Workshop on Muon Physics at the Intensity and Precision Frontiers (MIP 2024)

MuSR study on the quantum magnetism of 2D frustrated compounds

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2024.04.21.

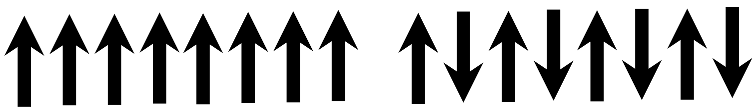
Outline

1. Geometrically Frustrated Magnet
2. Quantum effect in triangular lattice
3. Disorder state of Honeycomb lattice
4. Conclusion and outlook

Geometrically Frustrated Magnet

- Competing or contradictory constraints on a large fraction of the magnetic sites – **geometric magnetic frustration**

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



J < 0 J > 0

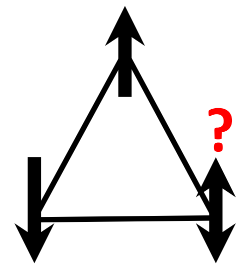


Fig. 2D frustrated model.

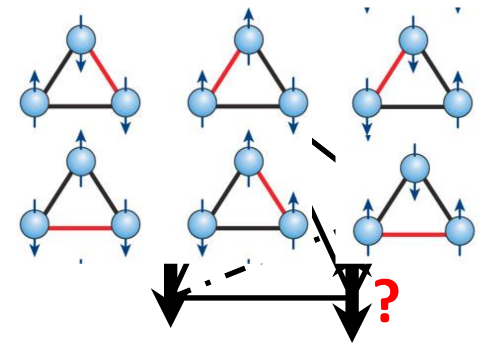


Fig. 3D model.

Geometrically Frustrated Magnet

- Competing or contradictory constraints on a large fraction of the magnetic sites – **geometric magnetic frustration**

- Examples of frustrated lattice
  - Edge/corner sharing triangles
    triangular/kagome lattices (2D)
  - Corner sharing tetrahedra
    Spinel/Pyrochlore lattices (3D)

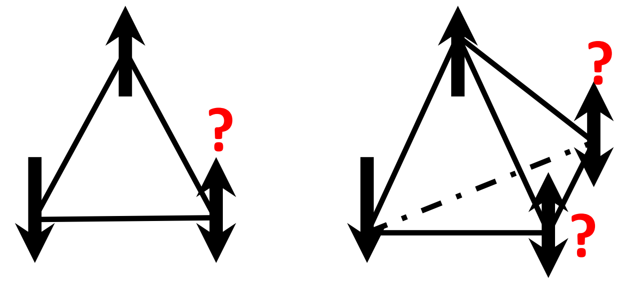


Fig. Frustrated model of 2D and 3D system.

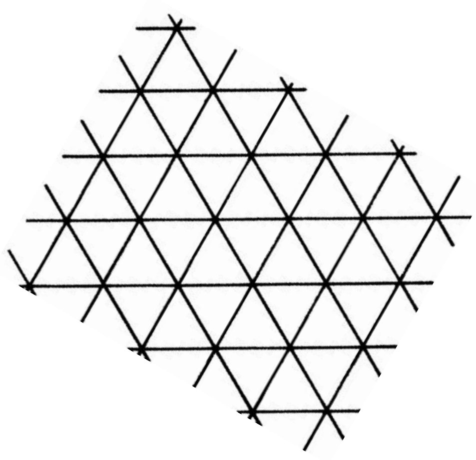


Fig. Triangular lattice.

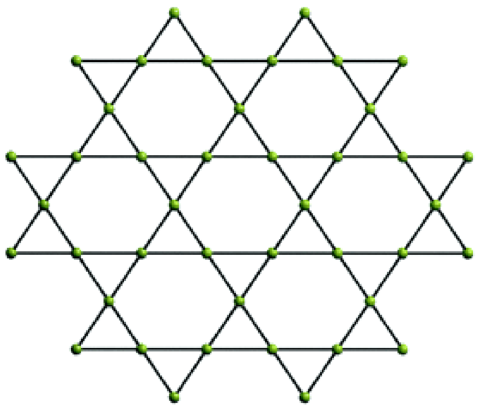


Fig. Kagome lattice.

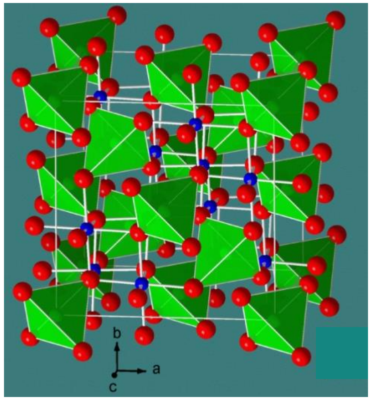


Fig. Spinel lattice AB_2O_4.

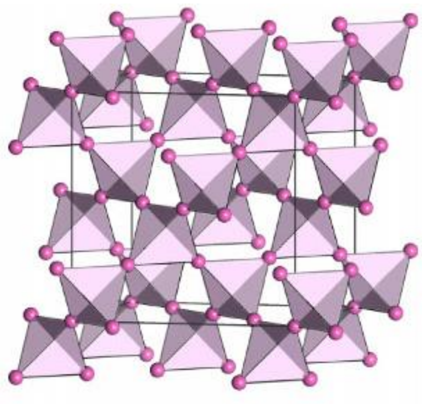


Fig. Pyrochlore lattice $A_2B_2O_7$.

Geometrically Frustrated Magnet

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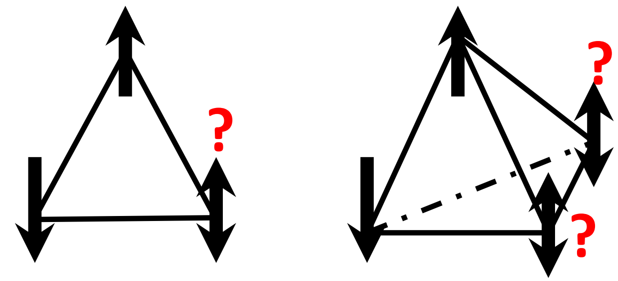
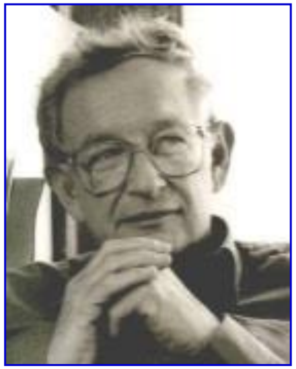
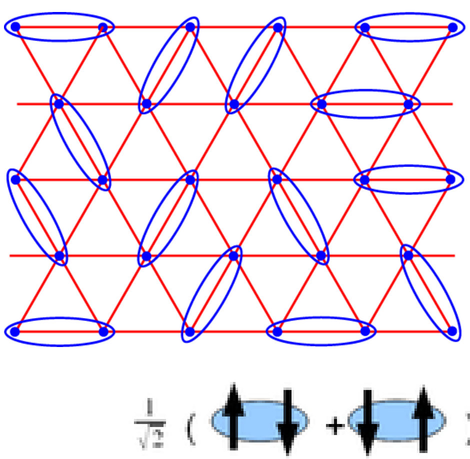


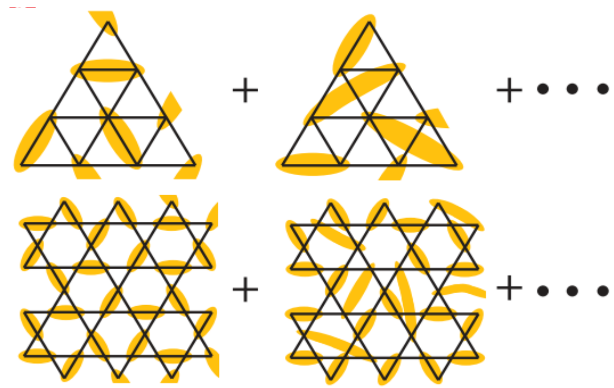
Fig. Frustrated model of 2D and 3D system.

- Superconductor



P.W. Anderson
1973、1987





Physics Today 69, 30 (2016).

Quantum Spin Liquid

- Competing or contradictory constraints on a large fraction of the magnetic sites – **geometric magnetic frustration**
- Quantum Spin Liquid
  – No magnetic long-range ordering
  – No symmetry broken
  – Fractional excitation: spinon
  – Strong fluctuation at low energy

- Examples of quantum-spin-liquid

Material	Lattice	Θ_{cw} (K)	J (K)
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$	anisotropic triangular	-375	250
EtMe$_3$Sb[Pd(dmit)$_2$]$_2$	anisotropic triangular	-375~-325	220-250
YbMgGaO$_4$	Triangular	-4	1.5
Na$_4$Ir$_3$O$_8$	Hyperkagome	-650	430
PbCuTe$_2$O$_6$		-22	15
ZnCu$_3$(OH)$_6$Cl$_2$ (herbertsmithite)	Kagome	-314	170
Cu$_2$Zn(OH)$_6$FBr (barlowite)	Kagome	-200	170
Rb$_2$Cu$_3$SnF$_{12}$	Kagome	-100	154.4
Ca$_3$Cr$_7$O$_{28}$	Distorted Kagome		-9

PRL 91, 107001 (2003); PRL 100, 087202 (2008); PRL 95, 177001 (2005); JACS 130, 2922 (2008); Nature Phys. 6, 865 (2010); Nature 464, 199 (2010); npj Quant. Mater. 4, 12 (2019); npj Comp. Mater. 8, 10 (2022).

Quantum Spin Liquid

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- Quantum Spin Liquid
  - No magnetic long-range ordering
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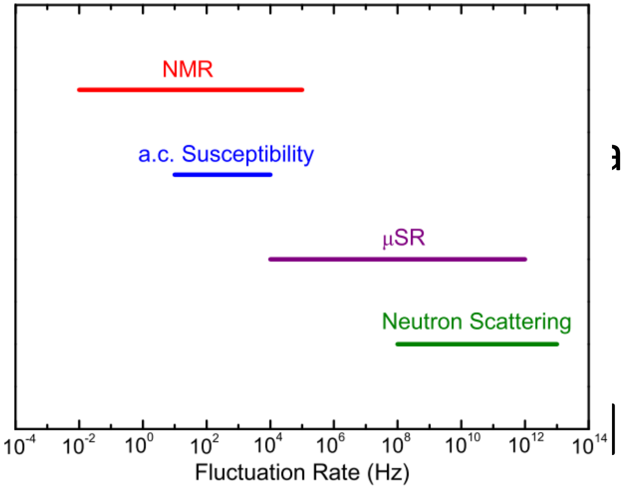
  - Strong fluctuation at low energy

- Identification of quantum-spin-liquid

• **Specific heat measurements**: the low-ener
• **Thermal transport measurements**: localize
• **Neutron scattering/Nuclear-magnetic resc** ations;
• **Reflectance measurements**: power-law op



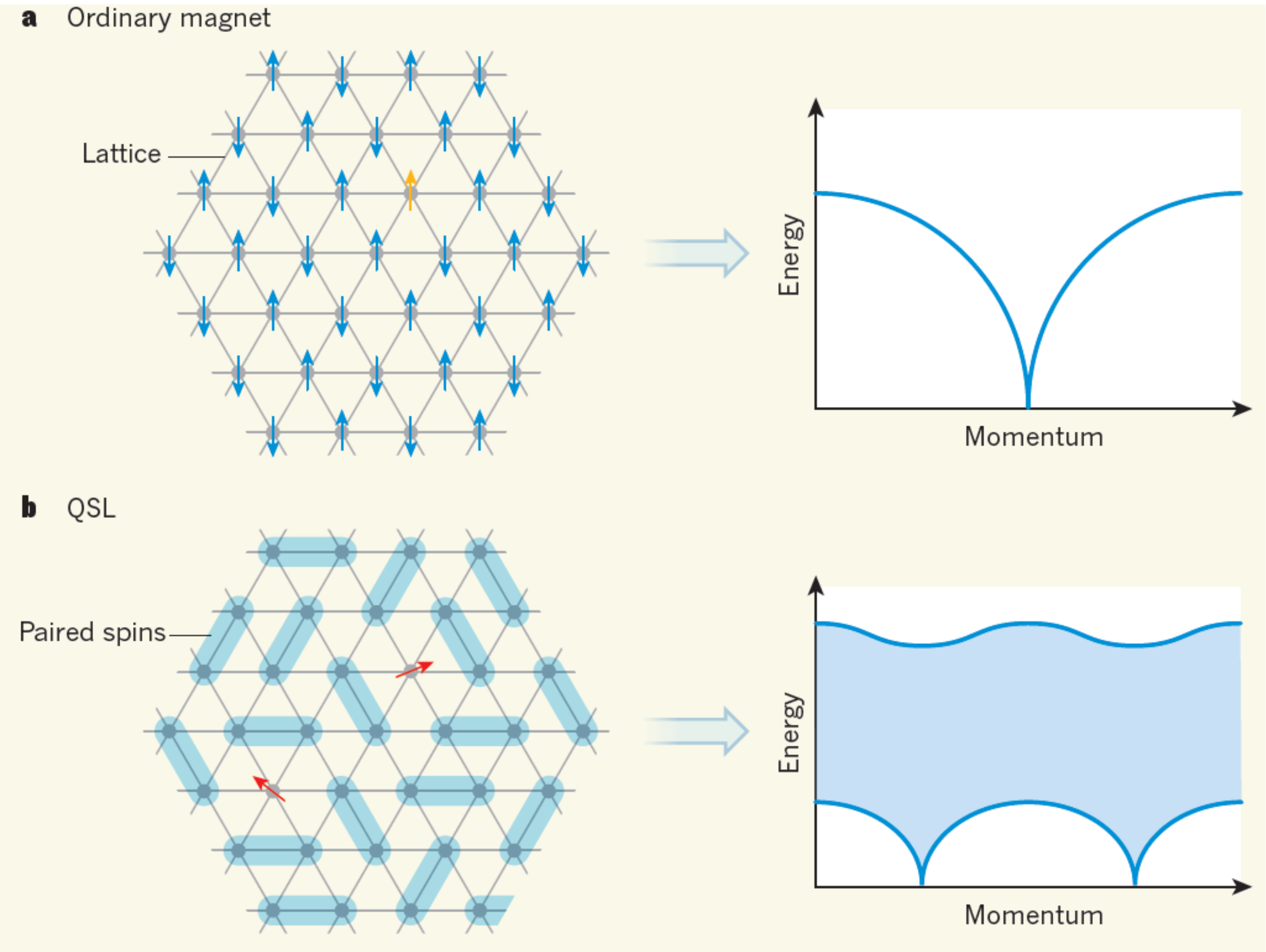
Frustration

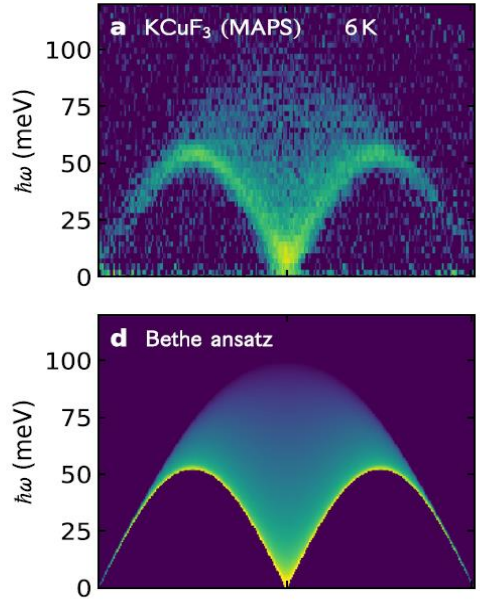
$$f = \frac{|\Theta_{cw}|}{T_c}$$

S|

Geometrically Frustrated Magnet

- Competing or contradictory constraints on a large fraction of the magnetic sites – **geometric magnetic frustration**
- Examples of frustrated lattice

