

Quantum Spin Liquid and Quantum Spin Ice: Introduction II

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Thermodynamic signatures in quantum spin liquids

Does spinon pairing exist ?

No

Yes

U(1) Spin Liquid

Emergent U(1) gauge field

Z₂ Spin Liquid (BCS of spinons)

U(1) gauge field gapped
(Meissner effect)

gapped
spinon

gapless
spinon

gapped
spinon

gapless
spinon

$$C_{\text{gauge}}^{3D}(T) \sim T^3$$

(unstable in 2D)

$$C_{\text{dirac}}(T) \sim T^2$$

$$C_{\text{FS}}(T) \sim T \quad \text{mean field}$$

$$C_{\text{FS}}^{2D} \sim T^{2/3} \quad \text{including fluctuations}$$
$$C_{\text{FS}}^{3D} \propto T \ln T$$

$$C(T) \sim e^{-\Delta/T}$$

$$C_{\text{d-wave}}(T) \sim T^2$$

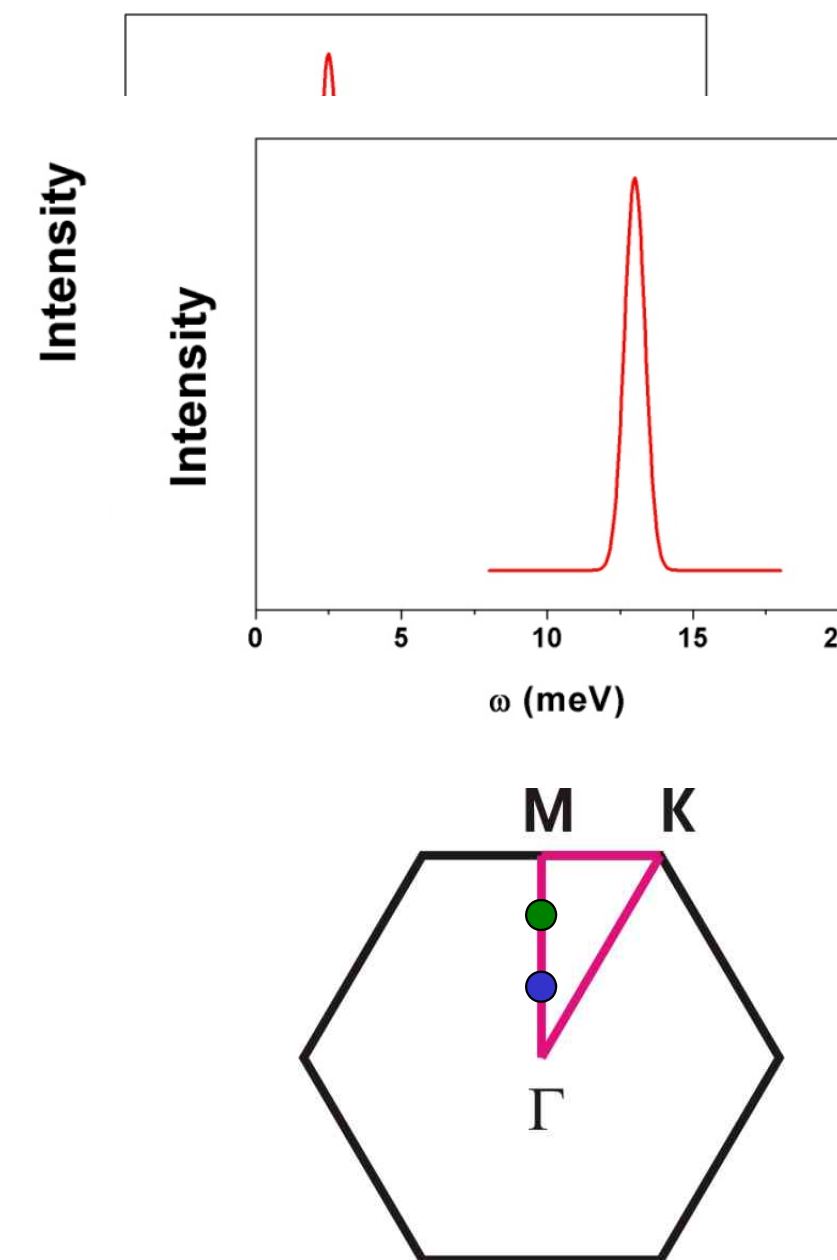
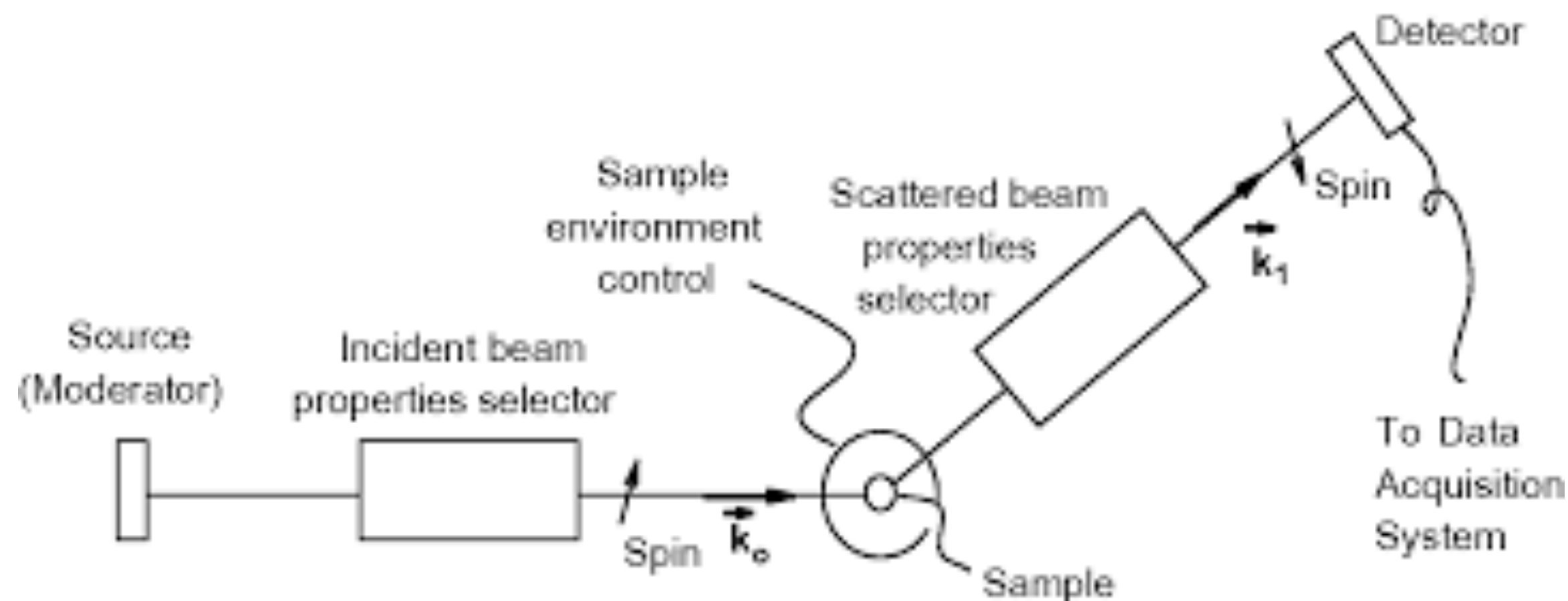
Neutron Scattering

Time-dependent correlator

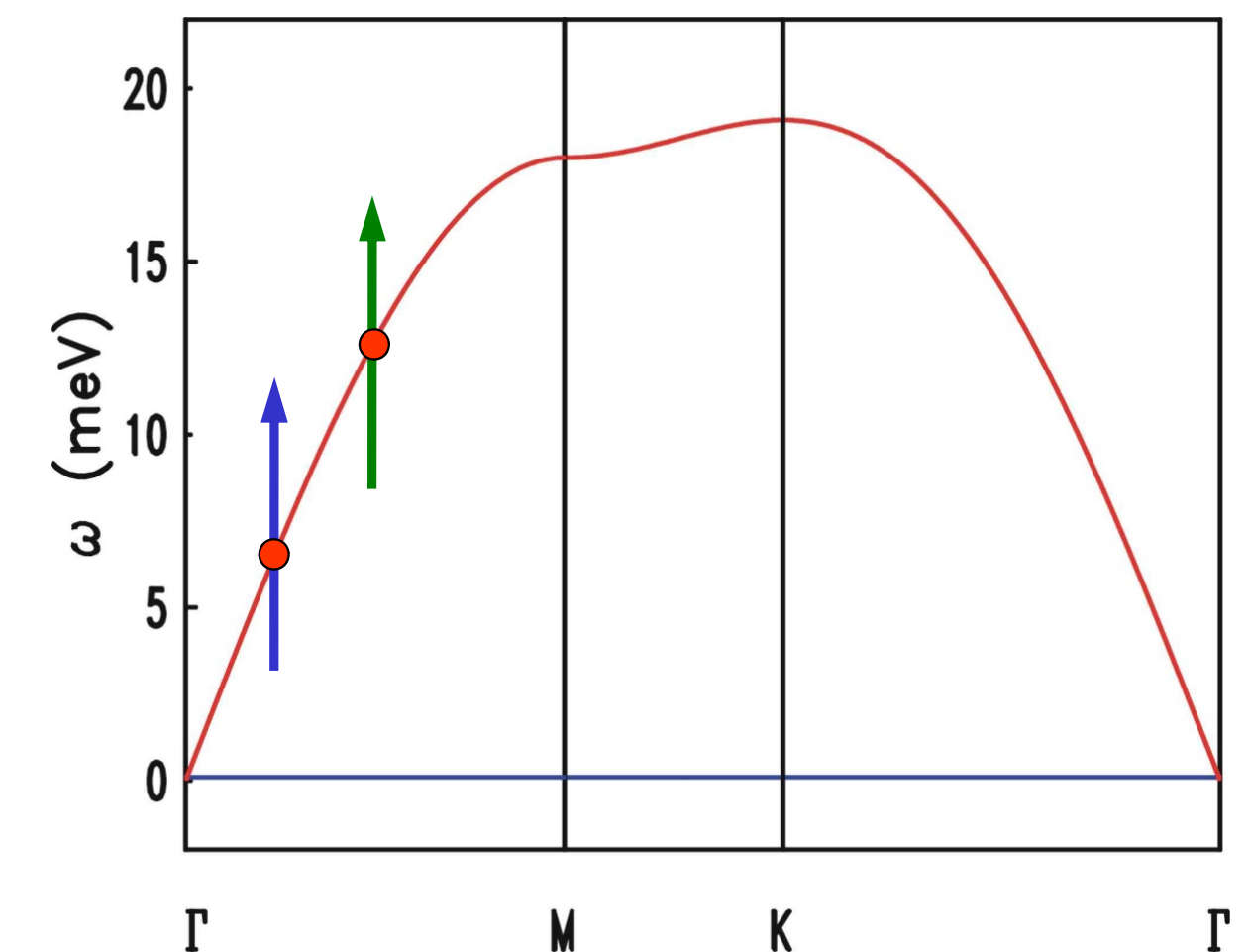
$$\sum_{\alpha=x,y,z} \langle S^\alpha(\mathbf{x}, t) S^\alpha(\mathbf{x} = 0, t = 0) \rangle$$

Dynamical spin structure factor (DSSF)

$$\longrightarrow \mathcal{S}(\mathbf{q}, \omega) \sim \int dt \int d^3\mathbf{x} e^{i\omega t} e^{i\mathbf{q} \cdot \mathbf{x}} \sum_{\alpha=x,y,z} \langle S^\alpha(\mathbf{x}, t) S^\alpha(\mathbf{x} = 0, t = 0) \rangle$$



Magnons



How to detect excitations in quantum spin liquids ?

Neutron Scattering: Spin-1 excitations

Spinon-Antispinon pair excitations

Scattering continuum

$$\omega_{\mathbf{q}} \sim \min \left[\varepsilon_{\frac{\mathbf{q}}{2} + \frac{\mathbf{p}}{2}} + \varepsilon_{\frac{\mathbf{q}}{2} - \frac{\mathbf{p}}{2}} \right]$$

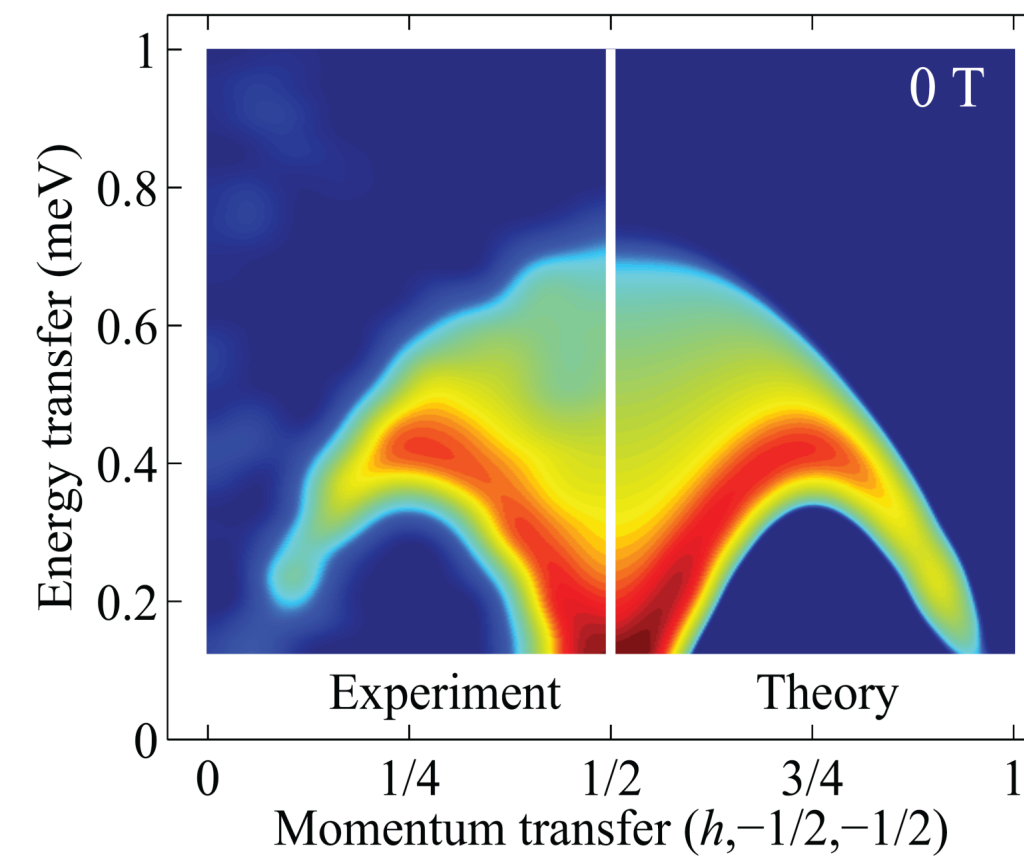
for all possible \mathbf{p}

Harder to confirm in 2D and 3D

Energy resolution may not be good enough

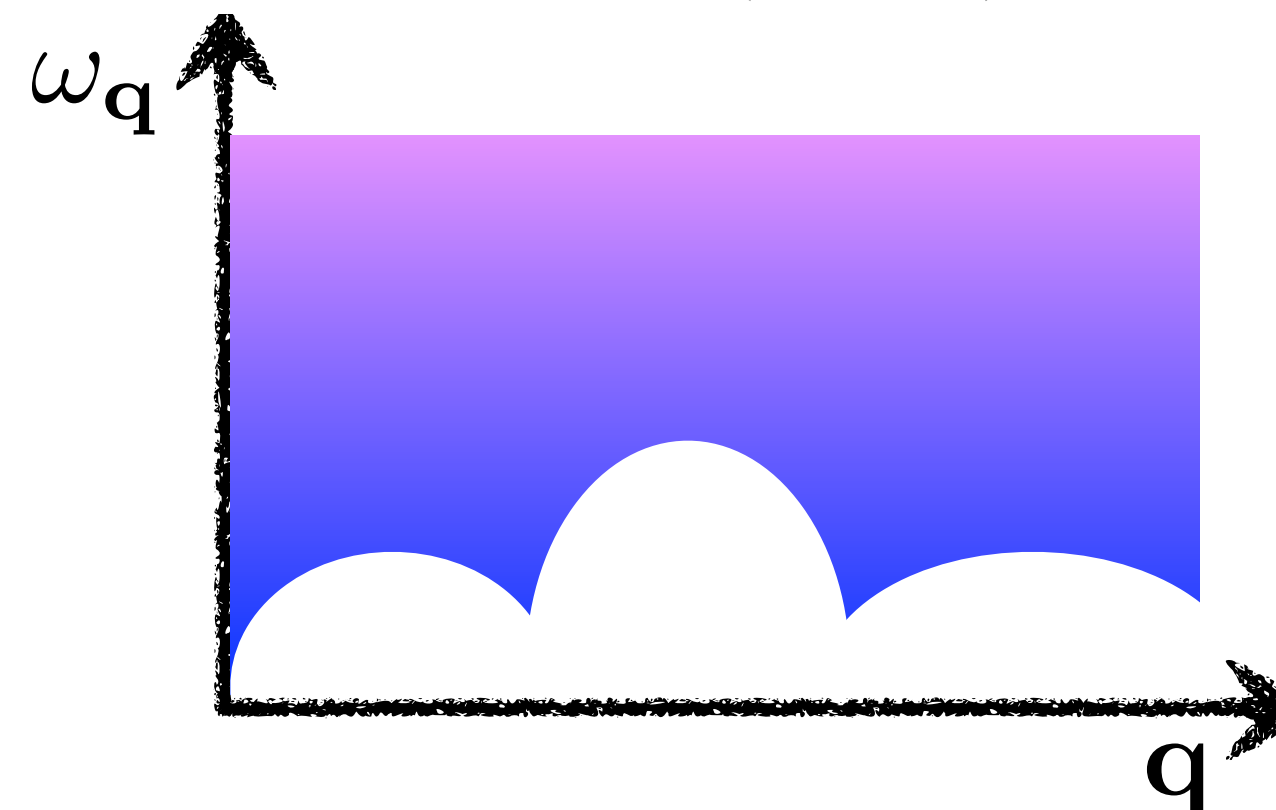
Well-defined dispersion -->
Threshold energy for pair excitations

CuSO4.5D2O



One dimensional spin liquid

Mourigal et al,
Nat. Phys. (2013)



?

Spin flip excitations in Kitaev Model

Define bond fermions

$$\chi_{\langle jk \rangle}^{\alpha} = \frac{1}{2} (b_j^{\alpha} + i b_k^{\alpha})$$

$$\chi_{\langle jk \rangle}^{\alpha\dagger} = \frac{1}{2} (b_j^{\alpha} - i b_k^{\alpha})$$

Baskaran, Mandal, Shankar (2007)

$$\tilde{H} = \sum_{\alpha\text{-bond}} i \hat{u}_{jk}^{\alpha} c_j c_k$$

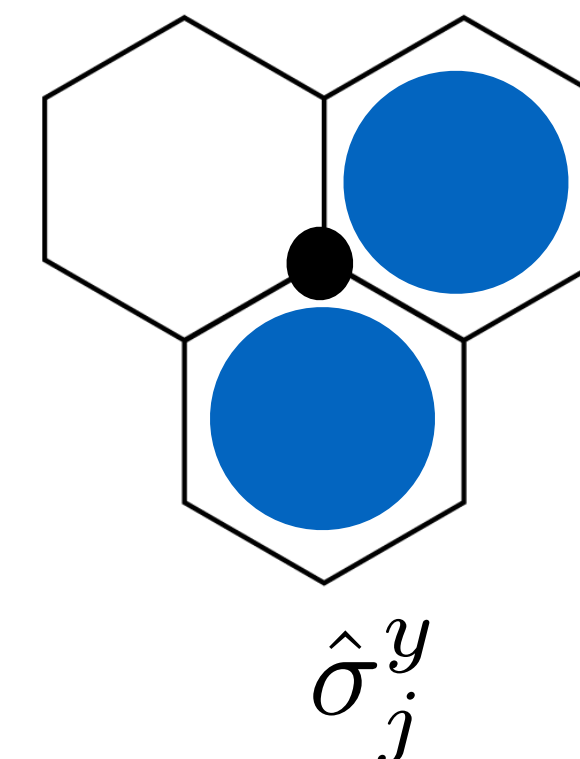
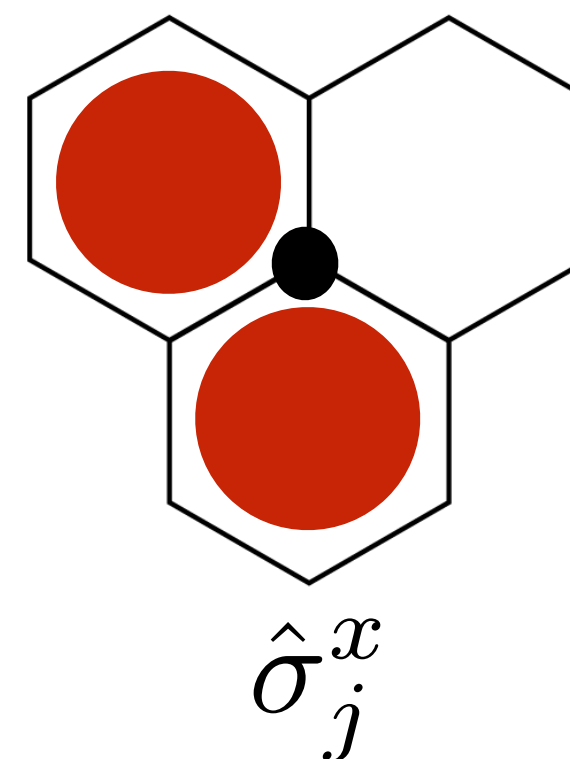
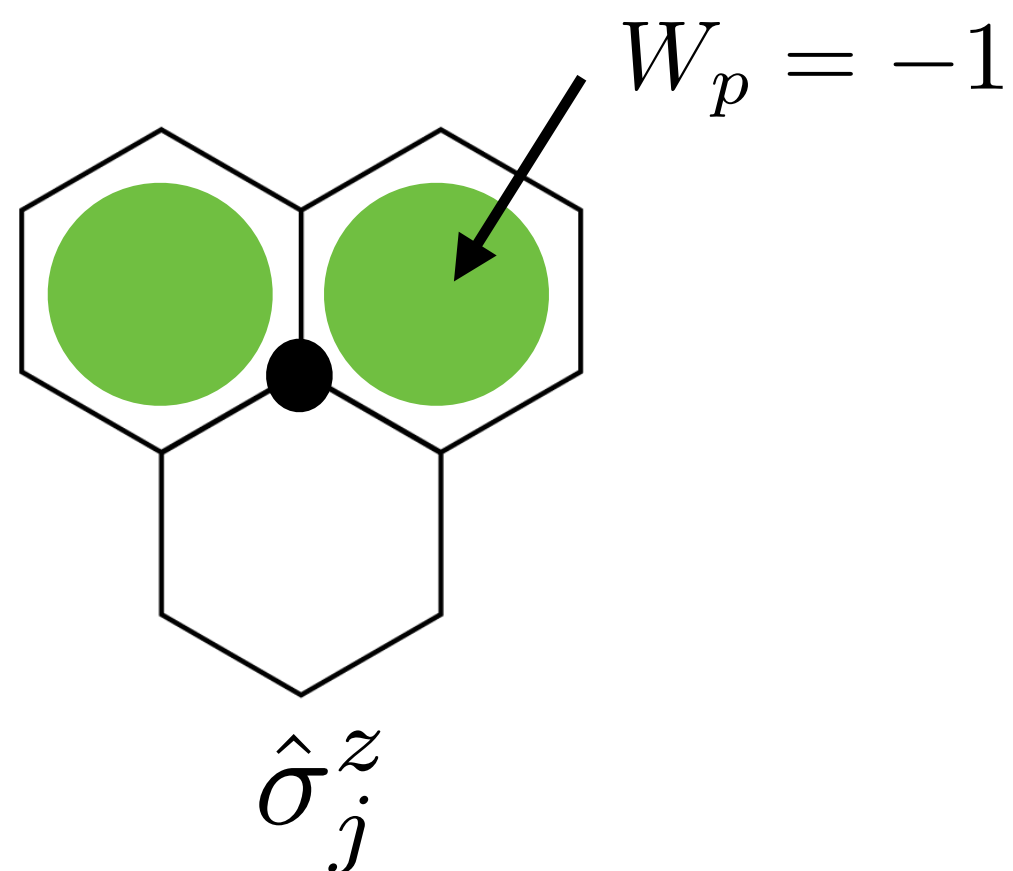
$$\hat{u}_{jk}^{\alpha} = 2 \chi_{\langle jk \rangle_{\alpha}}^{\dagger} \chi_{\langle jk \rangle_{\alpha}} - 1$$

Ground state

$$\chi_{\langle jk \rangle_{\alpha}}^{\dagger} \chi_{\langle jk \rangle_{\alpha}} = 1 \quad \hat{u}_{jk}^{\alpha} = +1$$

$$\chi_{\langle jk \rangle_{\alpha}}^{\dagger} \chi_{\langle jk \rangle_{\alpha}} = 1, 0 \quad \longleftrightarrow \quad \hat{u}_{jk}^{\alpha} = \pm 1$$

