Symmetry & Strategy to extract Kitaev interaction in spin-S Kitaev Materials

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Review article on "Beyond Kitaev physics", N. Perkins, I. Rousochatzakis, Q. Luo, HYK, in preparation

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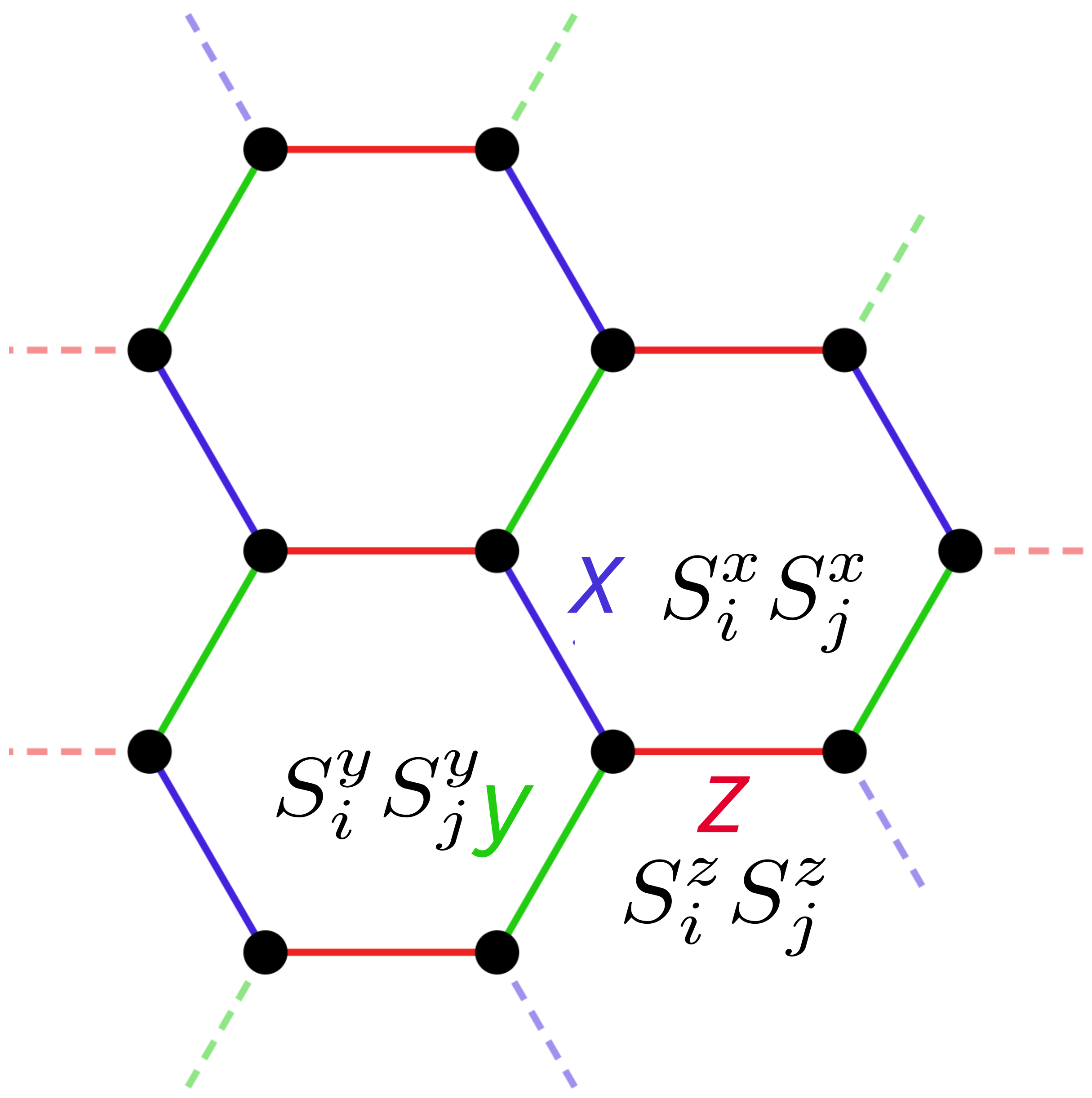
S= 1/2 Kitaev interaction & Kitaev spin liquid

Kitaev Exchange

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

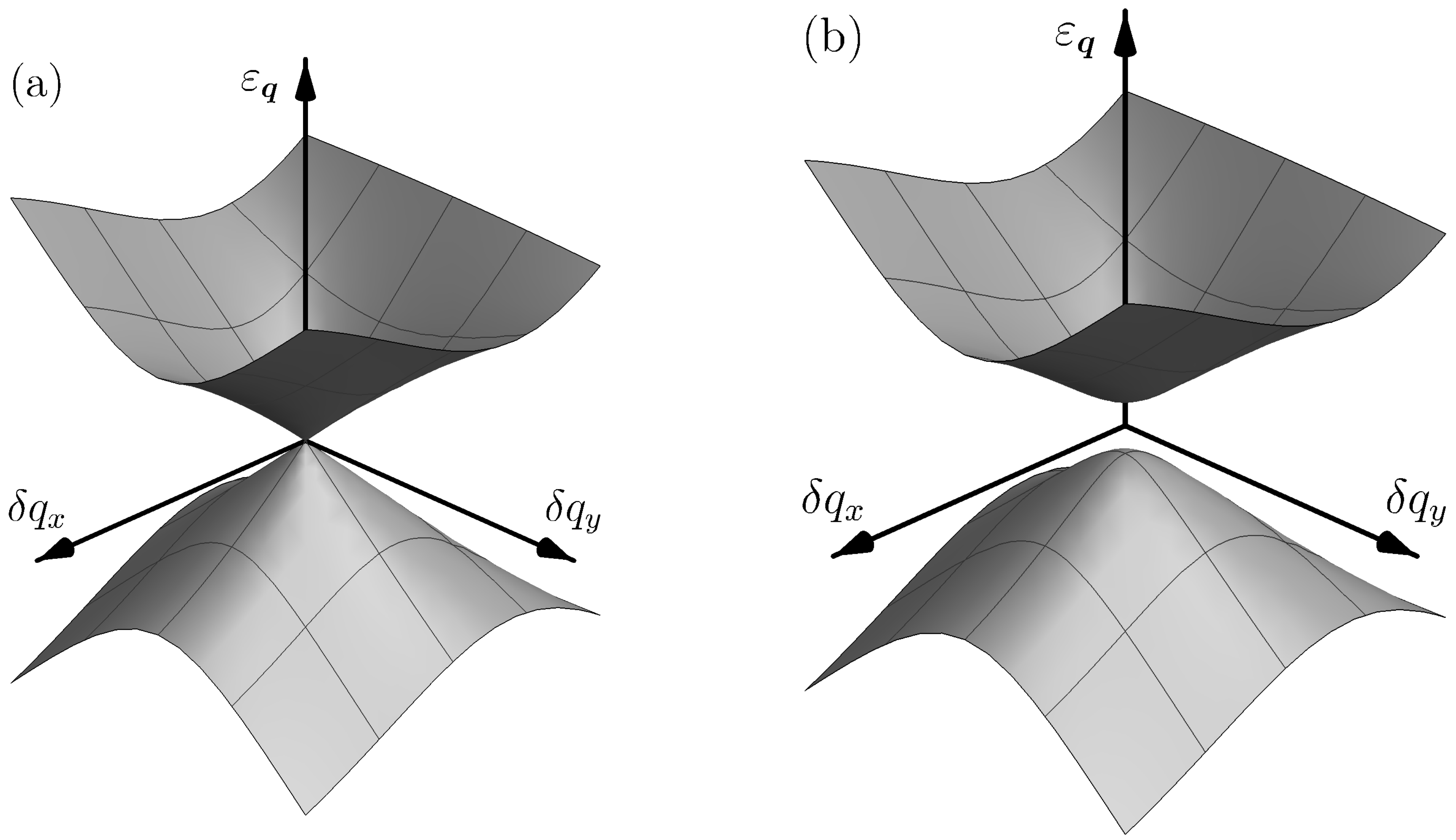
$$\boxed{\text{where} \quad \gamma = x, y, z}$$

bond-dependent Ising interaction



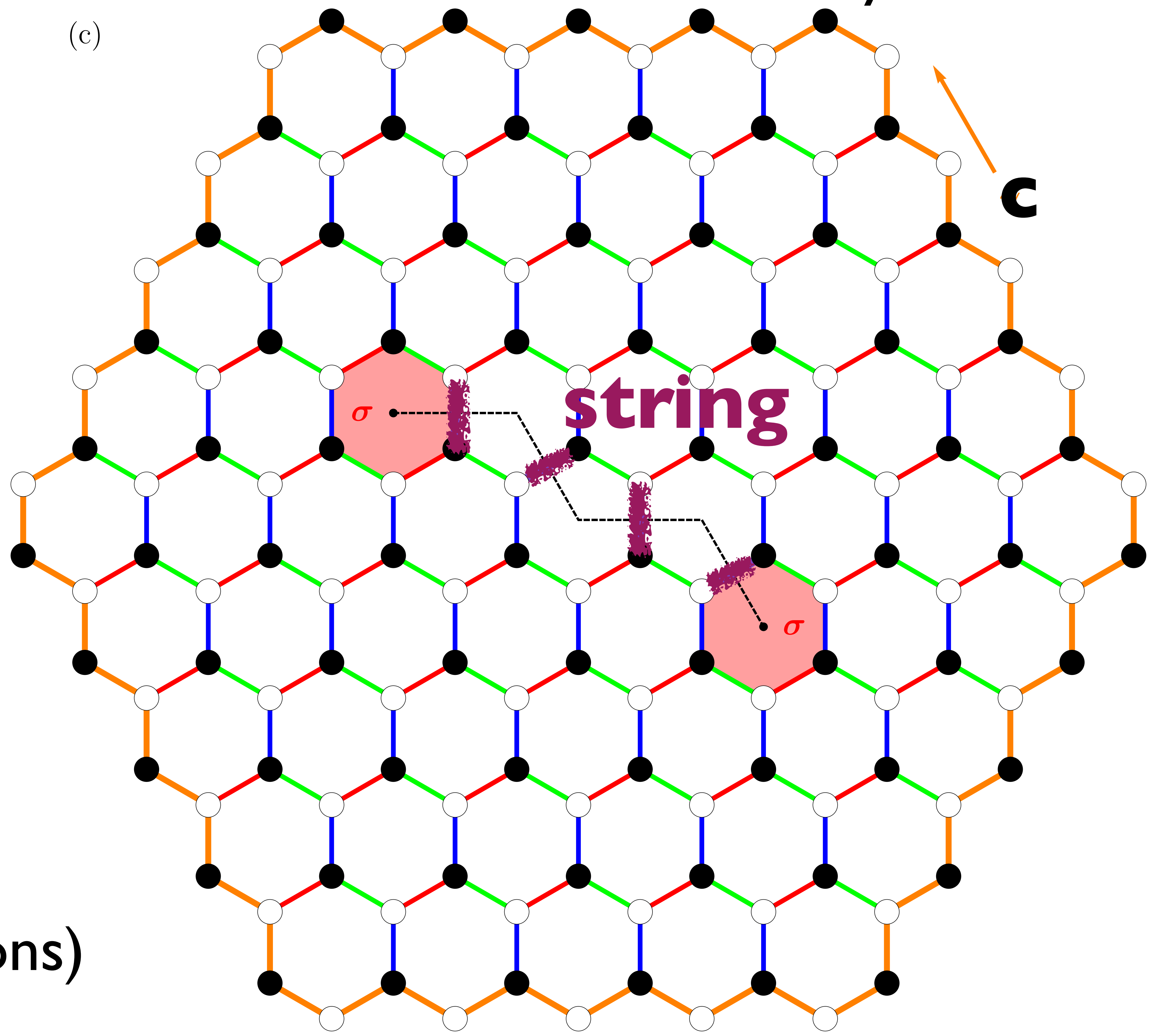
Kitaev spin liquid: emergent particles - Majorana fermion and vortices

A. Kitaev, Annals of Physics 321, 2 (2006)



under magnetic field

unpaired Majorana:
carry heat



Example: Ising Topological Order (Z2 vortex & Fermions)

$\psi \times \psi = 1 \qquad \psi \times \sigma = \sigma \qquad \boxed{\sigma} \times \sigma = 1 + \psi$

vortex carrying unpaired MF

Spin Interaction: low energy model of t + U

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

SU(2): U and single-orbital

When orbital & spin are not conserved via SOC

$$H_{ij} = \begin{pmatrix} S_i^x & S_i^y & S_i^z \end{pmatrix} \begin{pmatrix} J_{xx} & J_{xy} & J_{xz} \\ J_{yx} & J_{yy} & J_{yz} \\ J_{zx} & J_{zy} & J_{zz} \end{pmatrix} \begin{pmatrix} S_j^x \\ S_j^y \\ S_j^z \end{pmatrix}$$

: symmetry constrains matrix elements

insufficient to understand why one term is larger over other

Kitaev Materials

: Kitaev interaction is the largest interaction in full H

Necessary (not sufficient) Requirements

- honeycomb Mott insulator : strong e-e interaction
  transition metals

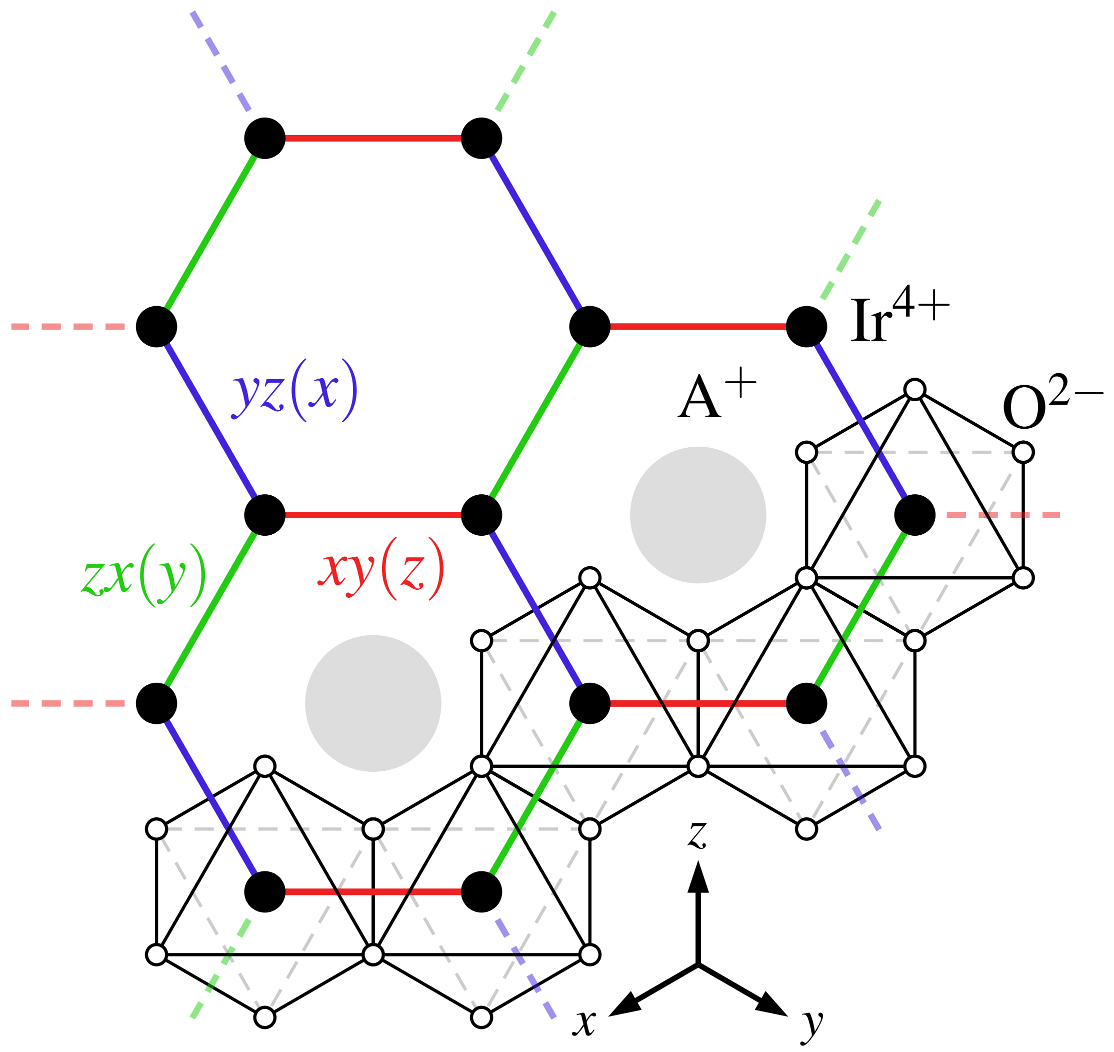
- bond-dependent spin interaction for $S=1/2$
  multi-orbital systems with Hund's coupling
  spin-orbit coupling

G. Khaliullin on triangular lattice (2005);
G. Jackeli, G. Khaliullin, Phys. Rev. Lett. 102, 017205 (2009)

Generic Spin Model in 2D honeycomb

nearest neighbour:
ideal honeycomb

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



another bond-dep. interaction

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

+ other interactions allowed by the symmetry

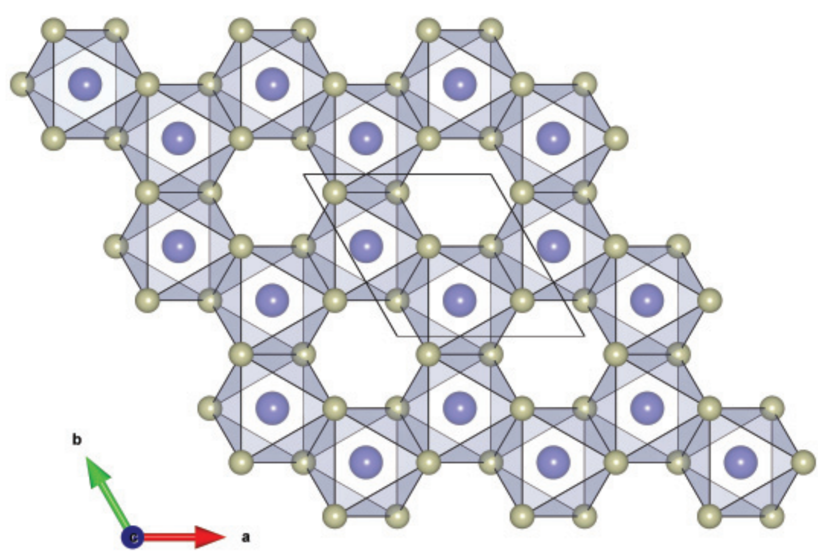
$$H^x = H^z (x \to y \to z \to x)$$

J. Rau, E. Lee, HYK, PRL 112, 077204 (2014)

other ref. - V. Katukuri, et al, New J. Phys. 16, 013056 (2014); Y. Yamaji, et al, PRL 113, 107201 (2014)

Kitaev interaction in Candidates with Jeff=1/2 (doublet)

Ti	V	Cr	Mn	Fe	Co	Ni
Zr	Nb	Mo	Tc	Ru	Rh	Pd
Hf	Ta	W	Re	Os	Ir	Pt

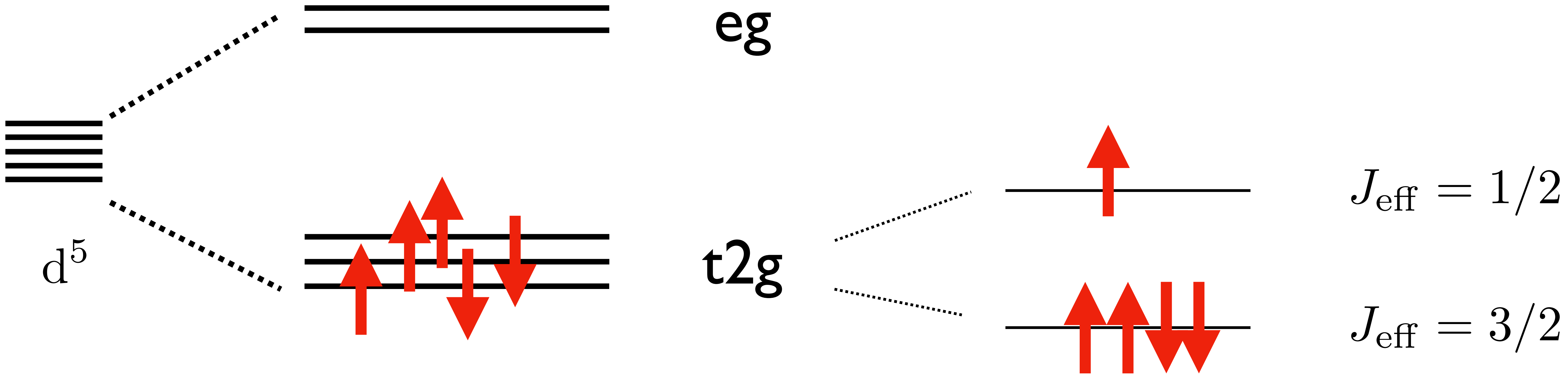


alpha-RuCl3

A2IrO3 (A = Na, Li): Y. Singh, et al, PRB 82, 064412 (2010);…

alpha-RuCl3 : K. Plumb, et al, PRB 90 041112(R) (2014); …

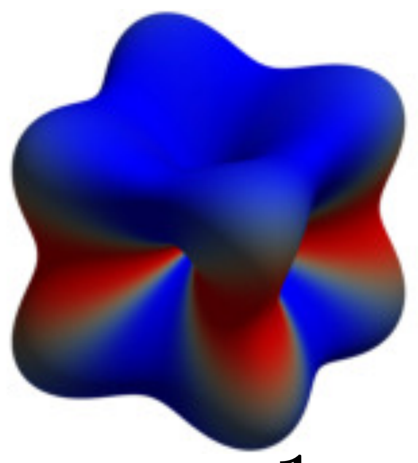
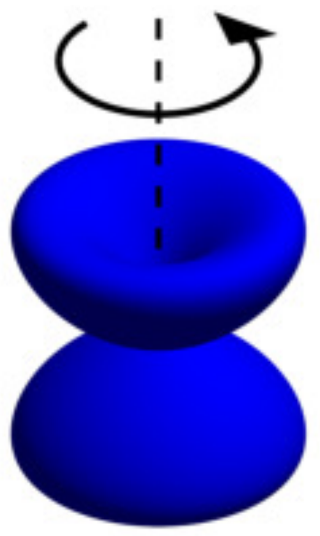
AIrO3 (A = Mg, Zn): Y. Haraguchi, et al, PRM 2, 054411 (2018),….



