Anisotropy of exchange interactions in Ir oxides from band structure calculations

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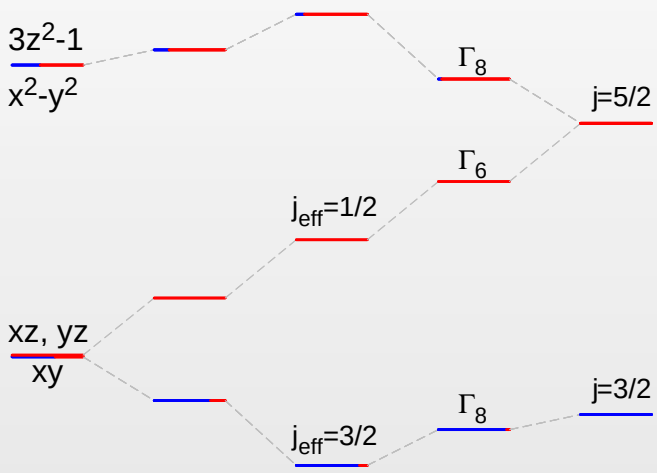
motivations: j_{eff}=1/2 physics

cubic crystal field + spin-orbit coupling (SOC)



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

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

$$\chi_{\Gamma_8}(t_{2g}) \approx \chi_{\Gamma_8}(d_{3/2})$$

which mixes with $\chi_{\Gamma_8}(e_g) \approx \chi_{\Gamma_8}(d_{5/2})$

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

$$\chi_{\Gamma_6} = \sqrt{\frac{1}{6}}\chi_{\frac{5}{2}\pm\frac{5}{2}} - \sqrt{\frac{5}{6}}\chi_{\frac{5}{2}\mp\frac{3}{2}}$$

or

$$\chi_{\Gamma_6} = \sqrt{\frac{1}{3}}\left[d_{xy}\chi_{\mp\frac{1}{2}} \mp d_{yz}\chi_{\pm\frac{1}{2}} + id_{zx}\chi_{\pm\frac{1}{2}}\right]$$

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

j_{eff}=1/2 half-filled

j_{eff}=3/2 completely filled

$\Downarrow$

Mott insulator already for moderate U

j_{eff}=1/2 magnetism

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

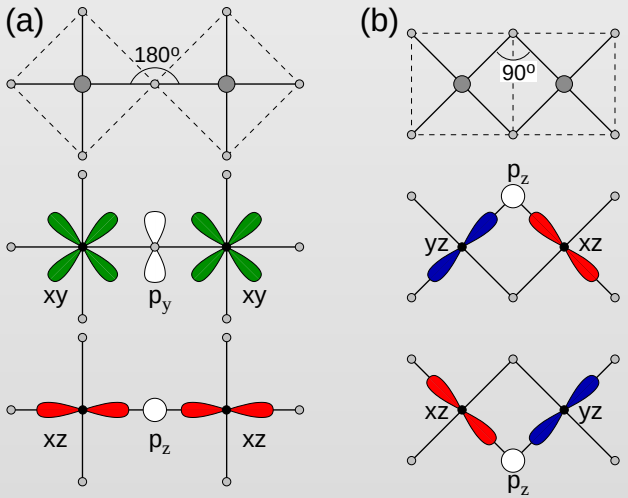
Mott Insulators in the Strong Spin-Orbit Coupling Limit: From Heisenberg to a Quantum Compass and Kitaev Models

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.



edge-sharing octahedra:

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 $K^{\alpha\beta}$

Heisenberg-Kitaev model:

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

where $S^{\gamma} \perp \alpha\beta$ plane

with exotic spin-liquid ground state

Can one estimate J and $K^{\alpha\beta}$ from LSDA($+U$) band structure calculations?

L(S)DA+U when SOC is strong

$n^{\sigma\sigma'}_{mm'}$: occupation matrix of localized states

$N_{\sigma\sigma'} = \sum_m n^{\sigma\,-\sigma}_{mm} \Rightarrow$ new spin frame in which $N_{\sigma\sigma'}$ is diagonal: $N_{\sigma\,-\sigma} = 0$:

strong SOC $\Rightarrow$ even in the new frame with $z' || \mathbf{m}$ $n^{\sigma\,-\sigma}_{mm'} \neq 0$

Coulomb energy:

$$
\begin{aligned}
E^U \;=\; & \frac{1}{2} \sum_{\sigma,\{m\}} \Big[n^{\sigma\sigma}_{m_1 m_2} (\langle m_1 m_3 | V_{ee} | m_2 m_4 \rangle - \langle m_1 m_3 | V_{ee} | m_4 m_2 \rangle) n^{\sigma\sigma}_{m_3 m_4} \\
& + n^{\sigma\sigma}_{m_1 m_2} \langle m_1 m_3 | V_{ee} | m_2 m_4 \rangle n^{-\sigma\,-\sigma}_{m_3 m_4} - n^{\sigma\,-\sigma}_{m_1 m_2} \langle m_1 m_3 | V_{ee} | m_4 m_2 \rangle n^{-\sigma\sigma}_{m_3 m_4} \Big]
\end{aligned}
$$

σ, m-dependent potential

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

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

problems:

- spin-spiral calculations cannot be done with SOC
- how to constrain the magnetization direction in $n^{\sigma\sigma'}_{mm'}$?

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$$+n^{\sigma\sigma}_{m_1m_2}\langle m_1m_3|V_{ee}|m_2m_4\rangle n^{-\sigma\,-\sigma}_{m_3m_4} - n^{\sigma\,-\sigma}_{m_1m_2}\langle m_1m_3|V_{ee}|m_4m_2\rangle n^{-\sigma\sigma}_{m_3m_4}\Big]$$

