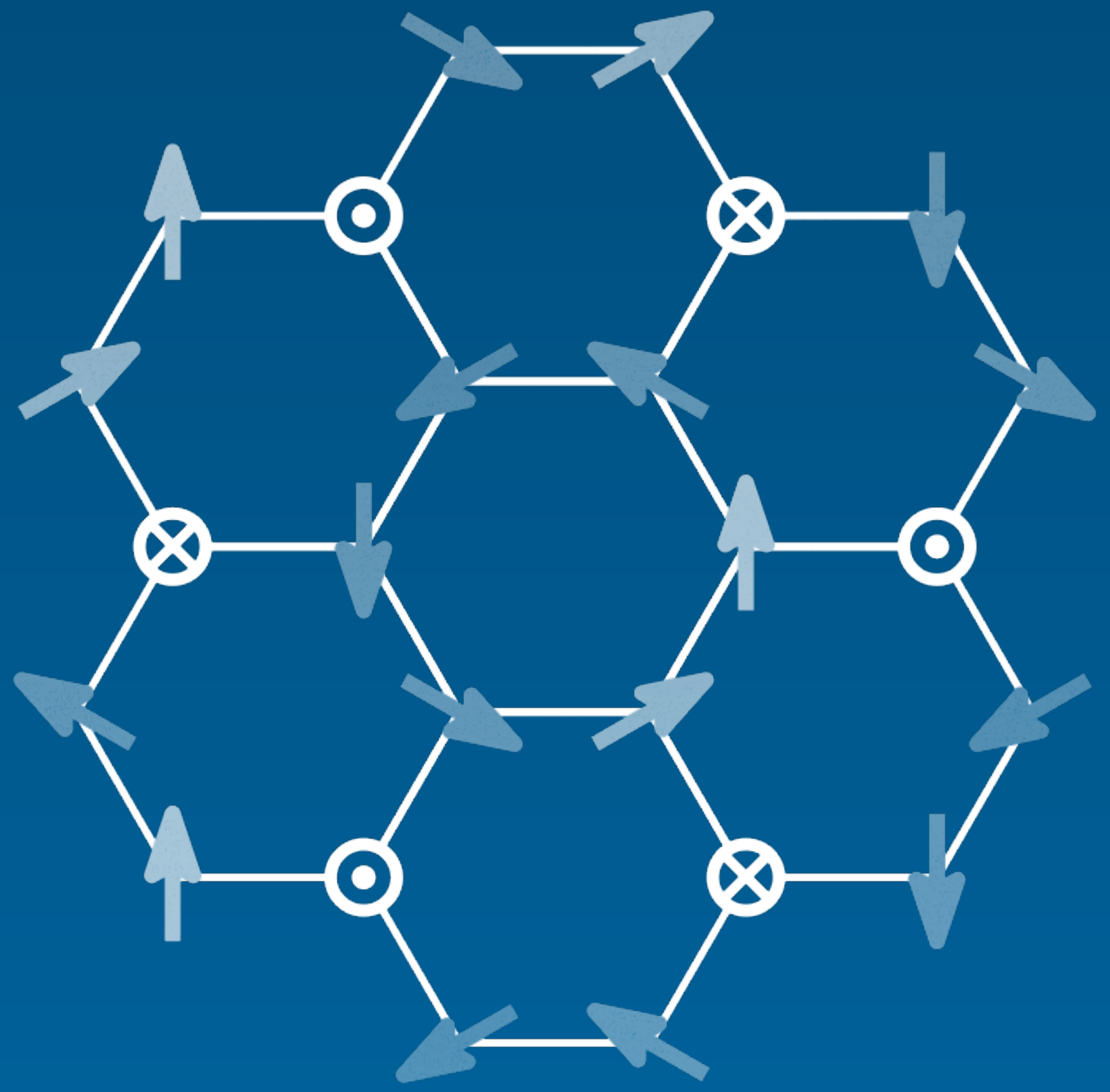
TECHNISCHE UNIVERSITÄT DRESDEN

DRESDEN concept



Ferrimagnetism from triple-q order in $Na_2Co_2TeO_6$

Lukas Janssen

Niccolò Francini

Pedro M. Cônsoli

Emmy Noether-Programm

DFG Deutsche Forschungsgemeinschaft

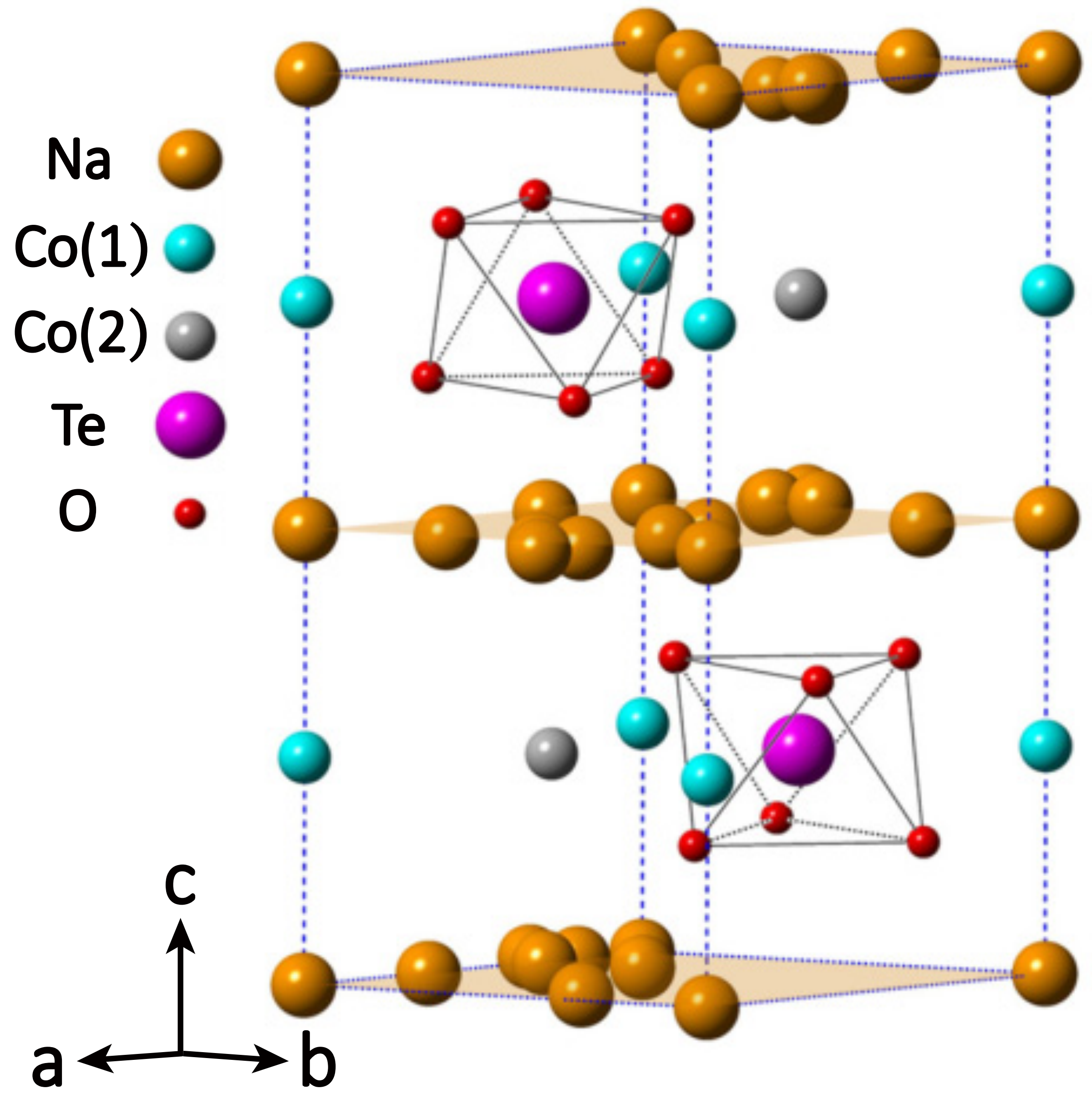
N. Francini, LJ, PRB **110**, 235118 (2024)

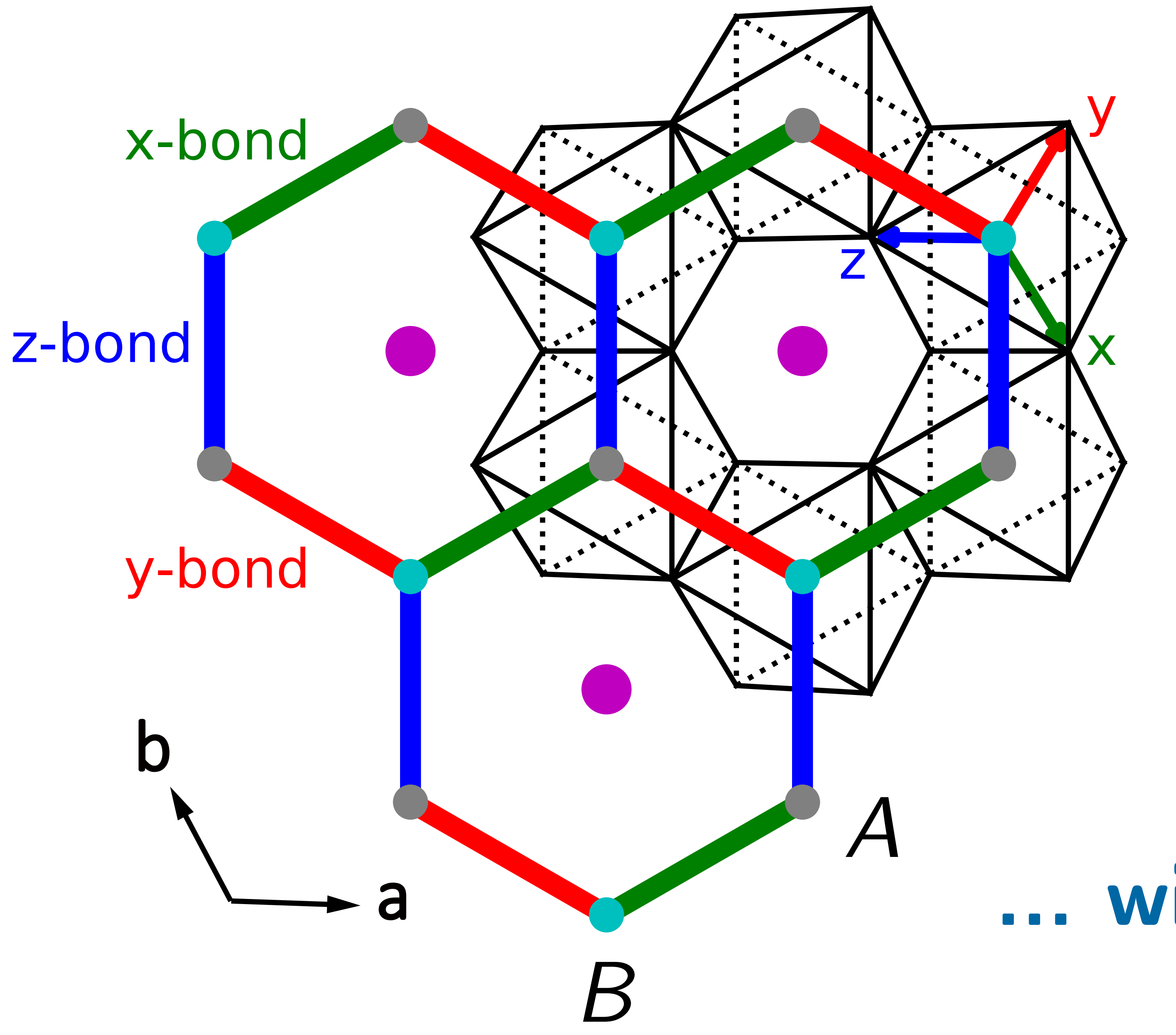
N. Francini, P. M. Cônsoli, LJ, *in preparation*

ct.qmat

Complexity and Topology in Quantum Matter

Magnetic Mott insulator $Na_2Co_2TeO_6$

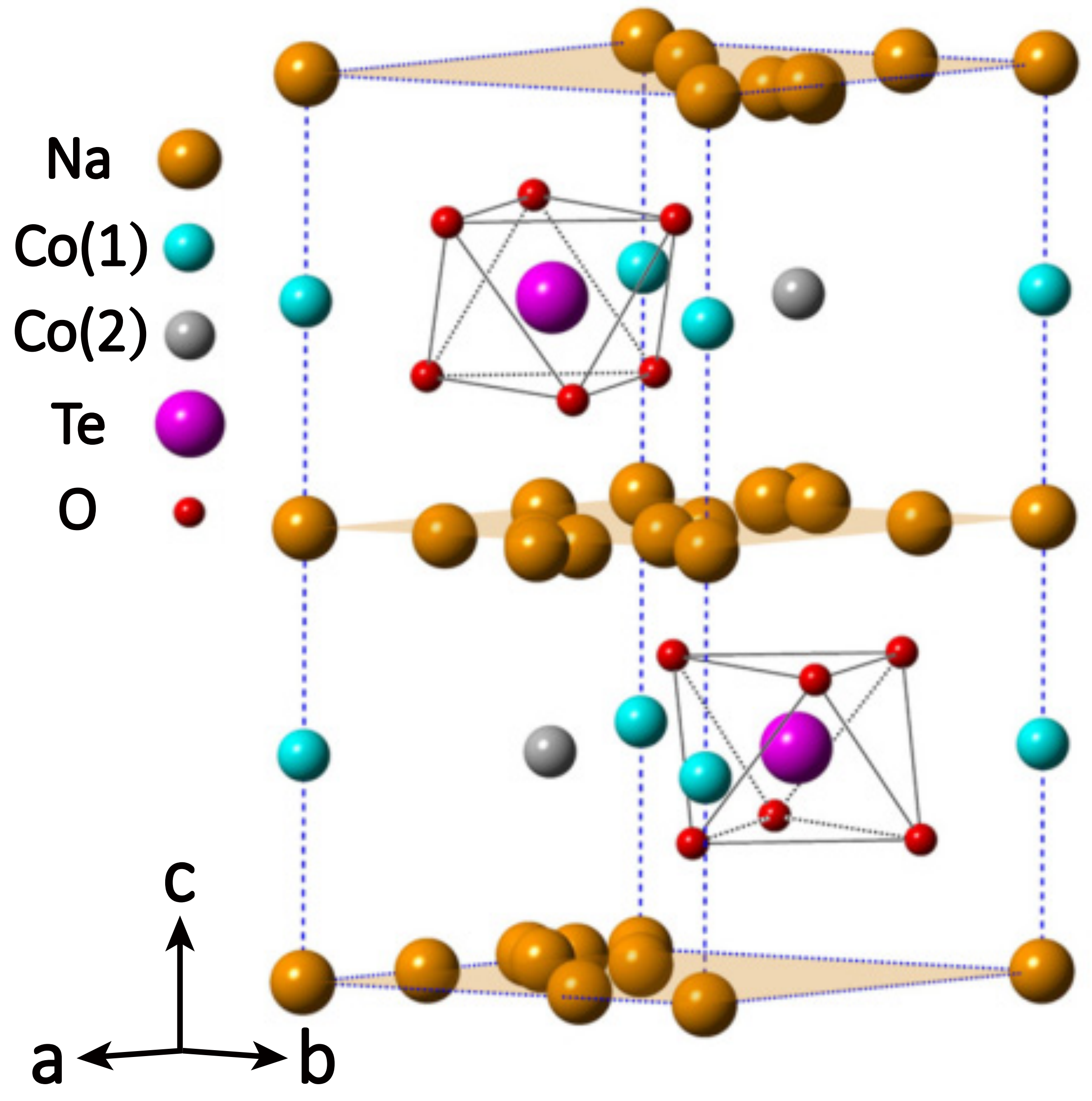


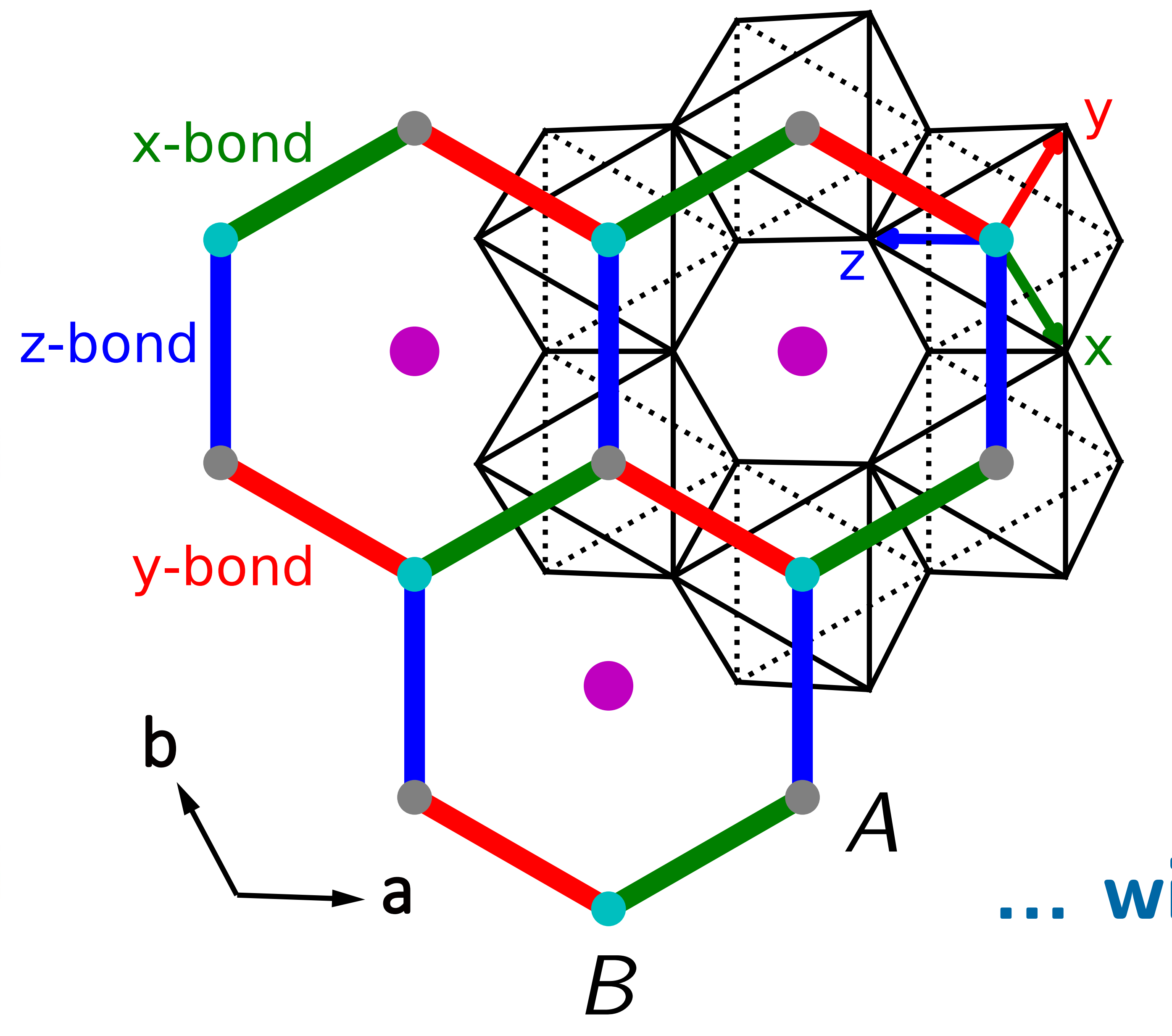


… with broken sublattice symmetry, $A \neq B$

[Yao, Li, PRB '20]

Magnetic Mott insulator $Na_2Co_2TeO_6$

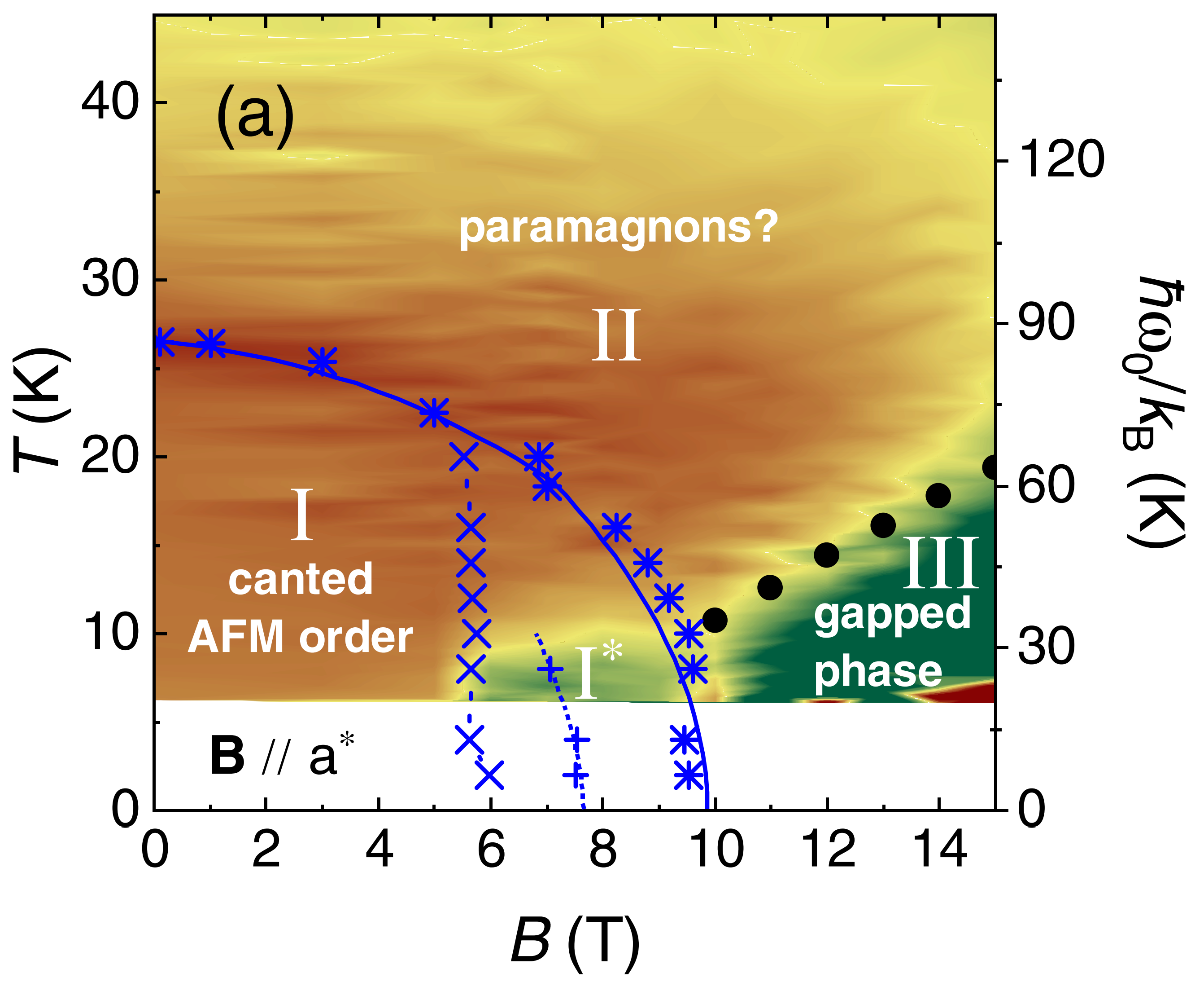


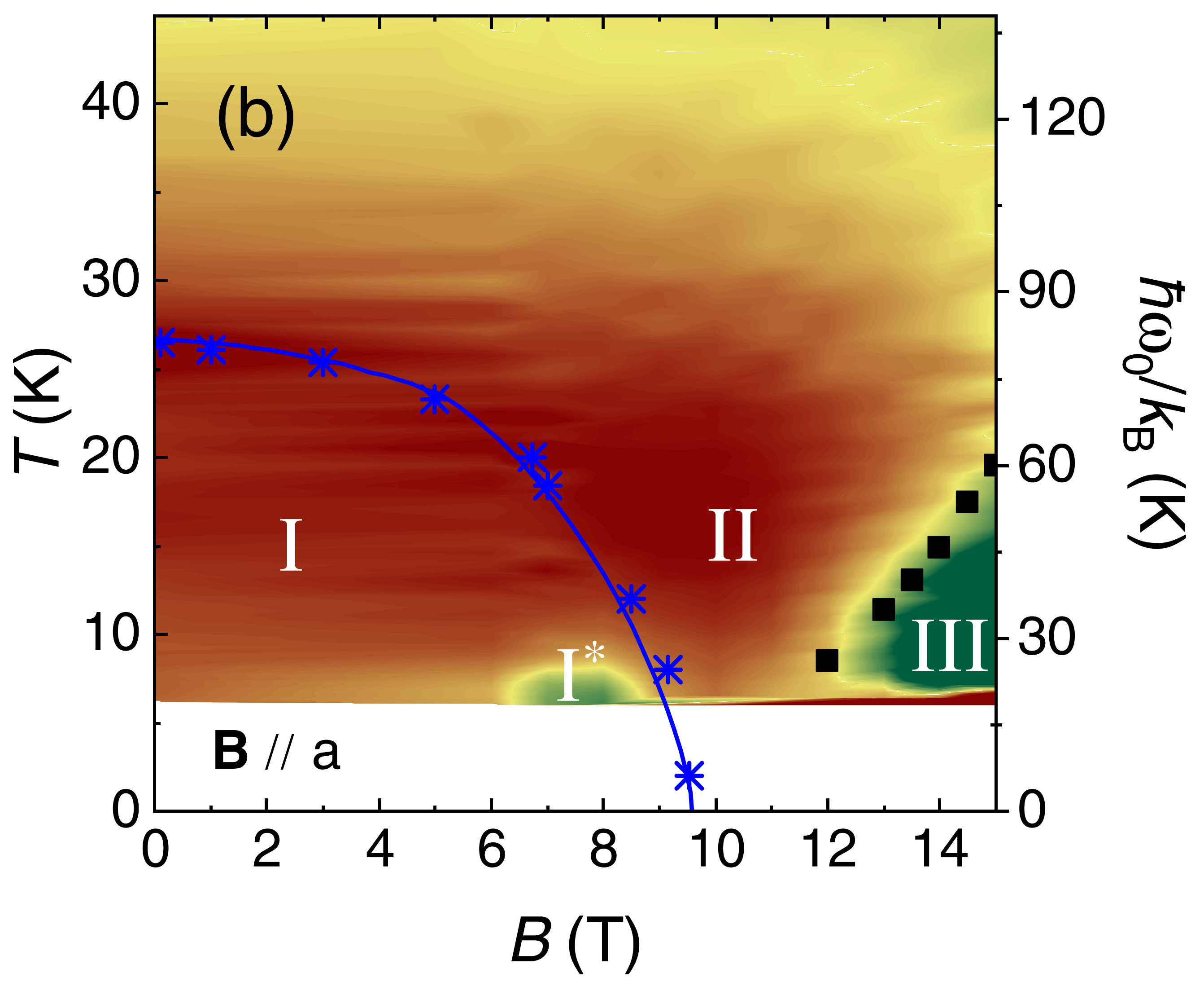


... with broken sublattice symmetry, *A* ≠ *B*

[Yao, Li, PRB '20]

Phase diagram:





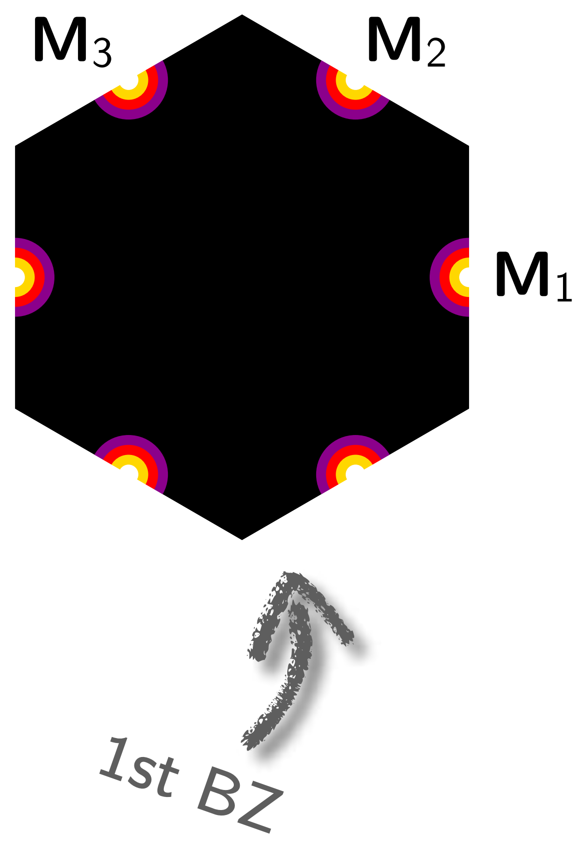
[Hong *et al.*, PRB '21]

Single-q vs multi-q order: Bragg peaks

Static spin structure factor:

... e.g., from neutron diffraction

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



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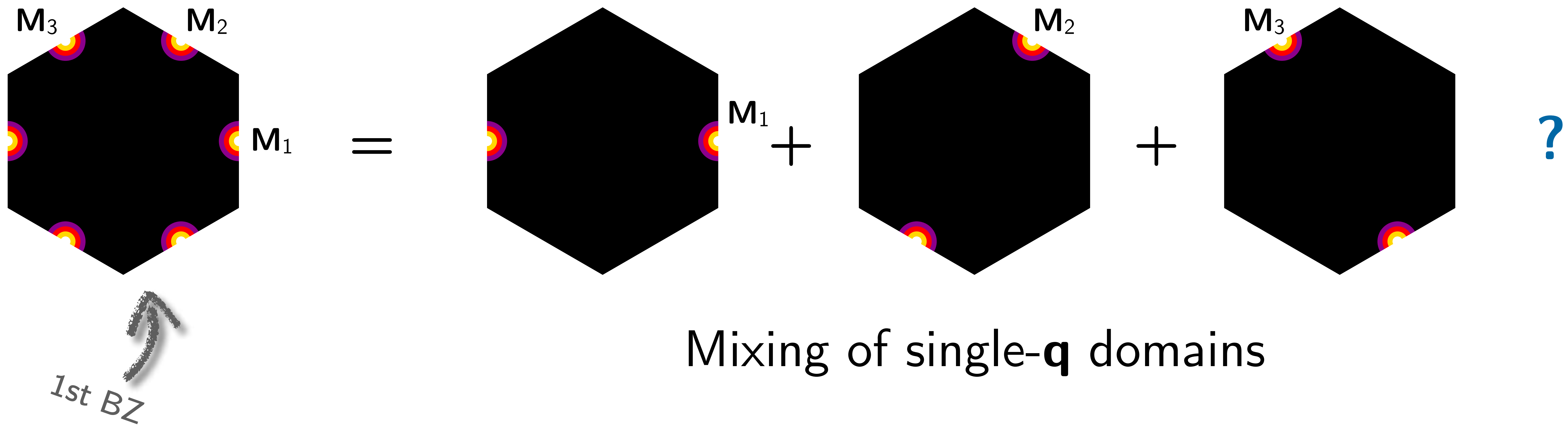

Mixing of single-**q** domains

Single-q vs multi-q order: Bragg peaks

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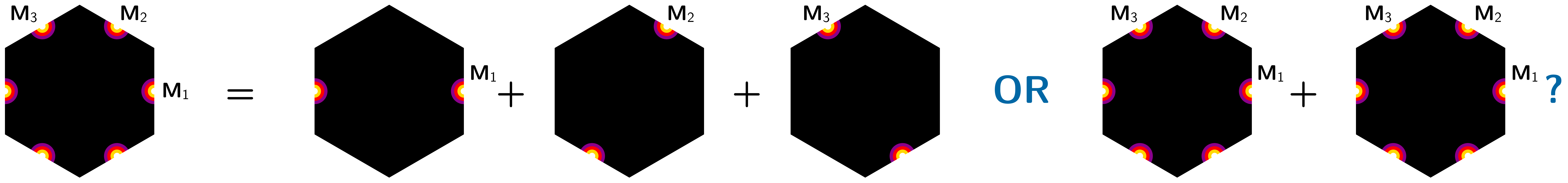

Mixing of single-**q** domains

True triple-**q** order

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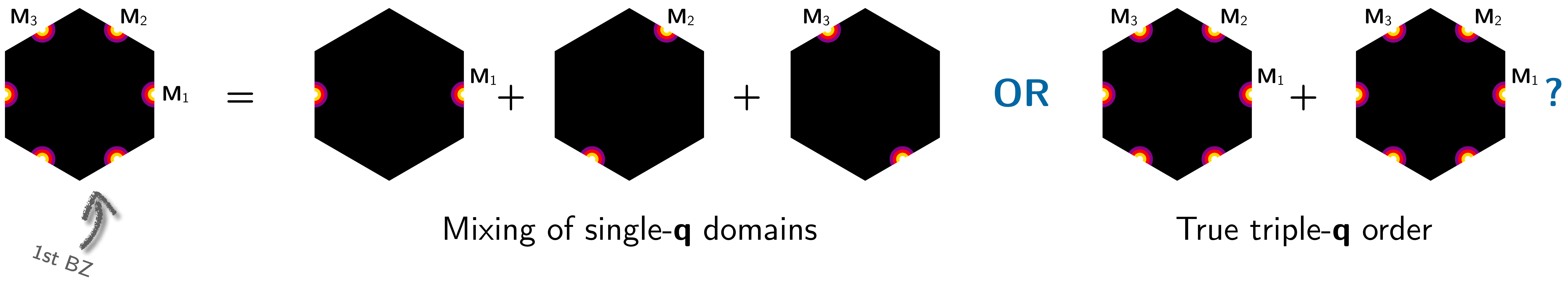
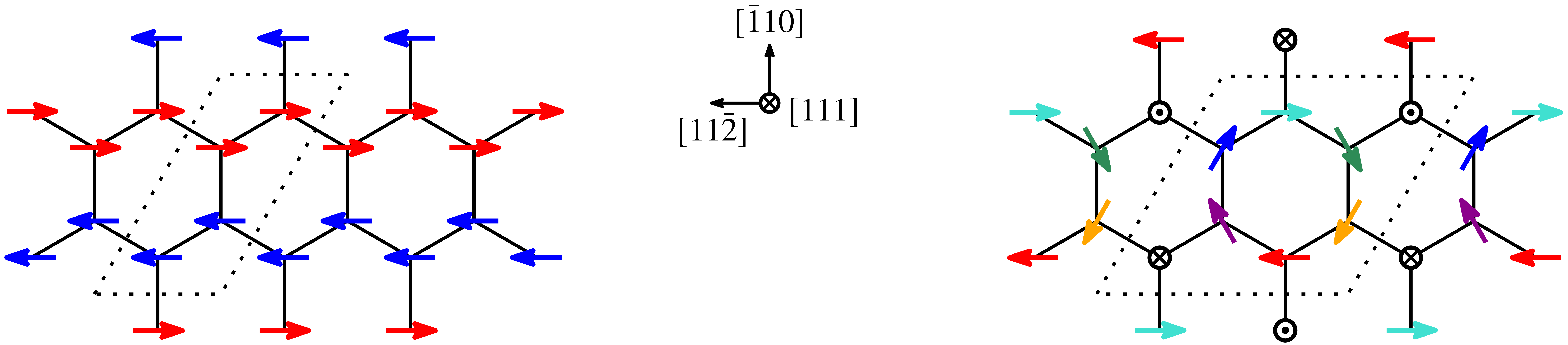


Mixing of single-**q** domains

True triple-**q** order

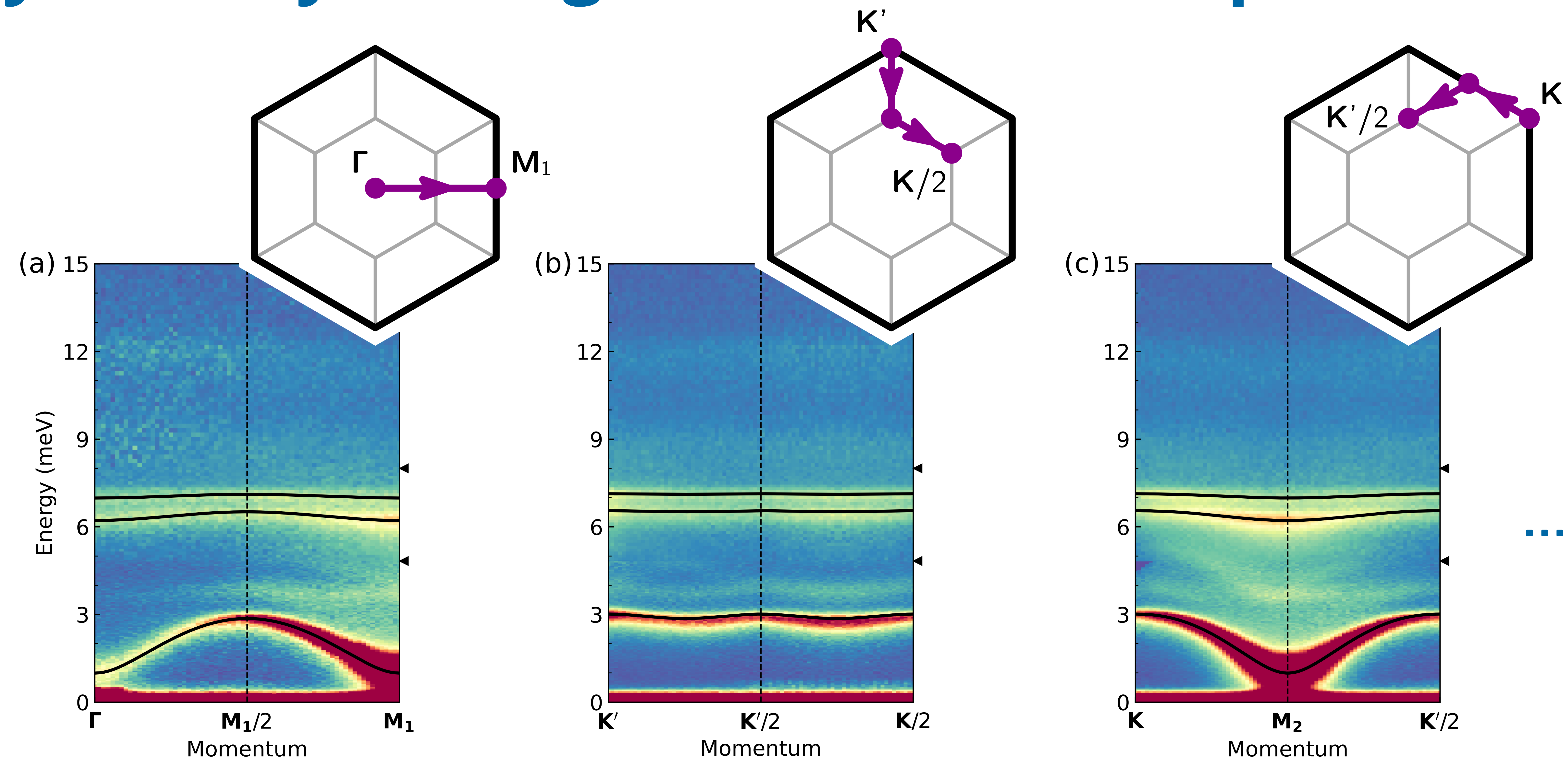
Real space:



α-$RuCl_3$, Na_2IrO_3, ...
[Choi *et al.*, PRL '12]
[Johnson *et al.*, PRB '15]
[Balz *et al.*, PRB '21]

Kitaev-Heisenberg in field: [LJ *et al.*, PRL '16]
Bilinear-Biquadratic Kitaev-Heisenberg: [Pohle *et al.*, PRB '23]

Symmetry in magnetic excitation spectrum



... perfectly symmetric magnon bands

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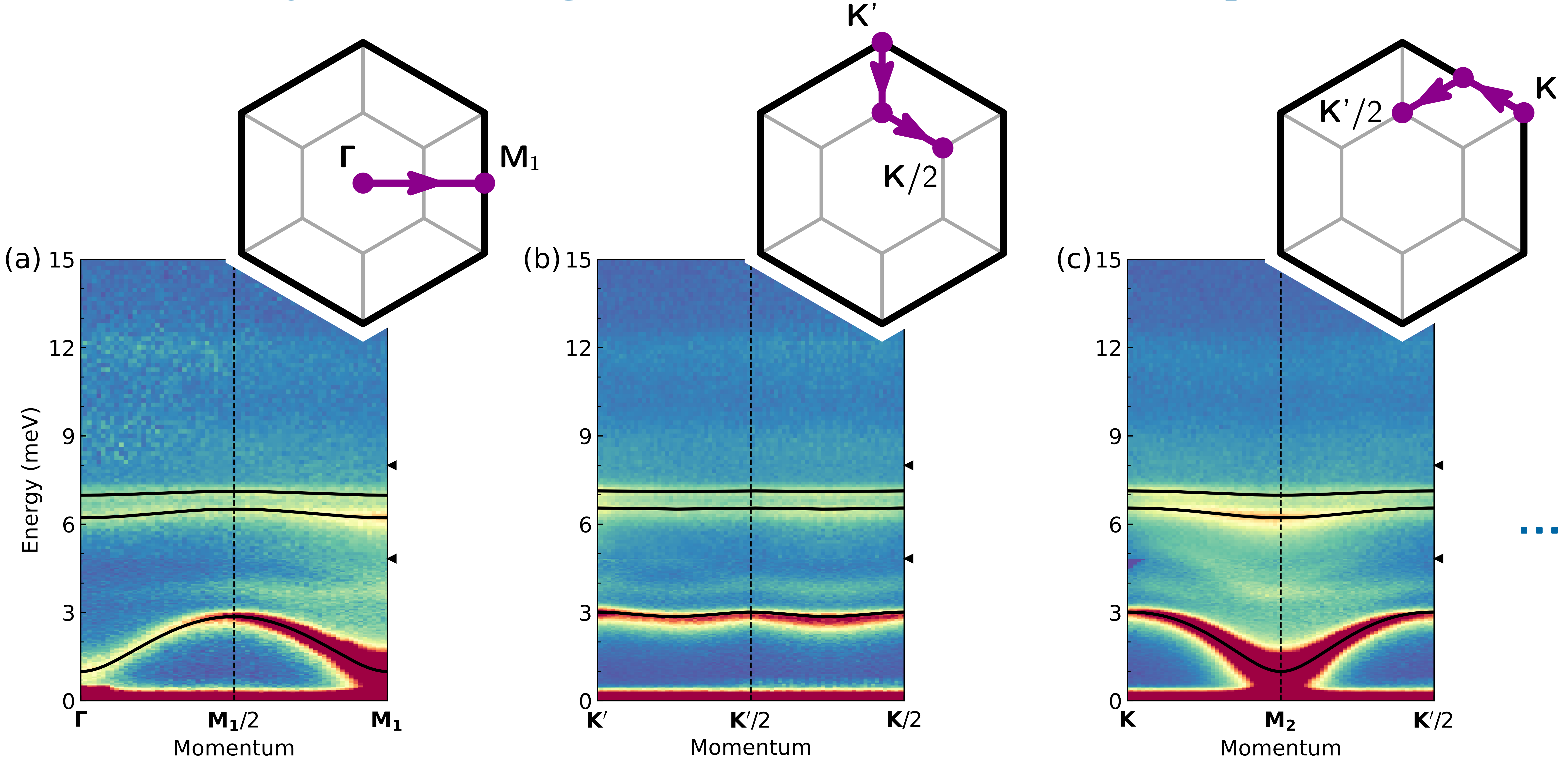
[Krüger, Chen, Jin, Li, LJ, PRL '23]

Symmetry in magnetic excitation spectrum



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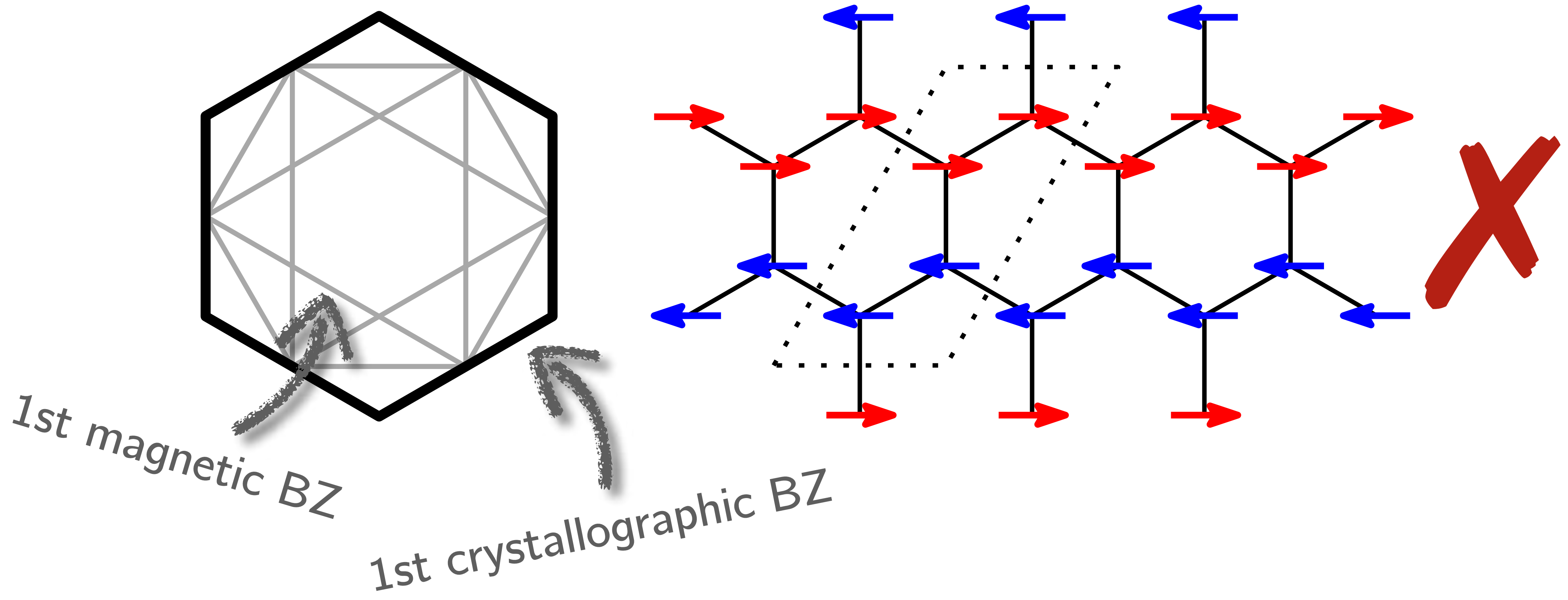
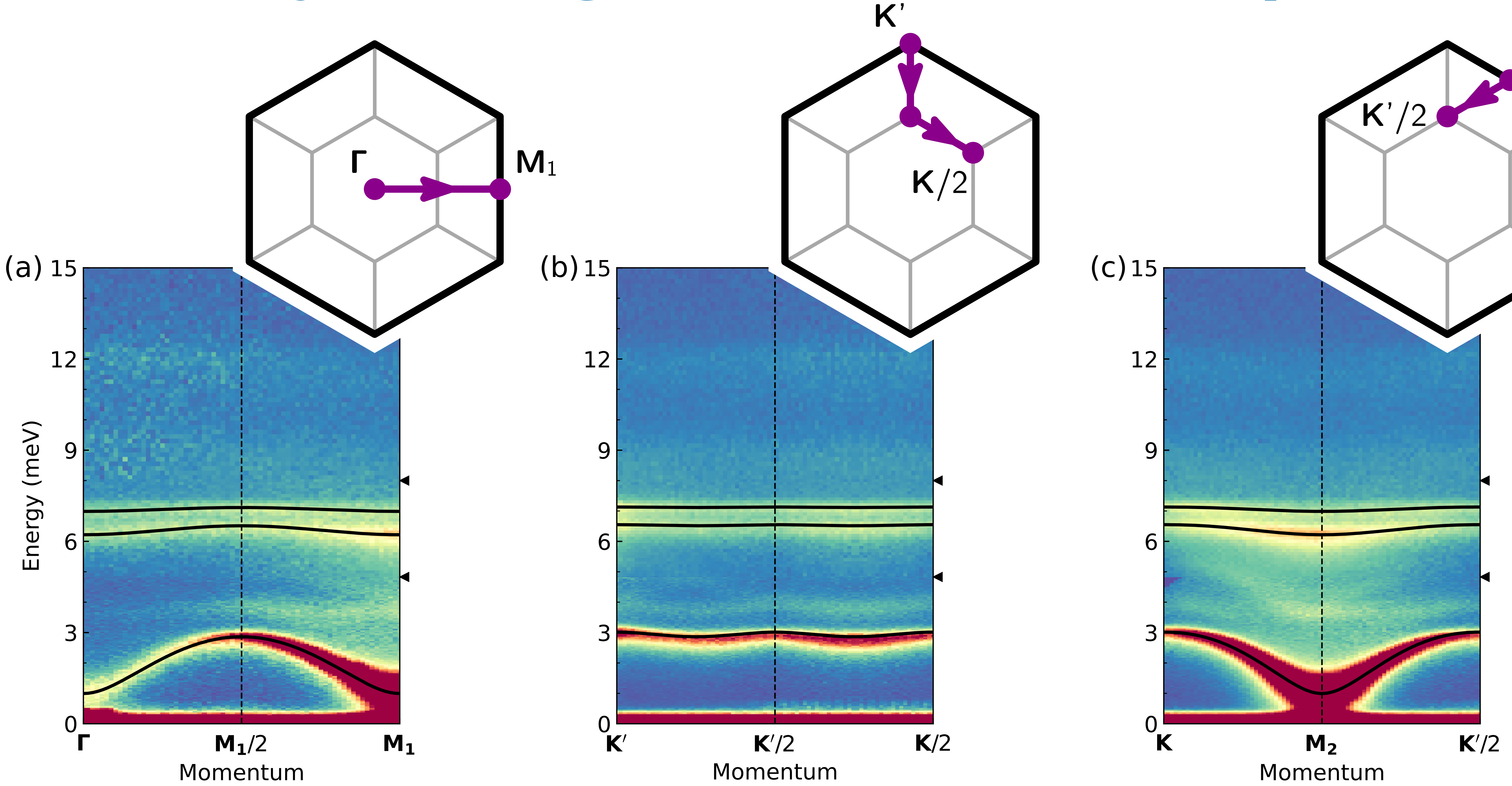
Mixing of zigzag domains



[Krüger, Chen, Jin, Li, LJ, PRL '23]

Symmetry in magnetic excitation spectrum



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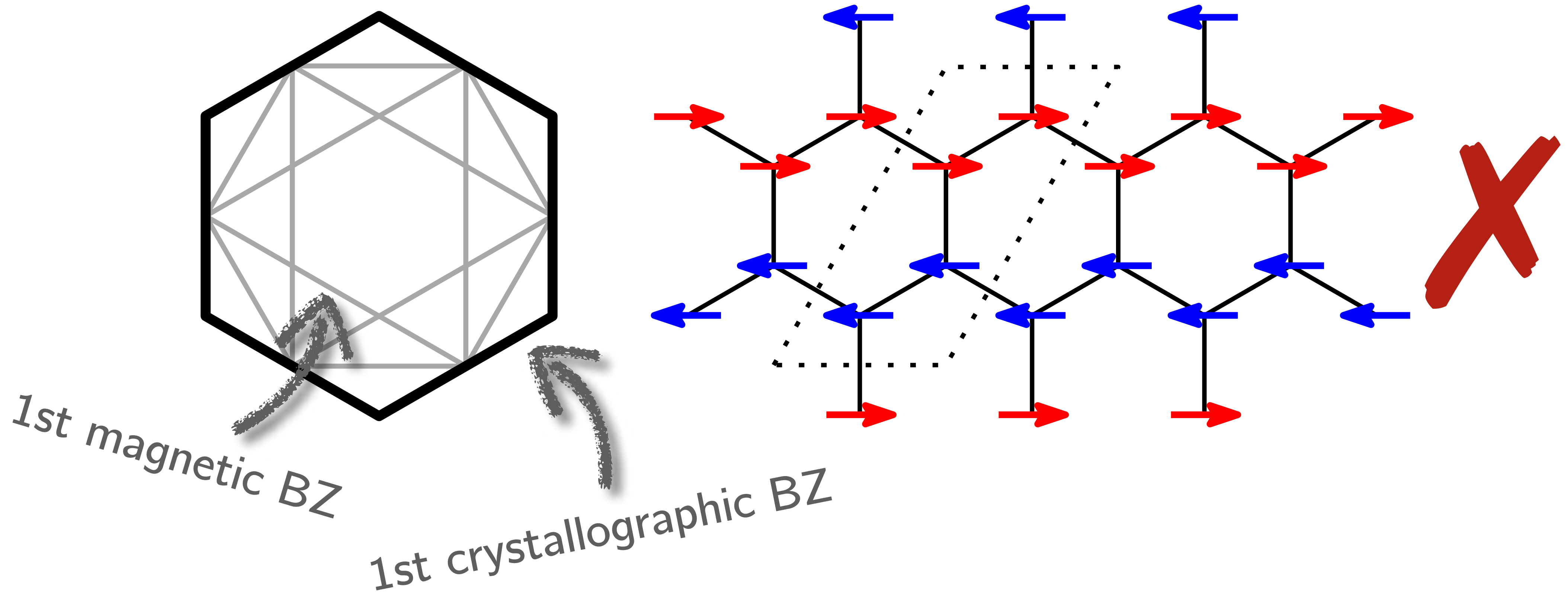
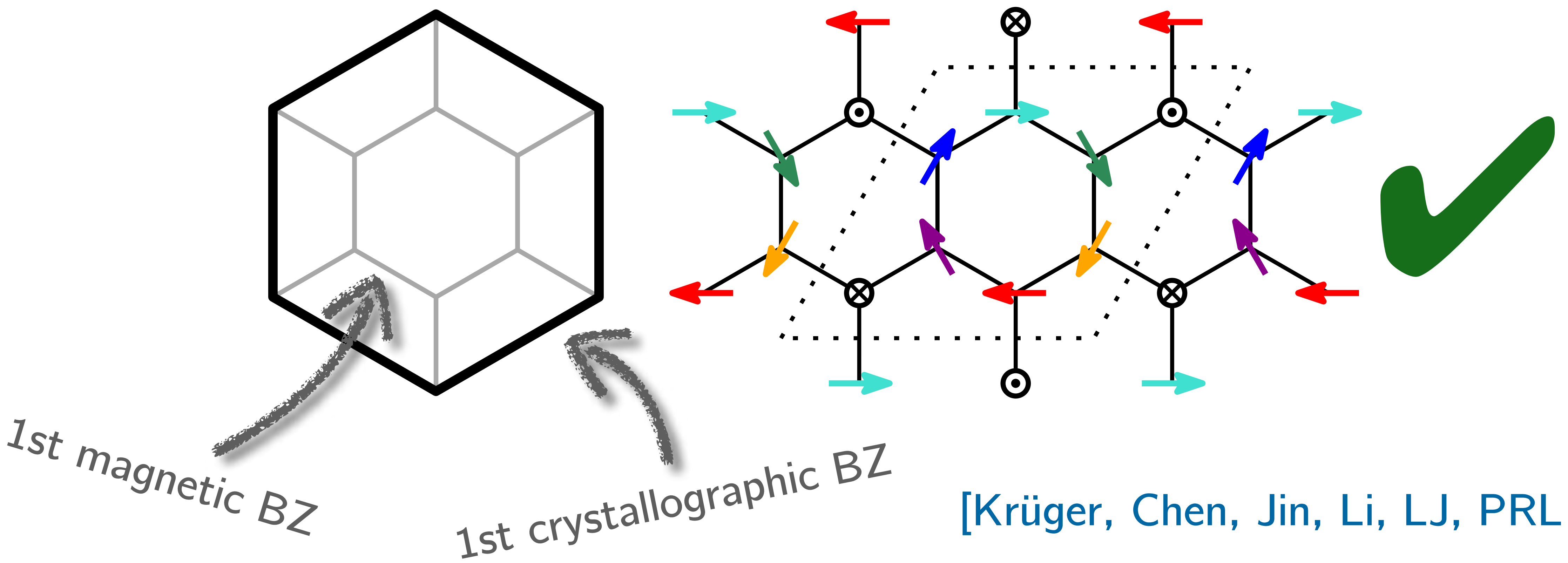


True triple-**q** order



[Krüger, Chen, Jin, Li, LJ, PRL '23]

Toroidal moment

Toroidal moment (“spin vorticity”):

$$\vec{t} = \frac{1}{N} \sum_i \vec{r}_i \times \vec{S}_i$$

... odd under inversion & time reversal

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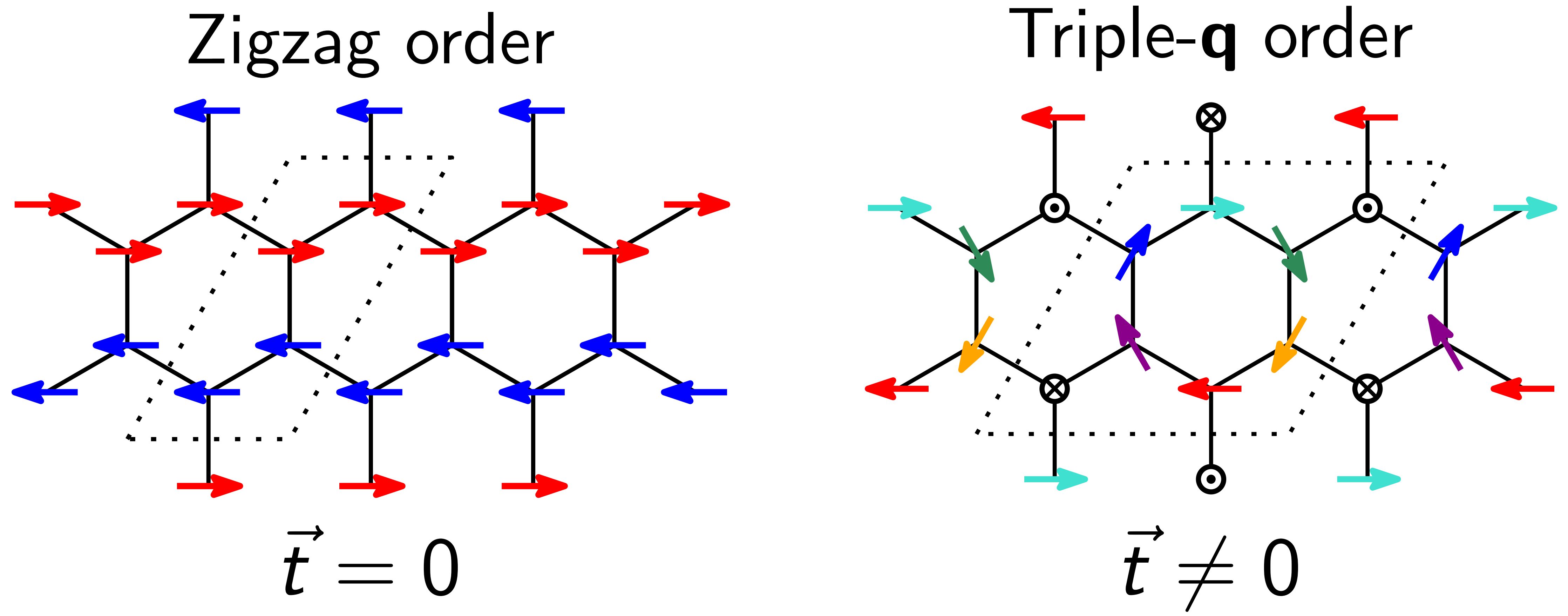

