TECHNISCHE UNIVERSITÄT DRESDEN

DRESDEN concept

Hidden symmetries and exotic orders in quantum magnets

Lukas Janssen

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Wilhelm Krüger

Experiments (Peking U):

Wenjie Chen

Xianghong Jin

Yuan Li

Quantum Critical Matter

JA 2306/4-1

Emmy Noether-Programm

DFG Deutsche Forschungsgemeinschaft

ct.qmat

Complexity and Topology in Quantum Matter

Würzburg-Dresden Cluster of Excellence

SFB 1143

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Hidden symmetries and exotic orders in ~~quantum magnets~~ the Kitaev magnet $Na_2Co_2TeO_6$

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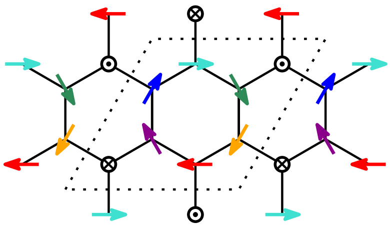
Outline

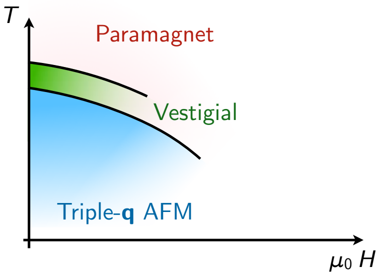
Message #1: Kitaev magnet $Na_2Co_2TeO_6$ features triple-**q** AFM order at low temperatures

[Krüger, Chen, Jin, Li, LJ, PRL '23]

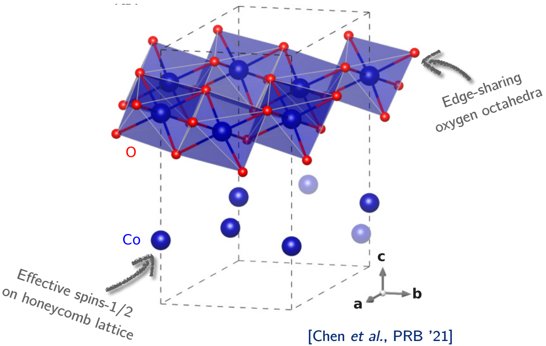


Message #2: Triple-**q** order can melt in two stages

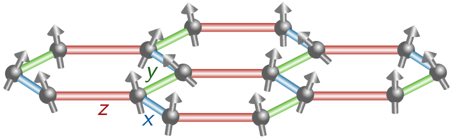
[Francini, LJ, PRB '24]



Magnetic Mott insulator $Na_2Co_2TeO_6$

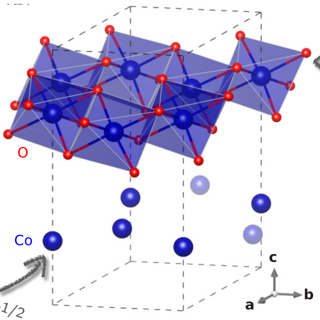


[Chen *et al.*, PRB '21]

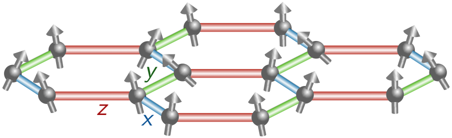


[Liu, Khaliullin, PRB '18]
[Sano, Kato, Motome, PRB '18]

Magnetic Mott insulator $Na_2Co_2TeO_6$



[Chen *et al.*, PRB '21]



Effective spin-1/2 model:

$$\mathcal{H} = K \left(\sum_{\langle ij \rangle_x} S_i^x S_j^x + \sum_{\langle ij \rangle_y} S_i^y S_j^y + \sum_{\langle ij \rangle_z} S_i^z S_j^z \right) + \dots$$

Kitaev exchange

Non-Kitaev terms

[Liu, Khaliullin, PRB '18]
[Sano, Kato, Motome, PRB '18]

Magnetic Mott insulator $Na_2Co_2TeO_6$

Non-Kitaev terms

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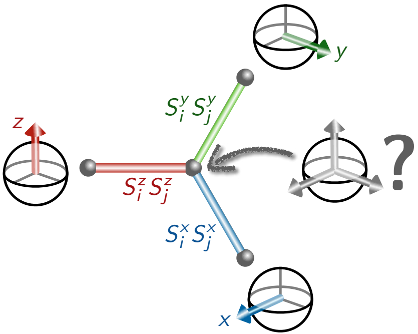
Kitaev exchange

[Kitaev, Ann. Phys. '06]

Frustration:

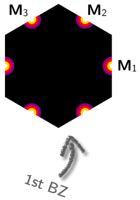


Multi-q vs single-q order: Bragg peaks

Static spin structure factor:

... e.g., from neutron diffraction

[Chen *et al.*, PRB '21]



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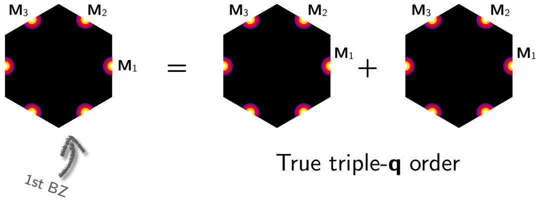

True triple-**q** order

Multi-q vs single-q order: Bragg peaks

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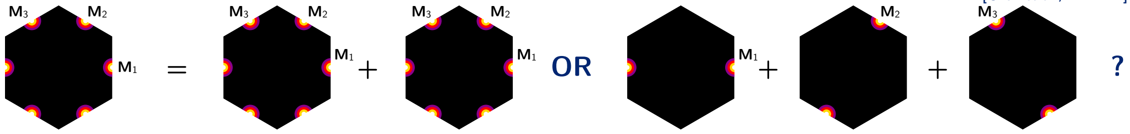


1st BZ

True triple-**q** order

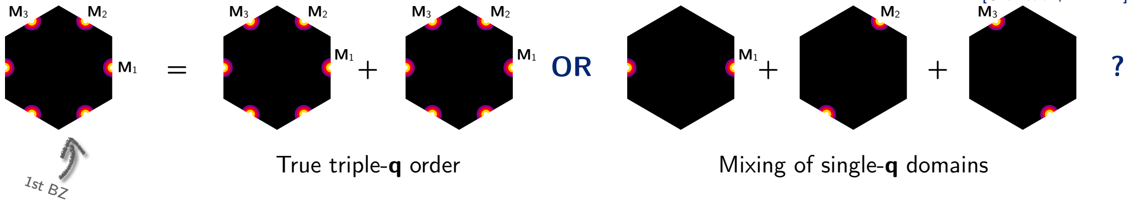
Mixing of single-**q** domains

Multi-q vs single-q order: Bragg peaks

Static spin structure factor:

... e.g., from neutron diffraction

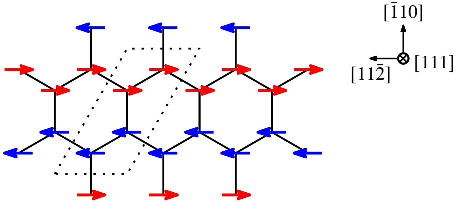
[Chen *et al.*, PRB '21]



True triple-**q** order

Mixing of single-**q** domains

Real space:



α-$RuCl_3$, Na_2IrO_3, ...

