Resonant Inelastic X-ray Scattering on Elementary Excitations

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Resonant Inelastic X-ray Scattering on Elementary Excitations

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

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Outline

1. Introducing RIXS

2. Magnetic RIXS on low dimensional magnets
1. Introducing RIXS

- Basic Scattering Process
- Direct & Indirect RIXS
- 5 features of RIXS
- Elementary Excitations Accessible to RIXS
- Progress in past decade
2. Magnetic RIXS on low dimensional magnets

- **Quasi 2D cuprates**
- **Quasi 1D cuprates**
- **Quasi 2D iron pnictide**
- **Quasi 2D iridate**
- **Doped Cu & Fe systems**
Basic Scattering Process

Direct and Indirect RIXS
What is X-ray scattering

Resonant Inelastic RIXS
**What is X-ray scattering**

\[ \text{Resonant, Inelastic, RIXS} \]

**X-ray scattering:** photon in \( \rightarrow \) solid \( \rightarrow \) photon out

**inelastic:** \( \omega_{\text{out}} < \omega_{\text{in}} \)

**resonant:** tune \( \omega_{\text{in}} \) to an atomic absorption edge
**What is X-ray scattering?**

**Resonant X-ray scattering (RIXS):** photon in $\rightarrow$ solid $\rightarrow$ photon out

- **Inelastic:** $\omega_{\text{out}} < \omega_{\text{in}}$
- **Resonant:** tune $\omega_{\text{in}}$ to an atomic absorption edge

**Cu K-edge:**

- $4p$
- $\sim 9 \text{ KeV}$
- $1s$
What is X-ray scattering:

- **Resonant RIXS**
  - Inelastic: $\omega_{\text{out}} < \omega_{\text{in}}$
  - Resonant: tune $\omega_{\text{in}}$ to an atomic absorption edge

X-ray scattering: \textit{photon in} $\rightarrow$ solid $\rightarrow$ \textit{photon out}

Cu K-edge

$4\ p$ $\sim 9\ KeV$

$1\ s$
What is X-ray scattering? X-ray scattering: photon in $\rightarrow$ solid $\rightarrow$ photon out

- Inelastic: $\omega_{\text{out}} < \omega_{\text{in}}$
- Resonant: tune $\omega_{\text{in}}$ to an atomic absorption edge

Cu K-edge

Energy loss

$\sim$9 KeV

Momentum transfer: $q$

1s

4p
What is X-ray scattering?

**Inelastic**

\[ \omega_{\text{out}} < \omega_{\text{in}} \]

**Resonant**

Tune \( \omega_{\text{in}} \) to an atomic absorption edge

**Cu K-edge**

Energy loss

Momentum transfer: \( q \)

\( 4p \)

\( \sim 9 \text{ KeV} \)

\( 1s \)

 INDIRECT
What is X-ray scattering?

**Resonant RIXS**

X-ray scattering: photon in → solid → photon out

- **Inelastic:** $\omega_{\text{out}} < \omega_{\text{in}}$

- **Resonant:** tune $\omega_{\text{in}}$ to an atomic absorption edge

**Cu L-edge**

**Momentum transfer:** $q$

**Energy loss**
What is X-ray scattering?

X-ray scattering: photon in $\rightarrow$ solid $\rightarrow$ photon out

Inelastic:

$\omega_{\text{out}} < \omega_{\text{in}}$

Resonant:

Tune $\omega_{\text{in}}$ to an atomic absorption edge

Energy loss

Momentum transfer: $q$

Cu L-edge

$2p$
What is X-ray scattering?

Inelastic:
\[ \omega_{\text{out}} < \omega_{\text{in}} \]

Resonant: tune \( \omega_{\text{in}} \) to an atomic absorption edge

Energy loss

Momentum transfer: \( q \)

Cu L-edge

\(~900 \text{ eV}\)

\(2\ p\)
What is X-ray scattering?

**Resonant Inelastic RIXS**

**X-ray scattering:** photon in → solid → photon out

**Inelastic:** $\omega_{\text{out}} < \omega_{\text{in}}$

**Resonant:** tune $\omega_{\text{in}}$ to an atomic absorption edge

**Cu**

**L-edge**

$3 \, d$

$\sim 900 \, \text{eV}$

$2 \, p$

**Momentum transfer:** $q$

**Energy loss**
What is X-ray scattering?

X-ray scattering: photon in → solid → photon out

Inelastic: $\omega_{\text{out}} < \omega_{\text{in}}$

Resonant: tune $\omega_{\text{in}}$ to an atomic absorption edge

Cu L-edge

$3d \sim 900 \text{ eV}$

$2p$

DIRECT

Momentum transfer: $q$

Energy loss

Resolution $< 100 \text{ meV}$
Direct and Indirect RIXS

**DIRECT**

- **INITIAL**
  - Photon in: $k, \omega_k$
  - Energy gain
  - Valence band
  - Core level

- **FINAL**
  - Photon out: $k', \omega_{k'}$

**Diagram Description:**
- The diagram illustrates the process of direct and indirect RIXS.
- Initially, a photon enters with momentum $k$ and energy $\omega_k$, exciting the electron from the core level to the valence band.
- The final state shows the electron transitioning back to the core level, with a change in momentum to $k'$ and energy $\omega_{k'}$.
**Direct and Indirect RIXS**

**DIRECT** scattering via absorption-emission matrix elements
Direct and Indirect RIXS

DIRECT scattering via absorption-emission matrix elements

INDIRECT
**Direct and Indirect RIXS**

**DIRECT**
- Scattering via absorption-emission matrix elements

**INDIRECT**
- Scattering via intermediate state core-hole shake-up
Direct RIXS @ TM L-edges

Cu L-edge

$\sim 900 \text{ eV}$

$3d$

$2p$

Momentum transfer

Energy loss
Direct RIXS @ TM L-edges

Cu L-edge

3d

~900 eV

2p

Momentum transfer

Energy loss
Direct RIXS @ TM L-edges

Cu L-edge

3d

\( \sim 900 \text{ eV} \)

2p

Momentum transfer

Energy loss
Direct RIXS @ TM L-edges

Cu L-edge

3d

~900 eV

2p

Momentum transfer

Energy loss
Direct RIXS @ TM L-edges

Cu L-edge

3d

~900 eV

2p

s=1/2

l=1

Momentum transfer

Energy loss
Direct RIXS @ TM L-edges

Cu L-edge

~900 eV

3d

2p

s=1/2

l=1

l•s

Momentum transfer

Energy loss
Direct RIXS @ TM L-edges

Cu L-edge

3d

~900 eV

2p

s=1/2
l=1

l•s

Momentum transfer
Energy loss
Direct RIXS @ TM L-edges

Cu L-edge

3d

~900 eV

2p

dd excitation

spin flip

Momentum transfer

Energy loss
**Direct RIXS @ TM L-edges**

- **Cu L-edge**: 
  - $3d$ orbital
  - $~900\text{ eV}$
  - $2p$ orbital

- **Ir L-edge**: 
  - $~11.2\text{ keV}$

- **Momentum transfer**
- **Energy loss**
- **spin flip**
- **dd excitation**
$\textit{RIXS} = |GS\rangle \rightarrow \textit{XAS} \rightarrow |\textit{INTERMEDIATE}\rangle \rightarrow \textit{XES} \rightarrow |FS\rangle$
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*Local atomic transition*
$\text{RIXS} = |GS\rangle \rightarrow \text{XAS} \rightarrow |\text{INTERMEDIATE}\rangle \rightarrow \text{XES} \rightarrow |FS\rangle$

*Complicated state with strong core-hole potential*

*Local atomic transition*
$\text{RIXS} = |GS\rangle \rightarrow \text{XAS} \rightarrow |\text{INTERMEDIATE}\rangle \rightarrow \text{XES} \rightarrow |FS\rangle$

