We want to study the exchange of energy and angular momentum between thermodynamic reservoirs!

- Optical excitation
- \( \tau_e < 0.1 \text{ps} \)
- Quenched Spin-Orbit coupling
  \( \zeta \sim \text{few } \mu\text{eV} \rightarrow \tau_{ls} \sim 100 \text{ ps} \)
- "bottleneck"
- "Atomic" Spin-Orbit coupling
  \( \zeta \sim \text{few meV} \rightarrow \tau_{es} \)?
Flow of Angular Momentum

Needed:
1. Information about the flow of spin and orbital momentum
2. Ultrafast time resolution
Conventional magnetic recording:
Magnetization reversal by *spin precession* and *damping*

Dream: *Fs laser* pulses *optically* induce magnetization reversal (ultrafast?)
Beamline & Experimental Techniques

- Beamline 11.0.1
- X-ray Magnetic Circular Dichroism
- Ultrafast Measurements with a Streak Camera

Normalized absorption

Normalized absorption vs photon energy (eV)
Ultrafast Magnetization End Station at BL 11.0.1
An Apple-II type Elliptically Polarizing Undulator (EPU) for full control of the x-ray polarization.

A Variable Line Space (VLS) grating design is used to produce moderate resolving power with a high efficiency.
The optical design incorporates several bendable and retractable mirrors, yielding a beamline which can be used for both microscopy (PEEM) and ultrafast magnetization research.
The experiment resides in a laser safety enclosure. . .

. . . which houses a 60 fs Ti:Sapphire laser oscillator/amplifier. . .

. . . and optics to trigger the camera, produce uv fiducialization pulses, and initiate the magnetic transitions.
Alternating the polarization (or the anisotropy) causes the x-ray absorption spectrum to vary.
• Changing the polarization of the x-rays from right to left elliptical polarization yields the x-ray magnetic circular dichroism (XMCD) spectrum.

• Shown here are the intensity-normalized spectra of the Gd M-edges at 1200 eV. Below is the difference spectrum.

• The beamline energy range allow measurement of rare earth and transition metal elements.
Circular Dichroism Measurements

XMCD is element specific and can measure complicated structures and compositions.
MCD Sum Rules Allow a Quantitative Analysis

\[ m_{\text{orb}} = -\frac{4}{3} \frac{\int_{L_3 + L_2} (\mu_+ - \mu_-) \, d\omega}{\int_{L_3 + L_2} (\mu_+ + \mu_-) \, d\omega} (10 - n_{3d}) \]

\[ m_{\text{spin}} = -\frac{6}{\int_{L_3} (\mu_+ - \mu_-) \, d\omega - 4 \int_{L_3 + L_2} (\mu_+ - \mu_-) \, d\omega} \frac{7\langle T_z \rangle}{\frac{2}{2} \langle S_z \rangle} \times (10 - n_{3d}) \left(1 + \frac{7\langle T_z \rangle}{\frac{2}{2} \langle S_z \rangle}\right)^{-1}, \quad (2) \]

Using the absorption and MCD spectra, the orbital and spin components of the magnetic moment can be determined.

P. Carra, B.T. Thole, M. Alterelli, and X. Wang PRL 70, 694 (1993)
For a ferromagnet, we get a different absorption for left or right circularly polarized light.

Integrals over the dichroism and absorption spectra (q, p & r) reveal the spin and orbital moments separately per atom.
Time Structure of ALS x-rays

- Multibunch operation with $\approx 1.5$ mA per bunch
- 2-bunch operation available with $\approx 30$ mA per bunch
- Streak camera synchronizes on one pulse and resolves its temporal structure.
Streak Camera Experiment at BL11.0.1

- X-ray intensity is time resolved by the detector
- Time resolution limited by the streak camera
Current Streak camera operates in normal incidence
Time-resolution approximately 1 pS
The x-rays produce a train of electrons, which are then accelerated by the cathode potential and deflected by the sweep plates to the observation plane.
How does a streak camera work?

Each electron sees a different sweep voltage...
How does a streak camera work?

Photocathode (-10 kV)

X-rays → Photocathode

Photoelectrons

Sweep Voltage $V(t)$ → Time

$V(t)$ so is deflected to a different part of the detector plane.
How does a streak camera work?

In this way, the temporal characteristics of the electron pulse. . .
How does a streak camera work?

Photocathode (-10 kV)

X-rays

Photoelectrons

Sweep Voltage $V(t)$

time

... are translated into a spatial dispersion ...
How does a streak camera work?

