Anisotropic magnetic interactions in Iridium oxides from LDA+U calculations

Alexander Yaresko

Max Planck Institute for Solid State Research,
Stuttgart, Germany

What about U?
Effects of Hubbard Interactions and Hunds Coupling in Solids
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outline

1. motivations: j_{eff}=1/2 beauty
2. anisotropic exchange in honeycomb Na_2IrO_3
3. noncollinear ground state in Sr_2IrO_4 and $Sr_3Ir_2O_7$

anisotropic exchange interaction

for general bi-linear pair interaction between spins: $H = \sum_{i \neq j} \mathbf{S}_i^T \mathfrak{J}_{ij} \mathbf{S}_j$

where $\mathfrak{J}_{\alpha\beta}$ is a real 3×3 matrix: $\mathfrak{J} = \mathfrak{J}^+ + \mathfrak{J}^-$

with symmetric $\mathfrak{J}^+ = (\mathfrak{J} + \mathfrak{J}^T)/2$ and antisymmetric $\mathfrak{J}^- = (\mathfrak{J} - \mathfrak{J}^T)/2$

- no spin-orbit coupling (SOC) ($\mathfrak{J}^+_{\alpha\alpha} = J$, $\mathfrak{J}^+_{\alpha\beta} = 0$, $\mathfrak{J}^-_{\alpha\beta} = 0$)

$$\mathfrak{J} = \begin{pmatrix} J & 0 & 0 \\ 0 & J & 0 \\ 0 & 0 & J \end{pmatrix}; \qquad H = J \mathbf{S}_i \cdot \mathbf{S}_j$$

isotropic Heisenberg

- weak SOC, no inversion ($\mathfrak{J}^-_{xy} = D \neq 0$)

$$\mathfrak{J} = \begin{pmatrix} J & D & 0 \\ -D & J & 0 \\ 0 & 0 & J \end{pmatrix}; \qquad H = J \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot [\mathbf{S}_i \times \mathbf{S}_j]$$

antisymmetric Dzyaloshinsky-Moriya

- strong SOC (e.g., $\mathfrak{J}^+_{xx} = \mathfrak{J}^+_{yy} = J$, $\mathfrak{J}^+_{zz} = J + K$, $\mathfrak{J}^+_{\alpha\beta} \neq 0$,...)

$$\mathfrak{J} = \begin{pmatrix} J & 0 & 0 \\ 0 & J & 0 \\ 0 & 0 & J + K \end{pmatrix}; \qquad H = J \mathbf{S}_i \cdot \mathbf{S}_j + K S_i^z S_j^z$$

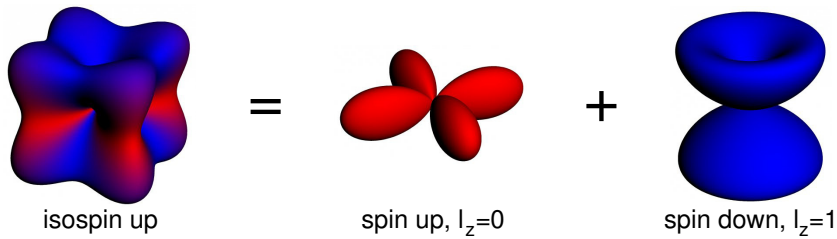
cubic crystal field + spin-orbit coupling (SOC)

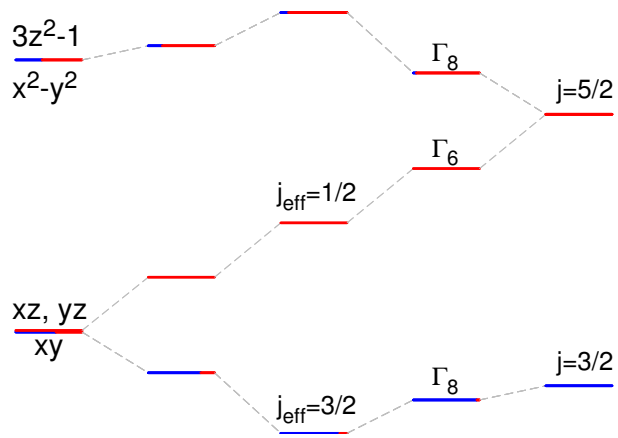
split t_{2g} states into a Γ_8 ($j_{\text{eff}}=3/2$; $d_{3/2}+d_{5/2}$) quartet:

$$\chi_{\Gamma_8} = \begin{cases} \sqrt{\frac{1}{2}}\left(d_{yz}\chi_{\pm\frac{1}{2}} \pm i d_{zx}\chi_{\pm\frac{1}{2}}\right) \\ \sqrt{\frac{1}{6}}\left(2d_{xy}\chi_{\mp\frac{1}{2}} \pm d_{yz}\chi_{\pm\frac{1}{2}} + i d_{zx}\chi_{\pm\frac{1}{2}}\right) \end{cases}$$

and a Γ_6 ($j_{\text{eff}}=1/2$; pure $d_{5/2}$) doublet:

$$\chi_{\Gamma_6} = \sqrt{\frac{1}{3}}\left(d_{xy}\chi_{\mp\frac{1}{2}} \mp d_{yz}\chi_{\pm\frac{1}{2}} + i d_{zx}\chi_{\pm\frac{1}{2}}\right)$$





$d_{3/2}$ and $d_{5/2}$

Ir^{4+} $5d^5$ ion in octahedral environment:

j_{eff}=1/2 half-filled

j_{eff}=3/2 completely filled

- Mott insulator already for moderate U
- $j_{\text{eff}} = \pm 1/2$ splitting is caused by U instead of J_H

j_{eff}=1/2 magnetism

PRL **102,** 017205 (2009) PHYSICAL REVIEW LETTERS week ending 9 JANUARY 2009

Mott Insulators in the Strong Spin-Orbit Coupling Limit: From Heisenberg to a Quantum Compass and Kitaev Models

G. Jackeli[1,*] and G. Khaliullin[1]

[1]*Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany*

(Received 21 August 2008; published 6 January 2009)

We study the magnetic interactions in Mott-Hubbard systems with partially filled t_{2g} levels and with strong spin-orbit coupling. The latter entangles the spin and orbital spaces, and leads to a rich variety of the low energy Hamiltonians that extrapolate from the Heisenberg to a quantum compass model depending on the lattice geometry. This gives way to ''engineer'' in such Mott insulators an exactly solvable spin model by Kitaev relevant for quantum computation. We, finally, explain ''weak'' ferromagnetism, with an anomalously large ferromagnetic moment, in Sr_2IrO_4.

corner-sharing octahedra (a):

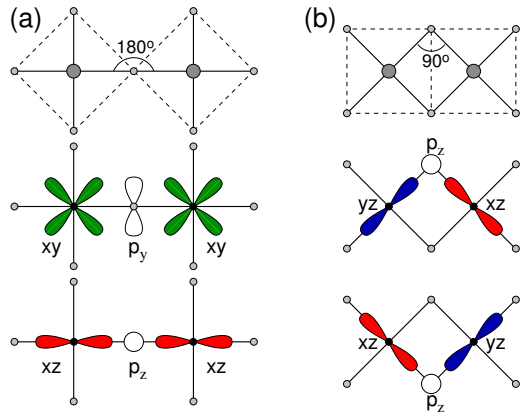
- Heisenberg + weak pseudo-dipolar interaction

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

- additional anisotropic terms if $\phi \neq 180°$

$$H = J_1 \mathbf{S}_i \cdot \mathbf{S}_j + J_z S_i^z S_j^z + \mathbf{D} \cdot [\mathbf{S}_i \times \mathbf{S}_j]$$

are responsible for weak FM in Sr_2IrO_4



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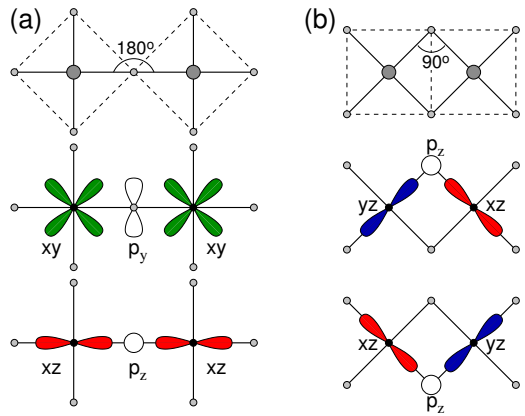
edge-sharing octahedra (b):

j_{eff}=1/2 hoppings via O_1 p_z and O_2 p_z cancel out

- isotropic superexchange J is suppressed
- strongly anisotropic interaction $K^{\alpha\beta}$



Kitaev-Heisenberg model:

$$H_{\text{HK}} = K^{\alpha\beta} S_i^\gamma S_j^\gamma + J \mathbf{S}_i \cdot \mathbf{S}_j$$

with exotic spin-liquid ground state

Can one get correct ground state and estimate $\mathfrak{J}$ from LDA+U calculations?

