Investigating Kitaev physics in Co honeycomb-lattice oxides

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Spin wave calculations:
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Neutron/X-ray scattering:
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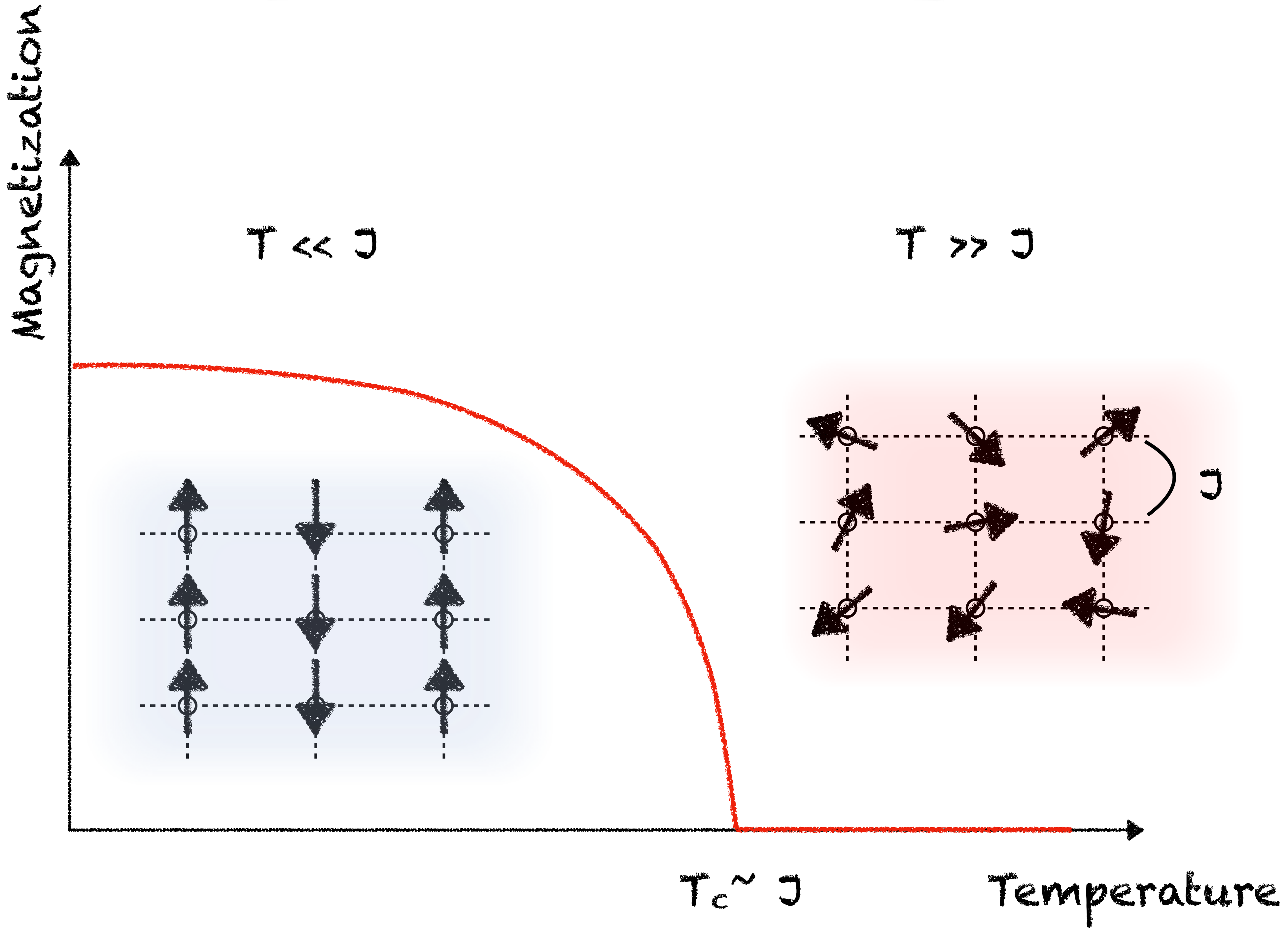
Jose Rodriguez, William Ratcliff (NIST)
Monica Jiménez-Ruiz (ILL)
Françoise Damay (Laboratoire Léon Brillouin)
Victor Balédent (Laboratoire de Physique des Solides)

Beyond conventional magnetism

Conventional magnetic order
Landau's formalism



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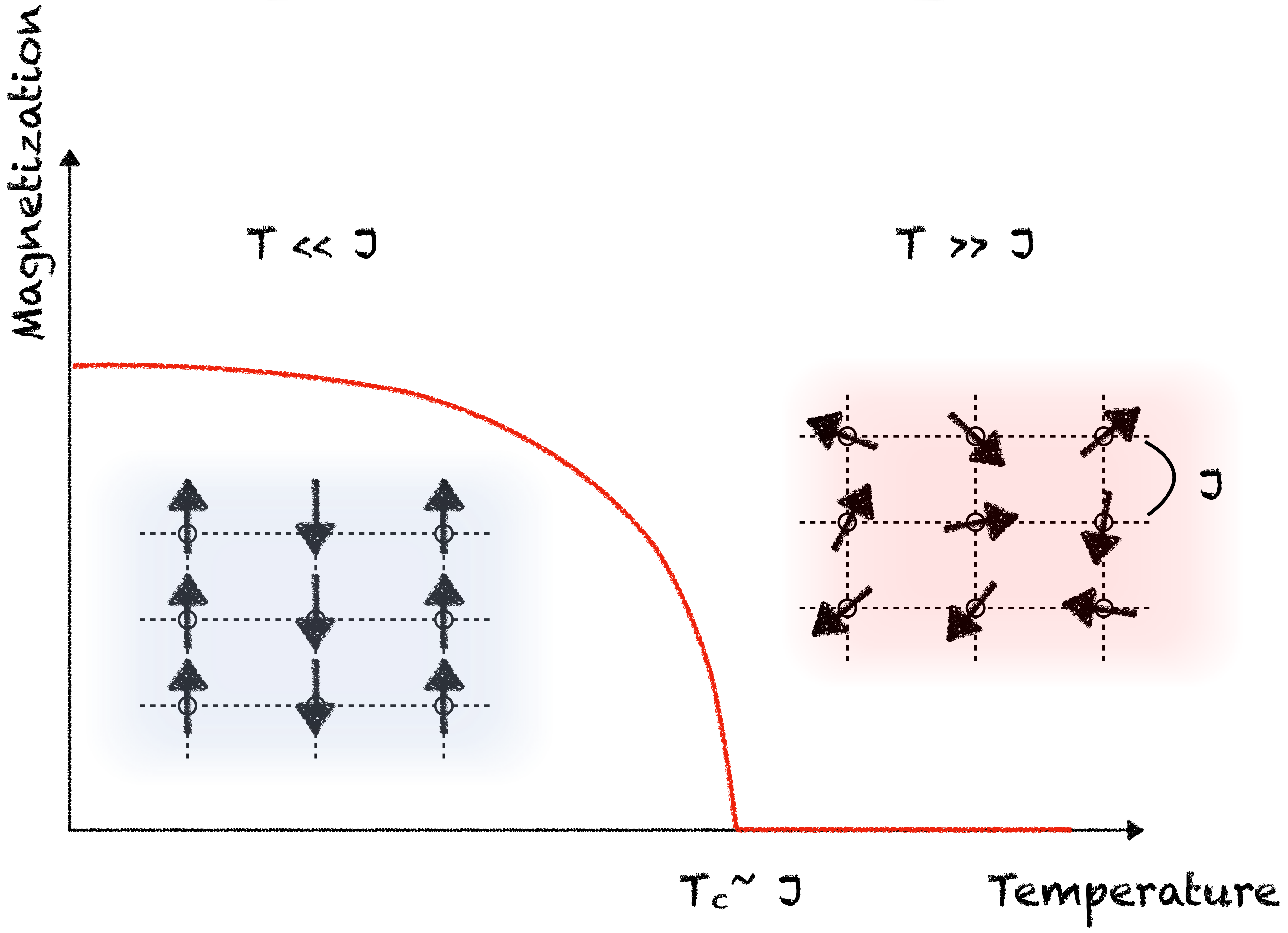
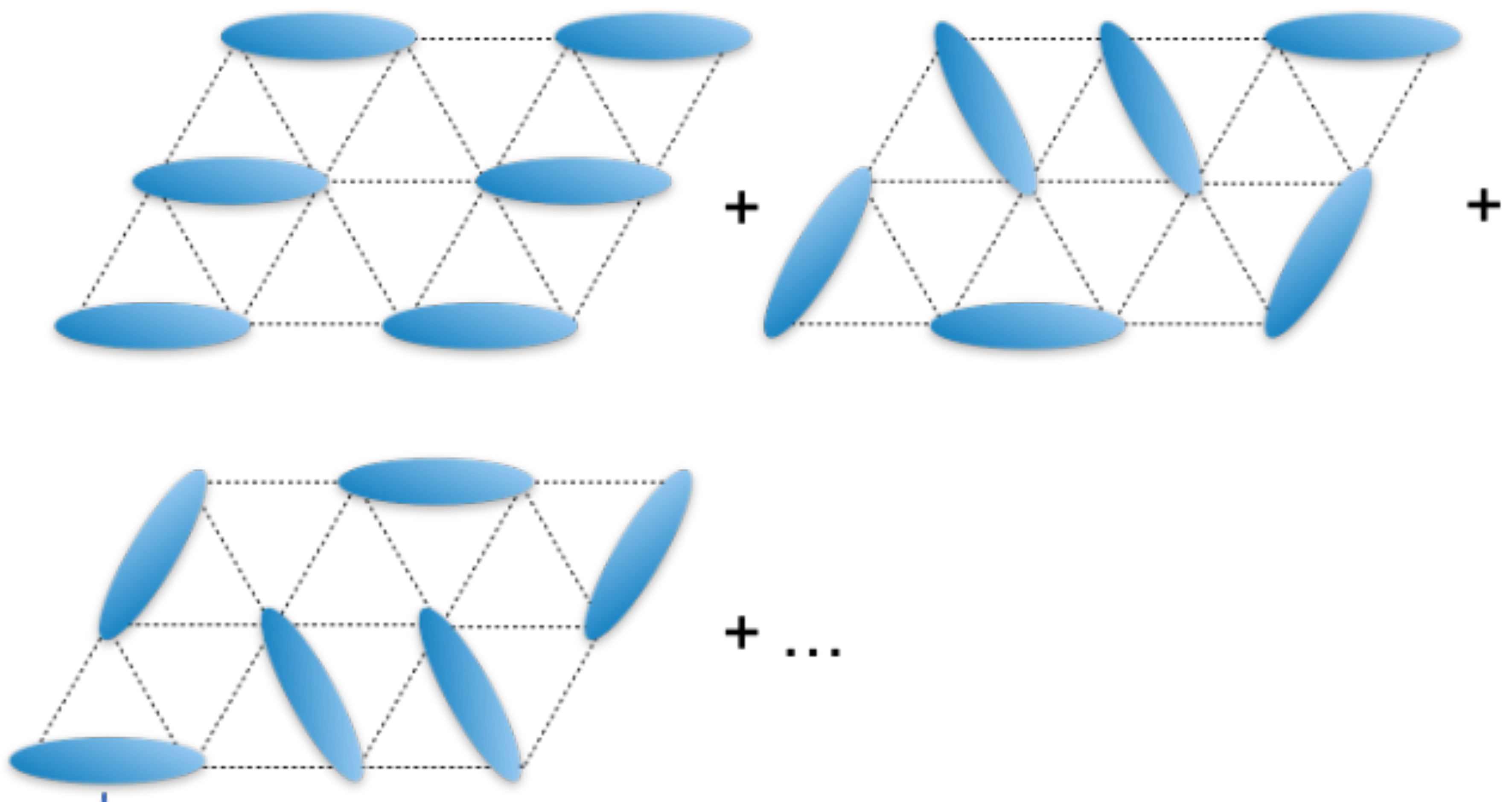
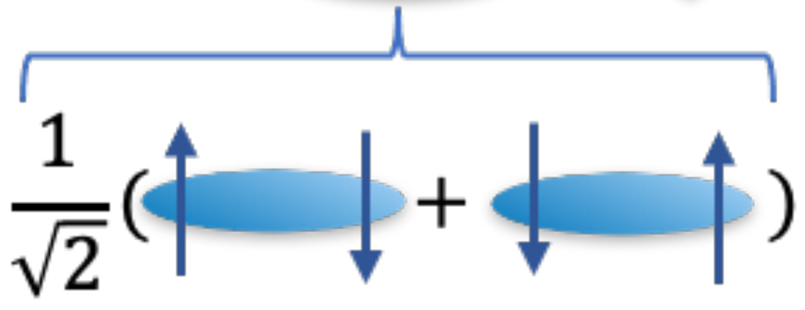


Quantum spin liquids
Beyond Landau's model





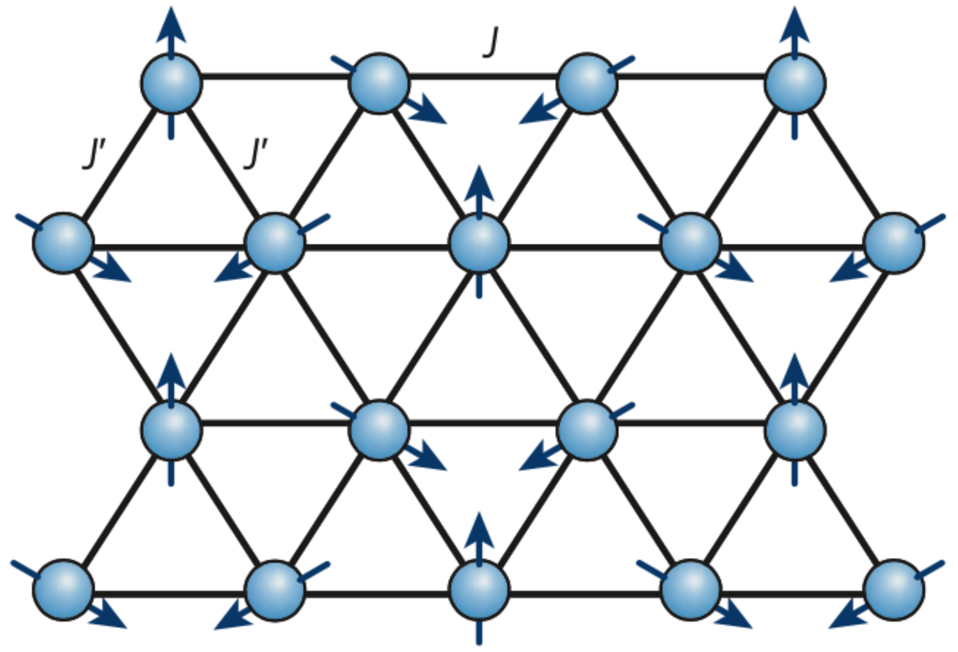
X No long-range order down to T=0 K
✓ Spin entanglement
✓ Emergent fractional excitations

Why are quantum spin liquids interesting?

It's a new state of matter:

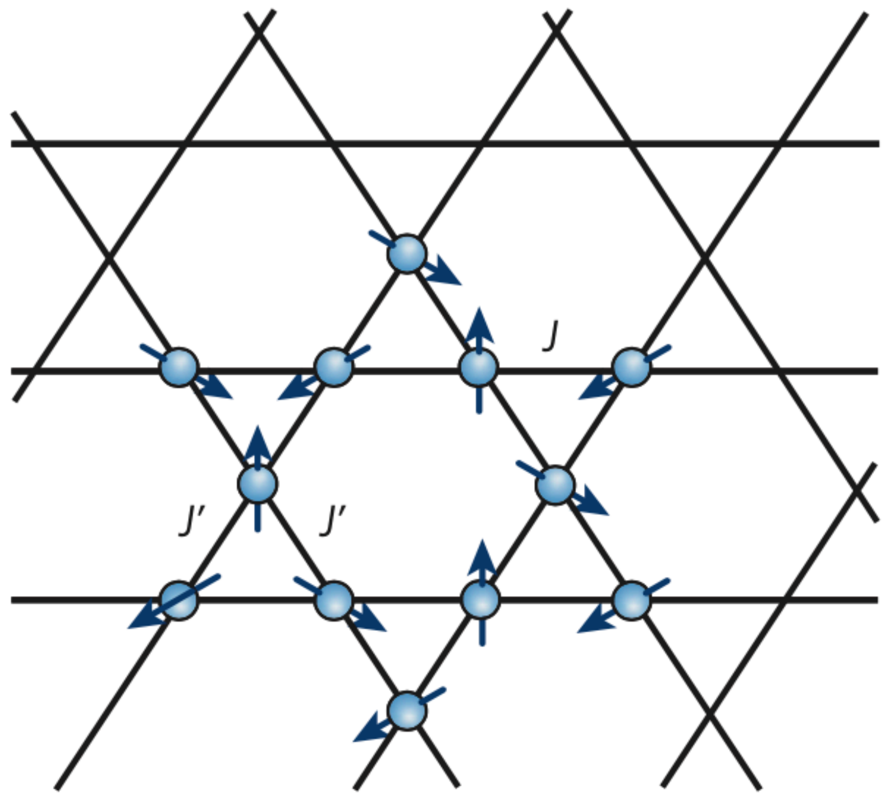
- Collective behaviours and emergent phenomena
  - *Electromagnetism*
  - *Quantum electrodynamics*
  - *High energy physics…*
- How to classify these new states? Topological orders…
- Experimental challenge: what are the signatures?
- Very few experimental realizations in dimension d > 1

Achieving quantum spin liquids: geometrical frustration



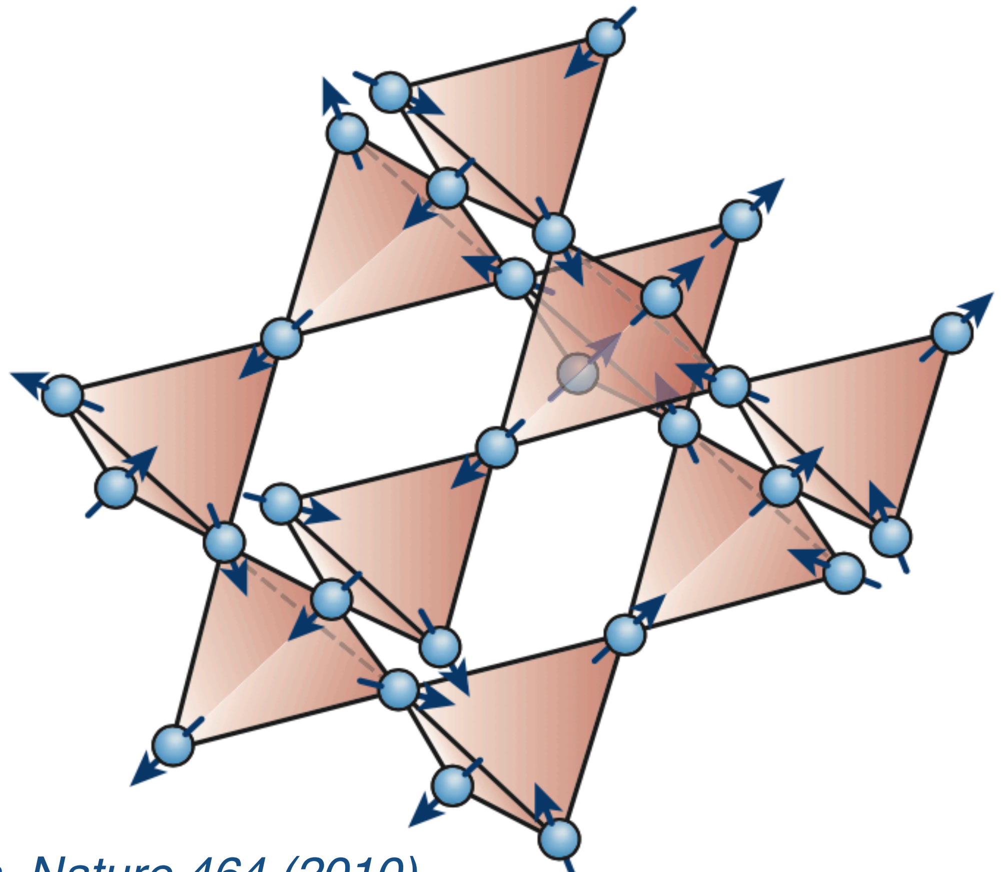
Triangular lattice

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$
EtMe$_3$Sb[Pd(dmit)$_2$]$_2$
Y. Shimizu et al., Phys. Rev. Lett. 91, 107001 (2003)
T. Itou et al., Nature Physics 6, 673 (2010)



Kagome lattice

Herbertsmithite ZnCu$_3$(OH)$_6$Cl$_2$
T.-H. Han at al., Nature 492,504 (2012)
M. P. Shores et al., J. Am. Chem. Soc. 127, 13462 (2005)



Pyrochlore lattice

Pyrochlore Dy$_2$Ti$_2$O$_7$, Ho$_2$Ti$_2$O$_7$...
M. P. J. Gingras and P. A. McClarty, Rep. Prog. Phys. 77, 056501 (2014)

Hyperkagome Na$_4$Ir$_3$O$_8$
Y. Okamoto et al., Phys. Rev. Lett. 99, 137207 (2007)

L. Balents, Nature 464 (2010)

A promising route: The Kitaev model on honeycomb lattice

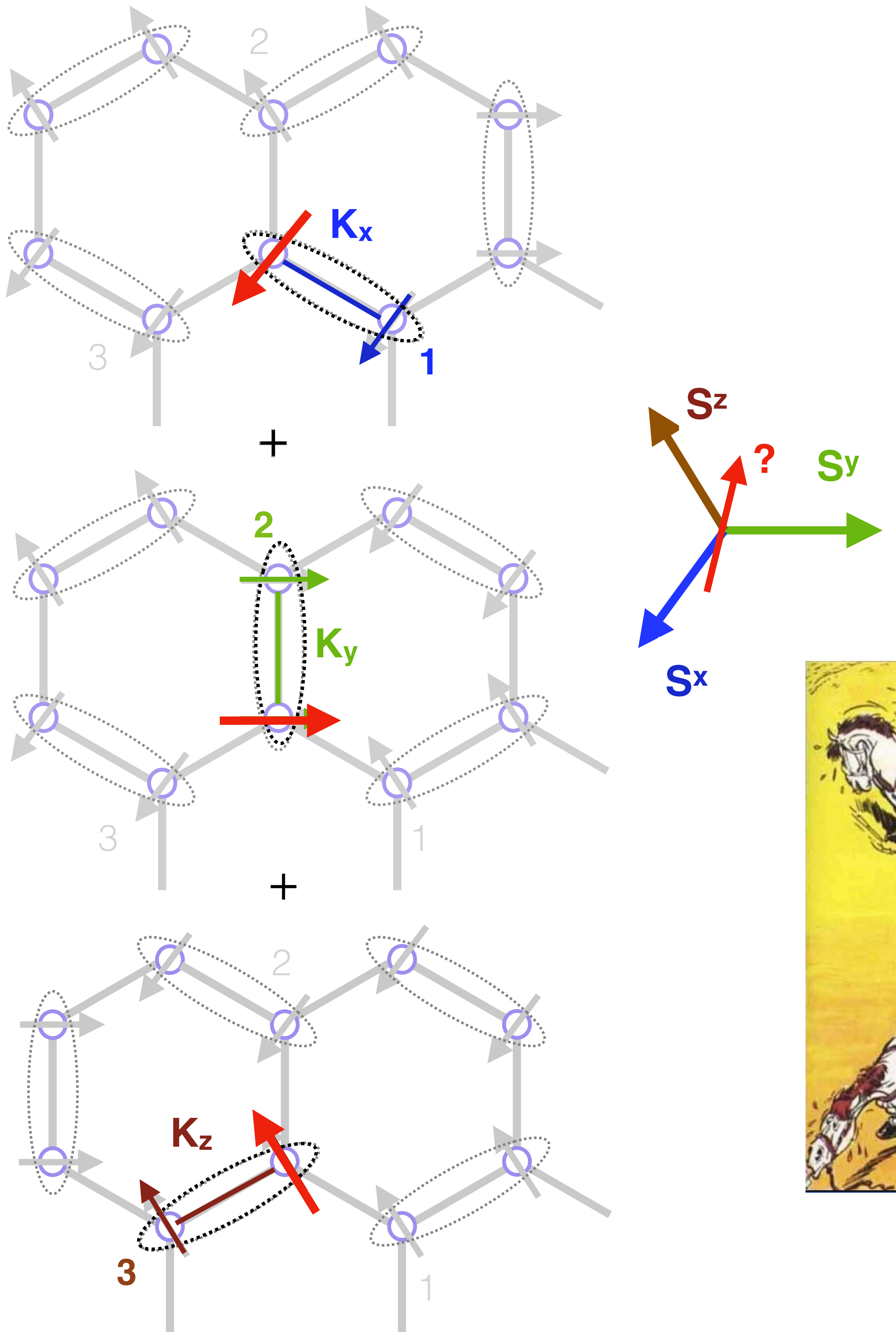
A. Kitaev, Annals of Physics 321 (2006)

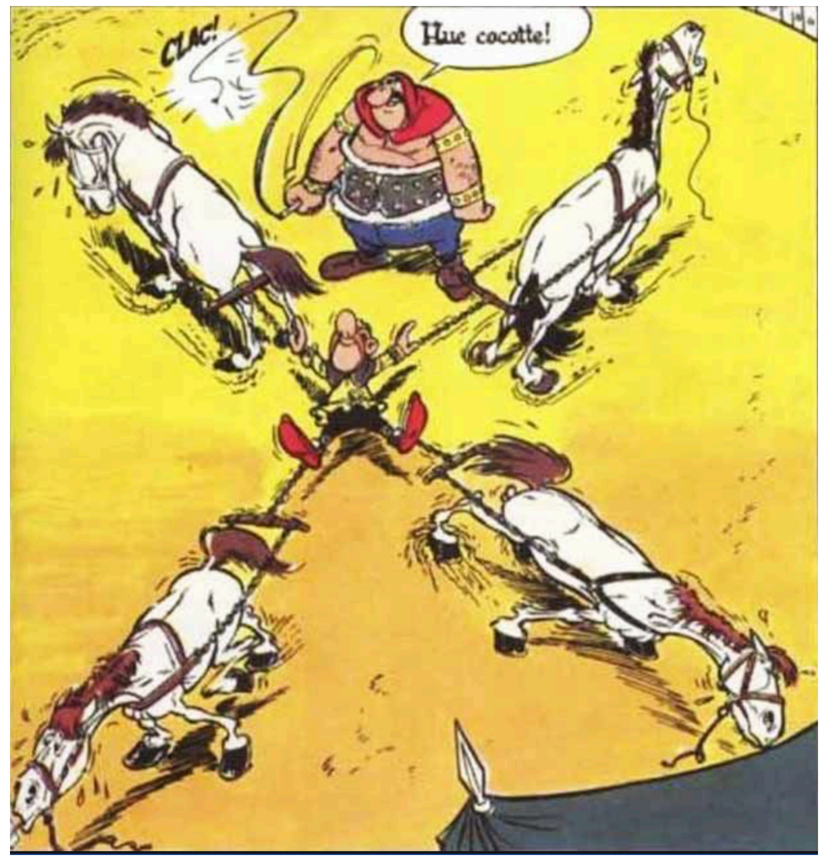
- S=1/2 honeycomb lattice
- **Exact ground state : quantum spin liquid**
- Emergent fractional excitations: Majorana modes and flux excitations
- Rich phase diagram

Anisotropic bond-dependent interactions

$$H_{Kitaev} = -\sum_{i,\alpha=1,2,3} (K_x S_i^x S_\alpha^x + K_y S_i^y S_\alpha^y + K_z S_i^z S_\alpha^z)$$

An exact model





The Kitaev model

A. Kitaev, Annals of Physics 321 (2006)

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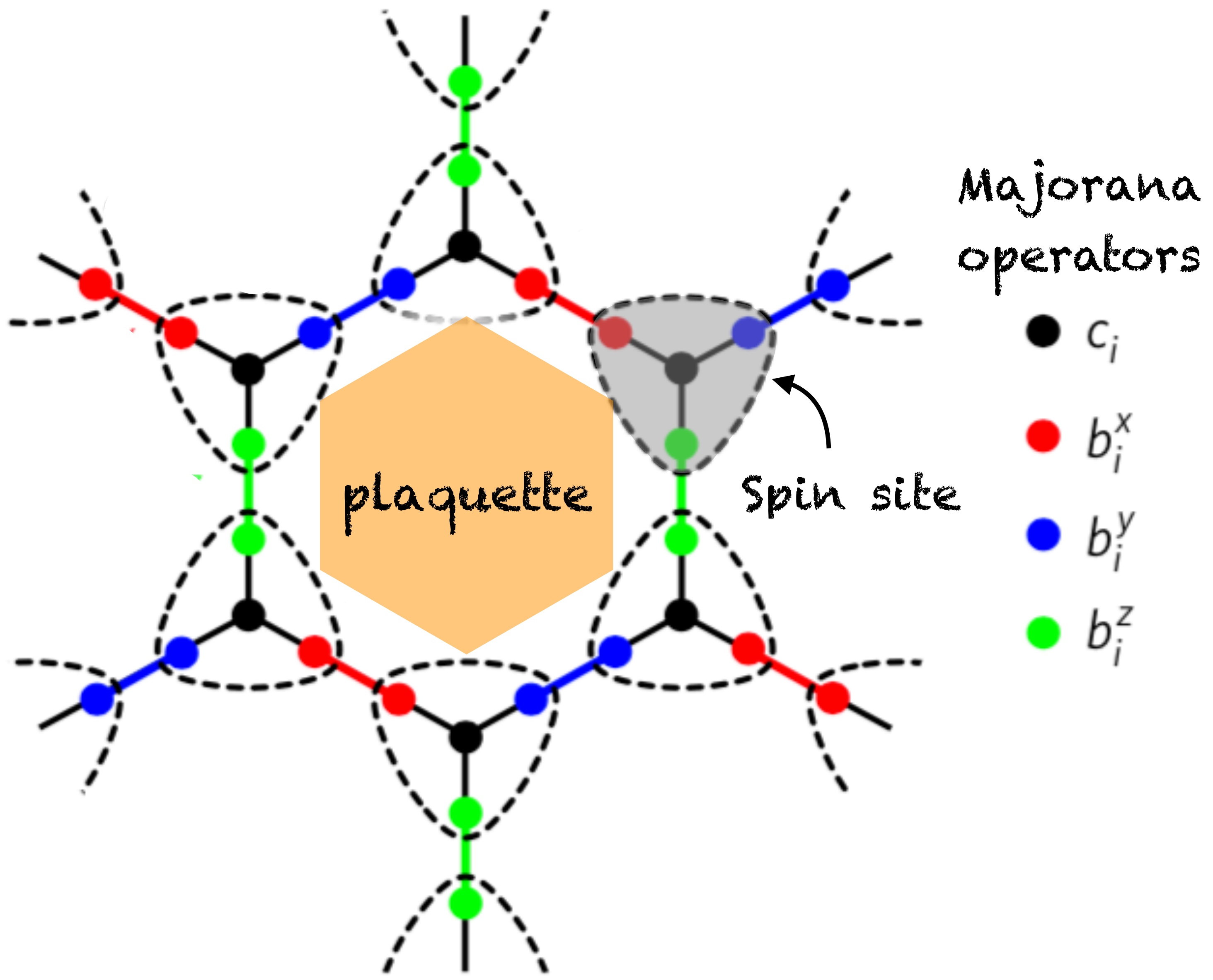
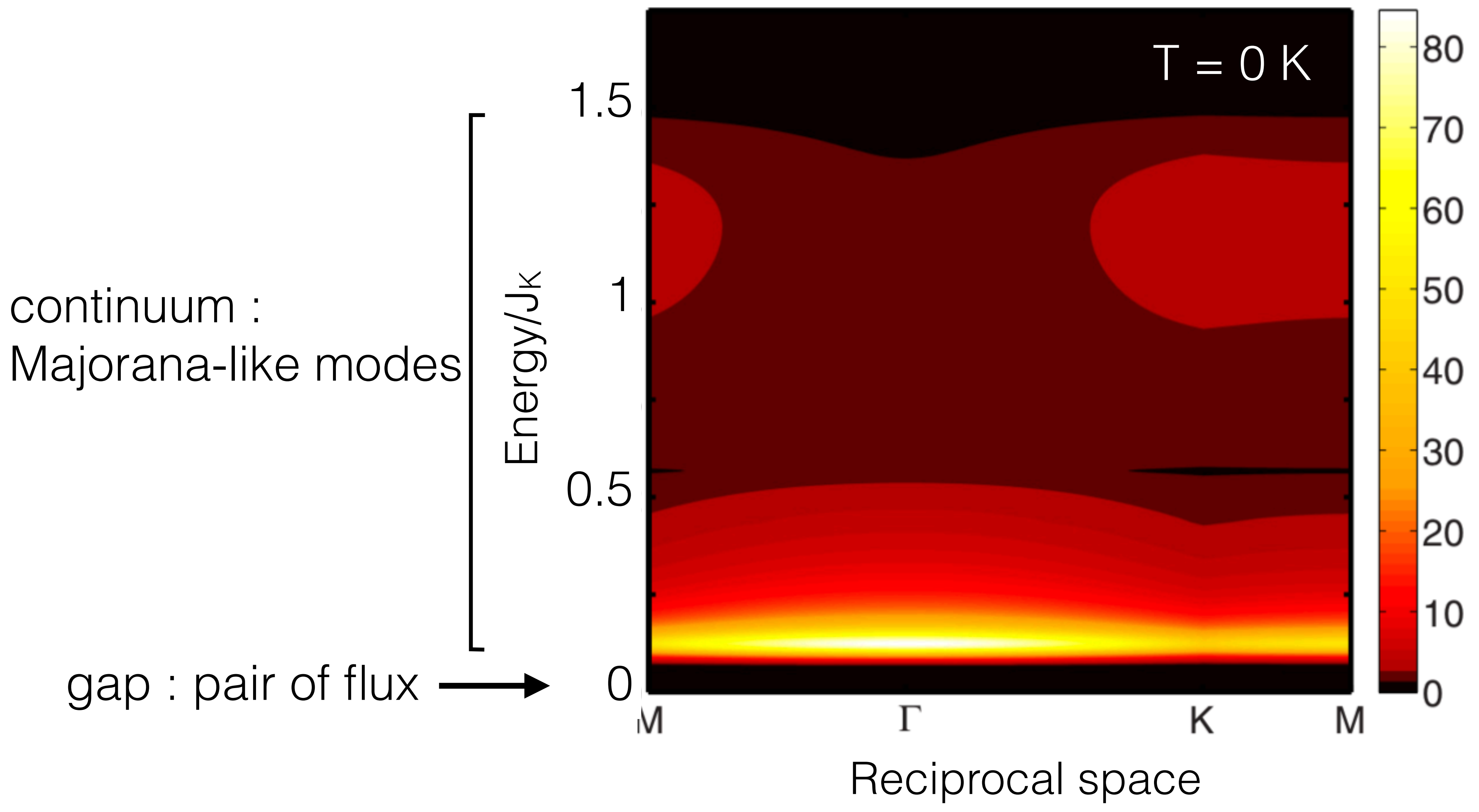


S.-H. Do et al., Nature Physics 13, 1079 (2017)

Theoretical prediction for the dynamical structure factor in neutron scattering



J. Knolle et al., Phys. Rev. B (2015)

The Kitaev model

A. Kitaev, Annals of Physics 321 (2006)

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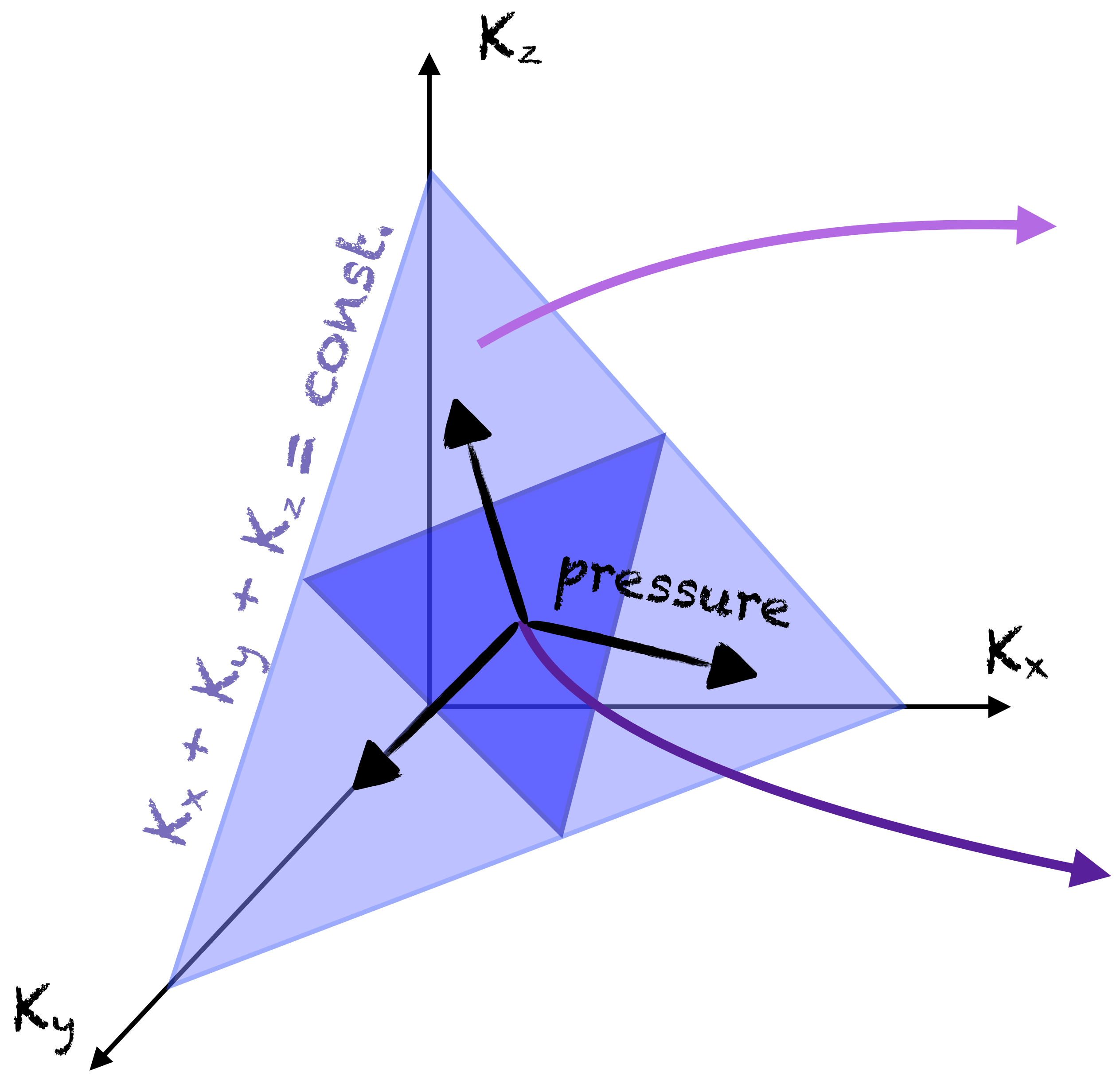
- Exact ground state : quantum spin liquid
- Emergent fractional excitations: Majorana modes and flux excitations
- **Rich phase diagram**

Tuning the ground state through external parameters (magnetic field, pressure)



Gapped spin liquid

Gapless spin liquid

- Becomes topologically non-trivial **under magnetic field** (non-Abelian excitations)

The Kitaev model

An alternative route to achieve quantum spin liquids

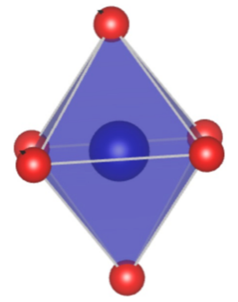
- Exact solution : spin liquid ground state
- Emergence of new fractional excitations with interesting statistical properties
- Possibility to navigate between different QSL regimes with external parameters
- Tools for quantum computing: the Toric code (c.f. *A. Kitaev, Annals of Physics 321 (2006)*)

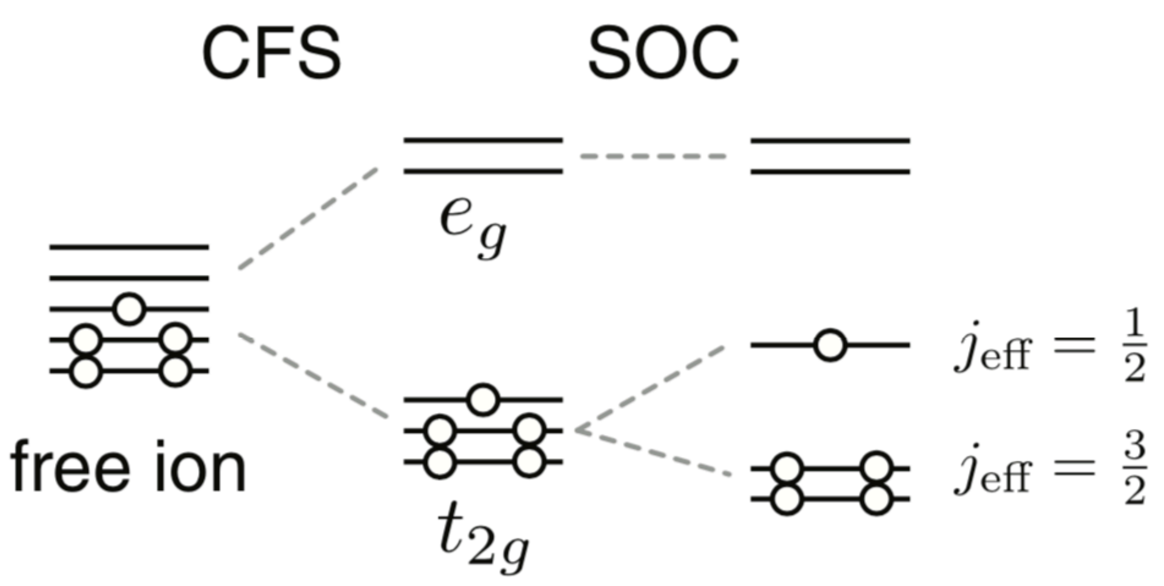
C. Nayak et al., Rev. Mod. Physics 80, 3 (2008)
A. Stern, Nature 464, 7286 (2010)

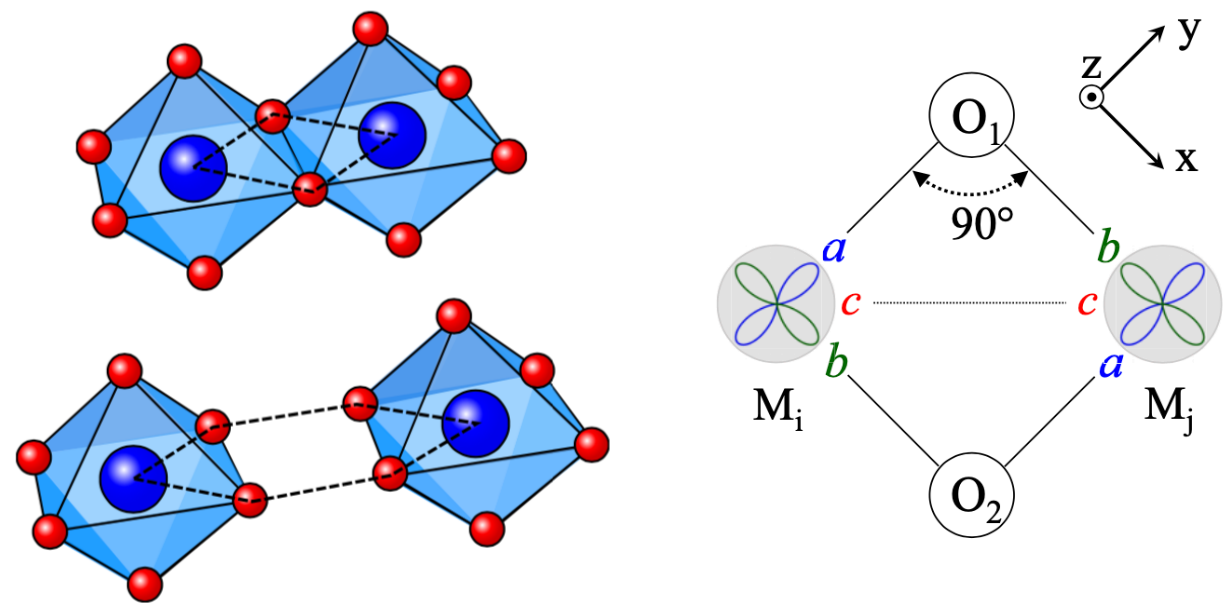
The Kitaev model: experimental realization?

