International Workshop on Muon Physics at the Intensity and Precision Frontiers

μSR studies of spin dynamics in frustrated rare-earth magnets

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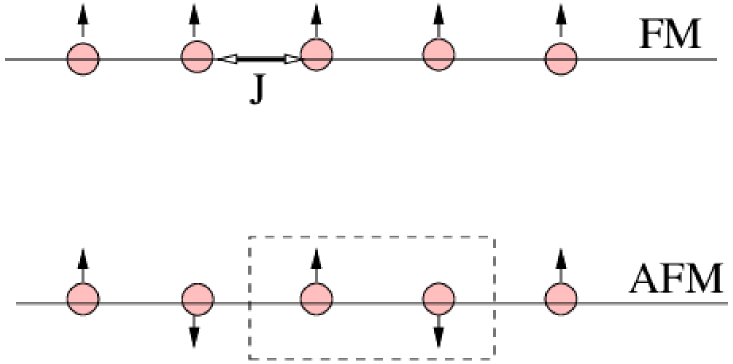
Huizhou

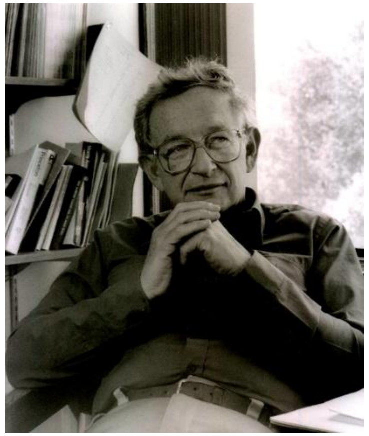
Outline

- ☐ Introduction: what is a quantum spin liquid, and the role of muSR
- ☐ QSL candidates hexaaluminates RE(Zn,Mg)$Al_{11}O_{19}$
- ☐ Proximity to a QSL in the Shastry-Sutherland magnet $Nd_2Be_2GeO_7$

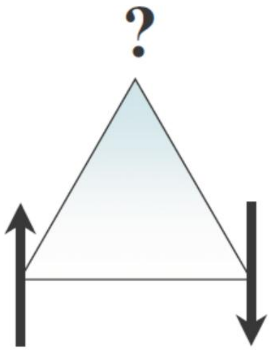
Quantum Spin Liquids

conventional spin ordering





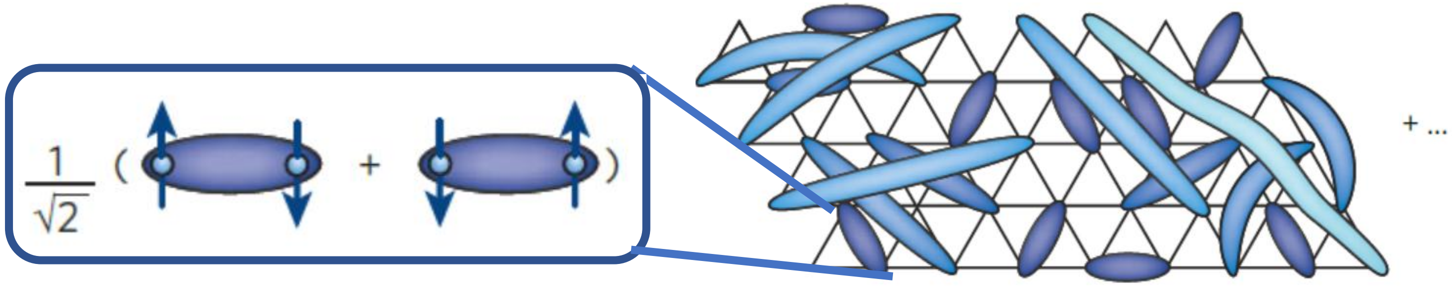
P. W. Anderson



geometric frustration

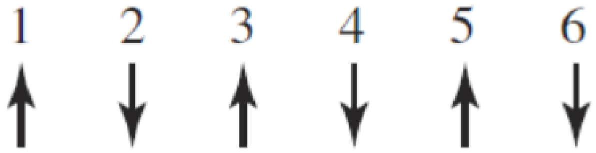
highly degenerate QSL

- ☐ No long-range magnetic ordering
- ☐ No spontaneous breaking of translation or rotation symmetry

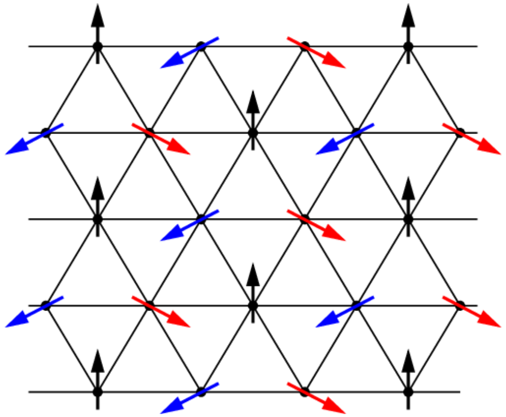


Quantum Spin Liquids

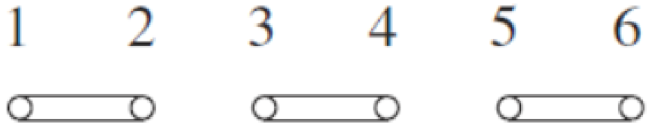
$$\mathcal{H} = J\sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1}$$



$$E = -\frac{1}{4}JN$$

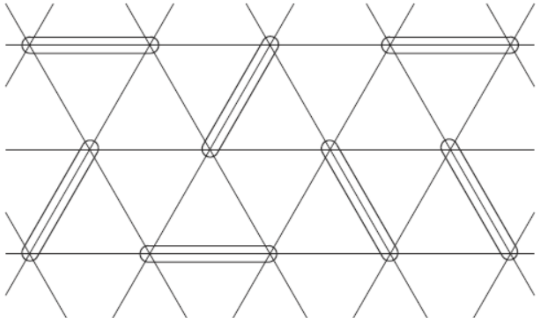


$$E = \frac{1}{4}J\cos\frac{2}{3}\pi \cdot \frac{1}{2}Nz = -\frac{3}{8}JN$$



$$\frac{1}{\sqrt{2}}(\uparrow\downarrow - \downarrow\uparrow)$$

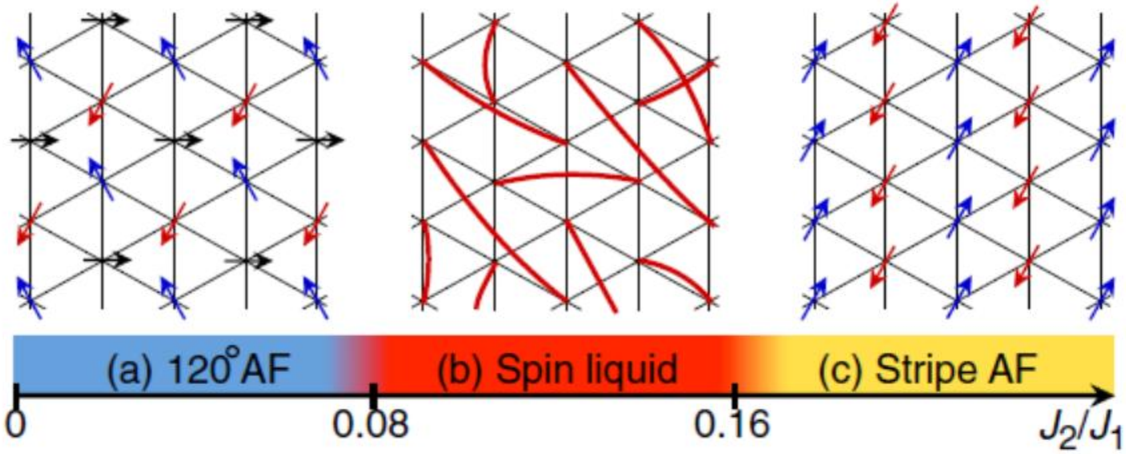
$$E = -\frac{3}{4}J \cdot \frac{N}{2} = -\frac{3}{8}JN$$



$$E = -\frac{3}{4}J \cdot \frac{1}{2}N = -\frac{3}{8}JN$$

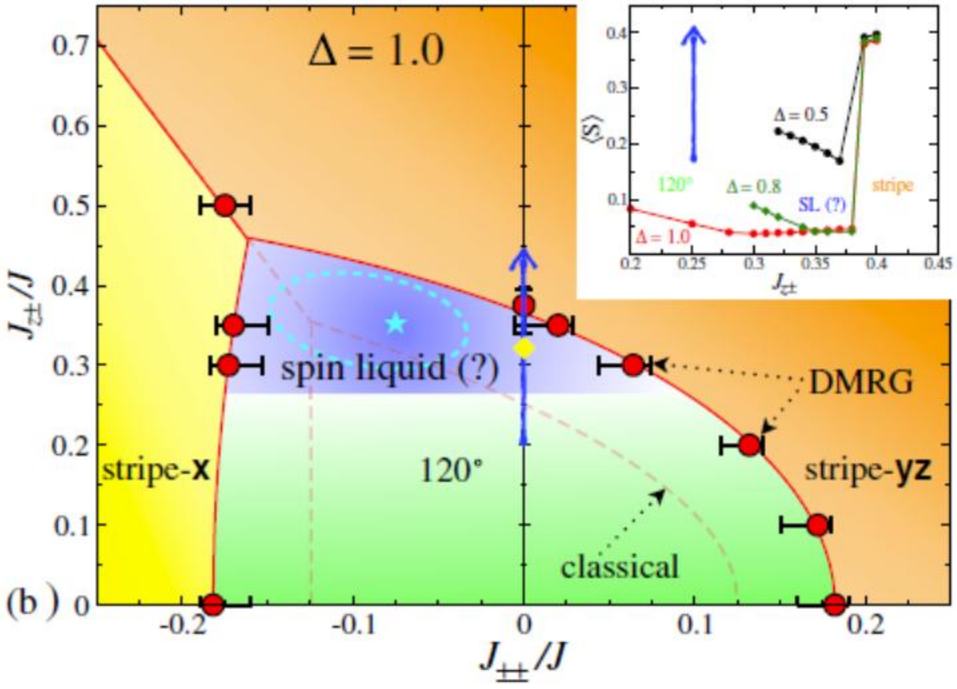
Quantum Spin Liquids

next nearest neighbor



Y. Iqbal et al., PRB, 93, 144411 (2016)

anisotropy

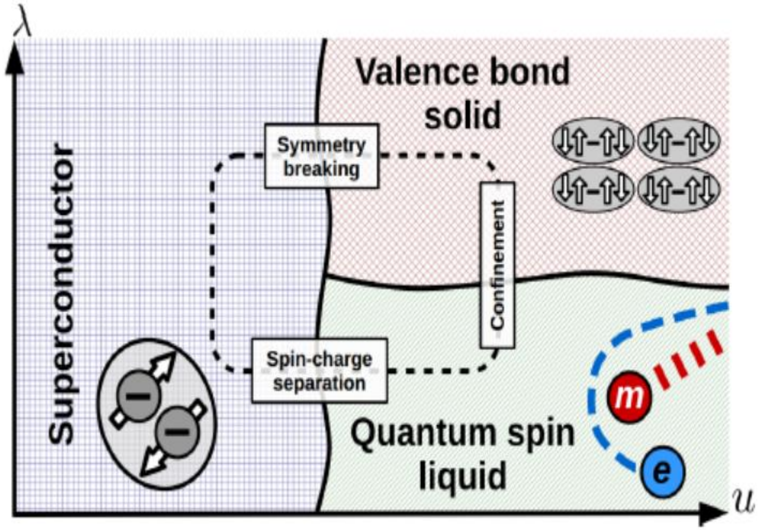


Z. Zhu et al., PRL, 120, 207203 (2018)

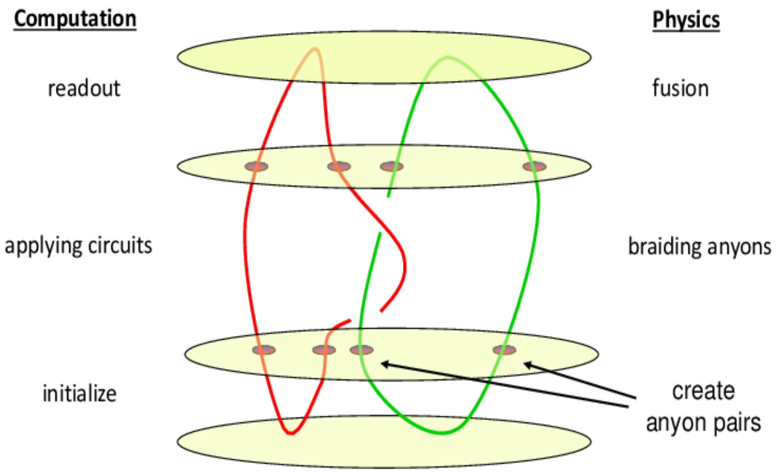
Quantum Spin Liquids

Introduction

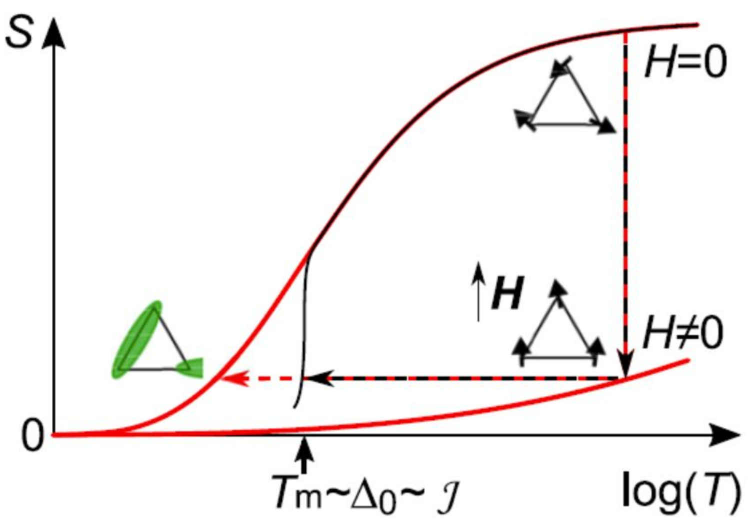
closely related SC



potential for quantum computation



adiabatic demagnetization refrigeration



Quantum Spin Liquids

How to detect? Or what to exclude?

- no order parameters

exclude any long-range magnetic ordering, spin freezing, valence bond solid

- spinon fractional excitations

→ excitation continua

(ordered magnets may show similar behavior)

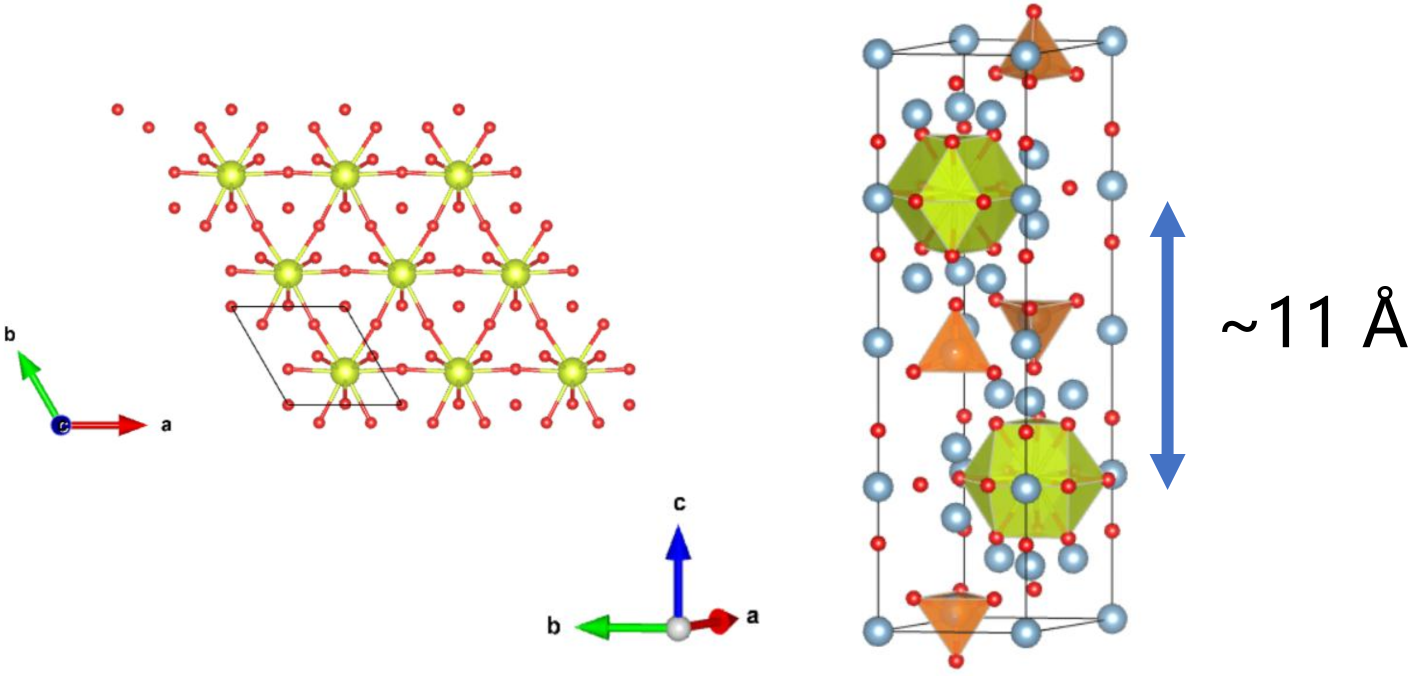
specific heat
susceptibility
neutron diffraction
……

