Resonant Soft X ray scattering

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Collaborators

Scattering:

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E.Isaacs, G.Bloomberg, A. Gozar ------ Bell

Samples:

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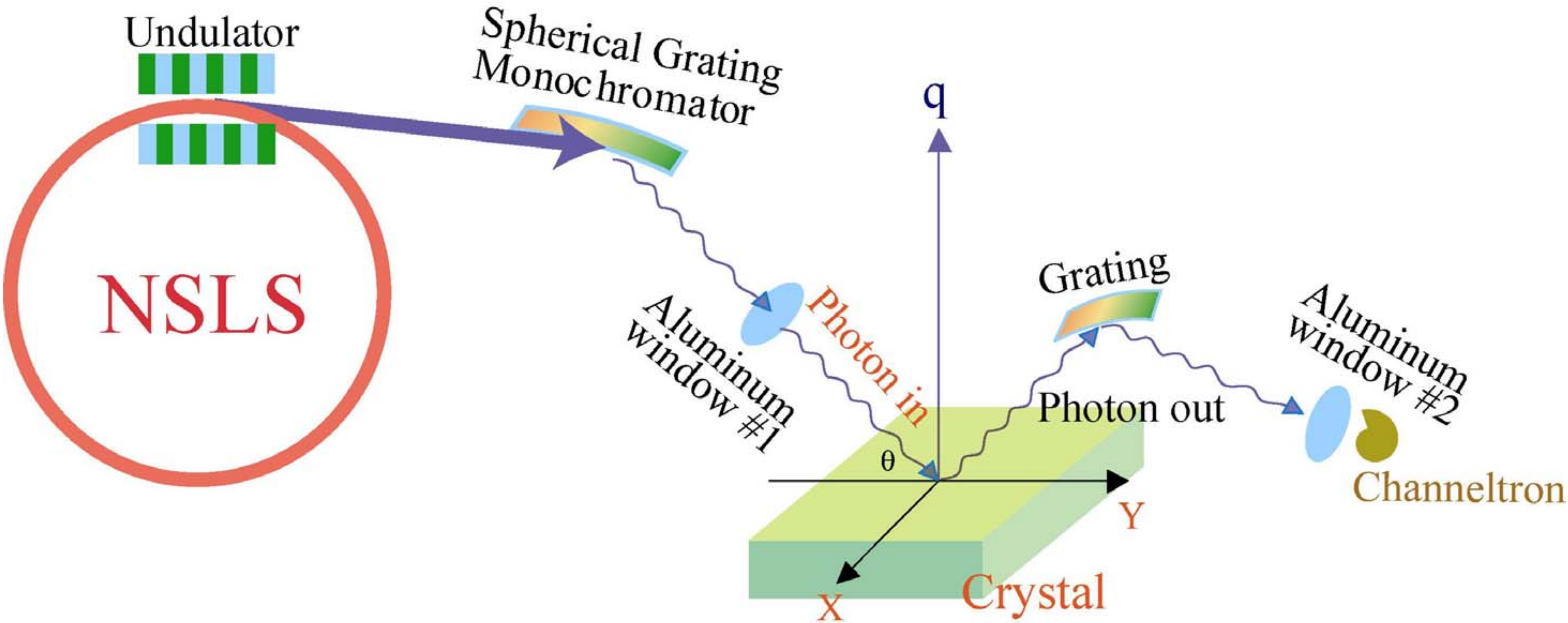
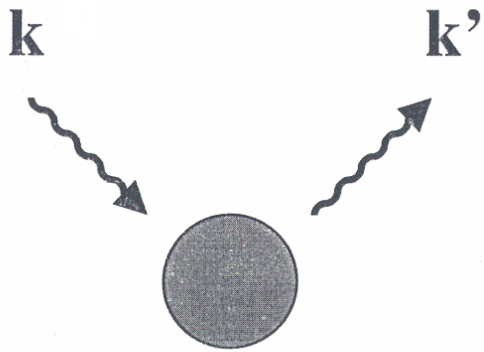
H.Eisaki, S.Uchida ---- Tokyo

Experimental Setup



- NSLS, undulator beamline X1B, photon energy: 85~1800 eV
- about $5*10^{12}$ photon/sec at a typical 1000 resolving power
- Removable Aluminum windows to block low energy stray light
- XAS taken in Fluorescence/Electron yield modes
- Removable 2^{nd} grating to select the primary energy
- 2.4 m sized UHV chamber, 10 motions, T=30~400K

X-ray Scattering

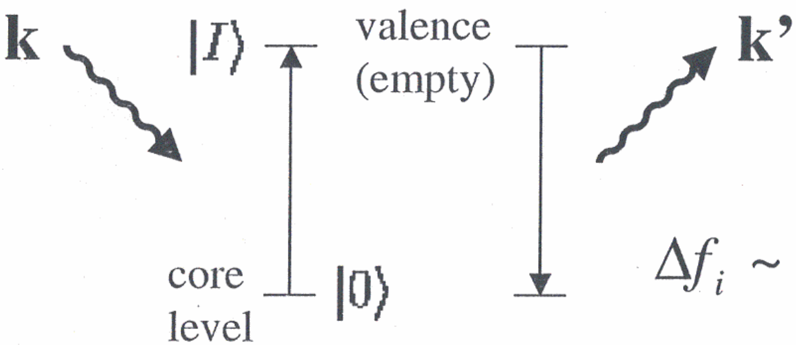


$$\mathbf{G} = \mathbf{k}' - \mathbf{k}$$

$$f_i^{\,0} = 4\pi \int dr\, n_i(r)\, r^2 \frac{\sin(\vec{G}\cdot\vec{r})}{|\vec{G}\cdot\vec{r}|}$$

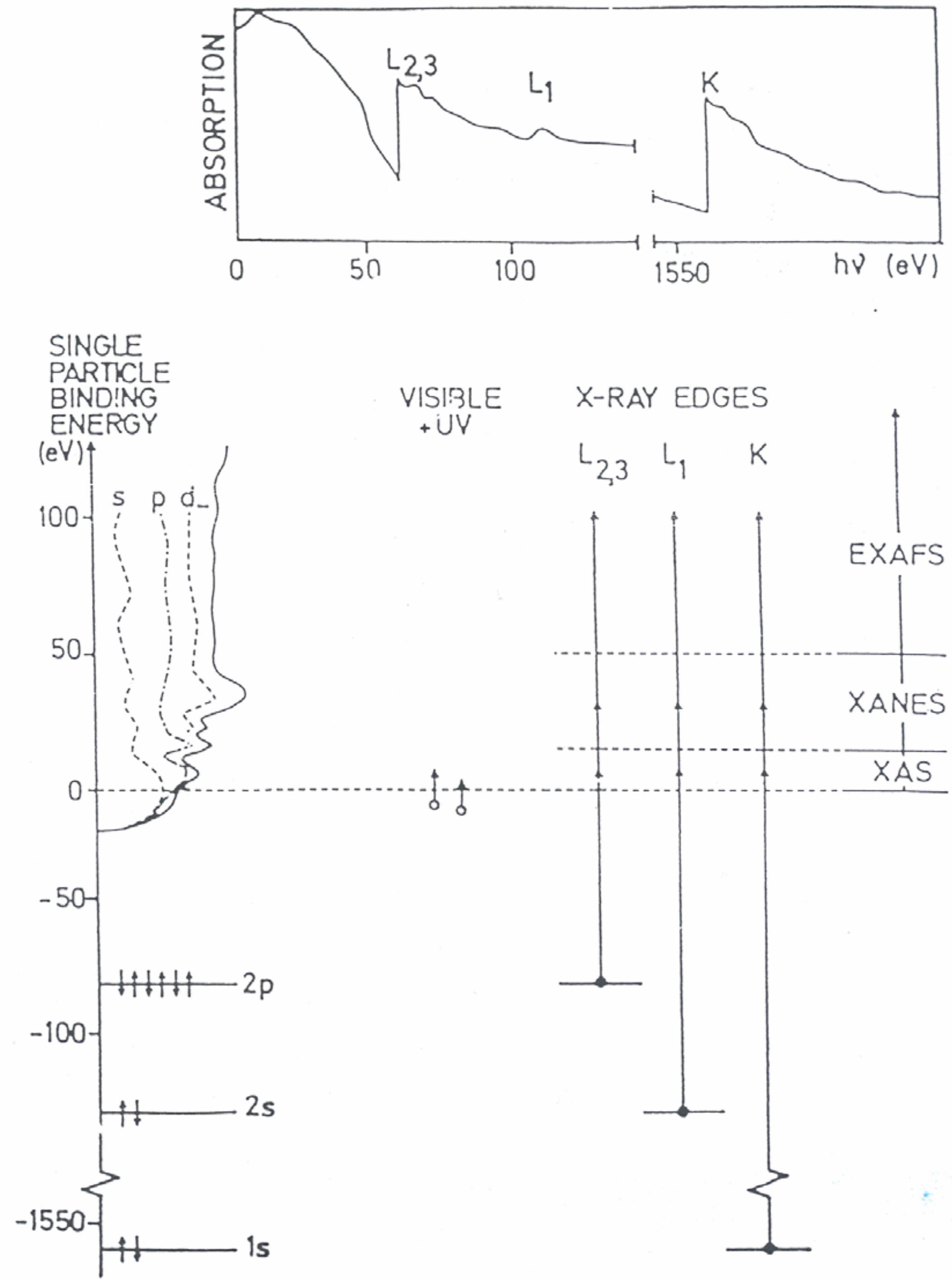
$n_i(r)$ → electron number density

Resonant X-ray Scattering



$$\Delta f_i \sim \sum_I \frac{\langle 0|\vec{\varepsilon}\cdot\vec{r}\exp(i\vec{k}\cdot\vec{r})|I\rangle\langle I|\vec{\varepsilon}\,'\cdot\vec{r}\exp(i\vec{k}\,'\cdot\vec{r})|0\rangle}{\hbar\omega - (E_I - E_0) - i\Gamma}$$

We learn about the spatial distribution of states $|I\rangle$



ABSORPTION
$L_{2,3}$
L_1
K
0
50
100
1550
hν (eV)
SINGLE PARTICLE BINDING ENERGY (eV)
s
p
d
100
50
0
-50
-100
-1550
2p
2s
1s
VISIBLE +UV
X-RAY EDGES
$L_{2,3}$
L_1
K
EXAFS
XANES
XAS

At resonance we have contrast for:

- Elements
- Valence electron density
- Bond orientation; orbital ordering
  quadrupole moment orientation
  [linear pol. light]
- Spin density [circular pol. light and
  p or *d* core level]

- Small crystals
- (ultra) thin films [few atoms]
- Multilayers
- Nano structures

Types of Materials

- Magnetic – Spintronics
- Superconductors
- Molecular solids – CMT, DNA
- Molecular Magnets

Some applications

Magnetic nano structures (Domains) (Speckle)

Charge density waves

Intrinsic phase separation (high T_C, Colosal Mag. Resis.)

