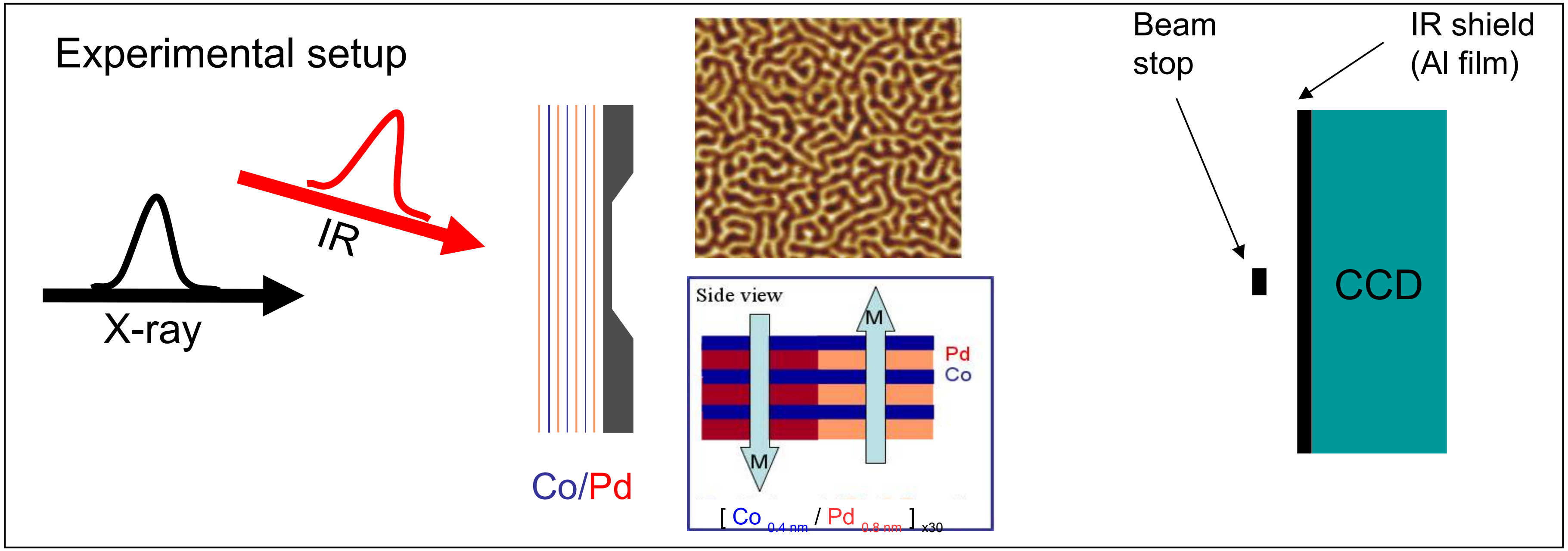

- Requires ~10 nm spatial resolution
- Element sensitivity
- Access to buried layers
- Strong dichroism signal

**→ X-ray based techniques ideally suited**



# Resonant scattering for local probing of magnetization

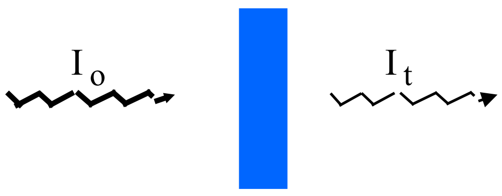
**IR** (EUV/THz) ***pump*** – Resonant (magnetic) **X-ray** (small angle) scattering ***probe***




**Integrated intensity → measure of the local magnetization**

# XMCD in Absorption and Scattering

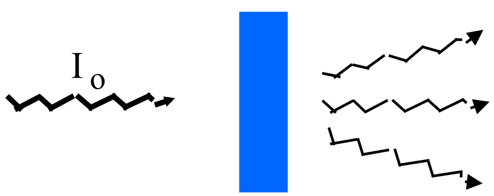
## Absorption




Sample density ρ

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

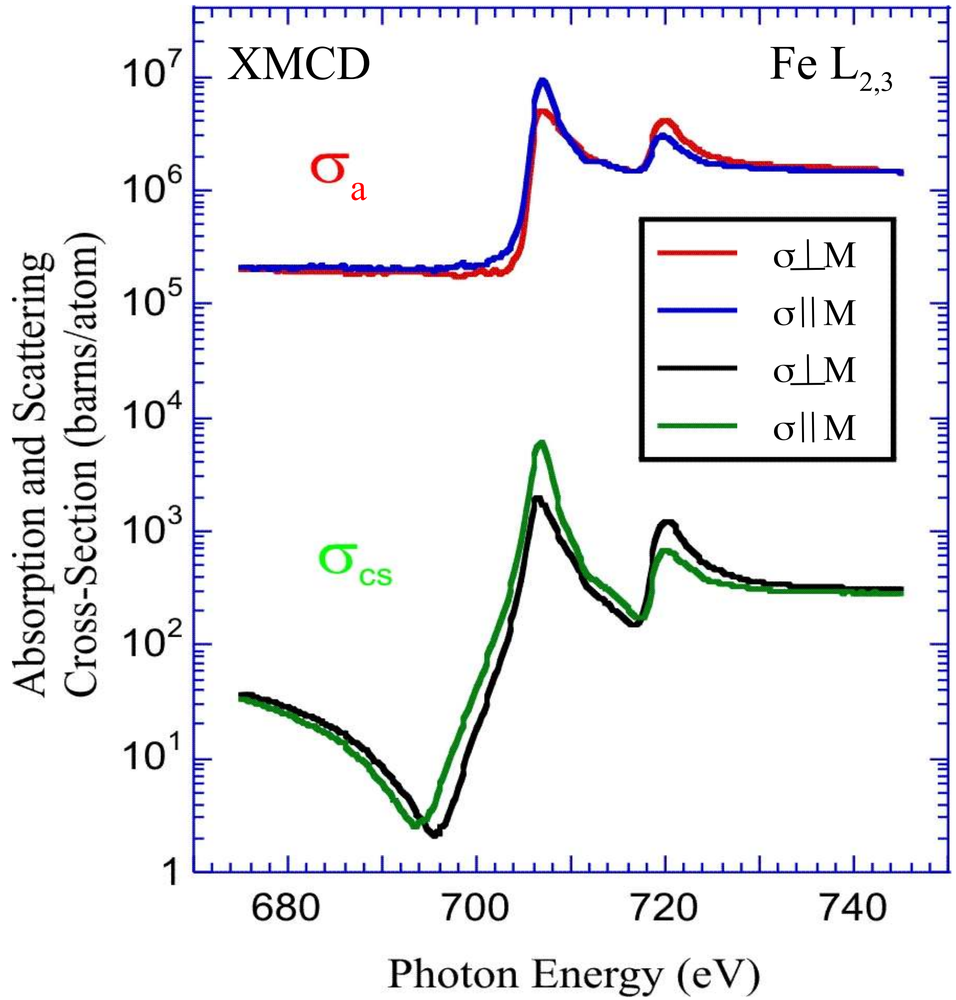
## Small Angle Scattering




Sample density ρ

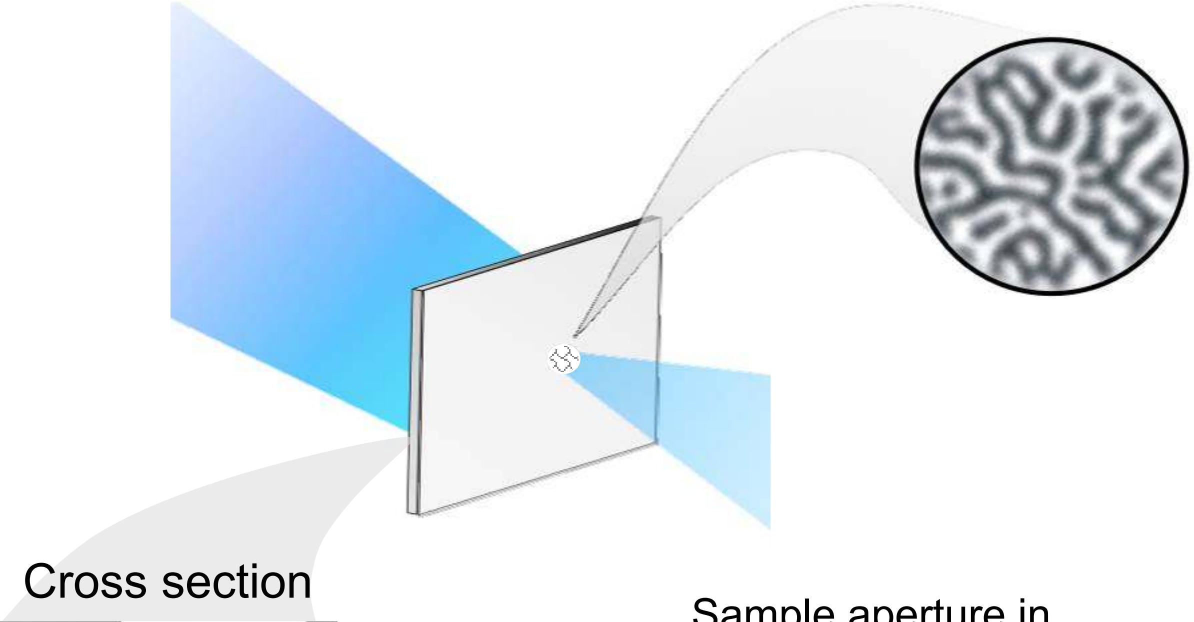
$$I_{cs} \sim I_o \Delta\sigma_{cs} / \sigma_a$$

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

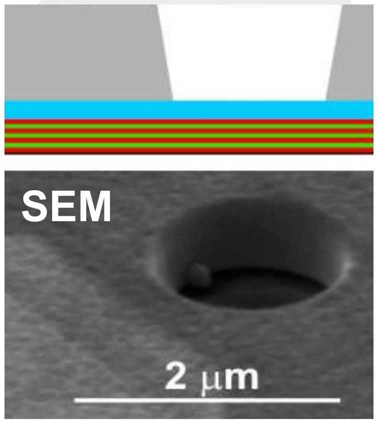



Data from Jeff Kortright (LBNL)

# Experimental geometry



Cross section



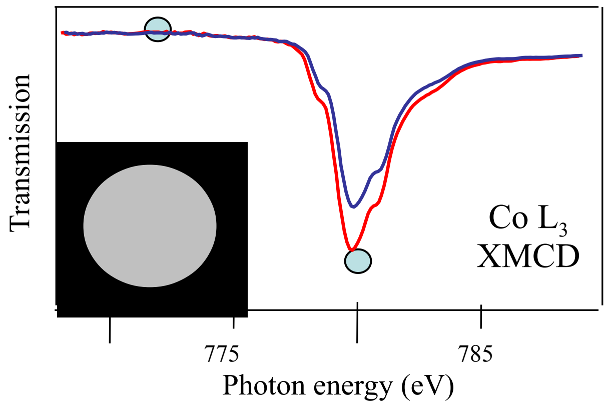

Au
SiN
Magn film

Sample aperture in X-ray opaque Au film is ‘drilled’ with focused ion beam

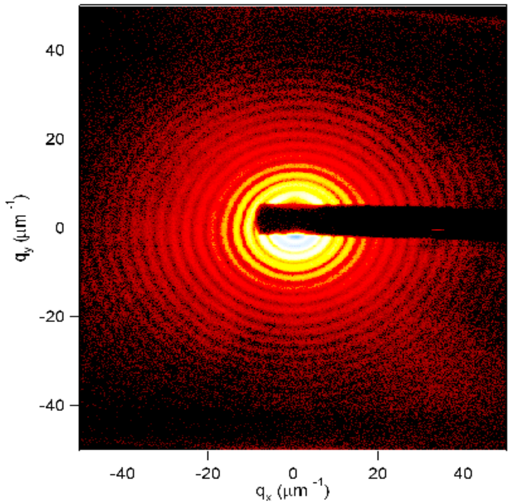
# Magnetic scattering contrast



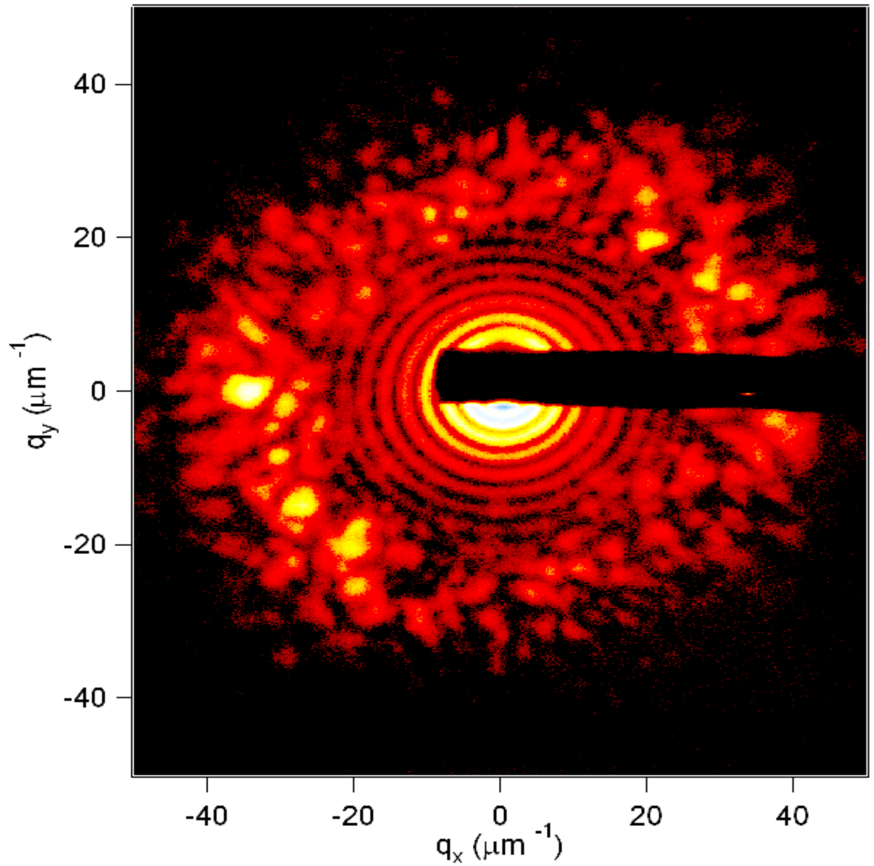
Scattering of coherent X-rays yields
Fourier Transformation of scatterin object




