Higher spin Kitaev model

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Topological Quantum Matter:
From Fantasy to Reality,
KITP, October 2, 2019

Quantum Spin liquids

: Long Range Entanglement
with fractional excitations

- S=1/2 Kitaev Spin liquids
- S=1/2 Toric code

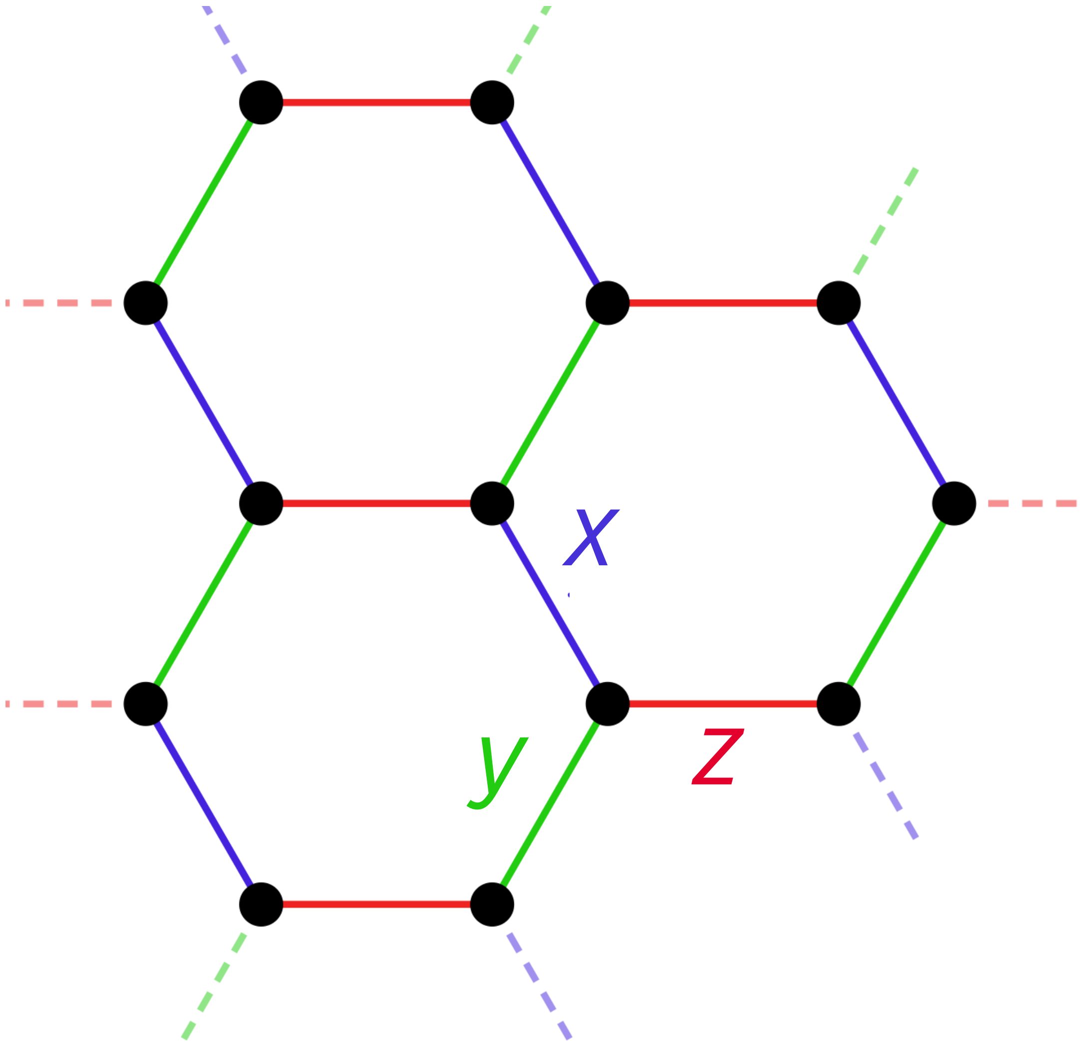
Kitaev spin liquid

Kitaev Exchange

$$K \sum_{\langle ij\rangle \in \gamma} S_i^\gamma S_j^\gamma$$

where $\gamma = x, y, z$

bond-dep. interaction



Exactly solvable: Z2 spin liquid ground state

A. Kitaev, Annals of Physics 321, 2 (2006):
Anyones in exactly solved model and beyond

Outline

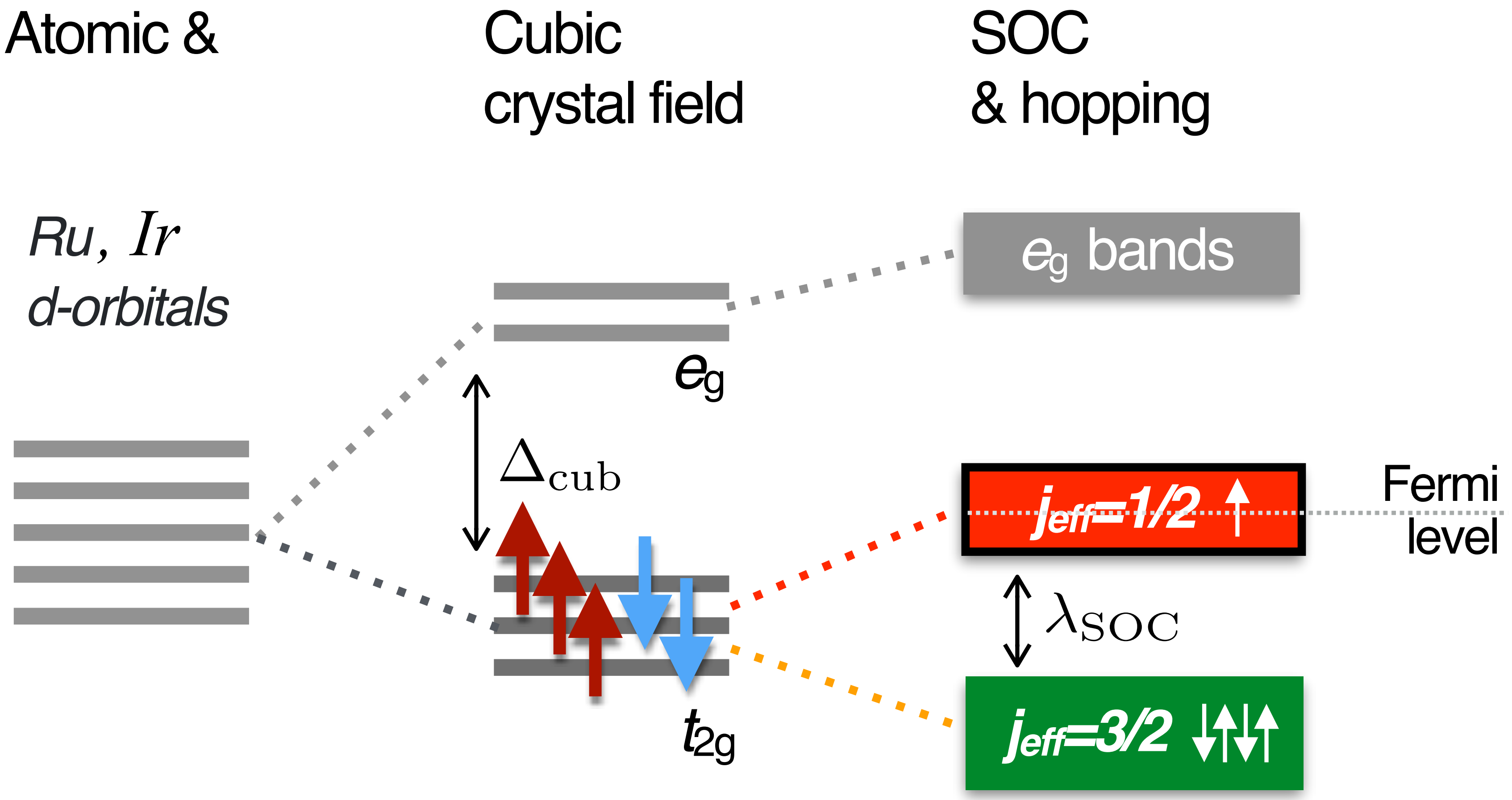
Review on S=1/2 bond-dep. interactions

Derivation of Kitaev interaction for S=1

S=1 Field induced spin liquid states?

Review: S=1/2 Kitaev materials

J_{eff}=1/2 — spin-orbit coupling (SOC)

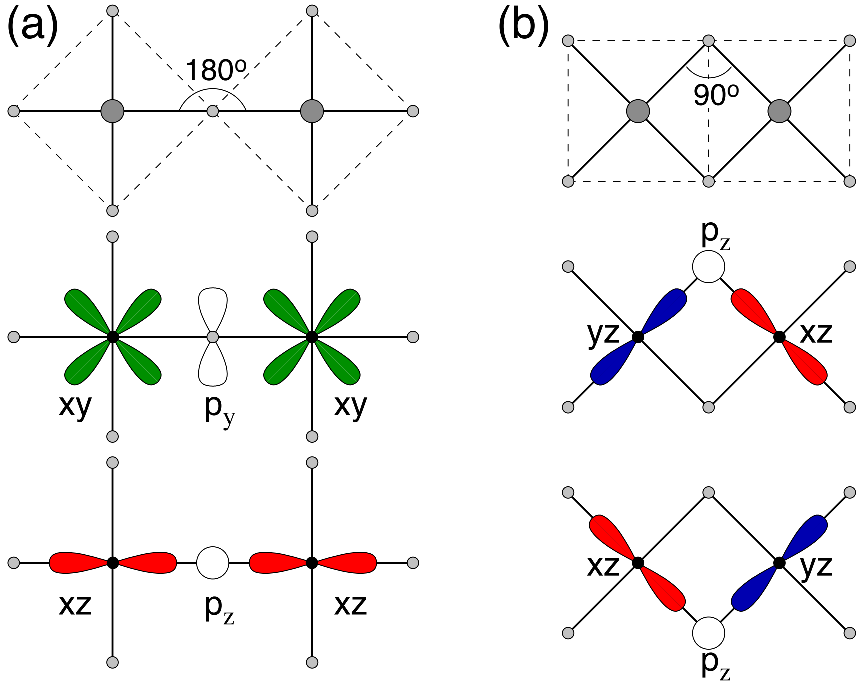


d^5 -- half filled Jeff=1/2 bands

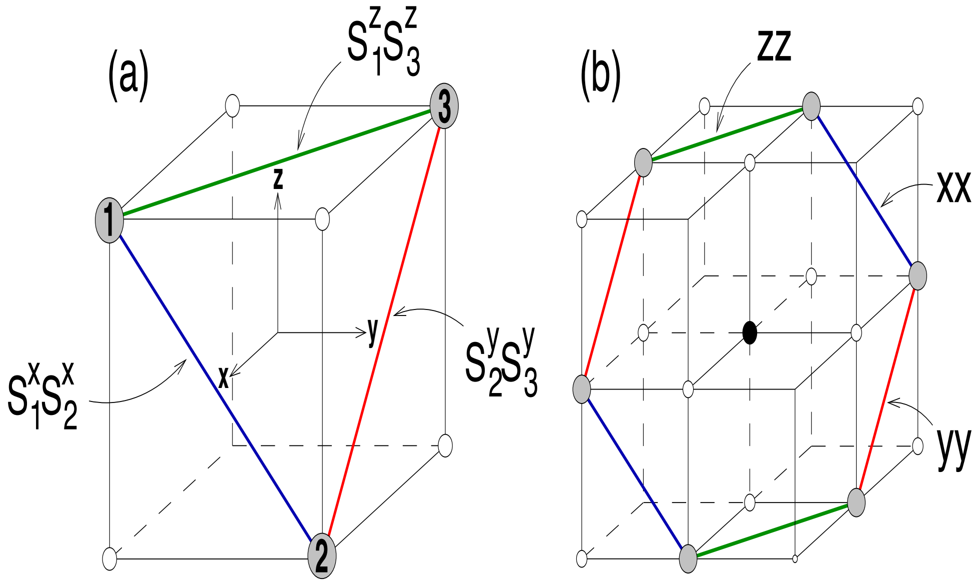
Sr2IrO4: Mott insulator, BJ Kim... W. Noh, PRL (2008);
B.J. Kim...H. Takagi, Science (2009)

Compass model

Jackeli and Khaliullin, Phys. Rev. Lett. 102, 017205 (2009)



edge-shared octahedra



strong SOC in t2g states:

Kitaev-Heisenberg model

$$H_{ij}^{\gamma} = -K S_i^{\gamma} S_j^{\gamma} + J\mathbf{S}_i \cdot \mathbf{S}_j \quad \text{where} \quad \mathrm{K} = \frac{8 J_H t_0^2}{3U^2}$$

Material candidates: honeycomb Iridates (5d)

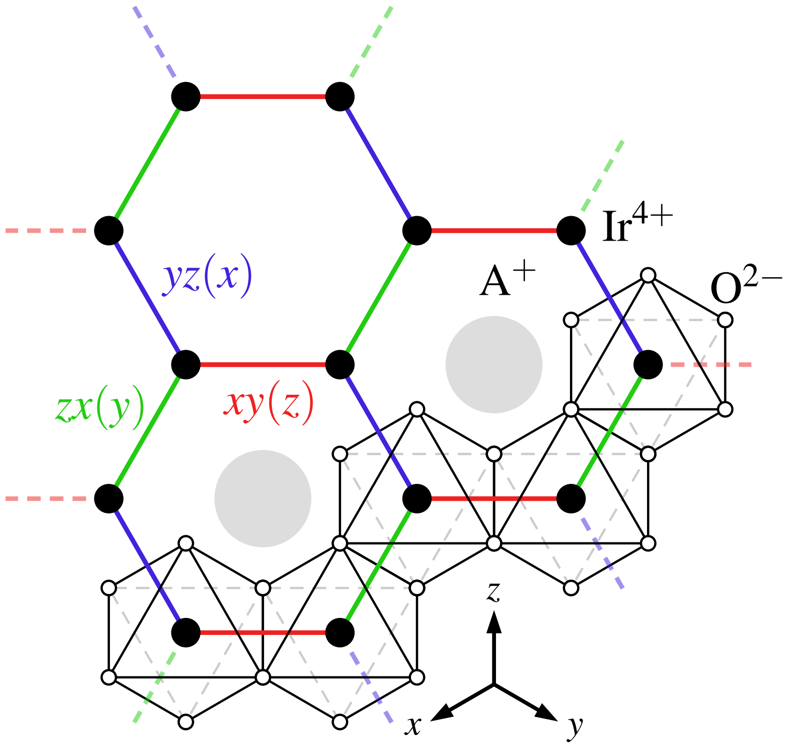
Na_2IrO_3, Li_2IrO_3

Generic Spin Model

J. Rau, E. Lee, HYK, Phys. Rev. Lett. 112, 077204 (2014)

nearest neighbour:
ideal honeycomb

$$H = \sum_{\gamma \in x,y,z} H^{\gamma},$$



bond-dep. interaction

$$H^z = \sum_{\langle ij \rangle \in z-bond} [\boxed{K_z S_i^z S_j^z + \Gamma_z (S_i^x S_j^y + S_i^y S_j^x)]} + J \mathbf{S}_i \cdot \mathbf{S}_j$$

$$H^x = H^z(x \to y \to z \to x)$$

How to get bond-dependent spin interaction?

