

Resonant x-ray scattering as a probe for exotic orders in condensed matter



B. J. Kim

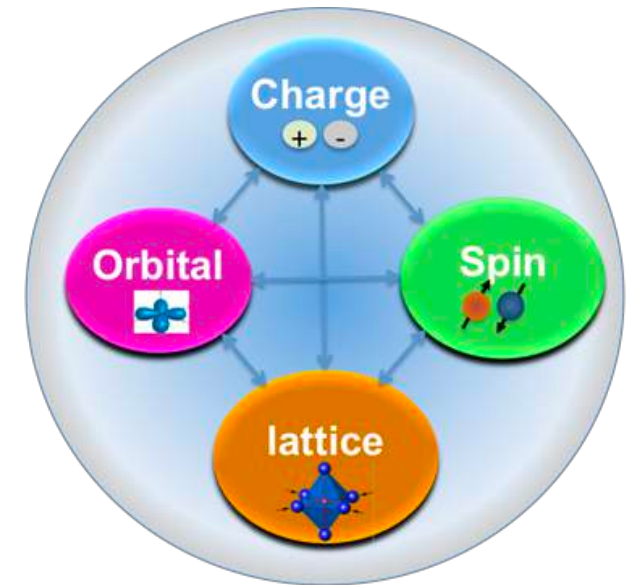
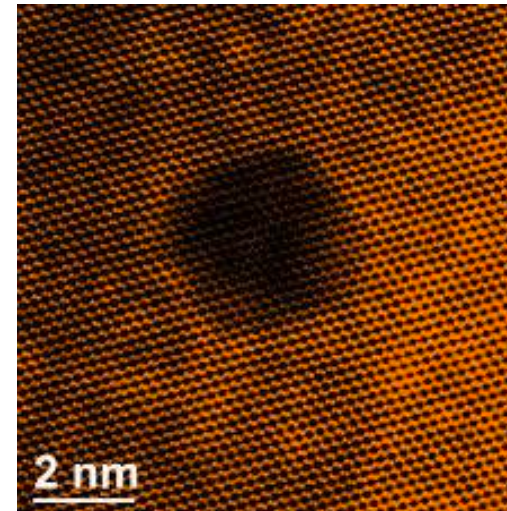
Pohang University of Science and Technology (POSTECH)

Center for Artificial Low-Dimensional Electronic Systems, Institute of Basic Science (IBS)

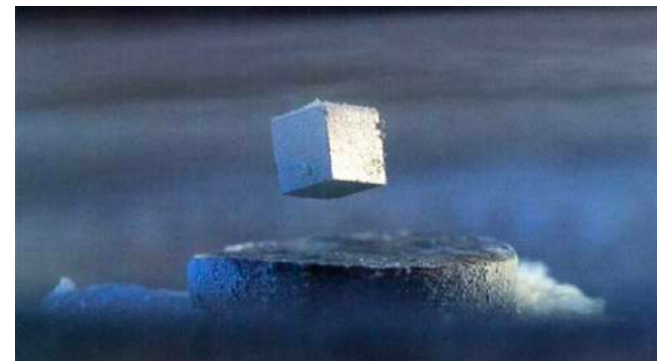
Colloquium
Seoul National University
Nov. 22, 2017

Condensed matter systems

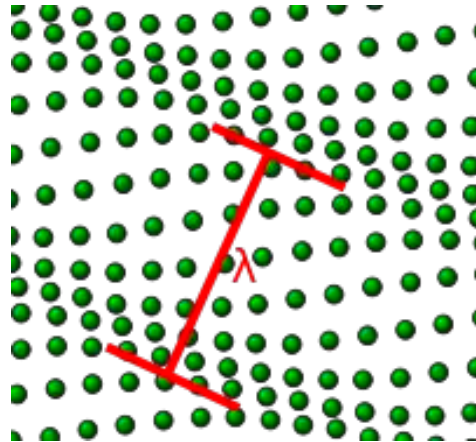
- Large number of interacting degrees of freedom
- Symmetry of the underlying lattice (230 space groups)
- Spontaneous breaking of the symmetry by electronic order
- Emergent physics



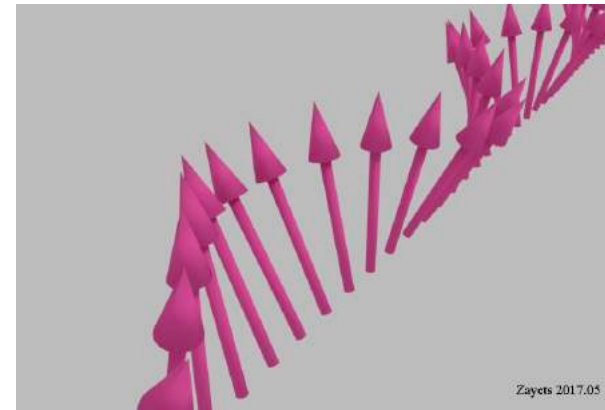
“More is different”



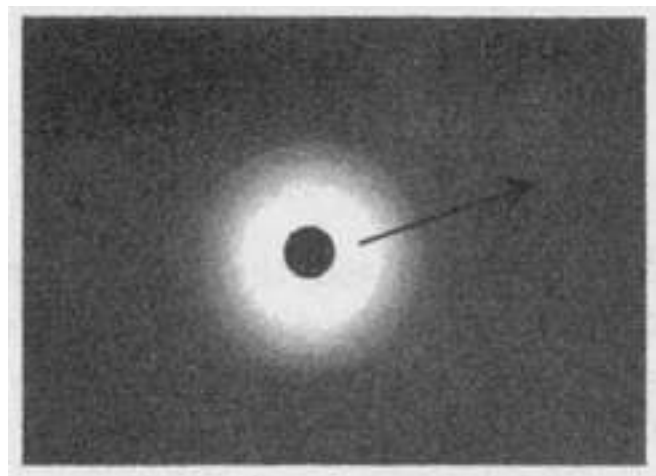
Elementary excitations in solids



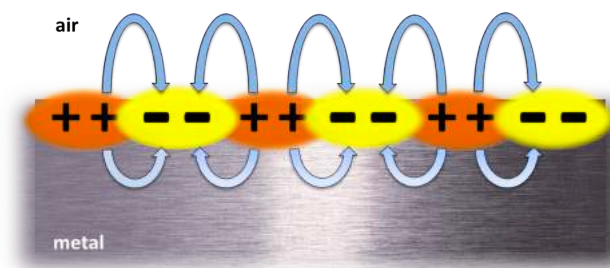
phonon



magnon

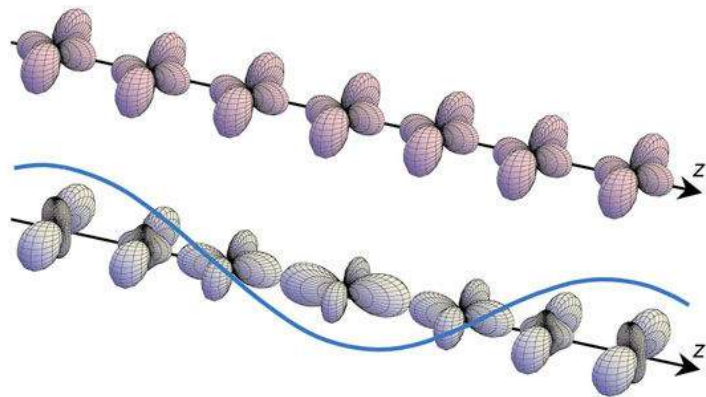


quasi-electron

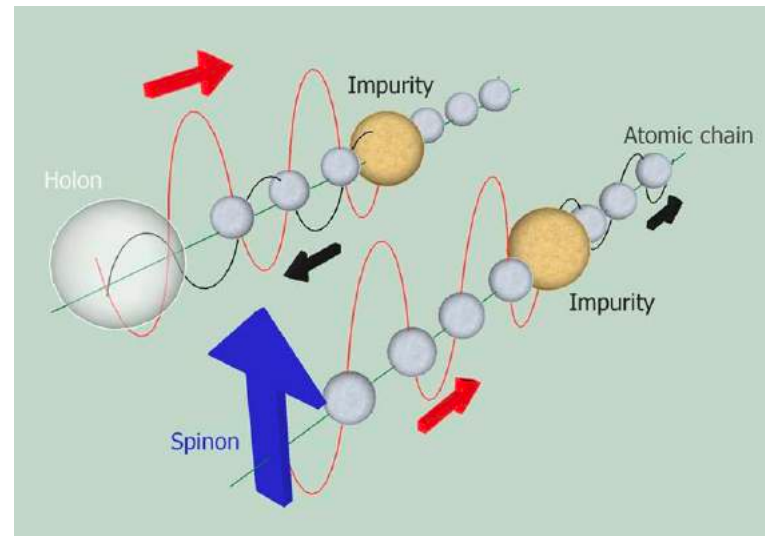


plasmon

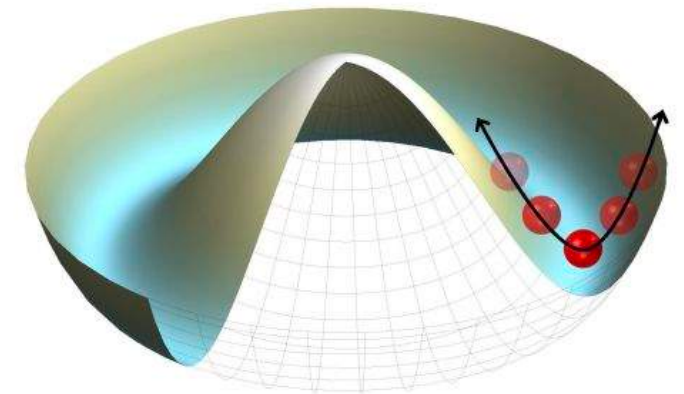
And more exotic particles..



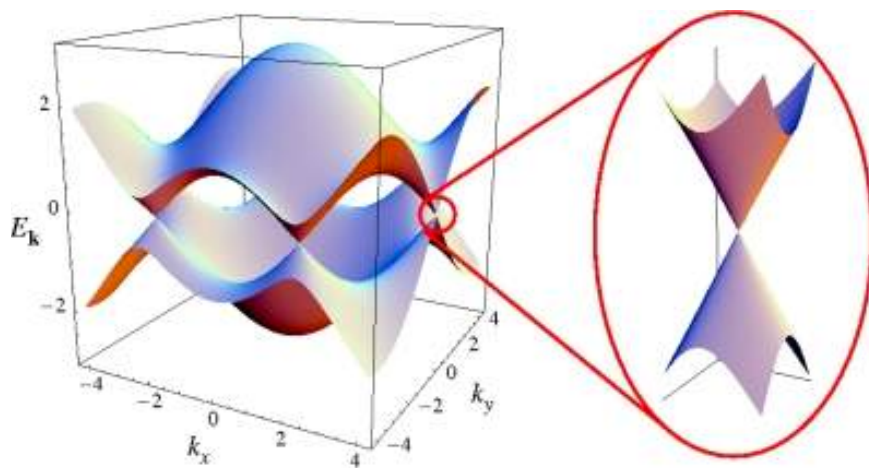
orbiton



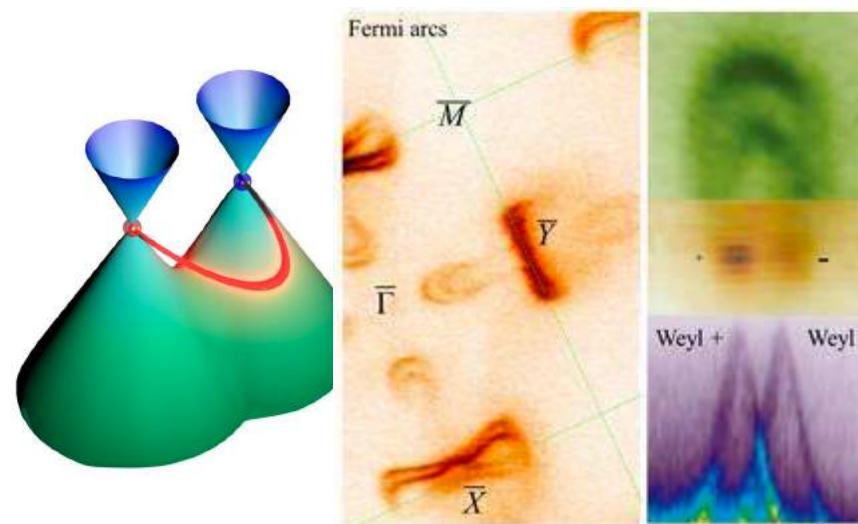
Spinon and holon



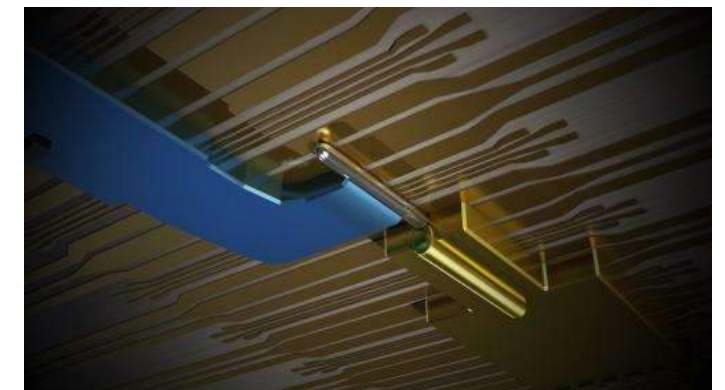
Higgs mode



Dirac fermion

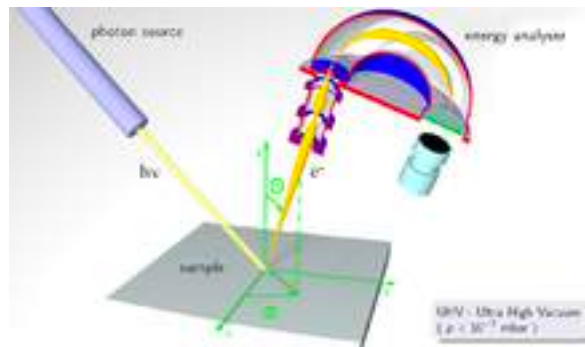


Weyl fermion

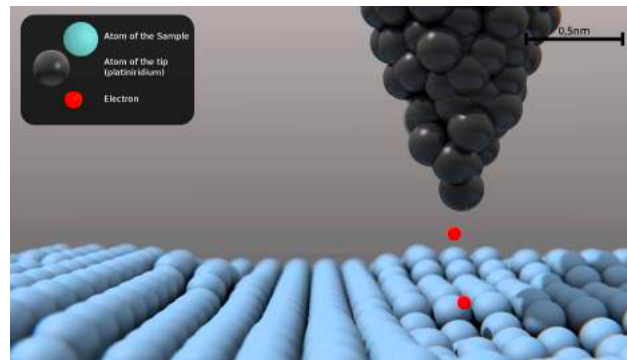


Majonara fermion

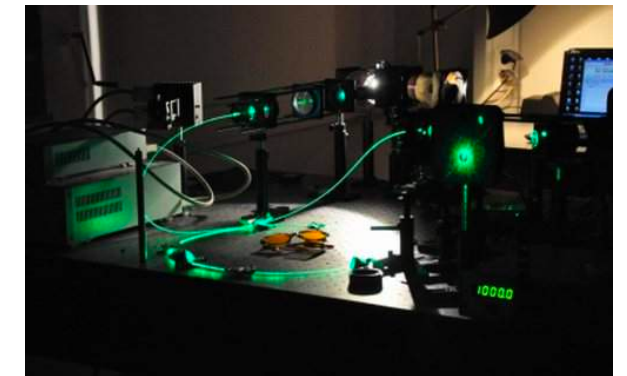
Standard probes



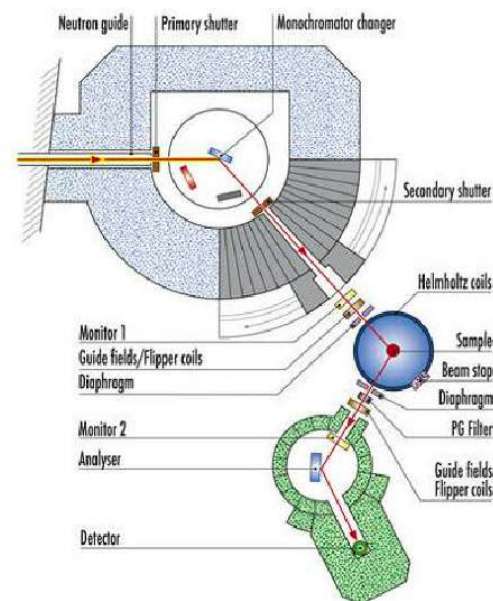
ARPES



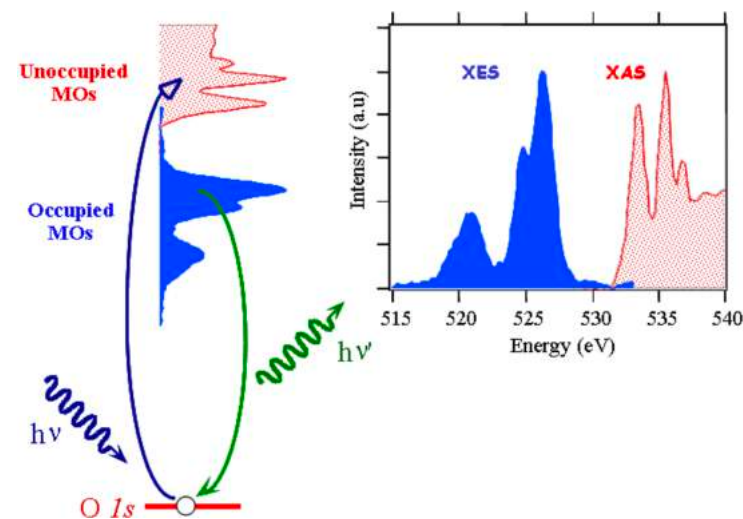
STM



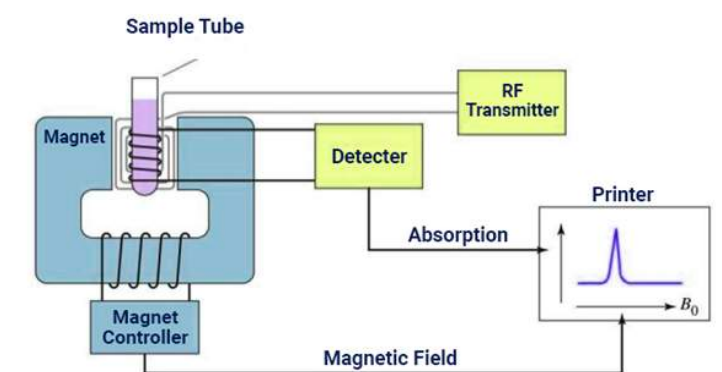
optical/Raman



INS

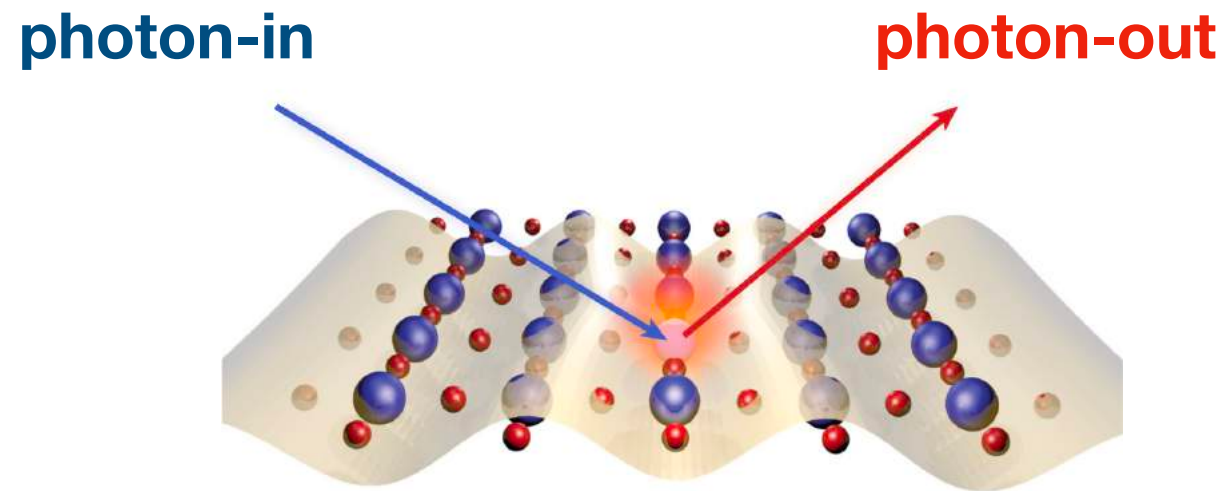


XAS/XES



NMR

Resonant inelastic x-ray scattering (RIXS)



Pros

- Sensitive to spin, orbital, charge excitations
- Sensitive to “hidden” orders
- Can measure up to hexadecapole (2^4 pole)
- Can measure tiny crystals/thin films

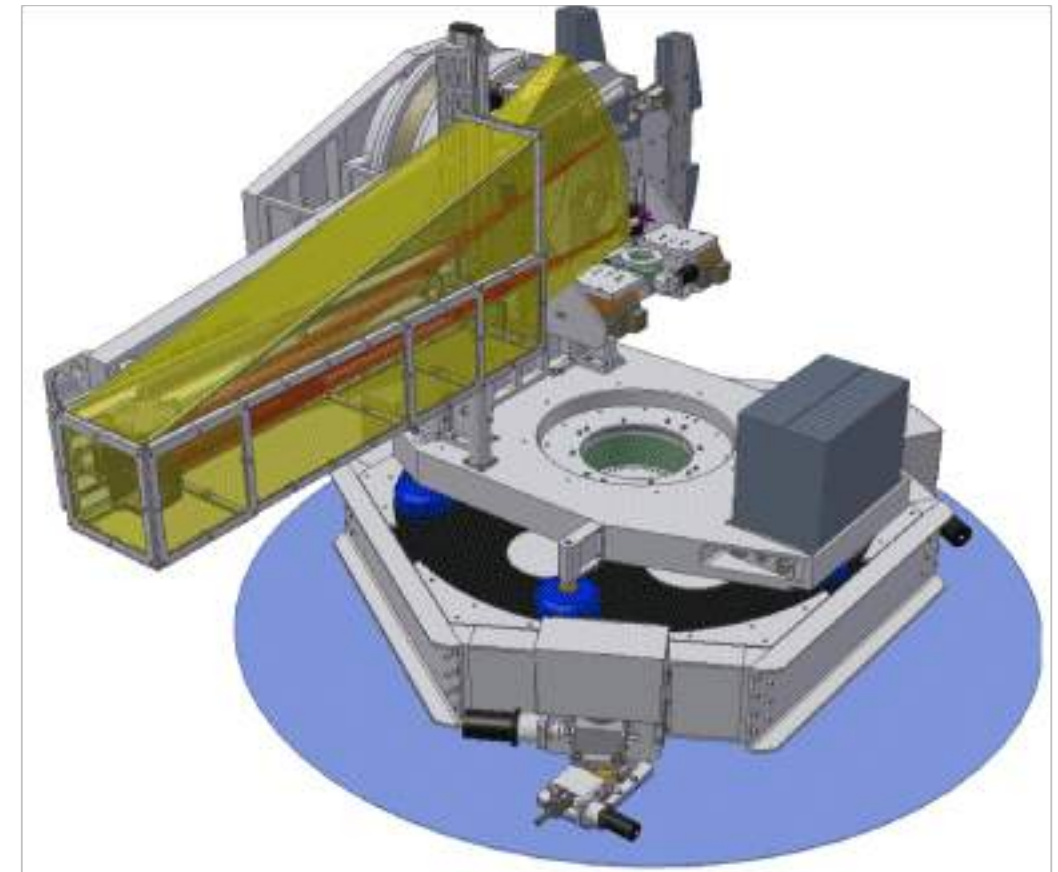
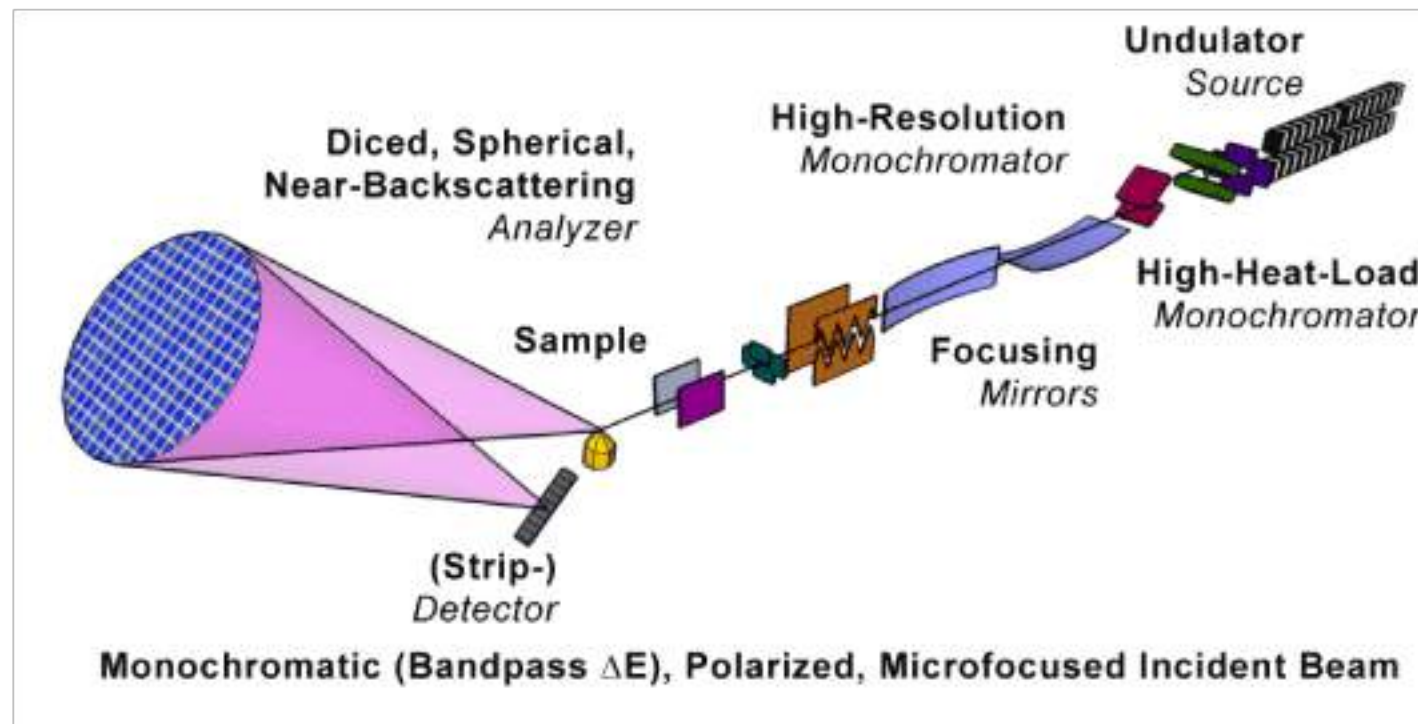
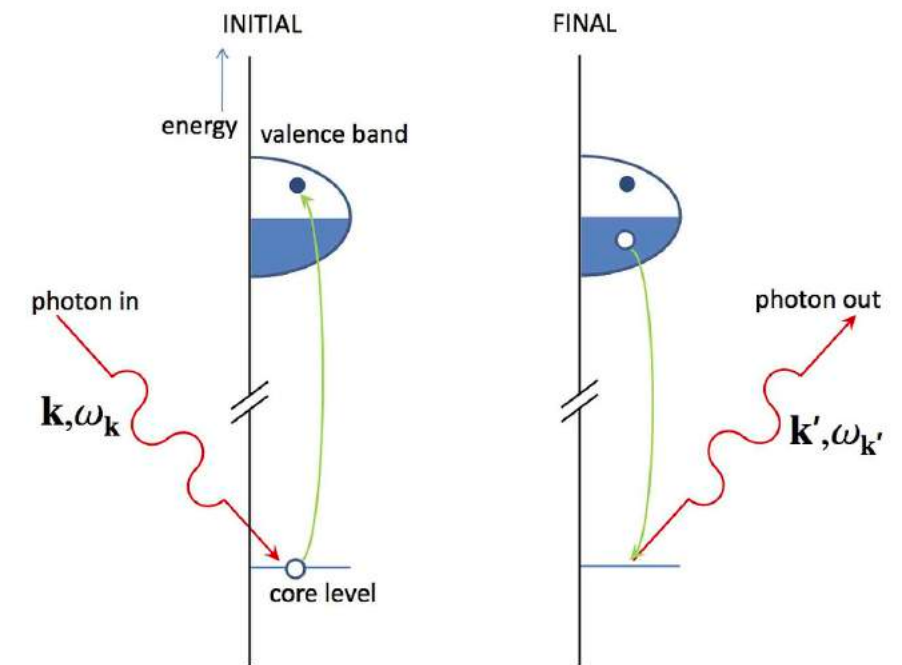
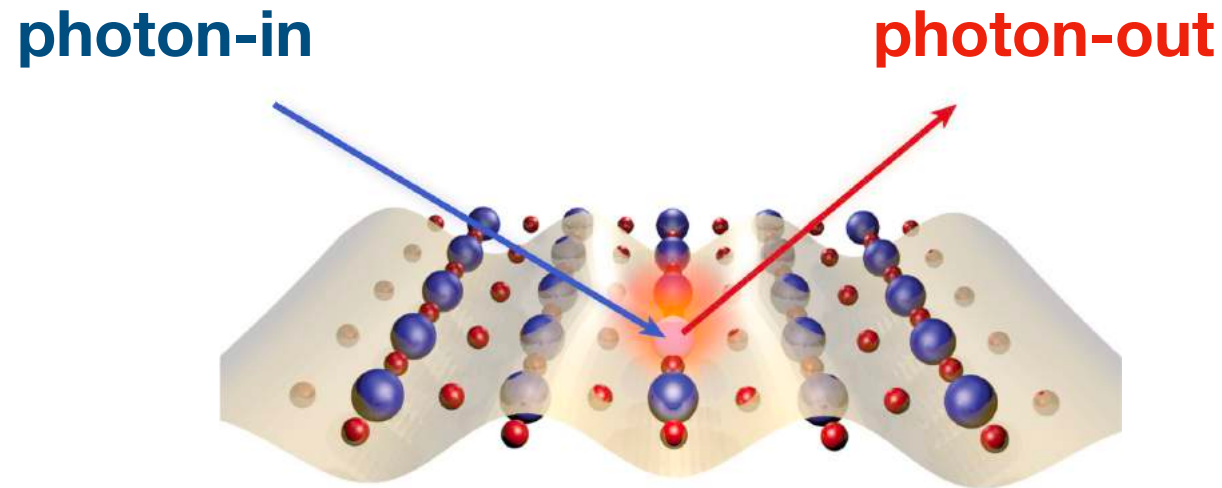
Cons

