Exploring quantum spin-frustrated materials

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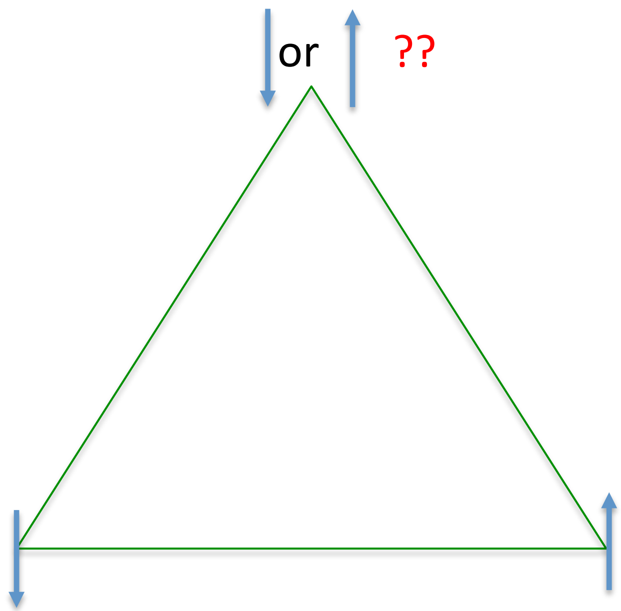
J. Phys.: Condens. Matter 25, 026003 (2013)
Chem. Phys. Lett. 570, 37 (2013)
New J. of Phys. 16, 093011 (2014)
Scientific Reports 5, 16419 (2015)
Phys. Rev. B 93, 060405 (R) (2016)
Phys. Rev. B 94, 024438 (2016)
Phys. Rev. Lett. 115, 167203 (2015)
Phys. Rev. Lett. 117, 097201 (2016)
Phys. Rev. Lett. 117, 267202 (2016)
Phys. Rev. Lett. 118, 107202 (2017)
Nature 540, 559-562 (2016)

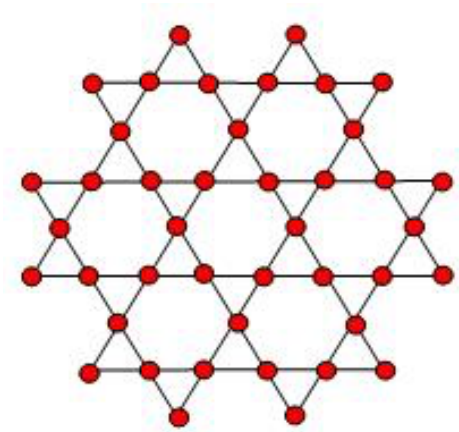
Outline

- A short introduction
- $(Zn,Co,Ni)Cu_3(OH)_6Cl_2$: two-dimensional kagome system
- $ZnCu_3(OH)_6SO_4$: anisotropic kagome system
- $YbMgGaO_4$: structurally perfect triangular system
- Summary

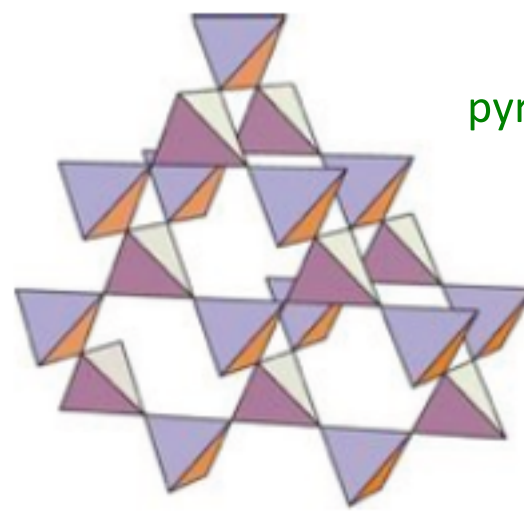
Geometrical frustration



Building block for spin frustrated systems
kagome, pyrochlore, spinel…



kagome



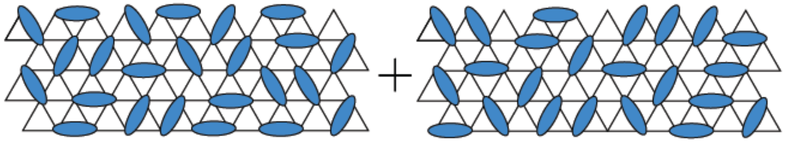
pyrochlore

Quantum spin liquid—RVB state

P. W. Anderson, 1973

Resonating Valence Bond state

$$\Psi = $$



$$+ \ldots$$

$$\text{(singlet bond)} = \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$$

X.-G. Wen

Buckley Prize winner 2017

RVB state – an example of a topological phase

U(1) quantum spin liquid

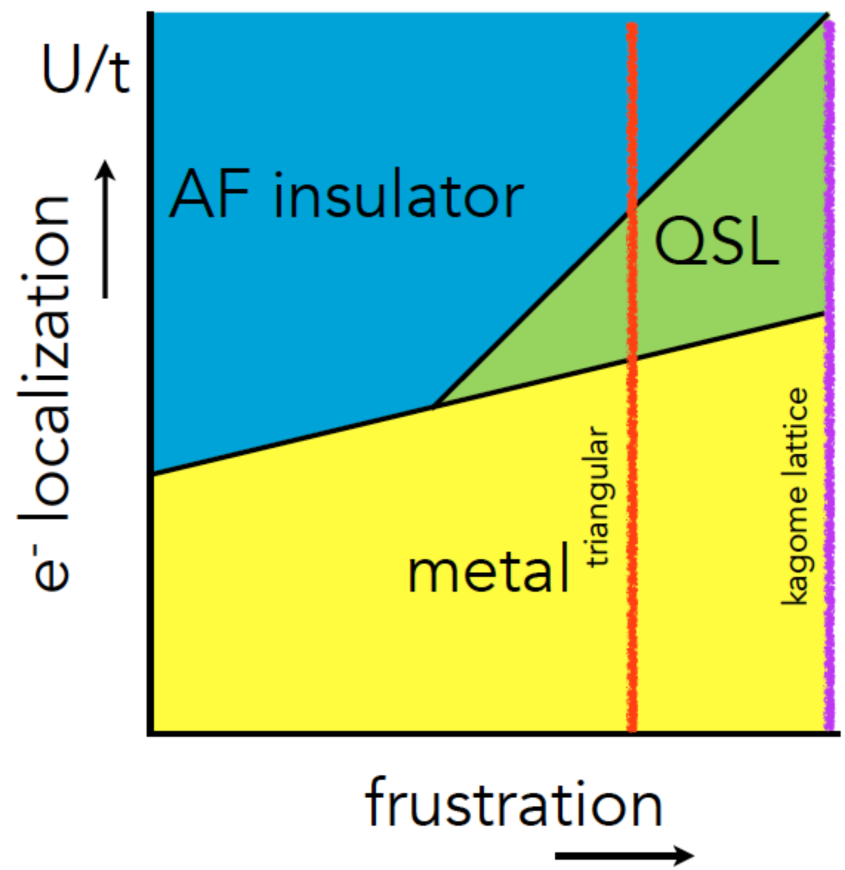
Z2 quantum spin liquid

SU(2) quantum spin liquid

"Sheepskin Scroll"

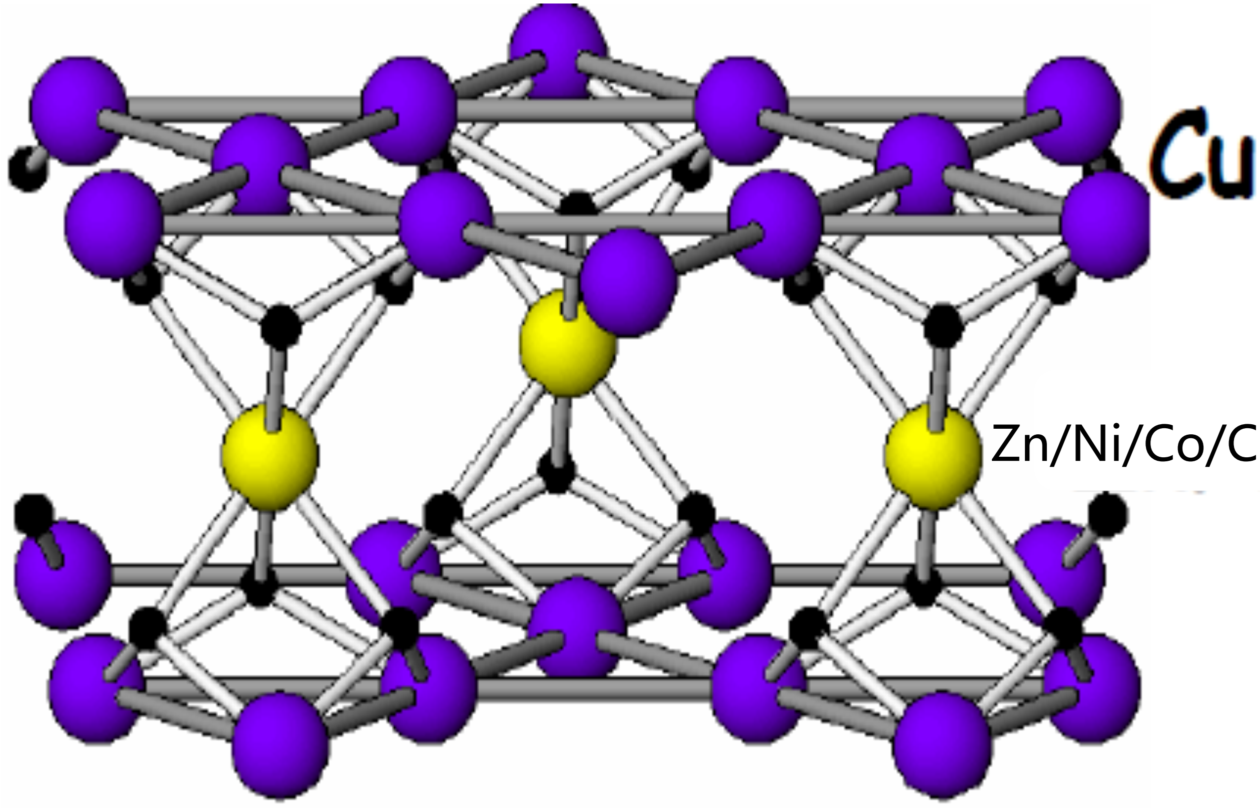
Where to find the mysterious "liquid" in the desert of condensed matter?