Complicated state with strong core-hole potential

Local atomic transition

Local atomic transition
\[ RIXS = |GS\rangle \to XAS \to |\text{INTERMEDIATE}\rangle \to XES \to |FS\rangle \]

- Complicated state with strong core-hole potential
- Local atomic transition
- Local atomic transition
- Contains chemical detail and atom specific physics

But:
$\text{RIXS} = |GS\rangle \rightarrow \text{XAS} \rightarrow |\text{INTERMEDIATE}\rangle \rightarrow \text{XES} \rightarrow |FS\rangle$

Complicated state with strong core-hole potential

Local atomic transition

Contains chemical detail and atom specific physics

But: $\text{RIXS} = |GS\rangle \rightarrow \cdots \rightarrow |FS\rangle$
\[ \text{RIXS} = |GS\rangle \rightarrow \text{XAS} \rightarrow |\text{INTERMEDIATE}\rangle \rightarrow \text{XES} \rightarrow |FS\rangle \]

- **Complicated state with strong core-hole potential**
- **Local atomic transition**
- **Contains chemical detail and atom specific physics**
- **But:** \[ \text{RIXS} = |GS\rangle \rightarrow \ldots \rightarrow |FS\rangle \]
- **Carries low energy, long wavelength, elementary excitations**
\[ RIXS = |GS\rangle \rightarrow XAS \rightarrow |INTERMEDIATE\rangle \rightarrow XES \rightarrow |FS\rangle \]

- Complicated state with strong core-hole potential

Local atomic transition

- Contains chemical detail and atom specific physics

**But:**

\[ RIXS = |GS\rangle \rightarrow \ldots \rightarrow |FS\rangle \]

- Carries low energy, long wavelength, elementary excitations

**Universal effective low energy behavior**
5 distinguishing features of RIXS
Why Inelastic Scattering with X-rays at a resonance
**Why** Inelastic Scattering

**X-rays:** momentum $\sim \text{Å}^{-1}$
Why Inelastic Scattering with X-rays at a resonance

X-rays: momentum $\sim \text{Å}^{-1}$

Solid:
- Lattice spacings $\sim \text{Å}$
- Brioullin zone $\sim \text{Å}^{-1}$

[Diagram showing particle energy vs. momentum]
Why Inelastic Scattering with X-rays at a resonance

**X-rays:** momentum $\sim \text{Å}^{-1}$

**Solid:**
- Lattice spacings $\sim \text{Å}$
- Brilliouin zone $\sim \text{Å}^{-1}$

**Visible Photons:**
- Momentum $\sim 10^{-3} \text{Å}^{-1}$
Why Inelastic Scattering

with X-rays at a resonance

X-rays: momentum $\sim \text{Å}^{-1}$

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- Lattice spacings $\sim \text{Å}$
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Visible Photons:
- momentum $\sim 10^{-3} \text{Å}^{-1}$

X-rays at 10 keV:
- momentum $\sim 5 \text{Å}^{-1}$
- several BZ’s
Why Inelastic Scattering

with X-rays at a resonance

probe charged excitations

X-rays:

momentum $\sim \text{Å}^{-1}$

Solid:

Lattice spacings $\sim \text{Å}$

Brioullin zone $\sim \text{Å}^{-1}$

Visible Photons:

momentum $\sim 10^{-3} \text{ Å}^{-1}$

X-rays at 10 keV

momentum $\sim 5 \text{ Å}^{-1}$

several BZ’s
Why Inelastic Scattering with X-rays at a resonance probe charged excitations have angular momentum $l=1$

X-rays: momentum $\sim \text{Å}^{-1}$

Solid: Lattice spacings $\sim \text{Å}$
Brioullin zone $\sim \text{Å}^{-1}$

Visible Photons: momentum $\sim 10^{-3} \text{Å}^{-1}$

X-rays at 10 keV momentum $\sim 5 \text{Å}^{-1}$
several BZ’s
**Why**

Inelastic Scattering

with X-rays at a resonance

probe charged excitations

have angular momentum \( l=1 \)

polarization dependence

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**X-rays:**

momentum \( \sim \) \( \text{Å}^{-1} \)

---

**Solid:**

Lattice spacings \( \sim \) \( \text{Å} \)

Brioullin zone \( \sim \) \( \text{Å}^{-1} \)

Visible Photons:

momentum \( \sim 10^{-3} \) \( \text{Å}^{-1} \)

---

**X-rays at 10 keV**

momentum \( \sim 5 \) \( \text{Å}^{-1} \)

several BZ’s
1. RIXS exploits both the *energy* and *momentum* dependence of the photon scattering cross-section. Comparing the energies of a neutron, electron, and photon, each with a wavelength on the order of the relevant length scale in a solid, *i.e.* the interatomic lattice spacing, which is on the order of a few Angstroms, it is obvious that an x-ray photon has much more energy than an equivalent neutron or electron.
1. RIXS exploits both the *energy and momentum* dependence of the photon scattering cross-section. Comparing the energies of a neutron, electron, and photon, each with a wavelength on the order of the relevant length scale in a solid, *i.e.* the interatomic lattice spacing, which is on the order of a few Angstroms, it is obvious that an x-ray photon has much more energy than an equivalent neutron or electron. The scattering phase space (the range of energies and momenta that can be transferred in a scattering event) available to x-rays is therefore correspondingly larger and is in fact without equal. For instance, unlike photon scattering experiments with visible or infrared light, RIXS can probe the full dispersion of low energy excitations in solids.
2. RIXS can utilize the \textit{polarization} of the photon: the nature of the excitations created in the material can be disentangled through polarization analysis of the incident and scattered photons, which allows one, through the use of various selection rules, to characterize the symmetry and nature of the excitations. To date, no experimental facility allows the polarization of the scattered photon to be measured, though the incident photon polarization is frequently varied.
2. RIXS can utilize the *polarization* of the photon: the nature of the excitations created in the material can be disentangled through polarization analysis of the incident and scattered photons, which allows one, through the use of various selection rules, to characterize the symmetry and nature of the excitations. To date, no experimental facility allows the polarization of the scattered photon to be measured, though the incident photon polarization is frequently varied. It is important to note that a polarization change of a photon is necessarily related to an angular momentum change. Conservation of angular momentum means that any angular momentum lost by the scattered photons has been transferred to elementary excitations in the solid.
Why Inelastic Scattering with X-rays at a resonance
Why Inelastic Scattering

At resonance:

enhanced loss features

with X-rays at a resonance
Why Inelastic Scattering with X-rays at a resonance

At resonance:
- enhanced loss features
- choose element & electronic shell

X-ray Absorption Edges
Why Inelastic Scattering

At resonance:

- enhanced loss features
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X-ray Absorption Edges

with X-rays at a resonance

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Atomic #
Why Inelastic Scattering

At resonance:
- enhanced loss features
- choose element & electronic shell

with X-rays at a resonance

X-ray Absorption Edges

K-edges

Atomic #
**Why** Inelastic Scattering

At resonance:
- enhanced loss features
- choose element & electronic shell

with X-rays at a resonance

**X-ray Absorption Edges**

![Graph showing X-ray absorption edges with K-edges, L-edges, and TM regions.](image)

Atomic #
Why Inelastic Scattering

At resonance:
- enhanced loss features
- choose element & electronic shell

with X-rays at a resonance

X-ray Absorption Edges

X-ray penetration depth: ~microns
Why Inelastic Scattering

At resonance:

- enhanced loss features
- choose element & electronic shell

with X-rays

at a resonance

X-ray Absorption Edges

K-edges

L-edges

TM

X-ray penetration depth: ~microns

RIXS is bulk sensitive

Atomic #
Tunable X-ray sources

synchrotron

ESRF Grenoble
Tunable X-ray sources

synchrotron
Tunable X-ray sources

synchrotron

X-ray laser

LCLS, Stanford
3. RIXS is *element and orbital specific*: Chemical sensitivity arises by tuning the incident photon energy to specific atomic transitions of the different types of atoms in a material. Such transitions are called absorption edges. RIXS can even differentiate between the same chemical element at sites with inequivalent chemical bondings, with different valencies or at inequivalent crystallographic positions if the absorption edges in these cases are distinguishable.
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4. RIXS is *bulk sensitive*: the penetration depth of resonant x-ray photons is material and scattering geometry-specific, but typically is on the order of a few \( \mu \text{m} \) in the hard x-ray regime (for example at transition metal \( K \)-edges) and on the order of 0.1 \( \mu \text{m} \) in the soft x-ray regime (e.g transition metal \( L \)-edges).
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5. RIXS needs only *small sample volumes*: the photon-matter interaction is relatively strong, compared to for instance the neutron-matter interaction strength. In addition, photon sources deliver many orders of magnitude more particles per second, in a much smaller spot, than do neutron sources. These facts make RIXS possible on very small volume samples, thin films, surfaces and nano-objects, in addition to bulk single crystal or powder samples.
Elementary Excitations
accessible to RIXS
Elementary Excitations in TMO: Schematic

- Phonons
- Magnons
- Charge Transfer

Energy scale:
- 50 meV
- 500 meV
- 1.5 eV
- 2 eV
Elementary Excitations in TMO: Schematic

Couple to Charge

Phonons

Magnons

Charge Transfer

Energy

50 meV

500 meV

1.5 eV

2 eV
Elementary Excitations in TMO: Schematic

Couple to Charge

dipolar
dipolar
quadru polar
Elementary Excitations in TMO: Schematic

Couple to Charge

dipolar

quadru polar

Couple to Angular Momentum
Elementary Excitations in TMO: Schematic

**Couple to Charge**

- dipolar
- quadrupolar
- d-d

**Couple to Angular Momentum**

- Phonons
- Magnons

Energy scale:
- 50 meV
- 500 meV
- 1.5 eV
- 2 eV

Charge Transfer
In principle RIXS can probe a very broad class of intrinsic excitations of the system under study – as long as these excitations are overall charge neutral. This constraint arises from the fact that in RIXS the scattered photons do not add or remove charge from the system under study.
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Progress in the Past Decades
Progress @ Cu K-edge resolution

Zahid Hasan et al., (2000)
Progress @ Cu K-edge resolution

Zahid Hasan et al., (2000)

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

Cu K-edge on La$_2$CuO$_4$
Progress @ Cu K-edge resolution II

Cu K-edge

J. Hill et al., (2008)
Progress @ Cu K-edge resolution II

Cu K-edge

Yavas, et al., JPCM 22, 485601 (2010)

J. Hill et al., (2008)
Progress @ Cu K-edge resolution III

Hard x-ray regime: Cu K-edge

resolution (meV) count rate (1/sec) count rate/resolution (1/sec-eV)


1500 1000 500 100 0

10^3 10^2 10^1 10^0

10^4 10^3 10^2 10^1 10^0 10^{-1}

Progress in soft x-ray RIXS resolution at the Cu L-edge at 931 eV (a) (Ichikawa et al., 1996), BLBB @ Photon Factory (b) I511-3 @ MAX II (Duda et al., 2000b); (c) AXES @ ID08, ESRF (Ghiringhelli et al., 2004) (d) AXES @ ID08, ESRF (Braicovich et al., 2009), (e) SAXES @ SLS (Ghiringhelli et al., 2010). Courtesy of G. Ghiringhelli and L. Braicovich.
Progress in X-ray sources...
Progress in X-ray sources...

synchrotrons
Progress in X-ray sources...

- X-ray lasers
- Synchrotrons
Summary part I
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• Direct and Indirect RIXS
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• RIXS measures excitation energy & momentum
Summary part I

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Summary part I

• Direct and Indirect RIXS
• RIXS measures excitation energy & momentum
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• Measures charge neutral elementary excitations
  spin, orbital, lattice, charge excitons
Summary part I

- Direct and Indirect RIXS
- RIXS measures excitation energy & momentum
- Polarization in/out dependence can be studied
- Element and orbital sensitive
- Bulk sensitive & needs small sample volumes
- Measures charge neutral elementary excitations: spin, orbital, lattice, charge excitons
- Great progress in resolution in the past decade
2. Magnetic RIXS on low dimensional magnets

- Quasi 2D cuprates
- Quasi 1D cuprates
- Quasi 2D iron pnictide
- Quasi 2D iridate
- Doped Cu & Fe systems
Quasi 2D cuprates
La$_2$CuO$_4$ crystal structure

- CuO$_2$ layer
- CuO$_2$ layer
- CuO$_2$ layer

- (LaO)$_2$ layer
- (LaO)$_2$ layer
- (LaO)$_2$ layer
La$_2$CuO$_4$ magnetic structure

strongly correlated antiferromagnet

spin 1/2
La$_2$CuO$_4$ magnetic structure

- strongly correlated antiferromagnet
- spin 1/2 insulator
La$_2$CuO$_4$ magnetic structure

- strongly correlated antiferromagnet
- spin 1/2 insulator

$t U \sim 6 \text{ eV}$
La$_2$CuO$_4$ magnetic structure

- Strongly correlated antiferromagnet
- Spin 1/2 insulator
- Gap $\sim$ 2 eV
- $U \sim$ 6 eV
Atomic Model: Local d-d orbital splitting: Cu$^{2+}$

Cubic Crystal field splitting

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

5x

$e_g$

$\uparrow\uparrow\uparrow\uparrow\uparrow$

$t_{2g}$

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

Cubic Crystal field splitting

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

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5x

- - - - - - - - - - - - - - - - -

- - - - - - - - - - - - - - - - -

- - - - - - - - - - - - - - - - -

- - - - - - - - - - - - - - - - -

$\text{eg}$ orbitals

$3z^2-r^2$

$x^2-y^2$

$t_{2g}$ orbitals

$xy$

$yz$

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

Cubic Crystal field splitting

Cu^{2+} 3d^9

- \( e_g \) orbitals
- \( t_{2g} \) orbitals

5x

\( 3z^2 - r^2 \)
\( x^2 - y^2 \)
\( xy \)
\( yz \)
\( zx \)
Atomic Model: Local d-d orbital splitting: Cu$^{2+}$

Cubic Crystal field splitting

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

- $e_g$ orbitals
  - $3z^2 - r^2$
  - $x^2 - y^2$

- $t_{2g}$ orbitals
  - $xy$
  - $yz$
  - $zx$
RIXS amplitude @ transition metal L-edge

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

RIXS amplitude @ transition metal L-edge

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

RIXS amplitude @ transition metal L-edge

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

Marra, Wohlfeld & JvdB,
PRL 109, 117401 (2012)
RIXS amplitude @ transition metal L-edge

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

RIXS amplitude @ transition metal L-edge

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

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

Moretti, Bisogni, Aruta, Balestrino, Berger, Brookes, Luca, Castro, Grioni, Guarise, Medaglia, Miletto, Minola, Perna, Radovic, Salluzzo, Schmitt, Zhou, Braicovich & Ghiringhelli, NJP 13, 043026 (2011)
Orbital excitations by direct RIXS on $\text{La}_2\text{CuO}_4$

Moretti, Bisogni, Aruta, Balestrino, Berger, Brookes, Luca, Castro, Grioni, Guarise, Medaglia, Miletto, Minola, Perna, Radovic, Salluzzo, Schmitt, Zhou, Braicovich & Ghiringhelli, NJP 13, 043026 (2011)
RIXS spin-flip amplitude @ transition metal L-edge

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RIXS spin-flip amplitude @ transition metal L-edge

$x^2 - y^2$ spin NOT || $z$: pure spin flip

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

$x^2 - y^2$ spin NOT // $z$:

RIXS amplitude/intensity

Interaction light & matter
RIXS amplitude $F$ and intensity $I$

$$I(\omega, k, k', \epsilon, \epsilon') = \sum_{f} |F_{fg}(k, k', \epsilon, \epsilon', \omega_k)|^2$$

$$\times \delta(E_f + \hbar \omega_{k'} - E_g - \hbar \omega_k)$$
RIXS amplitude $F$ and intensity $I$

\[ I(\omega, k, k', \epsilon, \epsilon') = \sum_f |F_{fg}(k, k', \epsilon, \epsilon', \omega_k)|^2 \]
\[ \times \delta(E_f + \hbar \omega_{k'} - E_g - \hbar \omega_k) \]
$I(\omega, k, k', \epsilon, \epsilon') = \sum_f |F_{fg}(k, k', \epsilon, \epsilon', \omega_k)|^2 \times \delta(E_f + \hbar \omega_{k'} - E_g - \hbar \omega_k)$
**RIXS amplitude $F$ and intensity $I$**

\[
I(\omega, k, k', \epsilon, \epsilon') = \sum_f |\mathcal{F}_{fg}(k, k', \epsilon, \epsilon', \omega_k)|^2 \times \delta(E_f + \hbar \omega_{k'} - E_g - \hbar \omega_k)
\]

- **Intensity**
- **Amplitude**

- $\omega_{\text{loss}}$
- $q_{\text{loss}} = k' - k$
- **momentum in**
- **out**
RIXS amplitude \( F \) and intensity \( I \)

Intensity \( I(\omega, k, k', \epsilon, \epsilon') = \sum_{f} |F_{fg}(k, k', \epsilon, \epsilon', \omega_k)|^2 \times \delta(E_f + \hbar \omega_k' - E_g - \hbar \omega_k) \)

polarization in

momentum in

\( \omega \text{loss} \)

out

out

out
\[ I(\omega, k, k', \epsilon, \epsilon') = \sum_f |\mathcal{F}_{fg}(k, k', \epsilon, \epsilon', \omega_k)|^2 \times \delta(E_f + \hbar \omega_k' - E_g - \hbar \omega_k) \]
\[ I(\omega, k, k', \epsilon, \epsilon') = \sum_f |F_{fg}(k, k', \epsilon, \epsilon', \omega_k)|^2 \times \delta(E_f + \hbar \omega_{k'} - E_g - \hbar \omega_k) \]

- **Polarization in**
- **Intensity**
- **Amplitude**
- **Energy conservation**
- **Momentum in**
- **\( \omega_{\text{loss}} \)**
- **\( \omega_{\text{out}} \)**
- **\( \omega_{\text{in}} \)**
$I(\omega, k, k', \epsilon, \epsilon') = \sum_f |F_{fg}(k, k', \epsilon, \epsilon', \omega_k)|^2 \times \delta(E_f + \hbar \omega_k' - E_g - \hbar \omega_k)$

- **Intensity**
- **Amplitude**
- **Energy conservation**
- **Final energy**
- **Initial energy**
- **Polarization in**
- **Momentum in**
- **Out**
- **$\omega_{loss}$**

$\omega_{in}$ and $\omega_{out}$
Interaction of light and matter: Hamiltonian

\[ H = \sum_{i=1}^{N} \left[ \frac{(p_i + eA(r_i))^2}{2m} + \frac{e\hbar}{2m} \sigma_i \cdot B(r_i) + \frac{e\hbar}{2(2mc)^2} \times \sigma_i \cdot \left( E(r_i) \times (p_i + eA(r_i)) - (p_i + eA(r_i)) \times E(r_i) \right) \right] \\
+ \frac{e\hbar^2 \rho(r_i)}{8(mc)^2 \varepsilon_0} + H_{\text{Coulomb}} + \sum_{\kappa, \varepsilon} \hbar \omega_\kappa \left( a_{\kappa \varepsilon}^\dagger a_{\kappa \varepsilon} + \frac{1}{2} \right) \]
Interaction of light and matter: Hamiltonian

\[ H = \sum_{i=1}^{N} \left[ \frac{(p_i + eA(r_i))^2}{2m} + \frac{e\hbar}{2m} \sigma_i \cdot B(r_i) + \frac{e\hbar}{2(2mc)^2} \times \right. \\
\left. \sigma_i \cdot \left( E(r_i) \times (p_i + eA(r_i)) - (p_i + eA(r_i)) \times E(r_i) \right) \right] \\
+ \frac{e\hbar^2 \rho(r_i)}{8(mc)^2 \epsilon_0} + H_{\text{Coulomb}} + \sum_{\kappa, \epsilon} \hbar \omega_{\kappa} \left( a_{\kappa \epsilon}^\dagger a_{\kappa \epsilon} + \frac{1}{2} \right) \]
Interaction of light and matter: Hamiltonian

\[ H = \sum_{i=1}^{N} \left[ \frac{(p_i + eA(r_i))^2}{2m} + \frac{e\hbar}{2m} \sigma_i \cdot B(r_i) + \frac{e\hbar^2}{2(2mc)^2} \times \sigma_i \cdot \left( E(r_i) \times (p_i + eA(r_i)) - (p_i + eA(r_i)) \times E(r_i) \right) \right] + \frac{e\hbar^2 \rho(r_i)}{8(mc)^2 \epsilon_0} + H_{\text{Coulomb}} + \sum_{\kappa, \epsilon} \hbar \omega_{\kappa} \left( a_{\kappa \epsilon}^\dagger a_{\kappa \epsilon} + \frac{1}{2} \right) \]
Interaction of light and matter: Hamiltonian

\[ H = \sum_{i=1}^{N} \left[ \frac{(p_i + eA(r_i))}{2m} \right]^2 + \frac{e\hbar}{2m} \sigma_i \cdot B(r_i) + \frac{e\hbar}{2(2mc)^2} \times \right. \\
\left. \sigma_i \cdot \left( E(r_i) \times (p_i + eA(r_i)) - (p_i + eA(r_i)) \times E(r_i) \right) \right] \\
+ \frac{\hbar^2 \rho(r_i)}{8(mc)^2 \varepsilon_0} + H_{\text{Coulomb}} + \sum_{\kappa, \varepsilon} \hbar \omega_\kappa \left( a_{\kappa \varepsilon}^\dagger a_{\kappa \varepsilon} + \frac{1}{2} \right) \]

kinetic  Zeeman

Darwin  free photons
spin-orbit coupling
electron-nucleus  electron-electron
Interaction of light and matter: Hamiltonian

\[ H = \sum_{i=1}^{N} \left[ \frac{(p_i + eA(r_i))^2}{2m} + \frac{e\hbar}{2m} \sigma_i \cdot B(r_i) + \frac{e\hbar}{2(2mc)^2} \times \sigma_i \cdot \left( E(r_i) \times (p_i + eA(r_i)) - (p_i + eA(r_i)) \times E(r_i) \right) \right] + \frac{e\hbar^2 \rho(r_i)}{8(mc)^2 \epsilon_0} + H_{\text{Coulomb}} + \sum_{\kappa, \epsilon} \hbar \omega_\kappa \left( a_{\kappa \epsilon}^\dagger a_{\kappa \epsilon} + \frac{1}{2} \right) \]

