Realization of a Quantum Spin Liquid in a bilayer Kagome lattice $Ca_{10}Cr_7O_{28}$

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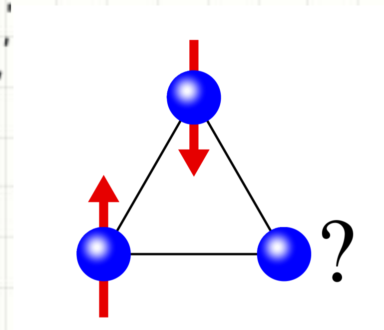
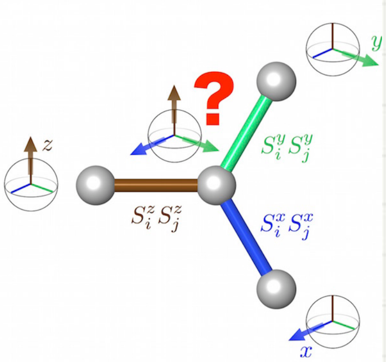
IISER Mohali





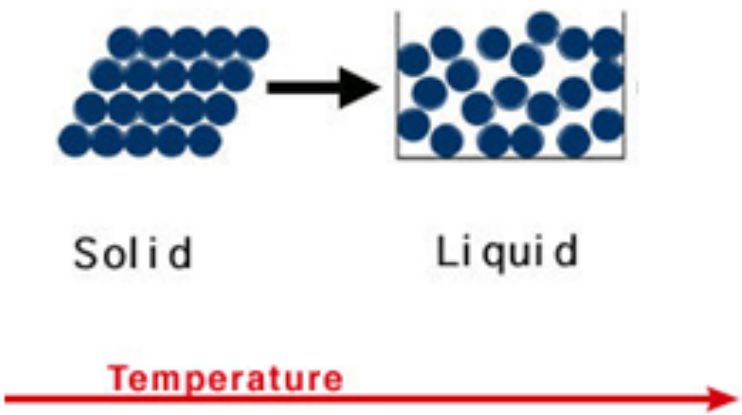
OUTLINE

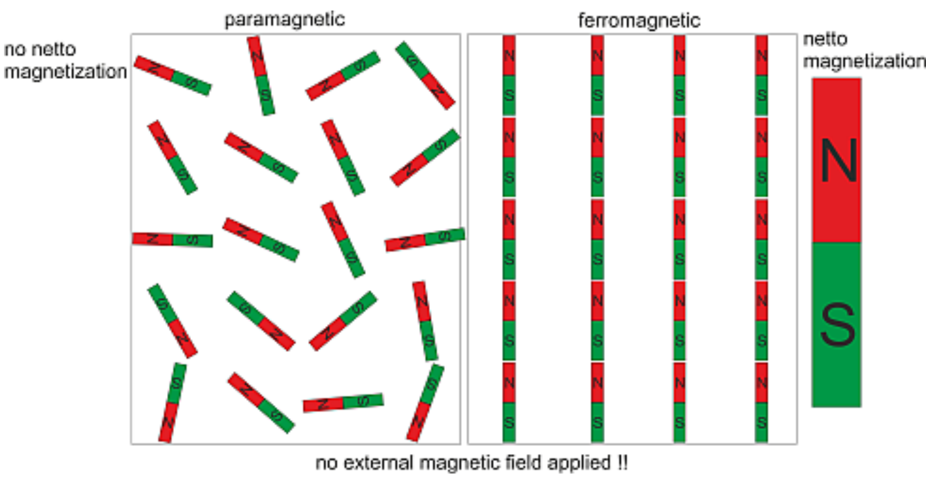
- Spin Liquids : Geometrical Frustration, No LRO, Fractionalization, Spinons,….
- $Ca_{10}Cr_7O_{28}$, a new complex oxide
- Thermodynamic Measurements: Magnetism, Heat Capacity, AC chi
- Microscopic Measurements: μSR
- Magnetic Excitations: Inelastic neutron scattering
- The Hamiltonian
- FRG: SL ground state

Collaborators

C. Balz, A. T. M. N. Islam, Bella Lake (*Helmholtz Zentrum Berlin*)	Crystal Growth, INS, low temp. thermodynamics
Chris Baines and Hubertus Luetkens (*Paul Scherrer Institut, Switzerland*)	μSR
Elisa M. Wheeler (ILL, Grenoble, France)	INS
J. A. Rodriguez-Rivera, T. Guidi, G. G. Simeoni (NIST, ISIS, TU Muenchen)	INS
Rico Schoenemann, Thomas Herrmannsdoerfer (*Helmholtz-Zentrum Dresden*)	AC Chi
Hanjo Ryll (*Helmholtz Zentrum Berlin*)	Low T heat capacity
J. Reuther (*Freie Universita¨t Berlin, Germany*)	FRG calculations