Due to strong spin-orbit coupling:
mixture of different orbitals and different spins

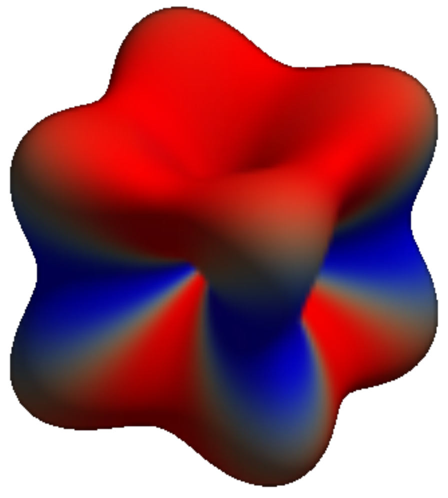
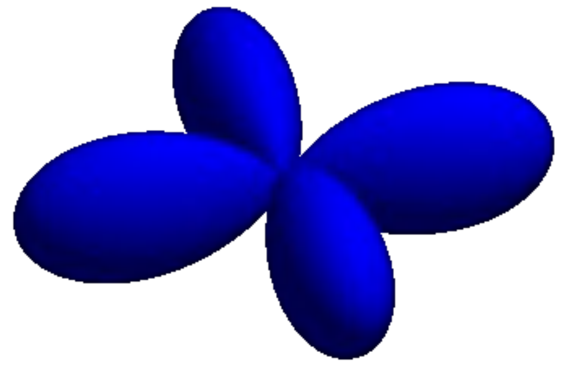
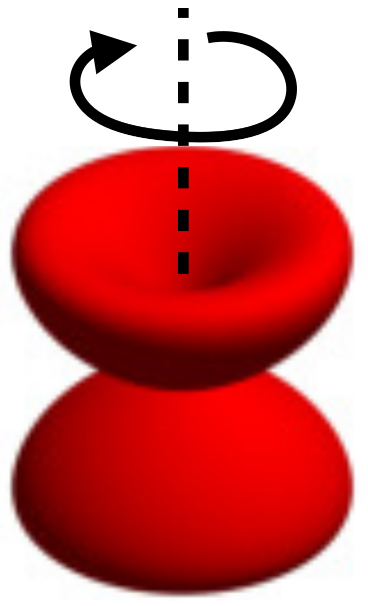
J_{eff} = 1/2 basis

spin down — spin up

 $= +\frac{1}{\sqrt{3}}$  $-\sqrt{\frac{2}{3}}$ 

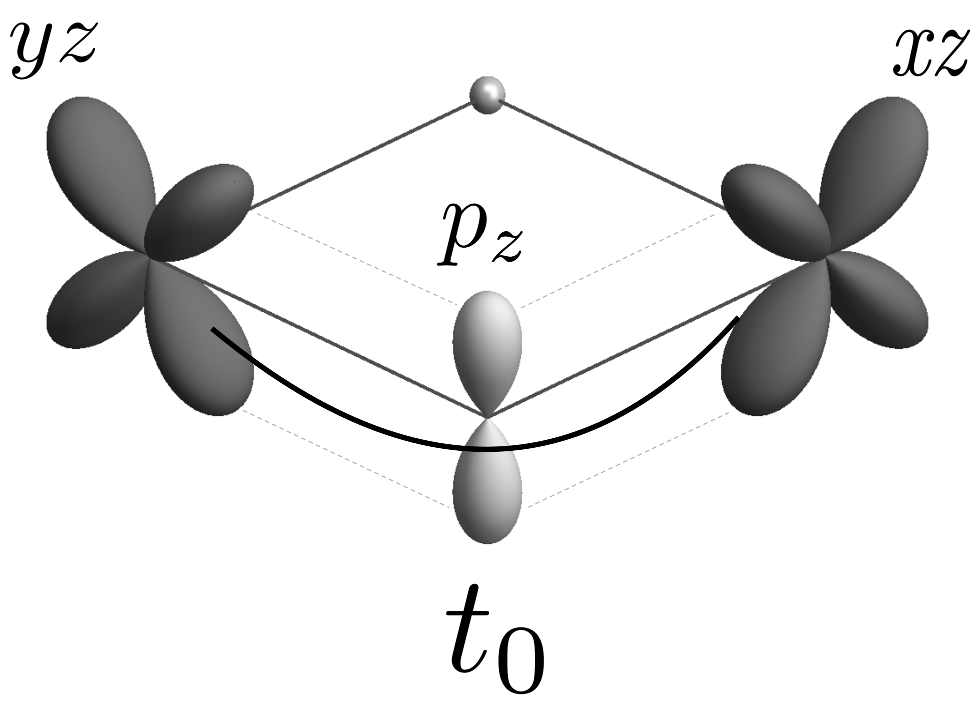
$J_z = \frac{1}{2}$ $L_z = 0$ $L_z = 1$

dxy **dxz + i dyz**

 $= \frac{1}{\sqrt{3}}$  $-\sqrt{\frac{2}{3}}$ 

$J_z = -\frac{1}{2}$ $L_z = 0$ $L_z = -1$

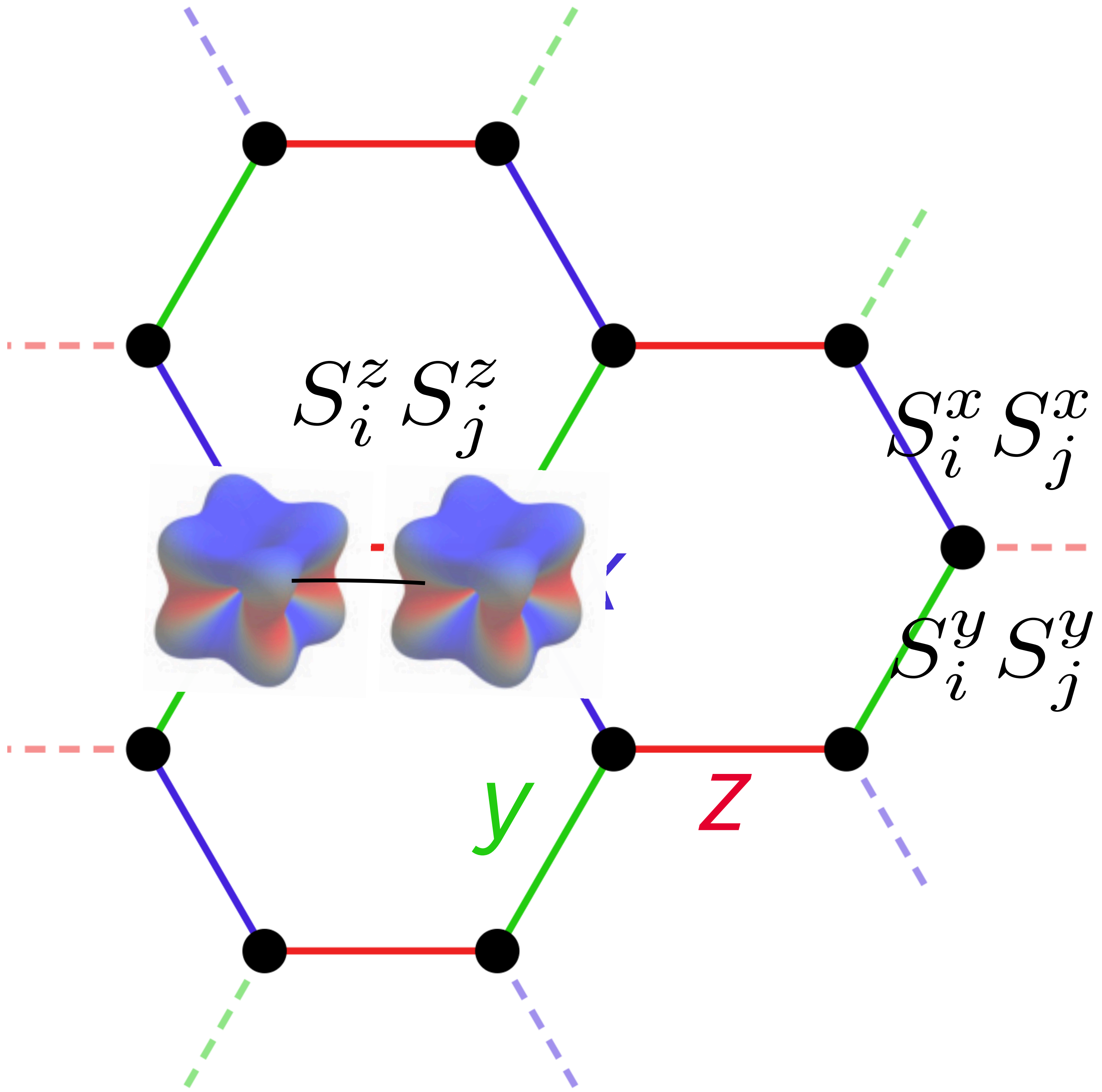
$$\frac{1}{\sqrt{3}}\left(|xy\rangle|\uparrow\rangle + |yz\rangle|\downarrow\rangle + i|xz\rangle|\downarrow\rangle\right)$$



$$\delta L^z = \pm 2 \qquad \delta J^z_{\text{eff}} = \pm 2$$

orbital non-conserving hopping

~~Heisenberg term~~



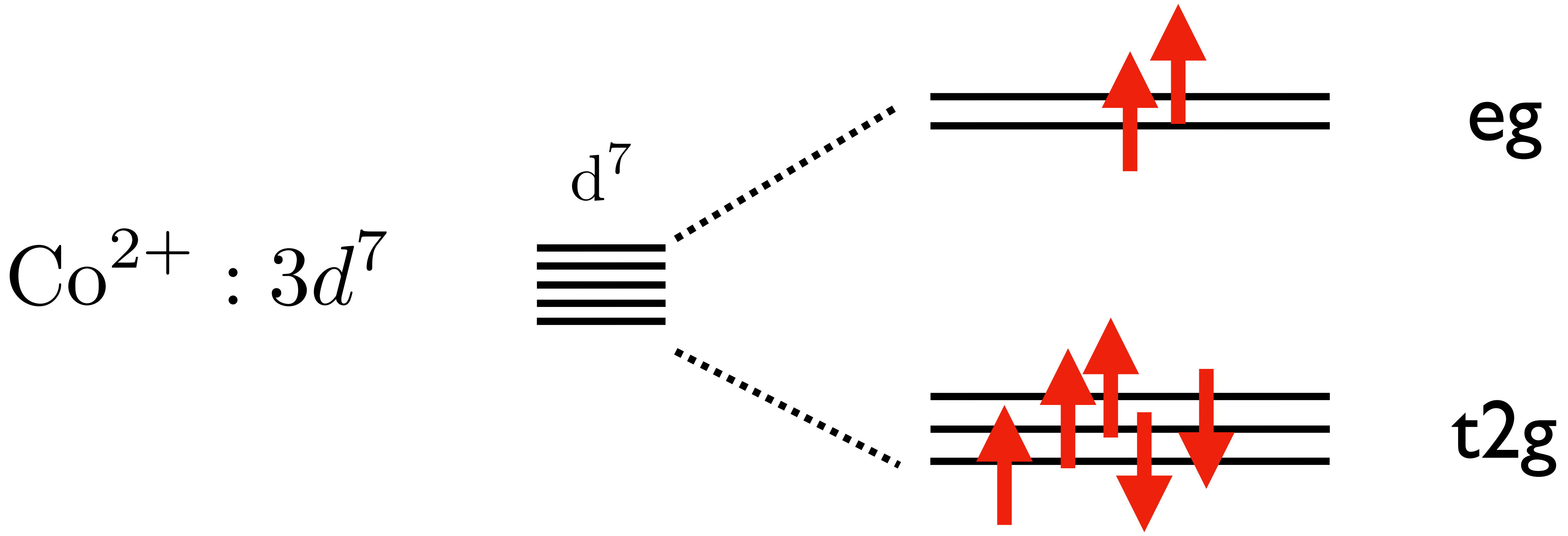
$$H_K = K S_i^z S_j^z$$

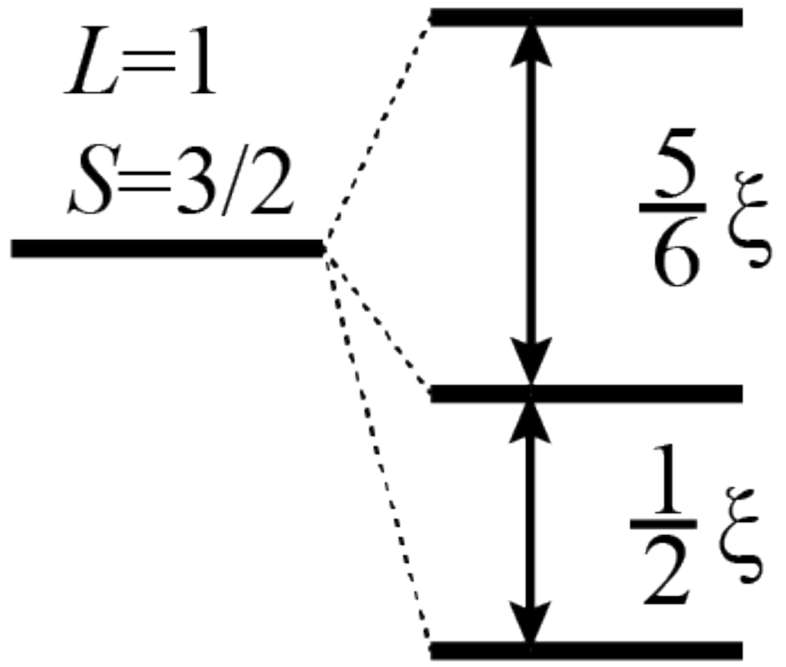
G. Jackeli, G. Khaliullin, Phys. Rev. Lett. 102, 017205 (2009)

orbital conserving hopping: Heisenberg + ...

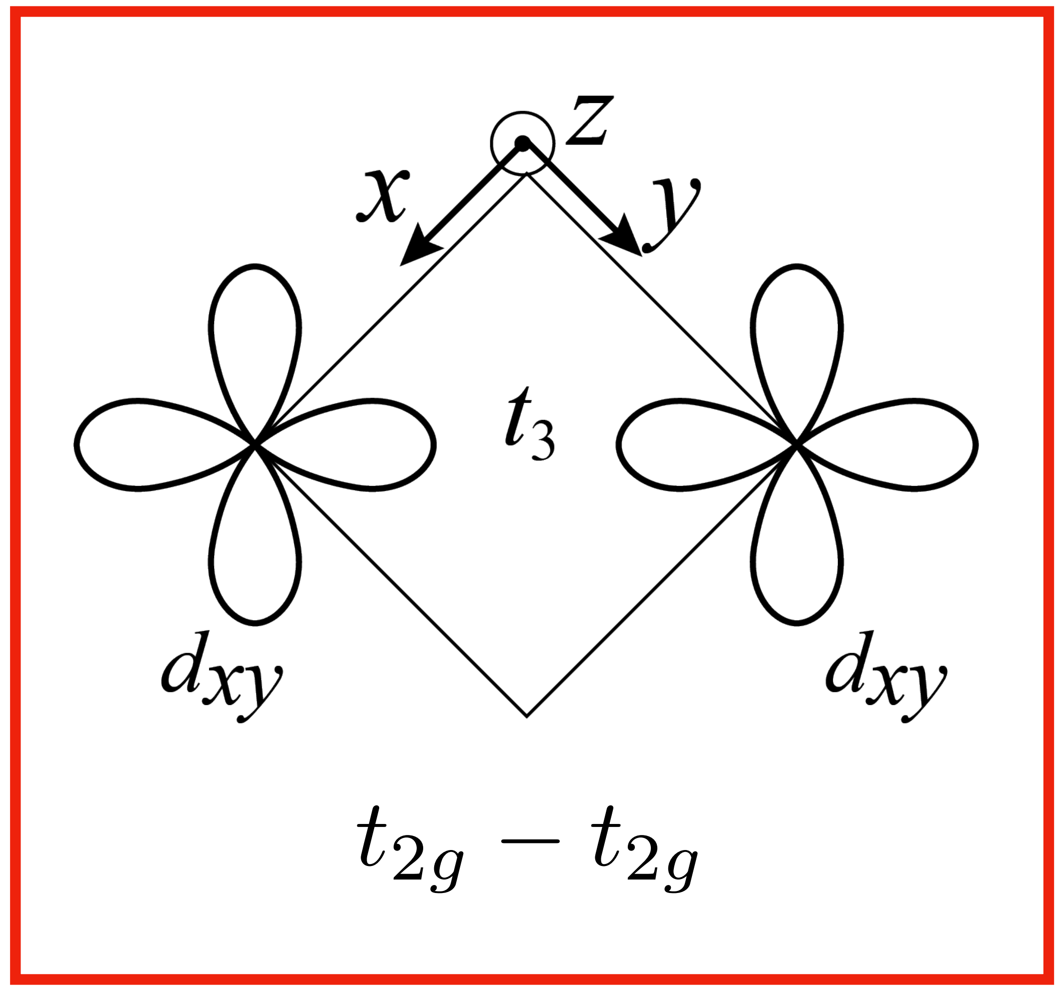
3d7: Jeff=1/2 honeycomb Cobaltates:

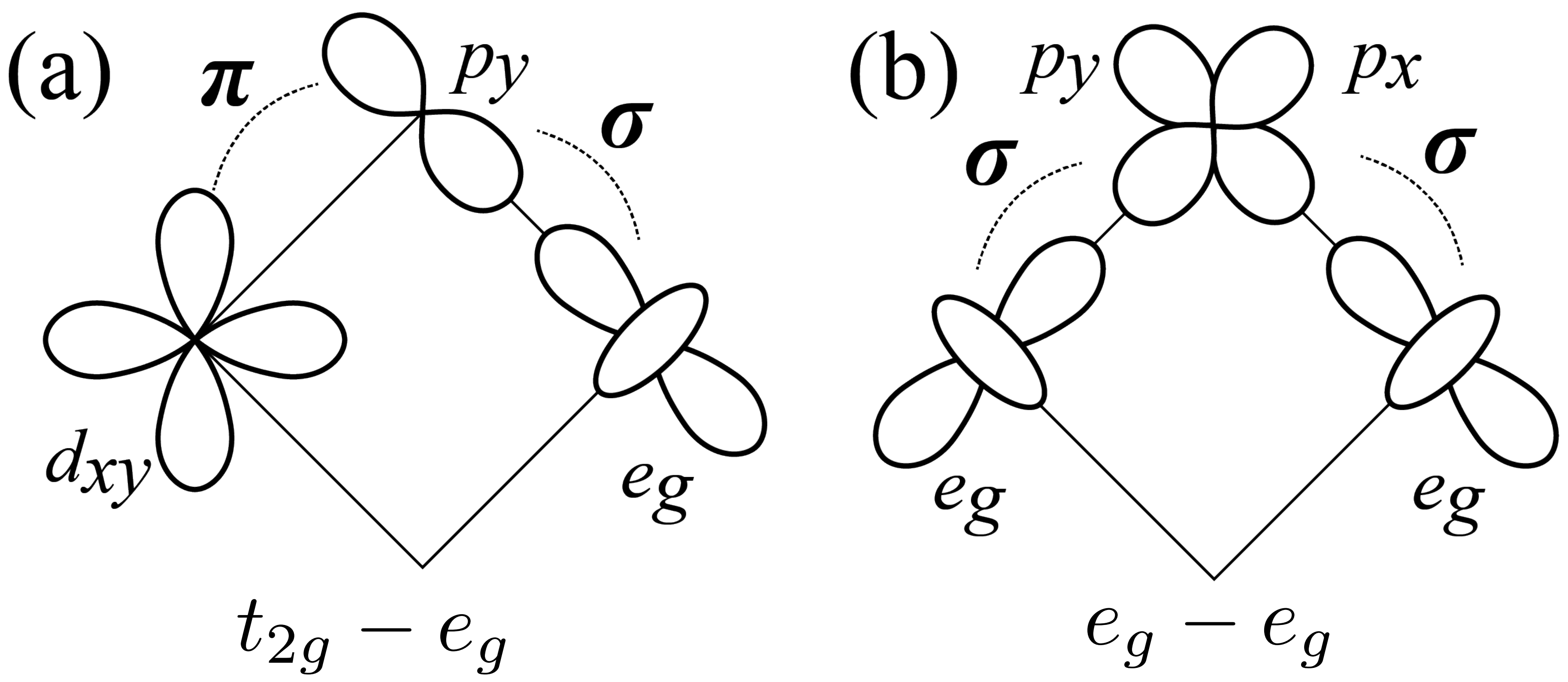
R. Sano et al, PRB 97, 014408 (2018), H. Liu, G. Khaliulin, PRB 97, 014407 (2018), ….





