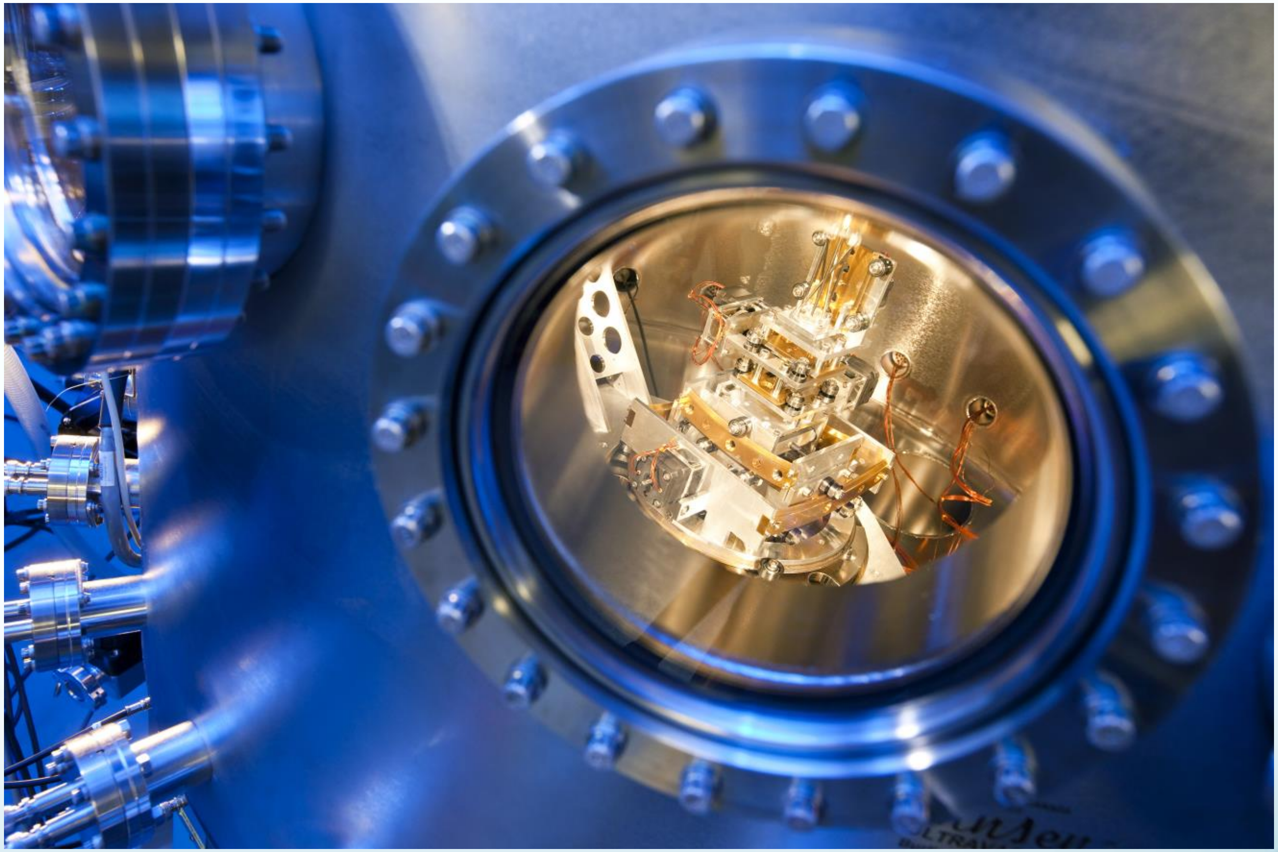
Resonant X-ray Scattering of Quantum Materials at the Canadian Light Source

David Hawthorn
University of Waterloo

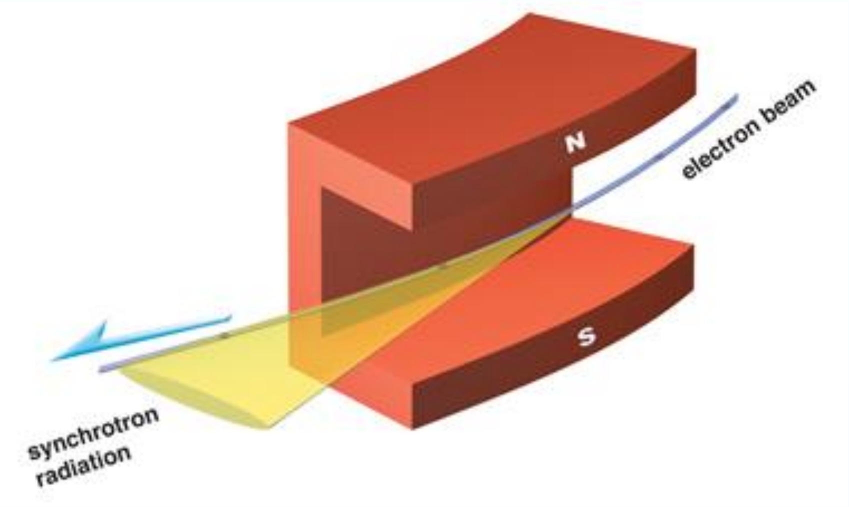


REIXS elastic scattering endstation

Canadian Light Source Inc

NSERC CRSNG
Canadian Light Source Centre canadien de rayonnement synchrotron
Canada Foundation for Innovation
Fondation canadienne pour l'innovation

Synchrotron Radiation



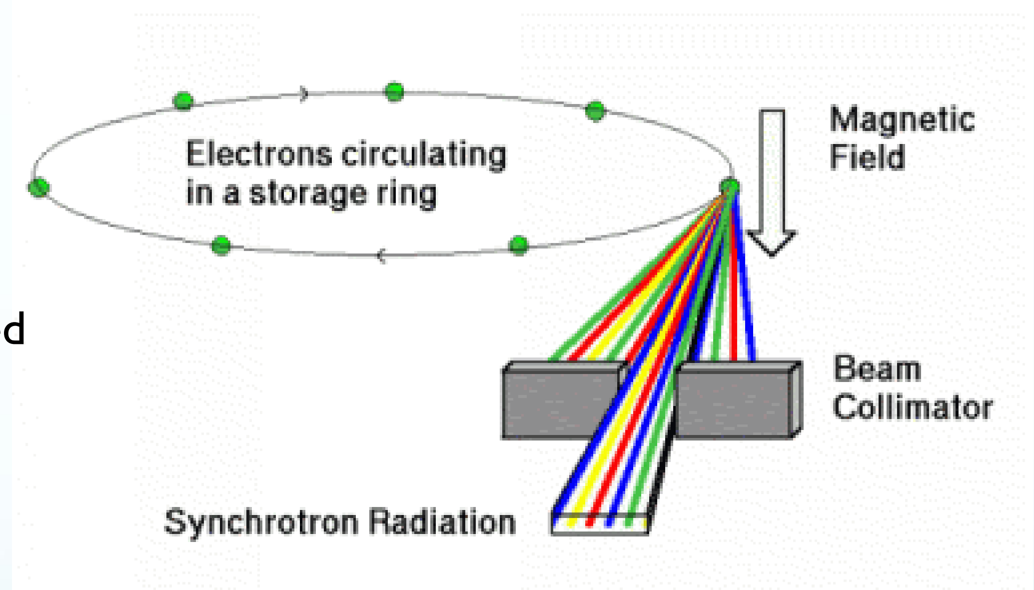
When a charged particle (electron, positron, ion, …) is accelerated it emits light.

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

Synchrotron Radiation
How is it produced?

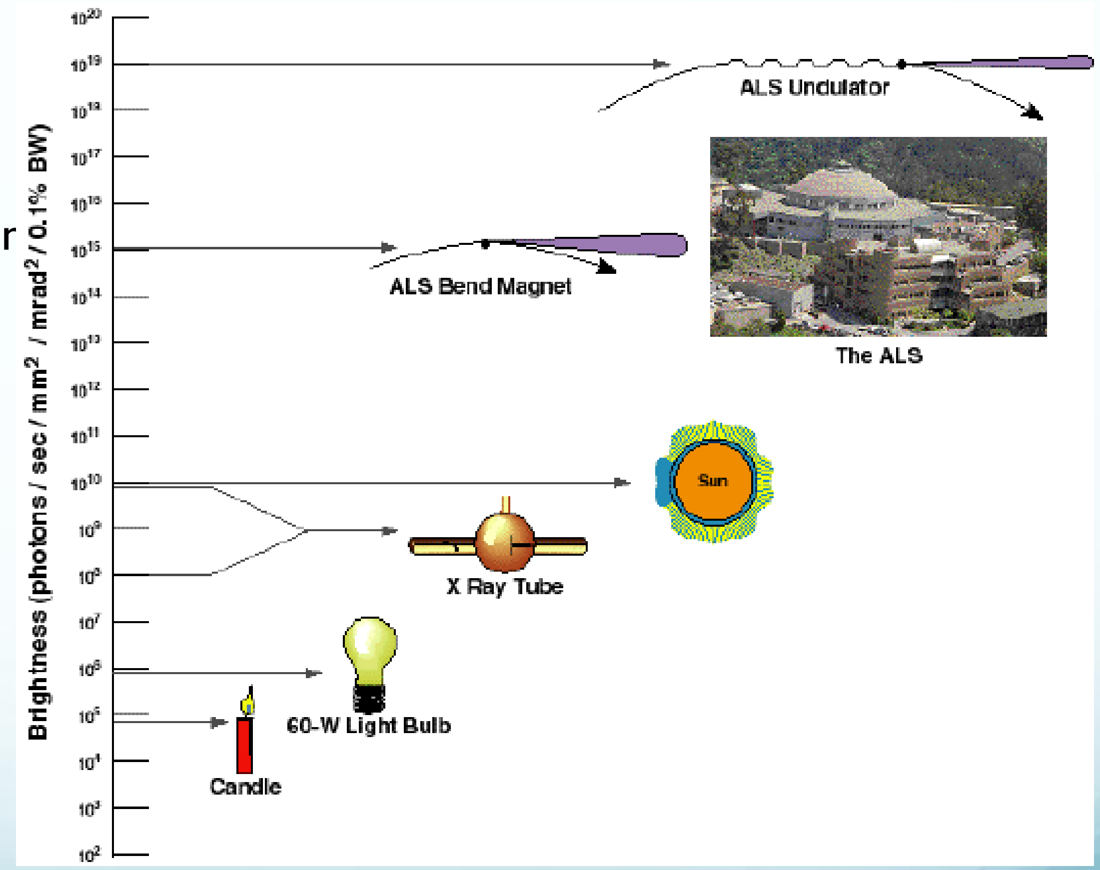
Magnetic fields
are used to make
electrons
travel in a ring

Electrons are accelerated
to high energy using
microwave radiation



Why Synchrotron Radiation?

Synchrotron radiation has a number of usefull 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