back to U_{1234}

Ir d occupation matrix $n^{\sigma\sigma'}_{mm'}$ has off-diagonal in spin terms ($n^{\sigma\,-\sigma}_{mm'} \neq 0$):

Coulomb energy:

$$E^U = \frac{1}{2}\sum_{\sigma,\{m\}}\Big[n^{\sigma\sigma}_{m_1m_2}(\langle m_1m_3|V_{ee}|m_2m_4\rangle - \langle m_1m_3|V_{ee}|m_4m_2\rangle)n^{\sigma\sigma}_{m_3m_4} + n^{\sigma\sigma}_{m_1m_2}\langle m_1m_3|V_{ee}|m_2m_4\rangle n^{-\sigma\,-\sigma}_{m_3m_4} - n^{\sigma\,-\sigma}_{m_1m_2}\langle m_1m_3|V_{ee}|m_4m_2\rangle n^{-\sigma\sigma}_{m_3m_4}\Big]$$

σ, m-dependent potential:

$$V^{\sigma\sigma'}_{mm'} = \frac{\partial(E^U - E^{dc})}{\partial n^{\sigma\sigma'}_{mm'}}, \quad E^{dc} = \frac{1}{2}UN(N-1) - \frac{1}{2}J\sum_{\sigma} N_{\sigma\sigma}(N_{\sigma\sigma}-1)$$

A. Liechtenstein, *et al* PRB **52**, R5467 (1995), AY, *et al* PRB **67**, 155103 (2003), ...

rotationally invariant LDA+U+SOC

- split $j_{\text{eff}} = 1/2$ states
- but does not change their wavefunction

how to estimate $\mathfrak{J}$?

1. to calculate the total (band) energy as a function of angle between spins using spin-spiral calculations and/or constraining magnetization direction
2. to map $\varepsilon(\{\phi\})$ onto an appropriate Heisenberg+Kitaev+... model

- spin-spiral calculations do not work with SOC
- tricky to impose constraints on magnetization direction in LDA+U calculations

but we can do:

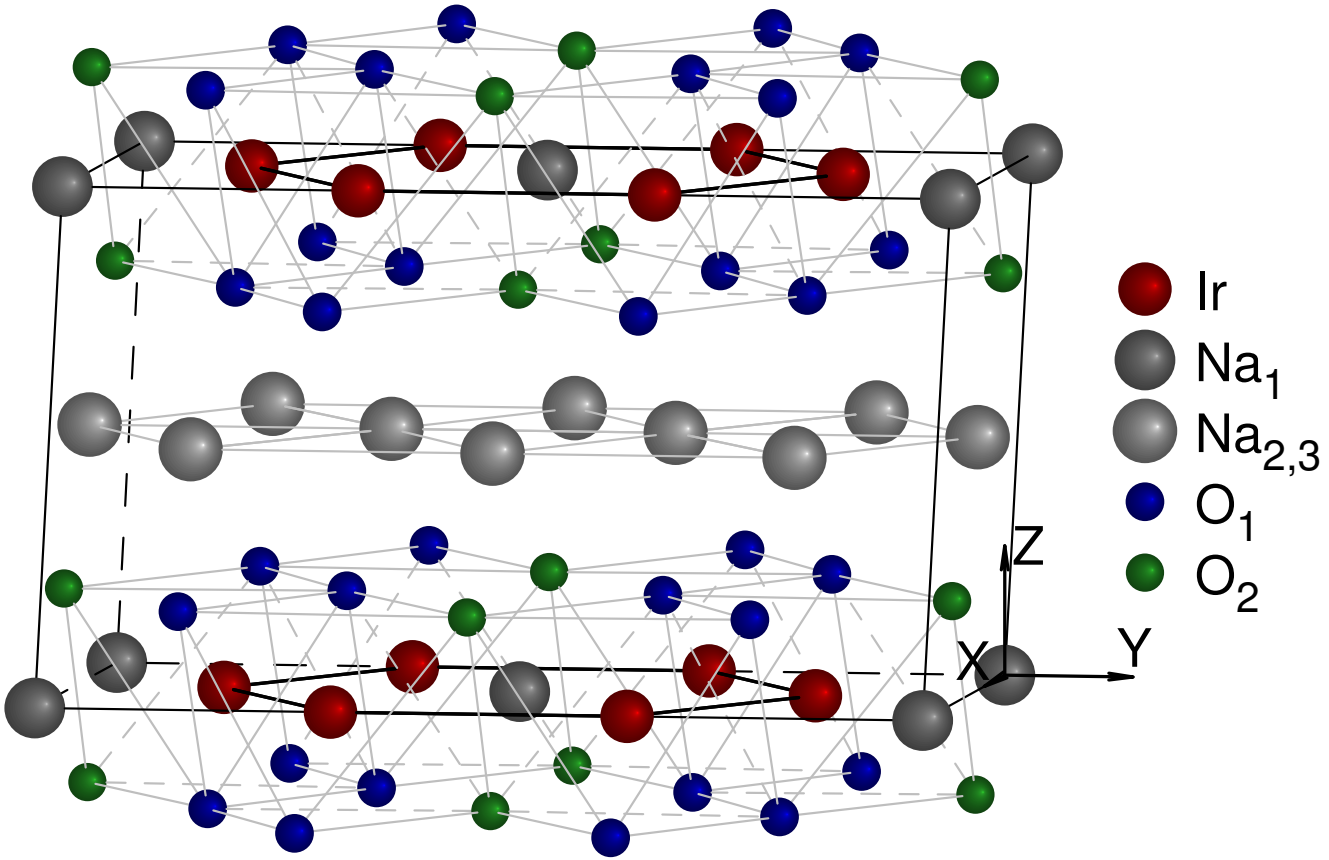
- calculations for magnetic configurations constrained by symmetry
  - limited number of magnetic configurations
  - not all exchange parameters can be determined simultaneously

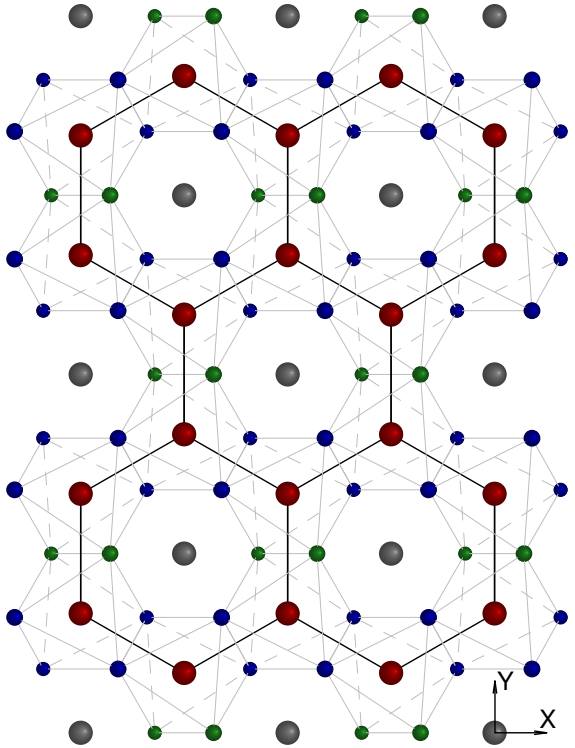
1. motivations: j_{eff}=1/2 beauty

2. anisotropic exchange in honeycomb Na_2IrO_3

3. noncollinear ground state in Sr_2IrO_4 and $Sr_3Ir_2O_7$

crystal structure

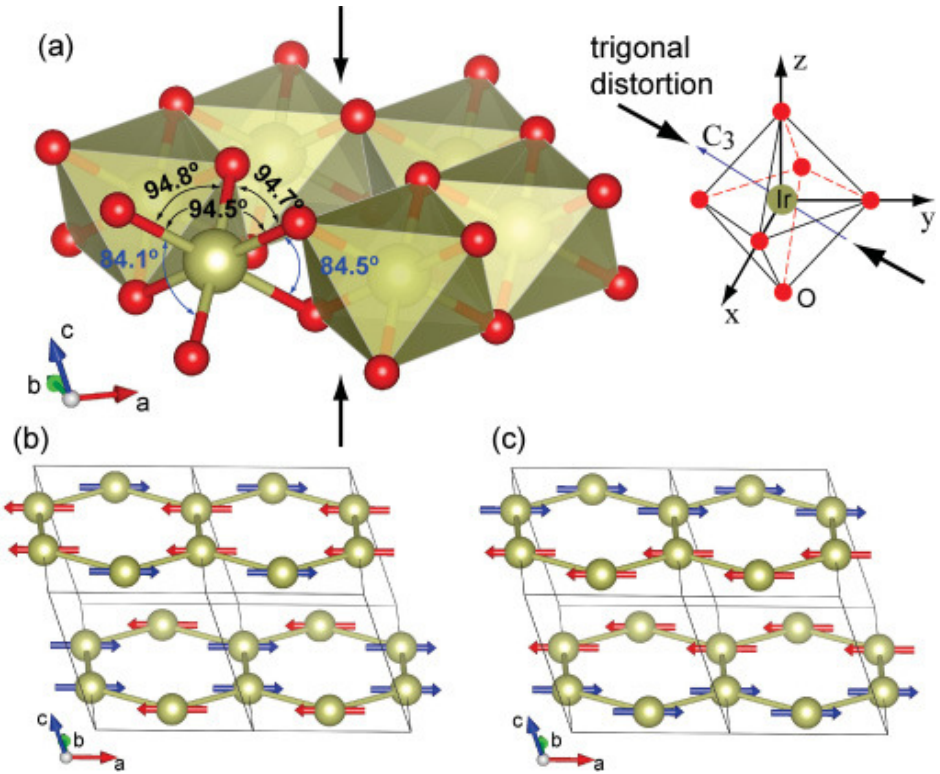




- monoclinic $C/2m$ space group
- honeycomb Ir layers separated by triangular Na layers
- trigonally distorted IrO_6 octahedra; Ir^{4+} d^5 with half-filled j_{eff}=1/2 states