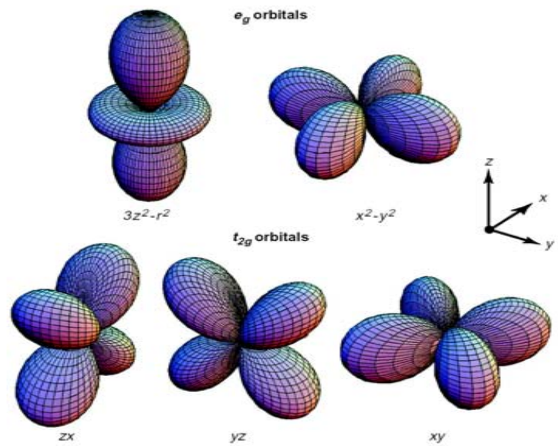
Structure of Self assembled Molecular Systems (thin films)

Distribution of Specific groups in Cells – Macromolecules
(follow folding)

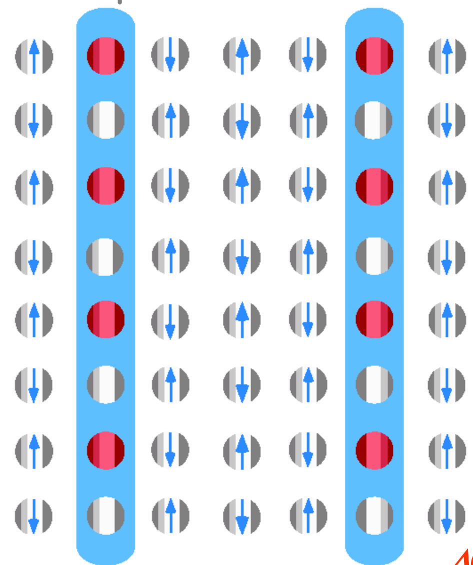
Orbital ordering and phase separation

T @ H Dep. local magnetic moments and long range order
(ultra thin films)

Ordering in strongly correlated systems



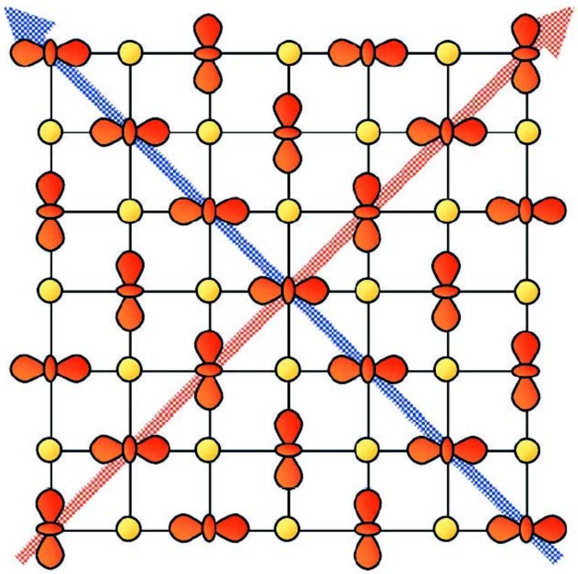
Stripes in Nd-LSCO



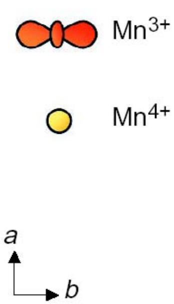
$\Delta Q < 0.5$ e

$\Delta Q/Q_{total} \sim 1/500$

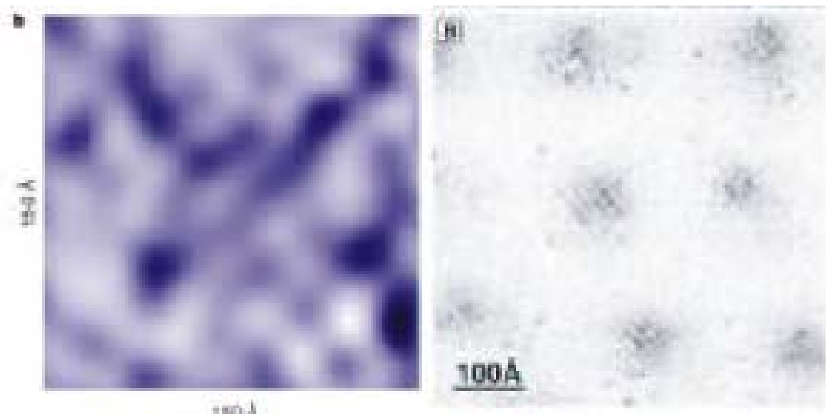
charge/orbital ordering in $La_{0.5}Sr_{1.5}MnO_4$



$\Delta Q_C \sim 1$ e
$\Delta Q_O \sim 0$



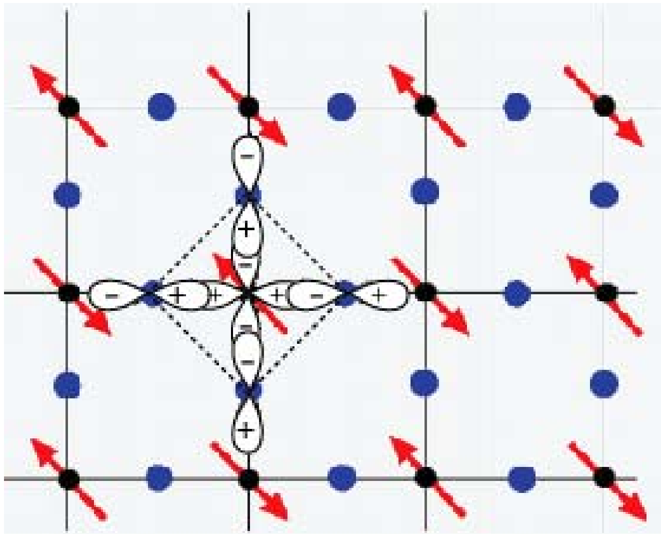
Charge inhomogeneity in Bi2212

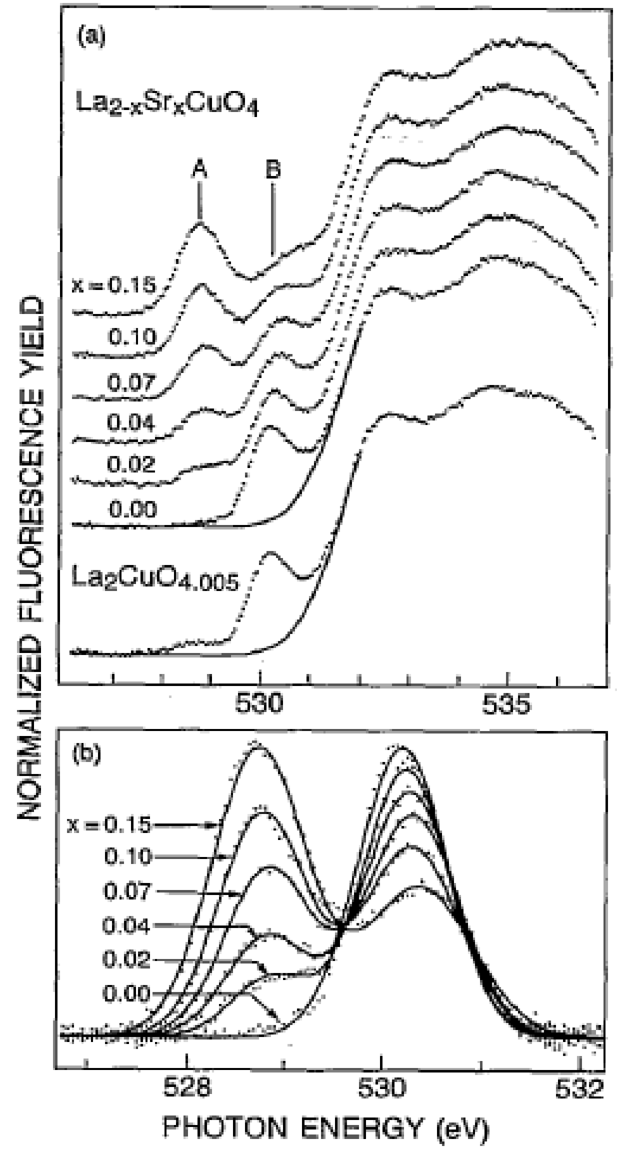


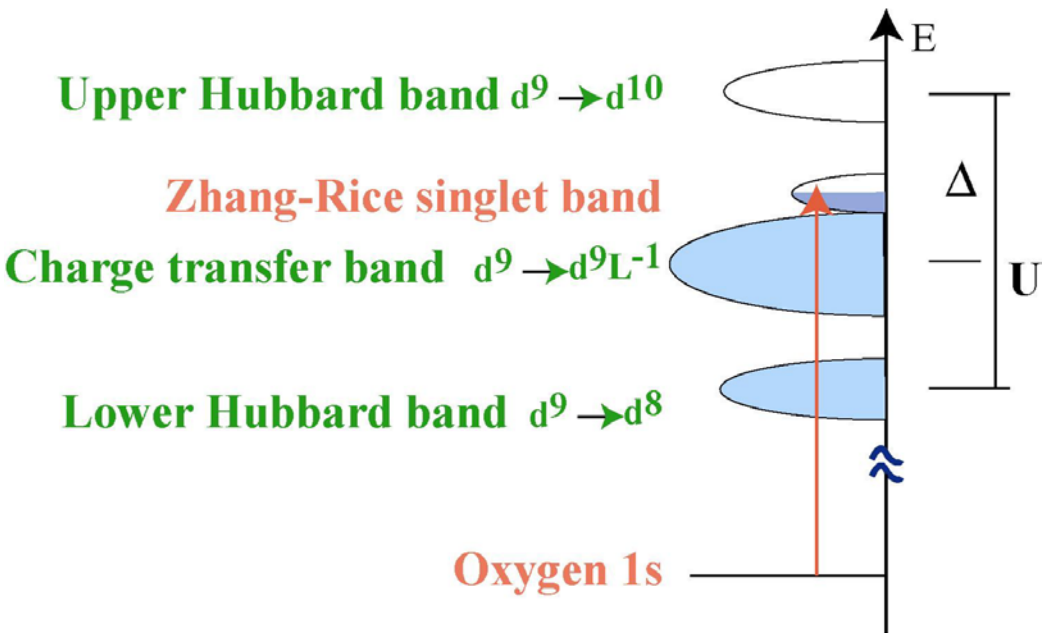
Pan, *Nature*, **413**, 282 (2001);
Hoffman, *Science*, **295**, 466 (2002)