Below Resonance




On Resonance

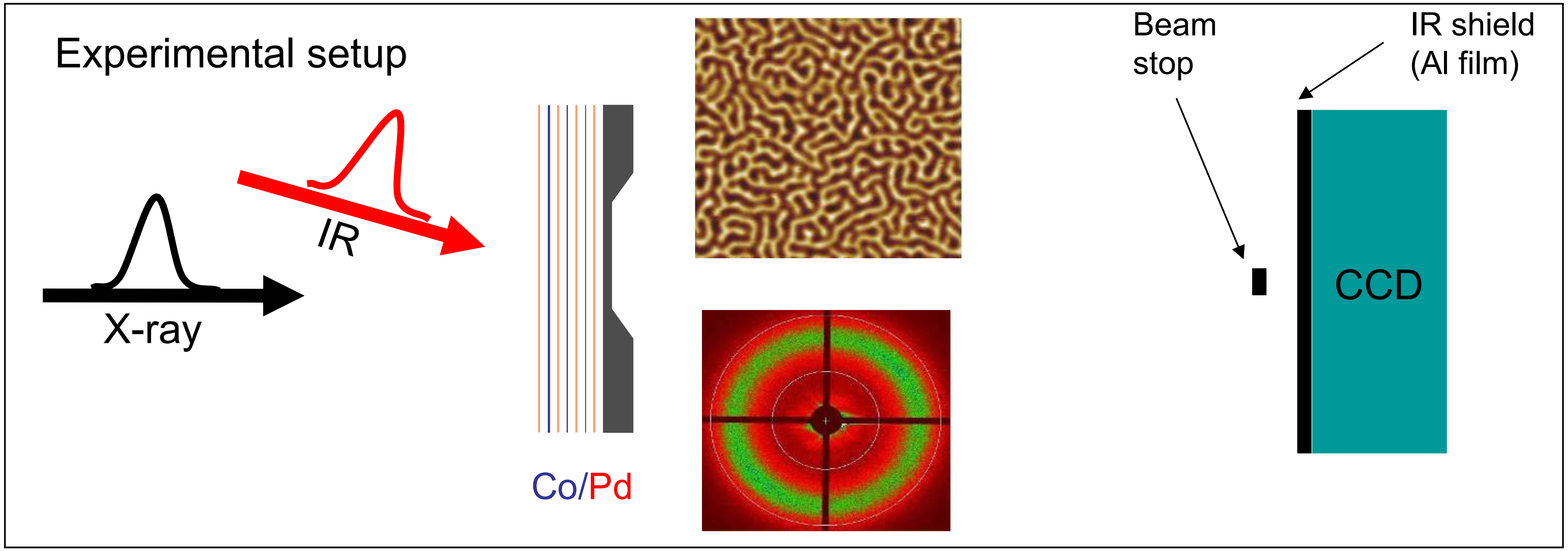



$\lambda$ = 1.59 nm, 2.5 mm ⌀ Pinhole
fully coherent illumination: visibility = 1, M = 1

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

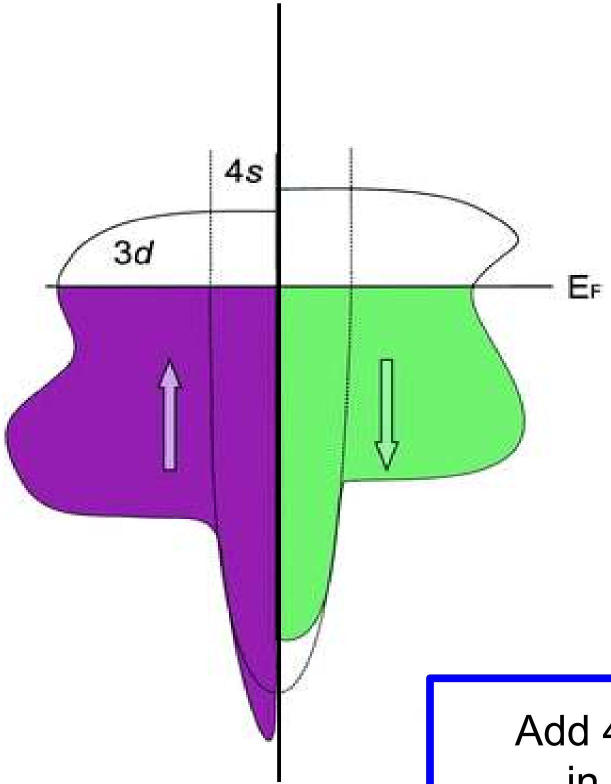
Magnetically dichroic absorption edges of transition metals:

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



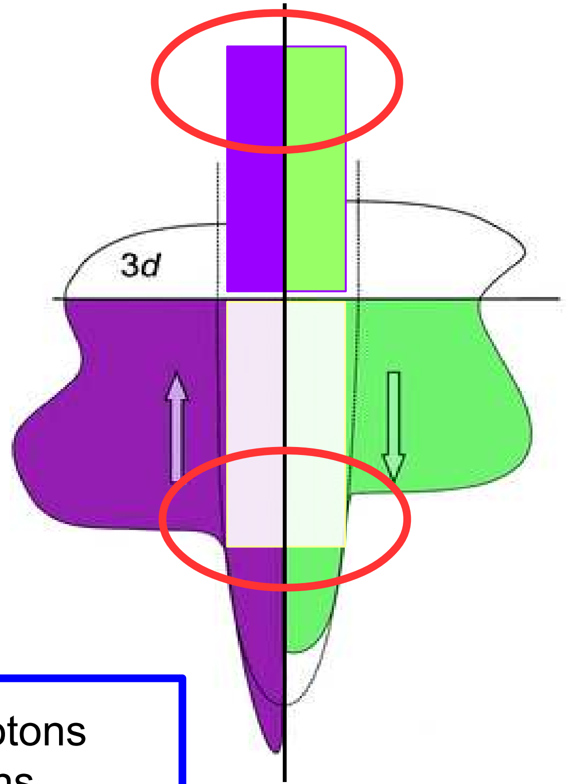

**Integrated intensity → measure of the local magnetization**

# Relevance of hot, directly excited valence electrons

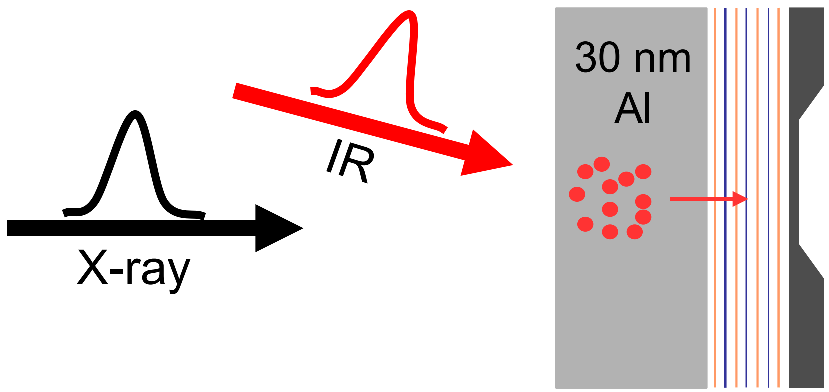



**1.5 eV laser**

**excitation**



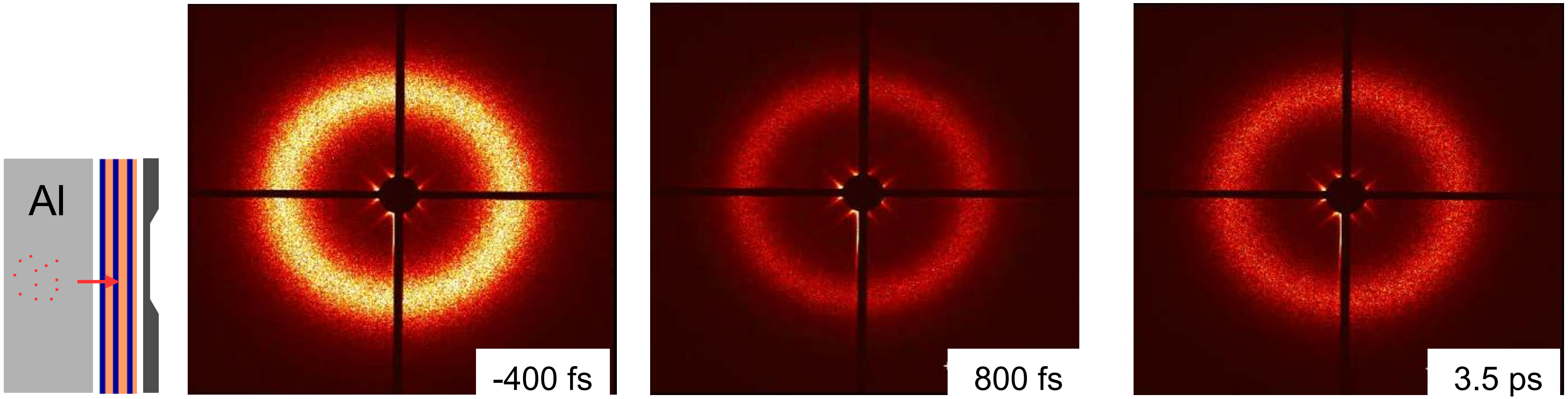

Add 40 nm Alu cap layer to convert IR photons
in avalanche of excited valence electrons

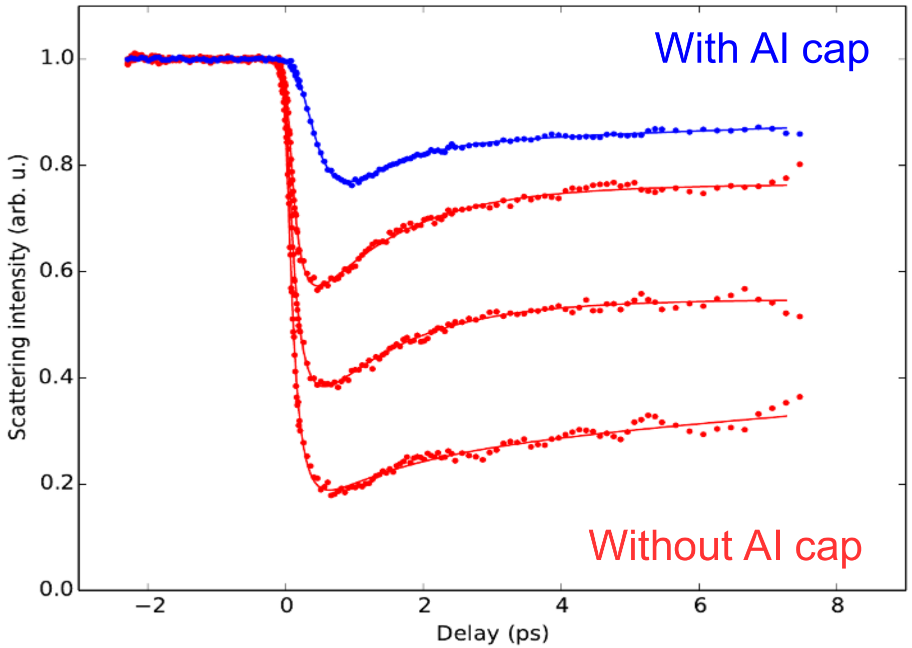


# Hot electron excited ultrafast magnetization dynamics

*SXR @ LCLS*

B. Vodungbo, to be published (2015)




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

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

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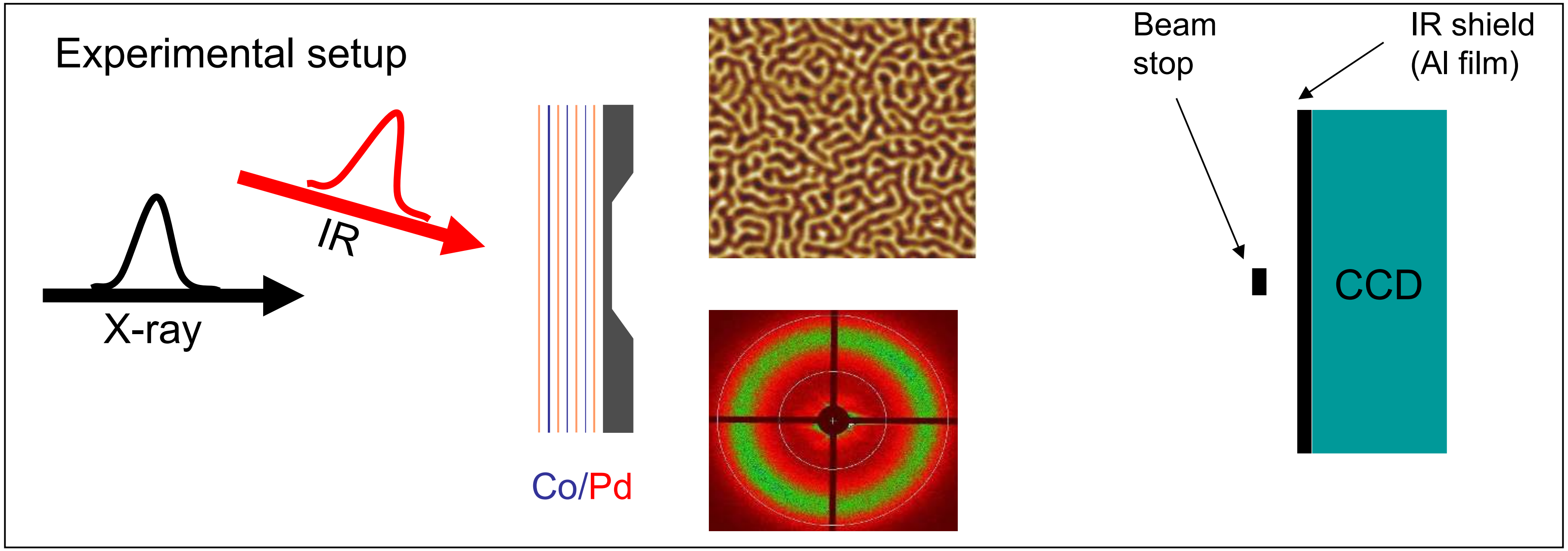
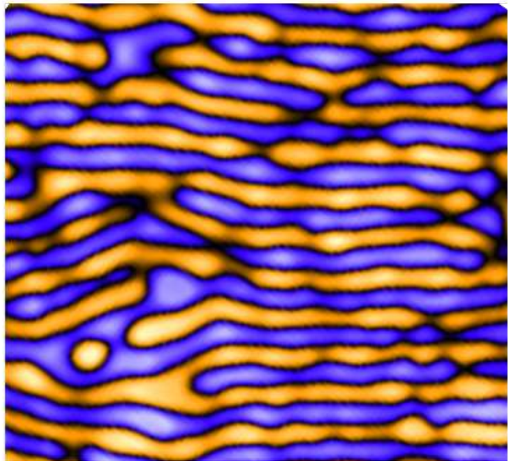



Integrated intensity → measure of the local magnetization

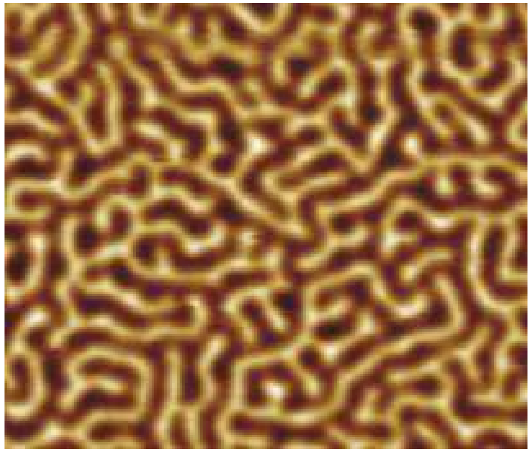
**Form of scattering pattern → spatial information**

