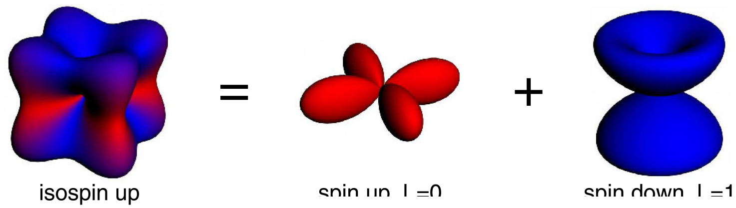
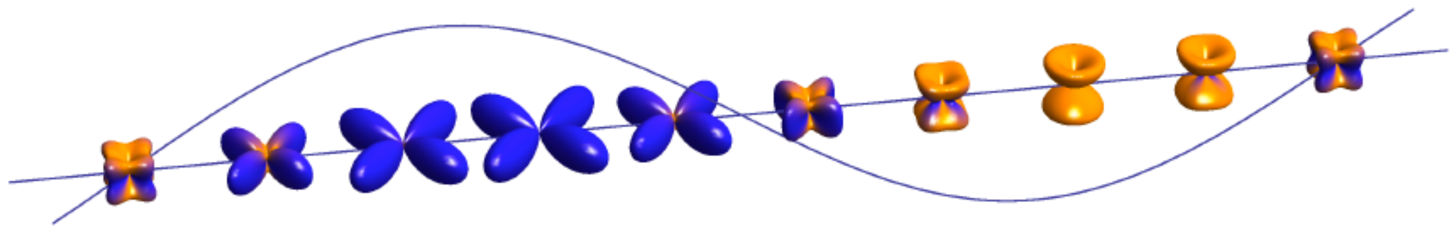
Resonant X-ray Scattering: Application to Iridates





B. J. Kim

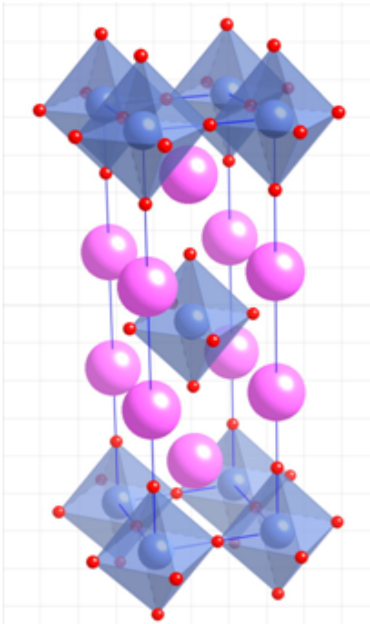
Max Planck Institute for Solid State Research

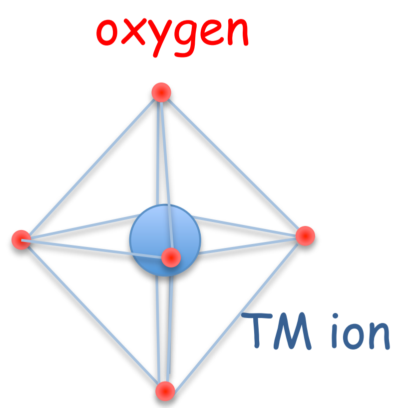
HSC18: Neutron and Synchrotron Radiation for Magnetism

Grenoble,14-18 Sep. 2015

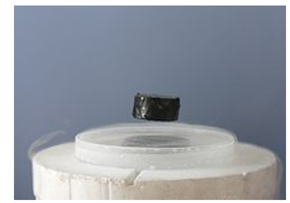
Transition metal oxides: Diverse Properties

(Layered) Perovskite structure

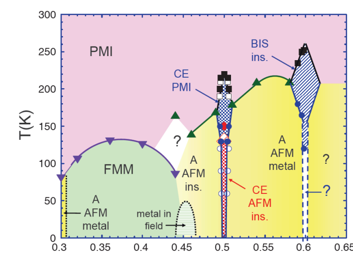




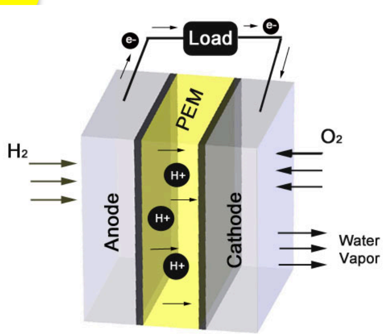
Cu: high-Tc superconductivity



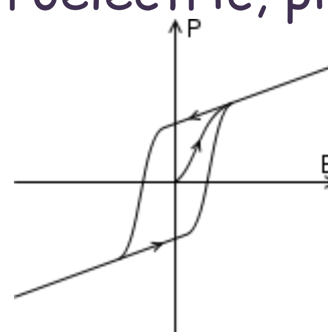
Mn: Colossal magnetoresistance



Ni: Fuel cell cathodes

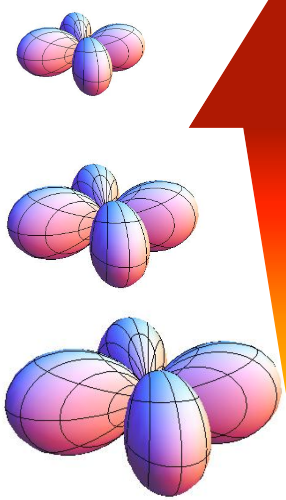


Ti: ferroelectric, piezoelectric



Transition metal oxides

Strong electron correlation

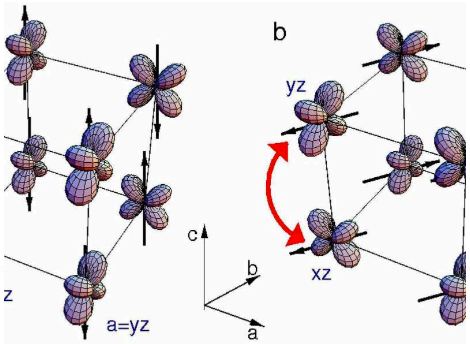


Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	*3d*
Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	*4d*
Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	*5d*

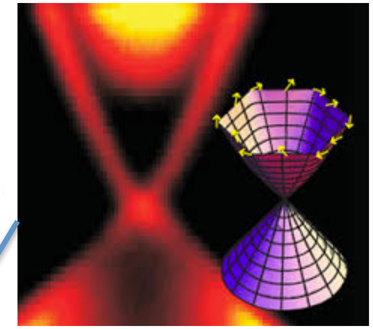
U~2 eV
ζ_{SO}*~0.5eV*

Strong spin-orbit coupling

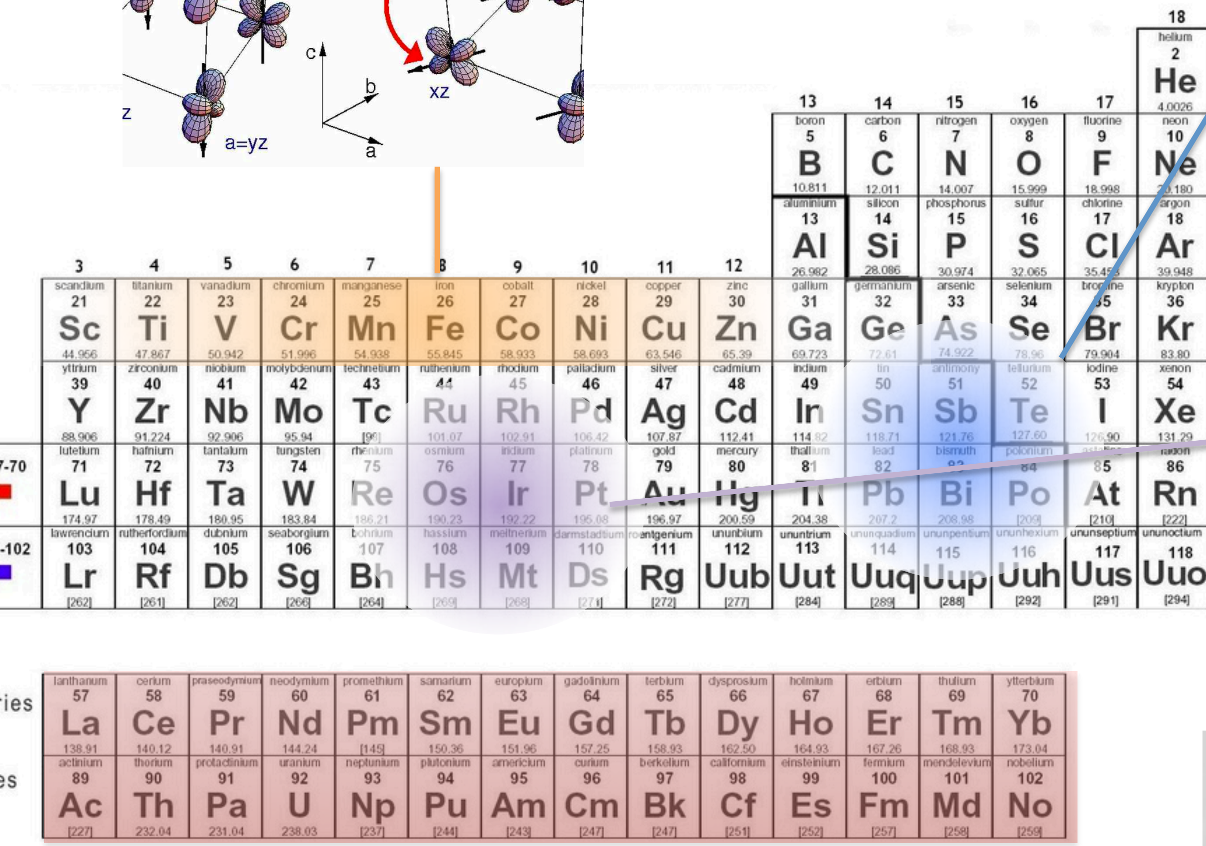
Correlation + Spin-Orbit Coupling



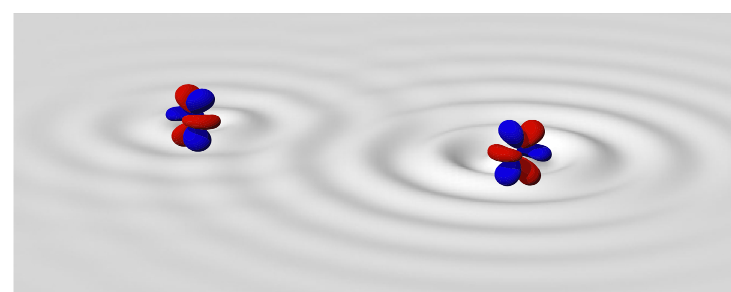
- high Tc superconductivity
- colossal magnetoresistance



- topological insulator



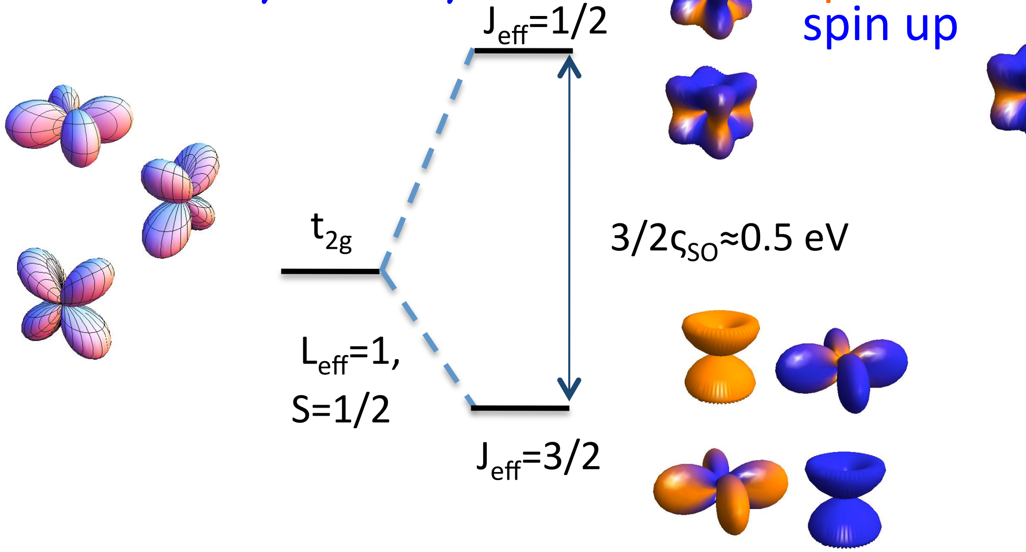
- novel topological phases
- Kitaev quantum spin liquid
- unconventional superconductivity

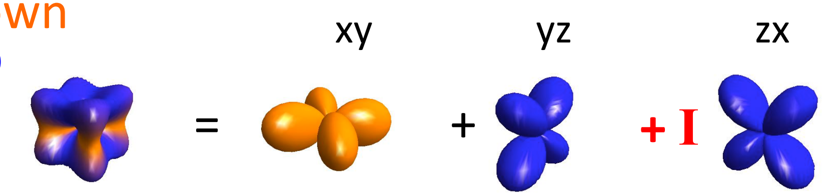


- Kondo physics
- heavy fermion

SOC Induced Mott Transition

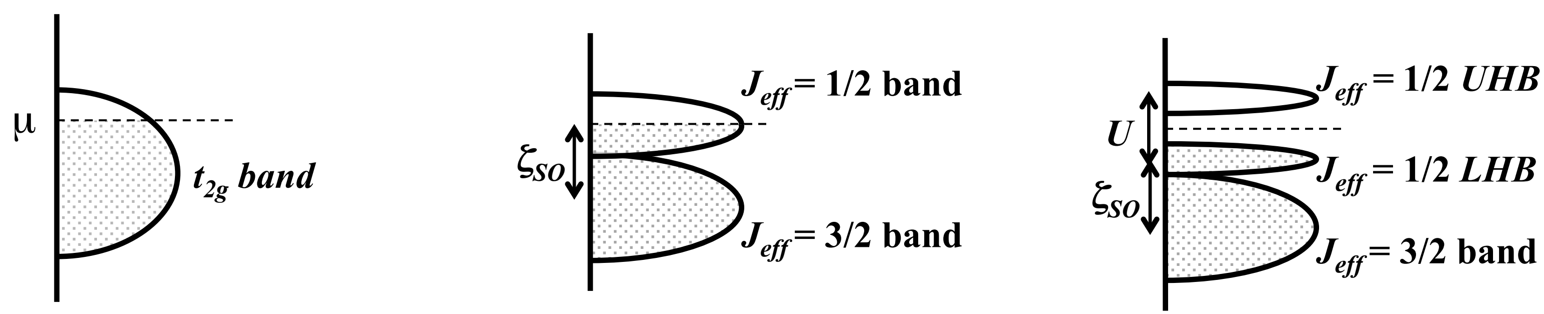
In cubic symmetry,





$$\langle L+2S\rangle = 1\mu_B$$

$$\langle L\rangle = \frac{2}{3}\mu_B, \langle 2S\rangle = \frac{1}{3}\mu_B$$



wide t_{2g}–band Metal **J_{eff} band split due to SO** **J_{eff}= 1/2 Mott ground state**

B. J. Kim et al. PRL 2009

J_{eff}=1/2 states

t_{2g} maps onto p(l=1, s=1/2) manifold (TP equivalence)

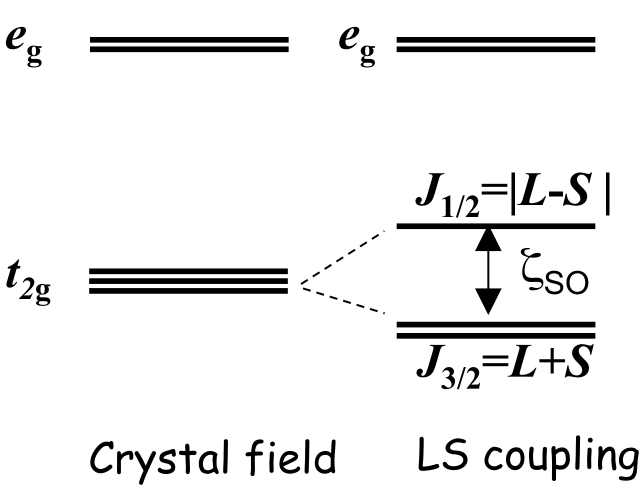
Taking only the t2g subspace, angular momentum operator **L** is given by

