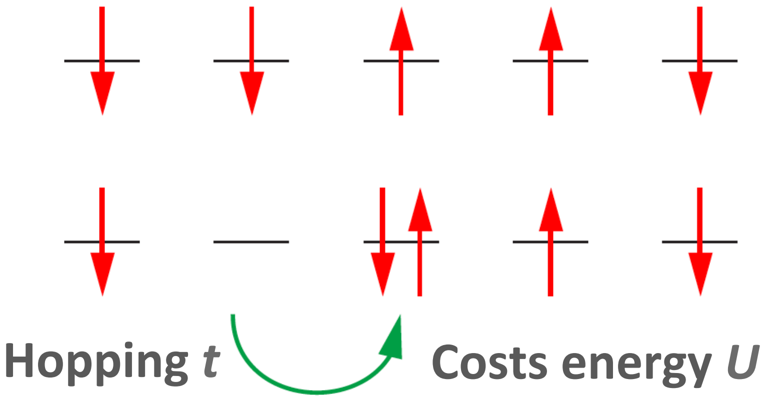
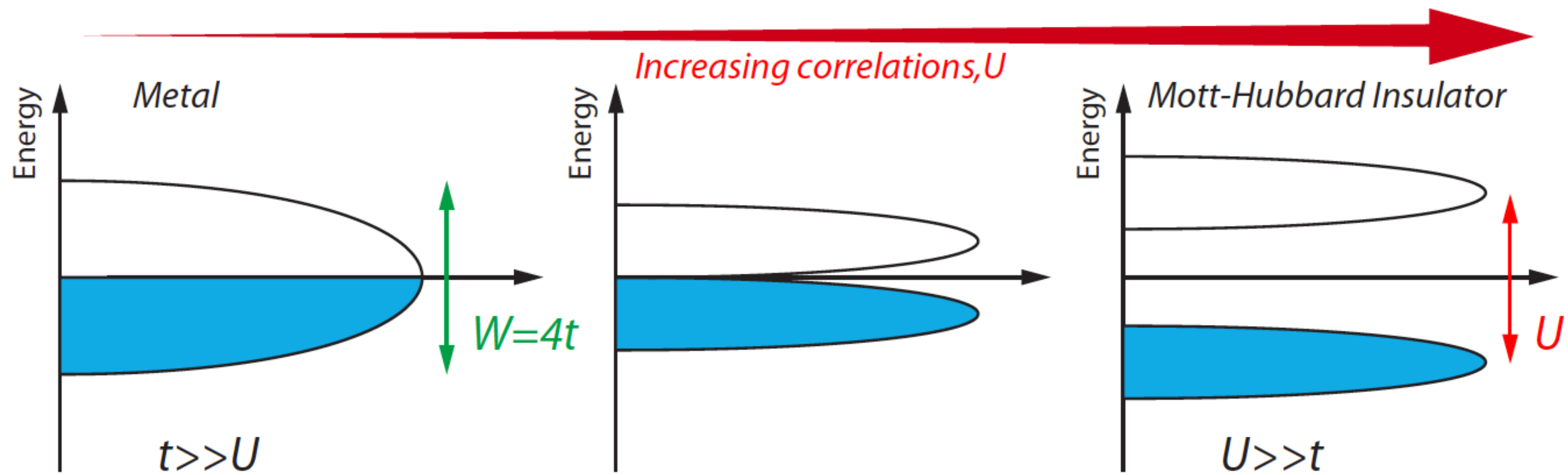
Strongly Correlated Systems and Mott-Hubbard-Heisenberg paradigm



$$\mathcal{H}_{Hubbard} = \mathcal{H}_{kinetic}(t) + \mathcal{H}_{repulsion}(U)$$

$$\mathcal{H} = J \sum_{<ij>} \mathbf{S}_i \cdot \mathbf{S}_j \qquad J = \frac{4t^2}{U}$$

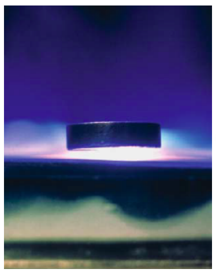


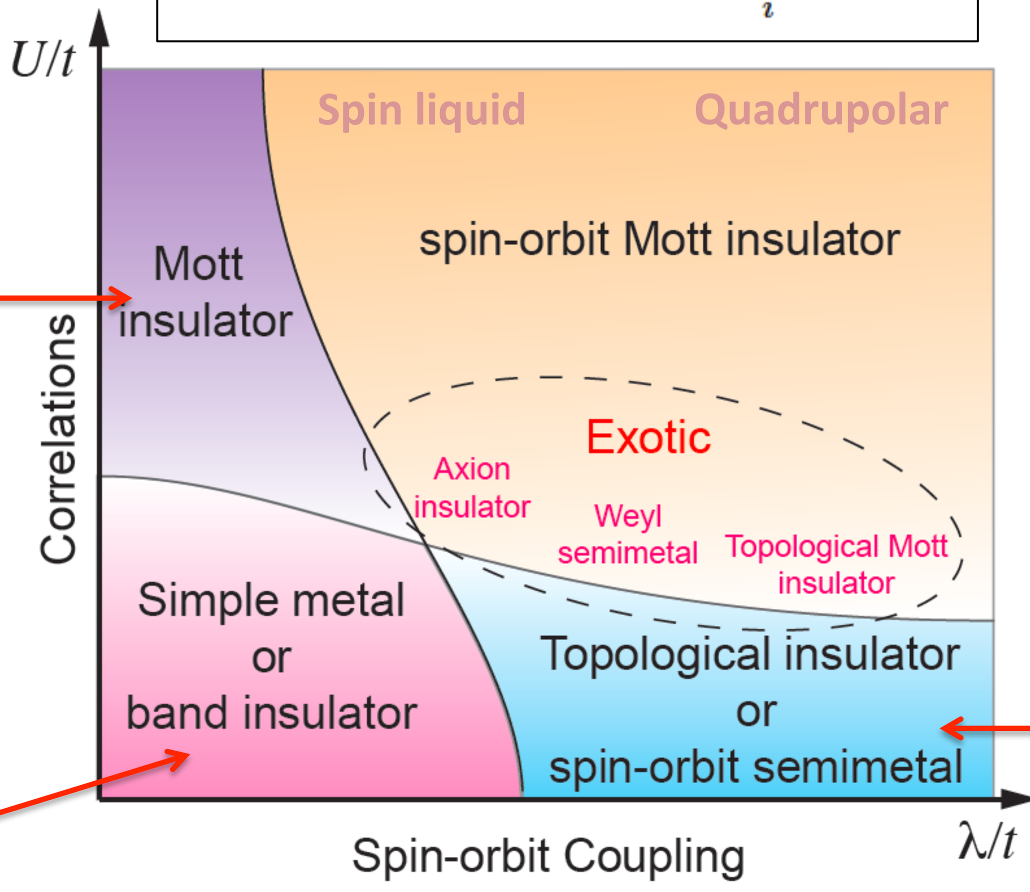
- Mott-Hubbard insulating state arises from electron-electron interactions
- Mott-Hubbard metal-insulator transition independent of magnetic order
- Low-energy spin physics described by isotropic Heisenberg Hamiltonian

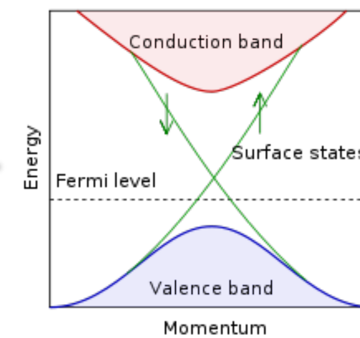
Beyond the Mott-Hubbard-Heisenberg Paradigm

Strong Spin-Orbit Coupling Limit

$$\mathcal{H} = \mathcal{H}(t, U)_{Hubbard} + \lambda \sum_i \mathbf{S}_i \cdot \mathbf{L}_i$$