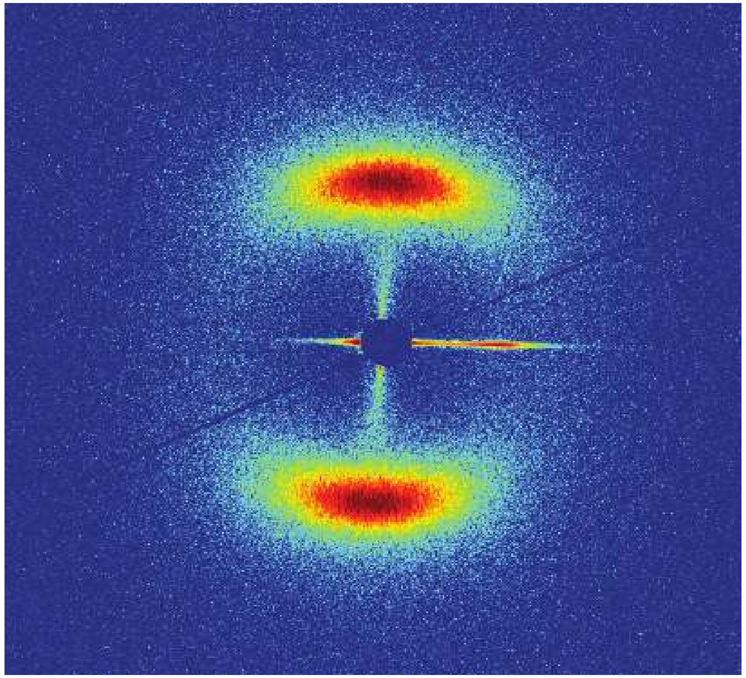
# Limit of ***very strong*** IR pump



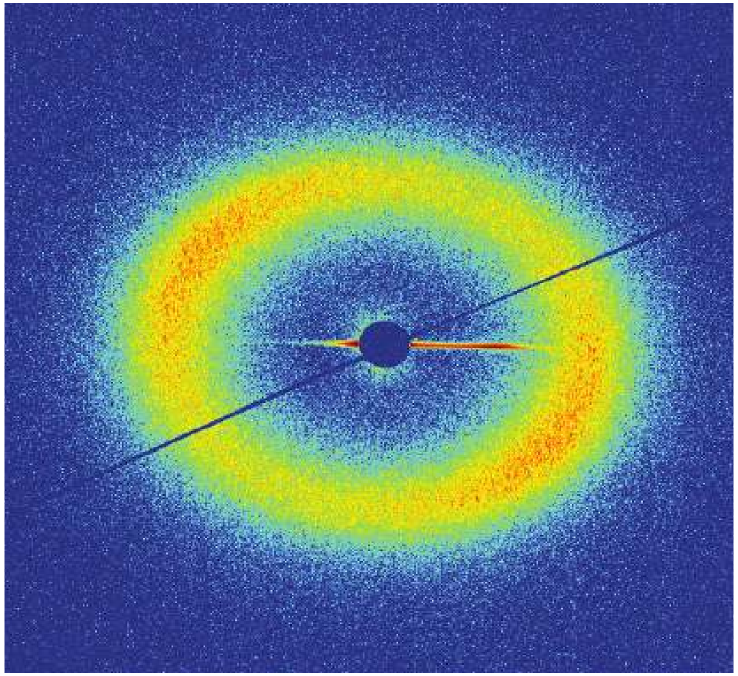


?



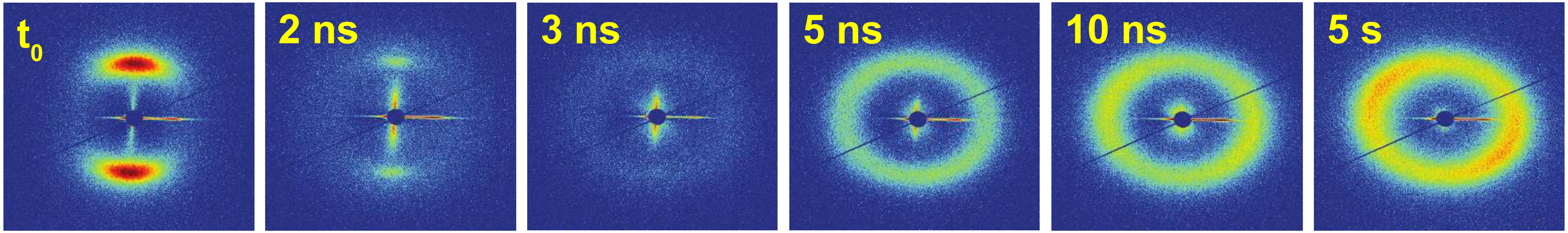


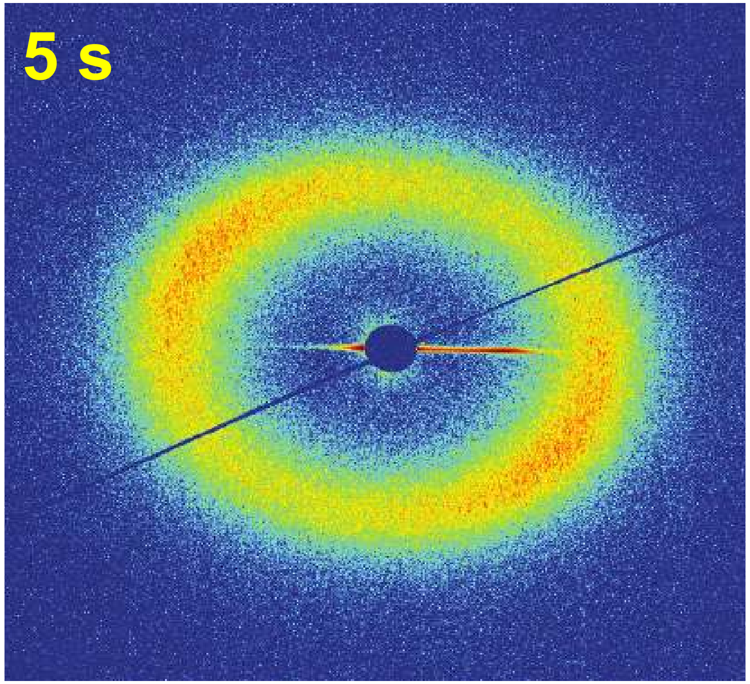
**Single, very intense IR pulse**



# Studying non-reproducible magnetization dynamics

C. Boeglin et al., LCLS (2012)



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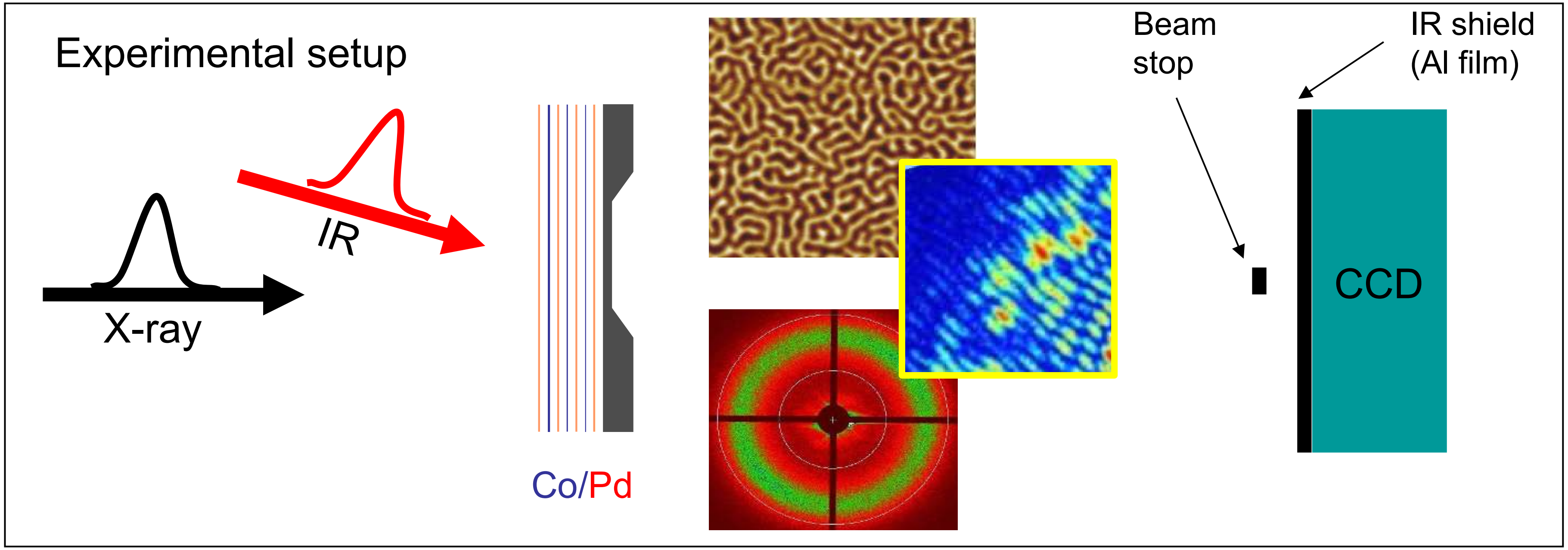
Magnetically dichroic absorption edges of transition metals:

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

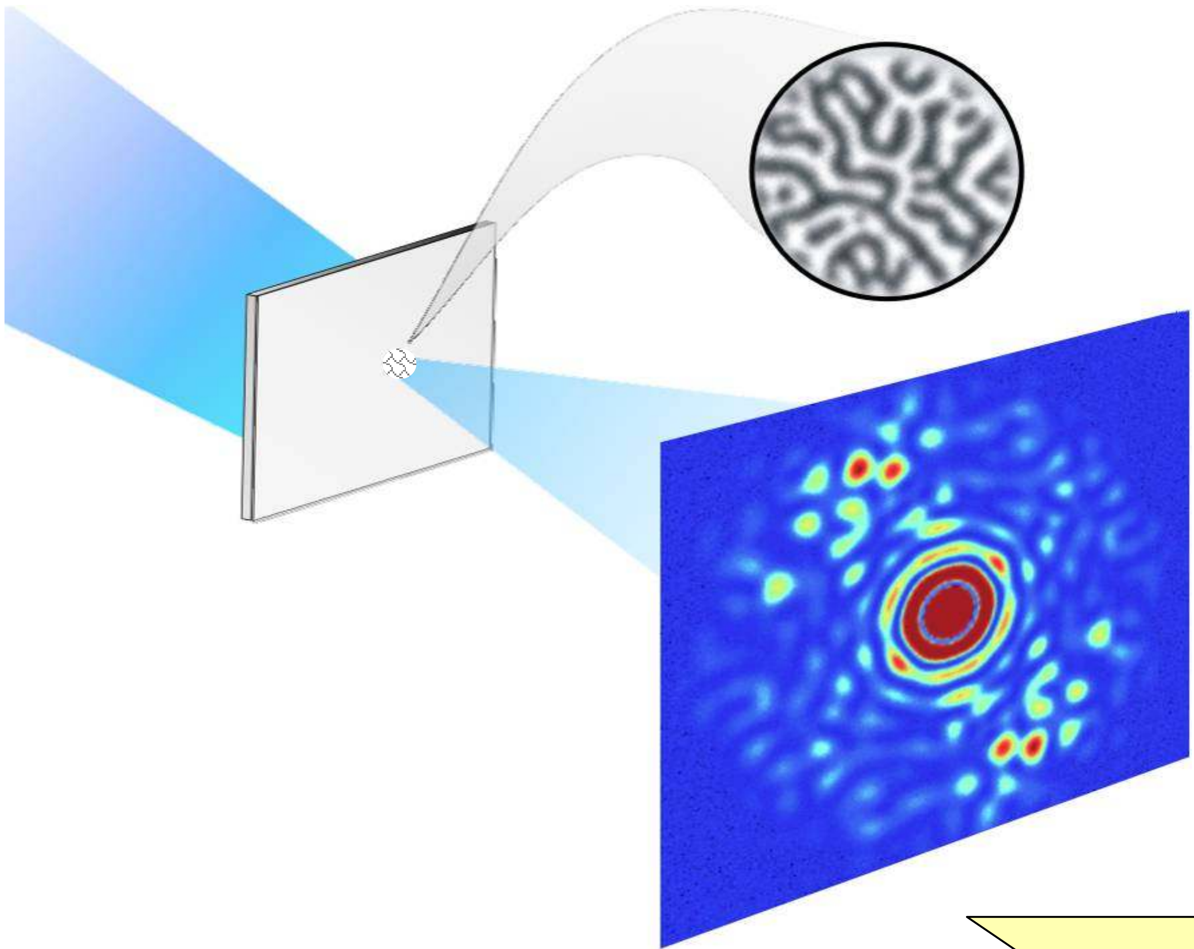



Integrated intensity → measure of the local magnetization

Form of scattering pattern → spatial information

**Speckle → imaging**

# Phase problem in X-ray scattering

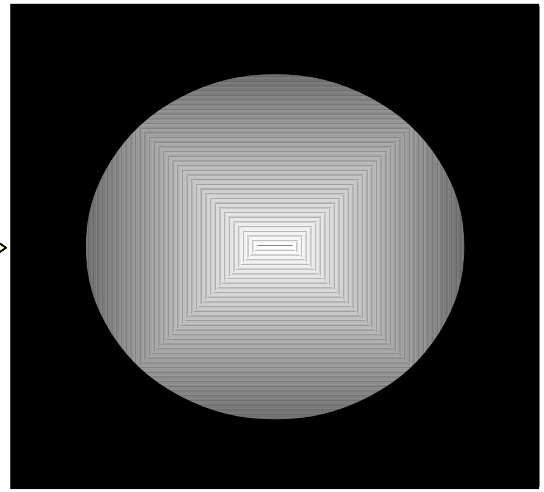


Scattering amplitude
is complex, but only
intensities are detected

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

**Fourier Transform**

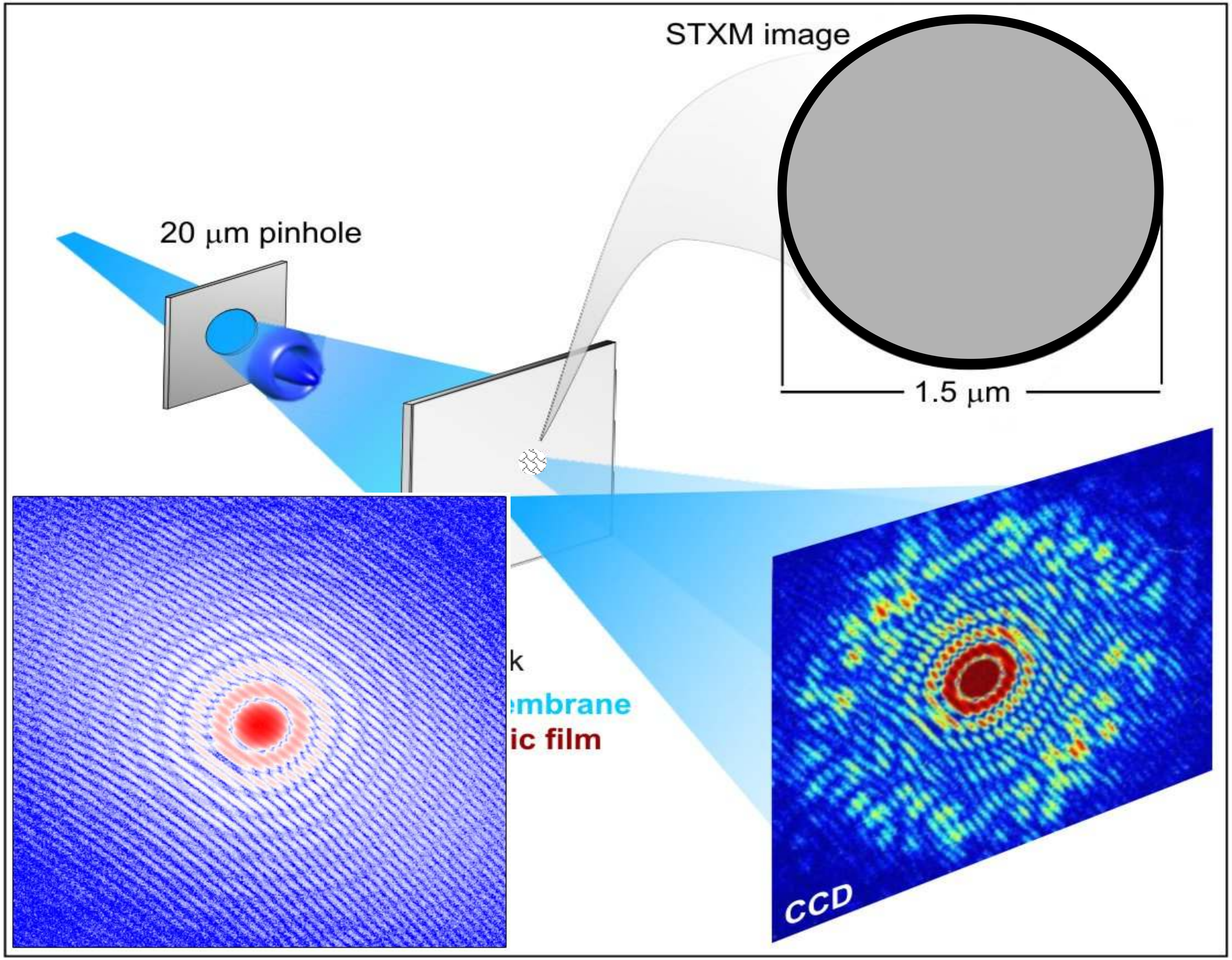
Auto-correlation



**Convolution theorem applied to diffraction**

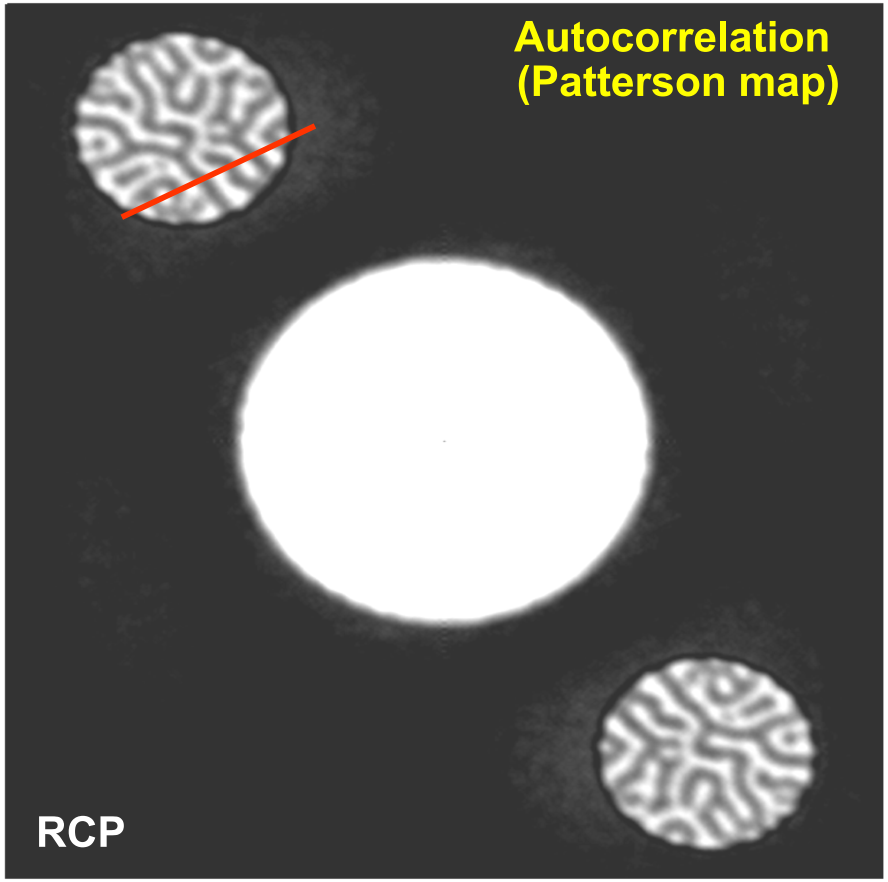
$$\mathbf{(a \otimes a) = FT^{-1}\{FT(a) \cdot FT(a)\}}$$

# Fourier transform X-ray spectro-holography

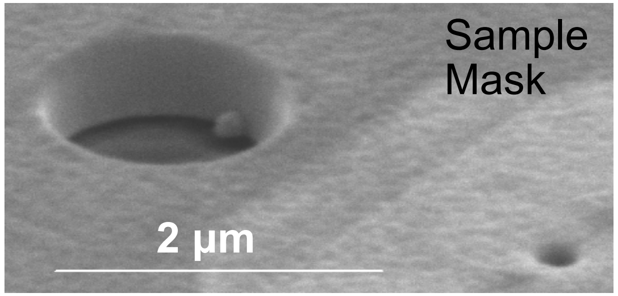


# Digital image reconstruction

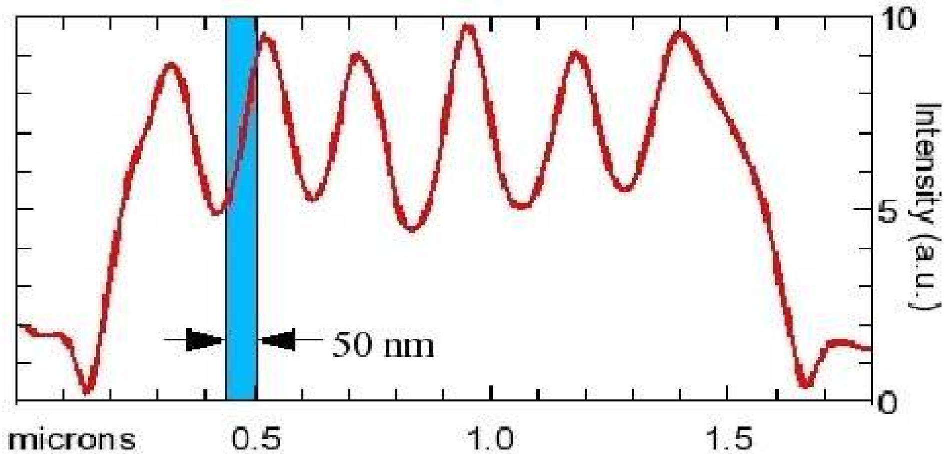
Single Fourier transformation of scattering intensities yields the auto-correlation of sample, which contains image of sample due to the off-axis geometry in FT holography (convolution theorem).




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



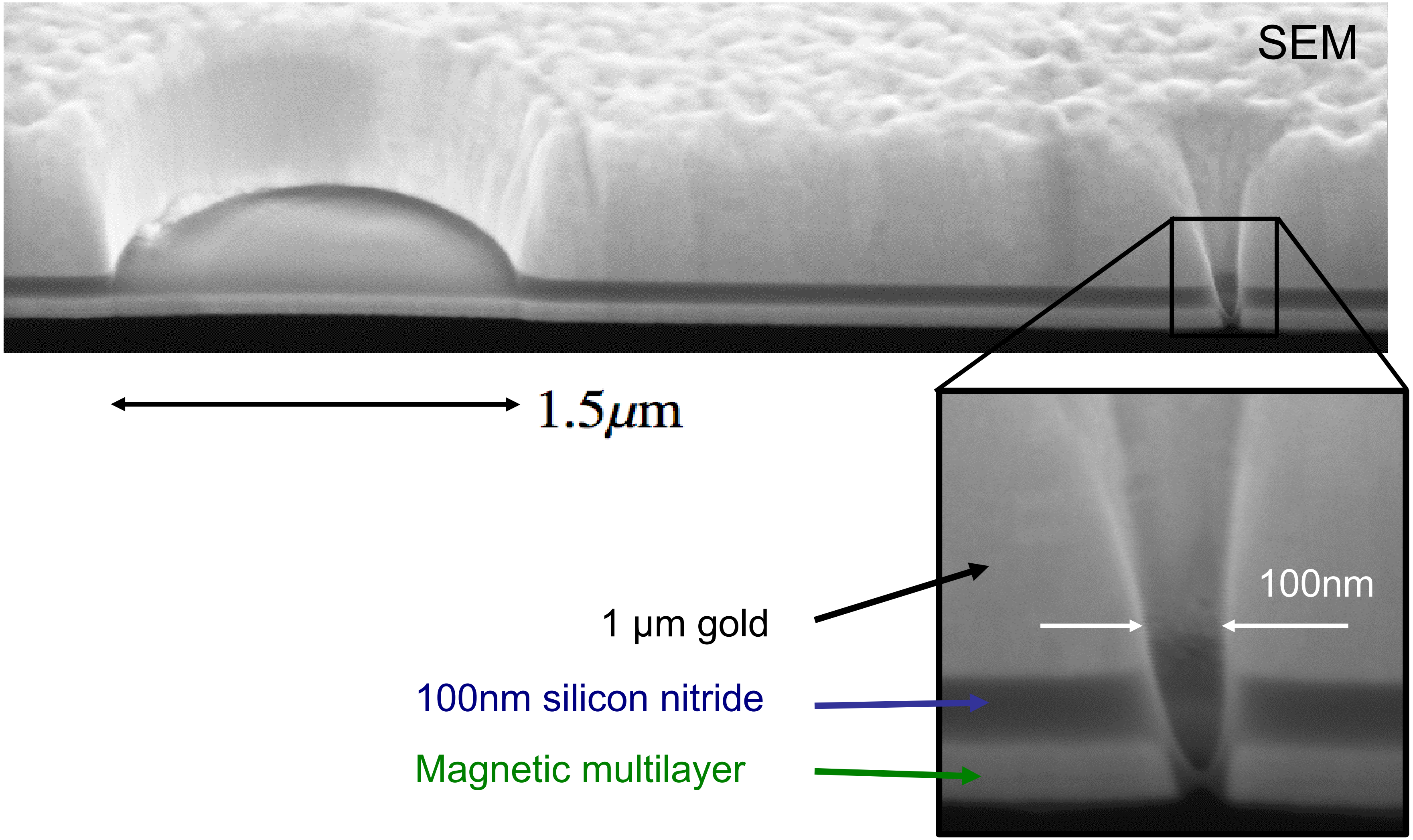

10% - 90% intensity rise over about 50 nm



# Integrated mask sample structure



Patterned with focused ion beam



# Key properties of Fourier transform X-ray holography

- ***True imaging technique***

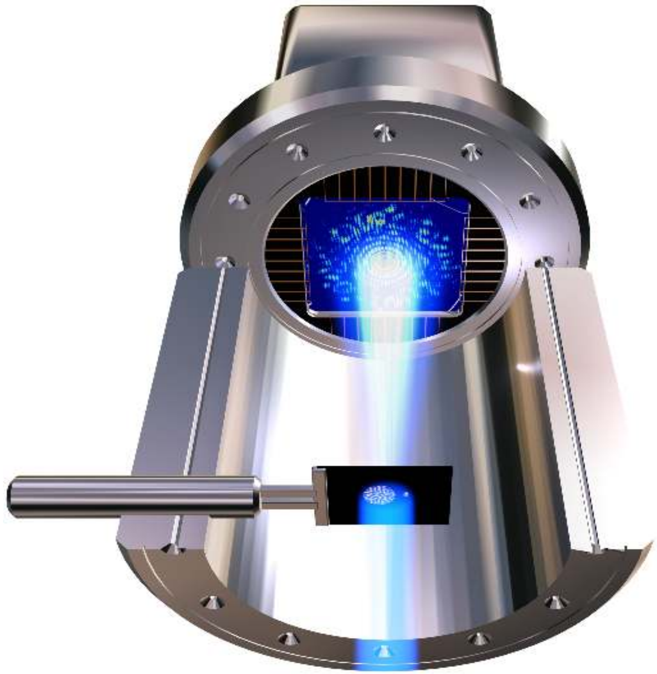
- ***Wavelength limited spatial resolution***
  Deconvolution and phase retrieval algorithm

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

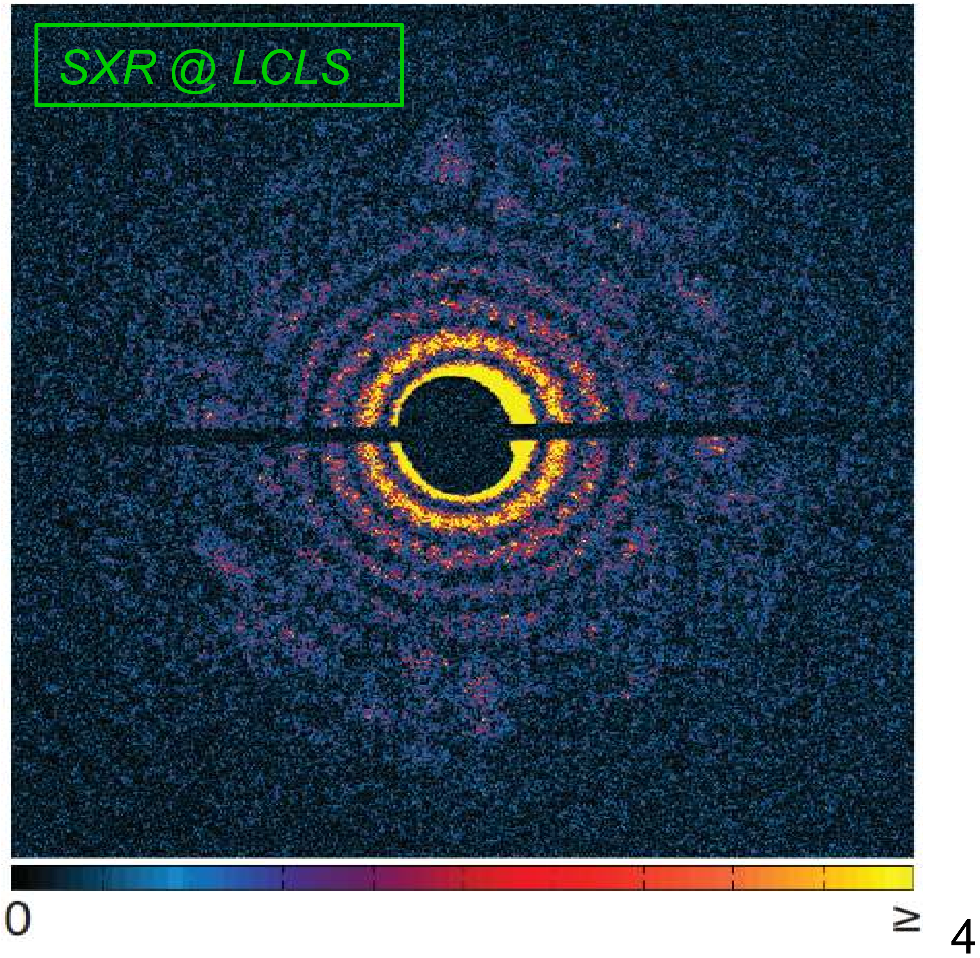
- ***Nanometer resolution with micron stability***
  Setup is basically insensitive to vibrations
  or thermal drifts