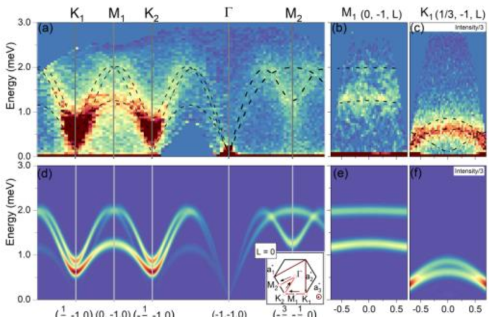




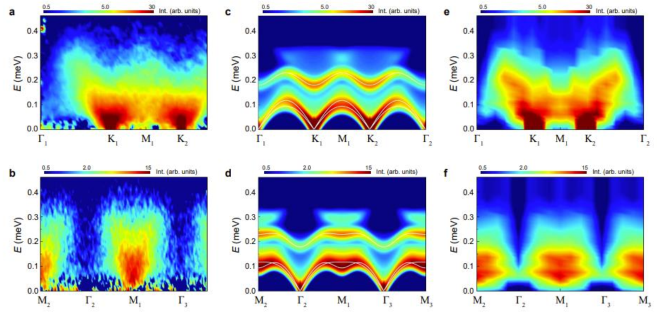
Nature 540 22 (2016); Nat. Phys. 17, 726 (2021); Nat. Commn. 13, 5796 (2022)

Is there a pure QSL?

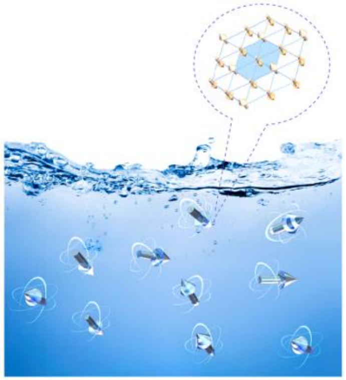
$Ba_3MnSb_2O_9$

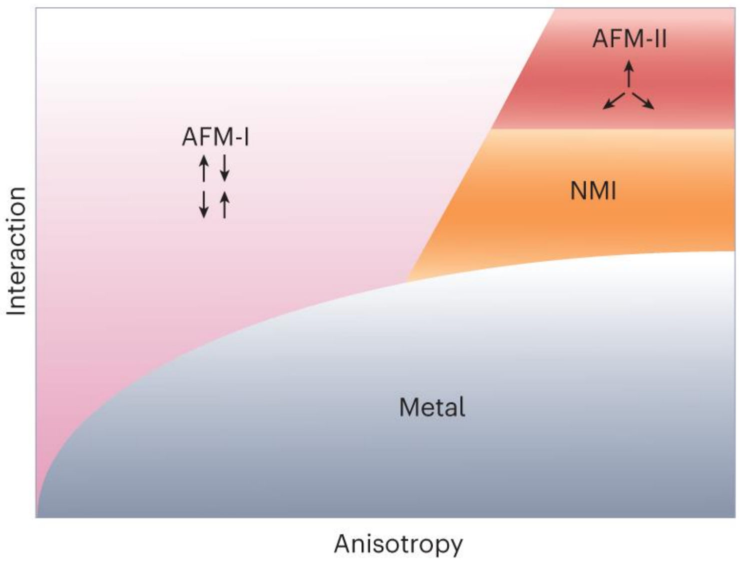


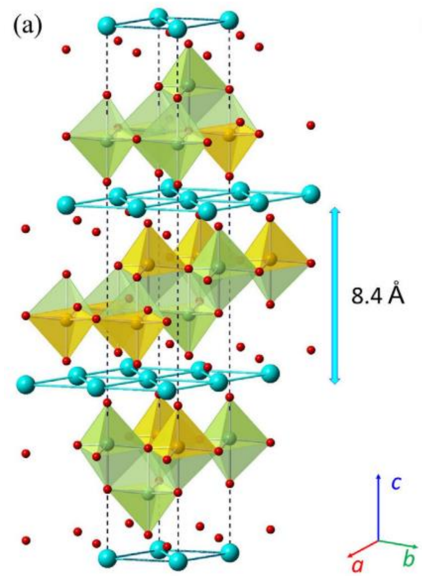
$Na_2BaCo(PO_4)_2$

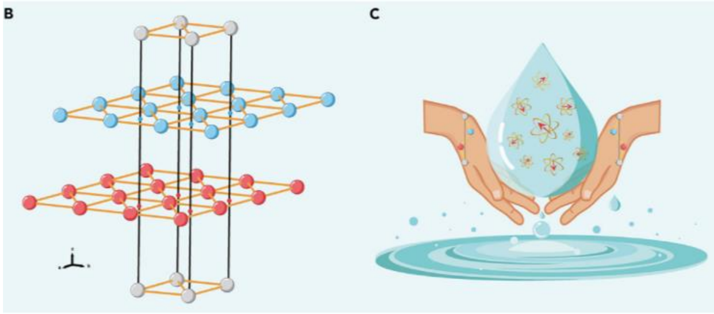


PRB 108, 174424 (2023); arxiv 2402.07730 (2024)



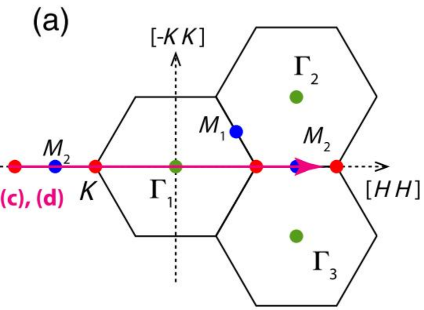


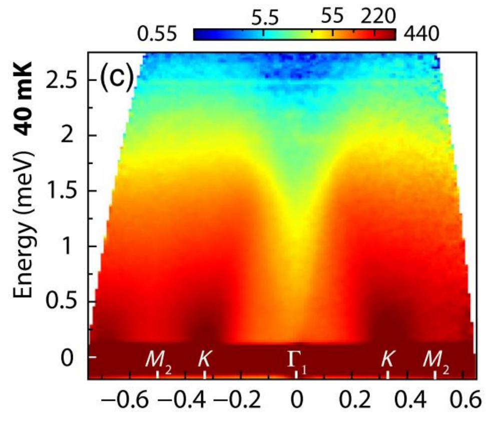


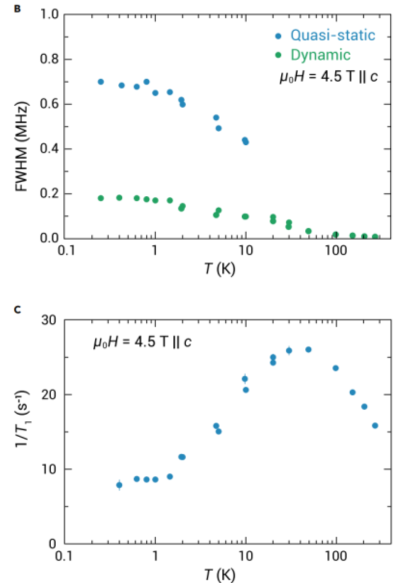


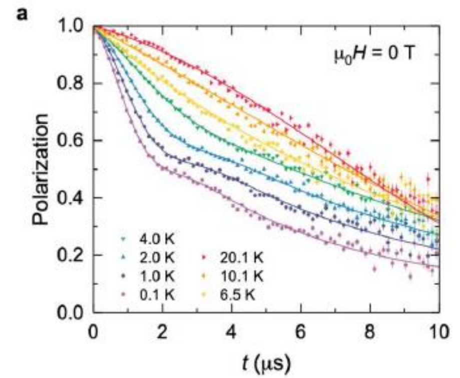
Adv. Quant. Technol., 1900089 (2019); The Innov. **4**, 100484, (2023); Nat. Phys. 19, 922 (2023).

NaYbSe$_2$

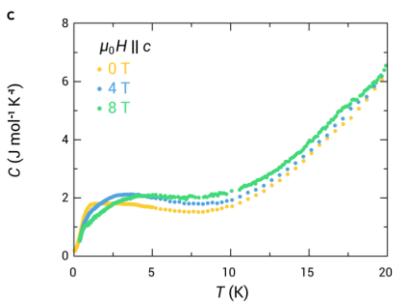


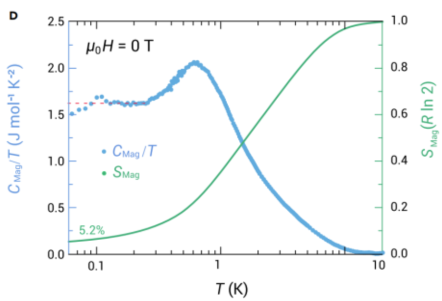


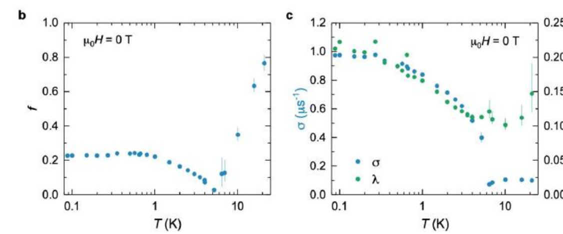




$$P(t) = fG_{KT}(\sigma, t) + (1 - f)\, exp(-\lambda t),$$





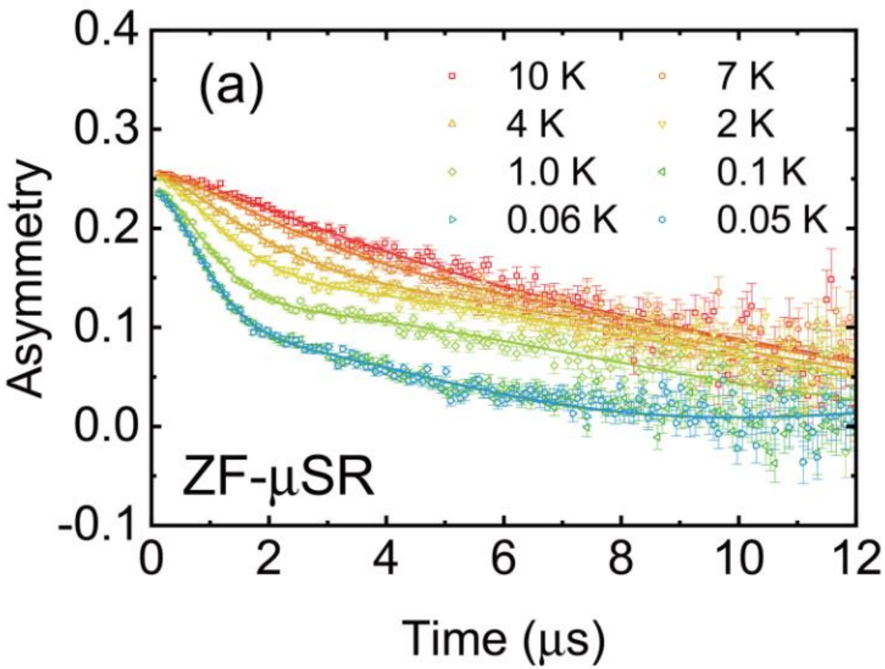


P. Dai, et al., PRX 11, 021044 (2021); Z. Zhu et al., The Innov. 4, 100459 (2023).

(a) QSL's evidences:

1) C_m/T (<0.5 K) is almost a constant, indicating the spinon Fermi surface spin liquid;
2) the μSR spectra show no spin freezing with robust dynamics and strong quantum fluctuations;
3) Continuous spin excitations by INS.
4) NMR indicates the coexistence of short-range magnetic order and QSL

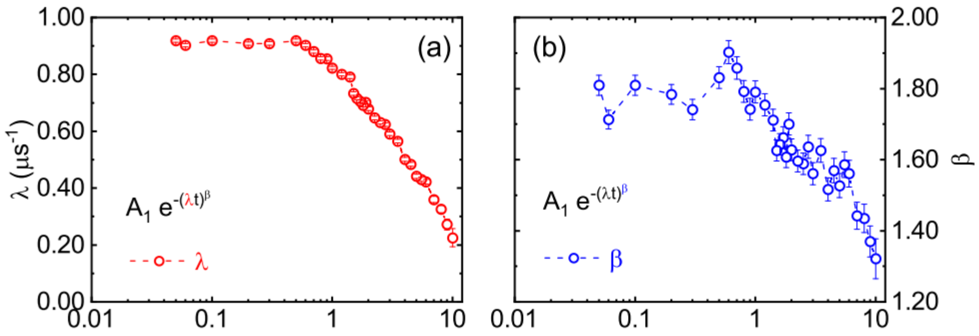
μSR study of $NaYbSe_2$



$$A(t) = A_0 + A_{SE} + A_{KT},$$

$$A_{SE} = A_1 e^{-(\lambda t)^{\beta}},$$

$$A_{KT} = A_2\left[\tfrac{2}{3}(1-\sigma^2 t^2)e^{-\frac{1}{2}\sigma^2 t^2} + \tfrac{1}{3}\right],$$



Z. Zhang, et al., PRB 106, 085115 (2022

1) No oscillation is seen in the ZF-μSR spectra down to 50mK;
2) No spin-glass-like freezing
3) Both λ and β go up to a plateau below 0.3 K, indicating the emergence of a stable magnetically disordered QSL ground state.