- S=1/2
- Antiferromagnetic coupling and frustration geomtry
- Large frustration factor f (=Θ_W/T_N)
- No long-range magnetic ordering
- No spontaneous symmetry breaking
- Strong charge fluctuation and/or exotic interactions
- Fractional excitation: spinon



From L. Balents

Herbertsmithite

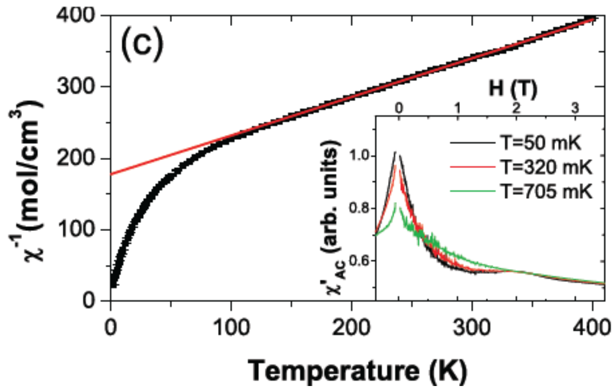


$ZnCu_3(OH)_6Cl_2$

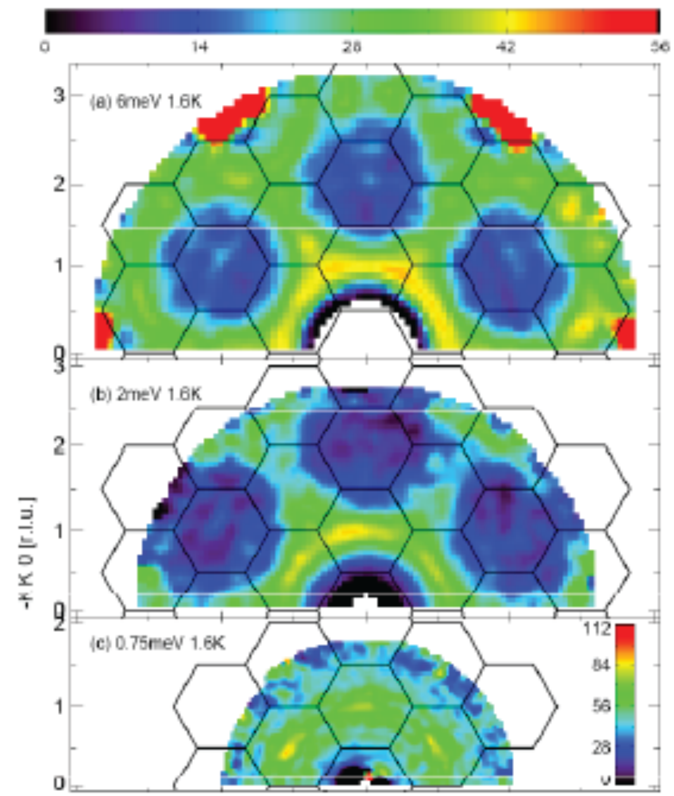
Antisite disorder/
impurities

Herbertsmithite

- AF coupling J~200K
- No magnetic ordering down to 50 mK
- Linear magnetic specific heat



J. S. Helton et al., Phys. Rev. Lett. 98, 107204 (2007)

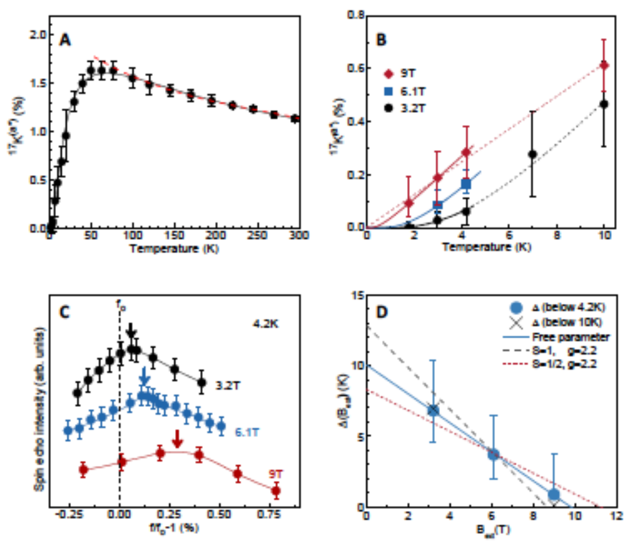


T. H. Han et al., Nature 492, 406 (2012)

Continuum of spin excitations

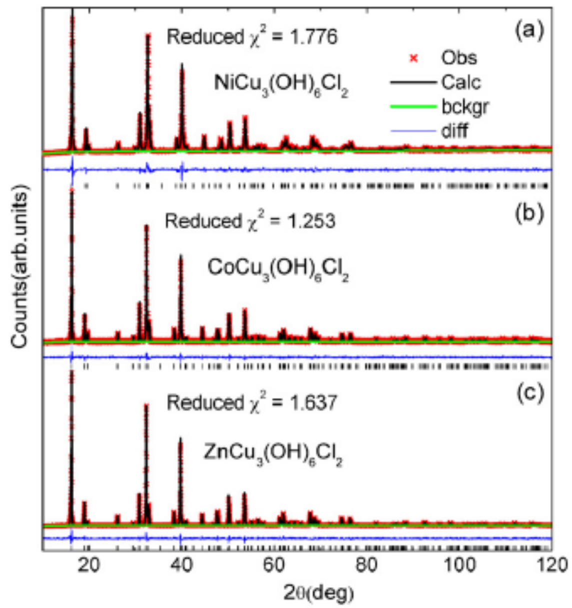
Single crystal NMR

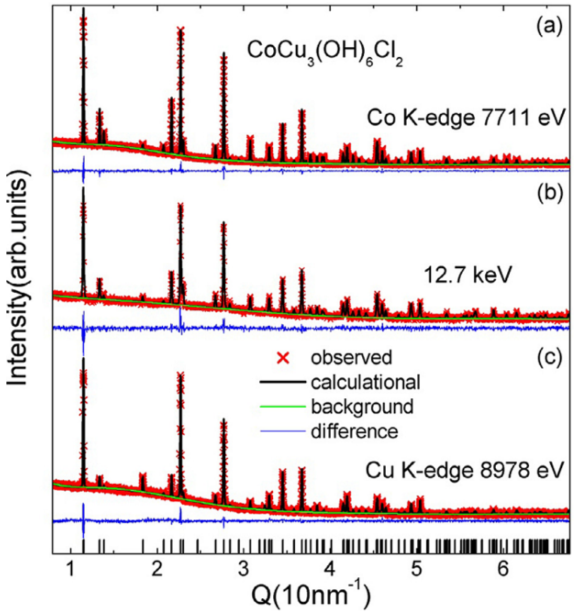
- Gapped spin liquid
- Gap ~ 10 K

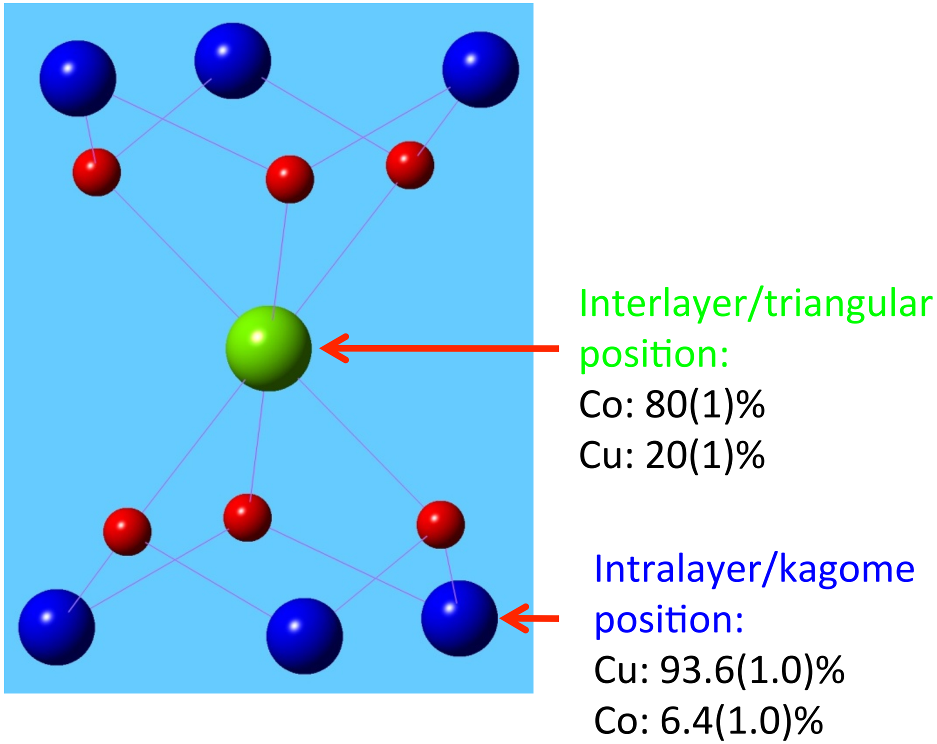


M. Fu et al., Science 350, 655 (2015)

$MCu_3(OH)_6Cl_2$ (M=Zn, Co, Ni)





