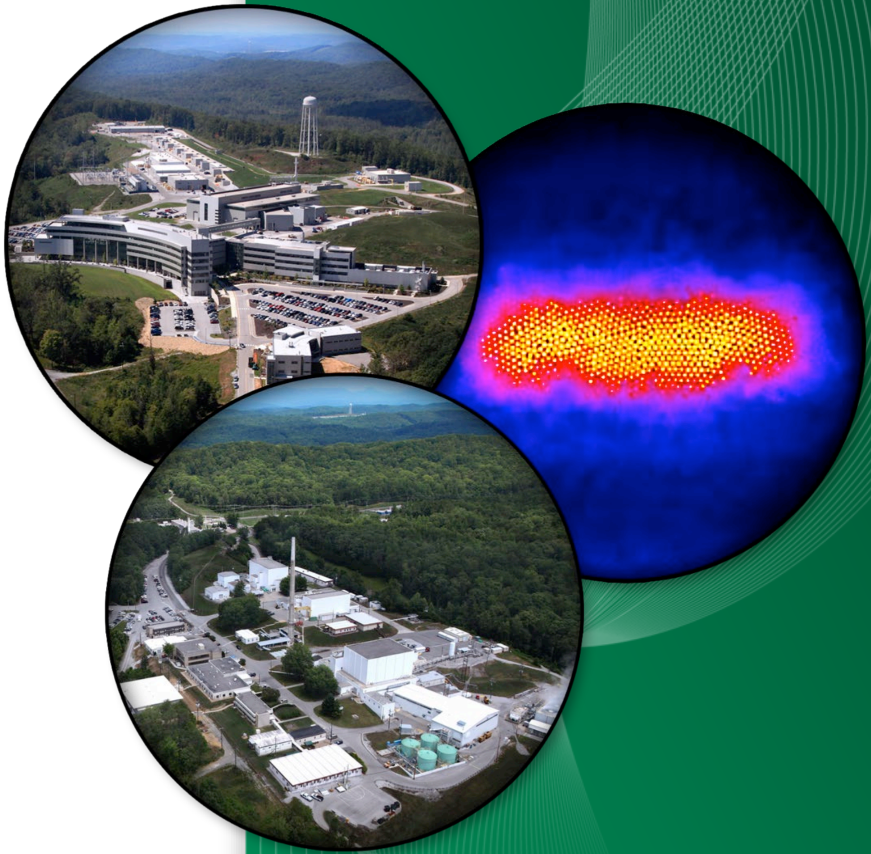
Magnetic excitations and possible spin liquid physics in α-$RuCl_3$



Stephen Nagler
Oak Ridge National Laboratory

Outline

I. Kitaev's model & materials

II. Ordering in α-RuCl$_3$

III. Inelastic neutron scattering in α-RuCl$_3$

IV. Higher fields, magnetocaloric effect, T-B phase diagram

Neutron Scattering Collaborators:

A. Banerjee, A. Aczel, C. Balz, C. Batista, S. Bhattacharjee, C. Bridges, H. Cao, B. Chakoumakos, G. Ehlers, O. Garlea, G. Granroth, Y. Kamiya, J. Knolle, D. Kovrizhin, P. Lampen-Kelley, L. Li, Y. Liu, Z. Lu, M. Lumsden, D. Mandrus, R. Moessner, M. Stone, D, Pajerowski, A. Samarakoon, D. A. Tennant, B. Winn, J.-Q. Yan, Y. Yiu, S. Zhang.

Most recent: Christian Balz et al.,

PRB 100 060405(R), 2019

MCE collaborators:
X. Hu, S. M. Yadav, Y. Takano

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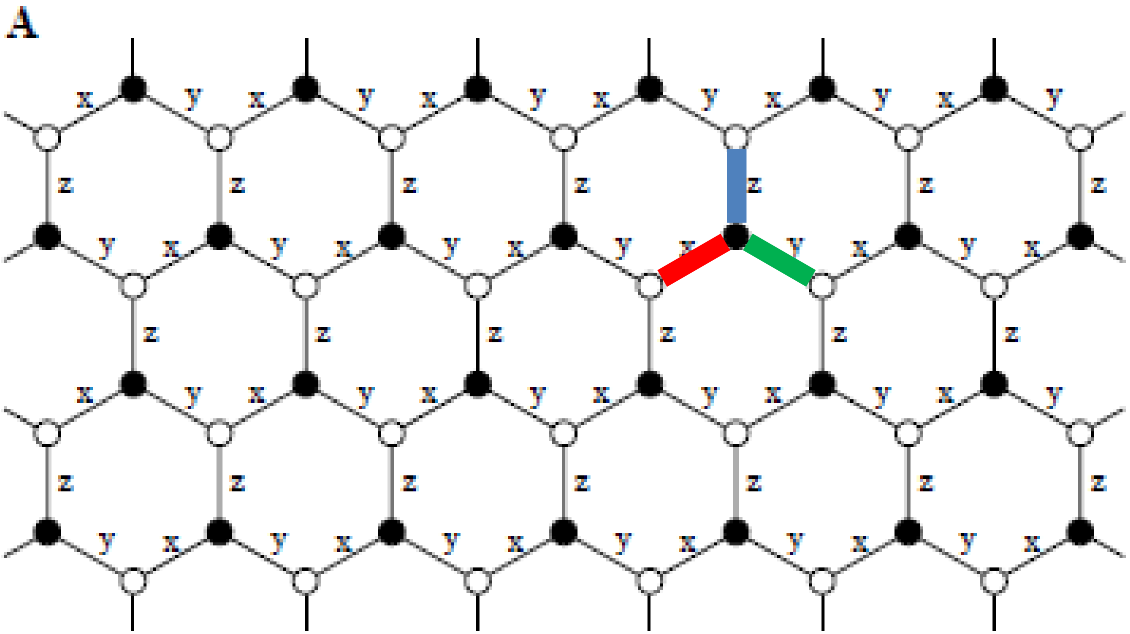
Kitaev's model on honeycomb lattice – a special QSL

$$H_{\text{Kitaev}} = -\sum_{\gamma-\text{bonds}} K_\gamma \, S_i^\gamma S_j^\gamma$$

- Kitaev interaction: Bond-directional dependent Ising coupling
- Exactly solvable Hamiltonian

→ ***quantum spin liquid*** ground state



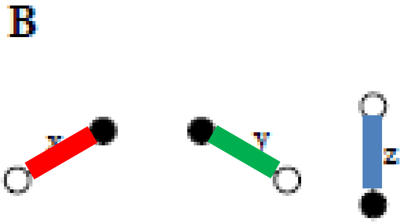
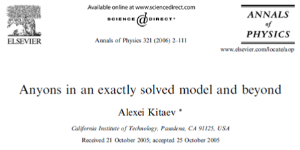


Fig. 3. Three types of links in the honeycomb lattice.



Anyons in an exactly solved model and beyond

Alexei Kitaev *

California Institute of Technology, Pasadena, CA 91125, USA

Received 21 October 2005; accepted 25 October 2005

Kitaev interactions in materials

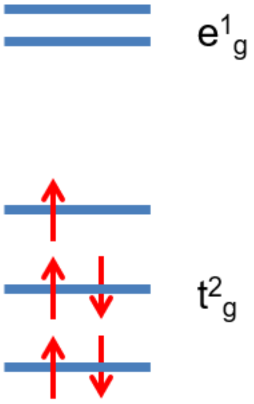
PRL **102,** 017205 (2009) PHYSICAL REVIEW LETTERS week ending 9 JANUARY 2009

Mott Insulators in the Strong Spin-Orbit Coupling Limit: From Heisenberg to a Quantum Compass and Kitaev Models

G. Jackeli[1,*] and G. Khaliullin[1]

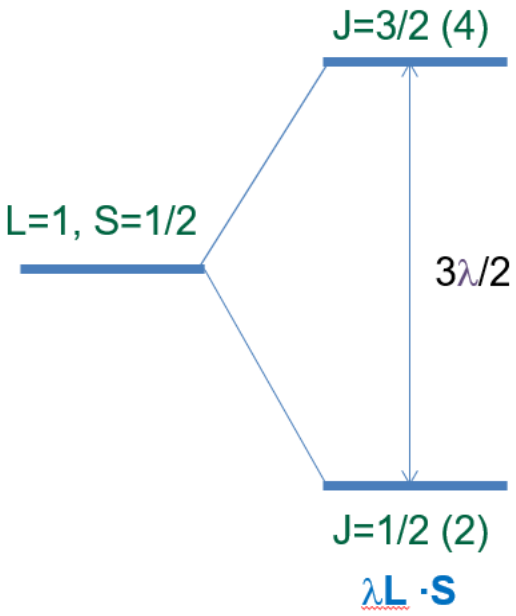
See also:
H. Takagi *et al.*,
Nature Reviews Physics 1, (2019)

d^5 in low spin octahedral configuration

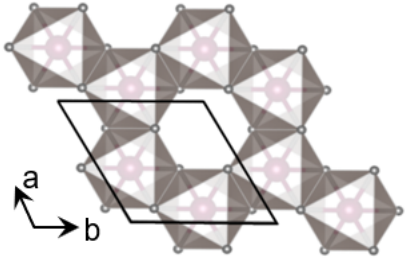


strong field limit
S=1/2, L_{eff} =1
e.g. ($5d^5$) Ir^{4+}
($4d^5$) Ru^{3+}

strong spin-orbit coupling



edge-sharing octahedra



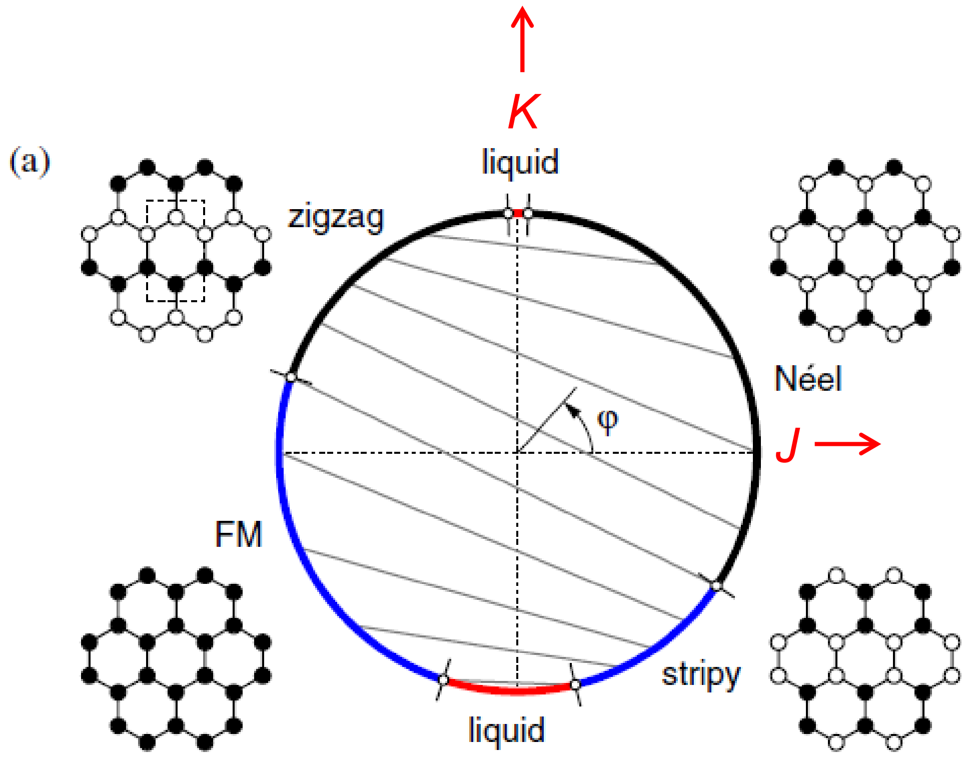
Heisenberg – Kitaev Phase Diagram

PRL **110,** 097204 (2013) PHYSICAL REVIEW LETTERS week ending 1 MARCH 2013

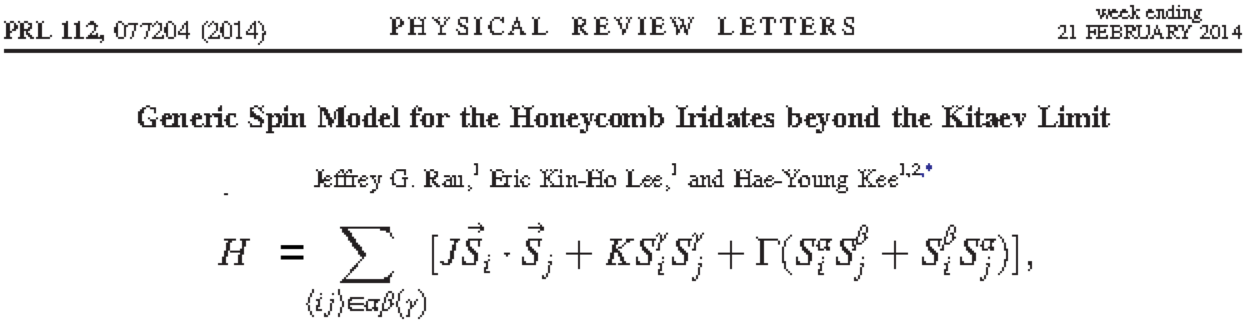
Zigzag Magnetic Order in the Iridium Oxide Na_2IrO_3

Jiří Chaloupka,[1,2] George Jackeli,[1,*] and Giniyat Khaliullin[1]

$$\mathcal{H}_{ij}^{(\gamma)} = 2KS_i^\gamma S_j^\gamma + J\mathbf{S}_i \cdot \mathbf{S}_j.$$



Effect of additional interactions

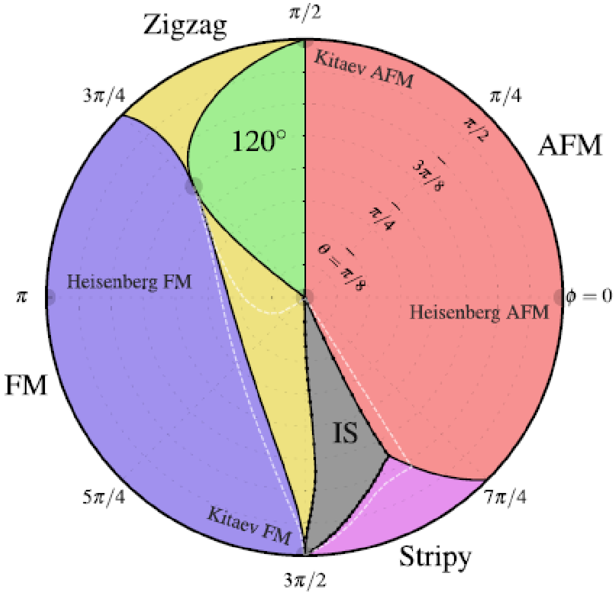


PRL 112, 077204 (2014) PHYSICAL REVIEW LETTERS week ending 21 FEBRUARY 2014

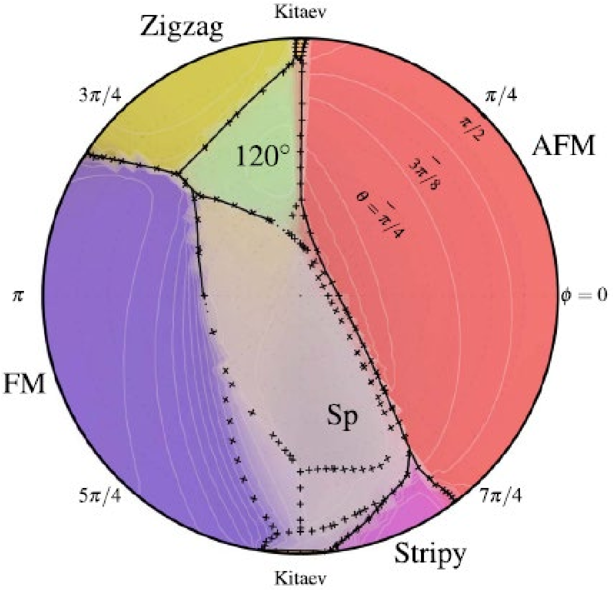
Generic Spin Model for the Honeycomb Iridates beyond the Kitaev Limit

