Scanning Transmission Soft X-ray Microscopy

Tolek Tyliszczak
Advanced Light Source
zone plates and achieved resolution

25 nm zone plate  
20 nm (vertical) Au lines

20 nm zone plate  
12 nm lines Si/Mo

17 nm zone plate  
10 nm lines Si/Mo

9 nm lines Si/Mo  
Imaged by 17 nm zone plate

All images taken at 700 eV
Structures have equal line/space nominal dimensions
Dimensions of half periods are quoted
Scanning Soft X-ray Microscopes have been operational at the ALS since 1997.

For magnetism and environmental science at 11.0.2 with 10nm resolution

For polymer chemistry at 5.3.2.2

“Classical” STXMs

For concrete chemistry and environmental science at 5.3.2.1 (commissioning)
3 ALS STXMs

- **Beamline 5.3.2.2 – “polymer STXM”**
  - Bending magnet
  - SGM 250 eV – 600 eV
  - Dedicated beamline

- **Beamline 11.0.2 – “MES STXM”**
  - Elliptically Polarizing Undulator
    - 80 eV – 2100 eV PGM
  - Possibility to scan the sample also at 30 deg to the beam
  - Time resolved measurements
  - External magnetic field

- **Beamline 5.3.2.1 – is being commissioned**
  - Bending magnet
  - SGM 250 eV – 2300 eV (possible 2600 eV)
  - Dedicated beamline
  - UHV
  - Scanned zone plate
  - Low temperature
75 - 2100 eV energy range
- K edge B, C, N, Na, Mg, Al
- L edge Al, Si, S, Ca, Ti, V, Cr, Fe, Ni, Cu, Ge, As, Se, Br
- M edge Br, Rb, Sr, Y, Zr, Mo, Tc, Ru, Pd, Ag, Cd, In, Sn, Sb, Te, I, Cs, Ba
- N edge Hf, Ta, W, Re, Os, Ir, Pt, Au, Hg, Ti, Pb
- Actinides & Lanthanides
- R>7500
- Linear and circular polarization control

2.8 fte beamline scientists, 1 post-doc
David Shuh – leader (in his spare time), Mary Gilles (0.8 fte), Hendrik Blum – APPES, Tolek Tyliszczak - STXM
Scanning Transmission X-ray Microscope (STXM)

0.5 – 25 mm (energy dependent)
Quest for resolution and precision

Current 11.0.2 STXM

Need better solutions:
• Interferometers
• Motion controllers (piezo stage)
• Thermal control (stability)
• Environment – noise, vibrations
• Pumps

20 nm steps
Low vibration and very good zone plate – key to high resolution

The interferometer works in a feedback loop of the piezo scanning stage reducing low frequency vibration and keeping the beam on the sample in a position with precision <50 nm for 500 eV energy change.

Relative vibrations of sample and zone plate measured with an interferometer.
Mirror alignment requirements

25 nm zone plate has to be shifted about 490 μm with 100 eV energy change. To keep the X-ray spot on the sample in full energy range, zone plate stage mirror has to be aligned with precision better than 3 μrad to the X-ray beam direction.
Scan tests – speed, precision

**Fast scan**

- 3 \( \mu \)m – 50 Hz
- 100 x 100 pixels image in 1 sec

**Full range 40 \( \mu \)m – 5 Hz**

Very low vibrations – some drift due to air turbulence
X axis sample scan 25 nm\(_{pp}\), 5Hz, ALS floor, optical table, no other vibration isolation

**Open loop test scans**

- Horizontal scan 30 nm
- Vertical scan 30 nm
Expected outcome: Photoelectron spectrometer with <100 nm spatial resolution, operating at gas pressures > 10 Torr in 280 eV – 1600 eV X-ray energy range, using a single zone plate.

Realization: Scanning zone plate microscope module for existing ambient pressure photoemission spectrometer

Challenges: zone plate illumination, vibrations, precision of scanning, precision of moving the zone plate along beam direction ( >10mm at < 200 nm run out), confined space
Sample will be placed on a standard UHV manipulator (XYZΦ) but to improve stability, the manipulator tube is supported close to the sample holder. Sample coarse positioning and scans are done using the manipulator motorized stages. OSA is placed on xyz stages to center on the electron energy analyzer focal point. Zone plate has coarse xyz stages for positioning and xy scanning stage with 40 μm range. The zone plate scanning motion controller uses a single beam xy interferometers in the feedback loop to reduce (eliminate) shift with energy change to have reproducible motion.