The Jackeli and Khaliullin proposal for experimental realization

G. Jackeli and G. Khaliullin, Phys. Rev. Lett. 102, 017205 (2009)







d^5 metal transition in octahedral environment

Ir^{4+} ($5d^5$), Ru^{3+} ($4d^5$)

+

Spin-orbit coupling

λ~ 400-500 meV for Ir^{4+}

λ~ 100 meV for Ru^{3+}

+

Edge-sharing octahedra

Suppress super-exchange processes

+

Hund's coupling

=

j_{eff} =1/2 pseudospin with anisotropic exchange interactions

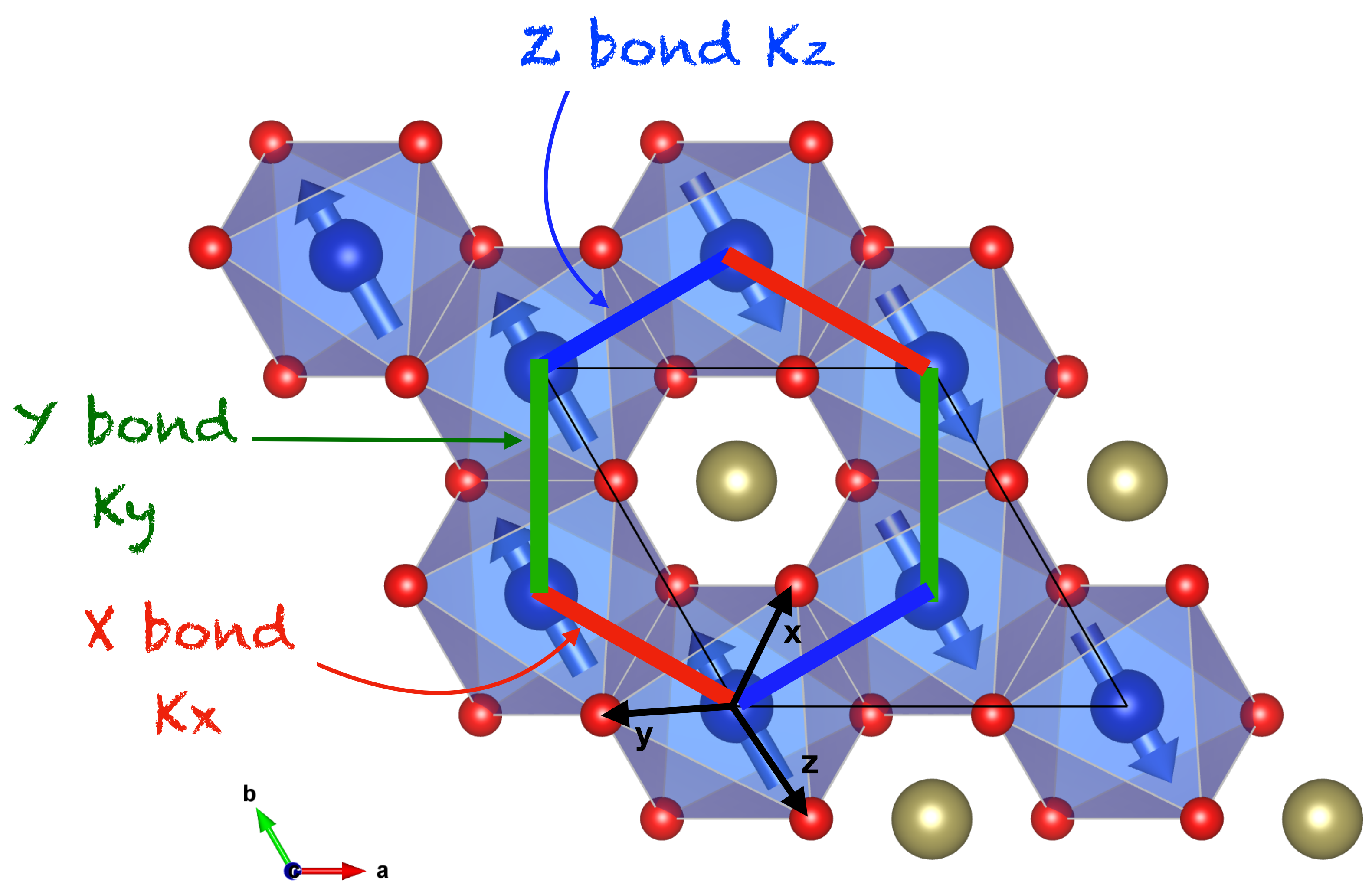
Material candidates:

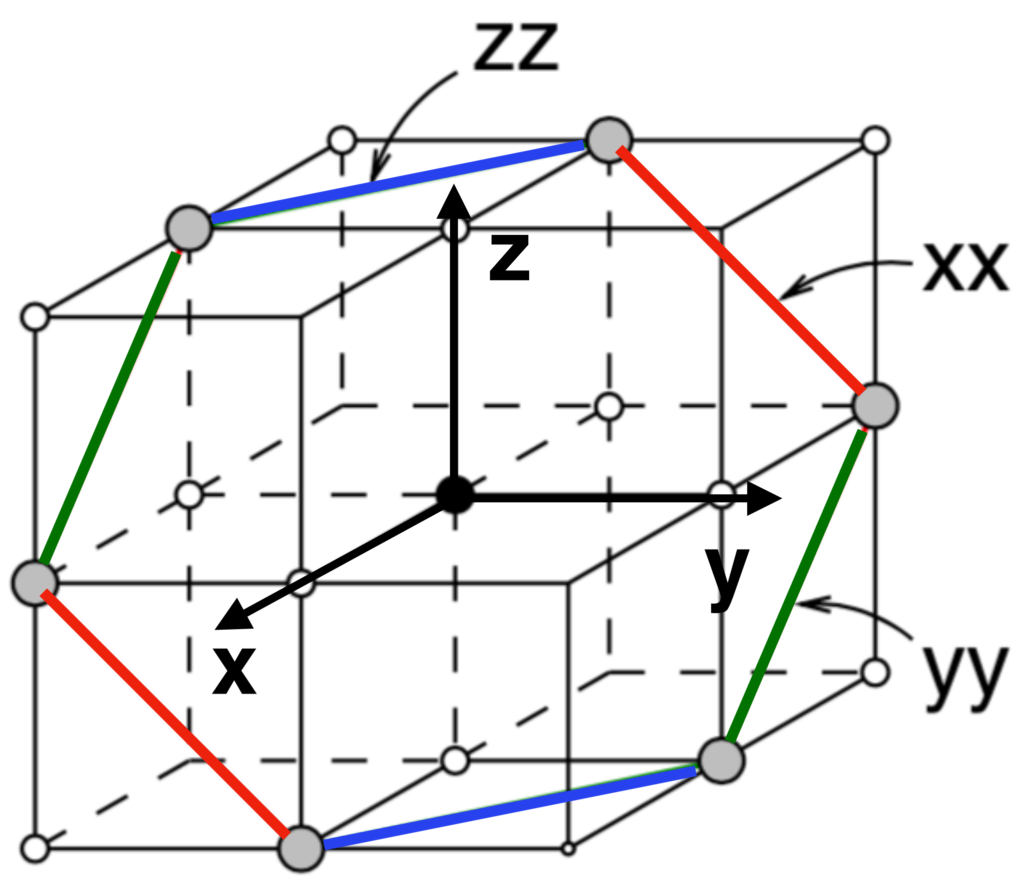
α-$RuCl_3$, Na_2IrO_3, α-Li_2IrO_3, Li_2RhO_3 …

S. M. Winter et al., J. Phys.: Condens. Matter 29, 493002 (2017)

The Kitaev model: experimental realization?

Bond-dependent Kitaev interactions





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→ Kitaev quantum spin liquid

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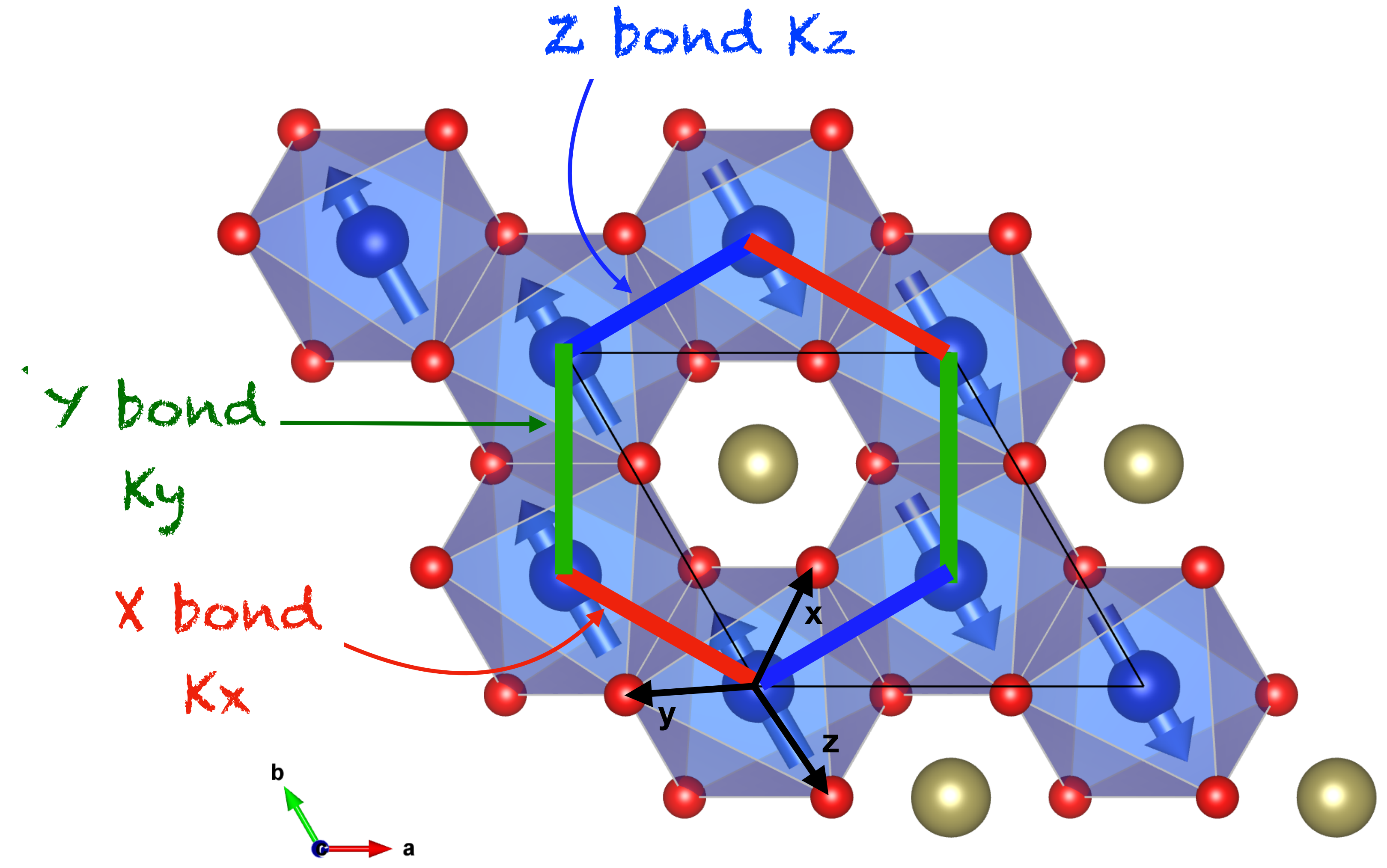
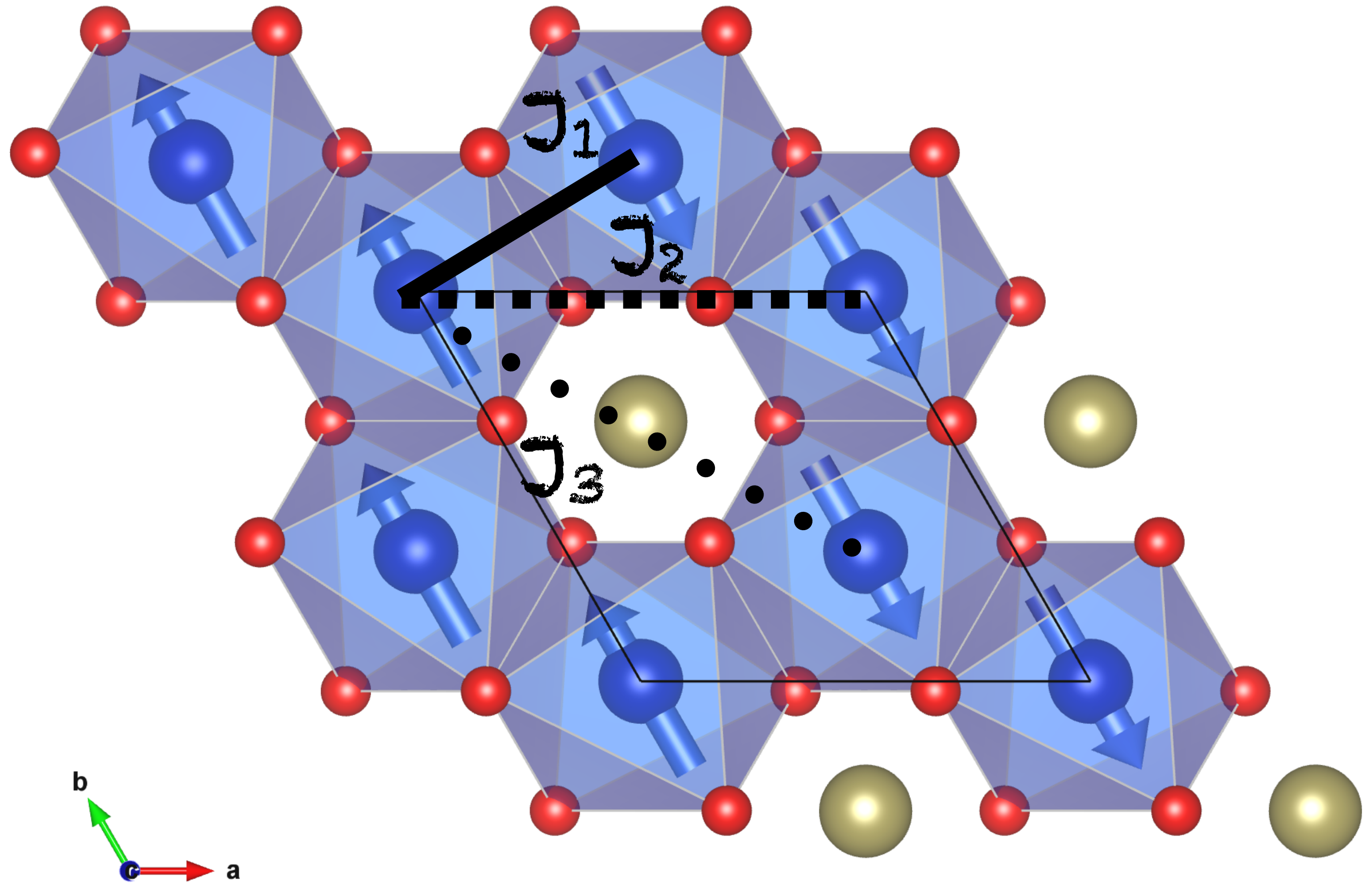
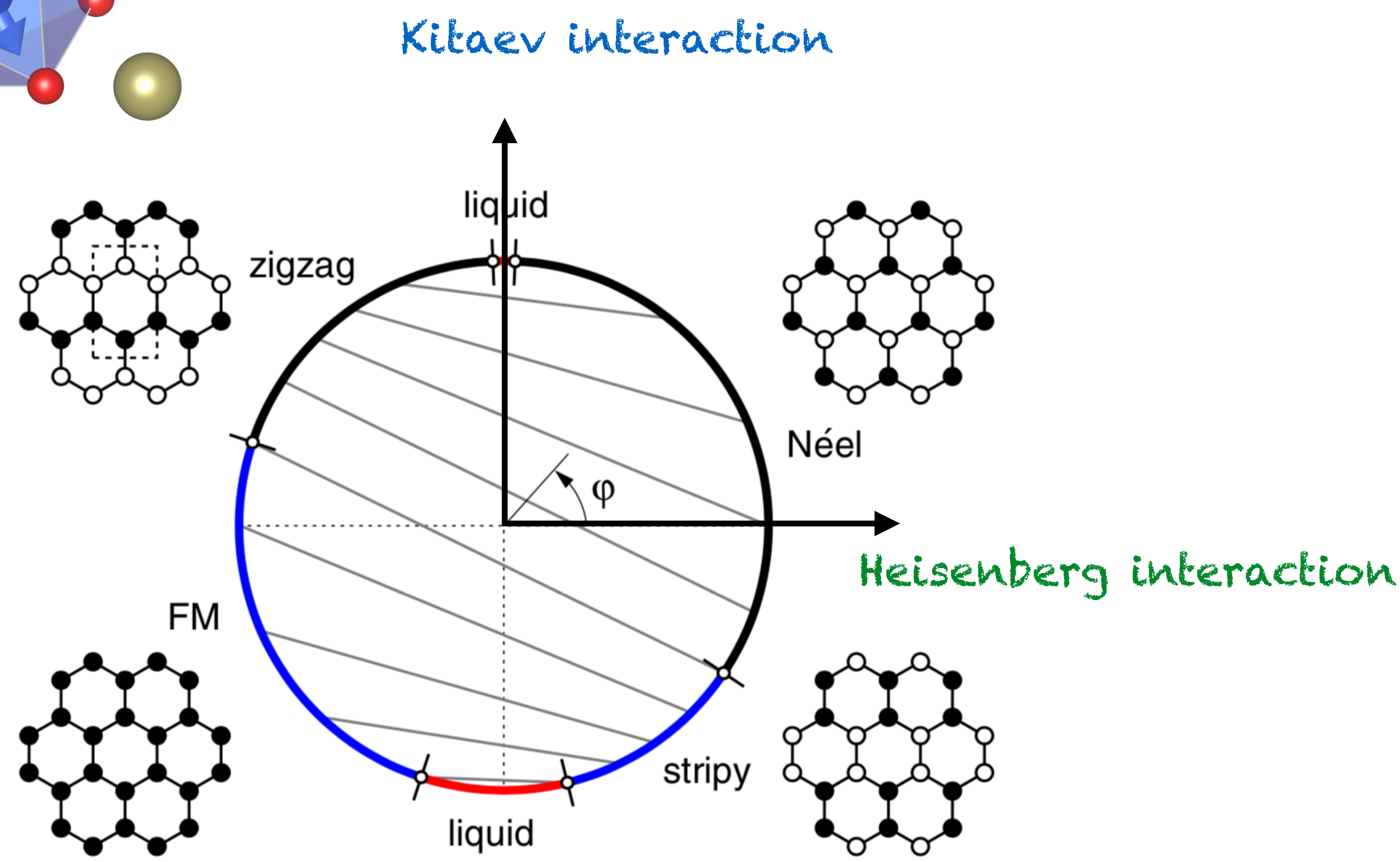


AND

Isotropic Heisenberg interactions



- Direct exchange
- Distortions : bond angles away from 90°

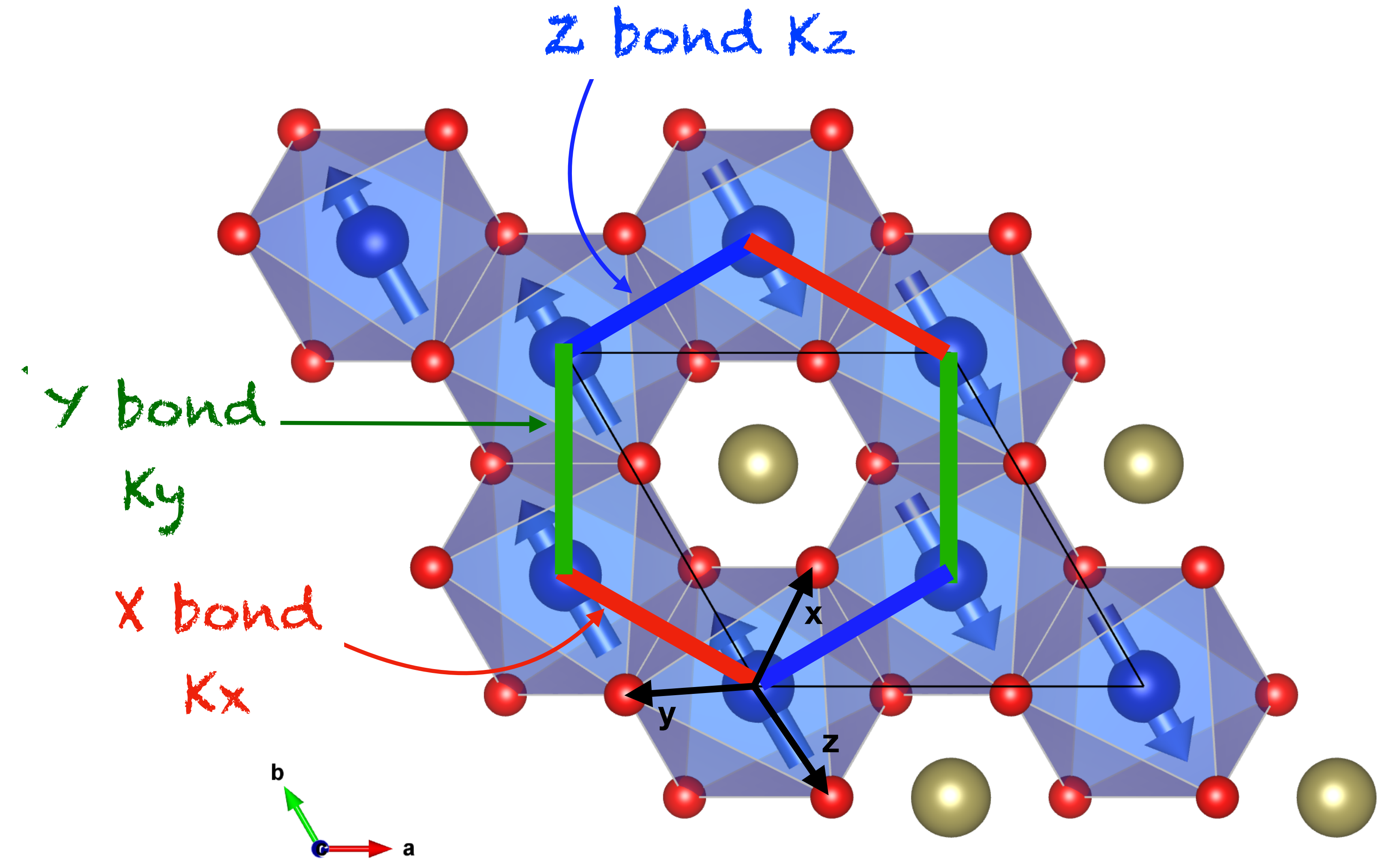


→ Long-range magnetic order at low temperature

J. Chaloupka et al., Phys. Rev. Lett. 110, 097204 (2013)

The Kitaev model: experimental realization?

Bond-dependent Kitaev interactions



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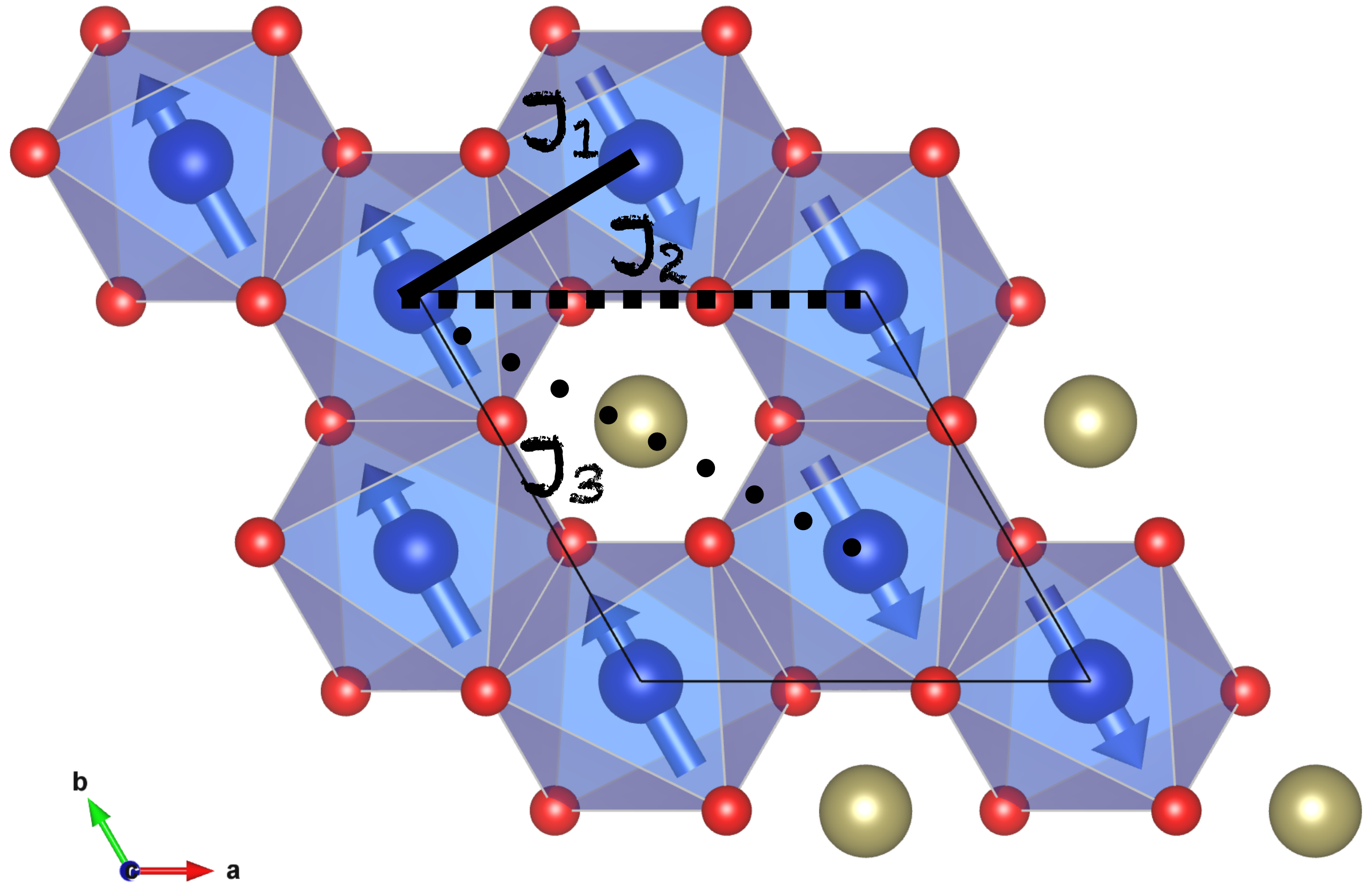

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→ Long-range magnetic order at low temperature

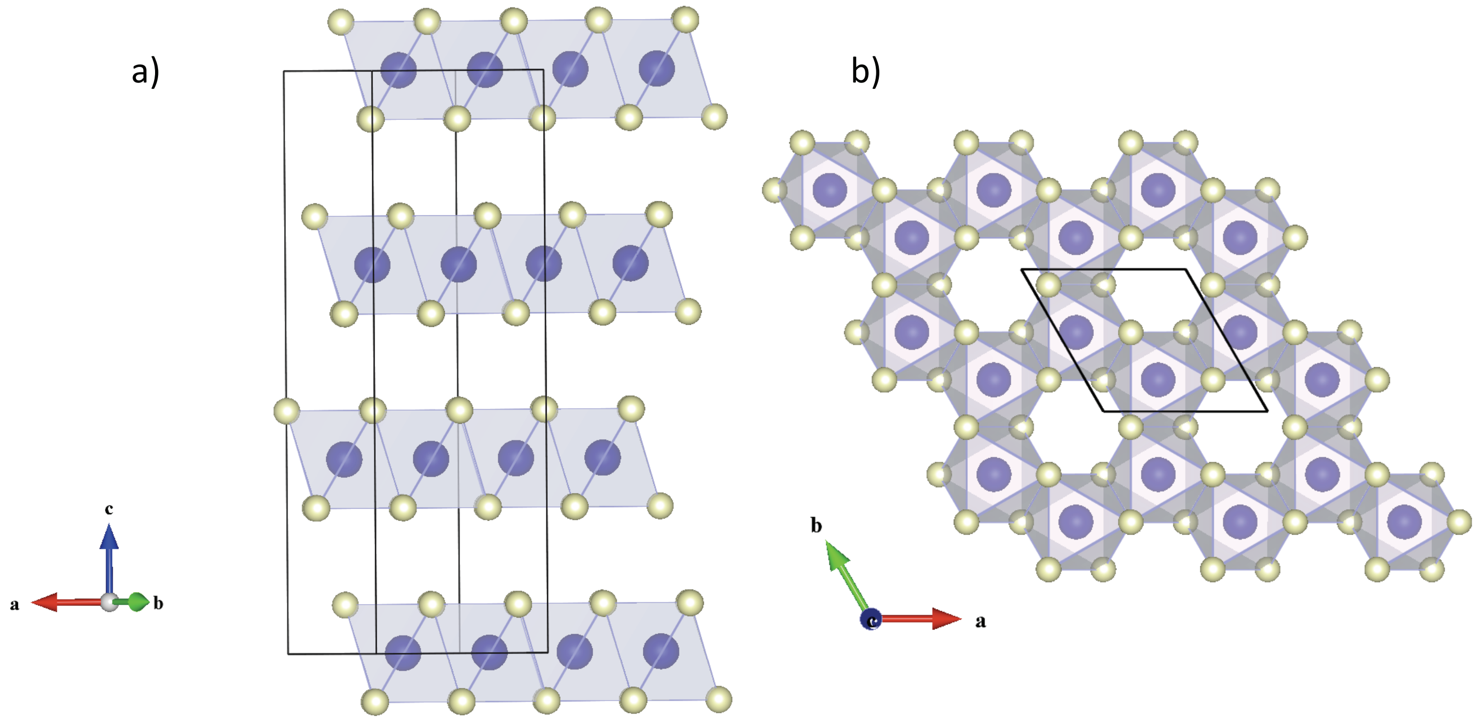
Exchange tensor:

$$\begin{pmatrix} K_x + J & \Gamma + \Gamma' & \Gamma + \Gamma' \\ \Gamma + \Gamma' & K_y + J & \Gamma + \Gamma' \\ \Gamma + \Gamma' & \Gamma + \Gamma' & K_z + J \end{pmatrix}$$

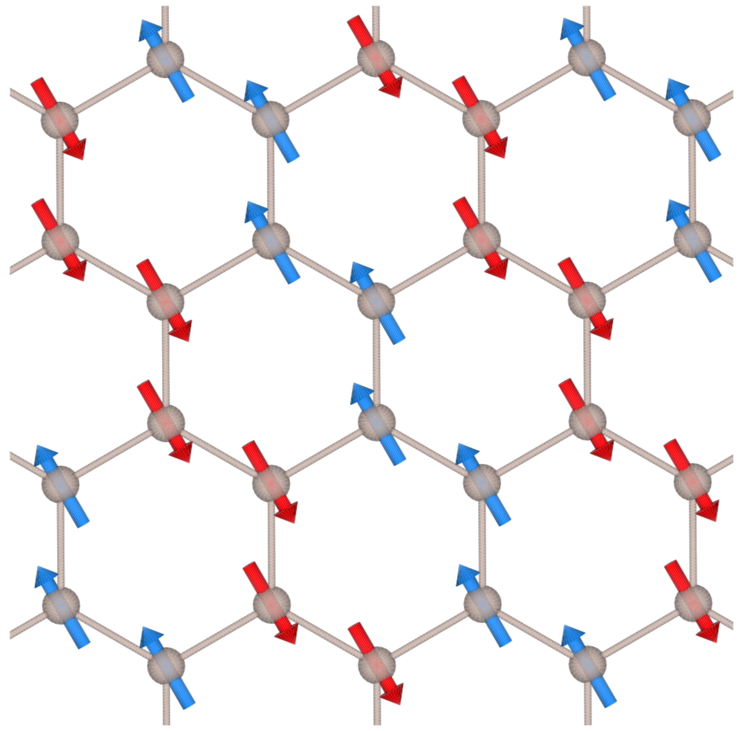
P. A. Maksimov et al., Rhys. Rev. Research 2, 03311 (2020)
H. Liu et al., Phys. Rev. Letters 125, 047201 (2020)

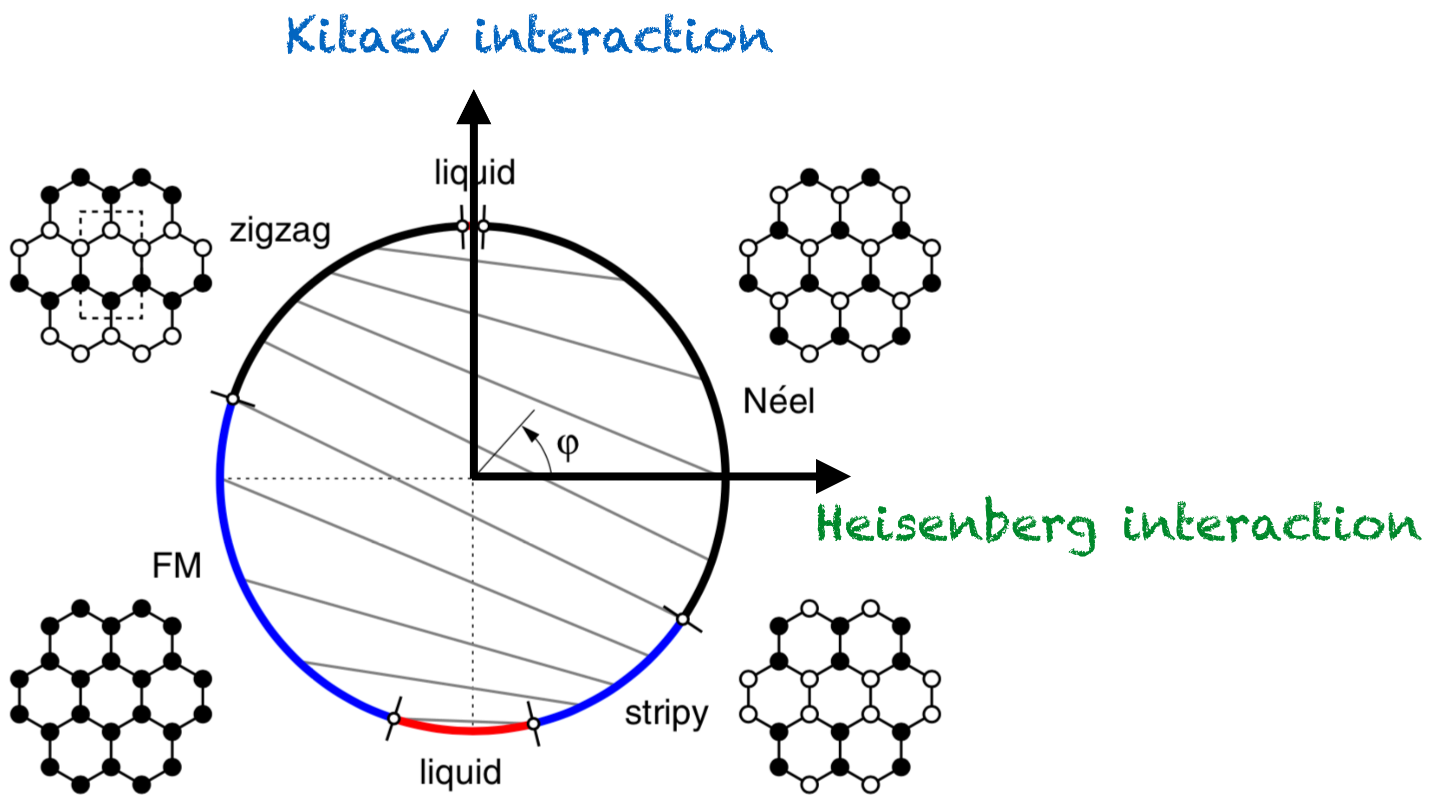
A promising material candidate: α-$RuCl_3$

A Van der Waals crystal



Zigzag magnetic order





J. A. Sears, M. Songvilay et al., Phys. Rev. B 91, 144420 (2015)
R. D. Jonhson et al., Phys. Rev. B 92, 235119 (2015)
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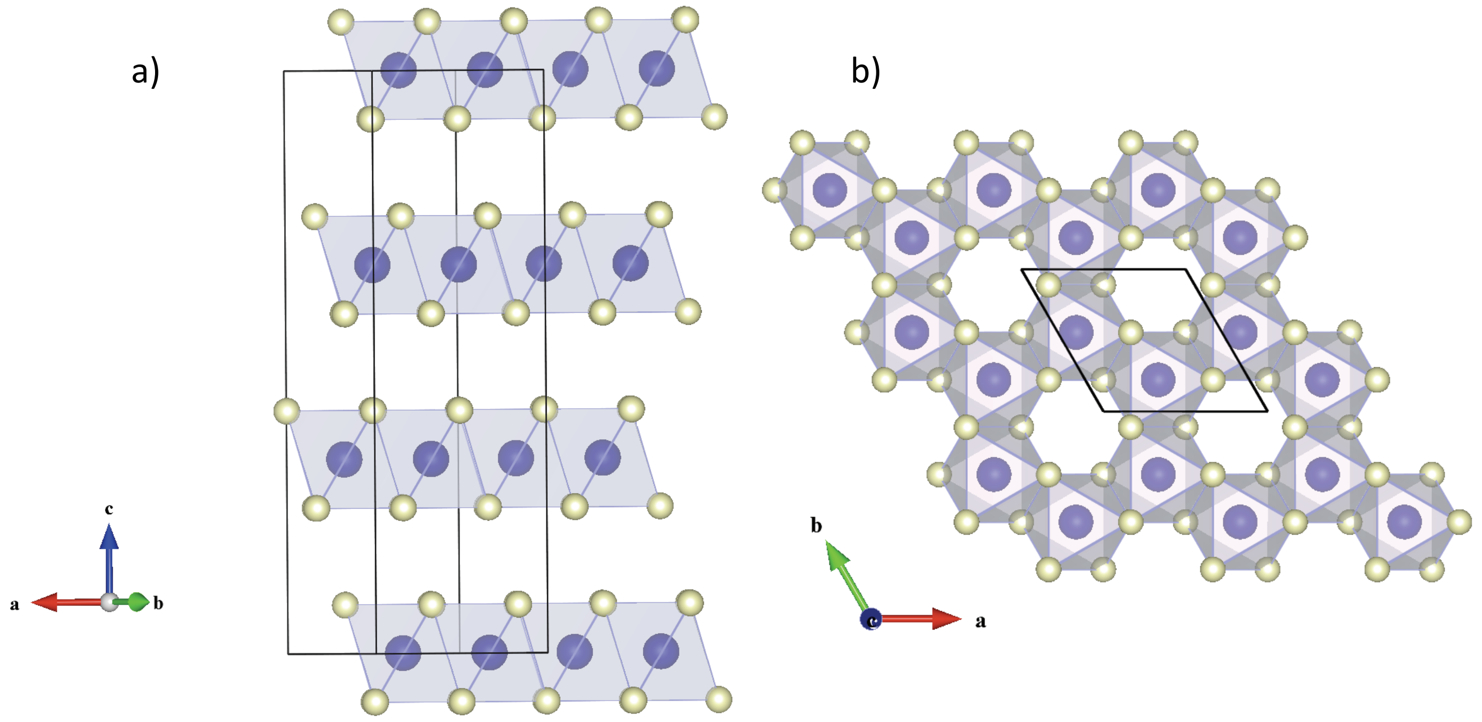