Jeffrey G. Rau,[1] Eric Kin-Ho Lee,[1] and Hae-Young Kee[1,2,*]

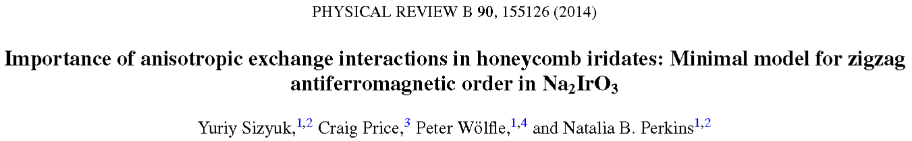
$$H = \sum_{\langle ij\rangle\in\alpha\beta(\gamma)} [J\vec{S}_i\cdot\vec{S}_j + KS_i^\gamma S_j^\gamma + \Gamma(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha)],$$



(a) Classical phase diagram with $\Gamma > 0$



(a) Phase diagram for $\Gamma > 0$



PHYSICAL REVIEW B 90, 155126 (2014)

Importance of anisotropic exchange interactions in honeycomb iridates: Minimal model for zigzag antiferromagnetic order in Na_2IrO_3

Yuriy Sizyuk,[1,2] Craig Price,[3] Peter Wölfle,[1,4] and Natalia B. Perkins[1,2]

Outline

I. Kitaev's model & materials

II. Ordering in α-RuCl$_3$

III. Inelastic neutron scattering in α-RuCl$_3$

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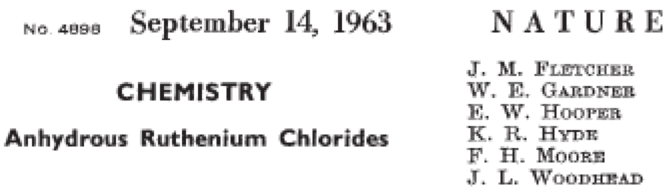
α-$RuCl_3$: quasi - 2D honeycomb material

- Honeycomb lattice
- Ru^{3+} in octahedral low spin
- $J_{1/2} \rightarrow J_{3/2}$ transition ≈ 200 meV

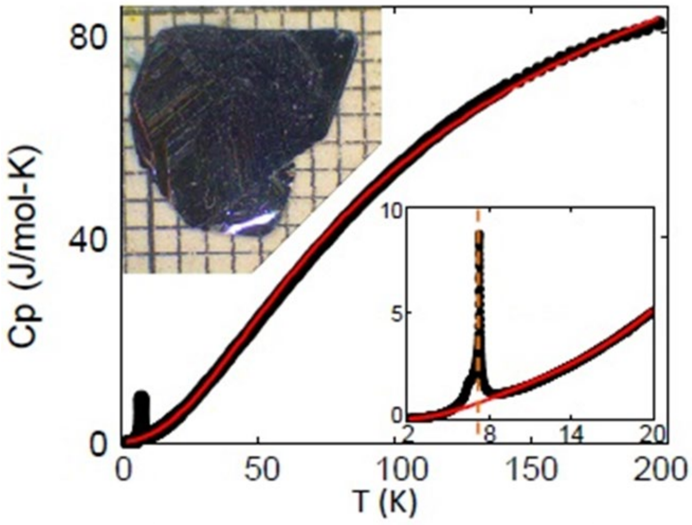


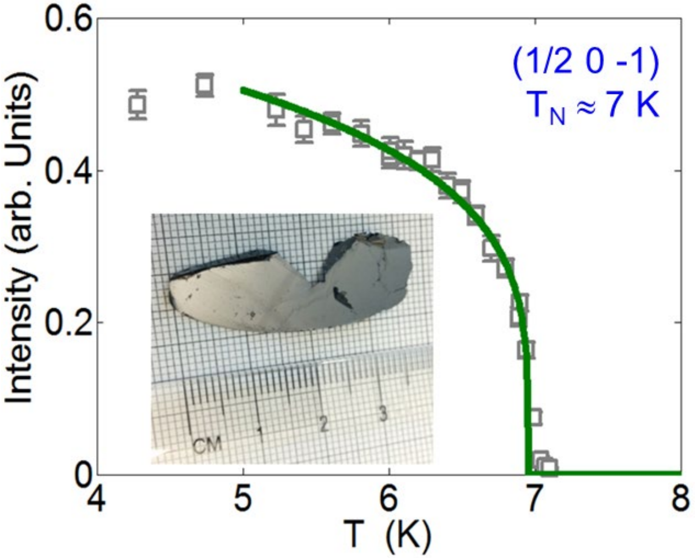
No. 4898 September 14, 1963 NATURE

CHEMISTRY

Anhydrous Ruthenium Chlorides

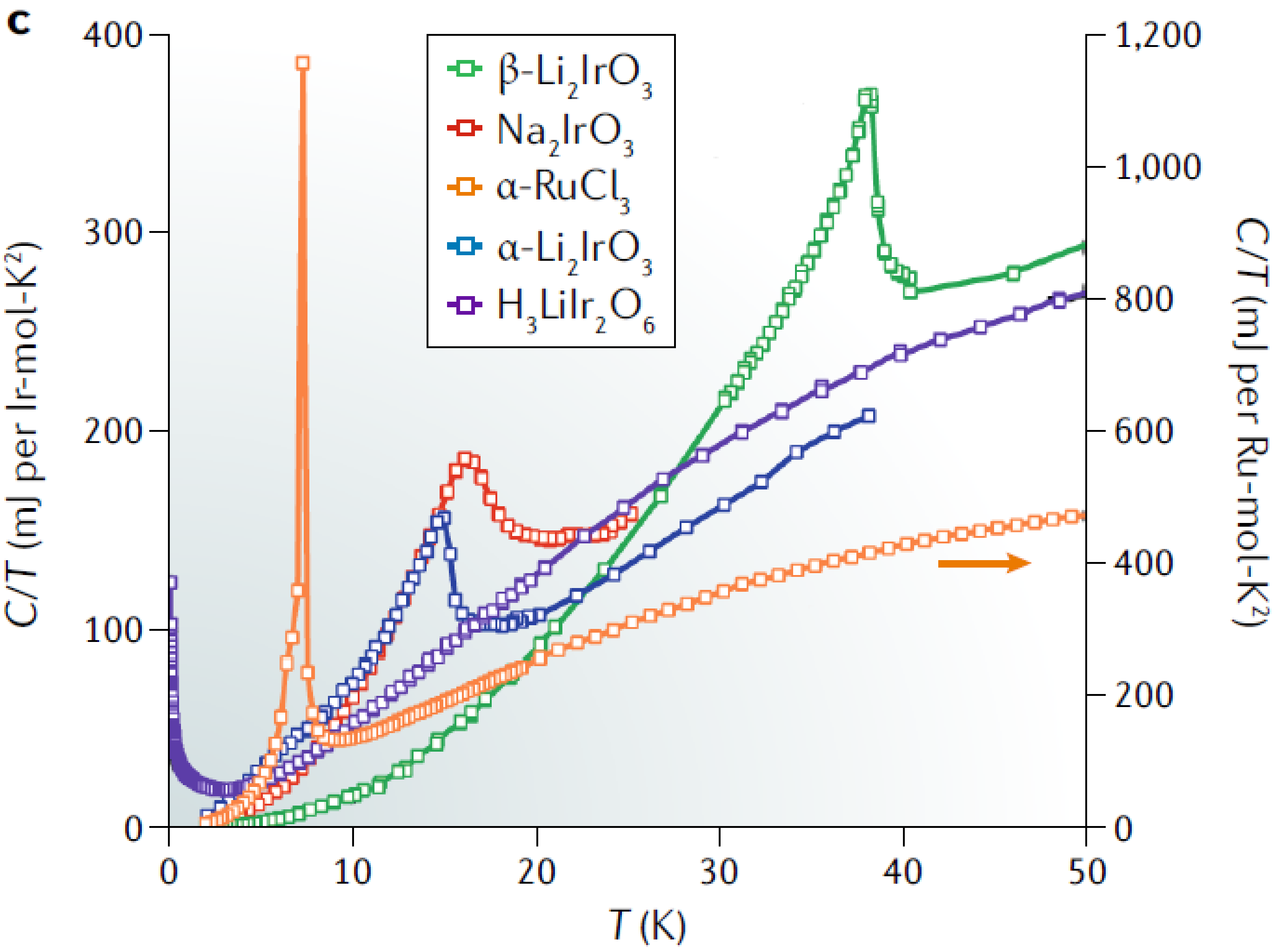
J. M. Fletcher
W. E. Gardner
E. W. Hooper
K. R. Hyde
F. H. Moore
J. L. Woodhead



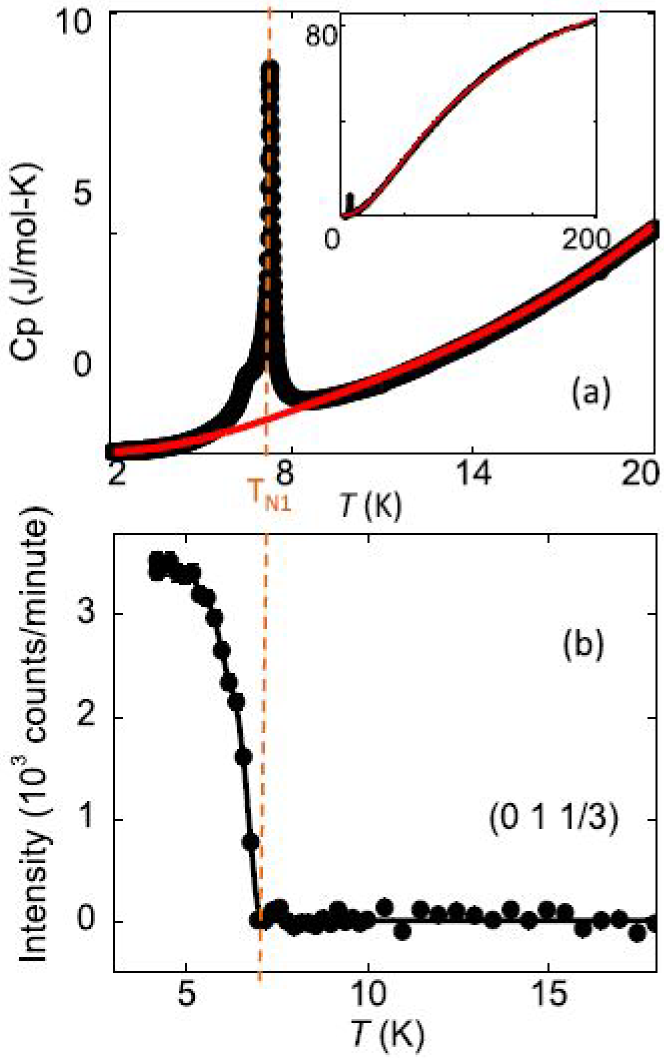


Transition to zig-zag order at $T_N = 7$ K

Comparisons of specific heat



Effect of stacking faults in α-$RuCl_3$



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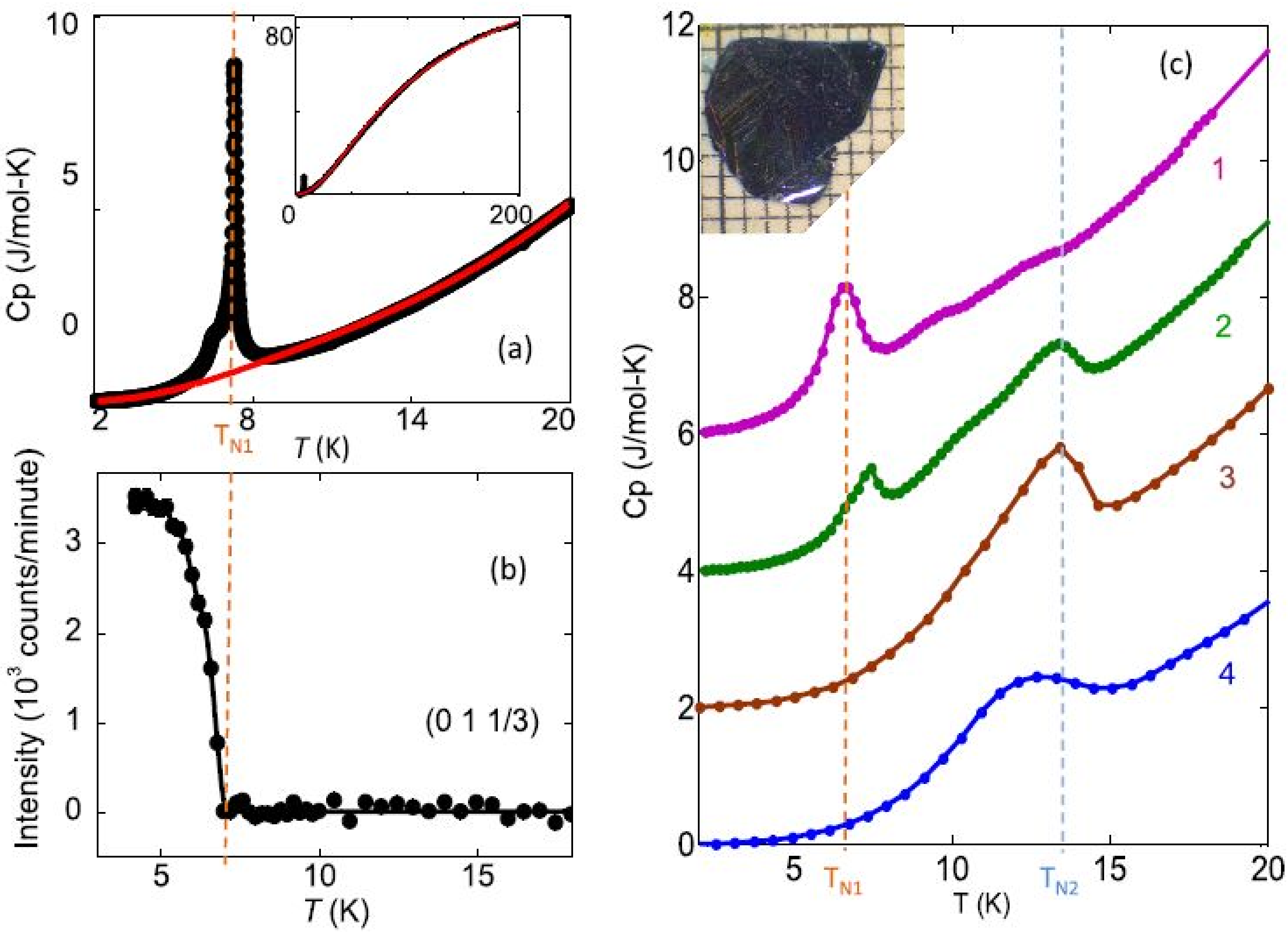
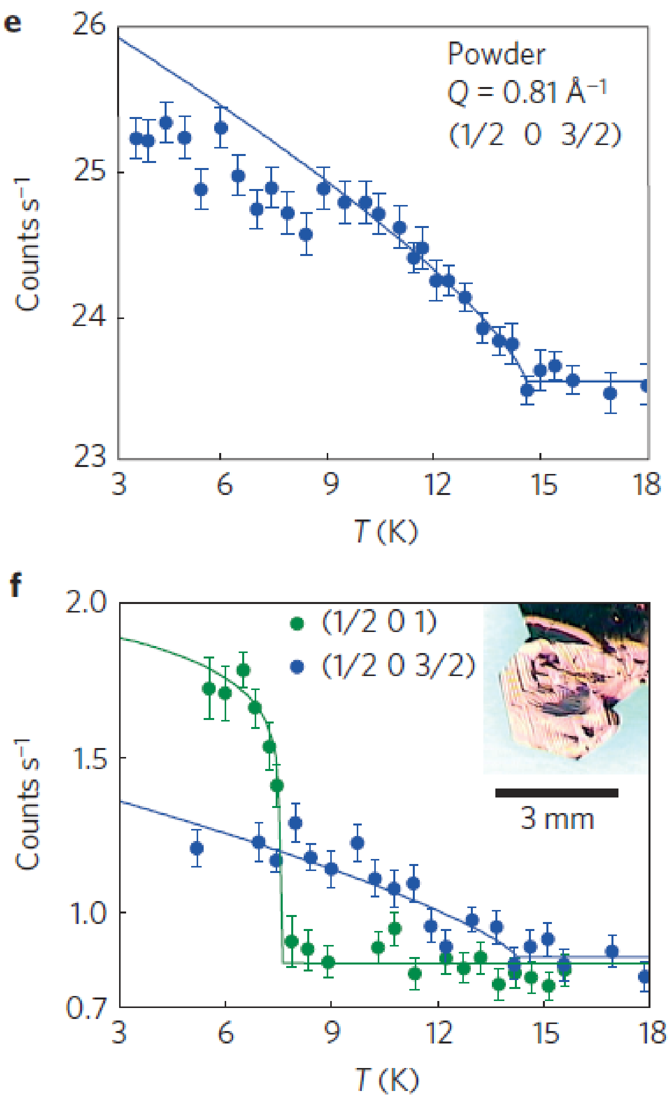
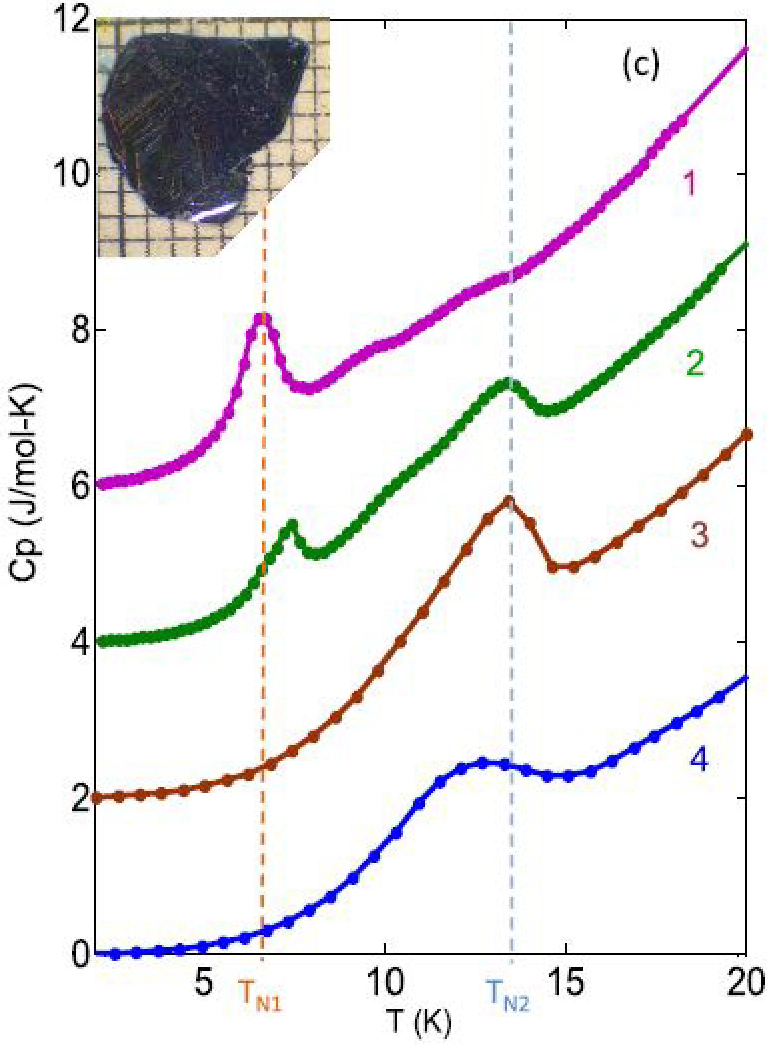