Outline

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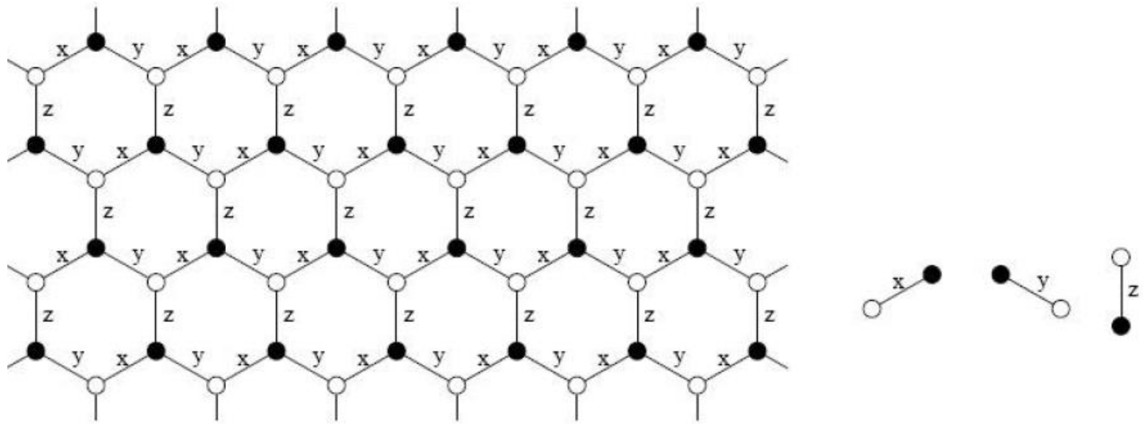
2. Quantum effect in triangular lattice
3. Disorder state of Honeycomb lattice
4. Conclusion and outlook

Kitaev model

- Anyons in an exactly solved model



$$H = \sum_{<ij>_x} J_x S_i^x S_j^x + \sum_{<ij>_y} J_y S_i^y S_j^y + \sum_{<ij>_z} J_z S_i^z S_j^z$$

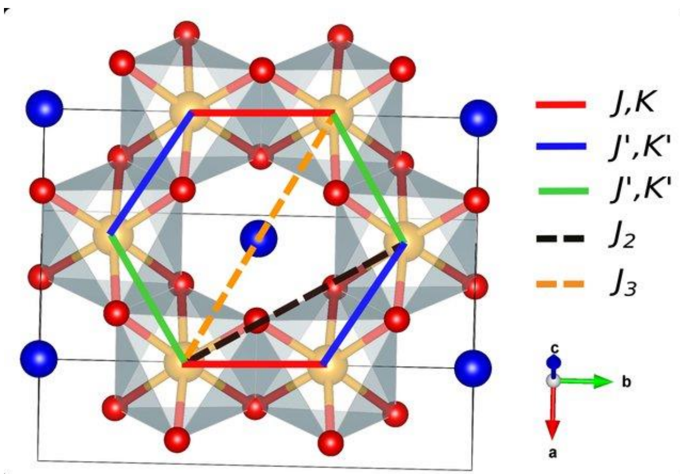
arXiv: cond-mat/0506438v3

- **Energy gaps:** local excitations
- **Topological Quantum Numbers:** excitations stable
- **Topological Order**

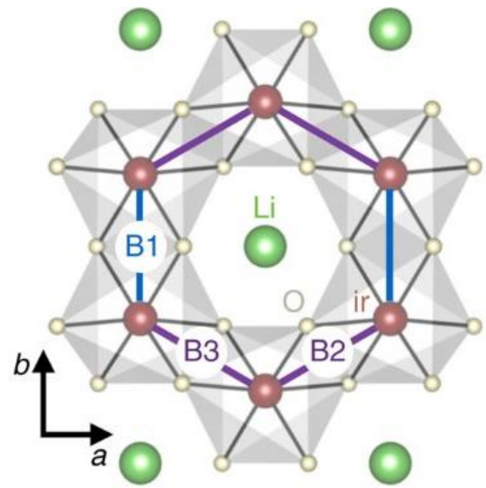
Honeycomb-Kitaev

$4d/5d$ transition mental

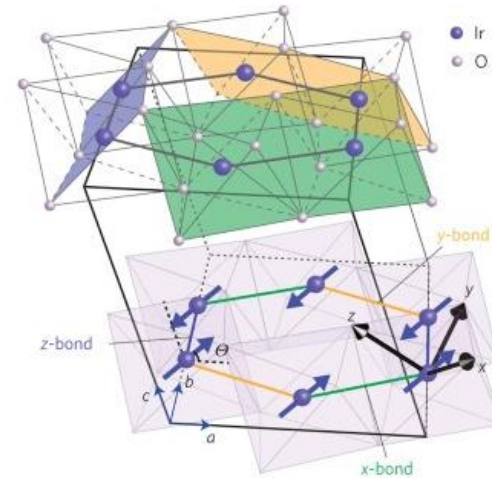
- $H_3LiIr_2O_6$



- α-Li_2IrO_3



- α-Na_2IrO_3



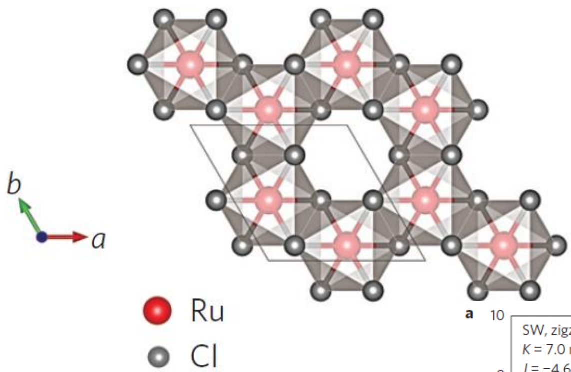
R. Yadav, et. al., PRL **121**, 197203 (2018); S. Nishimoto, et. al., Nat. Comm. **7**, 10273 (2016);
S. Hwan Chun, et al., Nat. Phys. **11**, 462 (2015).

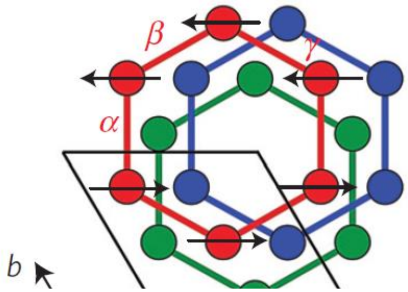
The Kitaev model extended to a Heisenberg-Kitaev model:

1) Kitaev term K,
2) off-diagonal symmetric exchange term Γ and Γ',
3) J (J_{NN}), and the third NN coupling J_3

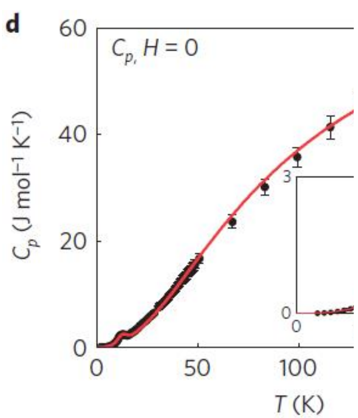
α-$RuCl_3$

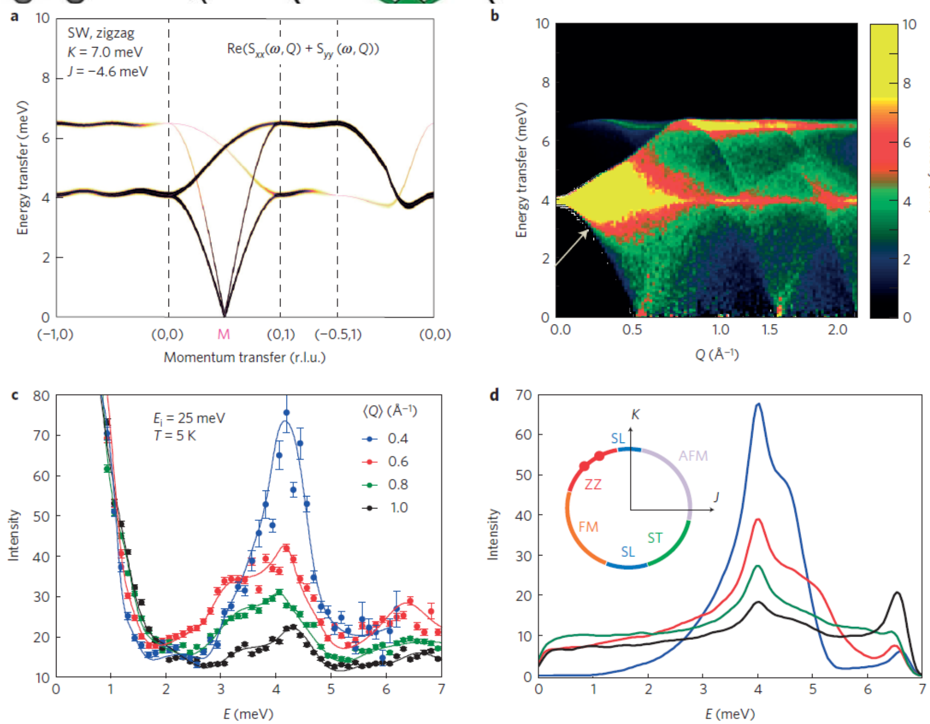
- **α-$RuCl_3$**

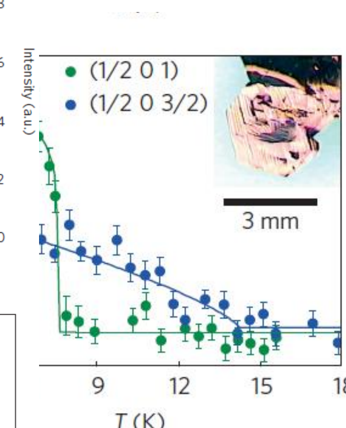




- Dominant Kitaev term *K*
- Magnetic ordered at 0 T while disordered at 7 T (potential QSL)
- Quantum spin liquid



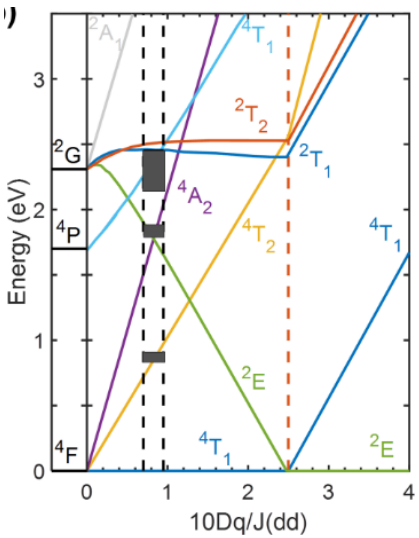


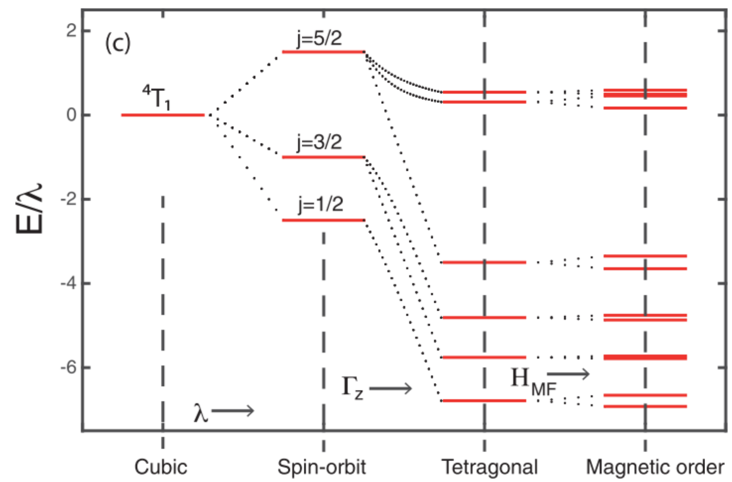


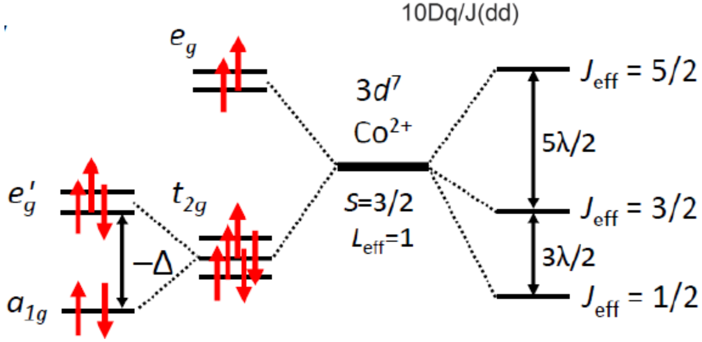
al. Nat. Mater. **15**, 733 (2016)

