Magnetization dynamics and fs pulsed X-ray sources

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

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Synchrotron SOLEIL
fs pulsed X-ray sources

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

**HHG**
\[ \sim 10^5 \text{ photons/pulse on sample} \]

**Femtoslicing (BESSY, SLS, **SOLEIL**)**
\[ \sim 10^6 \text{ photons/pulse on sample} \]

**FLASH / LCLS / FERMI / SACLA**
\[ \sim 10^{12} \text{ photons/pulse on sample} \]
Magnetization reversal implies change of (orbital and spin) angular momentum!
Transfer to lattice via magnon-phonon scattering!
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

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

\[ H(t) \]

\[ \tau \sim 100's \text{ fs} \]

\[ \tau \sim 1-10 \text{ ps} \quad (TM) \]

TM : \( m_i \approx 0 \)

Laser

3 temperature model

Electrons

Phonons

Spin

H(t)_{ext}

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

QUESTION: Could we manipulate magnetization with **only** a laser pulse?
1992: Spin-lattice coupling in Gd ~100 ps

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

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

Photoelectron spin polarization

60 ps laser pulses

Electrons

Phonons

Spin

τ ~ 100 ps (Gd)
1996: Discovery of ultrafast magnetization dynamics

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

\[ \tau \sim 1 - 10 \text{ ps} \]
1996: Discovery of ultrafast magnetization dynamics

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 ….

\[ \tau \sim 1 - 10 \text{ ps} \]

\[ \text{Normalized remanence} \]

\[ \text{\Delta t (ps)} \]

\[ \text{E. Baurepaire et al.} \]
\[ \text{PRL 76, 4250 (1996)} \]

… to ultrafast magnetization CONTROL

\[ \text{C.D. Stanciu et al., PRL 99, 047601 (2007)} \]
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
- Strong dichroism signal

→ properties of X-ray based techniques
Insertion devices of 3\textsuperscript{rd} generation sources provide X-ray beams with:

- **Flux:** $10^{14}$ ph / (sec$\cdot$0.1\% BW)
  $\rightarrow$ $10^6$ – $10^8$ pulses / sec

- **Brilliance:**
  $10^{22}$ ph / (sec$\cdot$0.1\%$\cdot$BW$\cdot$mrad$^2\cdot$mm$^2$)
  $\rightarrow$ low coherence degree (deg. < 1)

- **Polarization control**

- **Time structure:**
  $\sim$50 ps X-ray flashes, ns-$\mu$s spacing
  with few photons:
  - few ps in low-alpha
  - $\sim$150 fs in femtoslicing
  $\rightarrow$ inadequate for fs dynamics
Synchrotron radiation of an undulator

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

\[ N_e \sim 10^9 \quad I \sim N_e \cdot N^2 \quad N \sim 10^2 \]
Self Amplified Spontaneous Emission (SASE XFEL)

Spontaneous radiation

\[ I \sim (N_e) \cdot (N_u)^2 \]

Coherent radiation

\[ I \sim (N_e)^2 \cdot (N_u)^2 \]

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

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

\( E_x(t), v_x(t) \) const. \( \Rightarrow \) acceleration & deceleration
SASE-XFEL – a very long undulator

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

FLASH (Hamburg) • Built as the Tesla Test Facility
Successive accelerator upgrades (2000 – 2011) pushed shortest wavelength to 4.1 nm (300 eV)
2005: User facility FLASH
Today: FLASH, LCLS, SACLA, FERMI
Soon: E-XFEL, Swiss-FEL, Pohang
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
- ...

![Diagram showing peak brilliance vs energy for various X-ray sources, including XFEL, FLASh, PETRA III, BESSY II, and others.](image-url)
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

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Resonant scattering for local probing of magnetization

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

**Experimental setup**

- Co/Pd
- Beam stop
- IR shield (Al film)
- CCD

Side view: [Co 0.4 nm / Pd 0.8 nm] \times 30 \ldots
X-ray Magnetic Circular Dichroism in Absorption / Scattering

Absorption

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

Sample density \( \rho \)

Small Angle Scattering

\[ I_{cs} = I_o \frac{\sigma_{cs}}{\sigma} \]

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

Data from Jeff Kortright (LBNL)
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)

Experimental setup

Integrated intensity → measure of the local magnetization
Is direct photon excitation and presence of hot electrons needed?

Add 40 nm Alu cap layer to convert IR photons in avalanche of excited valence electrons.
Hot electron excited ultrafast magnetization dynamics

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:
Resonant scattering for local probing of magnetization

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

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→ *combines femtosecond temporal with nanometer spatial resolution*

**Experimental setup**

- X-ray
- IR
- Co/Pd
- Beam stop
- IR shield (Al film)
- CCD

Integrated intensity → measure of the local magnetization

Form of scattering pattern → spatial information
UF demagnetization in the high IR pump power limit


- FLASH -

The diagram shows a plot of normalized intensity against q (μm⁻¹). There is also a graph indicating the wall profile (nm) with M/Mₐ on the y-axis.
Angular momentum transport by hot polarized electrons


Monte Carlo simulation

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

Probing domain wall evolution directly

Higher scattering orders → more detailed insight in magnetic domain structure


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**: \( \text{L}_{2,3} (700 – 850 \text{ eV}) \)
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**Experimental setup**

- **X-ray**
- **IR**
- **Co/Pd**
- **Beam stop**
- **IR shield (Al film)**
- **CCD**

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:

\[ I_{p,q} = |M_{p,q}| \cdot e^{-i\phi_{p,q}}^2 \]
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
Imaging ultrafast demagnetization dynamics after a spatially localized optical excitation

Imaging ultrafast demagnetization dynamics after a spatially localized optical excitation

Signature of hot electron diffusion

NOTE: These are not single shot images!
The Linac Coherent Light Source at SLAC

- $10^{13}$ photons/pulse
- fs 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


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”


• 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!
Basic idea:

\[ \Delta x + x \]

Arrival time encoded in angular direction
Snapshot streaking of ultrafast demagnetization dynamics

Image of streaked single X-ray pulse

1st ZP dimension

Time axis

2nd ZP dimension

incidence angle

BL3 @ FLASH

ΔR/R

Time Delay (fs)

1 Shot demagnetisation curve
2 exponential fit

ΔR/R

Demagnetisation Time $\tau_M$

(130 ± 20) fs

$\tau_M$ in 200 shots
Gaussian Fit
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
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)

FLASH X-ray pulse: 100 fs, 60 eV (Co M₃)
Sample: (Co/Pt) multilayer on SiN membrane

Breakdown of the Resonant Magnetic Scattering Signal

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
Ultrafast evolution of the atomic 'oxidation' state

- 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!

R. Santra (CFEL)

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

2) stimulation of competing processes → reduction in yield of resonant magn. scat.
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