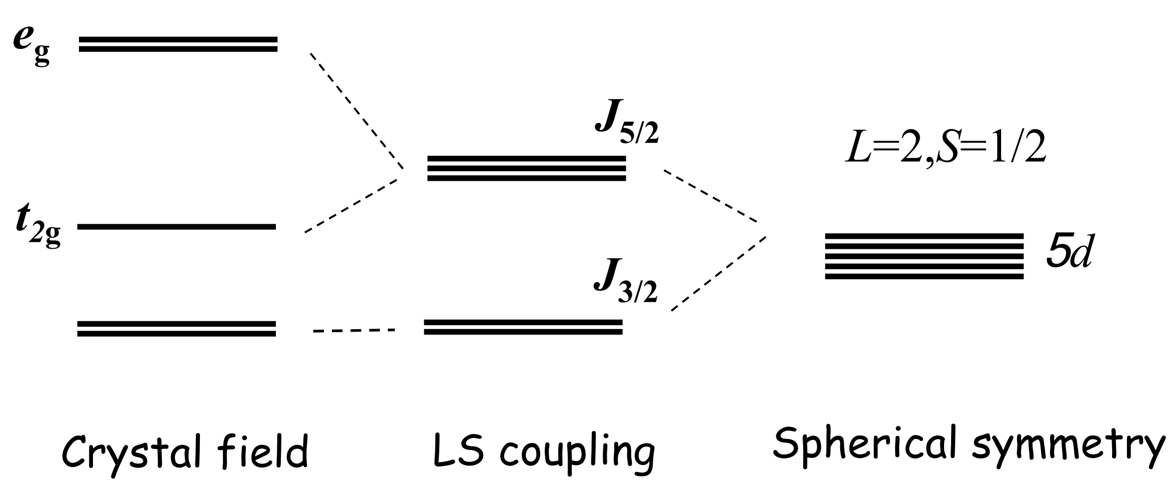
$$\begin{matrix} & xy & yz & zx \end{matrix}$$

$$L_x = \begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix} \qquad L_y = \begin{pmatrix} 0 & i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \qquad L_z = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & i \\ 0 & -i & 0 \end{pmatrix}$$

which can be mapped onto **L**=1 manifold with

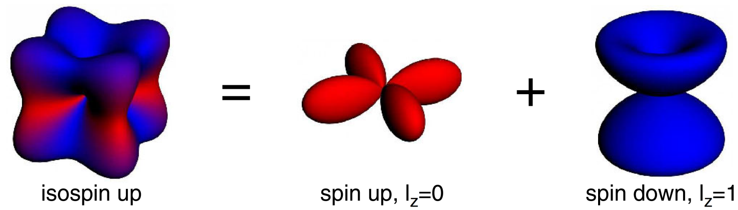
$$p_z \to xy\,,\ p_x \to yz\,,\ p_y \to zx \qquad \mathbf{L}_{t_{2g}} \to -\mathbf{L}_p$$





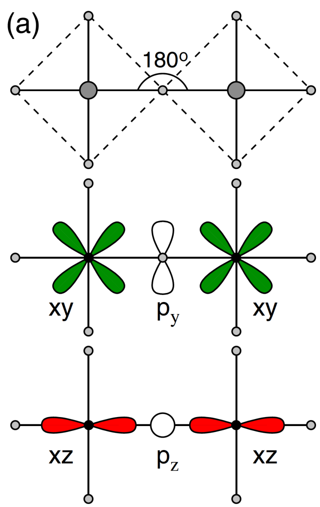
Goodenough-Kanamori theory on J_{eff}=1/2 states

J_{eff}=1/2 states



$$|xy\uparrow\rangle + |yz\downarrow\rangle + i|zx\downarrow\rangle$$

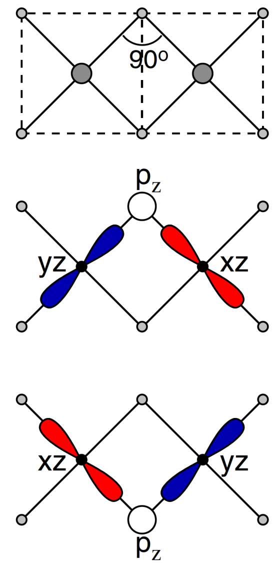
180° bond



Heisenberg

High-Tc superconductivity?

90° bond



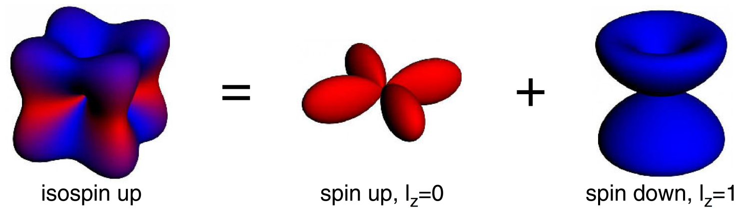
complete suppression of Heisenberg

Kitaev spin liquid?

G. Jackeli and G. Khaliullin PRL 2009

Goodenough-Kanamori theory on J_{eff}=1/2 states

J_{eff}=1/2 states

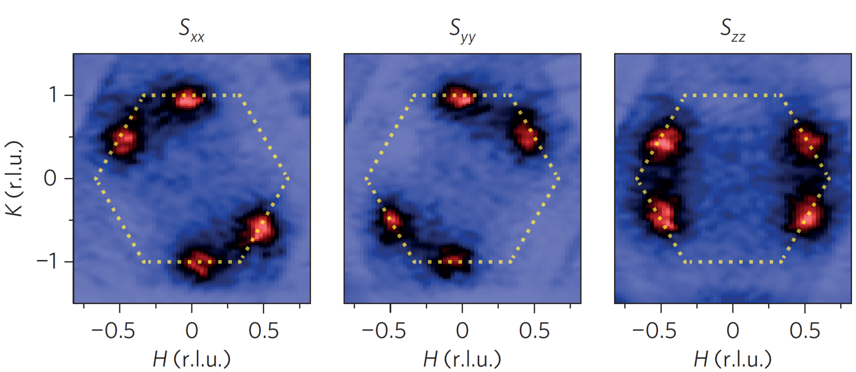


$$|xy \uparrow\rangle + |yz \downarrow\rangle + i|zx \downarrow\rangle$$

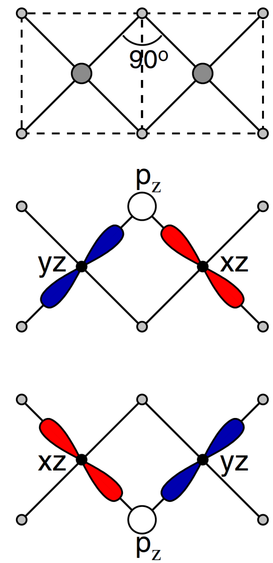
nature physics
LETTERS
PUBLISHED ONLINE: 11 MAY 2015 | DOI: 10.1038/NPHYS3322

Direct evidence for dominant bond-directional interactions in a honeycomb lattice iridate Na_2IrO_3

Sae Hwan Chun[1], Jong-Woo Kim[2], Jungho Kim[2], H. Zheng[1], Constantinos C. Stoumpos[1], C. D. Malliakas[1], J. F. Mitchell[1], Kavita Mehlawat[3], Yogesh Singh[3], Y. Choi[2], T. Gog[2], A. Al-Zein[4], M. Moretti Sala[4], M. Krisch[4], J. Chaloupka[5], G. Jackeli[6,7], G. Khaliullin[6] and B. J. Kim[6]*



90° bond



complete suppression of Heisenberg

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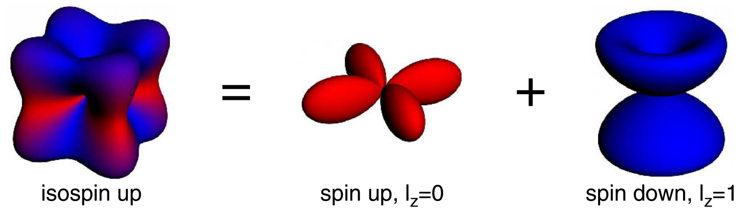
G. Jackeli and G. Khaliullin PRL 2009

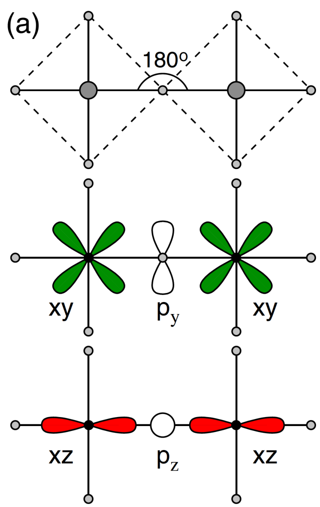
Goodenough-Kanamori theory on J_{eff}=1/2 states

J_{eff}=1/2 states



$$|xy \uparrow\rangle + |yz \downarrow\rangle + i|zx \downarrow\rangle$$

180° bond



Heisenberg

High-Tc superconductivity?

SUPERCONDUCTIVITY

Fermi arcs in a doped pseudospin-1/2 Heisenberg antiferromagnet

Y. K. Kim,[1] O. Krupin,[1] J. D. Denlinger,[1] A. Bostwick,[1] E. Rotenberg,[1] Q. Zhao,[2] J. F. Mitchell,[2] J. W. Allen,[3] B. J. Kim[2,3,4*]

arXiv.org > cond-mat > arXiv:1506.06639

Search

Condensed Matter > Strongly Correlated Electrons

Observation of a d-wave gap in electron-doped Sr_2IrO_4

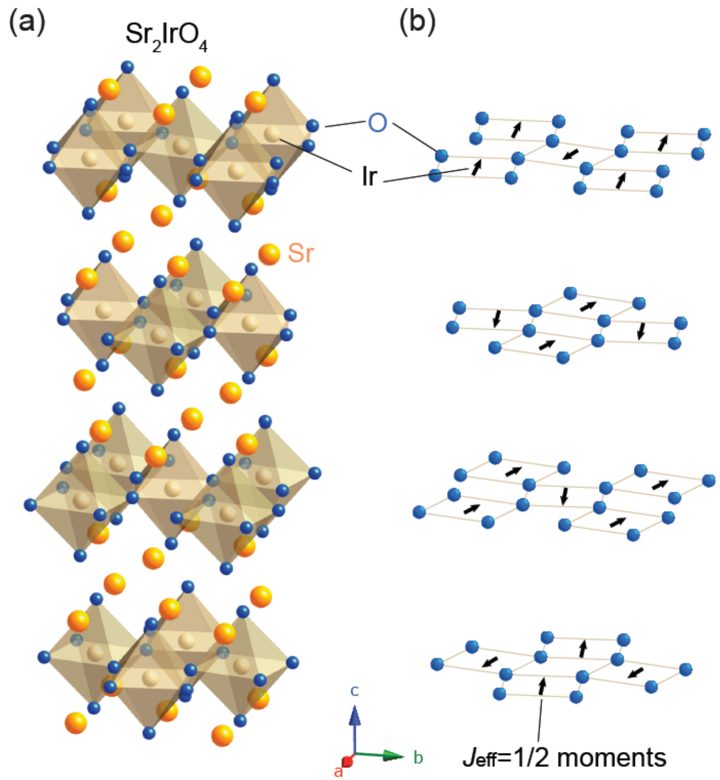
Y. K. Kim, N. H. Sung, J. D. Denlinger, B. J. Kim

(Submitted on 22 Jun 2015 (v1), last revised 25 Jun 2015 (this version, v2))

High temperature superconductivity in cuprates emerges out of a highly enigmatic `pseudogap' metal phase. The mechanism of high temperature superconductivity is likely encrypted in the elusive relationship between the two phases, which spectroscopically is manifested as Fermi arcs---disconnected segments of zero-energy states---collapsing into d-wave point nodes upon entering the superconducting phase. Here, we reproduce this distinct cuprate phenomenology in the 5d transition-metal oxide Sr_2IrO_4. Using angle-resolved photoemission, we show that clean, low-temperature phase of 6-8% electron-doped Sr_2IrO_4 has gapless excitations only at four isolated points in the Brillouin zone with a predominant d-wave symmetry of the gap. Our work thus establishes a connection between the low-temperature d-wave instability and the previously reported high-temperature Fermi arcs in electron-doped Sr_2IrO_4. Although the physical origin of the d-wave gap remains to be understood, Sr_2IrO_4 is a first non-cuprate material to spectroscopically reproduce the complete phenomenology of the cuprates, thus off?ering a new material platform to investigate the relationship between the pseudogap and the d-wave gap.

Ruddelsden-Popper Series Iridates

Sr_2IrO_4 T_N=240 K

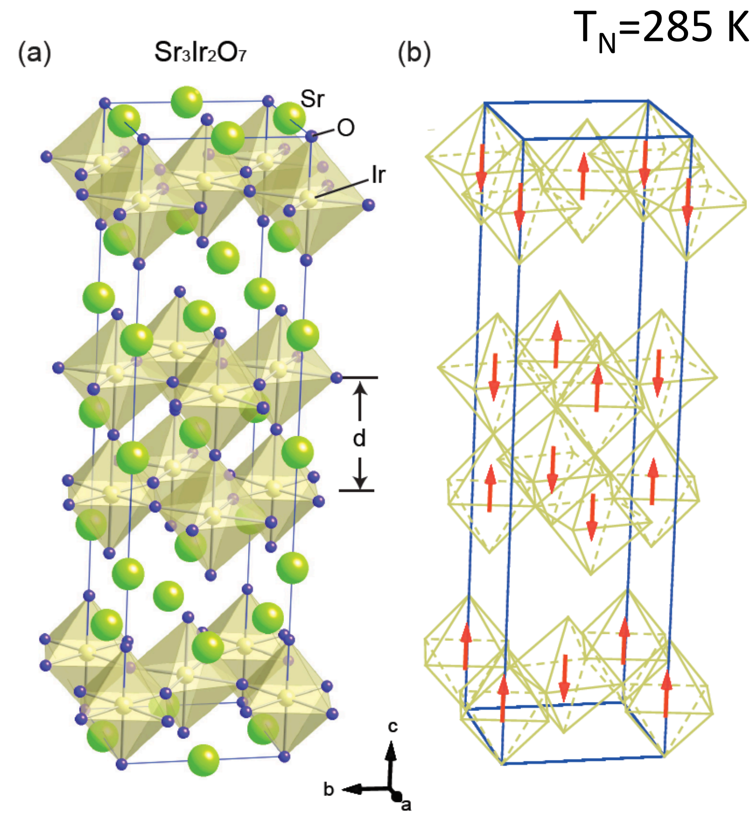


canted AF with
in-plane moments

B. J. Kim et al. Science (2009)

$Sr_3Ir_2O_7$

T_N=285 K



G-type AF
c-axis collinear

J. -W. Kim, BJK et al. PRL (2012),
S. Boseggia et al, J. Phys. Condens. Matter (2015)
J. P. Clancy et al., arxiv (2012)
S. Fujiyama et al. PRB (2012)

Resonant x-ray scattering

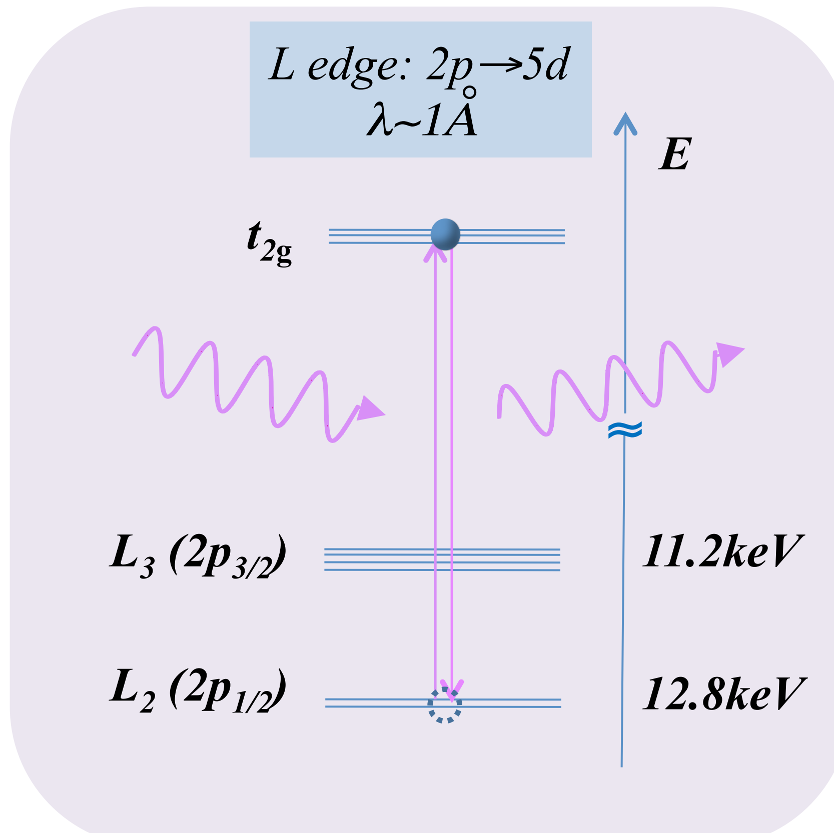
Direct probe for the 5d state responsible for magnetism

$$H' = \frac{e^2}{2m_e c^2}\sum_i A(\boldsymbol{r}_i)^2 - \frac{e^2\hbar}{2m_e^2 c^4}\sum_i \boldsymbol{s}_i \cdot \left(\frac{\partial A}{\partial t} \times A\right) - \frac{e}{m_e c}\sum_i A(\boldsymbol{r}_i)\cdot \boldsymbol{p} - \frac{e^2\hbar}{2m_e^2 c^4}\sum_i \boldsymbol{s}_i \cdot (\nabla \times A(\boldsymbol{r}_i))$$

$$F_{E1}^{(reso)} = \frac{e^2}{m_e c^2}\sum_{\boldsymbol{n},\boldsymbol{m}} e^{i\boldsymbol{K}\bullet(\boldsymbol{n}+\boldsymbol{d}_m)} \sum_{\alpha\beta} \varepsilon'_\alpha \varepsilon_\beta f_{\boldsymbol{m}}^{\alpha\beta}$$

$$f_m^{\alpha\beta} = \sum_c \frac{m_e \omega_{ca}^3}{\omega} \frac{\langle a|R_m^\alpha|c\rangle\langle c|R_m^\beta|a\rangle}{\hbar\omega - \hbar\omega_{ac} + i\Gamma/2}$$