$J_{eff} = 1/2$ basis

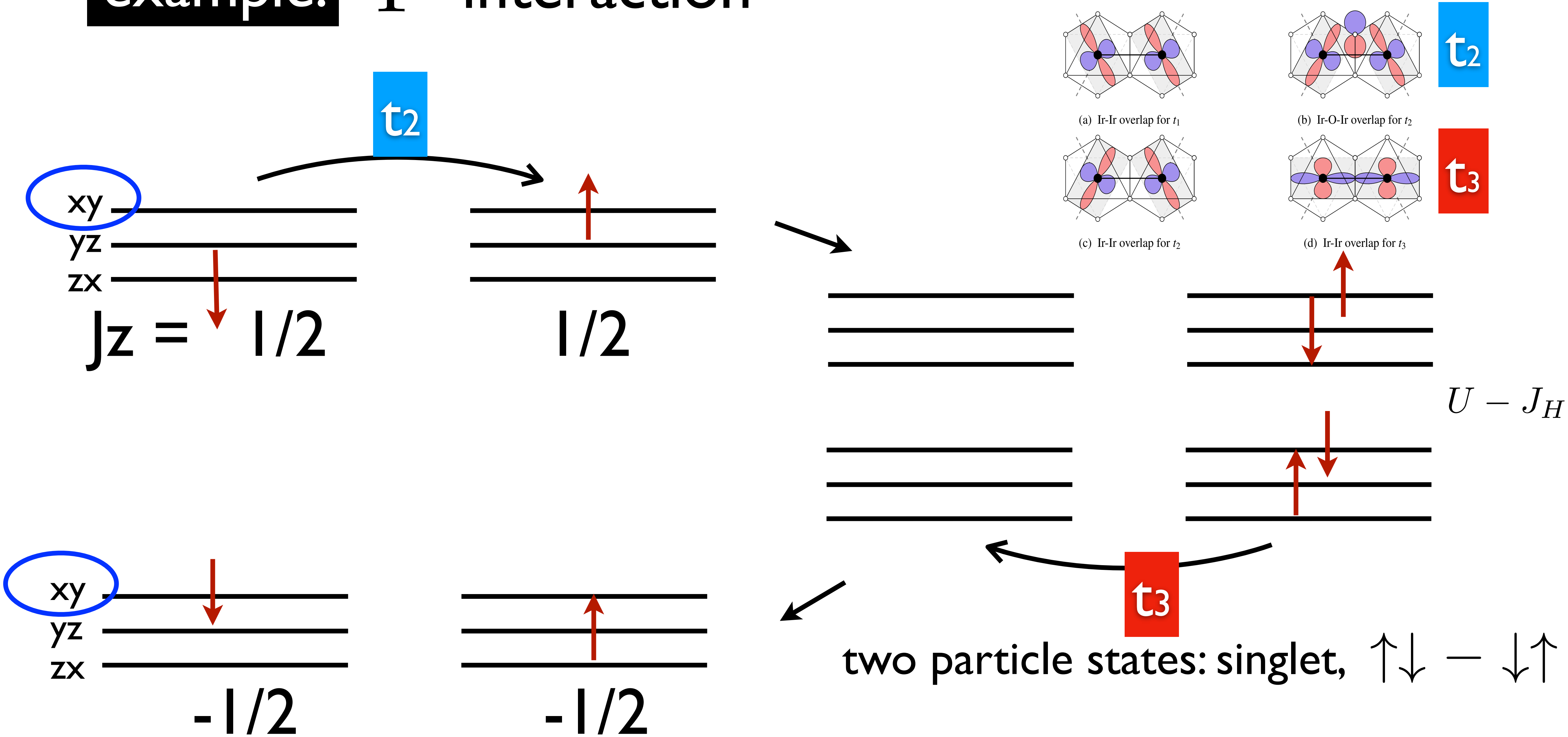
$$
\begin{aligned}
\left|+\tfrac{1}{2}\right\rangle &= \sqrt{\frac{1}{3}}\boxed{\left(|yz\rangle\,|\downarrow\rangle + i\,|zx\rangle\,|\downarrow\rangle + |xy\rangle\,|\uparrow\rangle\right)} \\
&= \sqrt{\frac{2}{3}}\,|1,+1\rangle\,|\downarrow\rangle - i\sqrt{\frac{1}{3}}\,|1,0\rangle\,|\uparrow\rangle
\end{aligned}
$$

TR ↓

$$
\begin{aligned}
\left|-\tfrac{1}{2}\right\rangle &= \sqrt{\frac{1}{3}}\left(|yz\rangle\,|\uparrow\rangle - i\,|zx\rangle\,|\uparrow\rangle - |xy\rangle\,|\downarrow\rangle\right) \\
&= \sqrt{\frac{2}{3}}\,|1,-1\rangle\,|\uparrow\rangle + i\sqrt{\frac{1}{3}}\,|1,0\rangle\,|\downarrow\rangle
\end{aligned}
$$

mixture of different orbitals and different spins

example: Γ interaction



(a) Ir-Ir overlap for t_1

(b) Ir-O-Ir overlap for t_2

(c) Ir-Ir overlap for t_2

(d) Ir-Ir overlap for t_3

two particle states: singlet, $\uparrow\downarrow - \downarrow\uparrow$

one particle

connect up-up and down-down Jz states:

$$\boxed{S_1^+ S_2^+ - S_1^- S_2^- \propto i(S_1^x S_2^y + S_1^y S_2^x)}$$

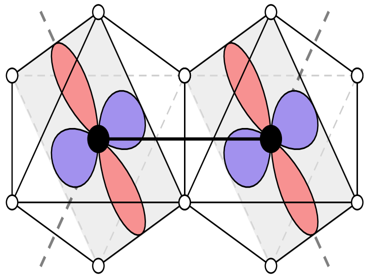
Generic spin model including d-p & d-d hopping

$$H = \sum_{\langle ij \rangle \in \alpha\beta(\gamma)} \left[J \vec{S}_i \cdot \vec{S}_j + \boxed{K S_i^\gamma S_j^\gamma + \Gamma \left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha \right)} \right]$$

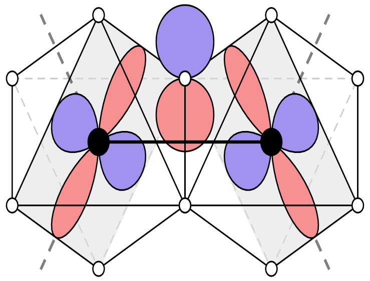
$$J = \frac{4}{27} \left[\frac{6t_1(t_1 + 2t_3)}{U - 3J_H} + \frac{2(t_1 - t_3)^2}{U - J_H} + \frac{(2t_1 + t_3)^2}{U + 2J_H} \right],$$

$$K = \frac{8J_H}{9} \left[\frac{(t_1 - t_3)^2 - 3t_2^2}{(U - 3J_H)(U - J_H)} \right],$$

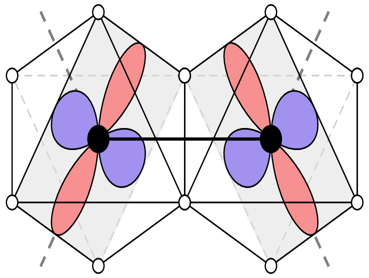
$$\Gamma = \frac{16J_H}{9} \left[\frac{t_2(t_1 - t_3)}{(U - 3J_H)(U - J_H)} \right].$$



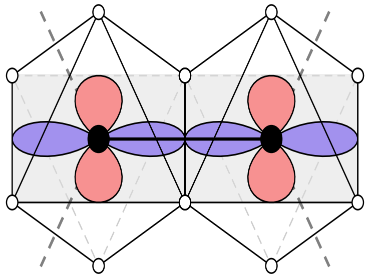
(a) Ir-Ir overlap for t_1



(b) Ir-O-Ir overlap for t_2



(c) Ir-Ir overlap for t_2



(d) Ir-Ir overlap for t_3

$$t_1 = \frac{1}{2}(\boxed{t_{dd\pi}} + t_{dd\delta}), \ t_2 = \frac{1}{2}(t_{dd\pi} - t_{dd\delta}) + \overset{t_0}{\boxed{\frac{t_{pd\pi}^2}{\Delta_{pd}}}}, \ t_3 = \frac{1}{4}(\boxed{3t_{dd\sigma}} + t_{dd\delta}).$$

$t_{dd\pi} : t_{dd\sigma}$

different signs

$$J \propto t_{dd\pi}^2 / U$$

$$K \propto -t_o^2 J_H / U^2$$

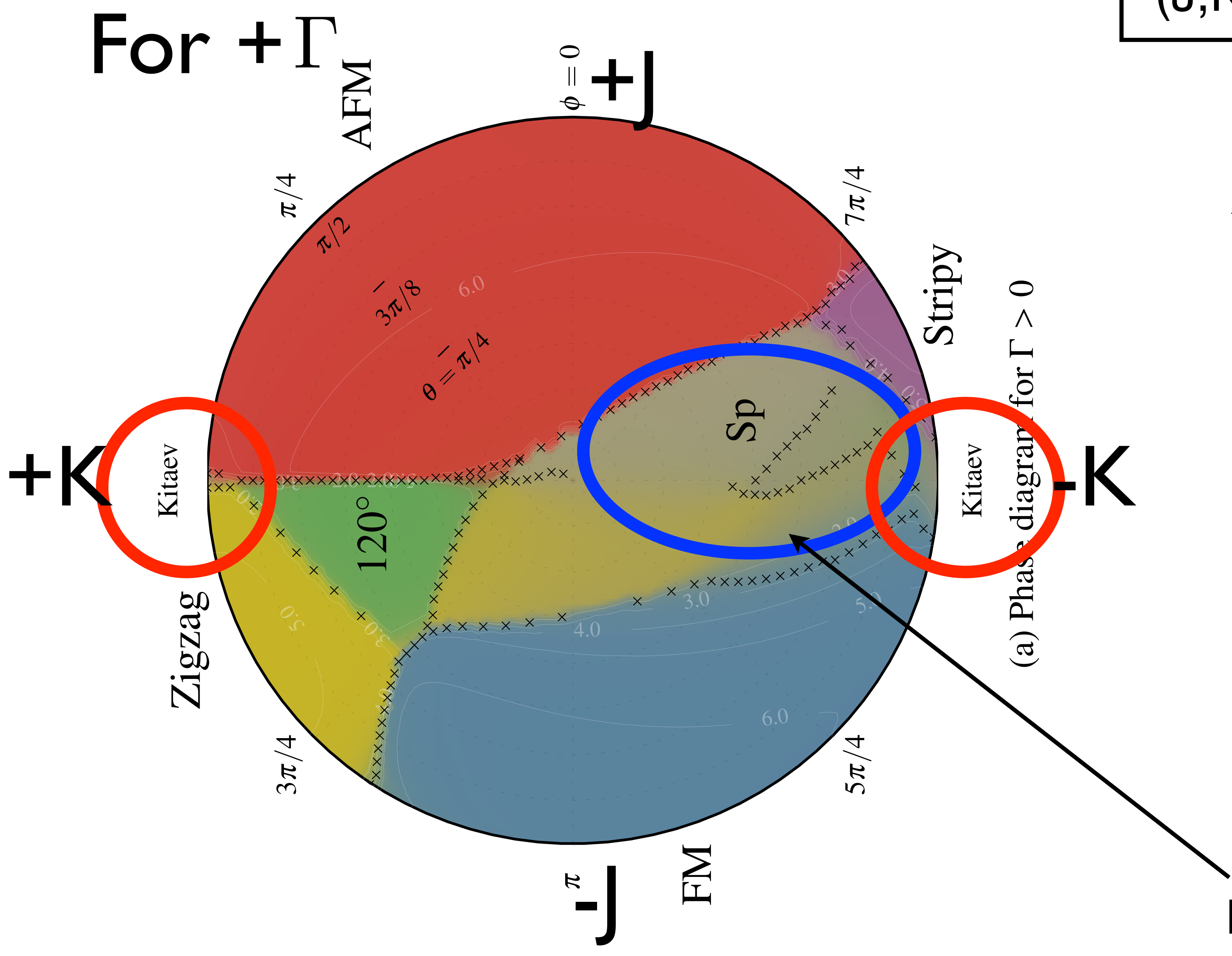
$$\boxed{\Gamma \propto t_o t_{dd\sigma} J_H / U^2}$$

