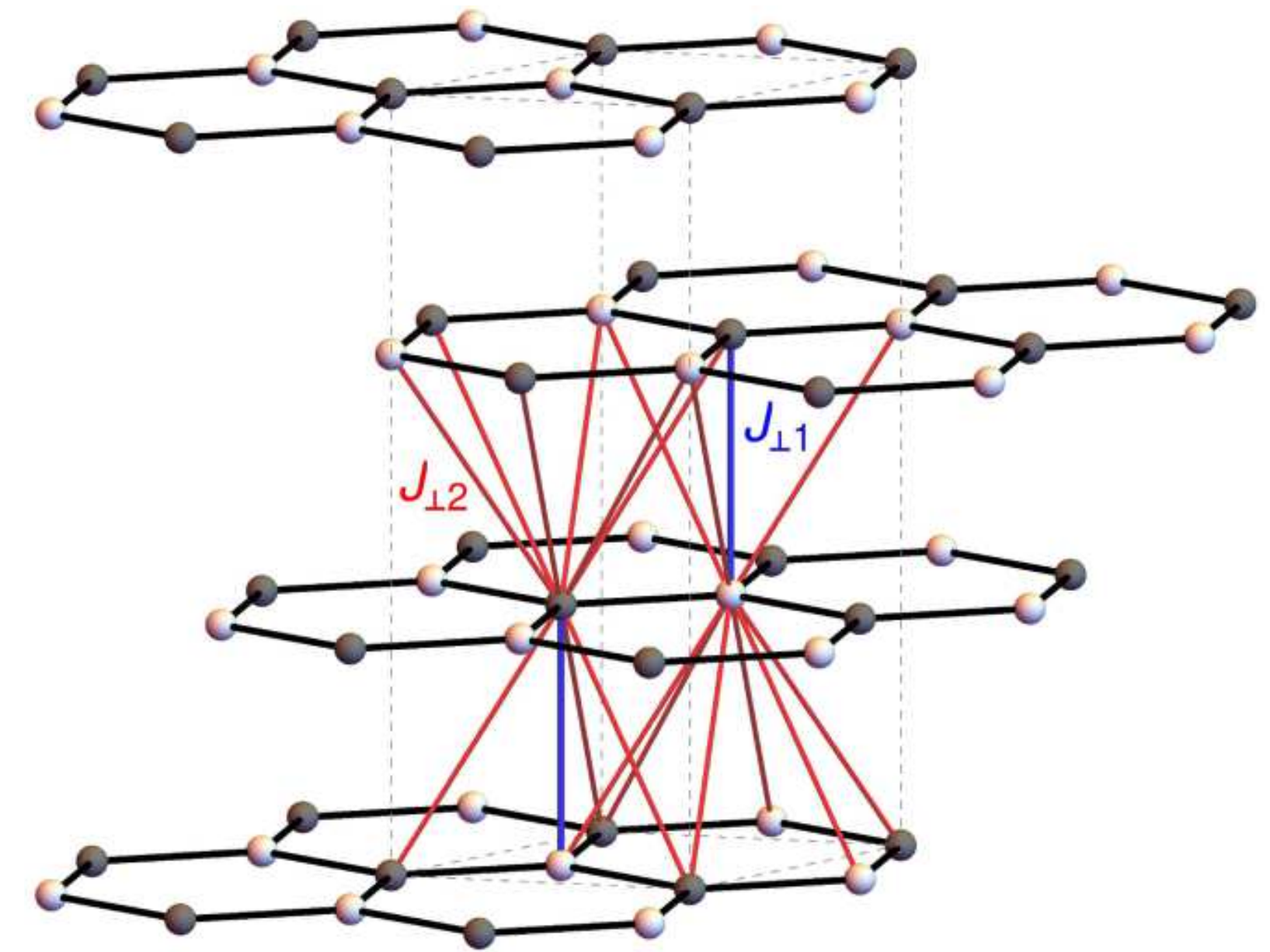


# Interlayer interactions in $\alpha$ - $\text{RuCl}_3$

Lukas Janssen  
TU Dresden



## Theory:

S. Koch

M. Vojta

P. M. C $\hat{o}$ nsoli

E. C. Andrade

## Experiments:

C. Balz

P. Lampen-Kelley

S. E. Nagler

S. Gass

A. U. B. Wolter

B. B $\ddot{u}$ chner

R. Hentrich

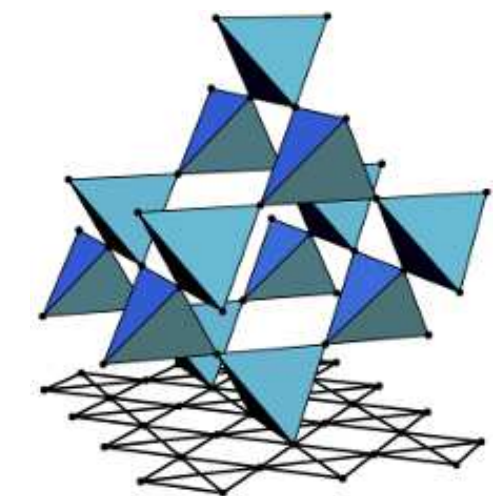
C. Hess



ct.qmat

Complexity and Topology  
in Quantum Matter

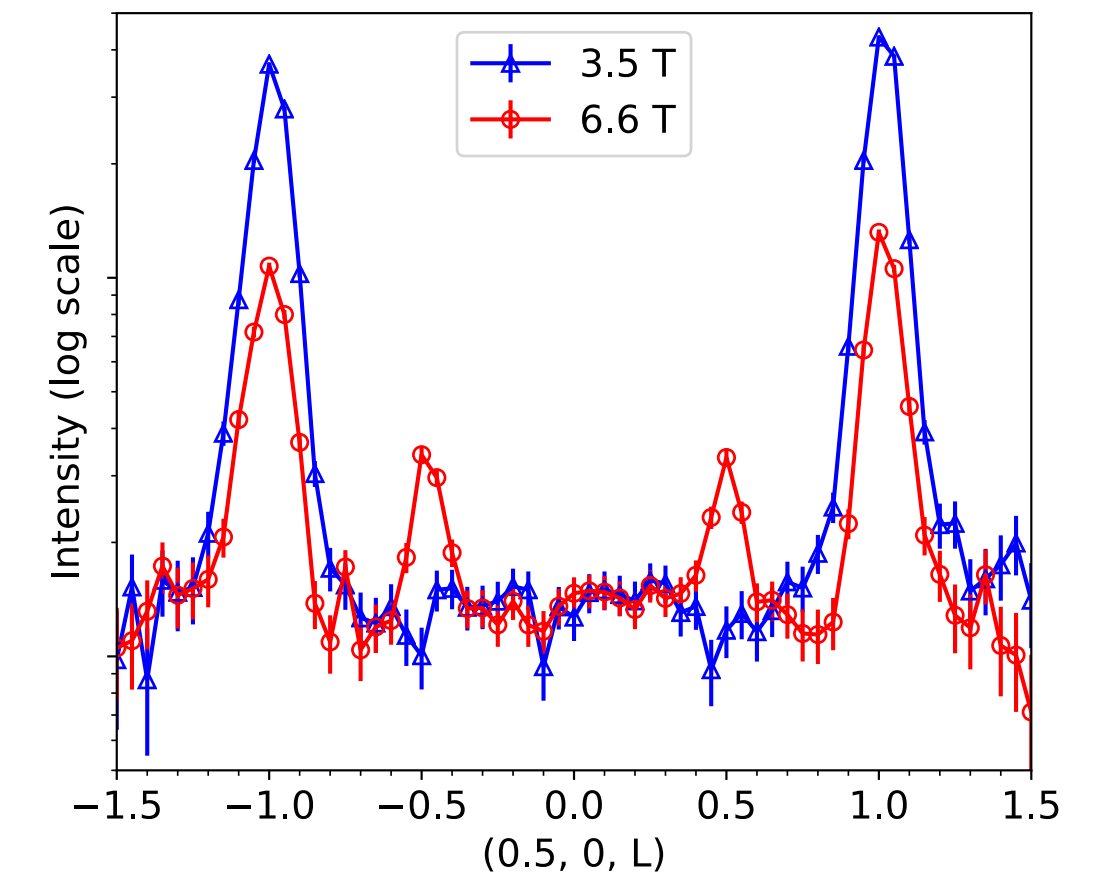
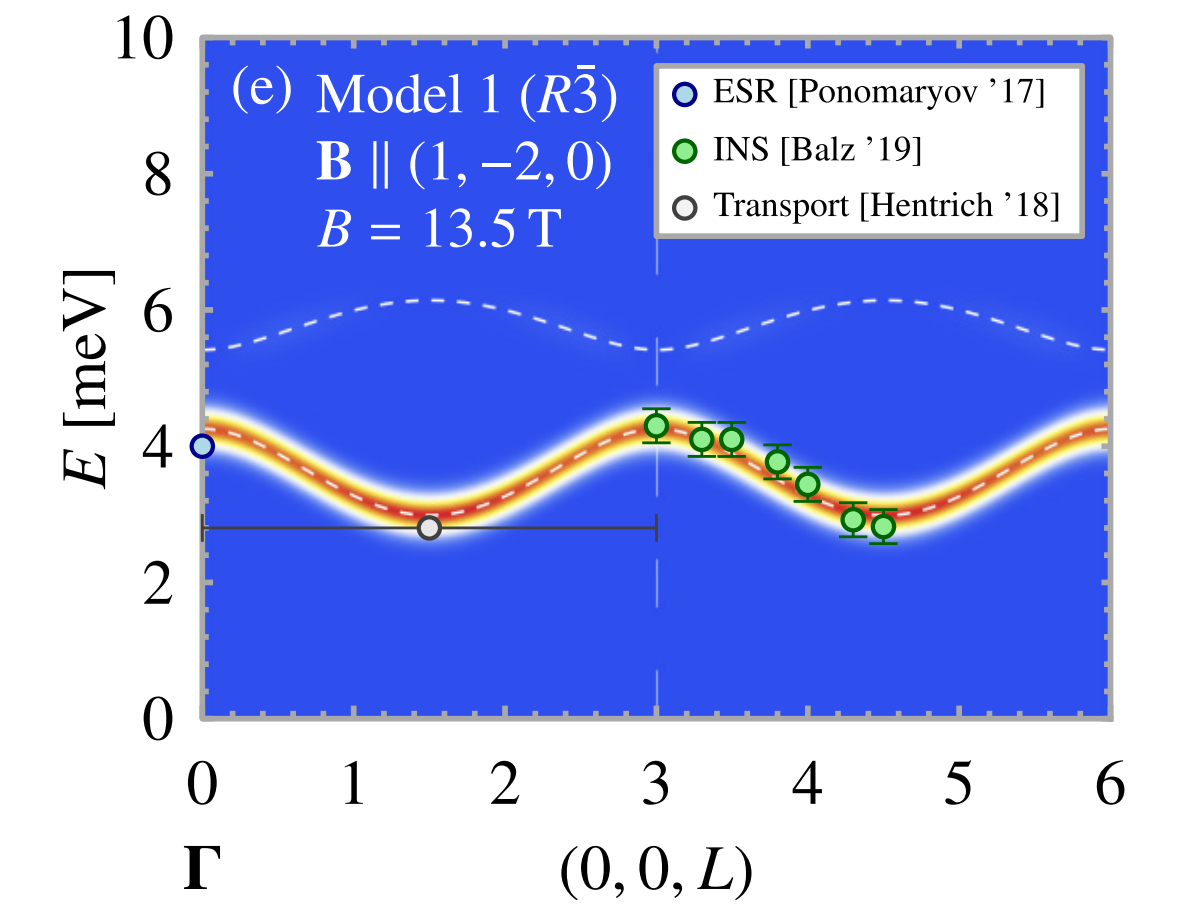
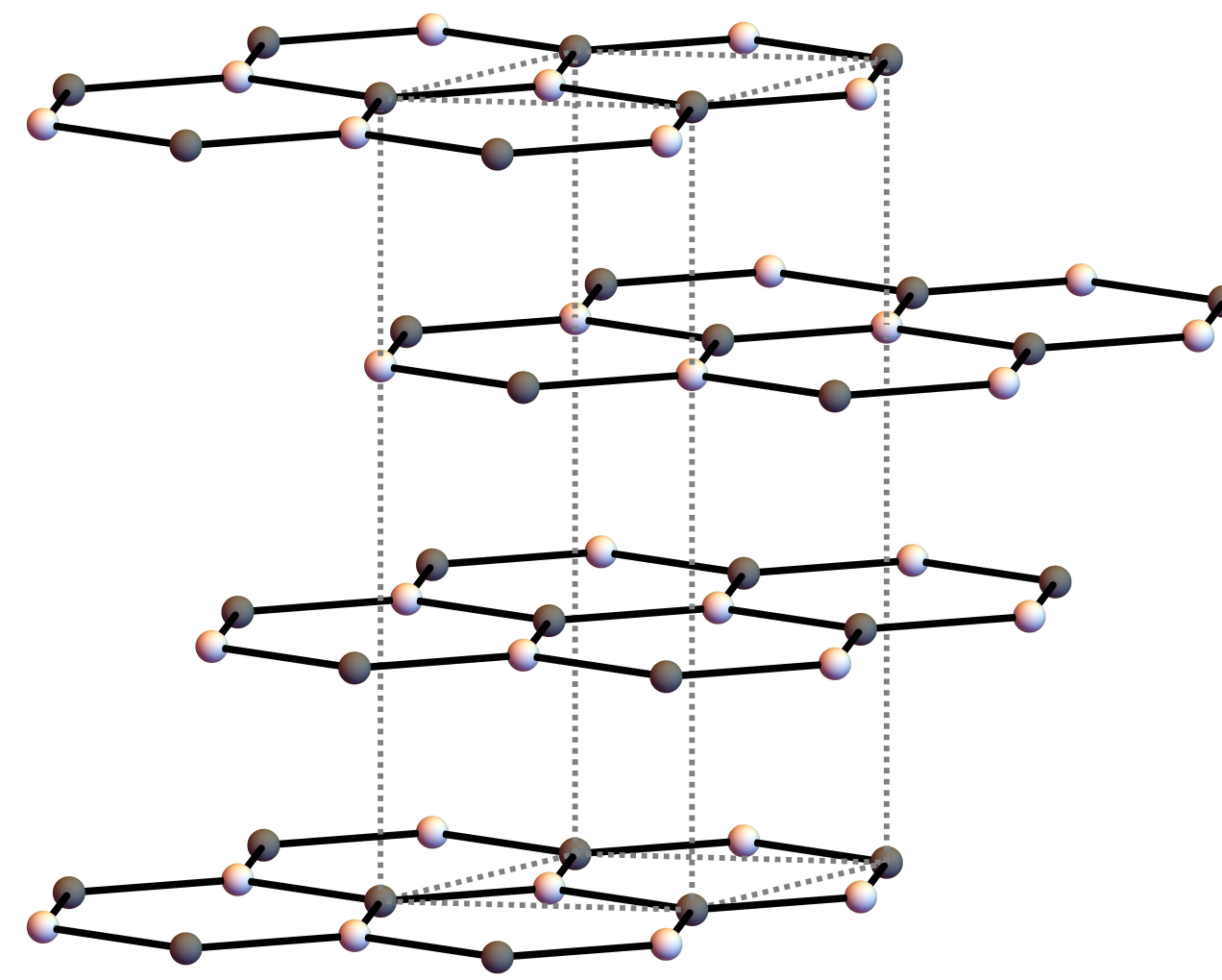
Würzburg-Dresden Cluster of Excellence



SFB 1143

# Outline

1. Introduction: Modeling  $\alpha$ -RuCl<sub>3</sub>
2. Evidence #1: Excitation spectra
3. Evidence #2: Field-induced intermediate order
4. Conclusions



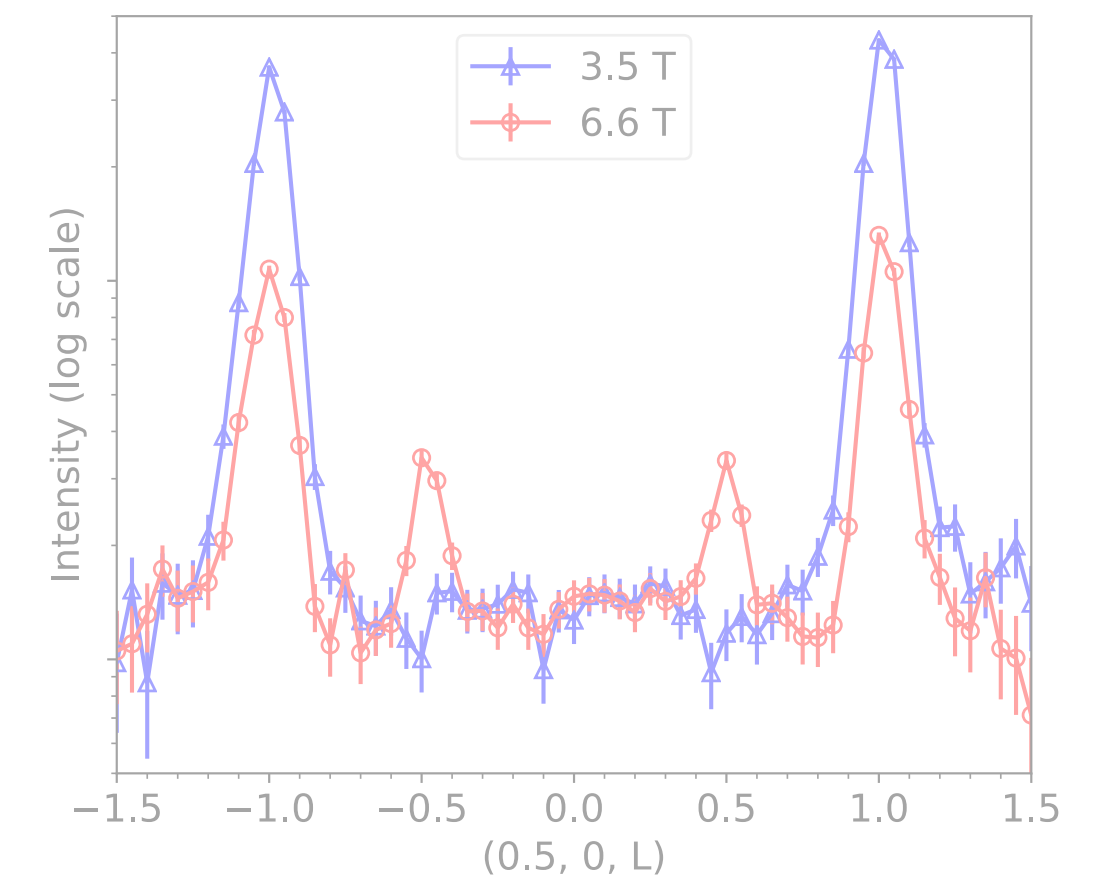
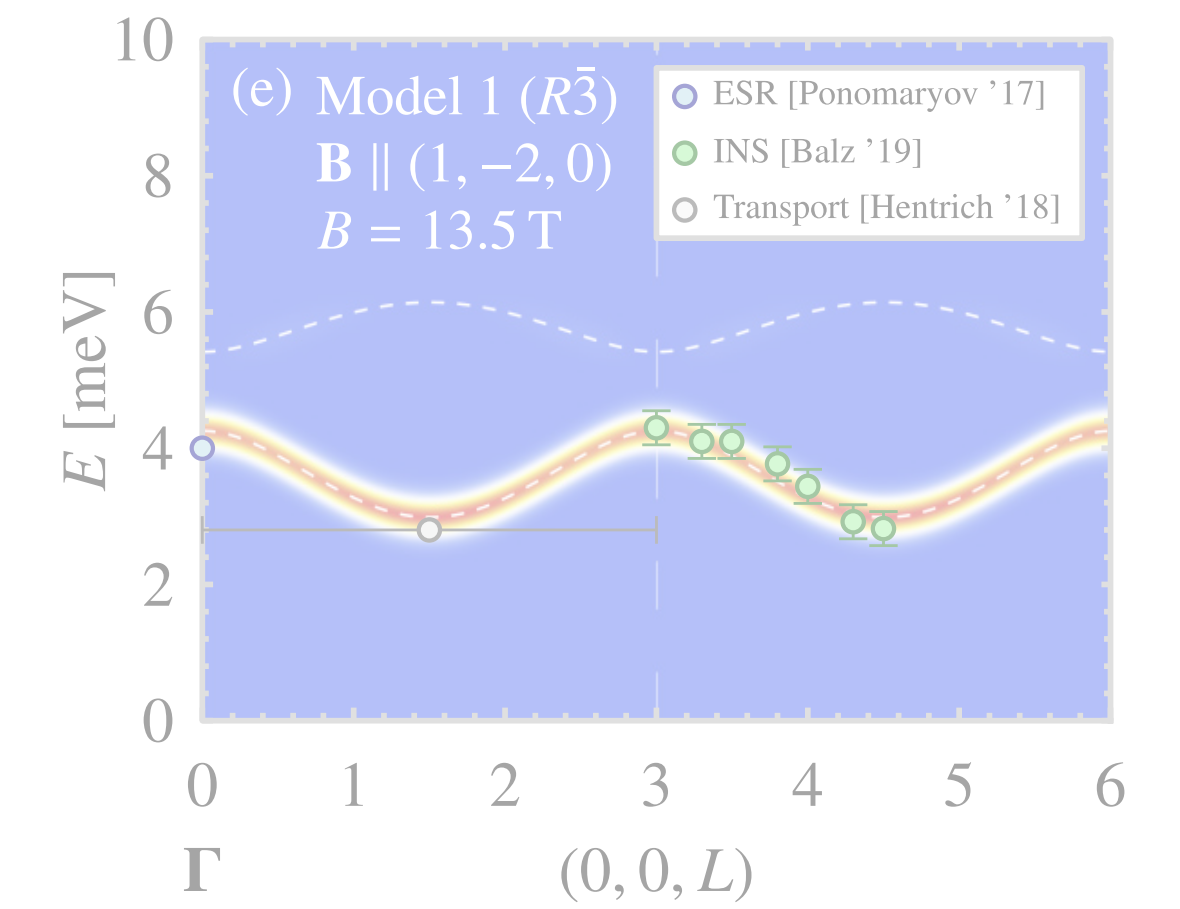
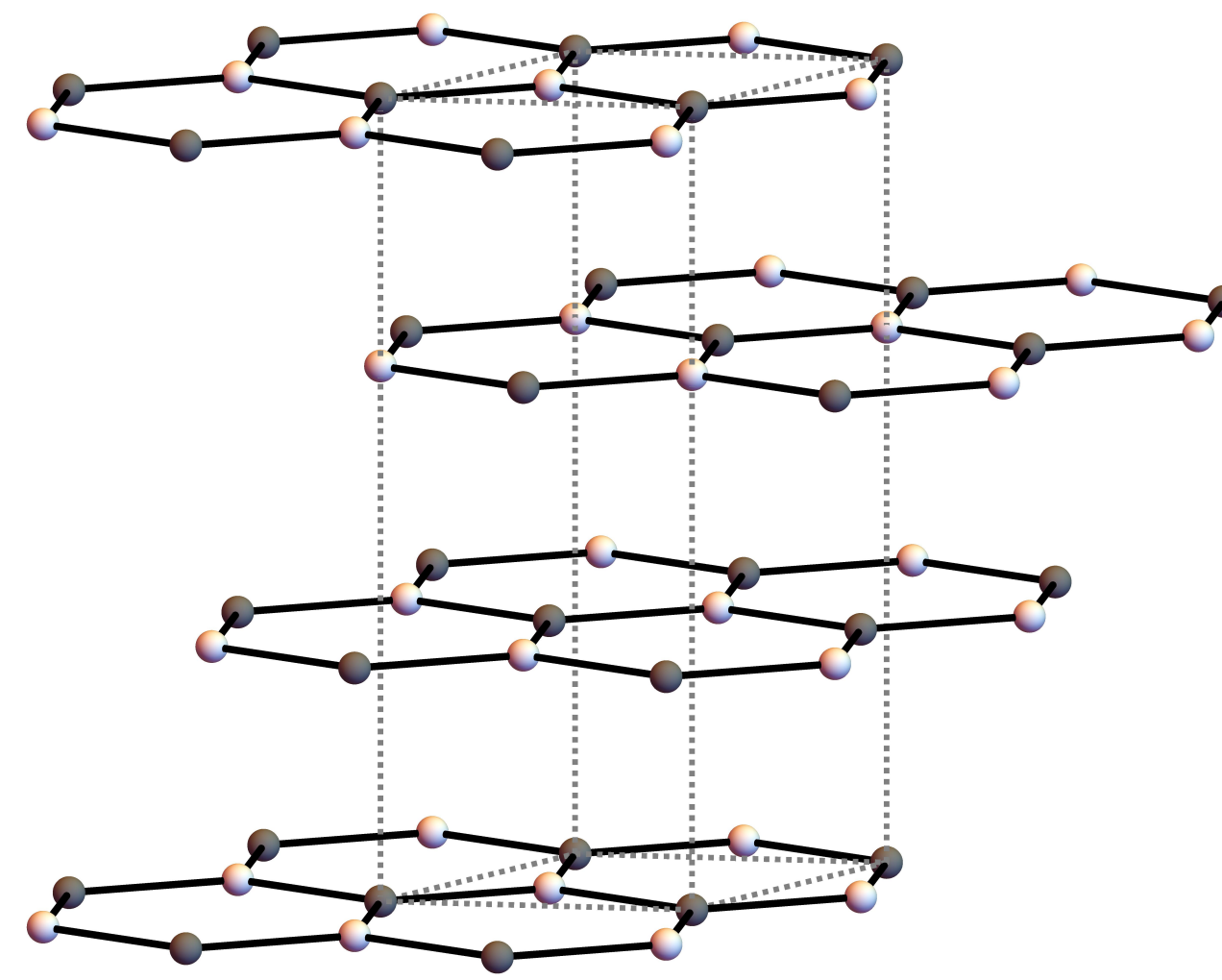
# Outline

1. Introduction: Modeling  $\alpha$ -RuCl<sub>3</sub>

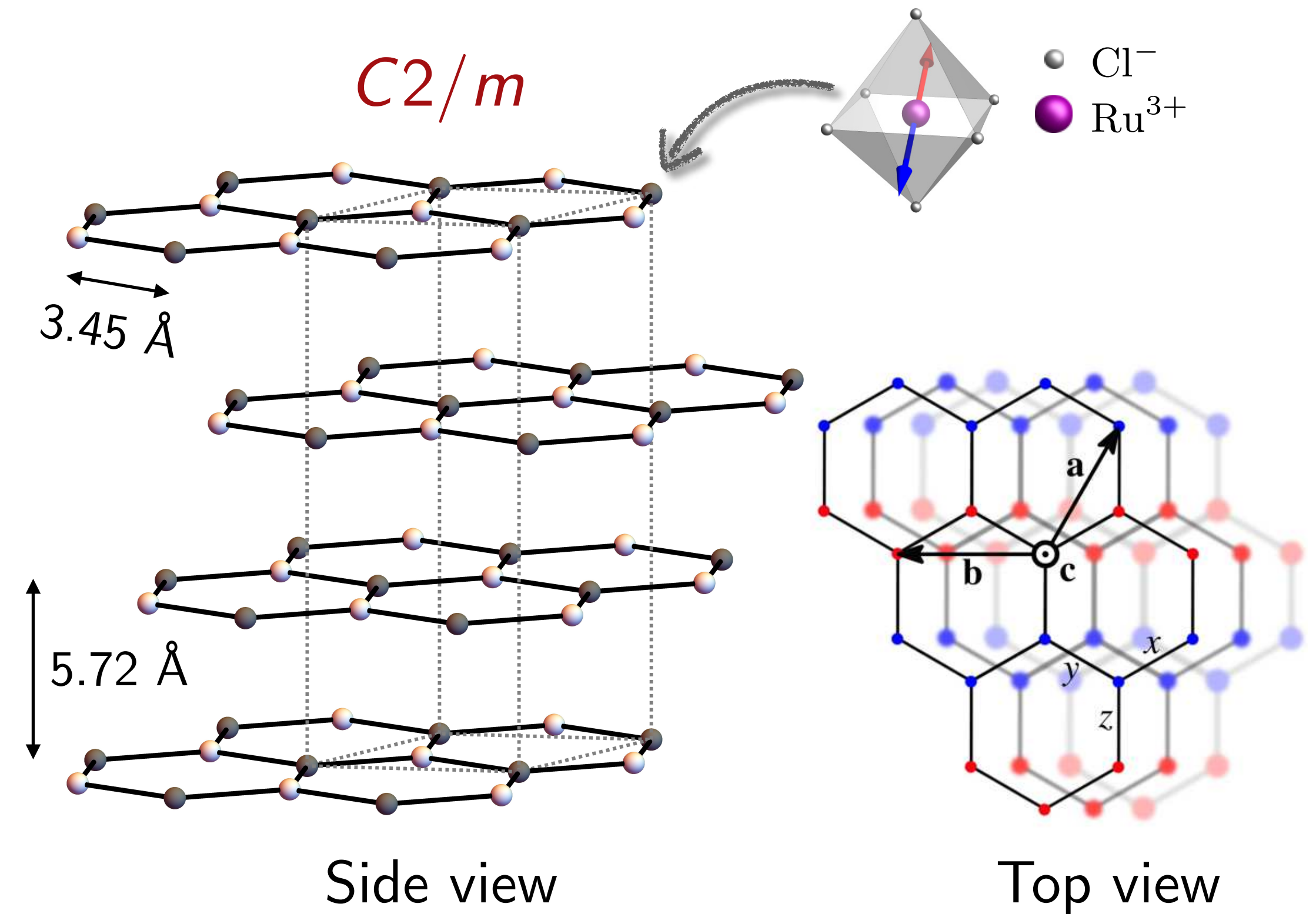
2. Evidence #1: Excitation spectra

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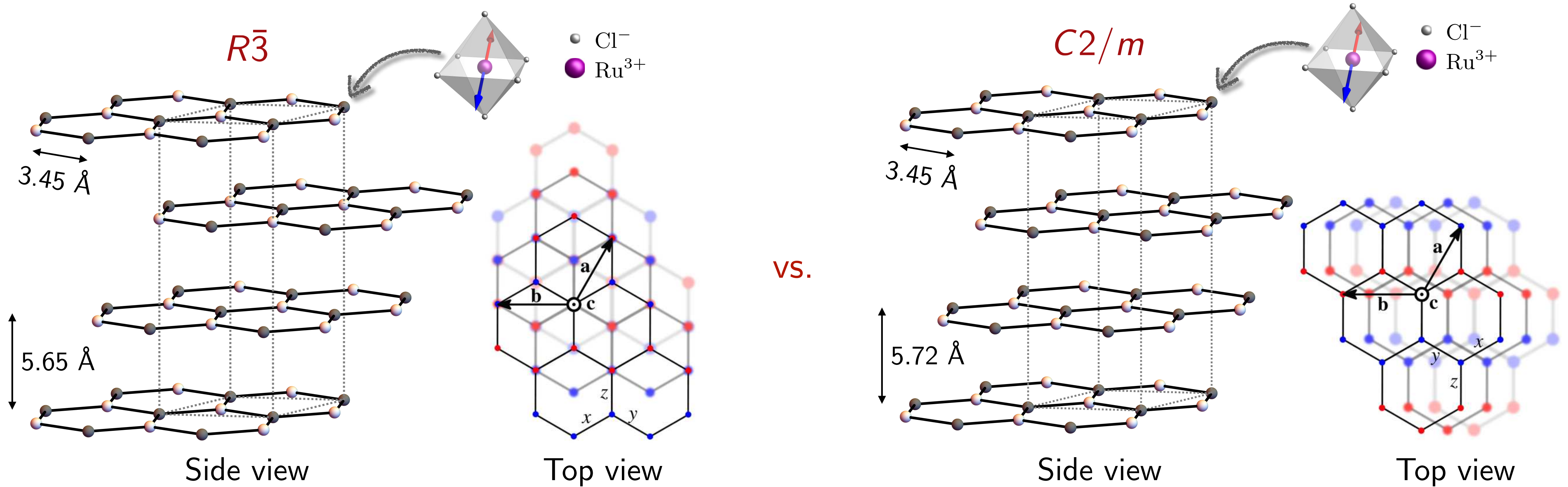
# $\alpha$ -RuCl<sub>3</sub>: Crystal structure



[Johnson *et al.*, PRB '15]

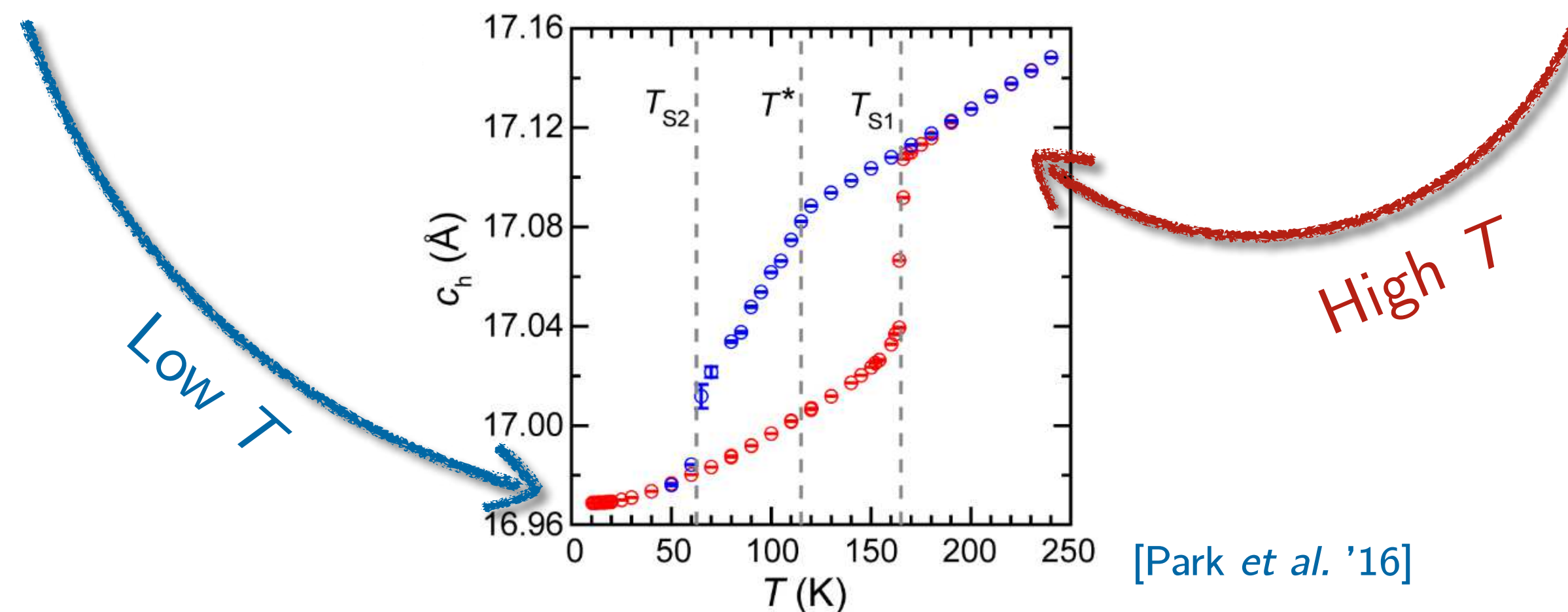
[Cao *et al.*, PRB '16]

# $\alpha$ -RuCl<sub>3</sub>: Crystal structure



[Johnson *et al.*, PRB '15]

[Cao *et al.*, PRB '16]



[Park *et al.* '16]

[Glamazda *et al.*, PRB '17]

[Do *et al.*, Nat. Phys. '17]

[Lampen-Kelley *et al.*, PRB '18]

[Gass, C onsoli, ..., LJ, *et al.*, PRB '20]

# $\alpha$ -RuCl<sub>3</sub>: Effective spin models

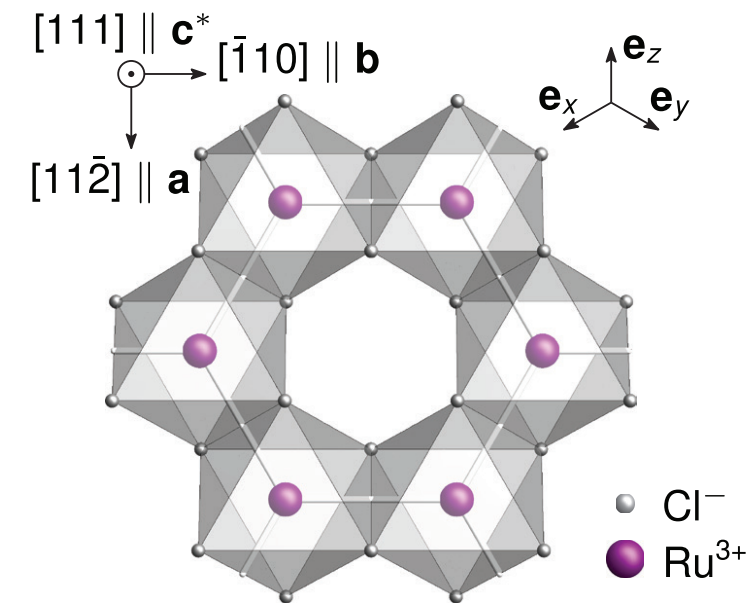
2D Hamiltonians:

$$\mathcal{H}_{2D} = \sum_{\langle ij \rangle} \left[ J_1 \vec{S}_i \cdot \vec{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \Gamma'_1 (S_i^\gamma S_j^\alpha + S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\beta + S_i^\beta S_j^\gamma) \right]$$

$$+ \sum_{\langle\langle ij \rangle\rangle} \left( J_2 \vec{S}_i \cdot \vec{S}_j + K_2 S_i^\gamma S_j^\gamma + \dots \right) + \sum_{\langle\langle\langle ij \rangle\rangle\rangle} \left( J_3 \vec{S}_i \cdot \vec{S}_j + \dots \right) + \dots$$

[Jackeli, Khaliullin, PRL '09]

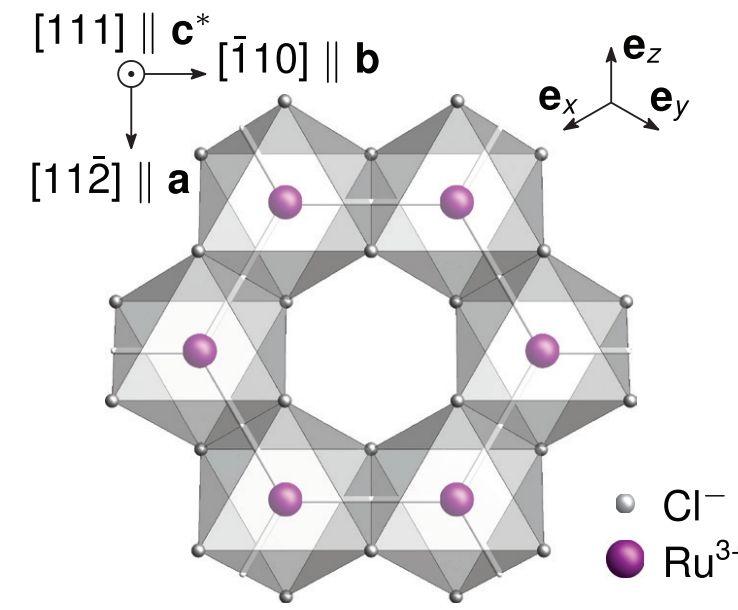
...