3*d* transition metal

- **Co^{2+} $3d^7$**







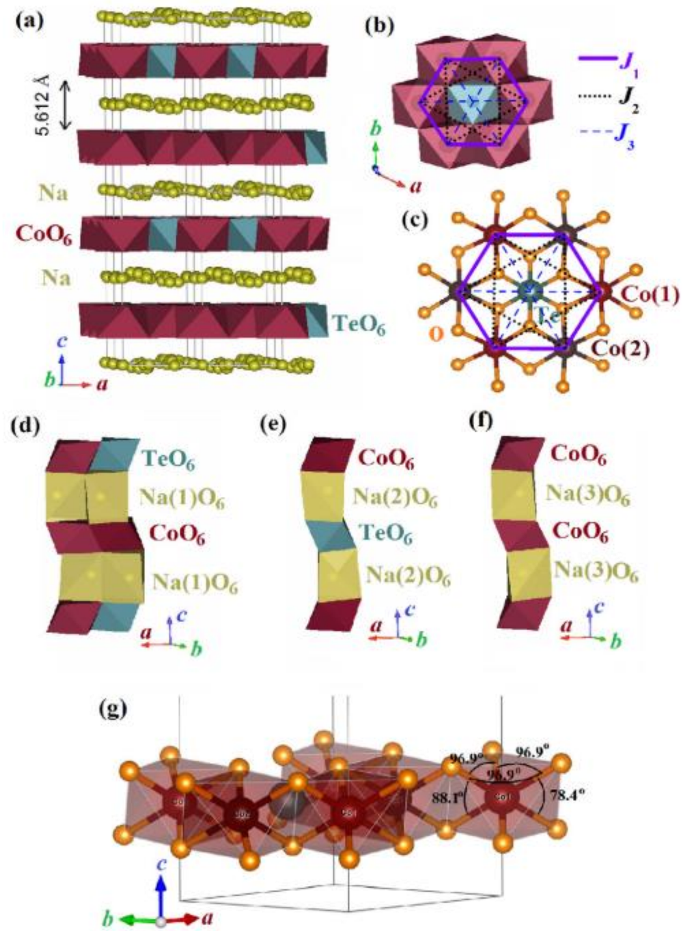
Splitting of the degenerate d^7 states

- Octahedral and Trigonal crystal field in a single-electron picture
- Spin-orbit coupling in a multi-electron picture

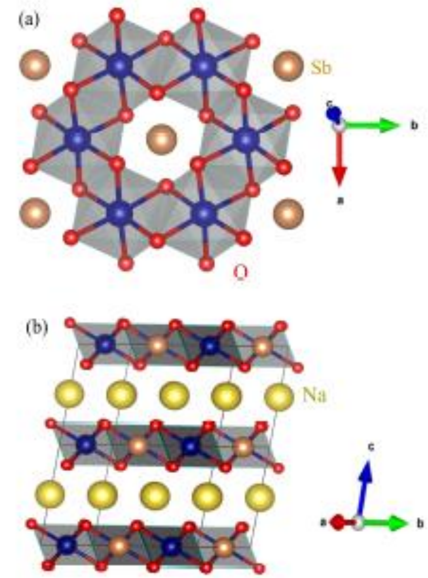
A. K. Bera, et al. Phys. Rev. B 95, 094424 (2017); C. Kim et al., arXiv:2012.06167 (2020).

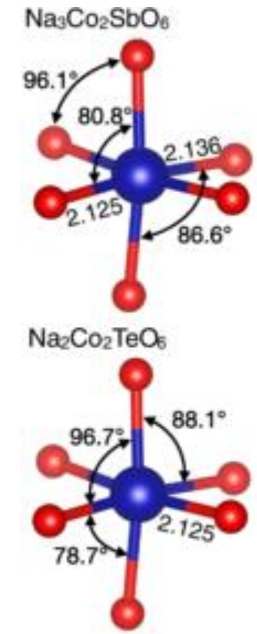
$Na_2Co_2TeO_6$ and $Na_3Co_2SbO_6$

- $Na_2Co_2TeO_6$



- $Na_3Co_2SbO_6$

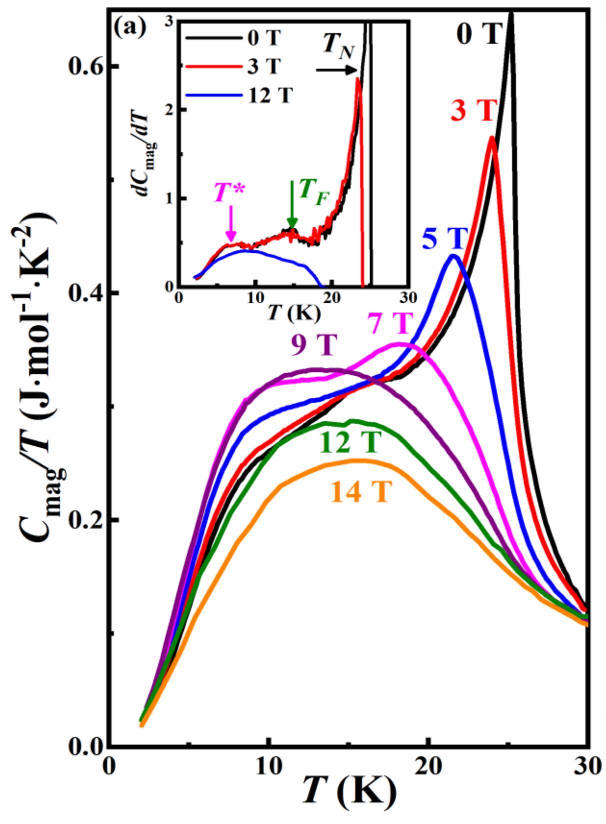


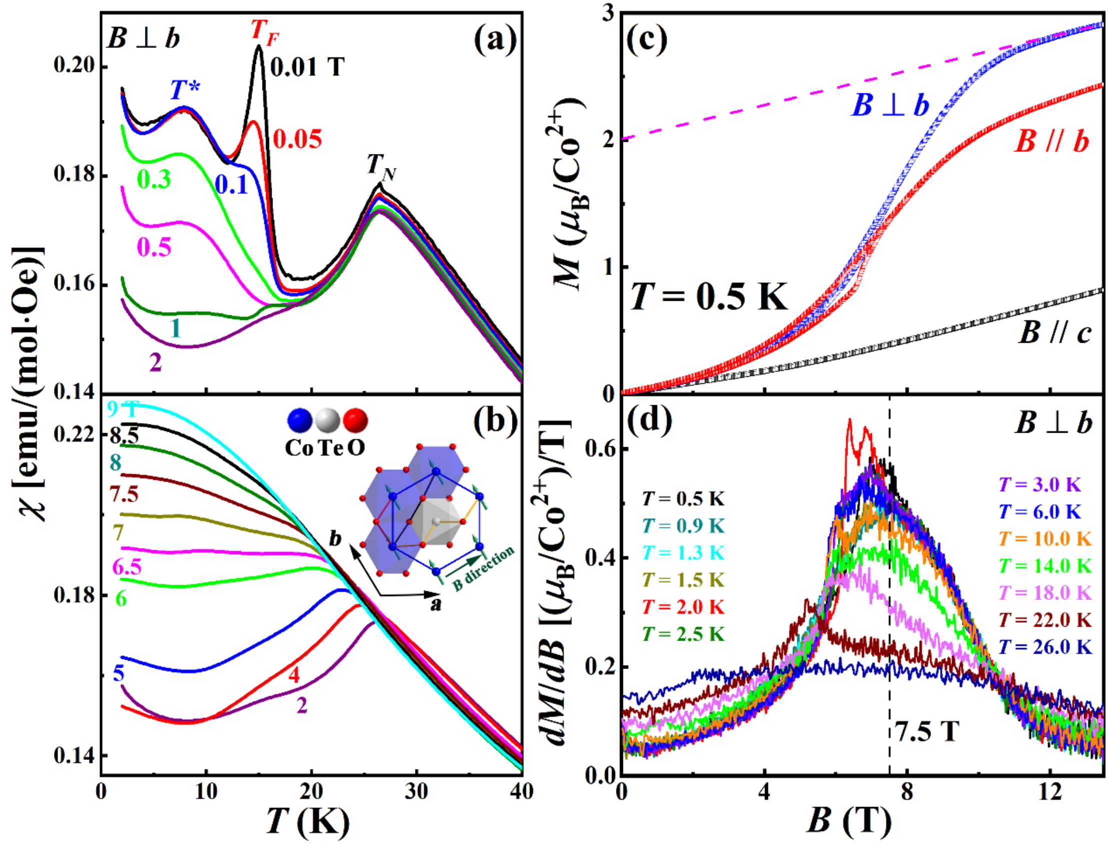


- $Na_3Co_2SbO_6$ and $Na_2Co_2TeO_6$ have the similar structure
- Distortions of $Na_2Co_2TeO_6$ and $Na_3Co_2SbO_6$ are different

M. Songvilay et al., PRB **102**, 224429 (2020); A. K. Bera, et al., PRB **95**, 094424 (2017); J. Yan, et al., PRM **3**, 074405 (2019)

Heat capacity & Magnetic susceptibility





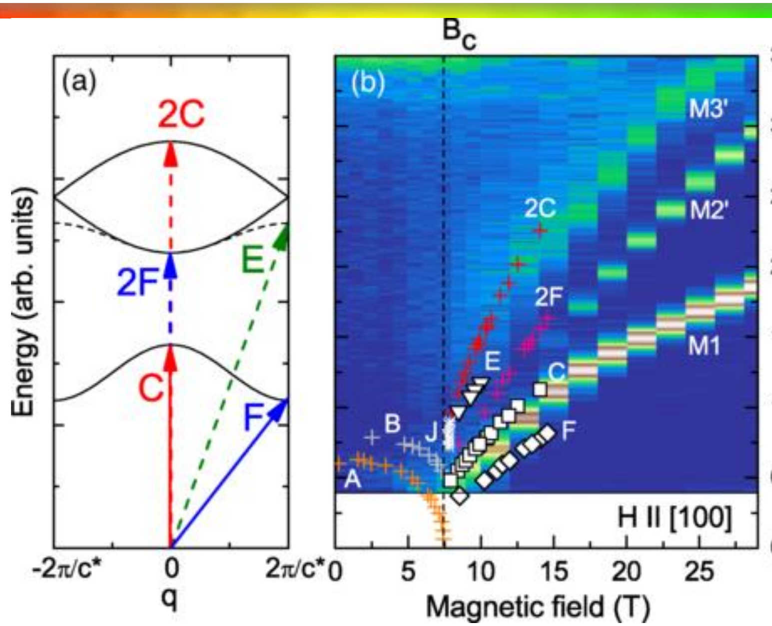
- With the field,

  1) Long-range ordering disappeared;

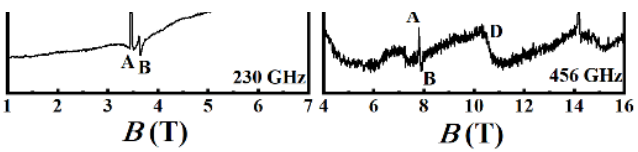
  2) Anisotropy between ab-plane and c-axis;

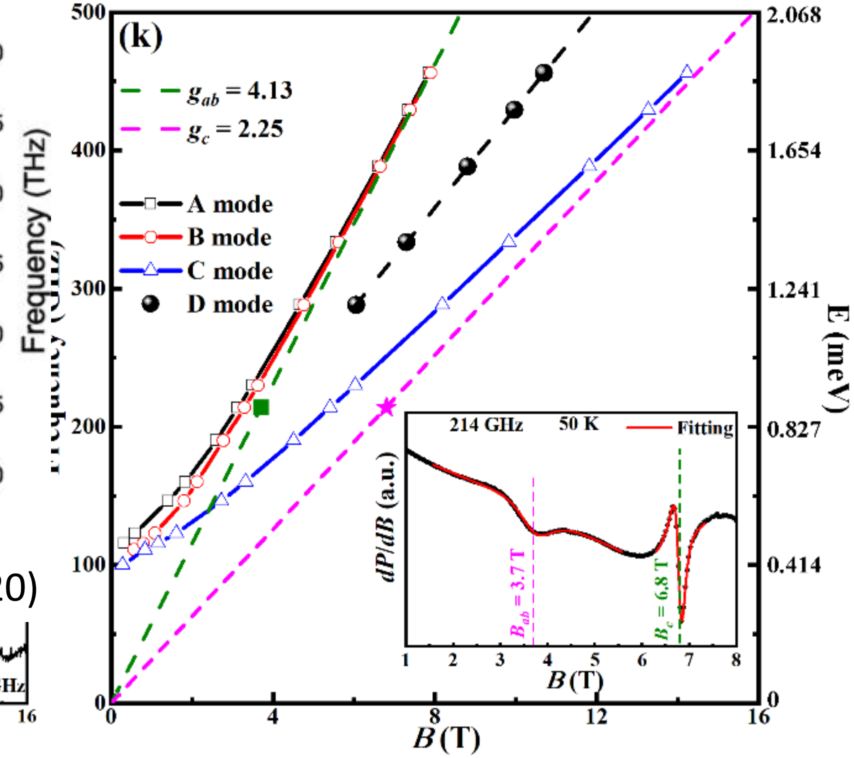
  3) All susceptibilities crossed at around 11.5 T.

