Topological spin liquids in honeycomb iridates and **$RuCl_3$** ?

Jeroen van den Brink

Leibniz Institute for Solid State and Materials Research Dresden

TECHNISCHE UNIVERSITÄT DRESDEN

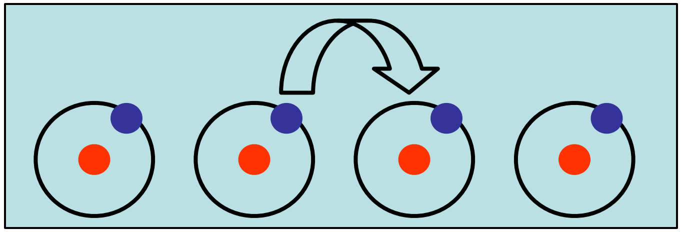
Ravi Yadav
Mohamed Eldeeb
Raajyavardhan Ray
Satoshi Nishimoto
Liviu Hozoi

SFB 1143

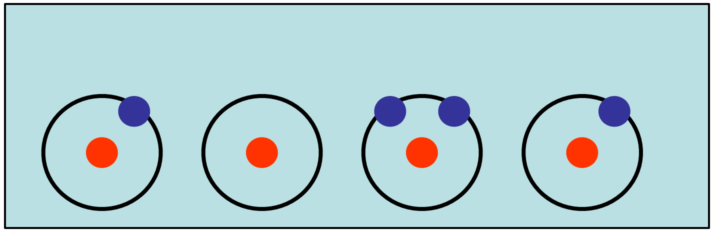
CENTER FOR TOPOLOGICAL QUANTUM MATTER RESEARCH

Topoquant19
KITP
Santa Barbara
28.08.2019

Chain of hydrogen atoms



Hopping amplitude: ***t***

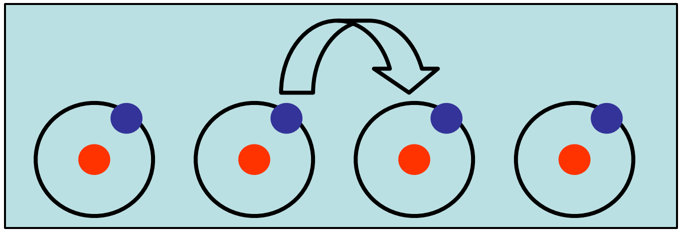


Coulomb interaction: ***U***

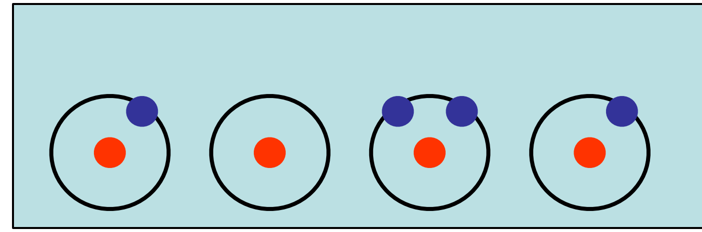
U = 0	***Bands: Metallic behaviour***
U >> t	***Mott-Hubbard Insulator*** → Antiferromagnetism

Hubbard Hamiltonian

$$H_{Hub} = t \sum_{\langle ij\rangle,\sigma} (c^{\dagger}_{i\sigma} c_{j\sigma} + h.c) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$



Hopping amplitude: ***t***

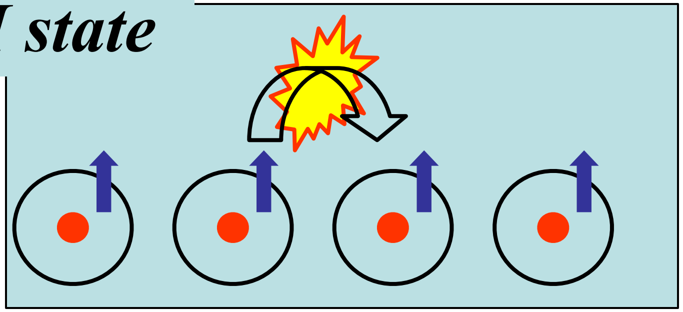


Coulomb interaction: ***U***

U = 0 — ***Bands: Metallic behaviour***

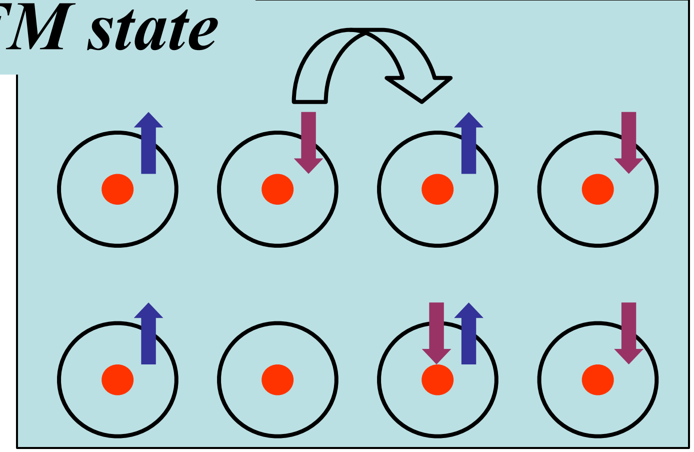
U >> t — ***Mott-Hubbard Insulator*** → Antiferromagnetism

FM state



E = 0

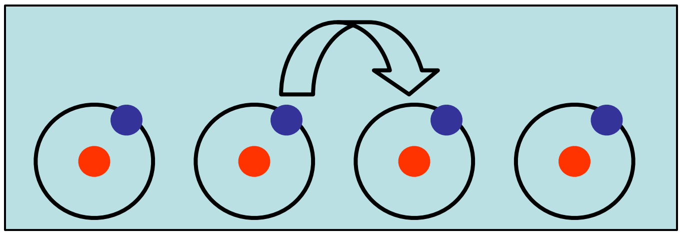
AFM state



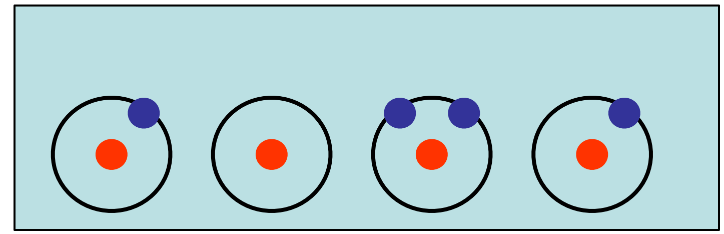
E = -t²/U

Hubbard Hamiltonian

$$H_{Hub} = t \sum_{\langle ij \rangle,\sigma} (c^{\dagger}_{i\sigma} c_{j\sigma} + h.c) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$



Hopping amplitude: ***t***



Coulomb interaction: ***U***

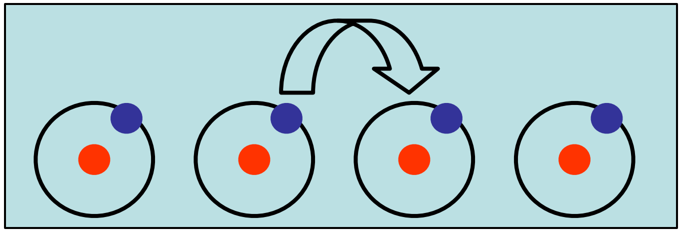
U >> t ***Mott-Hubbard Insulator*** → Antiferromagnetism

Heisenberg Hamiltonian

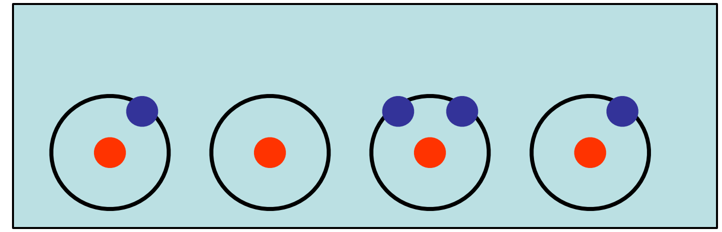
$$H_{Heis} = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j \qquad [S^x, S^y] = iS^z$$

Hubbard Hamiltonian

$$H_{Hub} = t \sum_{\langle ij \rangle, \sigma} (c^{\dagger}_{i\sigma} c_{j\sigma} + h.c) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$



Hopping amplitude: ***t***



Coulomb interaction: ***U***

U >> t ***Mott-Hubbard Insulator*** ⇒ Antiferromagnetism

Heisenberg Hamiltonian

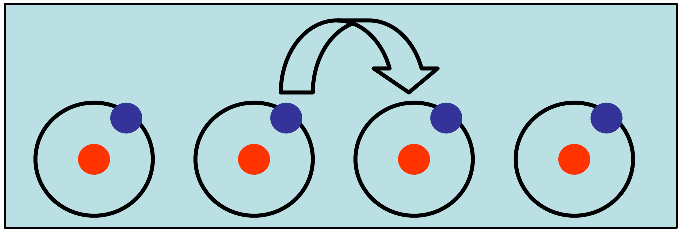
$$H_{Heis} = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j \qquad [S^x, S^y] = iS^z$$

↓

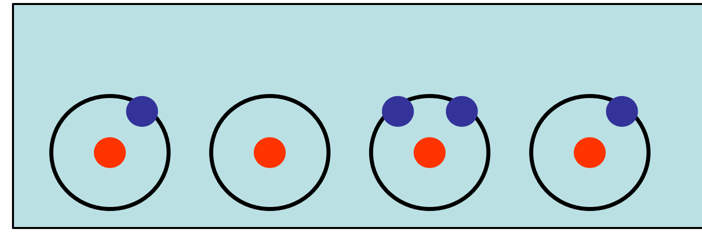
Rotational invariant

Hubbard Hamiltonian

$$H_{Hub} = t \sum_{\langle ij \rangle,\sigma} (c^\dagger_{i\sigma} c_{j\sigma} + h.c) + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



Hopping amplitude: ***t***



Coulomb interaction: ***U***

U >> t — ***Mott-Hubbard Insulator*** → Antiferromagnetism

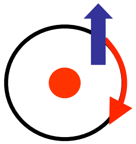
Heisenberg Hamiltonian

$$H_{Heis} = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j \qquad [S^x, S^y] = iS^z$$

Rotational invariant

In real materials: (easy axis) exchange anisotropy

Magnetic anisotropy

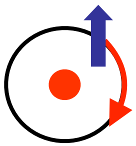


$$\vec{B} = \frac{\vec{v} \times \vec{E}}{c^2}, \quad \vec{E} = -\nabla V$$

$$Zeeman : \vec{B} \cdot \vec{S} \sim \vec{L} \cdot \vec{S}$$

spin-orbit coupling

Magnetic anisotropy



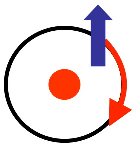
$$\vec{B} = \frac{\vec{v} \times \vec{E}}{c^2}, \quad \vec{E} = -\nabla V$$

$$Zeeman : \vec{B} \cdot \vec{S} \sim \vec{L} \cdot \vec{S}$$

spin-orbit coupling

1. When $c \to \infty$ anisotropy $\to 0$

Magnetic anisotropy



$$\vec{B} = \frac{\vec{v} \times \vec{E}}{c^2}, \quad \vec{E} = -\nabla V$$

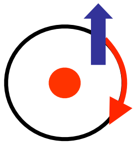
$$Zeeman : \vec{B} \cdot \vec{S} \sim \vec{L} \cdot \vec{S}$$

spin-orbit coupling

1. When $c \to \infty$ anisotropy $\to 0$

2. Total angular momentum $\vec{J} = \vec{L} + \vec{S}$

Magnetic anisotropy



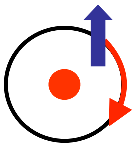
$$\vec{B} = \frac{\vec{v} \times \vec{E}}{c^2}, \quad \vec{E} = -\nabla V$$

$$Zeeman : \vec{B} \cdot \vec{S} \sim \vec{L} \cdot \vec{S}$$

spin-orbit coupling

1. When $c \to \infty$ anisotropy $\to 0$
2. Total angular momentum $\vec{J} = \vec{L} + \vec{S}$
3. ∇V large when Z large $\to$ heavy elements $\to$ 4d, 5d

Magnetic anisotropy



$$\vec{B} = \frac{\vec{v} \times \vec{E}}{c^2}, \quad \vec{E} = -\nabla V$$

$$Zeeman : \vec{B} \cdot \vec{S} \sim \vec{L} \cdot \vec{S}$$

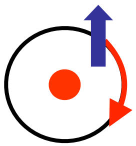
spin-orbit coupling

1. When $c \rightarrow \infty$ anisotropy $\rightarrow 0$
2. Total angular momentum $\vec{J} = \vec{L} + \vec{S}$
3. ∇V large when Z large $\rightarrow$ heavy elements $\rightarrow$ 4d, 5d

Ru, Mo

Ir, Os

Magnetic anisotropy



$$\vec{B} = \frac{\vec{v} \times \vec{E}}{c^2}, \quad \vec{E} = -\nabla V$$

$$Zeeman : \vec{B} \cdot \vec{S} \sim \vec{L} \cdot \vec{S}$$

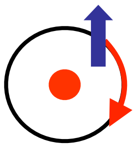
spin-orbit coupling

1. When $c \to \infty$ anisotropy $\to 0$
2. Total angular momentum $\vec{J} = \vec{L} + \vec{S}$
3. ∇V large when Z large $\to$ heavy elements $\to$ 4d, 5d
4. $\vec{J}$ has direction & breaks rotational invariance of H

4d → Ru, Mo

5d → Ir, Os

Magnetic anisotropy



$$\vec{B} = \frac{\vec{v} \times \vec{E}}{c^2}, \quad \vec{E} = -\nabla V$$

$$Zeeman : \vec{B} \cdot \vec{S} \sim \vec{L} \cdot \vec{S}$$

spin-orbit coupling

1. When $c \to \infty$ anisotropy $\to 0$

2. Total angular momentum $\vec{J} = \vec{L} + \vec{S}$

3. ∇V large when Z large $\to$ heavy elements $\to$ 4d, 5d

Ru, Mo

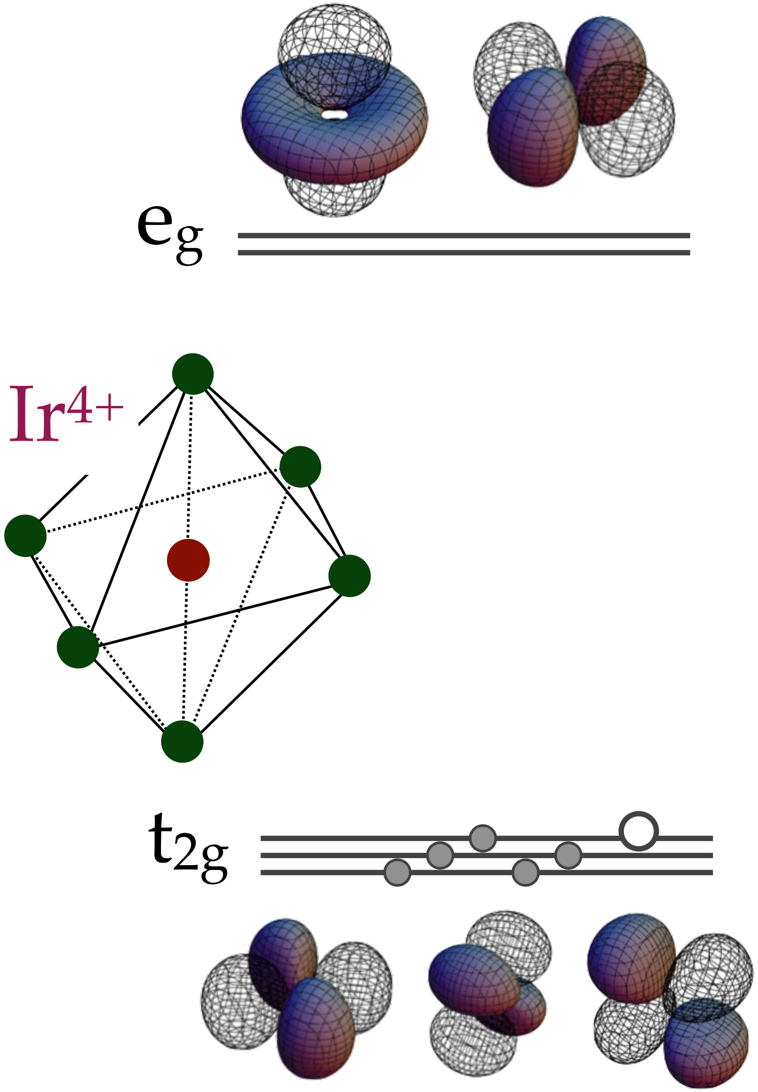
Ir, Os

4. $\vec{J}$ has direction & breaks rotational invariance of H

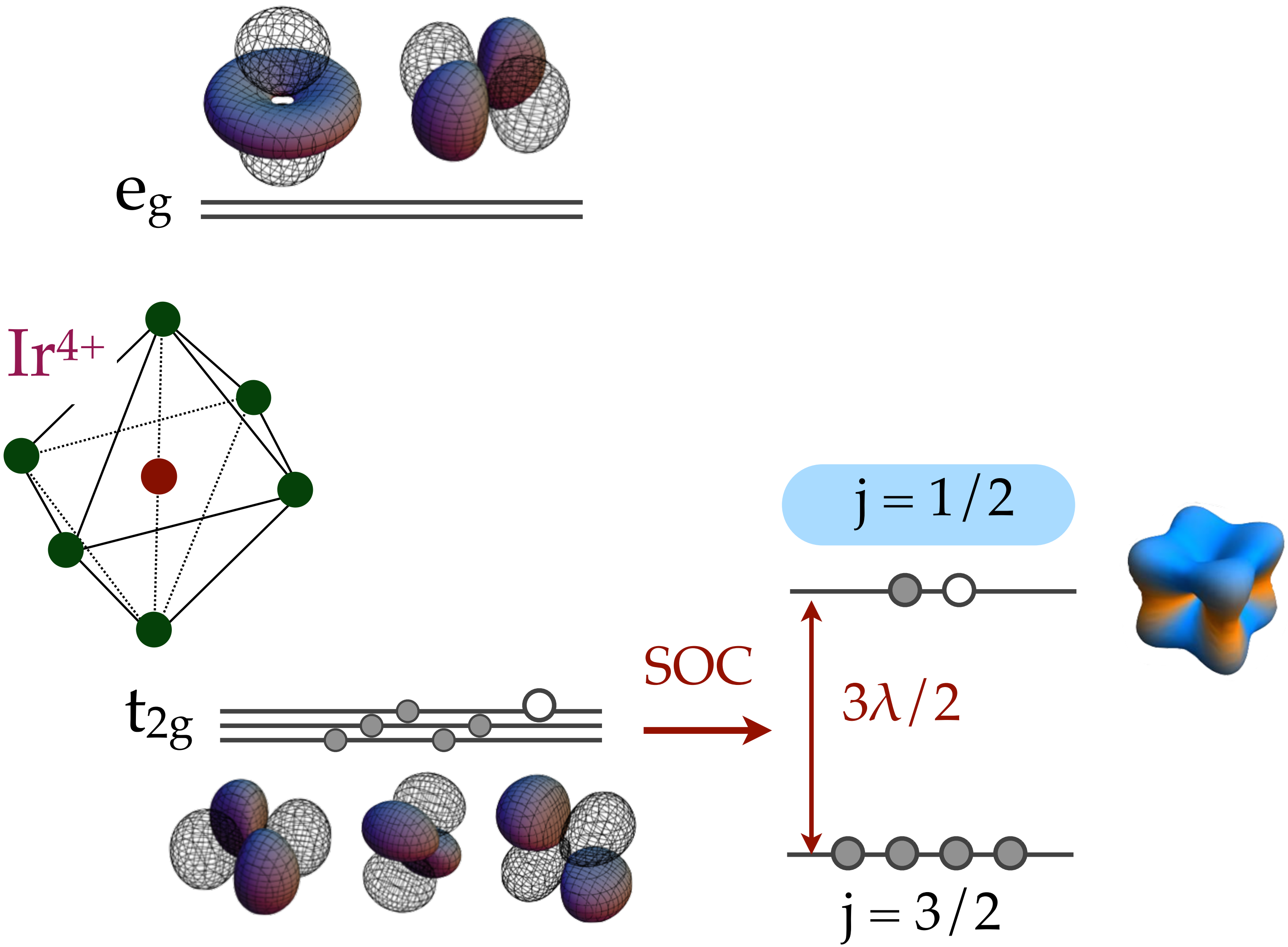
$S_i^z S_j^z$ instead of $\vec{S}_i \cdot \vec{S}_j$

(for $S = 1/2$ we have $(S_i^z)^2 = 1/4$)

Magnetic Iridium Oxides: Ir^{4+}



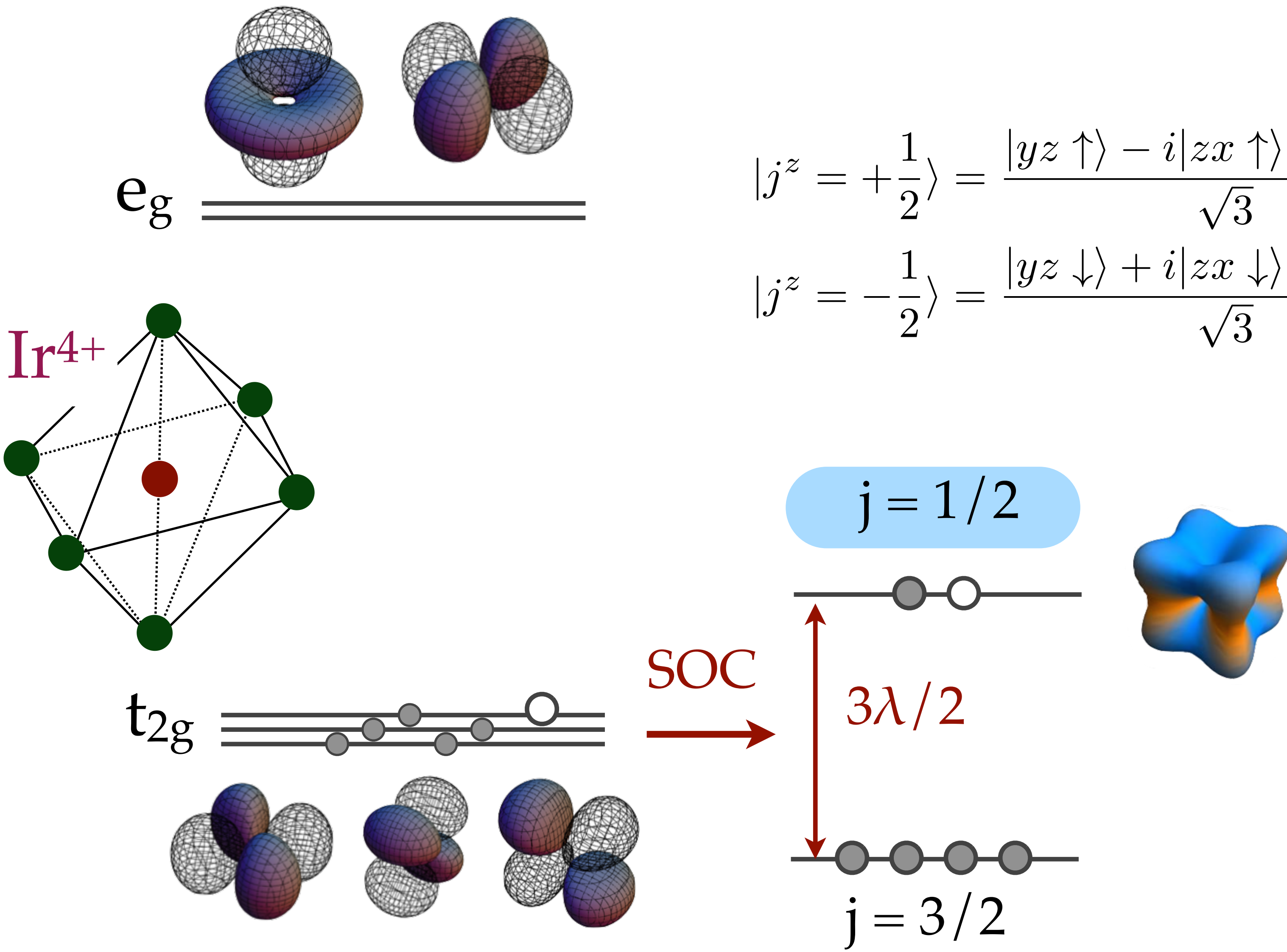
Magnetic Iridium Oxides: Ir^{4+}



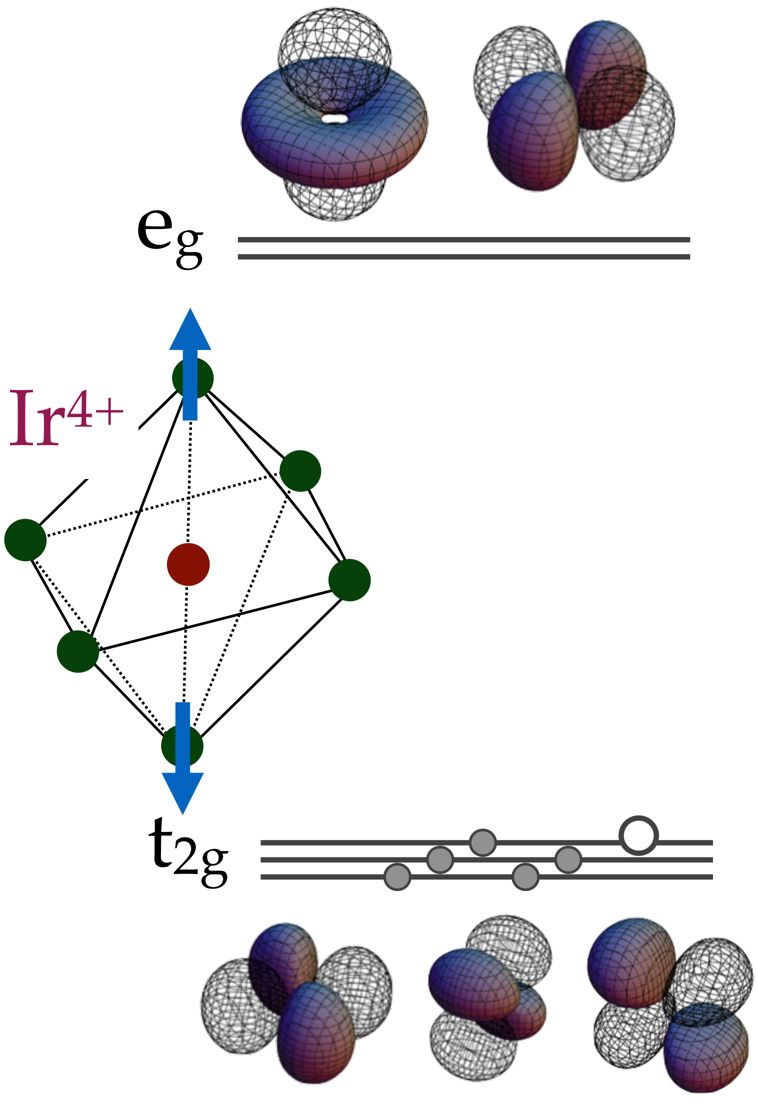
Magnetic Iridium Oxides: Ir^{4+}

$$|j^z = +\frac{1}{2}\rangle = \frac{|yz\uparrow\rangle - i|zx\uparrow\rangle - |xy\downarrow\rangle}{\sqrt{3}}$$

$$|j^z = -\frac{1}{2}\rangle = \frac{|yz\downarrow\rangle + i|zx\downarrow\rangle - |xy\uparrow\rangle}{\sqrt{3}}$$