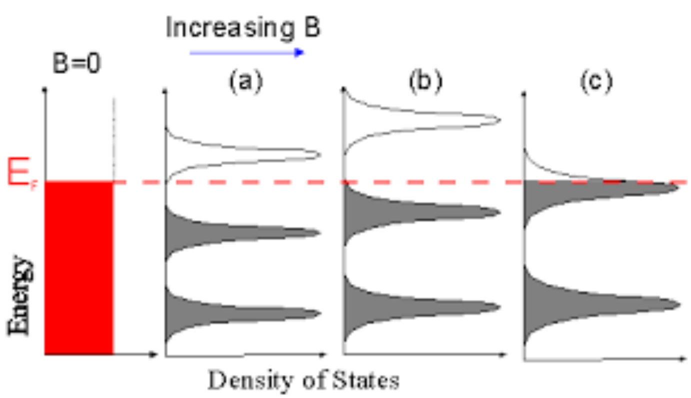
Broken Symmetry -----> Phases/particles

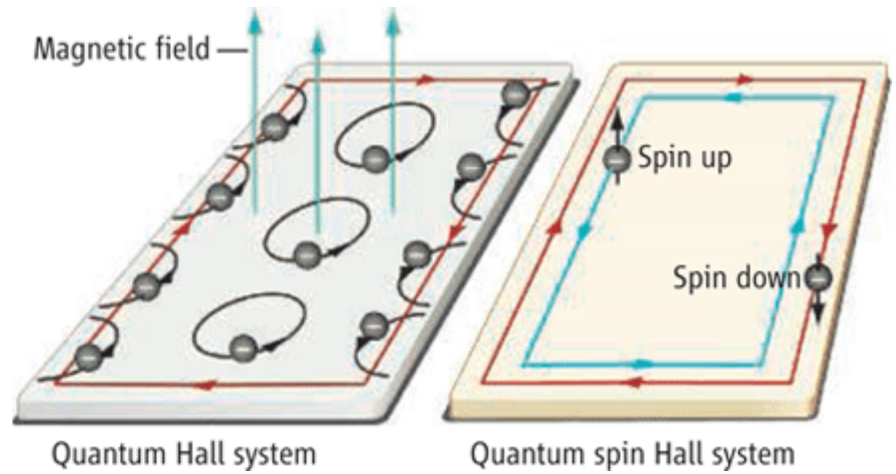


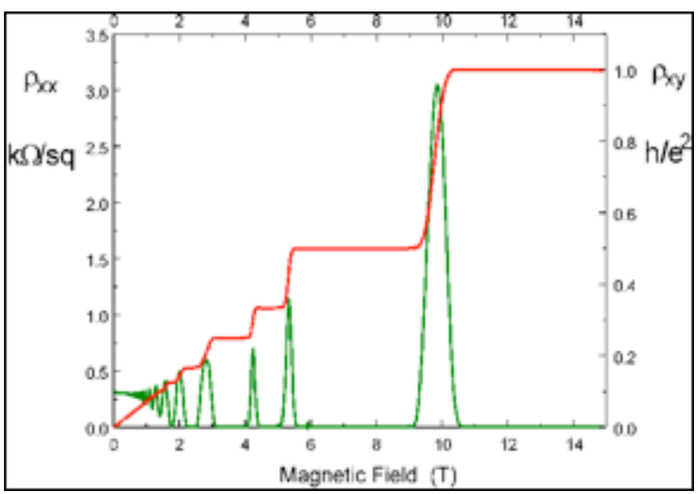


Can one have phases with no broken symmetry or no order parameter ??

Topological States: Quantum Hall Effect







$$\sigma_{xy} = Ne^2/h.$$

σ has been measured to 1 part in 10^9

$$N = n_m = \frac{1}{2\pi}\int d^2\mathbf{k}\,\mathcal{F}_m$$

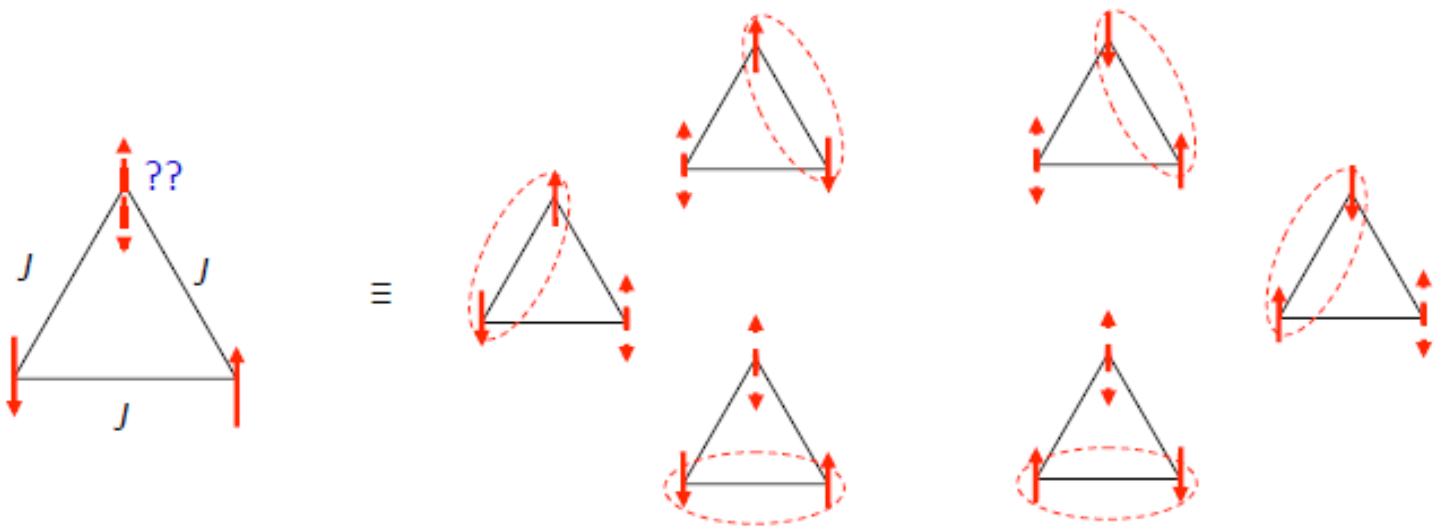
Berry flux $F_m = \nabla \times \boldsymbol{A}_m$

n_m = Chern #

This precision is a manifestation of the topological nature of σ_{xy}

Spin Liquids in Geometrically Frustrated Magnets

Huge degeneracy



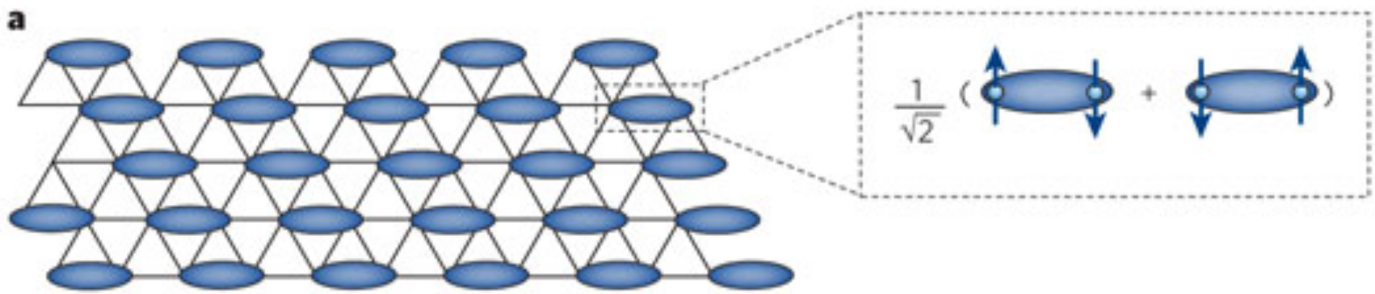
Spin–liquid: Quantum Disordered State of Strongly Interacting Spins

No "obvious" Order Parameter or Broken Symmetry

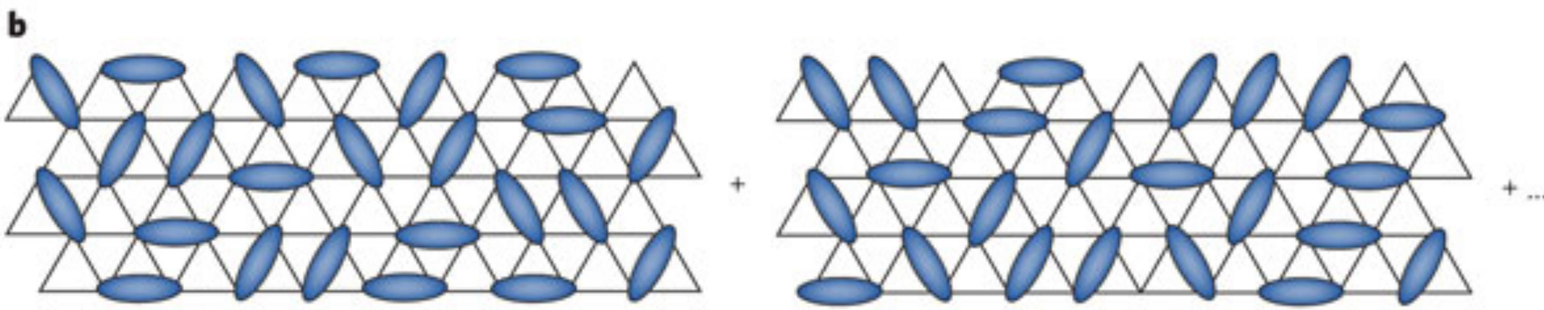
RVB state on a triangular lattice is a Z2 spin liquid