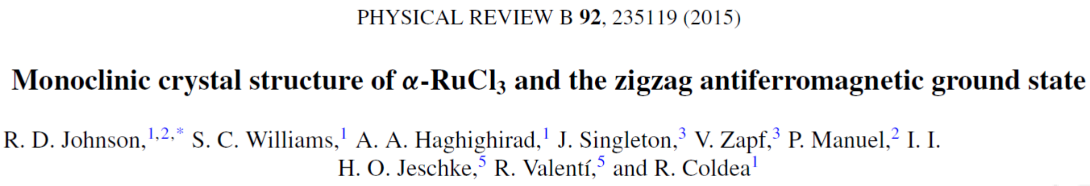
Stacking faults →14 K transition





Magnetic Field Effects

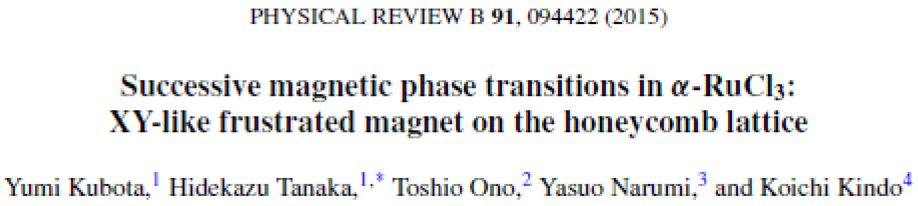
Kubota *et al*., Johnson *et al*. showed that a modest in-plane field kills magnetic order (out of plane needs > 50 Tesla to saturate)



PHYSICAL REVIEW B **92**, 235119 (2015)

Monoclinic crystal structure of α-$RuCl_3$ and the zigzag antiferromagnetic ground state

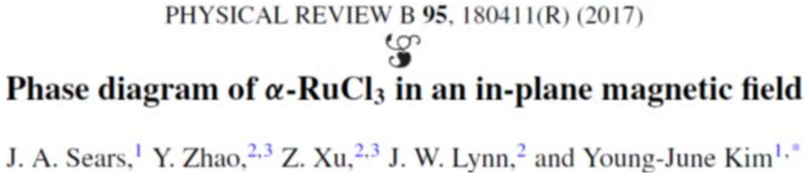
R. D. Johnson,[1,2,*] S. C. Williams,[1] A. A. Haghighirad,[1] J. Singleton,[3] V. Zapf,[3] P. Manuel,[2] I. I.
H. O. Jeschke,[5] R. Valentí,[5] and R. Coldea[1]



PHYSICAL REVIEW B **91**, 094422 (2015)

Successive magnetic phase transitions in α-$RuCl_3$: XY-like frustrated magnet on the honeycomb lattice

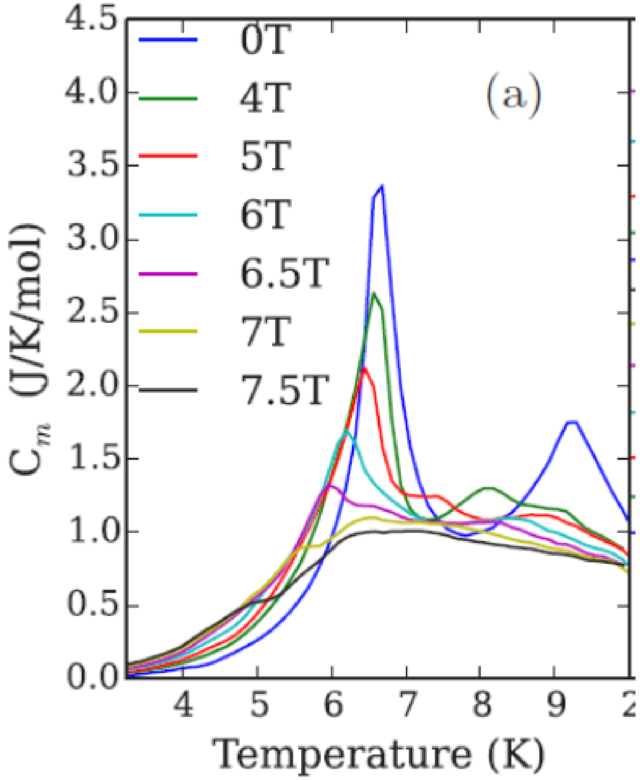
Yumi Kubota,[1] Hidekazu Tanaka,[1,*] Toshio Ono,[2] Yasuo Narumi,[3] and Koichi Kindo[4]



PHYSICAL REVIEW B **95**, 180411(R) (2017)

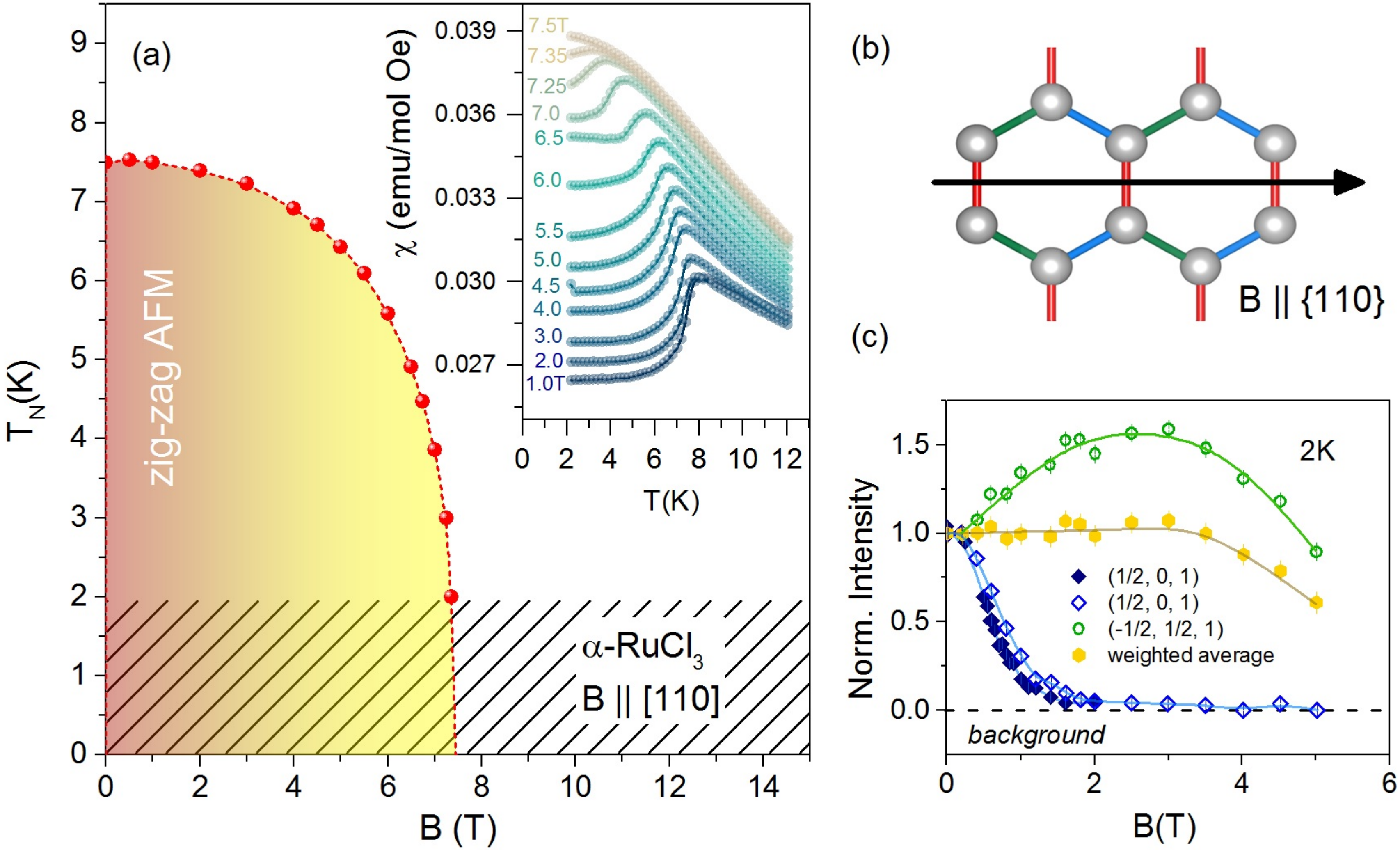
Phase diagram of α-$RuCl_3$ in an in-plane magnetic field

