The PEEM-3 Photoemission Electron Microscope at the Advanced Light Source

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- Introduction
- Advanced Light Source PEEM-3
- Cryo-PEEM
- X-ray Linear Dichroism (XLD)
- Examples
Ultrafast Dynamics and Nano Science

Ultra-Small

Nature
- Flea
- Human hair ~30 μm wide
- Red blood cells & white cell ~ 5μm
- Virus ~ 200 nm
- DNA helix ~3 nm width
- Water molecule
- Atom

Technology
- Head of a pin ~ 1mm
- Micro gears 10 - 100 μm diameter
- DVD track
- 1 μm Electrodes connected with nanotubes
- Carbon nanotube ~ 2nm diameter
- Atomic corral ~ 14 nm diameter

The Microworld

Ultra-Fast

Nature
- Hydrogen transfer time in molecules is ~ 1ns
- Spin precesses in 1 Tesla field is 10 ps
- Shock wave propagates by 1 atom in ~ 100 fs
- Water dissociates in ~10 fs
- Bohr period of valence electron is ~ 1 fs

Technology
- Computing time per bit is ~ 1 ns
- Optical network switching time per bit is ~ 100 ps
- Magnetic recording time per bit is ~ 2 ns
- Laser pulsed current switch ~ 1ps
- Shortest laser pulse is ~ 1 fs
- Oscillation period of visible light is ~ 1 fs

J. Stöhr, SLAC 2005
Applications of X-PEEM

Main applications of X-PEEM that differentiate it from TXM techniques:

• Magnetism, structure, electronic properties of single crystalline, epitaxial materials
• Study of surface properties, ultrathin films, growth, catalysis.
• Electronic structure using (AR)PES.
• Fast surveying at high resolution, time domain imaging.
• Cryogenic PEEM of surfaces.
• Properties of nanostructures requiring thick supports.

Other applications:

• Nanostructures (magnetism, composition, etc.)
• Ultrafast dynamics, pump probe studies
ALS PEEM-3 User Program

Distribution of beamtime proposals Spring 2011 (out of 31).

<table>
<thead>
<tr>
<th>Category</th>
<th>Count</th>
</tr>
</thead>
<tbody>
<tr>
<td>Materials – Magnetism</td>
<td>19</td>
</tr>
<tr>
<td>Materials – not Magnetism</td>
<td>12</td>
</tr>
<tr>
<td>Ferroelectric, Multiferroics</td>
<td>5</td>
</tr>
<tr>
<td>Nanostructures</td>
<td>6</td>
</tr>
<tr>
<td>Cryogenic PEEM</td>
<td>7</td>
</tr>
<tr>
<td>Time-resolved PEEM</td>
<td>3</td>
</tr>
</tbody>
</table>
• Variable linear and circular polarization
• High flux density using microfocusing (down to 10x10 microns)
• VLS plane grating monochromator
• Energy range 160 to 2000 eV with E/dE~4000

Undulator, monochromator are shared. PEEM has about 60% of the beamtime.
Advanced Light Source: PEEM-3

- Large machine with room inside to grow
- Fully automated sample transfer system

Beamline staff:
T. Young, A. Doran, A. Scholl
### A Look Inside the Microscope

**Components:***
- **Single Straight Electron Column**
- **Objective Lens**
- **X-rays**
- **Sample Stage**
- **CCD**
- **Space for Mirror & Separator**
- **Optics laid out on an in-vacuum optical table.**
- **Facilitates easy upgrades and modifications.**

**Features:**
- **Triple layer Mu metal shields**

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*Image description:*
- The image shows a detailed view of the microscope with labels indicating various components and features.
- The microscope includes a single straight electron column, an objective lens, and a sample stage, along with x-rays and a CCD camera.
- The space for the mirror and separator is also visible, along with triple-layer mu metal shields.
- The optics are laid out on an in-vacuum optical table, facilitating easy upgrades and modifications.
Without aberration correction
Element-resolved X-PEEM image of 25 nm thick Cr/Si layers.