Anderson’s Valence Bond States

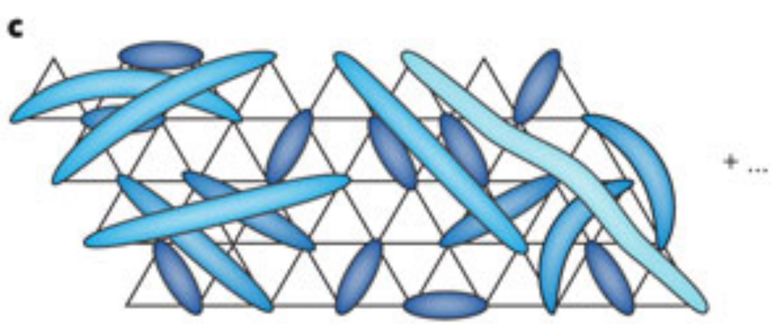
Valence Bond Solid



Resonating Valence Bond : Short Range



Resonating Valence Bond : Long Range

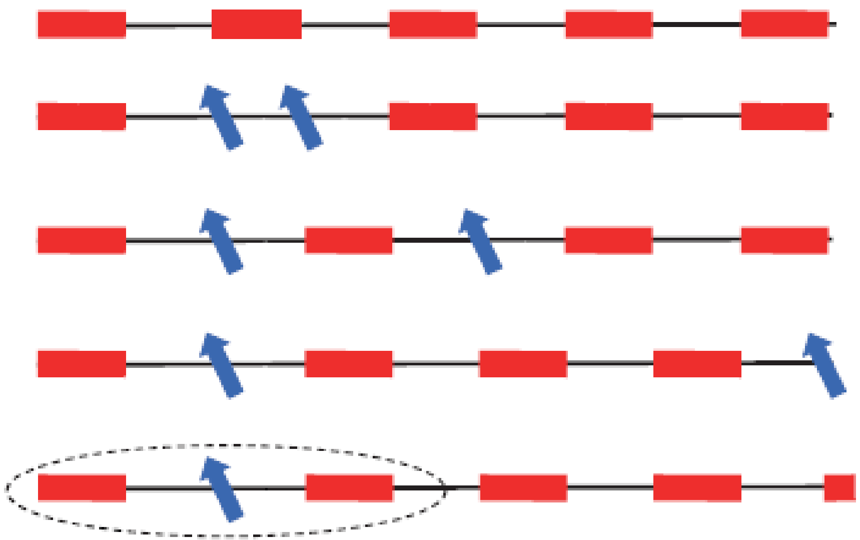


L. Balents, “Spin liquids in frustrated magnets,” Nature (2010).

Excitations in spin liquids: Fractional Particles

Federico Becca ('14)

- Majumdar-Gosh chain (1D): $\mathcal{H} = J \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} + \frac{J}{2} \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2}$
- The exact ground state is known (two-fold degenerate), perfect dimerization



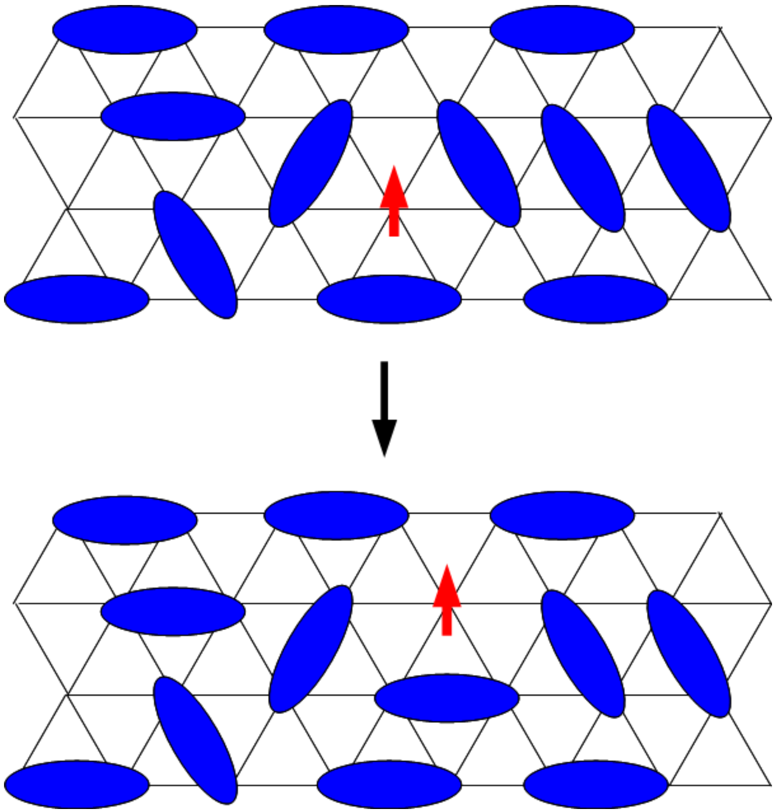
The "initial" $S = 1$ excitation can decay into two spatially separated spin-1/2 excitations (spinons)

Finite-energy state with an isolated spinon (the other is far apart) domain wall between two dimerization patterns

- A spinon is a neutral spin-1/2 excitation, "one-half" of a $S = 1$ spin flip.
  (it has the same spin as the electron, but no charge)
- Spinons can only be created by pairs in finite systems
  In one dimension, they can propagate at large distances, as two elementary particles

Excitations in spin liquids: Fractional Particles

In 2D a spinon is any unpaired spin which can move around the lattice "delocalize" with little or no energy cost



Experimental Signatures

ρ = insulating

C/T = metallic

χ = metallic

κ/T = metallic

Recipe for Quantum Fluctuations

- Small Spin : S = ½
- Low Dimensionality or co-ordination
- Geometric Frustration: mostly AF on triangular motifs

Most Spin Liquid Candidates are S = ½ Quasi-Low Dimensional Magnets with AF exchange