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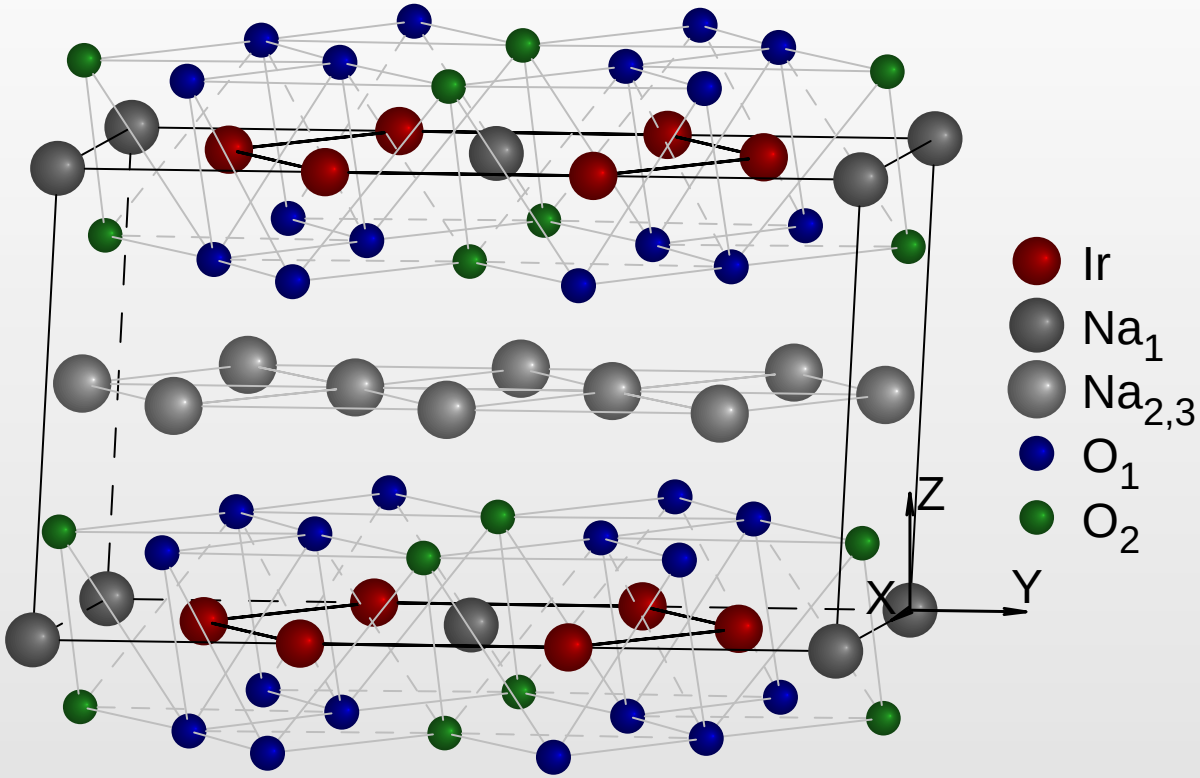
calculations for a limited number of magnetic configurations constrained by symmetry

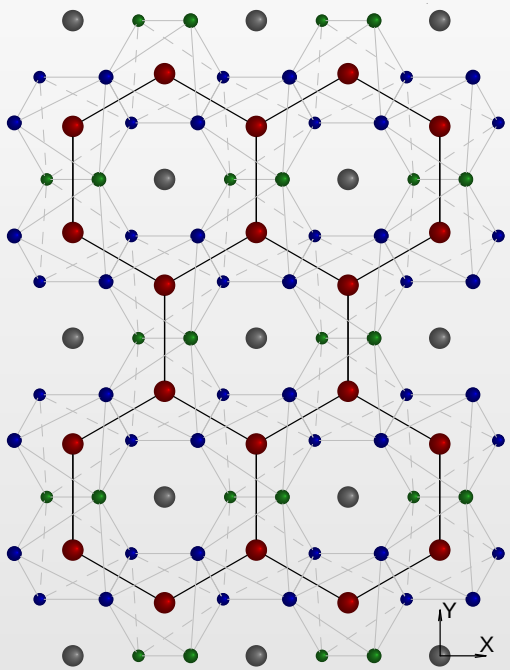
computational details

- relativistic ASA(+CC) LMTO code
- with rotationally invariant L(S)DA+U
- U=1.6, 2.1,2.6 eV; J=0.6 eV $\Rightarrow U_{\text{eff}}=U-J$=1.0, 1.5, 2.0 eV for Ir $5d$ states
- SOC was treated
  - either at the variational step ($|lm\sigma\rangle$ basis)
  - or by solving the 4-component Dirac equation ($|jm_j\rangle$ basis)
- commensurate (non)collinear magnetic structures
- with magnetization directions
  - constant in each atomic sphere
  - recalculated at each iteration until self-consistency

honeycomb Na_2IrO_3

crystal structure

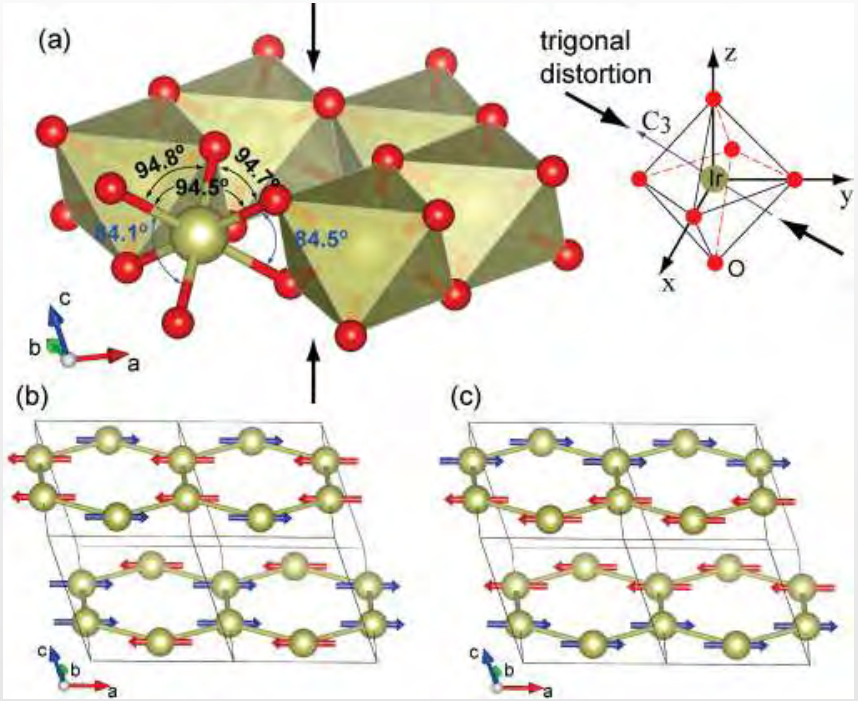




- monoclinic $C/2m$ space group
- honeycomb Ir layers separated by triangular Na layers
- trigonally distorted IrO_6 octahedra; Ir^{4+} d^5

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)