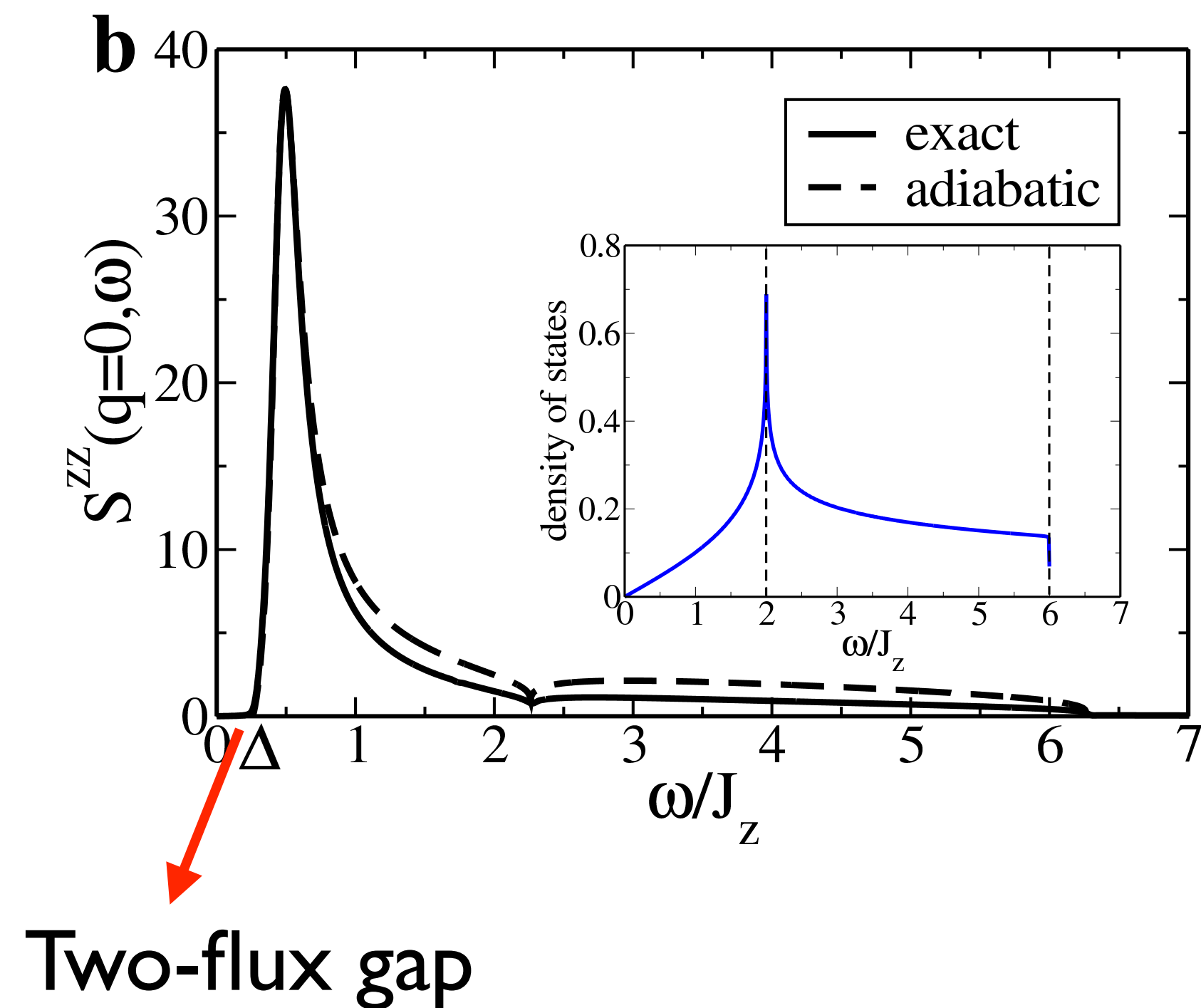
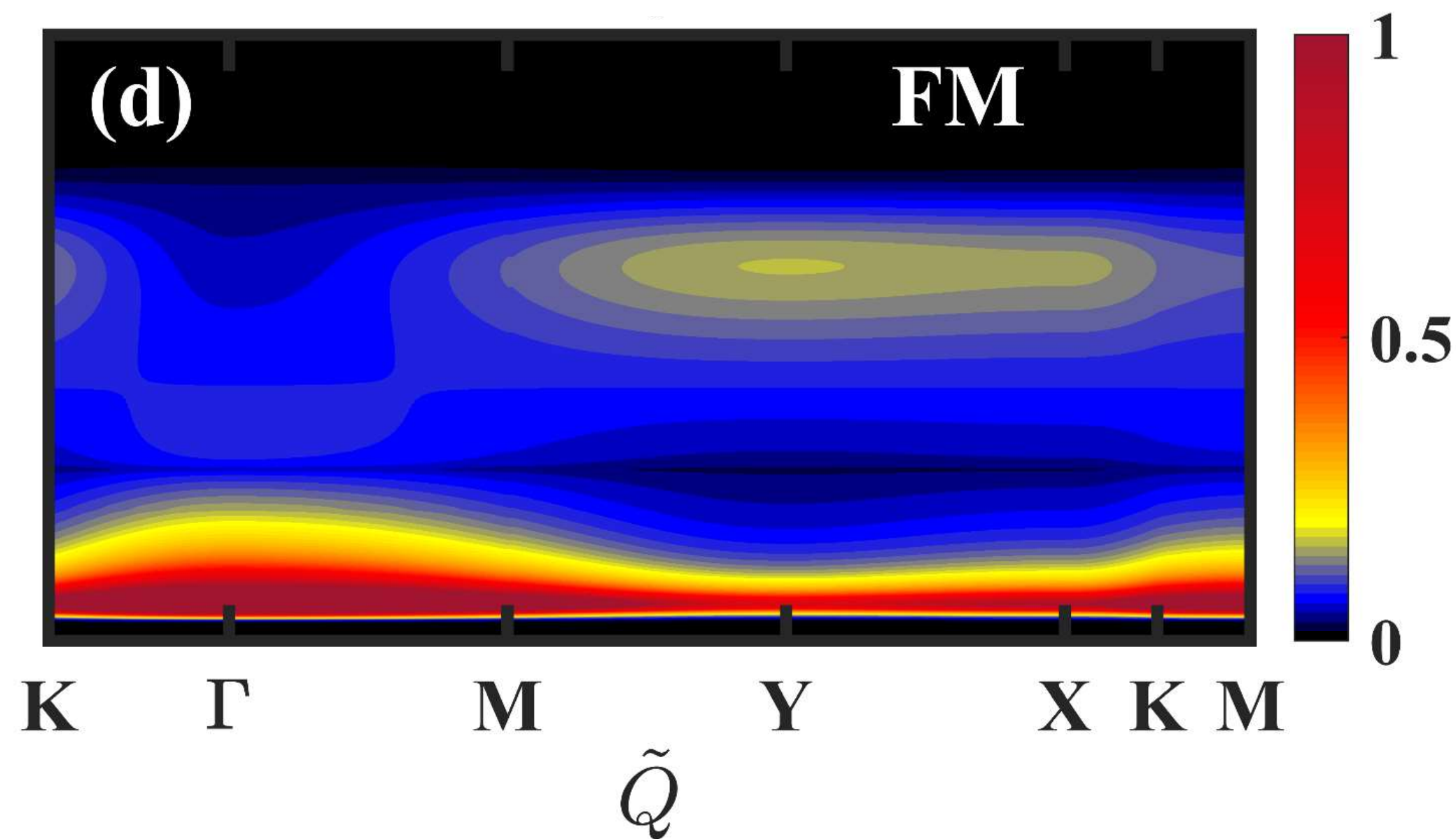
Single spin flip $\sigma_j^{\alpha} = i c_j \left(\chi_{\langle jk \rangle}^{\alpha} + \chi_{\langle jk \rangle}^{\alpha\dagger} \right) \quad \hat{u}_{jk}^{\alpha} \longrightarrow -\hat{u}_{jk}^{\alpha} \quad \longleftrightarrow \quad \text{one fermion and two fluxes}$



Dynamical Spin Structure Factor

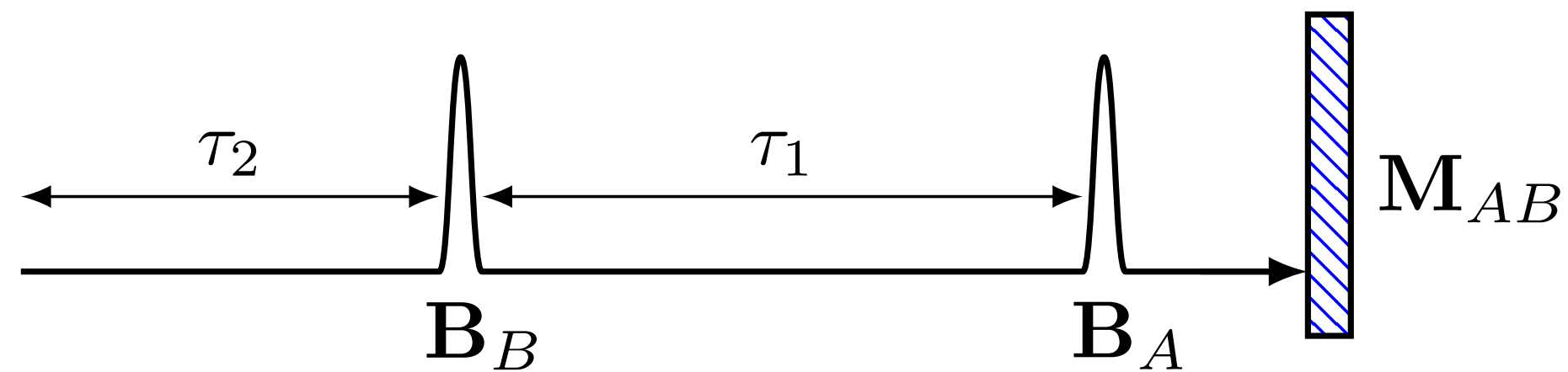
$$S_{ij}^{ab}(t) = \langle 0 | \hat{\sigma}_i^a(t) \hat{\sigma}_j^b(0) | 0 \rangle \xrightarrow{\text{F.T.}} S^{ab}(\mathbf{q}, \omega)$$

$$S(\mathbf{q}, \omega) = \sum_a S^{aa}(\mathbf{q}, \omega)$$



J. Knolle, D.L.Kovrizhin, J.T.Chalker, R. Moessner, PRL (2013)

2D nonlinear spectroscopy



$$\mathbf{B}(t) = B_A^\alpha \hat{e}_\alpha \delta(t) + B_B^\beta \hat{e}_\beta \delta(t - \tau_1)$$

Two pulses delayed by τ_1

Measure the nonlinear part of the transient magnetization

$$\mathbf{M}_{NL}(t) = \mathbf{M}_{AB}(t) - \mathbf{M}_A(t) - \mathbf{M}_B(t) \quad t = \tau_1 + \tau_2$$

Nonlinear transient magnetization - nonlinear susceptibilities

$$H_{\text{tot}} = H - \mathbf{B}(t) \cdot \mathbf{M}(t)$$

$$M_{NL}^\gamma(\tau_1 + \tau_2)/N = \chi_{\alpha\beta}^{(2),\gamma}(\tau_2, \tau_1) B_A^\alpha B_B^\beta \quad \text{AB}$$

$$+ \chi_{\alpha\alpha\beta}^{(3),\gamma}(\tau_2, \tau_1, 0) B_A^\alpha B_A^\alpha B_B^\beta + \chi_{\alpha\beta\beta}^{(3),\gamma}(\tau_2, 0, \tau_1) B_A^\alpha B_B^\beta B_B^\beta + \mathcal{O}(B^4)$$

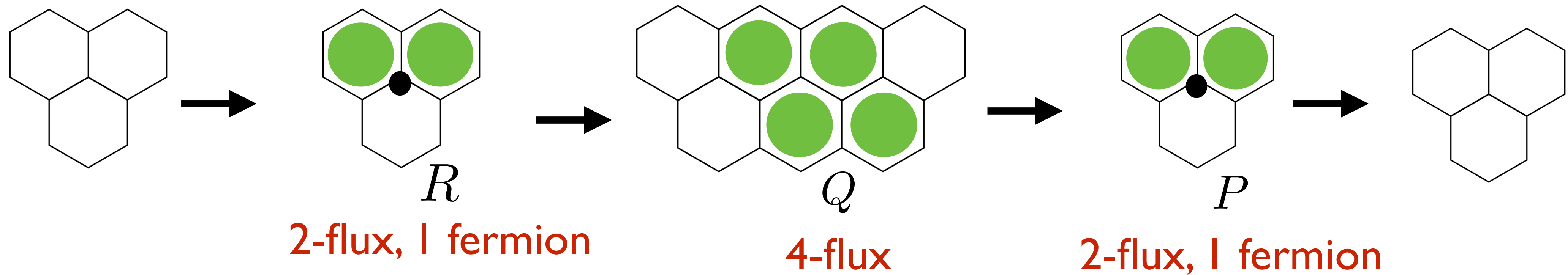
AAB ABB

$\alpha, \beta, \gamma = z, z, z$ case

3rd order susceptibility

$$R_{zzz}^{(1),z}(\tau_2, 0, \tau_1) = \sum_{jklm} \sum_{PQR} \langle 0 | \hat{\sigma}_j^z | P \rangle \langle P | \hat{\sigma}_k^z | Q \rangle \langle Q | \hat{\sigma}_l^z | R \rangle \langle R | \hat{\sigma}_m^z | 0 \rangle e^{-i(E_R - E_0)\tau_2 - i(E_P - E_0)\tau_1}$$

ABB



$$(\omega_2, \omega_1) = (E_R - E_0, E_P - E_0) = ((E_2 - E_0) + \varepsilon_R, (E_2 - E_0) + \varepsilon_P)$$

$$E_R = E_2 + \varepsilon_R$$

$$E_P = E_2 + \varepsilon_P$$

E_2 Two flux energy

ε_R Fermion energy

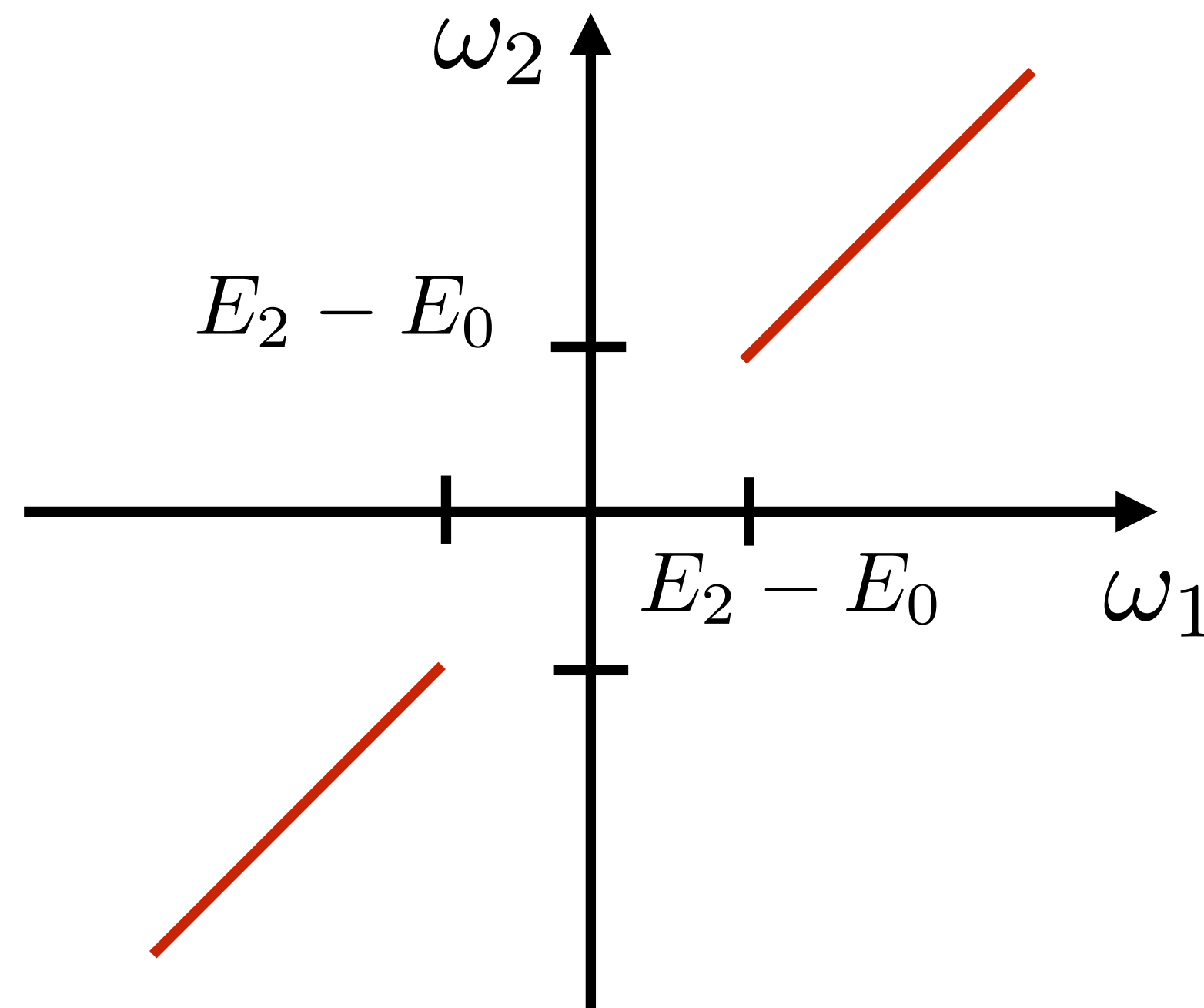
$\alpha, \beta, \gamma = z, z, z$ case

3rd order susceptibility

$$R_{zzz}^{(1),z}(\tau_2, 0, \tau_1) = \sum_{jklm} \sum_{PQR} \langle 0 | \hat{\sigma}_j^z | P \rangle \langle P | \hat{\sigma}_k^z | Q \rangle \langle Q | \hat{\sigma}_l^z | R \rangle \langle R | \hat{\sigma}_m^z | 0 \rangle e^{-i(E_R - E_0)\tau_2 - i(E_P - E_0)\tau_1}$$

ABB

$$(\omega_2, \omega_1) = (E_R - E_0, E_P - E_0) = ((E_2 - E_0) + \varepsilon_R, (E_2 - E_0) + \varepsilon_P)$$

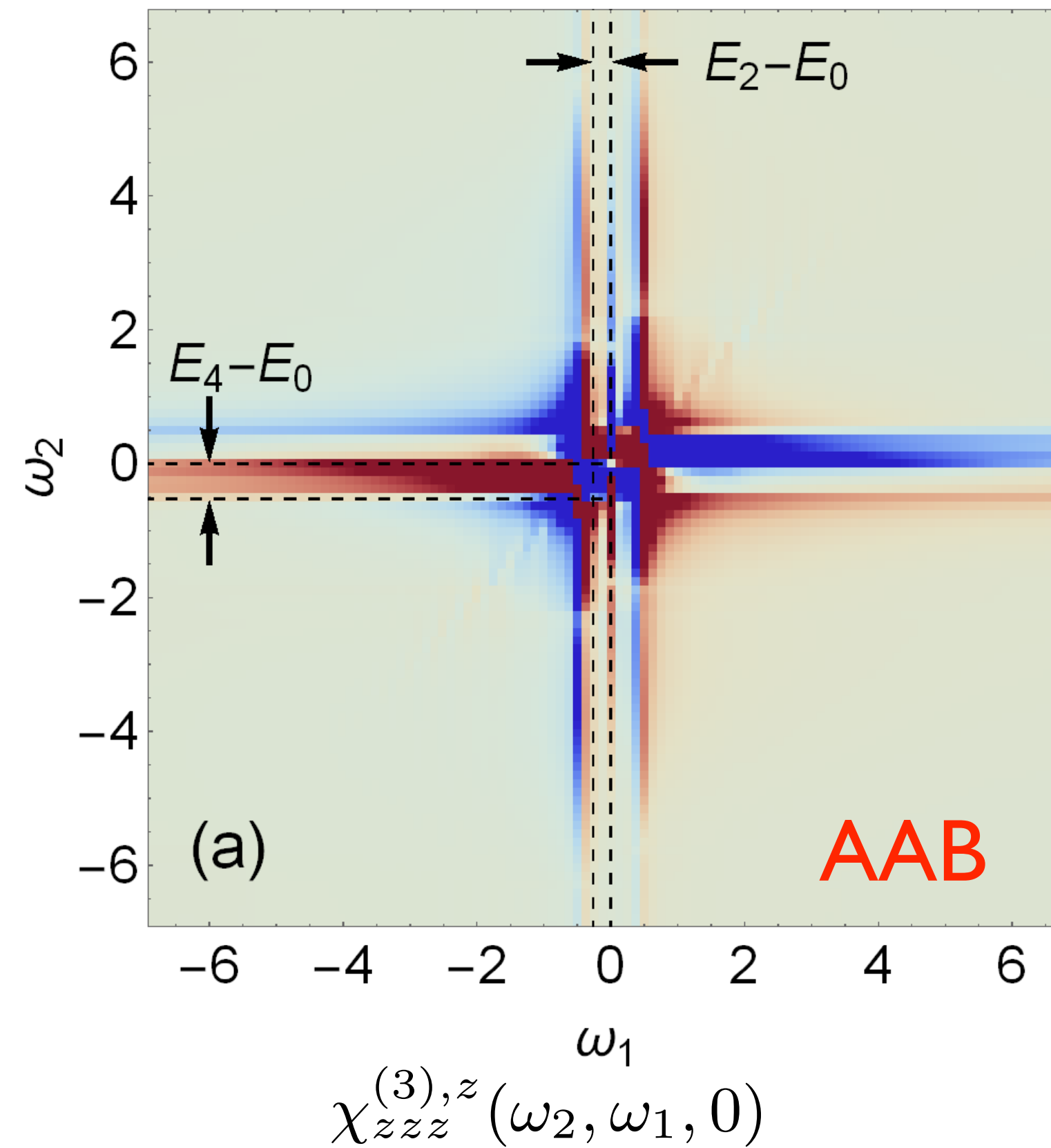


For $\varepsilon_R = \varepsilon_P$

$$\omega_2 = \omega_1 \geq E_2 - E_0$$

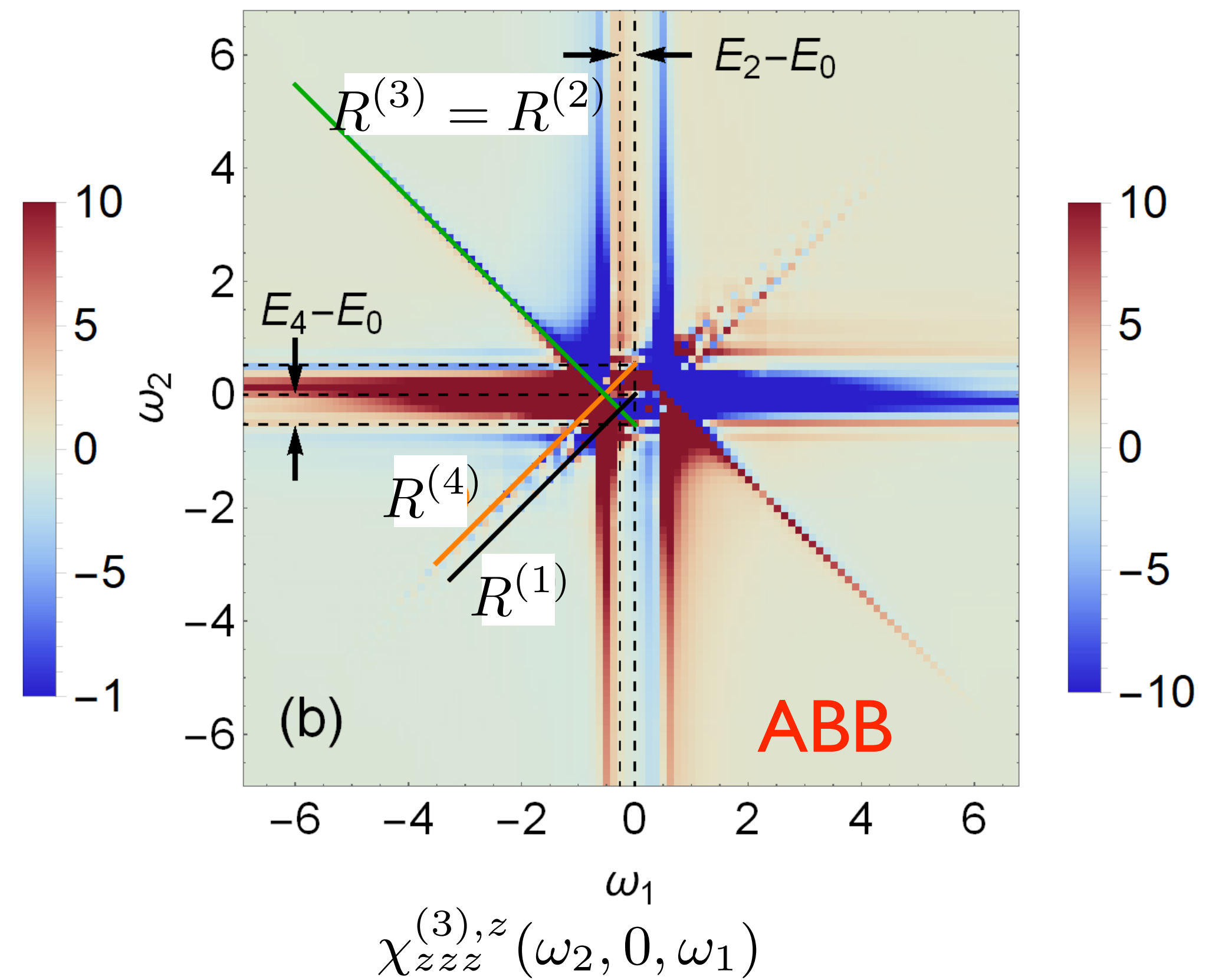
E_2 Two flux energy

2D Nonlinear susceptibility



$E_2 - E_0$ Two-flux gap
 $E_4 - E_0$ Four-flux gap

3rd order susceptibility



Wonjune Choi, Ki Hoon Lee, YBK,
 PRL 124, 117205, (2020)

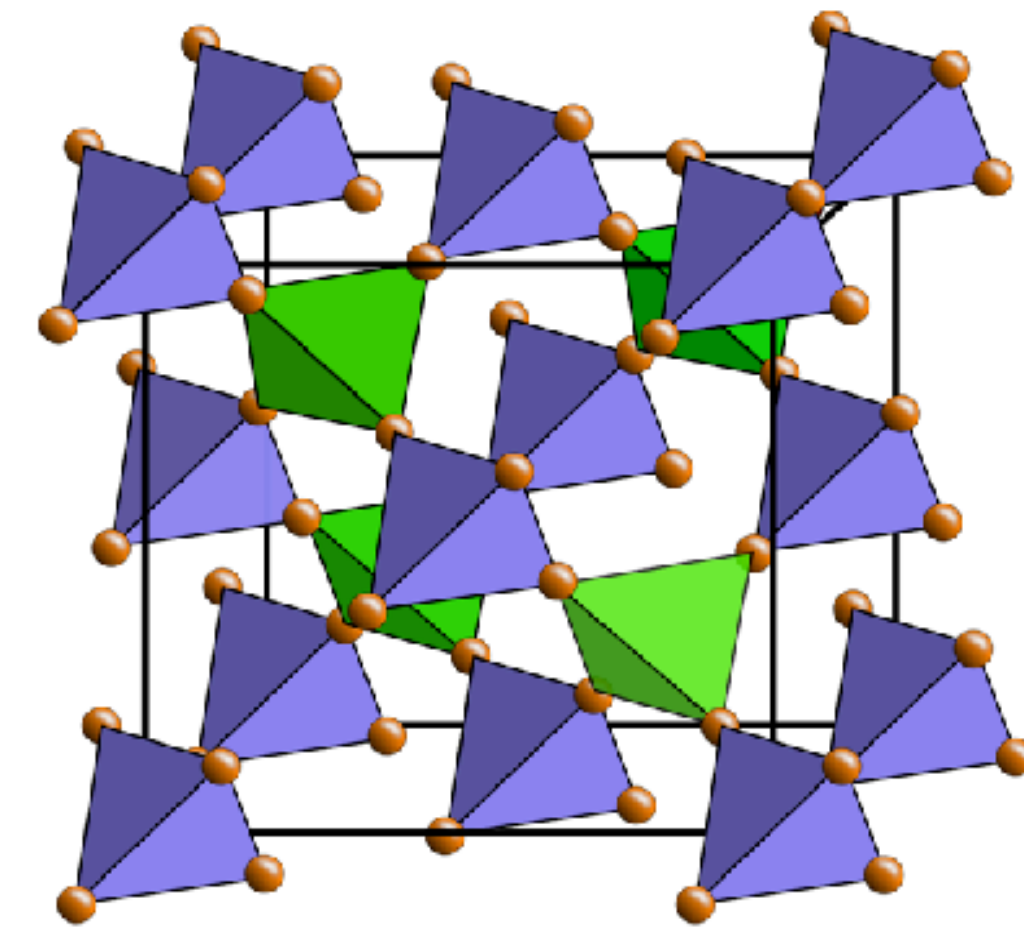
Emily Z. Zhang, Ciaran Hickey, YBK,
 PRB 110, 104415, (2024)