Aberration correction:
4-fold improvement of spatial resolution at a few % transmission (goal 10 nm)
10-fold improvement of transmission at several 10 nanometer spatial resolution.
Staged Buildup of Aberration Corrected Endstation

PEEM2.5 Mode
Now

PEEM3 Mode

Sample

Addition of Dipole Separator and Mirror Section results in aberration correction and improved resolution

Electrostatic dodecapole

Electrostatic Quadrupole

Dipole separator

Electrostatic mirror

CCD
Mechanical design complete, contingent on funding the corrector will be added in 2011.
Liquid He-Cooled Sample Manipulator and Cartridges

6-strut 5-axis manipulator using fast PI stages and flexural joints.

Large, easy to modify sample cartridges, suitable for fully automatic transfers.
Cooling Performance

Spring-loaded Cu clamps grab a polished Si nose mounted on a massive Cu sample cover. A one inch sapphire electrically isolates the sample. Co braids are used for vibration isolation.

Temperature as function of time using liquid He cooling.

Lowest temperature reached: 16 K.
Applications of Cryo-PEEM

- Antiferromagnetism of high Tc superconductors (pnictide)
- Antiferromagnetism and charge, orbital order in CMR materials
- Antiferromagnetic vortices in CoO, NiO
- **Interface exchange coupling in perovskite FM/AFMs**

- Dilute magnetic semiconductors
- Ferroelectrics, multiferroics
Total Electron Yield

(a) Transmission

\[ I_t = I_0 e^{-\mu x d} \]

Sample

(b) Electron yield

\[ I_e \sim I_0 \mu_x \]

J. Stohr, H. Siegmann: From Fundamentals to Nanoscale Dynamics
$\Delta I_{XLD} \propto I_{\text{charge}} Q_{zz} (1 - 3 \cos^2(\theta)) + I_{\text{magn}} \langle M^2 \rangle (1 - 3 \cos^2(\varphi))$

Magnetic and charge contribution need to be separated.
Linear Dichroism of a Partially Filled d-Electron System

C. T. Chen et al. PRL 68, 2543 (1992)
X-ray Magnetic Linear Dichroism (XMLD)

Ni$^{2+}$ in NiO

Angle dependence $\sim \cos^2 \theta$
XMCD and XMLD Imaging of Ferromagnets and Antiferromagnets

Spectroscopy

- Co
- LaFeO$_3$

Photon energy (eV)

TEY (a.u.)

F. Nolting et al., Nature 2000

Microscopy

- XMCD
- XMLD

F. Nolting et al., Nature 2000
Competing Interactions in FM/AFM Perovskite Superlattices

LSFO/LSMO multilayers (70/30)

F. Yang, Y. Takamura, et al., UC Davis

Spin flop coupling (BL 4, ALS)

Angle Dependent XMLD Contrast at Room Temperature

Xrays $\parallel$ <100>  
LSMO is paramagnetic

Experimental and simulated contrast

Xrays $\parallel$ <110>  
LSFO is antiferromagnetic

AFM axis is along the in-plane <100> directions.
Neither in a {111} plane (easy plane of LSFO)
Nor along <110> direction (easy axis of LSMO)
• Domain correlation and spin-flop coupling.
• Both LSMO and LSFO magnetic moments align along $<100>$. 
Magnetic Domain Formation in LSMO/LSFO multilayers

- Fe AFM are mobile while cooling through the LSMO Curie temperature.
- Crystal twin domains strongly pin AFM domains in thick single layers.
- Formation of large regions with preferred axis below the STO cubic to tretragonal transition at about 105 K.
X-ray Linear Dichroism Imaging

• Structure and formation of bio-minerals
  Gilbert group, U Wisconsin

• BiFeO$_3$, a room temperature multiferroic material
  R. Ramesh group, UC Berkeley
Linear Dichroism Study of Aragonite

R. Metzler, D. Zhou, M. Abrecht, S. Coppersmith, P. Gilbert
University of Wisconsin

Nacre: 95% aragonite (CaCO₃), + 5% organic matrix
3000x more resistant to fracture than aragonite

Goal:
- Determine the nacre microarchitecture (crystal orientation)
- Nacre formation mechanism

Light microscopy image of nacre.

x-ray linear dichroism as function of angle between linear x-ray polarization and the c-axis of aragonite.
X-ray linear dichroism (XLD) images at horizontal, vertical and diagonal polarization show the columnar growth and the crystal orientation of nacre, mother-of-pearl. The contrast varies with angle between the linear x-ray polarization direction and the stacking direction of the tablets.
High-resolution Ca and Mg mapping in the sea urchin tooth PIC map reveals that the plates (arrows) are slightly mis-oriented