High-field electron spin resonance



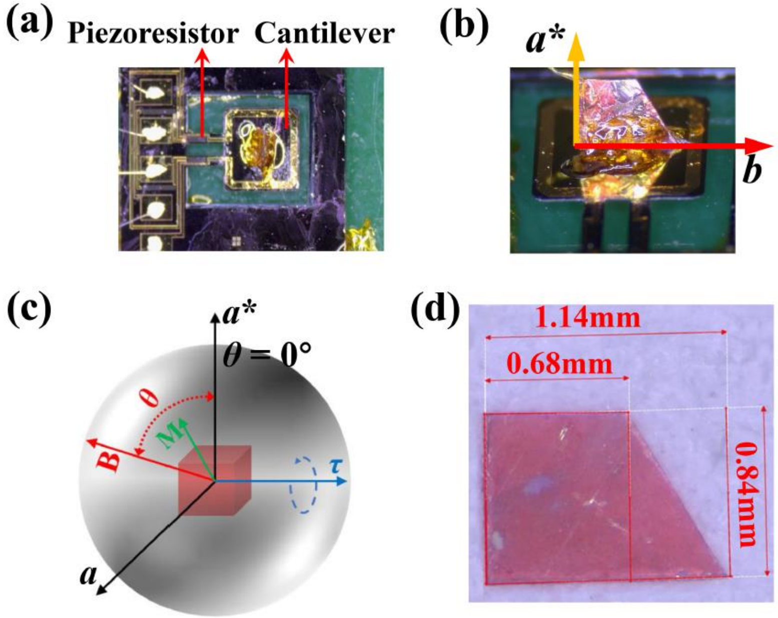
A. Ponomaryov, et. al., PRL **125**, 037202 (2020)

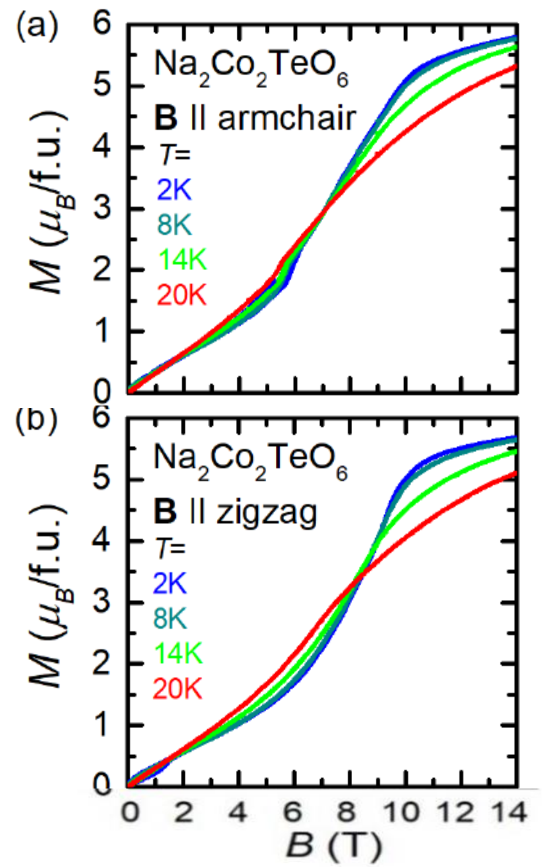


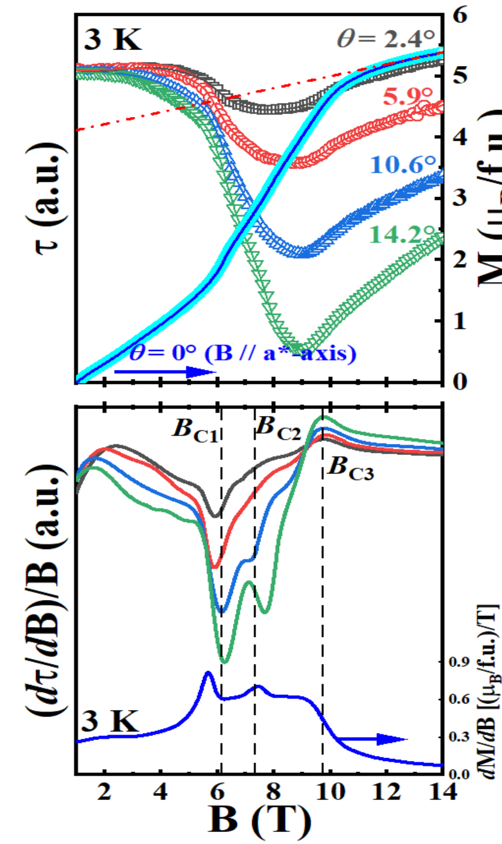


- Four modes are observed:
  1) A and B modes almost overplot on each other, g_{ab}~4.13;
  2) C mode is gapped with g_c~2.25;
  3) D mode is broad and weak, between A/B and C lines.

Magnetic torque measurements

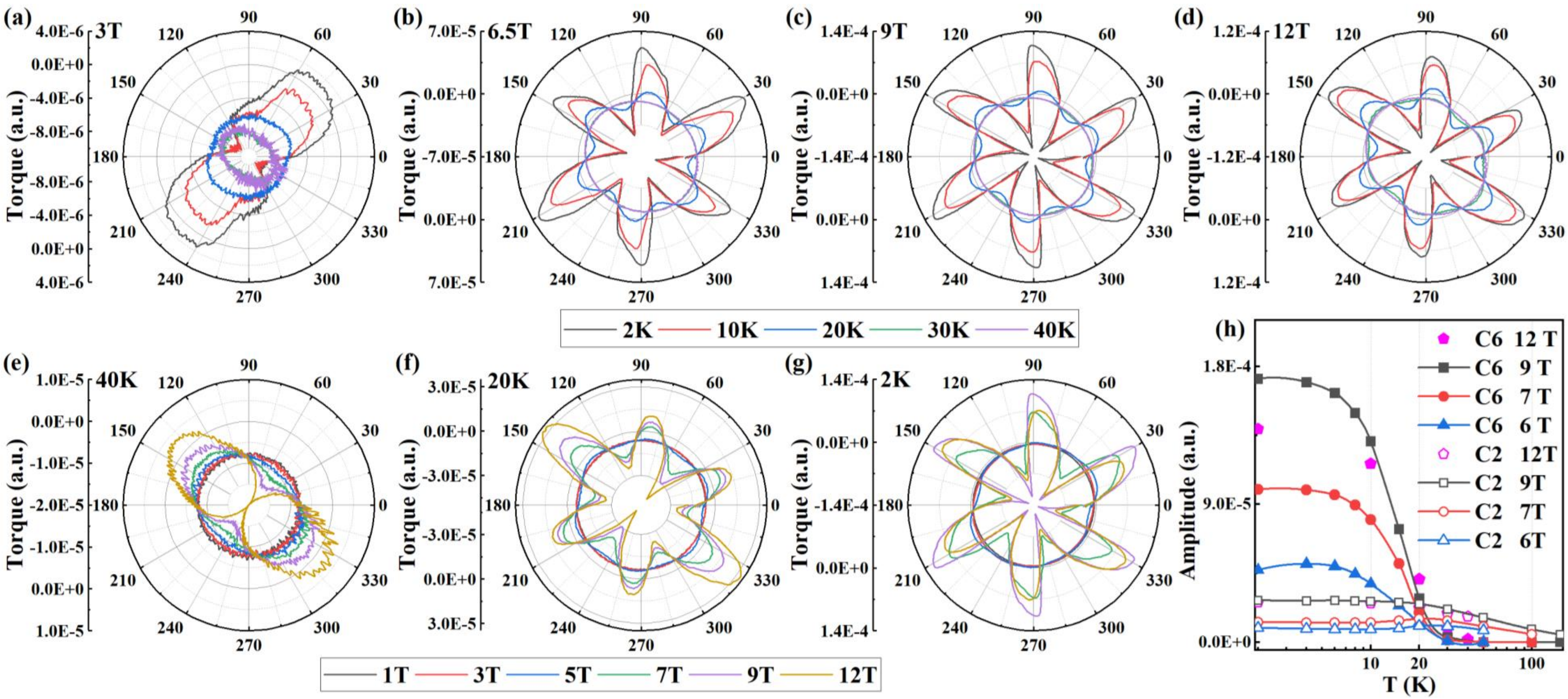






- Differential magnetic susceptibility and $\frac{1}{B}\frac{d\tau}{dB}$,
  - reflects the off-diagonal magnetic susceptibility；
  - three transition: B_{C1} ~ 6 T, B_{C2} ~ 7.5 T and B_{C3} ~ 10 T for B // **a***.
  - the region between B_{C2} and B_{C3} is a phase distinct from the trivial polarized phase.

Temperature- and field-dependence of torque



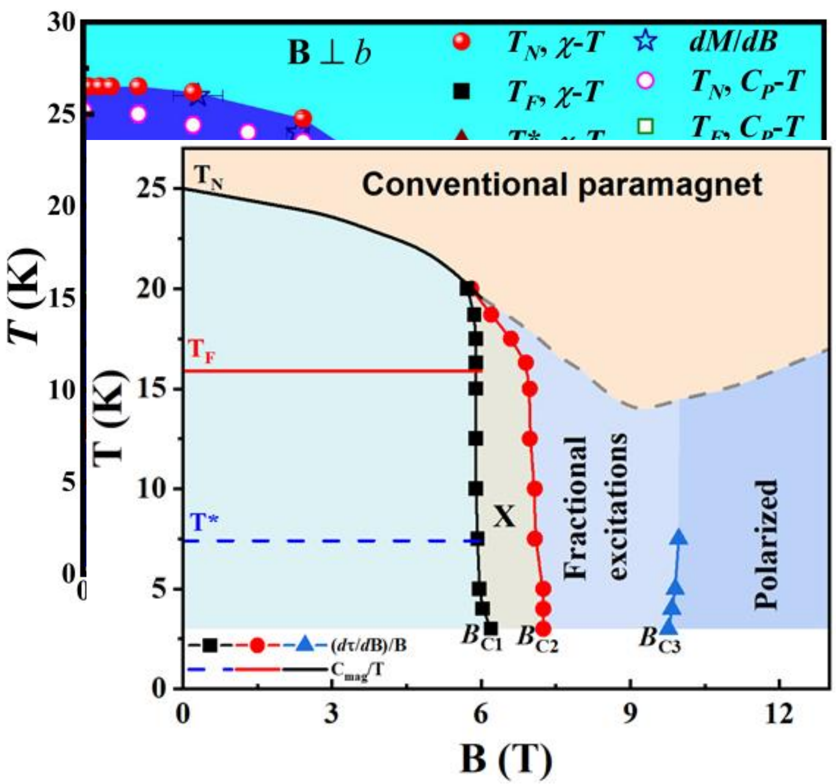
Between B_{C2} and B_{C3}, perfect 6-fold symmetry and the amplitude of the torque is larger than the polarized phase above B_{C3}, indicating a disordered ground state with obvious quantum fluctuations ;

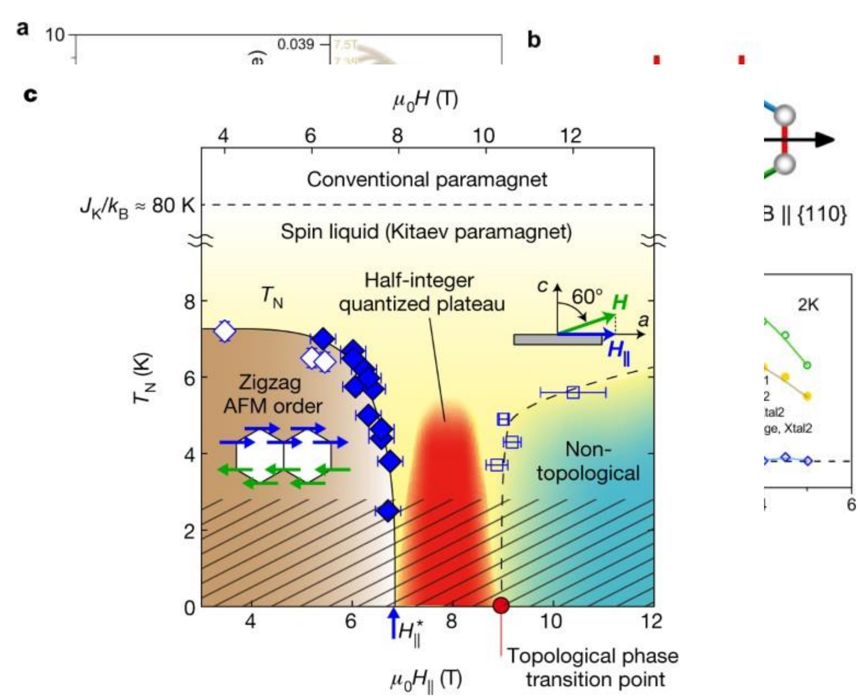
H vs T phase diagram, B // a*-axis

- Phases are complicated

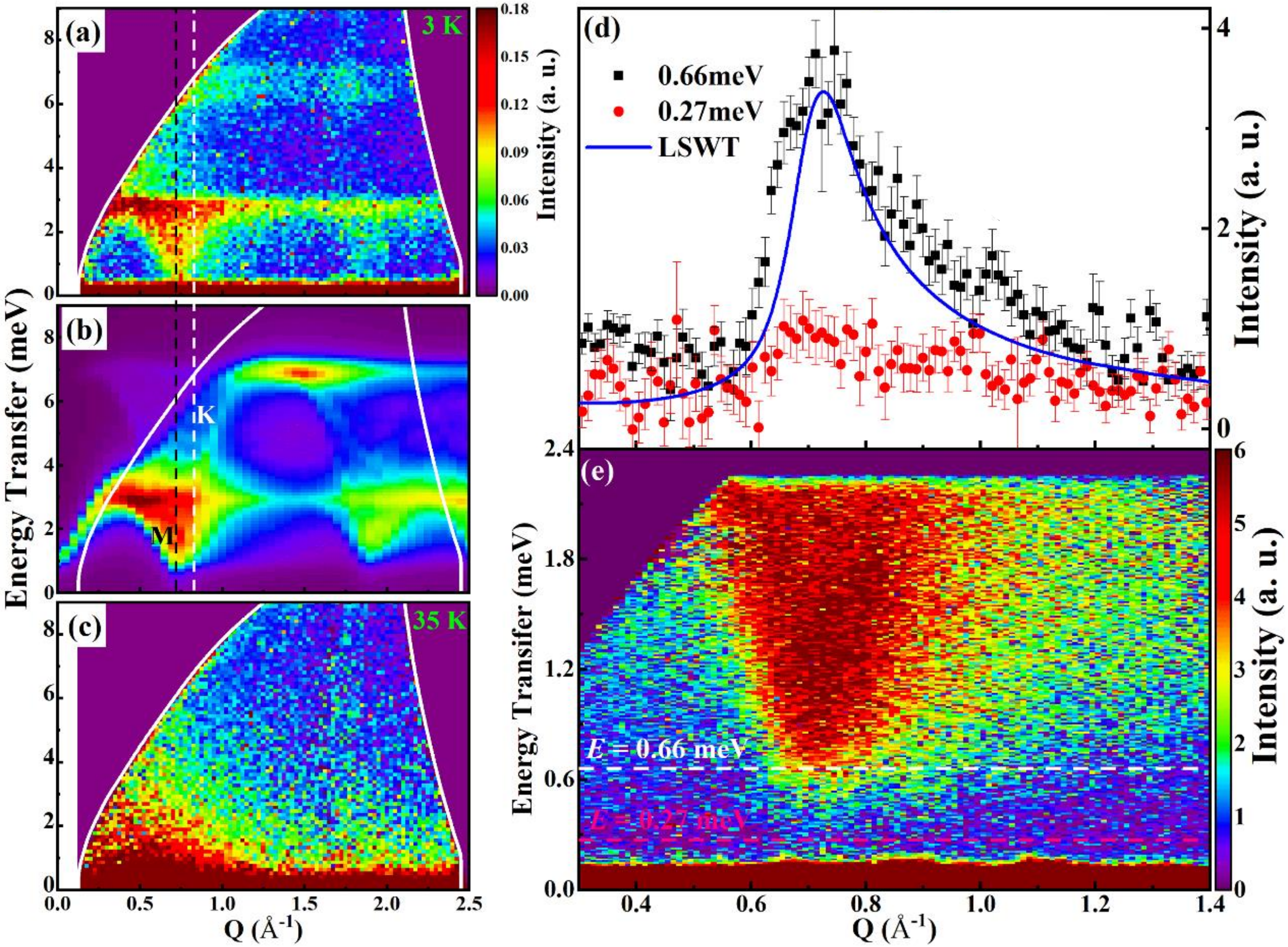
  1) QSL-like phase is observed

  2) No plateau is observed, only spin flip





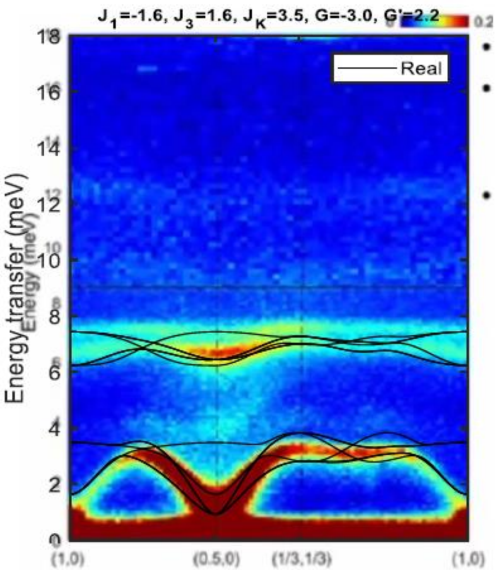
Spin-dynamics



$$H' = \sum_{\langle i,j \rangle} [J\mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha)$$