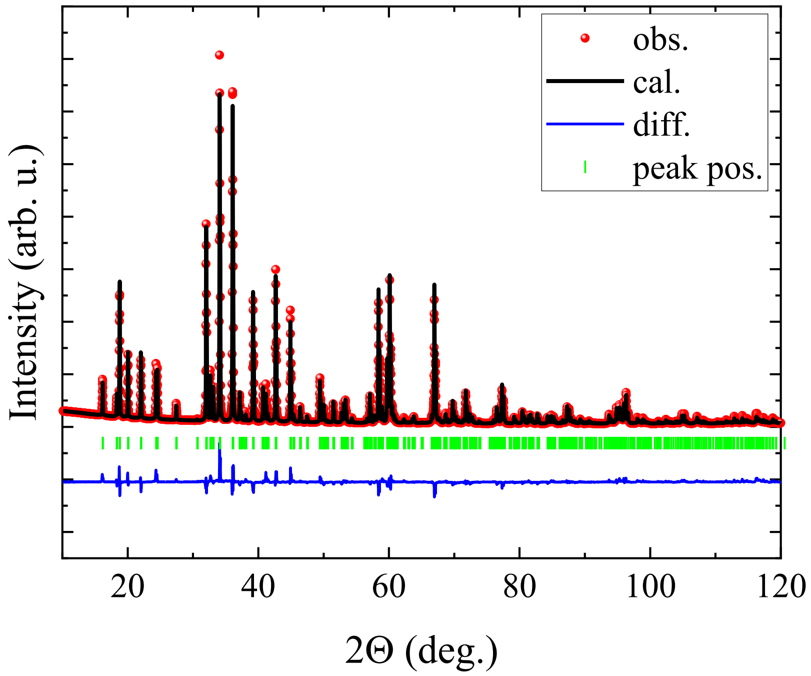
muon is very sensitive to magnetic fields!
distinguish between static and dynamic fields

inelastic neutron scattering
thermal transport

Hexaaluminates RE(Zn,Mg)$Al_{11}O_{19}$

general structural features





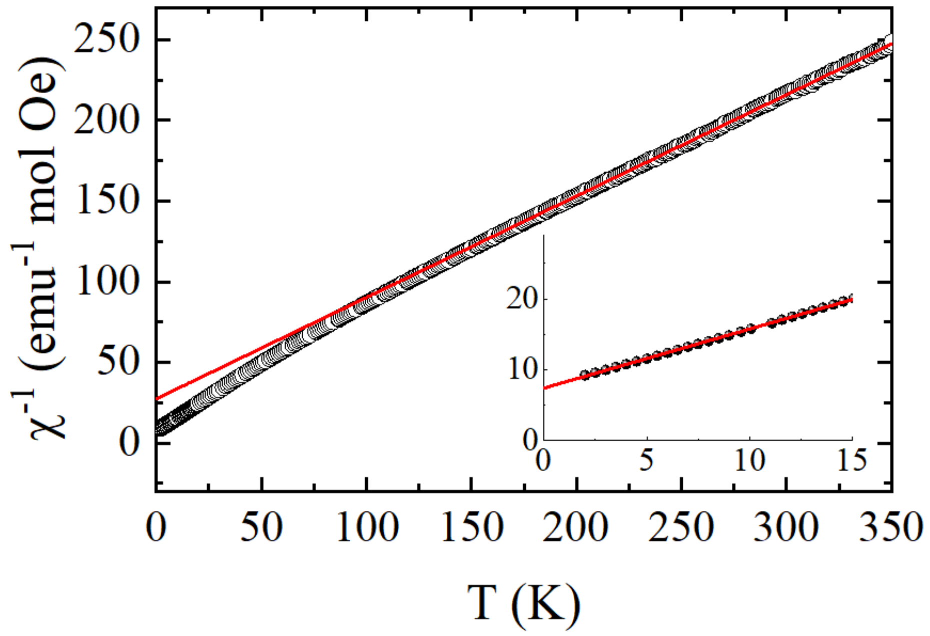
- well separated→ quasi-2D; minimize influence from interlayer disorder
- large differences in ionic radii→ no antisite disorder between magnetic and nonmagnetic ions
- versatile rare earth ions (RE = La-Tb)

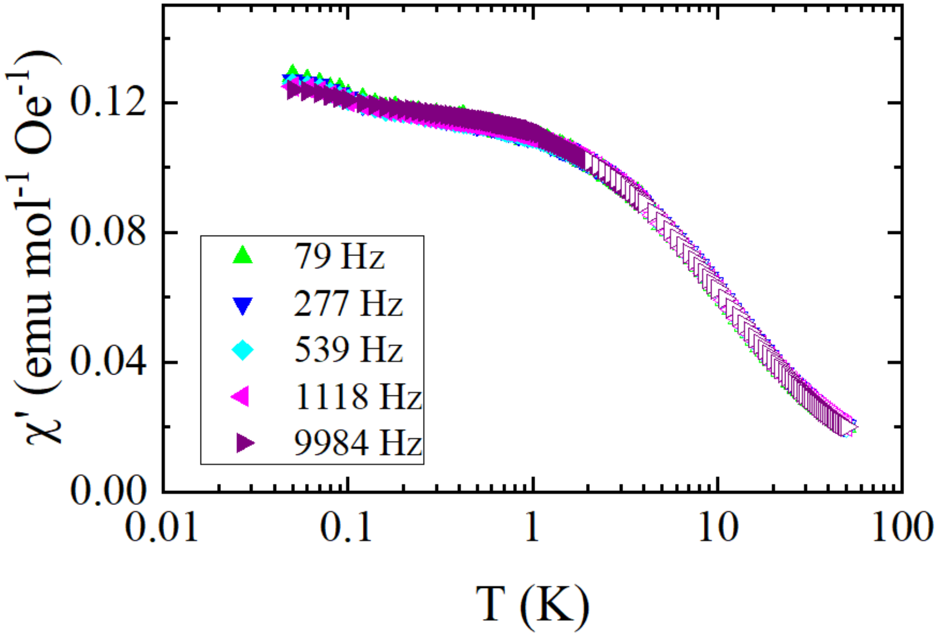
$P6_3/mmc$

a = b = 5.586026

c = 21.920176

$PrZnAl_{11}O_{19}$: susceptibility





dc susceptibility:

- Curie-Weiss fit (T < 15 K)

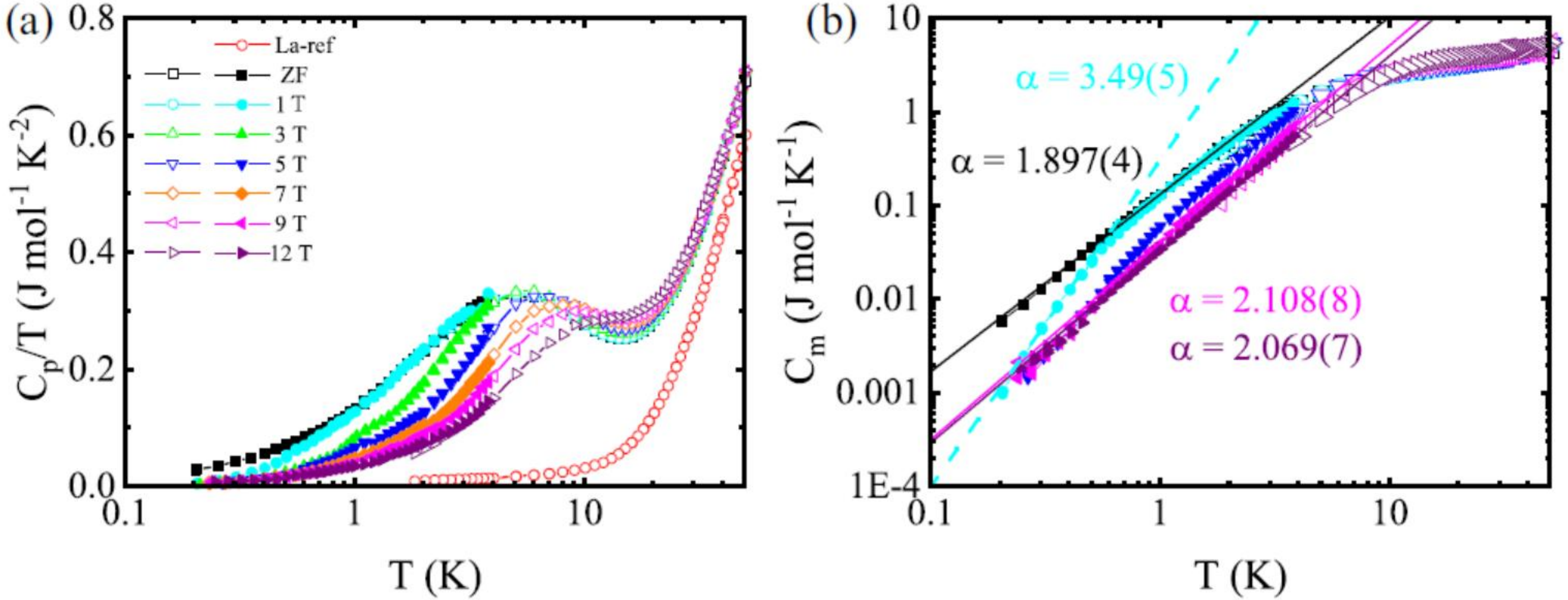
  θ_{CW} ~ -10 K → strong AFM coupling

ac susceptibility:

- no spin ordering down to 50 mK
- no frequency dependence → no spin freezing
- frustration index: *f* > 10/0.05 = 200

Bu, Guo et al., PRB 106, 134428 (2022)

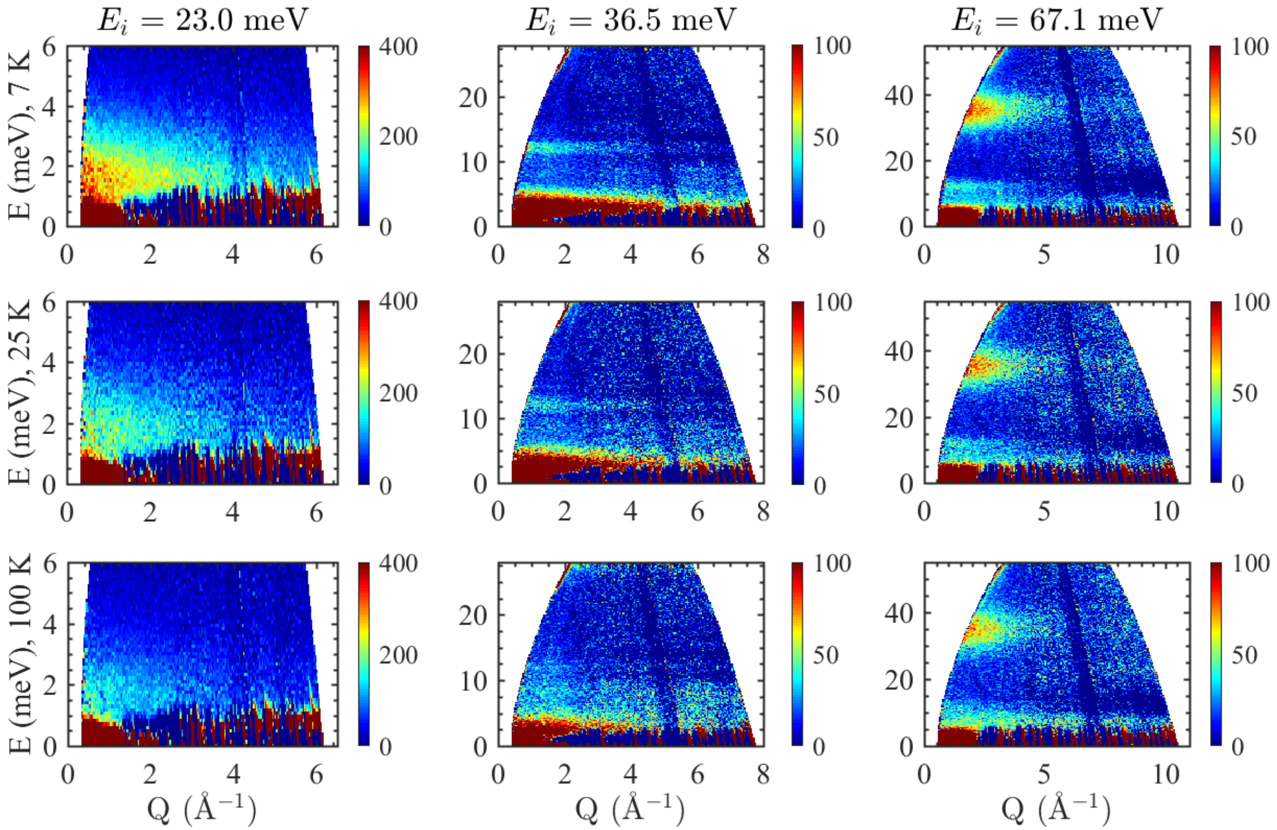
$PrZnAl_{11}O_{19}$: gapless excitations

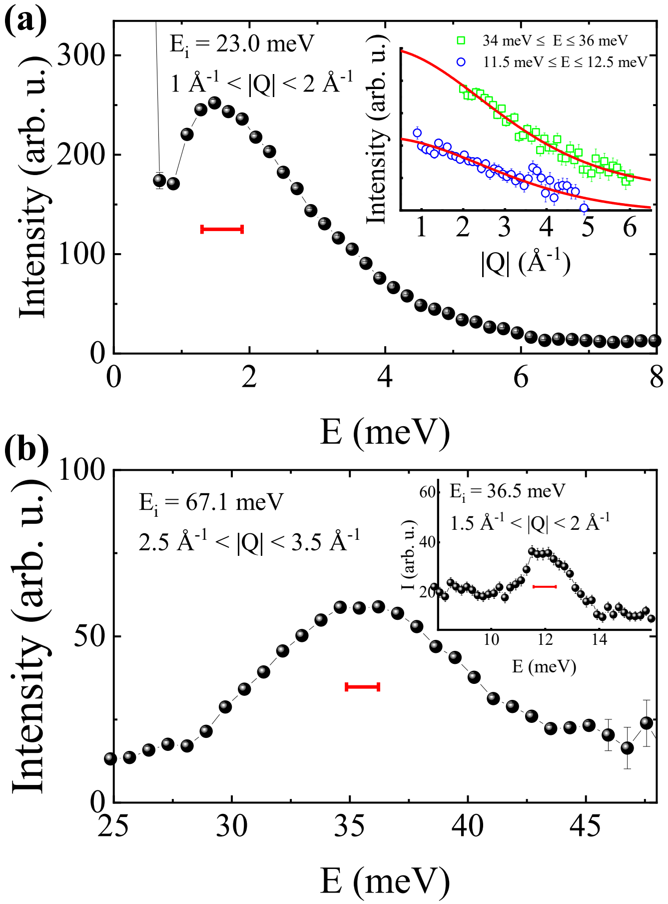


- ➢ $C_m(T) \propto T^{\alpha}$ → gapless excitation
- ➢ α ~ 2 (ZF)

gapless AFM: α = 3
gapless FM: α = 3/2
spinon Fermi surface: α = 2/3 or 1
Coulombic QSL: α = 3
Dirac QSL: α = 2 (ZF), α = 1 (fields)

$PrZnAl_{11}O_{19}$: low-E excitations

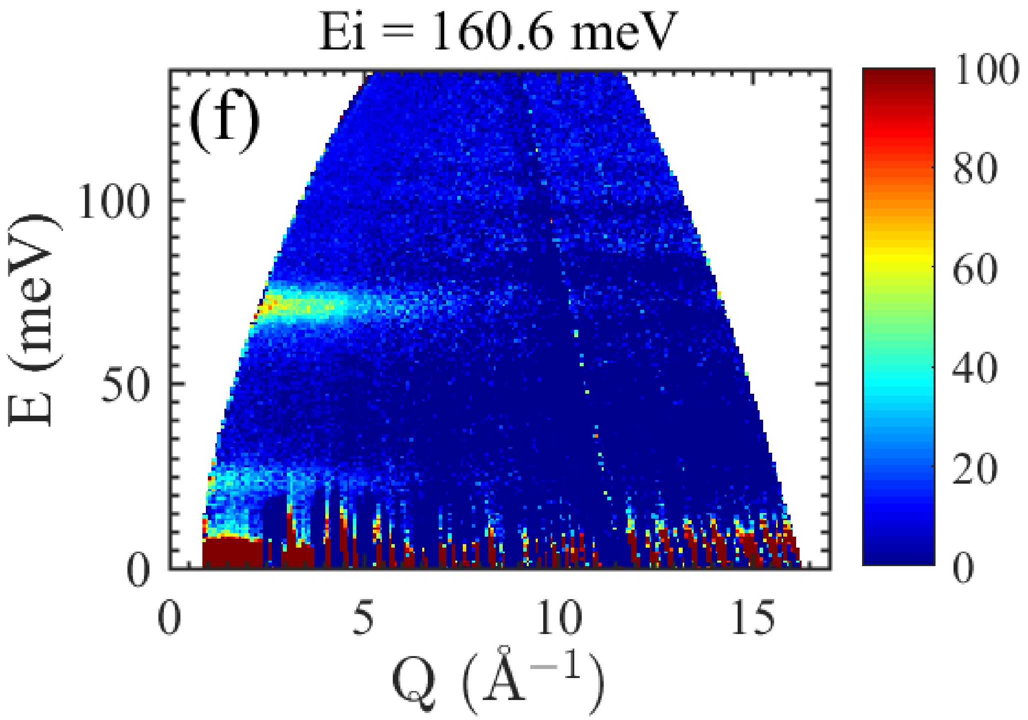


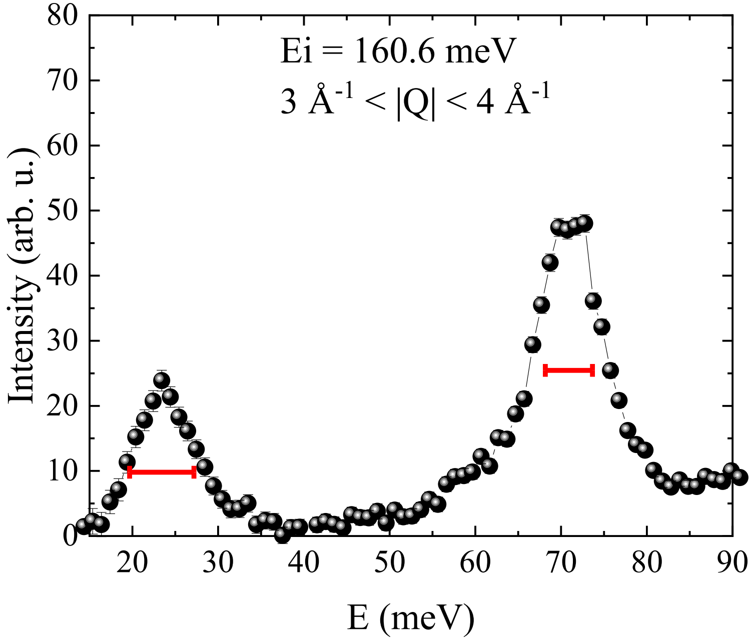


- Pr^{3+} (J = 4): non-Kramers ion
- D3h → 3 singlets, 3 doublets
- peak overlaps at 12 meV and 36meV