- kinetic
- Zeeman
- Darwin
- free photons
- spin-orbit coupling
- electron-nucleus coupling
- electron-electron coupling
- vector potential
- plane wave

\[ A(r) = \sum_{\kappa, \epsilon} \sqrt{\frac{\hbar}{2\sqrt{\epsilon_0 \omega_\kappa}}} \left( \epsilon a_{\kappa \epsilon} e^{i\kappa \cdot r} + \epsilon^* a_{\kappa \epsilon}^\dagger e^{-i\kappa \cdot r} \right) \]
Lowest order perturbing Hamiltonian: small A

\[ H' = \sum_{i=1}^{N} \left[ \frac{e}{m} \mathbf{A}(\mathbf{r}_i) \cdot \mathbf{p}_i + \frac{e^2}{2m} \mathbf{A}^2(\mathbf{r}_i) + \frac{\epsilon}{2m} \sigma_i \cdot \nabla \times \mathbf{A}(\mathbf{r}_i) \\
- \frac{e^2 \hbar}{(2mc)^2} \sigma_i \cdot \frac{\partial \mathbf{A}(\mathbf{r}_i)}{\partial t} \times \mathbf{A}(\mathbf{r}_i) \right], \]
Lowest order perturbing Hamiltonian: small $A$

\[
H' = \sum_{i=1}^{N} \left[ \frac{e}{m} \mathbf{A}(\mathbf{r}_i) \cdot \mathbf{p}_i + \frac{e^2}{2m} A^2(\mathbf{r}_i) + \frac{e\hbar}{2m} \sigma_i \cdot \nabla \times \mathbf{A}(\mathbf{r}_i) \right] - \frac{e^2\hbar}{(2mc)^2} \sigma_i \cdot \frac{\partial \mathbf{A}(\mathbf{r}_i)}{\partial t} \times \mathbf{A}(\mathbf{r}_i)
\]

small
Lowest order perturbing Hamiltonian: small $A$

$$H' = \sum_{i=1}^{N} \left[ \frac{e}{m} A(r_i) \cdot p_i + \frac{e^2}{2m} A^2(r_i) + \frac{e\hbar}{2m} \sigma_i \cdot \nabla \times A(r_i) - \frac{e^2\hbar}{(2mc)^2} \sigma_i \cdot \frac{\partial A(r_i)}{\partial t} \times A(r_i) \right],$$

Fermi Golden Rule, to second order:

$$w = \frac{2\pi}{\hbar} \sum_{f} \left| \langle f \mid H' \mid g \rangle \right|^2 + \sum_{n} \frac{\langle f \mid H' \mid n \rangle \langle n \mid H' \mid g \rangle}{E_g - E_n} \delta(E_f - E_g)$$
\[
\frac{e^2}{2m} \langle f | \sum_i A^2(r_i) | g \rangle = \frac{\hbar e^2}{2m \mathcal{V} \varepsilon_0} \frac{\epsilon'^* \cdot \epsilon}{\sqrt{\omega_k \omega_{k'}}} \langle f | \sum_i e^{iq \cdot r_i} | g \rangle
\]
1st Order: Thompson Scattering

\[ \frac{e^2}{2m} \langle f | \sum_i A^2(r_i) | g \rangle = \frac{\hbar e^2}{2m \sqrt{\epsilon_0}} \frac{\epsilon^* \cdot \epsilon}{\sqrt{\omega_k \omega_k'}} \langle f | \sum_i e^{i q \cdot r_i} | g \rangle \]

Fourier transform of charge density

Elastic part: causes Bragg scattering & diffraction
1st Order: Thompson Scattering

\[ \frac{e^2}{2m} \langle f | \sum_i A^2(r_i) | g \rangle = \frac{\hbar e^2}{2m \sqrt{\epsilon_0 \epsilon}} \cdot \epsilon' \cdot \epsilon \cdot \frac{1}{\sqrt{\omega_k \omega_{k'}}} \cdot \langle f | \sum_i e^{i \mathbf{q} \cdot \mathbf{r}_i} | g \rangle \]

- Elastic part: causes Bragg scattering & diffraction
- Inelastic part: causes Inelastic X-ray Scattering (IXS)
  = Non-Resonant Inelastic X-ray Scattering (NIXS)

Fourier transform of charge density
1st Order: Thompson Scattering

Elastic part: causes Bragg scattering & diffraction

Inelastic part: causes Inelastic X-ray Scattering (IXS)

= Non-Resonant Inelastic X-ray Scattering (NIXS)

transition operator

\[ e^{iqr} \approx 1 + iqr - \frac{1}{2} (qr)^2 + \ldots \]
1st Order: Thompson Scattering

\[
\frac{e^2}{2m} \langle f | \sum_i A^2(r_i) | g \rangle = \frac{\hbar e^2}{2m \mathcal{V} \varepsilon_0} \frac{\epsilon' \cdot \epsilon}{\sqrt{\omega_k \omega_k'}} \langle f | \sum_i e^{i \mathbf{q} \cdot \mathbf{r}_i} | g \rangle
\]

Fourier transform of charge density

**Elastic part:** causes Bragg scattering & diffraction

**Inelastic part:** causes Inelastic X-ray Scattering (IXS)

\[= \text{Non-Resonant Inelastic X-ray Scattering (NIXS)}\]

**transition operator**

\[e^{i \mathbf{q} \cdot \mathbf{r}} \approx 1 + i \mathbf{q} \cdot \mathbf{r} - \frac{1}{2} (\mathbf{q} \cdot \mathbf{r})^2 + \ldots\]

**quadrupole**

**dipole**
1st order: X-ray Absorption

\[ \langle f | A \cdot p | g \rangle \]
1st order: X-ray Absorption

\[ \langle f | A \cdot p | g \rangle \propto \langle f | e^{ikr} \cdot p | g \rangle \]
1st order: X-ray Absorption

\[
\langle f | A \cdot p | g \rangle \propto \langle f | e^{ikr} \varepsilon \cdot p | g \rangle \approx \langle f | \varepsilon \cdot p | g \rangle
\]

When expanding \( e^{iqr} \approx 1 + iqr - \frac{1}{2} (qr)^2 + \ldots \)
1st order: X-ray Absorption

\[ \langle f | A \cdot p | g \rangle \propto \langle f | e^{iqr} \cdot p | g \rangle \approx \langle f | \cdot p | g \rangle \]

when expanding \( e^{iqr} \approx 1 + iq - \frac{1}{2}(qr)^2 + \ldots \)

dipole:

transitions caused by momentum operator \( p \)
Fermi Golden Rule, to second order:

\[ H' = \sum_{i=1}^{N} \frac{e}{m} \mathbf{A}(\mathbf{r}_i) \cdot \mathbf{p}_i + \frac{e^2}{2m} \mathbf{A}^2(\mathbf{r}_i) + \frac{e\hbar}{2m} \mathbf{\sigma}_i \cdot \nabla \times \mathbf{A}(\mathbf{r}_i) \]

\[
w = \frac{2\pi}{\hbar} \sum_{f} \left| \langle f | H' | g \rangle \right|^2 
+ \sum_{n} \frac{\langle f | H' | n \rangle \langle n | H' | g \rangle}{E_g - E_n} \left| \delta(E_f - E_g) \right|^2
\]
2nd Order

\[ H' = \sum_{i=1}^{N} \frac{e}{m} A(r_i) \cdot p_i + \frac{e\hbar}{2m} \sigma_i \cdot \nabla \times A(r_i) \]

Fermi Golden Rule, to second order:

\[ w = \frac{2\pi}{\hbar} \sum_{f} \left| \langle f | H' | g \rangle \right|^2 + \sum_{n} \frac{\langle f | H' | n \rangle \langle n | H' | g \rangle}{E_g - E_n} \delta(E_f - E_g) \]
H.A. Kramers and W. Heisenberg, Z. Phys. 31, 681 (1925)
2nd Order: Resonant Scattering I

\[ \frac{e^2 \hbar}{2m^2 V \varepsilon_0 \sqrt{\omega_k \omega_{k'}}} \sum_n \sum_{i,j=1}^N \left( \langle f | e^{-ik' \cdot r_i} \left( \epsilon'^* \cdot p_i - \frac{i\hbar}{2} \sigma_i \cdot k' \times \epsilon'^* \right) | n \rangle \right. \\
\left. \times \frac{E_g + \hbar \omega_k - E_n + i \Gamma_n}{E_g + \hbar \omega_{k'} - E_n + i \Gamma_n} \langle n | e^{ik \cdot r_j} \left( \epsilon \cdot p_j + \frac{i\hbar}{2} \sigma_j \cdot k \times \epsilon \right) | g \rangle \right) \]

small

H.A. Kramers and W. Heisenberg, Z. Phys. 31, 681 (1925)
2nd Order: Resonant Scattering I

