Multipolar Spin Liquids

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References:

G. Khaliullin, D. Churchill, P. P. Stavropoulos, HYK, Phys. Rev. Research 3, 033163 (2021)

D. Churchill, HYK, Phys. Rev. B 105, 014438 (2022)

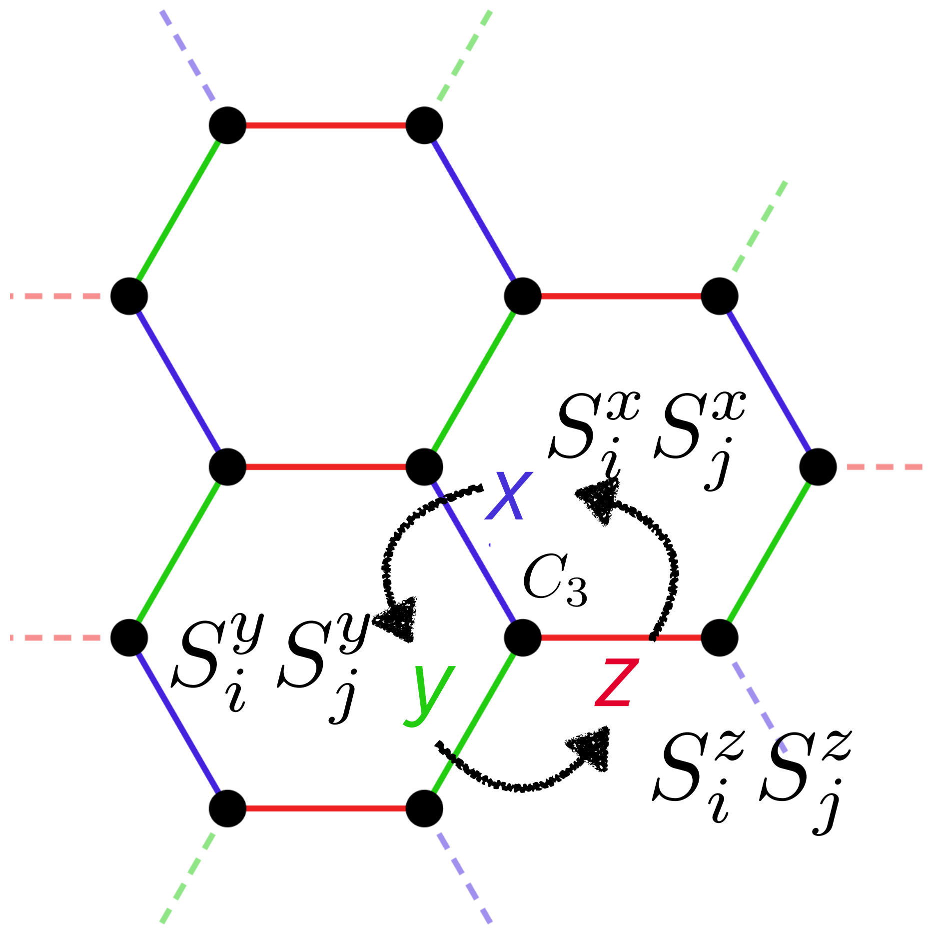
A. Rayyan, D. Churchill, HYK, Phys. Rev. B 107, L020408 (2023)

Kitaev model and Kitaev spin liquid in S=1/2 systems

Kitaev Exchange

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

where $\gamma = x, y, z$



bond-dependent Ising interaction

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

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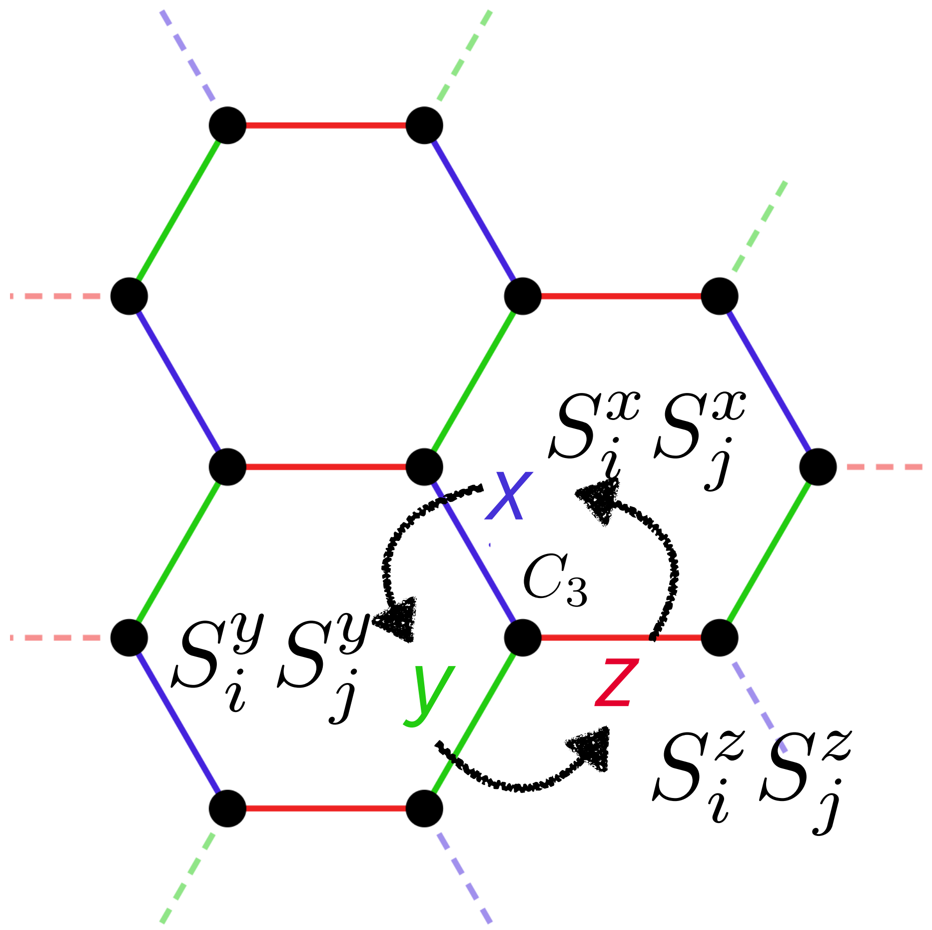

bond-dependent Ising interaction

Emergent particles: Majorana Fermions & vortex

particle = its own antiparticle

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Question:

Can we realize Kitaev model and quantum spin liquids in multipolar systems ?

Introduction: multipolar systems

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even number of electrons — free from Kramer's theorem

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Hund's rule + crystal field theory : non-Kramer doublet

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Under cubic crystal field $\Gamma_{J=4} = \Gamma_1 \oplus \Gamma_3 \oplus \Gamma_4 \oplus \Gamma_5$

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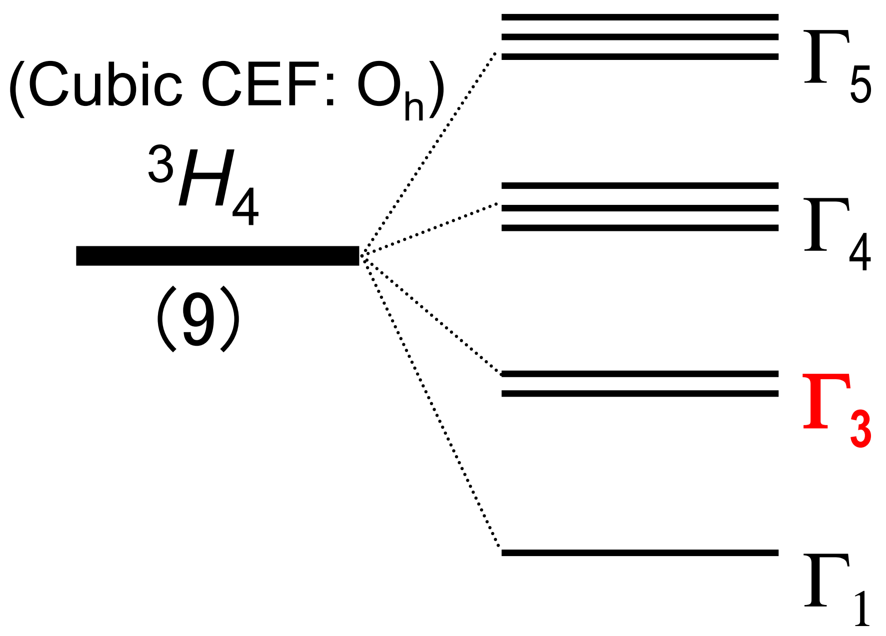

	Dipole	Quadru-pole	Octu-pole
Γ_5	○	○	×
Γ_4	○	○	×
Γ_3	×	○	○
Γ_1	×	×	×

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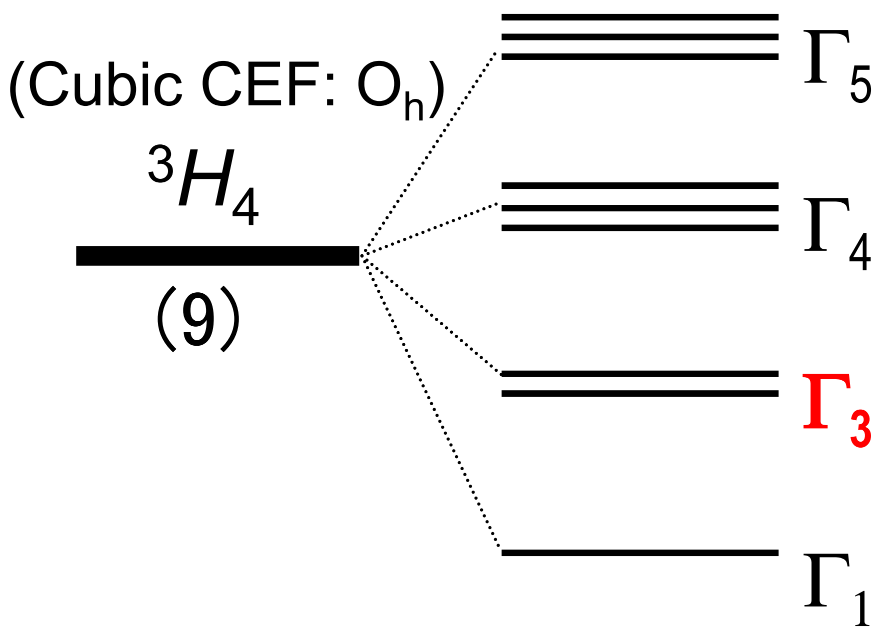

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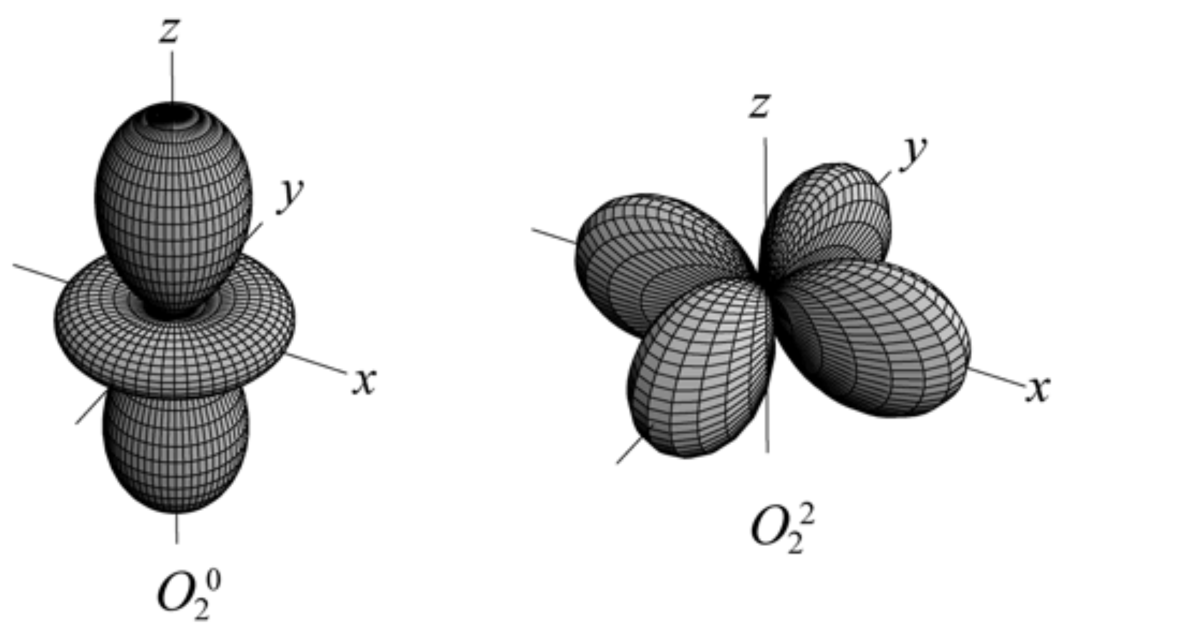
Γ_3	×	○	○
Γ_1	×	×	×

$$\sqrt{\tfrac{7}{24}}(|+4\rangle + |-4\rangle) - \sqrt{\tfrac{10}{24}}|0\rangle$$
$$\tfrac{1}{\sqrt{2}}(|+2\rangle + |-2\rangle)$$



$Q_{zz} \propto 3J_z^2 - J^2$ $(Q_{xx} - Q_{yy}) \propto (J_x^2 - J_y^2)$

$$\langle \mathbf{J} \rangle = 0$$

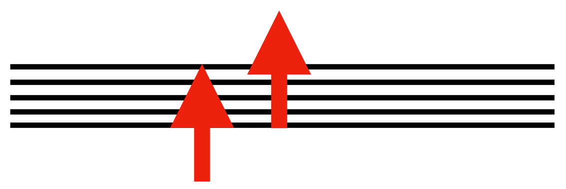
$$\langle Q_{ij} \rangle = \langle \frac{1}{2}\left(J_i J_j + J_j J_i\right) - J^2 \delta_{ij} \rangle$$

Focus: $5d^2$ systems

G. Chen and L. balents, PRB 84, -94420 (2011); D. D. Mahraj, et al, PRL 124, 087206 (2020); …..

Focus: 5d^2 systems

2 electrons in d orbitals



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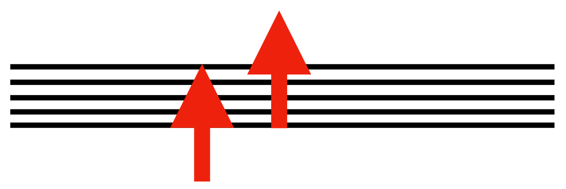

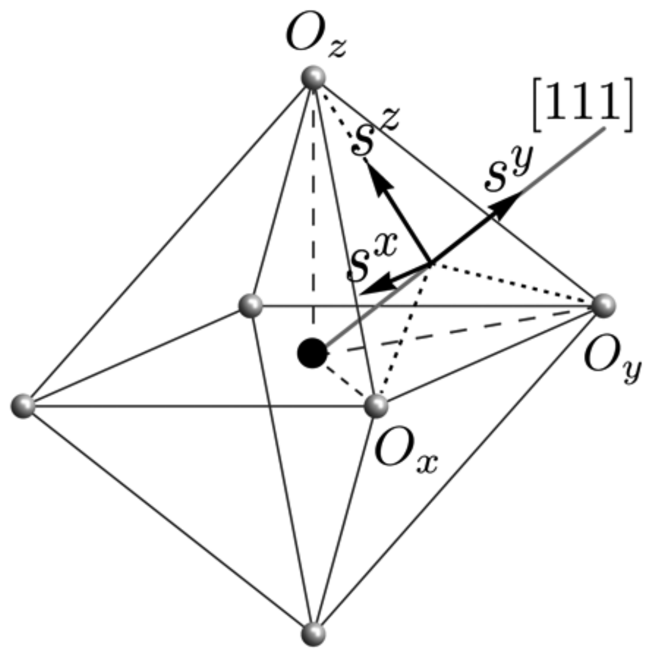

octahedral crystal field

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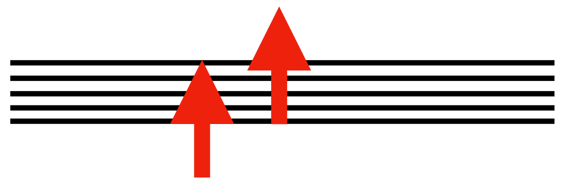

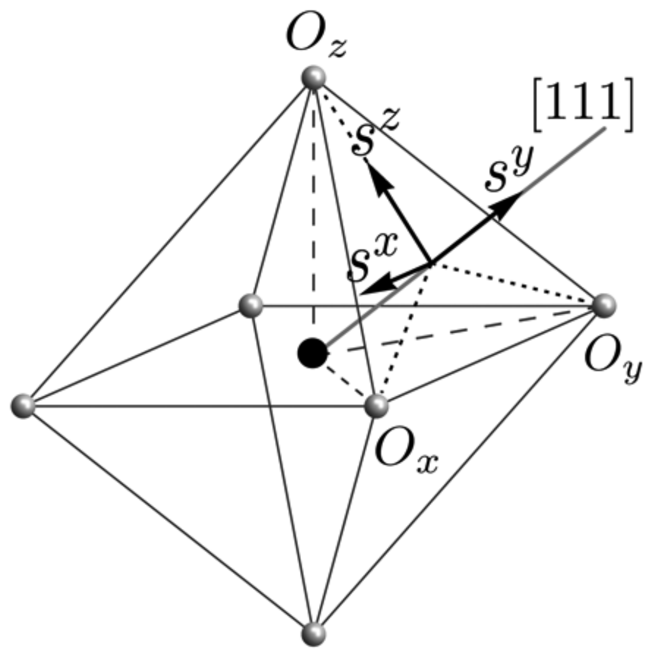

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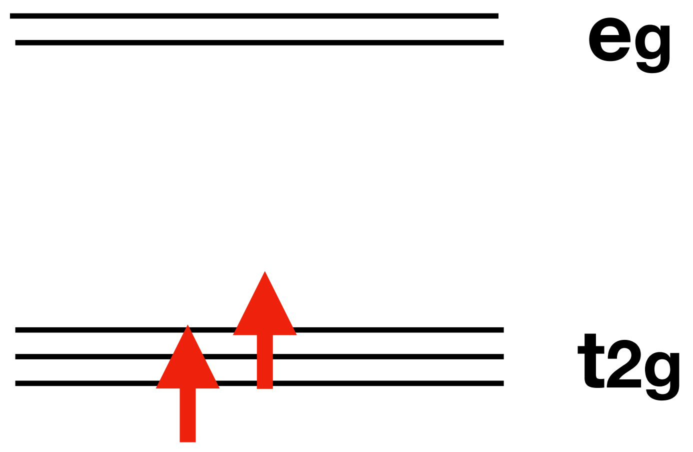

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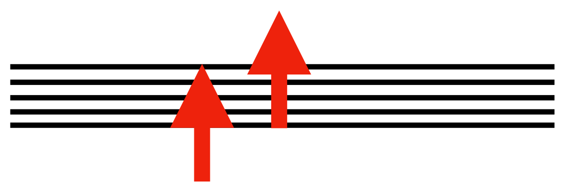

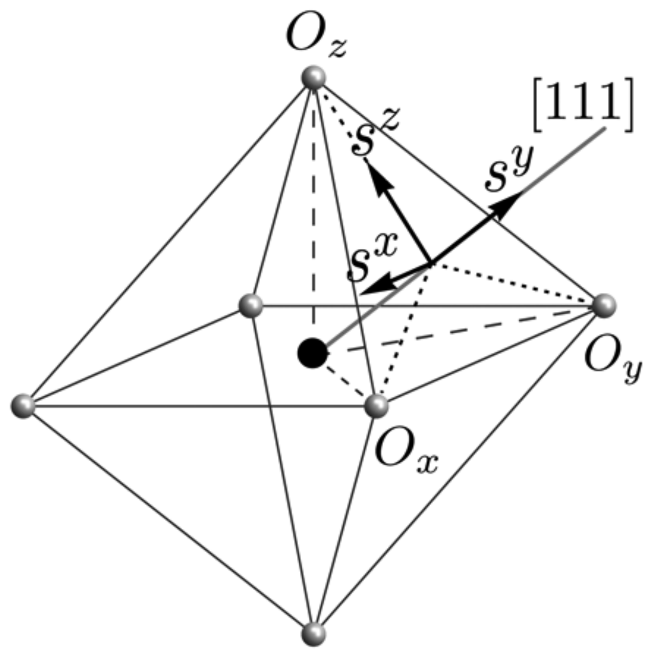

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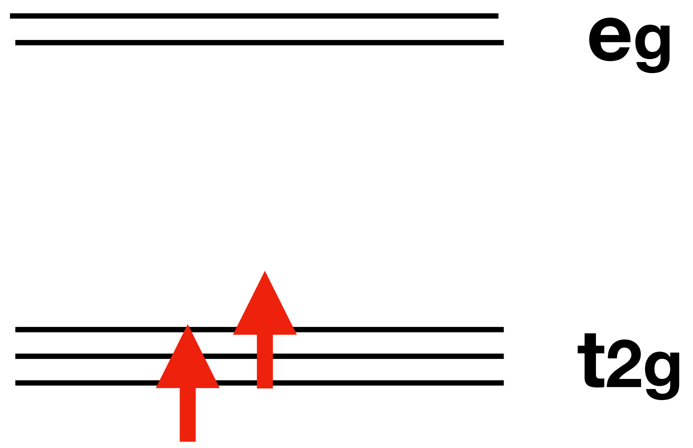

Hund's rule: S= 1, L =1, J=2

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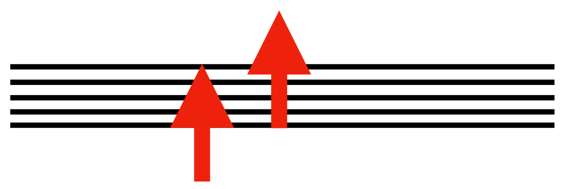

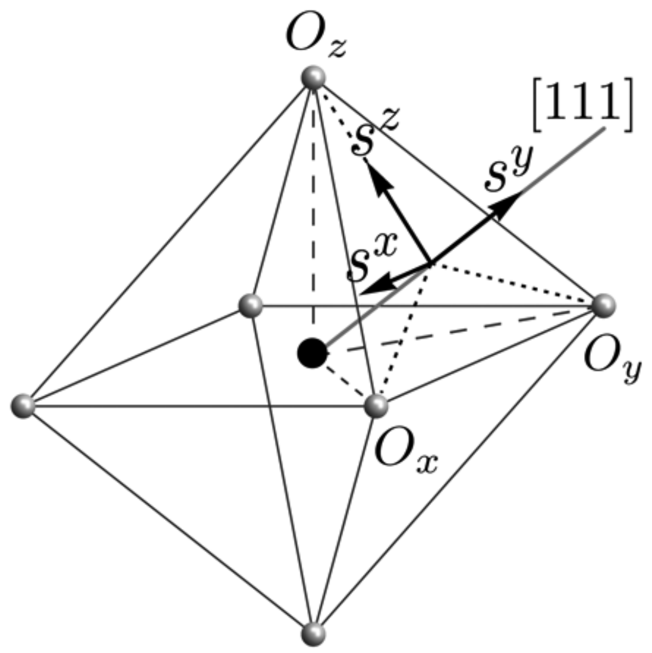

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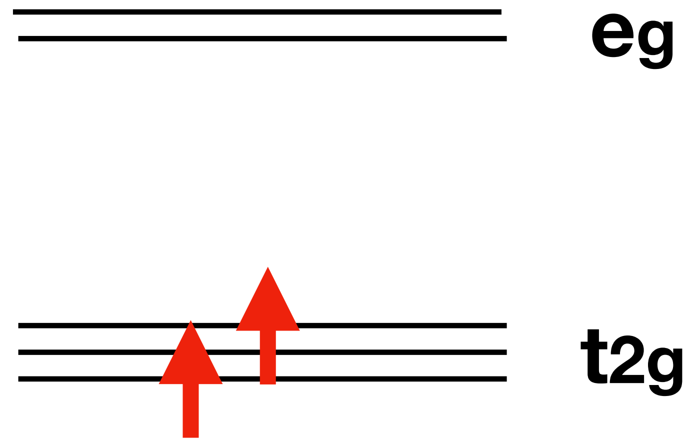

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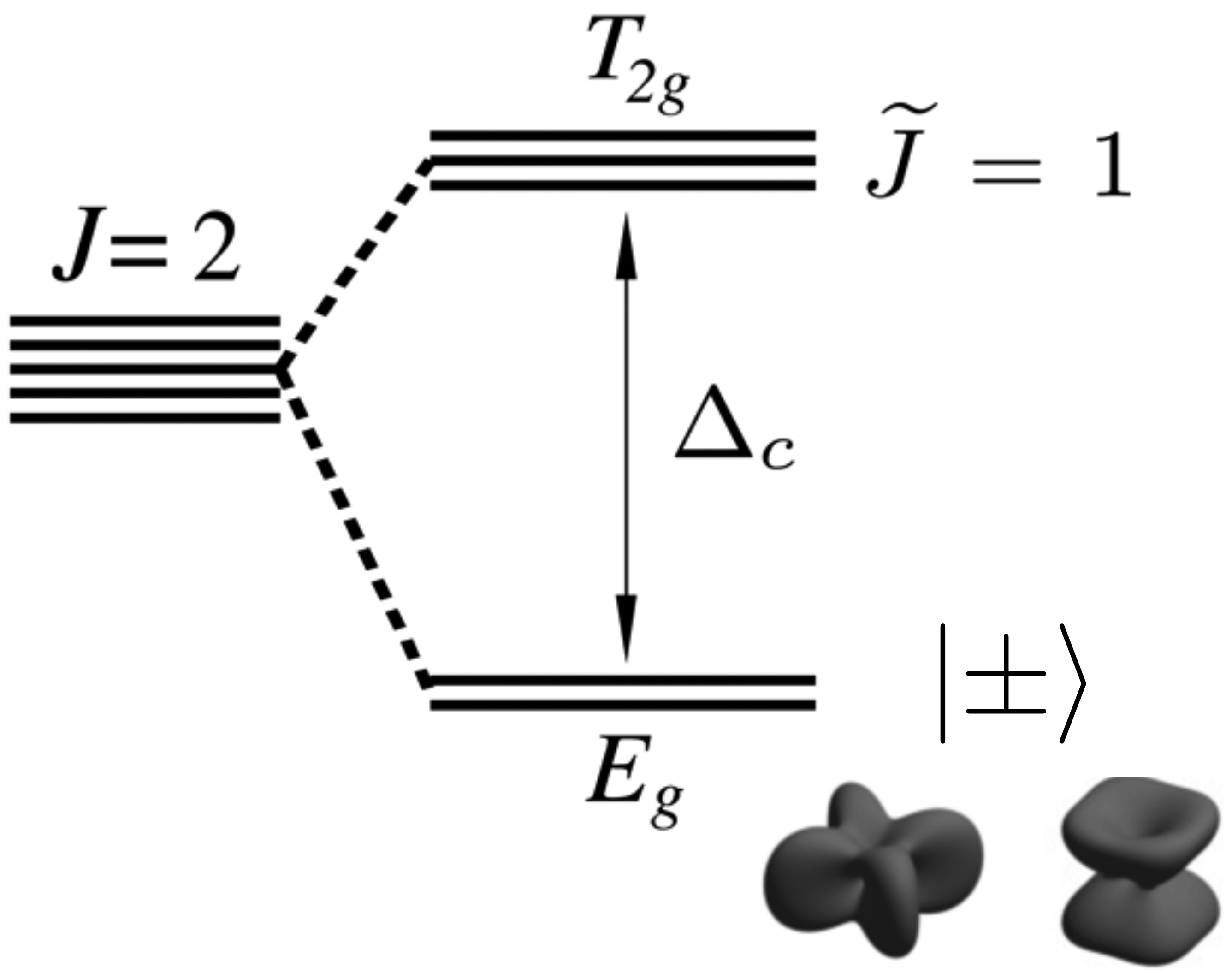

Non-Kramer doublet

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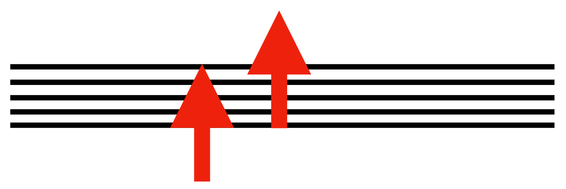

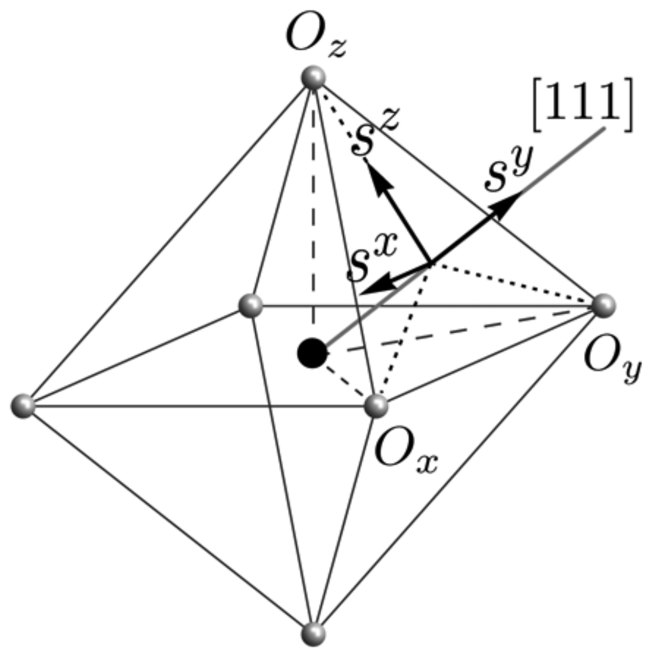

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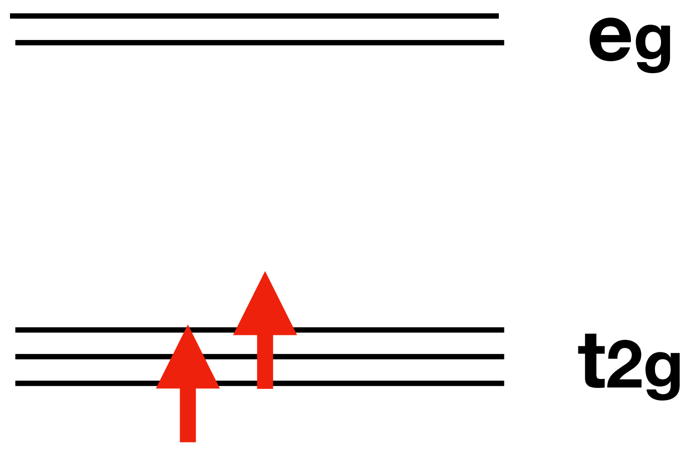

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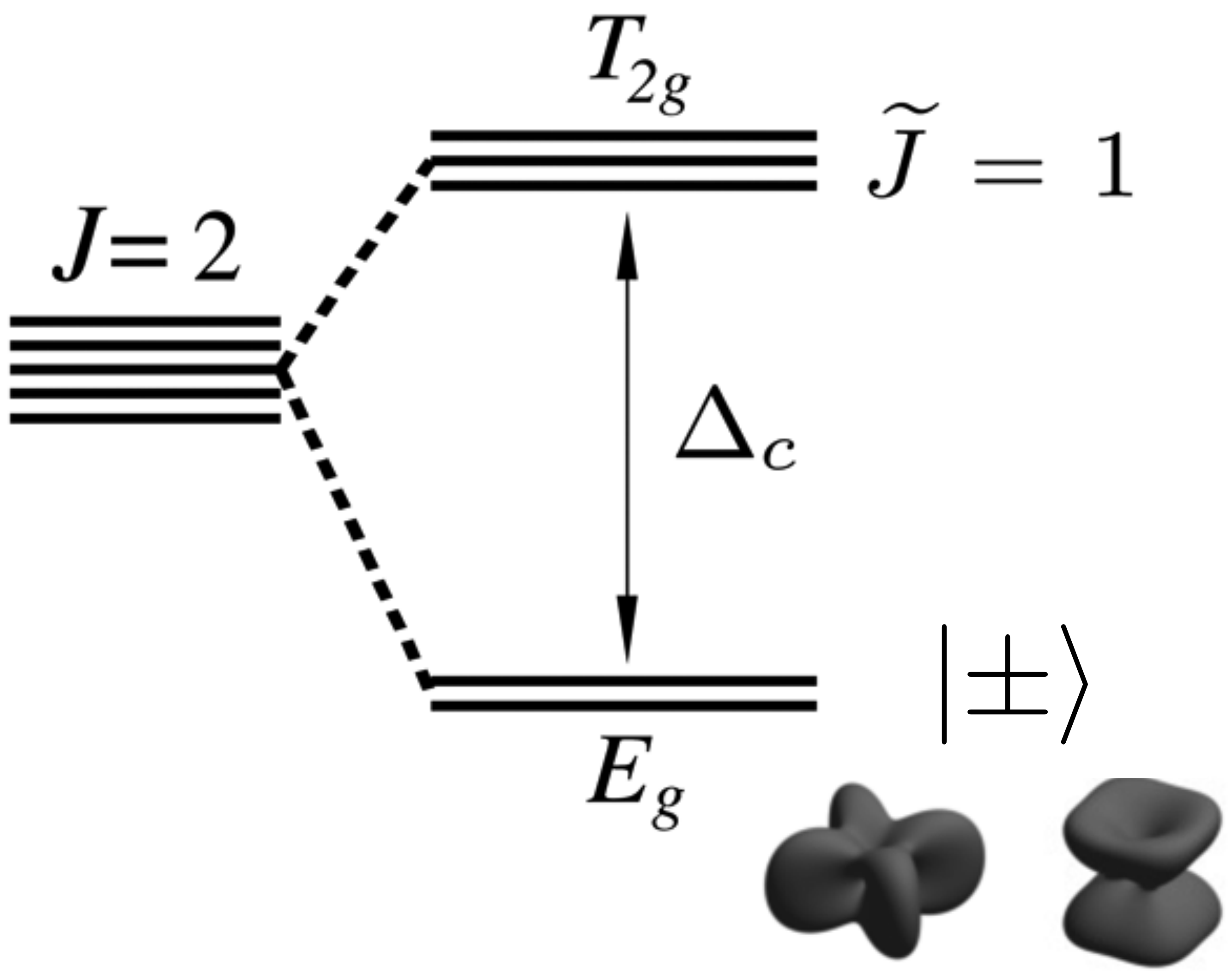