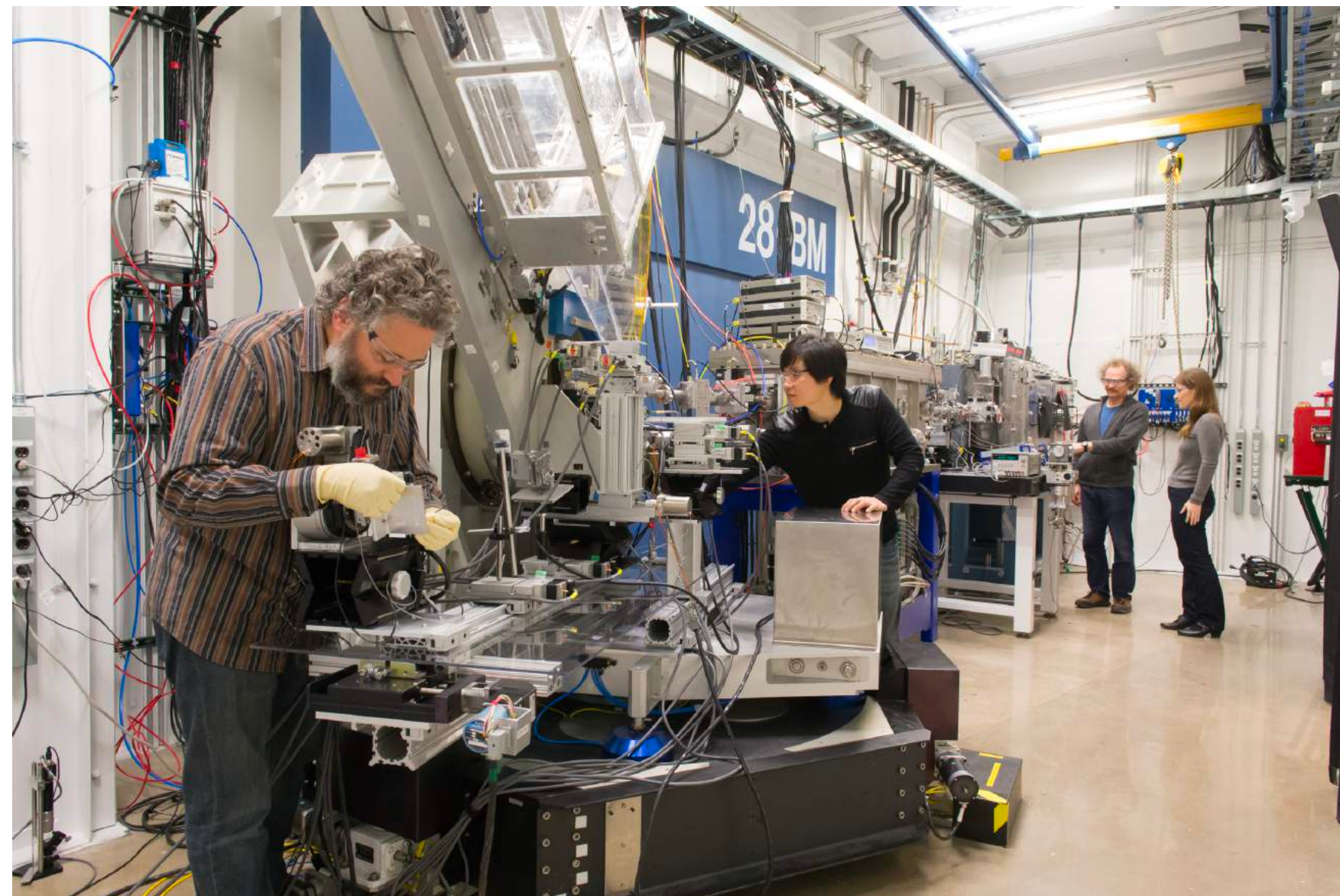
- Difficult to calculate the scattering cross section
- Energy resolution not as good as neutron scattering
- Difficult to get beamtime (not many user facilities)

Outline

- Short introduction to RIXS
- Applications to:
 1. Spin waves
 2. Excitons
 3. Fractional excitations
 4. Spin nematic
- Other interesting theoretical proposals to measure:
 5. Majorana fermions
 6. Gauge fields

Resonant inelastic x-ray scattering (RIXS)





Advanced
Photon
Source
ARGONNE NATIONAL LABORATORY




Soft X-Ray RIXS Spectrometer at ID32, ESRF



Brief history

VOLUME 33, NUMBER 5

PHYSICAL REVIEW LETTERS

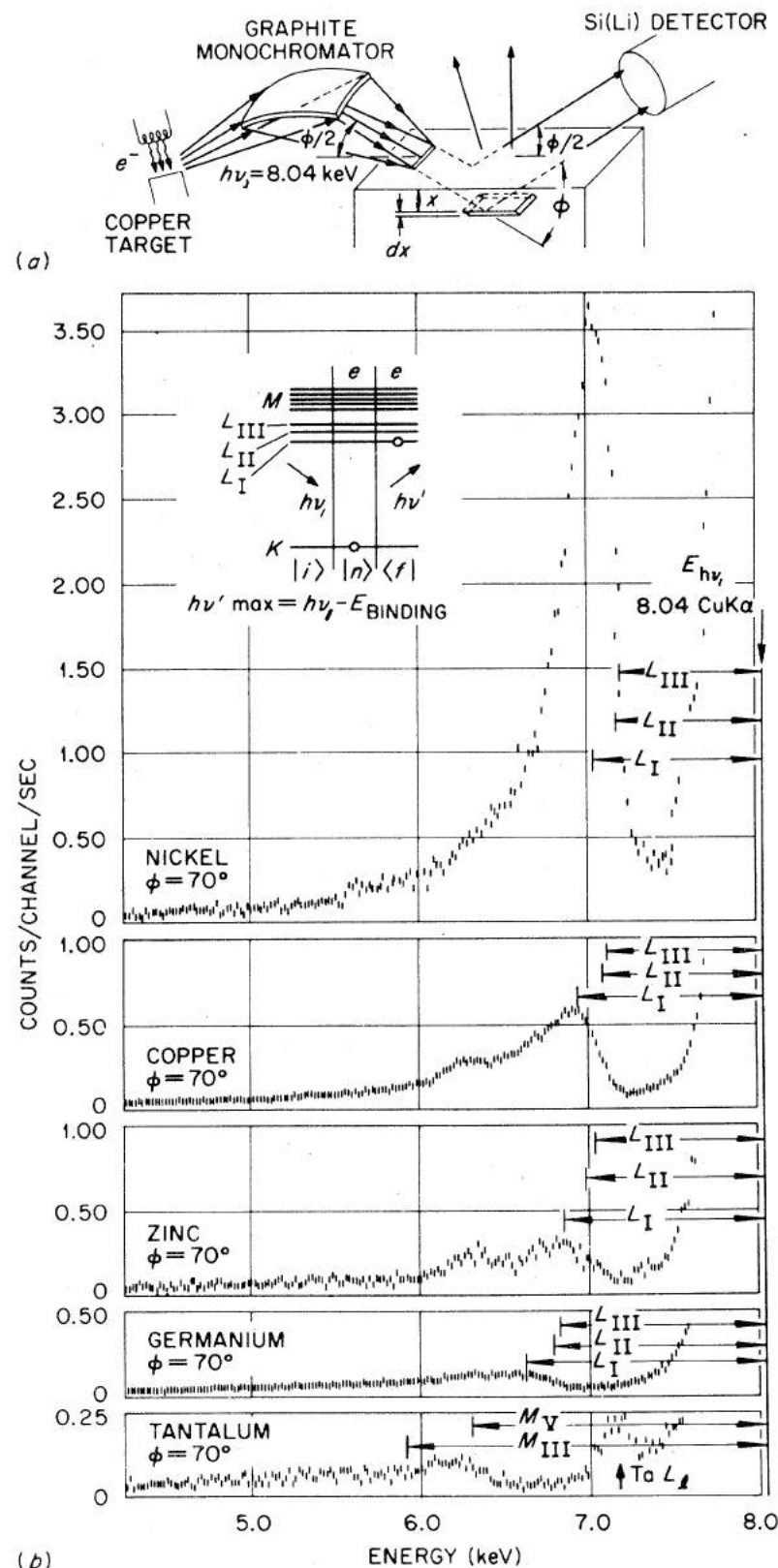
29 JULY 1974

Inelastic Resonance Emission of X Rays: Anomalous Scattering Associated with Anomalous Dispersion*

Cullie J. Sparks, Jr.

Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

(Received 13 May 1974)



190 eV resolution @ 5.9 keV

Brief history

our first experiment on iridate (hard x-ray)
first observation of single magnon (soft x-ray)
(Braicovich & Ghiringhelli)

2017: <10 meV
2012: 25 meV

Ir L3 edge 11.2 keV
resolving power $>10^6$

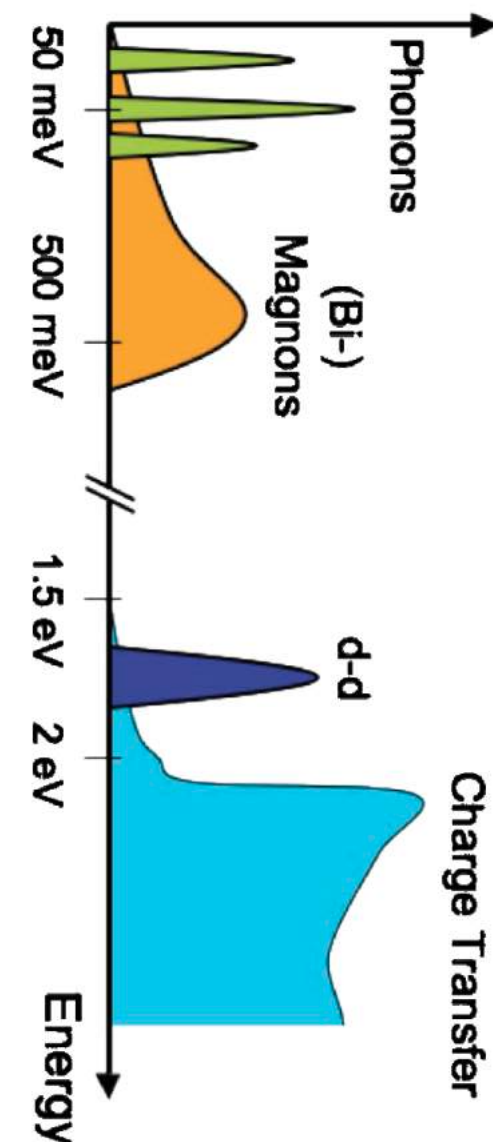
2010: 130 meV, 3000 cps

2010

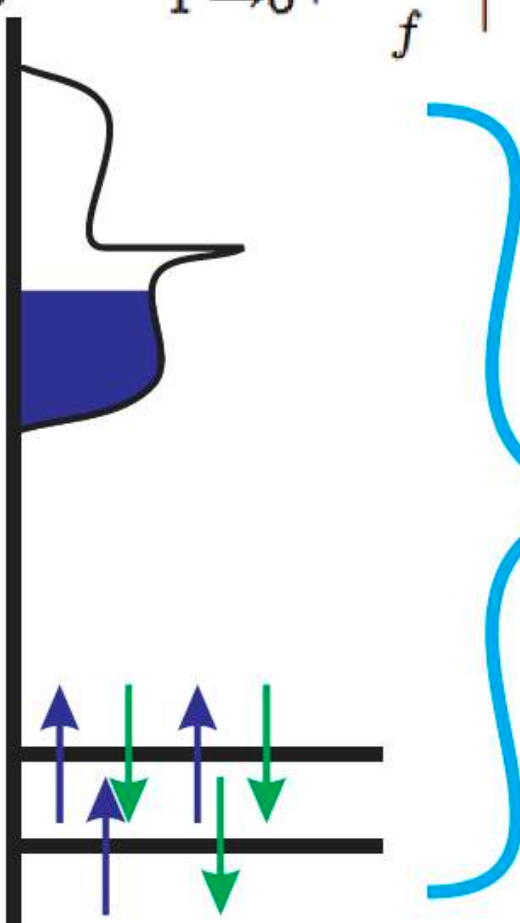
2007: 90 meV, 600 cps

2002: 300 meV, 6 cps

1999: 1500 meV, 1cps



RIXS process

$$\frac{\delta^2 \sigma}{\delta \Omega \delta \omega} \propto \lim_{\Gamma \rightarrow 0^+} \sum_f \left| \langle f | T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} | i \rangle \right|^2 \delta(\omega_i - \omega_o + E_i - E_f)$$


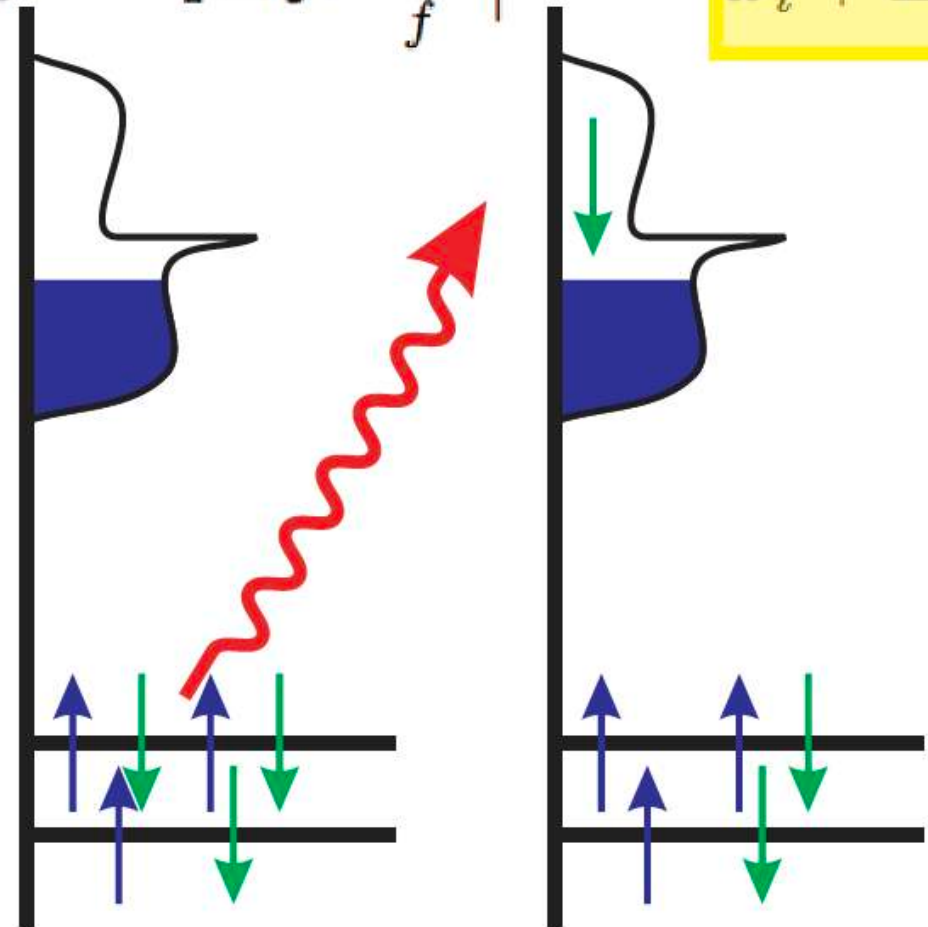
The diagram illustrates the RIXS process. On the left, a vertical black line represents the energy levels of the system. A blue shaded region indicates the initial state. Below this, two horizontal black lines represent the final state. Green arrows point upwards from the lower level to the upper level, and blue arrows point downwards from the upper level to the lower level. A large blue bracket connects the initial state to the final state, indicating the transition. A blue circle highlights the term $|i\rangle$ in the equation, which is connected to the initial state by a blue line.

RIXS process

$$\frac{\delta^2 \sigma}{\delta \Omega \delta \omega} \propto \lim_{\Gamma \rightarrow 0^+} \sum_f \left| \langle f | T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} | i \rangle \right|^2 \delta(\omega_i - \omega_o + E_i - E_f)$$

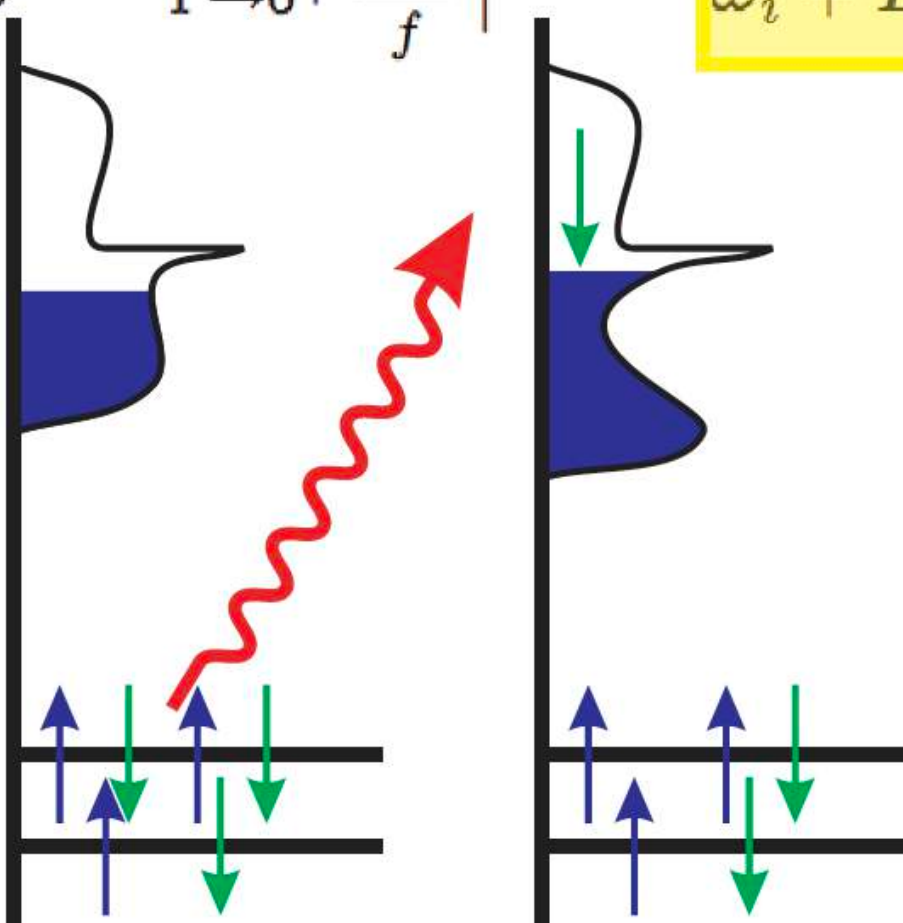
The diagram illustrates the RIXS process. It shows two electronic systems, each represented by a vertical line and a blue shaded region. The left system has two horizontal lines with blue arrows pointing up and green arrows pointing down. The right system has two horizontal lines with blue arrows pointing up and green arrows pointing down. A red wavy arrow points from the right system to the left system, representing the incoming photon. A red curved arrow points from the T_{ϵ_i} term in the equation to the right system, indicating the interaction of the incoming photon with the system.

RIXS process

$$\frac{\delta^2 \sigma}{\delta \Omega \delta \omega} \propto \lim_{\Gamma \rightarrow 0^+} \sum_f \left| \langle f | T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} | i \rangle \right|^2 \delta(\omega_i - \omega_o + E_i - E_f)$$


The diagram illustrates the RIXS process. It shows two energy levels, each represented by a vertical line with a blue shaded region. The left level has a red wavy arrow pointing upwards, while the right level has a green arrow pointing downwards. Below each level are horizontal lines with blue and green arrows, representing spin states. The equation above the diagram describes the RIXS process, showing the dependence of the second-order differential cross-section on the initial and final states and the energy transfer.

RIXS process

$$\frac{\delta^2 \sigma}{\delta \Omega \delta \omega} \propto \lim_{\Gamma \rightarrow 0^+} \sum_f \left| \langle f | T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} | i \rangle \right|^2 \delta(\omega_i - \omega_o + E_i - E_f)$$


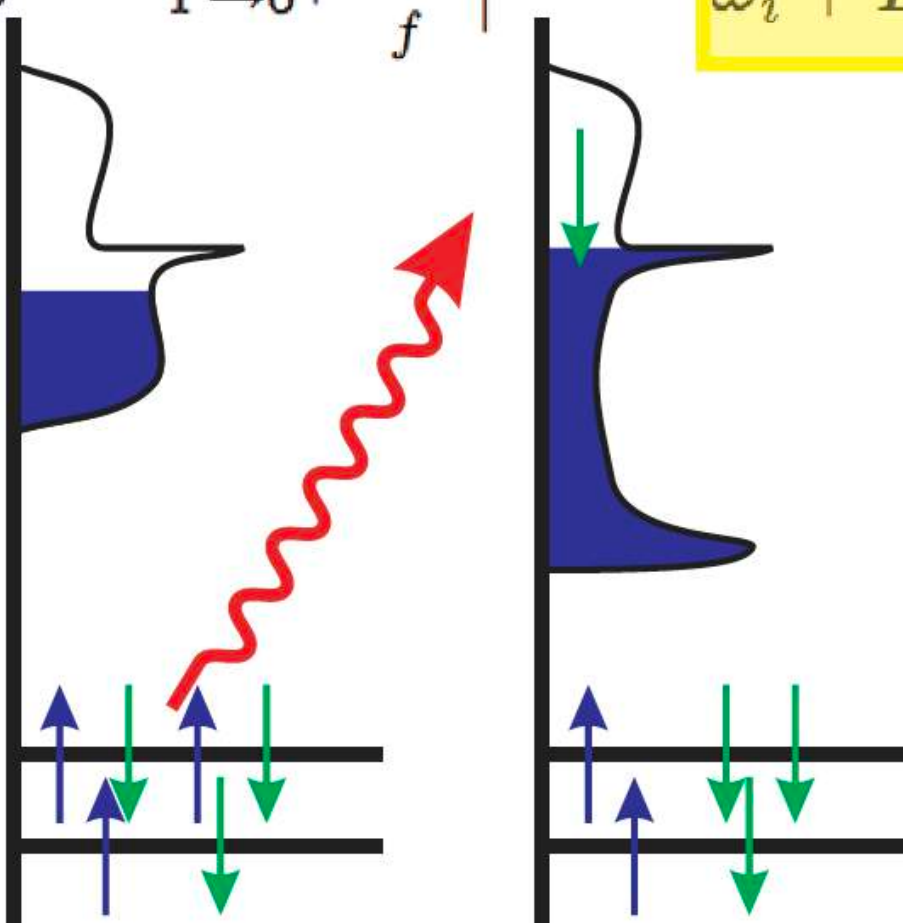
The diagram illustrates the RIXS process. It shows two energy levels, each represented by a vertical black line. The left energy level has a blue shaded region at the top and a red wavy arrow pointing upwards from a lower level. The right energy level has a blue shaded region at the top and a green arrow pointing downwards from a lower level. Below each energy level, there are two horizontal black lines representing electronic states. On the left, there are four blue arrows pointing up and four green arrows pointing down. On the right, there are four blue arrows pointing up and four green arrows pointing down. The red wavy arrow represents the scattering process, and the green arrow represents the emission of a photon.