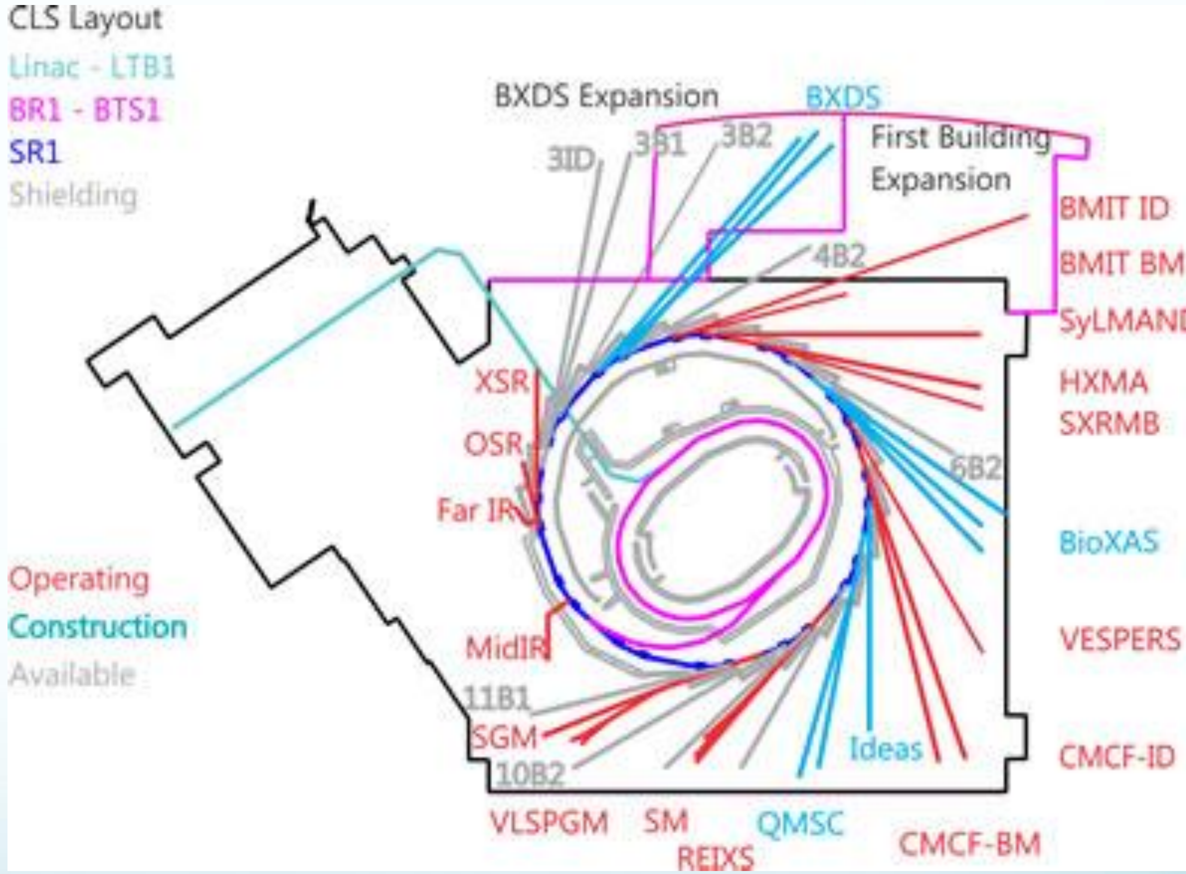
www.lightsource.ca

Electron energy: 2.9 GeV

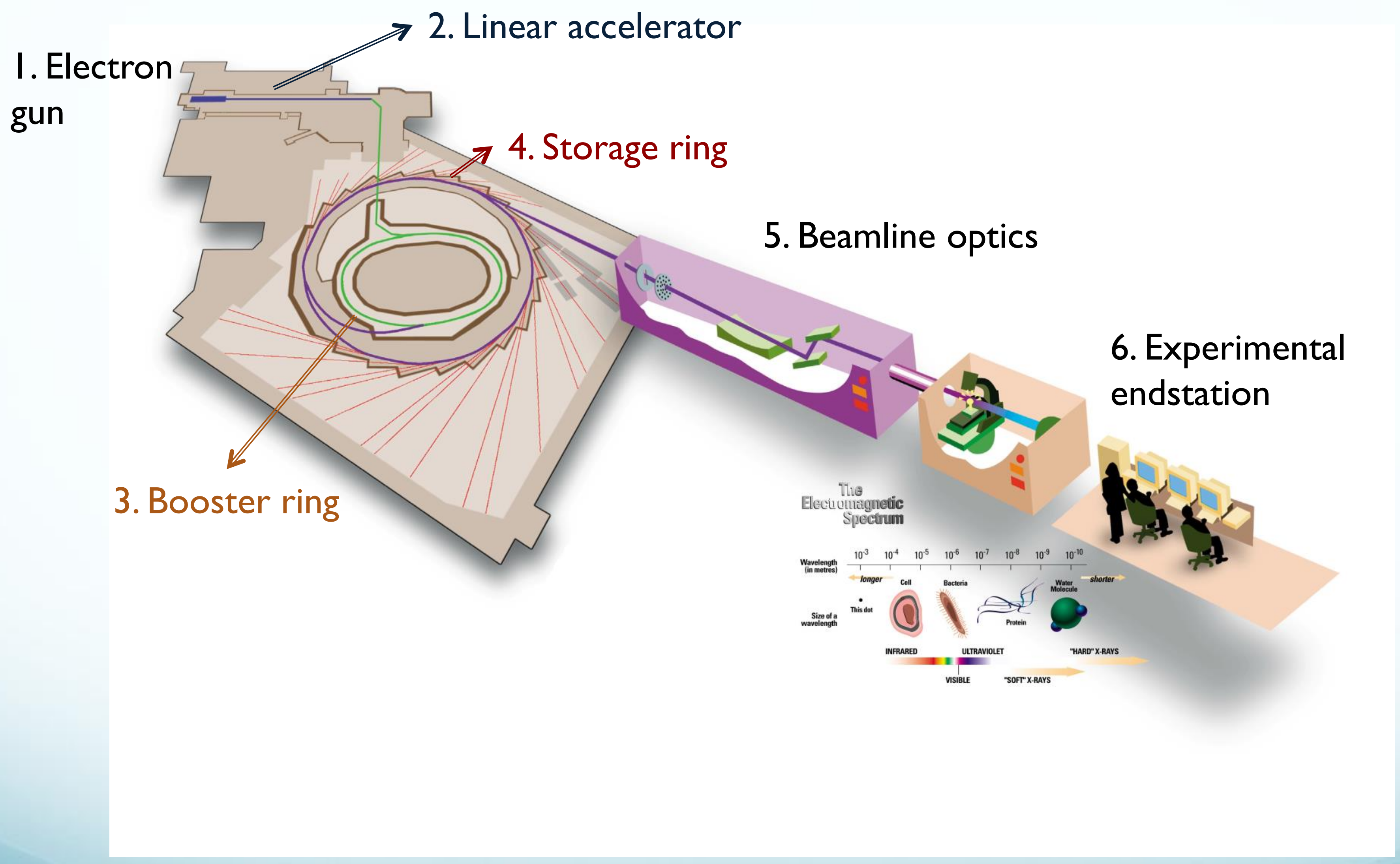
Storage ring circumference: 171 m

First Light: 2003

17 beamlines operational
3 under construction



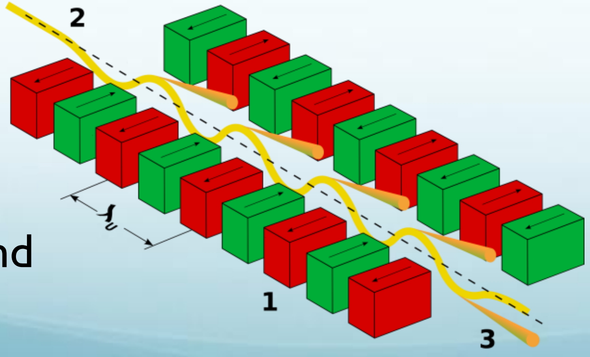
www.lightsource.ca


1. Electron gun
2. Linear accelerator
3. Booster ring
4. Storage ring
5. Beamline optics
6. Experimental endstation
The Electromagnetic Spectrum
Wavelength (in metres)
10-3 10-4 10-5 10-6 10-7 10-8 10-9 10-10
longer
shorter
Size of a wavelength
This dot
Cell
Bacteria
Protein
Water Molecule
INFRARED
ULTRAVIOLET
VISIBLE
"SOFT" X-RAYS
"HARD" X-RAYS

Bemline: Resonant Soft x-ray scattering

Undulator

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



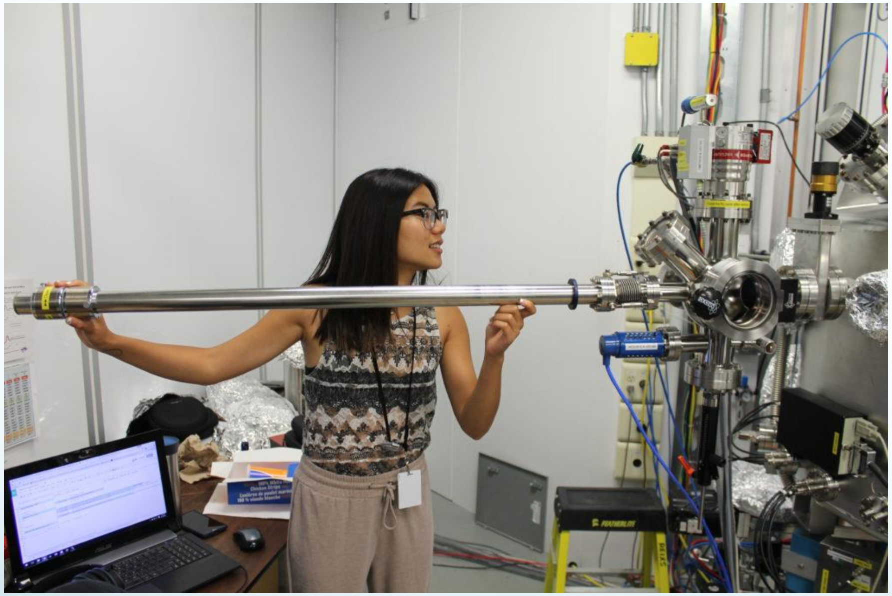
Monochromator

Define and vary the photon energy

+ mirrors to focus the light onto an endstation

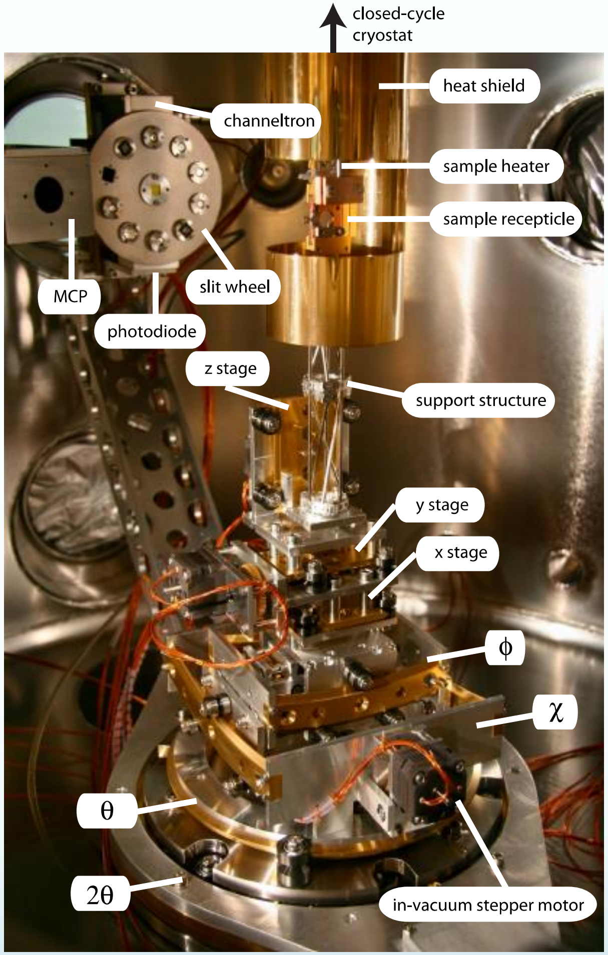
Resonant Soft x-ray scattering at the Canadian Light Source

Resonant Scattering endstation



Resonant Soft X-ray Scattering at the Canadian Light Source

- 4 circle diffractometer (9 in-vacuum motions)
- ultra-high vacuum (P ~ 2 x 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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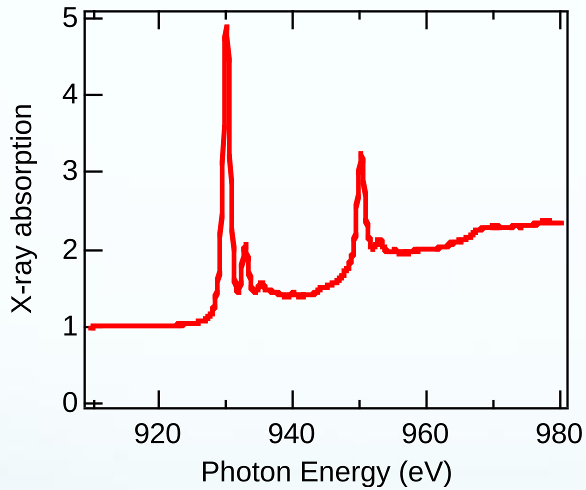
funded by Canada Foundation for Innovation, British Columbia Knowledge Development Fund and Western Economic Diversification

Hawthorn *et al.* Rev. Sci. Instrum. 2011

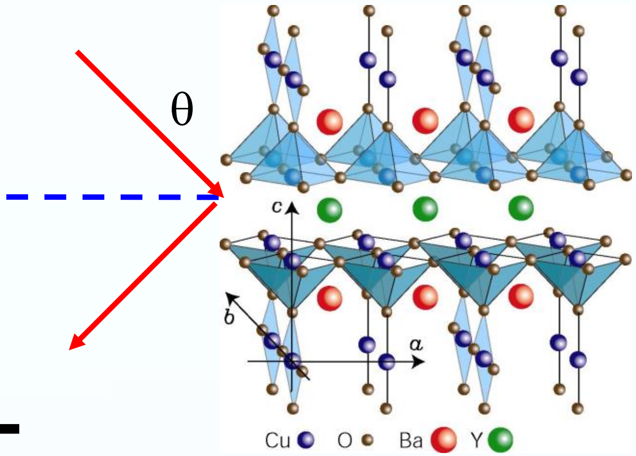
Resonant X-ray Diffraction and Reflectometry

x-ray spectroscopy



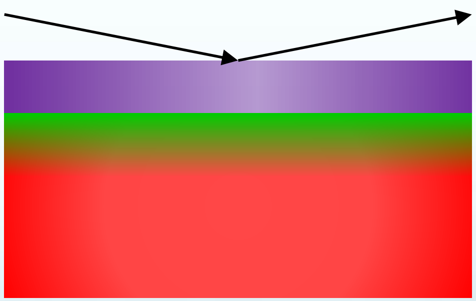
electronic structure

+



x-ray diffraction (in crystals)