Non-Kramer doublet

$$|+\rangle = \frac{1}{\sqrt{2}}(|1,1\rangle + |-1,-1\rangle),$$

$$|-\rangle = \frac{1}{\sqrt{6}}(|1,-1\rangle + 2|0,0\rangle + |-1,1\rangle).$$

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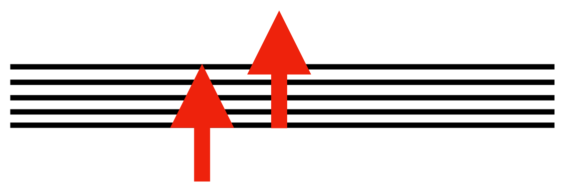

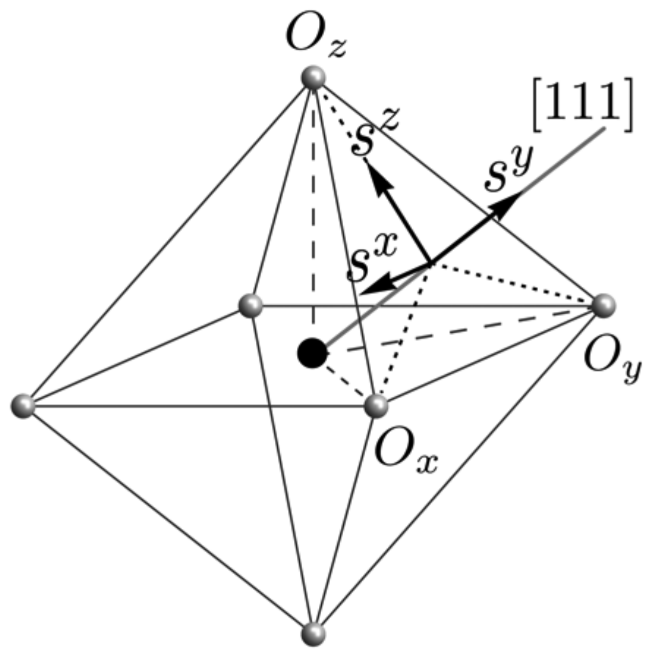

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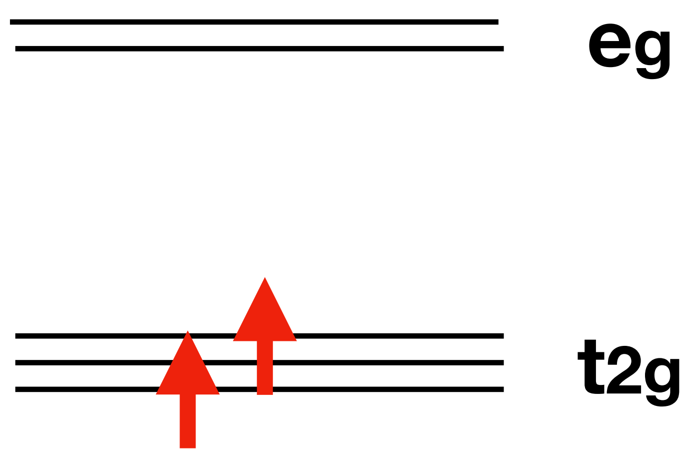

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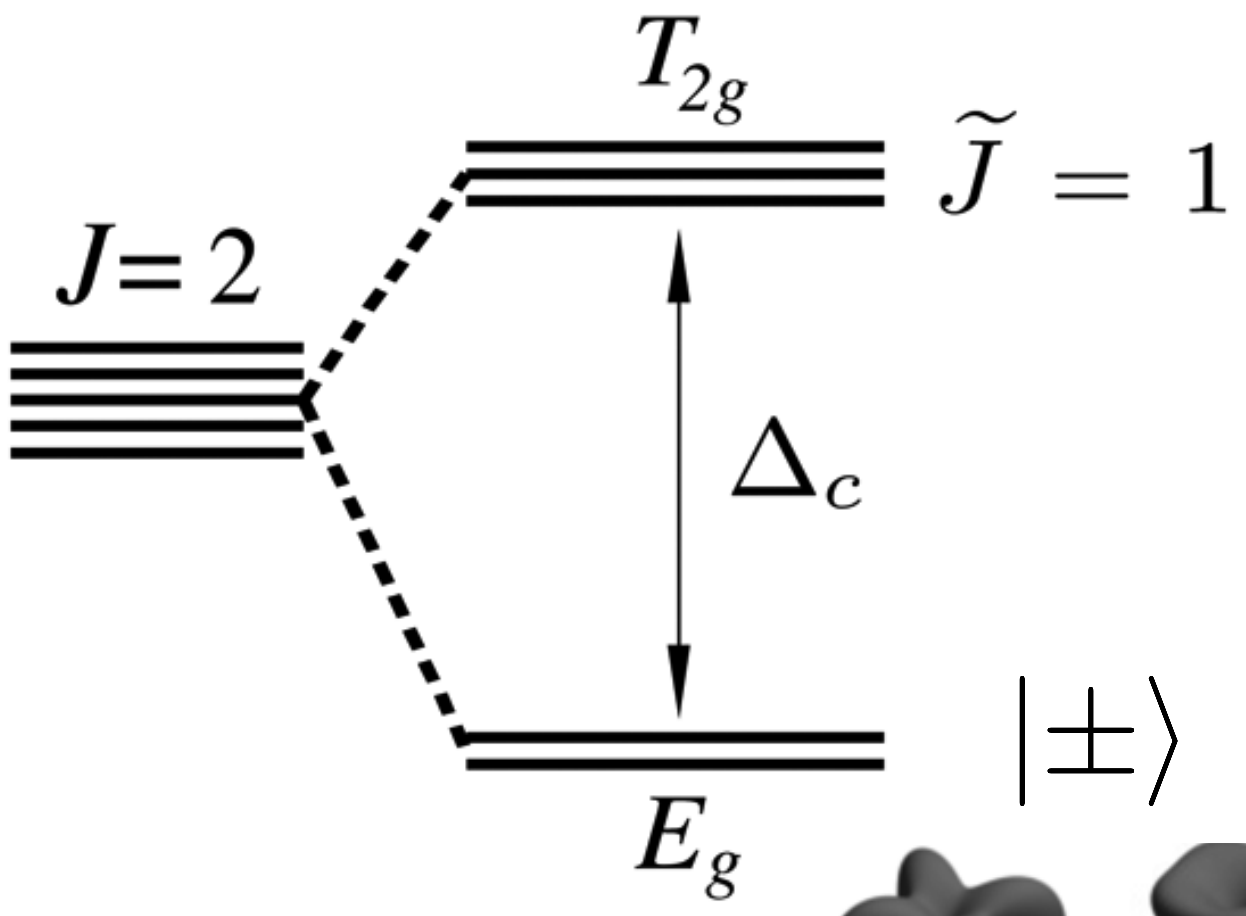

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Quadrupole operators

$$O_3 = \frac{1}{6}(2J_z^2 - J_x^2 - J_y^2) \qquad \langle \pm | O_3 | \pm \rangle = \pm 1$$

$$O_2 = \frac{1}{2\sqrt{3}}(J_x^2 - J_y^2) \qquad \langle \pm | O_2 | \mp \rangle = 1$$

Octuploar operator

$$T_{xyz} = \frac{1}{\sqrt{3}}\overline{J_x J_y J_z} \qquad \langle \pm \frac{1}{2} | T_{xyz} | \mp \frac{1}{2} \rangle = \mp i$$

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Pauli matrix

$$s^z = \frac{1}{2} O_3 \qquad s^x = \frac{1}{2} O_2$$

$$s^y = \frac{1}{2} T_{xyz}$$

$$|\pm\rangle =$$

Non-Kramer doublet

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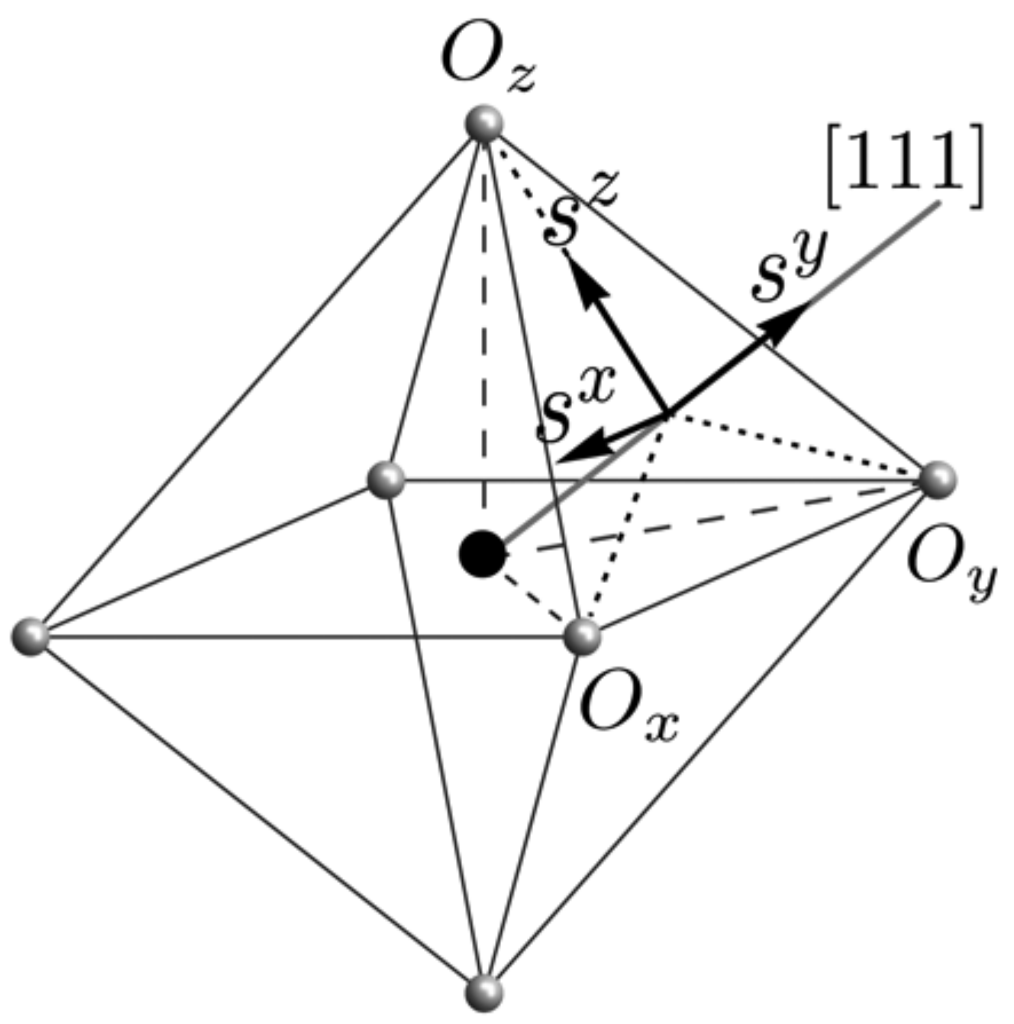

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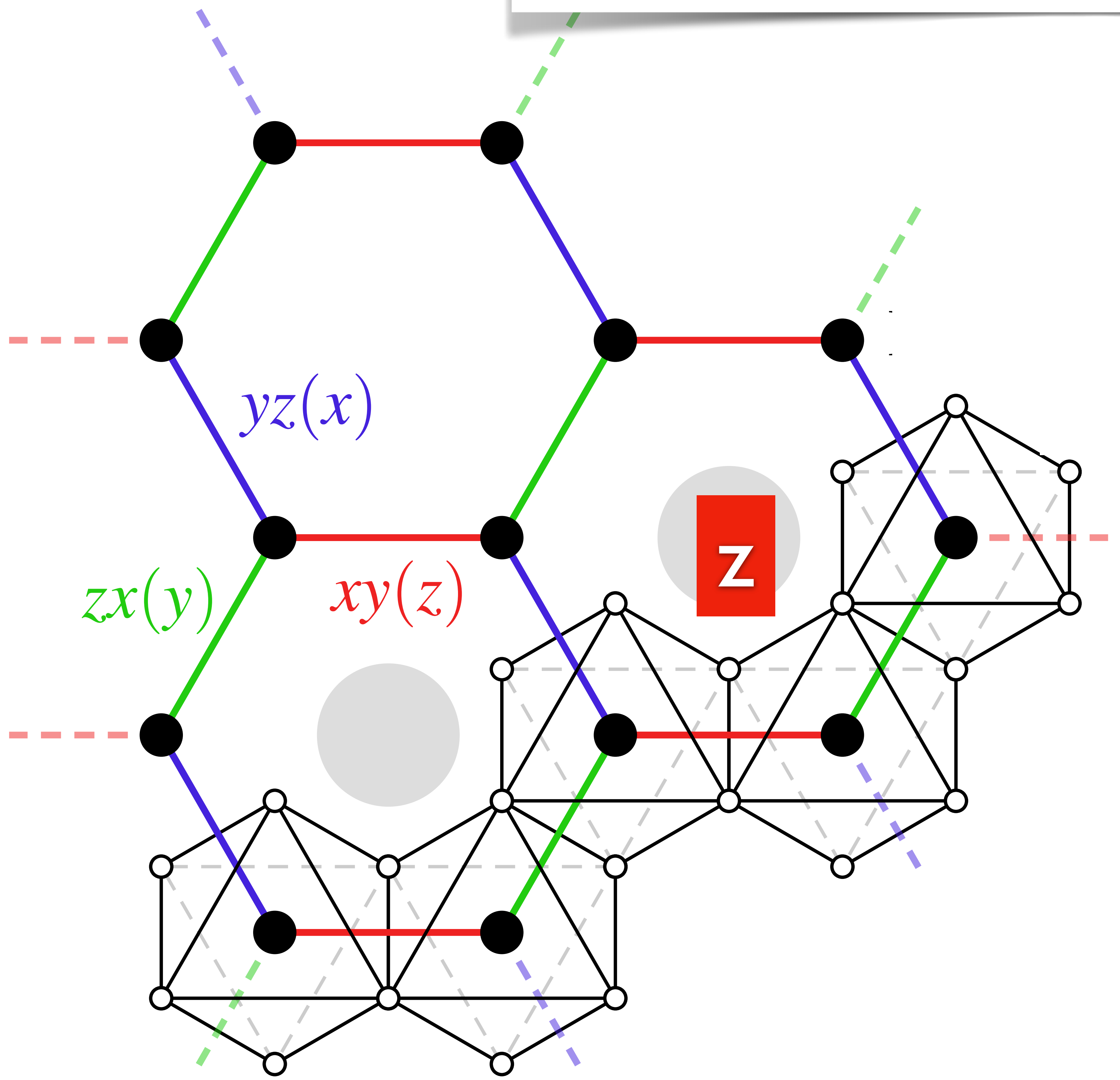
$|\pm\rangle =$ 

Non-Kramer doublet

intersite H

$$\mathcal{H}_{ij}$$

Microscopic Theory of 5d² systems



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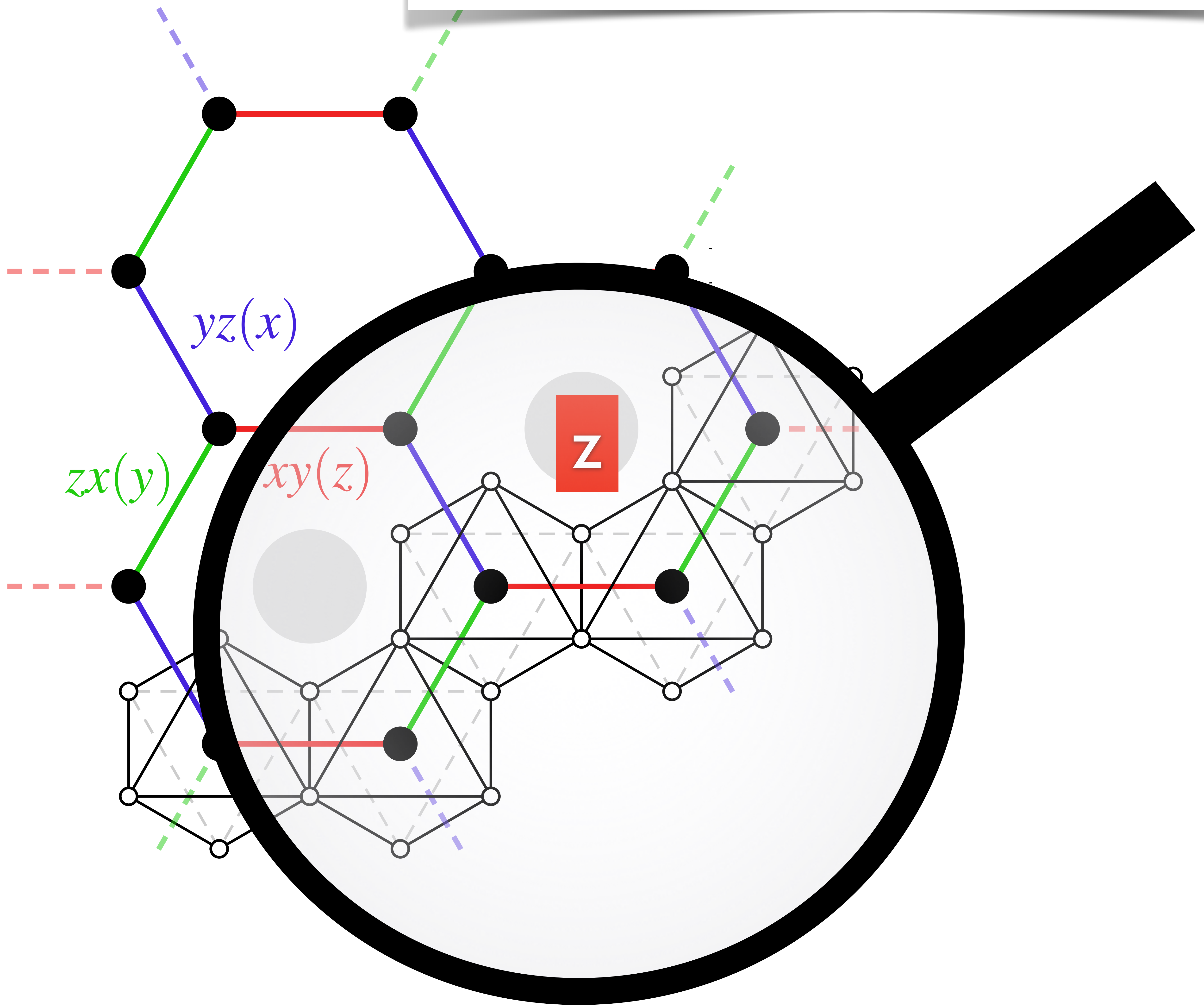

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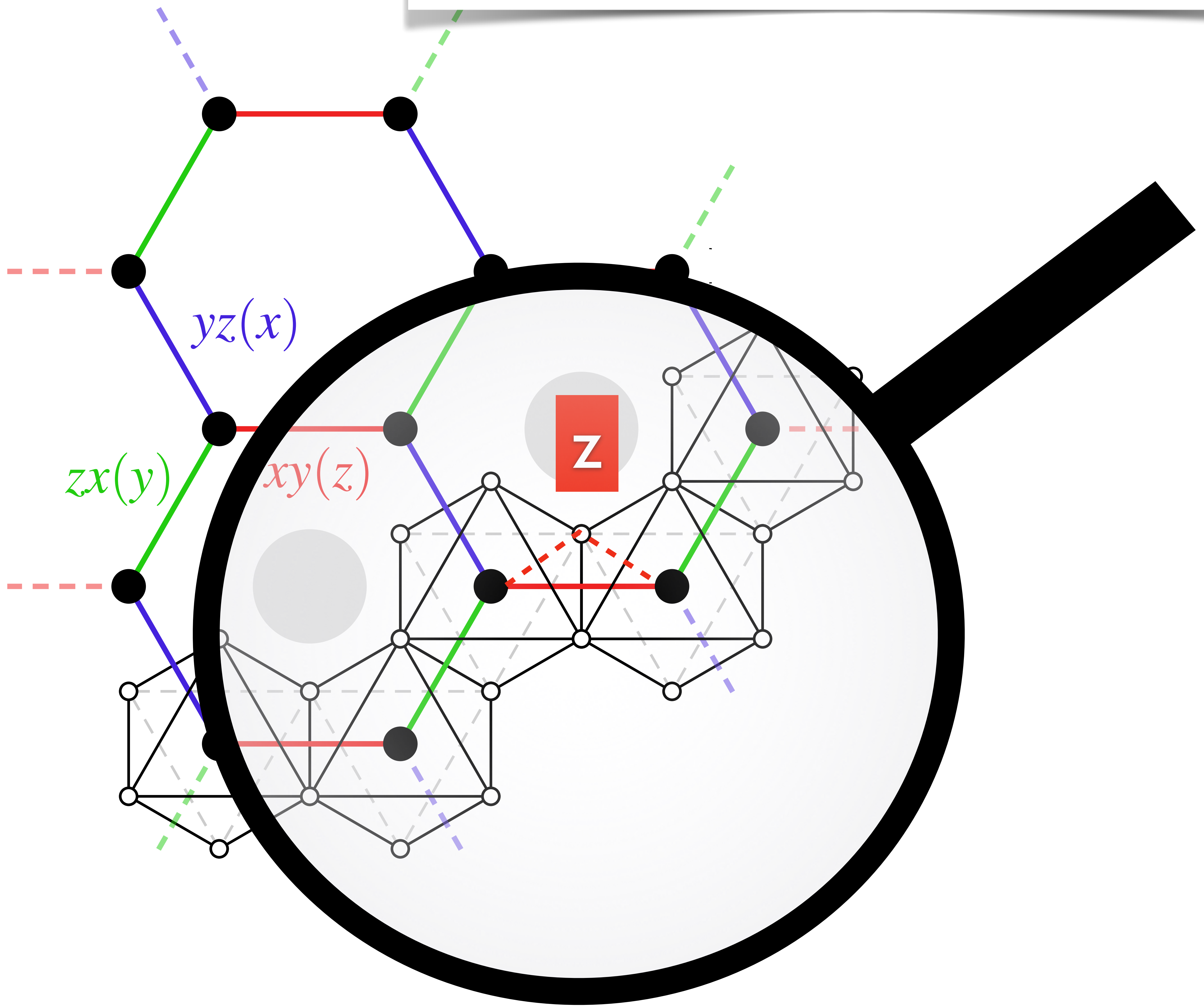

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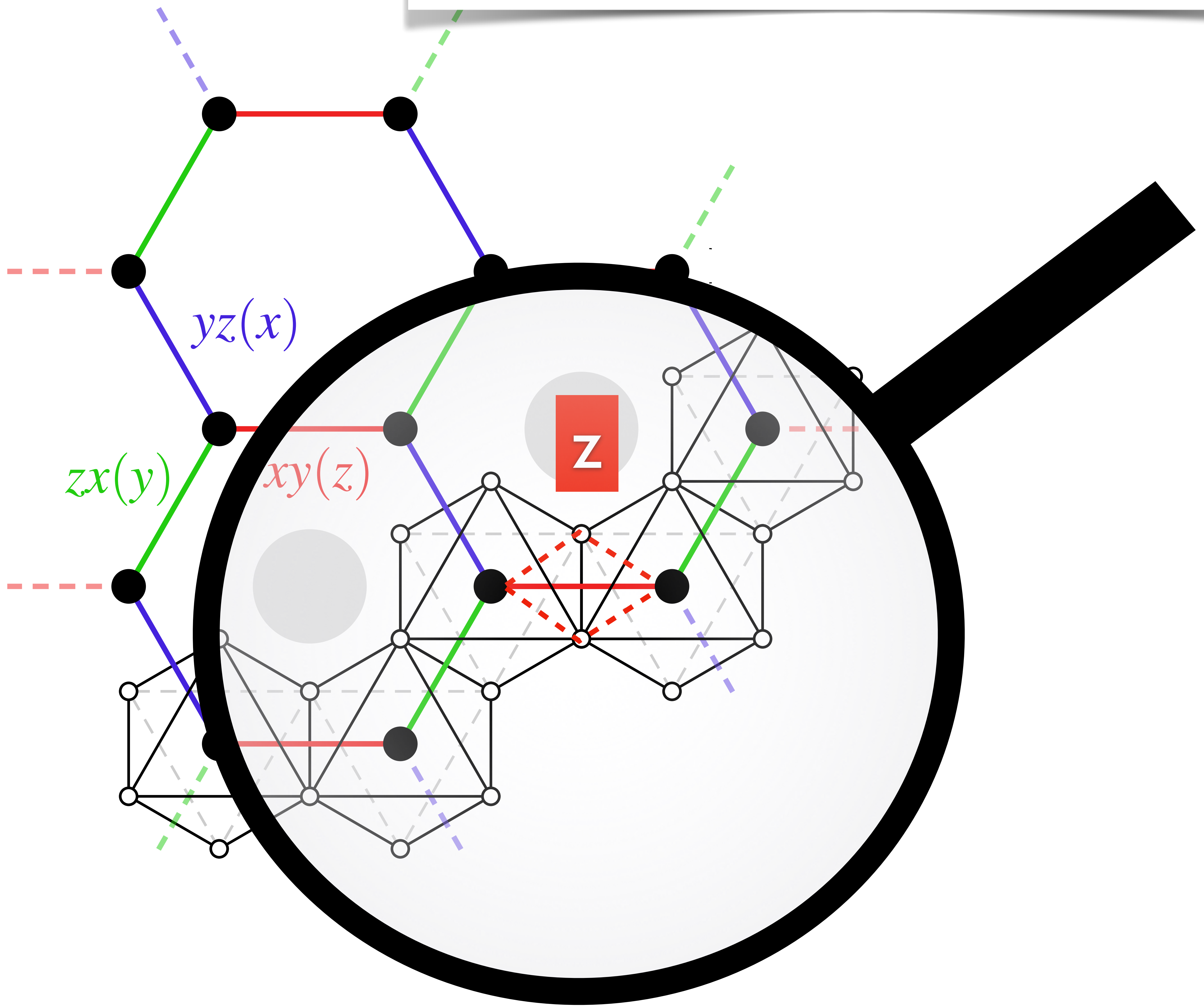

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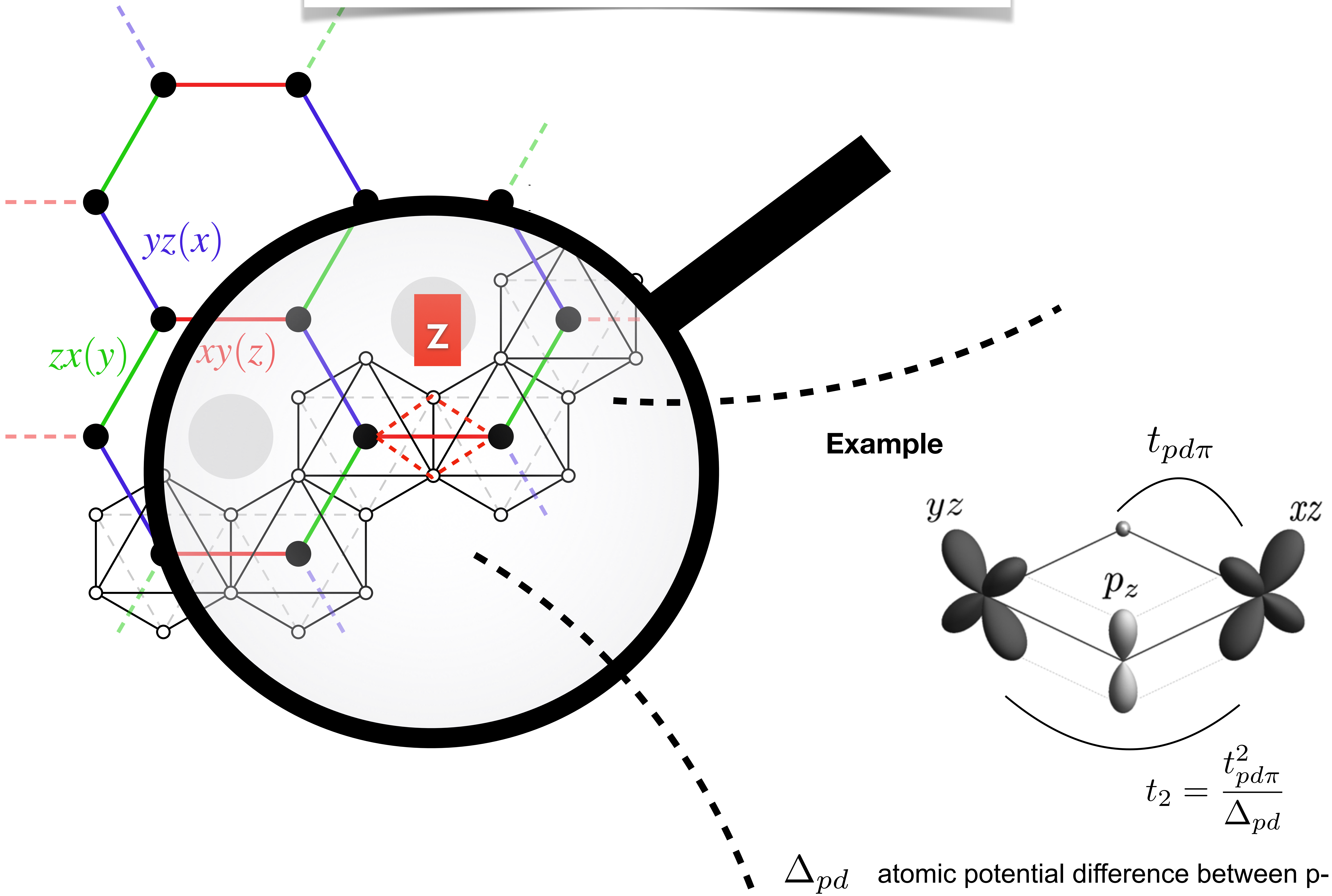


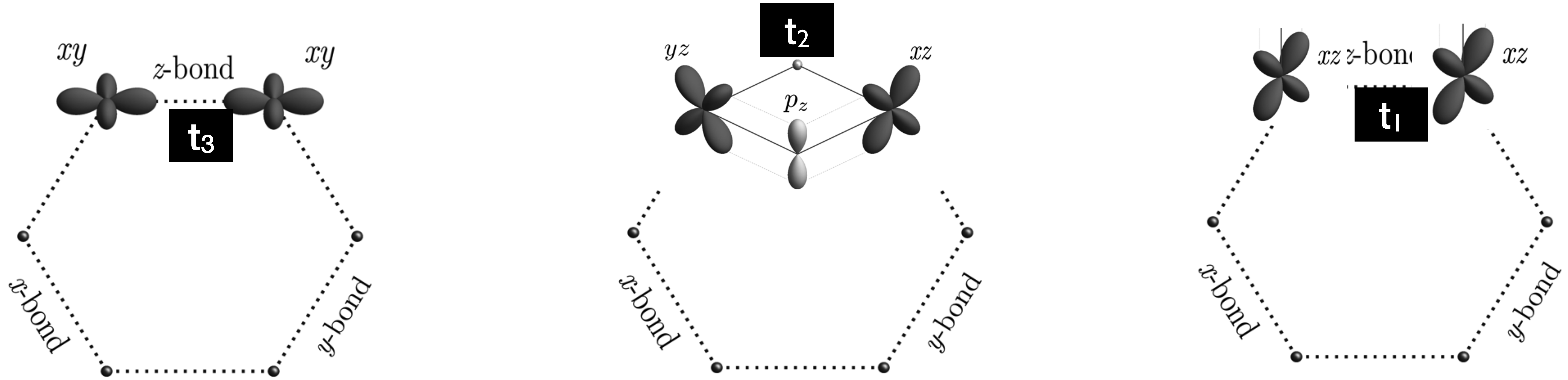
$$t_2 = \frac{t_{pd\pi}^2}{\Delta_{pd}}$$