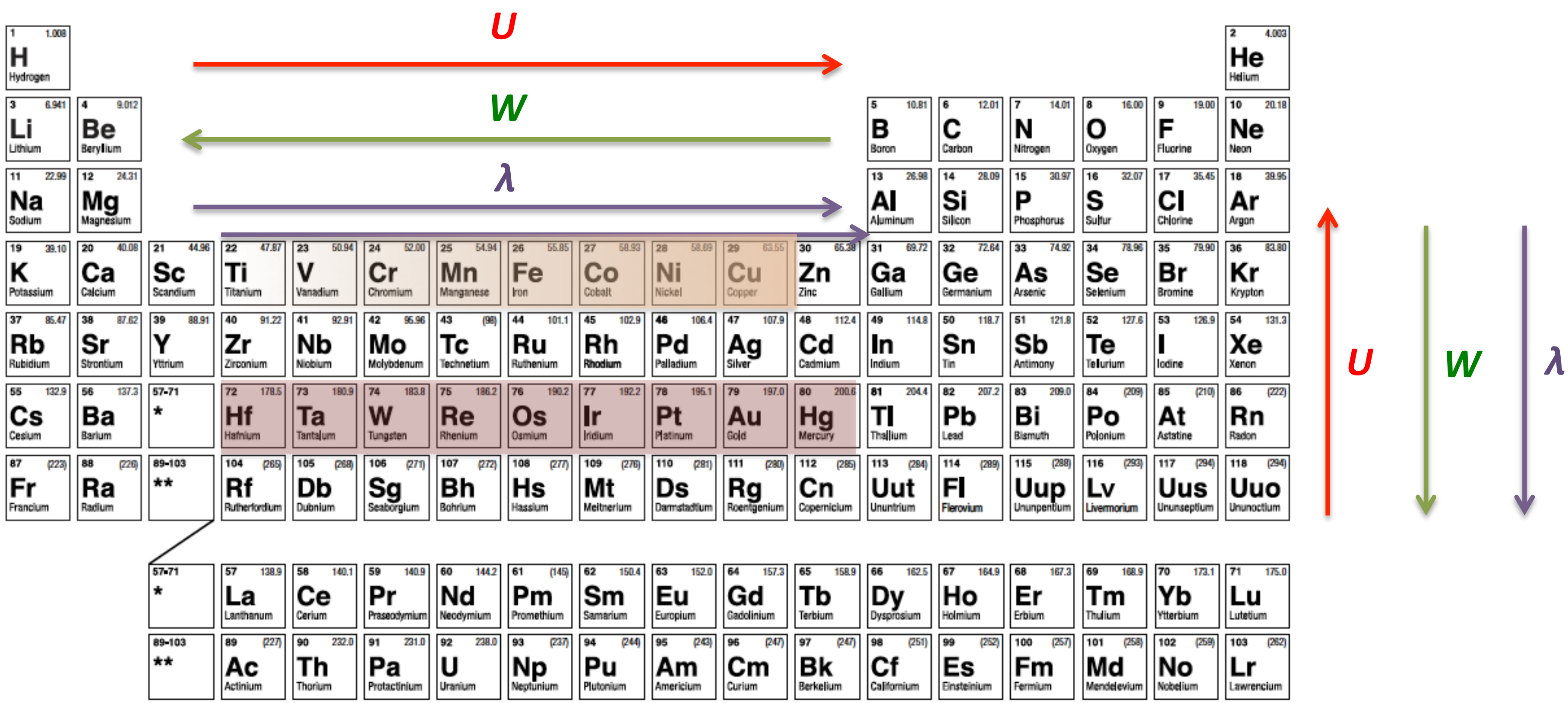






Witczak-Krempa et al., arXiv:1305.2193

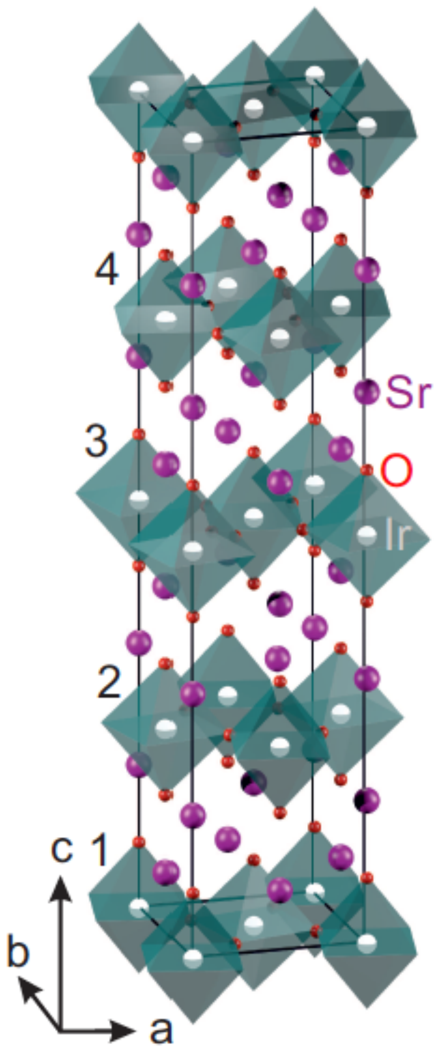
Z Dependence of Energy Scales



- For the late 3d series, U >> W, and are consequently strongly correlated: λ is small.
- In the 5d series, W~U, and in the absence of SOC, **generally expect metallic behaviour**
- However, the fact that U~λ have similar energy scale (1 eV) plays a decisive role
- Delocalisation implies covalency effects may also play an important role

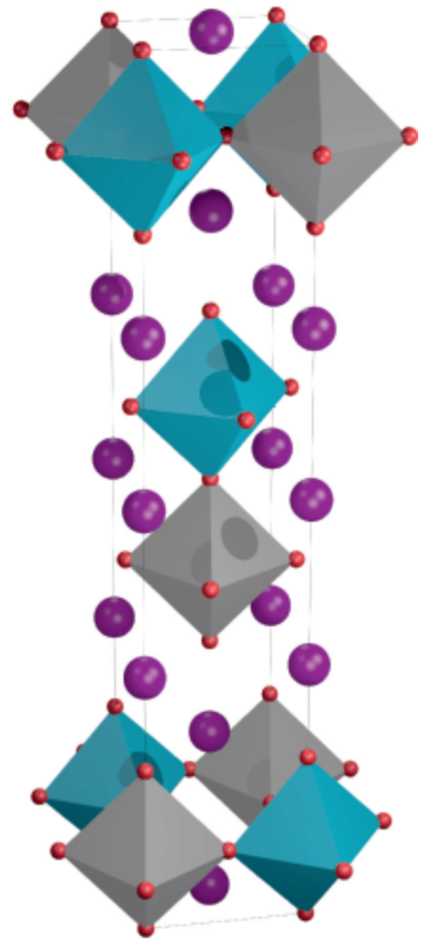
Nature and Evolution of the Mott-like insulating state in $Sr_{n+1}Ir_nO_{3n+1}$