Planned resolution – initially 100 nm (reduced resolution because of zone plate illumination problems) and in the second phase – 50 nm.
Ambient Pressure SPEM
First results

Transmission image

XPS image
Photoelectron spectroscopy

2 component maps

Carbon 1s
Photoelectron spectroscopy Au, Pt

SPEM_110519019.hdr
100x10 Image Scan 750.012 eV
0.4 ms Dwell Polariz = 0.0

SPEM_110519018.hdr
100x10 Image Scan 750.012 eV
0.4 ms Dwell Polariz = 0.0

SPEM_110519020.hdr
100x10 Image Scan 750.012 eV
0.4 ms Dwell Polariz = 0.0

Au

Pt

C

Unshifted Image

Unshifted Image

Unshifted Image

Lawrence Berkeley National Laboratory
### Zone plates and working distance

#### EY

<table>
<thead>
<tr>
<th></th>
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<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>at 300 eV</td>
<td>300</td>
</tr>
<tr>
<td>45</td>
<td>240</td>
<td>700</td>
<td>2810</td>
<td>6790</td>
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<tr>
<td>25</td>
<td>240</td>
<td>350</td>
<td>1467</td>
<td>3423</td>
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<td>17</td>
<td>120</td>
<td>130</td>
<td>570</td>
<td>1330</td>
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</table>
• Non transparent samples

• Use a projection lens like the SPECS
FY Hardware challenge
X-ray fluorescence in STXM

Tolek Tyliszczak (ALS), Martin Obst (Tuebingen), Adam Hitchcock (McMaster)

Arsenic (1 mM) exposure of Fe-metabolizing fresh water bacteria

X-ray fluorescence yield X-ray absorption provides a reliable As map in a case where the conventional transmission signal does not detect it.
Fluorescence yields in soft X-ray range

<table>
<thead>
<tr>
<th></th>
<th>K edge</th>
<th>L edge</th>
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<tbody>
<tr>
<td>3 Li</td>
<td>$[1.64(-4)]^a$</td>
<td>26 Fe</td>
</tr>
<tr>
<td>4 Be</td>
<td>$[4.51(-4)]^a$</td>
<td>0.0091±0.0014</td>
</tr>
<tr>
<td>5 B</td>
<td>$[.00101]^a$</td>
<td>0.0083±0.0016</td>
</tr>
<tr>
<td>6 C</td>
<td>$[.00198]^a$</td>
<td>29 Cu</td>
</tr>
<tr>
<td>7 N</td>
<td>$[.00351]^a$</td>
<td>0.0105±0.0011</td>
</tr>
<tr>
<td>8 O</td>
<td>$[.00579]^a$</td>
<td>6.0098±0.0019</td>
</tr>
<tr>
<td>9 F</td>
<td>$[.00902]^a$</td>
<td>30 Zn</td>
</tr>
<tr>
<td>10 Ne</td>
<td>$[.0134]^a$</td>
<td>0.0117±0.0018</td>
</tr>
<tr>
<td>11 Na</td>
<td>$[.0192]^a$</td>
<td>31 Ga</td>
</tr>
<tr>
<td>12 Mg</td>
<td>$[.0265]^a$</td>
<td>0.0129±0.0019</td>
</tr>
<tr>
<td>13 Al</td>
<td>.0357</td>
<td>32 Ge</td>
</tr>
<tr>
<td>14 Si</td>
<td>.0469</td>
<td>0.0139±0.0021</td>
</tr>
<tr>
<td>15 P</td>
<td>.0603</td>
<td>33 As</td>
</tr>
<tr>
<td>16 S</td>
<td>.0760</td>
<td>0.0156±0.0023</td>
</tr>
<tr>
<td>17 Cl</td>
<td>.0941</td>
<td>36 Kr</td>
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<tr>
<td></td>
<td></td>
<td>0.0210±0.002</td>
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<tr>
<td></td>
<td></td>
<td>37 Rb</td>
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<td></td>
<td></td>
<td>0.0186±0.0028</td>
</tr>
<tr>
<td></td>
<td></td>
<td>38 Sr</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.0213±0.0032</td>
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</table>