J. A. Sears,[1] Y. Zhao,[2,3] Z. Xu,[2,3] J. W. Lynn,[2] and Young-June Kim[1,*]

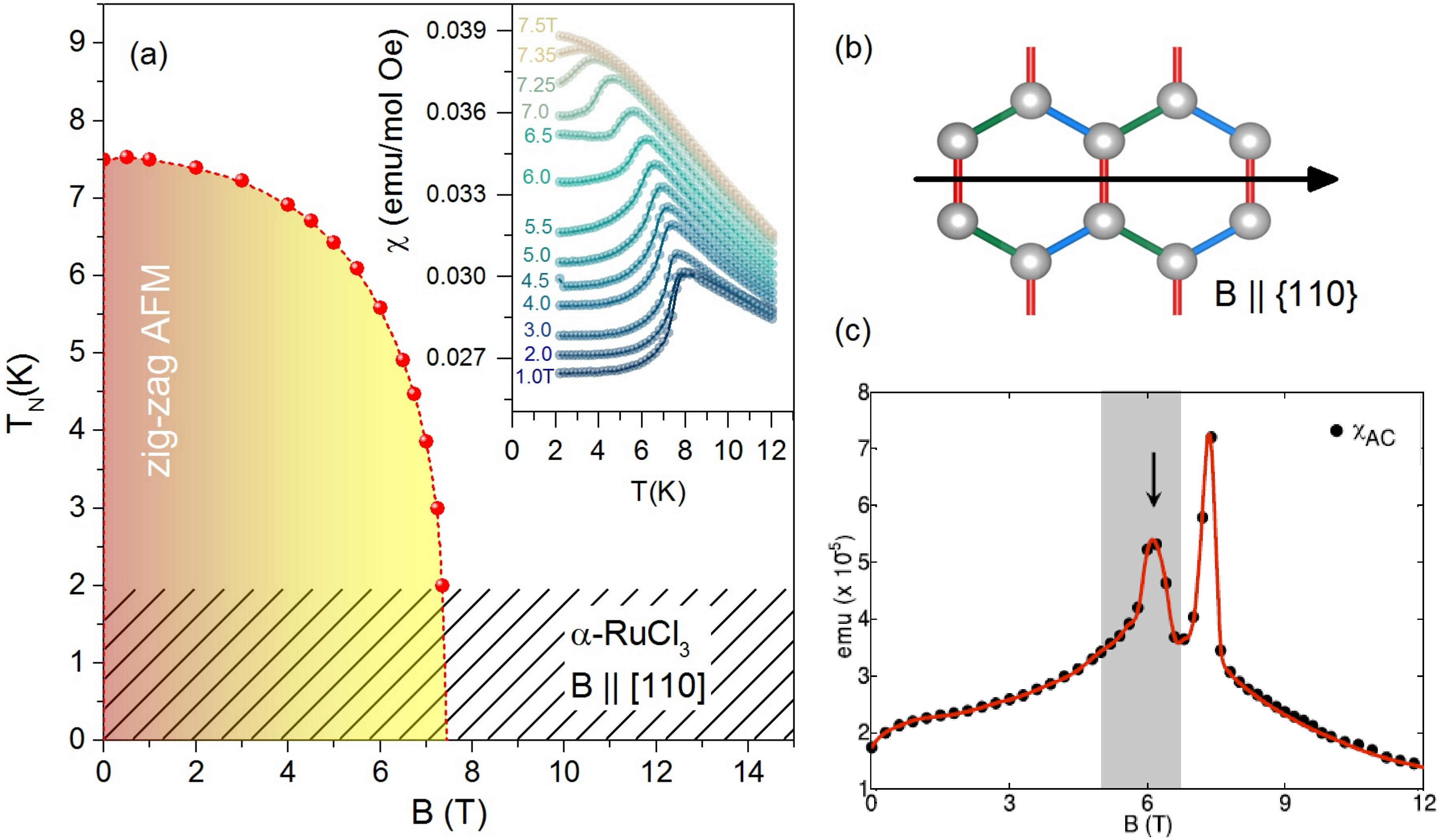


Field dependence of T_N

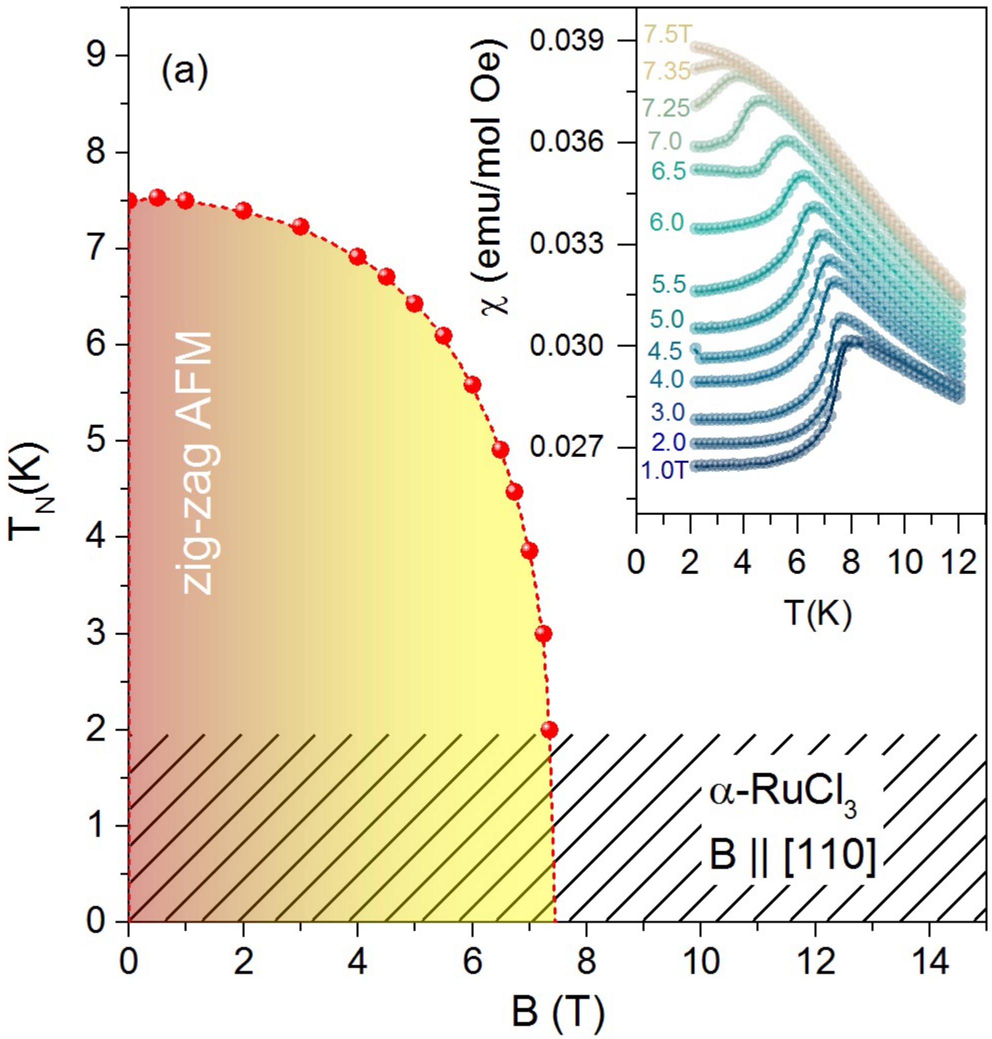
$B_C \approx 7.3$ T

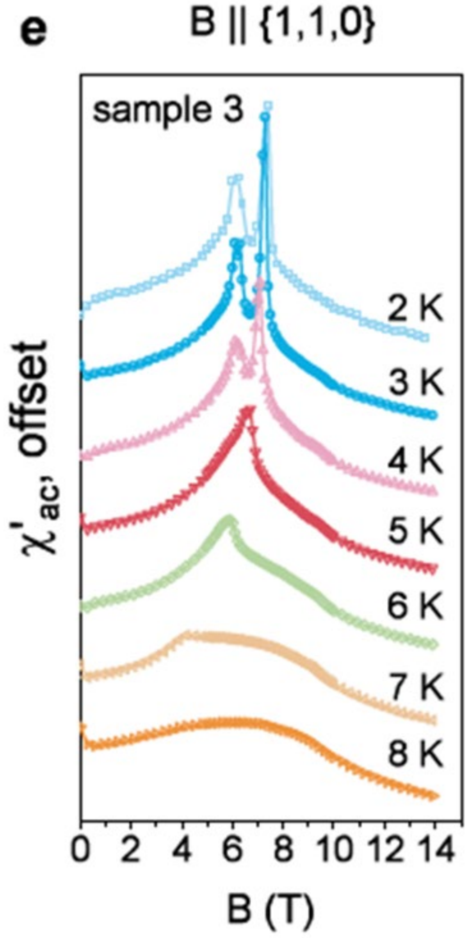


Additional ordered phase 6 – 7.3 T

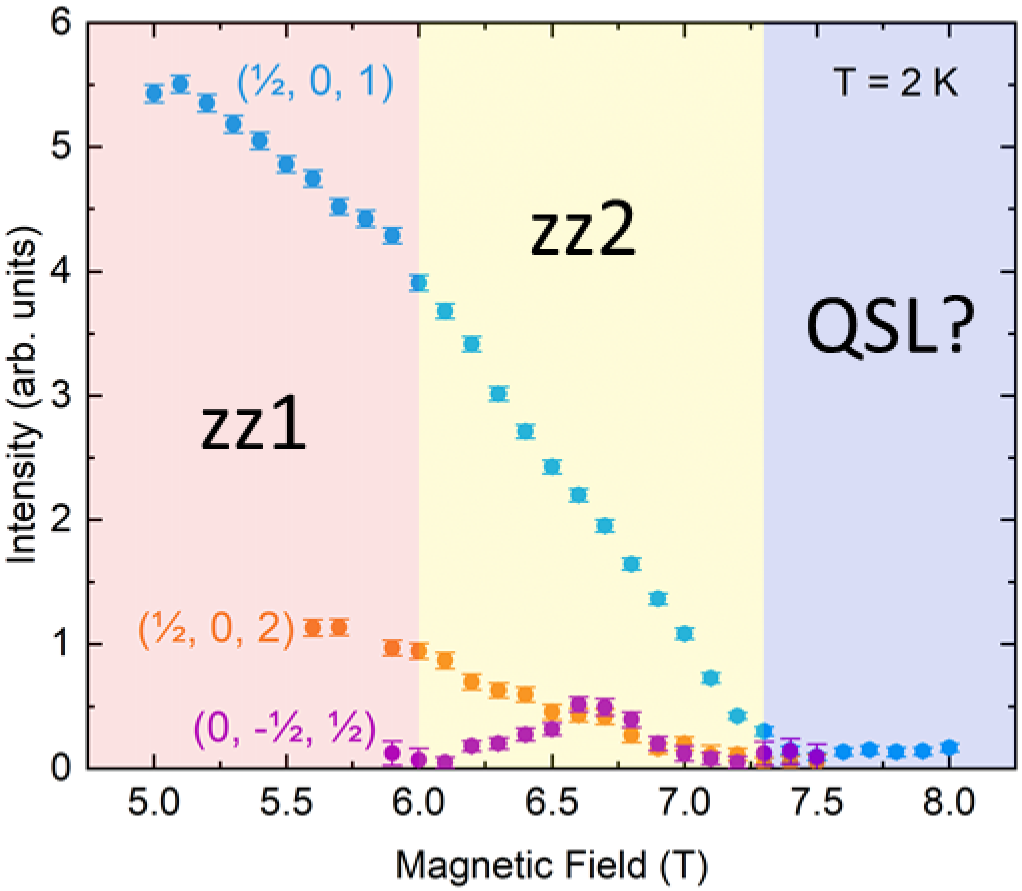


Additional ordered phase 6 – 7.3 T

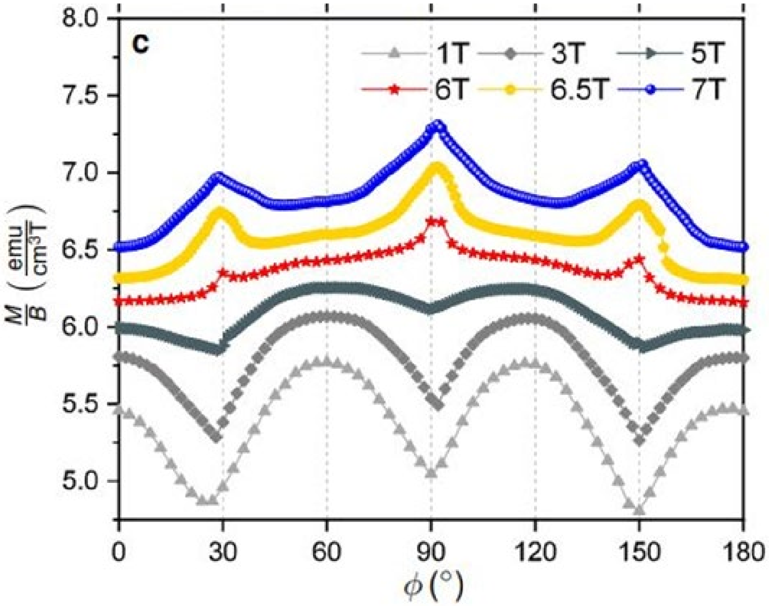


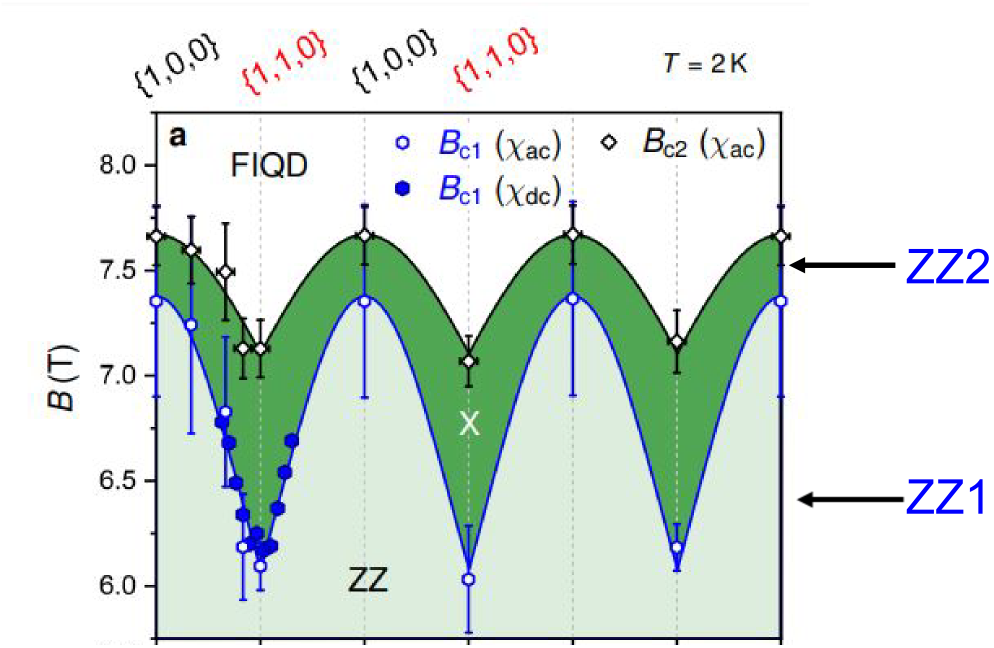


Diffraction: 2nd zigzag w/ different stacking



In-plane field direction dependence





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Unpolarized neutron intensity for magnetic scattering:

$$\mathrm{I}^{\mathrm{mag}}(\mathbf{Q},\omega) \propto \left|f(\mathbf{Q})\right|^2 \sum_{\alpha,\beta}\left(\delta^{\alpha\beta} - \hat{\mathrm{Q}}^{\alpha}\hat{\mathrm{Q}}^{\beta}\right)\mathrm{S}^{\alpha\beta}(\mathbf{Q},\omega)$$

magnetic form factor

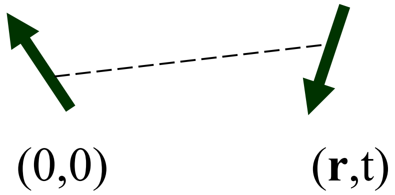
magnetic structure factor

components of spin α,β = x,y,z.

unit vector in direction of **Q**

Magnetic Structure Factor:

$$\mathrm{S}^{\alpha\beta}(\mathbf{Q},\omega) = \int \left\langle m^{\alpha}(0,0)m^{\beta}(\mathbf{r},t)\right\rangle e^{i(\mathbf{Q}\cdot\mathbf{r}-\omega t)}d\mathbf{r}dt$$



$$m^{\alpha} = L^{\alpha} + 2S^{\alpha}$$

$$\mathrm{S}^{\alpha\beta}(\mathbf{Q},\omega) \propto \mathrm{Im}\left\{\chi^{\alpha\beta}(\mathbf{Q},\omega)\right\}$$

Fluctuation dissipation theorem

For magnons usually $\mathrm{S}(\mathbf{Q},\omega) \propto \delta(\omega-\omega_{\mathbf{Q}})$

Exactly solvable quantum spin system

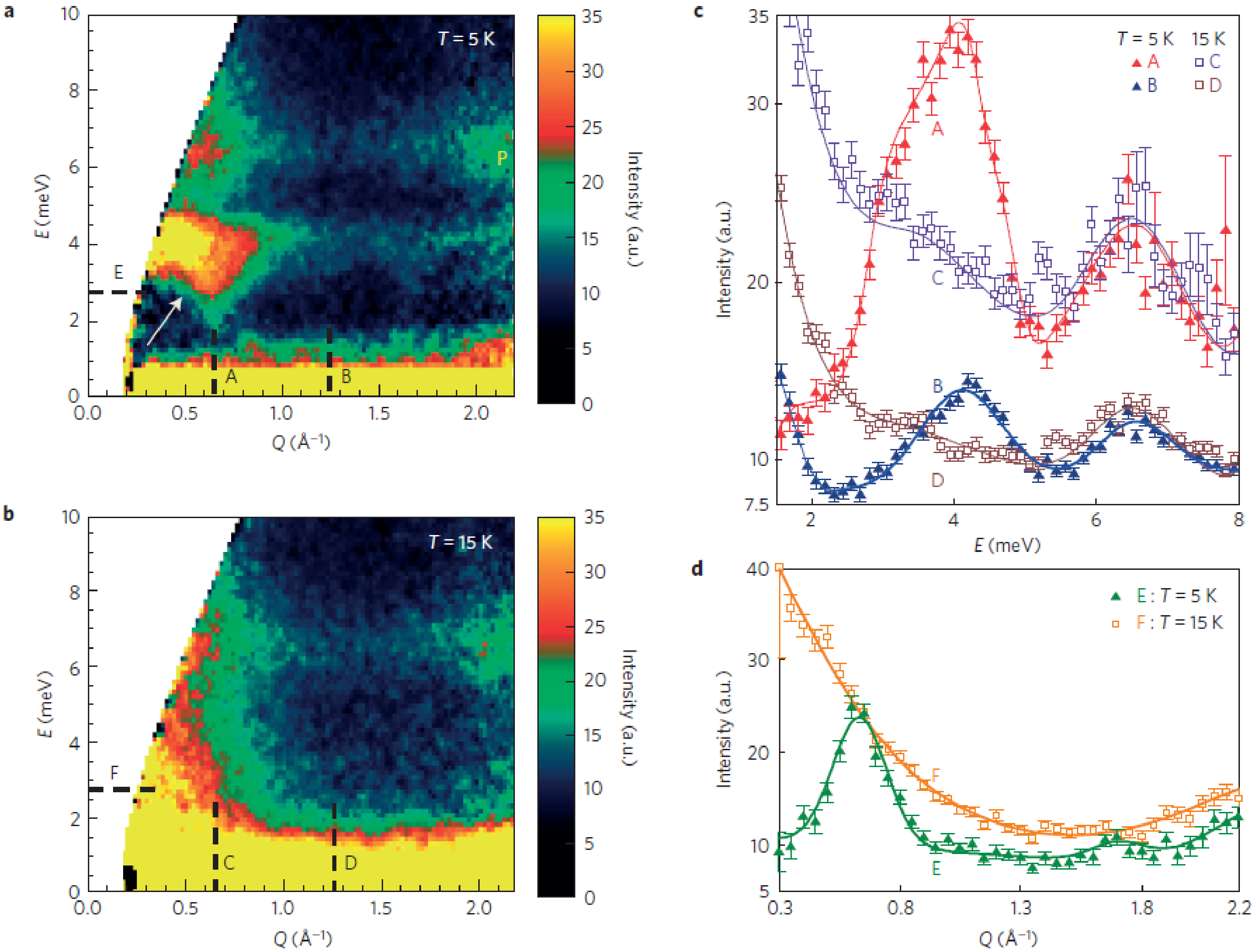
T=0: $$S^{\alpha\alpha}(\vec{Q},\omega) = \sum_{E} \left| \left\langle E \middle| S^{\alpha}_{\vec{Q}} \middle| G \right\rangle \right|^2 \delta(\omega - E)$$