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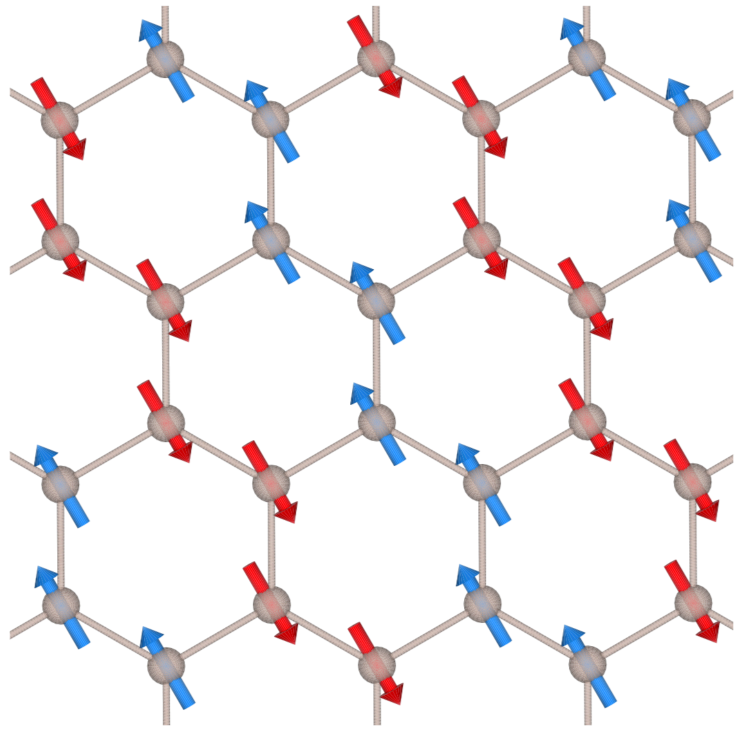

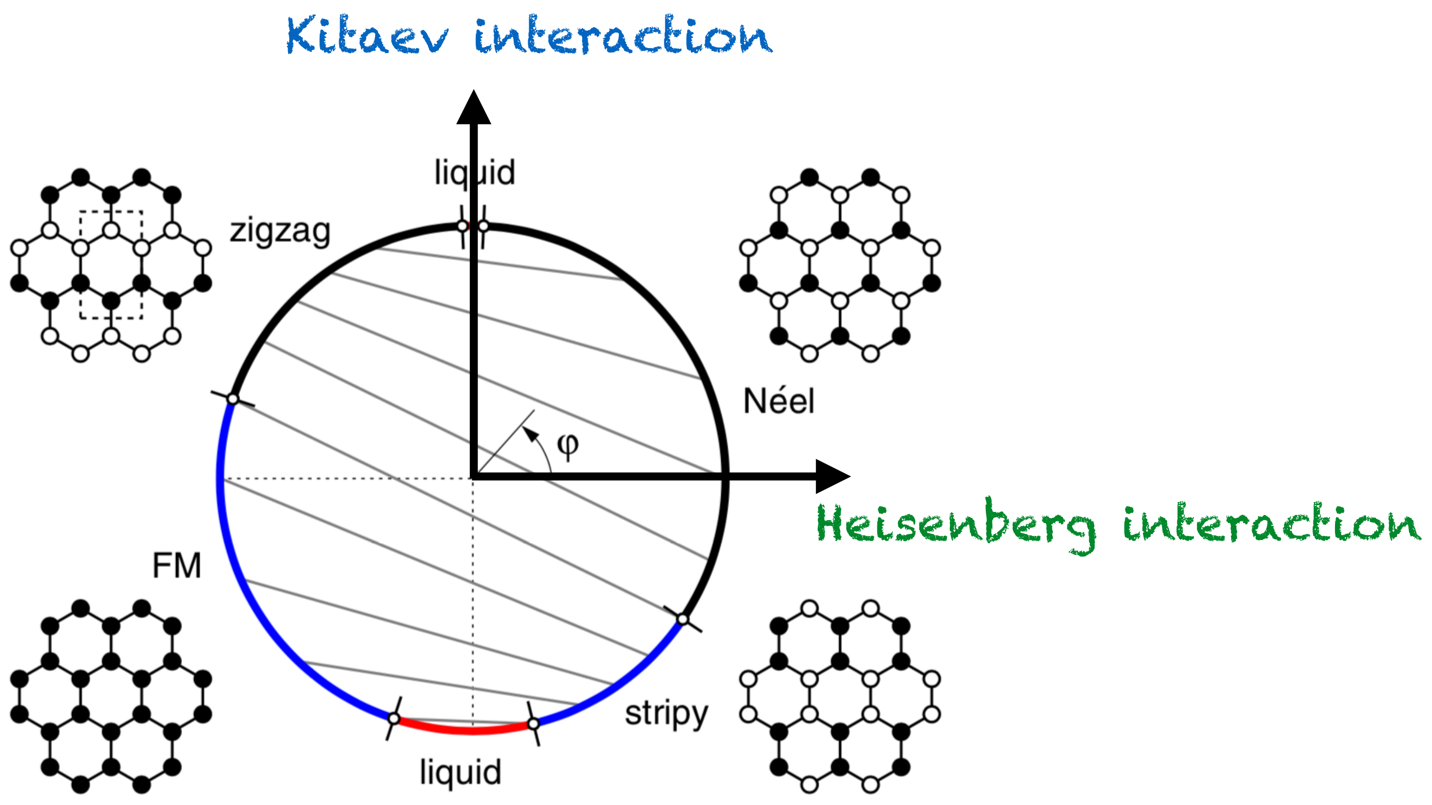

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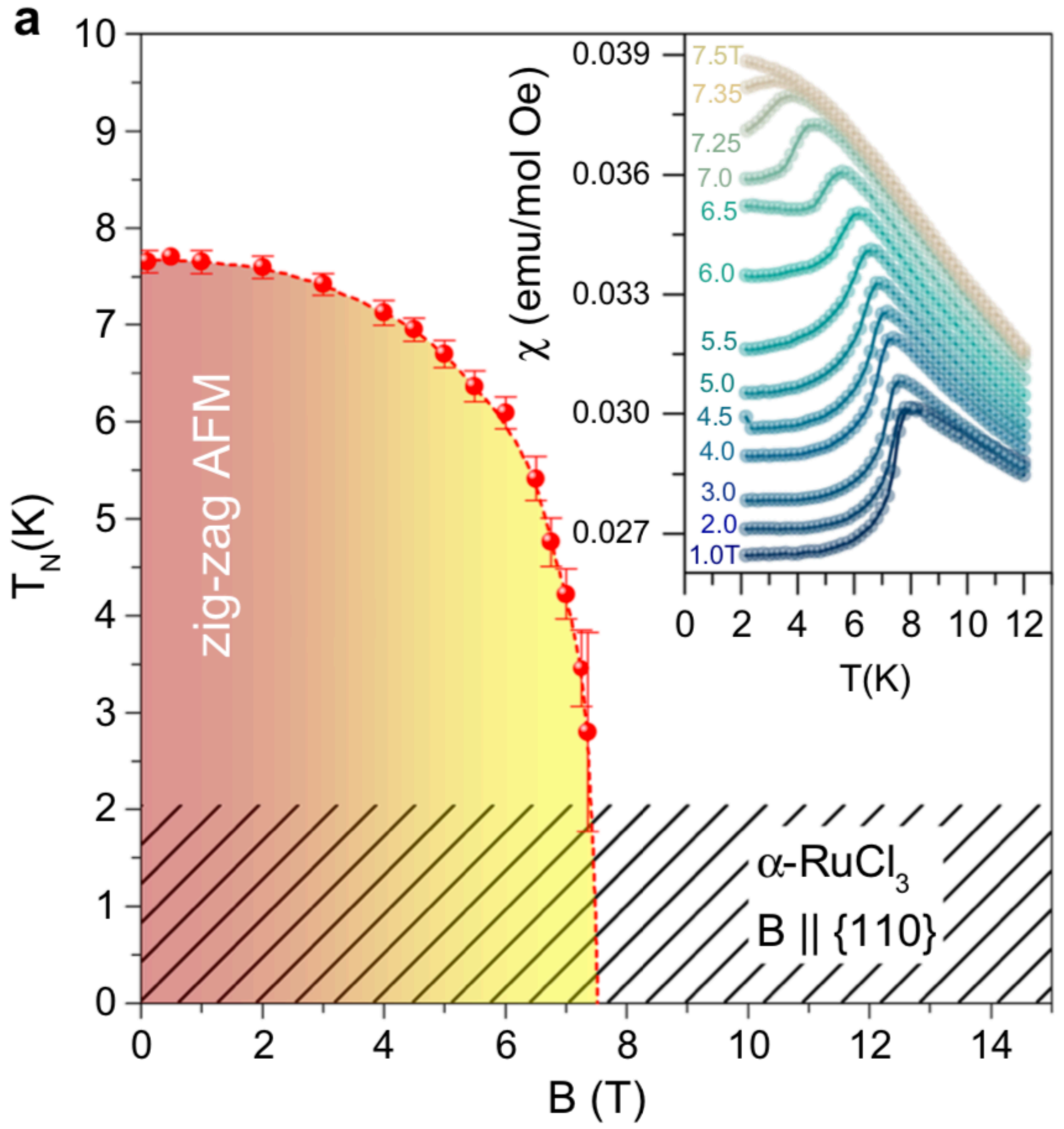
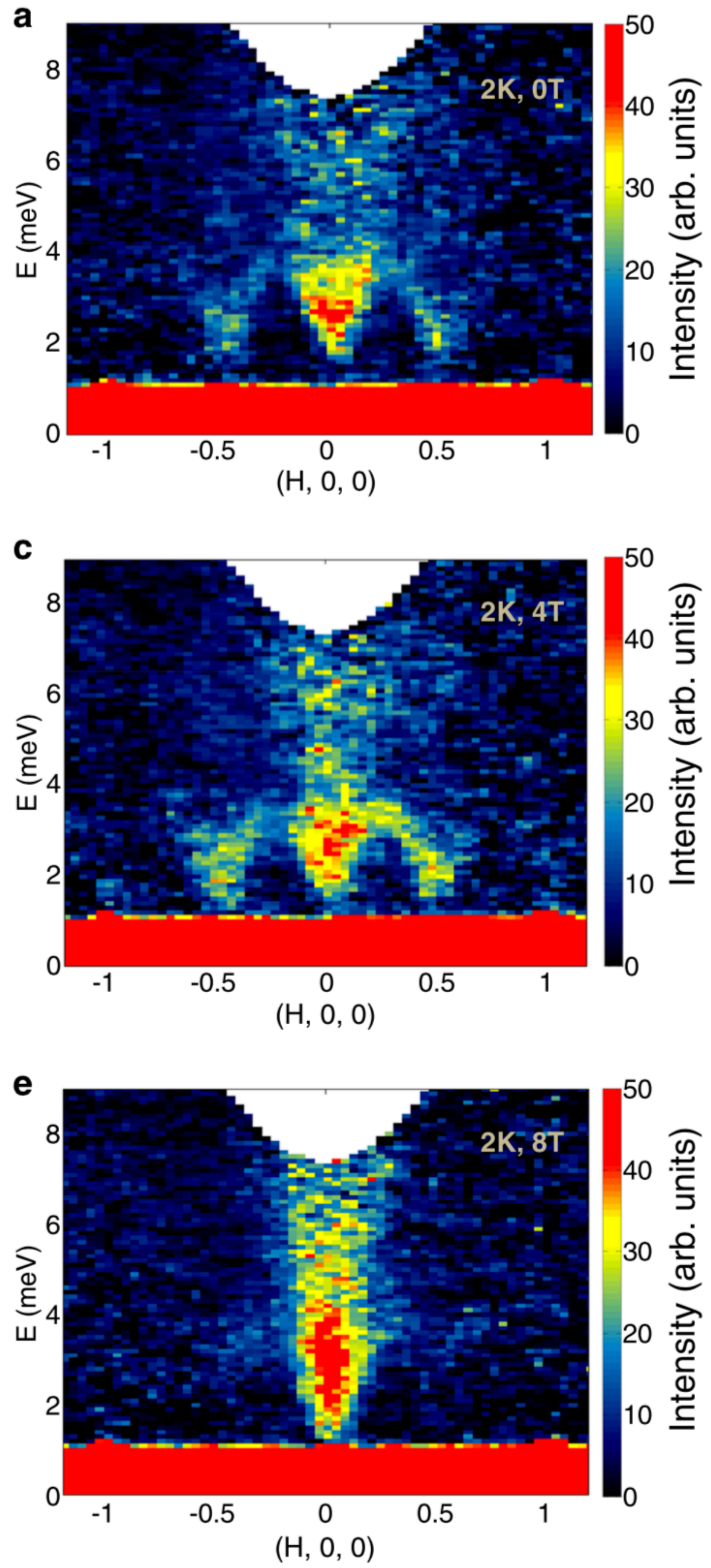
R. D. Jonhson et al., Phys. Rev. B 92, 235119 (2015)
A. Banerjee et al., Nat. Mat. 15, 733 (2016)

Possible field-induced quantum spin liquid state

- Suppression of magnetic order (in-plane field)
- gapped continuum of excitations (neutron scattering)

J. A. Sears et al., Phys. Rev. B 95, 180411(R) (2017)
A. Banerjee et al., npj Quantum Materials 3, 8 (2018)





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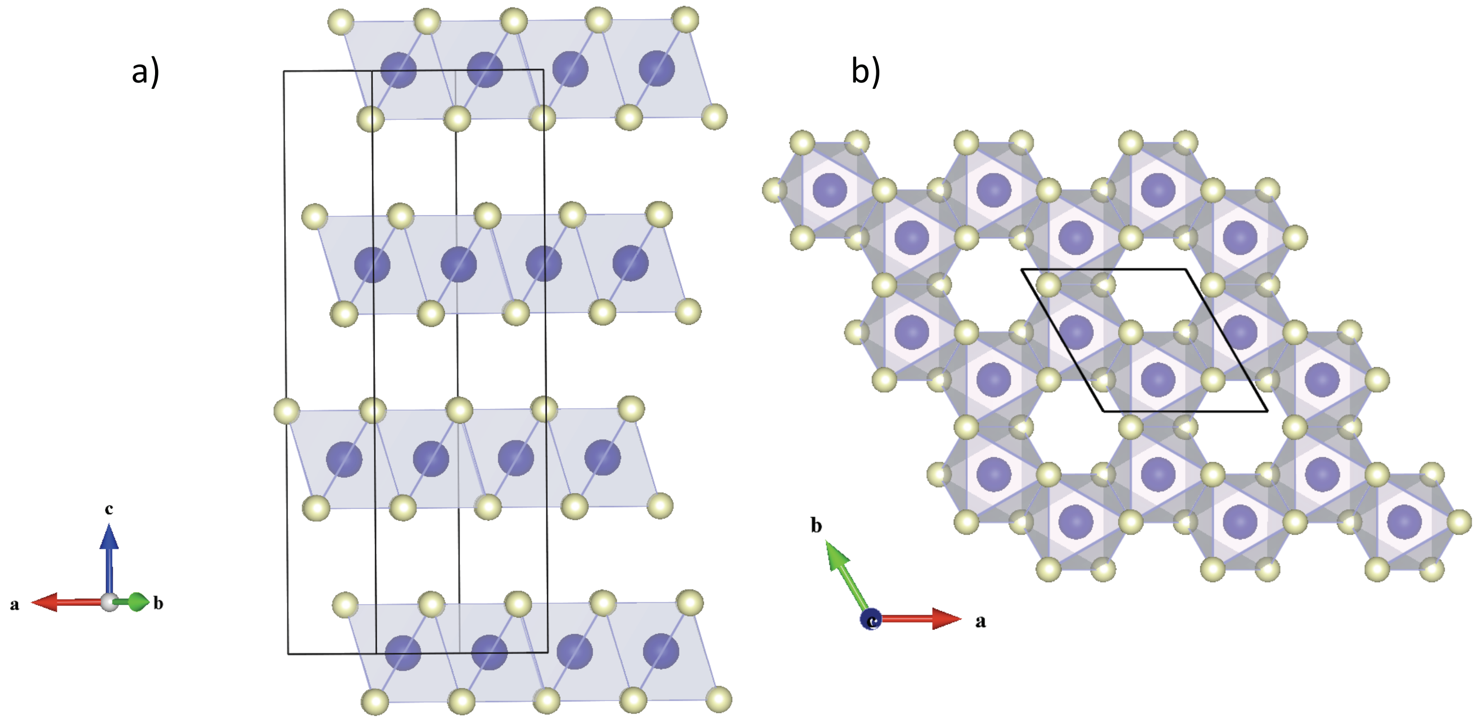

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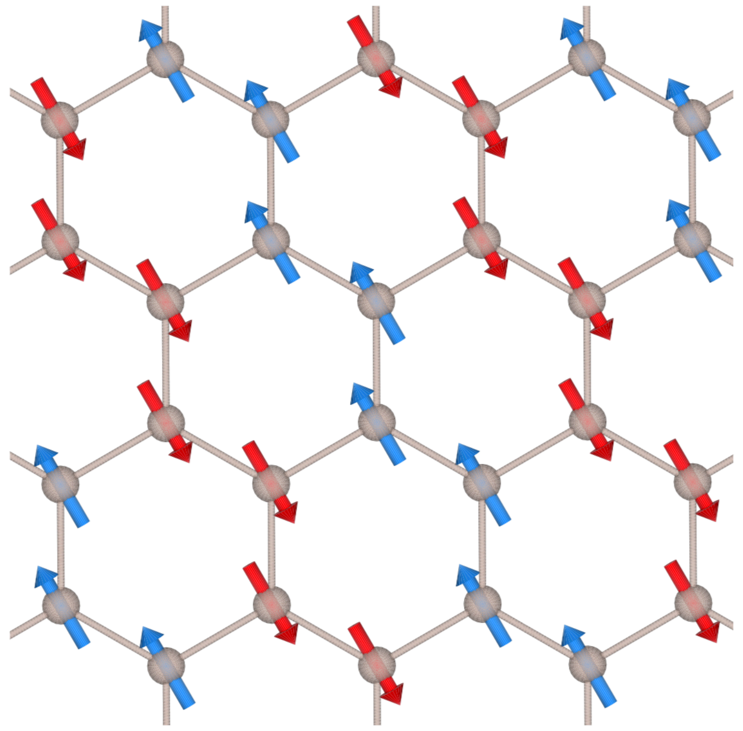

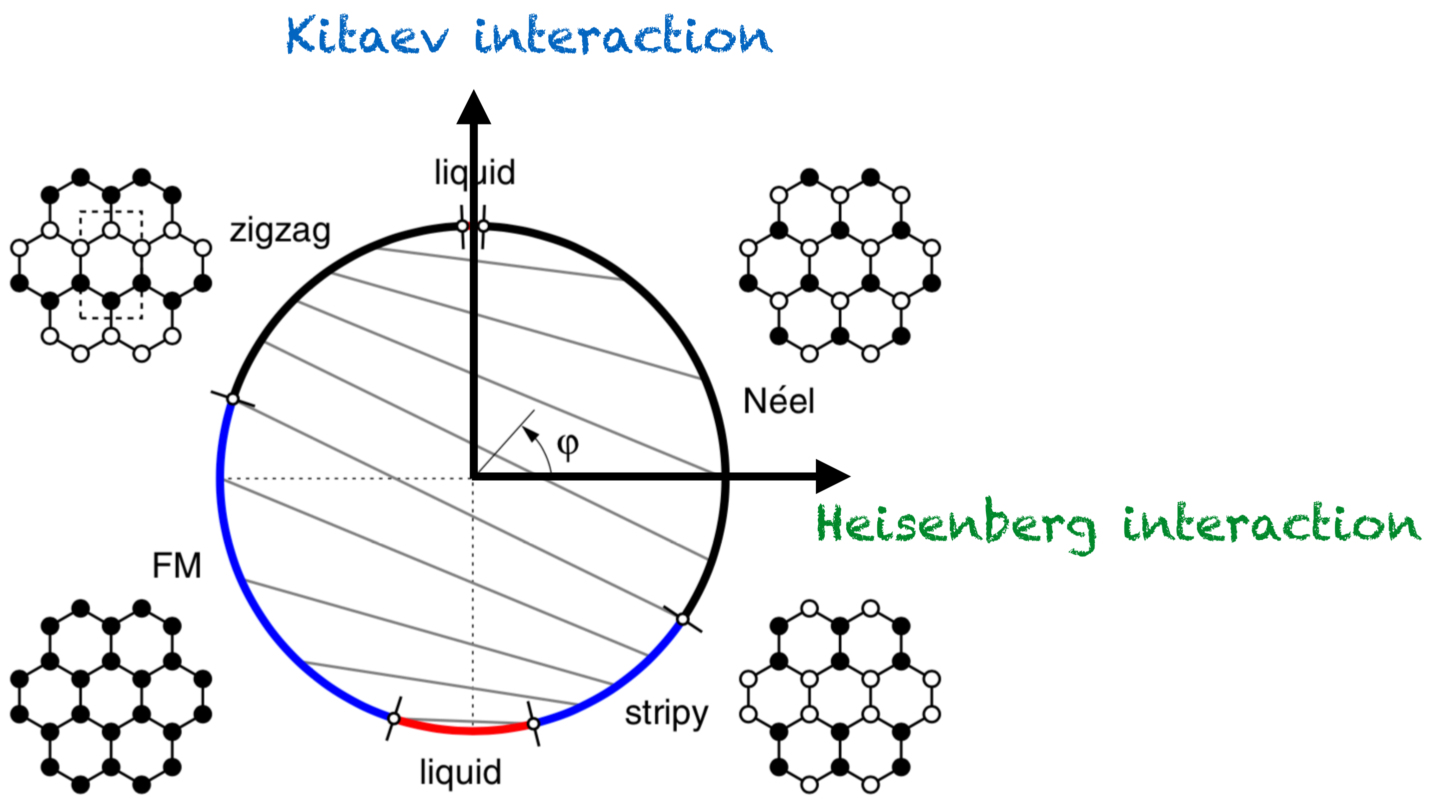

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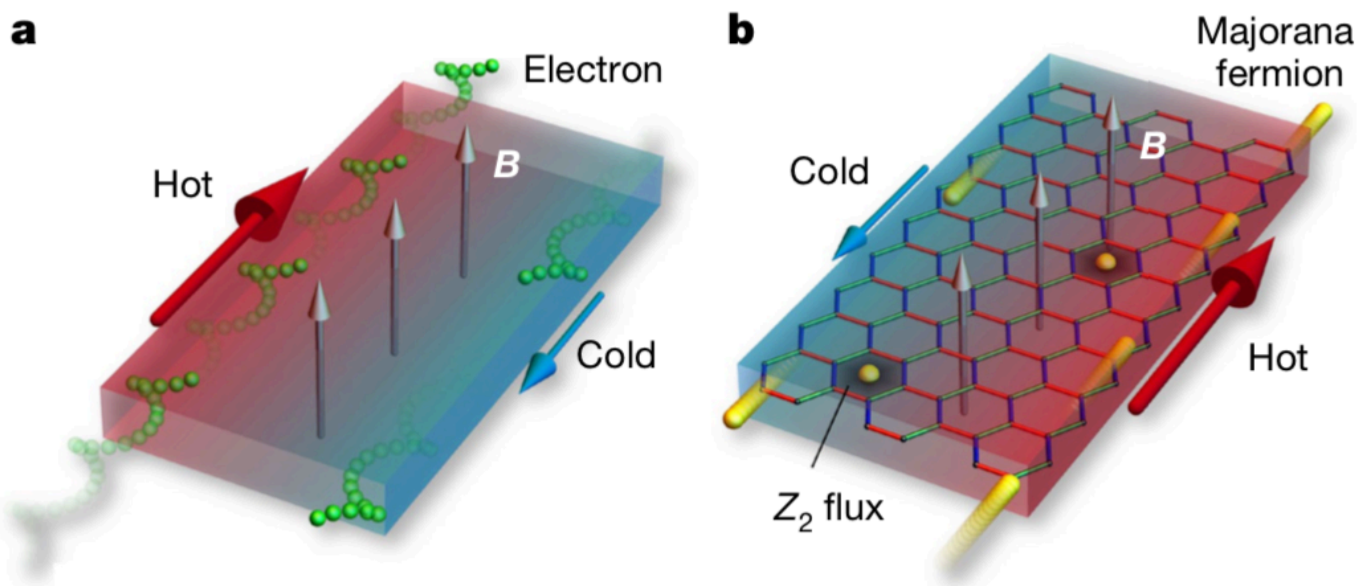
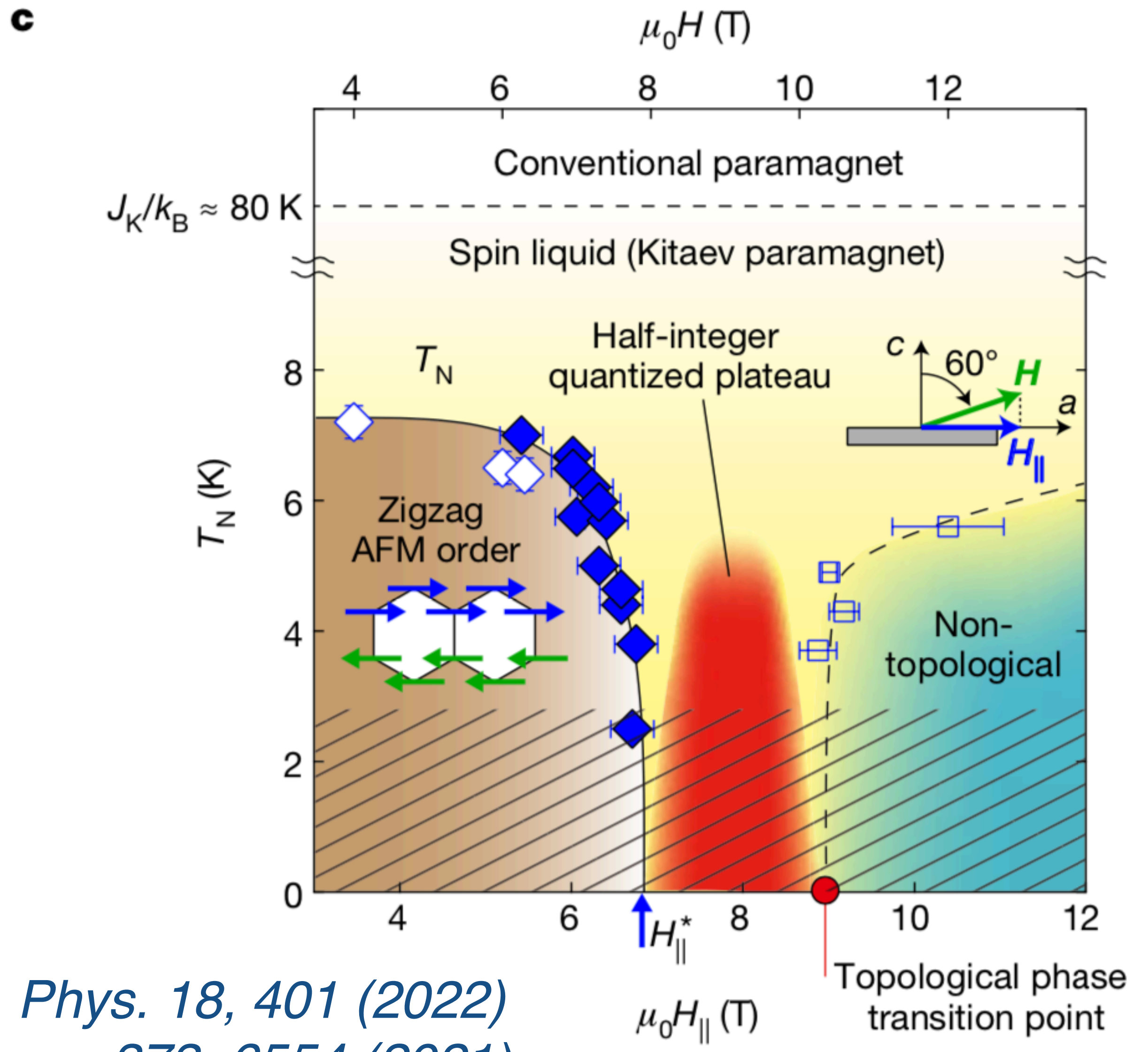
Half-integer thermal hall conductance : evidence for edge current Majorana-like excitations ?

- **Topological phase under magnetic field ?**

Y. Kasahara et al., Nature 559, 7713 (2018)





J. Bruin et al., Nat. Phys. 18, 401 (2022)
T. Yokoi et al., Science 373, 6554 (2021)

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▷ **Topological phase under magnetic field ?**

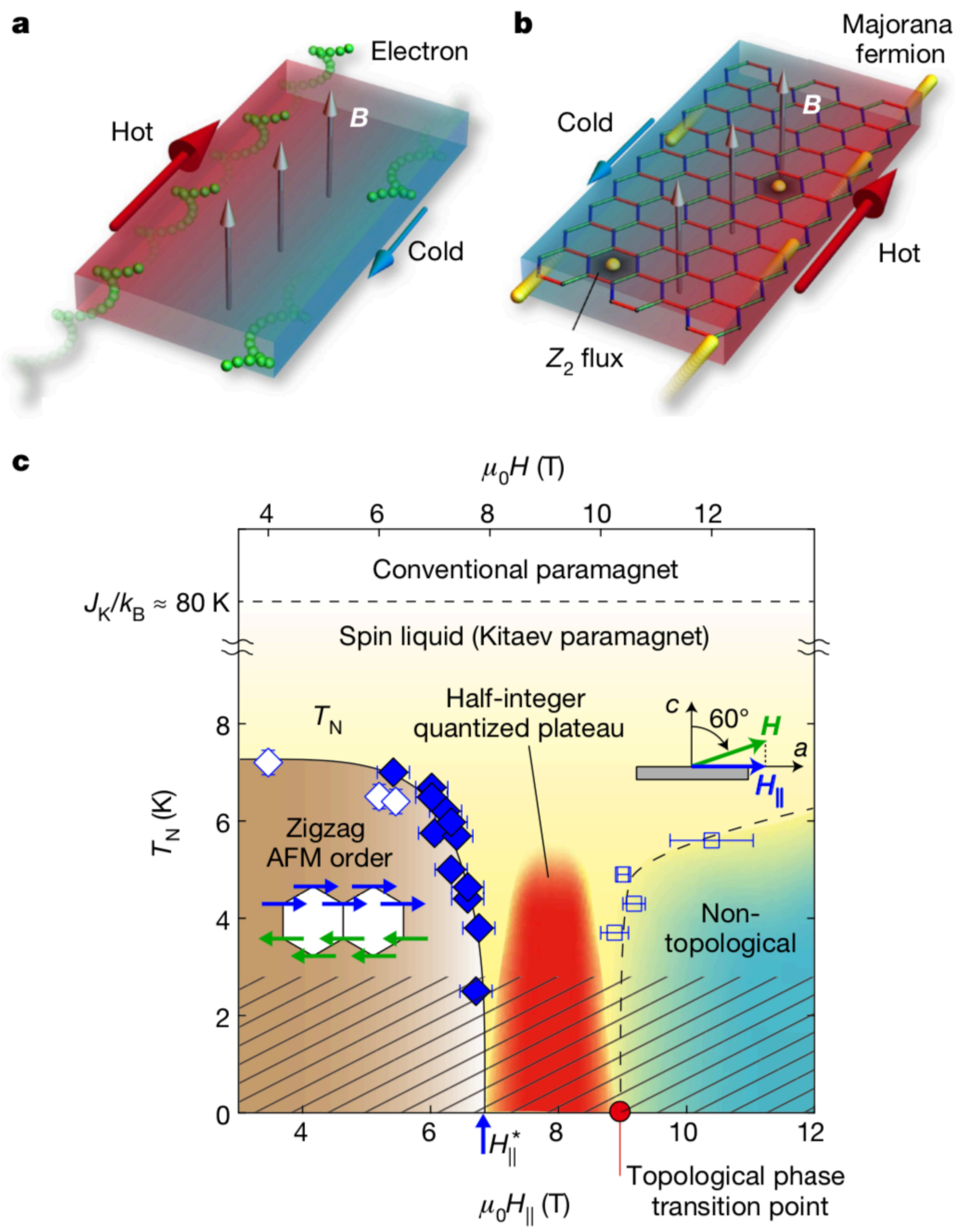
Y. Kasahara et al., Nature 559, 7713 (2018)

→ Other contributions to the thermal Hall effect?

E. Zhang et al., Phys. Rev. B 103, 174402 (2021)

→ Possible strong contribution of the phonons to the thermal Hall conductivity?

R. Hentrich et al., Phys. Rev. Lett. 120, 117204 (2018)
E. Lefrançois et al., Phys. Rev. X 12, 021025 (2022)



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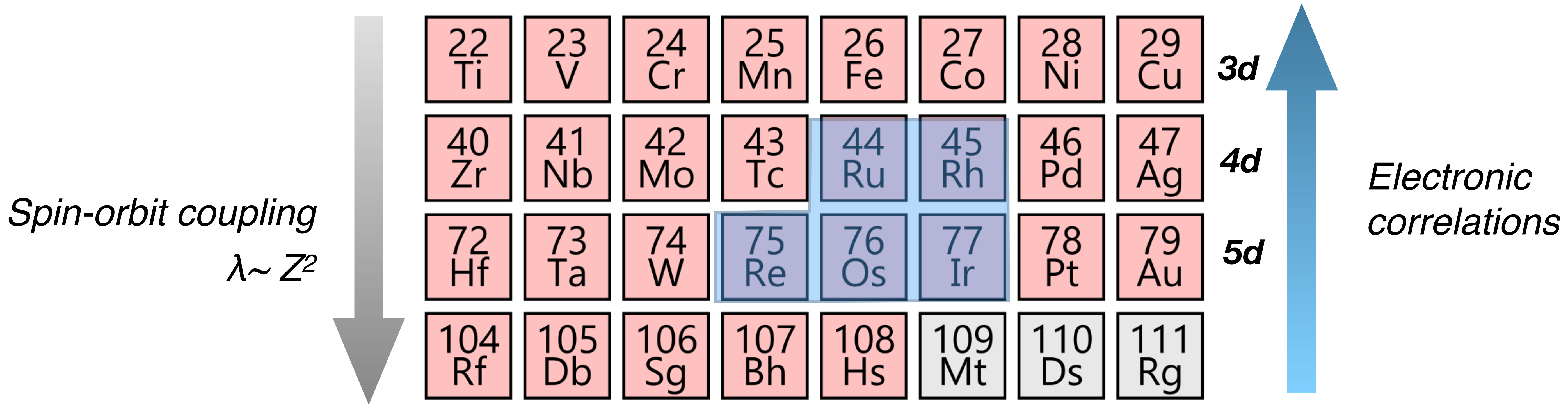
Extending the search for Kitaev materials

Beyond the Jackeli-Khaliullin proposal :

✓ Spin-orbit coupling/electron correlations : 3d transition metals, rare-earth, spin value…

✓ Other structures : organic materials, 3D lattices, triangular lattices…



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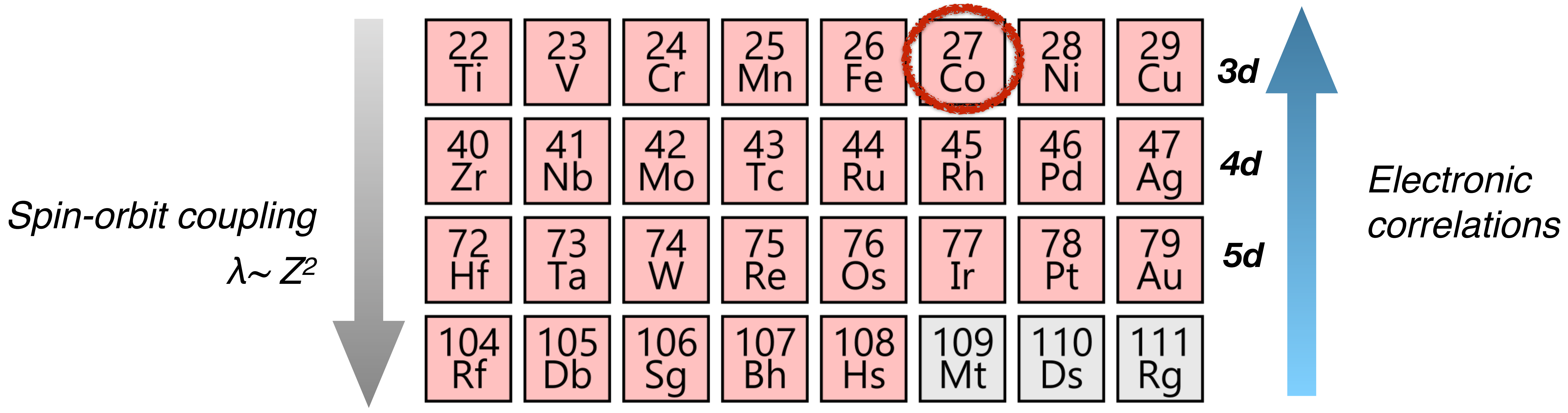


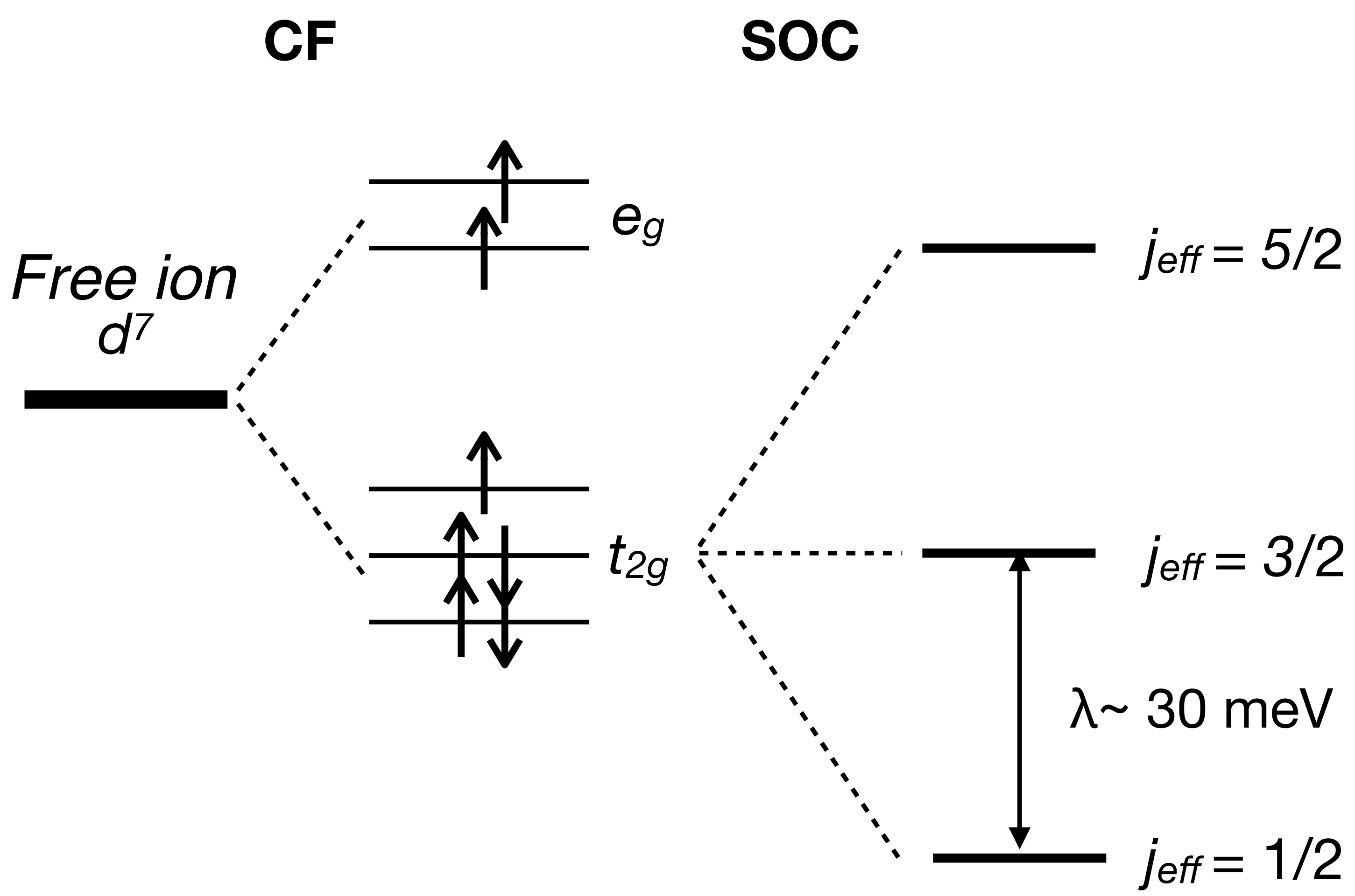
H. Liu and G. Khaliullin, Phys. Rev. B 97, 014407 (2018)
R. Sano et al., Phys. Rev. B 97, 014408 (2018)
H. Liu et al., Phys. Rev. Letters 125, 047201 (2020)

Beyond the Jackeli-Khaliullin proposal: cobalt materials

- Theoretical proposal to realise the Kitaev model in high-spin d7 systems

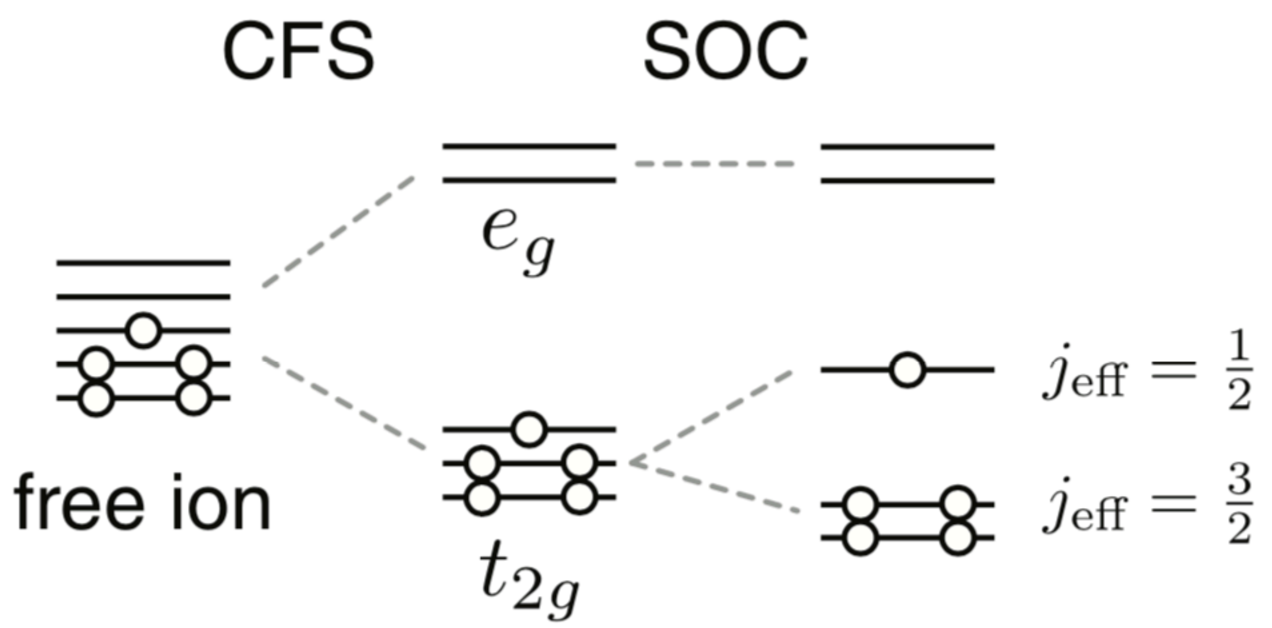
H. Liu and G. Khaliullin, Phys. Rev. B 97, 014407 (2018)
R. Sano et al., Phys. Rev. B 97, 014408 (2018)
H. Liu et al., Phys. Rev. Letters 125, 047201 (2020)

