Theory of Spectroscopy on Strongly Correlated Electron Systems

Jeroen van den Brink

Ament, van Veenendaal, Devereaux, Hill & JvdB
Rev. Mod. Phys. 83, 705 (2011)

7th MaNEP Winterschool
Saas Fee 09.01.2017
Lectures: outline

1. Intro to Spectroscopy, Correlations & RIXS

2. Theory for RIXS scattering amplitude

3. Magnetic RIXS: Theory and Experiment

High Tc superconducting cuprates

La$_2$CuO$_4$ crystal structure

- Introduce cuprates
- Bimagnon RIXS Cu K-edge
- Single Magnon RIXS Cu L-edge
- Magnetic RIXS on iridates
- Paramagnons in doped cuprates
**Correlation Effects: Zaanen-Sawatzky-Allen**

![Diagram of La$_2$CuO$_4$]

**Charge Transfer Insulator**

Zaanen, Sawatzky, Allen
PRL 55, 418 (1985)

**La$_2$CuO$_4$ magnetic structure**

![Diagram of magnetic structure]

strongly correlated antiferromagnet
spin $1/2$ insulator
gap $\sim 2$ eV

**Atomic Model: Local d-d orbital splitting: Cu$^{2+}$**

Cubic Crystal field splitting

Cu$^{2+}$
3d$^9$

Holes per CuO$_2$ Square

**Cu-O phase diagram**

High-T$_c$ Phase Diagram

very strange metal
messy insulator
antiferromagnet
less strange metal

Temperature

Ultra-short Core-hole Life-time expansion

- short life-time $\tau$ of the high energy core-hole
- large core-hole broadening $\Gamma = \hbar/\tau$

**RIXS amplitude**

$$F_{fg} = \langle f|D^d G(z_k) D|g\rangle$$

$$z_k = E_g + \hbar \omega_k + i\Gamma$$

large

$$G(z_k) = \frac{1}{2z_k - H} = \sum_n \frac{|n\rangle\langle n|}{z_k - E_n}$$

= constant

RIXS response governed by (dipole) transition operators

Direct **RIXS amplitude @ transition metal L-edge**

Ament, Ghiringhelli, Moretti, Braicovich & JvdB, PRL 103, 117003 (2009)


Ament, Ghiringhelli, Moretti, Braicovich & JvdB, PRL 103, 117003 (2009)

Orbital excitations by direct RIXS on $\text{La}_2\text{CuO}_4$

Moretti, Bisogni, Aruta, Balestrino, Berger, Brookes, Laca, Castro, Grioni, Guarise, Medaglia, Miletto, Minola, Perna, Radovic, Salluzzo, Schmitt, Zhou, Braicovich & Ghiringhelli, NJP 13, 043026 (2011)

**K-edge RIXS on $\text{La}_2\text{CuO}_4$**

500 meV peak! Phonons? d-d excitation? Magnons?

J. Hill et al., PRL 100, 097001 (2008)

Indirect Resonant Inelastic scattering mechanism

Initial: $1s$ core

Intermediate: Conduction, Valence, $4p$ empty

Final: $1s$ core

Indirect Resonant Inelastic scattering mechanism

Initial: $1s$ core

Intermediate: Conduction, Valence, $4p$ empty

Final: $1s$ core

$U_{\text{core}}$
Indirect Resonant Inelastic scattering mechanism

Initial state:
- 4p empty
- conduction
- valence
- 1s core

Final state:
- 1s core

Intermediate state:
- 4p empty
- conduction
- valence
- 1s core

\( U_{\text{core}} \) in the initial and final states.

Indirect Resonant Inelastic scattering mechanism

Initial state:
- 4p empty
- conduction
- valence
- 1s core

Final state:
- 1s core

Intermediate state:
- 4p empty
- conduction
- valence
- 1s core

\( (q, \omega) \) in the intermediate state.