T>0: $$S^{\alpha\alpha}(\vec{Q},\omega) = Z^{-1} \sum_{E,E'} e^{-\beta E} \left| \left\langle E' \middle| S^{\alpha}_{\vec{Q}} \middle| E \right\rangle \right|^2 \delta(\omega + E - E')$$

where $Z = \sum_{E} e^{-\beta E}$ and $S^{\alpha}_{\vec{Q}} = \sum_{\vec{R}} S^{\alpha}_{\vec{R}} e^{-i\vec{Q}\cdot\vec{R}}$

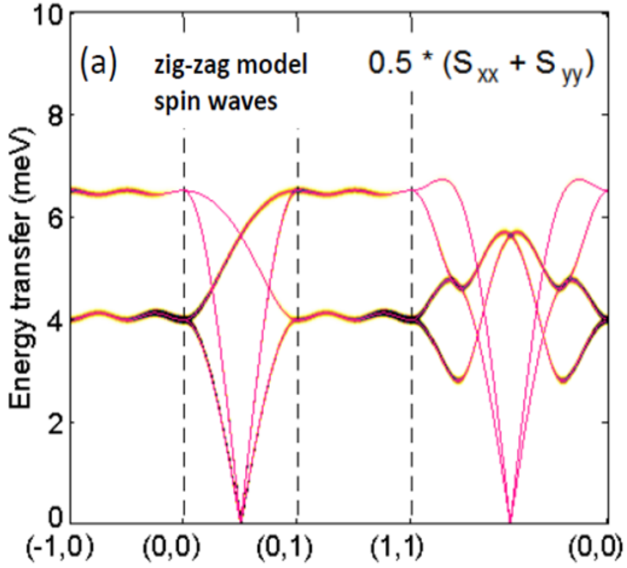
Neutron scattering is sensitive to matrix elements where ΔS or ΔL = 0,±1

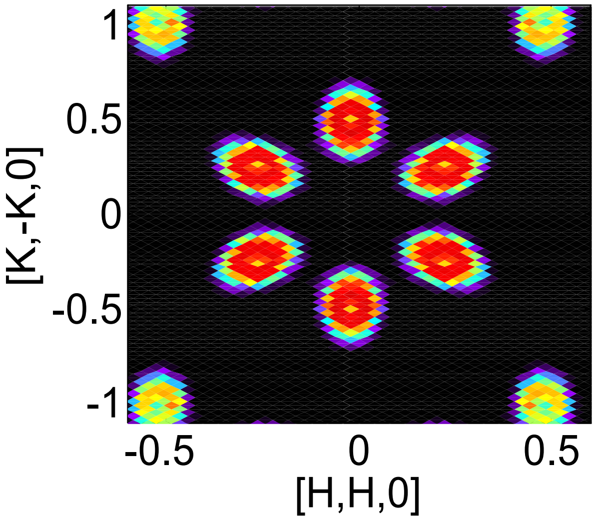
α-$RuCl_3$ powder – inelastic neutron scattering



Nature Materials **15**, 733 (2016).

Expectations for spin waves in a zigzag state





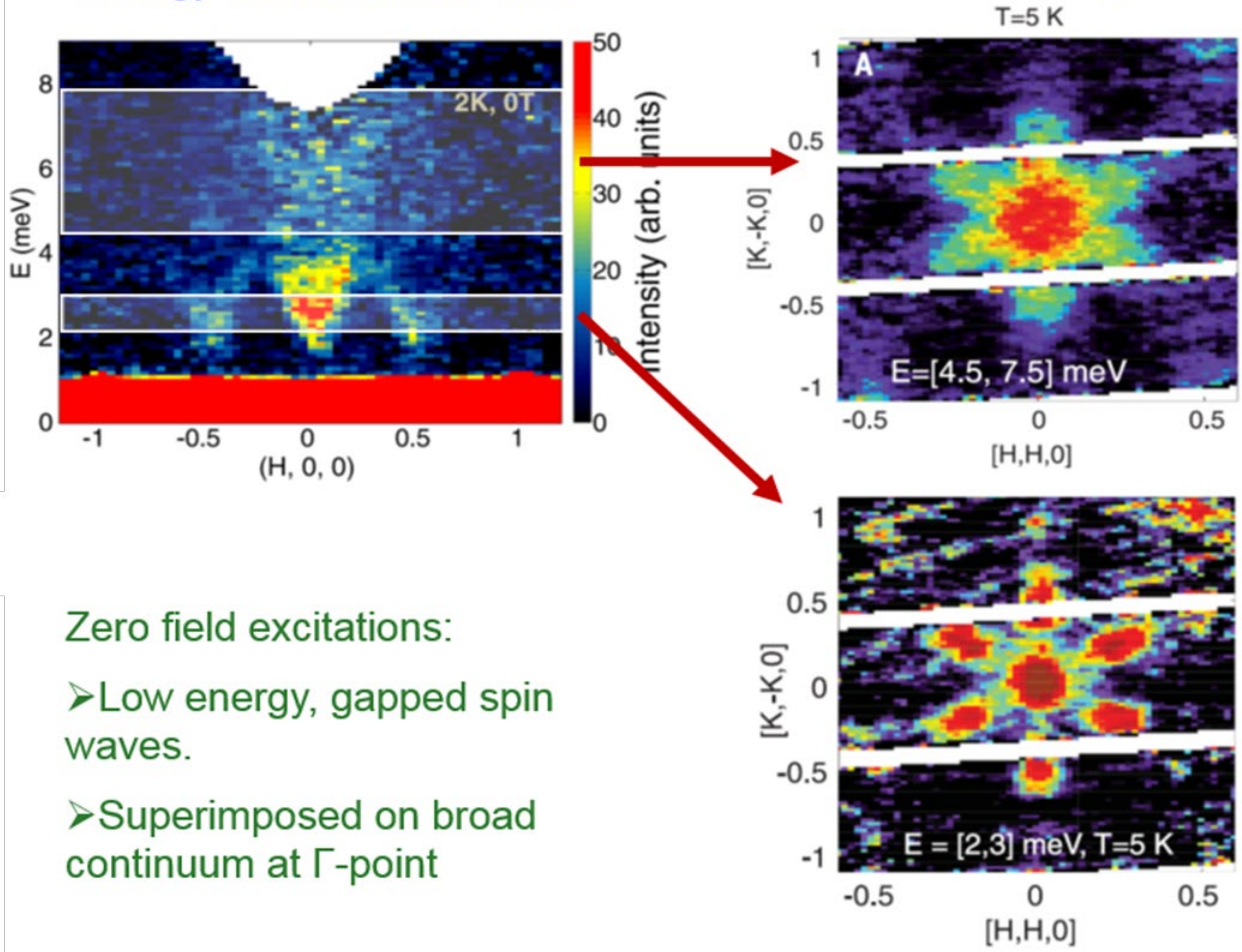
- Dispersion minima at ordering wavevectors (M points)
- Low energy constant E slices show cone shaped dispersion surfaces around the M points
- Less general, but true for Heisenberg-Kitaev model:

  Γ points show flat modes sharp in energy

α-RuCl$_3$ single crystal - INS

Energy vs wavevector slice

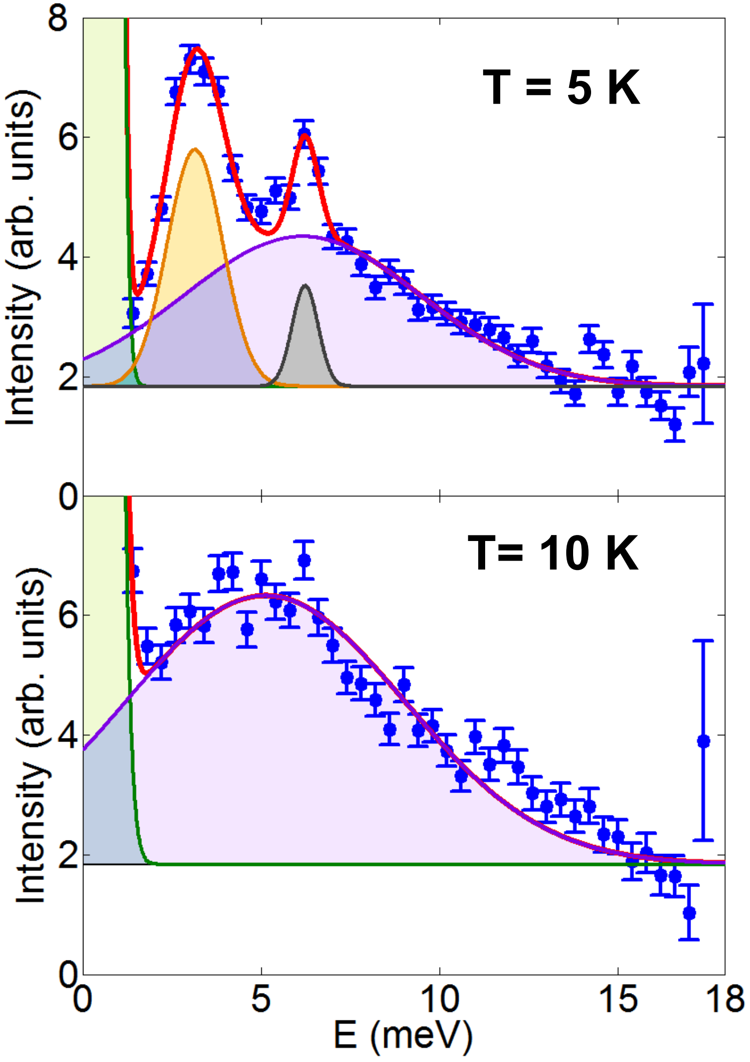
Constant energy slice

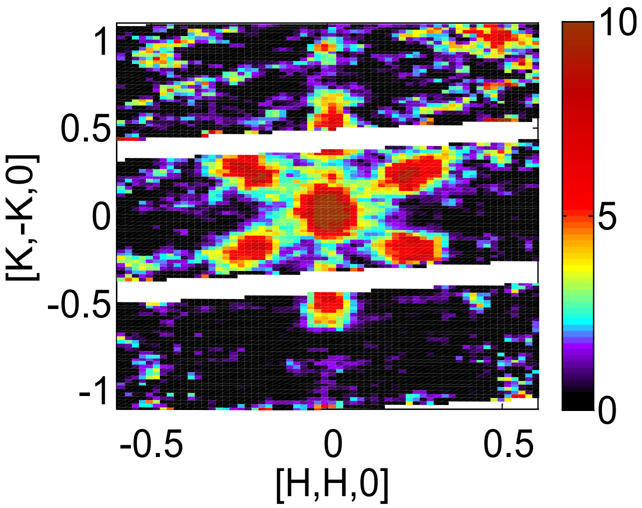


Zero field excitations:

- ➢Low energy, gapped spin waves.
- ➢Superimposed on broad continuum at Γ-point

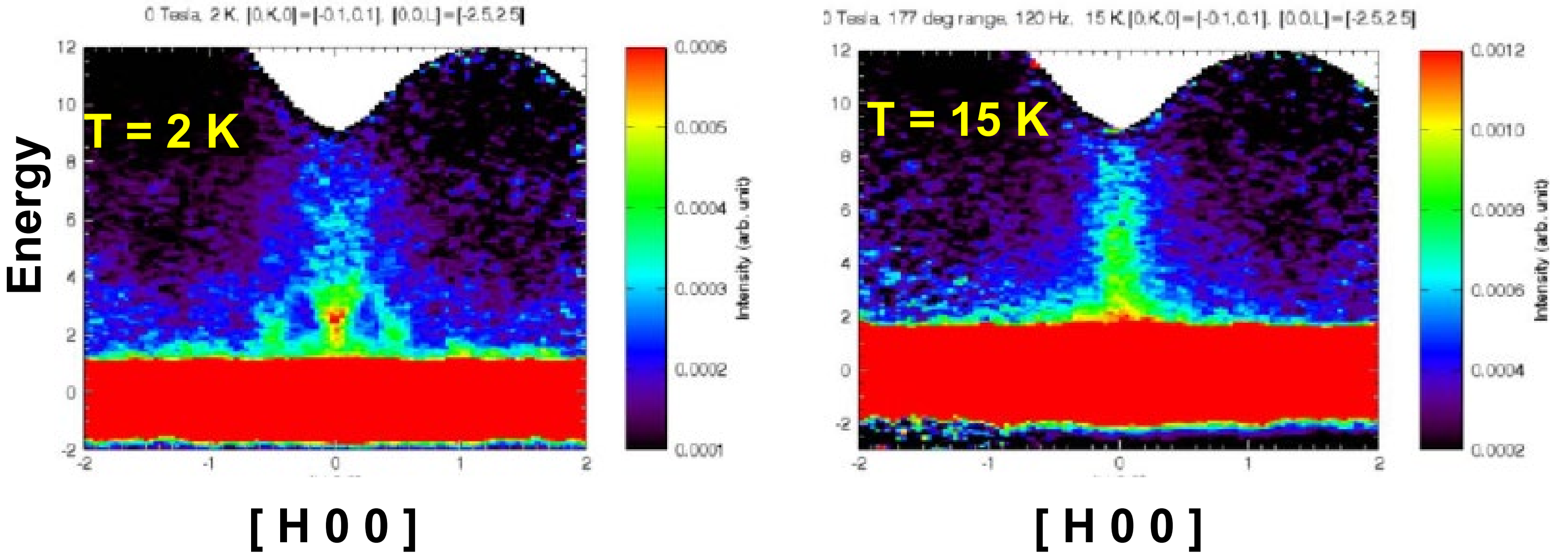
Experiment: Γ point signal inconsistent with SW