S.K. Choi, *et al* PRL **108**, 127204 (2012), F. Ye, *et al* PRB **85**, 180403 (2012)

experimental magnetic structure



- zigzag order (c)
- ordered Ir moment 0.22 μ_B

F. Ye, *et al* PRB **85**, 180403 (2012)

explanations:

- isotropic Heisenberg model with long-ranged interactions
- Kitaev-Heisenberg model
- Kitaev-Heisenberg model + additional anisotropic exchanges Γ, Γ'

diffuse magnetic x-ray scattering results

LETTERS
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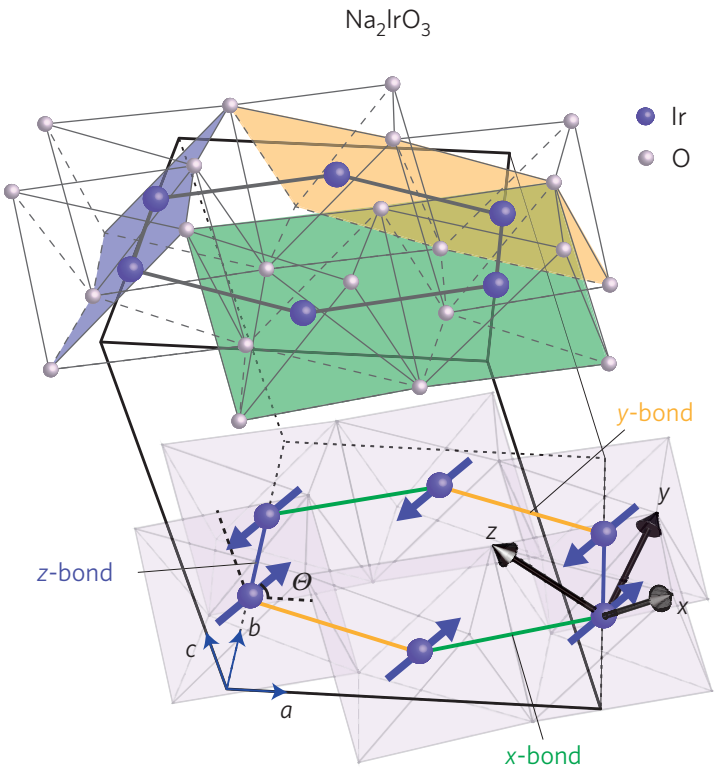
nature physics

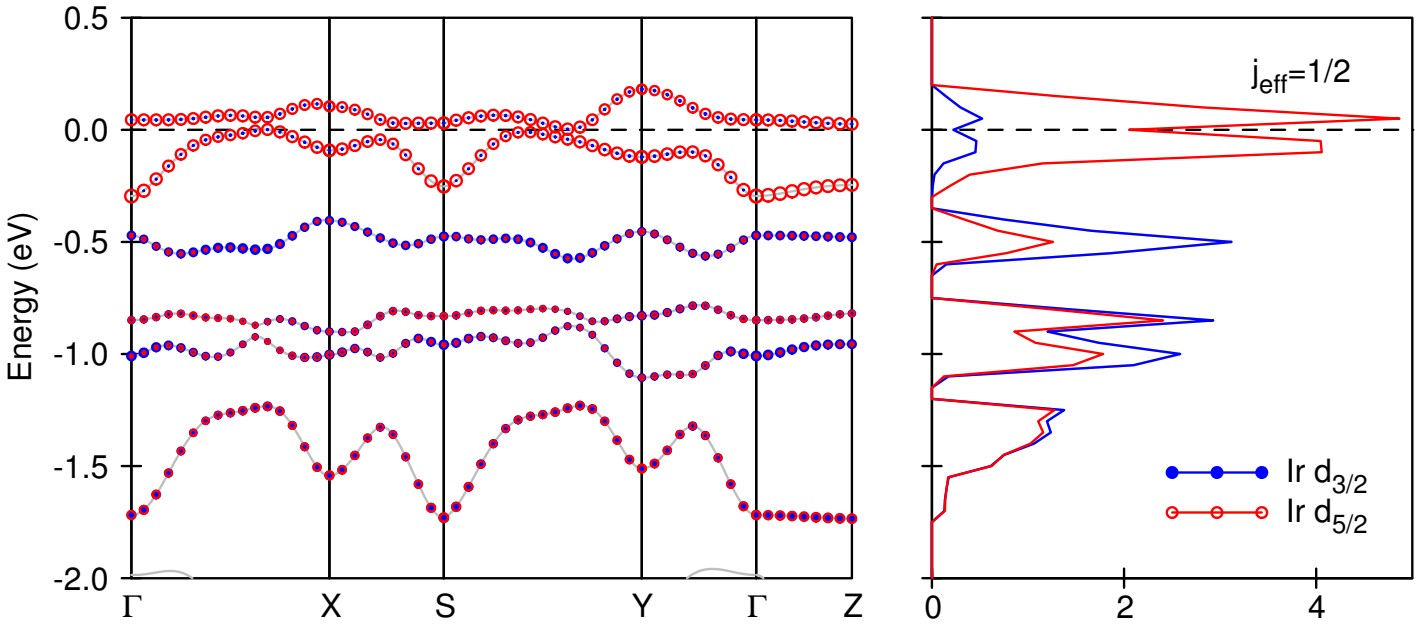
Direct evidence for dominant bond-directional interactions in a honeycomb lattice iridate Na_2IrO_3

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]*



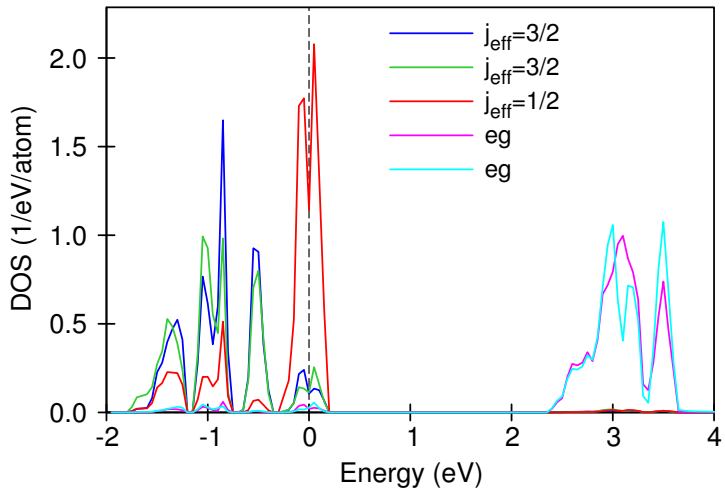
- zig-zag magnetic order
- Ir moments lie in ac plane
- form angle $\Theta = 44.3°$ with a axis
- scattering intensities above T_N are explained by strongly anisotropic exchange interactions

relativistic bands for Na_2IrO_3



- Ir t_{2g} bands are split into sub-bands due to formation of quasi-molecular orbitals (MO)

  I. Mazin, *et al* PRL **109**, 197201 (2012)
- highest MO are coupled by SOC
- dominant contribution of Ir $d_{5/2}$ states to bands crossing $E_F \Rightarrow \sim j_{\text{eff}} = 1/2$ states



8 distinct magnetic structures

- 4 inequivalent magnetic structures (magnetic groups) for the $C2/m$ cell and 4 for a doubled $P2/m$ cell
- all $\mathrm{Na}_{1,2,3}$, Ir, and $\mathrm{O}_{1,2}$ sites remain equivalent

C_{2y} rotation transforms each Ir site into itself: $C_{2y}\mathrm{Ir}_i = \mathrm{Ir}_i$

$C_{2y}m_x = -m_x$, $C_{2y}m_y = m_y$, $C_{2y}m_z = -m_z \quad \Rightarrow \quad m_x = m_z = 0$, $\mathbf{m}_{\mathrm{Ir}} || b$