- E ~ 1.5 meV, diffuse, long tail on the high-E side
- distinct from Schottky anomaly in HC → correlation effect

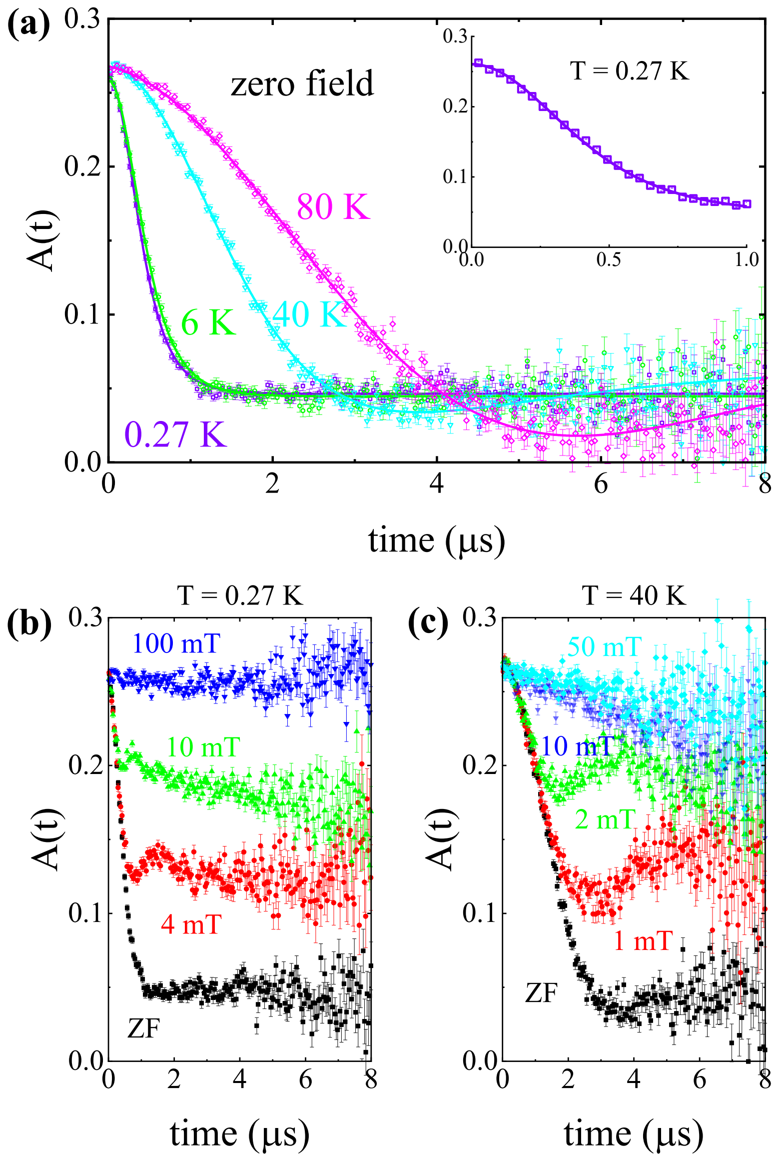
$NdZnAl_{11}O_{19}$: low-E excitations





☐ $NdZnAl_{11}O_{19}$: peak width comparable to instrument resolution ⇒ exclude disorder-induced peak broadening in $PrZnAl_{11}O_{19}$

$PrZnAl_{11}O_{19}$: enhanced nuclear magnetism



☐ ZF-μSR spectra: Gaussian-broaden-Gaussian

$$GbG(t) = f + (1-f)\left(\frac{1}{1+R^2\Delta_0^2t^2}\right)^{\frac{3}{2}}$$

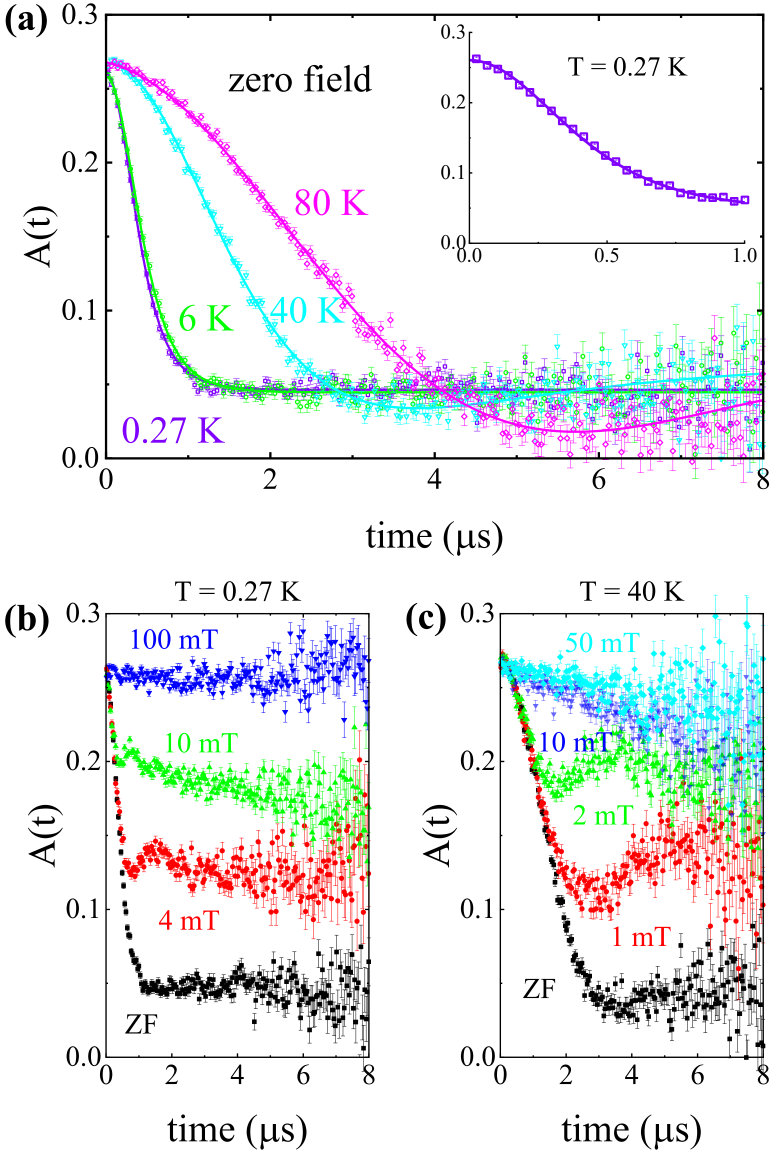
$$\times\left(1-\frac{\Delta_0^2t^2}{1+R^2\Delta_0^2t^2}\right)\exp[-\frac{\Delta_0^2t^2}{2(1+R^2\Delta_0^2t^2)}]$$

$$\mathrm{KT}^G(t) = \frac{1}{3}+\frac{2}{3}(1-\Delta^2t^2)\exp\left(-\frac{\Delta^2t^2}{2}\right)$$

Δ has a Gaussian distribution width W

☐ both ZF and LF data indicate a static ground state

$PrZnAl_{11}O_{19}$: enhanced nuclear magnetism



Two ingredients:

1. non-Kramers ion, singlet ground state → van Vleck paramagnetism
2. large hyperfine interaction

$$\mathcal{H}_{HF} = A_J \overset{\text{nuclear spin}}{I} \cdot \overset{\text{electronic angular momentum}}{J} = g_J \mu_B B_z J_z$$

$$B_z = A_J I_z / g_J \mu_B$$

$$m_z = \chi_{VV} B_z$$

Singlet ground state for the electronic system

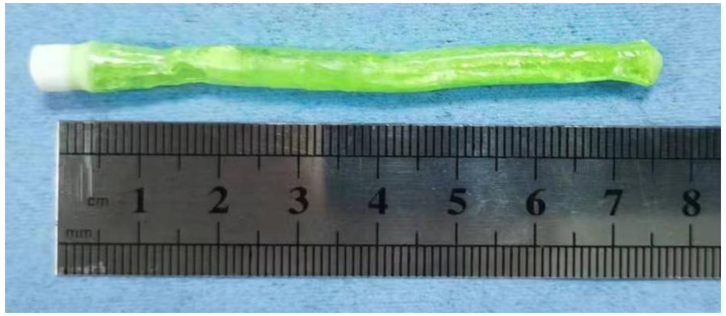
- ➢ intrinsic CEF ground state
- ➢ local distortions induced by implanted muons

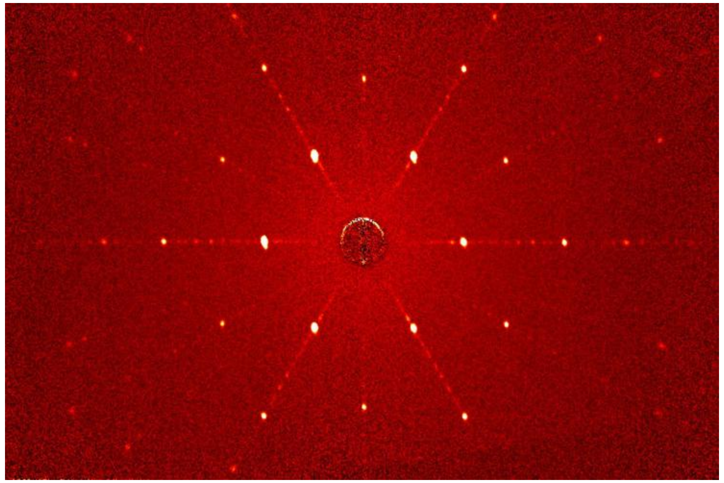
⇩

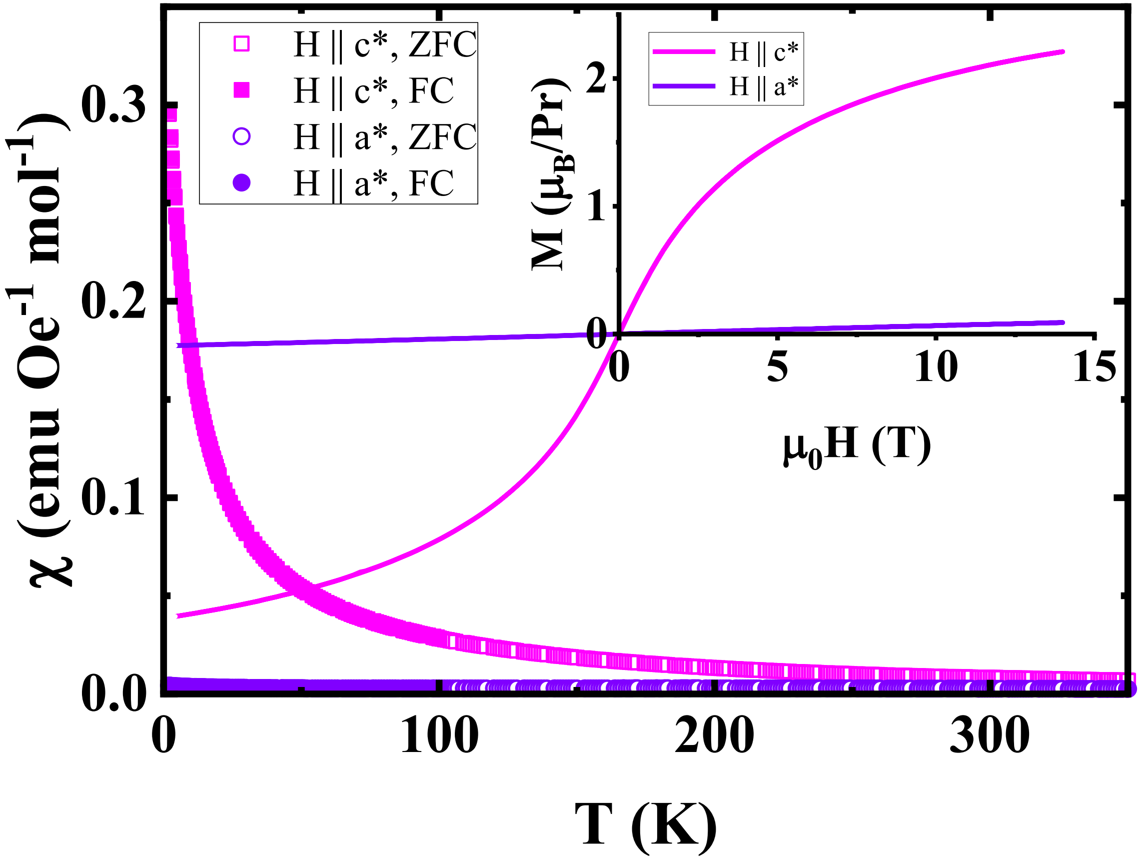
verified by following ESR measurements

$PrMgAl_{11}O_{19}$: Ising anisotropy

Hexaaluminate

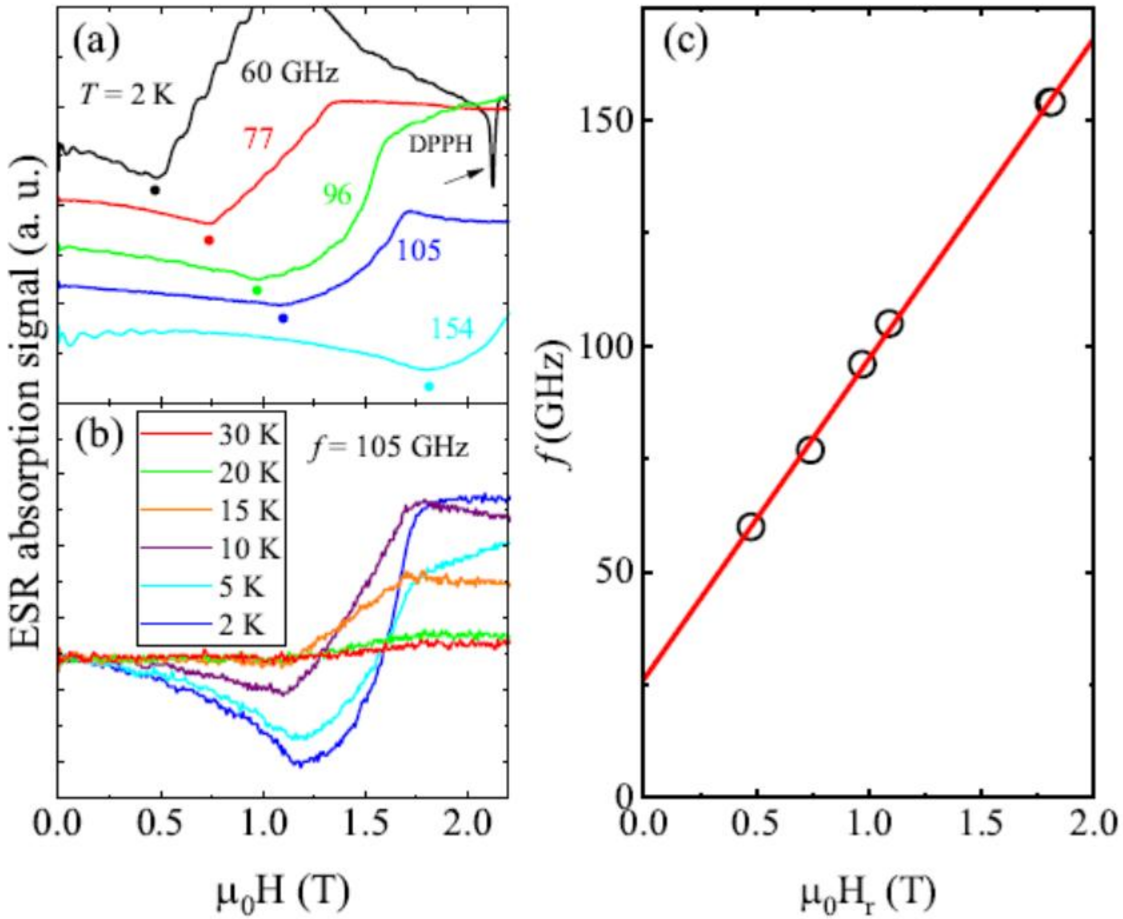


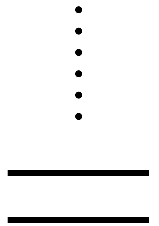




Cao, Guo et al., Mater. Futures 3, 035201 (2024)

Intrinsic transverse field Ising model





$\Delta \sim 0.1$ meV

In the frame work of intrinsic transverse-field Ising model

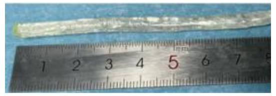
$$\mathcal{H} = \sum_{i,j} J S_i^z S_j^z - h \sum_i S_i^y$$