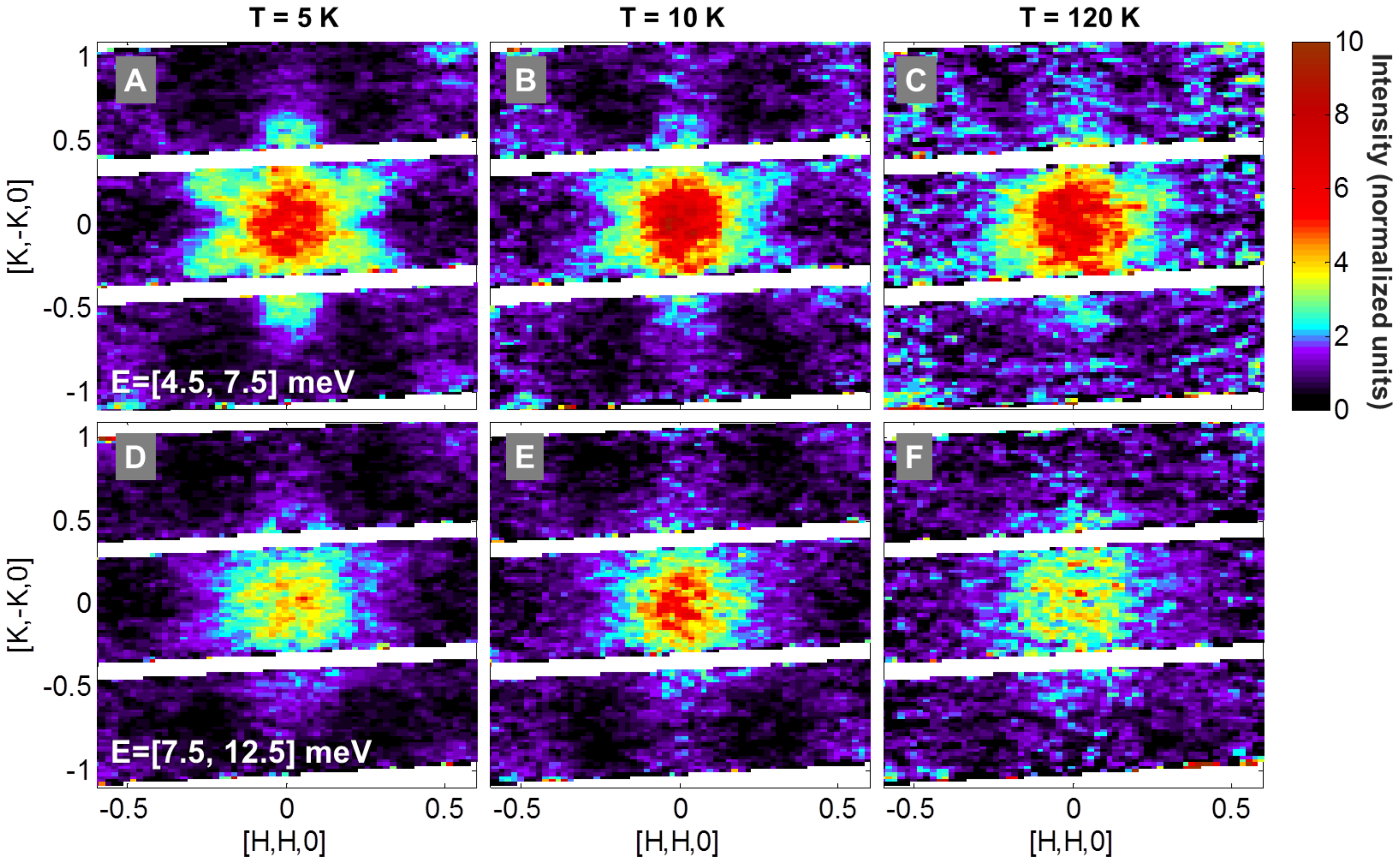
- Below TN - Γ point shows spin waves plus continuum
- Above TN – only continuum remains
- Large sustained continuum at Γ is absent in spin wave theory

Scattering through the Brillouin zone



- At $T_N \approx 7$ K the spin waves disappear throughout the Brillouin zone
- Above T_N the continuum near the Γ point persists

Q,T dependence of the continuum scattering

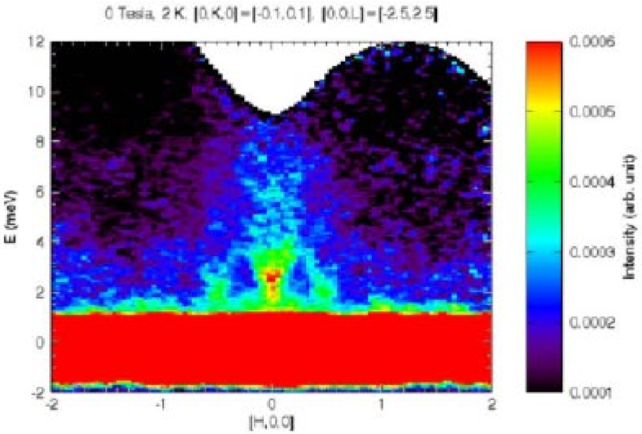


- circular column centered on H=K=0, extending to higher energies
- at low T, moderate energy SW peaks and column merge and scattering resembles a six pointed Star of David
- scattering persists to high T

How does field affect the magnetic excitations?

- Does killing order with an applied field lead to a QSL?

2K, 0T



How does field affect the magnetic excitations?

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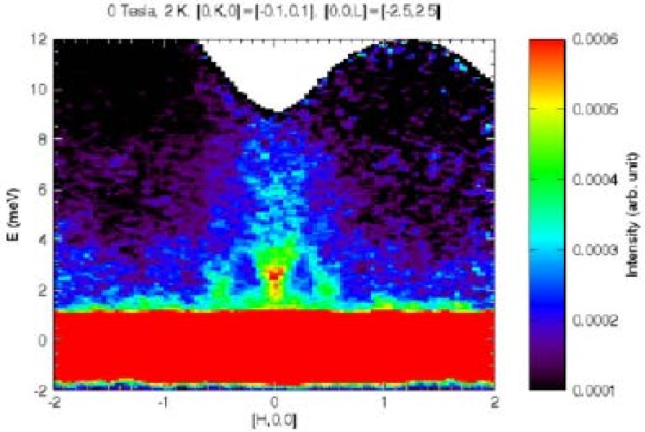
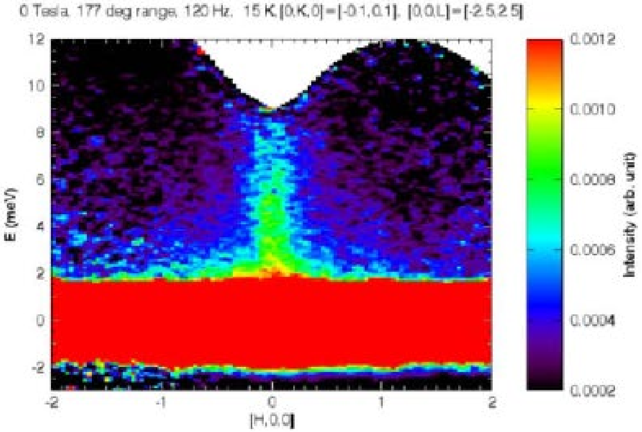
2K, 0T



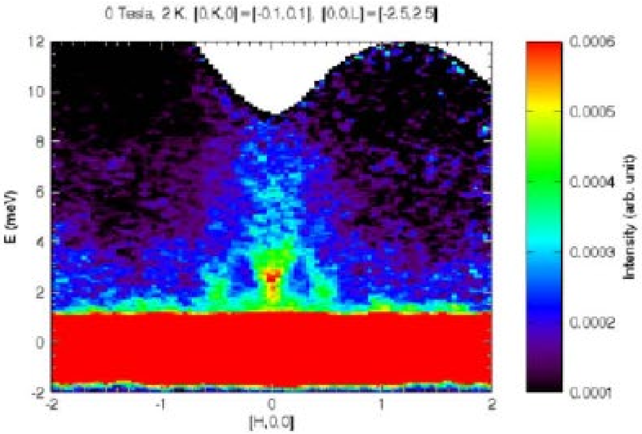


15K, 0T

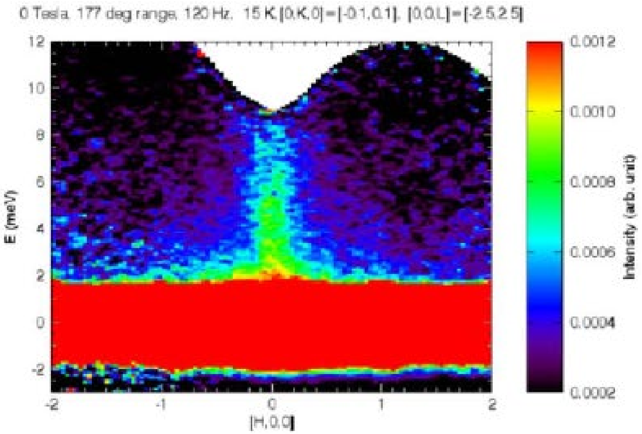
How does field affect the magnetic excitations?

- Does killing order with an applied field lead to a QSL?

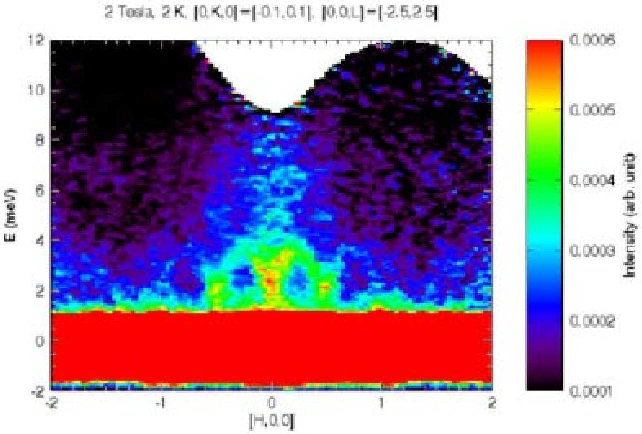
2K, 0T



15K, 0T

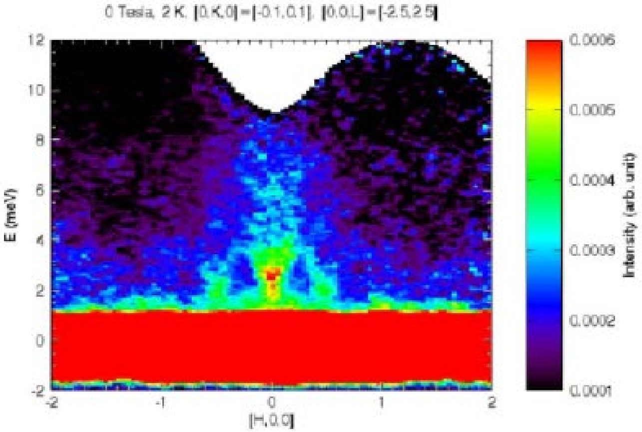


2K, 2T

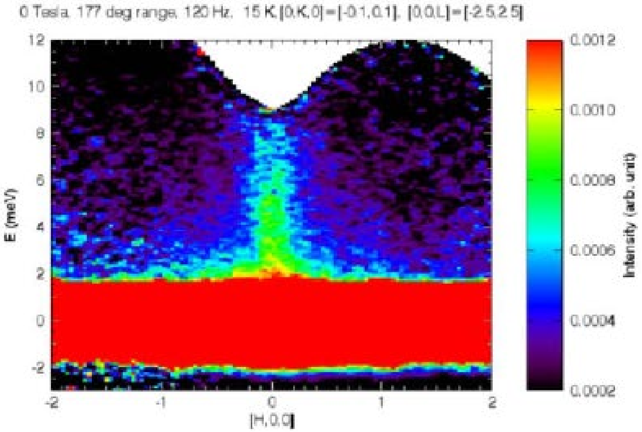


How does field affect the magnetic excitations?

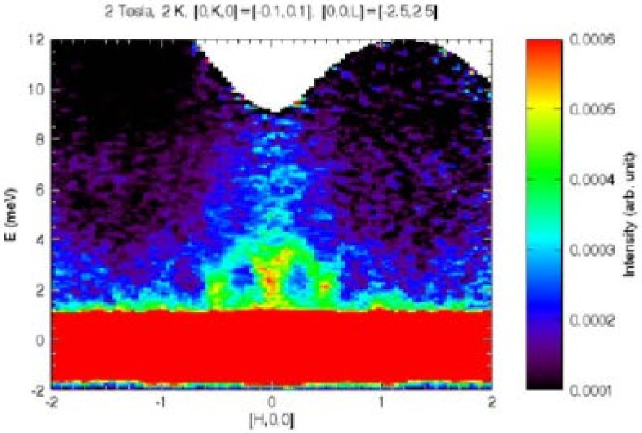
2K, 0T



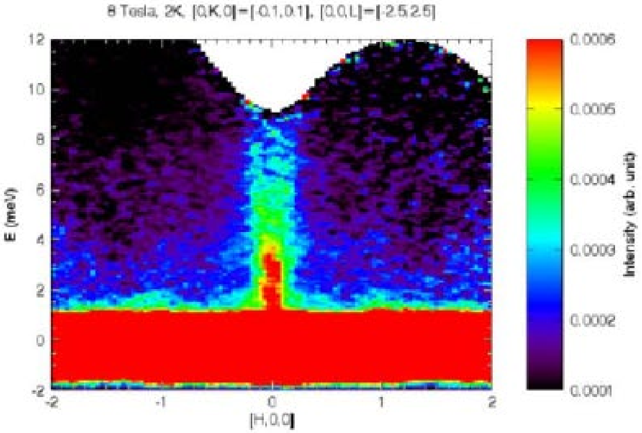
15K, 0T



2K, 2T

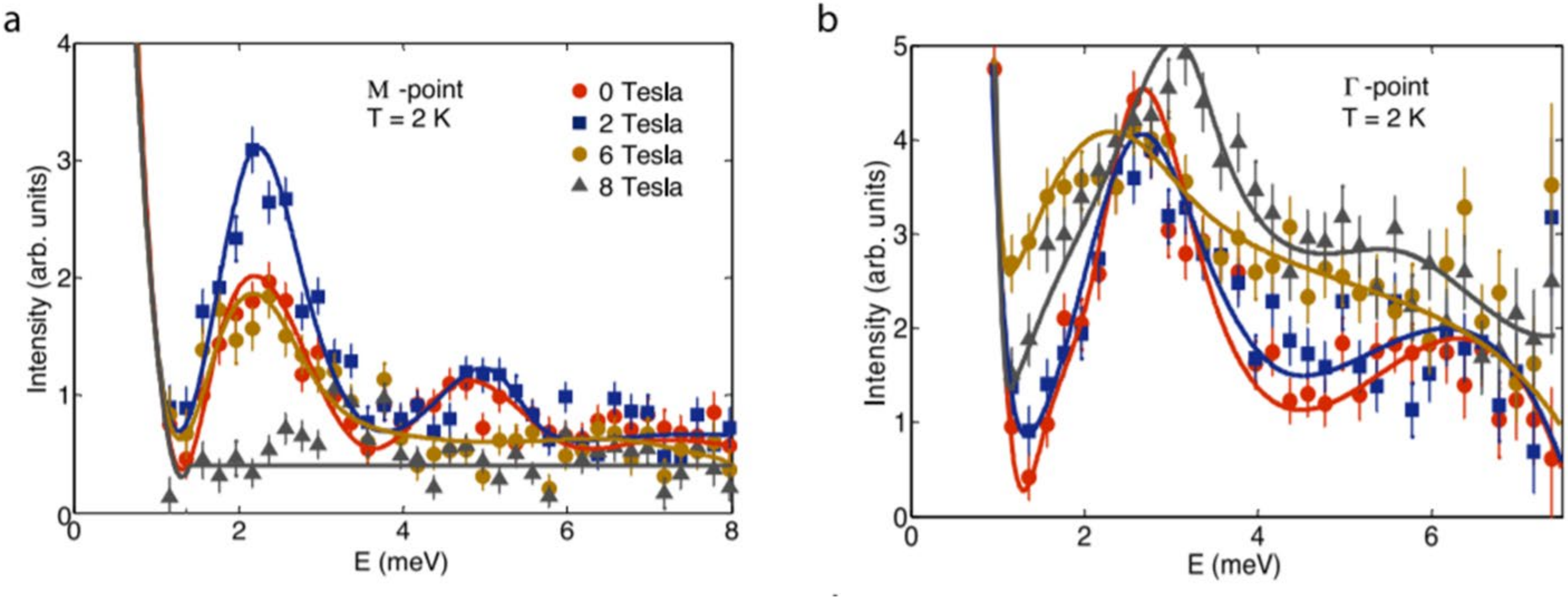


2K, 8T



Npj Quantum Materials **3**, 8 (2018).

Is the scattering gapped at the Γ point?



Npj Quantum Materials **3**, 8 (2018).