Debate over the strength of Kitaev interaction





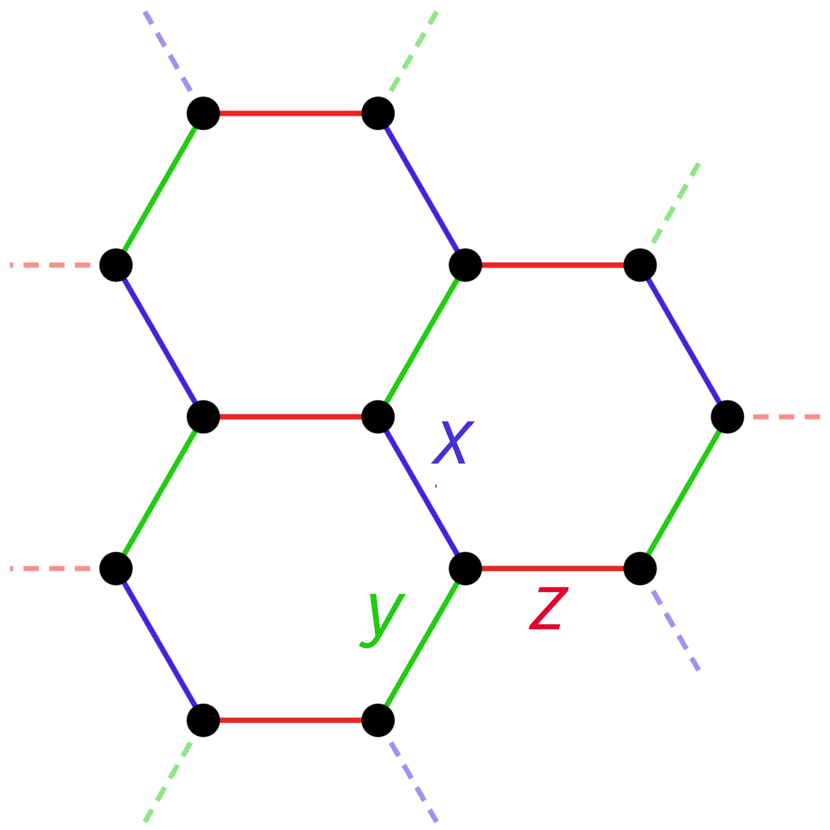
When dominated: Non-Kitaev, e.g., BaCo2(XO4)2, X=As, P : X. Liu, HYK, arXiv:2211.03737

Spin-S Kitaev

Kitaev Exchange

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

where $\gamma = x, y, z$



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 excitations?

half-integer vs. integer-S Kitaev?

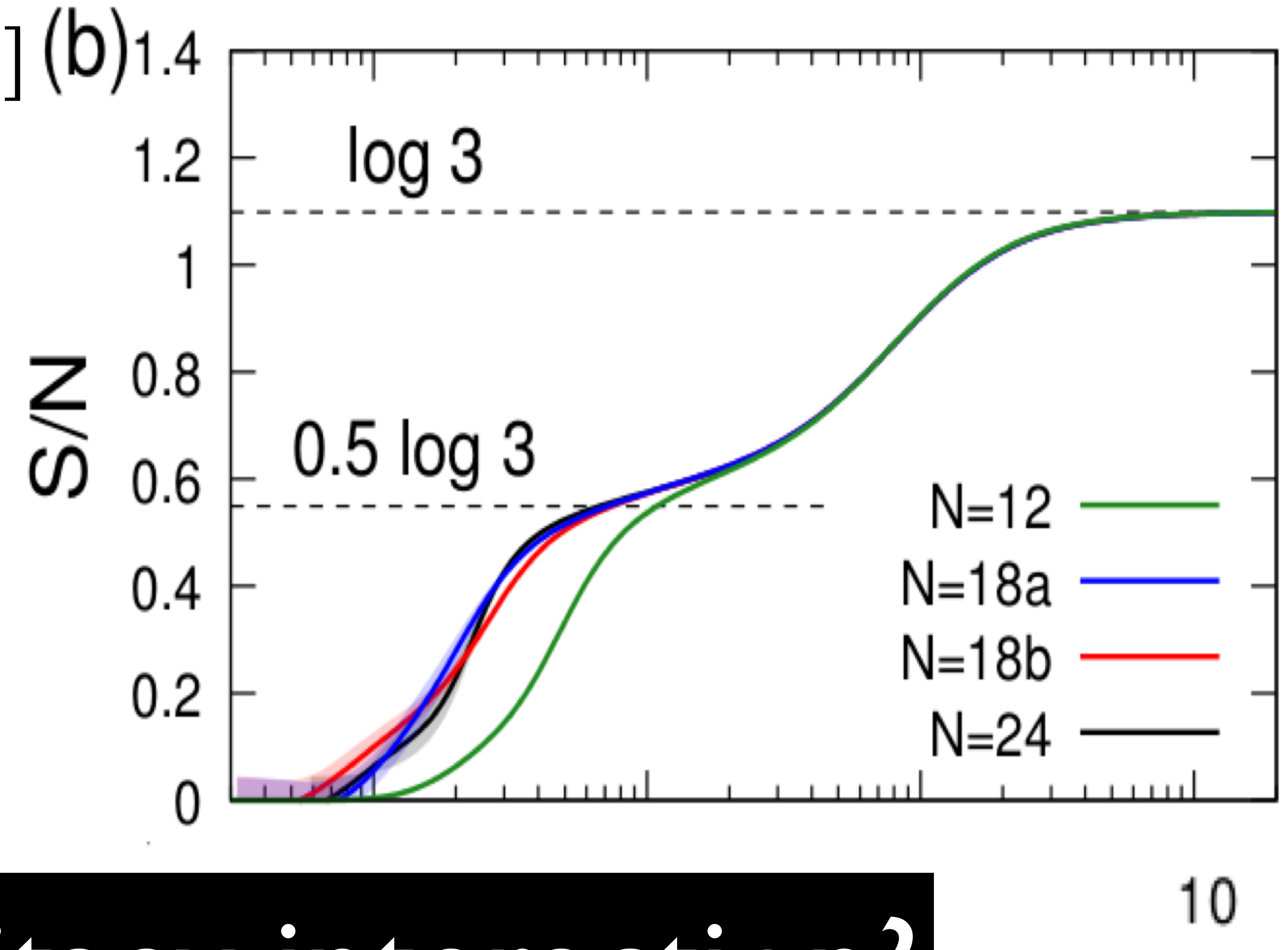
Spin S=1 Kitaev model in the literature.

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] →



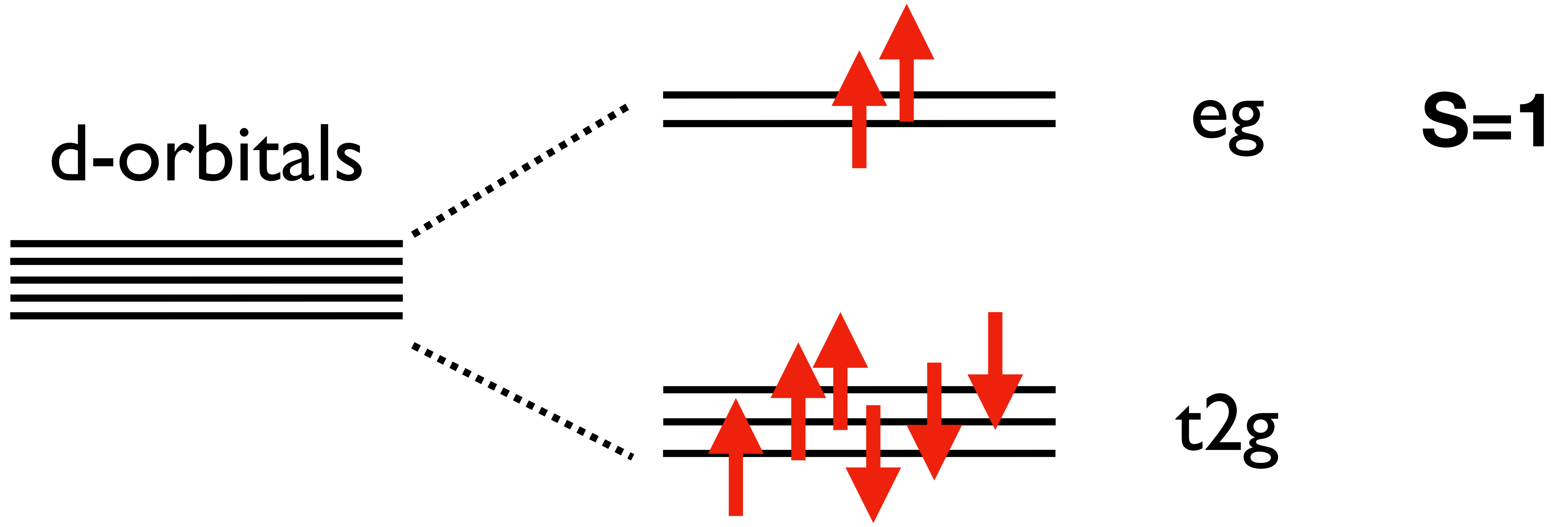
Q: How do we get S=1 Kitaev interaction?

[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).**

Kitaev interaction in honeycomb insulators with general S

Ti	V	Cr	Mn	Fe	Co	Ni
Zr	Nb	Mo	Tc	Ru	Rh	Pd
Hf	Ta	W	Re	Os	Ir	Pt

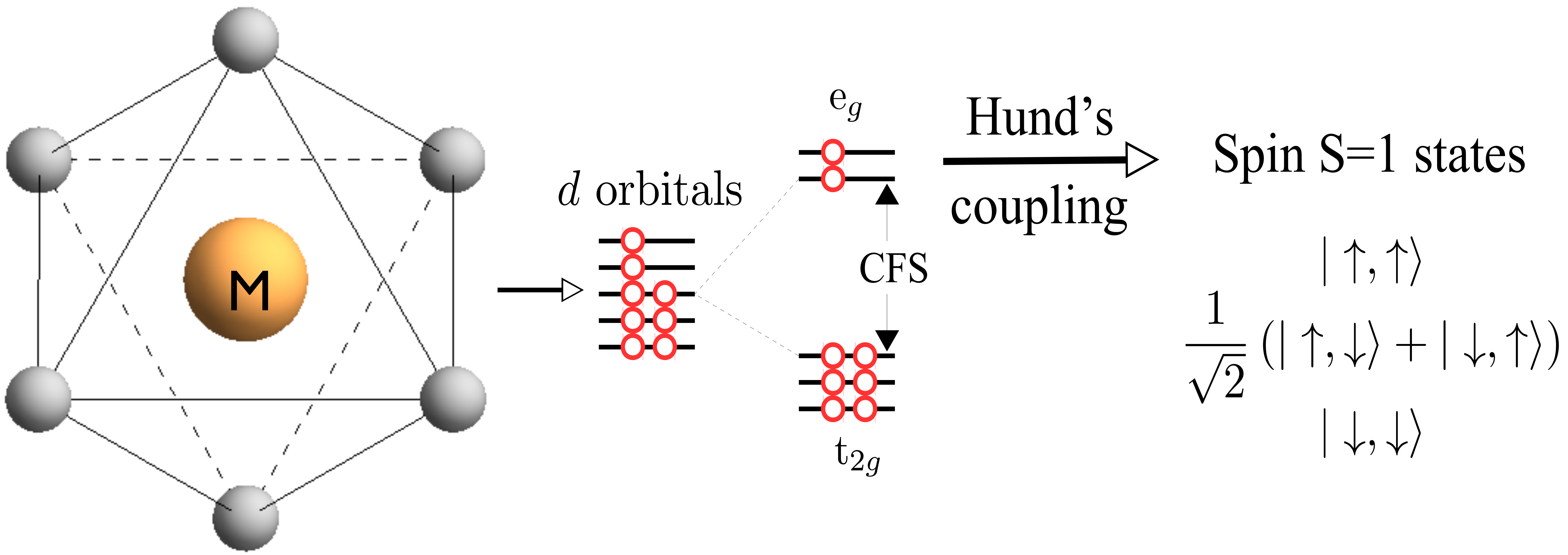
$\mathrm{Ni}^{2+} : 3d^8$



Higher-spin Kitaev interaction

P. Peter Stavropoulos, D. Pereira, HYK PRL 123, 037203 (2019)

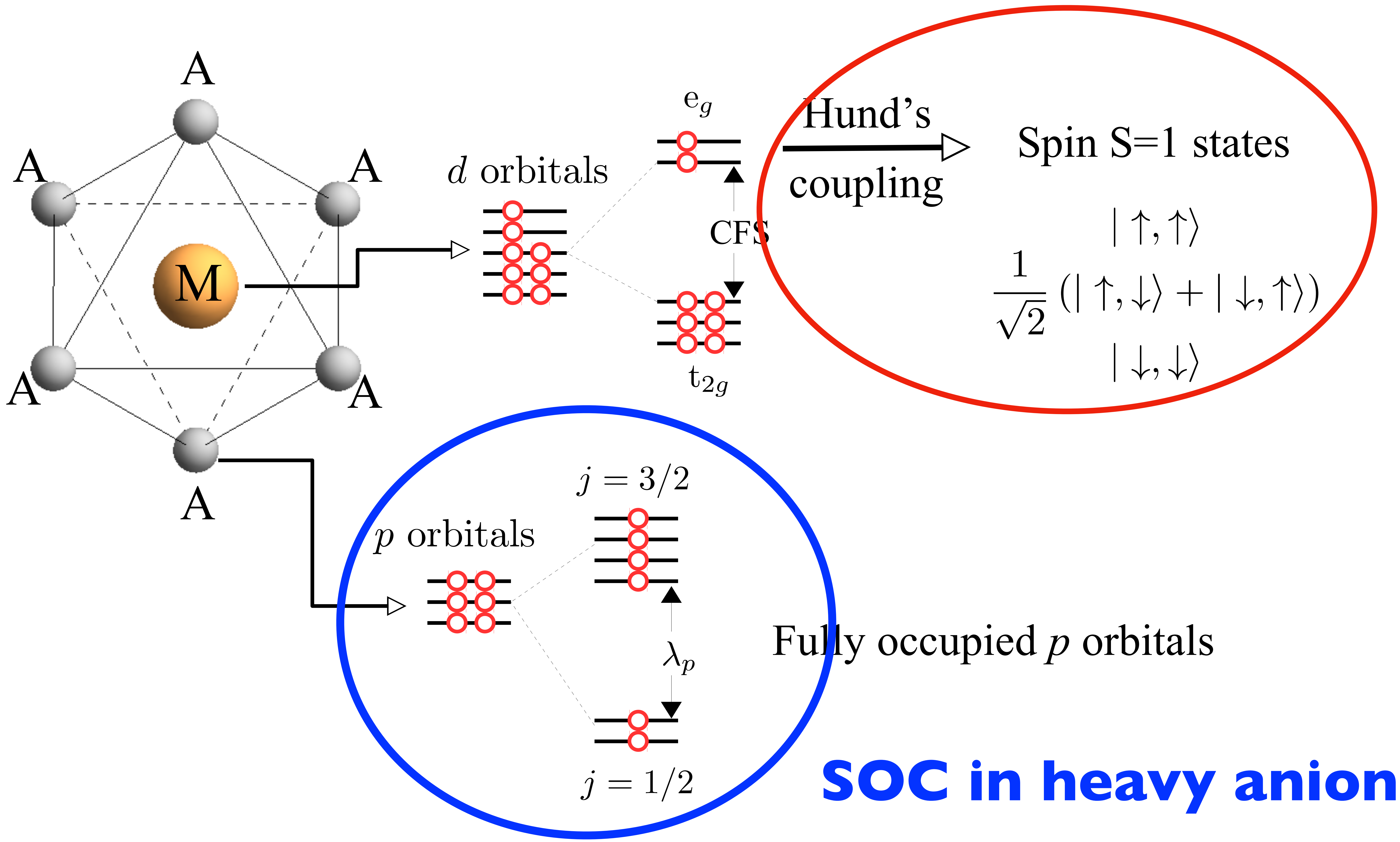
ex: spin one (d8)



Crystal field splitting > Hund's coupling >> spin-orbit coupling (SOC)

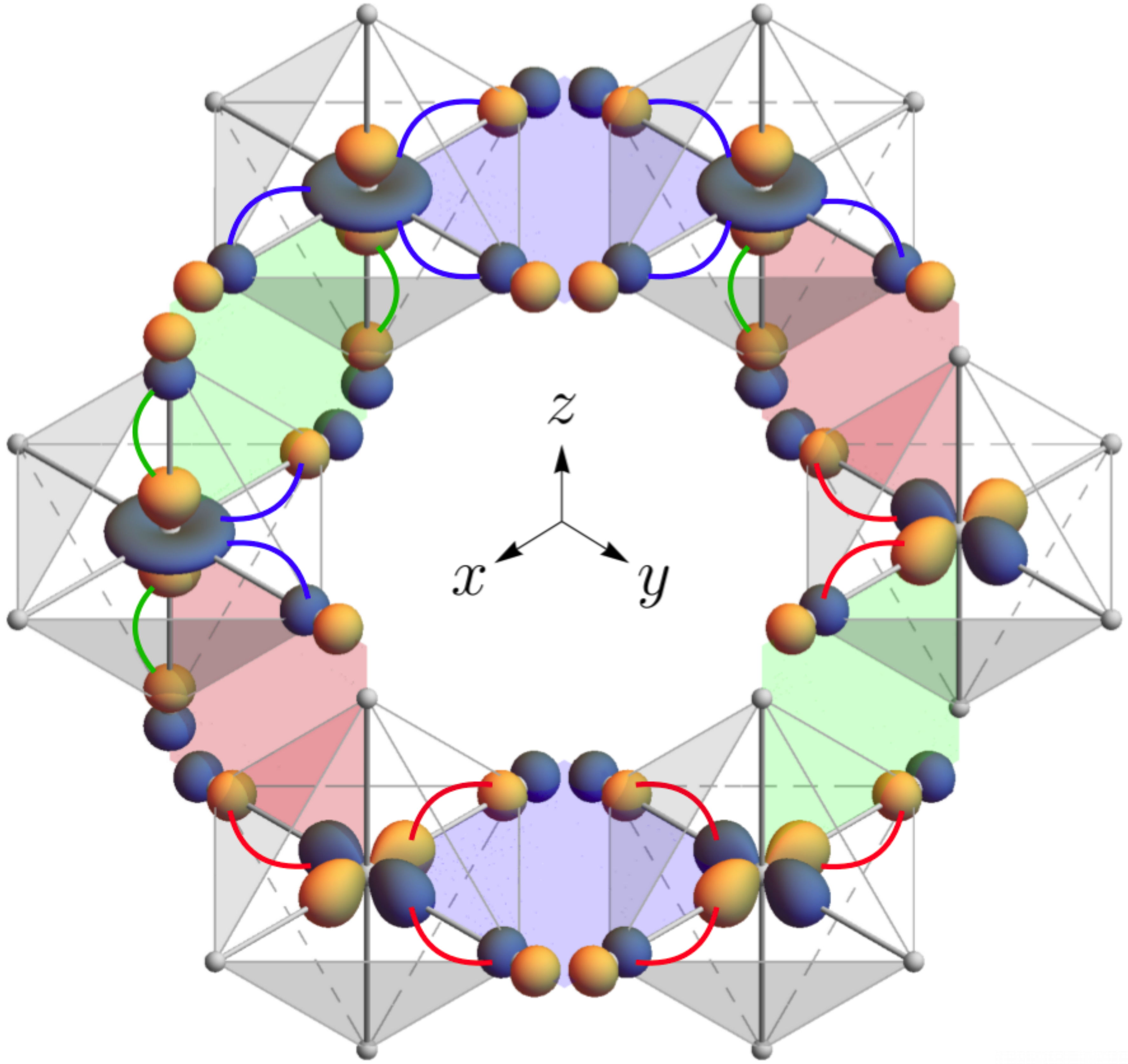
No mixture of different orbitals & spins - No Kitaev?

on-site H0 = Kanamori (U, U', Hund's) + SOC



SOC in heavy anions

perturbation theory: hopping between two M sites via heavy A sites



t_1 (red)

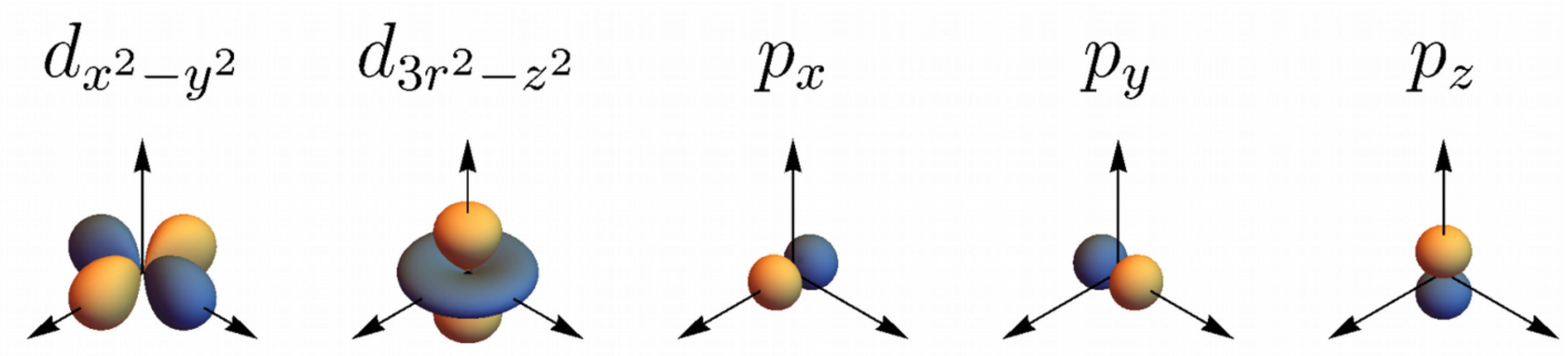
t_2 (blue)

t_3 (green)

Hopping integrals
M to A sites:

Cubic symmetry:

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



Indirect $K = -2J_{ind}$

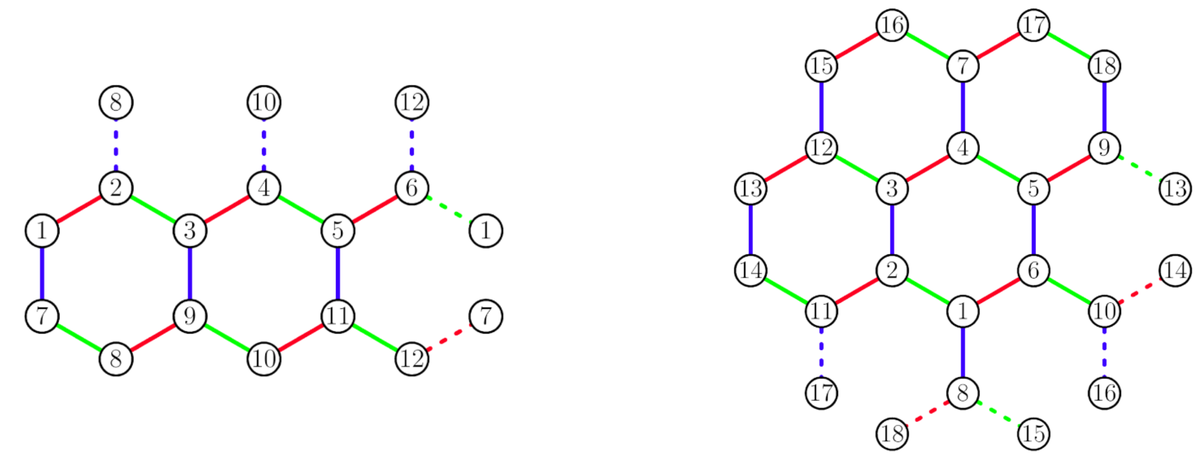
Direct $J_d = 4t^2/U$

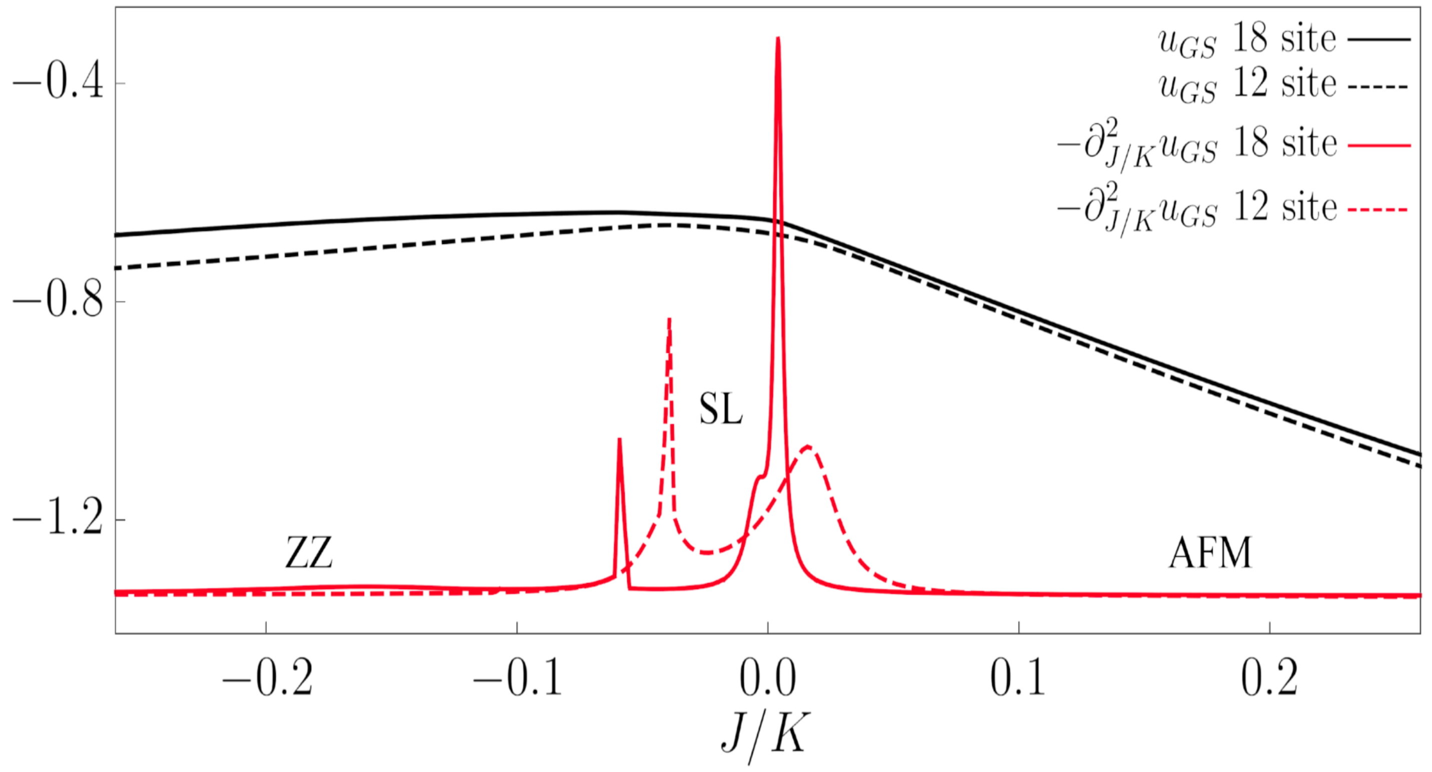
$\Gamma = 0$: up to 4th order

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

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

ED calculation: S=1 KJ model
12 & 18 sites



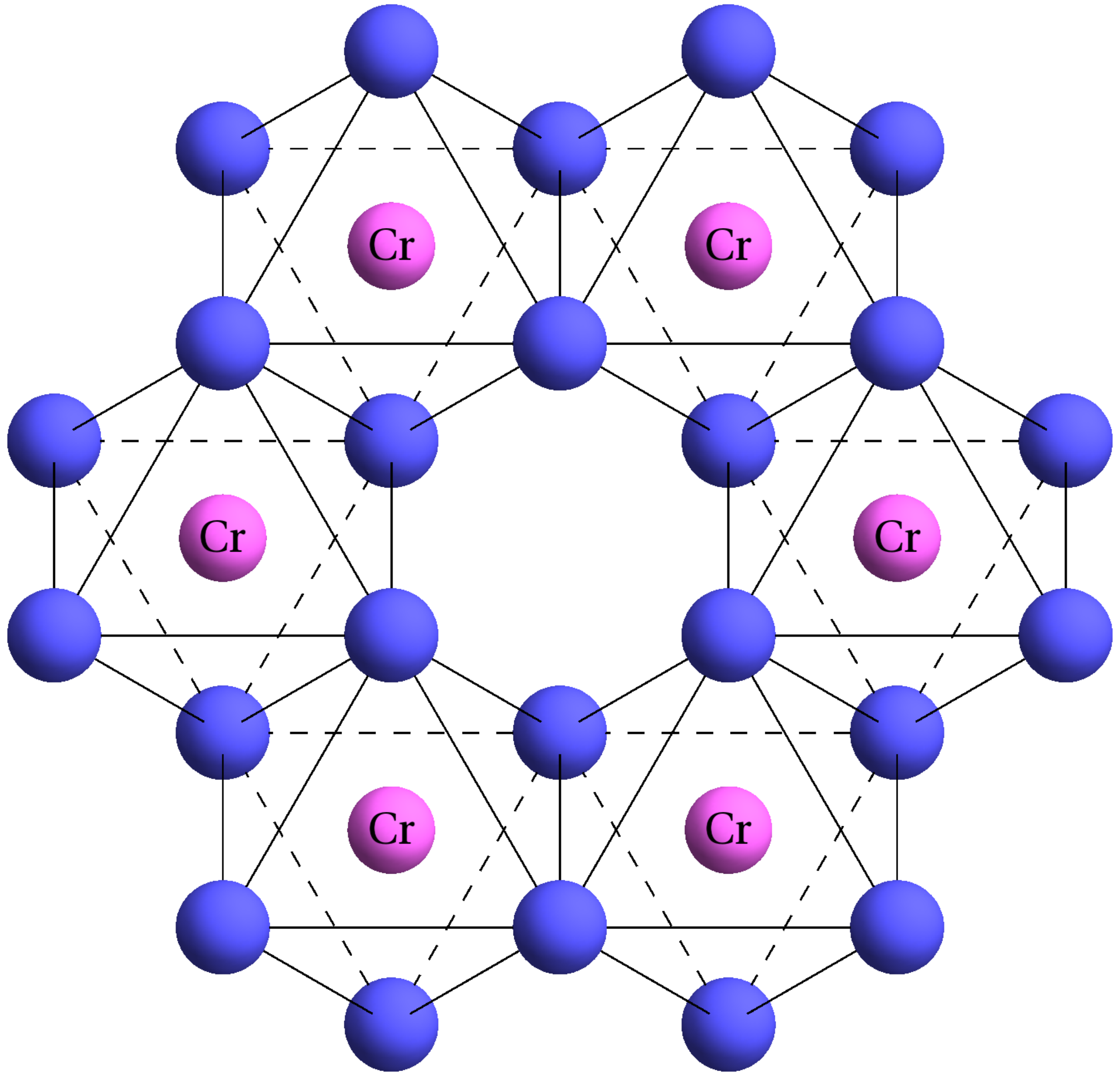


P. Peter Stavropoulos, D. Pereira, HYK, PRL 123, 037203 (2019)

Kitaev from heavy anion from ab-intio: C. Xu et al, npj Comput. Mater. 4, 57 (2018)

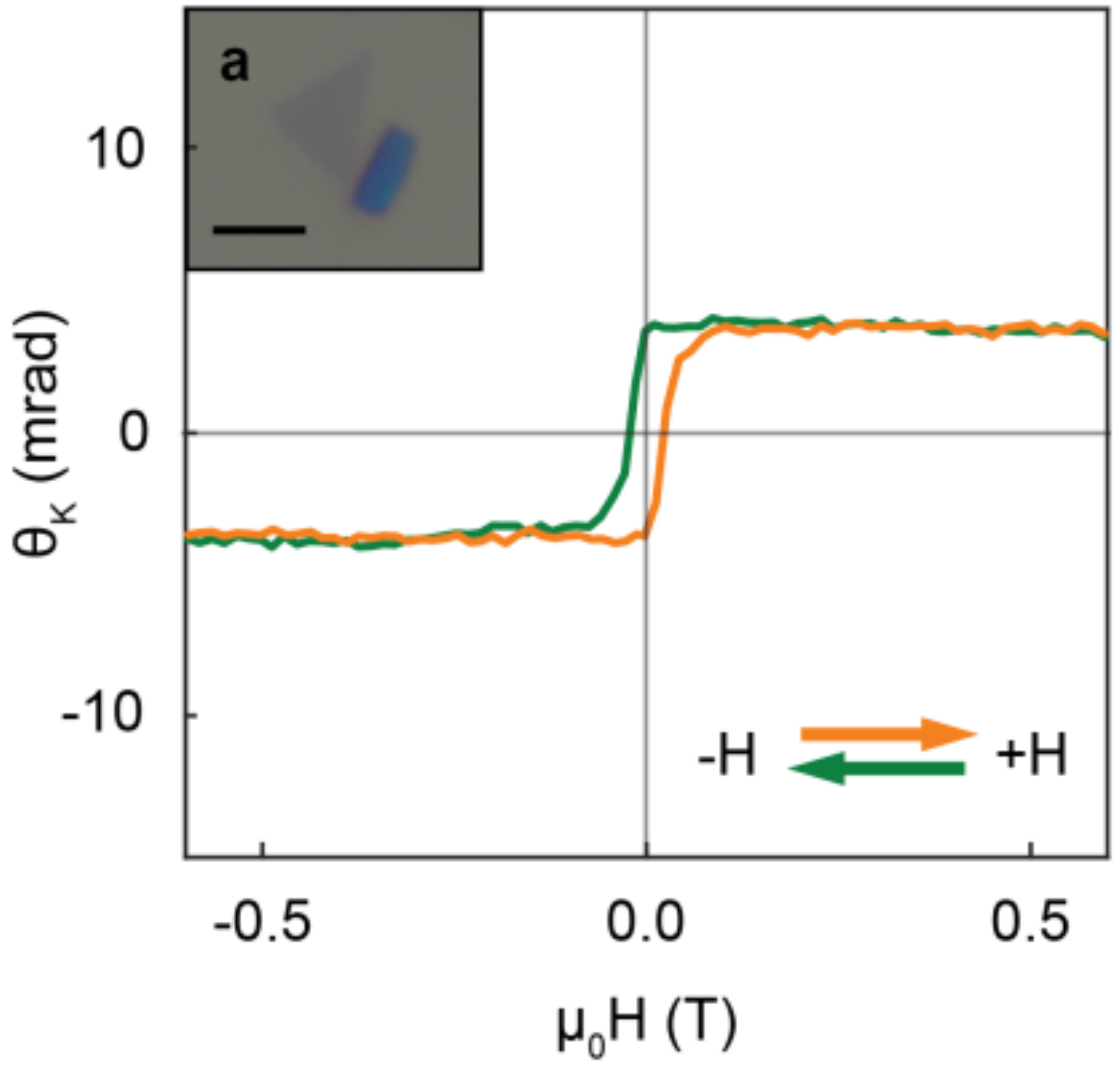
Example

Single layer of CrI3 (S=3/2 honeycomb):
Ferromagnetic insulator



Kitaev interaction?

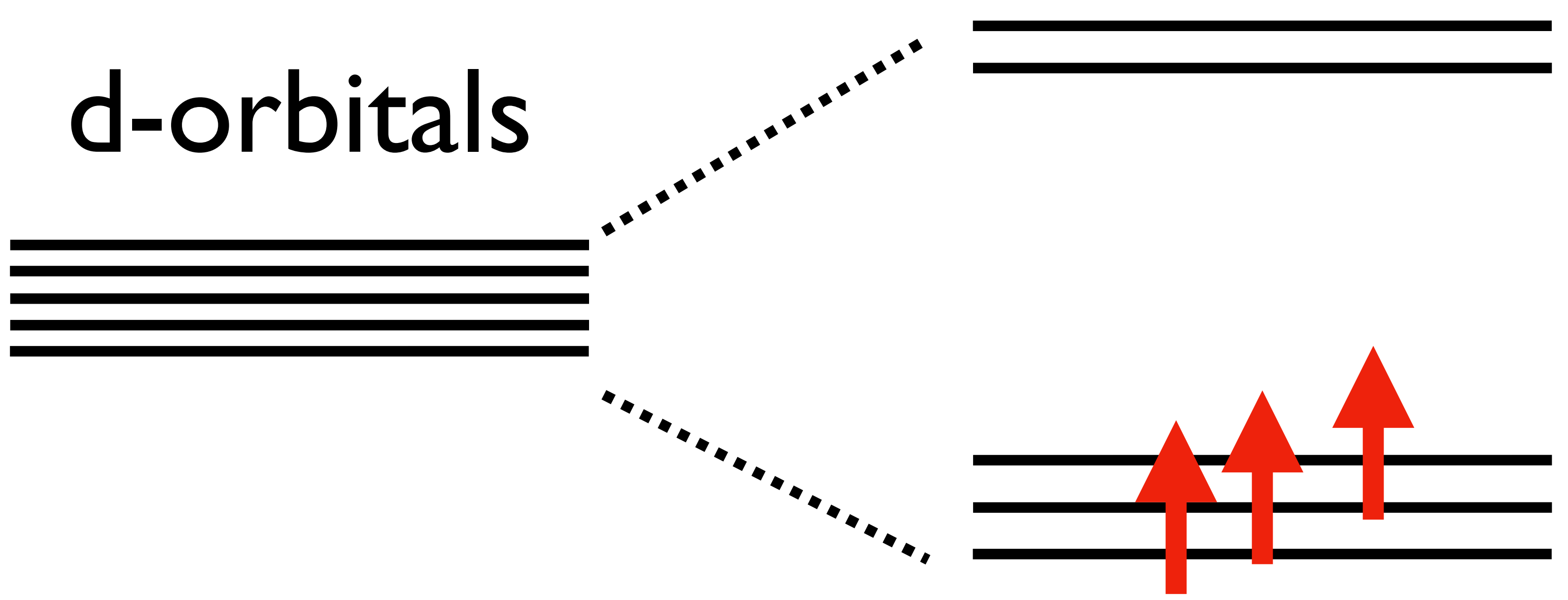
Kerr rotation



Single layer (2D) Ferromagnet

B. Huang et al, Nature 546, 7657 (2017)

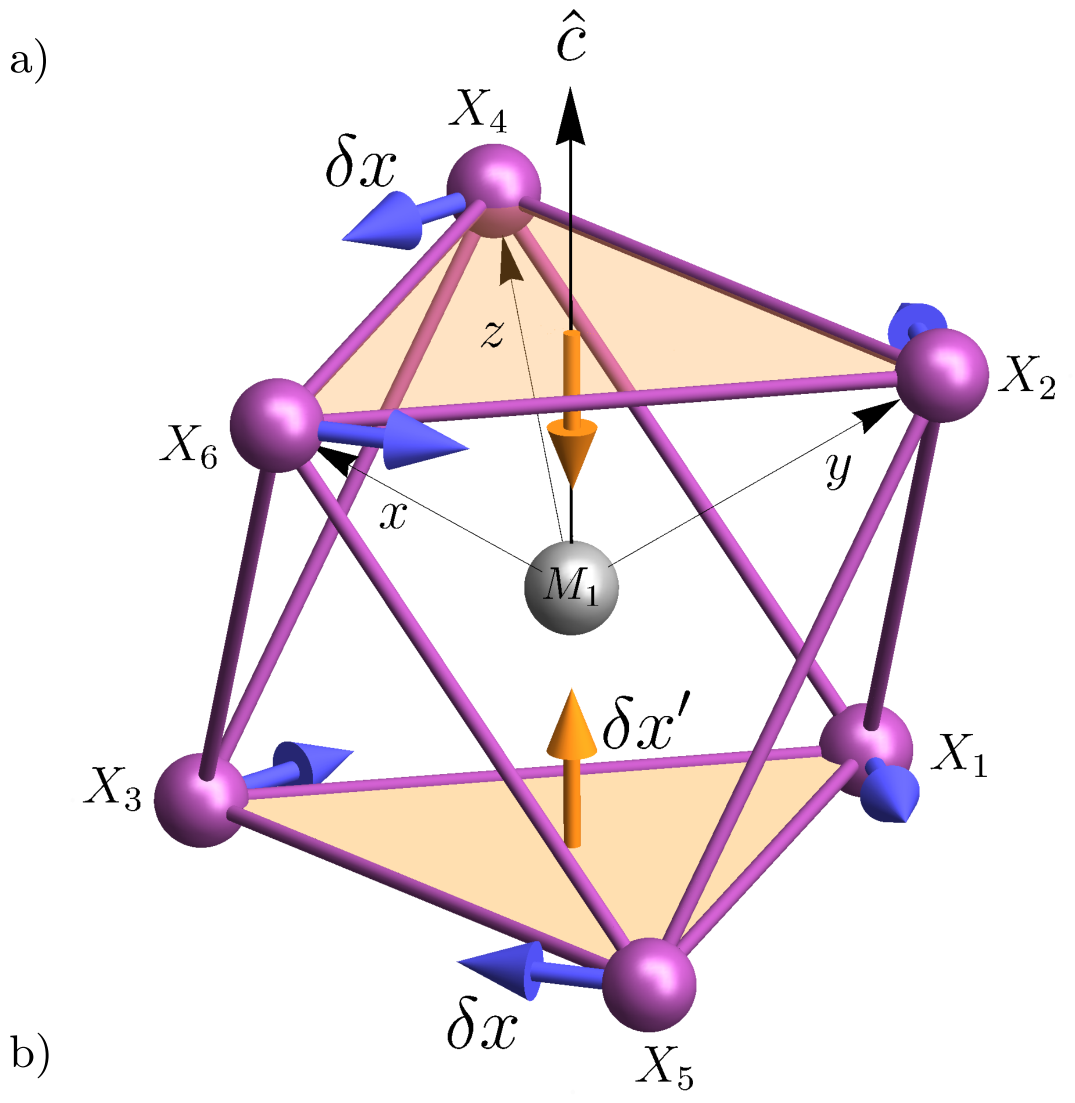
$Cr^{3+} : 3d^3$



S=3/2

n.n. model

$$H = \sum_{\langle ij \rangle \in \alpha\beta(\gamma)} \Big[J\mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \Gamma'(S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta) \Big] + \sum_i A_c(\mathbf{S}_i \cdot \hat{\mathbf{c}})^2;$$



trigonal distortion

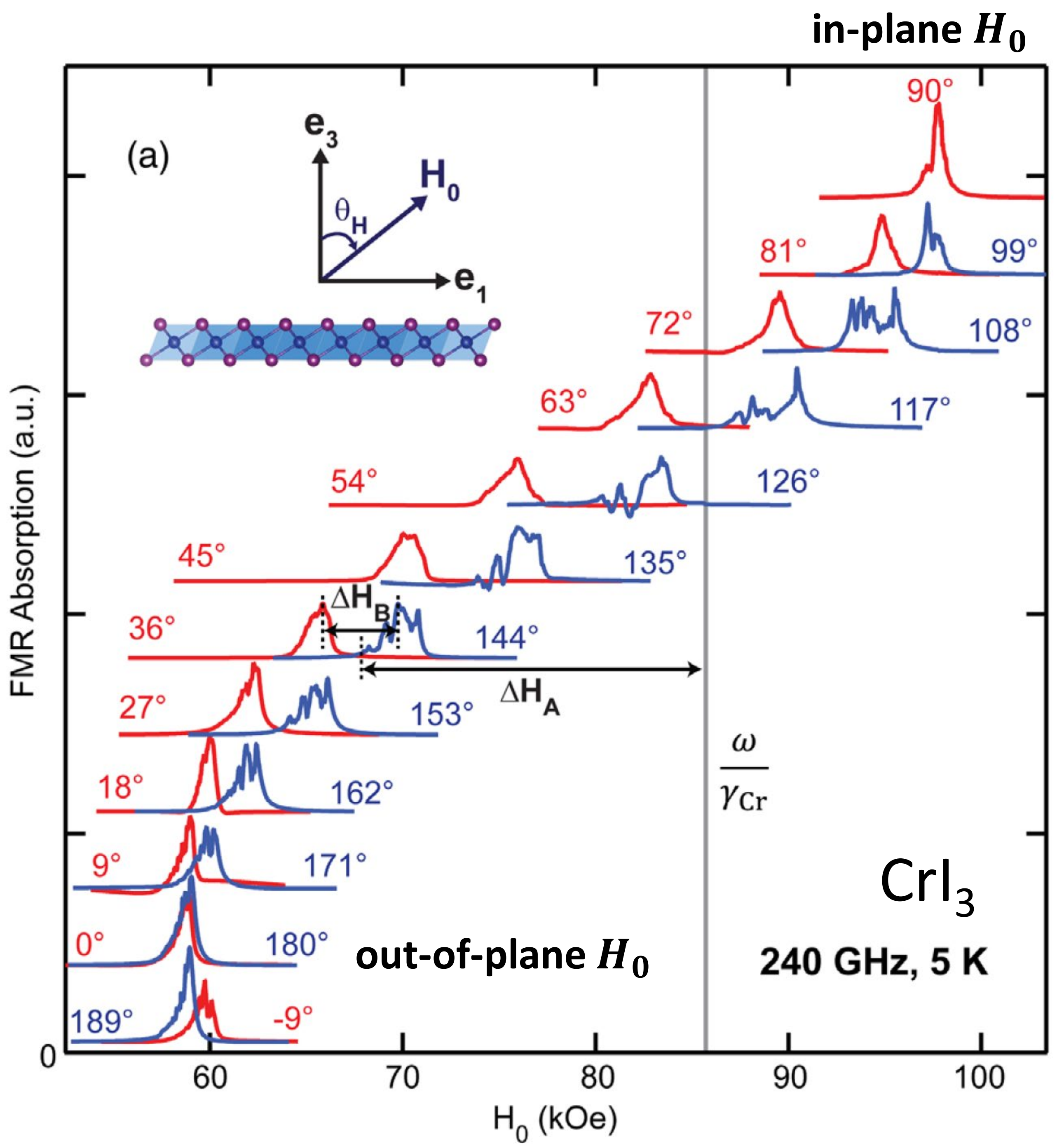
single-ion anisotropy, Γ, Γ'