Sr_2IrO_4 (n=1) monolayer



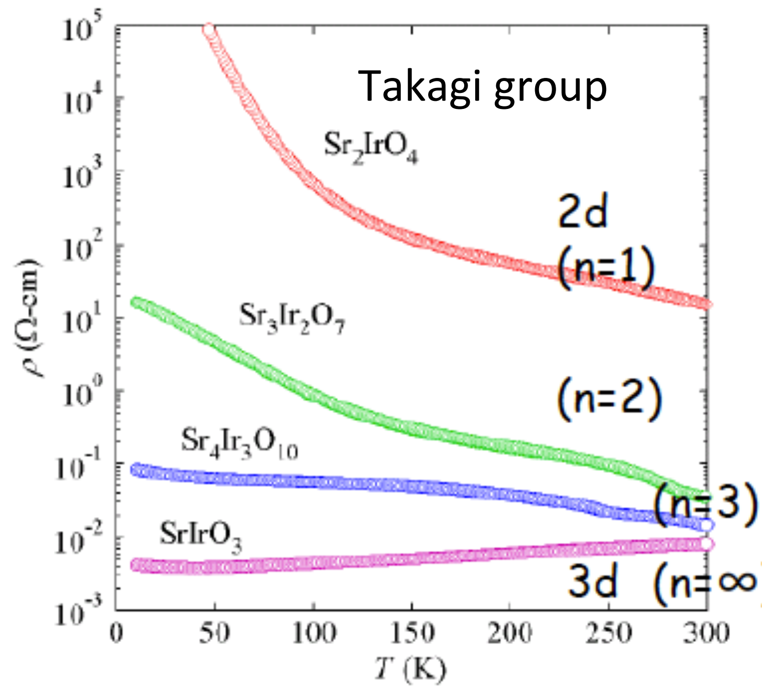
$I4_1/acd$

$Sr_3Ir_2O_7$ (n=2) bilayer



Bbcb or I4/mmm

Ruddlesden-Popper perovskite iridates

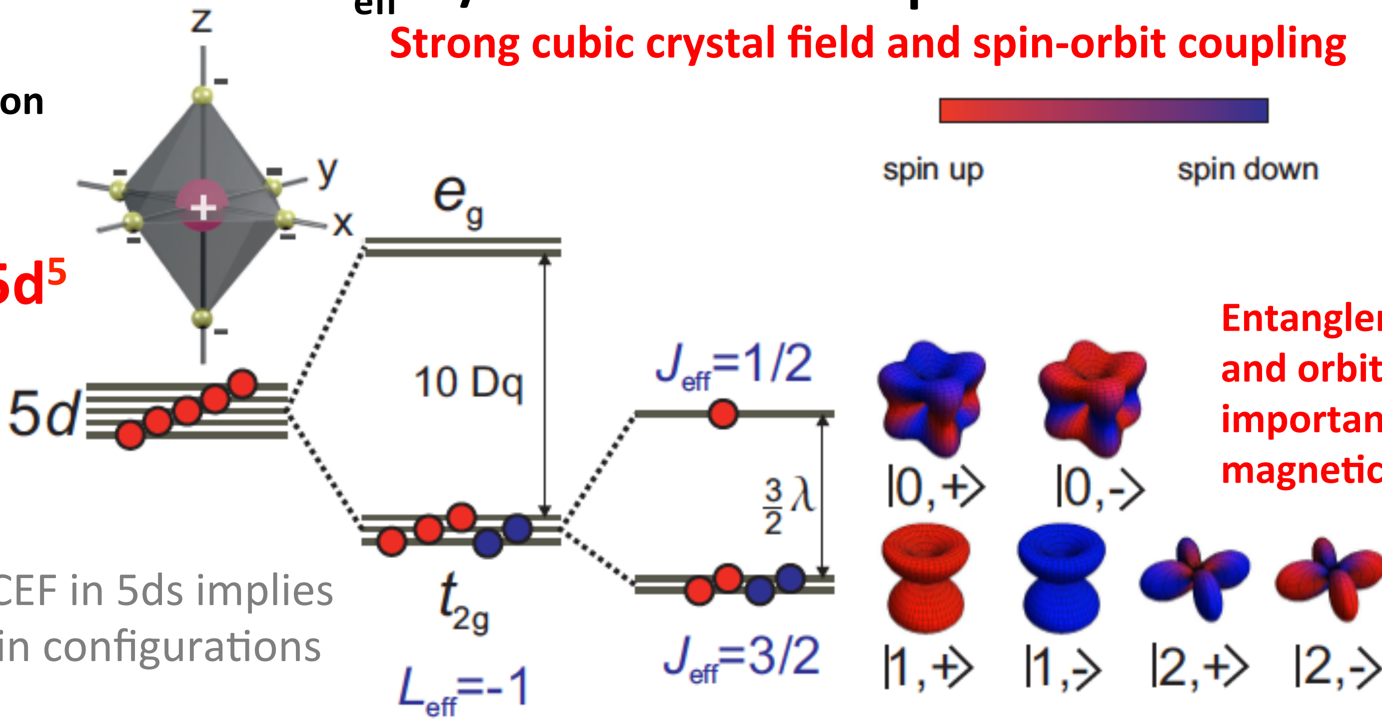


The J_{eff}=1/2 state and the spin-orbit Mott insulator

Strong cubic crystal field and spin-orbit coupling

Single-ion

Ir^{4+}, $5d^5$



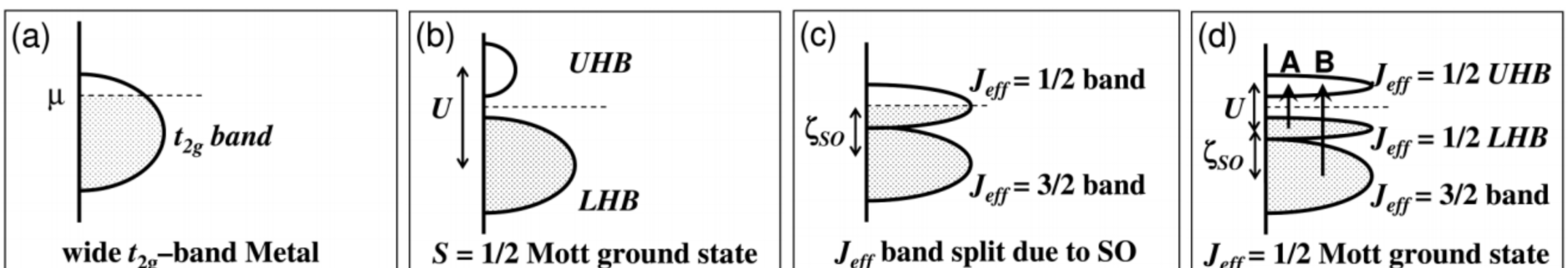
Large CEF in 5ds implies low spin configurations

Entanglement of spin and orbital moments => important consequences for magnetic interactions

B.J. Kim et al. PRL (2008)

$$|j_{\rm eff} = \frac{1}{2}\rangle_c = \frac{|xy,-\rangle + |yz,+\rangle - \imath|zx,+\rangle}{\sqrt{3}}$$

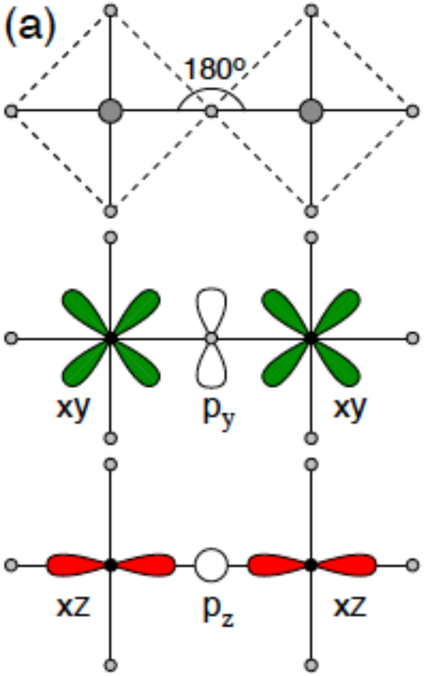
Electron band formation and the opening of a Mott gap



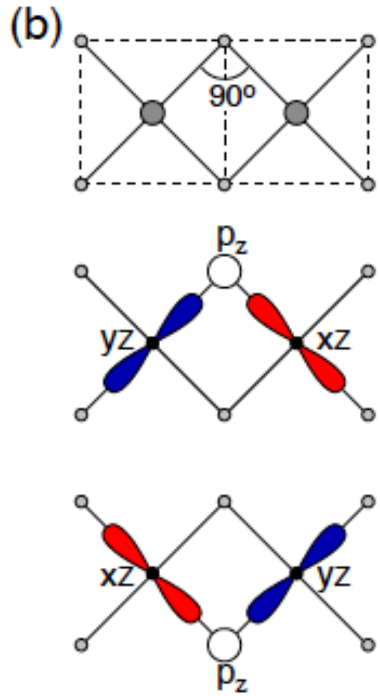
Novel Groundstates and Excitations in Iridates

Exquisite sensitivity of interactions for Jeff=1/2 state to lattice topology

Landmark paper by Jackeli and Khaliullin, PRL (2009)

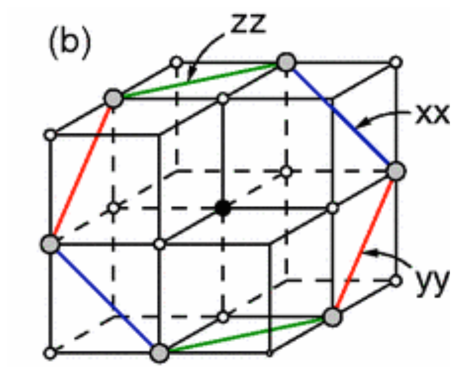


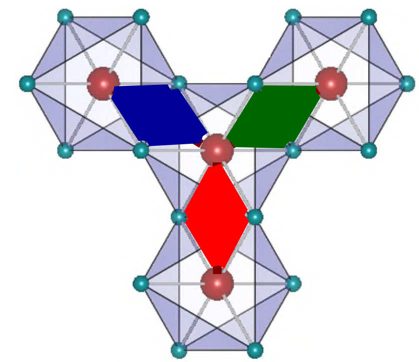
Isotropic
Heisenberg Exchange
$J_1 \mathbf{S}_i \cdot \mathbf{S}_j$



Anisotropic
Bond Directional
$K S_i^\gamma S_j^\gamma$

Honeycomb lattice





Kitaev Model

Here and henceforth S refers to the pseudo or isospin of the low energy projected model

$$\mathcal{H} = J\mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma$$

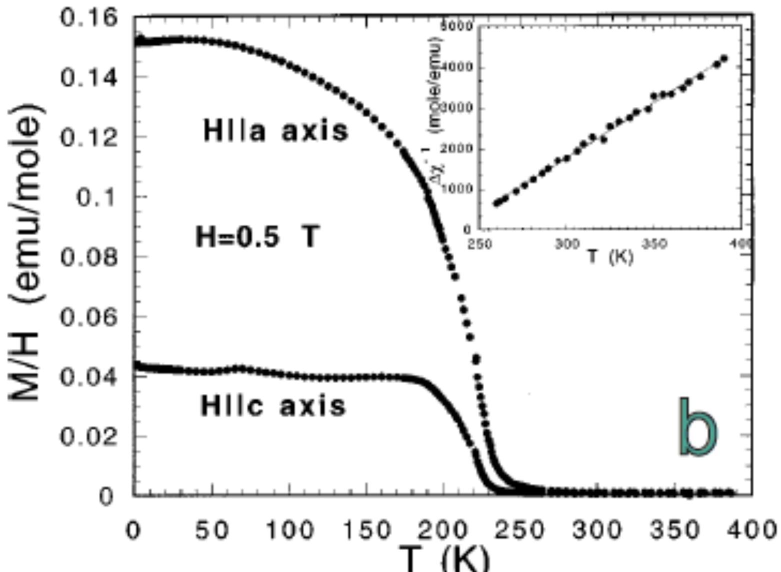
Kitaev-Heisenberg Model

Predicted to give rise to novel phases and excitations

Magnetism in $A_{n+1}Ir_nO_{3n+1}$ (A=Ba,Sr)

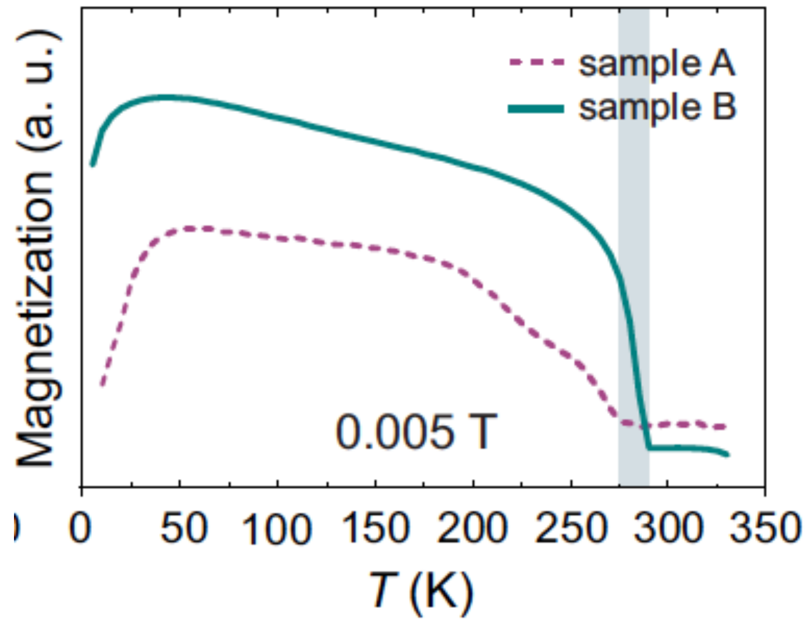
Bulk data

Sr_2IrO_4 (n=1)
monolayer



Cao et al. PRB (1998)