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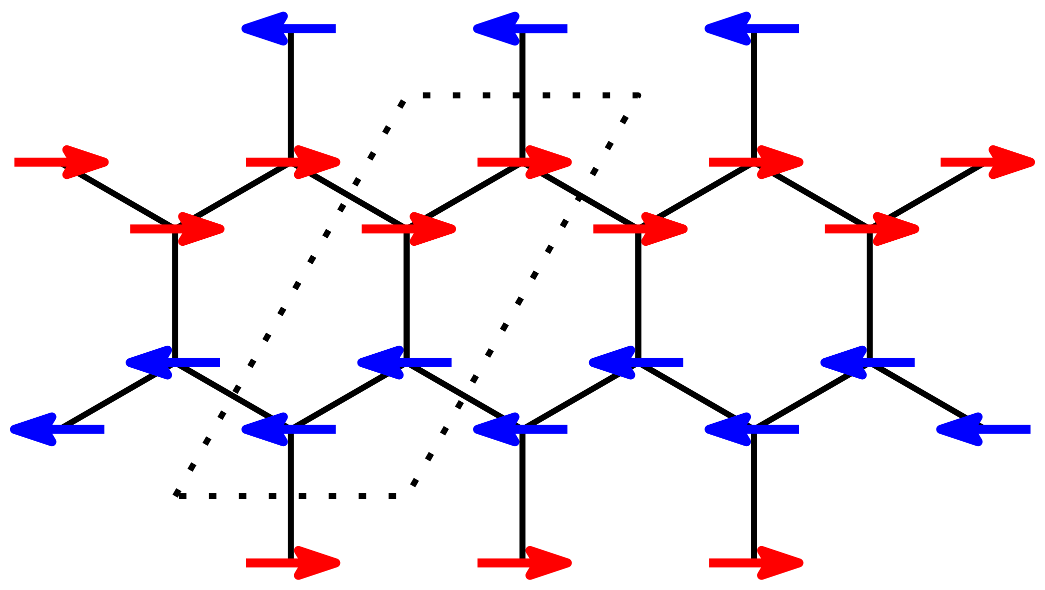
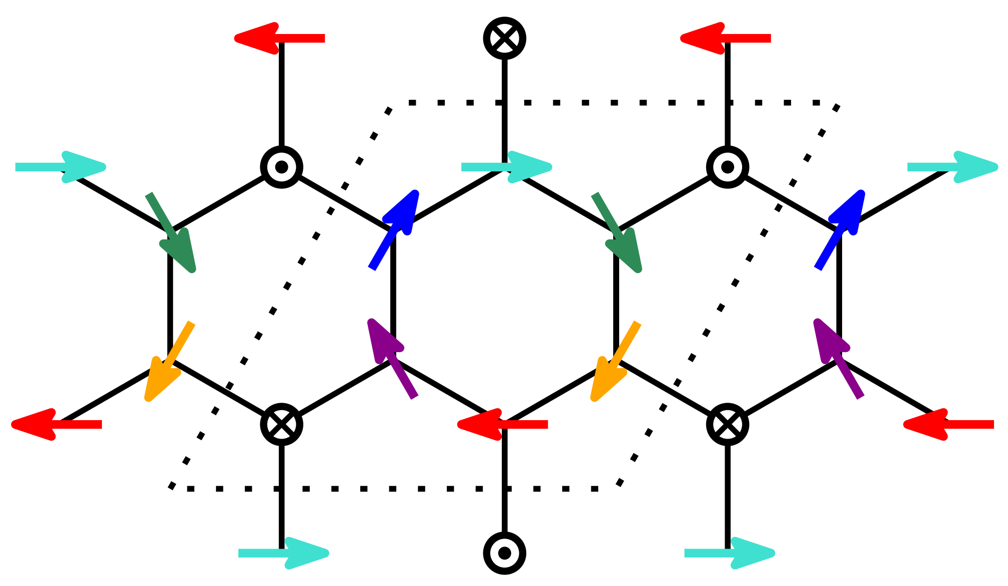
Zigzag order



$\vec{t} = 0$

Triple-**q** order



$\vec{t} \neq 0$

Electric polarization:

$$P_i = \alpha_{ij} H_j + \mathcal{O}(H^2, E)$$

magnetoelectric coupling

with $\alpha_{ij} \neq 0$ for $\vec{t} \neq 0$

[Spaldin *et al.*, JPCM '08]

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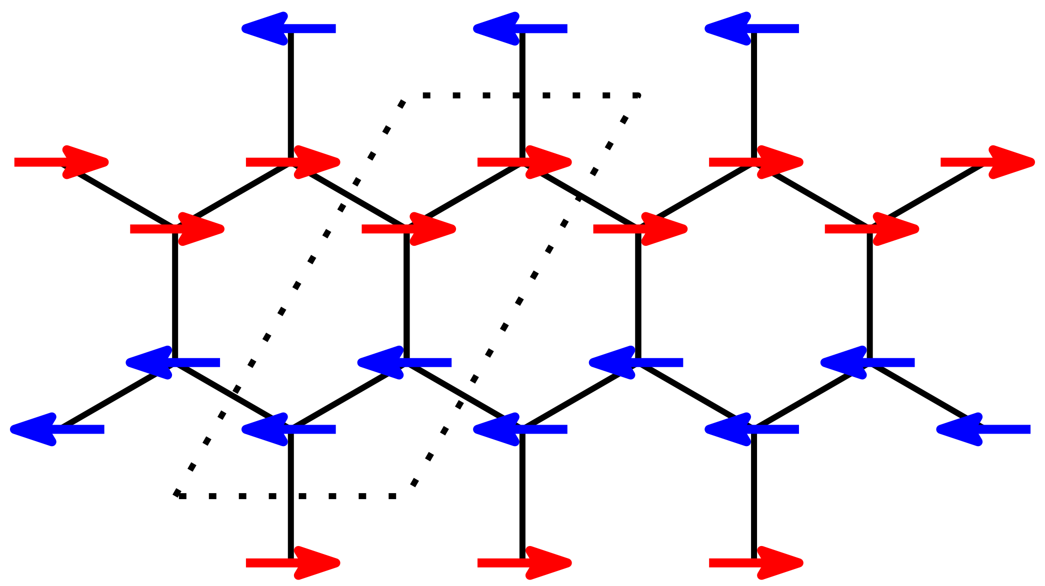

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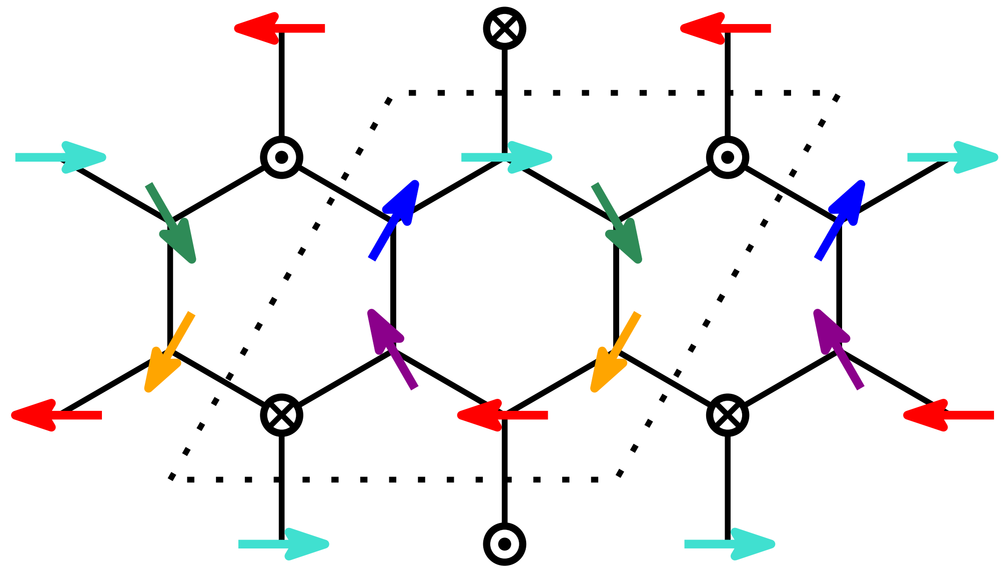
Triple-**q** order



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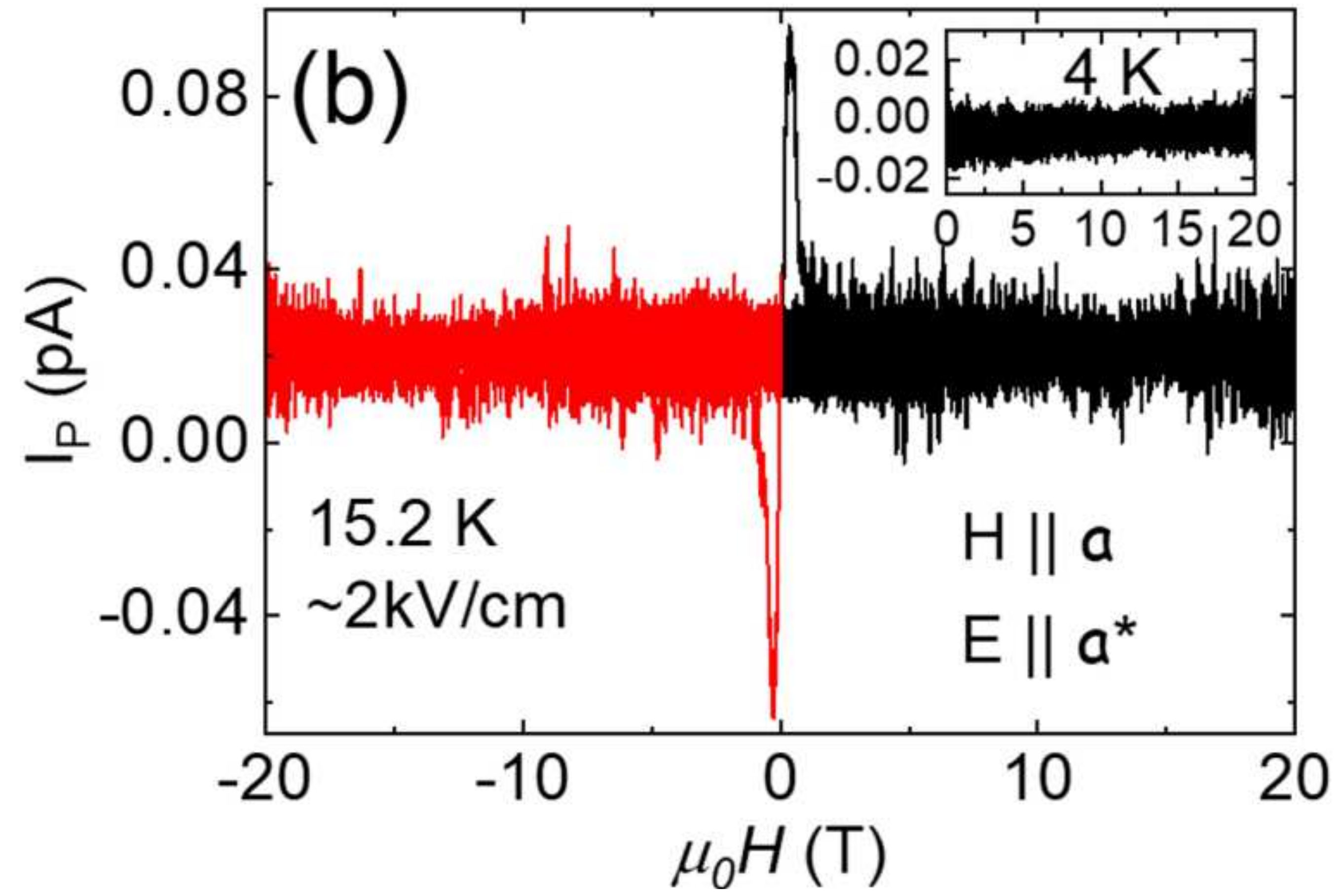
[Spaldin *et al.*, JPCM '08]

Magnetoelectric current:

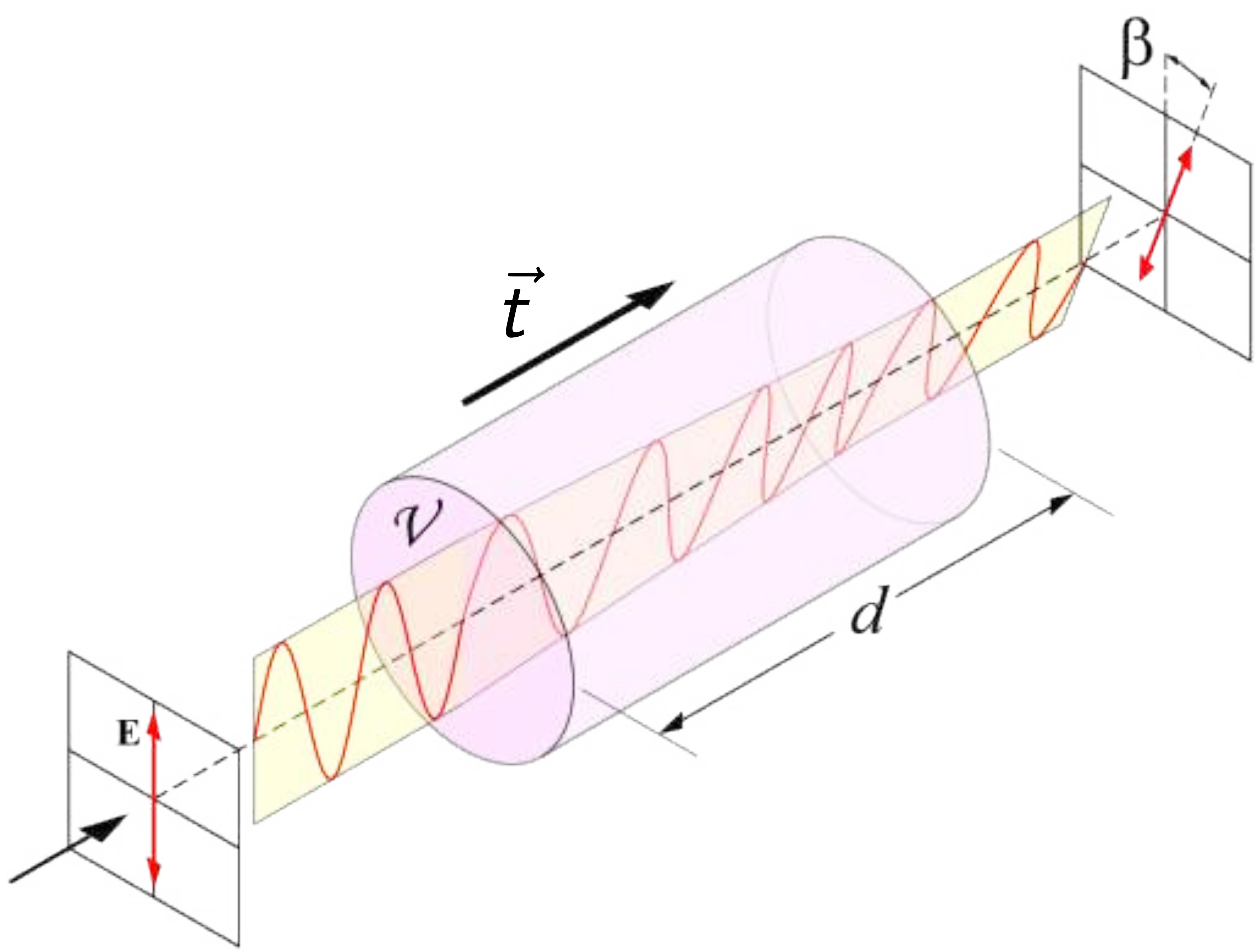


$\Rightarrow \quad \alpha_{ij} = 0$?

[Zhang *et al.*, PRB '23]

Toroidal moment from Faraday rotation

Faraday rotation:

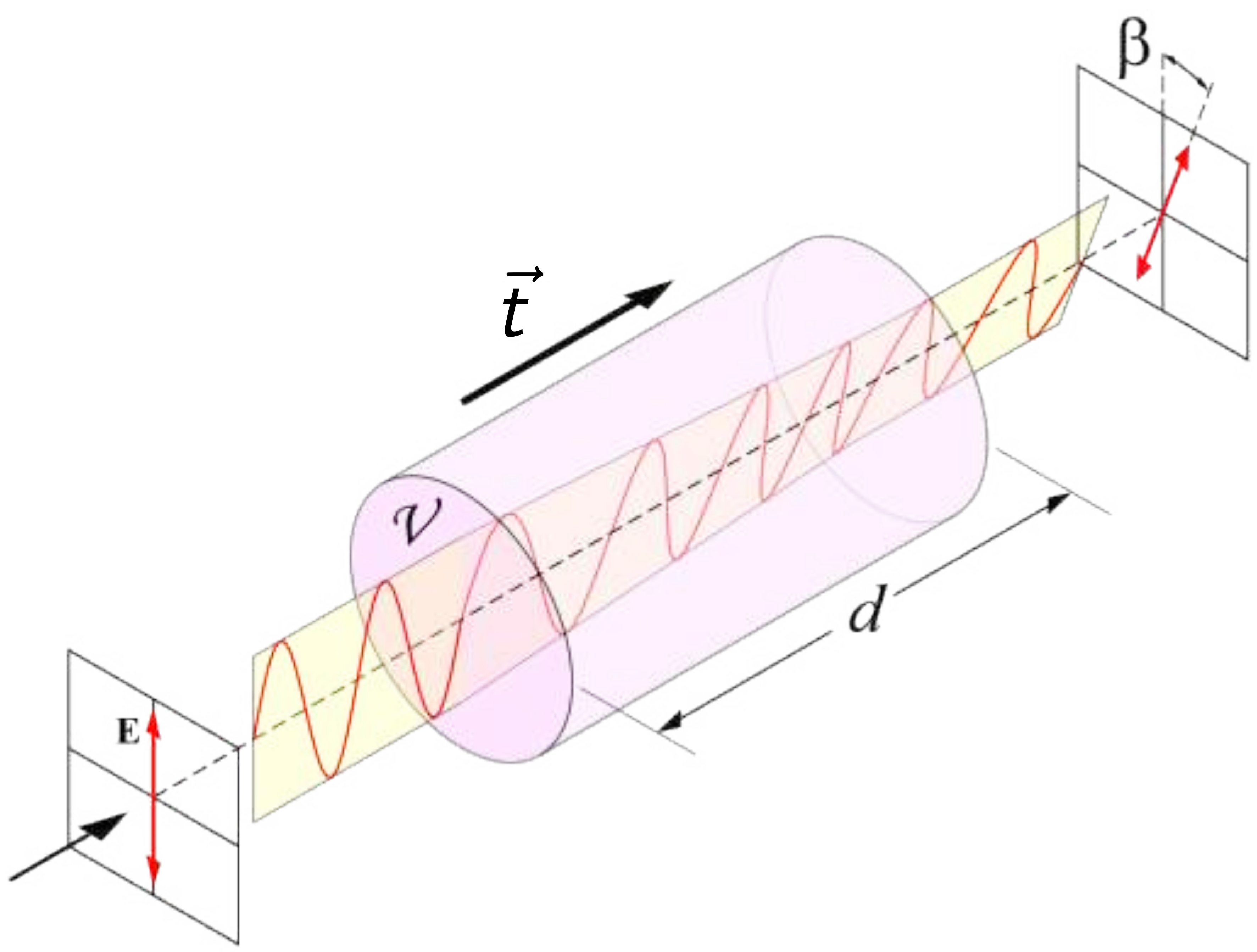


Polarization angle:

$$\beta \propto \vec{t} \cdot \vec{c}$$

Toroidal moment from Faraday rotation

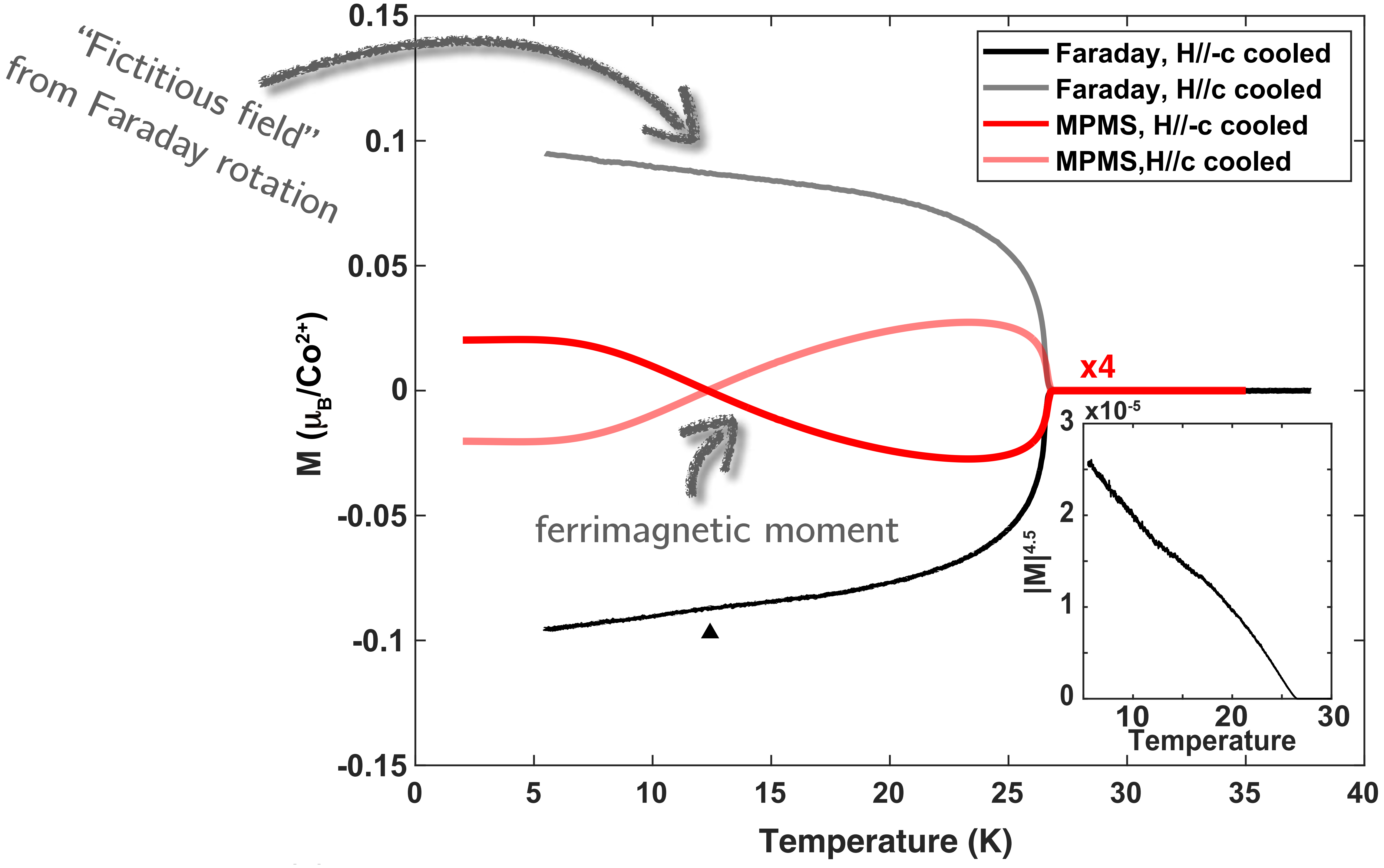
Faraday rotation:



Polarization angle:

$$\beta \propto \vec{t} \cdot \vec{c}$$

Toroidal & ferrimagnetic moments:

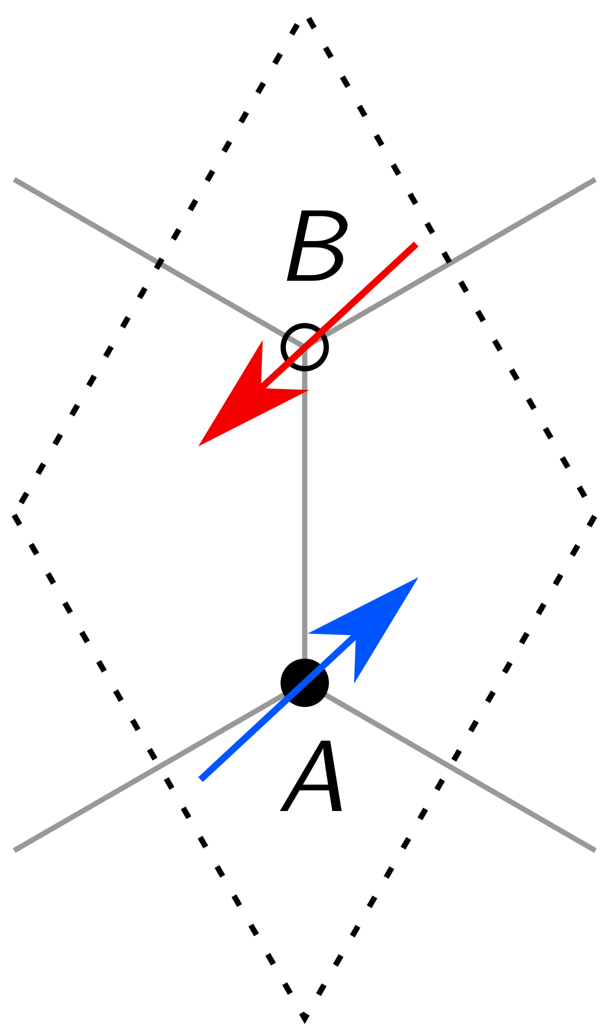


$\Rightarrow \quad \vec{t} \neq 0$
when $\vec{M} \neq 0$

[Jin *et al.*, arXiv '25]

Ferrimagnetism: Symmetry constraints

Néel



$\vec{M} \neq 0$ if $A \neq B$

$\vec{t} = 0$

Ferrimagnetism: Symmetry constraints

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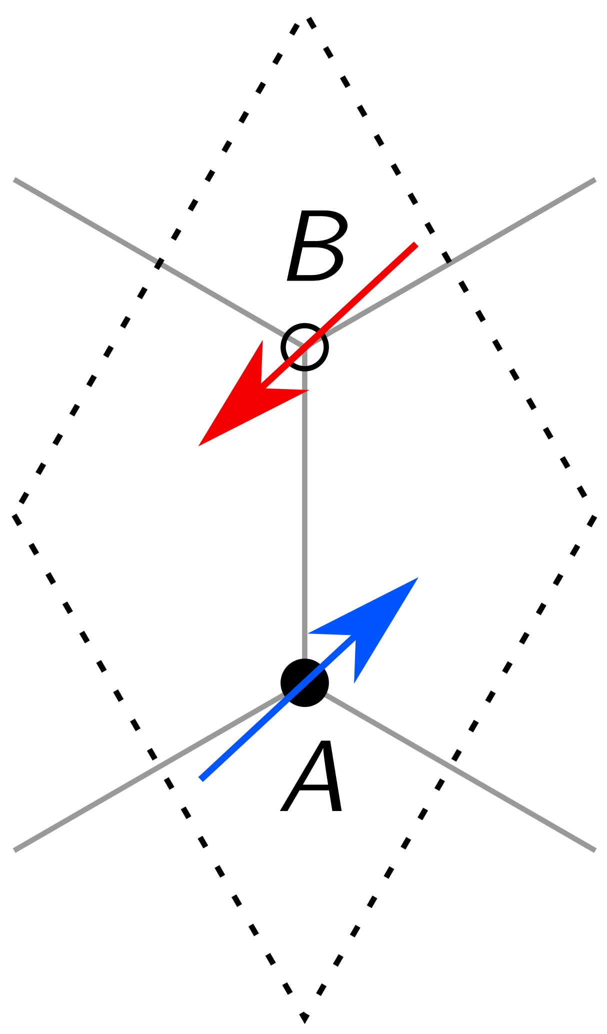