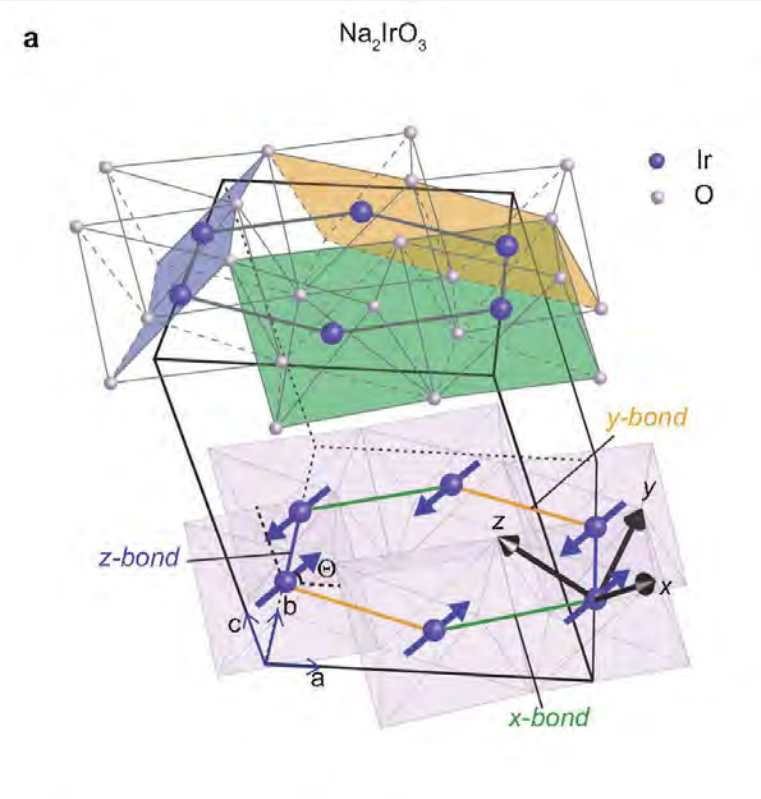
explanations:

- pure Heisenberg model with long-ranged interactions
- Kitaev-Heisenberg model
- Kitaev-Heisenberg model + additional anisotropic exchanges Γ, Γ'
- Ir t_{2g} form quasi-molecular orbitals on a hexagon which compete with SOC

diffuse magnetic x-ray scattering results

Direct Evidence for Dominant Bond-directional Interactions in a Honeycomb Lattice Iridate Na_2IrO_3

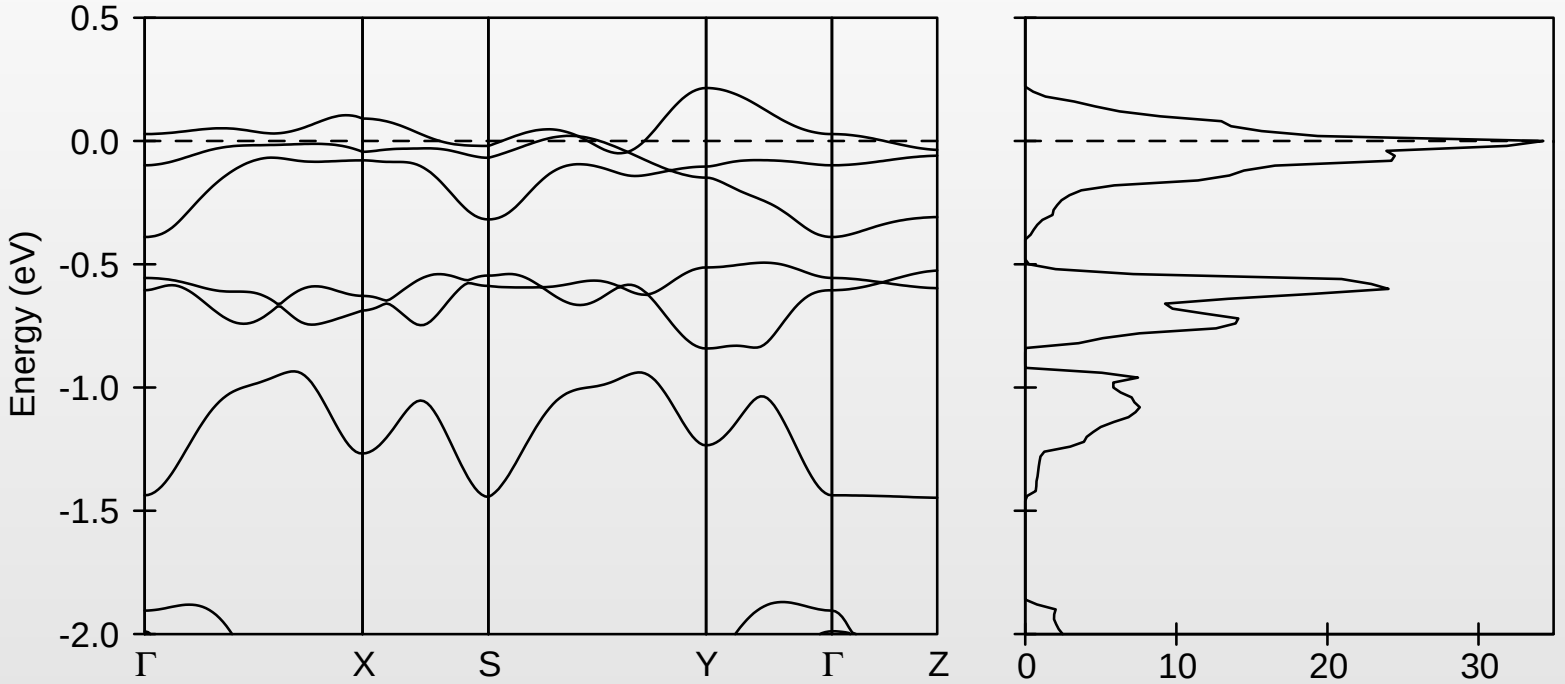
Sae Hwan Chun[1], Jong-Woo Kim[2], Jungho Kim[2], H. Zheng[1], Constantinos C. Stoumpos[1], C. D. Malliakas[1], J. F. Mitchell[1], Kavita Mehlawat[3], Yogesh Singh[3], Y. Choi[2], T. Gog[2], A. Al-Zein[4], M. Moretti Sala[4], M. Krisch[4], J. Chaloupka[5], G. Jackeli[6,7], G. Khaliullin[6] and B. J.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