$\Delta Q \sim 0.1$ e

Doped holes in cuprate







C. T. Chen et al. PRL 66, 104 (1991)

Reminder

$$\frac{\lambda}{1\text{Å}} = \frac{E}{12000\ \text{eV}}$$

<u>3*d* Transition Metal Compounds</u>

$L_{2,3}$ edge $2p \rightarrow 3d$

500 eV → 900 eV

20 Å → 12 Å

<u>Rare Earth's (4*f* compounds)</u>

$M_{4,5}$ edge $3d \rightarrow 4f$

800 eV → 1800 eV

12 Å → 8 Å

C_{1s} - 280 eV → 40 Å

N_{1s} - 390 eV → 35 Å

O_{1s} - 530 eV → 25 Å

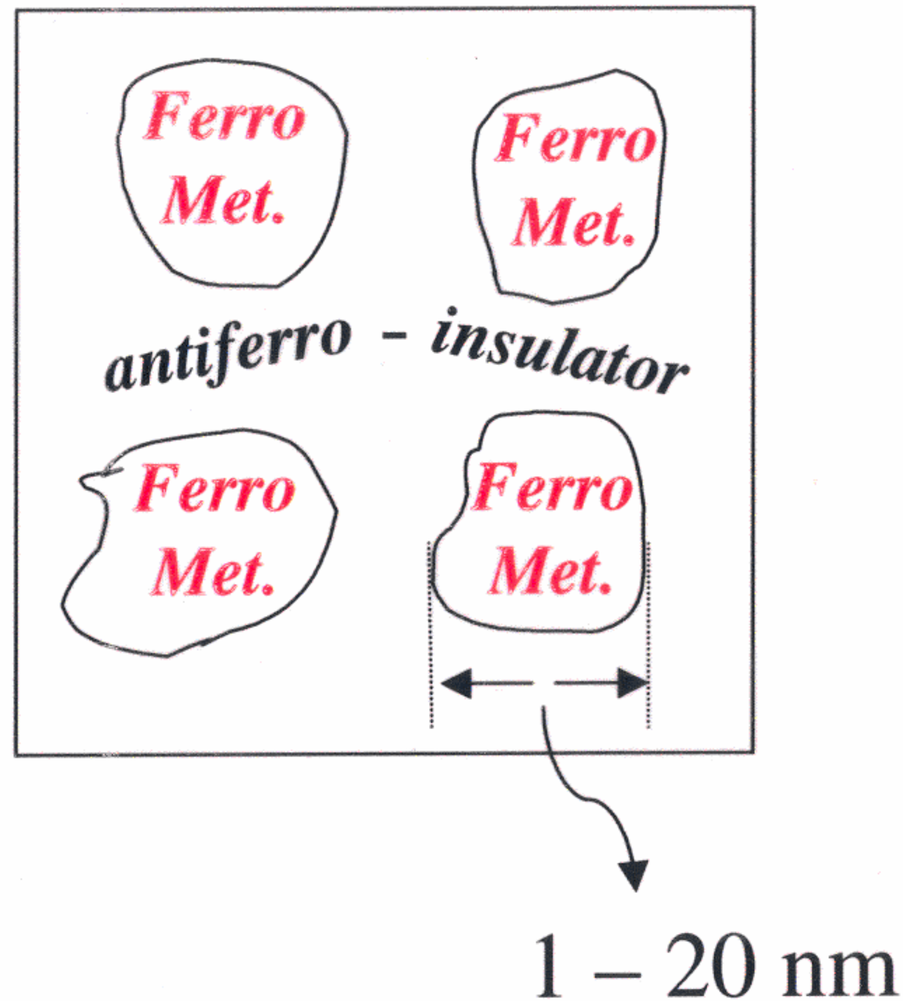
S_{1s} - 3000 eV → 4 Å

Phase Separation

- Stripes in High T_C's?
- Magnetic droplets in manganites

Diffuse Scattering

Exp. at $L_{2,3}$ edge would be much more sensitive than at the K edge



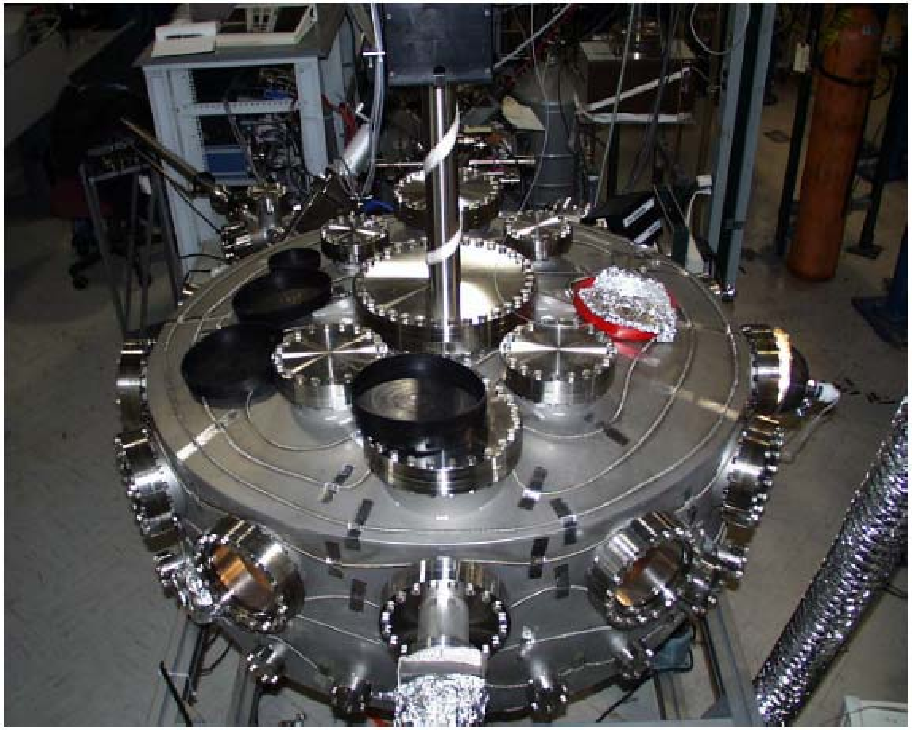
Resonant X-ray Scattering

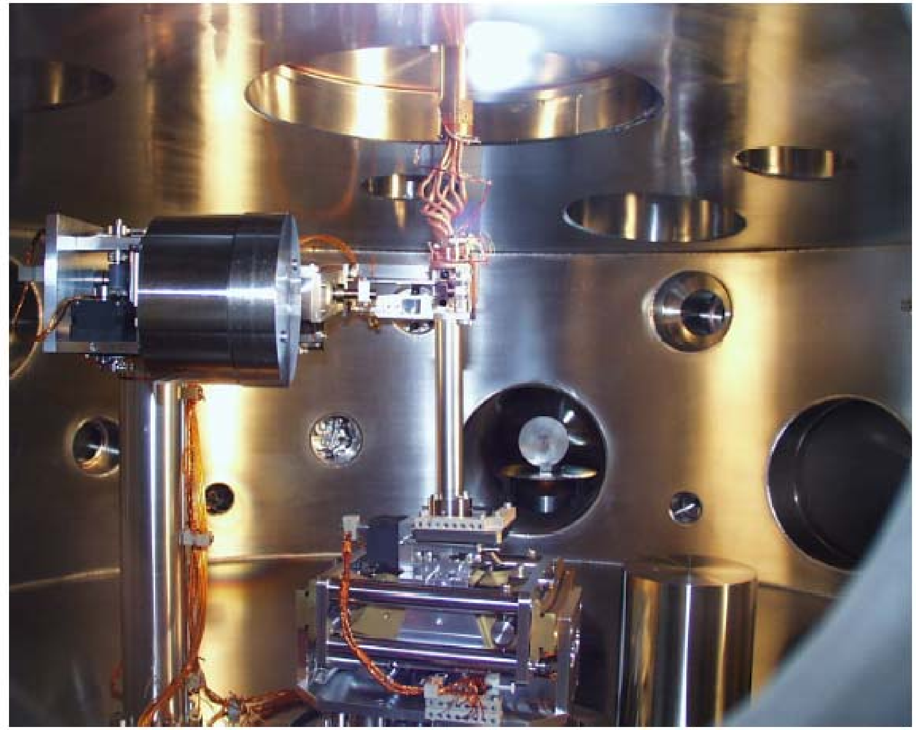
- Resonant High resolution X-ray diffraction
- Resonant diffuse scattering (Speckle)
- High resolution X-ray reflections
- Magnetic X-ray dichroism
- Polarization dependent X-ray absorption spectroscopy

Resonant X-ray diffraction

- Element specific structure
- Molecular group specific structure
- Spin, orbital, and charge dependent structure
- Surface and interface sensitivity

Experimental Station

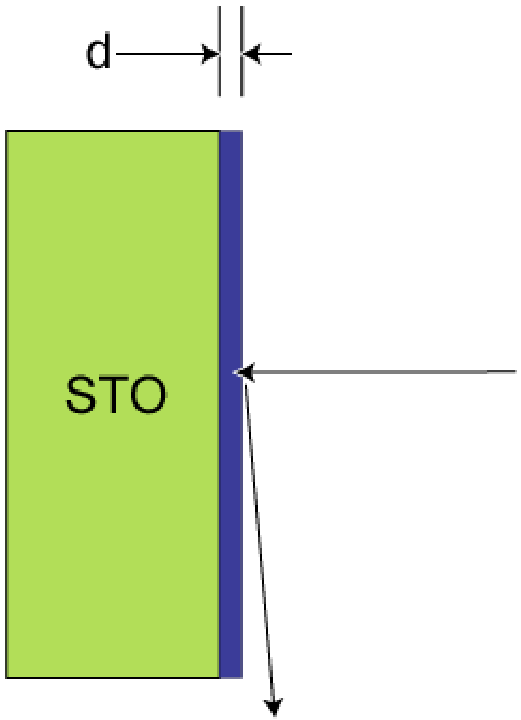




- 2.4m diameter vacuum chamber
- 5 circle geometry (10 motions)
- Integrated beamline + end station control

- Helium flow cryostat (15~400K)
- 5 Tesla magnet (vertical)
- UHV, Base pressure ~ 10^{-10} *torr*

XAS at Oxygen K and Copper L edge



Epitaxial $La_2CuO_{4+\delta}$ on $SrTiO_3$

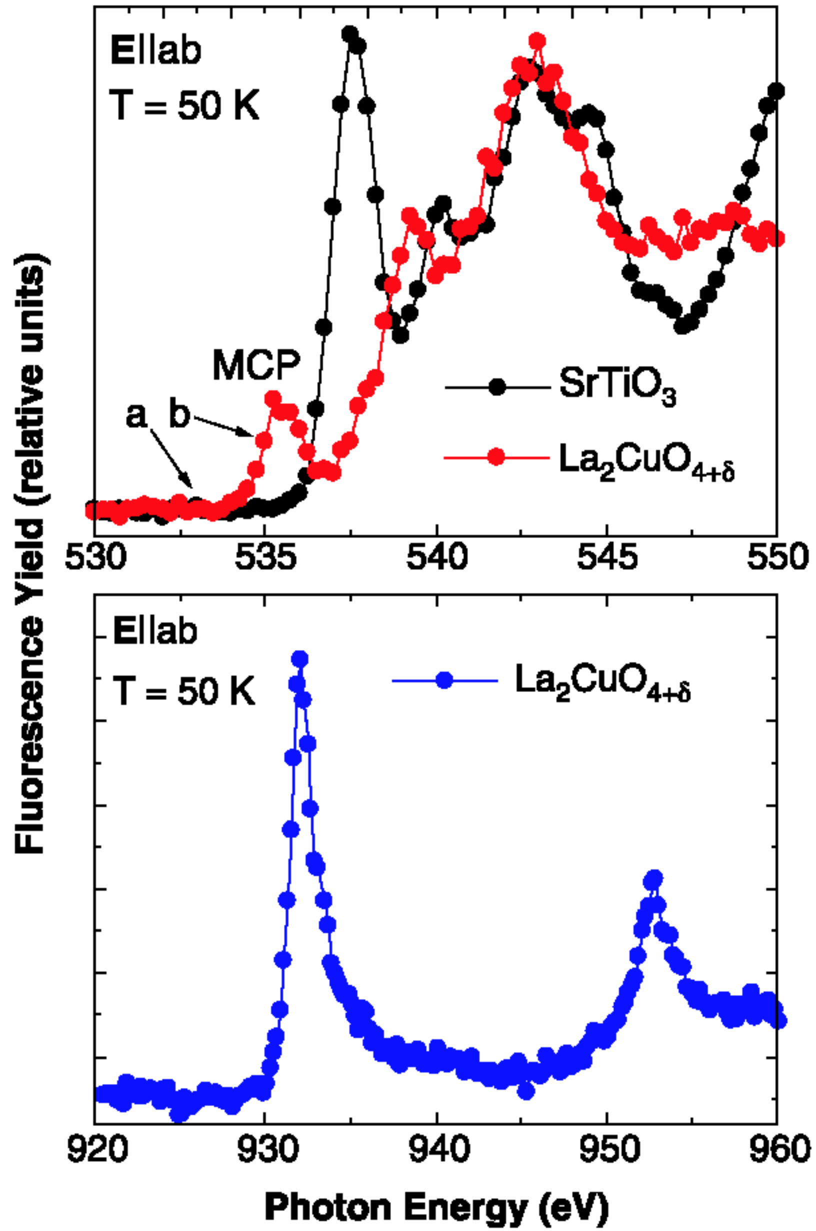
(d = 23.3 nm, Tc = 39 K)