\( \hat{O}_{\text{eff}} \) for low energy elementary excitations:

Core-hole potential is strong
but its life-time is ultra-short
**Theory: Kramers-Heisenberg**

**Fermi’s Golden Rule**

**Scattering amplitude:**
\[ A_{ij} \]

**Scattering intensity:**
\[ I_{\text{scatter}}(q,\omega) = \sum_j |A_{ij}|^2 \delta(\omega - \omega_{ji}) \]

**RIXS amplitude:**
\[ A_{ij} = \sum_n \frac{\langle f | \bar{D} | n \rangle \langle n | \bar{D} | i \rangle}{\omega_n - E_n - i\Gamma_n} \]

Core-hole life-time

H. Kramers and W. Heisenberg, Z. Phys. 31, 681 (1925)

**Theory: Ultra-short Corehole Life-time Expansion**

core-hole life time is ultra-short:
\[ \sim \text{femto sec} \]

core-hole broadening \( \Gamma \) is large:
\[ \sim 1-2 \text{ eV} \] \( K \)-edge
\[ \sim 0.5 \text{ eV} \] \( L \)-edge

suggests expansion parameter:
\[ \frac{1}{\omega_{in} - \omega_{res} - i\Gamma} = \frac{1}{\Delta} \]
with
\[ \Delta = \omega_{in} - \omega_{res} - i\Gamma \]


**Spin-photon coupling for Cu K-edge RIXS**

How does intermediate state core-hole couple to spins?
Go back to Hubbard model

Hopping amplitude \( t \)
Coulomb repulsion \( U \)

with core hole:
\[ \Delta E = U \Rightarrow J = \frac{4t^2}{U} \]

**Spin-photon coupling for Cu K-edge RIXS**

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intermediate state at \( U \cdot U_c \)
Spin-photon coupling for Cu K-edge RIXS

How does intermediate state core-hole couple to spins?
Go back to Hubbard model

Hopping amplitude $t$
Coulomb repulsion $U$

Core-hole locally modifies superexchange $J$!

intermediate state at $U - U_c$

K-edge RIXS on $\text{La}_2\text{CuO}_4$: two-magnon scattering

Intermediate state modification of $J$
leads in first order to

$$A_{fi} = \frac{\omega_{\text{res}}}{\Delta^2} \eta \langle f | \sum_{k,q} J_{k+q} S_k \cdot S_{-k+q} | i \rangle$$

2-magnon excitations are probed:

$$\propto \eta J \langle f | \sum_{k} f(k) \alpha_k \alpha_k^\dagger | 0 \rangle$$

JvdB, EPL 80, 47003 (2007)
F. Forte, L. Ament & JvdB, PRB 77, 134428 (2008)

+ magnon-magnon interactions:

T. Nagao, J. Igarashi, PRB 75, 214414 (2007)

Two-magnon scattering: experiment v. theory I

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

Two-magnon RIXS intensity

Two-magnon scattering: experiment v. theory II

F. Forte, L. Ament & JvdB, PRB 77, 134428 (2008)

Theory
Theory + experimental resolution

J. Hill et al., PRL 100, 097001 (2008)
Two-magnon scattering: experiment v. theory III

F. Forte, L. Ament & JvdB, PRB 77, 134428 (2008)

Direct Magnetic RIXS

Cu L-edge

Direct RIXS @ TM L-edges

Cu L-edge

3d

~900 eV

Energy loss

Momentum transfer

2p

3d

~900 eV

Energy loss

Momentum transfer
Direct RIXS @ TM L-edges

Cu L-edge

$\sim 900 \text{ eV}$

Momentum transfer

Energy loss

2p

3d

Direct RIXS @ TM L-edges

Cu L-edge

$\sim 900 \text{ eV}$

Momentum transfer

Energy loss

2p

$s=1/2$

$l=1$

l•s

Direct RIXS @ TM L-edges

Cu L-edge

$\sim 900 \text{ eV}$

Momentum transfer

Energy loss

2p

$s=1/2$

$l=1$

dd excitation

spin flip

Ir L-edge

$\sim 11.2 \text{ keV}$
**Atomic RIXS on Cu L-edge**

- Ground state
- Final states
- Orbital+spin flip
- Spin // z
- No spin flip without orbital flip

**RIXS spin-flip amplitude @ transition metal L-edge**

- Ament, Ghiringhelli, Moretti, Braicovich & JvdB, PRL 103, 117003 (2009)

- Ament, Ghiringhelli, Moretti, Braicovich & JvdB, PRL 103, 117003 (2009)
**RIXS spin-flip amplitude @ transition metal L-edge**

\[ x^2 - y^2 \]