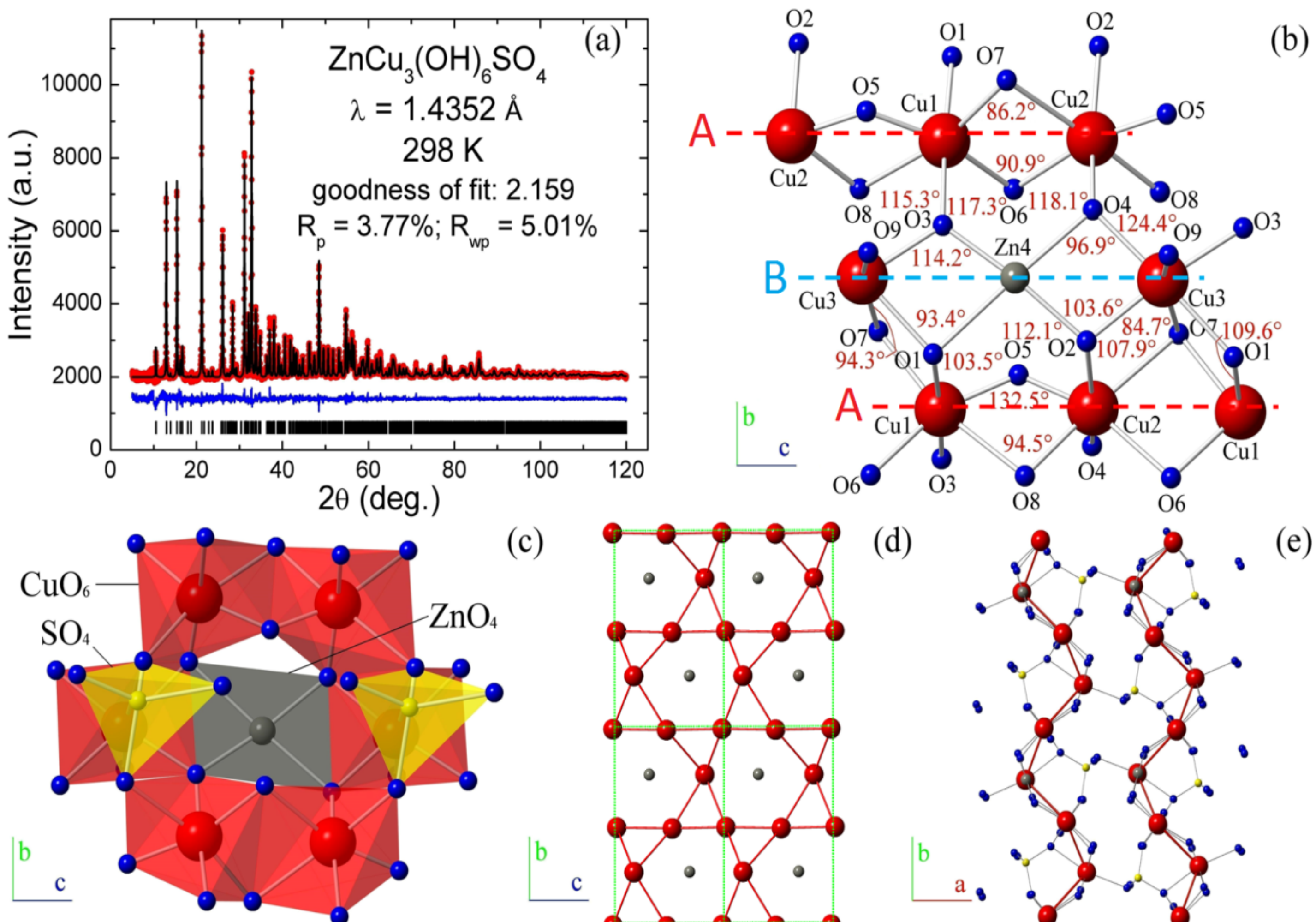


Significant site mixing

Combined structural refinements

Y. Li et al., J. Phys.: Condens. Matter 25, 026003 (2013)
Y. Li et al., Chem. Phys. Lett. 570, 37 (2013)

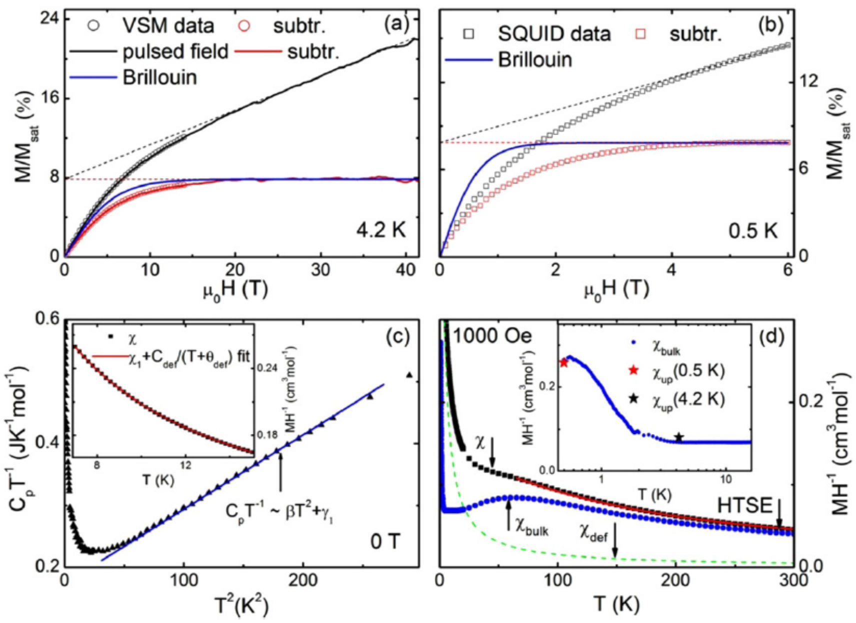
$ZnCu_3(OH)_6SO_4$

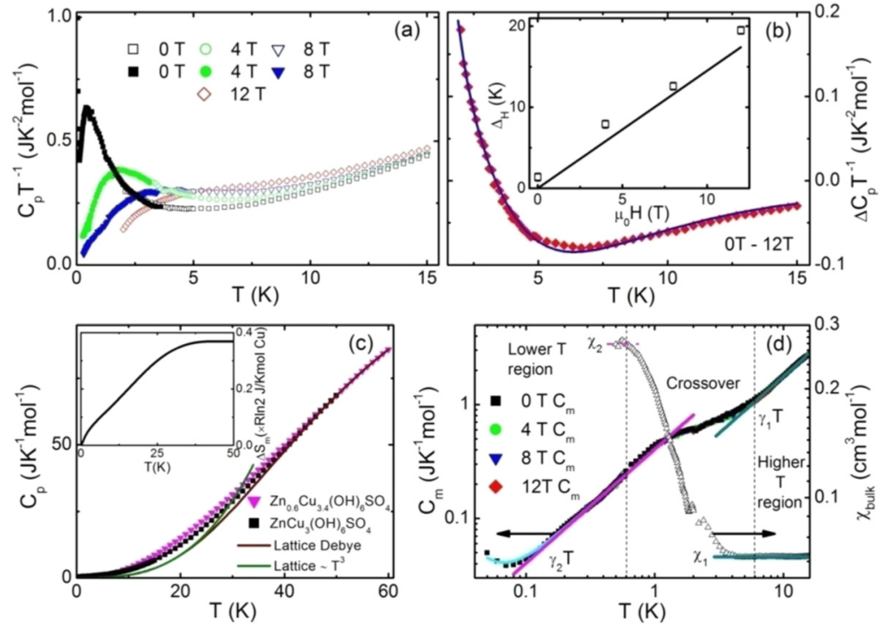


Li et al., New J. of Phys. 16 (2014) 093011

Thermodynamics of $ZnCu_3(OH)_6SO_4$

$Zn_xCu_{4-x}(OH)_6SO_4$	Weiss temperature (K)	transition temperature (K)	frustration factor ($\|\Theta_w\|/T_c$)	Curie constant (Kcm^3/mol)	μ_{eff} (μ_B/Cu)	g
x = 0	–100	7.5	13	22.2	1.88	2.17
x = 0.6	–90	3.5	26	19.5	1.91	2.21
x = 1	–79	<0.05	>1580	17.1	1.90	2.20