Co^{2+} in octahedral environment : high spin $3d^7$ configuration (S = 3/2, L = 1)



VS

Ir^{4+} in octahedral environment: $5d^5$ configuration

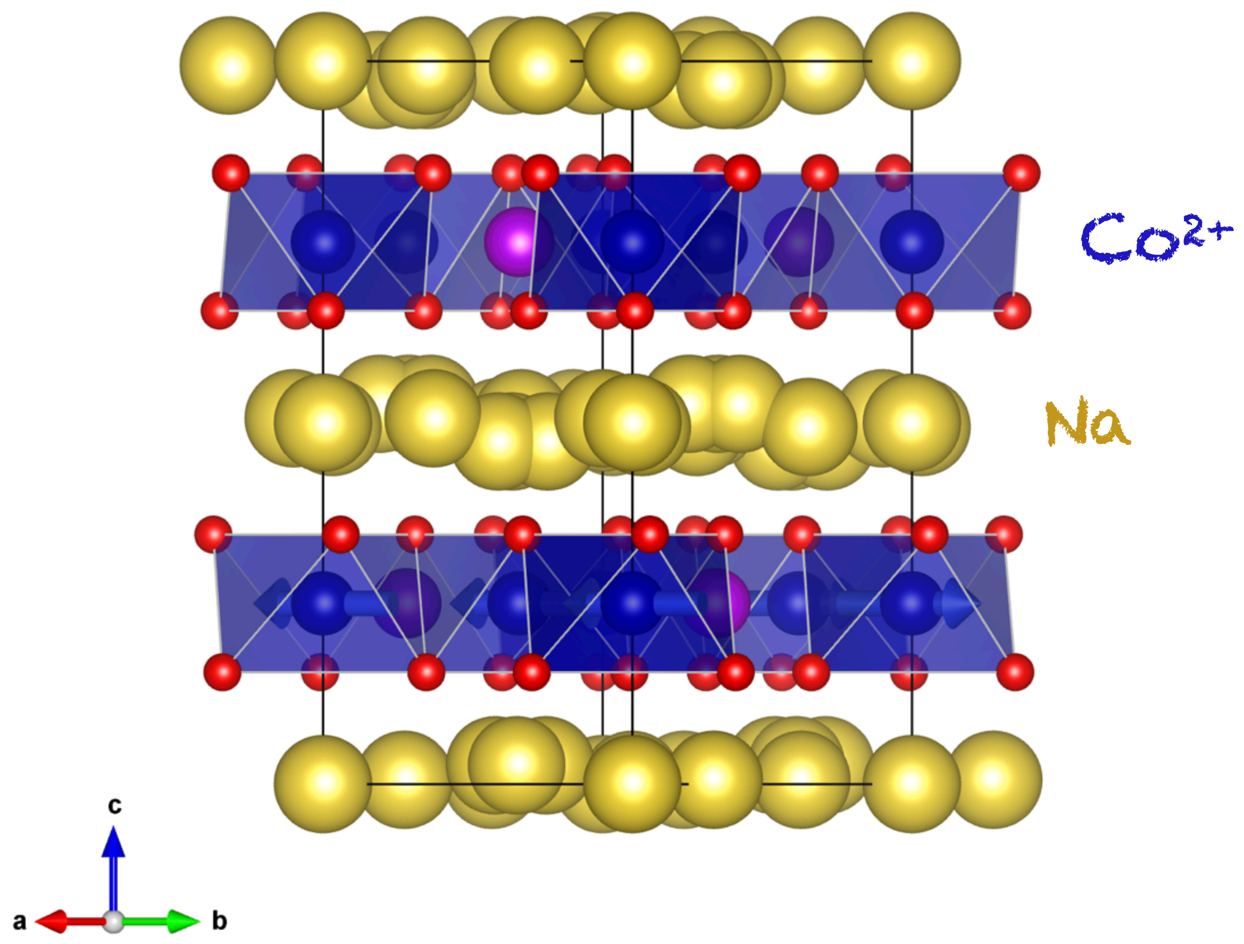


S. M. Winter et al., J. Phys.: Condens. Matter 29, 493002 (2017)

Beyond the Jackeli-Khaliullin proposal: cobalt materials

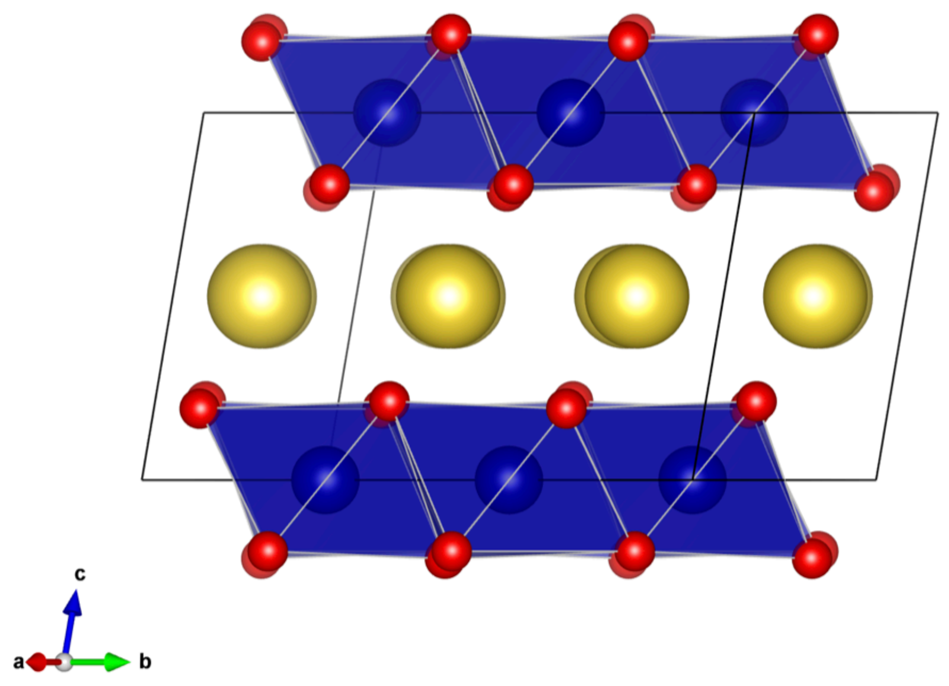
$Na_2Co_2TeO_6$

space group : $P6_322$ (hexagonal)



$Na_3Co_2SbO_6$

space group : C2/m (monoclinic)



E. Lefrançois et al., Phys. Rev. B 94, 214416 (2016)
A. K. Bera et al., Phys. Rev. B 95, 094424 (2017)
C. Wong et al., J. Sol. State. Chem. 243 (2016)
J.-Q Yan et al., Phys. Rev. Mat. 3, 074405 (2019)

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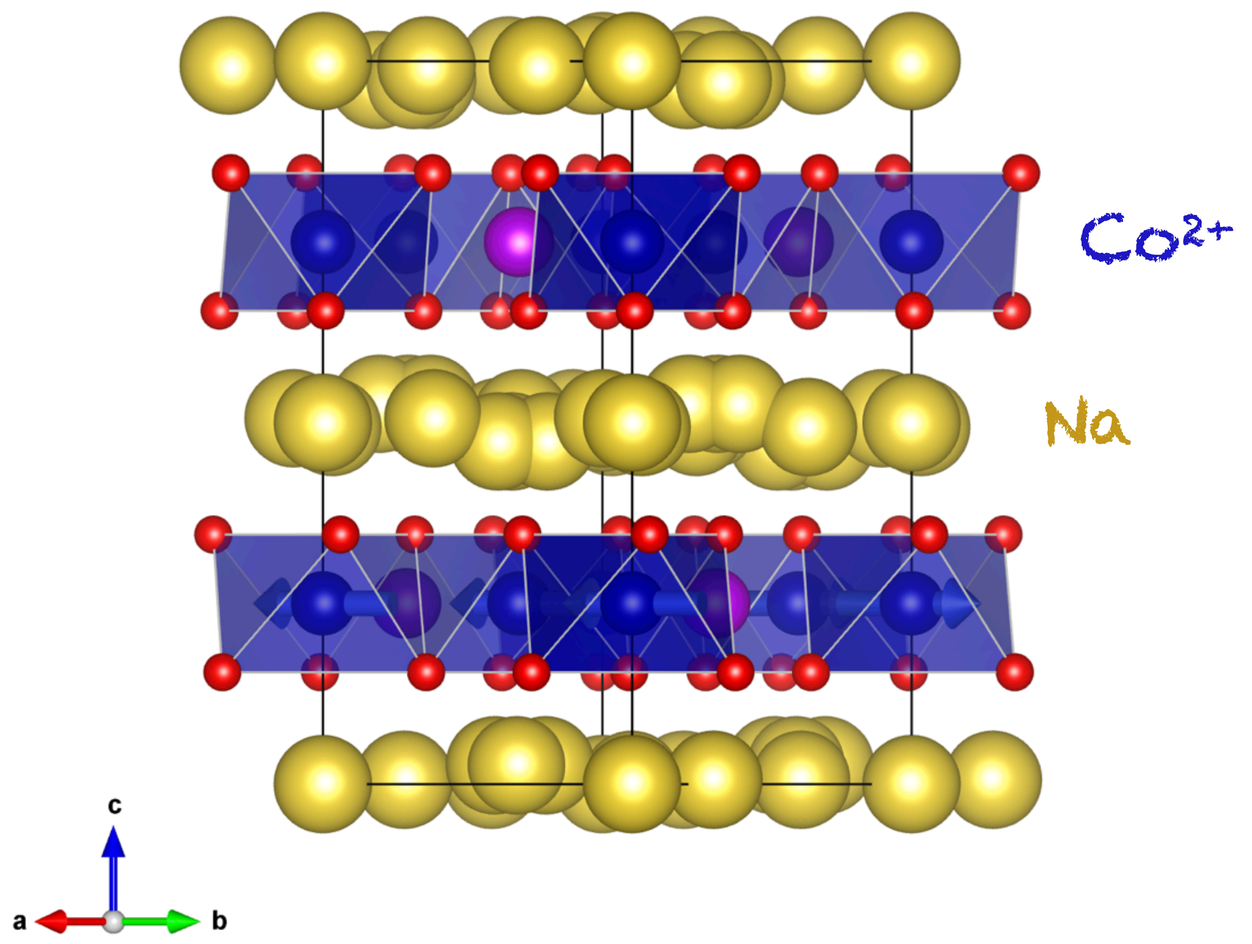

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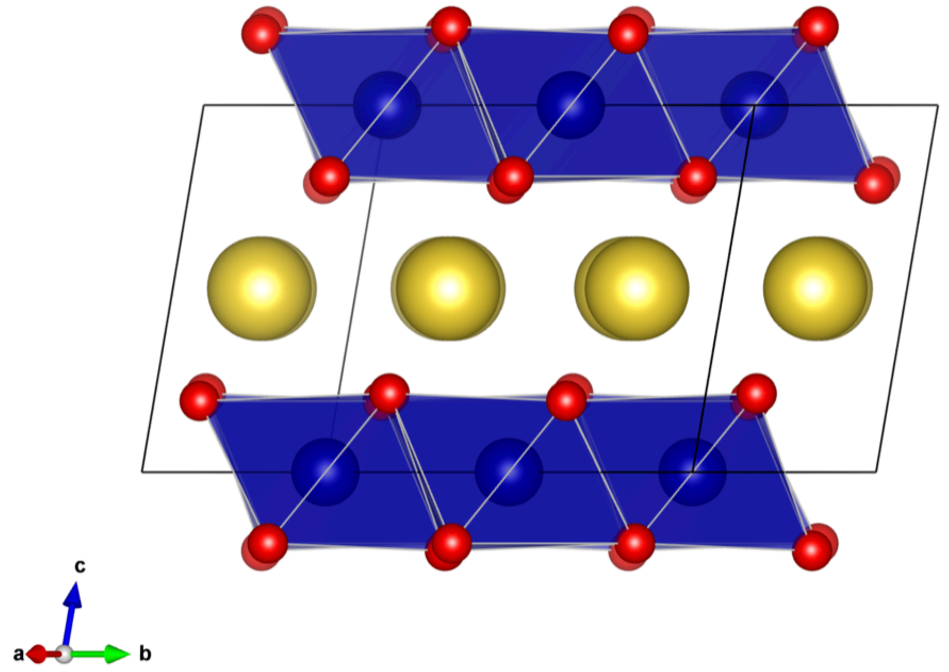
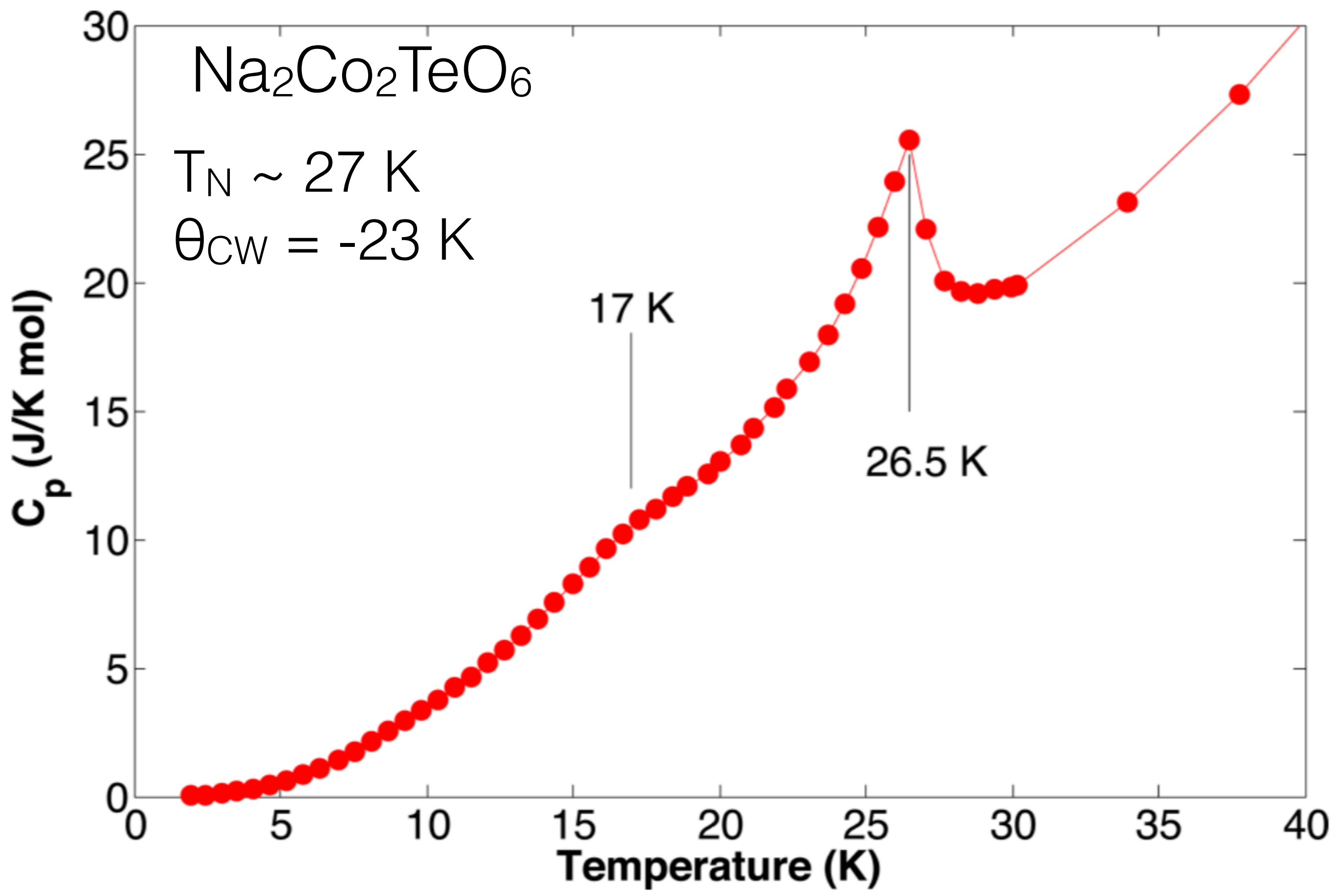
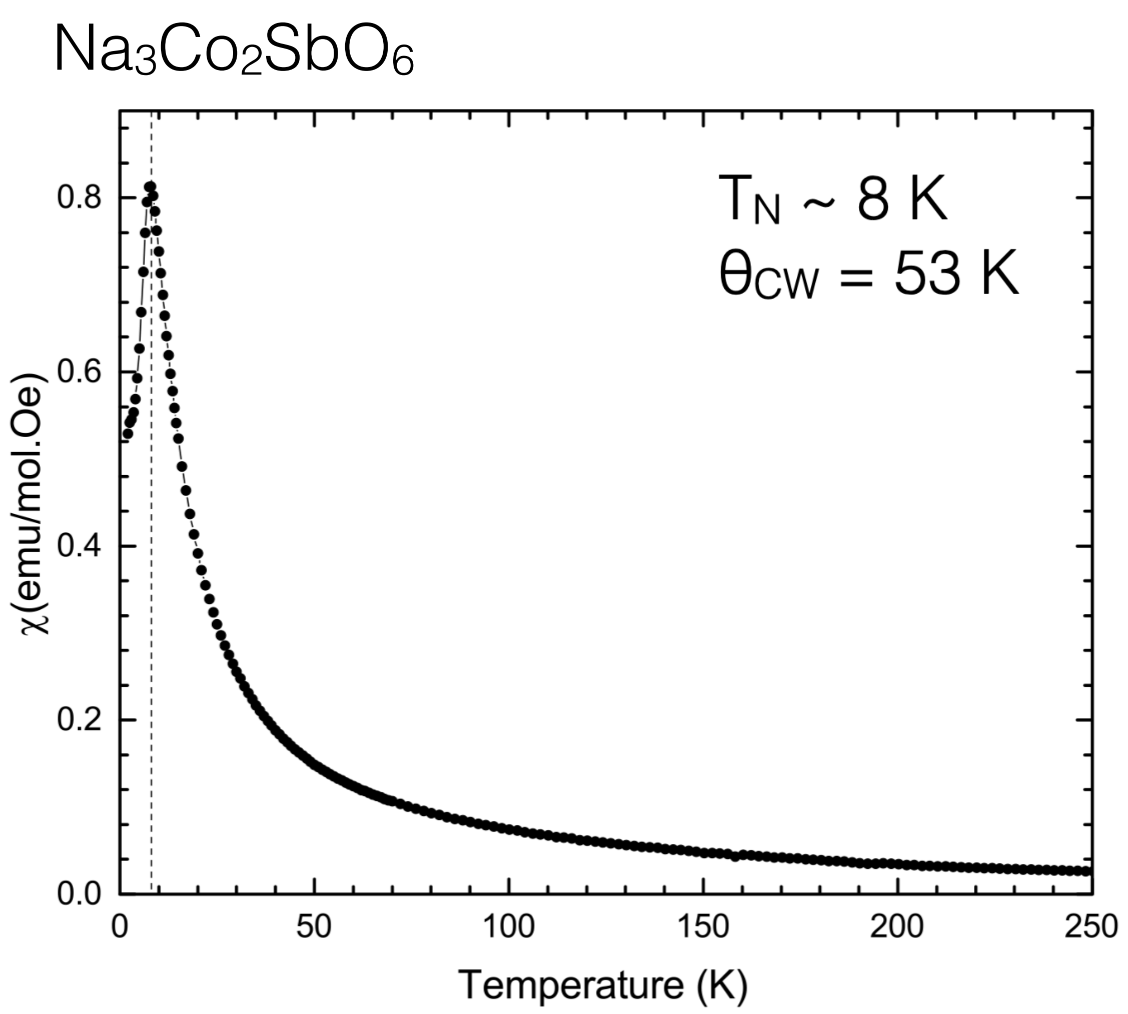


Magnetic properties





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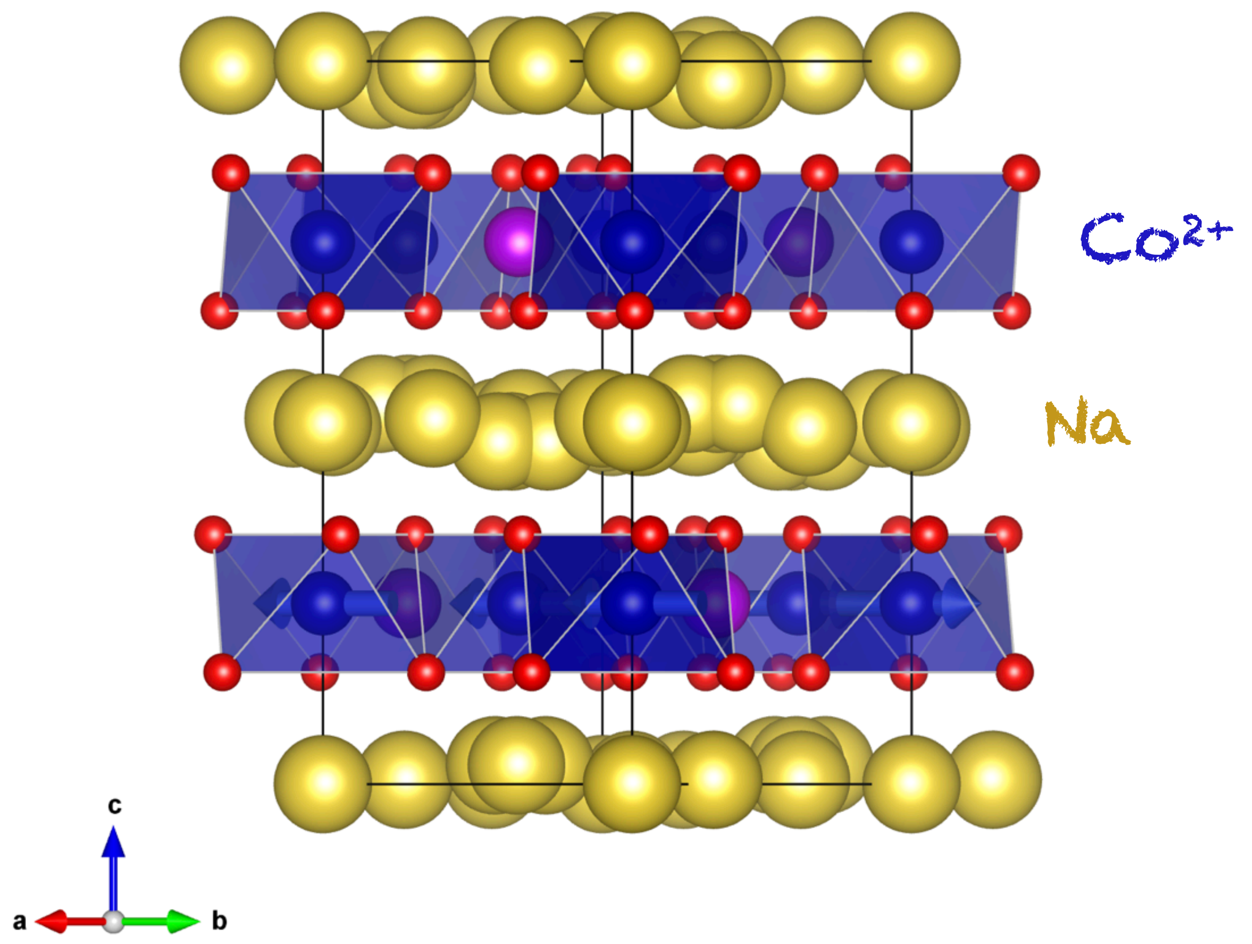
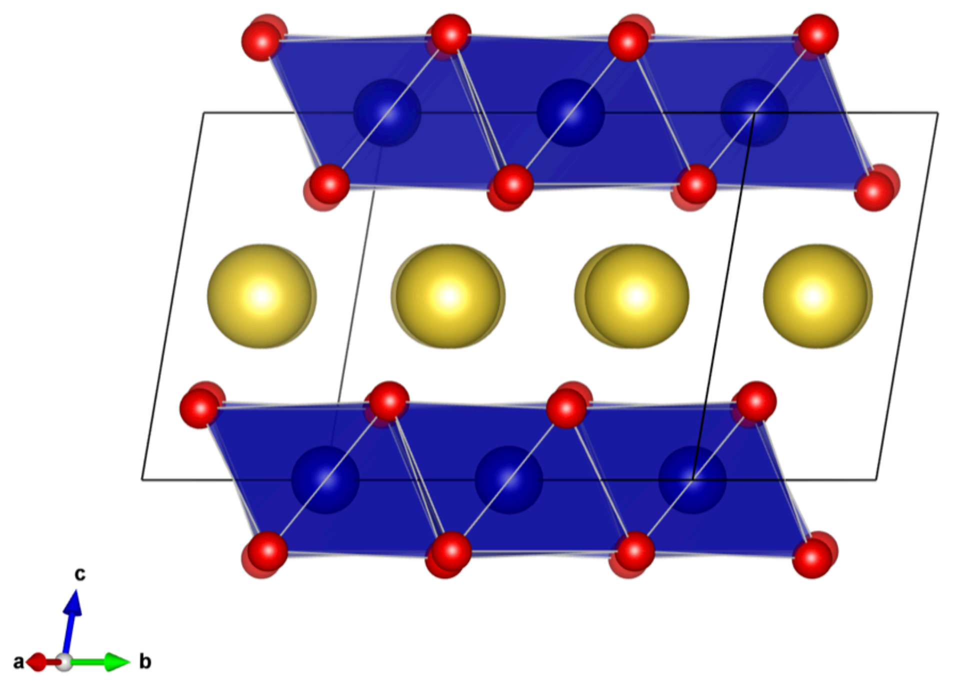
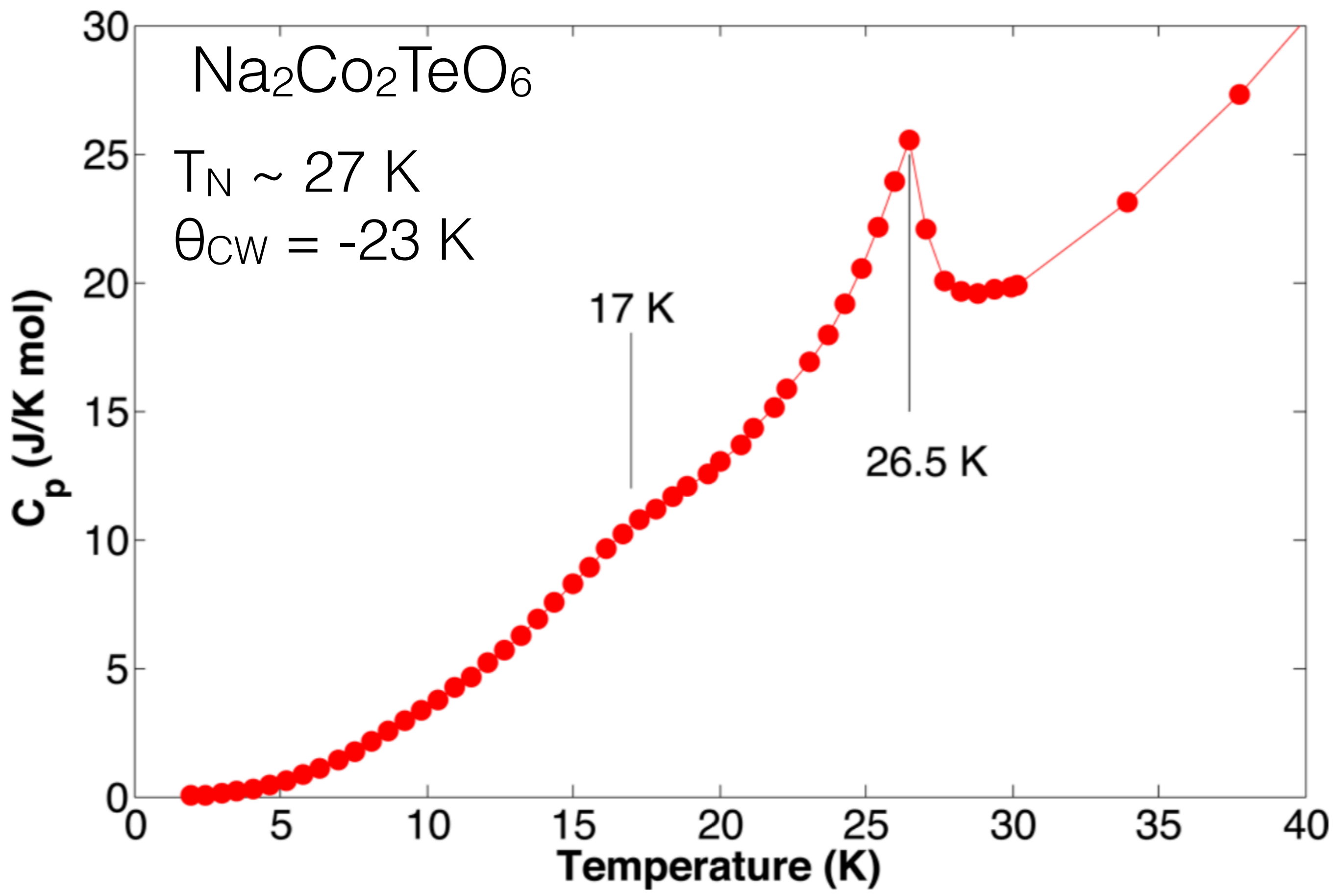


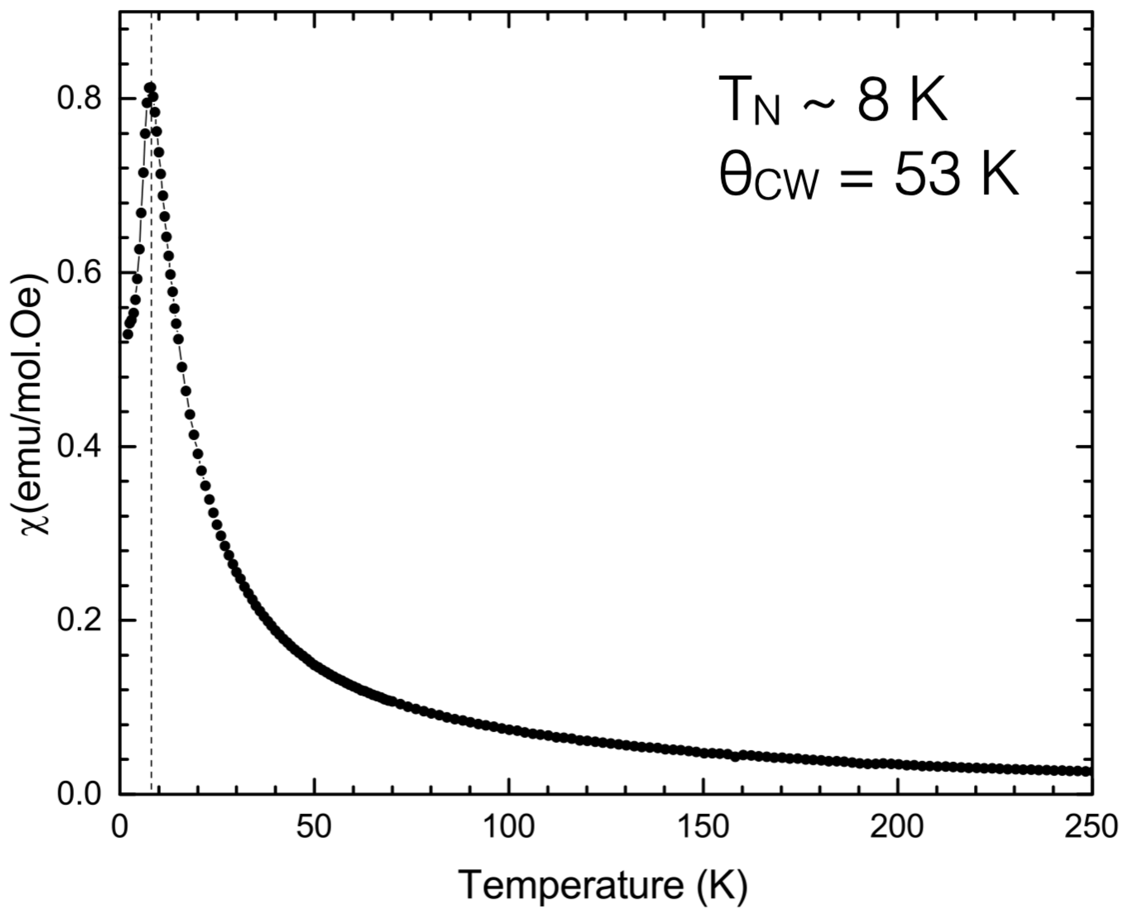
$Na_3Co_2SbO_6$

space group : C2/m (monoclinic)



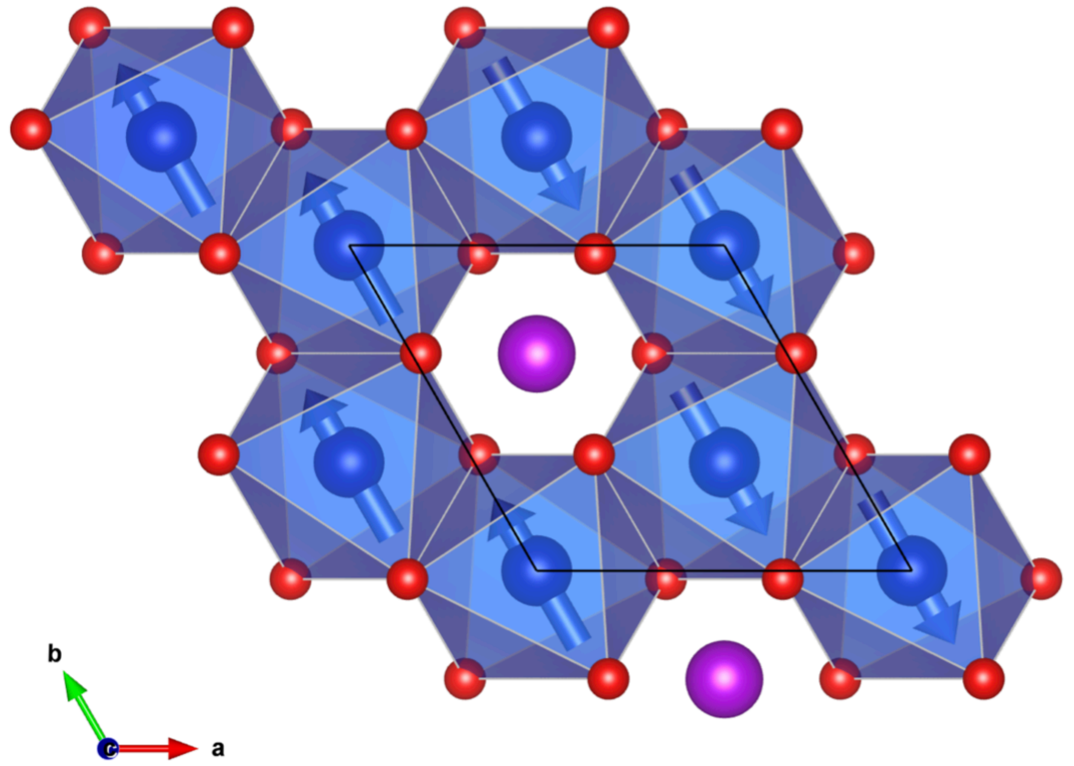
Magnetic properties





- Zigzag magnetic order at low temperature
- Anisotropic correlation lengths for $Na_2Co_2TeO_6$: short-range inter-plane correlations $J_{int}/J \sim 0.01$

Magnetic structure determined by neutron powder diffraction (D1B, ILL - France)



$Na_2Co_2TeO_6$
k = (1/2, 0, 0)

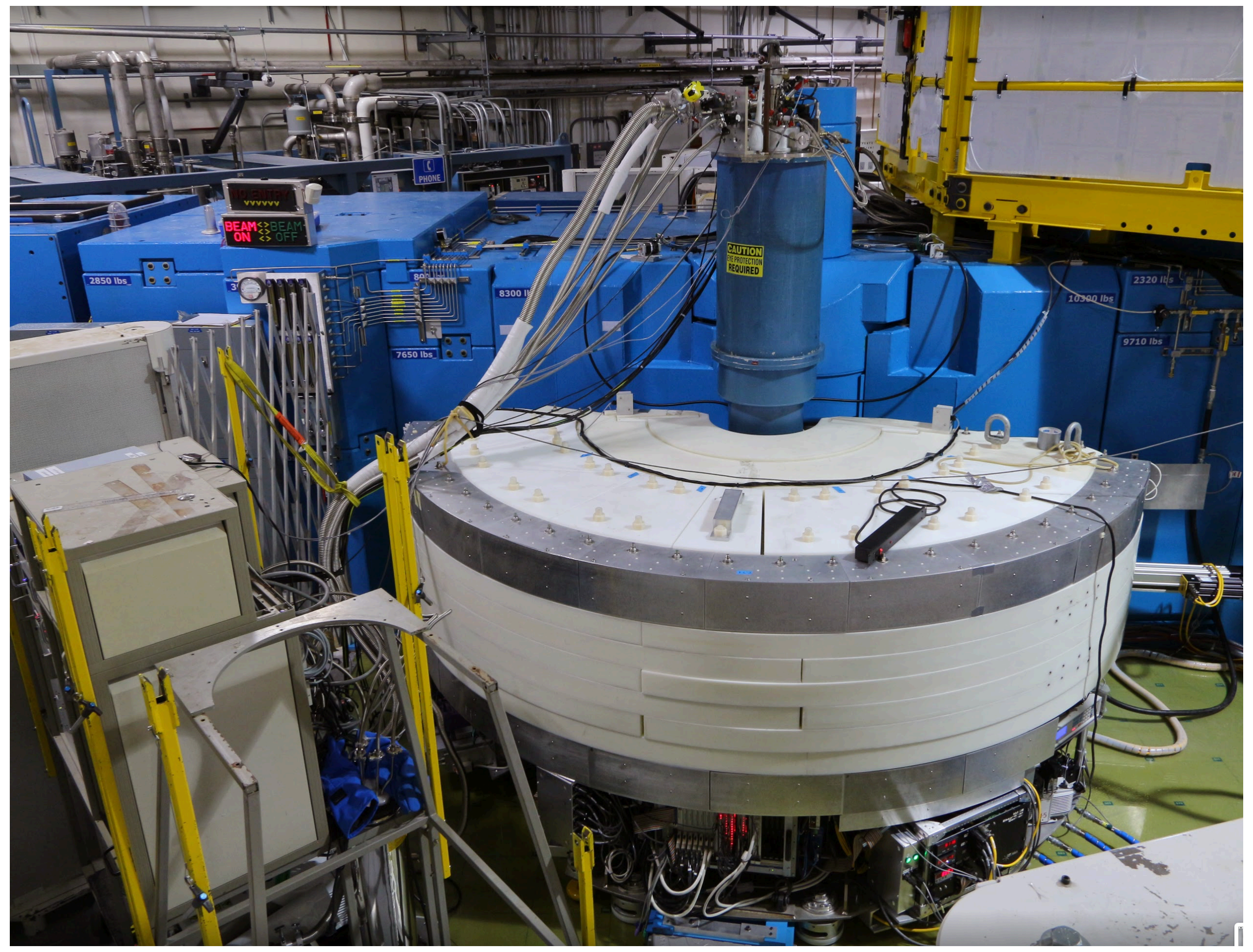
$Na_3Co_2SbO_6$
k = (1/2, 1/2, 0)

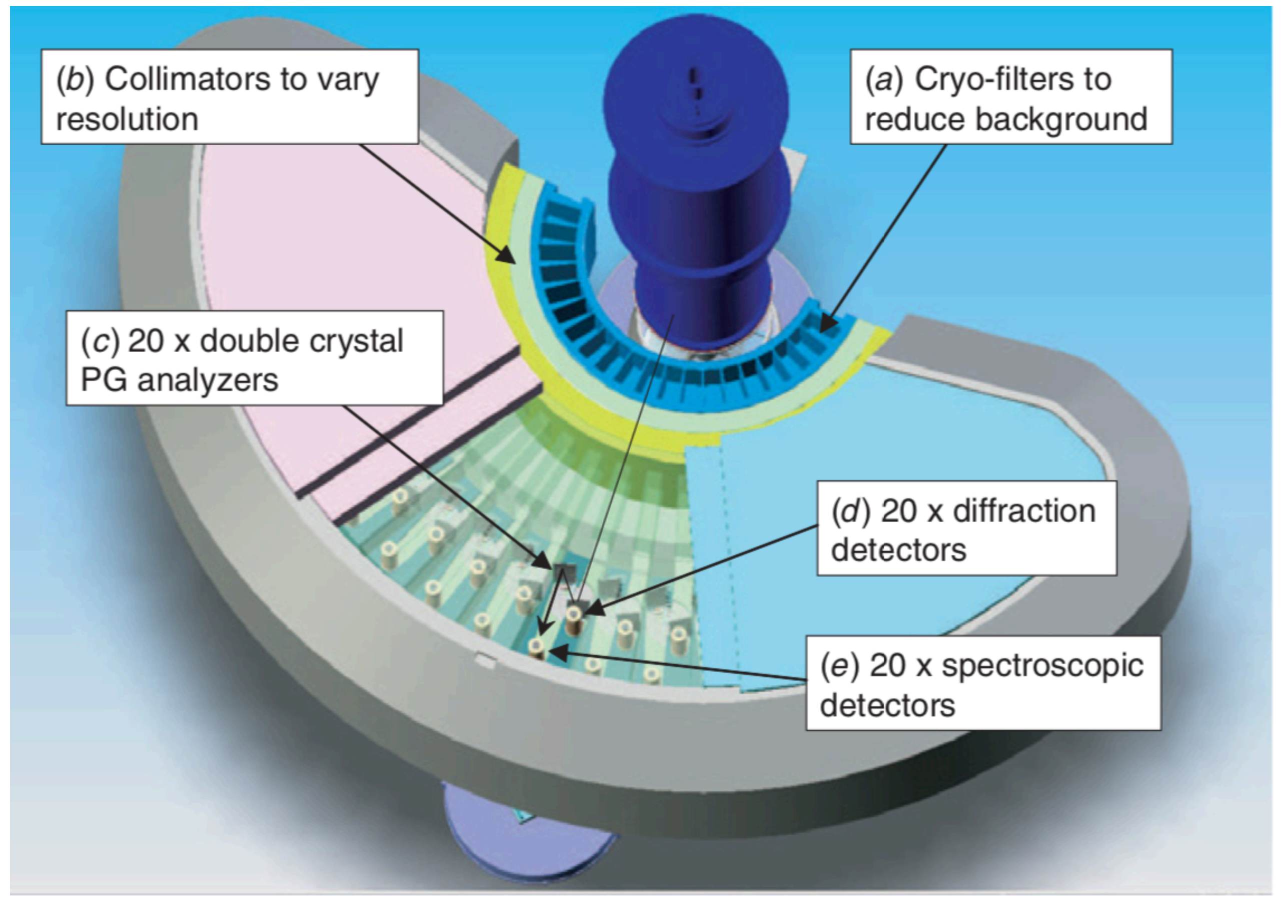
E. Lefrançois et al., Phys. Rev. B 94, 214416 (2016)
A. K. Bera et al., Phys. Rev. B 95, 094424 (2017)
C. Wong et al., J. Sol. State. Chem. 243 (2016)
J.-Q Yan et al., Phys. Rev. Mat. 3, 074405 (2019)