Δ_{pd} atomic potential difference between p- and d-orbitals

Recall : there are 2 electrons in t_{2g} orbitals at a site

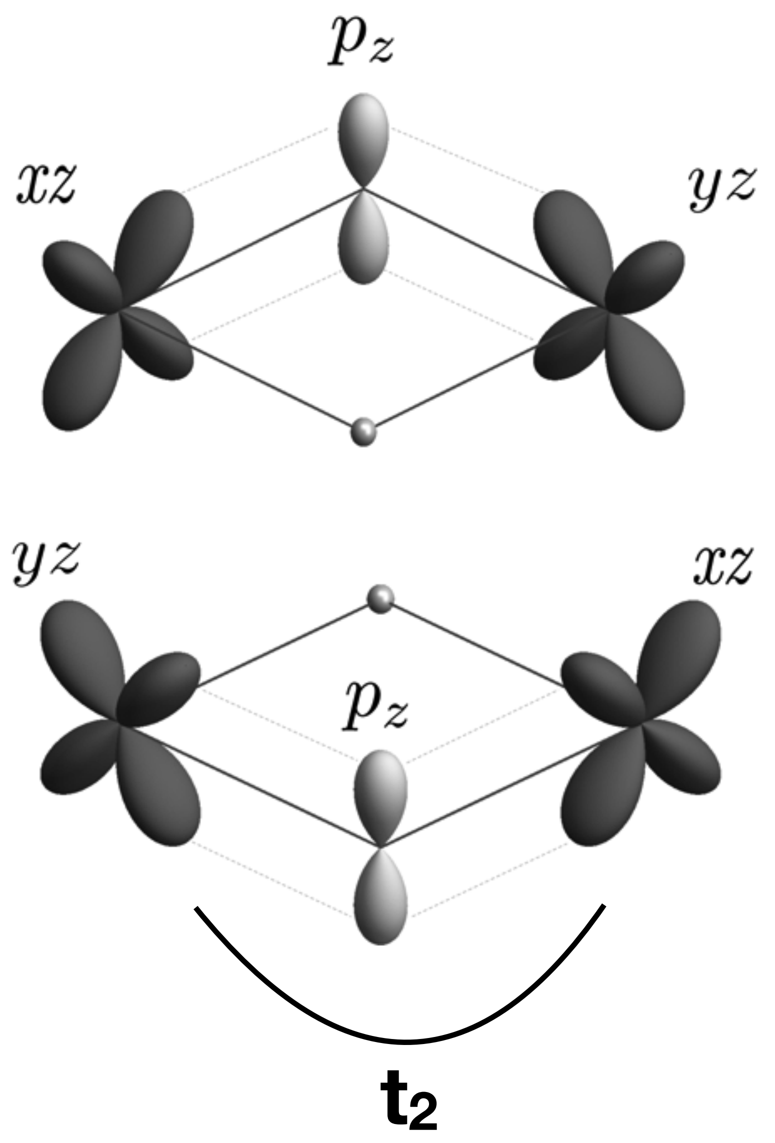
Consider hopping integrals between two sites (z bond)

$$t_{ij} = \begin{array}{c} \\ c^\dagger_{i,xy} \\ c^\dagger_{i,xz} \\ c^\dagger_{i,yz} \end{array} \begin{array}{c} \begin{array}{ccc} c_{j,xy} & c_{j,xz} & c_{j,yz} \end{array} \\ \begin{pmatrix} t_3 & 0 & 0 \\ 0 & t_1 & t_2 \\ 0 & t_2 & t_1 \end{pmatrix} \end{array}$$



I. inter-orbital super-exchange (t_2)

$$t_{ij} = \begin{matrix} & c_{j,xy} & c_{j,xz} & c_{j,yz} \\ \begin{matrix} c^\dagger_{i,xy} \\ c^\dagger_{i,xz} \\ c^\dagger_{i,yz} \end{matrix} & \multicolumn{3}{c}{\begin{pmatrix} t_3 & 0 & 0 \\ 0 & t_1 & t_2 \\ 0 & t_2 & t_1 \end{pmatrix}} \end{matrix}$$



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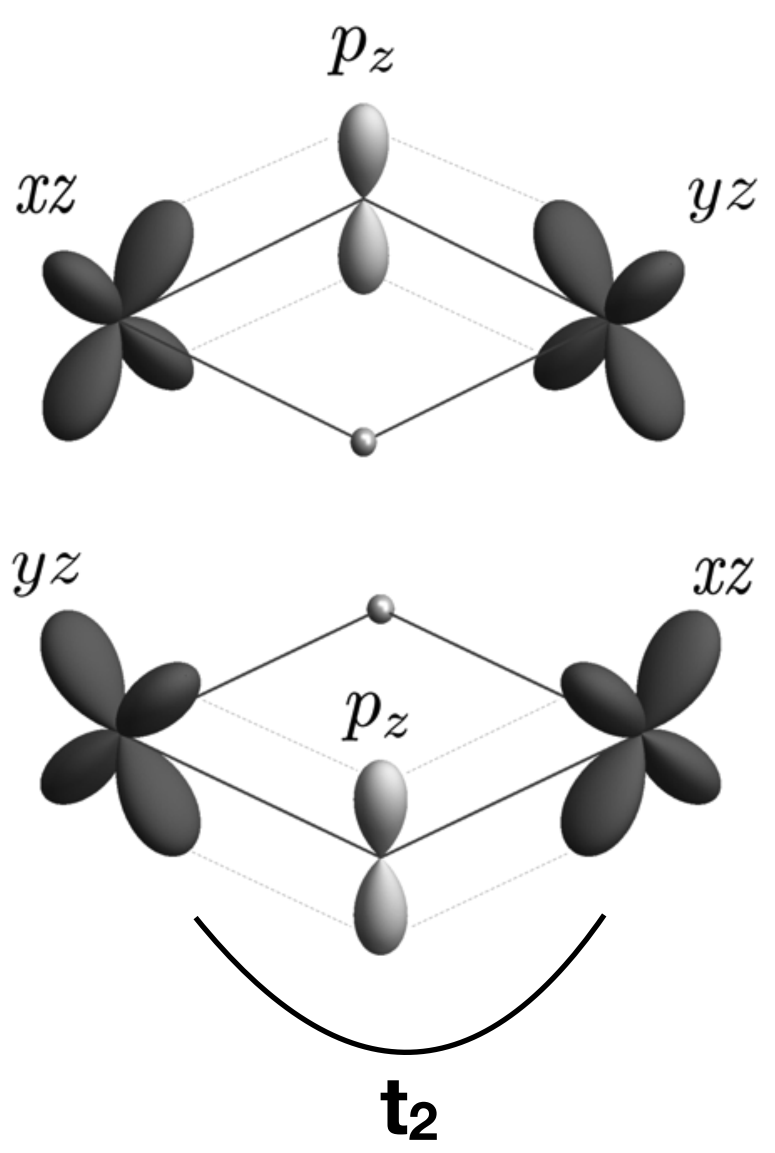

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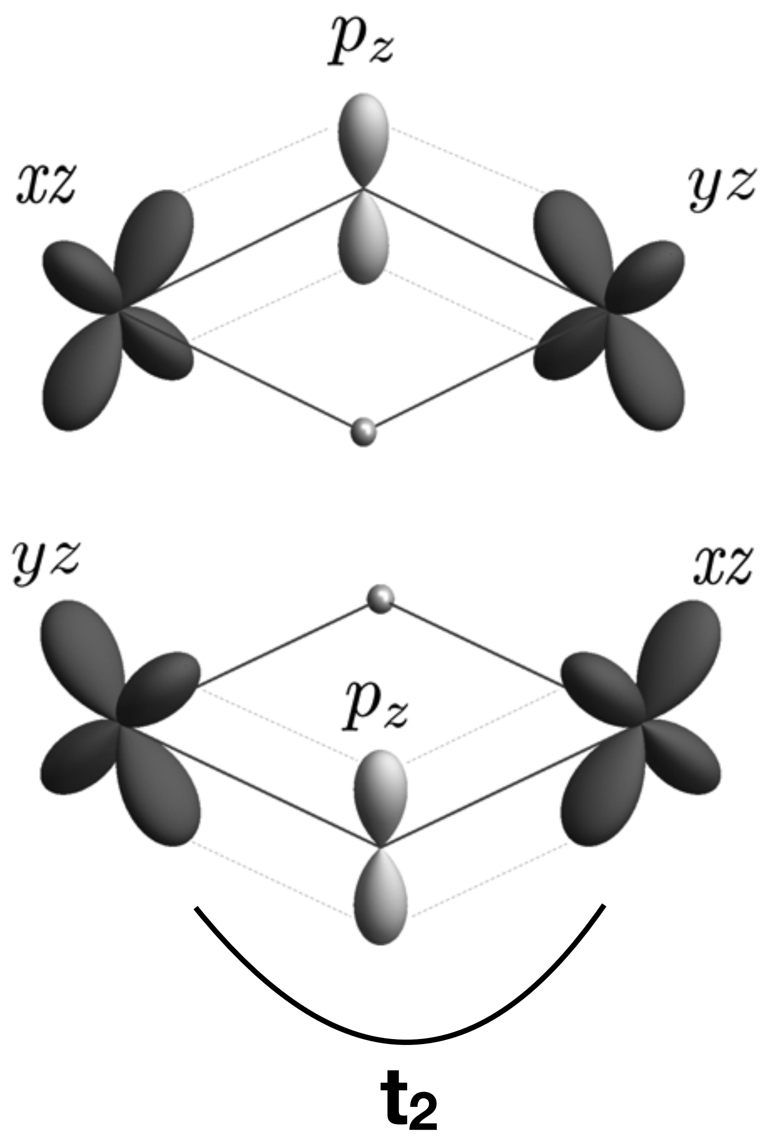
$$\mathcal{H}_{ij}(90^\circ) = J\left(s_i^y s_j^y - s_i^x s_j^x - s_i^z s_j^z\right), \qquad J = \frac{2t_2^2}{3U}$$



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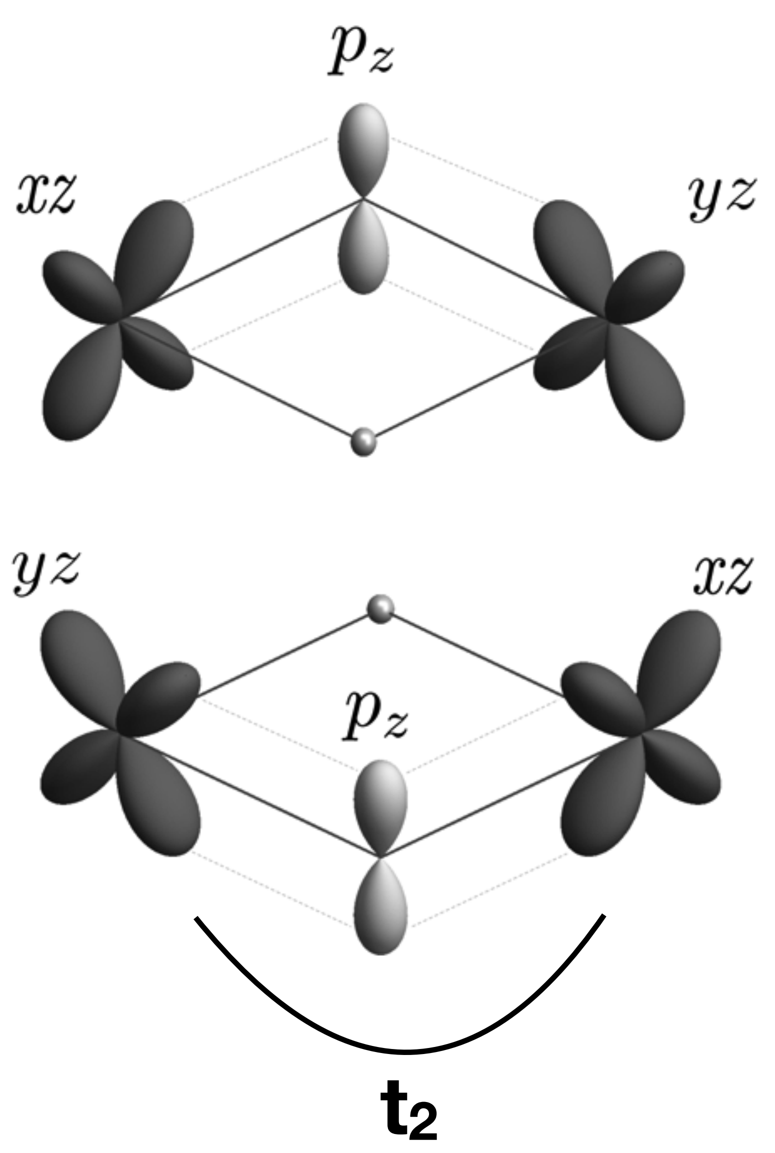
1/2(Pauli matrix) for non-Kramer doublet



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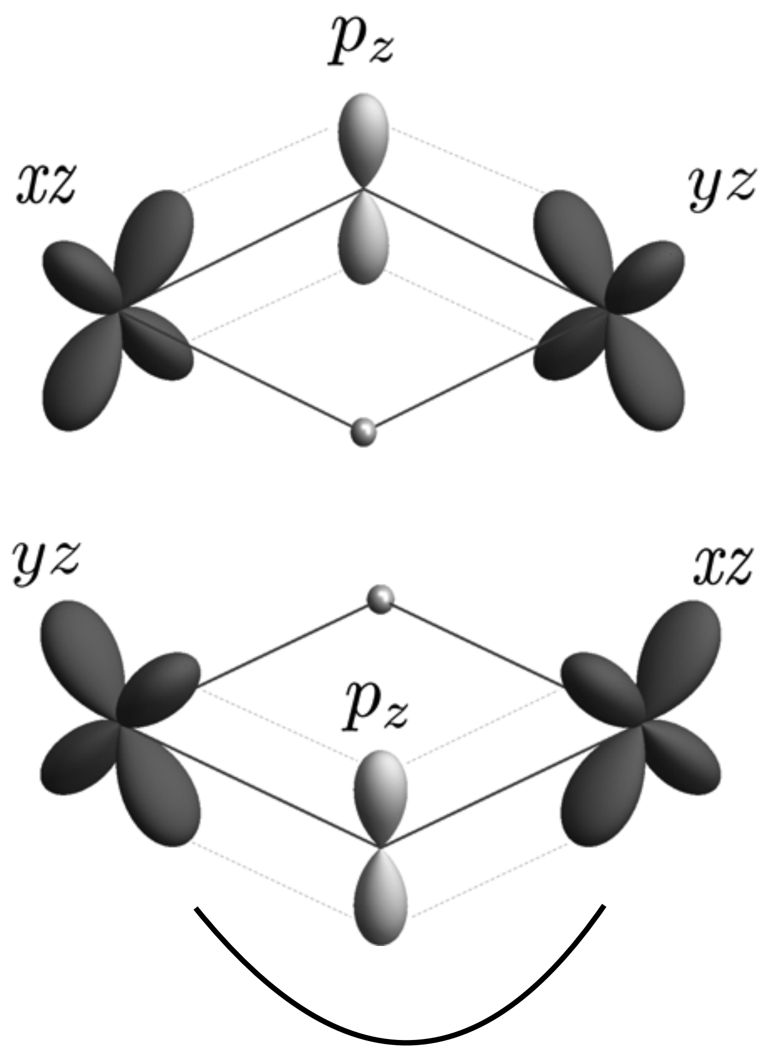
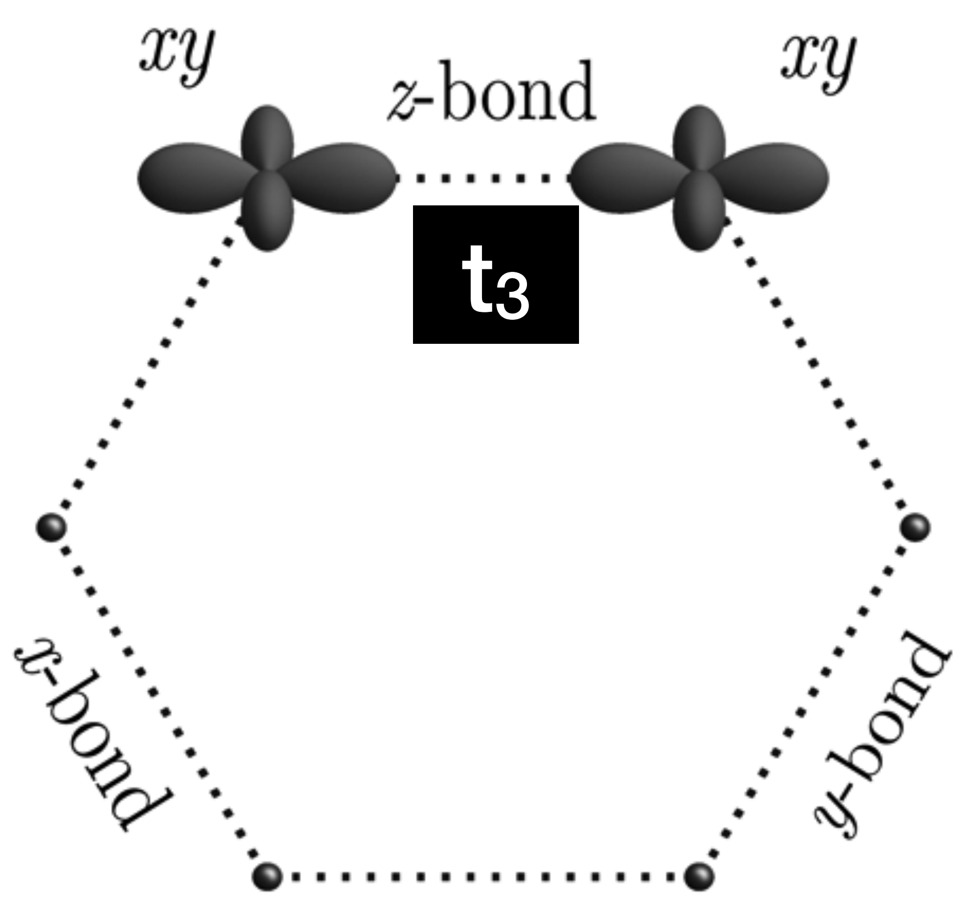


t_2

AFM octupolar and FM quadrupolar interaction

support AF Octupolar Order when J is large

2. direct exchange (t_3)



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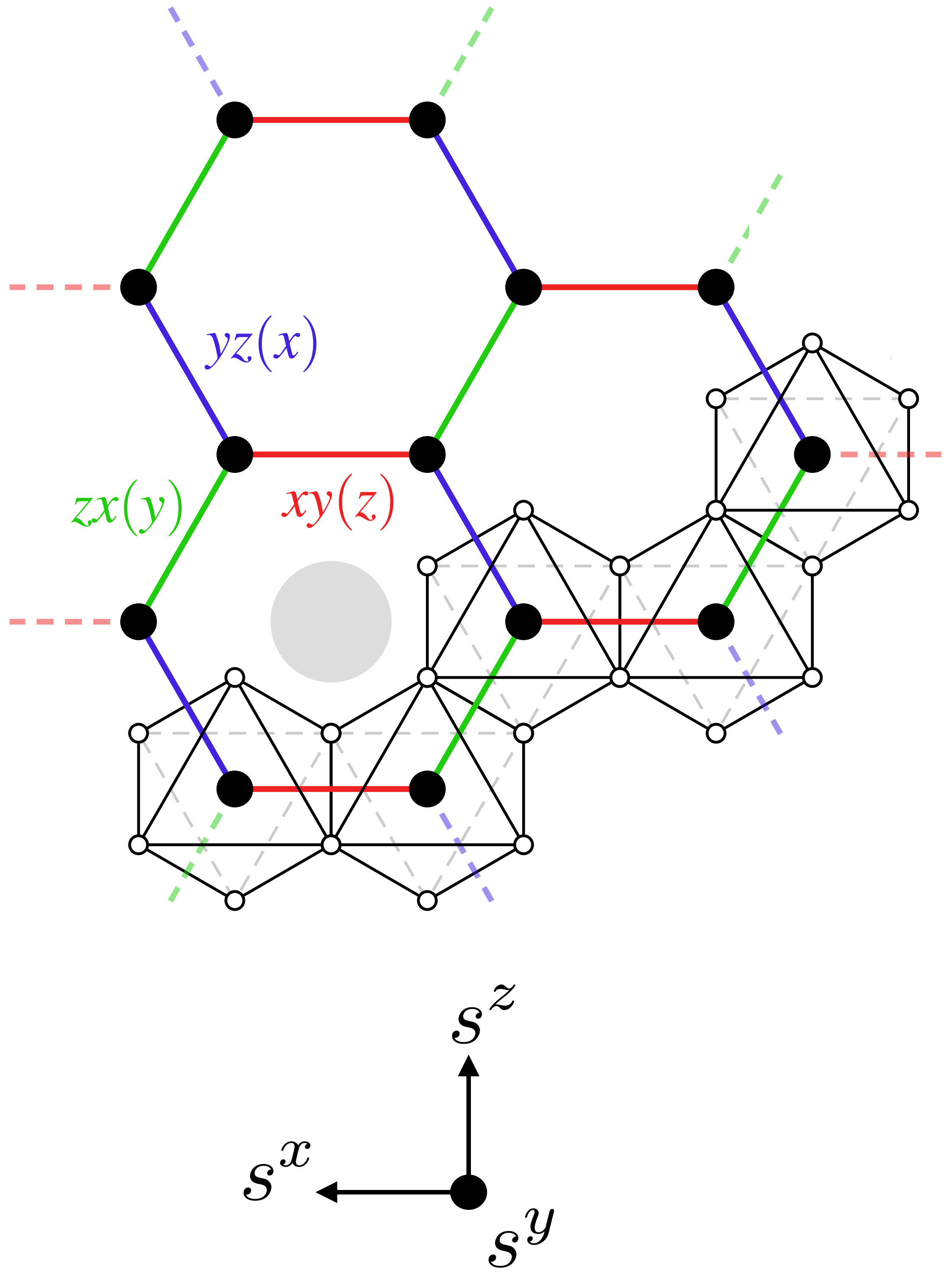

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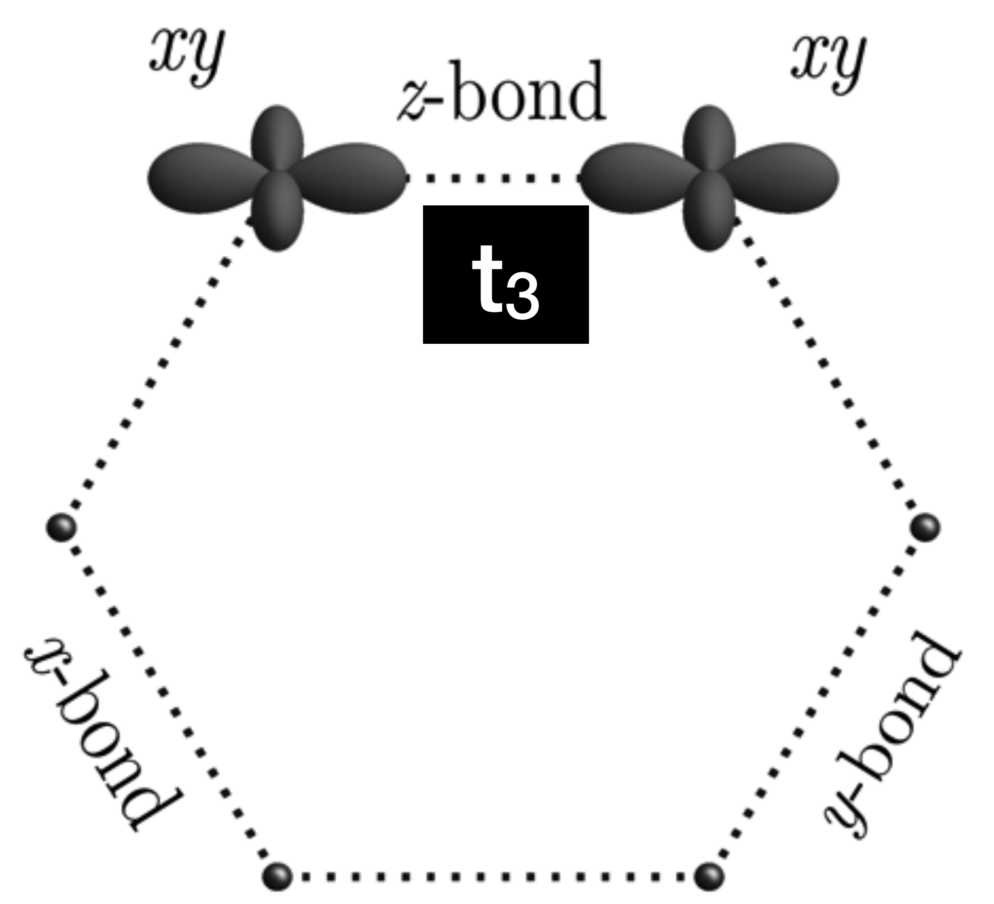
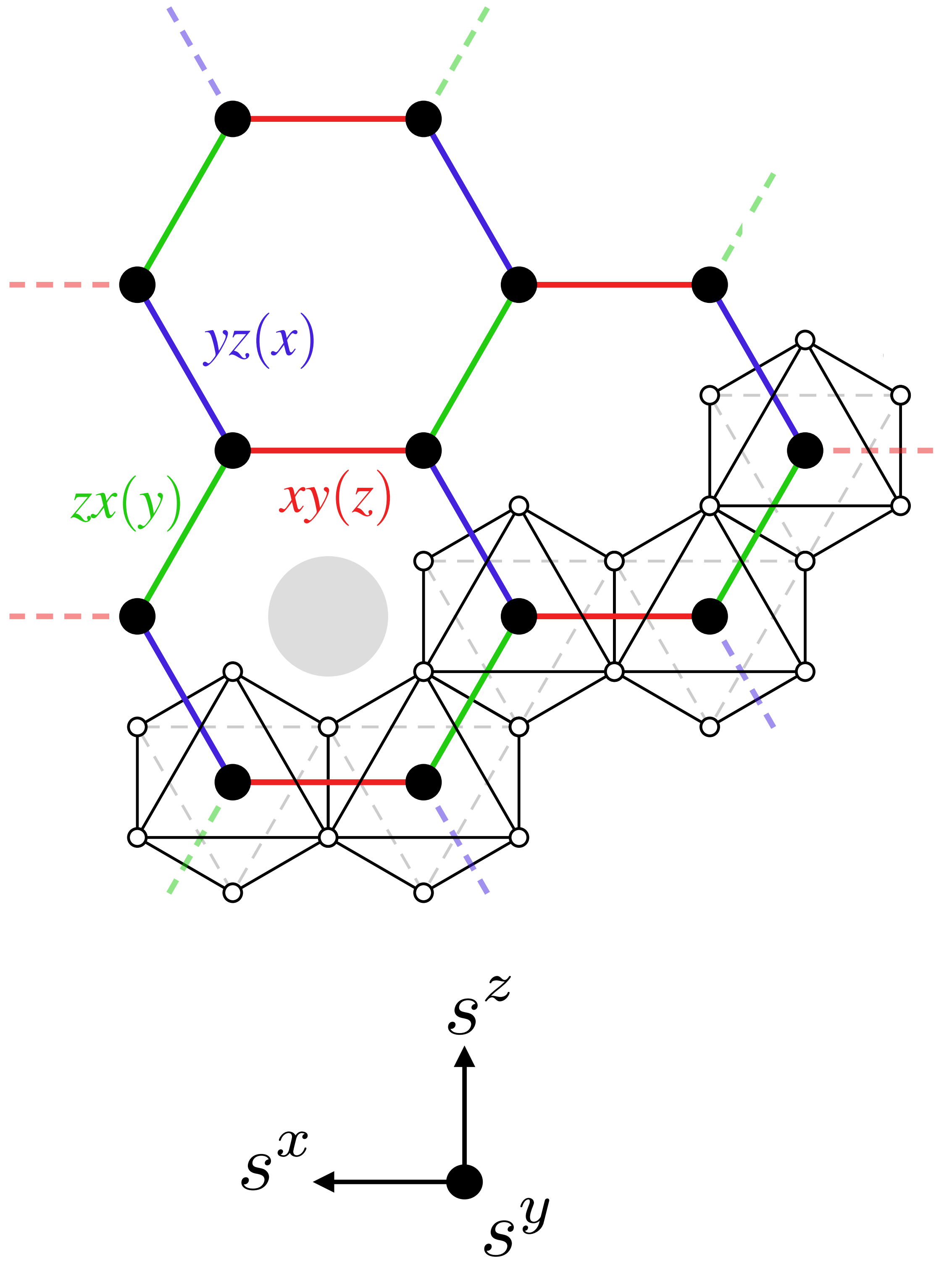


$$t_{ij} = \begin{array}{c} \\ c^\dagger_{i,xy} \\ c^\dagger_{i,xz} \\ c^\dagger_{i,yz} \end{array} \begin{array}{c} \begin{array}{ccc} c_{j,xy} & c_{j,xz} & c_{j,yz} \end{array} \\ \begin{pmatrix} t_3 & 0 & 0 \\ 0 & t_1 & t_2 \\ 0 & t_2 & t_1 \end{pmatrix} \end{array}$$



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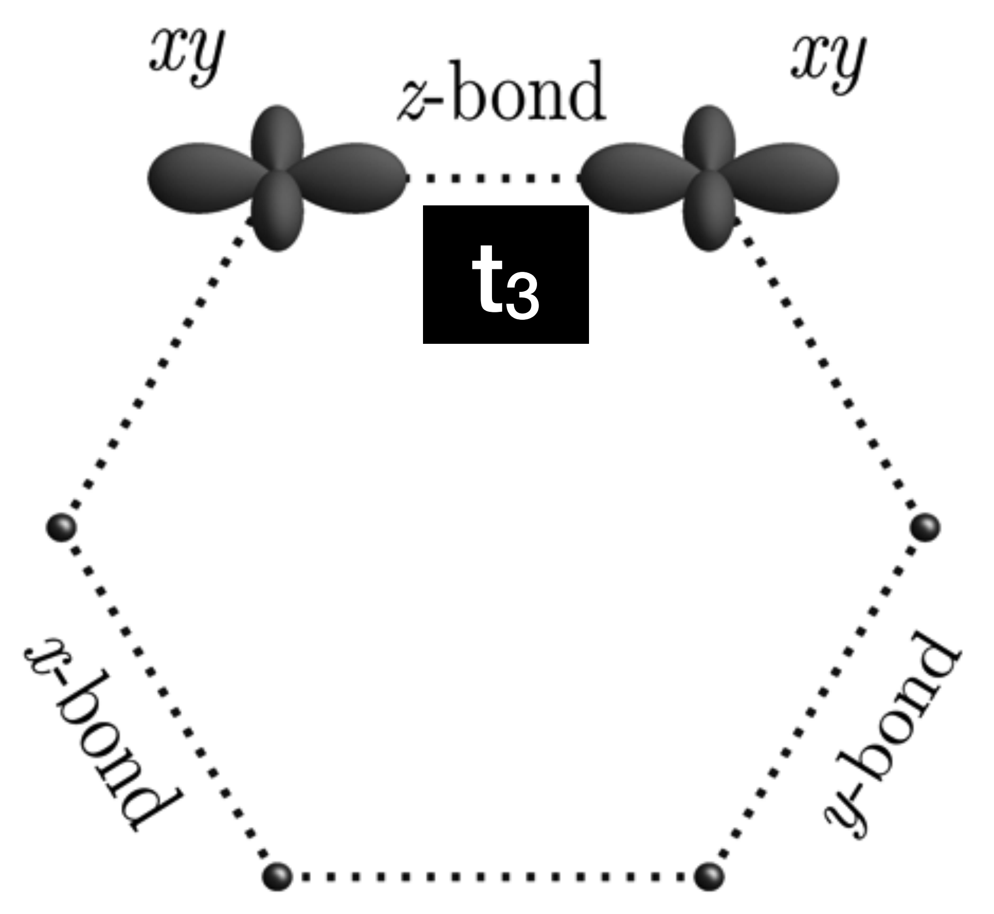