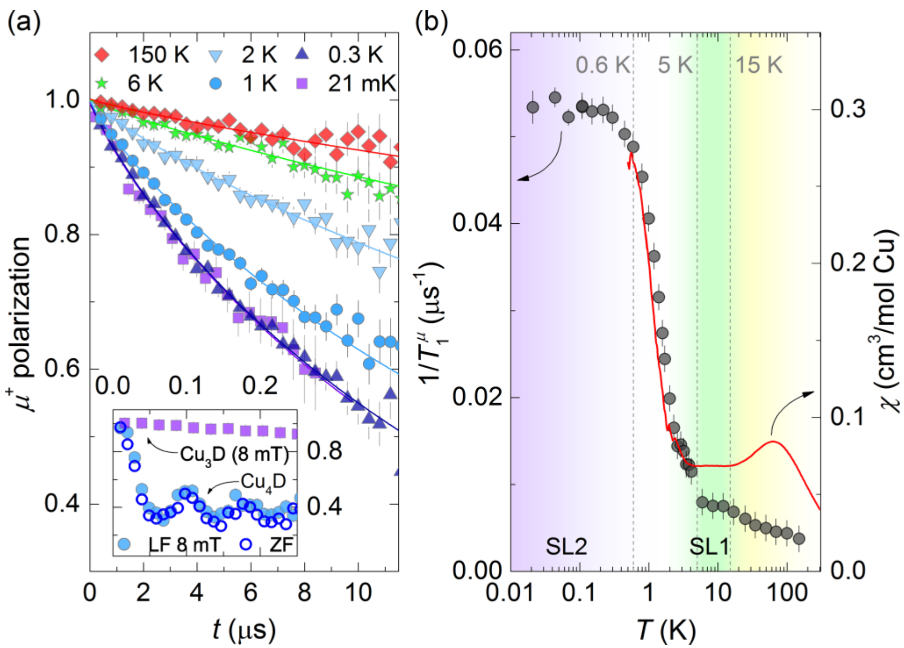




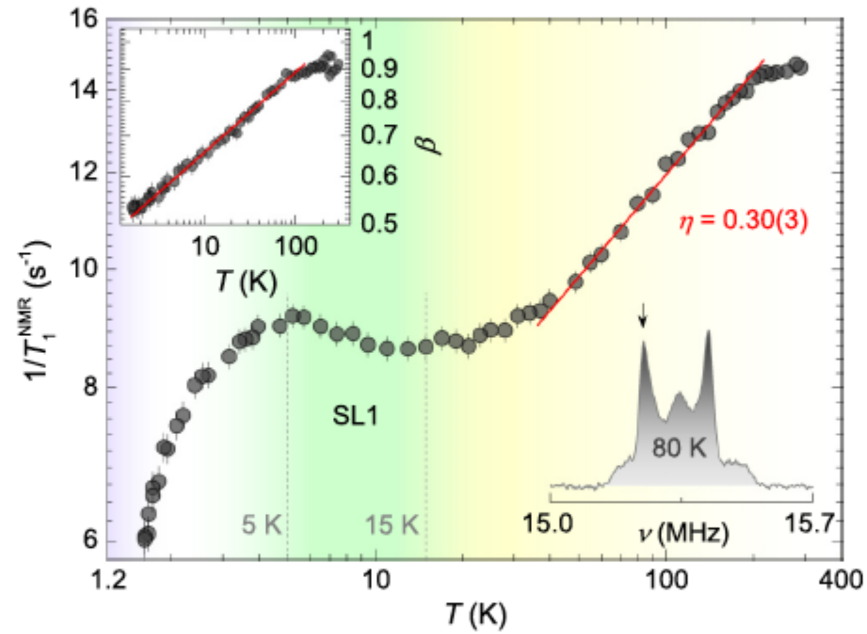
Li et al., New J. of Phys. 16 (2014) 093011

Spin dynamics of $ZnCu_3(OH)_6SO_4$

muSR



NMR



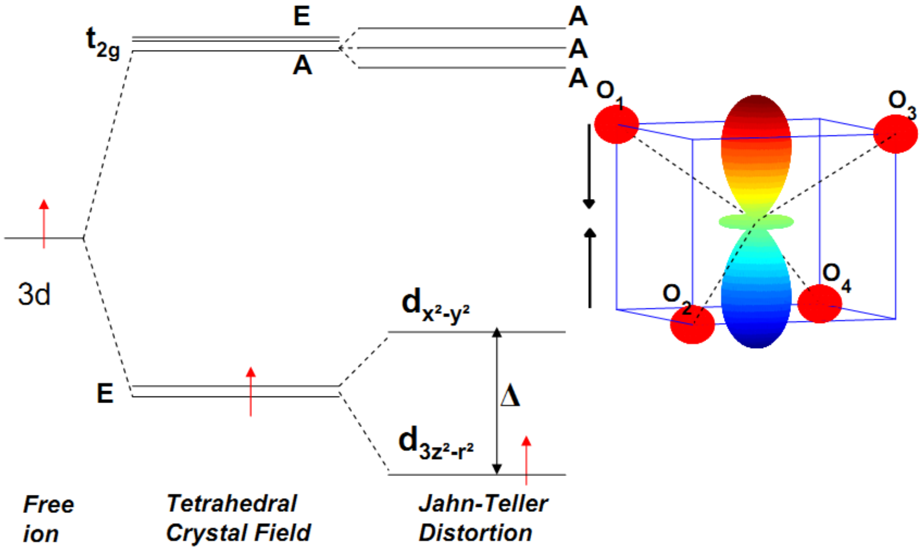
- $ZnCu_3(OD)_6SO_4$
- No oscillation down to 21 mK
- Two regions?

Sign of two regions

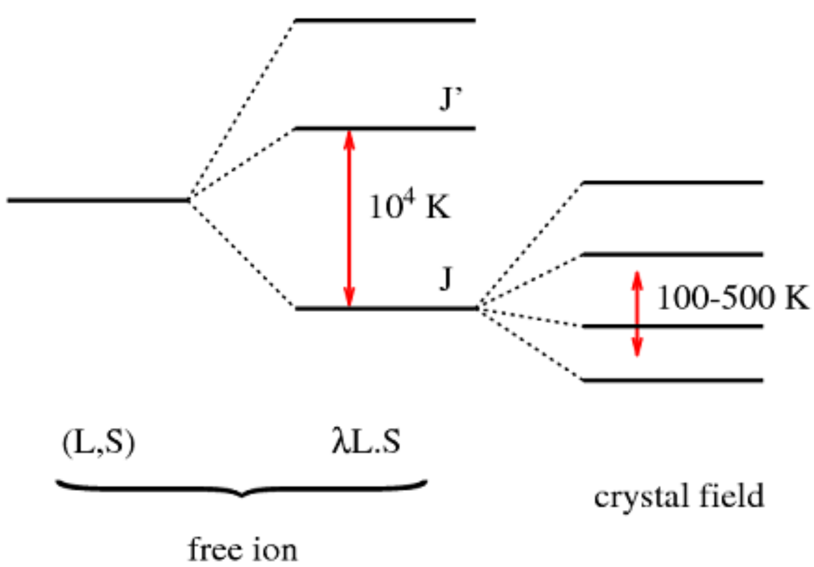
M. Gomilsek et al., Phys. Rev. B 93, 060405 (2016) (Rapid Commun.)
M. Gomilsek et al., Phys. Rev. B 94, 024438 (2016)