RIXS process

$$\frac{\delta^2 \sigma}{\delta \Omega \delta \omega} \propto \lim_{\Gamma \rightarrow 0^+} \sum_f \left| \langle f | T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} | i \rangle \right|^2 \delta(\omega_i - \omega_o + E_i - E_f)$$

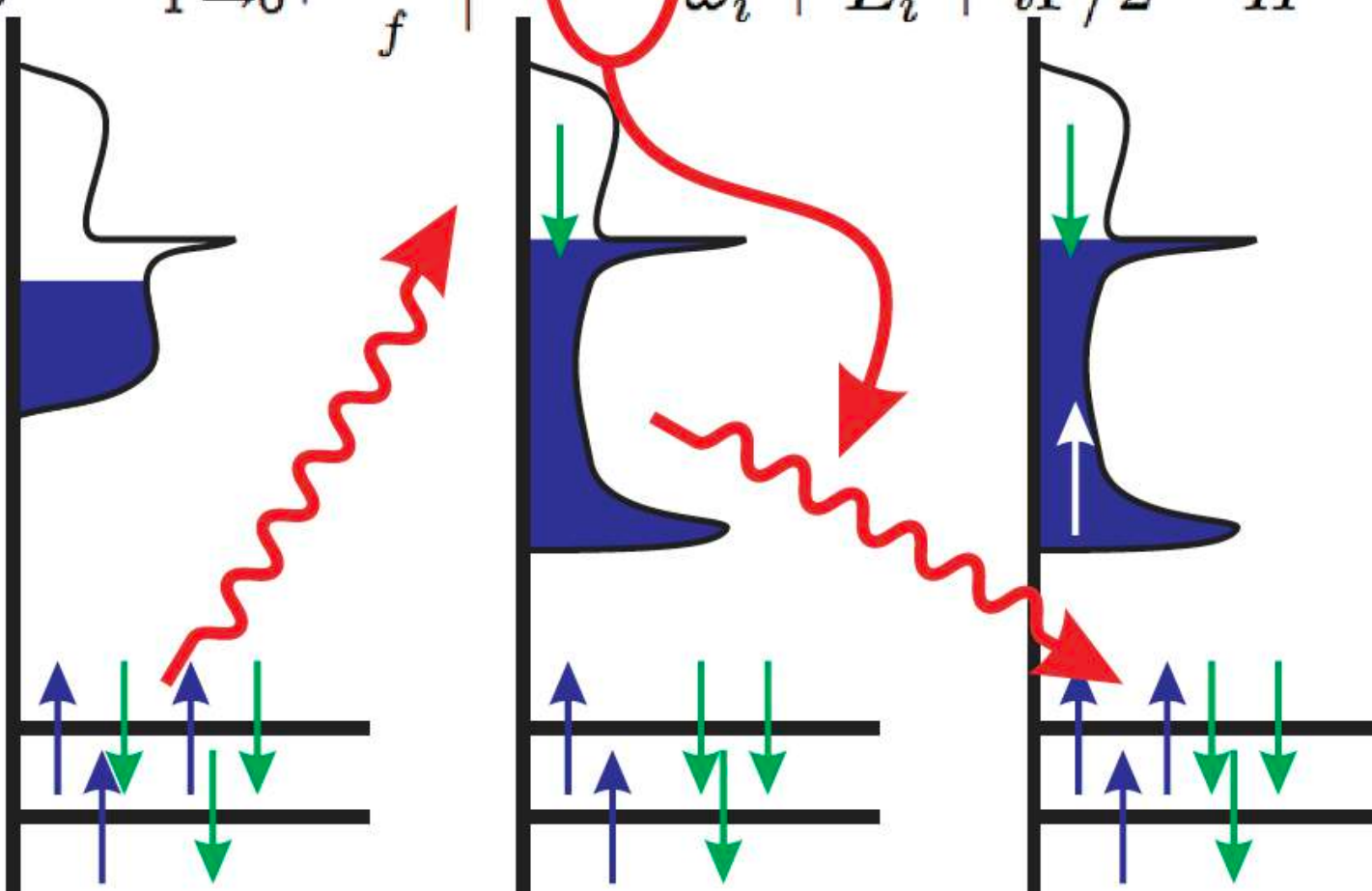
The diagram illustrates the RIXS process. On the left, the initial state shows two occupied orbitals (blue arrows) and two empty orbitals (green arrows). A red wavy arrow indicates an incoming photon. On the right, the final state shows one occupied orbital (blue arrow) and one empty orbital (green arrow). A blue shaded region represents the energy distribution of the scattered photon.

RIXS process

$$\frac{\delta^2 \sigma}{\delta \Omega \delta \omega} \propto \lim_{\Gamma \rightarrow 0^+} \sum_f \left| \langle f | T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} | i \rangle \right|^2 \delta(\omega_i - \omega_o + E_i - E_f)$$


The diagram illustrates the RIXS process. It shows two energy levels, each represented by a vertical black line. The left energy level has a blue shaded region at the top, representing an excited state. The right energy level has a blue shaded region at the bottom, representing a ground state. A red wavy arrow points from the left energy level to the right energy level, indicating the transfer of energy. Below each energy level, there are horizontal black lines representing electronic states. On the left, there are four blue arrows pointing up and four green arrows pointing down. On the right, there are four blue arrows pointing up and four green arrows pointing down. The red wavy arrow originates from the bottom of the left energy level and points towards the top of the right energy level.

RIXS process

$$\frac{\delta^2 \sigma}{\delta \Omega \delta \omega} \propto \lim_{\Gamma \rightarrow 0^+} \sum_f \left| \langle f | T_{\varepsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\varepsilon_i} | i \rangle \right|^2 \delta(\omega_i - \omega_o + E_i - E_f)$$


The diagram illustrates the RIXS process across three stages. Each stage shows a set of energy levels (black lines) with blue arrows indicating electron occupancy. A red wavy arrow represents the incident photon, and a green wavy arrow represents the scattered photon. The first stage shows the initial state. The second stage shows the intermediate state after the first transition. The third stage shows the final state after the second transition. A red circle highlights the term $T_{\varepsilon_o}^\dagger$ in the equation, which corresponds to the first transition in the diagram.

RIXS process

$$\frac{\delta^2 \sigma}{\delta \Omega \delta \omega} \propto \lim_{\Gamma \rightarrow 0^+} \sum_f \left| \langle f | T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} | i \rangle \right|^2 \delta(\omega_i - \omega_o + E_i - E_f)$$

The diagram illustrates the RIXS process through four stages, each showing energy levels (black lines) and electron occupancy (blue shaded regions). Blue arrows represent spin-up electrons, and green arrows represent spin-down electrons. A red wavy arrow indicates the incident X-ray photon, and a red arrow indicates the scattered X-ray photon.

- Stage 1:** Initial state with two occupied levels. An incident photon (red wavy arrow) is absorbed, promoting an electron to a higher energy level.
- Stage 2:** Intermediate state where an electron has been promoted to a higher energy level. A scattered photon (red arrow) is emitted, and an electron transitions back to a lower level.
- Stage 3:** Final state where the electron has transitioned to a different level, creating a spin excitation (indicated by a white arrow pointing up).
- Stage 4:** Final state showing the resulting spin excitation (white arrow pointing up) and the scattered photon.

RIXS process

$$\frac{\delta^2 \sigma}{\delta \Omega \delta \omega} \propto \lim_{\Gamma \rightarrow 0^+} \sum_f \left| \langle f | T_{\epsilon_o}^\dagger \frac{R_{\omega_i}^{\epsilon_i \epsilon_o}}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} | i \rangle \right|^2 \delta(\omega_i - \omega_o + E_i - E_f)$$

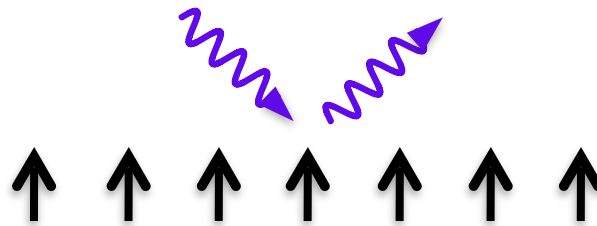
$$R_{\omega_i}^{\epsilon_i \epsilon_o} = T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i}$$

antisymmetric $(\epsilon_\alpha \epsilon'_\beta - \epsilon_\beta \epsilon'_\alpha)$ dipole

symmetric $(\epsilon_\alpha \epsilon'_\beta + \epsilon_\beta \epsilon'_\alpha)$ quadrupole

courtesy of M. Haverkort

Spin waves (or magnons)



Spin waves (or magnons)



Spin waves (or magnons)



Spin waves (or magnons)



Spin waves (or magnons)



$(r, t) \rightarrow (q, \omega)$

dynamic structure factor $S(q, \omega)$

RIXS operators for t_{2g} orbital systems

unquenched orbital angular momentum

“fast collision approximation”

$$R_{\omega_i}^{\epsilon_i \epsilon_o} = T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} \quad \rightarrow \quad R \propto D^\dagger D = \frac{1}{3}(R_Q + iR_M)$$

c number

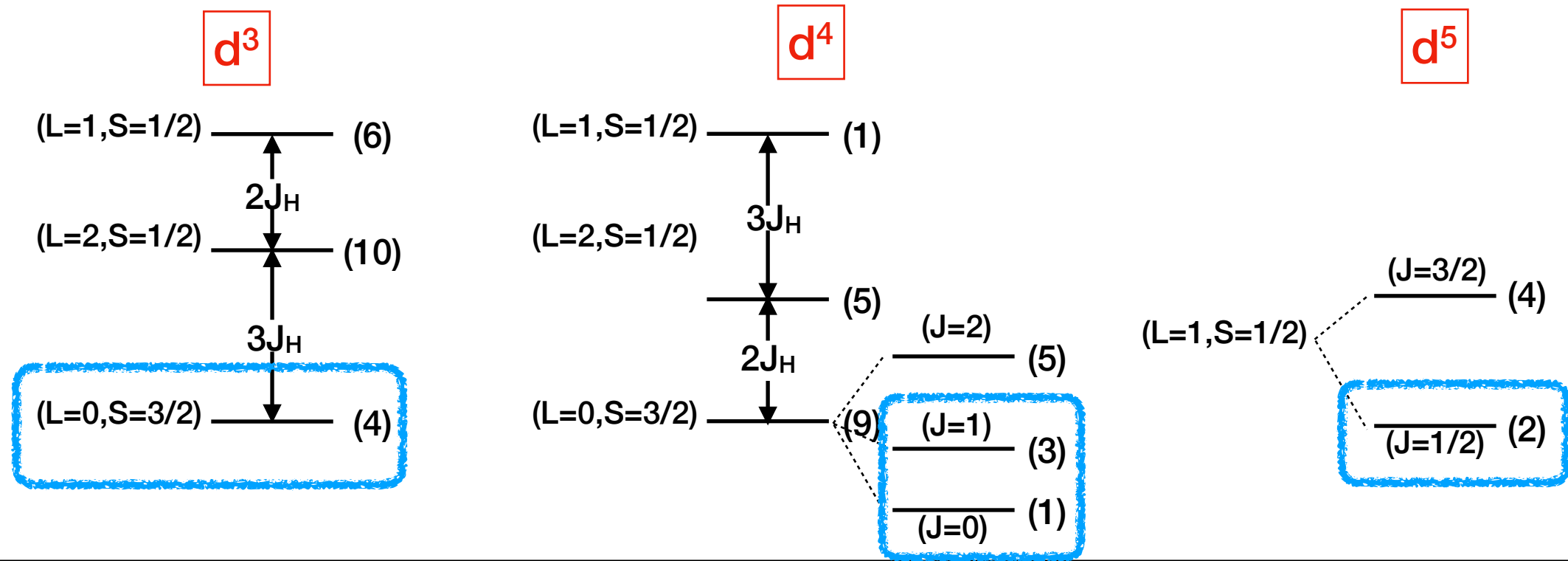
$$R_Q = \sum_{\alpha} \epsilon_{\alpha} \epsilon'_{\alpha} Q_{\alpha\alpha} - \frac{1}{2} \sum_{\alpha > \beta} (\epsilon_{\alpha} \epsilon'_{\beta} + \epsilon_{\beta} \epsilon'_{\alpha}) Q_{\alpha\beta}$$

$$R_M = \frac{1}{2}(\boldsymbol{\epsilon} \times \boldsymbol{\epsilon}') \cdot \mathbf{N}.$$

Q_{zz} (quadrupole)	L_3 edge	L_2 edge
$d^1, (-1)d^5$	$-2L_z^2 + 2L_z S_z$	$-L_z^2 - 2L_z S_z$
$d^2, (-1)d^4$	$2L_z^2 + L_z S_z$	$L_z^2 - L_z S_z$
Q_{xy} (quadrupole)	L_3 edge	L_2 edge
$d^1, (-1)d^5$	$-2L_x L_y - 2L_y L_x + 2L_x S_y + 2L_y S_x$	$-L_x L_y - L_y L_x - 2L_x S_y - 2L_y S_x$
$d^2, (-1)d^4$	$2L_x L_y + 2L_y L_x + L_x S_y + L_y S_x$	$L_x L_y + L_y L_x - L_x S_y - L_y S_x$
N_z (magnetic)	L_3 edge	L_2 edge
d^1, d^5	$2L_z - 4S_z + 8L_z^2 S_z - 2L_z(\mathbf{L} \cdot \mathbf{S}) - 2(\mathbf{L} \cdot \mathbf{S})L_z$	$L_z + 4S_z - 8L_z^2 S_z + 2L_z(\mathbf{L} \cdot \mathbf{S}) + 2(\mathbf{L} \cdot \mathbf{S})L_z$
d^2, d^4	$2L_z - 4L_z^2 S_z + L_z(\mathbf{L} \cdot \mathbf{S}) + (\mathbf{L} \cdot \mathbf{S})L_z$	$L_z + 4L_z^2 S_z - L_z(\mathbf{L} \cdot \mathbf{S}) - (\mathbf{L} \cdot \mathbf{S})L_z$
d^3	$(4/3)S_z$	$-(4/3)S_z$

RIXS operators for t_{2g} orbital systems

Energy levels in cubic symmetry



Q_{zz} (quadrupole)	L_3 edge	L_2 edge
$d^1, (-1)d^5$	$-2L_z^2 + 2L_z S_z$	$-L_z^2 - 2L_z S_z$
$d^2, (-1)d^4$	$2L_z^2 + L_z S_z$	$L_z^2 - L_z S_z$
Q_{xy} (quadrupole)	L_3 edge	L_2 edge
$d^1, (-1)d^5$	$-2L_x L_y - 2L_y L_x + 2L_x S_y + 2L_y S_x$	$-L_x L_y - L_y L_x - 2L_x S_y - 2L_y S_x$
$d^2, (-1)d^4$	$2L_x L_y + 2L_y L_x + L_x S_y + L_y S_x$	$L_x L_y + L_y L_x - L_x S_y - L_y S_x$
N_z (magnetic)	L_3 edge	L_2 edge
d^1, d^5	$2L_z - 4S_z + 8L_z^2 S_z - 2L_z(\mathbf{L} \cdot \mathbf{S}) - 2(\mathbf{L} \cdot \mathbf{S})L_z$	$L_z + 4S_z - 8L_z^2 S_z + 2L_z(\mathbf{L} \cdot \mathbf{S}) + 2(\mathbf{L} \cdot \mathbf{S})L_z$
d^2, d^4	$2L_z - 4L_z^2 S_z + L_z(\mathbf{L} \cdot \mathbf{S}) + (\mathbf{L} \cdot \mathbf{S})L_z$	$L_z + 4L_z^2 S_z - L_z(\mathbf{L} \cdot \mathbf{S}) - (\mathbf{L} \cdot \mathbf{S})L_z$
d^3	$(4/3)S_z$	$-(4/3)S_z$

RIXS measures (pseudo) spin-spin correlations

“Spin-Orbit Mott insulator”

Iridates

$$R_M = \frac{1}{2}(\epsilon \times \epsilon') \cdot \mathbf{N}$$

$$N_\alpha = f_\alpha \tilde{S}_\alpha$$

L_3

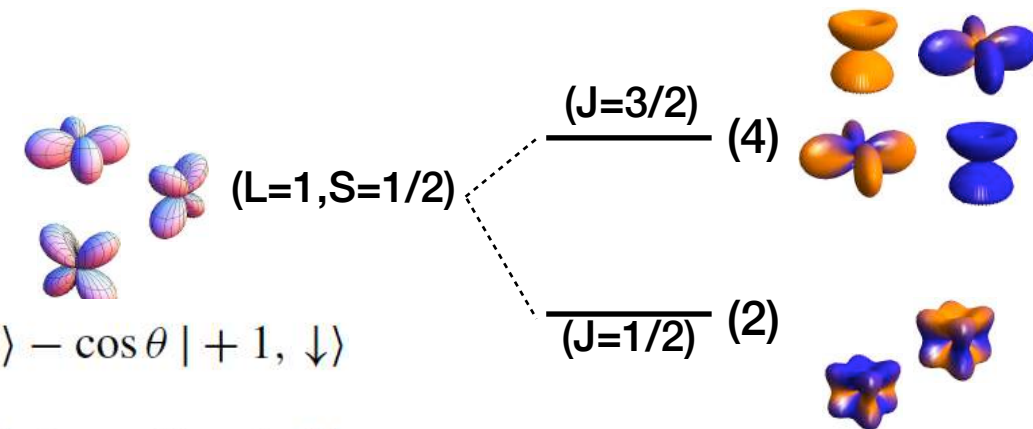
$$f_{x/y} = -3\sqrt{2} \sin 2\theta,$$

$$f_z = -2\sqrt{2}(\sin 2\theta + \sqrt{2} \cos 2\theta)$$

L_2

$$f_{x/y} = 0,$$

$$f_z = -3 + \cos 2\theta + 2\sqrt{2} \sin 2\theta$$

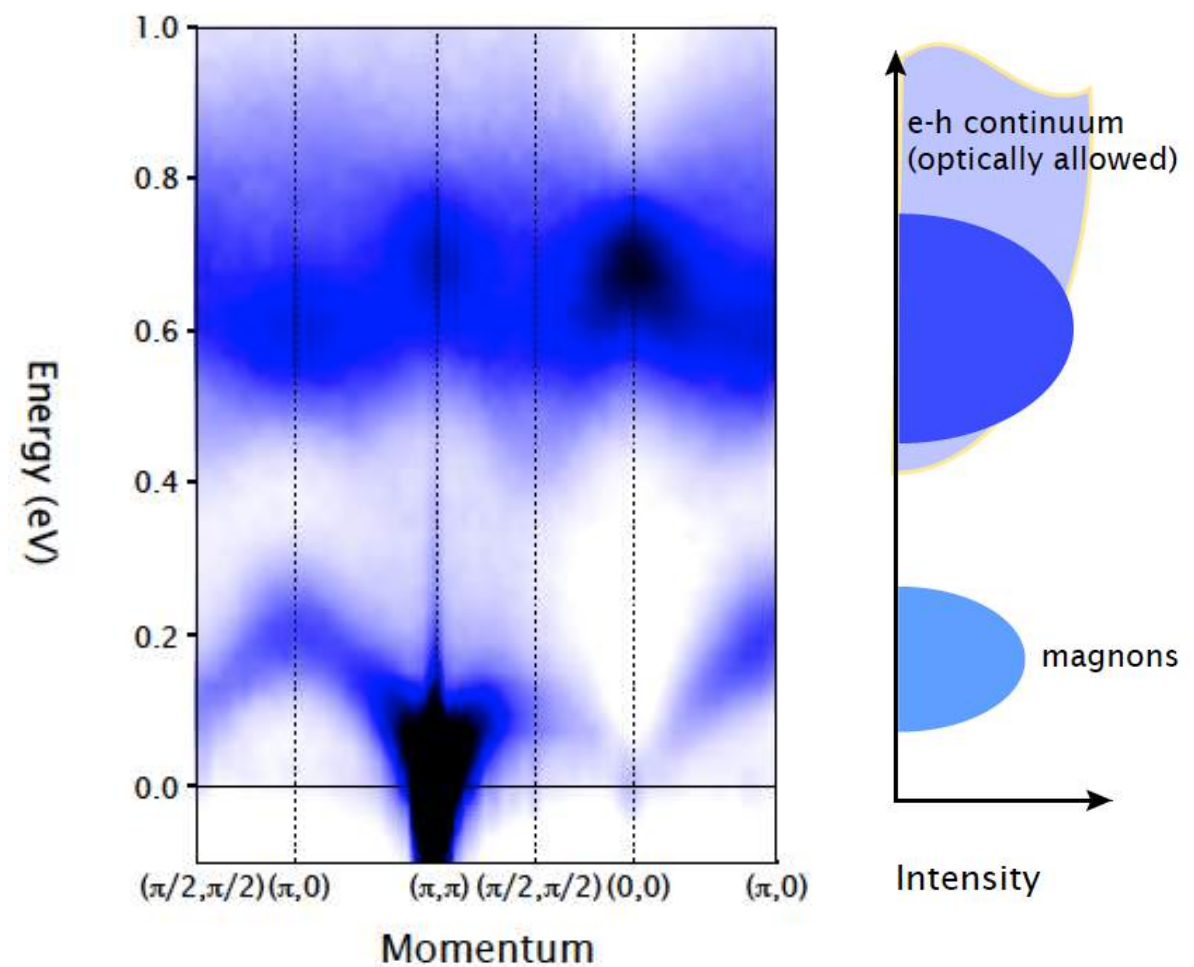
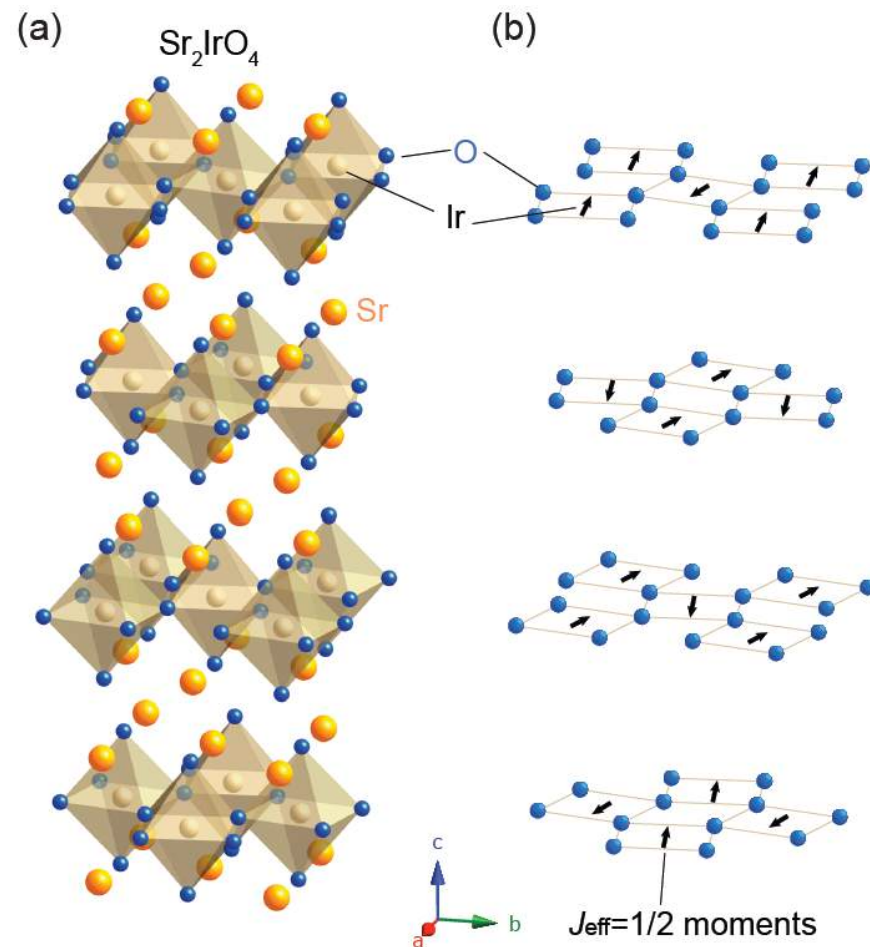


$$|\tilde{\uparrow}\rangle = +\sin\theta |0, \uparrow\rangle - \cos\theta | +1, \downarrow\rangle$$

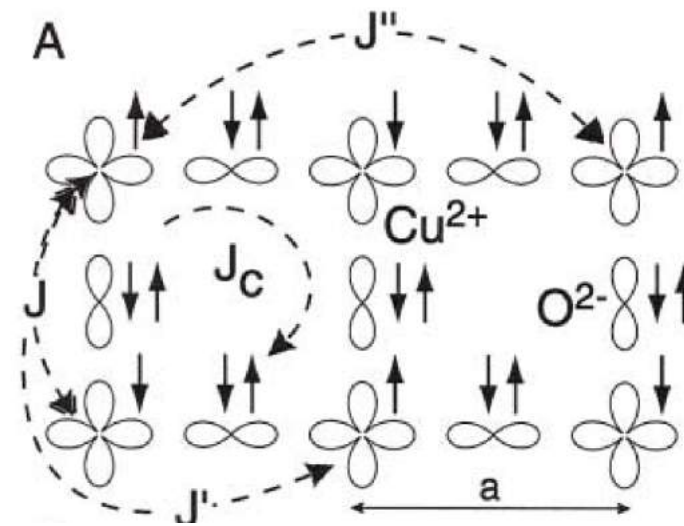
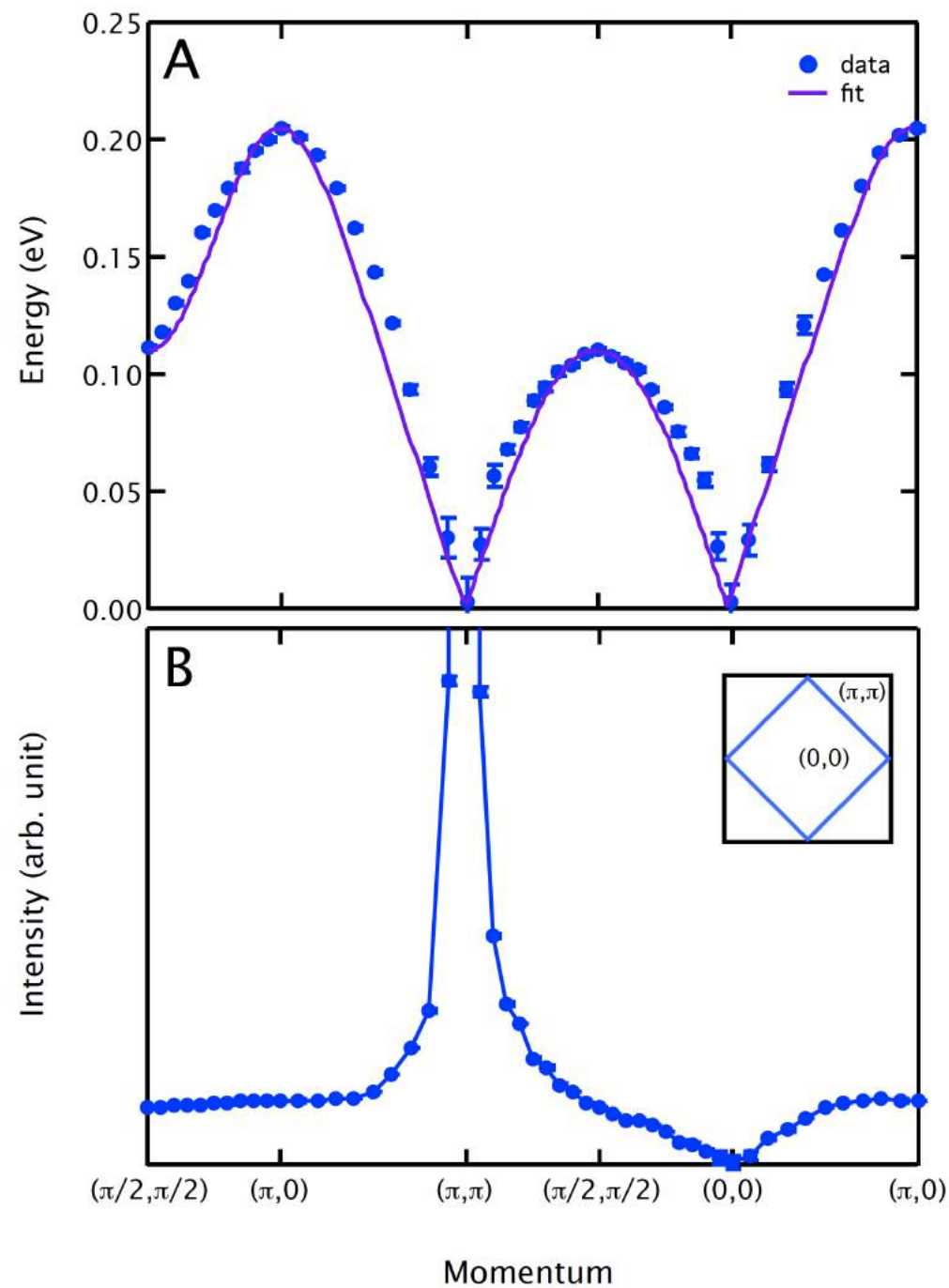
$$|\tilde{\downarrow}\rangle = -\sin\theta |0, \downarrow\rangle + \cos\theta | -1, \uparrow\rangle$$