- ***Ideally suited for in-situ studies***
  **-** No space constraint around sample
  - Application of extreme temperatures and fields
  - In-situ sample growth or self-assembly
  - Operation of electric or magnetic devices

- ***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




FFT

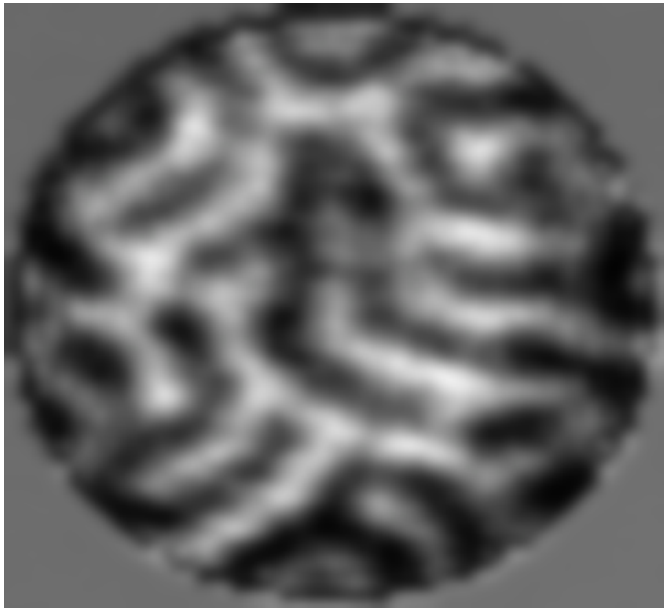


Image of magnetic domain structure obtained from a single X-ray pulse

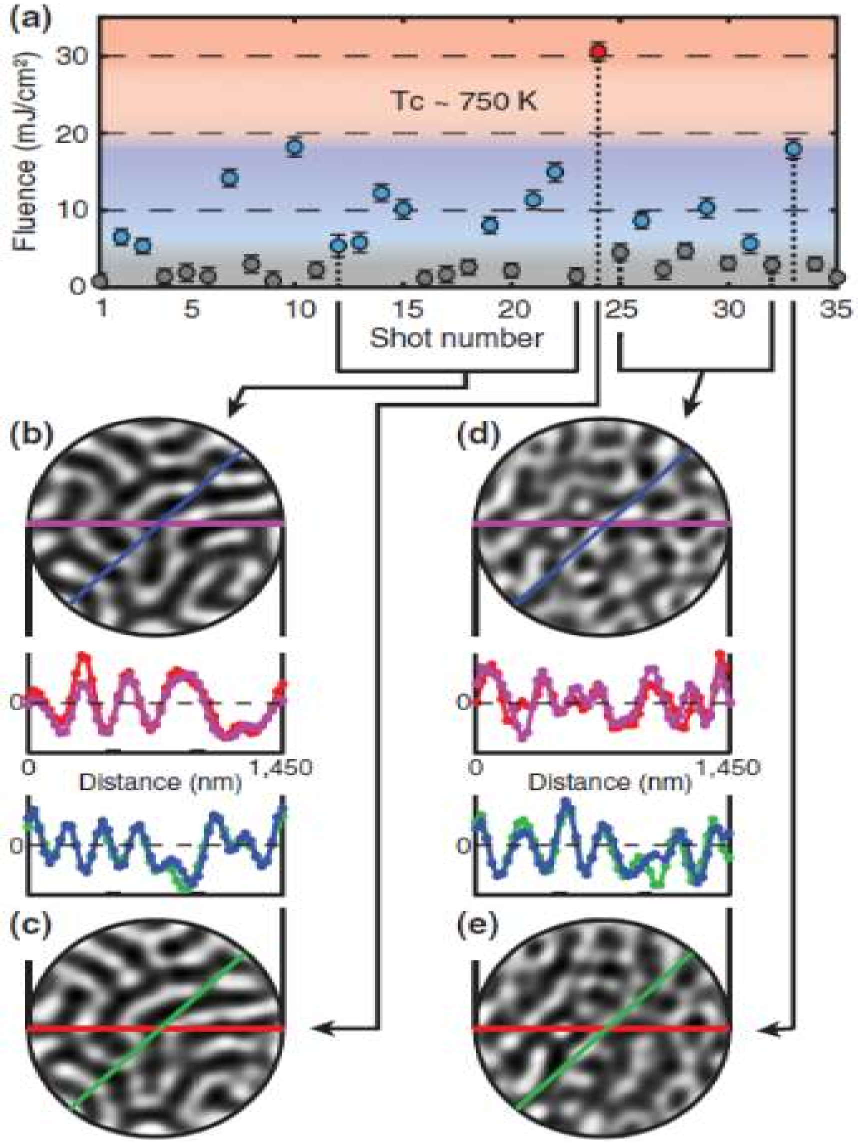
~ 50 nm spatial resolution
~ < 80 fs temporal resolution

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

# X-ray induced *“modifications”*

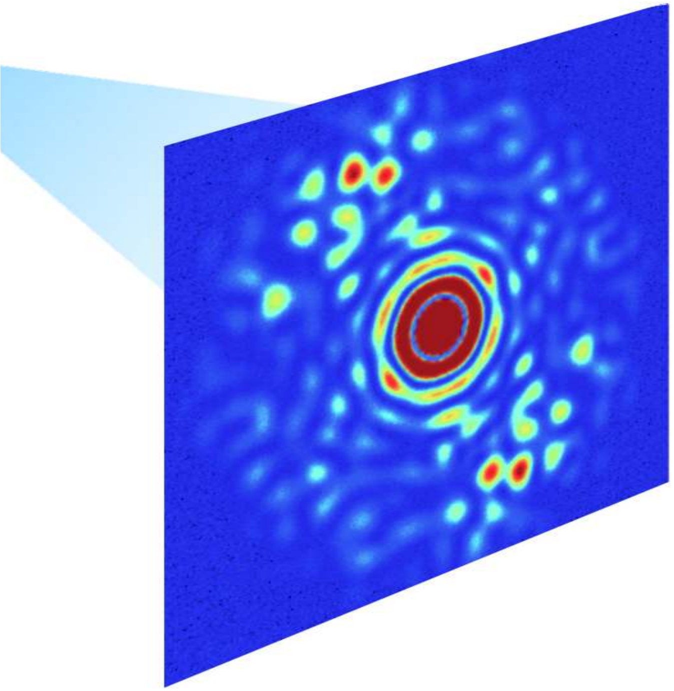
T. Wang et al., PRL **108**, 267403 (2012)

- 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.




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

# Solving the phase problem



**Phase problem in X-ray scattering:**

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

**Solutions:**

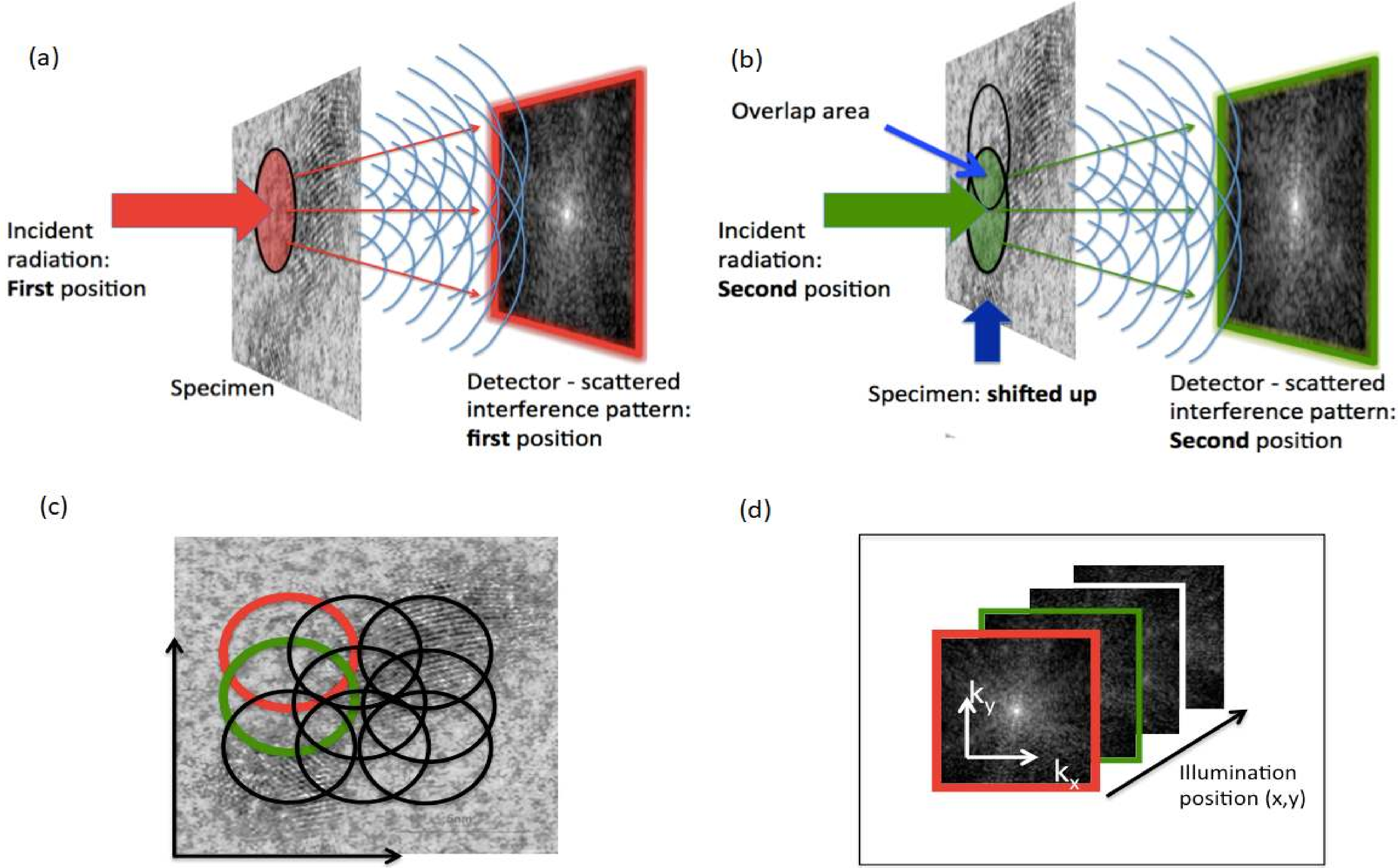
**1) X-ray Holography** (Gabor 1948, Stroke 1965)

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

**2) Iterative Phase Retrieval** (Sayers 1952)

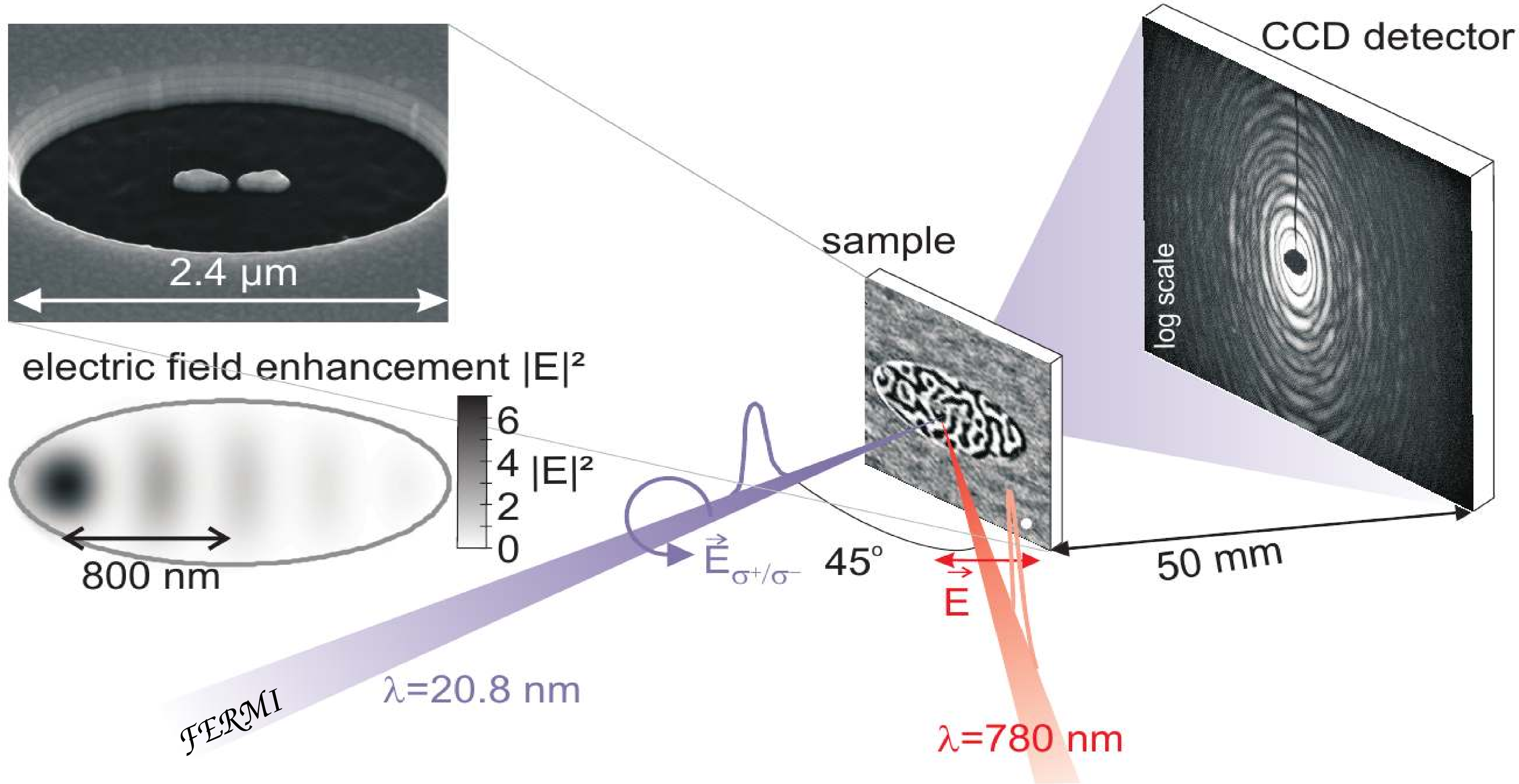
- Surround sample with ‘known’ support
- Measure additional scattering intensities (‘oversampling’)
- Use iterative algorithm to retrieve scattering phases from additional scattering intensities