3d vs 4f

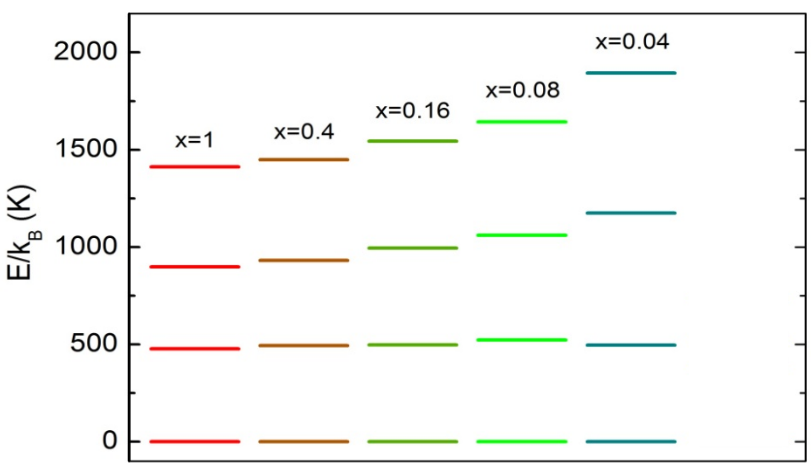
Cu^{2+}



Yb^{3+}

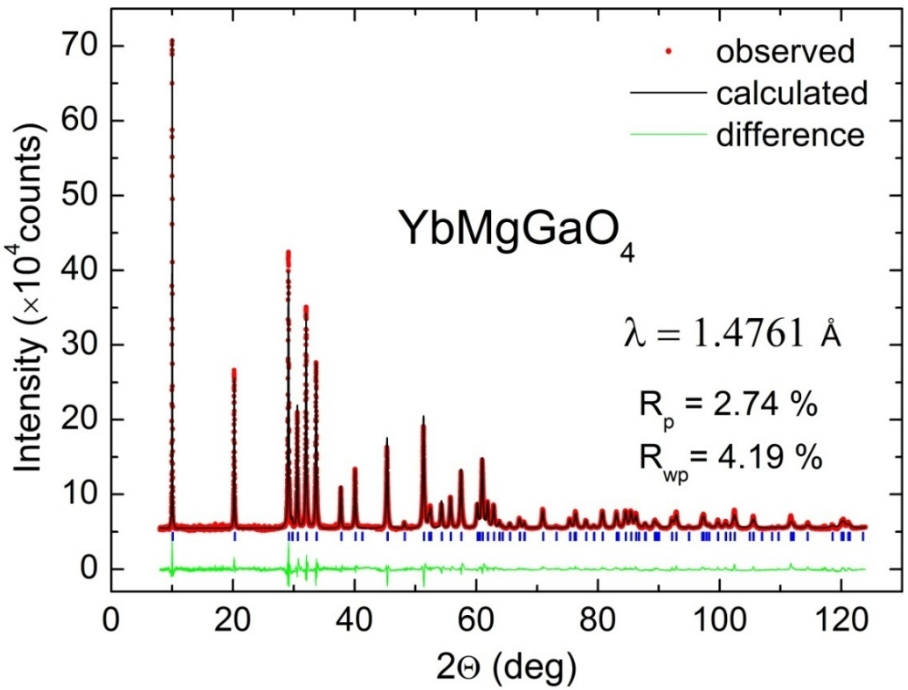


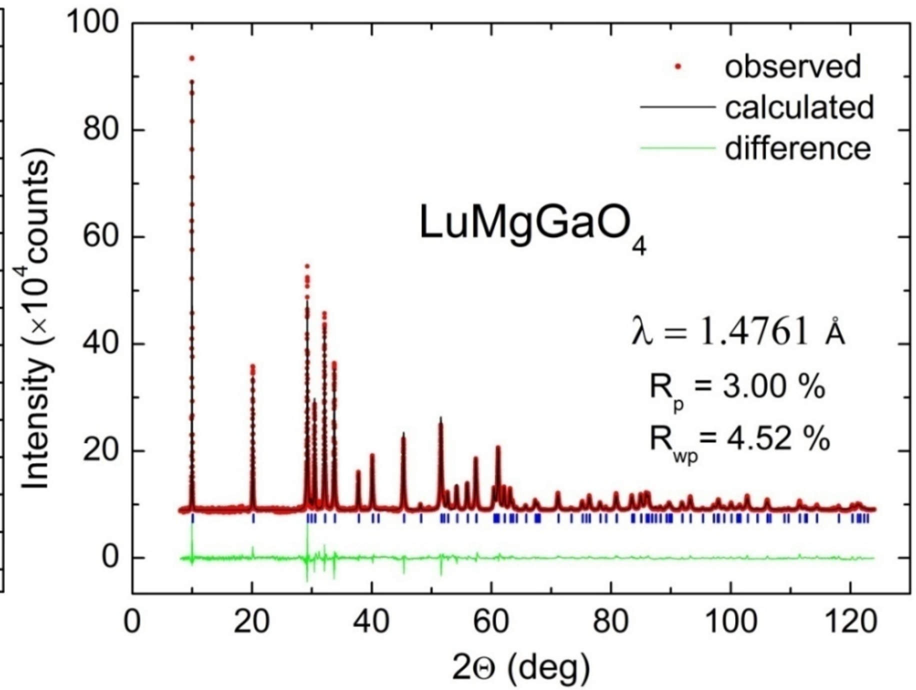
Kramers doublets



$4f^{13}$ L=3, S=1/2, J=7/2

Powder diffraction





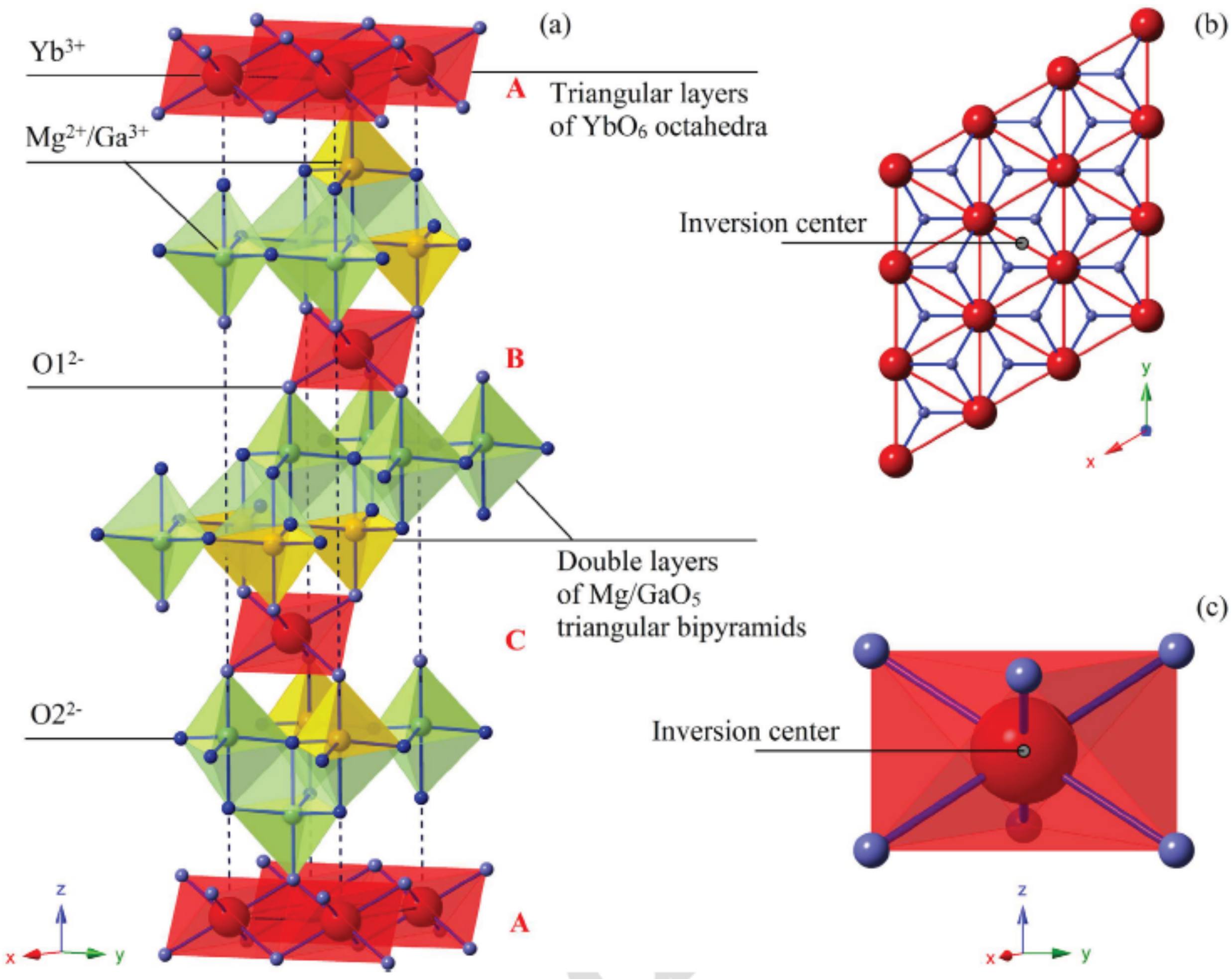
Yb^{3+}: $4f^{13}$, S=1/2
m=173

Lu^{3+}: $4f^{14}$, S=0
m=175

Lanthanides	6	58 **Ce** Cerium 140.115	59 **Pr** Praseodymium 140.9076	60 **Nd** Neodymium 144.24	61 **Pm** Promethium (145)	62 **Sm** Samarium 150.36	63 **Eu** Europium 151.965	64 **Gd** Gadolinium 157.25	65 **Tb** Terbium 158.9253	66 **Dy** Dysprosium 162.50	67 **Ho** Holmium 164.9303	68 **Er** Erbium 167.26	69 **Tm** Thulium 168.9342	70 **Yb** Ytterbium 173.04	71 **Lu** Lutetium 174.967	
Actinides	7	90 **Th** Thorium 232.0381	91 **Pa** Protactinium 231.0359	92 **U** Uranium 238.0289	93 **Np** Neptunium (237)	94 **Pu** Plutonium (244)	95 **Am** Americium (243)	96 **Cm** Curium (247)	97 **Bk** Berkelium (247)	98 **Cf** Californium (251)	99 **Es** Einsteinium (252)	100 **Fm** Fermium (257)	101 **Md** Mendelevium (258)	102 **No** Nobelium (259)	103 **Lr** Lawrencium (260)	7

Li et al., Scientific Reports 5, 16419 (2015)