Q_{zz} (quadrupole)	L_3 edge	L_2 edge
$d^1, (-1)d^5$	$-2L_z^2 + 2L_z S_z$	$-L_z^2 - 2L_z S_z$
$d^2, (-1)d^4$	$2L_z^2 + L_z S_z$	$L_z^2 - L_z S_z$
Q_{xy} (quadrupole)	L_3 edge	L_2 edge
$d^1, (-1)d^5$	$-2L_x L_y - 2L_y L_x + 2L_x S_y + 2L_y S_x$	$-L_x L_y - L_y L_x - 2L_x S_y - 2L_y S_x$
$d^2, (-1)d^4$	$2L_x L_y + 2L_y L_x + L_x S_y + L_y S_x$	$L_x L_y + L_y L_x - L_x S_y - L_y S_x$
N_z (magnetic)	L_3 edge	L_2 edge
d^1, d^5	$2L_z - 4S_z + 8L_z^2 S_z - 2L_z(\mathbf{L} \cdot \mathbf{S}) - 2(\mathbf{L} \cdot \mathbf{S})L_z$	$L_z + 4S_z - 8L_z^2 S_z + 2L_z(\mathbf{L} \cdot \mathbf{S}) + 2(\mathbf{L} \cdot \mathbf{S})L_z$
d^2, d^4	$2L_z - 4L_z^2 S_z + L_z(\mathbf{L} \cdot \mathbf{S}) + (\mathbf{L} \cdot \mathbf{S})L_z$	$L_z + 4L_z^2 S_z - L_z(\mathbf{L} \cdot \mathbf{S}) - (\mathbf{L} \cdot \mathbf{S})L_z$
d^3	$(4/3)S_z$	$-(4/3)S_z$

Magnetic excitations in Sr_2IrO_4



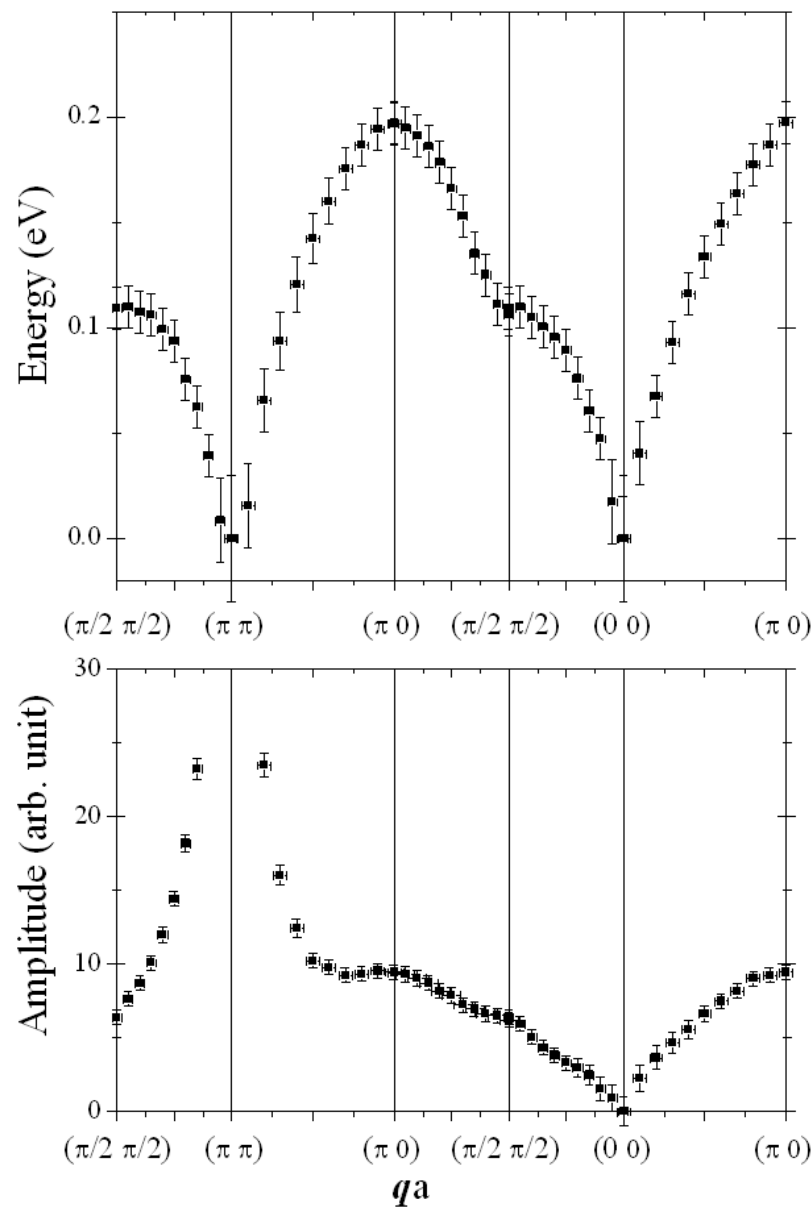
Fit to Heisenberg model



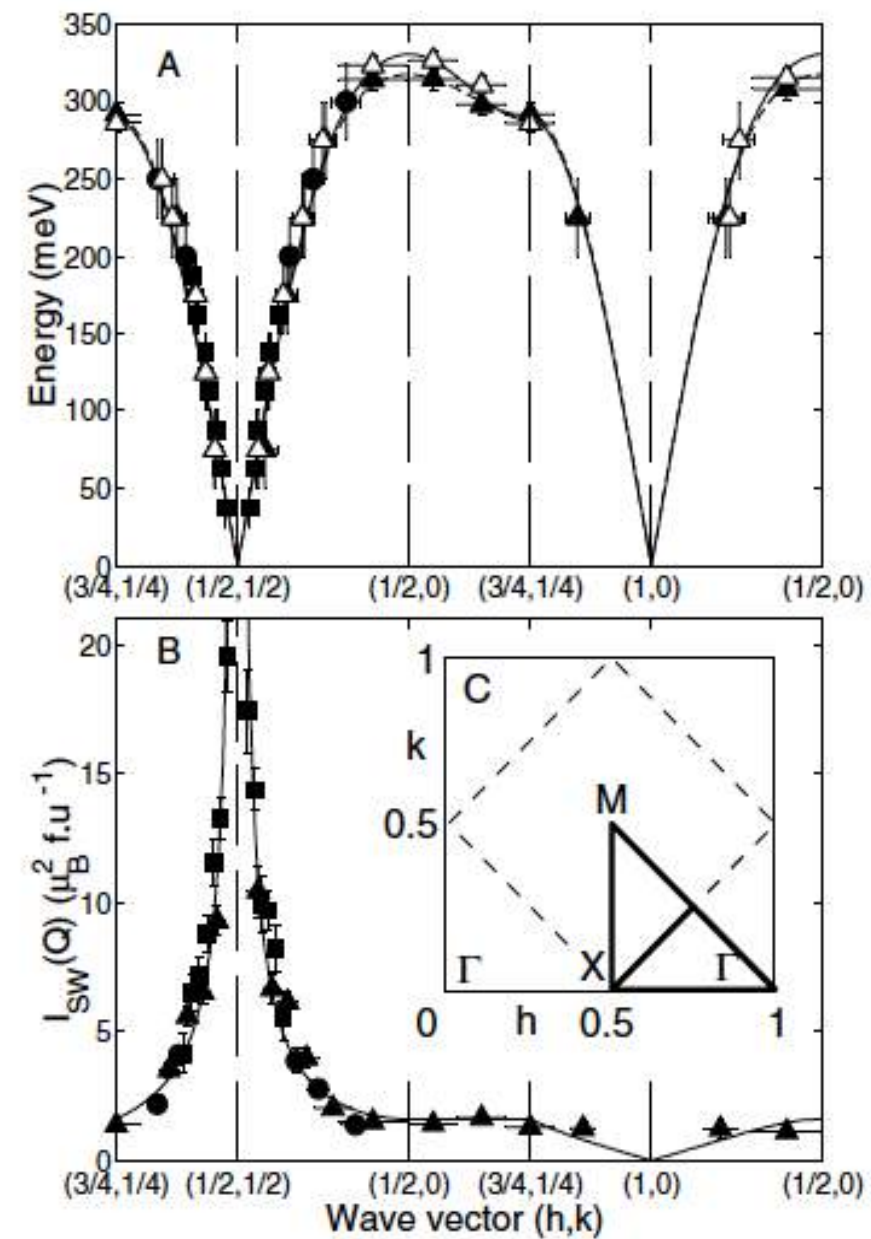
$$\begin{aligned} J &= 60 \\ J' &= -20 \\ J'' &= 15 \\ J_c &= 0 \text{ (suppressed)} \end{aligned}$$

Very similar spin dynamics

Our RIXS data on Sr_2IrO_4



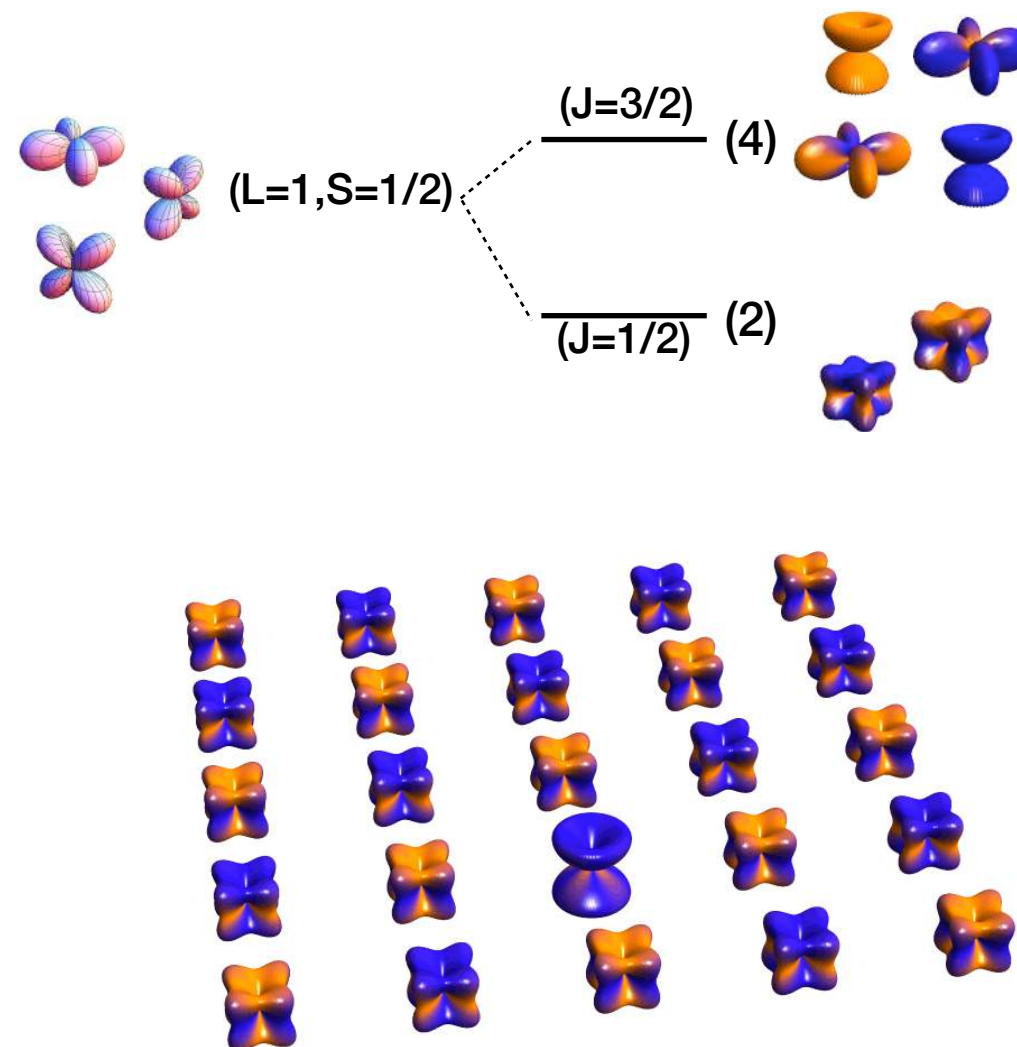
Neutron data on La_2CuO_4



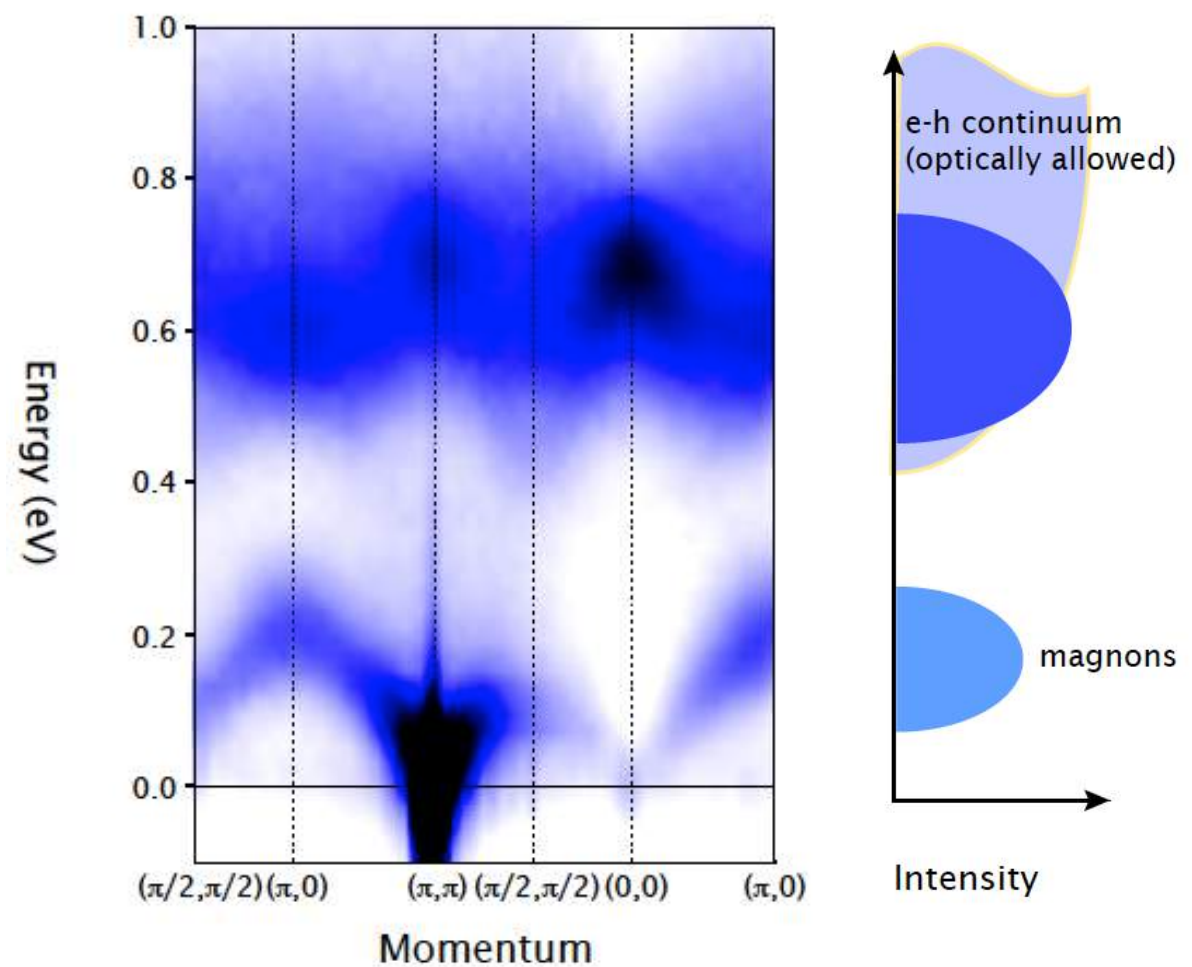
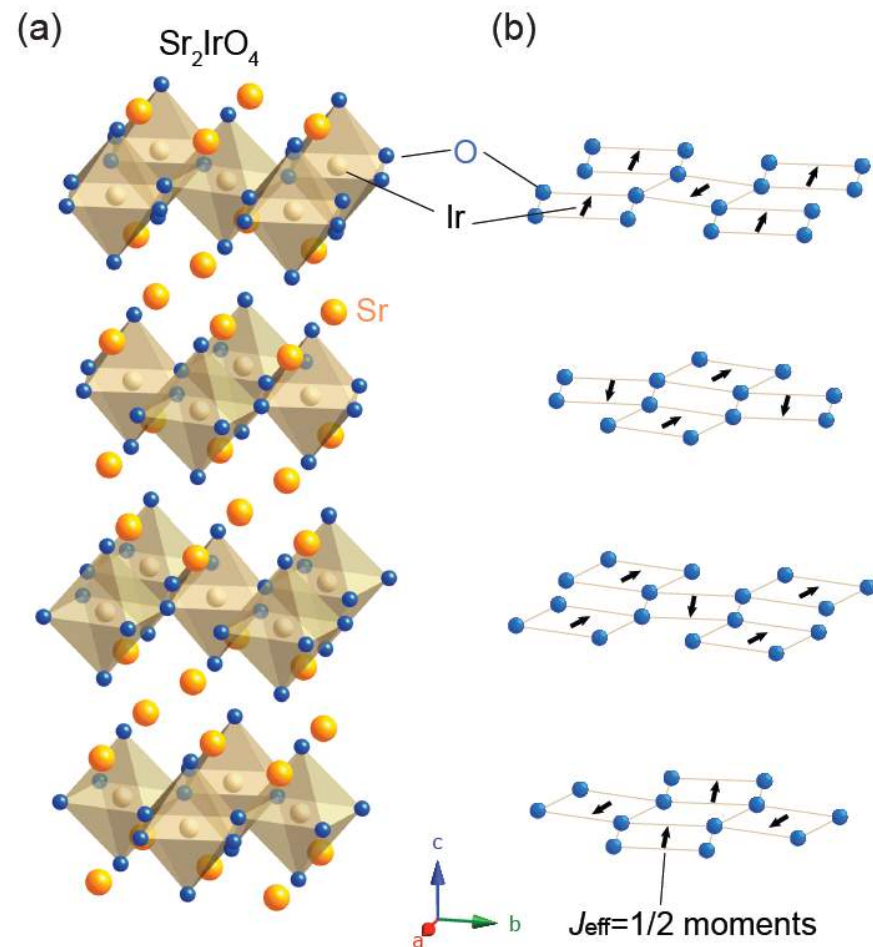
Coldea et al., PRL (2001)

Superconductivity in Sr_2IrO_4 upon carrier doping?

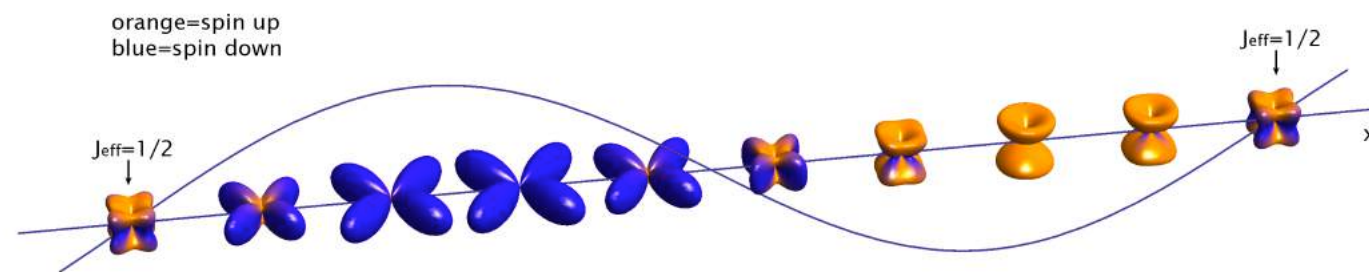
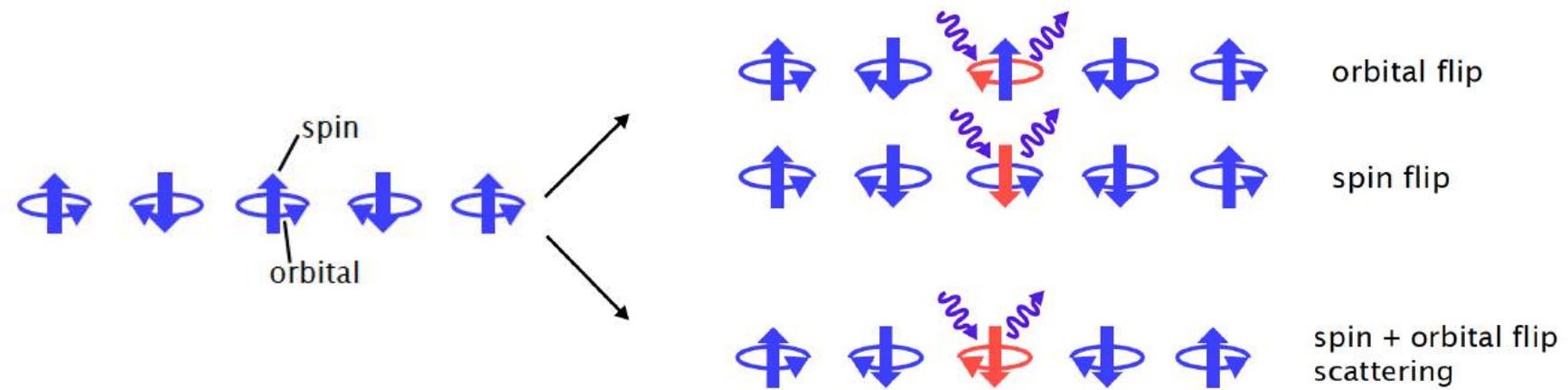
Spin-orbit exciton



Magnetic excitations in Sr_2IrO_4

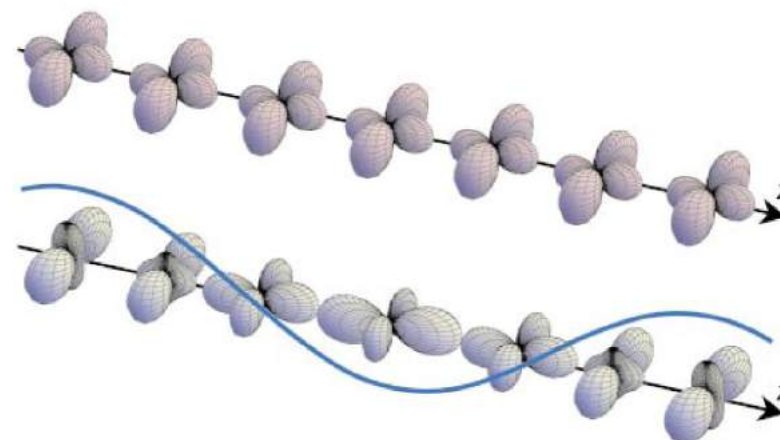


Spin-orbital waves

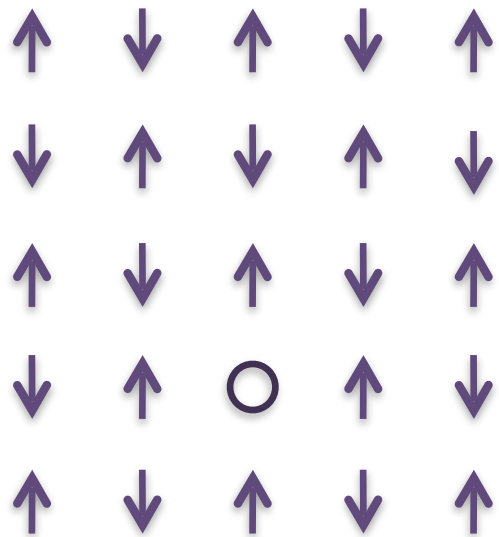


“Orbiton”

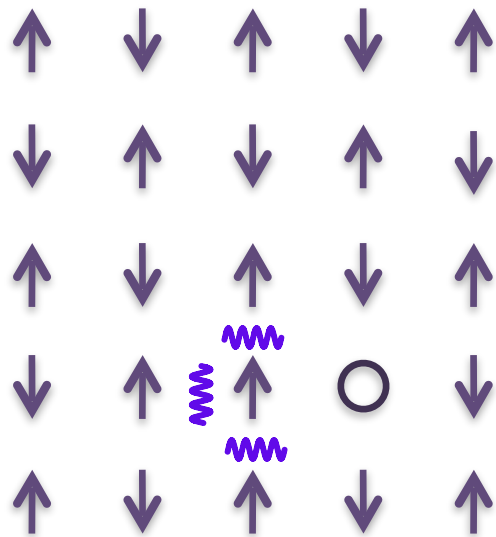
E. Saitoh et al., Nature (2001)