Neutron scattering experiments

NIST
National Institute of Standards and Technology

MACS: Multi-Axis Crystal Spectrometer





Neutron scattering, an ideal tool for magnetic excitations:

- No charge : probes the bulk and not the surface
- Interaction with matter : nuclear and magnetic contribution
- Energy of thermal neutrons : few meV
- Cross-section directly proportional to spin-spin correlation function
- Allows measurements throughout the reciprocal space

Momentum and energy conservation

$$\vec{Q} = \vec{k_i} - \vec{k_f}$$

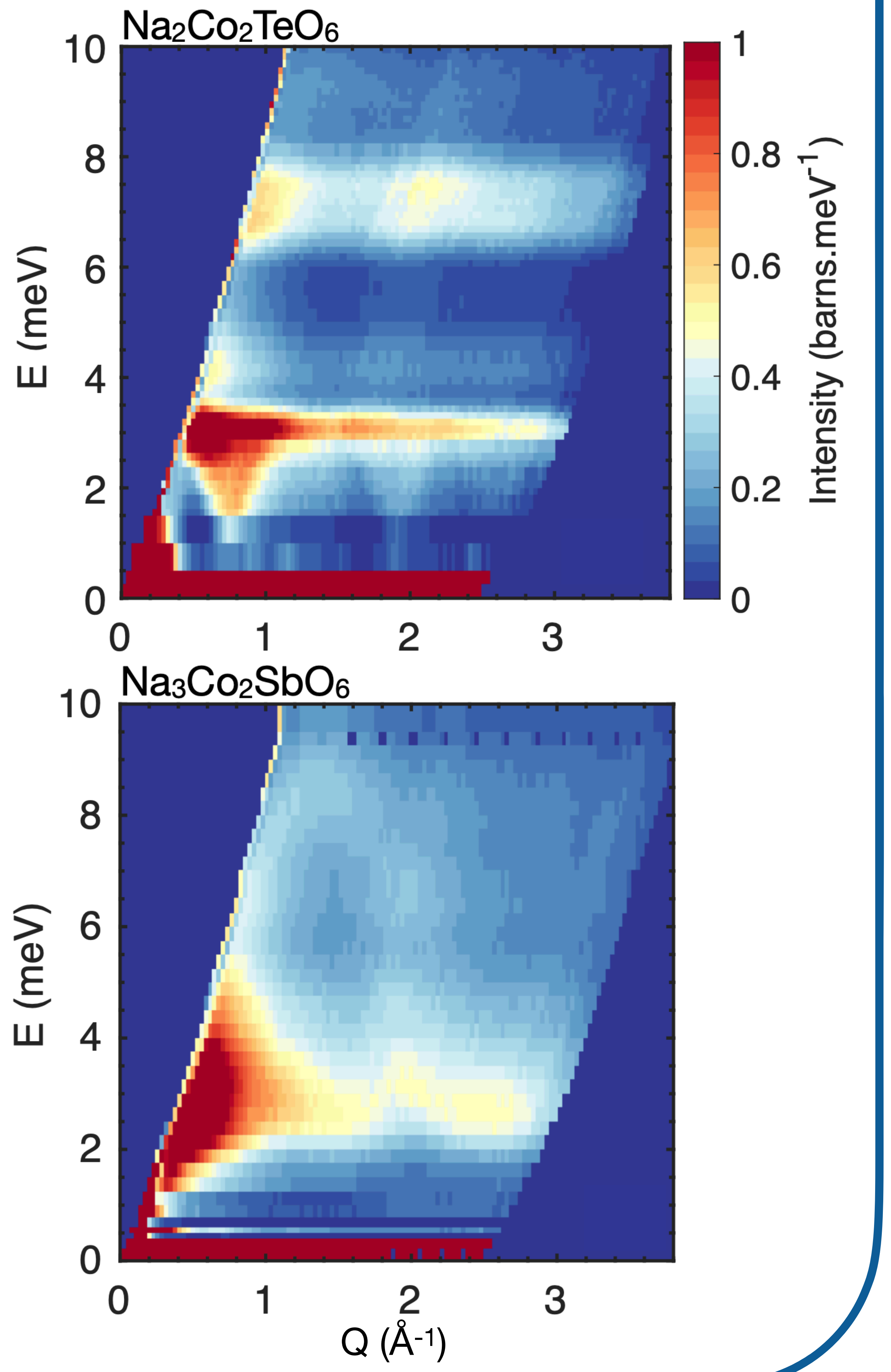
$$E = \hbar\omega = E_i - E_f = \frac{\hbar^2}{2m}(k_i^2 - k_f^2)$$

Neutron inelastic study (powder)

Low Energy:

- Excitations within the j_{eff} = 1/2 manifolds : exchange interactions

MACS (NIST), E_f = 5 meV
T = 1.5 K



Testing the spin model : spin wave calculations

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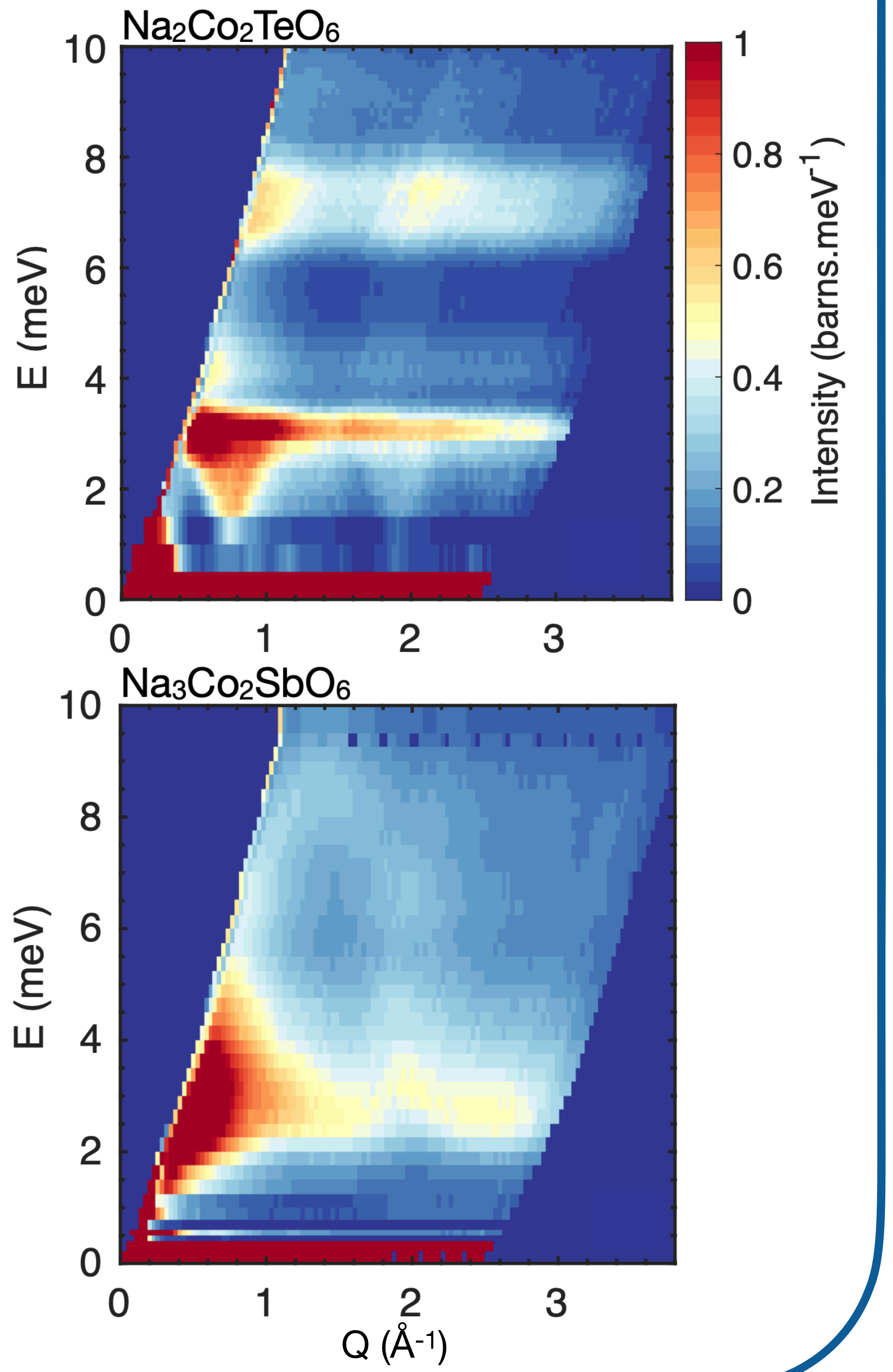
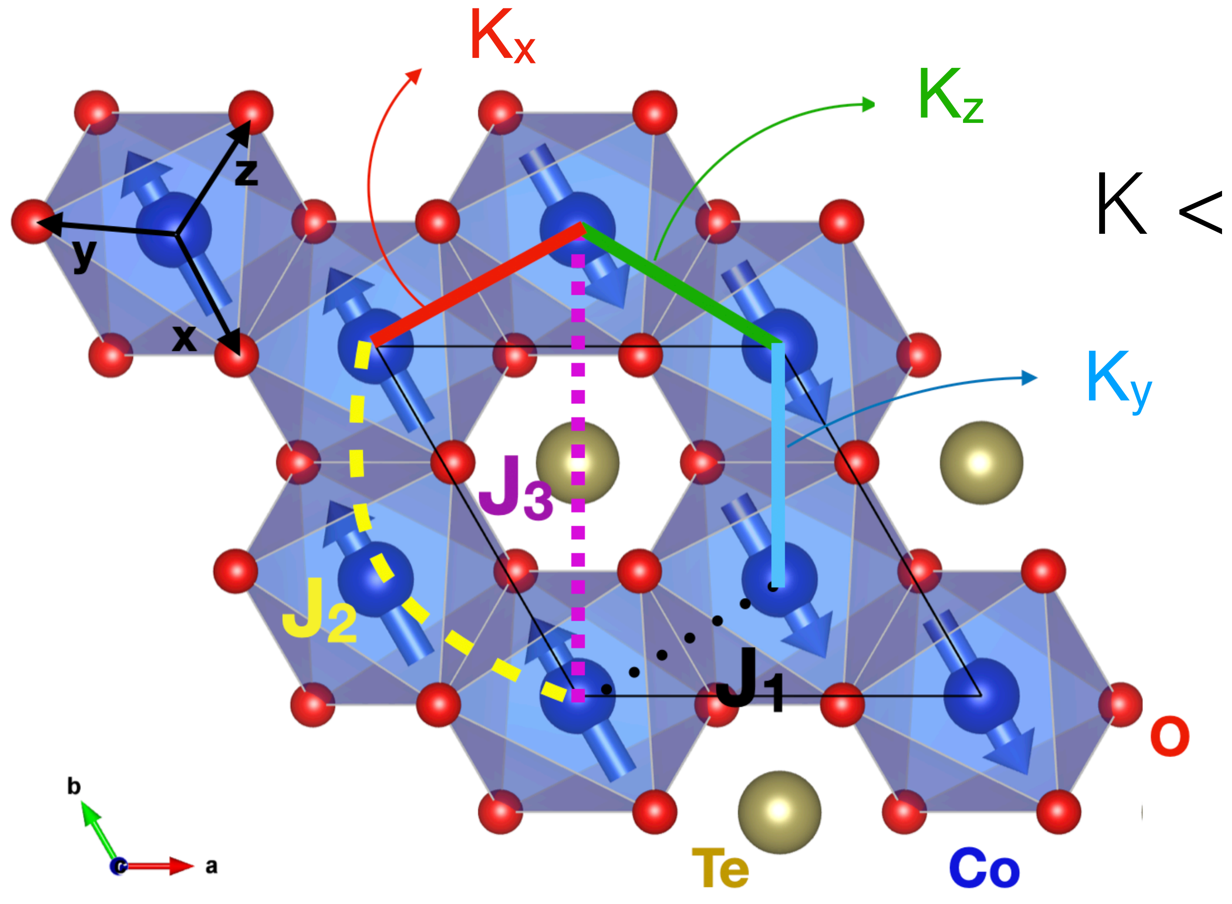


Spin Hamiltonian : Kitaev-Heisenberg model



K < 0 (ferro)

$$H_{K-H} = \sum_{n=1}^{3} J_n \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i,j} K S_i^\gamma S_j^\gamma + \sum_{i,j} \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \sum_{i,j} \Gamma' (S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\beta S_j^\gamma + S_i^\gamma S_j^\beta),$$

$$\{\alpha, \beta, \gamma\} = \{y, z, x\}, \{z, x, y\}, \{x, y, z\}$$

H. Liu et al., Phys. Rev. Letters 125, 047201 (2020)

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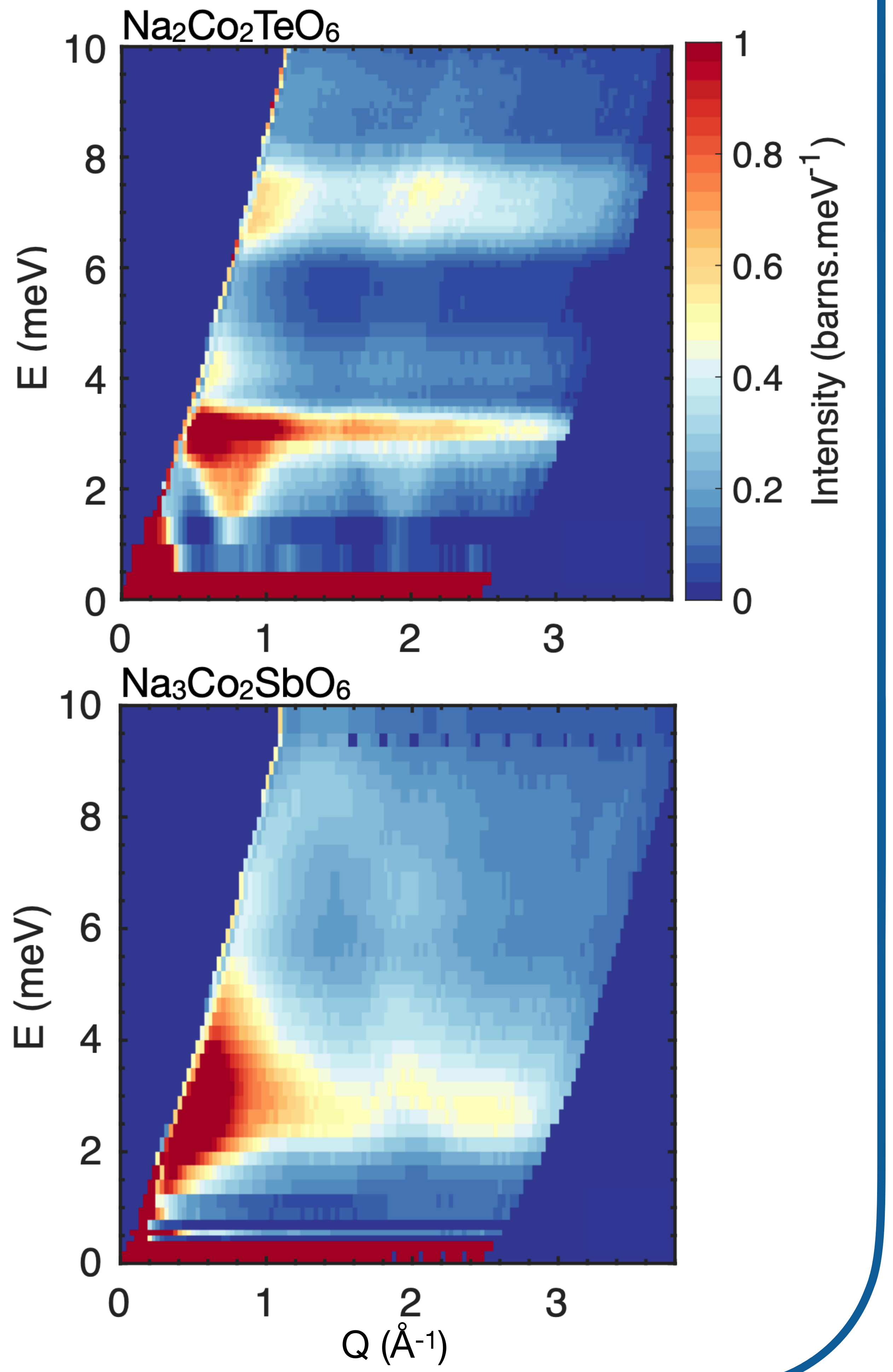

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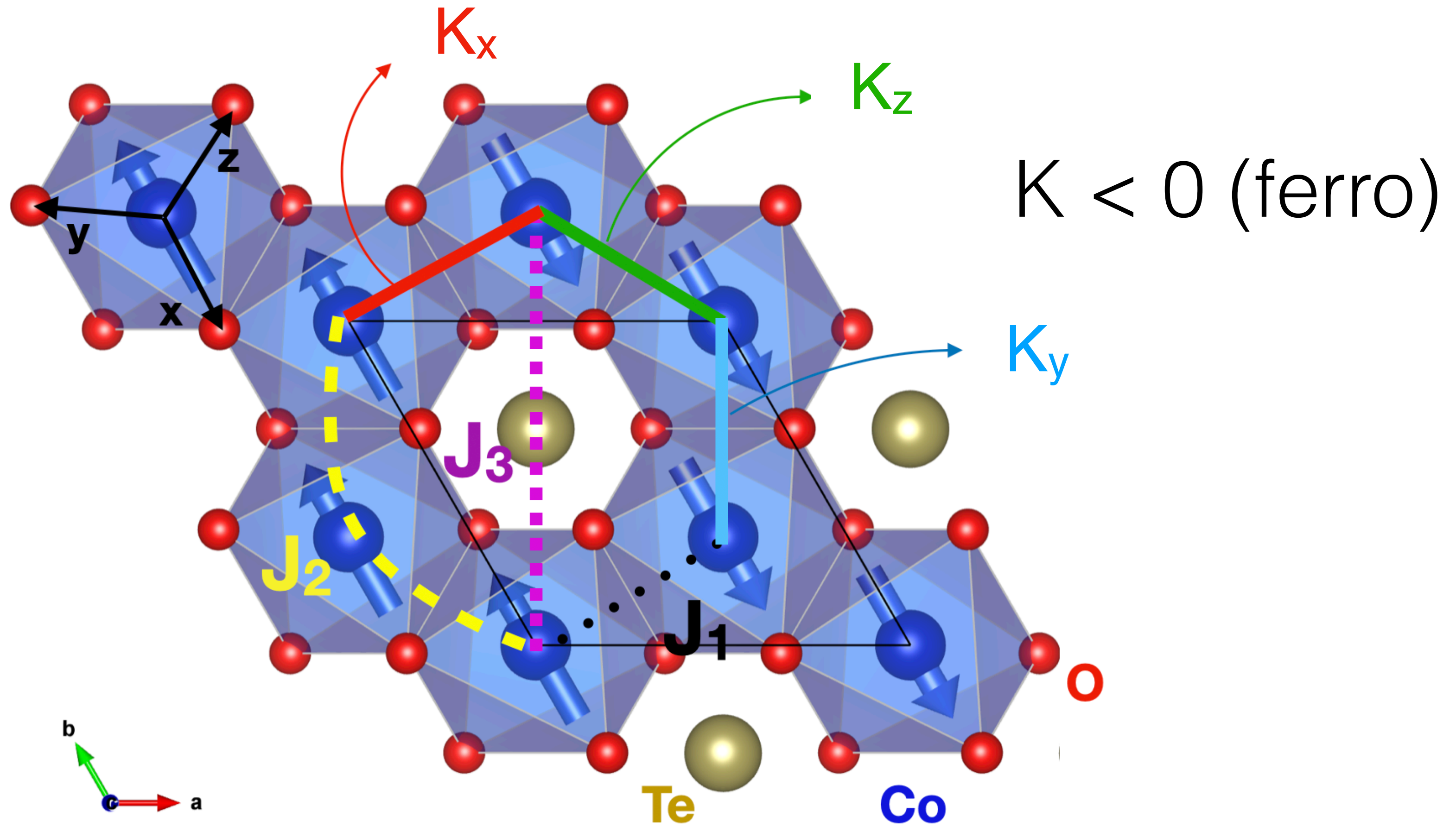
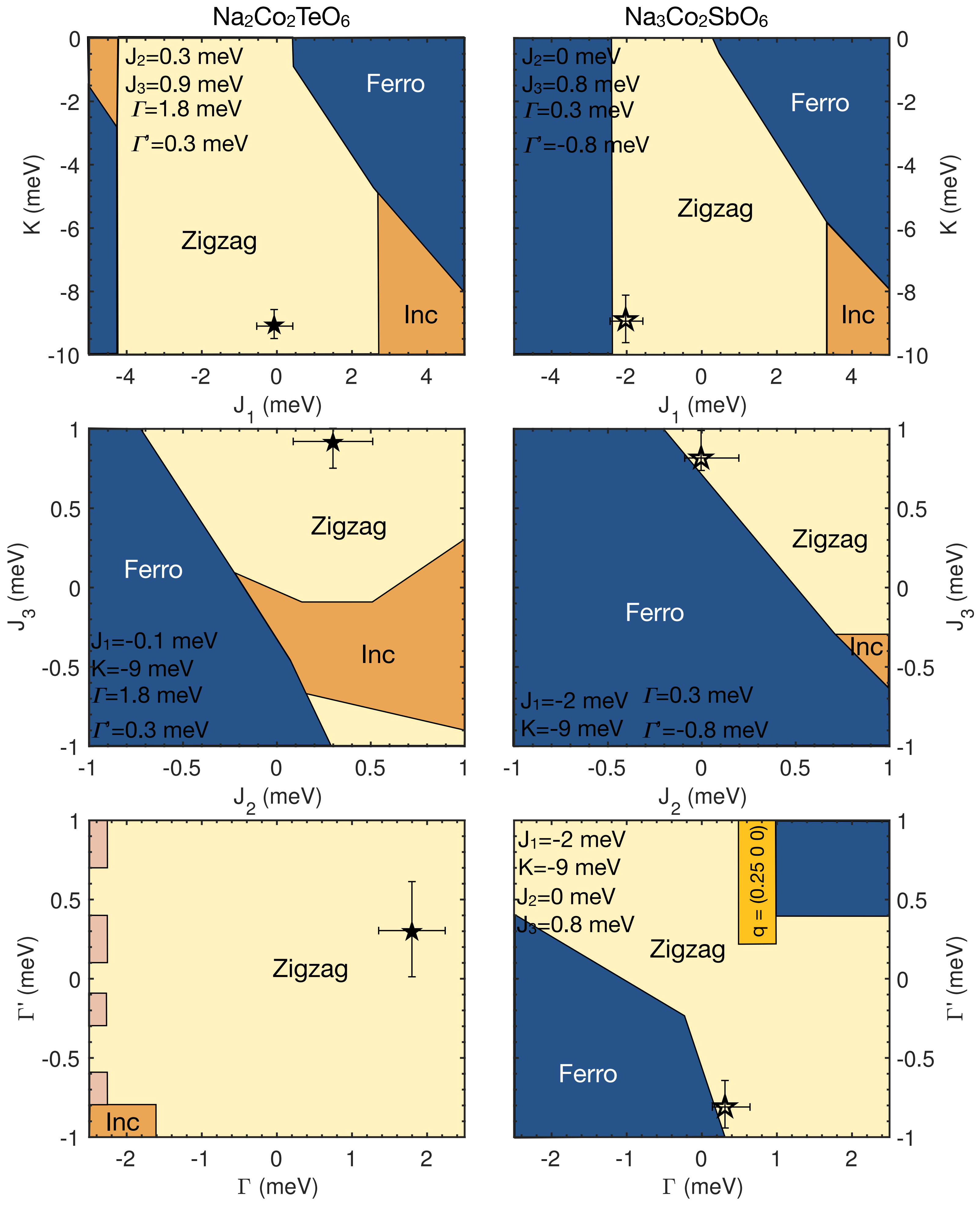


$$H_{K-H} = \sum_{n=1}^{3} J_n \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i,j} K S_i^{\gamma} S_j^{\gamma}$$
$$+ \sum_{i,j} \Gamma (S_i^{\alpha} S_j^{\beta} + S_i^{\beta} S_j^{\alpha})$$
$$+ \sum_{i,j} \Gamma' (S_i^{\alpha} S_j^{\gamma} + S_i^{\gamma} S_j^{\alpha} + S_i^{\beta} S_j^{\gamma} + S_i^{\gamma} S_j^{\beta}),$$

$$\{\alpha, \beta, \gamma\} = \{y, z, x\}, \{z, x, y\}, \{x, y, z\}$$

H. Liu et al., Phys. Rev. Letters 125, 047201 (2020)

Phase diagram as function of exchange parameters



Testing the spin model : spin wave calculations

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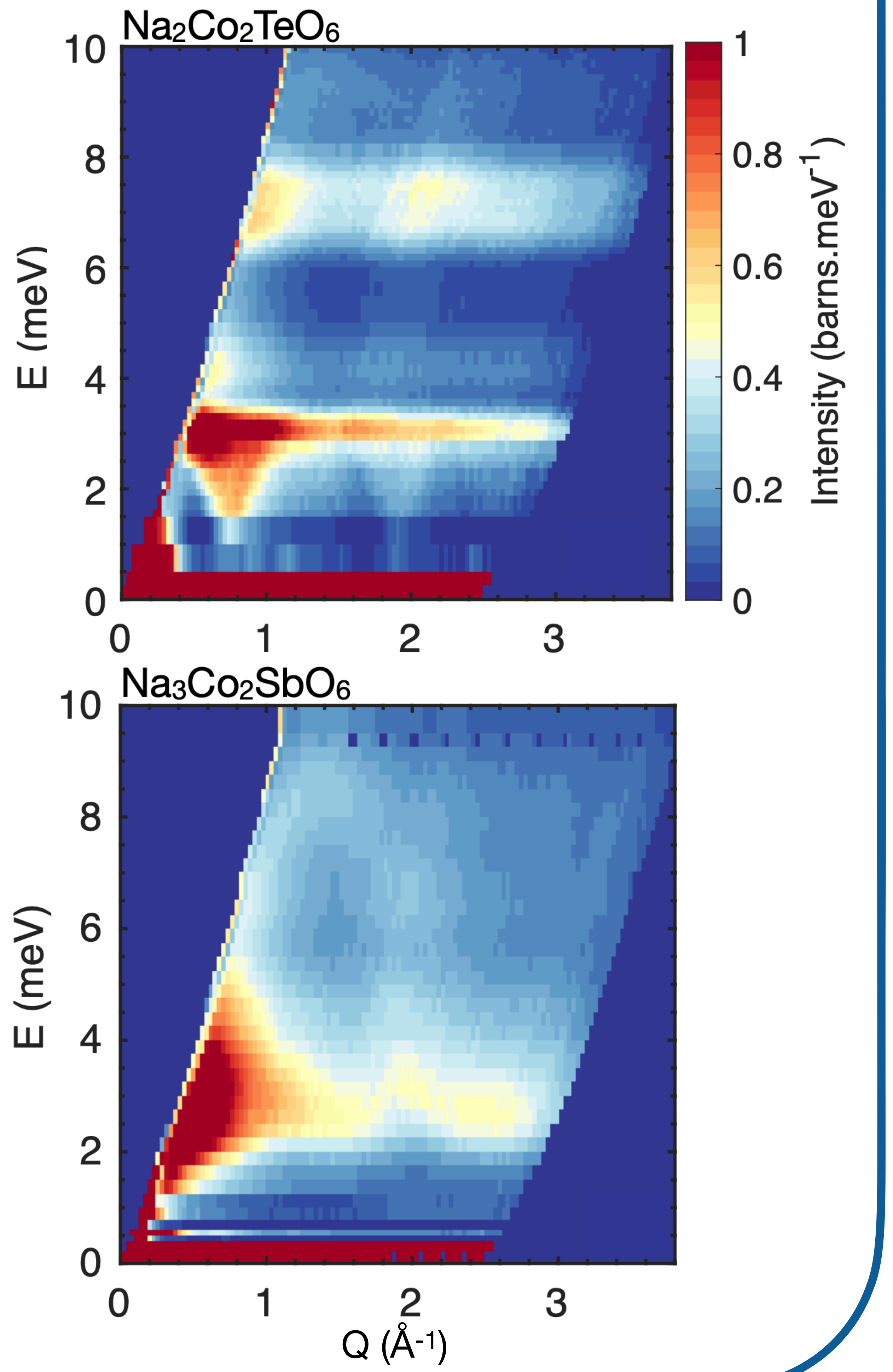
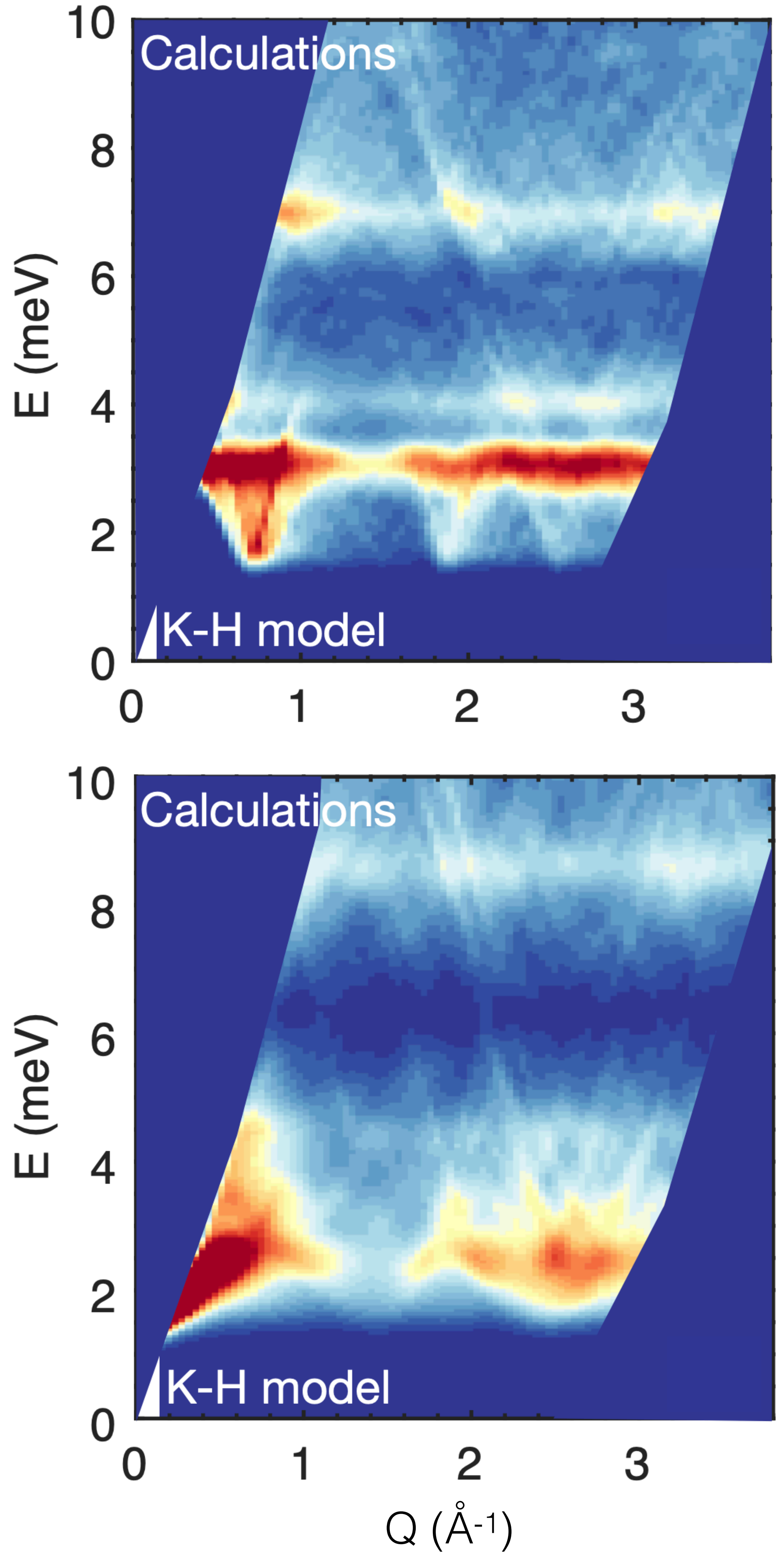


Spin wave calculations

Spinwave software
S. Petit, Collection SFN 12, 105 (2011)



	J_1	J_2	J_3	K	Γ	Γ'
$Na_2Co_2TeO_6$	−0.1(5)	0.3(3)	0.9(3)	−9.0(5)	1.8(5)	0.3(3)
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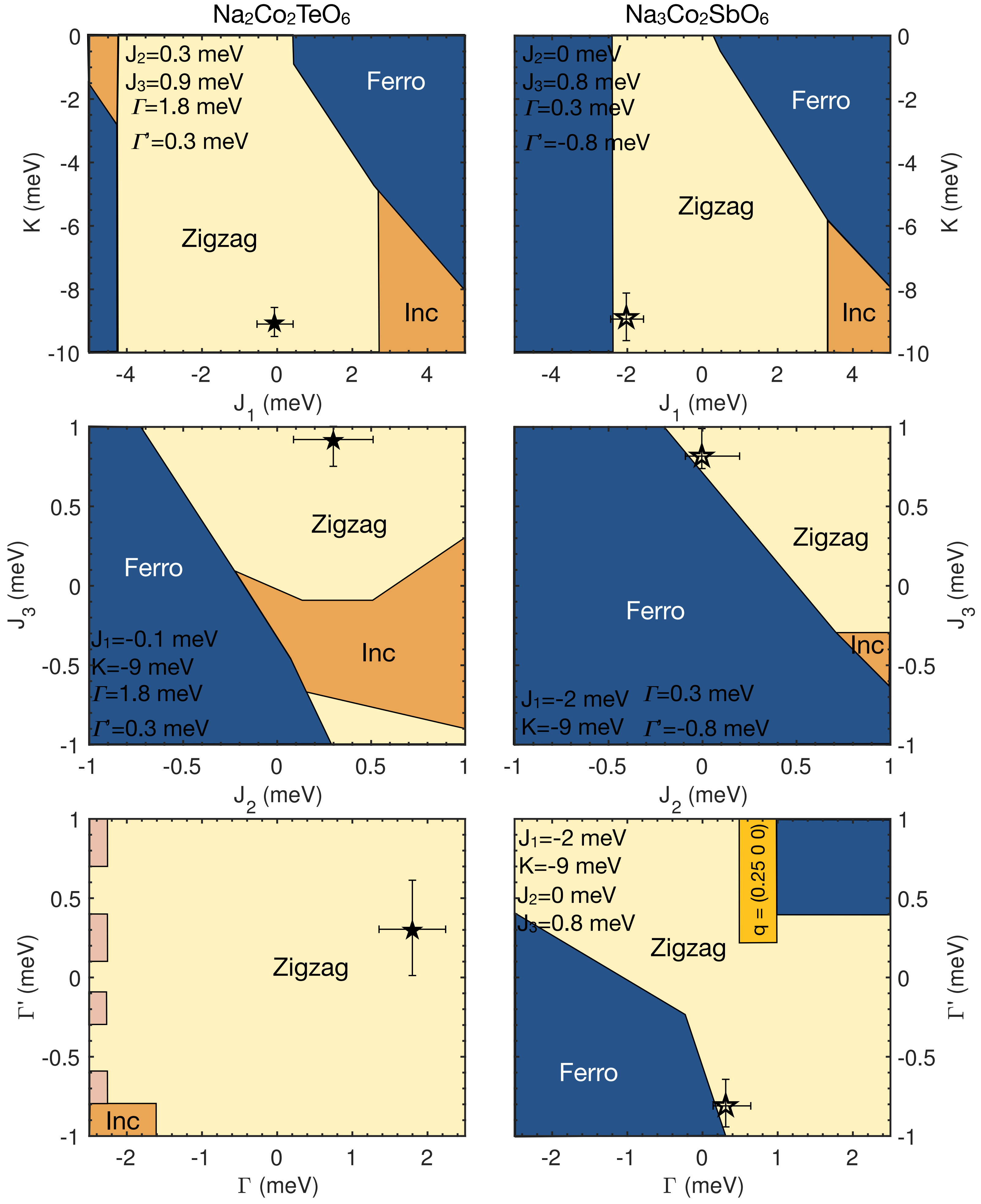

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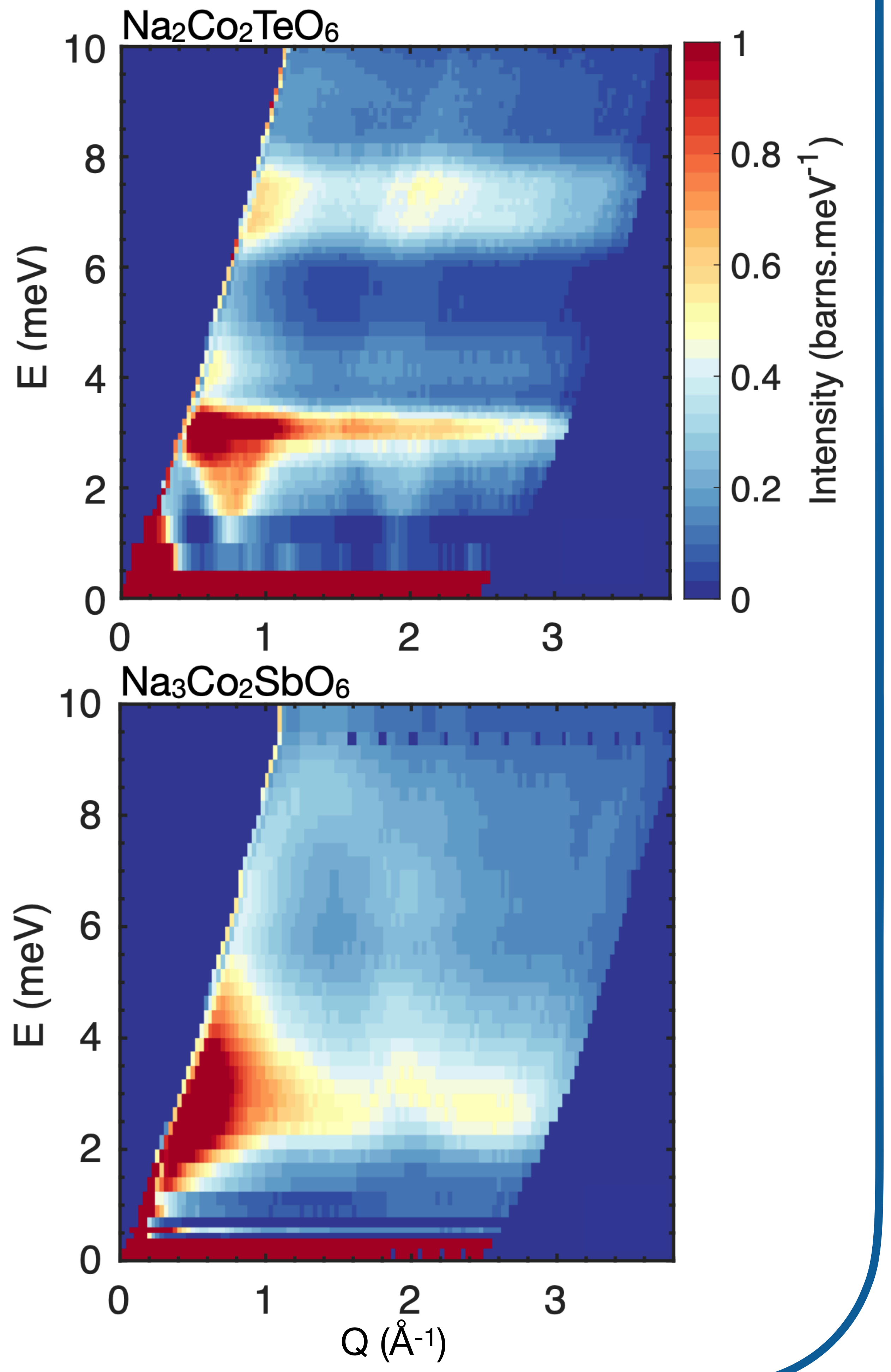

