Resonant X-ray Scattering of Quantum Materials at the Canadian Light Source

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REIXS elastic scattering endstation

Canadian Light Source Inc
When a charged particle (electron, positron, ion, ...) is accelerated it emits light.

Synchrotron radiation is light produced when a particle is accelerated along a curved trajectory at relativistic speeds (close to the speed of light)
Synchrotron Radiation
How is it produced?

Magnetic fields are used to make electrons travel in a ring.

Electrons are accelerated to high energy using microwave radiation.
Why Synchrotron Radiation?

Synchrotron radiation has a number of useful properties:

**High brightness**: synchrotron radiation is extremely intense (hundreds of thousands of times higher than conventional X-ray tubes) and highly collimated (similar to a laser).

**Tunable photon polarization and energy**

**Coherence**

**Pulsed source**
The Canadian Light Source

Location:
Saskatoon, SK

www.lightsource.ca

Electron energy:
2.9 GeV

Storage ring circumference:
171 m
The Canadian Light Source
Location: Saskatoon, SK

www.lightsource.ca

Electron energy: 2.9 GeV

Storage ring circumference: 171 m
First Light: 2003

17 beamlines operational
3 under construction

www.lightsource.ca
1. Electron gun
2. Linear accelerator
3. Booster ring
4. Storage ring
5. Beamline optics
6. Experimental endstation
Bemline: Resonant Soft x-ray scattering

**Undulator**

An adjustable periodic array of magnets that controls the photon energy and polarization

**Monochromator**

Define and vary the photon energy

+ mirrors to focus the light onto an endstation
Resonant Soft x-ray scattering at the Canadian Light Source

Resonant Scattering endstation
Resonant Soft X-ray Scattering at the Canadian Light Source

- 4 circle diffractometer (9 in-vacuum motions)
- Ultra-high vacuum ($P \sim 2 \times 10^{-10}$ mBar)
- Photodiode, channeltron, channelplate and polarization sensitive detector
- Cooling to < 20 K
- Full polarization control of incident light (EPU)

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Resonant X-ray Diffraction and Reflectometry

x-ray spectroscopy

x-ray diffraction (in crystals)

x-ray reflectometry (in thin films and multilayers)

electronic structure

spatial structure
Near an absorption edge, the atomic scattering form factor, $f$, (how strongly an x-ray scatters from an element), becomes strongly dependent on photon energy and polarization.
Nematicity in stripe-ordered cuprates probed via resonant x-ray scattering


Science 351, 576 (2016).
What are the ordered phases of the cuprates? - superconductivity - anti-ferromagnetism - charge density wave order - spin-density wave order - nematic order? - loop current order?

How do different types of order interact?
Density wave order in the cuprates

La$_{1.475}$Nd$_{0.4}$Sr$_{0.125}$CuO$_4$ Elastic Neutron scattering

Unidirectional Spin and charge order (stripes) first observed in the cuprates by neutron scattering (Tranquada et al., Nature 1995)

Charge density wave (CDW) peak

Half-filled charge stripe

Undoped AF regions
Charge Density Wave Order in Cuprate Superconductors

\[ \text{La}_{1.475}\text{Nd}_{0.4}\text{Sr}_{0.125}\text{CuO}_4 \]

- Bragg peak due to spatial modulation in the electronic structure
- Intensity enhanced on resonance

Achkar et al. PRL 110, 017001 (2013)
Structure, nematicity and CDW order in \((\text{La,X})_2\text{CuO}_4\)

Neutron scattering

La_{1.475}\text{Nd}_{0.4}\text{Sr}_{0.125}\text{CuO}_4

Charge density wave order onsets below 1\text{st} order structural phase transition: low temperature orthorhombic (LTO) to low temperature tetragonal (LTT)

Structure, nematicity and stripes in 
\((\text{La,}X)\text{CuO}_4\)

1\textsuperscript{st} order LTO to LTT phase transition measured by x-ray and neutron scattering

Axe PRL 1989
Suzuki Physica C 1989
Tranquada 1995
Zhao PRB 2007
Kim PRB 2008
Fink PRB 2011
Wilkins PRB 2011
Hucker PRB 2011
...
Tilt direction of octahedra alternates between neighboring planes.
Unidirectional CDW order: stripes

La-based cuprates

LTT tilts and stripe order

LTT distortion stabilizes stripes that alternate in direction between neighboring CuO$_2$ planes

Tranquada 1995
Tranquada 1996
(001) Bragg reflection

Conventional x-ray diffraction
(001) Bragg peak is forbidden

- Scattering from neighbouring planes destructively interferes

Resonant x-ray diffraction
(001) Bragg peak is allowed

Fink et al. PRB 2011
Wilkins PRB 2011
The (001) peak at the Cu L resonance measures electronic nematicity of the Cu 3d states.

The octahedral tilting breaks the $C_4$ symmetry of the orbitals in each plane.

Low temperature tetragonal (LTT) structure
(0 0 1) peak at different photon energies

Measure (0 0 1) at different photon energies.

→ Provides sensitivity to different atoms in the unit cell.
Relation to CDW order

Peak amplitude

La$_{1.65}$Eu$_{0.2}$Sr$_{0.15}$CuO$_4$

Achkar Science 2016

Intensity (arb. units)

Temperature (K)

(0 0 1) apical O

(0 0 1) Cu

(0.26 0 1.5) CDW, Cu
Distinct order parameters:

Electronic nematicity of the CuO$_2$ planes is coupled to, but distinct from the structural distortion of the (La,X)$_2$O$_2$ spacer layer.
Probing Emergent Phenomena at Oxide Interfaces using Resonant X-Ray Reflectometry
Emergent Phenomena at Oxide Interfaces

Control the proximity of different symmetry breaking phenomena to create new phases of matter

enhance desired properties

Ex: increase the superconducting transition temperature by changing dimensionality, applying epitaxial strain, or modifying the orbital symmetry

Hwang et al. 2012
Key Challenge in Studying Emergent Phenomena at Oxide Interfaces

It is experimentally difficult to examine spin, charge, orbital reconstruction of buried interfaces. Many conventional experimental tools are impractical, lack sensitivity or are destructive.
Example: $\text{Mn}_{0.07}\text{Ga}_{0.93}\text{As}$ film on GaAs substrate

$\text{Mn}_{0.07}\text{Ga}_{0.93}\text{As}$ forms a magnetic semiconductor that is potentially useful for new generations electronics, spintronics, that make use of magnetic degrees of freedom.
Example: $\text{Mn}_{0.07}\text{Ga}_{0.93}\text{As}$ film on GaAs substrate

Interference fringes due to the 45 nm thickness of the film

Measuring on resonance provides contrast, letting you “see” to the thin film

Example: $\text{Mn}_{0.07}\text{Ga}_{0.93}\text{As}$ film on GaAs substrate

Resonant reflectivity lets you probe the electronic structure

Chemical and electronic structure depth profiling

Example: Electronic reconstruction on the surface of LaCoO₃ film on an NdGaO₃ substrate

Hamann-Borrero et al. npj Quantum Materials (2016)
Chemical and electronic structure depth profiling

Example: Atomic layer resolved stoichiometry AND magnetic structure

Conclusions

Resonant x-ray diffraction and reflectometry provide a unique, element and orbital specific probe of spin, charge and orbital symmetry breaking in crystals and thin films.