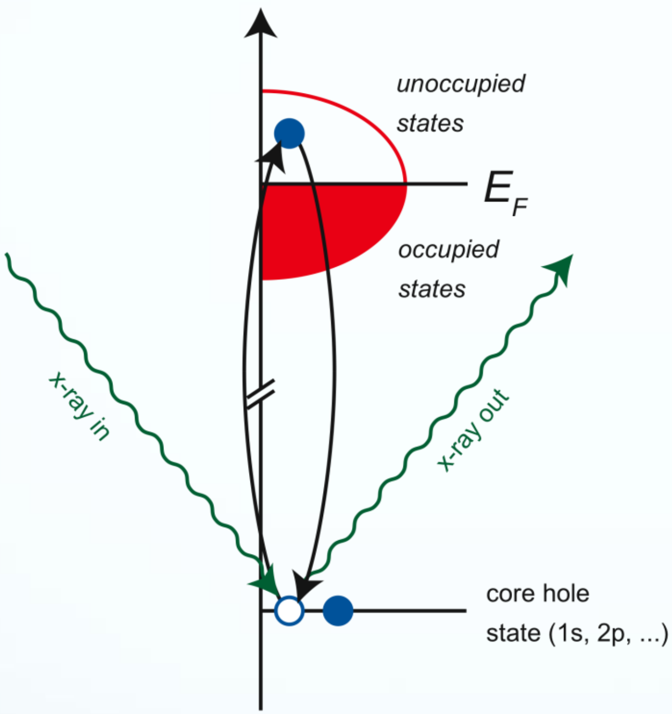
OR

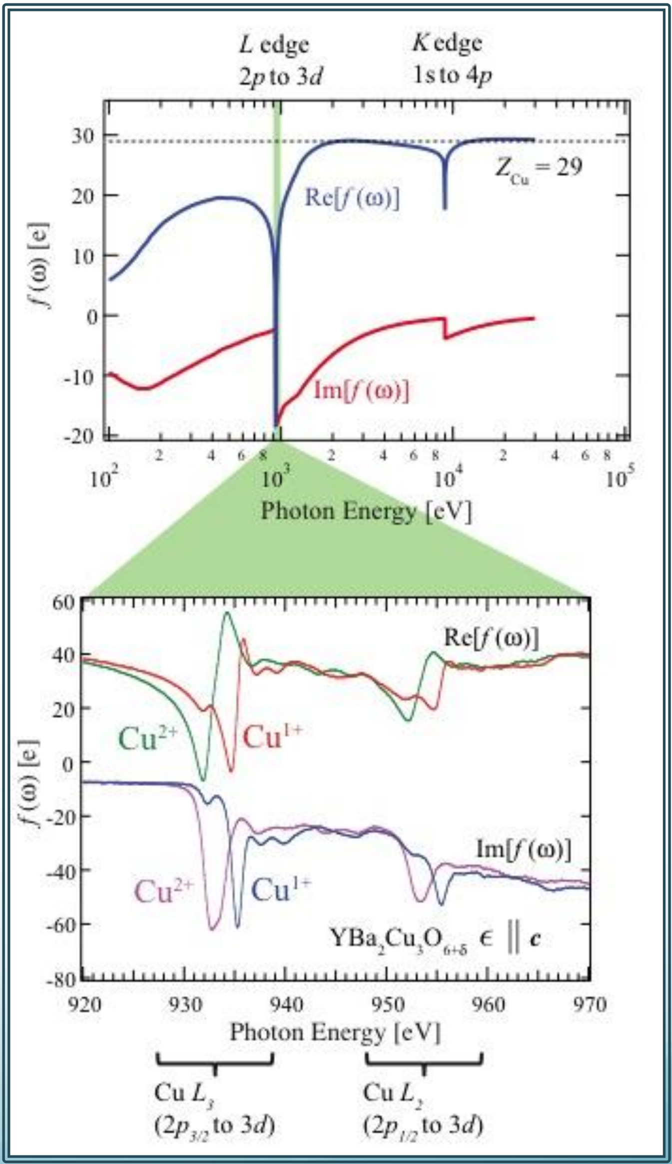


x-ray reflectometry (in thin films and multilayers)

spatial structure

Resonant Elastic X-ray Scattering

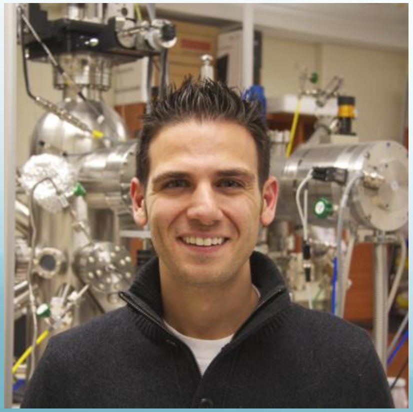




Near an absorption edge, the atomic scattering form factor, f (how strongly an x-ray scatters from an element), becomes strongly dependent on photon energy and polarization

Nematicity in stripe-ordered cuprates probed via resonant x-ray scattering

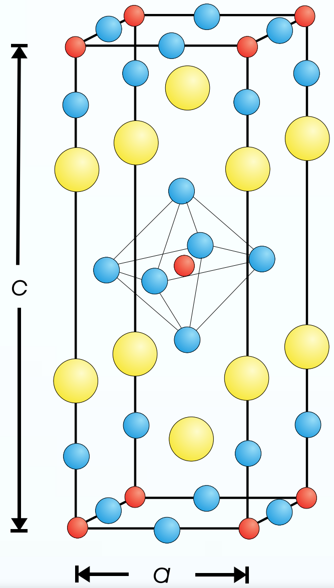
A. J. Achkar, M. Zwiebler, Christopher McMahon, F. He, R. Sutarto, Isaiah Djianto, Zhihao Hao, Michel J. P. Gingras, M. Hücker, G. D. Gu, A. Revcolevschi, H. Zhang, Y.-J. Kim, J. Geck, D. G. Hawthorn

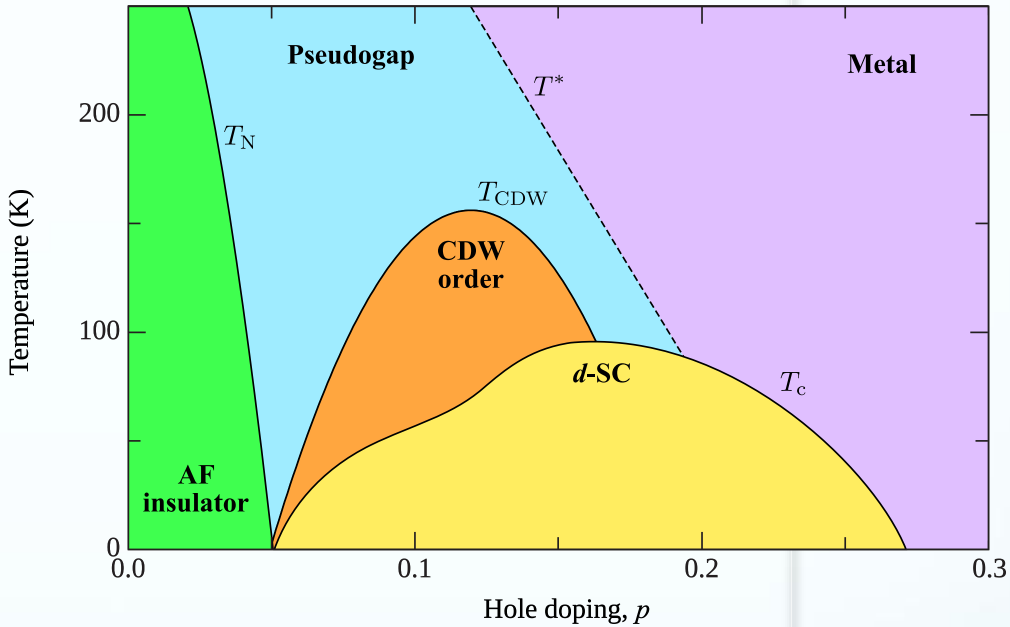


Science **351**, 576 (2016).

Cuprate High-Temperature Superconductors

$La_{2-x}Sr_xCuO_4$





What are the ordered phases of the cuprates?

-superconductivity

-anti-ferromagnetism

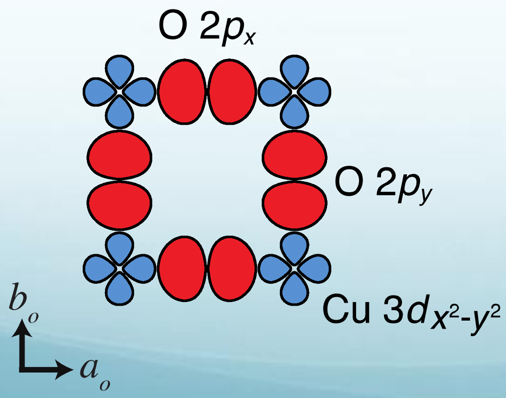
-charge density wave order

-spin-density wave order

-nematic order?

-loop current order?

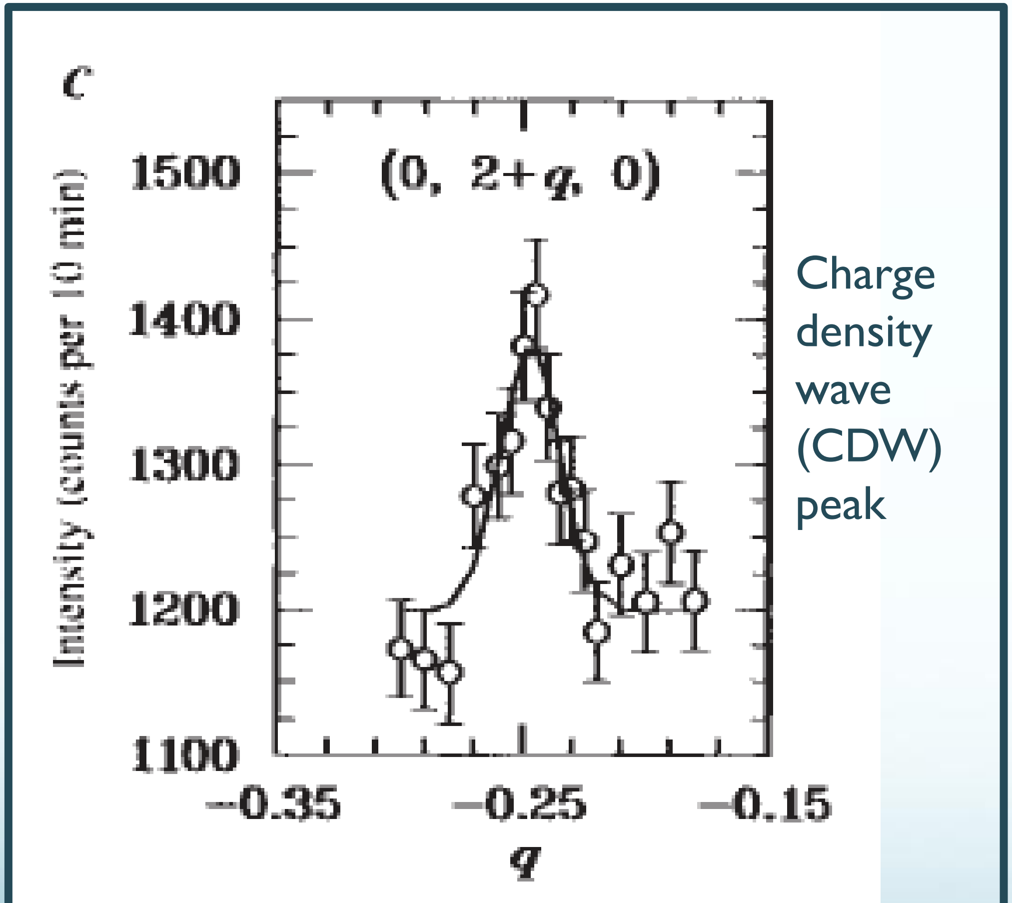
Low energy physics is dominated by the CuO_2 planes



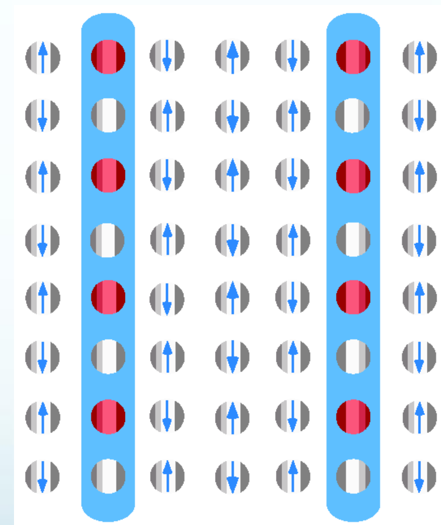
How do different types of order interact?