# $\alpha$ -RuCl<sub>3</sub>: Effective spin models

2D Hamiltonians:

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[Jackeli, Khaliullin, PRL '09]

Exchange couplings:

Set	Material	$J_1$ (meV)	$K_1$ (meV)	$\Gamma_1$ (meV)	$J_2$ (meV)	$K_2$ (meV)	$J_3$ (meV)	Method	Ref.	Year
1	$\alpha$ -RuCl <sub>3</sub>	-4.6	+7.0	-	-	-	-	Fit to neutron scattering	[35,36]	2016
1'	Na <sub>2</sub> IrO <sub>3</sub>	-4.0	+10.5	-	-	-	-	Fit to susceptibility and neutron scattering	[30]	2013
1 + $\Gamma$	$\alpha$ -RuCl <sub>3</sub>	-12	+17	+12	-	-	-	DFT + $t/U$ expansion	[44]	2015
2	Na <sub>2</sub> IrO <sub>3</sub>	0	-17	0	0	-	+6.8	DFT + exact diagonalization	[32]	2016
2 + $\Gamma$	Na <sub>2</sub> IrO <sub>3</sub>	+3	-17	+1	-3	+6	+1	DFT + $t/U$ expansion, direction of moments	[40,45]	2016
(2 + $\Gamma$ )'	Na <sub>2</sub> IrO <sub>3</sub>	+3	-17.5	+1	+5	-	+5	MRCI, fit to $\theta_{CW}$	[47]	2014
(2 + $\Gamma$ )''	$\alpha$ -RuCl <sub>3</sub>	+1.2	-5.6	+1	+0.3	-	+0.3	MRCI, fit to magnetization	[13]	2016
2/3	$\alpha$ -RuCl <sub>3</sub>	-1.7	-6.6	+6.6	0	-	+2.7	DFT + exact diagonalization	[32]	2016
3	$\alpha$ -RuCl <sub>3</sub>	-	-6.8	+9.5	-	-	-	Fit to neutron scattering	[33]	2017
3'	$\alpha$ -RuCl <sub>3</sub>	-	-5.5	+7.6	-	-	-	DFT + $t/U$ expansion	[34]	2016
3''	$\alpha$ -RuCl <sub>3</sub>	-1	-8	+4	-	-	-	DFT + $t/U$ expansion	[38]	2016
3 + $J_3$	$\alpha$ -RuCl <sub>3</sub>	-0.5	-5.0	+2.5	-	-	+0.5	Fit to neutron scattering	[39]	2017

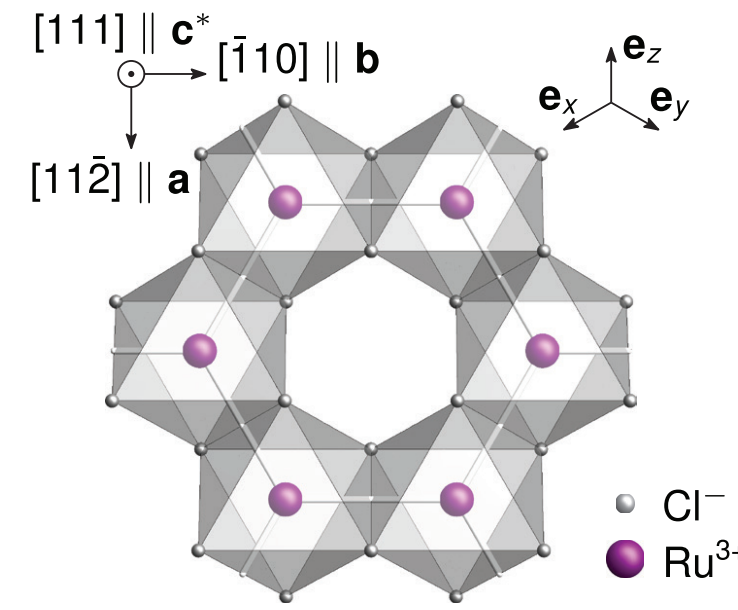
[LJ, Andrade, Vojta, PRB '17]

[Winter *et al*, JPCM '17]

[LJ, Vojta, JPCM '19]

[Maksimov, Chernyshev, PRR '20]

# $\alpha$ -RuCl<sub>3</sub>: Effective spin models



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[Jackeli, Khaliullin, PRL '09]

Exchange couplings:

Dominant  $K_1 < 0$  of  $O(5\text{meV})$  and large  $\Gamma_1 > 0$

- ✓ Ab-initio estimates
- ✓ Zigzag ground state
- ✓ Magnetic anisotropy
- ✓ In-field phase diagram

$(z, \tau)$	Material	$J_1$	$J_2$	$J_3$	$K_1$	$K_2$	$\Gamma_1$	Method	Ref.	Year
2/3	$\alpha$ -RuCl <sub>3</sub>	-1.7	-6.6	+6.6	0	-	+2.7	DFT + exact diagonalization	[32]	2016
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3'	$\alpha$ -RuCl <sub>3</sub>	-	-5.5	+7.6	-	-	-	DFT + $t/U$ expansion	[34]	2016
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[LJ, Andrade, Vojta, PRB '17]

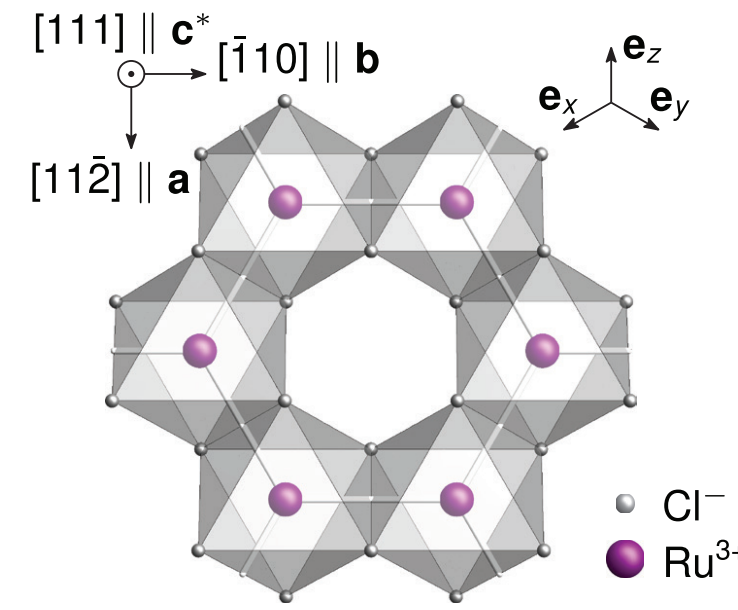
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[Jackeli, Khaliullin, PRL '09]

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- ✓ Zigzag ground state
- ✓ Magnetic anisotropy
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Today: 2D modeling **incomplete!**

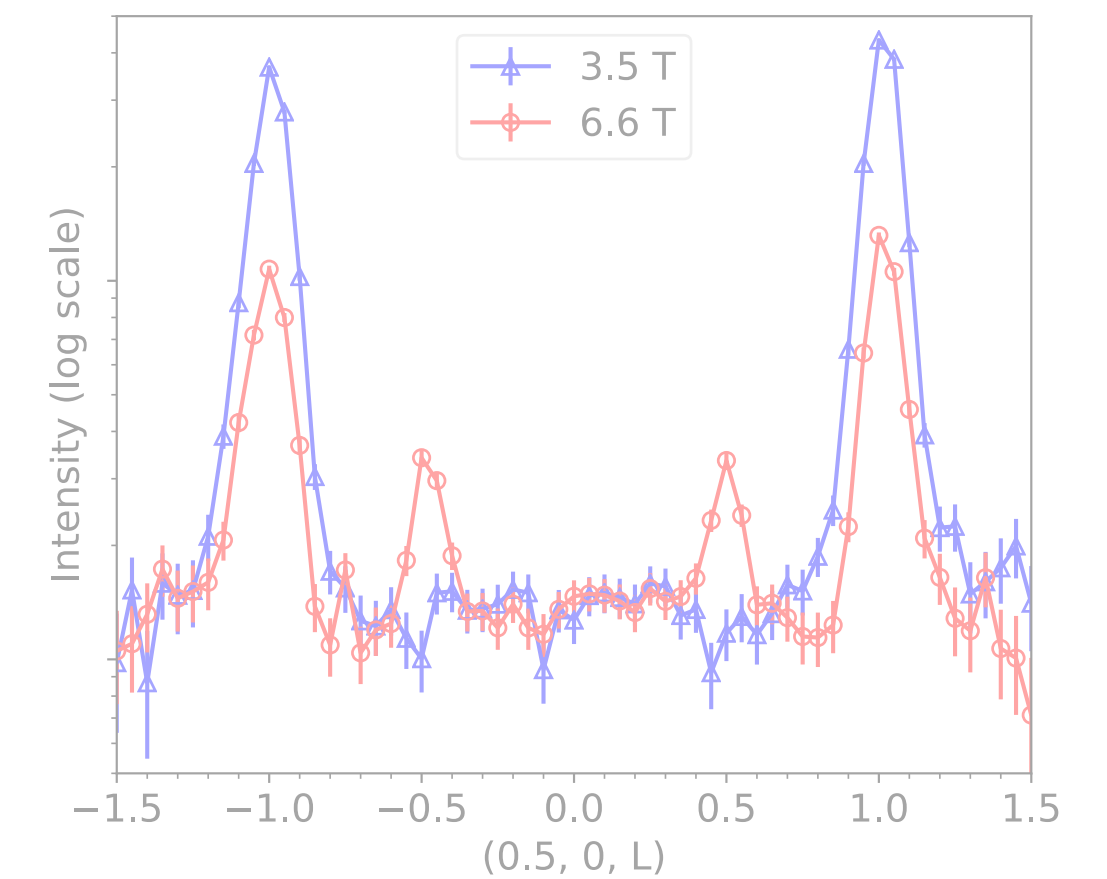
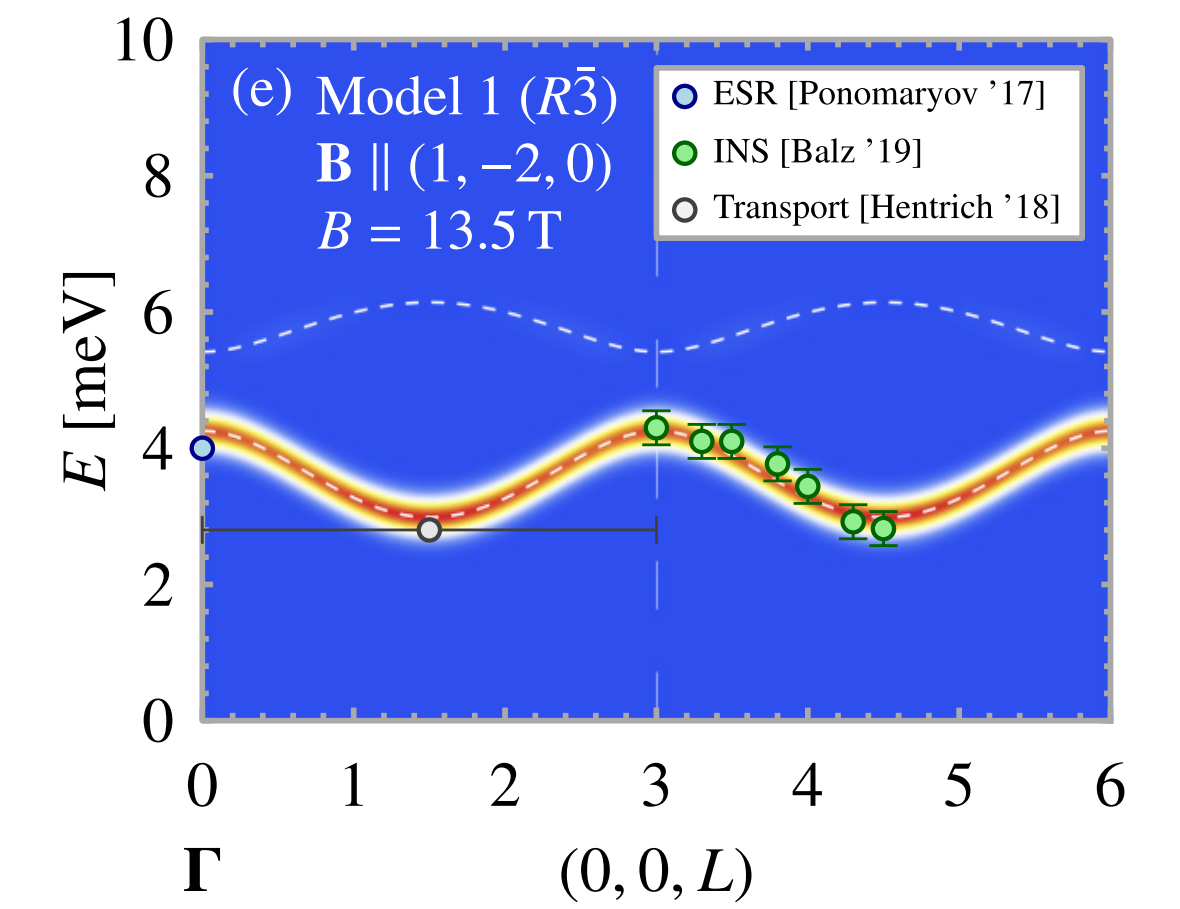
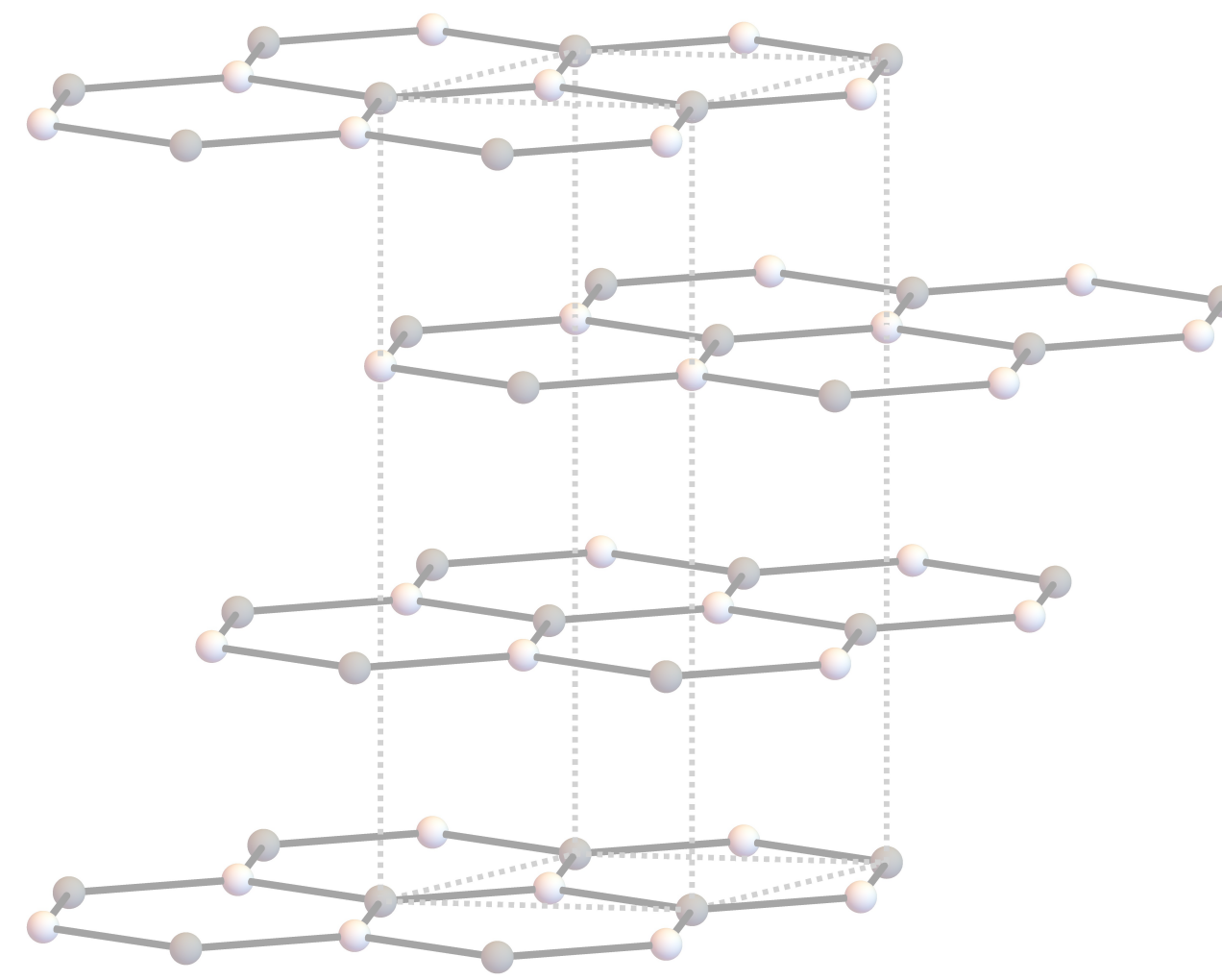
# Outline

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2. Evidence #1: Excitation spectra

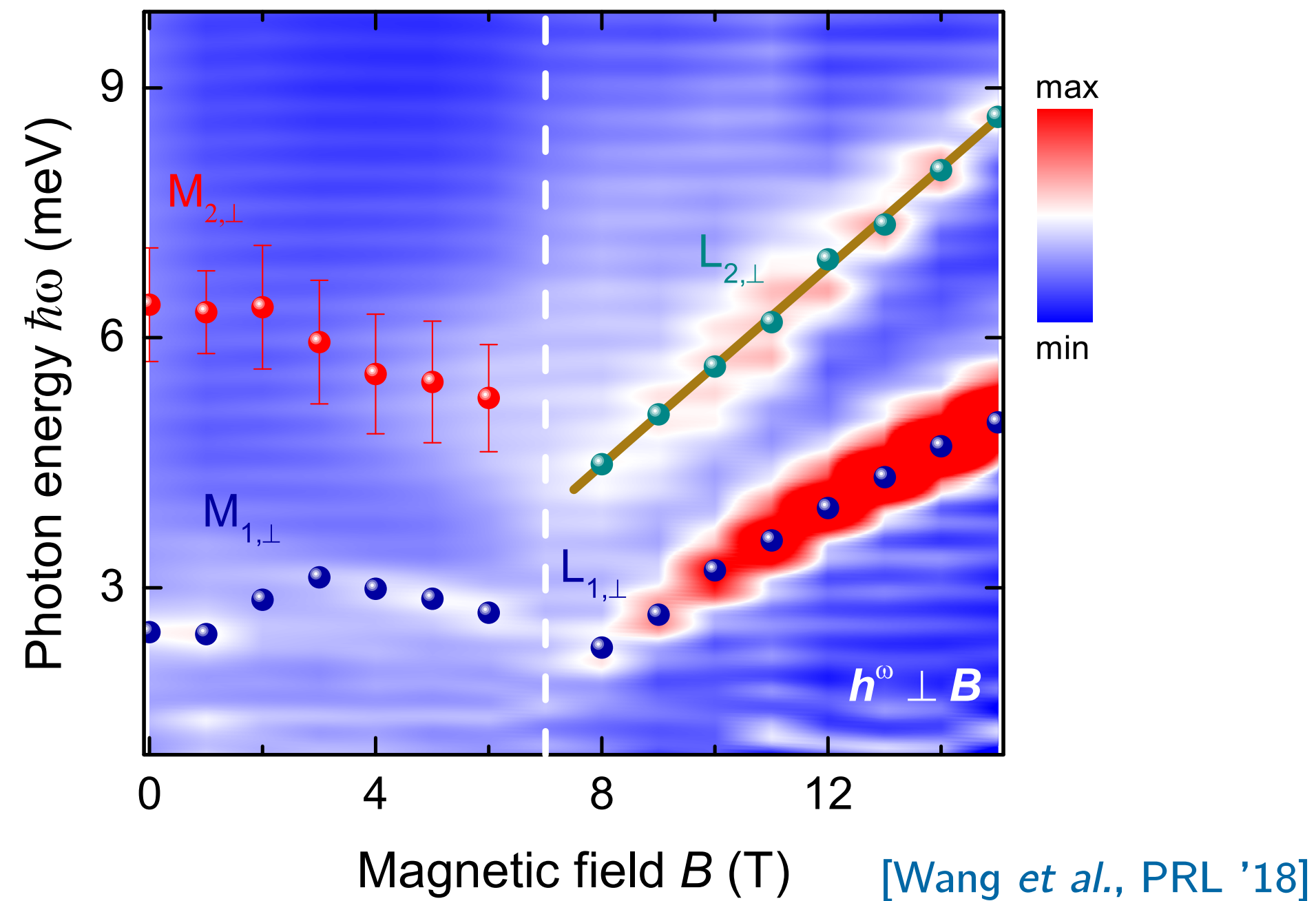
3. Evidence #2: Field-induced intermediate order

4. Conclusions



# Excitation spectrum in high-field phase I

THz spectroscopy

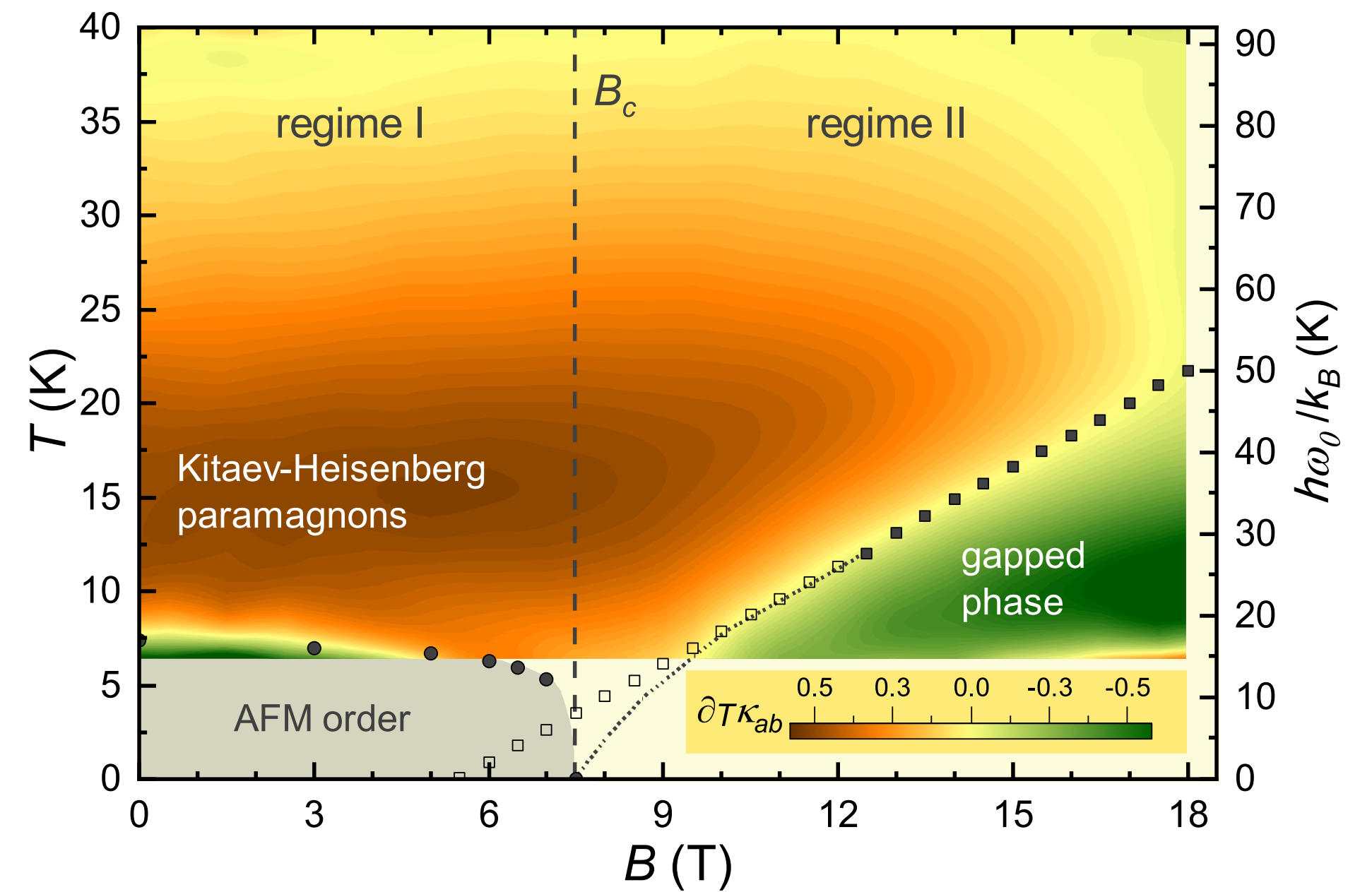


... consistent with ESR [Ponomaryov *et al.*, PRB '17, PRL '20]  
[Wellm *et al.*, PRB '18]

Gap @ 13.5T:  $\Delta(\vec{Q} = \Gamma) \simeq 4.5 \text{ meV}$

vs.

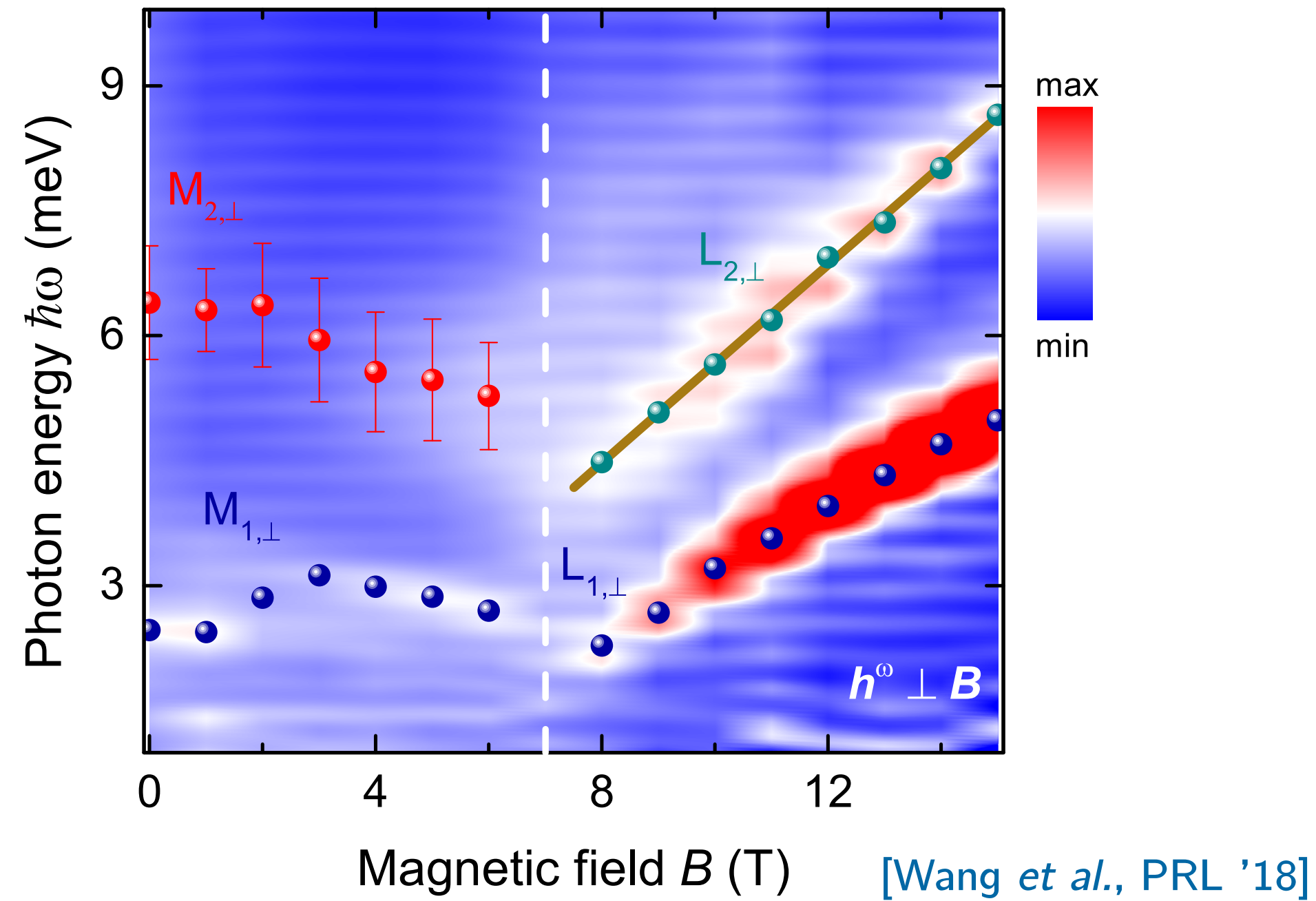
Heat transport



$\min_{\vec{Q}} \Delta(\vec{Q}) \simeq 32 \text{ K} \simeq 2.8 \text{ meV}$

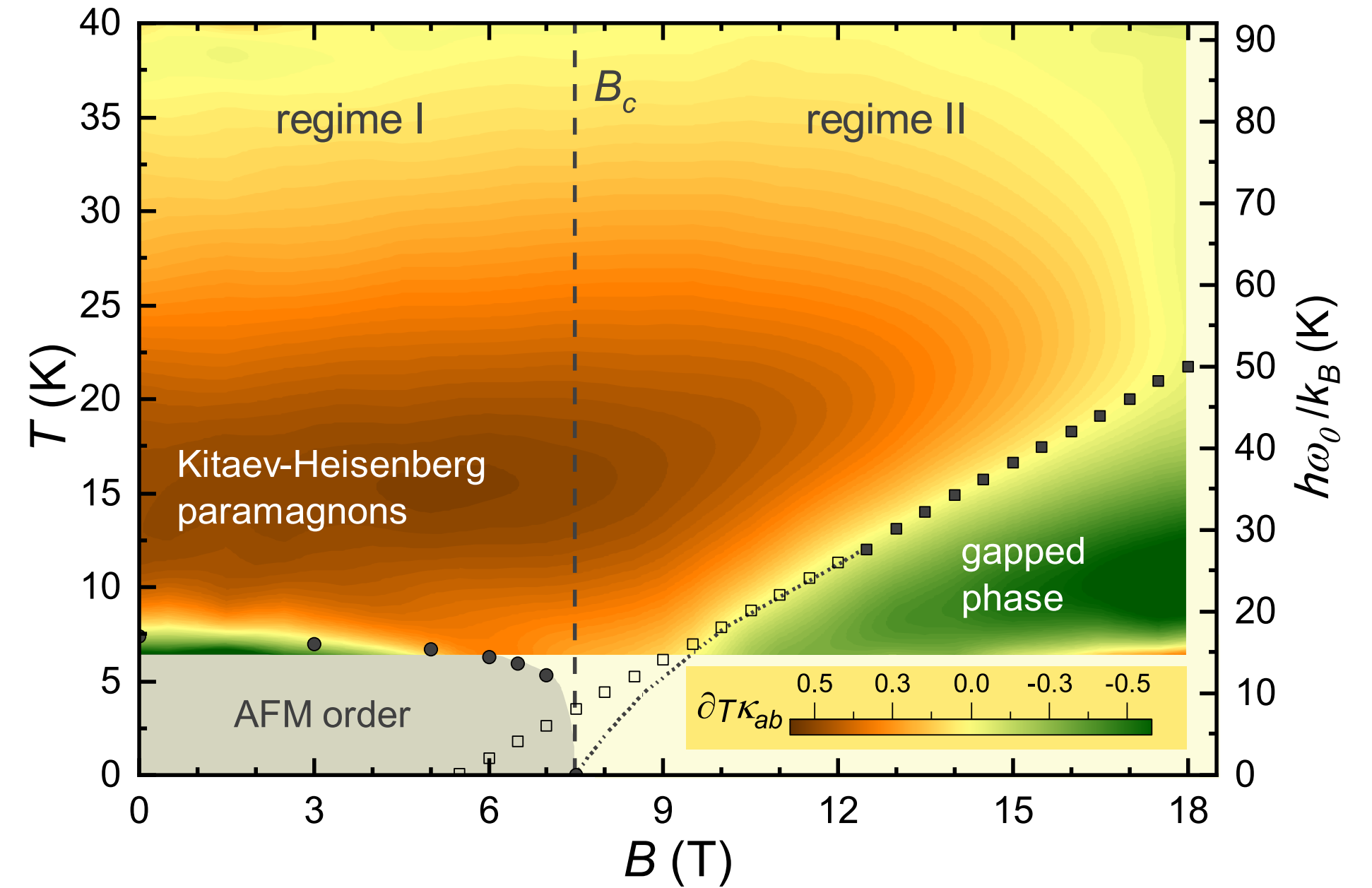
# Excitation spectrum in high-field phase I

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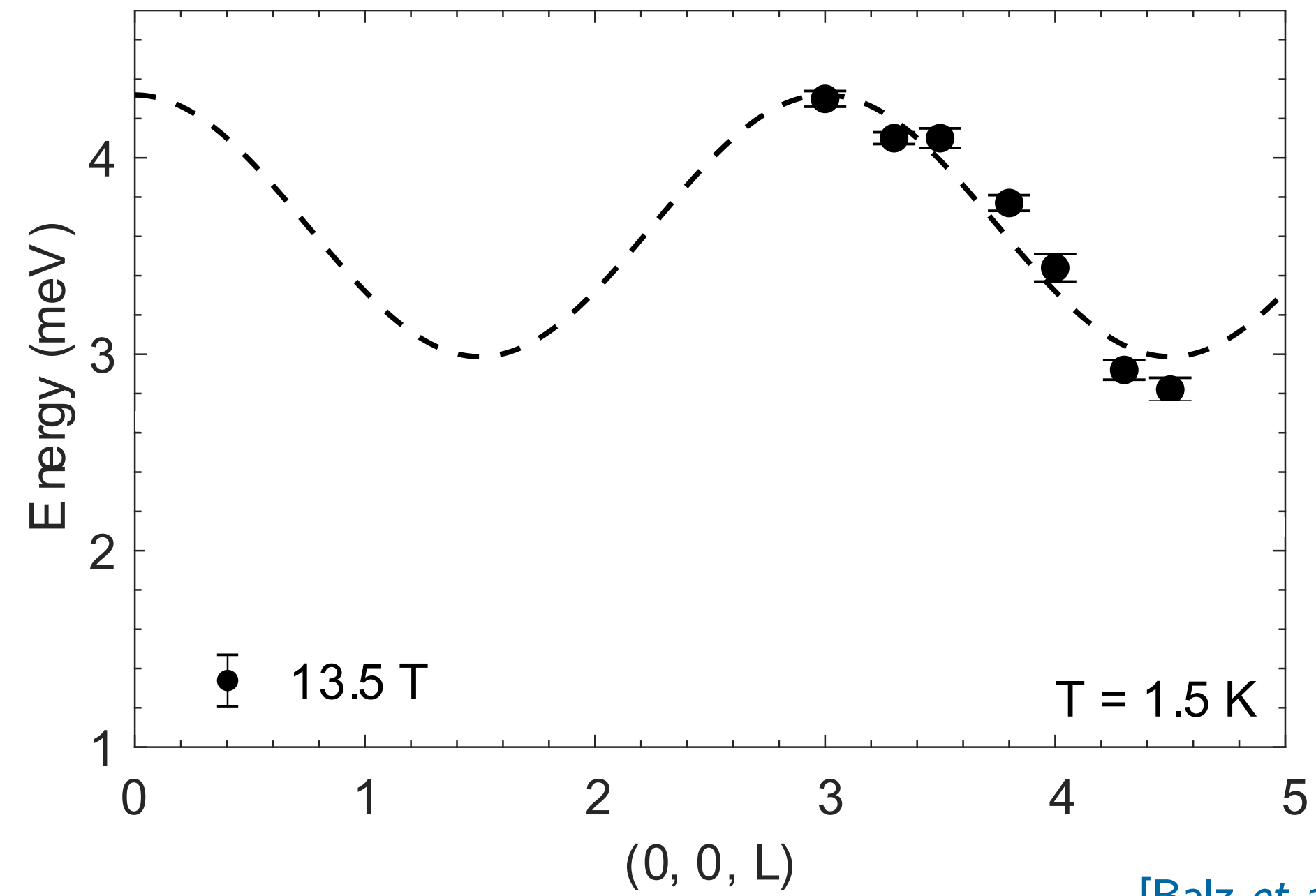
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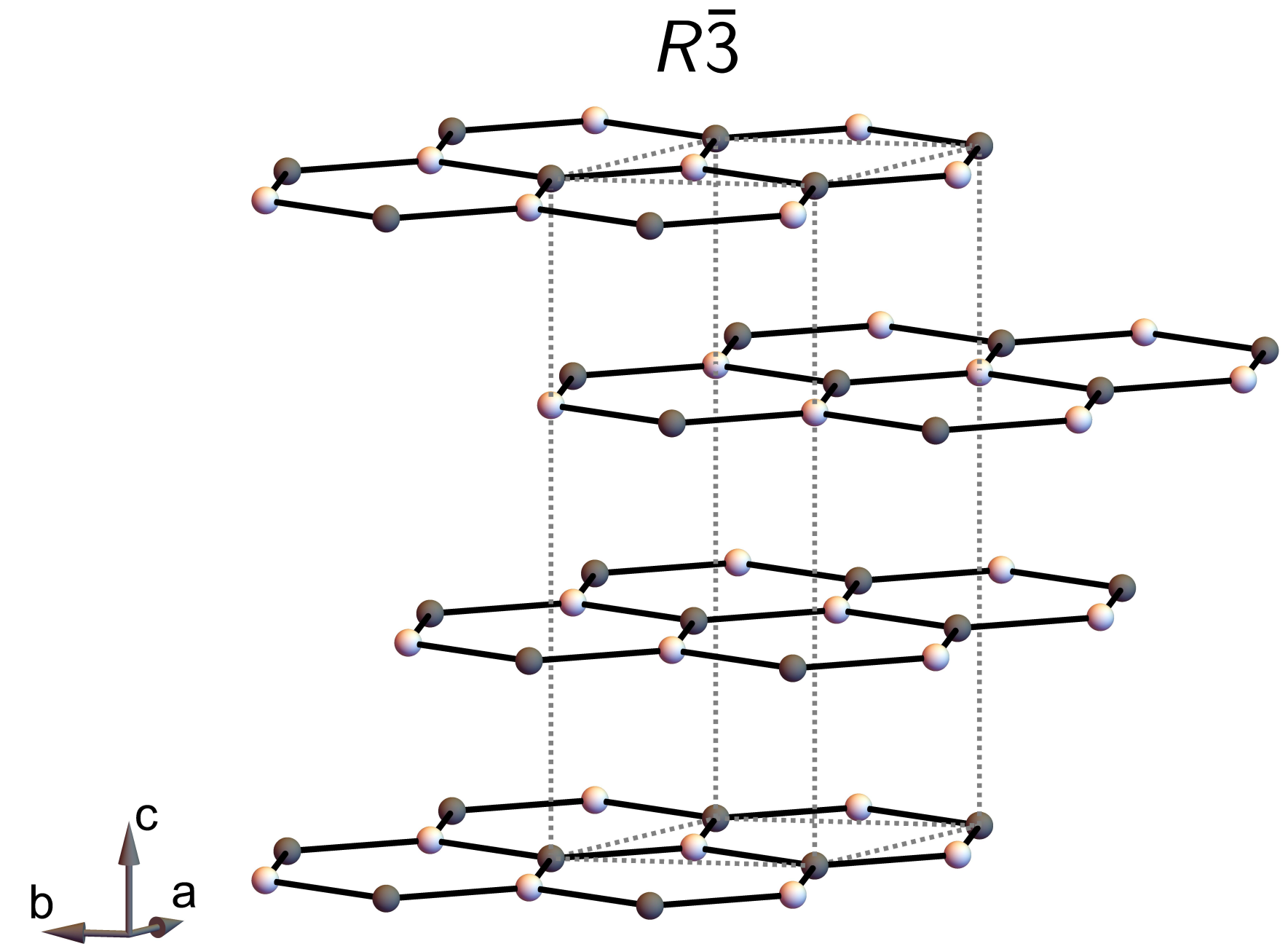
Lowest gap at  $\vec{Q} \neq \Gamma$ ?