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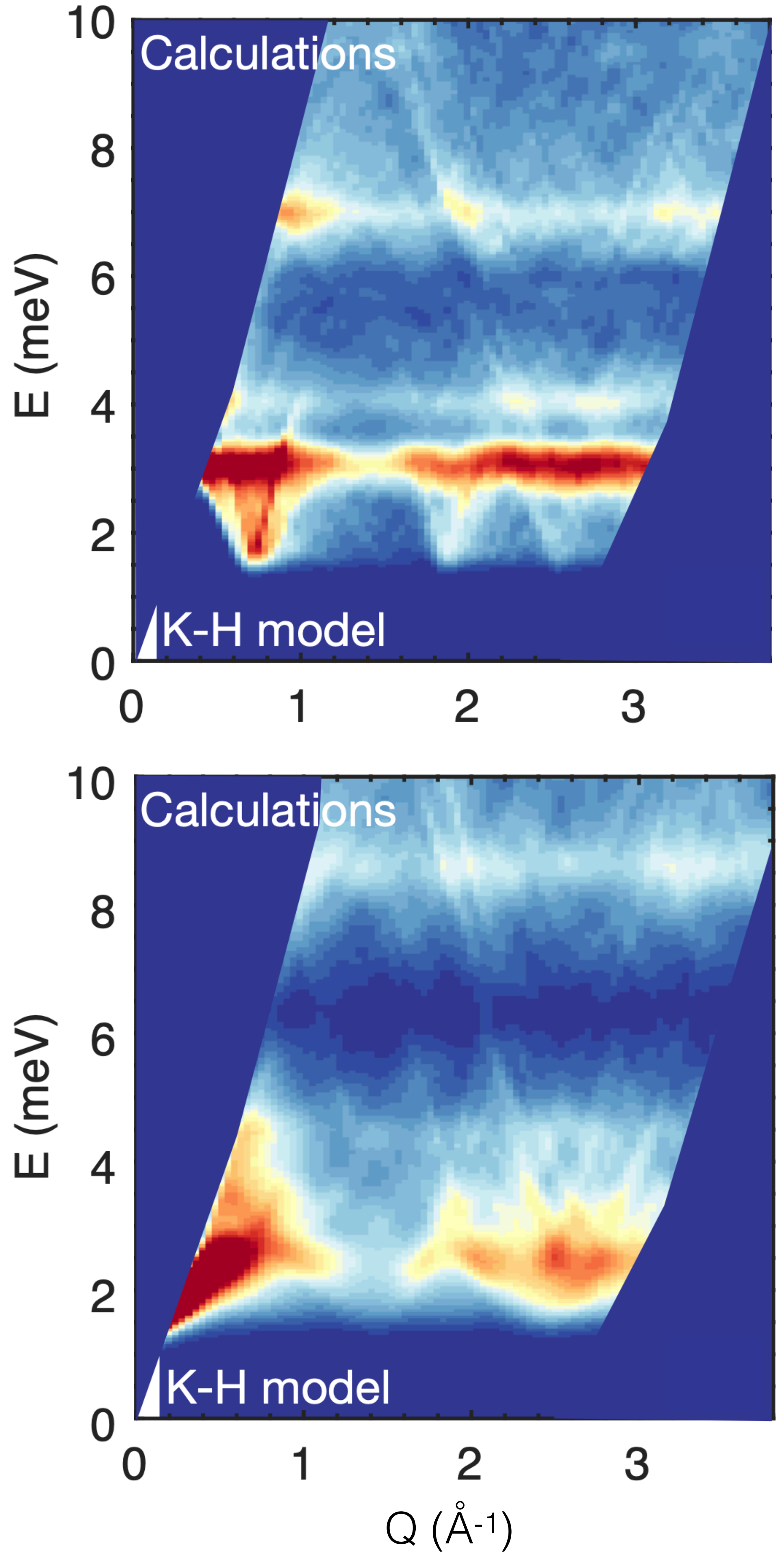

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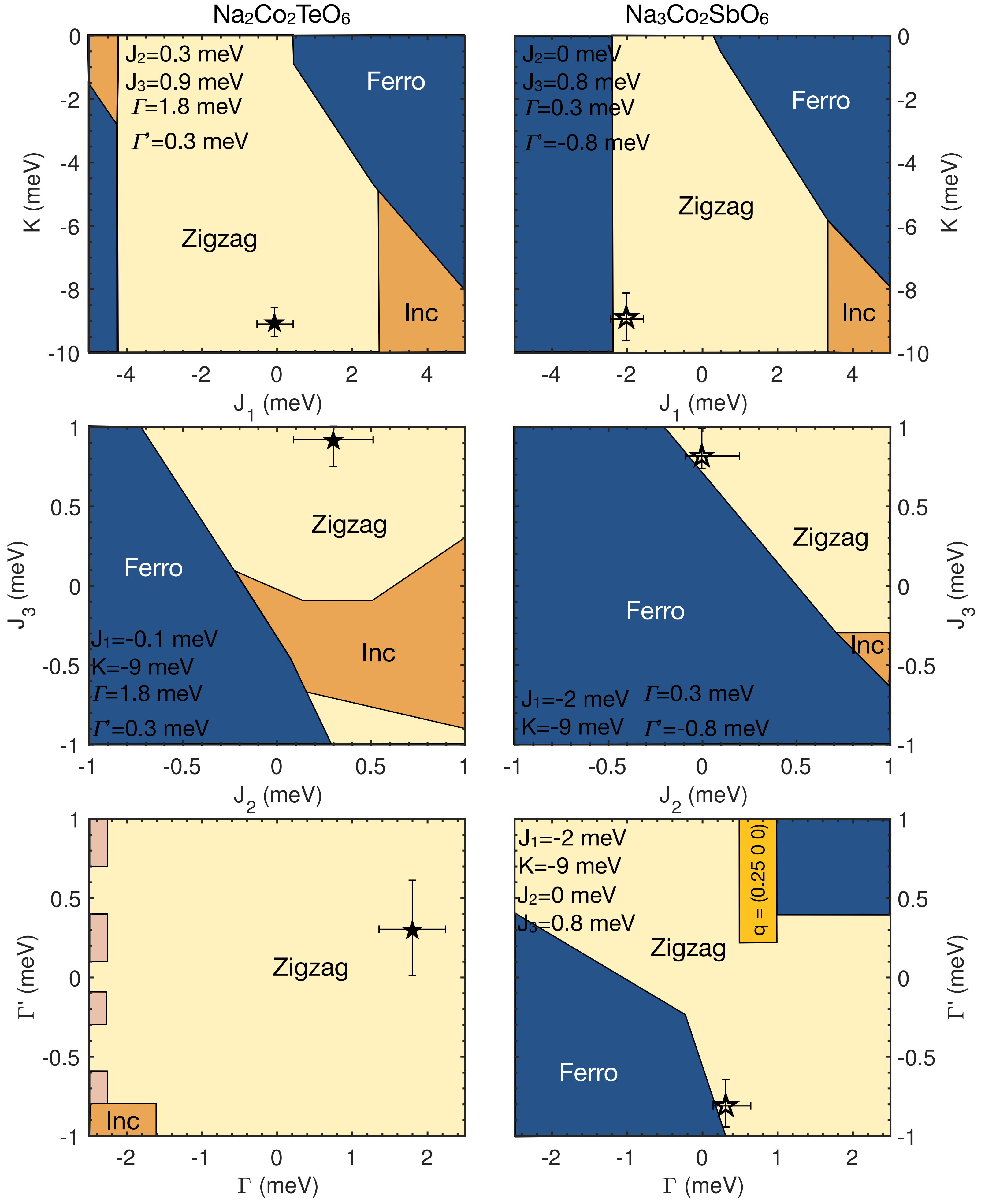

Comparison with theory: $Na_3Co_2SbO_6$

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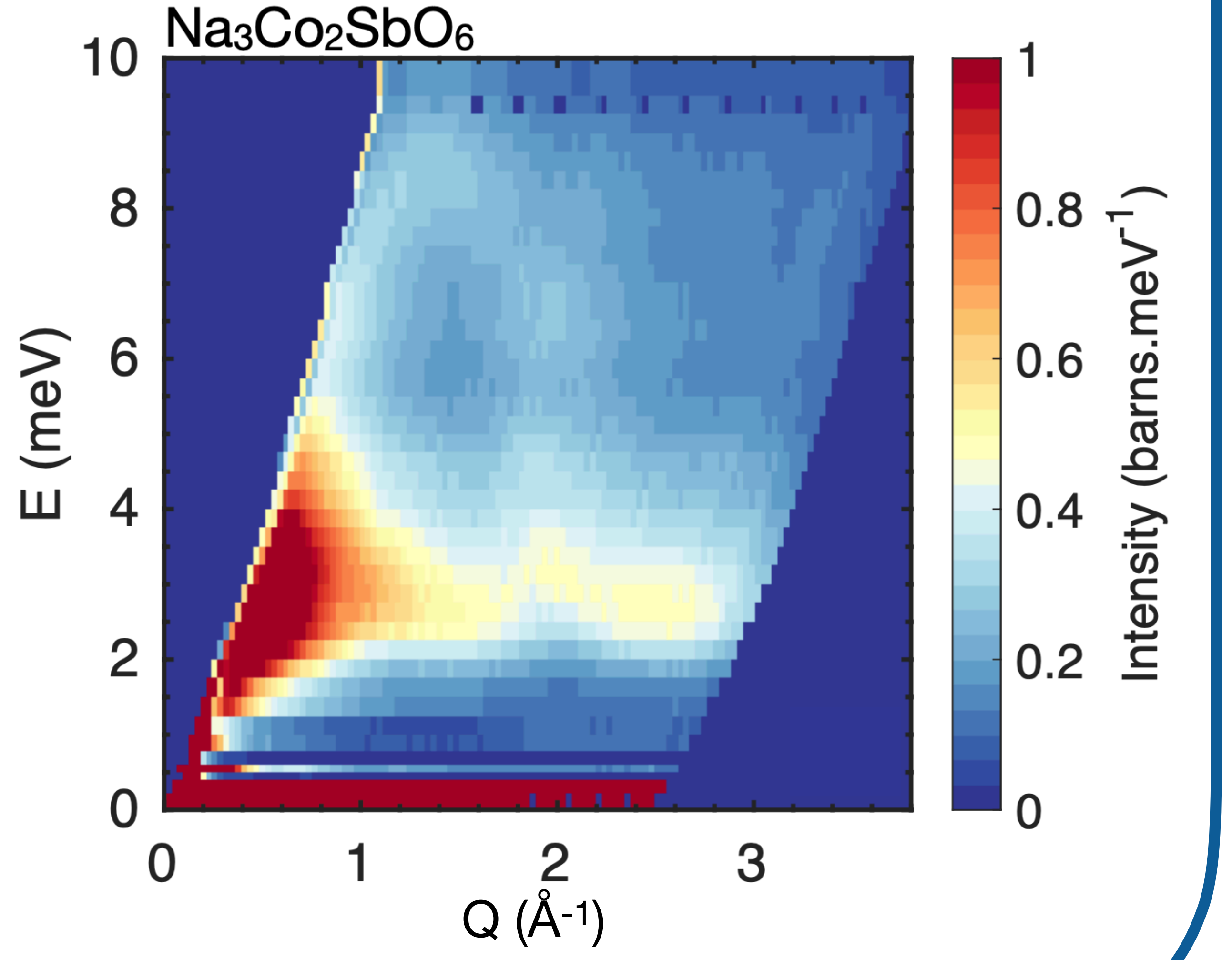
NIST National Institute of Standards and Technology



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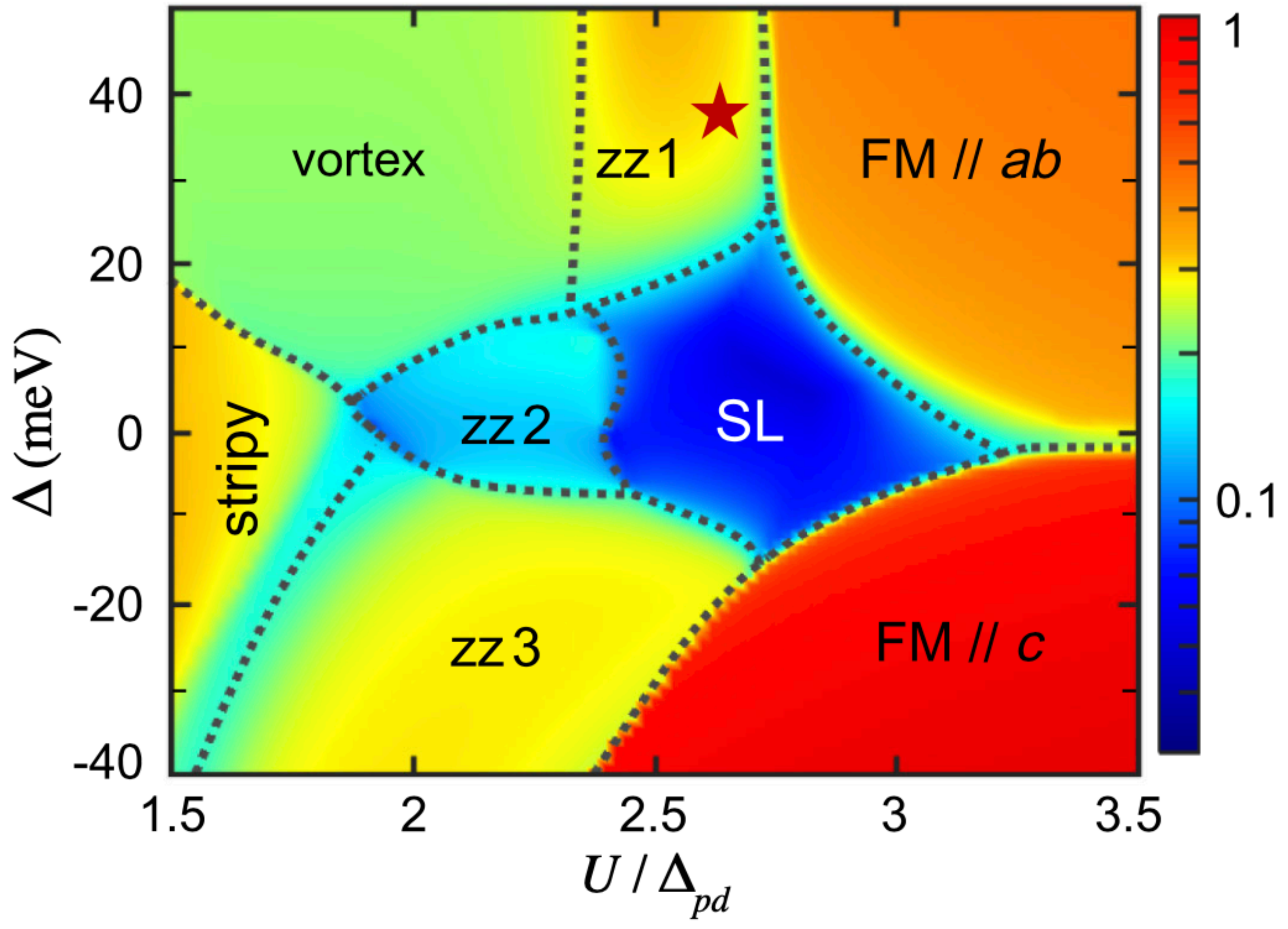
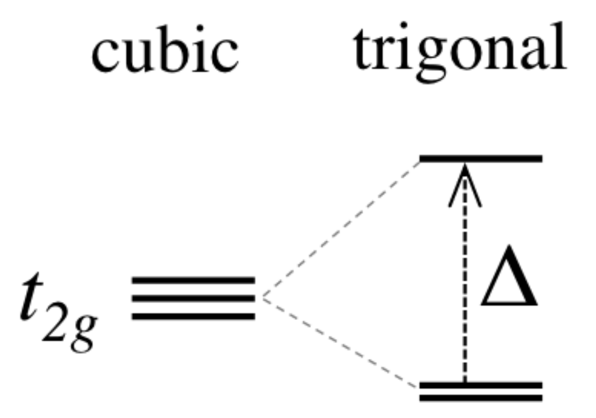
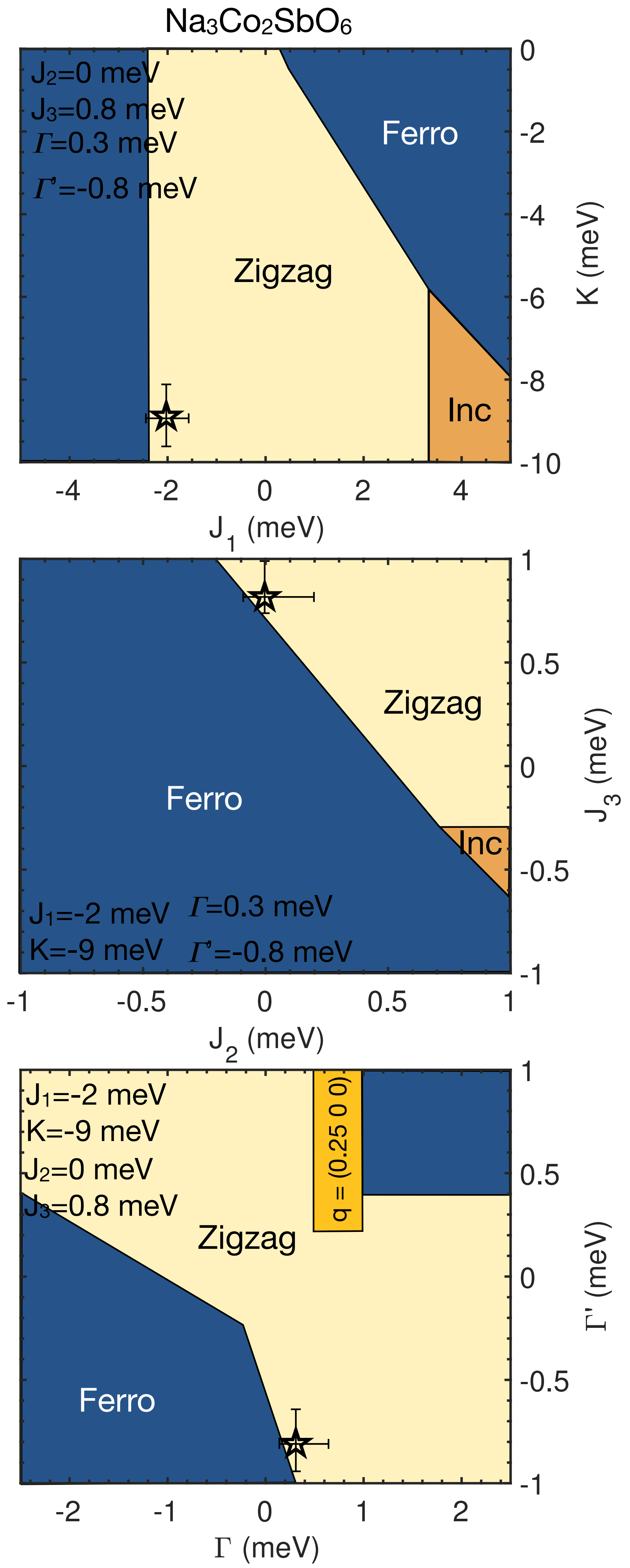
H. Liu et al., Phys. Rev. Letters 125, 047201 (2020)



Trigonal field
(octahedral distortion)



Our work



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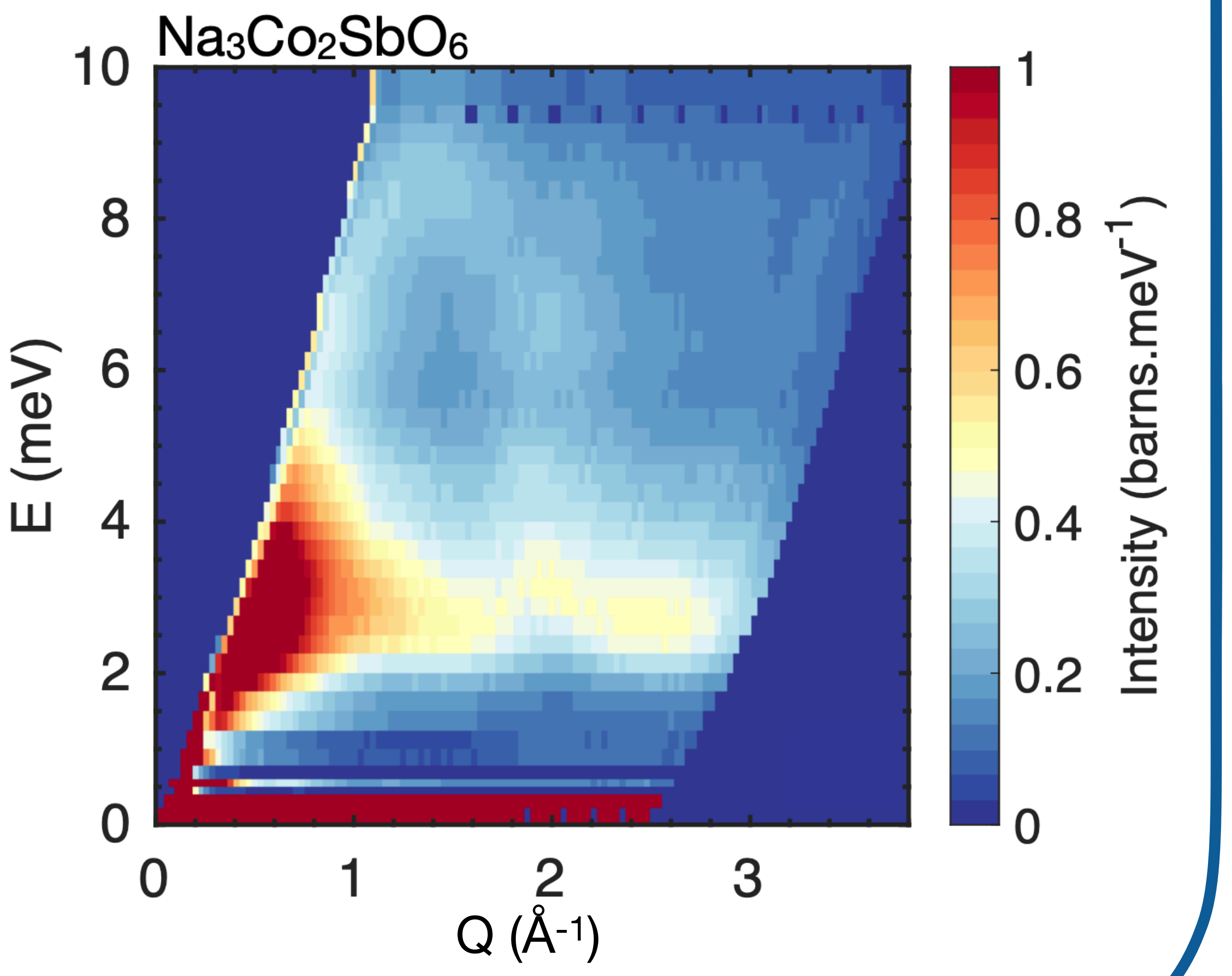

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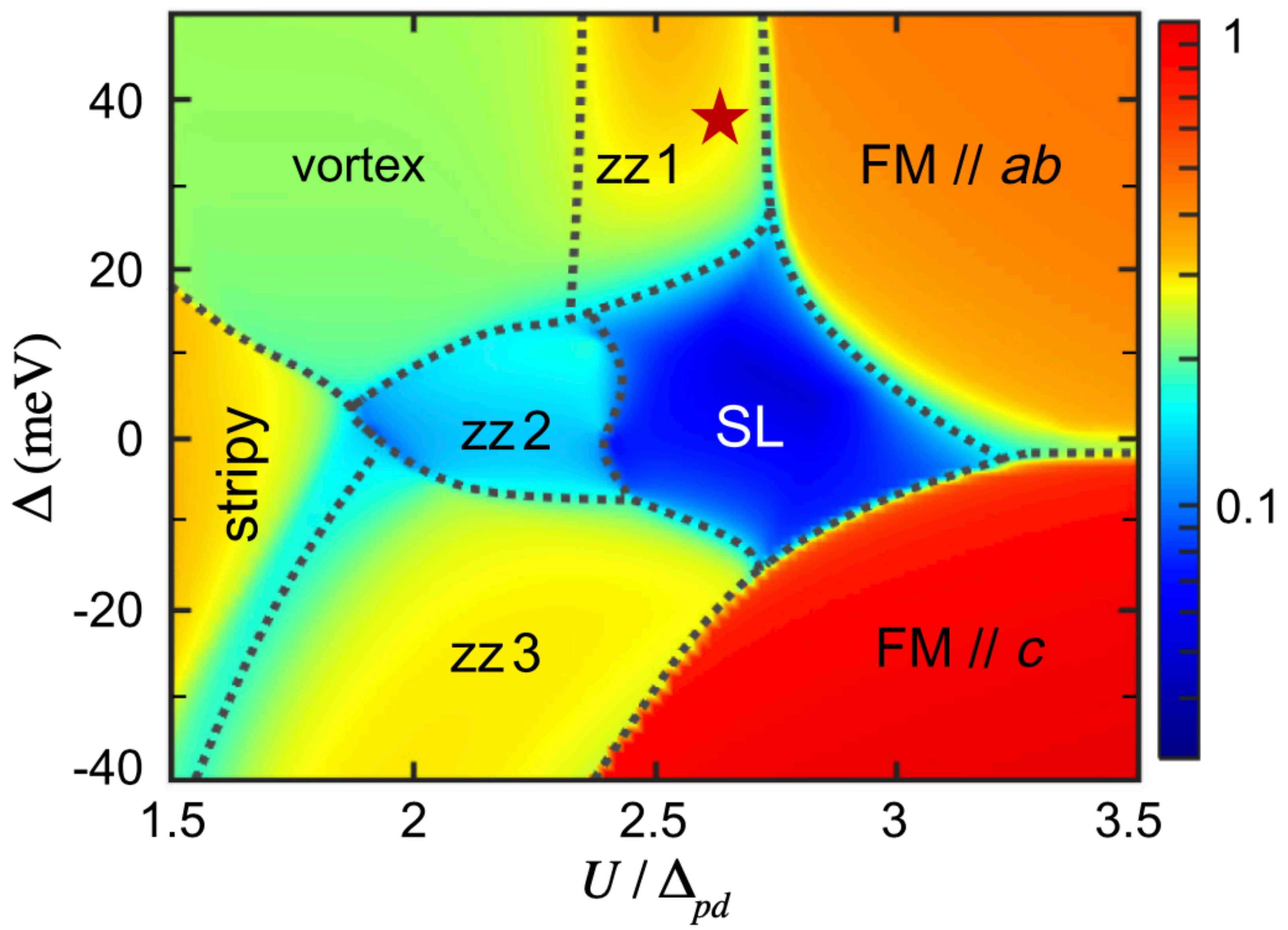

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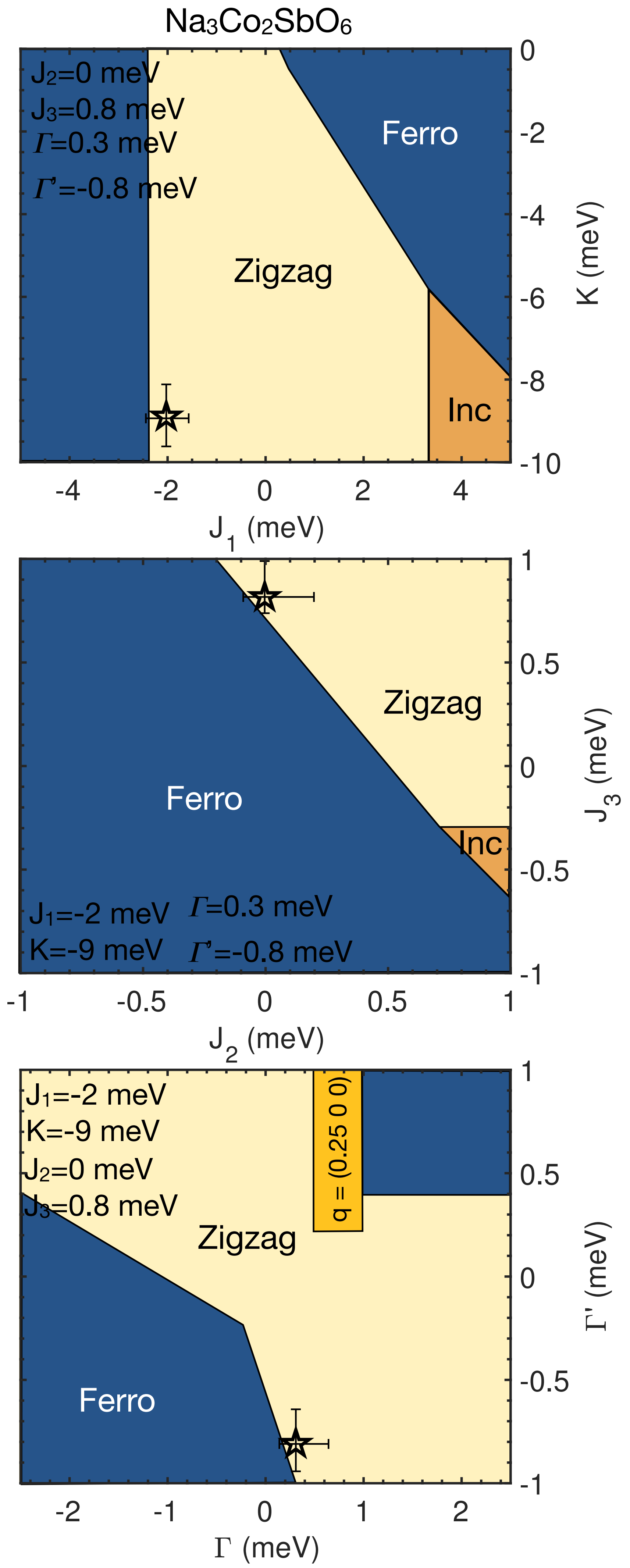


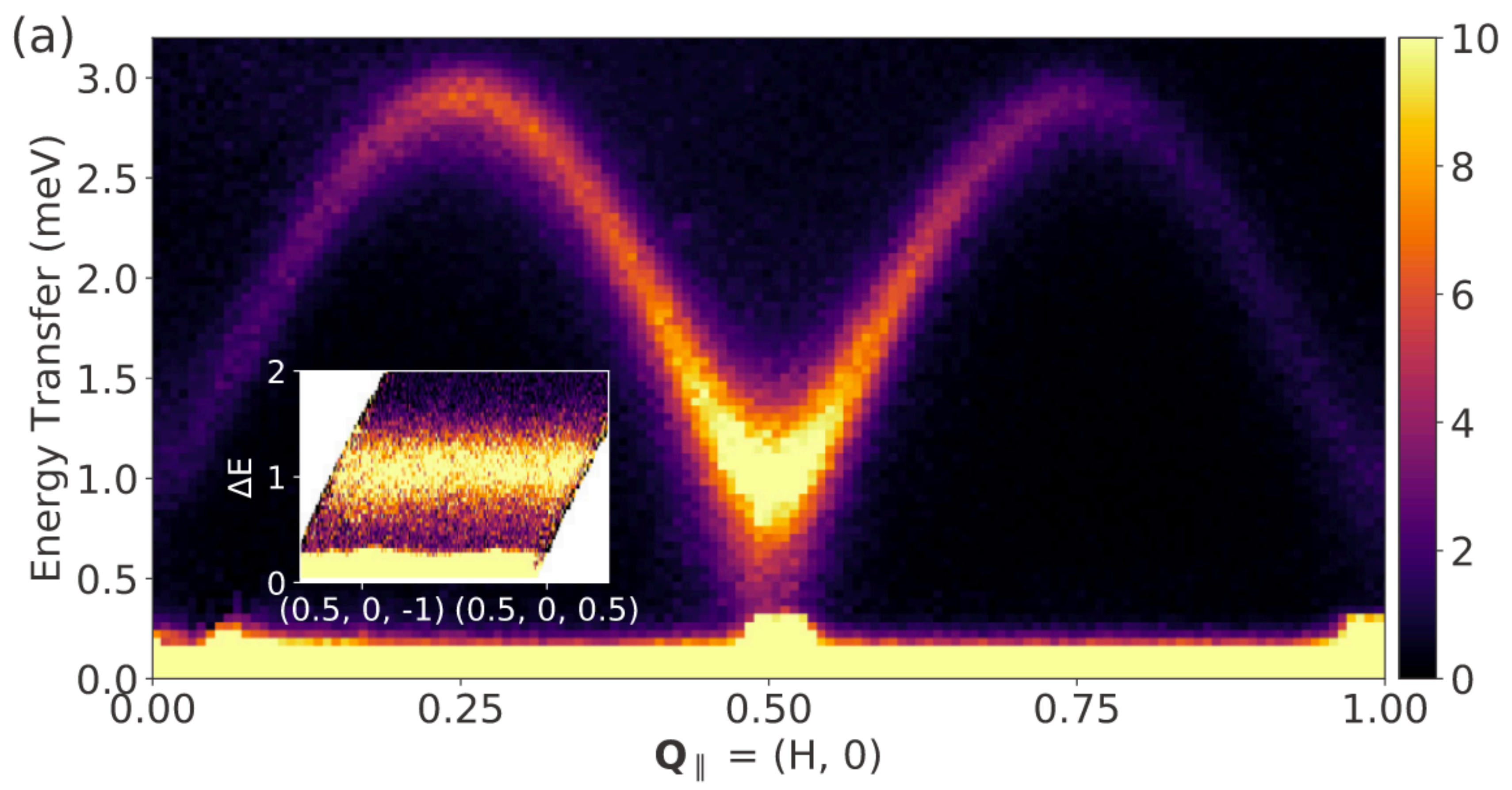
Confirmation in single crystal materials is essential !!

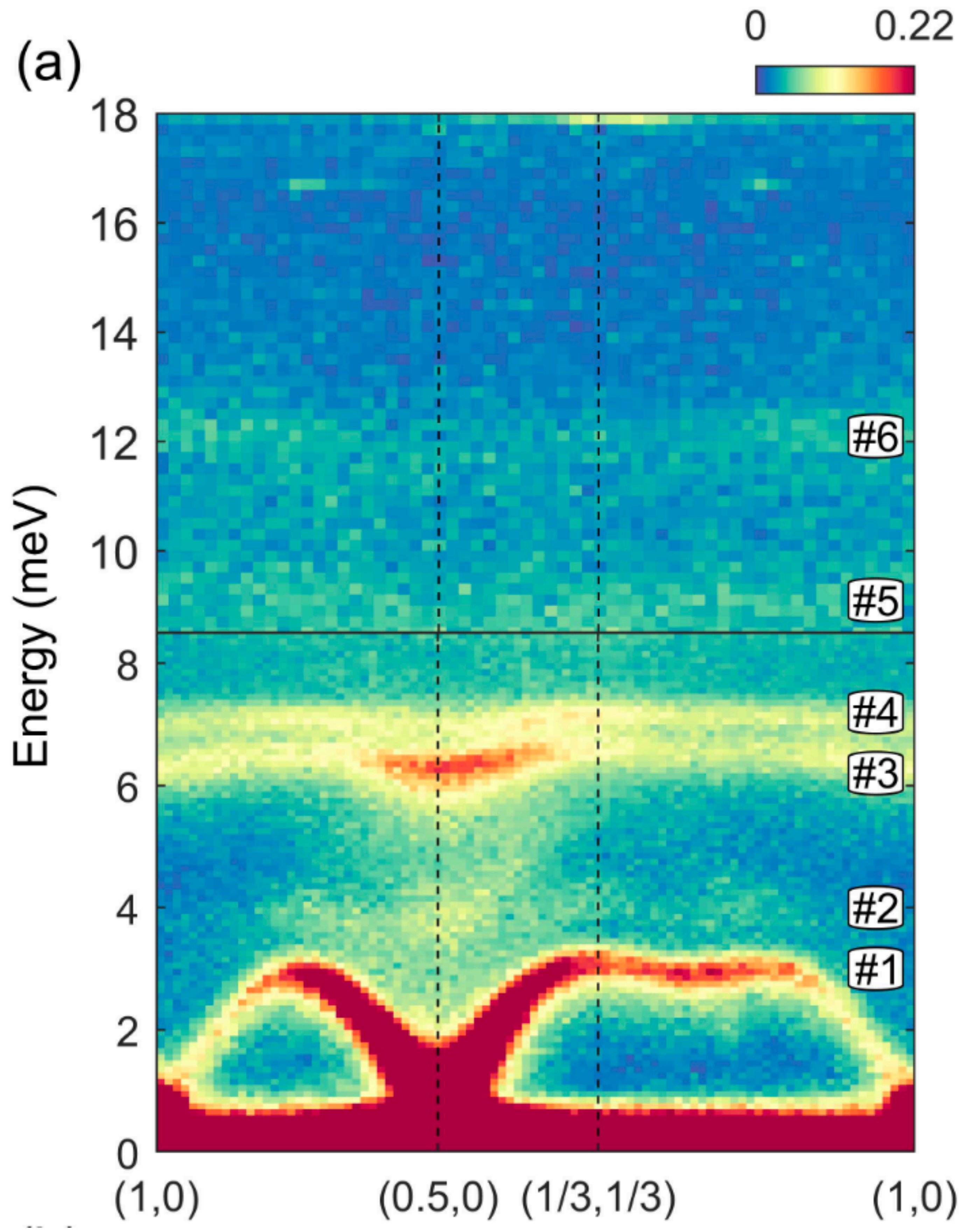
Theory	J/|K| ~ -0.14	Γ/|K| ~ -0.03	Γ'/|K| ~ -0.16
Experiment (our work)	1/2*(J_1/J_3)/|K| ~ -0.15	Γ/|K| ~ -0.03	Γ'/|K| ~ -0.08

Recent single crystal data : $Na_2Co_2TeO_6$

Neutron inelastic data

W. Chen et al., Phys. Rev. B 103, L180404 (2021)
W. Yao et al., arXiv 2203.00282v1 (2022)





- Flat band perpendicularly to the planes : quasi 2D-materials (J_{int} very weak)

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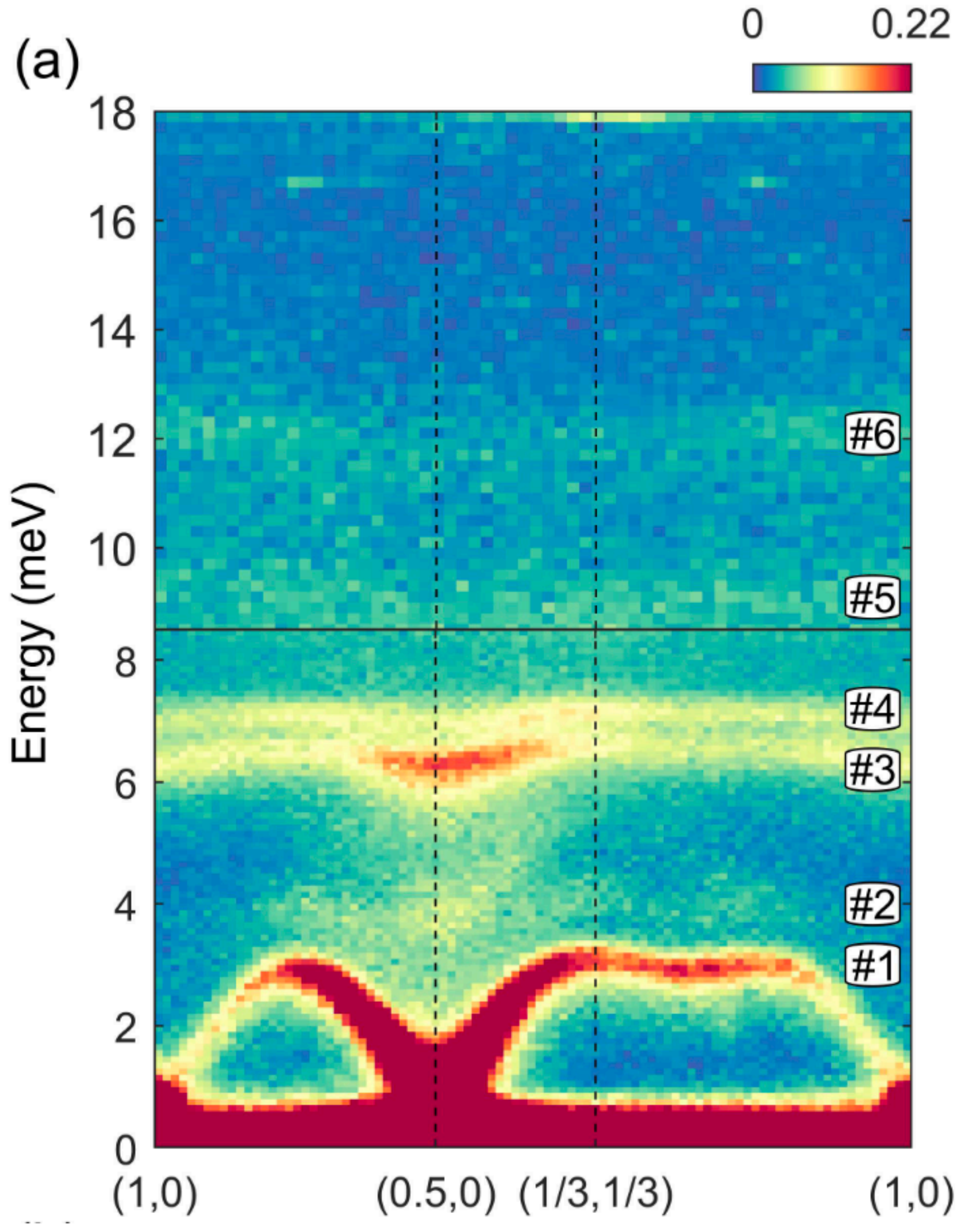
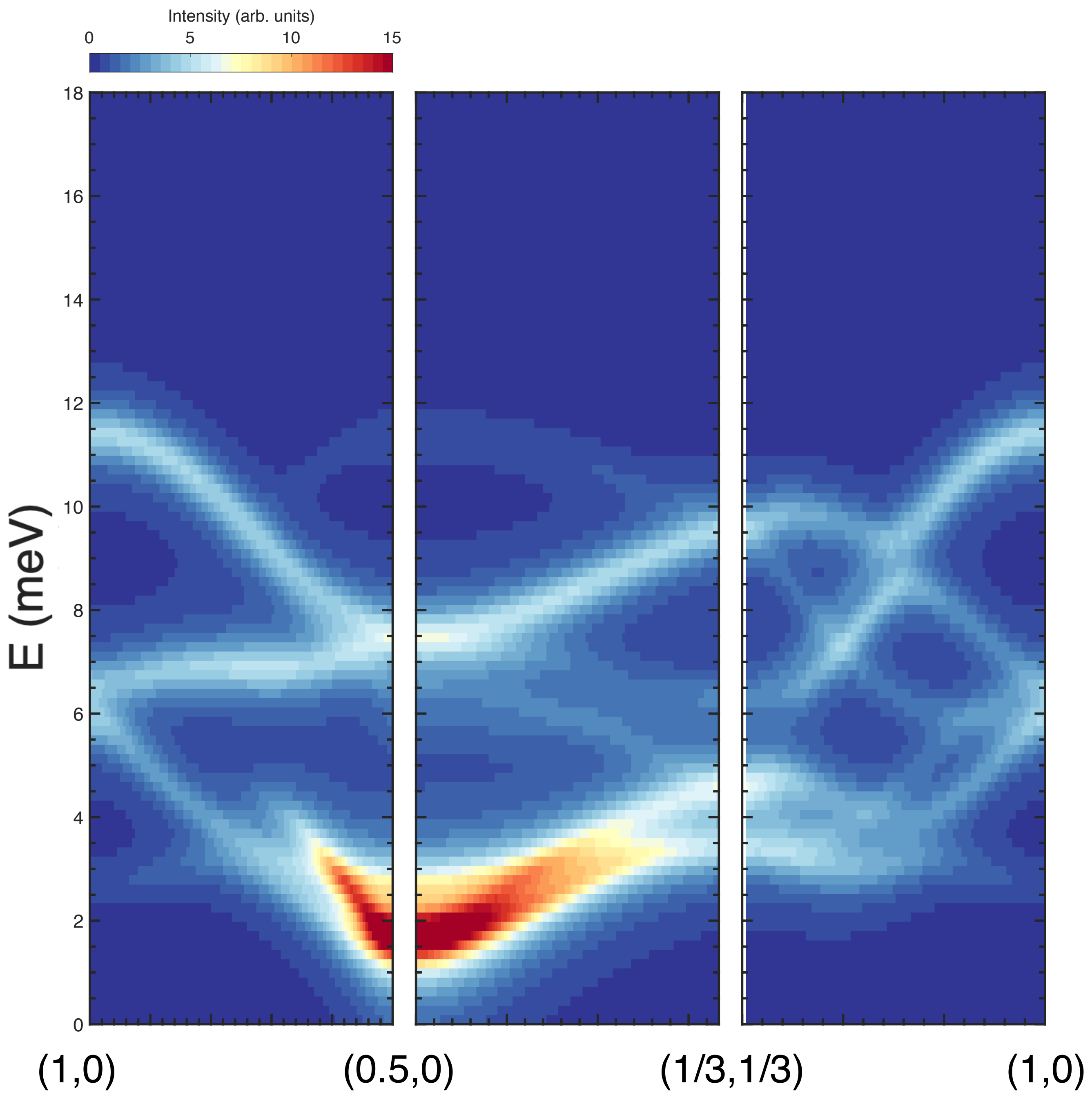


