Investigations on α-$RuCl_3$, a material proximate to the Kitaev quantum spin liquid

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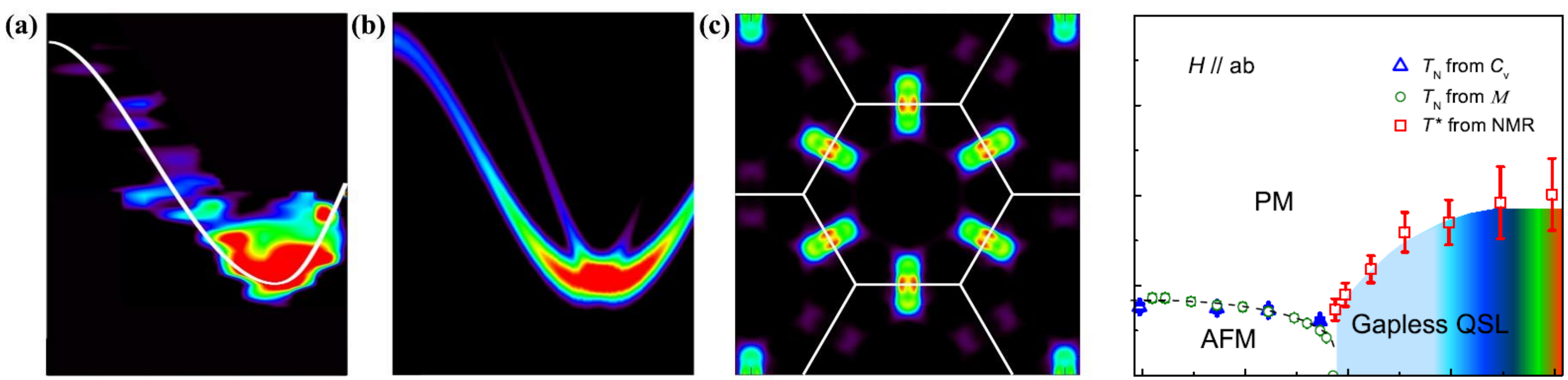
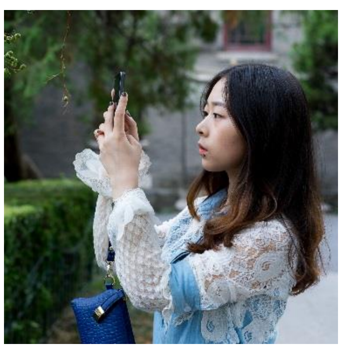


Acknowledgement

- **NJU**

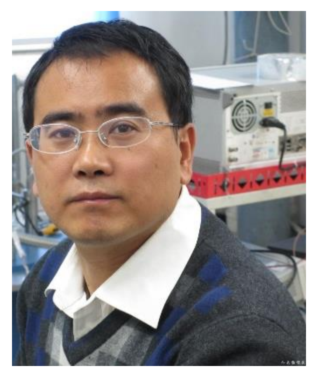
Neutron Scattering Group

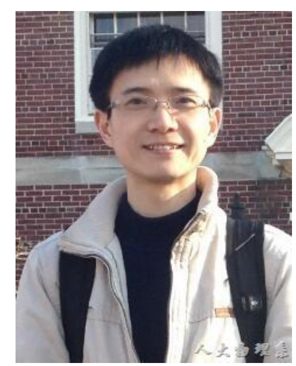


- **ANSTO, FRM II**

Theory: Jianxin Li's group

- **RUC, NMR+Theory**





Outline

- Introduction
- **Spin-wave excitations in α-$RuCl_3$**
- Fragile magnetic order

  Magnetic field, pressure, and doping
- Magnetism in two dimensional
- Summary

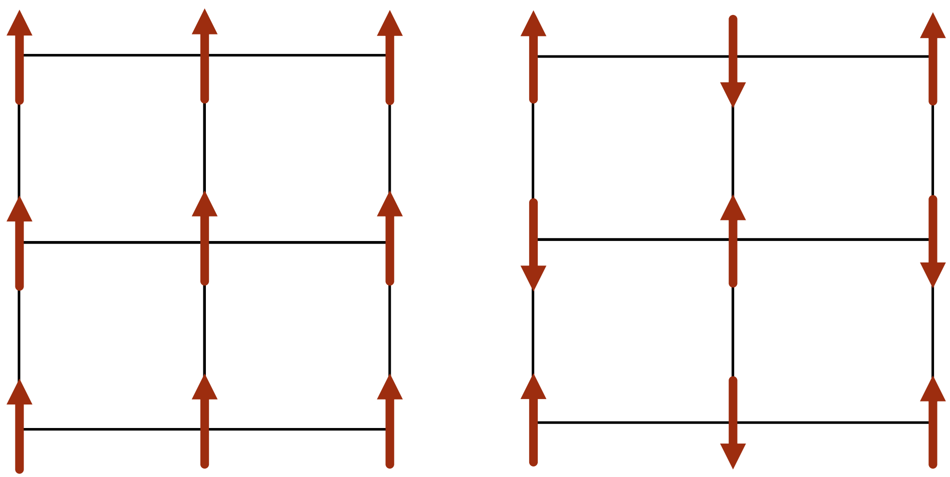
Quantum Spin Liquids

Heisenberg Model:

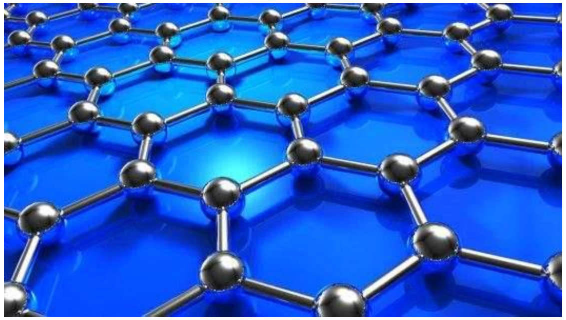
$$H = J \sum_{<i,j>} \mathbf{S}_i \cdot \mathbf{S}_j$$

Square lattice

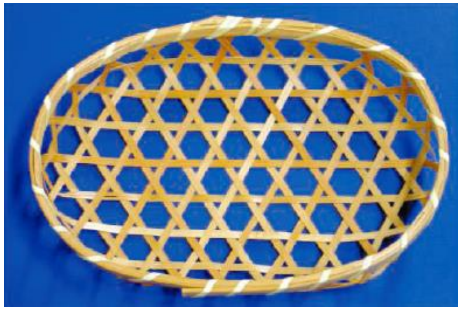
J<0 J>0

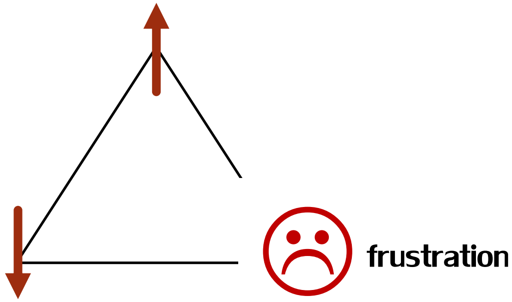


Honeycomb lattice



Kagome lattice

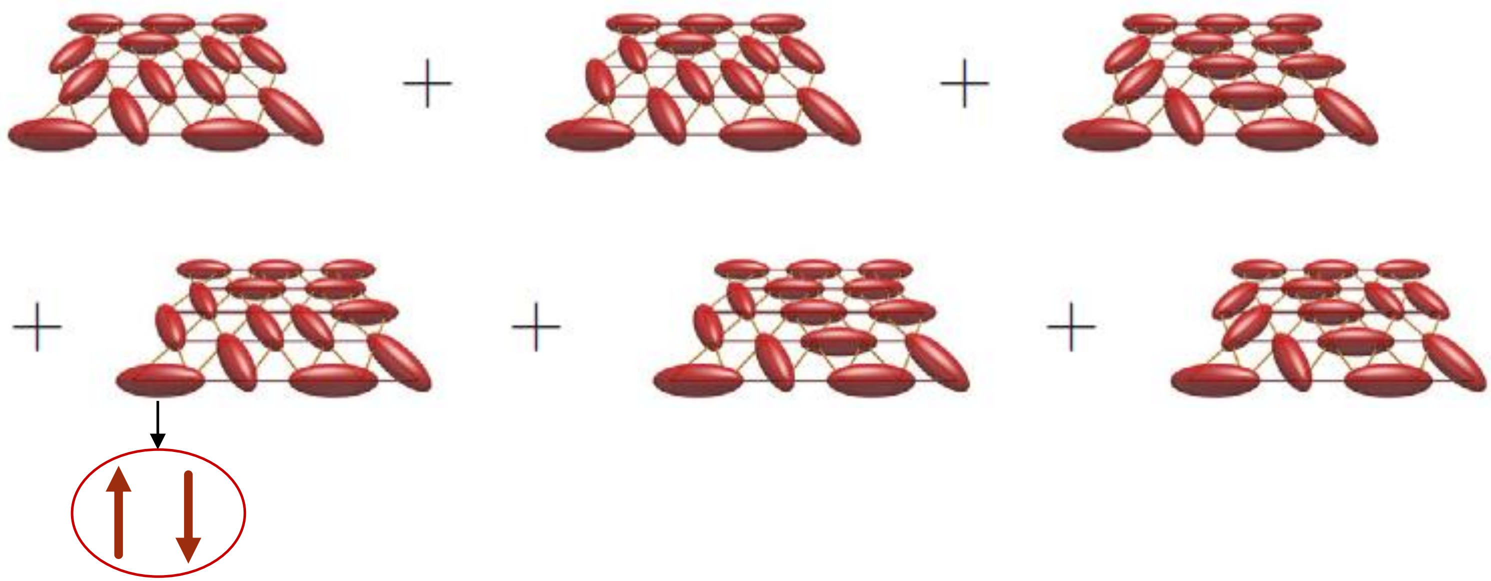




Triangular lattice

Quantum Spin Liquids

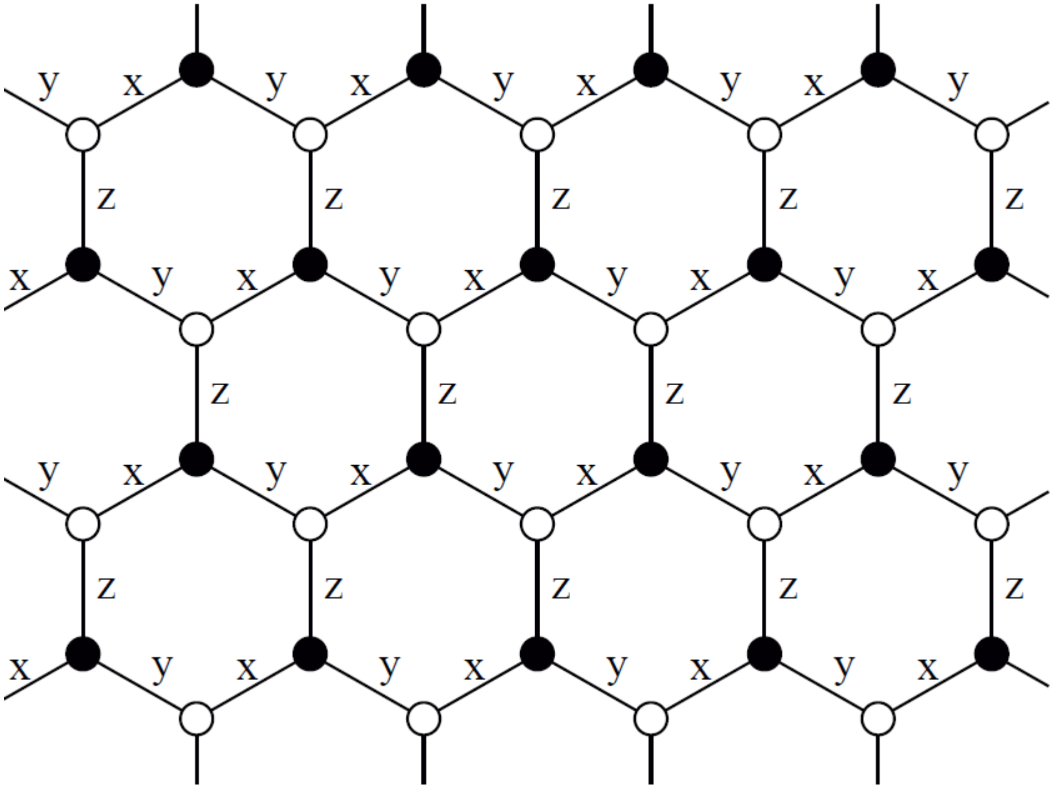
$$|\mathrm{RVB}\rangle =$$



P. W. Anderson, Mater. Res. Bull. 8, 153 (1973)
P. W. Anderson, Science 235, 1196 (1987)

- **A new quantum state of matter**
- **Quantum computing**
- **High-T_c superconductivity**

Kitaev model



$$H = -J_x \sum_{x\text{-links}} \sigma_j^x \sigma_k^x - J_y \sum_{y\text{-links}} \sigma_j^y \sigma_k^y - J_z \sum_{z\text{-links}} \sigma_j^z \sigma_k^z$$

A. Kitaev, Annals of Physics 321, 2 (2006)

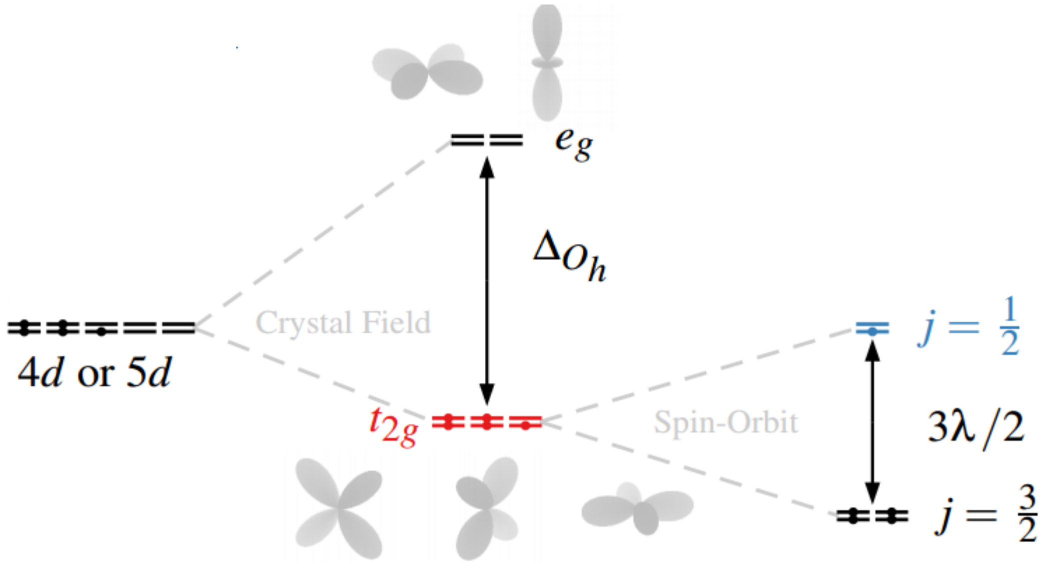
Kitaev model

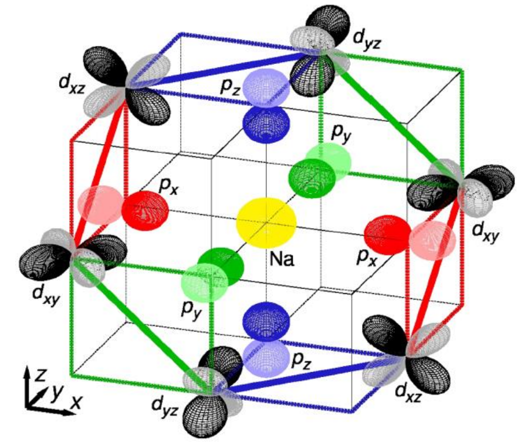
Alexei Kitaev
California Institute of Technology