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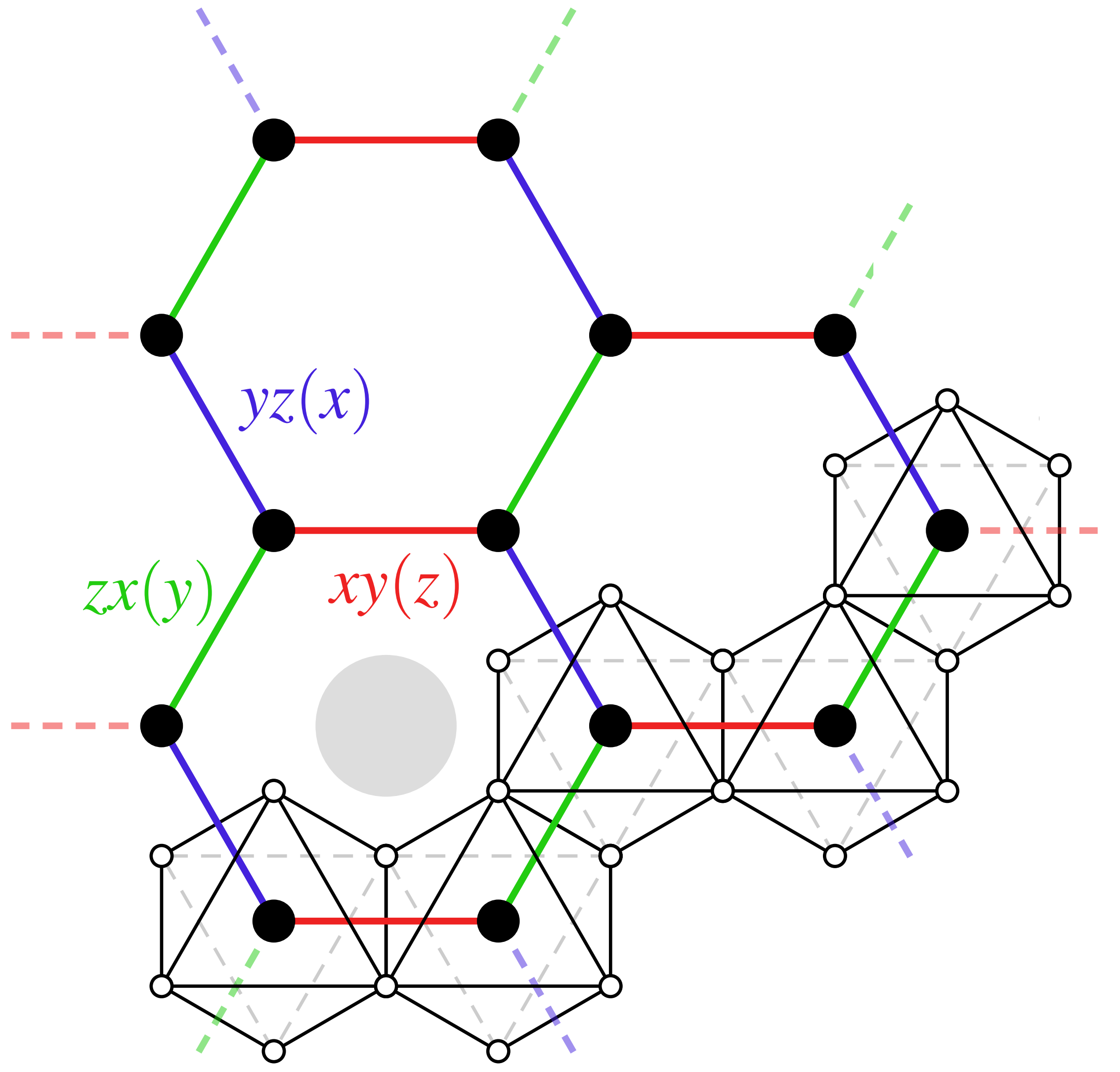
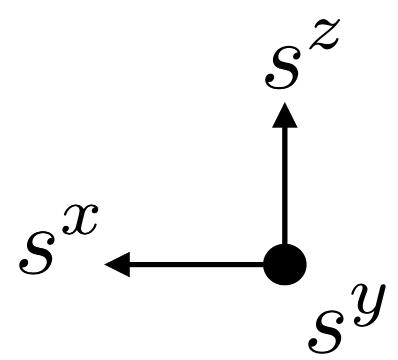


$$\boxed{\mathcal{H}^{(\gamma)}_{ij}(d) = J_\tau \; \tau_{i\gamma} \tau_{j\gamma},} \qquad J_\tau = \frac{4t_3^2}{9U}$$



$$\tau_\gamma = \cos\phi_\gamma \; s^z + \sin\phi_\gamma \; s^x,$$

$$\phi_\gamma = (0, 2\pi/3, 4\pi/3)$$

$$\gamma = (z, x, y)$$

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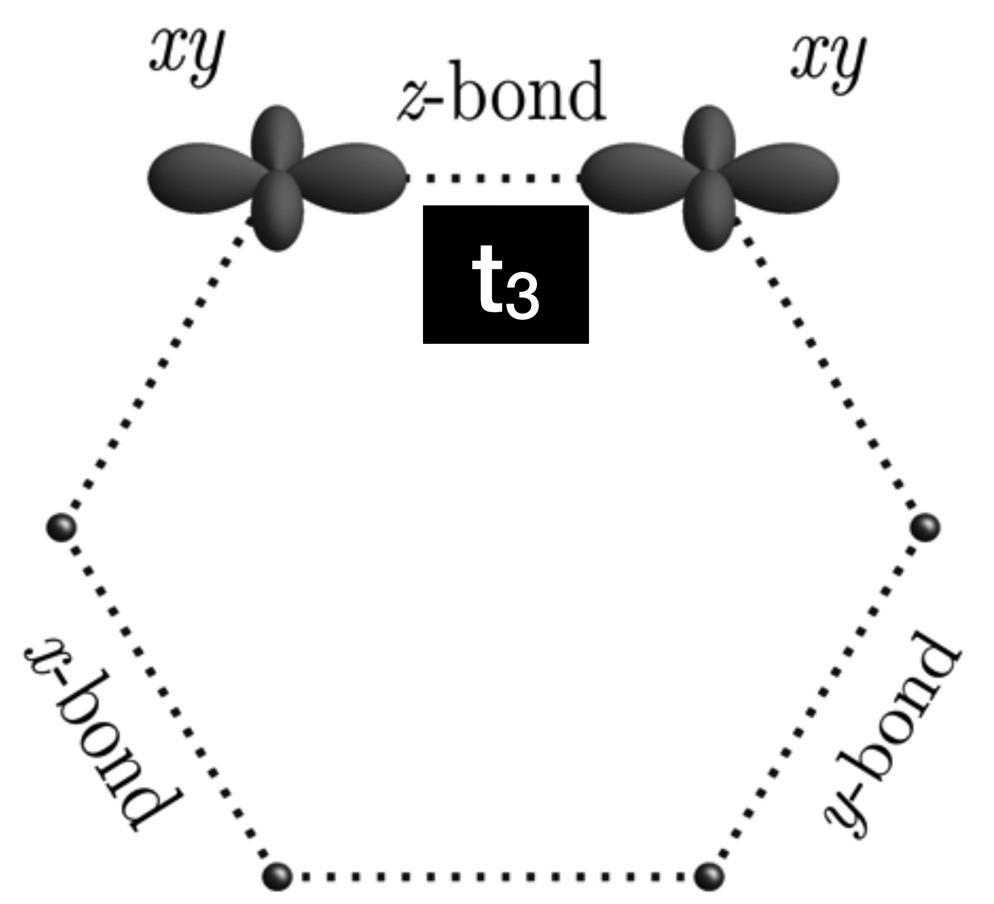

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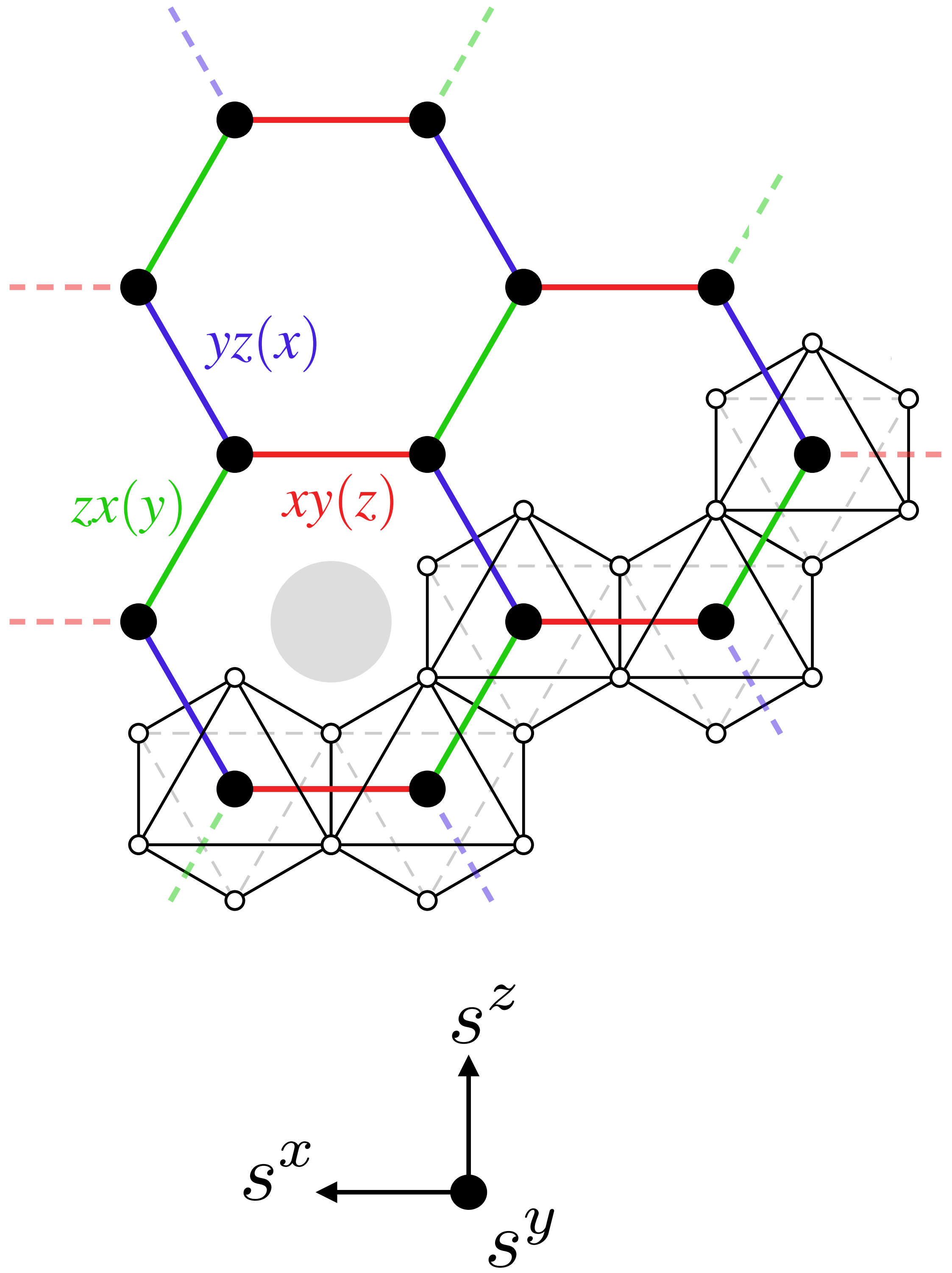

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bond-dependent quadrupole-quadrupole interaction

t_2+t_3 processes together:

$$H_{ij}^{\gamma} = \left(\frac{J_\tau}{2} - J\right)\left(s_i^z s_j^z + s_i^x s_j^x\right) + J s_i^y s_j^y$$
$$+\frac{J_\tau}{2}\left(\cos\phi_\gamma\left(s_i^z s_j^z - s_i^x s_j^x\right) - \sin\phi_\gamma\left(s_i^z s_j^x + s_i^x s_j^z\right)\right)$$

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G. Khaliullin, D. Churchill, P. Stavropoulos, HYK, PRR 3, 033163 (2021)

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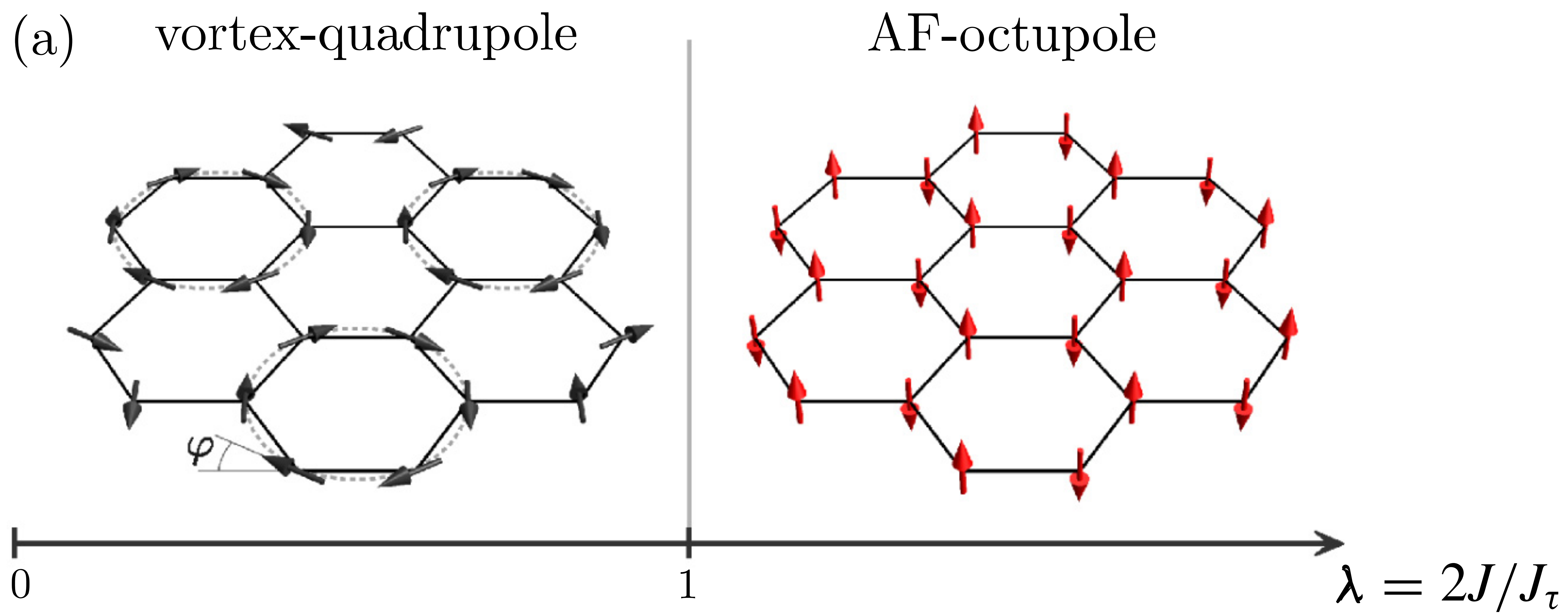

G. Khaliullin, D. Churchill, P. Stavropoulos, HYK, PRR 3, 033163 (2021)

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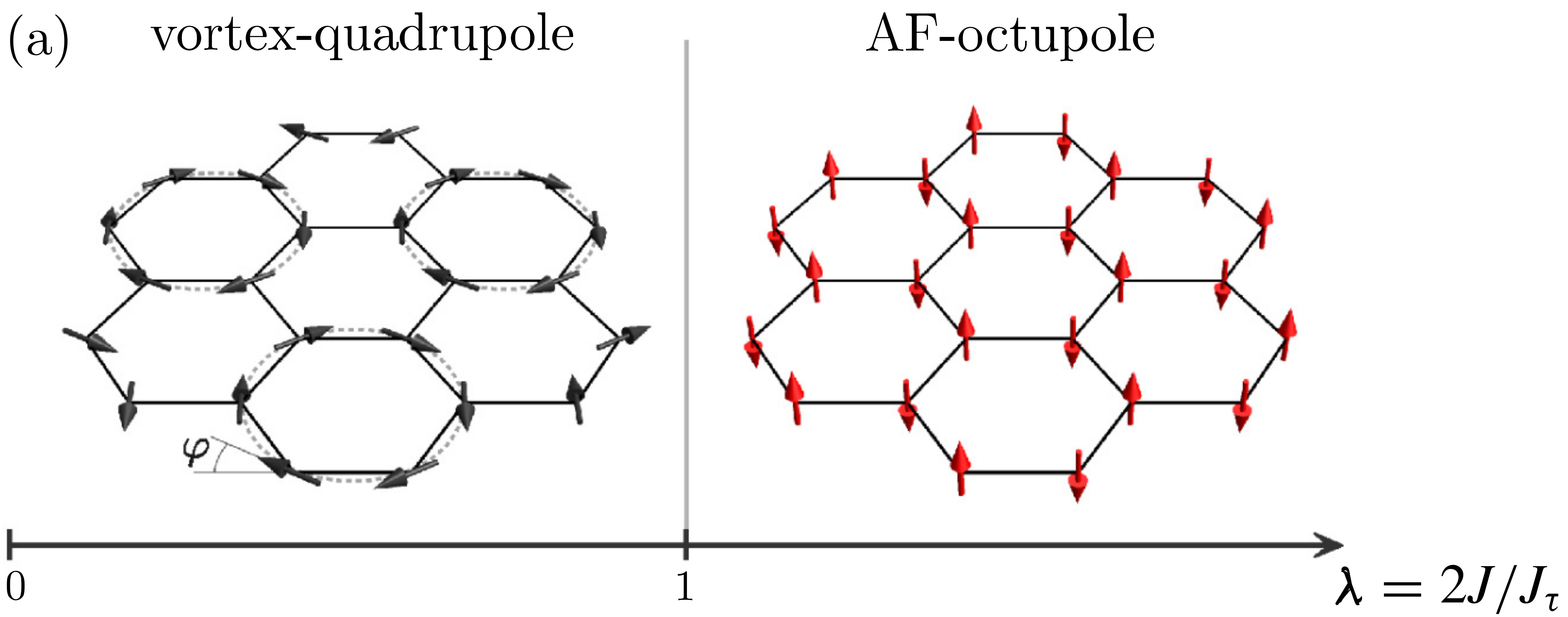


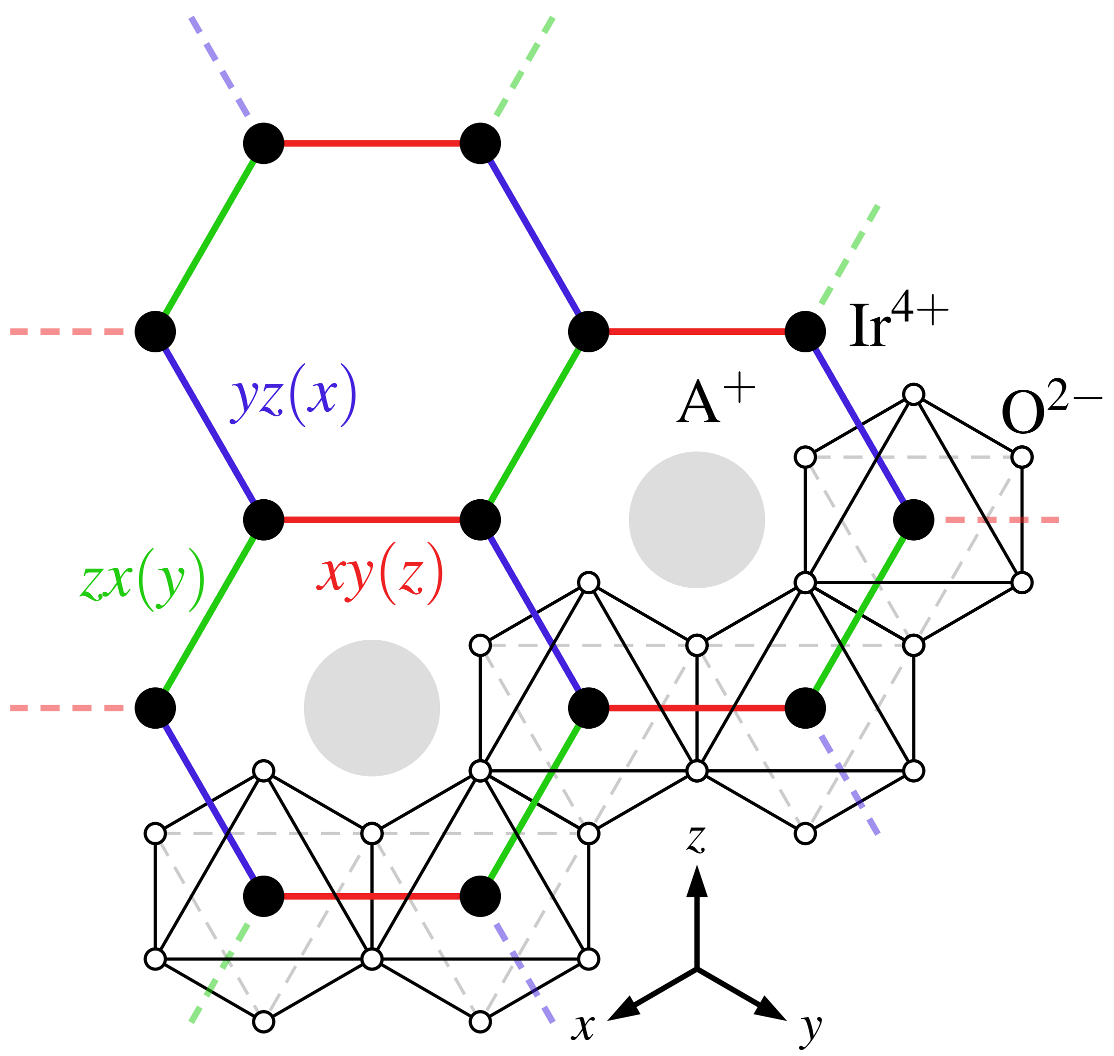
: Reminds the generic model in honeycomb lattice

G. Khaliullin, D. Churchill, P. Stavropoulos, HYK, PRR 3, 033163 (2021)

Generic Spin Model in 2D honeycomb $J-K-\Gamma$ model

nearest neighbour:
ideal honeycomb

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



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

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

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

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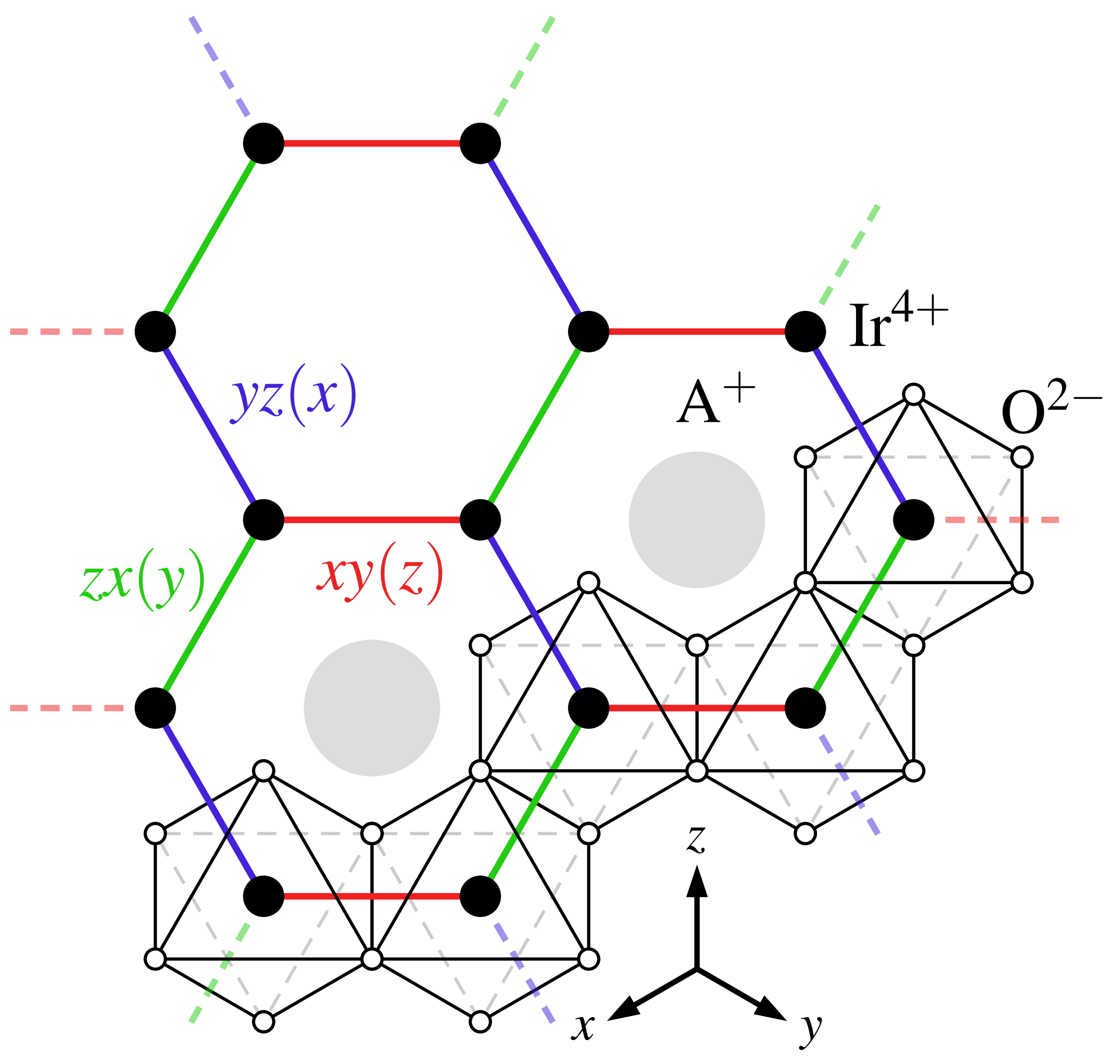

bond-dep. interaction

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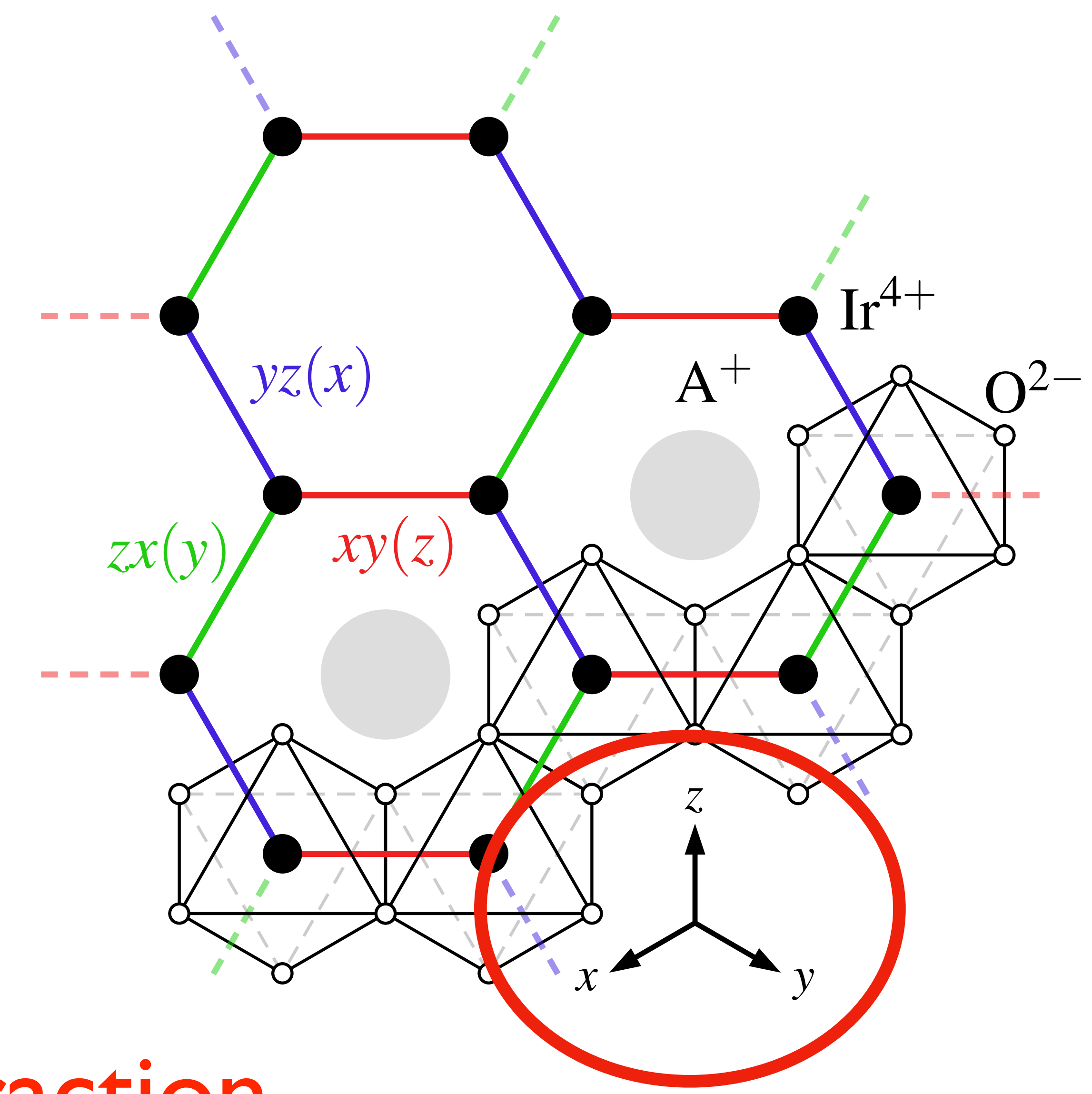

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J. Rau, E. Lee, HYK, PRL (2014)

(xyz)-coordinate

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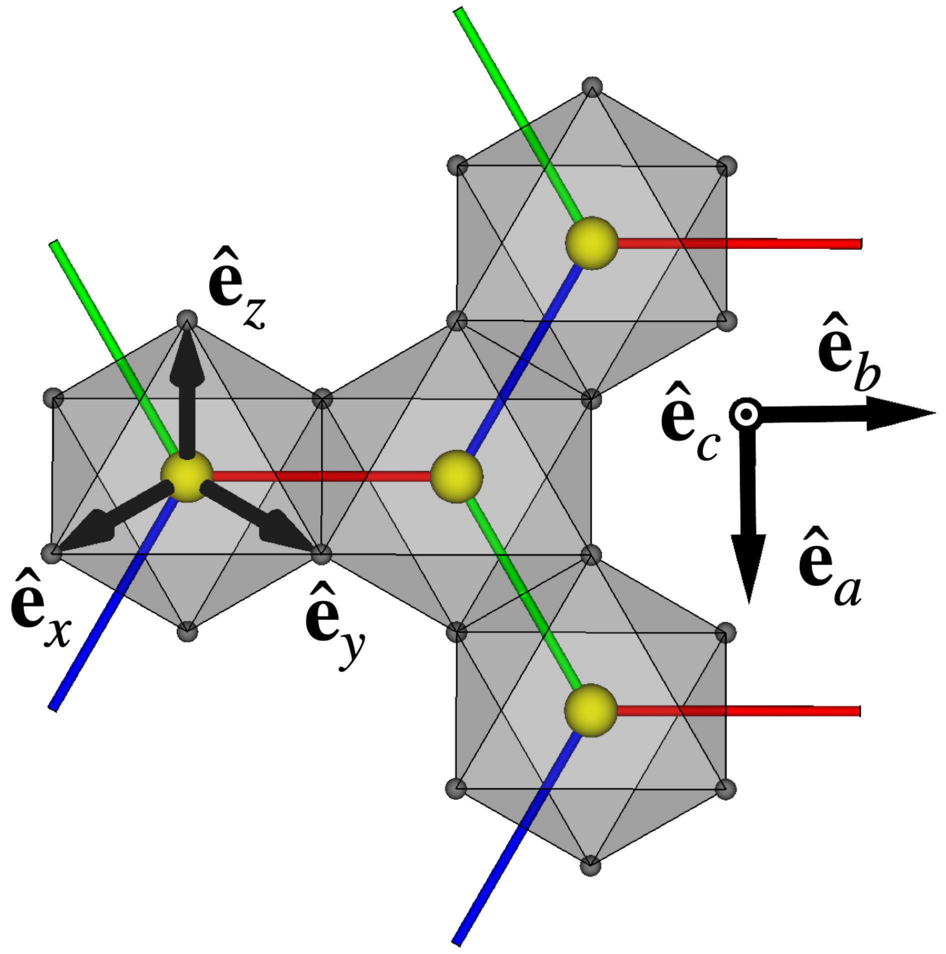
Compass model



$$\begin{pmatrix} s^x \\ s^y \\ s^z \end{pmatrix} = \begin{pmatrix} \frac{1}{\sqrt{6}} & -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{3}} \\ \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{3}} \\ -\frac{2}{\sqrt{6}} & 0 & \frac{1}{\sqrt{3}} \end{pmatrix} \begin{pmatrix} s^a \\ s^b \\ s^c \end{pmatrix}$$