Some Examples

Table 1 | Some experimental materials studied in the search for QSLs

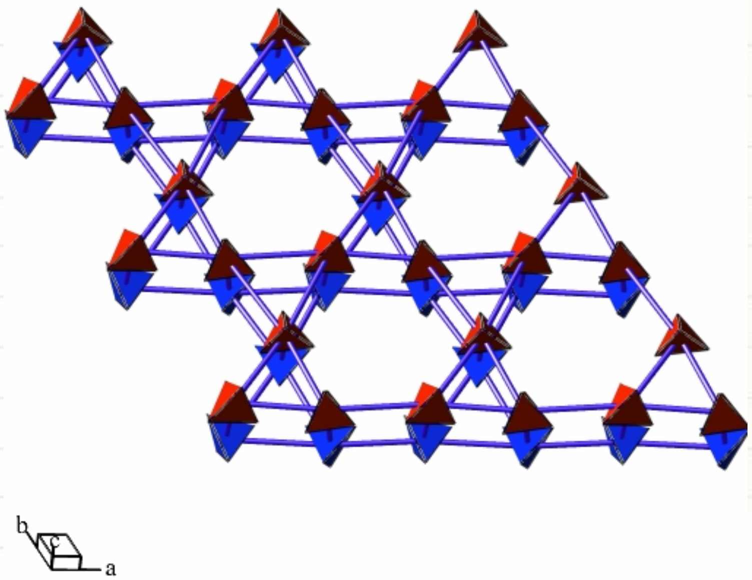
Material	Lattice	S	Θ_{CW} (K)	R^*	Status or explanation
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$	Triangular†	½	−375‡	1.8	Possible QSL
EtMe$_3$Sb[Pd(dmit)$_2$]$_2$	Triangular†	½	−(375–325)‡	?	Possible QSL
Cu$_3$V$_2$O$_7$(OH)$_2$•2H$_2$O (volborthite)	Kagomé†	½	−115	6	Magnetic
ZnCu$_3$(OH)$_6$Cl$_2$ (herbertsmithite)	Kagomé	½	−241	?	Possible QSL
BaCu$_3$V$_2$O$_8$(OH)$_2$ (vesignieite)	Kagomé†	½	−77	4	Possible QSL
Na$_4$Ir$_3$O$_8$	Hyperkagomé	½	−650	70	Possible QSL
Cs$_2$CuCl$_4$	Triangular†	½	−4	0	Dimensional reduction
FeSc$_2$S$_4$	Diamond	2	−45	230	Quantum criticality

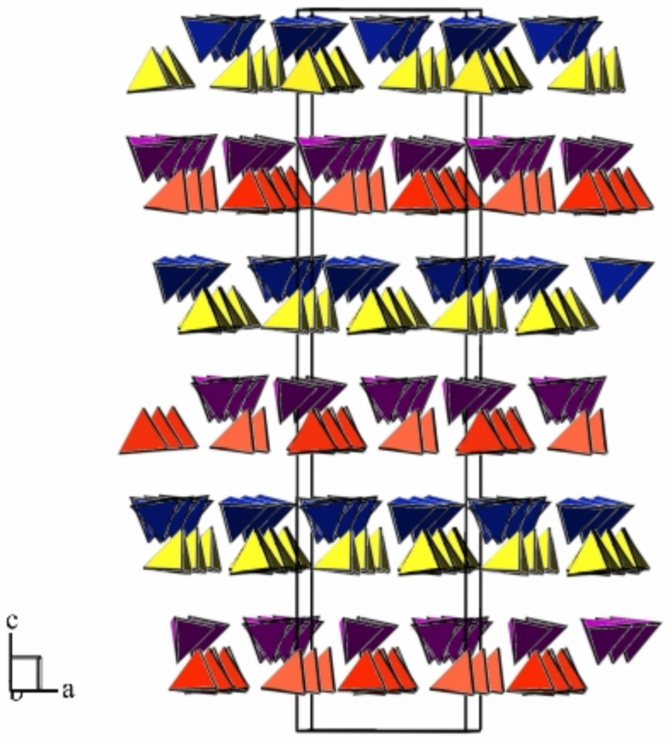
L. Balents, "Spin liquids in frustrated magnets," Nature (2010).