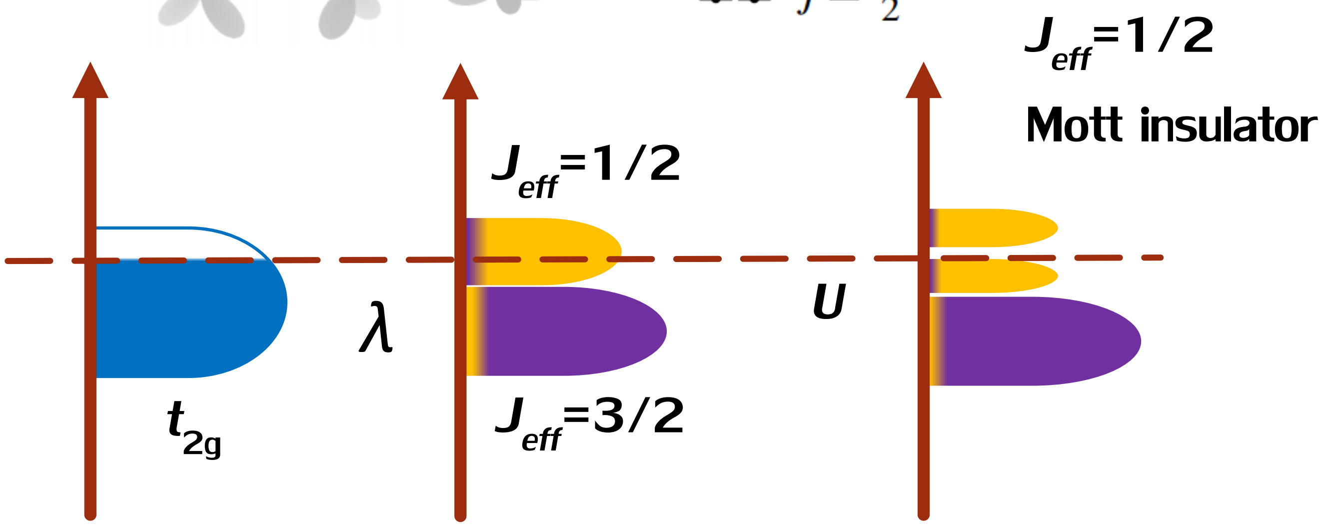
Xiao-Gang Wen
Massachusetts Institute of Technology

"For theories of topological order and its consequences in a broad range of physical systems, including the fractional quantum Hall effect, frustrated magnets, and topological states protected by symmetry."

Kitaev interaction in real materials

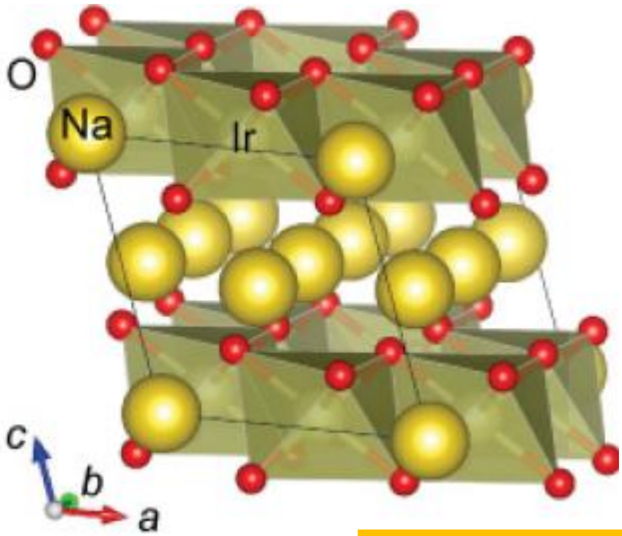


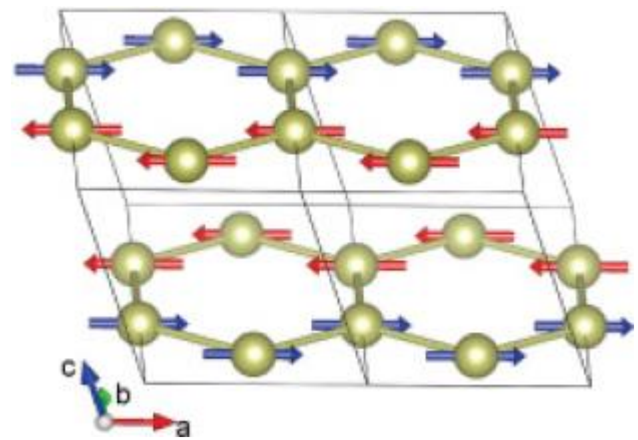




Rau *et al.*, Annu. Rev. Conden. Matter Phys. 7, 195 (2016).

Kitaev interaction in real materials





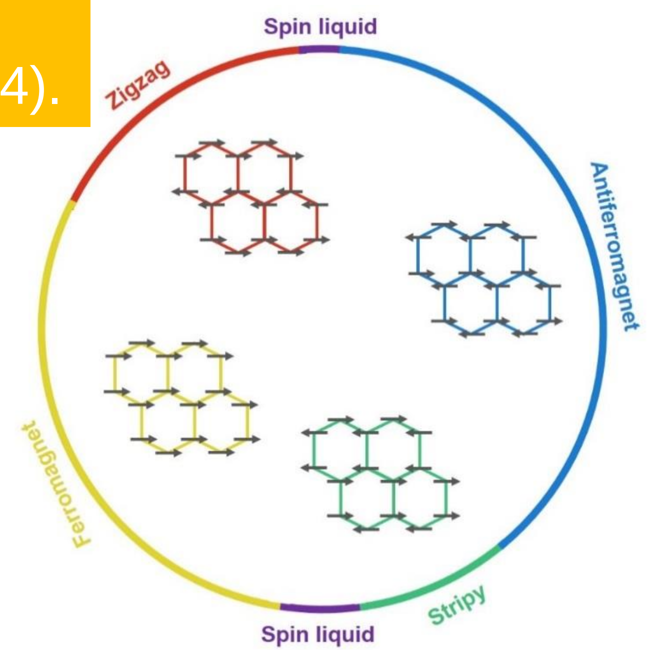
Liu *et al*

Ye *et al.*

α-RuCl3

Phys. Rev. B 90, 041112(R) (2014).

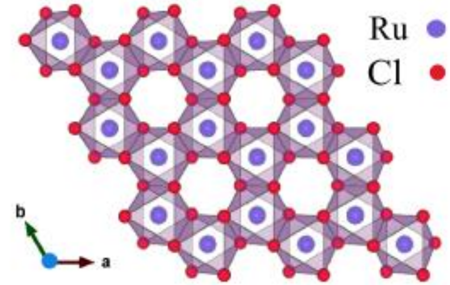
$$H = K \sum_{\gamma = x,y,z} S_i^{\gamma} S_j^{\gamma} + J \sum_{<ij>} \vec{S}_i \bullet \vec{S}_j$$



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

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

$\alpha-RuCl_3$



CHEMISTRY

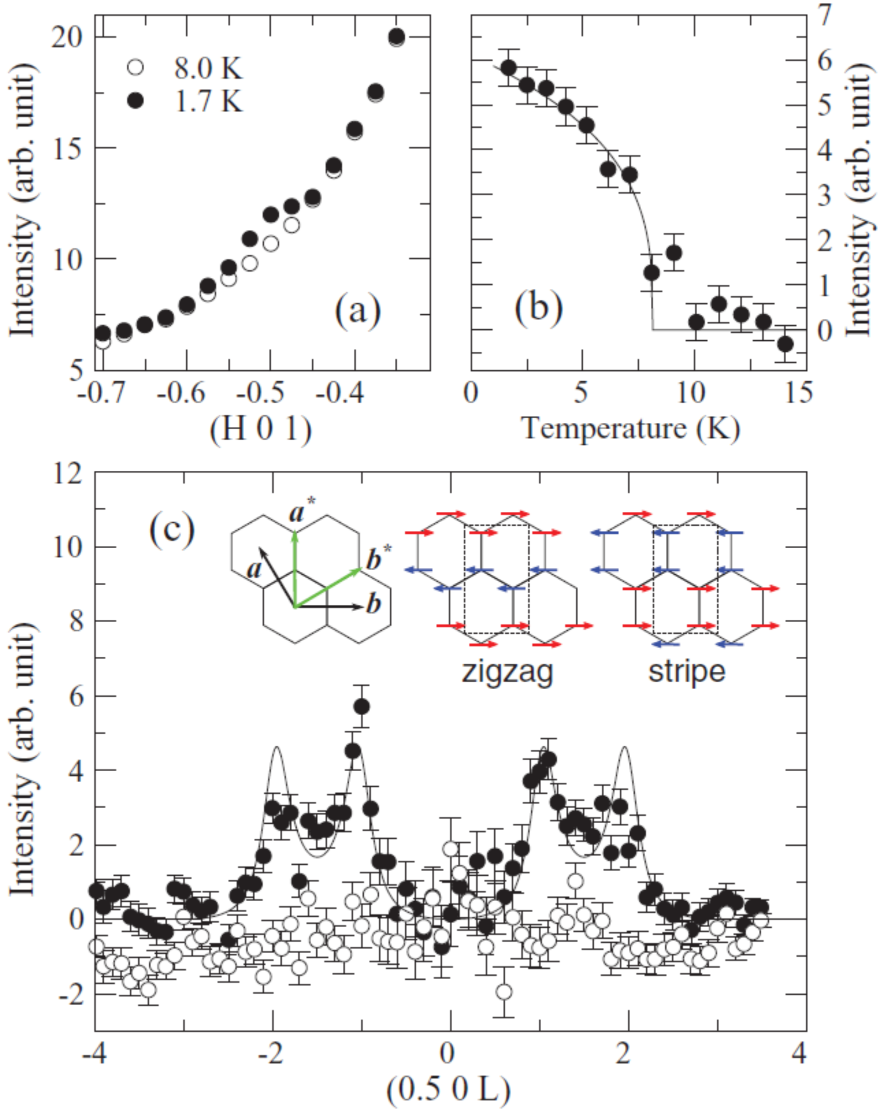
Anhydrous Ruthenium Chlorides

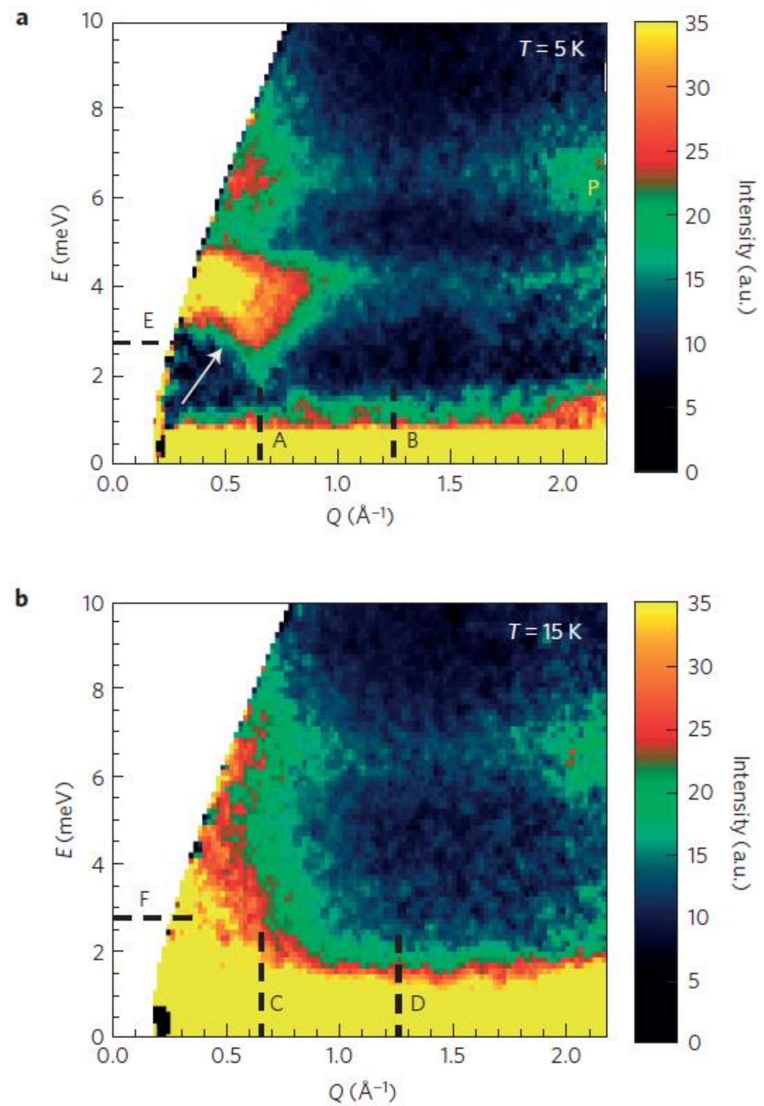
THE contrasting magnetic behaviour (Table 1 and Fig. 1) of the α- and β-forms of $RuCl_3$ is of interest in relation to the different types of stereochemical arrangements[1] shown by the transition metal halides: it has also allowed the presence of α-$RuCl_3$, produced in the usual method of preparation of β-$RuCl_3$, to be detected. A material, hitherto considered[2,3] as yet another form of $RuCl_3$, is now shown to be $Ru_2^{IV}OCl_6$ (Table 1), a member of a class of oxide chlorides of binuclear ruthenium in which the average oxidation state of ruthenium changes from +4 through +3·5 to +3.