Cartesian tensor of rank 2



Resonant x-ray scattering and x-ray absorption

-Related through the optical theorem

$$
\begin{aligned}
F_{\varepsilon_{in}\varepsilon_{out}} &= F^{(0)}(\varepsilon_{in} \cdot \varepsilon^*_{out}) \\
&+ F^{(1)}(\varepsilon_{in} \times \varepsilon^*_{out} \cdot \hat{m}) \\
&+ F^{(2)}((\varepsilon^*_{out} \cdot \hat{m})(\varepsilon_{in} \cdot \hat{m}) - \frac{1}{3}(\varepsilon_{in} \cdot \varepsilon^*_{out}))
\end{aligned}
$$

charge

magnetic dipole

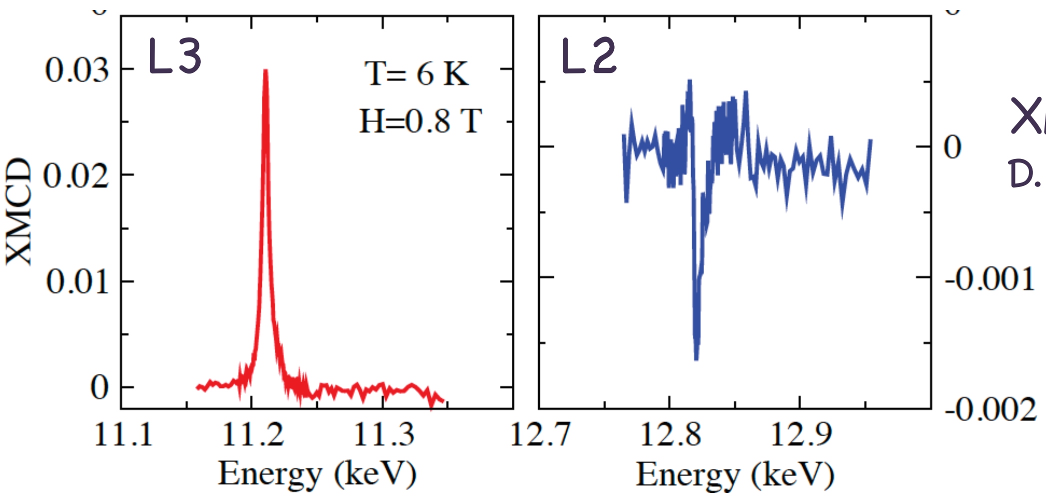
electric quadrupole

F0, describes the isotropic absorption
F1, describes the XMCD spectra
F2, describes the XMLD spectra

G. van der Laan, J. Phys. Soc. Jpn. 63, 2393 (1994)
M. Haverkort, PRL 105, 167404 (2010)

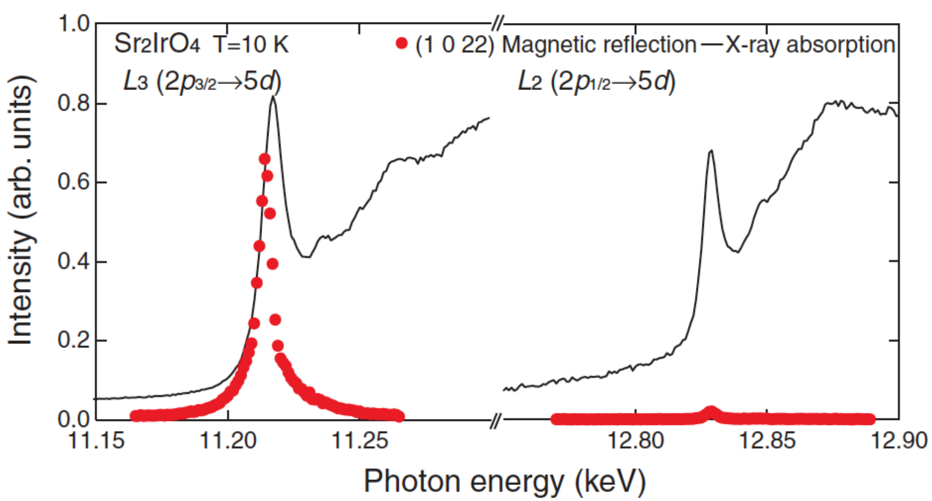
Resonant x-ray scattering contains information about spin and orbital structure of scattering ion

XMCD and RXD on Sr_2IrO_4



XMCD

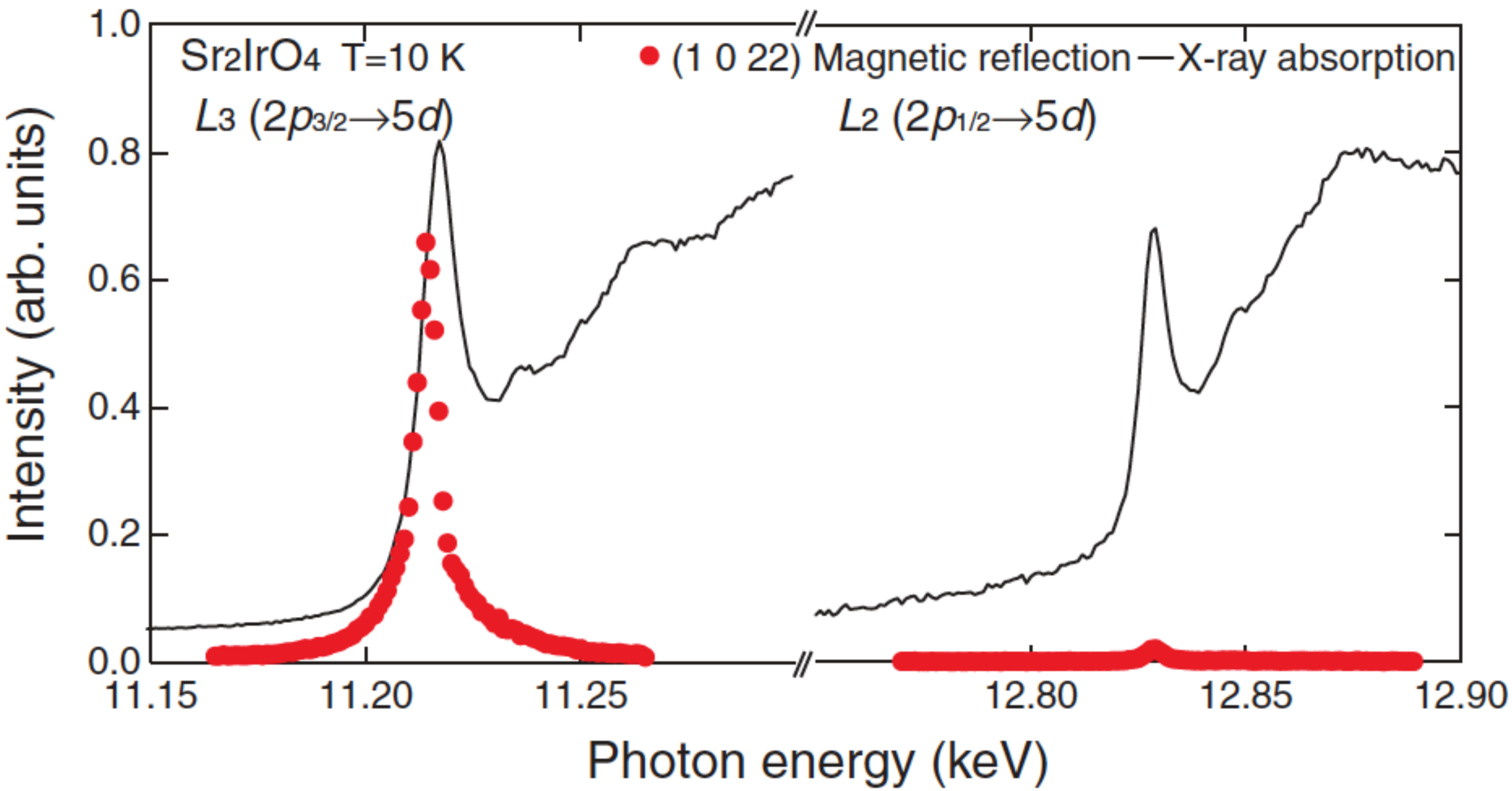
D. Haskel et al. PRL (2012)



RXD

B. J. Kim et al. Science (2009)

Evidence for J_{eff}=1/2 states



Almost no resonance at L_2:

L_2 scattering intensity is only about 1% of that of the L_3.

B. J. Kim et al., Science **323**, 1329 (2009)

RIXS cross section

To calculate $$O^{\alpha\beta}_{l_z,l_{z'}} = \sum_{j_z} \langle l, l_z | R^\alpha | j, j_z \rangle \langle j, j_z | R^\beta | l, l_{z'} \rangle$$

we need $\langle l, l_z | T^k_q | j, j_{z'} \rangle$ → Spherical tensor of rank 1

$$z = T^1_0$$

$$x = -\frac{1}{\sqrt{2}}\left(T^1_1 - T^1_{-1}\right)$$

$$y = \frac{1}{\sqrt{2}i}\left(T^1_1 + T^1_{-1}\right)$$

$$\langle l = 2, s = 1/2; l_z, s_z | T^k_q | l' = 1, s' = 1/2; j, j_{z'} \rangle$$

$$= \sum_{l'_z, s'_z} \langle l = 2, s = 1/2; l_z, s_z | T^k_q | l' = 1, s' = 1/2; l'_z, s'_z \rangle \underline{\langle l' = 1, s' = 1/2; l'_z, s'_z | j, j_z \rangle}$$

Clebsch-Gordan Coefficient

$$= \sum_{l'_z, s'_z} \langle l, s; l_z, s_z | T^k_q | l', s'; l'_z, s'_z \rangle \delta_{s_z s'_z} \langle l', s'; l'_z, s'_z | j, j_z \rangle$$

Wigner-Eckart theorem

$$\propto \sum_{l'_z} \langle l, k; l'_z, q | j, j_z \rangle \langle l', s'; l'_z, s_z | j, j_z \rangle$$

$$\langle l, l_z | T^k_q | l', l'_z \rangle = \langle l, k; l'_z, q \mid l, k; l, l_z \rangle \frac{\langle l' \parallel T^k \parallel l \rangle}{\sqrt{2l+1}}$$

$J_{\mathrm{eff}}=1/2 \xrightarrow[\text{Yes!}]{} \text{Experiment}$

$\text{Experiment} \xrightarrow{?} J_{\mathrm{eff}}=1/2$

IOP FTC
IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER
J. Phys.: Condens. Matter **23** (2011) 252201 (5pp) doi:10.1088/0953-8984/23/25/252201

FAST TRACK COMMUNICATION

The magnetic motif and the wavefunction of Kramers ions in strontium iridate (Sr_2IrO_4)

L C Chapon[1] and S W Lovesey[1,2]

PRL **112,** 026403 (2014) PHYSICAL REVIEW LETTERS week ending 17 JANUARY 2014

Resonant X-Ray Scattering and the $j_{\mathrm{eff}} = 1/2$ Electronic Ground State in Iridate Perovskites

M. Moretti Sala,[1,*] S. Boseggia,[2,3] D. F. McMorrow,[2,4] and G. Monaco[1]

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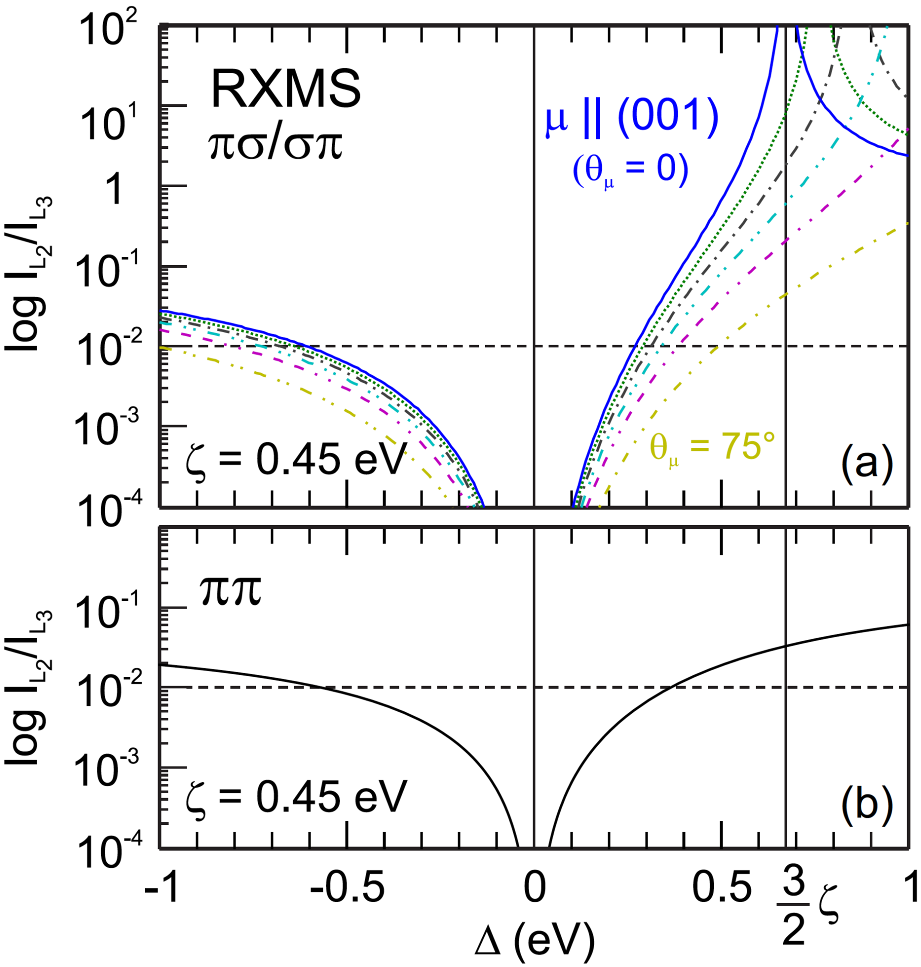

- valid only if M is along the axis of distortion (c-axis)
- L2 intensity vanishes for any Δ if M is in the ab-plane

PRL 112, 026403 (2014) PHYSICAL REVIEW LETTERS week ending 17 JANUARY 2014

Resonant X-Ray Scattering and the $j_{\mathrm{eff}} = 1/2$ Electronic Ground State in Iridate Perovskites