- 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

S.H. Chun, *et al* arXiv:1504.03618

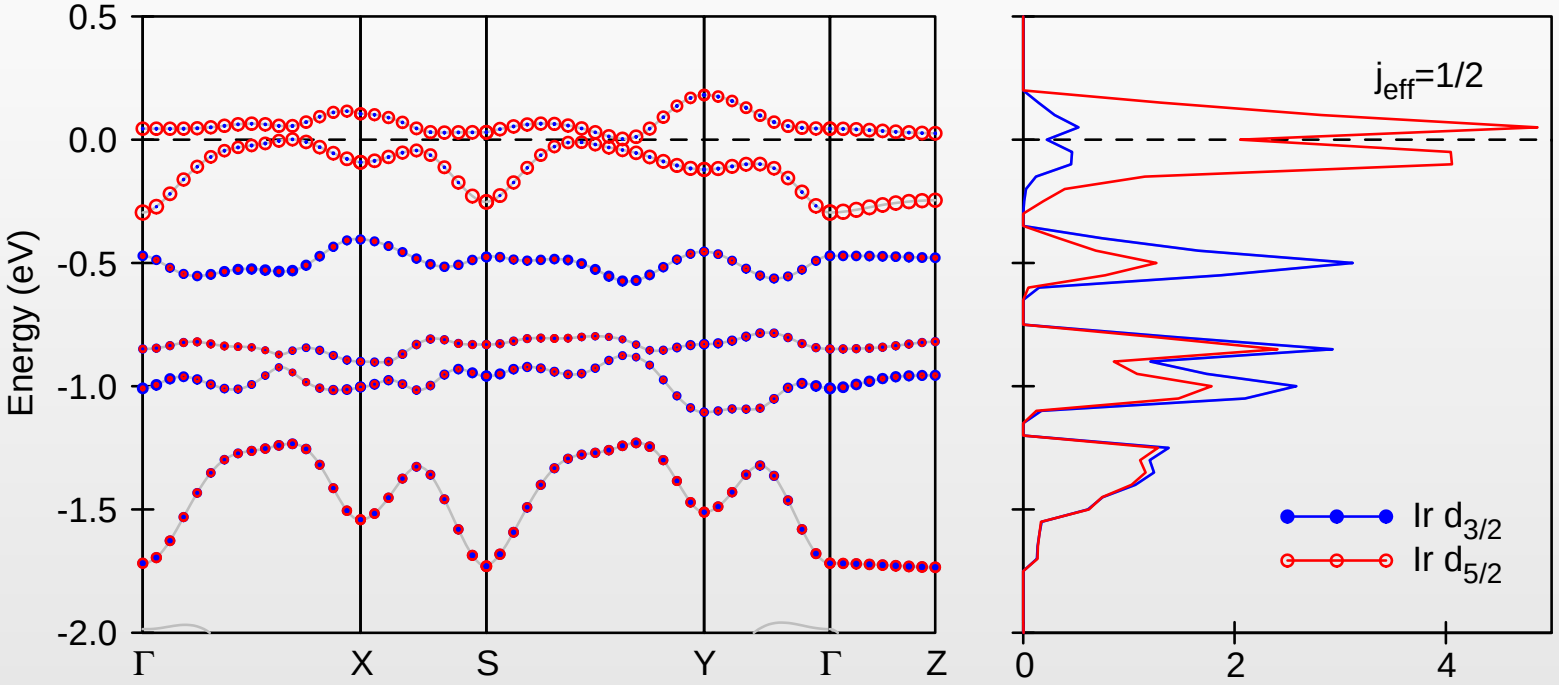
scalar-relativistic bands for Na_2IrO_3



Ir t_{2g} states form quasi-molecular orbitals

I. Mazin, *et al* PRL **109**, 197201 (2012), K. Foyevtsova, *et al* PRB **88**, 035107 (2013)

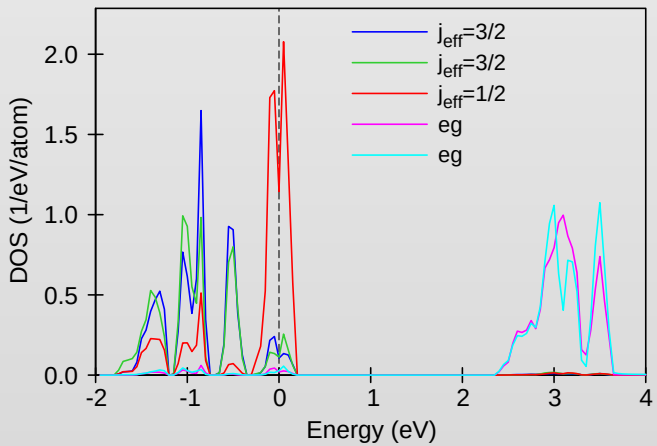
relativistic bands for Na_2IrO_3



dominant contribution of Ir $d_{5/2}$ states to bands crossing E_F

$\Downarrow$

$\sim j_{\text{eff}} = 1/2$ states



calculations for 8 magnetic structures

- 4 inequivalent magnetic structures for the $C2/m$ cell
- 4 inequivalent magnetic structures for a doubled $P2/m$ cell
- all $\mathrm{Na}_{1,2,3}$, Ir, and $\mathrm{O}_{1,2}$ sites remain equivalent
- "$-$" means that rotation is followed by time reversal $\hat{\Theta} = -i\sigma_y \hat{K}$