(xyz)-coordinate

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

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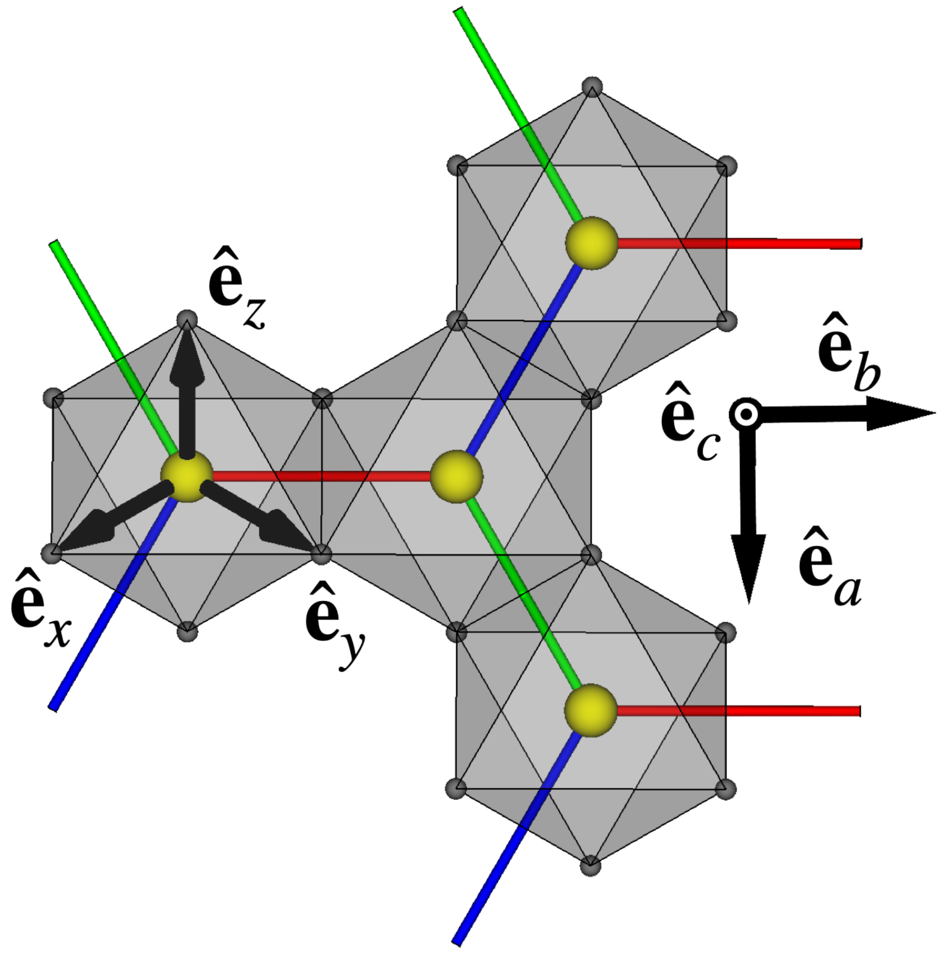

(abc)-coordinate

$$H_{ij}^{\gamma} = J_{xy}(S_i^x S_j^x + S_i^y S_j^y) + J_z S_i^z S_j^z$$
$$+A \left[\cos(\phi_\gamma)(S_i^x S_j^x - S_i^y S_j^y) - \sin(\phi_\gamma)(S_i^x S_j^y + S_i^y S_j^x)\right]$$
$$-B\sqrt{2} \left[\cos(\phi_\gamma)(S_i^x S_j^z + S_i^z S_j^x) + \sin(\phi_\gamma)(S_i^y S_j^z + S_i^z S_j^y)\right]$$

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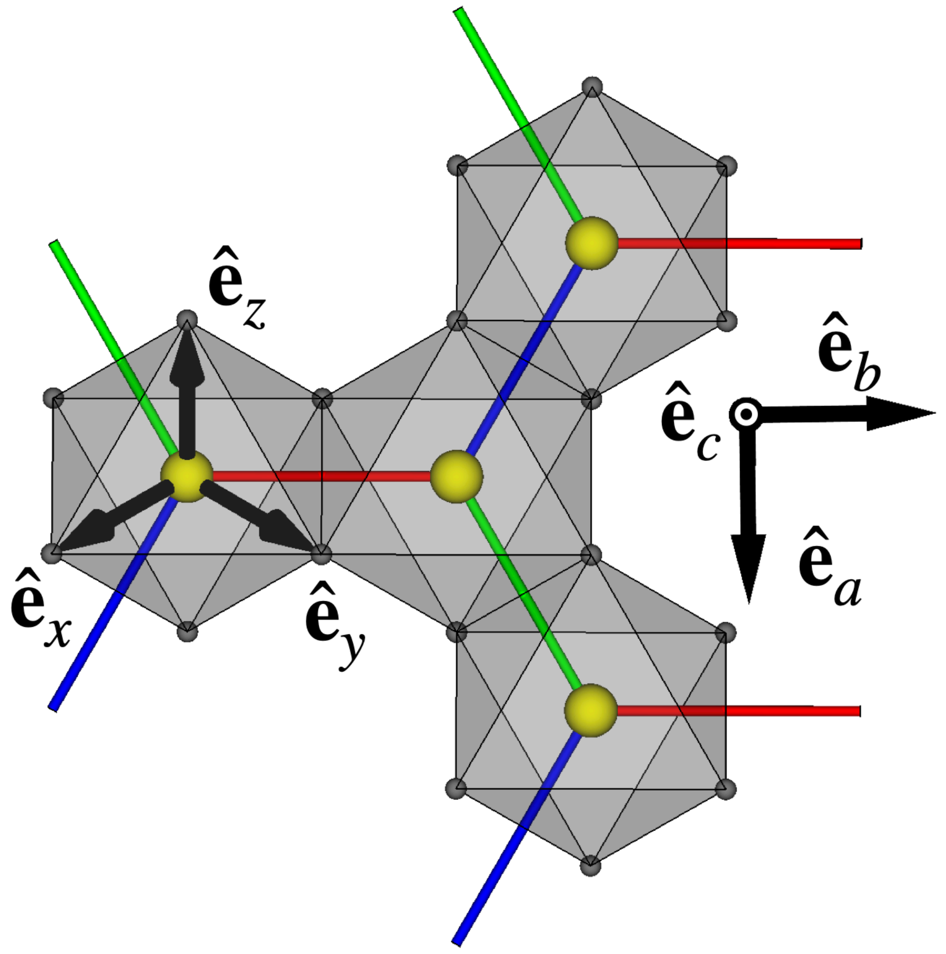

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$$J_{XY} = \tilde{J} + B \qquad J_Z = \tilde{J} + A$$

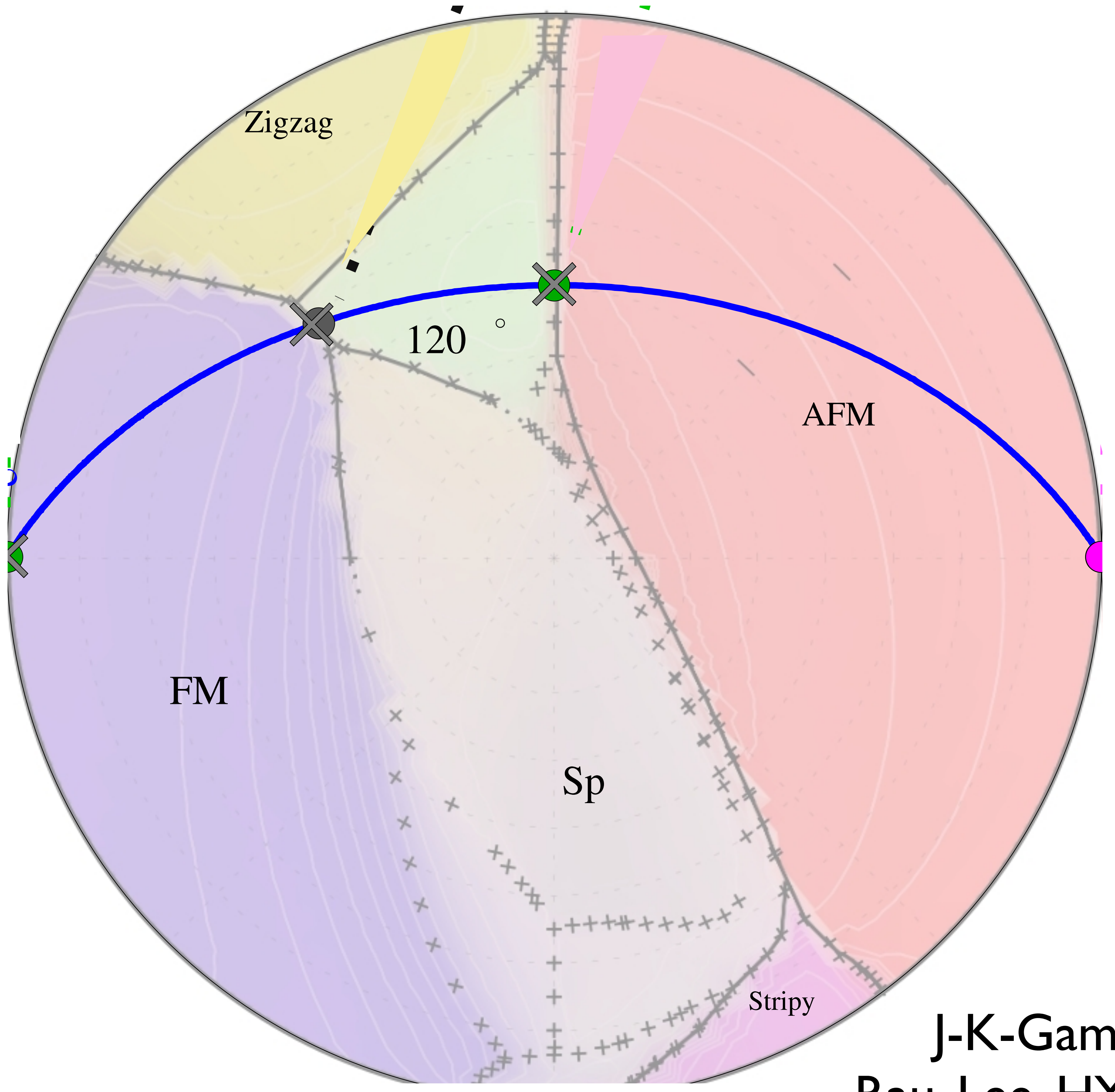
$$A = \frac{1}{3}(K + 2\Gamma) \qquad B = \frac{1}{3}(K - \Gamma)$$

In this model: Kitaev limit $J_{xy} = J_z = A = B$

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

Our model is a special line of JKGamma model

$$H_{ij}^{\gamma} = \left(\frac{J_\tau}{2} - J\right)\left(s_i^z s_j^z + s_i^x s_j^x\right) + J s_i^y s_j^y$$
$$+\frac{J_\tau}{2}\left(\cos\phi_\gamma\left(s_i^z s_j^z - s_i^x s_j^x\right) - \sin\phi_\gamma\left(s_i^z s_j^x + s_i^x s_j^z\right)\right)$$



J-K-Gamma model;
Rau, Lee, HYK, PRL (2014)

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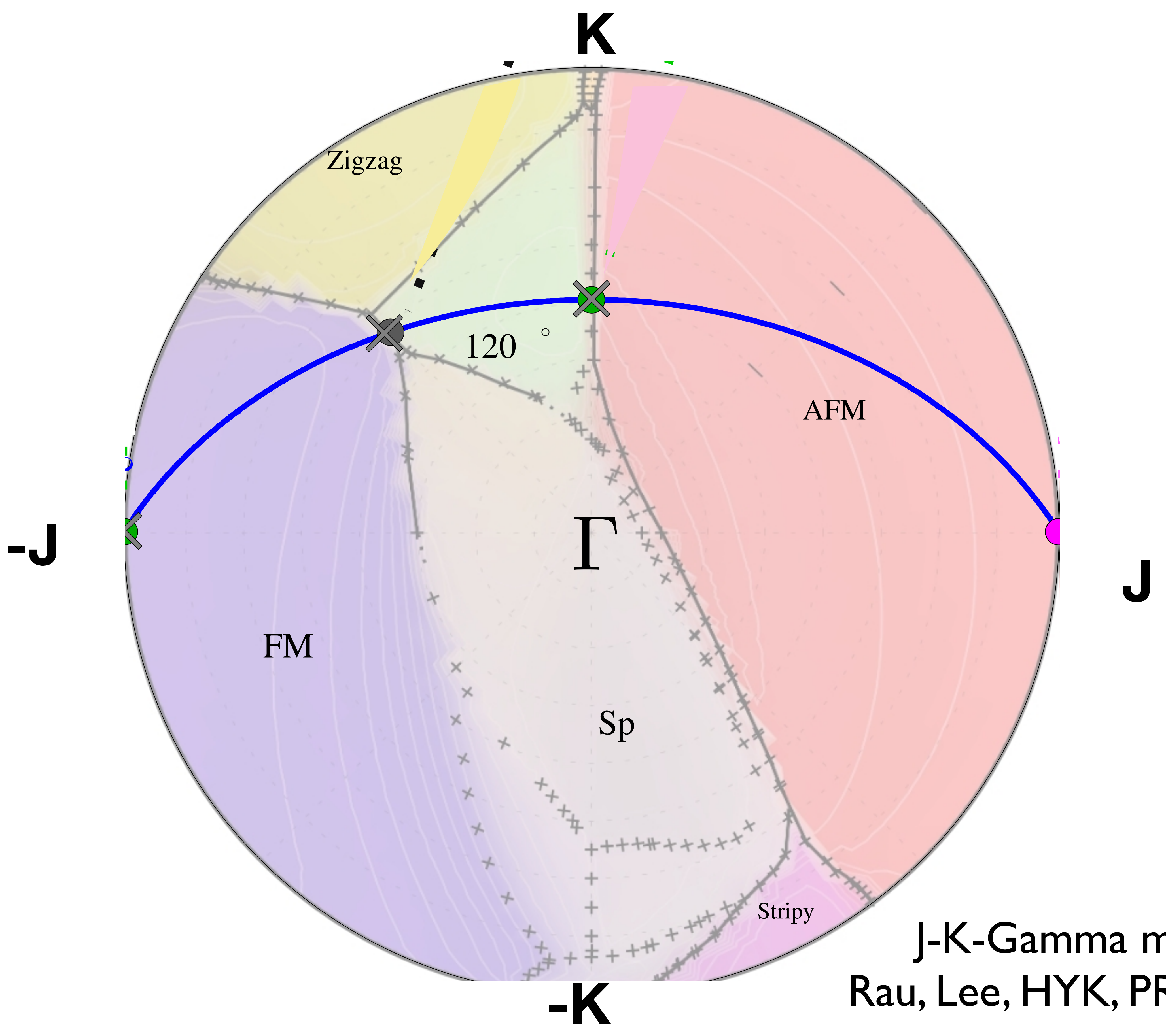

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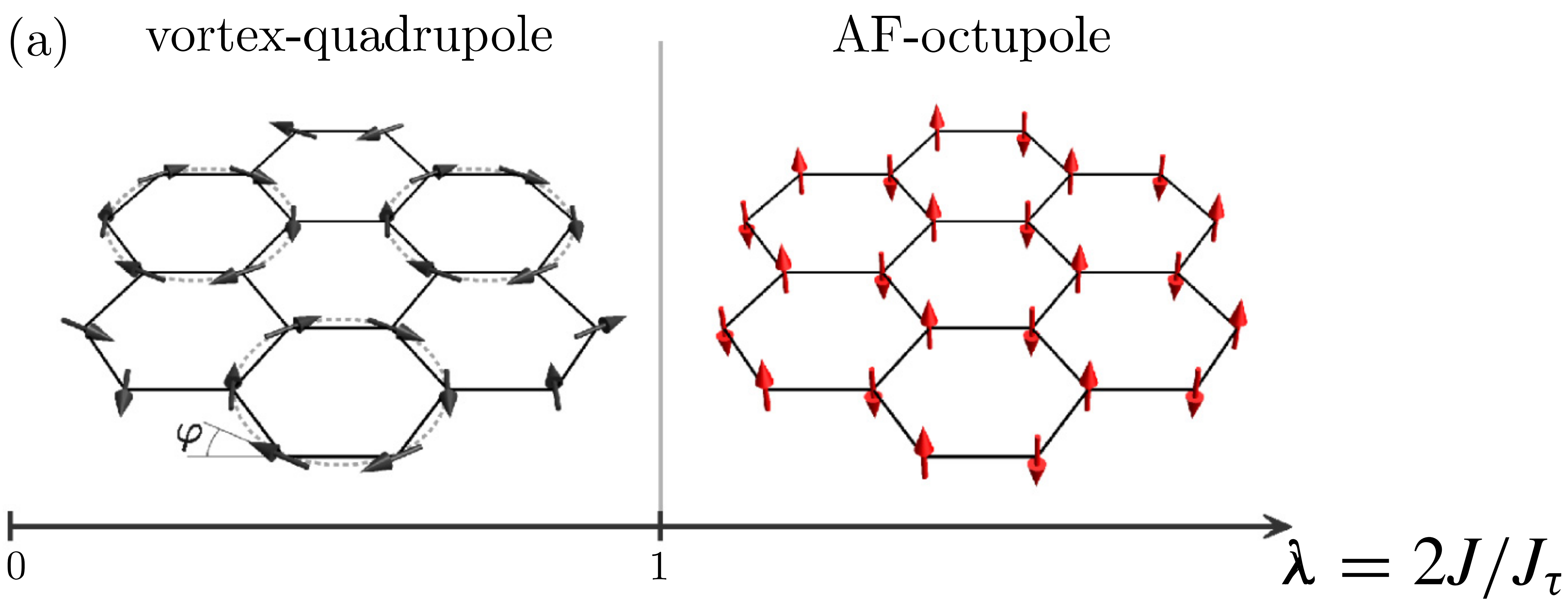

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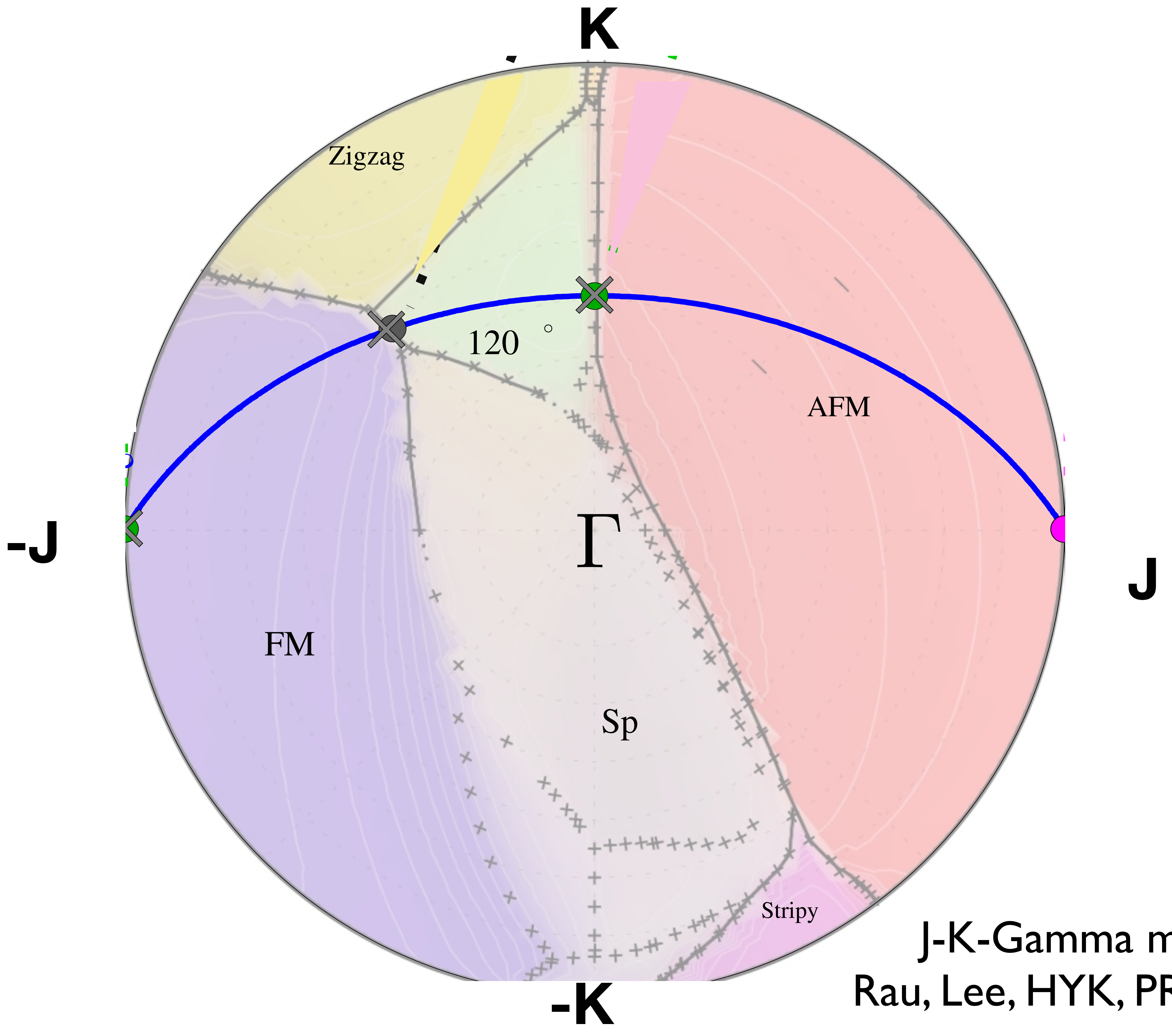

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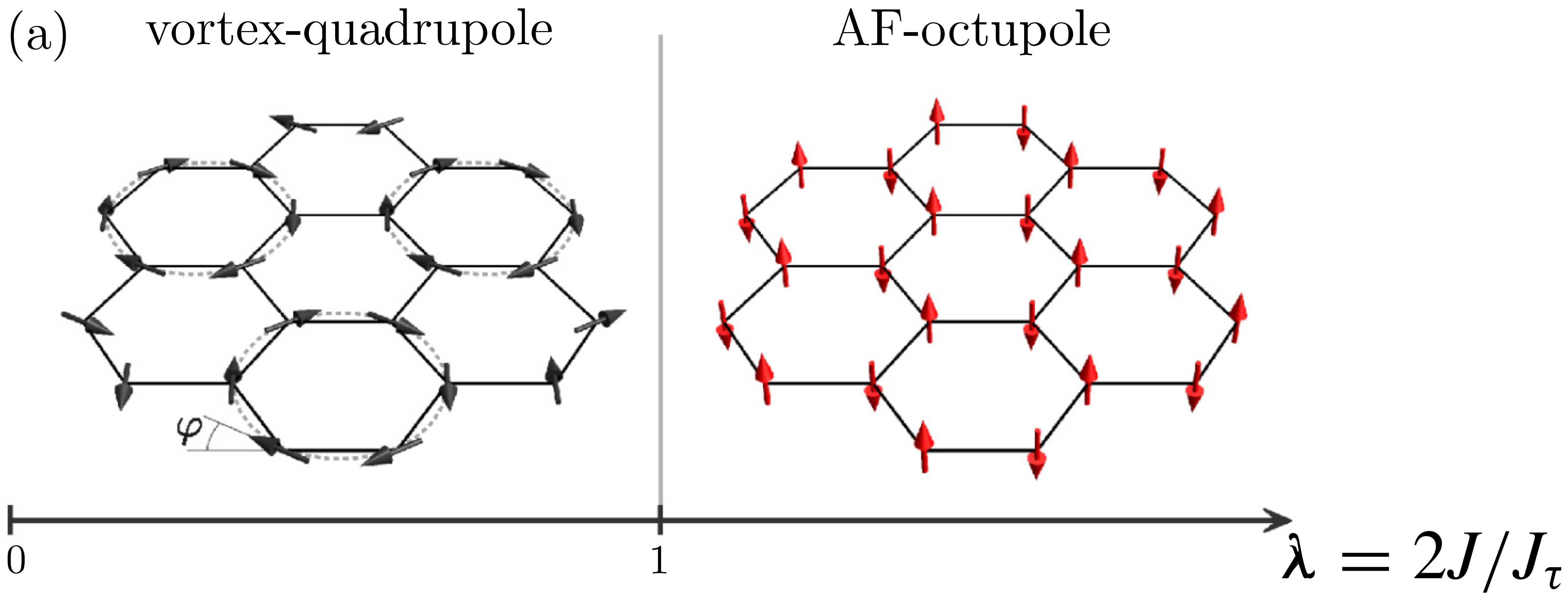

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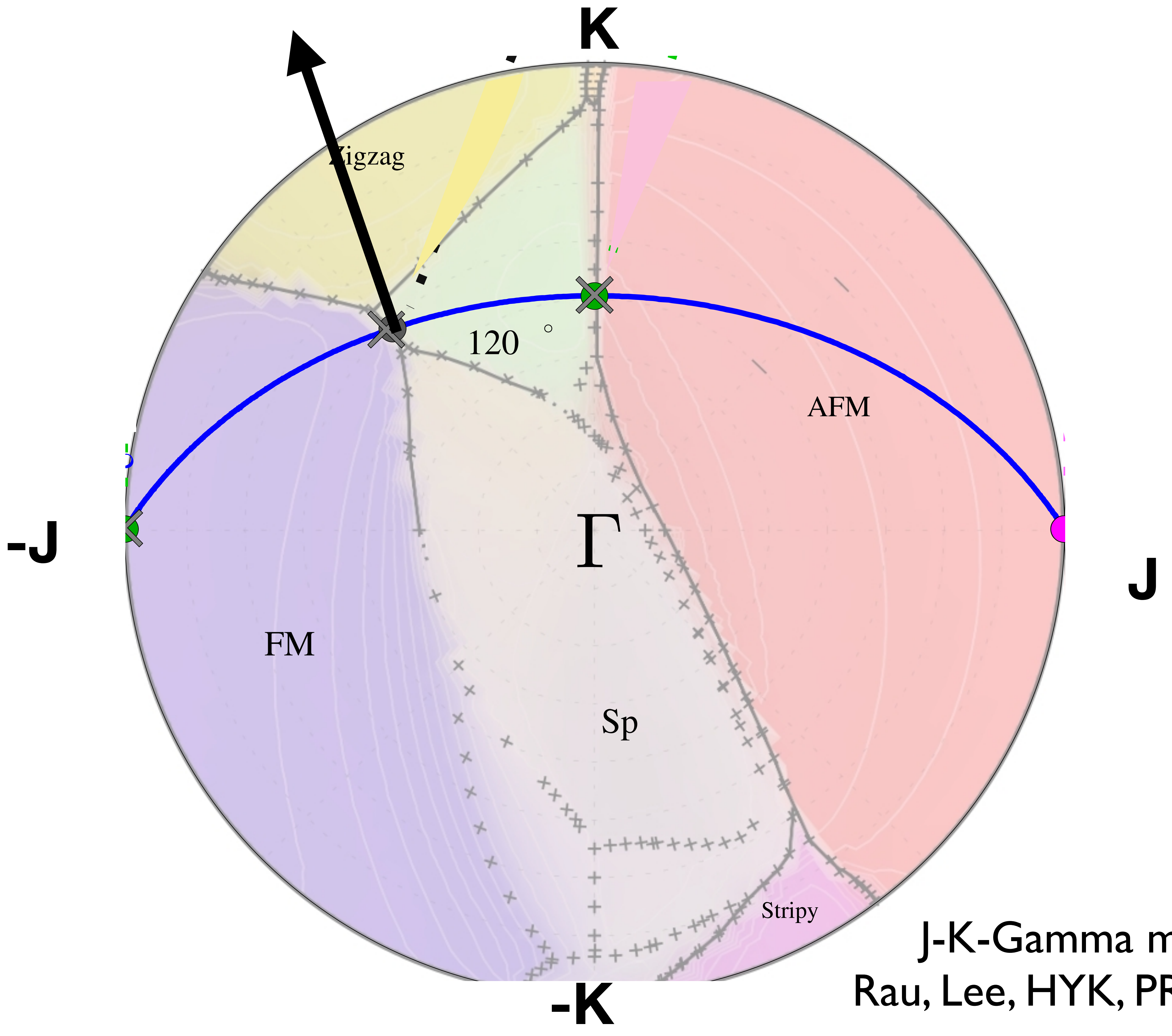

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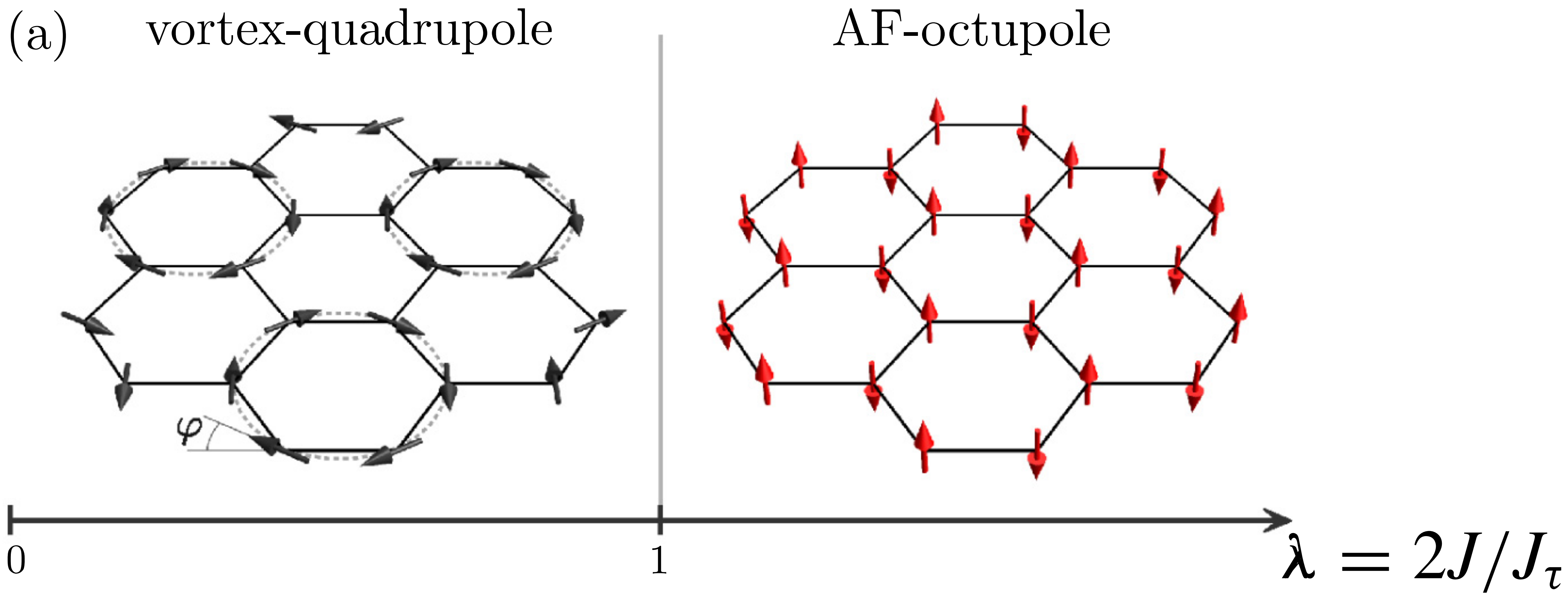