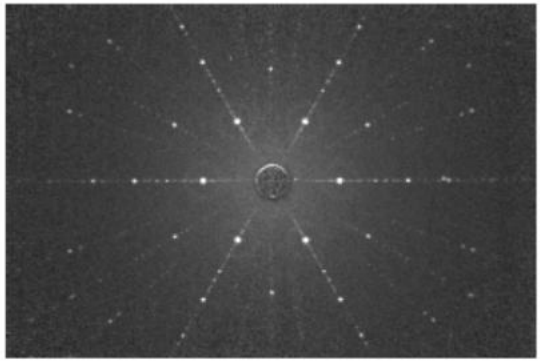
$$\Theta S_x \Theta^{-1} = S_x;\ \Theta S_y \Theta^{-1} = S_y;\ \Theta S_z \Theta^{-1} = -S_z$$

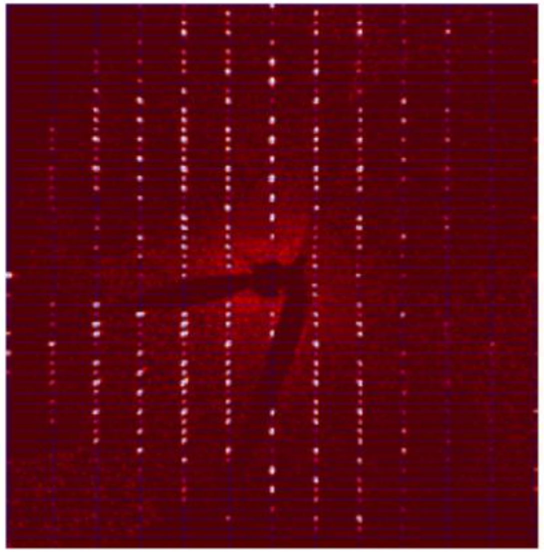
S_z: dipole

S_x, S_y: multipole

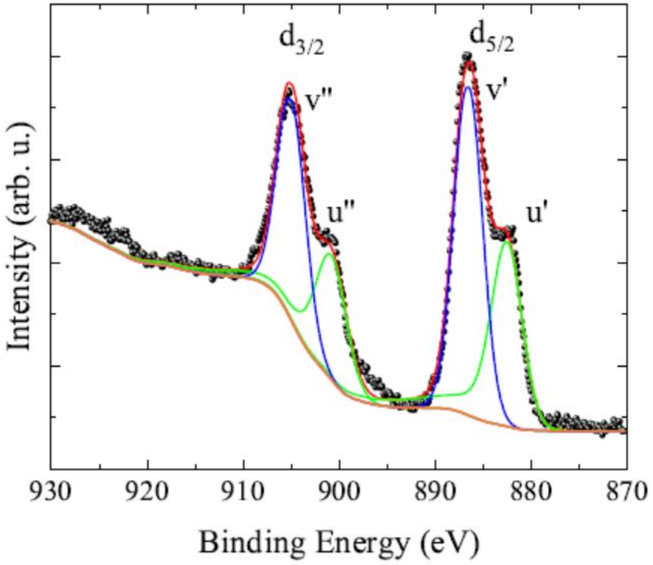
$CeMgAl_{11}O_{19}$: U(1) Dirac QSL candidate

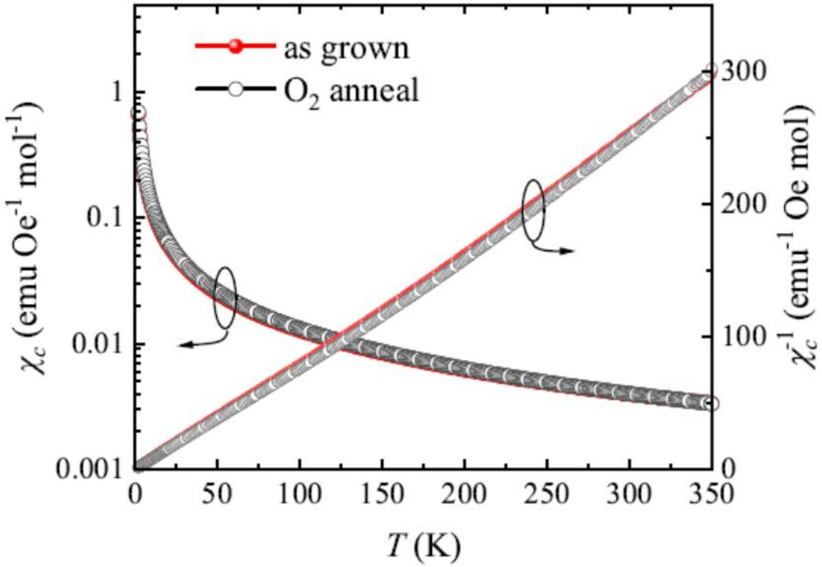


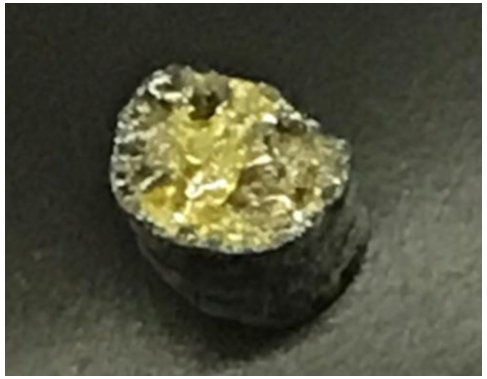




Very stable Ce^{3+} state







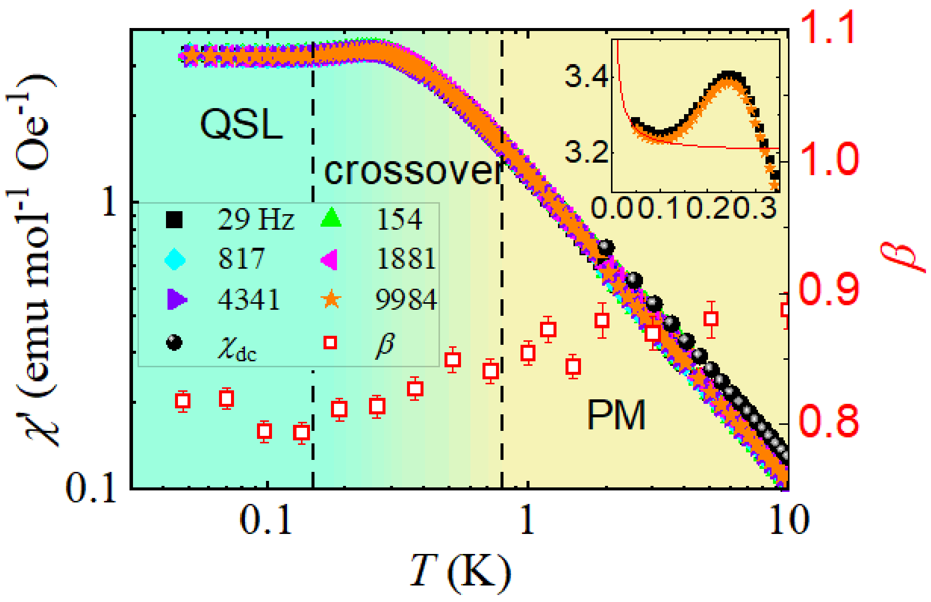
Ce2Zr2O7

oxidized Ce^{4+} on the surface

PRL 122, 187201 (2019)

CeMgAl$_{11}$O$_{19}$: U(1) Dirac QSL candidate

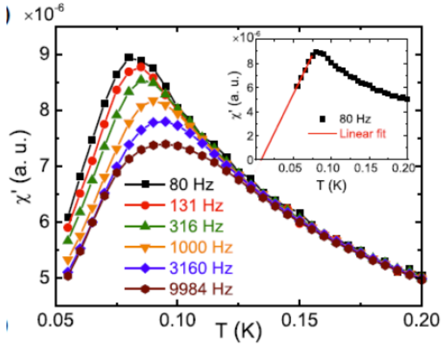
Hexaaluminate



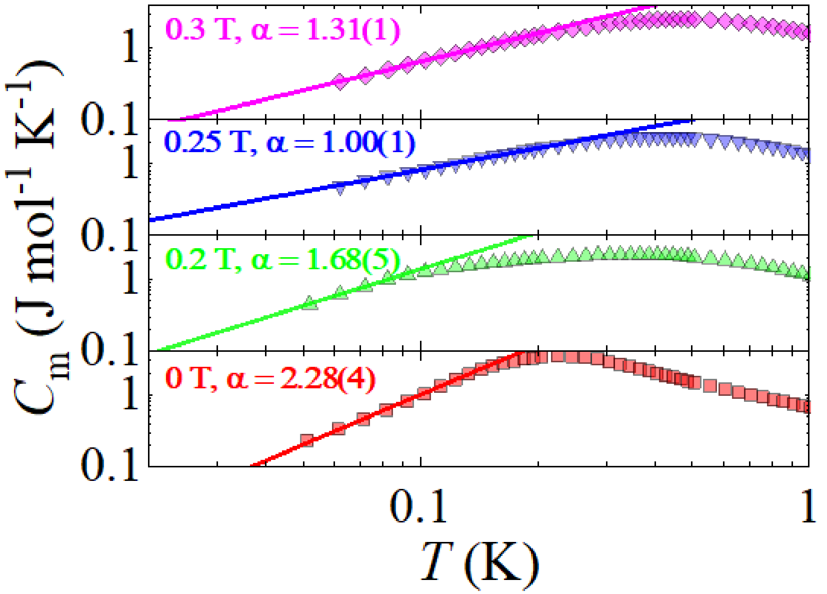
ac susceptibility:

magnetic correlations below ~0.2 K

no spin freezing



YbZn2GaO5
PRL 133, 266703 (2024)



specific heat:

gapless excitations $C_m = AT^{\alpha}$

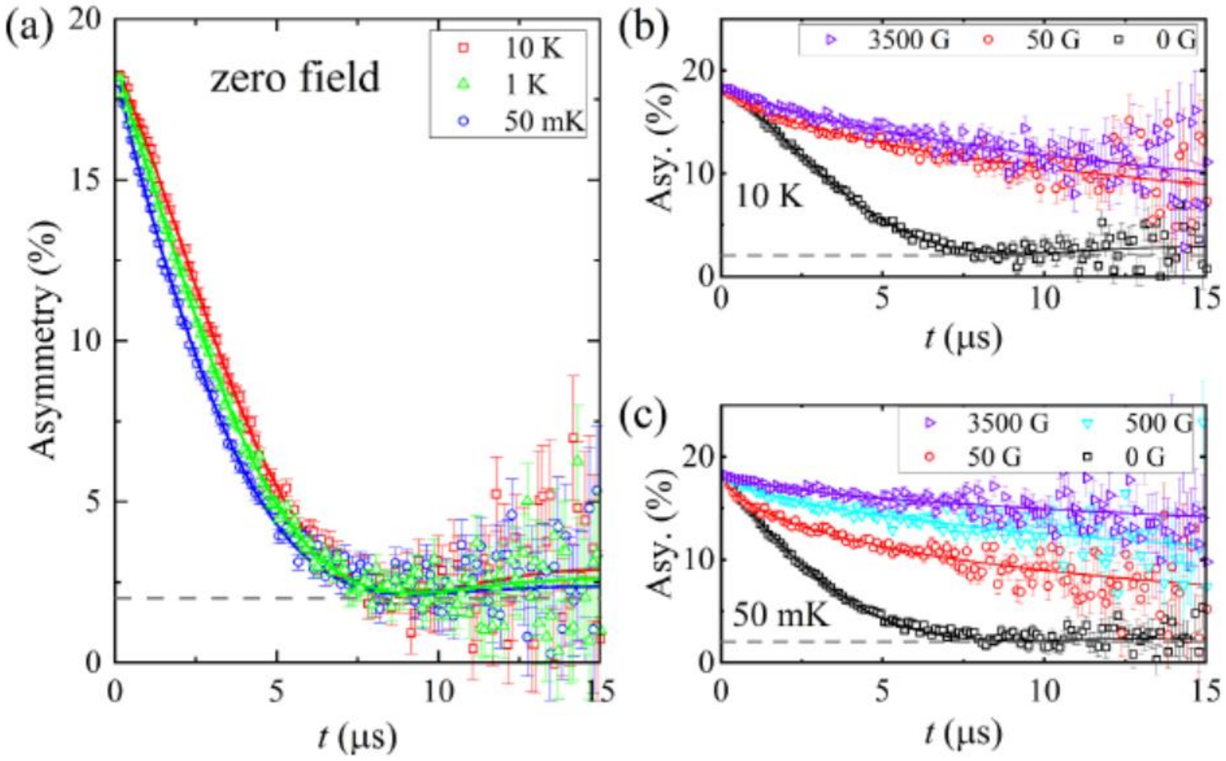
$\alpha = 2$, Dirac nodes

$\alpha = 1$, Fermi pocket

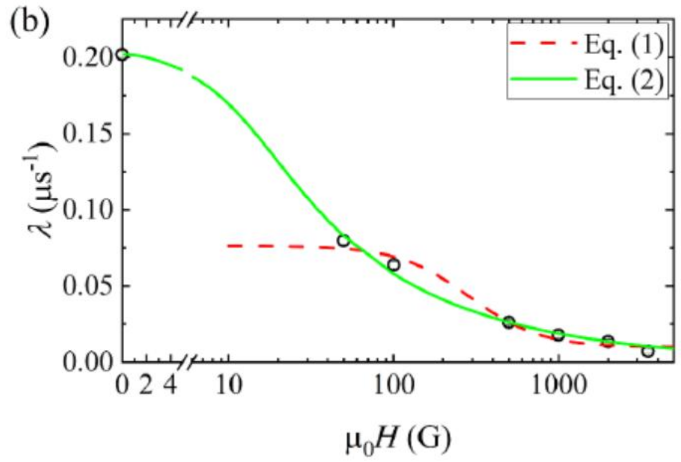
A(0.25 T)/A(0 T) =
0.041 (exp.) vs. 0.053 (theory)

Cao, Guo et al., Sci. China: PMA 68, 267011 (2025)

$CeMgAl_{11}O_{19}$: slow spin dynamics



$$A(t) = A_1 G_{KT}(t,\Delta)\exp[-(\lambda t)^{\beta}]$$



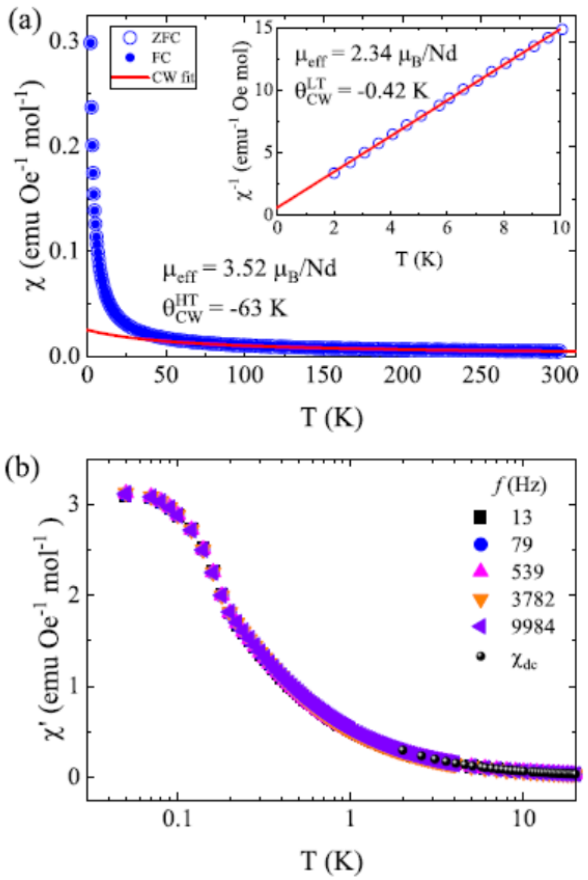
$$\lambda(H) = 2\Delta^2\tau^x \int_0^{\infty} t^{-x}\exp(-\nu t)\cos(\gamma_\mu \mu_0 H t)\mathrm{d}t$$

$x = 0.53(3)$, $\nu = 1.0(2)$ MHz, $\Delta = 9(1)$ MHz →

- out of the fast fluctuation limit for muSR
- less than canonical spin glass systems （10^8 Hz）