# Excitation spectrum in high-field phase II

Inelastic neutron scattering

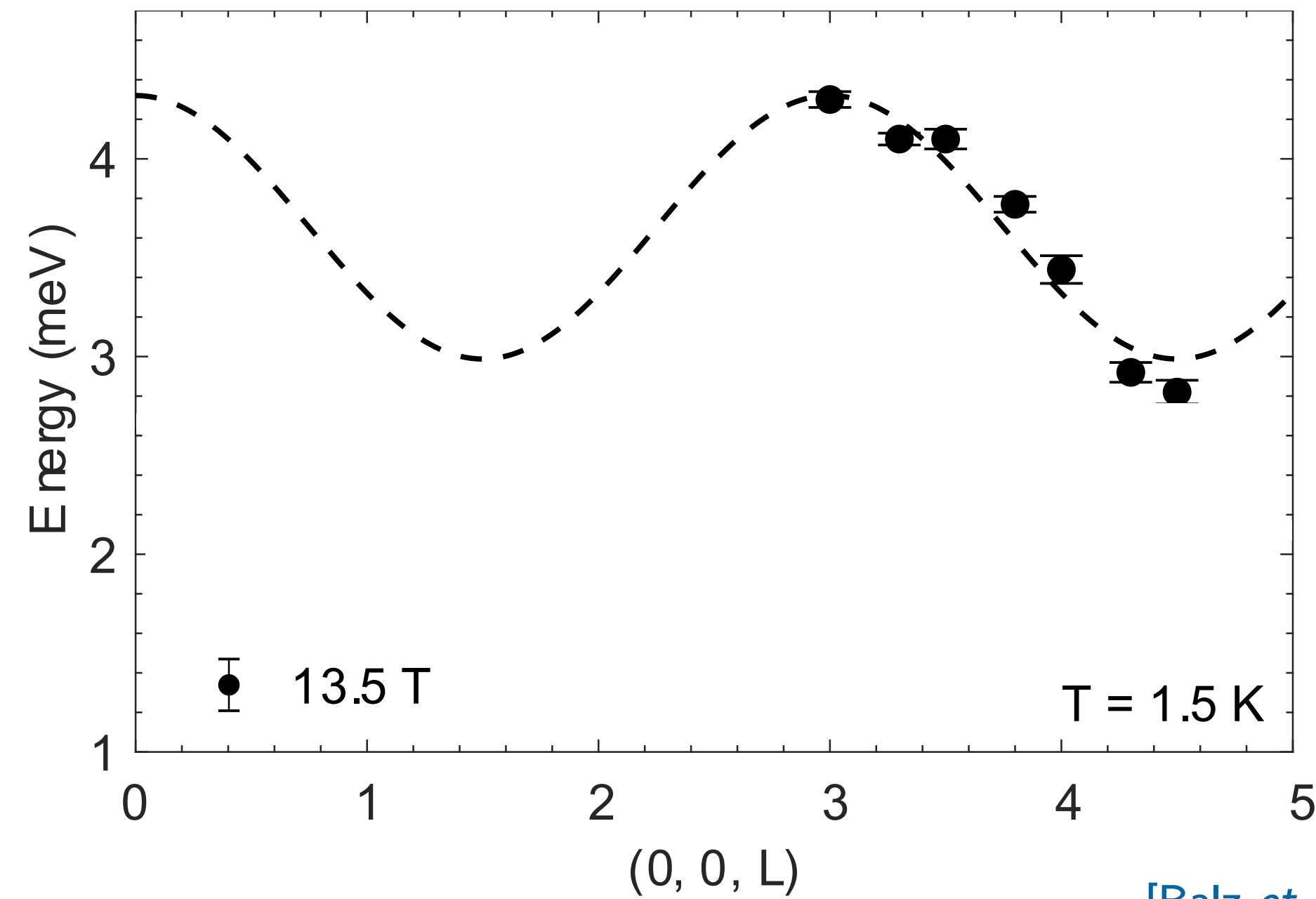


[Balz *et al.*, PRB '20]

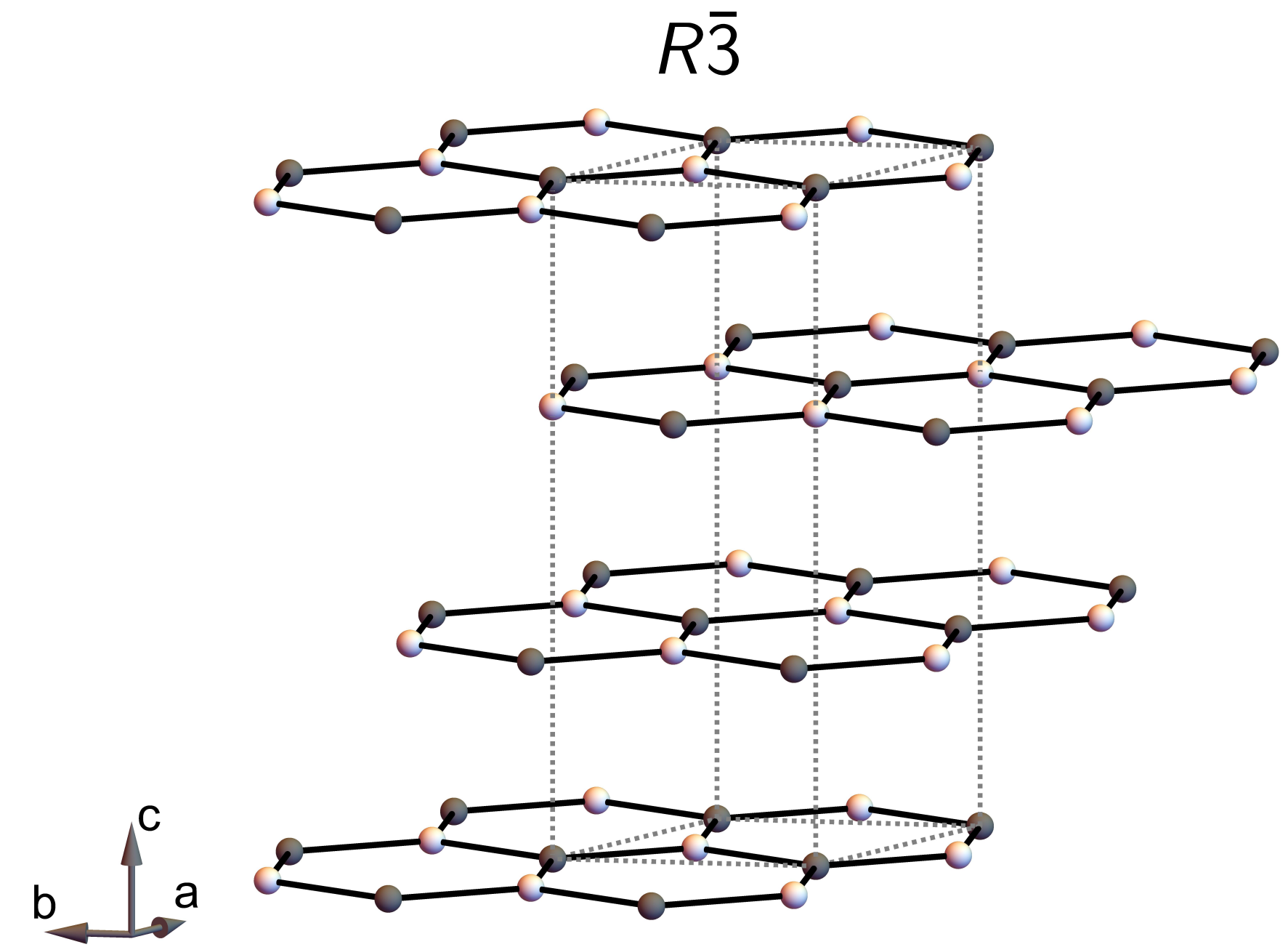


# Excitation spectrum in high-field phase II

Inelastic neutron scattering



[Balz *et al.*, PRB '20]

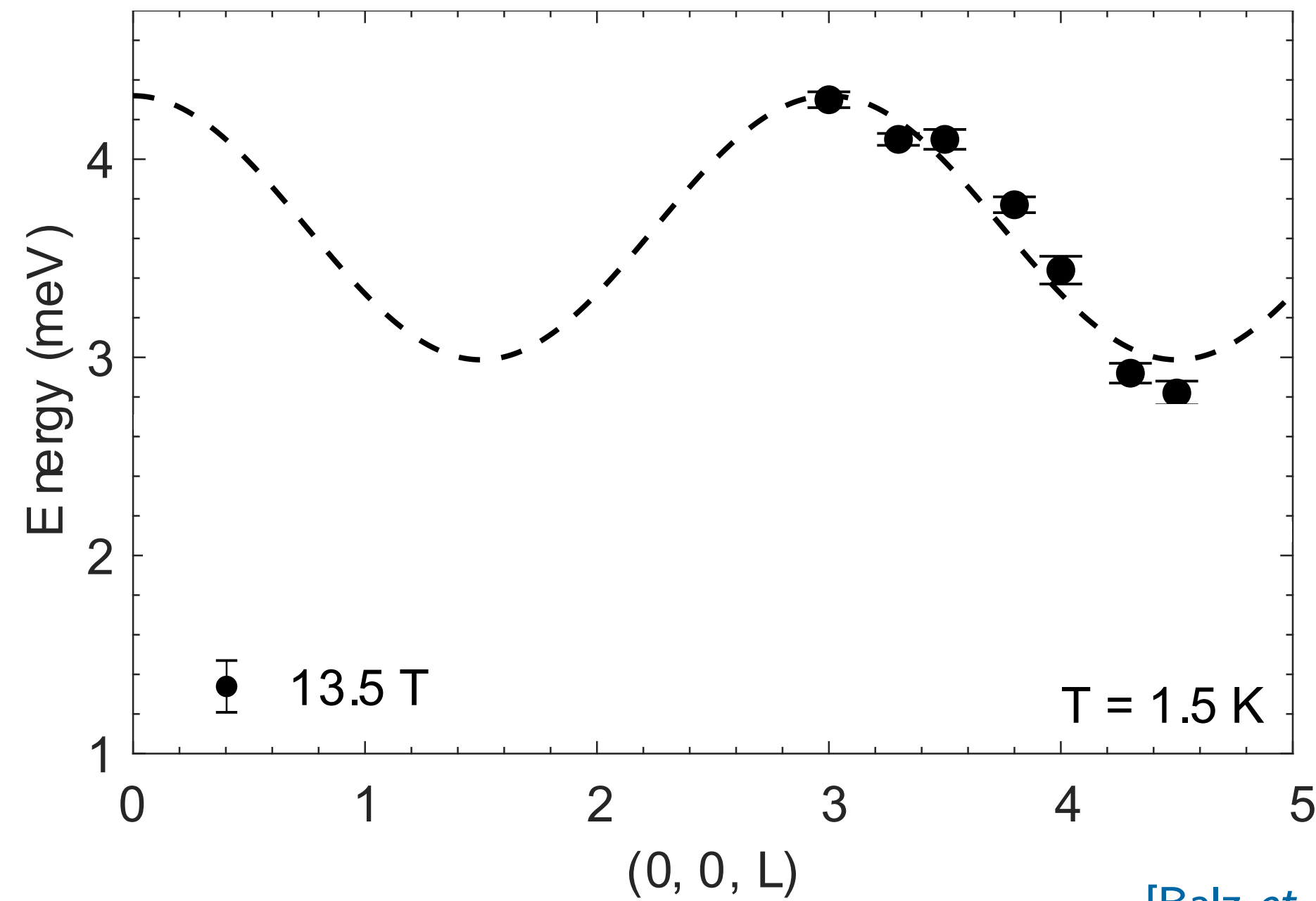


Out-of-plane bandwidth @ 13.5 T  $\simeq$  1.3 meV

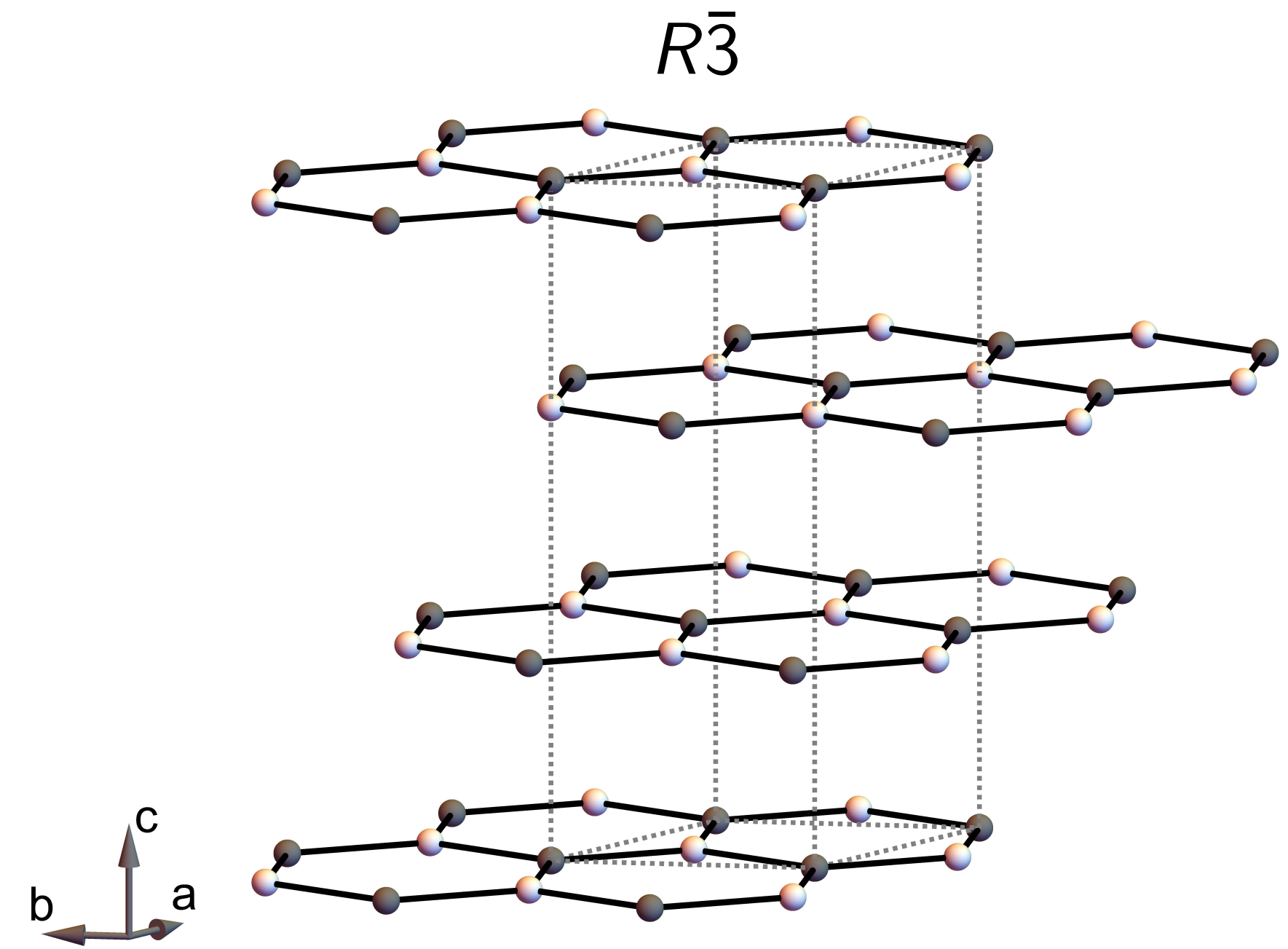
... not significantly smaller than in-plane bandwidth

# Excitation spectrum in high-field phase II

Inelastic neutron scattering



[Balz *et al.*, PRB '20]



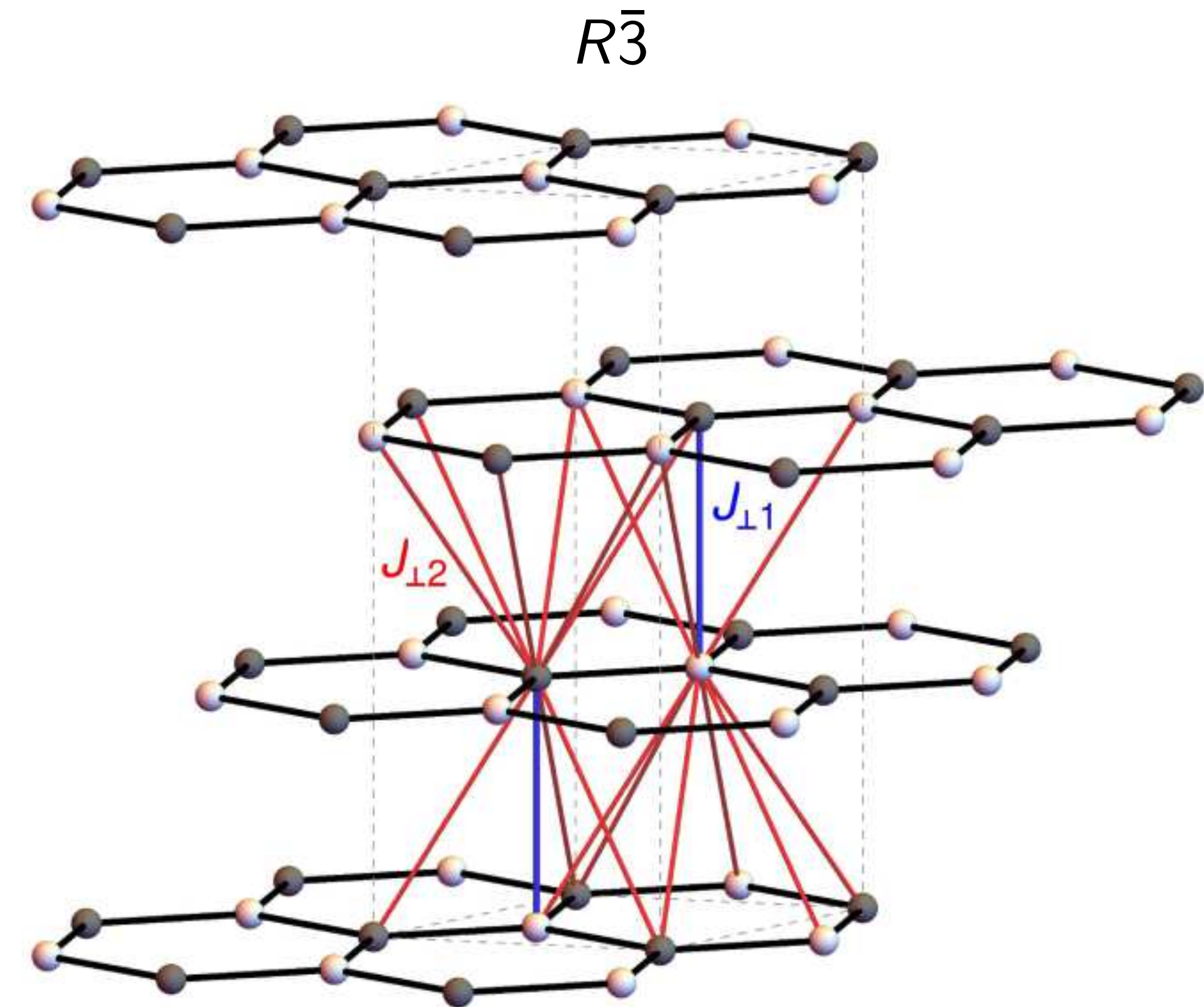
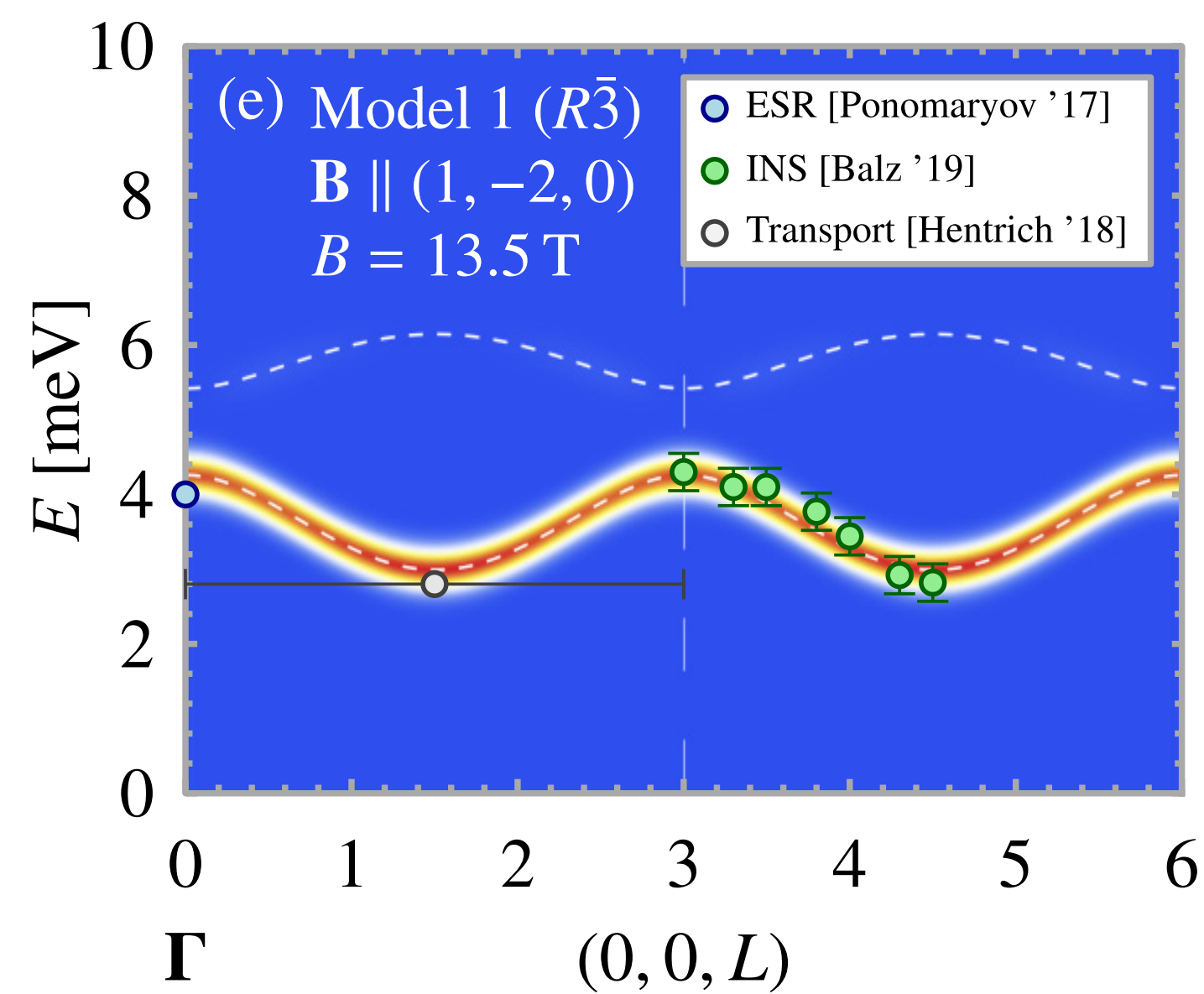
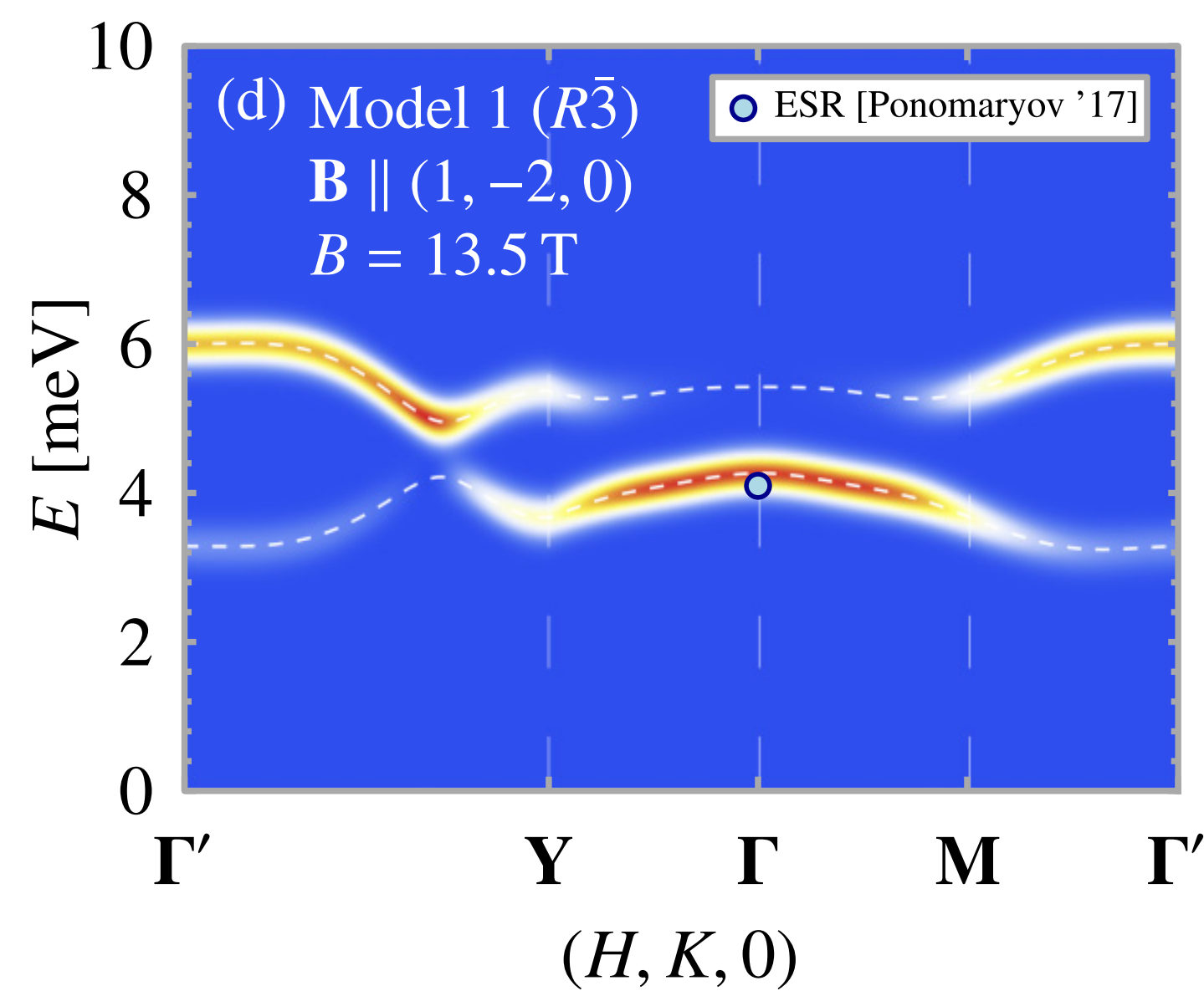
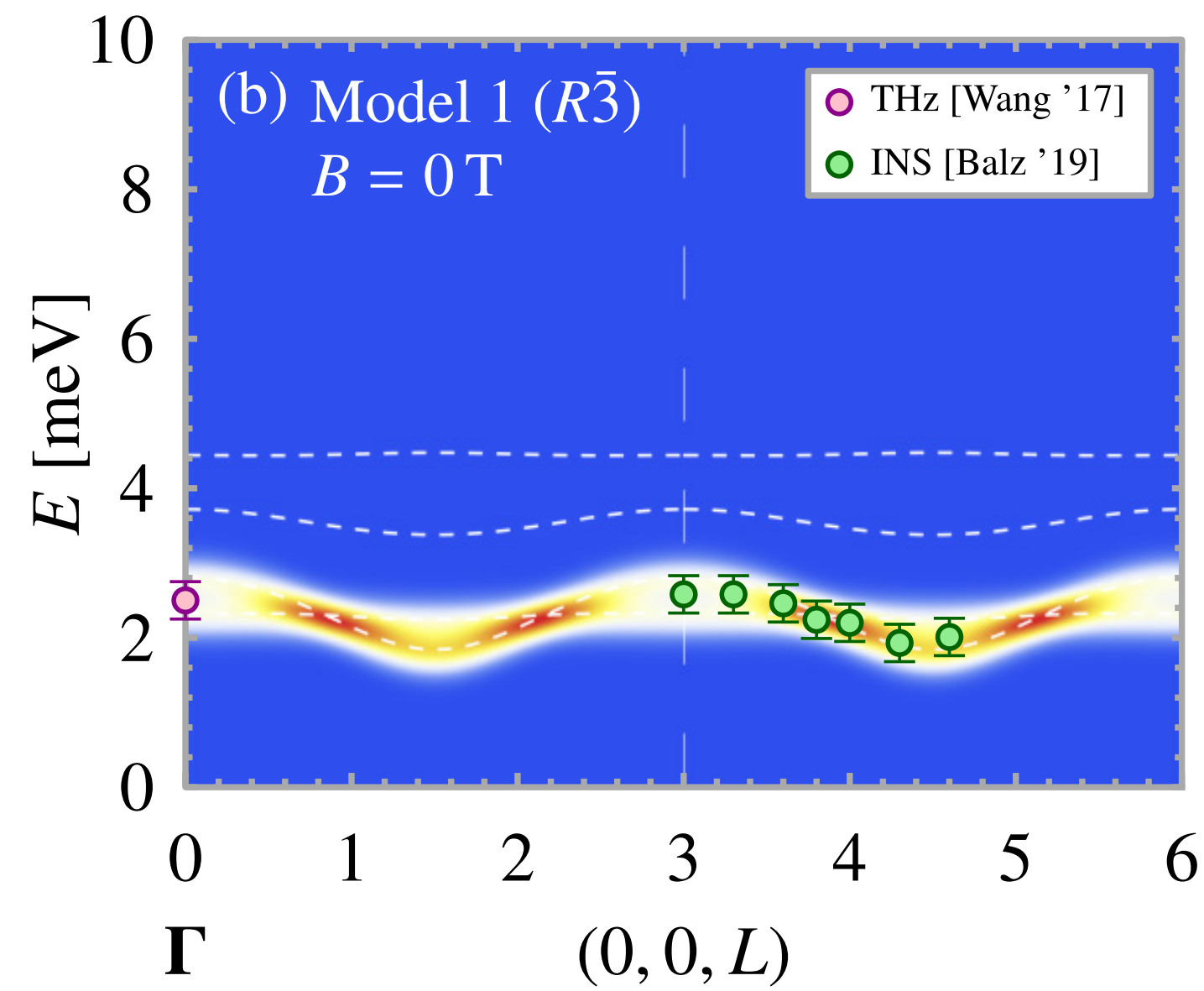
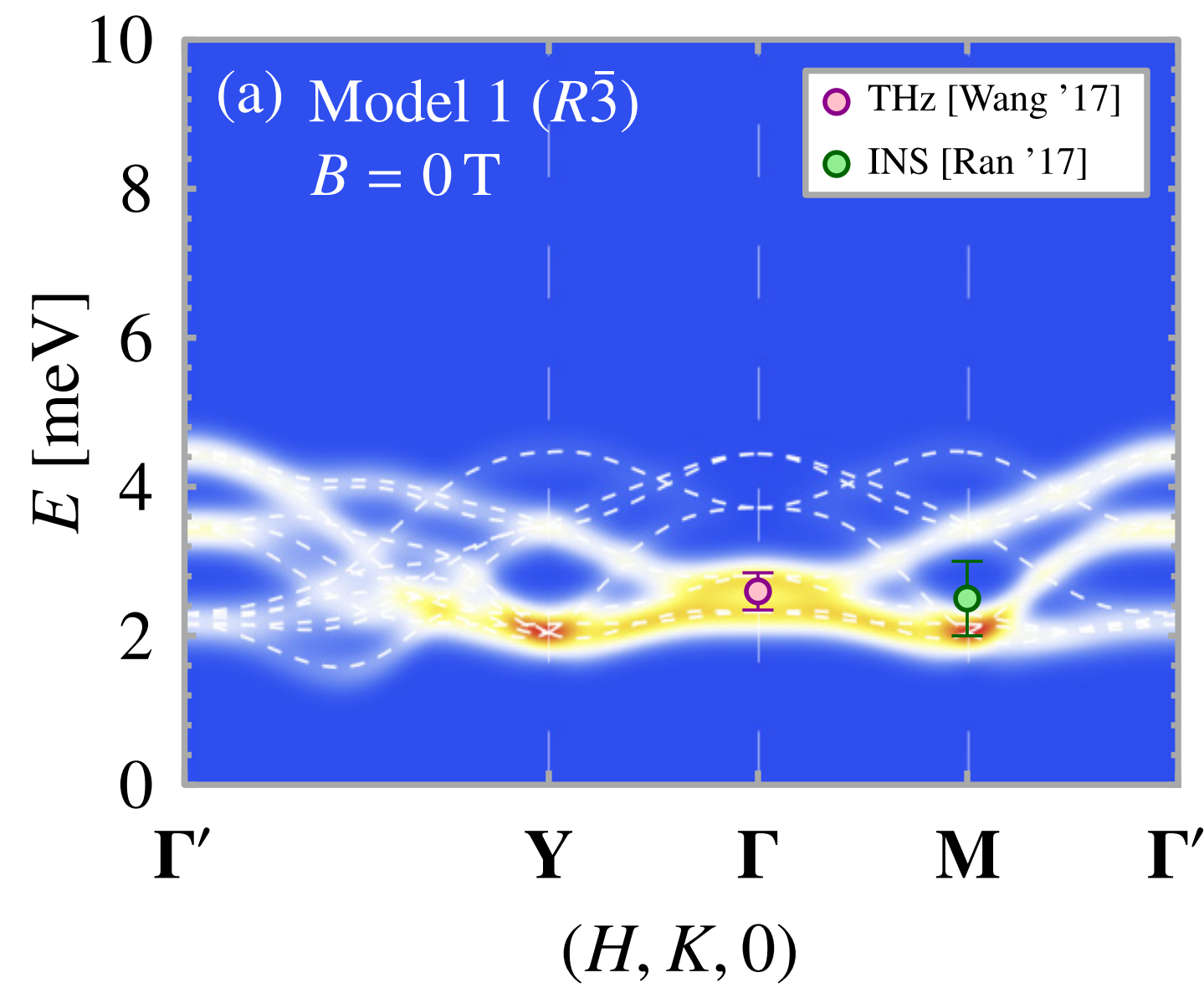
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Out-of-plane interactions important in  $\alpha$ -RuCl<sub>3</sub>!