from SOC + distortion

P. P. Stavropoulos, Liu, HYK, PRR3, 013216 (2021)

CrI3 : debate

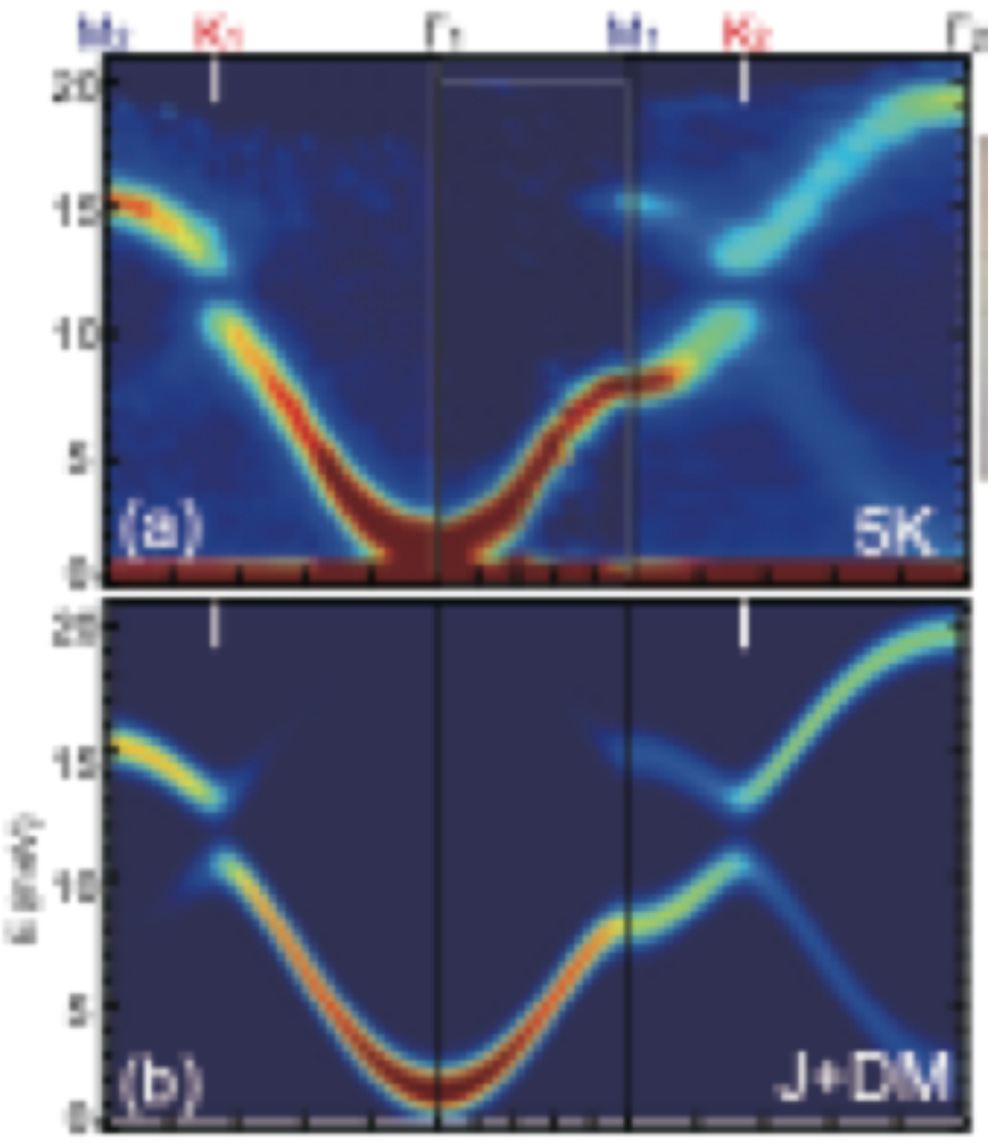
Angle dependent Ferromagnetic Resonance (FMR): CrI3



Kitaev dominant

Inhee Lee, et al, Phys. Rev. Lett. 124, 017201 (2020)

Inelastic neutron scattering



J + DM interaction

L. Chen et al, PRX 11, 031047 (2021)

Kitaev Materials

: Kitaev interaction is the largest interaction in full H

Necessary (not sufficient) Requirements

- honeycomb Mott insulator : strong e-e interaction

Debate over the strength of Kitaev interaction

- bond-dependent spin interaction for S=1/2

multi-orbital systems with Hund's coupling

spin-orbit coupling

S > 1/2 : spin-orbit coupling in heavy ligand + Hund's coupling in transition metal

Goal: how to determine the Kitaev strength?

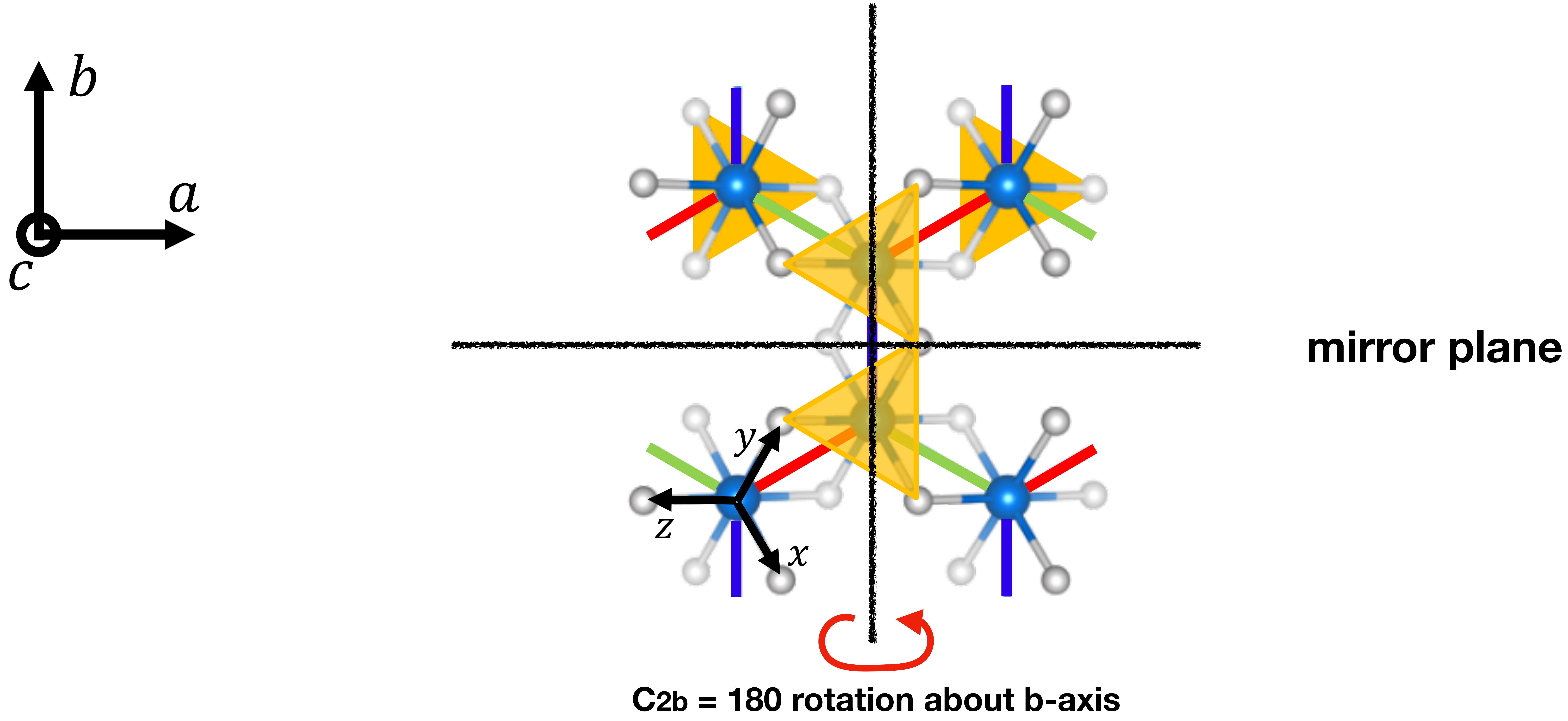
Use Symmetry of full Hamiltonian

Jiefu Cen, HYK, Commun. Phys. 5, 119 (2022);

Jiefu Cen, HYK, arXiv:2208.13807 (2022)

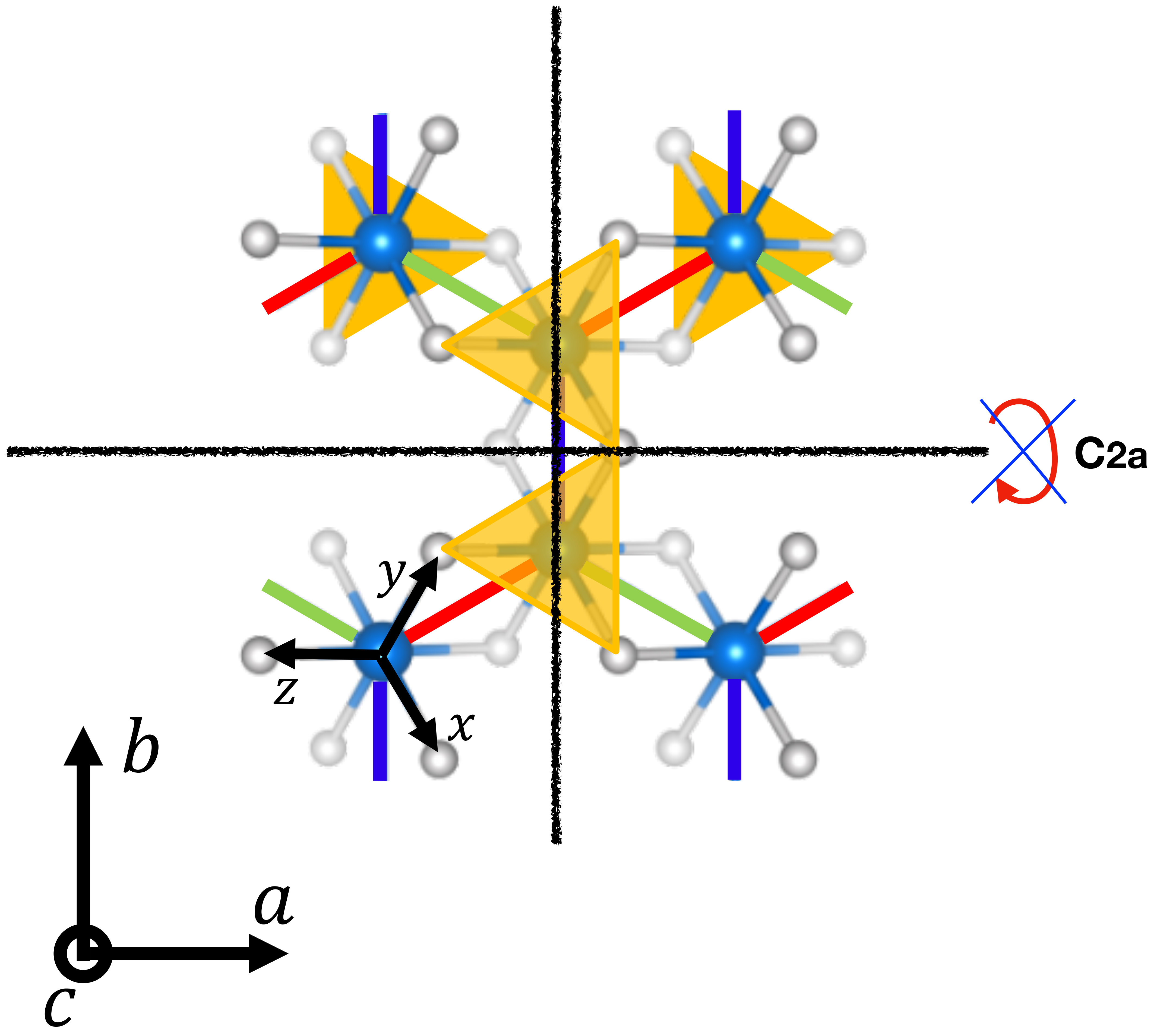
Generic n.n. ideal octahedra model

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



b is parallel to the bond; a/c-axis are perp. to the bond/plane

NO mirror plane (bc plane)



C2a= 180 rotation about a-axis

$$\begin{pmatrix} \hat{e}_a \\ \hat{e}_b \\ \hat{e}_c \end{pmatrix} = \frac{1}{\sqrt{6}} \begin{pmatrix} 1 & 1 & -2 \\ -\sqrt{3} & \sqrt{3} & 0 \\ \sqrt{2} & \sqrt{2} & \sqrt{2} \end{pmatrix} \begin{pmatrix} \hat{e}_x \\ \hat{e}_y \\ \hat{e}_z \end{pmatrix}.$$

b is parallel to the bond; a/c-axis are perp. to the bond/plane

H in abc coordinate

$$\mathcal{H} = \sum_{\langle i,j\rangle} \Big[J_{XY} \left(S_i^a S_j^a + S_i^b S_j^b \right) + J_Z S_i^c S_j^c$$
$$+ J_{ab} \left[\cos\phi_\gamma \left(S_i^a S_j^a - S_i^b S_j^b \right) - \sin\phi_\gamma \left(S_i^a S_j^b + S_i^b S_j^a \right) \right]$$
$$- \sqrt{2} J_{ac} \left[\cos\phi_\gamma \left(S_i^a S_j^c + S_i^c S_j^a \right) + \sin\phi_\gamma \left(S_i^b S_j^c + S_i^c S_j^b \right) \right] \Big],$$

$\phi_\gamma = 0, \frac{2\pi}{3}, \frac{4\pi}{3}$ for $\gamma =$ z,x,y- bond

$$J_{XY} = J + J_{ac}, \; J_Z = J + J_{ab},$$
$$J_{ab} = \frac{1}{3}K + \frac{2}{3}\Gamma, \; J_{ac} = \frac{1}{3}K - \frac{1}{3}\Gamma.$$

Note: $J_Z - J_{XY} = J_{ab} - J_{ac} = \Gamma$

in-plane vs. out-of-plane anisotropy due to Gamma

J. Chaloupka, G. Khaliullin, PRB 92, 24413 (2015);
Pyrochlore lattice, S. Onoda, J. Phys.:Conf. Ser. 320, 012065 (2011); K. Ross, et al, PRX 1, 021002 (2011)

$$C_{2a} : (S^a, S^b, S^c) \rightarrow (S^a, -S^b, -S^c) \text{ and } \phi_x \leftrightarrow \phi_y$$

Consequence of broken C2a

$$\mathcal{H} = \sum_{\langle i,j \rangle} \Big[J_X (S_i^a S_j^a + S_i^b S_j^b) + J_Z S_i^c S_j^c$$

$$+ J_{ab} \left[\cos\phi_\gamma (S_i^a S_j^a - S_i^b S_j^b) - \sin\phi_\gamma (S_i^a S_j^b + S_i^b S_j^a) \right]$$

$$- \sqrt{2} J_{ac} \left[\cos\phi_\gamma (S_i^a S_j^c + S_i^c S_j^a) + \sin\phi_\gamma (S_i^b S_j^c + S_i^c S_j^b) \right] \Big]$$

+ single-ion anisotropy, DM on 2nd n.n. with d//c, further n.n. Heisenberg + …

All interactions are invariant under C_{2a} *Except*

$J_{ac} \rightarrow -J_{ac}$

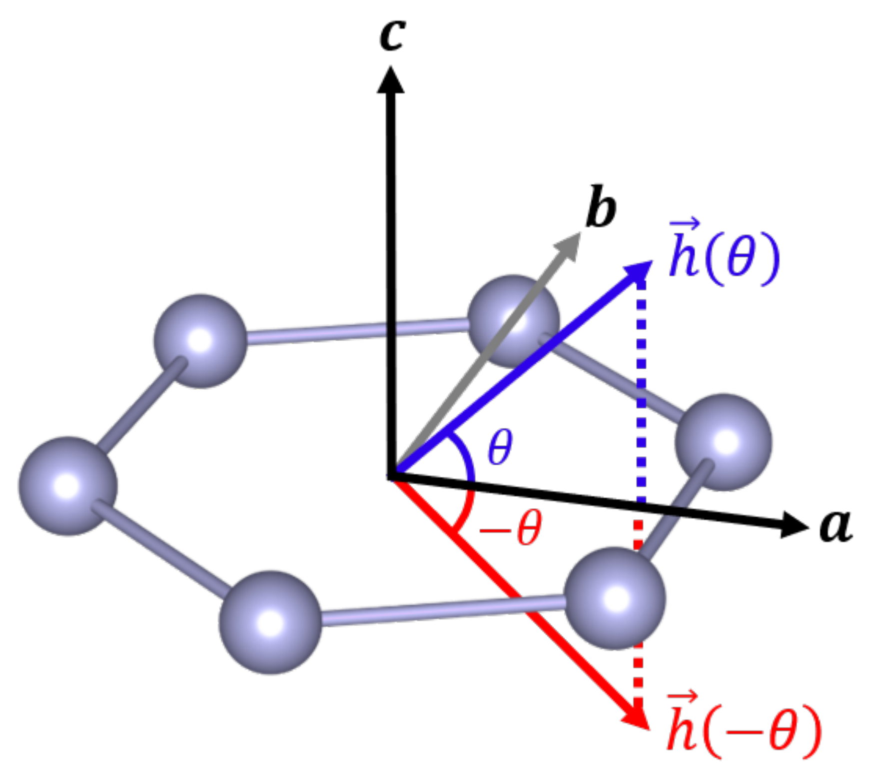
$$J_{ac} = \frac{1}{3} K - \frac{1}{3} \Gamma.$$

- A magnetic field can detect the broken C_{2a} symmetry and be used to isolate the interaction that breaks C_{2a}.

$$H_{total} = \mathcal{H} - g\mu_B \vec{h} \cdot \vec{S}$$

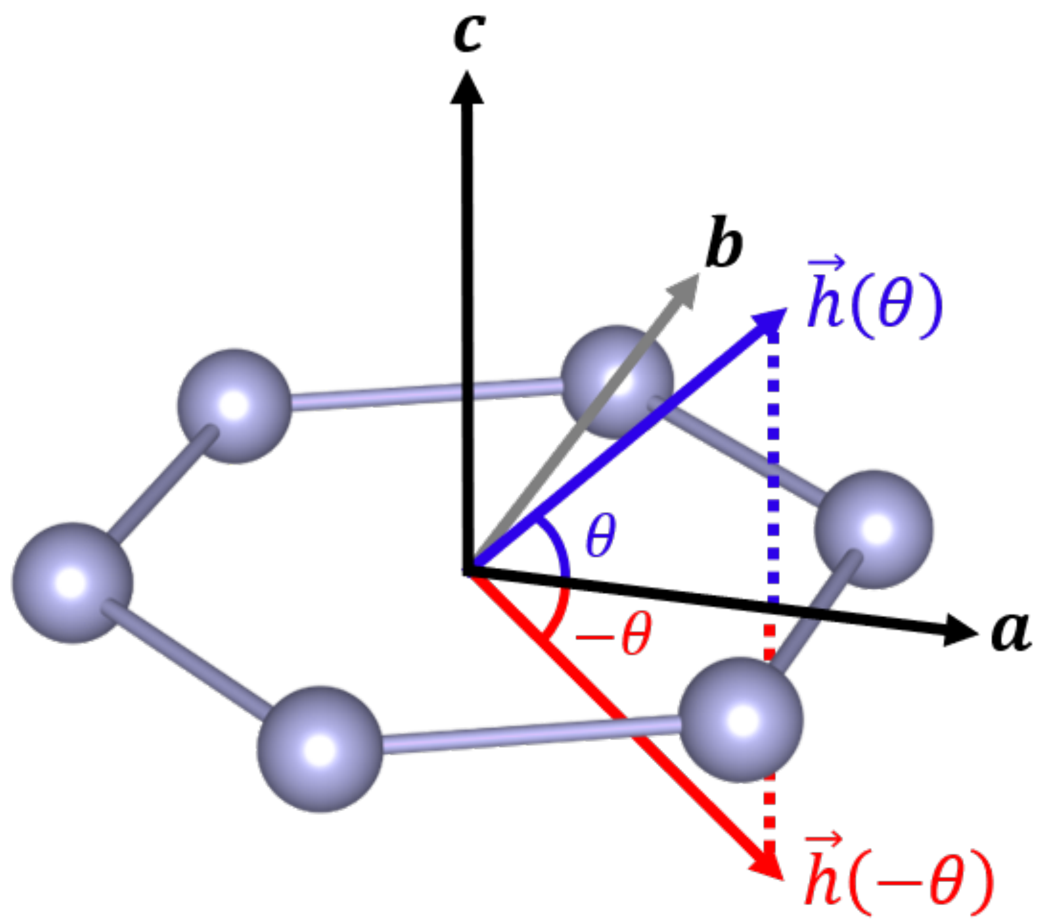
field is in the ac-plane

Two different directions of field related by C2a:
different excitation energies only if Jac finite



$$E_n(\theta) - E_n(-\theta) \propto J_{ac}$$

$$J_{ac} = \frac{1}{3}K - \frac{1}{3}\Gamma.$$ **needs to isolate both Gamma and K**