$2d\ sin(\theta) = n\ \lambda$

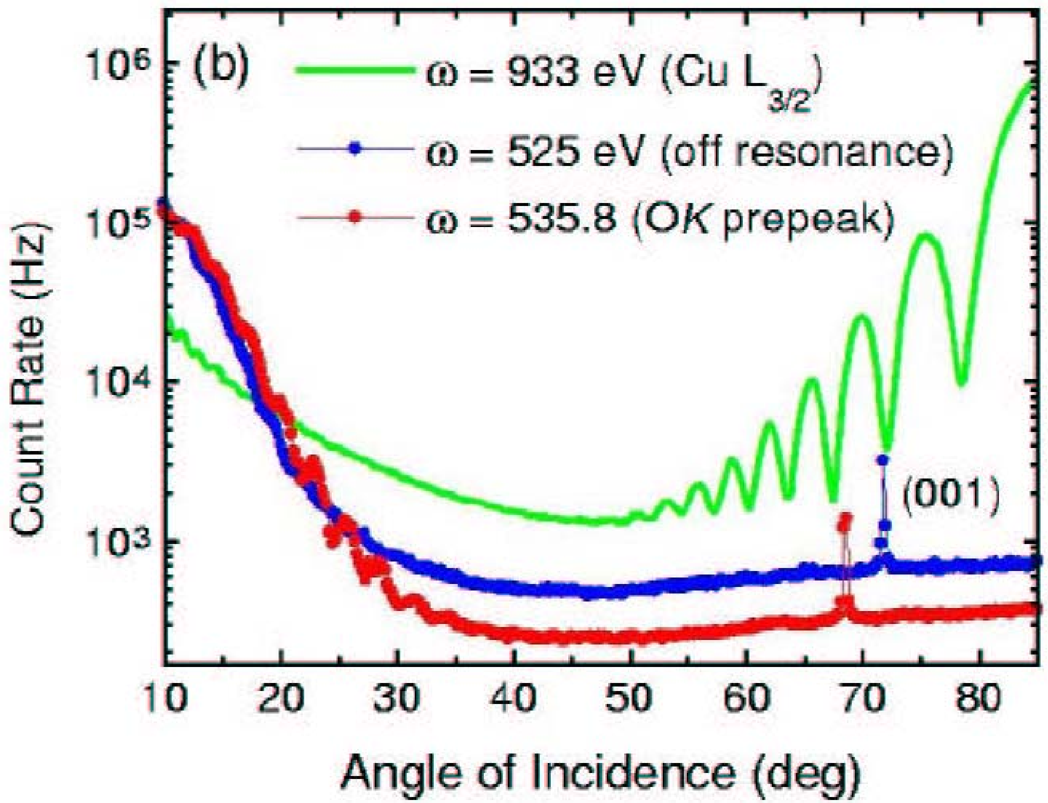
n_{STO} = 5.12 g/cm3

n_{LSCO} ~ 5.5 g/cm3

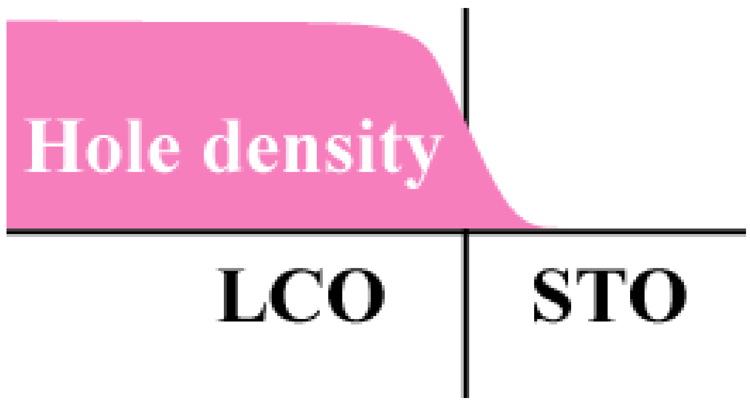
T=50K

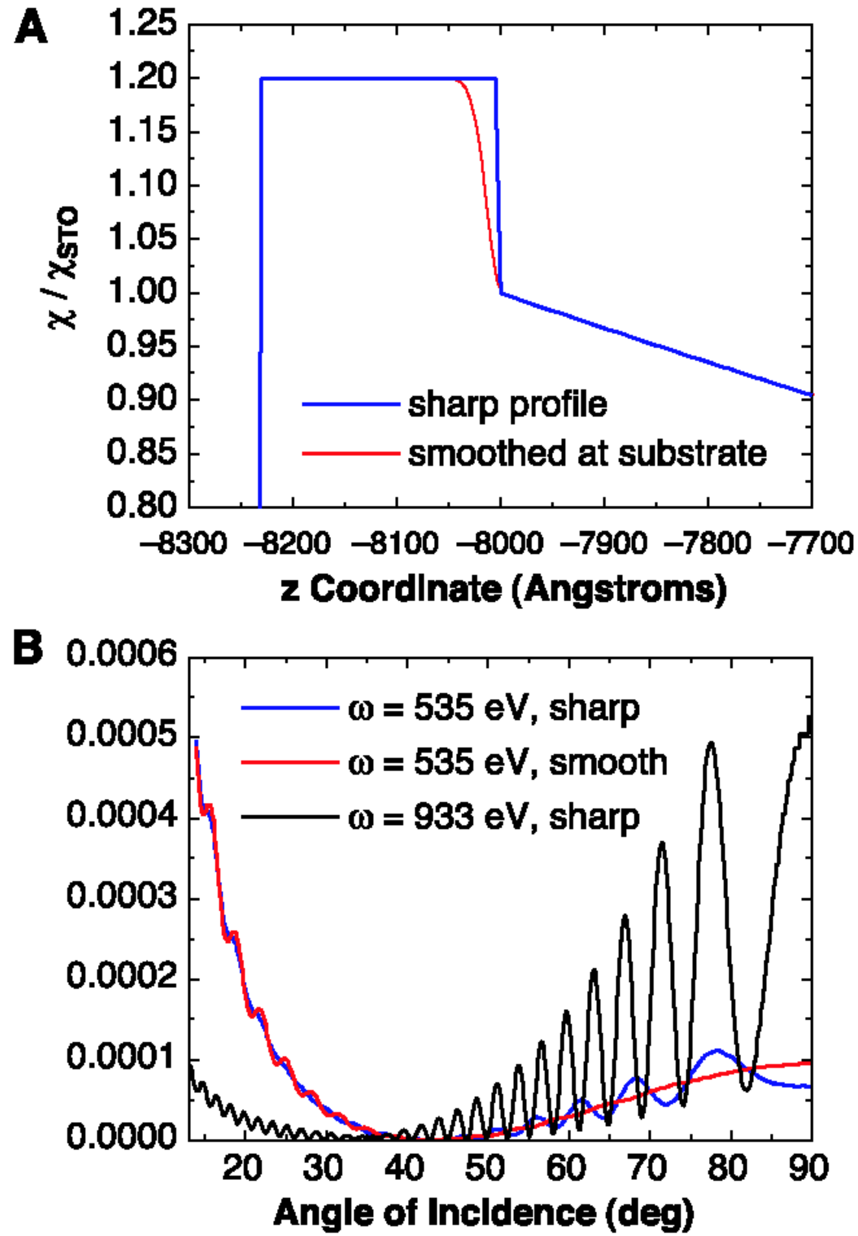


Anomaly in Fringe Envelope



Hole depletion model

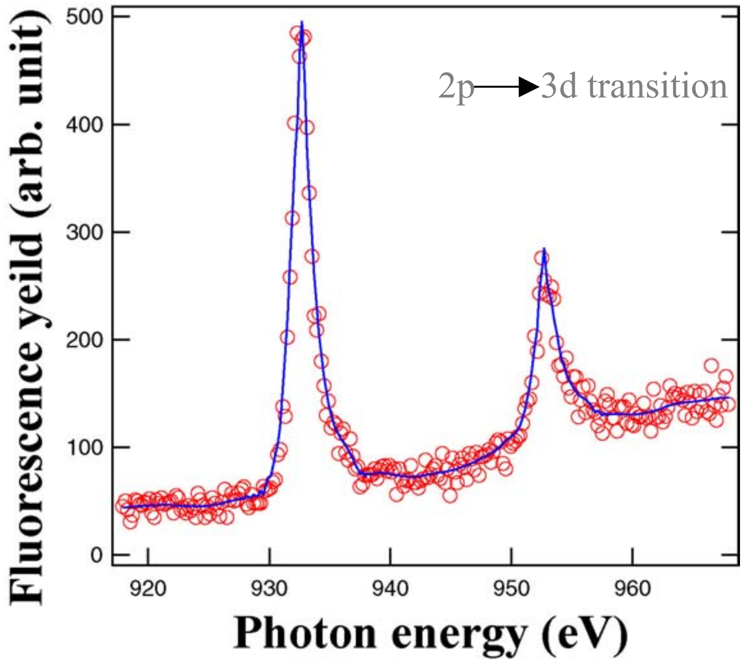


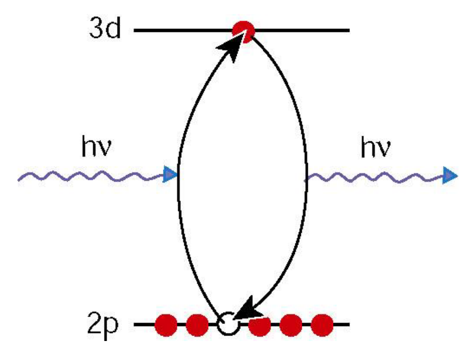


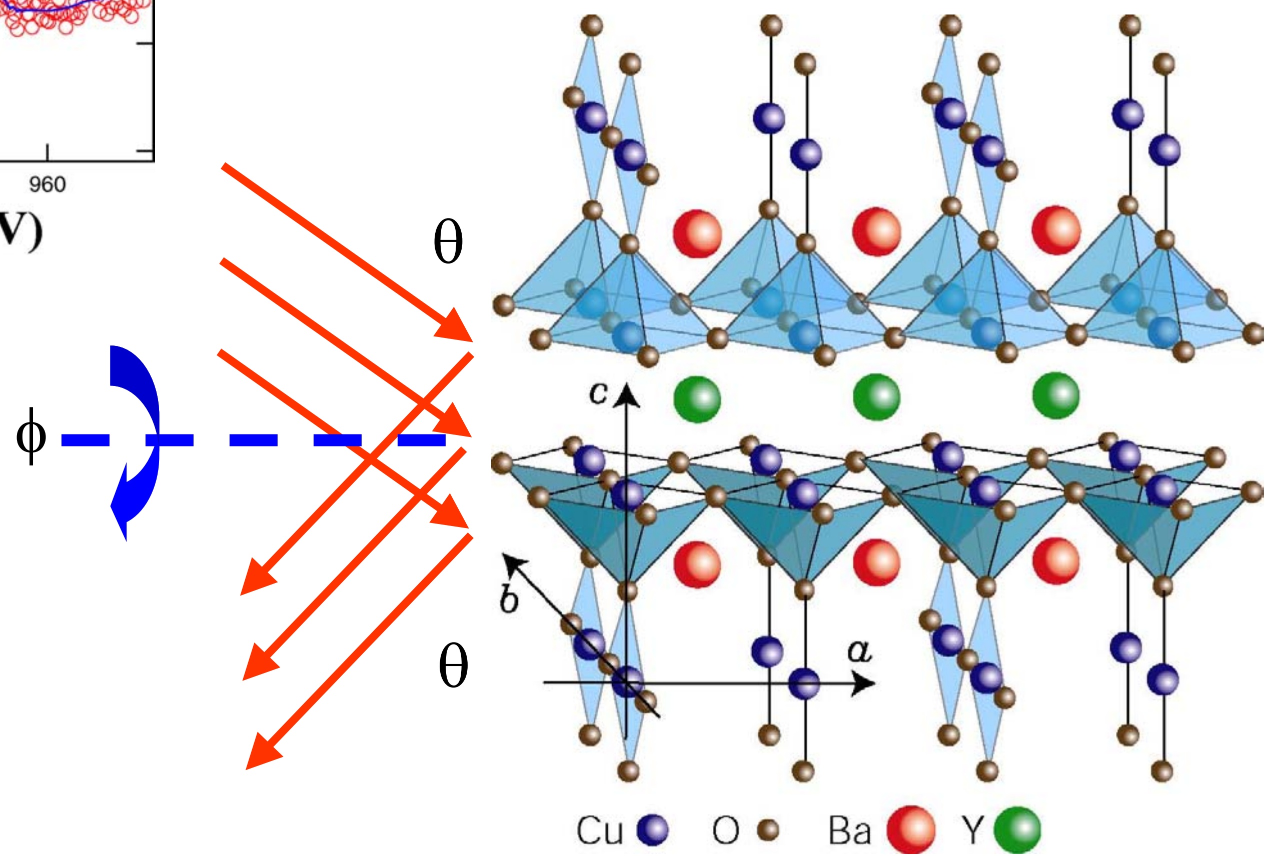
ϕ_{STO} = 3.2 eV Reihl, *PRB* **30**, 803 (1984)

ϕ_{LSCO} = 4.7 eV v. d. Marel, *Physica C* **241**,273 (1995)