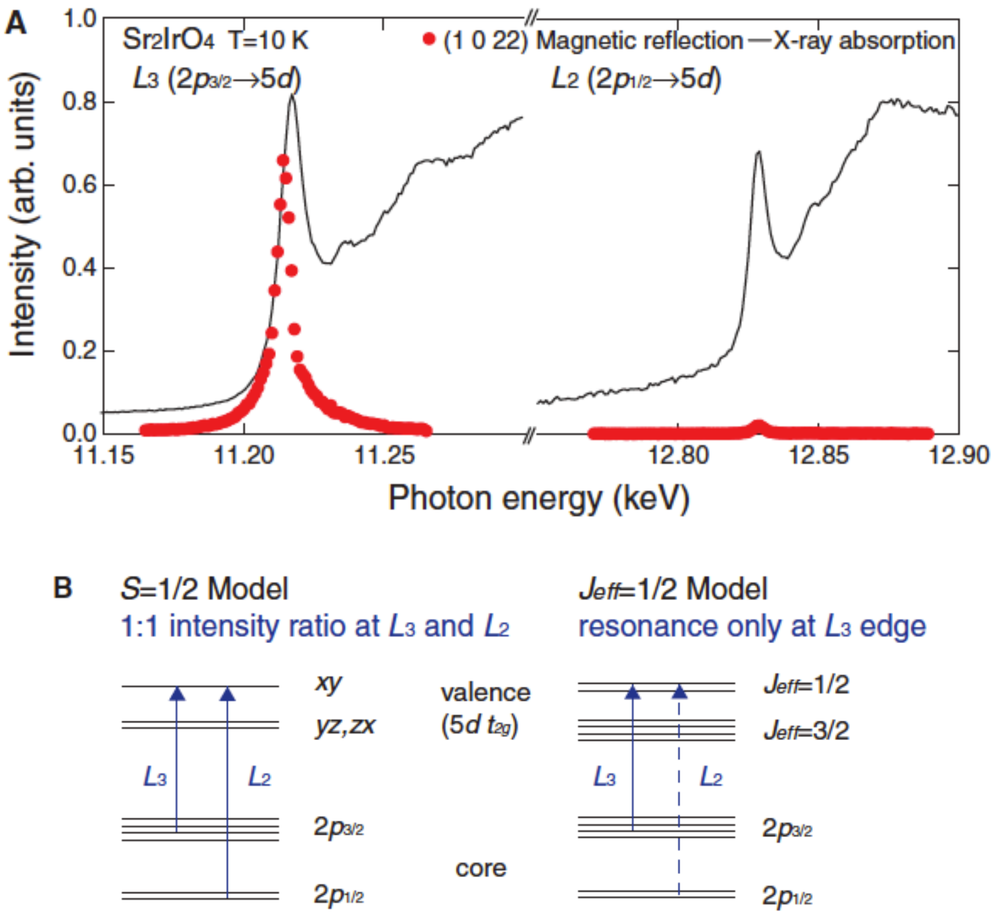
$Sr_3Ir_2O_7$ (n=2)
bilayer

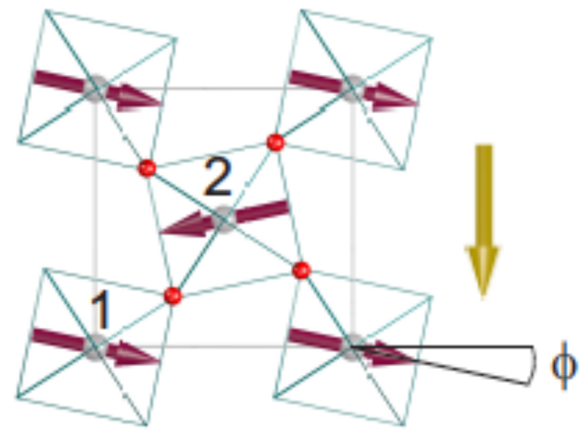


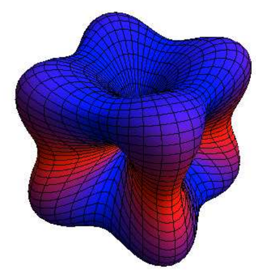
- Excellent at identifying the existence of phase transitions
- Very difficult to deduce anything detailed concerning nature of the groundstate or excitations

Sr_2IrO_4: Evidence for J_{eff}=1/2 model

B.J. Kim *et al.*, Science, 323 (2009), X-ray Resonant Magnetic Scattering







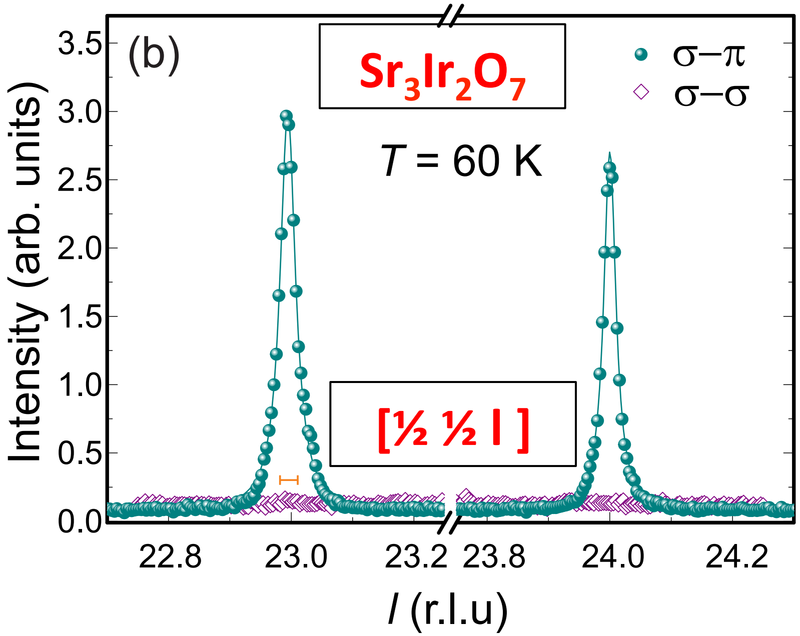
Conclusion: Large L3/L2 branching ratio => J_{eff}=1/2 model

$$|j_{\text{eff}} = \frac{1}{2}\rangle_c = \frac{|xy,-\rangle + |yz,+\rangle - \imath|zx,+\rangle}{\sqrt{3}}$$

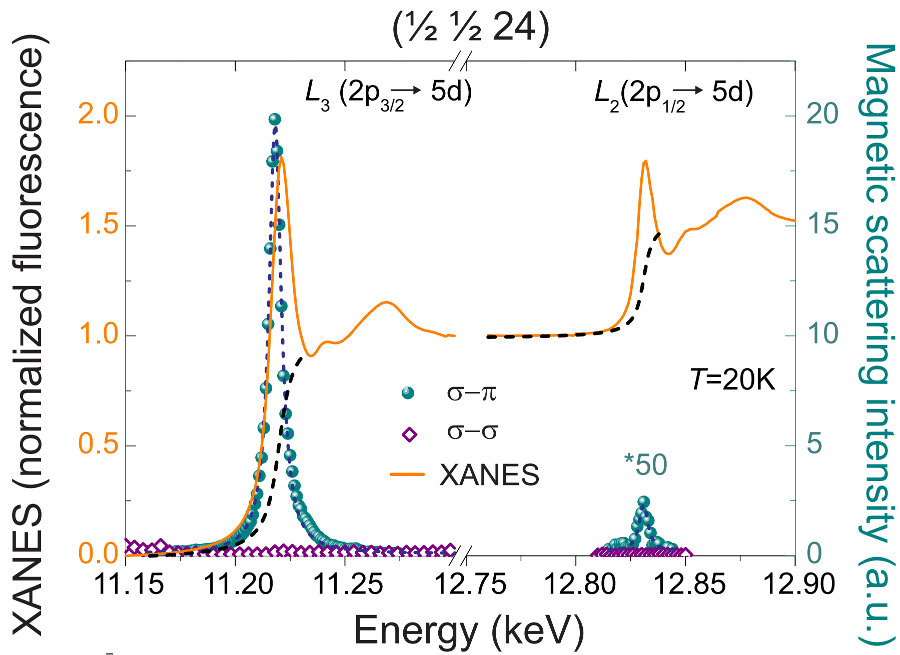
How to determine the magnetic structure using REXMS

Step 1: Determine the ordering wavevector

Find a candidate magnetic peak



Determine energy and polarization dependence



Scattering length for dipole resonances (E1)

$$f_{E1}^{XMRS} = -iF_{E1}^{(1)} \begin{pmatrix} \sigma \to \sigma' & \pi \to \sigma' \\ \sigma \to \pi' & \pi \to \pi' \end{pmatrix} = -iF_{E1}^{(1)} \begin{pmatrix} 0 & z_1 \cos\theta + z_3 \sin\theta \\ -z_1 \cos\theta + z_3 \sin\theta & -z_2 \sin 2\theta \end{pmatrix}$$

Expect REXMS (dipole) scattering in rotated polarization channel only

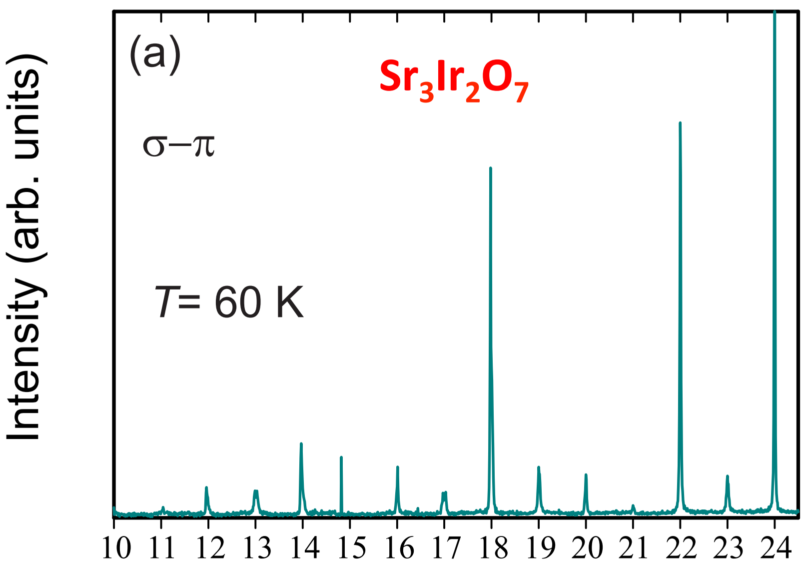
Magnetic ordering wavevector k = (½ ½ 0)
=> Antiferromagnetic coupling in a-b plane