# Ptychography (→ Wikipedia)



# Imaging ultrafast demagnetization dynamics

## after a spatially localized optical excitation



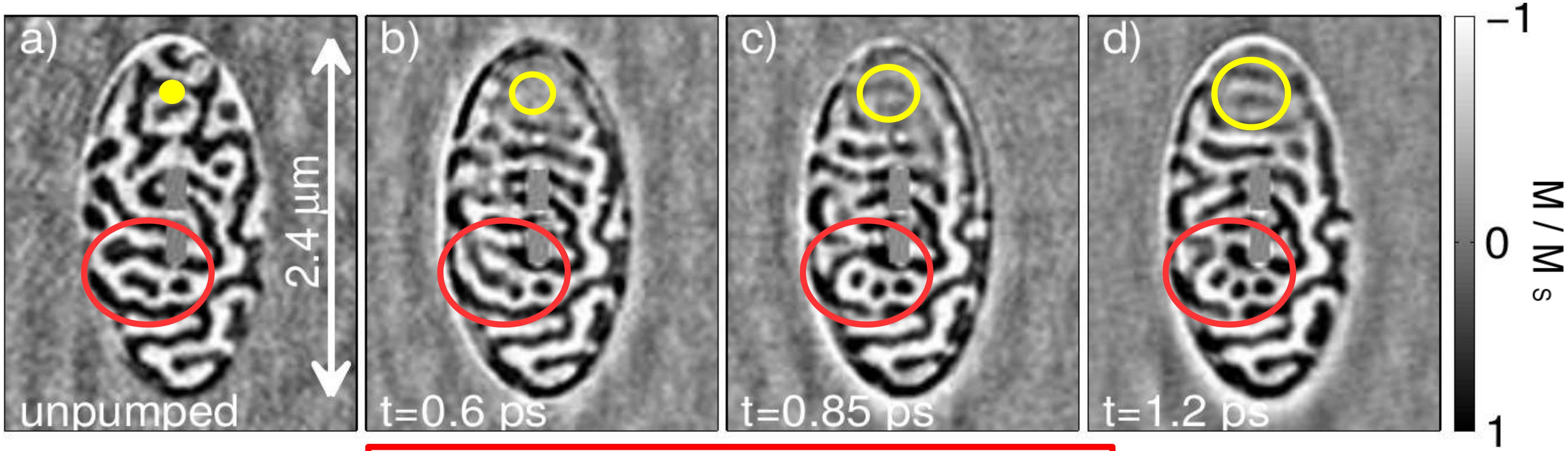

C. von Korff Schmising et al., Phys. Rev. Lett. **112**, 217203 (2014)

# Imaging ultrafast demagnetization dynamics

*DiProI @ FERMI*

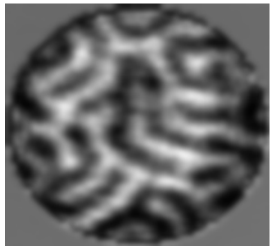
## after a spatially localized optical excitation

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




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

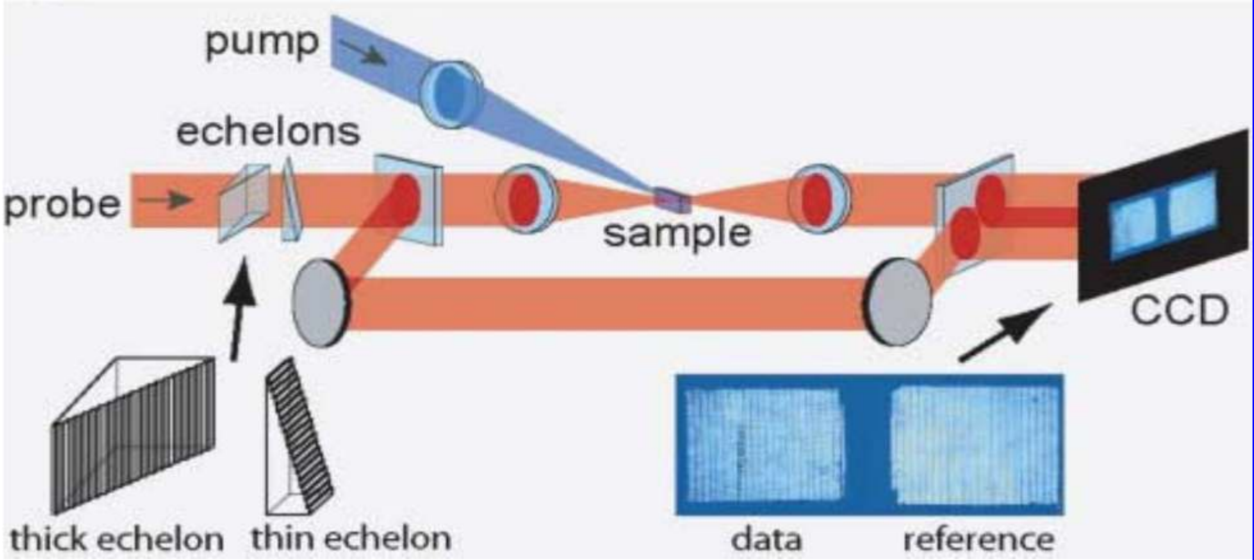
Excellent signal-to-noise due to very high pulse intensity, even for single pulse (snapshot) probing



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

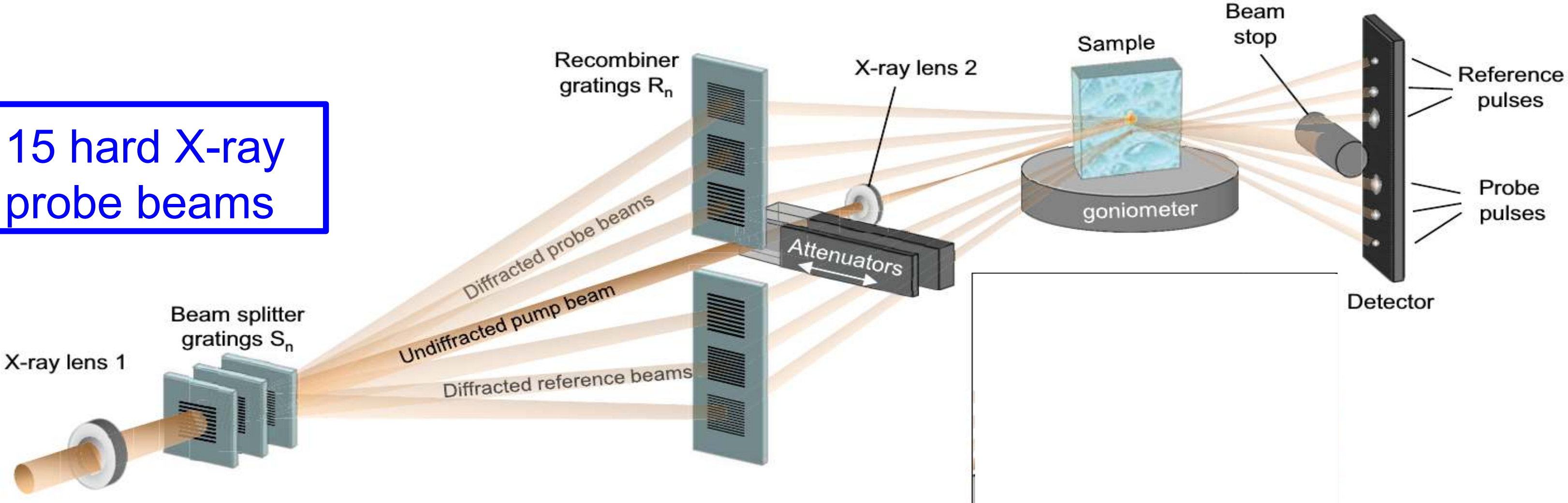
# Sampling several pump-probe delays at once

400 optical probe beams



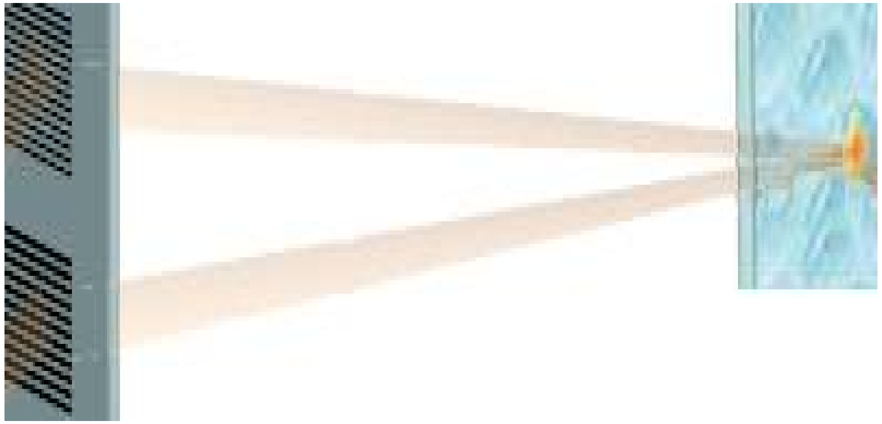

P.R. Poulin & K.A. Nelson, Science 313, 1756 (2006).