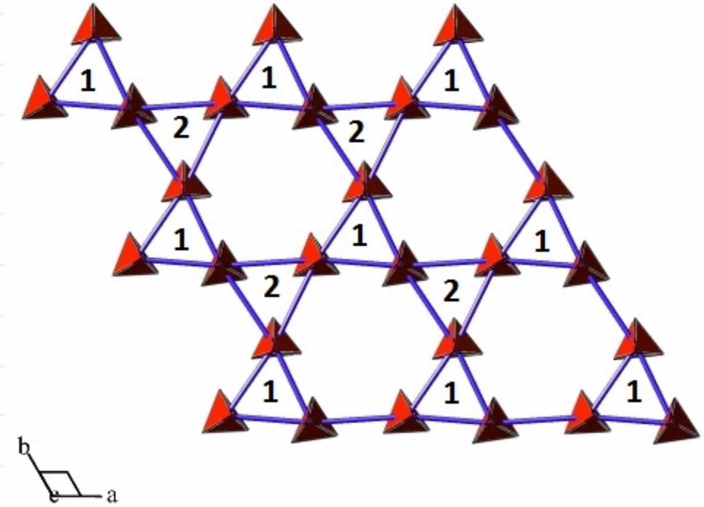
New Quantum Magnet $Ca_{10}Cr_7O_{28}$

Crystal Structure of $Ca_{10}Cr_7O_{28}$

Lattice Parameters		Space Group
a	10.76892(3) Å	$R3c$
c	38.09646(12) Å	

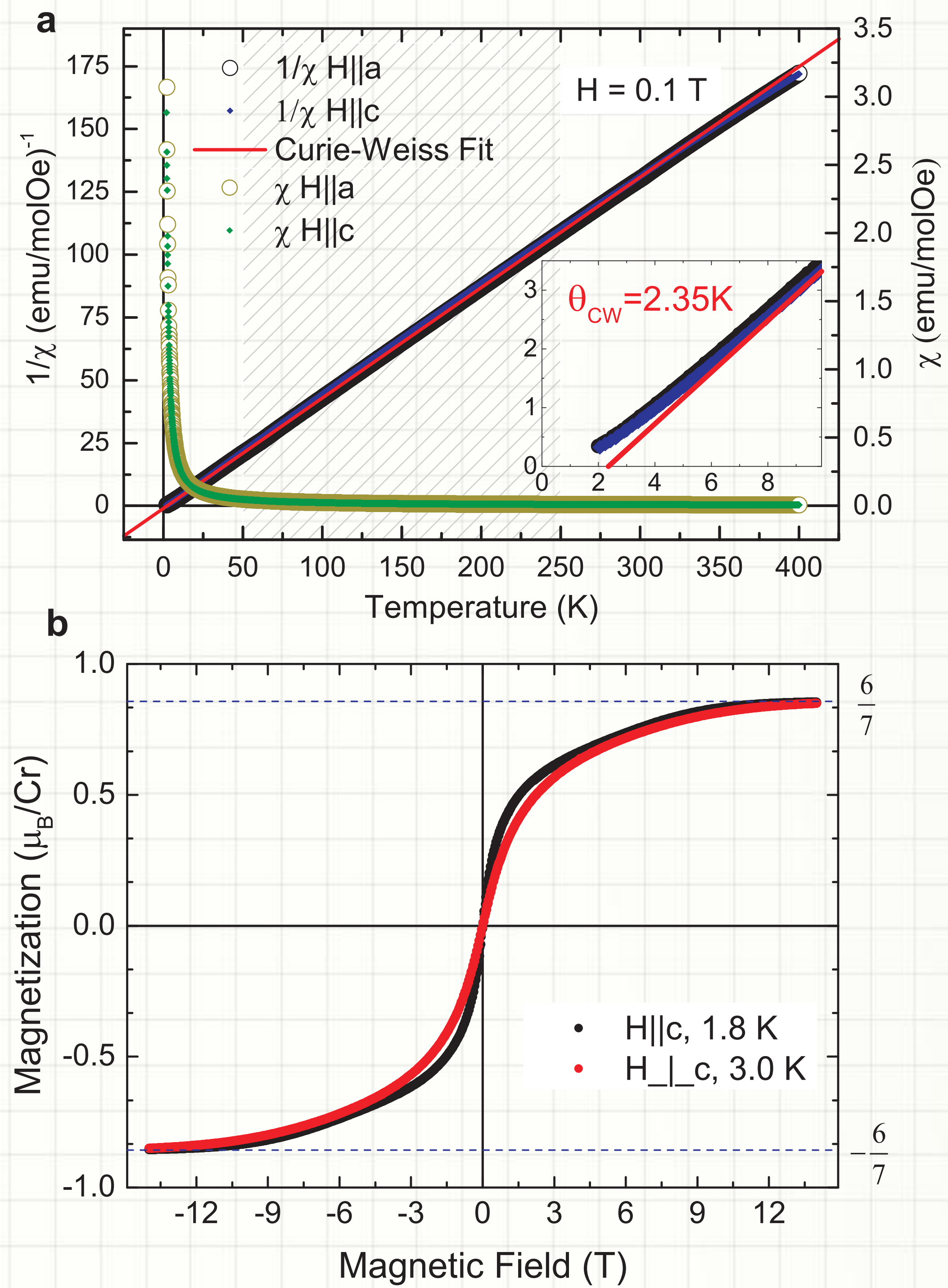




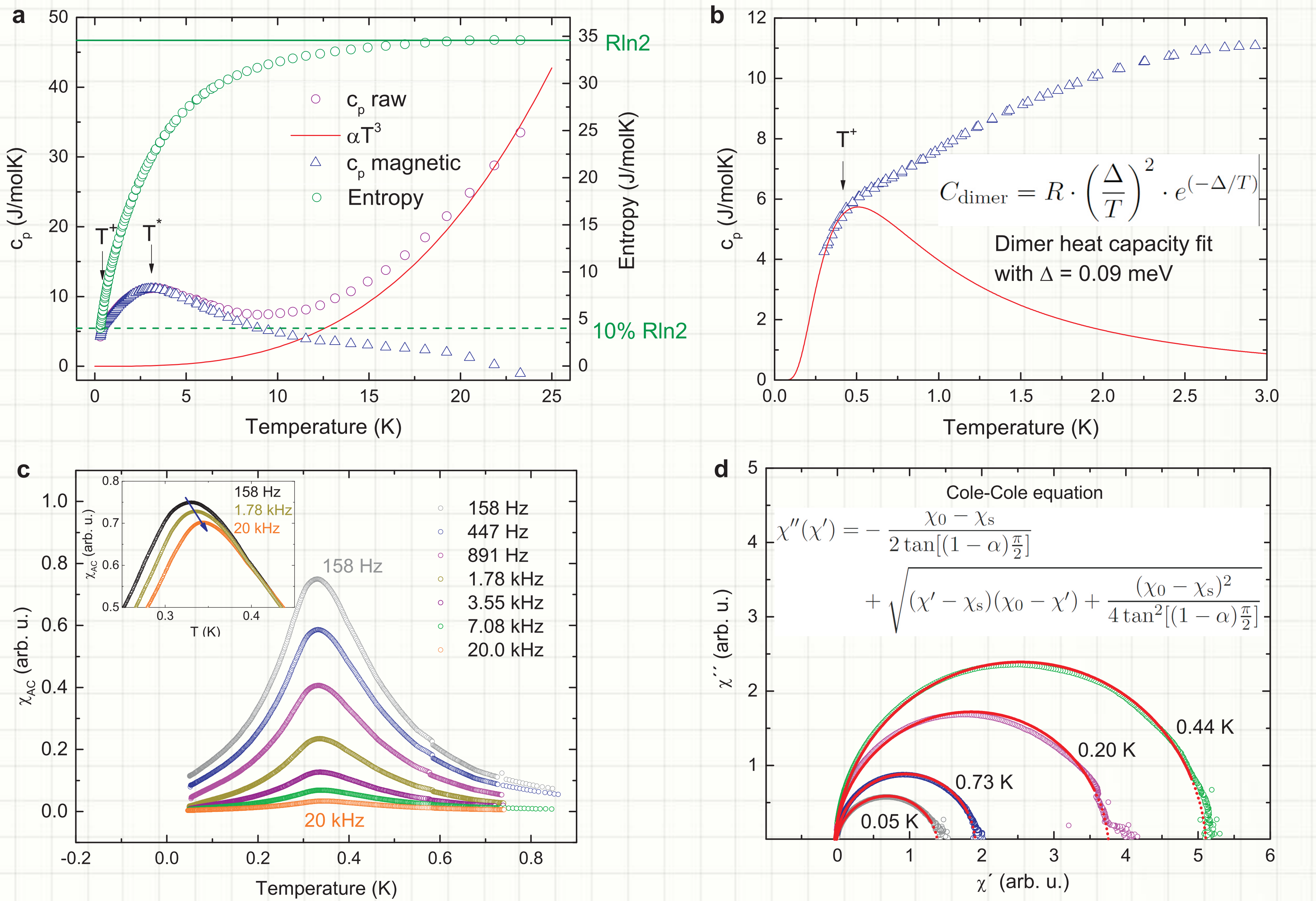


Magnetic Susceptibility + Magnetization

- Local Moment behavior down to 1.8 K with μ_{eff} = 1.49μ_B = 6/7 x 1.74(2) μ_B
- θ = 2.35 K. No LRO.
- No anisotropy between $\chi_{||}$ and χ_{perp}
- Deviations below ~ 5K of χ from CW fit suggest AF
- Magnetization saturates at 12T (= 17 K) to 6/7 μ_B = 6 Cr^{5+} (S = ½) and 1 Cr^{6+} (non-magnetic)