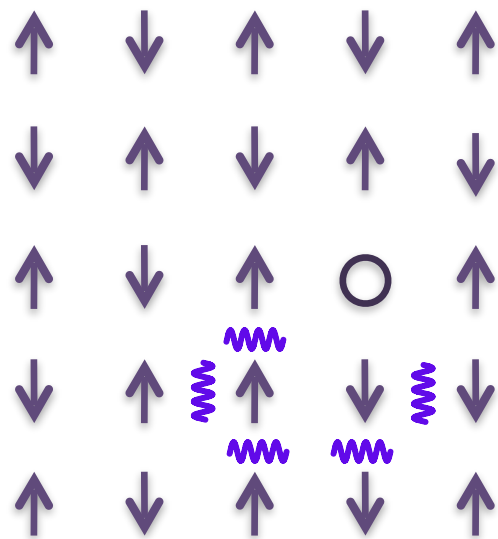
One-hole propagation



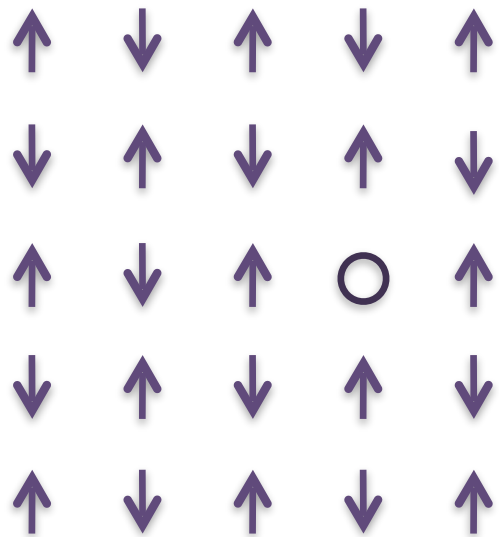
One-hole propagation



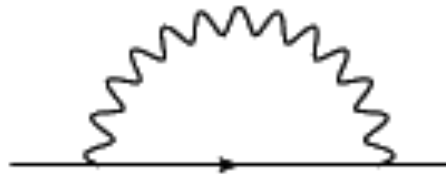
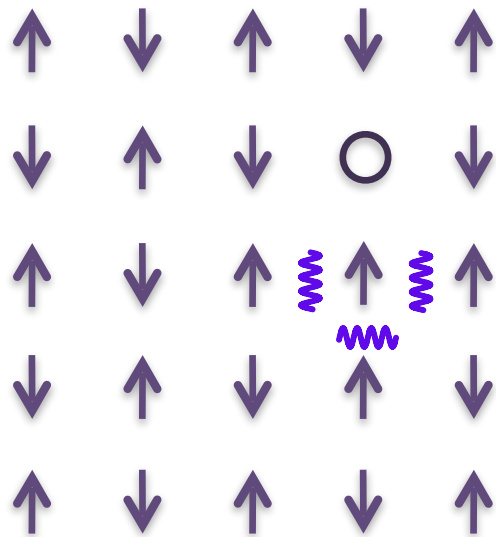
One-hole propagation



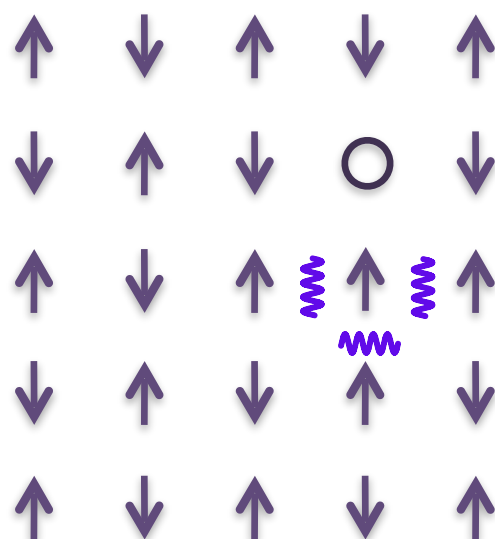
One-hole propagation



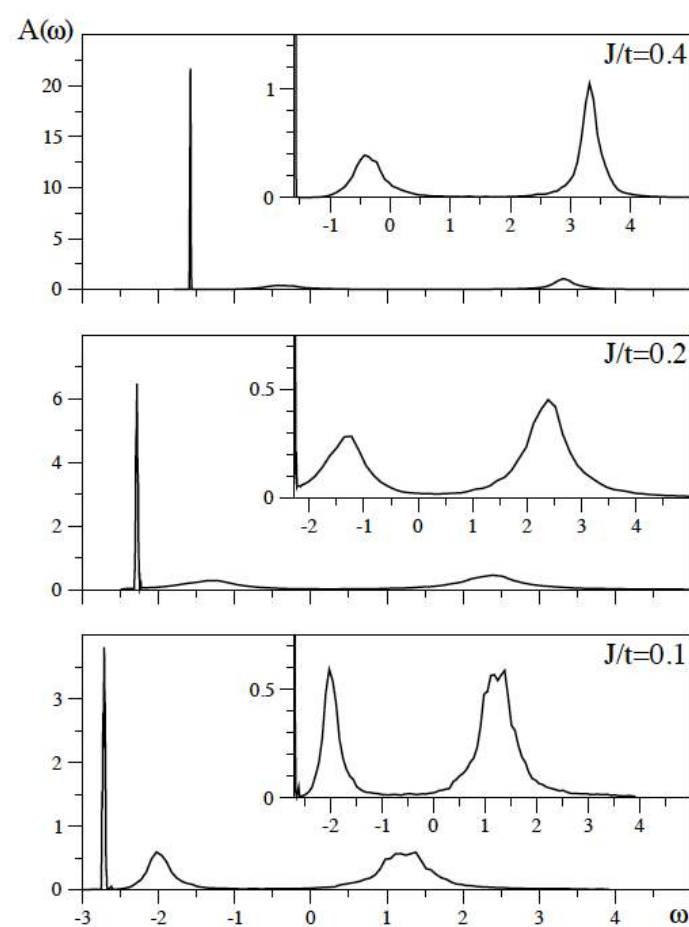
One-hole propagation



One-hole propagation

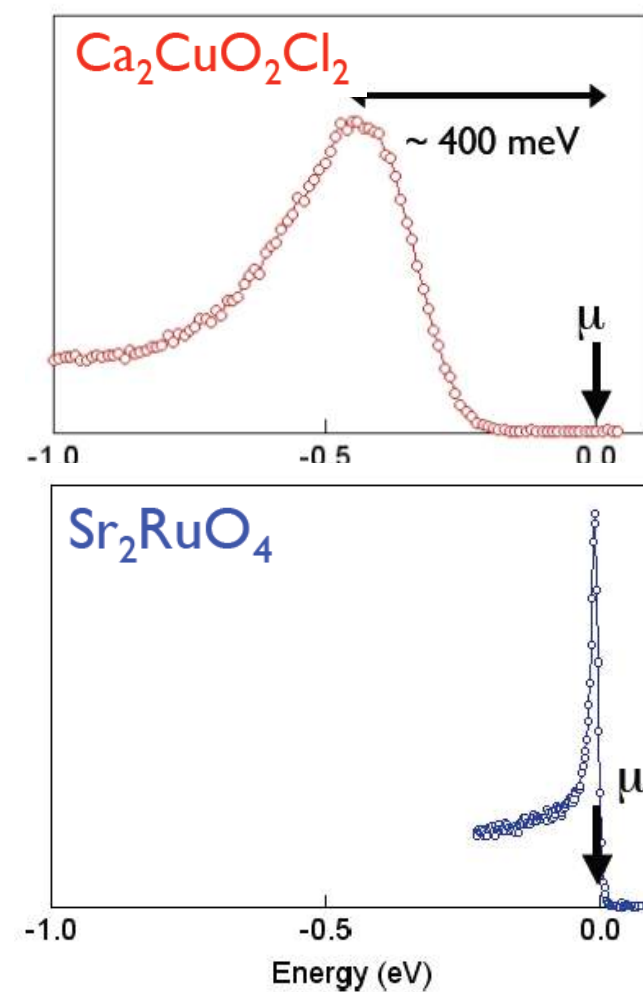


theory (t-J model)



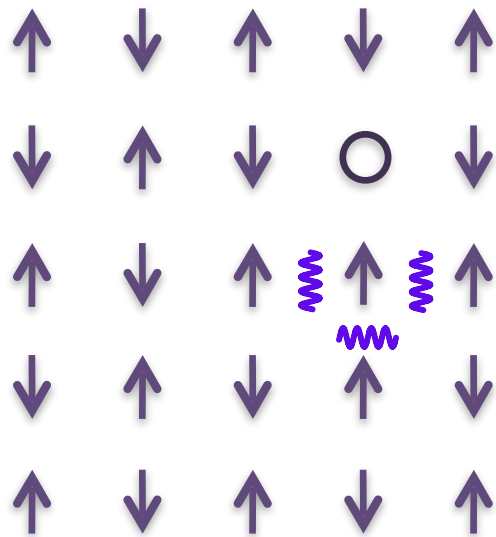
A. S. Mischenko et al. (2001)

ARPES

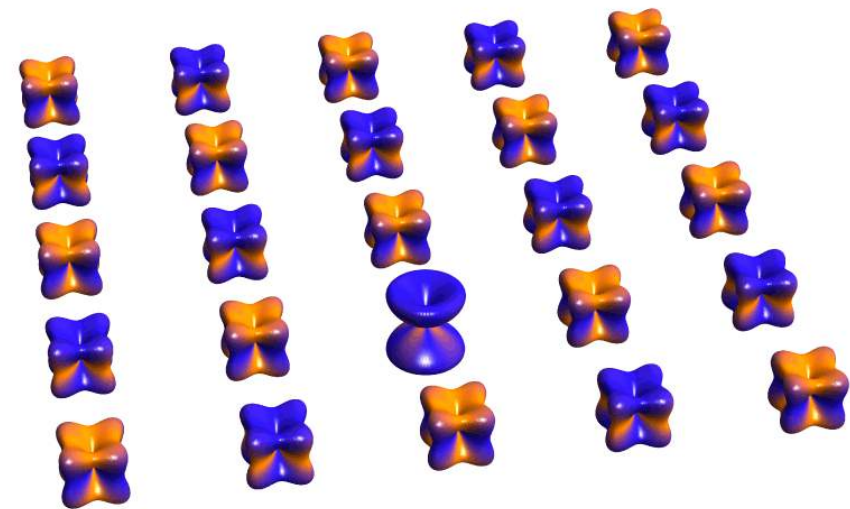


Shen group

One-hole propagation



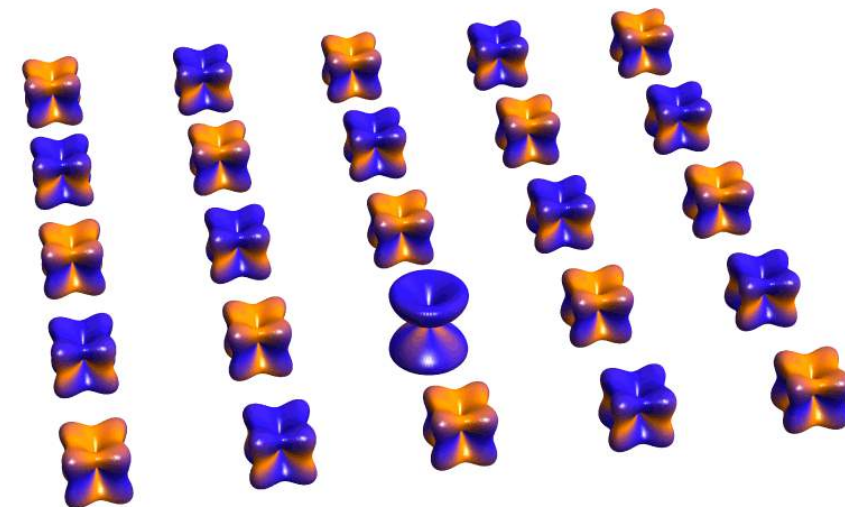
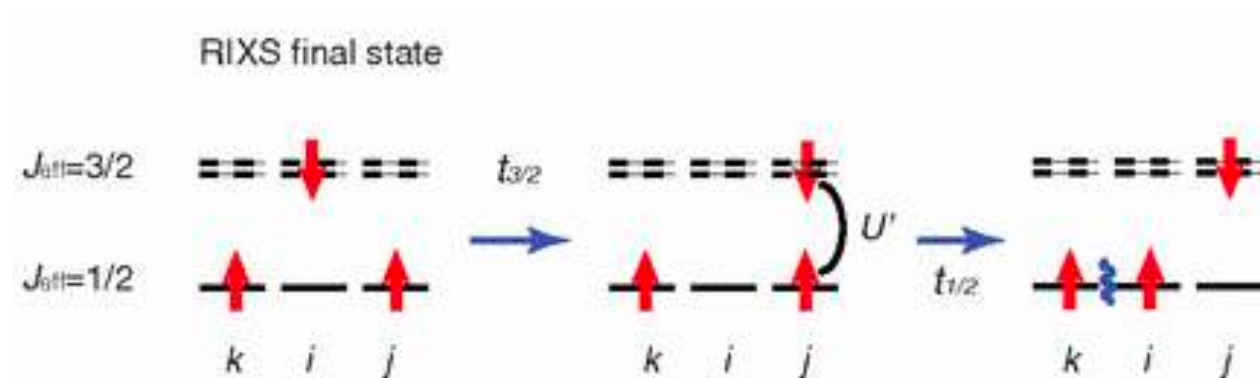
charge e^- , spin-1/2 fermion



charge neutral, hard-core boson

One-hole propagation

hopping via superexchange process



charge neutral, hard-core boson

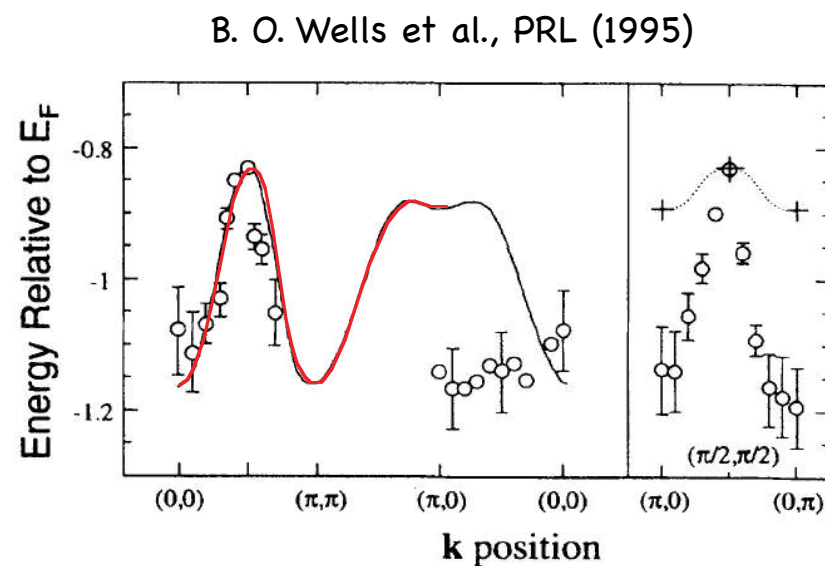
One-hole propagation

ARPES

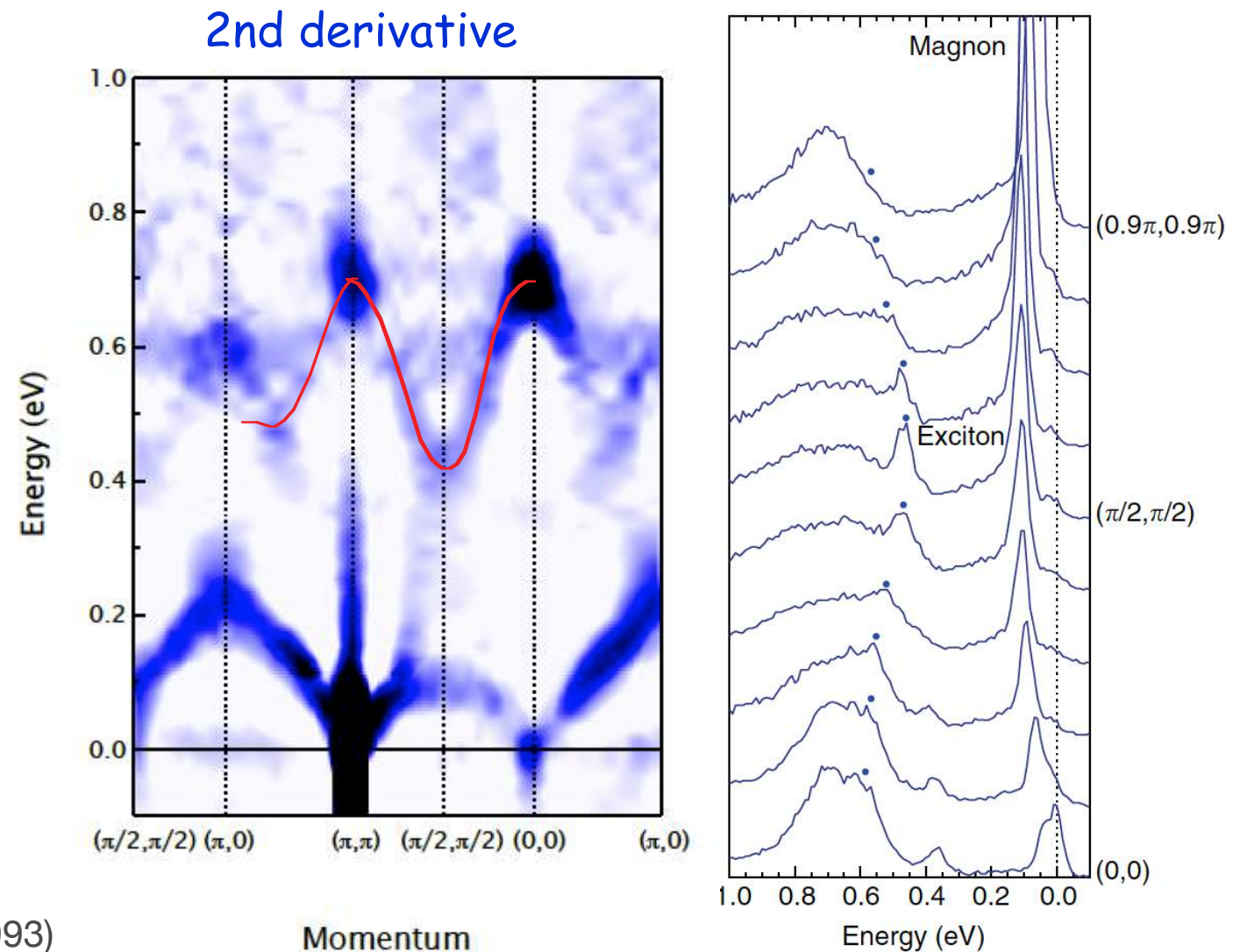
one-hole propagation in $\text{Sr}_2\text{CuO}_2\text{Cl}_2$

RIXS

one-exciton propagation in Sr_2IrO_4

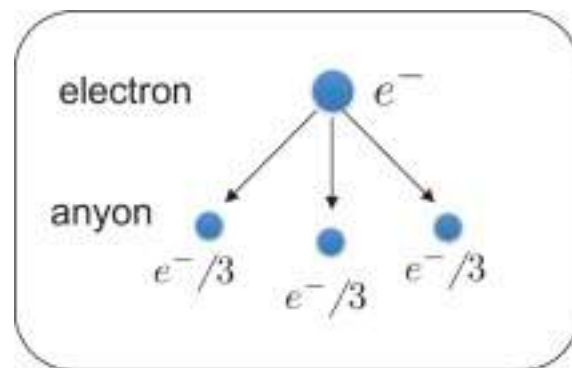


Schmitt-Rink, Varma, Ruckenstein, Phys. Rev. Lett. (1993)
 Lee, Nagaosa & Wen, Rev. Mod. Phys. (2006)