Experimental Geometry

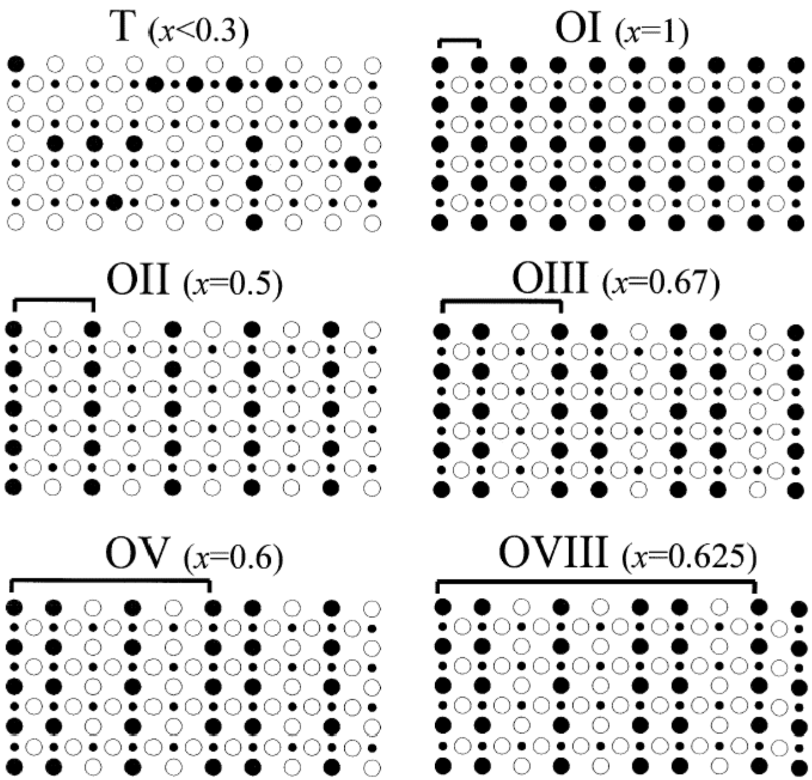




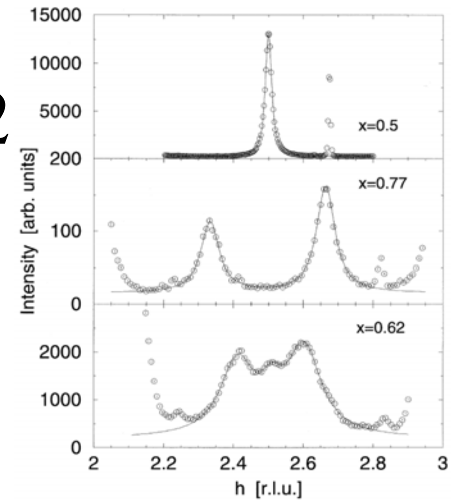


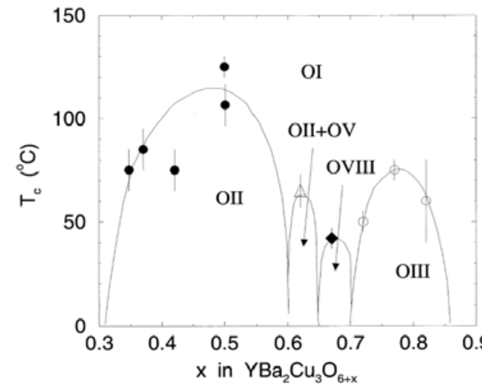
Chain (oxygen) ordering in YBa_2

Model



Hard X-ray diffraction

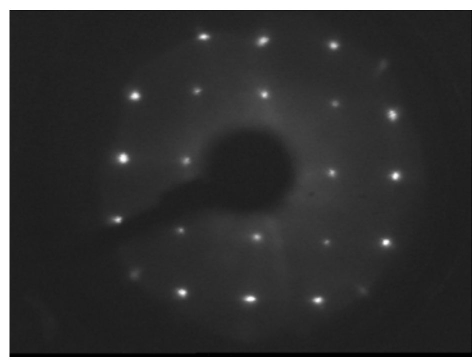




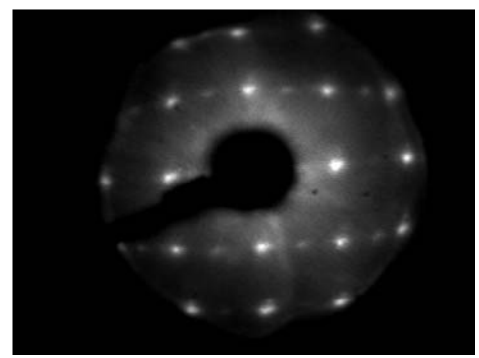
N.H. Andersen et al. / Physica C 317–318 (1999) 259–269

LEED

$YBa_2Cu_3O_{6.93}$

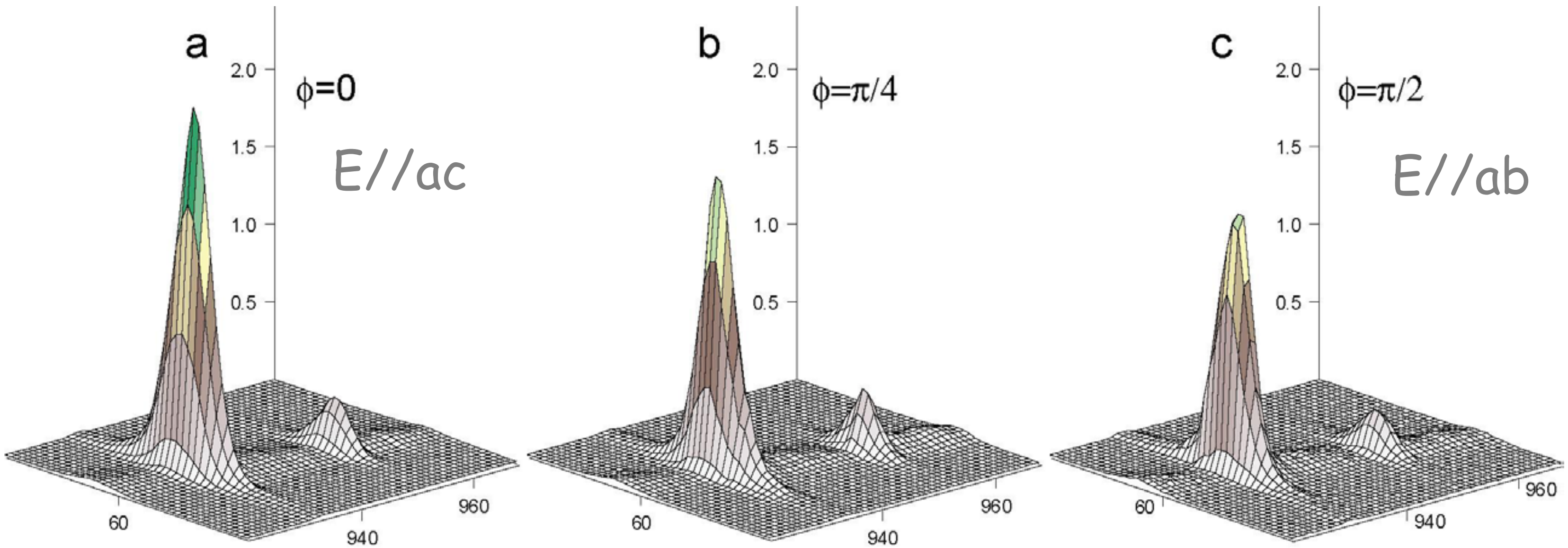


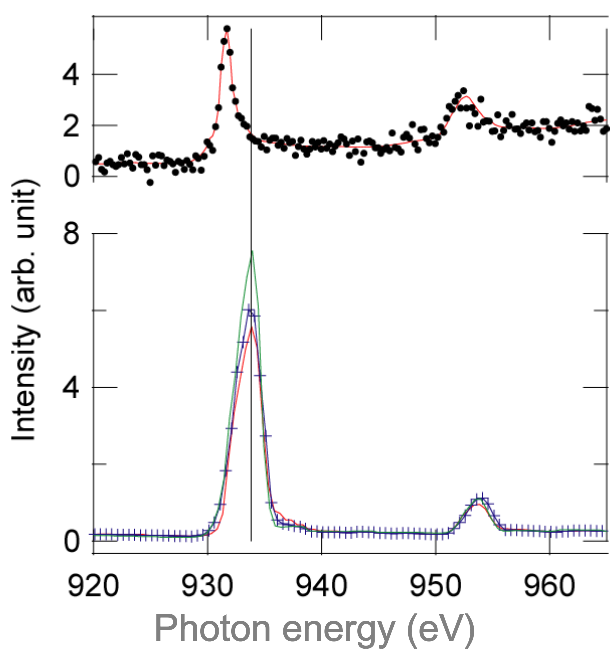
$YBa_2Cu_3O_{6.5}$



By courtesy of Donghui Lu, Stanford U.