[Choi *et al.*, PRL '12]
[Johnson *et al.*, PRB '15]
[Balz *et al.*, PRB '21]

Multi-q vs single-q order: Bragg peaks

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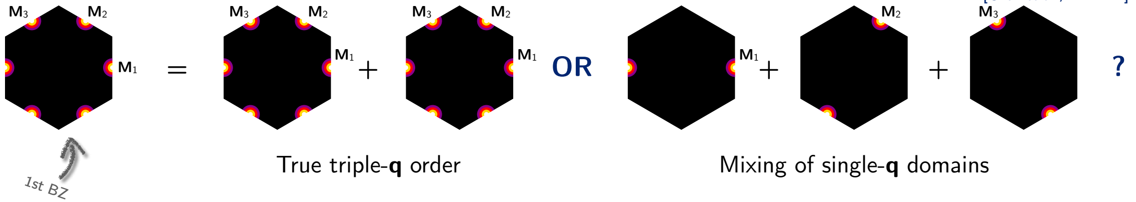

True triple-**q** order

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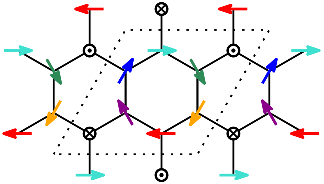
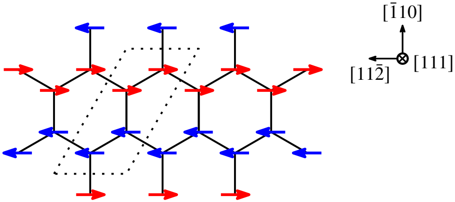


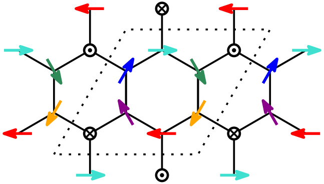
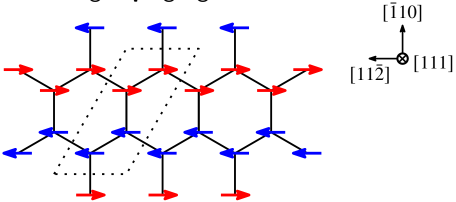
Kitaev-Heisenberg in field: [LJ, Andrade, Vojta, PRL '16]
Bilinear-Biquadratic Kitaev-Heisenberg: [Pohle, Shannon, Motome, PRB '23]



α-$RuCl_3$, Na_2IrO_3, ...

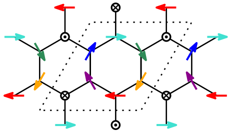
[Choi *et al.*, PRL '12]
[Johnson *et al.*, PRB '15]
[Balz *et al.*, PRB '21]

Multi-q vs single-q order: Symmetries

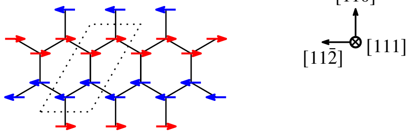
	Triple-**q** AFM	Single-**q** zigzag AFM
		
Time reversal:	✗	✗
Spin-lattice rotation C_3^*:	✓	✗
Translation $T_{\mathbf{a}_1}$:	✗	✗
Translation $T_{\mathbf{a}_2}$:	✗	✓

Multi-q vs single-q order: Magnetic excitation spectrum

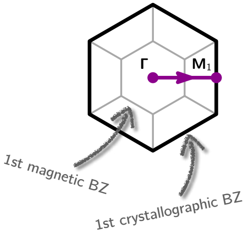
Triple-**q** AFM



Single-**q** zigzag AFM

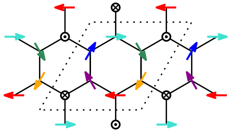


Brillouin zone path:

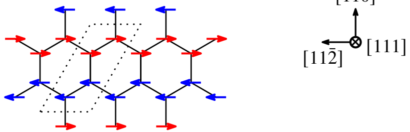


Multi-q vs single-q order: Magnetic excitation spectrum

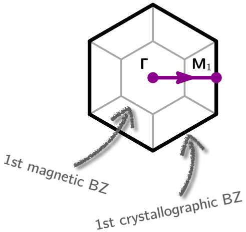
Triple-**q** AFM



Single-**q** zigzag AFM



Brillouin zone path:

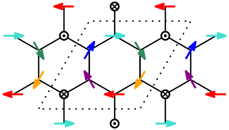


Spectrum necessarily symmetric

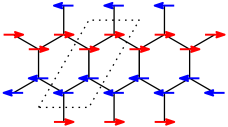
... independent of modeling

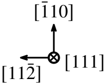
Multi-q vs single-q order: Magnetic excitation spectrum

Triple-**q** AFM

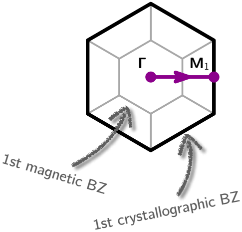


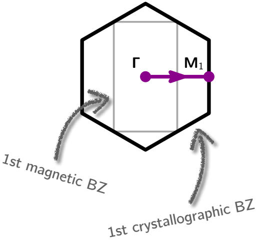
Single-**q** zigzag AFM





Brillouin zone path:





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Multi-q vs single-q order: Magnetic excitation spectrum

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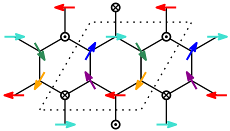
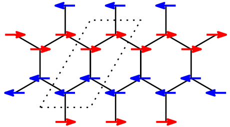
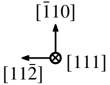


Single-**q** zigzag AFM





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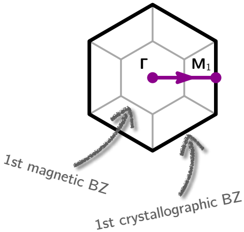

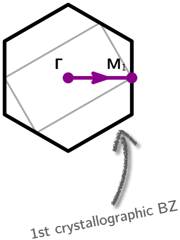

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Multi-q vs single-q order: Magnetic excitation spectrum

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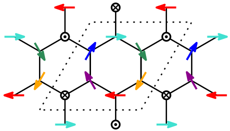
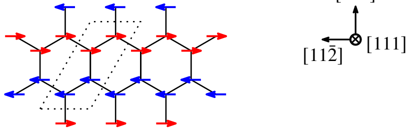
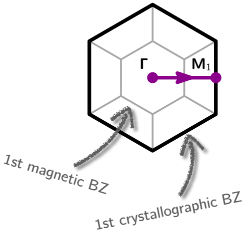
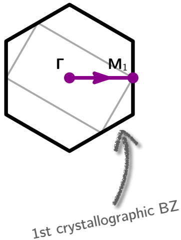


Single-**q** zigzag AFM



Brillouin zone path:





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Multi-q vs single-q order: Magnetic excitation spectrum

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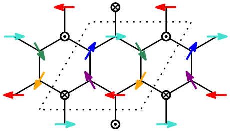
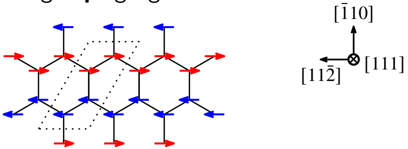