How to determine the magnetic structure using REXMS

Step 2: Determine the relative phases

Collect integrated peak intensities

(½ ½ *l*)



$$\left|\mathcal{F}^{\mathrm{A,AF}}_{(hkl)}\right|^2 \propto \left|e^{\imath 2\pi l z} - e^{-\imath 2\pi l z} + e^{\imath 2\pi(\frac{h}{2}+\frac{k}{2}+\frac{l}{2}-lz)} - e^{\imath 2\pi(\frac{h}{2}+\frac{k}{2}+\frac{l}{2}+lz)}\right|^2$$

$$\propto \left|2\imath \sin(2\pi l z)\left(1 - e^{\imath 2\pi(\frac{h}{2}+\frac{k}{2}+\frac{l}{2})}\right)\right|^2,$$

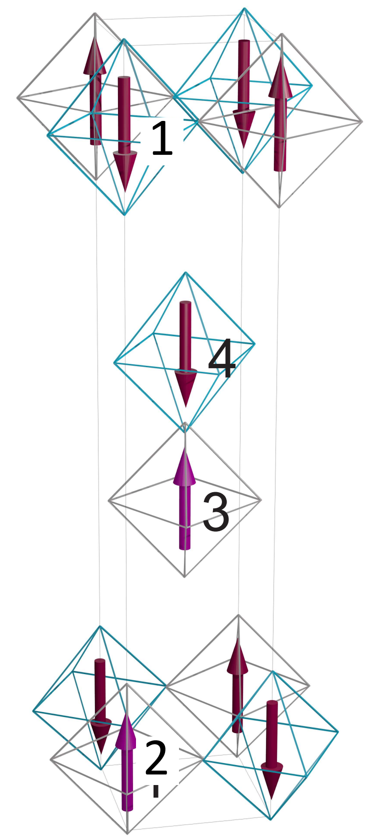
For (½ ½ l)

$$\left|\mathcal{F}^{\mathrm{A,AF}}_{(\frac{1}{2}\,\frac{1}{2}\,l)}\right|^2 \propto \left|2\imath \sin(2\pi l z)\left(1 + e^{\imath \pi l}\right)\right|^2$$

Non-zero for even l peaks

Compare with calculated structure Factors

$$\left|\mathcal{F}\right|^2 = \left|\sum_j \mathcal{A}_j \exp^{i\mathbf{Q}\cdot\mathbf{r}_j}\right|^2$$

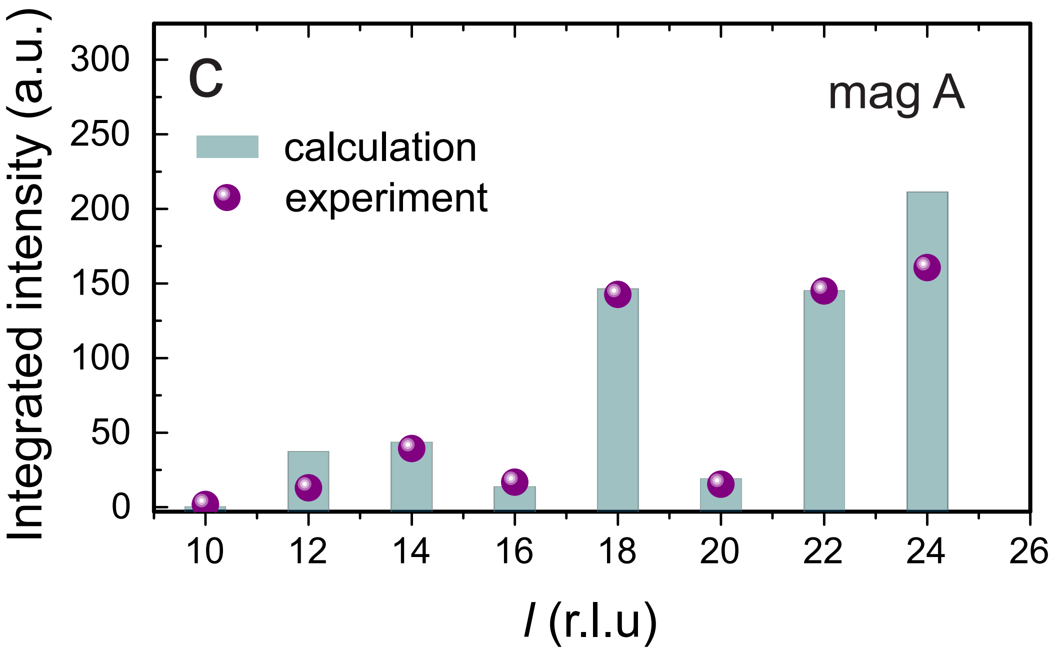


$$\mathcal{A}_2 = \mathcal{A}_3 = -\mathcal{A}_1 = -\mathcal{A}_4$$

How to determine the magnetic structure using REXMS

Step 2: Determine the relative phases (continued)

Compare Measured and Calculated Intensities



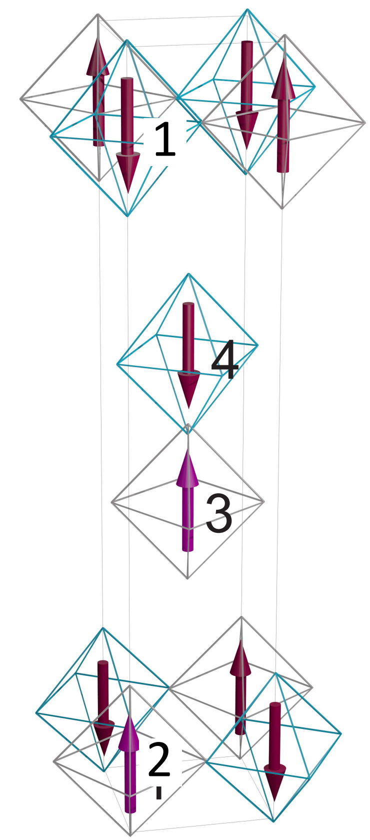
Apply corrections for:

- Absorptiion
- Geometrical terms in the Cross-section

$$f_{E1}^{XMRS} = -iF_{E1}^{(1)} \begin{pmatrix} 0 & z_1\cos\theta + z_3\sin\theta \\ -z_1\cos\theta + z_3\sin\theta & -z_2\sin 2\theta \end{pmatrix}$$

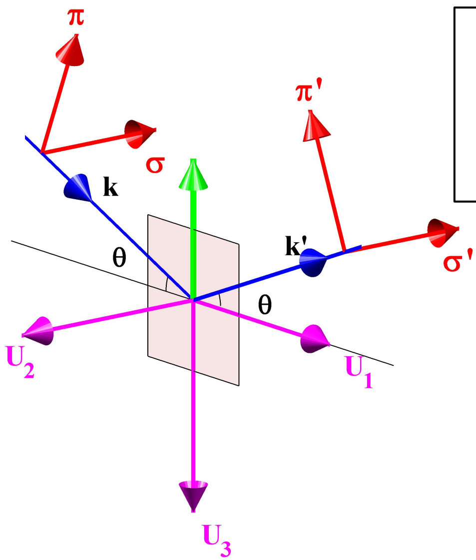
- Etc.

Candidate magnetic structure

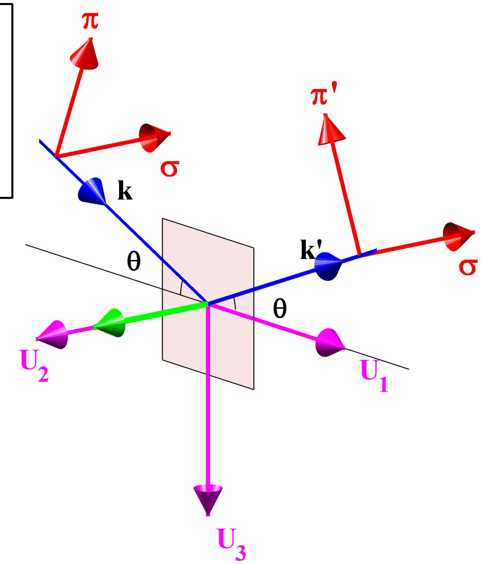


How to determine the magnetic structure using REXMS

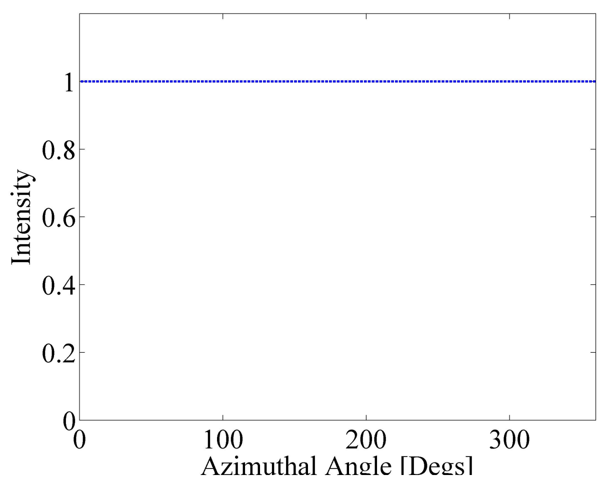
Step 3: Determine the directions of the magnetic moments