Quantum Spin Ice: a primer

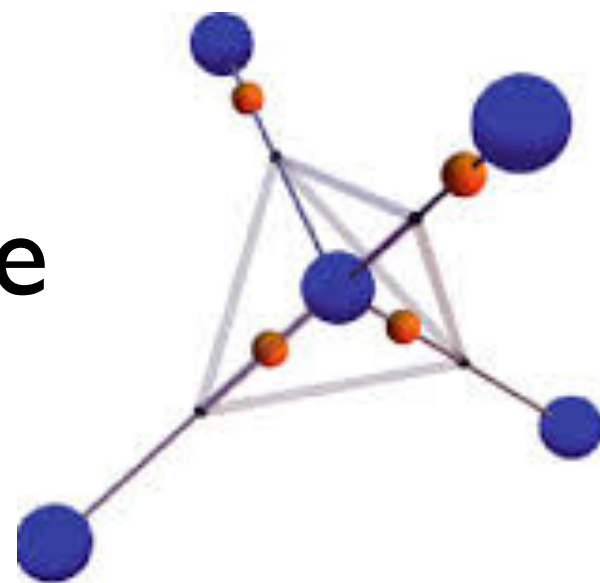
Classical Spin Ice

$$\mathcal{H}_I = J_z \sum_{\langle ij \rangle} S_i^z S_j^z = \frac{J_z}{2} \sum_{\triangleleft} (S_{\triangleleft}^z)^2 + \text{constant}$$

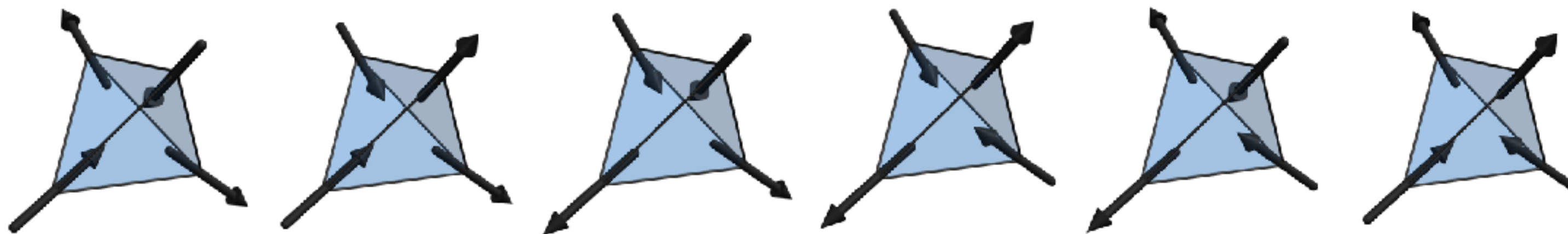
$$S_{\triangleleft}^z = \sum_{i \in \triangleleft} S_i^z = 0 \quad \text{2-in/2-out}$$



Water ice

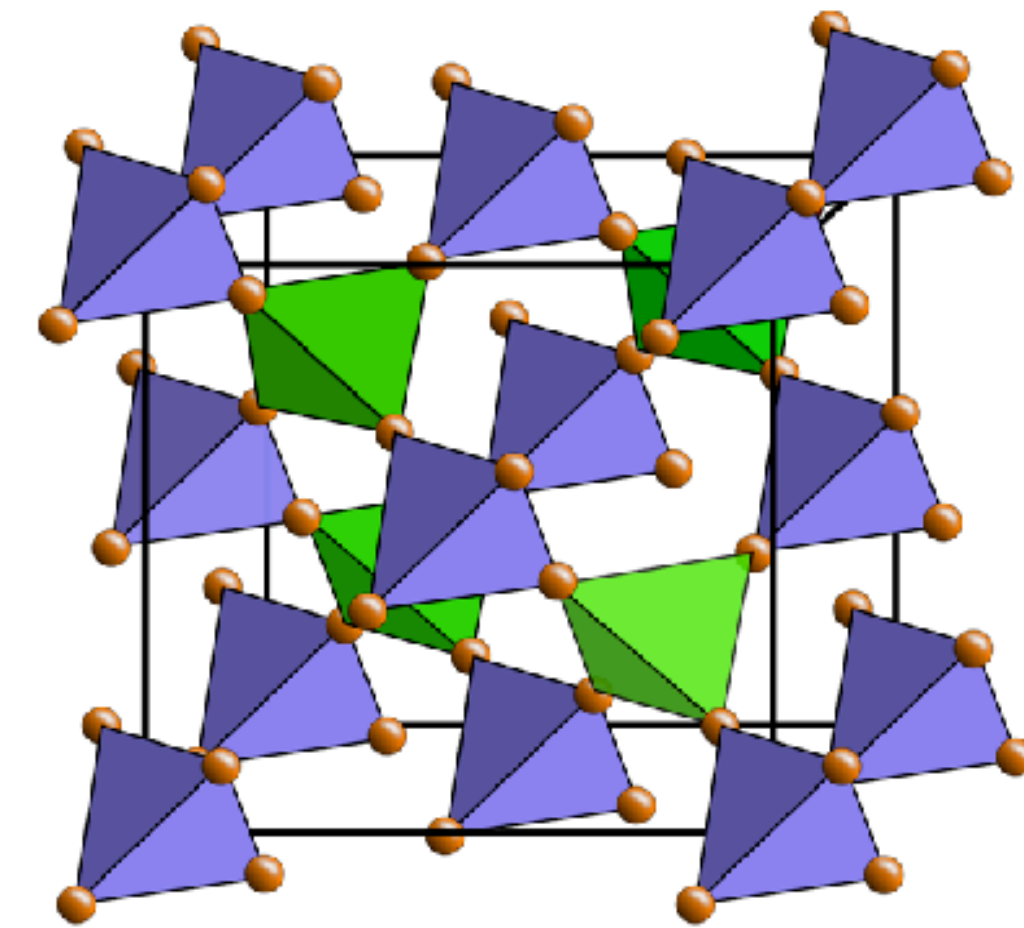


Spin ice



Massive degeneracy

Quantum Spin Ice



$$\mathcal{H}_I = J_z \sum_{\langle ij \rangle} S_i^z S_j^z = \frac{J_z}{2} \sum_{\triangleleft} (S_{\triangleleft}^z)^2 + \text{constant}$$

$$S_{\triangleleft}^z = \sum_{i \in \triangleleft} S_i^z = 0 \quad \text{2-in/2-out}$$

$$\mathcal{H}' = -\frac{J_{\perp}}{2} \sum_{\langle ij \rangle} (S_i^+ S_j^- + h.c.) \quad \text{Hermele, Balents, Fisher '03}$$

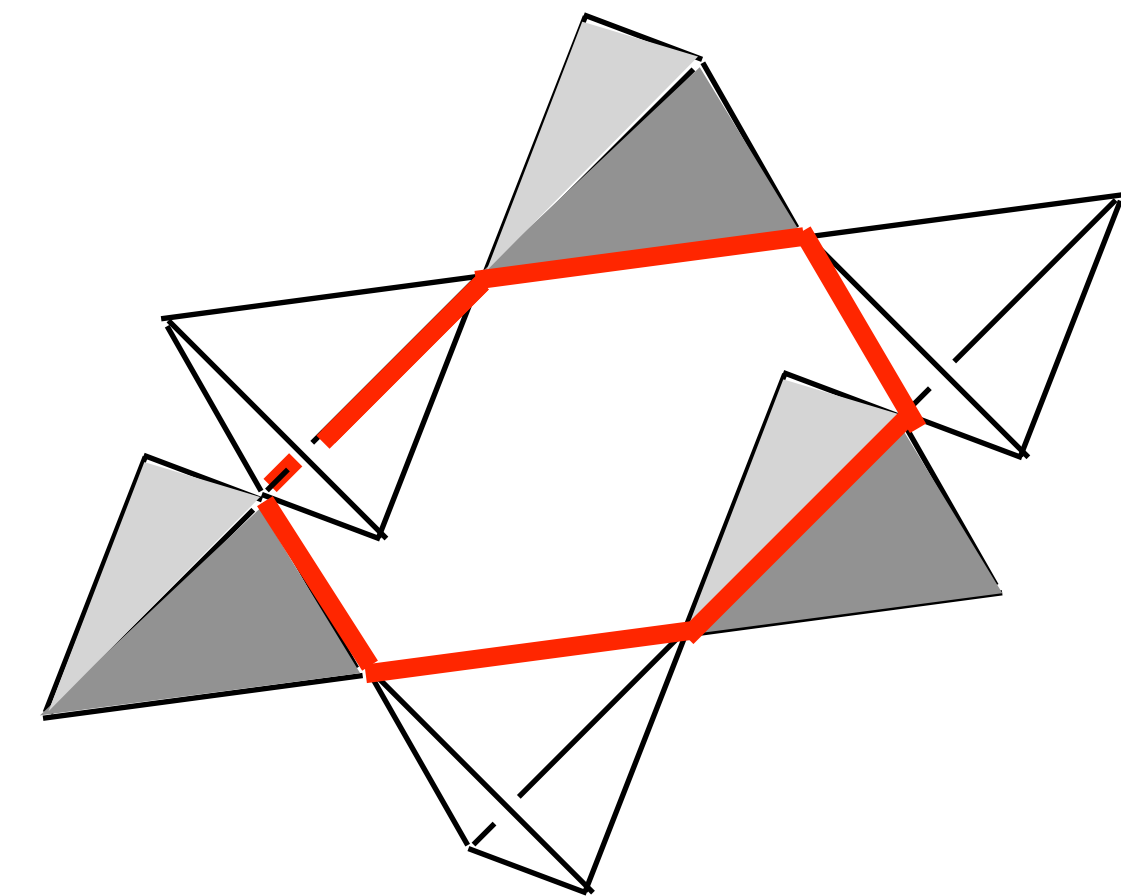
$J_z \gg J_{\perp}$ degenerate perturbation theory

$$\mathcal{H}_{eff} = -J_{ring} \sum_{\hexagon} (S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + h.c.)$$

$$J_{ring} = 12J_{\perp}^3 / J_z^2$$

Banerjee, Isakov, Damle, YBK '08

Benton, Sikora, Shannon '12



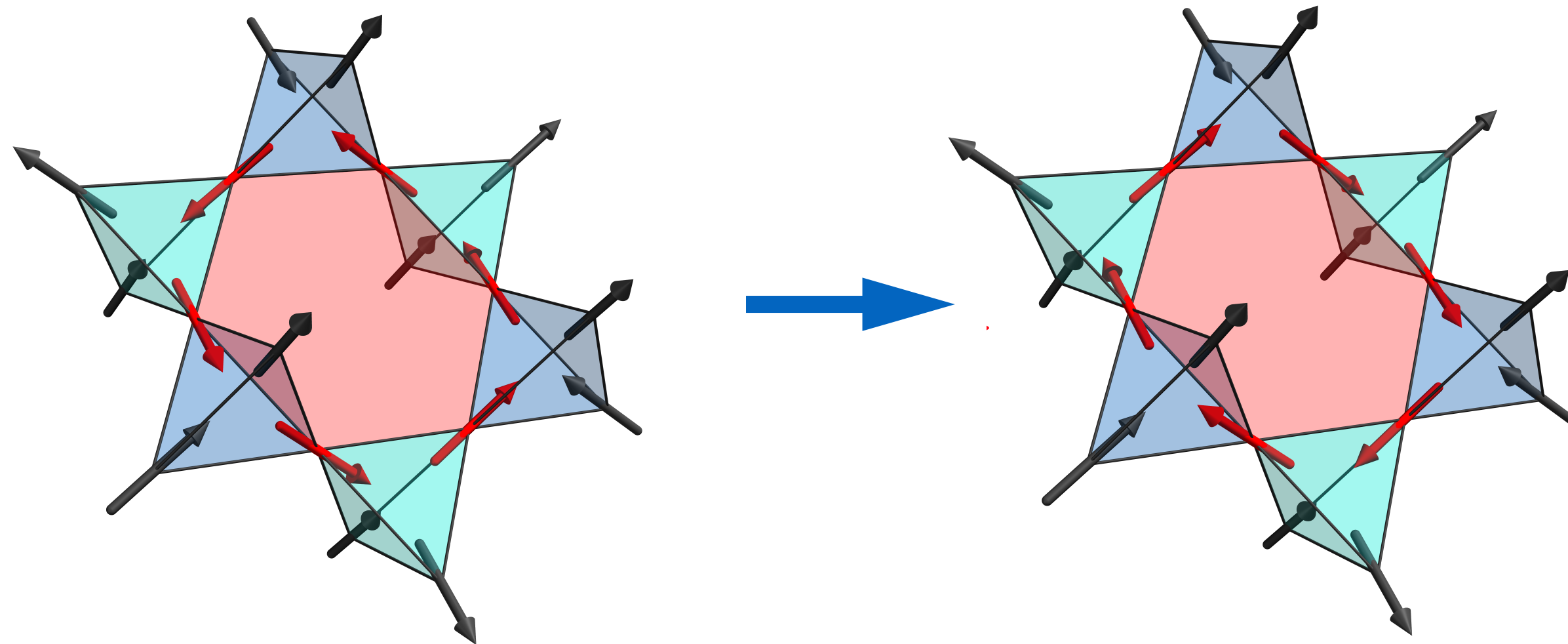
QMC

Connecting classically degenerate ground states

$$\mathcal{H}_{eff} = J_{ring} \sum_{\hexagon} (S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + h.c.)$$

$$K \sim -J_{ring} \propto -J_{\perp}^3 / J_{\parallel}^2$$

Hermele, Balents, Fisher '03



Quantum ground state = massive superposition of classically degenerate states
=> Quantum Spin Liquid

Quantum Electrodynamics

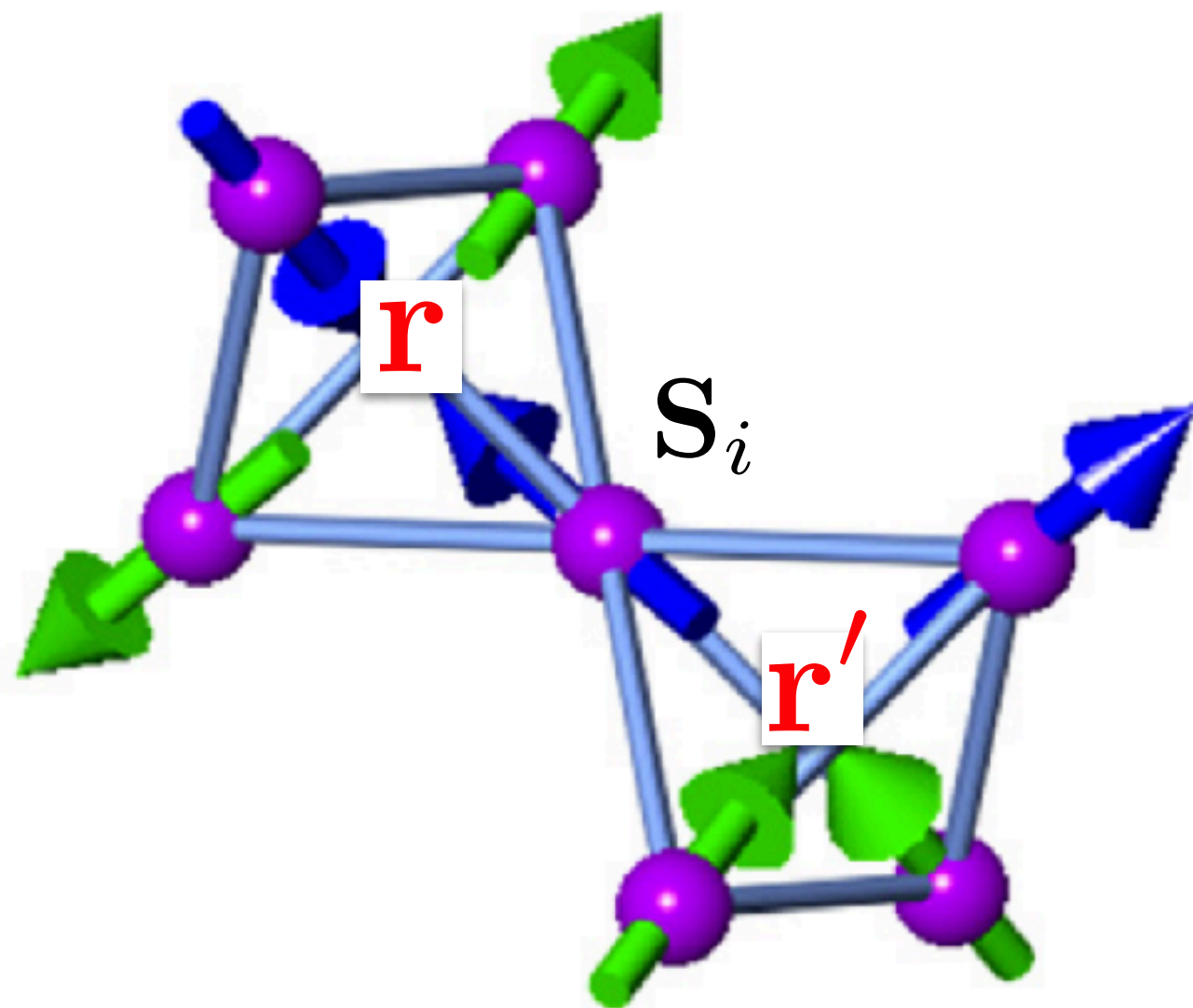
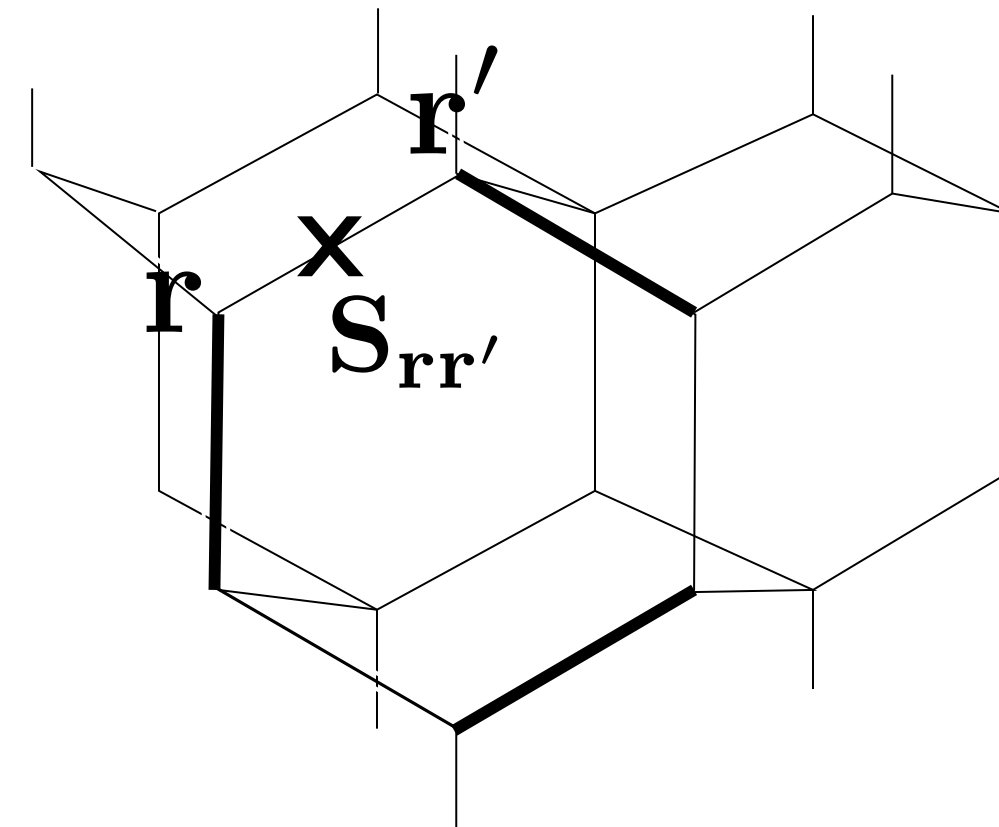
$$S_i^z = E_{rr'}$$

$$(\nabla \cdot E)_r = \sum_{r' \leftarrow r} E_{rr'} = \pm S_i^z$$

$$(\nabla \cdot E)_r = 0 \quad \text{Gauss's law}$$

$$S_i = S_{rr'}$$

link on the dual
diamond lattice sites



$$\nabla \cdot E = 0$$

Quantum Electrodynamics

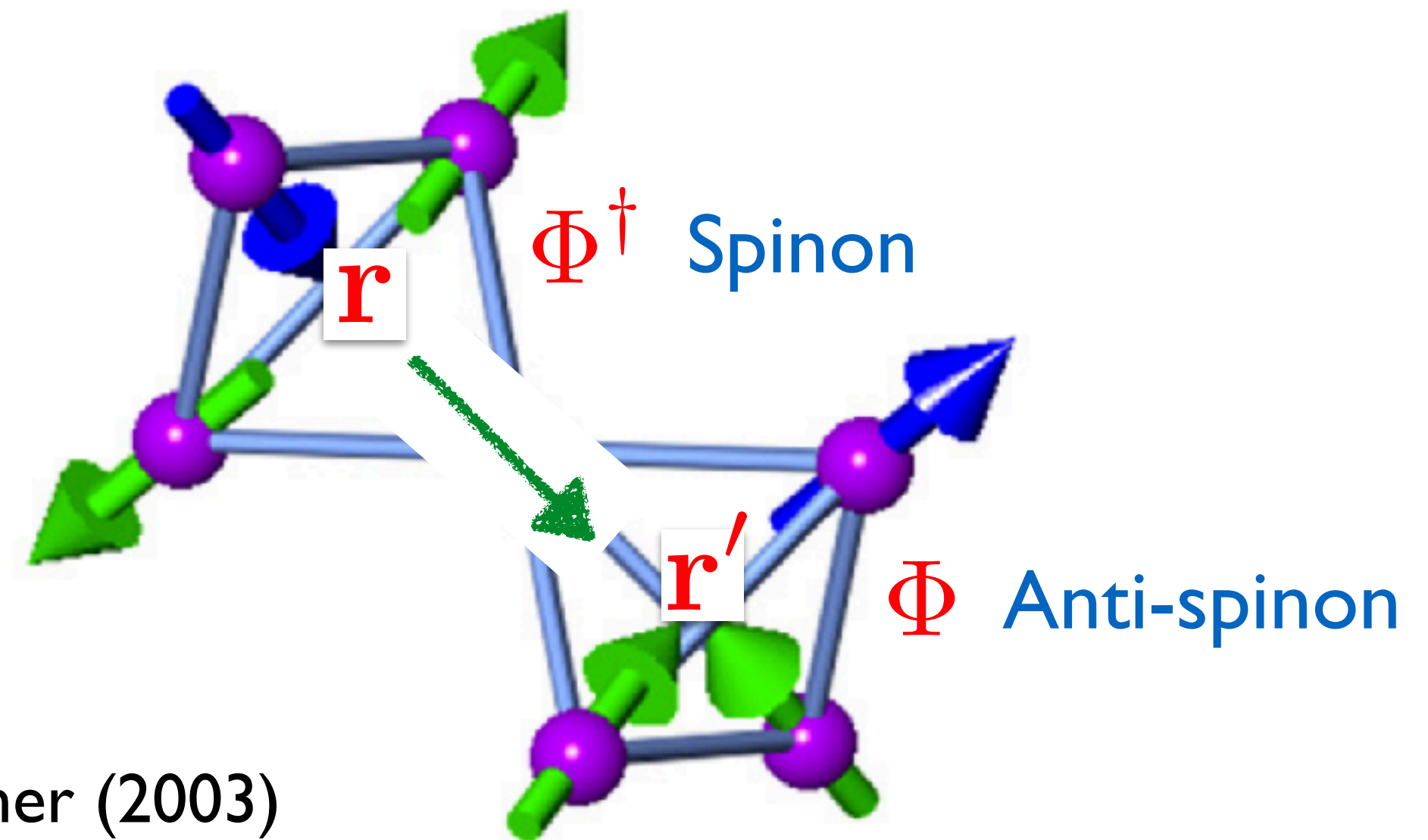
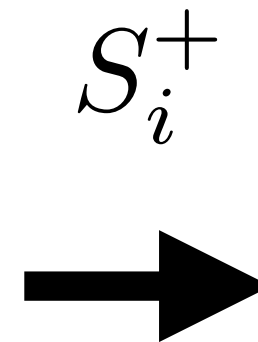
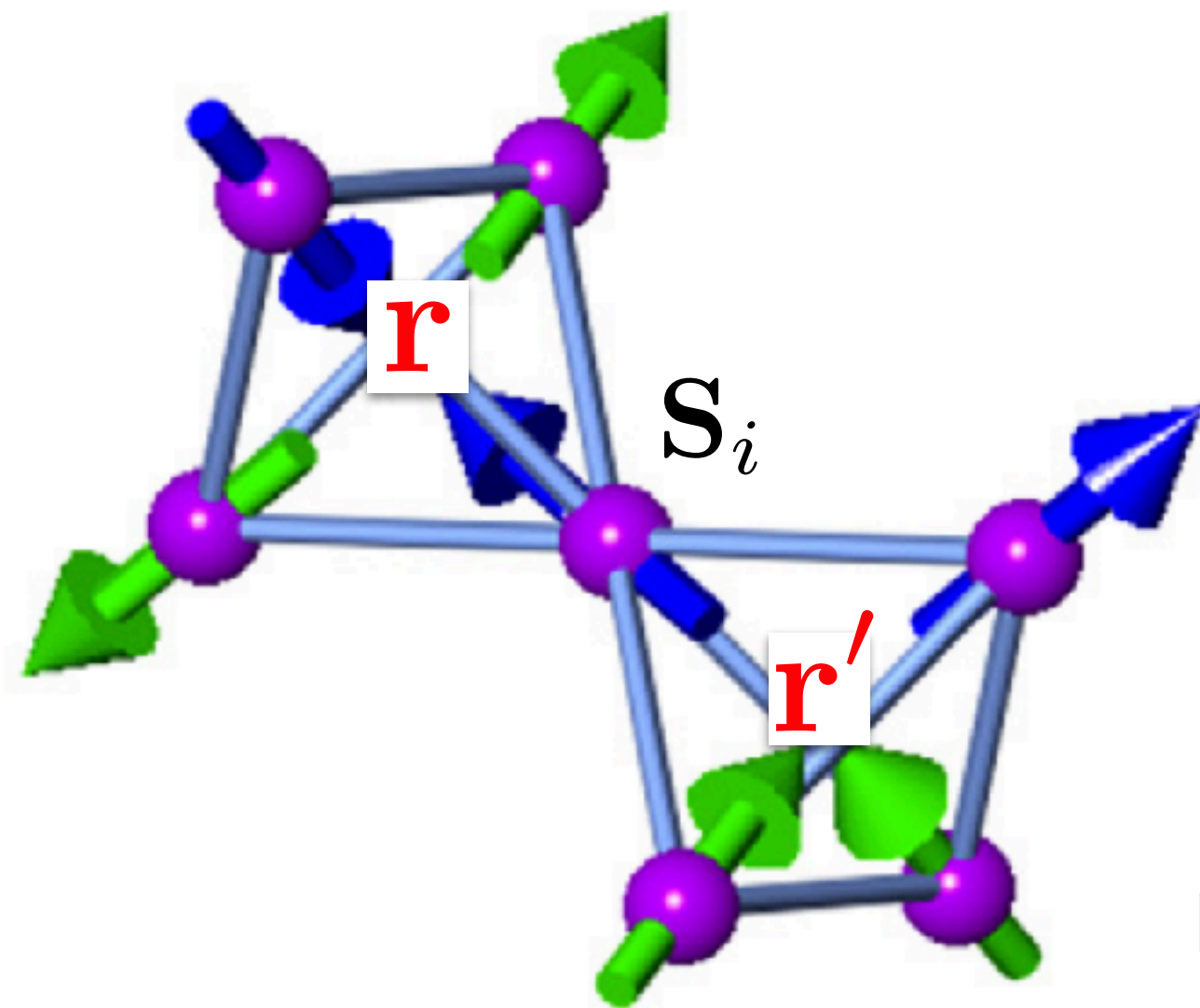
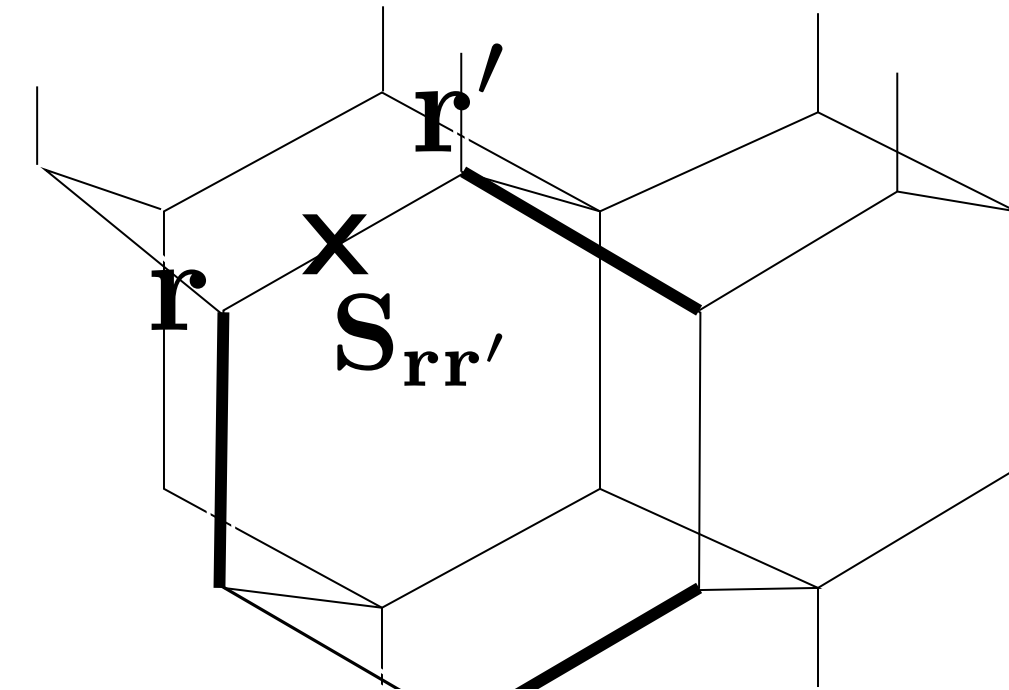
$$S_i^z = E_{rr'} \quad S_i^+ = \Phi_r^\dagger e^{iA_{rr'}} \Phi_{r'} \quad \pm \mathbf{r} \in A/B \quad [S^+, S^z] = -S^+ \quad \longrightarrow \quad [A_{rr'}, E_{rr'}] = i$$

$$(\nabla \cdot E)_r = \sum_{r' \leftarrow r} E_{rr'} = \pm S_r^z$$

$$(\nabla \cdot E)_r = 0 \quad \text{Gauss's law}$$

$$\mathbf{S}_i = \mathbf{S}_{rr'}$$

link on the dual
diamond lattice sites



$$\nabla \cdot E = 0$$

Hermele, Balents, Fisher (2003)