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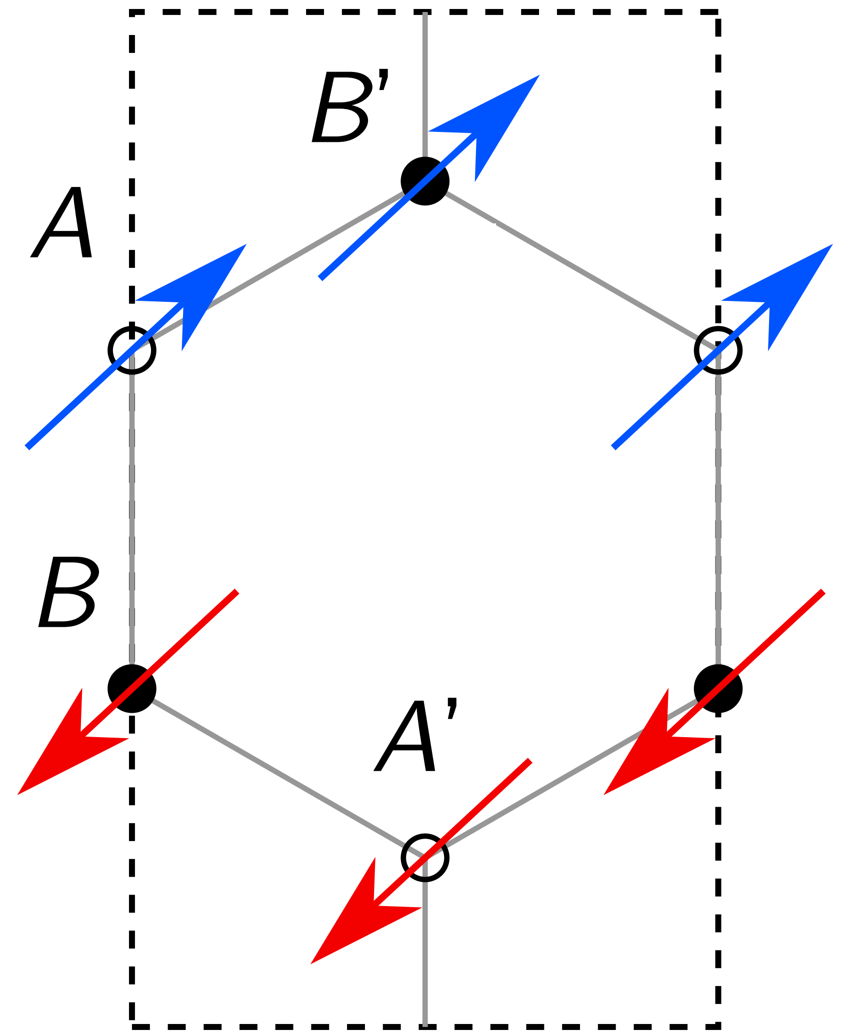
Zigzag



$\vec{M} = 0$ as long as $A = A'$, $B = B'$

$\vec{t} = 0$

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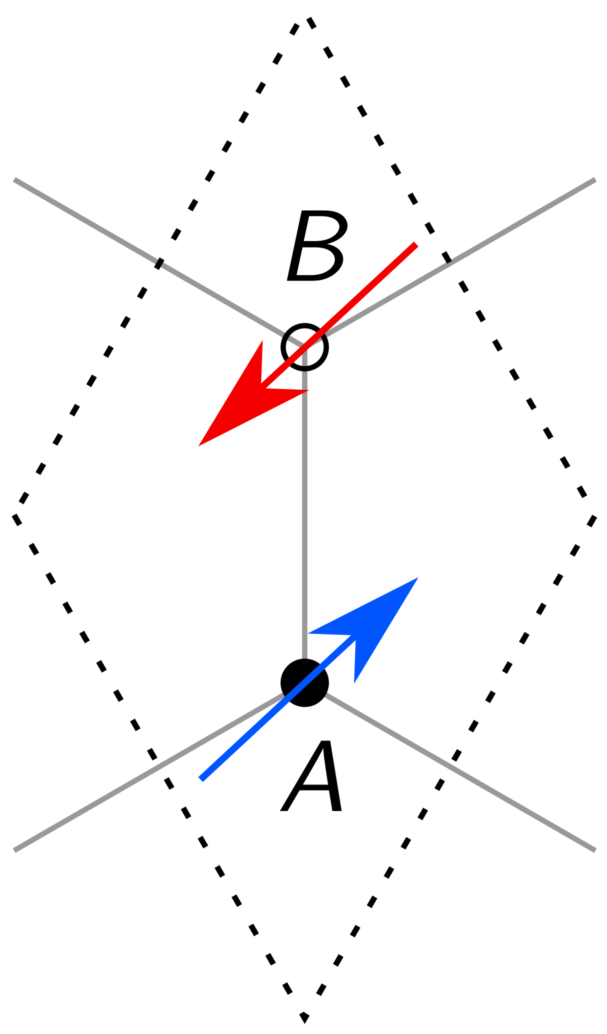

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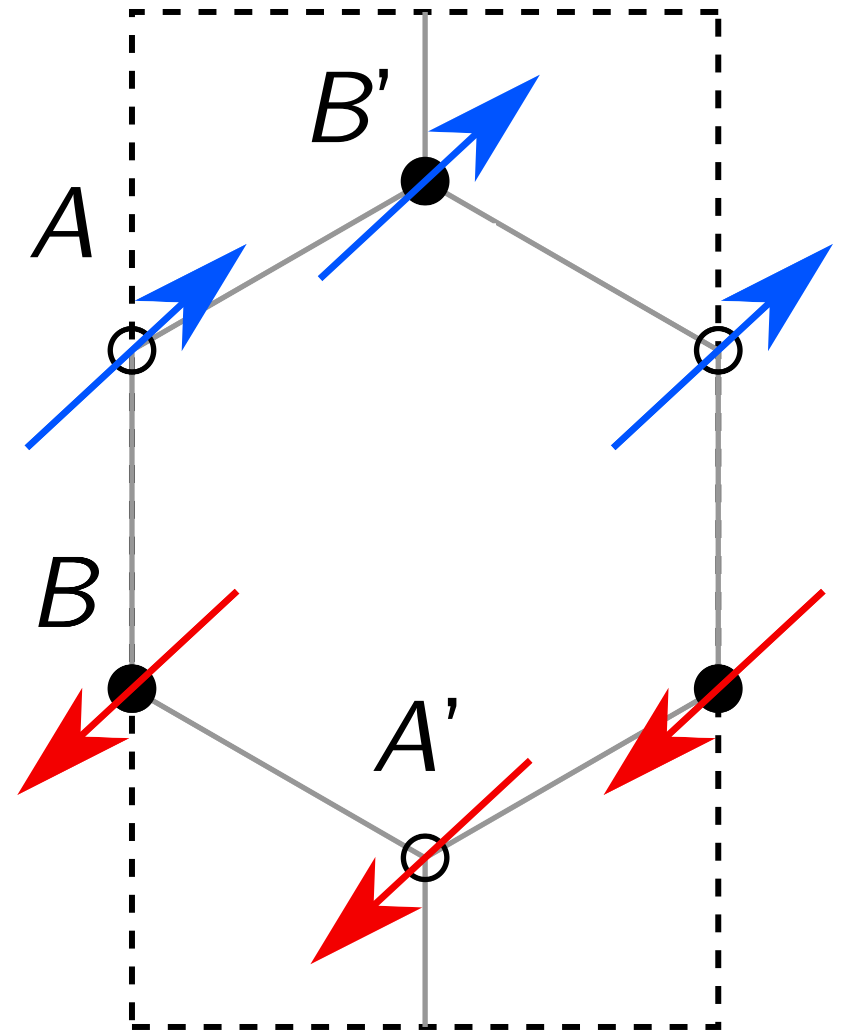

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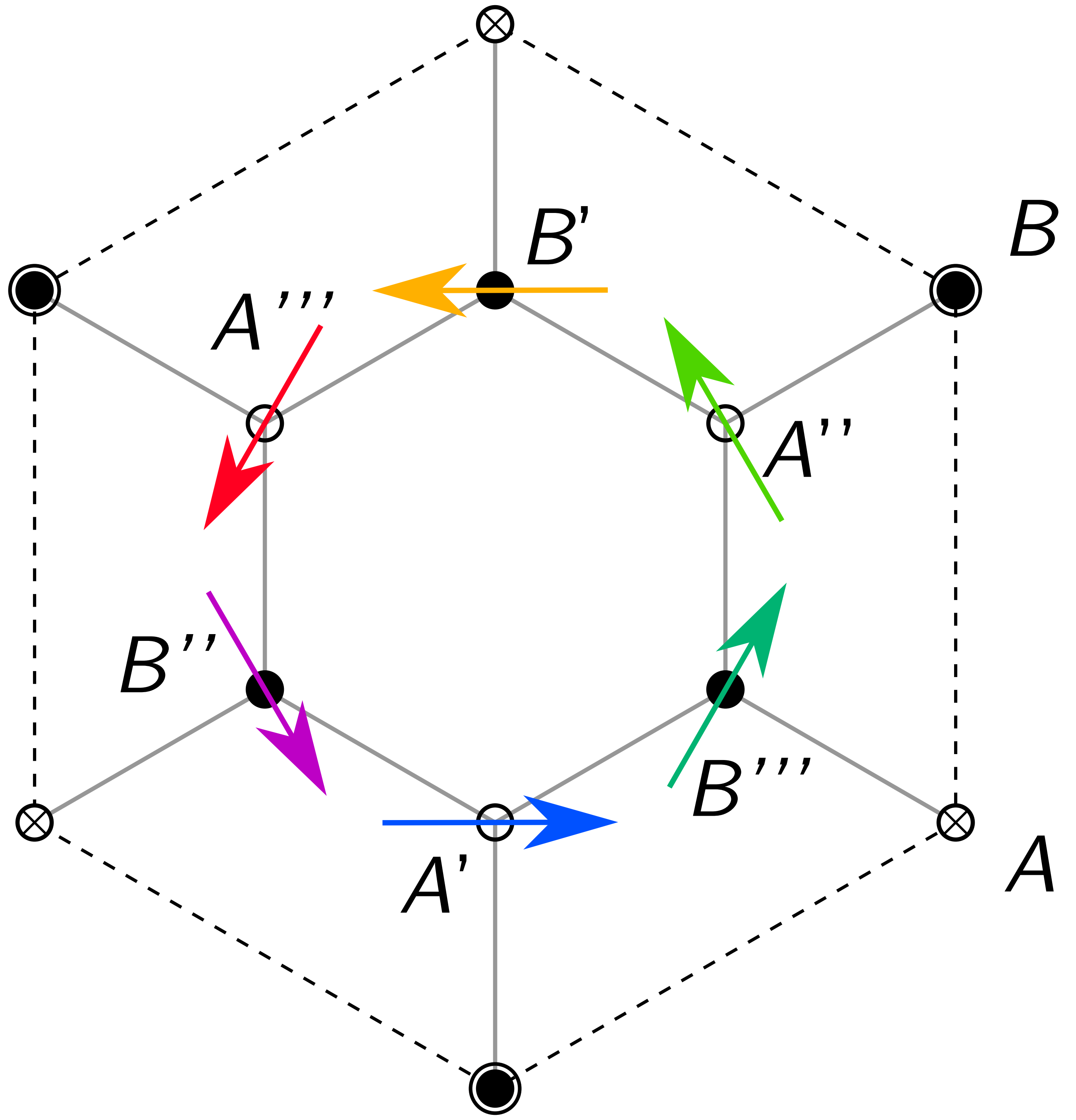
Triple-**q**

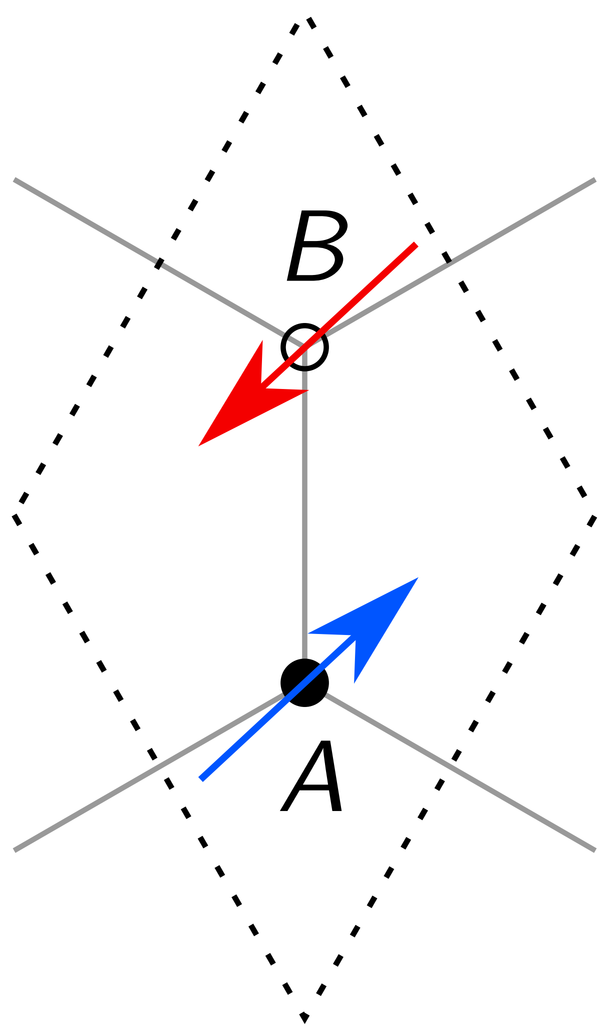


$\vec{M} \neq 0$ if $A \neq B$, $A' \neq B'$, etc.

$\vec{t} \neq 0$

Ferrimagnetism: Symmetry constraints

Néel



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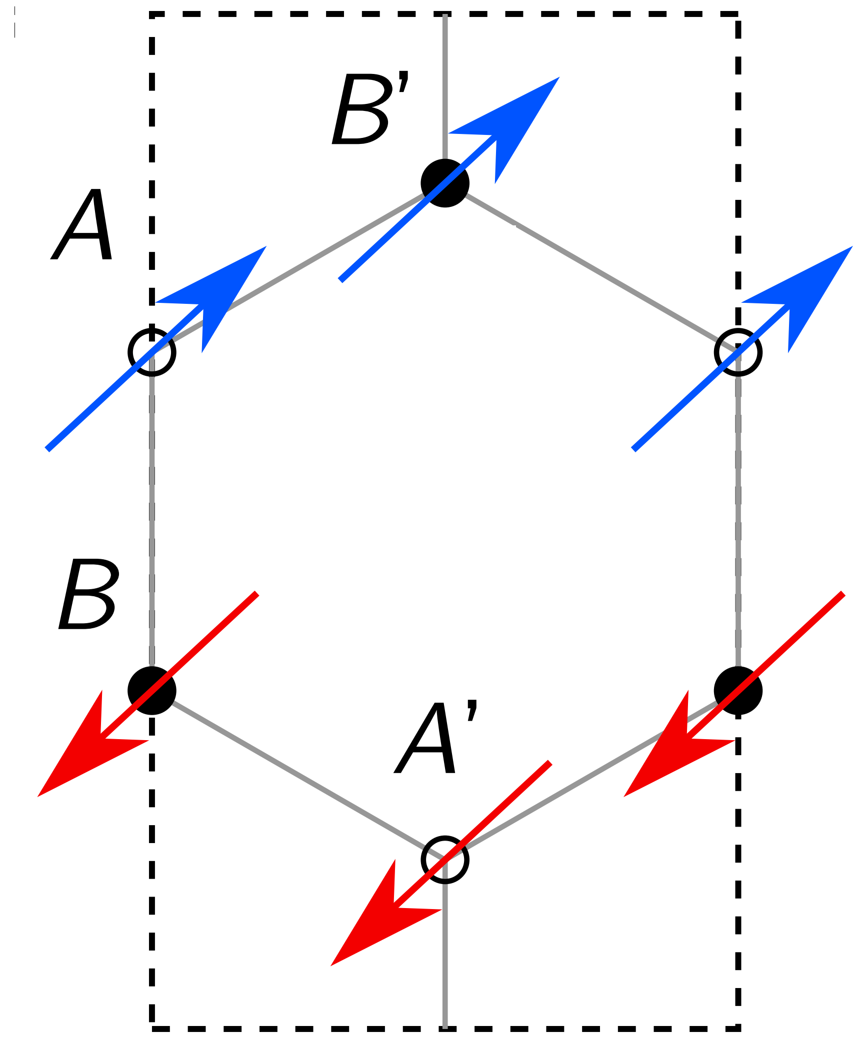

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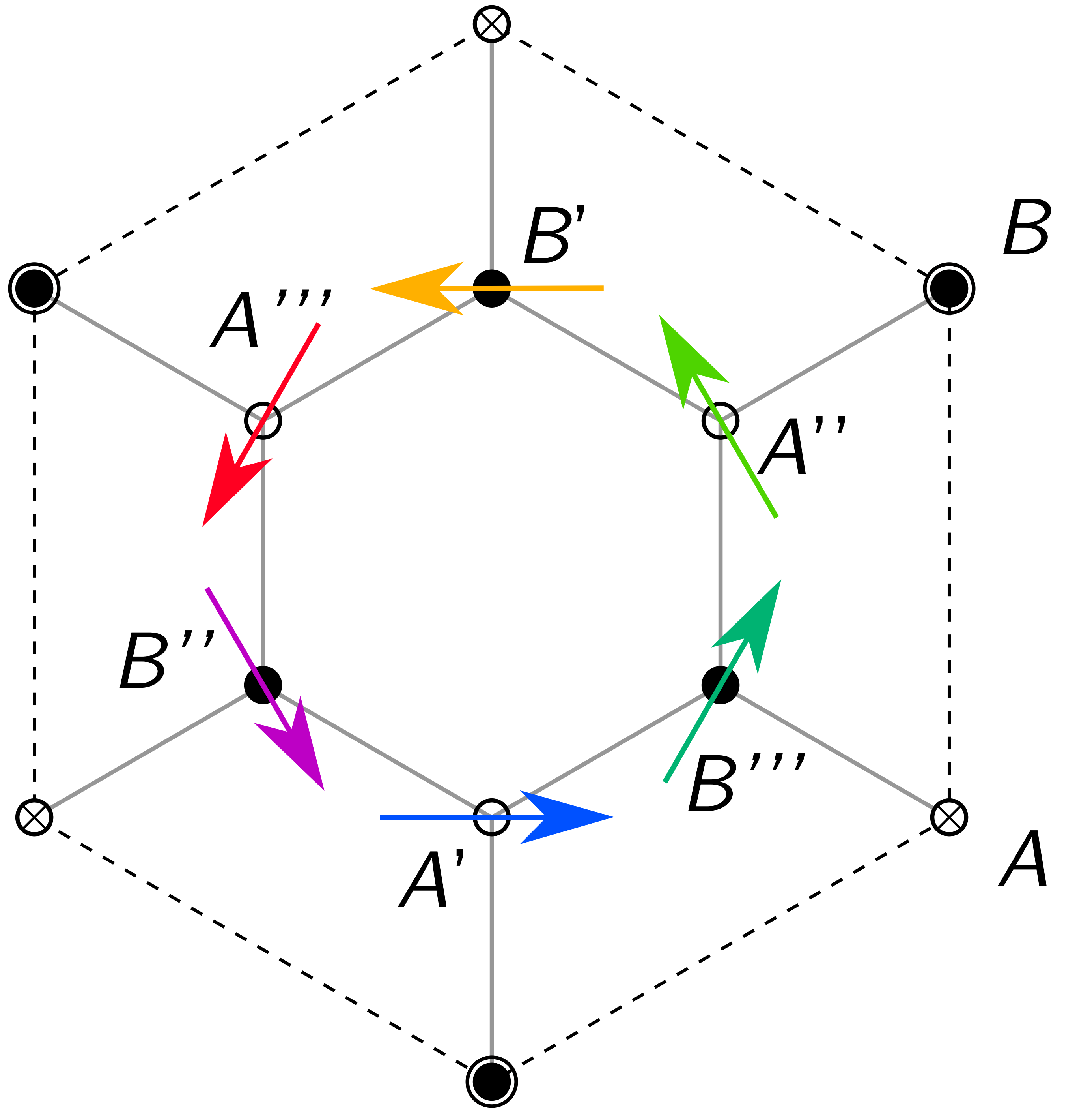
Triple-**q**



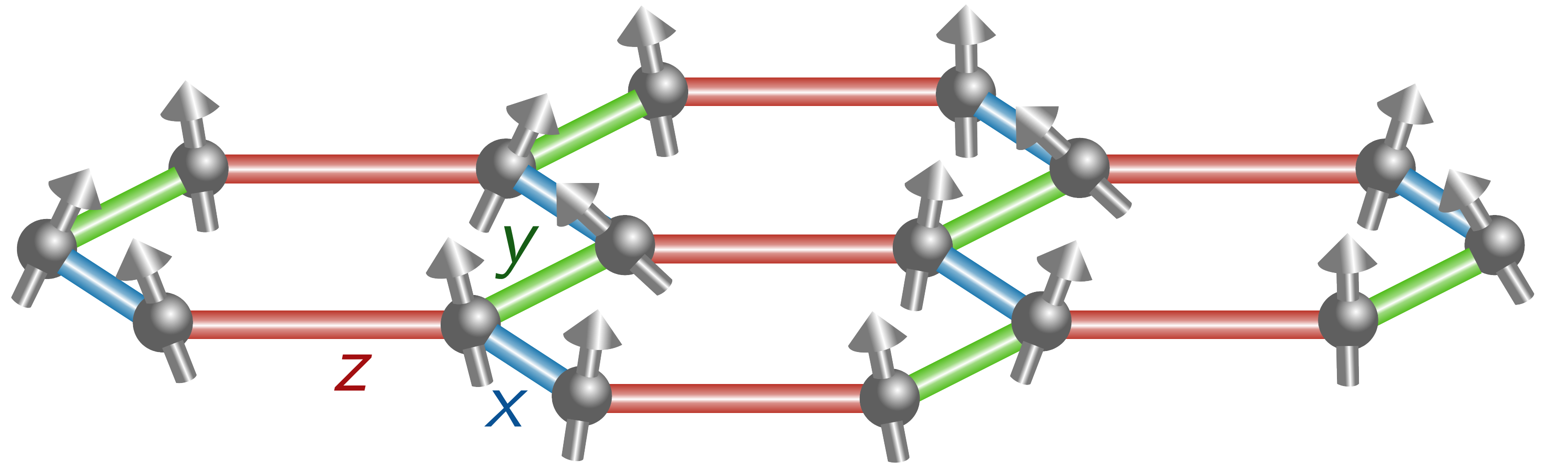
$\vec{M} \neq 0$ if $A \neq B$, $A' \neq B'$, etc.

$\vec{t} \neq 0$

No ferrimagnetic moment in zigzag state!

... independent of modeling

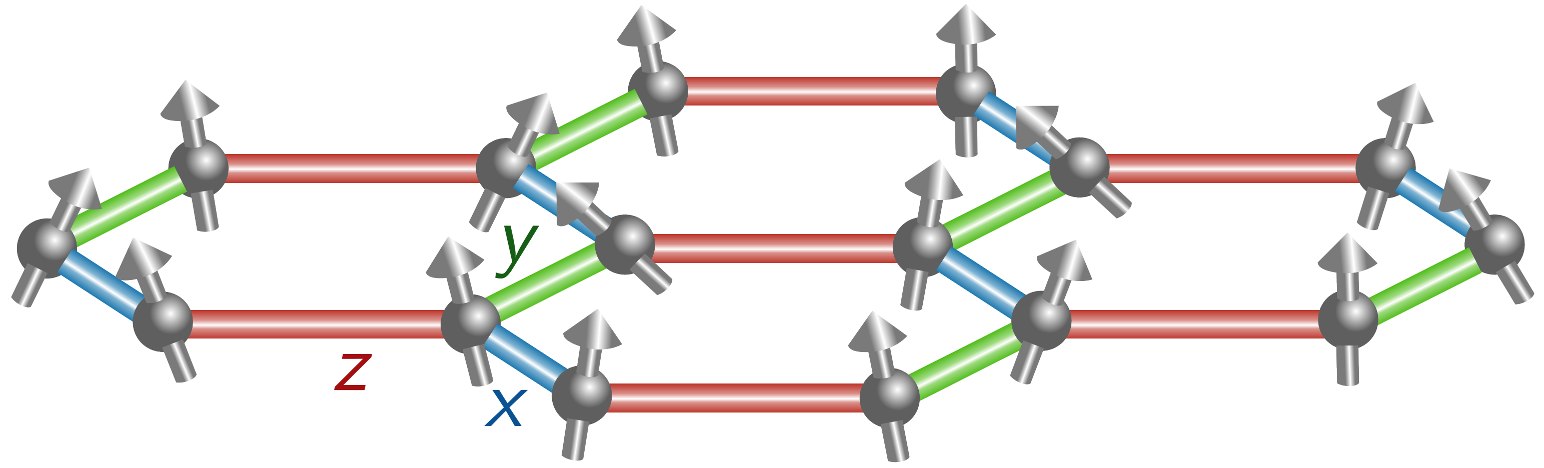
Extended Heisenberg-Kitaev model



Toy Hamiltonian:

$$\mathcal{H} = \mathcal{H}^{(0)}_{\vec{h}} + \mathcal{H}^{(1)}_{JK\Gamma'\Gamma'} + \mathcal{H}^{(2)}_{J_2^A J_2^B} + \mathcal{H}^{(3)}_{J_{\hexagon}}$$

Extended Heisenberg-Kitaev model



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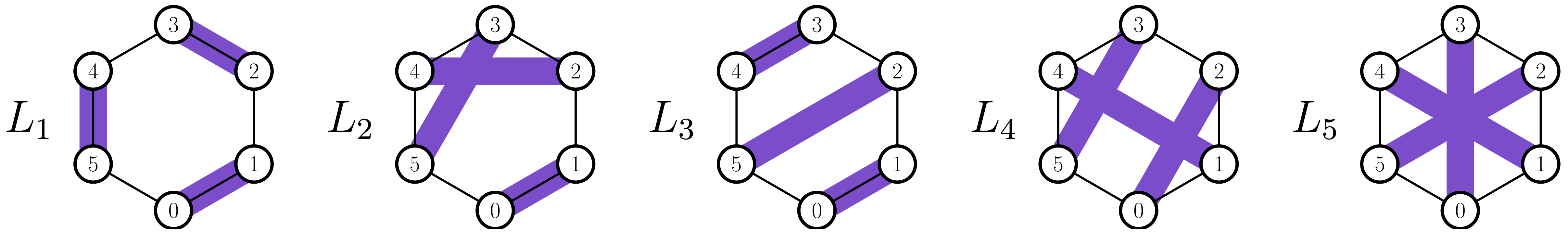
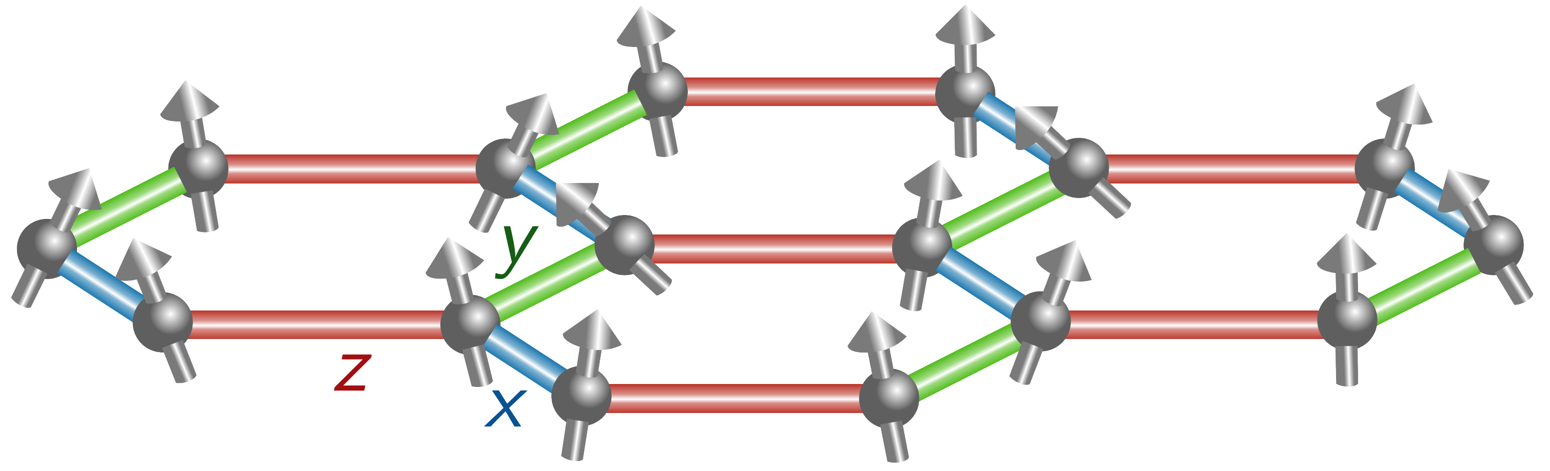
Ring exchange:



$$\mathcal{H}^{(3)}_{J_{\hexagon}} = J_{\hexagon} \sum_{\langle ijklmn \rangle} L_{ijklmn} (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l)(\mathbf{S}_m \cdot \mathbf{S}_n)$$