Single-**q** zigzag AFM



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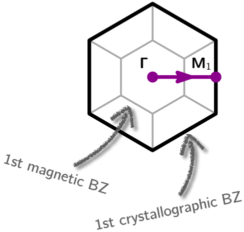

Spectrum necessarily symmetric

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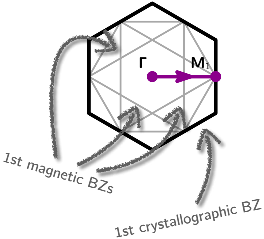
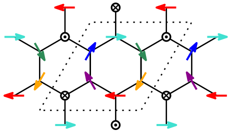


Spectrum generically asymmetric

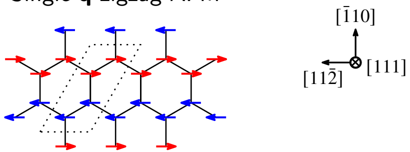
... unless fine-tuned

Multi-q vs single-q order: Magnetic excitation spectrum

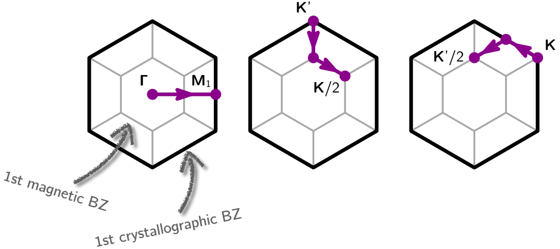
Triple-**q** AFM

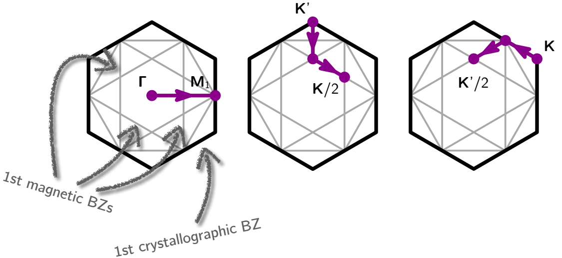


Single-**q** zigzag AFM



Brillouin zone path:





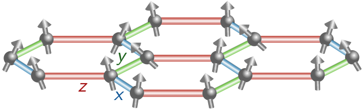
Spectrum necessarily symmetric

... independent of modeling

Spectrum generically asymmetric

... unless fine-tuned

Example: HKΓΓ' model @ hidden SU(2) point



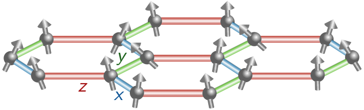
Hamiltonian:

$$\mathcal{H}_0 = \sum_{\langle ij \rangle_\gamma} \Big[J_1 \mathbf{S}_i \cdot \mathbf{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \Gamma_1' (S_i^\gamma S_j^\alpha + S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\beta + S_i^\beta S_j^\gamma) \Big]$$

... with (J_1, K_1, Γ_1, Γ'_1) = (-1/9, -2/3, 8/9, -4/9) A

[Chaloupka, Khaliullin, PRB '15]

Example: HKΓΓ' model @ hidden SU(2) point



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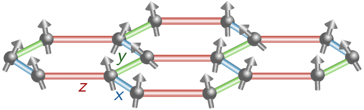
... with (J_1, K_1, Γ_1, Γ'_1) = (-1/9, -2/3, 8/9, -4/9) A

[Chaloupka, Khaliullin, PRB '15]

Hidden SU(2) symmetry: $\mathbf{S}_i \mapsto \tilde{\mathbf{S}}_i = T_{14} \mathbf{S}_i$: $\mathcal{H}_0 \mapsto \tilde{\mathcal{H}}_0 = A \sum_{\langle ij \rangle} \tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j$

Local rotation

Example: HKΓΓ’ model @ hidden SU(2) point



Hamiltonian:

$$\mathcal{H}_0 = \sum_{\langle ij\rangle_\gamma} \Big[J_1 \mathbf{S}_i \cdot \mathbf{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \Gamma_1' (S_i^\gamma S_j^\alpha + S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\beta + S_i^\beta S_j^\gamma) \Big]$$

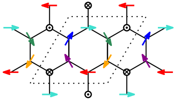
... with $(J_1, K_1, \Gamma_1, \Gamma'_1) = (-1/9, -2/3, 8/9, -4/9)\ A$

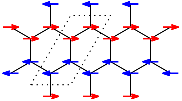
[Chaloupka, Khaliullin, PRB ’15]

Hidden SU(2) symmetry: $\mathbf{S}_i \mapsto \tilde{\mathbf{S}}_i = T_{14} \mathbf{S}_i$: $\mathcal{H}_0 \mapsto \tilde{\mathcal{H}}_0 = A \sum_{\langle ij\rangle} \tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j$

Ground state manifold:

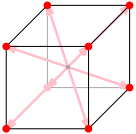
$\mathbf{S}_i$:



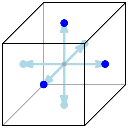


$\updownarrow$ $\updownarrow$

$\tilde{\mathbf{S}}_i$:

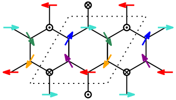


4 diagonals

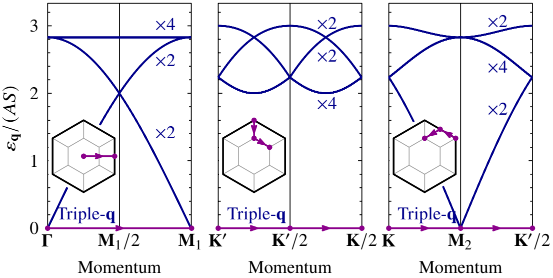


3 cubic axes

Magnon spectrum @ hidden SU(2) point

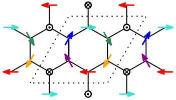


Triple-**q** AFM

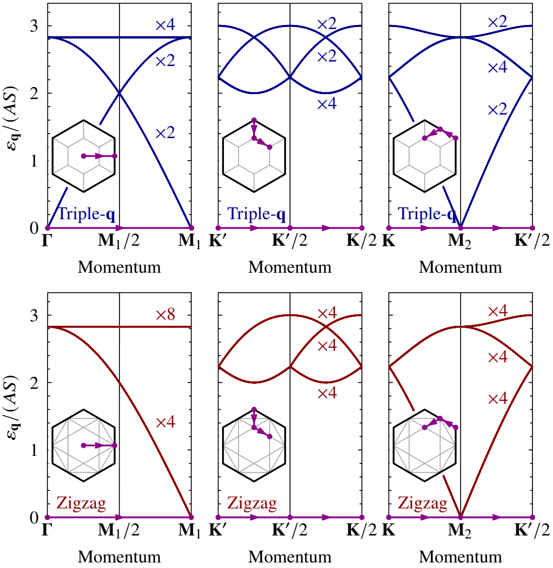


… fully symmetric

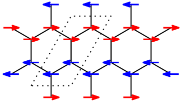
Magnon spectrum @ hidden SU(2) point



Triple-**q** AFM



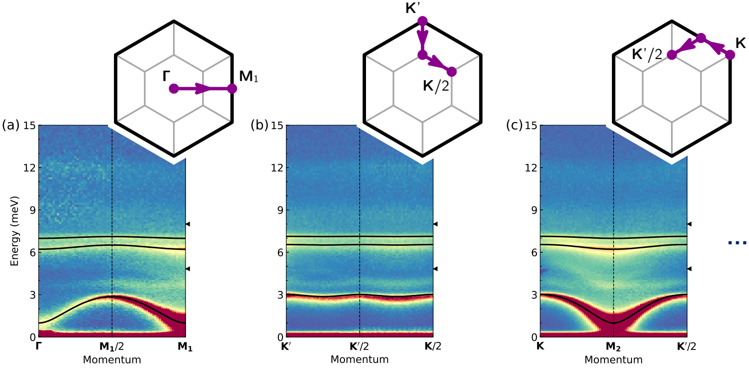
... fully symmetric



Zigzag AFM

... at least some bands asymmetric

$Na_2Co_2TeO_6$: Inelastic neutron scattering

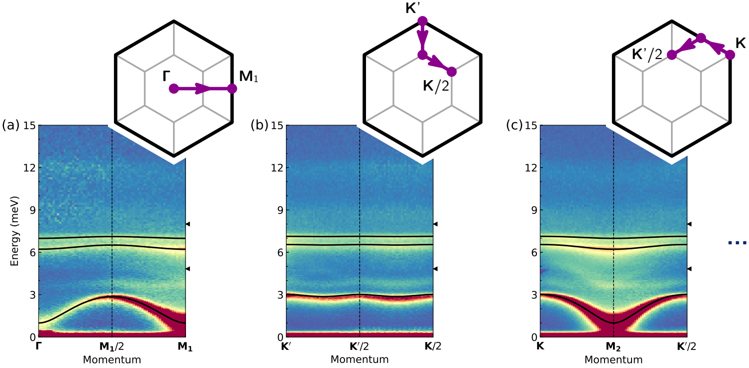


... perfectly symmetric magnon bands

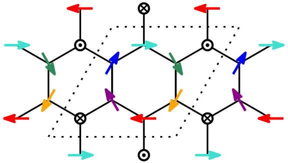
[Krüger, Chen, Jin, Li, LJ, PRL ’23]

→ Poster by W. Krüger

$Na_2Co_2TeO_6$: Inelastic neutron scattering



... perfectly symmetric magnon bands



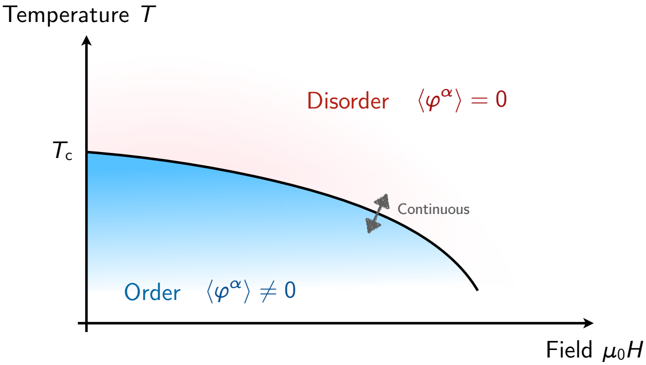
Message #1: $Na_2Co_2TeO_6$ features triple-**q** AFM order at low temperatures

[Krüger, Chen, Jin, Li, LJ, PRL '23]

→ Poster by W. Krüger

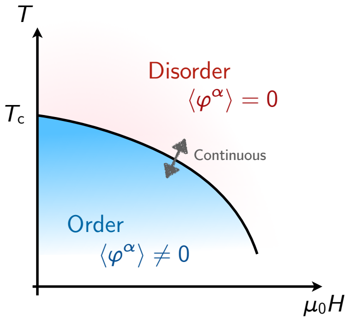
Finite-temperature phase diagram: Simple system

... order parameter has only 1 component
or breaks only 1 symmetry



Finite-temperature phase diagram: Multicomponent system

... order parameter has > 1 component
and breaks > 1 symmetry



Single continuous ✘

[Fernandes, Orth, Schmalian, ARCMP '19]

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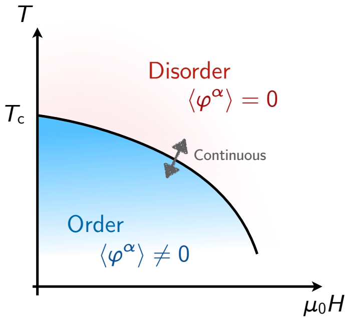

Single continuous ✗

First order ✓

[Fernandes, Orth, Schmalian, ARCMP '19]

Finite-temperature phase diagram: Multicomponent system

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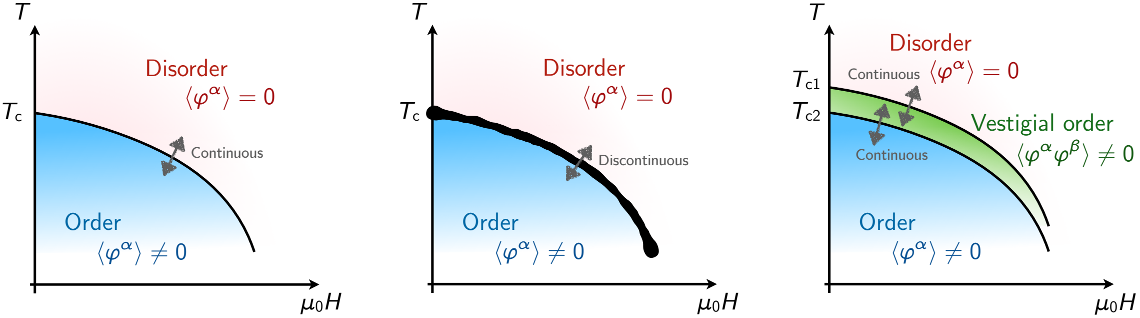
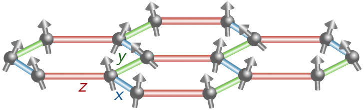


Single continuous ✗

First order ✓ **OR** Two-stage melting ✓

[Fernandes, Orth, Schmalian, ARCMP '19]

HKΓΓ' model with ring perturbation



Hamiltonian:

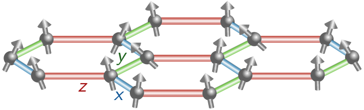
$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\hexagon}$$

Bilinear exchange:

$$\mathcal{H}_0 = \sum_{\langle ij \rangle_\gamma} \Big[J_1 \mathbf{S}_i \cdot \mathbf{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \Gamma_1' (S_i^\gamma S_j^\alpha + S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\beta + S_i^\beta S_j^\gamma) \Big]$$

... with (J_1, K_1, Γ_1, Γ'_1) = (-1/9, -2/3, 8/9, -4/9) A

HKΓΓ' model with ring perturbation



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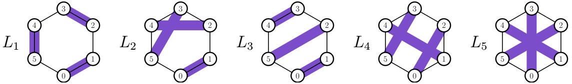
... with (J_1, K_1, Γ_1, Γ'_1) = (-1/9, -2/3, 8/9, -4/9) A