No LRO : Heat Capacity and AC chi



Other Examples of Cole-Cole plot analysis

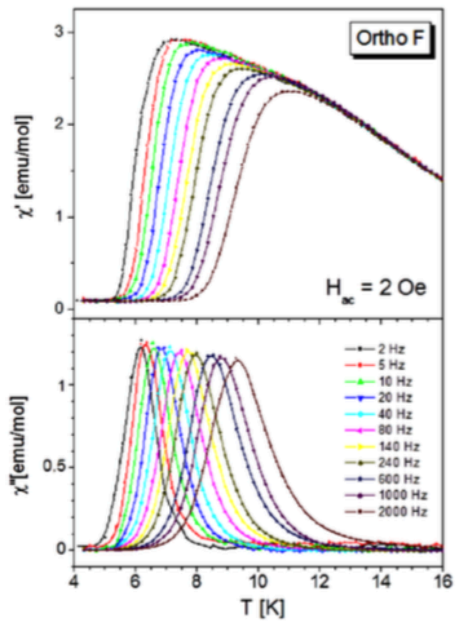


Fig. 12. Frequency dependent AC susceptibility of the [MnF$_4$TPP][TCNE] single chain magnet; fluorine is substituted to porphyrin in the *ortho* position.

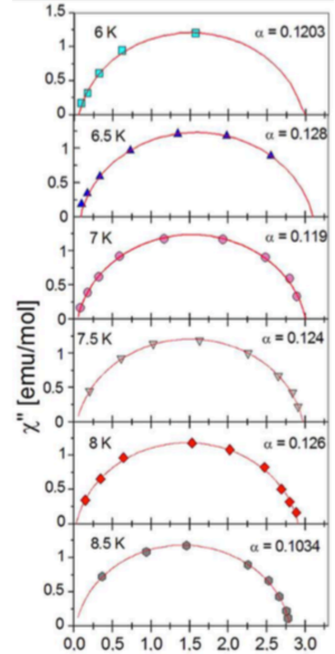
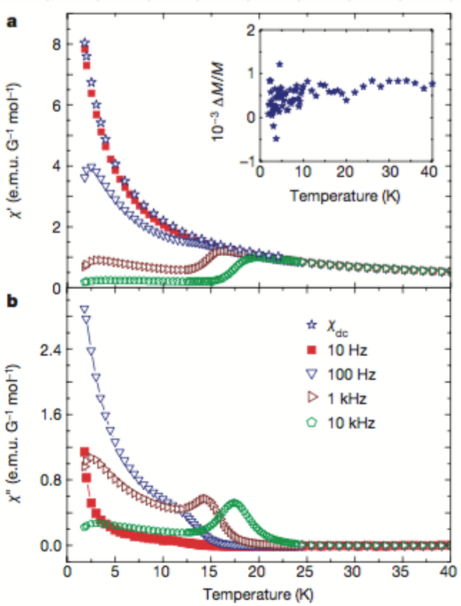
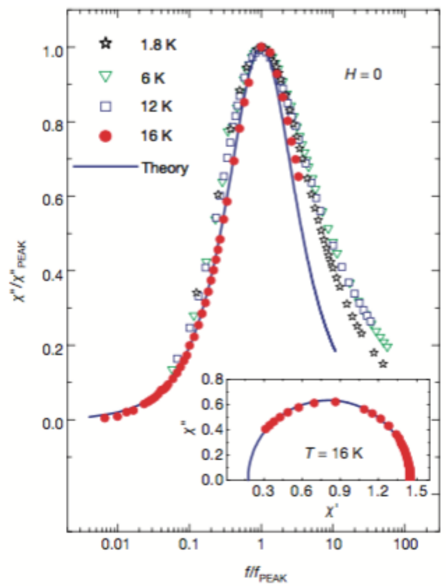


Fig. 13. Argand plots at several temperatures for the [MnF$_4$TPP][TCNE] SCM obtained from the data shown in Fig. 12.

How `spin ice' freezes

NATURE **413** , (2001)





PHYSICAL REVIEW B **91**, 054423 (2015)

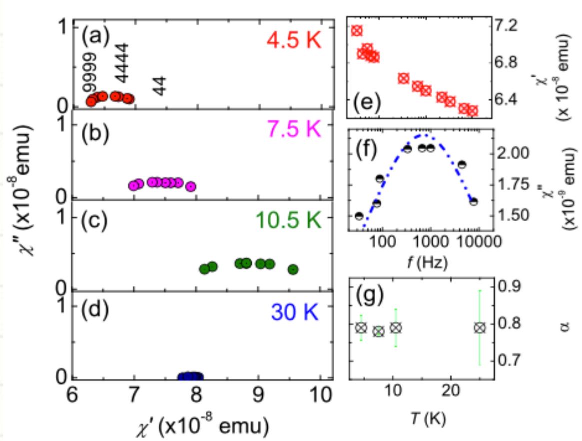
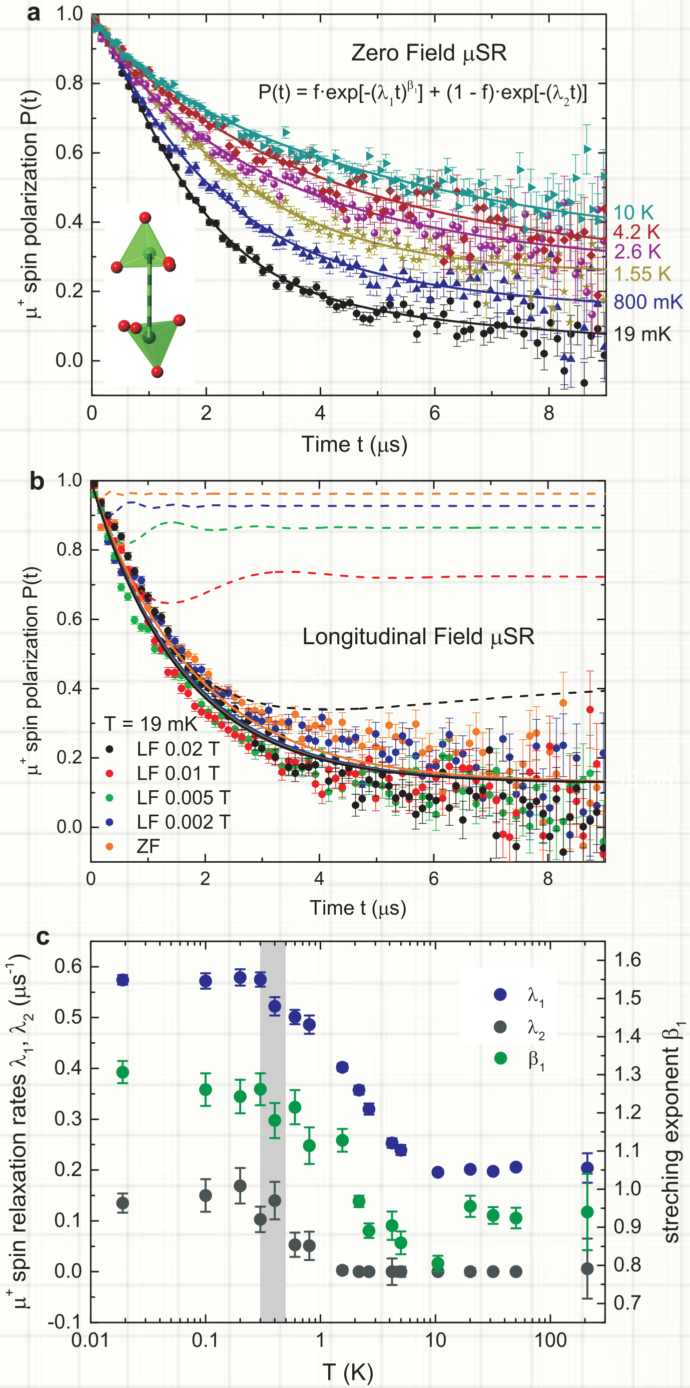