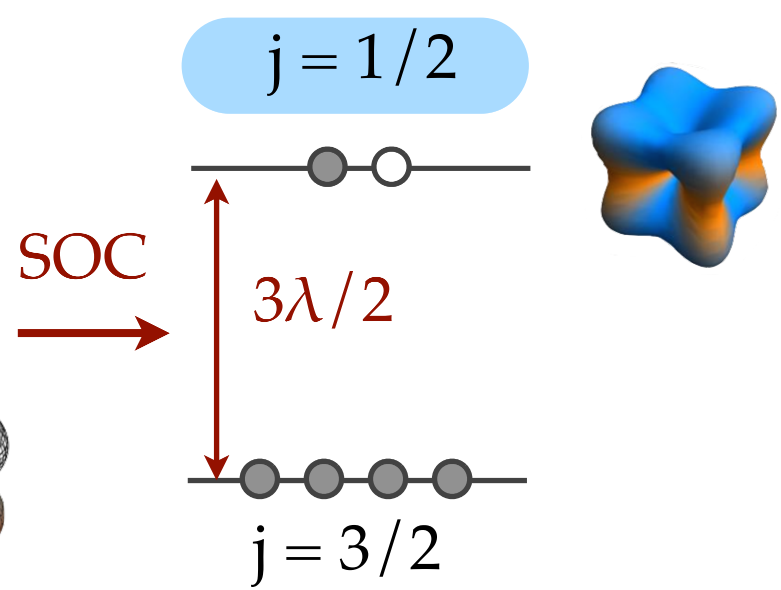


Magnetic Iridium Oxides: Ir^{4+}



$$|j^z = +\frac{1}{2}\rangle = \frac{|yz\uparrow\rangle - i|zx\uparrow\rangle - |xy\downarrow\rangle}{\sqrt{3}}$$

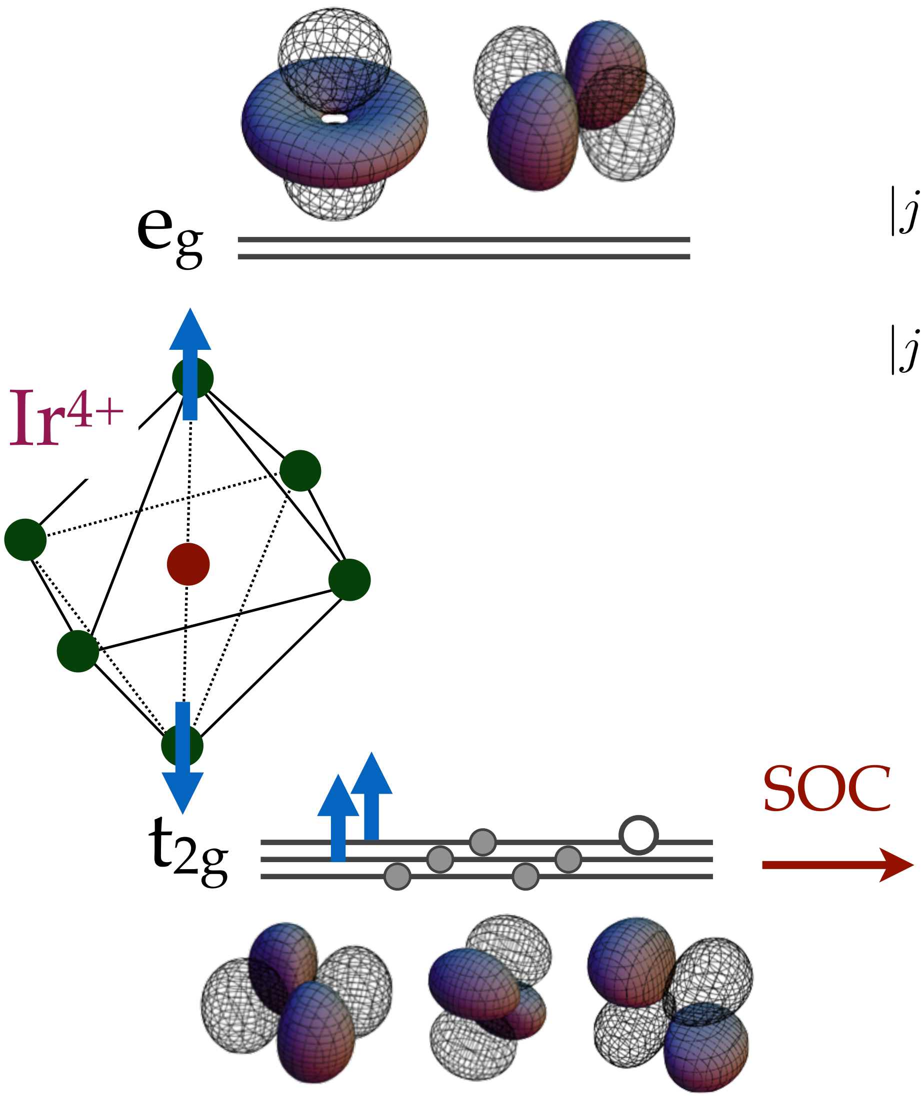
$$|j^z = -\frac{1}{2}\rangle = \frac{|yz\downarrow\rangle + i|zx\downarrow\rangle - |xy\uparrow\rangle}{\sqrt{3}}$$

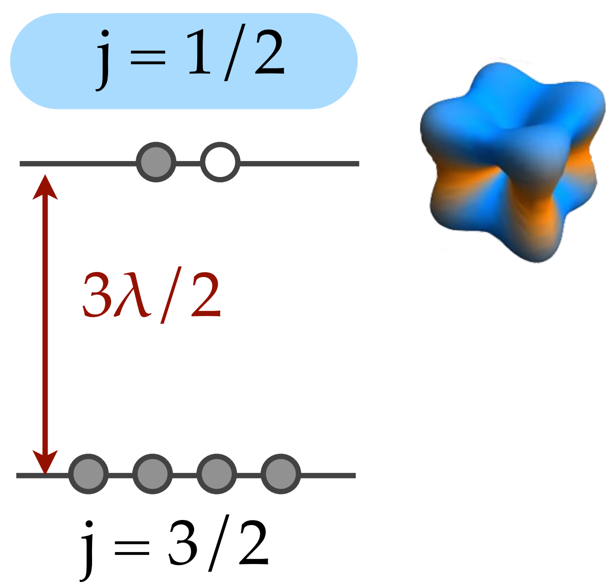


Magnetic Iridium Oxides: Ir^{4+}

$$|j^z = +\frac{1}{2}\rangle = \frac{|yz\uparrow\rangle - i|zx\uparrow\rangle - |xy\downarrow\rangle}{\sqrt{3}}$$

$$|j^z = -\frac{1}{2}\rangle = \frac{|yz\downarrow\rangle + i|zx\downarrow\rangle - |xy\uparrow\rangle}{\sqrt{3}}$$

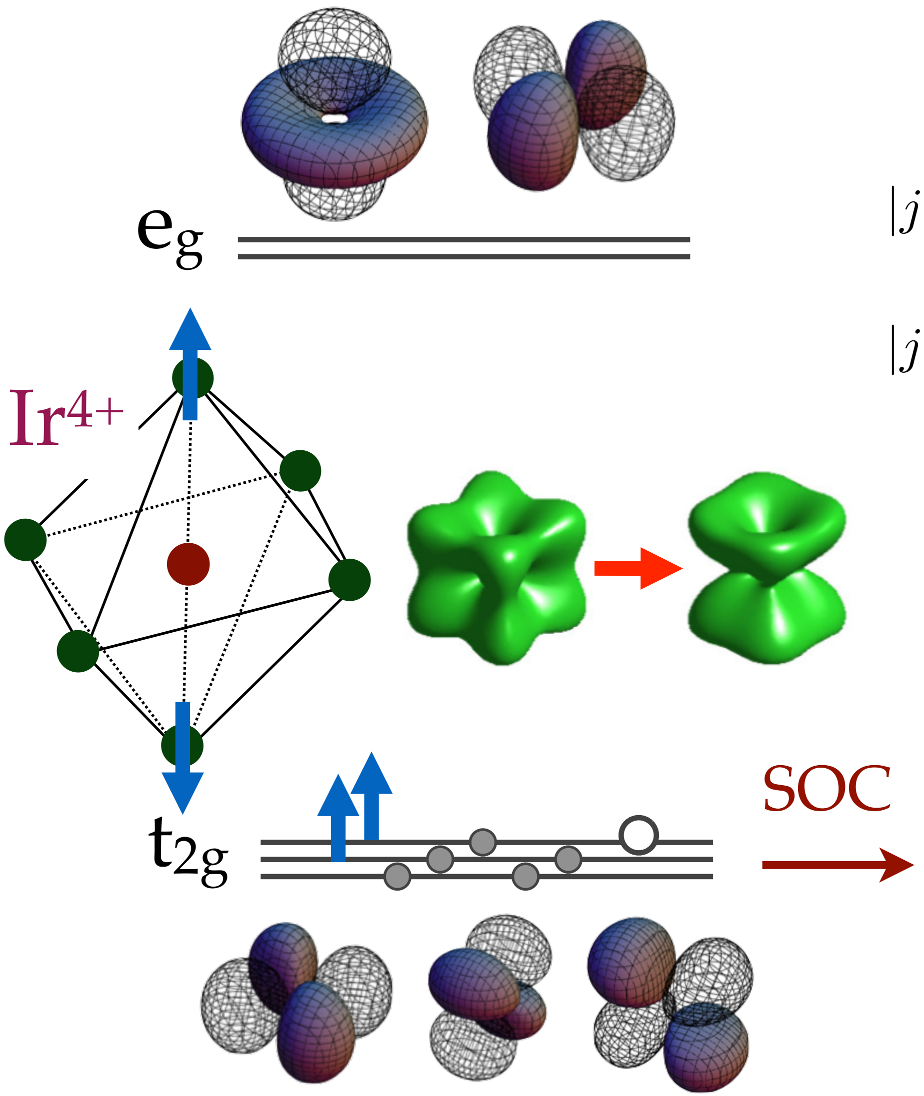


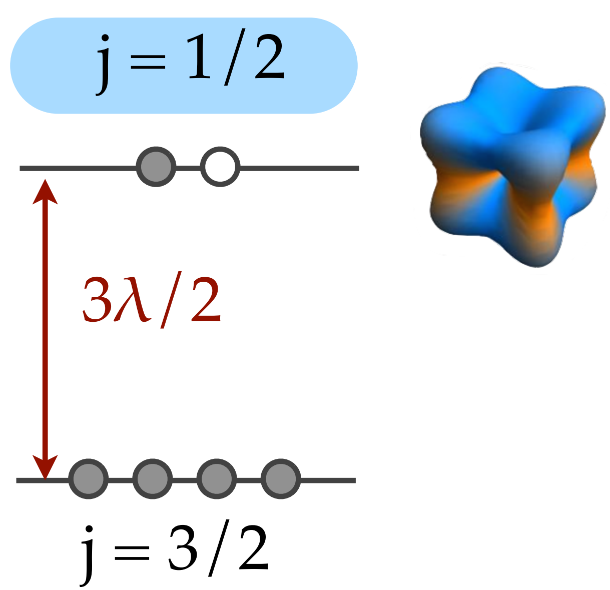


Magnetic Iridium Oxides: Ir^{4+}

$$|j^z = +\frac{1}{2}\rangle = \frac{|yz\uparrow\rangle - i|zx\uparrow\rangle - |xy\downarrow\rangle}{\sqrt{3}}$$

$$|j^z = -\frac{1}{2}\rangle = \frac{|yz\downarrow\rangle + i|zx\downarrow\rangle - |xy\uparrow\rangle}{\sqrt{3}}$$

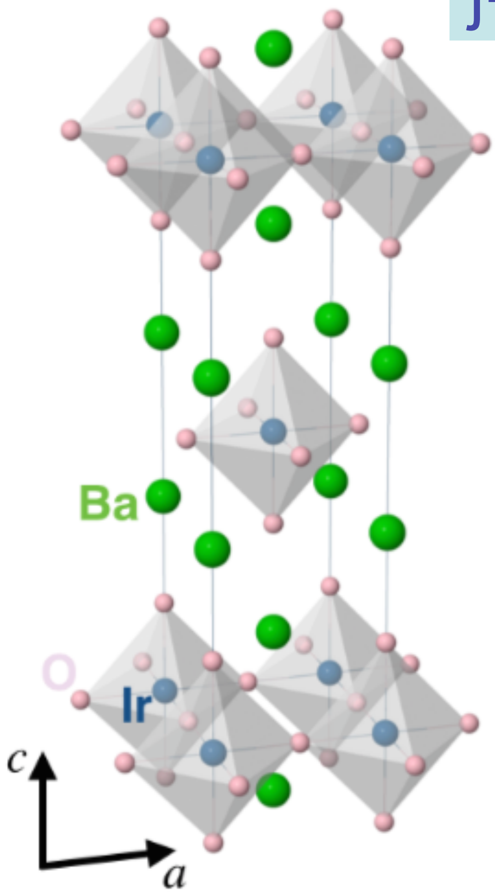


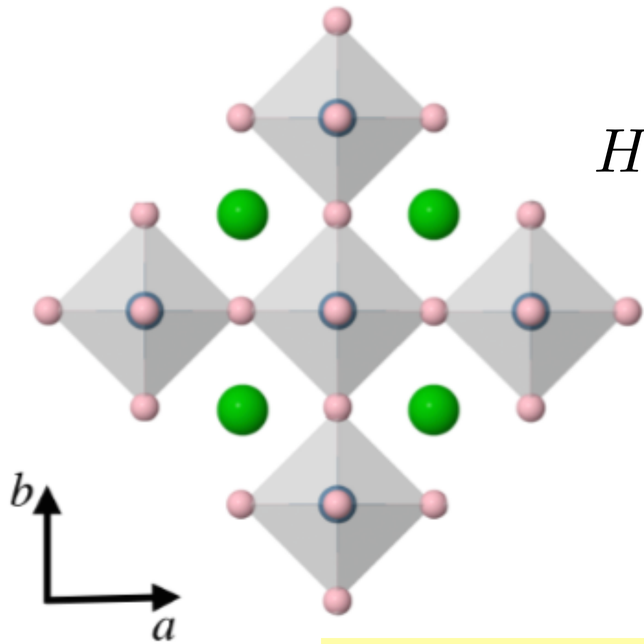


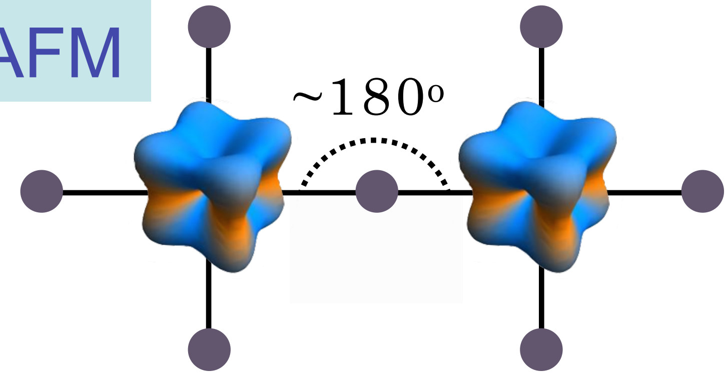
214 Magnetic Iridium Oxides

Sr_2IrO_4: equivalent of cuprate La_2CuO_4

j=1/2 square lattice AFM







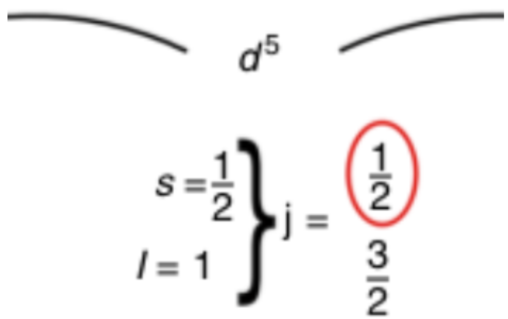
$$H_{Heis} = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j$$

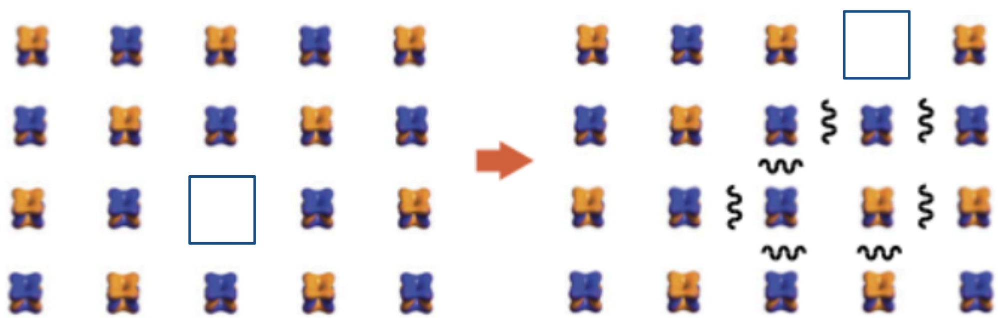
between j=1/2 moments

Jackeli & Khaliullin, PRL 102, 017205 (2009)

Electron/hole propagation in Sr_2IrO_4

hole hopping in s=1/2 AFM creates string of spin flips

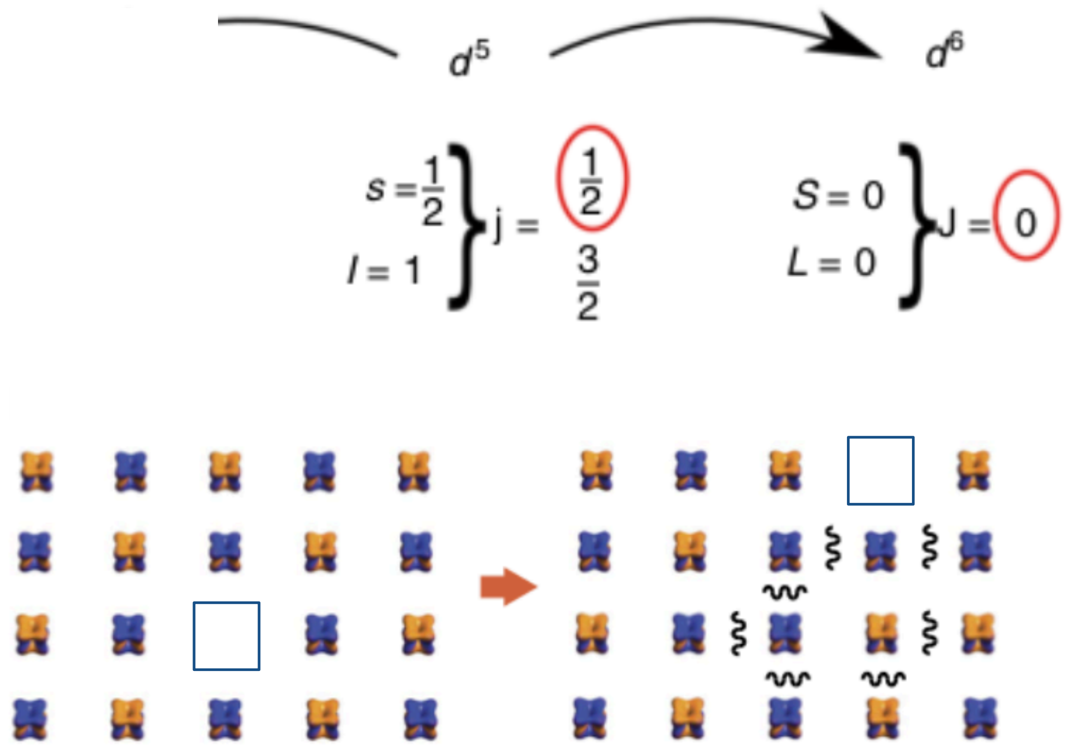




Pärschke, Wohlfeld, Foyevtsova & JvdB,
Nature Comm. 8, 686 (2017)

Electron/hole propagation in Sr_2IrO_4

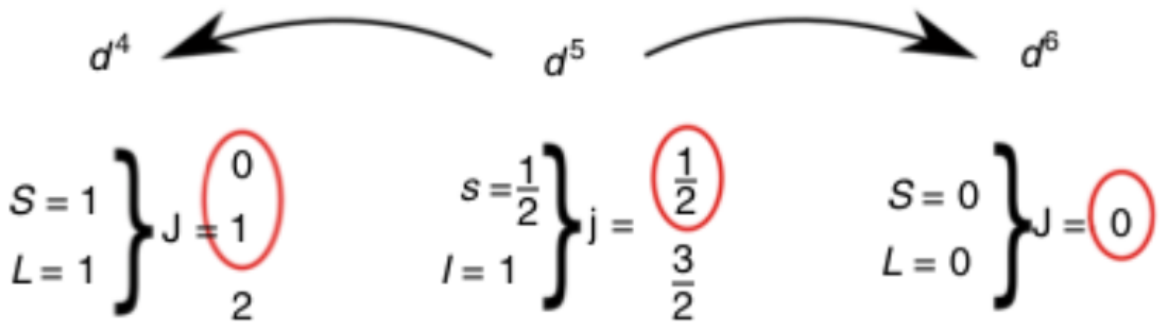
hole hopping in s=1/2 AFM creates string of spin flips

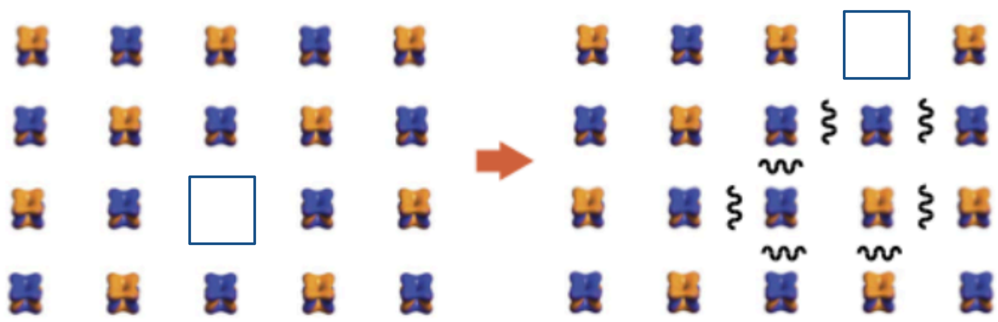


Pärschke, Wohlfeld, Foyevtsova & JvdB,
Nature Comm. 8, 686 (2017)

Electron/hole propagation in Sr_2IrO_4

hole hopping in s=1/2 AFM creates string of spin flips

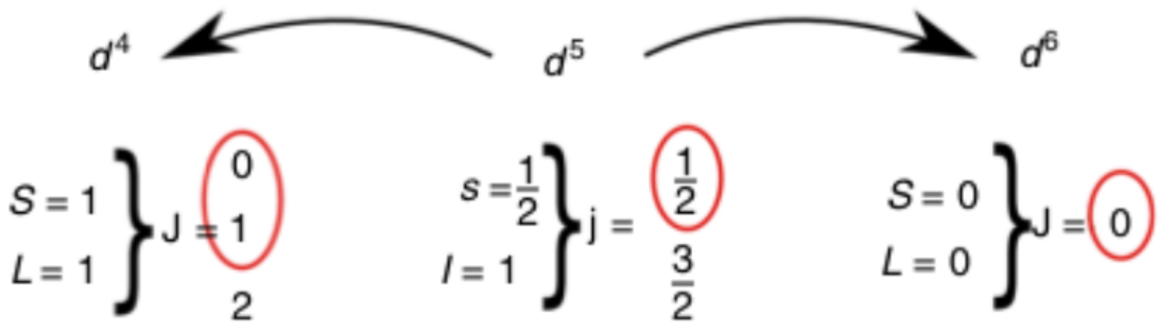




Pärschke, Wohlfeld, Foyevtsova & JvdB, Nature Comm. 8, 686 (2017)

Electron/hole propagation in Sr_2IrO_4

hole hopping in s=1/2 AFM creates string of spin flips

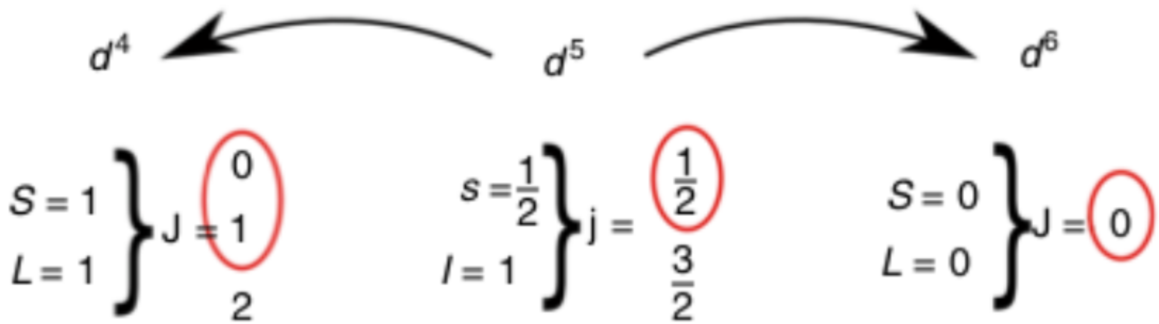


$$\mathcal{H}_t^{d} = \sum_k V_k^0 \left(d_{kA}^\dagger d_{kA} + d_{kB}^\dagger d_{kB}\right) + \sum_{k,q} V_{k,q}\left(d_{k-qB}^\dagger d_{kA}\alpha_q^\dagger + d_{k-qA}^\dagger d_{kB}\beta_q^\dagger + h.c.\right)$$

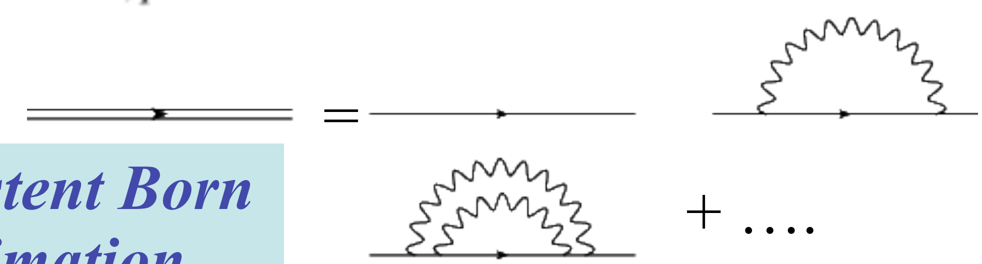
Pärschke, Wohlfeld, Foyevtsova & JvdB,
Nature Comm. 8, 686 (2017)

Electron/hole propagation in Sr_2IrO_4

hole hopping in s=1/2 AFM creates string of spin flips



$$\mathcal{H}_t^d = \sum_k V_k^0 \left(d_{kA}^\dagger d_{kA} + d_{kB}^\dagger d_{kB}\right)$$
$$+ \sum_{k,q} V_{k,q}\left(d_{k-qB}^\dagger d_{kA} \alpha_q^\dagger + d_{k-qA}^\dagger d_{kB} \beta_q^\dagger + h.c.\right)$$