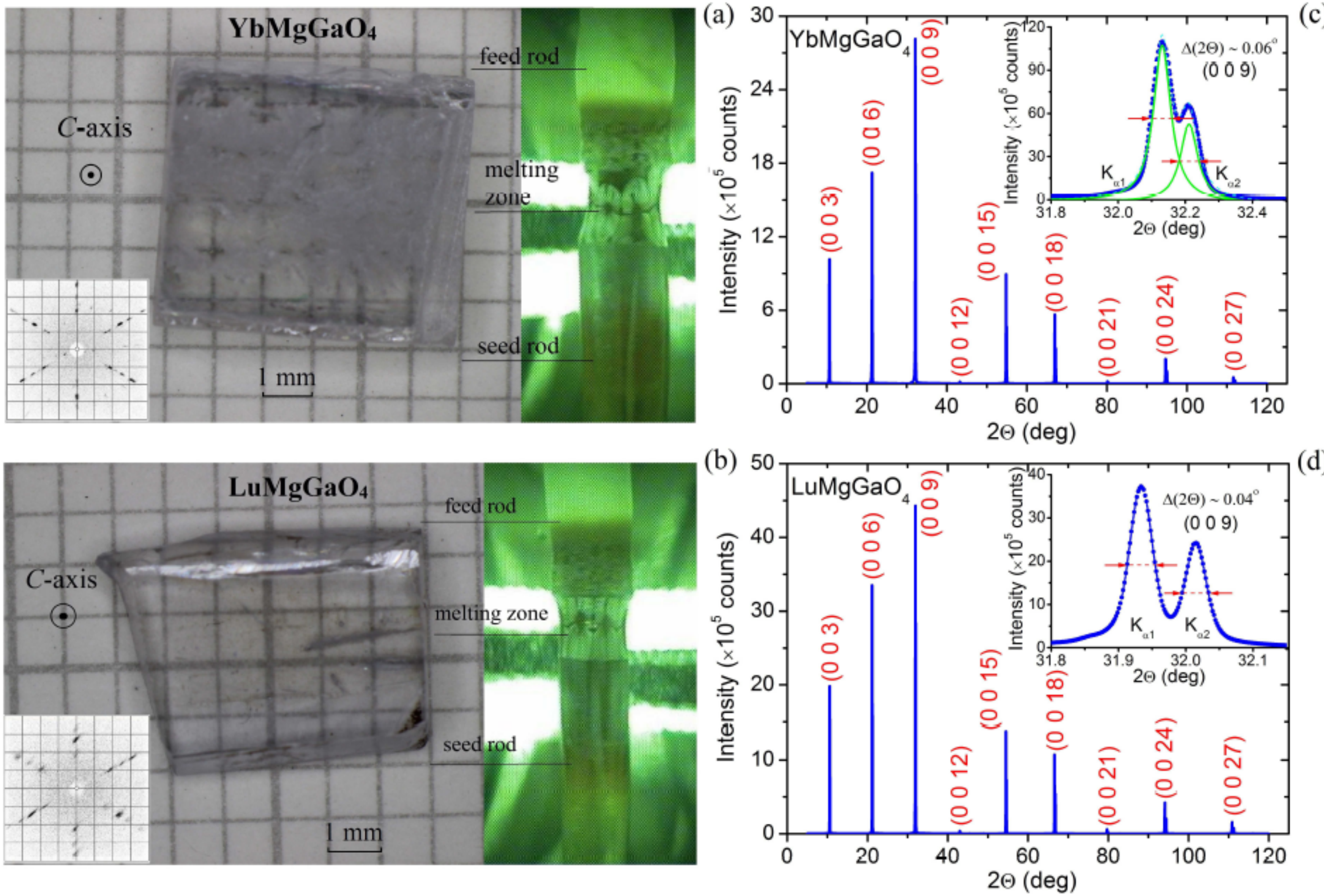
Structurally perfect $YbMgGaO_4$



- Strict R-3m symmetry
- Two-dimensional
- $S_{eff}=1/2$
- No antisite/impurity
- No DM interaction
- Perfect reference sample $LuMgGaO_4$
- High-quality crystals available
- Small J

Li et al., Scientific Reports 5, 16419 (2015)
Li et al., Phys. Rev. Lett. 115, 167203 (2015)

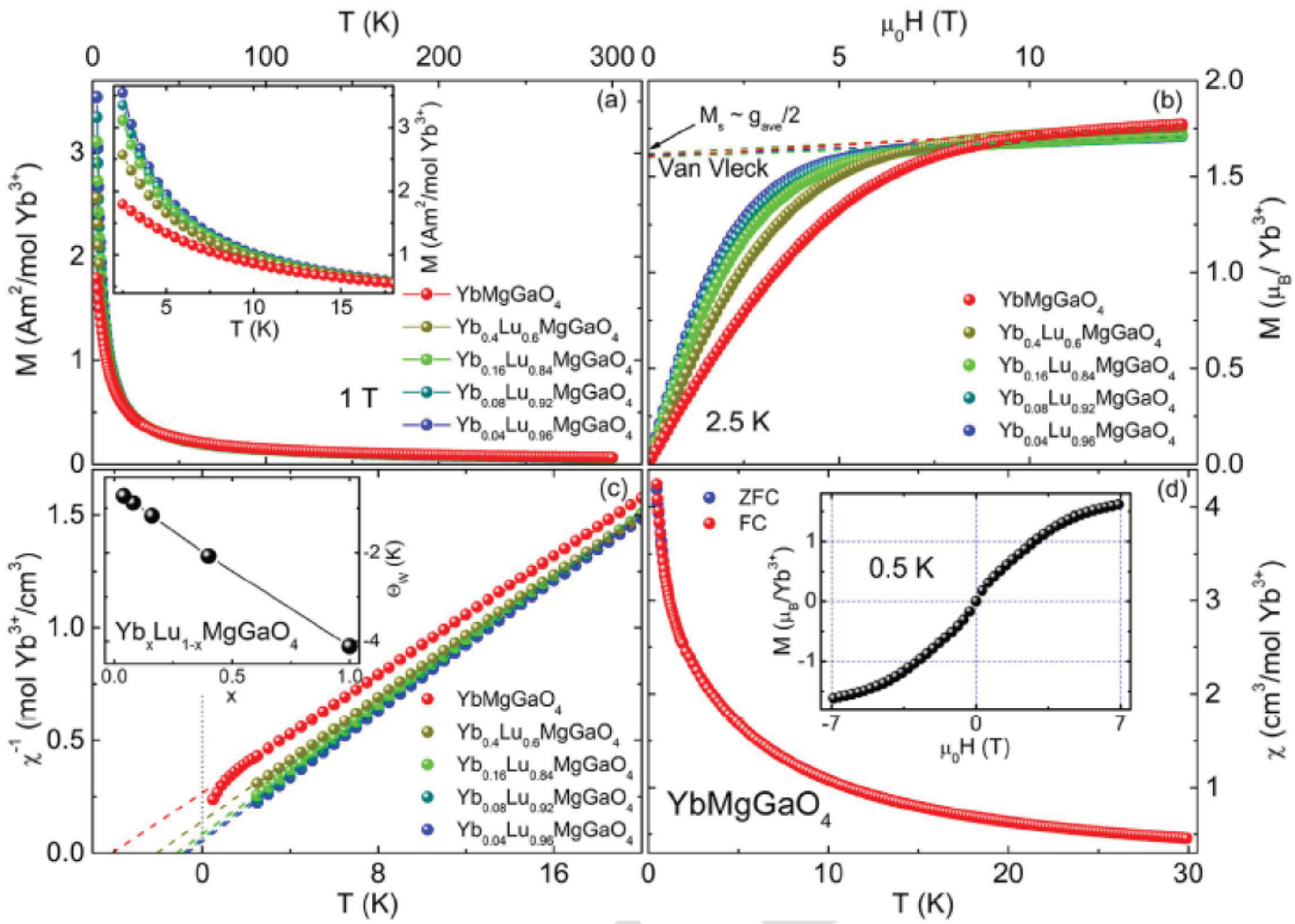
Single Crystals



Charge Gap～ 4 eV

Li et al., Phys. Rev. Lett. 115, 167203 (2015)

Magnetization



Θ_W~ -4 K

Li et al., Scientific Reports 5, 16419 (2015)
Li et al., Phys. Rev. Lett. 115, 167203 (2015)

Spin Hamiltonian

Under R-3m symmetry

$$\mathcal{H} = \sum_{\langle ij \rangle} [J_{zz} S_i^z S_j^z + J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+)$$

Anisotropic terms

$$+ J_{\pm\pm}(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-)$$

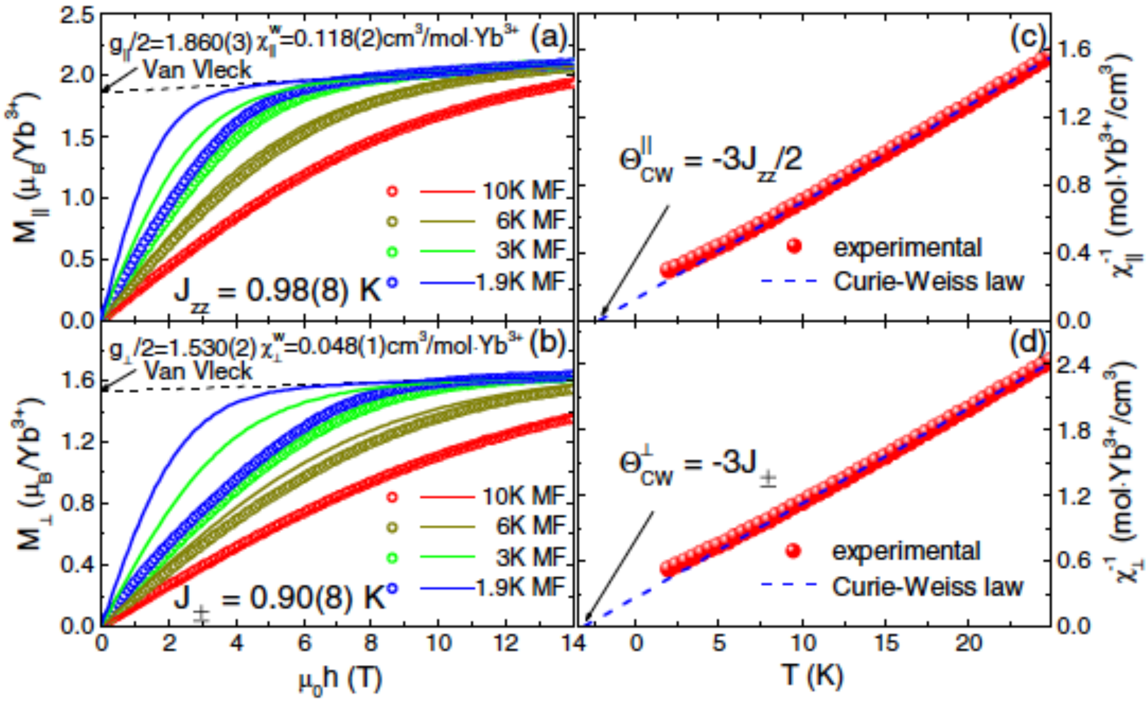
$$- \frac{iJ_{z\pm}}{2}(\gamma_{ij}^* S_i^+ S_j^z - \gamma_{ij} S_i^- S_j^z + \langle i \leftrightarrow j \rangle)],$$