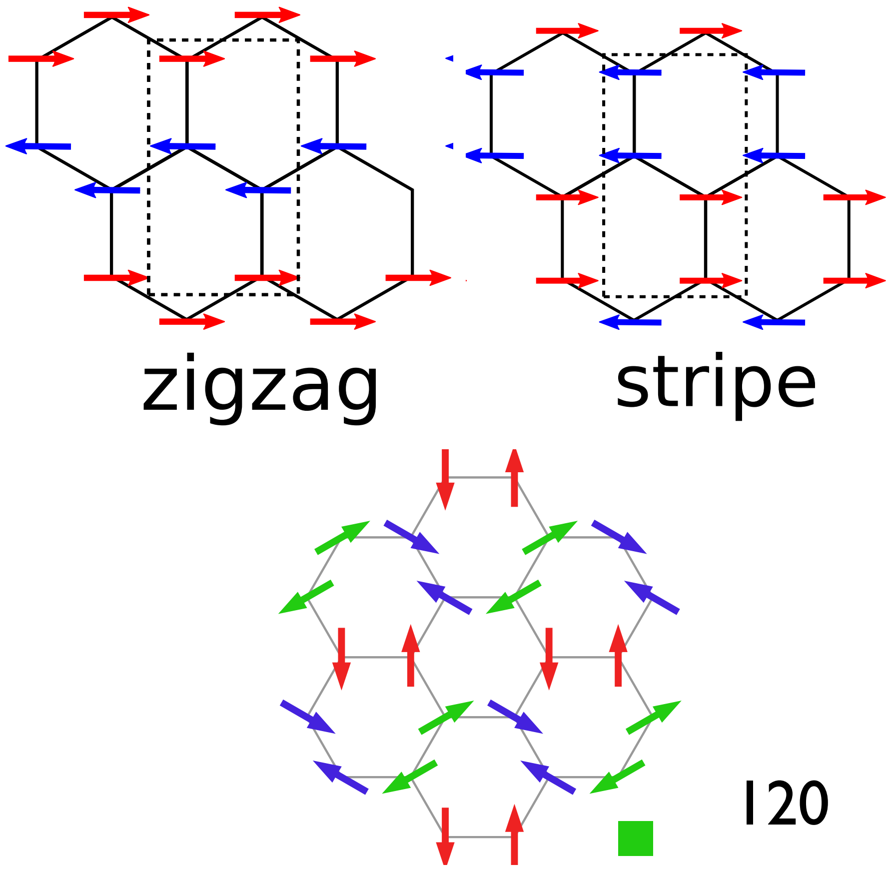
When d-p orbital overlap dominates; $|K|, |\Gamma| > |J|$

Ordered phases nearby KSL

ED 24-site cluster

(J,K,Γ) = (sinθcosφ,sinθsinφ,cosθ)





RuCl3, Na2IrO3, Li2IrO3:
NN+ J3 - nearby KSL!

extremely narrow range of KSL

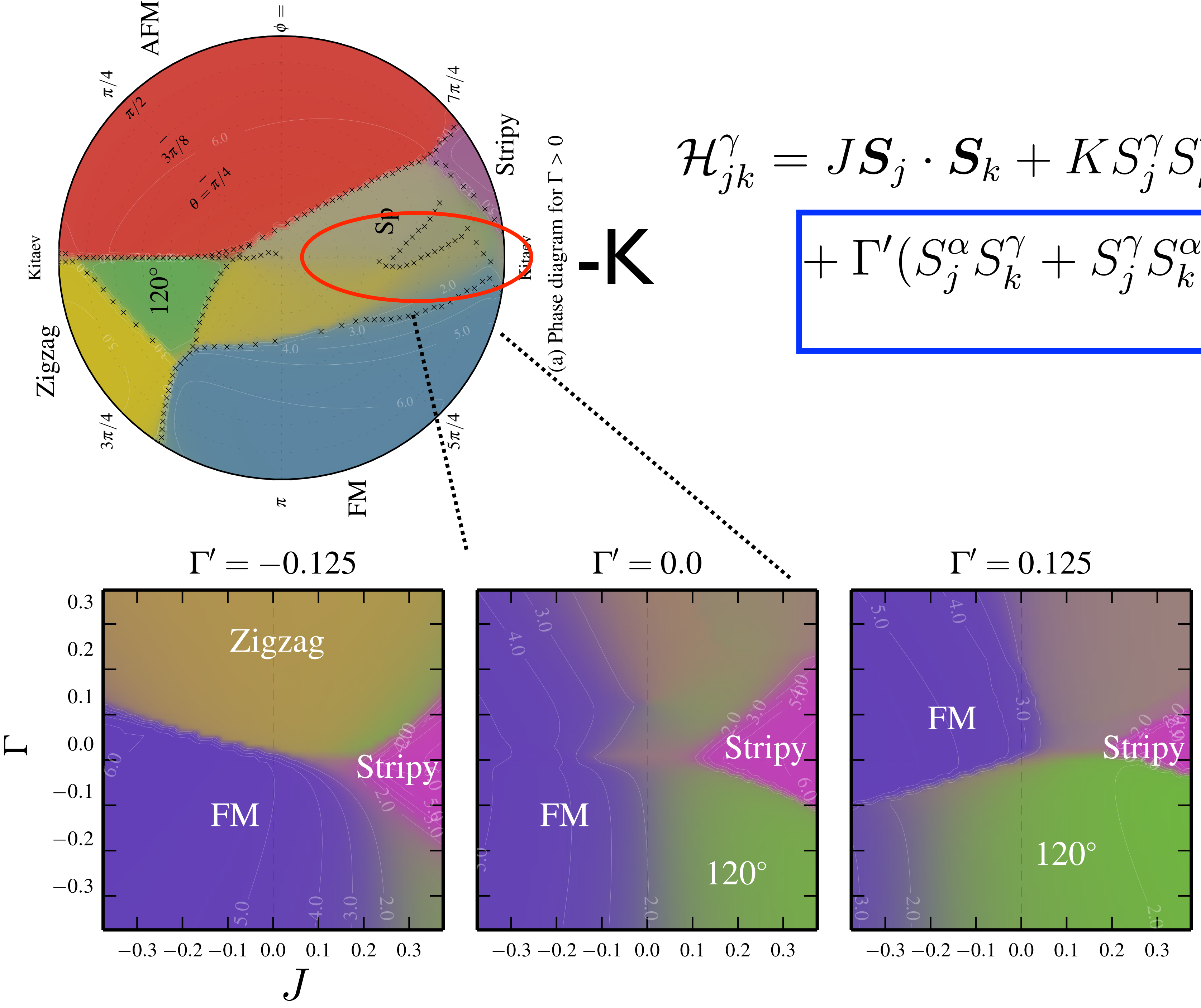
J. Rau, E. Lee, HYK, Phys. Rev. Lett. 112, 077204 (2014)

Nearest neighbour spin model

with trigonal distortion

$$\mathcal{H}_{jk}^{\gamma} = J\boldsymbol{S}_j \cdot \boldsymbol{S}_k + K S_j^{\gamma} S_k^{\gamma} + \Gamma(S_j^{\alpha} S_k^{\beta} + S_j^{\beta} S_k^{\alpha})$$

$$\boxed{+ \Gamma'(S_j^{\alpha} S_k^{\gamma} + S_j^{\gamma} S_k^{\alpha} + S_j^{\beta} S_k^{\gamma} + S_j^{\gamma} S_k^{\beta})}$$



effects of Γ'

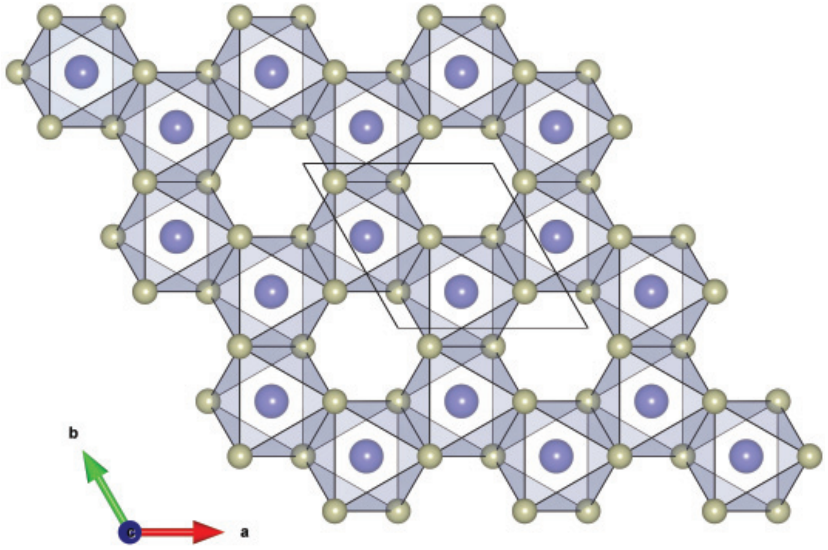
J. Rau, HYK, arXiv:1408.4811

Kitaev Materials: α-RuCl$_3$

PHYSICAL REVIEW B **90**, 041112(R) (2014)

α-RuCl$_3$: A spin-orbit assisted Mott insulator on a honeycomb lattice

K. W. Plumb,[1] J. P. Clancy,[1] L. J. Sandilands,[1] V. Vijay Shankar,[1] Y. F. Hu,[2] K. S. Burch,[1,3]
Hae-Young Kee,[1,4] and Young-June Kim[1,*]

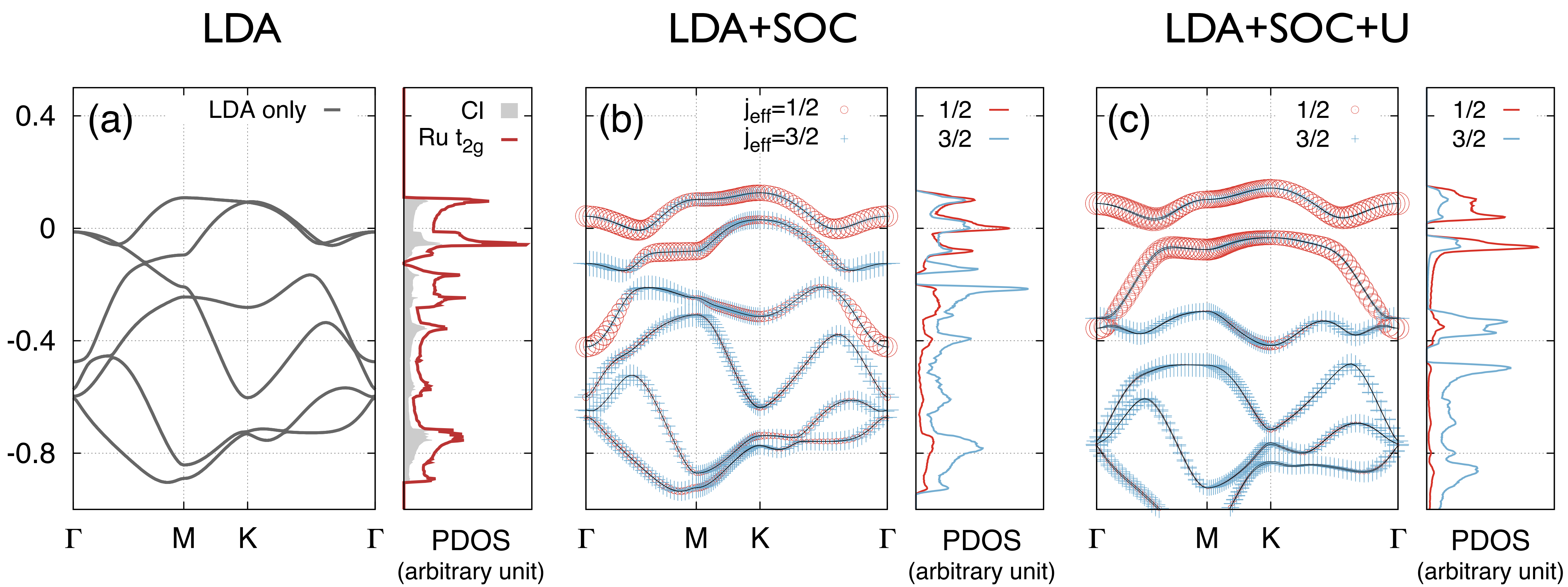


$$\boxed{\mathrm{Ru}^{3+} : 4d^5}$$

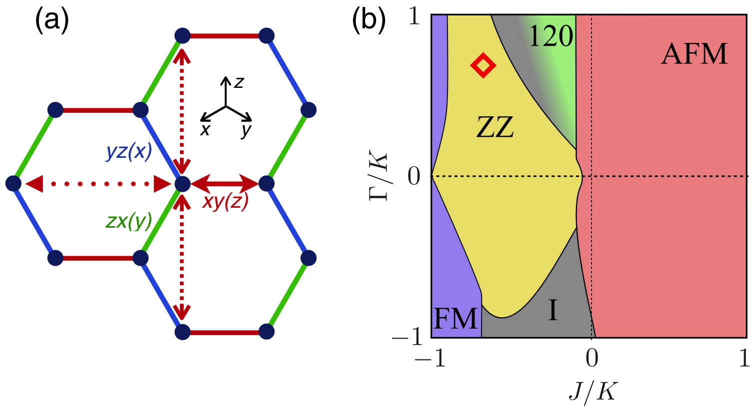
process in α-RuCl$_3$. Then a microscopic spin model relevant for α-RuCl$_3$ should be composed of both the nearest-neighbor Heisenberg and bond-dependent exchange terms denoted by Kitaev K and Γ [44–46].