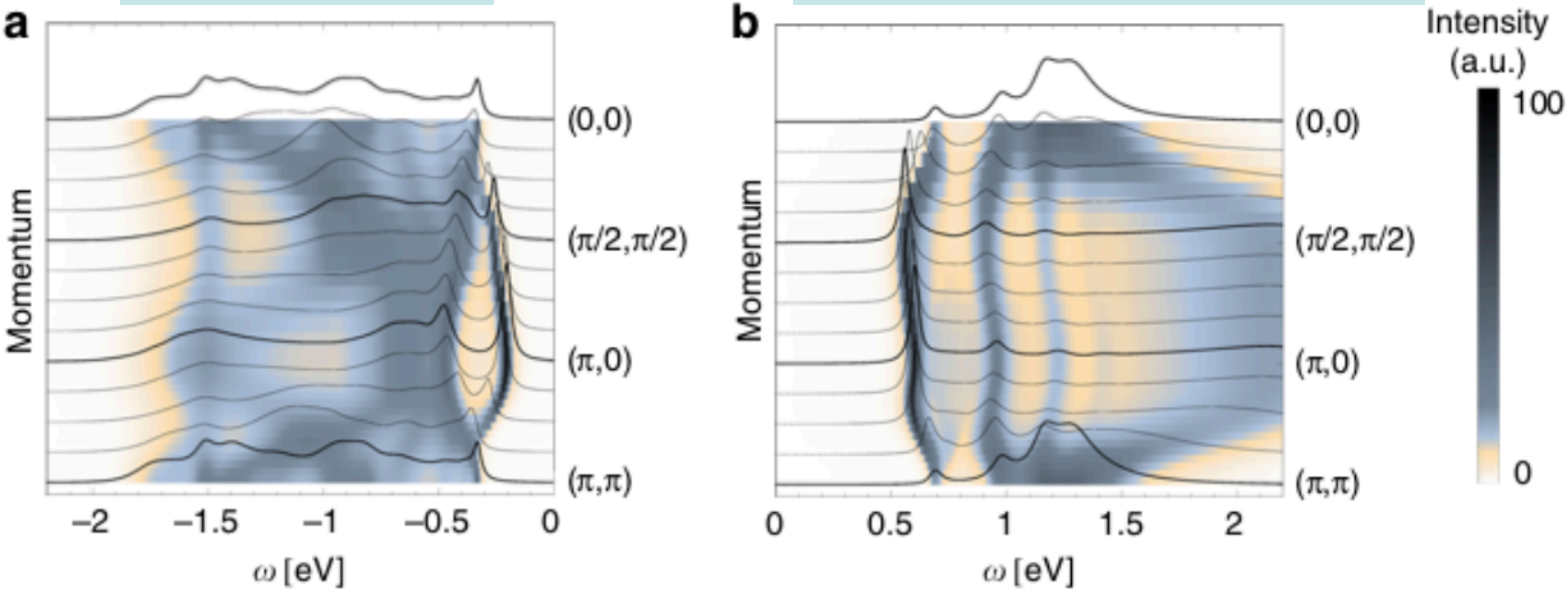


self-consistent Born approximation

Pärschke, Wohlfeld, Foyevtsova & JvdB, Nature Comm. 8, 686 (2017)

Electron/hole propagation in Sr_2IrO_4

photoemission | *inverse photoemission*

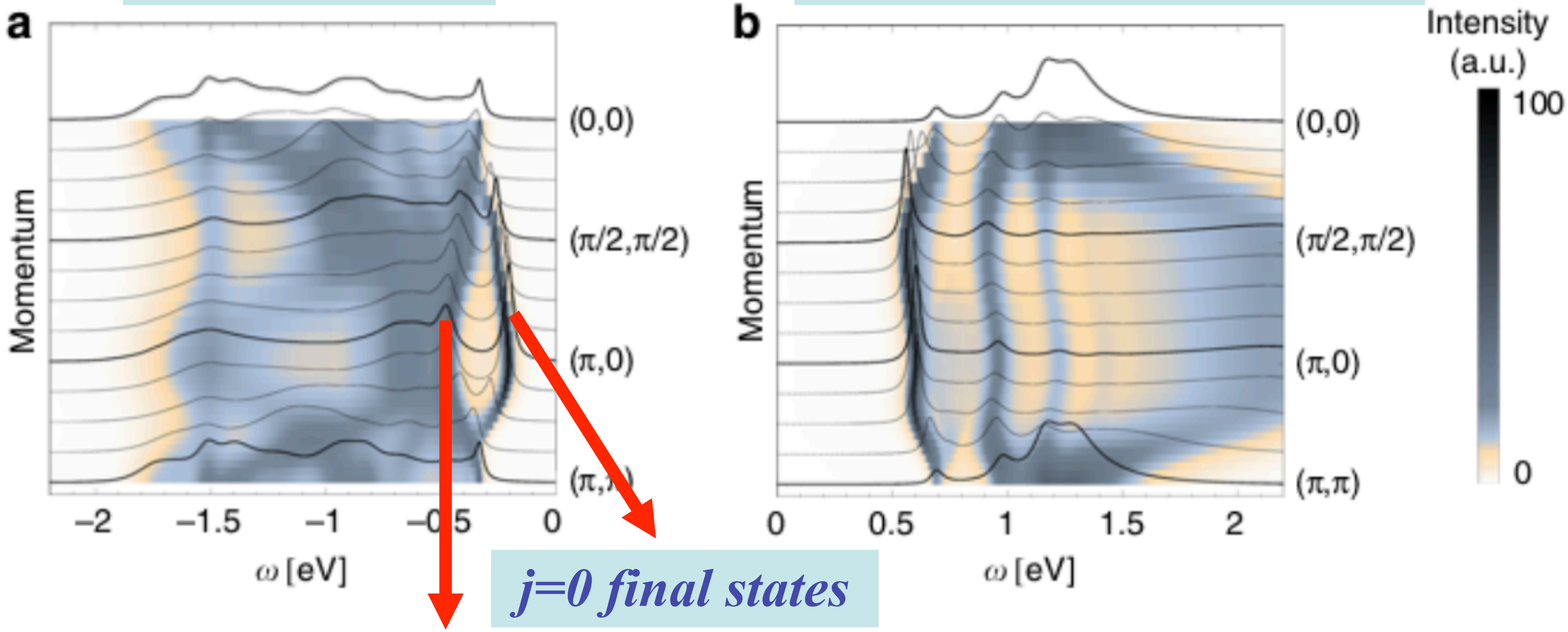


Strong electron-hole asymmetry

Pärschke, Wohlfeld, Foyevtsova & JvdB,
Nature Comm. 8, 686 (2017)
see also PRB 99, 121114(R)

Electron/hole propagation in Sr_2IrO_4

photoemission *inverse photoemission*



j=0 final states

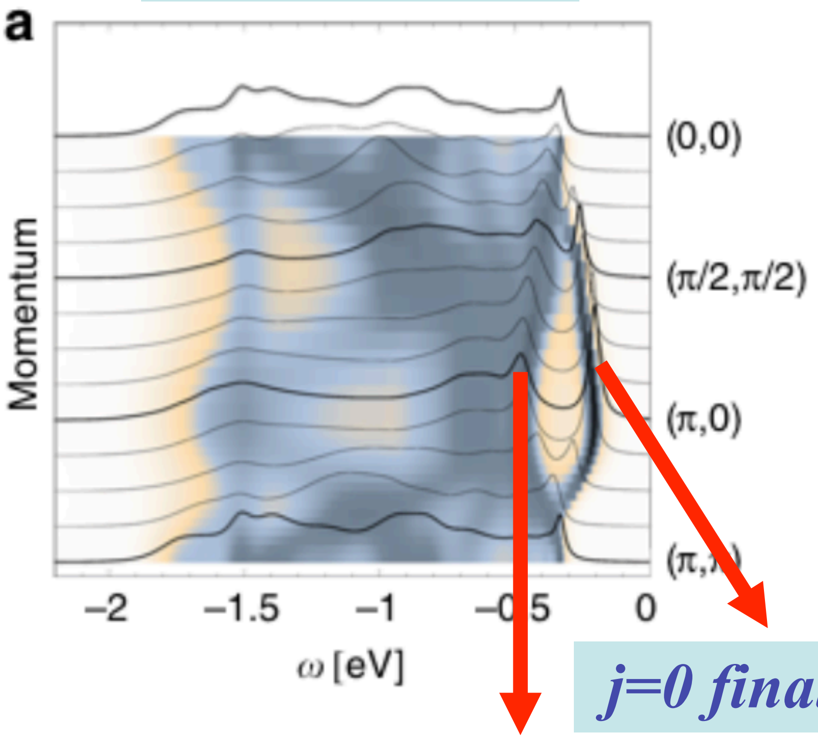
j=1 final states

Strong electron-hole asymmetry

Pärschke, Wohlfeld, Foyevtsova & JvdB,
Nature Comm. 8, 686 (2017)
see also PRB 99, 121114(R)

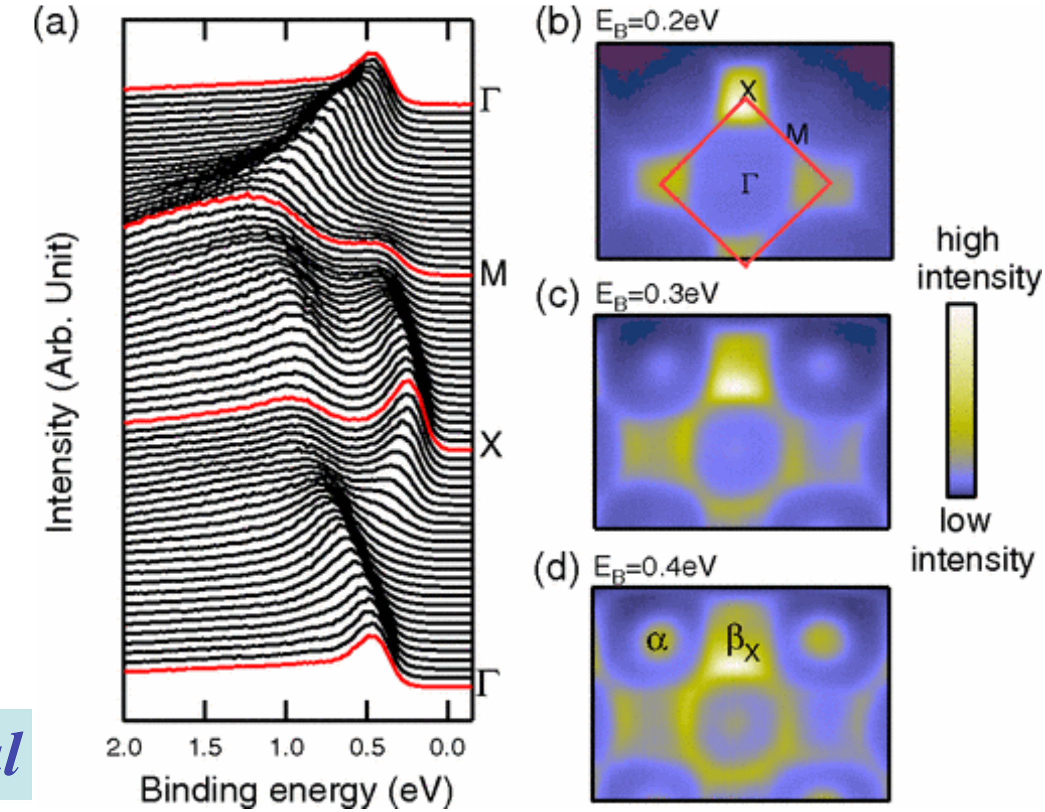
Electron/hole propagation in Sr_2IrO_4

photoemission



j=0 final

j=1 final states



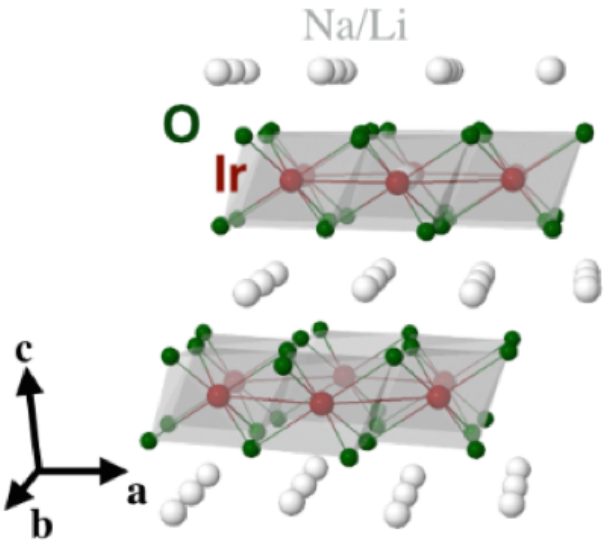
B. J. Kim et al., PRL 101, 076402 (2008)

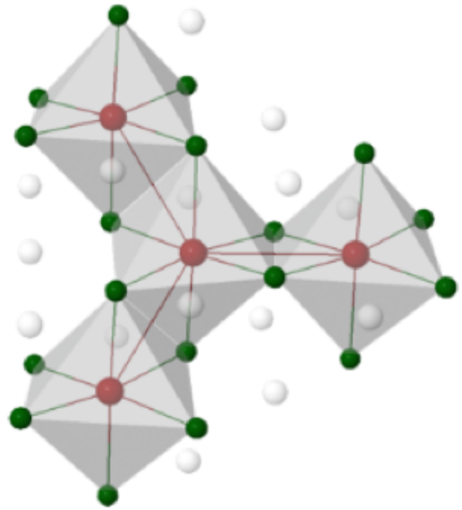
Strong electron-hole asymmetry

Pärschke, Wohlfeld, Foyevtsova & JvdB,
Nature Comm. 8, 686 (2017)
see also PRB 99, 121114(R)

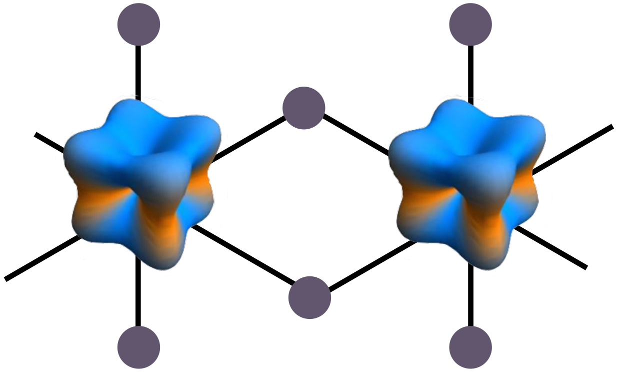
Exchange between edge-sharing j=1/2 moments

Na_2IrO_3 : honeycomb structure

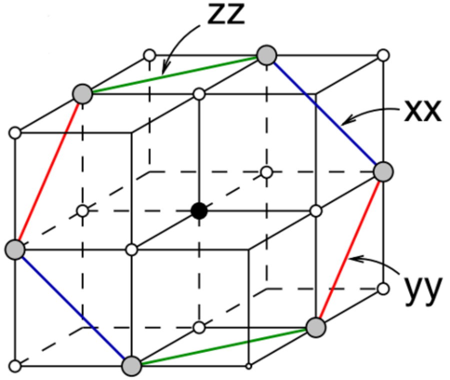




$\angle\, Ir - O - Ir \sim 90^o$



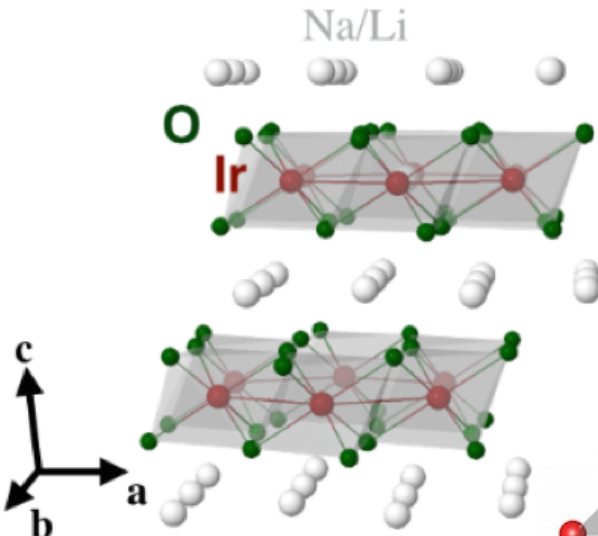
$$H_{ij}^{(1)} = K S_i^z S_j^z$$

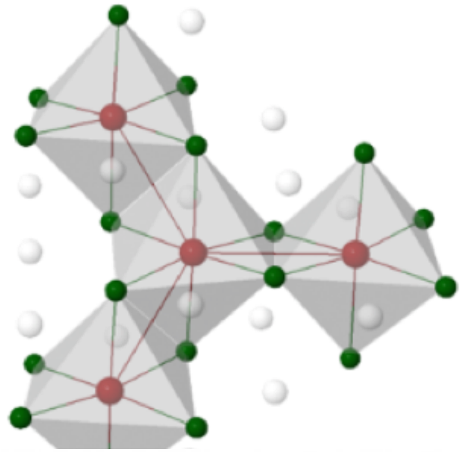


Jackeli & Khaliullin,
PRL 102, 017205 (2009)

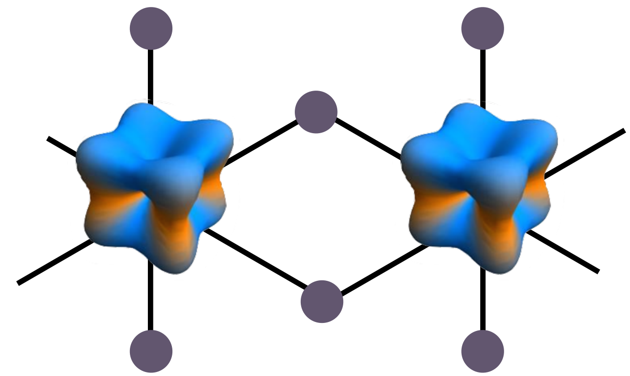
Exchange between edge-sharing j=1/2 moments

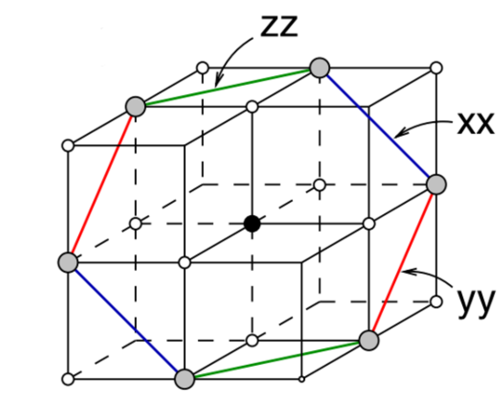
Na_2IrO_3 : honeycomb structure

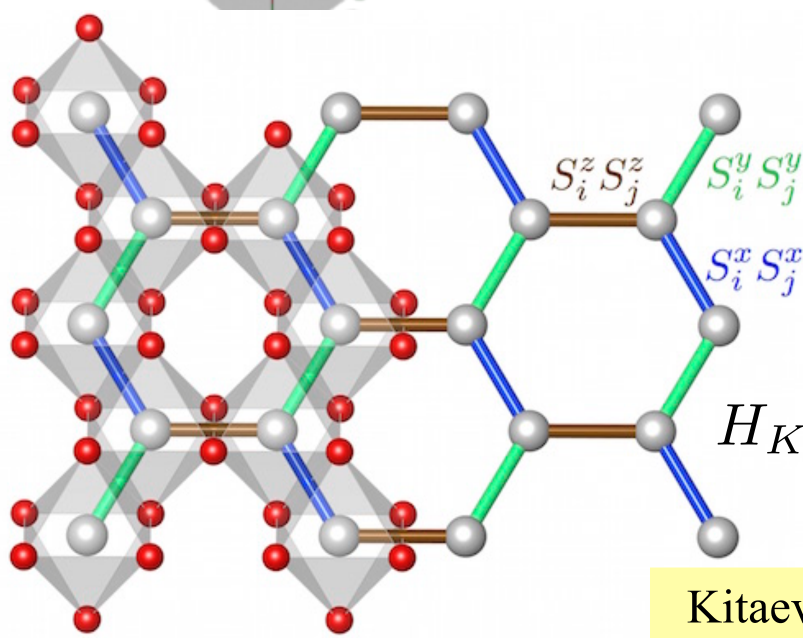




$\angle\, Ir - O - Ir \sim 90^o$







$$H_{ij}^{(1)} = K S_i^z S_j^z$$

$$H_{ij}^{(2)} = K S_i^y S_j^y$$

$$H_{ij}^{(3)} = K S_i^x S_j^x$$

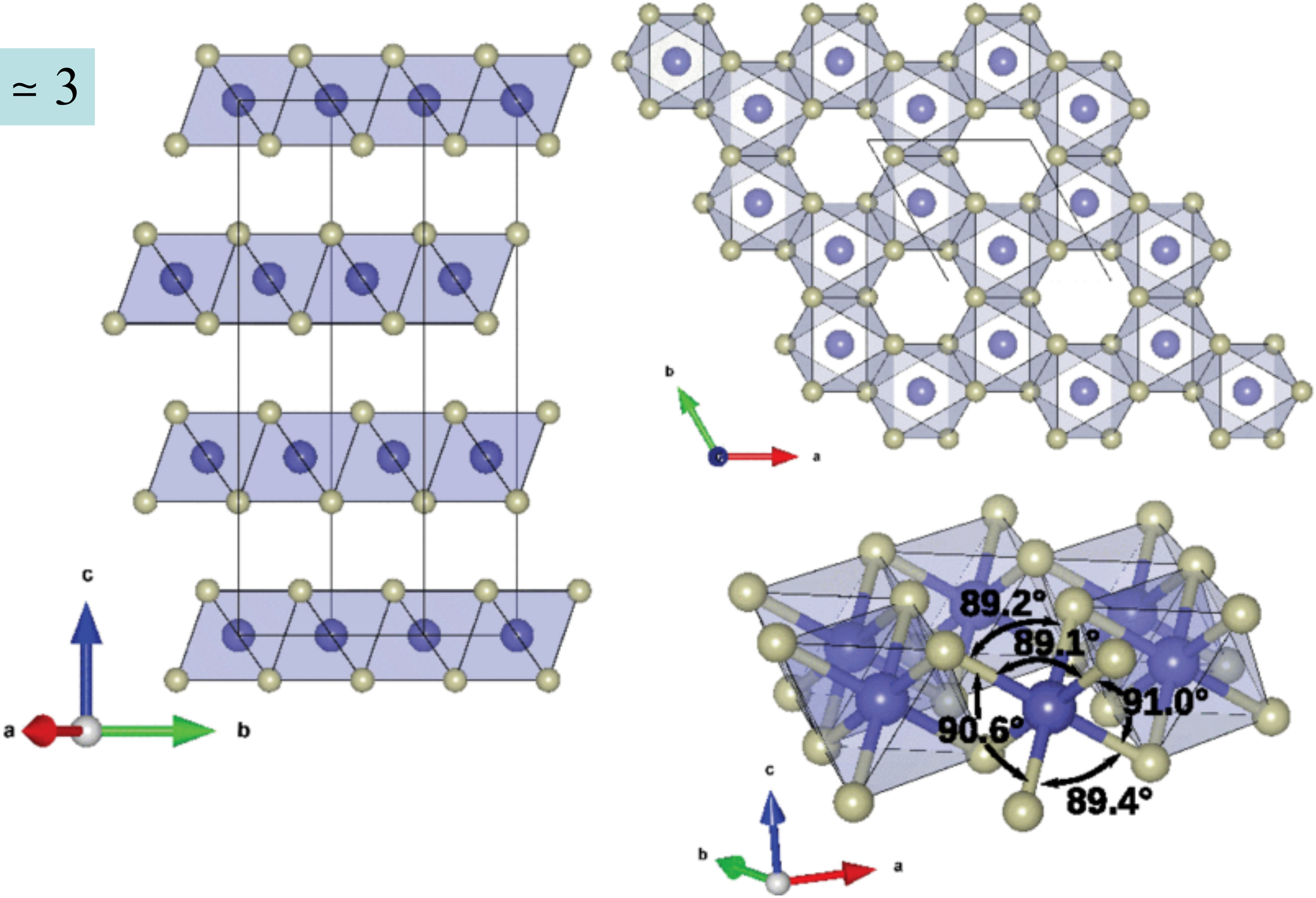
$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

Jackeli & Khaliullin,
PRL 102, 017205 (2009)

Kitaev, Ann. Phys. 321, 2 (2006)

Honeycomb α-$RuCl_3$

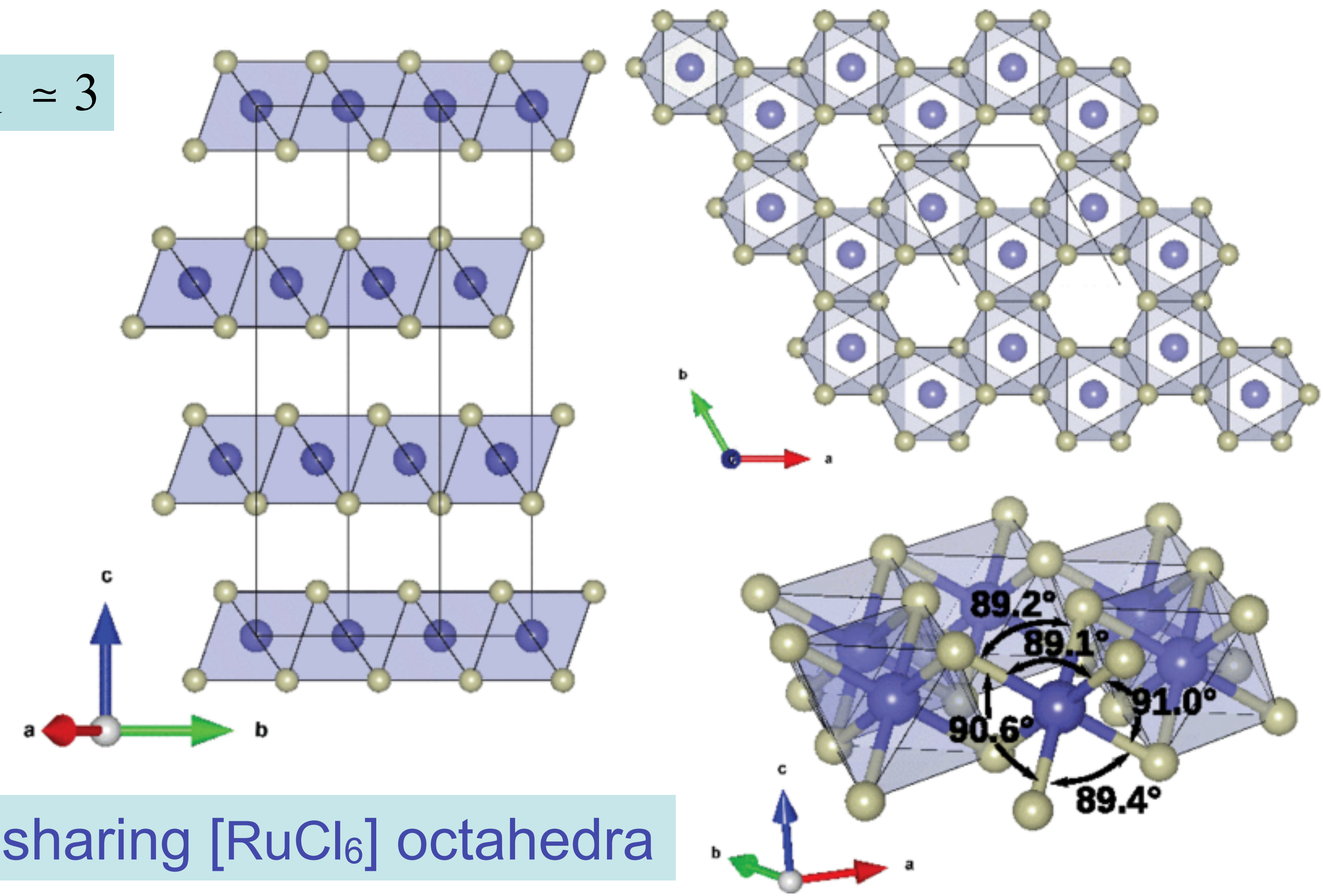
$\lambda_{Ir}/\lambda_{Ru} \simeq 3$



Plumb, Clancy, Sandilands, Shankar, Hu, Burch, H-Y Kee & Y-J Kim, PRB 90, 041112 (2014)