α-$RuCl_3$. This is produced[3,4] by a slow reaction between the metal and chlorine in siliceous vessels at temperatures above 600° C. However, contact of the metal with the

Fletcher *et al.*, Nature 199, 1089 (1963)

α−$RuCl_3$

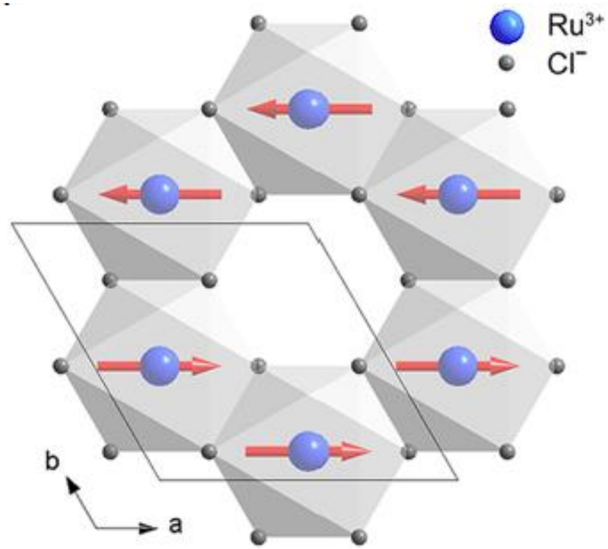




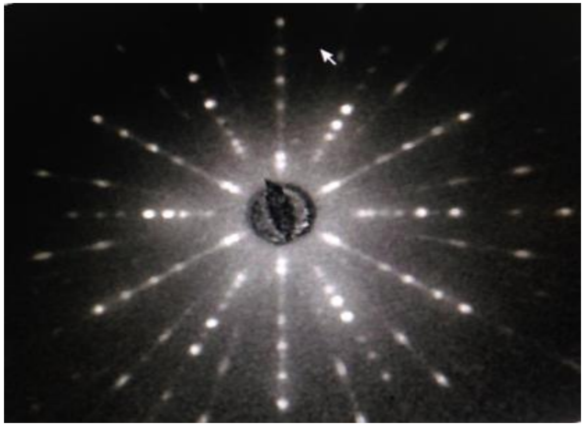
Phys. Rev. B 90, 041112(R) (2014).
Phys. Rev. B 91, 144420 (2015).

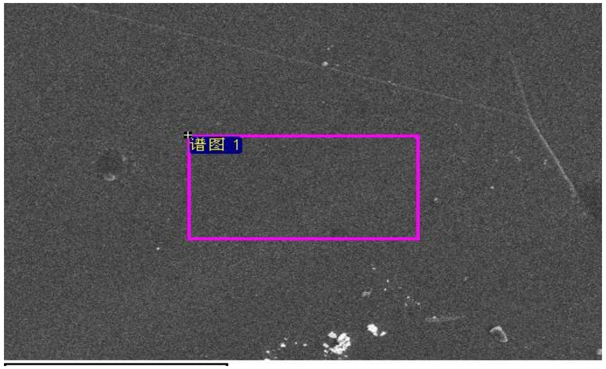
A. Banerjee et al., Nature Mat. 15, 733 (2016).

α-$RuCl_3$

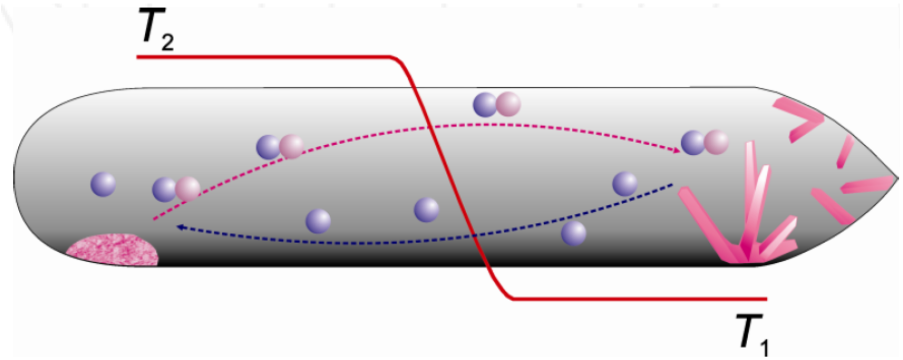


Ru^{3+}
Cl^-
b
a

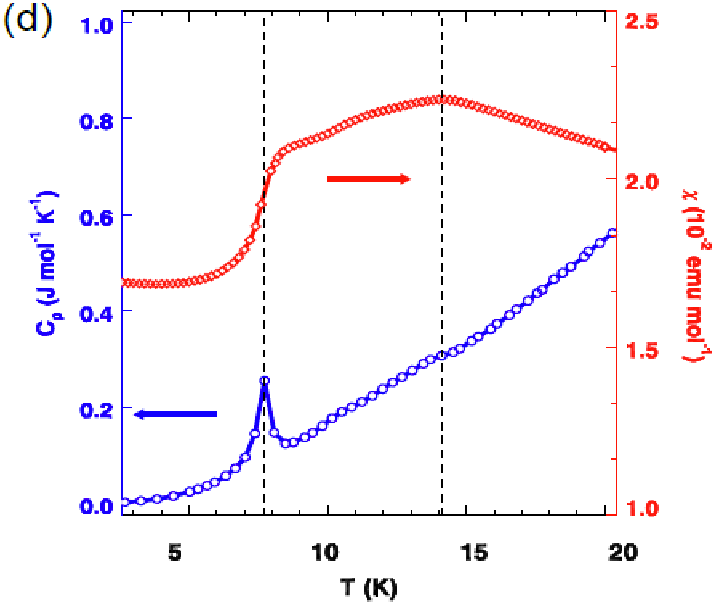




谱图 1



T_2
T_1

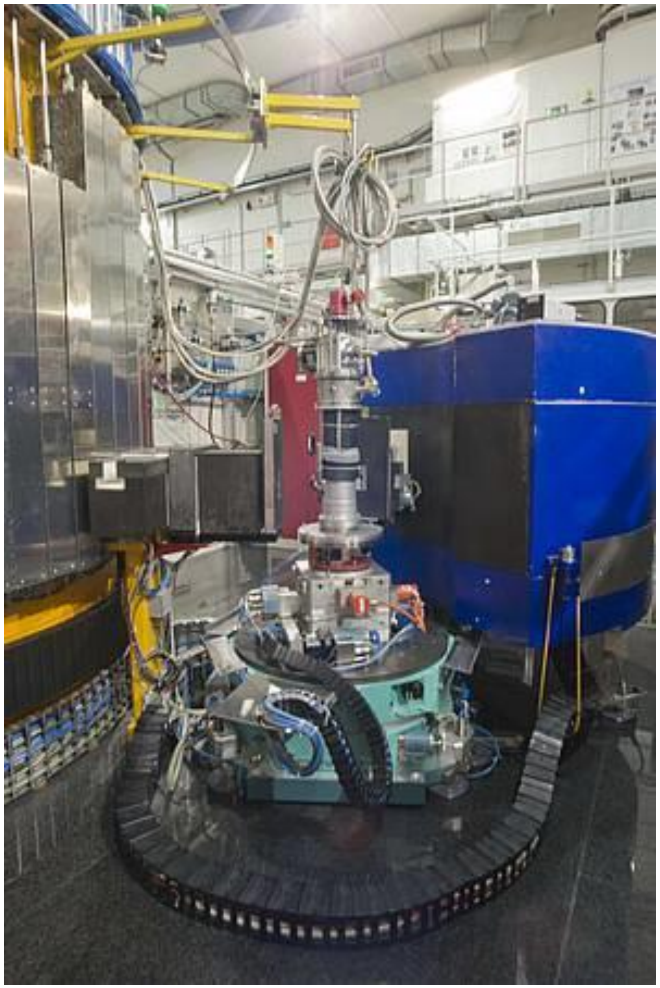


(d)
C_p (J mol^{-1} K^{-1})
χ (10^{-2} emu mol^{-1})
T (K)
0.0
0.2
0.4
0.6
0.8
1.0
1.0
1.5
2.0
2.5
5
10
15
20