Kitaev Magnetism in honeycomb RuCl3 with intermediate SOC

HS Kim, ... HYK, PRB 91, 241110 (2015)

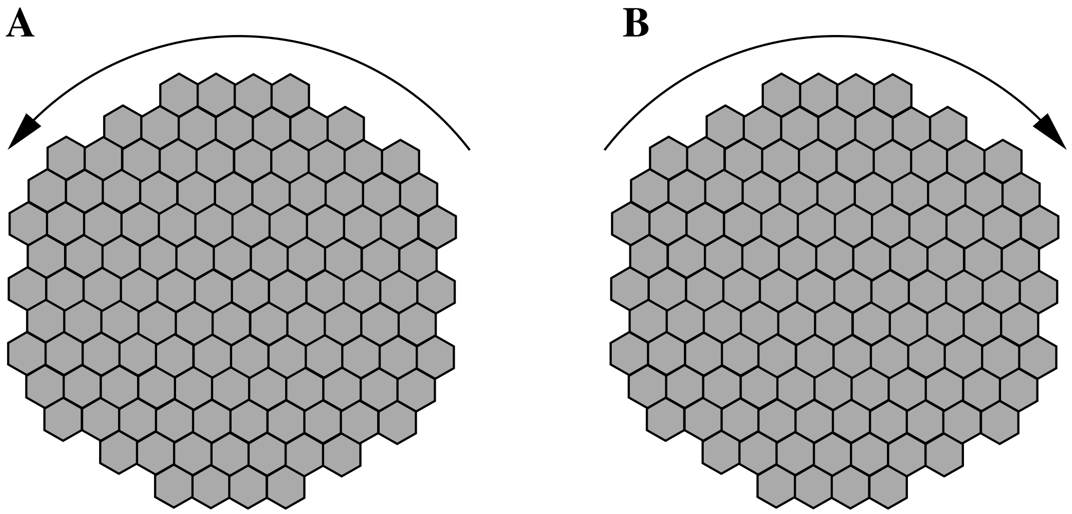


Kitaev Magnetism: U increases effective SOC



Zig-zag ordering
due to other interactions

smoking-gun signature

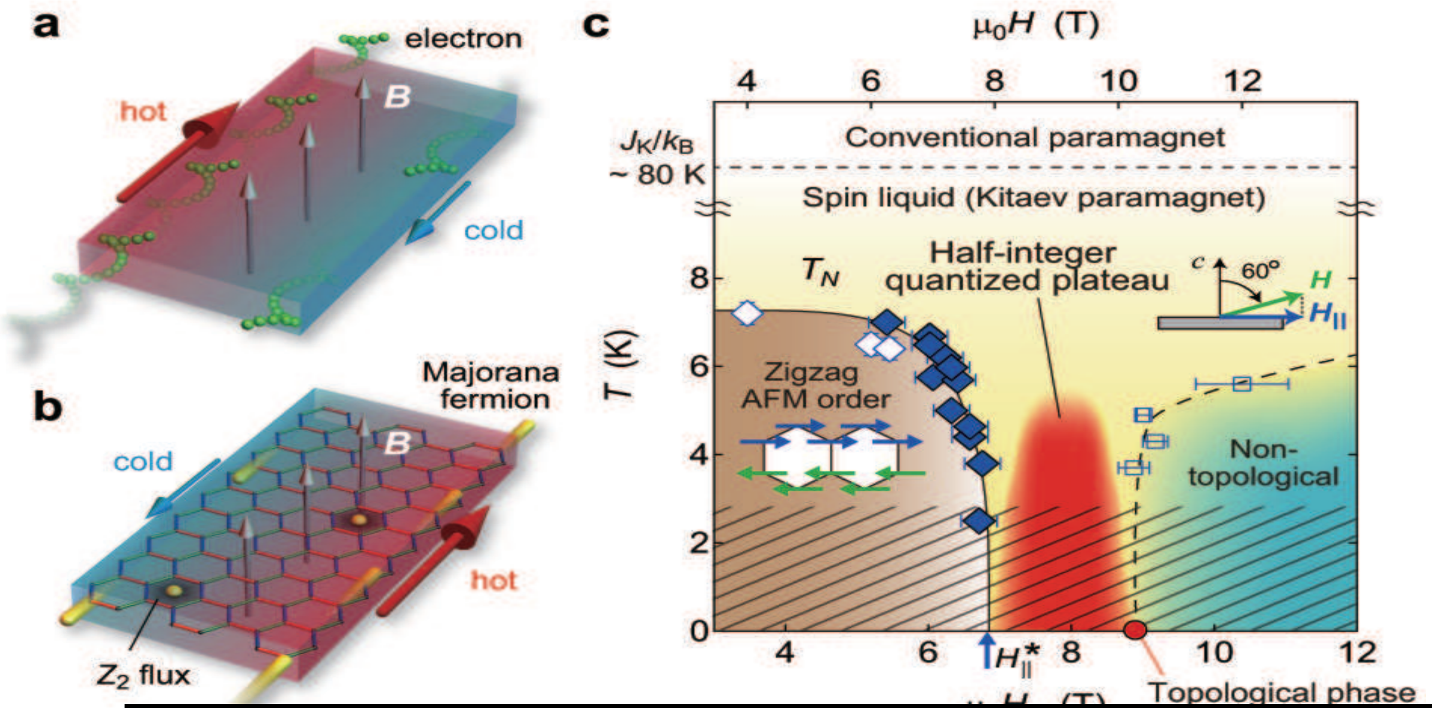


Chiral edge mode : 1/2 quantized thermal Hall conductivity

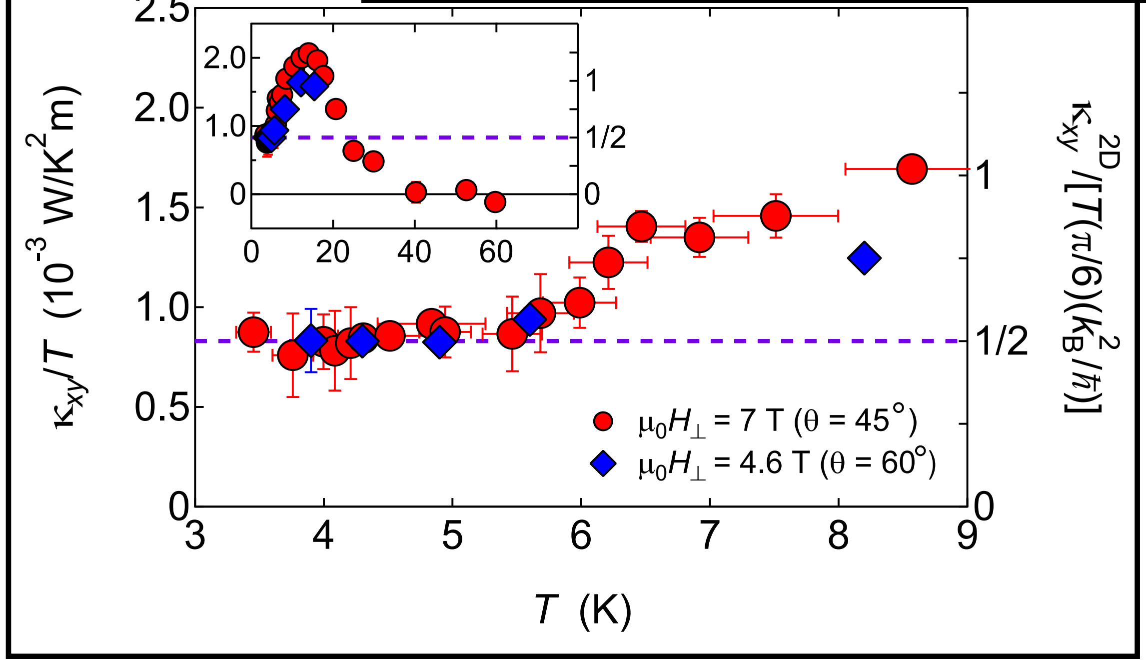
> Chiral edge modes can carry energy, leading to potentially measurable thermal transport. (The temperature T is assumed to be much smaller than the energy gap in the bulk, so that the effect of bulk excitations is negligible.) For quantum Hall systems, this phenomenon was discussed in [56,57]. The energy current along the edge in the left (counterclockwise) direction is given by the following formula:
>
> $$\boxed{I = \frac{\pi}{12} c_{-} T^2,} \qquad (57)$$

A. Kitaev, Annals of Physics 321, 2 (2006):
Anyones in exactly solved model and beyond

Thermal Transport: α-RuCl$_3$



Exciting: To be continued



$$\frac{\kappa_{xy}^{2D}}{T} = c\,\frac{\pi}{6}\,\frac{k_B^2}{\hbar}$$

compare:

$$\sigma_{xy}^{QH} = \nu\,\frac{e^2}{h}$$

Kasahara,.. Y. Matsuda, Nature (2018)

Spin-S Kitaev?

For arbitrary S,

$$W_p = e^{i\pi(S_1^y + S_2^z + S_3^x + S_4^y + S_5^z + S_6^x)}$$

ultra-short range correlations

G. Baskaran, D. Sen, R. Shankar, PRB 78, 115116 (2008)

Quantum spin liquid?

Majorana fermion vs. boson excitations?

half-integer vs. integer S Kiteav?

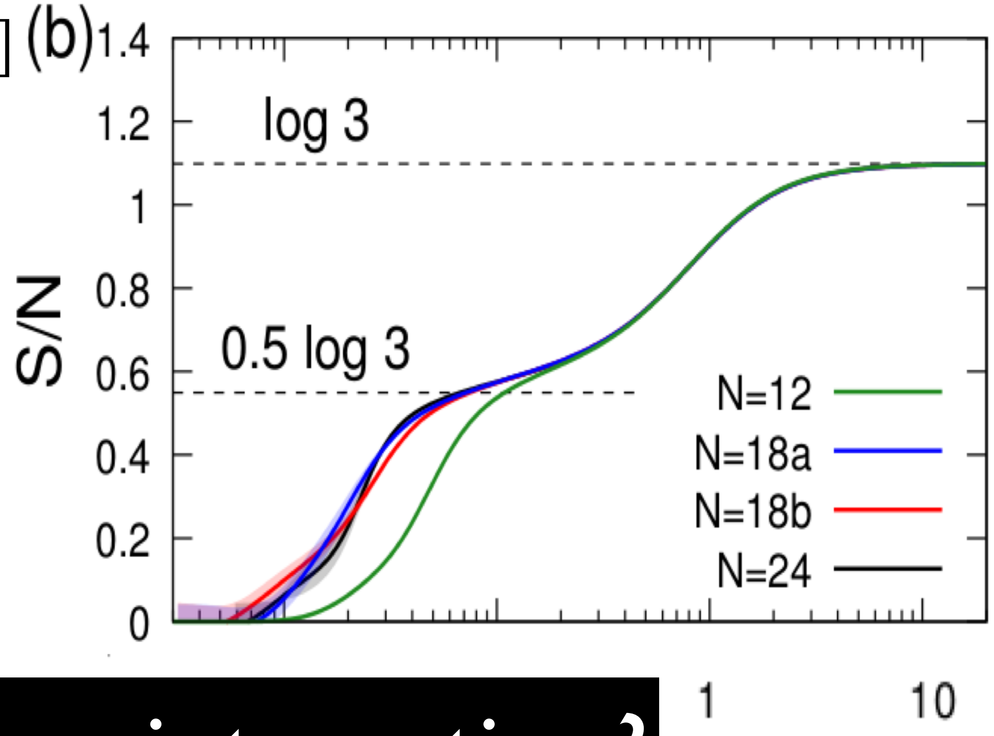
Spin S=1 Kitaev model in the literature.

S=1 Kitaev model:

Plaquette operators that commute with the Hamiltonian [1]

May be a gapless spin liquid [2]

Has incipient entropy plateau [2,3] ⟶



How do we get S=1 Kitaev interaction?

[1] G. Baskaran, D. Sen, and R. Shankar, **Phys. Rev. B 78, 115116 (2008).**
[2] A. Koga, H. Tomishige, and J. Nasu, **Journal of the Physical Society of Japan 87, 063703 (2018).**
[3] J. Oitmaa, A. Koga, and R. R. P. Singh, **Phys. Rev. B 98, 214404 (2018).**

Derivation of Kitaev interaction for S=1

$J_{eff} = 1/2$ basis

$$\begin{aligned} \left|+\tfrac{1}{2}\right\rangle &= \sqrt{\frac{1}{3}}\boxed{(|yz\rangle\,|\downarrow\rangle + i\,|zx\rangle\,|\downarrow\rangle + |xy\rangle\,|\uparrow\rangle)} \\ &= \sqrt{\frac{2}{3}}\,|1,+1\rangle\,|\downarrow\rangle - i\sqrt{\frac{1}{3}}\,|1,0\rangle\,|\uparrow\rangle \end{aligned}$$

TR

$$\begin{aligned} \left|-\tfrac{1}{2}\right\rangle &= \sqrt{\frac{1}{3}}(|yz\rangle\,|\uparrow\rangle - i\,|zx\rangle\,|\uparrow\rangle - |xy\rangle\,|\downarrow\rangle) \\ &= \sqrt{\frac{2}{3}}\,|1,-1\rangle\,|\uparrow\rangle + i\sqrt{\frac{1}{3}}\,|1,0\rangle\,|\downarrow\rangle \end{aligned}$$

mixture of t2g orbitals and different spins

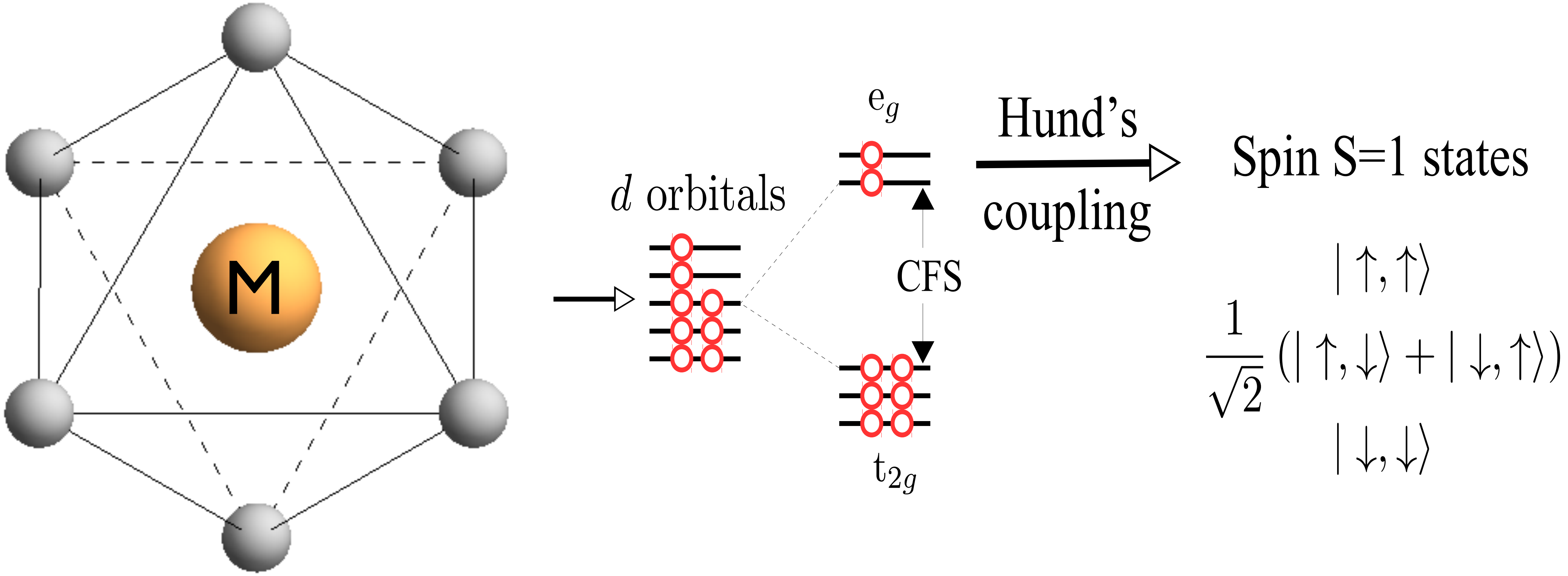
S=1; d2 or d8 with Hund's coupling

: no mixture of spin and orbitals?