Honeycomb α-$RuCl_3$

$\lambda_{Ir}/\lambda_{Ru} \simeq 3$



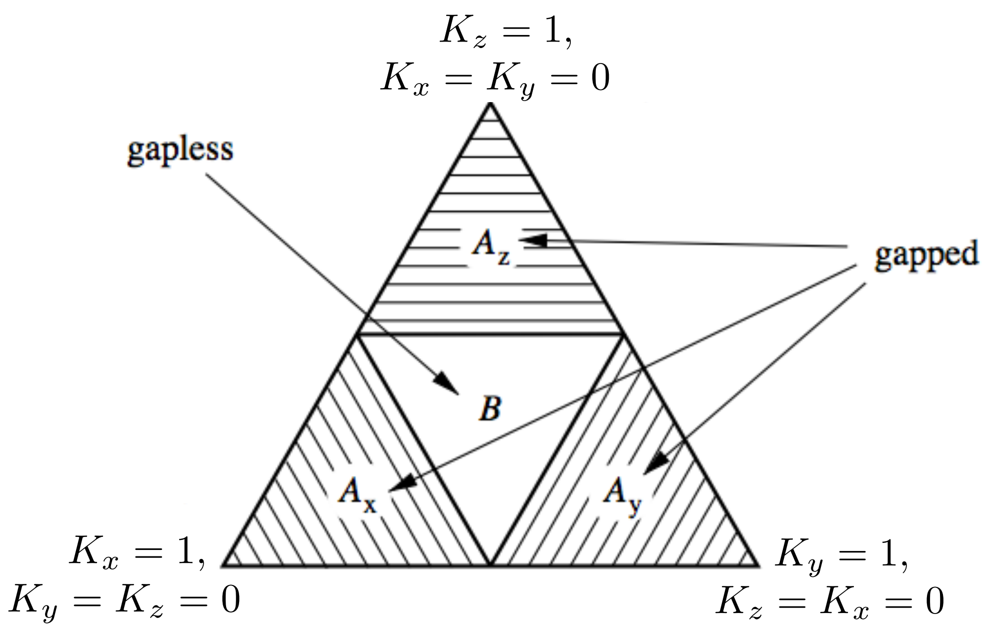
edge sharing [$RuCl_6$] octahedra

Plumb, Clancy, Sandilands, Shankar, Hu, Burch, H-Y Kee & Y-J Kim, PRB 90, 041112 (2014)

Honeycomb Kitaev model

$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

- phase diagram



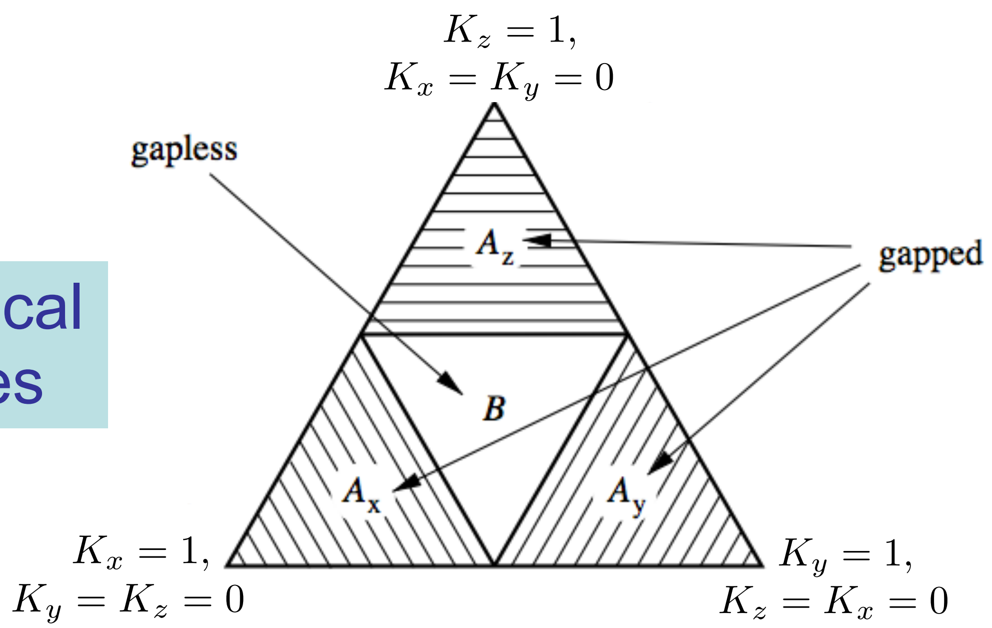
Kitaev, Ann. Phys. 321, 2 (2006)

Honeycomb Kitaev model

$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

- phase diagram

Abelian topological spin-liquid phases



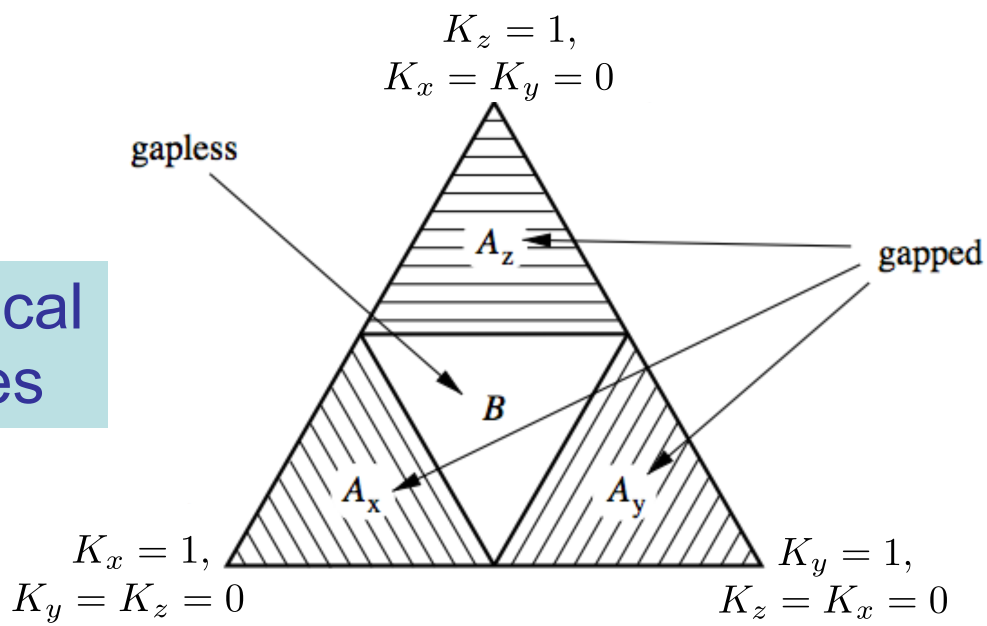
Kitaev, Ann. Phys. 321, 2 (2006)

Honeycomb Kitaev model

$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

- phase diagram

Abelian topological spin-liquid phases



- in magnetic field $H_{K-B} = K \sum_{\langle ij \rangle_\gamma} S_i^\gamma S_j^\gamma + B \sum_{i\gamma} S_i^\gamma$

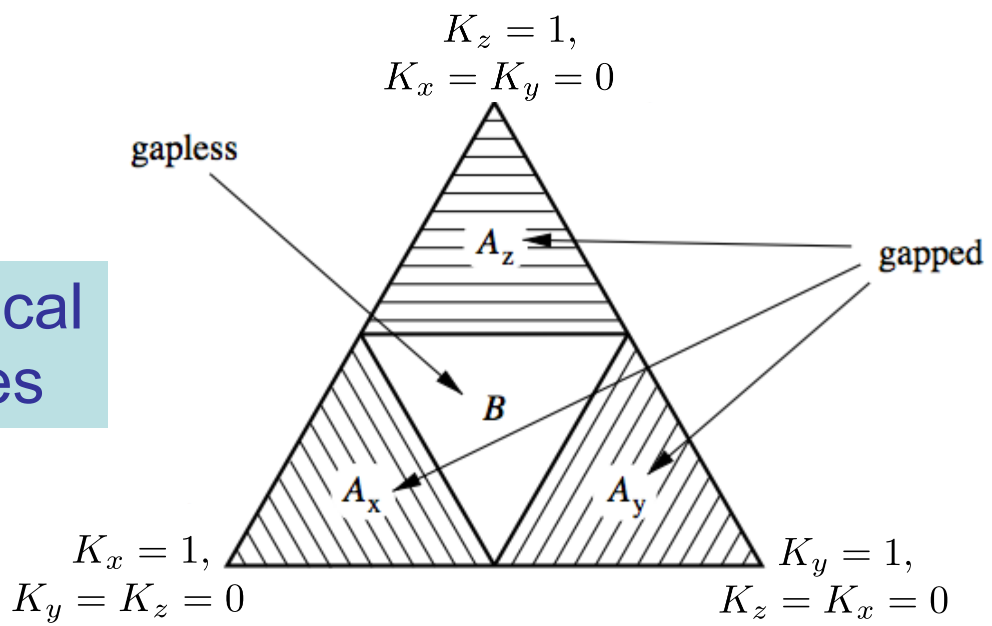
Kitaev, Ann. Phys. 321, 2 (2006)

Honeycomb Kitaev model

$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

- phase diagram

Abelian topological spin-liquid phases



- in magnetic field

$$H_{K-B} = K \sum_{\langle ij \rangle_\gamma} S_i^\gamma S_j^\gamma + B \sum_{i\gamma} S_i^\gamma$$

gapped non-Abelian topological spin-liquid phase

(perturbative in B/K)

Kitaev, Ann. Phys. 321, 2 (2006)

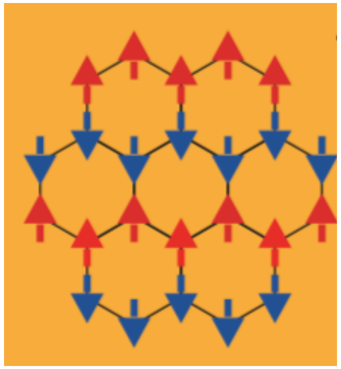
magnetic interactions in 213 iridates and α-$RuCl_3$

$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

magnetic interactions in 213 iridates and α-$RuCl_3$

$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

Experimentally: zig-zag order at low T

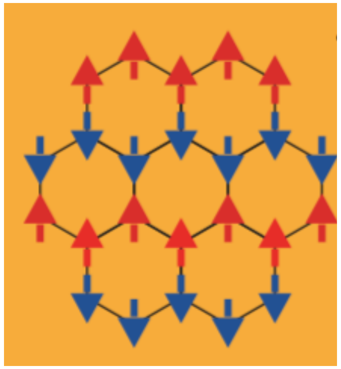


magnetic interactions in 213 iridates and α-$RuCl_3$

$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

$$H = K \sum_{\langle ij \rangle_\gamma} S_i^\gamma S_j^\gamma + J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + ...$$

Experimentally: zig-zag order at low T



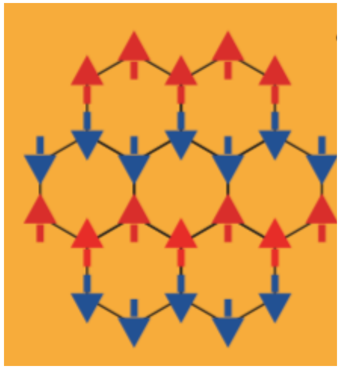
magnetic interactions in 213 iridates and α-RuCl$_3$

$$H_{Kitaev} = \sum_{\langle ij\rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

$$H = K \sum_{\langle ij\rangle_\gamma} S_i^\gamma S_j^\gamma + J \sum_{\langle ij\rangle} \vec{S}_i \cdot \vec{S}_j + \ldots.$$

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K \tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha\neq\beta} \Gamma_{\alpha\beta}(\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

Experimentally: zig-zag order at low T



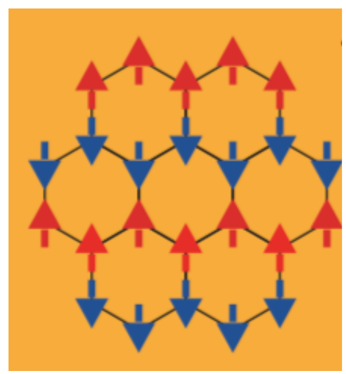
magnetic interactions in 213 iridates and α-$RuCl_3$

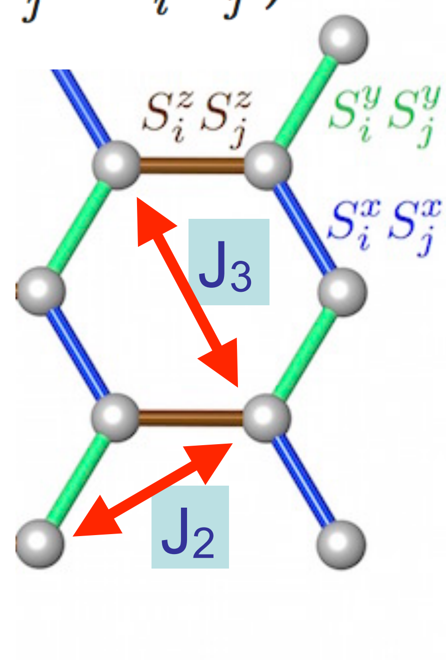
$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

$$H = K \sum_{\langle ij \rangle_\gamma} S_i^\gamma S_j^\gamma + J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + ...$$

$$\mathcal{H}_{i,j} = J\, \tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K \tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha \neq \beta} \Gamma_{\alpha\beta} (\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

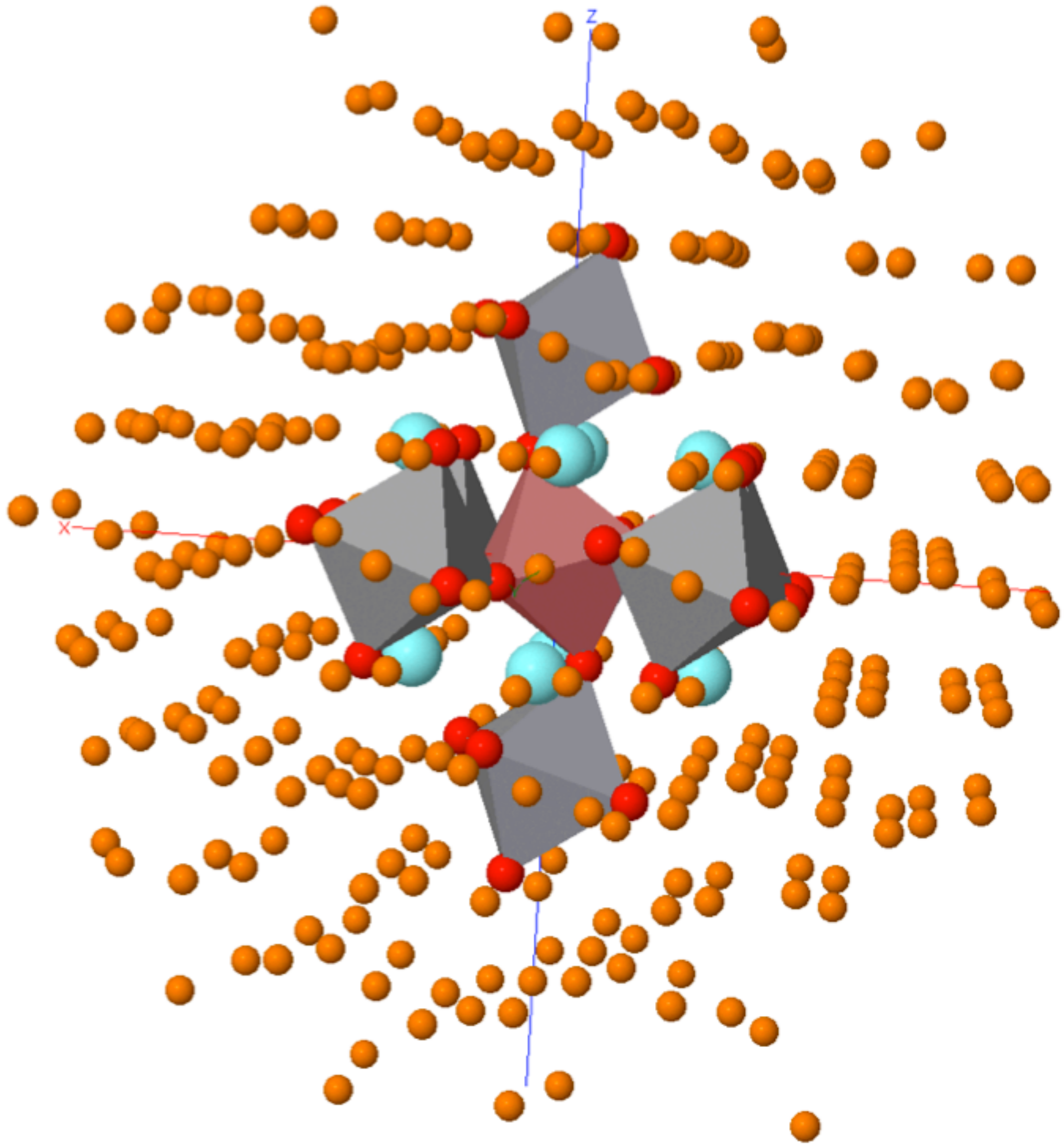
Experimentally: zig-zag order at low T





QC: wavefunction-based correlation methods

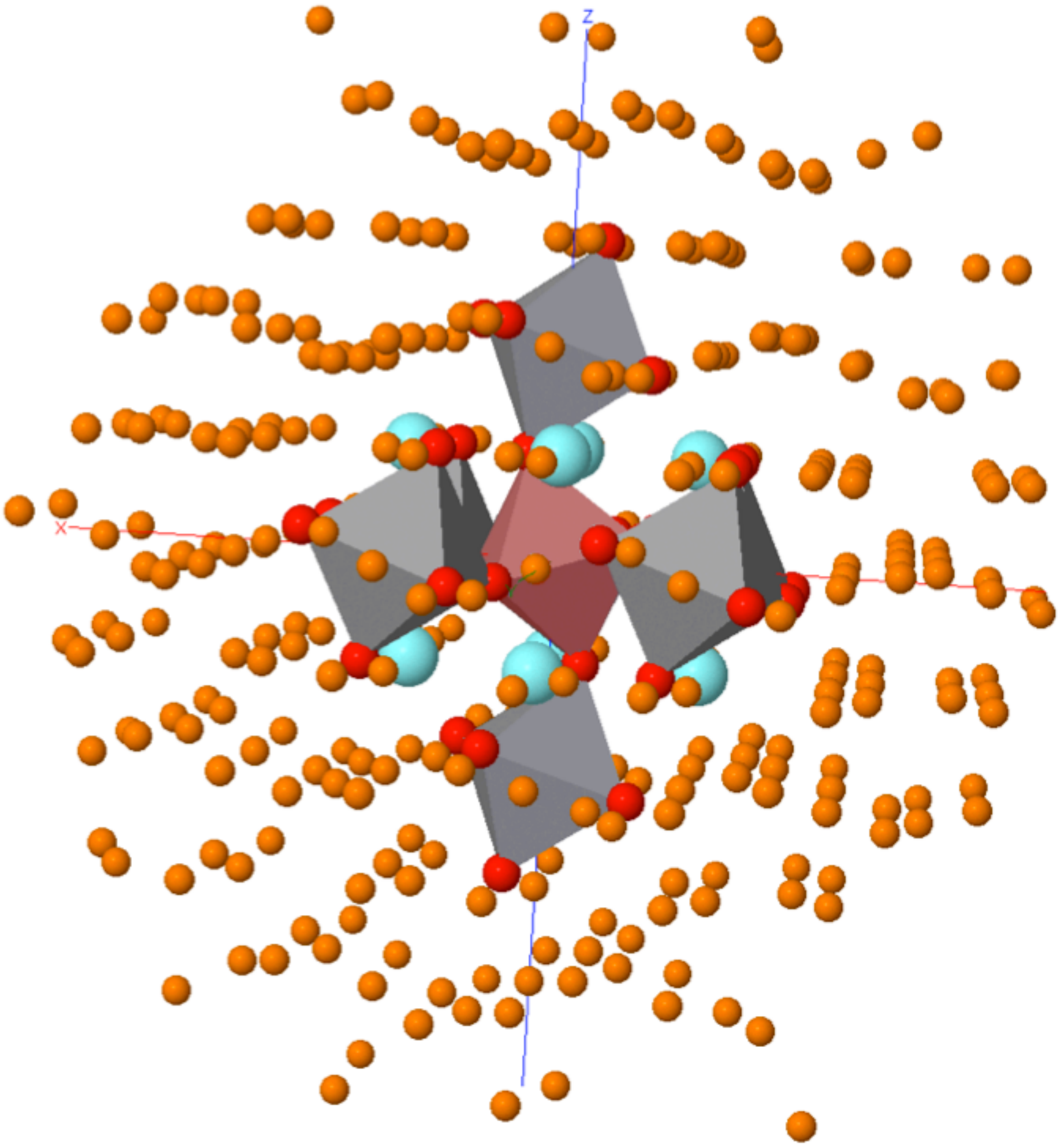
Finite embedded clusters



QC: wavefunction-based correlation methods

Fully ab initio for ground and excited states

Finite embedded clusters

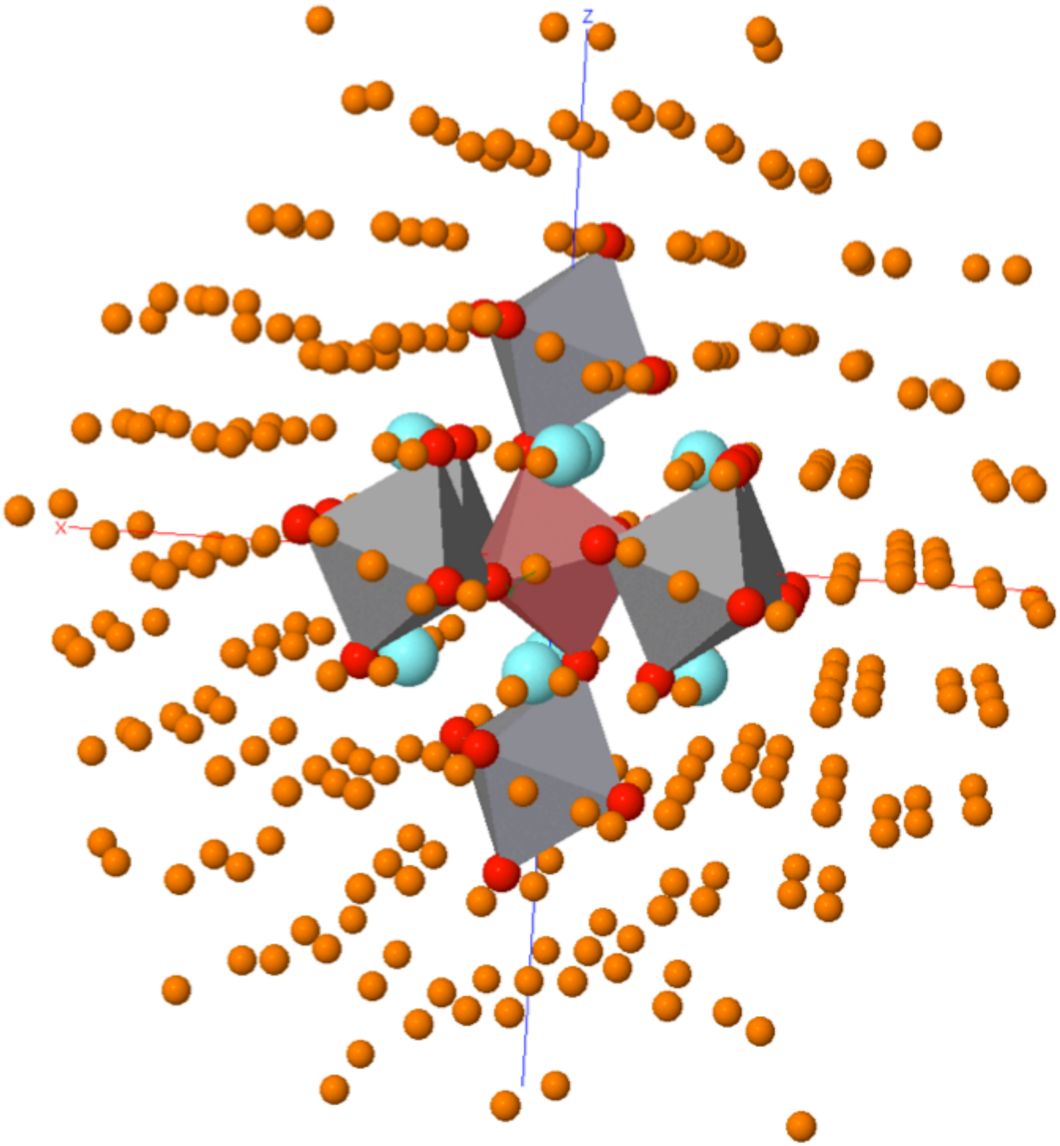


QC: wavefunction-based correlation methods

Fully ab initio for ground and excited states

Fully correlated: multi-configuration wave-functions

Finite embedded clusters



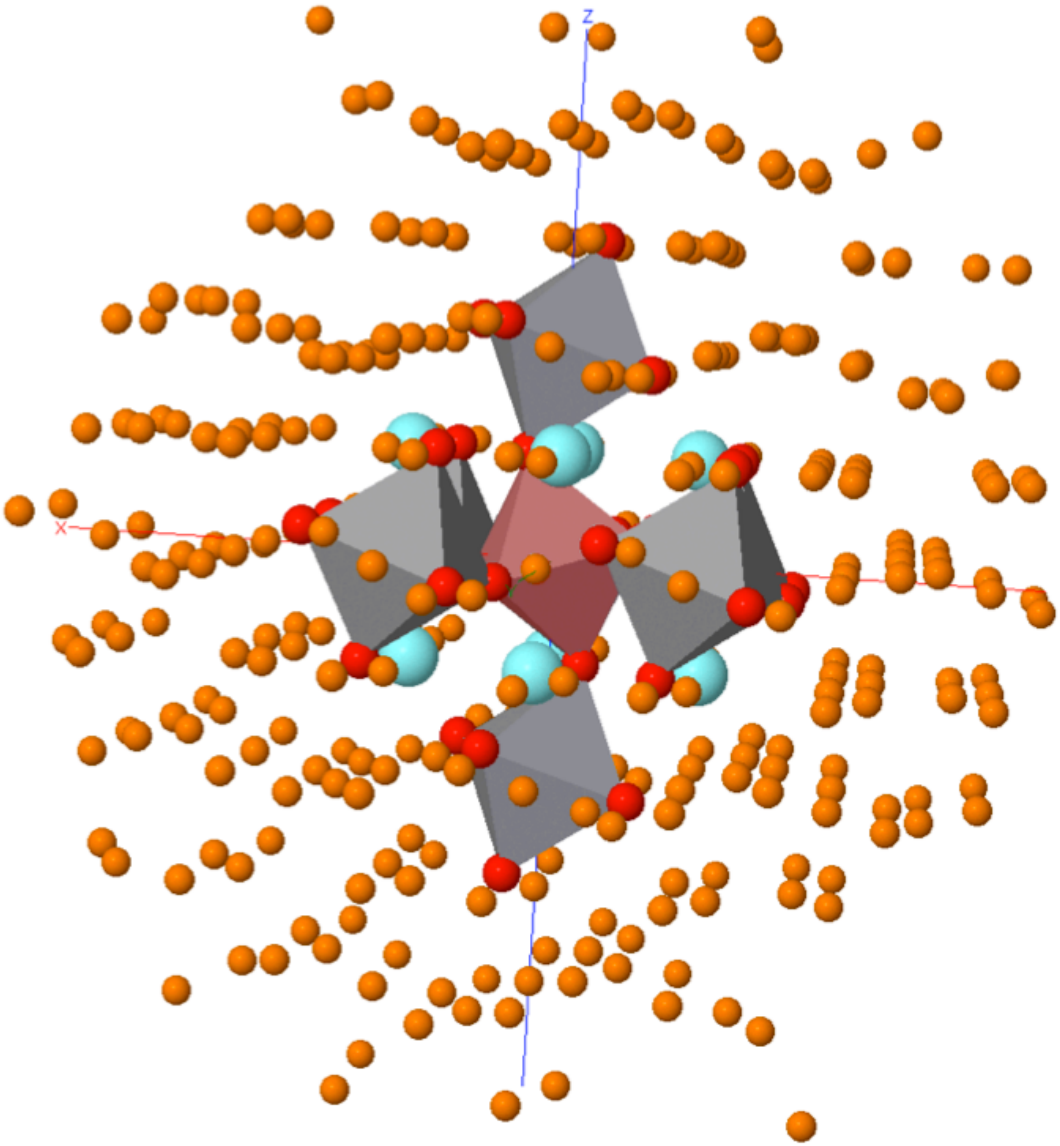
QC: wavefunction-based correlation methods