Are the FY STXM measurements feasible?
Expected count rate

- Flux = $10^8$ photons/s
- FY = 1%
- Collection angle (10 mm sample-detector, 30 mm$^2$ detector) = 0.6 %
- Detection time (dwell time) = 100 ms

If you have thick sample (total absorption)

600 photons / pixel

- What detection limits we can expect?
## 11.0.2 STXM general user proposals

<table>
<thead>
<tr>
<th>Beamline</th>
<th>% Allocated</th>
<th>Cutoff Score</th>
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<tbody>
<tr>
<td>1.4 (IR)</td>
<td>61</td>
<td>2.47</td>
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<tr>
<td>4.0.2 (EPU)</td>
<td>43</td>
<td>2.08</td>
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<tr>
<td>5.3.2 (Polymers XAFS)</td>
<td>65</td>
<td>2.40</td>
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<tr>
<td>6.0.1 (Femtosecond)</td>
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<tr>
<td>6.0.2 (Femtosecond)</td>
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<tr>
<td>6.1.2 (Soft X-Ray Microscopy)</td>
<td>67</td>
<td>2.77</td>
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<tr>
<td>6.3.1 (Materials Sciences)</td>
<td>53</td>
<td>2.19</td>
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<tr>
<td>6.3.2 (Calibration and Standards)</td>
<td>77</td>
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<tr>
<td>7.0.1 (XPS, STXM, SXF, SPEM)</td>
<td>36</td>
<td>2.10</td>
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<tr>
<td>7.3.3 (SAXS)</td>
<td>48</td>
<td>2.20</td>
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<tr>
<td>8.0.1 (SXF)</td>
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<td>8.3.2</td>
<td>70</td>
<td>2.88</td>
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<tr>
<td>9.0. (Chemical Dynamics, Coherent Imaging)</td>
<td>48</td>
<td>2.20</td>
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<tr>
<td>9.3.1 (XAMS)</td>
<td>--</td>
<td>--</td>
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<tr>
<td>9.3.2 (APSD/AMC, High-Pressure XPS)</td>
<td>56</td>
<td>2.28</td>
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<tr>
<td>10.0.1. (HERS/AMO)</td>
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<td>2.25</td>
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<tr>
<td>10.3.2 (Micro XAFS)</td>
<td>38</td>
<td>2.18</td>
</tr>
<tr>
<td>11.0.1 (Magnetic Microscopy, Spectromicroscopy; PEEM3)</td>
<td>65</td>
<td>2.47</td>
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<tr>
<td>11.0.2 (Molecular Environmental Sciences)</td>
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<td>1.72</td>
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<td>11.3.1 (Small Molecule Crystallography)</td>
<td>86</td>
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<td>12.0 (ARPES)</td>
<td>42</td>
<td>2.13</td>
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<td>12.2.2 (High Pressure)</td>
<td>59</td>
<td>2.22</td>
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<tr>
<td>12.3.2</td>
<td>58</td>
<td>2.48</td>
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</table>

7 AP, ~70 general proposals

Beamline 11.0.2 (endstations not separated for the beamtime allocation). oversubscription factor 5-6 to 1 (over last 5 years)

45 proposals with a score below 2.1 (total 69 proposals) – 11 allocated

A large field for the new, similar beamlines
The future SXM at the ALS

**In development:**
- Zone plate scanning
- New mechanical and piezo stages (to reduce vibrations, increase scanning speed)
- Increased resolution of interferometers
- Low temperature sample stage
- Tomography
- Ptychography
- Ambient Pressure SPEM

**Needed:**
- New microscope chamber – better vacuum mostly for low temperature
- Additional replacement stages – cleaner environment
- Environmental control inside and outside – improve stability
- Microscope computer control positioning (girder system) – alignment and zone plate illumination
- Better detectors (scintillators, avalanche photo diodes, fluorescence)
- Additional equipment for magnetization dynamic
Obvious: high performance microscope but there are important questions