15 hard X-ray probe beams

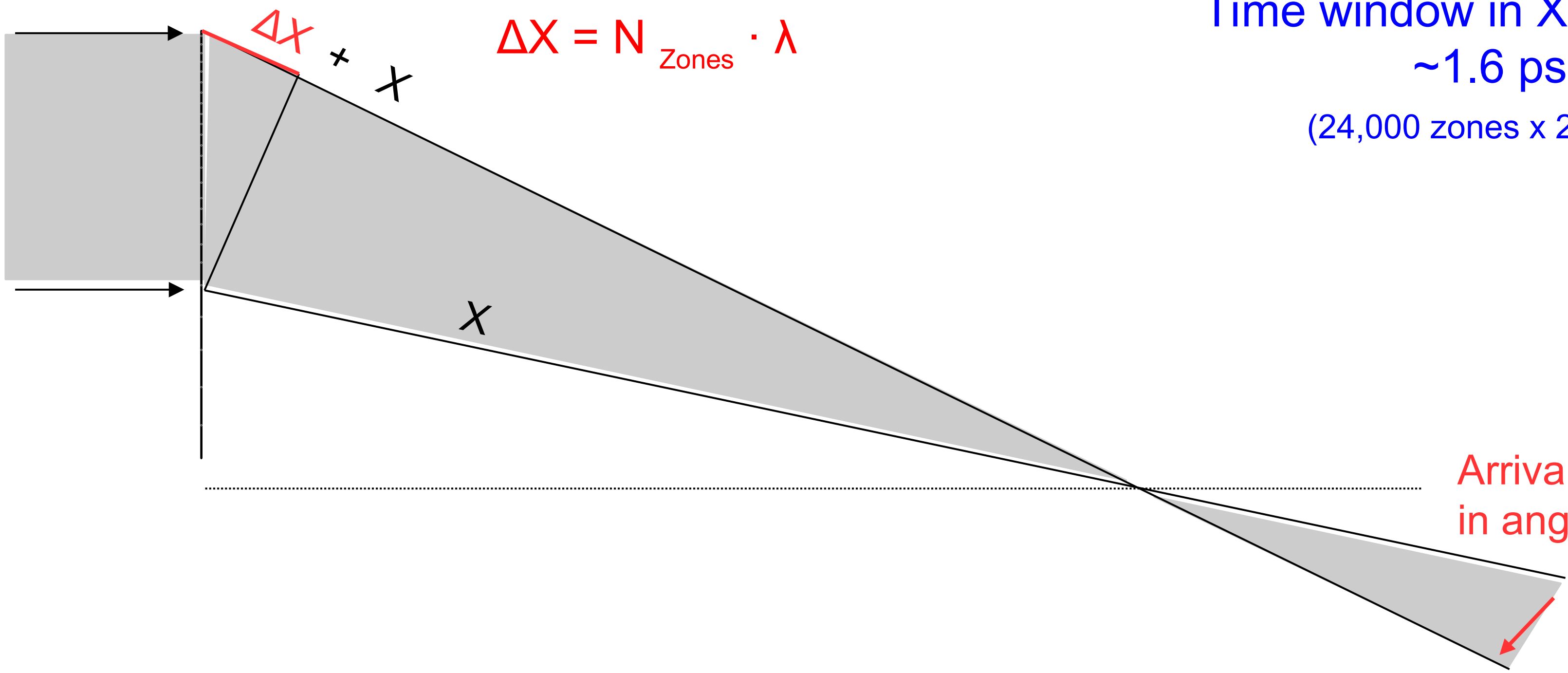



C. David et al., Scientific Reports 5, 7644 (2015)

# 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

Tw

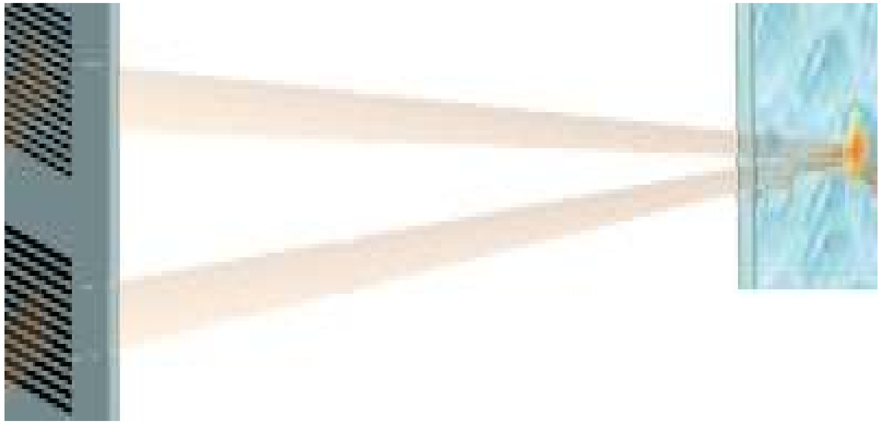
Mi
Re

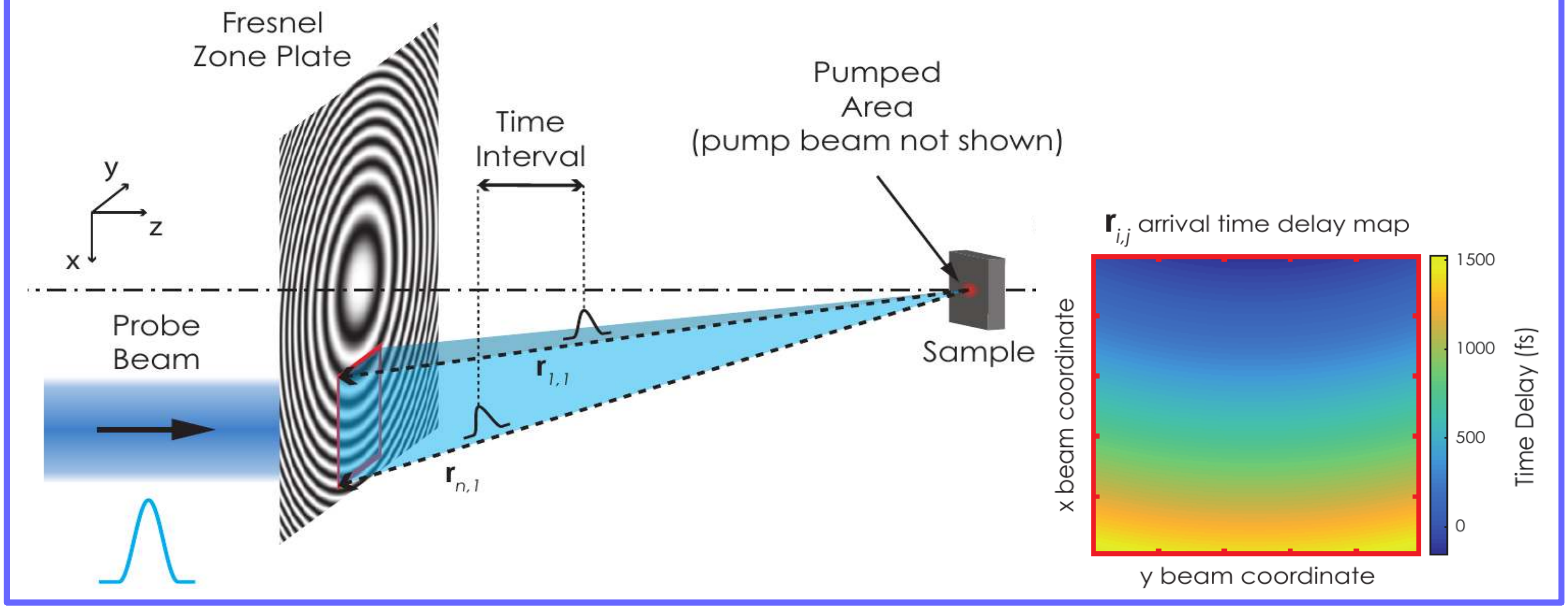
Mi
De

Ex
C.

# Snapshot recording of ultrafast dynamics







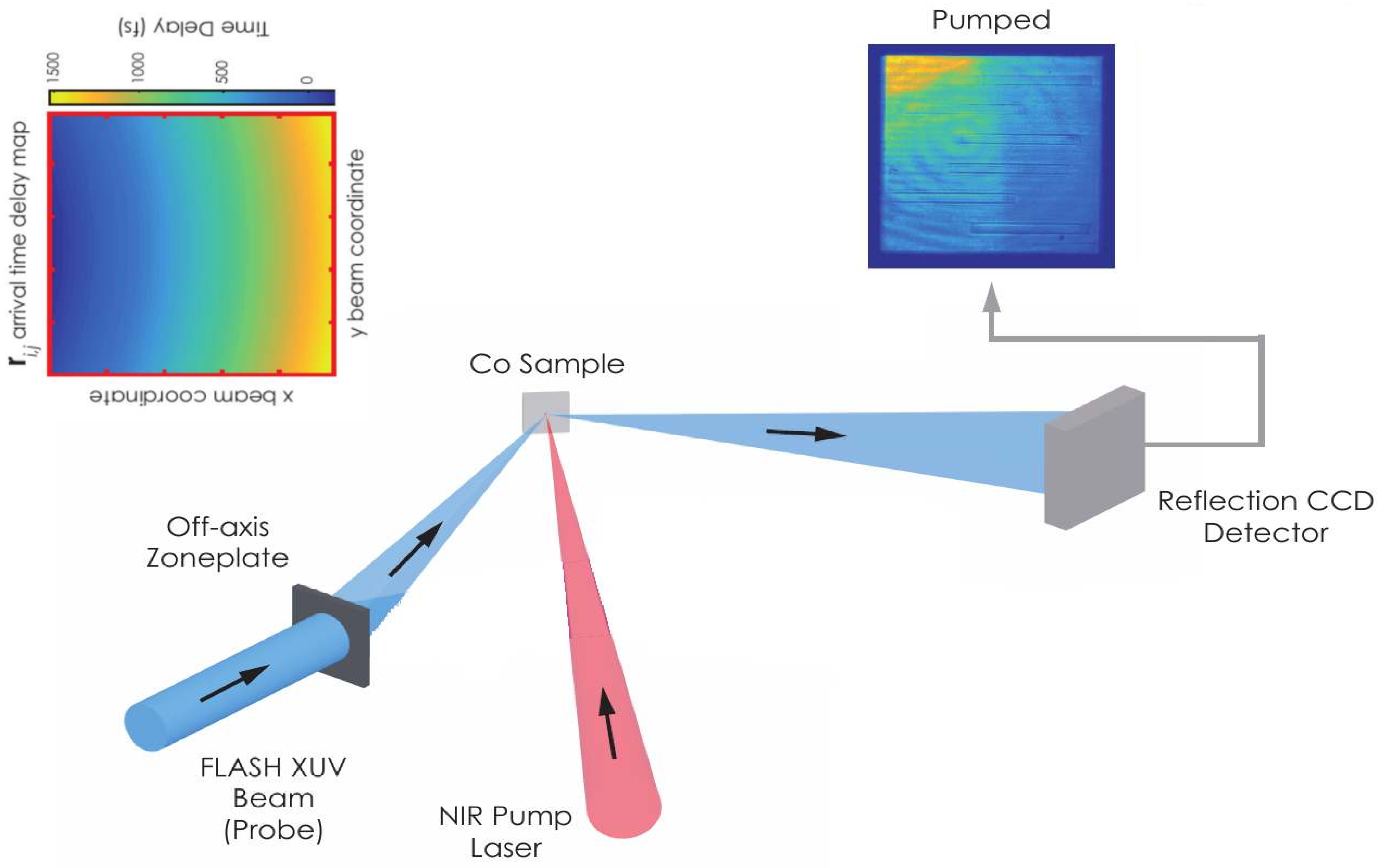

Tw

Mi
Re

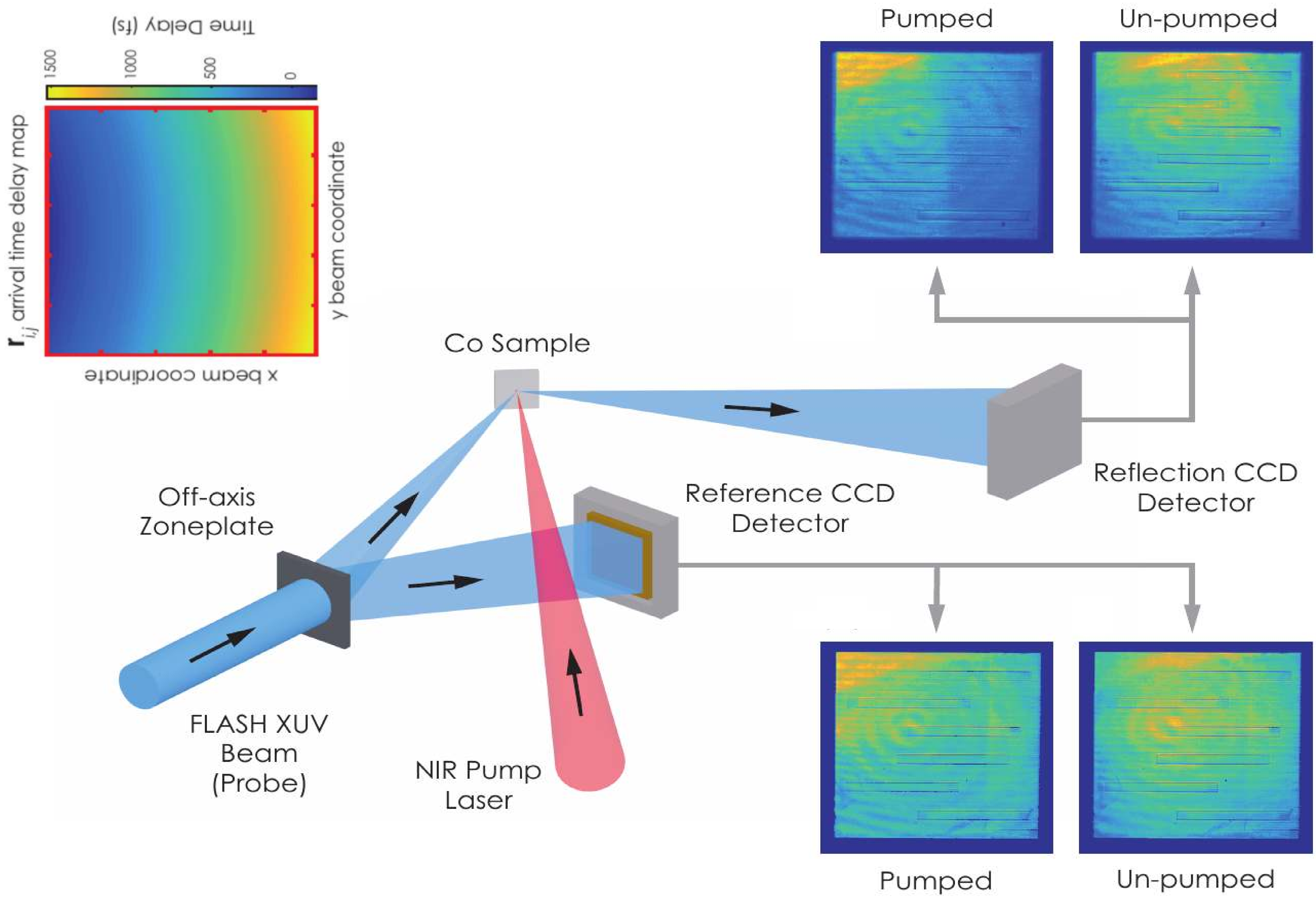
Mi
De

Ex
C.

# Snapshot streaking of ultrafast demagnetization dynamics

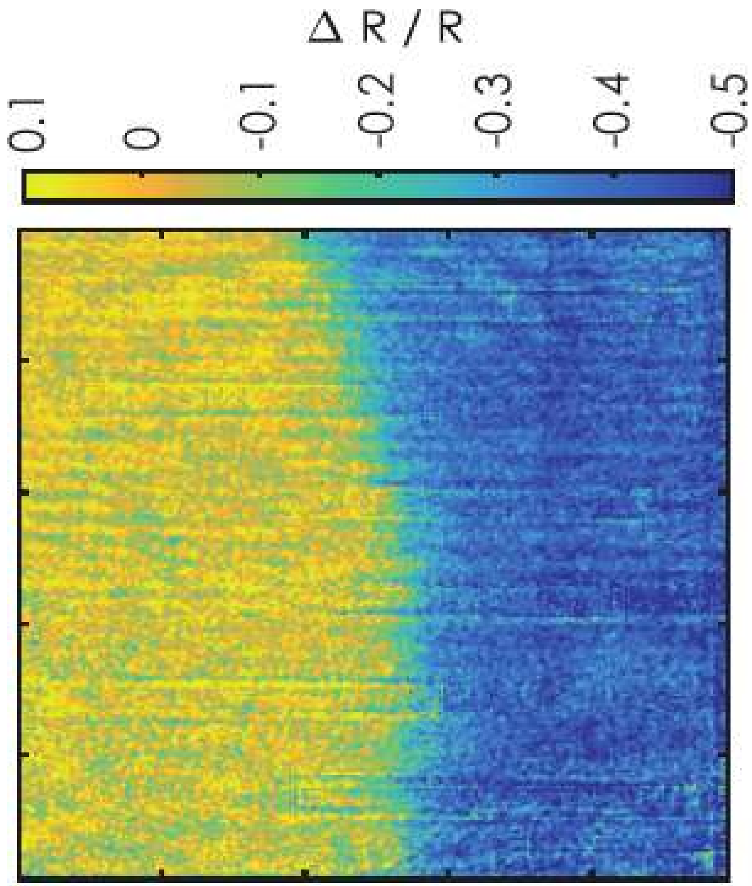


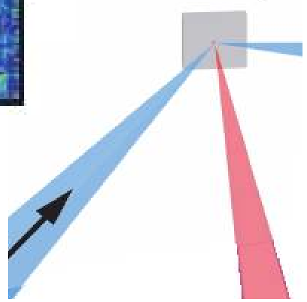
# Snapshot streaking of ultrafast demagnetization dynamics

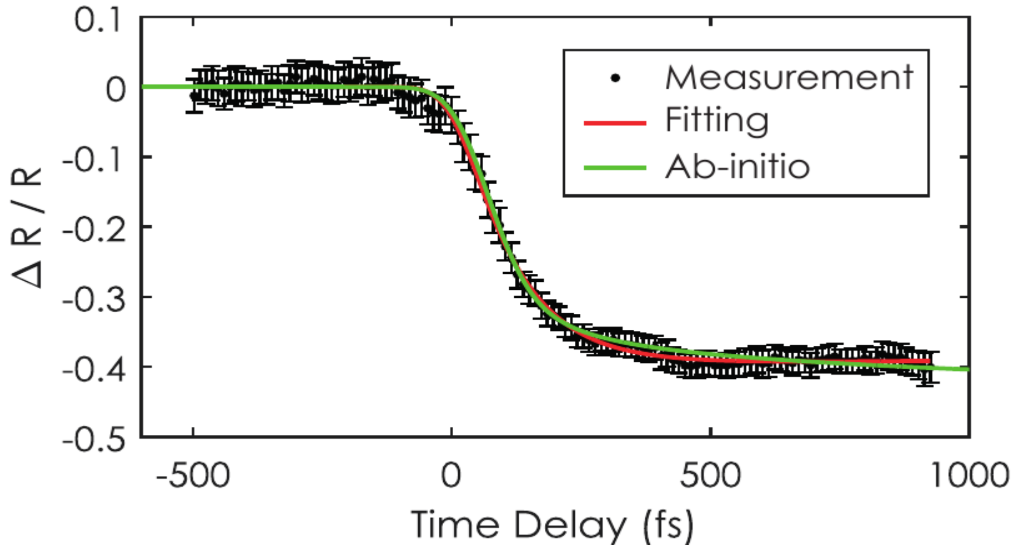


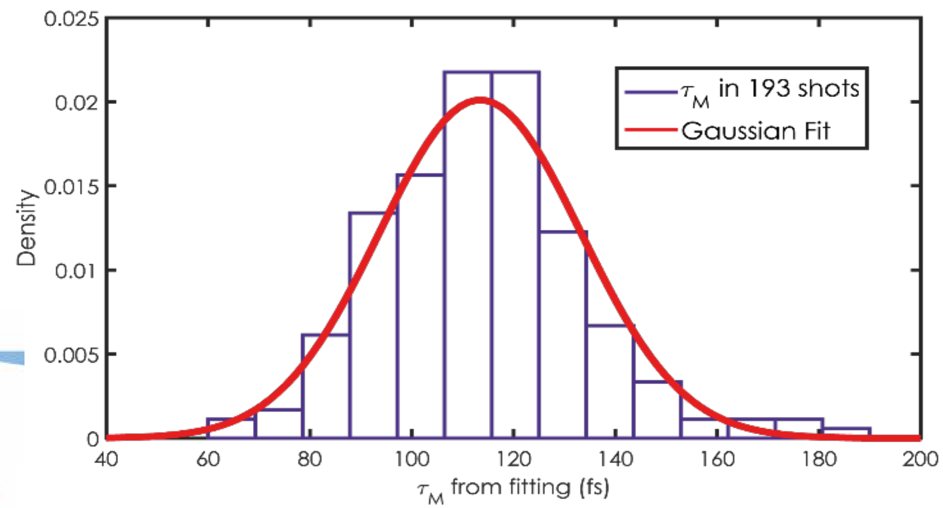
# Snapshot streaking of ultrafast demagnetization dynamics