→ slow collective fluctuations

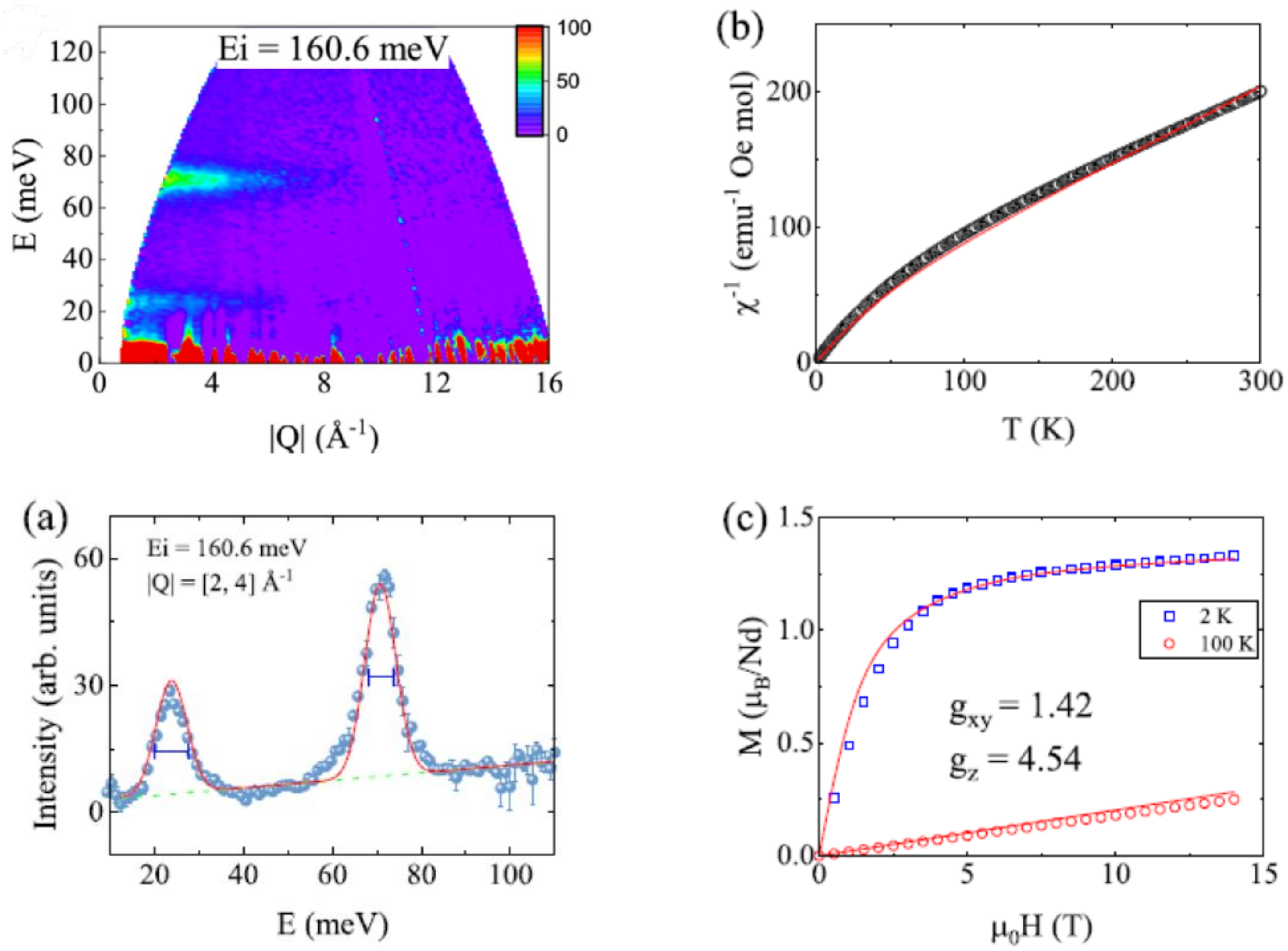
$NdZnAl_{11}O_{19}$: CEF ground state



☐ AFM dominant

☐ no order down to 50 mK

Cao, Guo et al., PRB 112, 144409 (2025)



☐ CEF fitting → Ising dominant anisotropy

$NdZnAl_{11}O_{19}$: CEF ground state

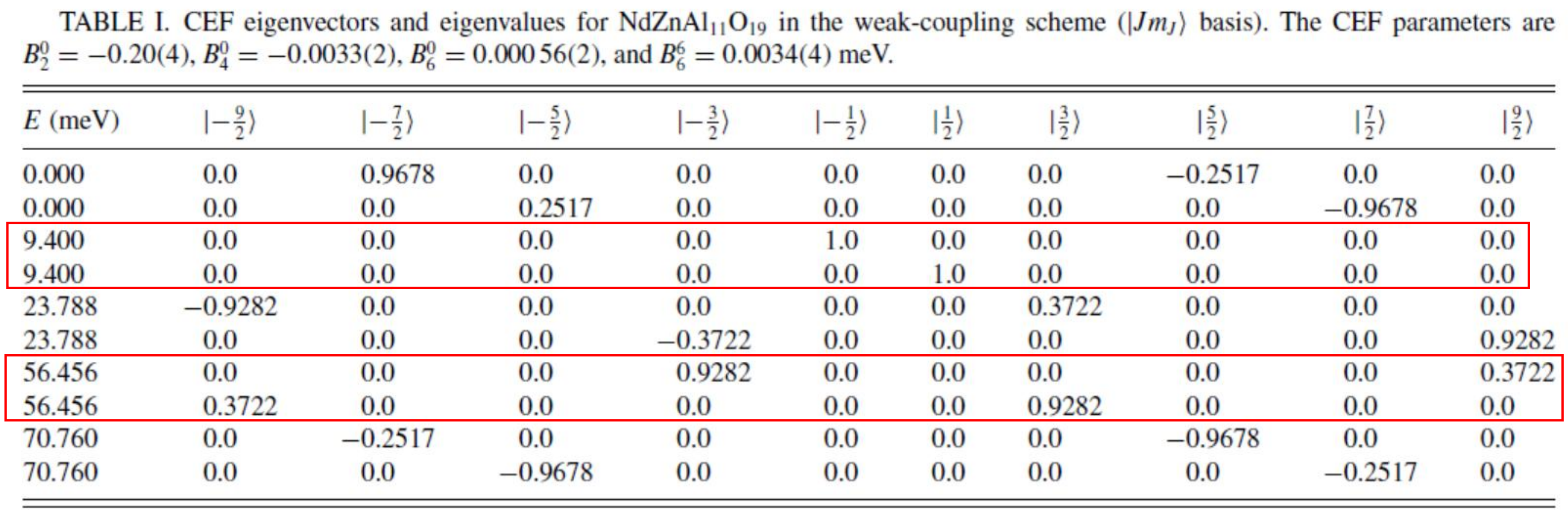
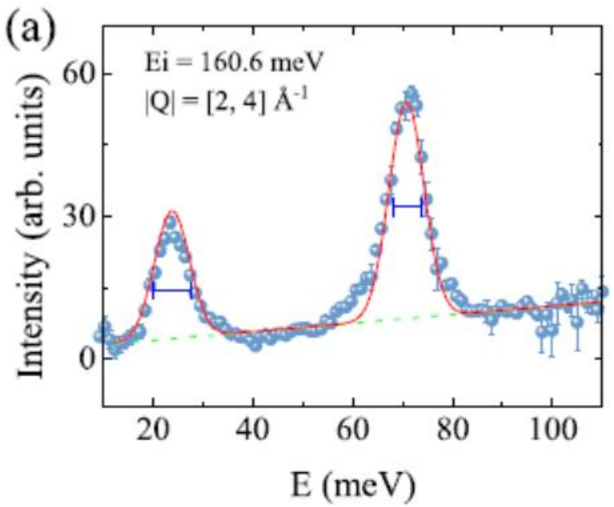


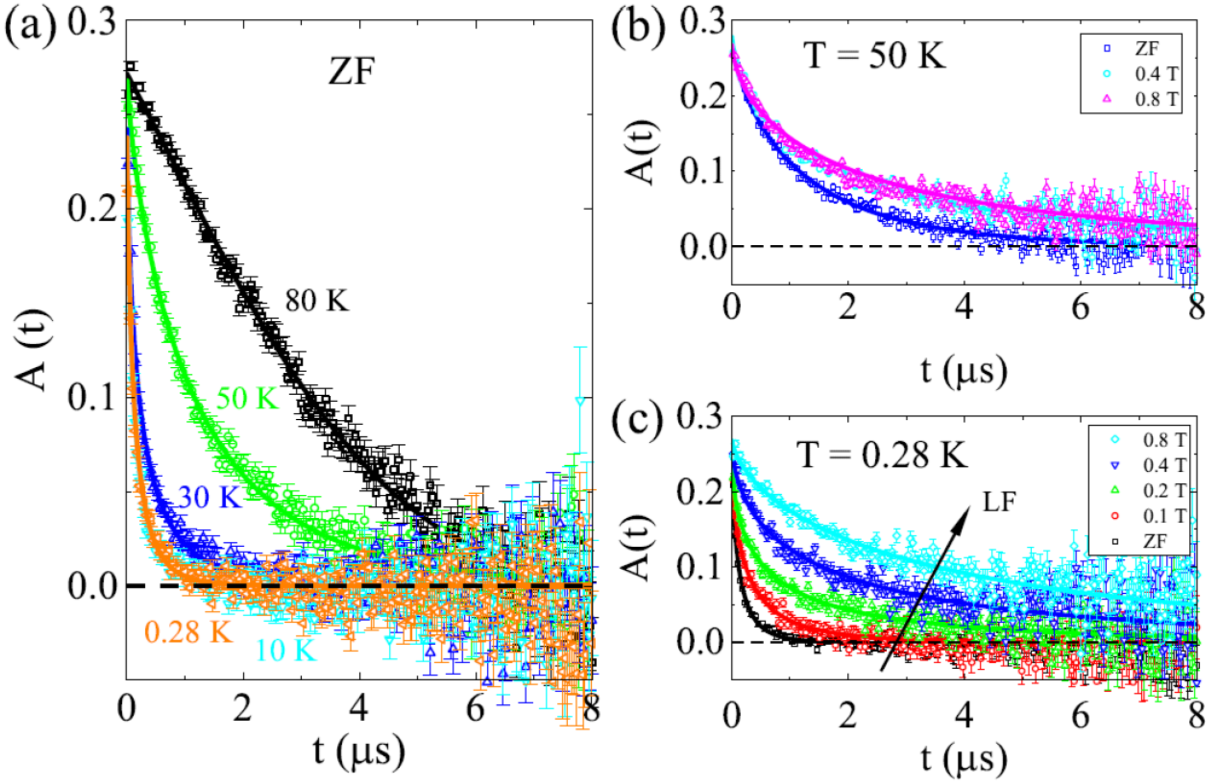
TABLE I. CEF eigenvectors and eigenvalues for $NdZnAl_{11}O_{19}$ in the weak-coupling scheme ($|Jm_J\rangle$ basis). The CEF parameters are $B_2^0 = -0.20(4)$, $B_4^0 = -0.0033(2)$, $B_6^0 = 0.000\,56(2)$, and $B_6^6 = 0.0034(4)$ meV.

E (meV)	$\|-\frac{9}{2}\rangle$	$\|-\frac{7}{2}\rangle$	$\|-\frac{5}{2}\rangle$	$\|-\frac{3}{2}\rangle$	$\|-\frac{1}{2}\rangle$	$\|\frac{1}{2}\rangle$	$\|\frac{3}{2}\rangle$	$\|\frac{5}{2}\rangle$	$\|\frac{7}{2}\rangle$	$\|\frac{9}{2}\rangle$
0.000	0.0	0.9678	0.0	0.0	0.0	0.0	0.0	−0.2517	0.0	0.0
0.000	0.0	0.0	0.2517	0.0	0.0	0.0	0.0	0.0	−0.9678	0.0
9.400	0.0	0.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0
9.400	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0
23.788	−0.9282	0.0	0.0	0.0	0.0	0.0	0.3722	0.0	0.0	0.0
23.788	0.0	0.0	0.0	−0.3722	0.0	0.0	0.0	0.0	0.0	0.9282
56.456	0.0	0.0	0.0	0.9282	0.0	0.0	0.0	0.0	0.0	0.3722
56.456	0.3722	0.0	0.0	0.0	0.0	0.0	0.9282	0.0	0.0	0.0
70.760	0.0	−0.2517	0.0	0.0	0.0	0.0	0.0	−0.9678	0.0	0.0
70.760	0.0	0.0	−0.9678	0.0	0.0	0.0	0.0	0.0	−0.2517	0.0

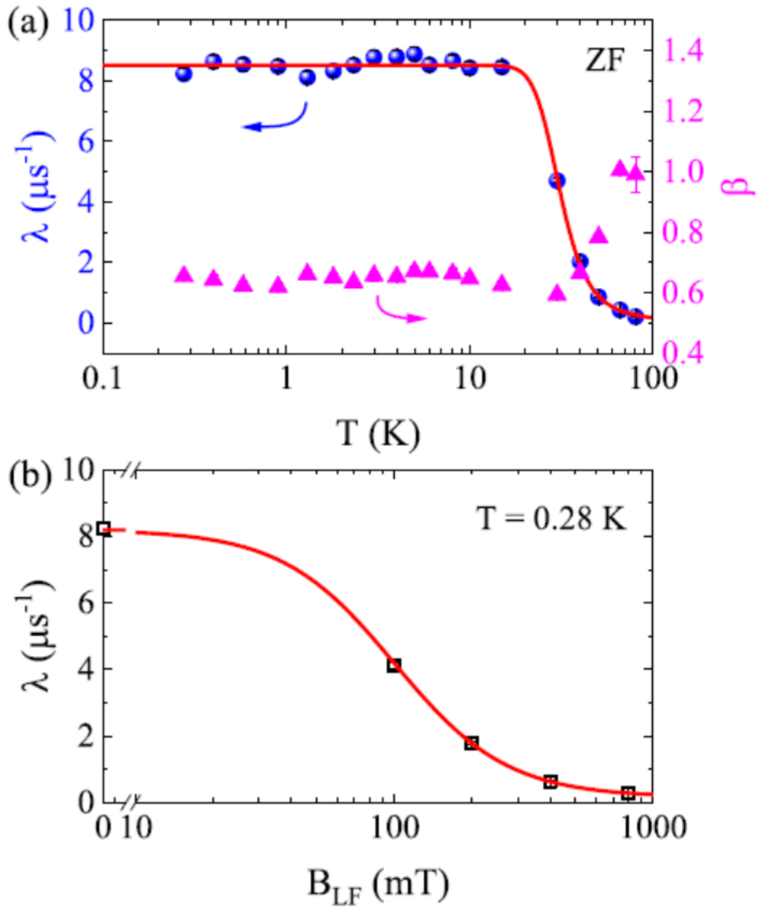
$$\sum_{\alpha} |\langle \Gamma_j | \hat{J}_\alpha | \Gamma_i \rangle|^2$$



persistent spin fluctuations

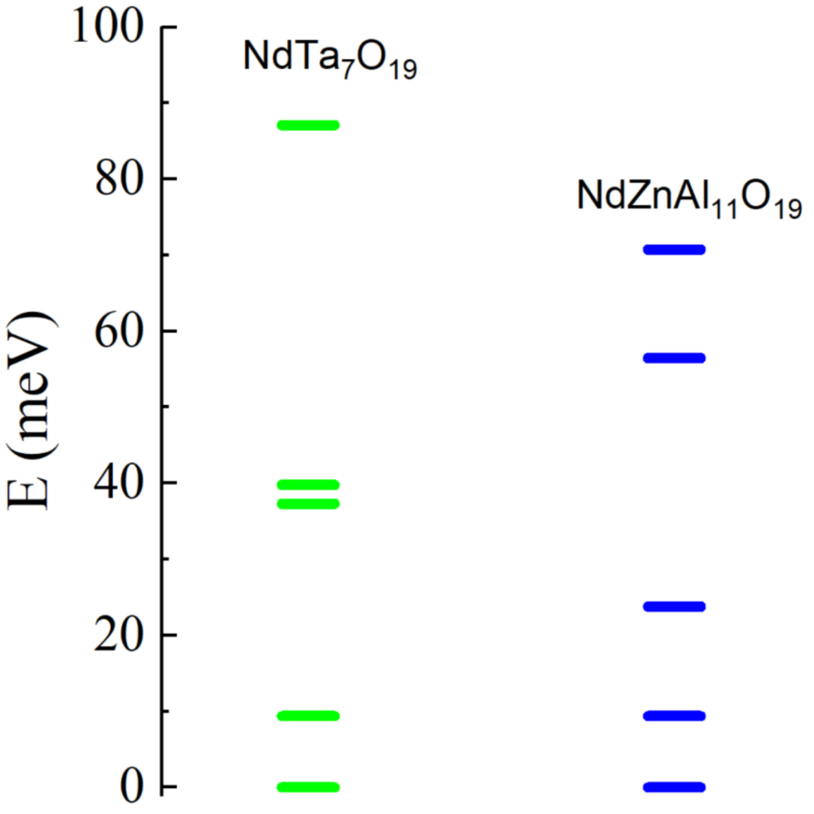


$$A(t) = A_1 \exp[-(\lambda t)^{\beta}]$$



ν = 85.3(8) MHz << 37 GHz（exchange fluctuation rate）

$NdTa_7O_{19}$ vs. $NdZnAl_{11}O_{19}$



$$\frac{g_z}{g_{xy}} = \frac{2.78}{1.22} = 2.28$$

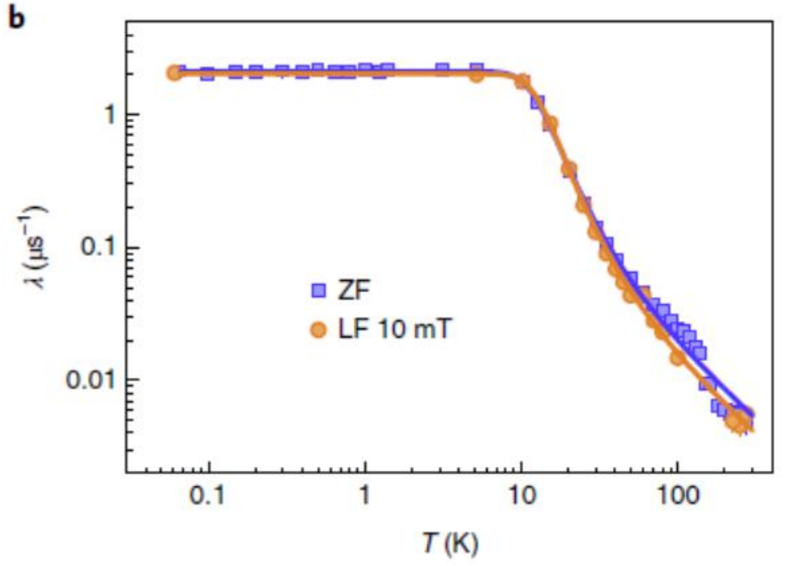
$$\theta_{CW} = -0.46\ \mathrm{K}$$

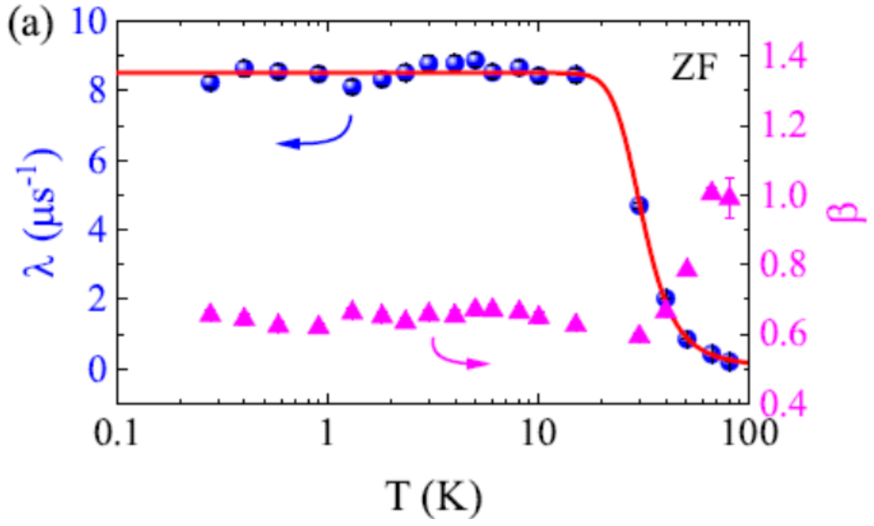
Nat. Mater. 21, 416 (2022)