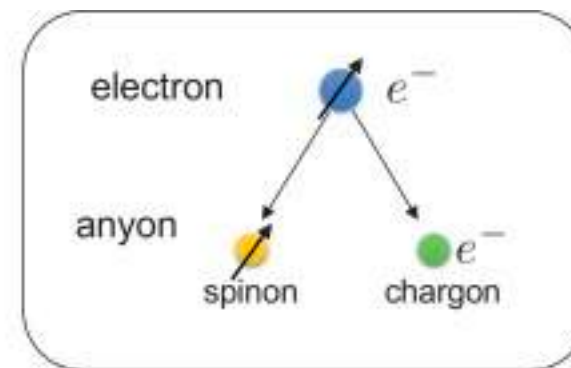


J. Kim & BJK et al. Nature Comm. (2014)

Fractional excitations

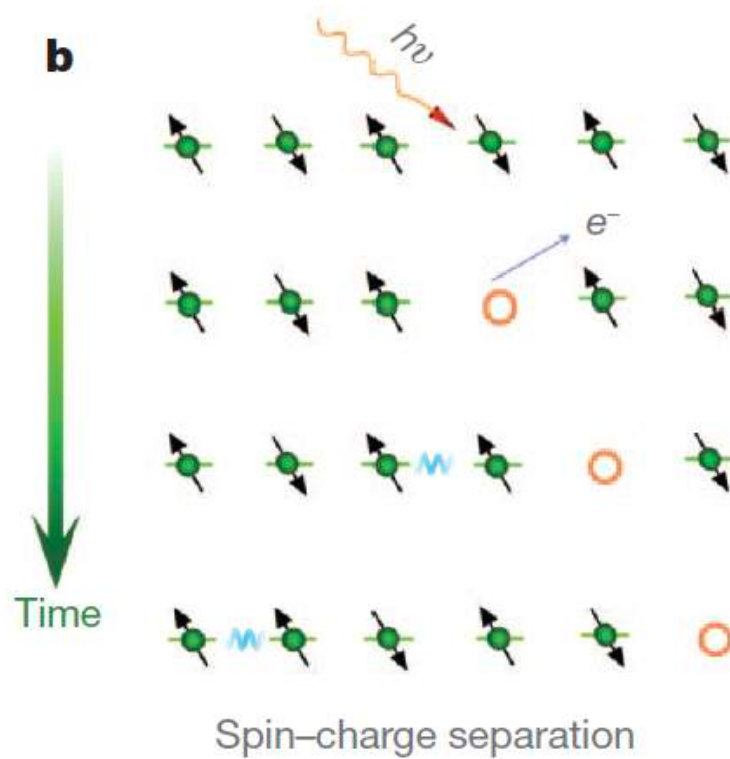


(a) $\nu = 1/3$ fractional quantum Hall

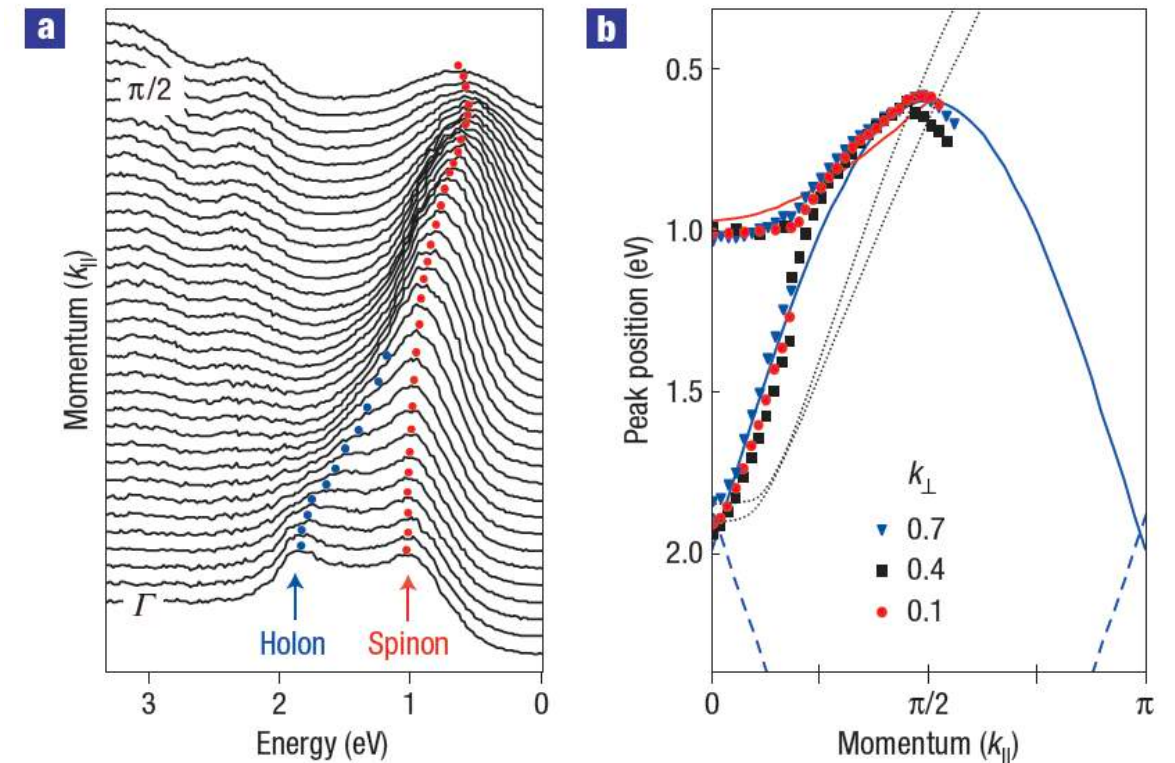


(b) spin-charge separation

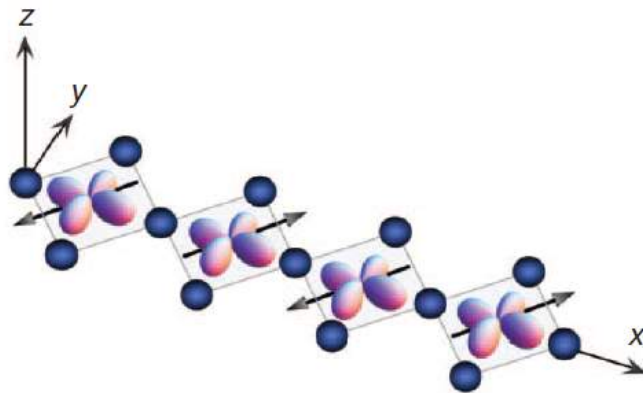
Spin-charge separation



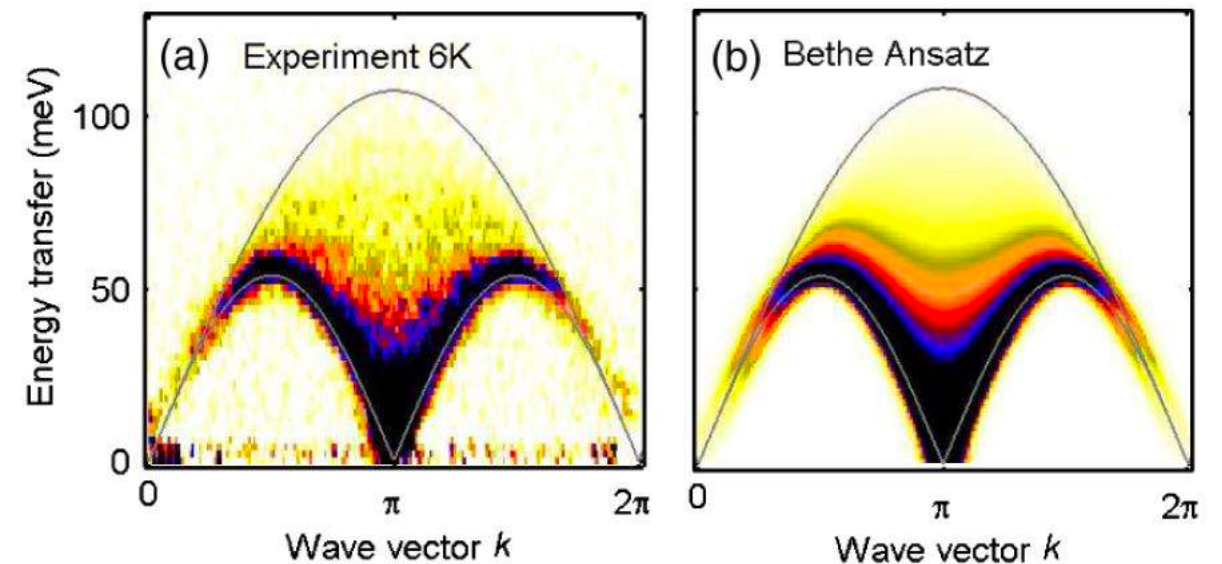
ARPES



1D chain structure in Sr_2CuO_3



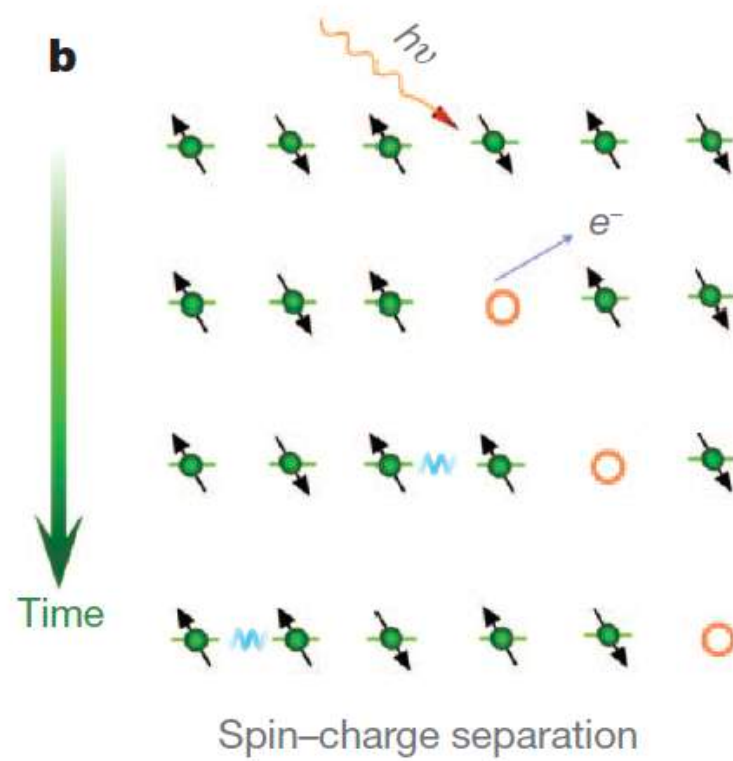
INS



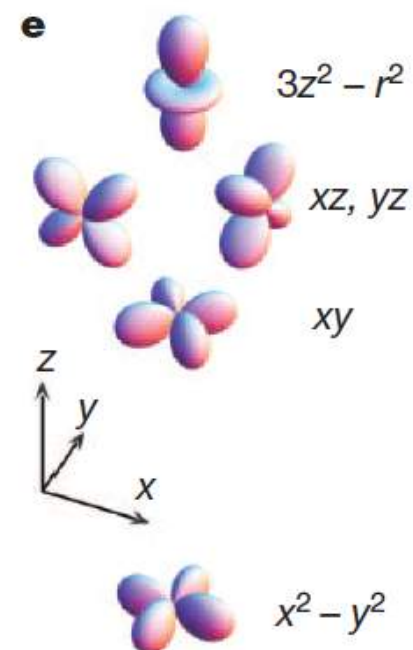
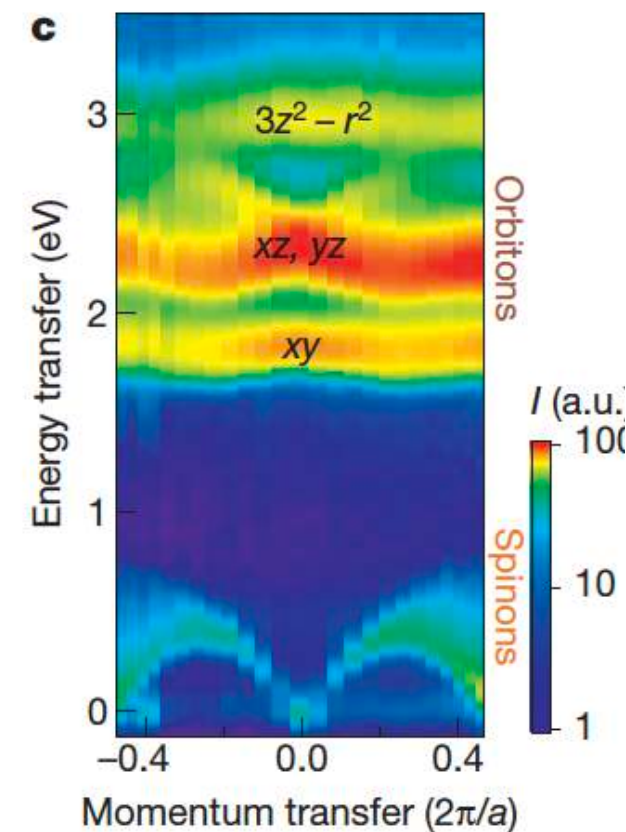
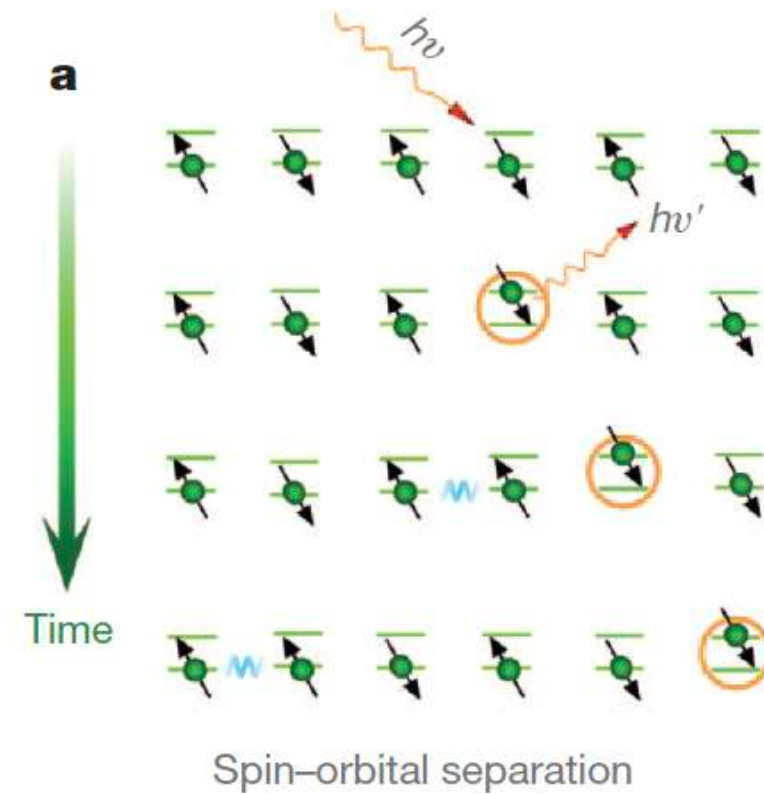
C. Kim et al. PRL (1996)

BJK, C. Kim et al. Nature Phys. (2006)

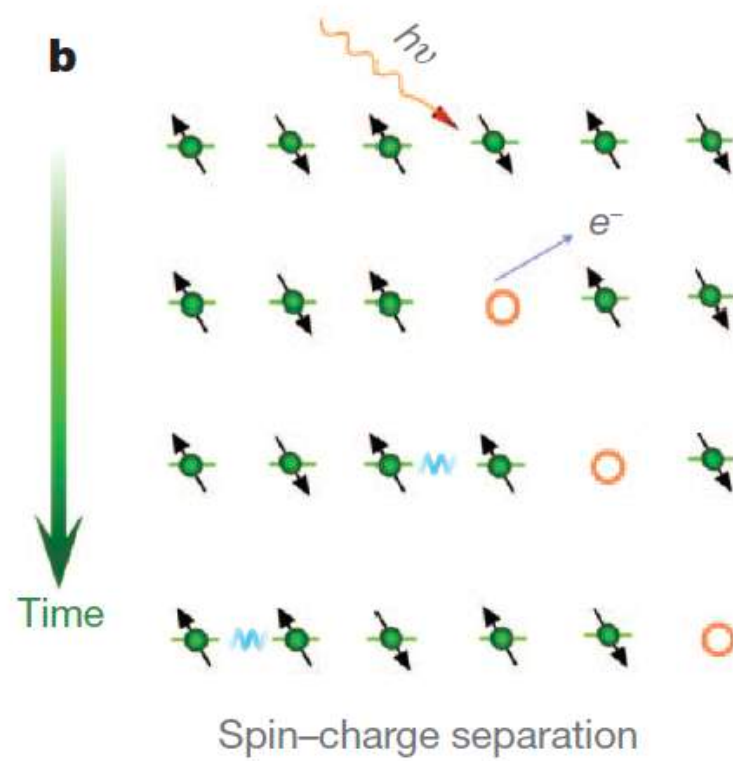
Spin-charge separation



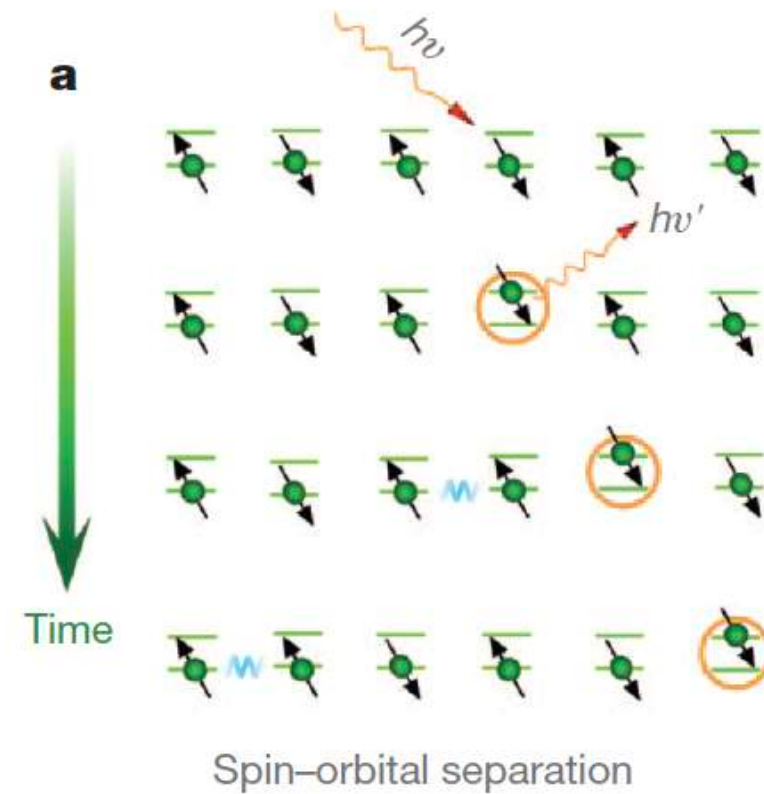
Spin-orbital separation



Spin-charge separation



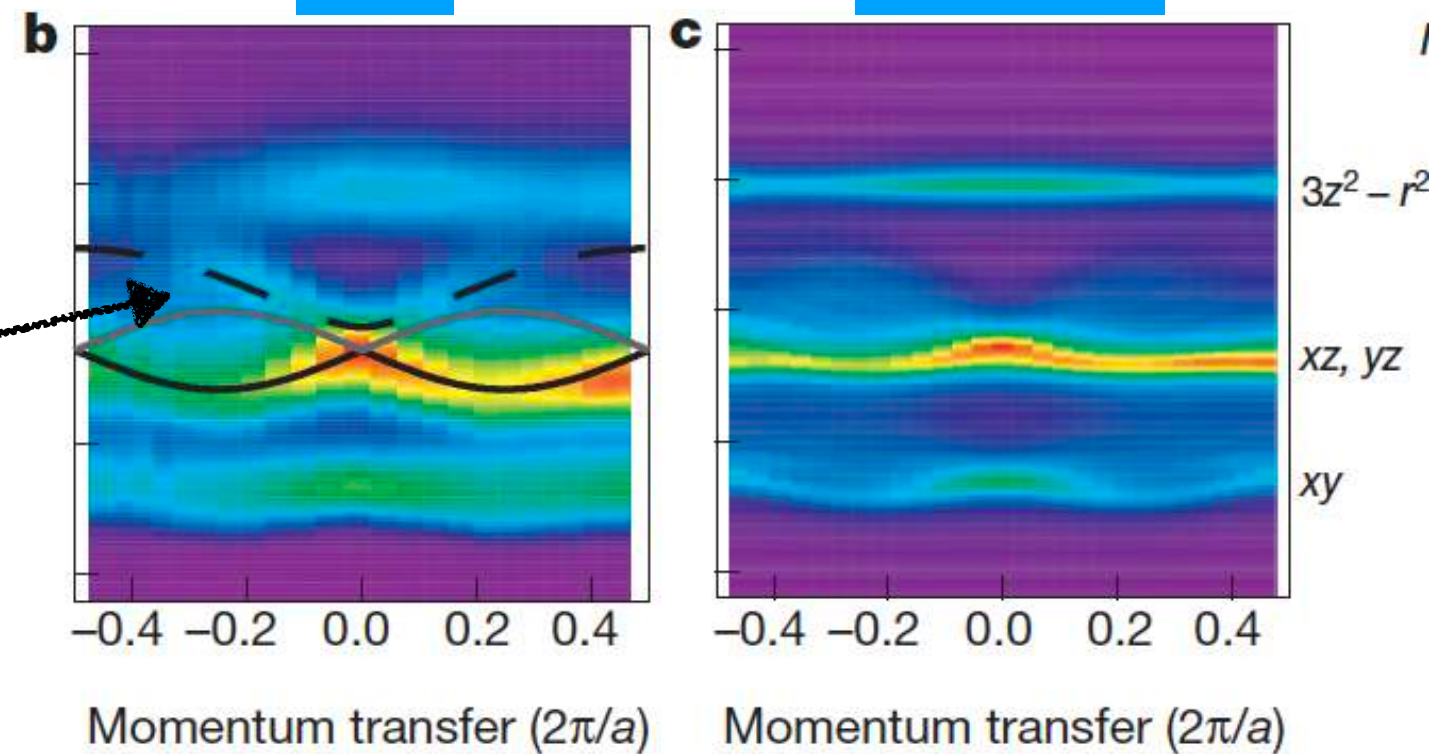
Spin-orbital separation



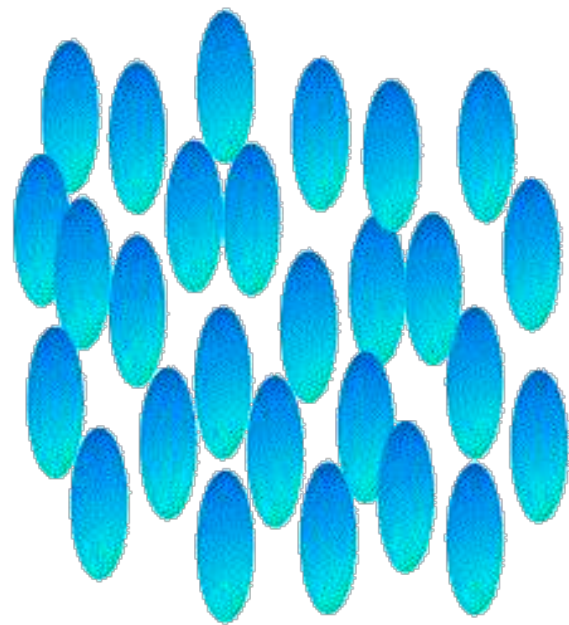
RIXS

Simulation

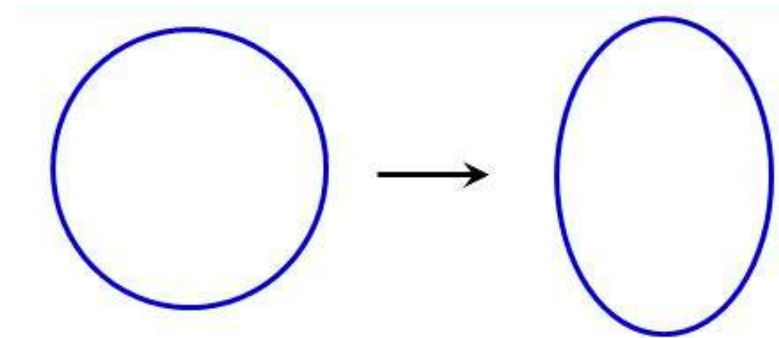
Spinon-orbiton continuum



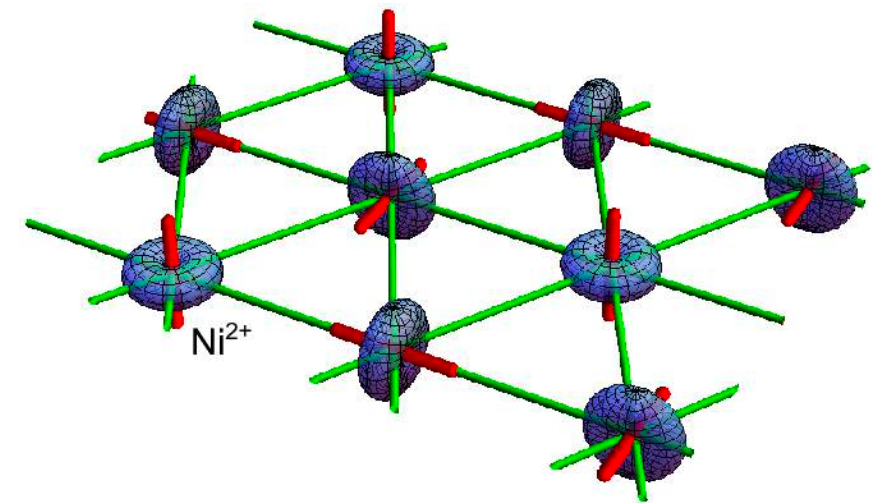
Spin nematic



Liquid crystal



Fermi surface
anisotropic distortions



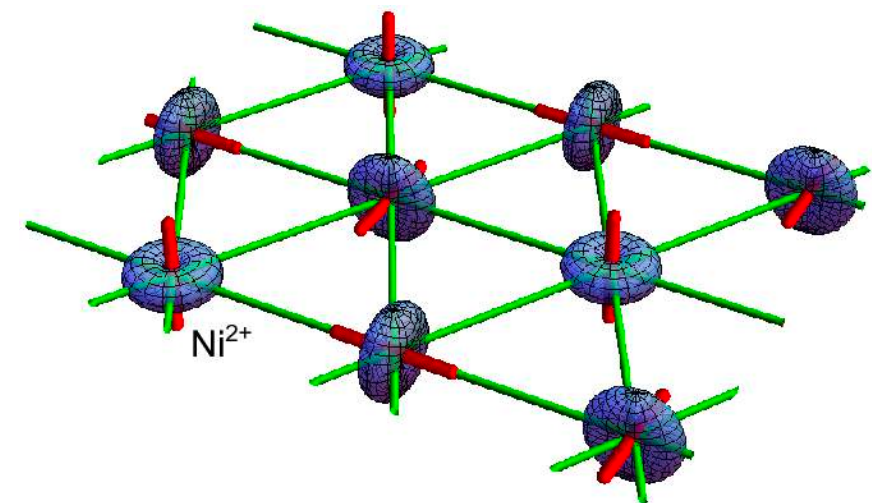
spin nematic

Spin nematic

$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + E \sum_i S_{zi}^2.$$

$S_z = \pm 1$ 

$S_z = 0$ 



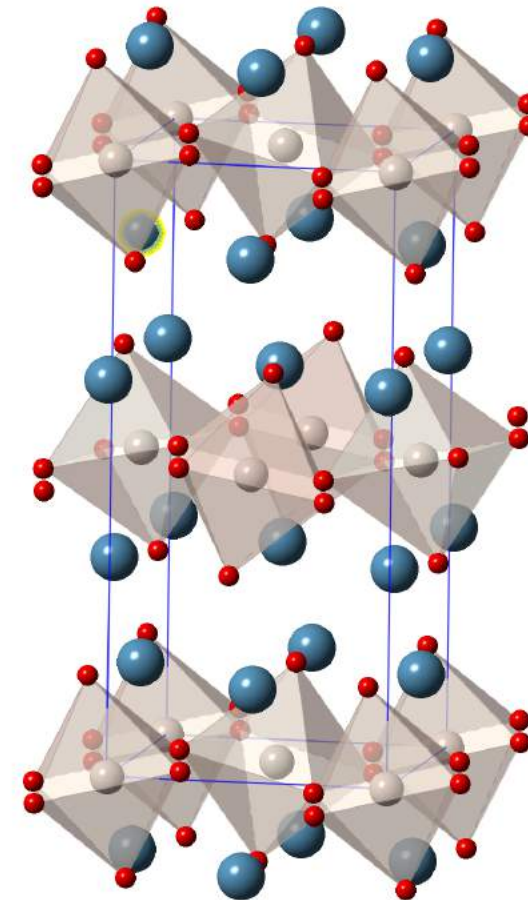
spin nematic

breaks rotational symmetry but preserves time-reversal symmetry

$$\langle \mathbf{S} \rangle = 0 \text{ and } \langle \mathbf{S}^2 \rangle \neq 0$$

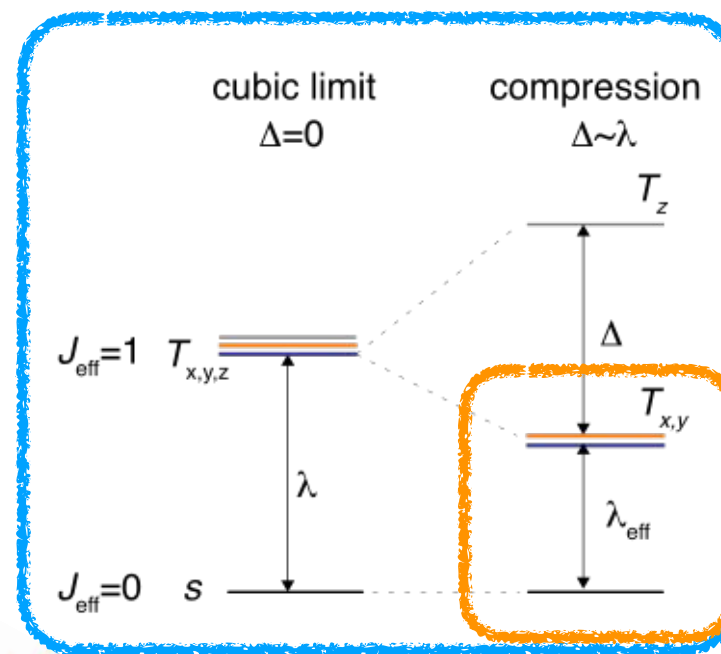
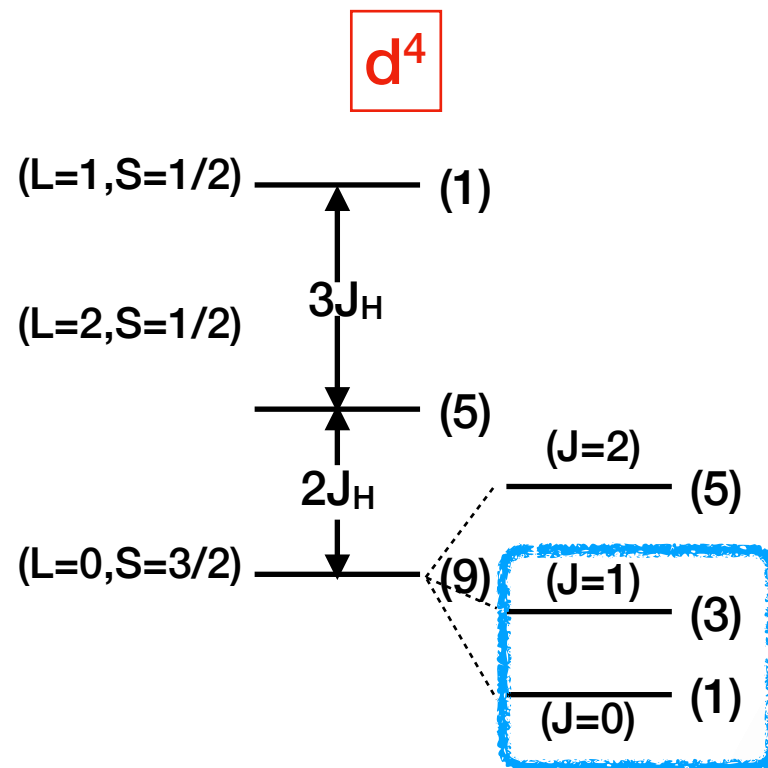
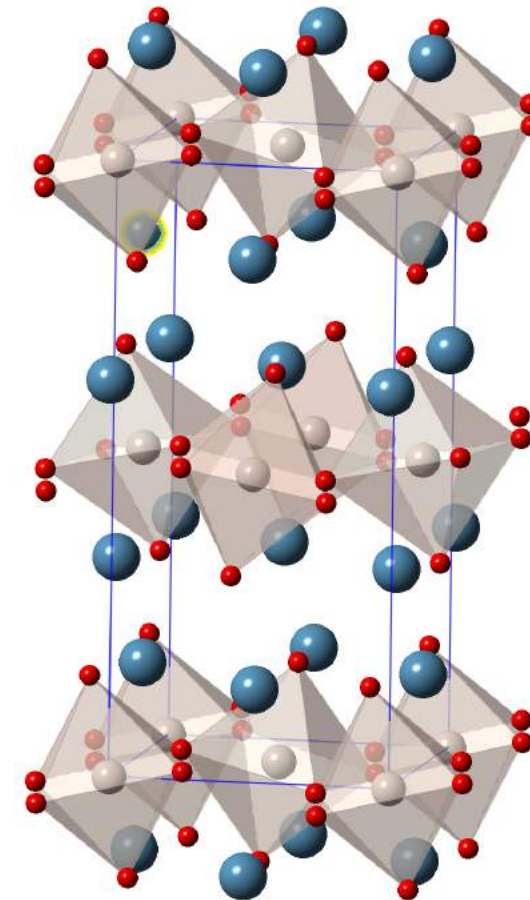
4d Mott insulator Ca_2RuO_4

- Metal-to-insulator transition at $T=360$ K
- (π,π) antiferromagnetic below $T_N=110$ K
- d^4 low-spin configuration, nominally $S=1$



4d Mott insulator Ca_2RuO_4

- Metal-to-insulator transition at $T=360$ K
- (π,π) antiferromagnetic below $T_N=110$ K
- d^4 low-spin configuration, nominally $S=1$