$$+ \Gamma' (S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\beta S_j^\gamma + S_i^\gamma S_j^\beta)] + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{\langle\langle\langle i,j \rangle\rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

Comparisons



Our parameter well agree with the measured neutron data

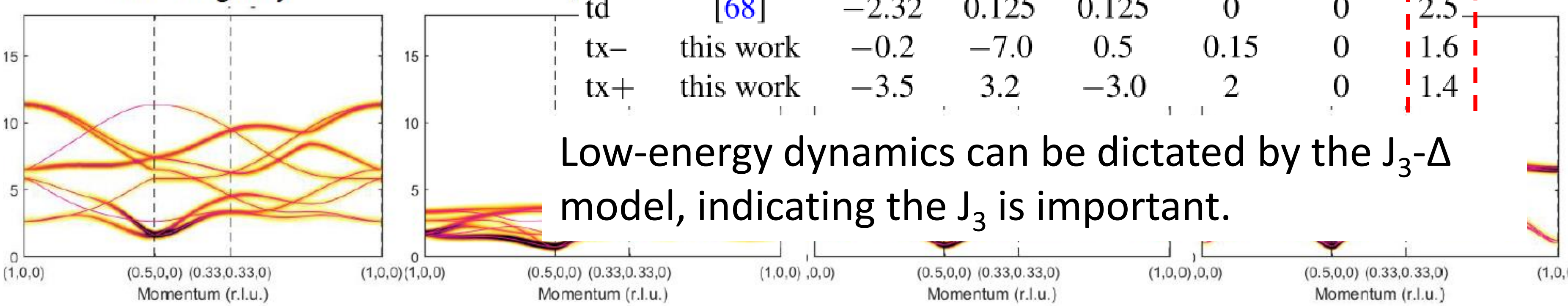
-> the flat-like continuous at ~9 meV and ~12 meV might came from the two-magnon continuum (it also agree with our calculation)

$Na_2Co_2TeO_6$		J	K	Γ	Γ′	J_2	J_3
ta−	[53]	−0.1	−9	1.8	0.3	0.3	0.9
tb−	[54]	−0.1	−7.4	−0.1	0.05	0	1.4
tb+	[54]	−1.5	3.3	−2.8	2.1	0	1.5
tc−	[55]*	−0.2	−7	0.02	−0.23	0.05	1.2
tc+	[55]*	−3.2	2.7	−2.9	1.6	0.1	1.2
td	[68]	−2.32	0.125	0.125	0	0	2.5
tx−	this work	−0.2	−7.0	0.5	0.15	0	1.6
tx+	this work	−3.5	3.2	−3.0	2	0	1.4

(meV)
Y. Li
M. Sor 3
Large 25
Small

M. Songvilay

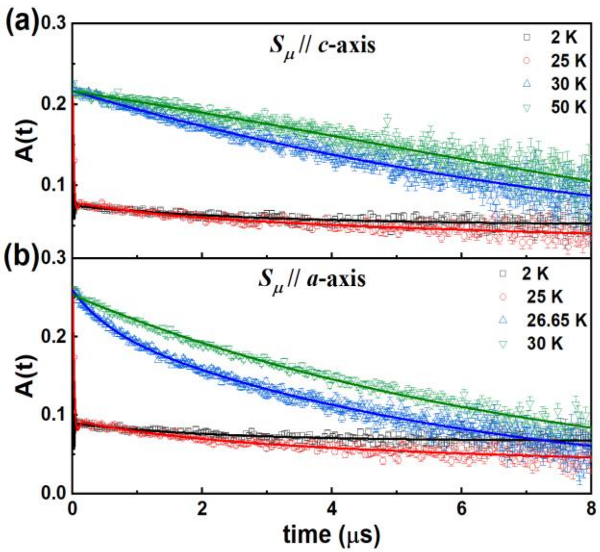
Y

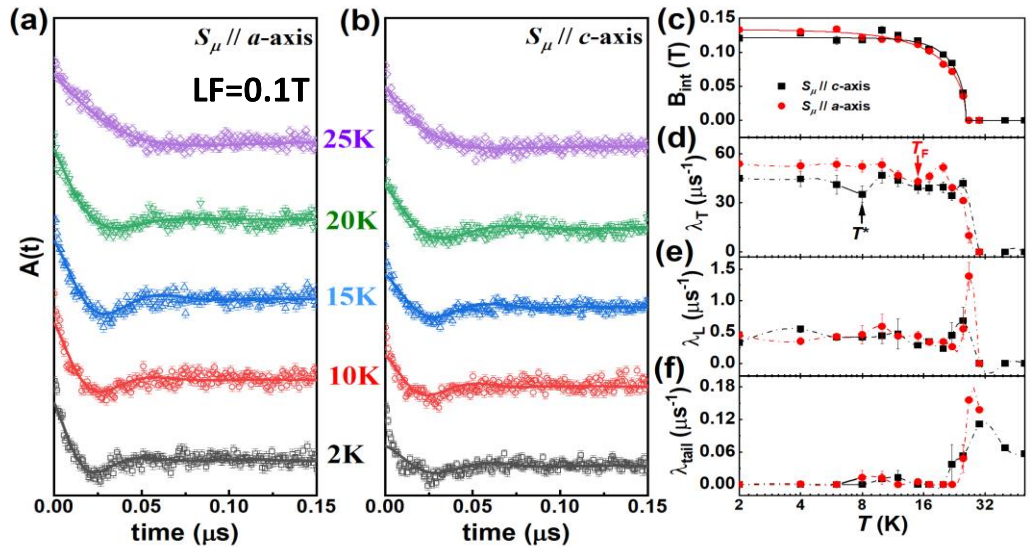


Low-energy dynamics can be dictated by the J_3-Δ model, indicating the J_3 is important.

PRB 103, L180404 (2021); PRB 102, 224429 (2020); JPCM 34, 045802(2021); PRL 129, 147202 (2022); PRB 106, 014413 (2022).

μSR by M20D at TRIUMF

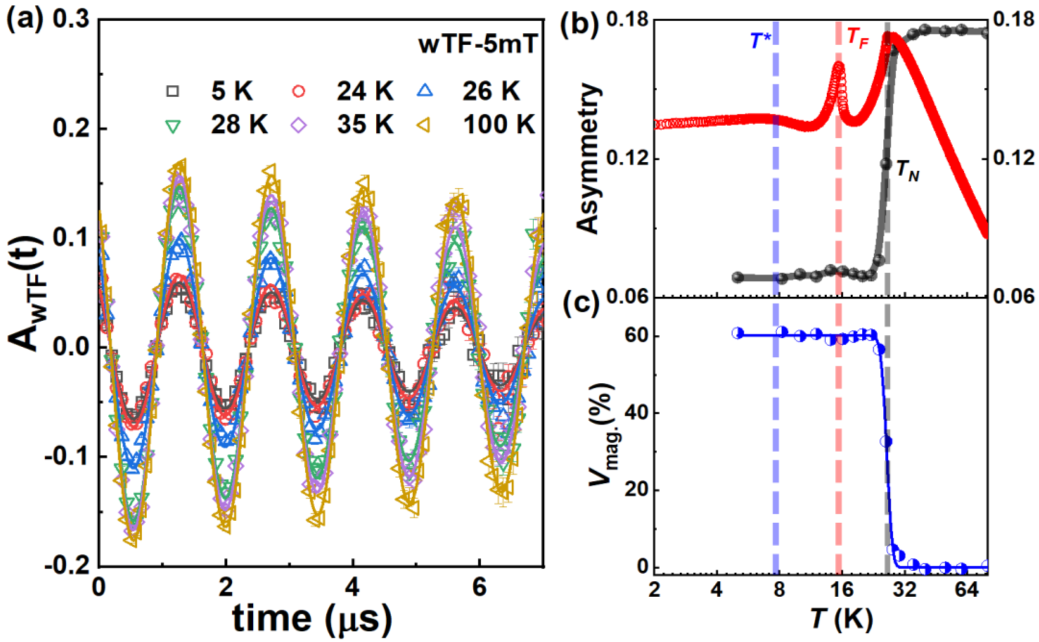


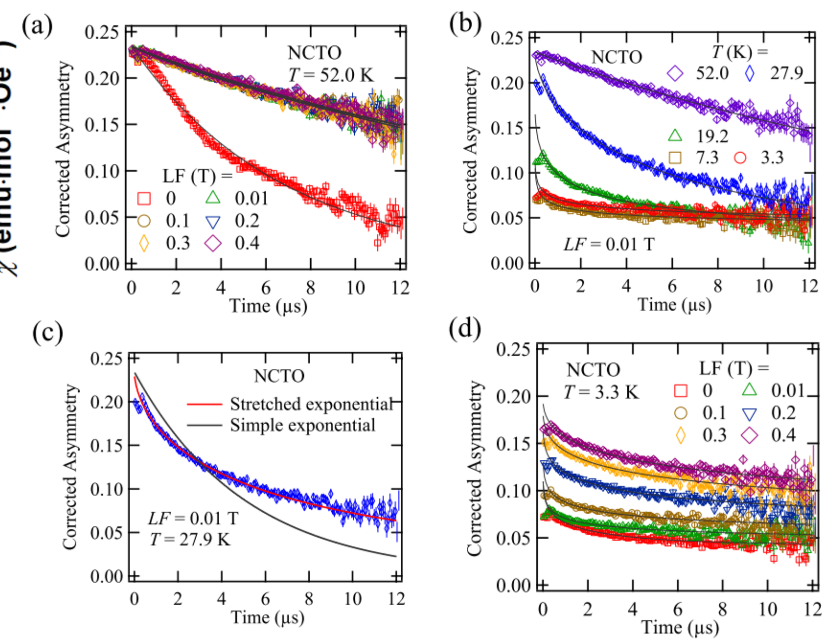


$$\mathbf{A}_{ZF}(t) = A_1 \cdot \left[\alpha \cos\left(\gamma_\mu B_{\text{int}} t + \varphi\right) \cdot e^{-\lambda_T t} + \left(1-\alpha\right) \cdot e^{-\lambda_L t} \right] + A_2 e^{-\lambda_{tail} t} \cdot G_{KT}$$

- ZF-μSR : very fast depolarization and superimposed oscillations within 50 ns below T_N, strong quasistatic internal field;
- λ_T reflects the width of static magnetic field distribution.
- T_F and T* present anomalies.

μSR by GPS at PSI





arXiv:2307.16451v1 (2023)

- V_{mag}(0K) = 60 % suggests the bond-dependent anisotropic frustrations (Kitaev interactions) and quantum fluctuations;
- Strong spin dynamics at low temperature.

