X-ray imaging

Mingyuan Ge

Beamline scientist (FXI, 18ID), NSLS-II, Brookhaven National Laboratory

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• Imaging using light

- Increasing energy $E$
- Increasing frequency $\nu$
- Increasing wavelength $\lambda$

- PET
- UV
- IR
- centimeter wavelength (Radar)
- Gamma
- X-ray
- visible light (GFP)
- EHT-submillimeter
- Wifi tomography (centimeter)

<table>
<thead>
<tr>
<th>Gamma</th>
<th>X-ray</th>
<th>Ultraviolet</th>
<th>Infrared</th>
<th>Terahertz</th>
<th>Microwave</th>
<th>Broadcast and wireless radio</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10^{-12}$</td>
<td>$10^{-11}$</td>
<td>$10^{-10}$</td>
<td>$10^{-9}$</td>
<td>$10^{-8}$</td>
<td>$10^{-7}$</td>
<td>$10^{-6}$</td>
</tr>
</tbody>
</table>

Wavelength $\lambda$ (m)
- X-ray imaging --- multimodal imaging technique

Direct imaging
(full frame)

- Transmission, CT

Indirect imaging (mostly scanning microscopy)

- X-ray fluorescence (XFM, BNL)

- Scattering (diffraction)

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**Nanoscale Adv., 2019, 1, 3009-3014**

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**Research at NSLS-II imaging group --- beamline capability**

- **Wide range of resolution**
- **Multi-mode detection (Absorption, fluorescence, diffraction)**
  - morphology, element species, valance states, stress/strain

<table>
<thead>
<tr>
<th>beamline</th>
<th>imaging method</th>
<th>energy range</th>
<th>max. res.</th>
<th>max. FOV</th>
<th>imaging modality</th>
<th>tomography</th>
</tr>
</thead>
<tbody>
<tr>
<td>FXI</td>
<td>full field</td>
<td>5-11 keV</td>
<td>20 nm</td>
<td>50 um</td>
<td>Absorption, XANES</td>
<td>Yes</td>
</tr>
<tr>
<td>HXN</td>
<td>scanning</td>
<td>6-20 keV</td>
<td>12 nm*</td>
<td>30 um</td>
<td>fluorescence, differential phase contrast, diffraction, ptychography, and XANES</td>
<td>Yes</td>
</tr>
<tr>
<td>SRX</td>
<td>scanning</td>
<td>4.5-20 keV</td>
<td>100 nm</td>
<td>~1.5mm</td>
<td>fluorescence, XANES, EXAFS, and diffraction</td>
<td>In 2021</td>
</tr>
<tr>
<td>XFM</td>
<td>scanning</td>
<td>4.7-20 keV</td>
<td>2 um</td>
<td>~100 mm</td>
<td>fluorescence, diffraction, absorption, XANES and EXAFS</td>
<td>Yes</td>
</tr>
<tr>
<td>TES</td>
<td>scanning</td>
<td>2.0-5.5 keV</td>
<td>5 um</td>
<td>~3 mm</td>
<td>fluorescence, XANES and EXAFS</td>
<td>No</td>
</tr>
</tbody>
</table>
• FXI beamline at BNL (18-ID)

• Home-designed transmission X-ray microscopy (TXM)

Energy range: 5-11 keV
Energy resolution: $\Delta E/E = 10^{-4}$
• TXM capabilities

TXM optics

• Fast nano 3D tomography (nano-CT)
  • 1 min
  • 30 nm resolution
  • 50 um x 50 um field of view

• 2D/3D XANES (X-ray absorption near edge spectrum)
  • Imaging the chemical species
**TXM capabilities**

- Fast nano 3D tomography (nano-CT)
  - 1 min
  - 30 nm resolution
  - 50 um x 50 um field of view

**TXM optics**

- phase ring
- sample
- capillary condenser
- X-ray
- detector
- objective ZP
- pinhole

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**Periodic Table**

- Group 1: H, Li, Be, B, C, N, O, F, Ne
- Group 2: He, Ne, Ar
- Group 3: Li, Be, B, C, N, O, F, Ne
- Group 4: Na, Mg, Al, Si, P, S, Cl, Ar
- Group 5: K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Kr
- Group 6: Rb, Sr, Ba, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb
- Group 7: Th, Pa, U, Np, Pu, Am, Cm, Bk, Cf, Es, Fr, Ra, Ac
- Group 8: Ra, Am, Cm, Bk, Cf, Es, Fr, Ra, Ac

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• TXM application: Fast 3D tomography (1min)