# 3D model for $\alpha$ -RuCl<sub>3</sub>

Dynamical structure factor (LSWT)



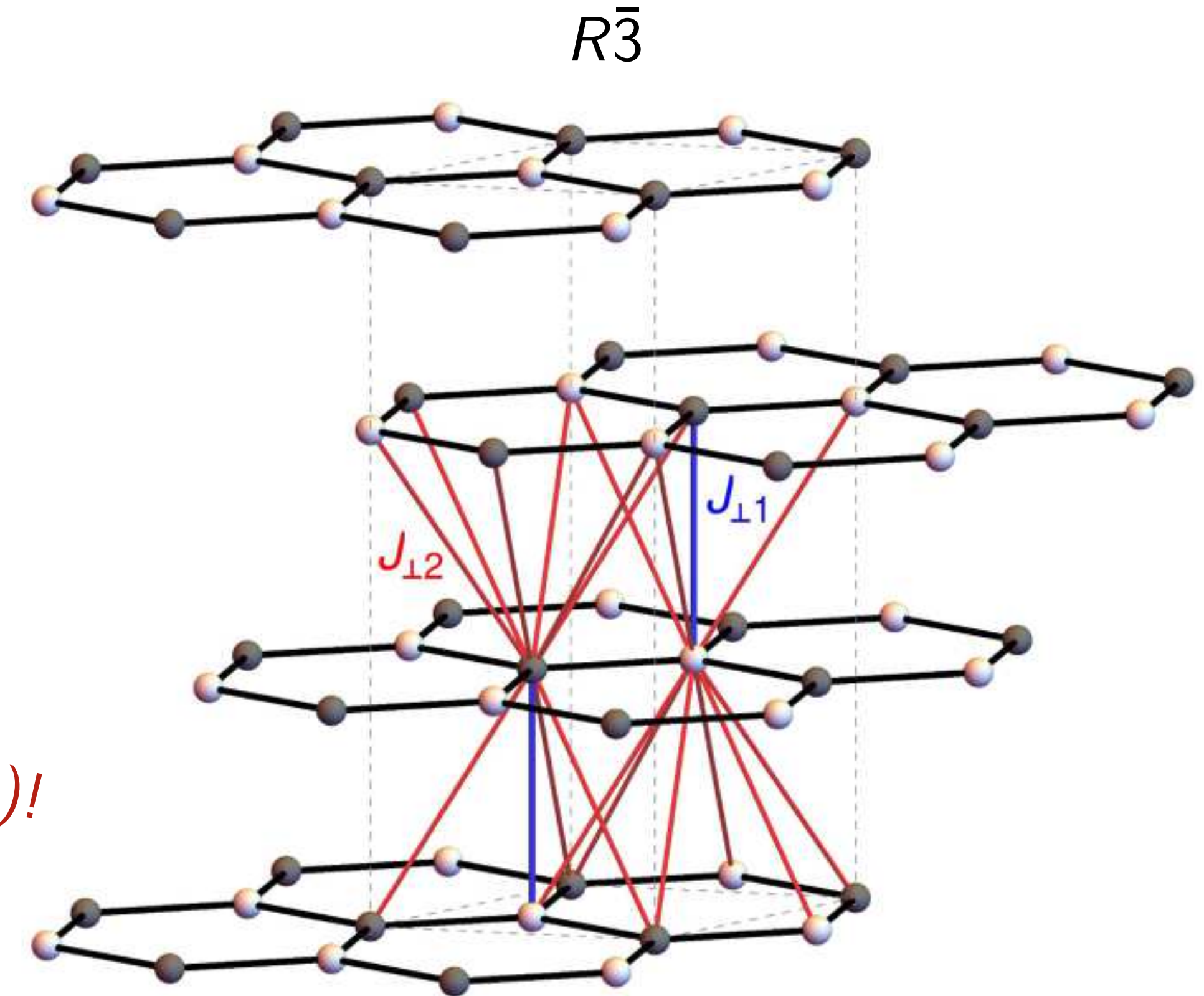
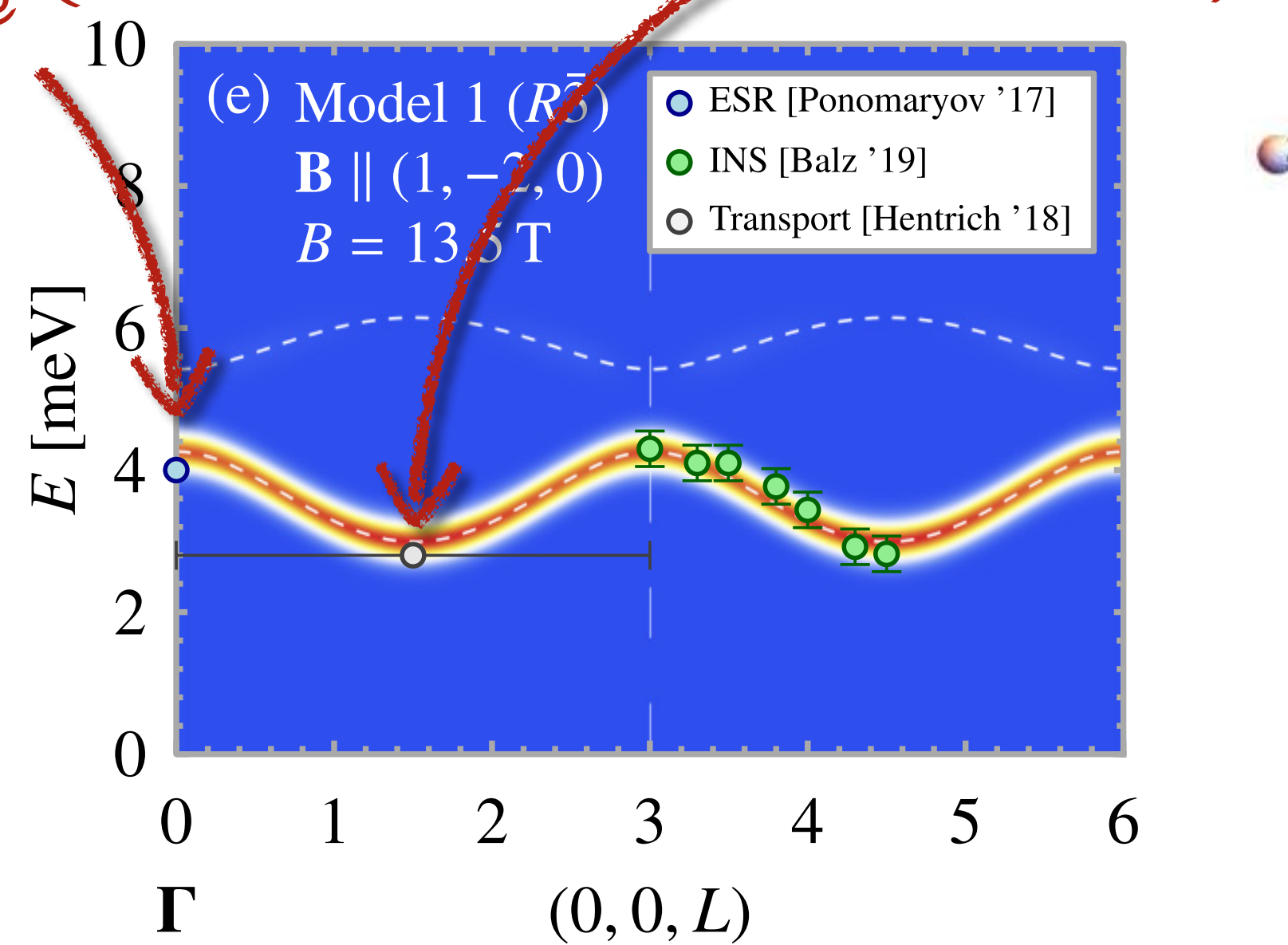
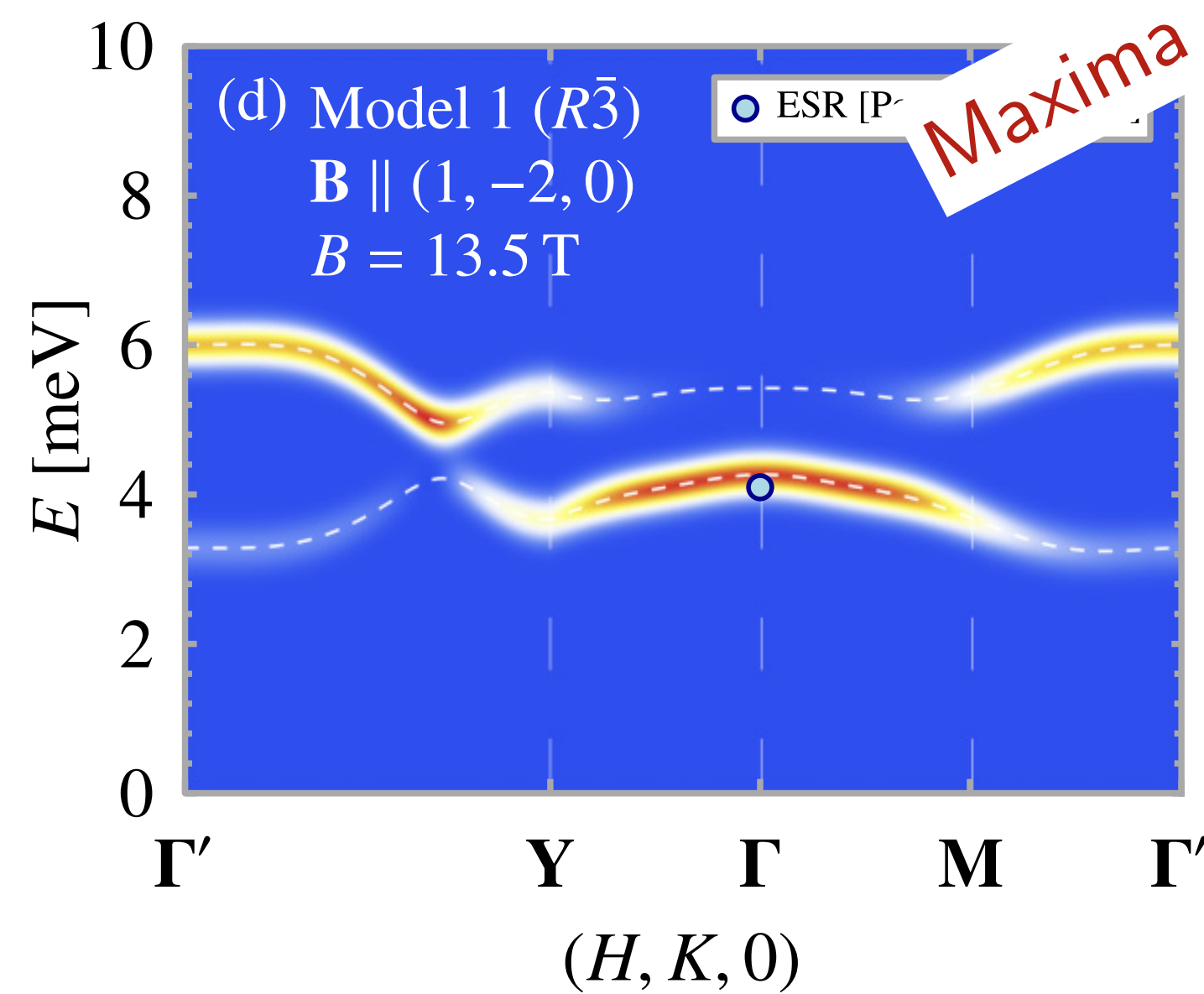
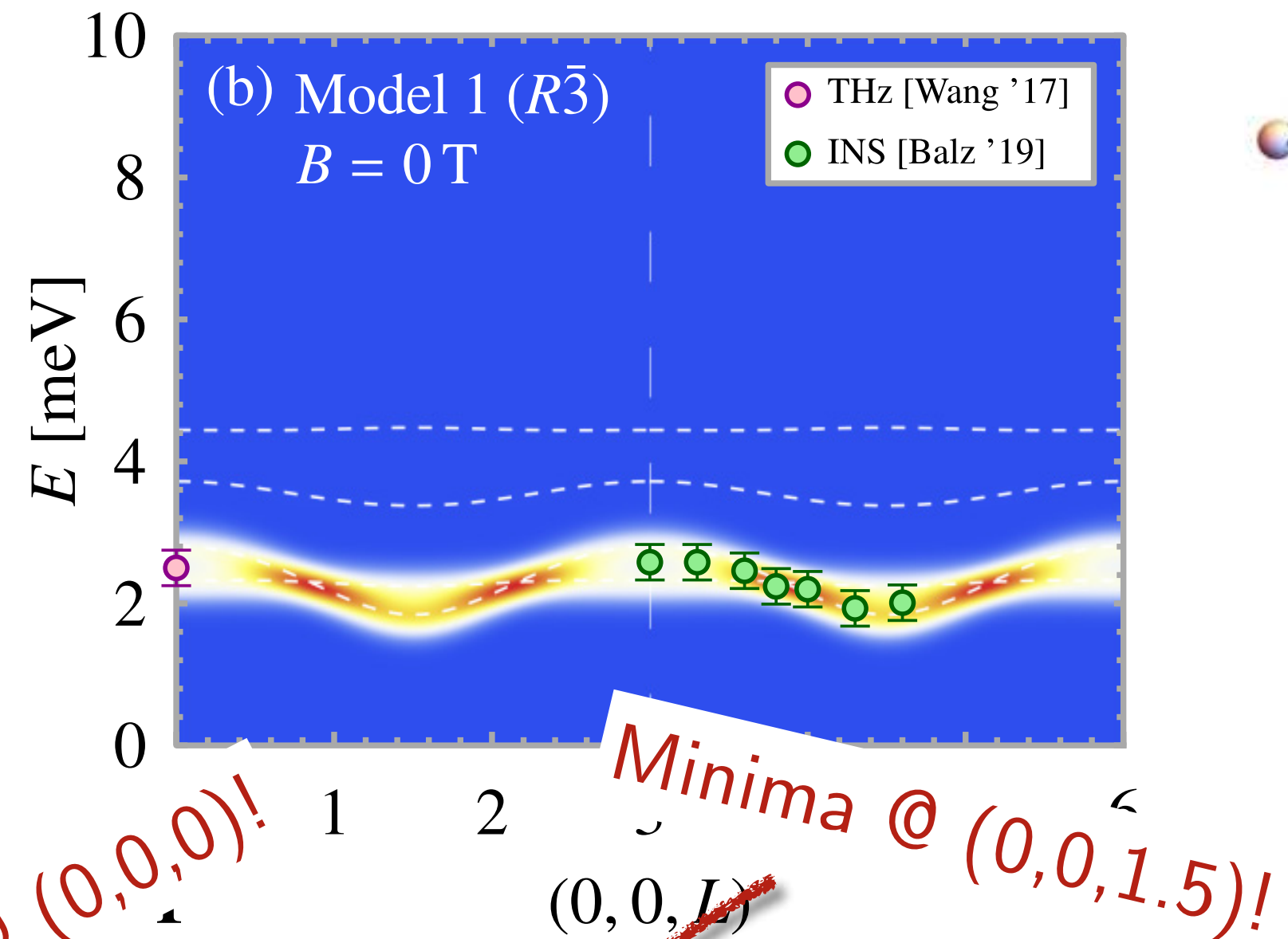
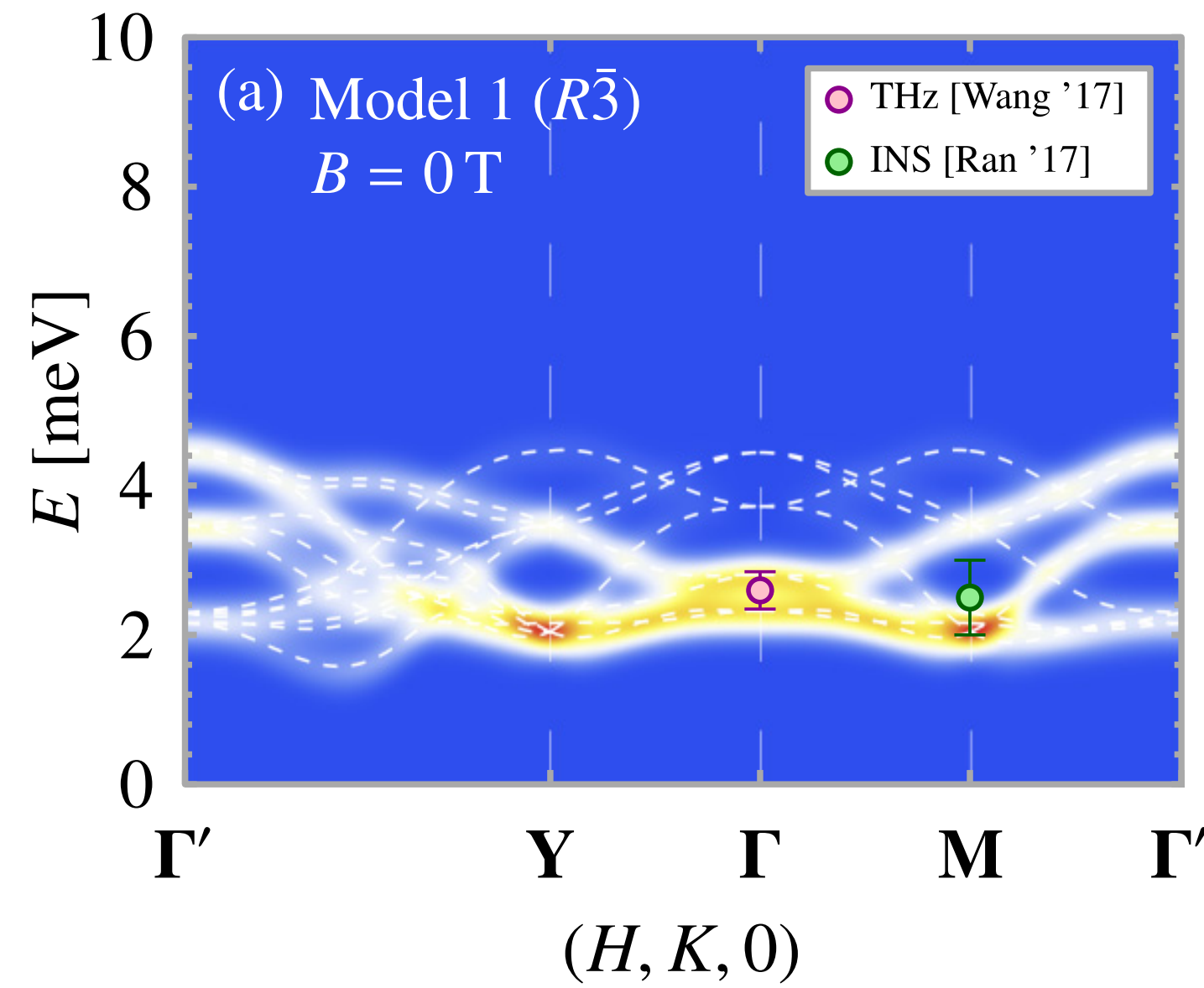
$$J_{\perp 1} \simeq +0.80 \text{ meV}$$

$$J_{\perp 2} \simeq +0.04 \text{ meV}$$



# 3D model for $\alpha$ -RuCl<sub>3</sub>

## Dynamical structure factor (LSWT)

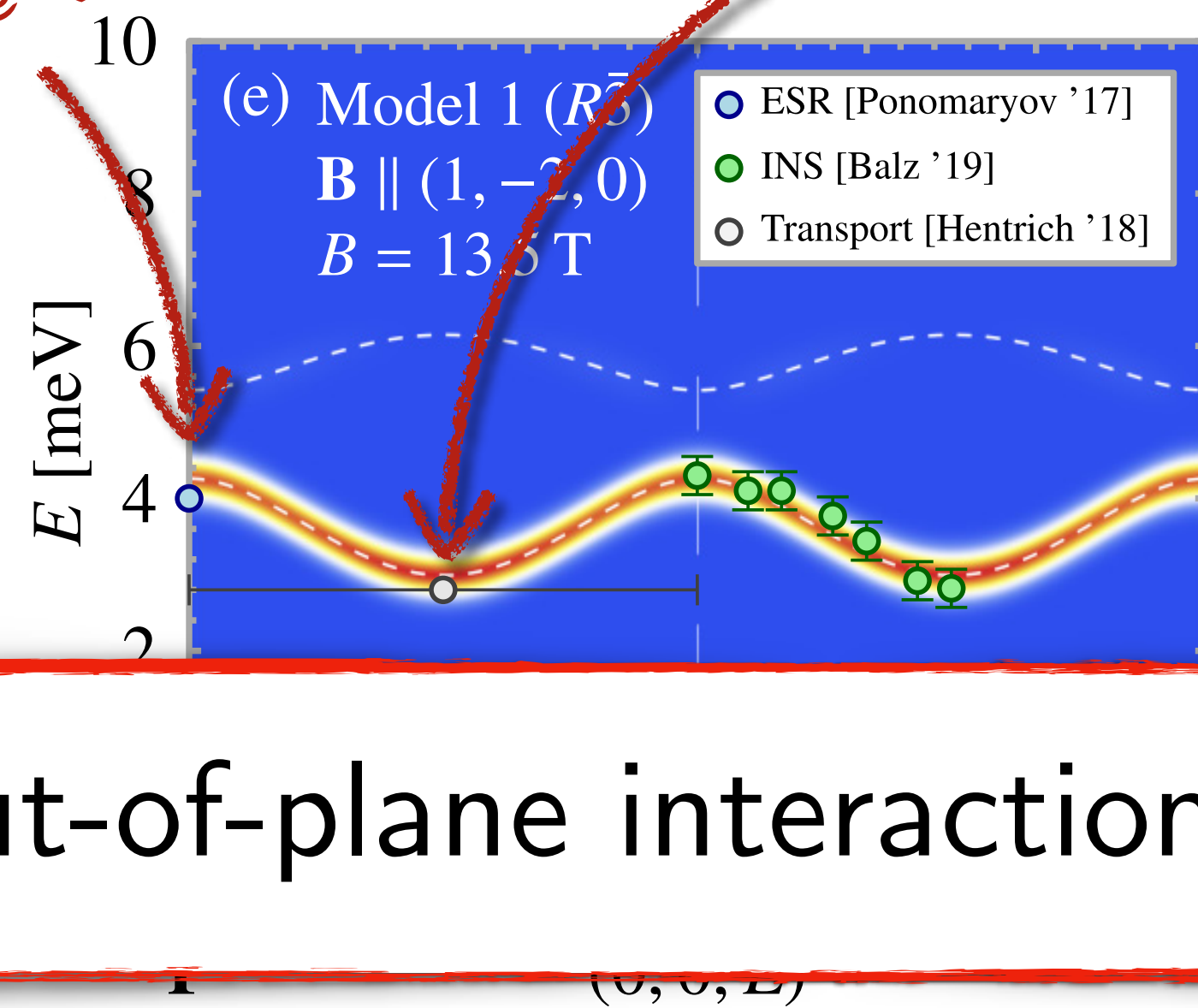
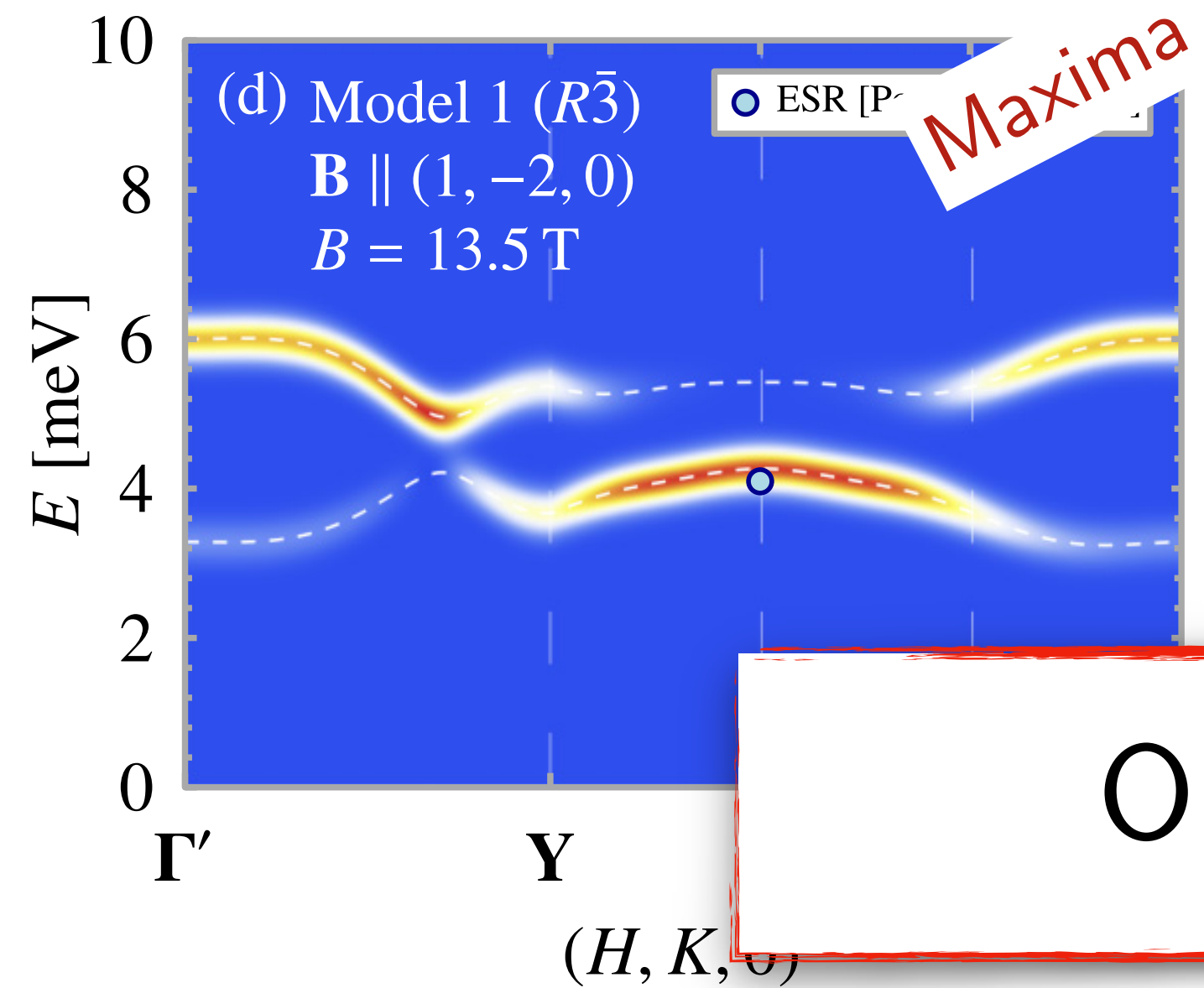
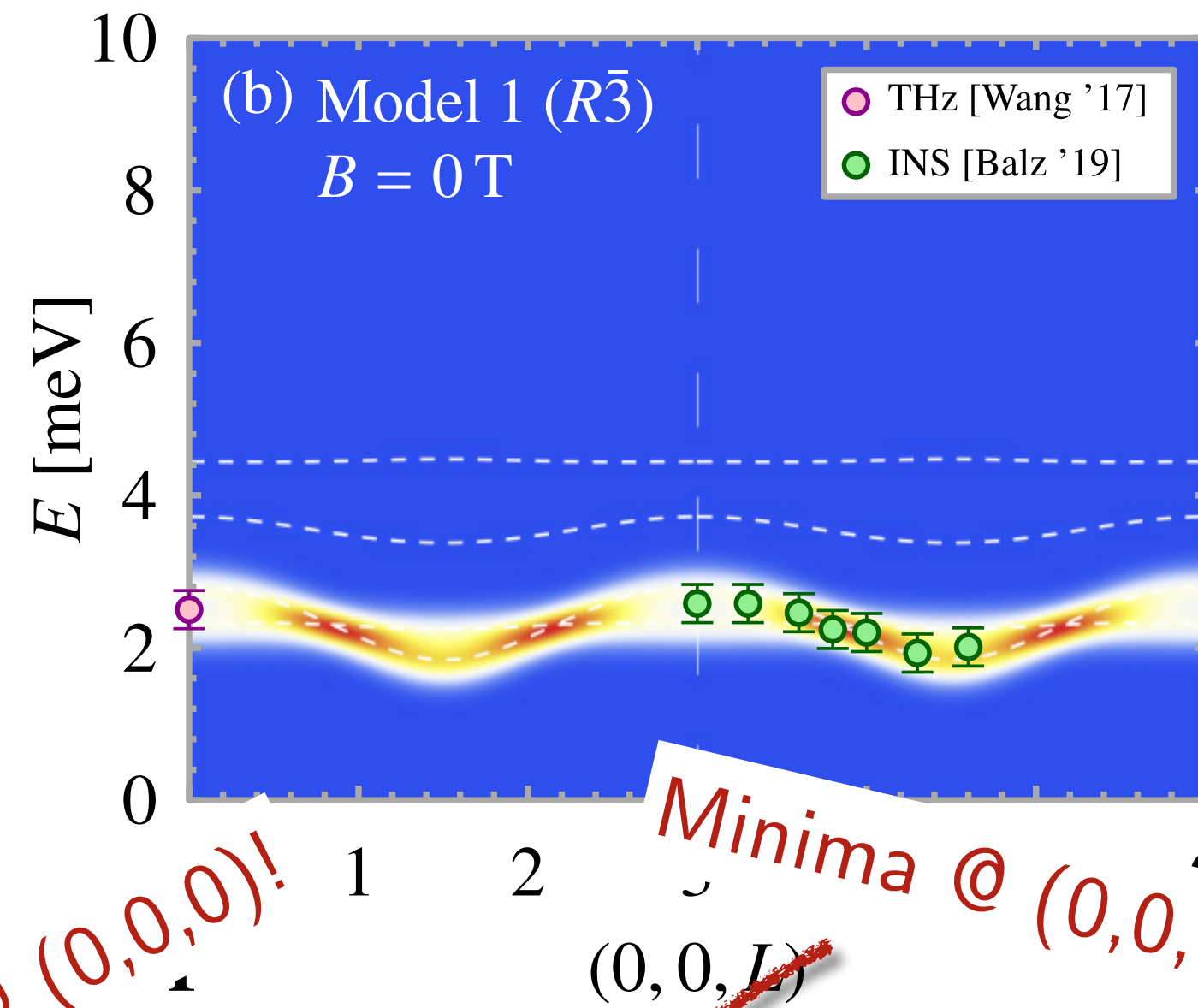
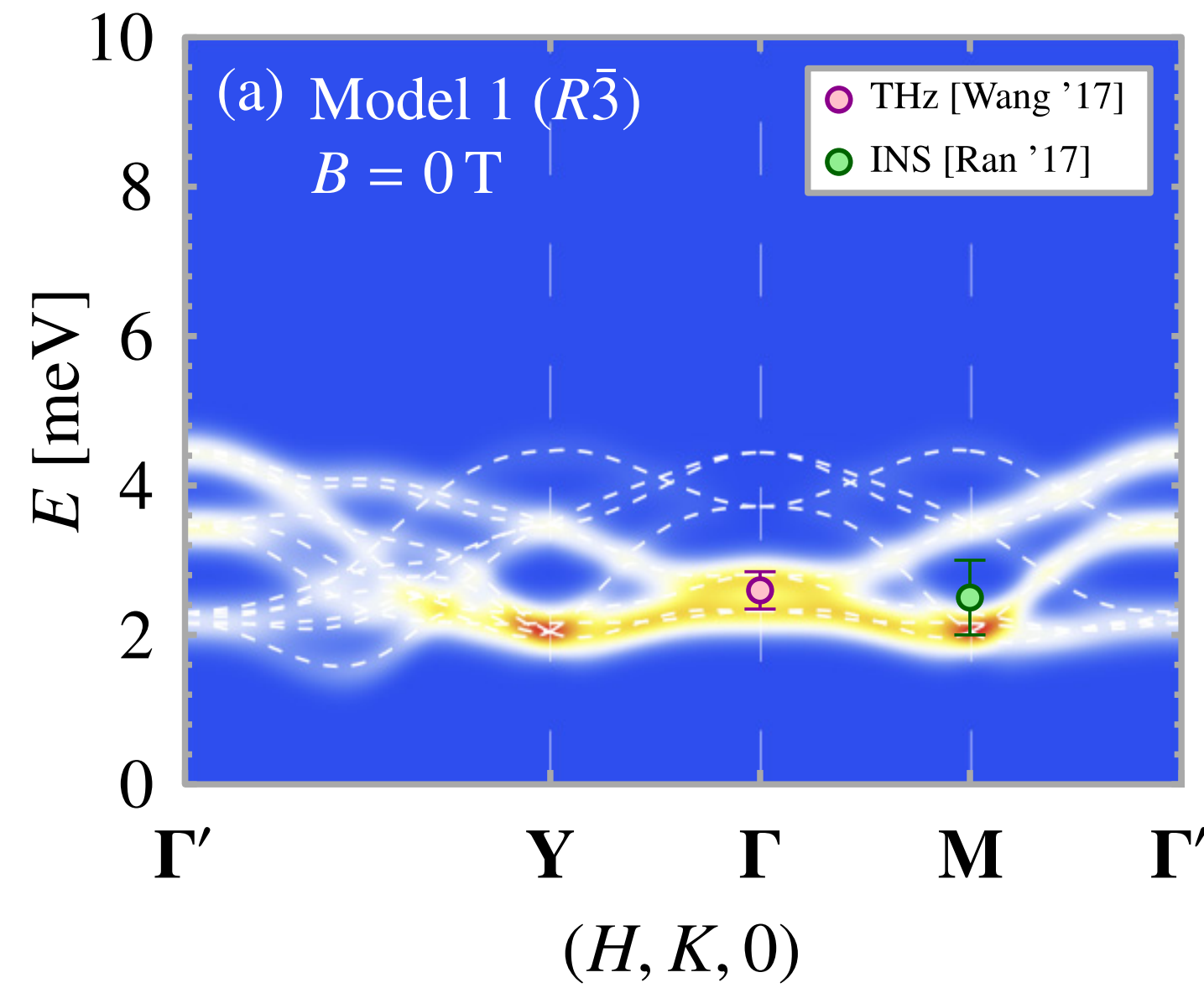


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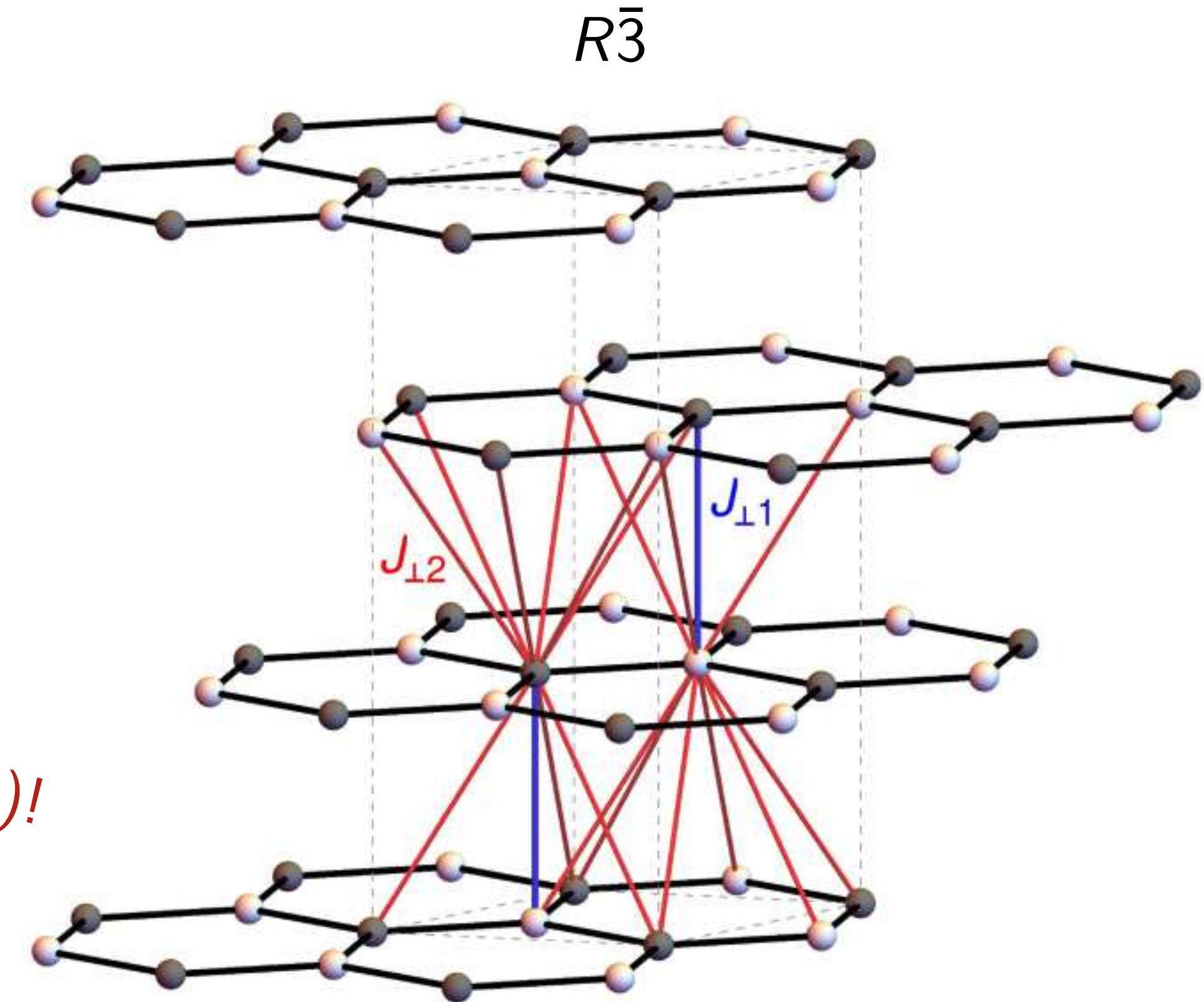
# 3D model for $\alpha$ -RuCl<sub>3</sub>

Dynamical structure factor (LSWT)



Maxima @ (0,0,0)!

Minima @ (0,0,1.5)!