Savary + Balents (2012)

S Lee, Onoda, Balents(2012)

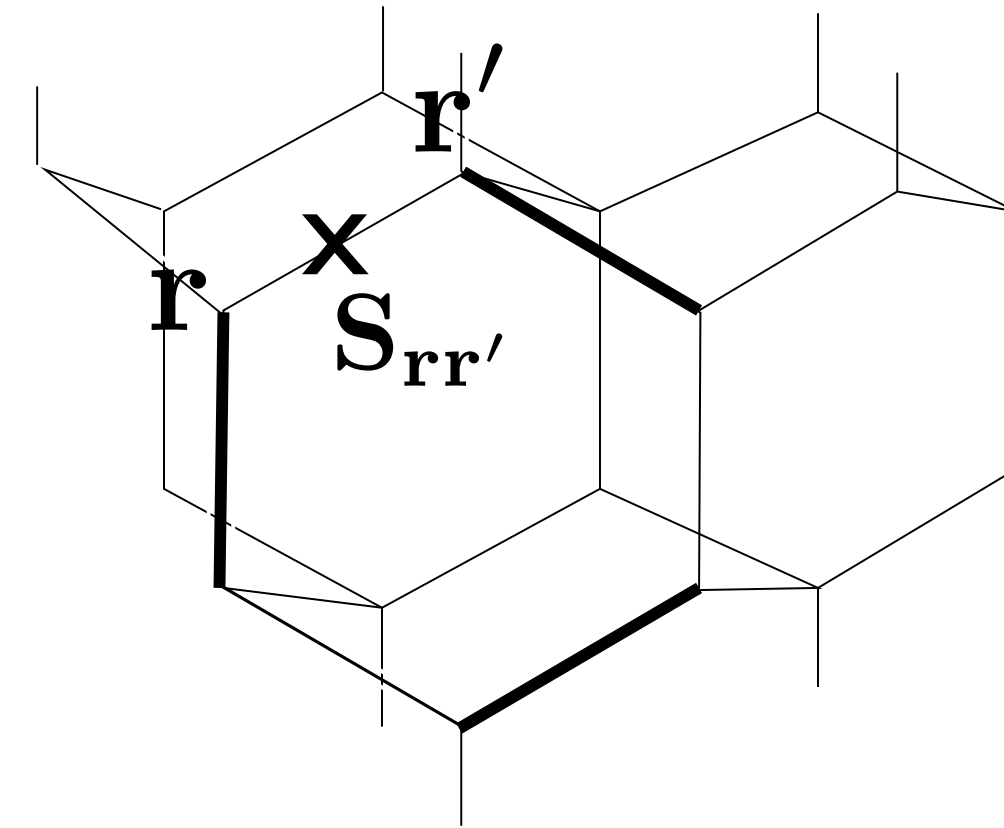
Quantum Electrodynamics

$$S_i^z = E_{rr'} \quad S_i^+ = \Phi_r^\dagger e^{iA_{rr'}} \Phi_{r'} \quad \pm \mathbf{r} \in A/B \quad [S^+, S^z] = -S^+ \quad \longrightarrow \quad [A_{rr'}, E_{rr'}] = i$$

$$(\nabla \cdot E)_{\mathbf{r}} = \sum_{\mathbf{r}' \leftarrow \mathbf{r}} E_{\mathbf{r}\mathbf{r}'} = \pm S_{\mathbf{r}}^z$$

$$(\nabla \cdot E)_{\mathbf{r}} = 0 \quad \text{Gauss's law}$$

$\mathbf{S}_i = \mathbf{S}_{\mathbf{r}\mathbf{r}'}$
link on the dual
diamond lattice sites



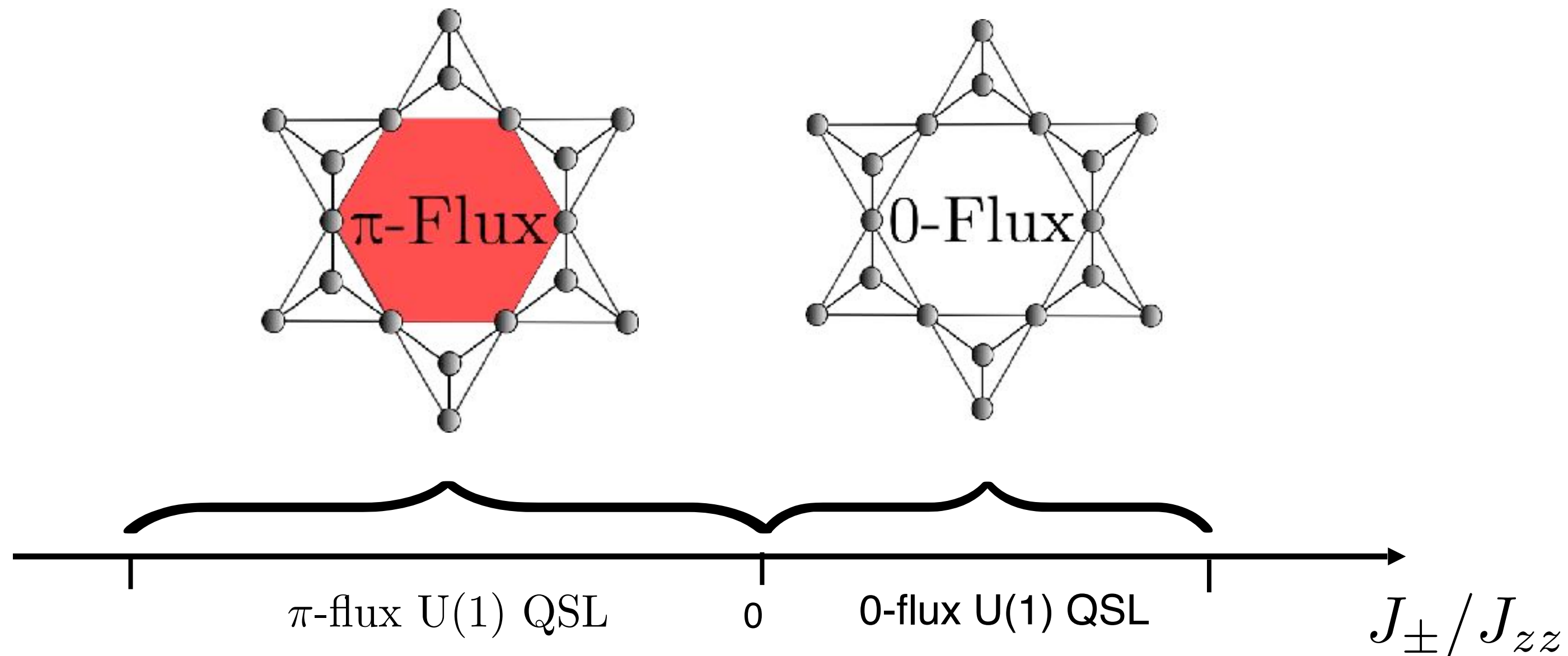
$$\mathcal{H}_{eff} = -J_{ring} \sum_{\hexagon} (S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + h.c.) \longrightarrow \sum_{\hexagon} 2 \cos(\nabla \times A)_{\hexagon}$$

$$\mathcal{H} = \frac{U}{2} \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} \left(E_{\mathbf{r}\mathbf{r}'}^2 - \frac{1}{4} \right) - K \sum_{\hexagon} \cos(\nabla \times A)_{\hexagon}$$

large U
 $K \sim J_{ring} \quad K \propto \frac{J_{\pm}^3}{J_{zz}^2}$

0-flux and π -flux Quantum Spin Ice

$$\mathcal{H} = \frac{U}{2} \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} \left(E_{\mathbf{r}\mathbf{r}'}^2 - \frac{1}{4} \right) - K \sum_{\hexagon} \cos(\nabla \times \mathbf{A})_{\hexagon} \quad K \propto \frac{J_{\pm}^3}{J_{zz}^2}$$



M. Hermele, et al. PRB (2004)

Excitations in the deconfined phase: U(1) quantum spin liquid (Quantum Spin Ice)

$$\mathcal{H} = \frac{U}{2} \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} \left(E_{\mathbf{r}\mathbf{r}'}^2 - \frac{1}{4} \right) + \frac{K}{2} \sum_{\square} [(\nabla \times \mathbf{A})_{\square}]^2$$

electric monopoles (spinons)

$$2\Delta_{\text{spinon}} \sim J_z$$

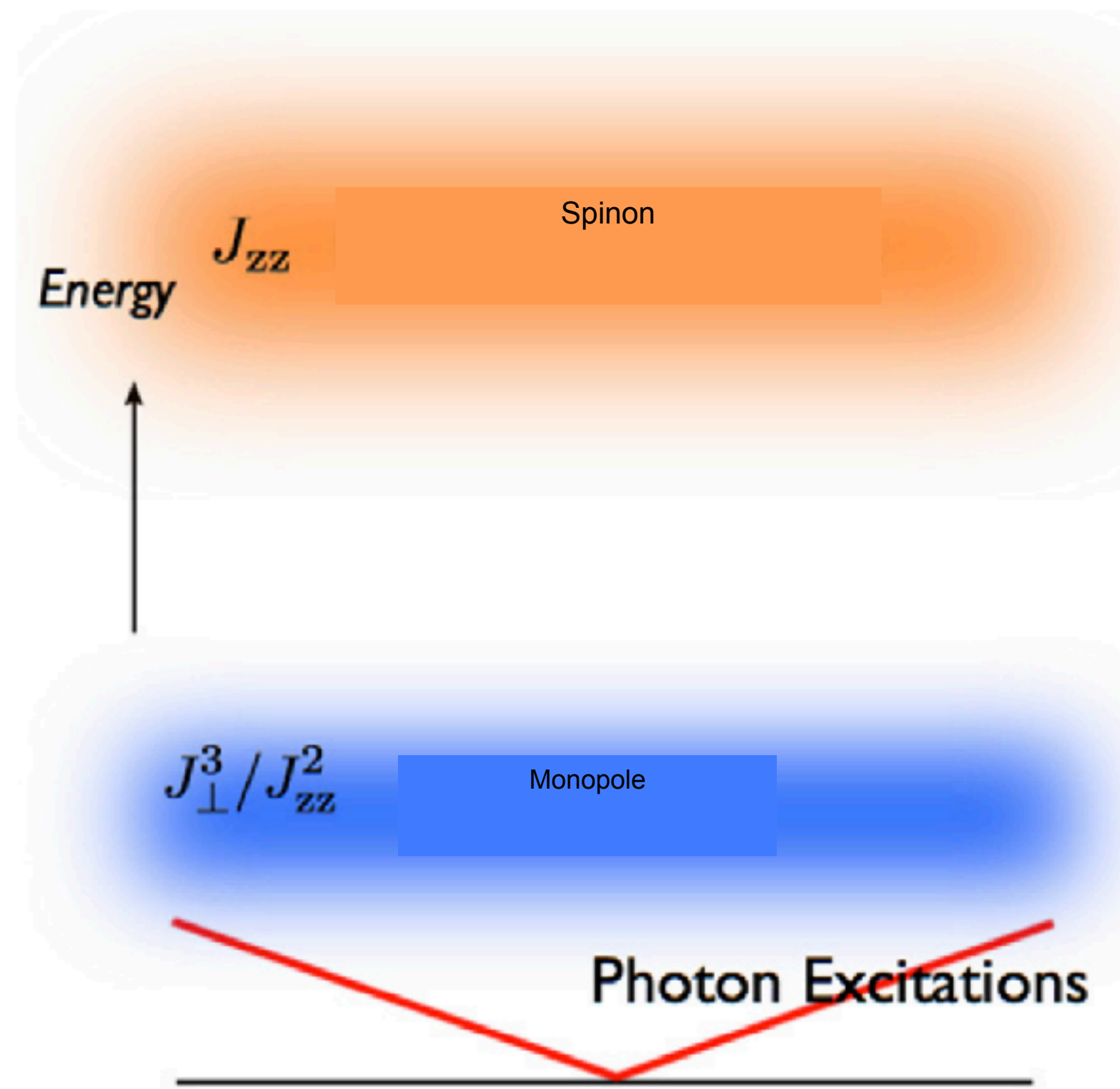
magnetic monopoles (visons)

$$\Delta_{\text{mon}} \sim J_{\pm}^3 / J_z^2$$

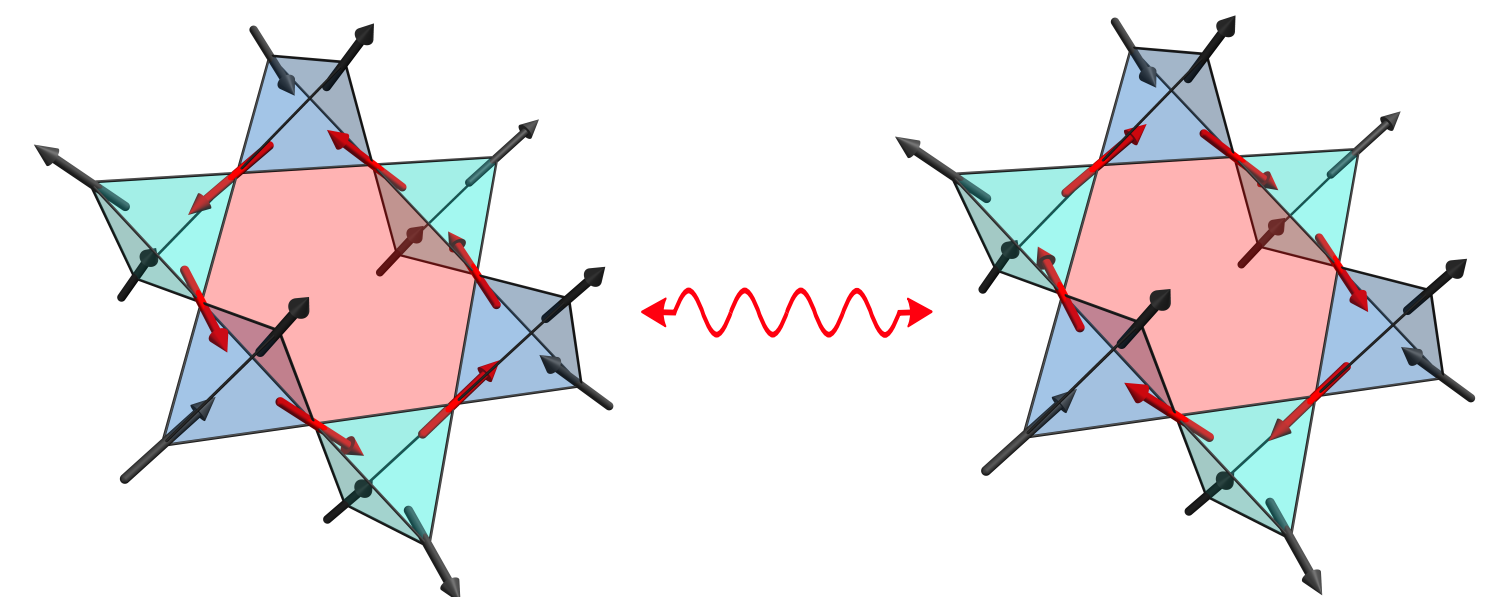
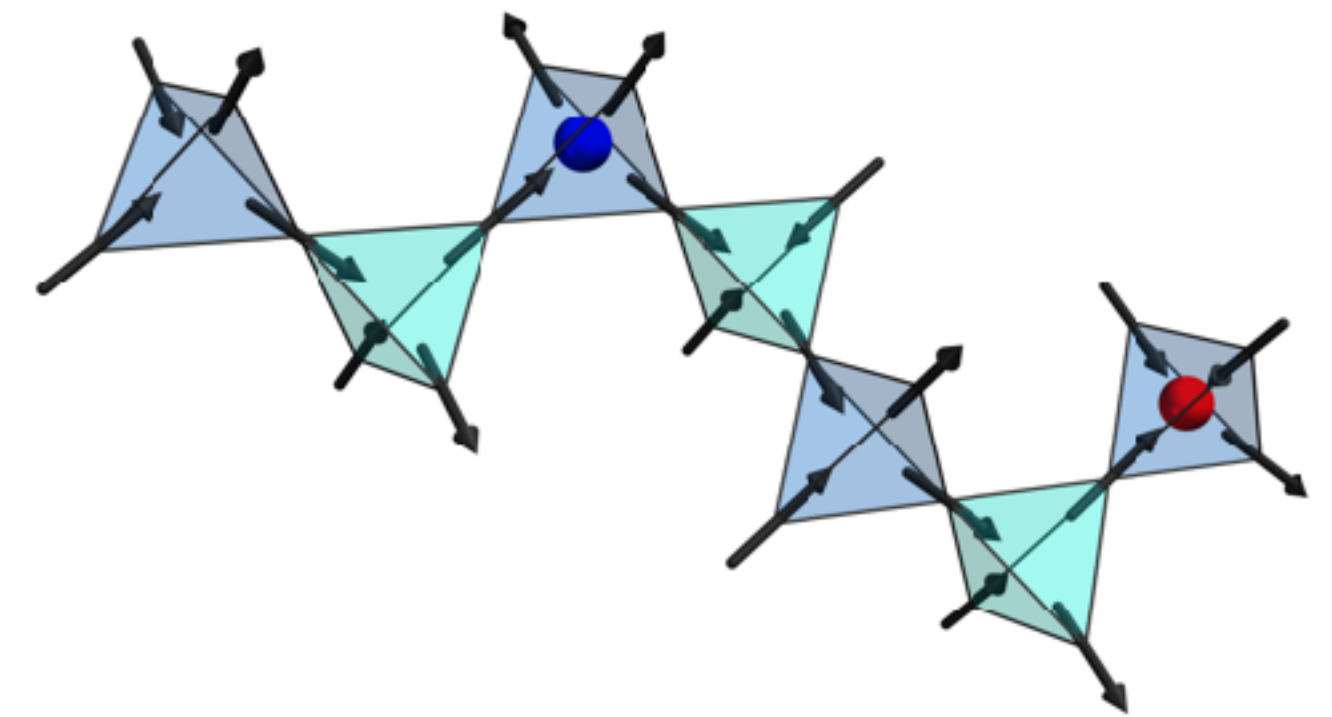
emergent photons

$$\omega(\mathbf{k}) \approx c|\mathbf{k}| \quad c \propto \sqrt{UK}a_0/\hbar$$

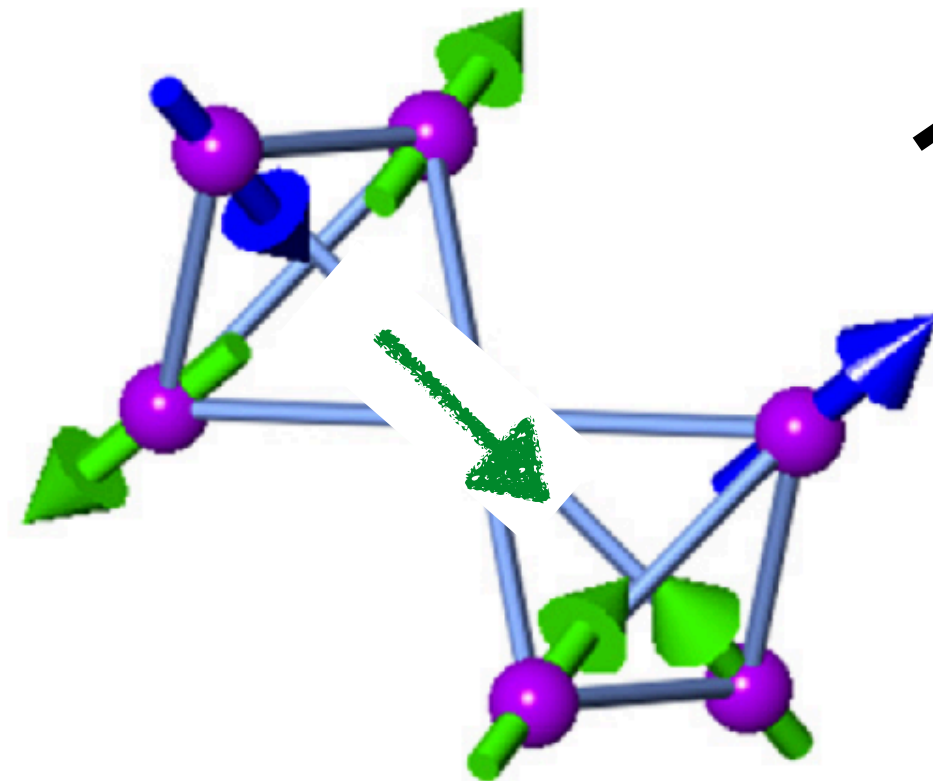
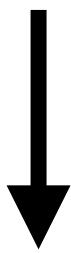
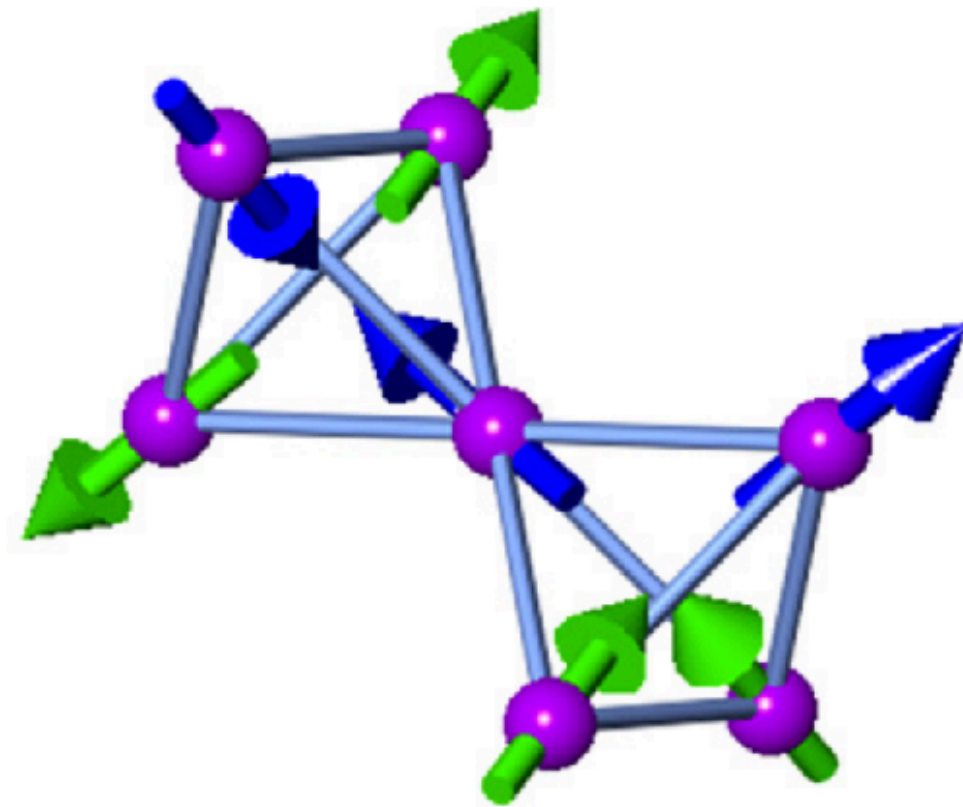
$$C(T) \propto \frac{1}{c^3} T^3$$