Fully ab initio for ground and excited states

Fully correlated: multi-configuration wave-functions

Heavy machinery

Finite embedded clusters



QC: wavefunction-based correlation methods

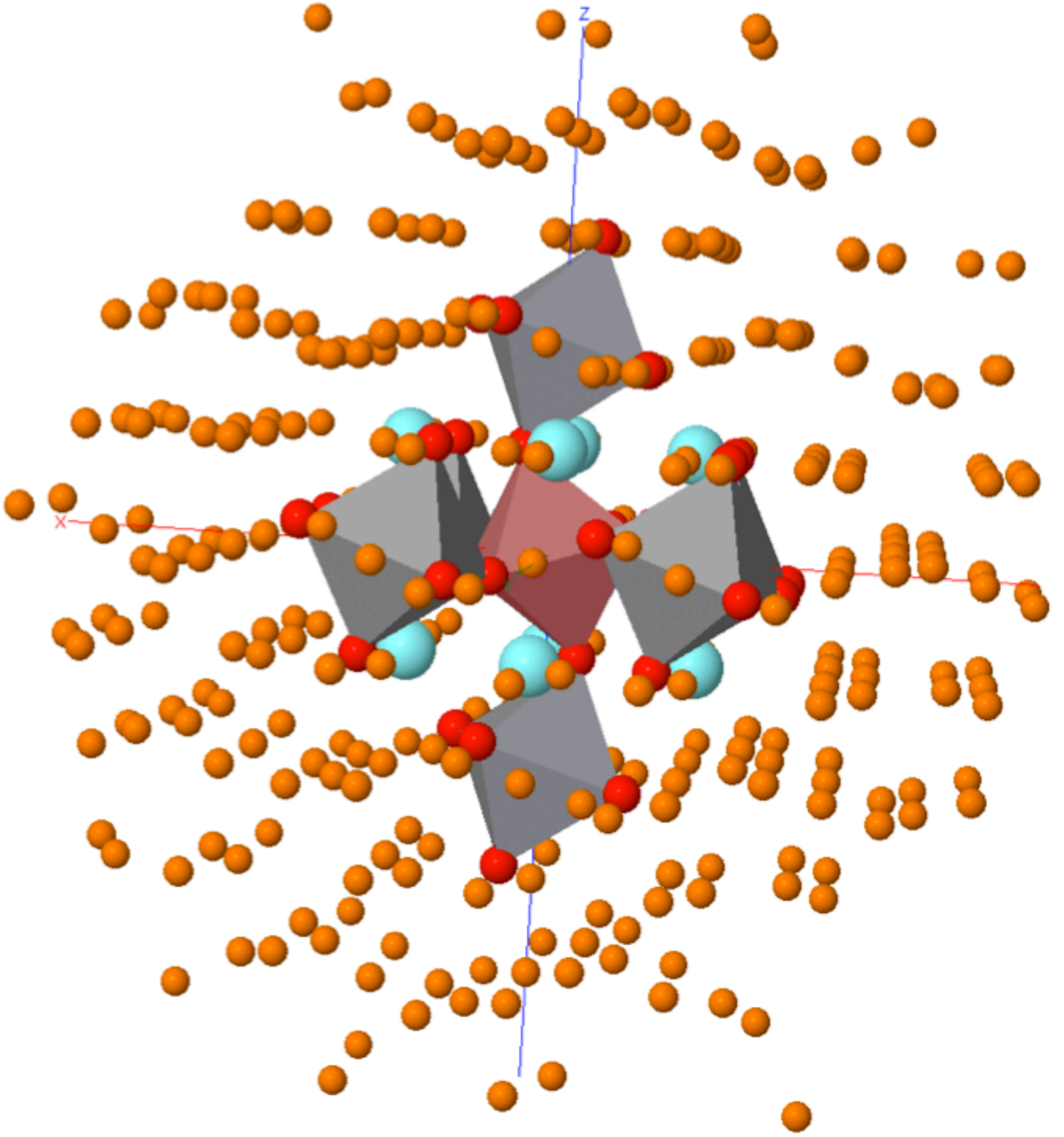
Fully ab initio for ground and excited states

Fully correlated: multi-configuration wave-functions

Heavy machinery

Excellent for systems with localized electrons

Finite embedded clusters



QC: wavefunction-based correlation methods

Fully ab initio for ground and excited states

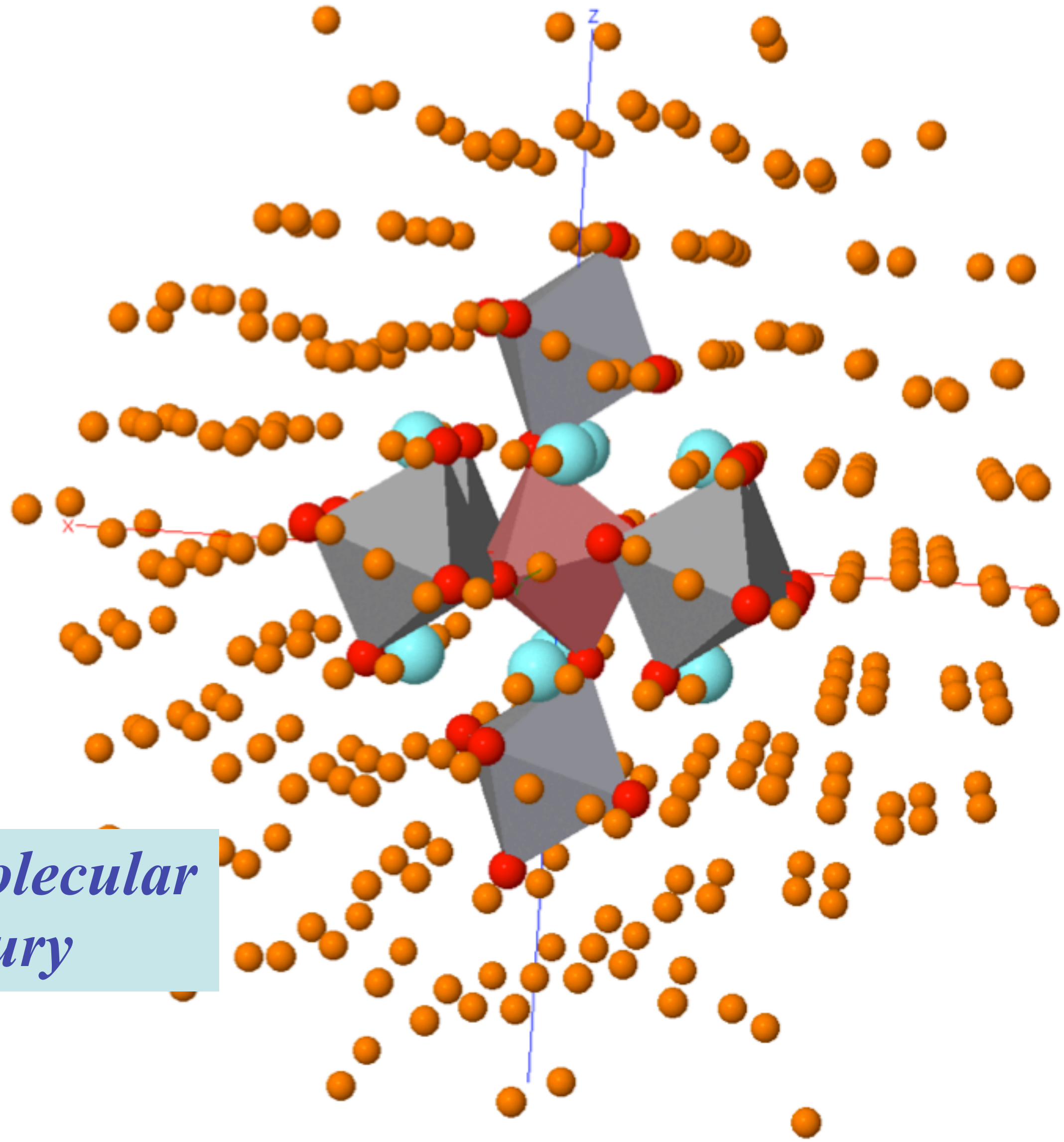
Fully correlated: multi-configuration wave-functions

Heavy machinery

Excellent for systems with localized electrons

Approximations tested in molecular systems since half century

Finite embedded clusters



QC: wavefunction-based correlation methods

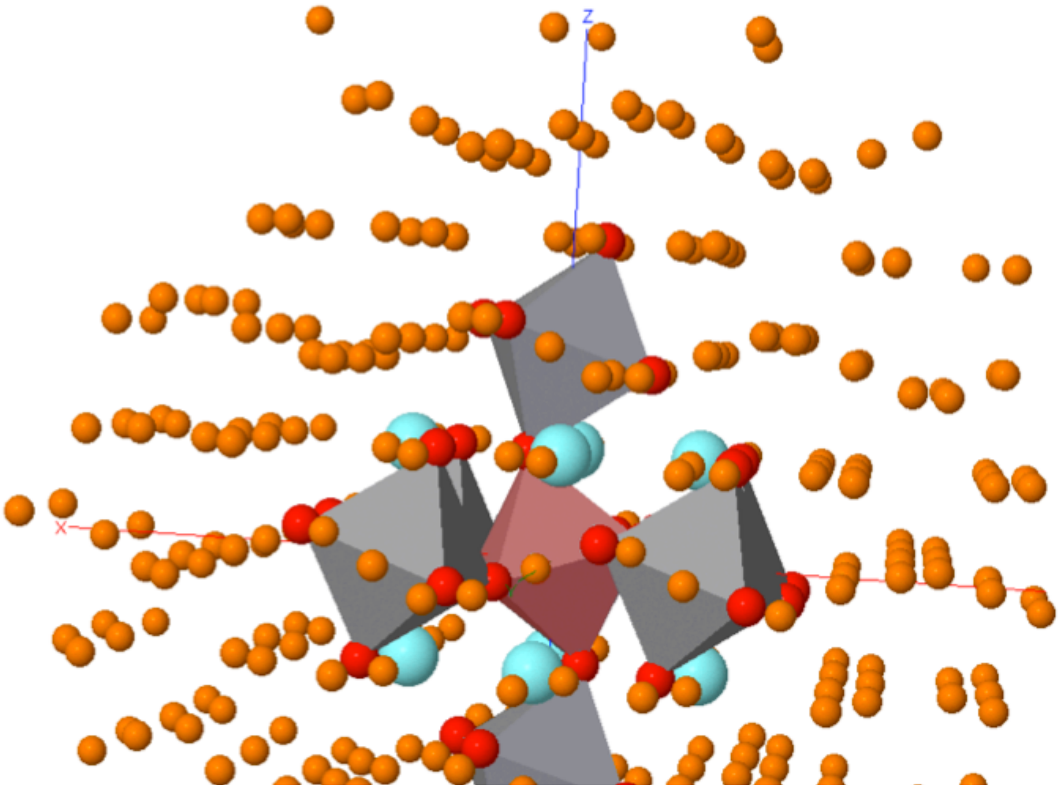
Fully ab initio for ground and excited states

Fully correlated: multi-configuration wave-functions

Heavy machinery

Excellent for systems with localized electrons

Finite embedded clusters



Our scheme : direct-space multireference CI, finite embedded clusters

The infinite solid-state environment : one-electron embedding potential

- simplest: point-charge array
- more advanced: based on prior periodic Hartree-Fock

honeycomb Kitaev materials to consider

Na_2IrO_3

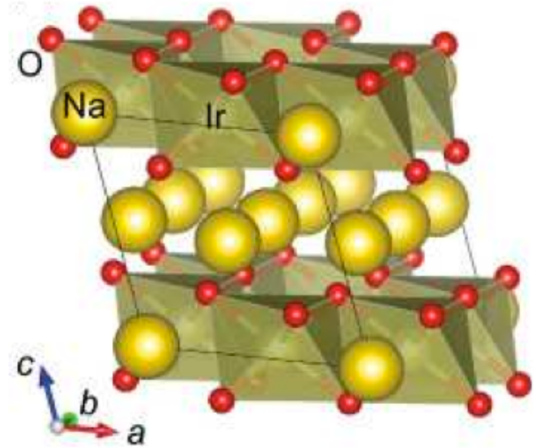
Li_2IrO_3

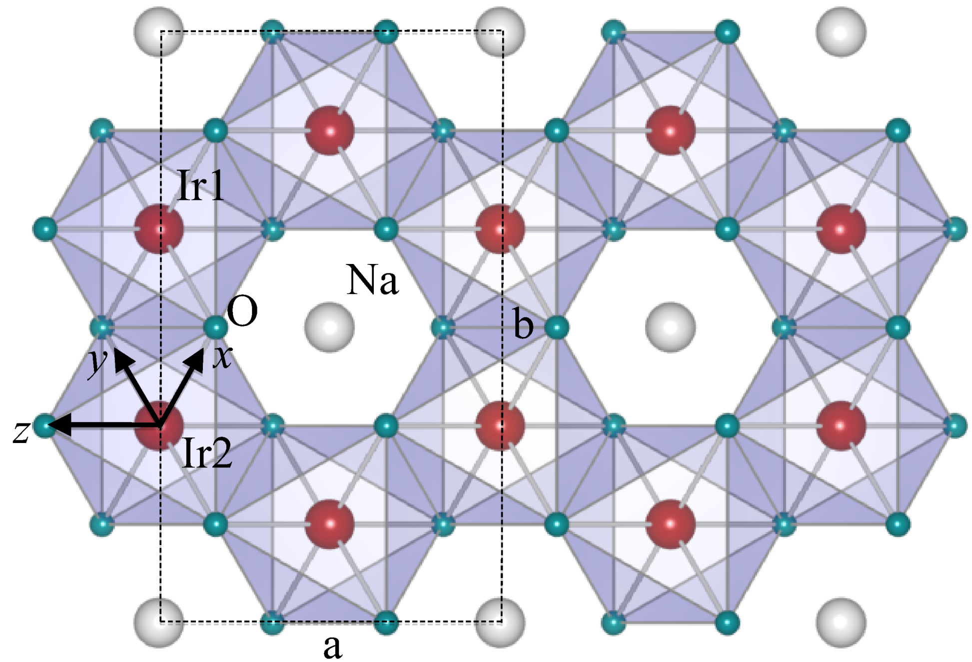
$H_3LiIr_2O_6$

K_2IrO_3

$RuCl_3$

honeycomb Na_2IrO_3





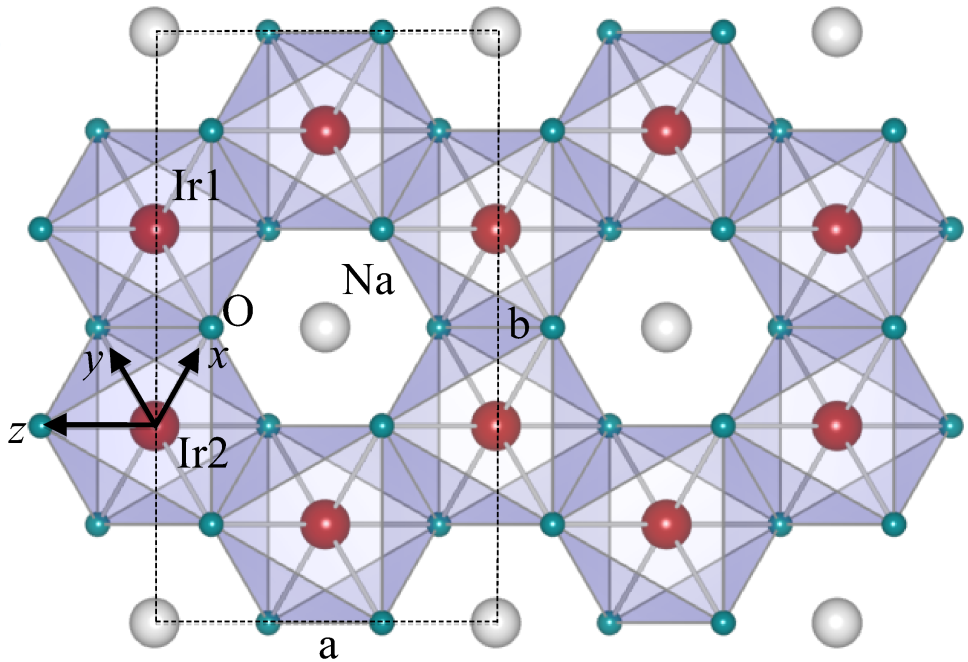
$$\mathcal{H}_{ij}^{\mathrm{D_{2h}}} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + J_{xy}\left(\tilde{S}_i^x \tilde{S}_j^y + \tilde{S}_i^y \tilde{S}_j^x\right)$$

Katakuri, Nishimoto, Yushankhai, Stoyanova, Kandpal, Choi, Coldea, Rousochatzakis, Hozoi & JvdB, NJP 16, 013056 (2014)

TABLE II. Energy splittings and effective parameters (meV) for the four lowest magnetic states of two NN IrO_6 octahedra in the $C2/m$ structure of 4. The weight of $(\uparrow\downarrow + \downarrow\uparrow)/\sqrt{2}$ and $(\uparrow\uparrow + \downarrow\downarrow)/\sqrt{2}$ in Ψ_1' and Ψ_2', respectively, is ≈98%, see text.

Method	CAS+SOC	MRCI+SOC
$\measuredangle$(Ir-O-Ir)=99.45°, $d(\mathrm{Ir_1\text{-}Ir_2})$=3.138 Å (×1)[a]:		
Ψ_2'	0.0	0.0
$\Psi_3 = (\uparrow\uparrow - \downarrow\downarrow)/\sqrt{2}$	0.2	0.5
$\Psi_S = (\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$	4.4	5.5
Ψ_1'	6.3	10.5
(J, K, J_{xy})	(1.9,−12.4,0.2)	(5.0,−20.5,0.5)
$\measuredangle$(Ir-O-Ir)=97.97°, $d(\mathrm{Ir_2\text{-}Ir_3})$=3.130 Å (×2)[b]:		
Ψ_2'	0.0	0.0
$\Psi_3 = (\uparrow\uparrow - \downarrow\downarrow)/\sqrt{2}$	0.3	1.2
$\Psi_S = (\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$	4.6	6.7
Ψ_1'	5.8	8.2
(J, K, J_{xy})	(1.2,−11.3,0.3)	(1.5,−15.2,1.2)

[a] $d(\mathrm{Ir\text{-}O_{1,2}})$=2.056 Å.
[b] $d(\mathrm{Ir\text{-}O_1})$=2.065 Å, $d(\mathrm{Ir\text{-}O_2})$=2.083 Å.



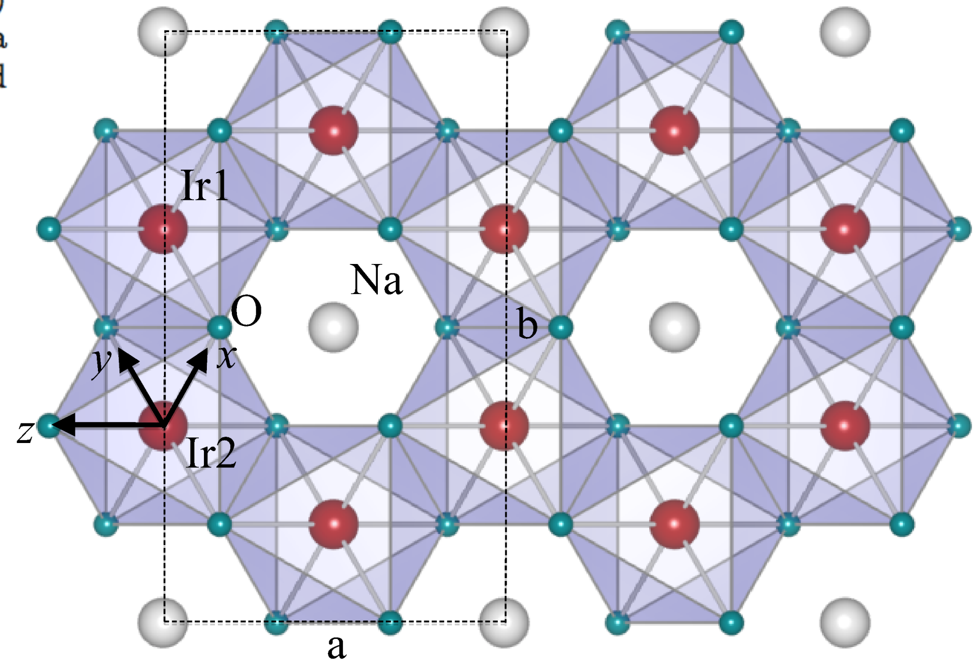
$$\mathcal{H}_{ij}^{\mathrm{D_{2h}}} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + J_{xy}\left(\tilde{S}_i^x \tilde{S}_j^y + \tilde{S}_i^y \tilde{S}_j^x\right)$$

Katakuri, Nishimoto, Yushankhai, Stoyanova, Kandpal, Choi, Coldea, Rousochatzakis, Hozoi & JvdB, NJP 16, 013056 (2014)

TABLE II. Energy splittings and effective parameters (meV) for the four lowest magnetic states of two NN IrO_6 octahedra in the $C2/m$ structure of 4. The weight of $(\uparrow\downarrow+\downarrow\uparrow)/\sqrt{2}$ and $(\uparrow\uparrow+\downarrow\downarrow)/\sqrt{2}$ in Ψ_1' and Ψ_2', respectively, is ≈98%, see text.

Method	CAS+SOC	MRCI+SOC
$\measuredangle$(Ir-O-Ir)=99.45°, $d(\mathrm{Ir_1\text{-}Ir_2})$=3.138 Å (×1)[a]:		
Ψ_2'	0.0	0.0
$\Psi_3=(\uparrow\uparrow-\downarrow\downarrow)/\sqrt{2}$	0.2	0.5
$\Psi_S=(\uparrow\downarrow-\downarrow\uparrow)/\sqrt{2}$	4.4	5.5
Ψ_1'	6.3	10.5
(J,K,J_{xy})	(1.9,−12.4,0.2)	(5.0,−20.5,0.5)
$\measuredangle$(Ir-O-Ir)=97.97°, $d(\mathrm{Ir_2\text{-}Ir_3})$=3.130 Å (×2)[b]:		
Ψ_2'	0.0	0.0
$\Psi_3=(\uparrow\uparrow-\downarrow\downarrow)/\sqrt{2}$	0.3	1.2
$\Psi_S=(\uparrow\downarrow-\downarrow\uparrow)/\sqrt{2}$	4.6	6.7
Ψ_1'	5.8	8.2
(J,K,J_{xy})	(1.2,−11.3,0.3)	(1.5,−15.2,1.2)

[a] $d(\mathrm{Ir\text{-}O_{1,2}})$=2.056 Å.
[b] $d(\mathrm{Ir\text{-}O_1})$=2.065 Å, $d(\mathrm{Ir\text{-}O_2})$=2.083 Å.



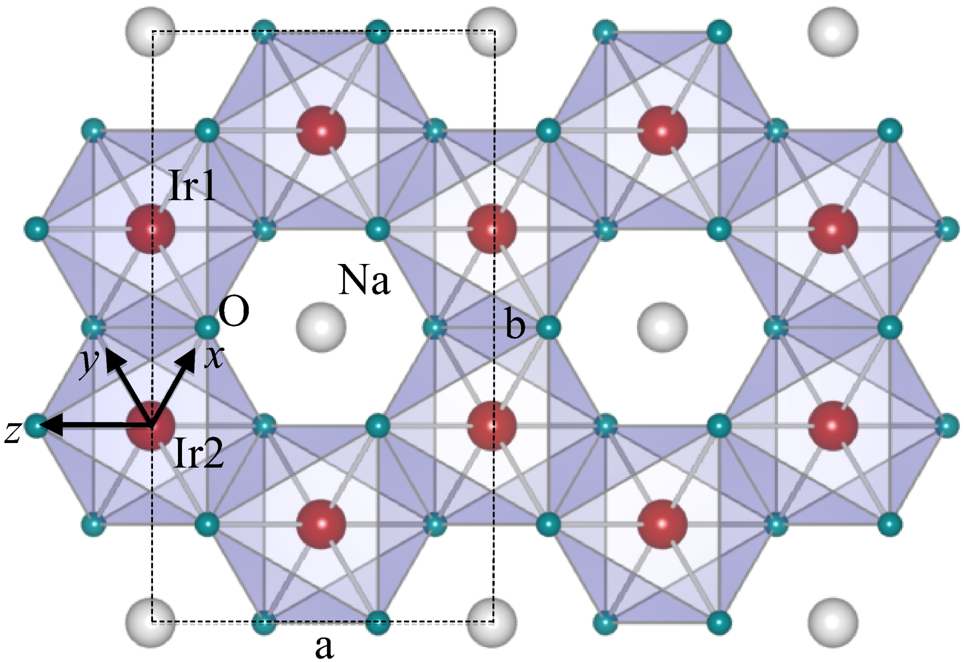
$$\mathcal{H}_{ij}^{\mathrm{D_{2h}}}=J\,\tilde{\mathbf{S}}_i\cdot\tilde{\mathbf{S}}_j+K\,\tilde{S}_i^z\tilde{S}_j^z+J_{xy}\left(\tilde{S}_i^x\tilde{S}_j^y+\tilde{S}_i^y\tilde{S}_j^x\right)$$

Katakuri, Nishimoto, Yushankhai, Stoyanova, Kandpal, Choi, Coldea, Rousochatzakis, Hozoi & JvdB, NJP 16, 013056 (2014)

TABLE II. Energy splittings and effective parameters (meV) for the four lowest magnetic states of two NN IrO_6 octahedra in the $C2/m$ structure of 4. The weight of $(\uparrow\downarrow+\downarrow\uparrow)/\sqrt{2}$ and $(\uparrow\uparrow+\downarrow\downarrow)/\sqrt{2}$ in Ψ_1' and Ψ_2', respectively, is ≈98%, see text.