Recipe to isolate Kitaev

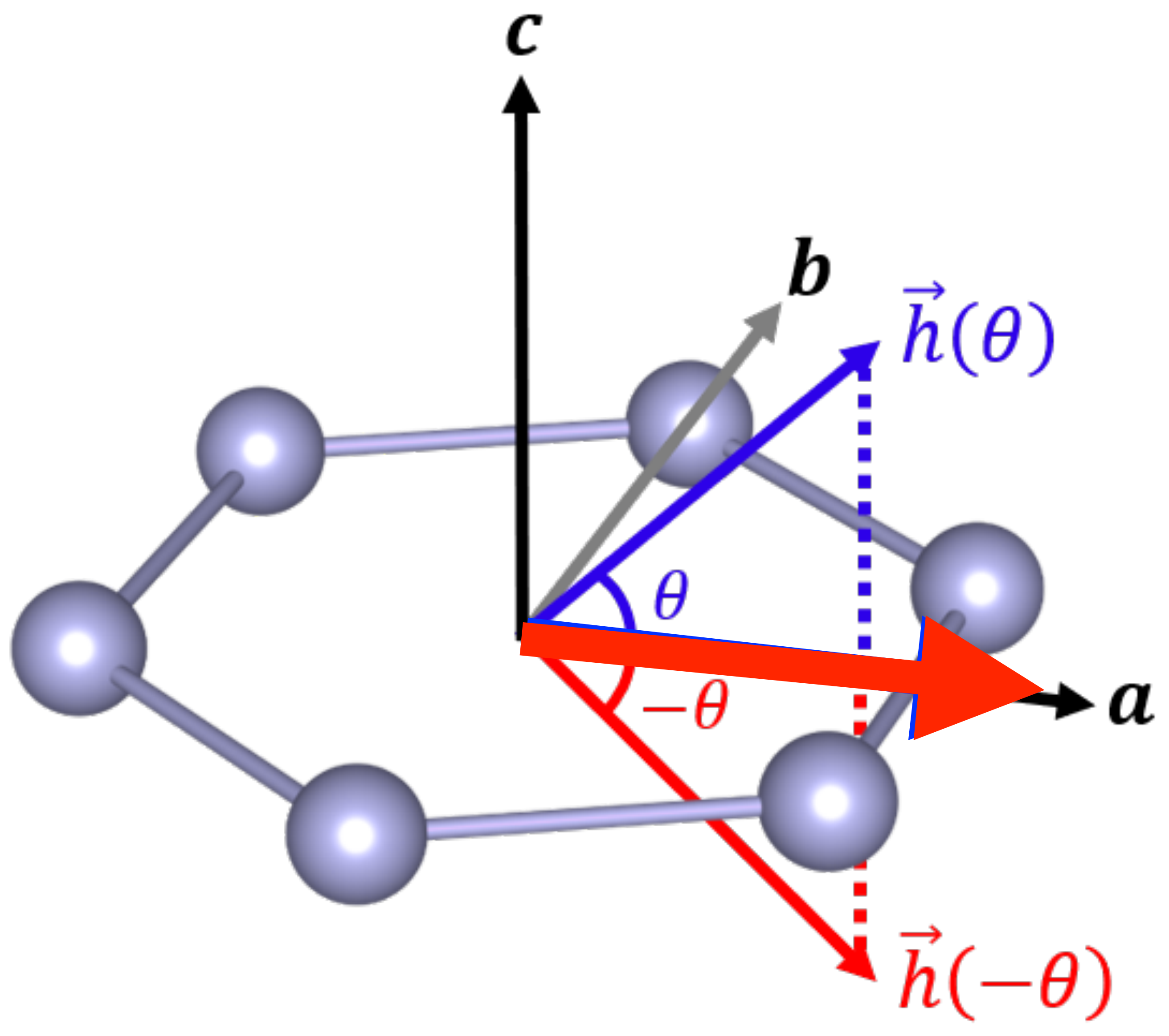
1. apply B in the ac-plane with θ
2. measure the spin excitations
3. rotate B with angle of $-\theta$
4. measure the spin excitations
5. energy difference between 2 & 4:

$$\delta\omega_K = \omega(\theta) - \omega(-\theta) \propto K - \Gamma$$

6. In-plane and out-of-plane energy excitation = Gamma

$$\delta\omega_A = \omega(\theta = 0) - \omega(\theta = 90^{\circ})$$ for S=1/2

7. subtract Gamma from 5, then single out the Kitaev interaction!

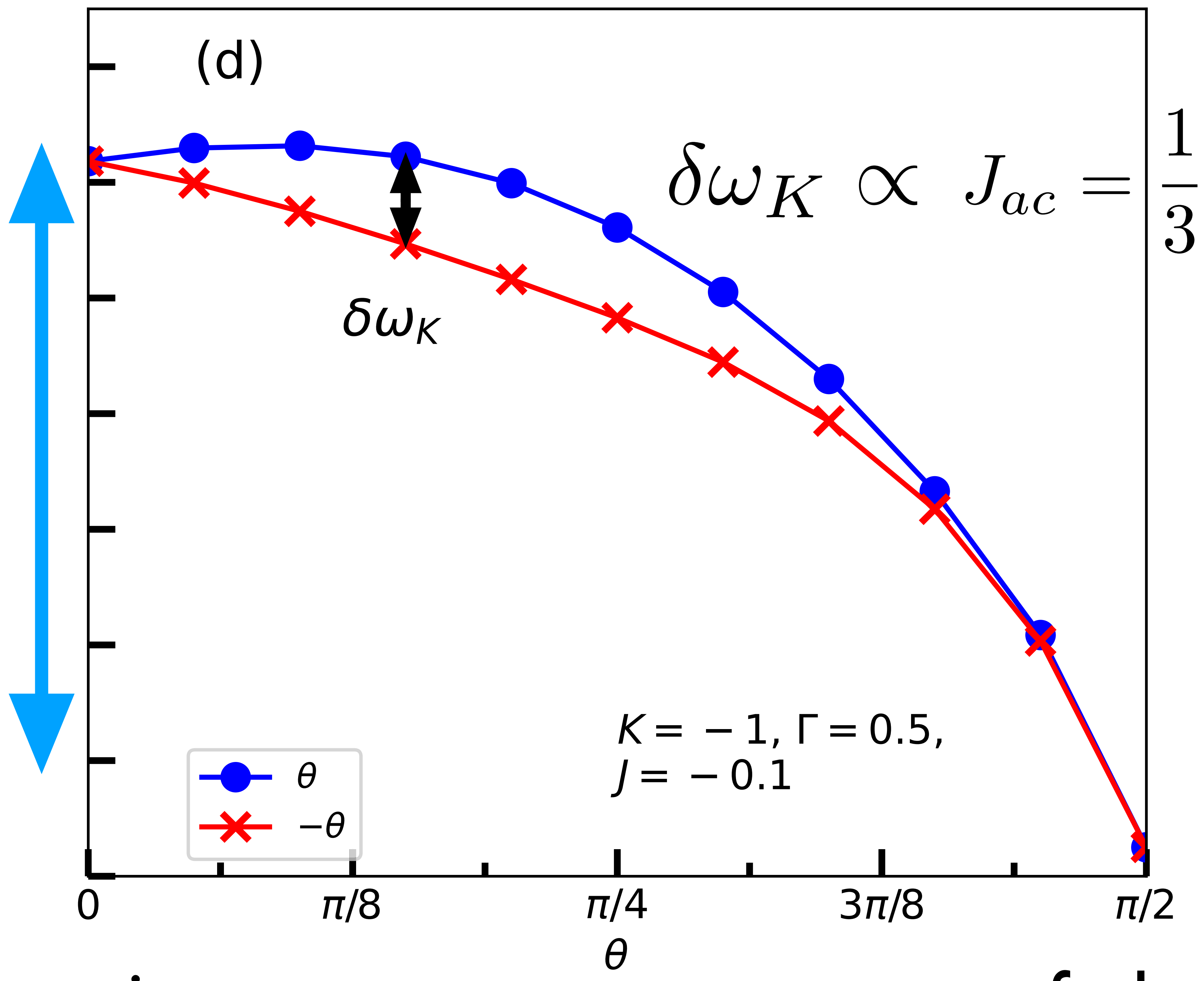


For S=1/2 $J_{ac} = \frac{1}{3}K - \frac{1}{3}\Gamma.$

Angle-dependent Ferromagnetic Resonance,
NMR, Optical spectroscopy

$$\delta\omega_A \equiv \omega(\theta = 0) - \omega(\theta = 90^\circ) \propto$$
$$J_{XY} - J_Z = \Gamma$$

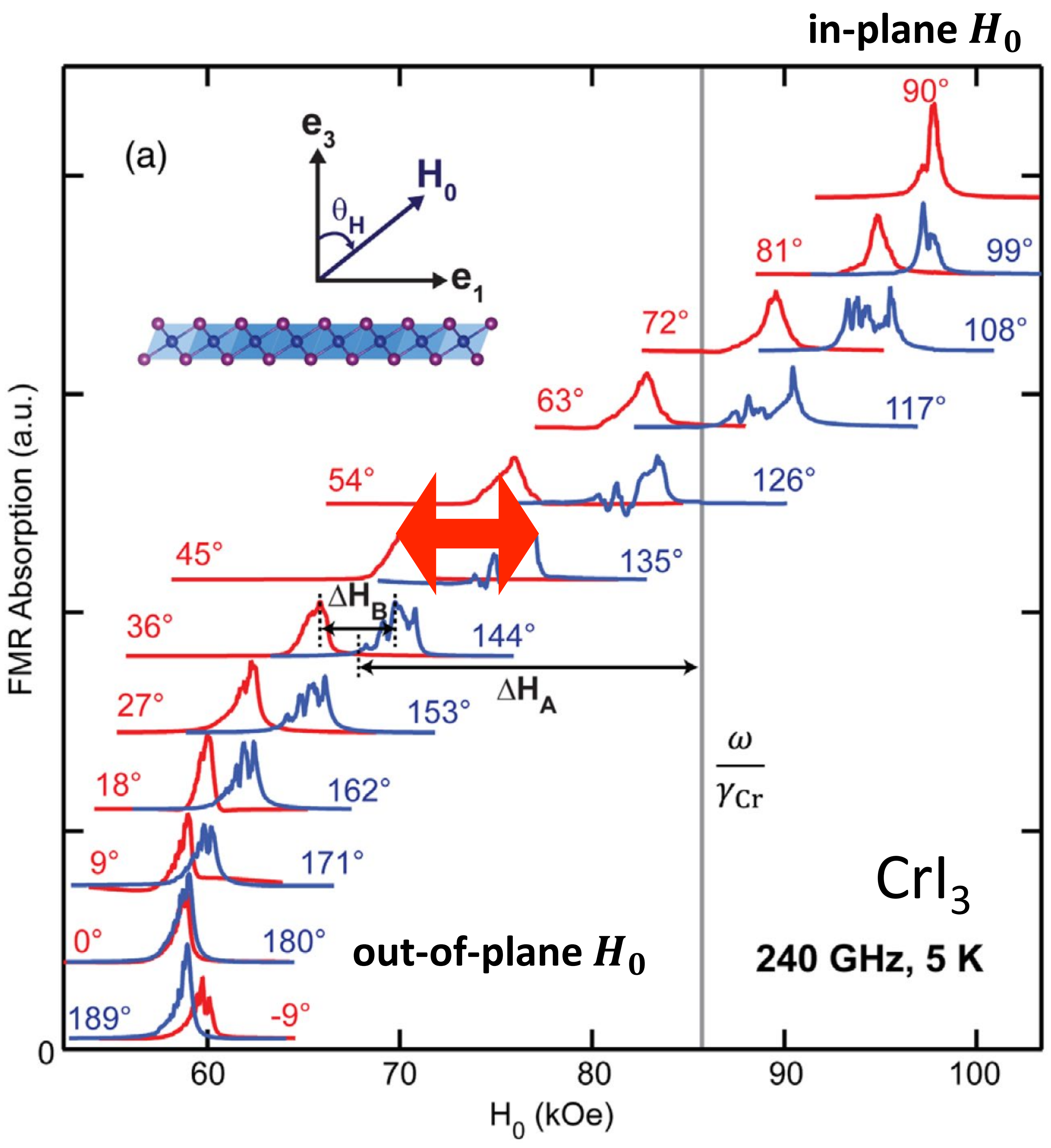
$$\delta\omega_K \propto J_{ac} = \frac{1}{3}(K - \Gamma)$$



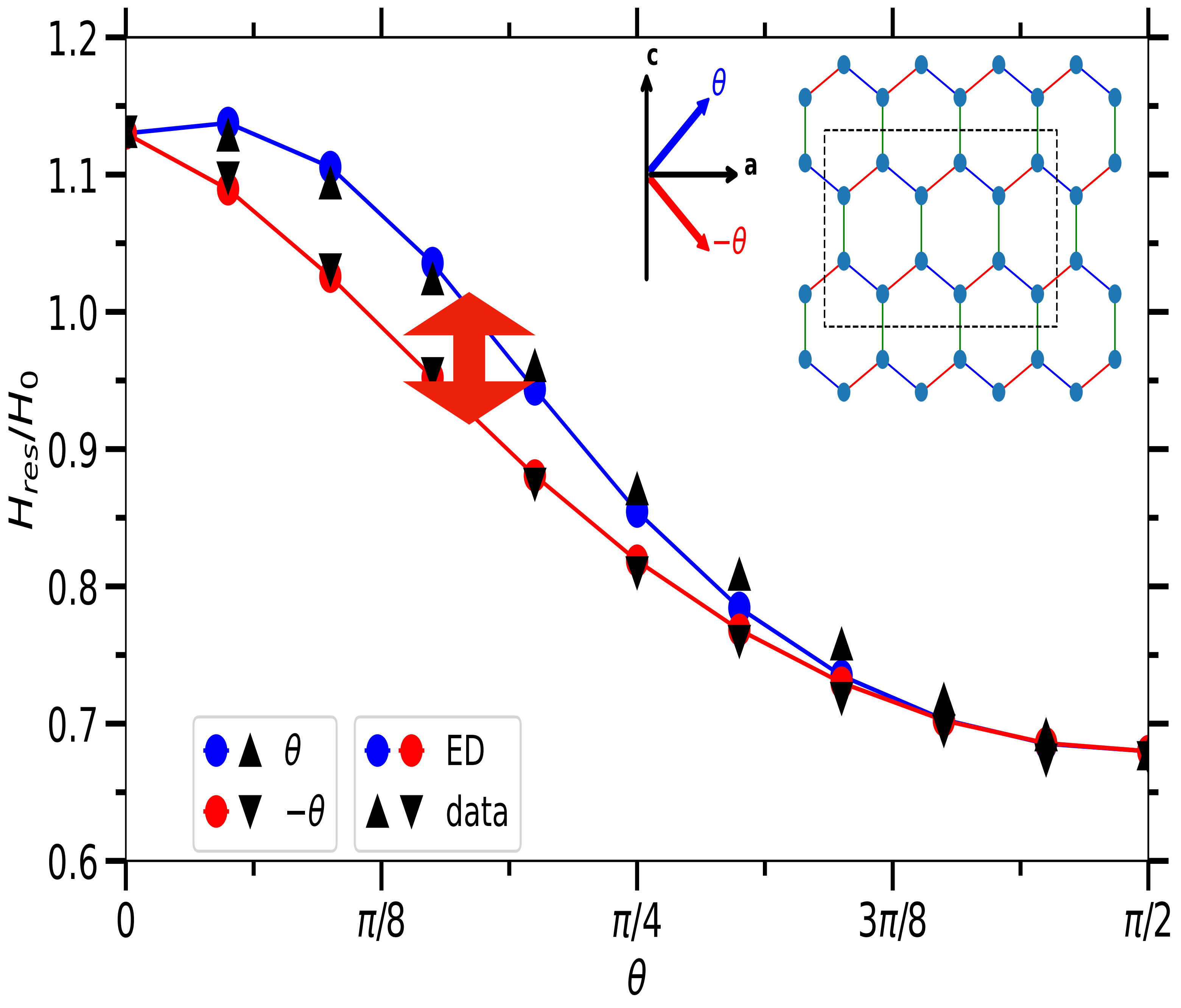
h//a-axis

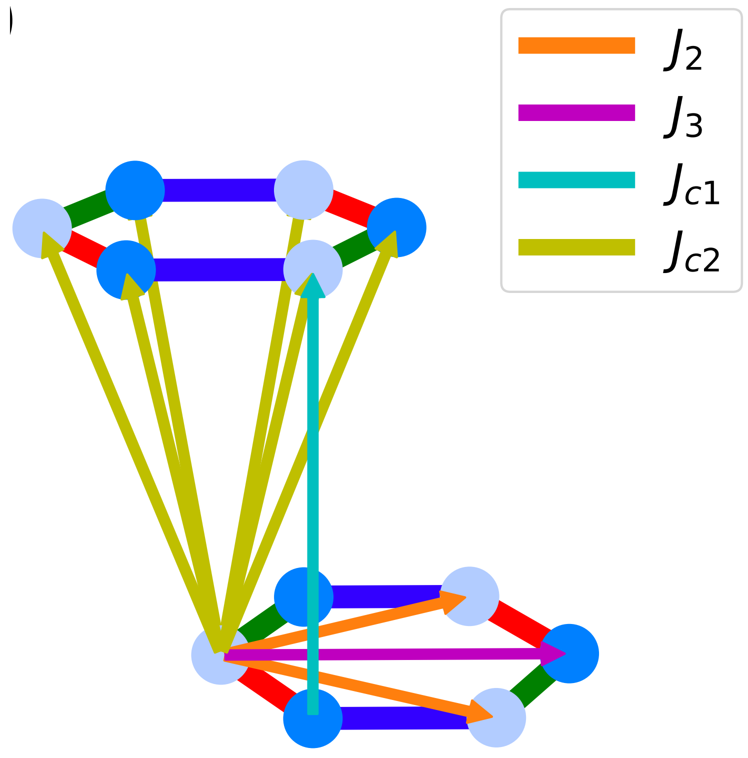
out-of-plane h//c

Application to CrI3

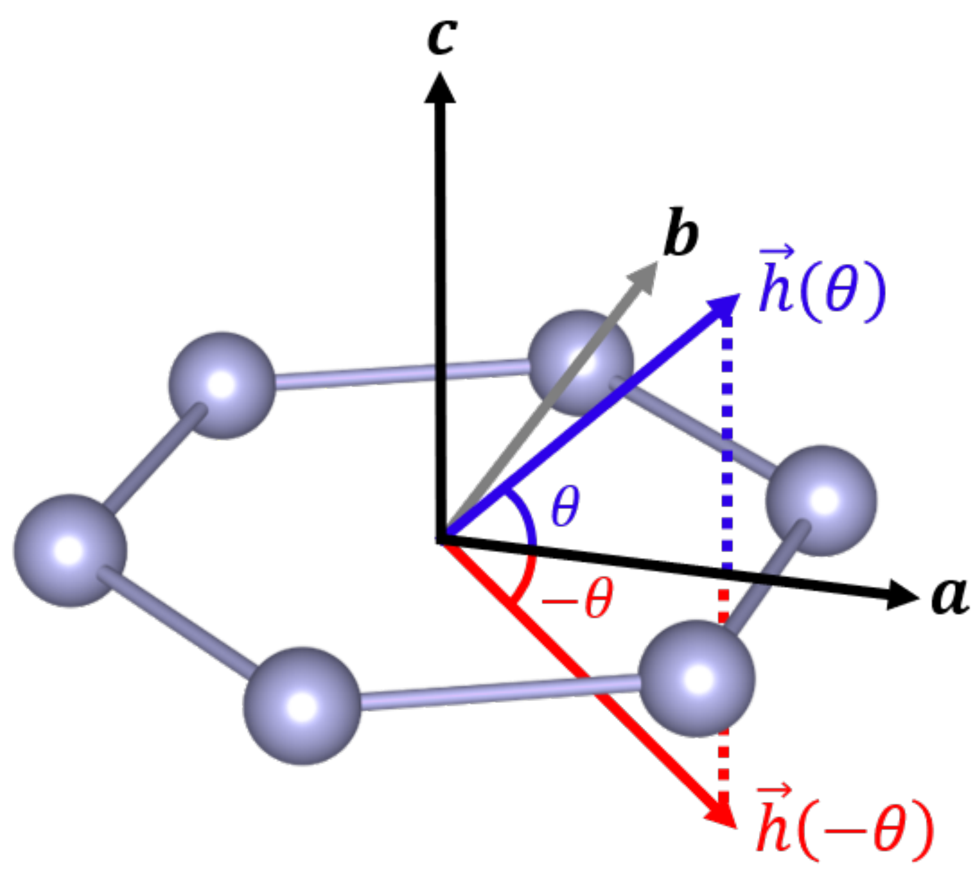


I. Lee et al, PRL 124, 017201 (2020)





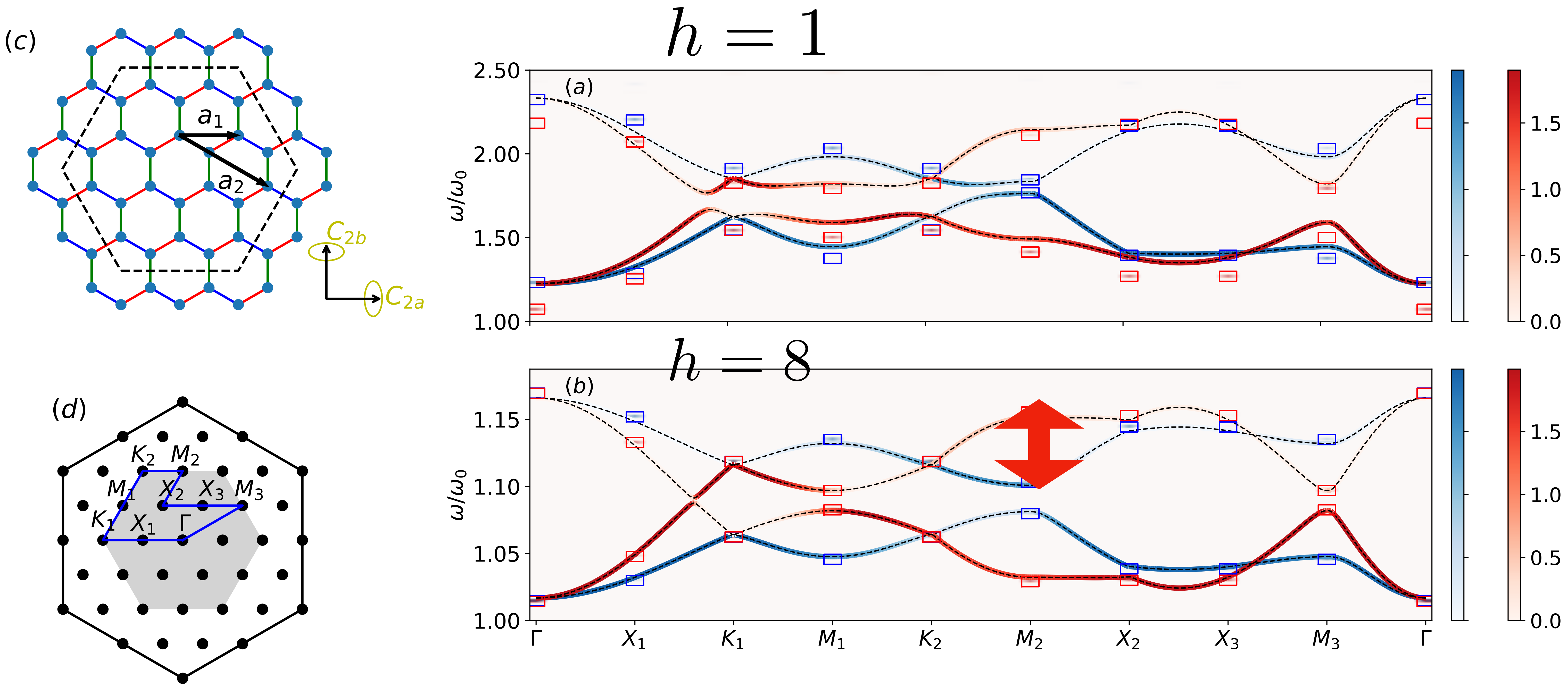
Interaction	value (meV)	Interaction	value (meV)
J	-2.5	J_2	-0.09
K	1.1	J_3	0.13
Γ	$\sim$0	J_{c1}	0.048
Γ'	$\sim$0	J_{c2}	-0.071
A_c	-0.23	D_c	0.17



Inelastic Neutron Scattering

$$K = -1, \Gamma = 0.5, J = -0.1$$

$$\theta = \pm 30^\circ$$



Jiefu Cen, HYK, Commun. Phys. 5, 119 (2022)

Can we use a "fixed" magnetic field direction?

