Ultrashort Lifetime Expansion for
Resonant Inelastic X-ray Scattering

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What is RIXS?
Resonant Inelastic X-ray Scattering

Synchrotron radiation

Interesting sample

Momentum transfer $q$

Energy loss $\omega$

$\omega_{\text{res}}$ (5-10 keV)

Core-hole

4p

1s
Why RIXS?

Complementary to Raman and neutrons

• Momentum dependence
• Tune X-rays to different resonances
• Small samples
• Why not?
Outline

1. Framework: the UCL expansion
   What can one measure?
2. Charge excitations
3. Magnetic RIXS on La$_2$CuO$_4$
   Coupling of core-hole to spins
   • Cu $K$-edge: indirect RIXS
   • Cu $L$-edge: direct RIXS
4. Orbital RIXS on LaMnO$_3$
Typical RIXS spectra

What happens in detail

4p empty

conduction

valence

1s core

$U_{core}$

$U_{core}$
What happens in detail

4p empty

Ultrashort Core-hole Lifetime (UCL):
Integrate out intermediate states!

1s core

U_{core}

U_{core}
Theory

- Kramers-Heisenberg relation

$$\left. \frac{d^2 \sigma}{d\Omega d\omega} \right|_{\text{res}} (q, \omega) \propto \left\langle \sum_f |A_{fi}|^2 \delta(\omega - \omega_{fi}) \right\rangle_T$$

with

$$A_{fi} = \omega_{\text{res}} \sum_n \frac{\langle f | \hat{O} | n \rangle \langle n | \hat{O} | i \rangle}{\omega_{\text{det}} - E_n - i\Gamma}$$

Detuning from resonance
Intermediate states
Inverse core-hole lifetime
$$\Gamma \approx 0.25 - 3 \text{ eV}$$
Theory - UCL expansion

Scattering amplitude

\[ A_{fi} = \omega_{\text{res}} \sum_n \frac{\langle f | \hat{O} | n \rangle \langle n | \hat{O} | i \rangle}{\omega_{\text{det}} - E_n - i\Gamma} \]

\[ \Delta := \omega_{\text{det}} - i\Gamma \]

Using the Ultrashort Core-hole Lifetime gives

\[ = \frac{\omega_{\text{res}}}{\Delta} \sum_n \sum_l \langle f | \hat{O} | n \rangle (E_n/\Delta)^l \langle n | \hat{O} | i \rangle \]

\[ = \frac{\omega_{\text{res}}}{\Delta} \sum_l \frac{1}{\Delta^l} \langle f | \hat{O}H^l_{\text{int}} \hat{O} | i \rangle \]
What does RIXS measure?

We obtain

$$A_{fi} = \frac{\omega_{\text{res}}}{\Delta} \sum_{l=0}^{\infty} \frac{1}{\Delta l} \langle f | \hat{O} H_{\text{int}}^l \hat{O} | i \rangle$$

$$H_{\text{int}} = H_0 + H'$$

Valence electrons

Interaction of valence electrons with core-hole

Assume immobile core-hole, and local core-hole potential

Arbitrary range hopping & Coulomb interactions

At $T = 0$, approximate $H_{\text{int}}^l$ for both strong and weak potentials

J. van den Brink & M. van Veenendaal, EPL 73, 121 (2006)
L. Ament, F. Forte & J. van den Brink, PRB 75, 115118 (2007)
What does RIXS measure?

Spinless fermions:

\[ A_{fi} = P(\omega) \langle f | \rho_{\mathbf{q}} | 0 \rangle \]

with \( P(\omega) = \frac{\omega_{\text{res}} U_c}{(\Delta - U_c)(\Delta - \omega)} \)

\[
\frac{d^2 \sigma}{d\omega d\Omega} \propto |P(\omega)|^2 S(\mathbf{q}, \omega)
\]

Fermions with spin:

\[ A_{fi} = P_1(\omega) \langle f | \rho_{\mathbf{q}} | i \rangle + P_2(\omega) \langle f | S_{\mathbf{q}}^2 | i \rangle \]

\( J. \) van den Brink & M. van Veenendaal, EPL 73, 121 (2006)
L. Ament, F. Forte & J. van den Brink, PRB 75, 115118 (2007)
Can we probe other excitations than charge ones?

What does RIXS measure?

Conclusion: core-hole couples to charge. RIXS measures charge density and longitudinal spin density (= higher order charge density correlation function).

Data on Cu K-edge of La$_2$CuO$_4$ below Mott gap (~ 2 eV)

- Phonons?
- d-d excitation?
- Nd$_2$CuO$_4$: no shift
- Magnons?

500 meV peak! Phonons?

J. van den Brink & M. van Veenendaal, EPL 73, 121 (2006)
L. Ament, F. Forte & J. van den Brink, PRB 75, 115118 (2007)
Magnetic RIXS on $\text{La}_2\text{CuO}_4$

Perovskite layers of $\text{CuO}_2$:

Cu 3d $e_g$ holes at low energy:

Single band Hubbard model \[ H_0 = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \]

Ground state is antiferromagnetically ordered
Collective excitations: magnons
Magnetic RIXS on La$_2$CuO$_4$

How does the core-hole couple to the spins?