- One microscope or 2-3
- Omnibus or dedicated/specialized microscopes
- Geared towards physics or multidiscipline general user instrument
- Low temperature – LN2 or LHe
- Environmental cells

Do not sell it cheap – it should be the best.
Publications:
• 34 publications
  — STXM - 18

Successful, multidiscipline beamline and endstation

A model for many beamline around the world
11.0.2 STXM- still best in the world

- Energy 90 eV – 2000 eV with resolving power > 6000
- Spatial resolution - can resolve 10 nm spaced lines (in 1st order)
- Time resolved measurements with 70 ps resolution
- Internal electromagnets (in plane and perpendicular field) for static magnetization measurements
- Normal and rotated (30 deg) scanning for polarization measurements
- Flexible sample mounting
- Fluorescence detection

In development
- Zone plate scanning
- New mechanical and piezo stages (to reduce vibrations, increase scanning speed)
- Increased resolution of interferometers
- Low temperature sample stage
- Tomography

Best working resolution of all X-ray microscopes
Observing In-situ catalytic reaction on nm scale at temperature up to 550 C and 2 bar pressure

Chemical contour maps of a region of a catalyst particle during the different stages of reaction. A: Before treatment at RT in 1 bar He. B: After 2h in H2 at 350oC. C: After 4h in CO/H2 at 250oC. The corresponding regions of the O K-edge (e, g, i) and Fe L2,3-edge spectra (d, f, h) are indicated in the figures. Dotted lines indicate the linear combination fits, with the bars representing the contribution of the different phases to the spectra.

Direct observation of magnetization reversal by spin injection – ultimate X-ray microscope challenge

Magnetization Reversal by Spin Injection

Sample prepared by Jordan Katine, Hitachi Global Storage

Challenge: measuring magnetization of thin magnetic layer buried in 250nm of metals with sub-ns time resolution!

From magnetization measured in 2 directions (Fig a and b) magnetization vectors are constructed (Fig c)

• X-rays allow us to image the switching process
• The vortex provides an alternative switching mechanism
• Smaller structures switch by a different mechanism (C-state flip-over?)

Time Resolved Imaging of Spin Transfer Switching: Beyond the Macro-Spin Concept,
Y. Acremann, J. P. Strachan, V. Chembroli, S. D. Andrews, T. Tyliszczak, J. A. Katine, M. J. Carey,

Measurements of magnetization distribution dynamics of 2 nm layer buried in 200 nm metal of 100 nm diameter pillar with 70 ps resolution

Sample prepared by Jordan Katine, Hitachi Global Storage

From magnetization measured in 2 directions (Fig a and b) magnetization vectors are constructed (Fig c)

Switching by vortex motion

The Oersted field creates the vortex
Spin injection moves the vortex


Software defined photon counting system for time resolved x-ray experiments

Vortex speed ≈ 180 m/s

Pos Switch: 600 ps
Neg Switch: 800 ps

The Oersted field creates the vortex
Spin injection moves the vortex

Challenge: measuring magnetization of thin magnetic layer buried in 250nm of metals with sub-ns time resolution!
Magnetization dynamic

Magnetic vortex core reversal by short bursts of an alternating field

A single period of AC magnetic field (< 1mT) can reverse magnetization direction of a vertex core (vortex dimension < 20 nm)


Established a new technique for study of magnetization dynamic (70ps, 20 nm resolution) 9 high profile publications (Nature, PRL)
Cometary and interstellar dust particles

TYPE IIA CHONDRULE FRAGMENT FROM COMET 81P/WILD2 IN STARDUST TRACK C2052,2,74

Optical and X-ray Fluorescence images

STXM element maps of Iris,9. Absorption difference maps, pixel size 100 nm. a) Ca-Al-Mg-. Yellow regions are Ca-Al rich, Purple regions are Ca-bearing Mg-Fe silicates. b) Al-Si-Mg map,

STXM provided detail understanding of chemical composition before the TEM work to identify crystalline structures.

High resolution characterization of aerogel embedded particles

Fluorescence of environmental samples

2 order higher sensitivity the in transmission for As
Highest spatial resolution.
Combination of elemental and chemical speciation

A. Hitchcock, M. Obst, T. Tyliszczak - work in progress