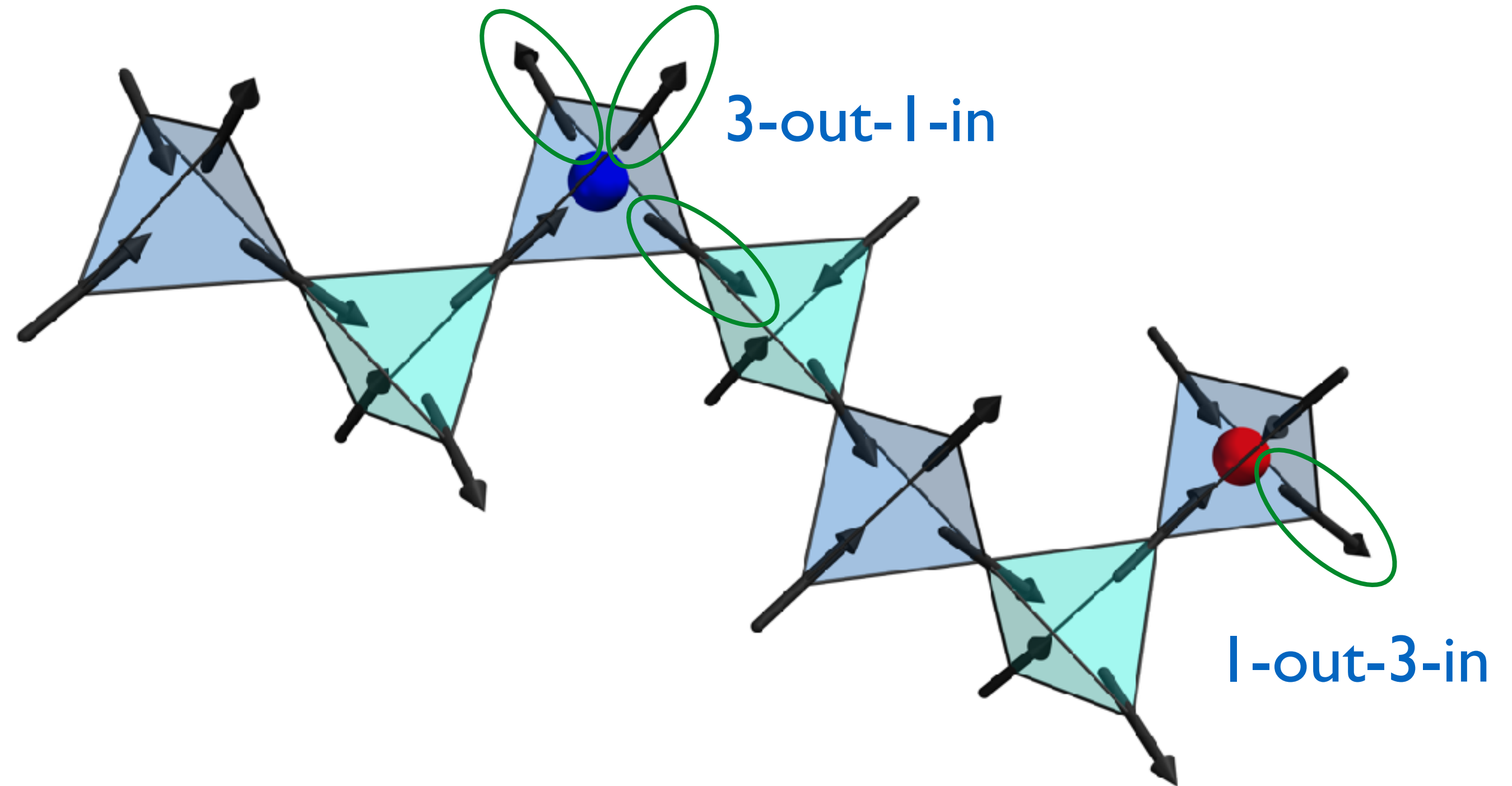
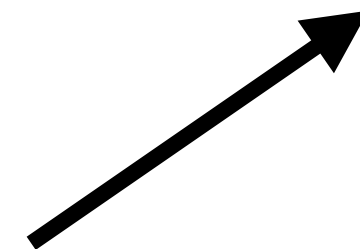
Gingras & McClarty (2014)



Spinons



$S=1$



“Two” $S=1/2$ excitations

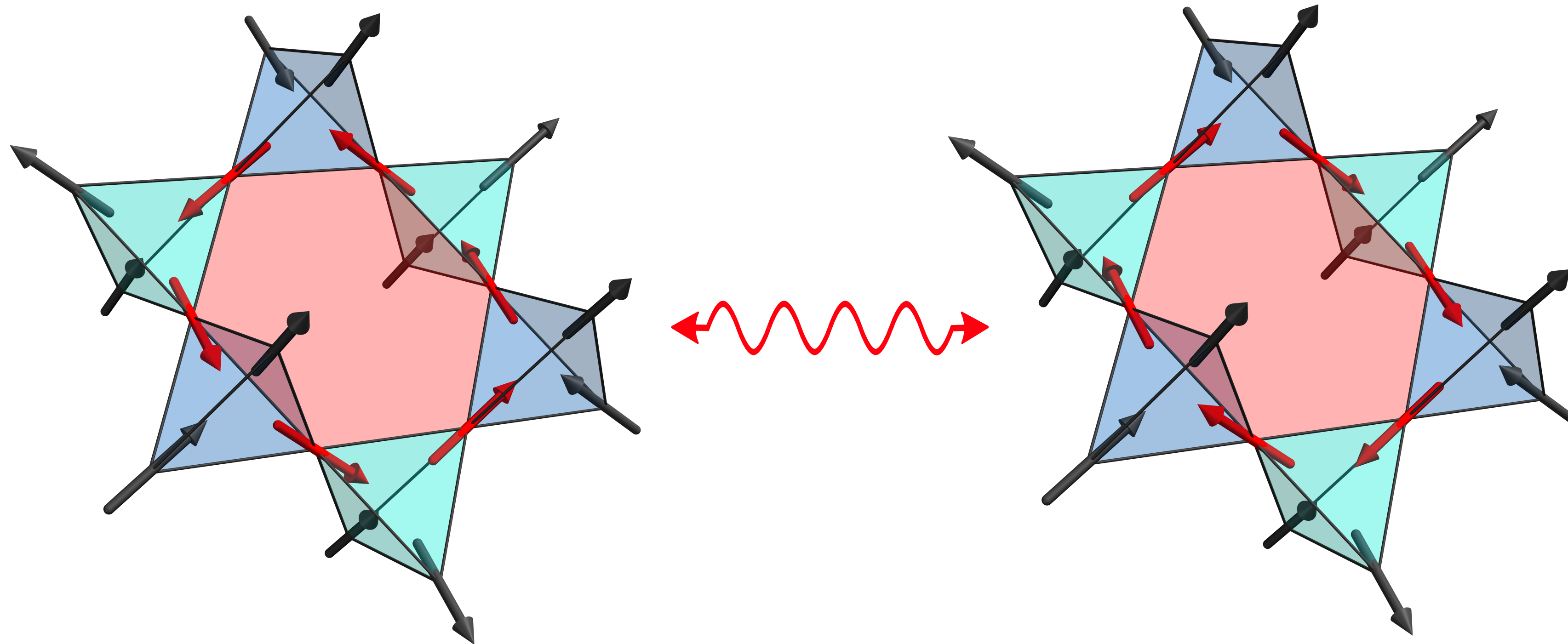
Spinon-Antispinon pair

Emergent Photons

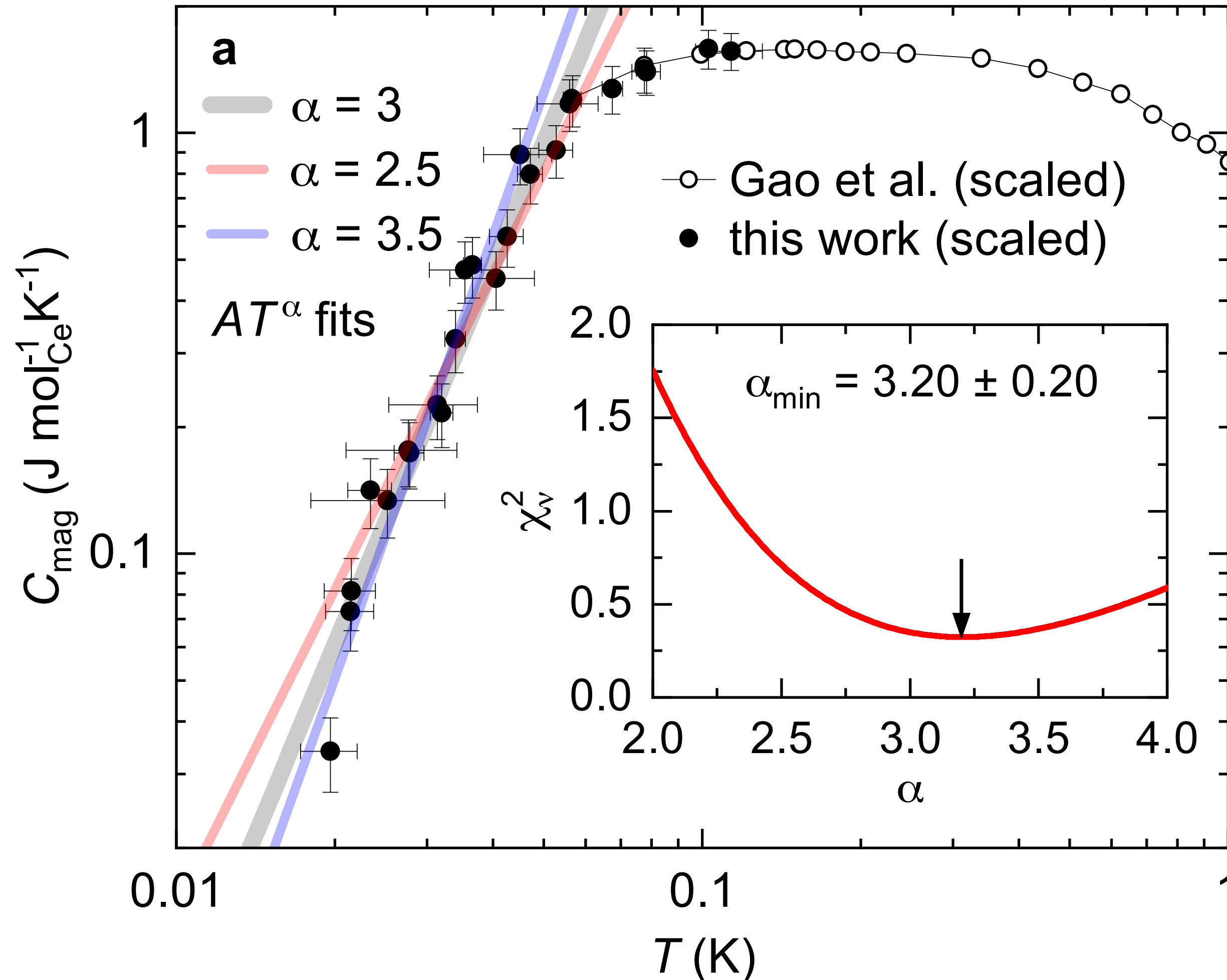
$$\mathcal{H}_{eff} = J_{ring} \sum_{\hexagon} (S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + h.c.) \longrightarrow -K \cos(\nabla \times A)$$

$$K \sim -J_{ring} \propto -J_{\perp}^3 / J_{\parallel}^2$$

Hermele, Balents, Fisher '03



Specific heat data at low temperatures



T^3 from emergent photons ?

Photon velocity comparable
to the estimation from
neutron scattering

$$c_{QSI} = 7.9 \pm 0.4 \text{ m/s}$$

B.Gao, F.Desrochers, ... S.Paschen, YBK, P.Dai.
arXiv:2404.04207, Nature Physics (2025)

Different routes to QSL: candidate materials

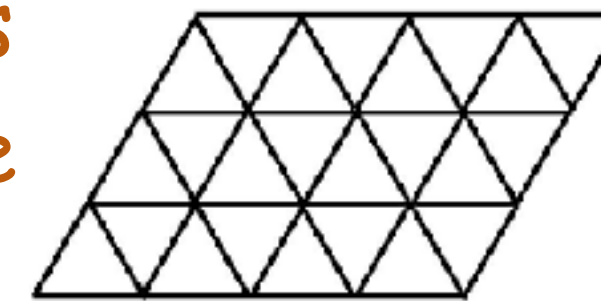
Candidate Materials (incomplete list)



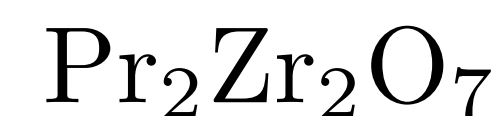
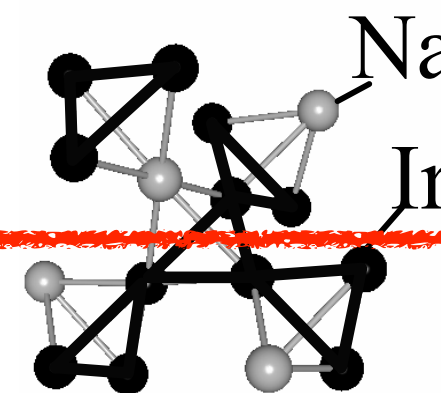
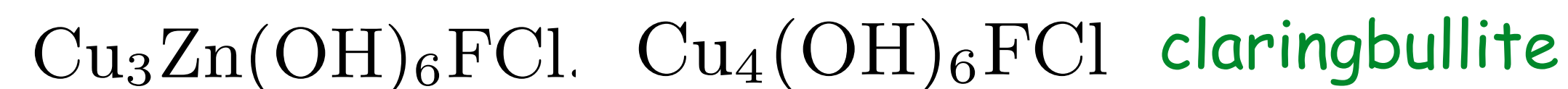
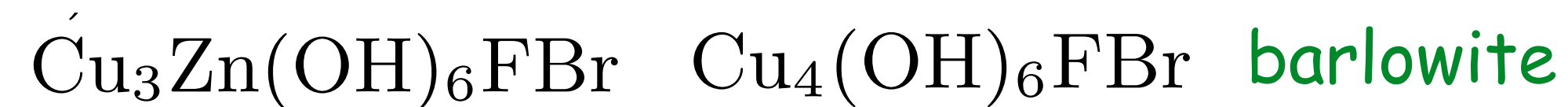
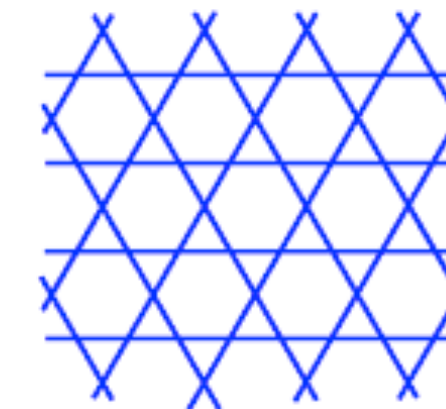
Geometric



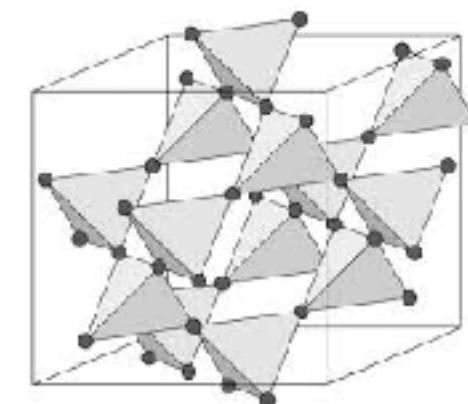
Organic Materials
Triangular Lattice



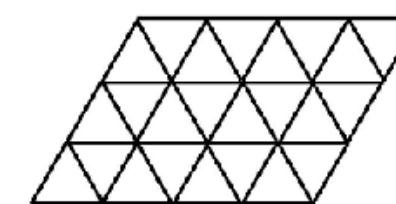
Kagome



pyrochlore



triangular



Kitaev Materials

honeycomb

hyper-honeycomb

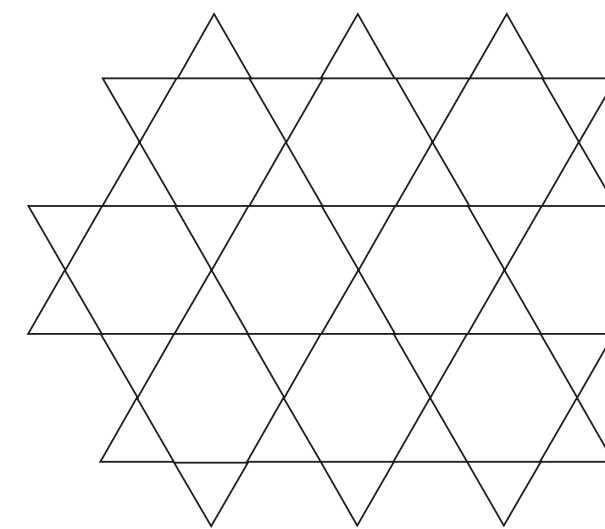
Spin-orbit

Different Routs to Quantum Spin Liquid

1. Geometric Frustration

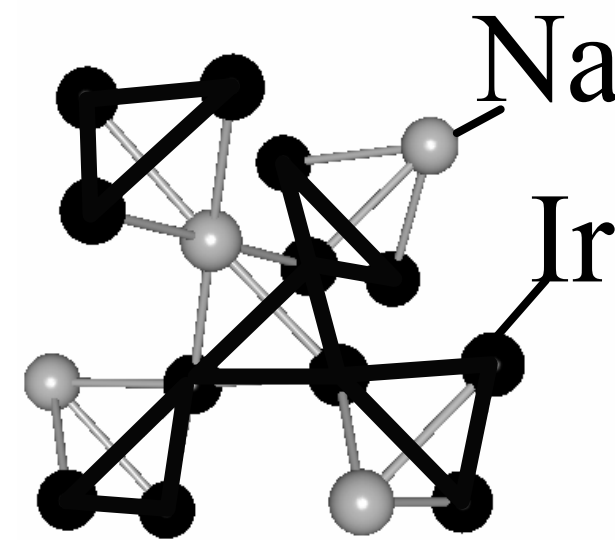
Herbertsmithite $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$

"Ideal" Kagome lattice



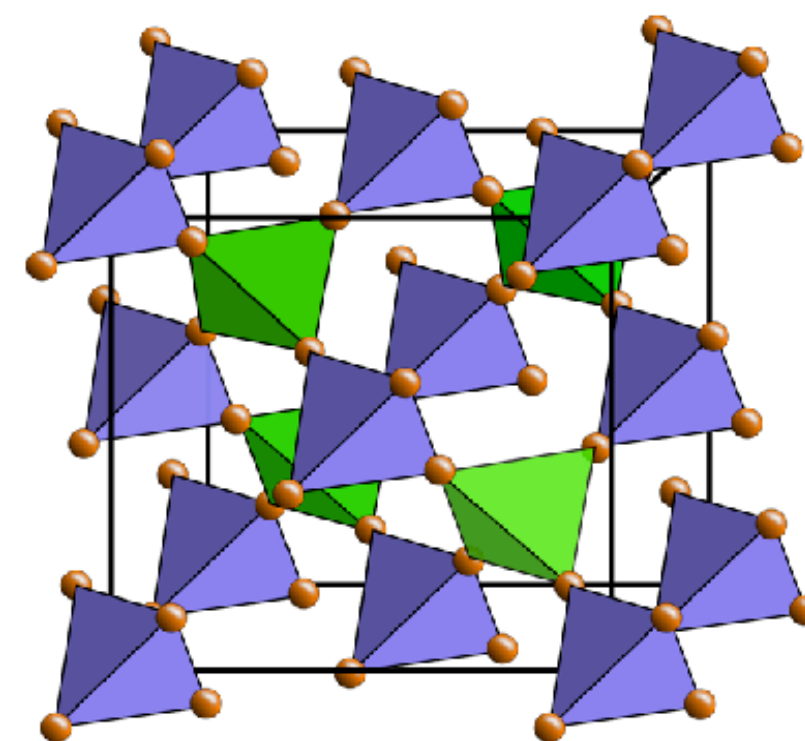
D. G. Nocera, Y. S. Lee
P. Mendels

Hyperkagome $\text{Na}_4\text{Ir}_3\text{O}_8$



H. Takagi (2007)
Y. Singh, Y. Tokiwa, P. Gegenwart
(2013)

Pyrochlore systems
Quantum spin ice $\text{Ce}_2\text{Zr}_2\text{O}_7$



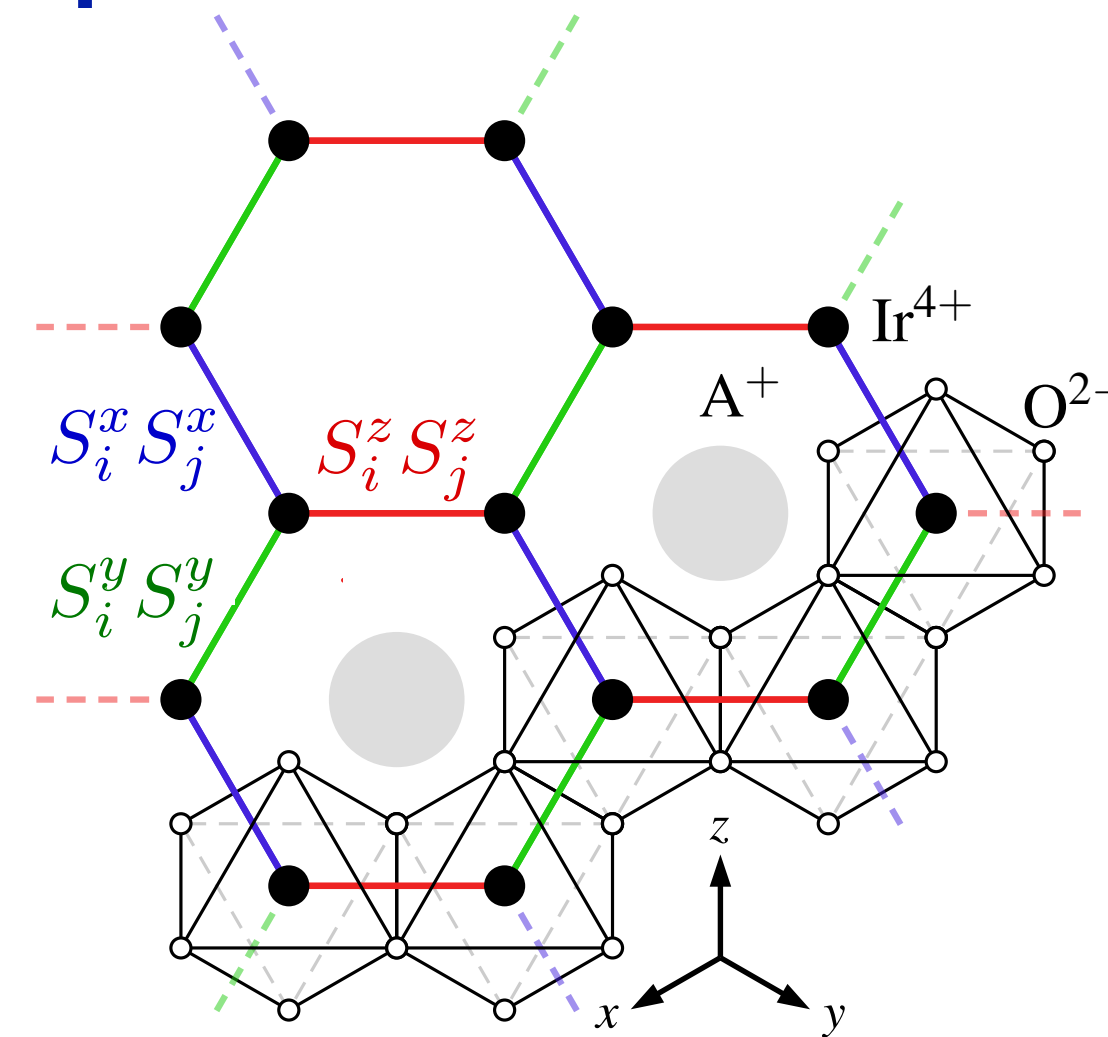
Pengcheng Dai, Bruce Gaulin, Romain
Sibille, Elsa Lhotel, Sylvain Petit ...