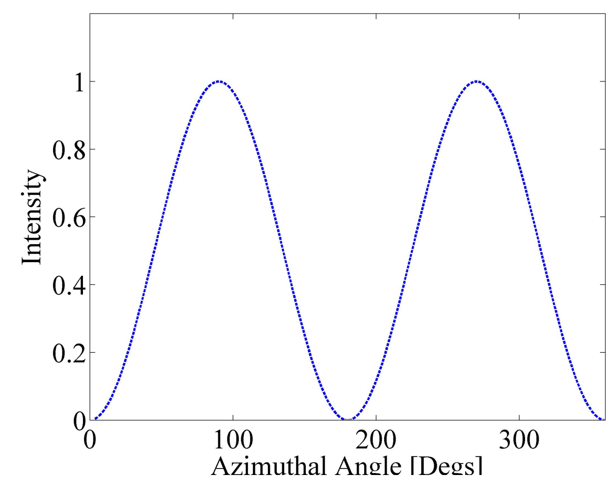


Rotate around wavevector transfer Q: Azimuthal Scans



$$f_{E1}^{XMRS} = -iF_{E1}^{(1)} \begin{pmatrix} \sigma \to \sigma' & \pi \to \sigma' \\ \sigma \to \pi' & \pi \to \pi' \end{pmatrix} = -iF_{E1}^{(1)} \begin{pmatrix} 0 & z_1 \cos\theta + z_3 \sin\theta \\ -z_1 \cos\theta + z_3 \sin\theta & -z_2 \sin 2\theta \end{pmatrix}$$



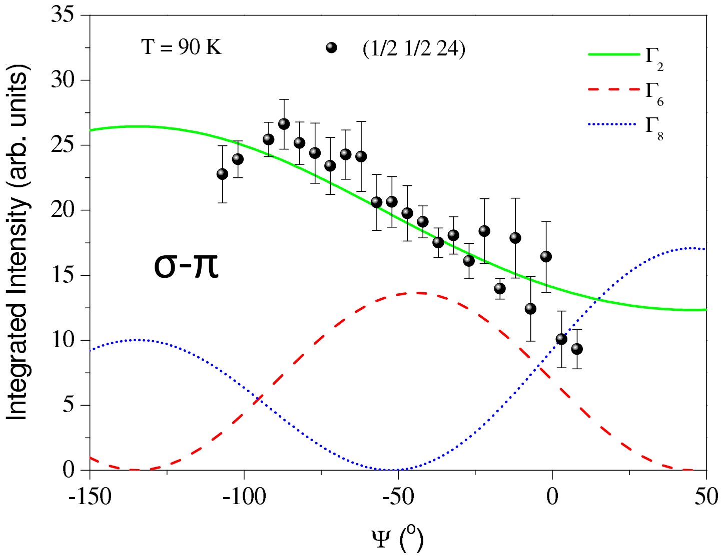


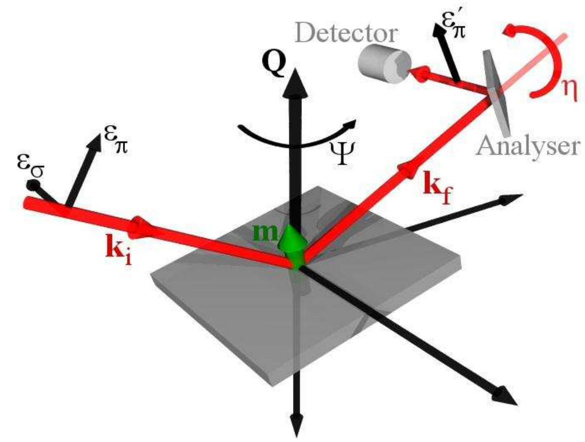
How to determine the magnetic structure using REXMS

Step 3: Determine the directions of the magnetic moments

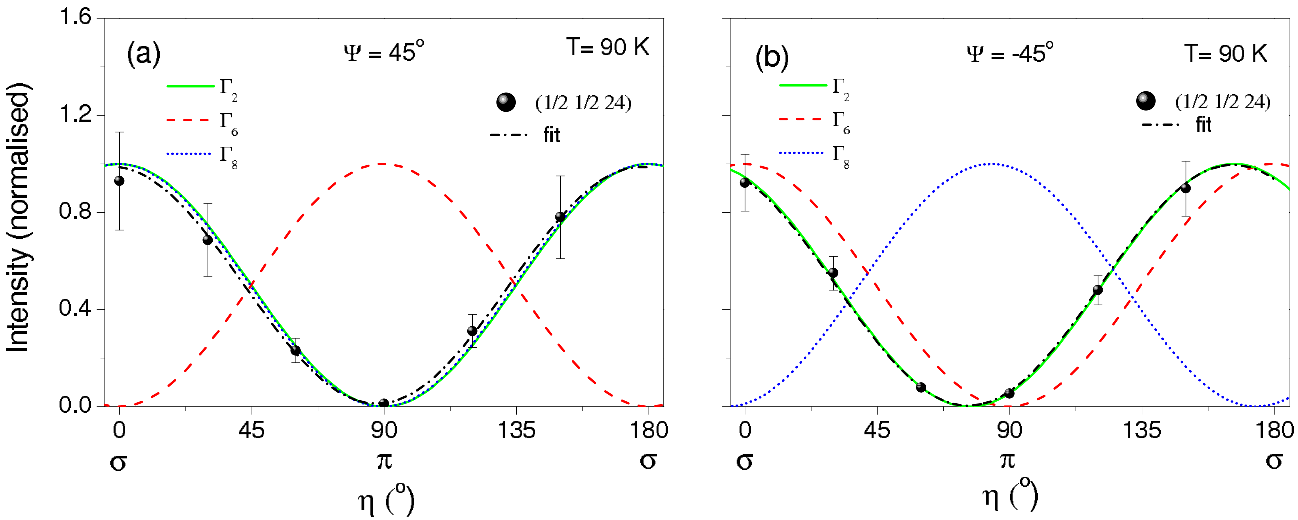
$Sr_3Ir_2O_7$

Γ_2 [001]
Γ_6 [-110]
Γ_8 [110]





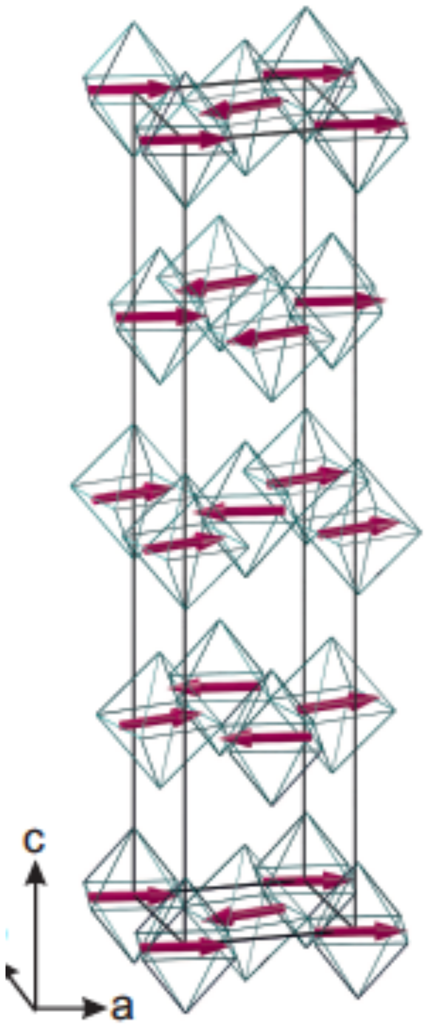
Horizontal geometry
π incident



Conclude that moments in $Sr_3Ir_2O_7$ are purely oriented along the c direction

Moment reorientation transition driven by dimensionality

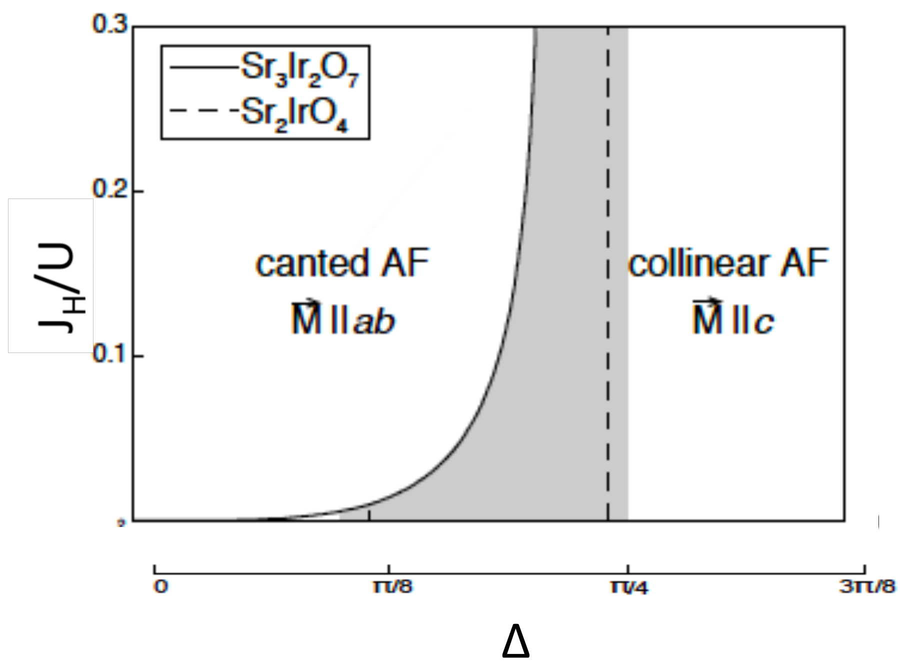
Sr_2IrO_4



J.W. Kim et al. PRL (2012)

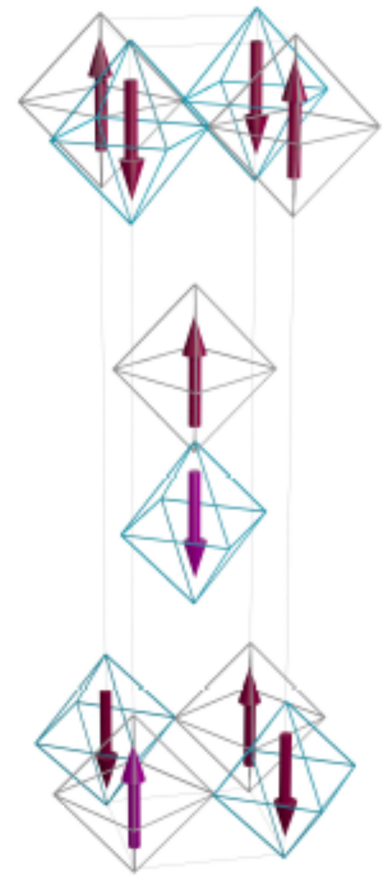
Moment reorientation for n=2 driven by inter-layer pseudo-diploar couplings arising from strong spin-orbit coupling