Zooming-in on different Cu's: Tuning Polarization

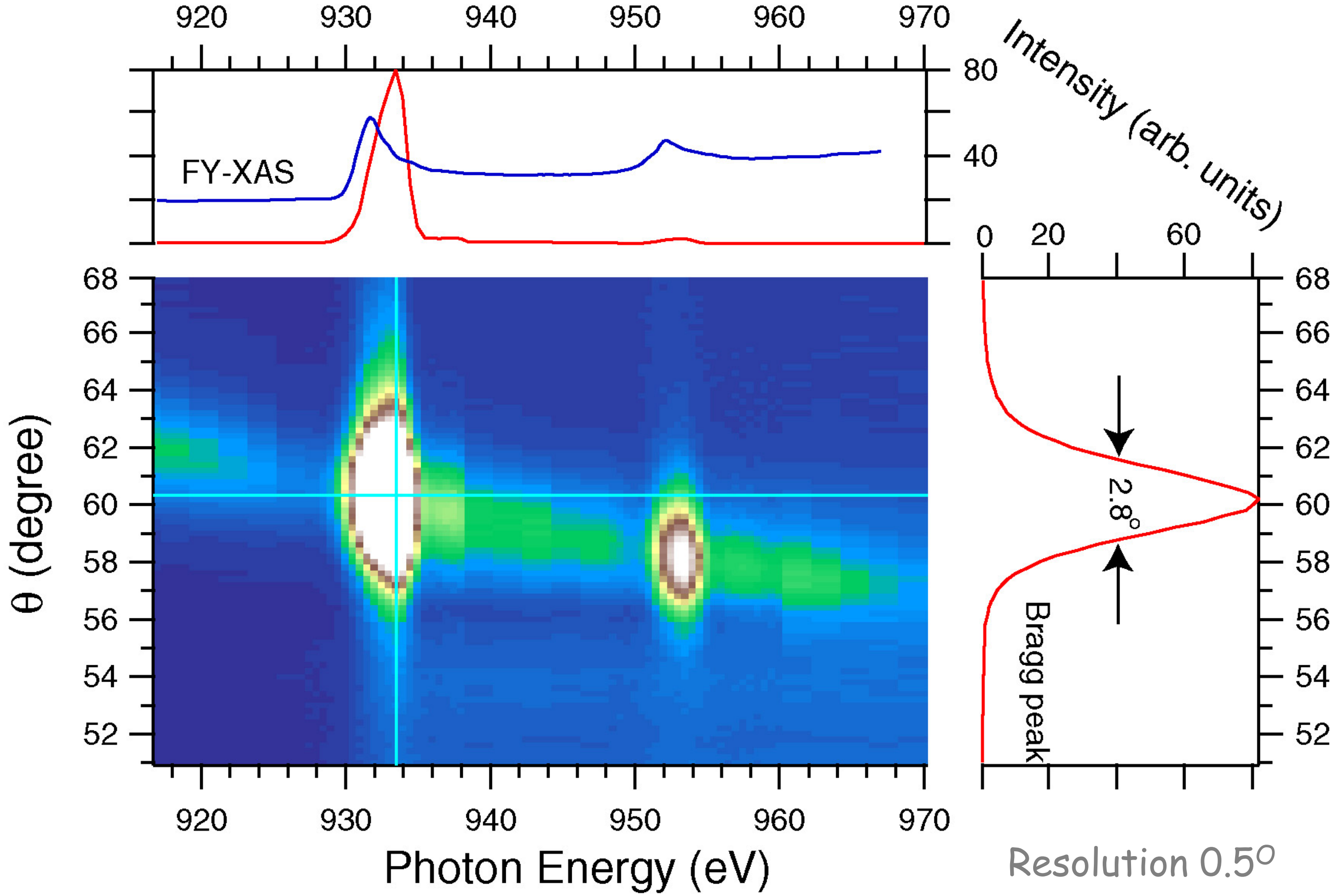




At L_3 edge, I(E//ac) / I(E//ab) = 1.3

Zooming-in on Cu: Tuning

Resonance Profile

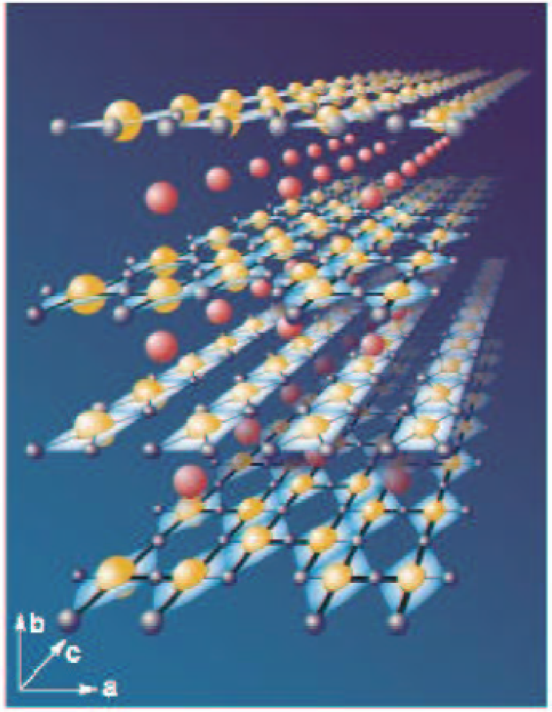


Resolution 0.5°

Bragg peak intensity is enhanced by 65 times at Cu-L_3, and 9 times at Cu-L_2

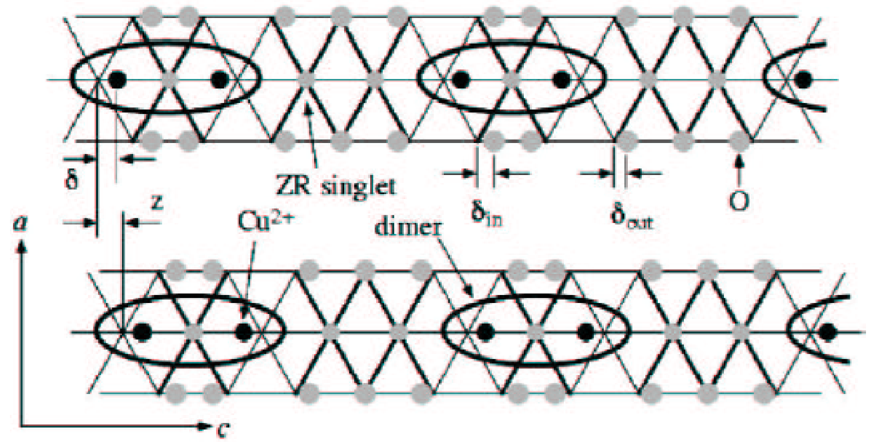
Cu contribution enhanced by much high than 65 times at Cu-L_3 !

$Sr_{14}Cu_{24}O_{41}$ – "telephone number" material

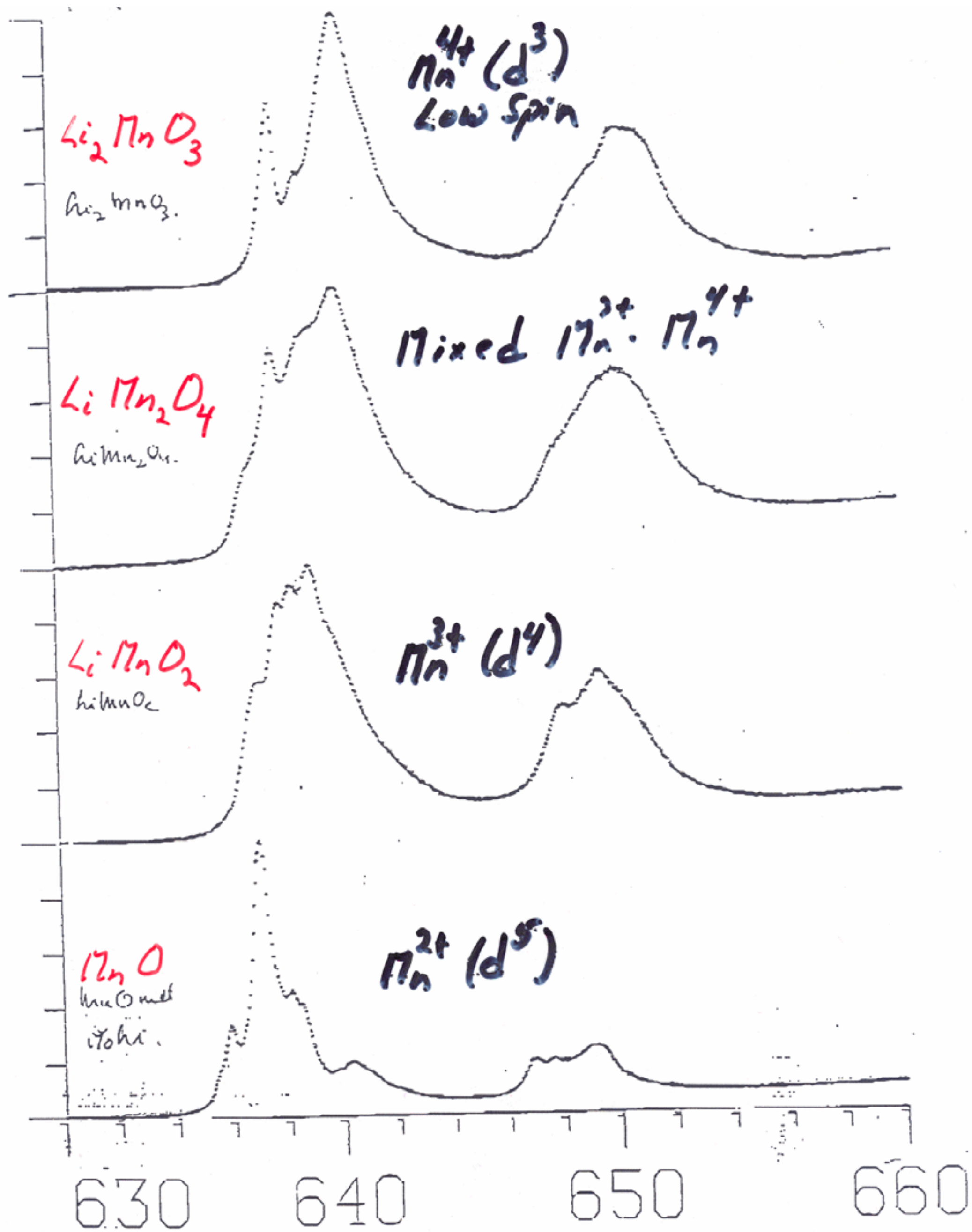


Girsh, *Science*, **297**, 584 (2002)

6 holes per unit cell 5 on chains
1 on ladder



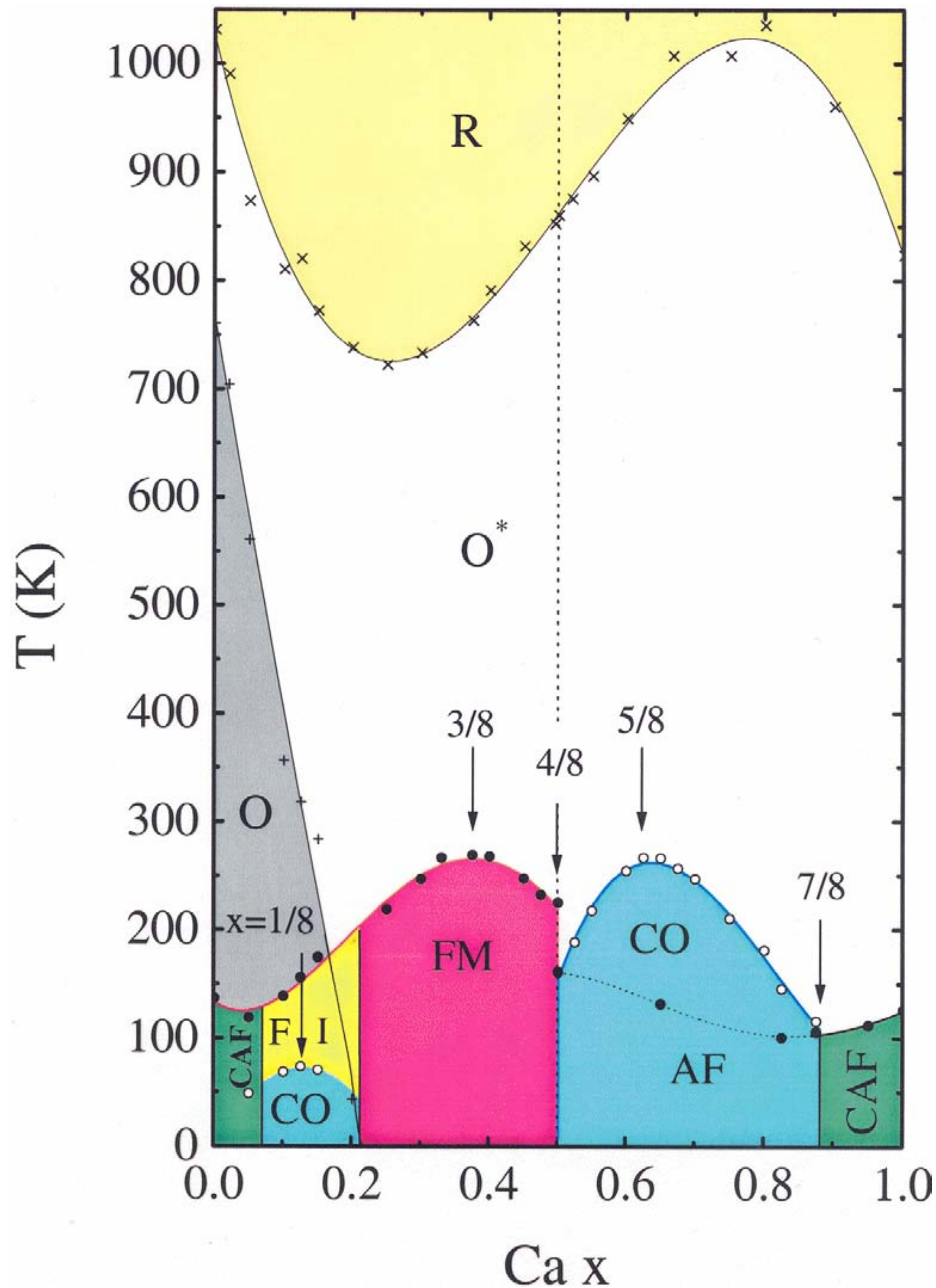
- Dimerized chain, CDW / SDW order
- Ladder $J_{\perp} = J_{||} \Rightarrow$ RVB state with spin gap and *different* CDW / SDW order.
  [Gozar, *PRL*, **87**, 197202 (2001)]



Li2MnO3
Mn4+ (d3)
Low Spin
LiMn2O4
Mixed Mn3+ : Mn4+
LiMnO2
Mn3+ (d4)
MnO
Mn2+ (d5)
630
640
650
660