Different Routs to Quantum Spin Liquid

2. Frustrating Interactions

Kitaev Materials
Bond-dependent
interaction

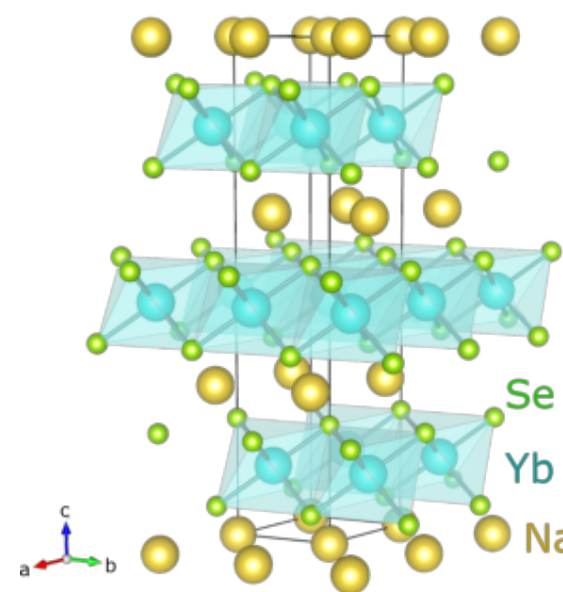
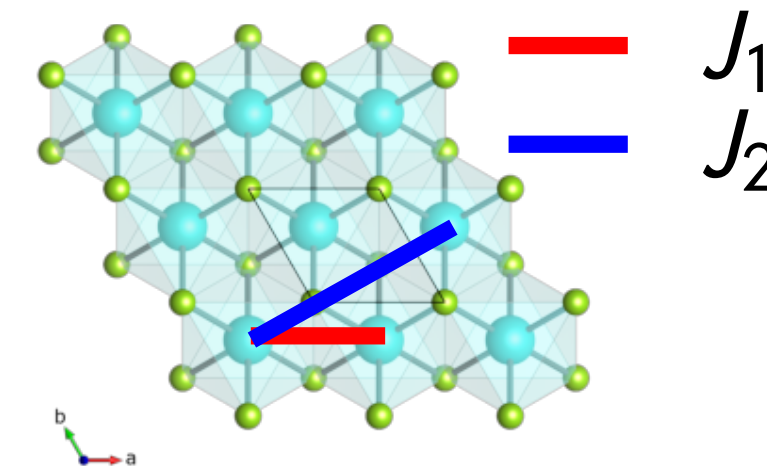
α - Na_2IrO_3
 α - RuCl_3



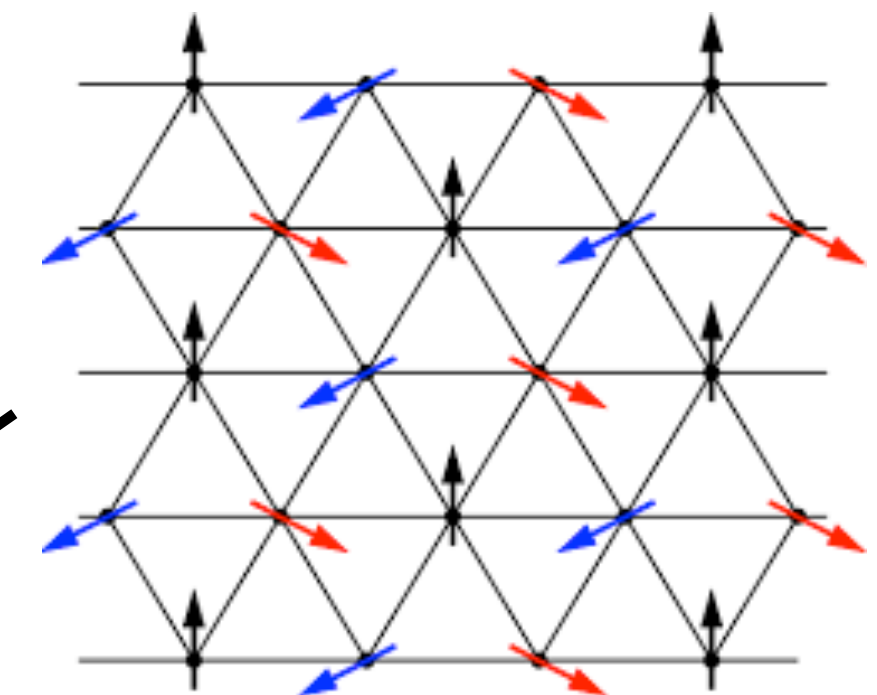
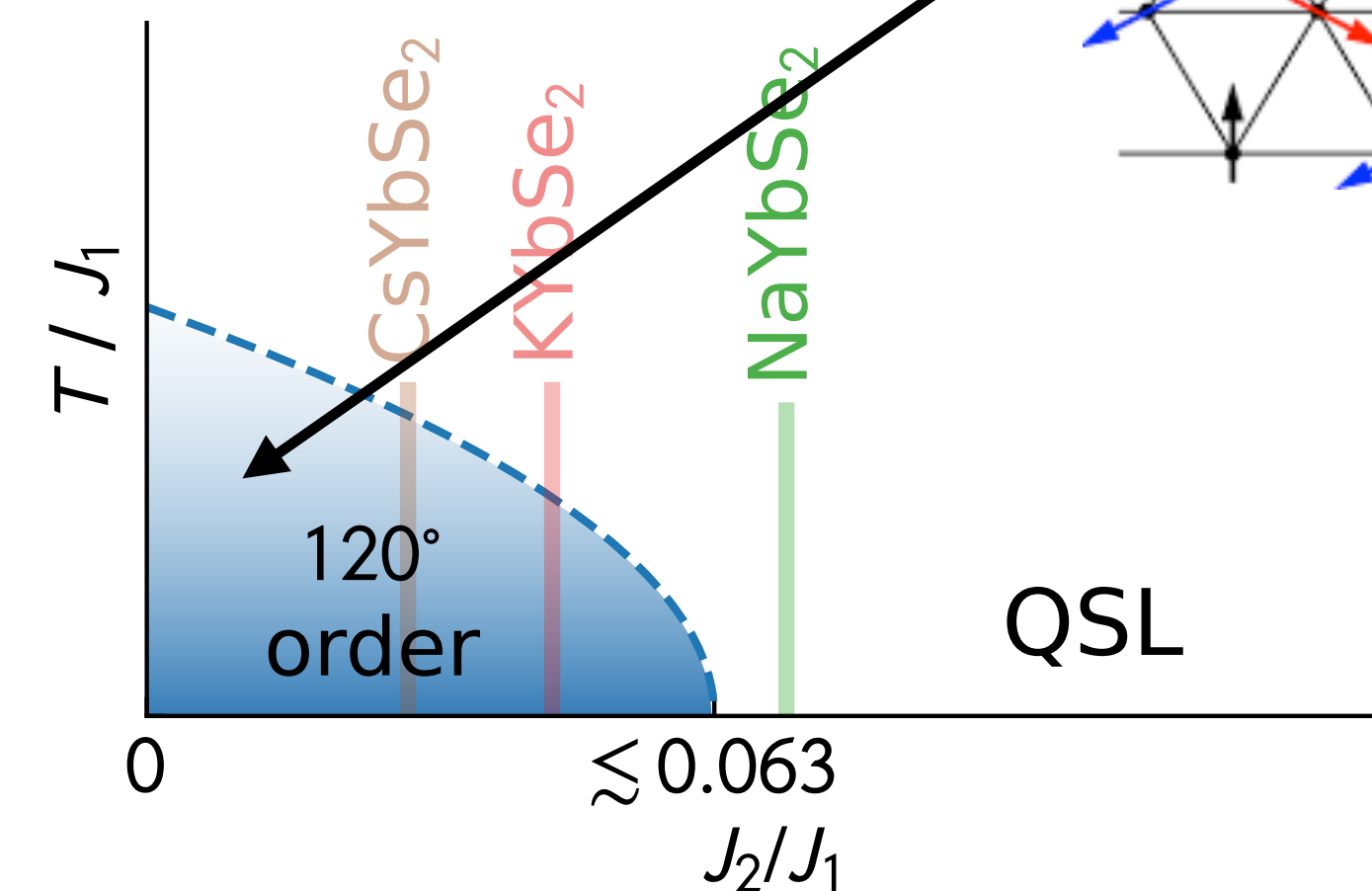
H. Takagi, R. Coldea,
P. Gegenwart,
YJ Kim, S. Nagler, ...

Longer range interactions
 J_1 - J_2 model

NaYbSe_2

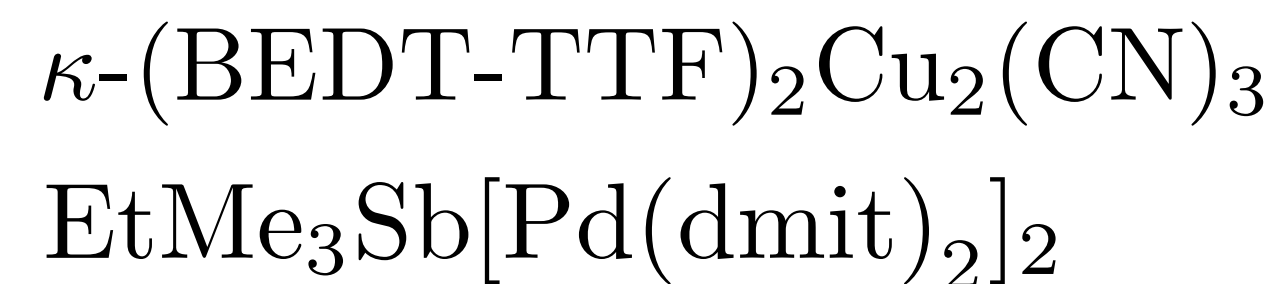


NaYbSe_2



Different Routs to Quantum Spin Liquid

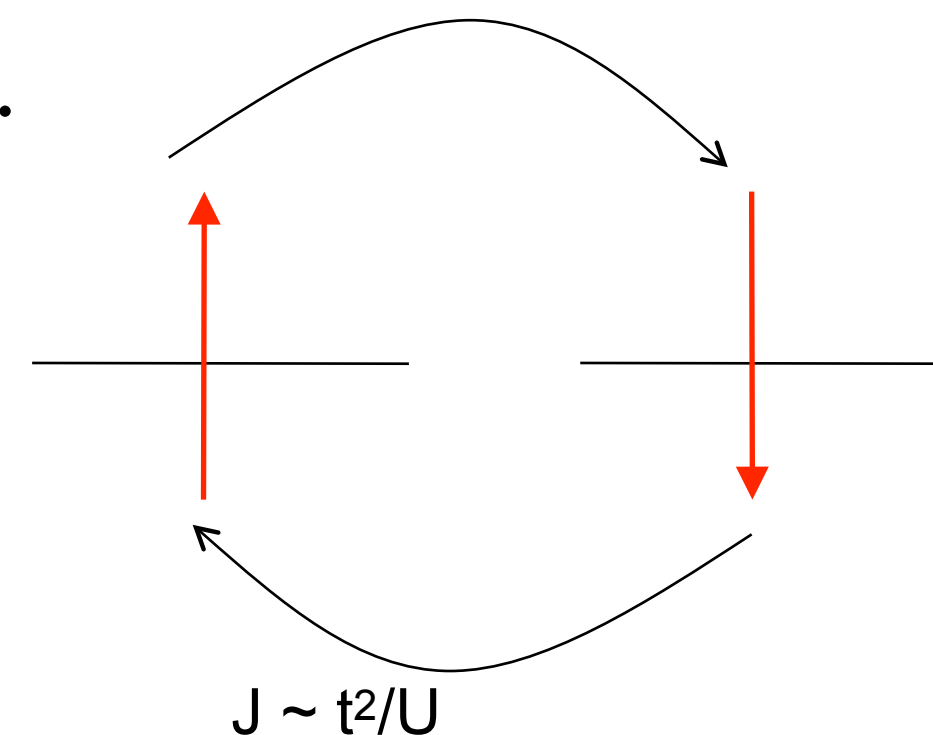
3. Weak Mott Insulator (charge fluctuations)



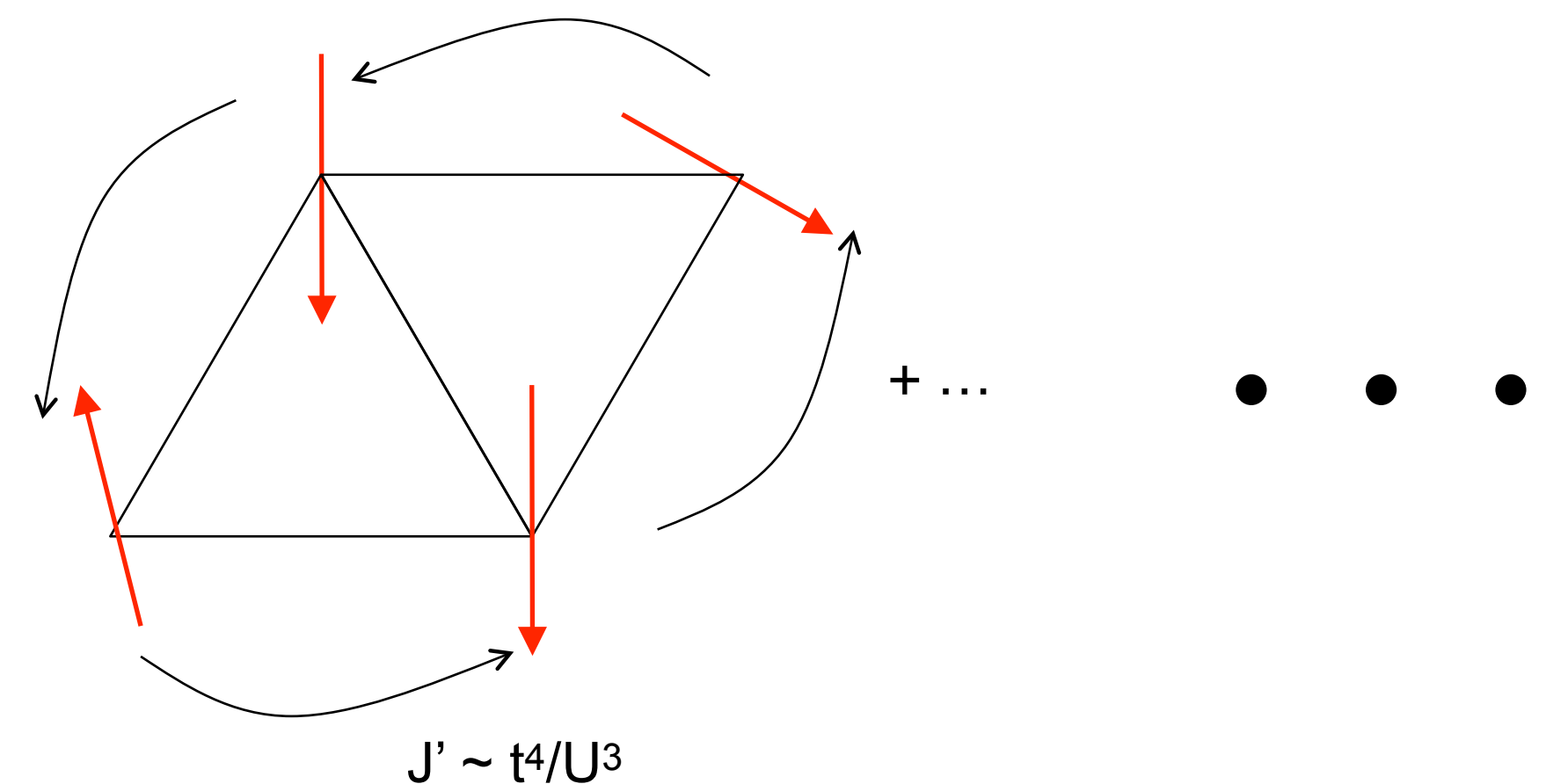
K. Kanoda
R. Kato

M. Yamashita,
Y. Matsuda,

$$H \sim H_{\text{heisenberg}} + H_{\text{ring}} + \dots$$



+

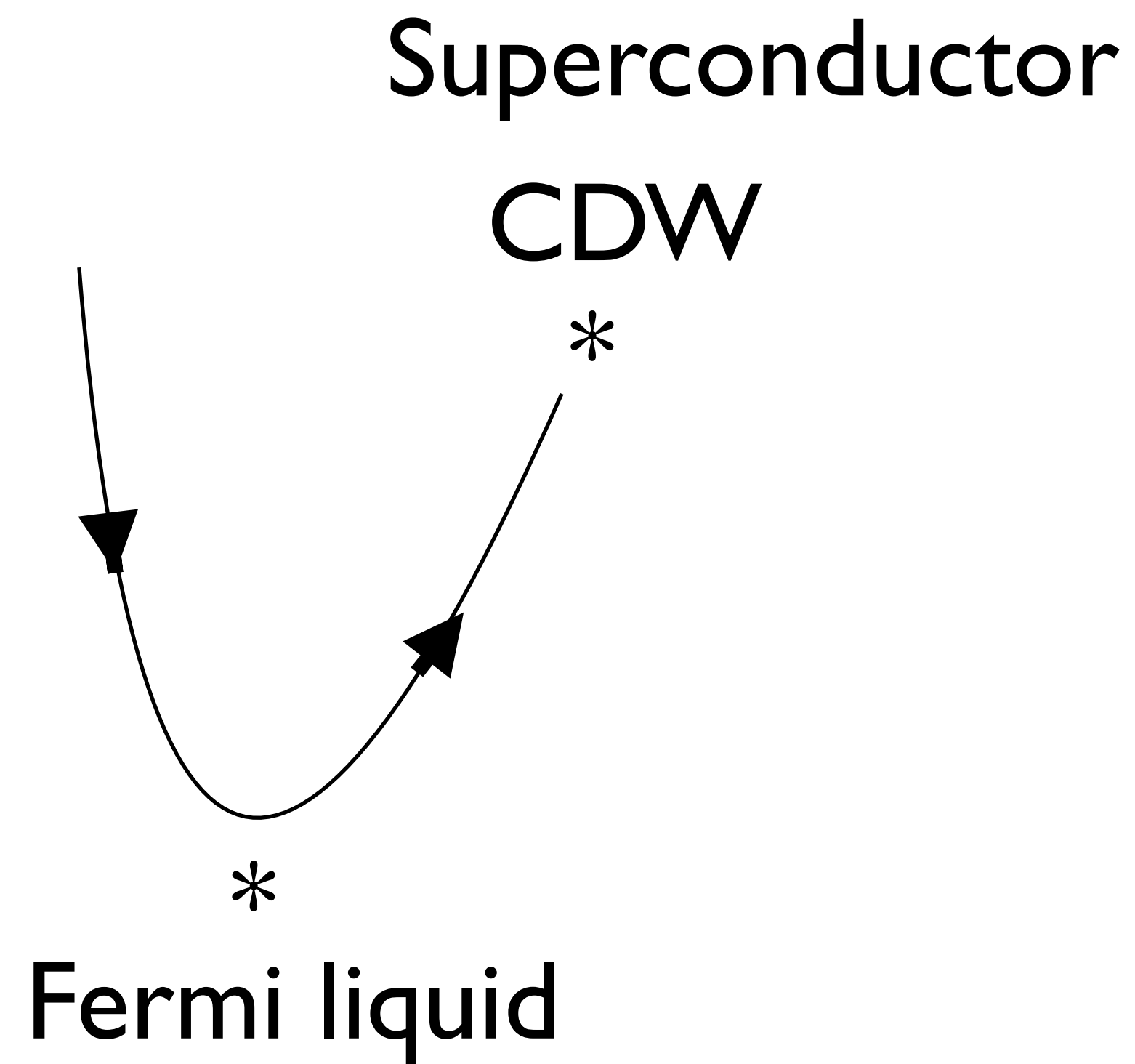


Organic Material
Triangular Lattice

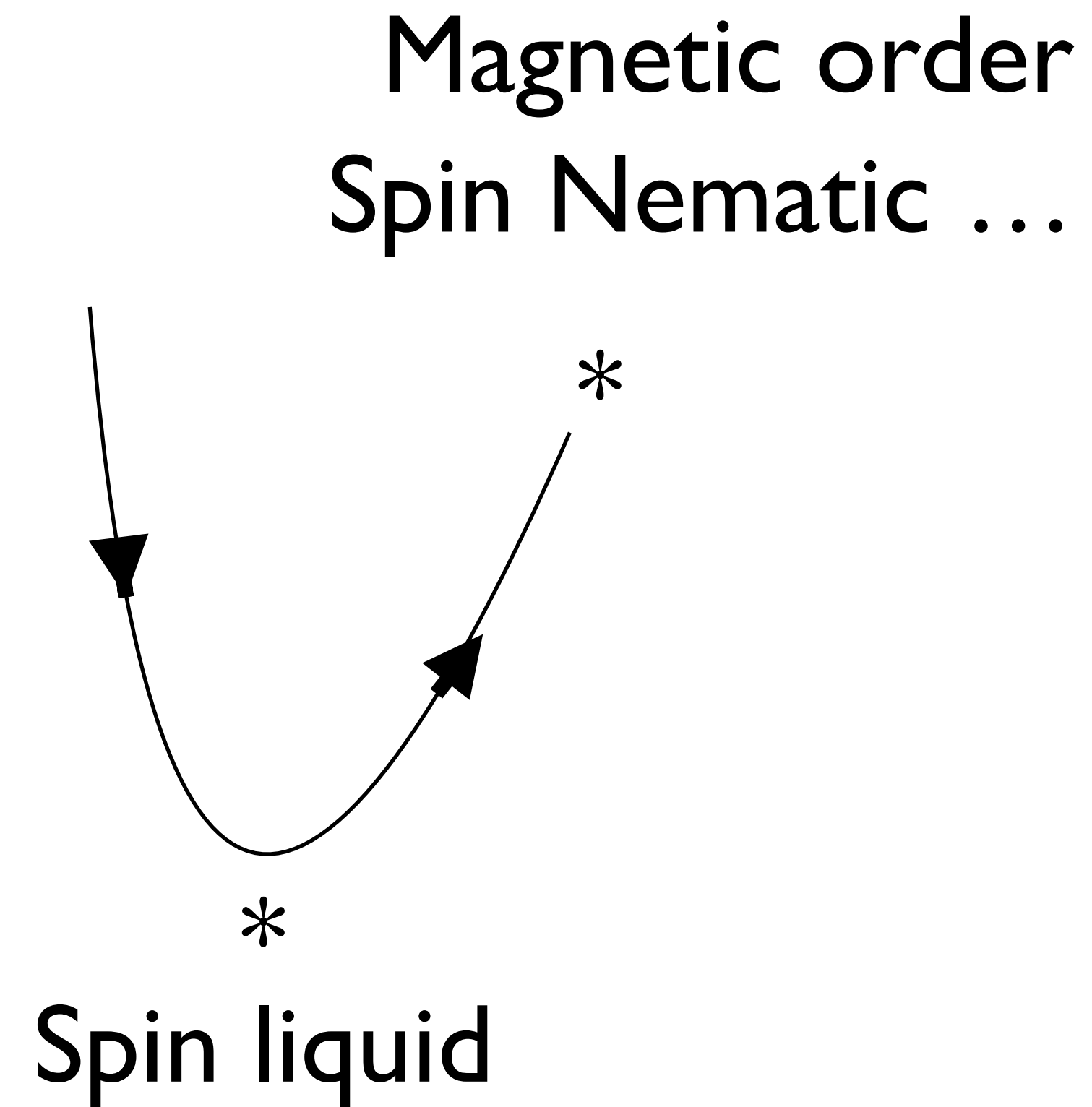


Charge fluctuations are important
near the Mott transition even in the
insulating phase

Fermi liquid behavior



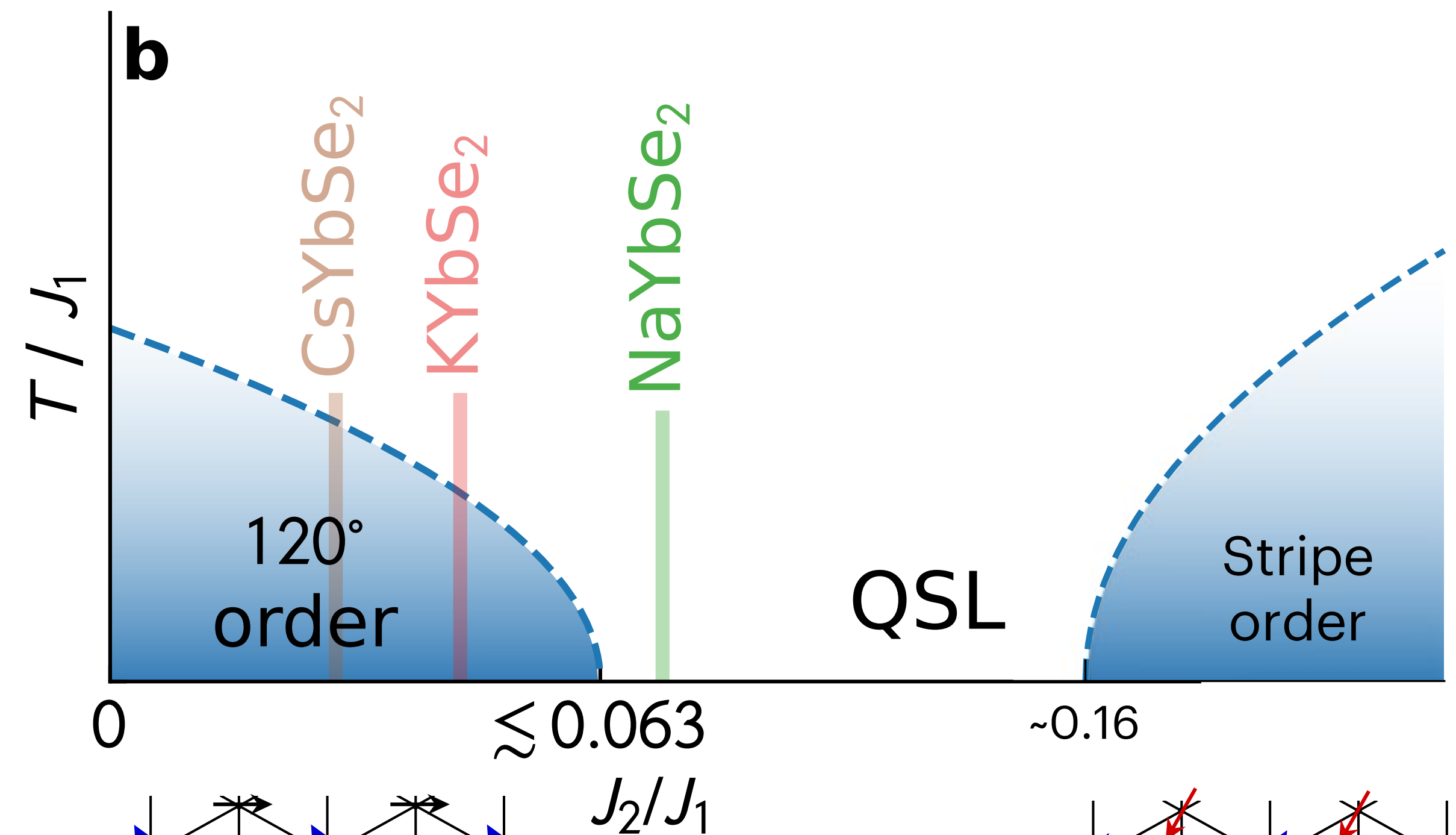
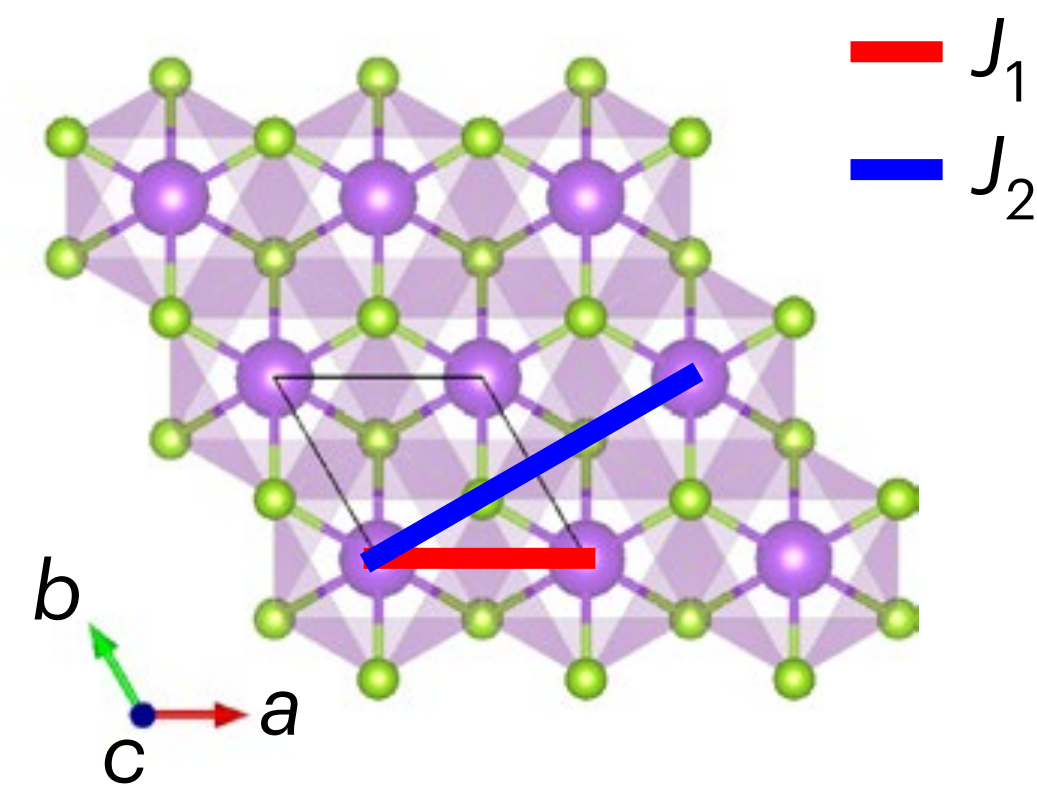
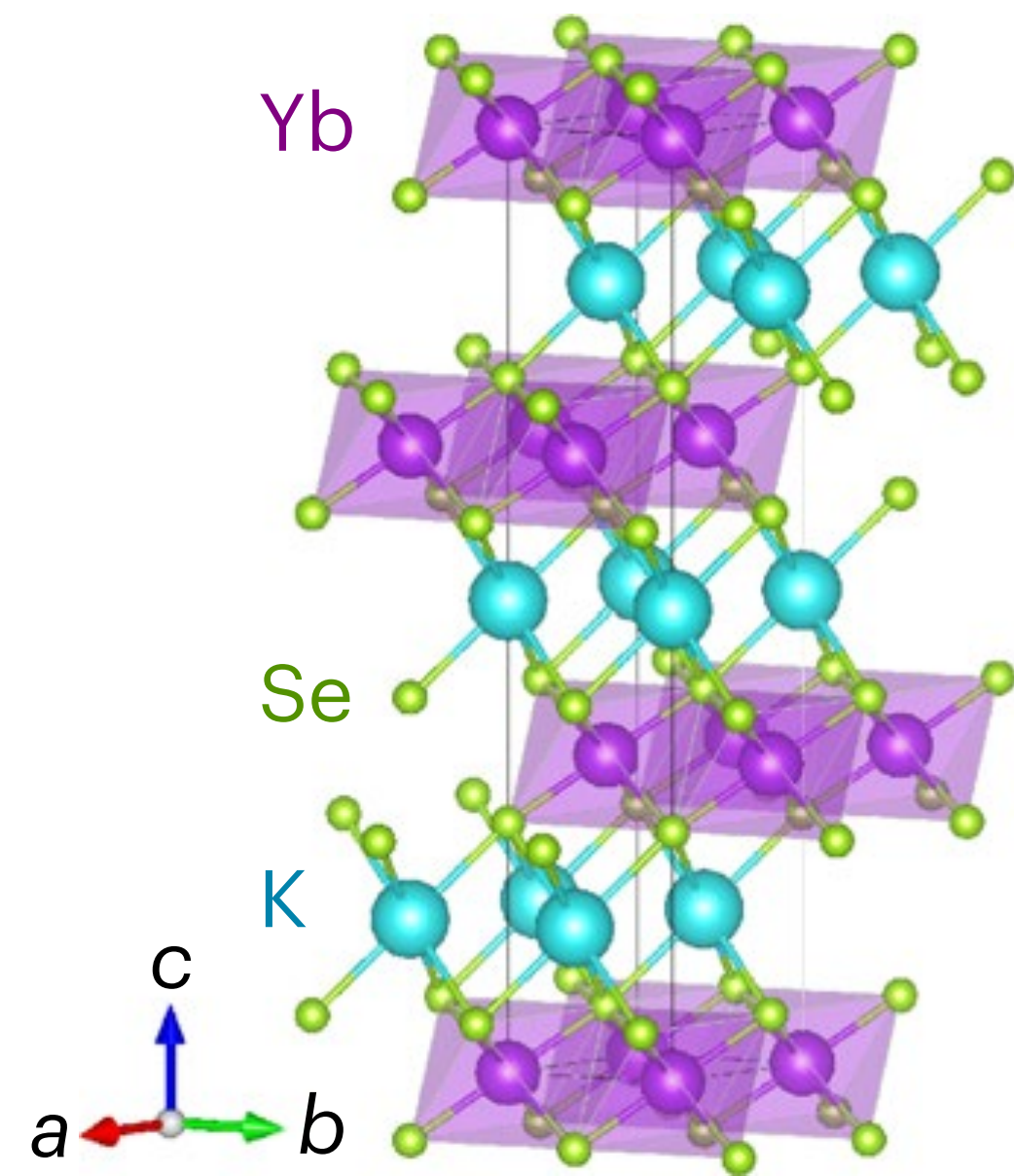
Spin liquid behavior ?