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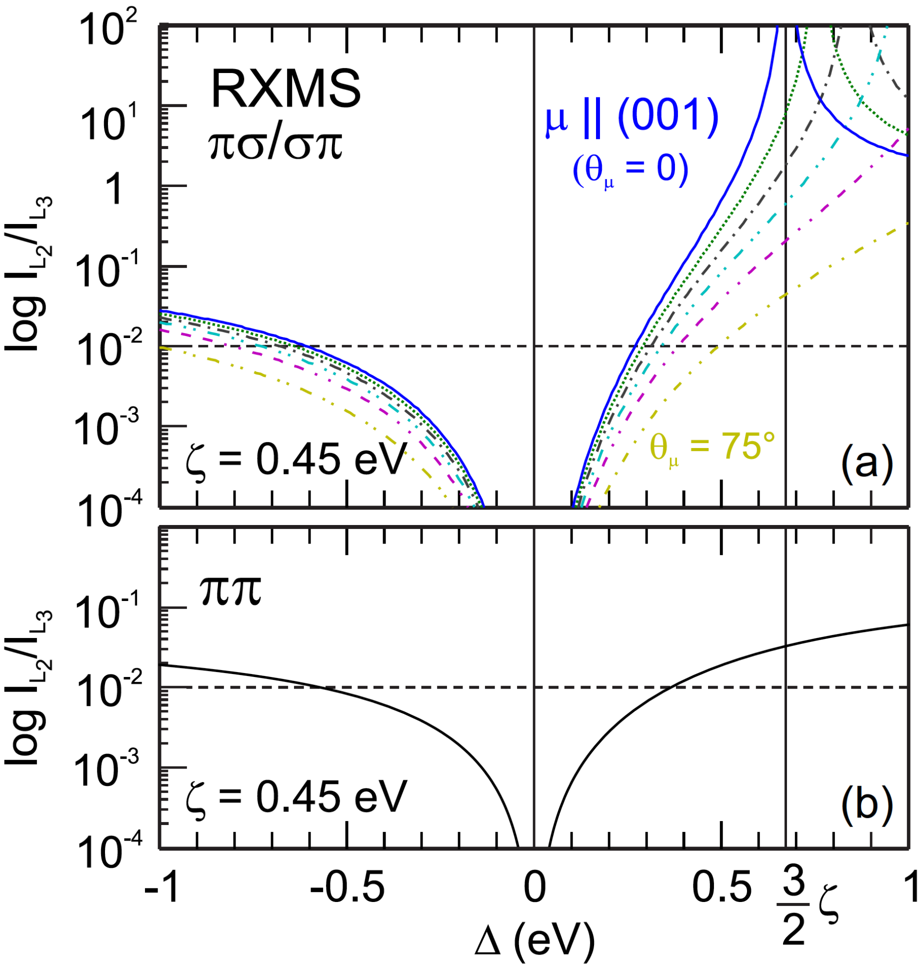
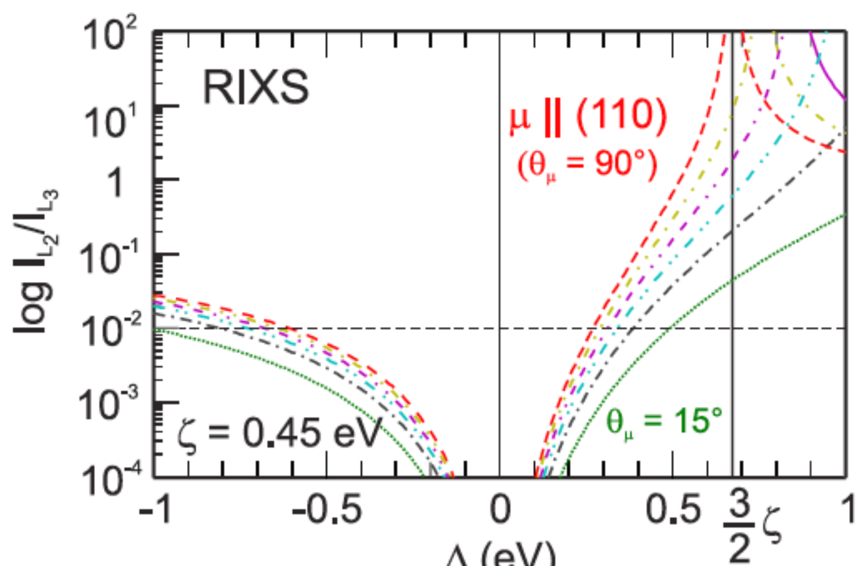
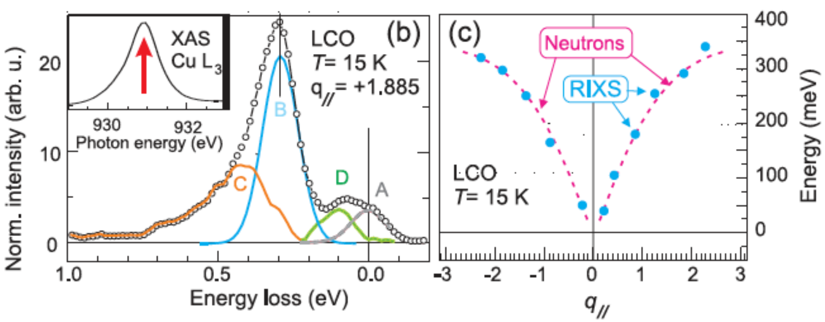


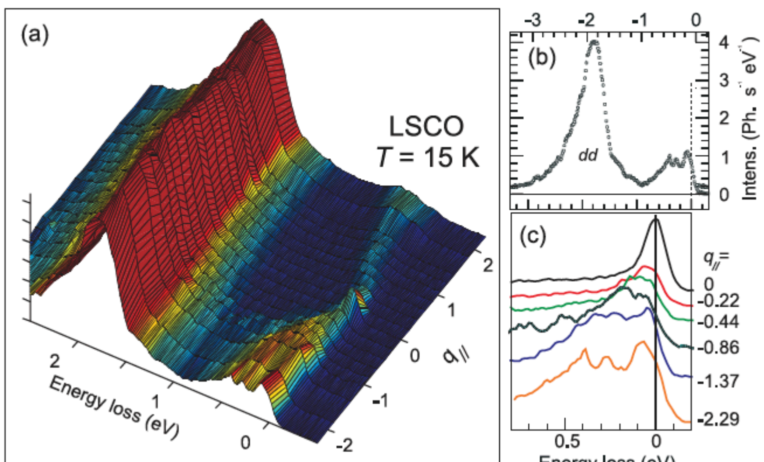


RIXS: New Tool to study Magnetic Excitations

soft x-ray (<1 keV)

RIXS experiment on thin film of La_2CuO_4





L. Braicovich et al., PRL (2010)

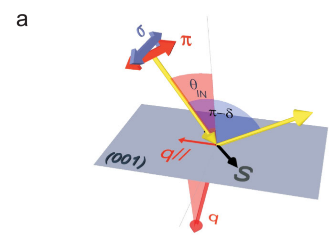
PRL 103, 117003 (2009) PHYSICAL REVIEW LETTERS week ending 11 SEPTEMBER 2009

Theoretical Demonstration of How the Dispersion of Magnetic Excitations in Cuprate Compounds can be Determined Using Resonant Inelastic X-Ray Scattering

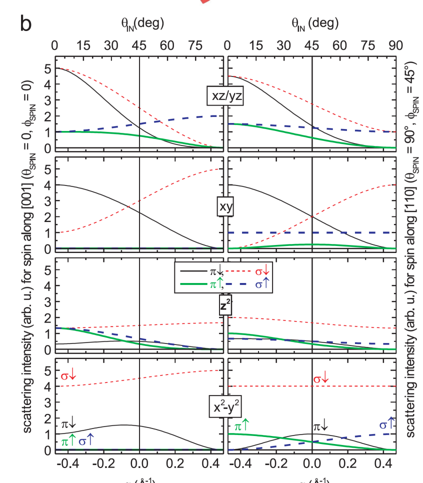
Luuk J. P. Ament,[1,4] Giacomo Ghiringhelli,[2] Marco Moretti Sala,[2] Lucio Braicovich,[2] and Jeroen van den Brink[1,3,4]
[1]Institute-Lorentz for Theoretical Physics, Universiteit Leiden, 2300 RA Leiden, The Netherlands
[2]INFM/CNR Coherentia and Soft-Dipartimento di Fisica, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy
[3]Institute for Molecules and Materials, Radboud Universiteit, 6500 GL Nijmegen, The Netherlands
[4]Stanford Institute for Materials and Energy Sciences, Stanford University and SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA
(Received 13 March 2009; published 11 September 2009)

We show that in resonant inelastic x-ray scattering (RIXS) at the copper *L* and *M* edge direct spin-flip scattering is in principle allowed. We demonstrate how this possibility can be exploited to probe the dispersion of magnetic excitations, for instance magnons, of cuprates such as the high T_c superconductors. We compute the relevant local and momentum dependent magnetic scattering amplitudes, which we compare to the elastic and *dd*-excitation scattering intensities. For cuprates these theoretical results put RIXS as a technique on the same footing as neutron scattering.

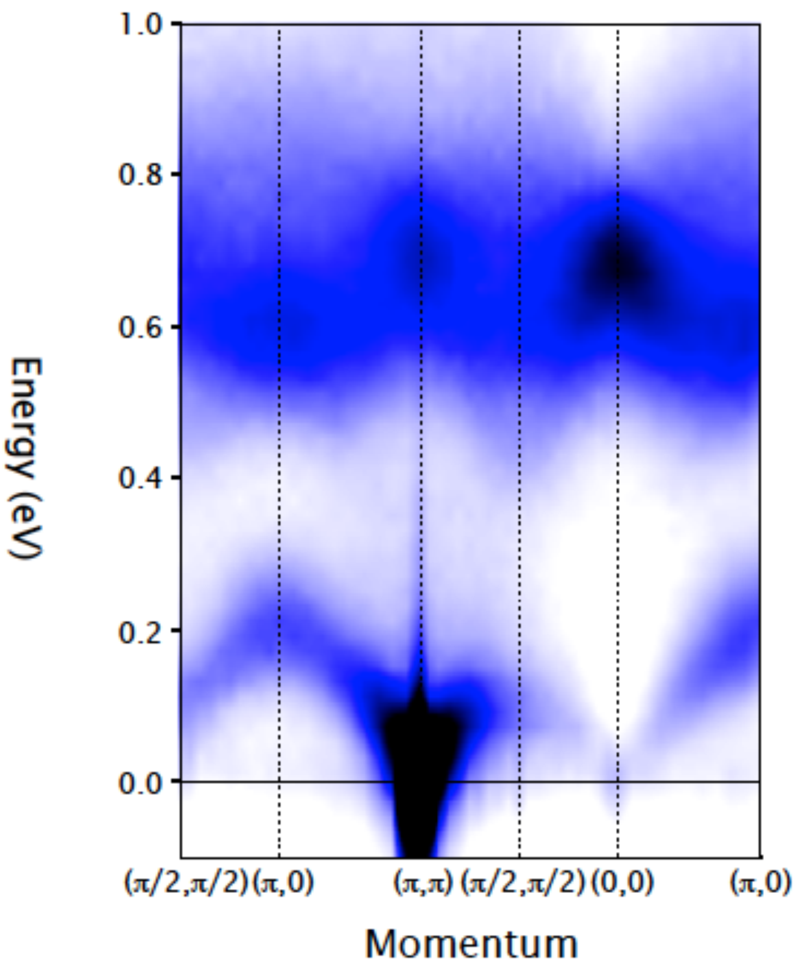
a



b



RIXS: New Tool to study Magnetic Excitations



5d TMO   hard x-ray (~11 keV)

2010

our first experiment on iridate using Si (844) analyzer @ APS

130 meV, 3000 cps

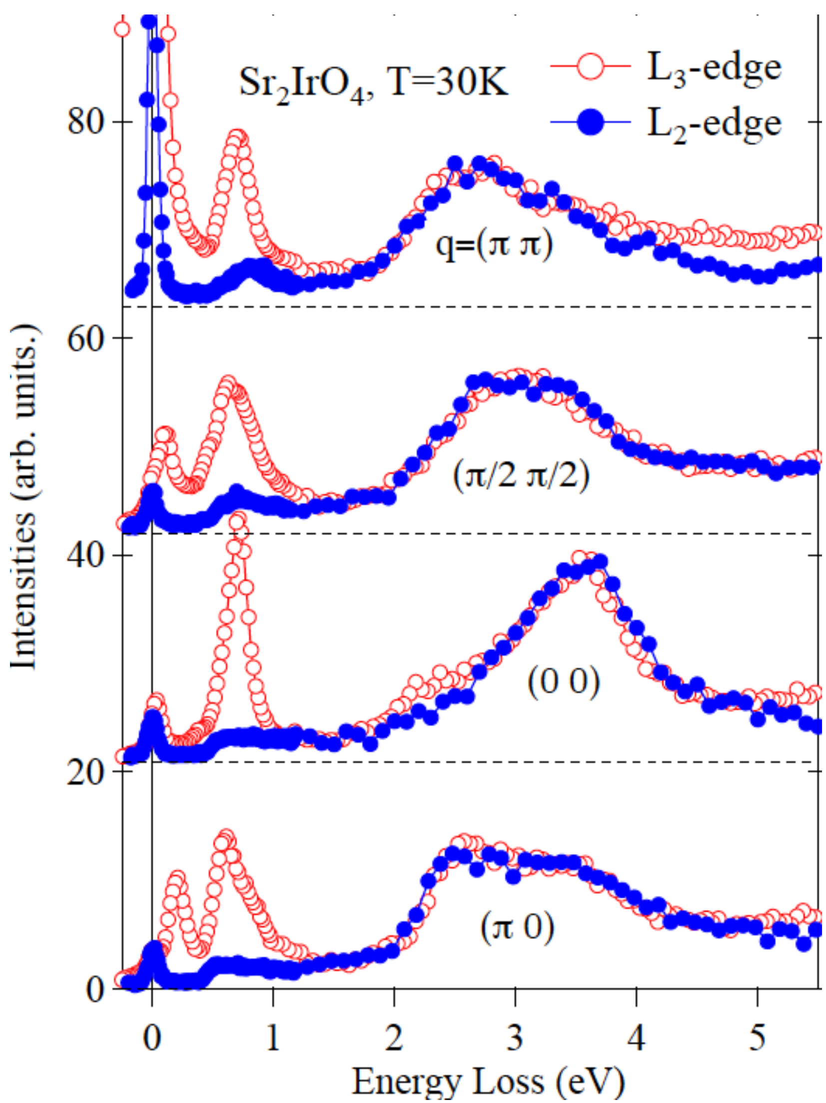
now

25 meV resolution routinely available at APS and ESRF

2016

sub 10-meV targeted in coming years

L2/L3 intensity ratio in RIXS

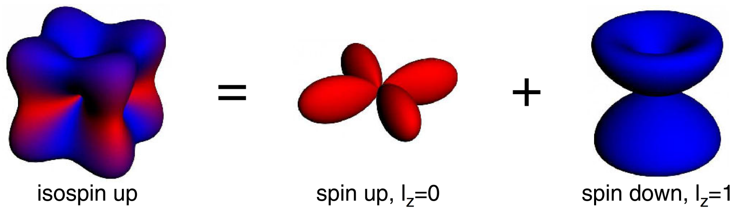


magnon at L2 edge suppressed below detection level!