... 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]

Extended Heisenberg-Kitaev model



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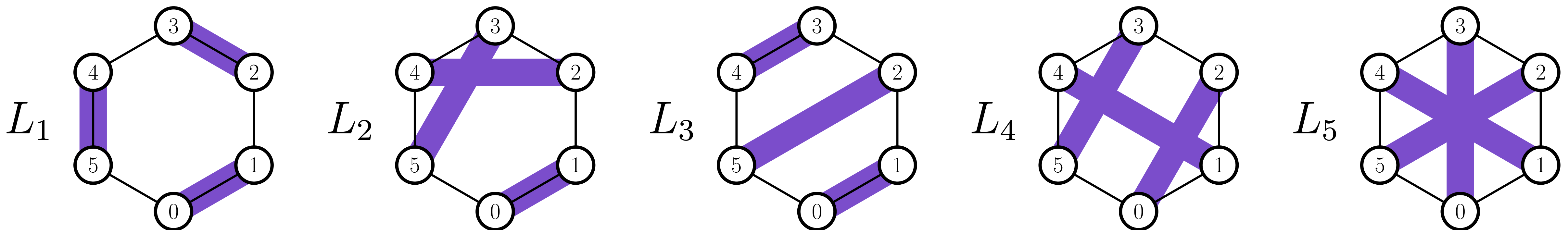

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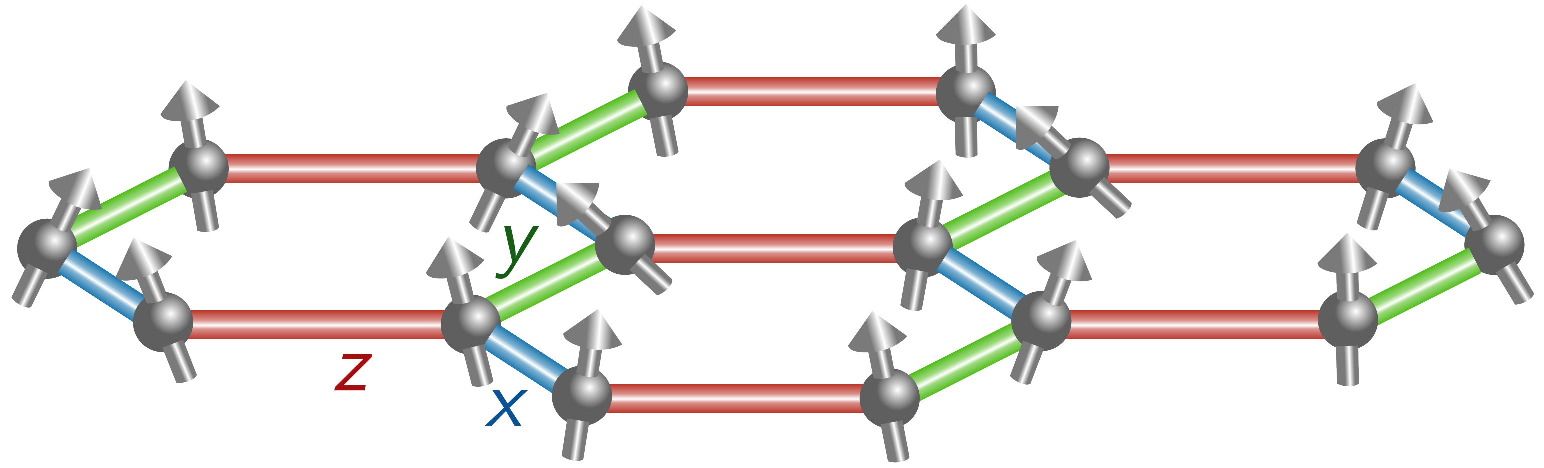
Training field:

$$\mathcal{H}^{(0)}_{\vec{h}} = -h\vec{c} \cdot \sum_i \vec{S}_i$$

... with $h \to 0$

Extended Heisenberg-Kitaev model



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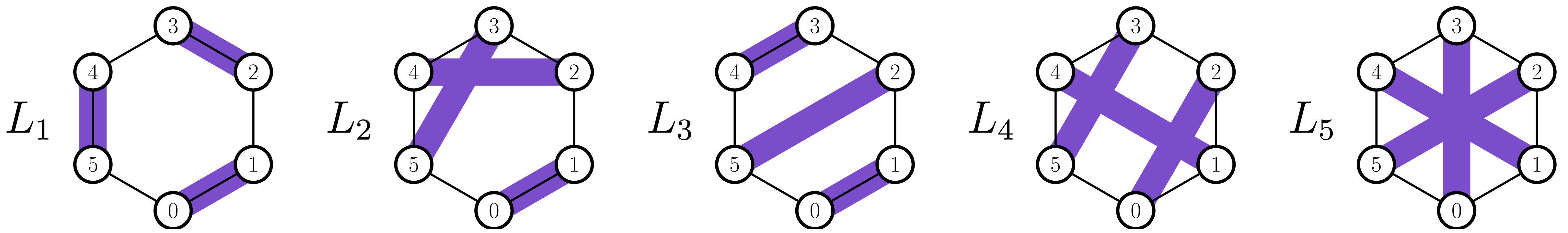
Ring exchange:



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Training field:

$$\mathcal{H}^{(0)}_{\vec{h}} = -h\vec{c} \cdot \sum_i \vec{S}_i$$

... with $h \to 0$

Representative parameter sets:

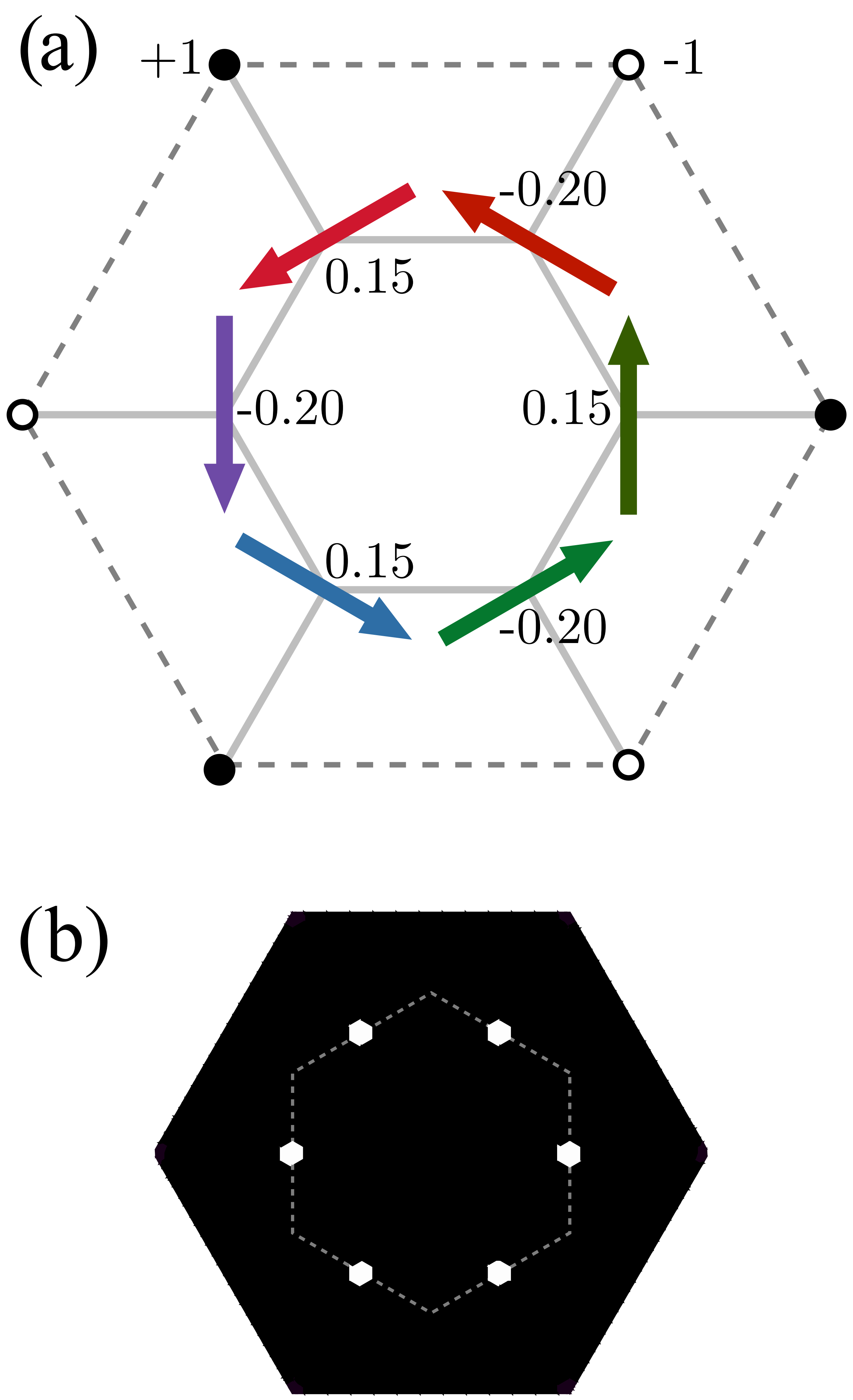
$$(J, K, \Gamma, \Gamma', J_2^A, J_2^B, J_{\hexagon} S^4)/J_0 =$$

$$\begin{cases} \left(-\frac{1}{5}, -\frac{2}{3}, 3, -\frac{1}{2}, -\frac{1}{4}, \frac{1}{4}, -\frac{1}{5}\right) & \text{“triple-}\mathbf{q}\text{ model”} \\ \left(-\frac{1}{5}, -\frac{2}{3}, 3, -\frac{1}{2}, -\frac{1}{4}, \frac{1}{4}, +\frac{1}{5}\right) & \text{“zigzag model”} \end{cases}$$

... inspired from [Krüger, Chen, Jin, Li, LJ, PRL '23]

Classical Monte Carlo simulations: Ground states

Triple-$\mathbf{q}$ model ($J_{\hexagon} < 0$)



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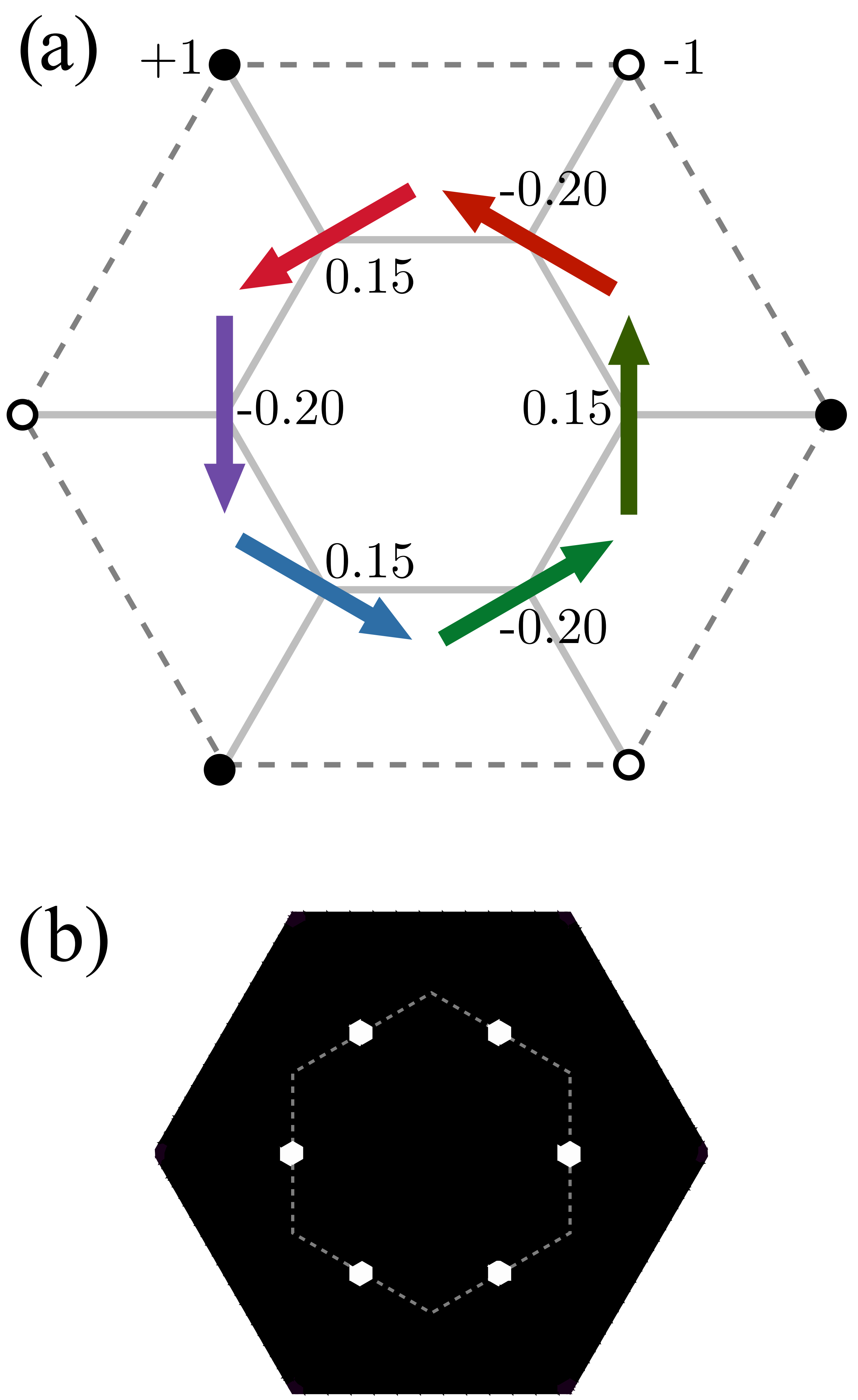
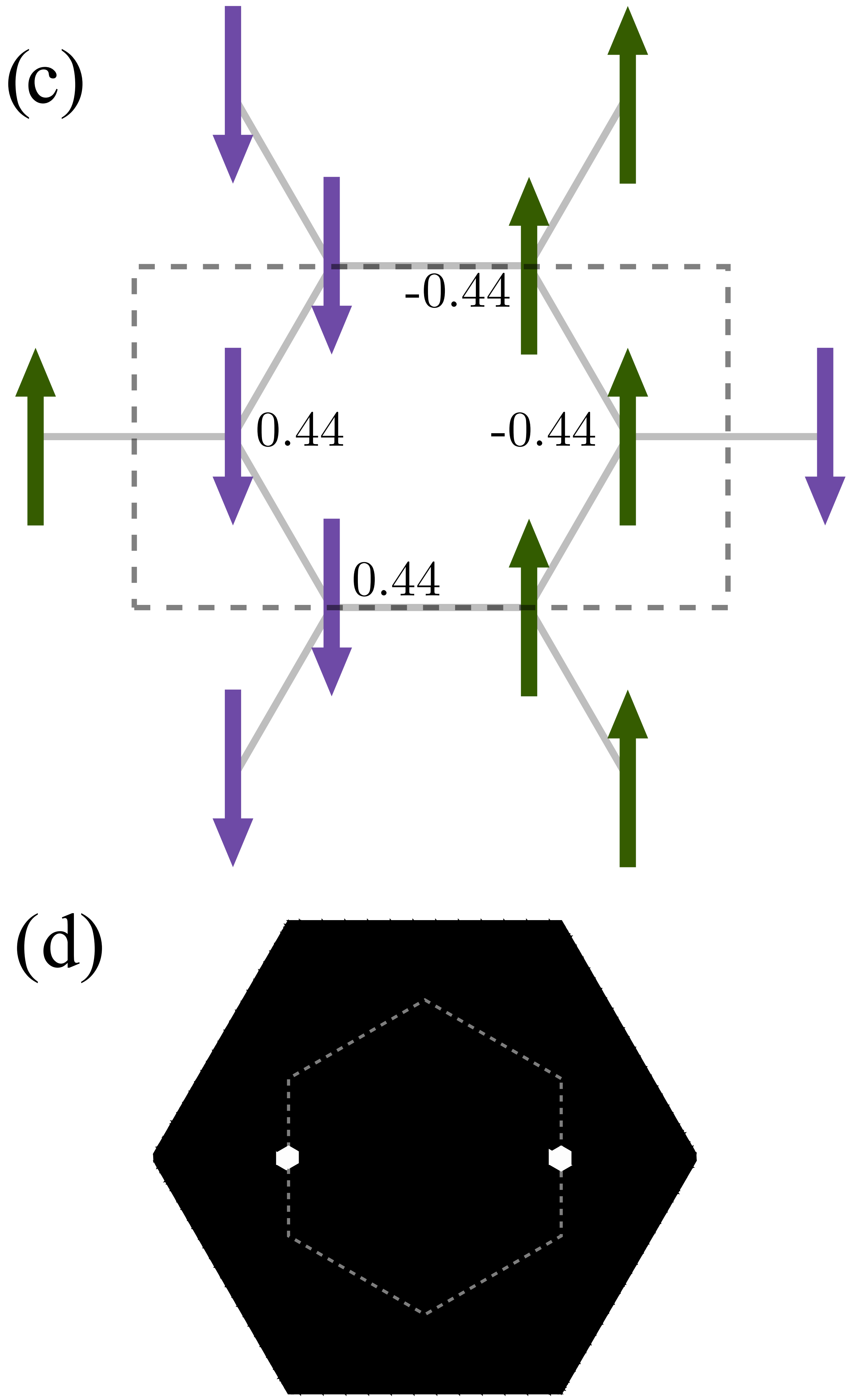


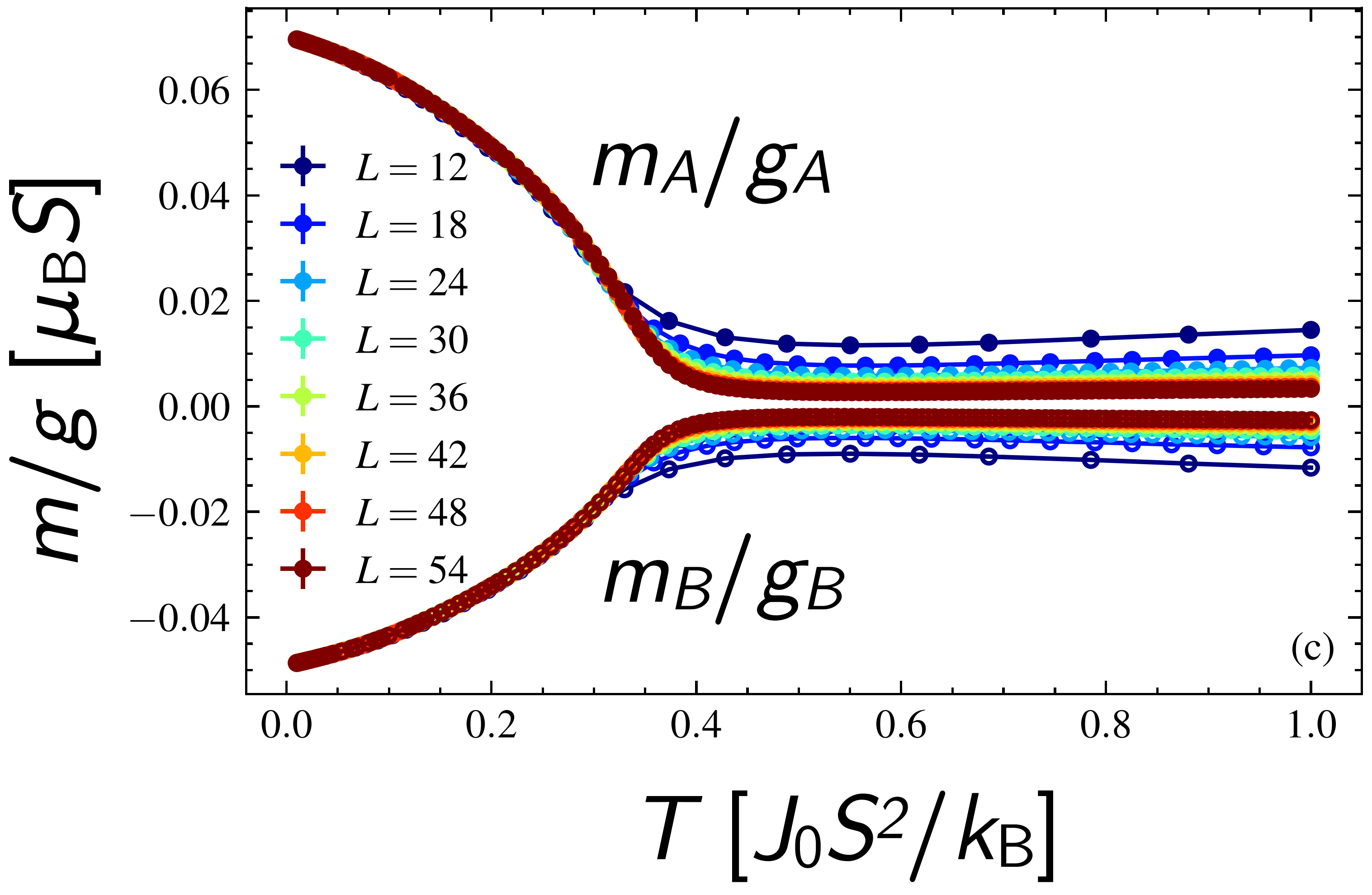
Zigzag model ($J_{\hexagon} > 0$)



Classical Monte Carlo simulations: Finite-*T* magnetization

Sublattice spin moments:

Triple-**q** model



Classical Monte Carlo simulations: Finite-T magnetization

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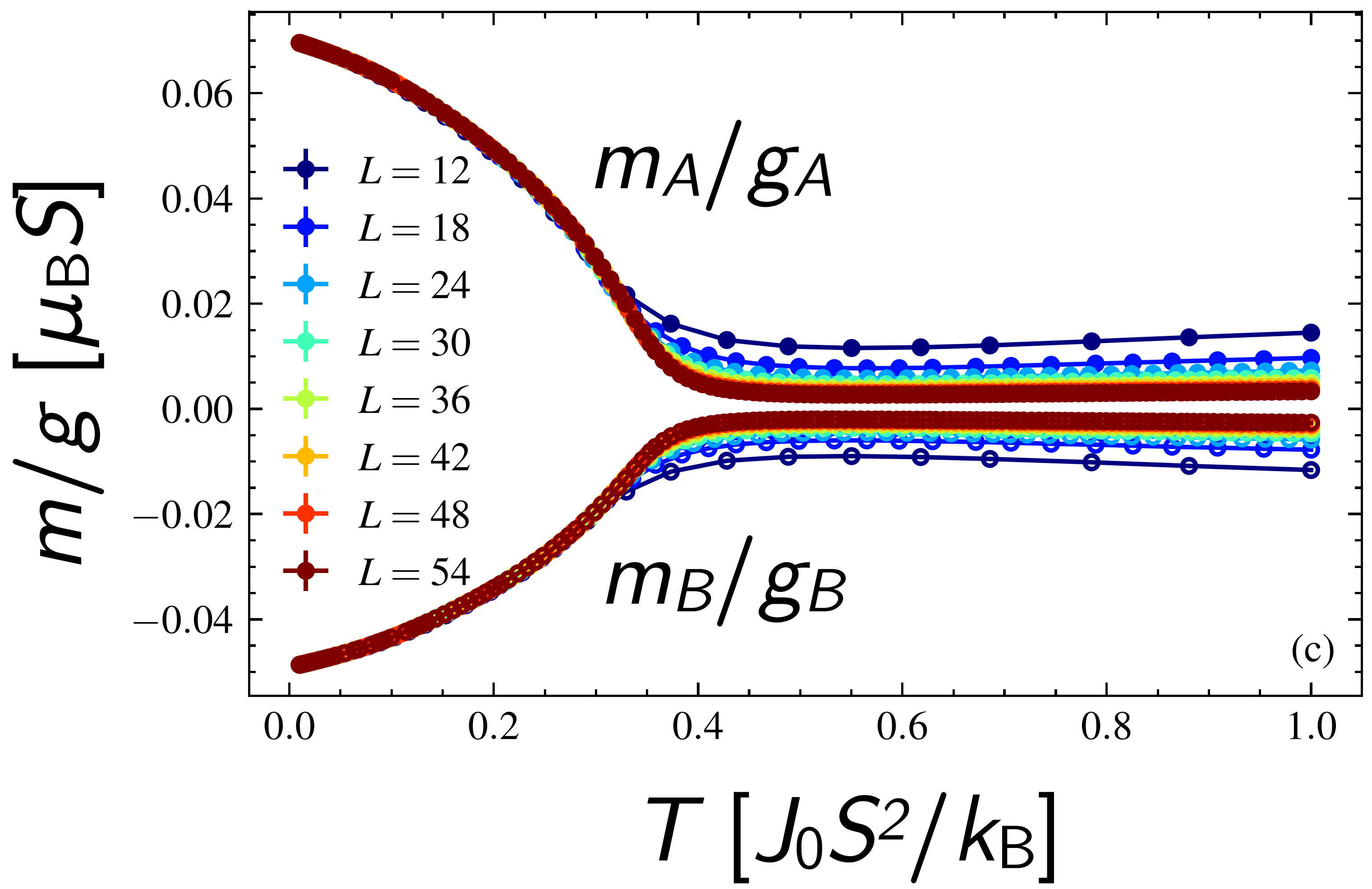
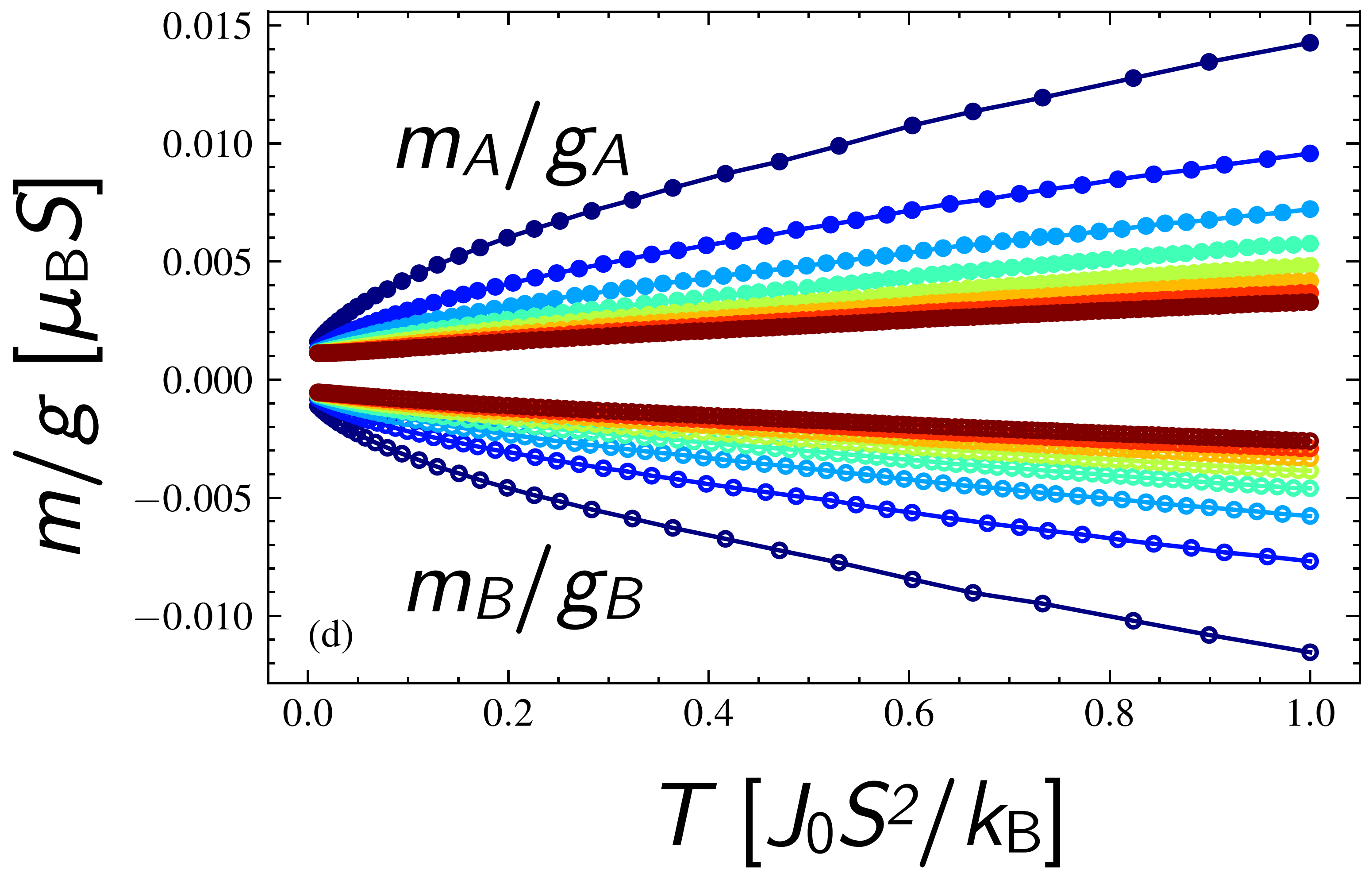
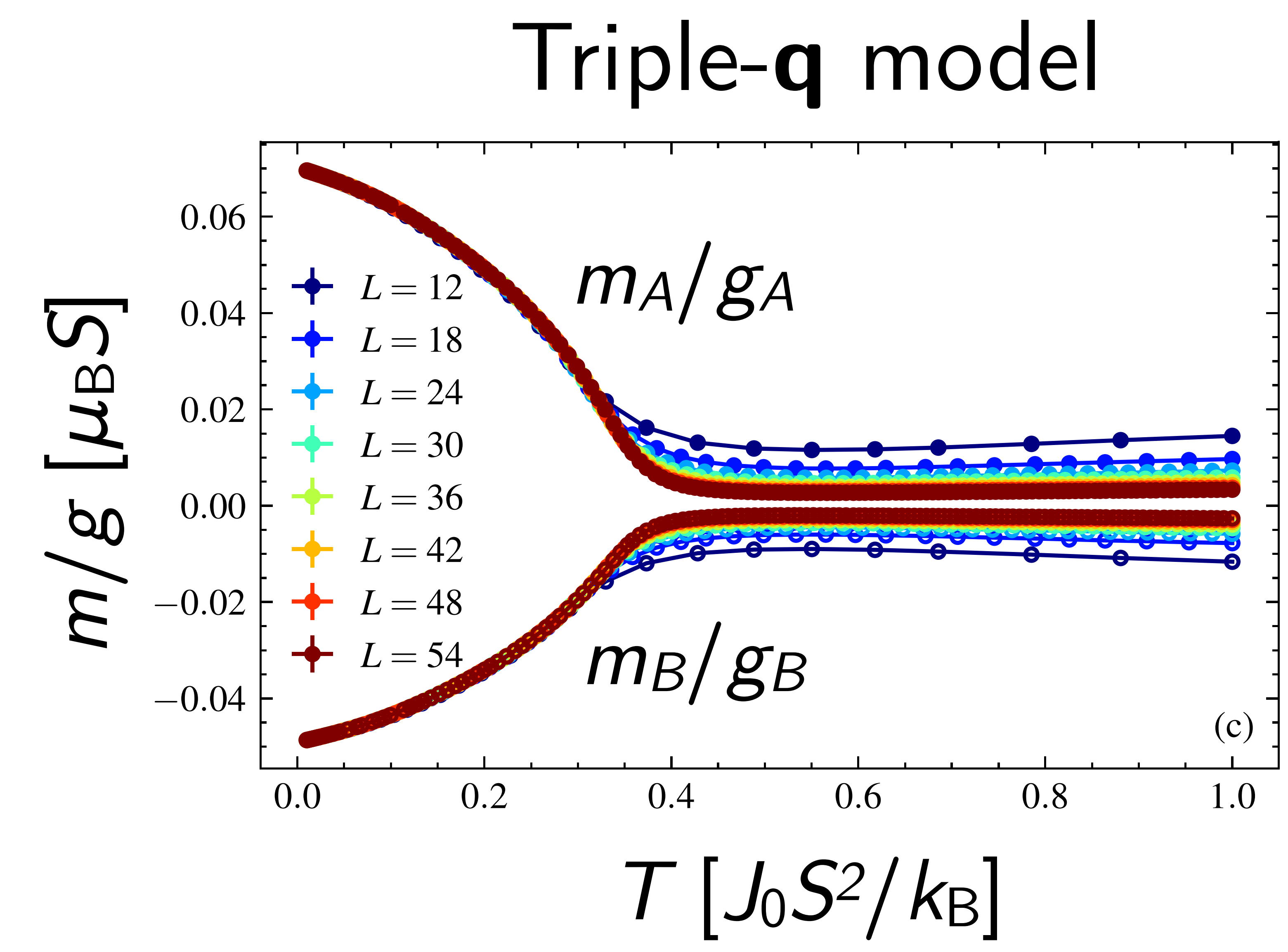