Phase Diagram of
$La_{1-x}Ca_xMnO_3$

Uehara, Kim and Cheong

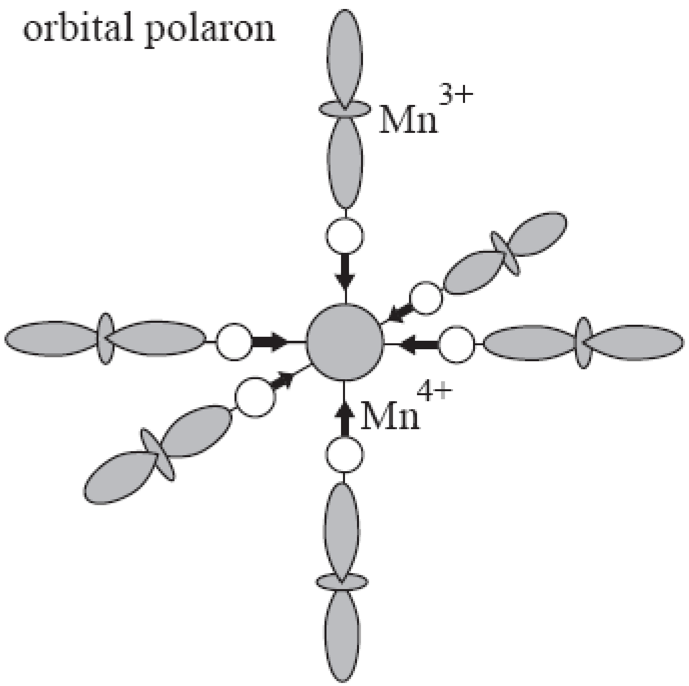


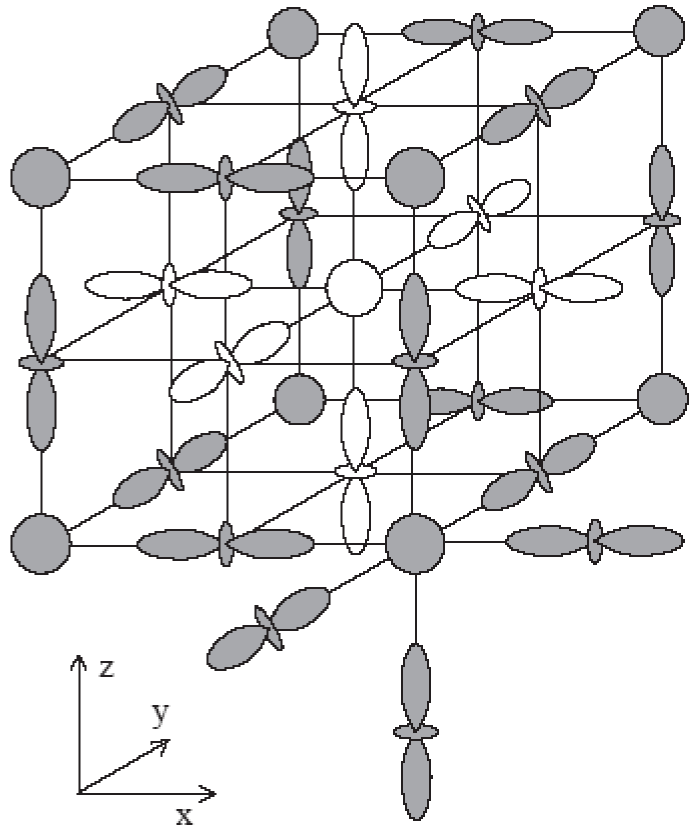
R: Rombohedral

O: Orthorhombic
(Jahn-Teller distorted)

O*: Orthorhombic
(Octahedron rotated)

Model for Charge, Spin and Orbital Correlations in Manganites



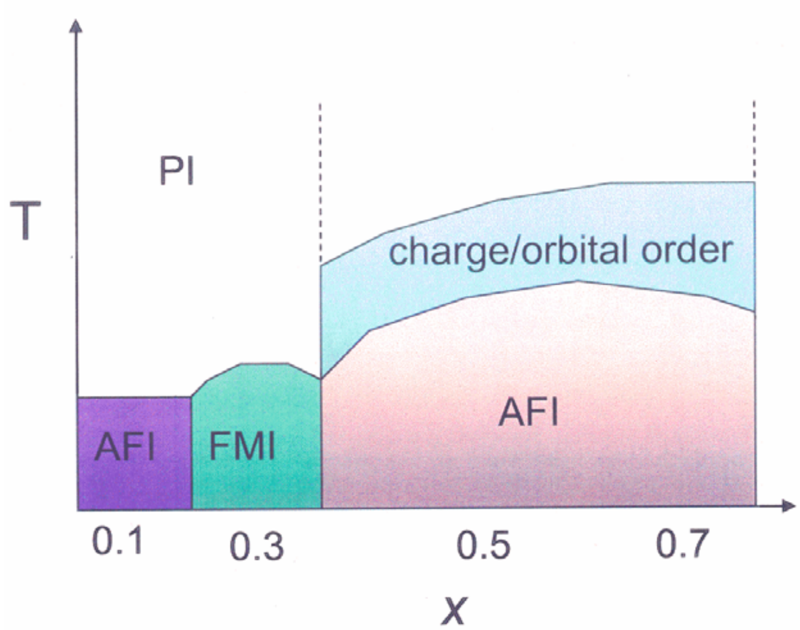


Mizokawa et al (2001)

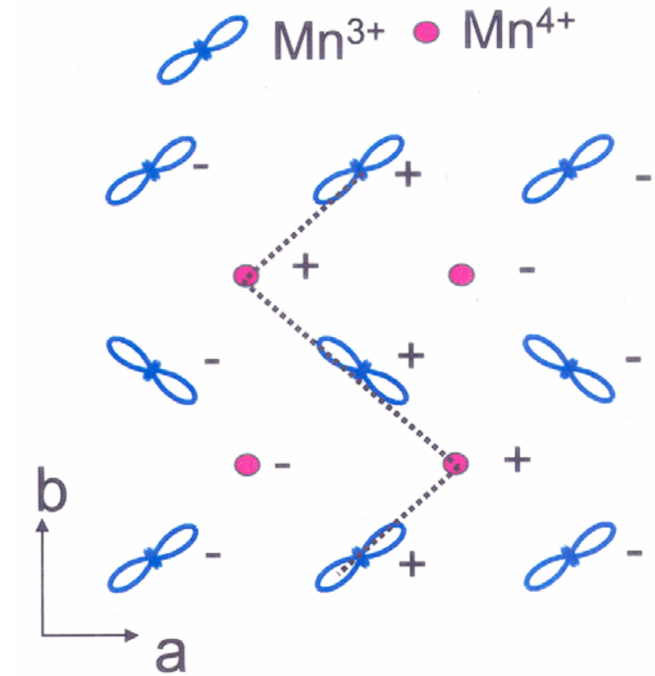
K.J. Thomas et al
NSLS/BNL

$Pr_{0.6}Ca_{0.4}MnO_3$ CE type charge, orbital and magnetic order

$Pr_{1-x}Ca_xMnO_3$



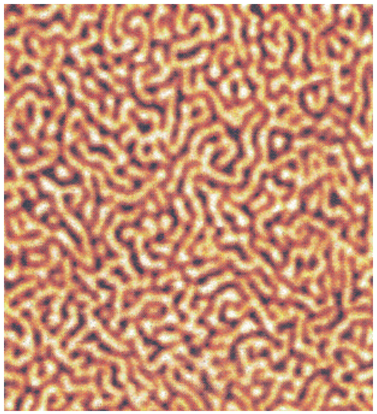
Goodenough (1955)



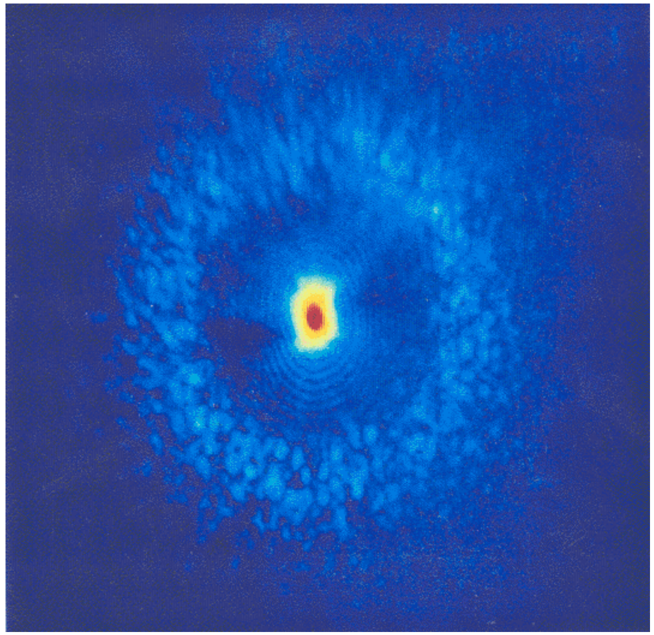
- Charge ordering below $T_{CO} \sim 240K$
- Cooperative orbital ordering + oxygen distortion at $T_{OO} = T_{CO}$
- Magnetic ordering below $T_N \sim 170K$

Coherent Soft X-ray Scattering

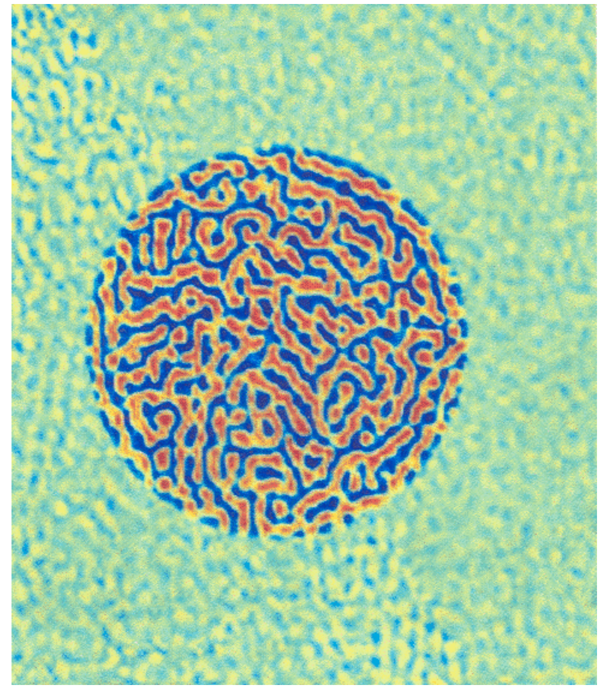
Magnetic "Worm" Domains



X-ray Speckle Pattern



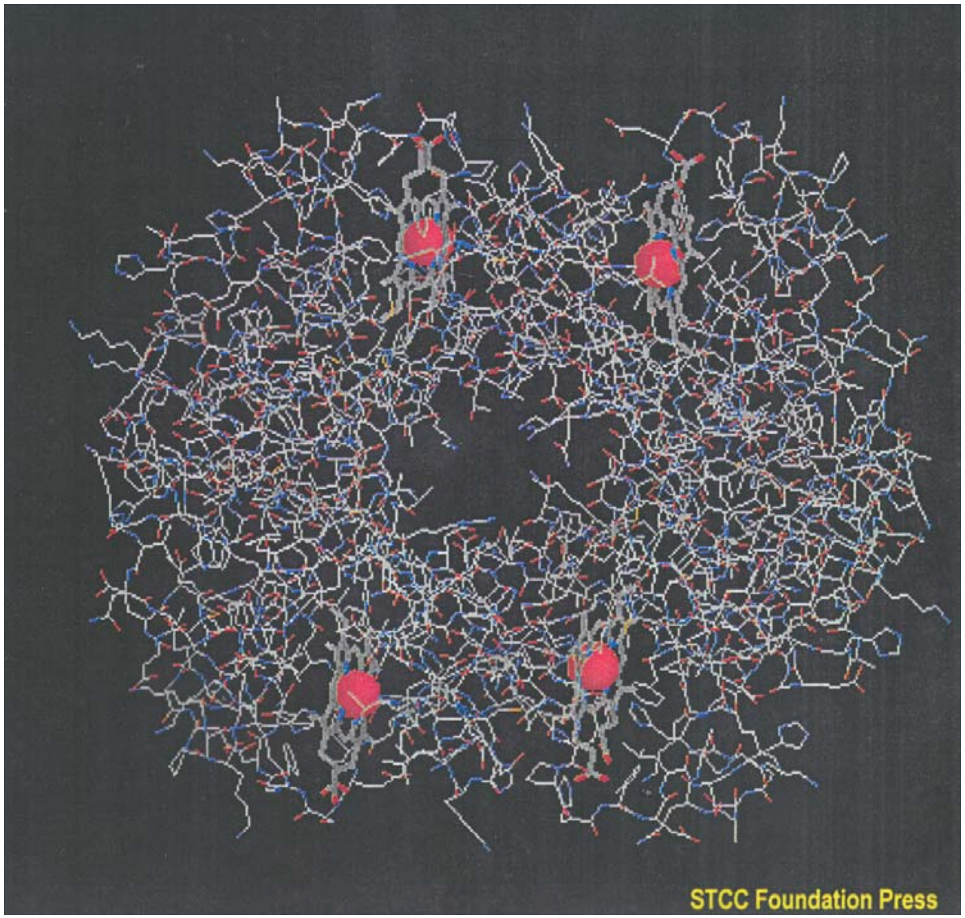
Domain Reconstruction



Example of a New Collaboration:

- oxide thin films, correlated electrons, nanomagnetism, electron spectroscopy
- UBC, SFU, FZ – Juelich, IBM Almaden, Stanford University, BESSY

Hemoglobin Studies



Thousands of atoms in hemoglobin molecules "communicate" with each other when an oxygen molecule is "dropped off"

Femto-second (10^{-15}) time slices allow observations of these "communications"