Ring exchange:



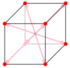
$$\mathcal{H}_{\hexagon} = J_{\hexagon} \sum_{\langle ijklmn \rangle} L_{ijklmn} (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l)(\mathbf{S}_m \cdot \mathbf{S}_n)$$

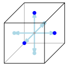
... with L_1 = 1/3, L_2 = -1, L_3 = 1/2, L_4 = 1/2, L_5 = -1/6
from strong-coupling expansion of honeycomb-lattice Hubbard model
[Yang, Albuquerque, Capponi, Läuchli, Schmidt, NJP '12]

Observables

Dual magnetization:

$$\tilde{M} = \langle |\tilde{\mathbf{M}}| \rangle \qquad \text{with} \qquad \tilde{\mathbf{M}} := \frac{1}{N}\sum_i (-1)^i T_{14}^{\top} \mathbf{S}_i = \begin{cases} \frac{1}{\sqrt{3}}(1,1,1), & \text{for triple-}\mathbf{q}\text{ order} \\ (0,0,1), & \text{for } z\text{-zigzag order} \end{cases}$$





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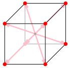
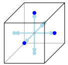




Composite order parameter:

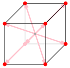
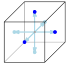
$$(Q_{e_g}, Q_{t_{2g}}) = (\langle|\mathbf{Q}_{e_g}|\rangle, \langle|\mathbf{Q}_{t_{2g}}|\rangle) \qquad \text{with} \qquad \begin{aligned} \mathbf{Q}_{e_g} &:= \tfrac{1}{2}\big(2\tilde{M}_z^2 - \tilde{M}_x^2 - \tilde{M}_y^2, \sqrt{3}[\tilde{M}_x^2 - \tilde{M}_y^2]\big) \\ \mathbf{Q}_{t_{2g}} &:= \sqrt{3}\big(\tilde{M}_y\tilde{M}_z, \tilde{M}_z\tilde{M}_y, \tilde{M}_x\tilde{M}_y\big) \end{aligned}$$

... 5 irreducible components of rank-2 tensor

Observables

Dual magnetization:

$$\tilde{M} = \langle|\tilde{\mathbf{M}}|\rangle \qquad \text{with} \qquad \tilde{\mathbf{M}} := \frac{1}{N}\sum_i (-1)^i T_{14}^\top \mathbf{S}_i = \begin{cases} \frac{1}{\sqrt{3}}(1,1,1), & \text{for triple-}\mathbf{q}\text{ order} \\ (0,0,1), & \text{for } z\text{-zigzag order} \end{cases}$$

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... 5 irreducible components of rank-2 tensor

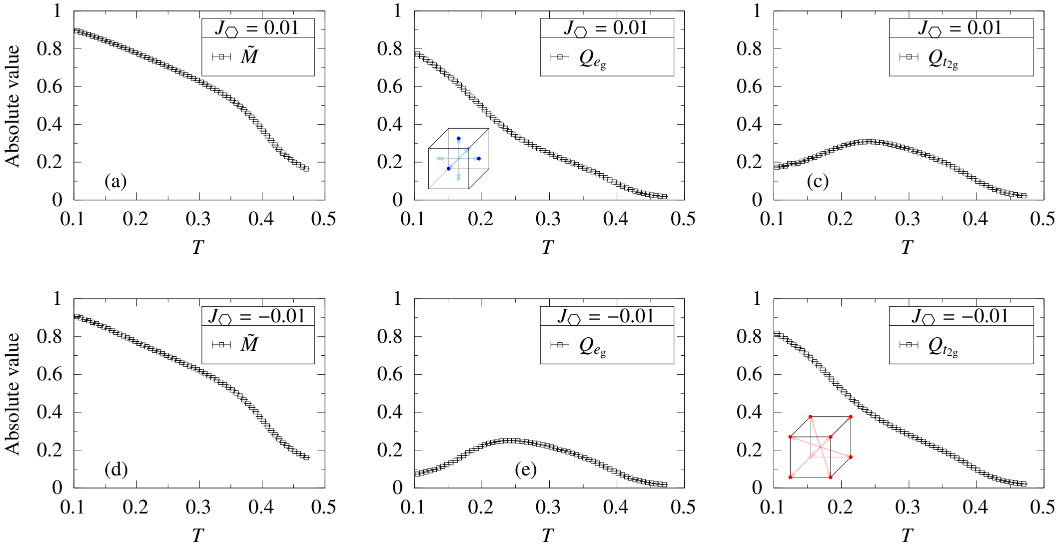
$$= \begin{cases} (0,1), & \text{for triple-}\mathbf{q}\text{ order} \\ (1,0), & \text{for zigzag order} \end{cases}$$

... measurable in finite-size simulations

Classical Monte Carlo simulations (L = 32)

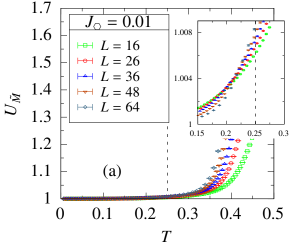
$J_{\hexagon} > 0$:

$J_{\hexagon} < 0$:

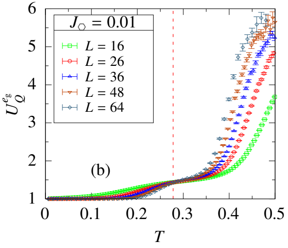


Critical temperatures

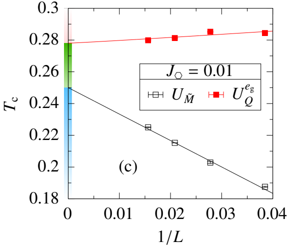
Primary Binder cumulant

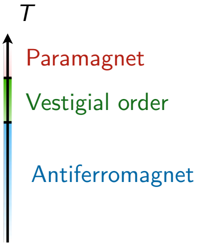


Composite Binder cumulant



Crossing points

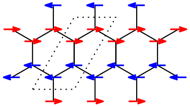
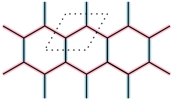
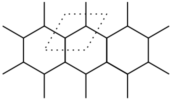




$$U_{\tilde{M}} = \frac{\langle \tilde{\mathbf{M}}^4 \rangle}{\langle \tilde{\mathbf{M}}^2 \rangle^2}$$

$$U_Q^{e_g} = \frac{\langle \mathbf{Q}_{e_g}^4 \rangle}{\langle \mathbf{Q}_{e_g}^2 \rangle^2}$$