— Two different momenta under a certain magnetic field

Symmetry of H

	$\mathcal{T}$	C_{2b}	$\mathcal{T}C_{2b}$	C_{2a}	$\mathcal{T}C_{2a}$
$\mathbf{h}=0$	○	○	○	× / ○	× / ○
$\mathbf{h}$ in $a-c$ plane	×	×	○	×	×
$\mathbf{h}$ in $b-c$ plane	×	×	×	×	× / ○

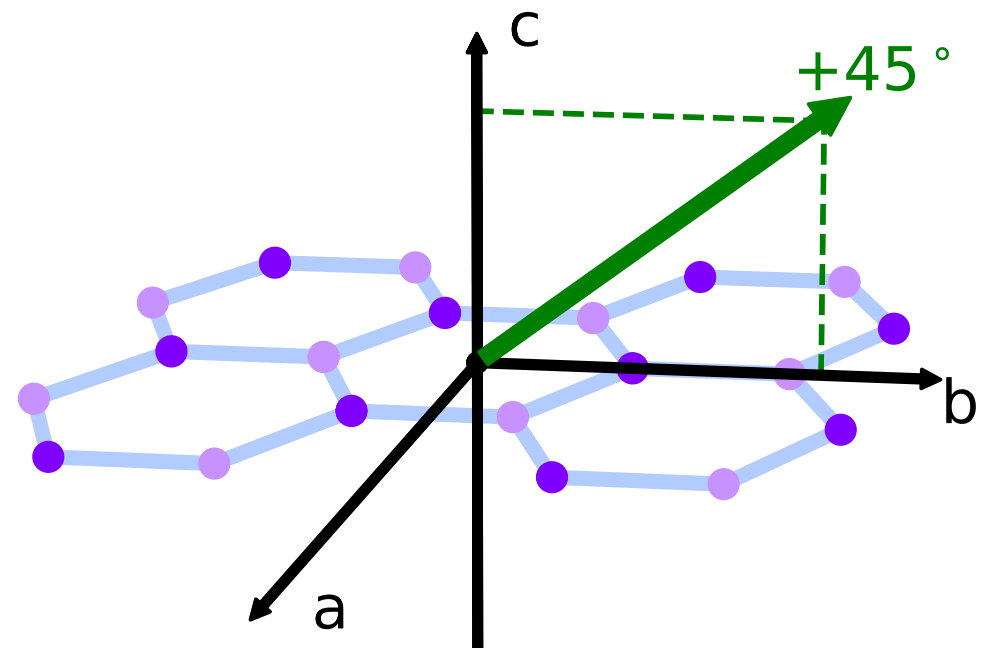
$J_{ac} \neq 0$ (×)

$J_{ac} = 0$ (○)

2nd Recipe to isolate Jac

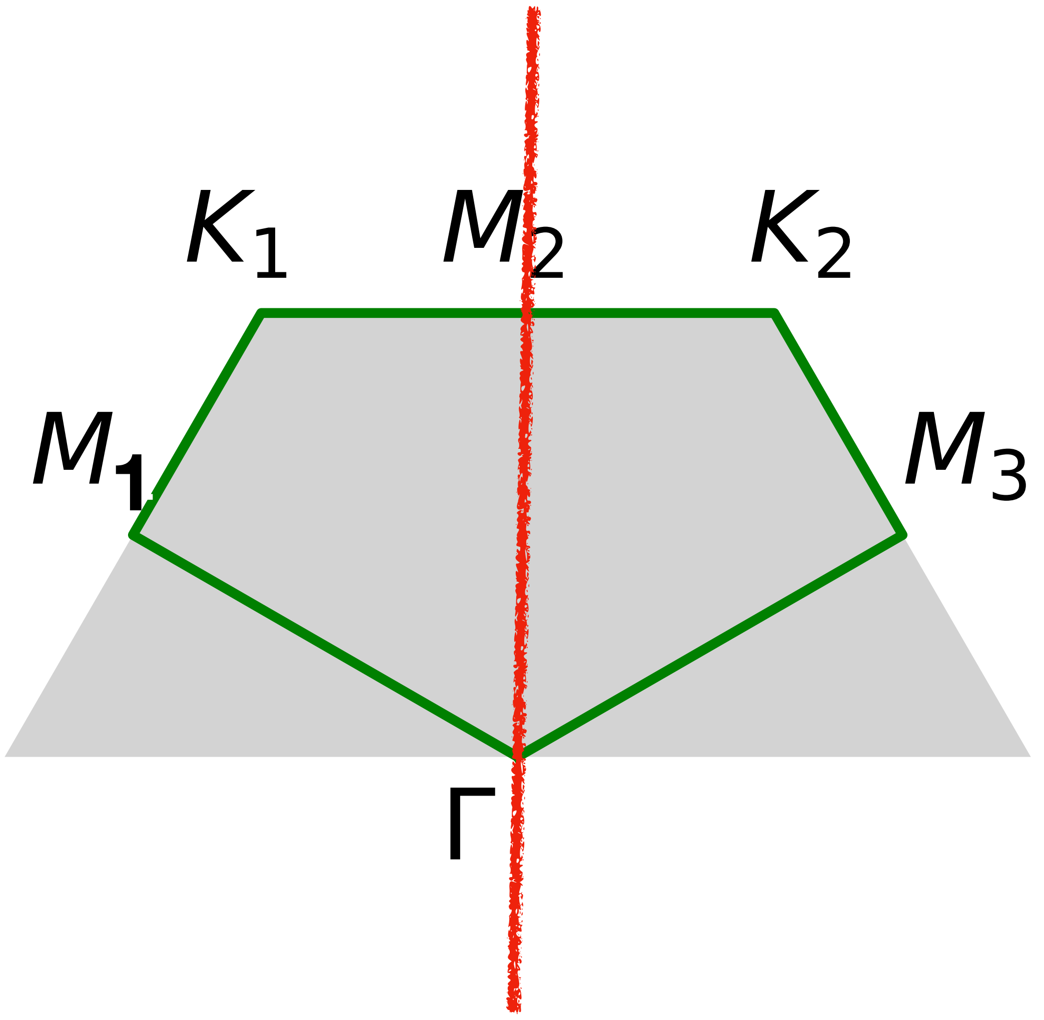
$$J_{ac} \propto (K - \Gamma)$$

1. apply B in the bc-plane with angle theta



2. measure the spin excitations at M1 and M3

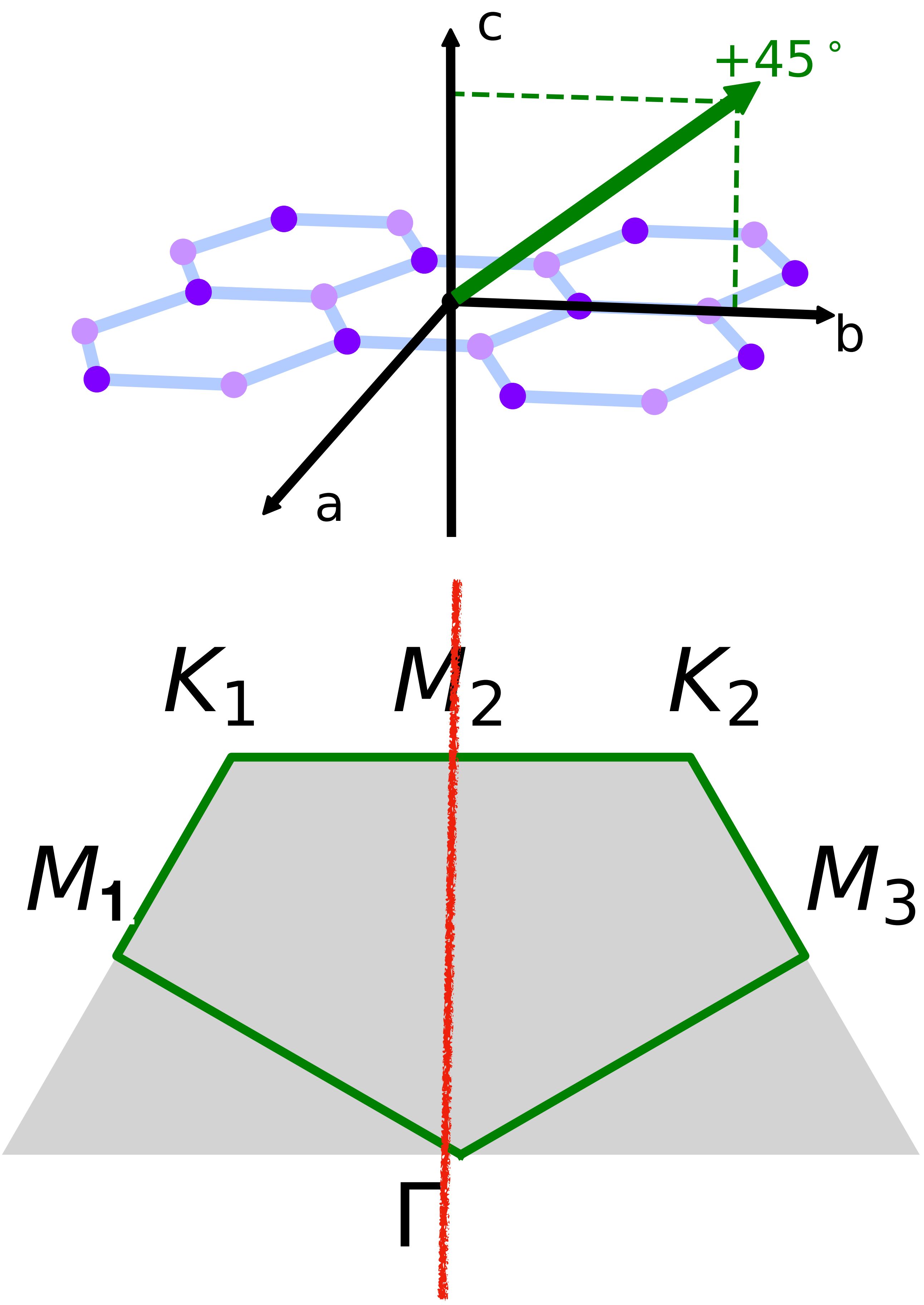
3. energy difference between M1 - M3 $\propto (K - \Gamma)$



bc-plane: no mirror symmetry

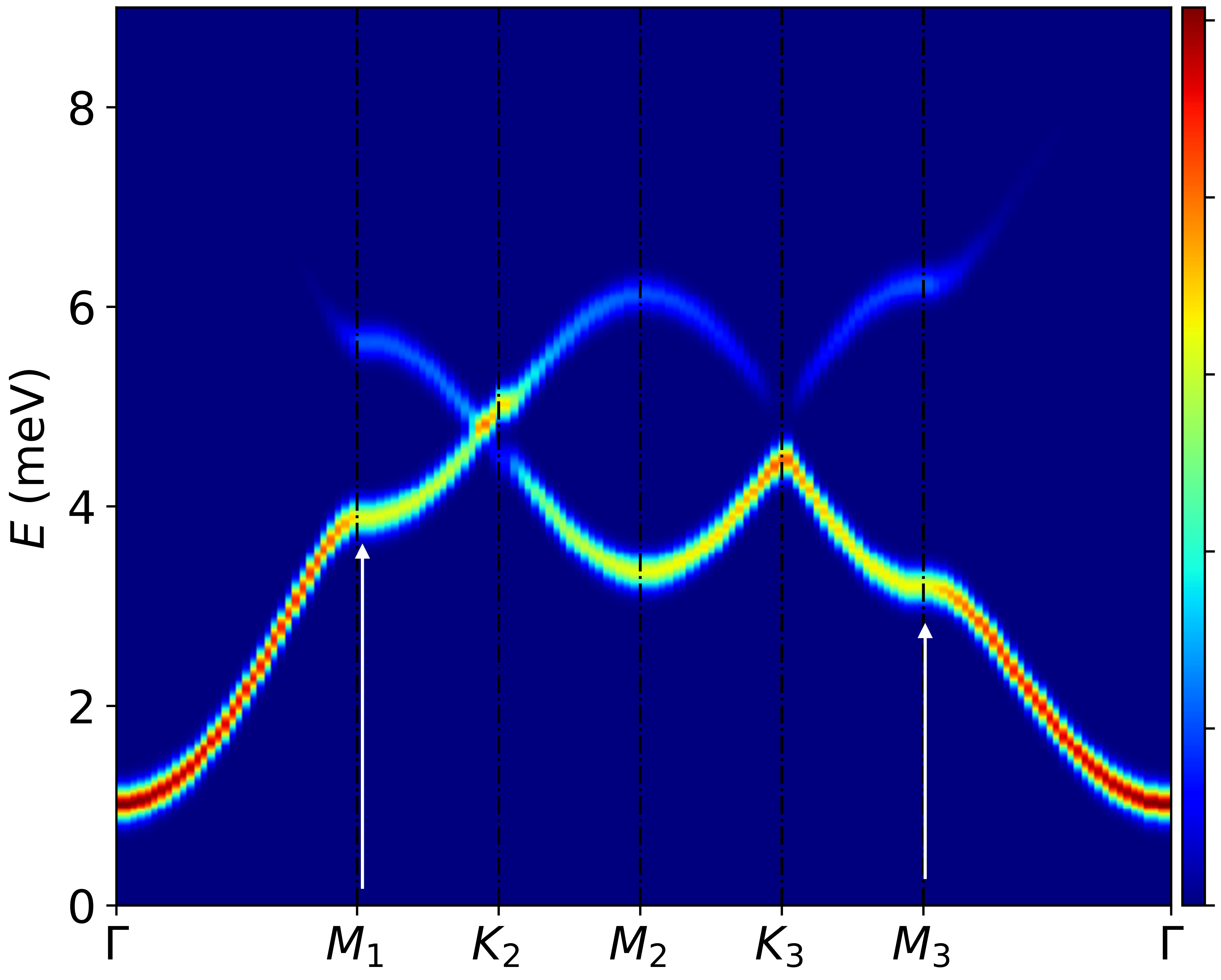
Jieful Cen, HYK, arXiv:2208.13807 (2022)

In higher-S, $\Gamma \sim 0 \ \rightarrow \ J_{ac} \sim K$



Example

J = -1, K=0.5, h =1

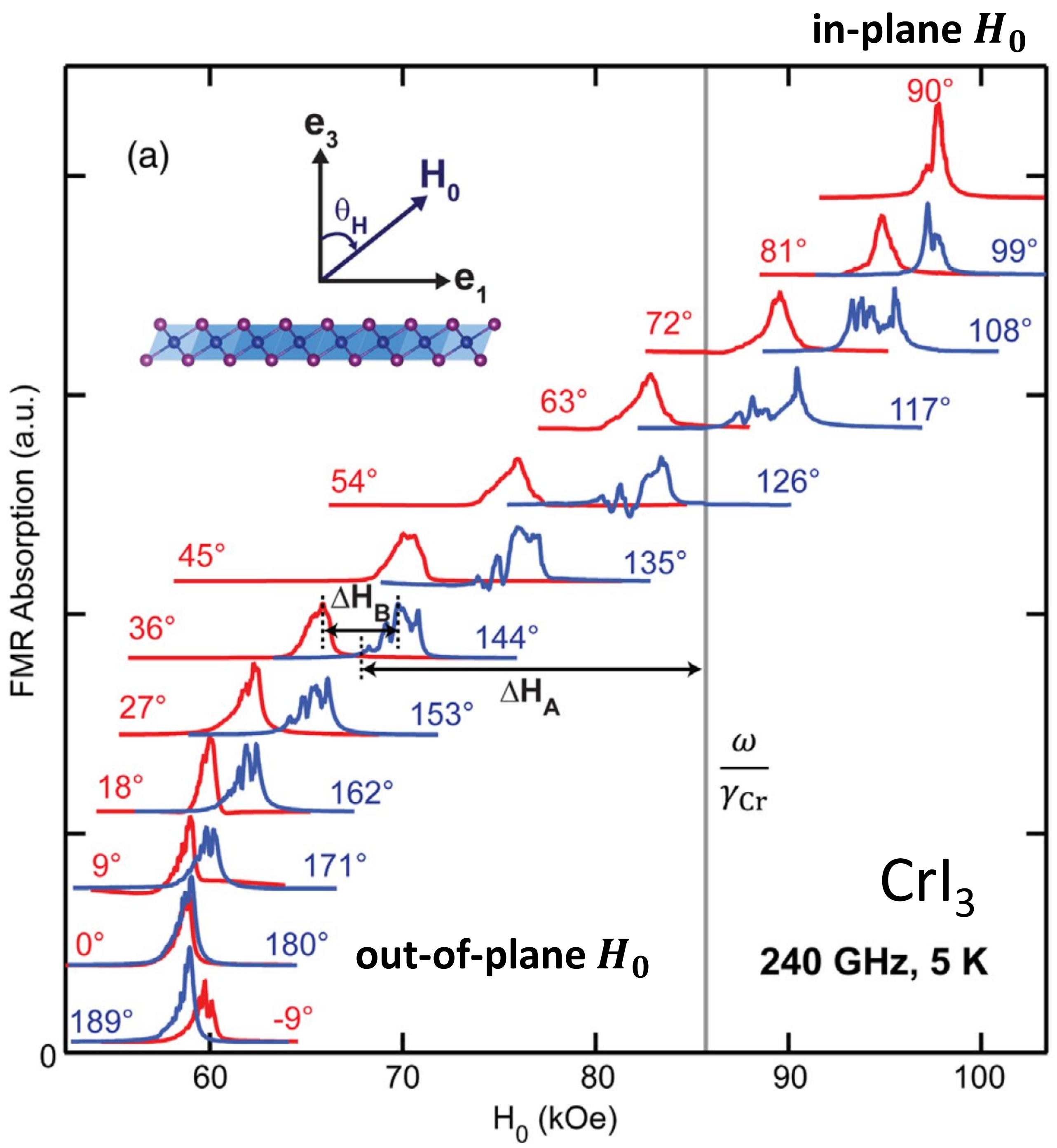


bc-plane: no mirror symmetry -> finite (Kitaev - Gamma) interaction

Application to CrI3

Current debate

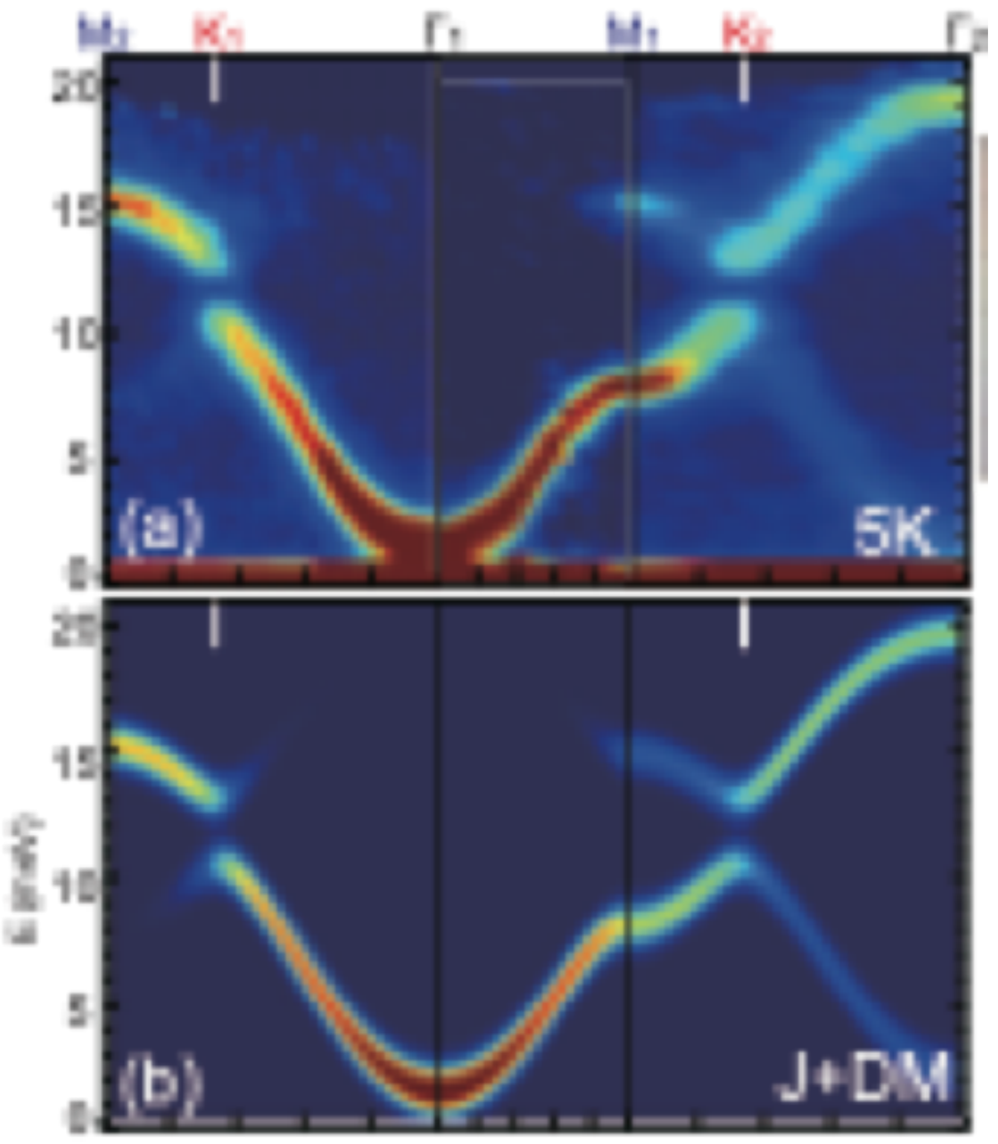
Angle dependent Ferromagnetic Resonance (FMR): CrI3