FIG. 4. (Color online) (a)–(d) The Cole-Cole plot (χ''-χ') at different temperatures below 30 K supports the claim of a broad distribution of relaxation times in $FeAl_2O_4$. The numbers in (a)

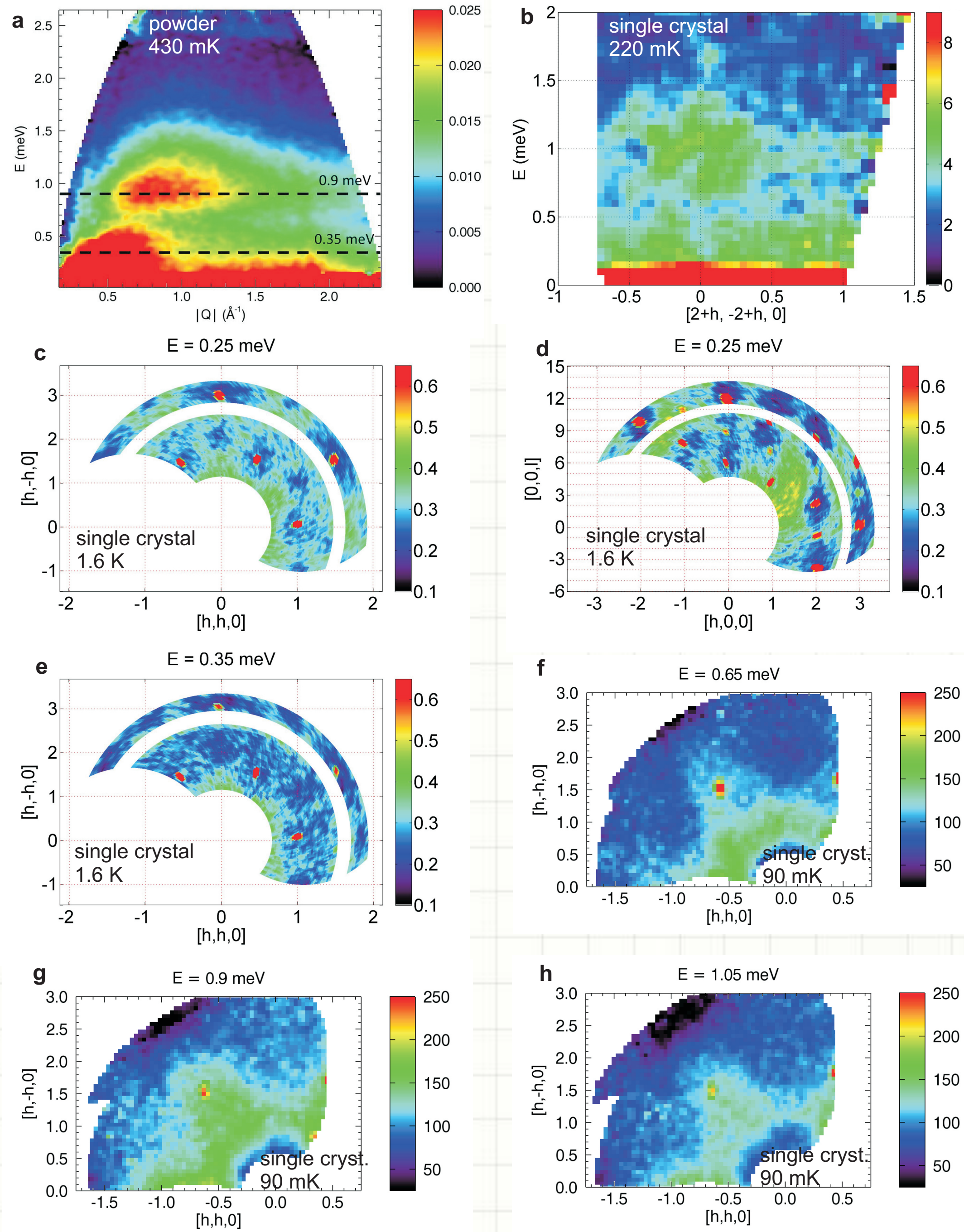
No LRO : μSR

- No oscillations in P(t) in ZF μSR down to 19 mK = no static fields
- SG would also have no oscillations!!
- LF μSR confirms absence of freezing
- Relaxation rates start to increase below about 3K and become constant below 0.3 – 0.5 K
- Persistent slow dynamics below 0.3 K