$$\frac{g_z}{g_{xy}} = \frac{4.54}{1.42} = 3.2$$

$$\theta_{CW} = -0.42\ \mathrm{K}$$

our work

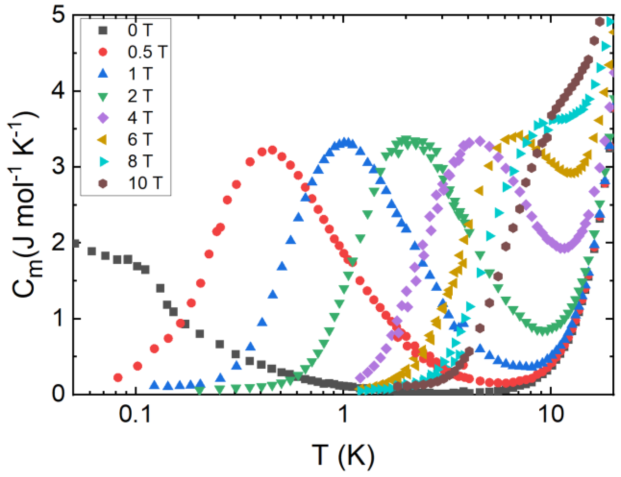


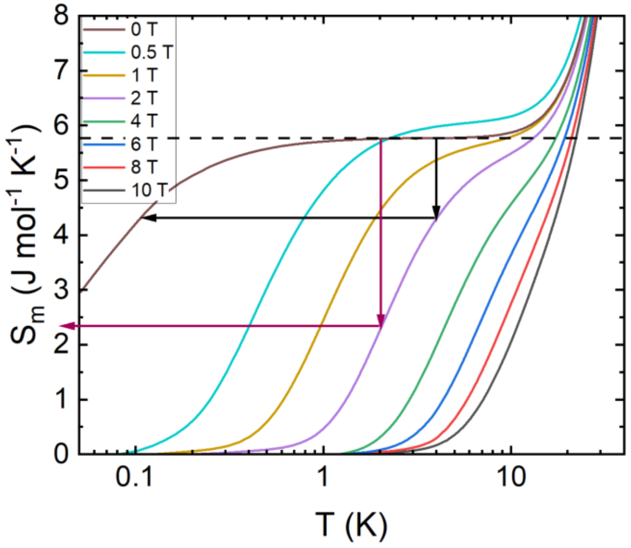


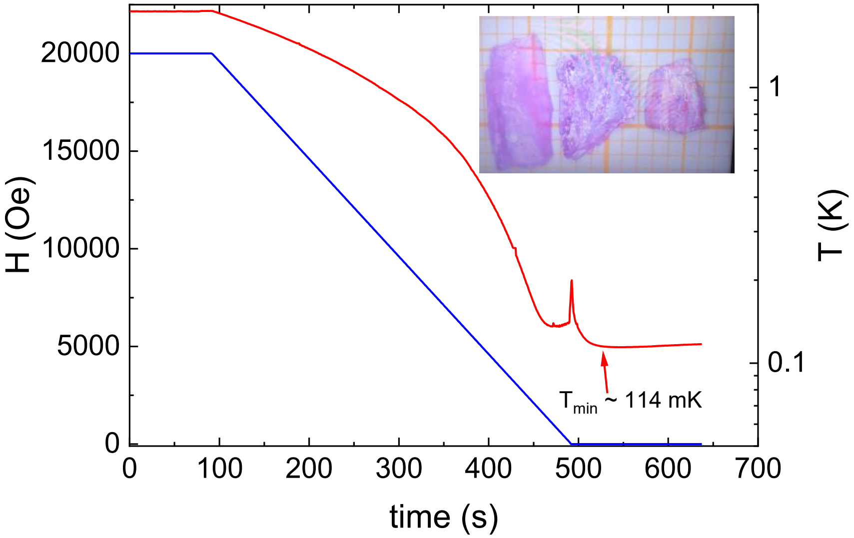
ADR performance

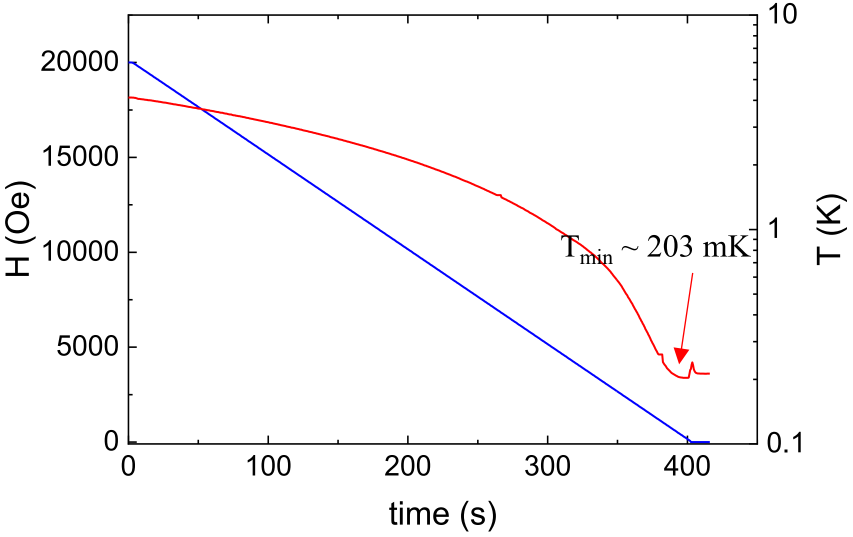
Hexaaluminate

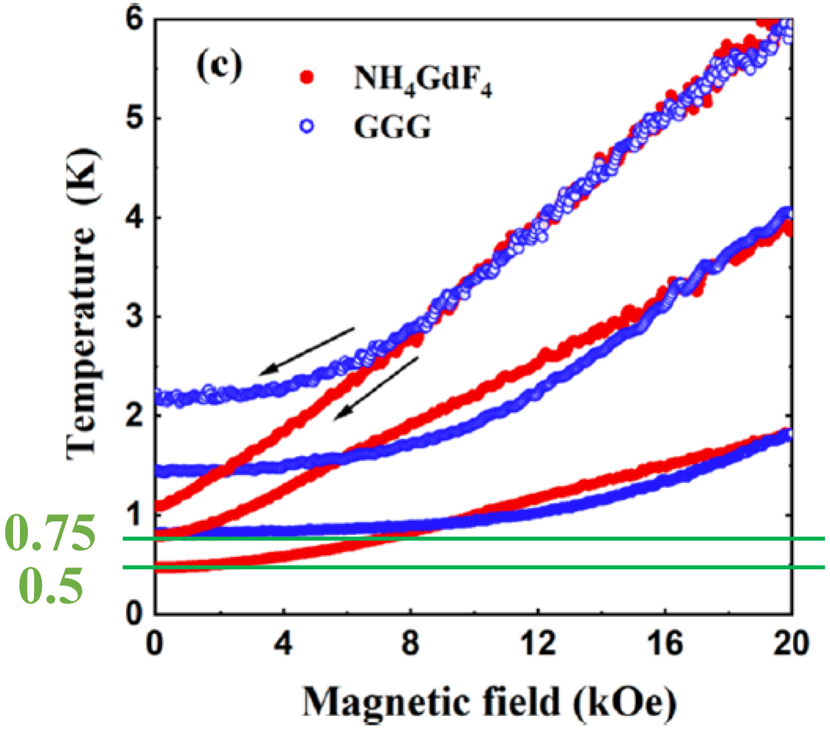
Superior ADR performance than some Gd-based compounds











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Summary for hexaaluminates

- Using muSR, combined with other techniques (specific heat, susceptibility, neutron…), we identify several quantum spin liquid candidates

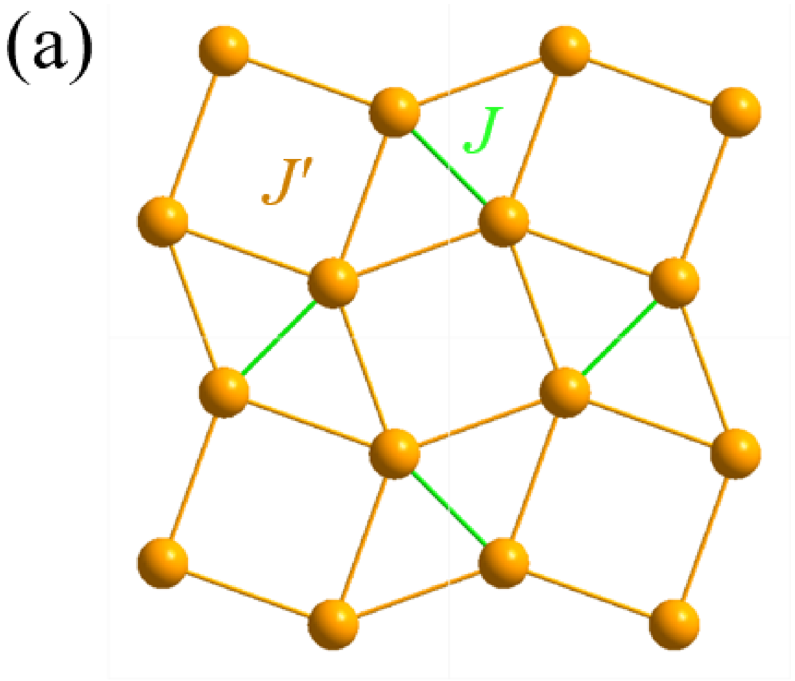
- The versatility of rare earth ions provides a fertile ground for studying different models (intrinsic transverse field Ising model, Ising dominant XXZ model…)

- QSLs retain a large amount of magnetic entropy at low temperatures;
  small exchange energy scale means a small magnetic field can easily polarize the spin system – remove entropy at low T
  → excellent for magnetic cooling

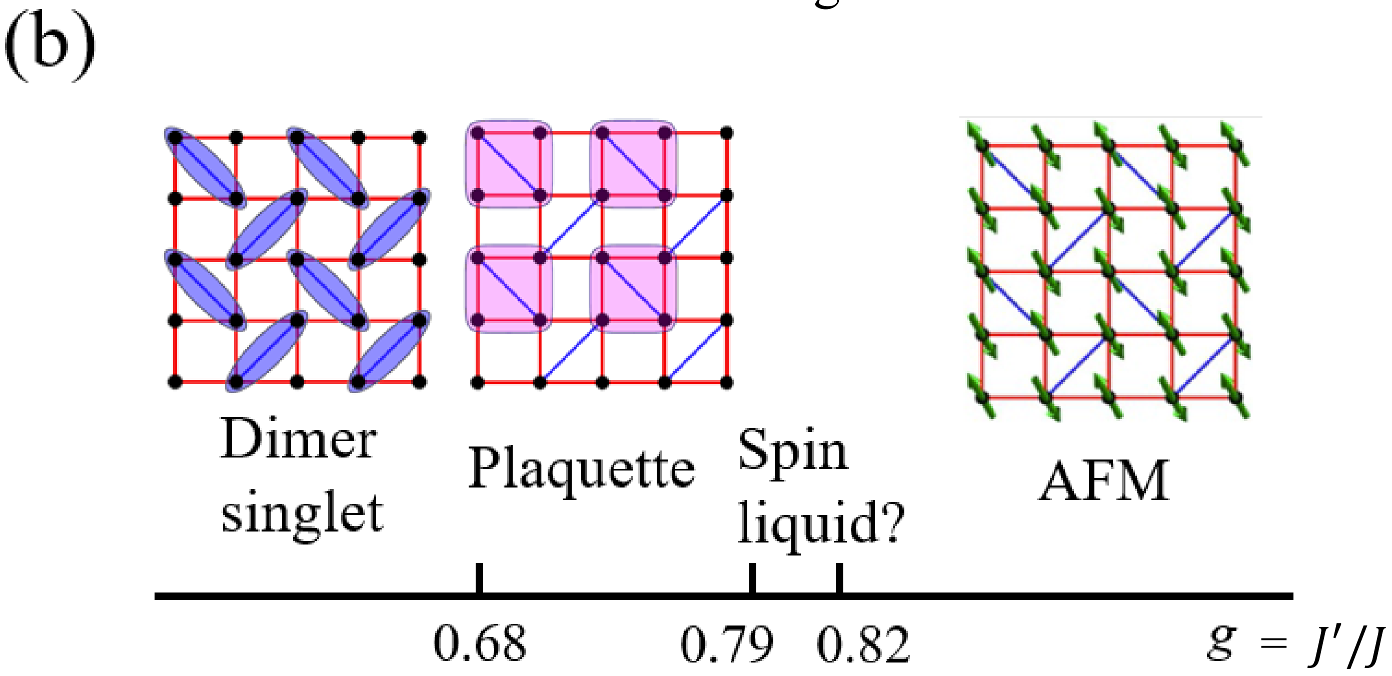
Proximity to a QSL in the SSL $Nd_2Be_2GeO_7$

phase diagram

Shastry-Sutherland lattice



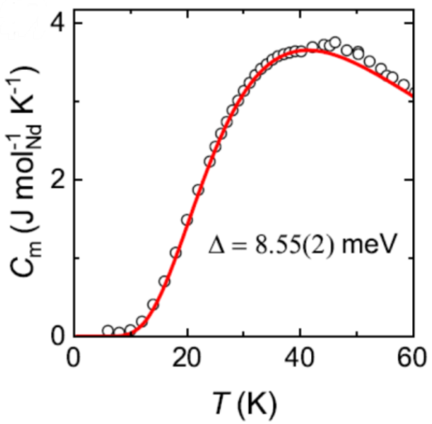
phase diagram based on the
Heisenberg model

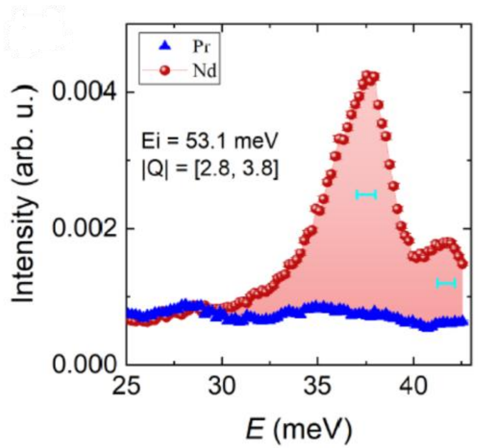


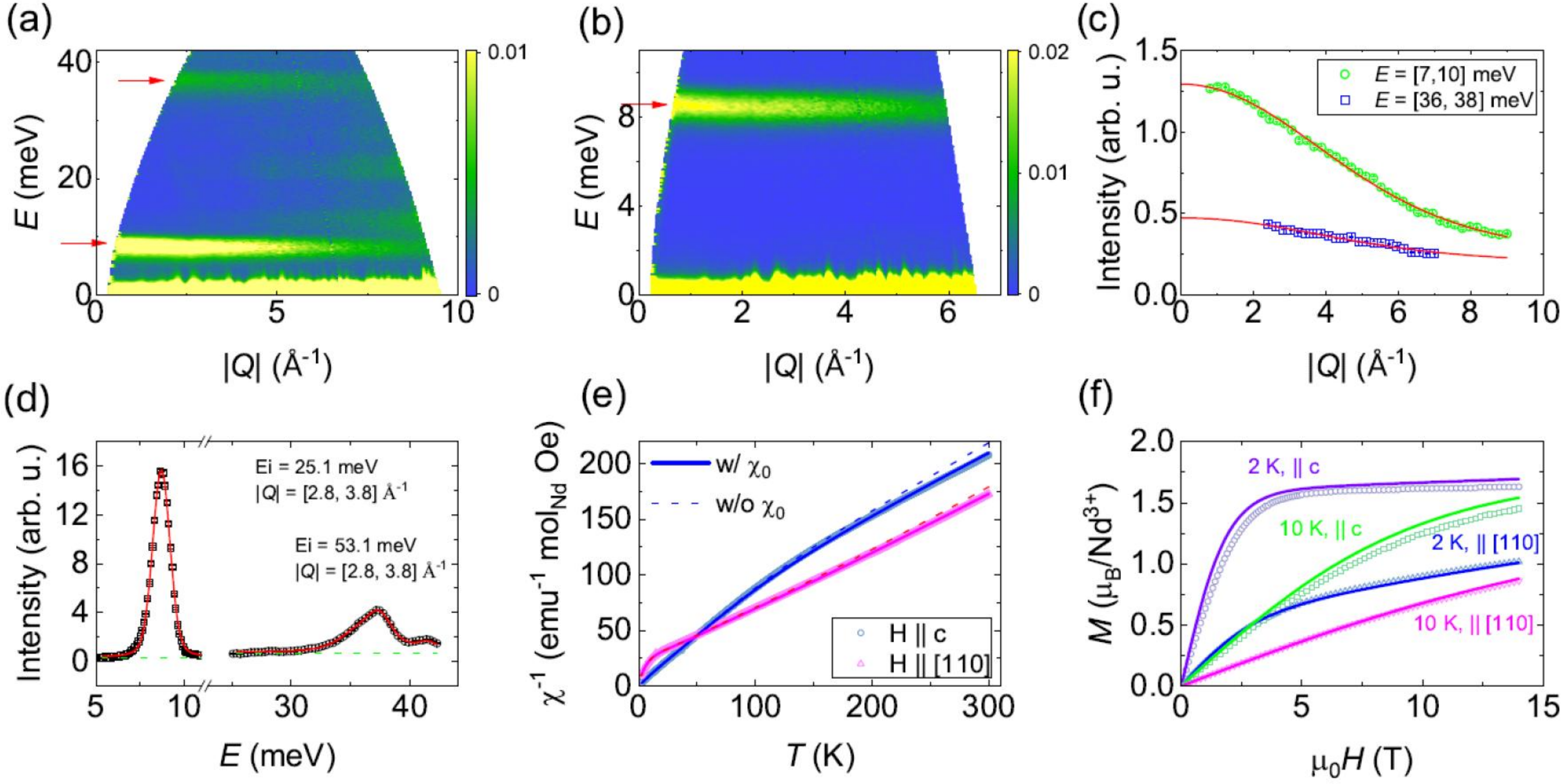
SOC-induced anisotropy ⇨

QSL still exists?
Other exotic phases?