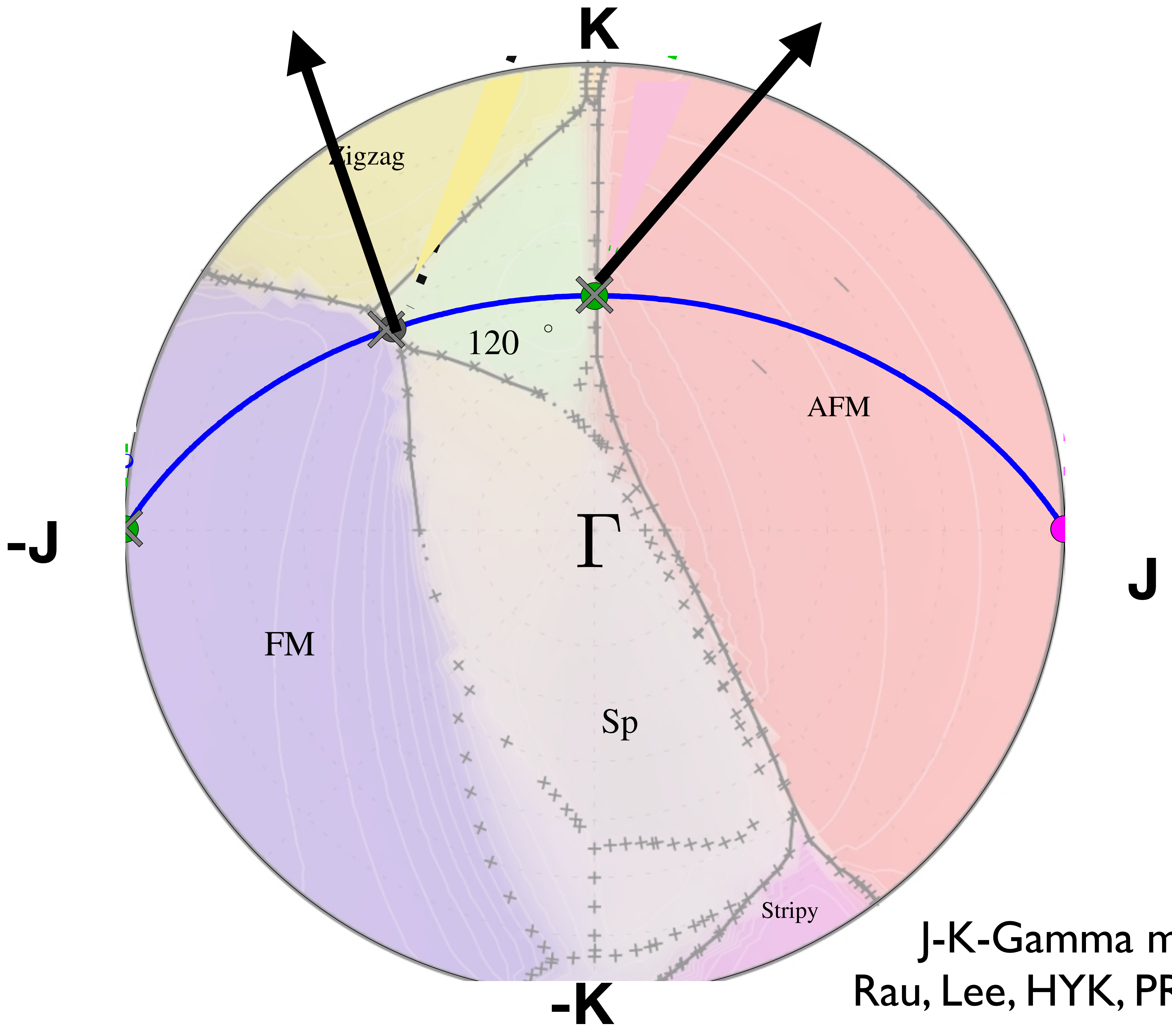

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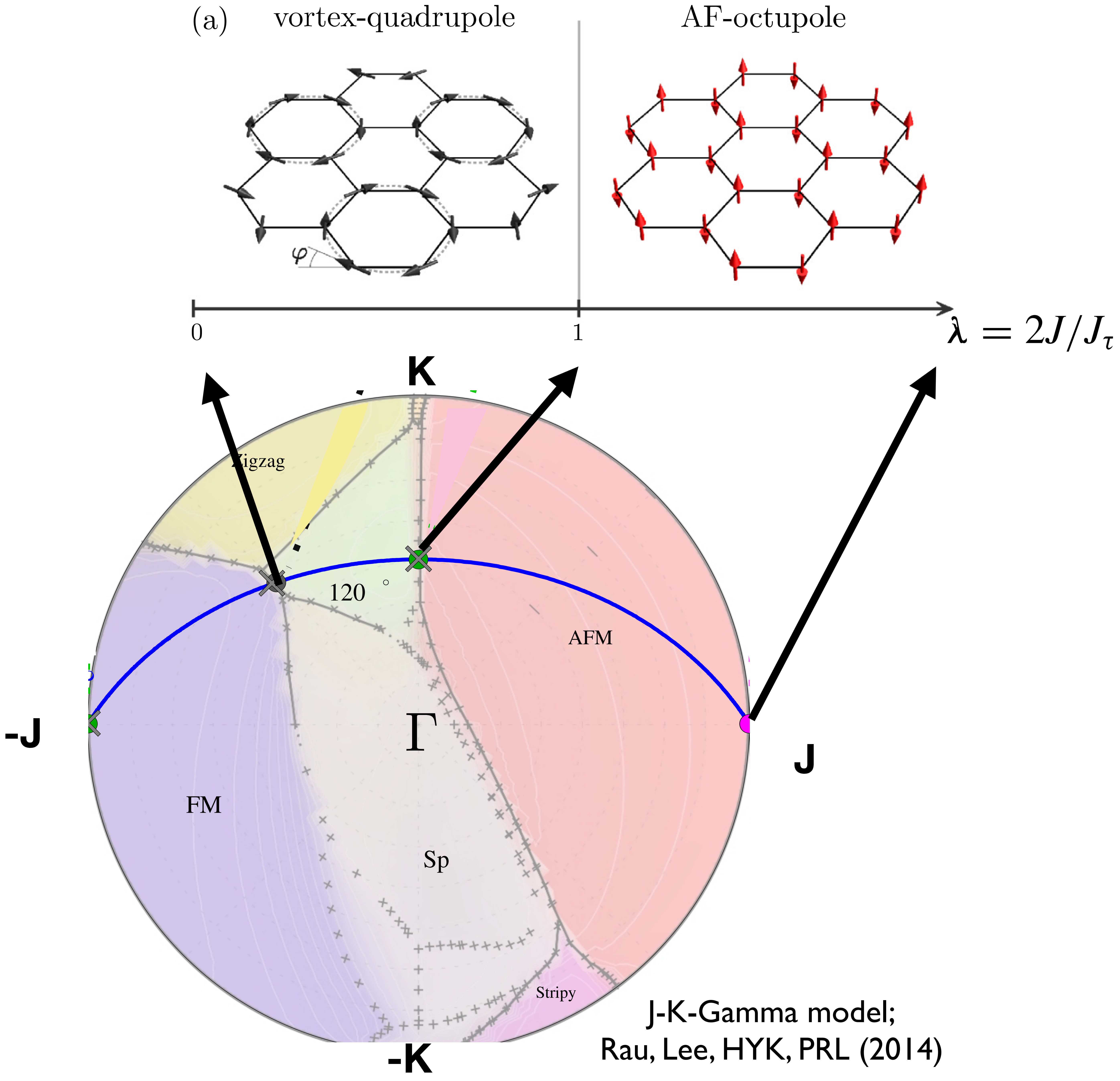
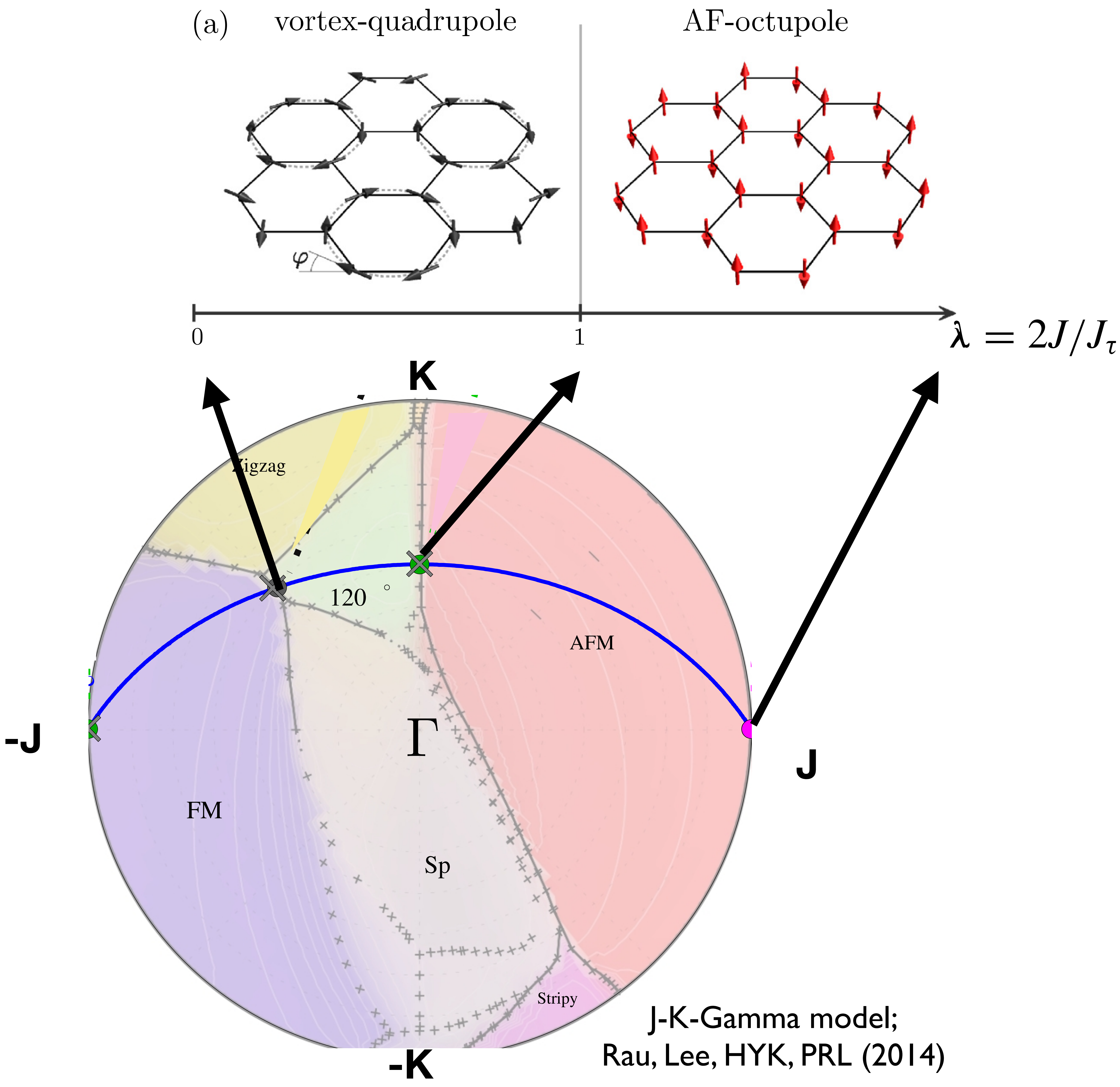


J-K-Gamma model;
Rau, Lee, HYK, PRL (2014)



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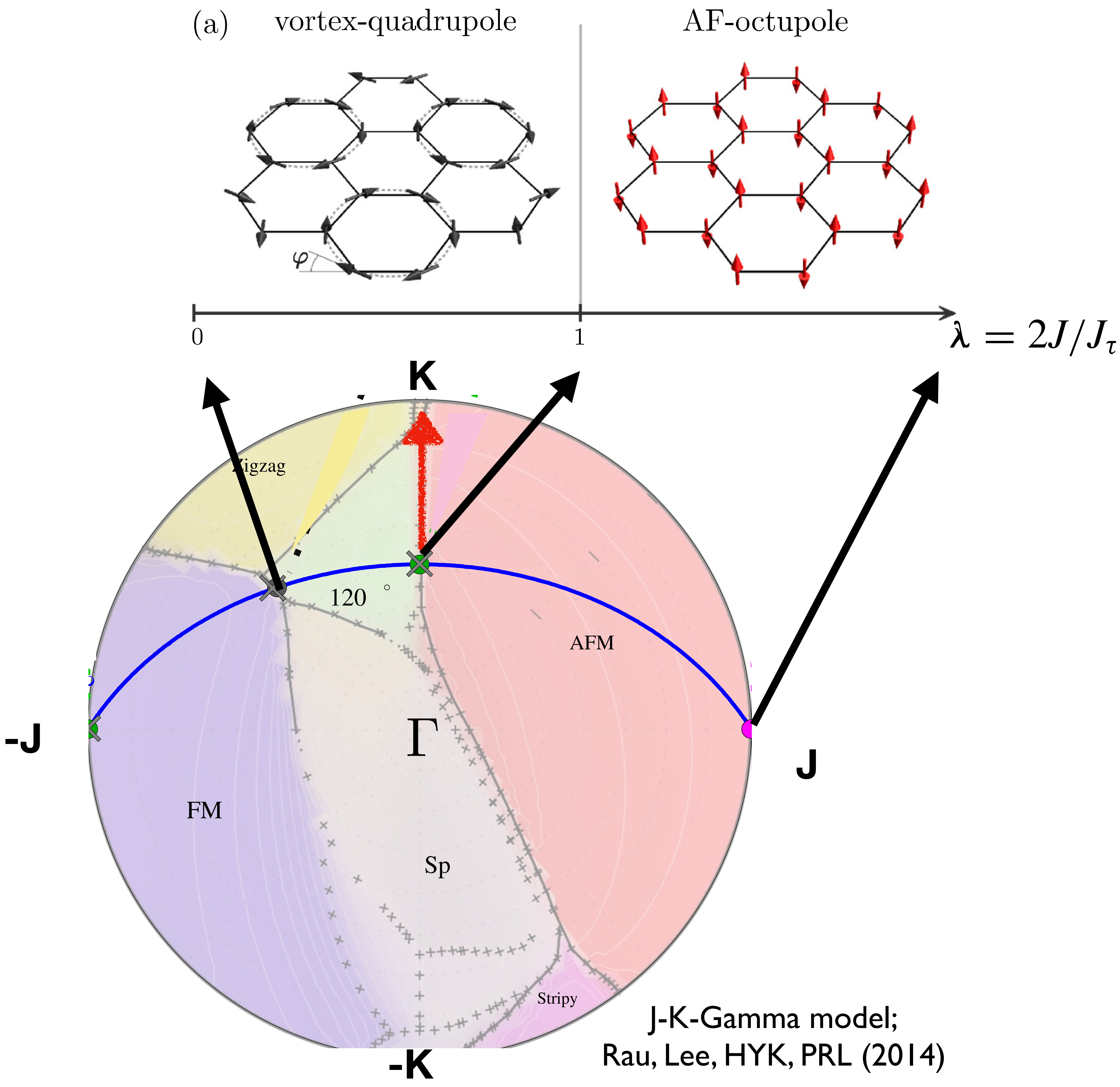
Can we tune to the Kitaev limit?
issues?



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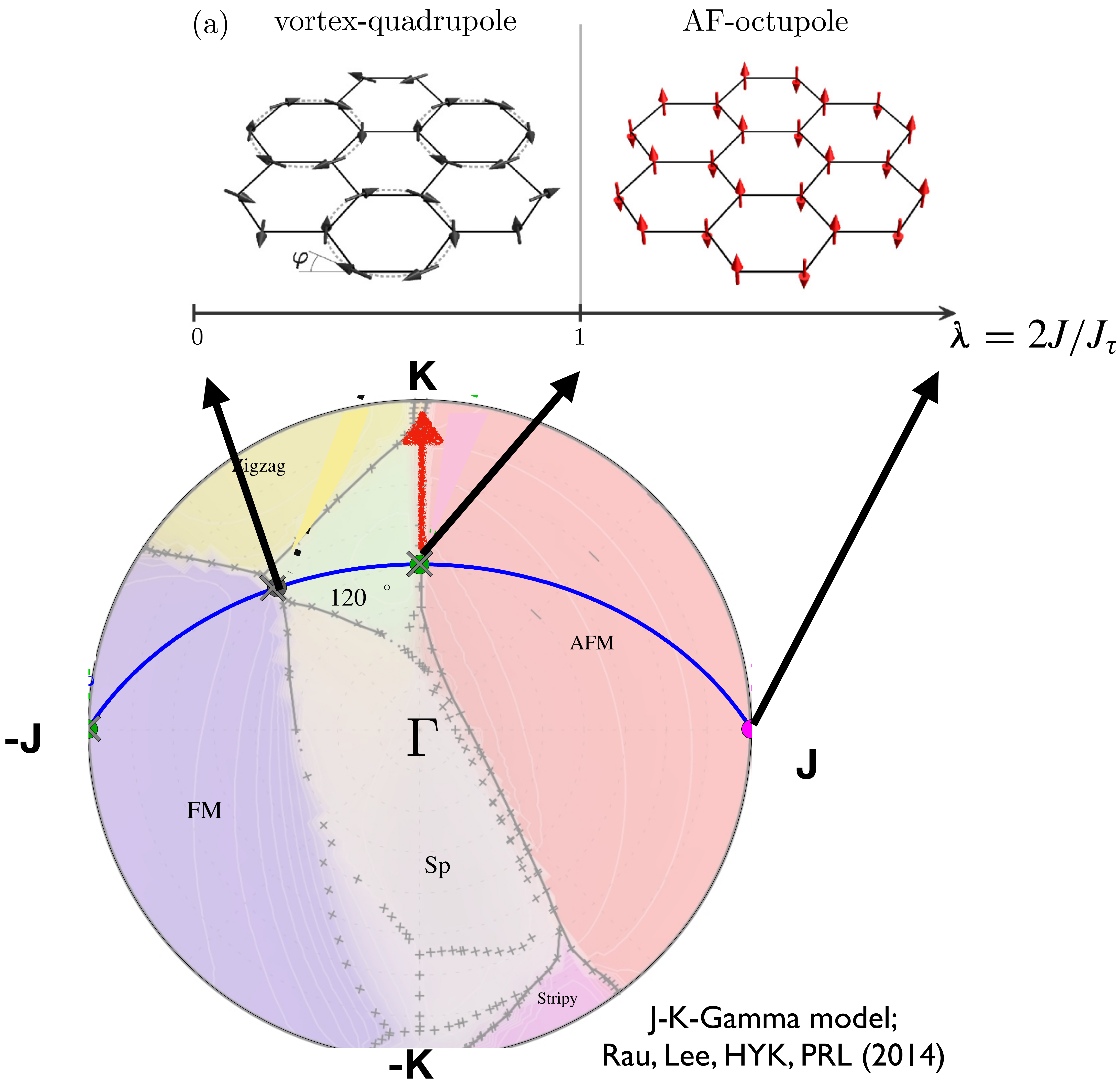

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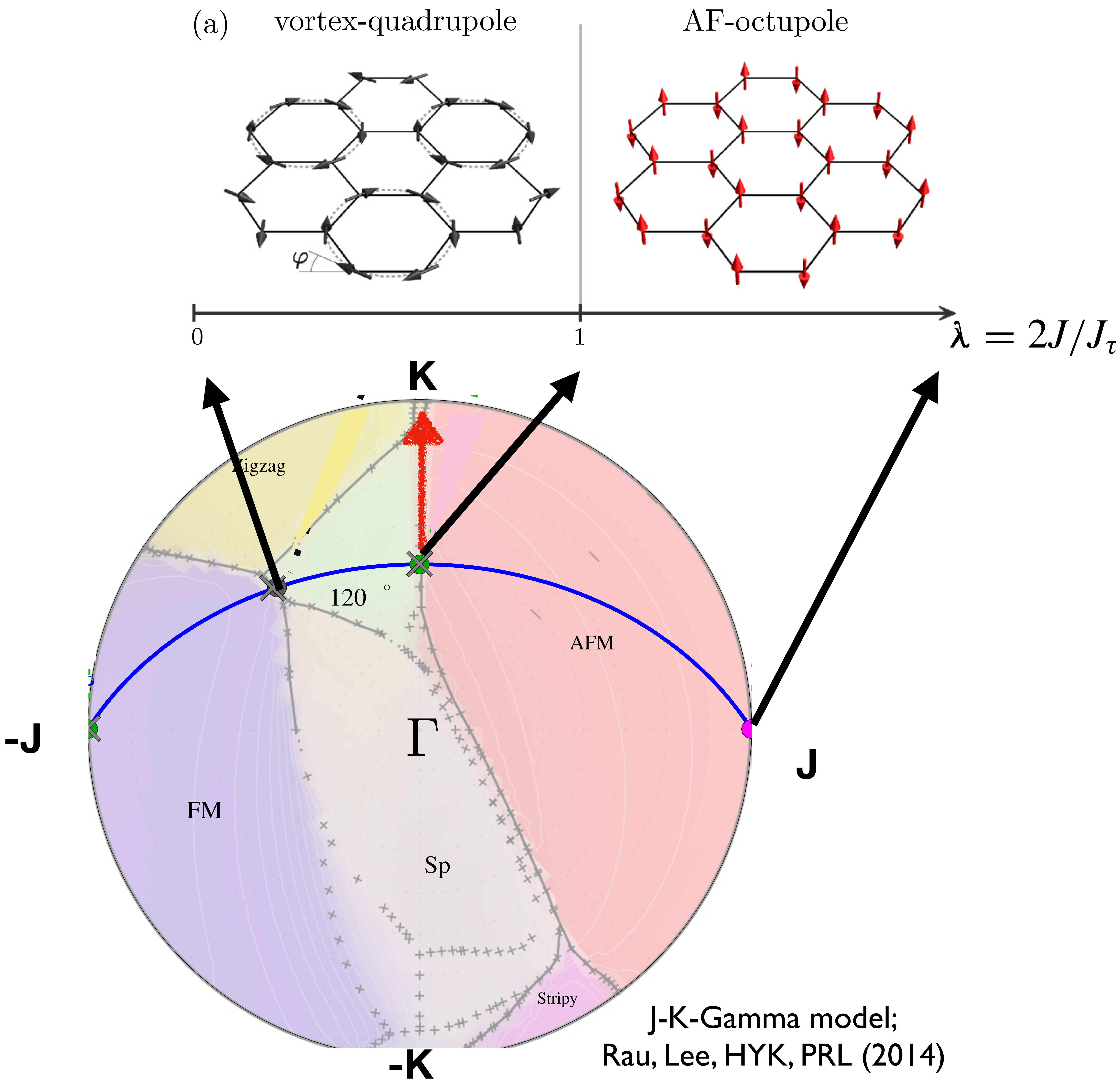
Can we tune to the Kitaev limit?
issues?

- **Kitaev limit** $J_{xy} = J_z = A$; cannot be satisfied



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$$H_{ij}^{\gamma} = \underbrace{\left(\frac{J_\tau}{2} - J\right)}_{J_{xy}} \left(s_i^z s_j^z + s_i^x s_j^x\right) + J s_i^y s_j^y + \frac{J_\tau}{2}\left(\cos\phi_\gamma\left(s_i^z s_j^z - s_i^x s_j^x\right) - \sin\phi_\gamma\left(s_i^z s_j^x + s_i^x s_j^z\right)\right)$$

Can we tune to the Kitaev limit?
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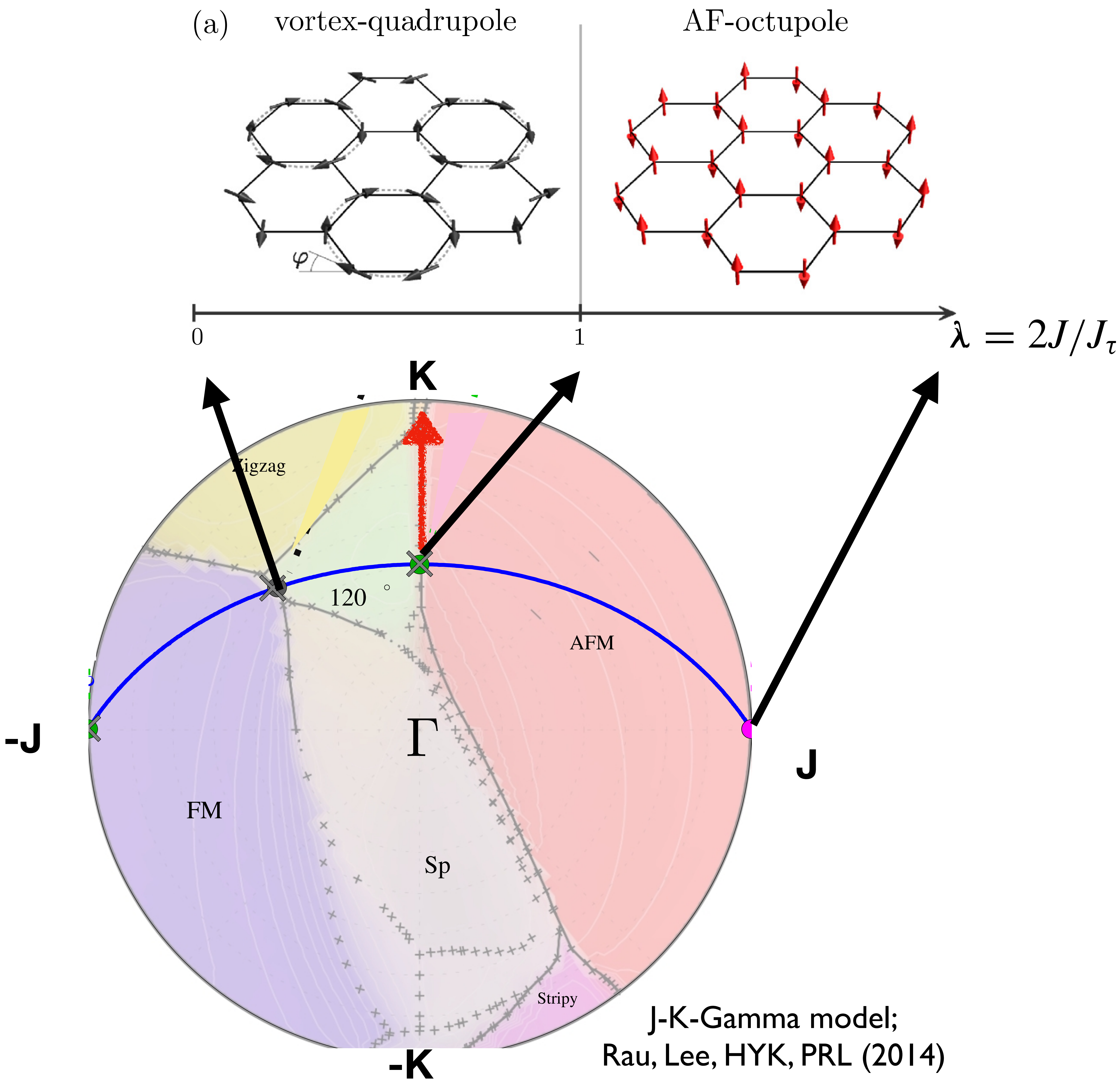

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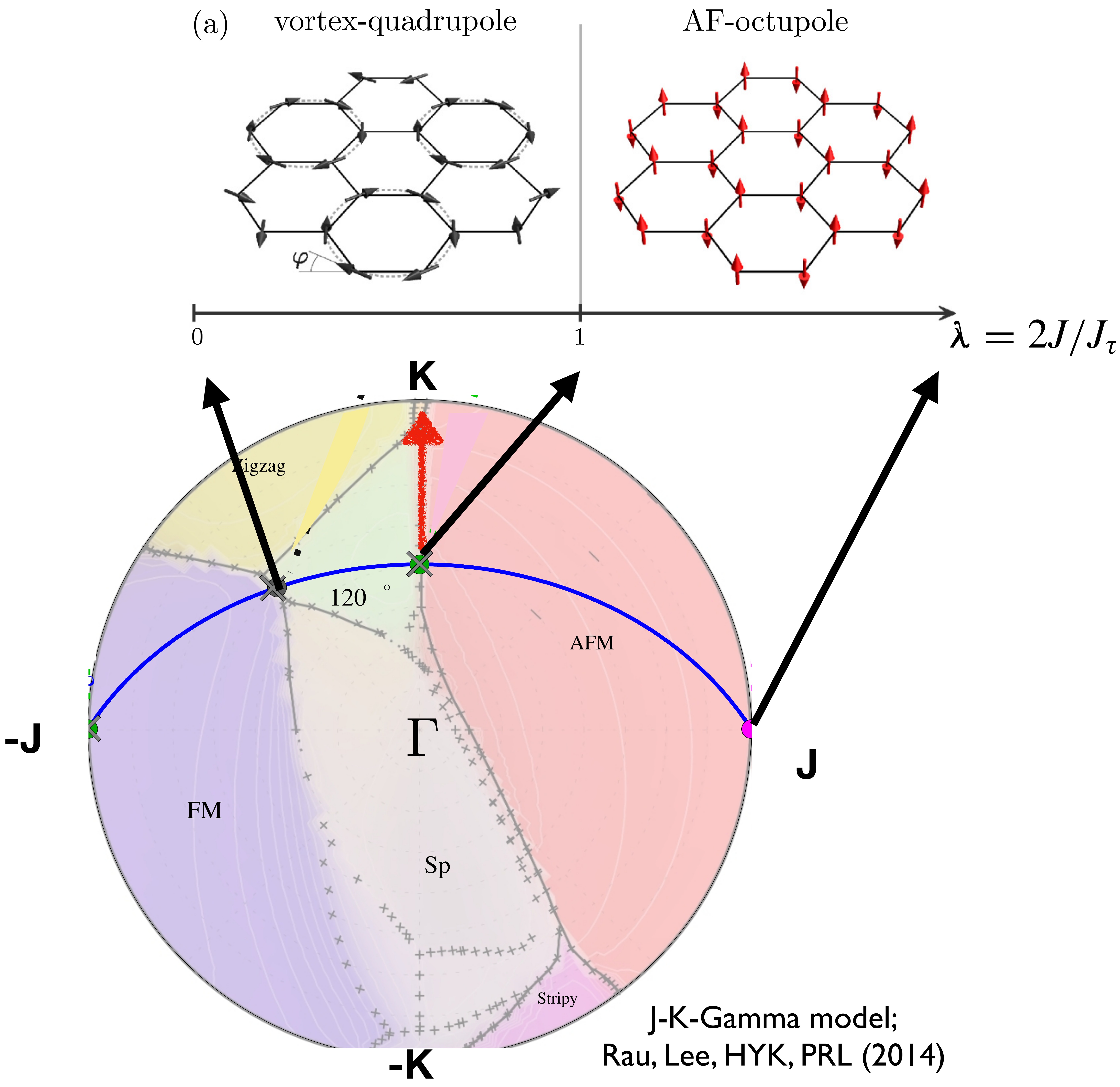

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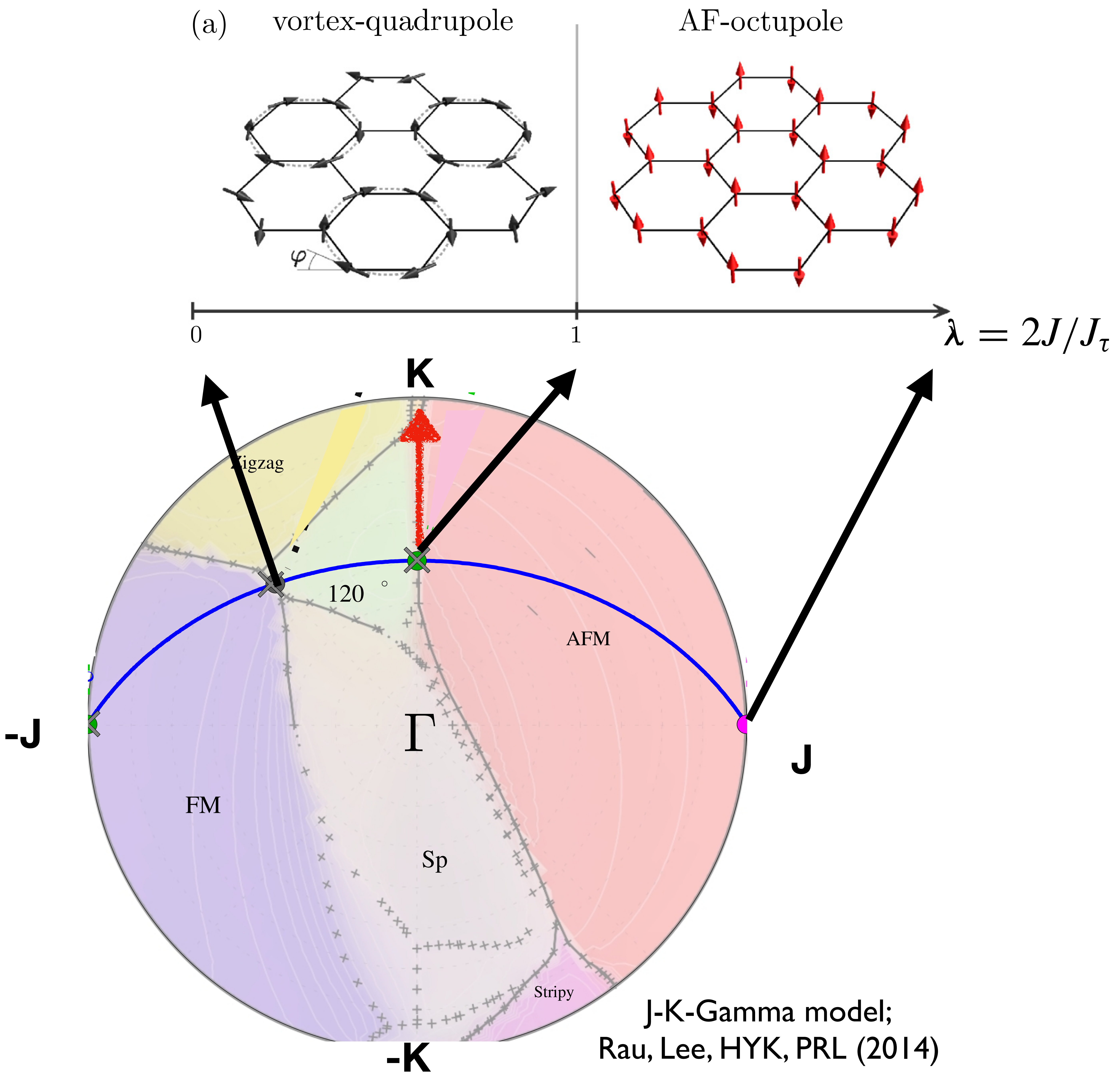