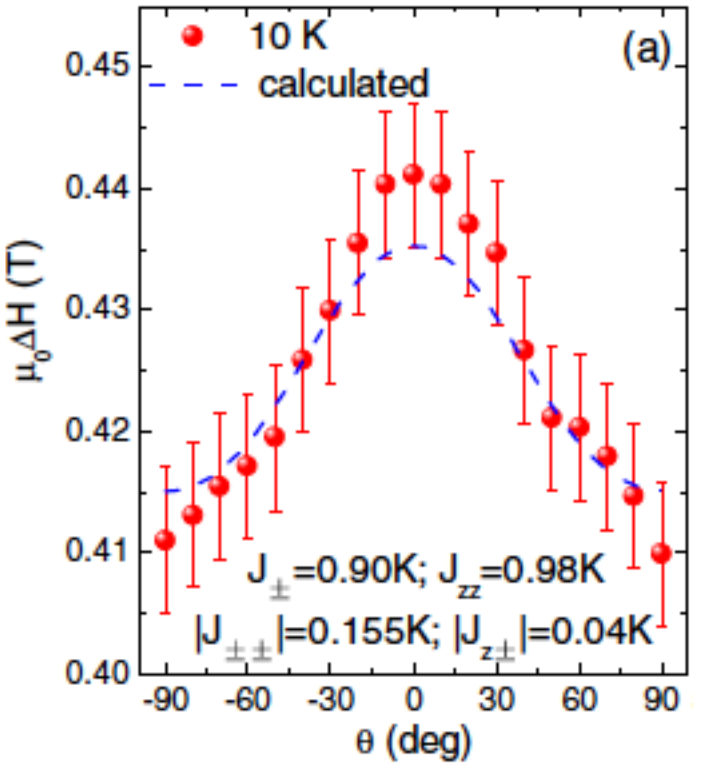
κ-$(ET)_2Cu_2(CN)_3$ $\quad$ $EtMe_3Sb[Pd(dmit)_2]_2$

Strong charge fluctuations → Ring exchange

Li et al., Phys. Rev. Lett. 115, 167203 (2015)

Exchange parameters

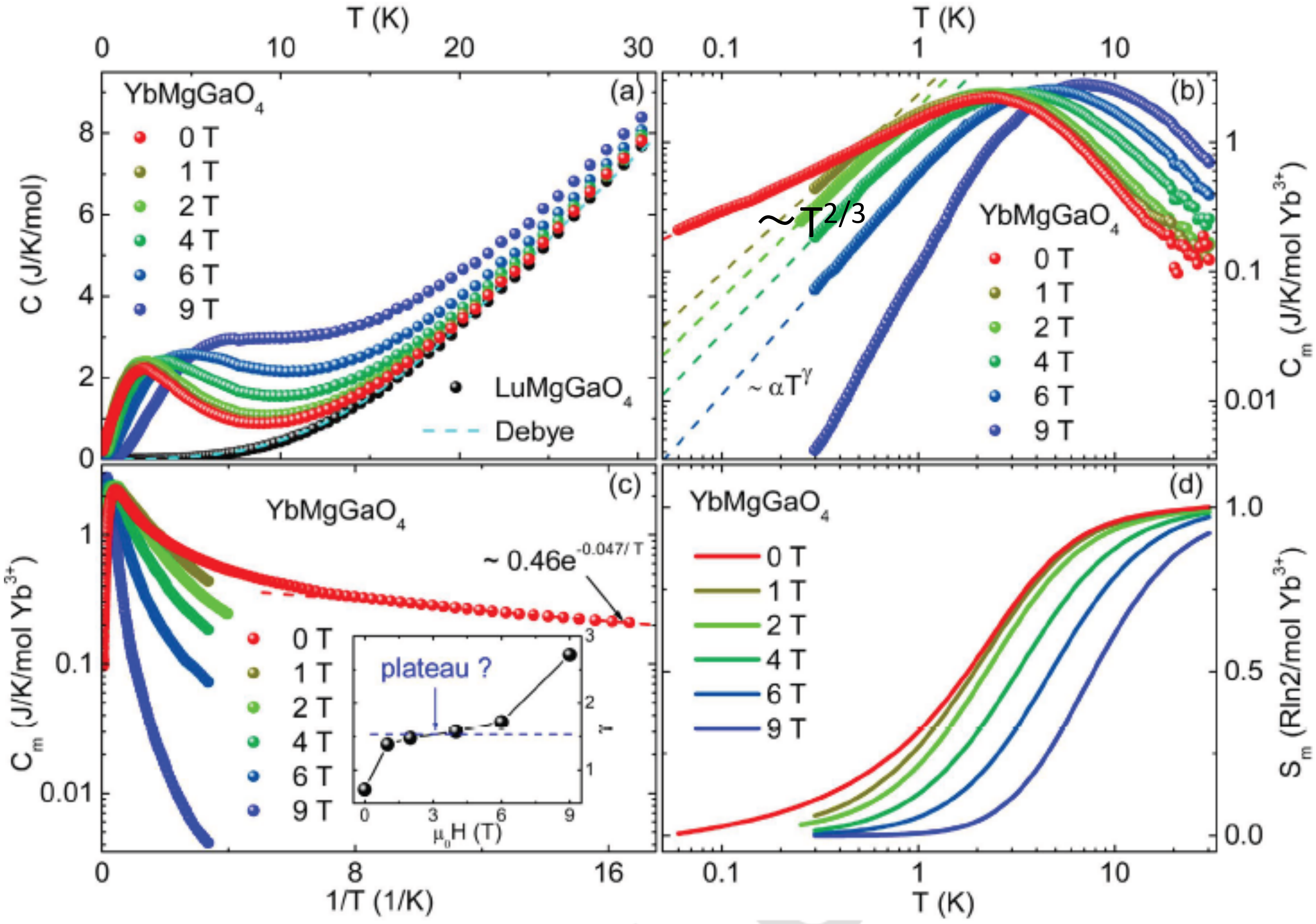




Li et al., Phys. Rev. Lett. 115, 167203 (2015)

Specific Heat

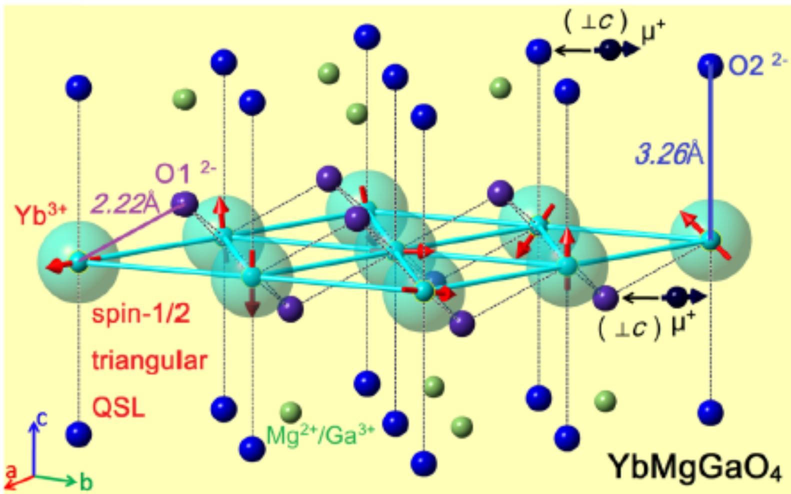
m_{Yb}=173
m_{Lu}=175

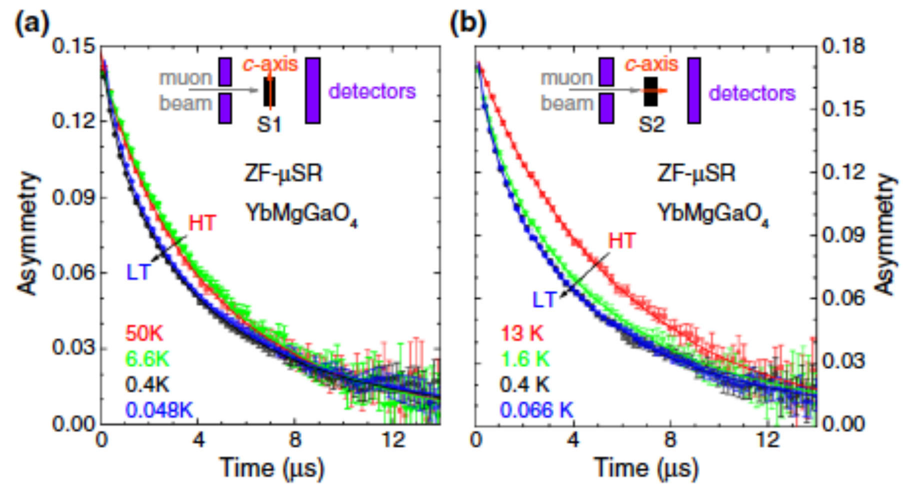


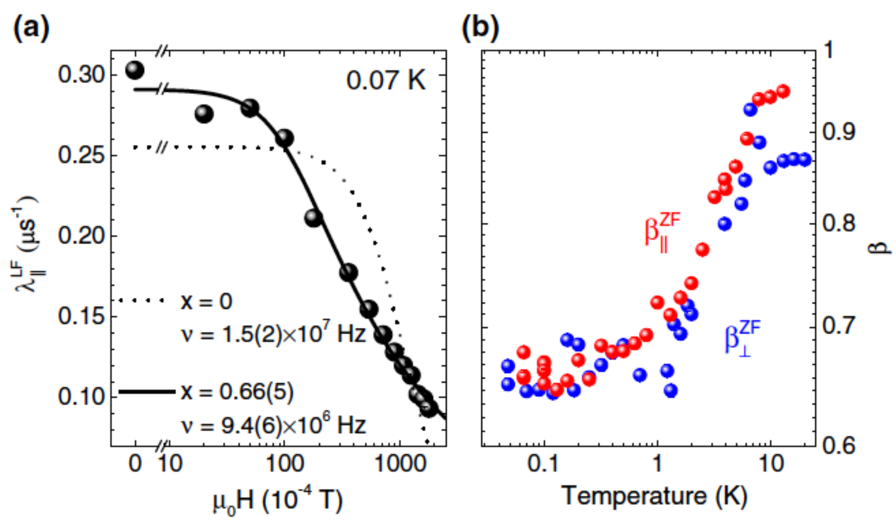
- No magnetic ordering down to 50 mK
- Accurate spin entropy
- Zero-entropy spin ground state (residual spin entropy below 50 mK < 0.6%)