CEF ground state







$$\mathcal{H}_{CEF} = \sum_{l,m} B_l^m O_l^m$$

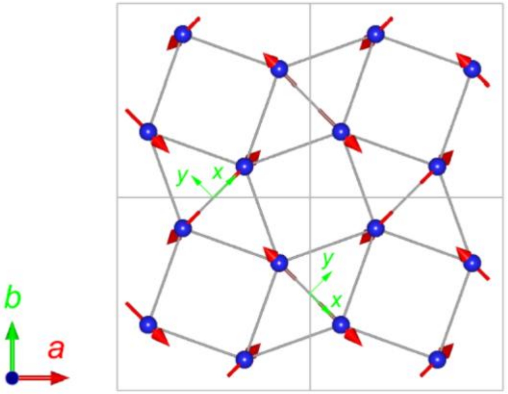
Nd^{3+}: J = 9/2

five Kramers doublets, four transitions at low T

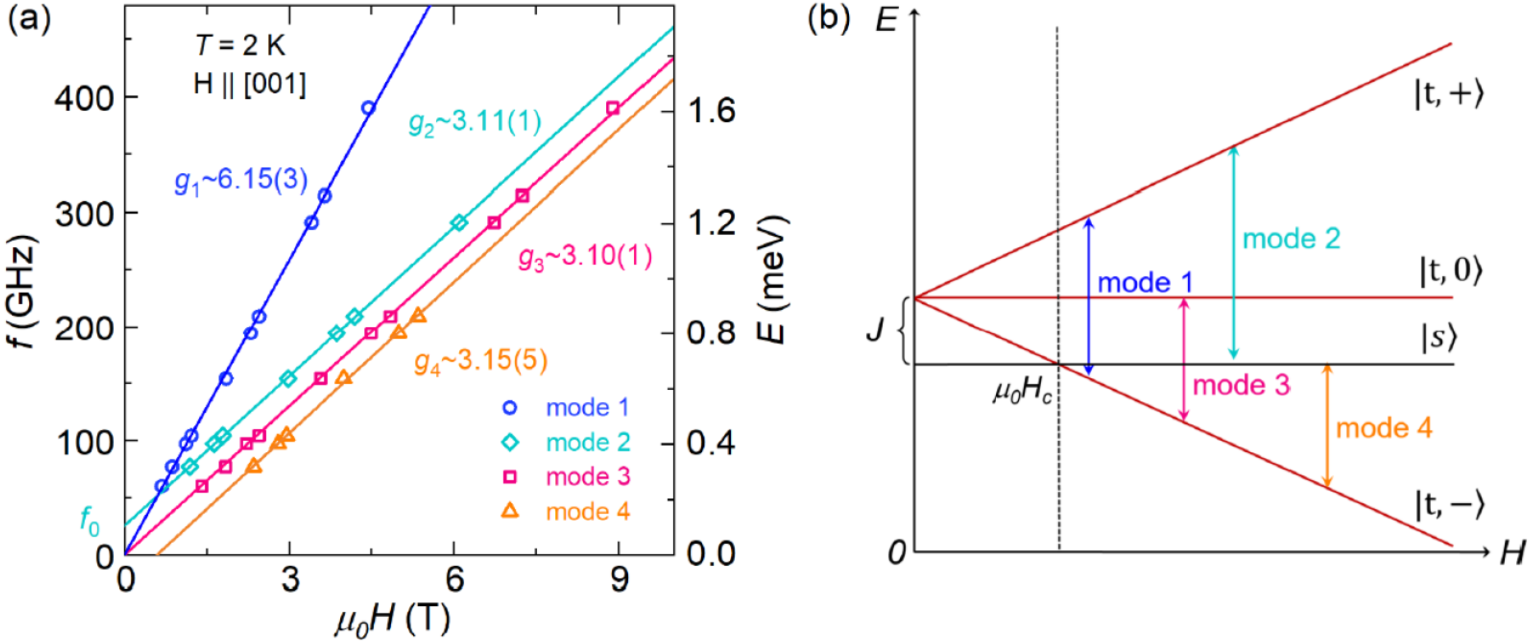
C_s point symmetry: up to 15 nonzero B_l^m

Liu, Guo et al., under review

dimer model



Electron spin resonance



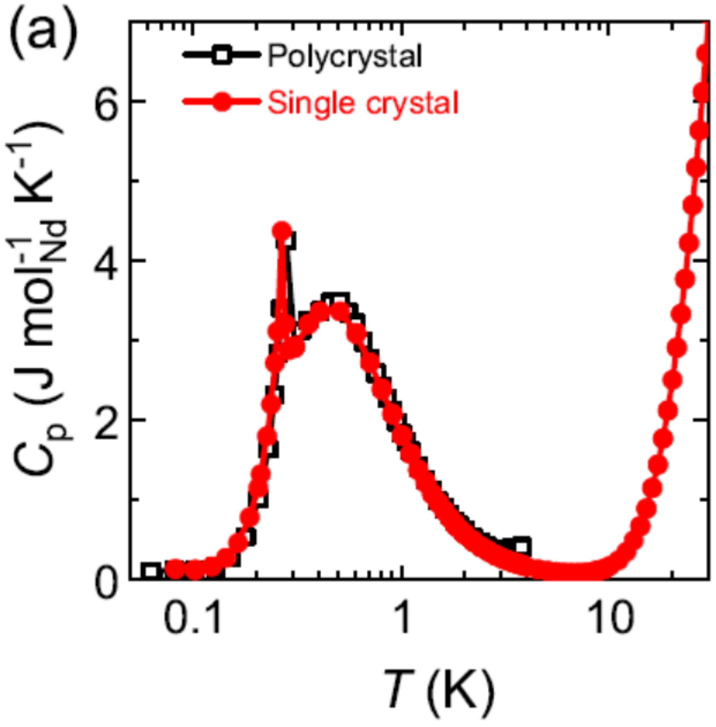
$$g_x^{CEF} = 2.02$$
$$g_y^{CEF} = 0.14$$
$$g_z^{CEF} = 3.18$$

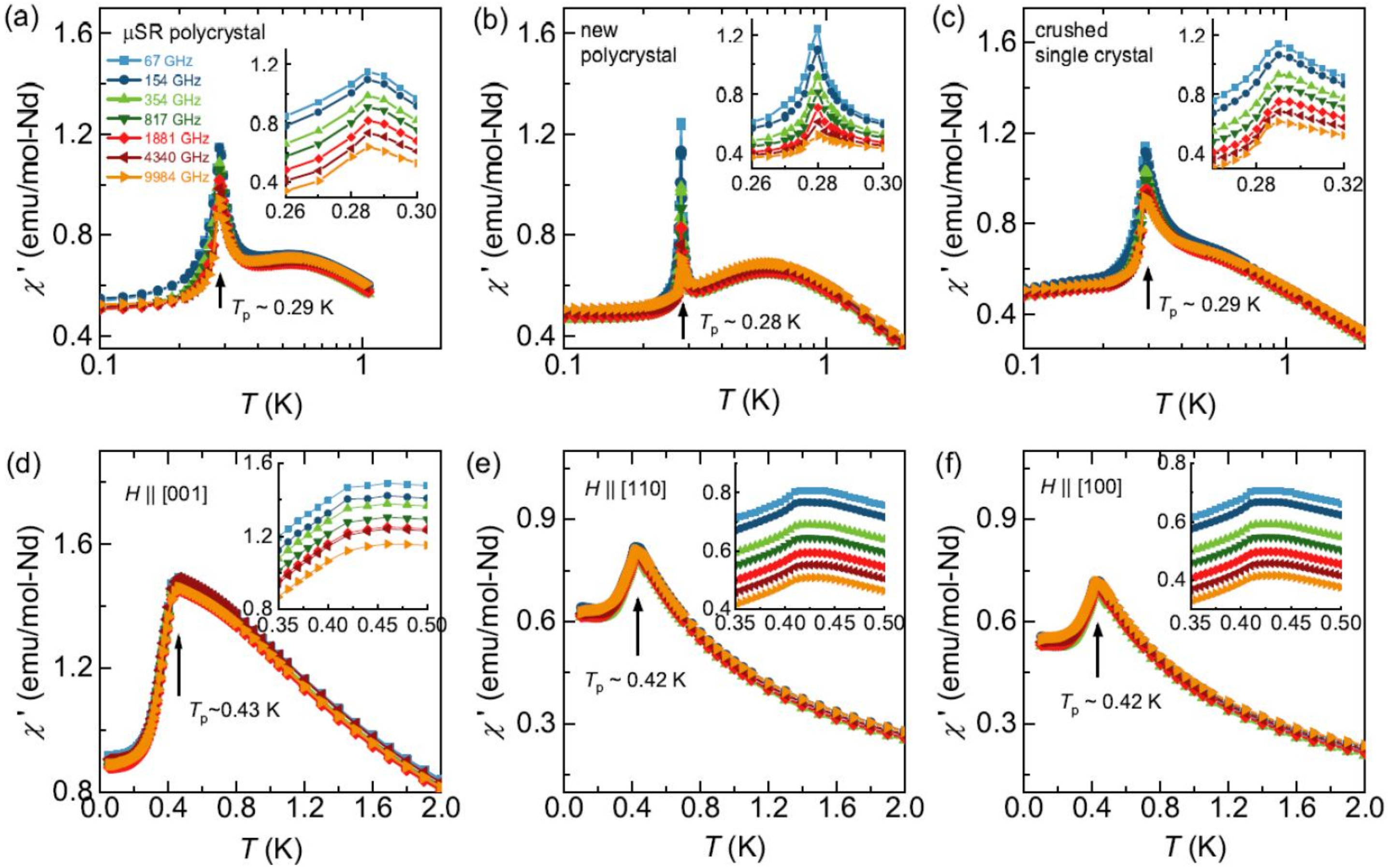
J = 0.106(2) meV

ESR transitions between the singlet and triplet are usually forbidden

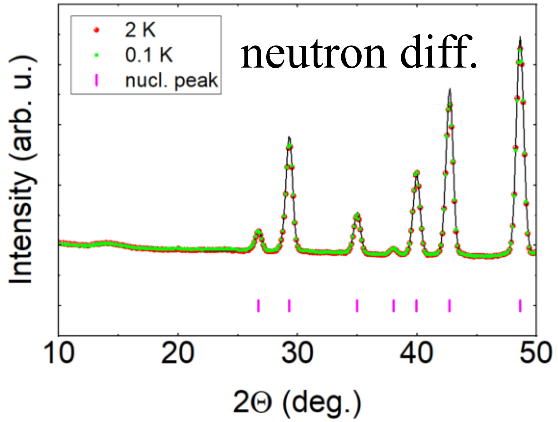
DM interaction ⇒ $|s\rangle = \alpha|00\rangle + \beta|10\rangle$

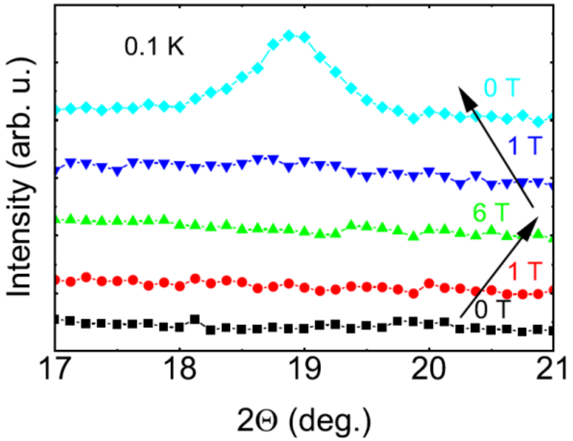
long-range magnetic ordering

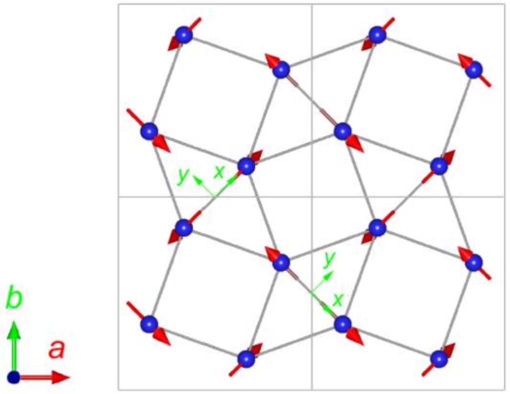


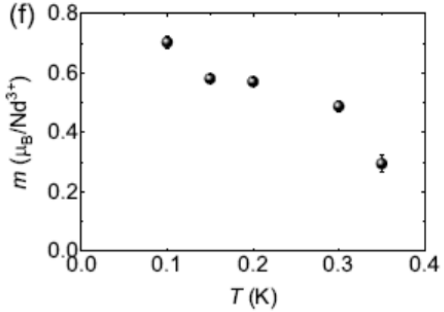


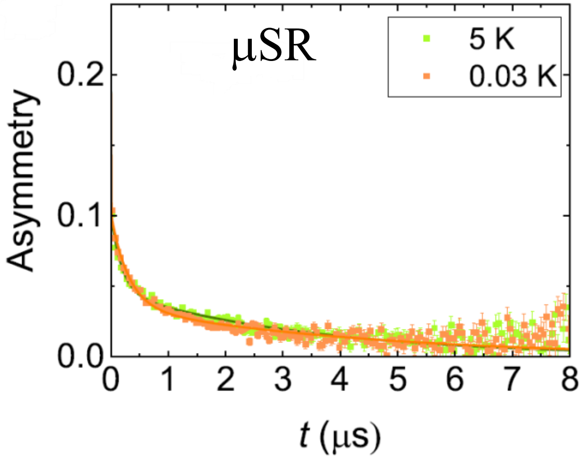
hidden order?









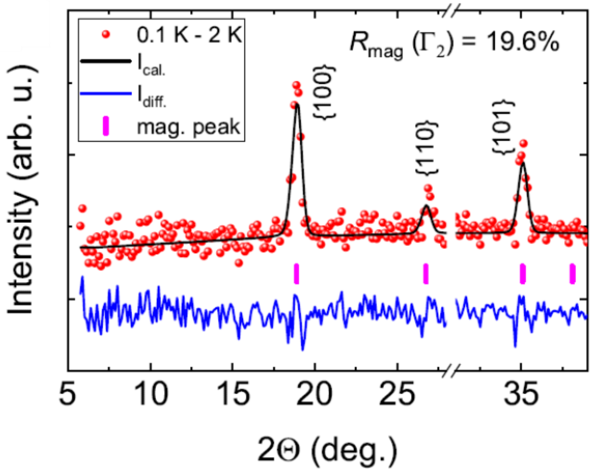


thermodynamic phase transition

✚

no order parameter （neutron, muon）

⇨ hidden order



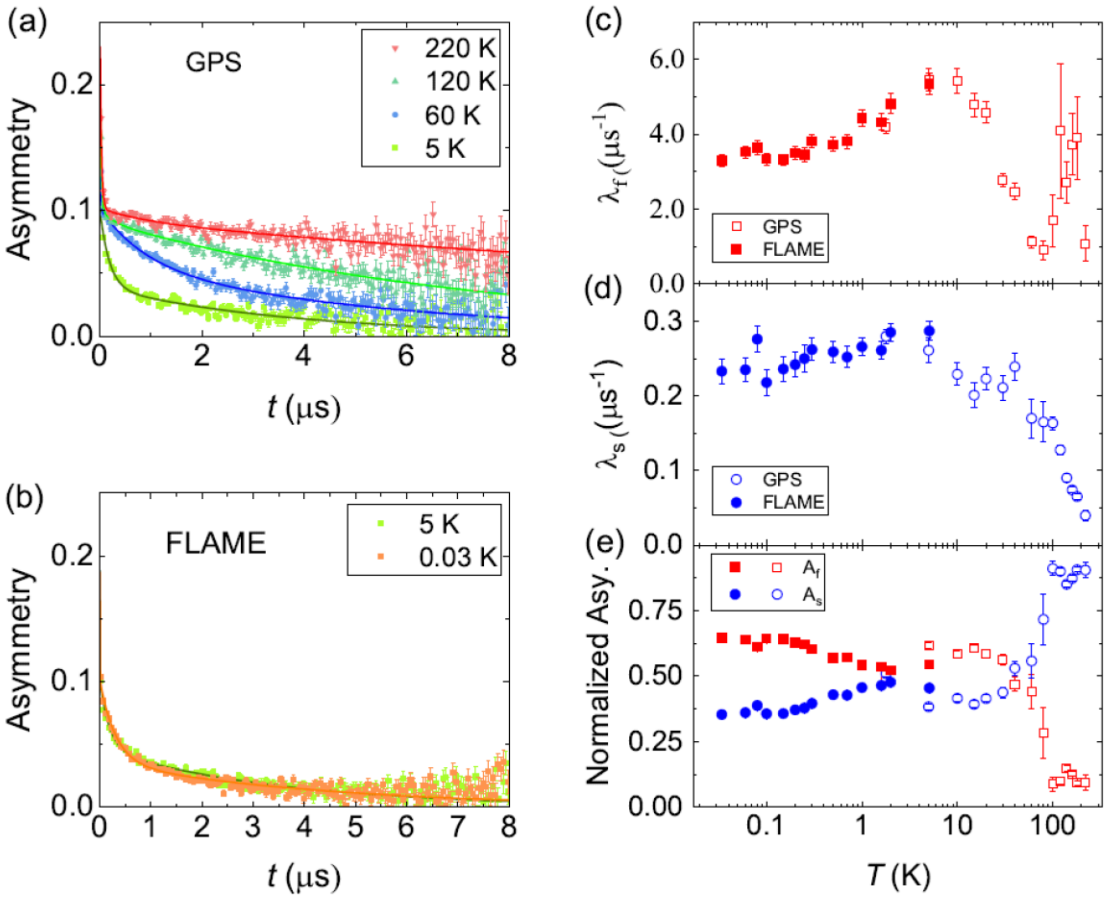
magnetic order revealed after field-training