Our model

Phys. Rev. B 102, 224429 (2020)

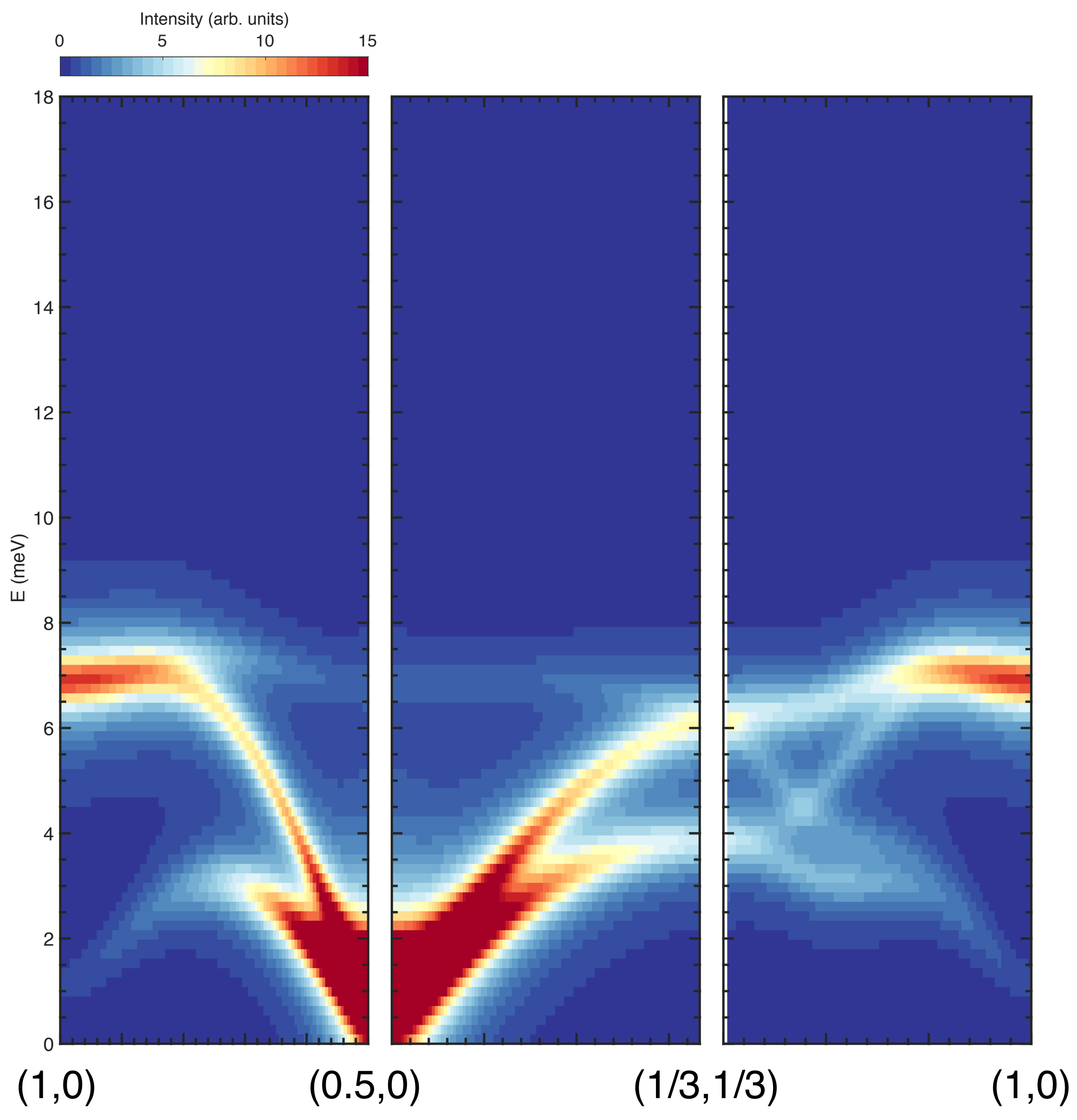
J_1	J_2	J_3	K	Γ	Γ'
-0.1(5)	0.3(3)	0.9(3)	-9.0(5)	1.8(5)	0.3(3)



Lin *et al.*

Nat. Comm. 12, 5559 (2021)

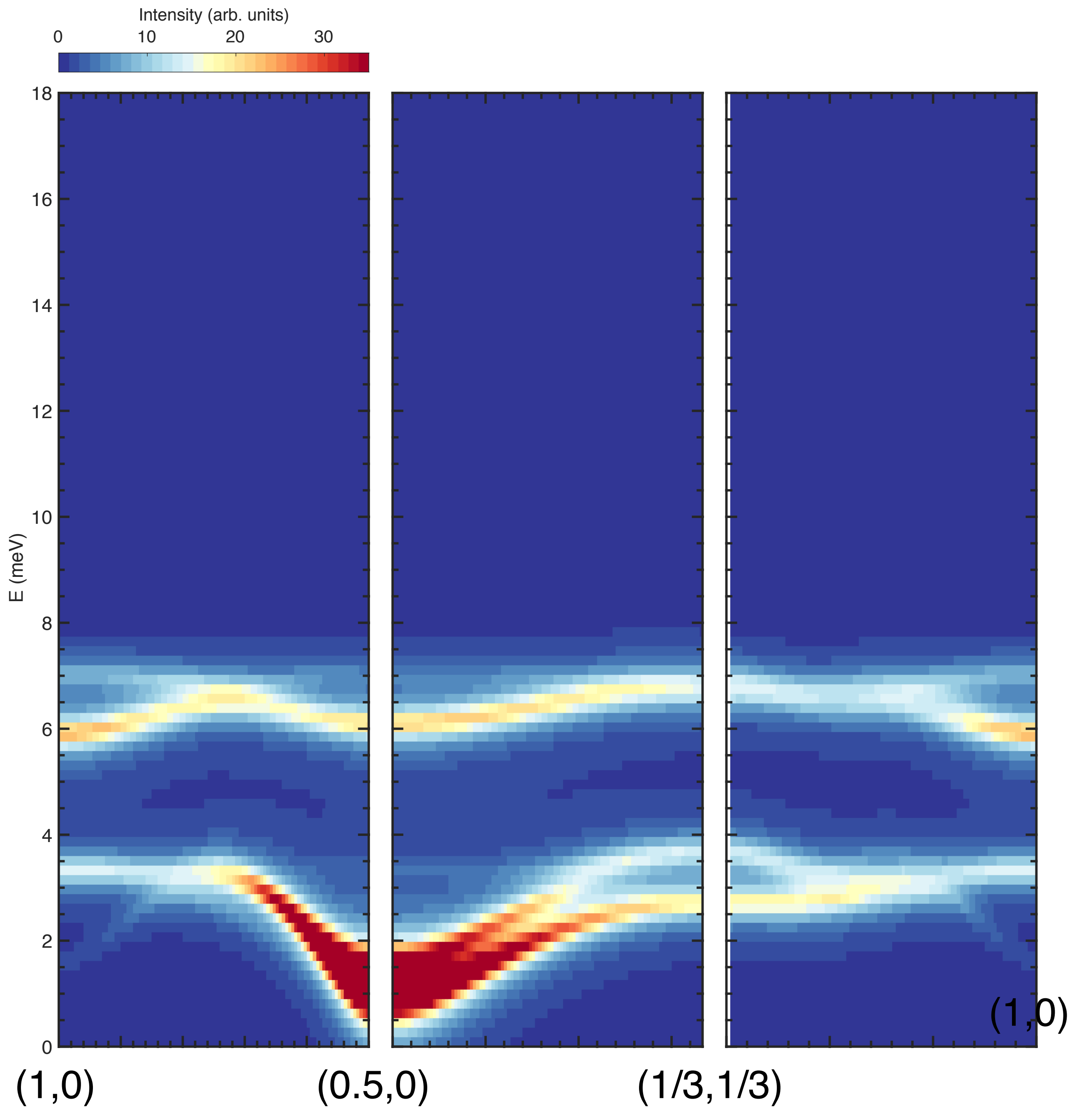
J_1	J_2	J_3	K	Γ	Γ'
2.35	0.0	-2.5	-0.125	-0.125	0.0



Kim *et al.*

J. Phys.: Condens. Matt. 34, 045802 (2021)

J_1	J_2	J_3	K	Γ	Γ'
-1.50(5)	0.0	1.50(2)	3.30(10)	-2.80(5)	2.10(7)



From powder studies

Recent single crystal data : $Na_2Co_2TeO_6$

Neutron inelastic data

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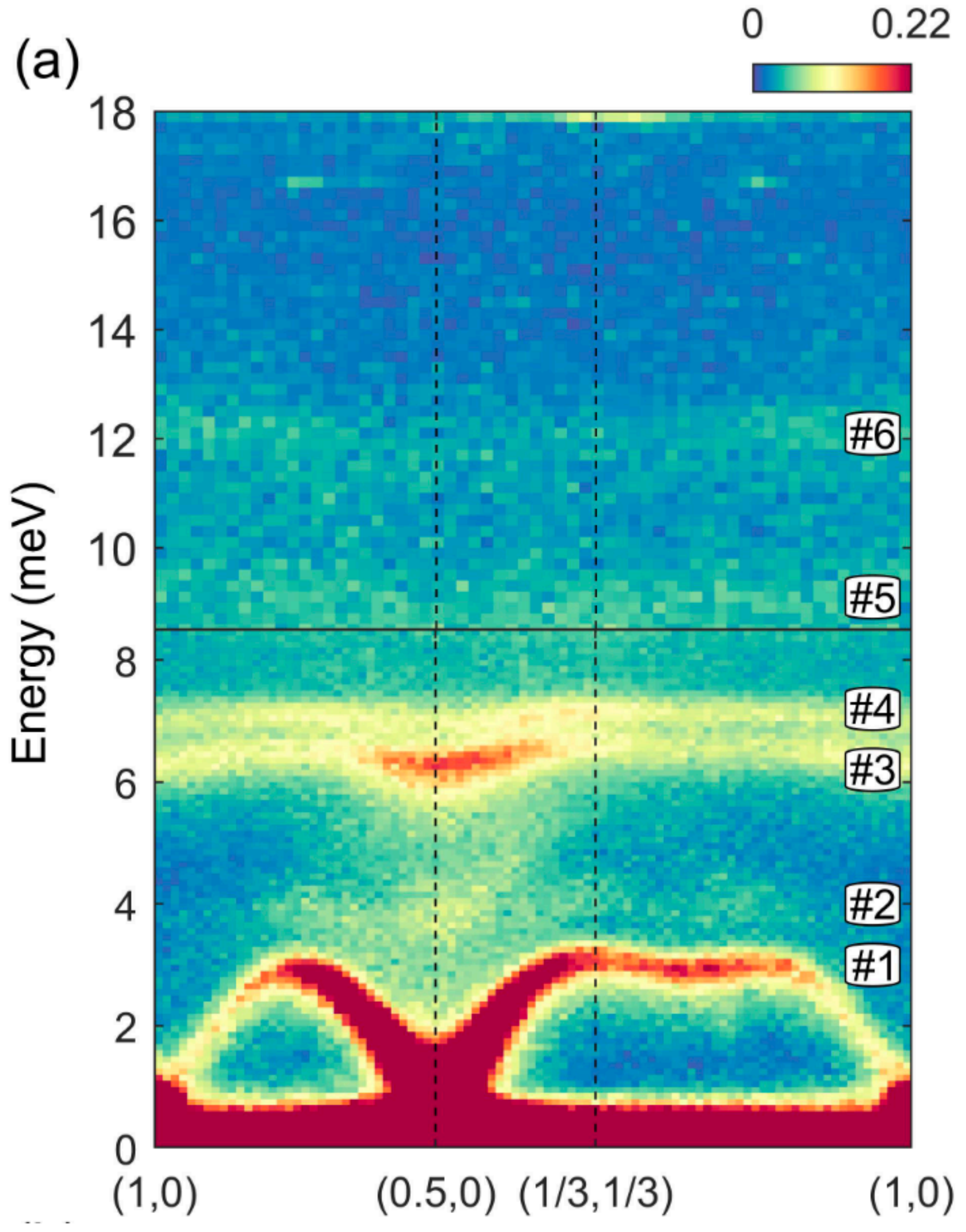

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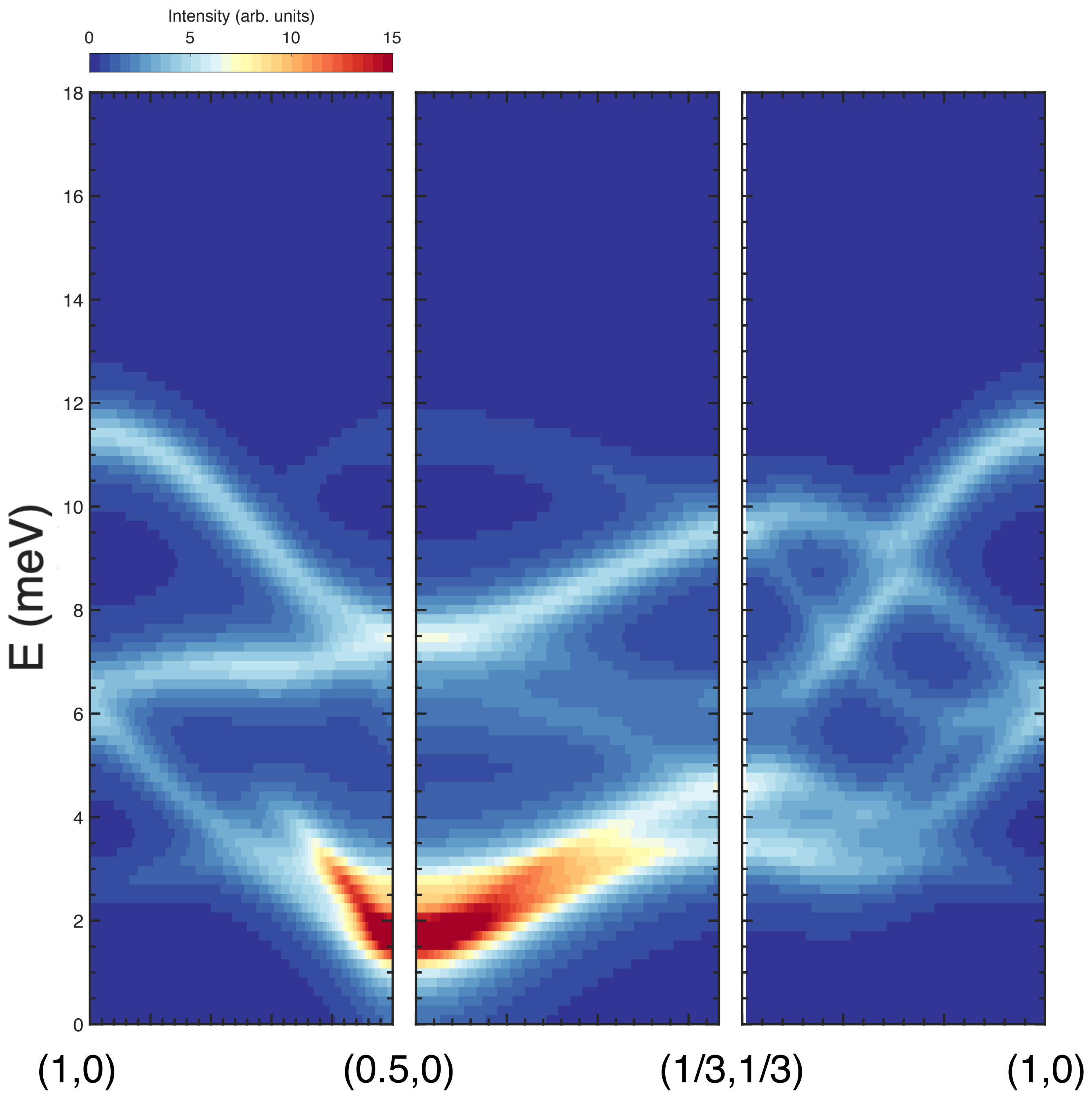
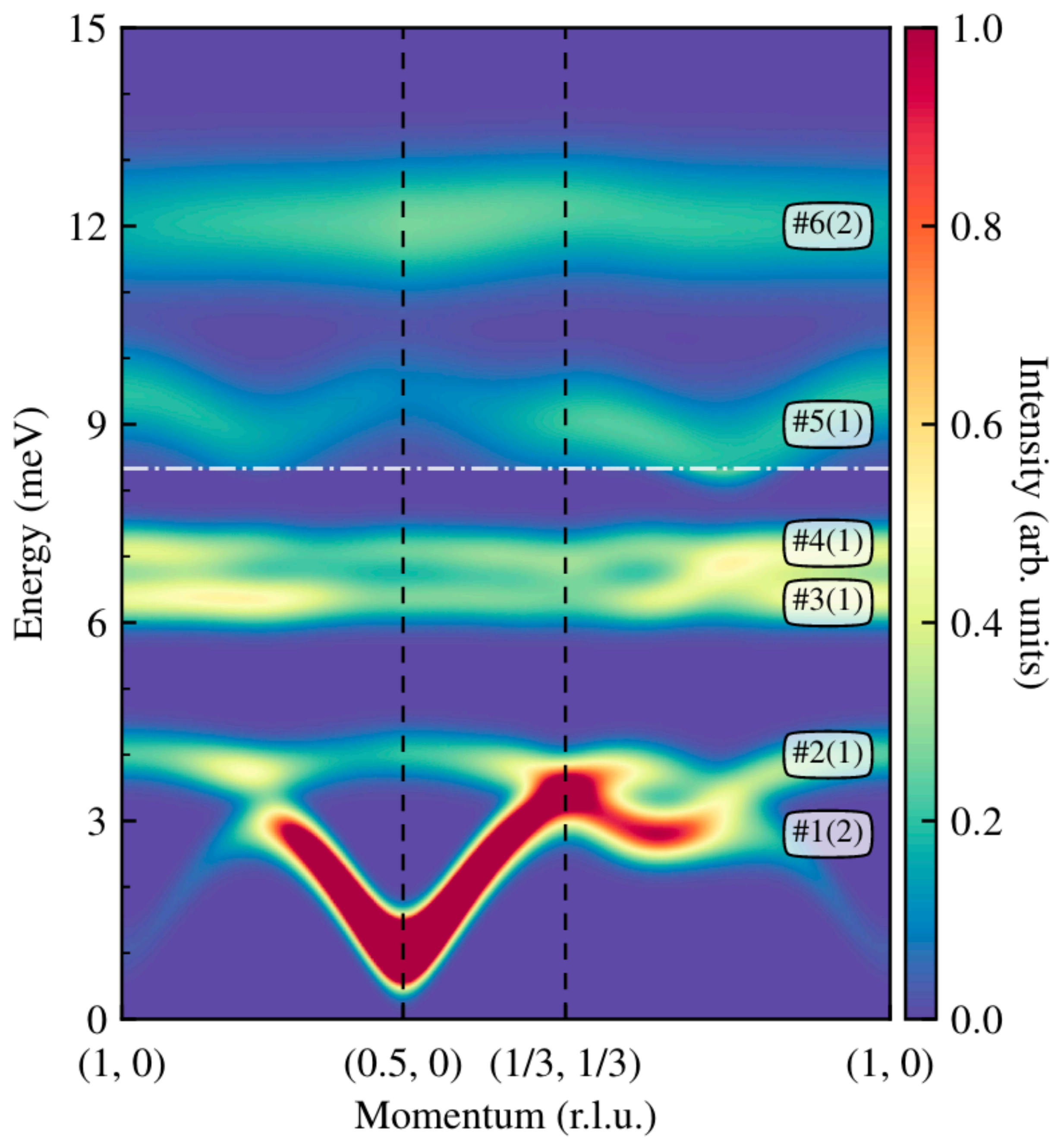


From powder study

Krüger et al.

arXiv 2211.16957v1 (2022)

J_1	J_2^A	J_2^B	J_3	K	Γ	Γ'
1.23	0.32	-0.24	0.47	-8.29	1.86	-2.27



- Proximity to a hidden-SU(2)-symmetric point
- Possible complex triple-Q magnetic ground state

nature COMMUNICATIONS

ARTICLE

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https://doi.org/10.1038/s41467-021-23851-0

OPEN

Order-by-disorder from bond-dependent exchange and intensity signature of nodal quasiparticles in a honeycomb cobaltate

M. Elliot[1], P. A. McClarty[2], D. Prabhakaran[1], R. D. Johnson[3], H. C. Walker[4], P. Manuel[4] & R. Coldea[1✉]

M. Elliot et al., Nature Comm. 12, 3936 (2021)
B. Yuan et al., PRX 10, 011062 (2020), PRB 102, 134404 (2020)

Conclusion

- Rich playground for material research/design: interplay between spin-orbit coupling, crystal field, Hund's coupling and lattice (3d/4d/5d transition metals, rare-earth, other geometries)
- Recent motivation re-visit Co materials: versatile materials with rich and complex magnetic states, beyond Kitaev physics (complex magnetic order, multi-q state, hidden order, topological magnons…)

$BaCo_2(AsO_4)_2$, $BaCo_2(PO_4)_2$, $CoPS_3$, $CoTiO_3$…

L. -P. Regnault et al., Heliyon 4 (2018)
H. Nair et al., PRB 97, 134409 (2018)
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A.R. Wildes et al., J. Phys.: Condens. Matter 29, 455801 (2017)
B.C. Kim et al., PRB 102, 184426 (2020)
and many others…

Other geometries (triangular, 1D chains, hyper-honeycomb…)

$Ba_3CoSb_2O_9$: PRB 102, 064421 (2020), PRL 108, 057205 (2012)
$Na_2BaCo(PO_4)_2$, $Na_2SrCo(PO_4)_2$ PNAS 116, 14505 (2019)

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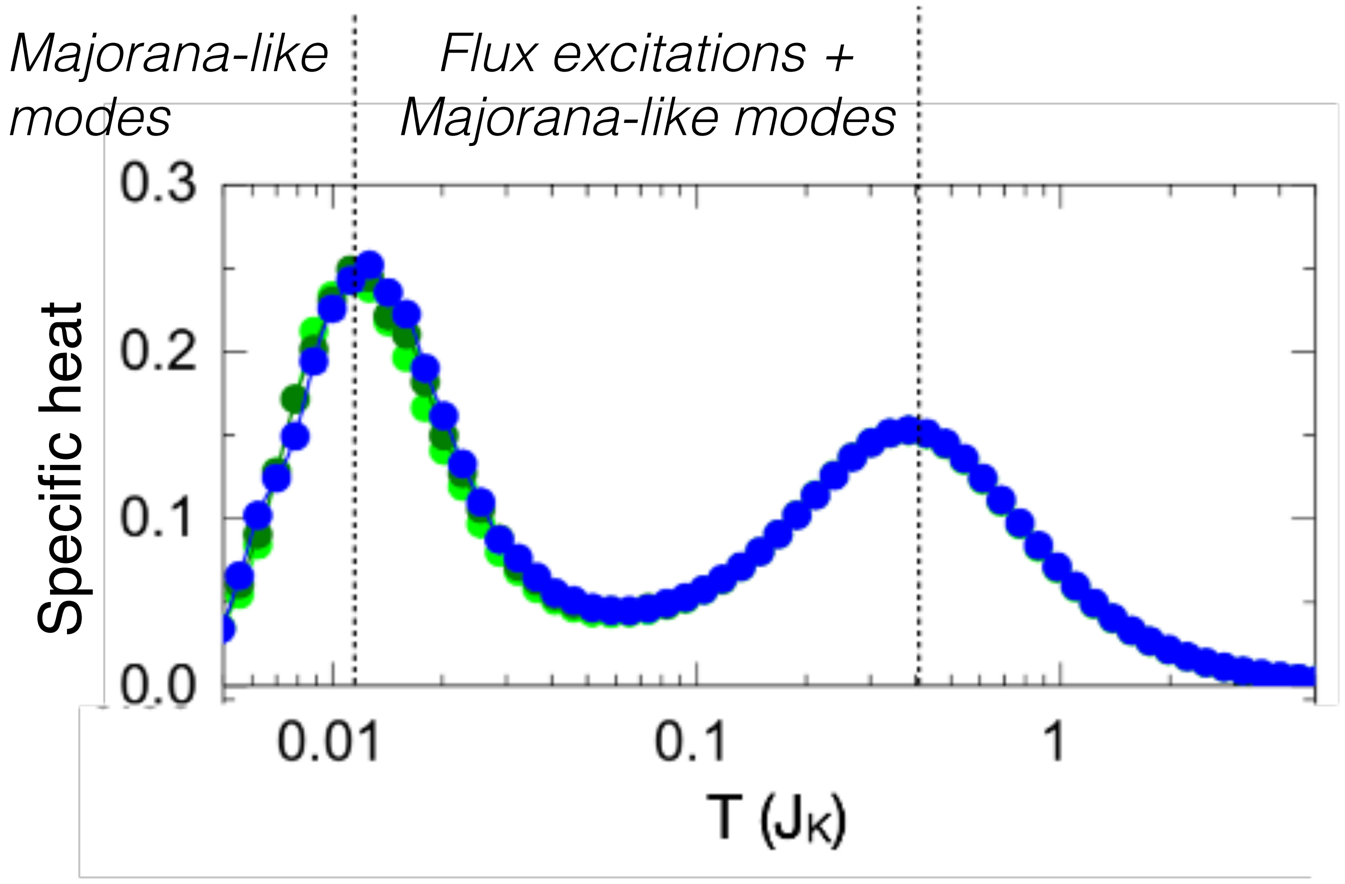
Thank you !

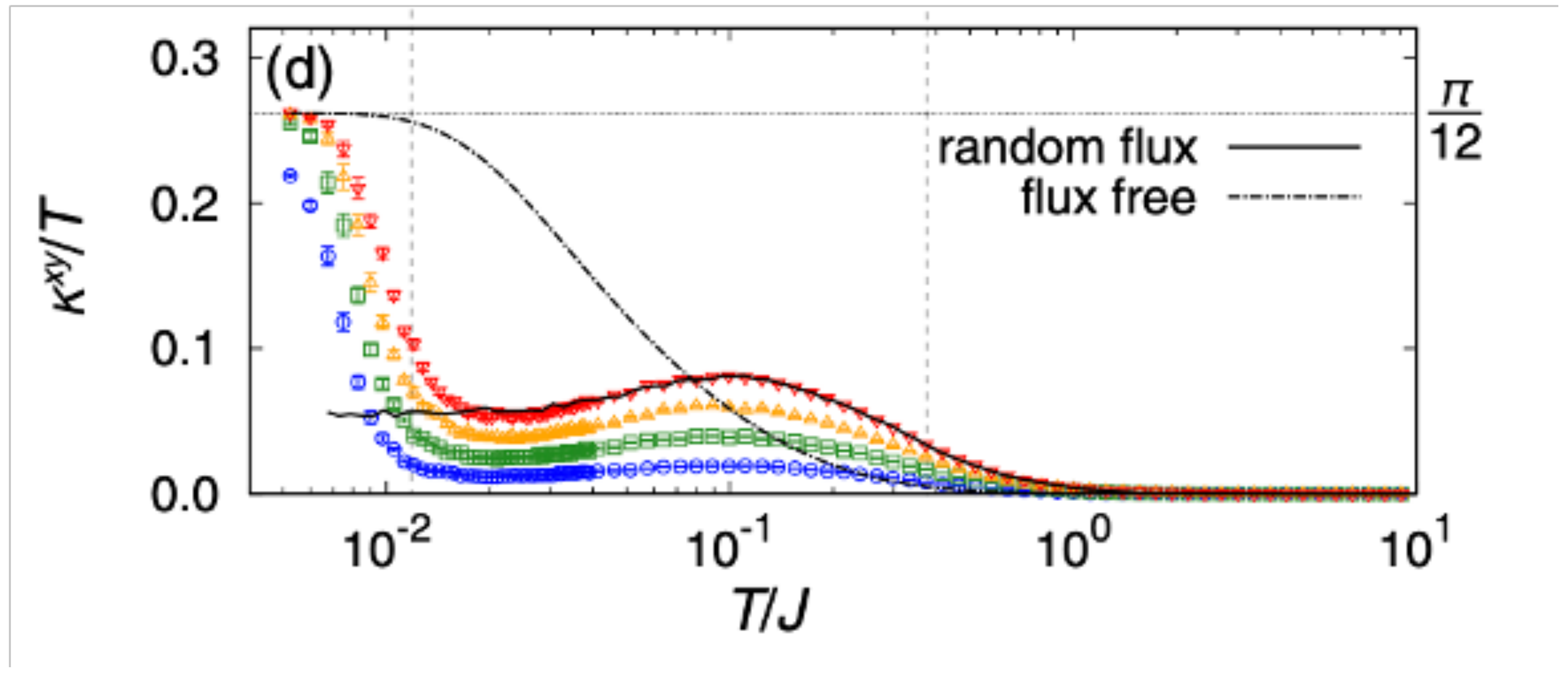
What experimental signatures?

An exact model: predictions for experimental signatures

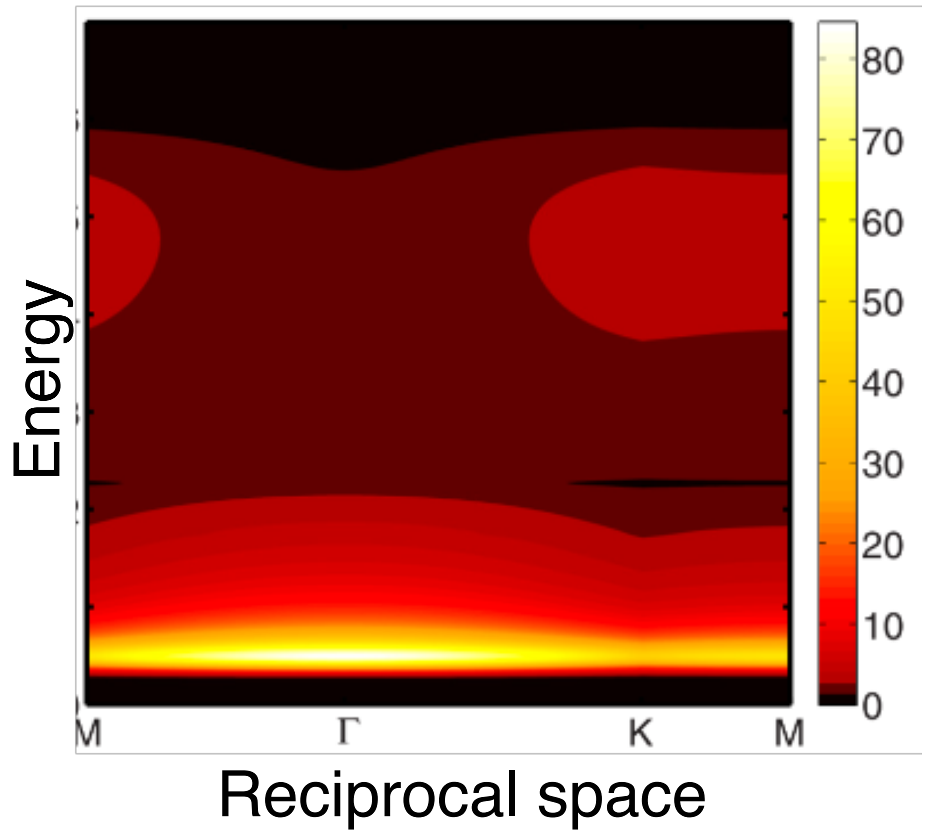
Magnetization, specific heat
Thermodynamic signatures of fractional excitations



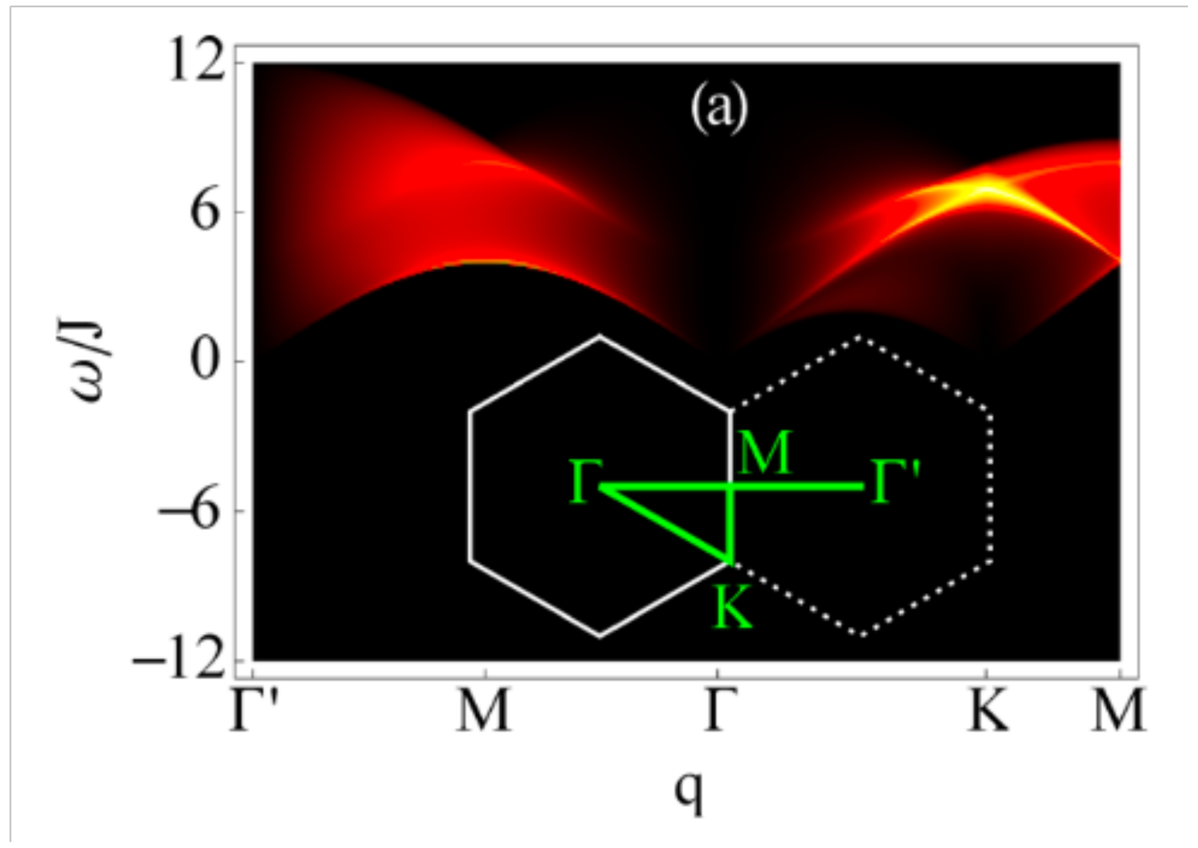
Thermal Hall effect
Signature for topological phase: chiral edge modes



Neutron scattering
Continuum of fractional excitations



Resonant X-ray scattering
Continuum of fractional excitations



J. Yoshitake et al., Phys. Rev. Letters (2016)
G. Halasz et al., Phys. Rev. B (2019)
J. Knolle et al., Phys. Rev. B (2015)

Neutron inelastic study (powders)