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

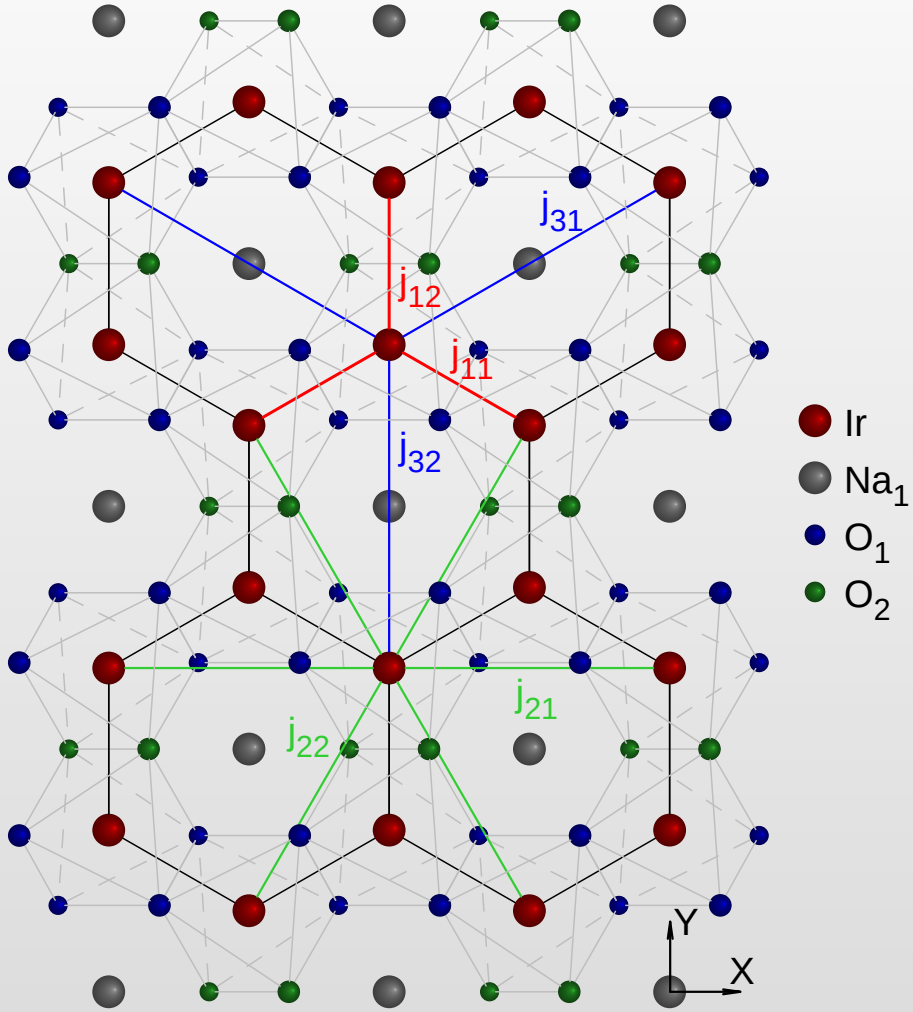
$$C_{2y}m_x = -m_x,\ C_{2y}m_y = m_y,\ C_{2y}m_z = -m_z \quad \Rightarrow \quad m_x = m_z = 0,\ \mathbf{m}_{\mathrm{Ir}} || b$$

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

	symmetry operations				$\mathbf{m}$	
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	

- $\mathbf{m}_{\mathrm{Ir}} || ac$: Ir magnetization direction is defined by polar angle θ
- all Ir moments are collinear, self-consistency in Ir, O, Na magnetization directions

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 exchange

For general bi-linear pair interaction between spins: $H = \frac{1}{2}\sum \mathbf{S}_i^T \mathfrak{J}_{ij} \mathbf{S}_j$

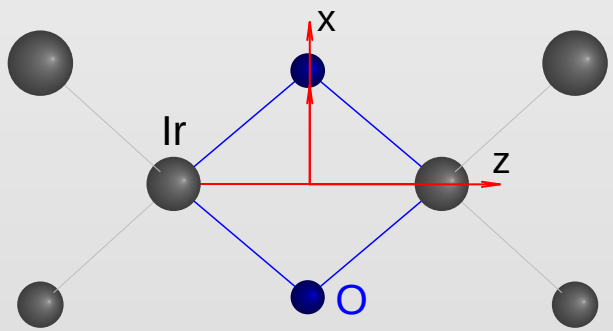
where $\mathfrak{J}_{\alpha\beta}$ is a real 3×3 matrix: $\mathfrak{J} = \mathfrak{J}^+ + \mathfrak{J}^-$

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

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



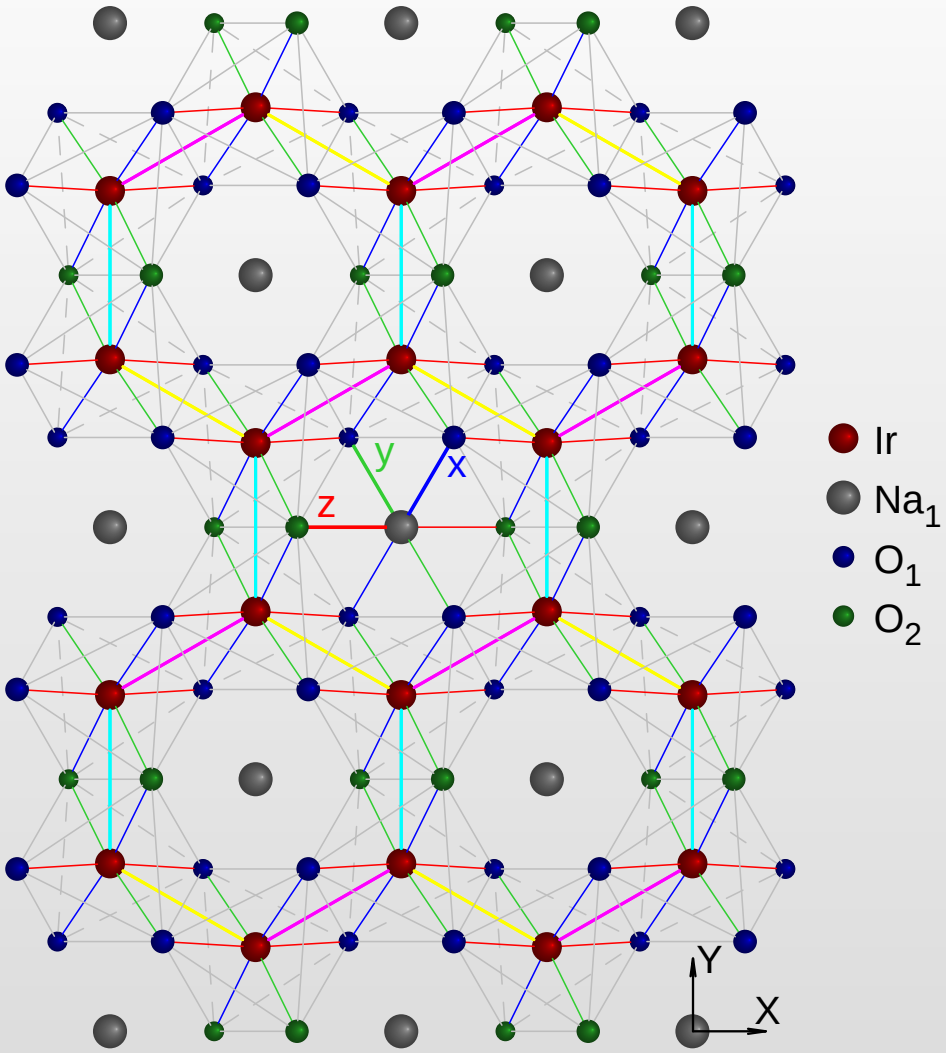
$$\mathfrak{J}^+ = \begin{pmatrix} \mathfrak{J}^+_{xx} & \mathfrak{J}^+_{xy} & 0 \\ \mathfrak{J}^+_{xy} & \mathfrak{J}^+_{yy} & 0 \\ 0 & 0 & \mathfrak{J}^+_{zz} \end{pmatrix}, \quad \mathfrak{J}^+_{xx} \neq \mathfrak{J}^+_{yy} \neq \mathfrak{J}^+_{zz}$$