RIXS amplitude

\[
\frac{e^2 \hbar}{2m^2 V\epsilon_0 \sqrt{\omega_k \omega_{k'}}} \sum_n \sum_{i,j=1}^N \langle f | e^{-i\mathbf{k'} \cdot \mathbf{r}_i} \left( \epsilon^{\prime \ast} \cdot \mathbf{p}_i - \frac{i\hbar}{2} \sigma_i \cdot \mathbf{k'} \times \epsilon^{\prime \ast} \right) | n \rangle \\
\times \frac{E_g + \hbar \omega_k - E_n + i\Gamma_n}{\epsilon \cdot \mathbf{p}_j + \frac{i\hbar}{2} \sigma_j \cdot \mathbf{k} \times \epsilon} \langle n | e^{ik \cdot \mathbf{r}_j} \left( \epsilon \cdot \mathbf{p}_j + \frac{i\hbar}{2} \sigma_j \cdot \mathbf{k} \times \epsilon \right) | g \rangle 
\]

\[D = \frac{1}{im\omega_k} \sum_{i=1}^N e^{ik \cdot r_i} \epsilon \cdot p_i,
\]

H.A. Kramers and W. Heisenberg, Z. Phys. 31, 681 (1925)
2nd Order: Resonant Scattering I

\[ \frac{e^2 \hbar}{2m^2 V \epsilon_0 \sqrt{\omega_k \omega_{k'}}} \sum_n \sum_{i,j=1}^{N} \]
\[ \langle f | e^{-i k' \cdot r_i} (\epsilon'^* \cdot p_i - \frac{i \hbar}{2} \sigma_i \cdot k' \times \epsilon'^*) | n \rangle \]
\[ \times \frac{1}{E_g + \hbar \omega_k - E_n + i \Gamma_n} \]
\[ \times \langle n | e^{i k \cdot r_j} (\epsilon \cdot p_j + \frac{i \hbar}{2} \sigma_j \cdot k \times \epsilon) | g \rangle \]

\[ D = \frac{1}{i m \omega_k} \sum_{i=1}^{N} e^{i k \cdot r_i} \epsilon \cdot p_i, \]

\[ F_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f | D'^{\dagger} | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i \Gamma_n} \]

H.A. Kramers and W. Heisenberg, Z. Phys. 31, 681 (1925)
2nd Order: Resonant Scattering I

\[ \frac{e^2 \hbar}{2m^2 V \epsilon_0 \sqrt{\omega_k \omega_k'}} \sum_n \sum_{i,j=1}^N \]
\[ \times \frac{\langle f | e^{-i k' \cdot r_i} \left( \epsilon' \cdot p_i - \frac{i \hbar}{2} \sigma_i \cdot k' \times \epsilon' \right) | n \rangle}{E_g + \hbar \omega_k - E_n + i \Gamma_n} \]
\[ \times \langle n | e^{i k \cdot r_j} \left( \epsilon \cdot p_j + \frac{i \hbar}{2} \sigma_j \cdot k \times \epsilon \right) | g \rangle \]

\[ D = \frac{1}{i m \omega_k} \sum_{i=1}^N e^{i k \cdot r_i} \epsilon \cdot p_i, \]

\[ F_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_k') = \sum_n \frac{\langle f | D^{\dagger} | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i \Gamma_n} \]

H.A. Kramers and W. Heisenberg, Z. Phys. 31, 681 (1925)
2nd Order: Resonant Scattering II

**RIXS amplitude**

\[
\mathcal{F}_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f | D| n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i\Gamma_n}
\]
2nd Order: Resonant Scattering II

RIXS amplitude

\[ \mathcal{F}_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f | D' \dagger | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i\Gamma_n} \]

RIXS intensity:

\[ I(\omega, k, k', \epsilon, \epsilon') = r_e^2 m^2 \omega_k^3 |\mathcal{F}_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'})|^2 \times \delta(E_g - E_f + \hbar \omega), \]
**2nd Order: Resonant Scattering II**

**RIXS amplitude**

\[
\mathcal{F}_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f | D'^\dagger | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i\Gamma_n}
\]

**RIXS intensity:**

\[
I(\omega, k, k', \epsilon, \epsilon') = r^2 m^2 \omega^3 k \sum_f |\mathcal{F}_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'})|^2 \times \delta(E_g - E_f + \hbar \omega),
\]

*This expression is essentially exact (non-relativistic limit)*
Resonant ELASTIC Scattering (REXS or RXS)

\[ \mathcal{F}_{fg}(\mathbf{k}, \mathbf{k}', \varepsilon, \varepsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f \mid \mathcal{D}^\dagger \mid n \rangle \langle n \mid \mathcal{D} \mid g \rangle}{E_g + \hbar \omega_k - E_n + i\Gamma_n} \]
Resonant ELASTIC Scattering (REXS or RXS)

REXS amplitude

\[ \mathcal{F}_{gg}(\mathbf{k}, \mathbf{k}', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle g | D^\dagger | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i\Gamma_n} \]
Resonant ELASTIC Scattering (REXS or RXS)

**REXS amplitude**

\[ F_{g g} (k, k', \epsilon, \epsilon', \omega_k, \omega_k') = \sum_n \frac{\langle g | D^\dagger | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i \Gamma_n} \]

**REXS intensity:**

\[ I(\omega, k, k', \epsilon, \epsilon') = r^2_e m^2 \omega_k^3 \omega_k \sum_f |F_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_k')|^2 \]

\[ \times \delta(E_g - E_f + \hbar \omega), \]
Resonant ELASTIC Scattering (REXS or RXS)

**REXS amplitude**

\[
\mathcal{F}_{gg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{|g \langle D^\dagger | n \rangle \langle n | D | g \rangle|}{E_g + \hbar \omega_k - E_n + i\Gamma_n}
\]

**REXS intensity:**

\[
I(k, k', \epsilon, \epsilon') = r_e^2 m^2 \omega_k^3 \omega_{k'} \sum_f |\mathcal{F}_{gg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'})|^2
\]
Greens function expression for $F$

$$F_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f | D^\dagger | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i\Gamma_n}$$
Greens function expression for $F$

$$\mathcal{F}_{fg}(\mathbf{k}, \mathbf{k}', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f | D^\dagger | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i\Gamma_n}$$

Greens function

$$G(z_k) = \frac{1}{z_k - H} = \sum_n \frac{|n\rangle \langle n|}{z_k - E_n}$$
Greens function expression for $F$

$$\mathcal{F}_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f | D^\dagger | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i\Gamma_n}$$

Greens function

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

with

$$z_k = E_g + \hbar \omega_k + i\Gamma$$
Greens function expression for $F$

$$F_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f | D^\dagger | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i\Gamma_n}$$

Greens function

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

= intermediate state propagator

with

$$z_k = E_g + \hbar \omega_k + i\Gamma$$
Greens function expression for $F$

$\mathcal{F}_{fg}(k, k', \epsilon, \epsilon', \omega_k, \omega_{k'}) = \sum_n \frac{\langle f | D'^\dagger | n \rangle \langle n | D | g \rangle}{E_g + \hbar \omega_k - E_n + i \Gamma_n}$

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

= intermediate state propagator

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

so that:

$\mathcal{F}_{fg} = \langle f | D'^\dagger G(z_k) D | g \rangle$
RIXS spin-flip amplitude @ transition metal L-edge

$x^2 - y^2$ spin NOT // z: pure spin flip

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

Quasi 2D Cuprates
Magnetic RIXS on $\text{La}_2\text{CuO}_4$ @ Cu L-edge
Magnetic RIXS on $\text{La}_2\text{CuO}_4 @ \text{Cu L-edge}$

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

CuO’s are such special cases...
Magnetic RIXS on $\text{La}_2\text{CuO}_4$ @ Cu L-edge

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

CuO’s are such special cases...
Magnetic RIXS on La$_2$CuO$_4$ @ Cu L-edge

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

CuO's are such special cases...