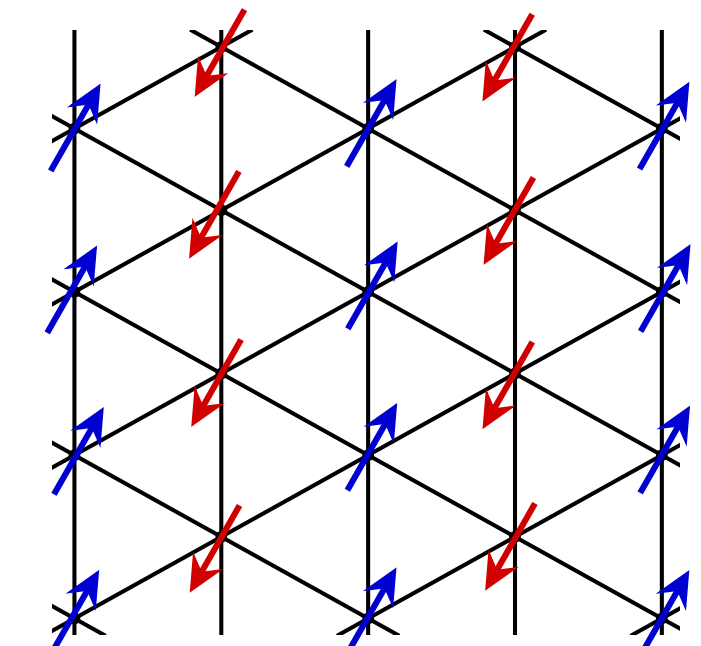
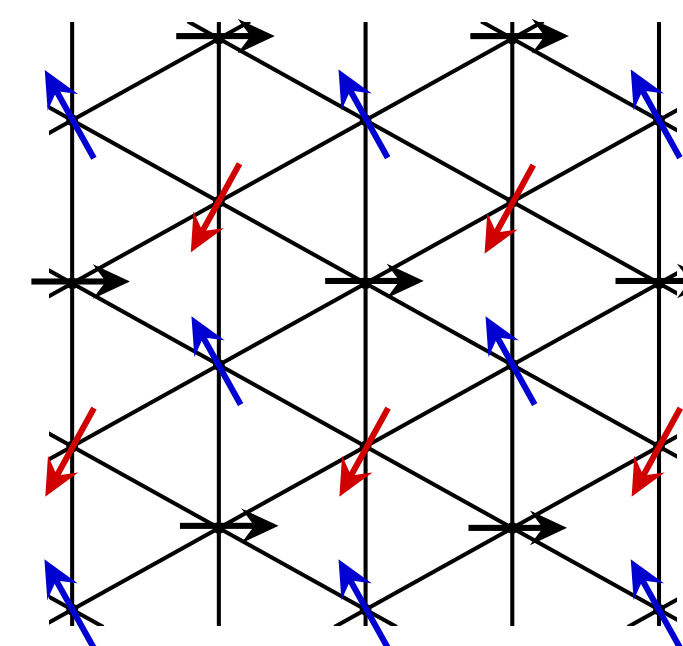
Triangular Lattice J_1 - J_2

AYbSe₂ A=Cs, K, Na

A. O. Scheie et al, arXiv:2406.1777



$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$



AYbSe₂ A=Cs, K, Na

$$\text{Yb}^{3+} \quad 4f^{13} \quad S = 1/2 \quad L = 3 \quad J = 7/2$$

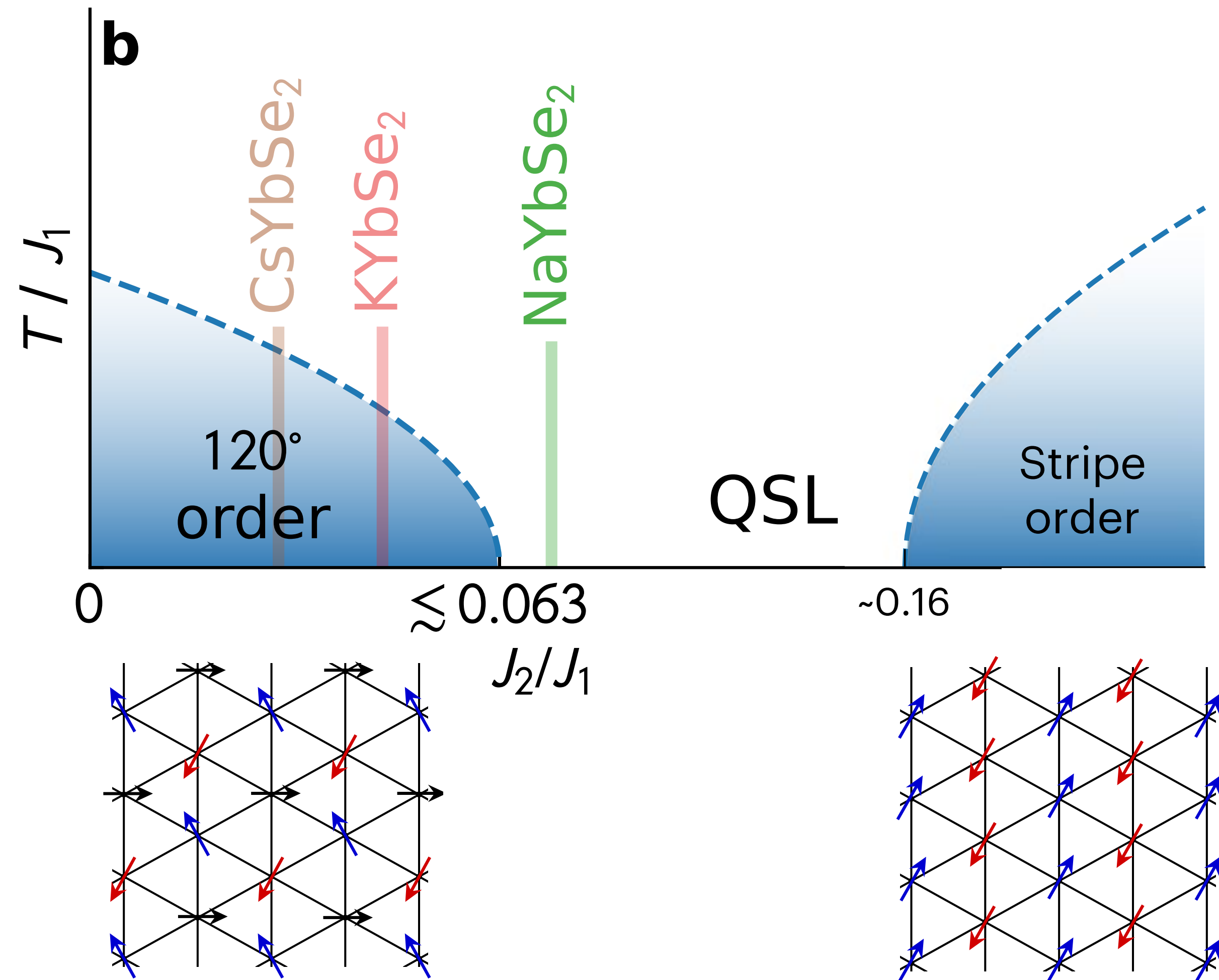
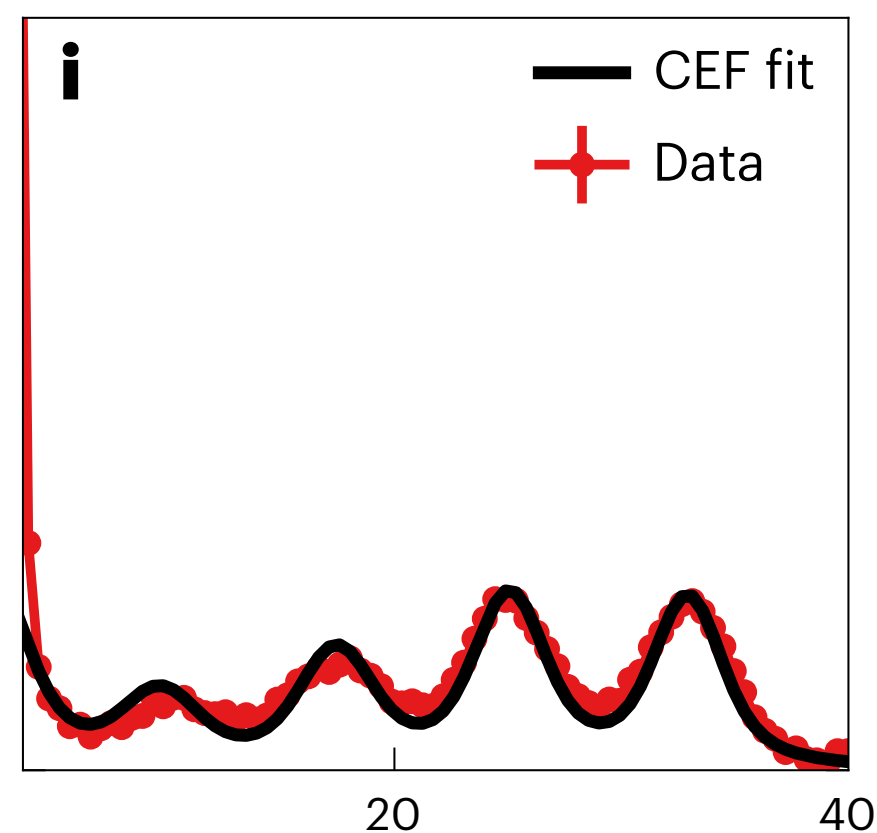
Lowest Kramers doublet in CEF
Pseudospin 1/2

$$|\psi_{\pm}\rangle = 0.78(3) \left| \mp \frac{5}{2} \right\rangle \mp 0.44(4) \left| \pm \frac{1}{2} \right\rangle - 0.44(3) \left| \pm \frac{7}{2} \right\rangle$$

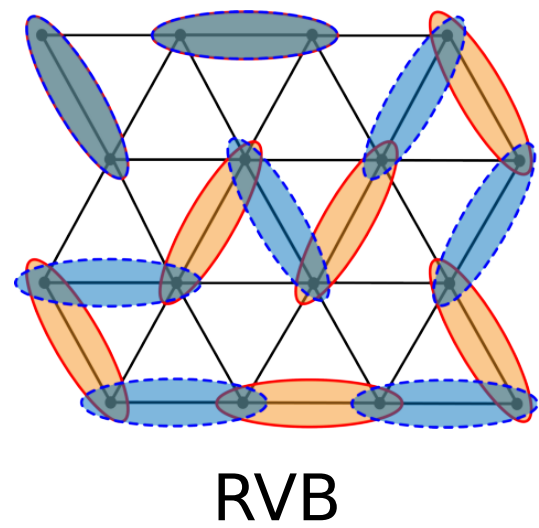
$$g_{xx} = g_{yy} = 3.0(2) \quad g_{zz} = 1.8(6)$$

Easy plane anisotropy

A. O. Scheie et al, arXiv:2406.1777



AYbSe₂ A=Cs, K, Na



Gapped Z₂ SL

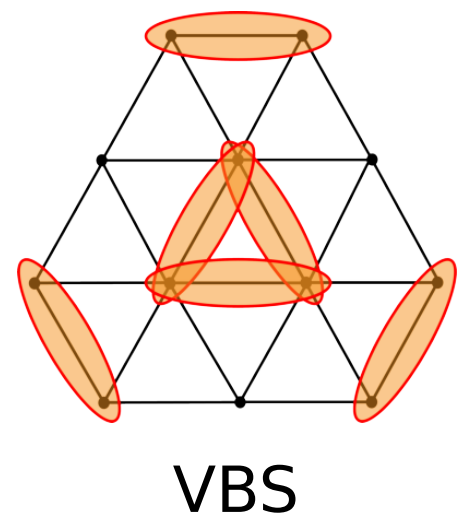
S. Sachdev, PRB 1992

Z. Zhu, S.R. White, PRB 2015 (DMRG)

U(1) Dirac SL

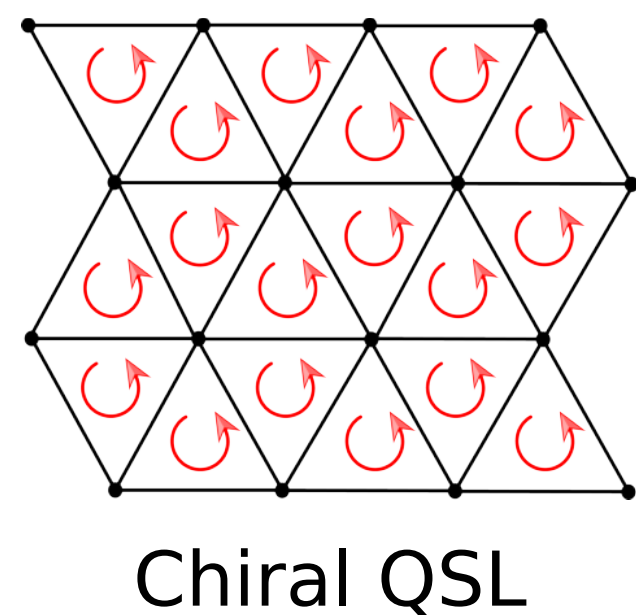
Y. Iqbal et al, PRB 2016 (VMC)

S. Hu et al, PRL 2019 (DMRG)



Valence Bond Solid

U. F. P. Seifert et al, arXiv:2307.12295

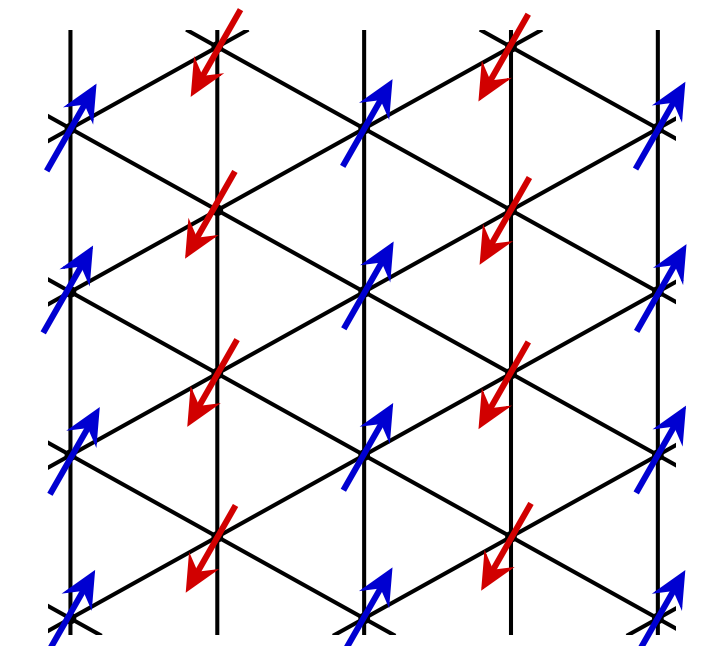
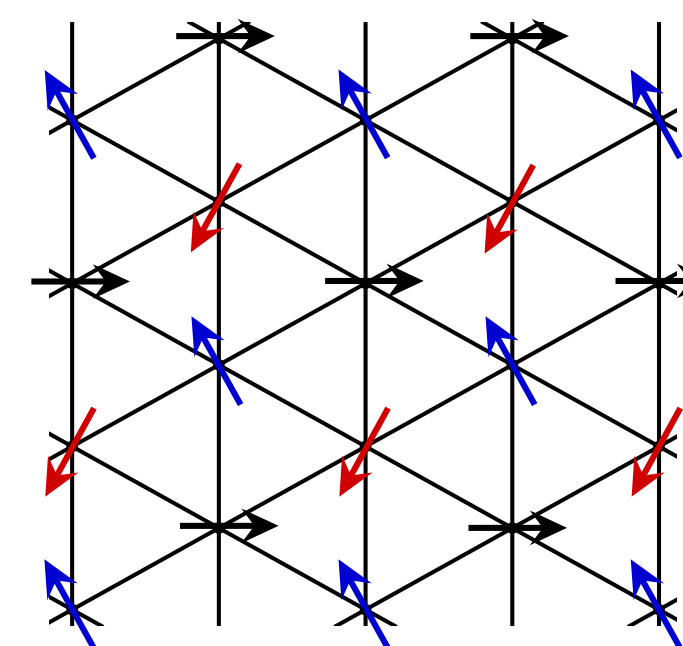
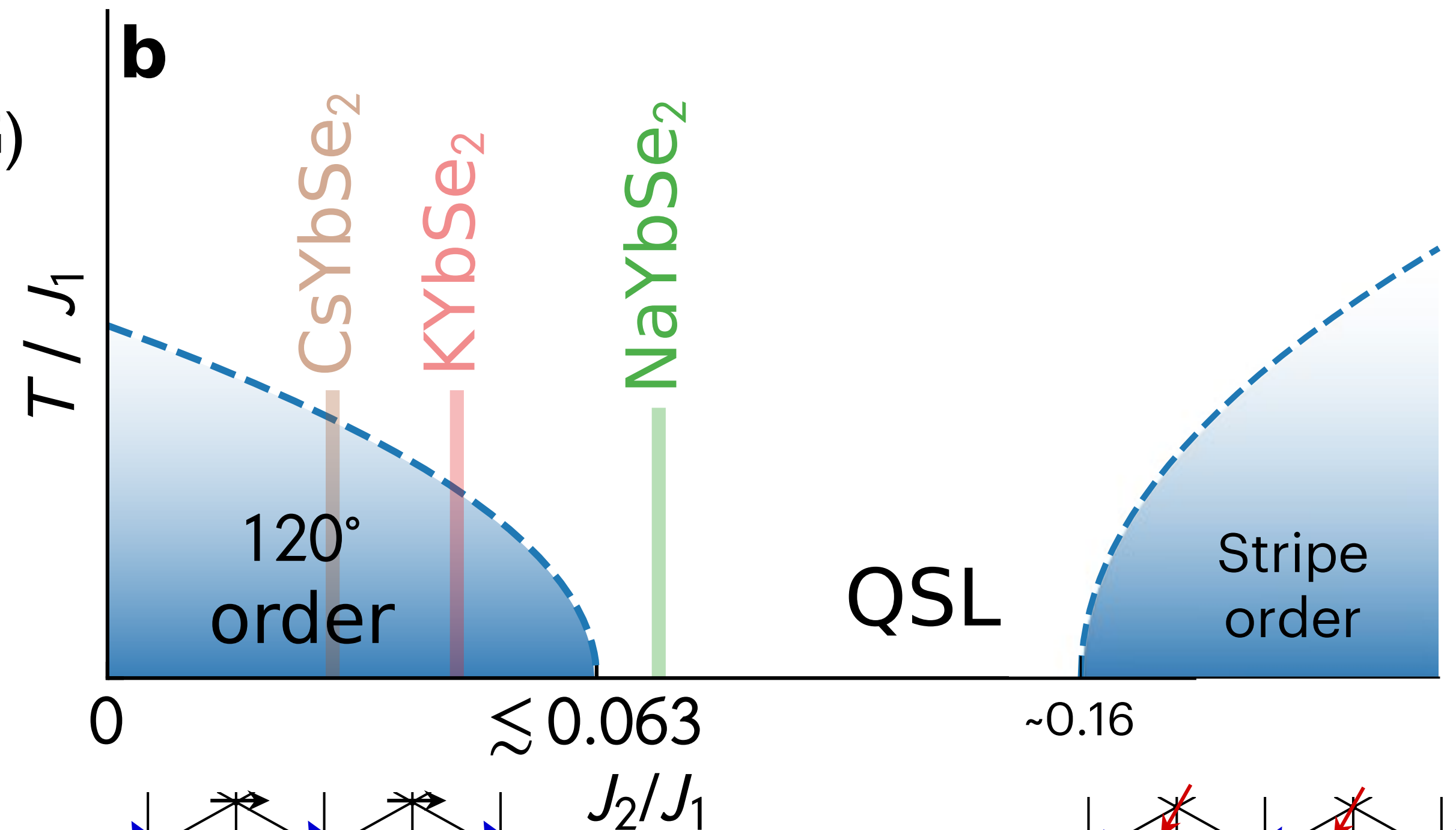


Hubbard Model Chiral Spin Liquid

A. Szasz et al, PRX (2020)

B.-B. Chen et al, PRB (2022)

A. O. Scheie et al, arXiv:2406.1777



Schwinger boson theory

$$H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \dots \quad S_i^a = \frac{1}{2} \sum_{\alpha, \beta} b_{i\alpha}^\dagger \sigma_{\alpha\beta}^a b_{i\beta} \quad \sum_{\alpha} b_{i\alpha}^\dagger b_{i\alpha} = 1$$

$$H_{MF} \sim \frac{1}{2} \sum_{ij} J_{ij} \left(-A_{ij}^* \sum_{\alpha, \beta} \epsilon_{\alpha\beta} b_{i\alpha} b_{j\beta} + B_{ij}^* \sum_{\alpha} b_{i\alpha}^\dagger b_{j\alpha} + h.c. \right)$$

$$A_{ij} = \frac{1}{2} \sum_{\alpha, \beta} \epsilon_{\alpha\beta} \langle b_{i\alpha} b_{j\beta} \rangle \quad B_{ij} = \frac{1}{2} \sum_{\alpha} \langle b_{i\alpha}^\dagger b_{j\alpha} \rangle$$

$$A_{ij} = 0 \quad \text{U(1) spin liquid}$$

$$A_{ij} \neq 0 \quad \text{Z}_2 \text{ spin liquid}$$

Bose condensation leads to magnetic ordering

Only gapped spin liquids are allowed

U(1) spin liquid possible only in 3D

Thermodynamic signatures in quantum spin liquids

Does spinon pairing exist ?