Method	CAS+SOC	MRCI+SOC
$\measuredangle$(Ir-O-Ir)=99.45°, $d(Ir_1\text{-}Ir_2)$=3.138 Å (×1)[a]:		
Ψ_2'	0.0	0.0
$\Psi_3=(\uparrow\uparrow-\downarrow\downarrow)/\sqrt{2}$	0.2	0.5
$\Psi_S=(\uparrow\downarrow-\downarrow\uparrow)/\sqrt{2}$	4.4	5.5
Ψ_1'	6.3	10.5
(J,K,J_{xy})	(1.9,−12.4,0.2)	(5.0,−20.5,0.5)
$\measuredangle$(Ir-O-Ir)=97.97°, $d(Ir_2\text{-}Ir_3)$=3.130 Å (×2)[b]:		
Ψ_2'	0.0	0.0
$\Psi_3=(\uparrow\uparrow-\downarrow\downarrow)/\sqrt{2}$	0.3	1.2
$\Psi_S=(\uparrow\downarrow-\downarrow\uparrow)/\sqrt{2}$	4.6	6.7
Ψ_1'	5.8	8.2
(J,K,J_{xy})	(1.2,−11.3,0.3)	(1.5,−15.2,1.2)

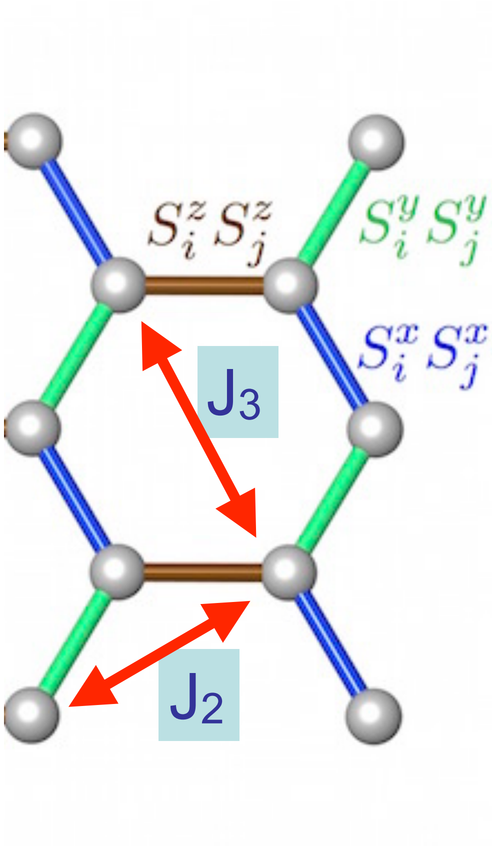
[a] $d(Ir\text{-}O_{1,2})$=2.056 Å.
[b] $d(Ir\text{-}O_1)$=2.065 Å, $d(Ir\text{-}O_2)$=2.083 Å.

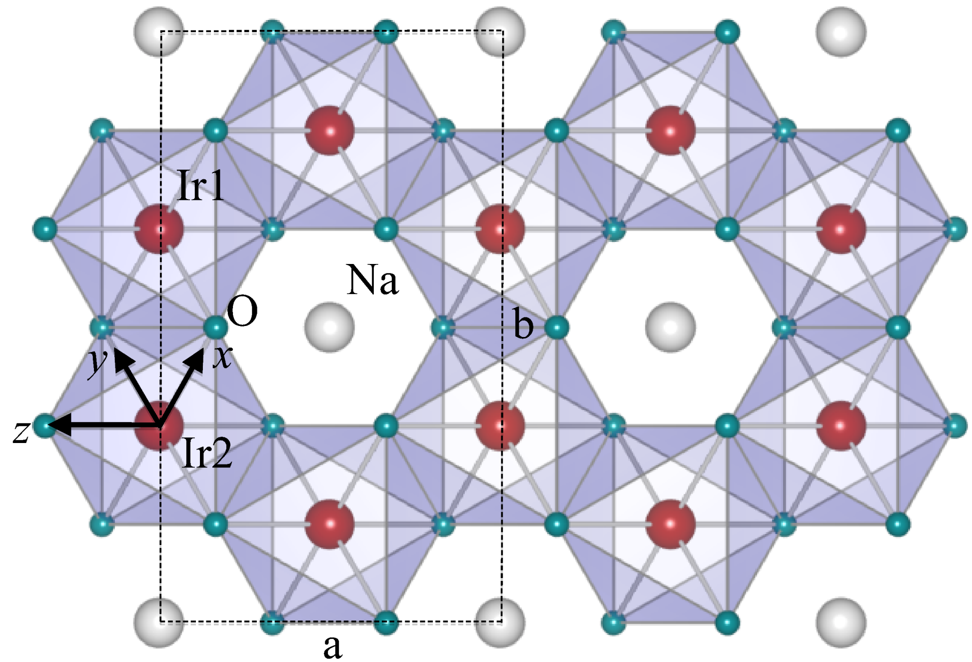


$$\mathcal{H}_{ij}^{\mathrm{D_{2h}}} = J\,\tilde{\mathbf{S}}_i\cdot\tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z\tilde{S}_j^z + J_{xy}\left(\tilde{S}_i^x\tilde{S}_j^y + \tilde{S}_i^y\tilde{S}_j^x\right)$$

K large and FM, J small and AFM substantial anisotropy between links

Katakuri, Nishimoto, Yushankhai, Stoyanova, Kandpal, Choi, Coldea, Rousochatzakis, Hozoi & JvdB, NJP 16, 013056 (2014)

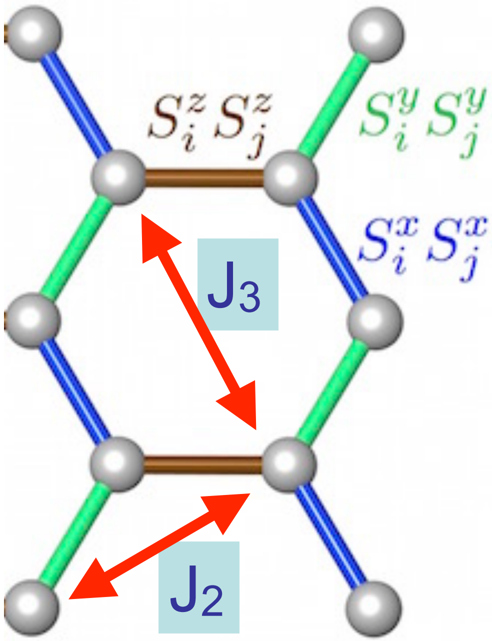


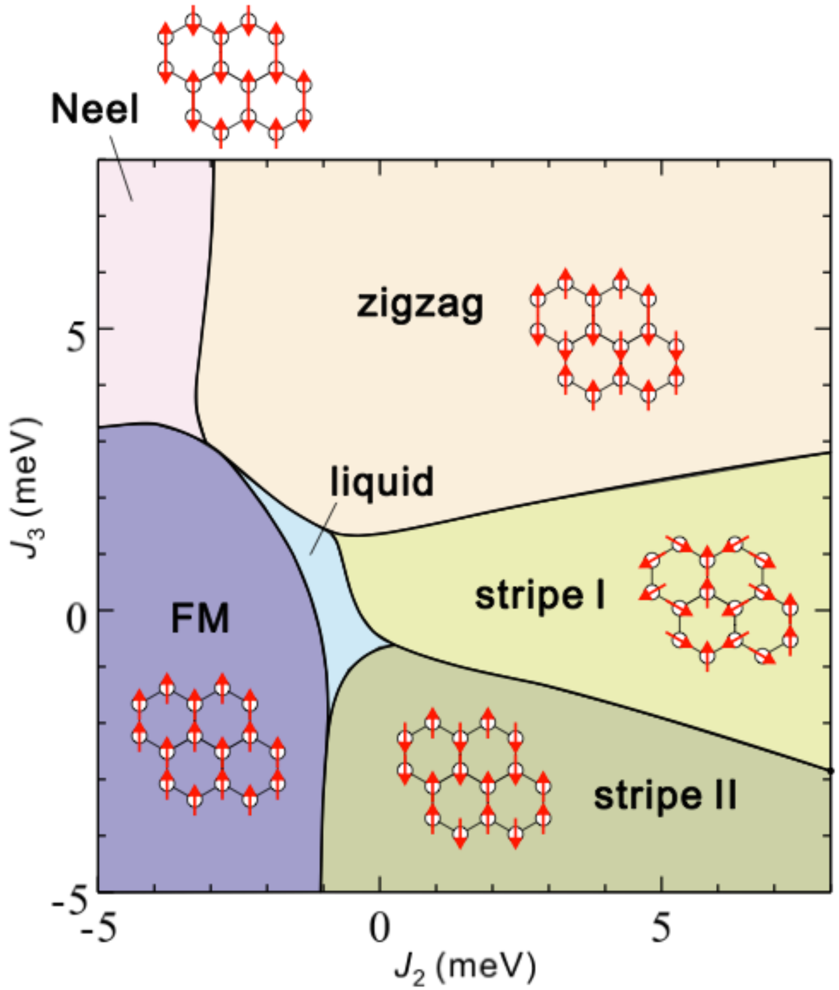


$$\mathcal{H}_{ij}^{\mathrm{D_{2h}}} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + J_{xy}\left(\tilde{S}_i^x \tilde{S}_j^y + \tilde{S}_i^y \tilde{S}_j^x\right)$$

+ longer range Heisenberg J_2 and J_3

Katakuri, Nishimoto, Yushankhai, Stoyanova, Kandpal, Choi, Coldea, Rousochatzakis, Hozoi & JvdB, NJP 16, 013056 (2014)



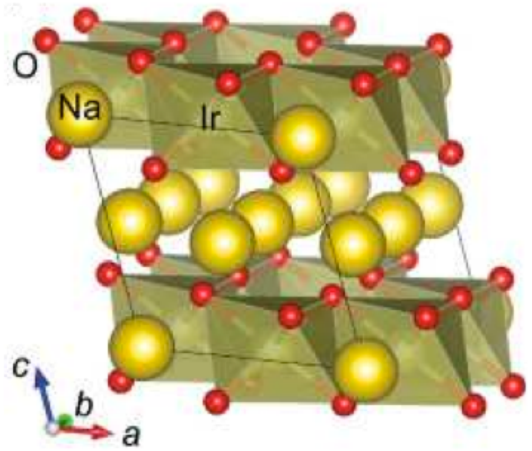


$$\mathcal{H}_{ij}^{\mathrm{D_{2h}}} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + J_{xy}\left(\tilde{S}_i^x \tilde{S}_j^y + \tilde{S}_i^y \tilde{S}_j^x\right)$$

+ longer range Heisenberg J_2 and J_3

Katakuri, Nishimoto, Yushankhai, Stoyanova, Kandpal, Choi, Coldea, Rousochatzakis, Hozoi & JvdB, NJP 16, 013056 (2014)

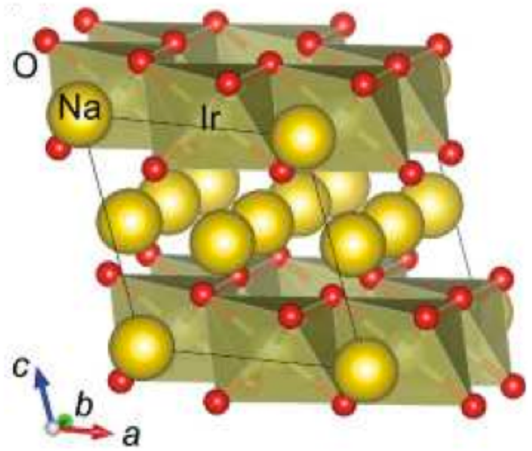
honeycomb Li_2IrO_3



Method	CASSCF+SOC	MRCI+SOC
B1, ∡(Ir-O-Ir)=95.3°, *d*(Ir-Ir)=2.98 Å (×1)[a] :		
Ψ_2	0.0	0.0
$\Psi_3 = \Phi_3 = (\uparrow\uparrow - \downarrow\downarrow)/\sqrt{2}$	1.6	3.2
Ψ_1	5.4	7.7
$\Psi_S = \Phi_S = (\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$	25.5	24.8
$(J, K, \Gamma_{xy}, \Gamma_{zx} = -\Gamma_{yz})$[b]		(−19.2,−6.0,−1.1,−4.8)
B2, ∡(Ir-O-Ir)=94.7°, *d*(Ir-Ir)=2.98 Å (×2)[c] :		
$\Psi_3 = \Phi_3 = (\uparrow\uparrow - \downarrow\downarrow)/\sqrt{2}$	0.0	0.0
Ψ_2	2.8	3.7
$\Psi_S = \Phi_S = (\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$	5.9	7.1
Ψ_1	5.7	8.4
$(J, K, \Gamma_{xy}, \Gamma_{zx} = -\Gamma_{yz})$[d]		(0.8,−11.6,4.2,−2.0)

Nishimoto, Katukuri, Yushankhai, Stoll, Rößler, Rousochatzakis, Hozoi & JvdB, Nat. Comm. 7, 10273 (2016)

honeycomb Li_2IrO_3

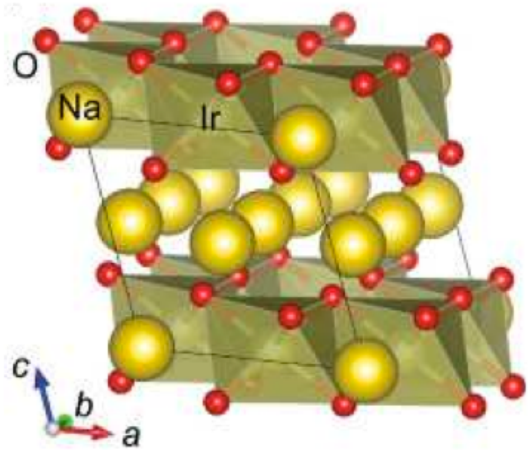


Method	CASSCF+SOC	MRCI+SOC
B1, ∡(Ir-O-Ir)=95.3°, *d*(Ir-Ir)=2.98 Å (×1)[a] :		
Ψ_2	0.0	0.0
$\Psi_3 = \Phi_3 = (\uparrow\uparrow - \downarrow\downarrow)/\sqrt{2}$	1.6	3.2
Ψ_1	5.4	7.7
$\Psi_S = \Phi_S = (\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$	25.5	24.8
$(J, K, \Gamma_{xy}, \Gamma_{zx} = -\Gamma_{yz})$[b]		(–19.2,–6.0,–1.1,–4.8)
B2, ∡(Ir-O-Ir)=94.7°, *d*(Ir-Ir)=2.98 Å (×2)[c] :		
$\Psi_3 = \Phi_3 = (\uparrow\uparrow - \downarrow\downarrow)/\sqrt{2}$	0.0	0.0
Ψ_2	2.8	3.7
$\Psi_S = \Phi_S = (\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$	5.9	7.1
Ψ_1	5.7	8.4
$(J, K, \Gamma_{xy}, \Gamma_{zx} = -\Gamma_{yz})$[d]		(0.8,–11.6,4.2,–2.0)

- ***bond with large FM J***
- ***bond with substantial FM K***

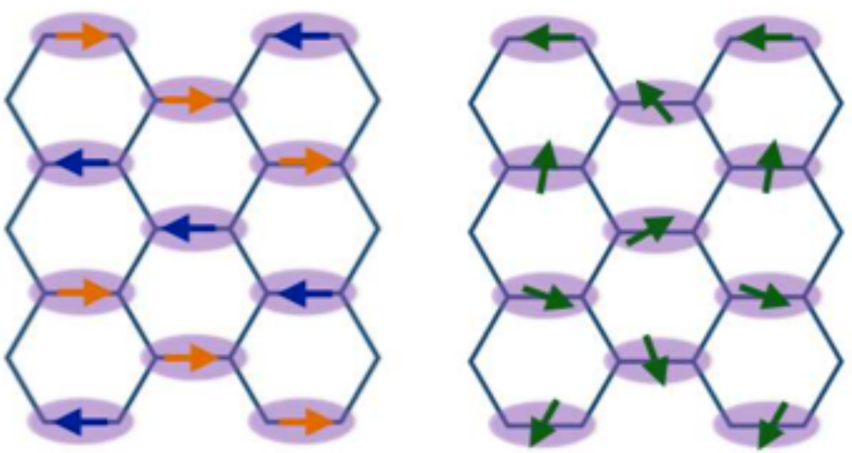
Nishimoto, Katukuri, Yushankhai, Stoll, Rößler, Rousochatzakis, Hozoi & JvdB, Nat. Comm. 7, 10273 (2016)

honeycomb Li_2IrO_3



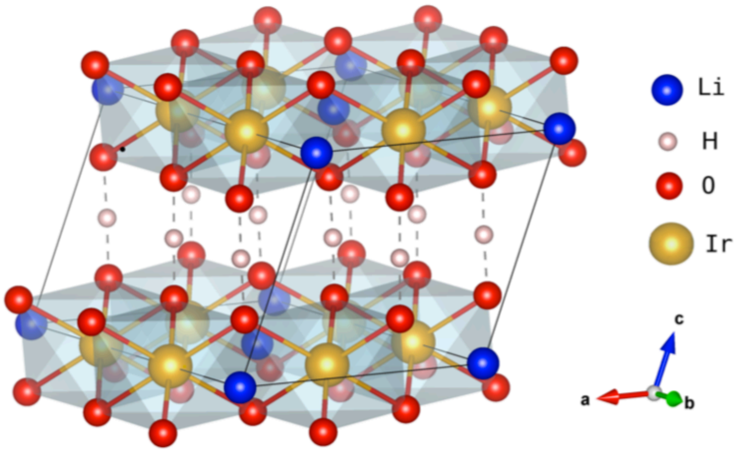
Method	CASSCF+SOC	MRCI+SOC
B1, $\measuredangle$(Ir-O-Ir)=95.3°, *d*(Ir-Ir)=2.98 Å (×1)[a] :		
Ψ_2	0.0	0.0
$\Psi_3 = \Phi_3 = (\uparrow\uparrow - \downarrow\downarrow)/\sqrt{2}$	1.6	3.2
Ψ_1	5.4	7.7
$\Psi_S = \Phi_S = (\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$	25.5	24.8
$(J, K, \Gamma_{xy}, \Gamma_{zx} = -\Gamma_{yz})$[b]		(−19.2,−6.0,−1.1,−4.8)
B2, $\measuredangle$(Ir-O-Ir)=94.7°, *d*(Ir-Ir)=2.98 Å (×2)[c] :		
$\Psi_3 = \Phi_3 = (\uparrow\uparrow - \downarrow\downarrow)/\sqrt{2}$	0.0	0.0
Ψ_2	2.8	3.7
$\Psi_S = \Phi_S = (\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$	5.9	7.1
Ψ_1	5.7	8.4
$(J, K, \Gamma_{xy}, \Gamma_{zx} = -\Gamma_{yz})$[d]		(0.8,−11.6,4.2,−2.0)

- ***bond with large FM J***
- ***bond with substantial FM K***
- ***triplet dimer formation!***



Nishimoto, Katukuri, Yushankhai, Stoll, Rößler, Rousochatzakis, Hozoi & JvdB, Nat. Comm. 7, 10273 (2016)

honeycomb $H_3LiIr_2O_6$



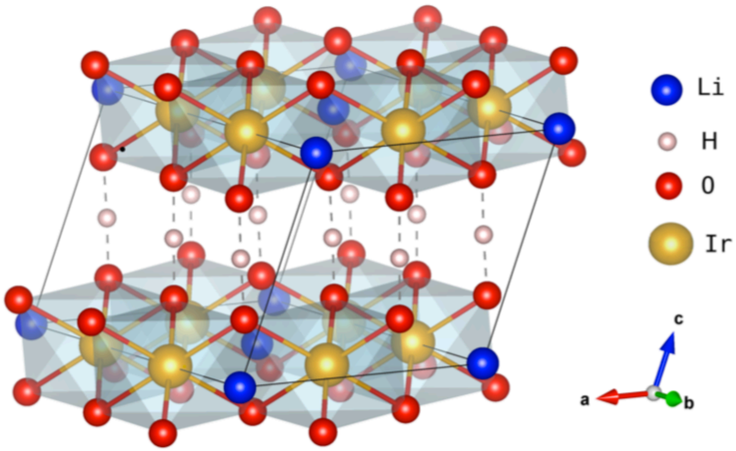
nature
International journal of science

A spin–orbital-entangled quantum liquid on a honeycomb lattice

K. Kitagawa, T. Takayama, Y. Matsumoto, A. Kato, R. Takano, Y. Kishimoto, S. Bette, R. Dinnebier, G. Jackeli & H. Takagi

Nature **554**, 341–345 (15 February 2018) | Received: 18 July 2017

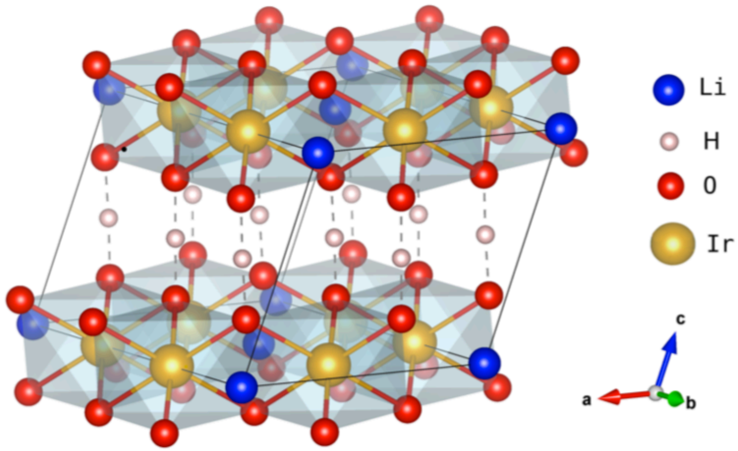
honeycomb $H_3LiIr_2O_6$



Experimental crystal structure					
Bond	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
B2 (3.10Å)	99.8°	**−12.0**	**1.8**	−0.2	−3.2
B1 (3.05Å)	99.0°	**−12.6**	**1.5**	−1.8	−0.65

Yadav, Ray, Eldeeb, Nishimoto, Hozoi & JvdB, PRL 121, 197203 (2018)

honeycomb $H_3LiIr_2O_6$

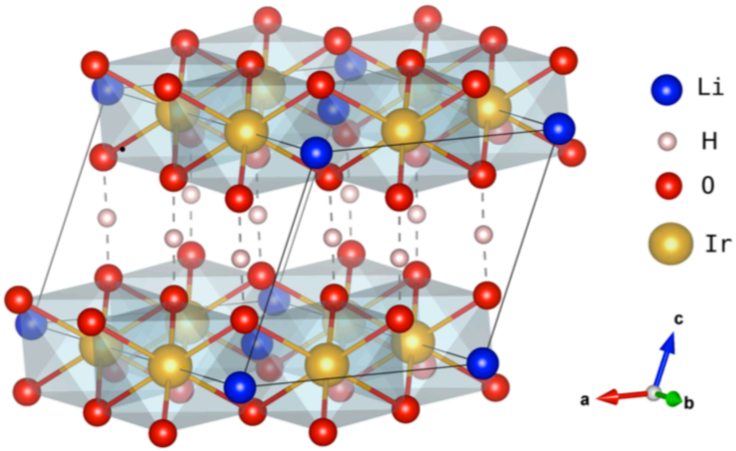


Bond	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
Experimental crystal structure					
B2 (3.10Å)	99.8°	**−12.0**	**1.8**	−0.2	−3.2
B1 (3.05Å)	99.0°	**−12.6**	**1.5**	−1.8	−0.65

- ***FM K, weak AFM J, large K/J***
- ***weak bond anisotropy***

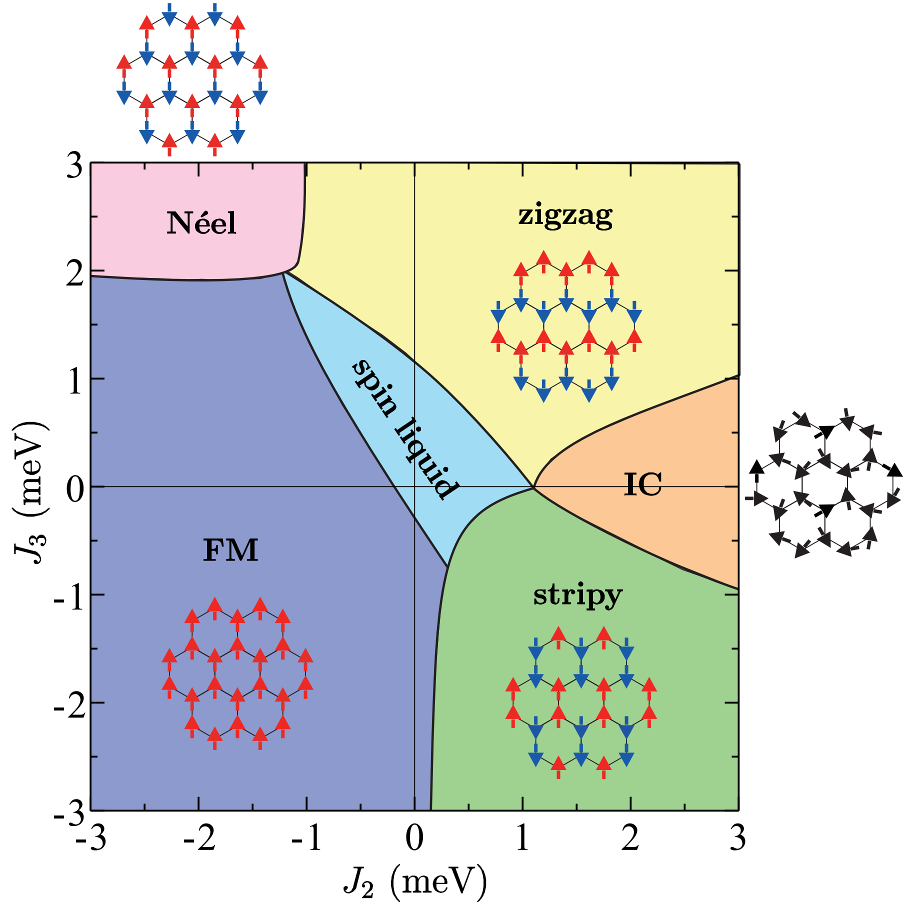
Yadav, Ray, Eldeeb, Nishimoto, Hozoi & JvdB, PRL 121, 197203 (2018)

honeycomb $H_3LiIr_2O_6$



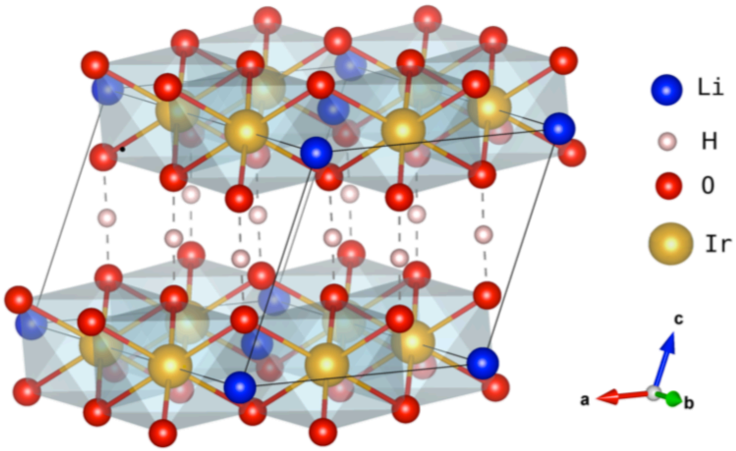
Experimental crystal structure					
Bond	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
B2 (3.10Å)	99.8°	**−12.0**	**1.8**	−0.2	−3.2
B1 (3.05Å)	99.0°	**−12.6**	**1.5**	−1.8	−0.65