J. Kim, BJK et al. unpublished

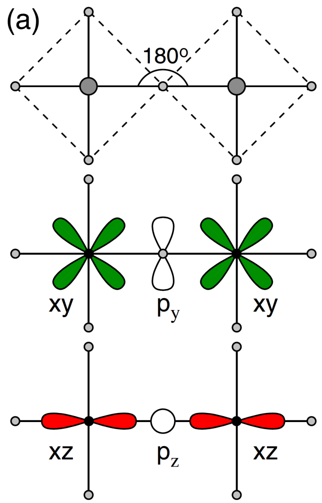
Goodenough-Kanamori theory on J_{eff}=1/2 states

J_{eff}=1/2 states



$$|xy\uparrow\rangle + |yz\downarrow\rangle + i|zx\downarrow\rangle$$

180° bond



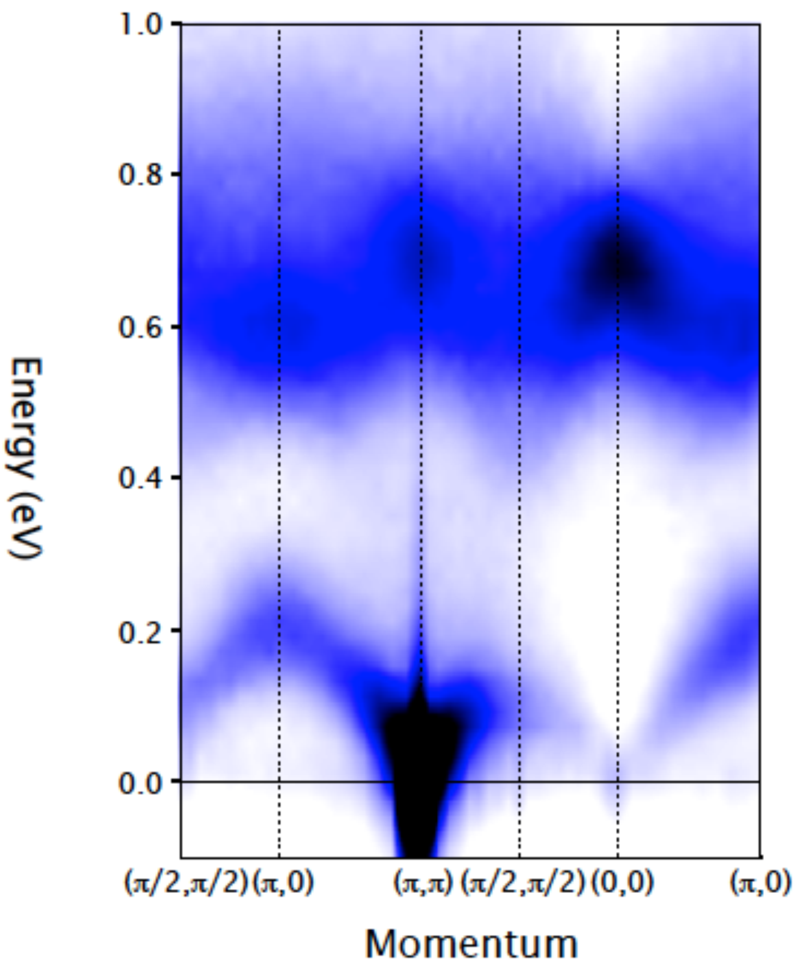
$$\mathcal{H}_{ij} = J_1 \vec{S}_i \cdot \vec{S}_j + J_2(\vec{S}_i \cdot \vec{r}_{ij})(\vec{r}_{ij} \cdot \vec{S}_j),$$

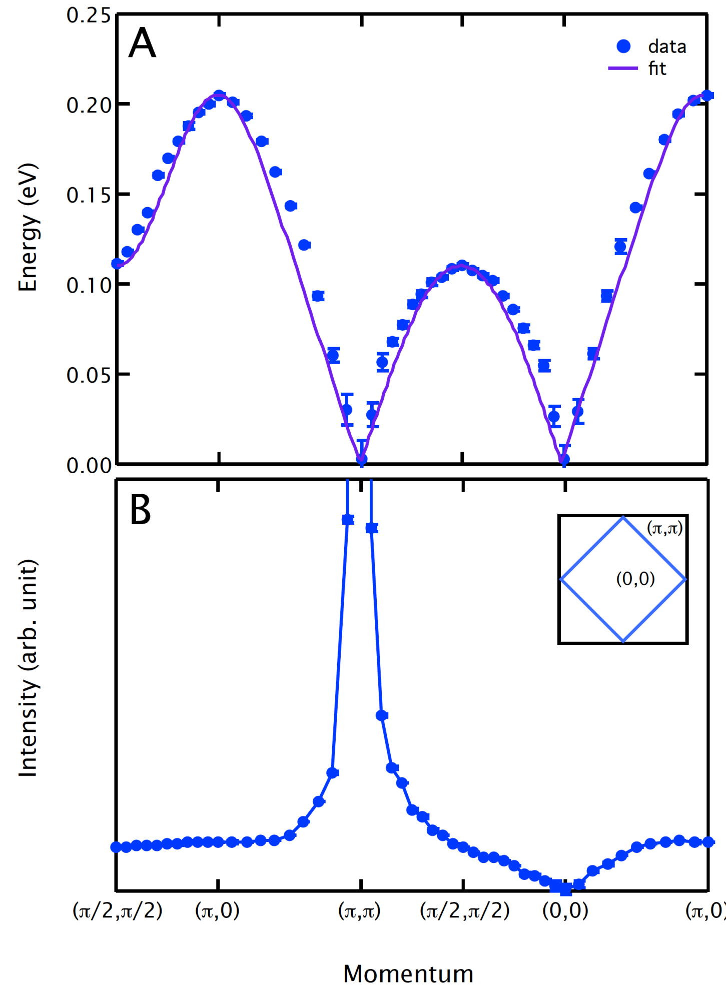
Predominantly
Heisenberg-like
J1 >> J2

G. Jackeli and G. Khaliullin PRL 2009

Spin wave- fitting to Heisenberg model

2010

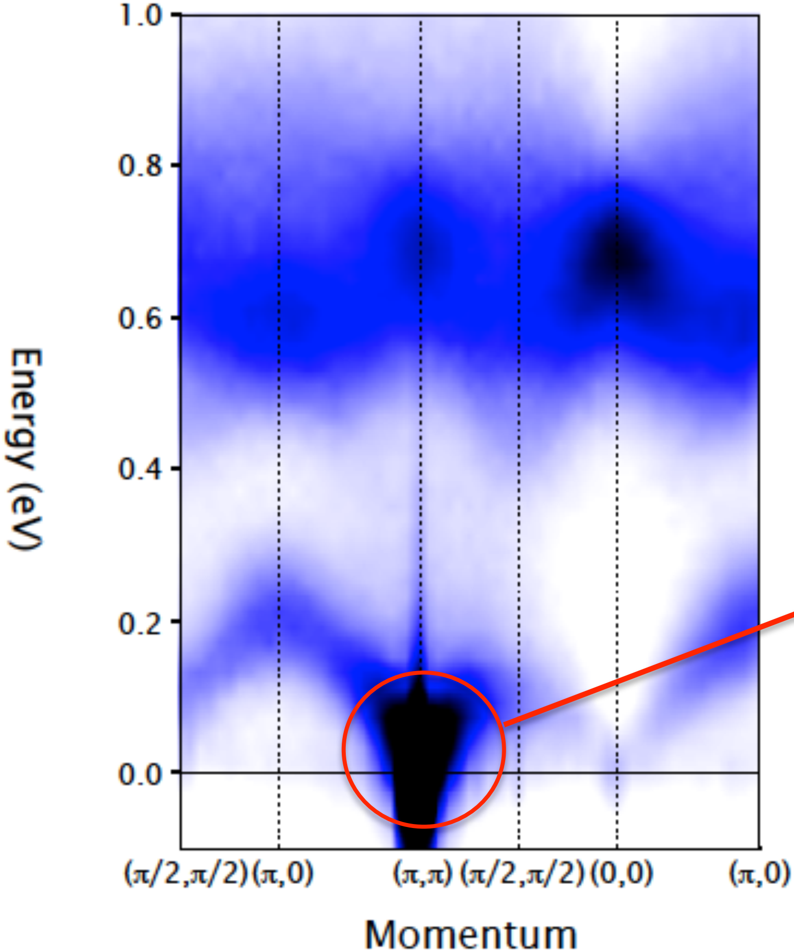




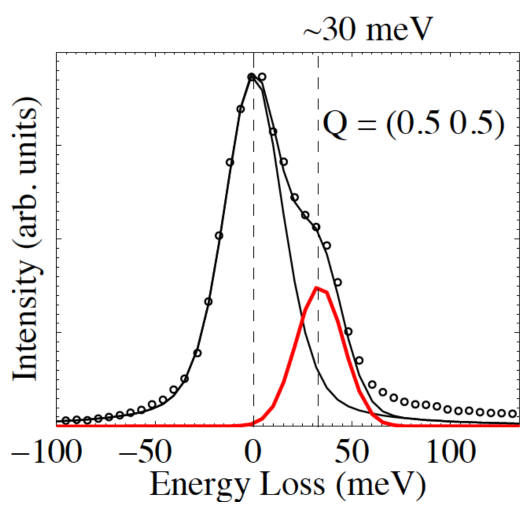
J=60
J′=-20
J″=15

Spin wave gap

2010



2015

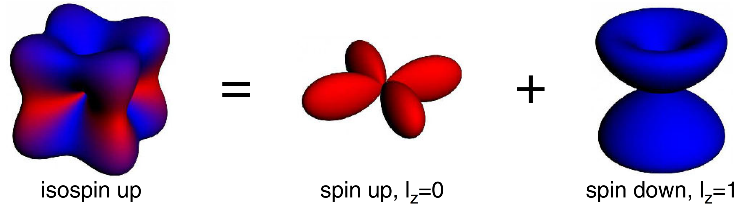


Sizable XY anisotropy!

see also J. Vale et al. PRB (2015)

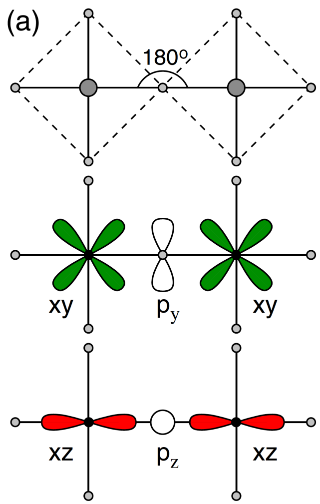
Goodenough-Kanamori theory on J_{eff}=1/2 states

J_{eff}=1/2 states



$$|xy\uparrow\rangle + |yz\downarrow\rangle + i|zx\downarrow\rangle$$

180° bond



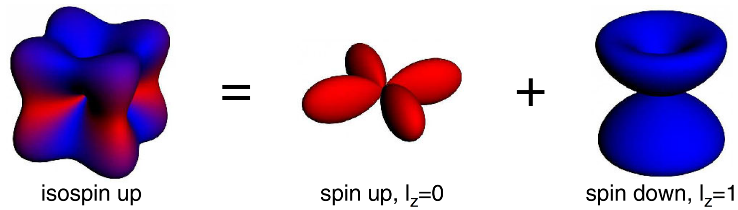
$$\mathcal{H}_{ij} = J_1 \vec{S}_i \cdot \vec{S}_j + J_2(\vec{S}_i \cdot \vec{r}_{ij})(\vec{r}_{ij} \cdot \vec{S}_j),$$

-Compass
-bond-directional
-Pseudodipolar

G. Jackeli and G. Khaliullin PRL 2009

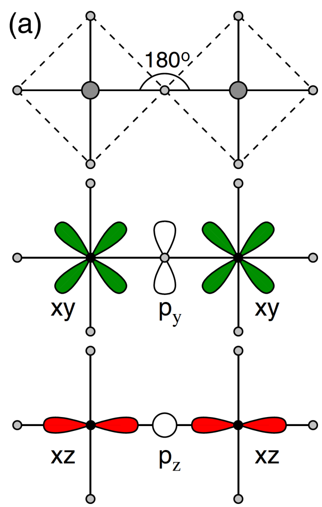
Goodenough-Kanamori theory on J_{eff}=1/2 states

J_{eff}=1/2 states



$$|xy\uparrow\rangle + |yz\downarrow\rangle + i|zx\downarrow\rangle$$

180° bond



$$\mathcal{H}_{ij} = J_1 \vec{S}_i \cdot \vec{S}_j + J_2(\vec{S}_i \cdot \vec{r}_{ij})(\vec{r}_{ij} \cdot \vec{S}_j),$$

-Compass
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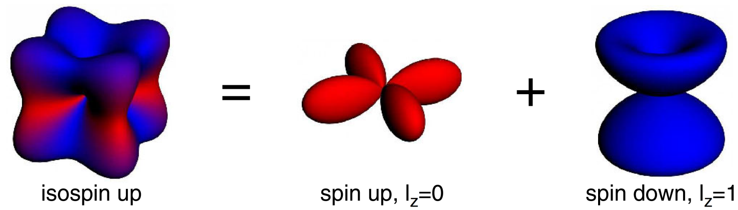
G. Jackeli and G. Khaliullin PRL 2009

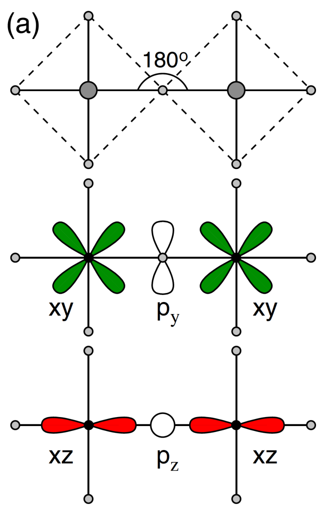
Goodenough-Kanamori theory on J_{eff}=1/2 states

J_{eff}=1/2 states



$$|xy\uparrow\rangle + |yz\downarrow\rangle + i|zx\downarrow\rangle$$

180° bond



$$\mathcal{H}_{ij} = J_1 \vec{S}_i \cdot \vec{S}_j + J_2(\vec{S}_i \cdot \vec{r}_{ij})(\vec{r}_{ij} \cdot \vec{S}_j),$$

-Compass
-bond-directional
-Pseudodipolar

responsible for magnetic anisotropy and magnon gap

weak SOC limit: scales with λ
strong SOC limit: scales with J_H/U

G. Jackeli and G. Khaliullin PRL 2009

Pseudodipolar term in a square lattice

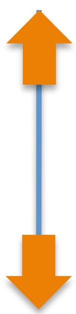
x

$$\mathcal{H}_{ij} = J_{ij}\vec{S}_i \cdot \vec{S}_j + \Gamma S_i^x S_j^x$$

$$\frac{\Gamma}{2}(S_i^x S_j^x + S_i^y S_j^y) + \frac{\Gamma}{2}(S_i^x S_j^x - S_i^y S_j^y)$$

A_{1g} $\qquad$ B_{1g}

y

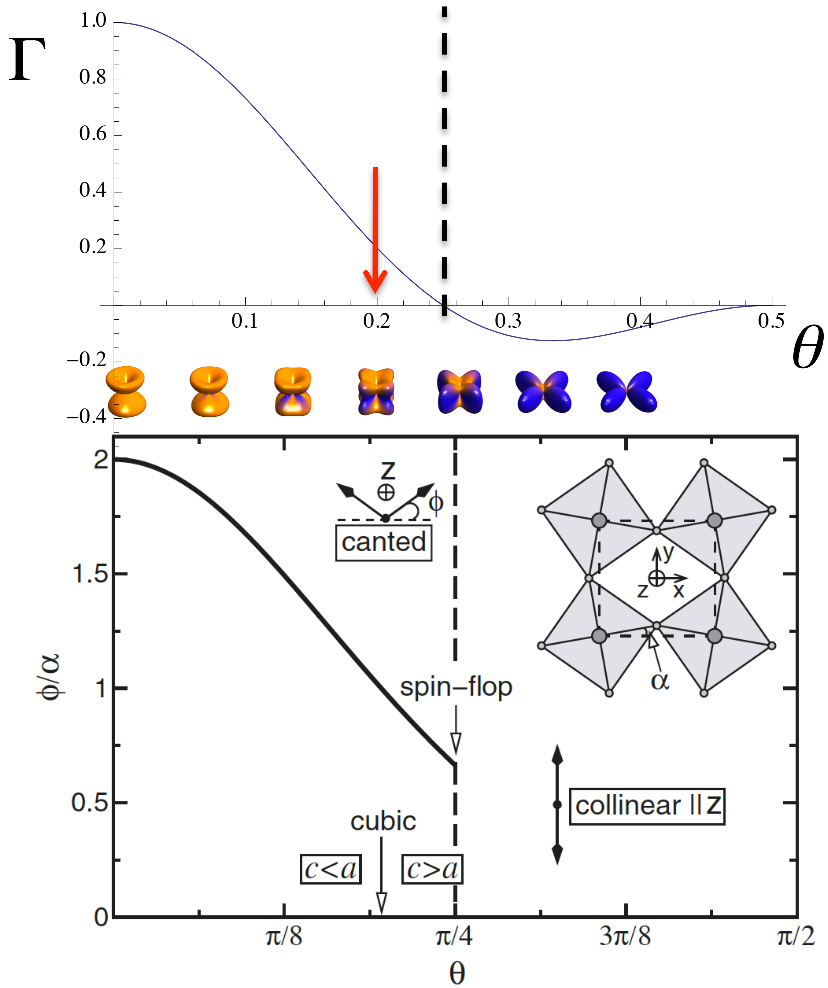


$$\mathcal{H}_{ij} = J_{ij}\vec{S}_i \cdot \vec{S}_j + \Gamma S_i^y S_j^y$$

$$\frac{\Gamma}{2}(S_i^x S_j^x + S_i^y S_j^y) - \frac{\Gamma}{2}(S_i^x S_j^x - S_i^y S_j^y)$$

$$\mathcal{H}_{ij} = (J_{ij} + \Gamma/2)\vec{S}_i \cdot \vec{S}_j - \frac{\Gamma}{2} S_i^z S_j^z$$