[Graph showing RIXS spectrum with annotations for phonon and zero-loss peaks]
Magnetic RIXS on La$_2$CuO$_4$ @ Cu L-edge

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

CuO's are such special cases...

[Diagram showing Cu L-edge spectra with peaks labeled as magnon, phonon, and zero-loss.]
Magnetic RIXS on $\text{La}_2\text{CuO}_4$ @ Cu L-edge

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

CuO’s are such special cases...

![Graph showing high resolution Cu L-edge RIXS spectrum.](image)
Magnetic direct RIXS on La$_2$CuO$_4$ @ Cu L-edge

Braicovich, JvdB et al., PRL 104, 077002 (2010)
Magnetic direct RIXS on La$_2$CuO$_4$ @ Cu L-edge

Braicovich, JvdB et al., PRL 104, 077002 (2010)
RIXS magnon dispersion of $\text{Sr}_2\text{CuO}_2\text{Cl}_2$

$\rightarrow$ transferred momentum $q$

Guarise et al.,
PRL 105, 157006 (2010)
RIXS magnon dispersion of $\text{Sr}_2\text{CuO}_2\text{Cl}_2$

development from
simple Heisenberg

$\rightarrow$ transferred momentum $q$

Guarise et al.,
PRL 105, 157006 (2010)
Quasi 1D Cuprate
Cuprate spin chain system $\text{Sr}_2\text{CuO}_3$

weakly coupled spin chains
Cuprate spin chain system $\text{Sr}_2\text{CuO}_3$

weakly coupled spin chains
Cuprate spin chain system $\text{Sr}_2\text{CuO}_3$

weakly coupled spin chains

$\text{Cu}^{2+}$
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad S^z = -\hbar/2 \]

\[ H = J S^z_i S^z_{i+1} \]

Allow for spin-exchange
Heisenberg antiferromagnet

\[ S_z^z = +\frac{\hbar}{2} \quad \text{and} \quad S_z^z = -\frac{\hbar}{2} \]

Allow for spin-exchange

\[ H = J S_z^z S_z^{z_{i+1}} \]
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad \text{and} \quad S^z = -\hbar/2 \]

\[ H = J \, S^z_i \, S^z_{i+1} \]

Allow for spin-exchange
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad \text{and} \quad S^z = -\hbar/2 \]

\[ H = J S^z_i S^z_{i+1} \]

Allow for spin-exchange
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad \text{or} \quad S^z = -\hbar/2 \]

\[ H = J S_i^z S_{i+1}^z + J S_i^y S_{i+1}^y + J S_i^x S_{i+1}^x \]

Allow for spin-exchange
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad S^z = -\hbar/2 \]

\[ H = J S^z_i S^z_{i+1} + J S^y_i S^y_{i+1} + J S^x_i S^x_{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1} \]

Allow for spin-exchange
Heisenberg antiferromagnet

$S^z = +\hbar/2 \quad S^z = -\hbar/2$

$H = J S^z_i S^z_{i+1} + J S^y_i S^y_{i+1} + J S^x_i S^x_{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1}$

Allow for spin-exchange

ground state
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad \text{or} \quad S^z = -\hbar/2 \]

\[ H = J S^z_i S^z_{i+1} + J S^y_i S^y_{i+1} + J S^x_i S^x_{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1} \]

Allow for spin-exchange

spin flip \( \Delta S^z = \hbar \)

\text{in } d=3: \text{ magnon}
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad S^z = -\hbar/2 \]

\[ H = J S^z_i S^z_{i+1} + J S^y_i S^y_{i+1} + J S^x_i S^x_{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1} \]

Allow for spin-exchange

spin flip \( \Delta S^z = \hbar \)

in \( d=3 \): magnon
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad S^z = -\hbar/2 \]

\[ H = J S^z_i S^z_{i+1} + J S^y_i S^y_{i+1} + J S^x_i S^x_{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1} \]

Allow for spin-exchange

Spin flip \( \Delta S^z = \hbar \)

In \( d=3 \): magnon
Heisenberg antiferromagnet

\[ S_z^z = \pm \hbar/2 \]

\[ H = J S_z^i S_z^{i+1} + J S_y^i S_y^{i+1} + J S_x^i S_x^{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1} \]

Allow for spin-exchange

Spin flip \( \Delta S_z = \hbar \)

In \( d=3 \): magnon
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad S^z = -\hbar/2 \]

\[
H = J S^z_i S^z_{i+1} + J S^y_i S^y_{i+1} + J S^x_i S^x_{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1}
\]

Allow for spin-exchange

Spin flip \( \Delta S^z = \hbar \) in \( d=3 \): magnon
Heisenberg antiferromagnet

\( S^z = +\hbar/2 \quad S^z = -\hbar/2 \)

\[ H = J S^z_i S^z_{i+1} + J S^y_i S^y_{i+1} + J S^x_i S^x_{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1} \]

Allow for spin-exchange

\( \Delta S^z = \hbar \)

in \( d=3 \): magnon

two domain walls, each with \( \Delta S^z = \hbar/2 \) → spinon
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad \text{and} \quad S^z = -\hbar/2 \]

\[ H = J \, S^z_i \, S^z_{i+1} + J \, S^y_i \, S^y_{i+1} + J \, S^x_i \, S^x_{i+1} = J \, \mathbf{S}_i \cdot \mathbf{S}_{i+1} \]

Allow for spin-exchange

Spin flip \( \Delta S^z = \hbar \)

In \( d=3 \): magnon

two domain walls, each with \( \Delta S^z = \hbar/2 \)

Spinon

\( d=1 \): magnon fractionalizes into spinons
Heisenberg antiferromagnet

\[ S^z = +\hbar/2 \quad \text{and} \quad S^z = -\hbar/2 \]

\[ H = J S^z_i S^z_{i+1} + J S^y_i S^y_{i+1} + J S^x_i S^x_{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1} \]

Allow for spin-exchange

Spin flip \( \Delta S^z = \hbar \)

In \( d=3 \): magnon

Two domain walls, each with \( \Delta S^z = \hbar/2 \)

Spinon

\( d=1 \): magnon fractionalizes into spinons

Magnon \( q, \omega \)

Spinon \( k_1, \omega_1 \), \( k_2, \omega_2 \)
Heisenberg antiferromagnet

$S^z = +\hbar/2$

$S^z = -\hbar/2$

$H = J S^z_i S^z_{i+1} + J S^y_i S^y_{i+1} + J S^x_i S^x_{i+1} = J \mathbf{S}_i \cdot \mathbf{S}_{i+1}$

Allow for spin-exchange

spin flip $\Delta S^z = \hbar$

in $d=3$: magnon

two domain walls, each with $\Delta S^z = \hbar/2$

spinon

d=1: magnon fractionalizes into spinons

magnon $q$, $\omega$  

spinon $k_1$, $\omega_1$

spinon $k_2$, $\omega_2$

$q = k_1 + k_2$, $\omega = \omega_1 + \omega_2$
Spinons in 1d Heisenberg antiferromagnet

magnon $q, \omega \rightarrow$ spinon $k_1, \omega_1$
spinon $k_2, \omega_2$

$q = k_1 + k_2, \omega = \omega_1 + \omega_2$
Spinons in 1d Heisenberg antiferromagnet

magnon $q, \omega$ \rightarrow spinon $k_1, \omega_1$
spinon $k_2, \omega_2$

$q = k_1 + k_2$, $\omega = \omega_1 + \omega_2$

relative momentum of spinons
$p = k_2 - k_1$ not yet determined
Spinons in 1d Heisenberg antiferromagnet

- magnon $q, \omega$ → spinon $k_1, \omega_1$
- spinon $k_2, \omega_2$
- $q = k_1 + k_2, \omega = \omega_1 + \omega_2$
- relative momentum of spinons $p = k_2 - k_1$ not yet determined

excitation continuum $q, \omega$
Spinon excitations in 1d Heisenberg antiferromagnet