$\hat{\Theta}C_{2y}m_x = m_x$, $\hat{\Theta}C_{2y}m_y = -m_y$, $\hat{\Theta}C_{2y}m_z = m_z \quad \Rightarrow \quad m_y = 0$, $\mathbf{m}_{\mathrm{Ir}} || ac$

	symmetry operations				$\mathbf{m}_{\mathrm{Ir}}\|\|$	
c_f	E	$-C_{2y}$	I	$-M_y$	ac	ferro
c_f	E	C_{2y}	I	M_y	b	
c_a	E	$-C_{2y}$	$-I$	M_y	ac	Néel
c_a	E	C_{2y}	$-I$	$-M_y$	b	
p_z	E	$-C_{2y}$	$-I$	M_y	ac	zig-zag
p_z	E	C_{2y}	$-I$	$-M_y$	b	
p_s	E	$-C_{2y}$	I	$-M_y$	ac	stripe
p_s	E	C_{2y}	I	M_y	b	

- "$-$" means that rotation is followed by time reversal $\hat{\Theta} = -i\sigma_y\hat{K}$
- $\mathbf{m}_{\mathrm{Ir}} || ac$: Ir magnetization direction is defined by polar angle θ
- Ir moments are collinear, self-consistency in Ir, O, Na magnetization directions

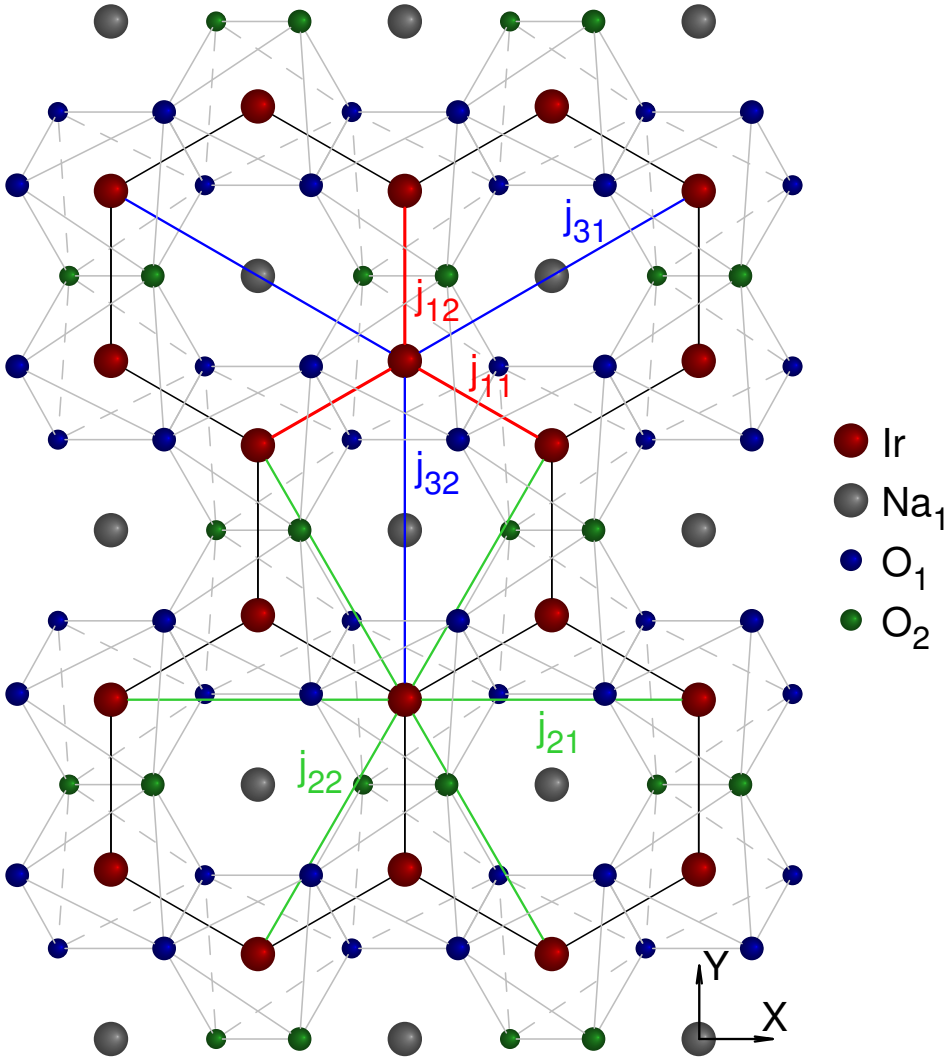
summary for 8 configurations

- LDA+U with U=2.1 eV; J=0.6 eV $\Rightarrow$ U_{eff}=$U - J$=1.5 eV
- for Sr_2IrO_4 U_{eff}=1.3 eV gives good agreement with optical spectra

	$\mathbf{M}$	ε (meV)	θ	ϕ	m_s (μ_B)	m_l (μ_B)
c_f	ac	-5.5	18.8	180	0.32	0.40
c_f	b	0.	90.0	90	0.20	0.43
c_a	ac	-10.9	14.2	180	0.24	0.35
c_a	b	-12.4	90.0	90	0.14	0.38
p_z	ac	-16.7	26.4	0	0.25	0.34
p_z	b	-14.9	90.0	90	0.18	0.39
p_s	ac	-8.7	138.5	0	0.26	0.36
p_s	b	0.5	90.0	90	0.16	0.42

- correct ground state with zig-zag order and Ir moments rotated away from a
- insulating solutions for all magnetic orders
- zig-zag ground state also for U_{eff}=1.0 and 2.0 eV
- θ does not depend on U_{eff} but depends on SOC strength ξ

Heisenberg model



- interlayer coupling is neglected
- ideal honeycomb lattice is assumed

isotropic exchange:

n: $J_{11} = J_{12} \equiv J_1$

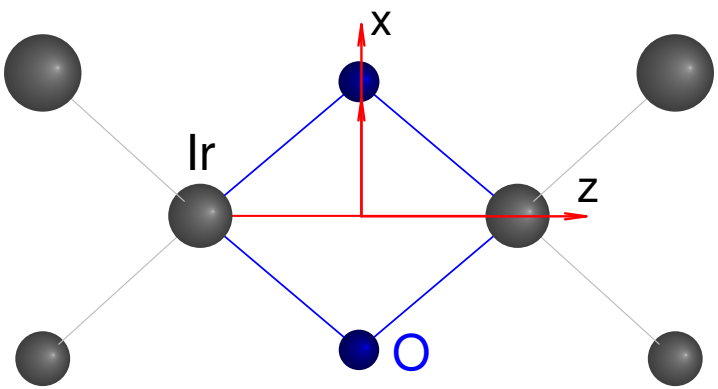
nn: $J_{21} = J_{22} \equiv J_2$

nnn: $J_{31} = J_{32} \equiv J_3$

anisotropic exchanges

Ir-Ir bond in ideal honeycomb lattice: D_{2h} $(E,\, C_{2x},\, C_{2y},\, C_{2z},\, I,\, M_x,\, M_y,\, M_z)$

Ir-Ir bond in Na_2IrO_3: C_{2h} $(E,\, C_{2z},\, I,\, M_z)$



inversion symmetry $\Rightarrow \mathfrak{J}^- = 0$

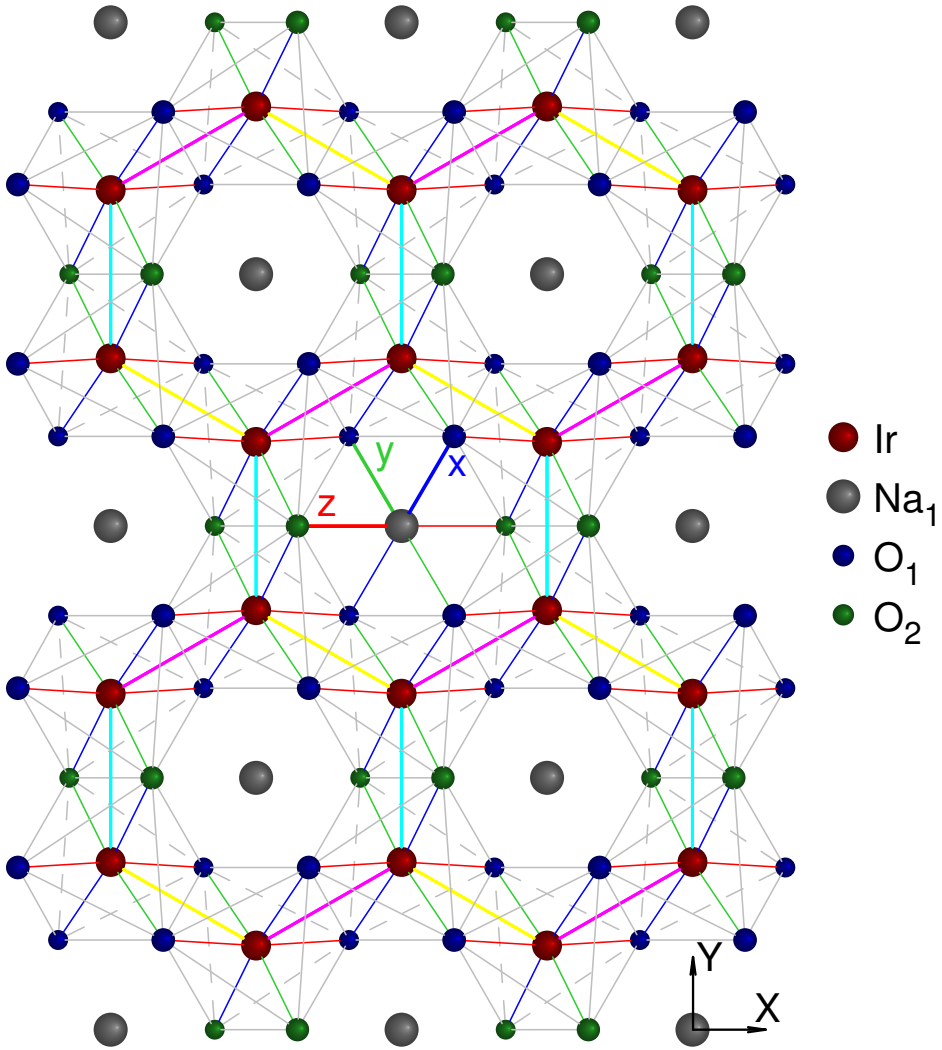
$$\mathfrak{J}^+ = \begin{pmatrix} \mathfrak{J}^+_{xx} & \mathfrak{J}^+_{xy} & 0 \\ \mathfrak{J}^+_{xy} & \mathfrak{J}^+_{yy} & 0 \\ 0 & 0 & \mathfrak{J}^+_{zz} \end{pmatrix}, \quad \mathfrak{J}^+_{xx} \neq \mathfrak{J}^+_{yy} \neq \mathfrak{J}^+_{zz}$$