*BL3 @ FLASH*





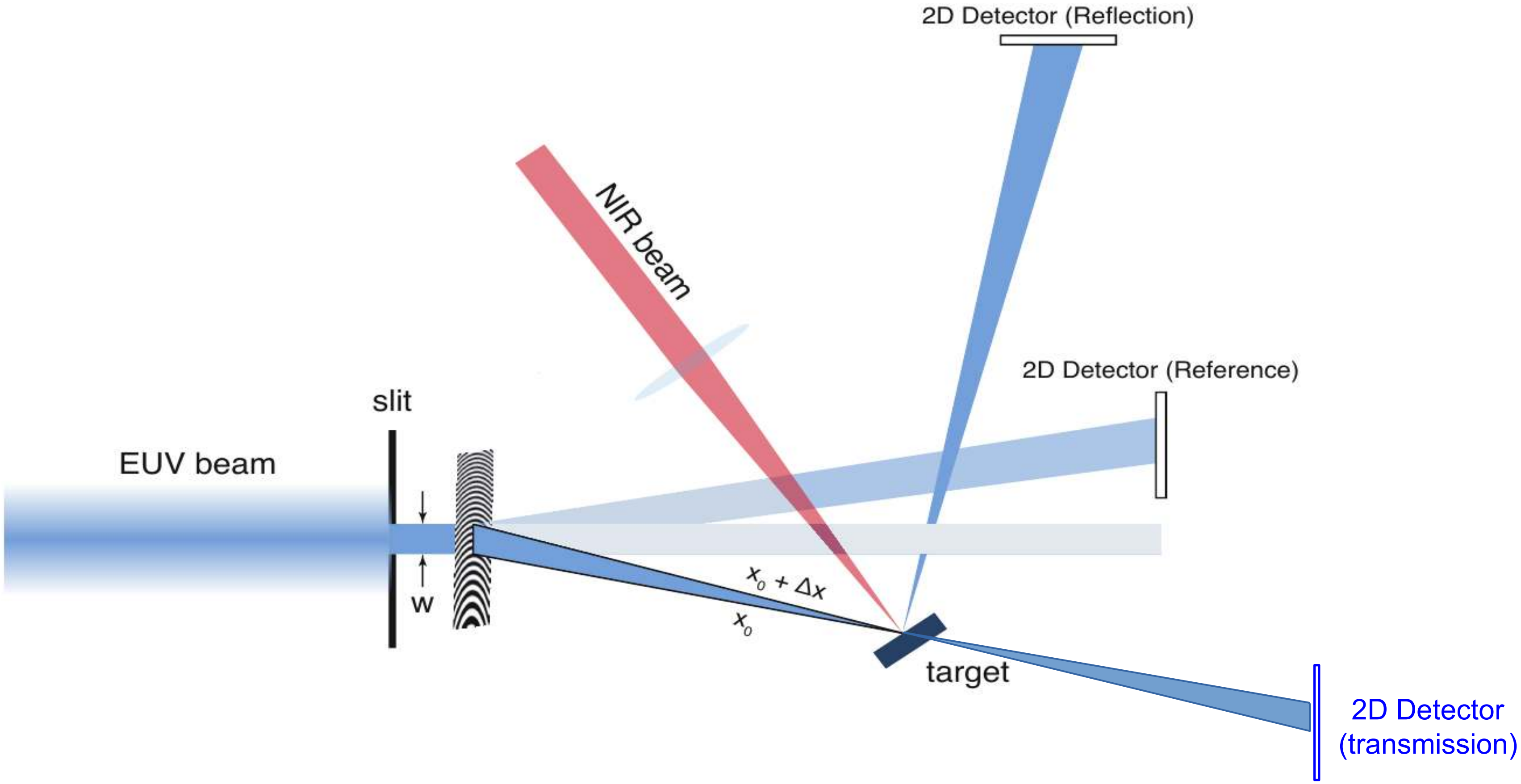




Time resolution today limited by IR pulse length

$\Rightarrow \tau_M$ = 113 fs ± 20 fs

# Reflectivity versus transmission geometry

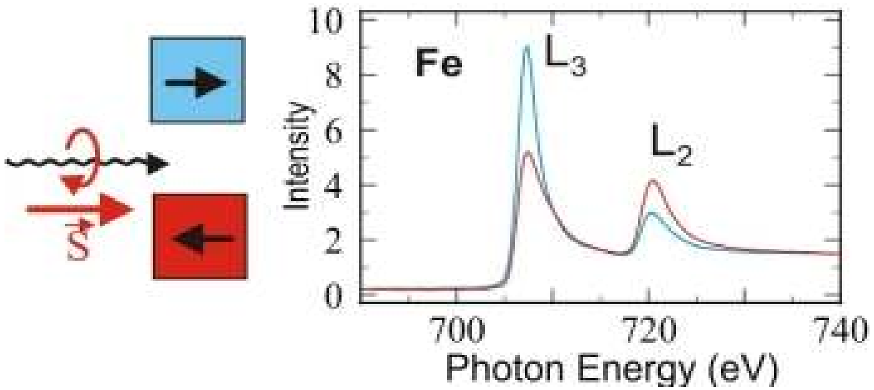
Reflectivity geometry limits applicability of technique
to other scientific domains




→ X-ray absorption spectroscopy in transmission geometry

# X-ray streaking at the seeded XUV-FEL FERMI

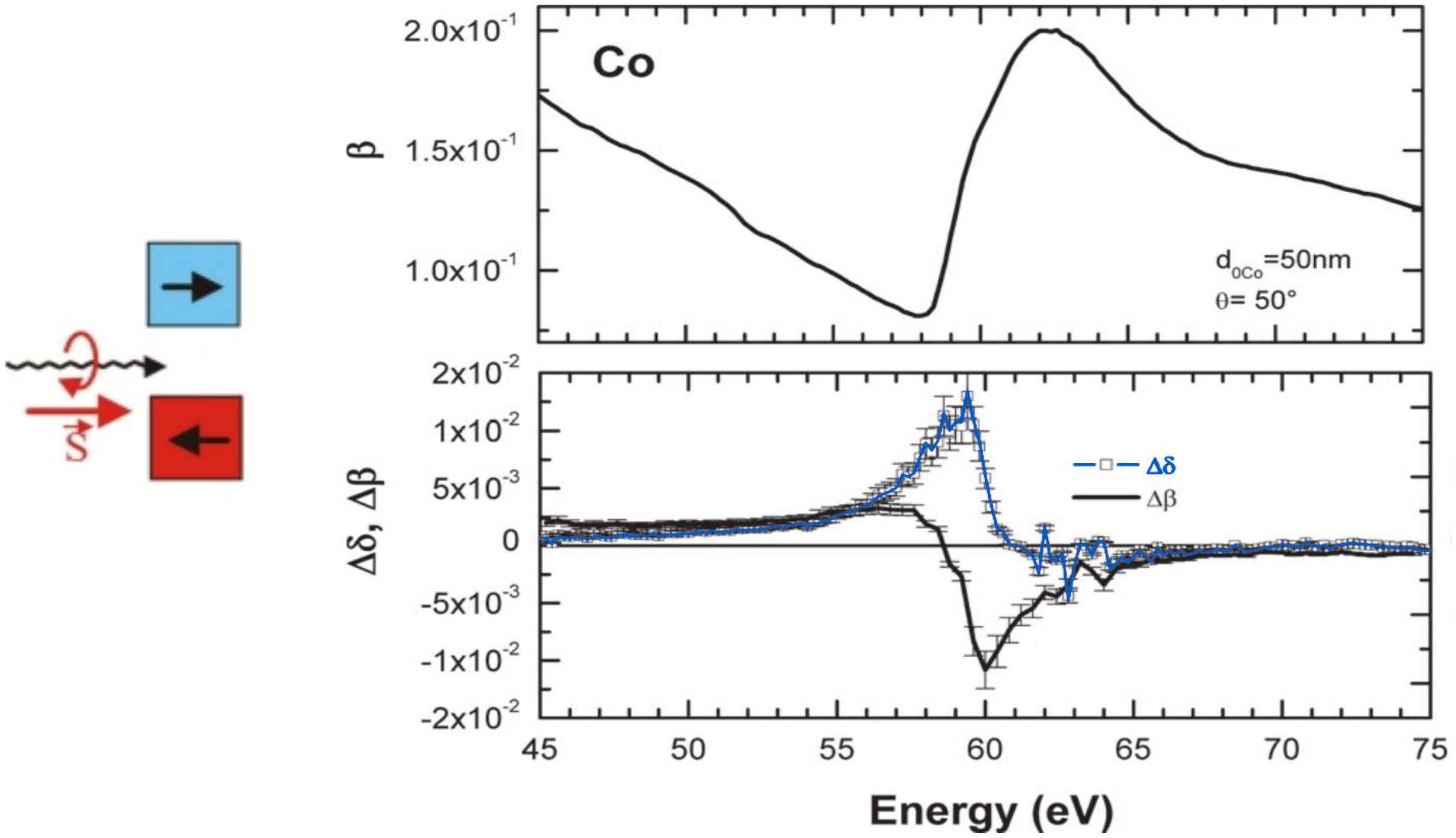
- Polarization control provides circularly polarized X-rays



# X-ray magnetic circular dichroism

FERMI XUV-FEL provides circularly polarized X-rays

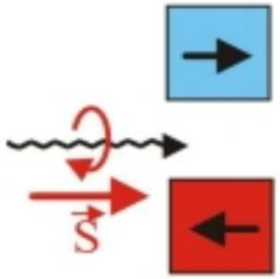
Co $M_{2,3}$ edge

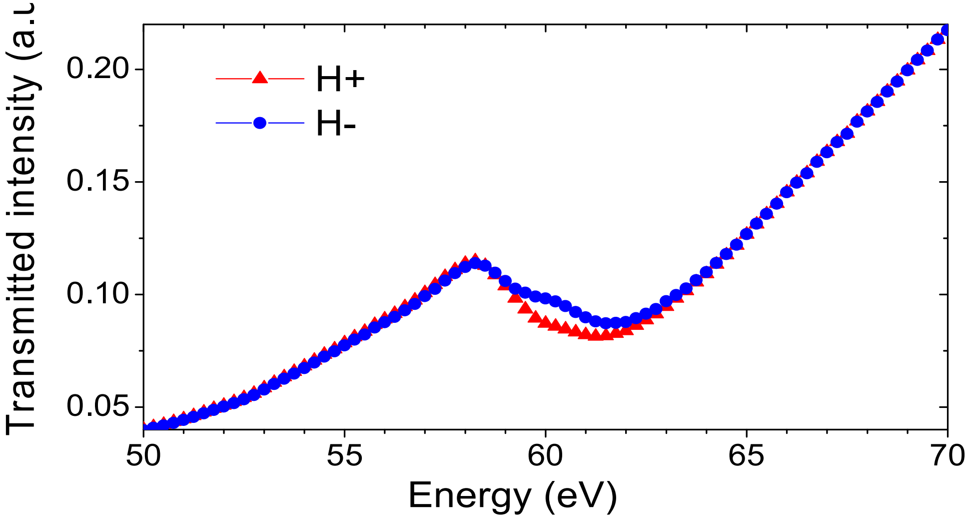



→ **weak** XMCD effect of **weak** resonance on **strong background**

# X-ray magnetic circular dichroism

Co $M_{2,3}$ edge



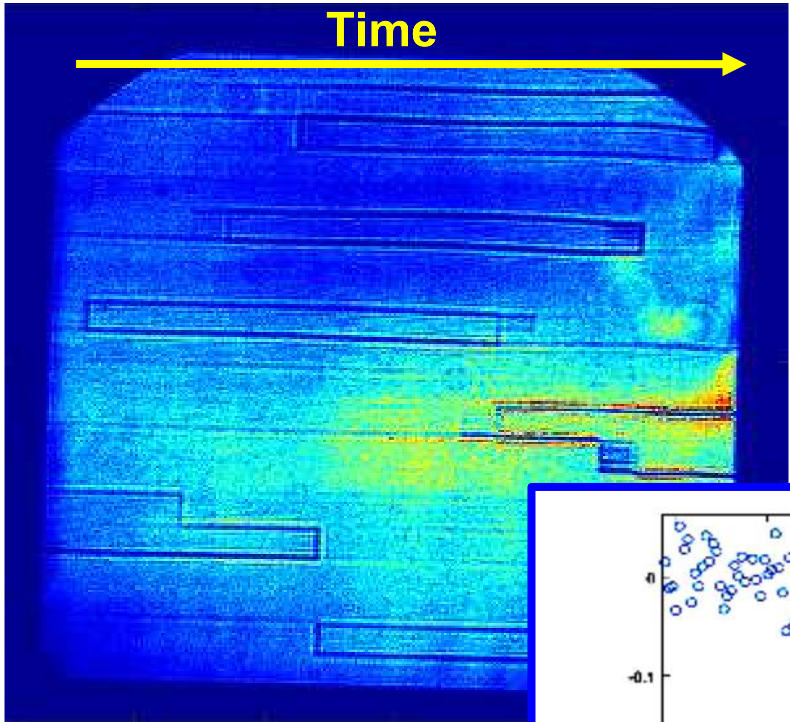




→ **weak** XMCD effect of **weak** resonance on **strong background**

# XMCD contrast evolution in transmission geometry



Transmission camera




Normalized image