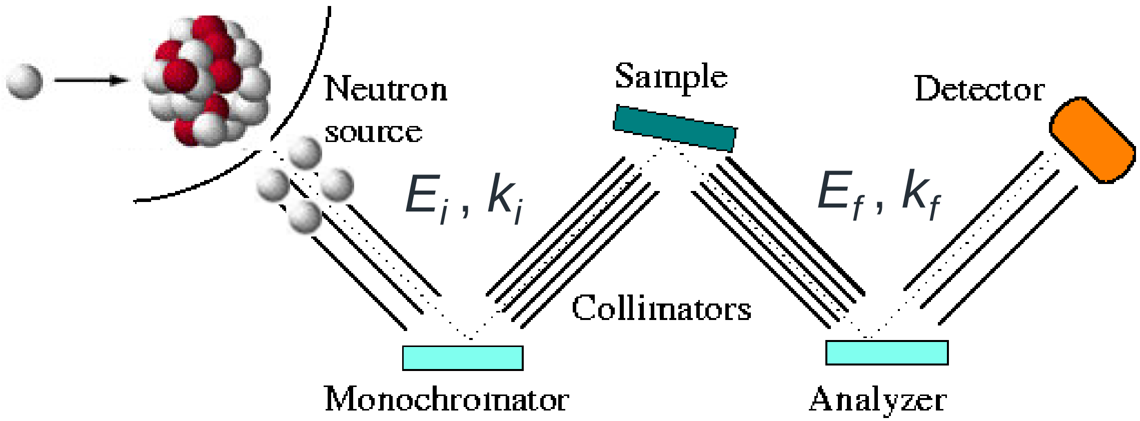
Neutron Scattering experiment

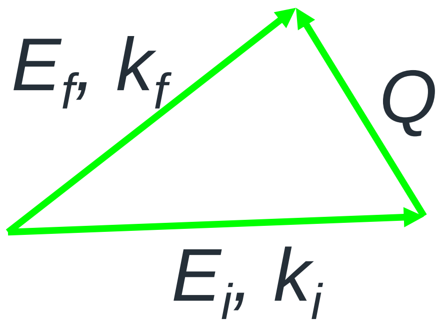
ANSTO: SIKA

Neutron Scattering experiment



PUMA, FRM II, TUM





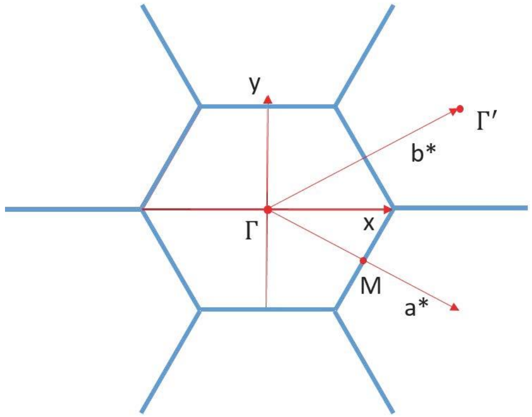
$\hbar\omega = E_i - E_f$

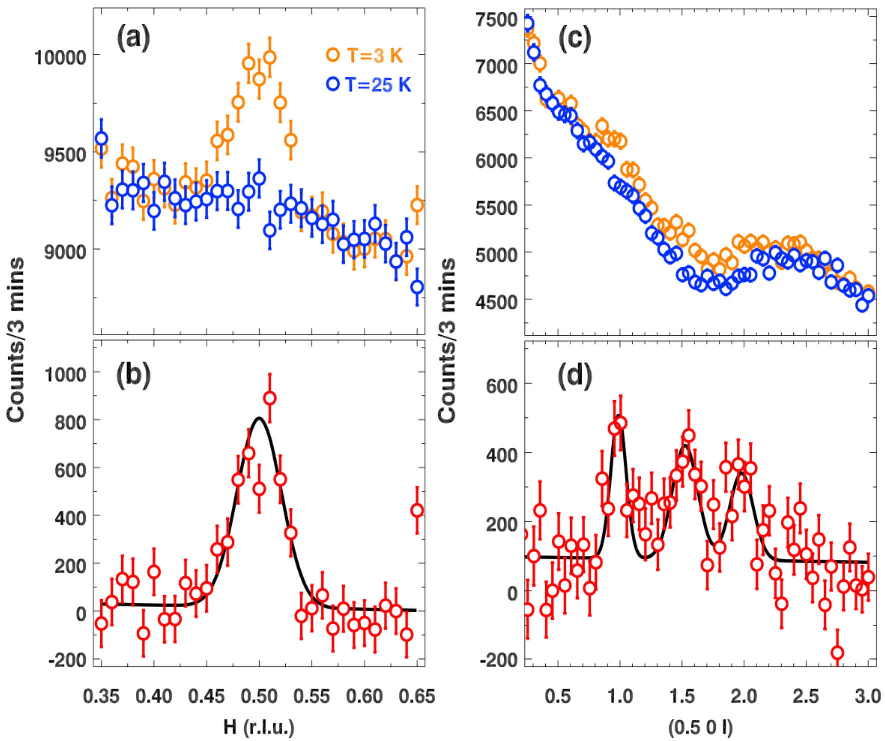
$Q = k_f - k_i$

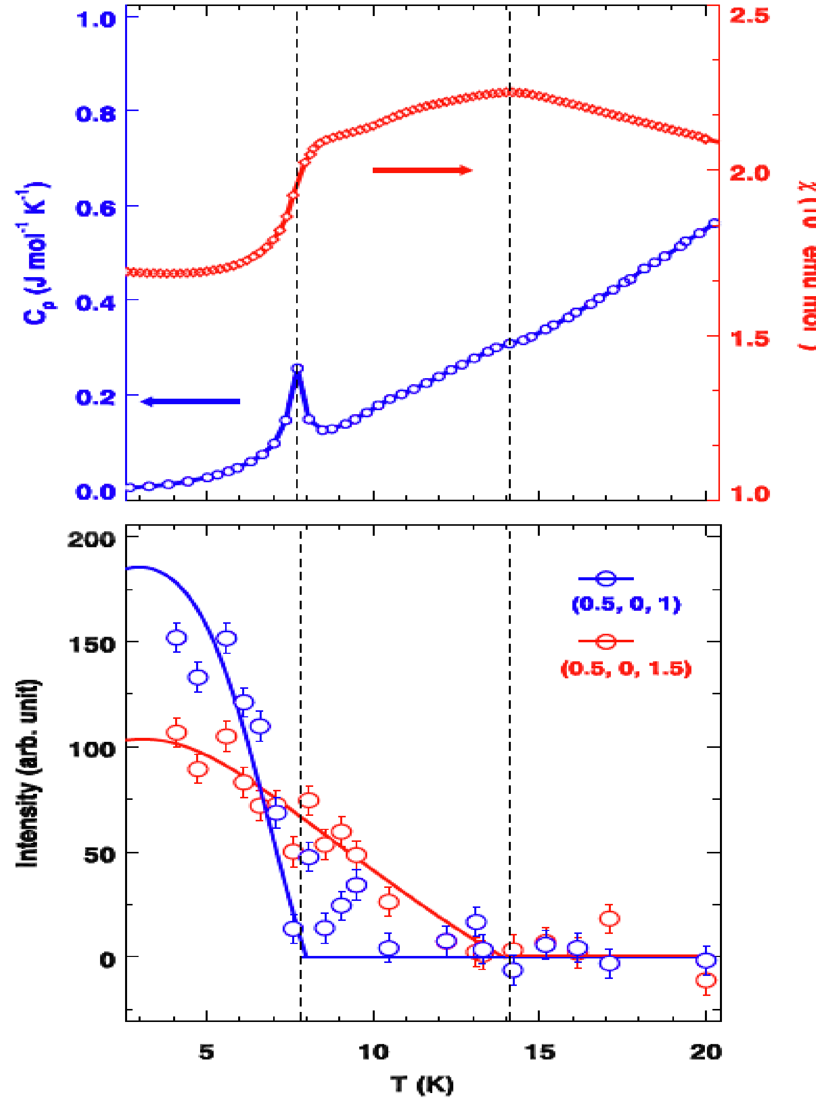
Elastic: $E_f = E_i$, structure

Inelastic: $E_f \neq E_i$ dynamics

Elastic Neutron Scattering Results

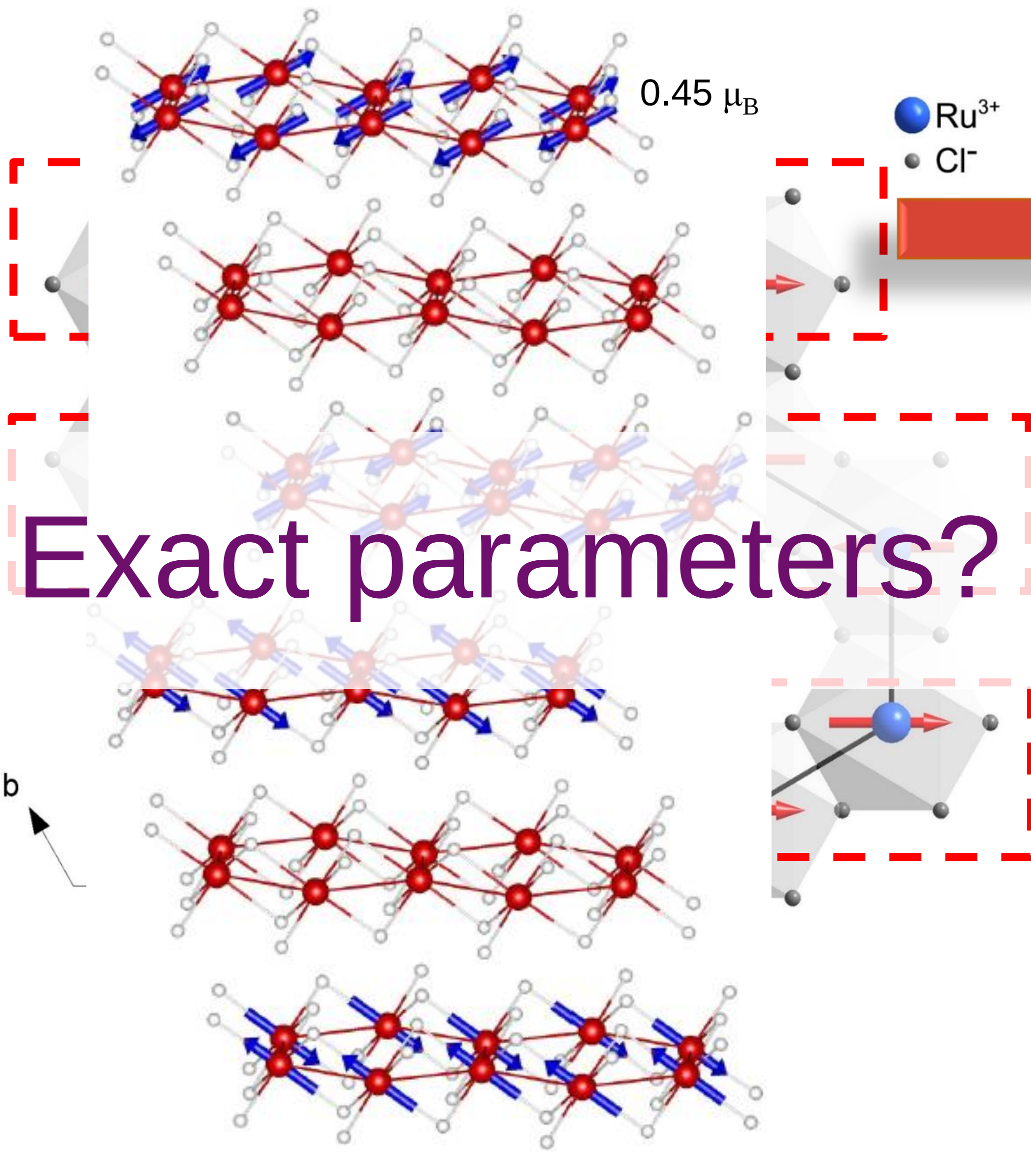


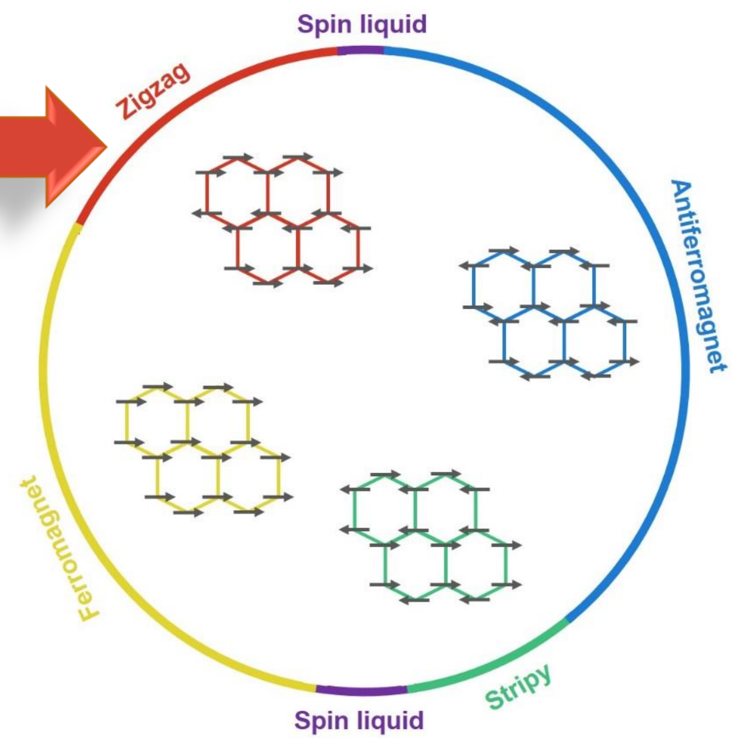




Phys. Rev. Lett. 118, 107203 (2017)

Zigzag magnetic order in $\alpha-RuCl_3$

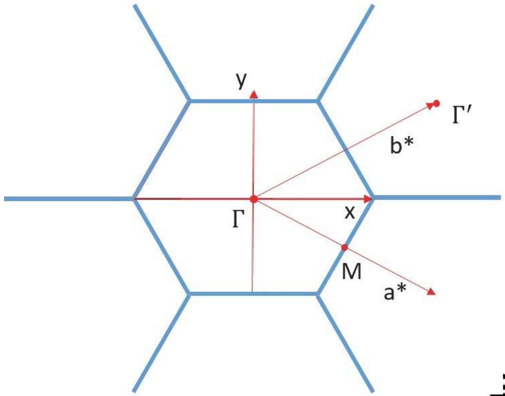




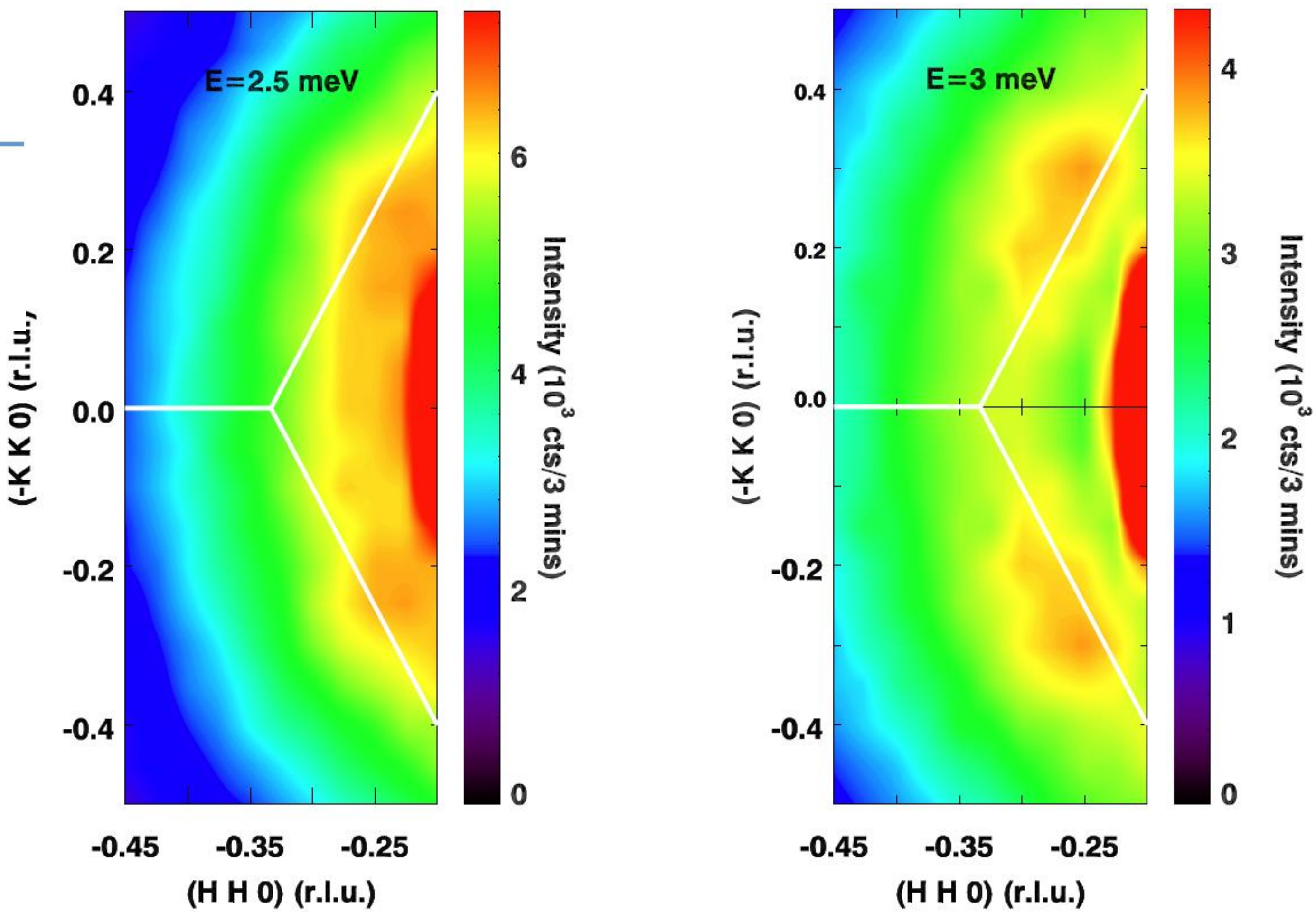
Exact parameters?

Cao *et al.*, Phys. Rev. B 93, 134423 (2016)

Inelastic Neutron Scattering Results

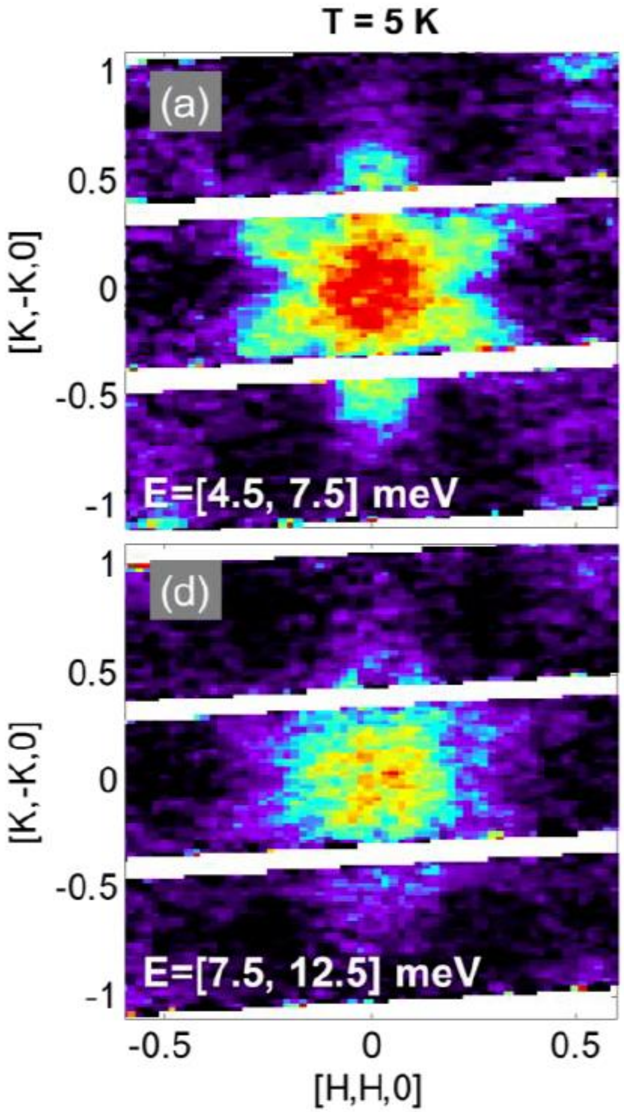


T= 3 K

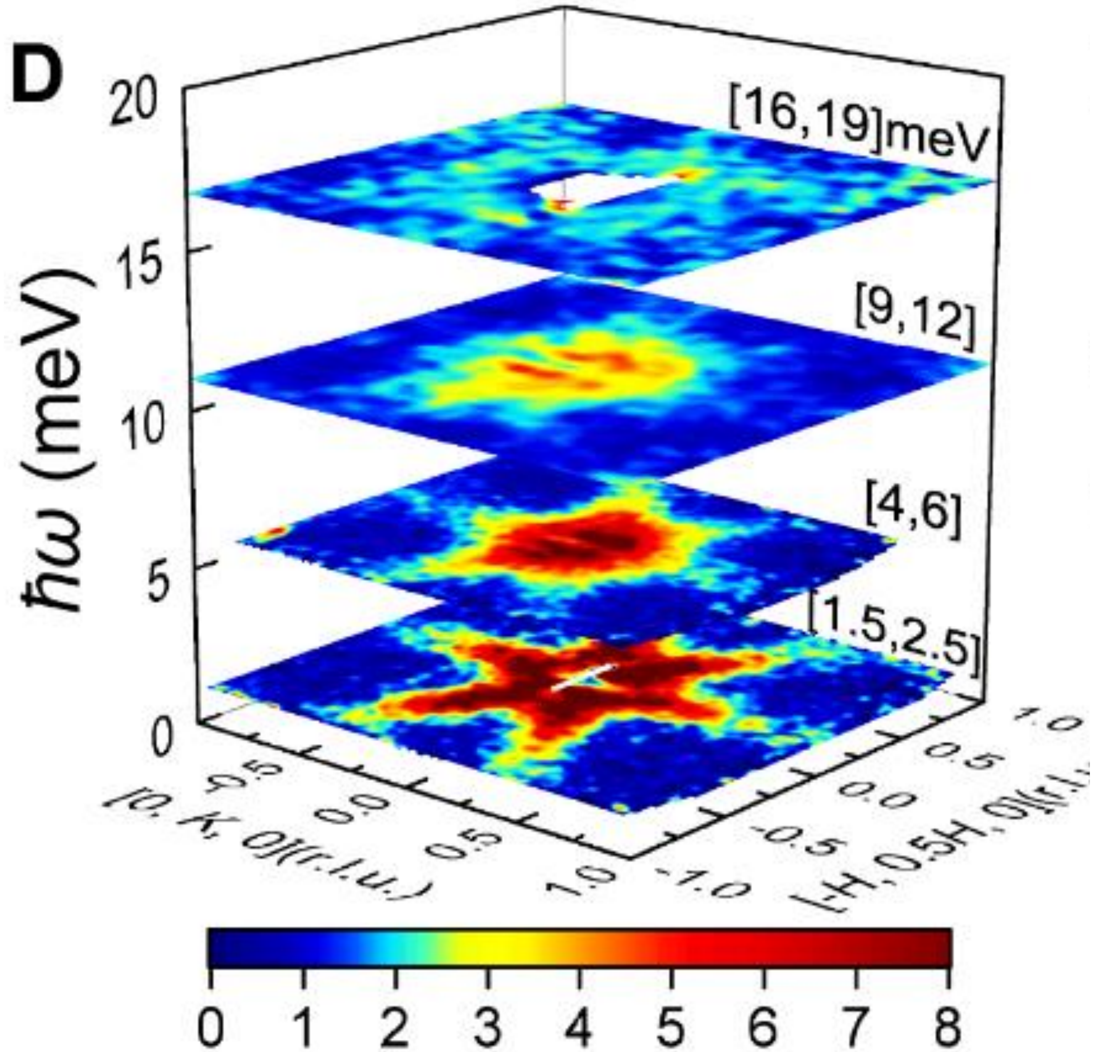


Phys. Rev. Lett. 118, 107203 (2017)

Majorana mode?