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



Tetragonal crystal field splitting

$Sr_3Ir_2O_7$



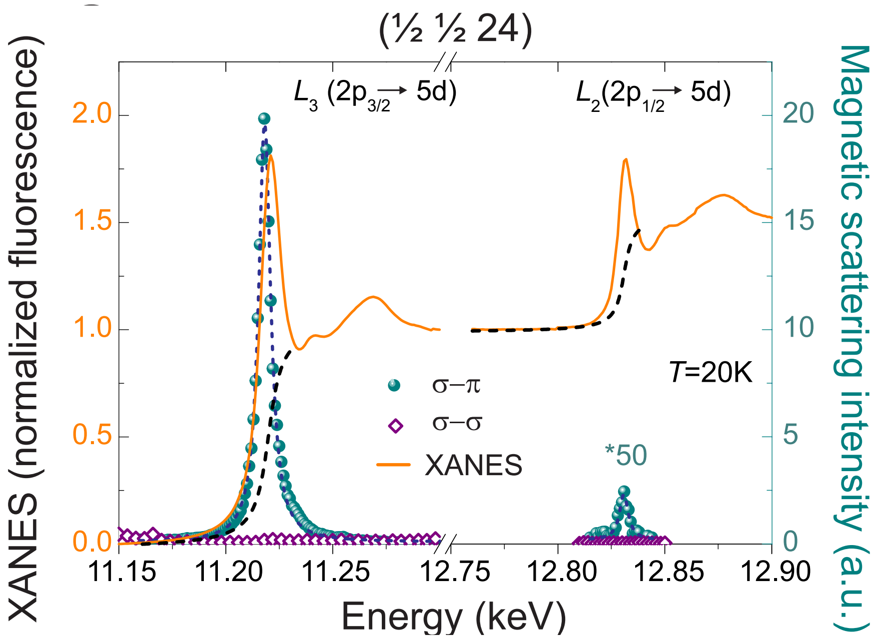
Kim et al. Science (2009)
Boseggia et al. PRL (2013)

Boseggia et al. JPCM (2013)

What else can we learn from REXMS?

Symmetry of the groundstate wavefunction

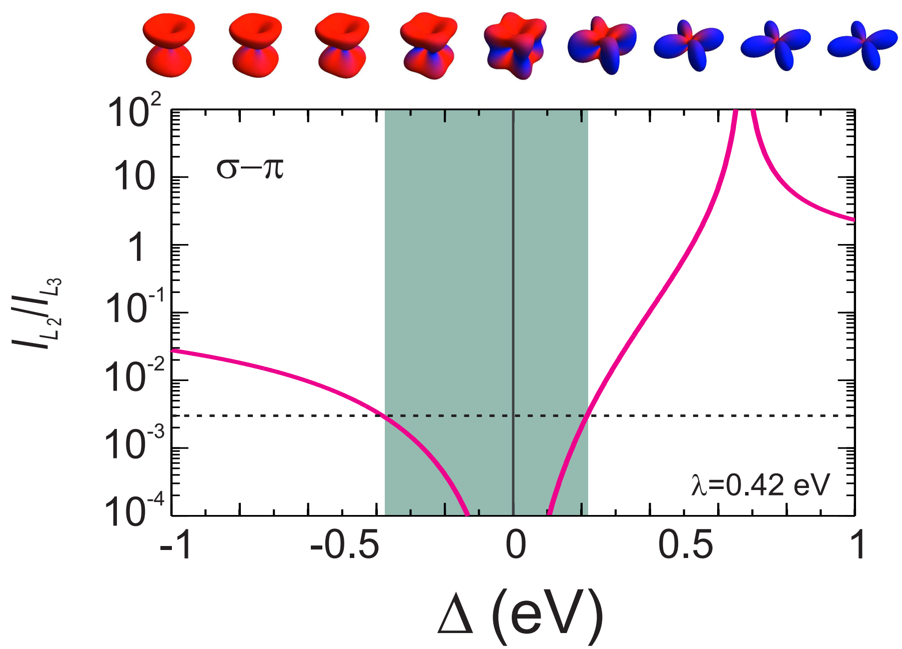
$Sr_3Ir_2O_7$



Resonant X-ray scattering and the jeff=1/2 electronic ground state

Morreti Sala et al. Phys. Rev. Lett. (2014)

$$\mathcal{H} = \zeta \mathbf{L} \cdot \mathbf{S} - \Delta \langle L_z \rangle^2$$



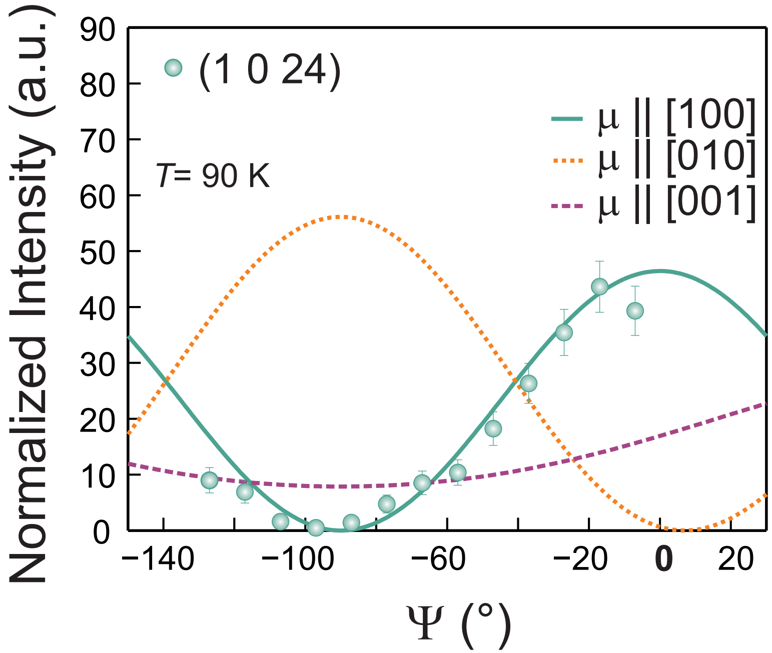
Conclude from branching ratio:

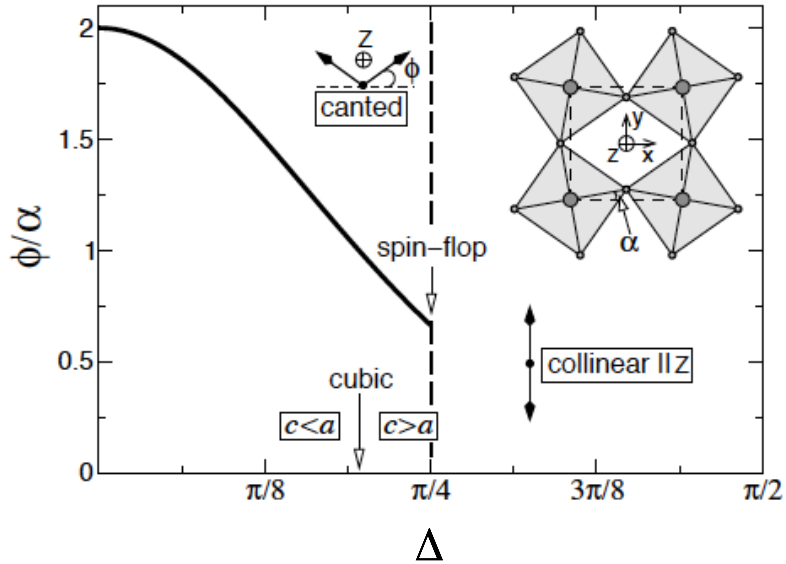
- **Jeff=1/2 state is realised in $Sr_3Ir_2O_7$**
- **Ambiguity in Sr_2IrO_4 as moments lie in basal plane**

The magnetic structure of Sr_2IrO_4

Kim et al., Science (2009), Boseggia et al. PRL (2013) Boseggia et al. JPCM (2013)

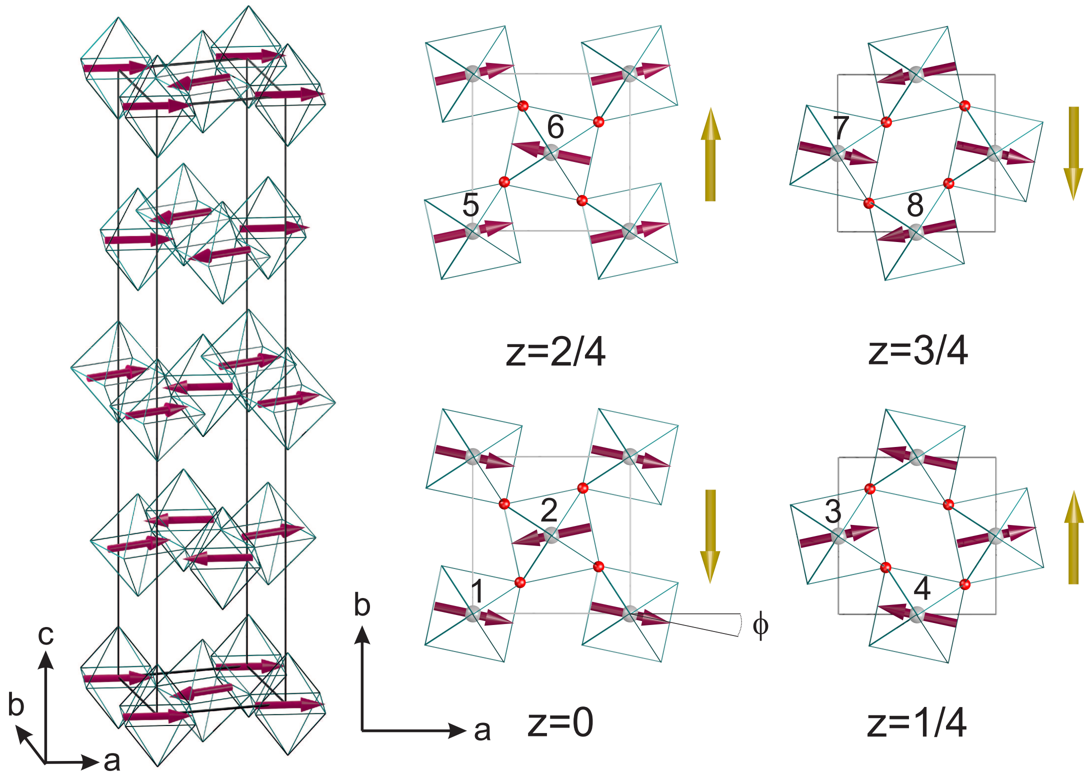
- REXS experiments establish that Ir moments are AF coupled along [100] direction and lie in the a-b plane
- Key prediction of Jeff=1/2 model by Jackeli and Khaliullin is that the moments are canted to follow rigidly the rotation of the IrO_6 octahedra
- Test the model by measuring reflections sensitive to canted AF component