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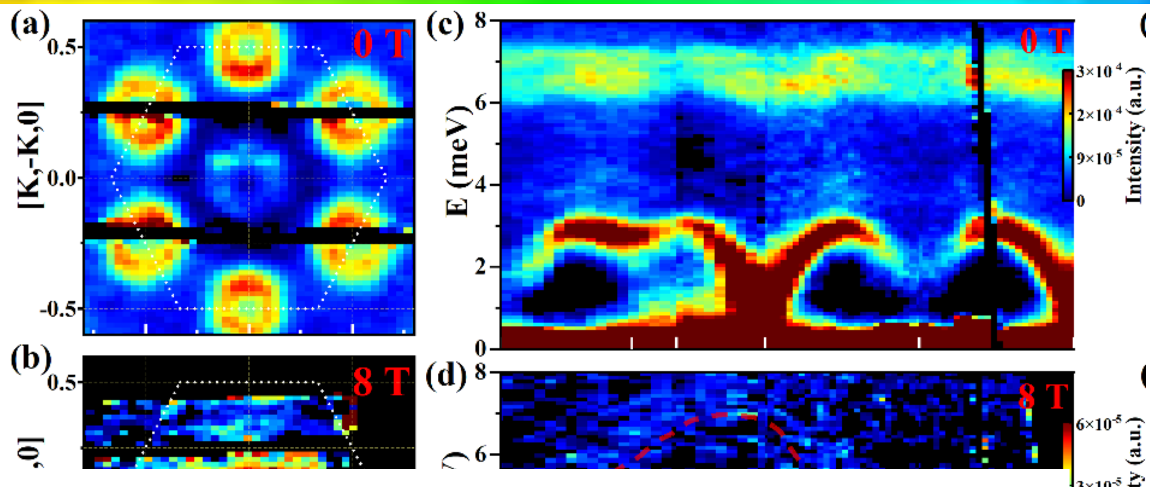
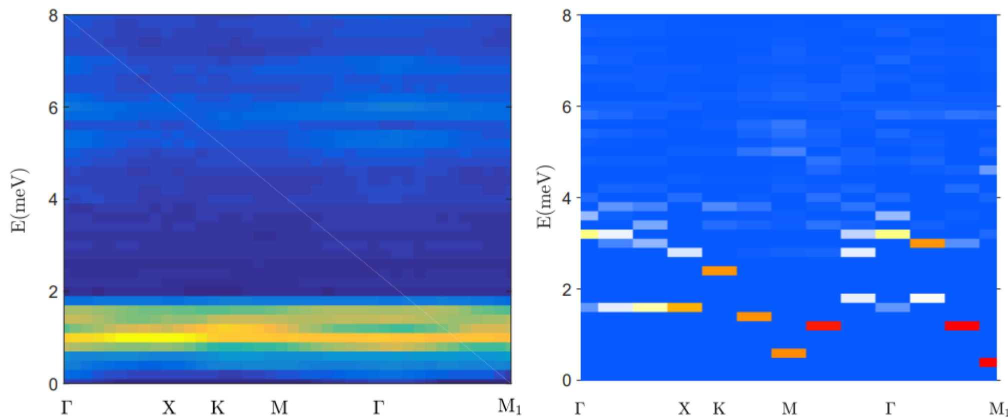
Inelastic Neutron Scattering





(a)$S(\boldsymbol{k},\omega)$ from Mean field

(b)$S^{zz}(\boldsymbol{k},\omega)$ from VMC

QSL mean-fi ... ld to enforce the ... aking

$$H_{\rm mf}^{\rm QSL} = \sum_{\langle i,j\rangle\in\gamma}\Big[i\rho_a {\rm Tr}(\psi_i^\dagger\psi_j \cdots$$

$$+i\rho_c{\rm Tr}(\psi_i^\dagger\psi_j+\tau^\gamma\psi_i^\dagger\sigma^\gamma\psi_j-\tau^\alpha\psi_i^\dagger\sigma^\alpha\psi_j-\tau^\beta\psi_i^\dagger\sigma^\beta\psi_j)+i\rho_d{\rm Tr}(\tau^\alpha\psi_i^\dagger\sigma^\beta\psi_j+\tau^\beta\psi_i^\dagger\sigma^\alpha\psi_j)$$

$$+i\rho_f{\rm Tr}(\tau^\alpha\psi_i^\dagger\sigma^\gamma\psi_j+\tau^\gamma\psi_i^\dagger\sigma^\alpha\psi_j+\tau^\beta\psi_i^\dagger\sigma^\gamma\psi_j+\tau^\gamma\psi_i^\dagger\sigma^\beta\psi_j)+{\rm H.c.}\Big]$$

$$+\sum_{\langle\langle\langle i,j\rangle\rangle\rangle}\Big[t_3{\rm Tr}(\tau^z\psi_i^\dagger\psi_j)+\Delta_3{\rm Tr}(\tau^x\psi_i^\dagger\psi_j)+{\rm H.c.}\Big]+\sum_i\boldsymbol{\lambda}_i\cdot{\rm Tr}(\psi_i\boldsymbol{\tau}\psi_i^\dagger)$$

$$\boldsymbol{M}_i = M\Big(\sin\phi\,[\hat{\boldsymbol{e}}_x\cos(\boldsymbol{Q}\cdot\boldsymbol{r}_i)+\hat{\boldsymbol{e}}_y\sin(\boldsymbol{Q}\cdot\boldsymbol{r}_i)]+\cos\phi\,\hat{\boldsymbol{e}}_z\Big),$$

$$H_{\rm mf}^{\rm total} = H_{\rm mf}^{\rm QSL}+\frac{1}{2}\sum_i(\boldsymbol{M}_i+\tilde{g}\mu_B\tilde{\boldsymbol{B}})\cdot{\rm Tr}(\psi_i^\dagger\frac{\boldsymbol{\sigma}}{2}\psi_i),$$

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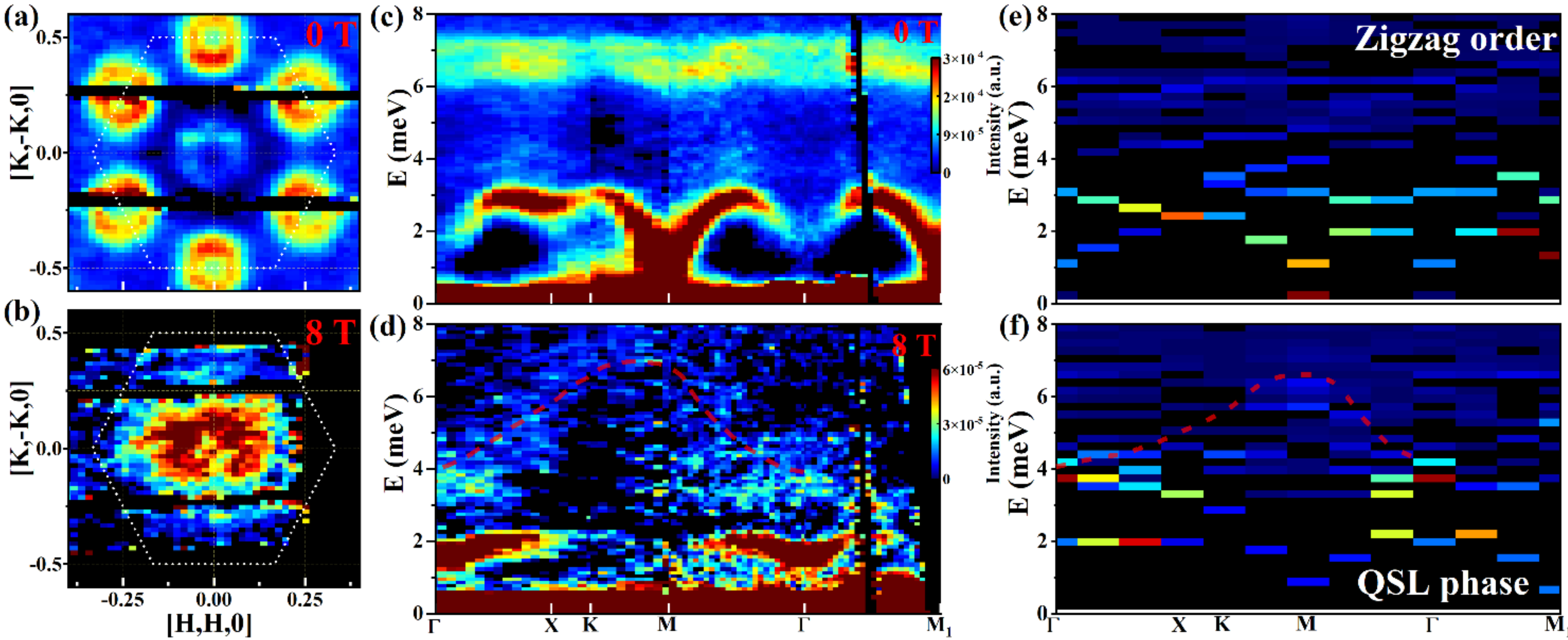
Inelastic Neutron Scattering



J_1 = −1.54 meV, J_3 = 1.32 meV, K = 1.408 meV, Γ = −1.32 meV, and Γ' = 0.88 meV,

J_1 = 0.066 meV, J_3 = 1.32 meV, K = −3.399 meV, Γ = 0.286 meV, and Γ' = 0.077 meV.

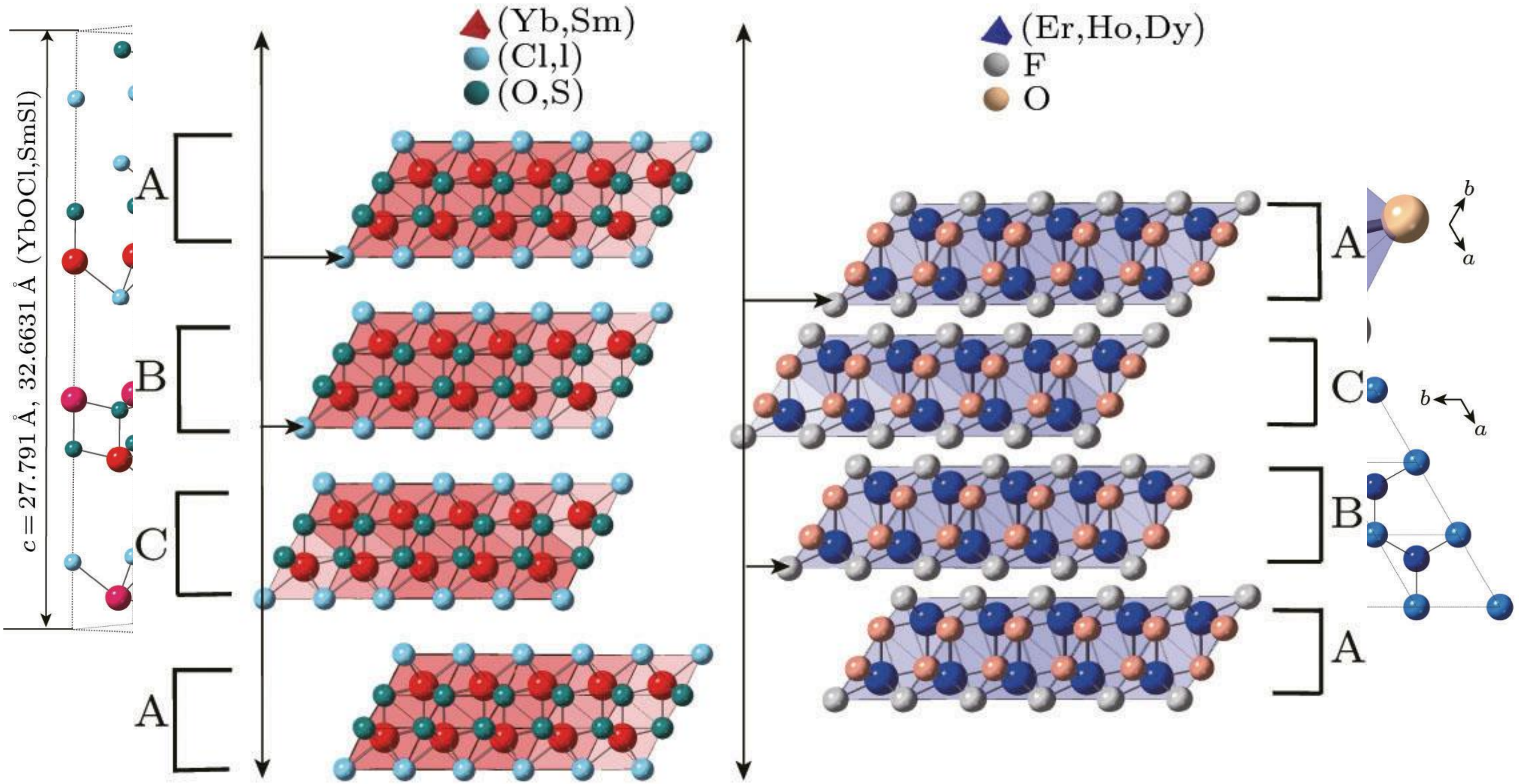
Conclusions

- Quantum effect in 2D triangular/honeycomb lattices could very strong
- New physical properties could be introduced with the complicated interactions of the quantum fluctuation and spin/orbital
- Spinons of RVB? or Vortex fermion excitations?
- muSR has a strong contribution to the proof of QSL