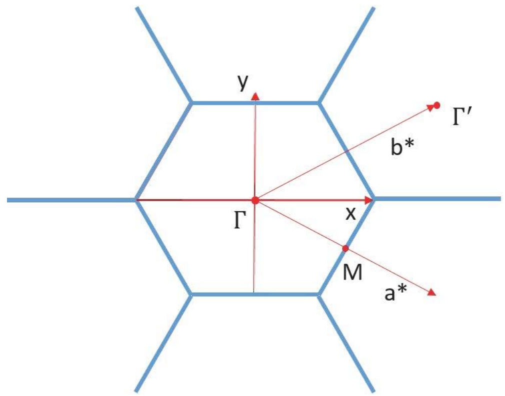


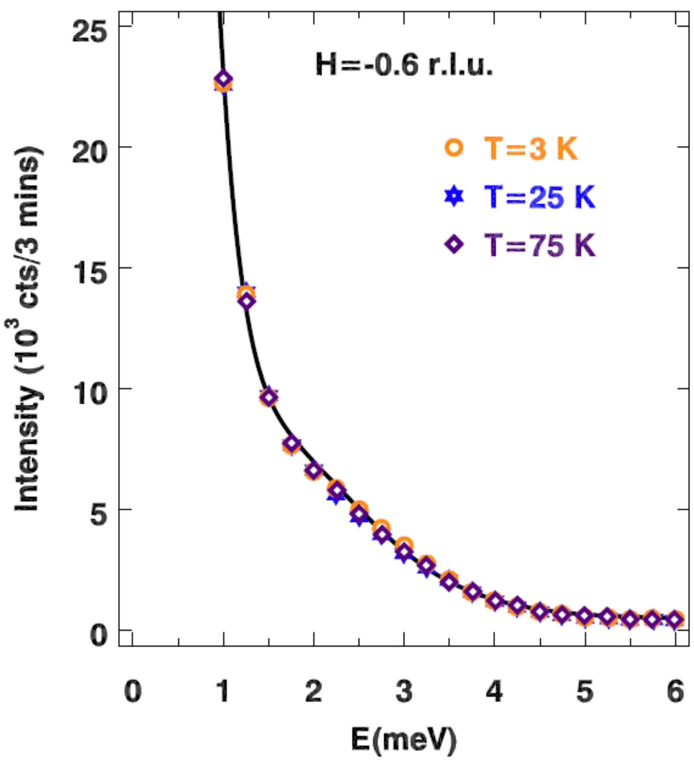
Banerjee *et al.*, Science 356, 1055 (2017)

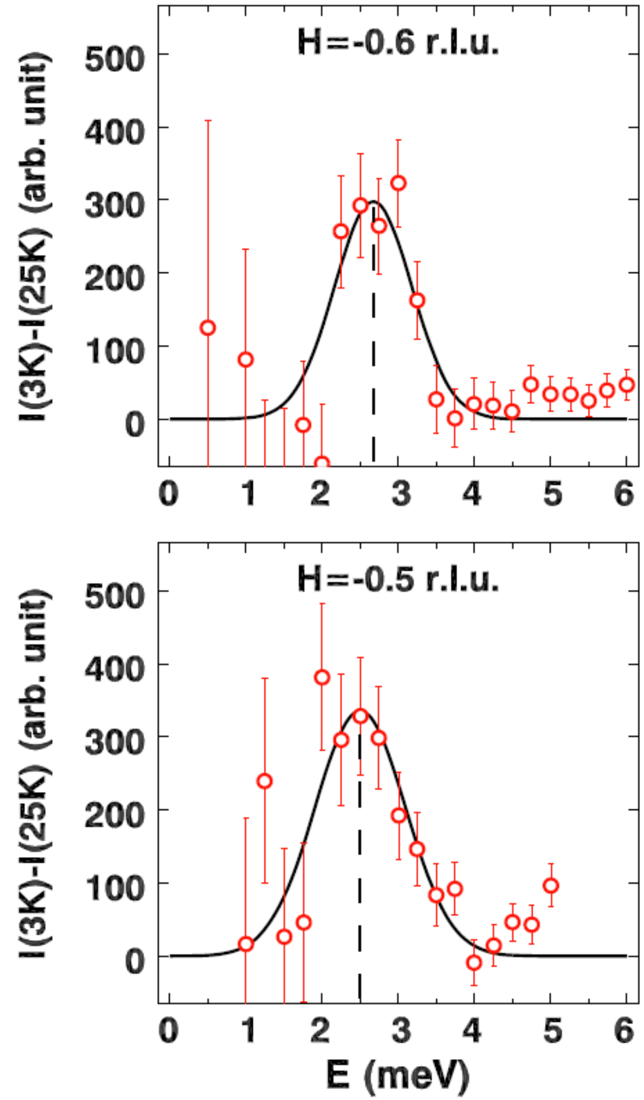


Do *et al.*, arXiv:1703.01081

Inelastic Neutron Scattering Results

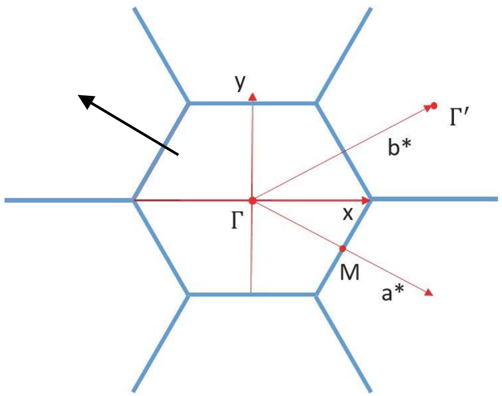


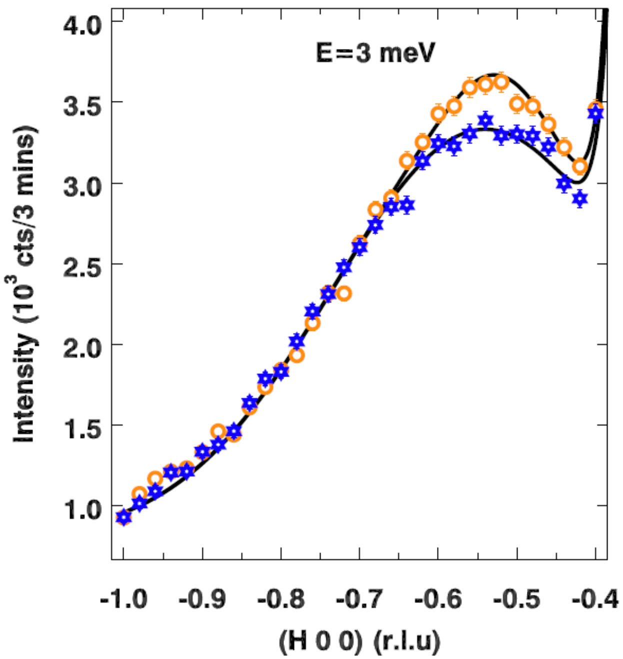


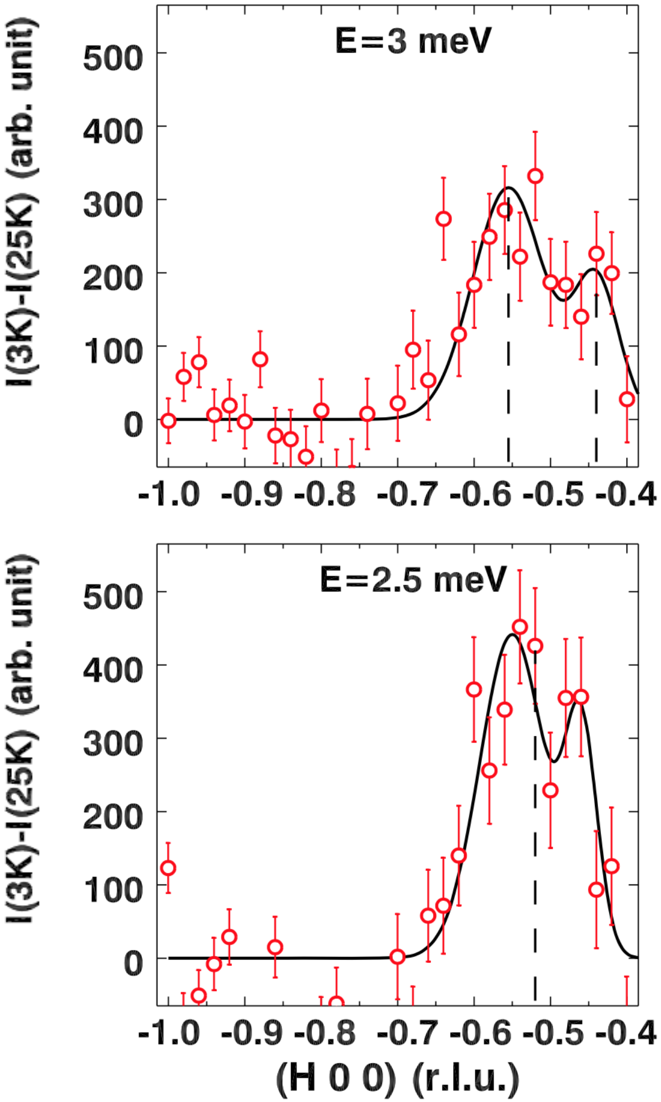


Phys. Rev. Lett. 118, 107203 (2017)

Inelastic Neutron Scattering Results

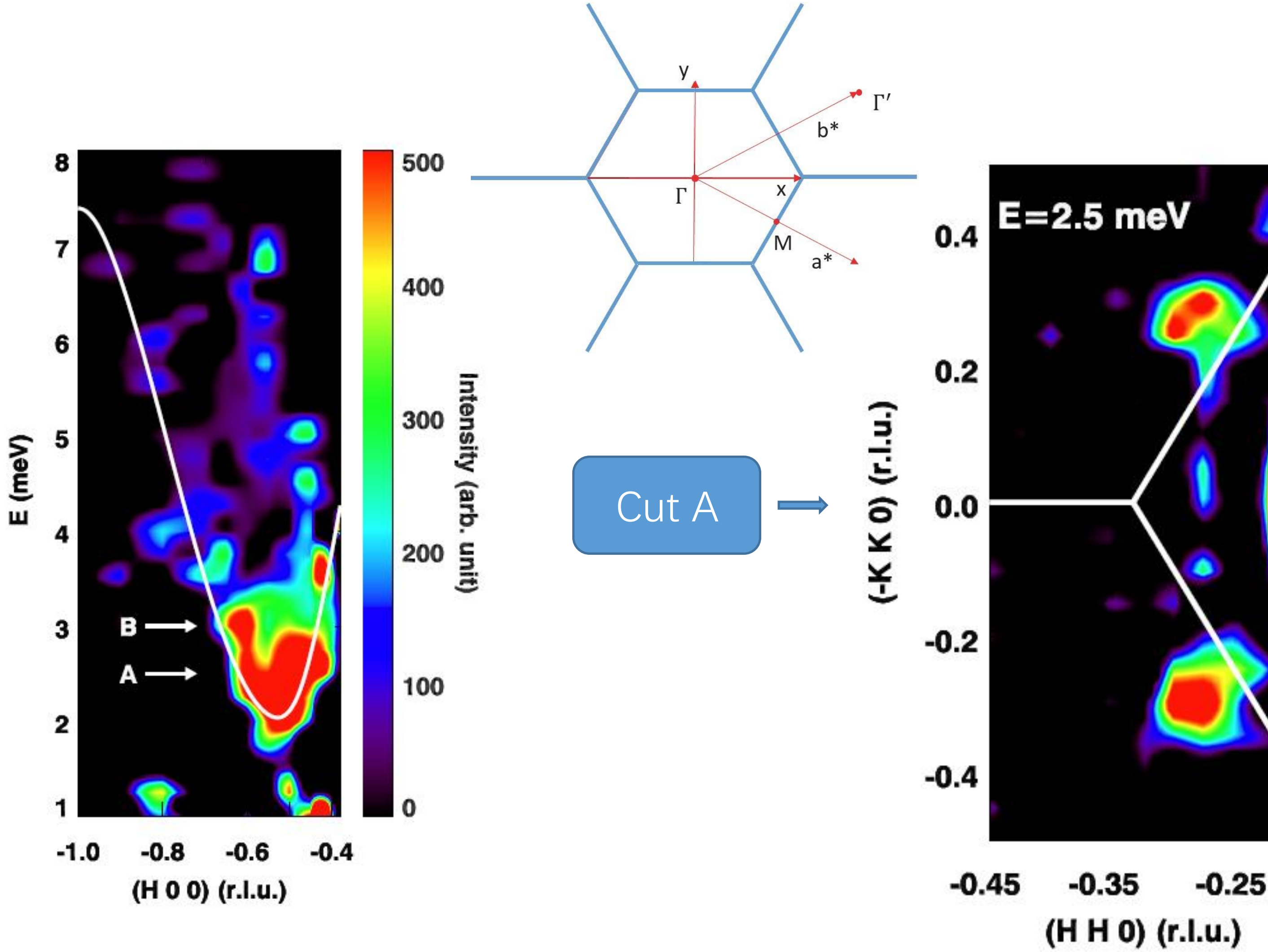






Phys. Rev. Lett. 118, 107203 (2017)