Higher spin model derivation

P. Peter Stavropoulos, D. Peira, HYK PRL (2019)

ex: spin one (d8)



d orbitals

e_g

CFS

t_{2g}

Hund's coupling

Spin S=1 states

$$|\uparrow,\uparrow\rangle$$

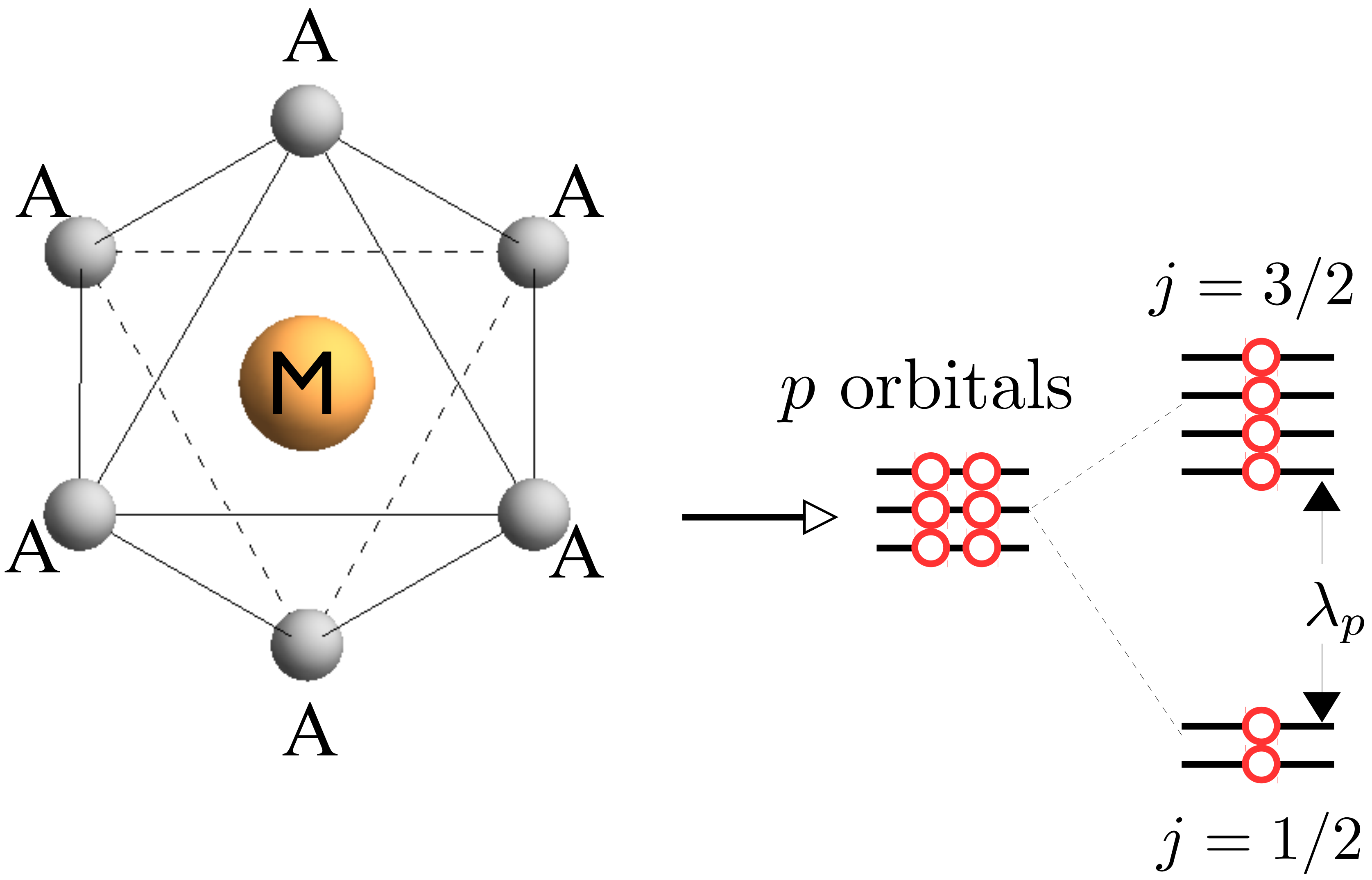
$$\frac{1}{\sqrt{2}}(|\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle)$$

$$|\downarrow,\downarrow\rangle$$

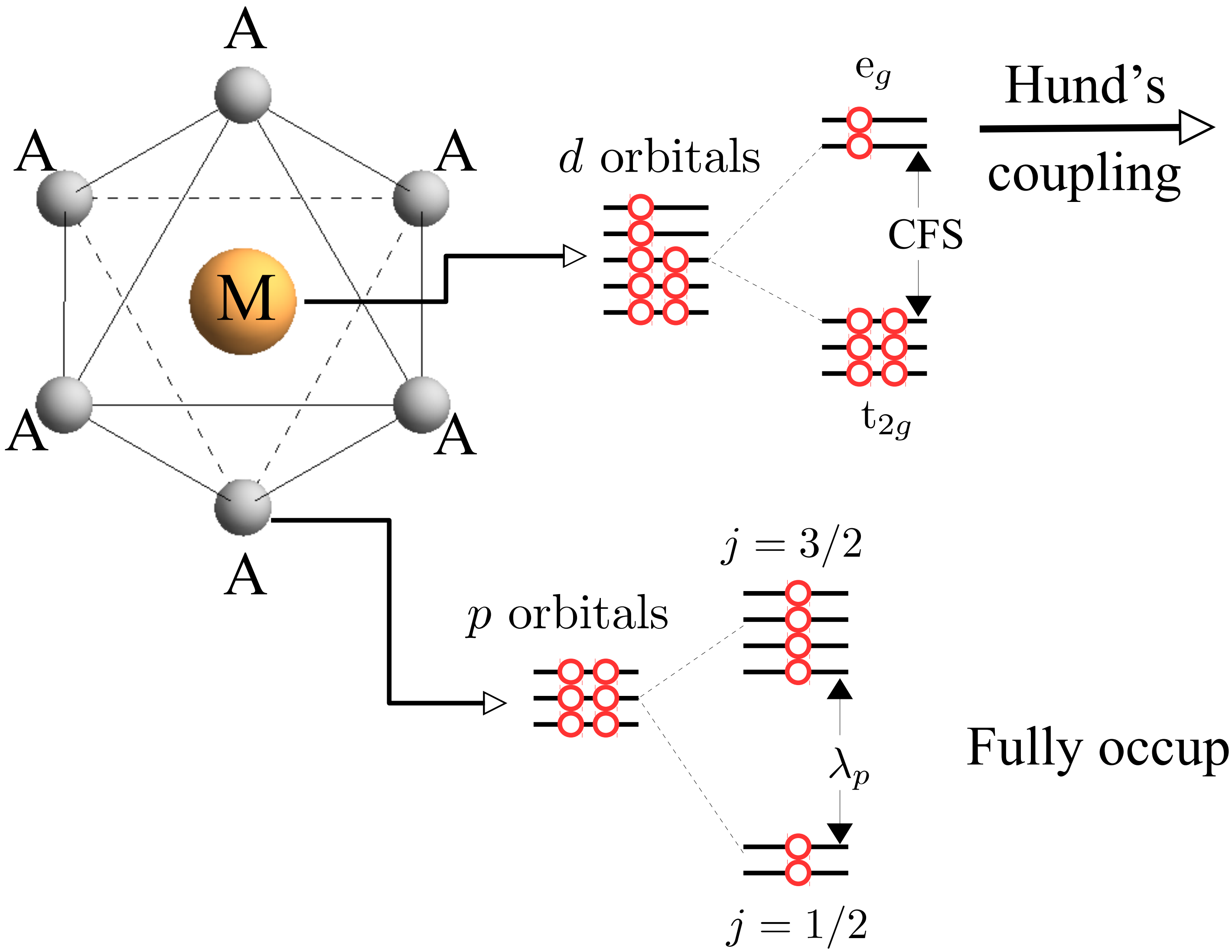
Crystal field splitting > Hund's coupling >> SOC

No mixture of different spins

Heavy Anions



on-site H_0 = Kanamori (U, U', Hund's) + SOC



Spin S=1 states

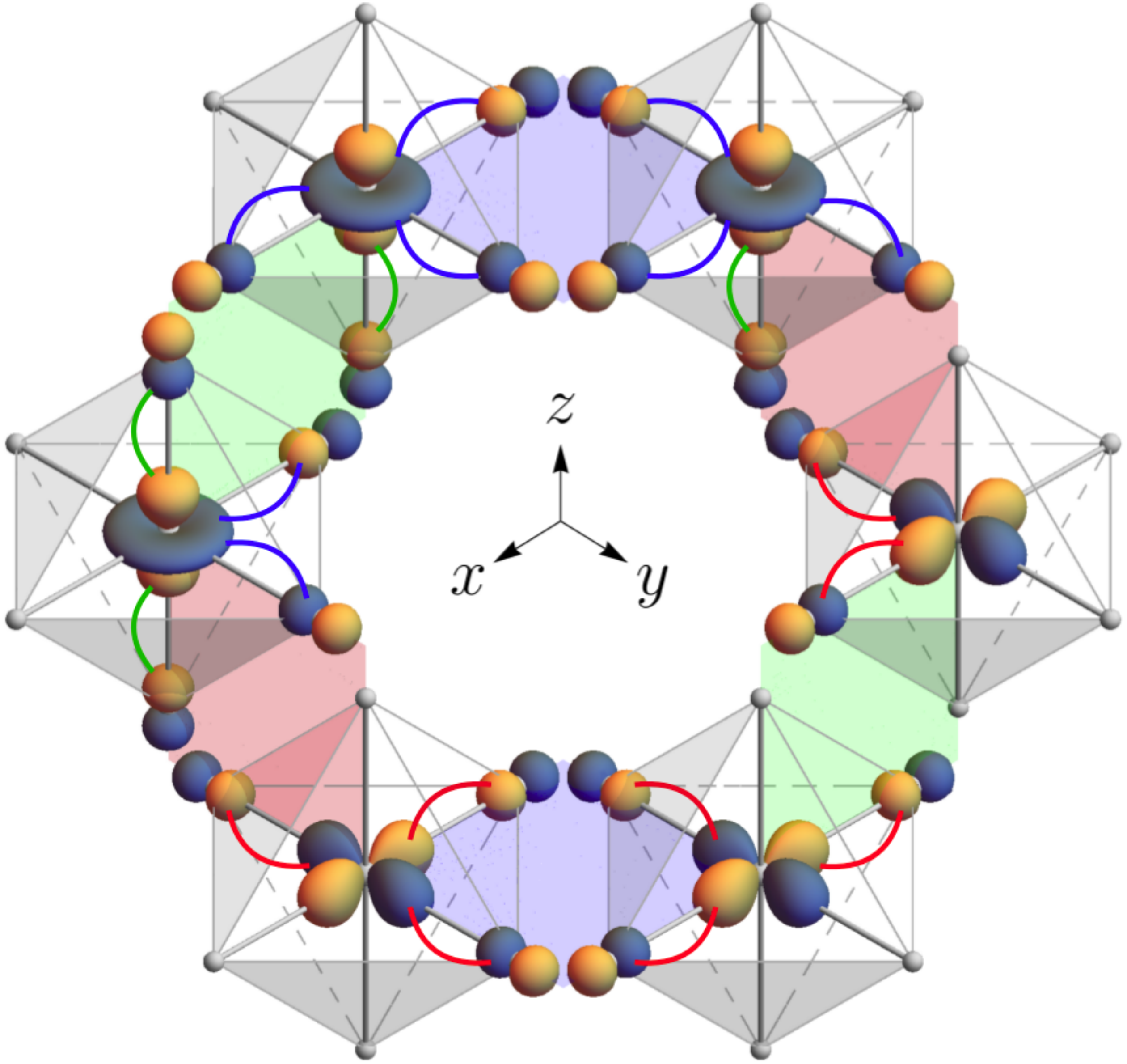
$$|\uparrow,\uparrow\rangle$$

$$\frac{1}{\sqrt{2}}(|\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle)$$

$$|\downarrow,\downarrow\rangle$$

Fully occupied *p* orbitals

Hopping between two M sites via heavy A sites



t_1 🟥

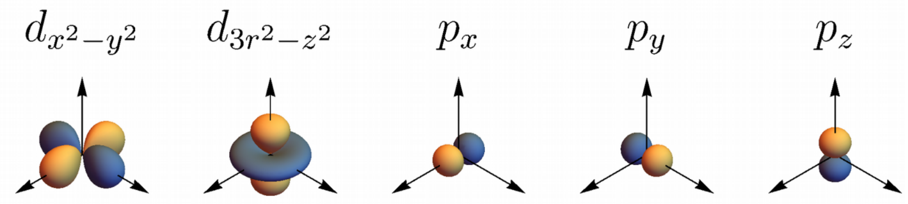
t_2 🟦

t_3 🟩

Hopping integrals
M to A sites:

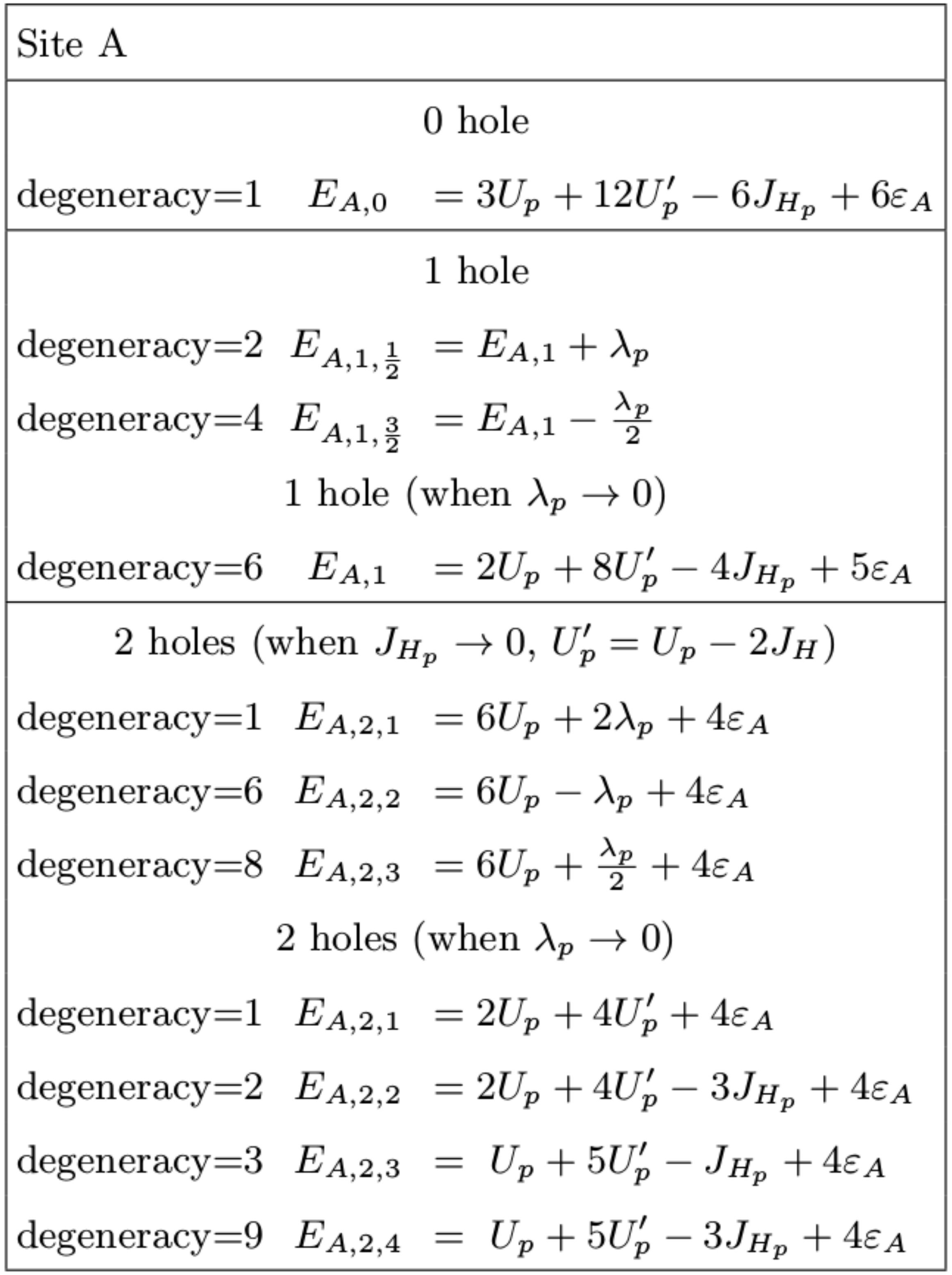
Cubic symmetry:

$$t_1 = \frac{\sqrt{3}}{2} t_{pd\sigma}\,,$$
$$t_2 = \frac{1}{2} t_{pd\sigma}\,,$$
$$t_3 = t_{pd\sigma}$$

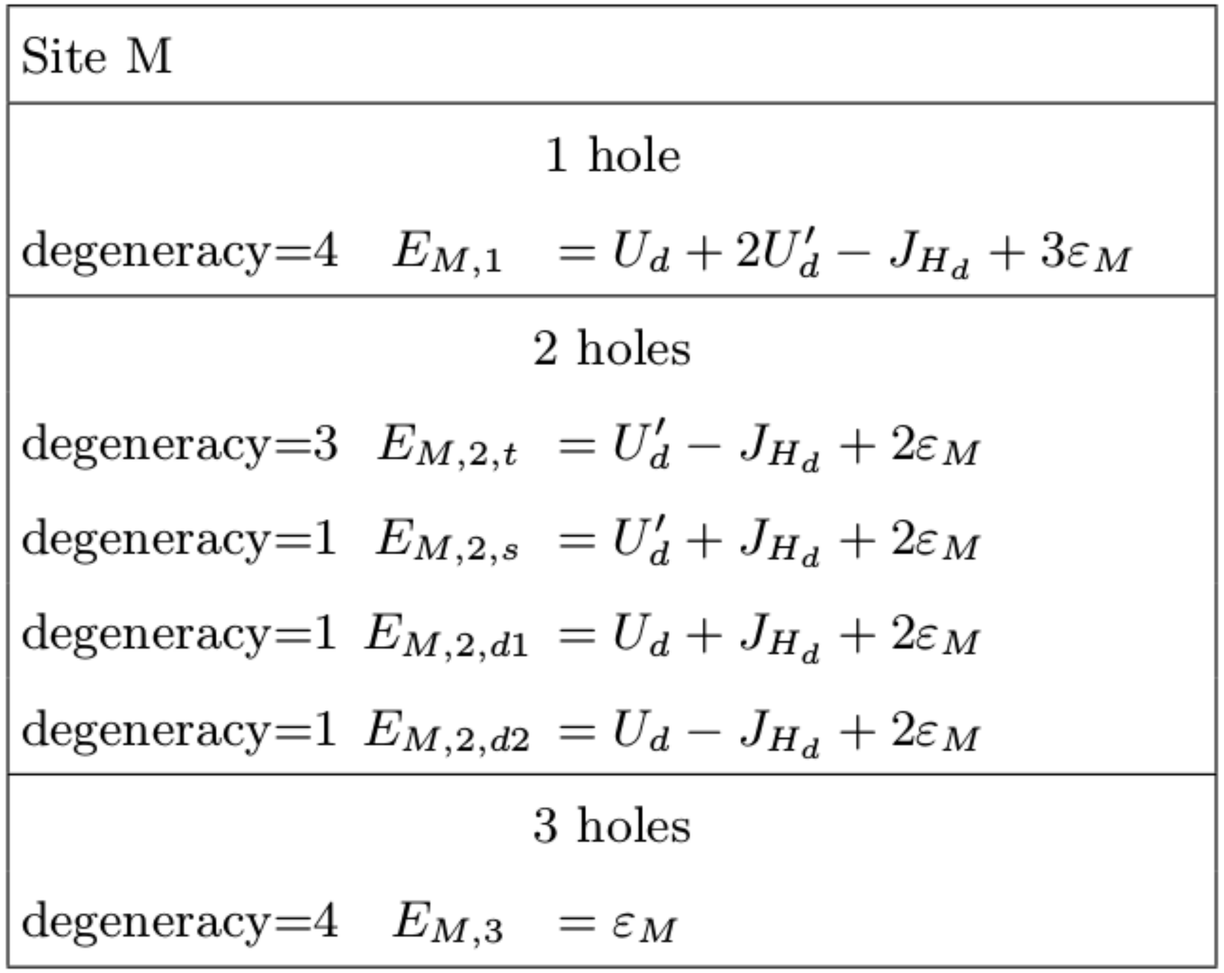


Perturbation theory

H_0 : on-site interaction

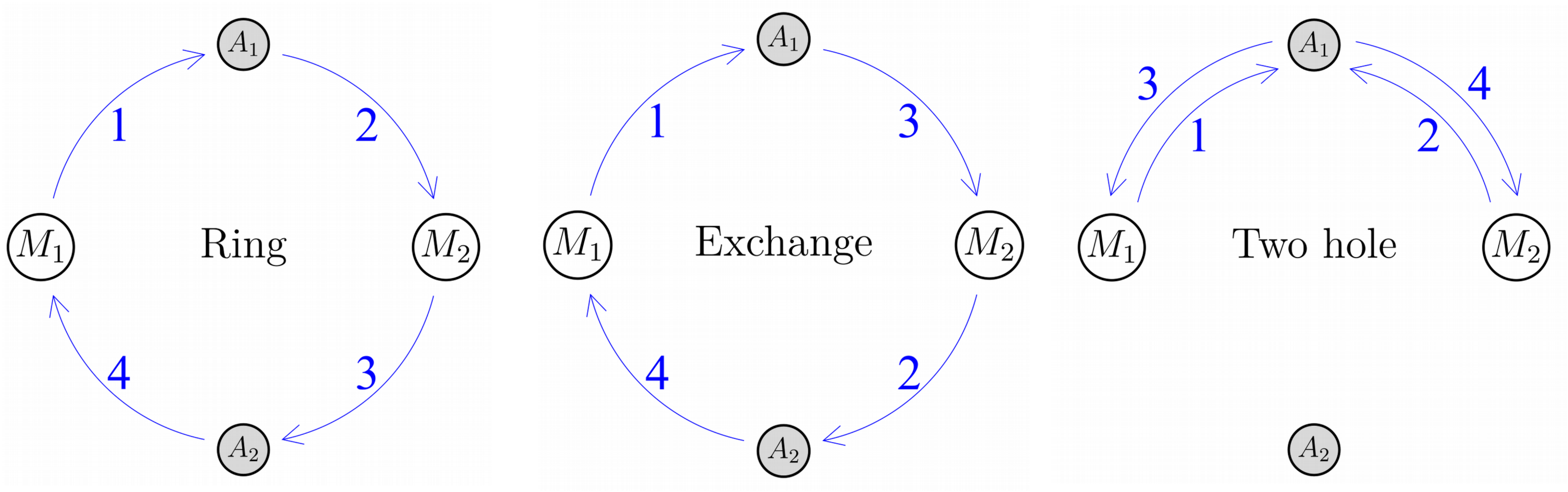


Site A		
0 hole		
degeneracy=1	$E_{A,0}$	$= 3U_p + 12U_p' - 6J_{H_p} + 6\varepsilon_A$
1 hole		
degeneracy=2	$E_{A,1,\frac{1}{2}}$	$= E_{A,1} + \lambda_p$
degeneracy=4	$E_{A,1,\frac{3}{2}}$	$= E_{A,1} - \frac{\lambda_p}{2}$
1 hole (when $\lambda_p \to 0$)		
degeneracy=6	$E_{A,1}$	$= 2U_p + 8U_p' - 4J_{H_p} + 5\varepsilon_A$
2 holes (when $J_{H_p} \to 0$, $U_p' = U_p - 2J_H$)		
degeneracy=1	$E_{A,2,1}$	$= 6U_p + 2\lambda_p + 4\varepsilon_A$
degeneracy=6	$E_{A,2,2}$	$= 6U_p - \lambda_p + 4\varepsilon_A$
degeneracy=8	$E_{A,2,3}$	$= 6U_p + \frac{\lambda_p}{2} + 4\varepsilon_A$
2 holes (when $\lambda_p \to 0$)		
degeneracy=1	$E_{A,2,1}$	$= 2U_p + 4U_p' + 4\varepsilon_A$
degeneracy=2	$E_{A,2,2}$	$= 2U_p + 4U_p' - 3J_{H_p} + 4\varepsilon_A$
degeneracy=3	$E_{A,2,3}$	$= U_p + 5U_p' - J_{H_p} + 4\varepsilon_A$
degeneracy=9	$E_{A,2,4}$	$= U_p + 5U_p' - 3J_{H_p} + 4\varepsilon_A$



Site M		
1 hole		
degeneracy=4	$E_{M,1}$	$= U_d + 2U_d' - J_{H_d} + 3\varepsilon_M$
2 holes		
degeneracy=3	$E_{M,2,t}$	$= U_d' - J_{H_d} + 2\varepsilon_M$
degeneracy=1	$E_{M,2,s}$	$= U_d' + J_{H_d} + 2\varepsilon_M$
degeneracy=1	$E_{M,2,d1}$	$= U_d + J_{H_d} + 2\varepsilon_M$
degeneracy=1	$E_{M,2,d2}$	$= U_d - J_{H_d} + 2\varepsilon_M$
3 holes		
degeneracy=4	$E_{M,3}$	$= \varepsilon_M$

keep up to 4th order



P. Peter Stavropoulos, D. Peira, HYK PRL (2019)

Indirect **Superexchange** paths using cubic symmetry and in the limit $\lambda_p \gg J_{H_p}$

$$\boxed{K = -2J_{\text{ind}}}$$

Ferromagnetic J_{ind}

When $\Delta = \epsilon_M - \epsilon_A$ and U_d dominant:

$$K \sim \frac{3}{2}\lambda_p^2 t_{pd\sigma}^4 \left(\frac{1}{(2U_d + \Delta)^5} + \frac{1}{2U_d\,(2U_d + \Delta)^4} \right) \xrightarrow[\text{limit}]{\text{Mott}} \frac{3}{4}\frac{\lambda_p^2 t_{pd\sigma}^4}{U_d \Delta^4} \equiv \frac{3}{4}\frac{t_{\text{eff}}^2}{U_d}$$

Antiferromagnetic Kitaev!

