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
Quantitative imaging with sensitivity to elemental and chemical distribution and charge/spin ordering
Motivation: Switching of magnetic memory cells (MRAM)
STXM image of spin injection structure

4 nm magnetic layer
buried in 250 nm of metals

~100 nm

Detector

leads for current pulses

Static images of the buried layer’s magnetization
Problem: Today not enough intensity for single shot experiments with nanometer spatial and picosecond time resolution.
Storage ring is filled with electron bunches → emission of X-ray pulses

Bunch spacing 2 ns

Bunch width ~ 50 ps

beam line

pulsed x-rays
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

Y. Acremann et al.,

= 950 Oersted
for 150x100nm,
j = 2x10^8 A/cm^2
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
Insertion devices of 3rd generation sources provide X-ray beams with:

- Flux: $10^{14}$ ph / (sec$\cdot$0.1% BW) → $10^6 - 10^8$ pulses / sec
- Brilliance:
  $10^{22}$ ph / (sec$\cdot$0.1%$\cdot$BW$\cdot$mrad$^2\cdot$mm$^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

LBNL/EXXON/SSRL (1982), SSRL Beamline VI
55 pole ($N = 27.5$), $\lambda_w = 7$ cm
fs pulsed X-ray sources

Combine nanometer spatial resolution with femtosecond temporal resolution

Femtoslicing (BESSY, SLS, SOLEIL) ~10^3 / 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!

\[ N_e \sim 10^9 \quad I \sim N_e \cdot N^2 \quad N \sim 10^2 \]
SASE-XFEL – a very long undulator

Coherent source → Intensity ~ (# of e⁻)²

FLASH (Hamburg)
- Built as the Tesla Test Facility

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

Soon: several FELs in China
X-ray Free Electron Lasers

- $\sim 10^{13}$ 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

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
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
1996: Discovery of ultrafast magnetization dynamics

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

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

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

→ 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**

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

Sample density \( \rho \)

**Small Angle Scattering**

\[ I_{cs} \sim I_o \Delta \sigma_{cs} / \rho \]

\[ \sigma_{cs} = c |f_1 + i f_2| \]

Sample density \( \rho \)

Data from Jeff Kortright (LBNL)
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 scattering object

Photon energy (eV)

Transmission

Co L₃ XMCD

Below Resonance

On Resonance

\( \lambda = 1.59 \text{ nm}, 2.5 \text{ mm } \odot \text{ Pinhole} \)

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: \( \text{L}_{2,3} \) (700 – 850 eV)
- FLASH, FERMI (HHG): \( \text{M}_{2,3} \) (55 - 65 eV ↔ 37\textsuperscript{th} – 41\textsuperscript{st} 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
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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**Experimental setup**

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)
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 ↔ 37th – 41st 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

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

Convolution theorem applied to diffraction

\[(a \otimes a) = \text{FT}^{-1} \{\text{FT}(a) \cdot \text{FT}(a)\} \]
Fourier transform X-ray spectro-holography
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

SEM

1.5 μm

1 μm gold

100nm silicon nitride

Magnetic multilayer

100nm
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

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

~ 50 nm spatial resolution
~ < 80 fs temporal resolution

X-ray induced “modifications”


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

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

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

Imaging ultrafast demagnetization dynamics after a spatially localized optical excitation


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


15 hard X-ray probe beams

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

\[ \Delta X = N_{\text{Zones}} \cdot \lambda \]

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
Time resolution today limited by IR pulse length

\[ \tau_M = 113 \text{ fs} \pm 20 \text{ fs} \]
Reflectivity geometry limits applicability of technique to other scientific domains

→ X-ray absorption spectroscopy in transmission geometry
• Polarization control provides circularly polarized X-rays
X-ray magnetic circular dichroism

FERMI XUV-FEL provides circularly polarized X-rays

→ 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

Time

$\Delta \text{MM (a.u.)}$

Time (fs)