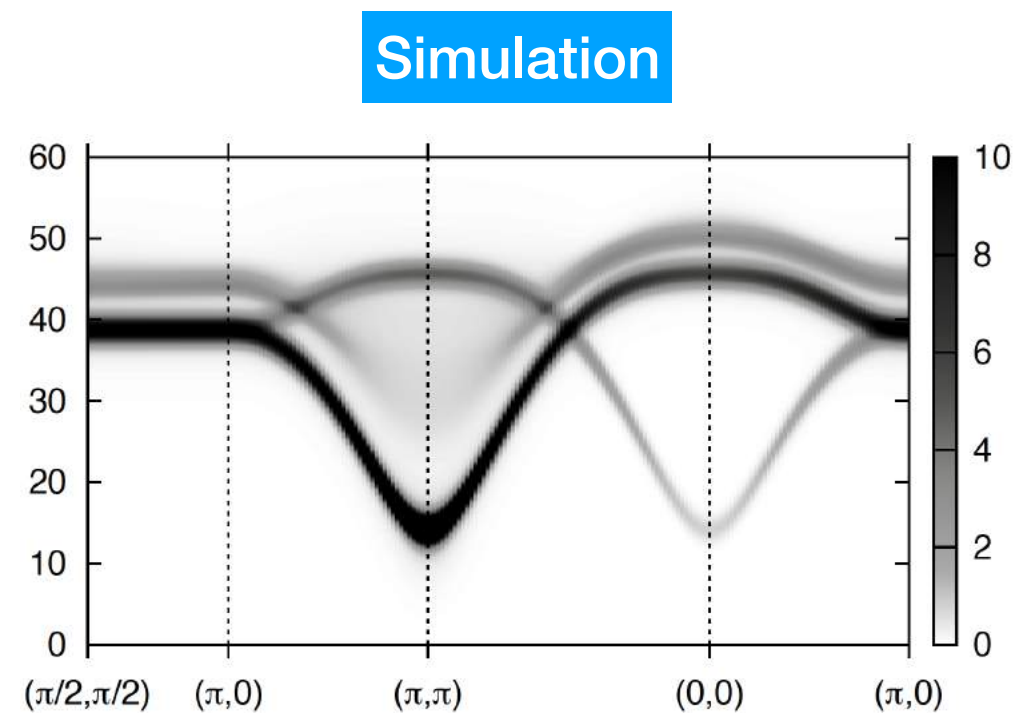
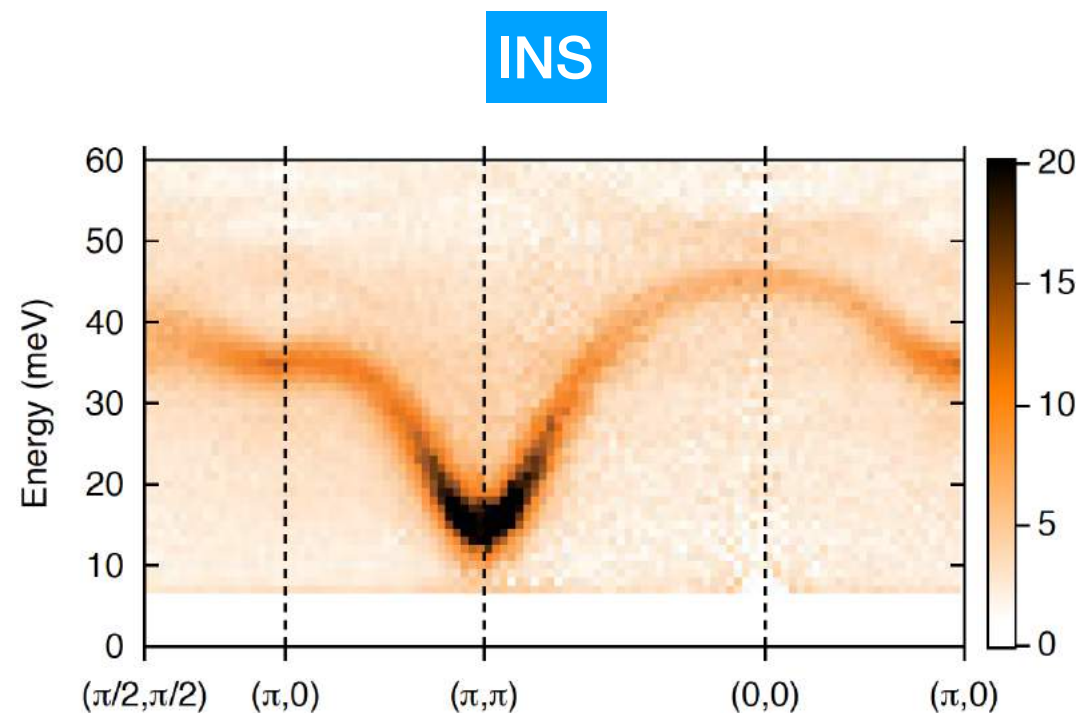
Pbca space group

Octahedra:
rotated about c axis
tilted about b axis,
compressed along c

singlet-triplet model

effective $S=1$ model

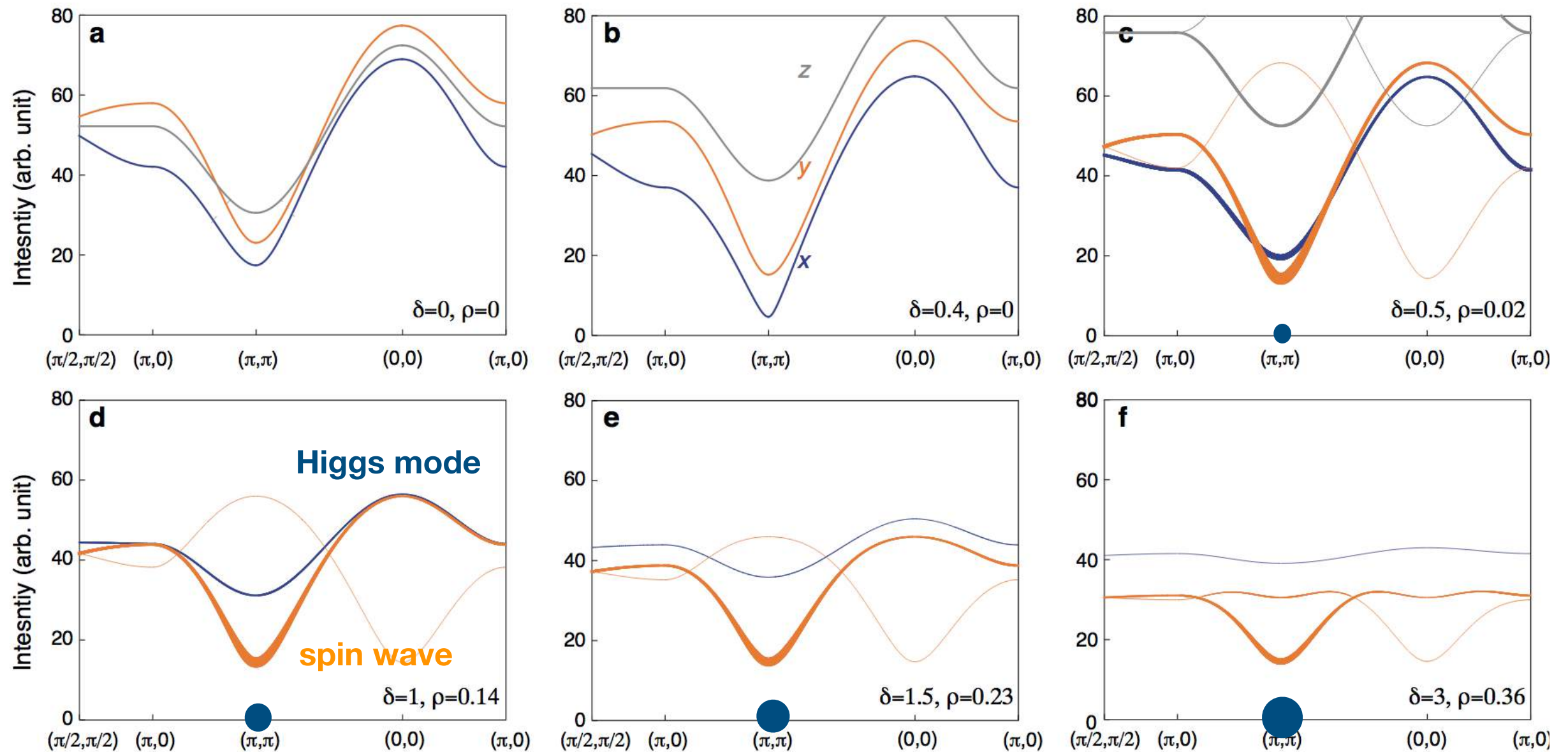
Anomalous dispersion



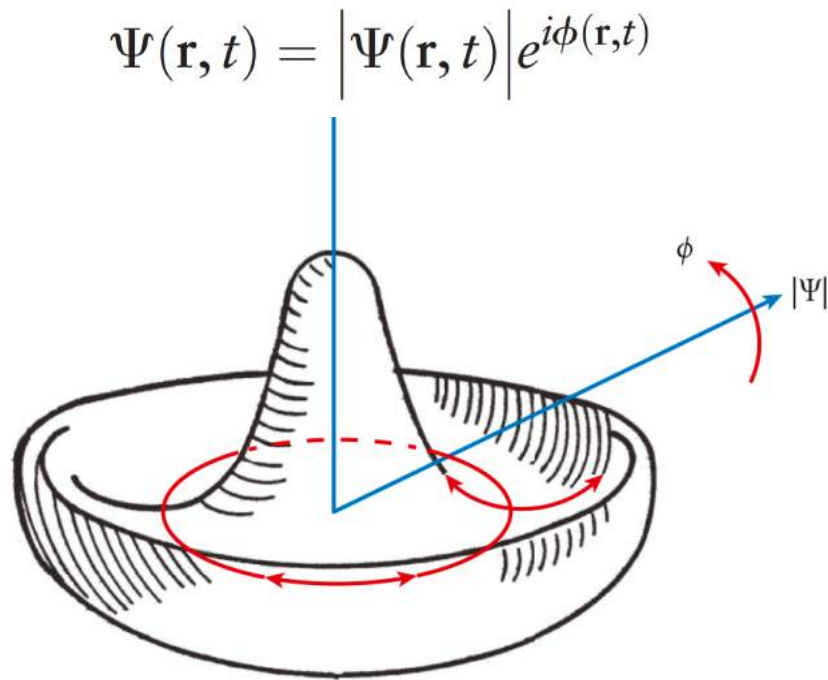
$$H = J \sum_{\langle ij \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j - \alpha S_{zi} S_{zj}) + E \sum_i S_{zi}^2 + \epsilon \sum_i S_{xi}^2.$$

$$E \simeq 24, J \simeq 5.6, \epsilon \simeq 4.0 \text{ meV, and } \alpha = 0$$

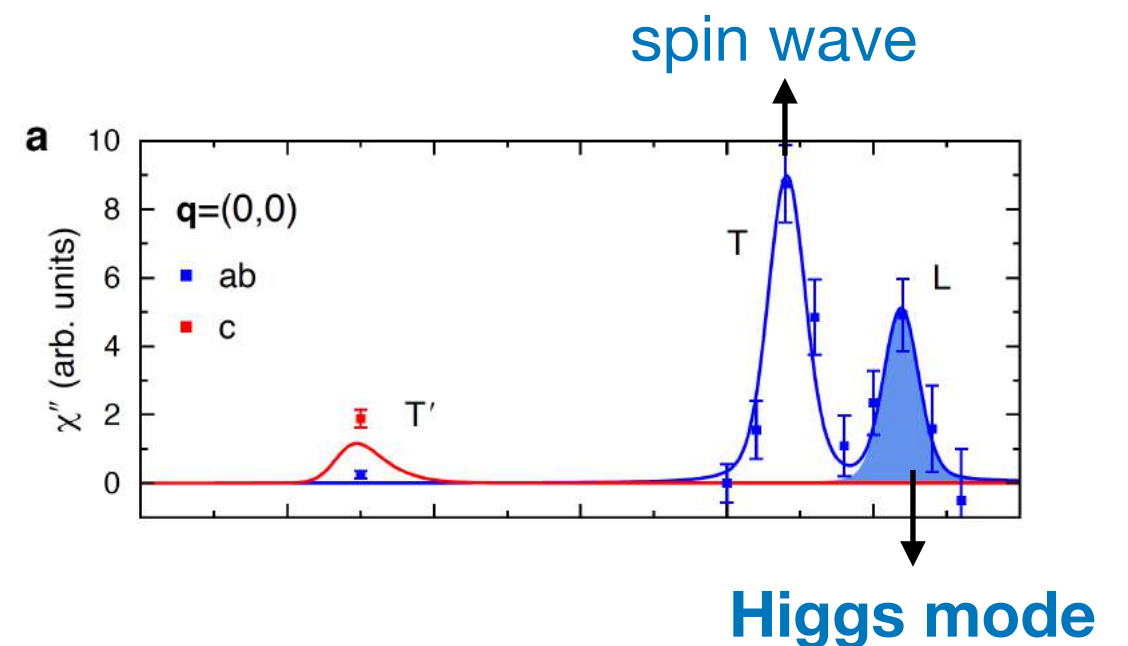
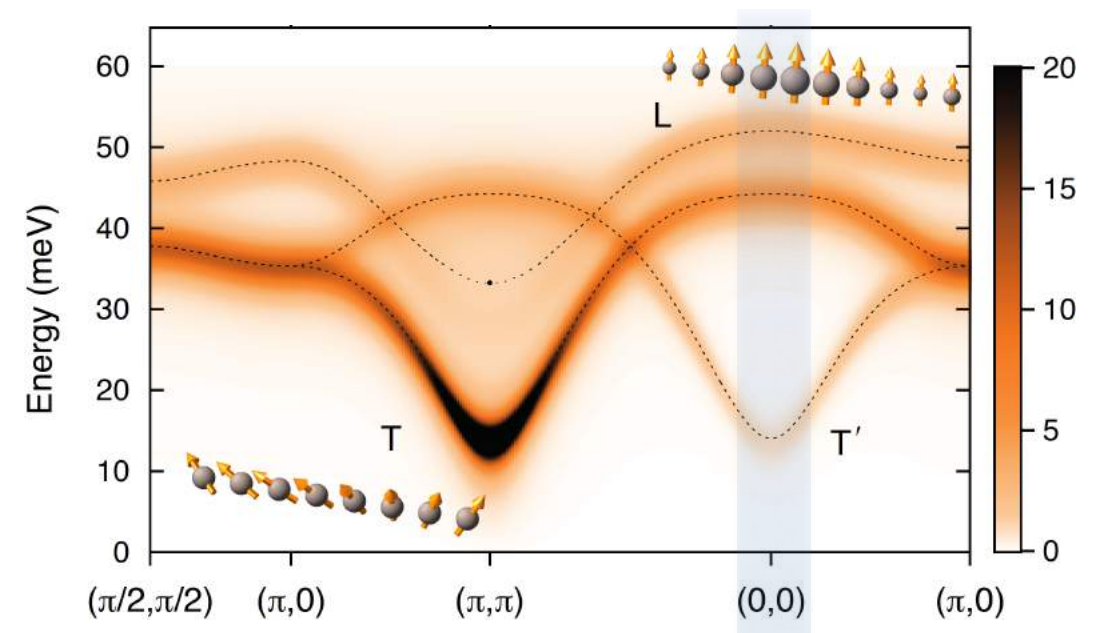
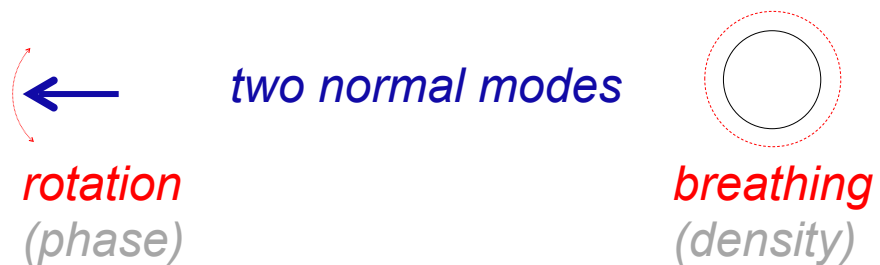
Magnetic order by exciton condensation



Higgs amplitude mode

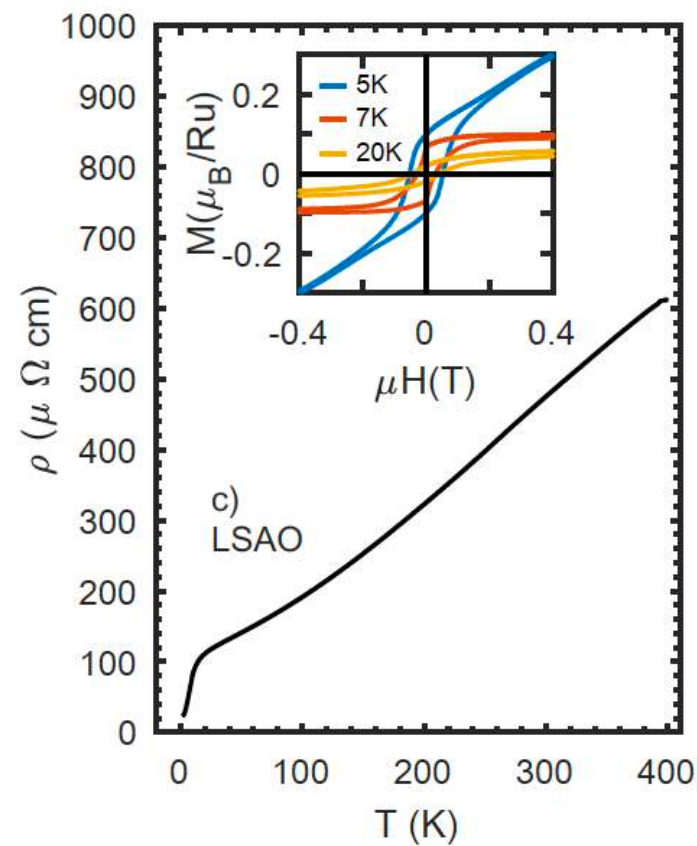
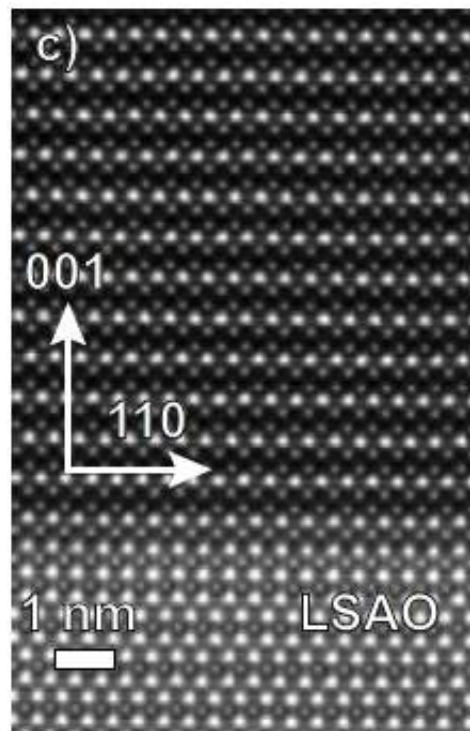


 Pekker D, Varma CM. 2015.
Annu. Rev. Condens. Matter Phys. 6:269–297

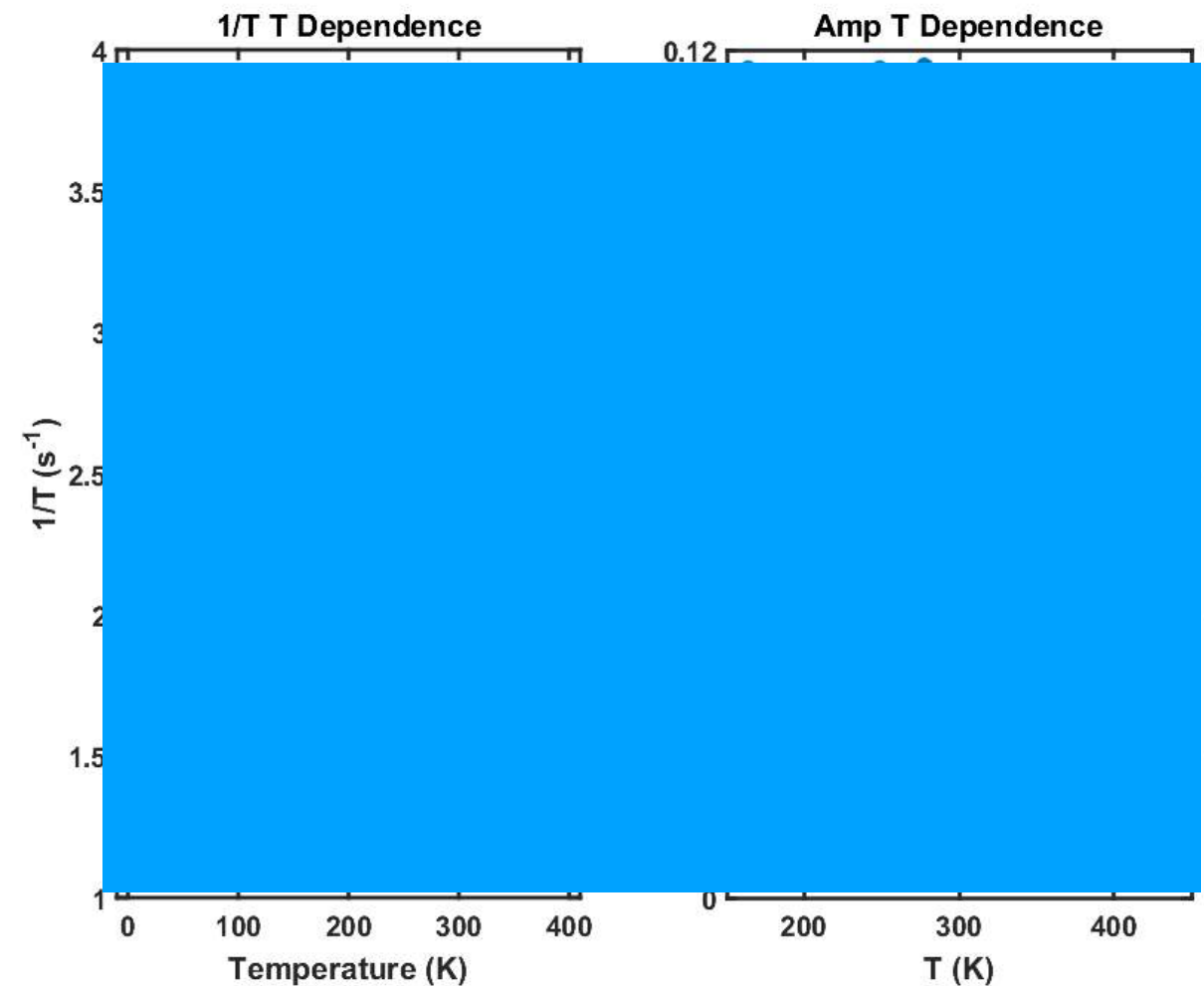


Magnetic order suppressed

CRO on LSAO



β -NMR indicates an electronic order!

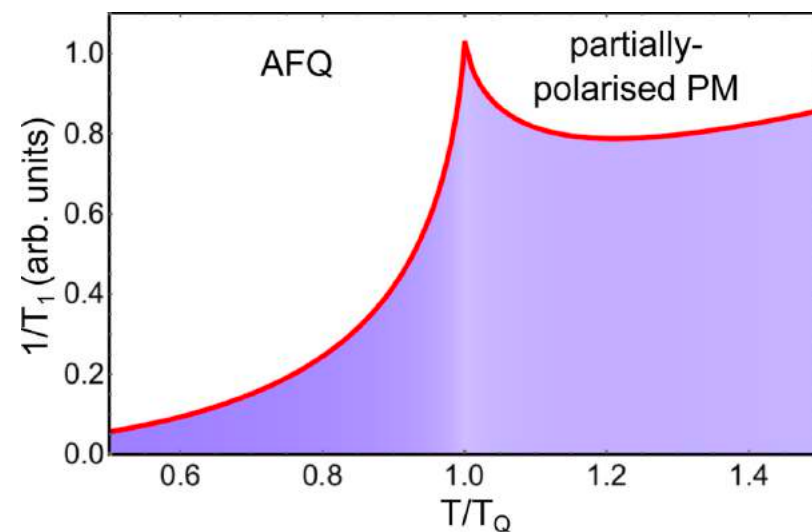


Spin nematic?

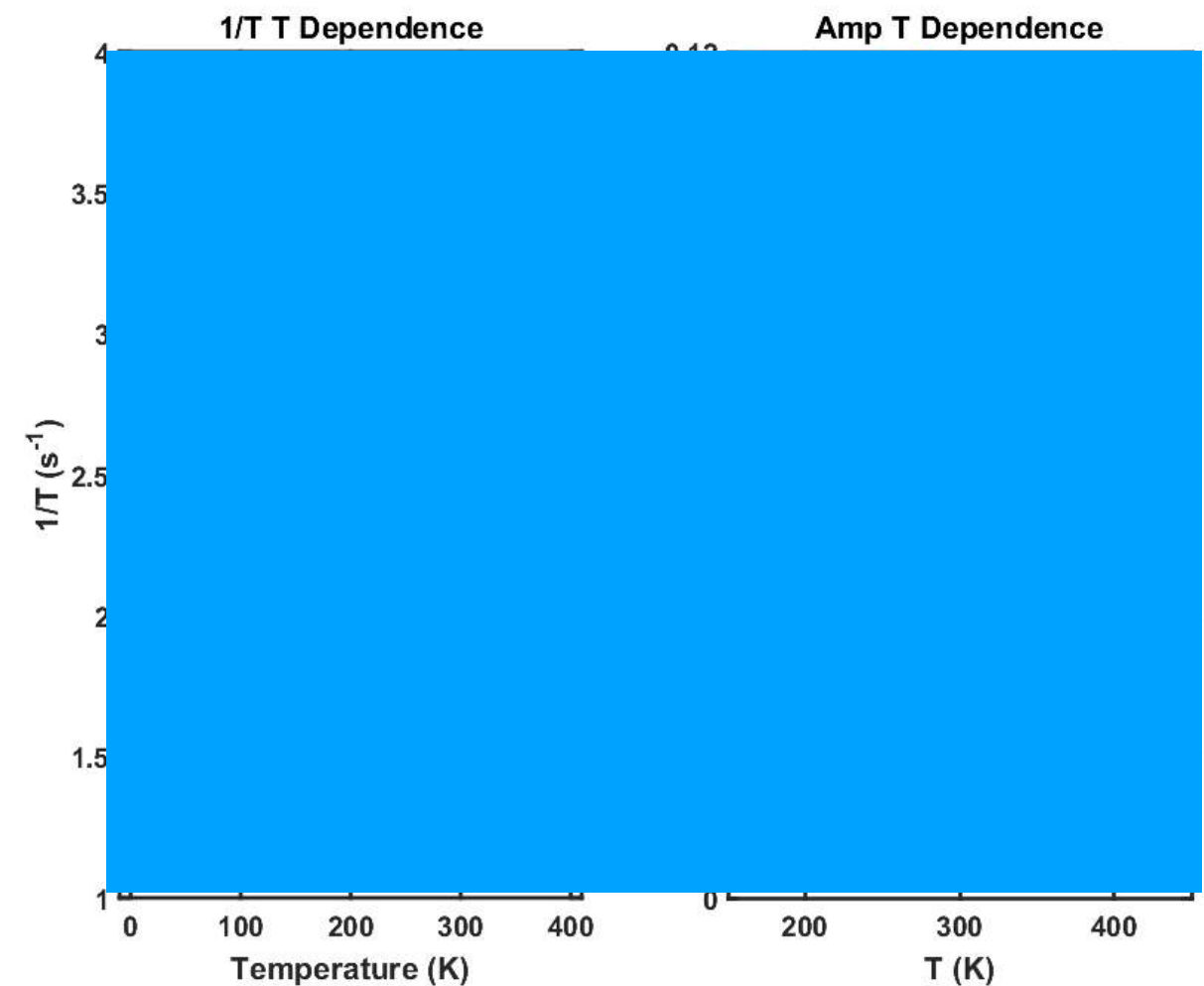
PHYSICAL REVIEW B **93**, 184419 (2016)

Theory of NMR $1/T_1$ relaxation in a quantum spin nematic in an applied magnetic field

Andrew Smerald^{1,2,3} and Nic Shannon^{2,3,4}



β -NMR indicates an electronic order!