Excitation continuum $q, \omega$

Relative momentum of spinons $q = k_2 - k_1$ not yet determined

Neutron scattering magnetic excitations

Bethe-Ansatz exact solution

Klauser, Mossel, Caux, JvdB, PRL 106, 157205 (2011)
RIXS spectrum of $\text{Sr}_2\text{CuO}_3$ spin chain

RIXS spectrum of $\text{Sr}_2\text{CuO}_3$ spin chain

**RIXS spectrum of Sr$_2$CuO$_3$ spin chain**

Spinons in $\text{Sr}_2\text{CuO}_3$

Spinons in \( \text{Sr}_2\text{CuO}_3 \)

Quasi 2D Iron Pnictide
Magnetic RIXS on $\text{BaFe}_2\text{As}_2$ @ Fe L-edge

Magnetic RIXS on BaFe$_2$As$_2$ @ Fe L-edge

Magnetic RIXS on BaFe$_2$As$_2$ @ Fe L-edge

Magnetic RIXS vs. Inelastic Neutron Scattering

- RIXS
- Neutrons

- Amount of material needed
- Magnon energy accessible
- Materials
Magnetic RIXS vs. Inelastic Neutron Scattering

- **amount of material needed**
  - RIXS: small
  - Neutrons: large

- **magnon energy accessible**
  - RIXS

- **materials**
**Magnetic RIXS vs. Inelastic Neutron Scattering**

- **amount of material needed**
  - **RIXS**: small
  - **Neutrons**: large

- **magnon energy accessible**
  - **RIXS**: high (>10^2 meV)
  - **Neutrons**: low (<10^2 meV)

- **materials**
**Magnetic RIXS vs. Inelastic Neutron Scattering**

- **amount of material needed**
  - RIXS: small
  - Neutrons: large

- **magnon energy accessible**
  - RIXS: high ($>10^2$ meV)
  - Neutrons: low ($<10^2$ meV)

- **materials**
  - RIXS: Cu, Fe ...
  - Neutrons: non-absorbers
Quasi 2D Iridate
Magnetic Iridium Oxides

$\text{Sr}_2\text{IrO}_4$: equivalent of cuprate $\text{La}_2\text{CuO}_4$

Jackeli & Khaliullin, PRL 102,017205 (2009)

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

B.J. Kim, Ohsumi, Komesu, Sakai, Morita, Takagi, Arima, Science 323, 1329 (2009)

Ir$^{4+}$

Ir (4+) = 5d$^5$

5x

e_g

t_{2g}
**Magnetic Iridium Oxides**

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\text{Sr}_2\text{IrO}_4 : \text{equivalent of cuprate } \text{La}_2\text{CuO}_4
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Jackeli & Khaliullin, PRL 102,017205 (2009)

B.J. Kim, Ohsumi, Komesu, Sakai, Morita, Takagi, Arima, Science 323, 1329 (2009)

\[t_{2g}^5: \text{single hole } s=1/2 \text{ in 3-fold degenerate } l=1 \text{ state}\]
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\( \text{Sr}_2\text{IrO}_4 \): equivalent of cuprate \( \text{La}_2\text{CuO}_4 \)

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

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

\[ J_{\text{eff}} = \frac{3}{2} \]

\[ 3\lambda/2 \quad \text{quartet} \]

\[ J=L+S \]

\[ J_{\text{eff}} = \frac{1}{2} \]

\[ \lambda/2 \quad \text{doublet} \]
Direct RIXS on $\text{Sr}_2\text{IrO}_4$
Direct RIXS on $\text{Sr}_2\text{IrO}_4$

- Spin-orbit exciton (optically forbidden)
- e-h continuum (optically allowed)
- Magnons

Energy (eV)

Intensity

3\hbar/2

Quartet

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

Ament, Khaliullin & JvdB
PRB 84, 020403 (2011)
Direct RIXS on Sr$_2$IrO$_4$

Magnon dispersion

π-pol

σ-pol

Ament, Khaliullin & JvdB
PRB 84, 020403 (2011)
Direct RIXS on $\text{Sr}_2\text{IrO}_4$

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

PRL 108, 177003 (2012)

Jungho Kim, D. Casa, M. H. Upton, T. Gog, Young-June Kim, J. F. Mitchell, M. van Veenendaal, M. Daghofer, J. van den Brink, G. Khaliullin, B. J. Kim
PRL 108, 177003 (2012)
Magnetic RIXS on
doped quasi-2D
Cu-oxides and Fe-pnictides
Magnetic L-edge RIXS on 8% doped La$_{2-x}$Sr$_x$CuO$_4$

$LSCO$

$T_c = 21K$

$T = 15 K$
Magnetic L-edge RIXS on 8% doped La$_{2-x}$Sr$_x$CuO$_4$

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d-d excitation
Magnetic L-edge RIXS on 8% doped La$_{2-x}$Sr$_x$CuO$_4$

$T_c = 21$ K

LSCO

$T = 15$ K

$d$-$d$ excitation

paramagnon

Braicovich, JvdB et al.
PRL 104, 077002 (2010)
Intense paramagnon excitations in a large family of high-temperature superconductors

magnon

M. Le Tacon¹, G. Ghiringhelli², J. Chaloupka¹, M. Moretti Sala², V. Hinkov¹, M. W. Haverkort¹, M. Minola², M. Bakr¹, K. J. Zhou⁴, S. Blanco-Canosa¹, C. Monney⁴, Y. T. Song¹, G. L. Sun¹, C. T. Lin¹, G. M. De Luca⁵, M. Salluzzo⁵, G. Khaliullin¹, T. Schmitt⁴, L. Braicovich² and B. Keimer¹
magnon

para-magnon

M. Le Tacon¹*, G. Ghiringhelli², J. Chaloupka¹, M. Moretti Sala², V. Hinkov¹,³, M. W. Haverkort¹,
M. Minola², M. Bakr¹, K. J. Zhou⁴, S. Blanco-Canosa¹, C. Monney⁴, Y. T. Song¹, G. L. Sun¹, C. T. Lin¹,
G. M. De Luca⁵, M. Salluzzo⁵, G. Khaliullin¹, T. Schmitt⁴, L. Braicovich² and B. Keimer¹*
Dynamical structure factor Hubbard model, QMC

\[ U = 8t \]

Jia, Nowadnick, Wohlfeld, Kung, Chen, Johnston, Tohyama, Moritz & Devereaux

Nat. Comm. 5, 3314 (2014)
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Nat. Comm. 5, 3314 (2014)
**Dynamical structure factor Hubbard model, QMC**

- **magnon**
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Jia, Nowadnick, Wohlfeld, Kung, Chen, Johnston, Tohyama, Moritz & Devereaux
Nat. Comm. 5, 3314 (2014)
RIXS on Bi-2212 cuprate

RIXS on Bi-2212 cuprate

RIXS on Bi-2212 cuprate

RIXS on Bi-2212 cuprate

RIXS on Bi-2212 cuprate

- **Magnon**
- **Paramagnon**

Related to presence of e-h continuum?

- Benjamin, Klich & Demler
  PRL 112, 247002 (2014)

Magnetic RIXS on $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$

Magnetic RIXS on $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$

Magnetic RIXS on $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$

Summary part 2
• RIXS sensitive to magnetic excitations of e.g. low D cuprates, iron pnictides and iridates


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- RIXS sensitive to magnetic excitations of e.g. low D cuprates, iron pnictides and iridates
- Magnons, spinons and paramagnons are observed
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• Dispersion of these modes can be determined
Summary part 2

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• Observed paramagnons are challenge for theory
Summary part 2

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• It can reasonably be assumed that the future of RIXS is even brighter than its past
Summary part 2

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  low D cuprates, iron pnictides and iridates

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• 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 beam-lines, experiments, theory