Li et al., Phys. Rev. Lett. 115, 167203 (2015)

U(1) quantum spin liquid ground state



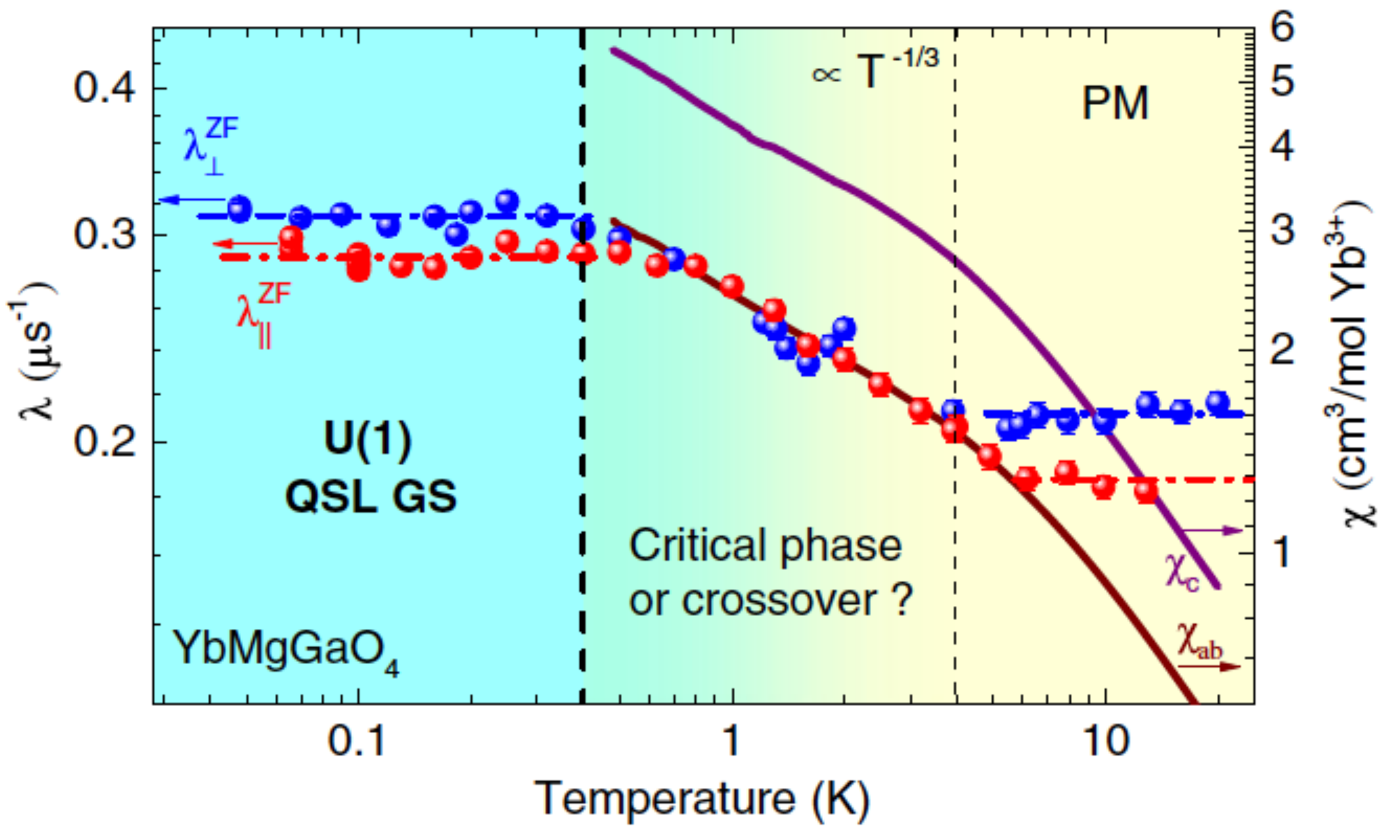




Rule out long-range magnetic ordering and spin freezing

Li et al., Phys. Rev. Lett. 117, 097201 (2016)

U(1) quantum spin liquid ground state

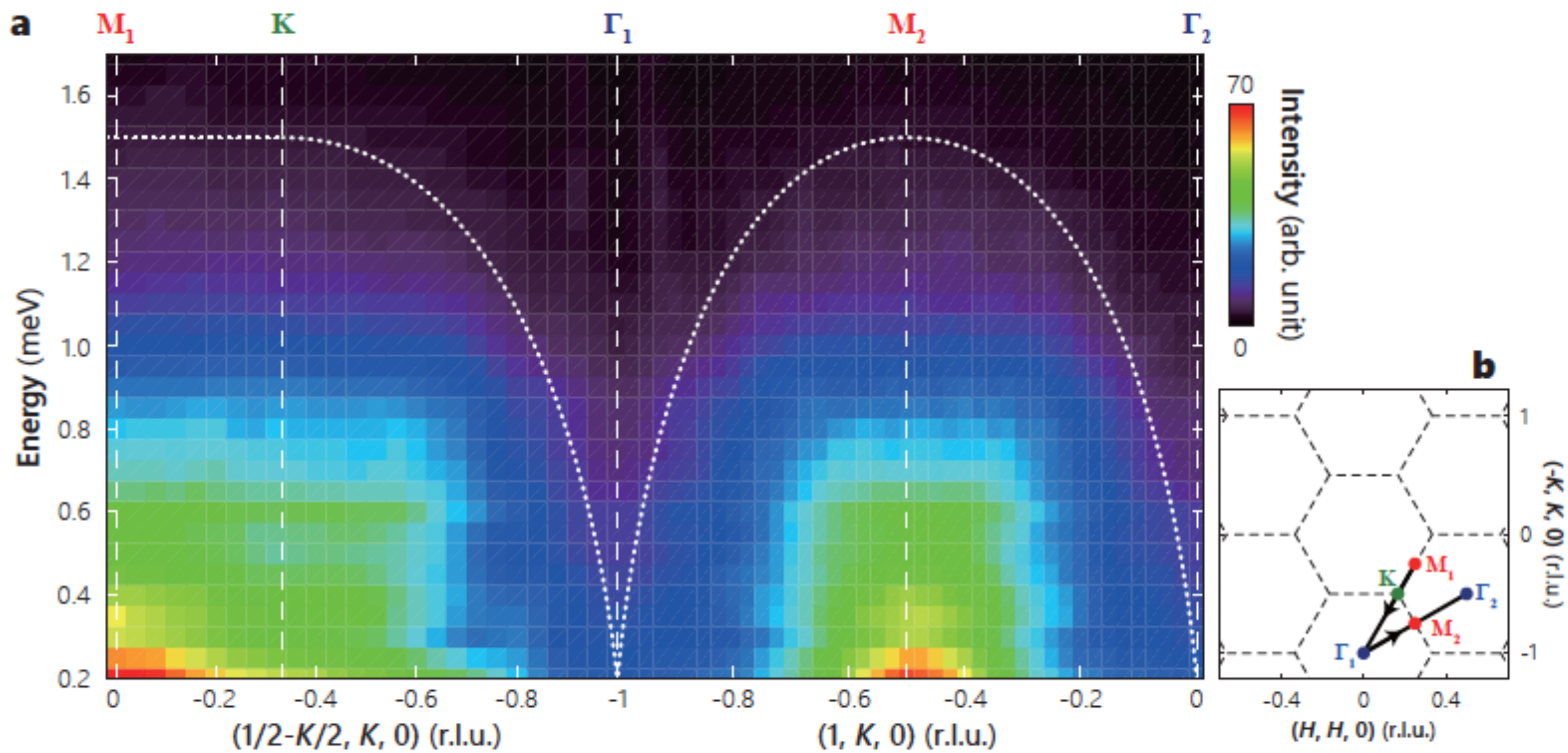


Zero spin entropy: spin ground state
$C_v \sim T^{2/3}$
Constant Muon spin relaxation rate

U(1) gapless quantum spin liquid

Li et al., Phys. Rev. Lett. 117, 097201 (2016)

Spin excitations



- Diffusive spin excitations
- Spinon Fermi surface
- Gapless U(1) quantum spin liquid

Y. Shen et al., Nature 540, 559-562 (2016)

Summary

- A new two-dimensional triangular compound $YbMgGaO_4$ with $S_{eff}=1/2$ and high-quality single crystals available
- Anisotropic spin Hamiltonian
- Gapless U(1) quantum spin liquid ground state
- Strong spin-orbit coupling may play a key role in forming QSL ground state and our work may inspire a new route to search for QSL materials