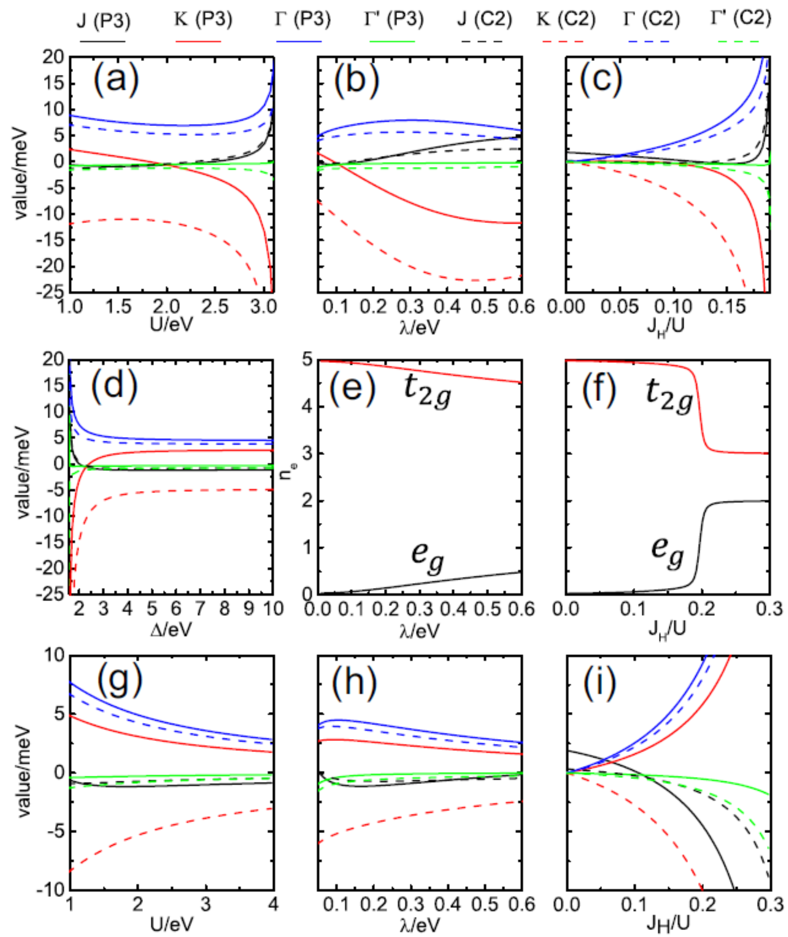
Inelastic Neutron Scattering Results



Minimal model

$$H_{\text{eff}} = \sum_{\langle ij\rangle\in\gamma(\alpha\beta)} [J^\gamma \boldsymbol{S}_i\cdot\boldsymbol{S}_j + K^\gamma S_i^\gamma S_j^\gamma + \Gamma^\gamma(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \Gamma'^\gamma(S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma + S_i^\gamma S_j^\beta + S_i^\gamma S_j^\alpha)] + \sum_{\langle\langle ij\rangle\rangle\in\gamma}(J_2^\gamma \boldsymbol{S}_i\cdot\boldsymbol{S}_j + K_2^\gamma S_i^\gamma S_j^\gamma) + \sum_{\langle\langle\langle ij\rangle\rangle\rangle\in\gamma} K_3^\gamma S_i^\gamma S_j^\gamma. \quad (10)$$

Structure	J	K	Γ	J_3
$C2$	-0.3	-10.9	6.1	0.03
$P3$	0.1	-5.5	7.6	0.1



K- Γ model

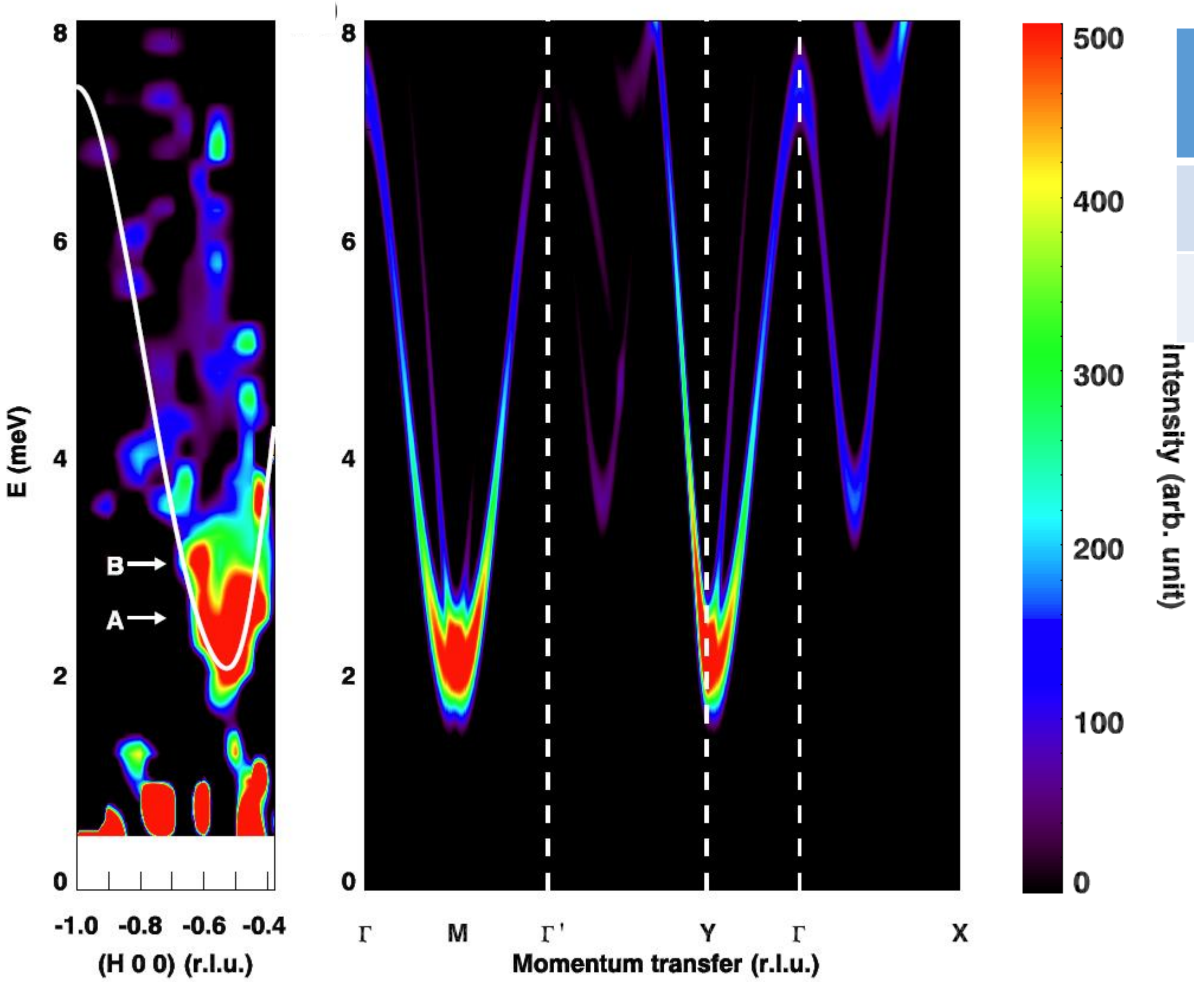
$$H = \sum_{\langle ij\rangle\in\alpha\beta(\gamma)} [K S_i^\gamma S_j^\gamma + \Gamma(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha)]$$

See Wang *et al.*, arXiv:1612.09515 for more details.

Spin−wave excitations in $RuCl_3$

K- Γ model

$$H = \sum_{\langle ij \rangle \in \alpha\beta(\gamma)} [K S_i^\gamma S_j^\gamma + \Gamma(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha)]$$



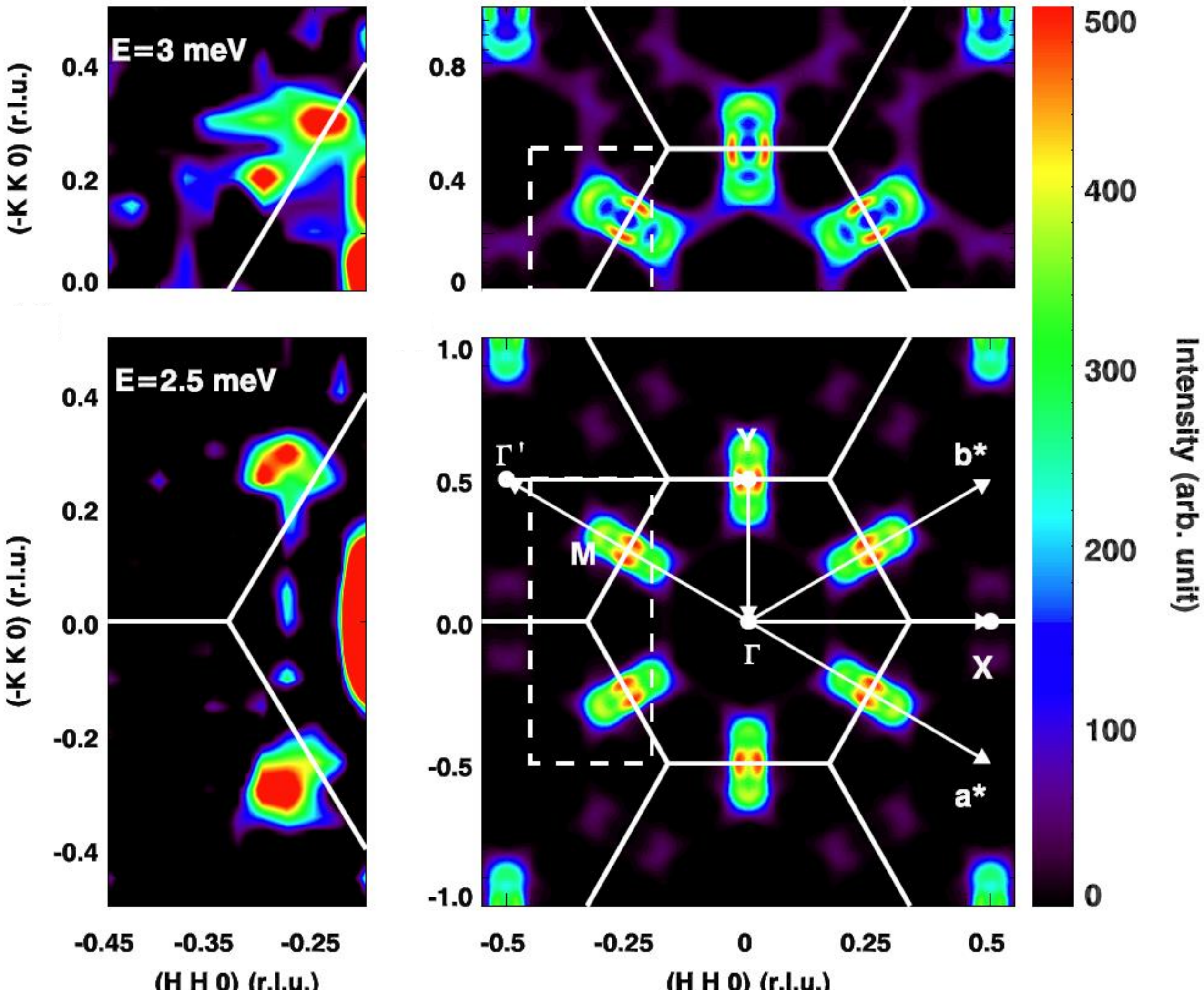
	K (meV)	*Γ* (meV)
Experiment	-6.8	9.5
Theory	-5.5	7.6

Phys. Rev. Lett. 118, 107203 (2017)

Spin-wave excitations in $RuCl_3$

K- Γ model

$$H = \sum_{\langle ij \rangle \in \alpha\beta(\gamma)} [K S_i^\gamma S_j^\gamma + \Gamma(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha)]$$



K= -6.8 meV

Γ= 9.5 meV

Small *J* may be needed

Phys. Rev. Lett. 118, 107203 (2017)

Which is the best?

➢ *J*–*K* model: may be incompatible with the order

Chaloupka *et al.*, Phys. Rev. Lett. 105, 027204 (2010)

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

➢ Extended *J*–*K* model: Γ necessary for the gap

Winter *et al.*, Phys. Rev. B 93, 214431 (2016)

Banerjee *et al.*, Nature Mater. 15, 733 (2016)

Hou et al., arXiv:1612.00761

➢ *K*–Γ–*J* model

Ran *et al.*, Phys. Rev. Lett. 118, 107203 (2017)

Wang *et al.*, arXiv:1612.09515

Winter *et al.*, arXiv:1702.08466

Banerjee *et al.*, Science 356, 1055 (2017)

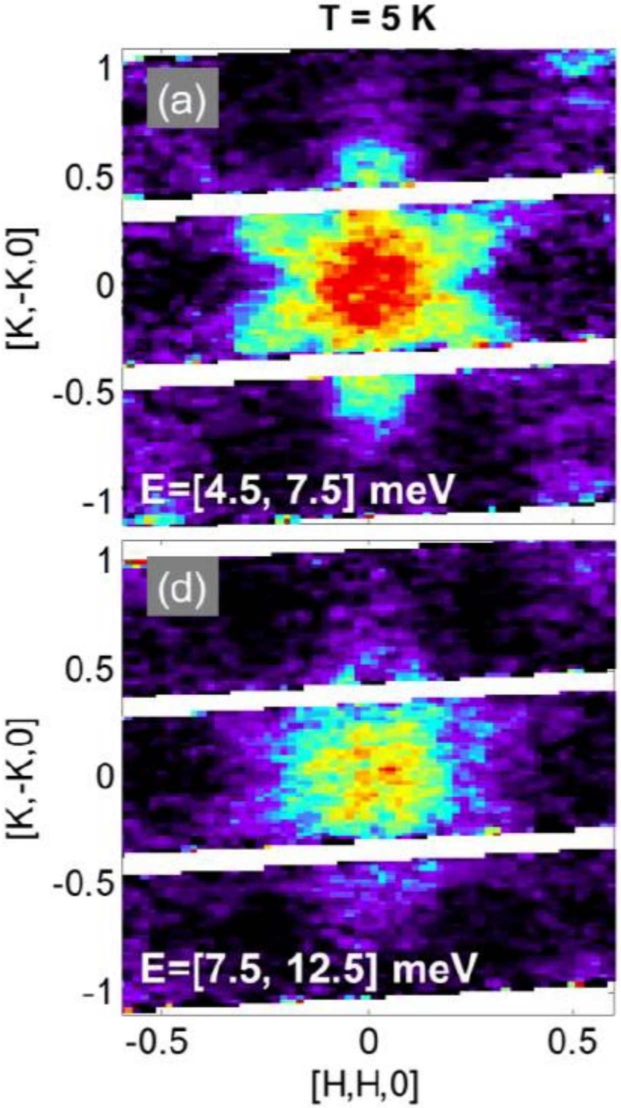
Catuneanu *et al.*, arXiv:1701.07837

Janssen *et al.*, arXiv:1706.0538

Gohlke *et al.*, arXiv:1706.09908

➢ Kitaev model: no magnetic order

Do *et al.*, arXiv:1703.01081



Banerjee *et al.*, Science 356, 1055 (2017)

Model

Breakdown of Magnons in a Strongly Spin-Orbital Coupled Magnet

Stephen M. Winter,[1] Kira Riedl,[1] Andreas Honecker,[2] and Roser Valentí[1]
[1] *Institut für Theoretische Physik, Goethe-Universität Frankfurt, Max-von-Laue-Strasse 1, 60438 Frankfurt am Main, Germany*
[2] *Laboratoire de Physique Théorique et Modélisation, CNRS UMR 8089, Université de Cergy-Pontoise, 95302 Cergy-Pontoise Cedex, France*
(Dated: March 1, 2017)

The description of quantized collective excitations stands as a landmark in the quantum theory of condensed matter. A prominent example occurs in conventional magnets, which support bosonic magnons - quantized harmonic fluctuations of the ordered spins. In striking contrast is the recent discovery that strongly spin-orbital coupled magnets, such as α-$RuCl_3$, may display a broad excitation continuum inconsistent with conventional magnons. Unraveling the nature of this continuum, however, remains challenging due to incomplete knowledge of the underlying interactions. While the most discussed explanation refers to a coherent continuum of fractional excitations analogous to the celebrated Kitaev spin-liquid, we present here a more general scenario. We propose the observed continuum represents incoherent excitations originating from strong magnetic anharmonicity that naturally occurs in such materials. This scenario fully explains the observed inelastic magnetic response of α-$RuCl_3$ and reveals the presence of nontrivial excitations in such materials extending well beyond the Kitaev state.