Density wave order in the cuprates

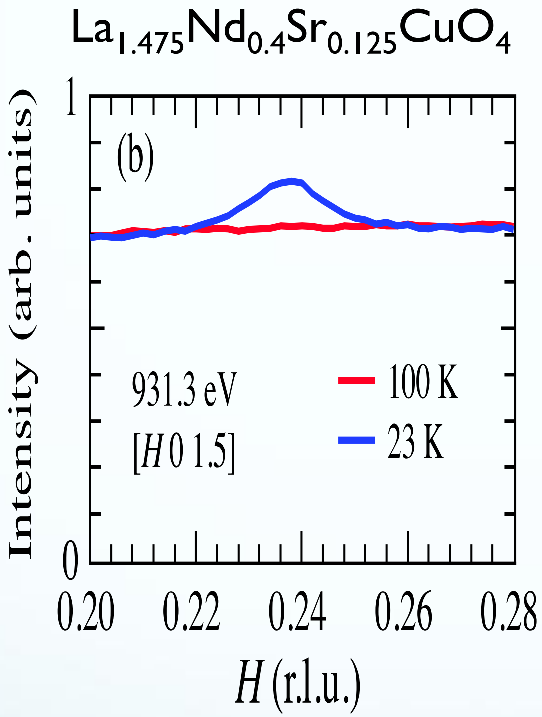
$La_{1.475}Nd_{0.4}Sr_{0.125}CuO_4$ Elastic Neutron scattering

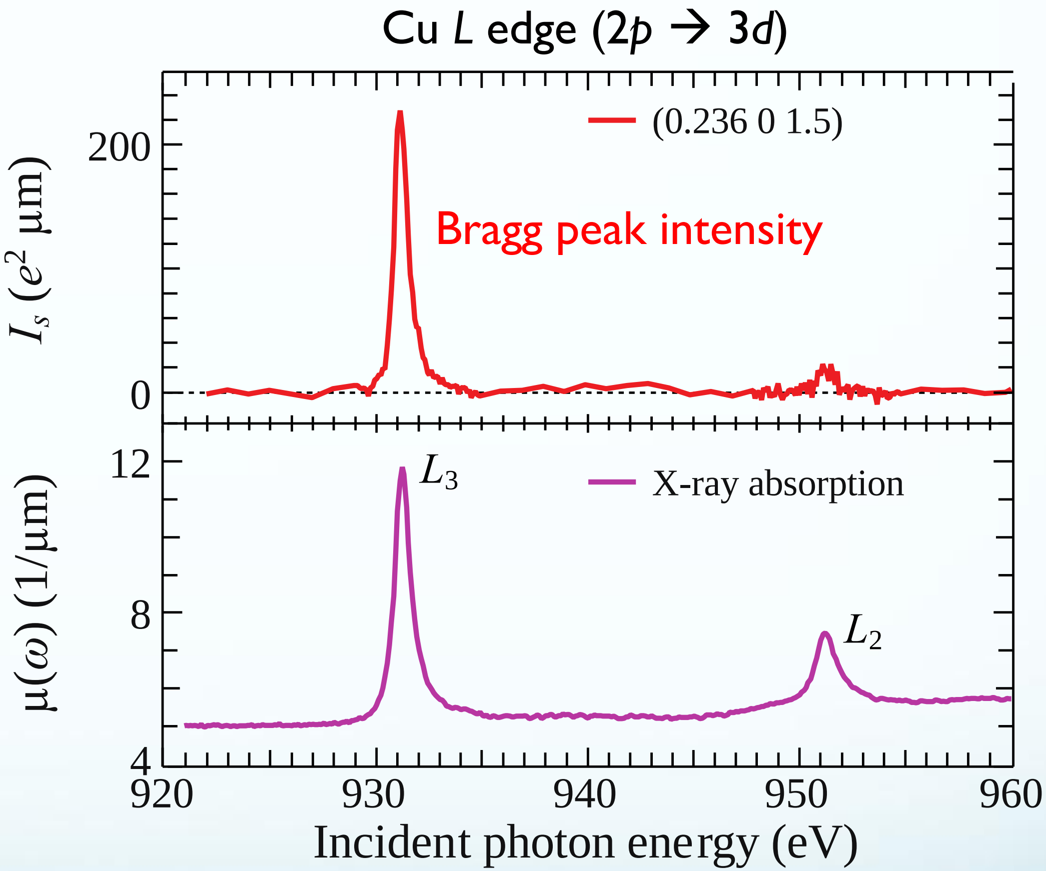


Unidirectional Spin and charge order (stripes) first observed in the cuprates by neutron scattering (Tranquada et al., Nature 1995)



Charge Density Wave Order in Cuprate Superconductors





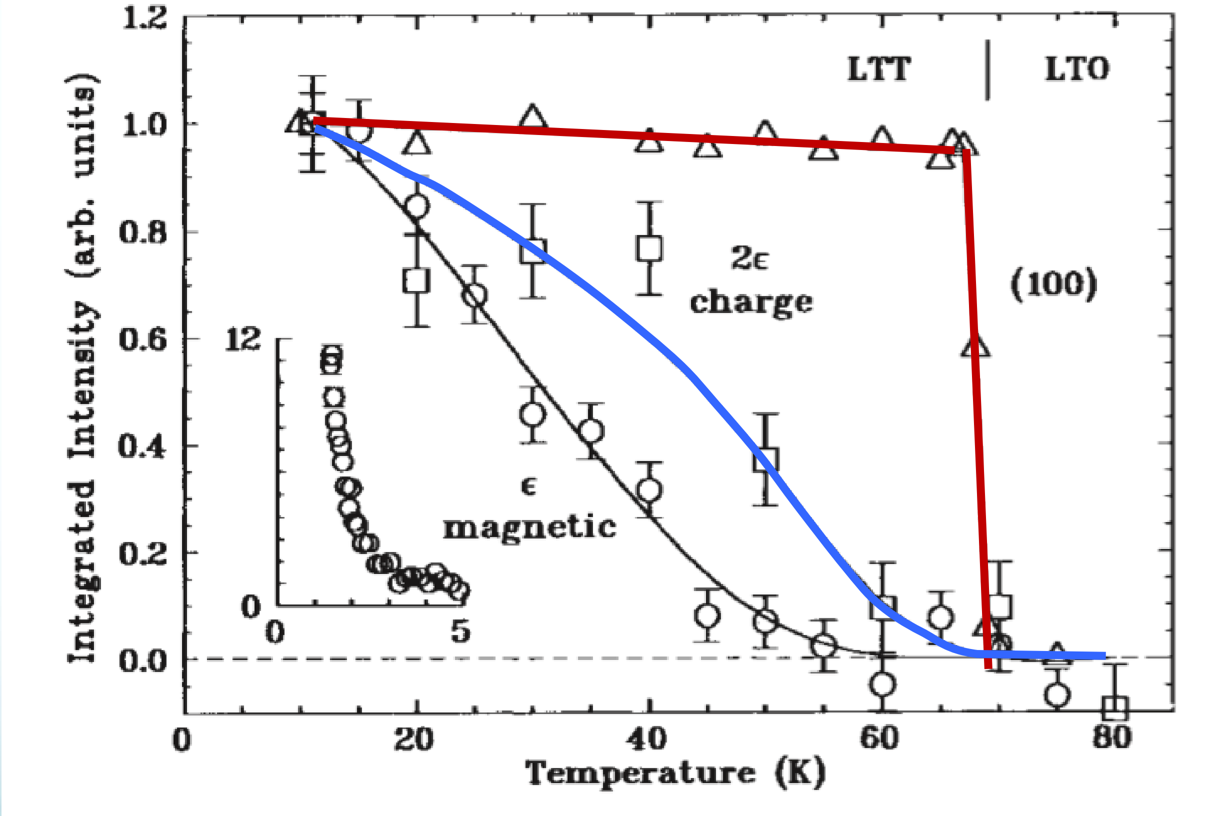
- Bragg peak due to spatial modulation in the electronic structure
- Intensity enhanced on resonance

Achkar et al. PRL **110**, 017001 (2013)

Structure, nematicity and CDW order in $(La,X)_2CuO_4$

Neutron scattering

$La_{1.475}Nd_{0.4}Sr_{0.125}CuO_4$

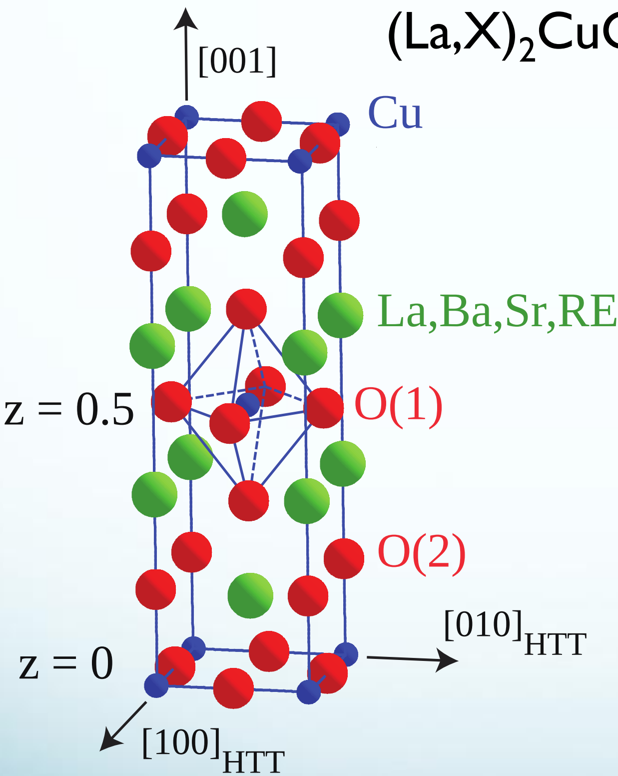


Charge density wave order onsets below 1st order structural phase transition: low temperature orthorhombic (LTO) to low temperature tetragonal (LTT)

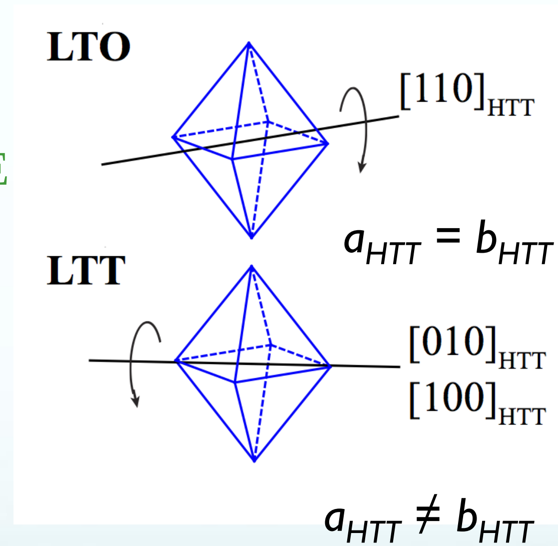
Tranquada Nature (1995)