Ament, Ghiringhelli, Moretti, Braicovich & JvdB,
PRL 103, 117003 (2009)

**Magnetic RIXS on Cu L-edge**

\[ \downarrow \text{hole in } x^2-y^2 \text{ orbital} \]

\[ \text{spin } // z \quad \text{spin } \perp z \]

Create Single Magnon

Ament, Ghiringhelli, Moretti, Braicovich & JvdB,
PRL 103, 117003 (2009)

**Magnetic RIXS on La}_2\text{CuO}_4 @ Cu L-edge**

In special cases direct spin-flip scattering is allowed at Cu L-edge

CuO's are such special cases...
**High resolution Cu L-edge RIXS spectrum**

- magnon
- phonon
- zero-loss
- bimagnon

**Magnetic L-edge RIXS on La$_2$CuO$_4$**

In special cases direct spin-flip scattering is allowed

**Magnetic L-edge RIXS on La$_2$CuO$_4$**

- In general: Single spin-flip amplitude related to circular magnetic dichroism in absorption
- CuO's are such cases!

**RIXS magnon dispersion of Sr$_2$CuO$_2$Cl$_2$**

- deviation from simple Heisenberg

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Braicovich, JvdB et al., PRL 104, 077002 (2010)

Haverkort, PRL 105, 167404 (2010)

Guarise et al., PRL 105, 157006 (2010)

Braicovich, JvdB et al., PRB 81, 174533 (2010)
**Magnetic RIXS vs. Inelastic Neutron Scattering**

<table>
<thead>
<tr>
<th>RIXS</th>
<th>Neutrons</th>
</tr>
</thead>
<tbody>
<tr>
<td>small</td>
<td>large</td>
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</tbody>
</table>

**What about magnons in iridates?**

**Sr$_2$IrO$_4$: equivalent of cuprate La$_2$CuO$_4$**

Ir (4+) = 5d5

$J=L+S$

$t_{2g}^5$: single hole $s=1/2$ in 3-fold degenerate $l=1$ state

**Sr$_2$IrO$_4$: equivalent of cuprate La$_2$CuO$_4$**

J$_{eff}$=3/2
doublet

J$_{eff}$=1/2
quartet

Jackeli & Khaliullin, PRL 102,017205 (2009)
B.J. Kim, Ohsumi, Komesu, Sakai, Morita, Takagi, Arima, Science 323, 1329 (2009)
Magnon dispersion

Direct RIXS on $\text{Sr}_2\text{IrO}_4$

- $3\lambda/2$
- doublet

Ament, Khaliullin & JvdB
PRB 84, 020403 (2011)

Direct RIXS on $\text{Sr}_2\text{IrO}_4$

- $3\lambda/2$
- quartet
- doublet

Jungho Kim,1 D. Casa,1 M. H. Upton,1 T. Gog,1 Young-June Kim,2 J. F. Mitchell,3 M. van Veenendaal,1,4 M. Daghofer,5 J. van den Brink,5
G. Khaliullin,6 B. J. Kim7,8,9 PRL 108, 177003 (2012)

Magnetic RIXS paramagnons

doped quasi-2D Cu-oxides

Jungho Kim,1 D. Casa,1 M. H. Upton,1 T. Gog,1 Young-June Kim,2 J. F. Mitchell,3 M. van Veenendaal,1,4 M. Daghofer,5 J. van den Brink,5
G. Khaliullin,6 B. J. Kim7,8,9 PRL 108, 177003 (2012)
Magnetic L-edge RIXS on 8% doped La$_{2-x}$Sr$_x$CuO$_4$

$T_c = 21K$

Dynamical structure factor Hubbard model, QMC

$U=8t$

Intense paramagnon excitations in a large family of high-temperature superconductors


Jia, Nowadnick, Wohlfeld, Kung, Chen, Johnston, Tohyama, Moritz & Devereaux
Nat. Comm. 5, 3314 (2014)
Summary part 3

• RIXS sensitive to magnetic excitations of e.g. low D cuprates, iron pnictides and iridates
• Magnons, spinons and paramagnons are observed
• Dispersion of these modes can be determined
• Observed paramagnons are challenge for theory
• It can reasonably be assumed that the future of RIXS is even brighter than its past
• More and better experiments, instruments, theory