or isotropic J_0 + traceless symmetric part

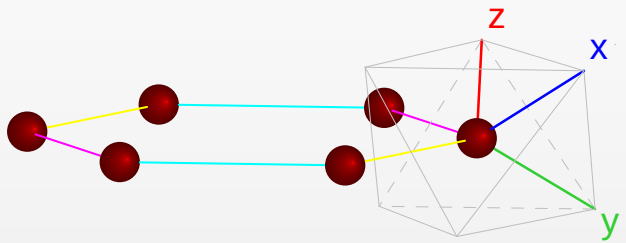
$$\mathfrak{J}^+ = J_0 \cdot \hat{I} + \begin{pmatrix} B & C & 0 \\ C & -B - A & 0 \\ 0 & 0 & A \end{pmatrix}$$

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:

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

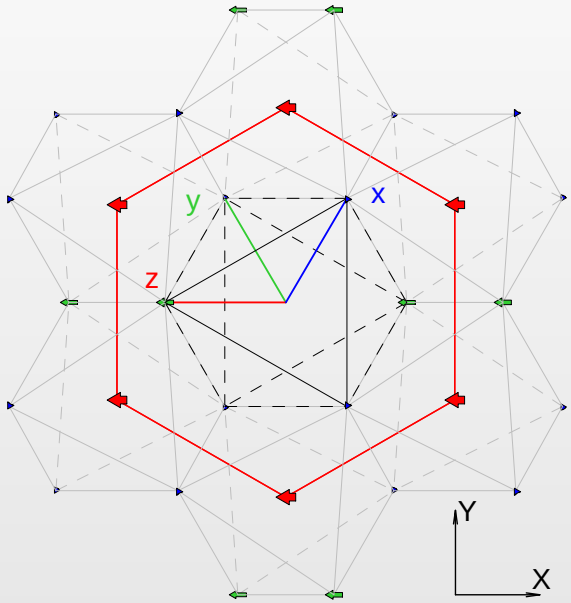
Γ-terms:

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

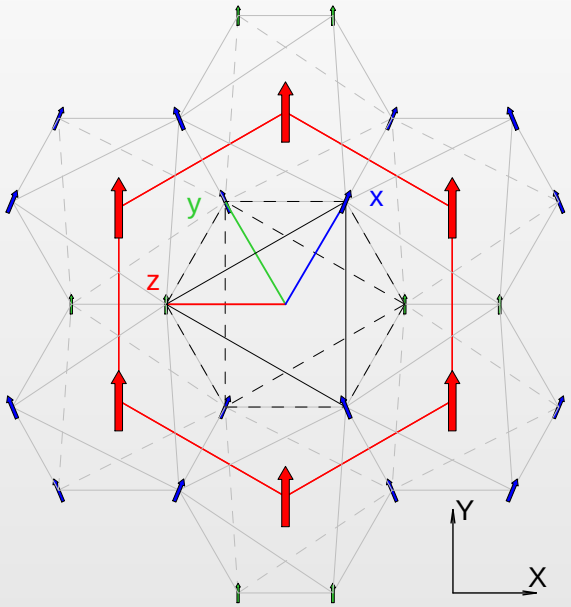
J.G. Rau, *et al* PRL **112**, 077204 (2014), J. Chaloupka and G. Khaliullin, arXiv:1502.02587

FM order (c_f)

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



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



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

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$\varepsilon = -5.5$ meV, $\theta = 18.8°$, $\phi = 180°$

$\varepsilon = 0.$

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 $K_a \neq K_b$?

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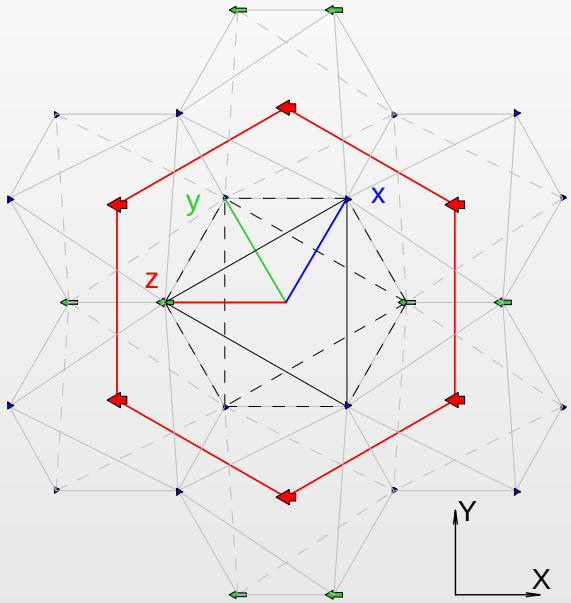
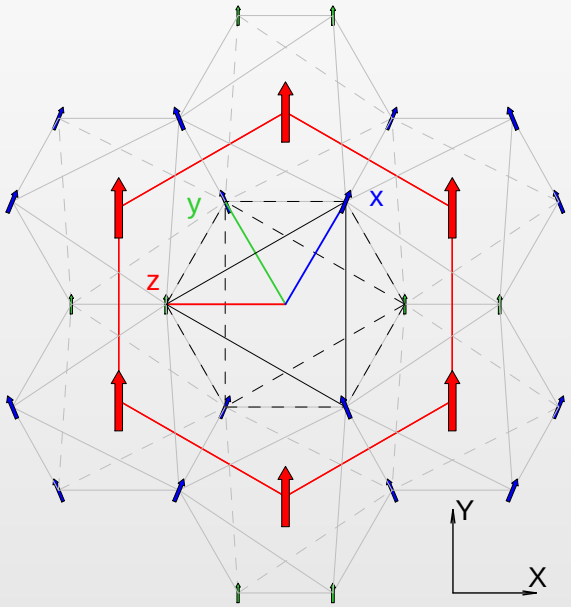