or isotropic J_0 + traceless symmetric part

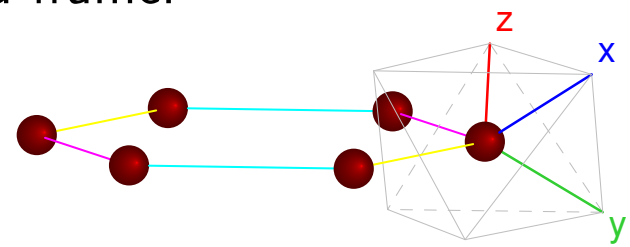
$$\mathfrak{J}^+ = J_0 \cdot \hat{I} + \begin{pmatrix} B & C & 0 \\ C & -B-A & 0 \\ 0 & 0 & A \end{pmatrix}$$

V. Katukuri, *et al* NJP **16**, 013056 (2014)

anisotropic exchange in a rotated fame



rotated frame:



x, y, and z point to nearest O

Ir-Ir bonds along xy, yz, zx

$$\mathfrak{J}^{xy} = \begin{pmatrix} J & \Gamma & -\Gamma' \\ \Gamma & J & \Gamma' \\ -\Gamma' & \Gamma' & J+K \end{pmatrix}$$

$$K = -\tfrac{3}{2}(A+B),\ J = J_0 - K/3,$$

$$\Gamma = (A-B)/2\ ,\ \Gamma' = C/\sqrt{2}$$

Kitaev-like terms:

$$K_{ij}^{xy} S_i^z S_j^z,\ K_{ij}^{yz} S_i^x S_j^x,\ K_{ij}^{zx} S_i^y S_j^y$$

$$K^{xy} \neq K^{yz} = K^{zx} \approx K$$

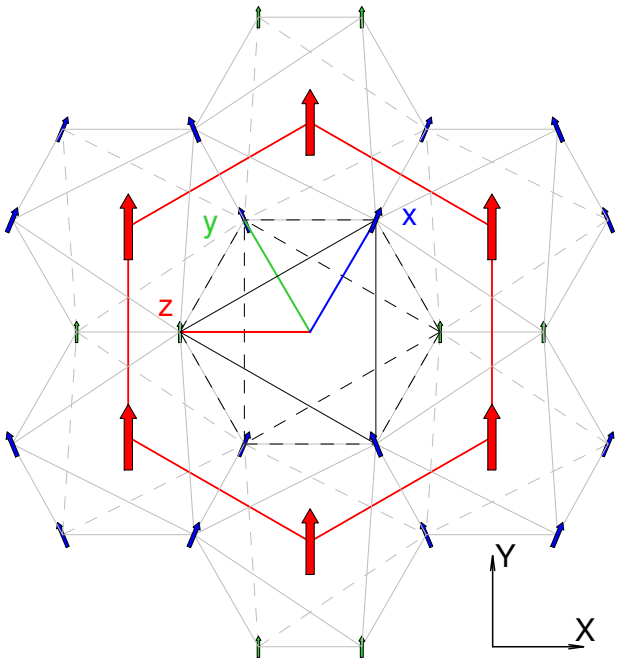
Γ-terms:

$$\Gamma_{ij}^{xy}(S_i^x S_j^y + S_i^y S_j^x), \ldots$$

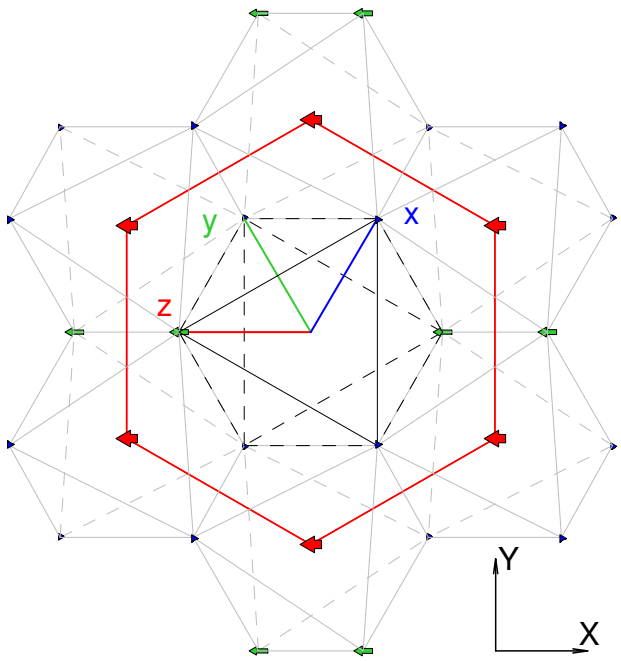
J.G. Rau, *et al* PRL **112**, 077204 (2014), J. Chaloupka and G. Khaliullin, PRB **92**, 024413 (2015)

FM order (c_f)

$\mathbf{M}||Y$ $(\mathbf{M}||b)$



$\mathbf{M} \perp Y$ $(\mathbf{M}||ac)$



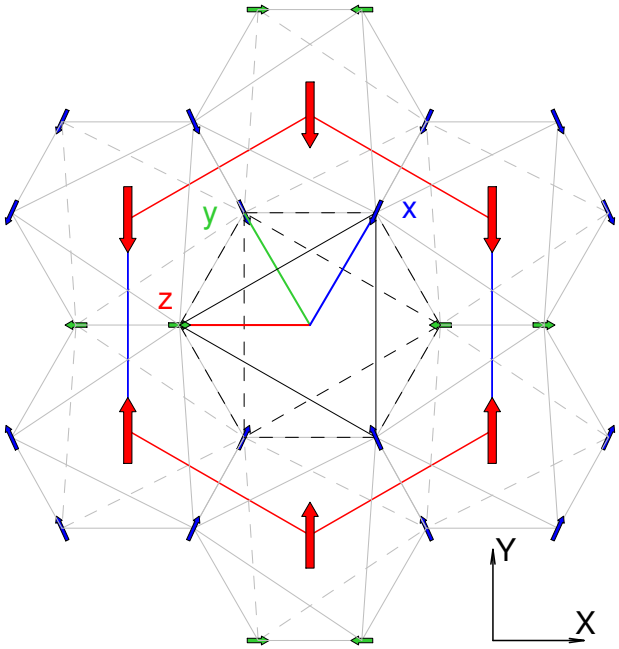
$$\varepsilon = \quad 6J_1 + 12J_2 + 6J_3$$
$$+2K$$
$$\varepsilon = 0.$$

$$\varepsilon = \quad 6J_1 + 12J_2 + 6J_3$$
$$+2K$$
$$\varepsilon = -5.5 \text{ meV}, \theta = 18.8°, \phi = 180°$$

- all energies are per f.u. and relative to $\varepsilon_{cf}(\mathbf{m}||b)$; FM (AF) bonds
- $\theta \neq 0$, $\Delta\varepsilon = \varepsilon(ac) - \varepsilon(b) \neq 0 \Rightarrow \Gamma$-term or $K_a \neq K_b$?
- noncollinear O moments (0.04–0.09 μ_B) $\Rightarrow$ anisotropic interactions