Previous Ca and Mg mapping in sea urchin tooth from Robach et al. JSB 2006

(SIMS data, bars are 10 µm)
Electrical Control of Magnetism in Multiferroic Materials

Multiferroics: Materials with simultaneous (anti-) ferro-magnetic, ferroelectric and/or ferroelastic ordering

Coupling between Different Ordering Parameters

Intriguing Fundamental Science & Novel Design of Electronic Devices (i.e. Electrically Controllable Ferromagnet)

Measure the magnetization microscopically
Magnetic Structure of BiFeO₃ Films

BiFeO₃ - Ferroelectric antiferromagnet

$T_C \sim 1103K$

$T_N \sim 643K$

Magnetic plane is believed to be perpendicular to the polarization direction.

Ederer and Spaldin, PRB 71(2005)

This sample: 600 nm BiFeO₃ / SrRuO₃ / SrTiO₃
Polarization Direction in BiFeO$_3$(001)

Polarization points in one of 8 possible \{111\} directions.
X-ray Magnetic (Linear) Dichroism

Spectroscopy

Co
LaFeO₃

Microscopy

XMCD
XMLD

F. Nolting et al., Nature 2000
Domain Structure of BiFeO₃ Imaged Using X-ray Linear Dichroism

- x-ray linear dichroism at Fe L₂ edge
- contrast reversal proves dichroic origin
- images match PFM images of FE structure
- magnetic or charge dichroism?

Natural Linear Dichroism in PbZrTiO$_3$

Charge dichroism!
Horizontal “a” domains

Out-of-plane polarization contrast

C.S. Ganpule et al., JAP 91, 1477 (2002)

\[ \Delta z \approx t_d \times \tan^{-1}\left(\frac{c}{a} - 1\right) \sim 20\text{Å} \]

XLD is indeed sensitive to ferroelectric structure.
Temperature Dependence Shows Contrast Dominated By Magnetism

Temperature Dependence Shows Contrast Dominated By Magnetism
Determine Antiferromagnetic Axis of BiFeO$_3$

Six unique low index axes in (111) plane.

M. Holcomb et al., PRB 2010
Polarization Angle Dependence

~5 um FOV

XLD Images

M. Holcomb et al., PRB 2010
Modeling the Angular Dependence

Dichroic Contrast

Polarization

Model (112 or 110 axis)

M. Holcomb et al., PRB 2010
Antiferromagnetic Axis


\[ \Delta I_{XLD} \propto I_{\text{charge}} Q_{zz} (1 - 3 \cos^2(\theta)) + I_{\text{magn}} \left\langle M^2 \right\rangle (1 - 3 \cos^2(\varphi)) \]

- Sign can be positive or negative making both [110] and [112] possible solutions.
- However: temperature dependence changes with sign.
Temperature dependence favors [11-2] direction

M. Holcomb et al., PRB 2010
Summary

Photoemission Electron Microscopy

High spatial resolution

Cryogenic temperatures

XMCD, novel materials

XNLD

XMLD
The Future of X-PEEM

Concentrate on our strengths:
- Study of materials, surfaces, thin films.
- Cryogenic studies, electronic structure.
- Fast full field imaging.

Our next steps:
- Modest improvement in spatial resolution
- Large improvement in transmission
- Flexible sample environment
- Improved user-friendliness
STXM, PEEM or something else?

• Selection needs to be driven by the user community
• Should be a work-horse type experiment not a totally new concept with large risk

• PEEM and STXM communities have some overlap
  strength of PEEM in materials, epitaxial systems
  strength of STXM: very versatile, only drawback is the requirement in thickness

My recommendation:
To broaden the user community: STXM
To deepen and empower the current community: PEEM
SPEM: Need to go all the way towards meV energy resolution, competitive k resolution, cooling, sample prep. This is very challenging.
Do both but probably STXM first, if a strong user community exist.

Staffing and technical resources is an important concern. User support, and technical development is never finished.
The Future of X-PEEM

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- Study of materials, surfaces, thin films.
- Cryogenic studies, electronic structure.
- Fast full field imaging.

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Options for AC correction:
- Home-built, Specs, Elmitec
- Resolution: similar, sub 10 nm probably
- Sample environment: home built or a modified commercial PEEM has advantages
- User-friendliness: Requires software development beyond what vendors offer.