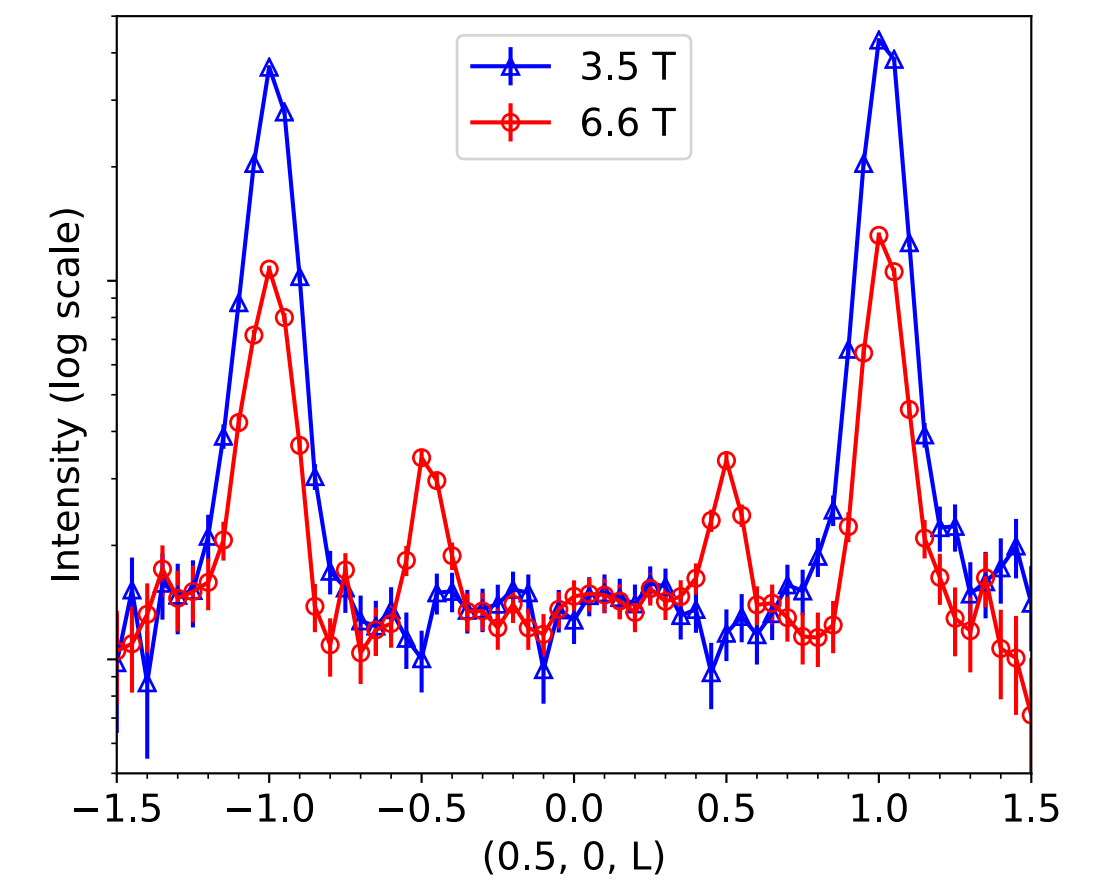
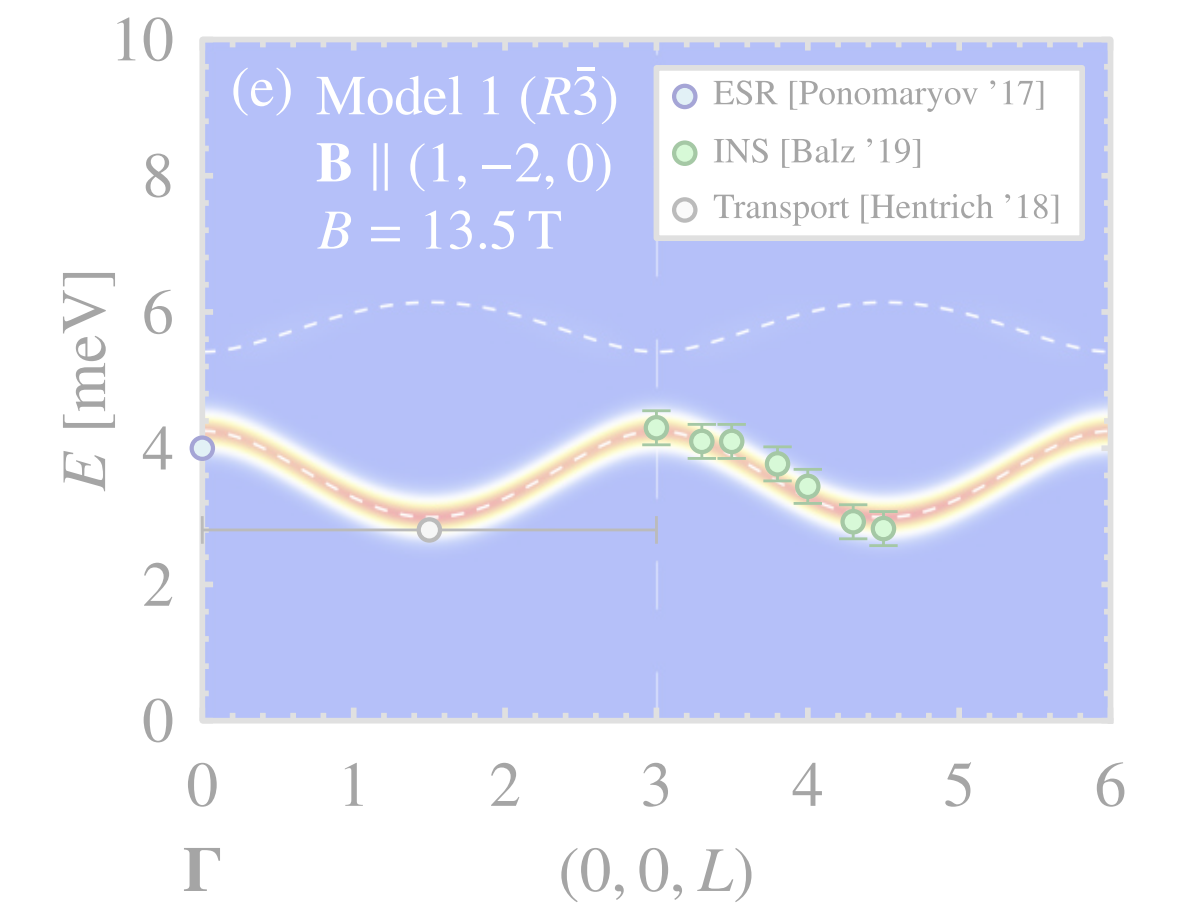
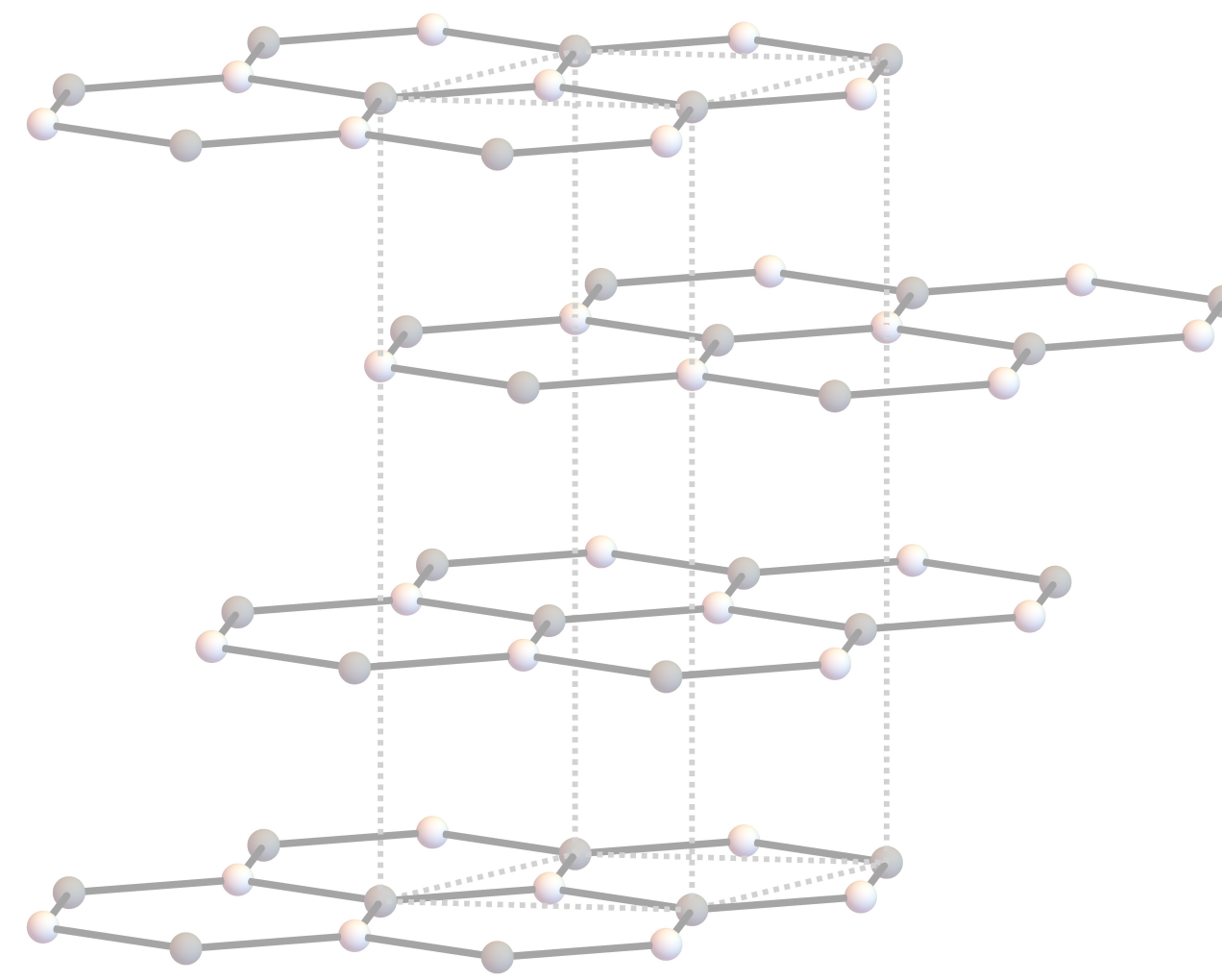
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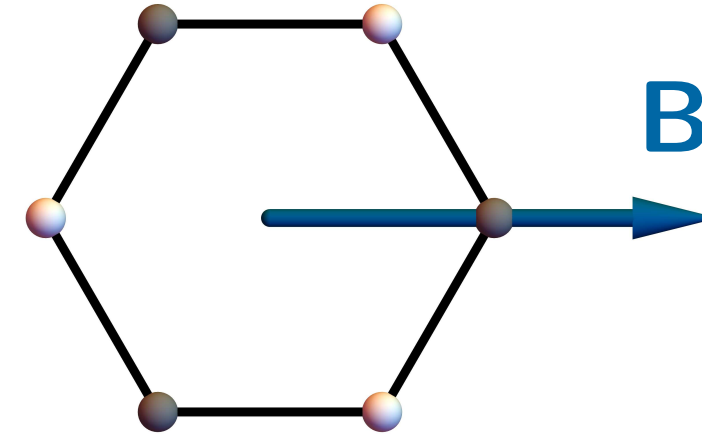
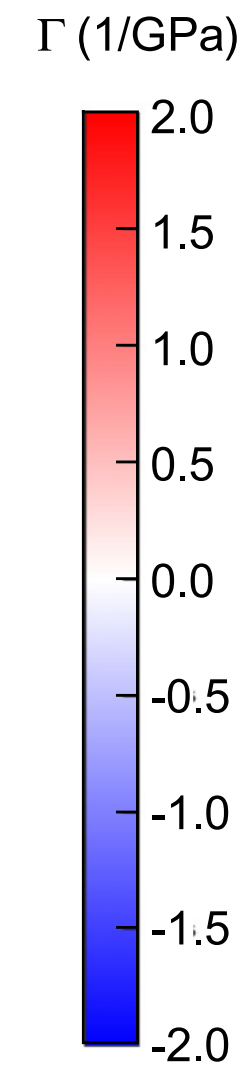
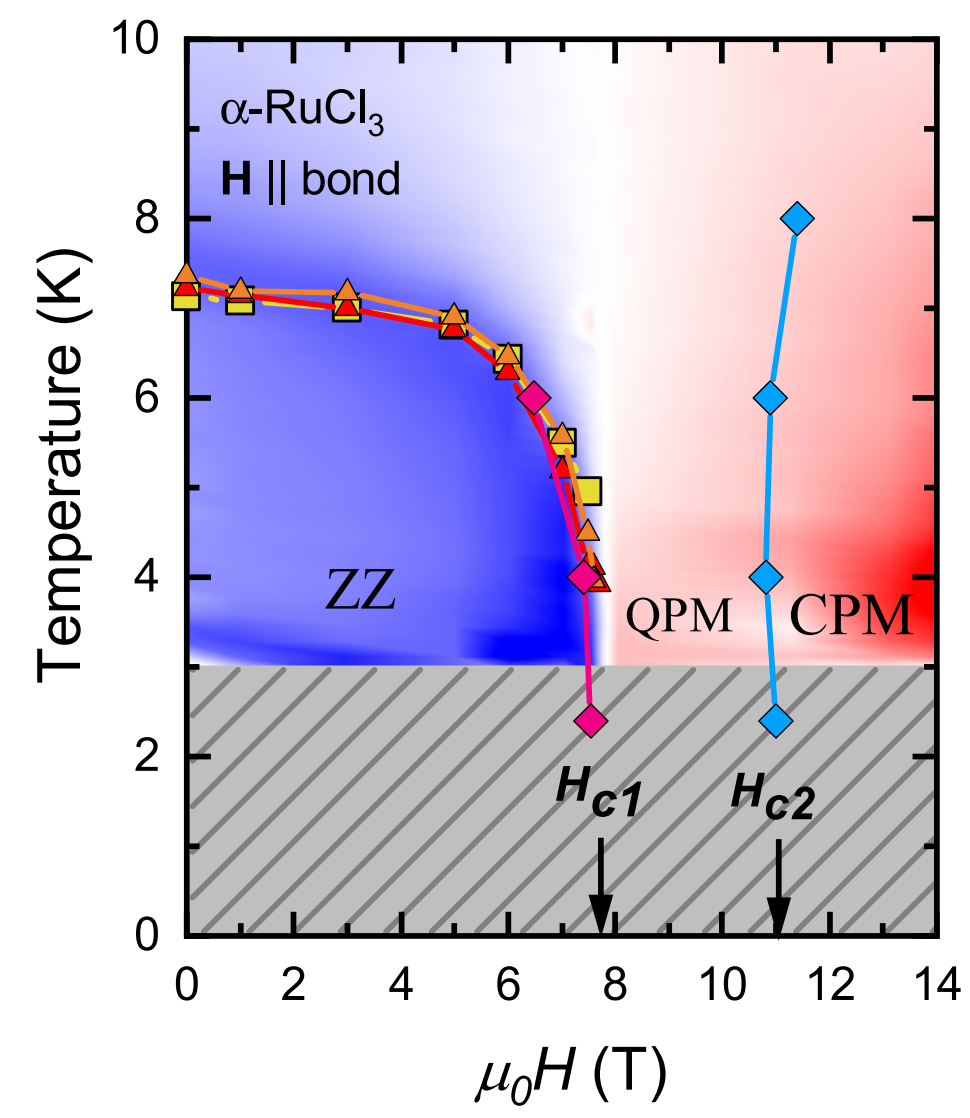
Out-of-plane interactions  $\sim 1\text{ meV}$

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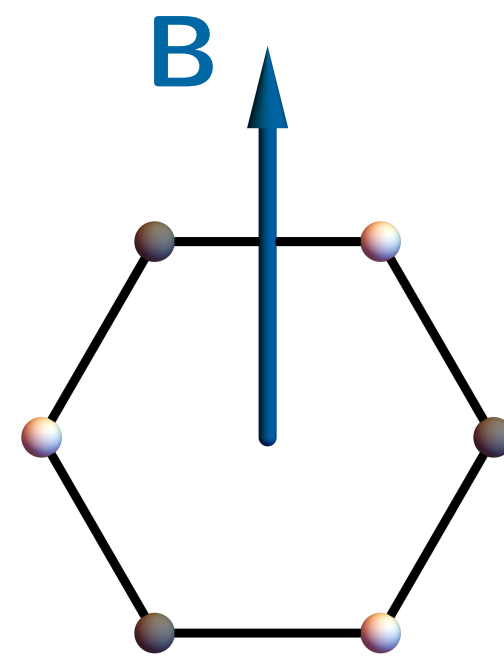
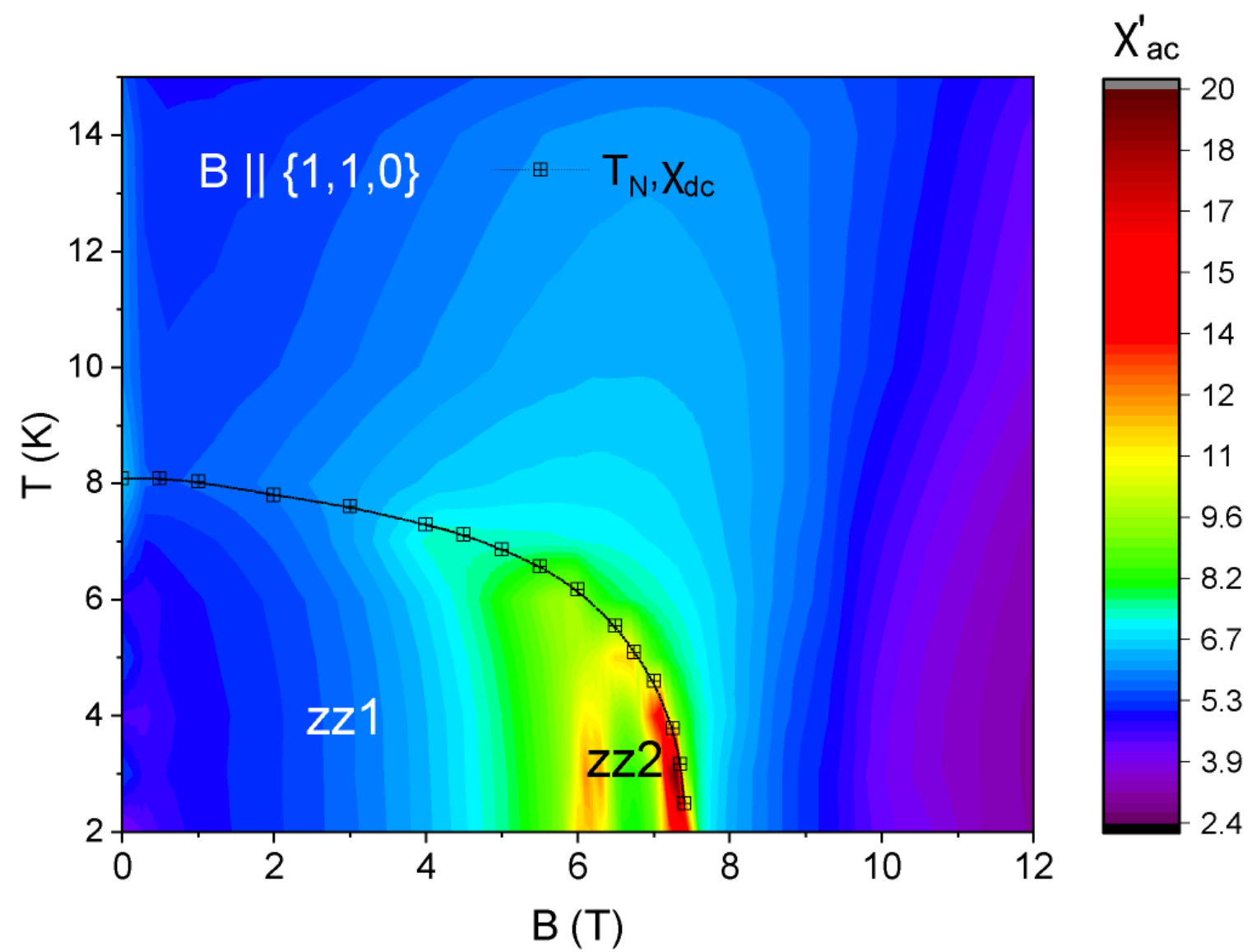
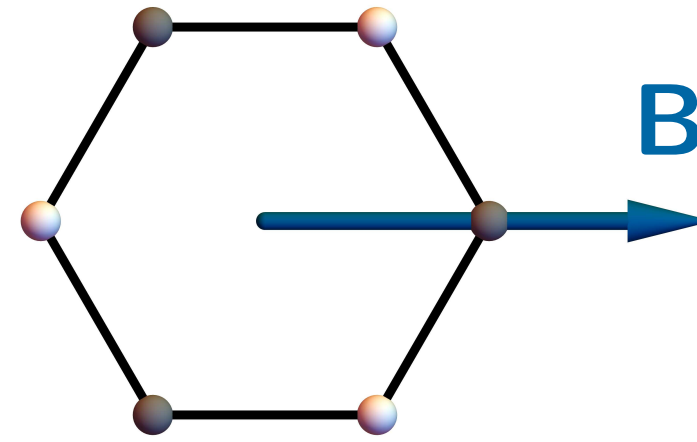
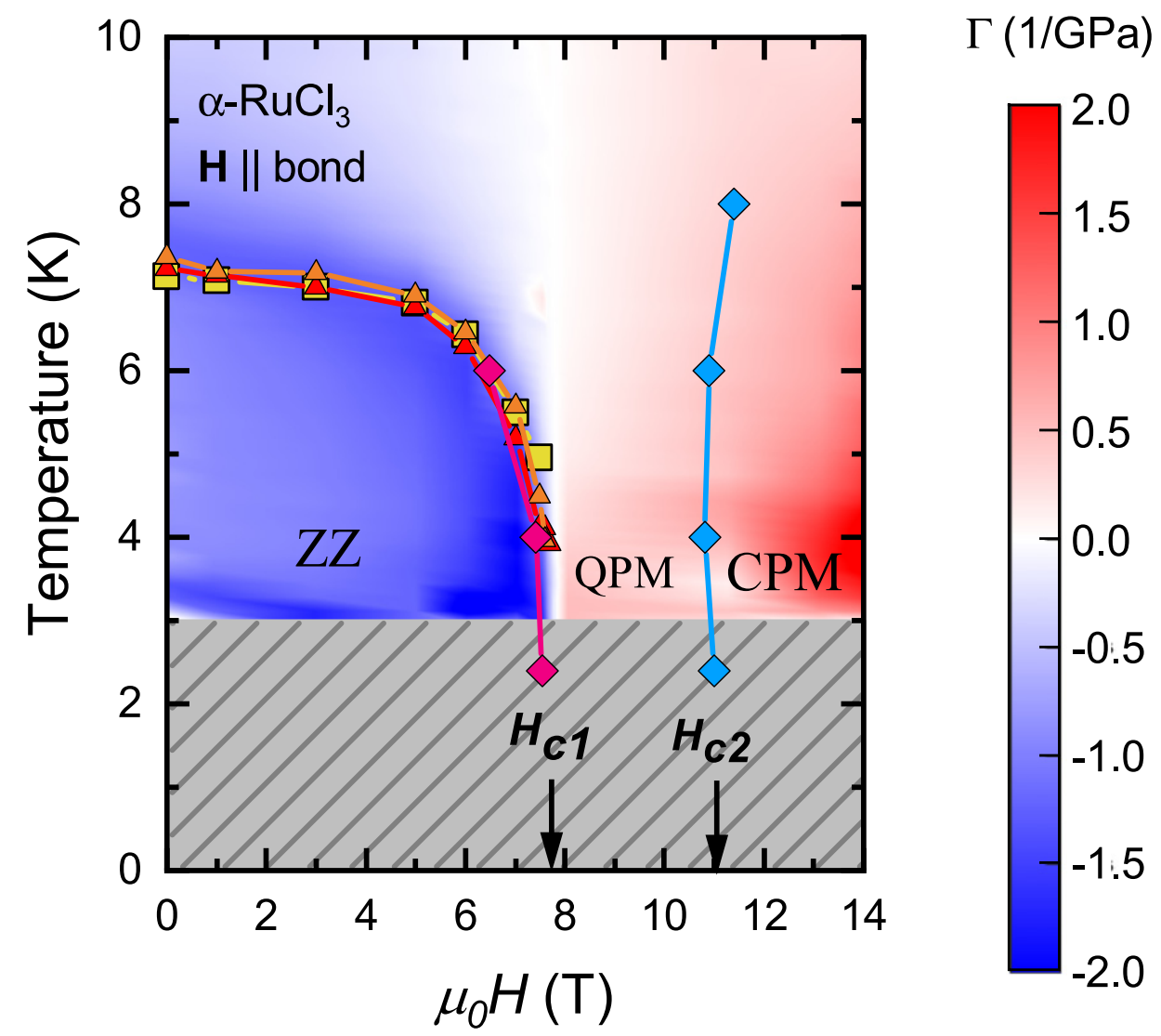
# Temperature-field phase diagram



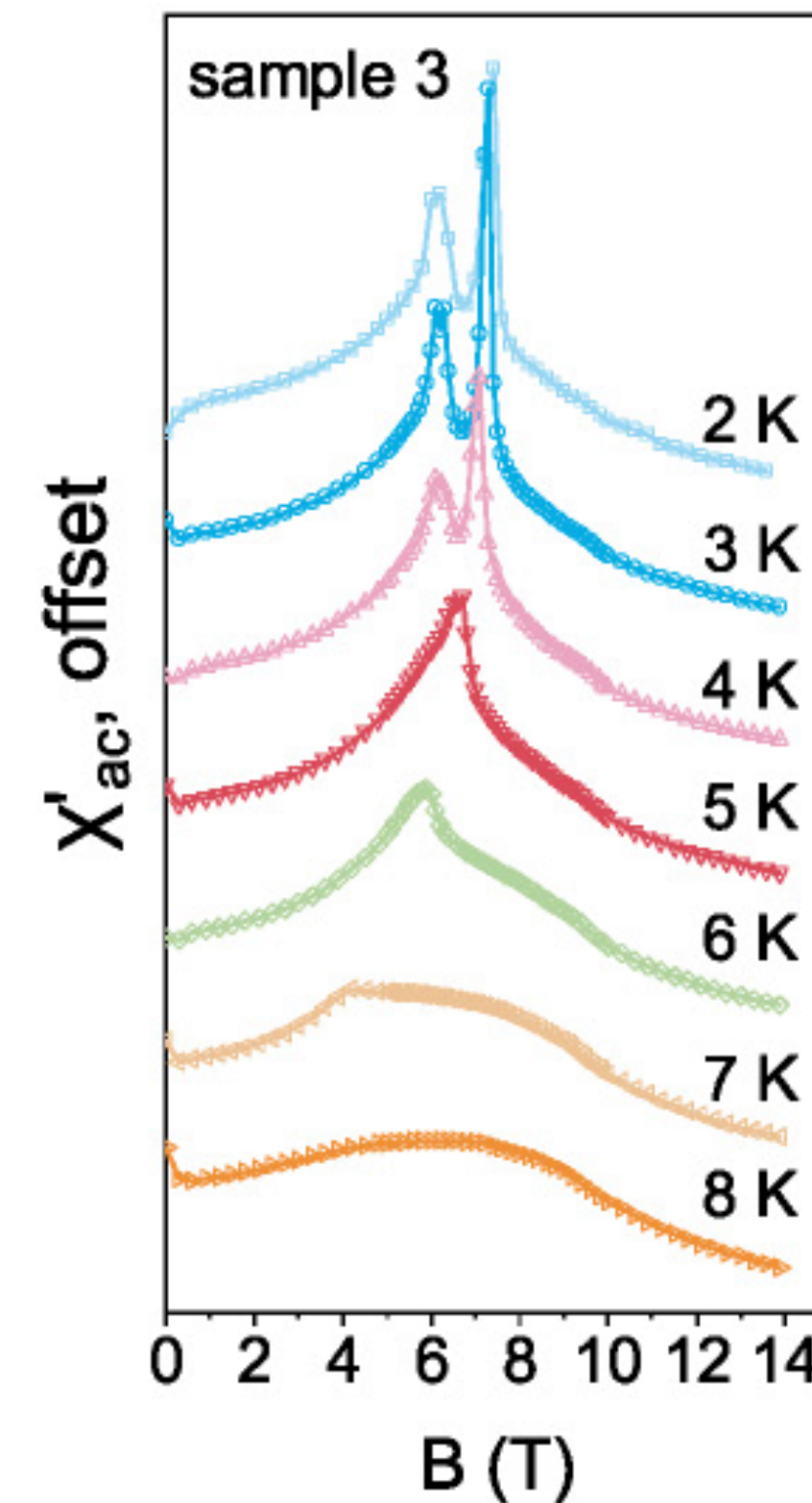
[Wolter, Corredor, LJ, *et al.*, PRB '17]  
[Gass, C onsoli, ..., LJ, *et al.*, PRB '20]

...

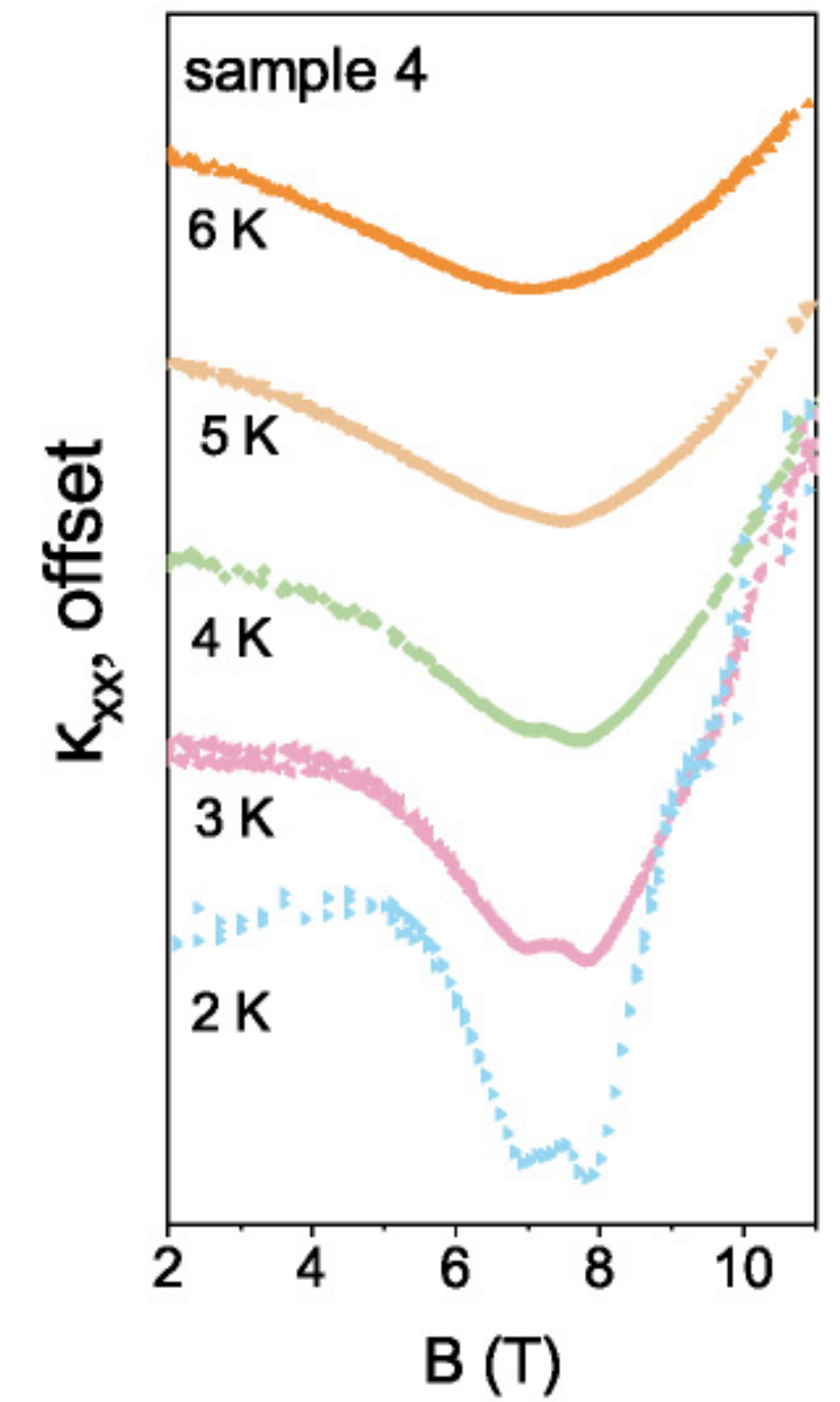
# Temperature-field phase diagram



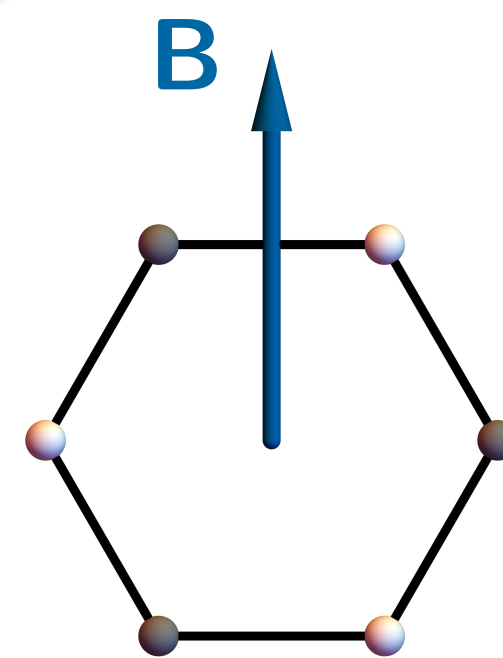
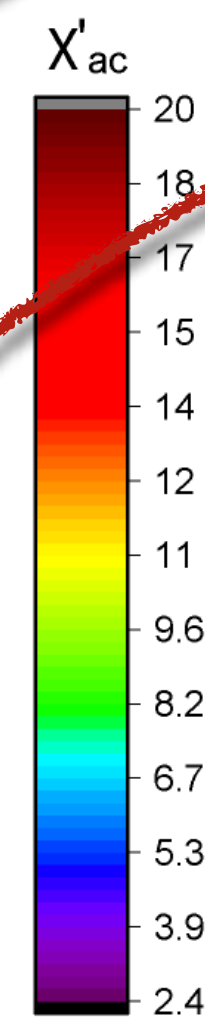
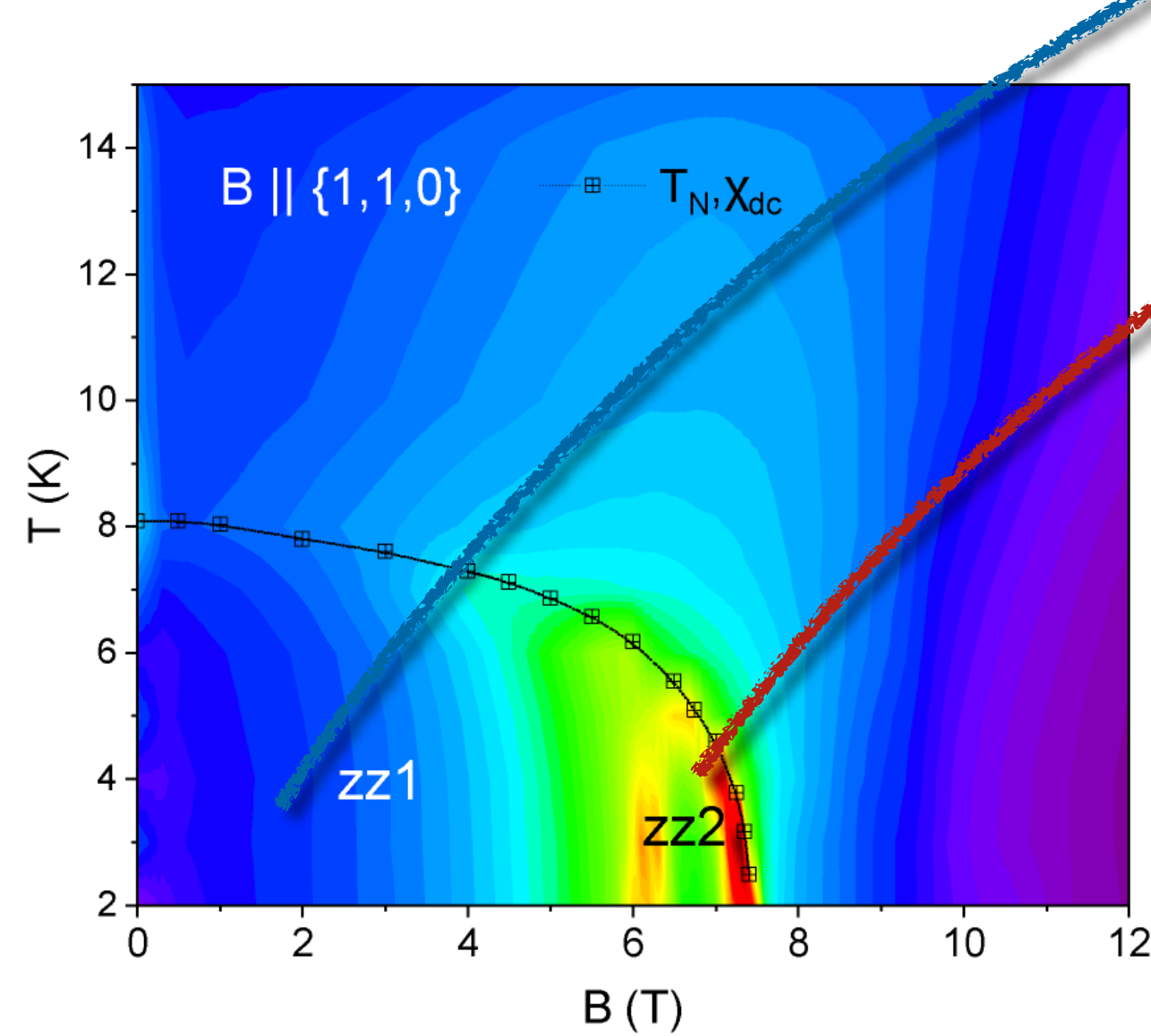
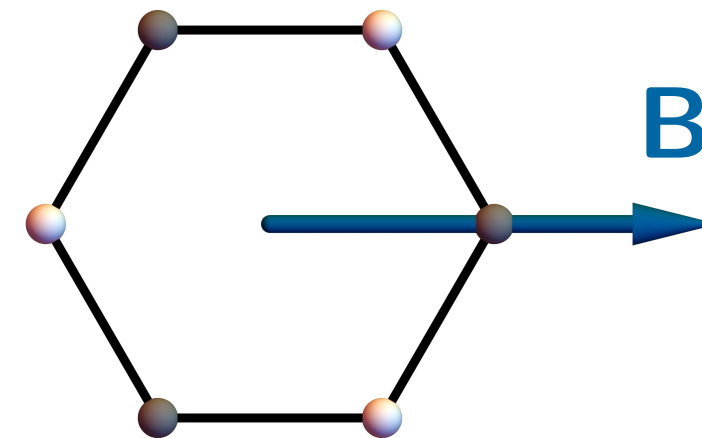
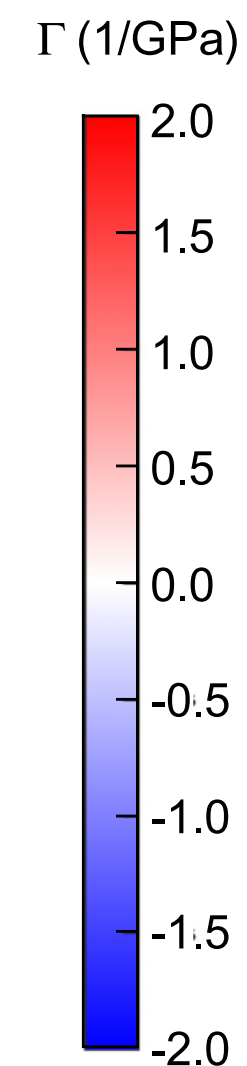
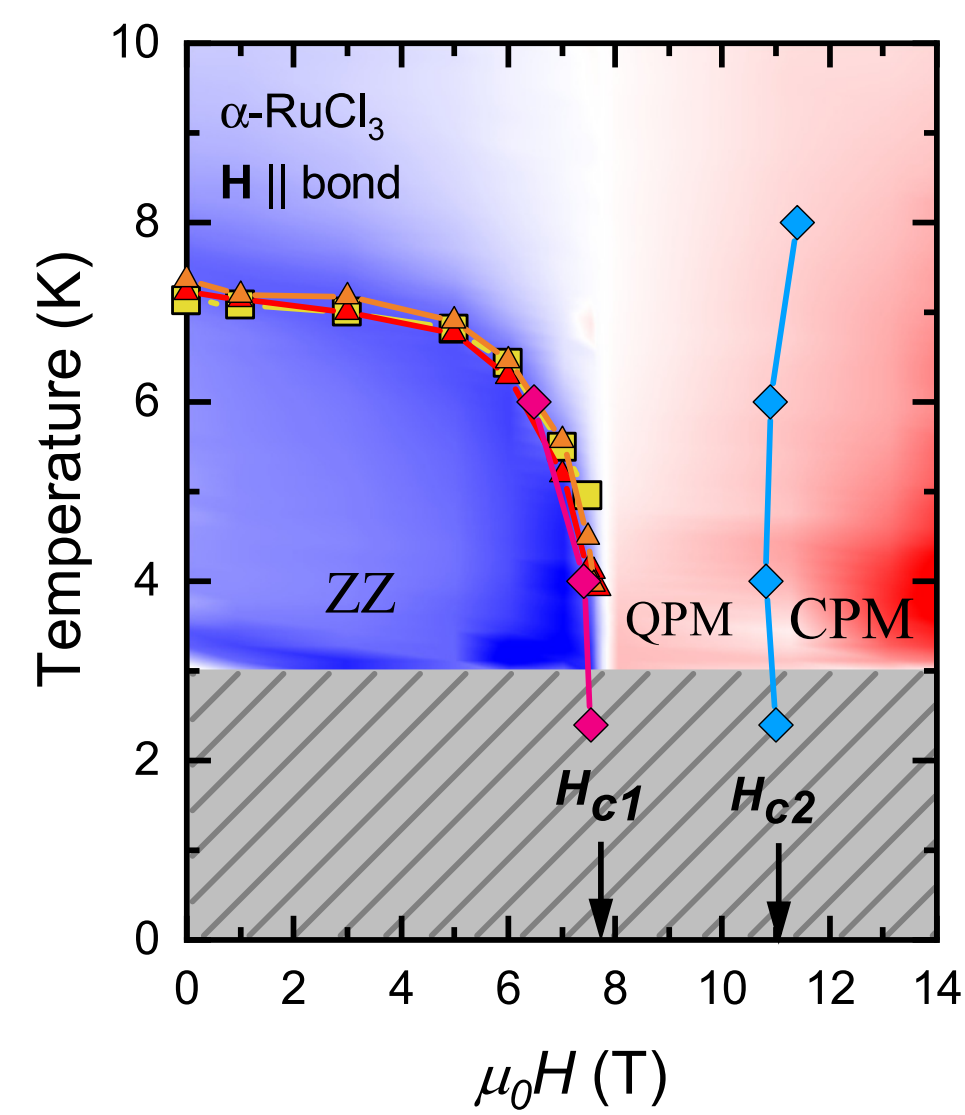
AC susceptibility