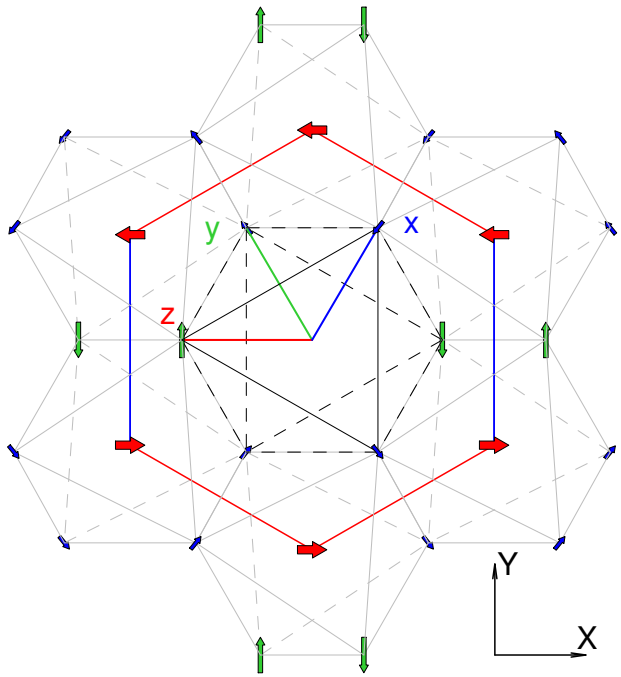
zig-zag (p_z)

$\mathbf{M}||Y$ $(\mathbf{M}||b)$



$$\varepsilon = \quad 2J_1 - \quad 4J_2 - 6J_3$$
$$+2K$$
$$\varepsilon = -14.9 \text{ meV}$$

$\mathbf{M} \perp Y$ $(\mathbf{M}||ac)$



$$\varepsilon = \quad 2J_1 - \quad 4J_2 - 6J_3$$
$$+2K(\cos 2\theta + 2\sqrt{2}\sin 2\theta)/3$$
$$\varepsilon = -16.7 \text{ meV}, \ \theta = 26.4°, \ \phi = 0$$

correct ground state with zig-zag order; $\theta = 26.4° < \theta_{exp} = 45.7°$ $(90° - 44.3°)$

least-square fits to J_{123}-K-Γ models

		ε_{calc}	ε_{JK}	$\varepsilon_{JK\Gamma}$	ε_{123}	$\varepsilon_{123K\Gamma}$
c_f	ac	-5.5	0.0	-3.8	0.0	-1.9
c_f	b	0.0	0.0	0.0	0.0	0.0
c_a	ac	-10.9	-10.5	-9.0	-11.7	-10.7
c_a	b	-12.4	-10.5	-13.0	-11.7	-12.6
p_z	ac	-16.7	-8.6	-6.5	-15.8	-14.3
p_z	b	-14.9	-9.0	-12.4	-15.8	-17.3
p_s	ac	-8.7	-17.1	-15.8	-4.1	-8.6
p_s	b	0.5	-1.6	-0.7	-4.1	0.4
χ			0.30	0.27	0.18	0.10

JK $J_1 = 9.0$, $K = -16.4$ meV

incorrect p_s^{ac} ground state (in agreement with the J-K model)

$JK\Gamma$ $J_1 = 6.7$, $K = -12.8$, $\Gamma = 5.6$ meV

the p_s^{ac} ground state is still incorrect

123 $J_1 = -0.1$, $J_2 = 1.0$, $J_3 = 3.9$ meV

correct p_z ground state but: $\varepsilon(ac) = \varepsilon(b)$; $J_3 > J_2 \gg J_1$

$123K\Gamma$ $J_1 = 2.7$, $J_2 = 0.5$, $J_3 = 3.2$, $K = -8.0$, $\Gamma = 2.8$ meV

best fit although $\varepsilon(p_z^b) > \varepsilon(p_z^{ac})$

least-square fits to J_{123}-K-Γ models

		ε_{calc}	ε_{JK}	$\varepsilon_{JK\Gamma}$	ε_{123}	$\varepsilon_{123K\Gamma}$
c_f	ac	-5.5	0.0	-3.8	0.0	-1.9
c_f	b	0.0	0.0	0.0	0.0	0.0
c_a	ac	-10.9	-10.5	-9.0	-11.7	-10.7
c_a	b	-12.4	-10.5	-13.0	-11.7	-12.6
p_z	ac	-16.7	-8.6	-6.5	-15.8	-14.3
p_z	b	-14.9	-9.0	-12.4	-15.8	-17.3
p_s	ac	-8.7	-17.1	-15.8	-4.1	-8.6
p_s	b	0.5	-1.6	-0.7	-4.1	0.4
χ			0.30	0.27	0.18	0.10

- $123K\Gamma$: $J_1 = 2.7$, $J_2 = 0.5$, $J_3 = 3.2$, $K = -8.0$, $\Gamma = 2.8$ meV
- $(J,K,D = -\Gamma)$=(1.1,−0.7,−0.7) meV for $\phi_{(\mathrm{IrOIr})} = 90°$
  (J,K,D)=(1.4,−10.9,−2.1) meV for $\phi_{(\mathrm{IrOIr})} = 98.5°$

  V. Katukuri, *et al* NJP **16**, 013056 (2014)
- $J = 5.3$ meV, $K = -7$ meV, $\Gamma = 9.3$ meV, $\sqrt{2}\Gamma' = -6.6$ meV

  J. Chaloupka and G. Khaliullin, PRB **92**, 024413 (2015)

conclusions for Na_2IrO_3

- LSDA+U calculations reproduce correct zig-zag magnetic order in Na_2IrO_3 although the Ir magnetization direction seems to be too far away from ab plane compared to the experiment
- Calculated total energies and magnetization directions cannot be explained using the isotropic J_1–J_3 Heisenberg model
- Best fit is obtained when the anisotropic K and Γ terms are added

1. motivations: j_{eff}=1/2 beauty

2. anisotropic exchange in honeycomb Na_2IrO_3

3. noncollinear ground state in Sr_2IrO_4 and $Sr_3Ir_2O_7$

canted AFM order in Sr_2IrO_4

Phase-Sensitive Observation of a Spin-Orbital Mott State in Sr_2IrO_4

B. J. Kim,[1,2]* H. Ohsumi,[3] T. Komesu,[3] S. Sakai,[3,4] T. Morita,[3,5] H. Takagi,[1,2]* T. Arima[3,6]

Measurement of the quantum-mechanical phase in quantum matter provides the most direct manifestation of the underlying abstract physics. We used resonant x-ray scattering to probe the relative phases of constituent atomic orbitals in an electronic wave function, which uncovers the unconventional Mott insulating state induced by relativistic spin-orbit coupling in the layered 5*d* transition metal oxide Sr_2IrO_4. A selection rule based on intra-atomic interference effects establishes a complex spin-orbital state represented by an effective total angular momentum = 1/2 quantum number, the phase of which can lead to a quantum topological state of matter.

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with Ir moments aligned in ab plane:

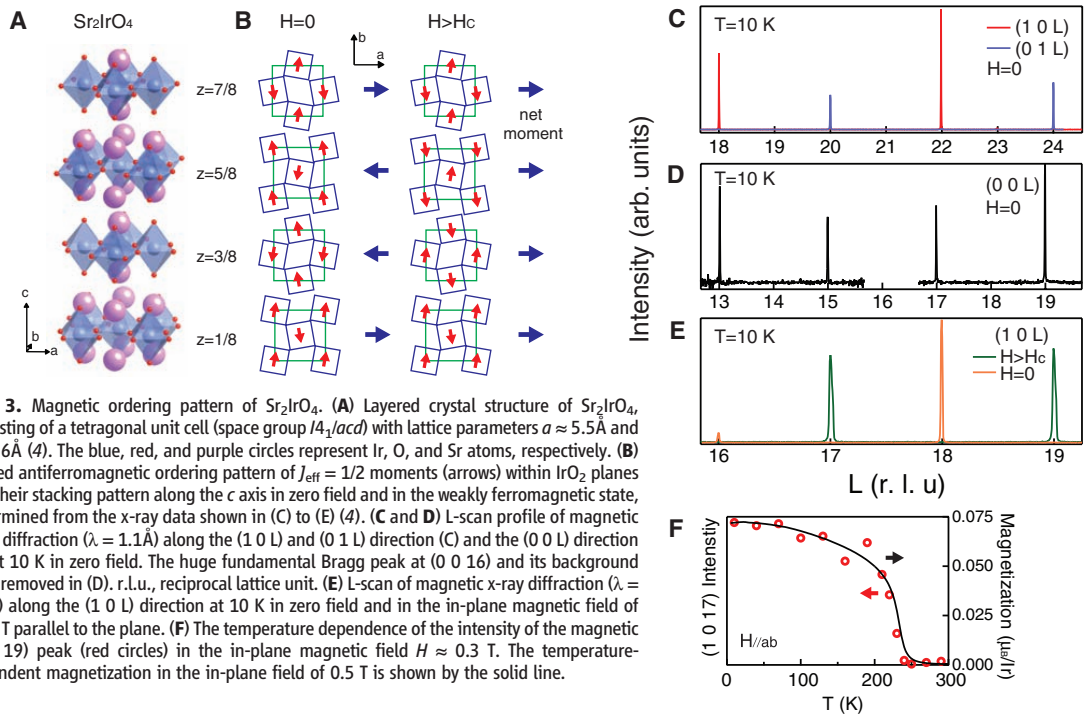

Fig. 3. Magnetic ordering pattern of Sr_2IrO_4. (**A**) Layered crystal structure of Sr_2IrO_4, consisting of a tetragonal unit cell (space group $I4_1/acd$) with lattice parameters $a \approx 5.5$Å and $c \approx 26$Å (*4*). The blue, red, and purple circles represent Ir, O, and Sr atoms, respectively. (**B**) Canted antiferromagnetic ordering pattern of $J_{eff} = 1/2$ moments (arrows) within IrO_2 planes and their stacking pattern along the *c* axis in zero field and in the weakly ferromagnetic state, determined from the x-ray data shown in (C) to (E) (*4*). (**C** and **D**) L-scan profile of magnetic x-ray diffraction ($\lambda = 1.1$Å) along the (1 0 L) and (0 1 L) direction (C) and the (0 0 L) direction (D) at 10 K in zero field. The huge fundamental Bragg peak at (0 0 16) and its background were removed in (D). r.l.u., reciprocal lattice unit. (**E**) L-scan of magnetic x-ray diffraction (λ = 1.1Å) along the (1 0 L) direction at 10 K in zero field and in the in-plane magnetic field of ≈0.3 T parallel to the plane. (**F**) The temperature dependence of the intensity of the magnetic (1 0 19) peak (red circles) in the in-plane magnetic field $H \approx 0.3$ T. The temperature-dependent magnetization in the in-plane field of 0.5 T is shown by the solid line.

collinear AFM order in $Sr_3Ir_2O_7$

with Ir moments aligned along c:

PRL **109,** 037204 (2012) PHYSICAL REVIEW LETTERS week ending 20 JULY 2012

Dimensionality Driven Spin-Flop Transition in Layered Iridates

J. W. Kim,[1] Y. Choi,[1] Jungho Kim,[1] J. F. Mitchell,[2] G. Jackeli,[3] M. Daghofer,[4] J. van den Brink,[4] G. Khaliullin,[3] and B. J. Kim[2,*]

[1]*Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, USA*
[2]*Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA*
[3]*Max Planck Institute for Solid State Research, Heisenbergstrasse 1, D-70569 Stuttgart, Germany*
[4]*Institute for Theoretical Solid Sate Physics, IFW Dresden, Helmholtzstrasse 20, 01069 Dresden, Germany*

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Using resonant x-ray diffraction, we observe an easy c-axis collinear antiferromagnetic structure for the bilayer $Sr_3Ir_2O_7$, a significant contrast to the single layer Sr_2IrO_4 with in-plane canted moments. Based on a microscopic model Hamiltonian, we show that the observed spin-flop transition as a function of number of IrO_2 layers is due to strong competition among intra- and interlayer bond-directional pseudodipolar interactions of the spin-orbit entangled $J_{\text{eff}} = 1/2$ moments. With this we unravel the origin of anisotropic exchange interactions in a Mott insulator in the strong spin-orbit coupling regime, which holds the key to the various types of unconventional magnetism proposed in 5d transition metal oxides.

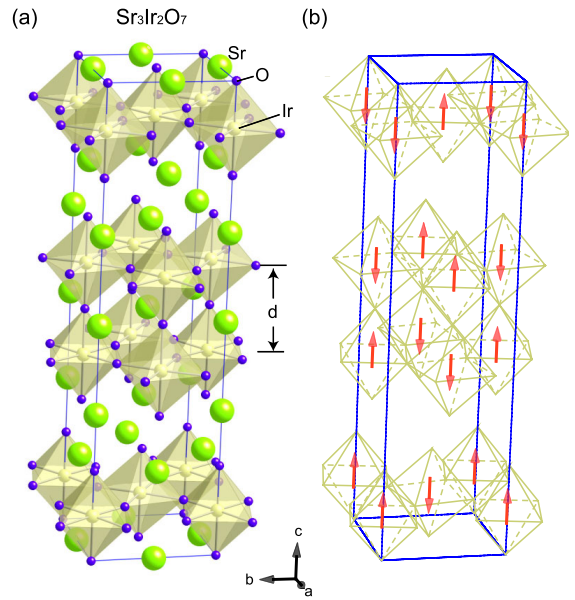


FIG. 1 (color online). (a) Crystal structure of $Sr_3Ir_2O_7$ as reported in Ref. [17]. Every neighboring IrO_6 octahedra are rotated in opposite sense about the c axis by $\simeq 12°$. (b) Magnetic order has a c-axis collinear G-type antiferromagnetic structure. The up and down magnetic moments correlate with counterclockwise and clockwise rotations of the IrO_6 octahedra, respectively.

$Sr_3Ir_2O_7$: crystal structure

$I4/mmm$ No. 139 space group: a=3.896 Å, c=20.879 Å

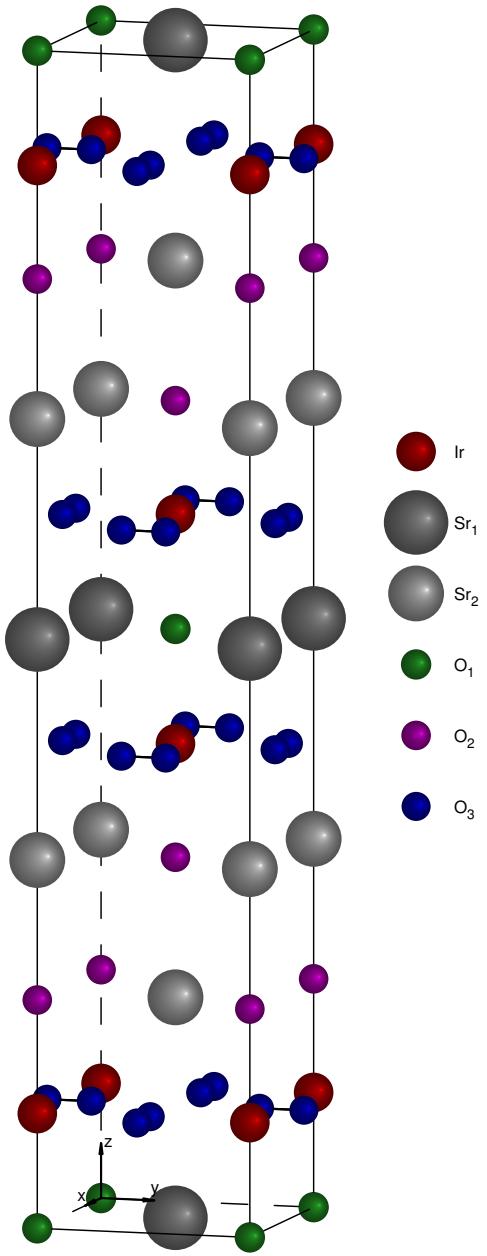
ion	W	x	y	z	occ.
Ir_1^{4+}	4e	0	0	0.09743	1.
Sr_1^{2+}	2b	0.5	0.5	0	1.
Sr_2^{2+}	4e	0.5	0.5	0.1872	1.
O_1^{2-}	2a	0	0	0	1.
O_2^{2-}	4e	0	0	0.1939	1.
O_3^{2-}	16n	0.1043	0.5	0.0960	0.5

M. A. Subramanian,. . . MRB **29**, 645 (1994)

Clock- or counterclockwise rotations of IrO_6 octahedra around c (ϕ=11.8°) $\Rightarrow$ averaged O_3 positions

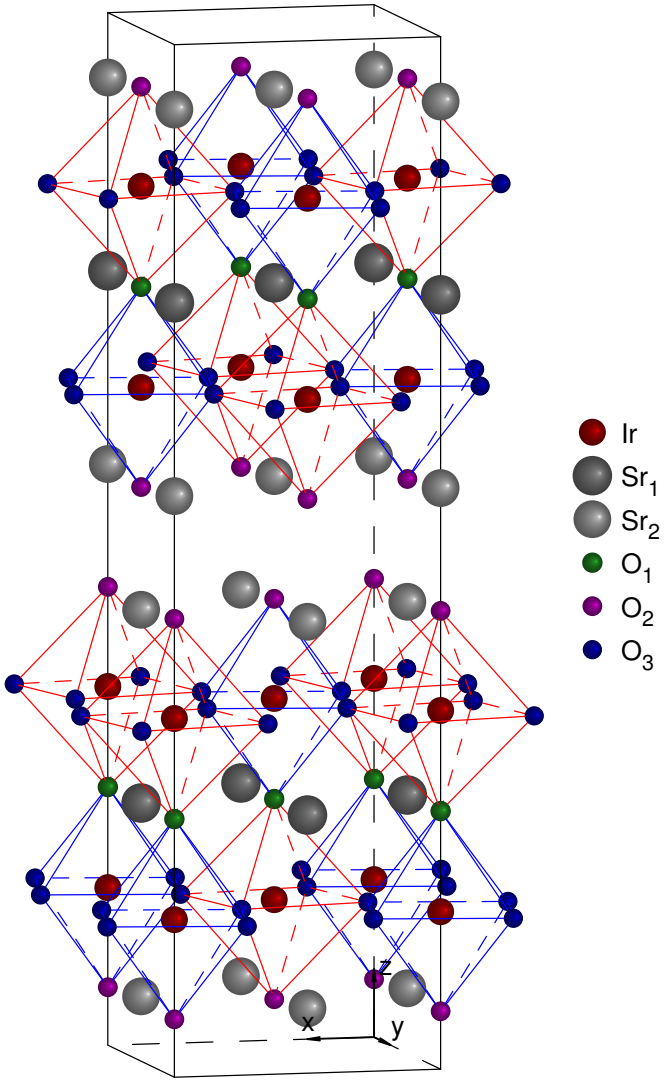
TEM: $Bbcb$ ($\equiv Acaa$) No. 68 space group
rotations in opposite senses in each bilayer