Structure, nematicity and stripes in $(La,X)_2CuO_4$

$(La,X)_2CuO_4$



orthorhombic
CuO_2 planes

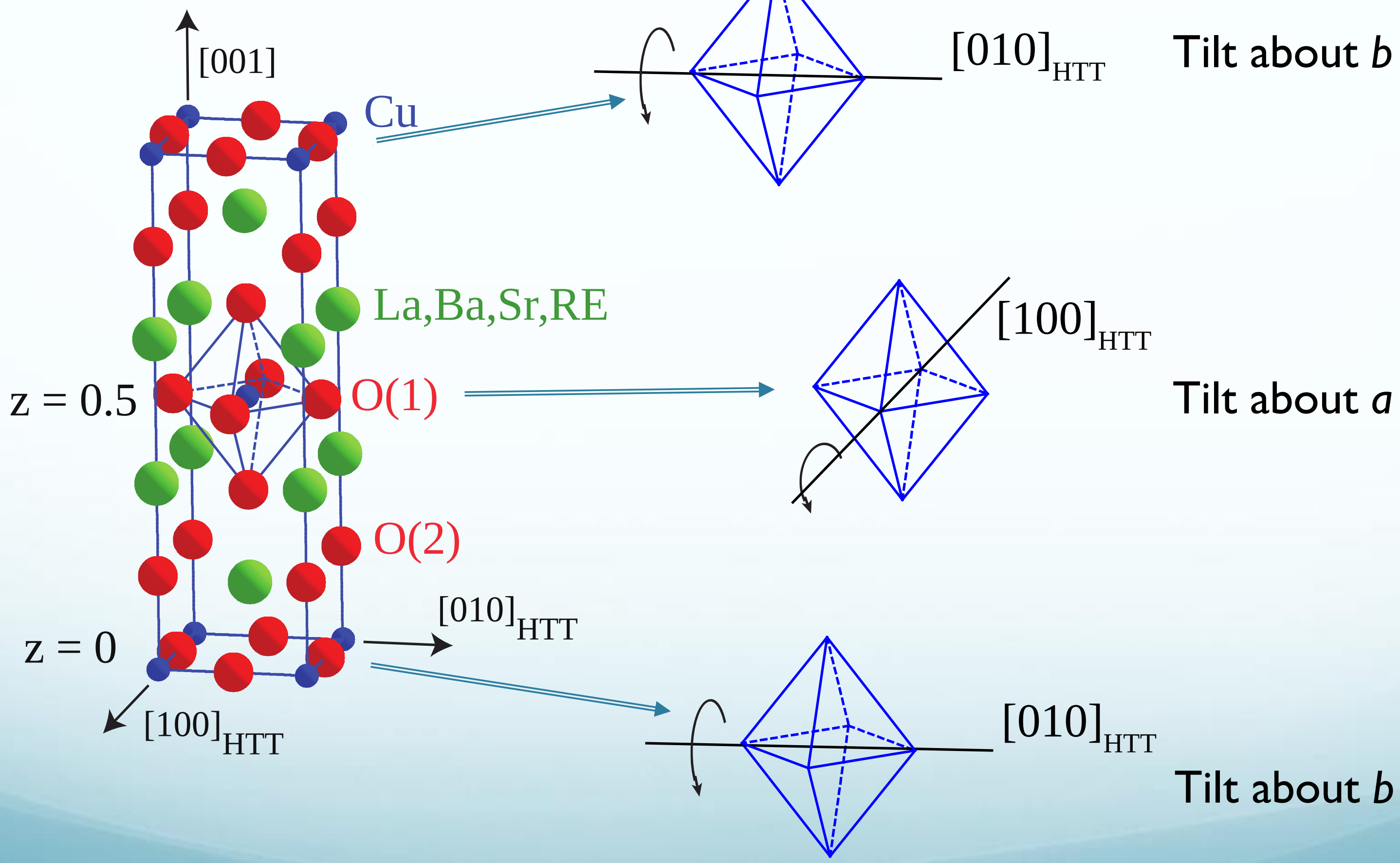


1st order LTO to LTT phase transition measured by x-ray and neutron scattering

Axe PRL 1989
Suzuki Physica C 1989
Tranquada 1995
Zhao PRB 2007
Kim PRB 2008
Fink PRB 2011
Wilkins PRB 2011
Hucker PRB 2011
…

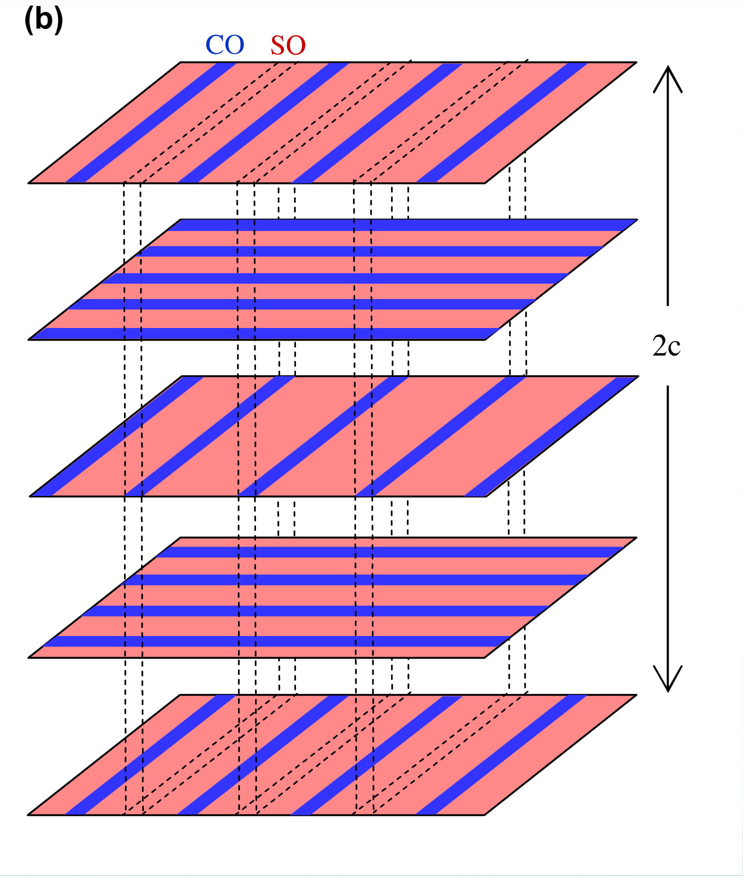
Low temperature tetragonal (LTT) structure

Tilt direction of octahedra alternates between neighboring planes



Unidirectional CDW order: stripes

La-based cuprates

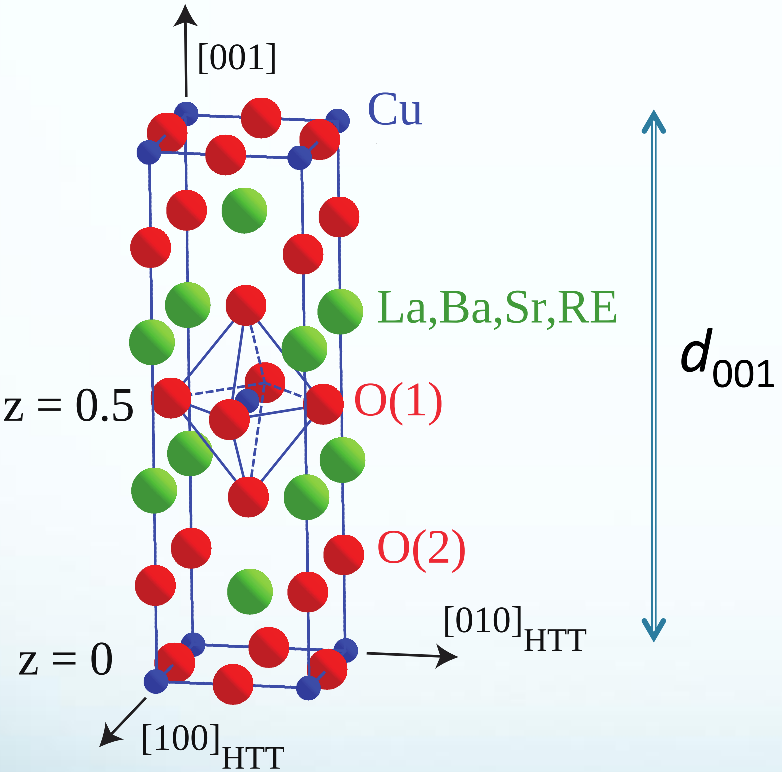


LTT tilts and stripe order

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

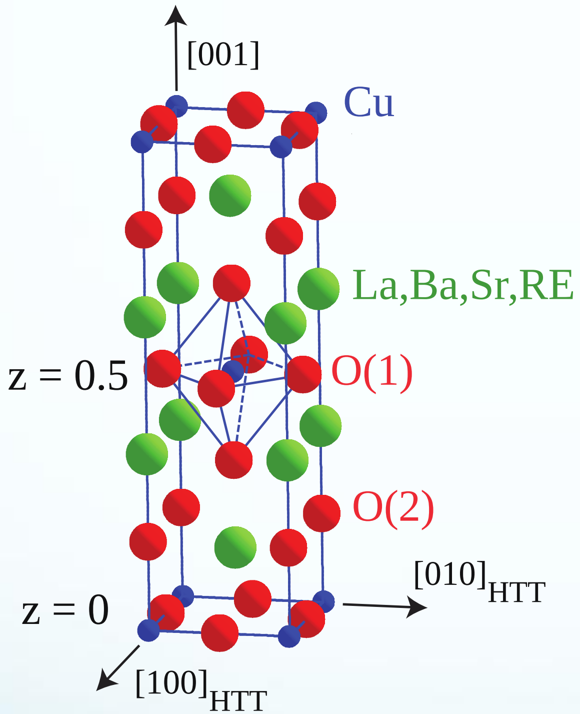
- Scattering from neighbouring planes destructively interferes