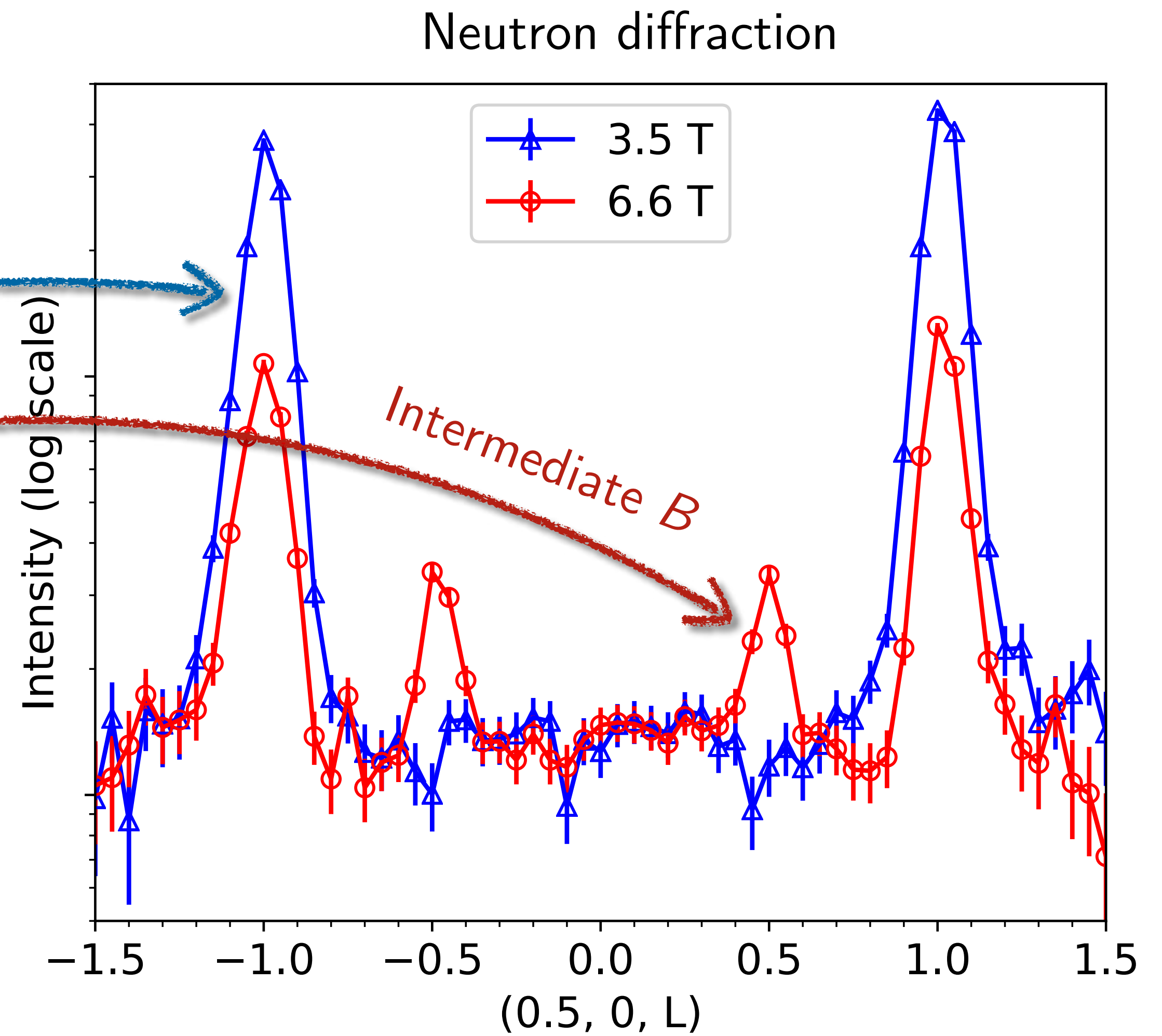
Thermal conductivity



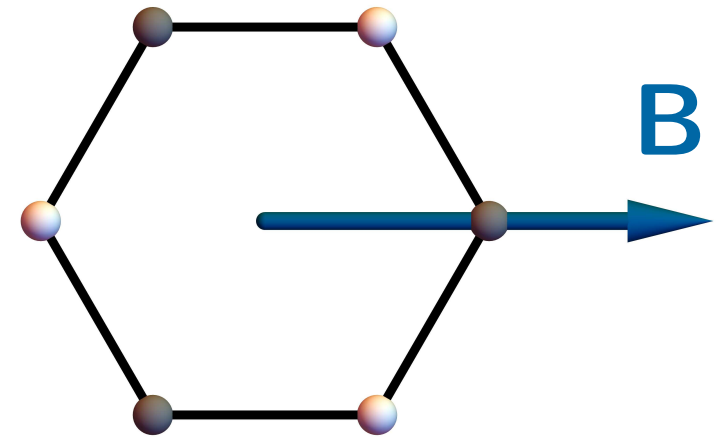
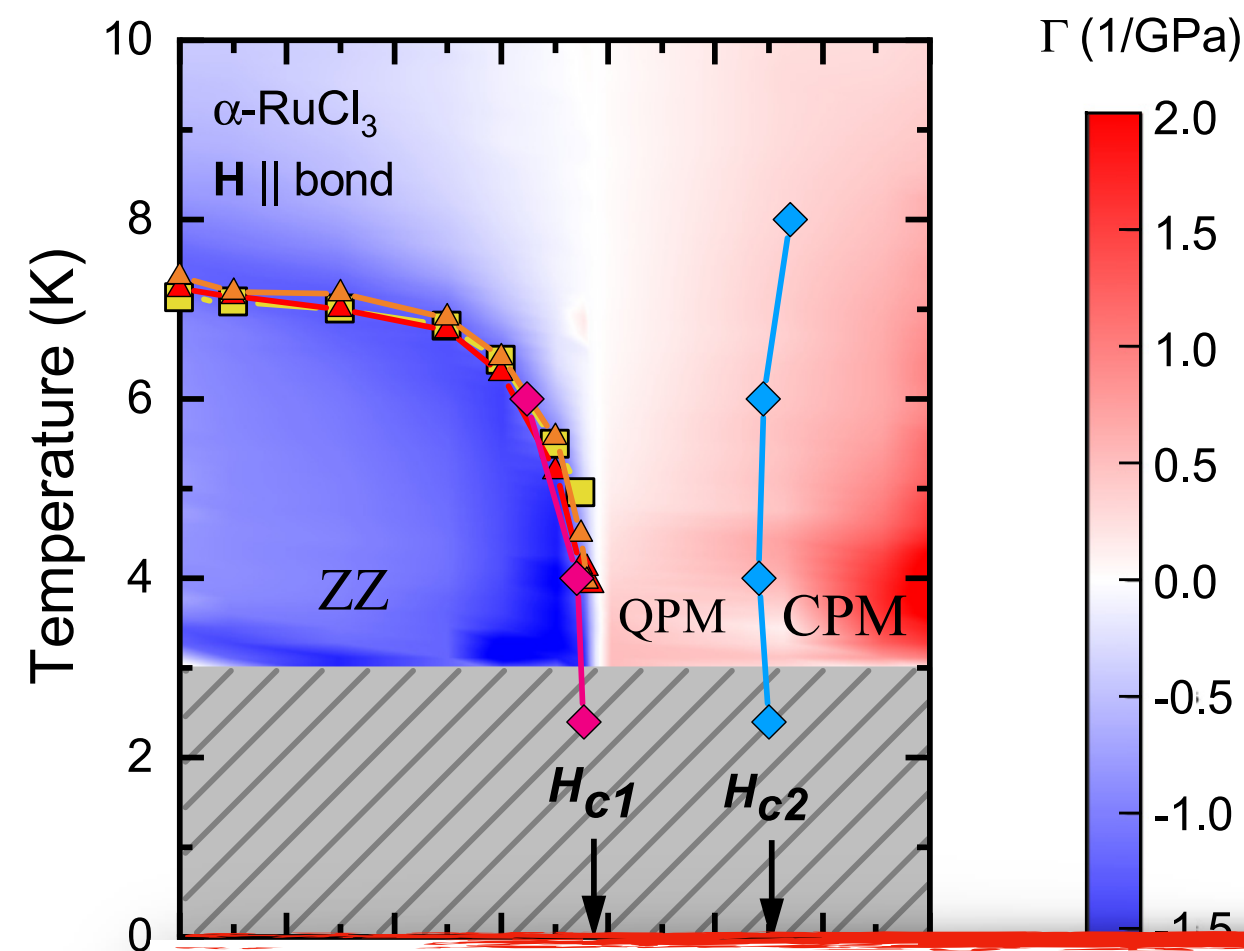
# Temperature-field phase diagram



Low  $B$

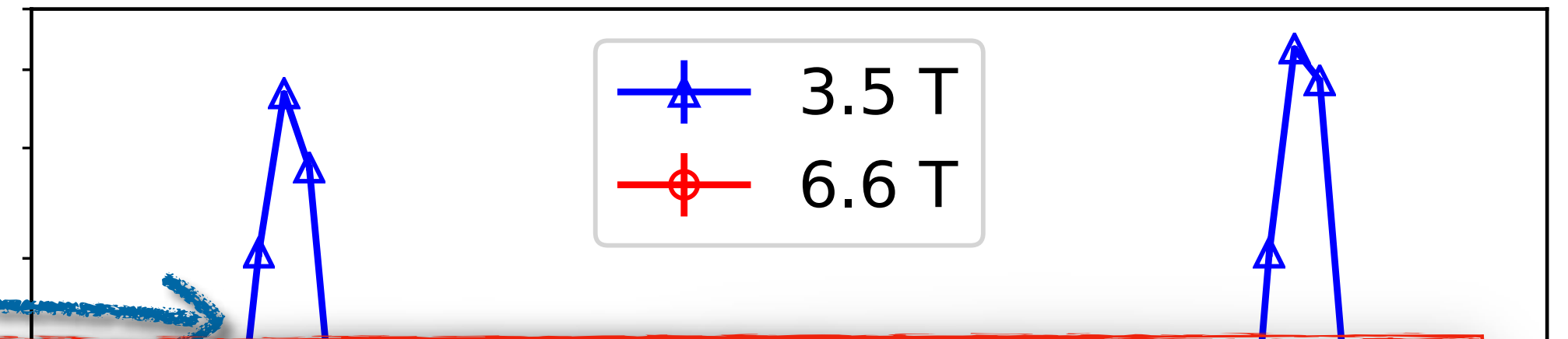


# Temperature-field phase diagram

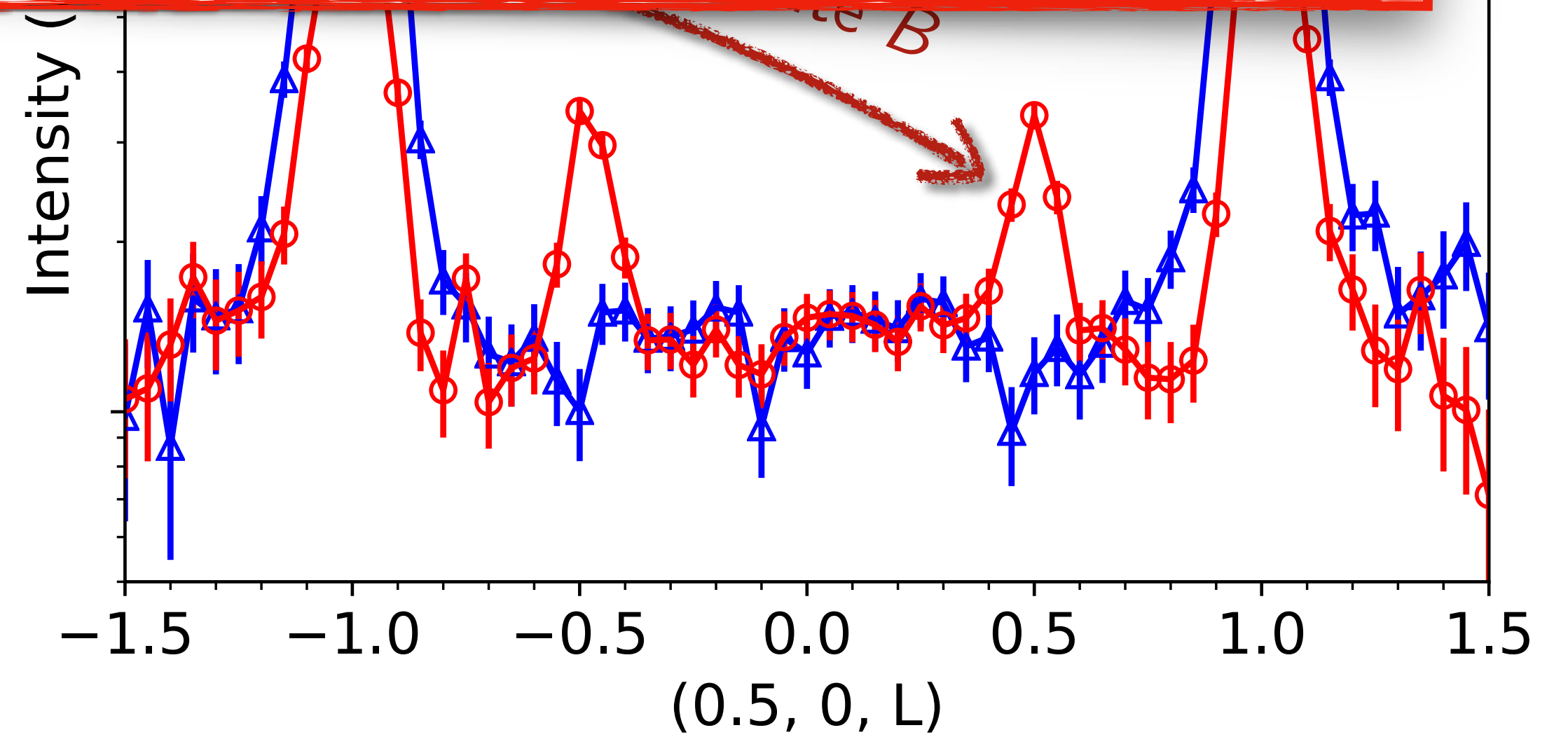
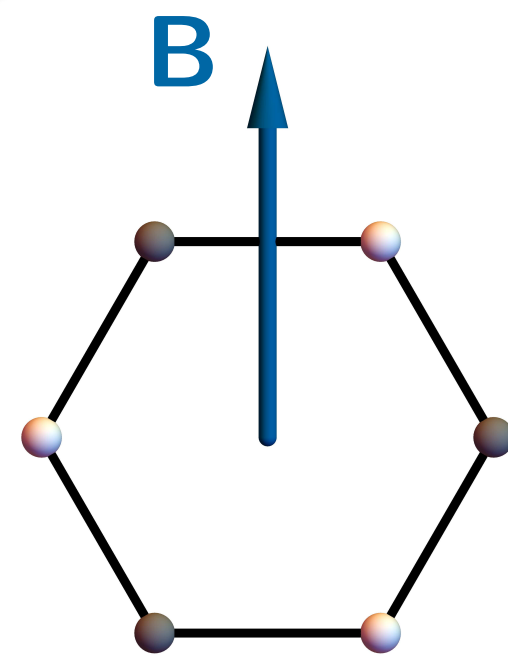
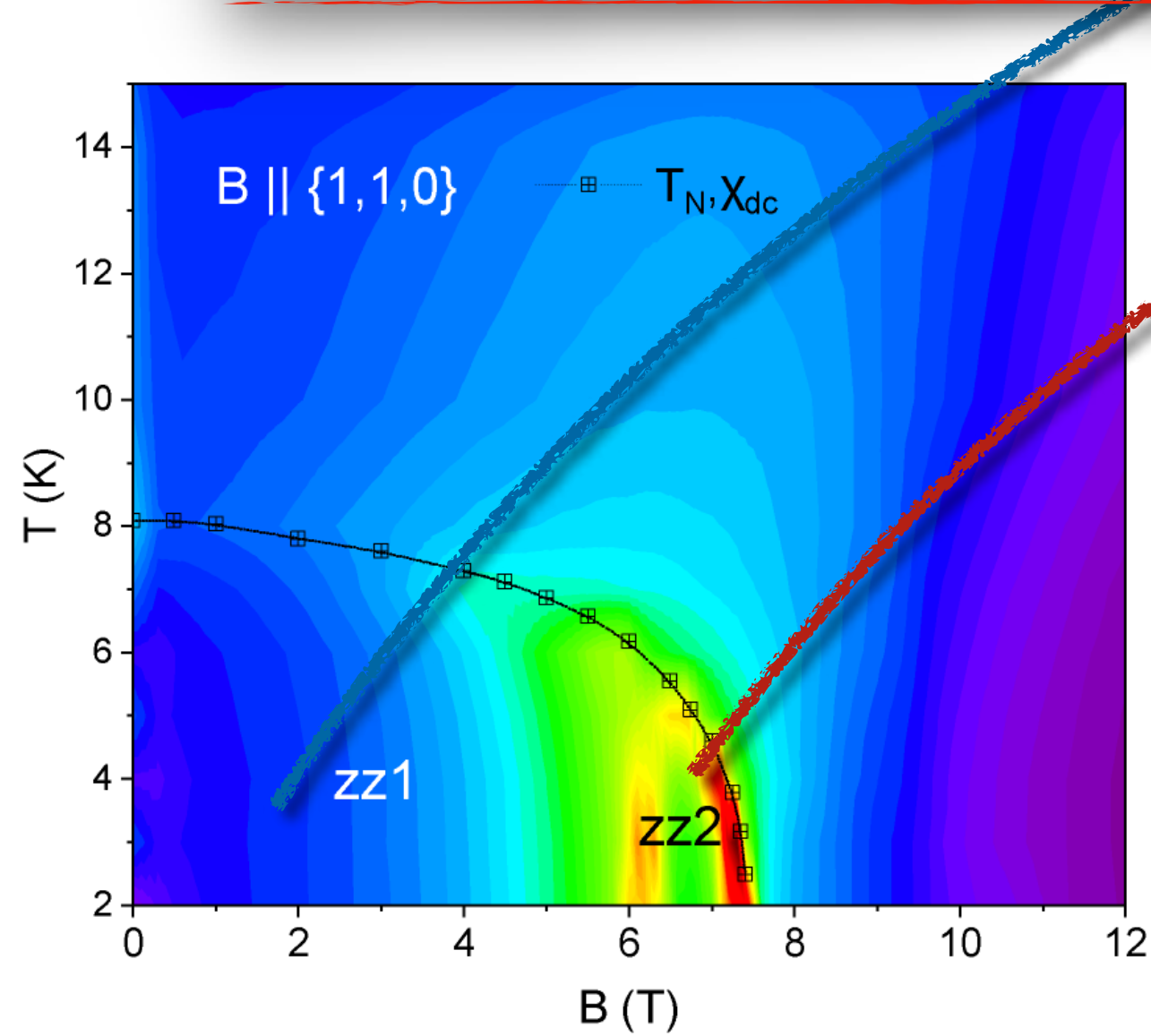


Low  $B$

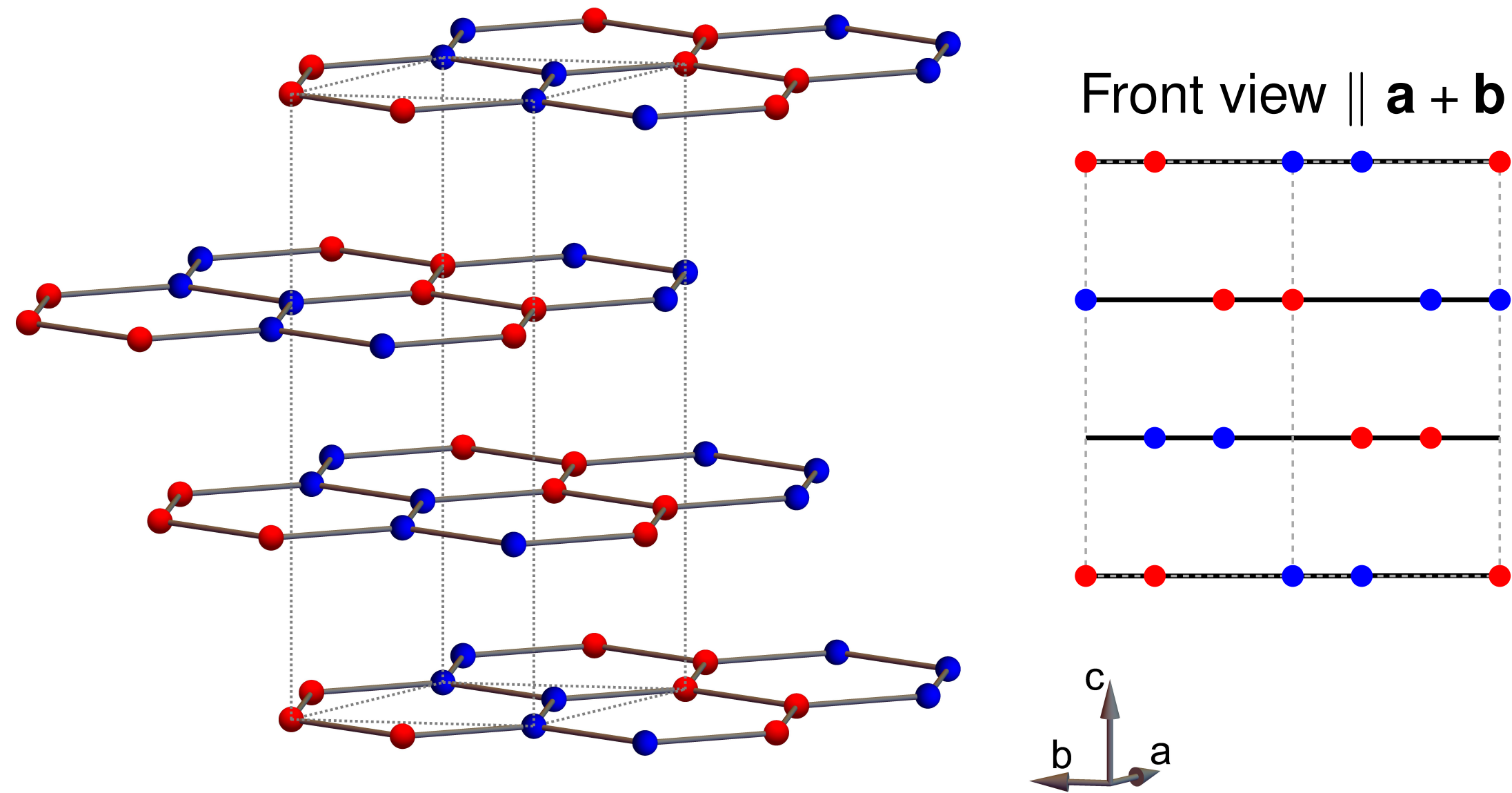
Neutron diffraction



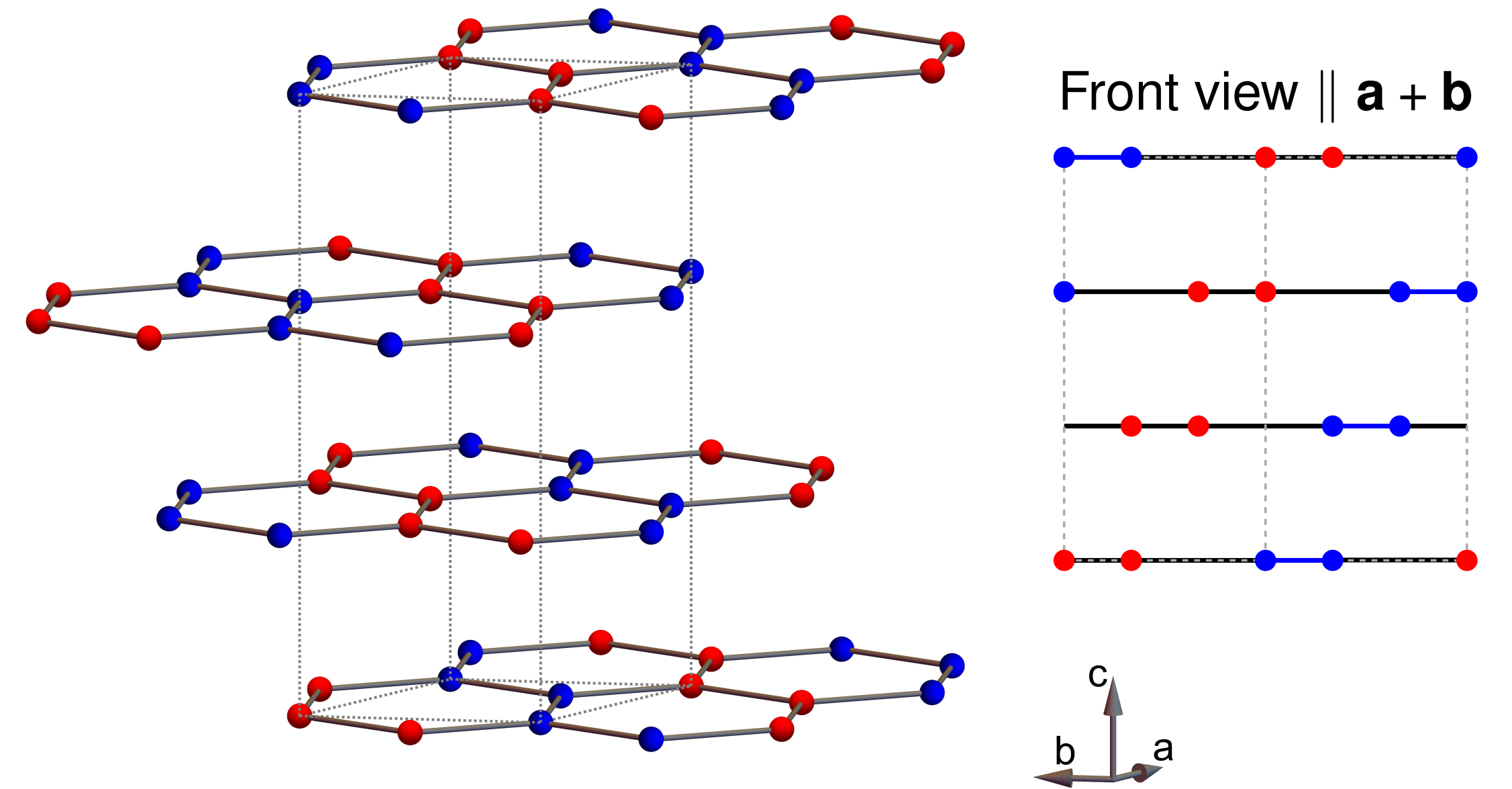
New zigzag phase with different out-of-plane stacking!



# Zigzag stackings



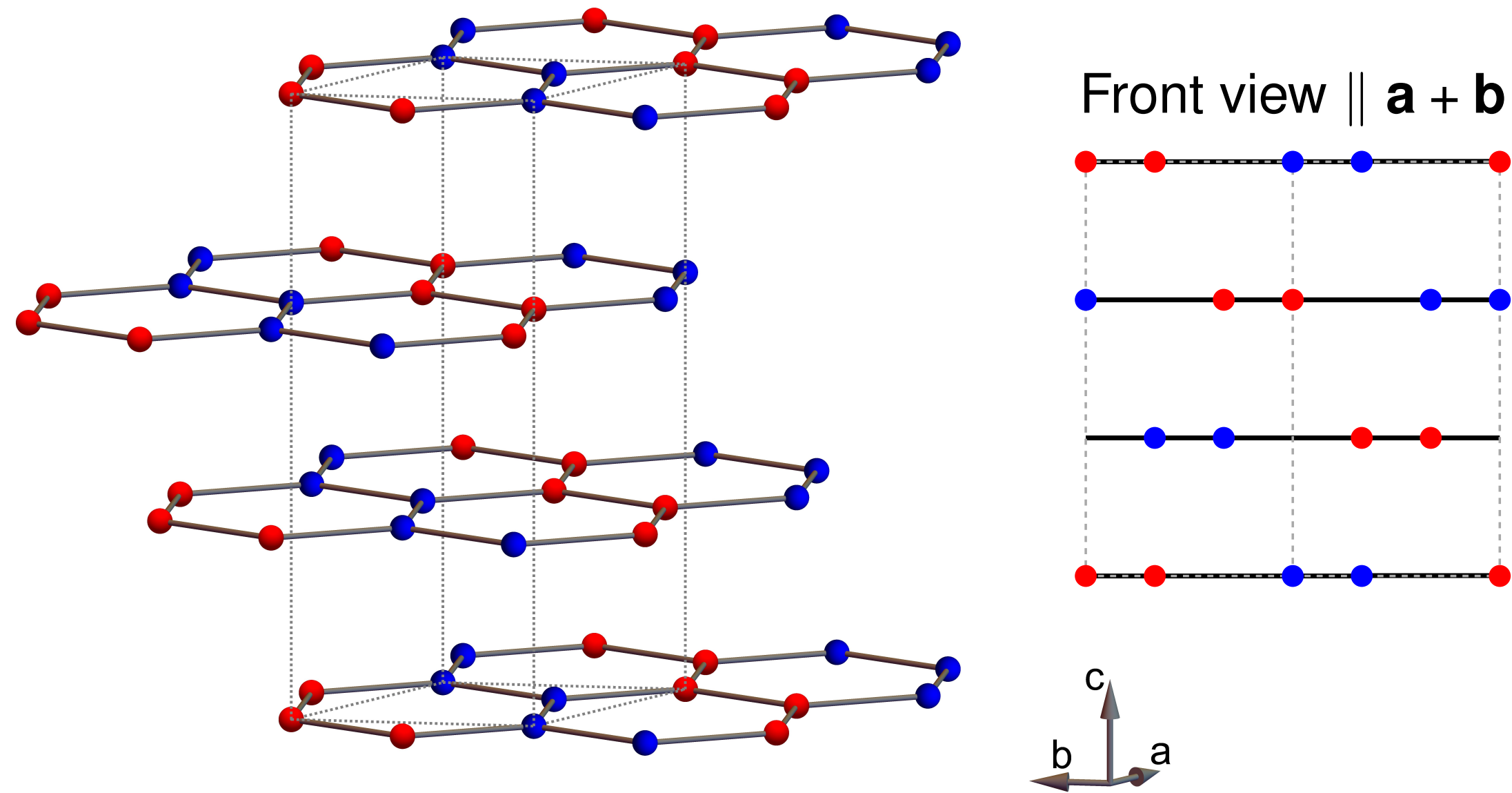
$B \lesssim 6T$  : “3-fold zigzag”  $\mathbf{Q} = (\frac{1}{2}, 0, \pm 1)$



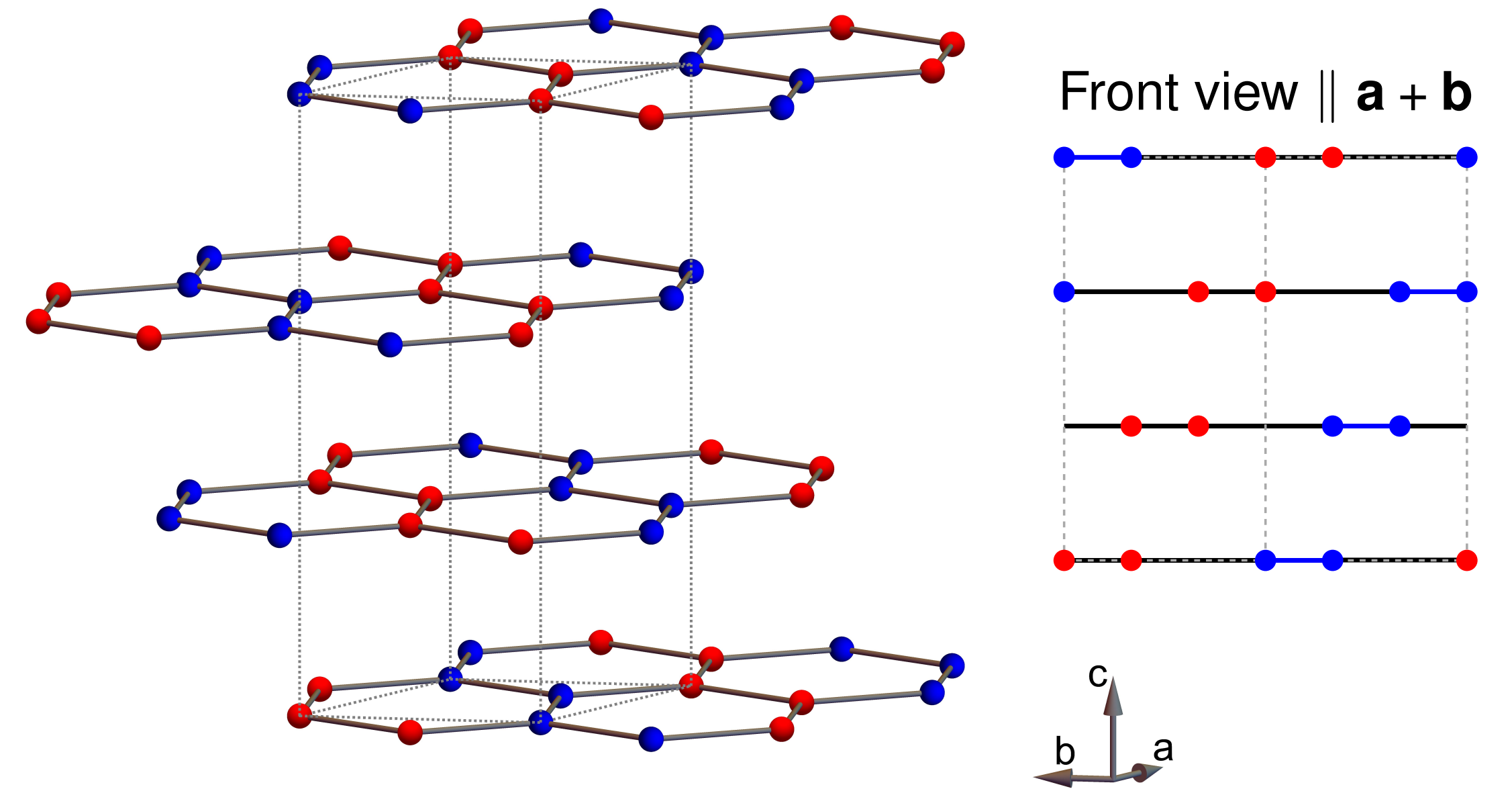
$6T \lesssim B \lesssim 7.5T$  : “6-fold zigzag”  $\mathbf{Q} = (\frac{1}{2}, 0, \pm \frac{1}{2})$



# Zigzag stackings



$B \lesssim 6 \text{ T} : \quad \text{“3-fold zigzag”} \quad \mathbf{Q} = \left(\frac{1}{2}, 0, \pm 1\right)$

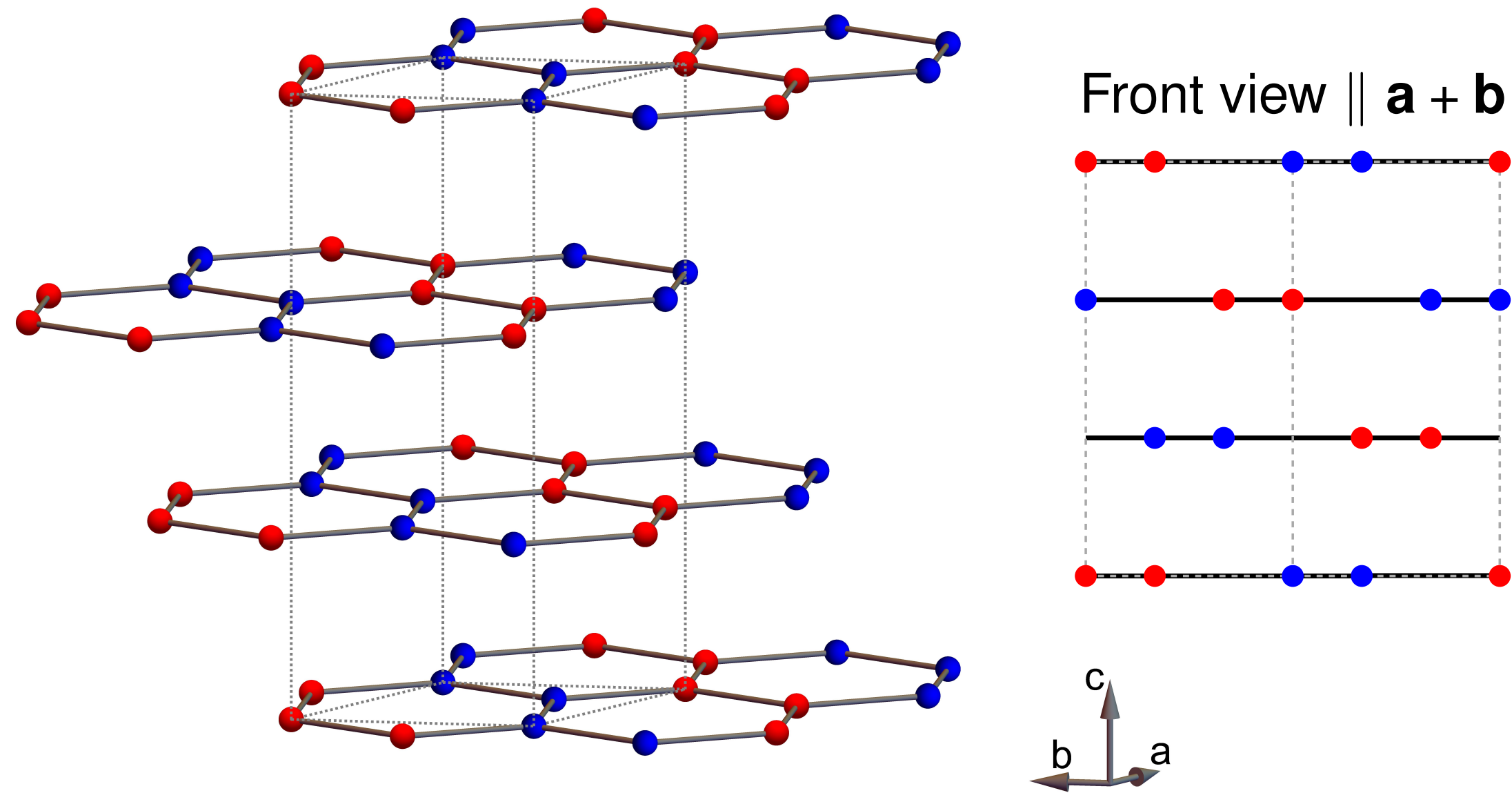


$6 \text{ T} \lesssim B \lesssim 7.5 \text{ T} : \quad \text{“6-fold zigzag”} \quad \mathbf{Q} = \left(\frac{1}{2}, 0, \pm \frac{1}{2}\right)$

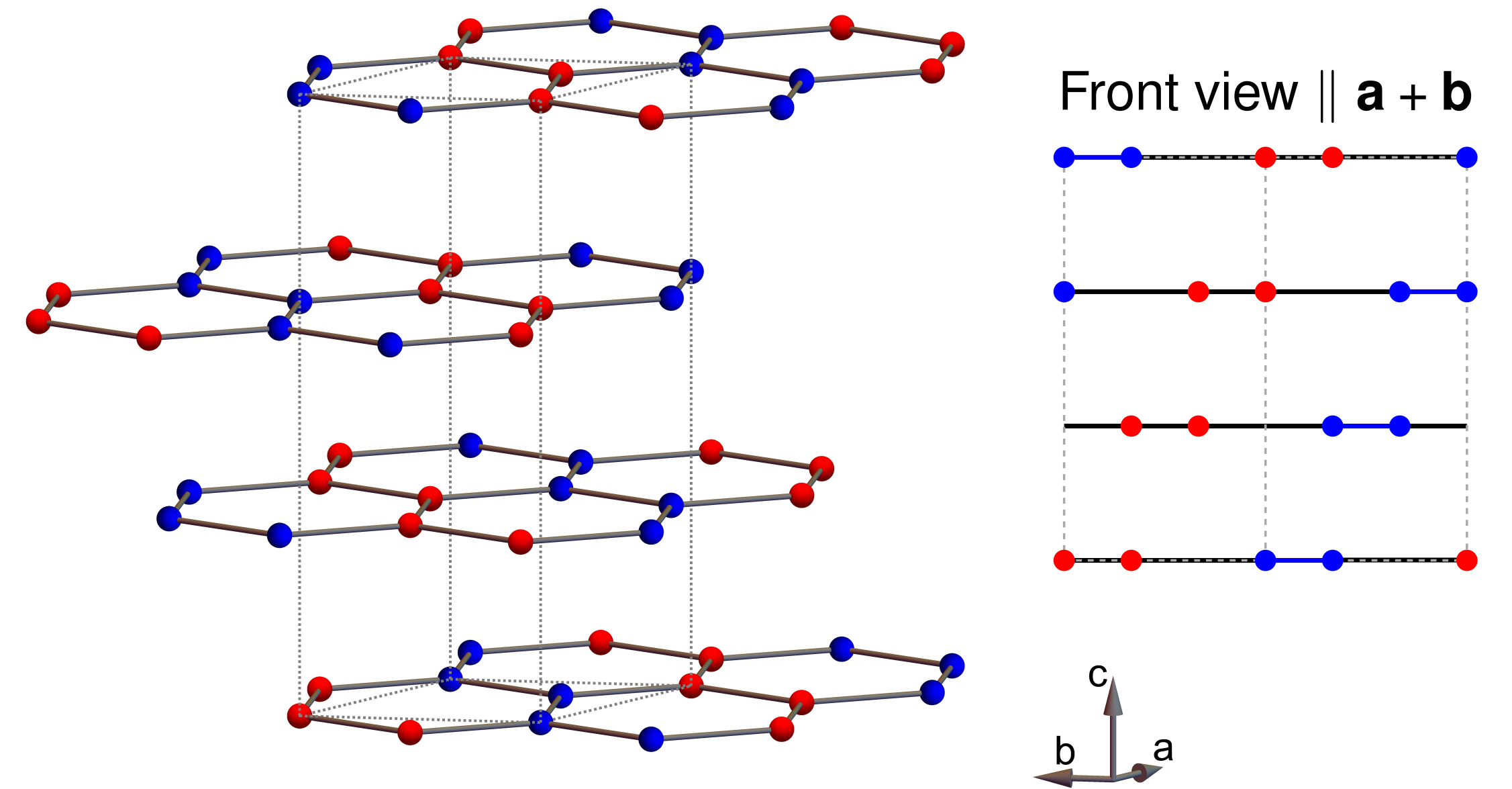
Possible mechanisms:

- Magnetoelastic coupling?
- Order from disorder?
- Anisotropic interactions?
- ...