C. Dietl, BJK et al. arxiv (2017),
C. Dietl, BJK (unpublished)

RIXS operators for t_{2g} orbital systems

unquenched orbital angular momentum

“fast collision approximation”

$$R_{\omega_i}^{\epsilon_i \epsilon_o} = T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} \quad \rightarrow \quad R \propto D^\dagger D = \frac{1}{3}(R_Q + iR_M)$$

c number

$$R_Q = \sum_{\alpha} \epsilon_{\alpha} \epsilon'_{\alpha} Q_{\alpha\alpha} - \frac{1}{2} \sum_{\alpha > \beta} (\epsilon_{\alpha} \epsilon'_{\beta} + \epsilon_{\beta} \epsilon'_{\alpha}) Q_{\alpha\beta}$$

$$R_M = \frac{1}{2}(\boldsymbol{\epsilon} \times \boldsymbol{\epsilon}') \cdot \mathbf{N}.$$

Q_{zz} (quadrupole)	L_3 edge	L_2 edge
$d^1, (-1)d^5$	$-2L_z^2 + 2L_z S_z$	$-L_z^2 - 2L_z S_z$
$d^2, (-1)d^4$	$2L_z^2 + L_z S_z$	$L_z^2 - L_z S_z$
Q_{xy} (quadrupole)	L_3 edge	L_2 edge
$d^1, (-1)d^5$	$-2L_x L_y - 2L_y L_x + 2L_x S_y + 2L_y S_x$	$-L_x L_y - L_y L_x - 2L_x S_y - 2L_y S_x$
$d^2, (-1)d^4$	$2L_x L_y + 2L_y L_x + L_x S_y + L_y S_x$	$L_x L_y + L_y L_x - L_x S_y - L_y S_x$
N_z (magnetic)	L_3 edge	L_2 edge
d^1, d^5	$2L_z - 4S_z + 8L_z^2 S_z - 2L_z(\mathbf{L} \cdot \mathbf{S}) - 2(\mathbf{L} \cdot \mathbf{S})L_z$	$L_z + 4S_z - 8L_z^2 S_z + 2L_z(\mathbf{L} \cdot \mathbf{S}) + 2(\mathbf{L} \cdot \mathbf{S})L_z$
d^2, d^4	$2L_z - 4L_z^2 S_z + L_z(\mathbf{L} \cdot \mathbf{S}) + (\mathbf{L} \cdot \mathbf{S})L_z$	$L_z + 4L_z^2 S_z - L_z(\mathbf{L} \cdot \mathbf{S}) - (\mathbf{L} \cdot \mathbf{S})L_z$
d^3	$(4/3)S_z$	$-(4/3)S_z$

Verifying spin nematic using RXS

unquenched orbital angular momentum

“fast collision approximation”

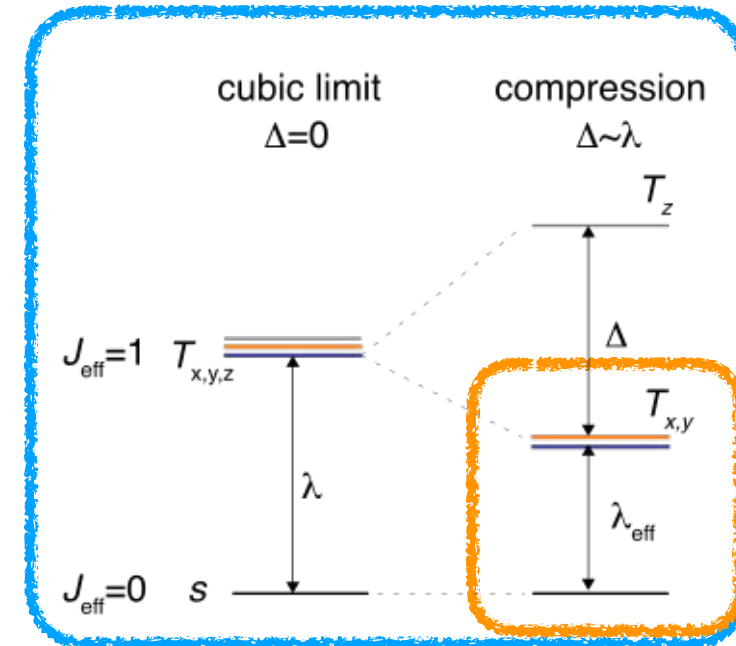
$$R_{\omega_i}^{\epsilon_i \epsilon_o} = T_{\epsilon_o}^\dagger \frac{1}{\omega_i + E_i + i\Gamma/2 - H} T_{\epsilon_i} \quad \text{c number} \quad \rightarrow \quad R \propto D^\dagger D = \frac{1}{3}(R_Q + i R_M)$$

$$R_Q = \sum_{\alpha} \epsilon_{\alpha} \epsilon'_{\alpha} Q_{\alpha\alpha} - \frac{1}{2} \sum_{\alpha > \beta} (\epsilon_{\alpha} \epsilon'_{\beta} + \epsilon_{\beta} \epsilon'_{\alpha}) Q_{\alpha\beta}$$

$$R_M = \frac{1}{2} (\boldsymbol{\epsilon} \times \boldsymbol{\epsilon}') \cdot \mathbf{N}.$$

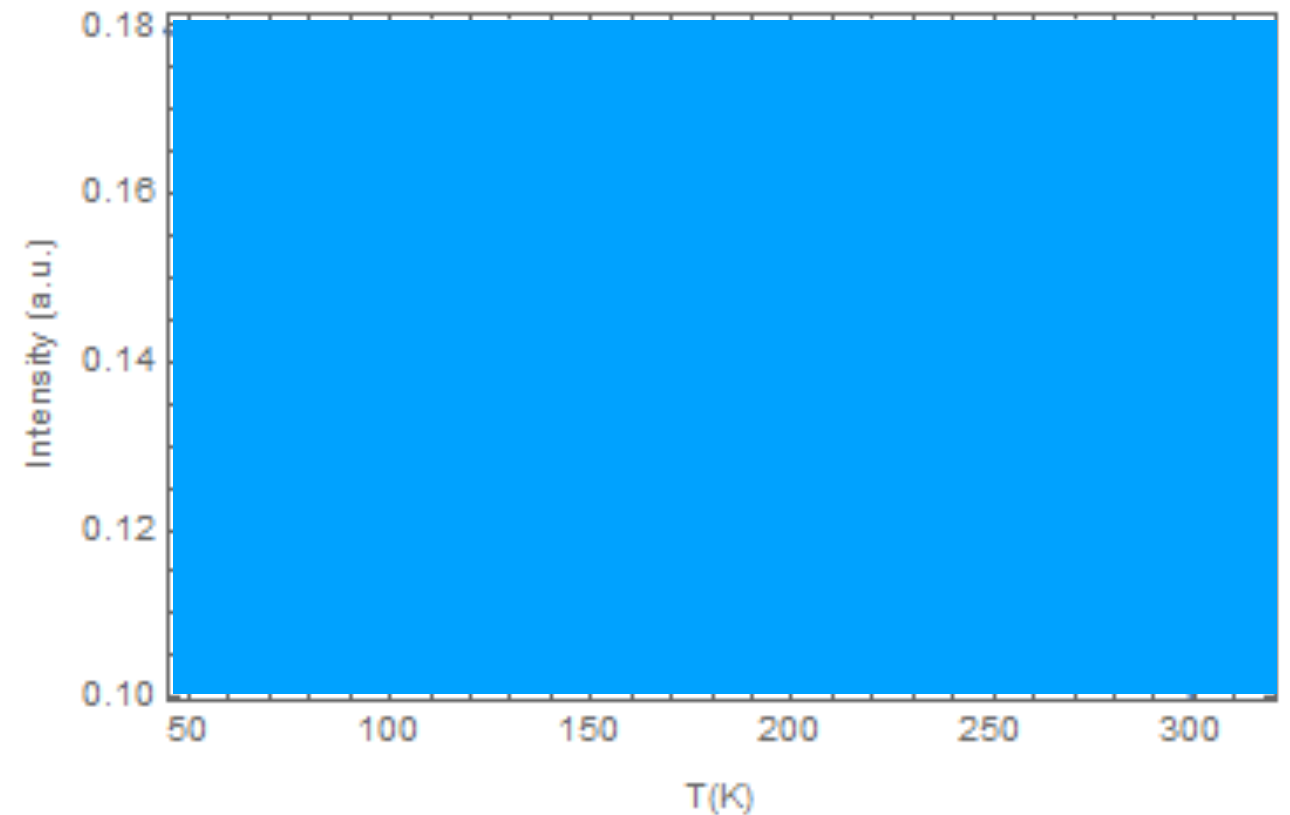
$$\begin{aligned} R_Q = & \frac{1}{2} [a_{xy}(\epsilon_x \epsilon'_x + \epsilon_y \epsilon'_y) + a_z \epsilon_z \epsilon'_z] \tilde{S}_z^2 \\ & + \frac{1}{2} b_{xy} [(\epsilon_x \epsilon'_x - \epsilon_y \epsilon'_y)(\tilde{S}_y^2 - \tilde{S}_x^2) \\ & + (\epsilon_x \epsilon'_y + \epsilon_y \epsilon'_x)(\tilde{S}_x \tilde{S}_y + \tilde{S}_y \tilde{S}_x)] \\ & + \frac{1}{2} b_z [(\epsilon_y \epsilon'_z + \epsilon_z \epsilon'_y)(\tilde{S}_y \tilde{S}_z + \tilde{S}_z \tilde{S}_y) \\ & + (\epsilon_z \epsilon'_x + \epsilon_x \epsilon'_z)(\tilde{S}_z \tilde{S}_x + \tilde{S}_x \tilde{S}_z)], \end{aligned}$$

$$\begin{aligned} R_M = & \frac{1}{2} c_{xy} [(\epsilon_y \epsilon'_z - \epsilon_z \epsilon'_y) \tilde{S}_x + (\epsilon_z \epsilon'_x - \epsilon_x \epsilon'_z) \tilde{S}_y] \\ & + \frac{1}{2} c_z (\epsilon_x \epsilon'_y - \epsilon_y \epsilon'_x) \tilde{S}_z. \end{aligned}$$

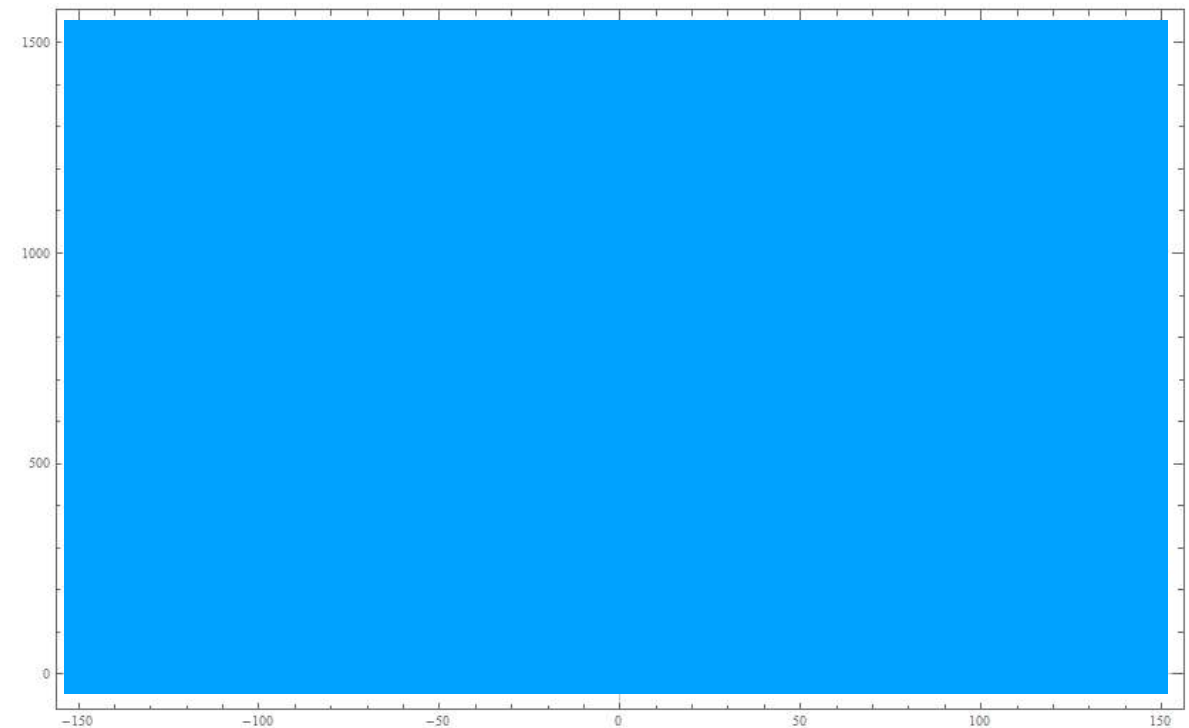


Verifying spin nematic using RXS

order parameter grows around $T=300$ K



xy quadrupole



Recent theoretical proposals

Majorana fermions in Kitaev spin liquid

Spins fractionalize into Majorana fermions and emergent gauge fluxes

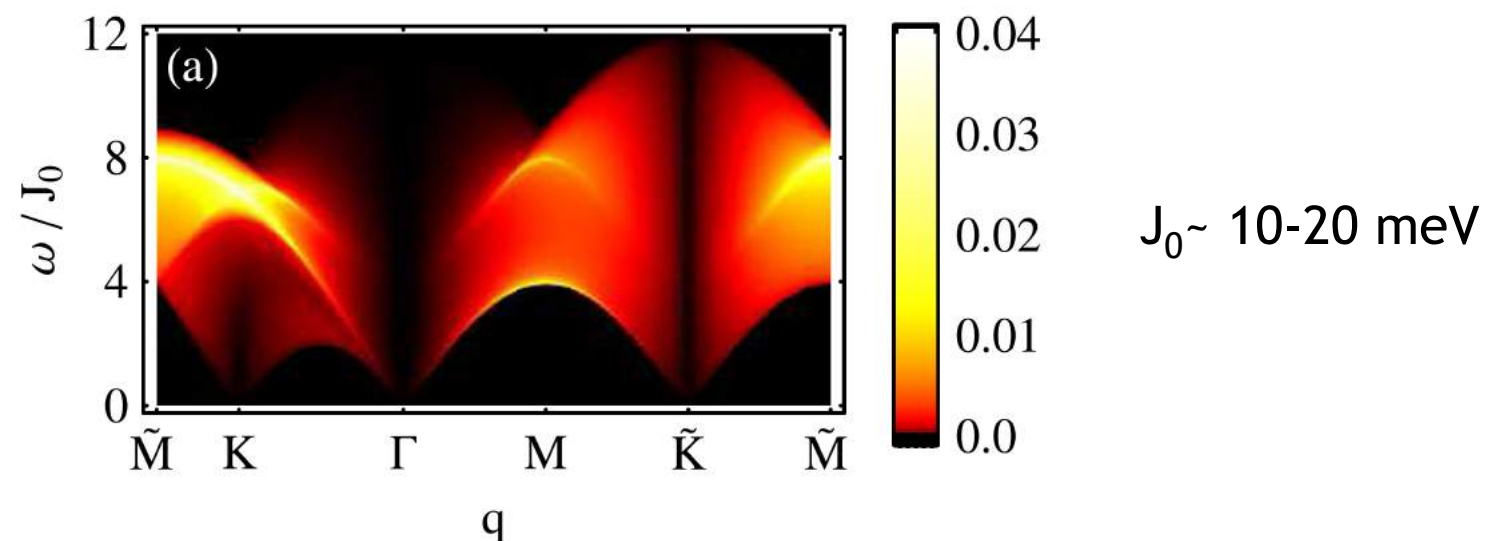
$$e^{-iHt} \text{ (on a honeycomb lattice with sites } i, j, k \text{ and fluxes } \pi) = \sum_j A_{ij}(t) \text{ (on a honeycomb lattice with sites } i, j, k \text{ and fluxes } \pi \text{ and a Majorana fermion at } j \text{)}$$

● Majorana Fermion

INS: dominated by static flux excitations

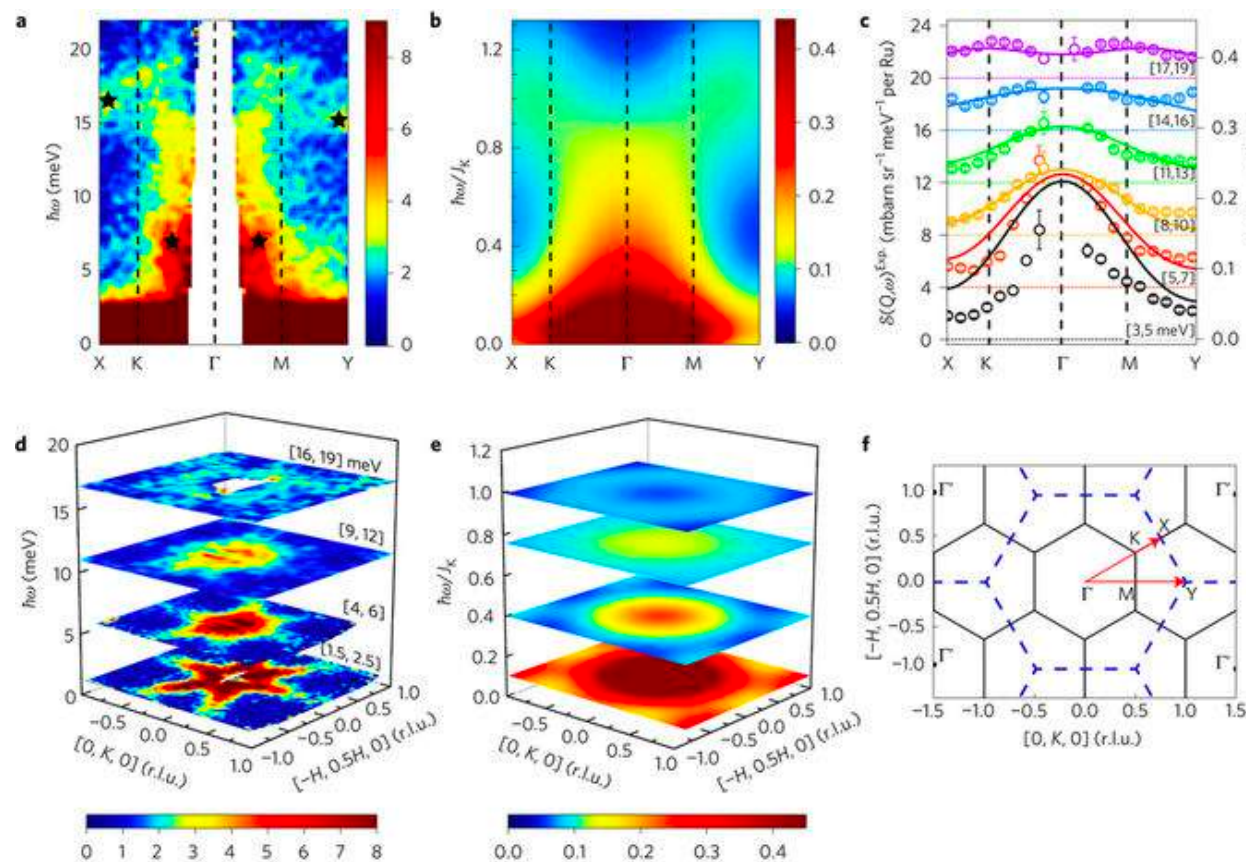
Raman scattering: two Majorana excitations but only at $q=0$

RIXS: Majorana (SC channel) and flux excitations (NSC channel)

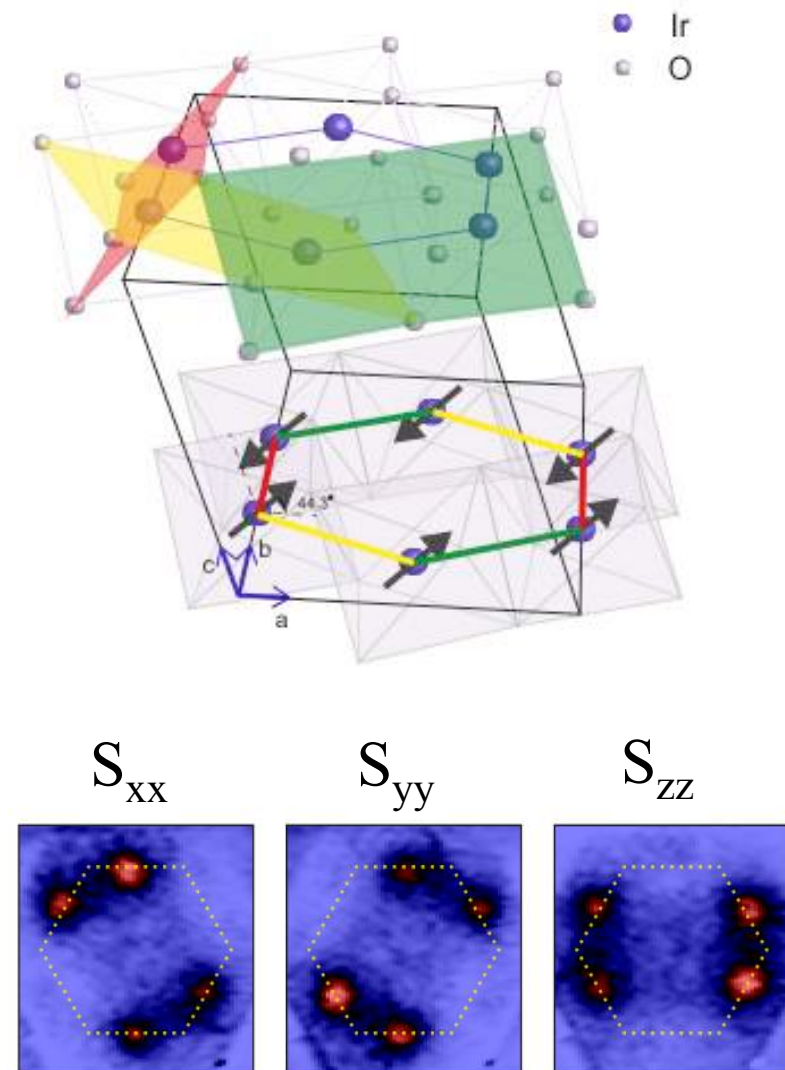


Candidate materials for Kitaev quantum spin liquid

RuCl_3



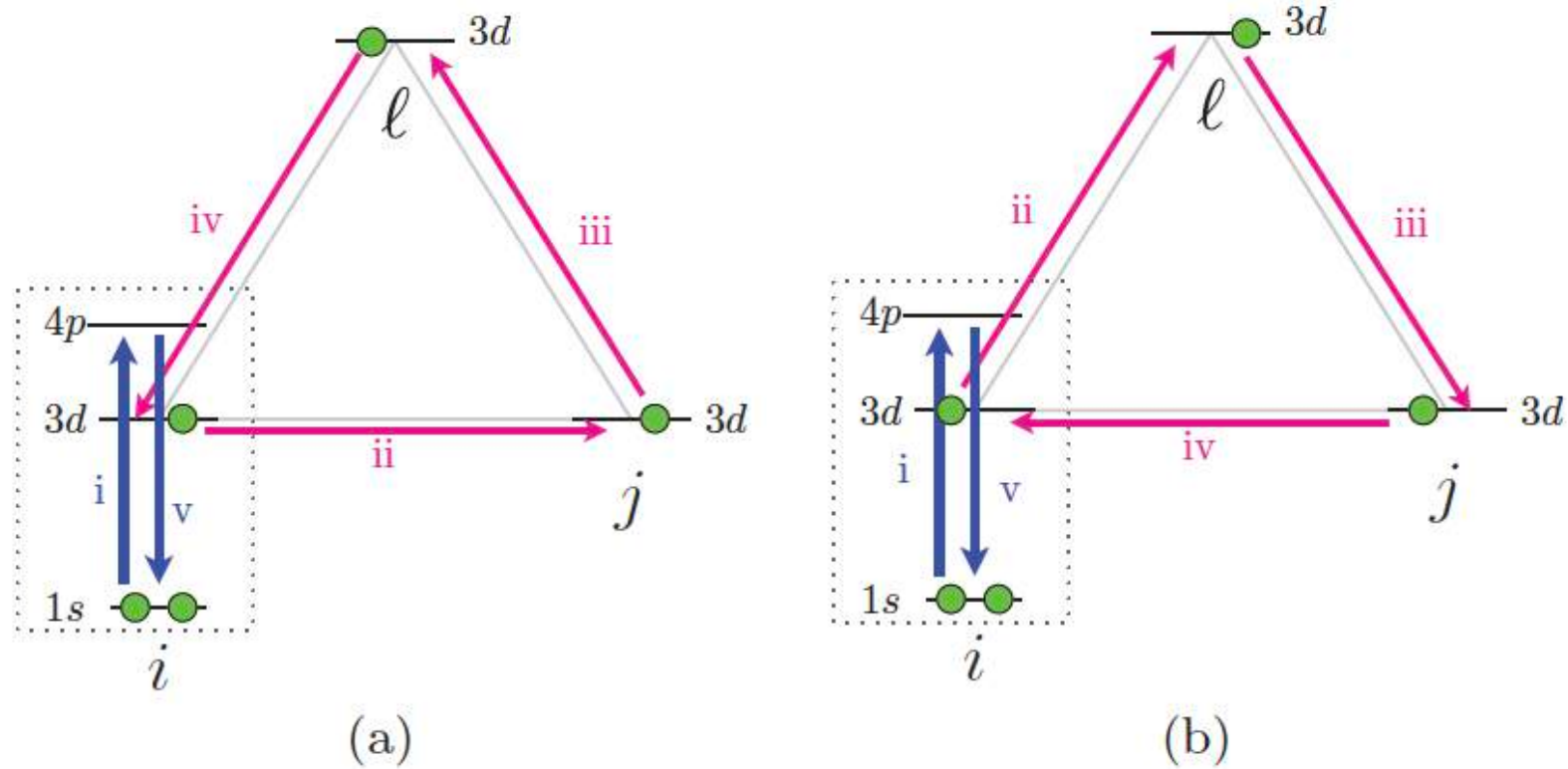
Na_2IrO_3



S.-H. Do et al., Nature Physics (2017)

S. H. Chun, BJK et al., Nature Physics (2015)

Gauge fields



$$\begin{aligned}
 T_{3\text{-sites}}^{(a)} &\propto (c_{is}^\dagger (J^\dagger)_{sp}^\beta c_{ip}) (t_{il} c_i^\dagger c_l) (t_{lj} c_l^\dagger c_j) (t_{ji} c_j^\dagger c_i) (c_{ip'}^\dagger J_{p's'}^\alpha c_{is'}) \\
 &= \text{tr}\{J^\alpha (J^\dagger)^\beta\} t_{il} t_{lj} t_{ji} \text{tr}\{\chi_l \chi_j \tilde{\chi}_i\} \\
 &= N^{\alpha\beta} t_{il} t_{lj} t_{ji} [2i \mathbf{S}_l \cdot (\mathbf{S}_j \times \mathbf{S}_i) + \dots], \tag{9}
 \end{aligned}$$

In a U(1) spin liquid, spin chirality translates into

$$\sim \langle i | b(\Delta \mathbf{k}, \Delta \omega) b(\mathbf{0}, 0) | i \rangle + \dots$$

where b is effective magnetic field associated with the emergent gauge boson

Summary

- RIXS is a powerful tool sensitive to charge, spin, and orbital degrees of freedom.
- As such, it is useful for measuring spin-wave (magnon) dispersions, but can also be used to detect more exotic particles not readily measured by conventional probes.

RIXS beamline @ PAL