• Cu + Ag$^+$ $\rightarrow$ Ag + Cu$^{2+}$
• X-ray: 8.96 keV, 0.02 s exposure time
• 3 deg/s (0-180 deg), 1060 projection images

• TXM application: XANES

Auger electron \rightarrow \text{Emitted photo-electron} \rightarrow \text{(XPS)}

M edges (n = 3)

3p

3s

L edges (n = 2)

2p

2s

K edge (n = 1)

1s

\( h\nu \rightarrow \text{Fluorescent photon} \rightarrow h\nu' \)

Core Hole

Dipole selection rule: \( \Delta l = \pm 1 \)

(e.g. ns ↔ np transition)
• **TXM application: chemical states**
• Chemical state mapping using X-ray absorption spectrum (XANES)

\[ I = I_0 \exp(-\mu \cdot t) \]

- **LiNi\(_{0.5}\)Mn\(_{0.3}\)Co\(_{0.2}\)O\(_2\)(NMC532)**

Red: Ni\(^{2+}\)

Green: Ni\(^{3+}\)

\[ \text{Ni}^{2+}/\text{Ni}^{3+} = 0.57/0.45 \]
Application:

- In-situ observation of Ni oxidation change during charging

Region 1

Region

LiNi$_x$Co$_y$O$_2$

Voltage (V vs Li/Li$^+$) vs Specific capacity (mAhg$^{-1}$)

Unpublished work

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Application: Chemical states in 3D

Cycled LiNi$_x$Co$_y$O$_2$ (Seong Min Bak, BNL)

• Color image: Ni$^{2+}$ concentration
• Cathode particle after long cycling, showing crack and enrichment of Ni$^{2+}$ at crack site

Unpublished work
Chemical states in 3D

NMC532 after 100 cycles, stop at 3.7V

Solid electrolyte–NMC interface (FengWang BNL)
**TXM application: Molten salt corrosion (reactor)**

**Historic background:**

**Fukushima Daiichi Accident (2011)**

[Image: Fukushima Daiichi Accident](https://ejatlas.org/conflict/fukushima)


"New" design: molten salt reactor (Th carried in molten salts)

\[ ^{232}\text{Th} (n,\gamma) \rightarrow \cdots \rightarrow ^{233}\text{U} (n,2n) \]


• *In situ* Study: Morphological Evolution of Ni, Ni-20Cr in MgCl$_2$-KCl

• Capturing time-dependent behavior to study kinetics

Summary of TXM capabilities

• TXM (Transmissional X-ray Microscopy)
  • Morphology (2D, and 3D tomography)
  • Chemicals (2D and 3D XANES)

• Capability at FXI beamline (NSLS-II 18-ID)
  • Fast tomography (1 min)
  • In-situ experiment (e.g., electrochemistry, heating)
HXN: Scanning probe microscopy

Highest special resolution: 11 nm
XRF imaging
LiNiMnCoO$_2$ particles: non-uniform element distribution
Ni, Mn, Co

XRF overlay

Atomic percentage overlay
XRF imaging

LiNiMnCoO$_2$ particles: non-uniform element distribution

Ni, Mn, Co

XRF overlay

Atomic percentage overlay

Unpublished work

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• 3D quantitative analysis

Solid electrolyte for fuel cell: 
\( \text{Gd}_x\text{Ce}_{1-x}\text{O}_2\text{-CoFe}_2\text{O}_4 \) (GDC-CFO) sintering at high temperature

Nature Comm. DOI: 10.1038/ncomms7824
Particle composition analysis → discovering new phases

Inter-diffusion of: Gd to CFO and Fe/Co to GDC

Unpublished work
Understand spatial distribution

CFO
Emerging phase
GDC matrix

Unpublished work
Other sample XRF images

Bio-cell

Solid electrolyte in fuel cells

Cr enrichment in steel after etching

Ti-Mg-Al co-doped LiCoO₂

Journal of the electrochemical society, 166(11), C3320, (2019)

Diffraction imaging

Tracking Bragg peak in nano-diffraction (LiCoO$_2$ particle)
Summary

• Full field imaging (FXI)

• Scanning microscopy (HXN)
Acknowledgement

- TXM group at NSLS-II
  - Wah-Keat Lee (leading scientist)
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  - David Scott Coburn (TXM design)
  - Zhijian Yin (control)
  - Kazimierz Gofron (control)
  - Huijuan Xu (control)
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  - Hanfei Yan
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  - Evgeny Nazaretski
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  - Weihe Xu

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Thanks!