Kitaev dominant

Inhee Lee, et al, Phys. Rev. Lett. 124, 017201 (2020)

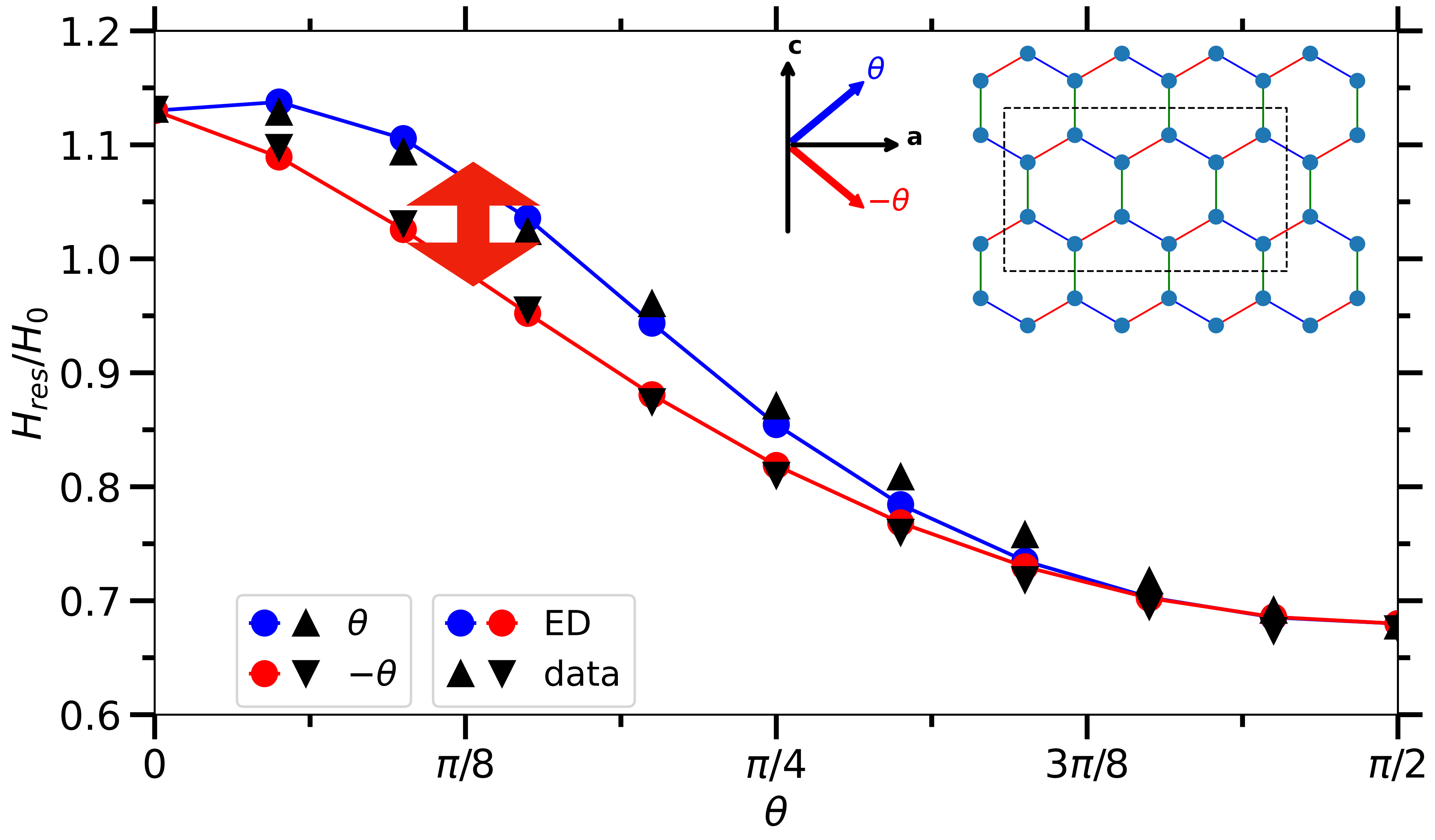
Inelastic neutron scattering

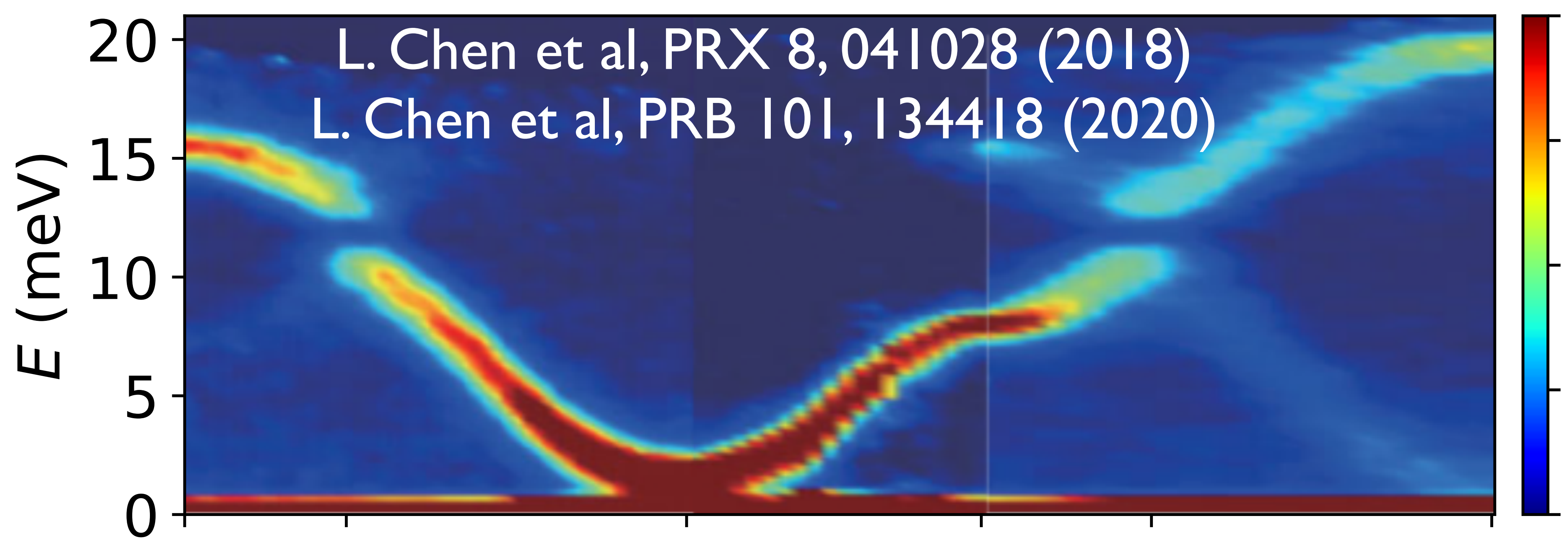


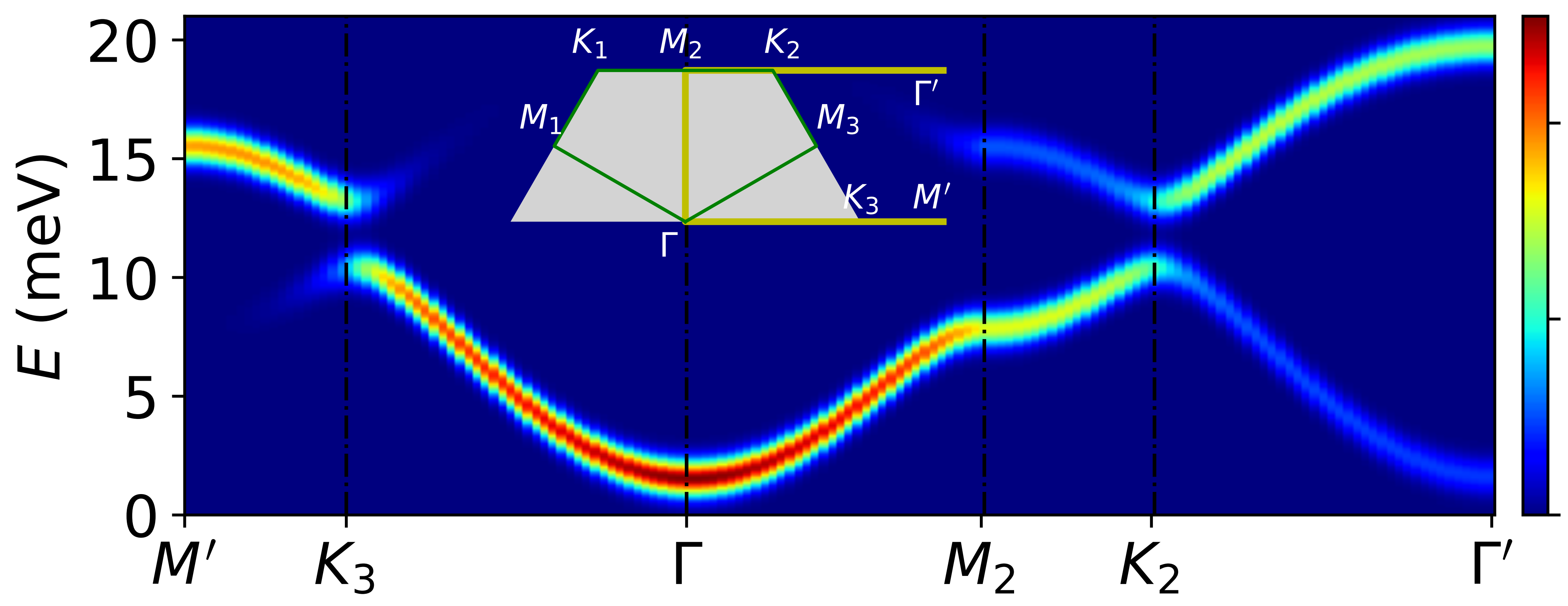
J + DM interaction

L. Chen et al, PRX 11, 031047 (2021)

Applying our theory







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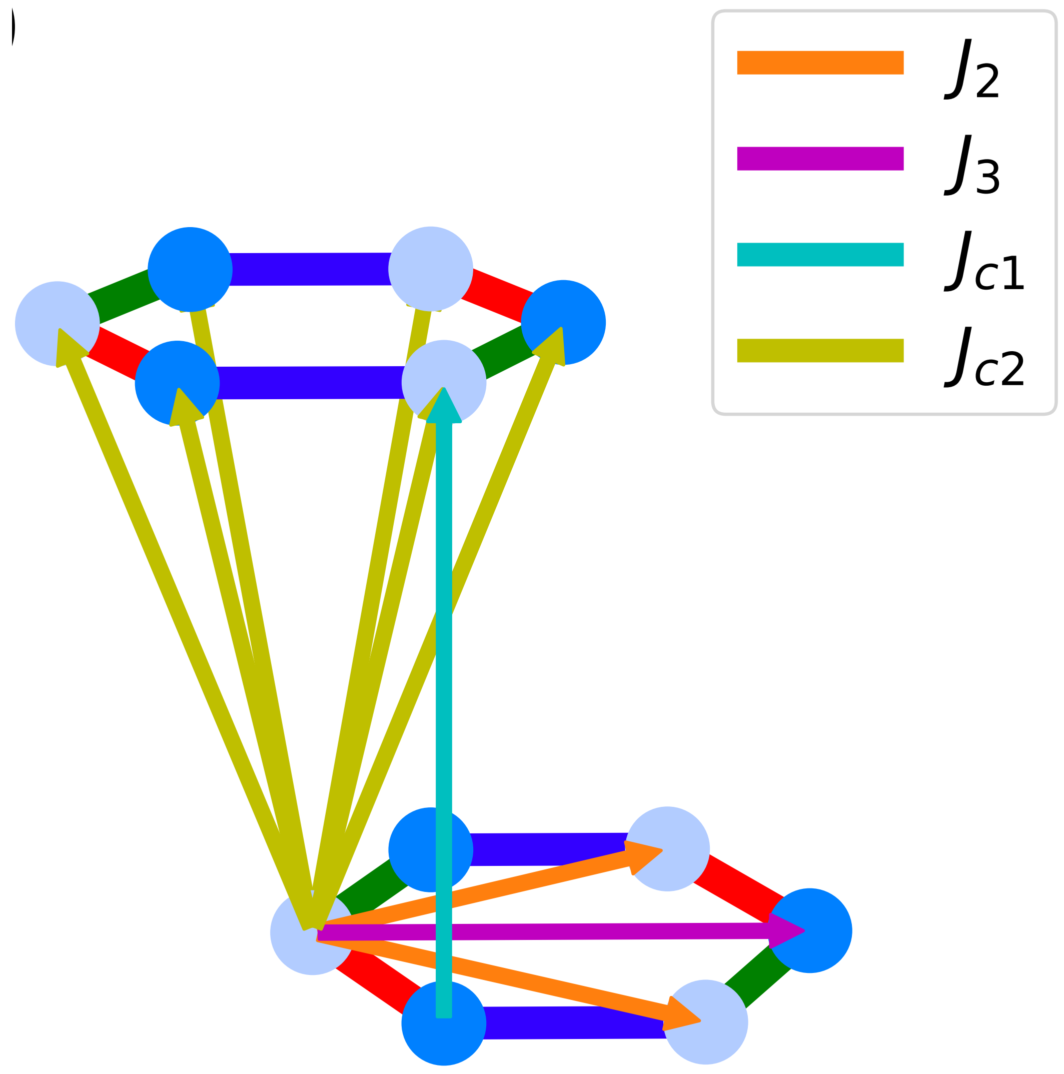
Prediction using our 2nd recipe: CrI3



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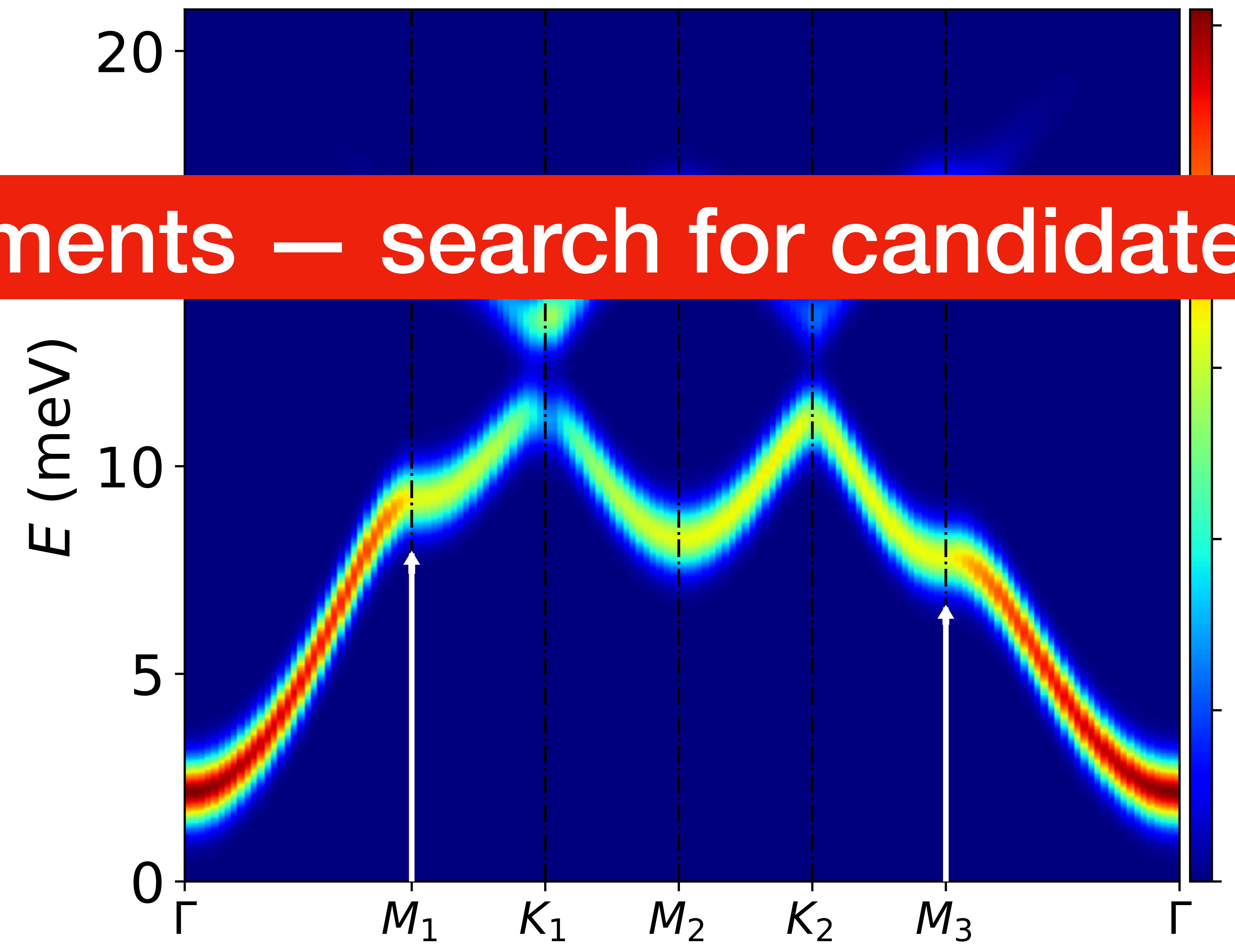
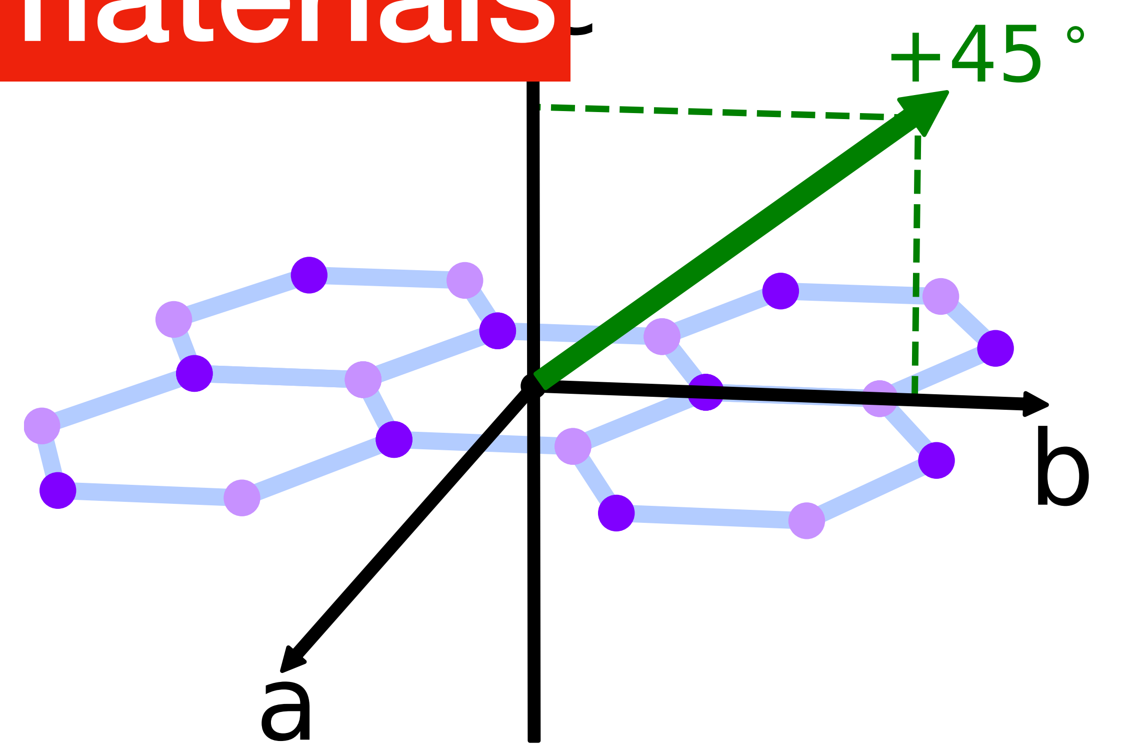
Future experiments — search for candidate materials





Jiefu Cen, HYK, arXiv:2208.13807 (2022)

Summary

- Higher-spin S Kitaev interaction:
  combination of Hund's at transition metal + SOC in ligand

- Proposal to estimate the Kitaev interaction for general S:
  apply magnetic field in the bc-plane;
  measure spin excitations at two momenta (M1 and M3) - broken mirror symmetry
  difference is due to Kitaev interaction (Gamma, Gamma' ~ 0 for spin > 1/2)