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3. another direct exchange

$$t_{ij} = \begin{matrix} & c_{j,xy} & c_{j,xz} & c_{j,yz} \\ \begin{matrix} c^\dagger_{i,xy} \\ c^\dagger_{i,xz} \\ c^\dagger_{i,yz} \end{matrix} & \multicolumn{3}{c}{\begin{pmatrix} t_3 & 0 & 0 \\ 0 & t_1 & t_2 \\ 0 & t_2 & t_1 \end{pmatrix}} \end{matrix}$$

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but ratio between J_q and J_o is no longer limited

D. Churchill, HYK, PRB 105, 014438 (2022)

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$$J_\tau = \frac{4(t_1 - t_3)^2}{9U} \qquad J_o = \frac{2[t_1(t_1 + 2t_3) + t_2^2]}{3U} \qquad J_q = \frac{2[t_1(t_1 + 2t_3) - t_2^2]}{3U} \qquad J_q \neq -J_o$$

D. Churchill, HYK, PRB 105, 014438 (2022)

Compare the two

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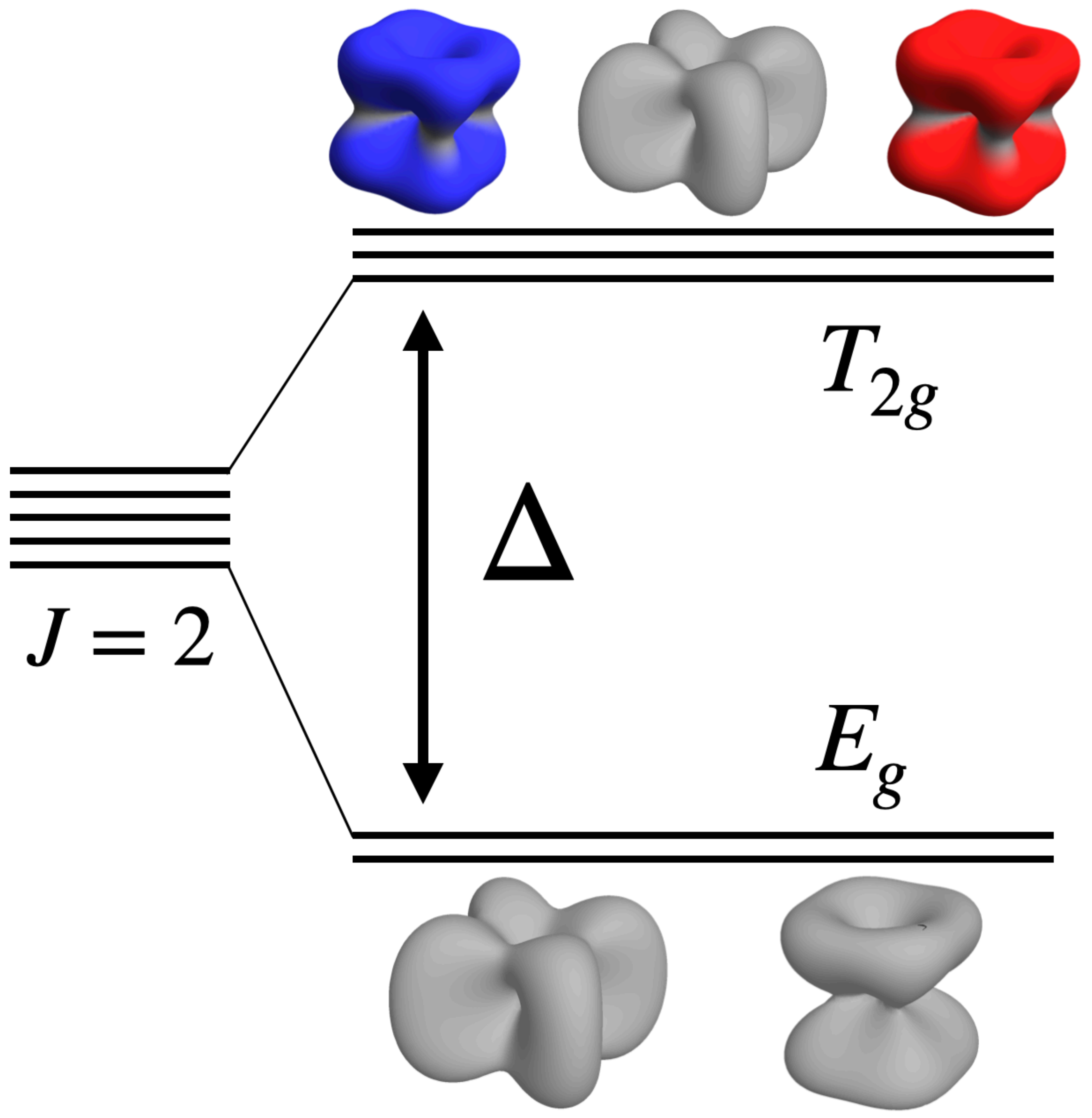
Kitaev model via magnetic field



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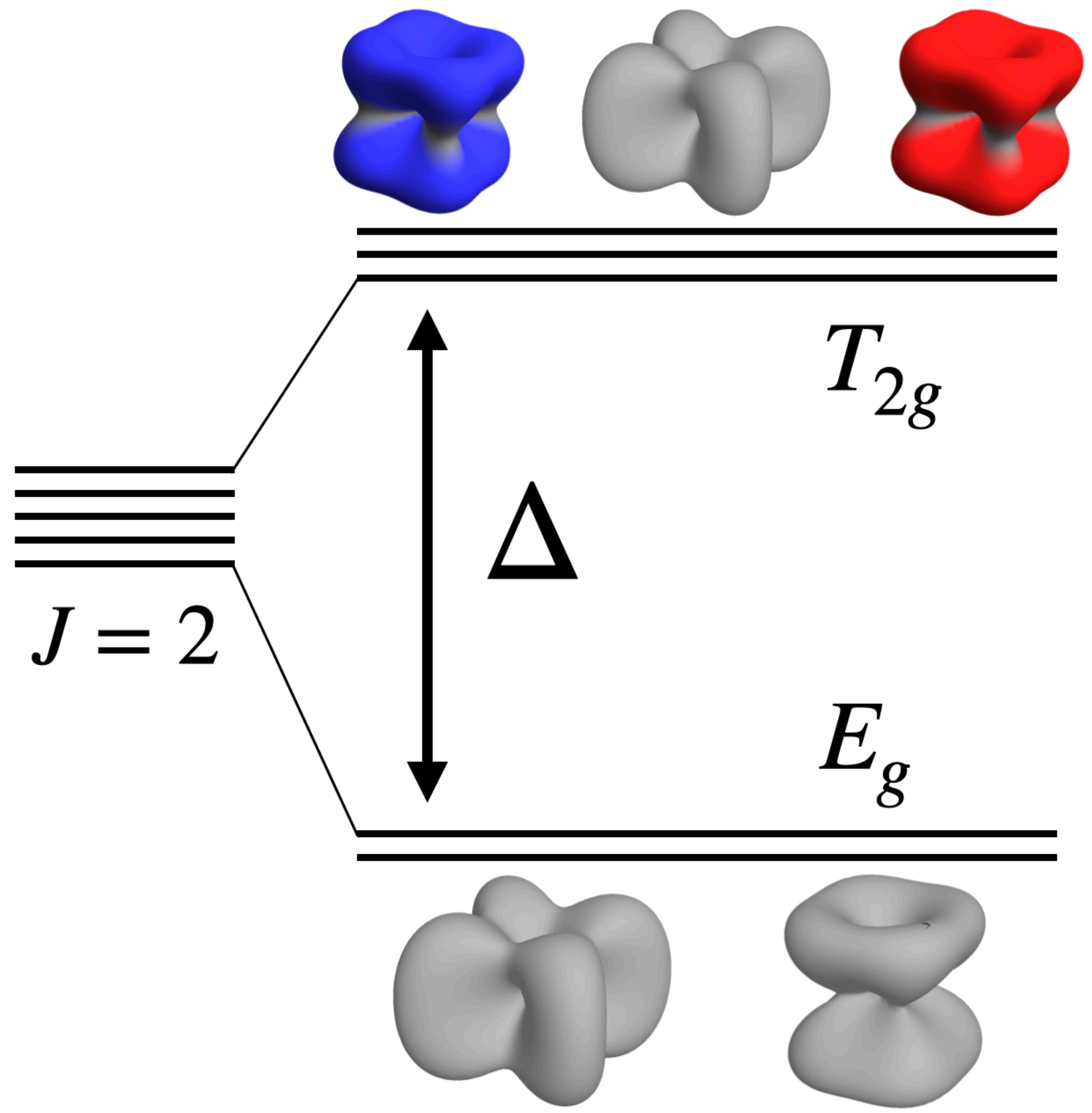

$$H^Z = g_J \mu_B \mathbf{J} \cdot \mathbf{h}$$

along [111]//c-axis

$$= g_J \mu_B \left(\begin{array}{cc|ccc} 0 & 0 & \frac{1}{\sqrt{2}} h^+ & \frac{1}{\sqrt{2}} h^- & 2h_z \\ 0 & 0 & \frac{\sqrt{3}}{2} h^- & \frac{\sqrt{3}}{2} h^+ & 0 \\ \hline \frac{1}{\sqrt{2}} h^- & \frac{\sqrt{3}}{2} h^+ & -h_z & 0 & -\frac{1}{\sqrt{2}} h^- \\ \frac{1}{\sqrt{2}} h^+ & \frac{\sqrt{3}}{2} h^- & 0 & h_z & \frac{1}{\sqrt{2}} h^+ \\ 2h_z & 0 & -\frac{1}{\sqrt{2}} h^+ & \frac{1}{\sqrt{2}} h^- & 0 \end{array} \right)$$

$$h^{\pm} = h_x + i h_y$$

Project to E_g doublet:

Field is perp to the plane // s_y (octuploar moment)

$$H_{ij}^{\gamma} = \left(\frac{J_\tau}{2} + J_q\right)(s_i^z s_j^z + s_i^x s_j^x) + J_o s_i^y s_j^y$$

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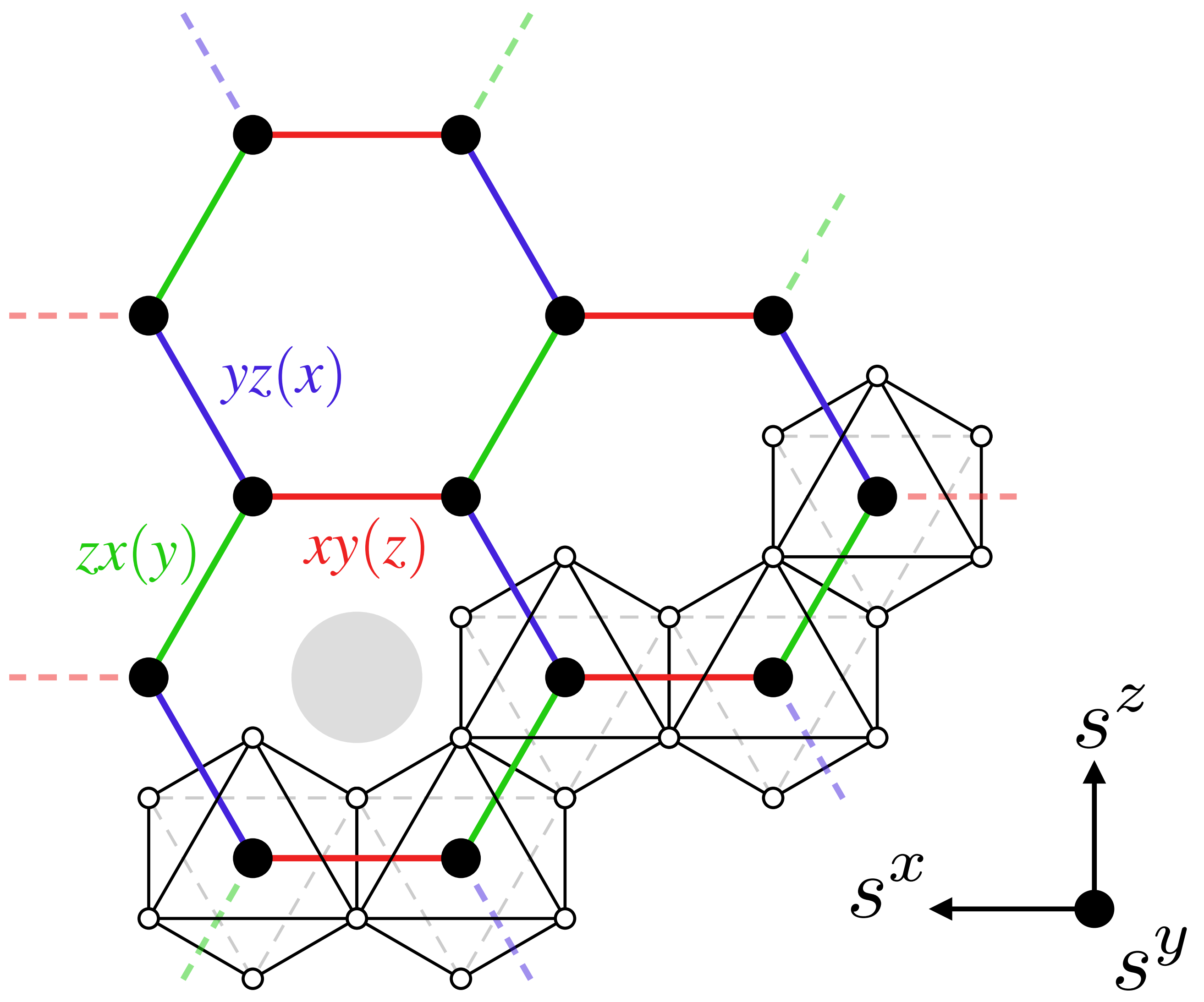

Bond-dependent quadrupole-octupole interaction

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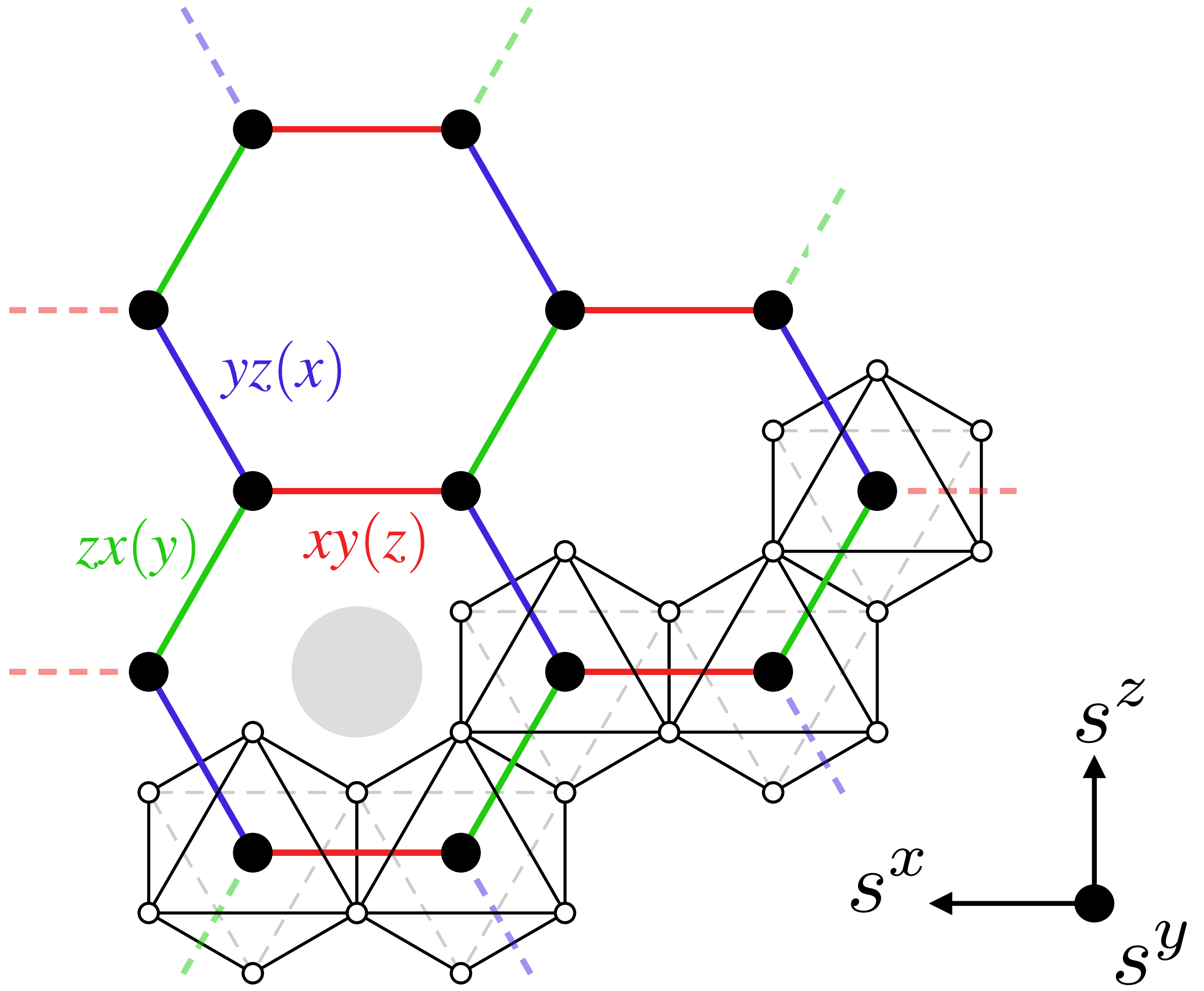
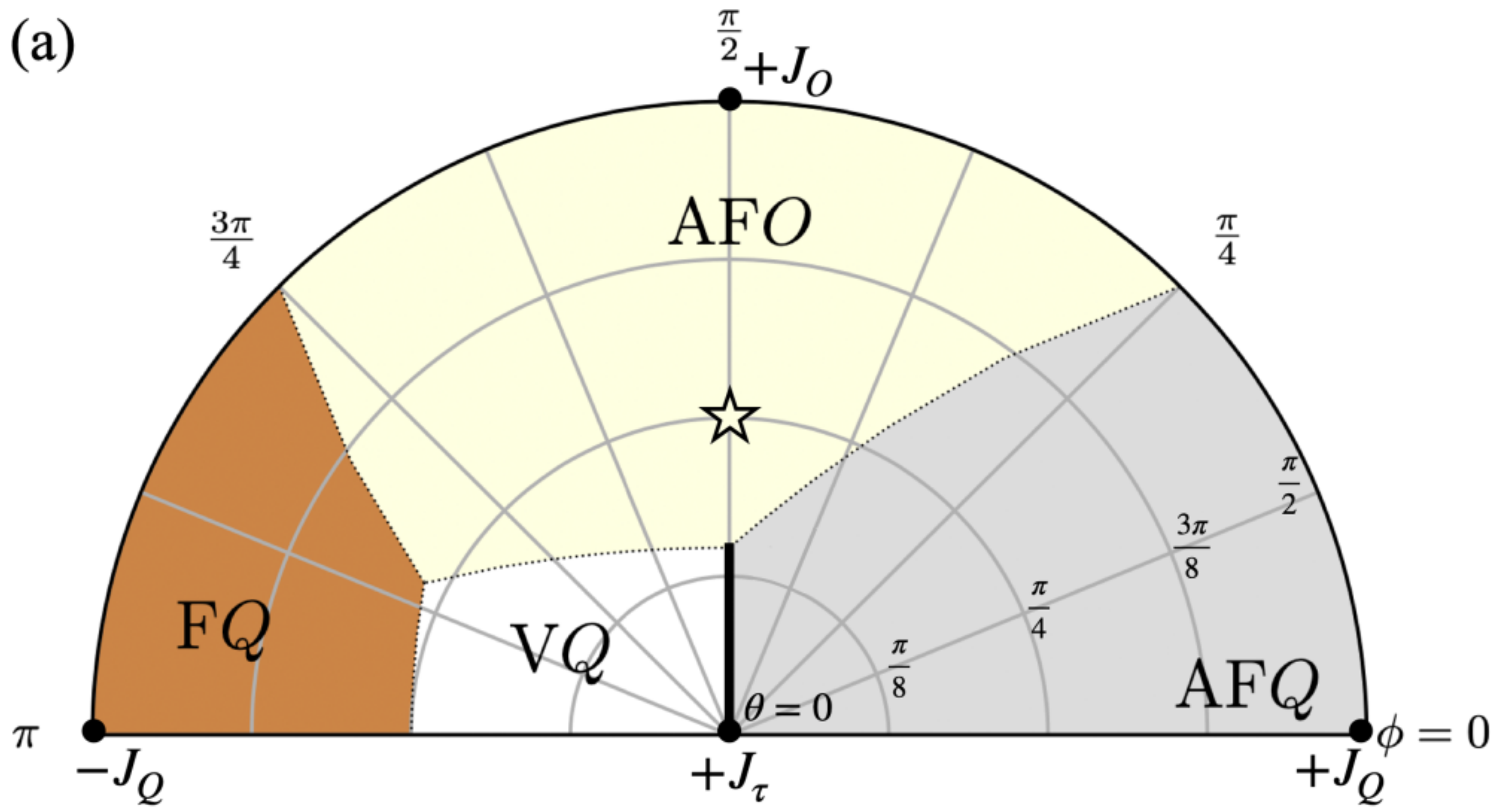


Bond-dependent quadrupole-octupole interaction

$$J_B \sim \frac{t_2\left(2t_1 + t_3\right)}{U}\frac{g_J\mu_B h}{\Delta} \qquad h_{eff}^{c} \sim \frac{t_2\left(t_1 - t_3\right)}{U}\frac{g_J\mu_B h}{\Delta} + \ldots$$

Classical phase diagram (Monte Carlo simulated annealing): $5d^2$ honeycomb lattice

Classical phase diagram (Monte Carlo simulated annealing): 5d² honeycomb lattice



$$J_B = 0$$

$$J_\tau = \cos\theta$$

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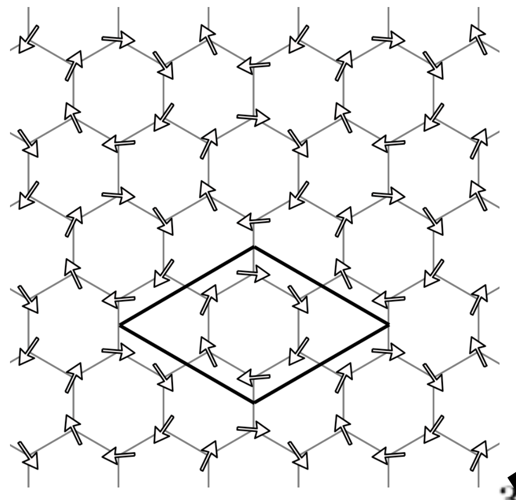
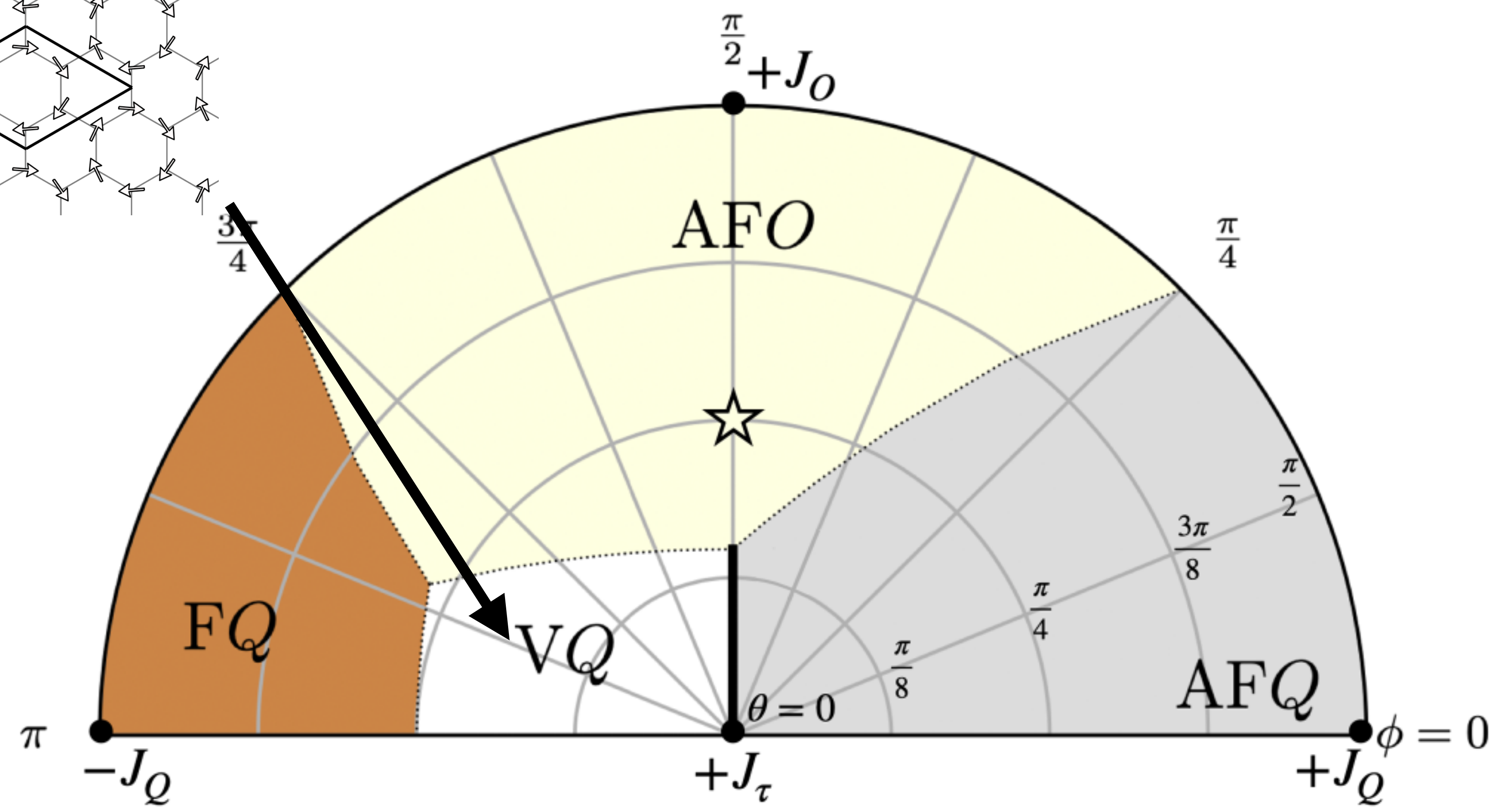
Classical phase diagram (Monte Carlo simulated annealing): 5d^2 honeycomb lattice





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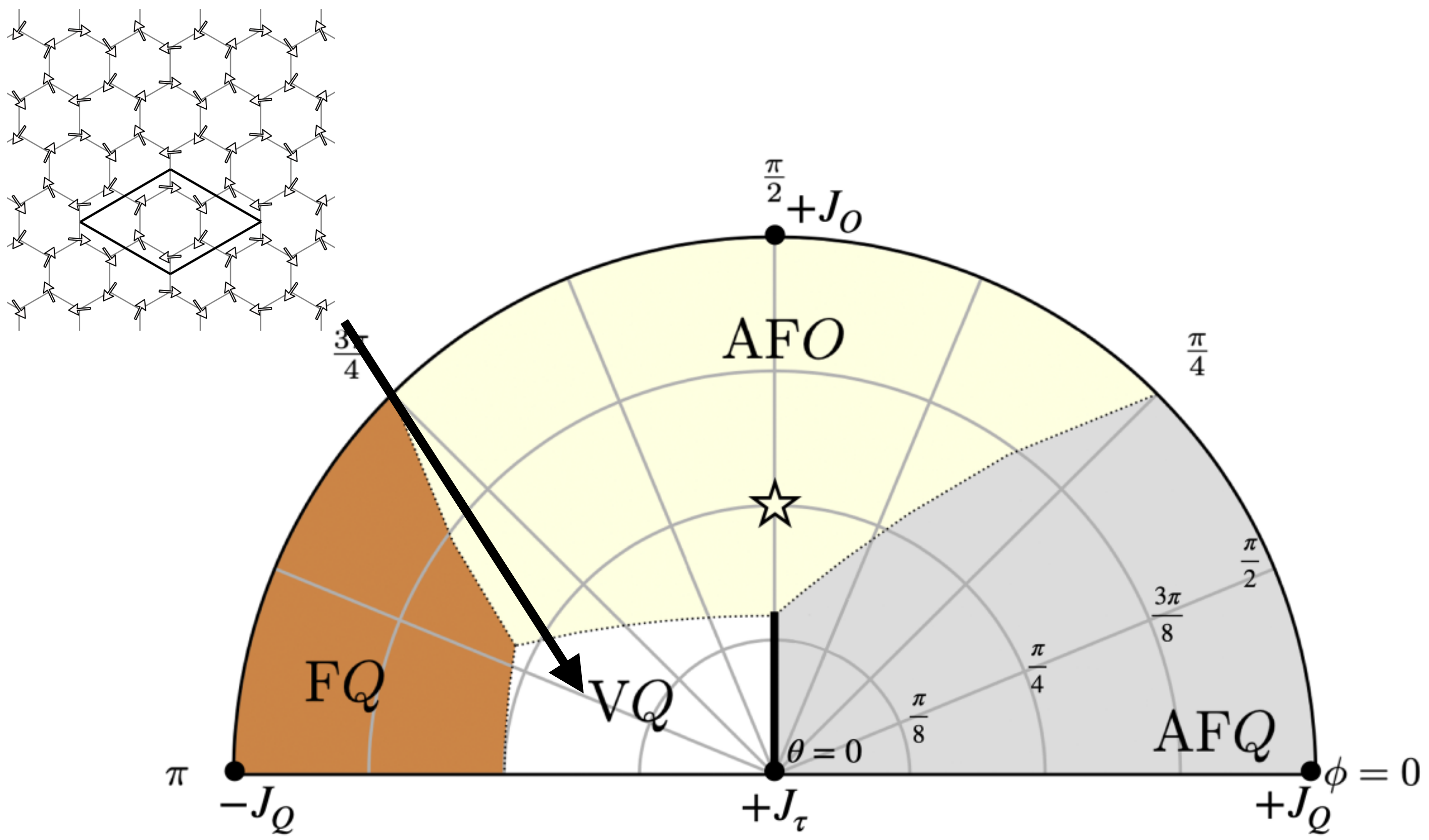