Where $t_{\text{eff}} = \dfrac{\lambda_p t_{pd\sigma}^2}{\Delta^2}$ effective hopping between the M and M site via A sites

Direct $J_d = 4t^2/U$

$\boxed{\Gamma = 0}$: up to 4th order

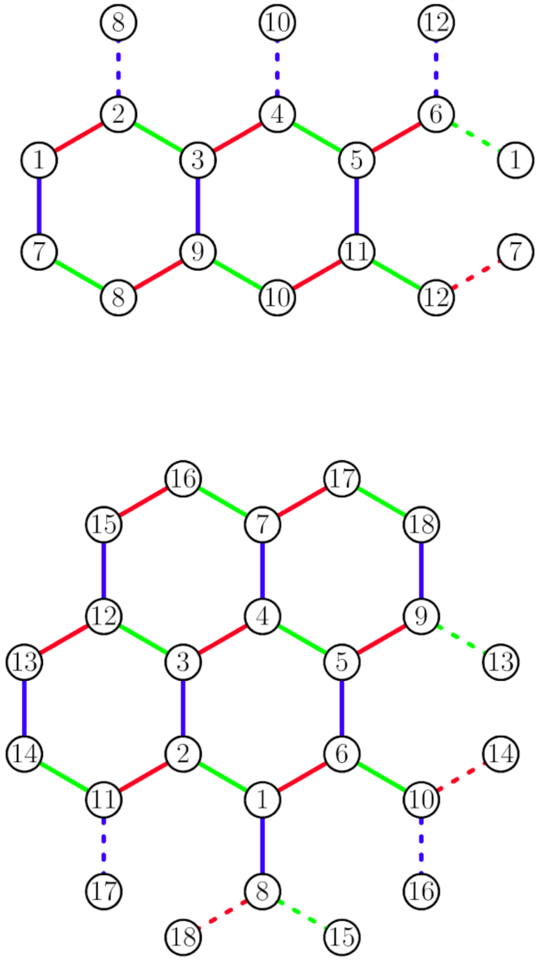
Hamiltonian $$H_{ij}^{\gamma} = \underline{K S_k^{\gamma} S_j^{\gamma}} + J\mathbf{S}_i \cdot \mathbf{S}_j$$

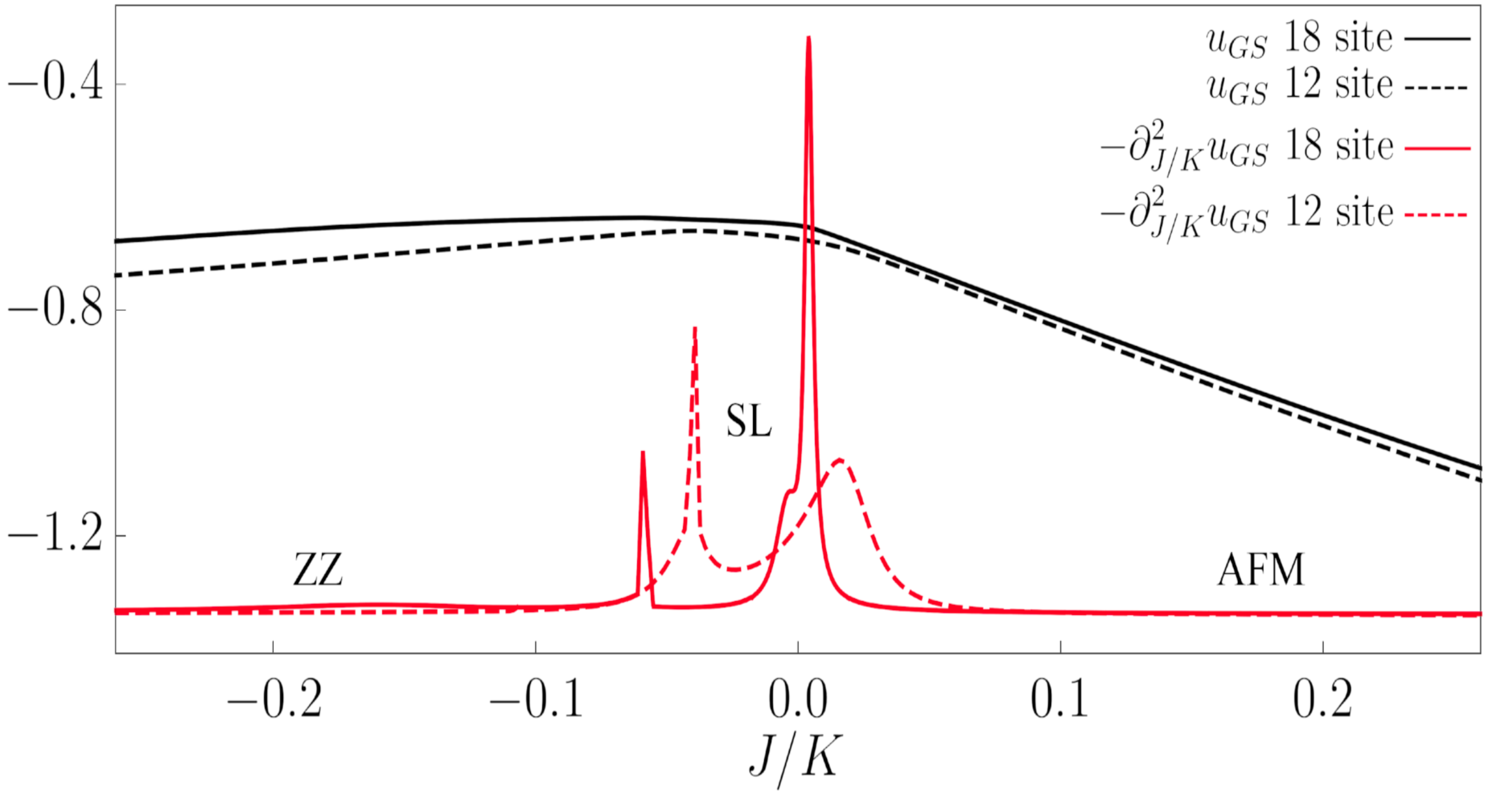
AF Kitaev

$$J = -|J_{ind}| + J_d$$

ED calculation: S=1 KJ model

12 & 18 sites

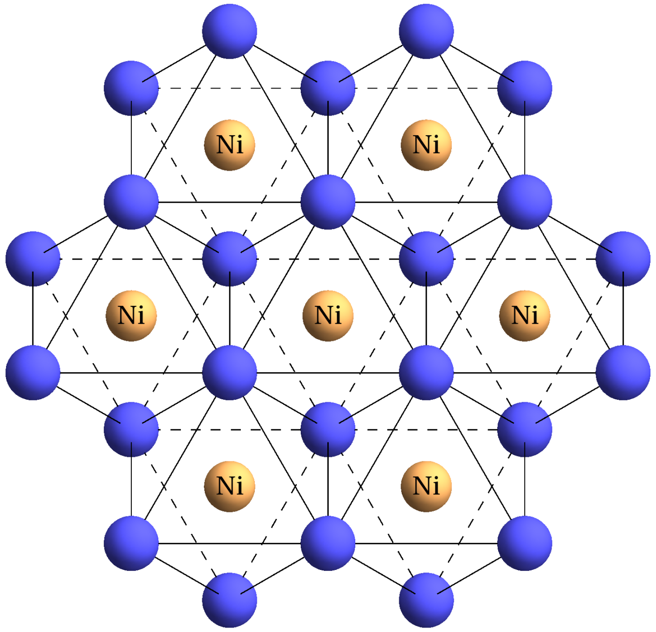




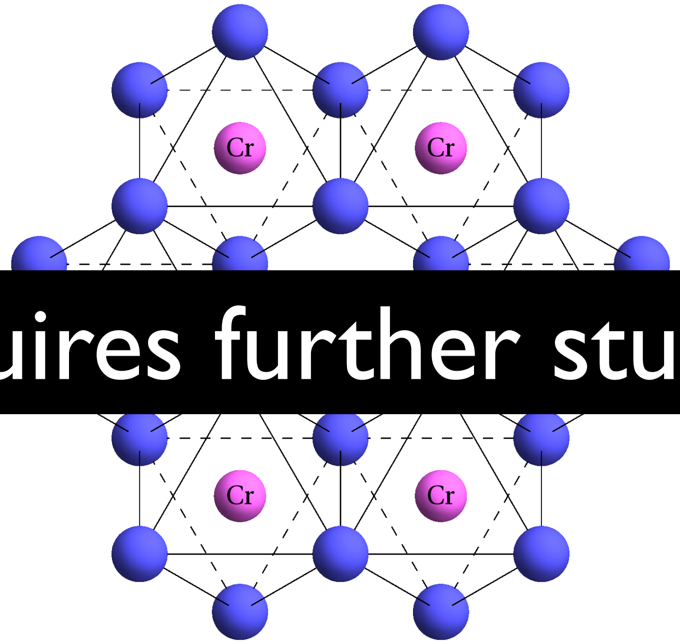
Candidate Materials

van der Waals Materials

NiI2 (S=1 triangle)



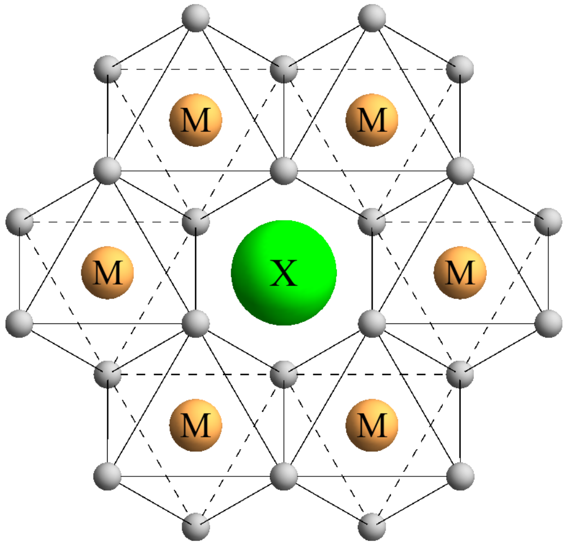
CrI3 (S=3/2 honeycomb)

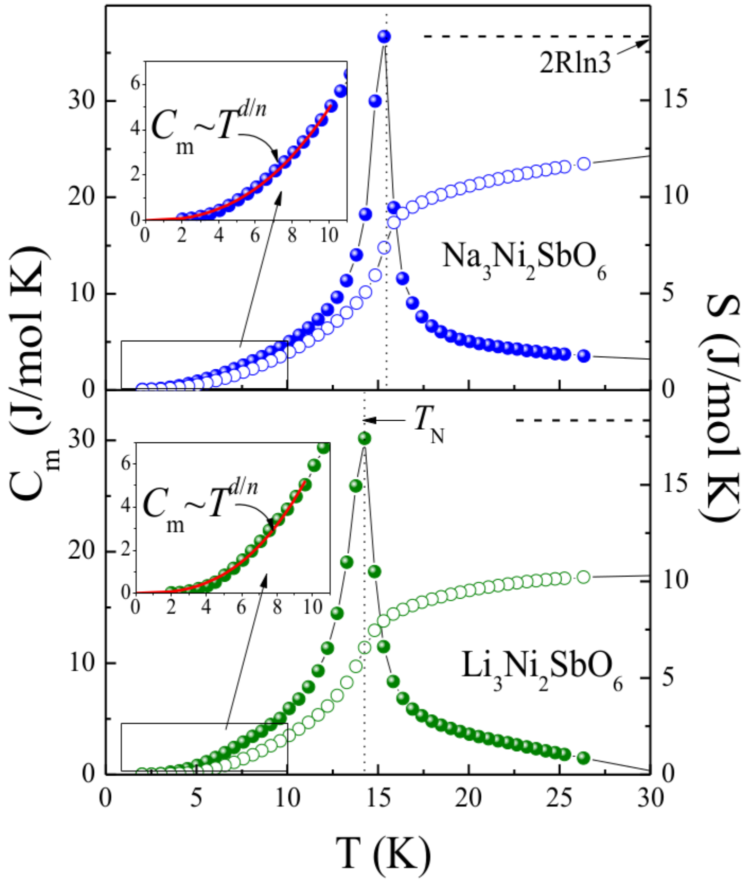


requires further studies

Transition metal oxides

$A_3M_2XO_6$ (A=Li, Na, X=Bi, Sb)