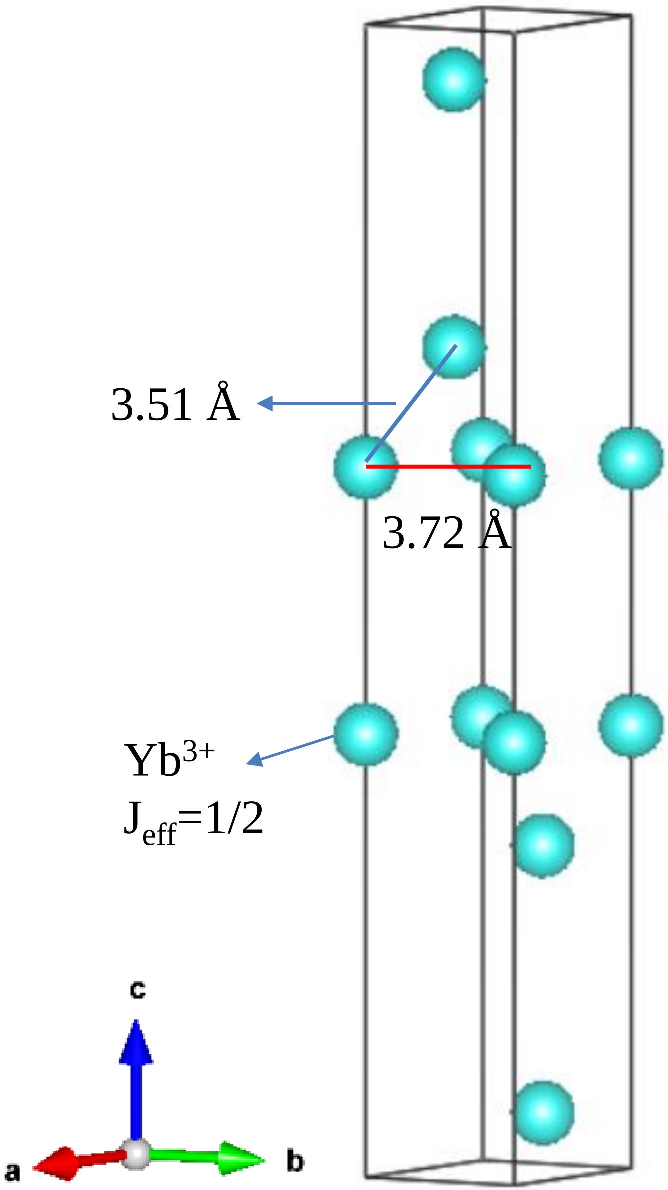
Quantum Spin Liquid Candidate

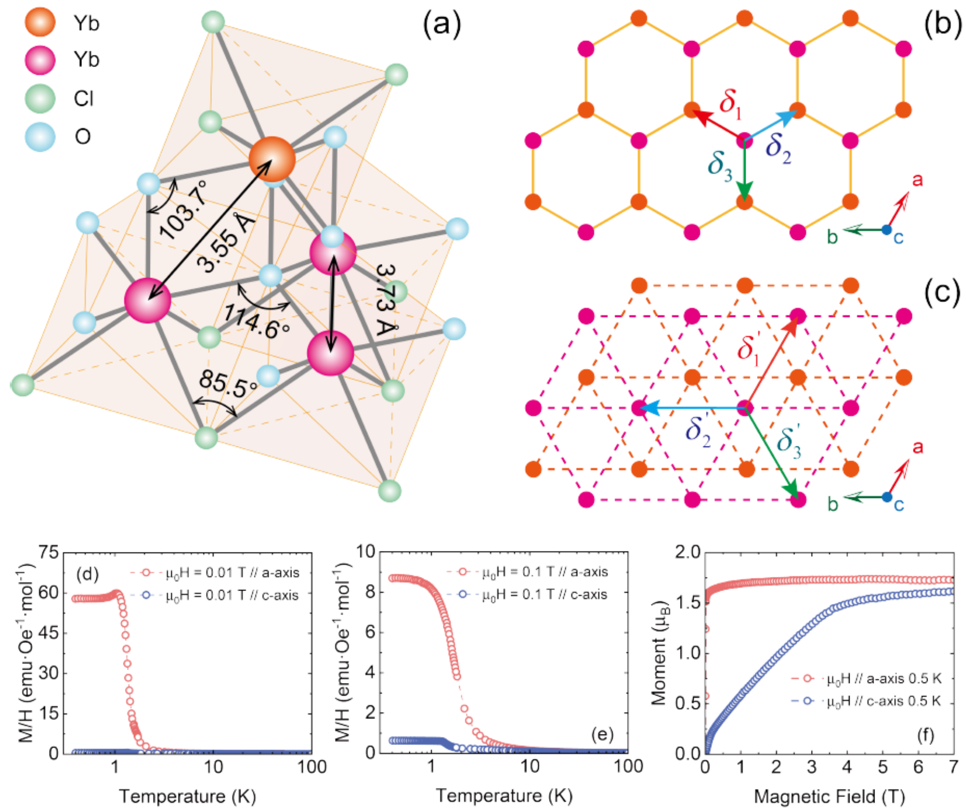
- REChX (RE = rare earth; Ch = O, S, Se, Te; X = F, Cl, Br, I)

Chin. Phys. Lett. 38， 047502 (2021)

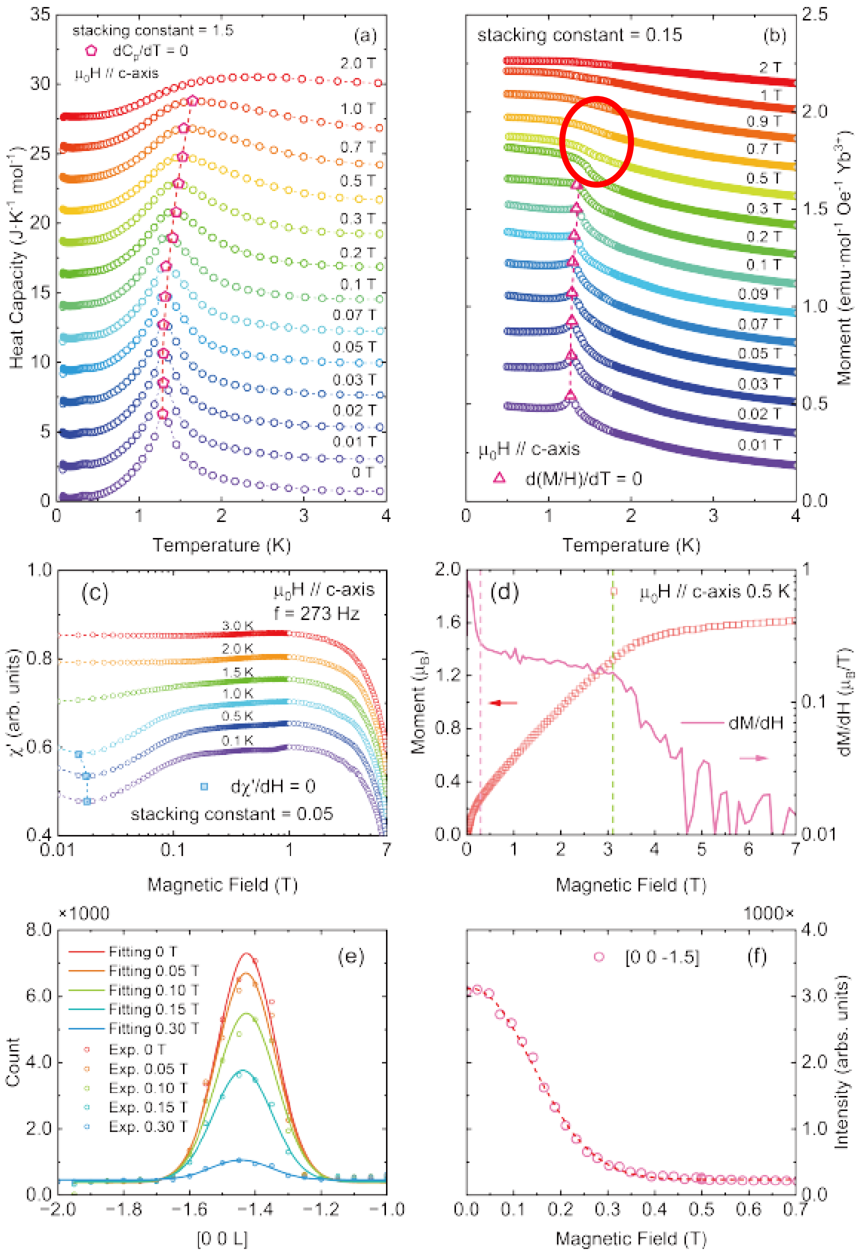


YbOCl





1. Triangular lattice in a single layer and the neighbor layers form a stacked honeycomb lattice.
2. An AFM transition was detected at about 1.3 K.
3. Strong anisotropy in M-H curves.



Thermodynamic measurements of YbOCl

$$H = H_{Honeycomb} + H_{Triangular} + H_{Zeeman} \quad (1)$$

$$\hat{H}_{Honeycomb} = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z + J_{\pm}\left(S_i^+ S_j^- + S_i^- S_j^+\right) + J_{\pm\pm}\left(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-\right) + J_{z\pm}\left(\gamma_{ij} S_i^+ S_j^z + \gamma_{ij}^* S_i^- S_j^z + \langle i \leftrightarrow j \rangle\right) \quad (2)$$

$$\hat{H}_{Triangular} = \sum_{\langle\langle ik \rangle\rangle} J'_{zz} S_i^z S_k^z + J'_{\pm}\left(S_i^+ S_k^- + S_i^- S_k^+\right) + J'_{\pm\pm}\left(\gamma'_{ik} S_i^+ S_k^+ + \gamma'^*_{ik} S_i^- S_k^-\right) - \frac{iJ'_{z\pm}}{2}\left(\gamma'^*_{ik} S_i^+ S_k^z + \gamma'_{ik} S_i^- S_k^z + \langle i \leftrightarrow k \rangle\right) \quad (3)$$

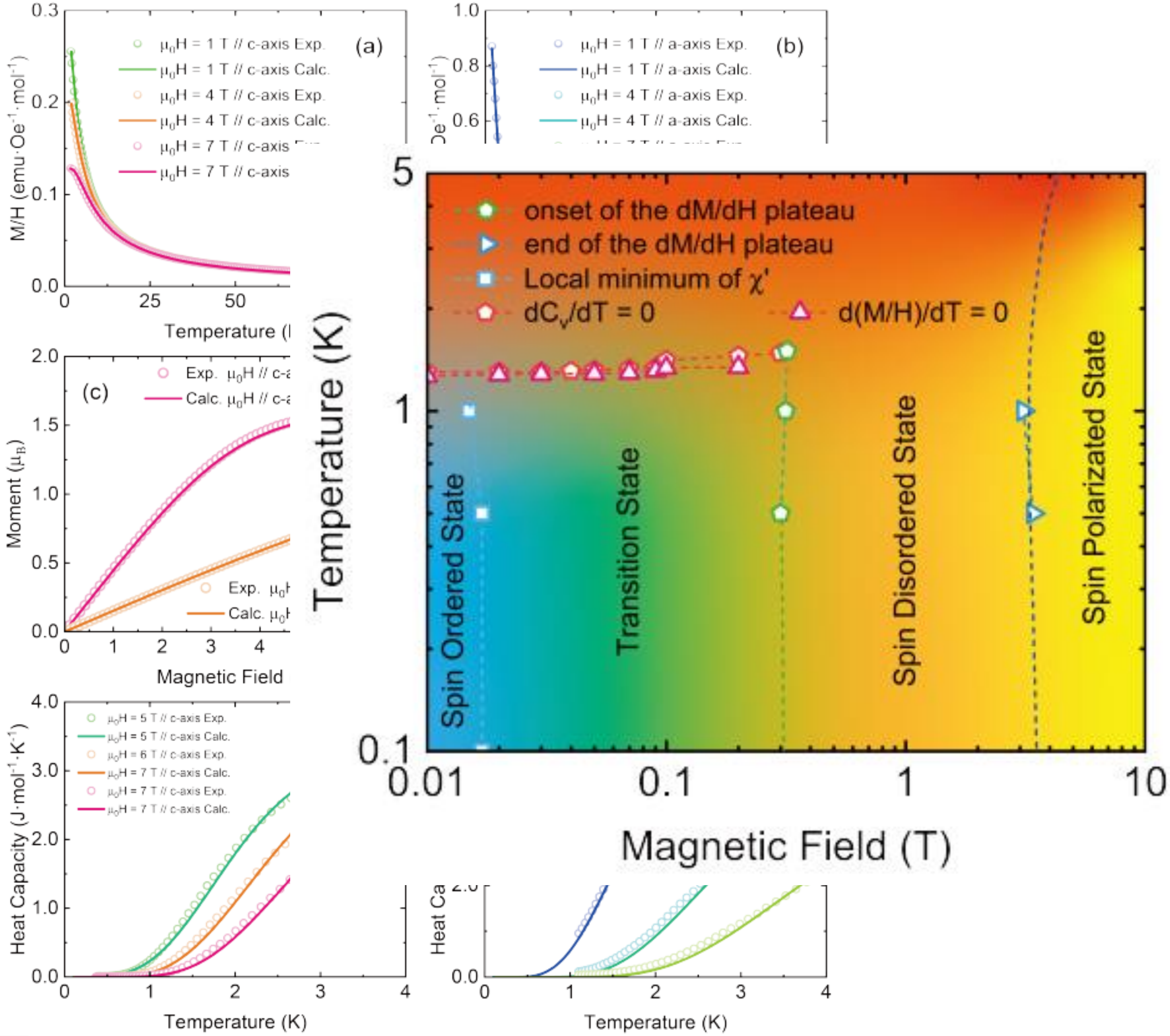
$$H_{zeeman} = -\mu_0 \mu_B \sum_i g_{ab}(h_x S_i^x + h_y S_i^y) + g_c h_c S_i^z \quad (4)$$

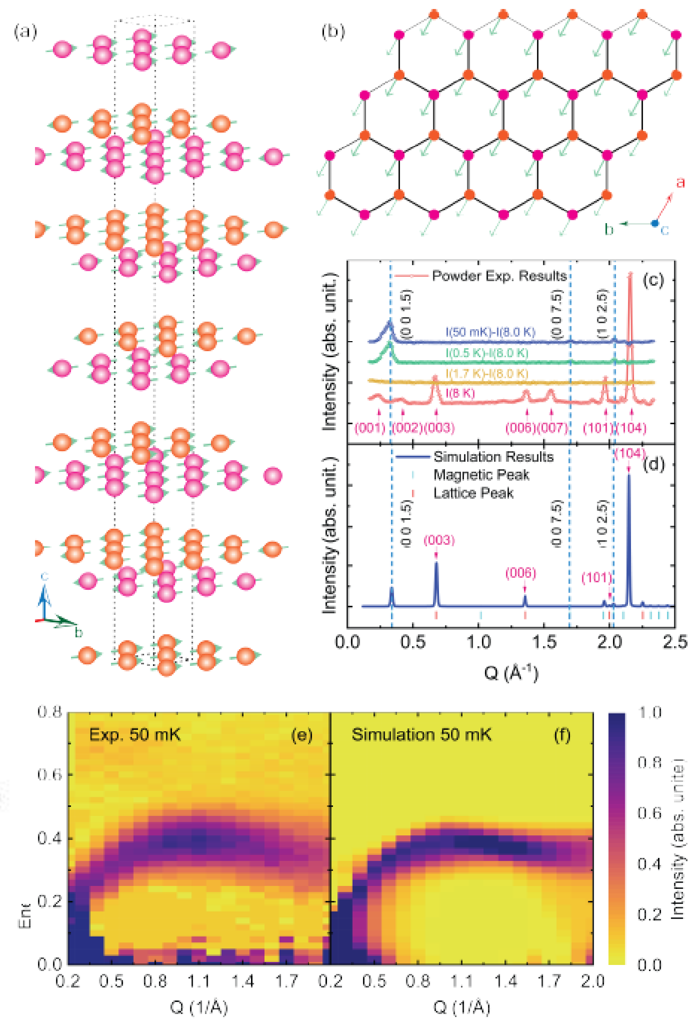
1. The AFM transition moves toward high temperature with increasing field.
2. A spin disordered state was detected.
3. The anisotropic spin Hamiltonian was constructed.

FD simulation results of YbOCl

Derived parameters

NPD and INS results





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R. Yu, Z. Liu, J. Park

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NCNR: N. Butch, Yiming Qiu

HZB: Z. L. Lu, J. Q. Xu, M. Russina, G. Günther

FRM-II, J-Parc, PSI, ANSTO, etc

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NMR* and *ESR:

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