	CEF GS	neutron diff.
m_x	0.82	0.60
m_y	0	0
m_z	1.55	0.66

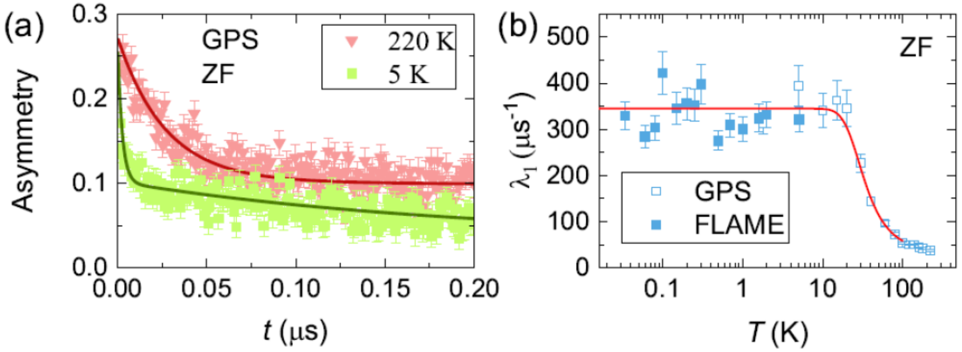
strong quantum fluctuations along c axis

spin dynamics

$Nd_2Be_2GeO_7$



$$A(t) = A_1 \exp(-\lambda_1 t) + A_f \exp(-\lambda_f t) + A_s \exp(-\lambda_s t)$$



Relaxed via the Orbach process
(first CEF excitation)

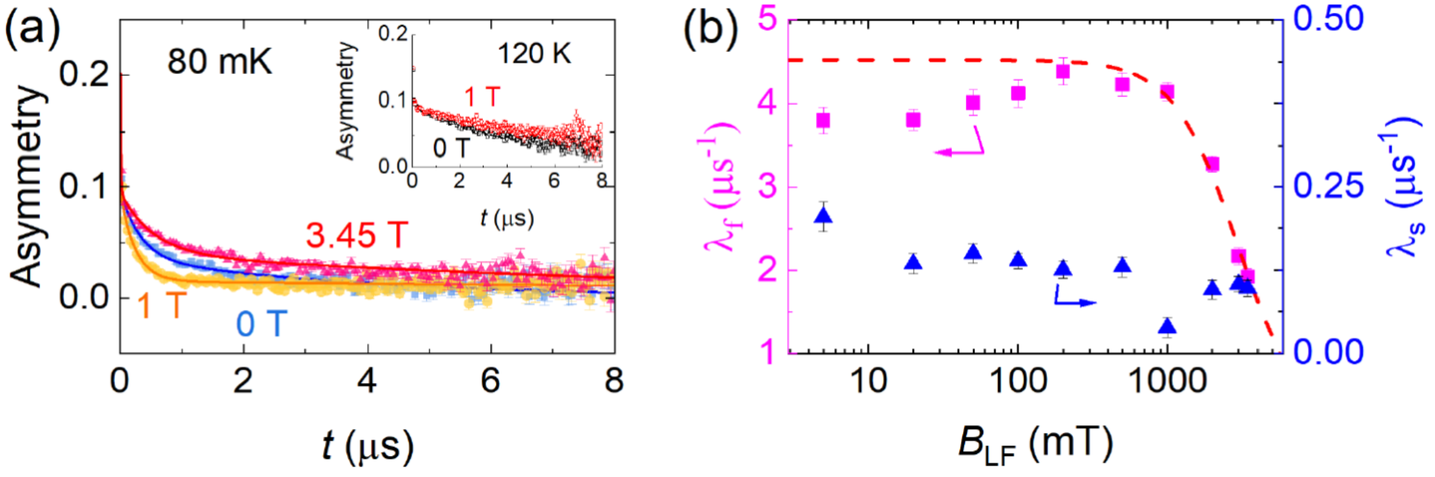
$$\lambda(B_{\mathrm{LF}}) = \frac{2(\gamma_\mu \Delta)^2 \nu}{\nu^2 + (\gamma_\mu B_{\mathrm{LF}})^2}$$

$$\lambda^{-1} = \lambda_0^{-1} + C\exp(-\delta/k_B T)$$

$\delta = 7.8(4)$ meV

unusual field-enhanced spectral weight

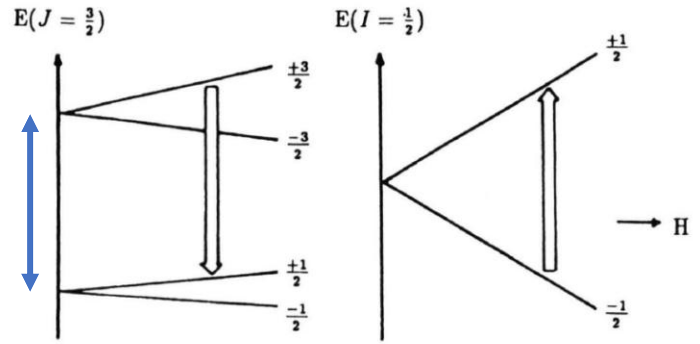
$Nd_2Be_2GeO_7$



nuclear spin

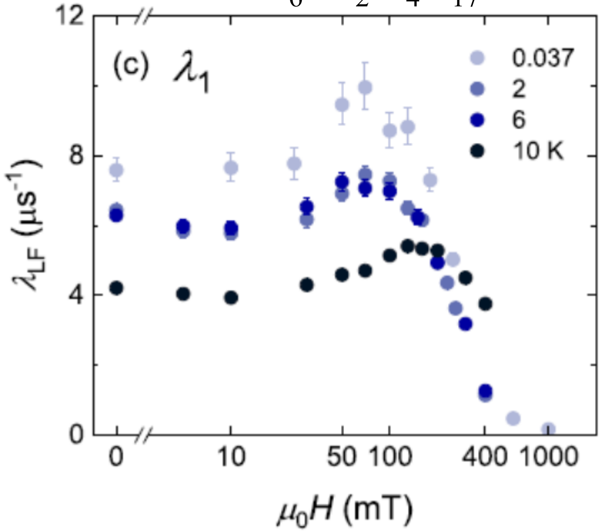
muon spin

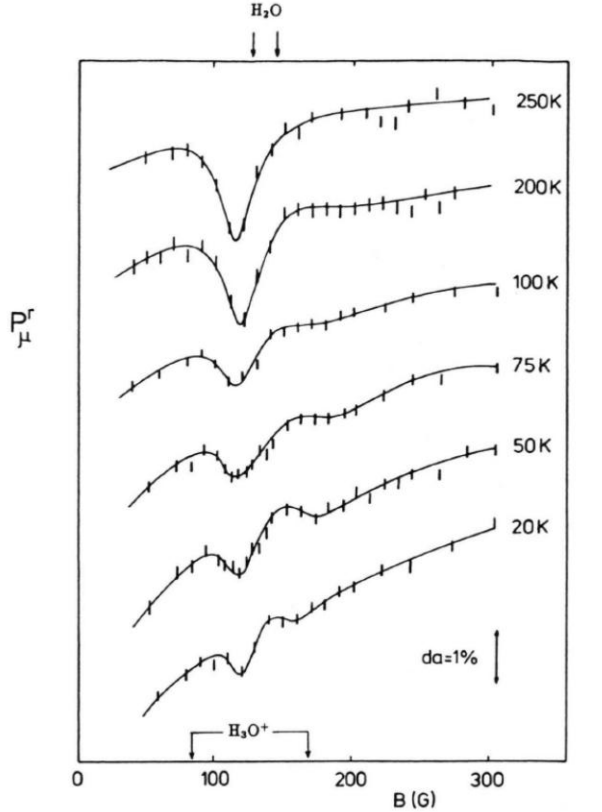
quadrupole splitting



Level crossing resonance:
flip-flop transition

$Ba_6Nd_2Ti_4O_{17}$



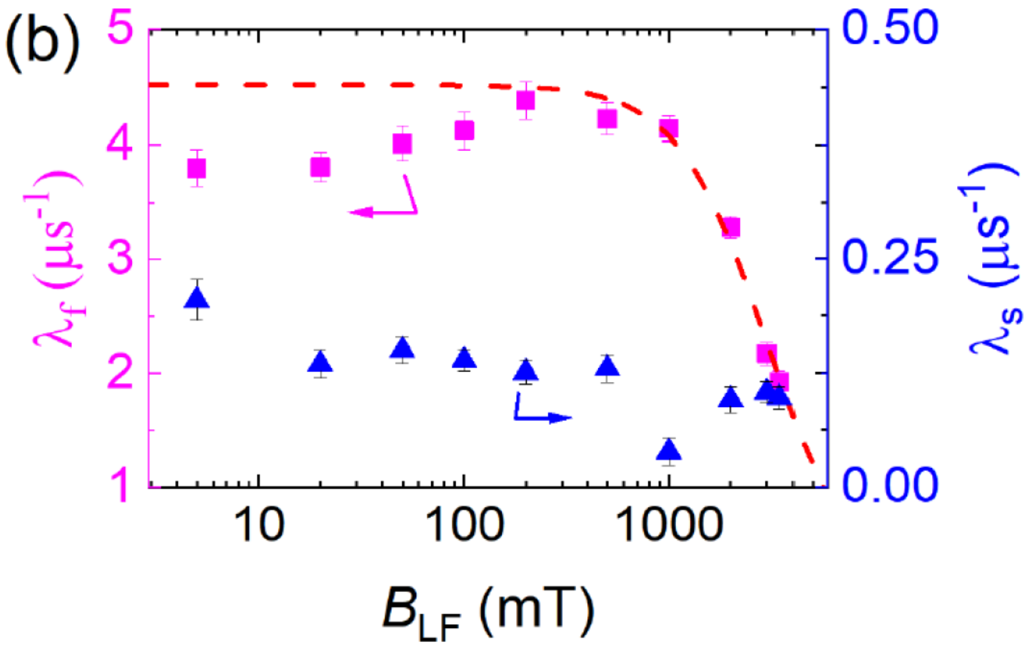


Z. Naturforsch. **47a,** 371-381 (1992)

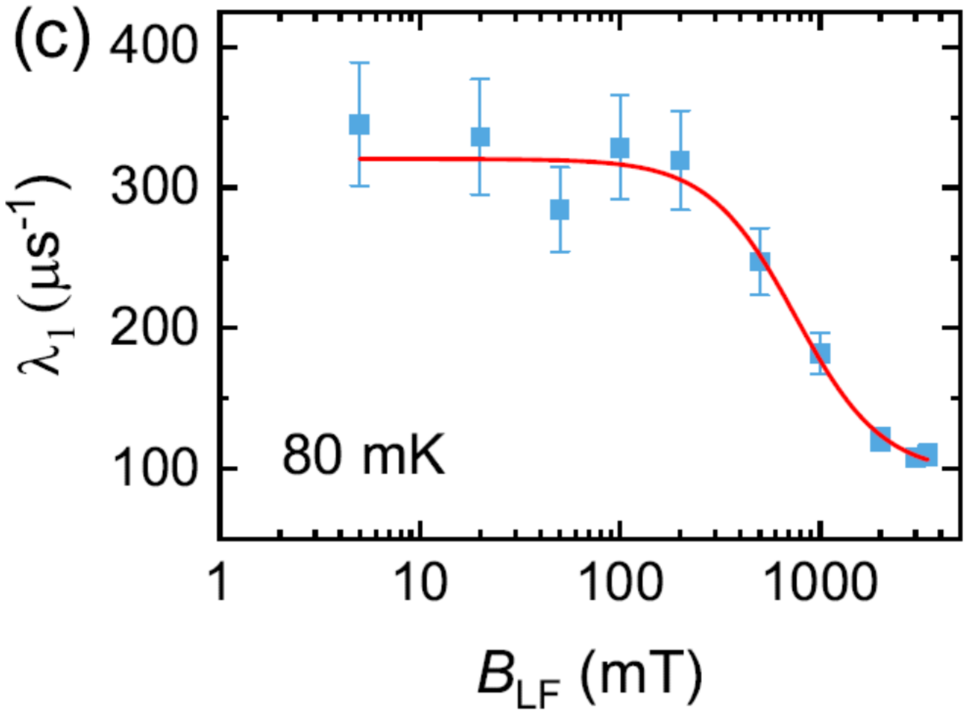
PRB 111, 155148 (2025)

spin dynamics

$Nd_2Be_2GeO_7$

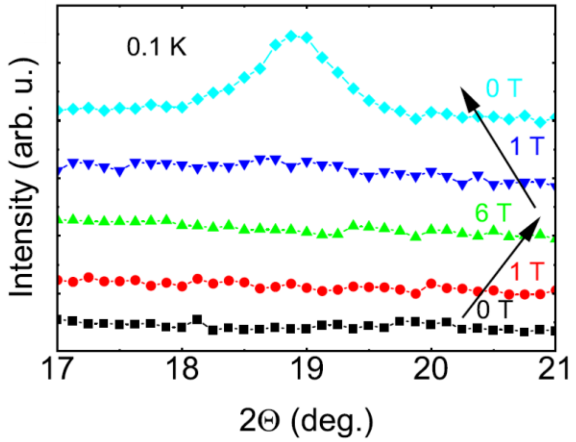


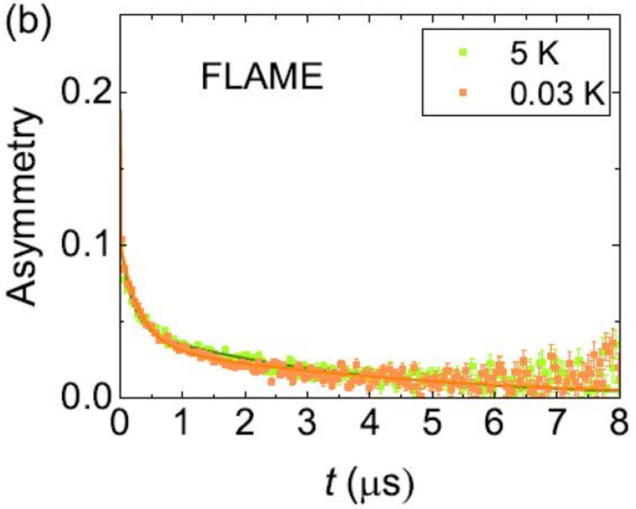
$\nu = 2.6(1)$ GHz



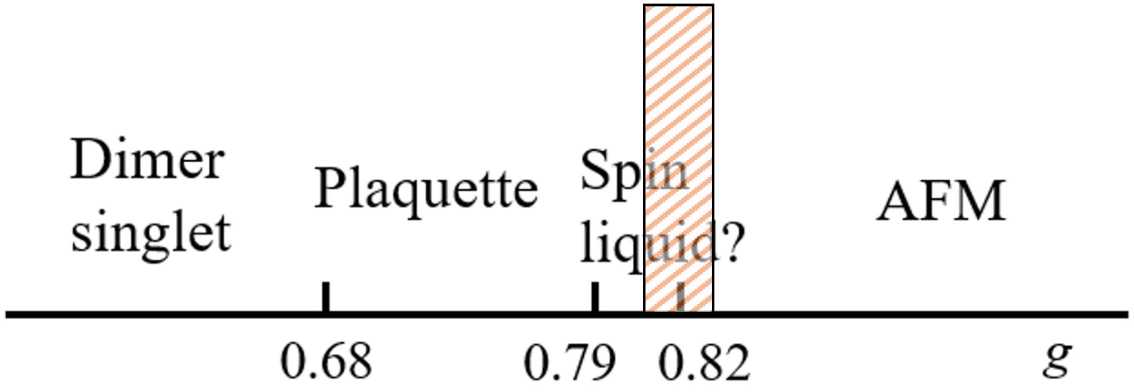
$\nu = 0.64(6)$ GHz

Discussion and Conclusion





- ➢ hidden-order-like transition
- ➢ the order parameter is revealed by neutron after field training, but not in muon experiment



- proximity to the QSL and AFM border, highly degenerate
- magnetic field selects a particular AFM phase, which can manifest itself only when the field is removed
- the stabilized AFM phase is dynamic in nature, but the moments fluctuate coherently in the 10^{-9} s time scale

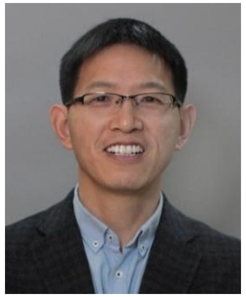
Acknowledgement

PhD Students

Yantao Cao

Andi Liu

Collaborators



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ADR measurements

Beamtime

J-PARC

Science & Technology Facilities Council
ISIS Neutron and Muon Source

PAUL SCHERRER INSTITUT
PSI

Thank you for your attention!