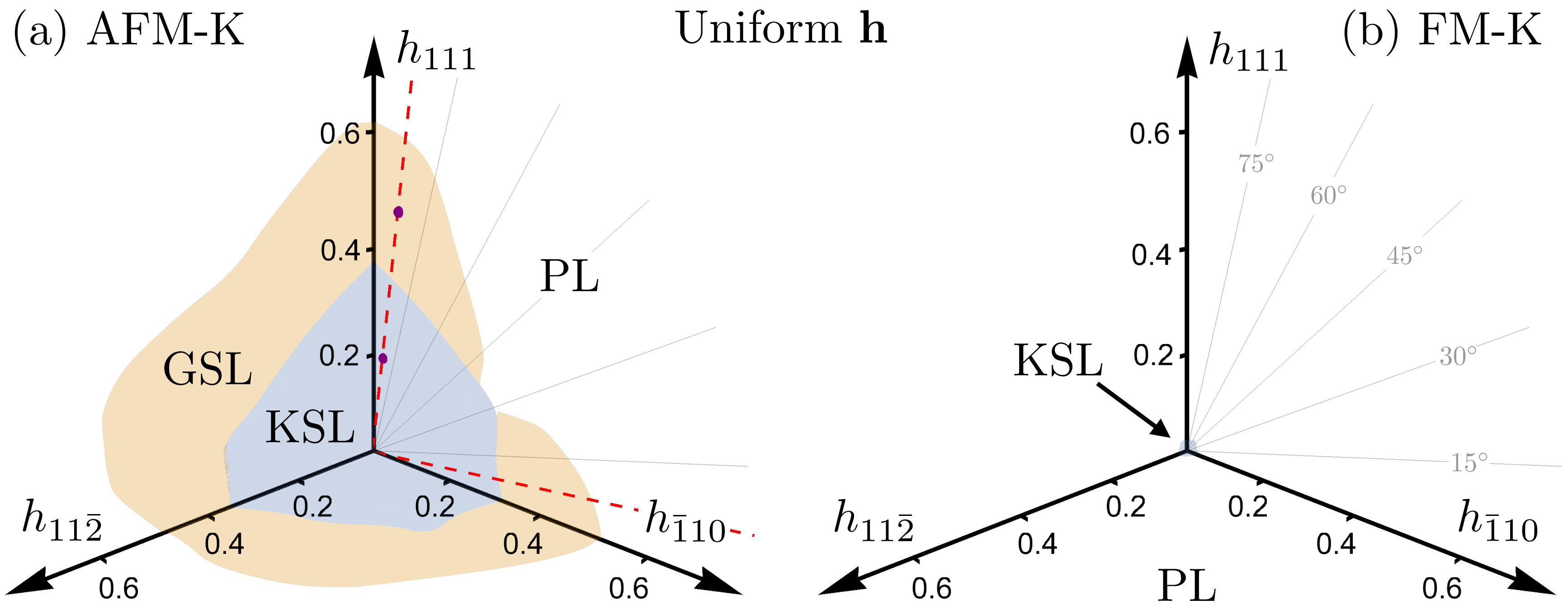




E.A.Zvereva, et al, PRB 92, 144401(2015);
A. I. Kurbakov, et al, PRB 96, 024417 (2017)

Field-driven U(1) spin liquid: transition from Kitaev to U(1) spin liquid near AF Kitaev region

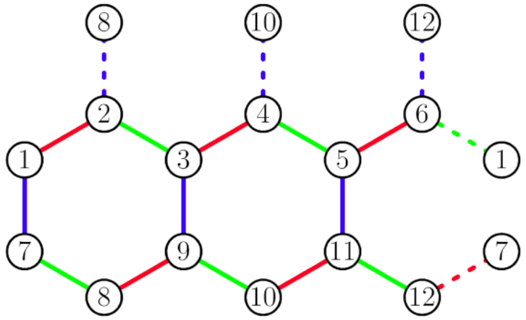
C. Hickey, S. Trebst, Nat. Comm. 10, 530 (2019);
OSU (Y.-M. Lu, N. Trivedi), PI (Y. He), & many others

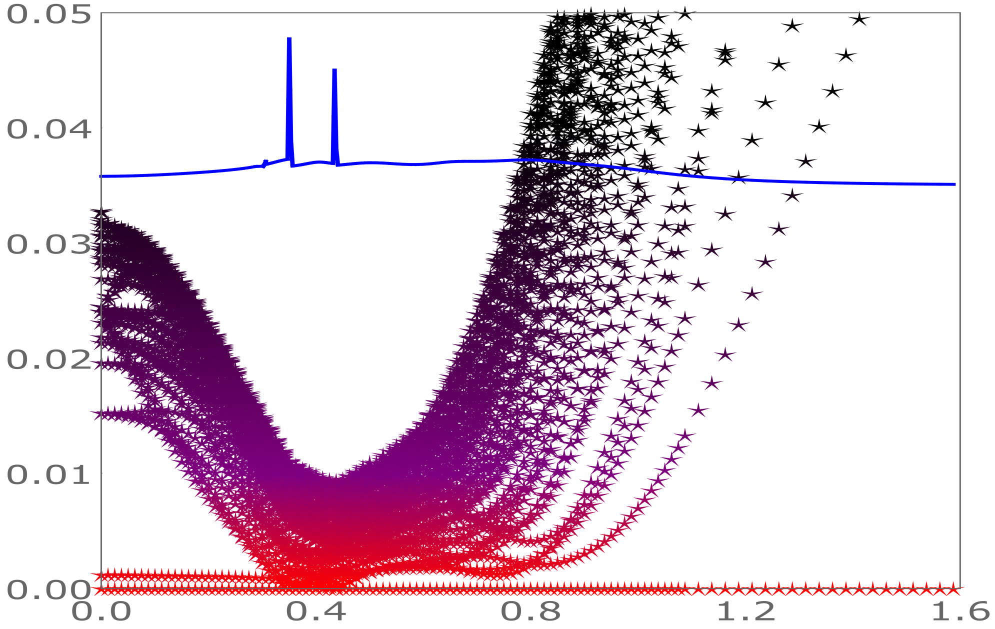


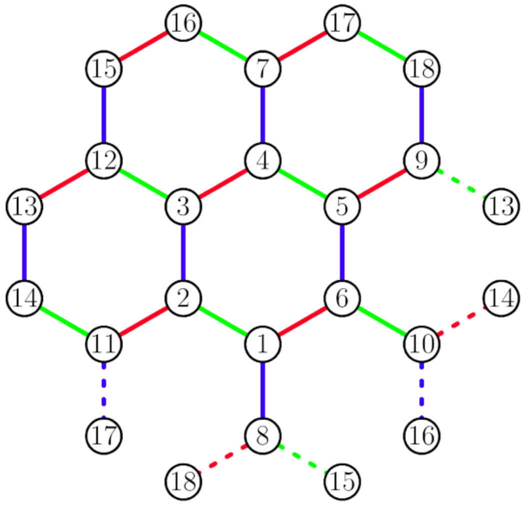
fascinating result; S=1 AF Kitaev?

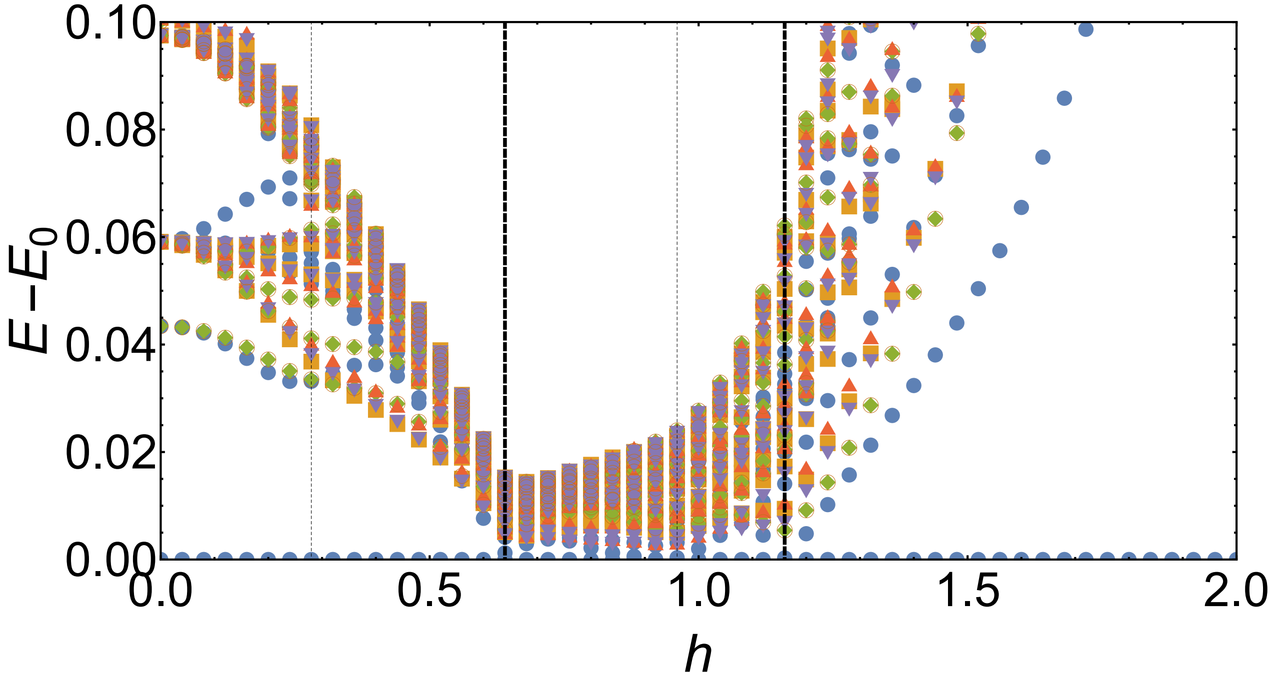
Field induced spin liquid states?

12 &18 sites ED AF Kitaev + field





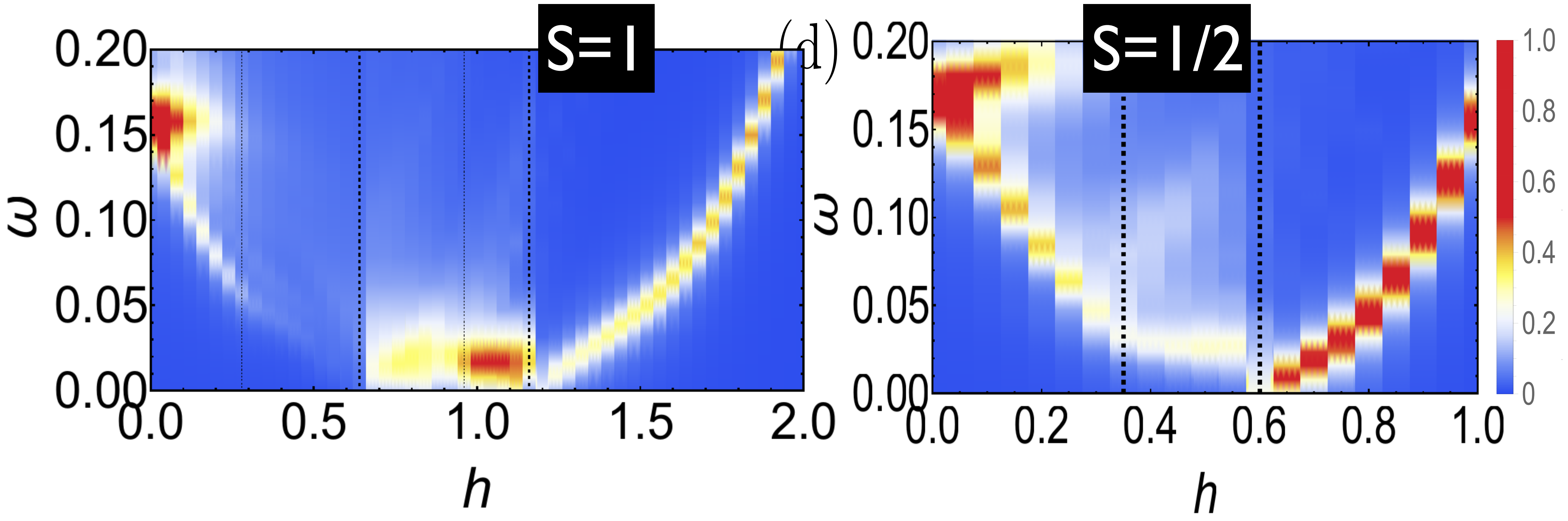




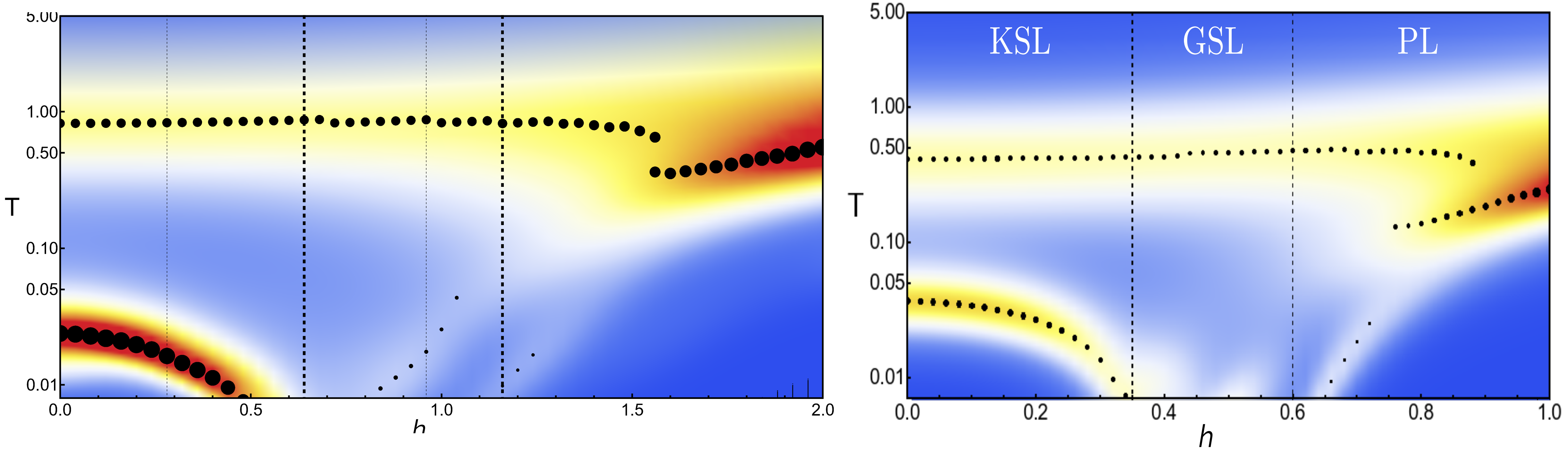
C. Hickey, P. Stavropoulos, C. Berke, S. Trebst, HYK, unpublished

Dynamical structure factor

C. Hickey, S. Trebst, Nat. Comm. 10, 530 (2019)



Specific heat



Summary

Higher spin Kitaev materials: 3d with strong Hund's coupling
+ heavy anions with strong SOC

AF Kitaev: gapless intermediate states under field

Open questions

S=1
fractional excitations in low field & intermediate field?