X-ray Scattering Studies of Dynamics in Strongly Correlated Electron Systems

From Femto-seconds to Minutes

J.P. Hill
Brookhaven National Laboratory
Collaborators

**Cuprate work:**
Y-J. Kim, D. Ellis, S. Wakimoto (U. Toronto, Canada)
R.J. Birgeneau (U. C. Berkeley, USA)
T. Gog (APS, USA)
D. Casa (APS, USA)
G. Blumberg (Lucent Technologies, USA)

**Manganite work**
J.J. Turner, S. Kevan (U. Oregon, USA)
M. A. Pfeifer (U. Latrobe, Australia)
K.J. Thomas (BNL, USA)
K. Chesnel (ALS, USA)
Y. Tomioka (CERC, Japan)
Y. Tokura (CERC, Japan)
Outline

1. Dynamics in the Energy Domain
   - Resonant Inelastic X-ray Scattering
   - Recent results in the cuprates

2. Dynamics in the Time Domain
   - Coherent x-ray Scattering
   - Recent results in the manganites

3. Future
   - NSLS-II
   - Instrumentation
1. Dynamics in the Energy Domain

Inelastic X-ray Scattering

\[ w_i, k_i \rightarrow w_f, k_f \]

\[ q = k_i - k_f \]

\[ w = w_i - w_f \]

Elastic scattering \((w = 0)\) gives static properties.

Inelastic scattering \((\omega \neq 0)\) gives dynamic properties.
Electronic Excitations

1) Excitation spectrum determines the dynamic response of material.
2) Excitation spectra provide stringent test of theory.
3) “High-energy physics” of strongly correlated electron systems controls their behavior:
   \[ t \sim 1 \text{ eV}, \quad U \sim 8 \text{ eV}, \quad D \sim 2 \text{eV} \]

Need a momentum and energy resolved probe \( \rightarrow \) IXS
Resonant Inelastic X-ray Scattering

But….IXS from electronic excitations is weak (unlike the case for phonons)

\[
\frac{d^2\sigma}{d\Omega d\omega} \propto \left| \langle f \mid H_{\text{int}} \mid i \rangle + \sum_{n} \frac{\langle f \mid H_{\text{int}} \mid n \rangle \langle n \mid H_{\text{int}} \mid i \rangle}{E_i - E_n + i\Gamma} \right|^2
\]

Non-resonant scattering (weak)

Kao et al, PRB 1996, NiO

Resonant IXS is > 100 x Non-Resonant IXS
10 years of RIXS Progress: CuGeO$_3$

$\text{Figure of merit} = \text{count rate}/\text{resolution}$, increased by $10^4$!

- **1999**: 1 cps, 1500 meV, X21, NSLS
- **2002**: 6 cps, 300 meV, 9IDB, APS
- **2006**: 50 cps, 115 meV, 30IDB, APS
- **2007**: 600 cps, 90 meV, 30IDB, APS
$\Delta E = 120 \text{ meV}$

91DB, APS
La$_2$CuO$_4$

**Characteristics of 500 meV mode:**

- Strongly peaked around $(p,0)$
- Strongly doping dependent
- Present only for $e$ parallel to $c$
- Rotates polarization of photon.

2-Magnon Scattering?

Single Magnon Dispersion Surface

R. Coldea (PRL, 2001)

Calculated 2-magnon DOS

2-magnon DOS has strong peaks at (p,0) at ~500 meV

Calculated energy and momentum behavior, and doping dependence all resemble two-magnon scattering

N.B. Predicted by Tsutsui, Tohyama and Maekawa (1999)
Summary: Cuprates

Used resonant inelastic x-ray scattering to look at excitations in mid-IR region of the cuprates

• Observed a new peak at 500 meV
• Peak occurs at (p,0). Softens, and broadens, away from there. Strongly doping dependent. Rotates the photon polarization
• Consistent with it being due to 2-magnon scattering.
• Future experiments will study the temperature dependence and detailed q-dependence to make the assignment unambiguous.
2. Dynamics in the Time Domain

Coherent X-ray Scattering
Coherent Scattering: Dynamic Speckle

Speckle pattern characteristic of domain structure:

Transverse coherence length

Longitudinal coherence length

Coherent flux
An Orbital Glass?

$\mathrm{Pr}_{0.5}\mathrm{Ca}_{0.5}\mathrm{MnO}_3$

$x \sim 300 \, \text{Å}$

$\xi_{\text{orb}} \sim 375 \, \text{Å}$

$\xi_{\text{mag}} \sim 780 \, \text{Å}$

Orbital Speckle Observed

$\text{Pr}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$

Mn LIII edge
10 mm pinhole

Speckle Patterns are Largely Static

\[ T = 205 \text{ K} \]

\[ T = 232 \text{ K} \]
Some Dynamics Close to Transition

Correlations between images at fixed temperature drop off slightly near the orbital ordering transition temperature:

Summary: Manganites

Used soft x-ray coherent scattering (correlation spectroscopy) to look at orbital domains in a half-doped manganite.

• Observed “orbital speckle”
• The speckle was static at low temperatures – therefore the domains are static at low temperatures
• Even very close to the transition, (correlation length has halved to ~ 100A), the speckle was still largely static
• BUT, there is a small component close to the transition that is dynamic. Time constant is ~ 5 mins
3. National Synchrotron Light Source-II

Operational: June 2015

E=3.0 GeV
I=500 mA
e=0.6 nm-rad

Brightness will be used to study dynamics:
- Better energy resolution
- Faster time scales
Inelastic X-ray Scattering Beamline

NSLS-II: Yong Cai
Hard X-ray Coherent Scattering

NSLS-II: Andrei Fluerasu

B = 20 x APS
Time scales = 400 x faster
Conclusions

Strongly correlated electron systems exhibit dynamics over a wide range of time and space scales.

*Inelastic x-ray scattering* is a powerful probe of the very fast dynamics:
- Now ~2 meV – 10’s eV
- NSLS-II 0.1 meV – 10’s eV

*Coherent x-ray scattering* is a powerful probe of long length scale, slow dynamics
- Now ms to minutes
- NSLS-II ms to minutes