Pseudodipolar term in a square lattice

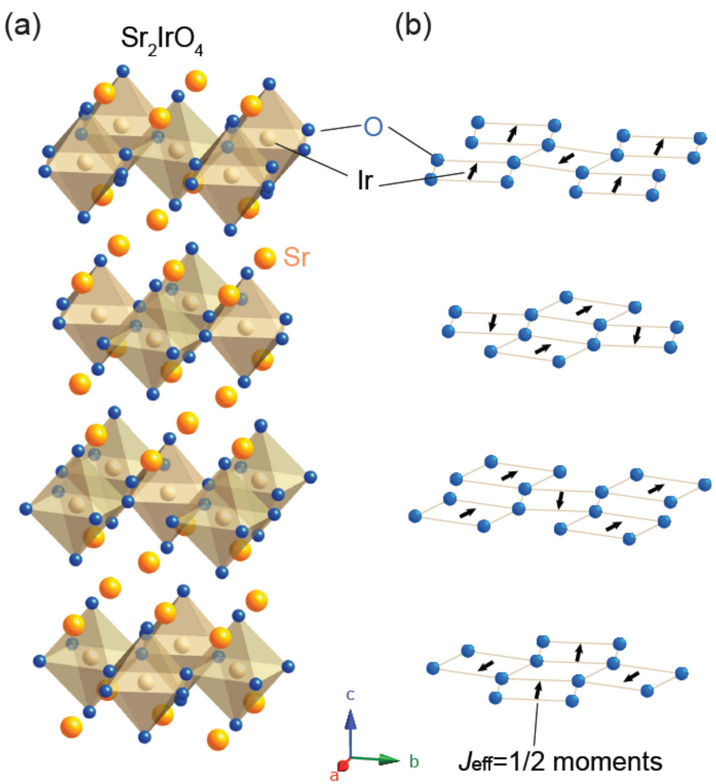


$$\mathcal{H}_{ij} = (J_{ij} + \Gamma/2)\vec{S}_i \cdot \vec{S}_j - \frac{\Gamma}{2} S_i^z S_j^z$$

G. Jackeli and G. Khaliullin PRL 2009

Ruddelsden-Popper Series Iridates

Sr_2IrO_4 T_N=240 K

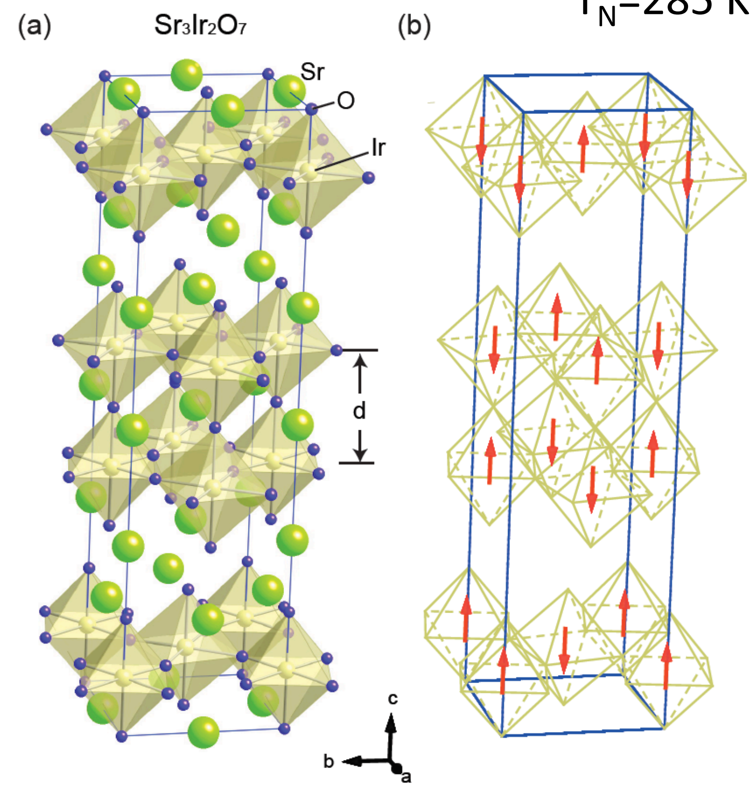


canted AF with
in-plane moments

B. J. Kim et al. Science (2009)

$Sr_3Ir_2O_7$

T_N=285 K

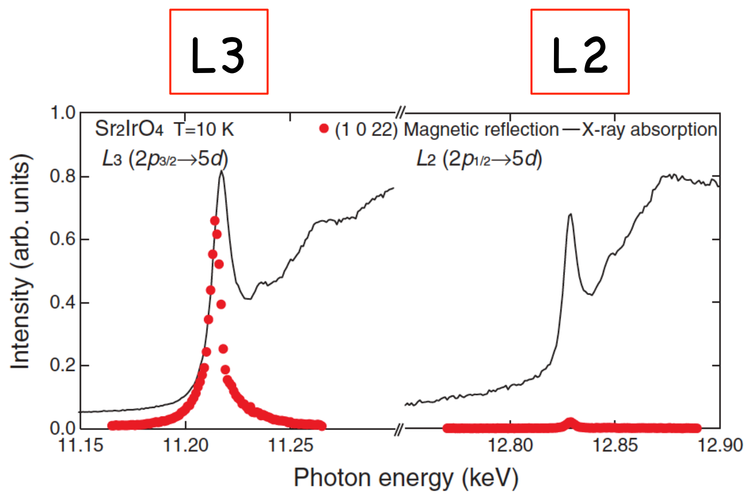


G-type AF
c-axis collinear

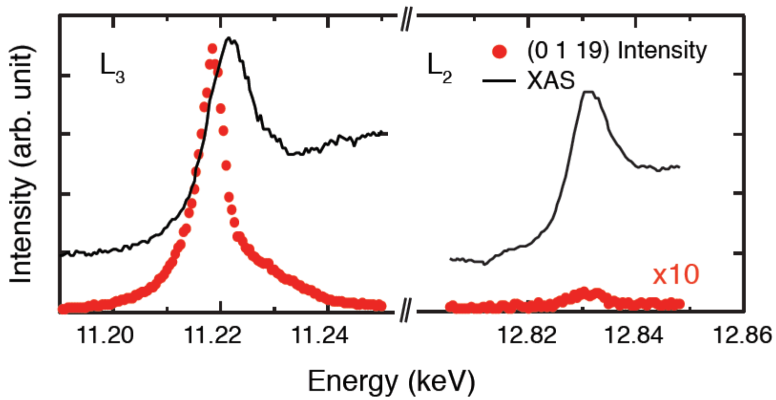
J. -W. Kim, BJK et al. PRL (2012),
S. Boseggia et al, J. Phys. Condens. Matter (2015)
J. P. Clancy et al., arxiv (2012)
S. Fujiyama et al. PRB (2012)

L2/L3 RXD Intensity

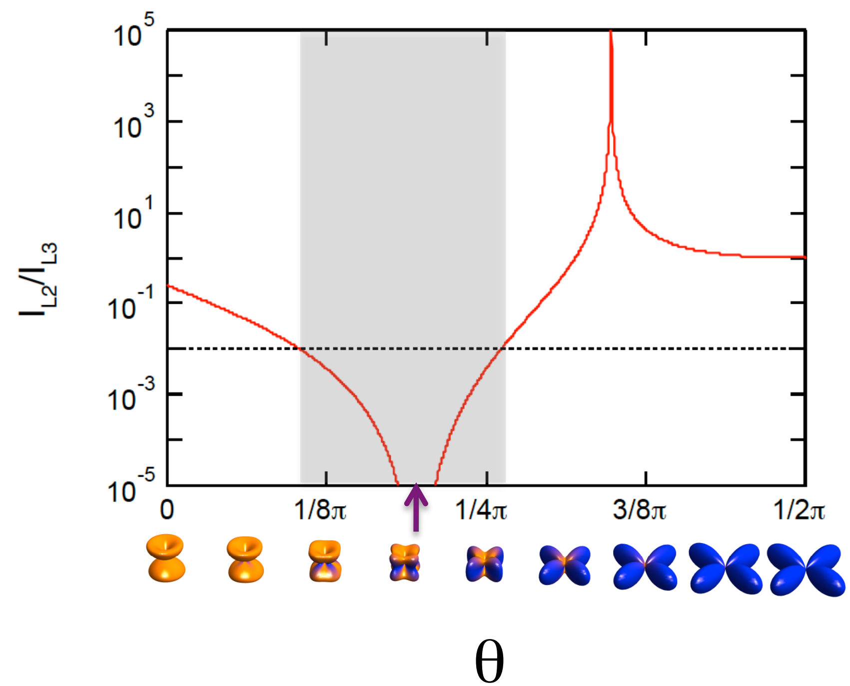
Sr_2IrO_4



$Sr_3Ir_2O_7$

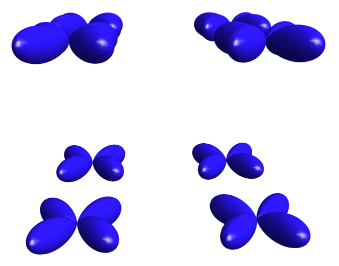


$I_{L2}/I_{L3} < 1\%$



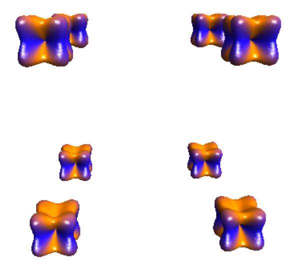
$$\mathcal{H}_{ij} = J_1 \vec{S}_i \cdot \vec{S}_j + J_2 (\vec{S}_i \cdot \vec{r}_{ij})(\vec{r}_{ij} \cdot \vec{S}_j),$$

Orbitals polarized, anisotropy in superexchange interactions



$J_{ab} >> J_{c,}$

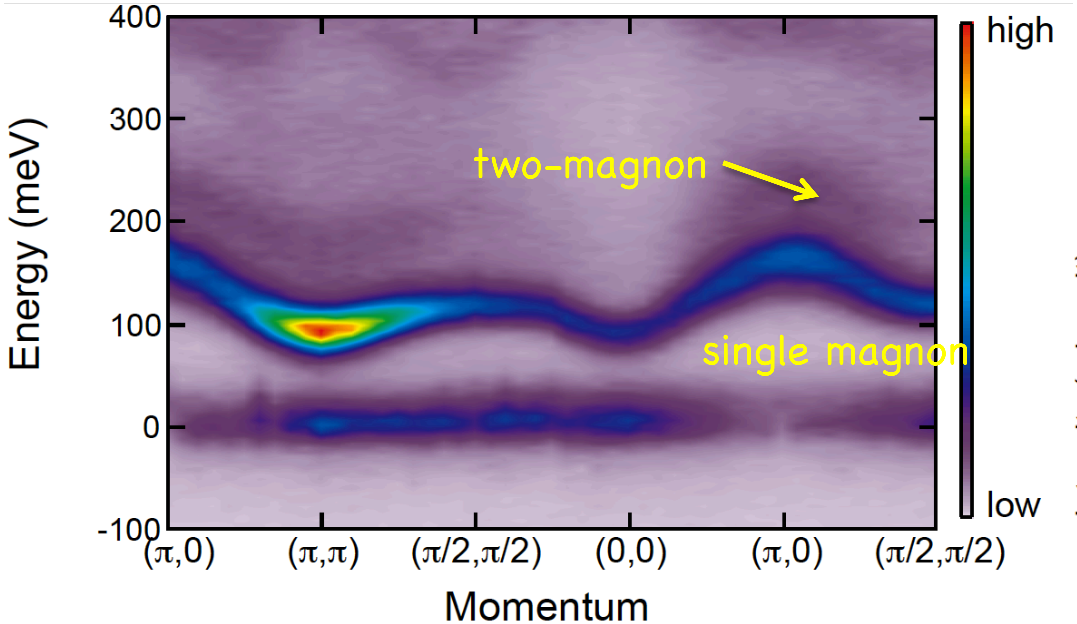
Superexchange interactions multidirectional

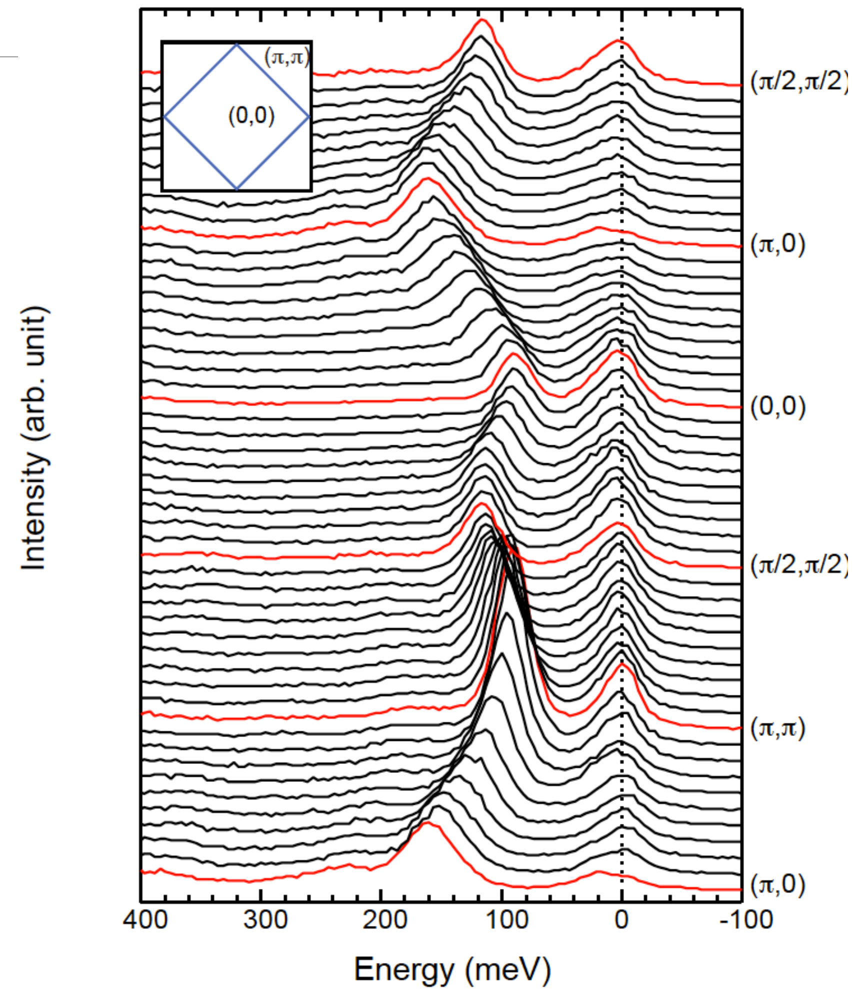


$J_{ab} \approx J_c$

Giant Magnon Gap in $Sr_3Ir_2O_7$

$q_c = \pi/3$





- Large magnon gap $\Delta_m \approx 90$ meV
- Total magnon bandwidth ≈ 70 meV

Jungho Kim et al. PRL (2012)