- ***FM K, weak AFM J, large K/J***
- ***weak bond anisotropy***



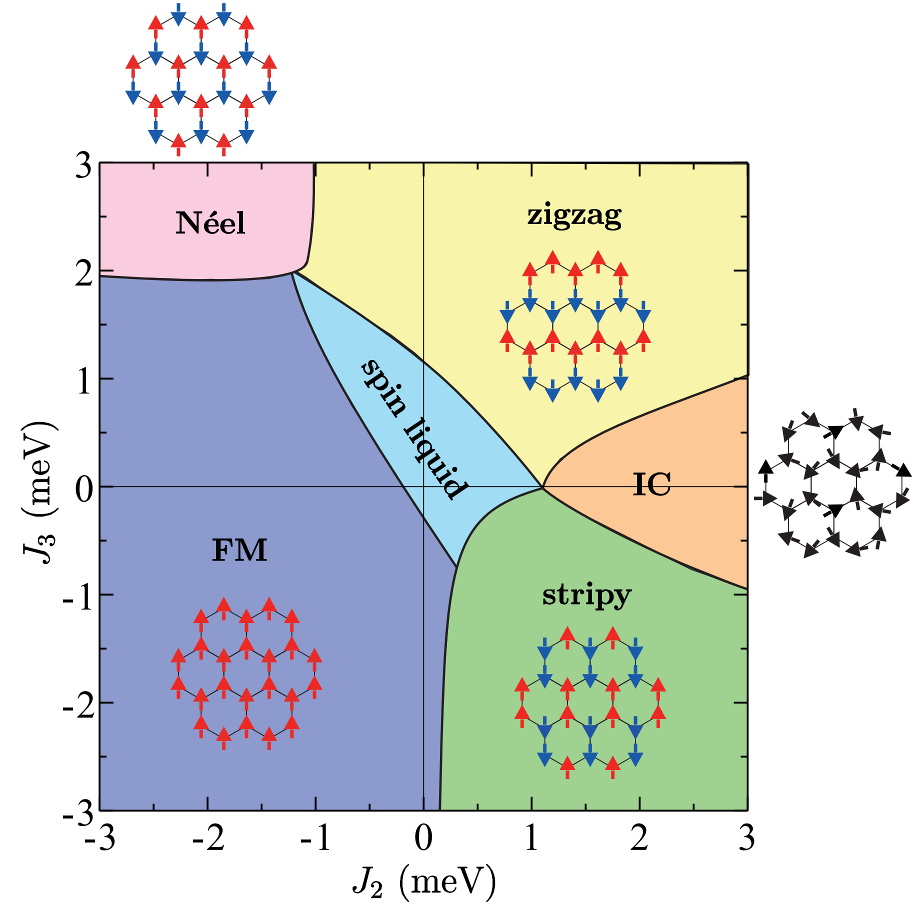
Yadav, Ray, Eldeeb, Nishimoto, Hozoi & JvdB, PRL 121, 197203 (2018)

honeycomb $H_3LiIr_2O_6$



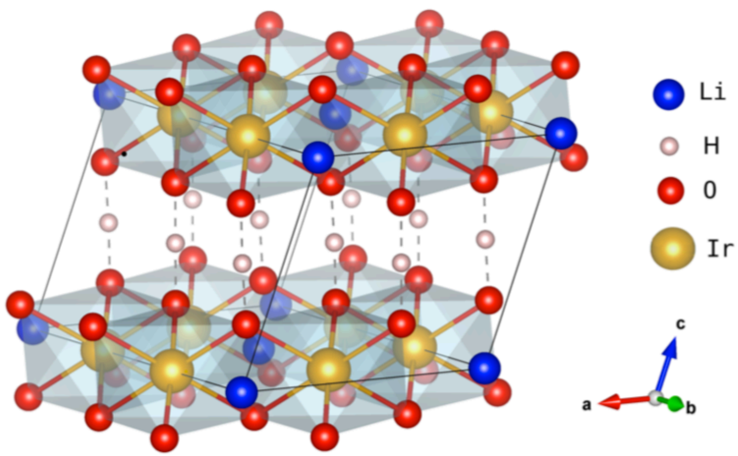
Experimental crystal structure					
Bond	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
B2 (3.10Å)	99.8°	**−12.0**	**1.8**	−0.2	−3.2
B1 (3.05Å)	99.0°	**−12.6**	**1.5**	−1.8	−0.65

- ***FM K, weak AFM J, large K/J***
- ***weak bond anisotropy***
- ***why no LRO by J_2 and J_3?***
- ***why K smaller than Na_2IrO_3 ?***



Yadav, Ray, Eldeeb, Nishimoto, Hozoi & JvdB, PRL 121, 197203 (2018)

honeycomb $H_3LiIr_2O_6$



Experimental crystal structure					
Bond	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
B2 (3.10Å)	99.8°	**−12.0**	**1.8**	−0.2	−3.2
B1 (3.05Å)	99.0°	**−12.6**	**1.5**	−1.8	−0.65

Remove H from Ir-O-Ir links

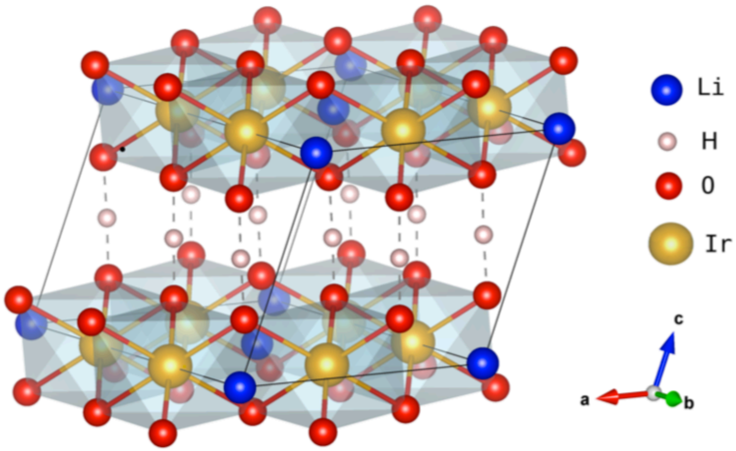
& smear out the + charge

Bond	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
B2 (3.10Å)	**−38.1**	5.9	5.0	−11.1
B1 (3.05Å)	**−40.0**	4.6	7.9	−14.0

Yadav, Ray, Eldeeb, Nishimoto, Hozoi & JvdB, PRL 121, 197203 (2018)

Yadav, Eldeeb, Ray, Aswartham, Sturza, Nishimoto, JvdB & Hozoi, Chemical Science 10,1866 (2019)

honeycomb $H_3LiIr_2O_6$



Experimental crystal structure					
Bond	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
B2 (3.10Å)	99.8°	**−12.0**	**1.8**	−0.2	−3.2
B1 (3.05Å)	99.0°	**−12.6**	**1.5**	−1.8	−0.65

Remove H from Ir-O-Ir links

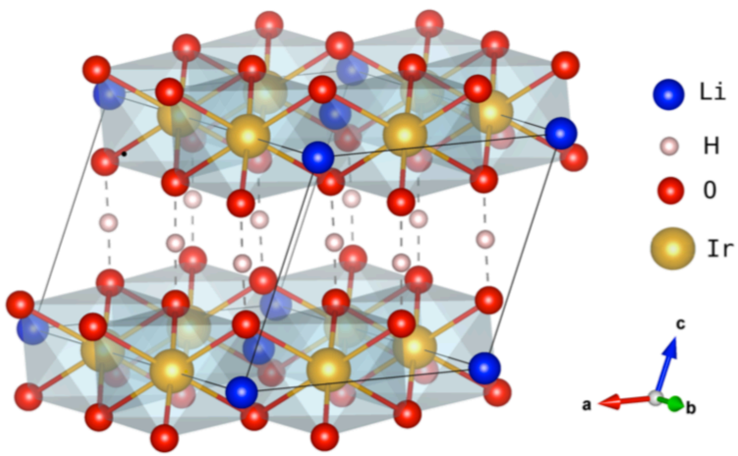
& smear out the + charge

Bond	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
B2 (3.10Å)	**−38.1**	5.9	5.0	−11.1
B1 (3.05Å)	**−40.0**	4.6	7.9	−14.0

Yadav, Ray, Eldeeb, Nishimoto, Hozoi & JvdB, PRL 121, 197203 (2018)

Yadav, Eldeeb, Ray, Aswartham, Sturza, Nishimoto, JvdB & Hozoi, Chemical Science 10,1866 (2019)

honeycomb $H_3LiIr_2O_6$



Experimental crystal structure					
Bond	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
B2 (3.10Å)	99.8°	**−12.0**	**1.8**	−0.2	−3.2
B1 (3.05Å)	99.0°	**−12.6**	**1.5**	−1.8	−0.65

Remove H from Ir-O-Ir links

& smear out the + charge

Bond	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
B2 (3.10Å)	**−38.1**	5.9	5.0	−11.1
B1 (3.05Å)	**−40.0**	4.6	7.9	−14.0

- *H polarizes oxygen orbital relevant for superexchange*
- *very strong effect of hydrogen disorder - affects QSL?*

Yadav, Ray, Eldeeb, Nishimoto, Hozoi & JvdB, PRL 121, 197203 (2018)

Yadav, Eldeeb, Ray, Aswartham, Sturza, Nishimoto, JvdB & Hozoi, Chemical Science 10,1866 (2019)

honeycomb K_2IrO_3

- *C_3 symmetry*
- *Defect structure $K_xIr_yO_2$*
- *Magnetic disorder above 2K*

Johnson, Broeders, Mehlawat, Li, Singh, Valenti, Coldea arXiv:1908.04584 (2019)

Mehlawat & Singh, arXiv:1908.08475 (2019)

honeycomb K_2IrO_3

- *C_3 symmetry*
- *Defect structure $K_xIr_yO_2$*
- *Magnetic disorder above 2K*

Johnson, Broeders, Mehlawat, Li, Singh, Valenti, Coldea arXiv:1908.04584 (2019)

Mehlawat & Singh, arXiv:1908.08475 (2019)

A_2IrO_3	$\angle$Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
A = K	95.0°(×3)	−6.3	1.3	**5.2**	**−8.9**
A = Na	99.5°(×1)	**−20.8**	**5.2**	−0.7	−0.8
	98.0°(×2)	**−15.6**	**2.2**	−1.1	0.8

Yadav, Nishimoto, Richter Jvdb, Ray, preprint (2019)

honeycomb K_2IrO_3

- *C_3 symmetry*
- *Defect structure $K_xIr_yO_2$*
- *Magnetic disorder above 2K*

Johnson, Broeders, Mehlawat, Li, Singh, Valenti, Coldea arXiv:1908.04584 (2019)

Mehlawat & Singh, arXiv:1908.08475 (2019)

A_2IrO_3	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
A = K	95.0°(×3)	−6.3	1.3	**5.2**	**−8.9**
A = Na	99.5°(×1)	**−20.8**	**5.2**	−0.7	−0.8
	98.0°(×2)	**−15.6**	**2.2**	−1.1	0.8

Yadav, Nishimoto, Richter Jvdb, Ray, preprint (2019)

honeycomb K_2IrO_3

- ***C_3 symmetry***
- ***Defect structure $K_xIr_yO_2$***
- ***Magnetic disorder above 2K***

Johnson, Broeders, Mehlawat, Li, Singh, Valenti, Coldea arXiv:1908.04584 (2019)

Mehlawat & Singh, arXiv:1908.08475 (2019)

A_2IrO_3	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
A = K	95.0°(×3)	−6.3	1.3	**5.2**	**−8.9**
A = Na	99.5°(×1)	**−20.8**	**5.2**	−0.7	−0.8
	98.0°(×2)	**−15.6**	**2.2**	−1.1	0.8

- ***MRCI: large off-diagonal magnetic interactions → C_3***

Yadav, Nishimoto, Richter Jvdb, Ray, preprint (2019)

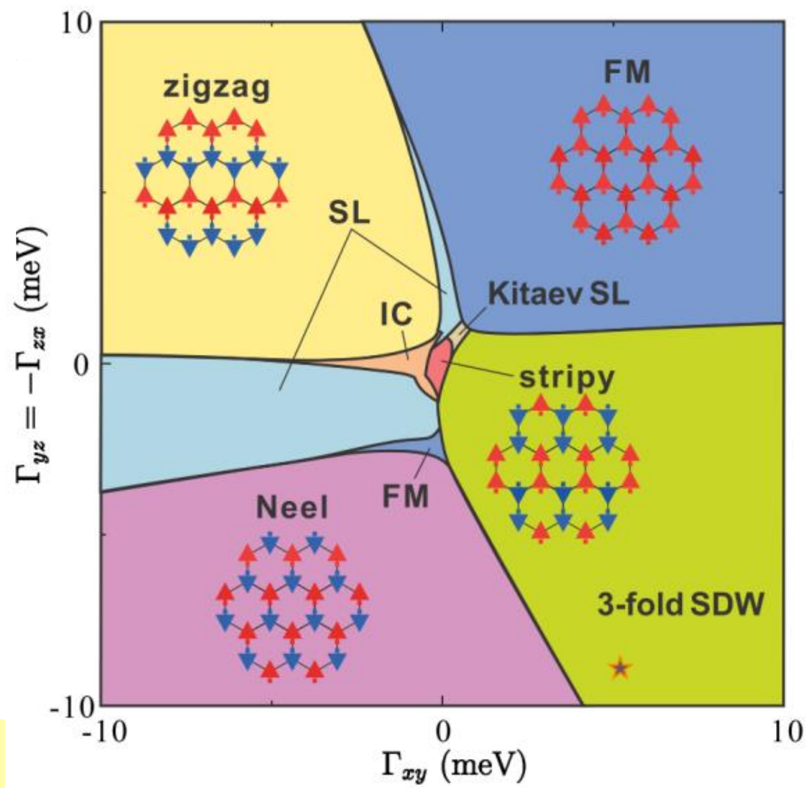
honeycomb K_2IrO_3

- *C_3 symmetry*
- *Defect structure $K_xIr_yO_2$*
- *Magnetic disorder above 2K*

Johnson, Broeders, Mehlawat, Li, Singh, Valenti, Coldea arXiv:1908.04584 (2019)

Mehlawat & Singh, arXiv:1908.08475 (2019)

A_2IrO_3	∠Ir-O-Ir	K	J	Γ_{xy}	$\Gamma_{yz} = -\Gamma_{zx}$
A = K	95.0°(×3)	−6.3	1.3	**5.2**	**−8.9**
A = Na	99.5°(×1)	**−20.8**	**5.2**	−0.7	−0.8
	98.0°(×2)	**−15.6**	**2.2**	−1.1	0.8



- *MRCI: large off-diagonal magnetic interactions → C_3*
- *Alternative route to stabilize (Kitaev) QSL*

Yadav, Nishimoto, Richter Jvdb, Ray, preprint (2019)

Electronic & magnetic structure of α-$RuCl_3$

Honeycomb $RuCl_3$

Quantum chemistry calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha \neq \beta} \Gamma_{\alpha\beta}(\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

Structure	∠Ru-Cl-Ru	K	J	Γ_{xy}	$\Gamma_{zx}=-\Gamma_{yz}$
$C2/m$ [30]	94°	−5.6	1.2	−1.2	−0.7
$C2/m$ [29]					
Link 1 (×2)	94°	−5.3	1.2	−1.1	−0.7
Link 2 (×1)	93°	−4.8	−0.3	−1.5	−0.7
$P3_112$ [28]	89°	−1.2	−0.5	−1.0	−0.4

Sears, Songvilay, Plumb, Clancy, Qiu, Zhao, Parshall & Y-J Kim, PRB 91, 144420 (2015)

Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Honeycomb $RuCl_3$

Quantum chemistry calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha\neq\beta} \Gamma_{\alpha\beta}(\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

Structure	∠Ru-Cl-Ru	K	J	Γ_{xy}	$\Gamma_{zx}=-\Gamma_{yz}$
$C2/m$ [30]	94°	−5.6	1.2	−1.2	−0.7
$C2/m$ [29]					
Link 1 (×2)	94°	−5.3	1.2	−1.1	−0.7
Link 2 (×1)	93°	−4.8	−0.3	−1.5	−0.7
$P3_112$ [28]	89°	−1.2	−0.5	−1.0	−0.4

K large FM, J small AFM

|K/J| ~ 5

Winter, Tsirlin, Daghofer, JvdB, Singh, Gegenwart & Valenti, JPCM 29, 493002 (2017)

Sears, Songvilay, Plumb, Clancy, Qiu, Zhao, Parshall & Y-J Kim, PRB 91, 144420 (2015)

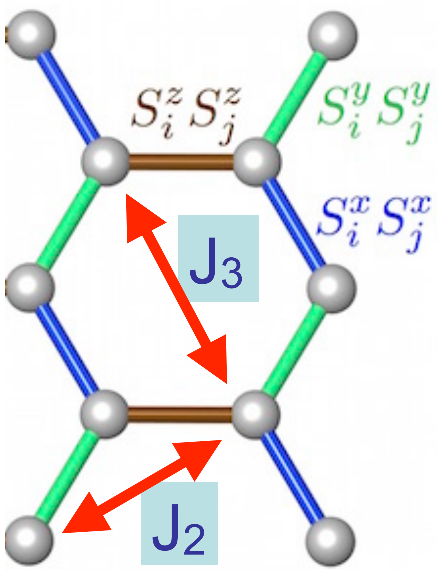
Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Honeycomb $RuCl_3$

Exact diagonalization calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha \neq \beta} \Gamma_{\alpha\beta} (\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

+ longer range Heisenberg J_2 and J_3

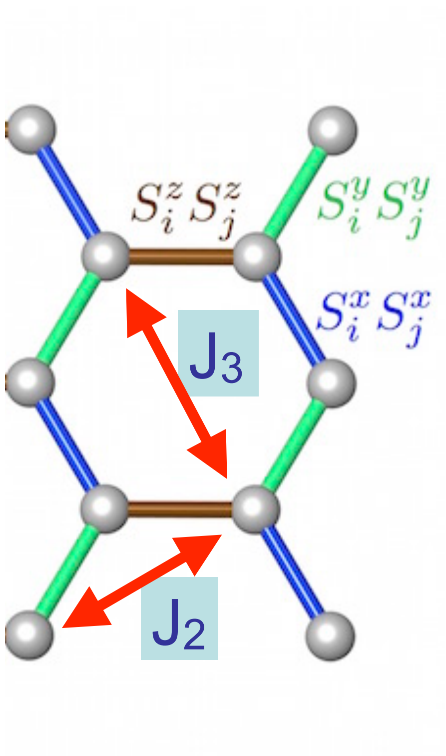


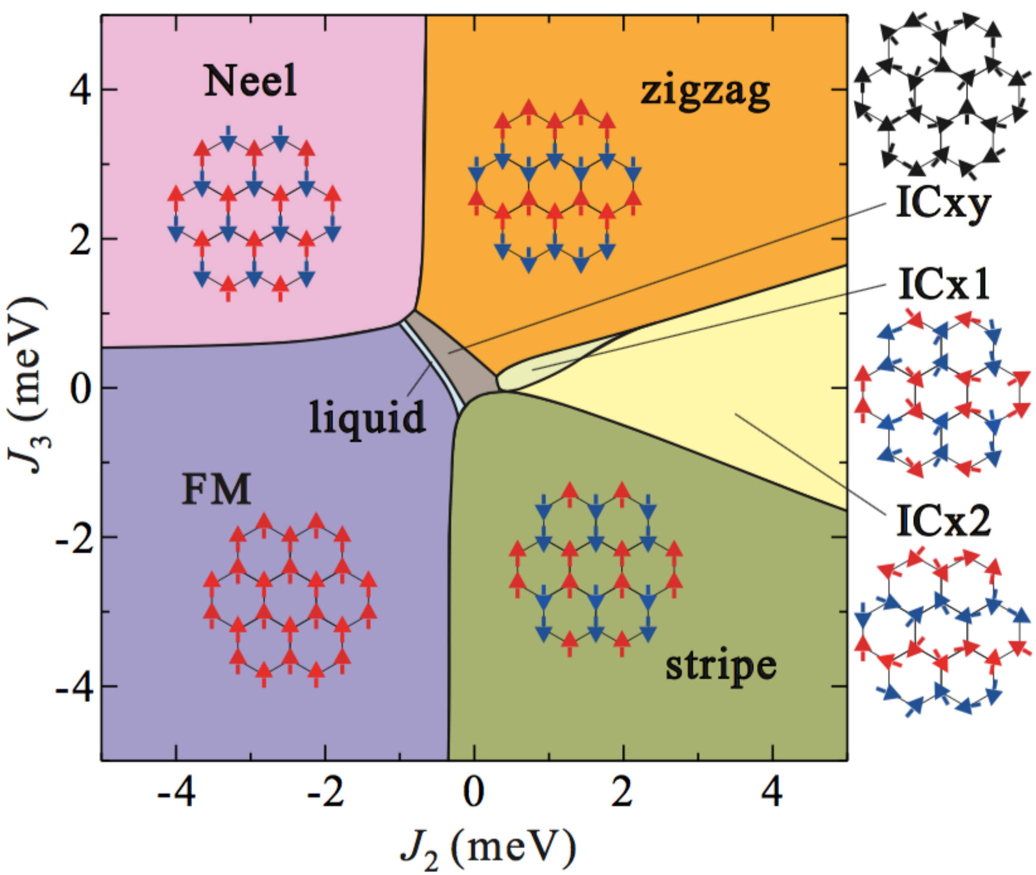
Honeycomb RuCl$_3$

Exact diagonalization calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha \neq \beta} \Gamma_{\alpha\beta}(\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

+ longer range Heisenberg J_2 and J_3





zig-zag order driven by J_2 & J_3

Local electronic structure of α-$RuCl_3$

Quantum chemistry calculations

Ru^{3+} $4d^5$ splittings	CASSCF	CASSCF +SOC	MRCI	MRCI +SOC
$^2T_2$ (t_{2g}^5)	0	0	0	0
	0.066	0.193	0.067	0.195
	0.069	0.232	0.071	0.234
$^4T_1$ $(t_{2g}^4 e_g^1)$	1.08	1.25	1.28	1.33
	1.12	|	1.30	|
	1.13	1.37	1.31	1.48
$^4T_2$ $(t_{2g}^4 e_g^1)$	1.76	1.90	1.97	2.09
	1.81	|	2.01	|
	1.83	1.98	2.03	2.17
$^6A_1$ $(t_{2g}^3 e_g^2)$	1.01	1.09 (×6)	1.51	1.74 (×6)

$$g_{xx} = g_{yy} = 2.51; g_{zz} = 1.09$$

Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Local electronic structure of α-$RuCl_3$

Quantum chemistry calculations

Ru^{3+} $4d^5$ splittings	CASSCF	CASSCF +SOC	MRCI	MRCI +SOC
$^2T_2$ (t_{2g}^5)	0	0	0	0
	0.066	0.193	0.067	0.195
	0.069	0.232	0.071	0.234
$^4T_1$ $(t_{2g}^4 e_g^1)$	1.08	1.25	1.28	1.33
	1.12	|	1.30	|
	1.13	1.37	1.31	1.48
$^4T_2$ $(t_{2g}^4 e_g^1)$	1.76	1.90	1.97	2.09
	1.81	|	2.01	|
	1.83	1.98	2.03	2.17
$^6A_1$ $(t_{2g}^3 e_g^2)$	1.01	1.09 (×6)	1.51	1.74 (×6)

$$g_{xx} = g_{yy} = 2.51; g_{zz} = 1.09$$

Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Local electronic structure of α-$RuCl_3$

Quantum chemistry calculations

Ru^{3+} $4d^5$ splittings	CASSCF	CASSCF +SOC	MRCI	MRCI +SOC
$^2T_2$ (t_{2g}^5)	0	0	0	0
	0.066	0.193	0.067	0.195
	0.069	0.232	0.071	0.234
$^4T_1$ $(t_{2g}^4 e_g^1)$	1.08	1.25	1.28	1.33
	1.12	|	1.30	|
	1.13	1.37	1.31	1.48
$^4T_2$ $(t_{2g}^4 e_g^1)$	1.76	1.90	1.97	2.09
	1.81	|	2.01	|
	1.83	1.98	2.03	2.17
$^6A_1$ $(t_{2g}^3 e_g^2)$	1.01	1.09 (×6)	1.51	1.74 (×6)

~0.2 eV splitting j=1/2 to j=3/2

$$g_{xx} = g_{yy} = 2.51; g_{zz} = 1.09$$

Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Magnetic nearest neighbor interactions in α-$RuCl_3$

Quantum chemistry calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha\neq\beta} \Gamma_{\alpha\beta}(\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

Sears, Songvilay, Plumb, Clancy, Qiu, Zhao, Parshall & Y-J Kim, PRB 91, 144420 (2015)

Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Magnetic nearest neighbor interactions in α-$RuCl_3$

Quantum chemistry calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha \neq \beta} \Gamma_{\alpha\beta}\left(\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha\right)$$

Structure	∠Ru-Cl-Ru	K	J	Γ_{xy}	Γ_{zx}=$-\Gamma_{yz}$
$C2/m$ [30]	94°	−5.6	1.2	−1.2	−0.7
$C2/m$ [29]					
Link 1 (×2)	94°	−5.3	1.2	−1.1	−0.7
Link 2 (×1)	93°	−4.8	−0.3	−1.5	−0.7
$P3_112$ [28]	89°	−1.2	−0.5	−1.0	−0.4

Sears, Songvilay, Plumb, Clancy, Qiu, Zhao, Parshall & Y-J Kim, PRB 91, 144420 (2015)

Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Magnetic nearest neighbor interactions in α-$RuCl_3$