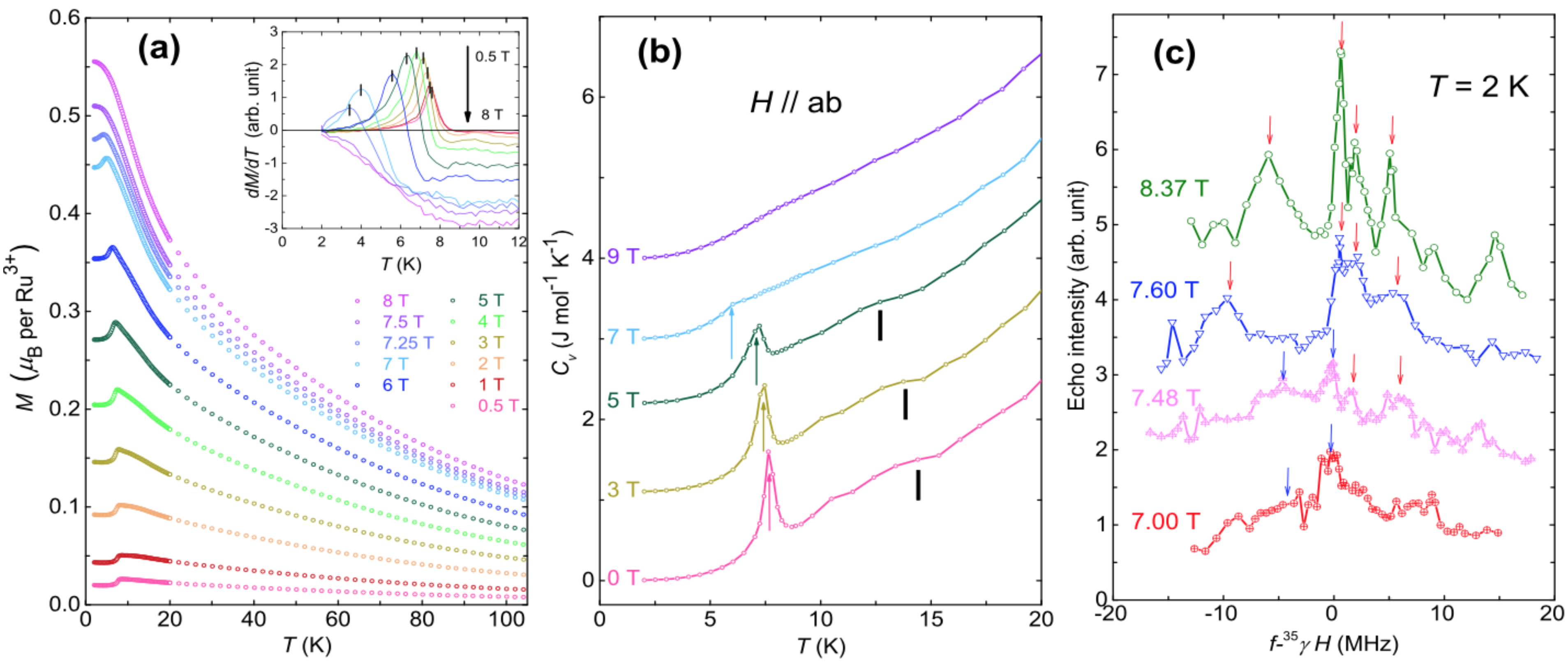
arXiv:1702.08466

Outline

- **Introduction**
- **Spin-wave excitations in α-RuCl3**
- **Fragile magnetic order**

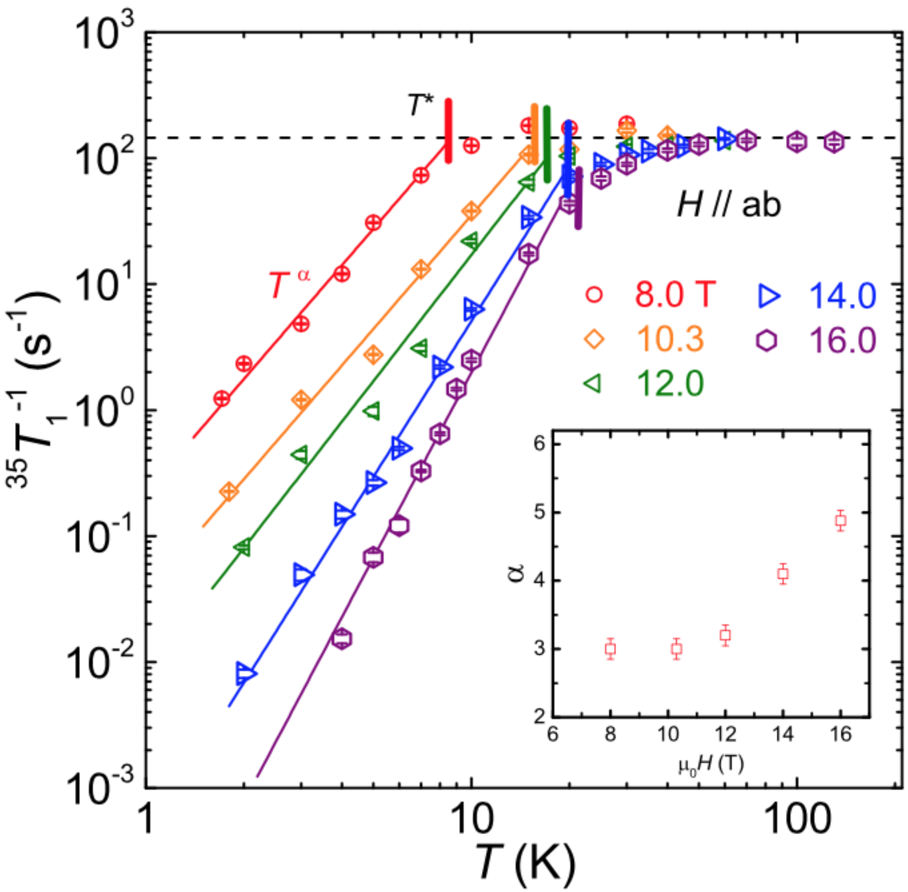
  Magnetic field, pressure, and doping
- **Magnetism in two dimensional**
- **Summary**

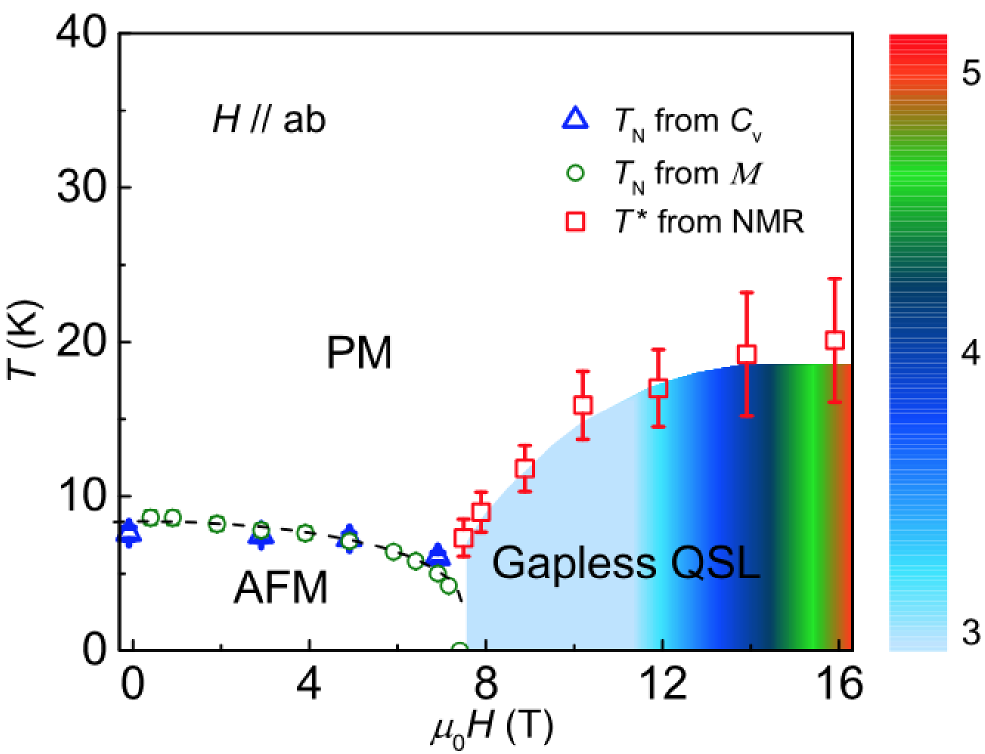
Fragile zigzag order: magnetic field



arXiv:1703.08474

Fragile zigzag order: magnetic field





arXiv:1703.08474

Fragile zigzag order: magnetic field

Phys. Rev. B 91, 094422 (2015)
Phys. Rev. B 91, 180401 (2015)
Phys. Rev. B 92, 235119 (2015)
Phys. Rev. Lett. 118, 187203 (2017)
Phys. Rev. B 95, 180411(R) (2017)
arXiv:1702.01671
arXiv:1703.08474
arXiv:1703.08623
arXiv:1704.03475
arXiv:1706.06157
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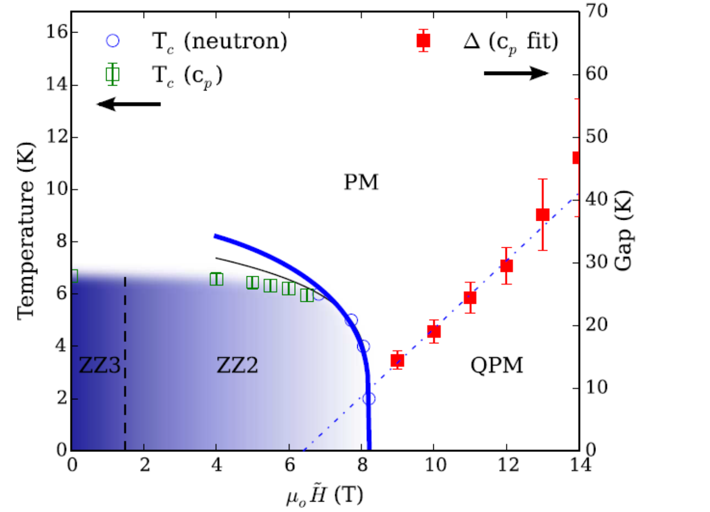
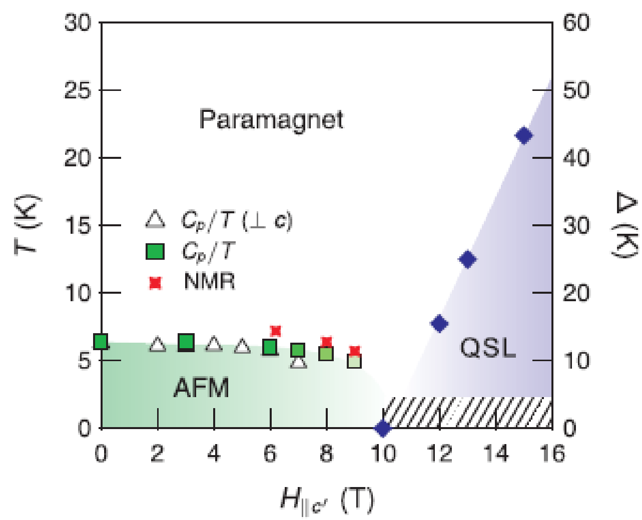
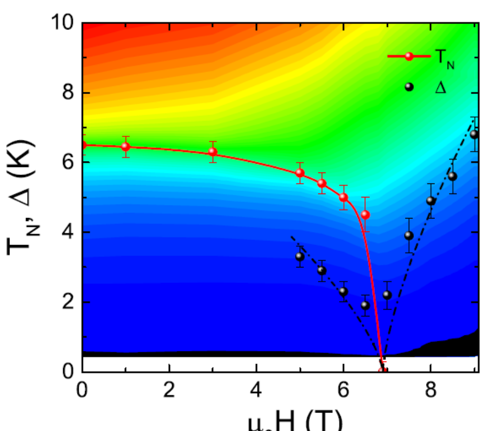
arXiv:1706.07240
arXiv:1706.08455



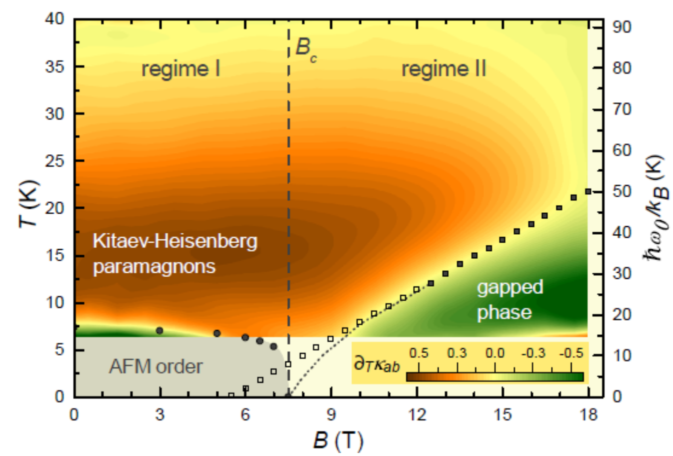
Sears *et al.*, Phys. Rev. B 95, 180411(R) (2017)



Baek *et al.*, arXiv:1702.01671 (2017)