Outline

I. Kitaev’s model & materials

II. Ordering in α-RuCl$_3$

III. Inelastic neutron scattering in α-RuCl$_3$

IV. Higher fields, magnetocaloric effect, T-B phase diagram

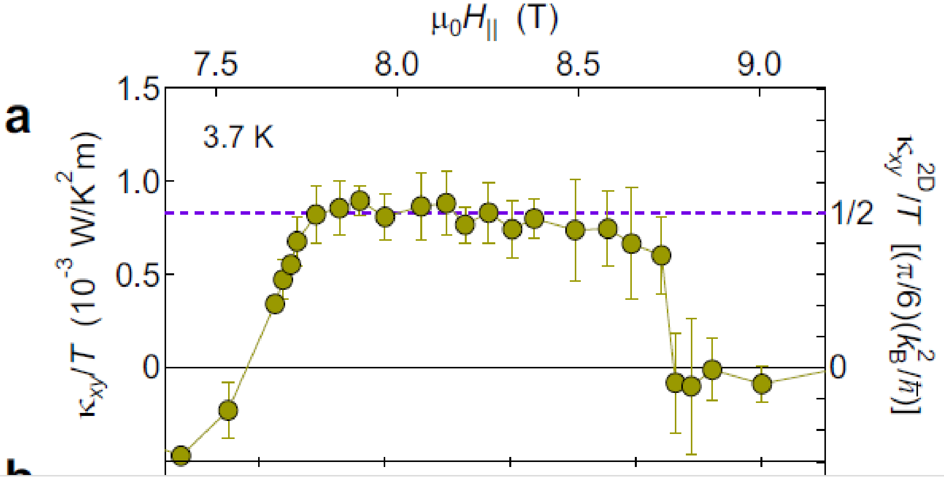
Evidence of fractionalization from thermal Hall ?

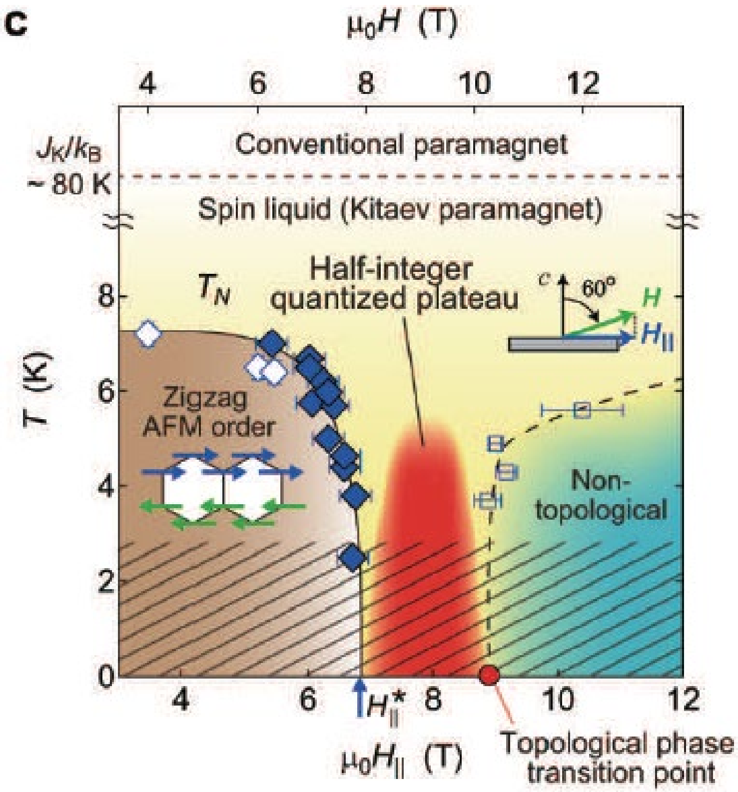
Majorana quantization and half-integer thermal quantum Hall effect in a Kitaev spin liquid

Y. Kasahara[1], T. Ohnishi[1], N. Kurita[2], H. Tanaka[2], J. Nasu[2], Y. Motome[3], T. Shibauchi[4], and Y. Matsuda[1]

κ_{xy}^{2D} reaches a quantum plateau as a function of applied magnetic field. That is, κ_{xy}^{2D}/T attains a quantization value of $(\pi/12)(k_B^2/\hbar)$, which is exactly half of κ_{xy}^{2D}/T in the integer QHE. This half-integer thermal Hall conductance observed in a bulk material is a direct signature of topologically protected chiral edge currents of charge neutral Majorana fermions, particles that are their own antiparticles, which possess half degrees of freedom of conventional fermions [13–16]. These signatures demonstrate the fractionalization of spins into itinerant Majorana fermions and Z_2 fluxes predicted in a Kitaev QSL [1, 3]. Above

Nature **559**, 227–231 (2018)





Field dependence of scattering at specific Γ points

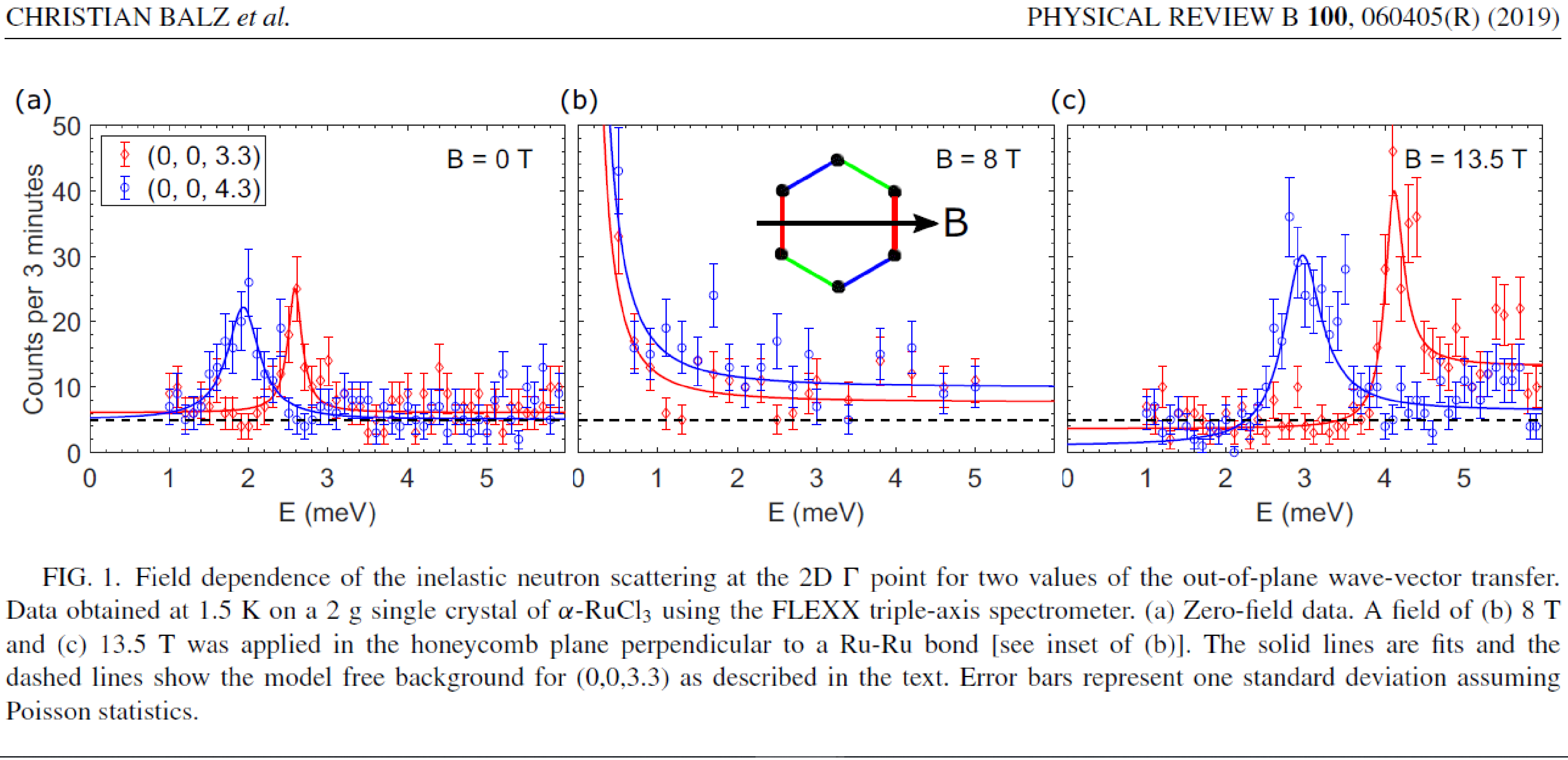


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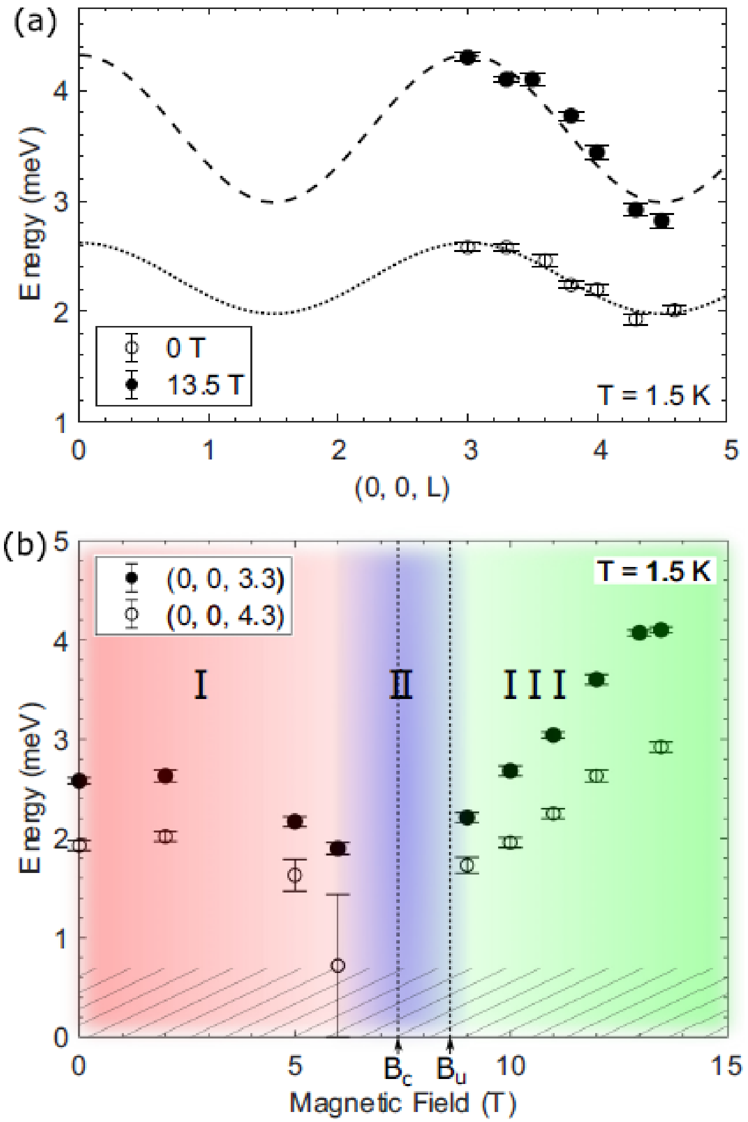
FIG. 1. Field dependence of the inelastic neutron scattering at the 2D Γ point for two values of the out-of-plane wave-vector transfer. Data obtained at 1.5 K on a 2 g single crystal of α-$RuCl_3$ using the FLEXX triple-axis spectrometer. (a) Zero-field data. A field of (b) 8 T and (c) 13.5 T was applied in the honeycomb plane perpendicular to a Ru-Ru bond [see inset of (b)]. The solid lines are fits and the dashed lines show the model free background for (0,0,3.3) as described in the text. Error bars represent one standard deviation assuming Poisson statistics.

L dispersion and band width

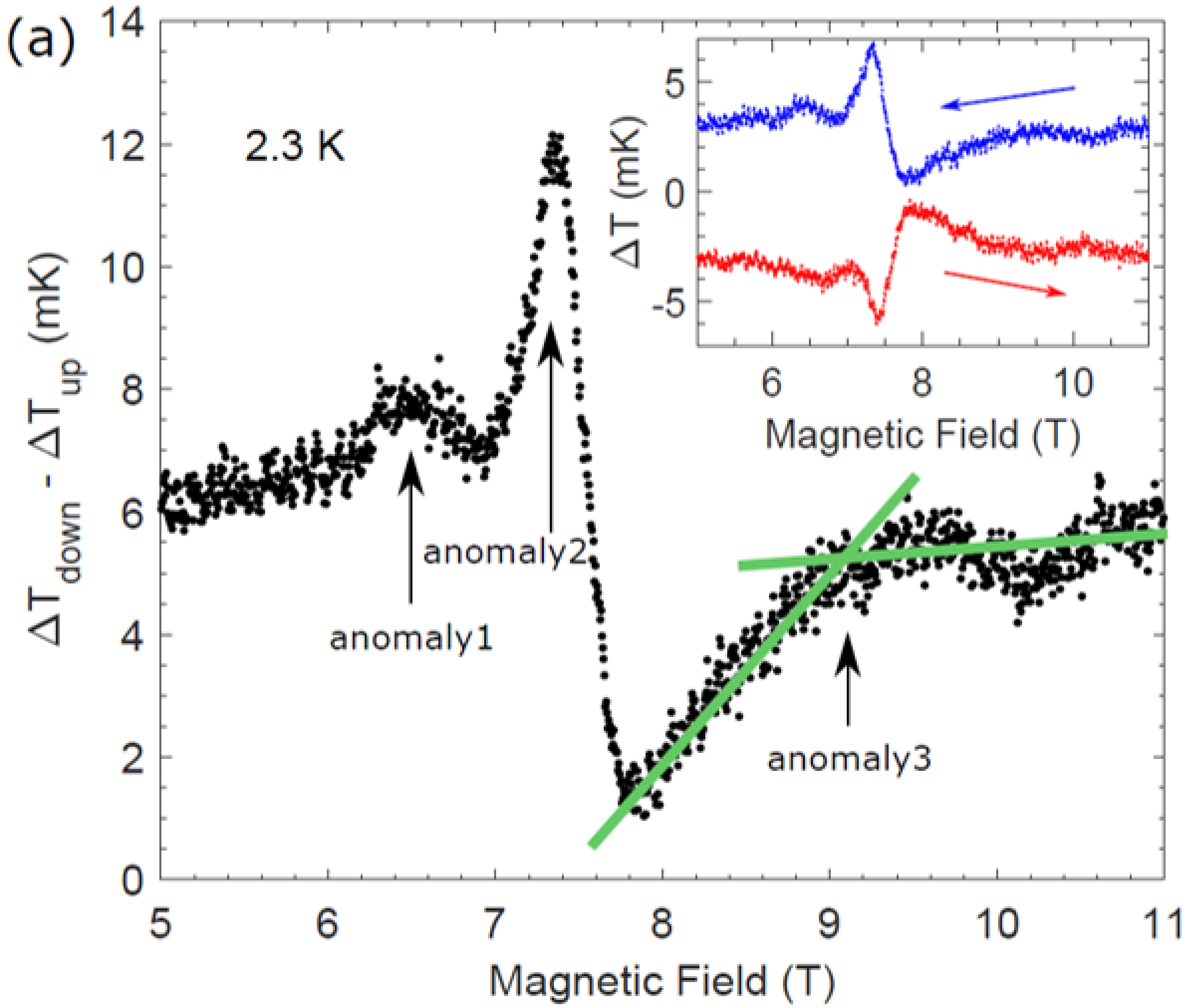


- The dispersion in L is a measure of the magnetic interactions perpendicular to the plane

- The reduction of the bandwidth near the region where magnons are not detected is a signature of enhanced two-dimensionality