Two-stage melting

$J_{\hexagon} > 0$:	Zigzag	$\mathbb{Z}_3$ spin nematic	Paramagnet
			
Rotation:	✗	✗	✓
Time reversal:	✗	✓	✓

Two-stage melting

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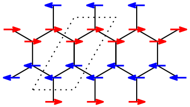
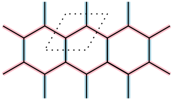
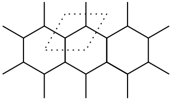
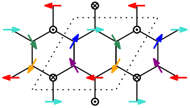
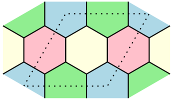
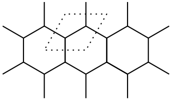
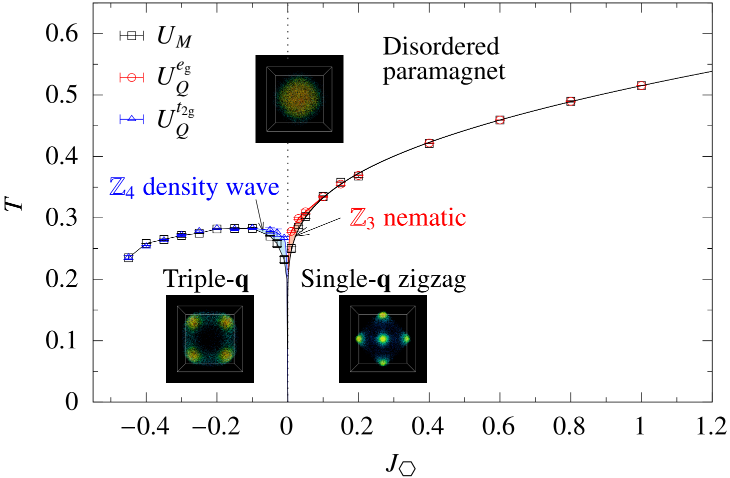
			
Rotation:	✗	✗	✓
Time reversal:	✗	✓	✓

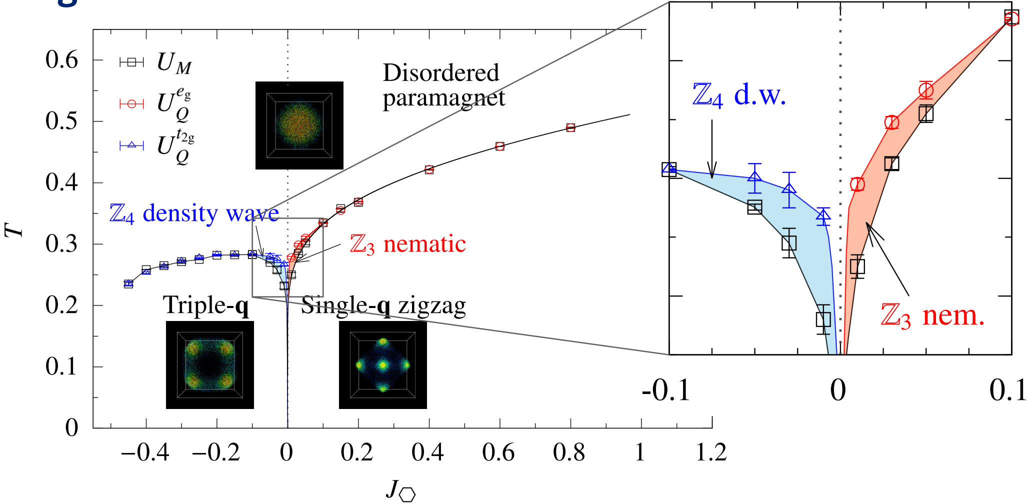
$J_{\hexagon} < 0$:	Triple-**q**	$\mathbb{Z}_4$ spin current density wave	Paramagnet
			
Rotation:	✓	✓	✓
Time reversal:	✗	✓	✓
Translation:	✗	✗	✓

Phase diagram: HKΓΓ’ model



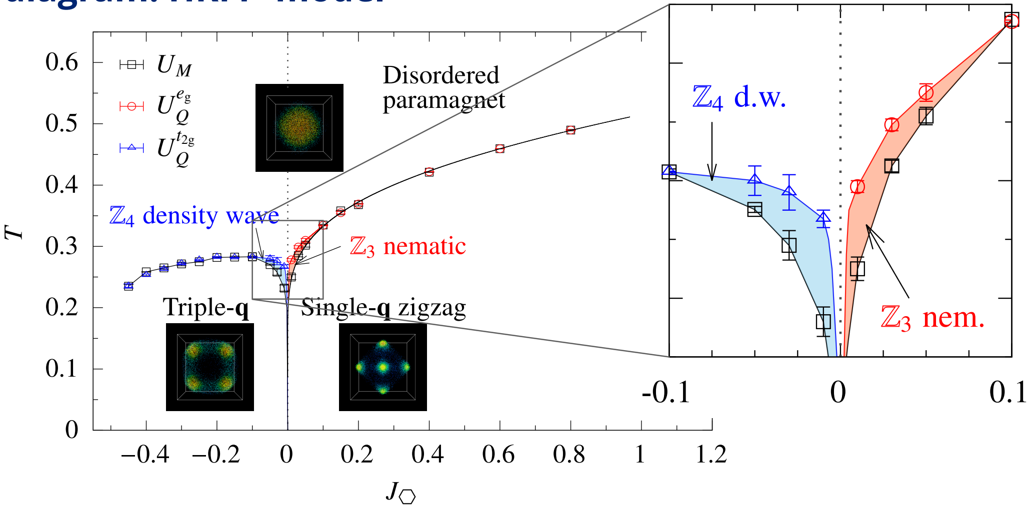
[Francini, LJ, PRB ’24]

Phase diagram: HKΓΓ’ model



[Francini, LJ, PRB ’24]

Phase diagram: HKΓΓ’ model



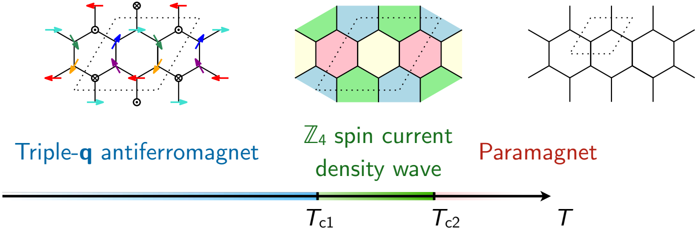
Message #2: Triple-**q** order can melt in two stages

… same is true for zigzag order

[Francini, LJ, PRB ’24]

Phase diagram: $Na_2Co_2TeO_6$

Theory:



[Francini, LJ, PRB ’24]

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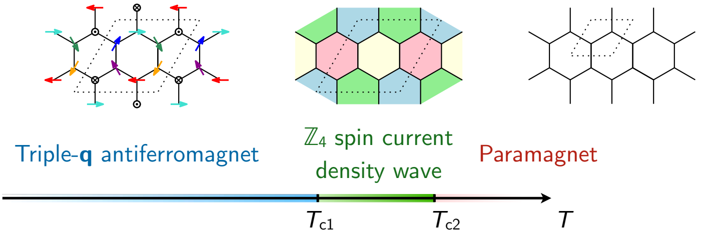
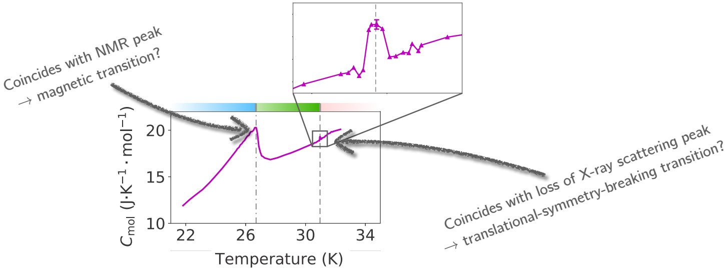