Quantum chemistry calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K \tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha \neq \beta} \Gamma_{\alpha\beta} (\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

Structure	∠Ru-Cl-Ru	K	J	Γ_{xy}	$\Gamma_{zx}=-\Gamma_{yz}$
$C2/m$ [30]	94°	−5.6	1.2	−1.2	−0.7
$C2/m$ [29]					
Link 1 (×2)	94°	−5.3	1.2	−1.1	−0.7
Link 2 (×1)	93°	−4.8	−0.3	−1.5	−0.7
$P3_112$ [28]	89°	−1.2	−0.5	−1.0	−0.4

K large FM, J small AFM

|K/J| ~ 5

Winter, Tsirlin, Daghofer, JvdB, Singh, Gegenwart & Valenti, JPCM (2018)

Sears, Songvilay, Plumb, Clancy, Qiu, Zhao, Parshall & Y-J Kim, PRB 91, 144420 (2015)

Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Magnetic nearest neighbor interactions in α-$RuCl_3$

Quantum chemistry calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\tilde{S}_i^z\tilde{S}_j^z + \sum_{\alpha\neq\beta}\Gamma_{\alpha\beta}(\tilde{S}_i^\alpha\tilde{S}_j^\beta + \tilde{S}_i^\beta\tilde{S}_j^\alpha)$$

Structure	∠Ru-Cl-Ru	K	J	Γ_{xy}	$\Gamma_{zx}=-\Gamma_{yz}$
$C2/m$ [30]	94°	−5.6	1.2	−1.2	−0.7
$C2/m$ [29]					
Link 1 (×2)	94°	−5.3	1.2	−1.1	−0.7
Link 2 (×1)	93°	−4.8	−0.3	−1.5	−0.7
$P3_112$ [28]	89°	−1.2	−0.5	−1.0	−0.4

K large FM, J small AFM

|K/J| ~ 5

However INS: K AFM

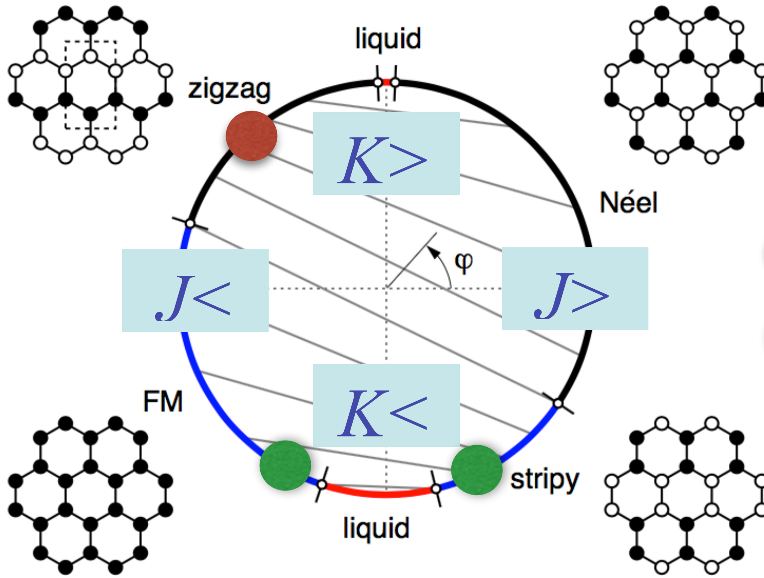
Winter, Tsirlin, Daghofer, JvdB, Singh, Gegenwart & Valenti, JPCM (2018)

Banerjee et al., Nat. Mater. 4604 (2016)

Sears, Songvilay, Plumb, Clancy, Qiu, Zhao, Parshall & Y-J Kim, PRB 91, 144420 (2015)

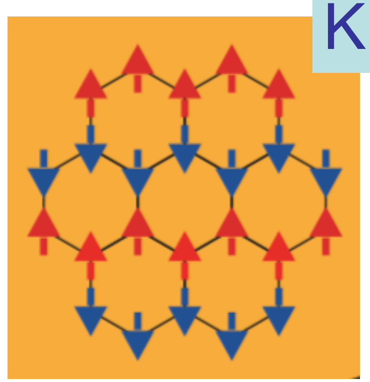
Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Magnetic nearest neighbor interactions in α-$RuCl_3$



Banerjee et al.

Yadav et al.



K large FM, J small AFM

|K/J| ~ 5

However INS: K AFM

Experimentally: zig-zag order below ~8K

Banerjee et al., Nat. Mater. 4604 (2016)

Sears, Songvilay, Plumb, Clancy, Qiu, Zhao, Parshall & Y-J Kim, PRB 91, 144420 (2015)

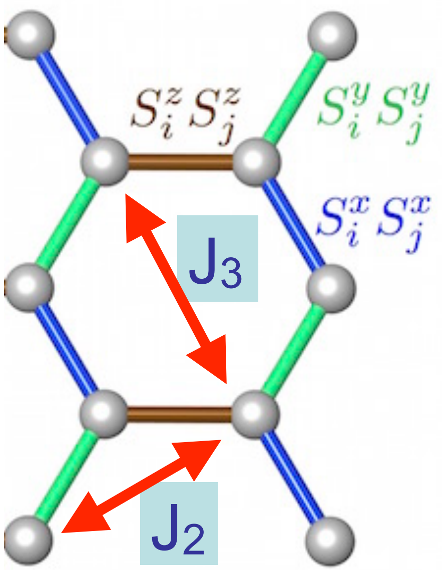
Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Magnetic nearest neighbor interactions in α-$RuCl_3$

Exact diagonalization calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha \neq \beta} \Gamma_{\alpha\beta} (\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

+ longer range Heisenberg J_2 and J_3

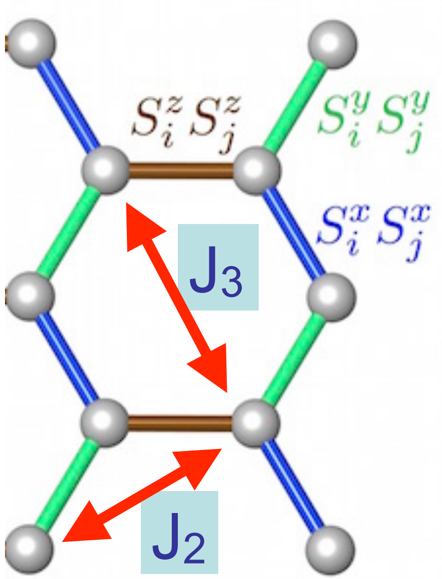


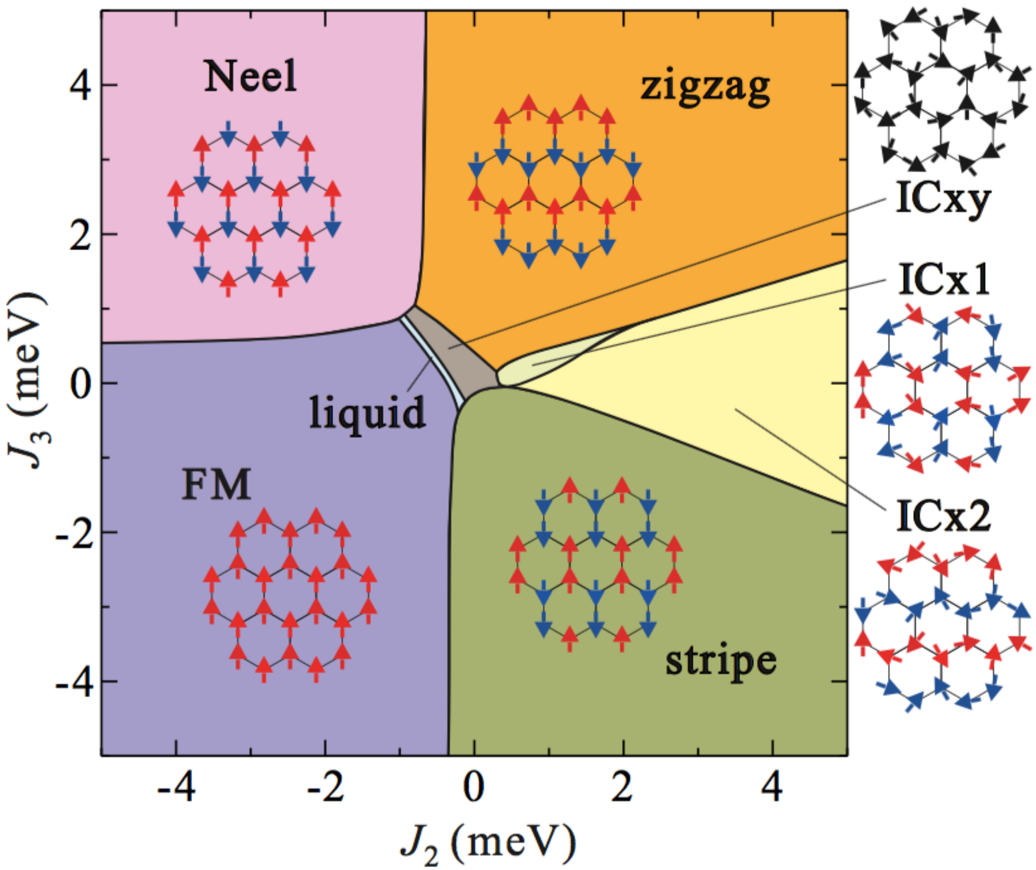
Magnetic nearest neighbor interactions in α-$RuCl_3$

Exact diagonalization calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha\neq\beta} \Gamma_{\alpha\beta}(\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

+ longer range Heisenberg J_2 and J_3





zig-zag order driven by J_2 & J_3

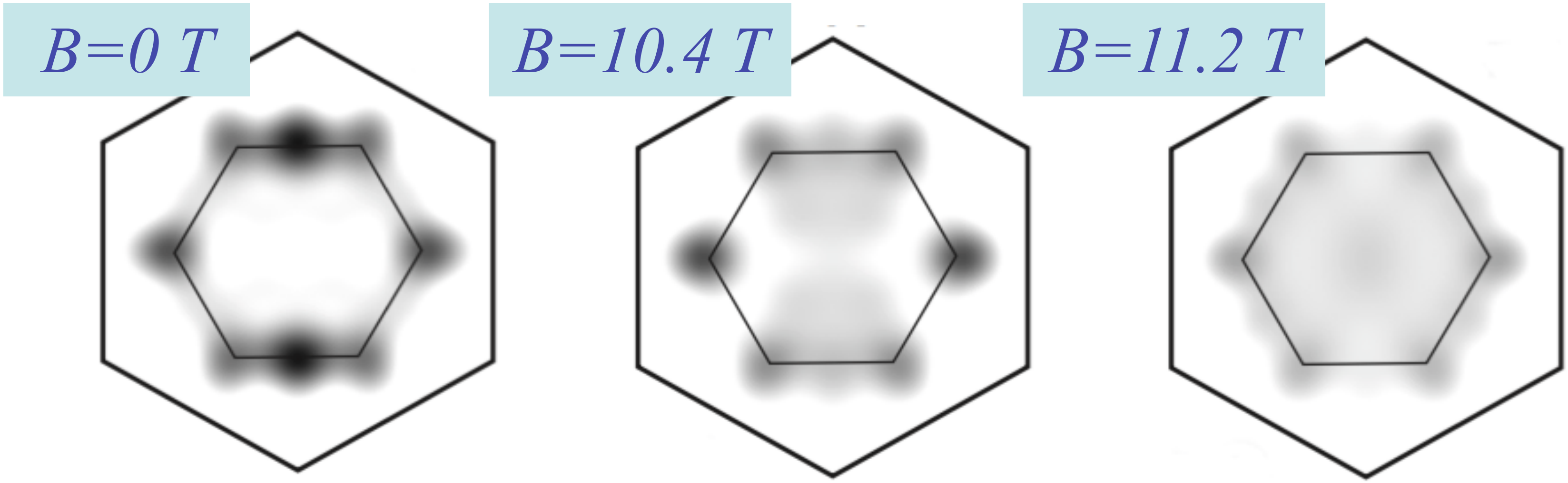
Magnetic nearest neighbor interactions in α-RuCl₃

Exact diagonalization calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha \neq \beta} \Gamma_{\alpha\beta}(\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

+ longer range Heisenberg J_2 and J_3

Static spin structure factor S(Q) from ED



Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Magnetic nearest neighbor interactions in α-$RuCl_3$

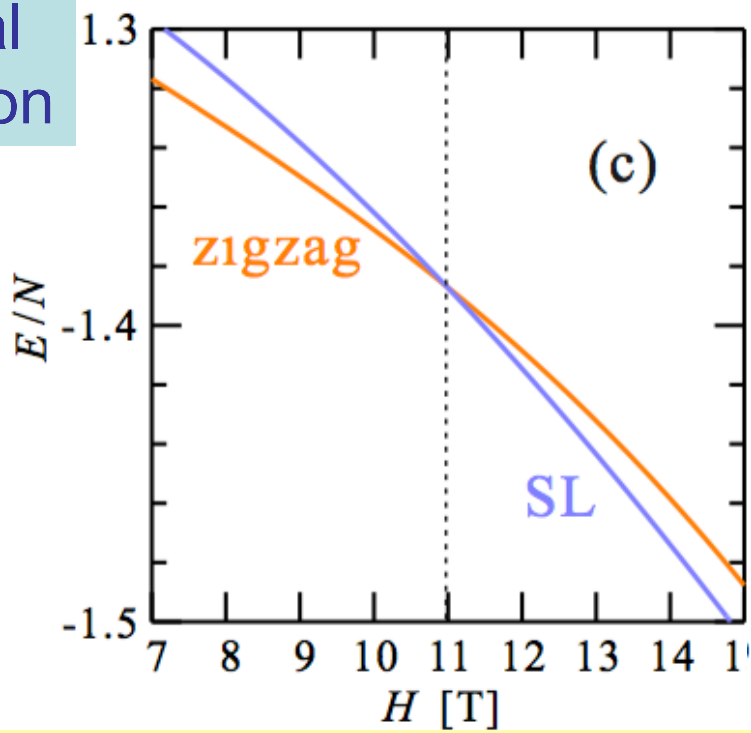
Exact diagonalization calculations

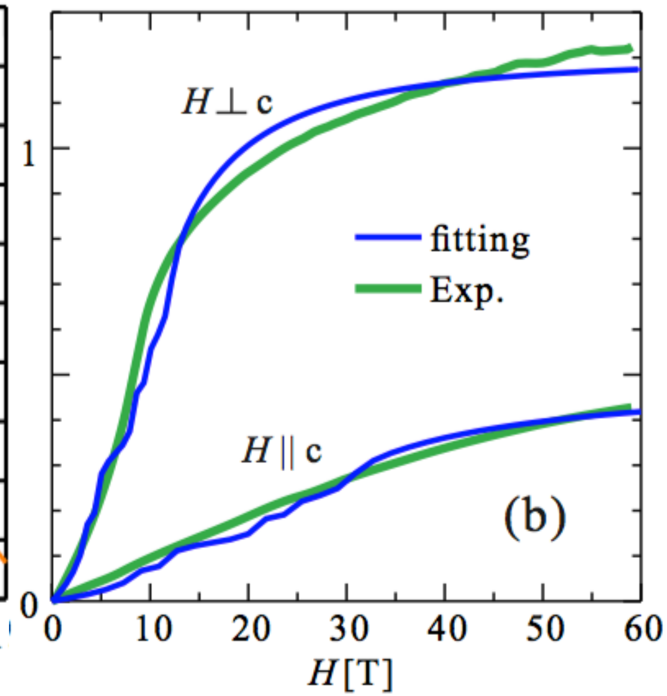
$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\,\tilde{S}_i^z \tilde{S}_j^z + \sum_{\alpha\neq\beta} \Gamma_{\alpha\beta}(\tilde{S}_i^\alpha \tilde{S}_j^\beta + \tilde{S}_i^\beta \tilde{S}_j^\alpha)$$

+ longer range Heisenberg J_2 and J_3

Compare to experimental magnetization

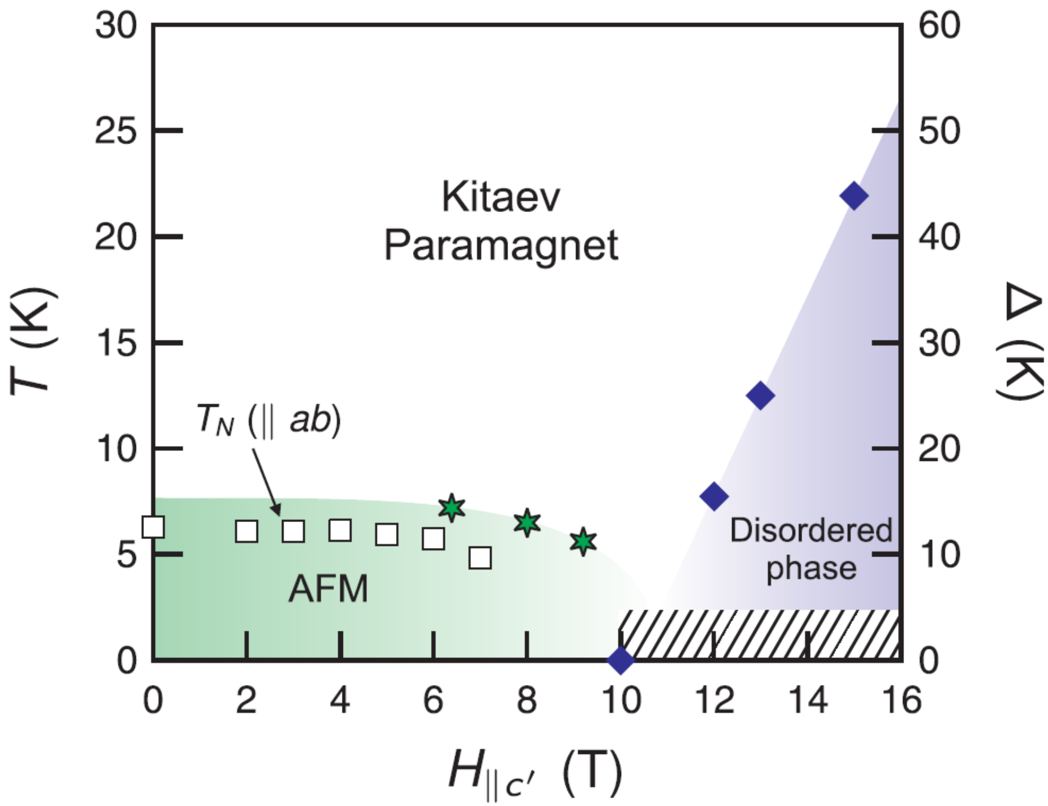
Fit g-factors



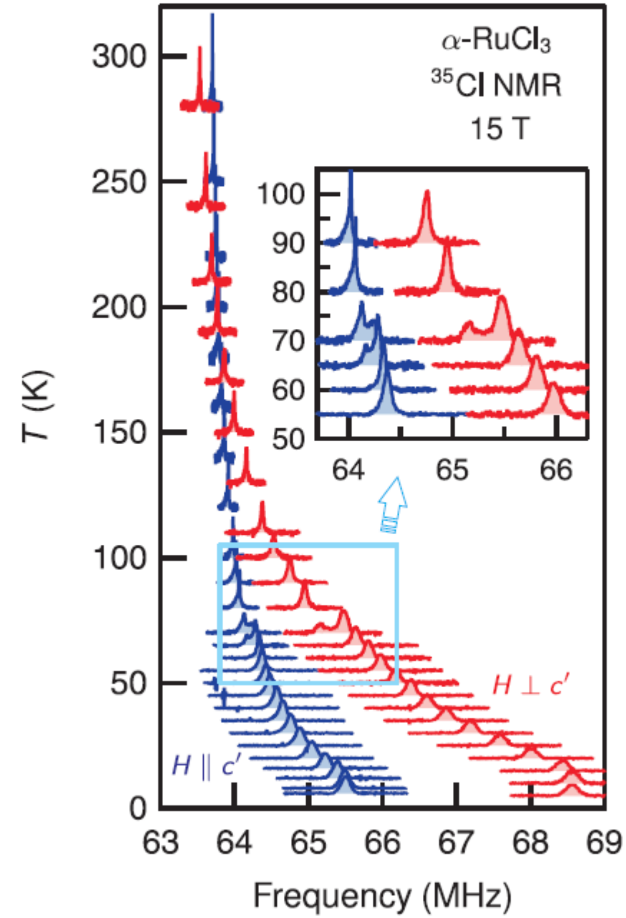


Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

Observation of B-induced spin liquid in $RuCl_3$



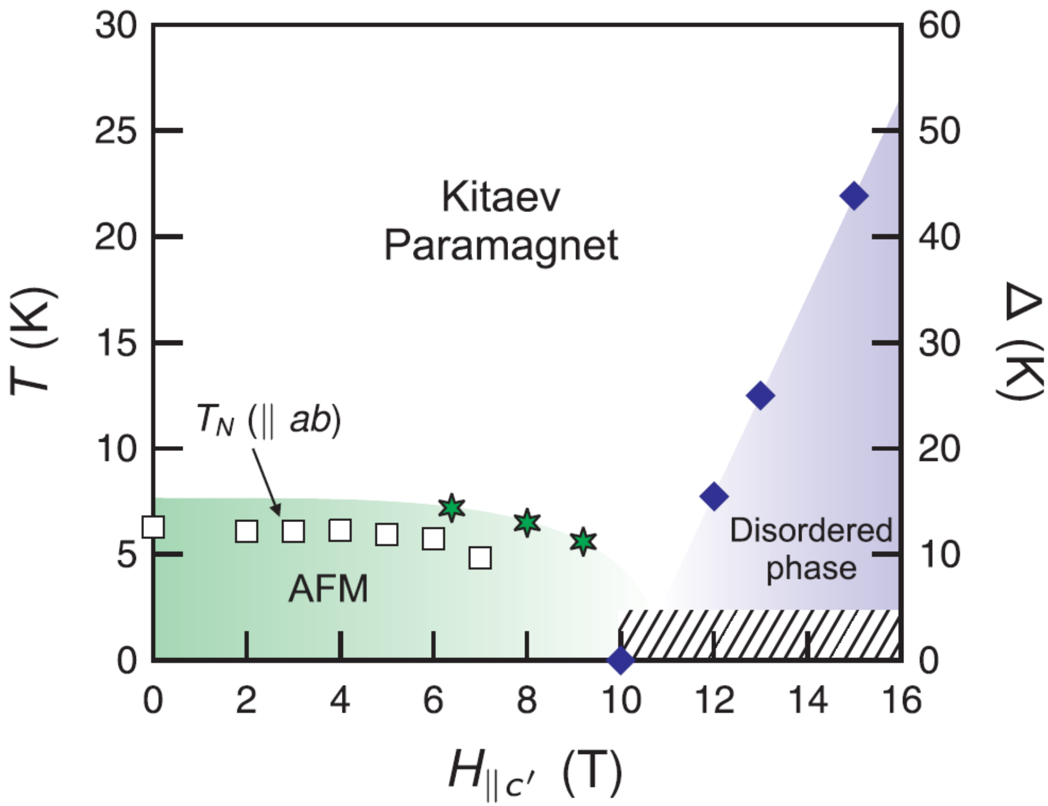
gapped spin liquid

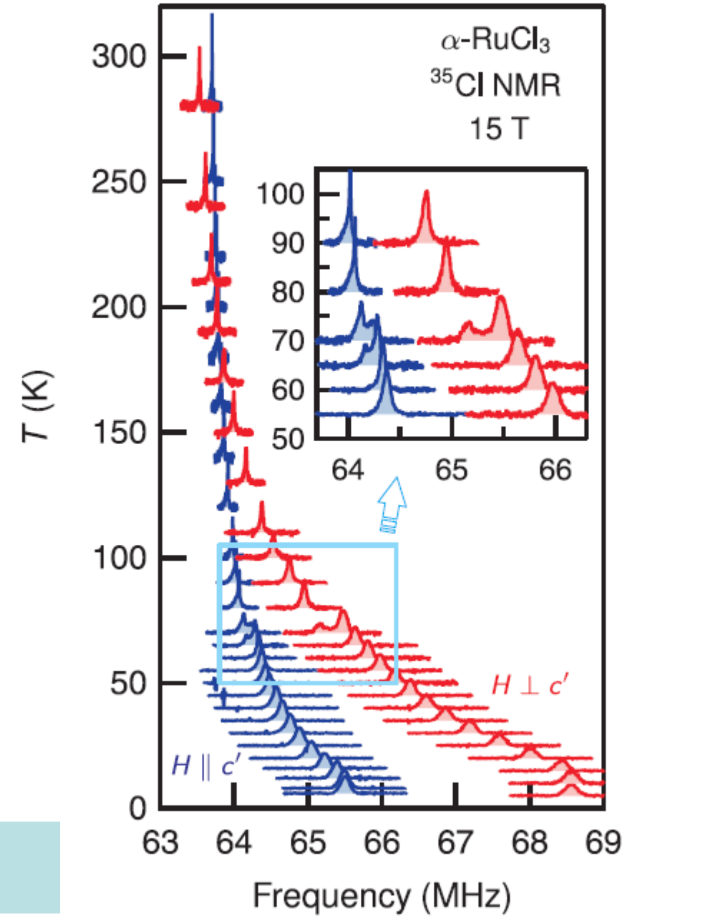


Baek, Do, Choi, Kwon,Wolter, Nishimoto, JvdB & Büchner, PRL 119, 037201 (2017)

Observation of B-induced spin liquid in $RuCl_3$

gapped spin liquid





now also reported by neutron scattering

Banerjee et al., Quantum Mat. 3, 8 (2018)

Baek, Do, Choi, Kwon, Wolter, Nishimoto, JvdB & Büchner, PRL 119, 037201 (2017)

Conclusions

Based on quantum chemistry & cluster ED:

Conclusions

Based on quantum chemistry & cluster ED:

213 honeycomb iridates: ***K ~ -15 meV (ballpark)***

interactions beyond Kitaev

strong bond anisotropies

Conclusions

Based on quantum chemistry & cluster ED:

213 honeycomb iridates: *$K \sim -15$ meV (ballpark)*

interactions beyond Kitaev *strong bond anisotropies*

honeycomb $H_3LiIr_2O_6$ *$K \sim -12$ meV, $|K/J| > 6$*

very strong effect of H disorder on magnetism

Conclusions

Based on quantum chemistry & cluster ED:

213 honeycomb iridates: *$K \sim -15$ meV (ballpark)*

interactions beyond Kitaev *strong bond anisotropies*

honeycomb $H_3LiIr_2O_6$ *$K \sim -12$ meV, $|K/J| > 6$*

very strong effect of H disorder on magnetism

honeycomb K_2IrO_3 *$K \sim -6$ meV, $|K/J| \sim 5$, $|K| \sim |\Gamma|$*

Conclusions

Based on quantum chemistry & cluster ED:

213 honeycomb iridates: *K ~ -15 meV (ballpark)*

interactions beyond Kitaev *strong bond anisotropies*

honeycomb $H_3LiIr_2O_6$ *K ~ -12 meV, |K/J| > 6*

very strong effect of H disorder on magnetism

honeycomb K_2IrO_3 *K ~ -6 meV, |K/J|~5, |K|~|Γ|*

ruthenium trichloride: *K ~ -5 meV, |K/J| ~ 5*

residual interactions weak, anisotropy weak

in-plane field above B=~8 T: gapped spin liquid