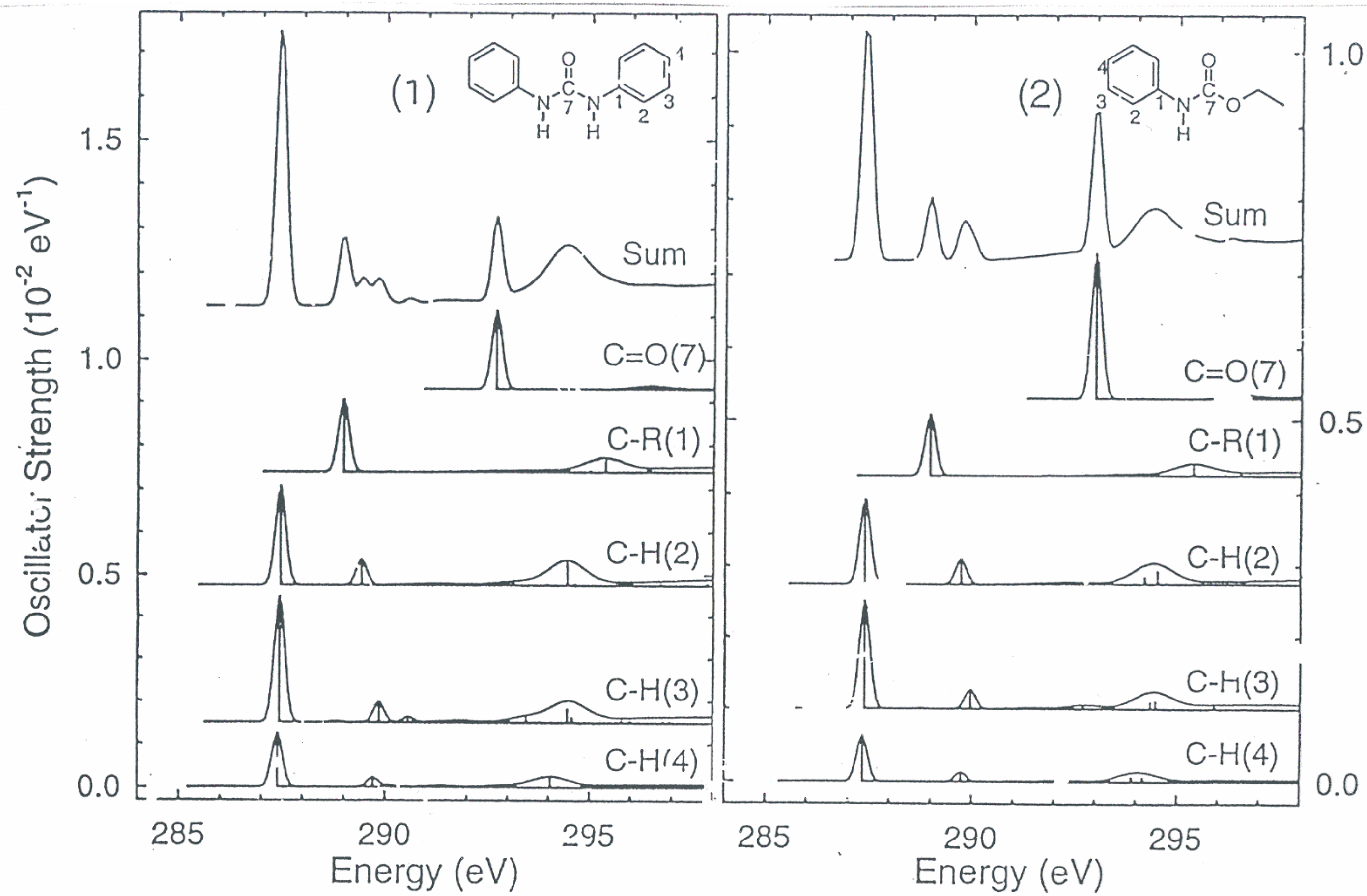


Fig. 7. Simulated C 1s spectra of *N,N'*-diphenyl urea (1) and ethyl *N*-phenyl carbamate (2) based on the results of GSCF3 calculations. The intensity of each unique carbon site is indicated, along with the appropriately weighted sum. The details of the calculations and the construction of the simulated spectra are presented in the text.

Lattice of fluxoids ($H > H_{C1} < H_{C2}$)

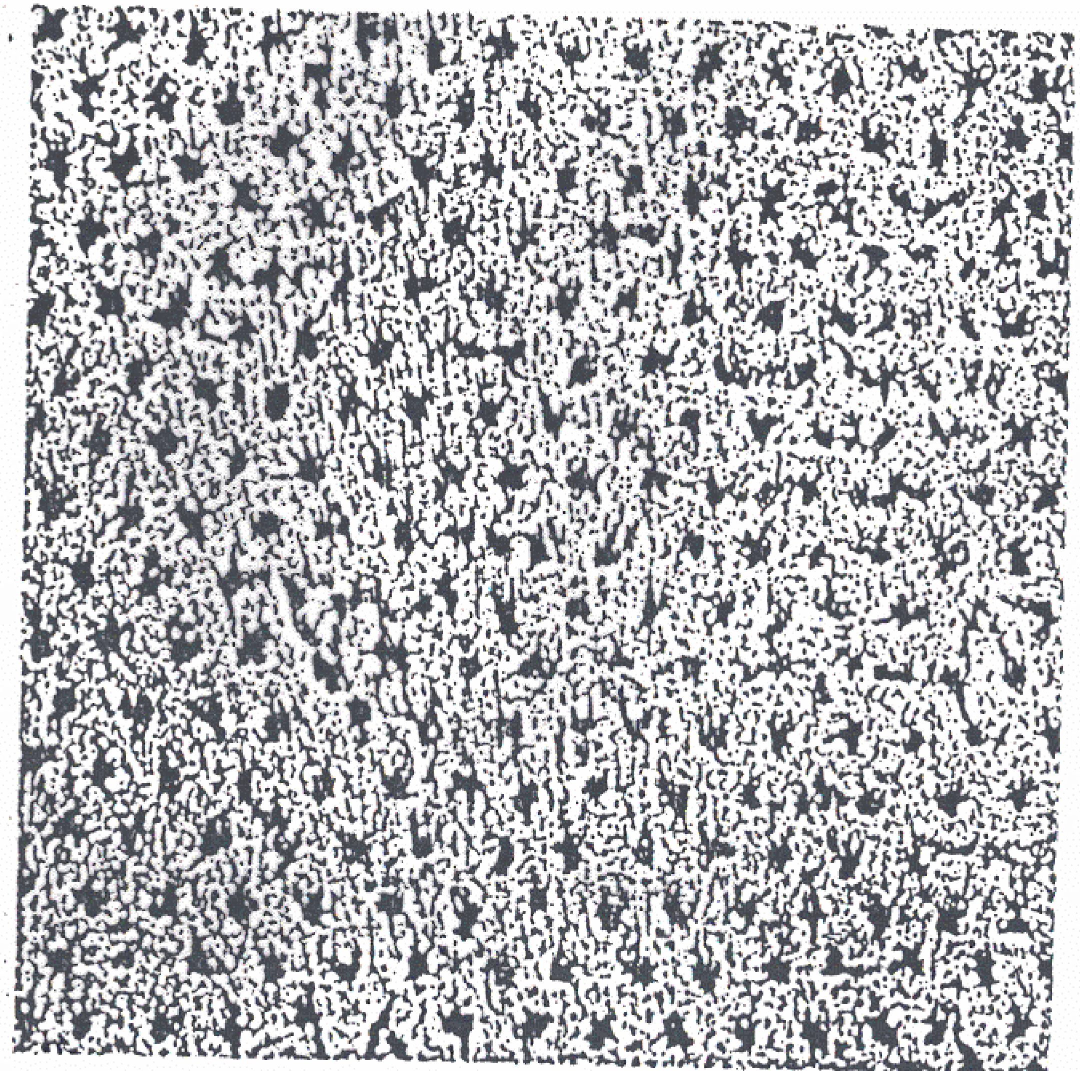


Figure 19 Triangular lattice of fluxoids through top surface of a superconducting cylinder. The points of exit of the flux lines are decorated with fine ferromagnetic particles. The electron microscope image is at a magnification of 8300, by U. Essmann and H. Träuble.

Vortex lattice of High T_C's

Type II S.C.

$$a = 1.072\sqrt{\frac{\phi_0}{B}} \qquad \phi_0 = \frac{h}{2e}$$

10 Tesla ~ 100Å

Chemical potential for electrons $T < T_C$ changes by

$$\mu(T) = \mu_0 - \frac{\Delta^2(T)}{4\mu_0} \qquad \mu_{S.C.} = \mu_{Npart} \qquad i.e$$

μ of S.C. = μ of vortex core

∴ Vorteces are charged

$$\delta n = \pi\xi^2 2N(\varepsilon_F)\delta\mu$$

For future studies we need:

- Circular polarized light
- Full polarization control [fast switching]
- Array of channelplate detectors or CCD
- Octopole superconducting magnet
- We will add two in-situ MBE systems for molecular and oxide ultra-thin film preparation
- Low Temperature

Near Future

1. Resonant X-ray Scattering
   - Orbital Ordering
   - Charge Ordering
   - Spin Ordering
   - J.-T. Distortions

   } T, x, H dep.

2. Large length scales

   (λ for 2p → 3d in 3d TM ~ 15 Å)
   - Vortex lattice in High T_C's
   - Charge and Spin density waves
   - (Dynamic) phase separation (stripes, puddles …)
   - Spatial distribution of selected organic groups
   - Structure of organic blends – self assembly

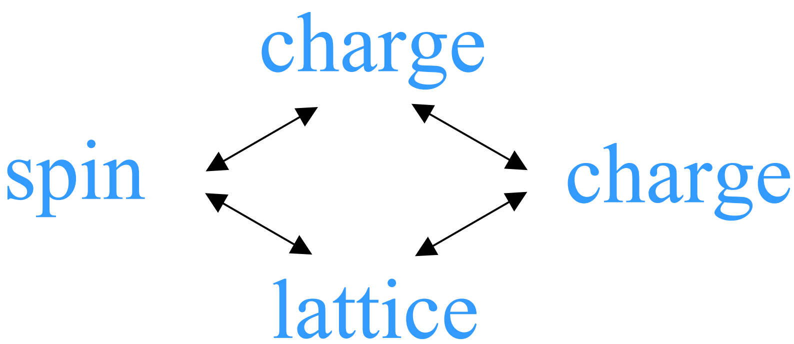
3. Use of coherence
   - Speckle – disordered systems

4. X-ray Raman
   - electronic excitations
   - Magnons
   - Phonons
   - Orbitons + Orbiton ⇄ Spin Wave
   - q dependence

5. All the above in Microscopy
   - focus to ~ 10 nM

Manganites Exhibit Interplay of Charge, Spin, Lattice and Orbital degrees of freedom

Interacting degrees of freedom
(complex electron systems)



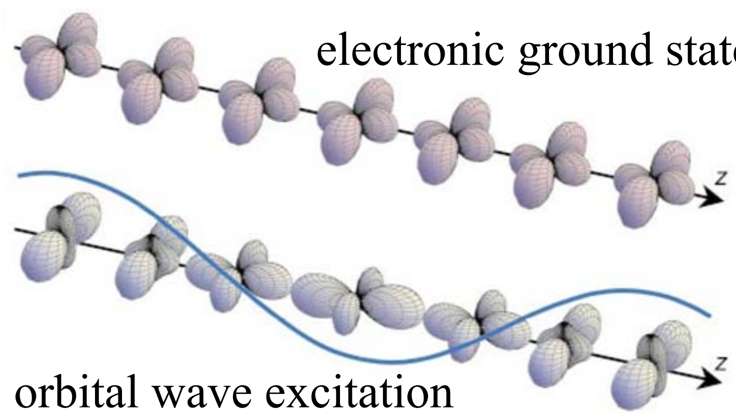
Competition among many Energy and Length scales

Determine the physics of these systems

An unexplored degree of freedom in transition metal oxides:

Orbital Density Waves

Ordering of orbitals produce long-range orbital density waves – a new type of collective excitation in crystals



E. Saitoh et al. Nature 410, 180 (2001)

$La_{0.6}Sr_{0.4}MnO_3$

Pellegrin et al.