[Francini, LJ, PRB '24]

Experiment:



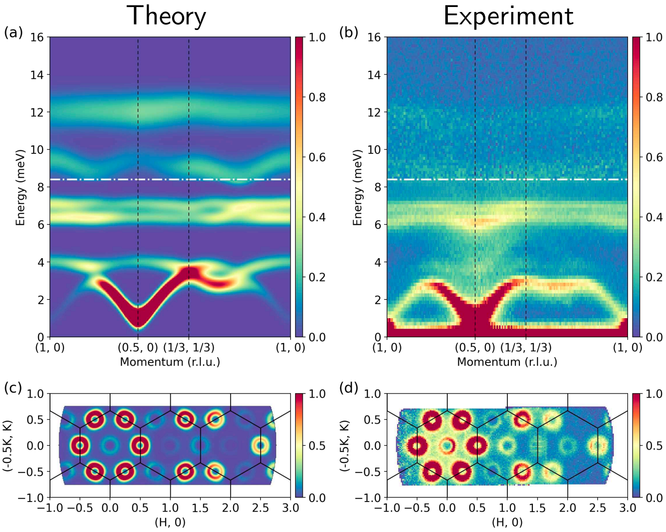
[Chen *et al.*, PRB '21]

$Na_2Co_2TeO_6$: Magnon excitation spectrum

$$(J, K, \Gamma, \Gamma') = (1.2, -8.3, 1.9, -2.3, 0.5)\,\text{meV}$$

$$(J_3, J_2^A, J_2^B) = (1.5, 0.32, -0.24)\,\text{eV}$$

$$+\ \text{nonbilinear exchange}$$



[Krüger, Chen, Jin, Li, LJ, PRL '23]

Candidate Kitaev magnets: Effective spin models

Minimal HKΓΓ' models:

Material	Reference	$\mathcal{H}_{\text{Kitaev}}$	$\mathcal{H}_{\text{H}\Gamma\Gamma'}$						α_{c}
		K_1 (meV)	J_1 (meV)	Γ_1 (meV)	Γ'_1 (meV)	J_2^A (meV)	J_2^B (meV)	J_3 (meV)	
$Na_2Co_2TeO_6$	this work	-8.29	1.23	1.86	-2.27	0.32	-0.24	0.47	0.135
Na_2IrO_3	[21]	-17.00	–	–	–	–	–	6.80	0.086
α-$RuCl_3$	[22]	-5.00	-0.50	2.50	–	–	–	0.50	0.175

$Na_2Co_2TeO_6$: [Krüger, Chen, Jin, Li, LJ, PRL '23]

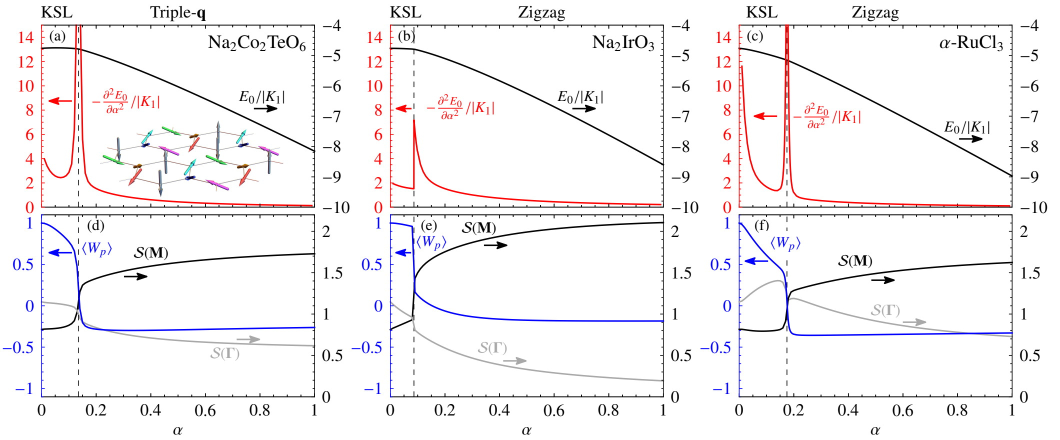
Na_2IrO_3: [Winter *et al.*, PRB '16]

α-$RuCl_3$: [Winter *et al.*, Nat. Commun. '17]

Candidate Kitaev magnets: Proximity to quantum spin liquid

1-parameter family of Hamiltonians: $\mathcal{H}(\alpha) = \mathcal{H}_{\text{Kitaev}} + \alpha \mathcal{H}_{\text{H}\Gamma\Gamma'}$

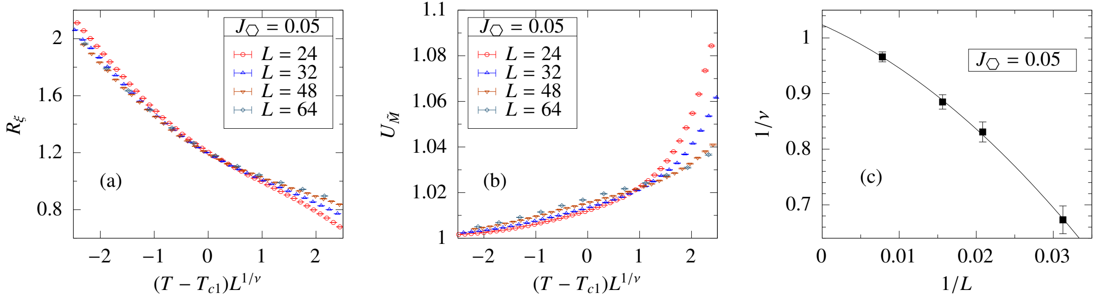
Phase diagrams:



[Krüger, Chen, Jin, Li, LJ, PRL '23]

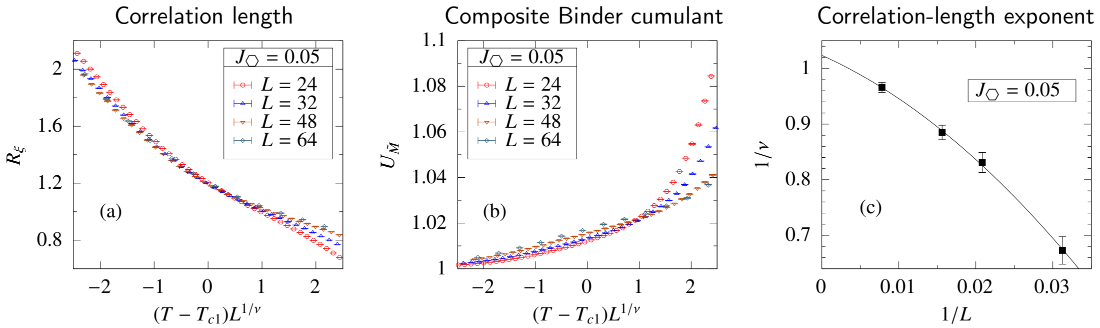
Critical scaling @ T_{c1} (primary-to-vestigial)