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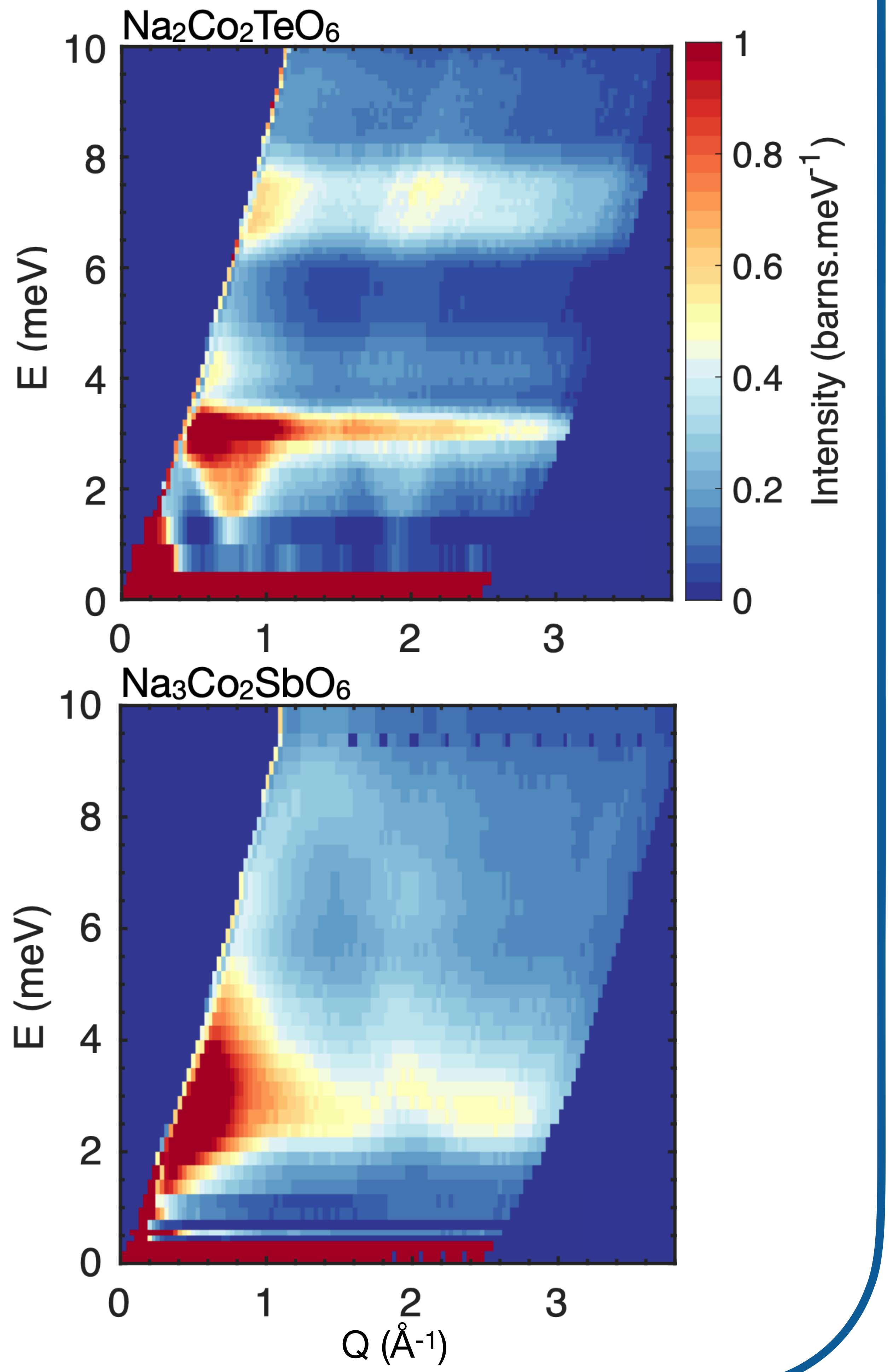
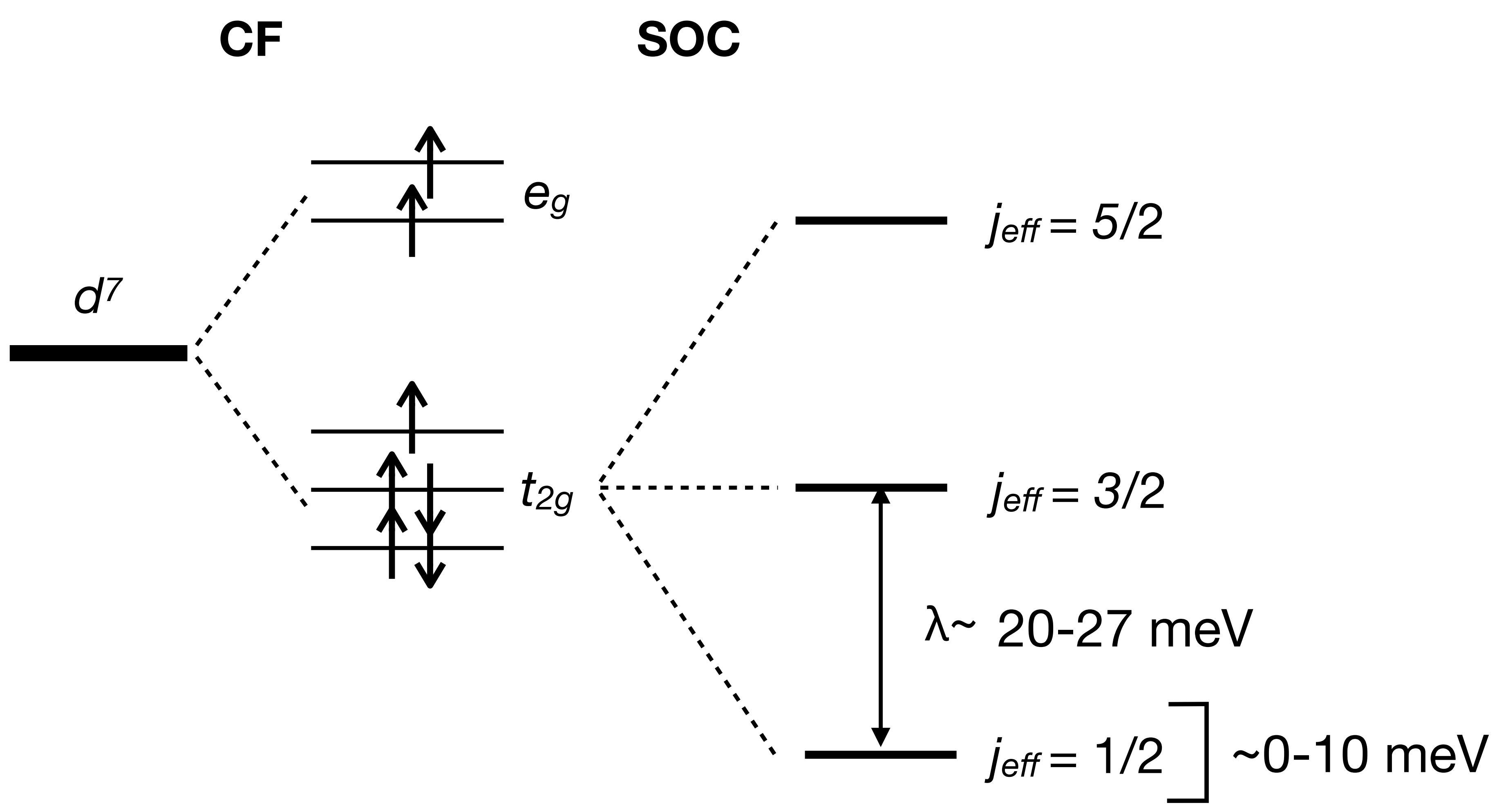
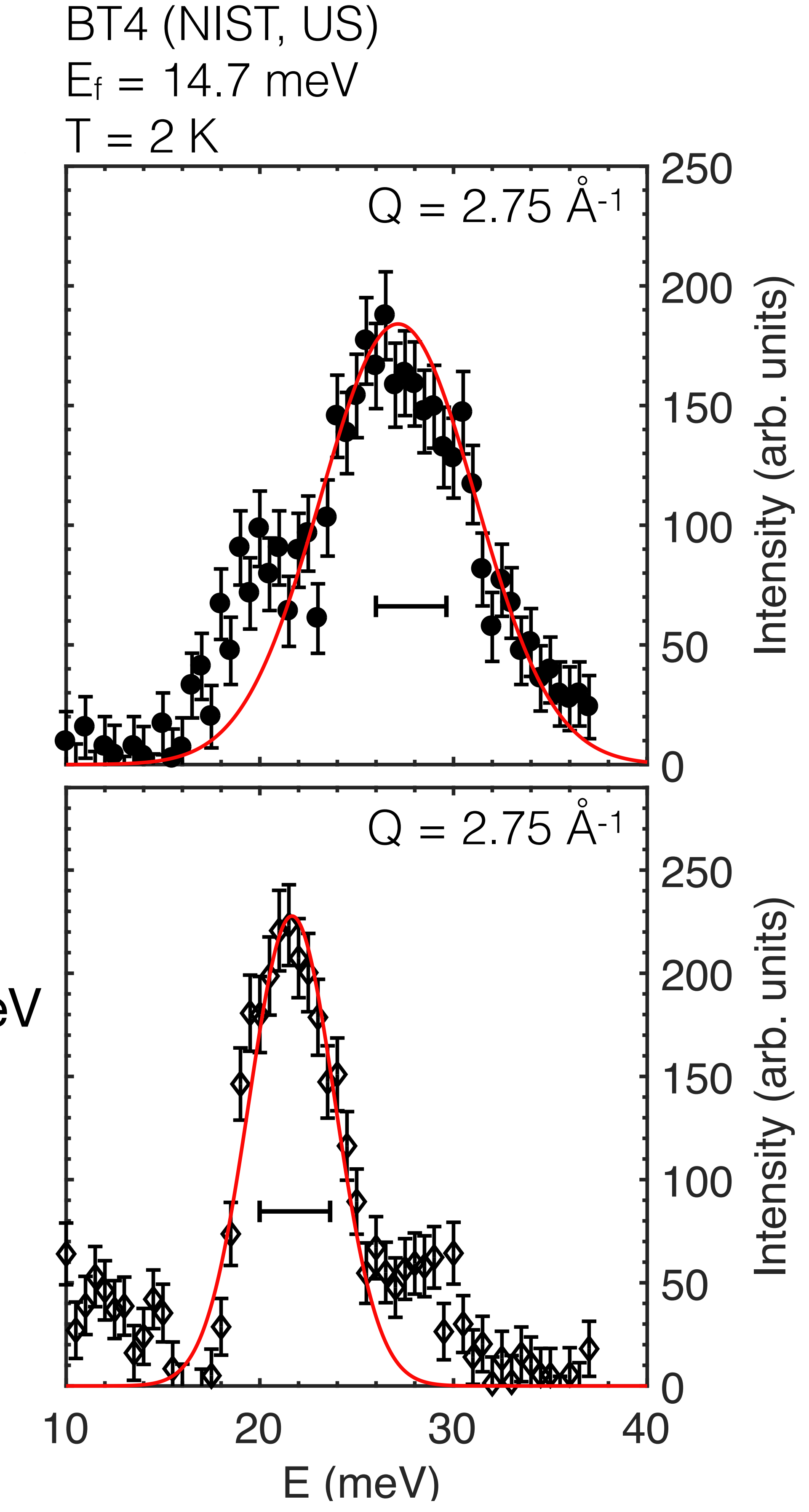


Higher Energy:

- Excitations between the j_{eff} = 1/2 and j_{eff} = 3/2 manifolds : spin-orbit coupling





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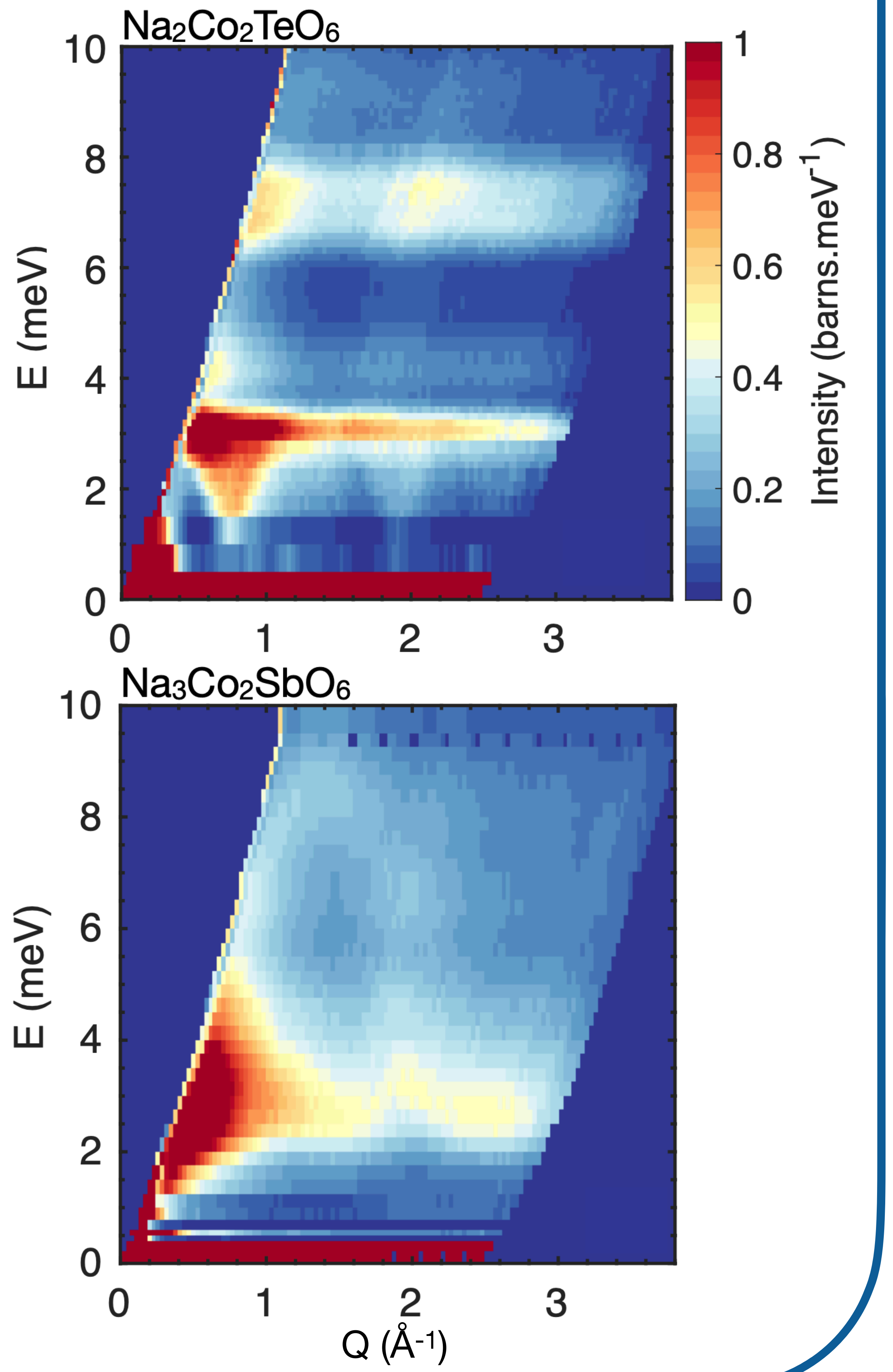
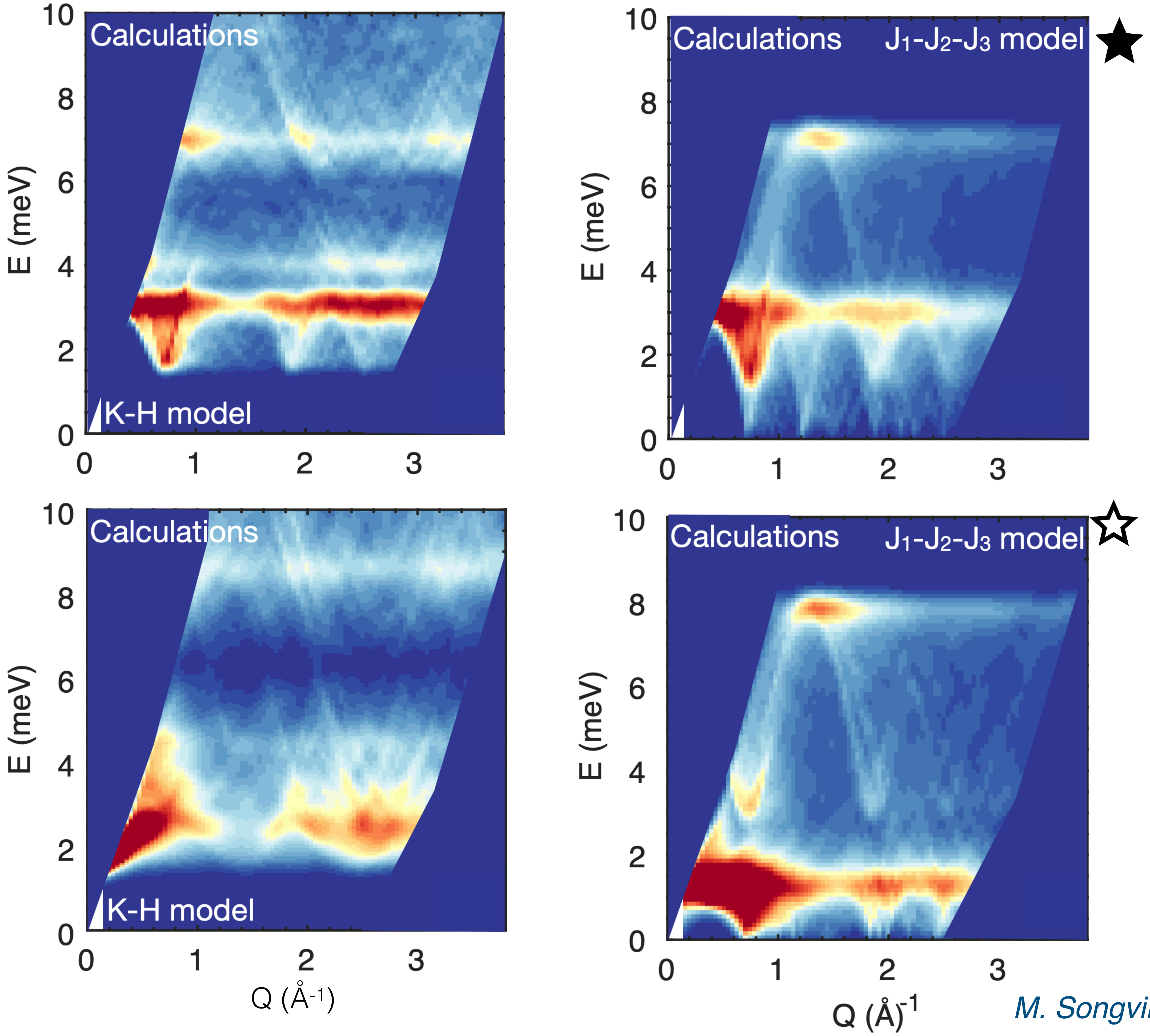
NIST
National Institute of Standards and Technology

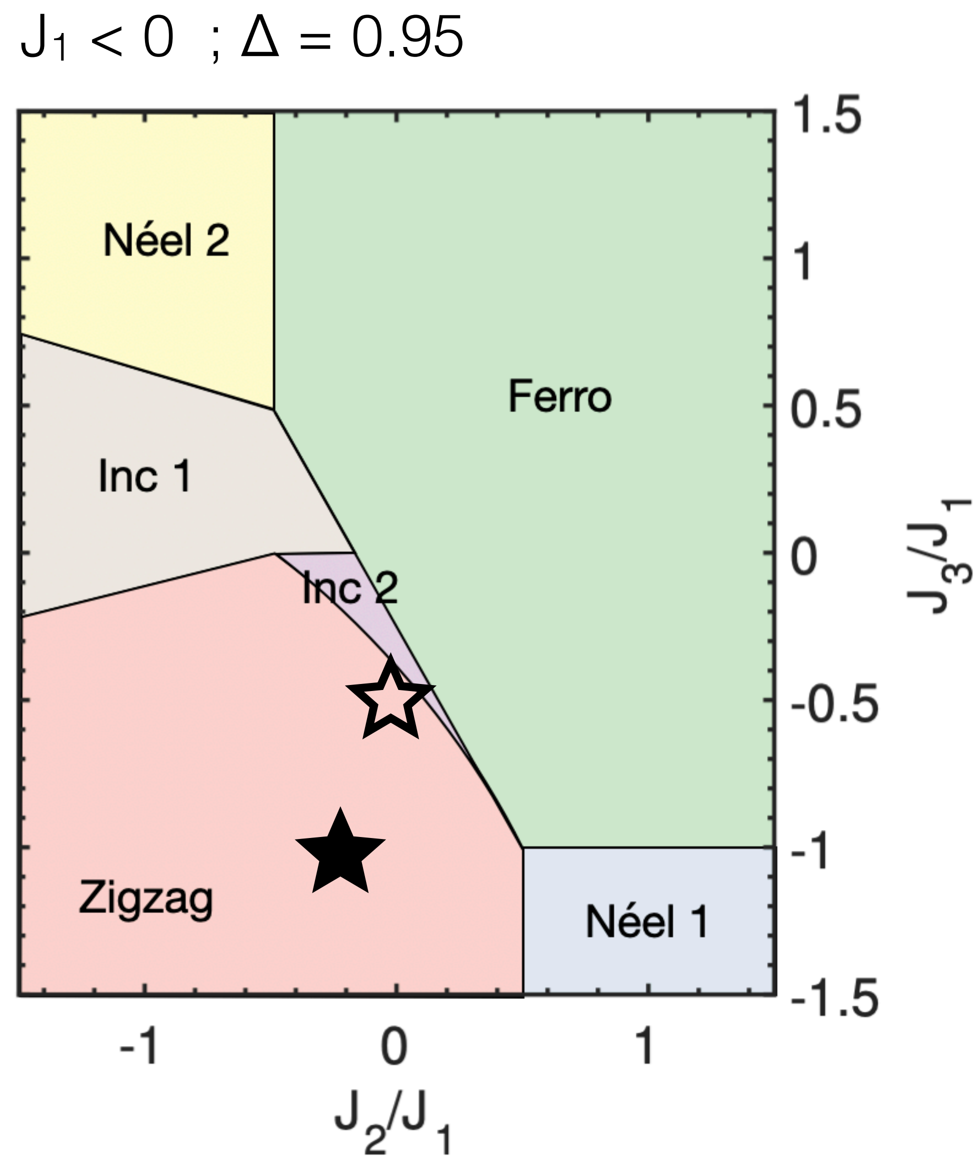


Spin wave calculations : Kitaev-Heisenger vs. Heisenberg XXZ models

Spinwave software
S. Petit, Collection SFN 12, 105 (2011)

$$H_{XXZ} = \sum_{n=1}^{3} J_n \sum_{i,j} \left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z \right)$$

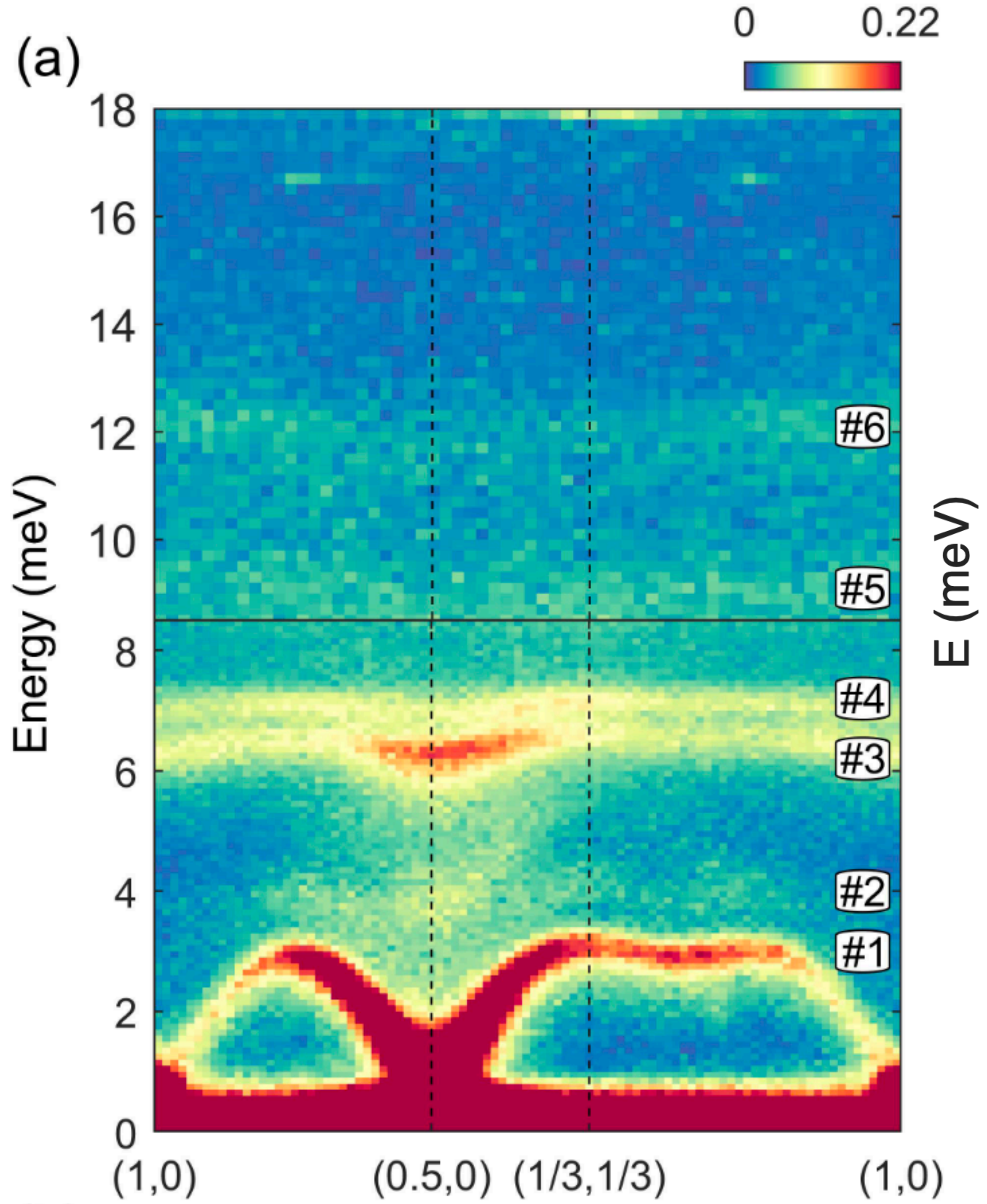


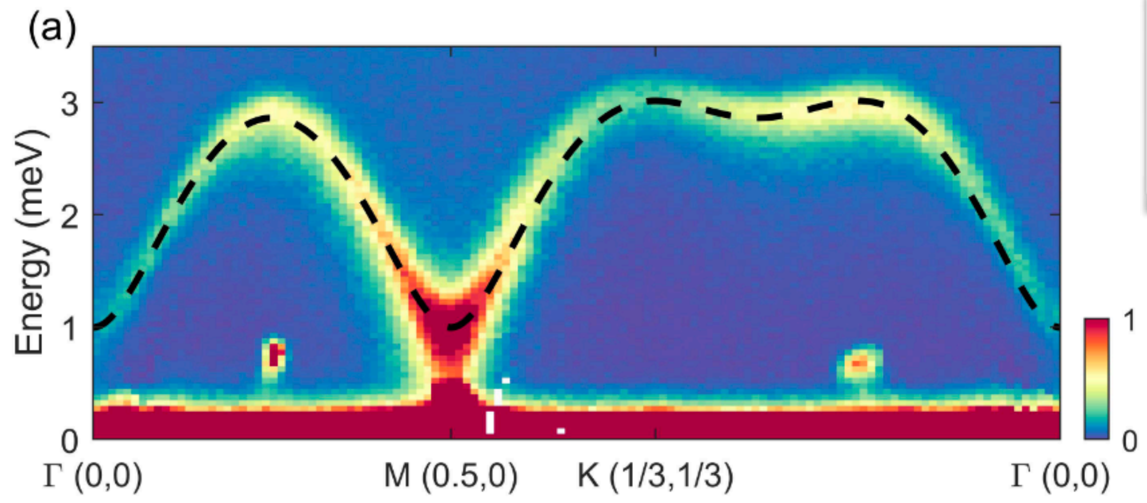


M. Songvilay et al., Phys. Rev. B 102, 224429 (2020)

$Na_2Co_2TeO_6$: relevance of Kitaev interactions?

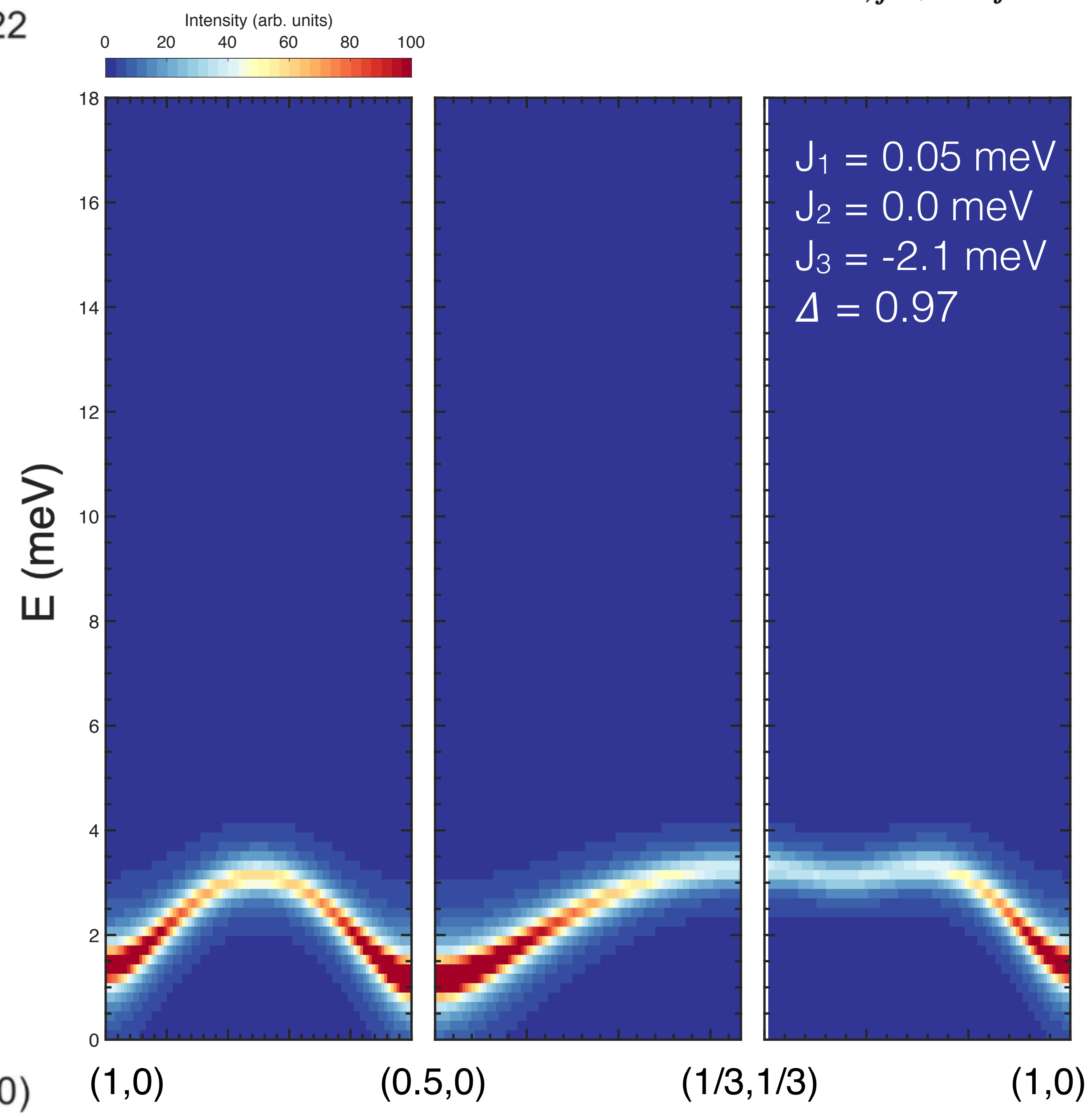
W. Chen et al., Phys. Rev. B 103, L180404 (2021)
W. Yao et al., arXiv 2203.00282v1 (2022)



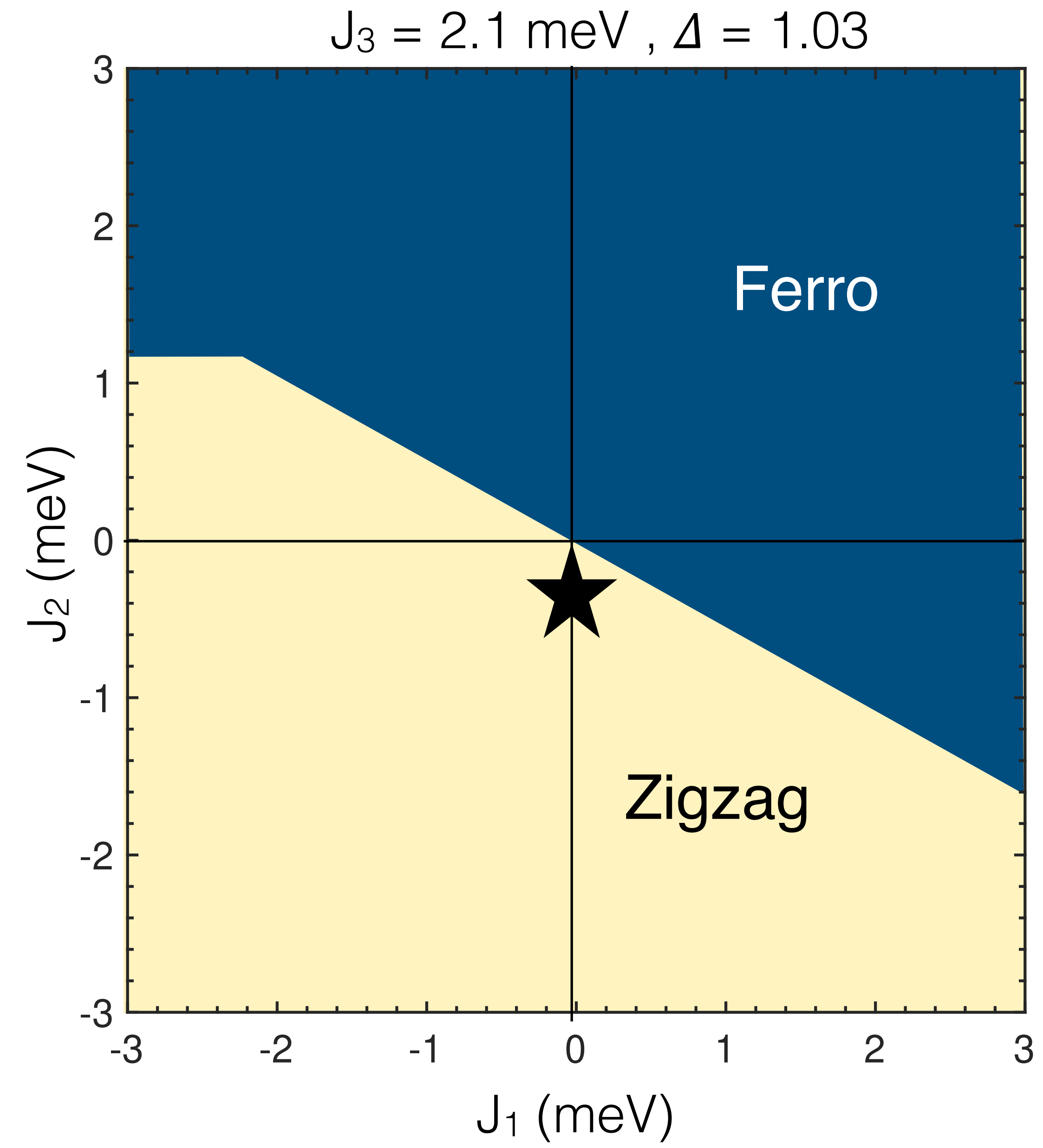


J_1-J_2-J_3 model with weak Ising anisotropy

$$H = \sum_{n=1}^{3} J_n \sum_{i,j} \left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z\right)$$



- A weak J_1 interaction necessary to stabilise the **k** = (1/2 0 0) phase



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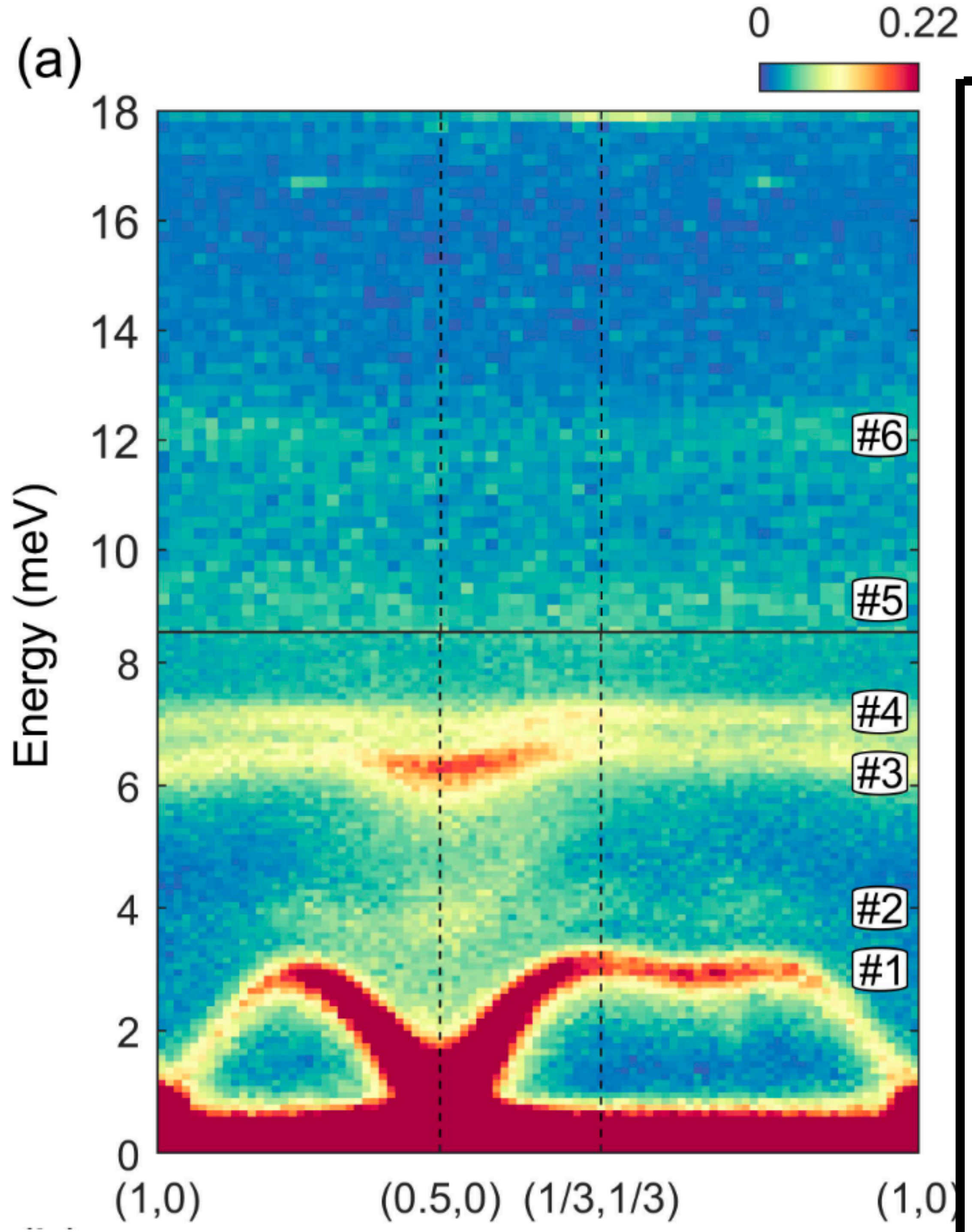
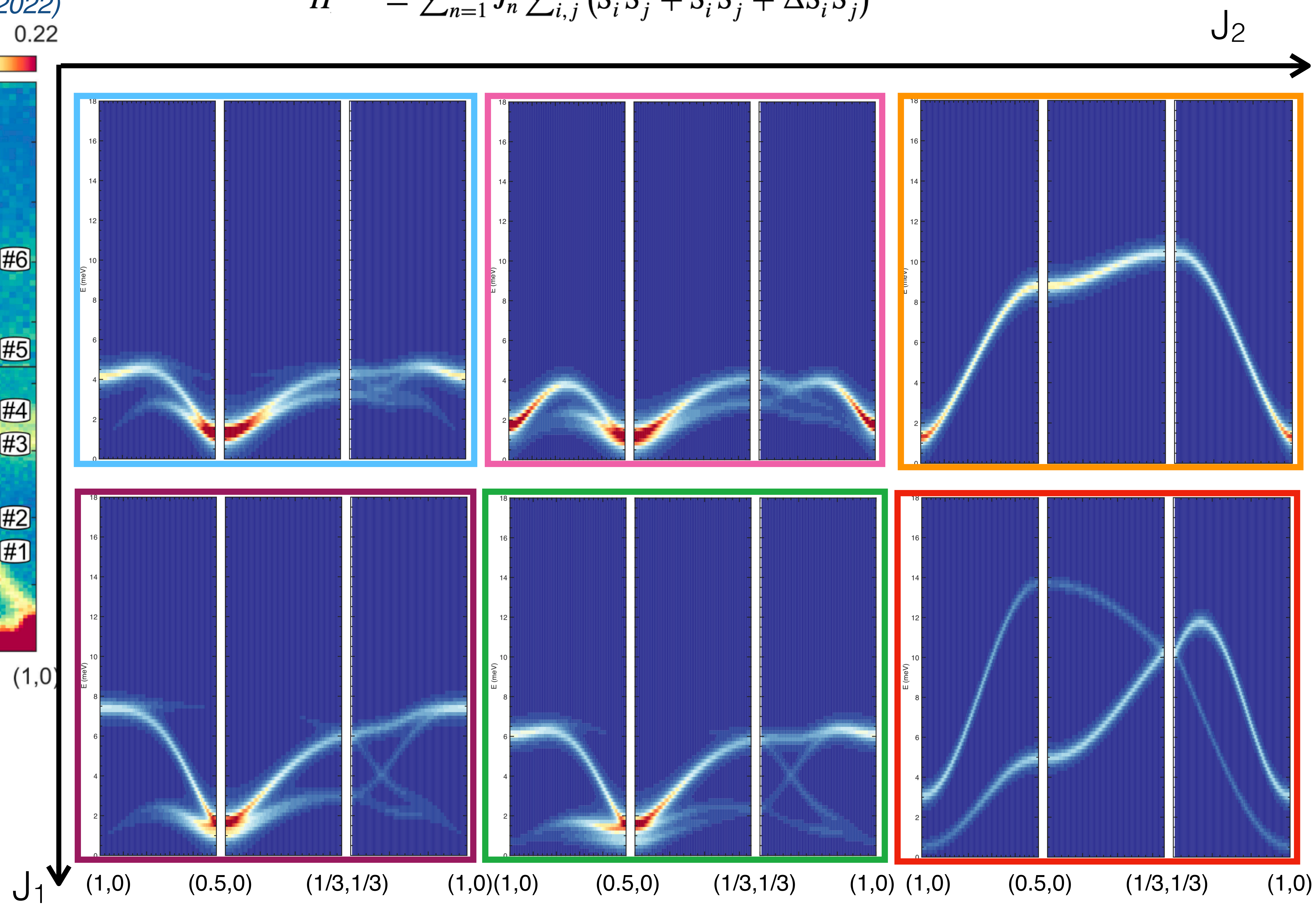
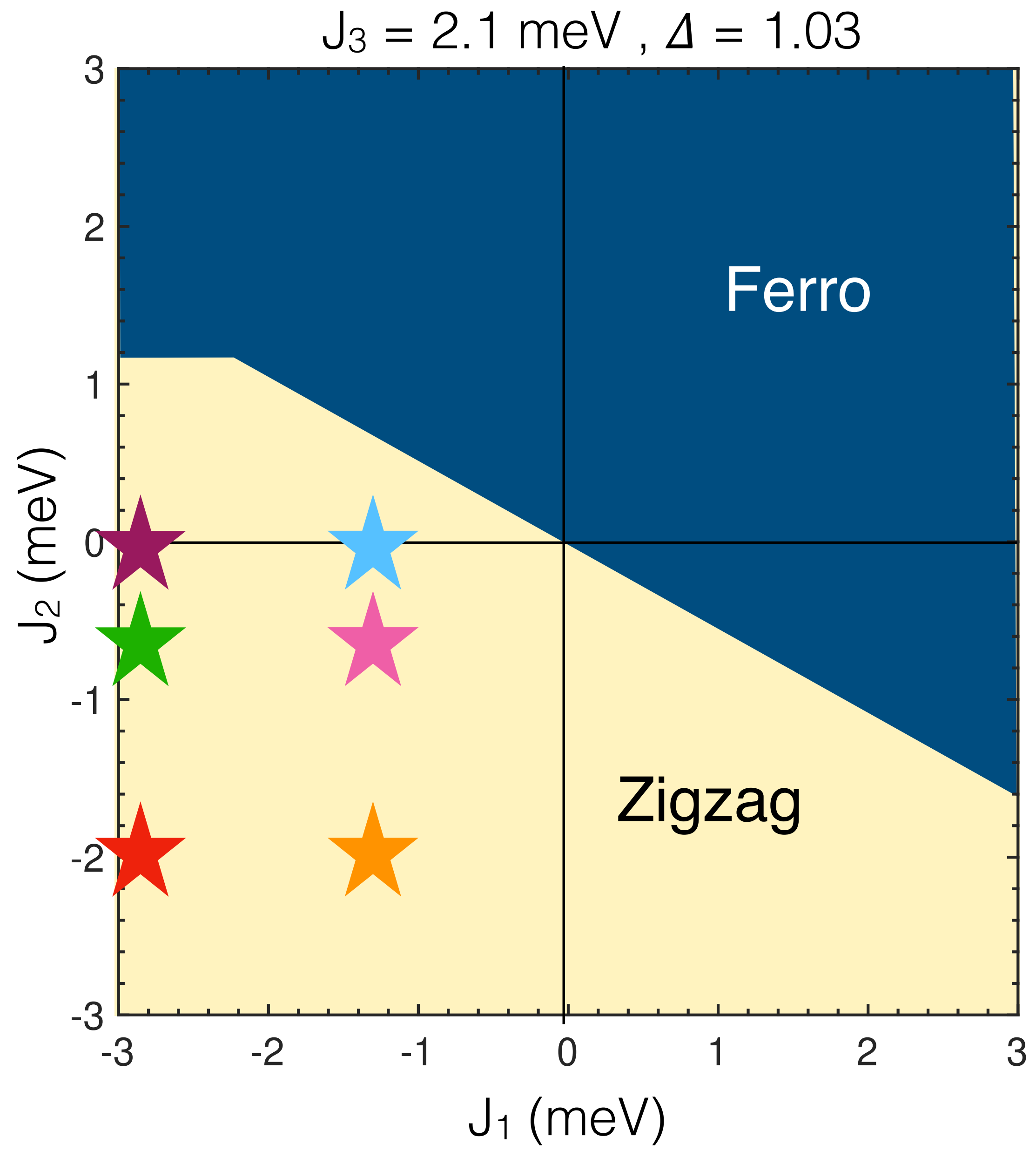


J_1-J_2-J_3 model with weak Ising anisotropy

$$H = \sum_{n=1}^{3} J_n \sum_{i,j} \left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z\right)$$





▷ Another ingredient is necessary: bond dependent interactions? … to be continued