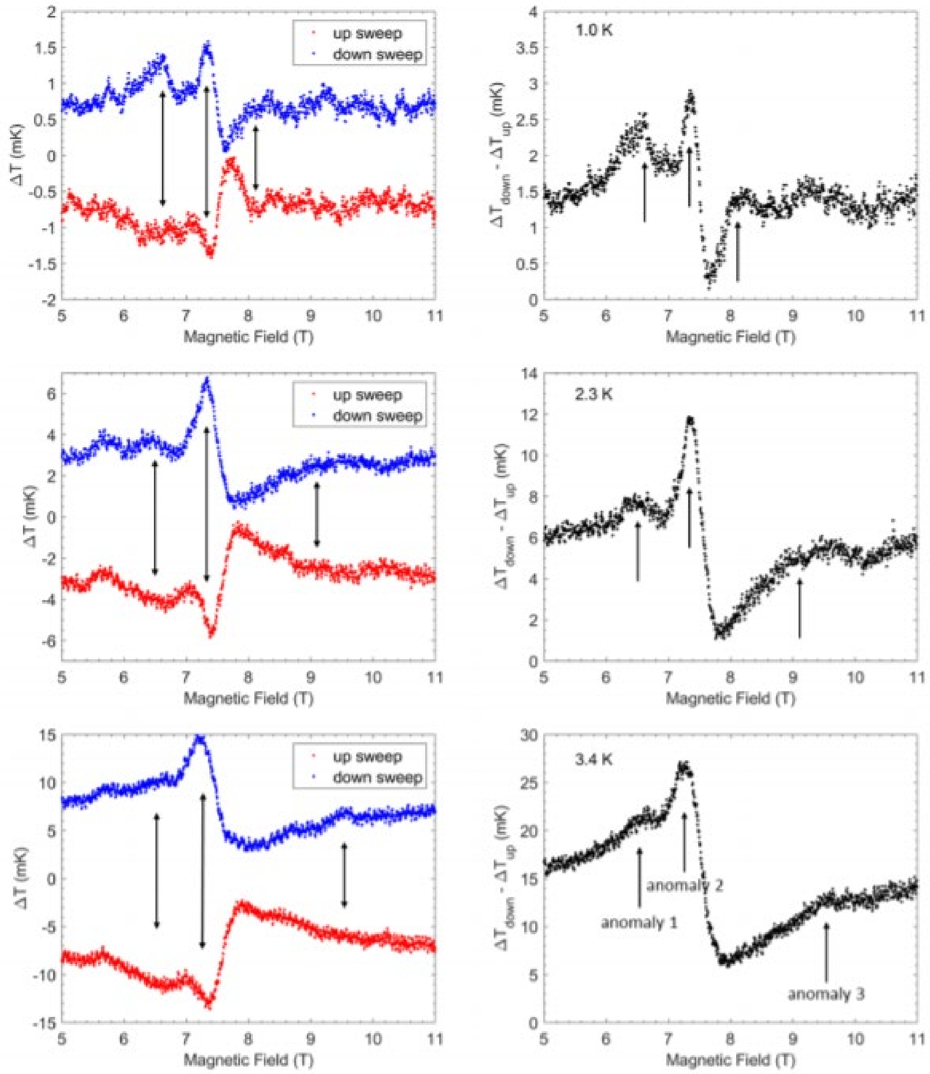
Magnetocaloric effect

Y. Takano group



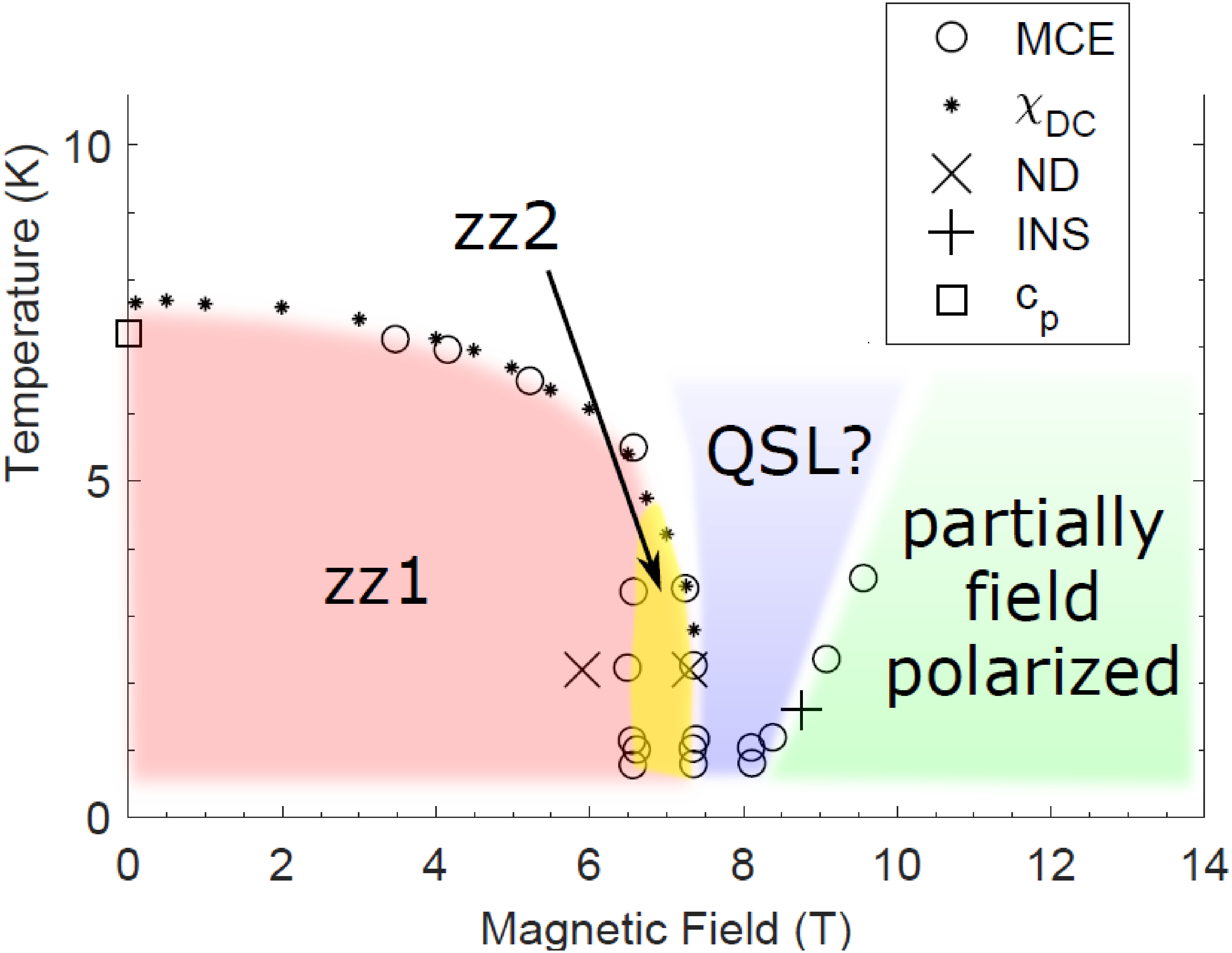
$$\Delta T \propto -T\left(\frac{\partial M}{\partial T}\right)_H \Delta H$$

Magnetocaloric effect at different T

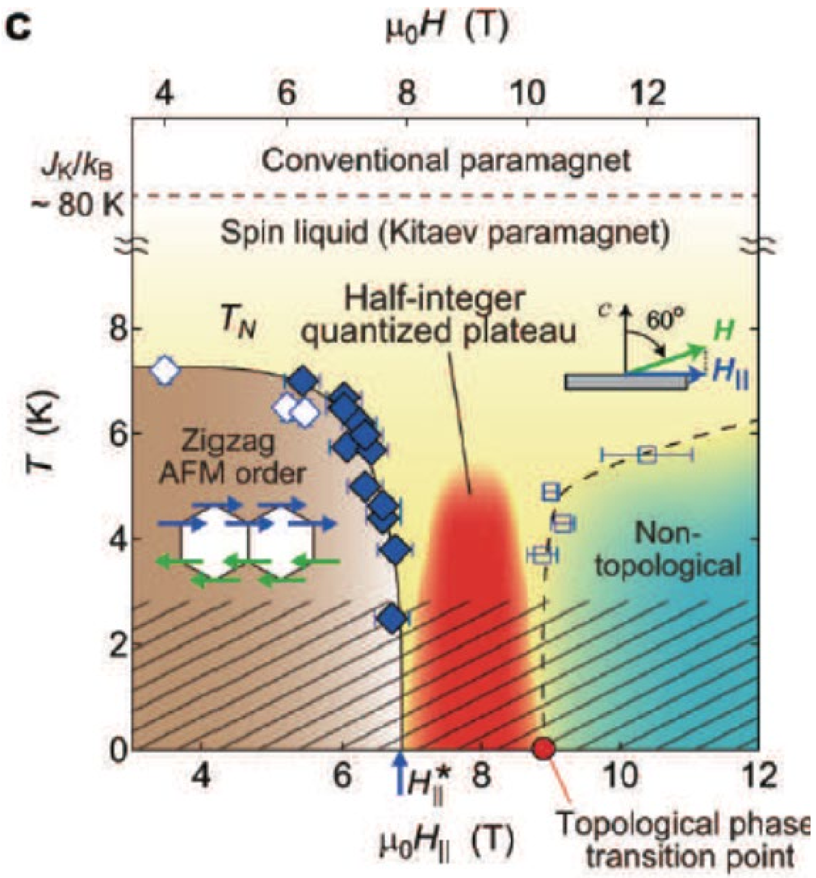


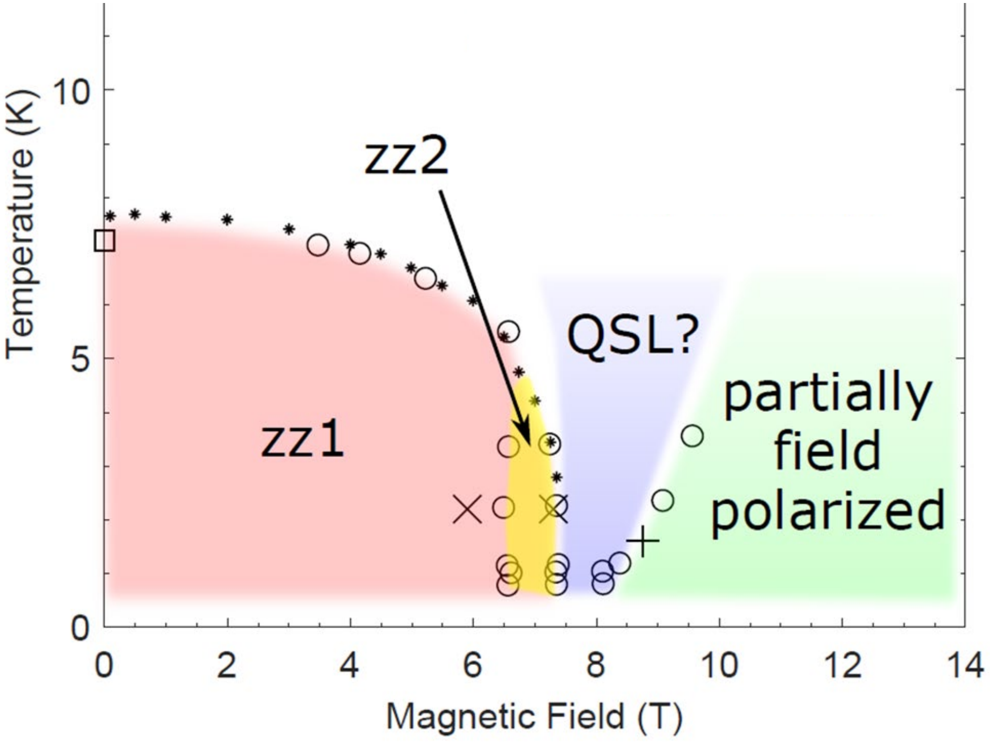
- Measurements at different temperatures allow one to construct a phase diagram

More complete phase diagram



Comparison with Kasahara *et al.* phase diagram





Some conclusions

- Inelastic neutron scattering in α-RuCl$_3$ is consistent with fractional excitations

- An external magnetic field applied in-plane leads to a magnetically disordered state, with a higher field transition to a state that seems to be partially polarized and supports magnons

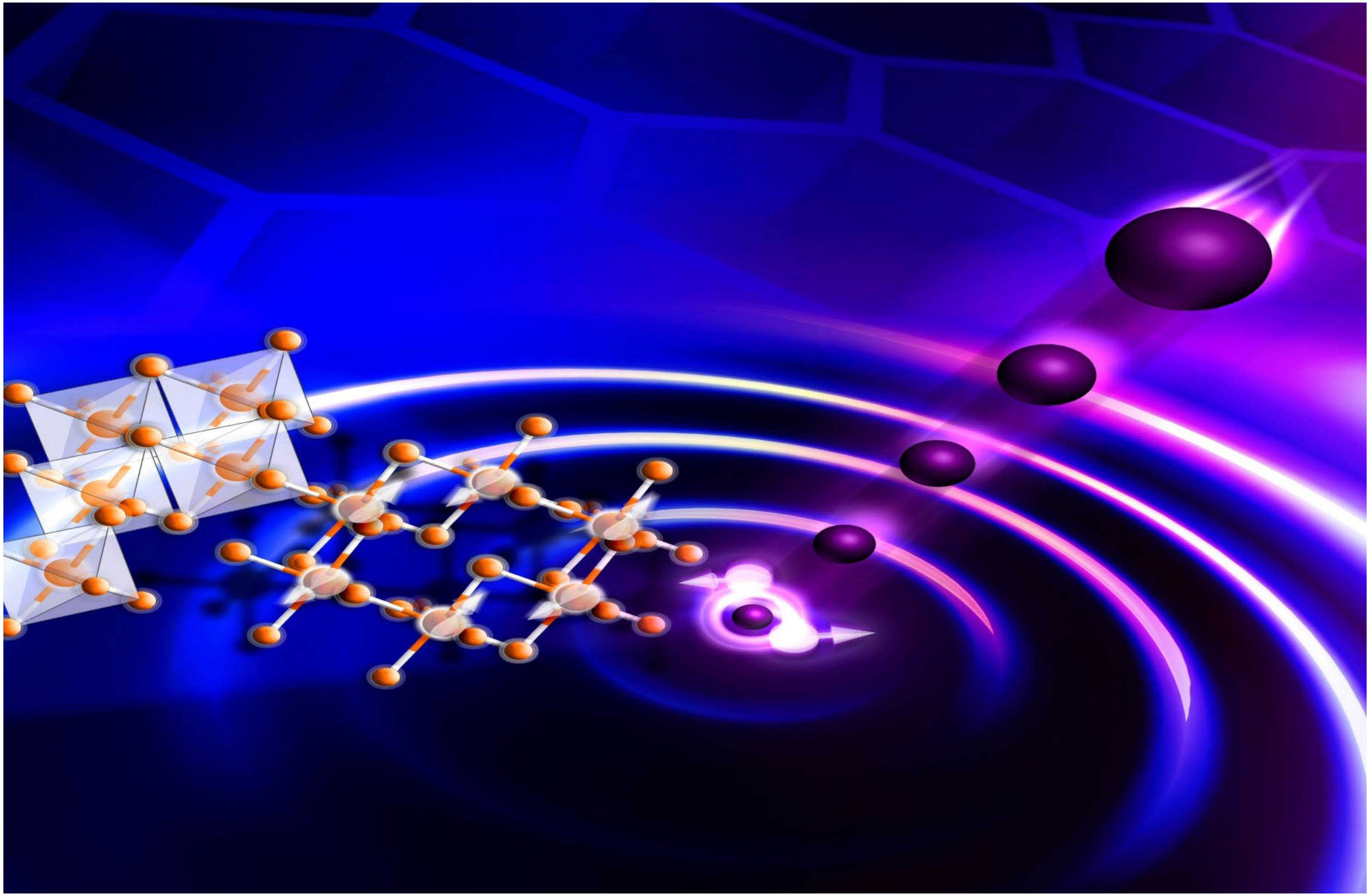
- The intermediate field disordered state is consistent with a QSL

Some ORNL references on α-$RuCl_3$

Neutron scattering experiments:

- A. Banerjee *et al.* Nature Materials **15**, 733(2016).
- H. Cao, A. Banerjee *et al.* PRB **93**, 134423 (2016).
- A. Banerjee *et al.* SCIENCE **356**, 1055 (2017).
- P. Lampen–Kelley *et al.* PRL **119**, 237203, (2017).
- A. Banerjee *et. al*., Npj Quantum Materials **3**, 8 (2018).
- C. Balz *et al*., PRB **100**, 060405(R) (2019).

Thank you for your attention



Questions?