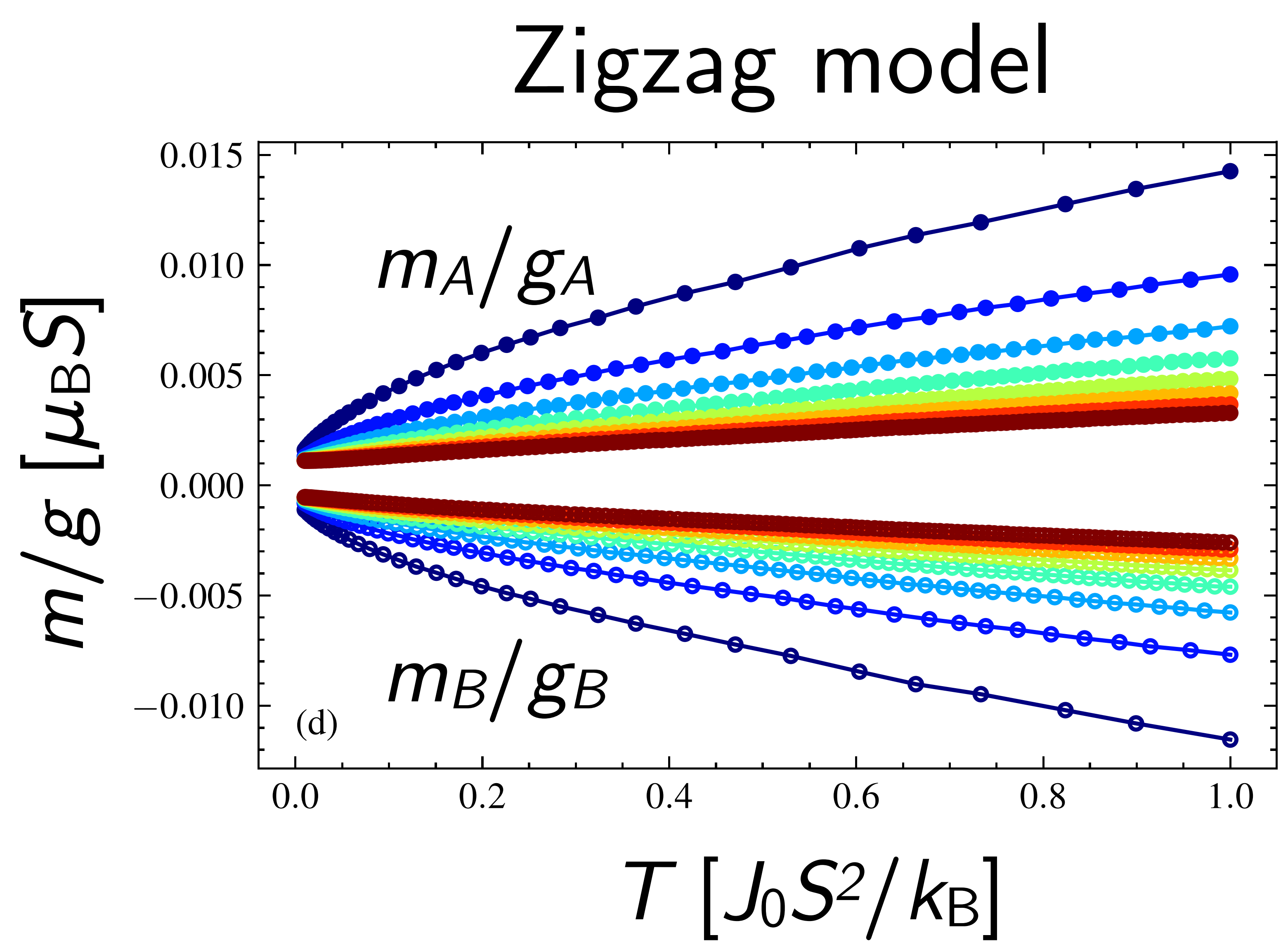
Zigzag model



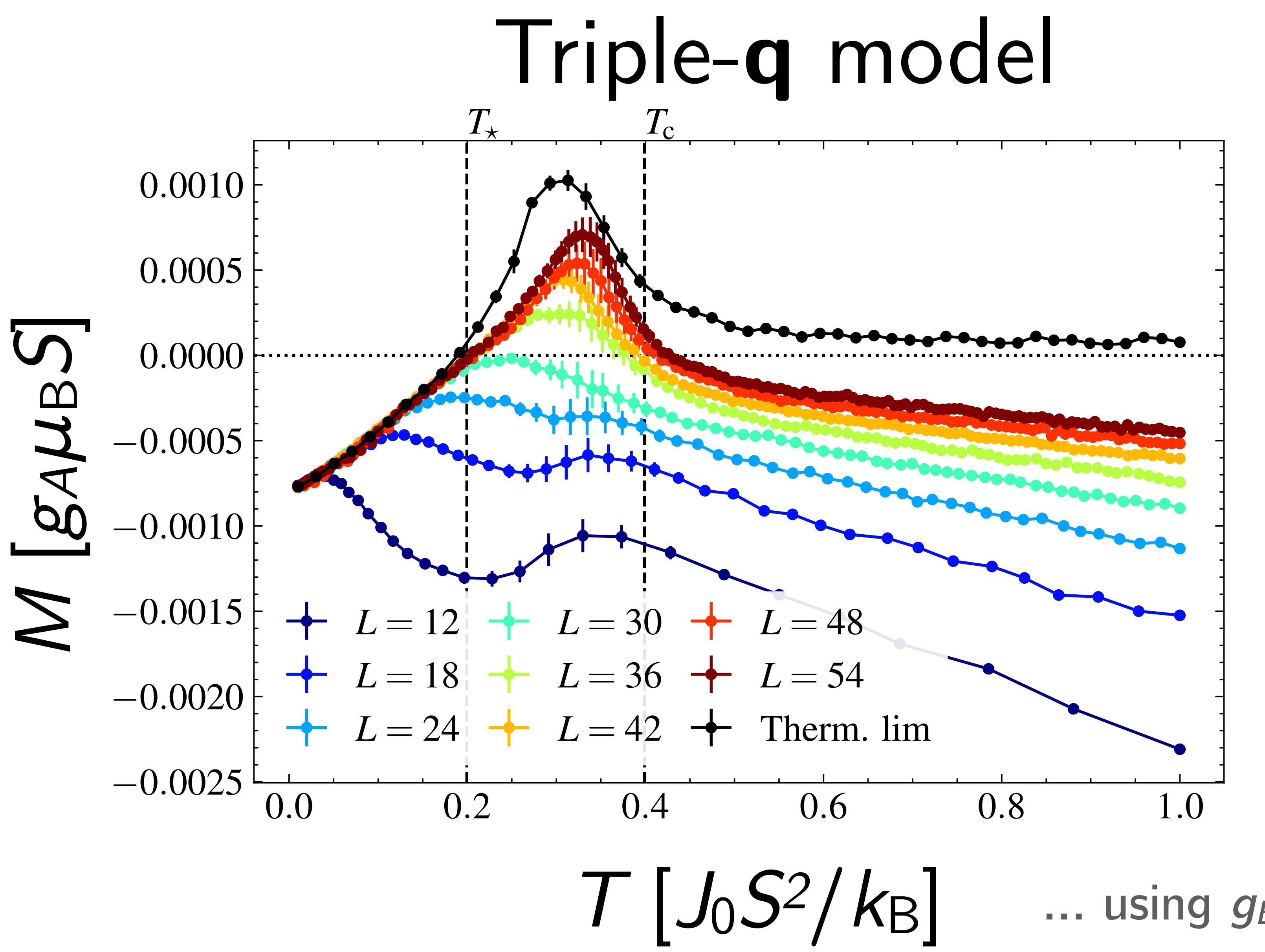
Classical Monte Carlo simulations: Finite-T magnetization

Sublattice spin moments:





Ferrimagnetic moment:



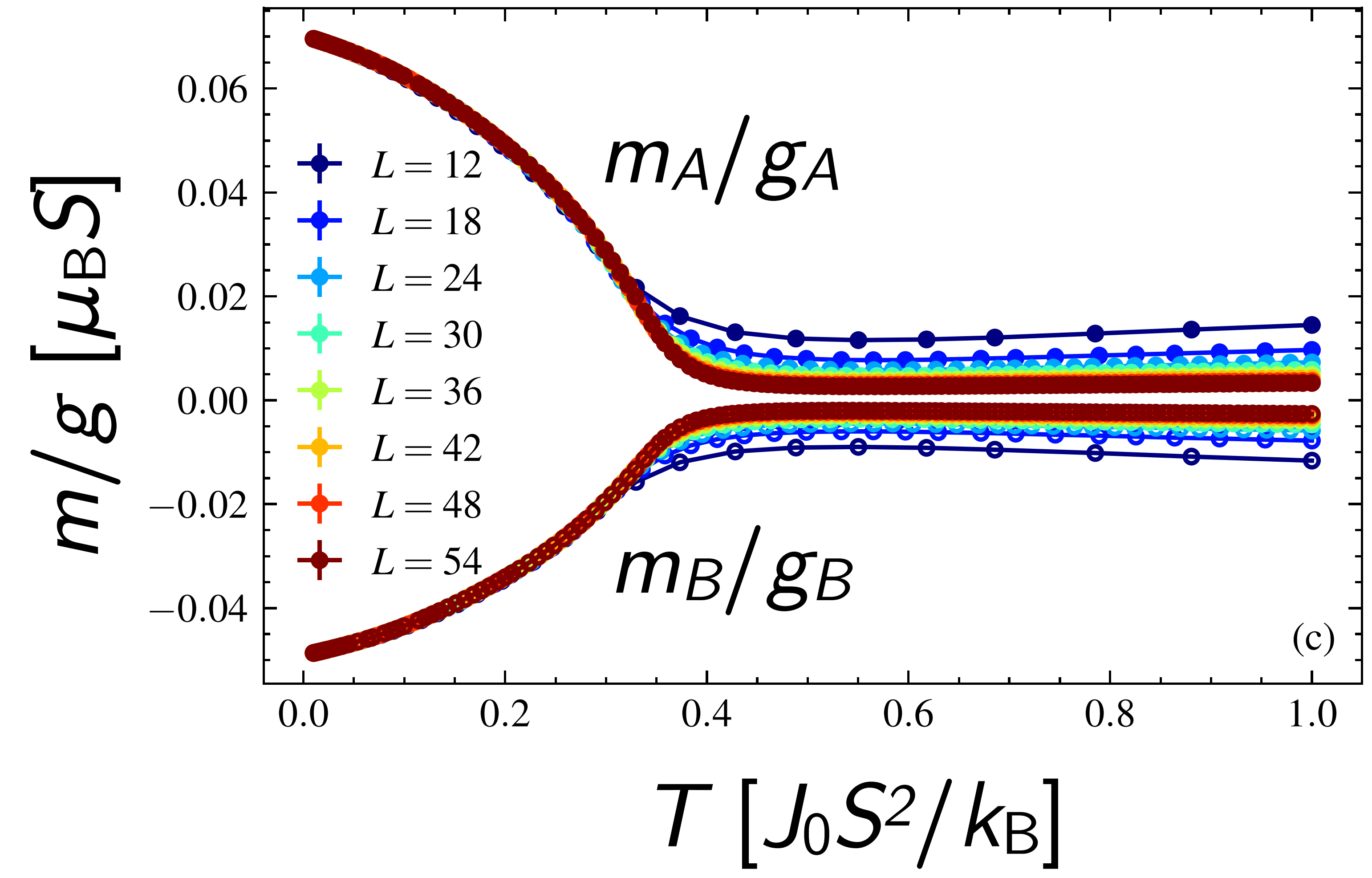
... using $g_B/g_A = 1.45$

[Francini, LJ, PRB '24]

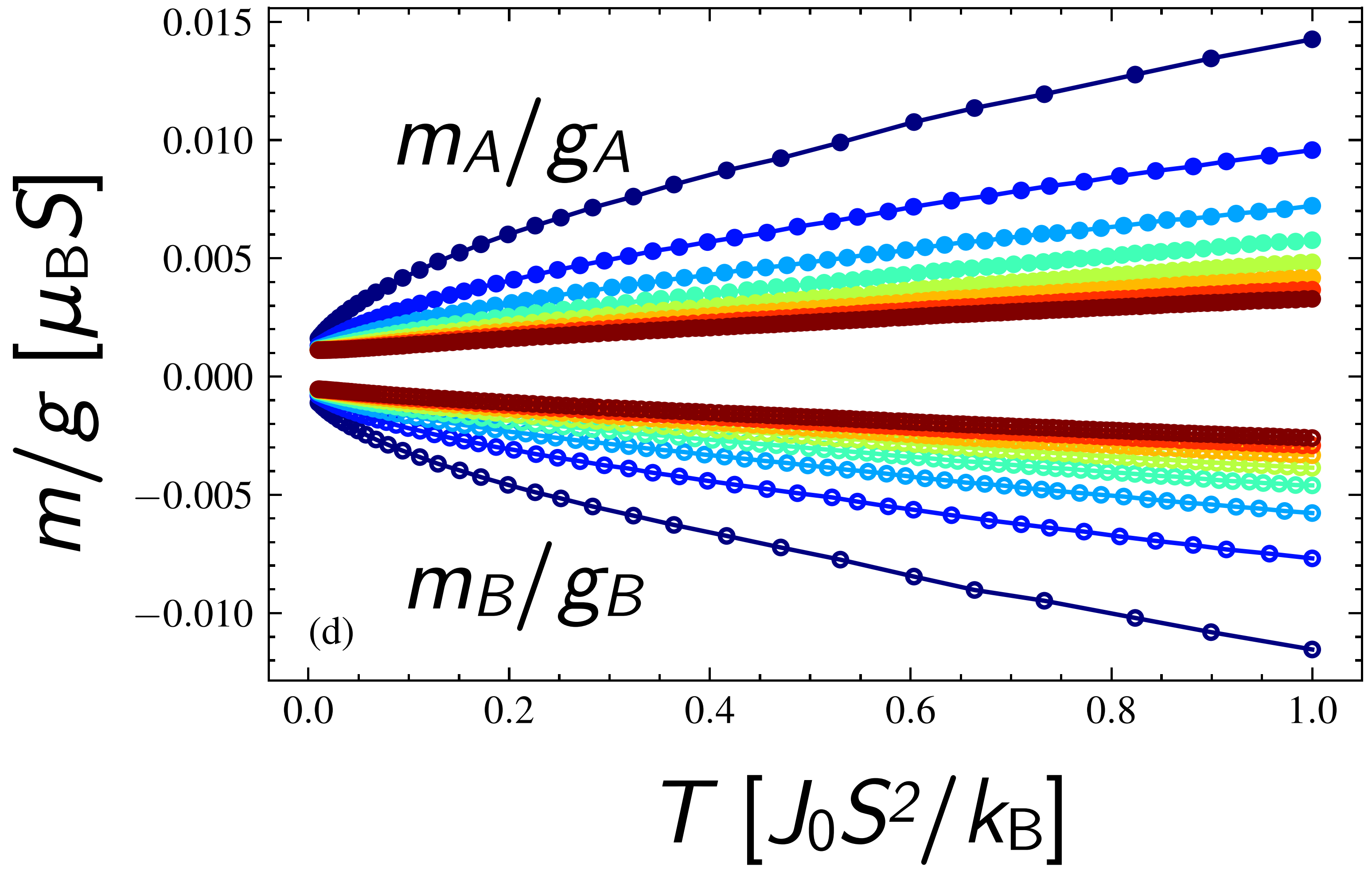
Classical Monte Carlo simulations: Finite-T magnetization

Sublattice spin moments:

Triple-**q** model



Zigzag model



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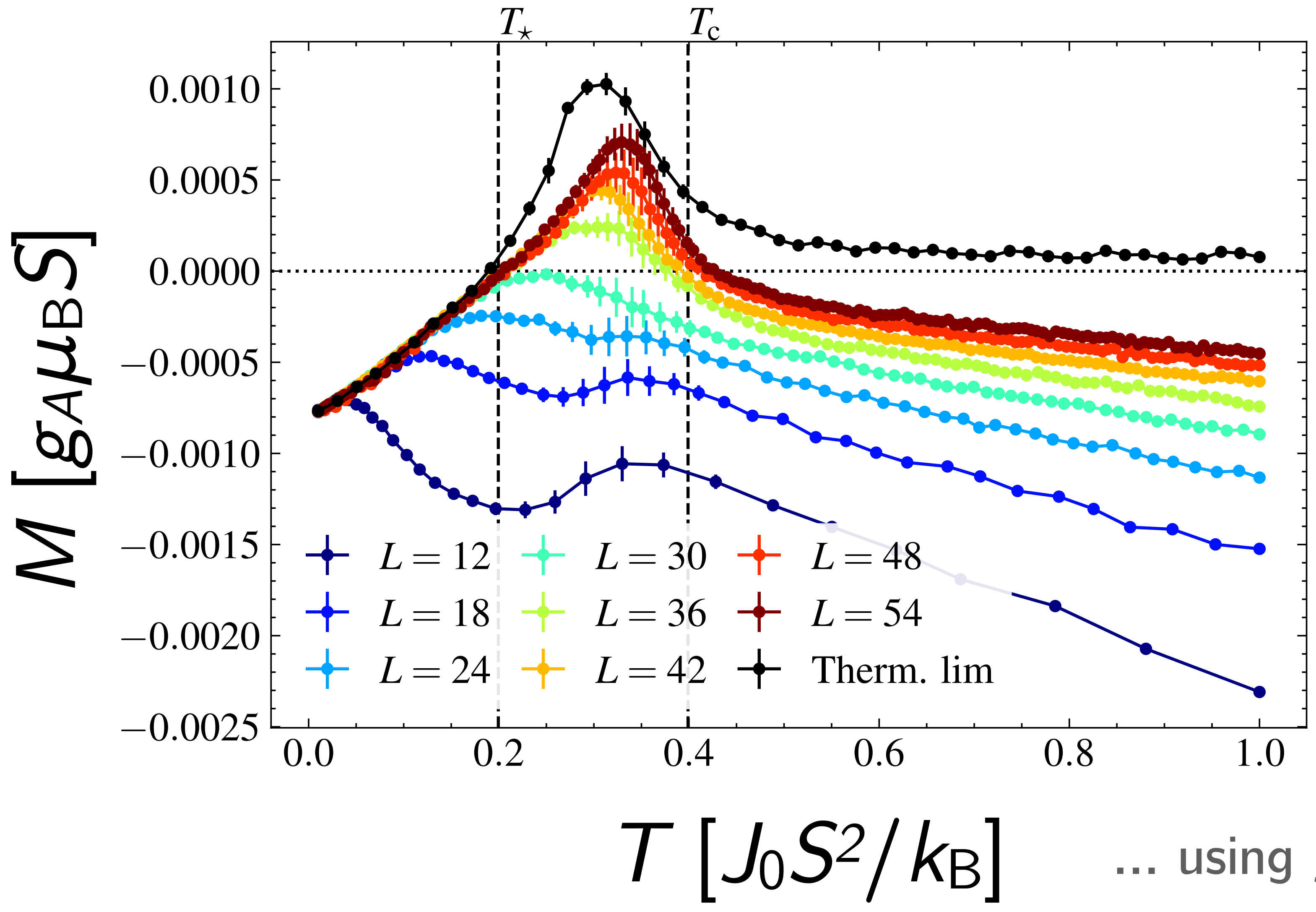

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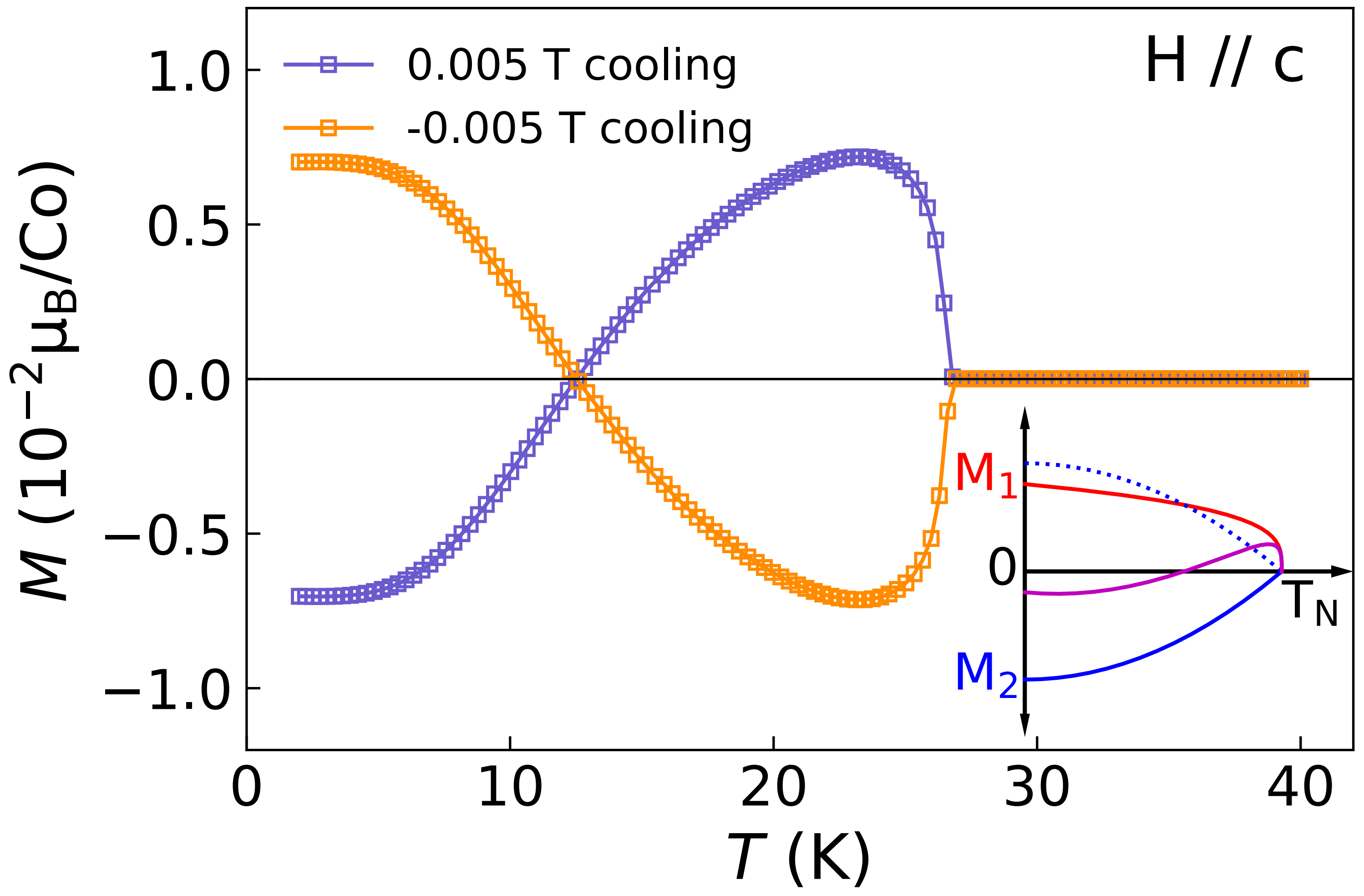
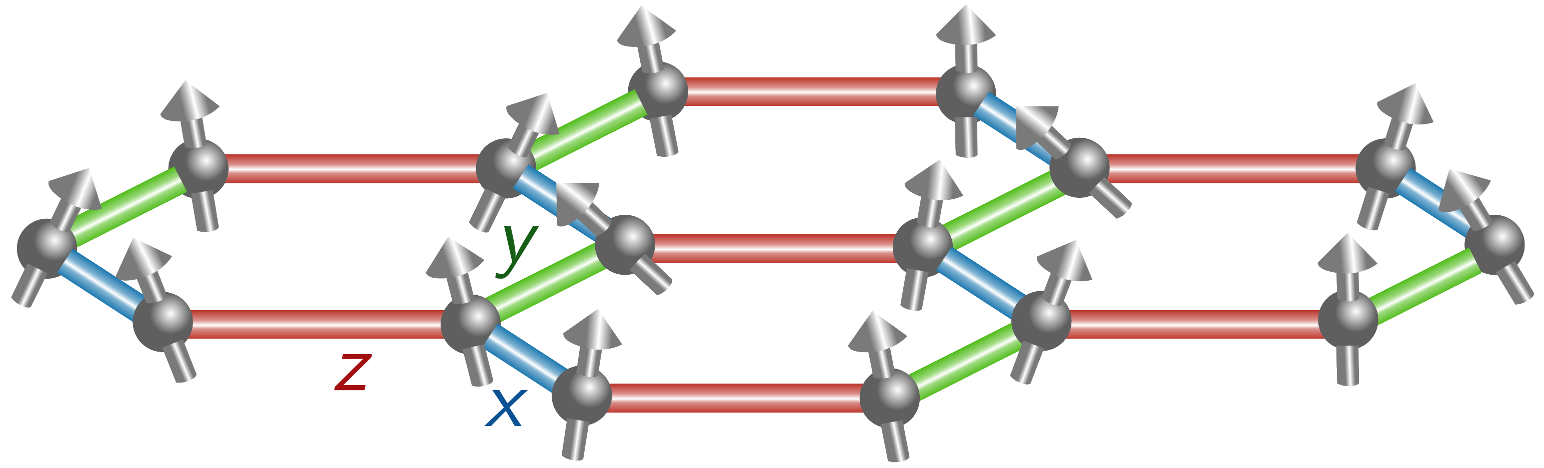
[Francini, LJ, PRB '24]