Optical spectroscopy on RP iridates

n=1: Sr_2IrO_4

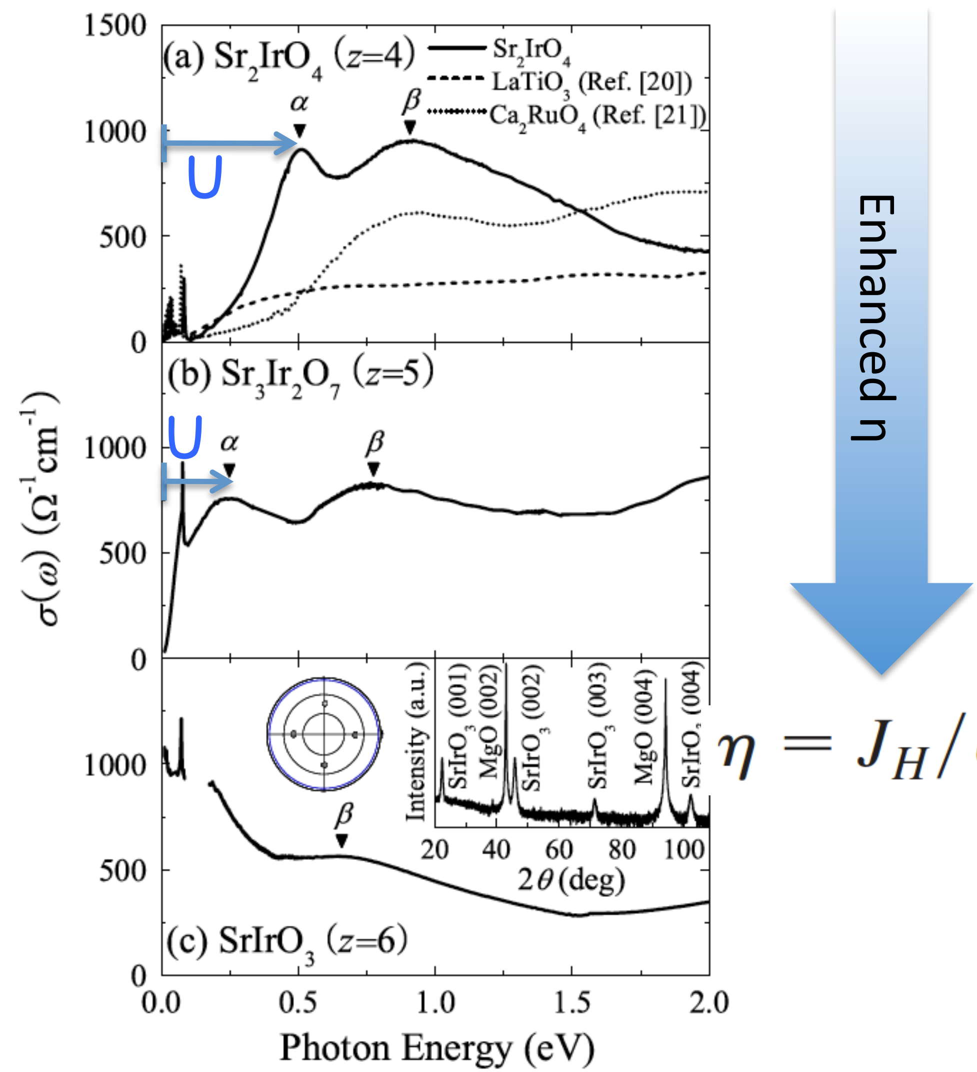
Δ_c=0.35 eV

n=2: $Sr_3Ir_2O_7$

Δ_c very small (unresolved)

n=∞: $SrIrO_3$

metal



$$\eta = J_H/U$$

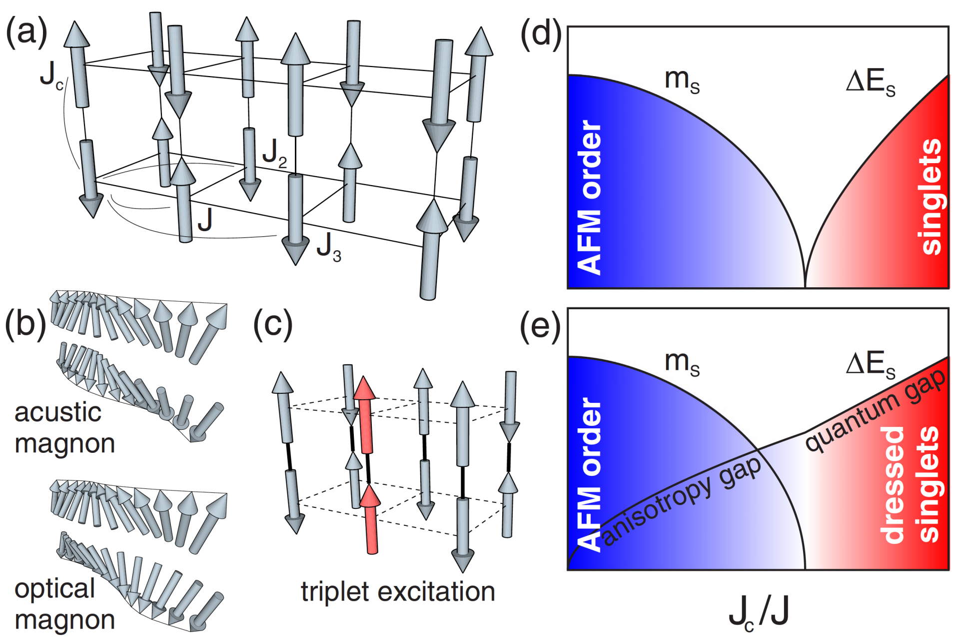
S. J Moon et al. PRL (2009)

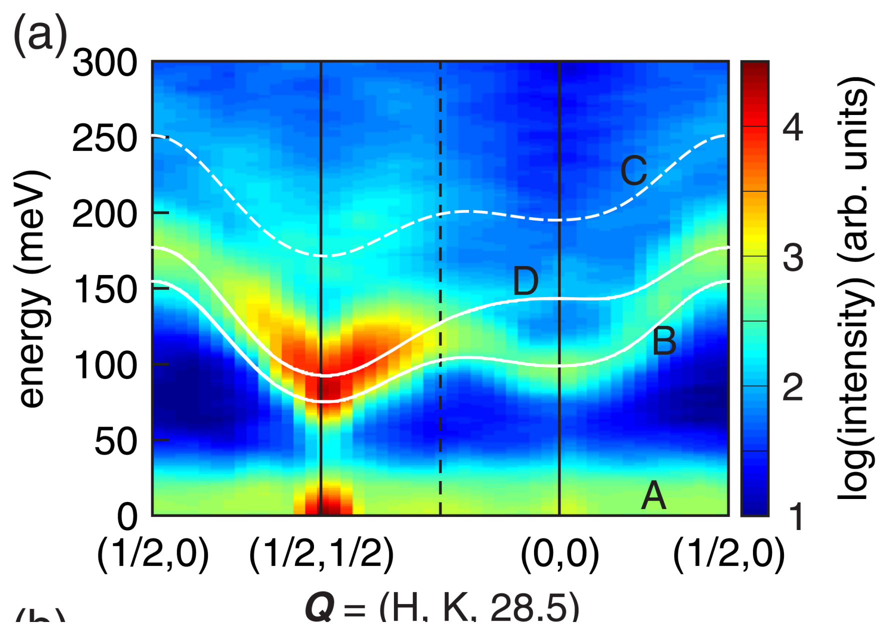
Alternative model

PHYSICAL REVIEW B **92**, 024405 (2015)

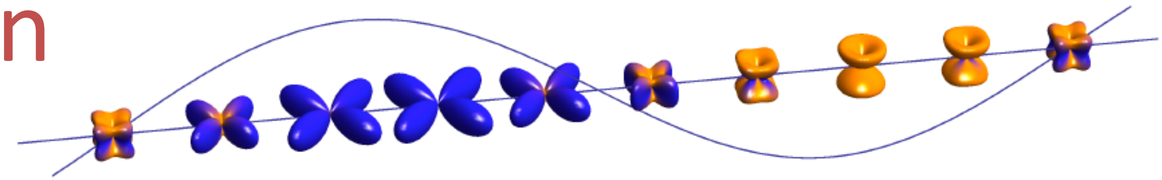
Evidence of quantum dimer excitations in $Sr_3Ir_2O_7$

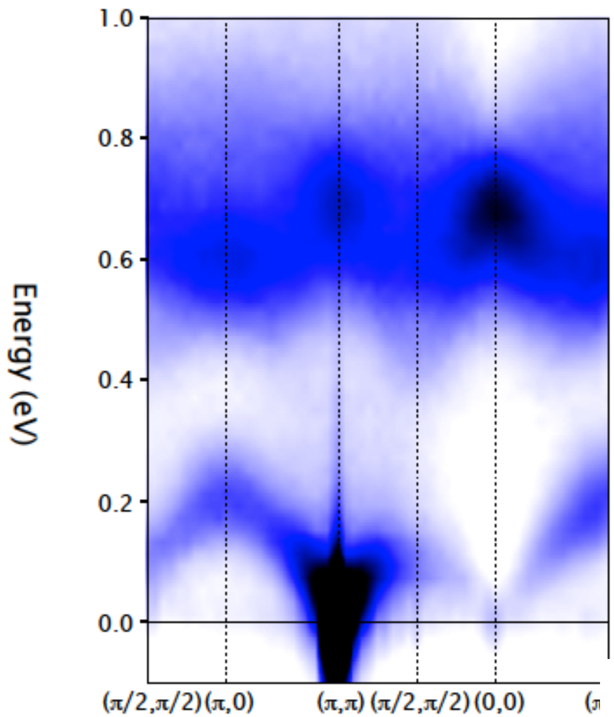
M. Moretti Sala,[1] V. Schnells,[2] S. Boseggia,[3,4] L. Simonelli,[1,5] A. Al-Zein,[1] J. G. Vale,[3] L. Paolasini,[1] E. C. Hunter,[6] R. S. Perry,[3] D. Prabhakaran,[7] A. T. Boothroyd,[7] M. Krisch,[1] G. Monaco,[1,8] H. M. Rønnow,[9,10] D. F. McMorrow,[3] and F. Mila[11]