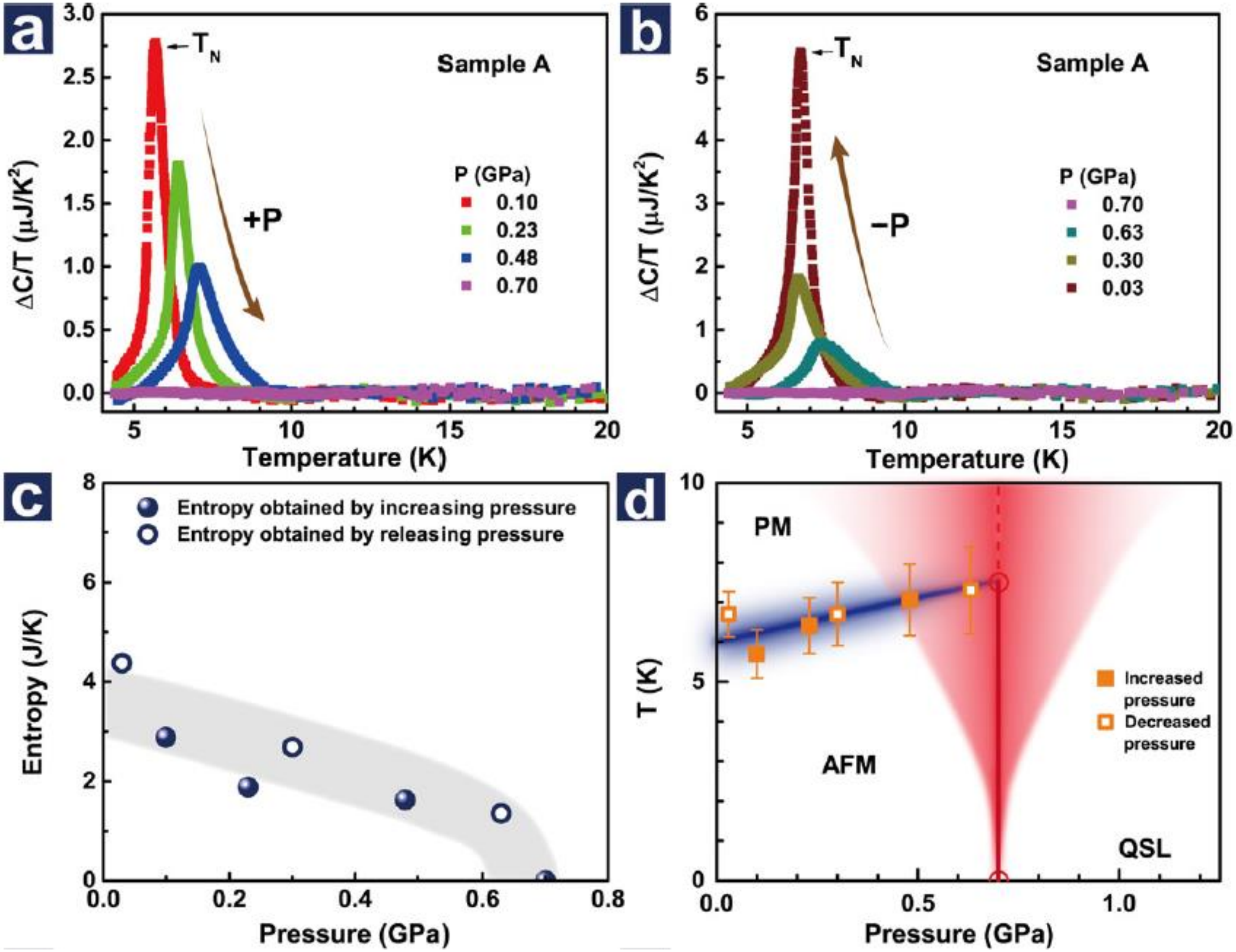


Wolter *et al.*, arXiv:1704.03475



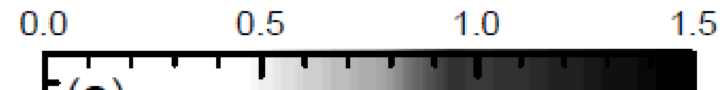
Hentrich *et al.*, arXiv:1703.08623 (2017)

Fragile zigzag order: pressure



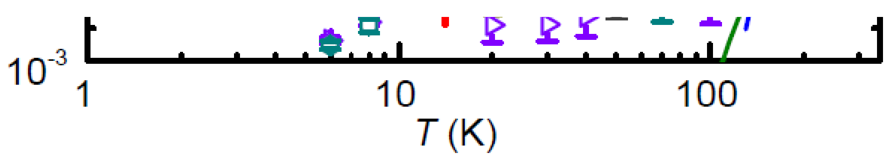
Wang *et al.*, arXiv:1705.06139 (2017)

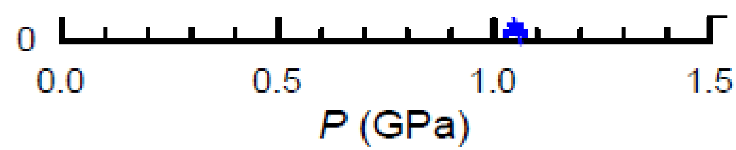
Fragile zigzag order: pressure



Jul. 20

Adam Nahum (Oxford)	Emergent SO(5) symmetry and dualities at deconfined critical points
Nic Shannon (Okinawa Inst. of Sci. and Tech.)	Frustrating quantum spin ice
Wei-Qiang Yu (Renmin Univ.)	Pressure-induced and field-induced Magnetically Disordered State of a-RuCl3 Evidenced by NMR

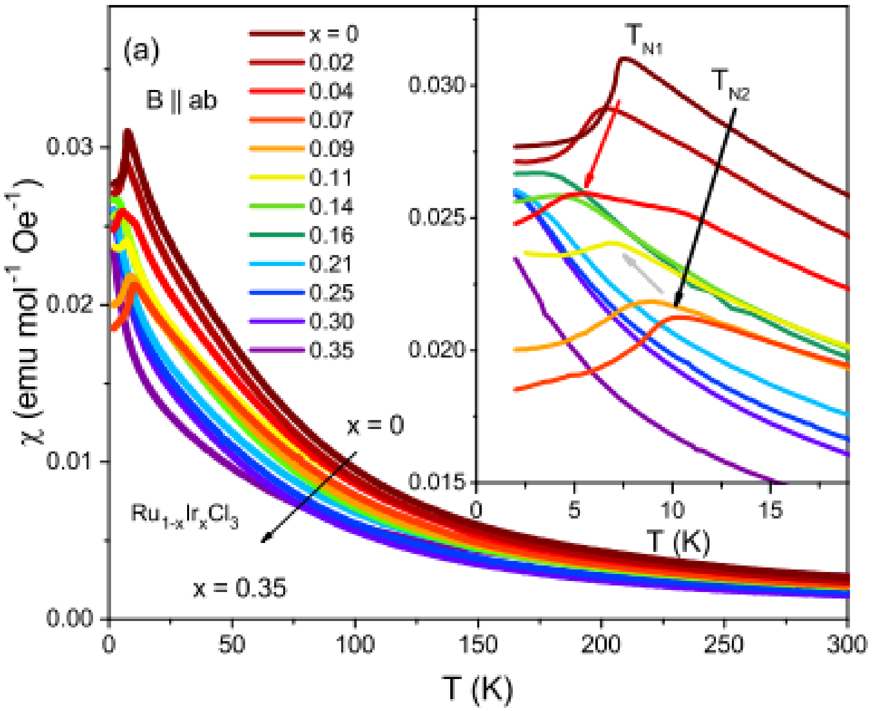


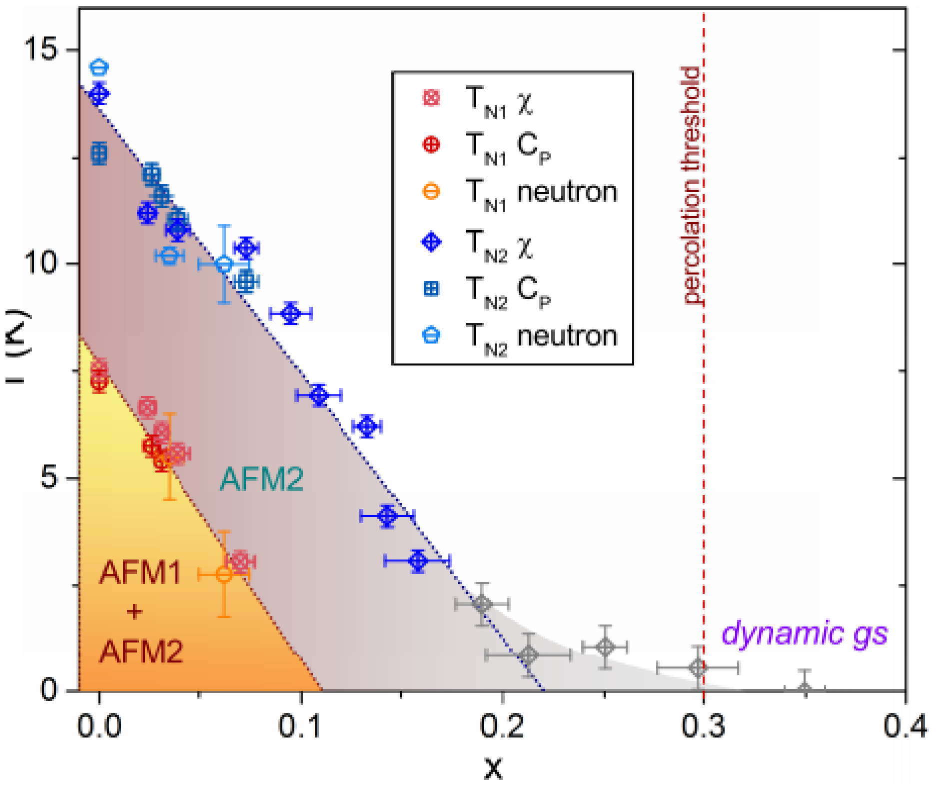


arXiv:1706.02697 (2017)

Fragile zigzag order: doping

Phase diagram of $Ru_{1-x}Ir_xCl_3$





Lampen-Kelley *et al.*, arXiv:1612.07202

Magnetism in 2D

LETTER

doi:10.1038/nature22391

CrI_3

Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit

Bevin Huang[1]*, Genevieve Clark[2]*, Efrén Navarro-Moratalla[3]*, Dahlia R. Klein[3], Ran Cheng[4], Kyle L. Seyler[1], Ding Zhong[1], Emma Schmidgall[1], Michael A. McGuire[5], David H. Cobden[1], Wang Yao[6], Di Xiao[4], Pablo Jarillo-Herrero[3] & Xiaodong Xu[1,2]

LETTER

doi:10.1038/nature22060

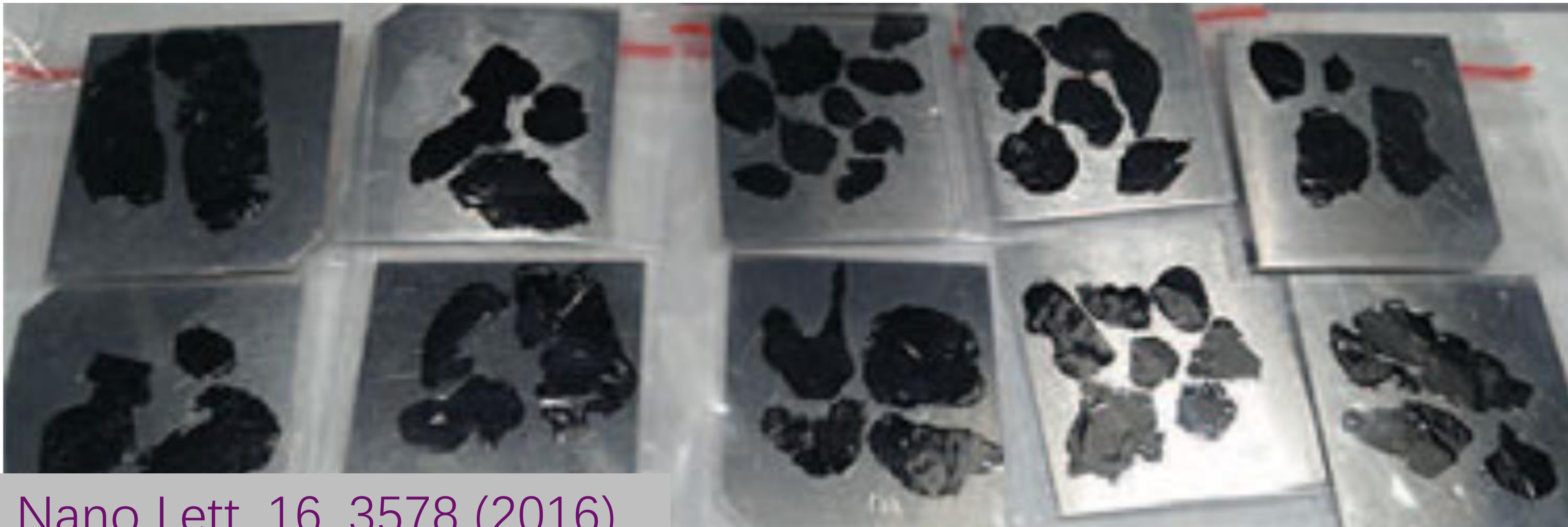
$Cr_2Ge_2Te_6$

Discovery of intrinsic ferromagnetism in two-dimensional van der Waals crystals

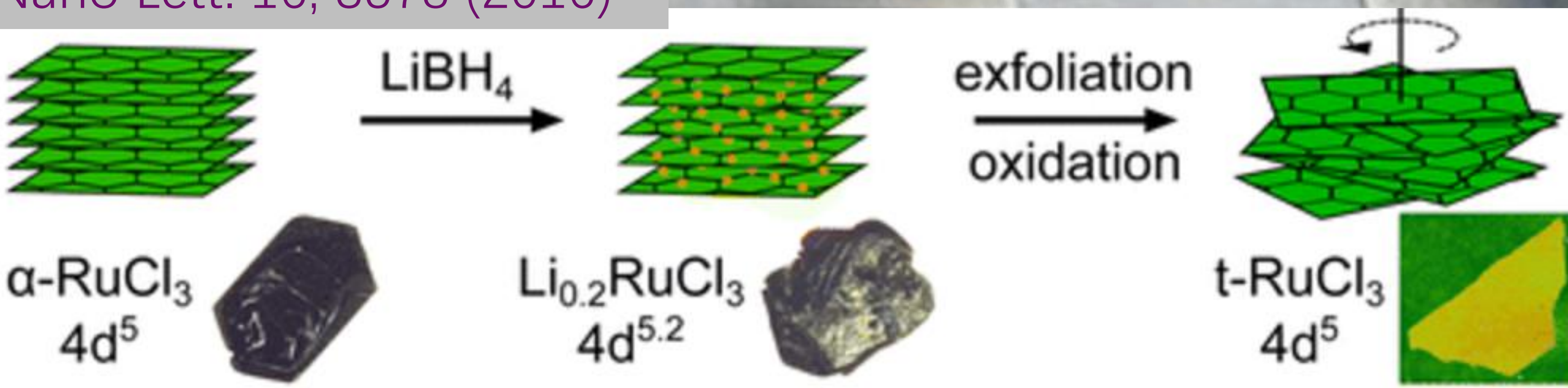
Cheng Gong[1]*, Lin Li[2]*, Zhenglu Li[3,4]*, Huiwen Ji[5], Alex Stern[2], Yang Xia[1], Ting Cao[3,4], Wei Bao[1], Chenzhe Wang[1], Yuan Wang[1,4], Z. Q. Qiu[3], R. J. Cava[5], Steven G. Louie[3,4], Jing Xia[2] & Xiang Zhang[1,4]

Magnetism in 2D

$\alpha-RuCl_3$



Nano Lett. 16, 3578 (2016)



Magnetism in 2D

$CrCl_3$, MX_3

Summary

- **Results on the spin-wave excitations indicate that the Kitaev interaction has been realized in a real material**

  Phys. Rev. Lett. 118, 107203 (2017)

- **High-field induces the quantum-spin-liquid state**

  arXiv:1703.08474

- **The zigzag magnetic order can be tuned by pressure**

  arXiv:1706.02697