$Na_2Co_2TeO_6$



[Yao, Li, PRB '20]

Ferrimagnetism from quantum fluctuations



[Francini, Cônsoli, LJ, *in preparation*]

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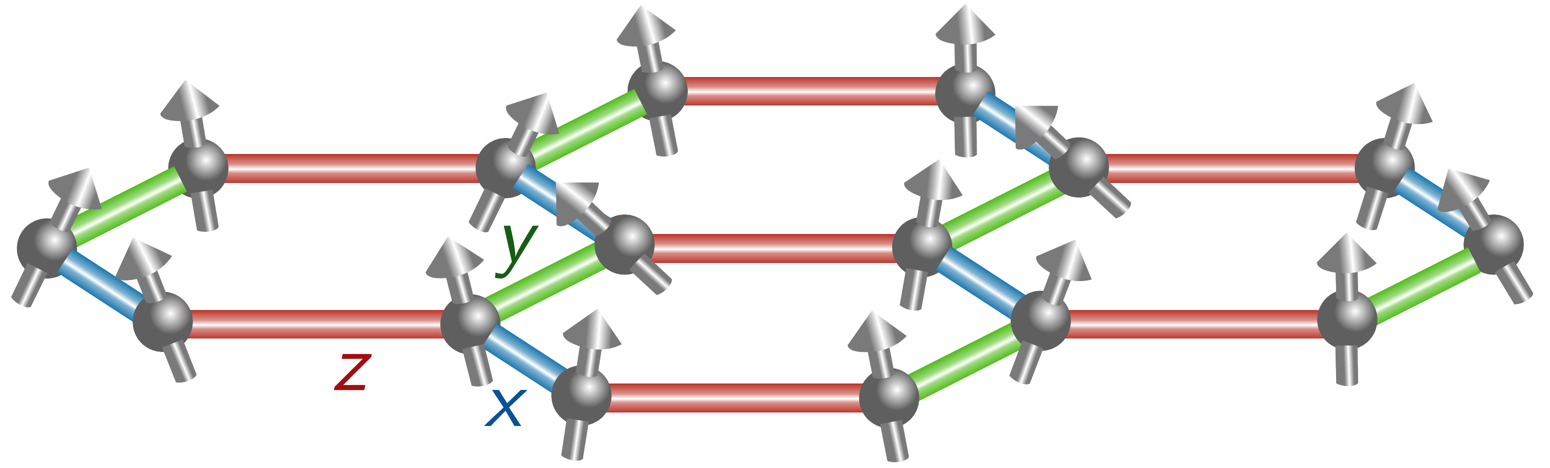

Heisenberg-Kitaev model with sublattice symmetry breaking:

$$(J, K, J_2^A, J_2^B) = J_0(\cos\varphi, \sin\varphi, -\tfrac{1}{10}, \tfrac{1}{5})$$

... with $\Gamma, \Gamma', J_{⬡} = 0$

[Francini, Cônsoli, LJ, *in preparation*]

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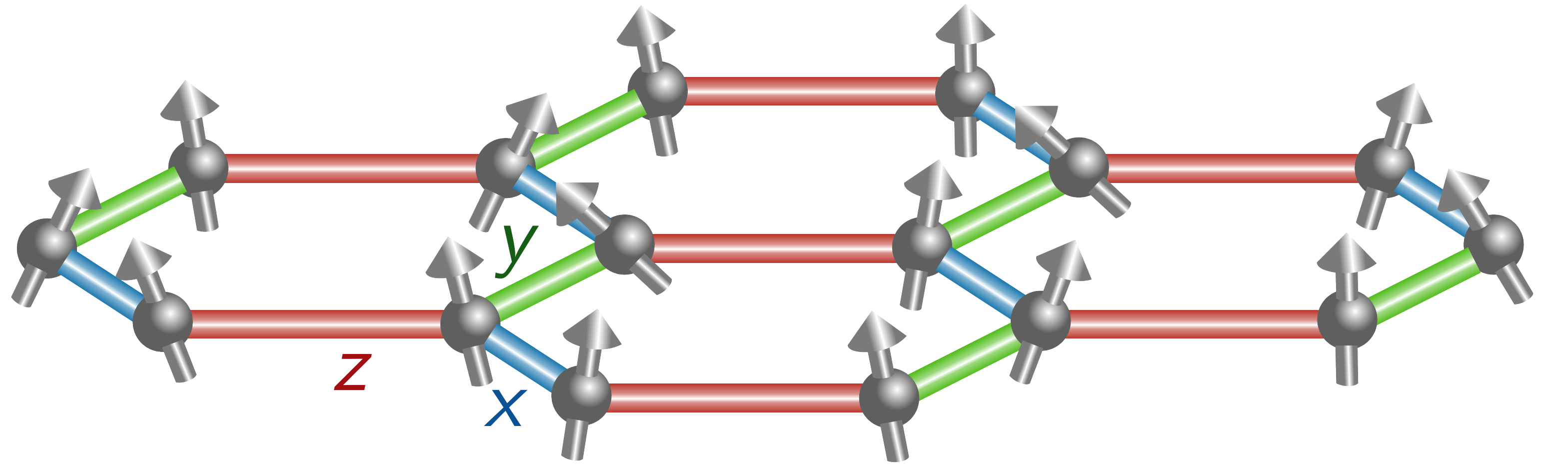

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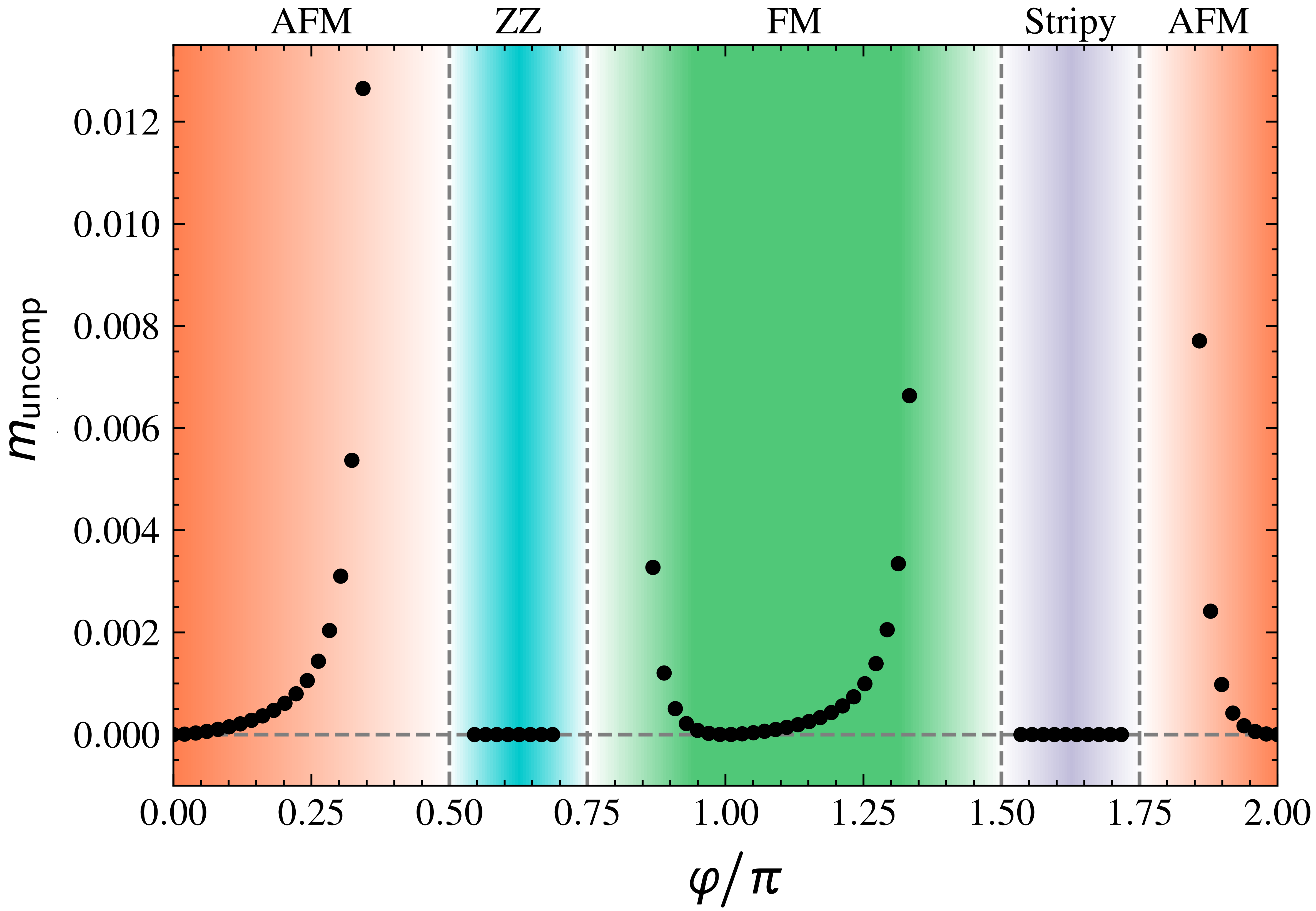
Uncompensated moments (linear spin-wave theory):



$$m_{\text{uncomp}} = \begin{cases} \frac{1}{2}|(\vec{S}_A + \vec{S}_B)\cdot\vec{c}| & \text{AFM} \\ \frac{1}{2}|(\vec{S}_A - \vec{S}_B)\cdot\vec{c}| & \text{FM} \end{cases}$$

... from $m_h(T=0) = -\frac{1}{N}\frac{\partial E_{\text{gs}}}{\partial h}$

[Francini, Cônsoli, LJ, *in preparation*]

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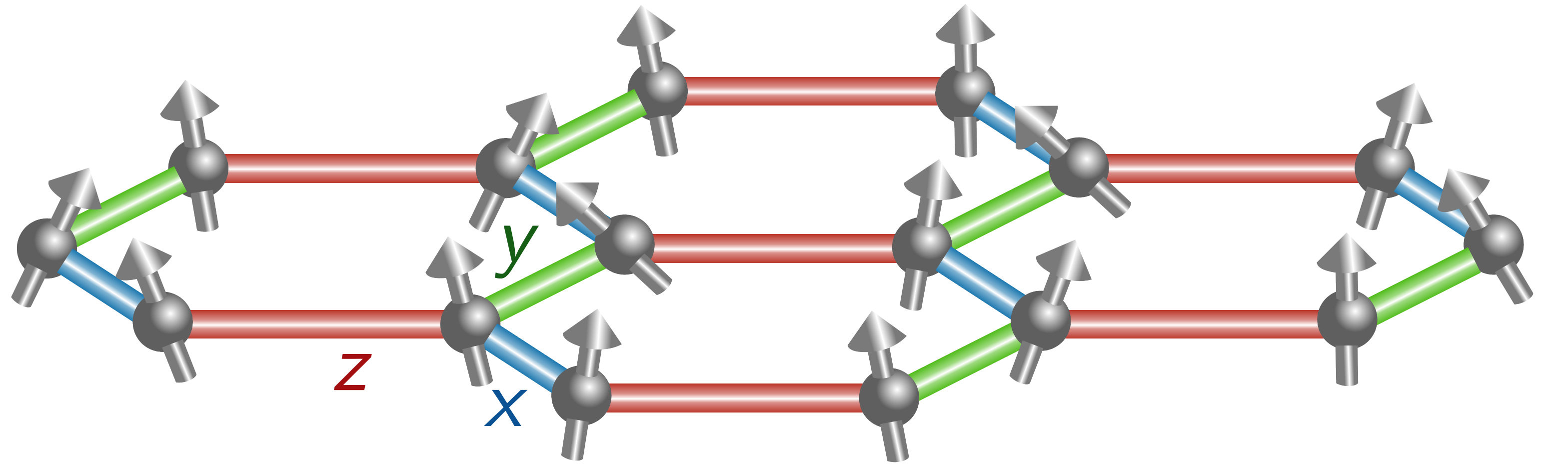

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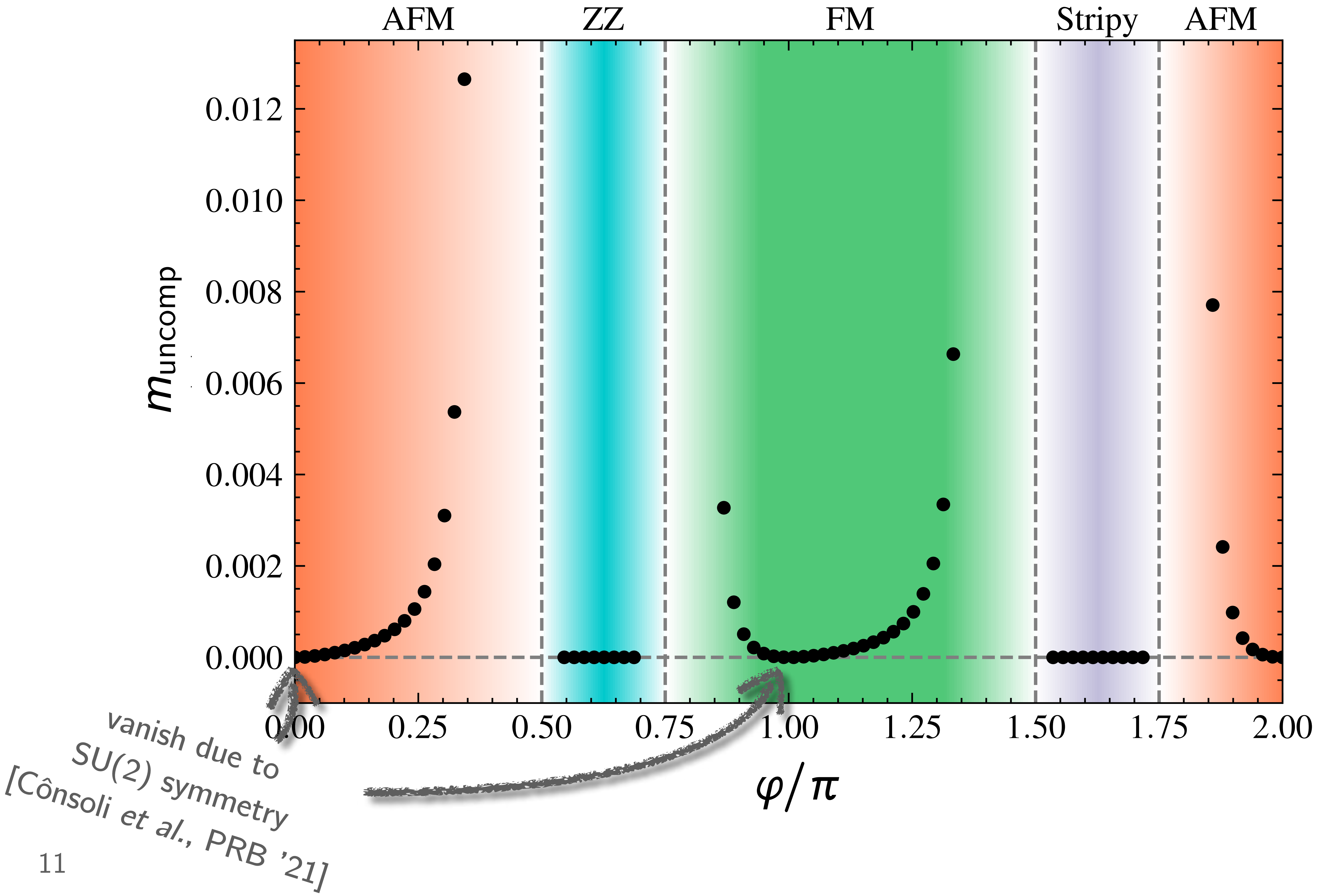

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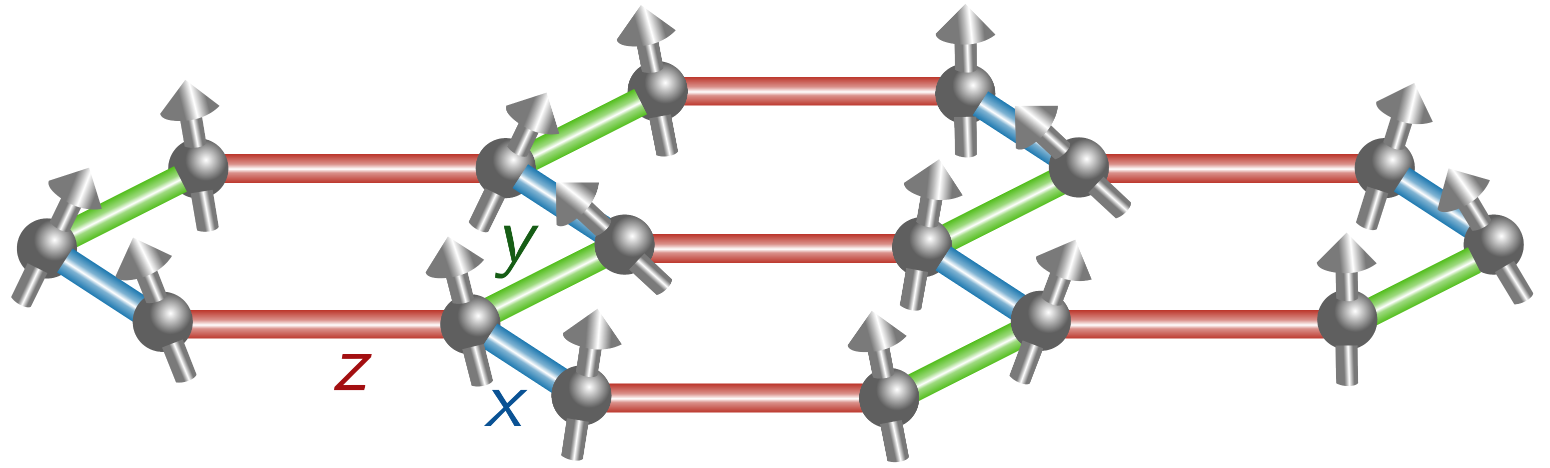

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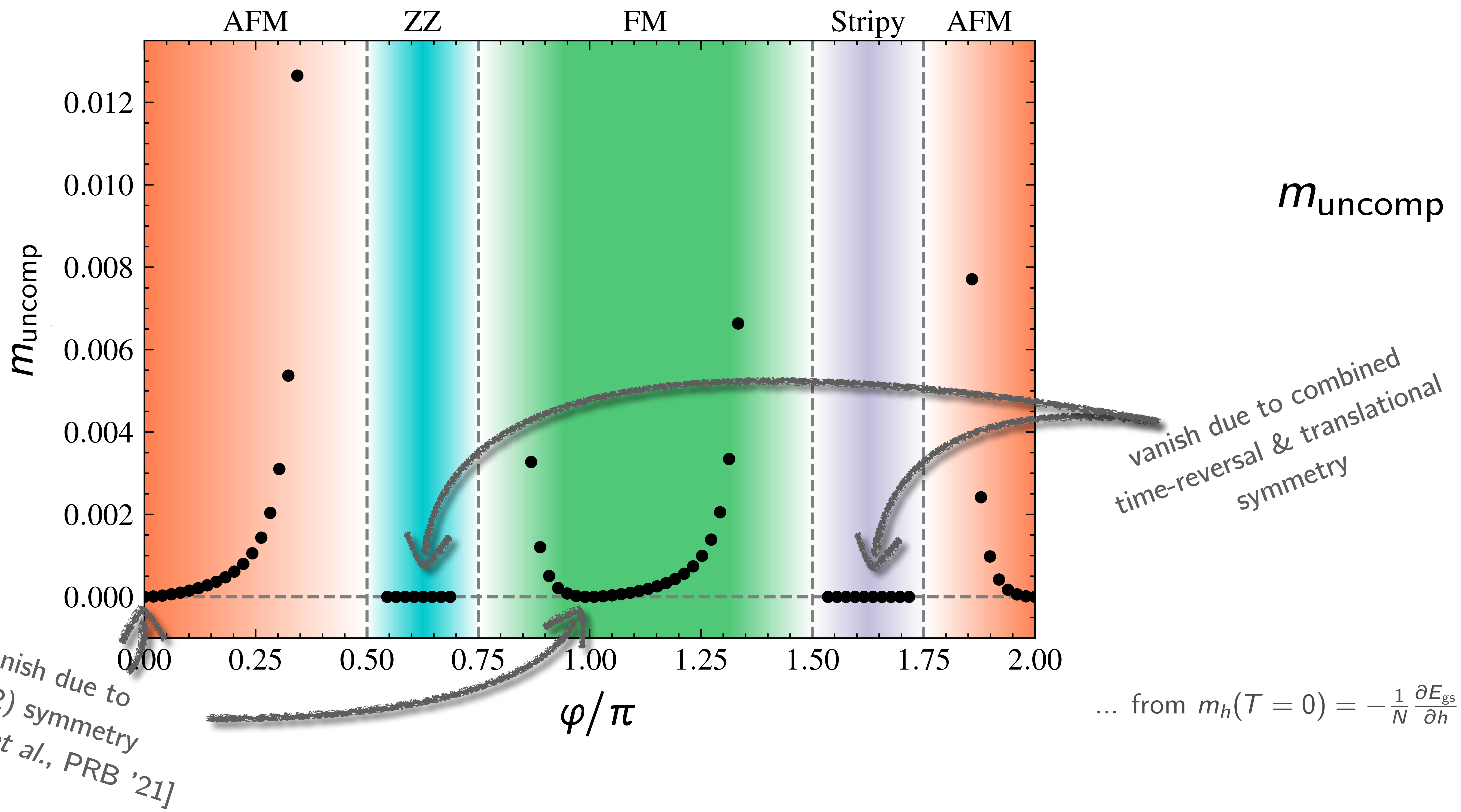

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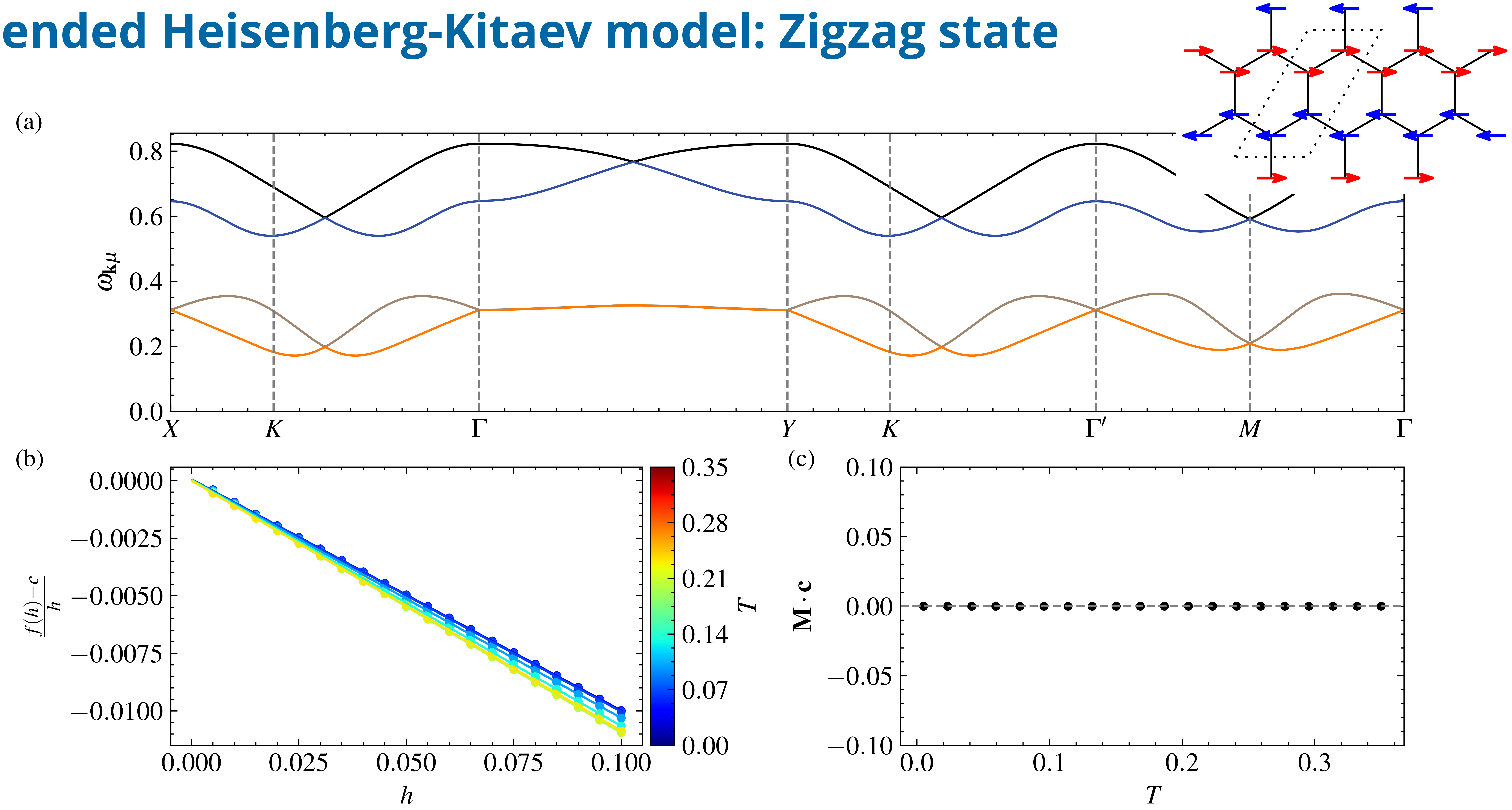
[Francini, Cônsoli, LJ, *in preparation*]

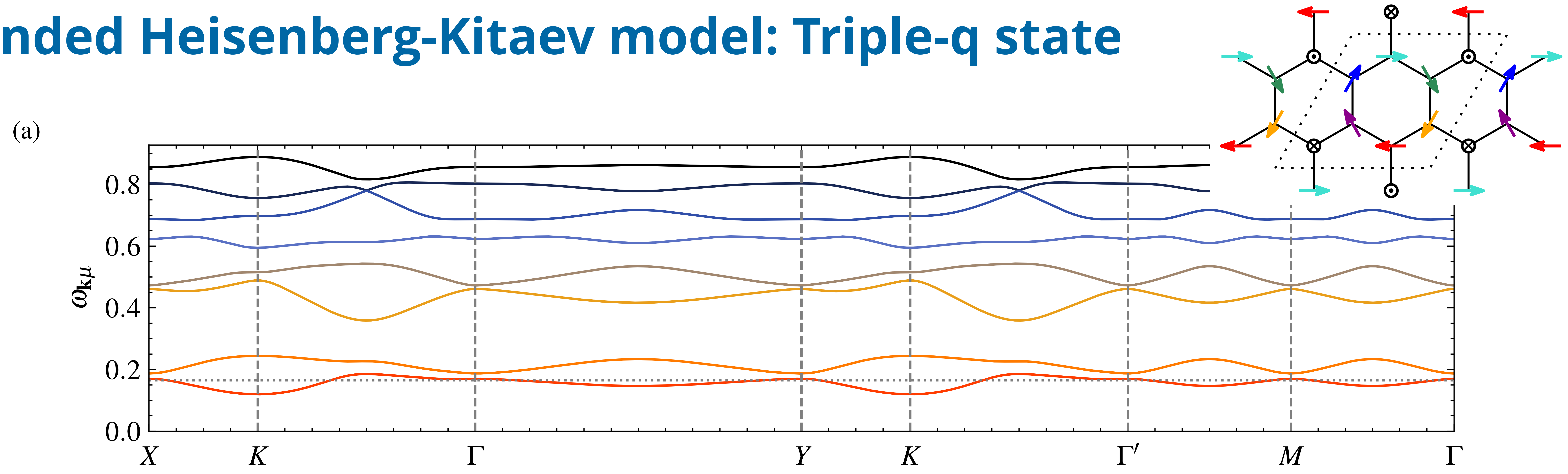
Extended Heisenberg-Kitaev model: Zigzag state