# Zigzag stackings



$B \lesssim 6 \text{ T}$  : “3-fold zigzag”  $\mathbf{Q} = (\frac{1}{2}, 0, \pm 1)$



$6 \text{ T} \lesssim B \lesssim 7.5 \text{ T}$  : “6-fold zigzag”  $\mathbf{Q} = (\frac{1}{2}, 0, \pm \frac{1}{2})$

Possible mechanisms:

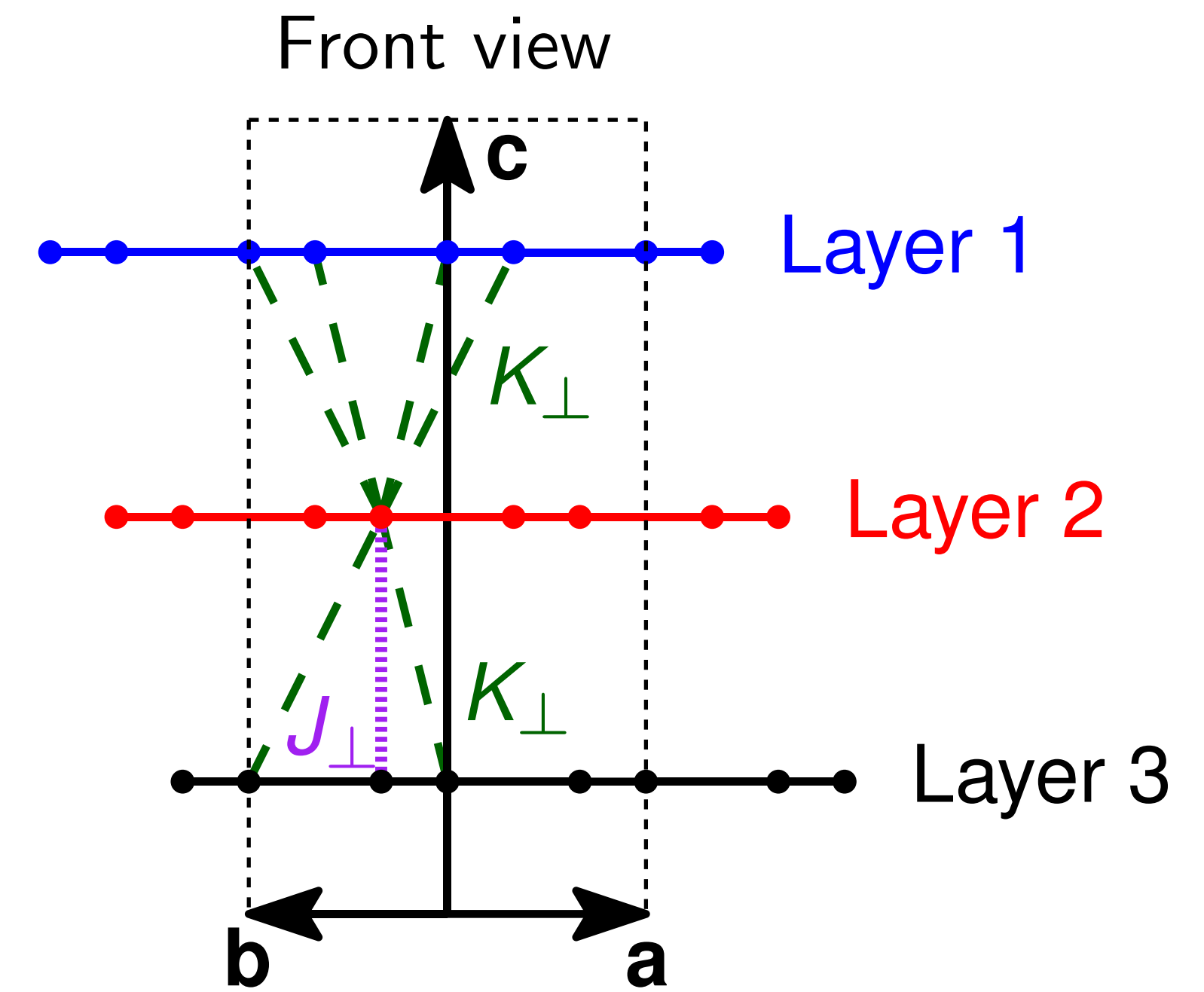
- Magnetoelastic coupling?
- Order from disorder?
- Anisotropic interactions?
- ...

Transition explainable within minimal classical spin model?

# Minimal 3D spin model

Hamiltonian:

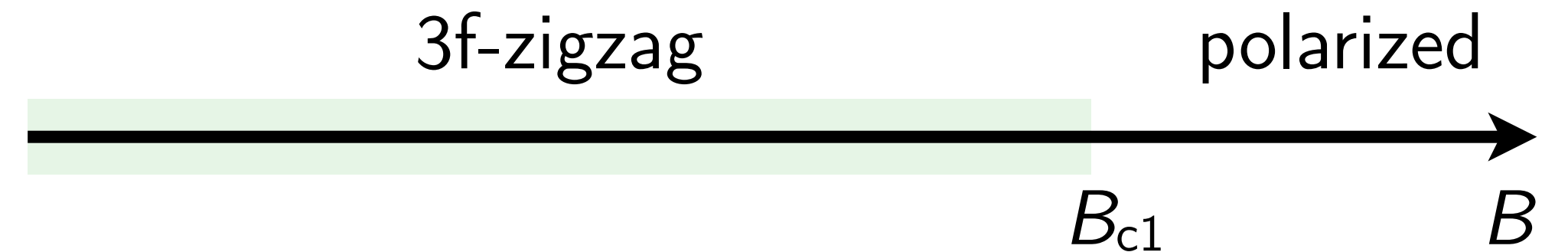
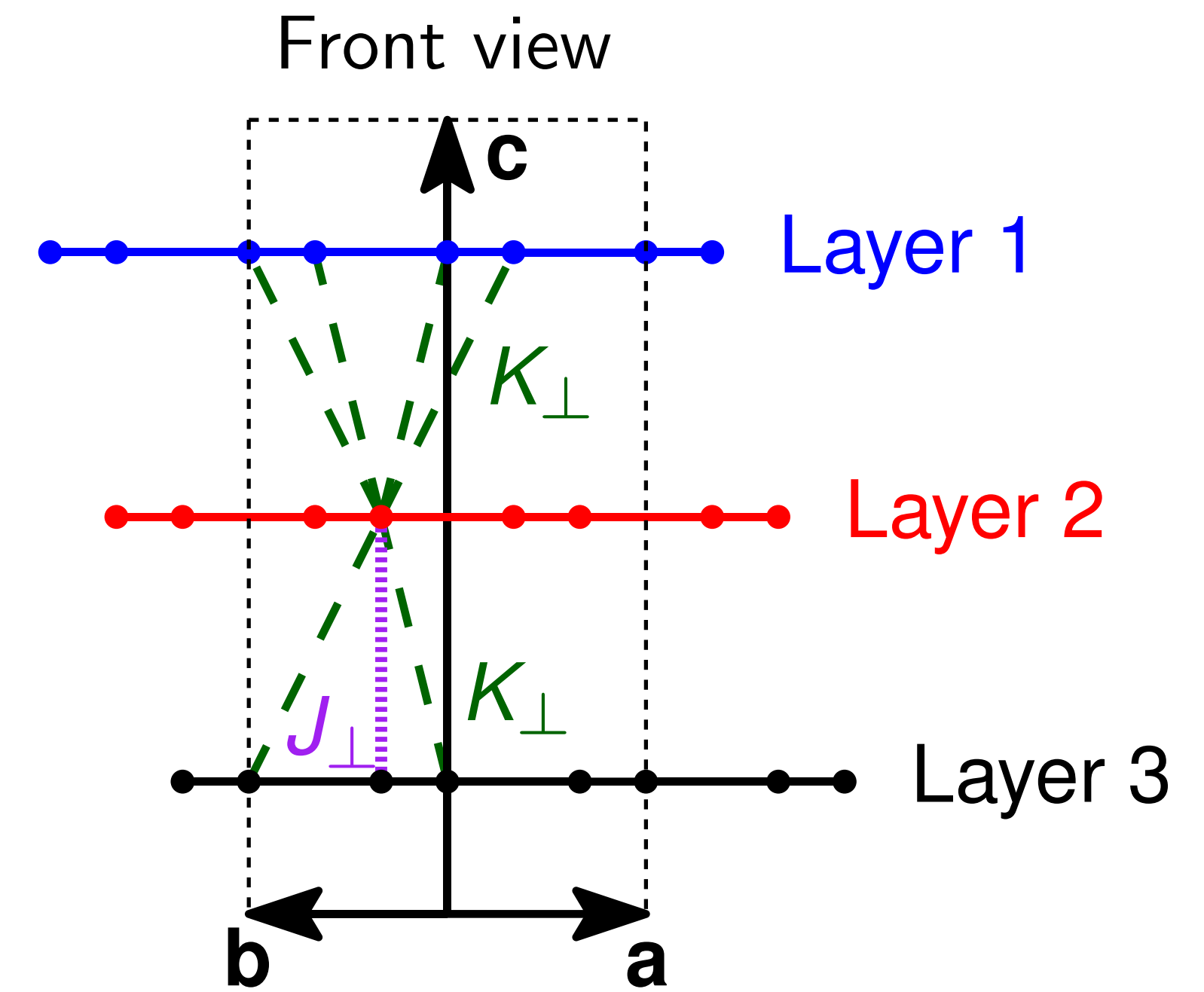
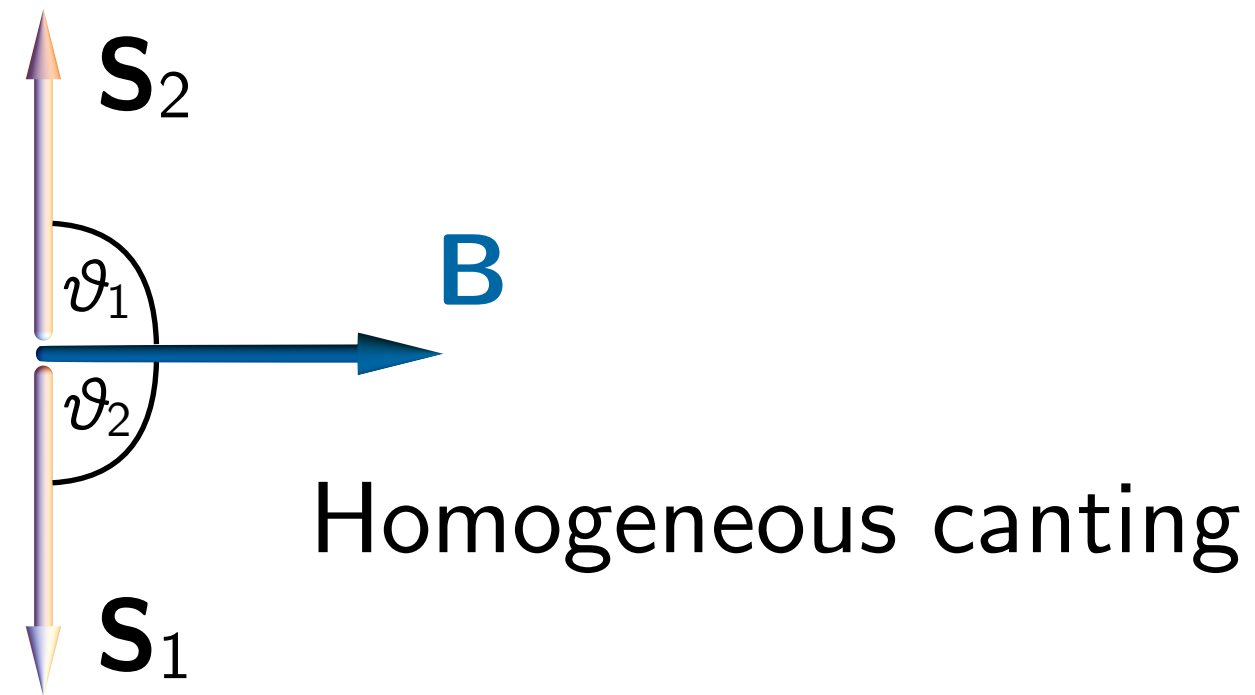
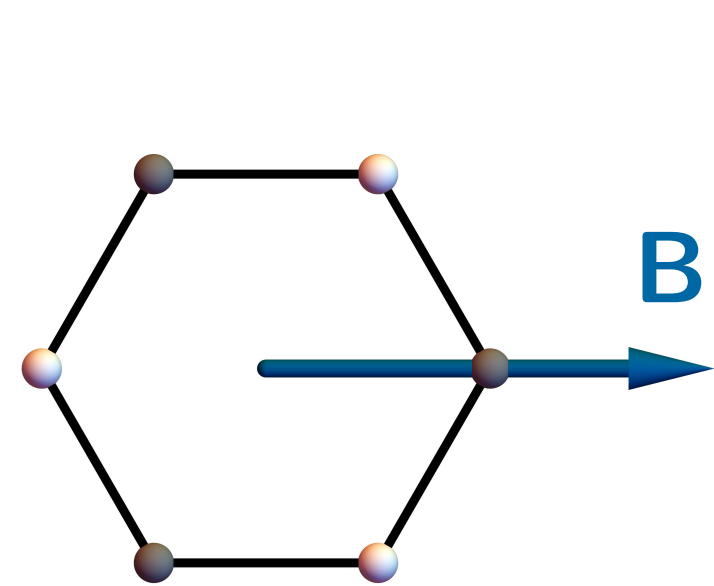
$$\mathcal{H}_{3D} = \mathcal{H}_{2D} + J_{\perp} \sum_{\langle ni, mi \rangle} \vec{S}_{n,i} \cdot \vec{S}_{m,i} + K_{\perp} \underbrace{\sum_{\langle\langle ni, mj \rangle\rangle_{\gamma}} S_{n,i}^{\gamma} S_{m,j}^{\gamma}}_{\text{Depends on absolute spin orientation}}$$



# Minimal 3D spin model

Hamiltonian:

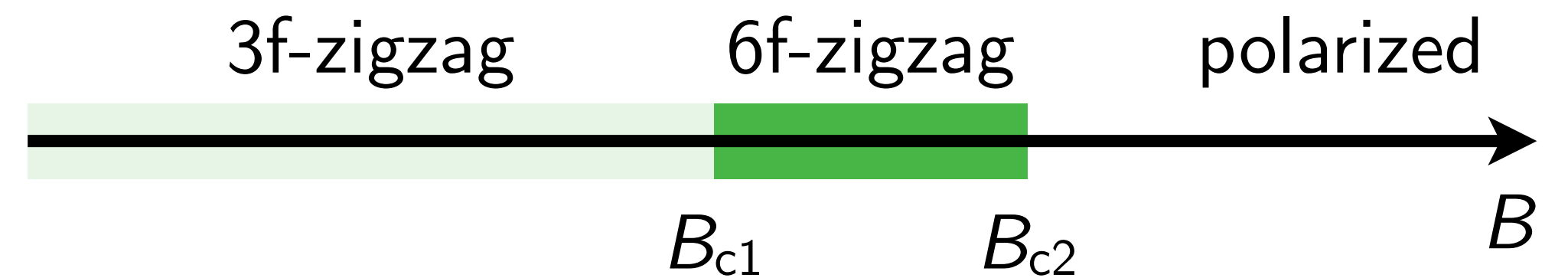
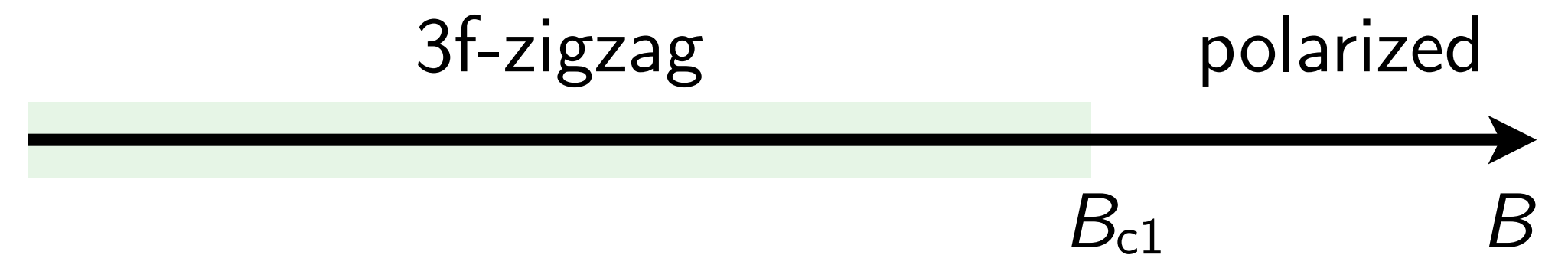
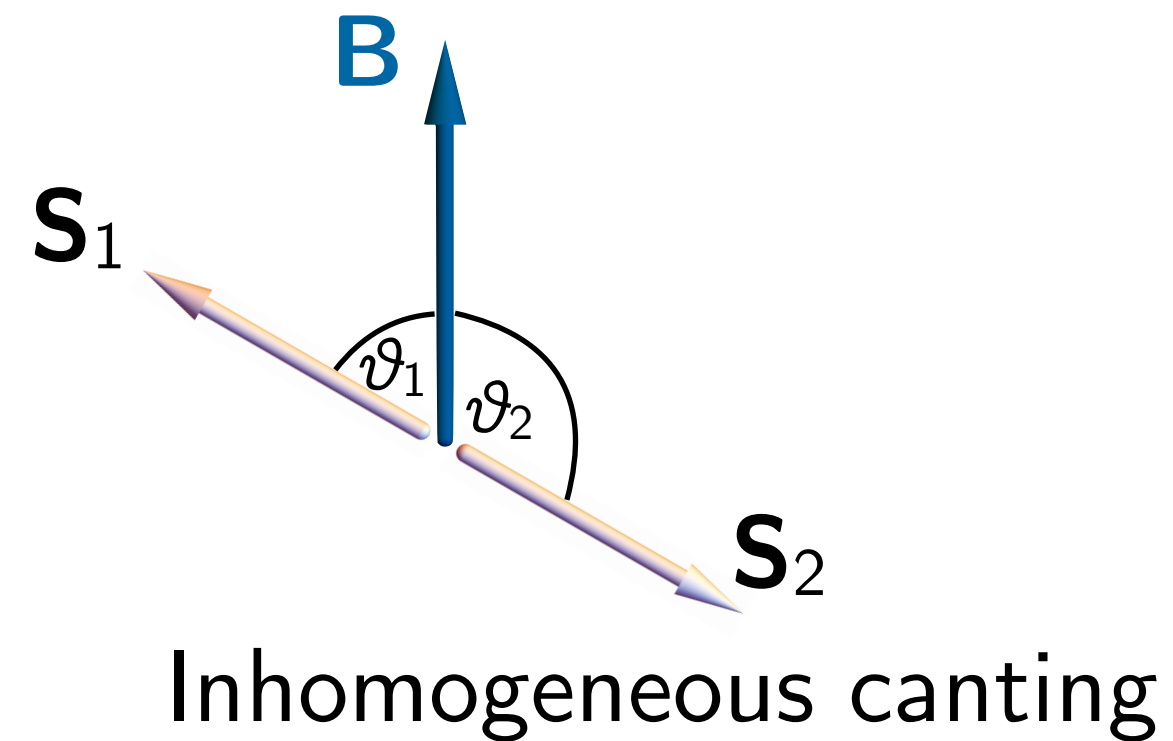
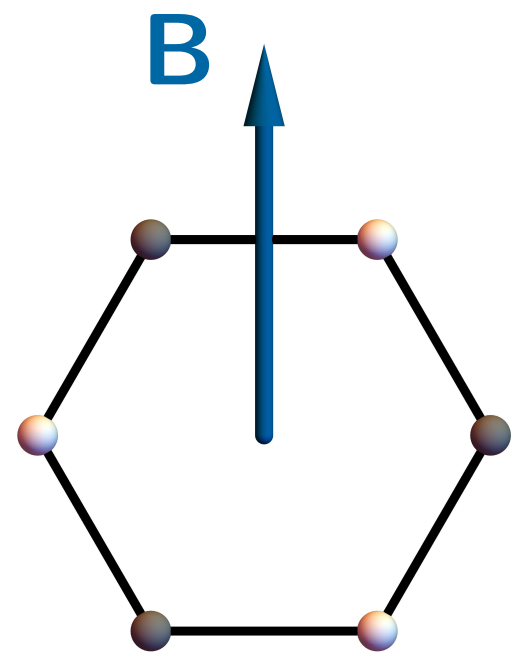
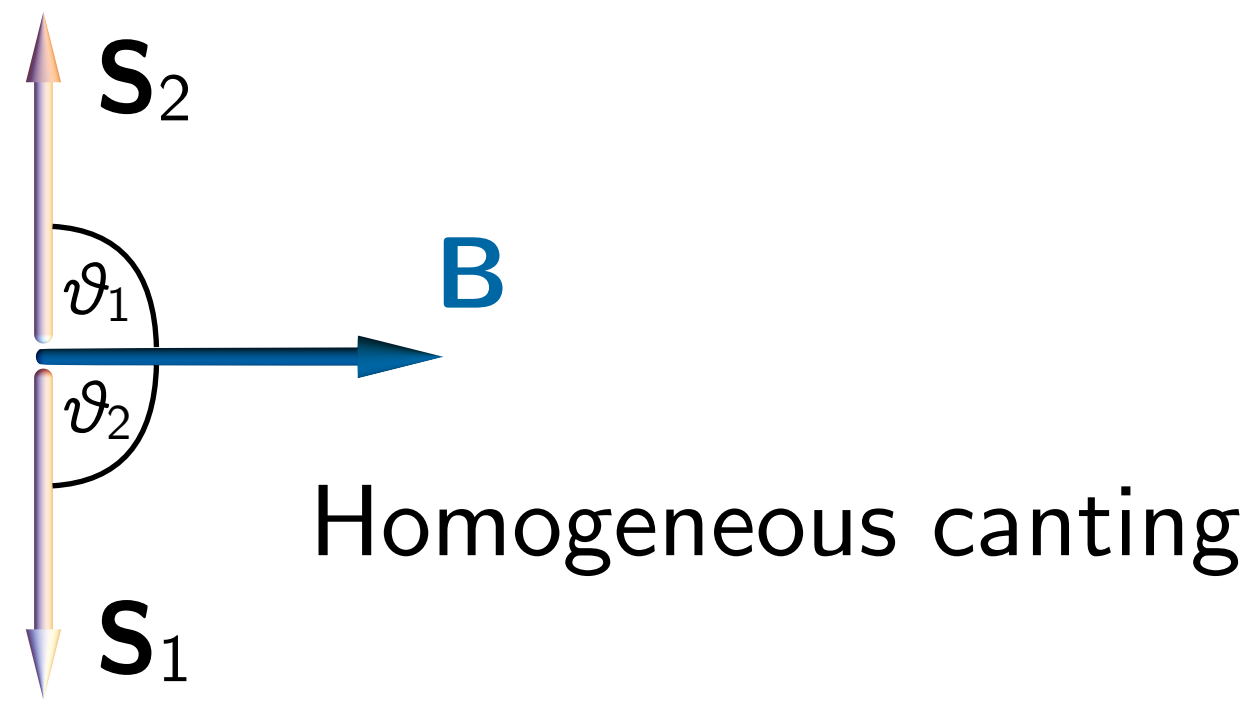
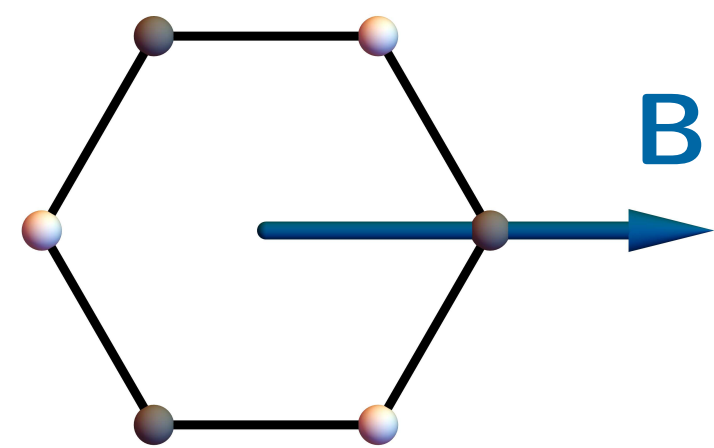
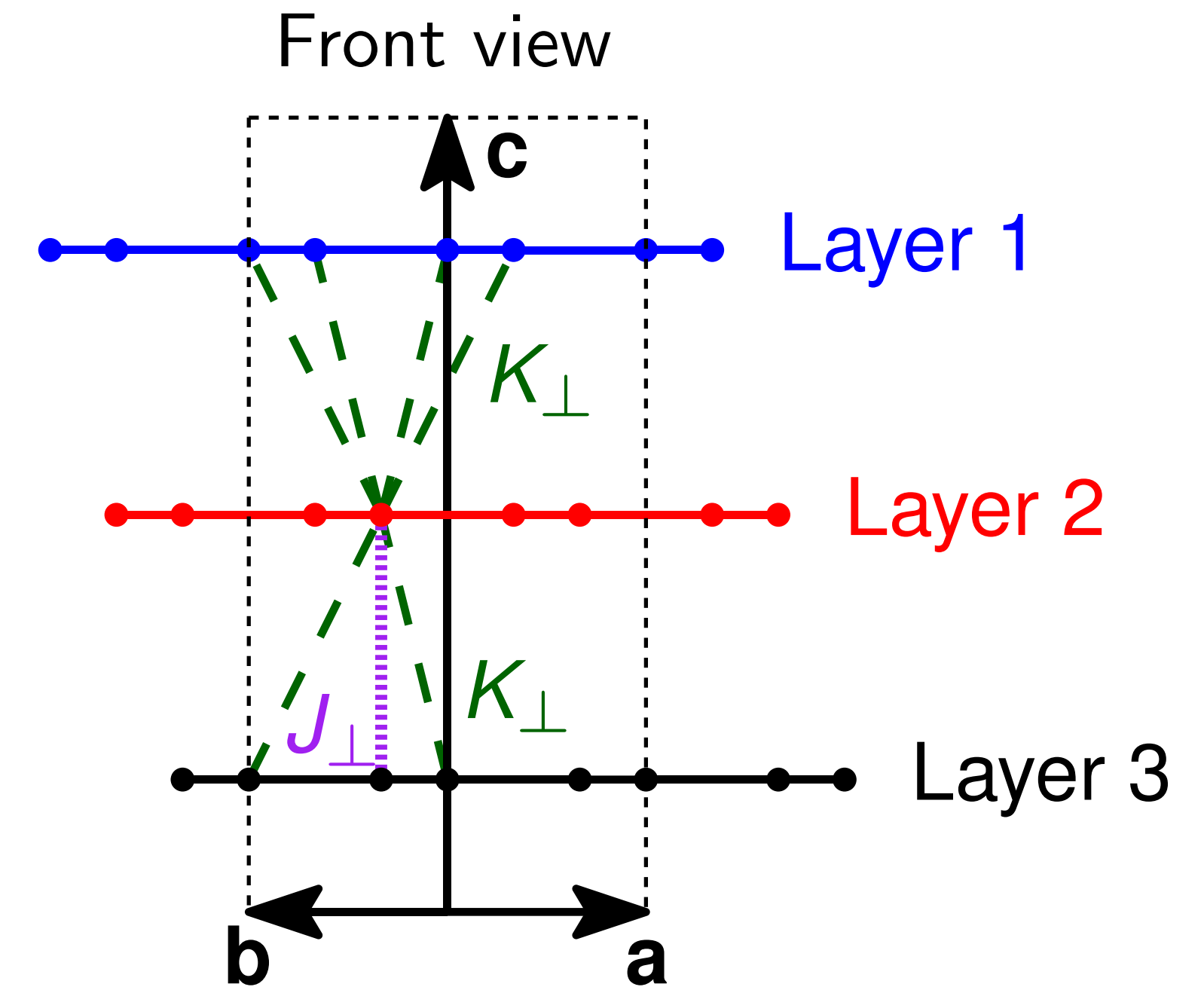
$$\mathcal{H}_{3D} = \mathcal{H}_{2D} + J_{\perp} \sum_{\langle ni, mi \rangle} \vec{S}_{n,i} \cdot \vec{S}_{m,i} + K_{\perp} \underbrace{\sum_{\langle\langle ni, mj \rangle\rangle_{\gamma}} S_{n,i}^{\gamma} S_{m,j}^{\gamma}}_{\text{Depends on absolute spin orientation}}$$



# Minimal 3D spin model

Hamiltonian:

$$\mathcal{H}_{3D} = \mathcal{H}_{2D} + J_{\perp} \sum_{\langle ni, mi \rangle} \vec{S}_{n,i} \cdot \vec{S}_{m,i} + K_{\perp} \underbrace{\sum_{\langle\langle ni, mj \rangle\rangle_{\gamma}} S_{n,i}^{\gamma} S_{m,j}^{\gamma}}_{\text{Depends on absolute spin orientation}}$$