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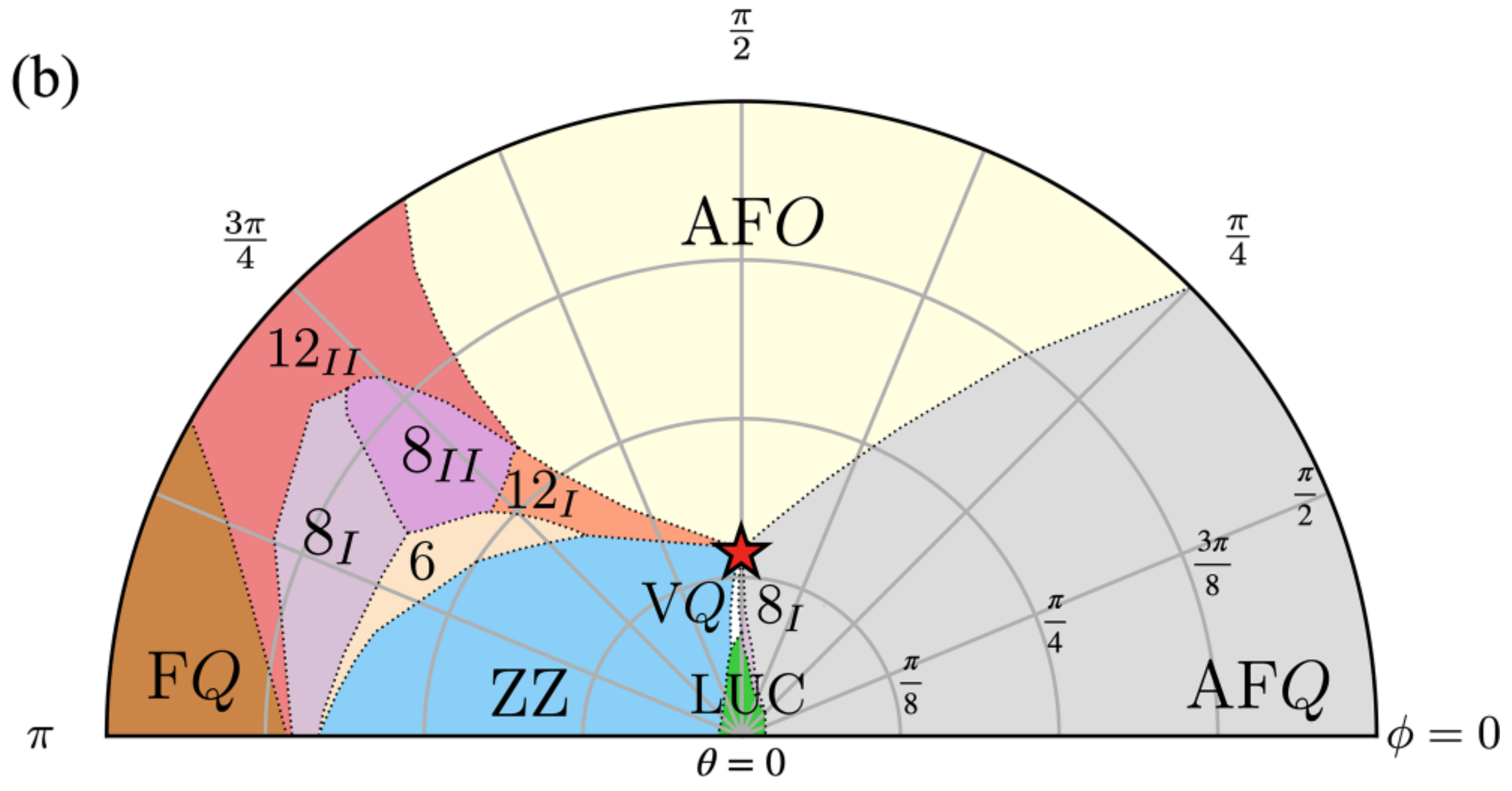

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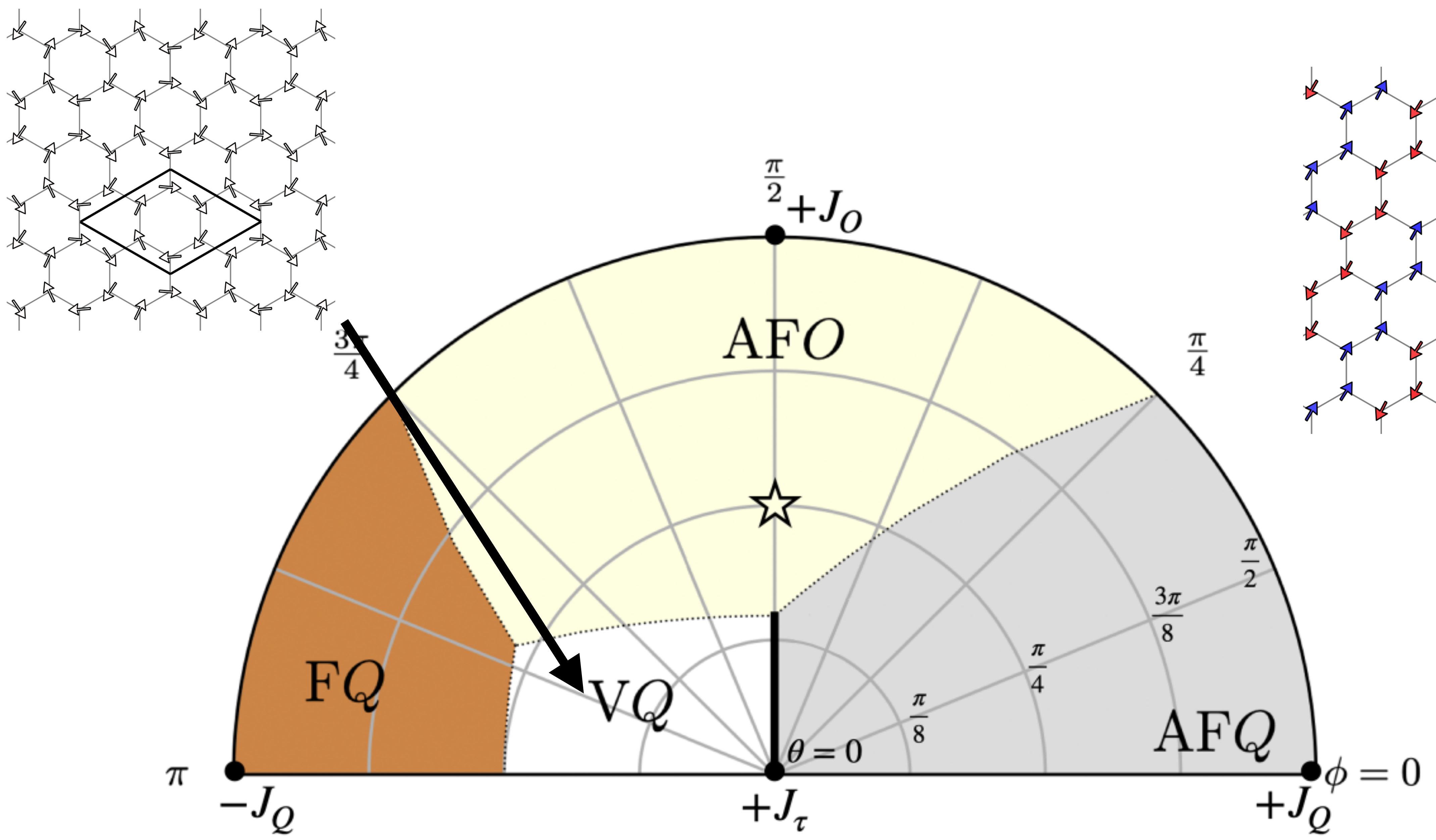
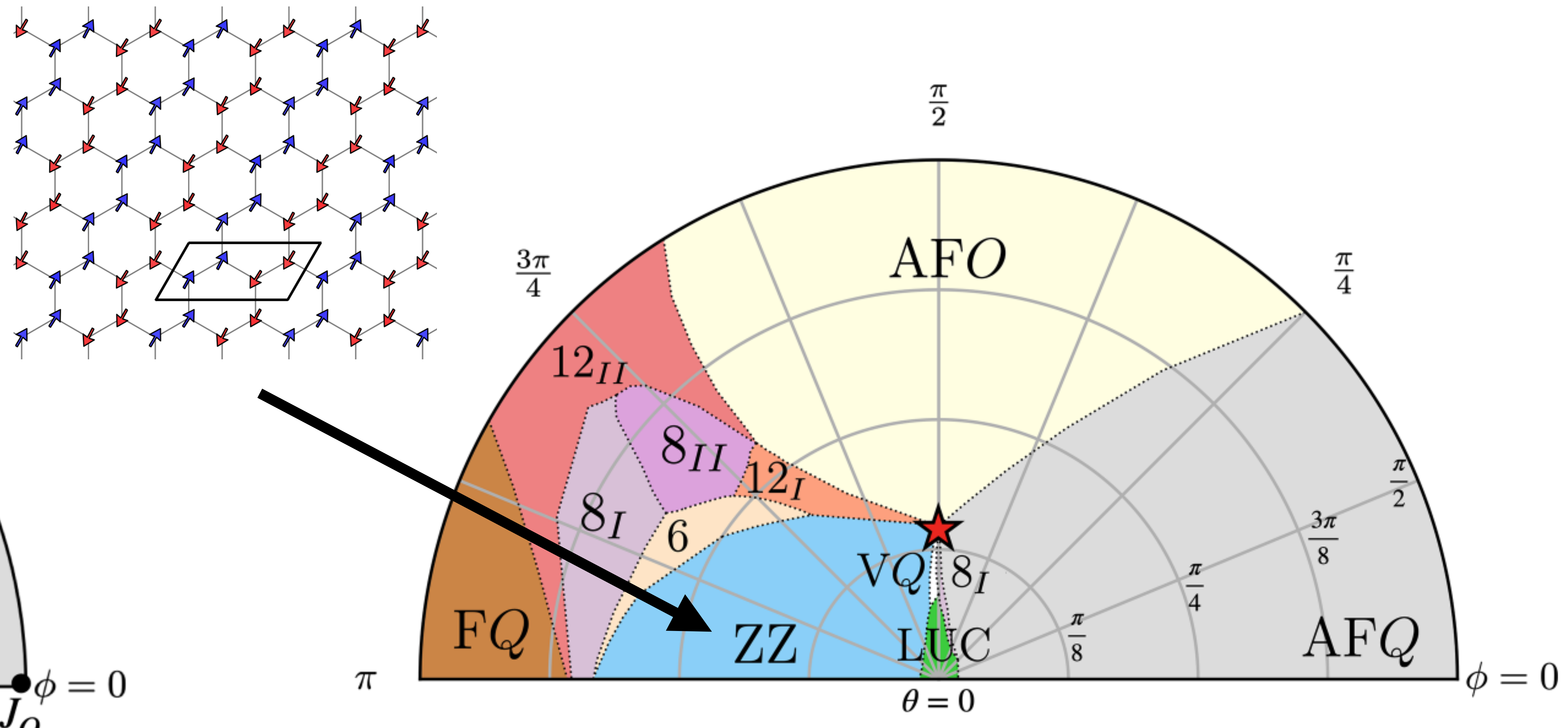


$J_B = 0$



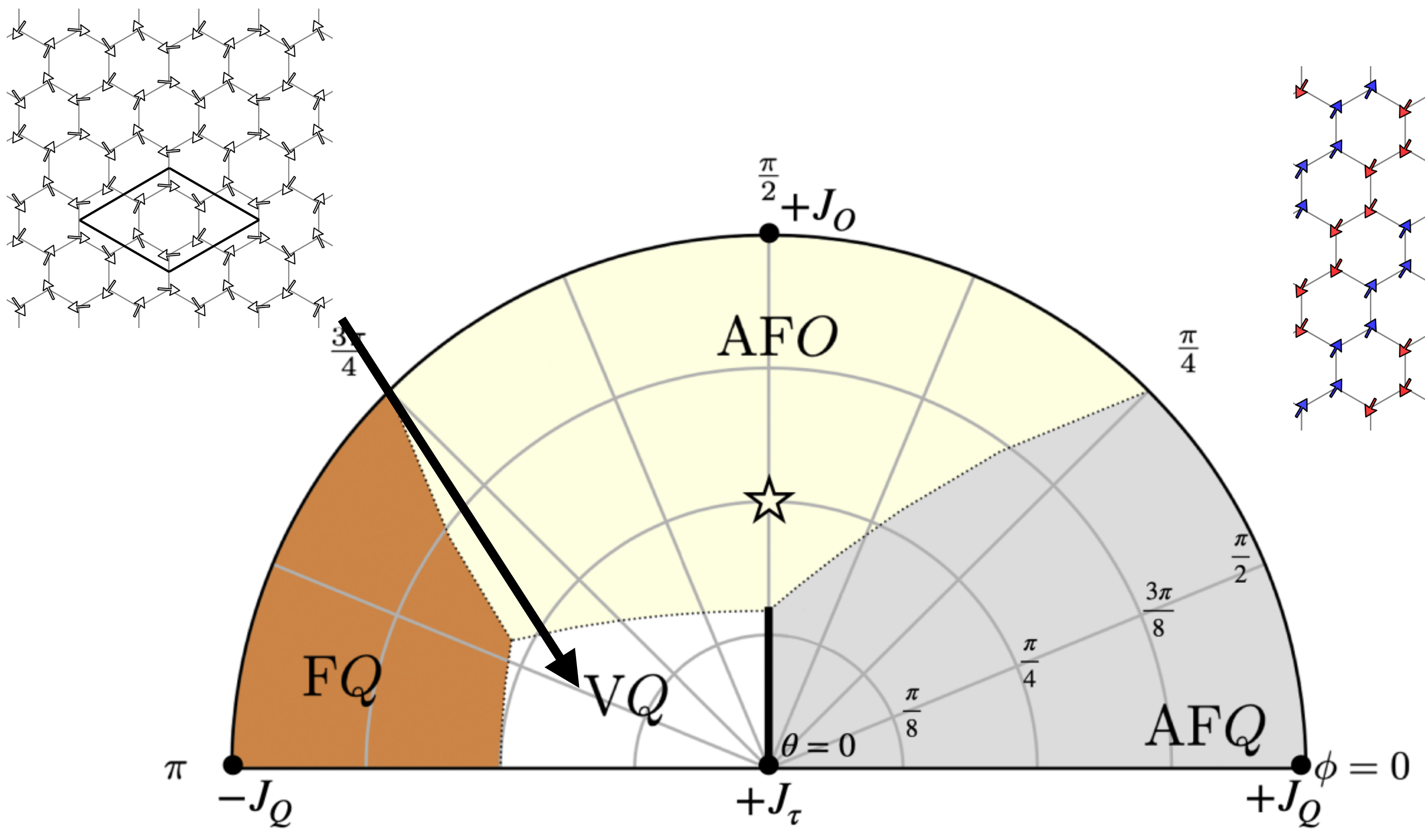
$J_B \neq 0$

$$J_\tau = \cos\theta$$

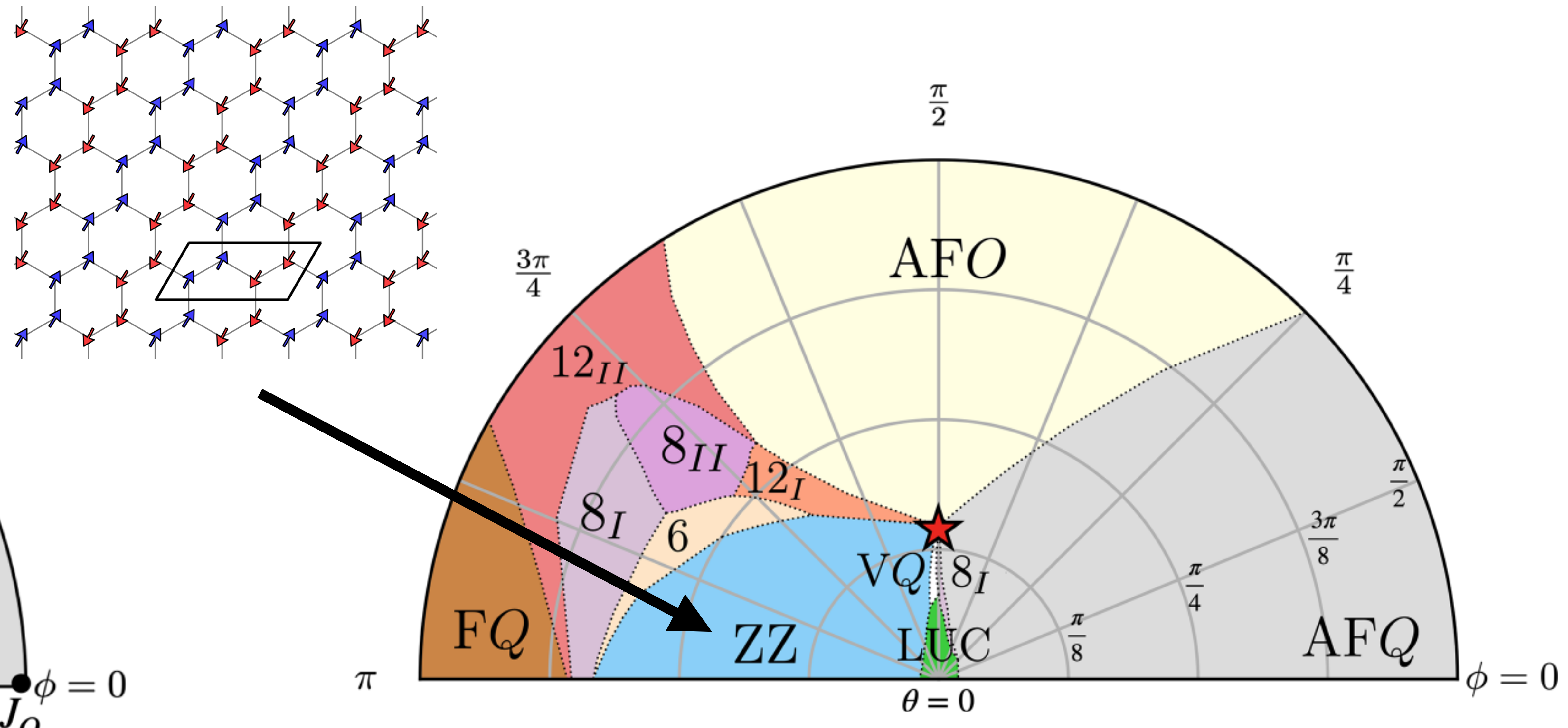
$$J_Q = \sin\theta\cos\phi$$

$$J_O = \sin\theta\sin\phi$$

Classical phase diagram (Monte Carlo simulated annealing): 5d² honeycomb lattice



$J_B = 0$



$J_B \neq 0$

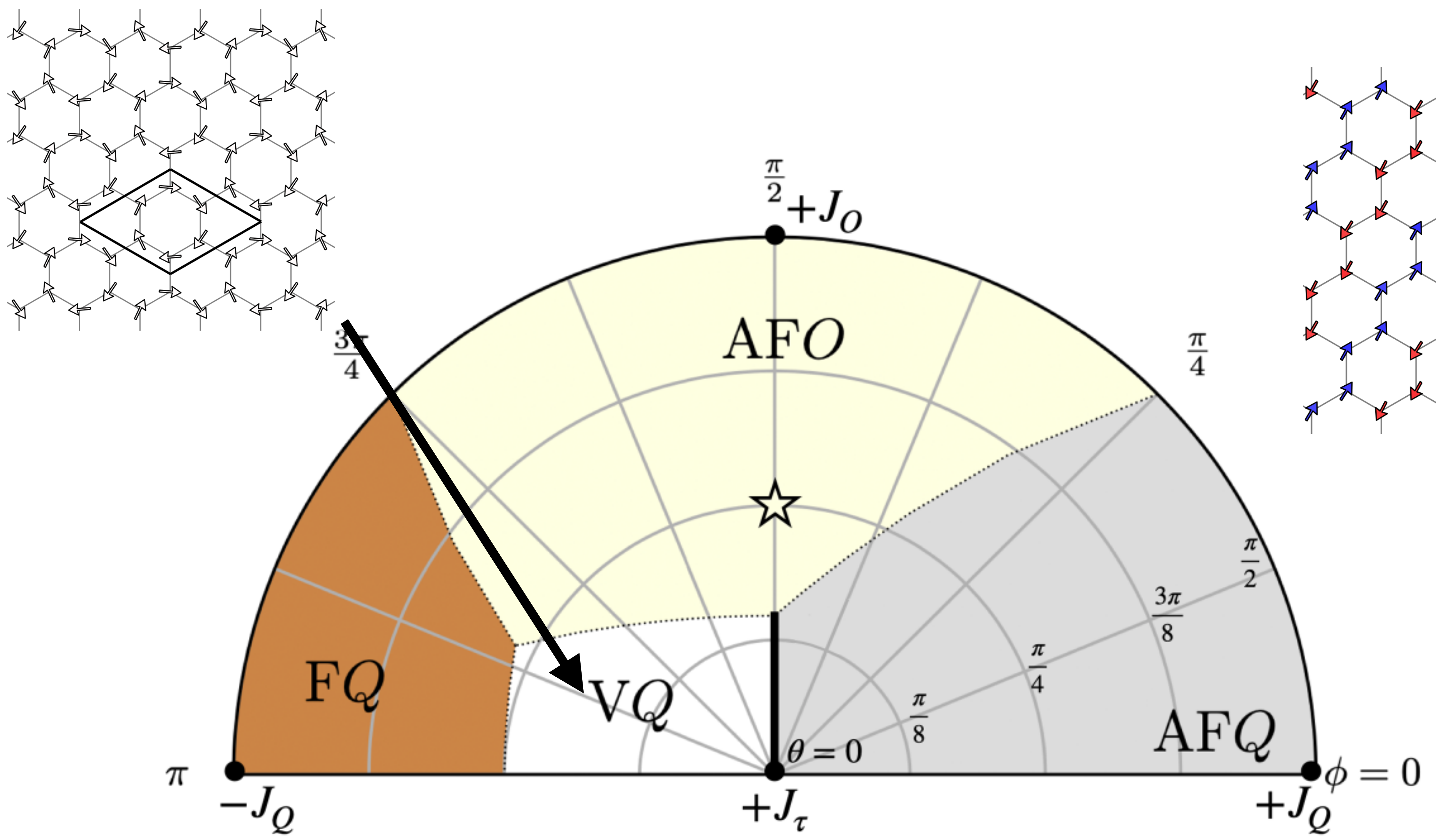
$J_\tau = \cos\theta$

$J_Q = \sin\theta\cos\phi$

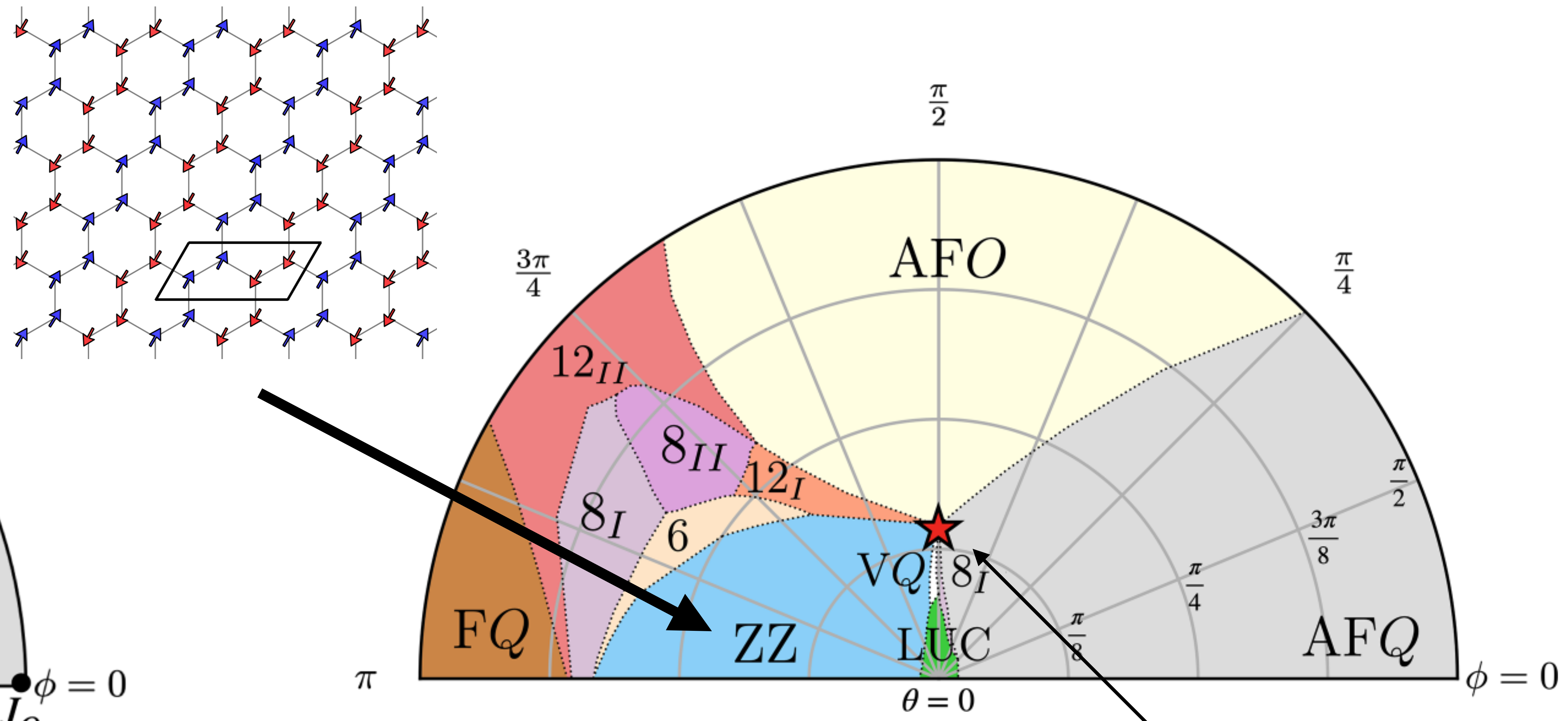
$J_O = \sin\theta\sin\phi$

magnetic field introduces frustration

Classical phase diagram (Monte Carlo simulated annealing): $5d^2$ honeycomb lattice



$J_B = 0$



$J_B \neq 0$

$J_\tau = \cos\theta$

$J_Q = \sin\theta\cos\phi$

$J_O = \sin\theta\sin\phi$

pure Kitaev limit

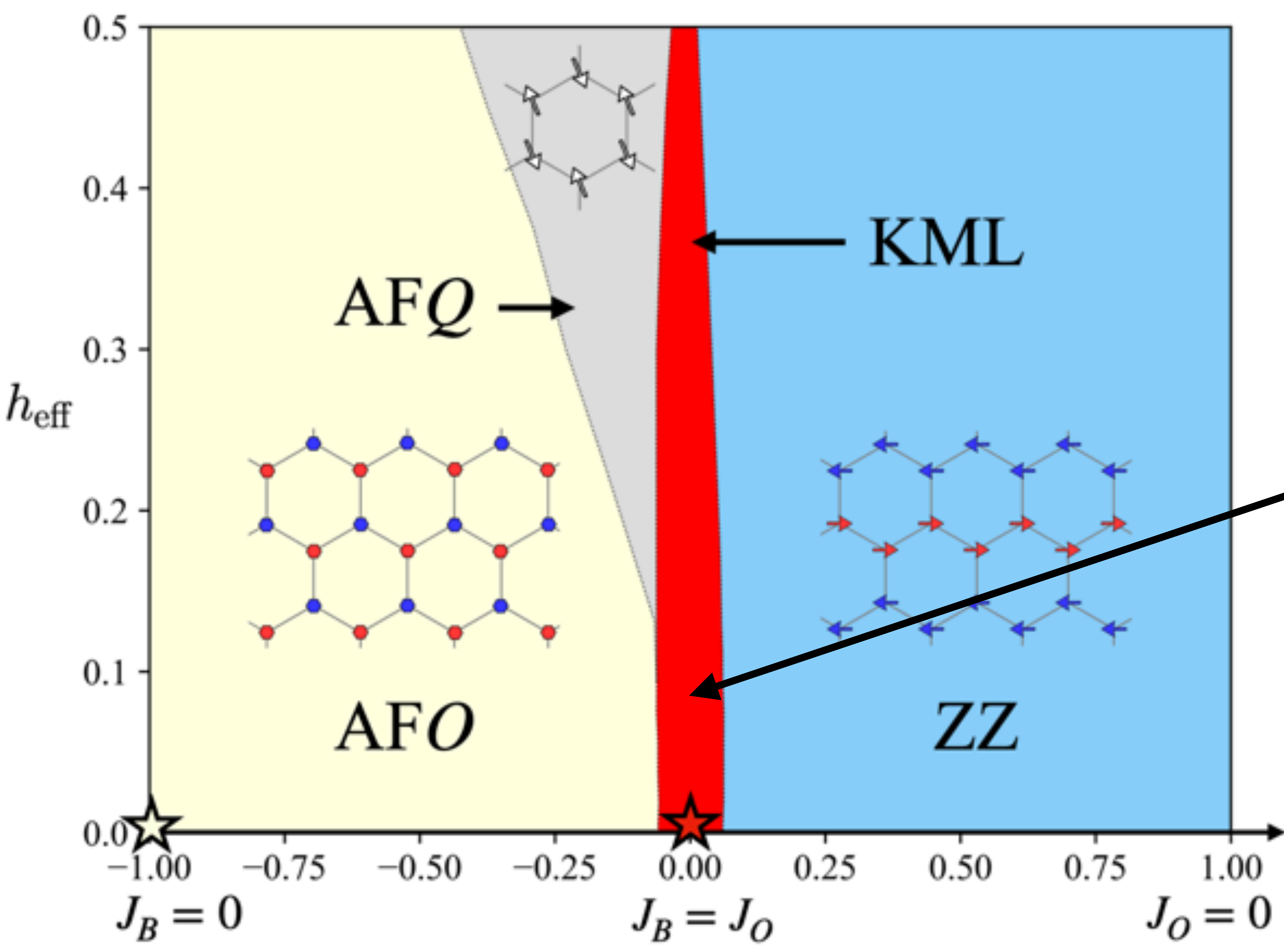
$J_q = 0$

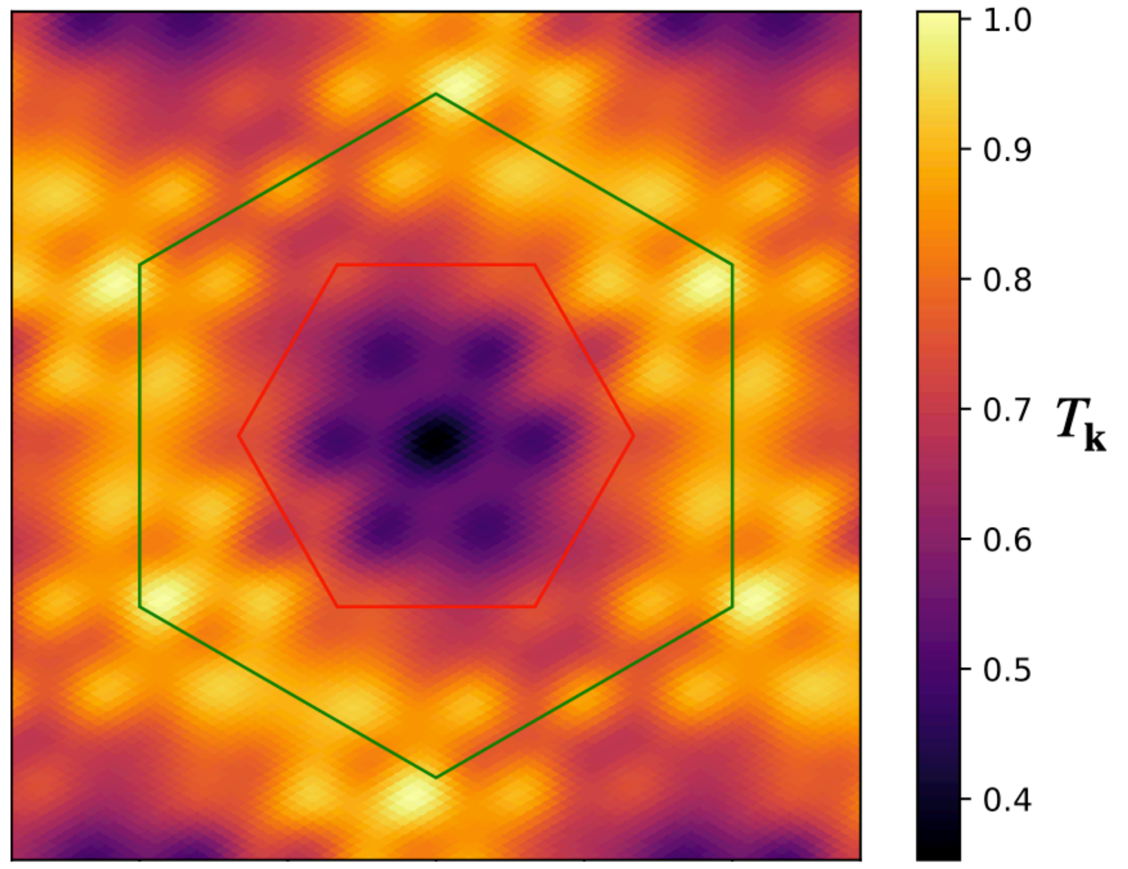
$J_\tau : J_o : J_B = 2 : 1 : 1$

magnetic field introduces frustration

Quantum phase diagram (24-site ED)

M. Kawamura et al. Computer Phys. Comm **217** (2017)





$$\xi = (J_B - J_o)/(J_B + J_o)$$

$$J_\tau = J_B + J_o = 1$$

A. Rayyan, D. Churchill, HYK, PRB 107, L020408 (2023)

Summary

- In $5d^2$ honeycomb Mott insulators, magnetic field introduces the frustration & moves toward the Kitaev + field limit
- $5d^2$ honeycomb Mott insulator offers Kitaev Multipolar liquids