Correlation length | Composite Binder cumulant | Correlation-length exponent



$$R_\xi = \xi/L$$

Critical scaling @ T_{c1} (primary-to-vestigial)

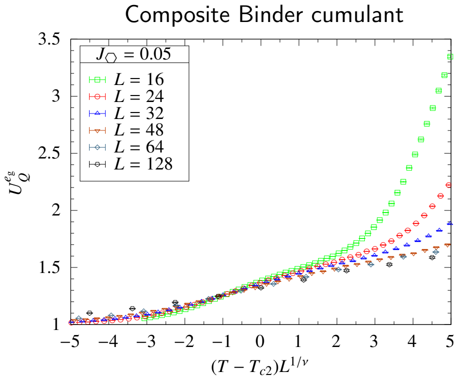


$R_\xi = \xi/L$

→ 2D Ising universality

... consistent with $\mathbb{Z}_2$ time reversal symmetry breaking

Critical scaling @ T_{c2} (vestigial-to-paramagnet)

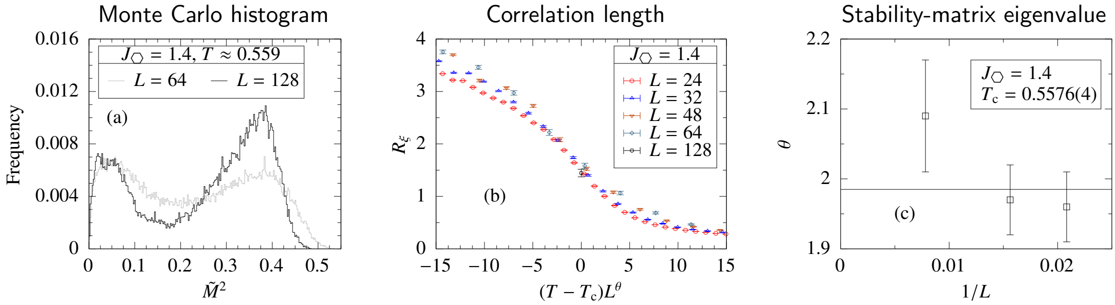


with $1/\nu = 6/5$

$\rightarrow$ 2D three-state Potts universality

… consistent with $\mathbb{Z}_3$ symmetry breaking

Phase coexistence @ primary-to-disorder transition

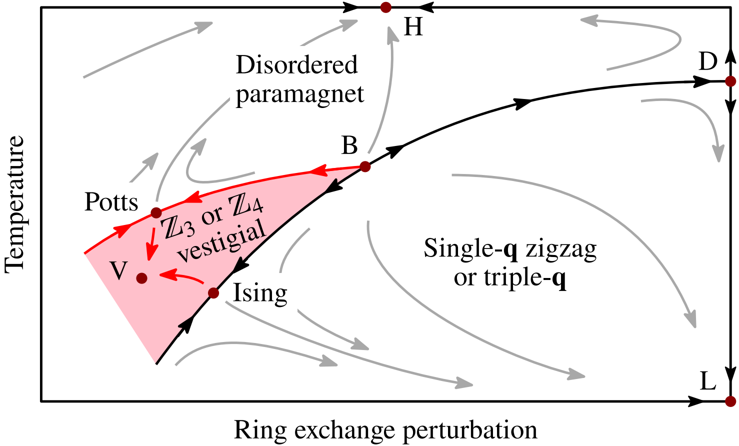


$\theta = 1.99(3) = d$

... spatial dimension

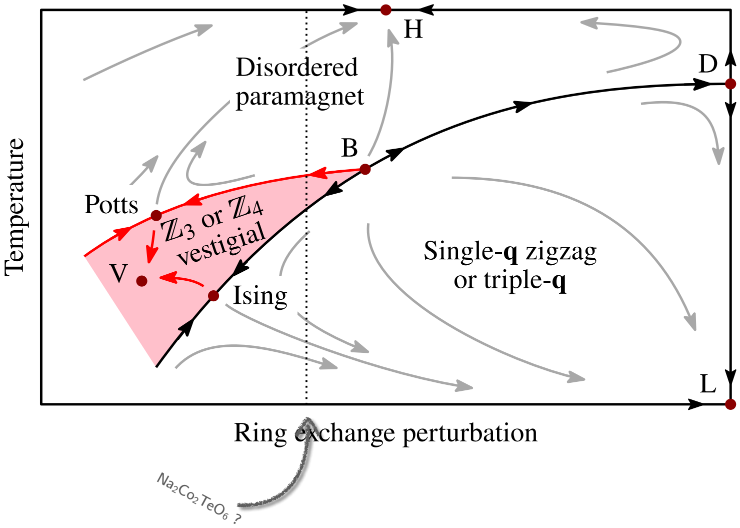
→ First-order transition from discontinuity fixed point

Schematic RG flow



[Francini, LJ, PRB ’24]

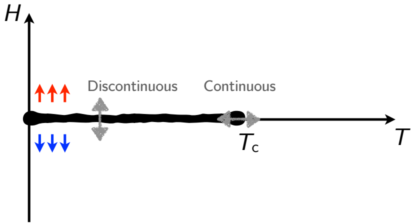
Schematic RG flow

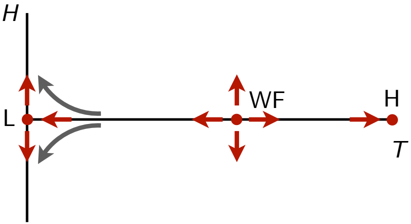


[Francini, LJ, PRB '24]

Digression: Discontinuity fixed point

Ising model in longitudinal field:





Flow linearization (relevant direction):

$$\beta_H = -\theta H + \mathcal{O}(H^2) \qquad \text{with} \qquad \theta = d = 2$$

Formal limit of continuous transition:

$$\begin{array}{lll} U \propto |t|^{1-\alpha} & & \alpha \to 1 \\ & \text{with} & \\ M \propto (-t)^{\beta} \quad (t < 0) & & \beta \to 0 \end{array} \qquad \Rightarrow \qquad 1/\nu \to d = 2$$

[Nienhuis, Nauenberg, PRL '75]

High-spin d^7 Mott insulators

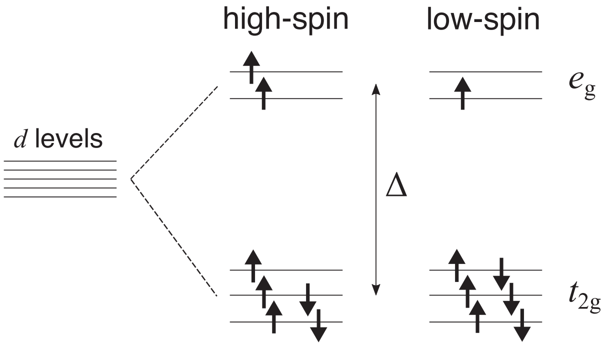


FIG. 1. Atomic d levels splitting into two groups under the octahedral CEF Δ: e_g levels at a higher energy and t_{2g} levels at a lower energy. The d^7 electron configuration can take either high-spin (middle) or low-spin state (right), depending on the strength of Coulomb interactions and Δ.

[Sano, Kato, Motome, PRB '18]