Photocathode (-10 kV)

X-rays

Photoelectrons

Sweep Voltage $V(t)$

(time)

$V(t)$

. . . which can then be recorded with a spatially sensitive electron detector.
Alternatively, the electrons can hit a phosphor, and the resulting visible fluorescence can be detected with a CCD camera.
Entire 70 pS fwhm captured with every laser shot
- 5 kHz laser repetition rate
- 500 MHz x-ray pulse repetition rate
- Streaks are currently averaged over ten’s of seconds
- UV generated by frequency tripling the femtosecond laser pulses provide resolution measurement and jitter compensation
Scientific Applications of Ultrafast Magnetic Measurements

- Dynamics of FeGd Multilayers
- Demagnetization of a Heusler Alloy

Graph showing XMCD (a.u.) vs. Photon energy (eV) and Spin moment (a.u.) for different times (ps).
Example-FeGd Multilayers

- Samples are transmission
- Mounted on SiN windows
- Variety of samples measured thin films of Ni and other transition metals, also more complicated materials

Sample from T. Eimuller, Ruhr-Universität Bochum
Ultrafast Demagnetization

60fs pump pulse

- Normalized transmitted intensity $\sigma^-$
- Normalized transmitted intensity $\sigma^+$
- Normalized dichroism

Dichroic contrast is lost in a few pico-seconds
Fast Demagnetization - FeGd Multilayers

- Demagnetization happens in less than 4 ps.

**FeGd**
- Magnetic contrast on L3 peak
- Demagnetization happens in less than 4 ps.

Temporal resolution indicated by broadening of 60fs UV pulses.
Each line in this image is from a separate streak at a different energy.

Time-resolved absorption spectra obtained by combining streaks at different photon energies.

Gd edges

Streak at one energy

H = +20 mT
Right photon polarization

M₄
M₅
From Streaks to Time Resolved XMCD Spectra

FeGd sample, Fe edge

H,σ parallel

H,σ antiparallel

difference
XMCD signal vanished in a few picoseconds
Heusler Alloys

- Materials of formula $X_2YM$ and L2$_1$ structures
- Half metallic, may be good candidates for spin injection
- Cubic and tetragonal unit cells
Half Metals—Slower dynamics??

Ultrafast Spin Dynamics and Critical Behavior in Half-Metallic Ferromagnet: \( \text{Sr}_2\text{FeMoO}_6 \)

T. Kise,\(^1\) T. Ogasawara,\(^1\) M. Ashida,\(^2\) Y. Tomioka,\(^3\) Y. Tokura,\(^{1,3}\) and M. Kuwata-Gonokami\(^{1,2,*}\)
Measurement of Heusler Alloy

\[ \tau = 1.2 \pm 0.6 \text{ ps} \]
- Future plans include the installation of an improved streak camera, featuring:
  • Photoelectron gating
  • Improved photocathode

- Improvements to the end station will included a variable temperature sample manipulator

- Beamline improvements will included:
  • Chopper to reduce extraneous x-rays
  • Improvements in spectral resolving power and spot size to match temporal resolution
We want to study the exchange of **energy** and **angular momentum** between thermodynamic reservoirs!

- Optical excitation
- \( \tau_e < 0.1 \text{ps} \)
- Electrons
  - non-thermal
  - \( \tau_e \)
  - \( \tau_{el} \approx 1 \text{ ps} \)
- Lattice
  - \( \tau_l = 1 \text{ ps} \)
- Spin
  - \( \tau_S \)
  - Quenched Spin-Orbit coupling
  - \( \zeta \approx \text{few } \mu\text{eV} \)
  - \( \tau_{ls} \approx 100 \text{ ps} \)
  - “Atomic” Spin-Orbit coupling
  - \( \zeta \approx \text{few meV} \)
  - \( \tau_{es} \)?
Brief Overview

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Time-Resolved Absorption Spectra

H = +20 mT
Right photon polarization

Time-resolved absorption spectra obtained by combining streaks at different photon energies

Each line in this image is from a separate streak at a different energy
From Streaks to Time Resolved XMCD Spectra

H,σ parallel

H,σ antiparallel

FeGd sample, Fe edge

difference

hν

time
Ultrafast Demagnetization

60fs pump pulse

Normalized transmitted intensity $\sigma^-$

Normalized transmitted intensity $\sigma^+$

Normalized dichroism

X-Rays

dichroic contrast is lost in a few pico-seconds