Resonant x-ray diffraction

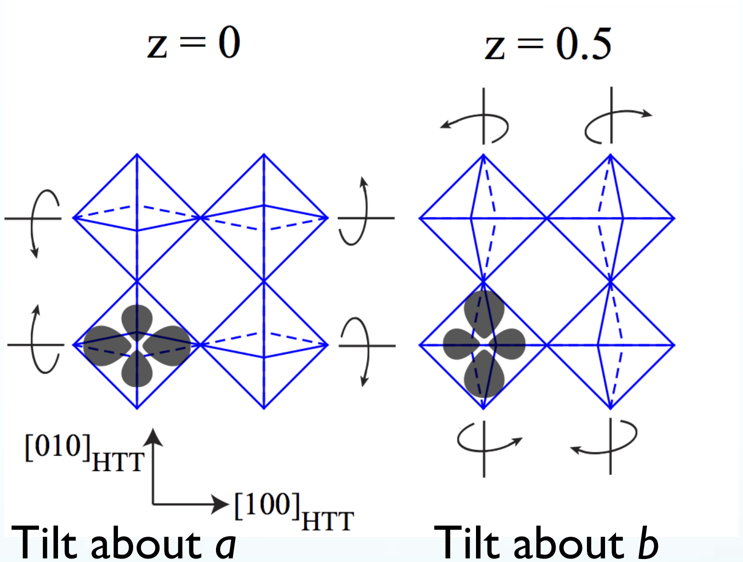
(001) Bragg peak is allowed

Fink et al. PRB 2011
Wilkins PRB 2011

Low temperature tetragonal (LTT) structure



Cu 3*d* states

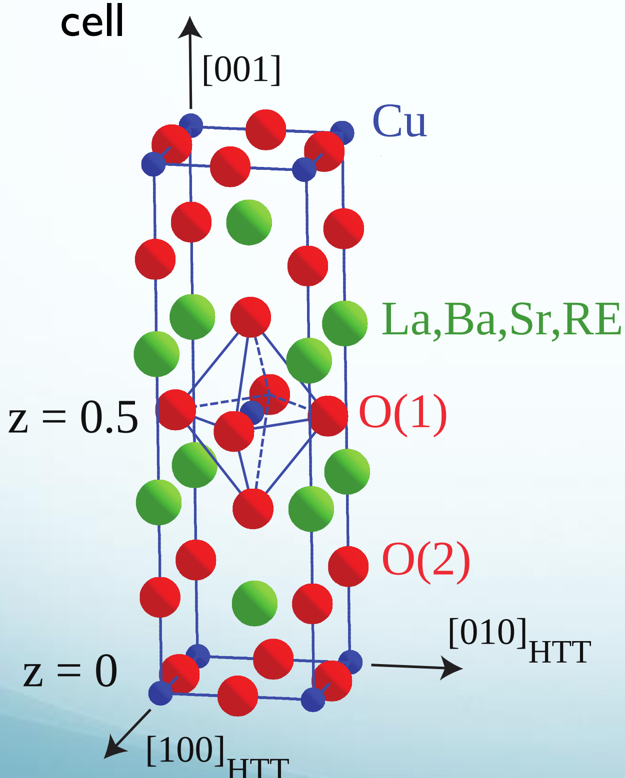


The octahedral tilting breaks the C_4 symmetry of the orbitals in each plane

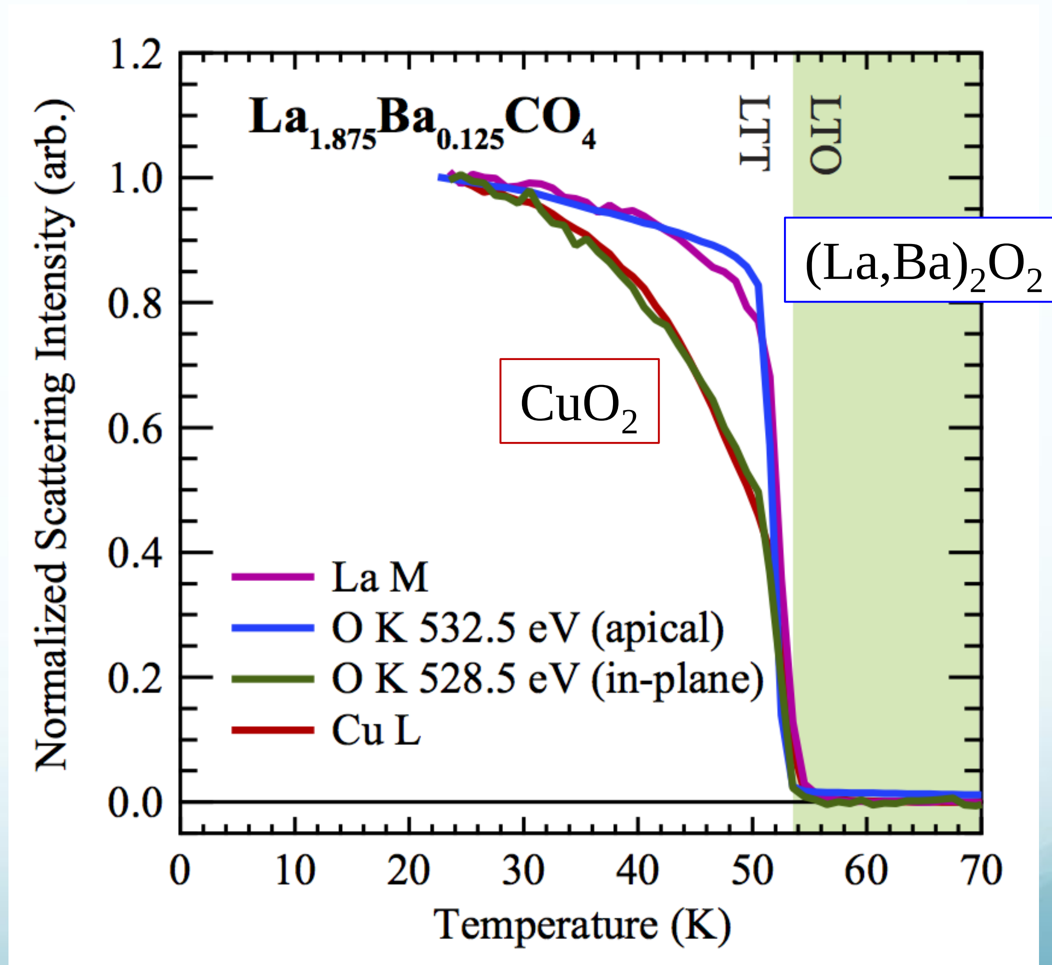
The (001) peak at the Cu *L* resonance measures electronic nematicity of the Cu 3*d* states

(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



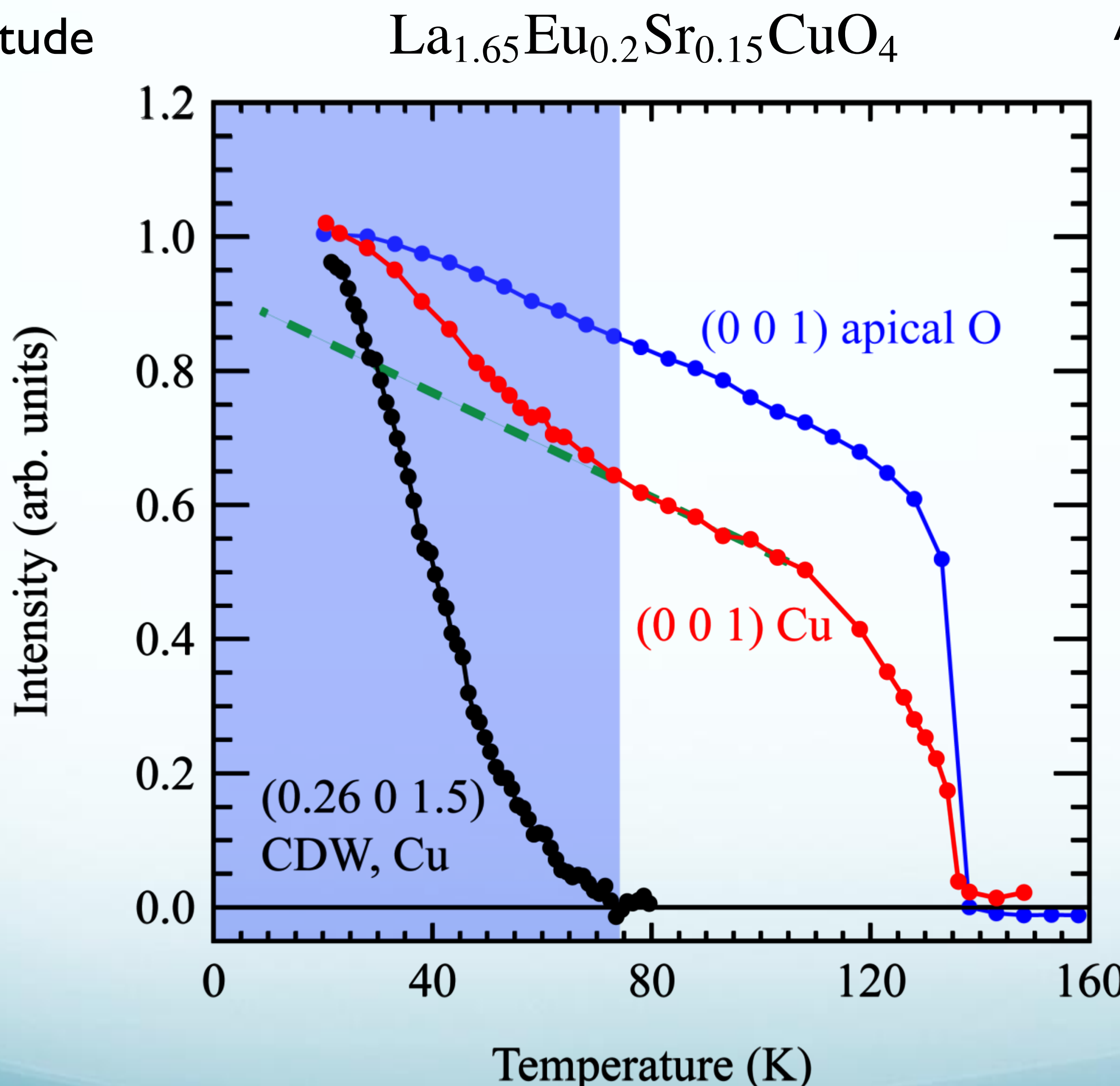
Achkar Science 2016



Relation to CDW order

Peak amplitude

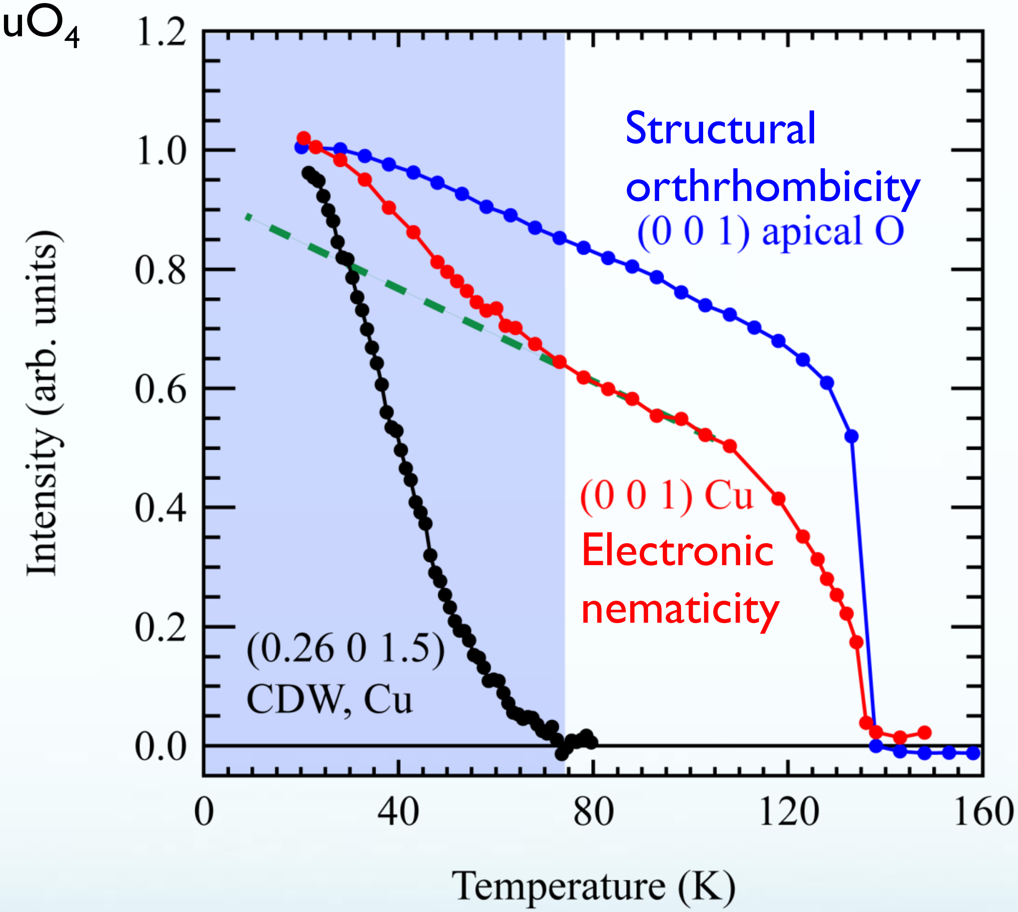
Achkar Science 2016



Distinct order parameters

$La_{1.65}Eu_{0.2}Sr_{0.15}CuO_4$

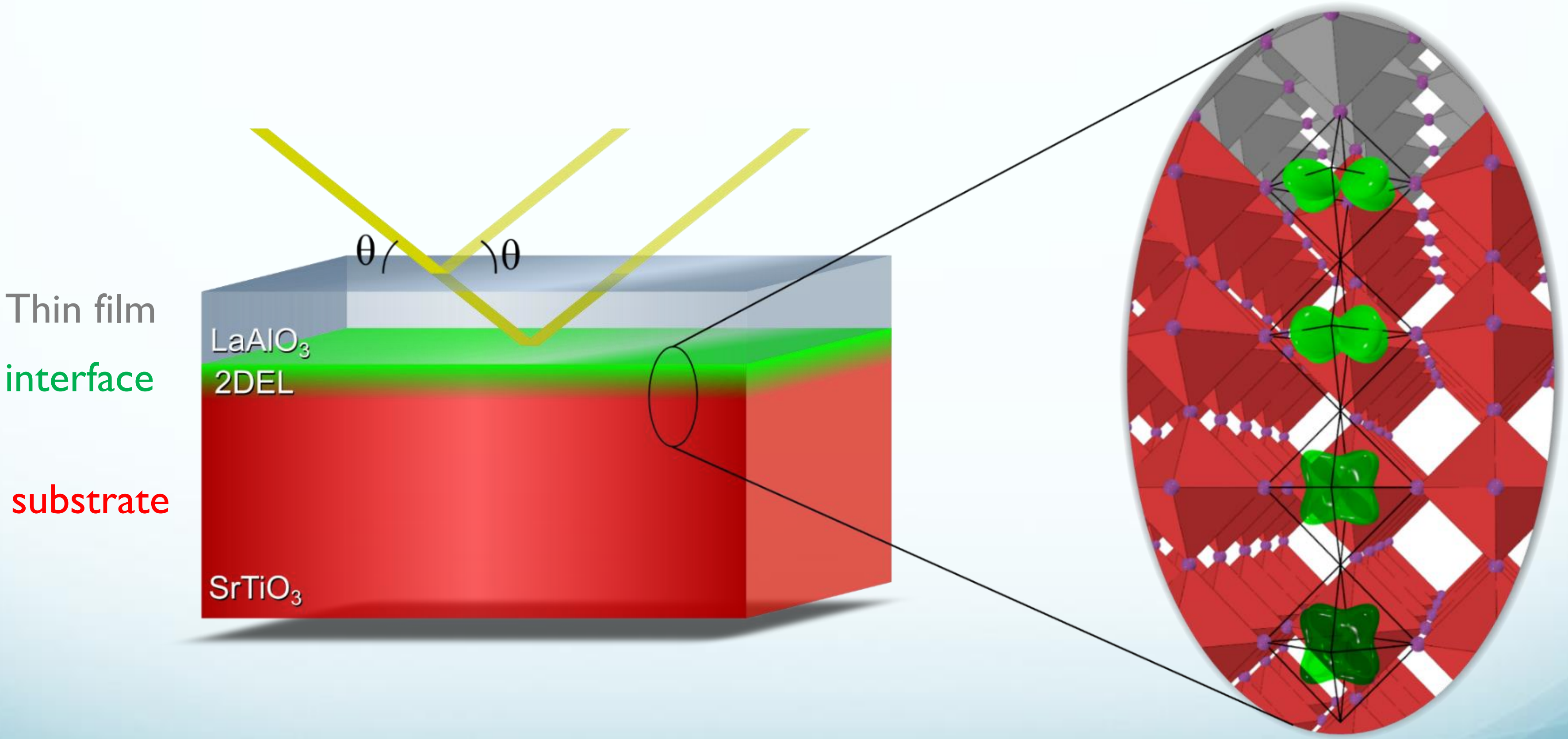
Achkar Science 2016



Distinct order parameters:

Electronic nematicity of the CuO_2 planes is coupled to, but distinct from the structural distortion of the $(La,X)_2O_2$ spacer layer

Probing Emergent Phenomena at Oxide Interfaces using Resonant X-Ray Reflectometry

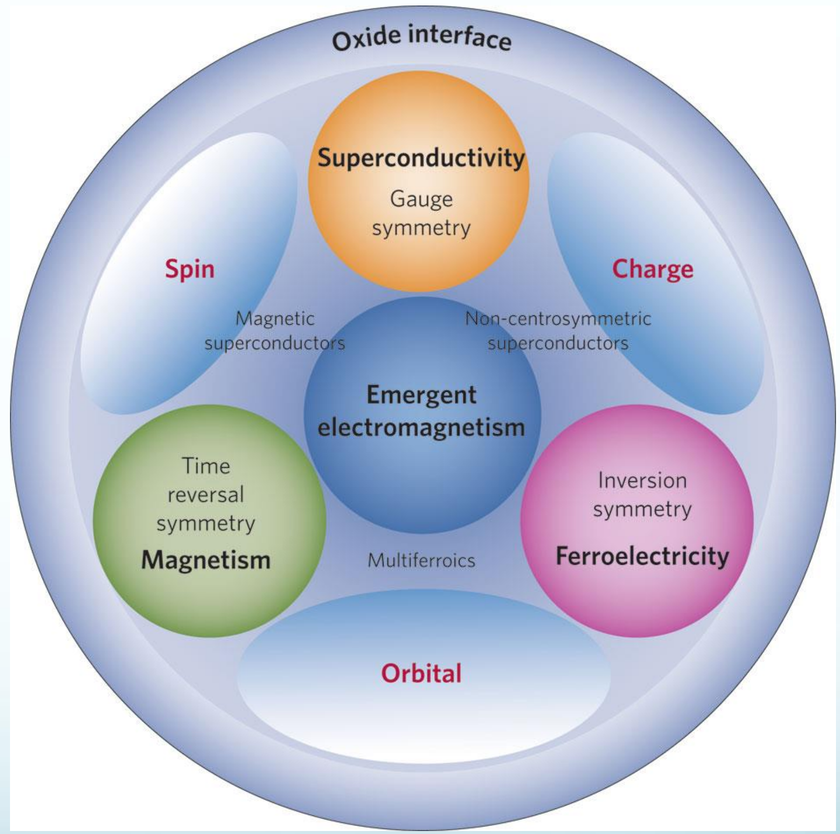


Emergent Phenomena at Oxide Interfaces

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

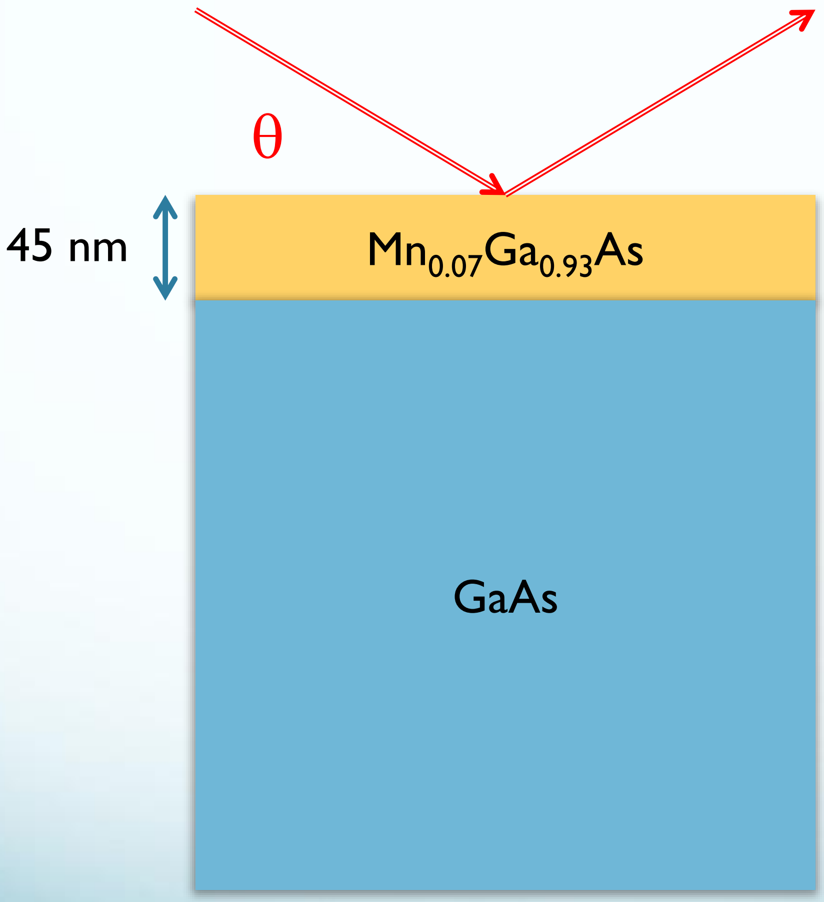


Hwang et al. 2012

Key Challenge in Studying Emergent Phenomena at Oxide Interfaces

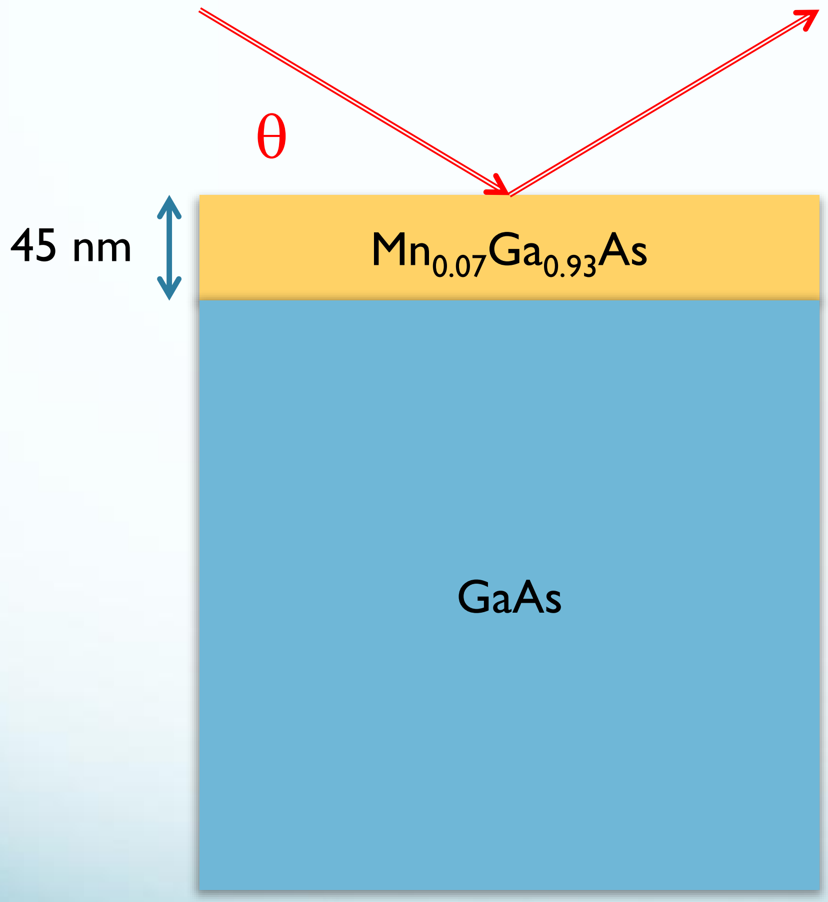
It is experimentally difficult to examine spin, charge, orbital reconstruction of buried interfaces. Many conventional experimental tools are impractical, lack sensitivity or are destructive.

Example: $Mn_{0.07}Ga_{0.93}As$ film on GaAs substrate

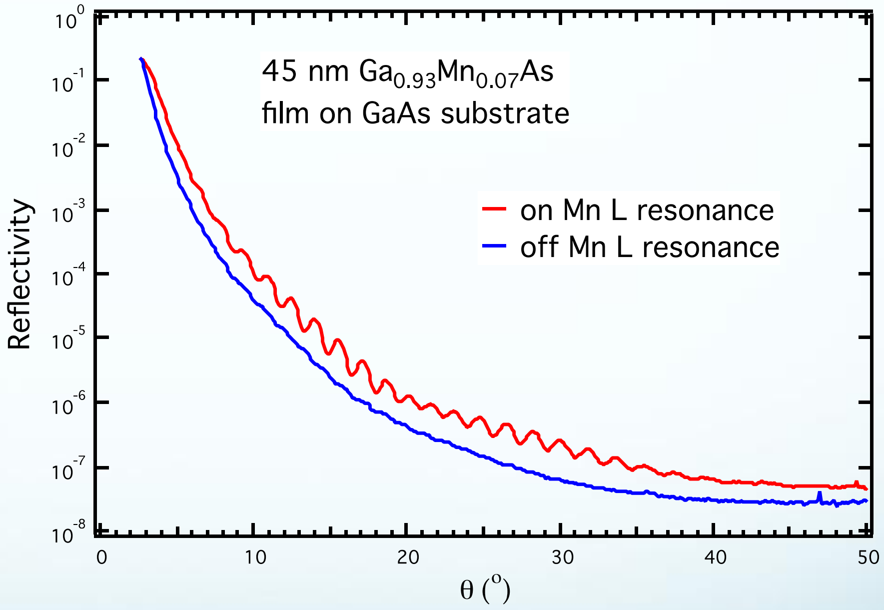


$Mn_{0.07}Ga_{0.93}As$ forms a magnetic semiconductor that is potentially useful for new generations electronics, spintronics, that make use of magnetic degrees of freedom

Example: $Mn_{0.07}Ga_{0.93}As$ film on GaAs substrate



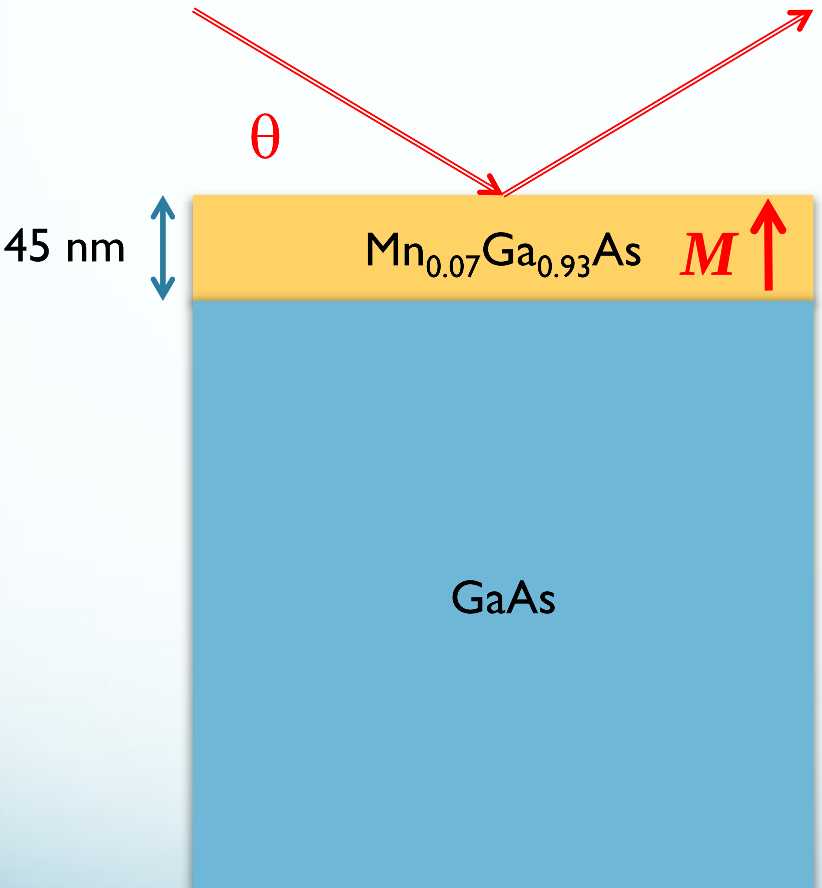
Interference fringes due to the 45 nm thickness of the film

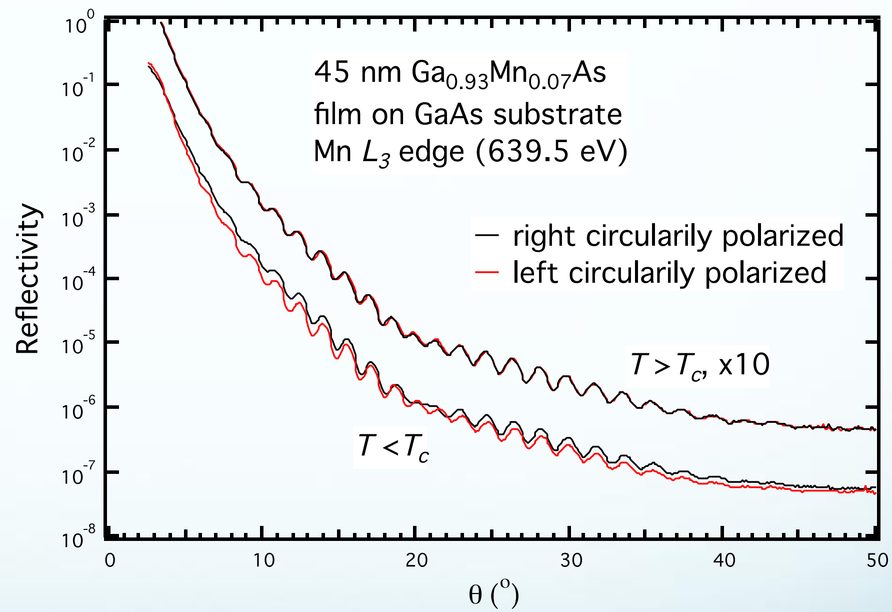


Measuring on resonance provides contrast, letting you "see" to the thin film

Hawthorn et al. Reviews of Scientific Instruments (2011).