$\mathbf{M}||Y$ $(\mathbf{M}||b)$



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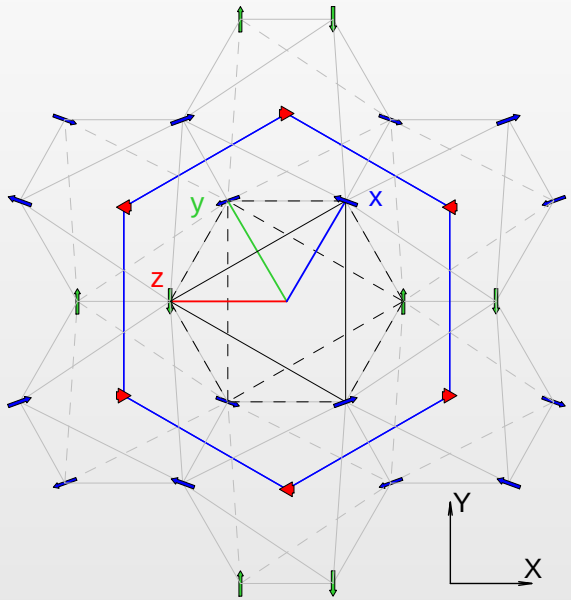
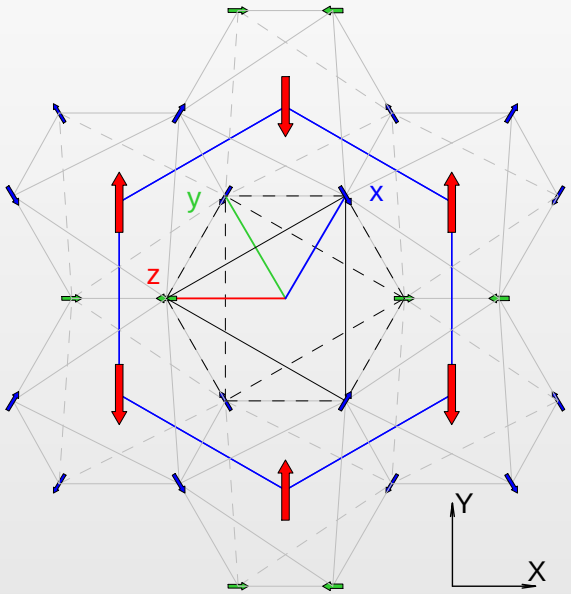
noncollinear O moments (0.04–0.09 μ_B) even for FM order $\Rightarrow$ anisotropic interactions?

Néel AFM order (c_a)

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



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



$$\varepsilon = -6J_1 + 12J_2 - 6J_3 - 2K$$

$$\varepsilon = -10.9, \quad \theta = 14.2°, \quad \phi = 180°$$

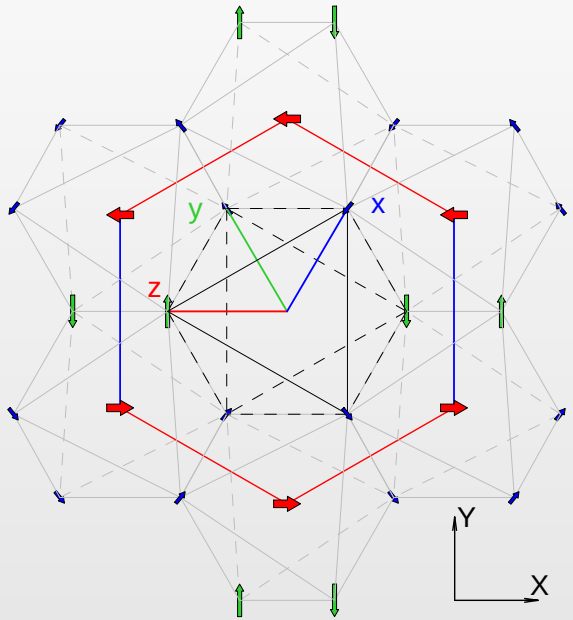
$$\varepsilon = -6J_1 + 12J_2 - 6J_3 - 2K$$

$$\varepsilon = -12.4 \text{ meV}$$

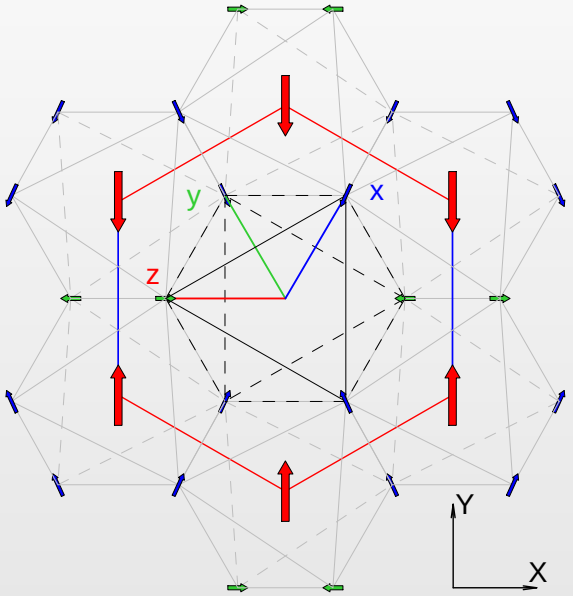
$\theta \neq 0$, $\Delta\varepsilon \neq 0$ but smaller than for c_f

zig-zag (p_z)

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



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



$$\varepsilon = 2J_1 - 4J_2 - 6J_3 + 2K(\cos 2\theta + 2\sqrt{2}\sin 2\theta)/3$$

$\varepsilon = -16.7$ meV, $\theta = 26.4°$, $\phi = 0$

$$\varepsilon = 2J_1 - 4J_2 - 6J_3 + 2K$$

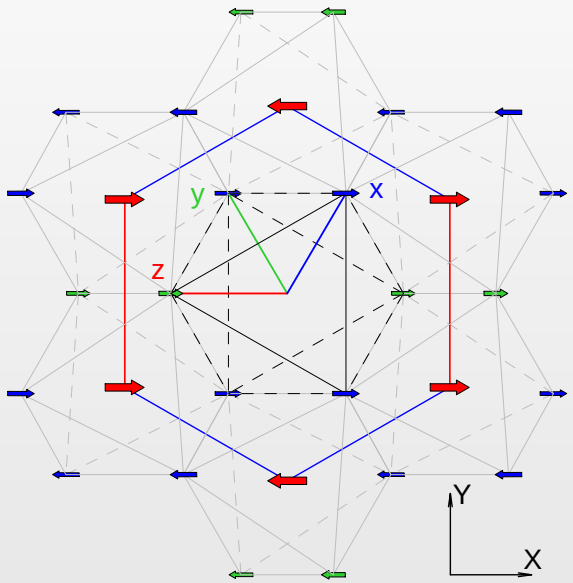
$\varepsilon = -14.9$ meV

extrema at $\tan 2\theta = 2\sqrt{2}$, $\theta_m = 35.3° + n90°$; $\varepsilon_k(\theta_m) = \pm 2K$