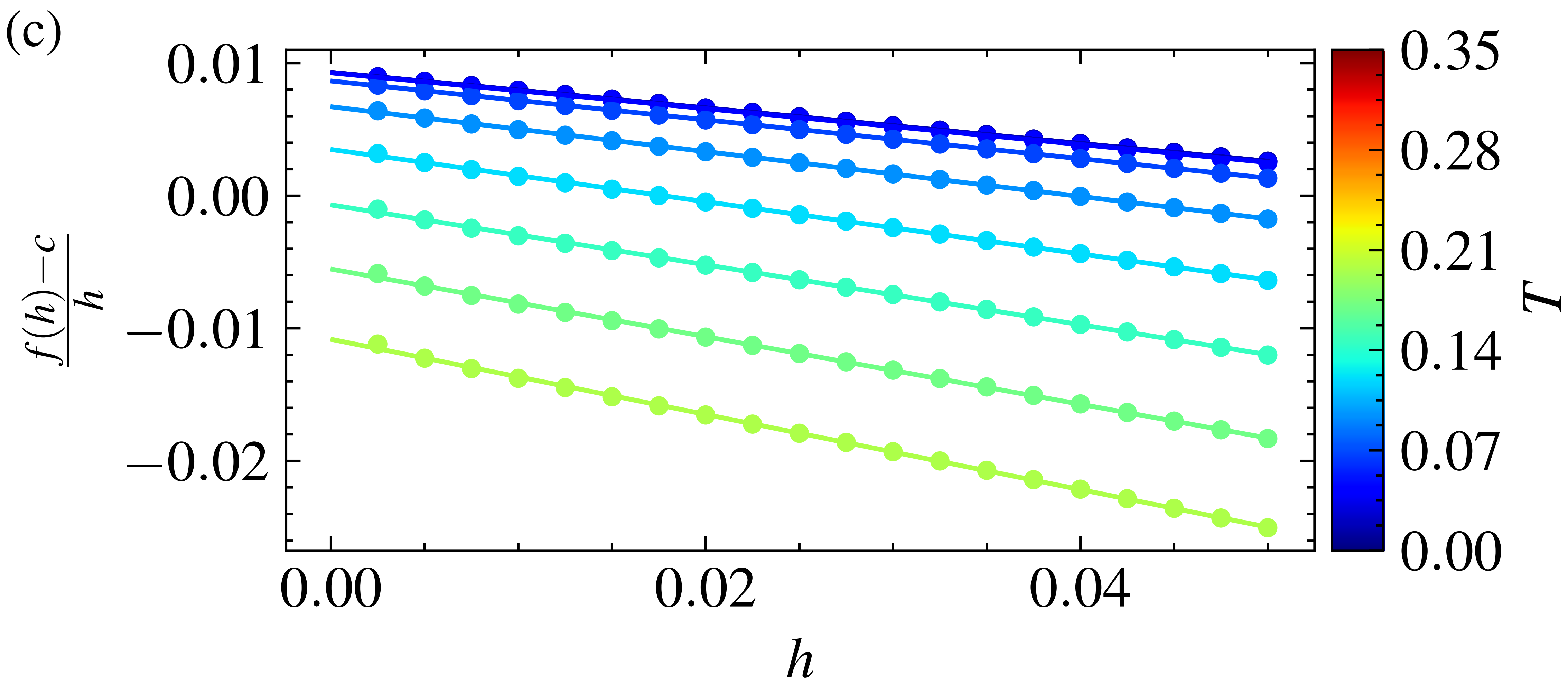


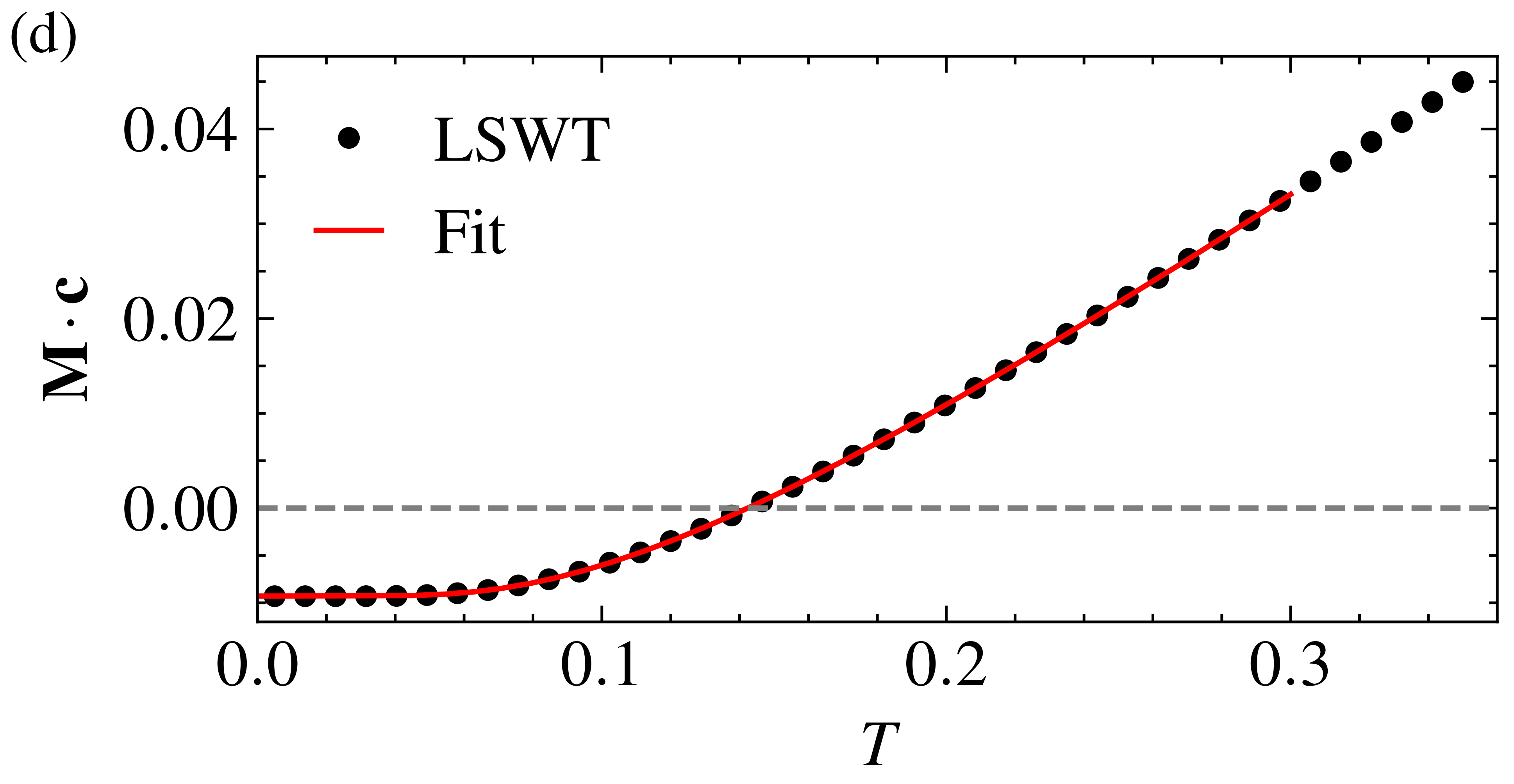
... using $(J, K, \Gamma, J_2^A, J_2^B) = J_0(-\sin(\frac{\pi}{16})/\sqrt{2}, -\sin(\frac{\pi}{16})/\sqrt{2}, \cos(\frac{\pi}{16}), -\frac{1}{10}, \frac{1}{5})$

... with $\Gamma', J_{\hexagon} = 0$

Extended Heisenberg-Kitaev model: Triple-q state







... using $(J, K, \Gamma, J_2^A, J_2^B) = J_0(-\sqrt{\frac{3}{5}}/2, \sqrt{\frac{3}{5}}, \frac{1}{2}, -\frac{1}{10}, \frac{1}{5})$

... with $\Gamma' = 0$ and $h_{\text{loc}} = \frac{2}{5} J_0 S$, mimicking $J_{\hexagon} < 0$

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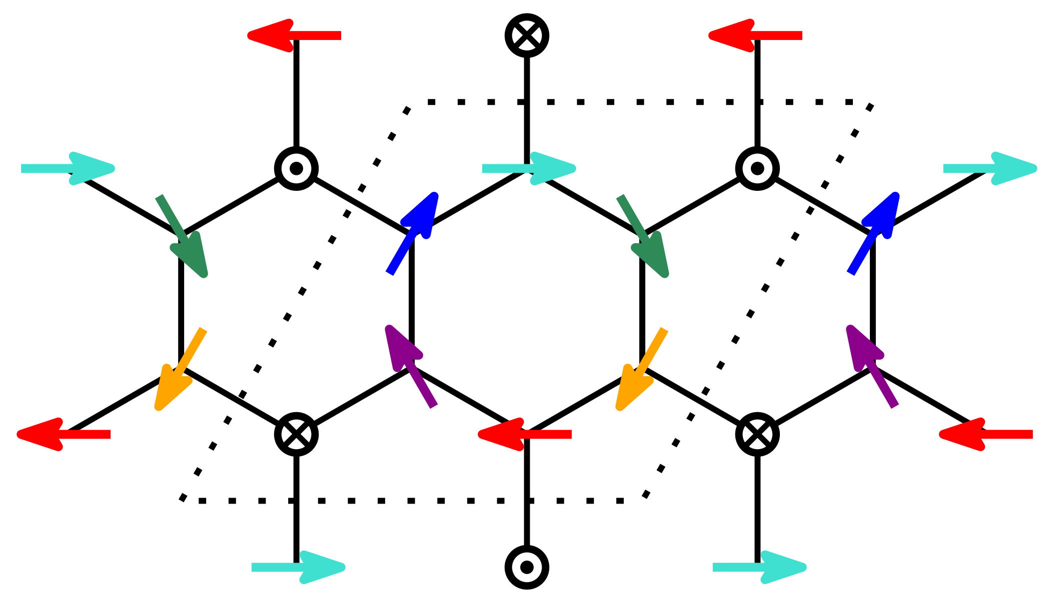

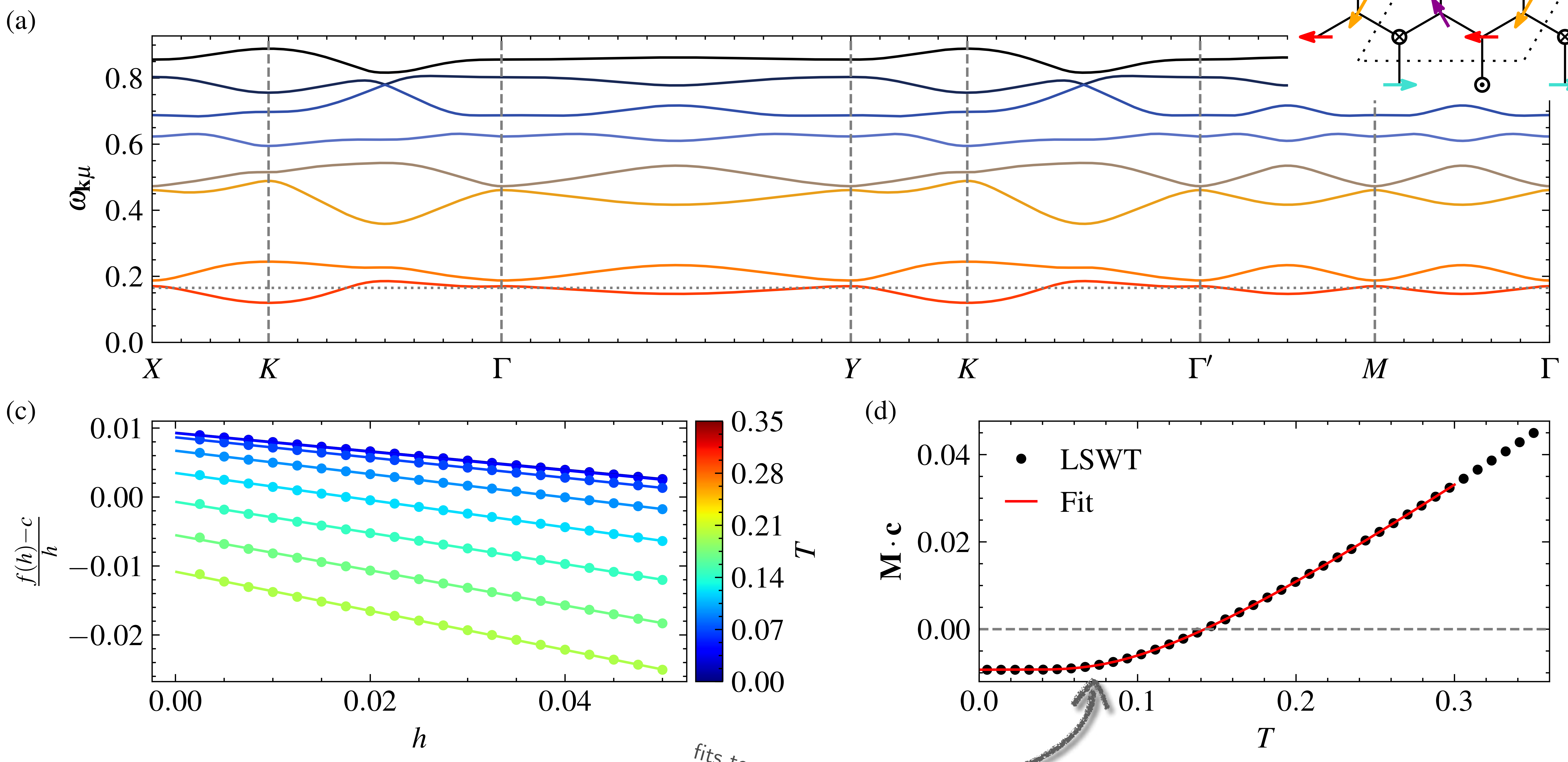

fits to $m(T) = m_0 + \frac{a}{e^{2\Delta/T}-1}$ with effective magnon gap Δ

… using $(J, K, \Gamma, J_2^A, J_2^B) = J_0(-\sqrt{\frac{3}{5}}/2, \sqrt{\frac{3}{5}}, \frac{1}{2}, -\frac{1}{10}, \frac{1}{5})$

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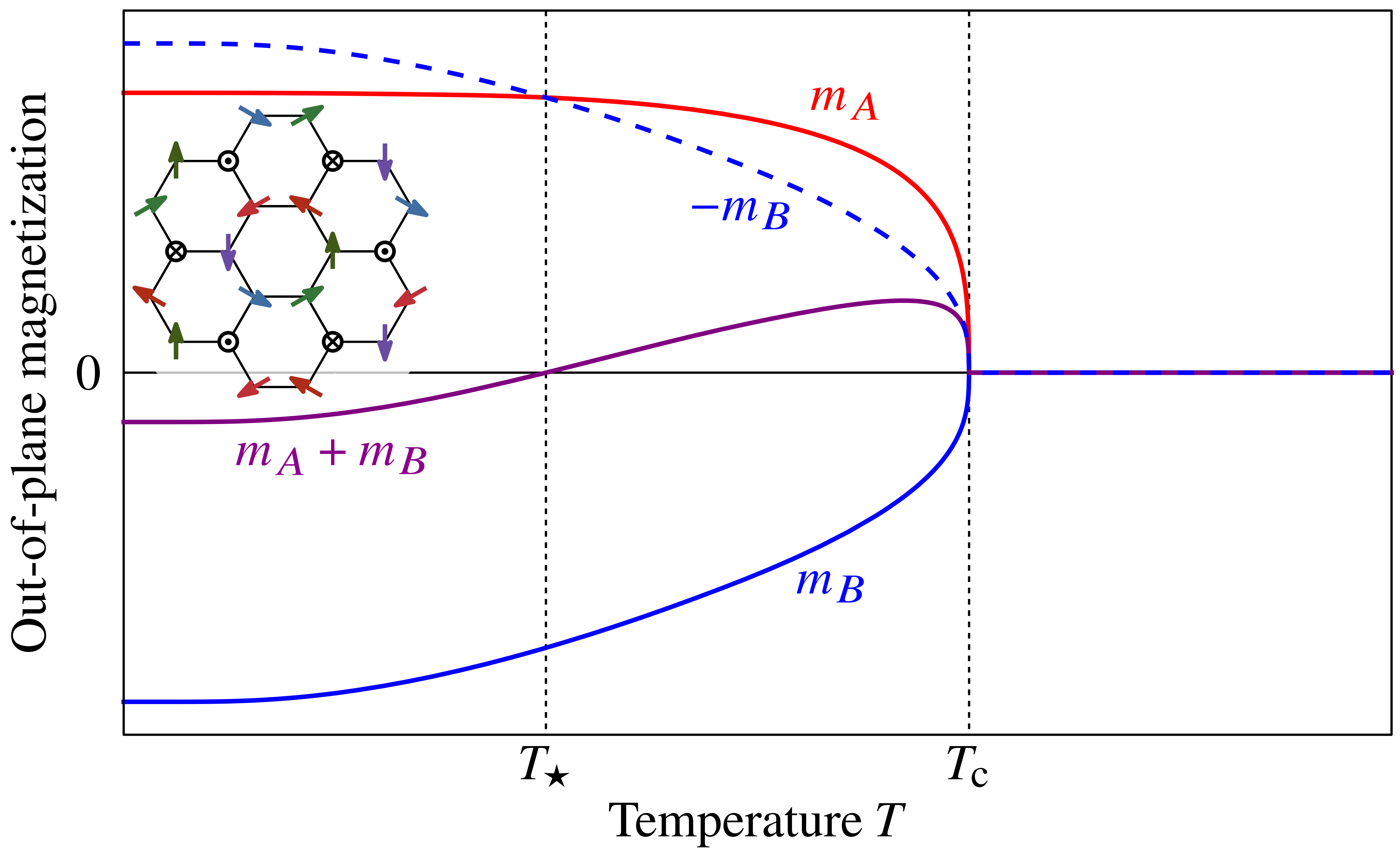
Summary: Ferrimagnetism from triple-q order in $Na_2Co_2TeO_6$

Triple-**q** model



[Francini, LJ, PRB '24]
[Francini, Cônsoli, LJ, *in preparation*]

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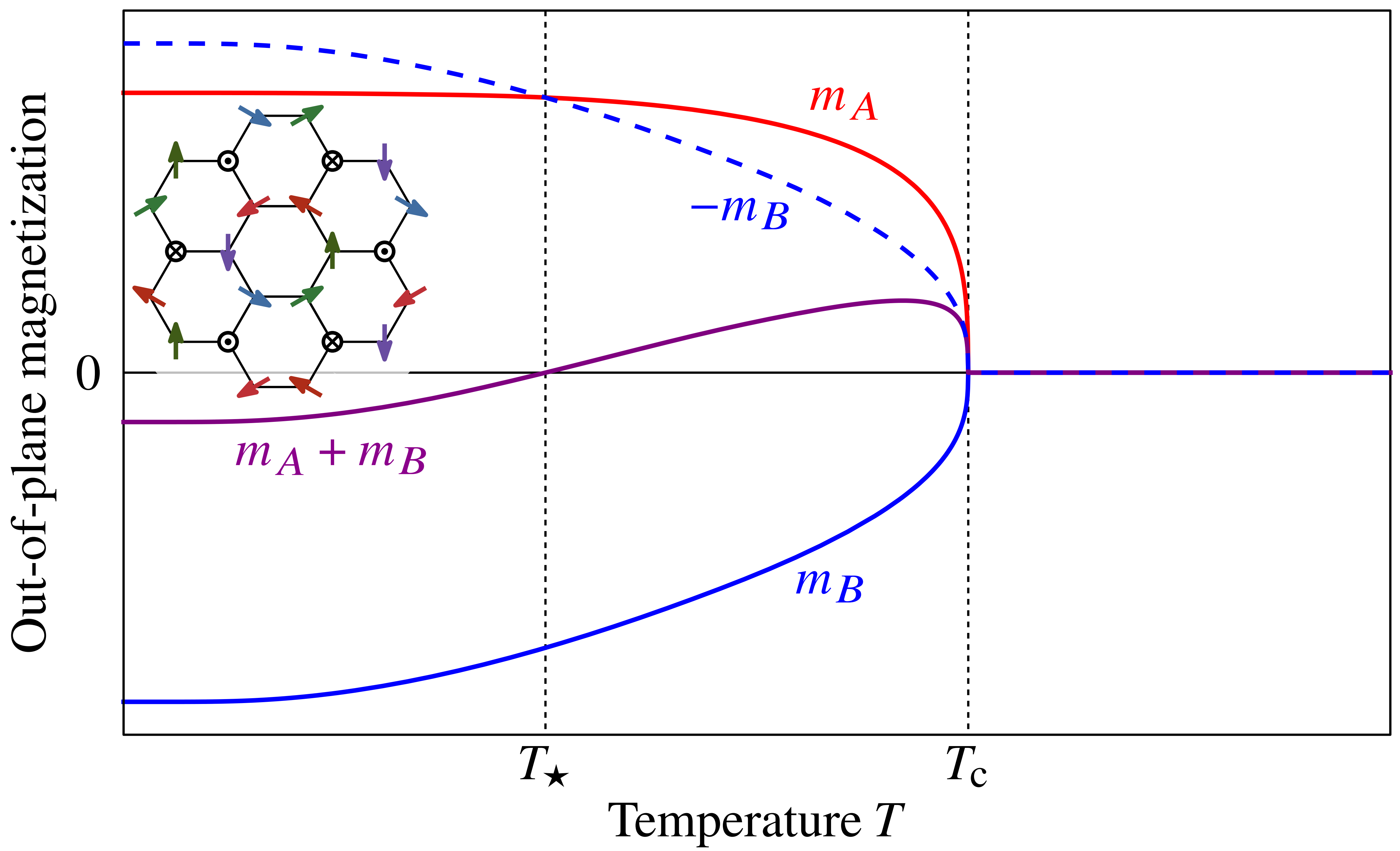
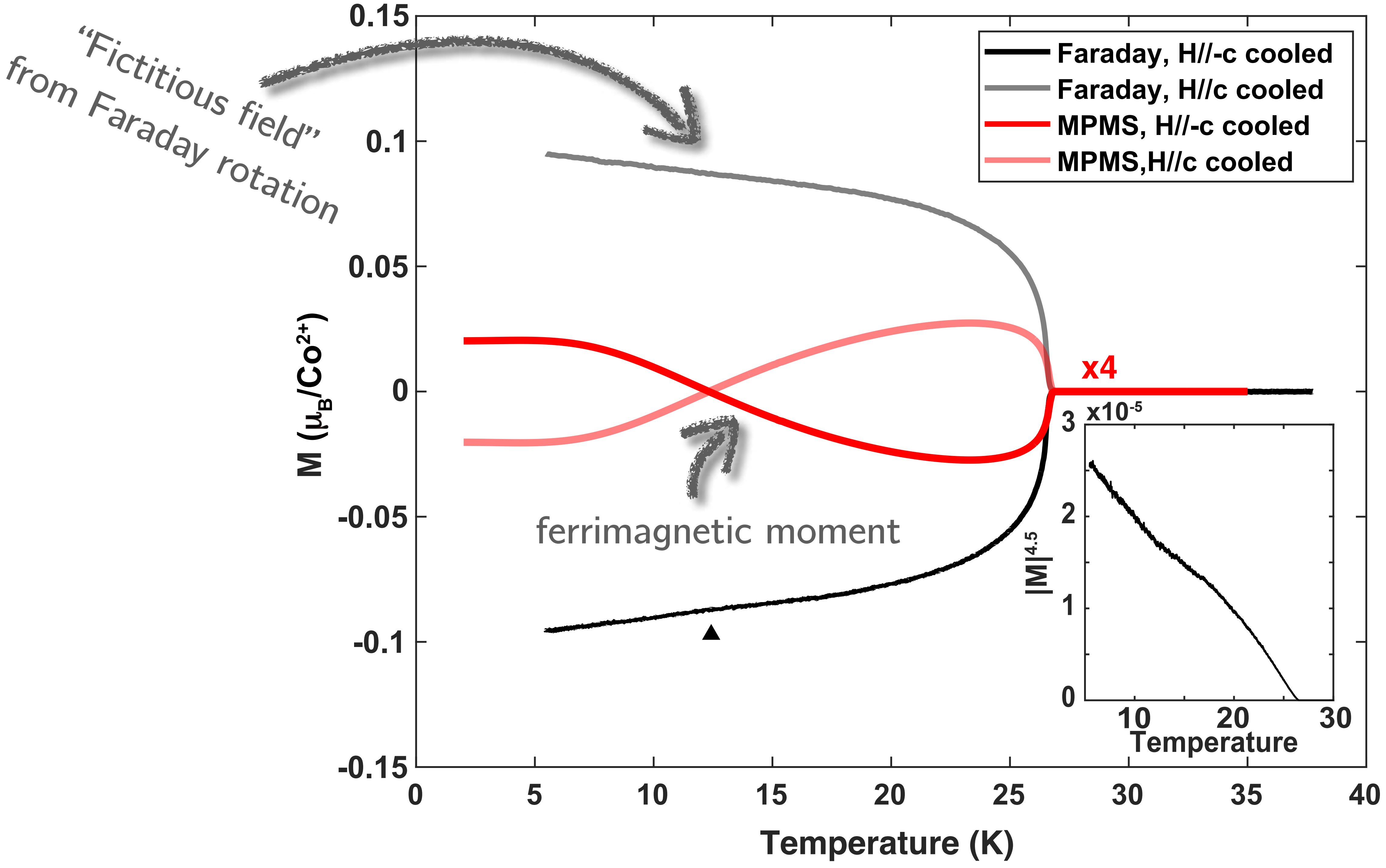


$Na_2Co_2TeO_6$



[Francini, LJ, PRB '24]
[Francini, Cônsoli, LJ, *in preparation*]

[Jin *et al.*, arXiv '25]

Ring exchange term in spin-wave theory

Ring exchange term equivalent to local term:

... at the level of linear spin-wave theory

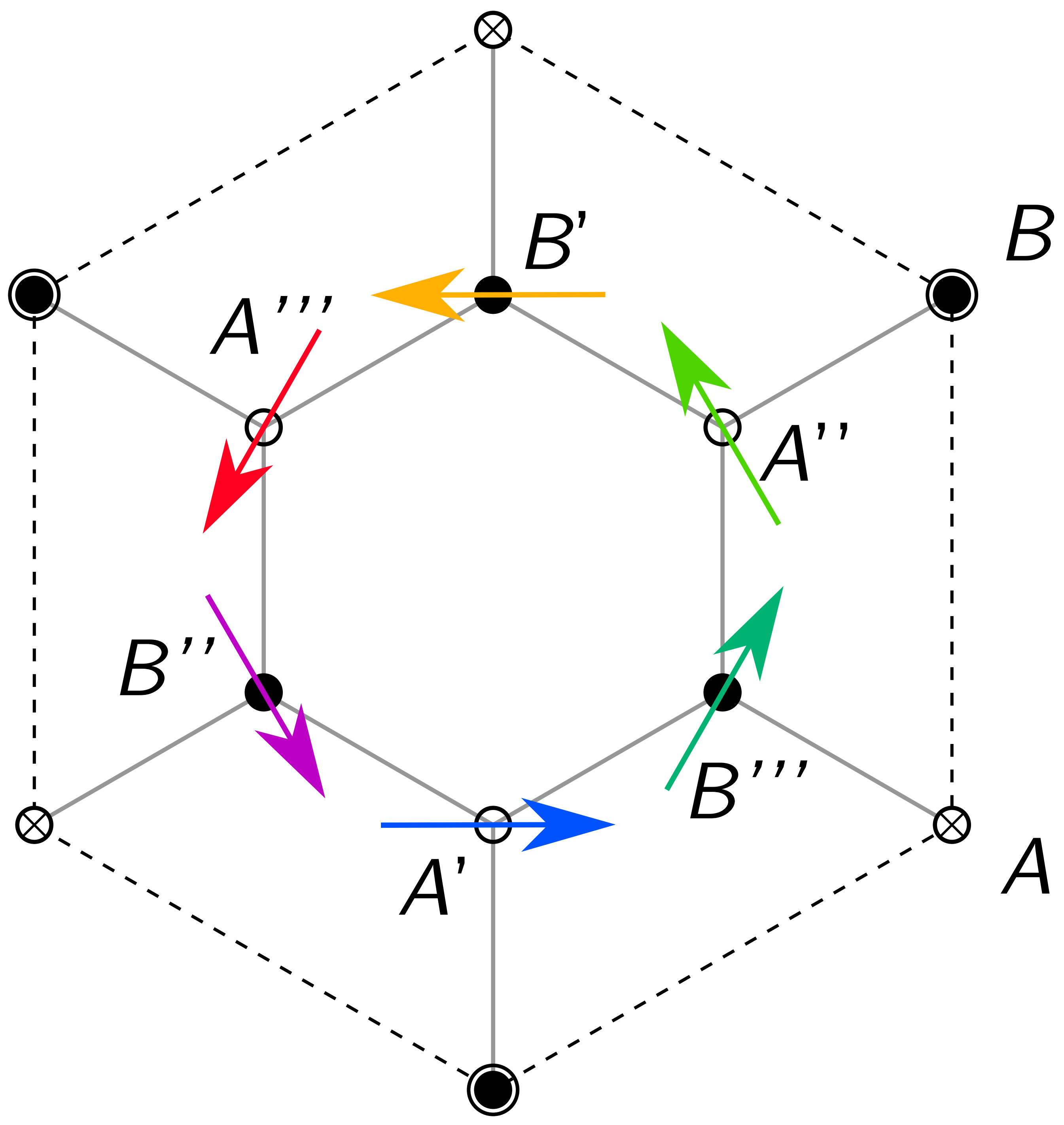
$$\mathcal{H}^{(3)}_{J_{\hexagon}} \simeq -h_{\text{loc}} \sum_i \hat{\mathbf{n}}_i \cdot \mathbf{S}_i$$

... up to renormalizations of bilinear couplings

with

$$\hat{\mathbf{n}}_A = -(1,1,1)/\sqrt{3} = -\hat{\mathbf{n}}_B$$
$$\hat{\mathbf{n}}_{A'} = (1,1,-1)/\sqrt{3} = -\hat{\mathbf{n}}_{B'}$$
$$\hat{\mathbf{n}}_{A''} = (-1,1,1)/\sqrt{3} = -\hat{\mathbf{n}}_{B''}$$
$$\hat{\mathbf{n}}_{A'''} = (1,-1,1)/\sqrt{3} = -\hat{\mathbf{n}}_{B'''}$$

... or C_2 rotated version



[Krüger, Chen, Jin, Li, LJ, PRL '23]