The magnetic structure of Sr_2IrO_4

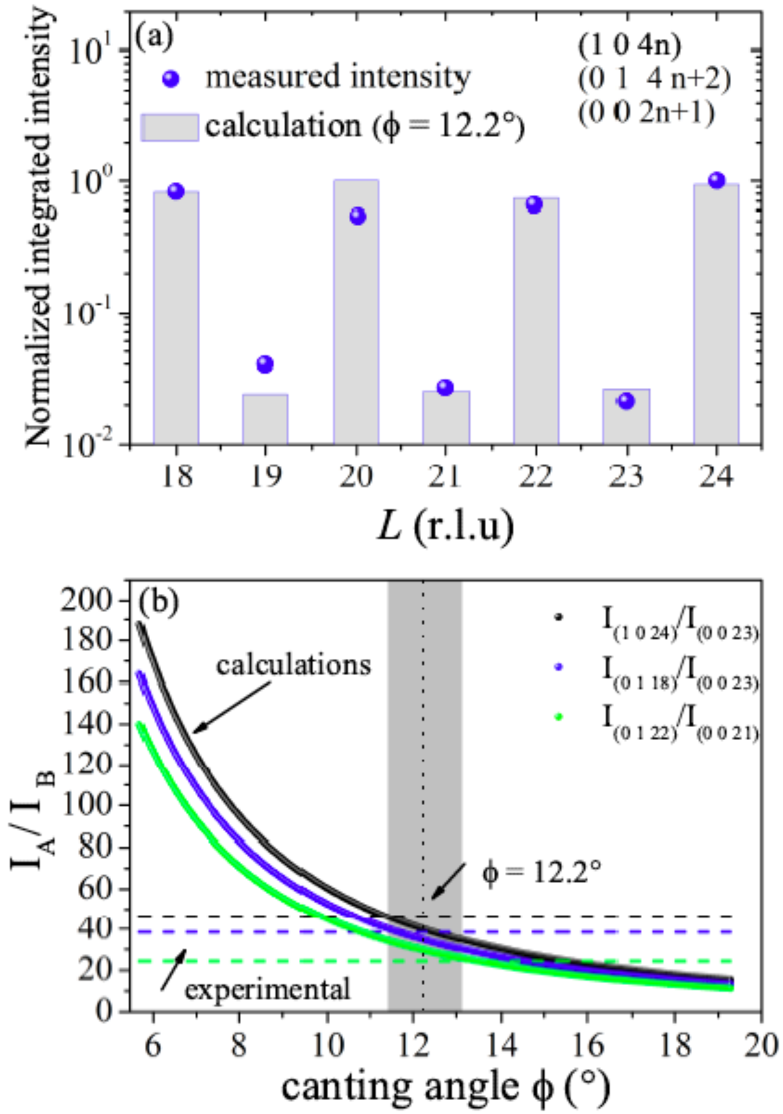
Boseggia et al. JPCM (2013)



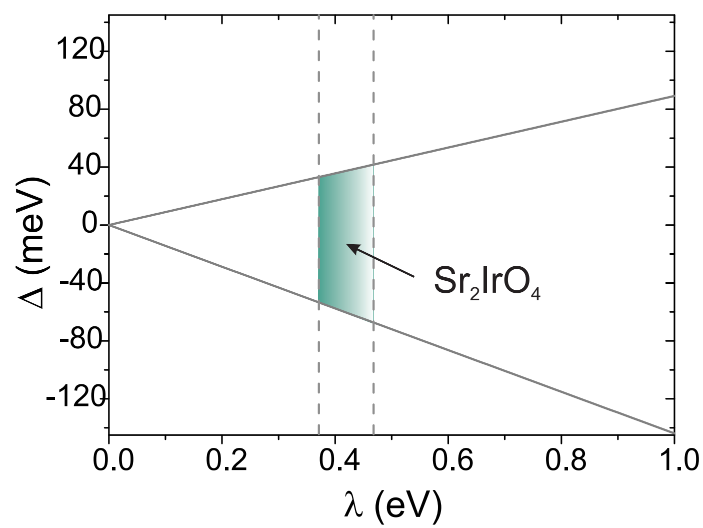
- A sublattice, dominant AF order along a axis: reflections of type (1 0 4n) or (0 1 4n+2)
- B sublattice, canted AF order along b axis: reflections of type (0 0 2n+1)
- Ratio of intensity of reflections can be used to determine canting angle

Locking of Ir magnetic moments to the correlated rotation of O octahedra in Sr_2IrO_4

Boseggia et al. JPCM (2013)



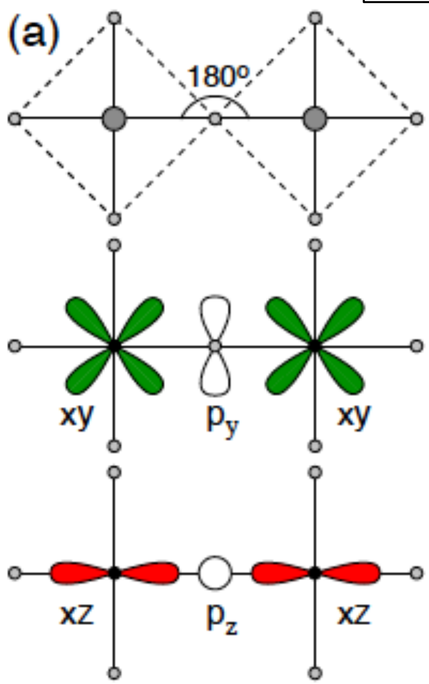
- Canting angle of magnetic moments of 12.2(8) degrees is within error equal to the rotation angle 11.8 degrees.
- Confirms key prediction of Jeff=1/2 model by Jackeli and Khaliullin
- Can used measured canting angle in theory to place constraints on tetragonal crystal field
- Conclude that Jeff=1/2 state is realised in Sr_2IrO_4



Novel Groundstates and Excitations in Iridates

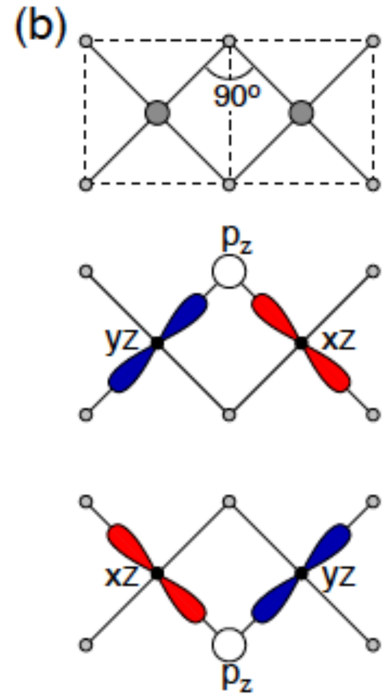
Exquisite sensitivity of interactions for Jeff=1/2 state to lattice topology

Landmark paper by Jackeli and Khaliullin, PRL (2009)



Isotropic
Heisenberg Exchange

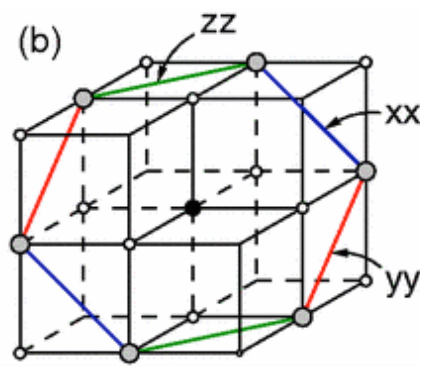
$$J_1 \mathbf{S}_i \cdot \mathbf{S}_j$$

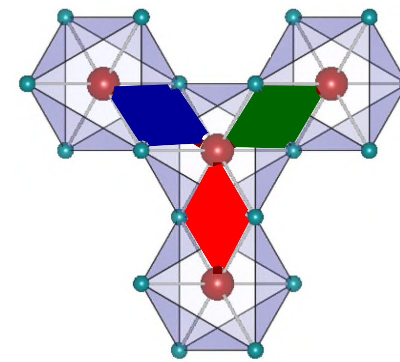


Anisotropic
Bond Directional

$$K \mathrm{S}_i^{\gamma} \mathrm{S}_j^{\gamma}$$

Honeycomb lattice





Kitaev Model

Kitaev-Heisenberg Model

$$\mathcal{H} = J \mathbf{S}_i \cdot \mathbf{S}_j + K \mathrm{S}_i^{\gamma} \mathrm{S}_j^{\gamma}$$

Predicted to give rise to novel phases and excitations