Go back to Hubbard model!

Hopping amplitude $t$

Coulomb repulsion $U$

Core-hole locally modifies superexchange!

$K$-edge:

Coulomb repulsion $U - U_c$
Magnetic RIXS on La$_2$CuO$_4$

How does the core-hole couple to the spins?
Indirect RIXS locally modifies superexchange!
To first order in UCL:

$$A_{fi} = \frac{\omega_{\text{res}}}{\Delta^2} \eta \langle f \mid \sum_{k,q} J_{k+q} \mathbf{S}_k \cdot \mathbf{S}_{-k+q} \mid i \rangle$$

$$\frac{d^2 \sigma}{d\omega d\Omega} \propto |A_{fi}|^2 \Rightarrow$$

4-spin correlation function

At T = 0, 2-magnon excitations are probed:

$$\propto \eta J \langle f \mid \sum_{k} f(k) \alpha^\dagger_k \alpha^\dagger_{-k-q} |0\rangle$$

J. van den Brink, cond-mat/0510140
F. Forte, L. Ament & J. van den Brink, arXiv:0705.0263
Magnetic RIXS on La$_2$CuO$_4$

Selection rule: vanishing intensity at $q = (0,0)$ and $q = (\pi,\pi)$

F. Forte, L. Ament & J. van den Brink, arXiv:0705.0263
Magnetic RIXS on La$_2$CuO$_4$

F. Forte, L. Ament & J. van den Brink, arXiv:0705.0263

Improve theory with longer range interactions:

$J = 146.3$ meV, $J' = J'' = 2$ meV, and $J_c = 61$ meV

Values from neutron scattering.
Coldea et al., PRL 86, 5377 (2001)
Magnetic RIXS on $\text{La}_2\text{CuO}_4$

F. Forte, L. Ament & J. van den Brink, arXiv:0705.0263

J. Hill et al., arXiv:0709.3274
Magnetic RIXS on La$_2$CuO$_4$

$L$-edge: direct RIXS - photoexcited electron in 3d-shell. G. Ghiringhelli *et al.* are measuring this right now!

Sneak preview:
1. Dispersive feature similar to $K$-edge RIXS, becoming elastic in the low-$q$ limit.
2. Another feature appearing at $q = 0$ around $\omega \approx 3J$.
3. This feature disappears off-resonance.

What about the theory?
Magnetic RIXS on La$_2$CuO$_4$

$L$-edge: direct RIXS - core-electron ends up in 3d-state

Back to Hubbard model!

\[ J' = 0 \]

The extra 3d-electron **locally blocks** superexchange

The UCL results for the K-edge and L-edge are the same!
Magnetic RIXS on La$_2$CuO$_4$

$L$-edge has more intensity than $K$-edge.

Can we go beyond the UCL approximation?

Off-resonance: $A_{fi}^{(1)}$ becomes more important.

Reminder

$\Delta := \omega_{in} - i\Gamma$

Currently, $q = 0$ is being measured by G. Ghiringhelli et al.

To be continued!
LaMnO$_3$ has a cubic structure:

MnO$_6$ octahedron induces crystal field:

LaMnO$_3$: A-type AFM order:
spins align FM in each layer.
Kugel-Khomskii model without Hund’s rule coupling:

\[ H \propto \sum_{\langle i,j \rangle} \left( S_i \cdot S_j + \frac{1}{4} \right) H_{\text{orb}}^{ij} \]

To first order, orbitals of different layers decouple!

- \(e_g\) orbitals order ‘antiferro-orbitally’:
  
  \[ \begin{align*}
    &e_g: \\
    &t_{2g}:
  \end{align*} \]

  Order by superexchange and/or Jahn-Teller distortion of octahedra

- Excitations: \(e_g\) orbital waves (analogous to spin waves)
Orbital RIXS

Initial $\rightarrow$ Intermediate $\rightarrow$ Final

$e_g$ $\rightarrow$ $\rightarrow$ $\uparrow$ $\rightarrow$ $\rightarrow$ $\uparrow$ $\rightarrow$ $\rightarrow$

$H_{\text{orb}} \sim S_i^+ S_j^z$

Looks like Heisenberg, but no conservation of pseudo-$S^z$. This leads to single orbiton excitations.

J. van den Brink, P. Horsch, F. Mack & A. M. Oles, PRB 59, 6795 (1999)
Orbital RIXS

\[ U' = U + U_c \]

How does the core-hole couple to the \( e_g \) orbitals?

Again, core-hole locally modifies superexchange!
Orbital RIXS spectrum for LaMnO$_3$

J = 25 meV

One-orbiton peak

Two-orbiton continuum

Experimental limitations:
Resolution = 100 meV, inelastic peak
Conclusions

• Experimental progress is enormous

• Theoretical spectra can be easily obtained with the UCL expansion using e.g. a model Hamiltonian

• We now know which charge correlation functions we measure in indirect RIXS

• Both direct and indirect RIXS are a new probe for magnons (La$_2$CuO$_4$)

• We can possibly detect orbitons in the manganites!