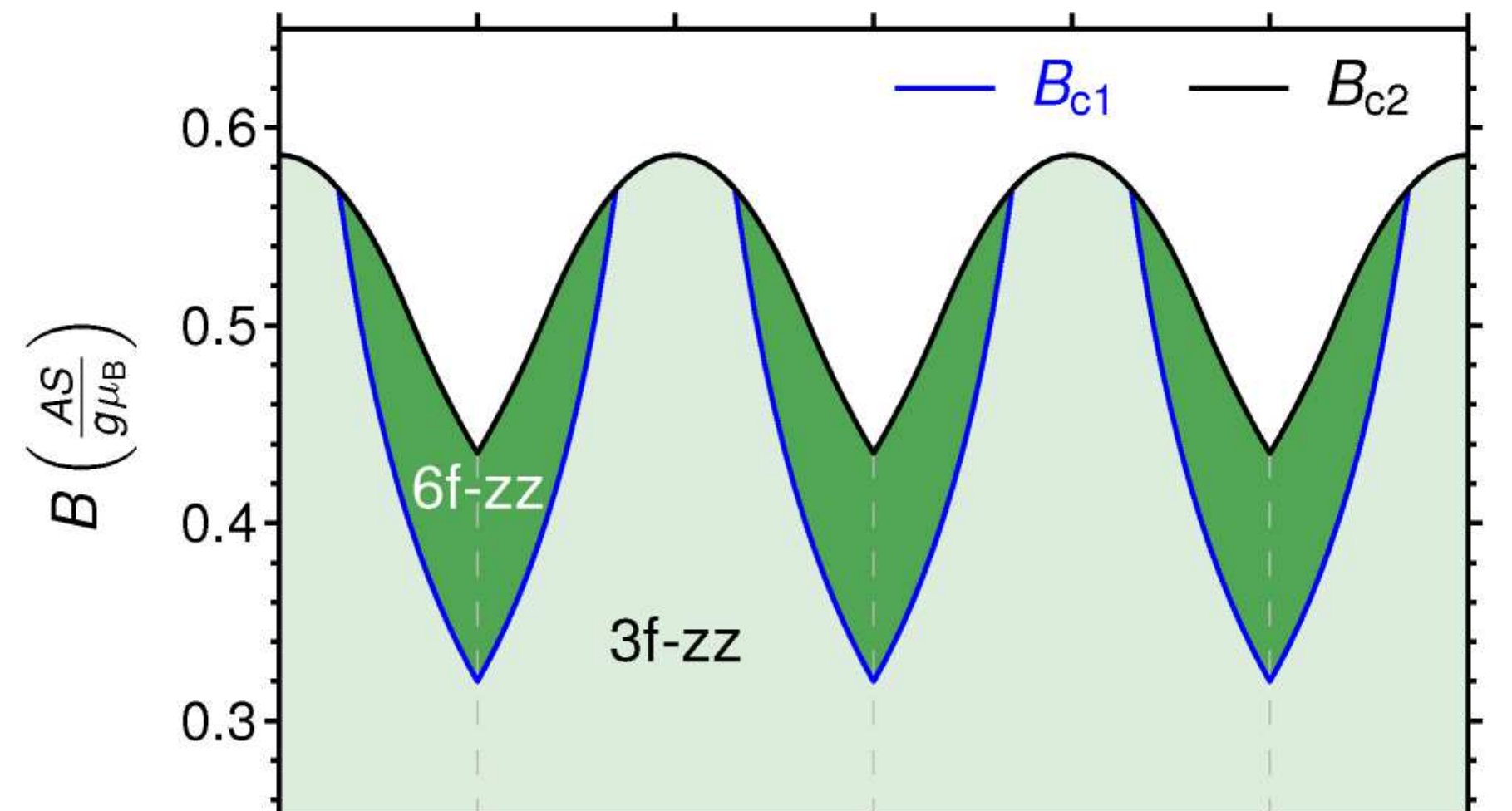


... for  $J_{\perp} > 0$  and  $K_{\perp} < 0$

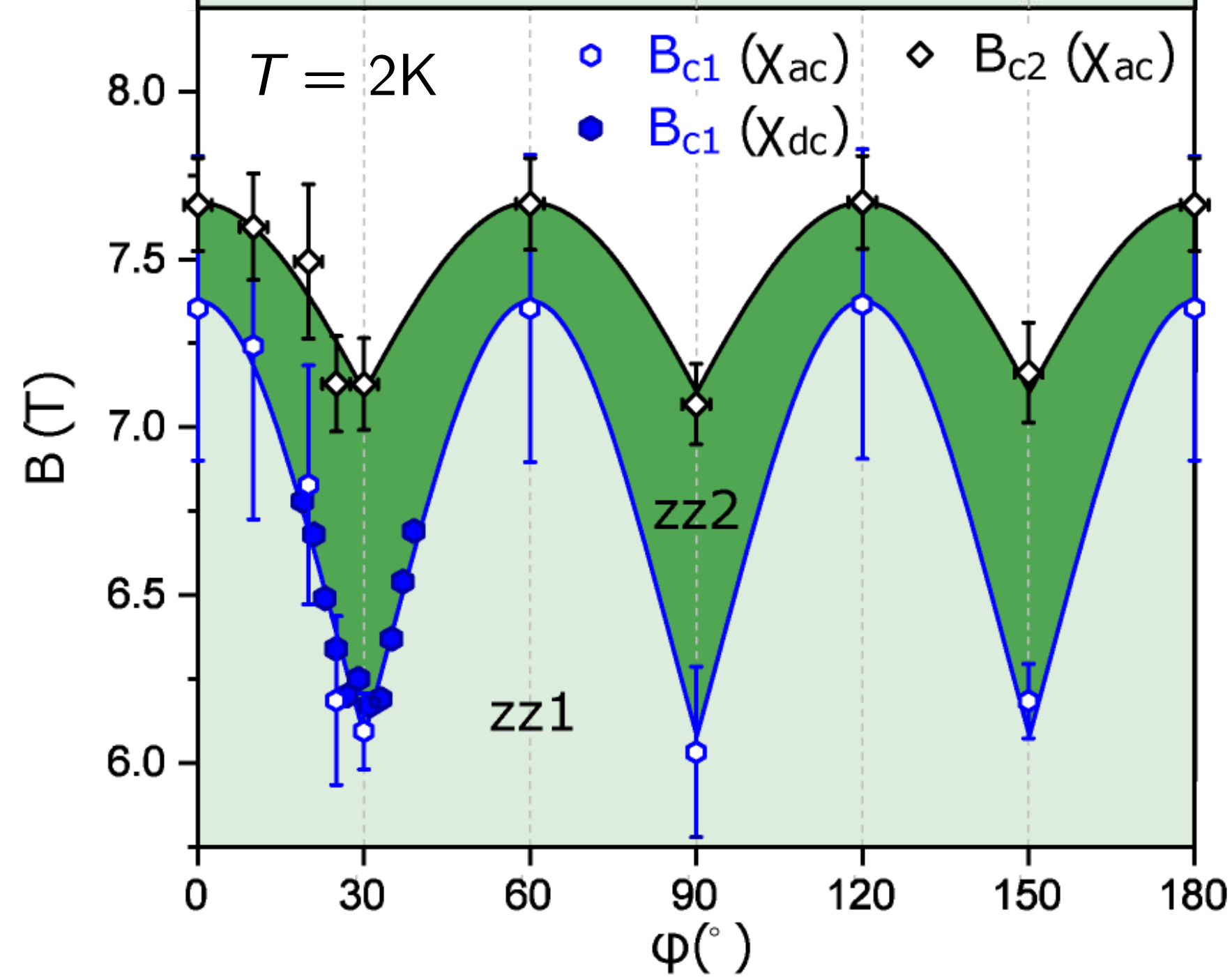
# Comparison model vs. experiment

Field-angle phase diagram

Interlayer model



Experiment  $\alpha$ -RuCl<sub>3</sub>



$$K_1 < 0$$

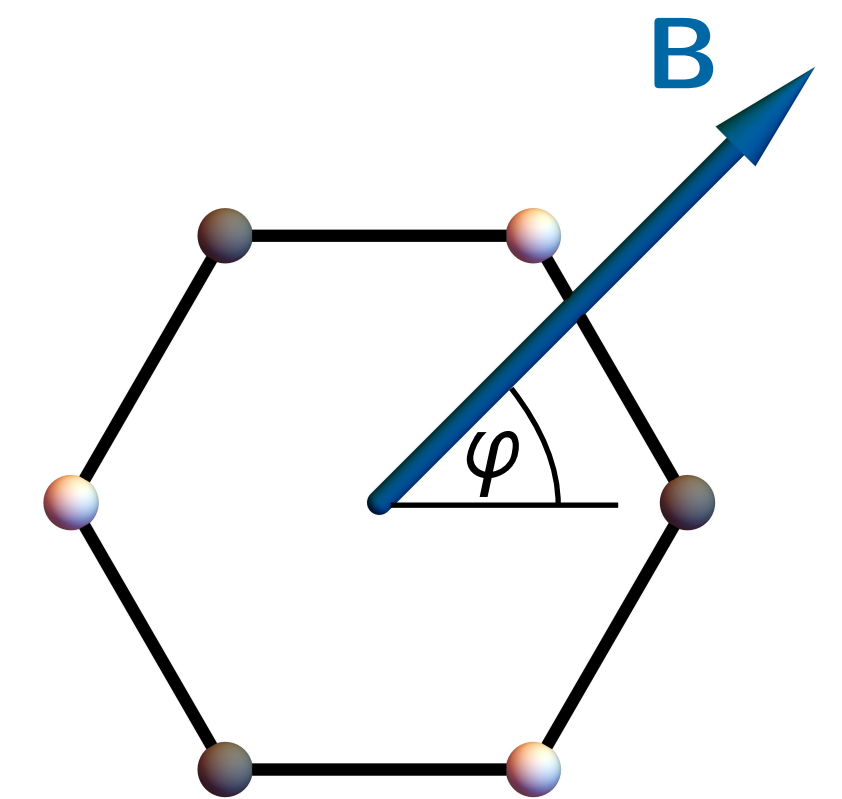
$$\Gamma_1/|K_1| = 0.5$$

$$J_1/|K_1| = -0.1$$

$$J_3/|K_1| = 0.1$$

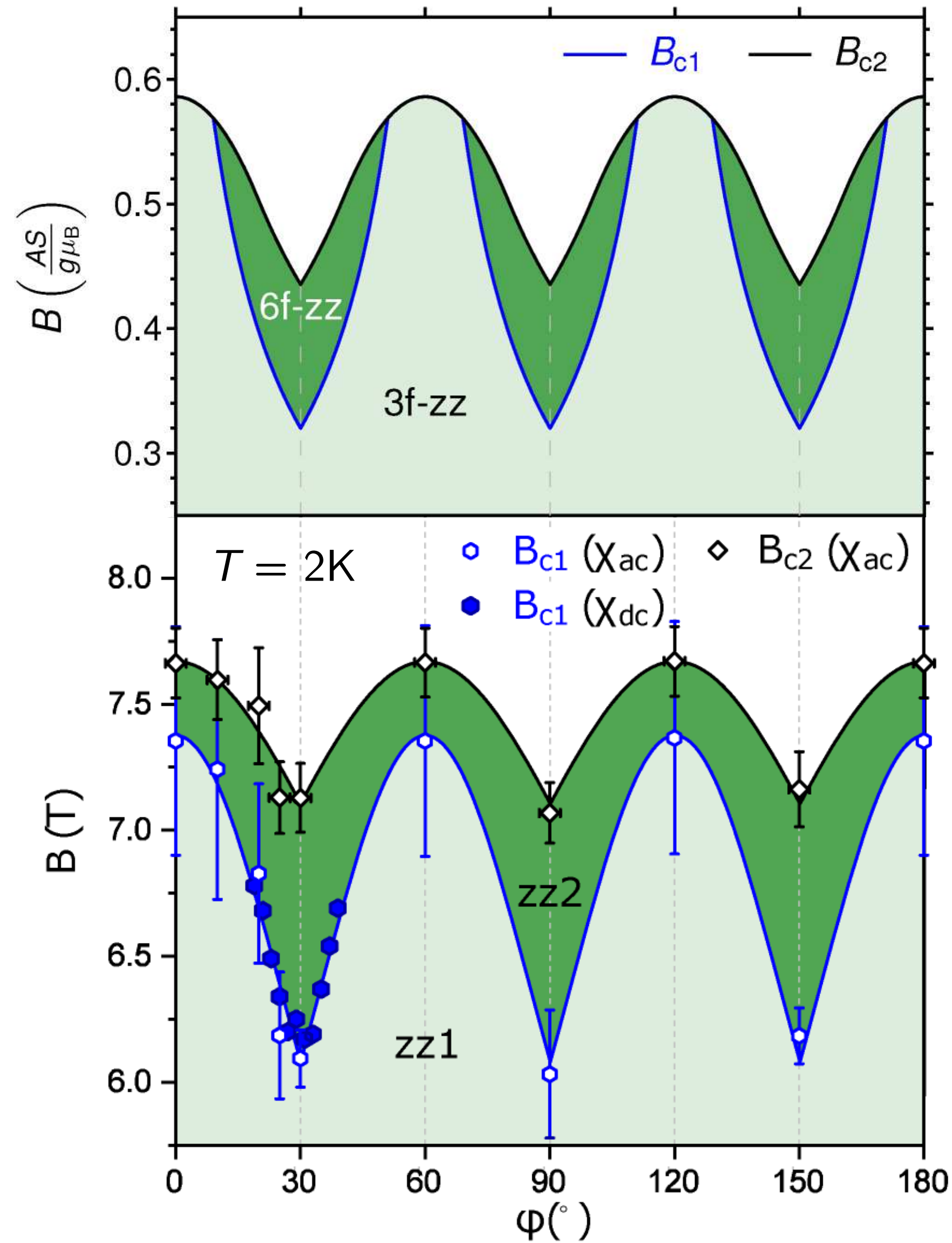
$$J_\perp > 0$$

$$K_\perp/J_\perp = -1.14$$

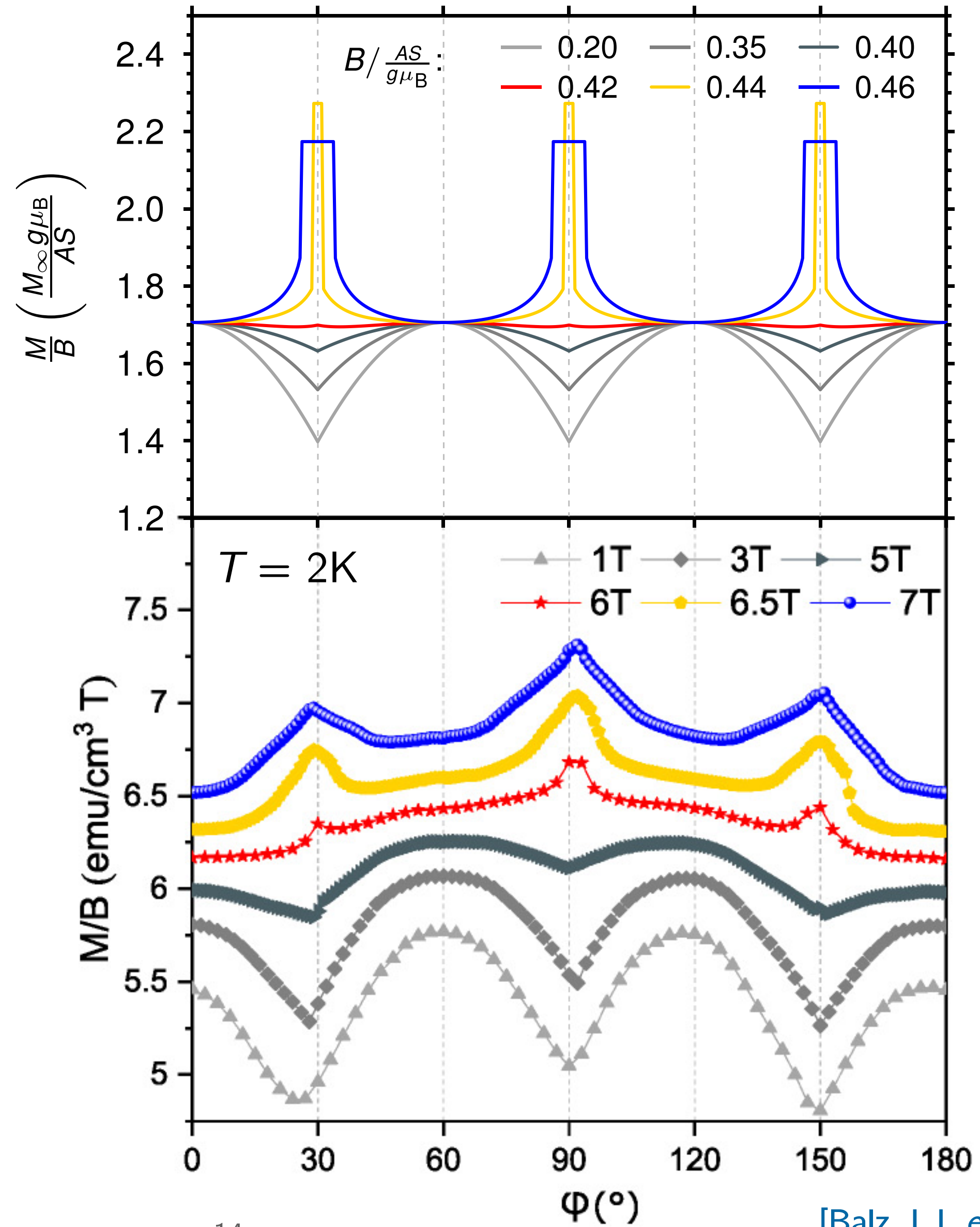


# Comparison model vs. experiment

Field-angle phase diagram



Magnetization vs. field angle



$$K_1 < 0$$

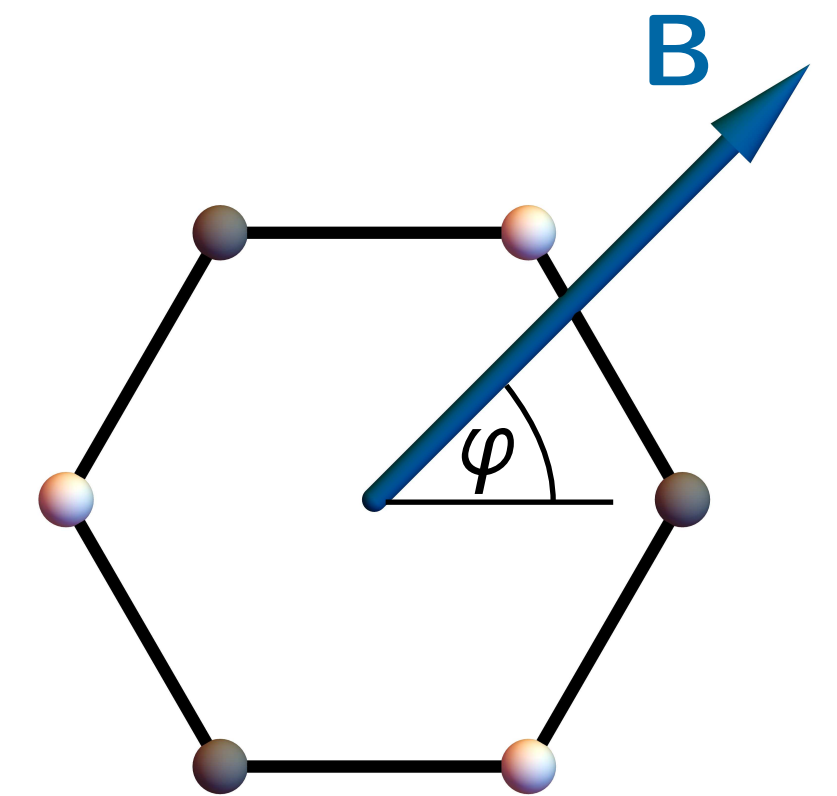
$$\Gamma_1/|K_1| = 0.5$$

$$J_1/|K_1| = -0.1$$

$$J_3/|K_1| = 0.1$$

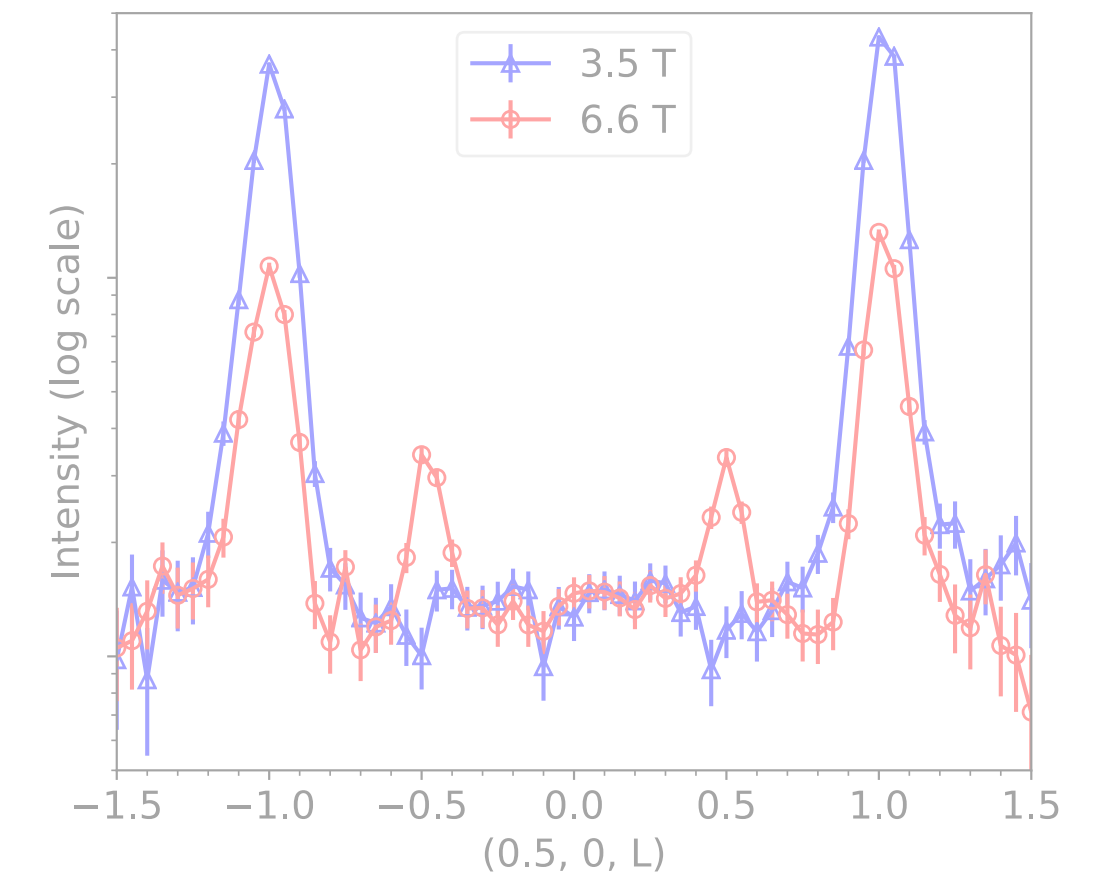
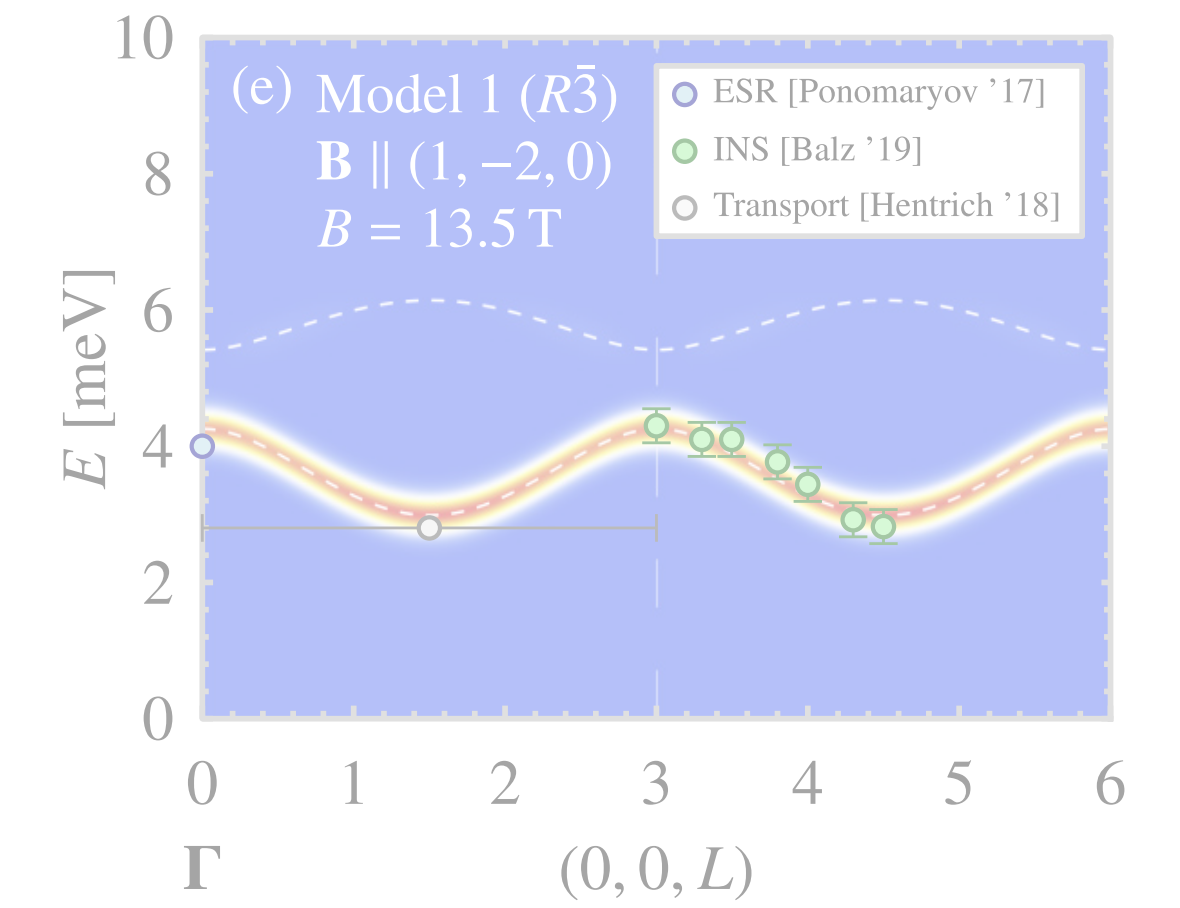
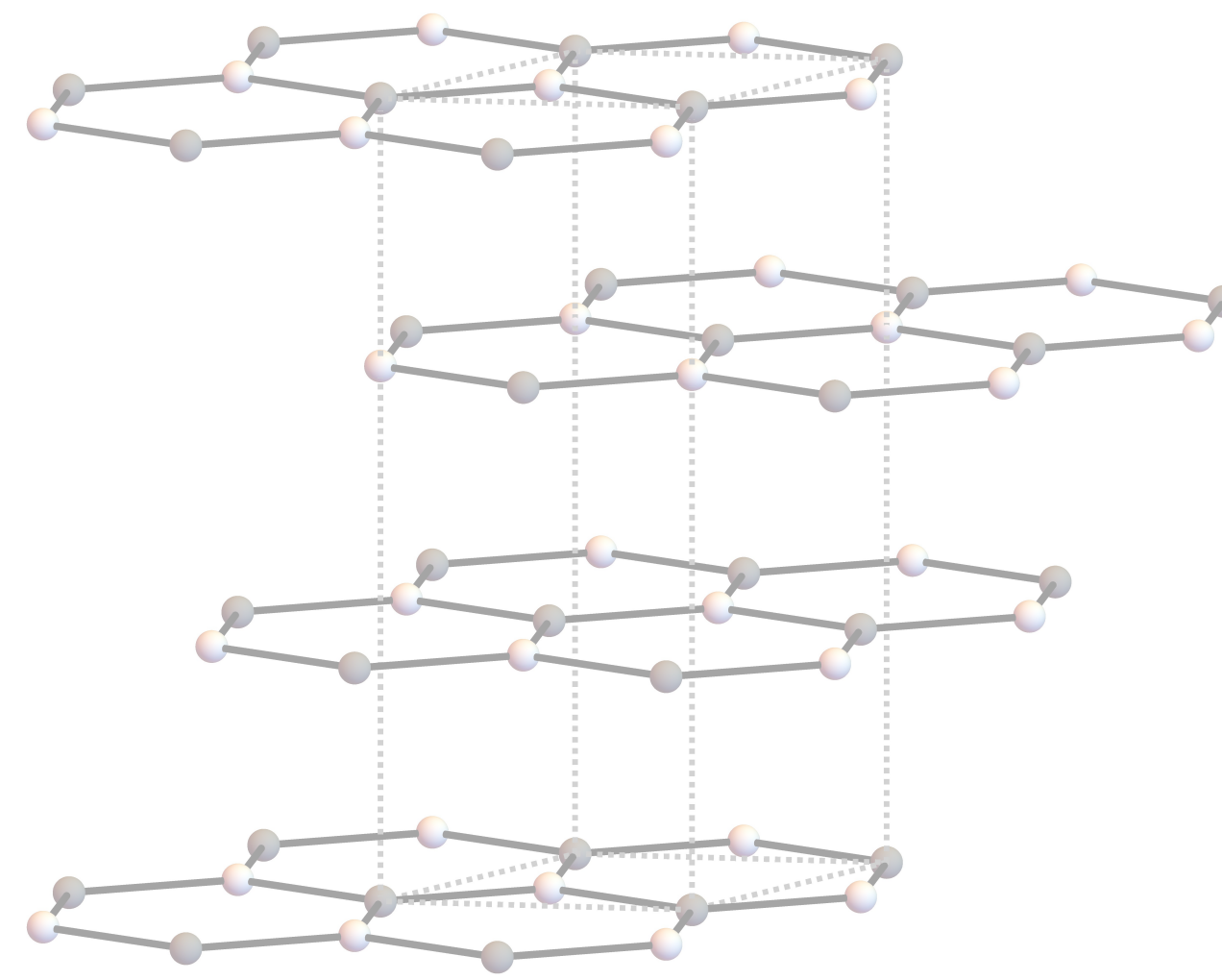
$$J_\perp > 0$$

$$K_\perp/J_\perp = -1.14$$



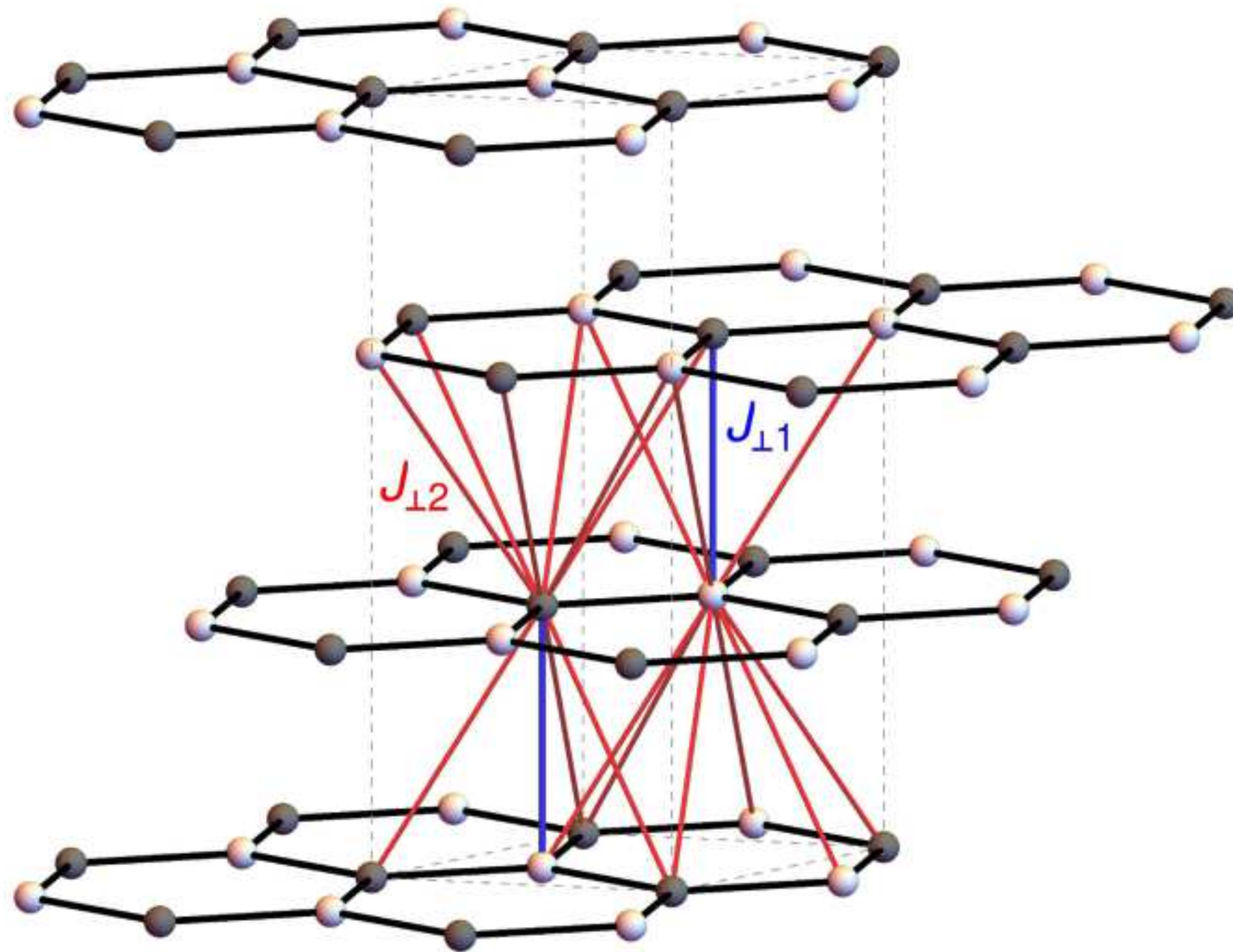
# Outline

1. Introduction: Modeling  $\alpha$ -RuCl<sub>3</sub>
2. Evidence #1: Excitation spectra
3. Evidence #2: Field-induced intermediate order
4. Conclusions



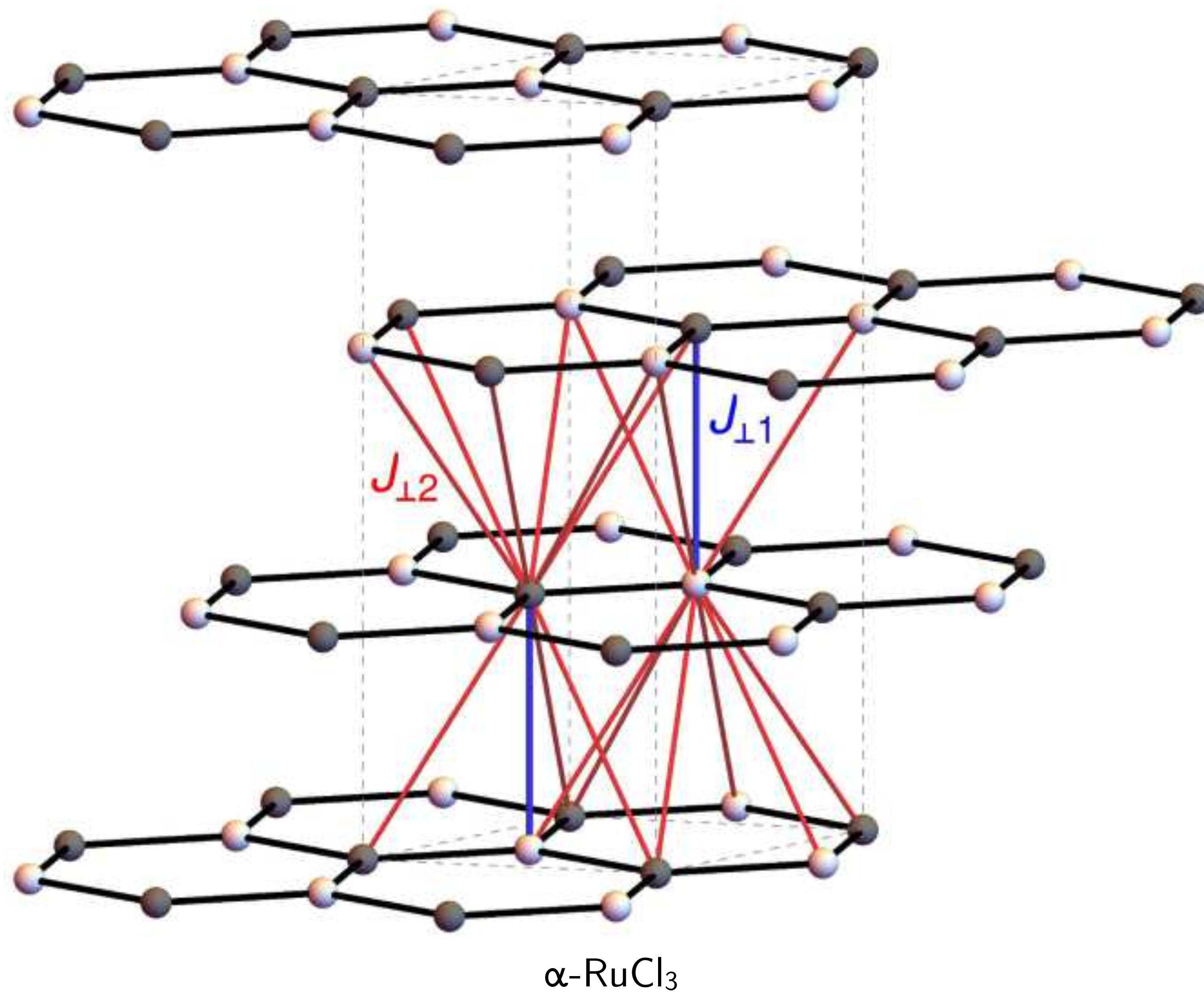


# Conclusions

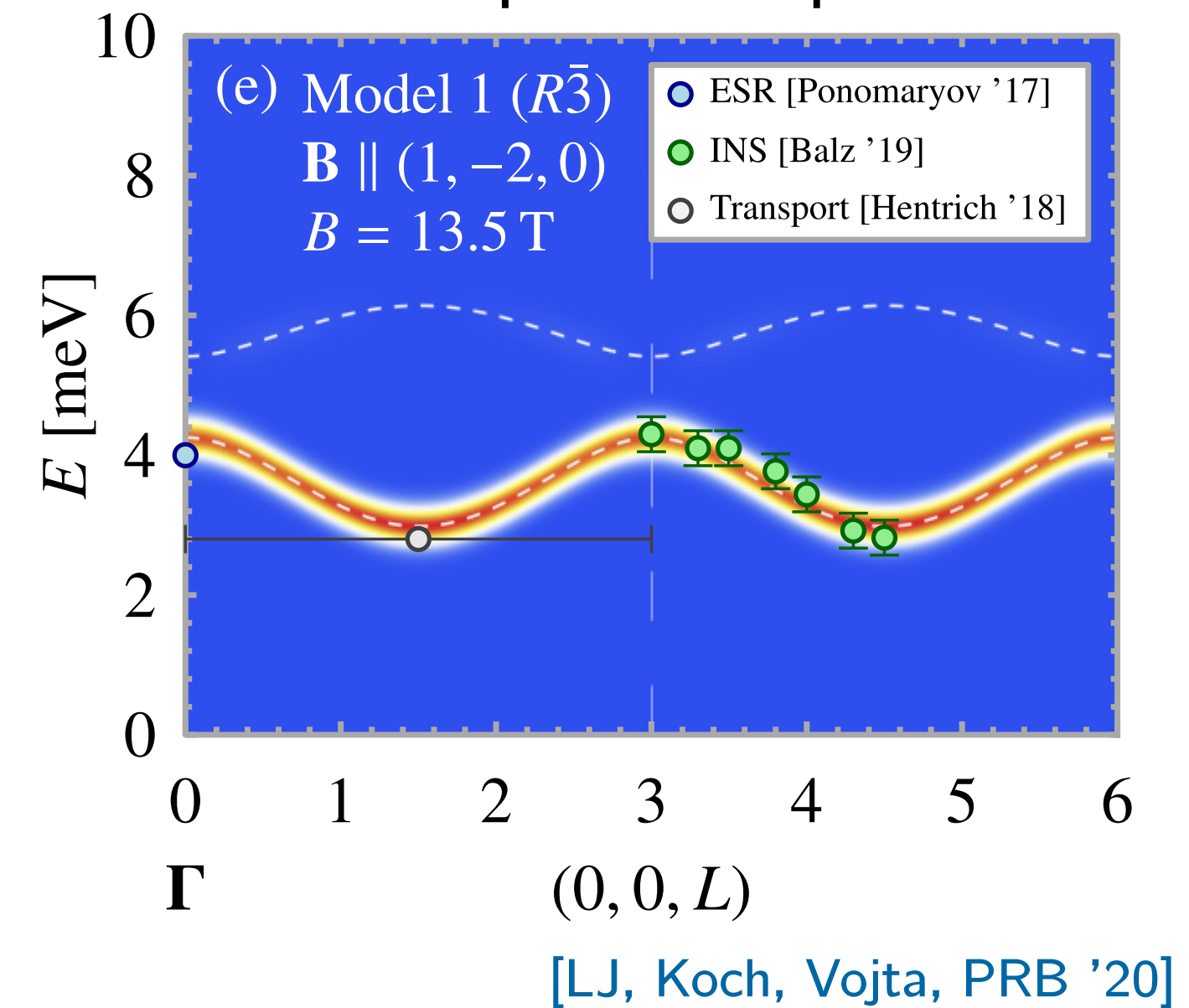


$\alpha\text{-RuCl}_3$

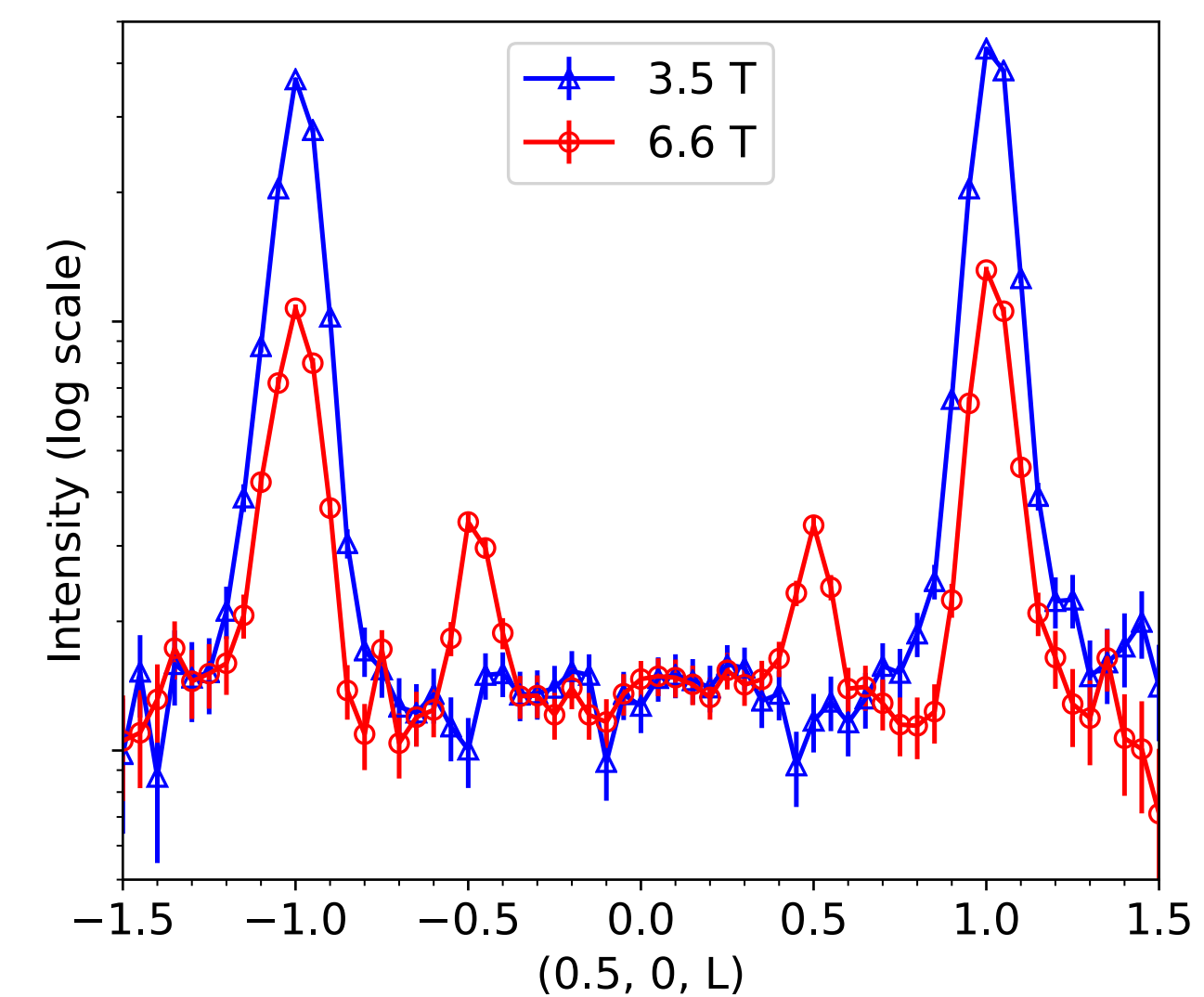
# Conclusions



## Out-of-plane dispersion



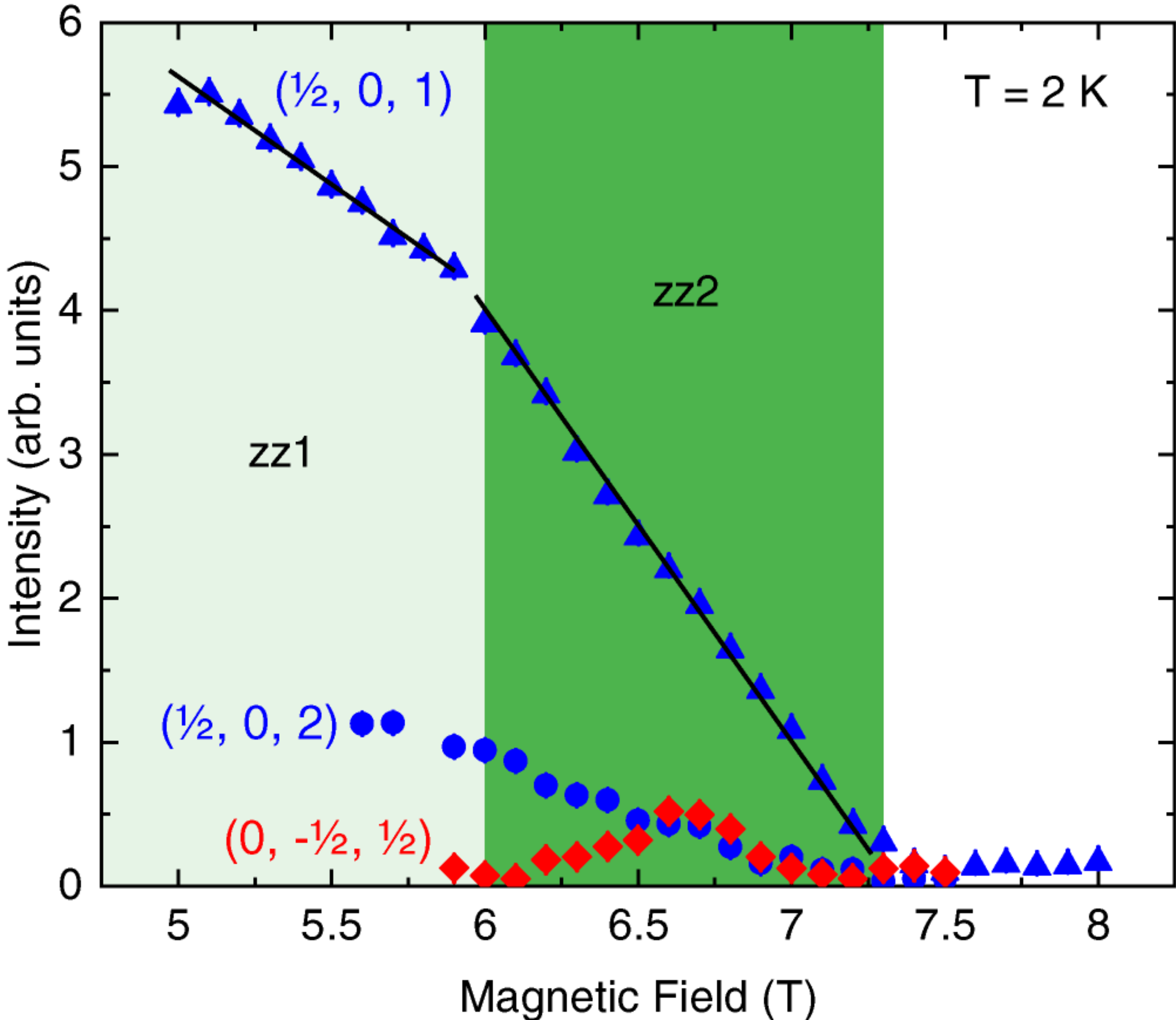
## Zigzag-to-zigzag transition



[Balz, LJ, *et al.*, PRB '21 (Editors' Suggestion)]



# Bragg peak intensity vs. field



[Balz, LJ, et al., PRB '21 (Editors' Suggestion)]

# Quantum oscillations in low- $T$ thermal conductivity