H. Matsuhata,. . . JSSC **177**, 3776 (2004)

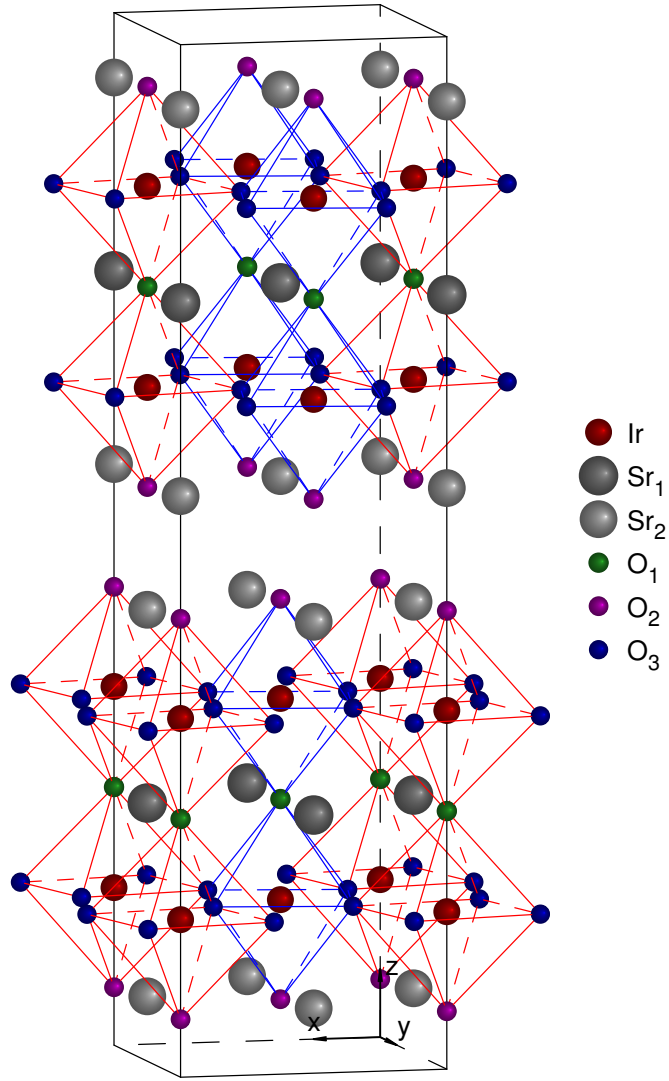


two structural models for $Sr_3Ir_2O_7$

$Acaa$ D_{2h} No. 68 (A_o)



$Acam$ D_{2h} No. 64 (F_o)



with clockwise or counterclockwise rotated octahedra

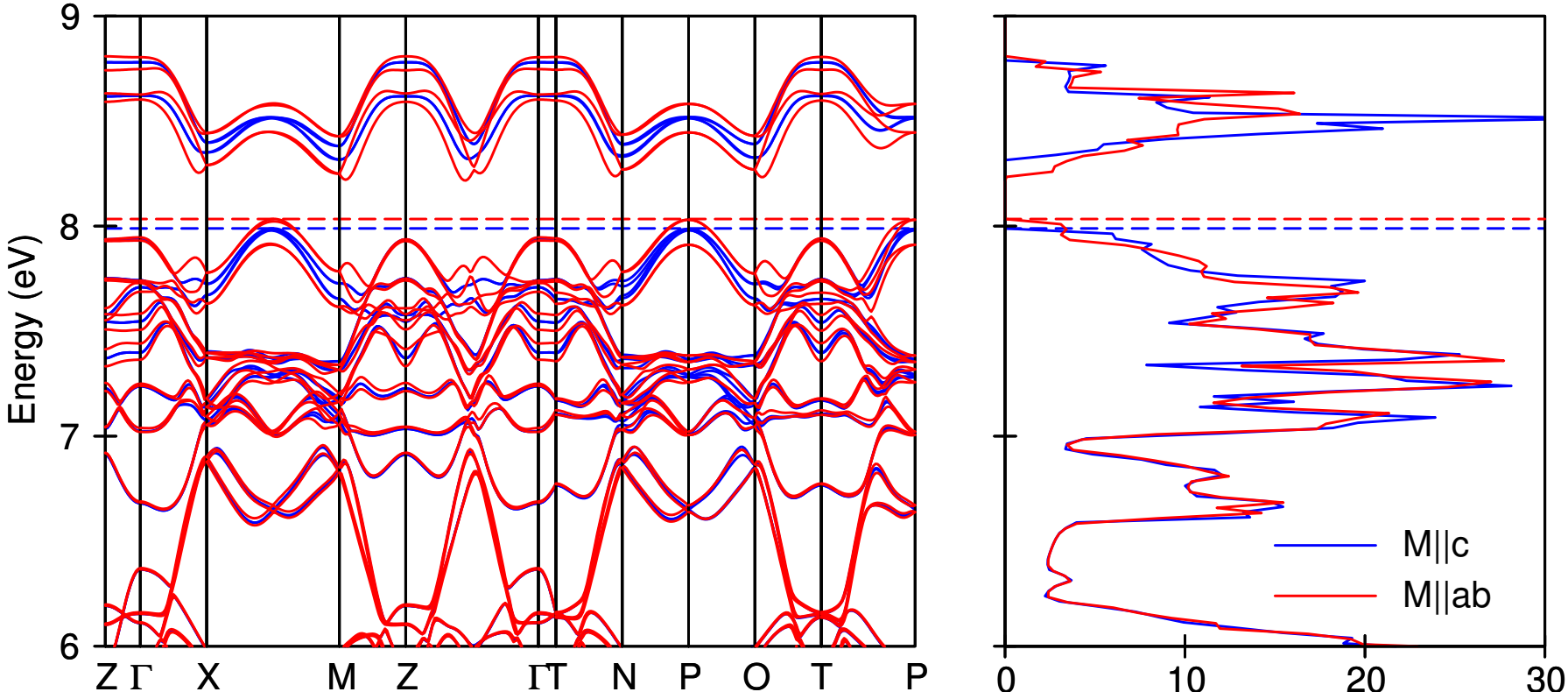
total energies

$Sr_3Ir_2O_7$			U_{eff}=1.0 eV		U_{eff}=1.5 eV		U_{eff}=2.0 eV	
OO	MO	$\mathbf{M}$	ΔE	E_g	ΔE	E_g	ΔE	E_g
A_o	AA_c	c	0	0.13	0	0.33	0	0.53
		ab	7.4	0	6.8	0.20	6.3	0.41
	AF_c	c	10.0	0	20.8	0.08	19.8	0.31
		ab	6.6	0.05	9.1	0.23	10.6	0.43
F_o	AA_c	c	54.8	0.14	50.4	0.33	48.1	0.52
		ab	52.9	0.15	49.0	0.33	47.5	0.51
	AF_c	c	67.7	0	71.3	0.12	68.5	0.35
		ab	67.0	0	69.6	0.13	67.7	0.35
Sr_2IrO_4								
	A	c	0	0.14	0	0.35	0	0.58
		ab	-1.3	0.15	-1.1	0.36	-0.7	0.58

- Sr_2IrO_4: lowest energy for $\mathbf{M}||ab$; $Sr_3Ir_2O_7$: lowest energy for $\mathbf{M}||c$
- $Sr_3Ir_2O_7$: A_o order of octahedra and A_c order in a bilayer are always favorable
- $Sr_3Ir_2O_7$: for F_o order of octahedra $\mathbf{M}||ab$ gives lower energy

$Sr_3Ir_2O_7$: bilayer splitting

AF order in a bilayer:



- $\mathbf{M}||c$: Ir moments are antiparallel $\Rightarrow$ small bilayer splitting
- $\mathbf{M}||ab$: the angle between moments $\phi_c = 137^\circ$
  $\Rightarrow$ bilayer splitting is much stronger

conclusions

- LSDA+U calculations reproduce magnetic ground states for Sr_2IrO_4 and $Sr_3Ir_2O_7$
- in $Sr_3Ir_2O_7$ bilayer splitting of unoccupied Ir t_{2g} bands increases strongly when Ir moments are in ab plane