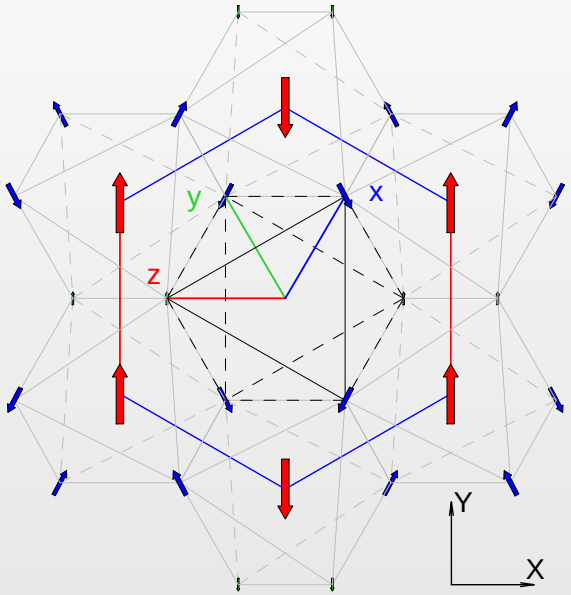
the lowest energy for zig-zag order; $\theta = 26.4° < \theta_{exp} = 45.7°$ $(90° - 44.3°)$

stripe (p_s)

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



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



$$\varepsilon = -2J_1 - \ 4J_2 + 6J_3 \\ -2K(\cos 2\theta + 2\sqrt{2}\sin 2\theta)/3$$

$\varepsilon = -8.7$ meV, $\quad \theta = 138.5°, \quad \phi = 0°$

$$\varepsilon = -2J_1 - \ 4J_2 + 6J_3 \\ -2K$$

$\varepsilon = 0.5$ meV

large $|\Delta\varepsilon| = 9.2$ meV can be explained by K exchange

summary for the 8 configurations

	$\mathbf{M}$	ε (meV)	θ	ϕ	m_s (μ_B)	m_l (μ_B)	$n_{j=1/2}$	
c_f	ac	-5.5	18.8	180	0.32	0.40	0.89	0.34
c_f	b	0.	90.0	90	0.20	0.43	0.89	0.34
c_a	ac	-10.9	14.2	180	0.24	0.35	0.89	0.35
c_a	b	-12.4	90.0	90	0.14	0.38	0.89	0.35
p_z	ac	-16.7	26.4	0	0.25	0.34	0.89	0.35
p_z	b	-14.9	90.0	90	0.18	0.39	0.89	0.35
p_s	ac	-8.7	138.5	0	0.26	0.36	0.89	0.34
p_s	b	0.5	90.0	90	0.16	0.42	0.89	0.34

- insulating solutions for all magnetic orders
- magnetic moments are larger than $m_{exp} = 0.22\ \mu_B$
- for j_{eff}=1/2: $m_s = 2\langle s_z \rangle = 1/3$, $m_l = \langle l_z \rangle = 2/3$
- spin moment is suppressed for $\mathbf{M}||b$
- occupations of j_{eff}=1/2 states do not depend on magnetic order

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 so far, but $\varepsilon(p_z^b) > \varepsilon(p_z^{ac})$

U dependence

		$U_{\text{eff}} = 1.0$ eV		$U_{\text{eff}} = 1.5$ eV		$U_{\text{eff}} = 2.0$ eV	
		calc	$123K\Gamma$	calc	$123K\Gamma$	calc	$123K\Gamma$
c_f	ac	-6.1	-2.1	-5.5	-1.9	-4.9	-1.6
c_f	b	0.0	0.0	0.0	0.0	0.0	0.0
c_a	ac	-14.1	-13.8	-10.9	-10.7	-8.8	-8.5
c_a	b	-15.9	-16.1	-12.4	-12.6	-10.0	-10.2
p_z	ac	-20.3	-17.8	-16.7	-14.3	-14.0	-11.8
p_z	b	-18.6	-21.1	-14.9	-17.3	-12.2	-14.3
p_s	ac	-9.6	-9.5	-8.7	-8.6	-7.8	-7.7
p_s	b	0.6	0.6	0.5	0.4	0.4	0.4
χ			0.09		0.10		0.11
J_1			3.1		3.2		2.5
J_2			.55		.53		.47
J_3			4.2		3.2		2.5
K			-8.9		-8.0		-7.3
Γ			3.2		2.8		2.3

- energy differences and exchange constants decrease with the increase of U_{eff}
- θ does not depend on U_{eff}, e.g., for p_z: $\theta_{1.0} = 27.8°$, $\theta_{1.5} = 26.4°$, $\theta_{2.0} = 26.4°$

dependence on SOC strength ξ for U_{eff}=1.5 eV

		$\xi = 0.75\xi_0$		$\xi = \xi_0$		$\xi = 1.5\xi_0$		$\xi = 2\xi_0$	
		calc	$123K\Gamma$	calc	$123K\Gamma$	calc	$123K\Gamma$	calc	$123K\Gamma$
c_f	ac	-8.4	-2.9	-5.5	-1.9	-2.9	-1.0	-1.9	-0.8
c_f	b	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
c_a	ac	-12.5	-12.2	-10.9	-10.7	-8.7	-8.6	-7.3	-7.2
c_a	b	-14.9	-15.2	-12.4	-12.6	-9.5	-9.7	-7.9	-8.0
p_z	ac	-20.0	-16.4	-16.7	-14.3	-12.5	-11.3	-9.9	-9.2
p_z	b	-17.3	-20.9	-14.9	-17.3	-11.8	-12.9	-9.8	-10.4
p_s	ac	-10.5	-10.4	-8.7	-8.6	-6.5	-6.5	-5.2	-5.2
p_s	b	0.3	0.1	0.5	0.4	0.5	0.5	0.2	0.2
θ_{p_z}	χ	24.4°	0.13	26.4°	0.10	32.0°	0.07	34.7°	0.05
J_1			2.9		3.2		2.2		1.8
J_2			.70		.53		.35		.27
J_3			3.6		3.2		2.6		2.1
K			-8.8		-8.0		-6.3		-4.9
Γ			4.3		2.8		1.6		1.2

- somewhat stronger dependence of θ on ξ
- χ decreases with increasing ξ, i.e. fit becomes better: closer to j_{eff}=1/2 limit?

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
- Adding the anisotropic K and Γ terms improves the fit

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- . . . but I would be cautions