No

Yes

U(1) Spin Liquid
Emergent U(1) gauge field

Z₂ Spin Liquid

gapped
spinon

$$C_{\text{gauge}}^{3D}(T) \sim T^3$$

(unstable in 2D)

~~gapless
spinon~~

$$C_{\text{dirac}}(T) \sim T^2$$
$$C_{\text{FS}}(T) \sim T \quad \text{mean field}$$

$$C_{\text{FS}}^{2D} \sim T^{2/3} \quad \text{including}$$
$$C_{\text{FS}}^{3D} \propto T \ln T \quad \text{fluctuations}$$

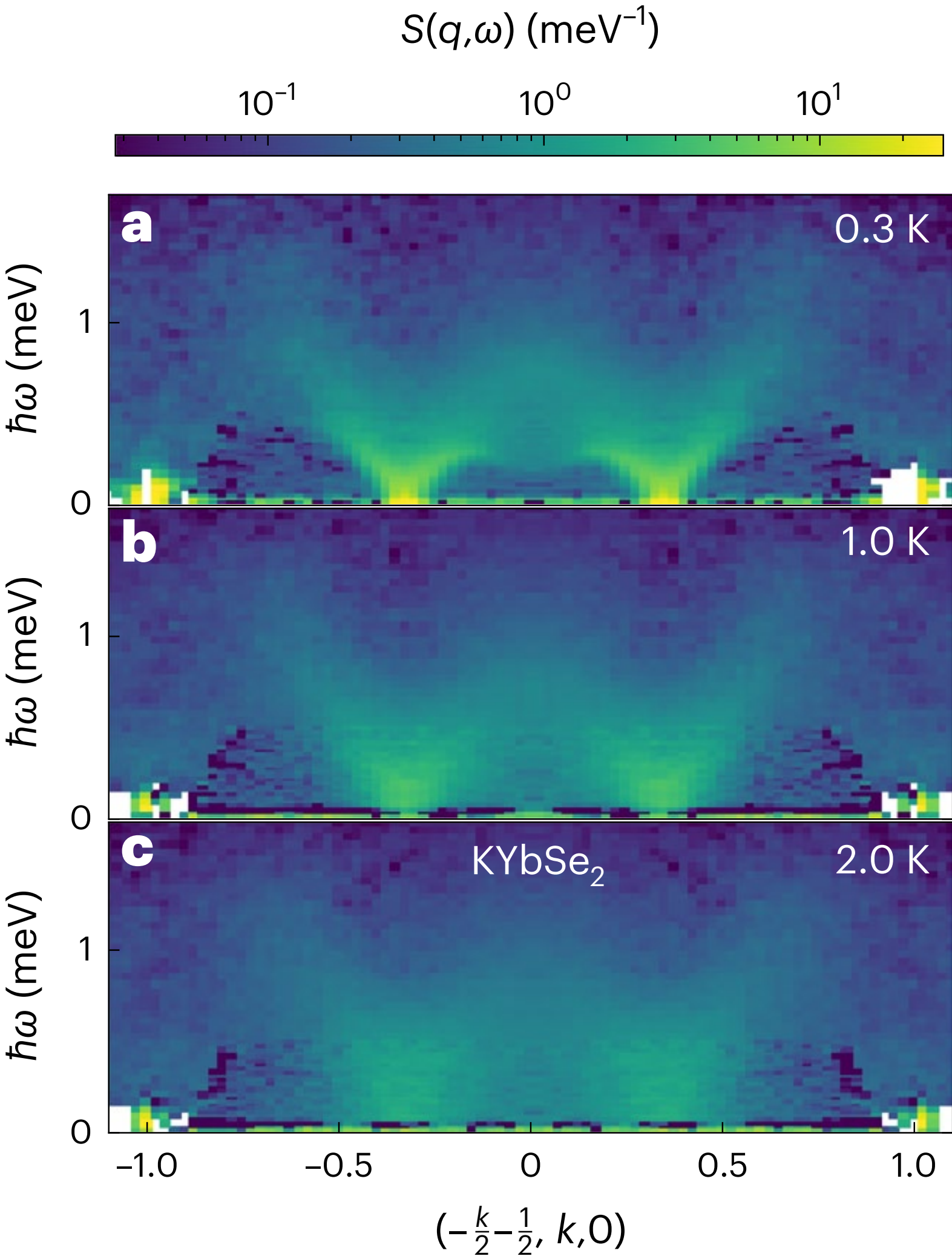
gapped
spinon

$$C(T) \sim e^{-\Delta/T}$$

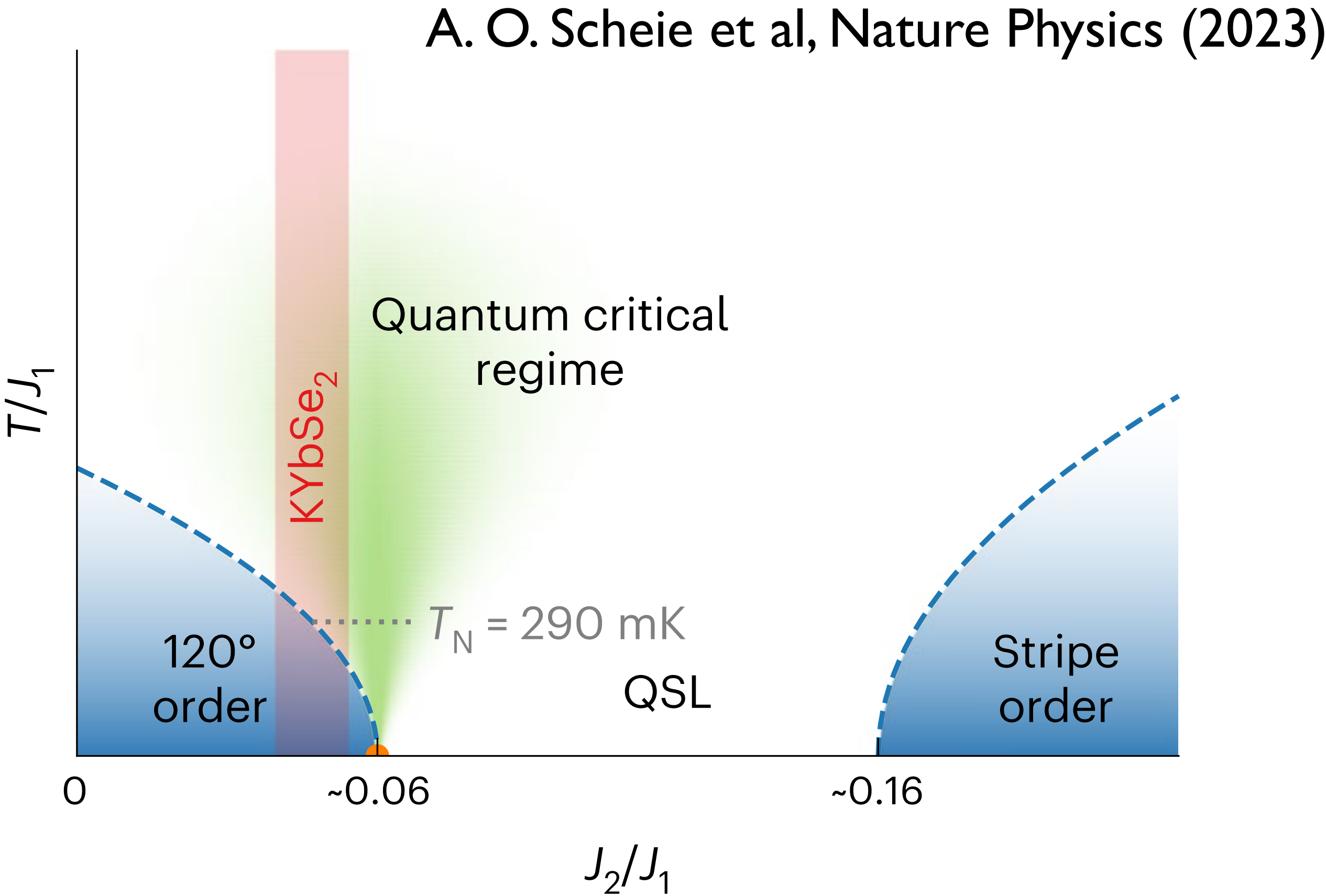
~~gapless
spinon~~

$$C_{\text{d-wave}}(T) \sim T^2$$

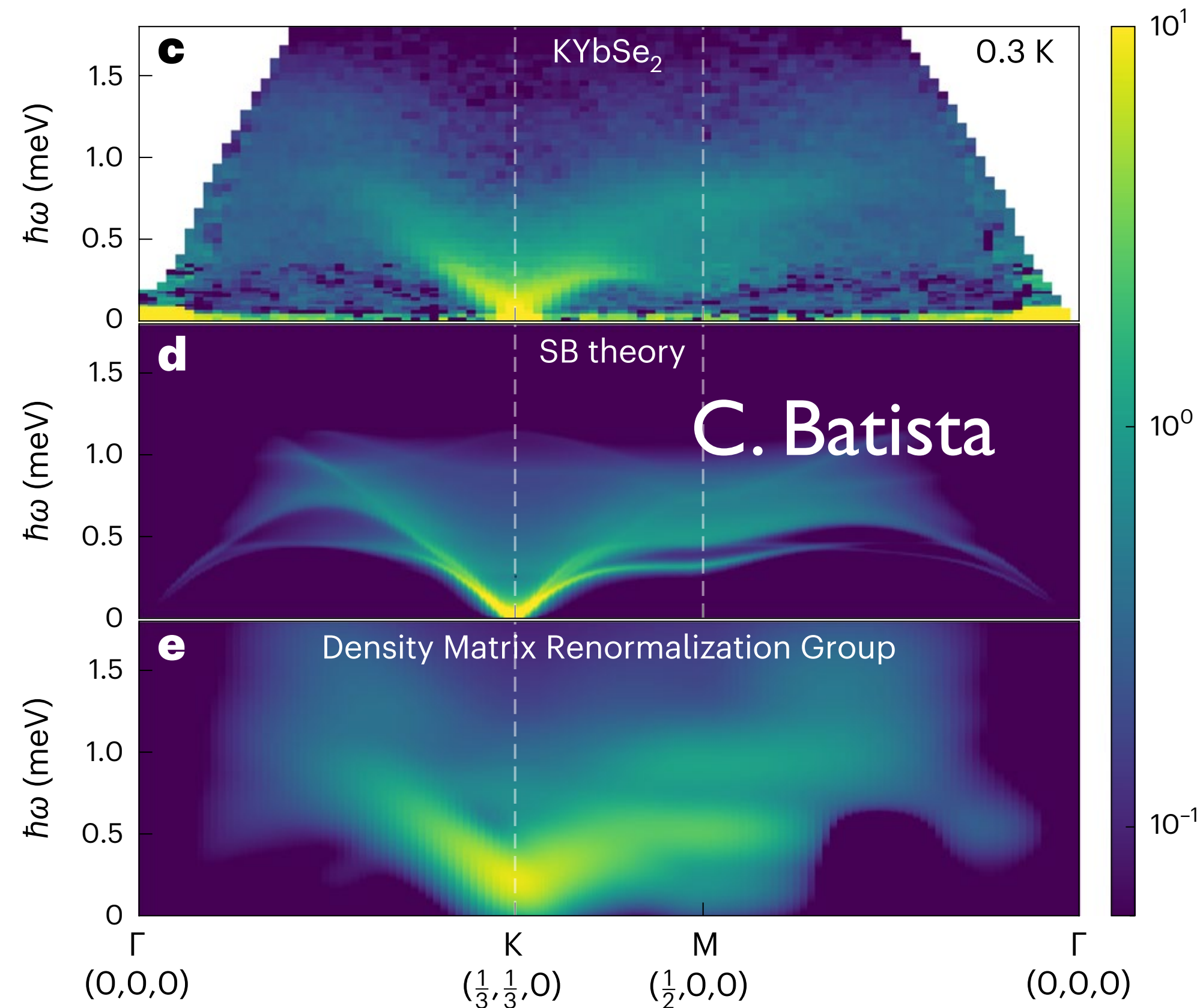
KYbSe₂



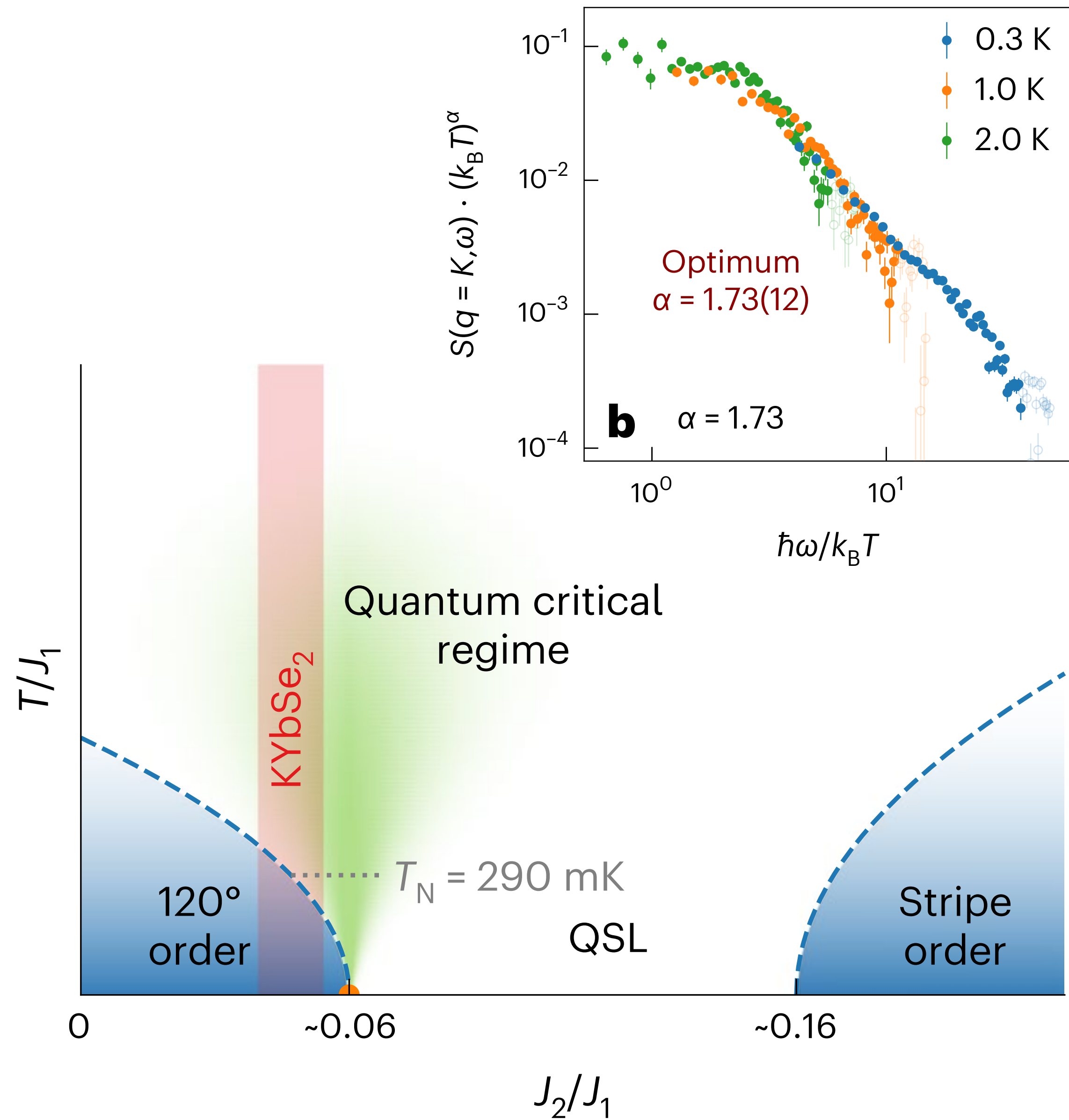
Theoretical technique	J_1 (meV)	J_2/J_1
Nonlinear spin waves	0.456 ± 0.013	0.043 ± 0.010
Heat capacity	0.429 ± 0.010	0.037 ± 0.013



KYbSe₂

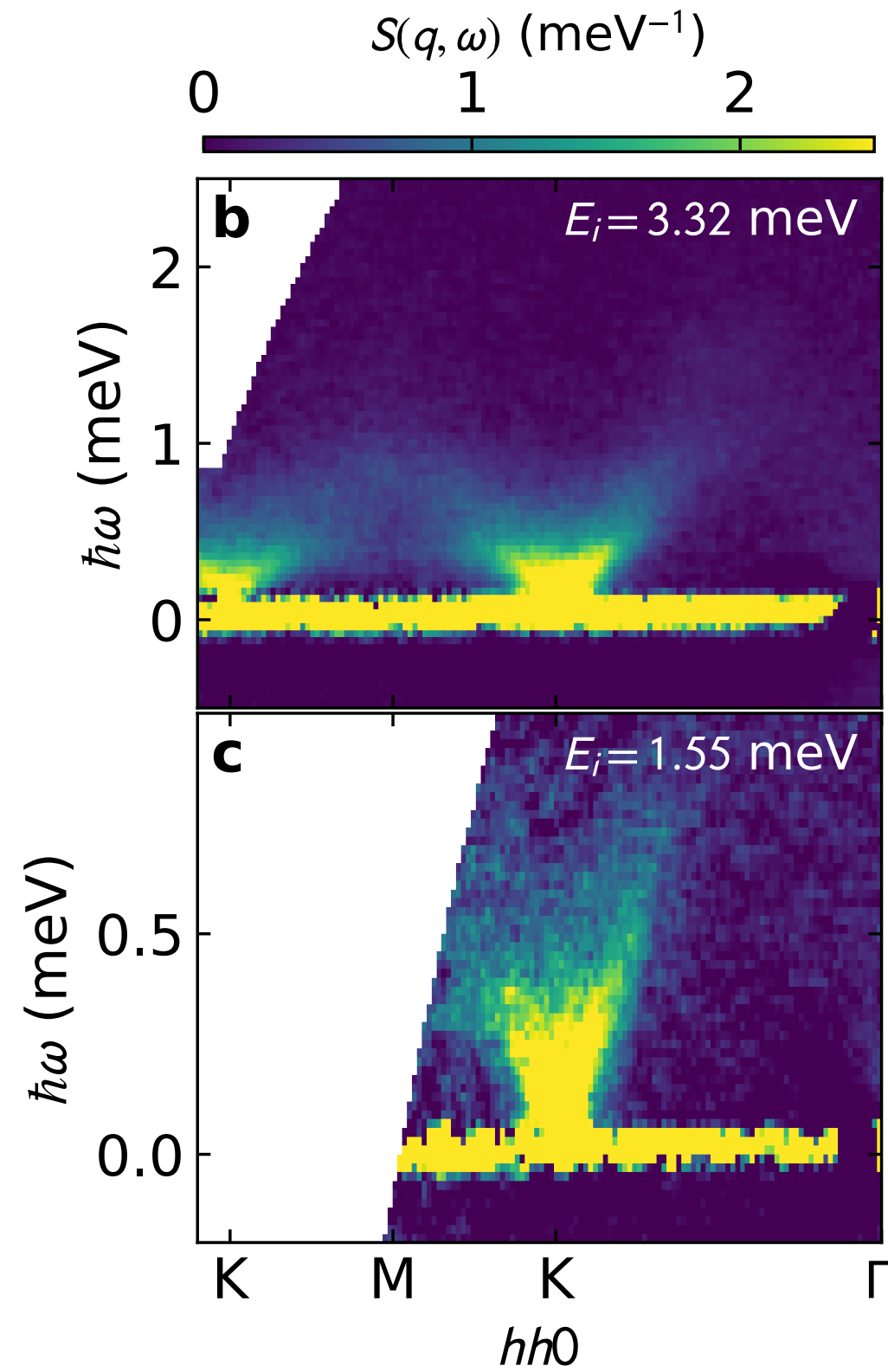
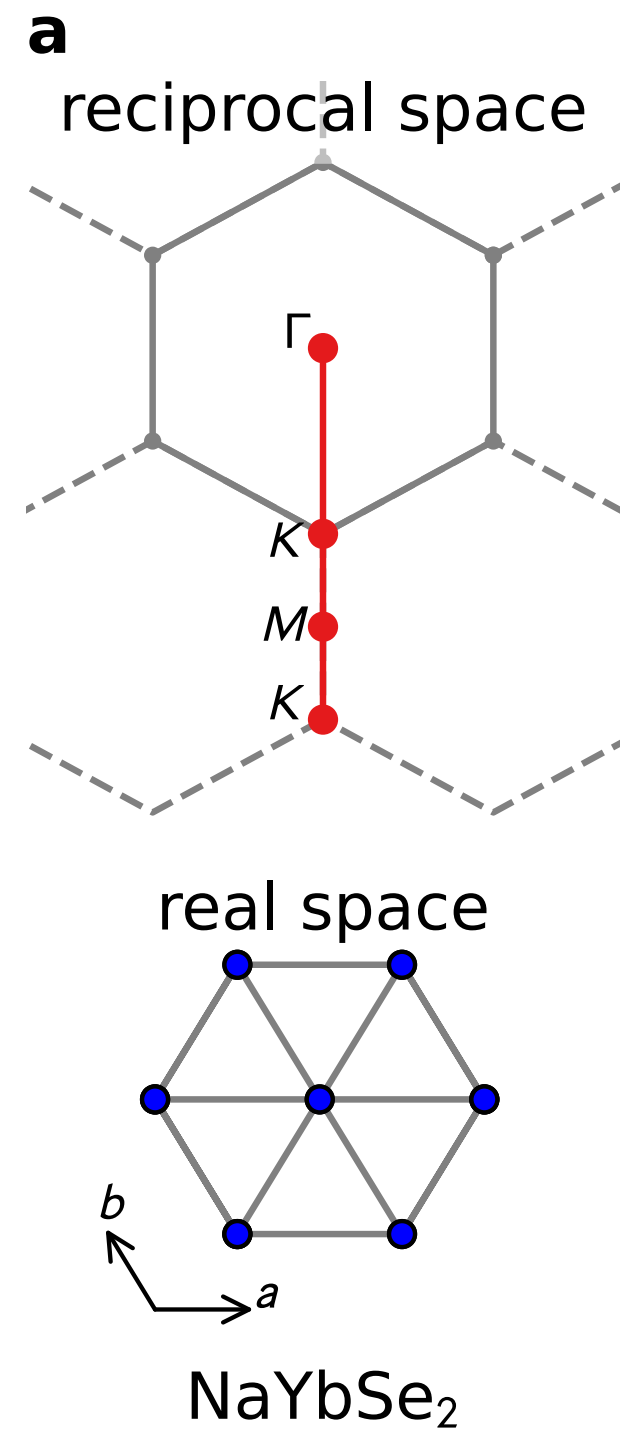


C. Batista

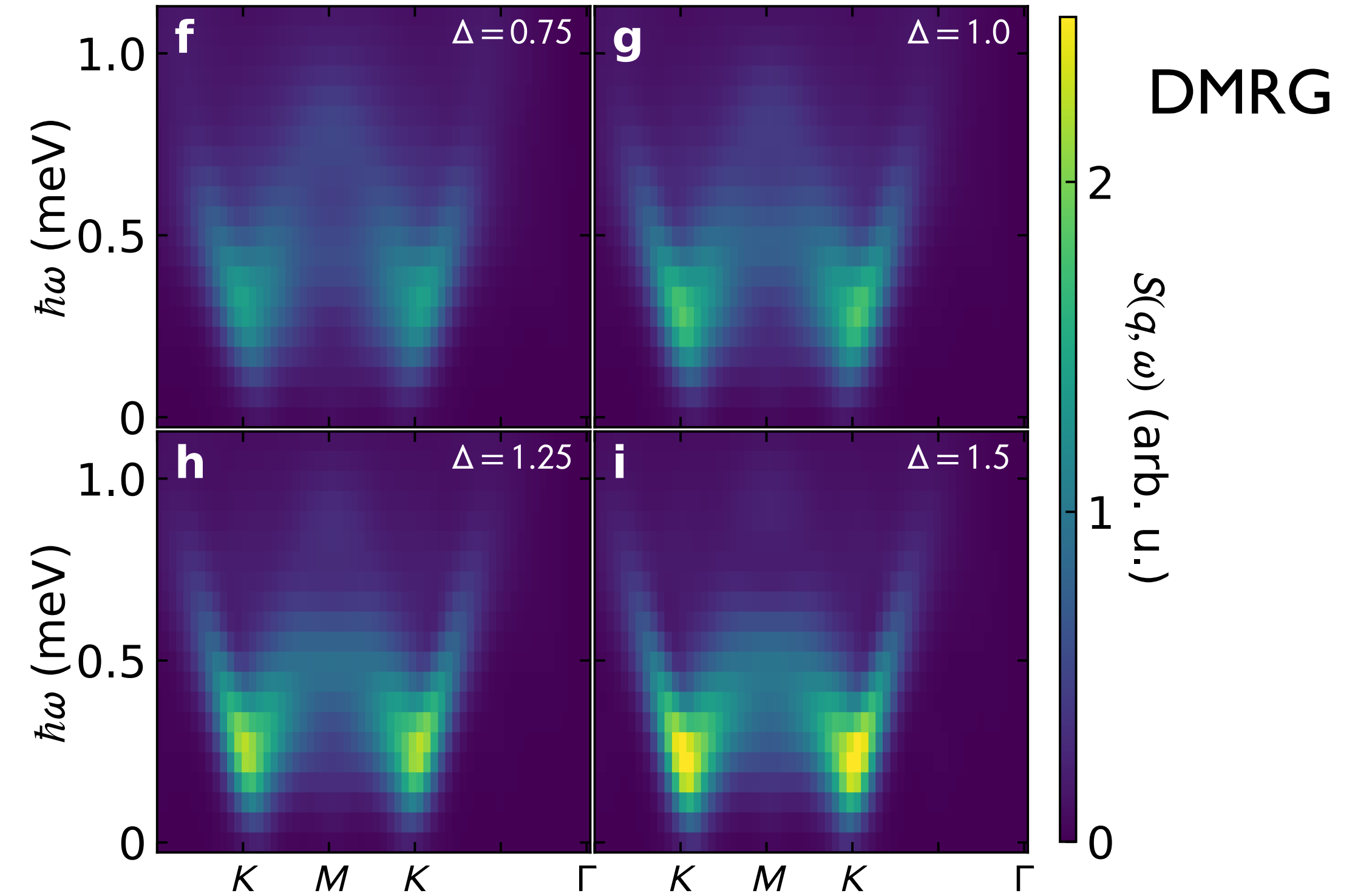


NaYbSe₂

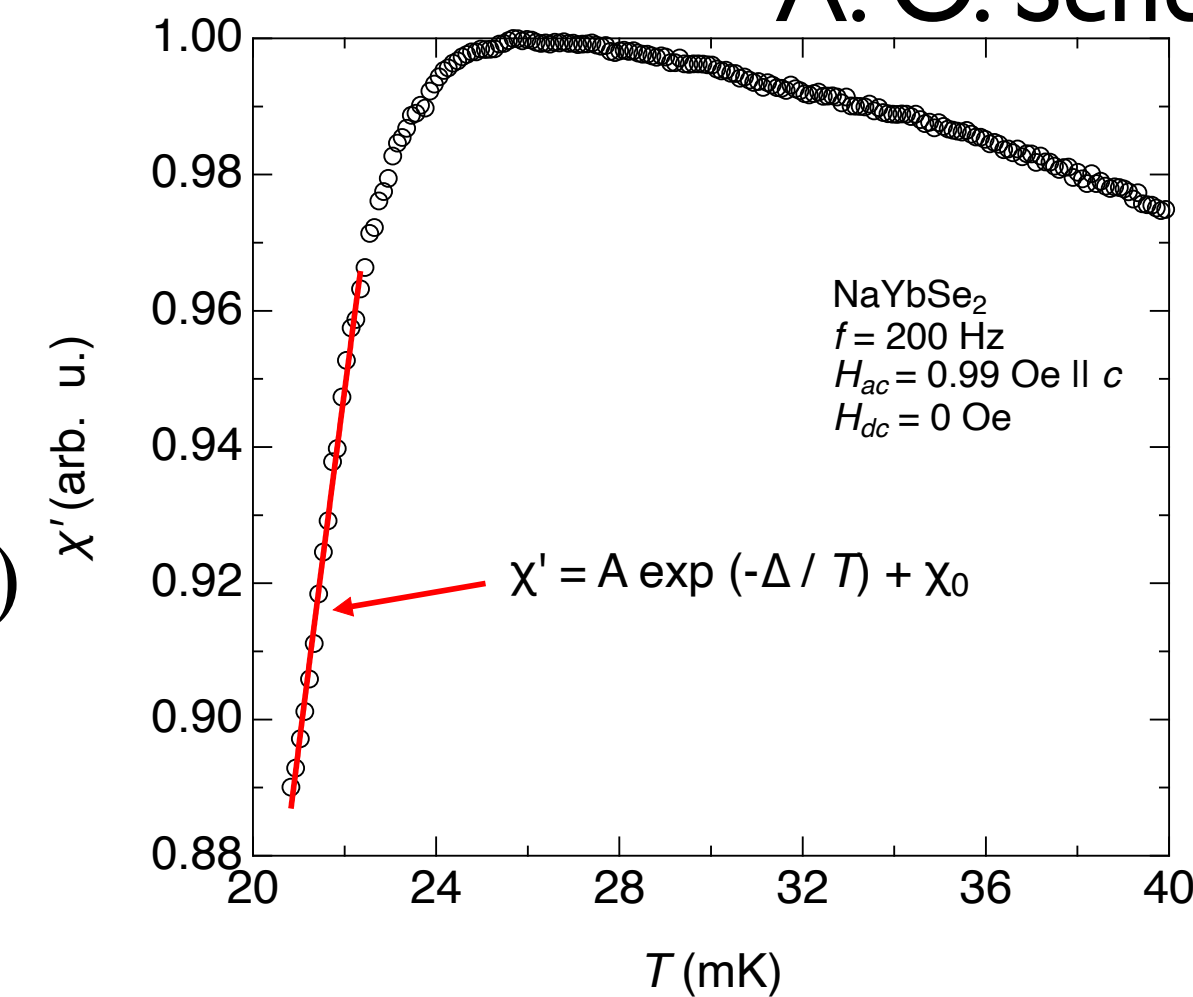
No order down to 100 *mK*



MPS simulations



A. O. Scheie et al, arXiv:2406.1777



Excitation gap
2.1 μeV .

$$H = J_1 \sum_{\langle i,j \rangle} (S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z) + J_2 \sum_{\langle\langle i,j \rangle\rangle} (S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z)$$

$$J_2/J_1 = 0.071$$