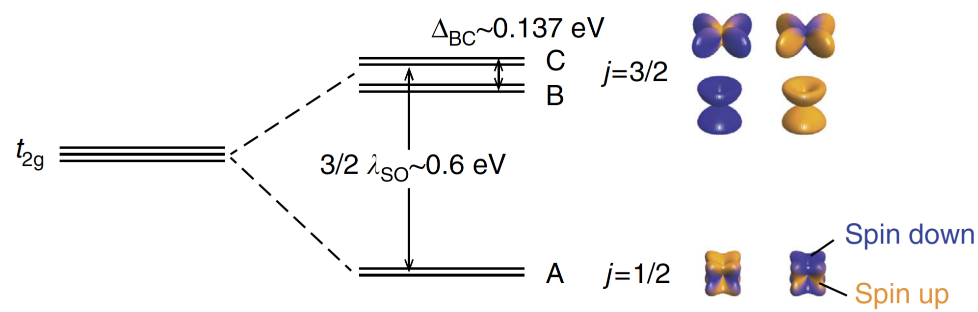


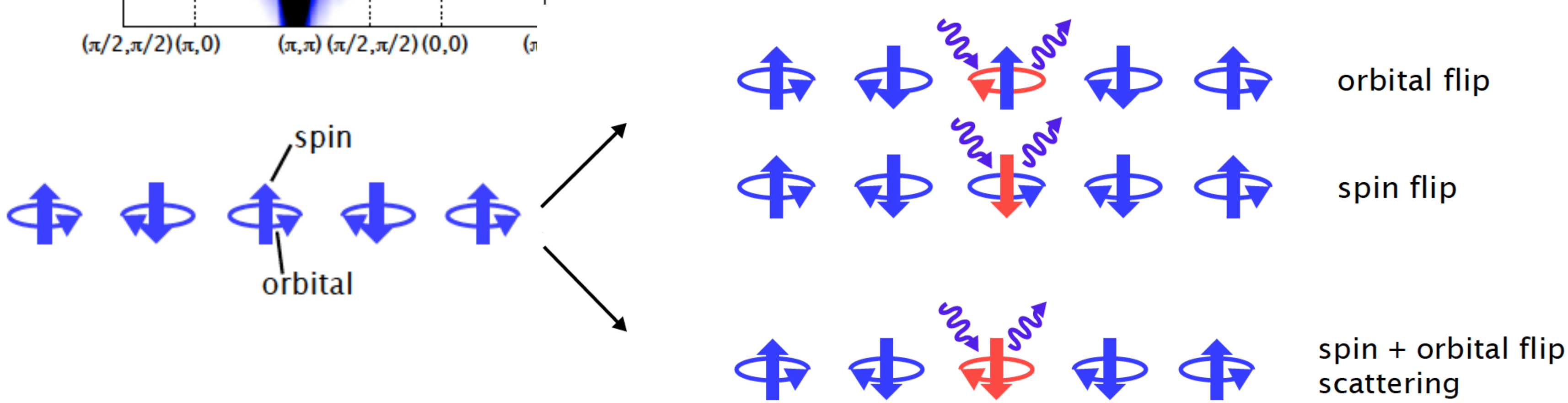


Spin-orbit exciton

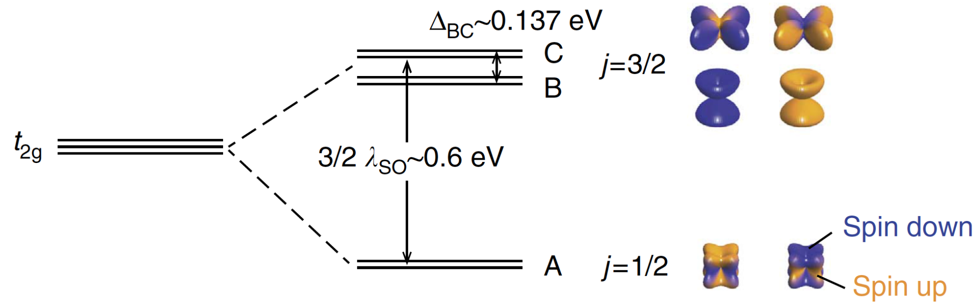


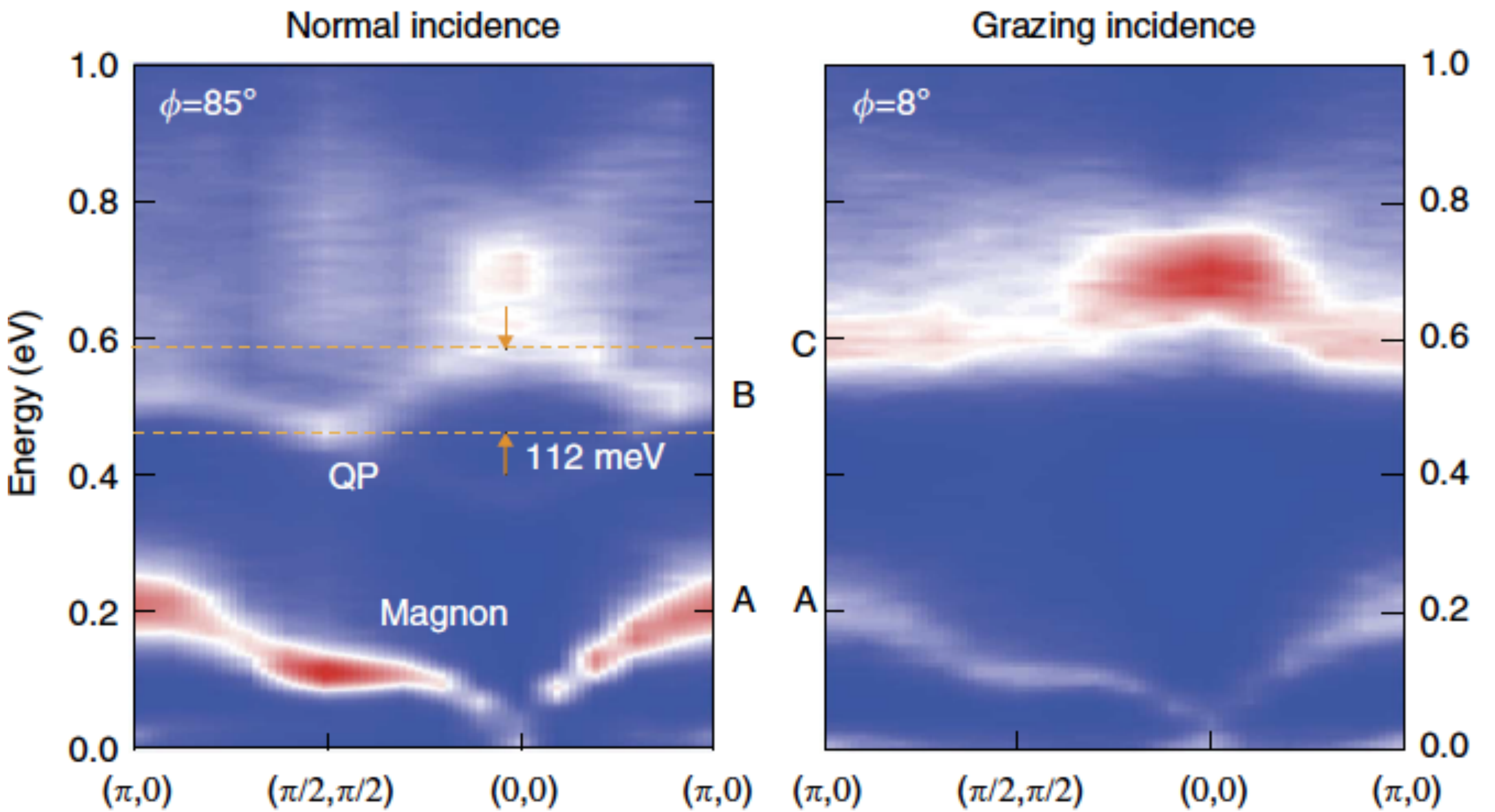




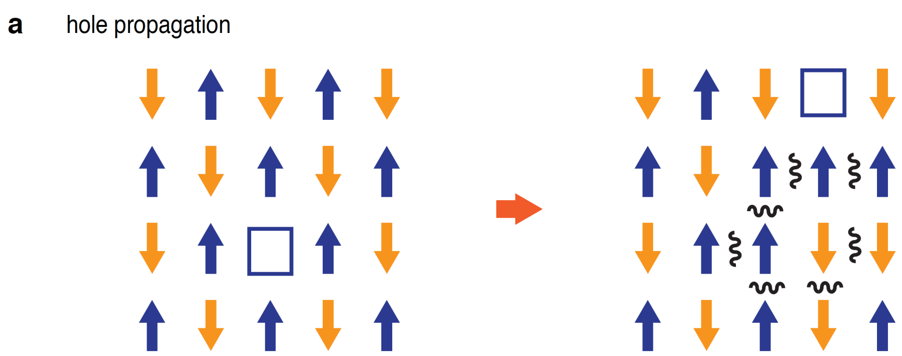


Spin-orbit exciton

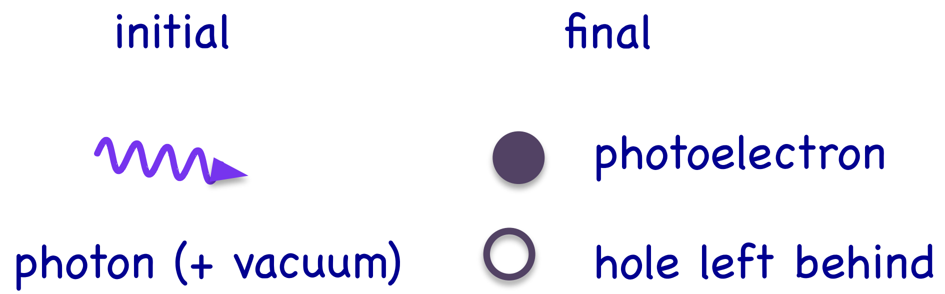




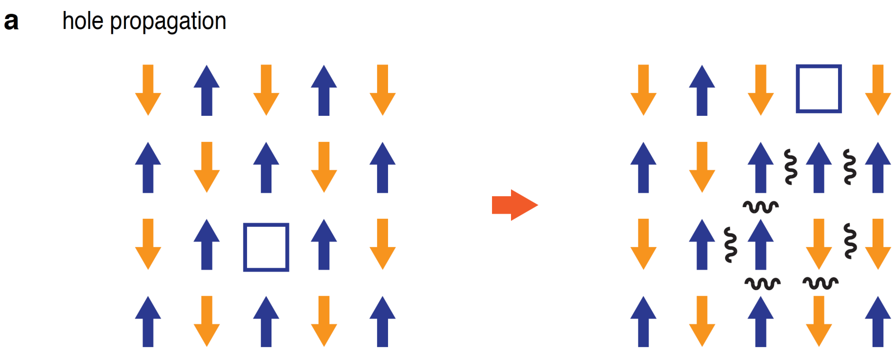
One-hole problem



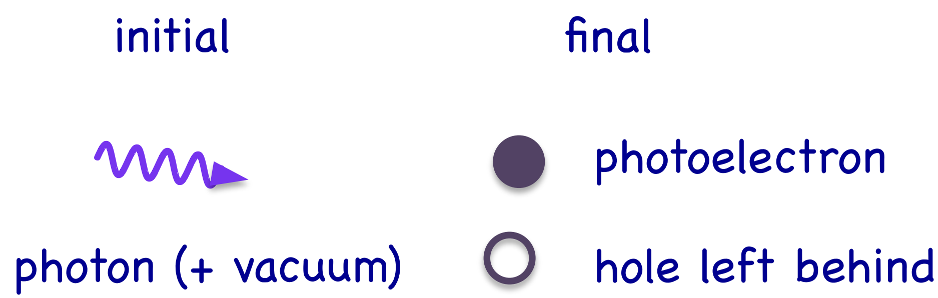
ARPES (photon-in electron-out)

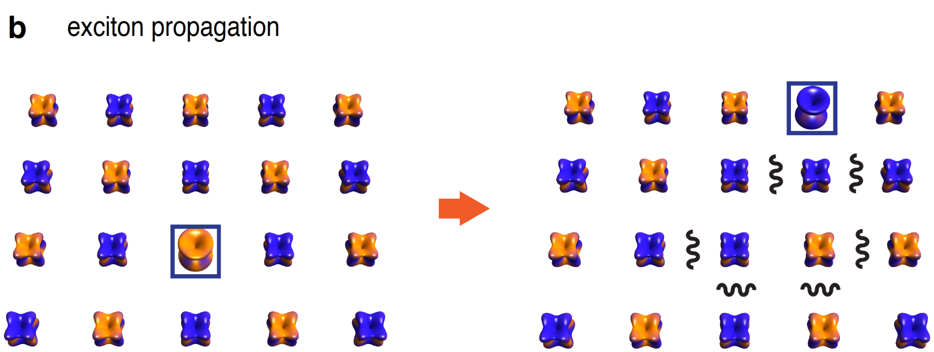


One-hole problem

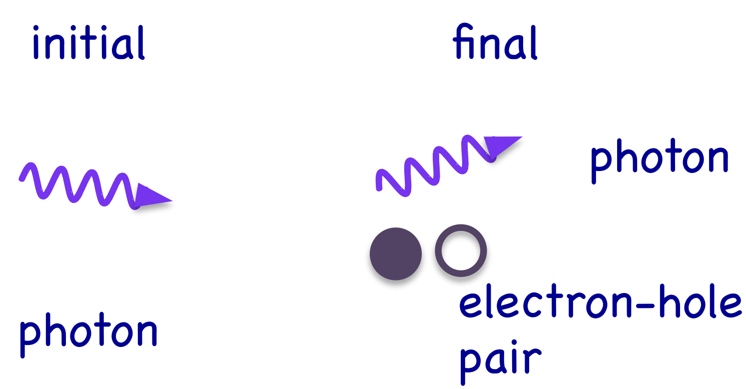


ARPES (photon-in electron-out)

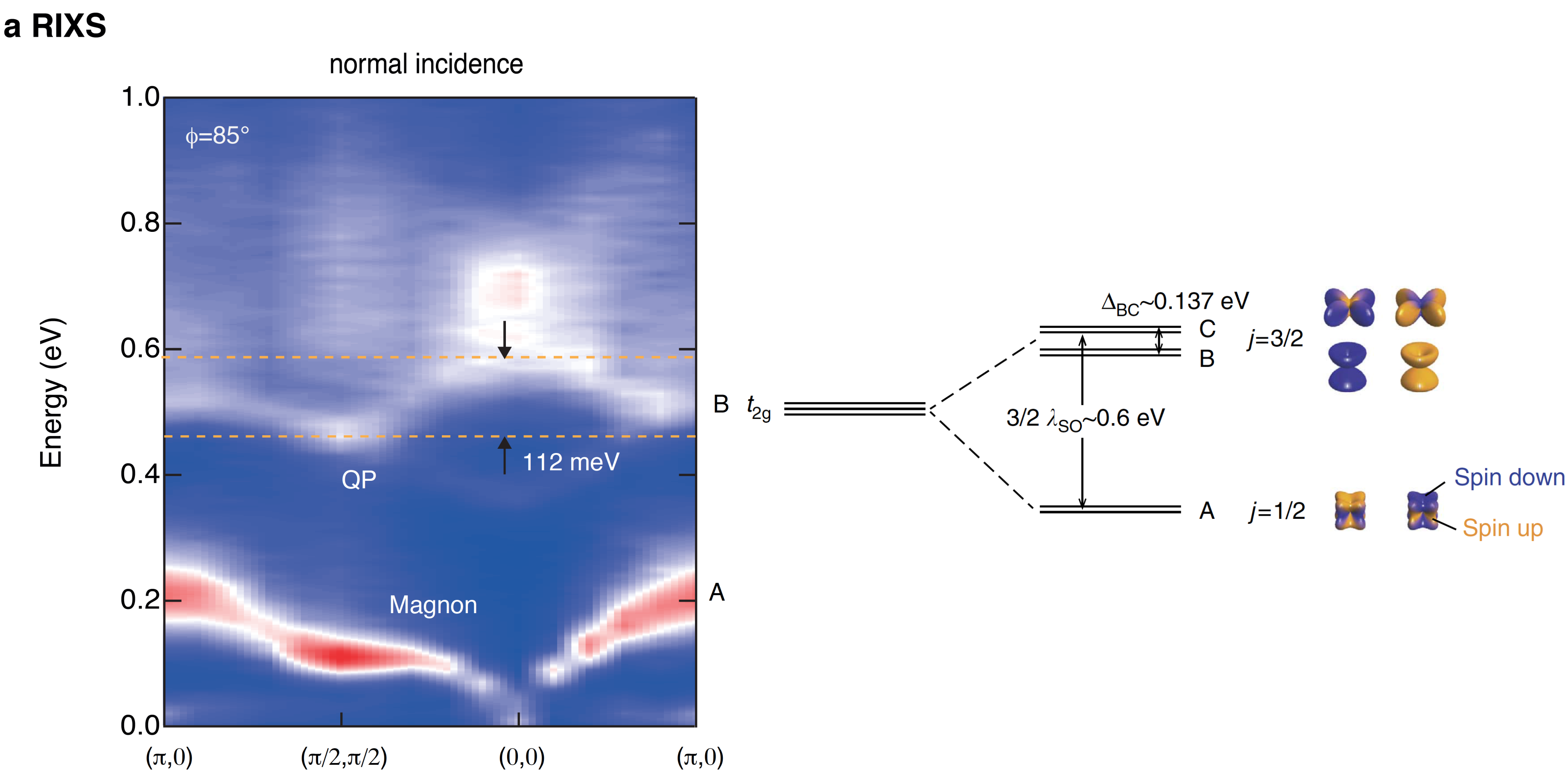




RIXS (photon-in photon-out)

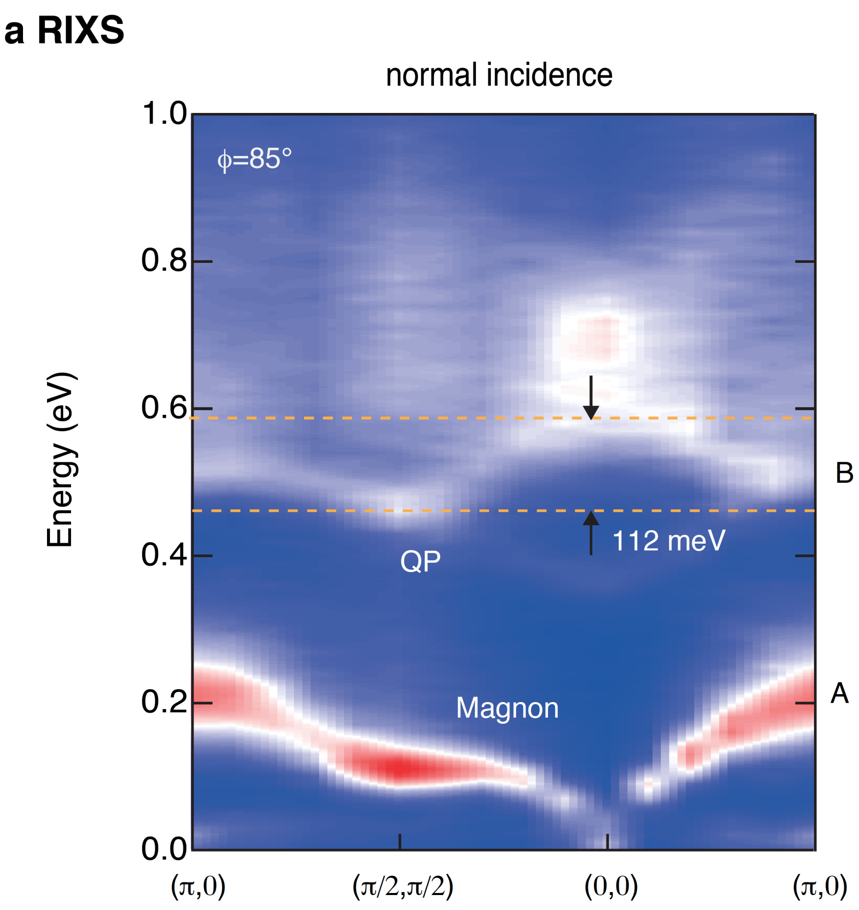


One-hole problem

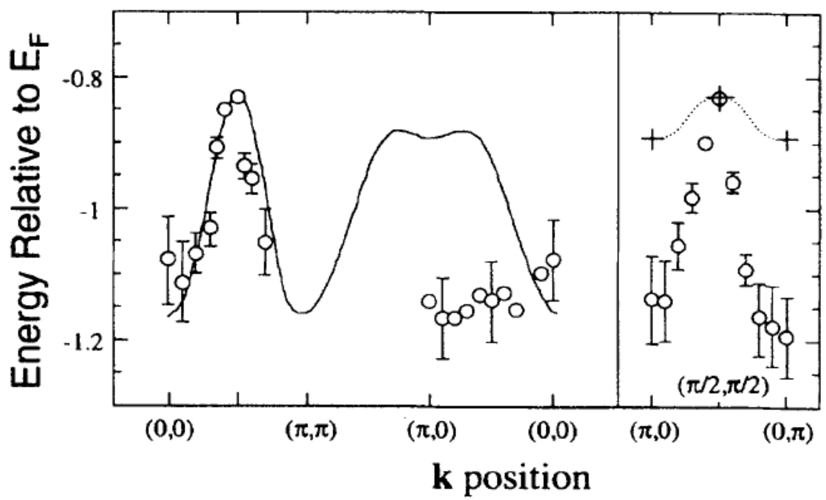


J. Kim, BJK et al. Nature Commun. (2013)

One-hole problem



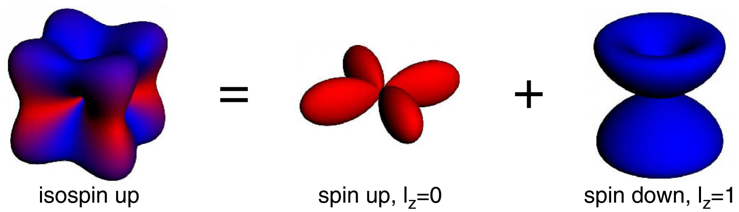
ARPES (cuprate)



B. O. Wells et al., PRL (1995)

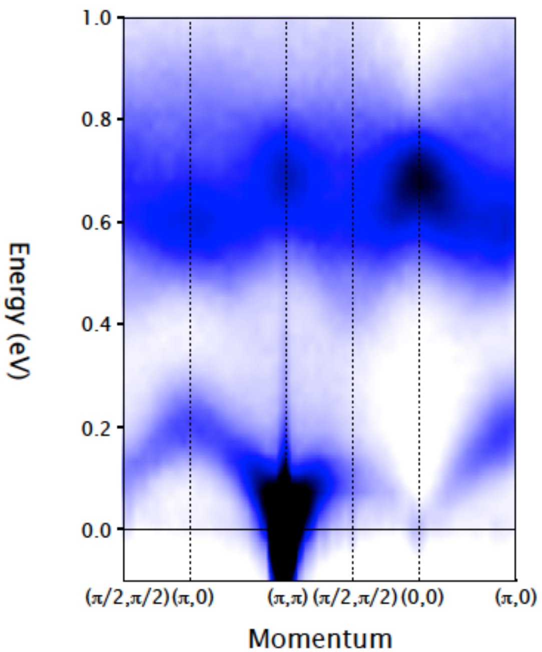
J. Kim, BJK et al. Nature Commun. (2014)

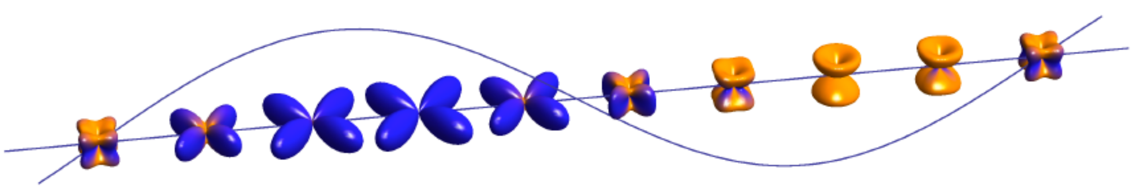
Summary



RXD can not only solve the magnetic structure but also the spin-orbital structure

RIXS is an excellent tool for 5d oxides to study magnetic excitations.





RIXS is sensitive to many other kinds of excitations beyond magnons