Example: $Mn_{0.07}Ga_{0.93}As$ film on GaAs substrate



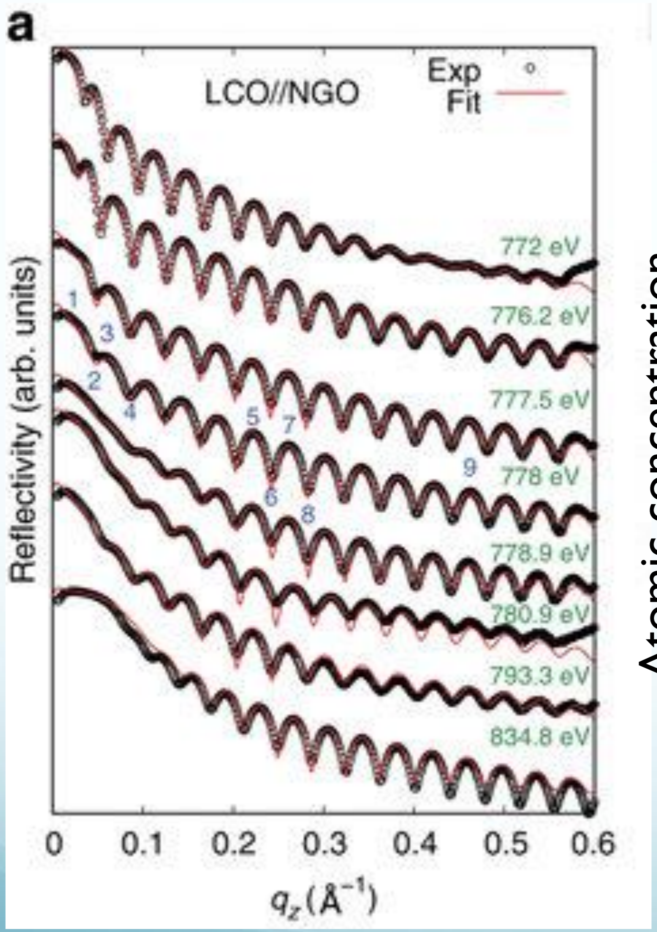


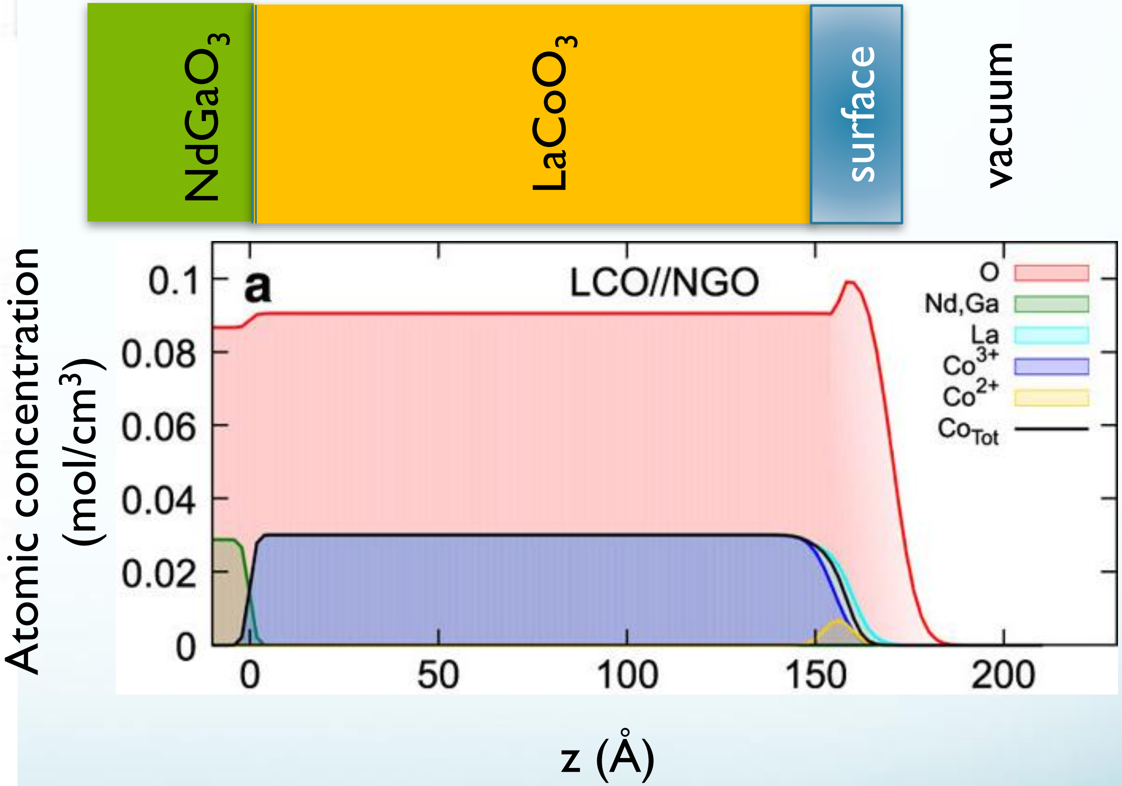
Resonant reflectivity lets you probe the electronic structure

Hawthorn et al. Reviews of Scientific Instruments (2011).

Chemical and electronic structure depth profiling

Example: Electronic reconstruction on the surface of $LaCoO_3$ film on an $NdGaO_3$ substrate

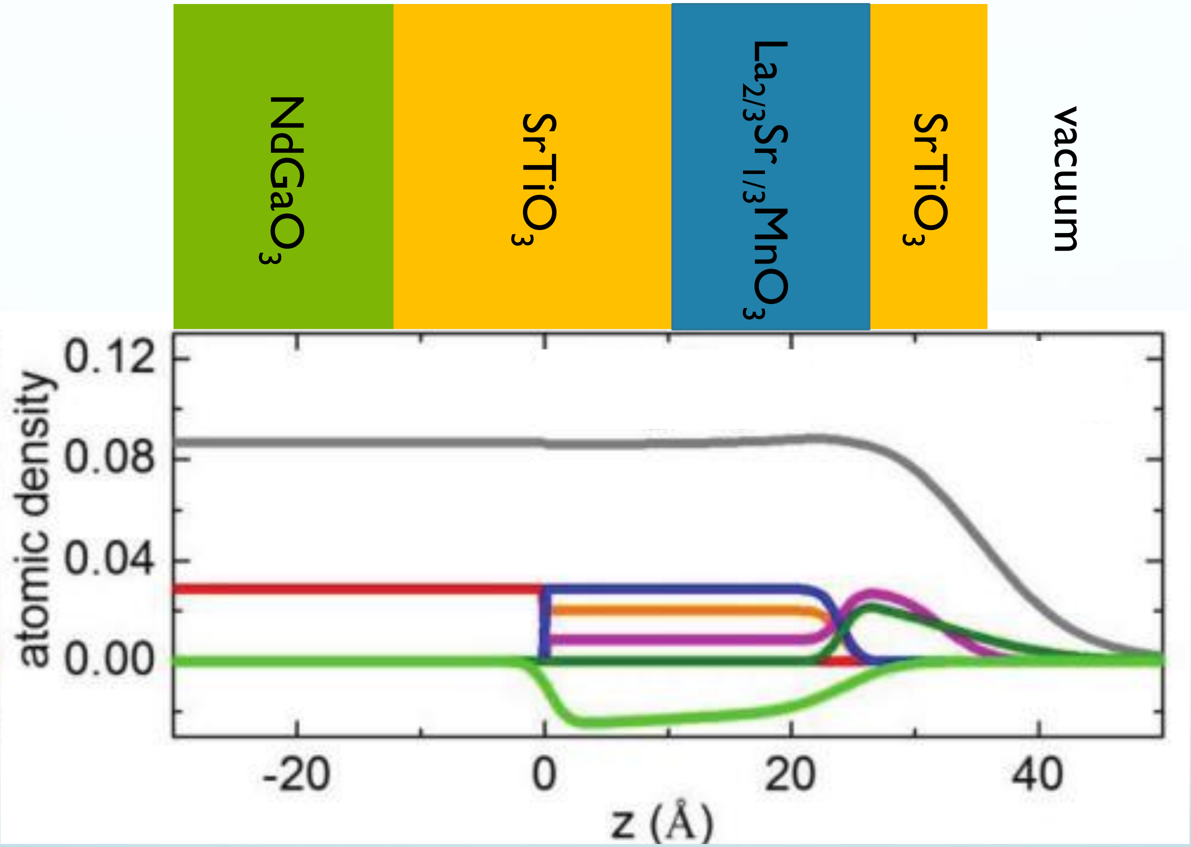




Hamann-Borrero et al. npj Quantum Materials (2016)

Chemical and electronic structure depth profiling

Example: Atomic layer resolved stoichiometry AND magnetic structure

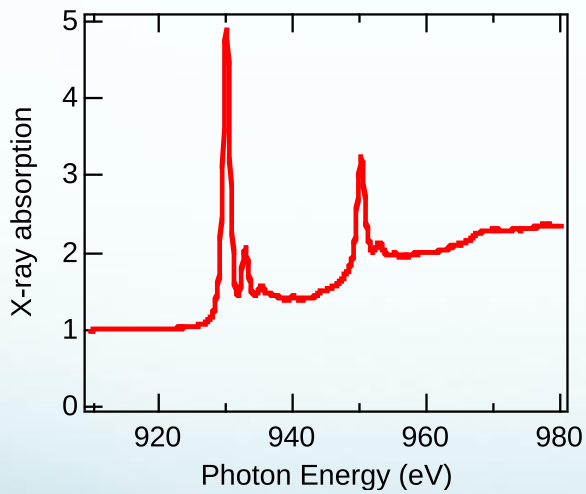


Liao et al. Advanced Functional Materials (2017)

Conclusions

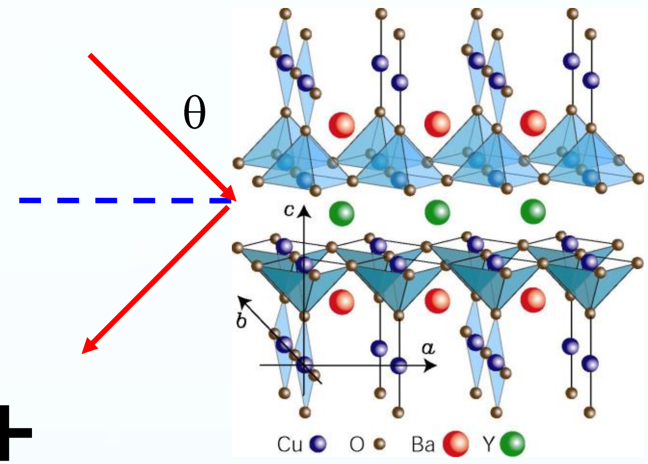
Resonant x-ray diffraction and reflectometry provide a unique, element and orbital specific probe of spin, charge and orbital symmetry breaking in crystals and thin films

x-ray spectroscopy



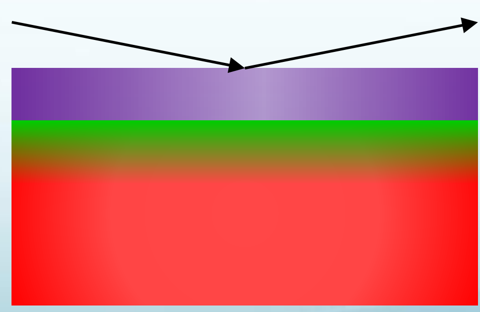
electronic structure

+



x-ray diffraction (in crystals)

OR



x-ray reflectometry (in thin films and multilayers)

spatial structure