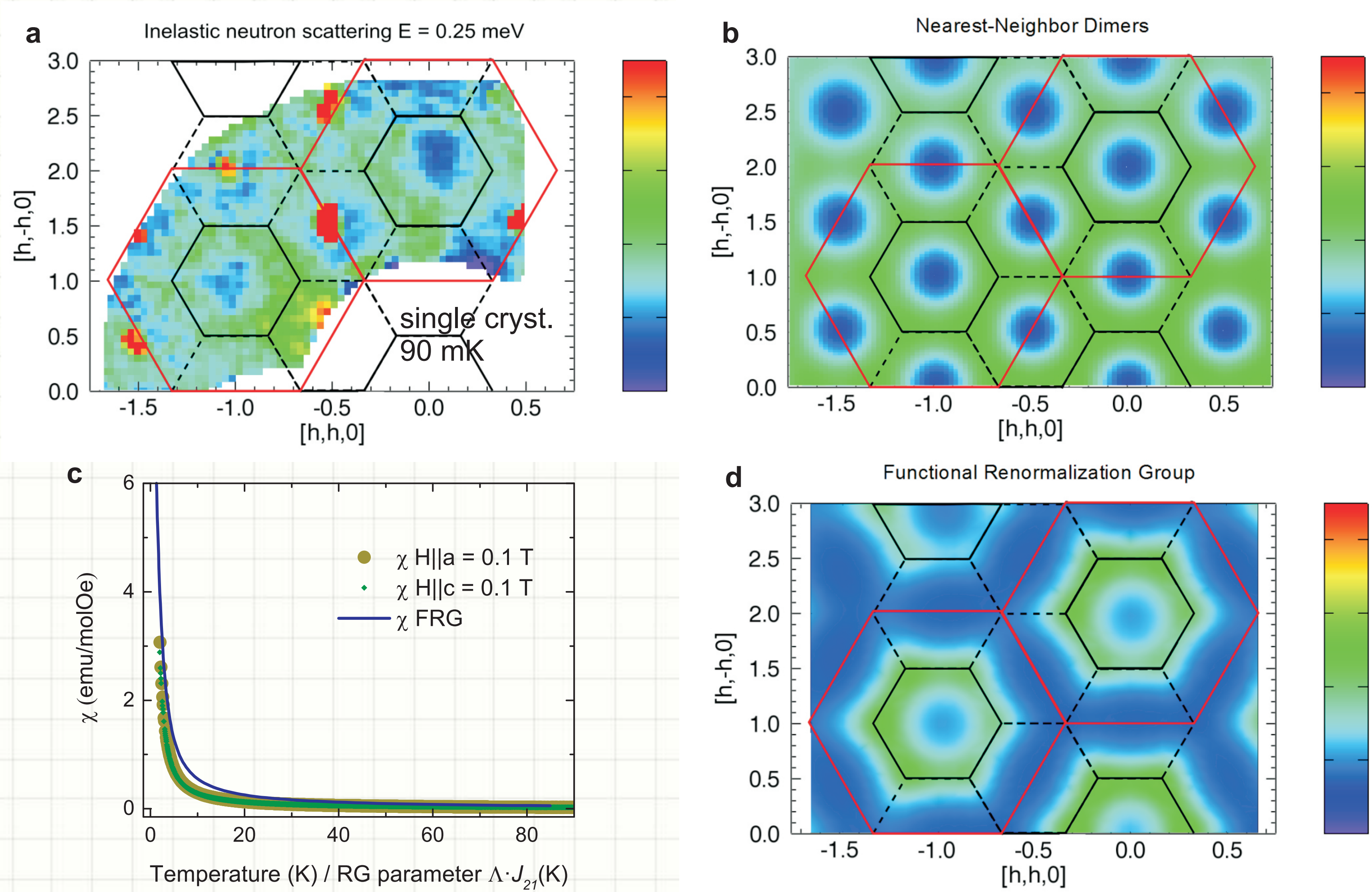
Inelastic Neutron Scattering

- Diffuse magnetic excitations
- excitations appear gapless
- two distinct bands with energy ranges 0.0-0.6 meV and 0.7-1.5 meV
- No magnetic scattering above 1.6 meV ~ 18 K
- Ring-like features in lower band (c & e)
- Block-like features in higher band (f- h)
- Fig.**d:** spectrum perp. to the kagome bilayers at 0.25 meV, no dispersion along the ([0,0,l]) = 2D-magnetism

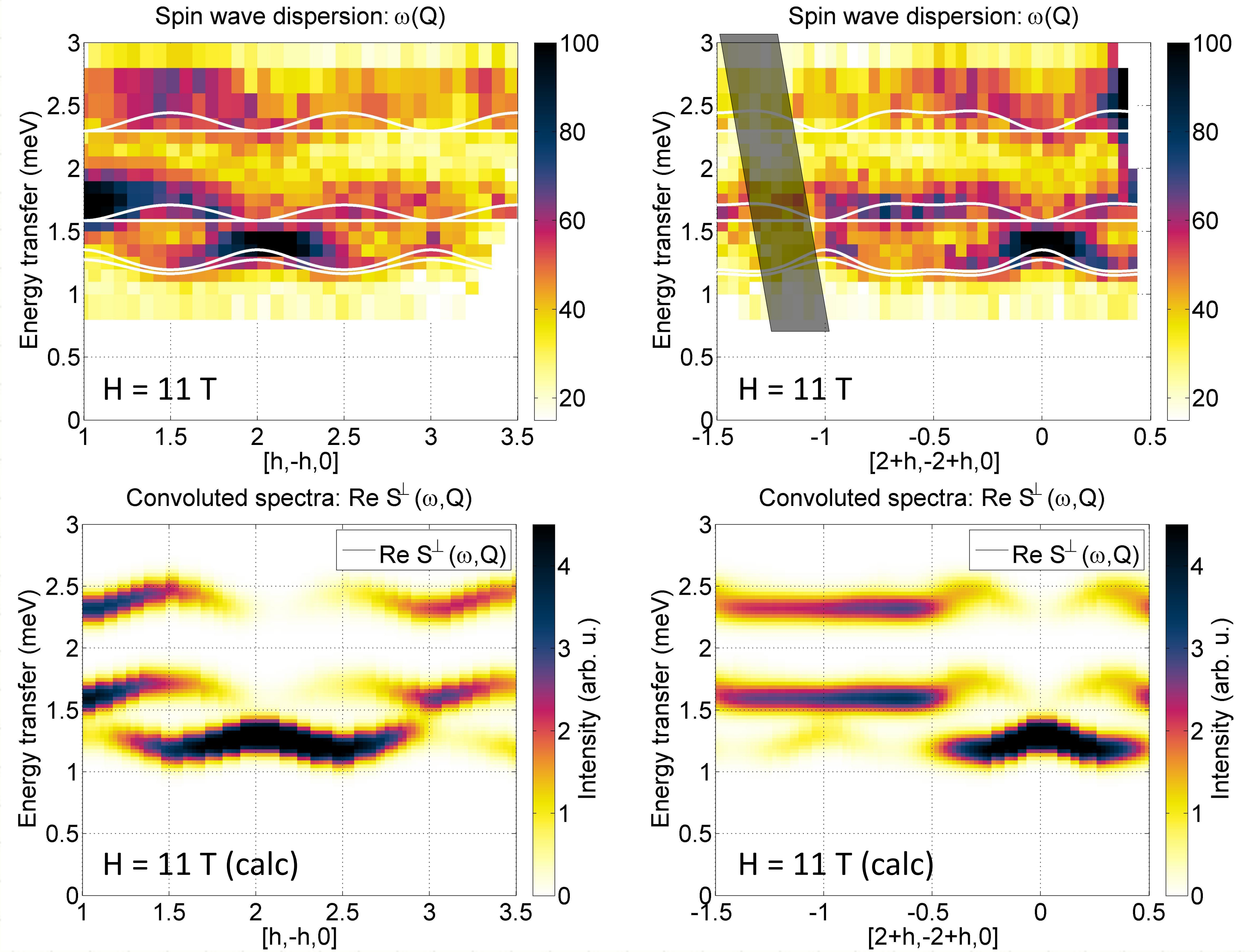


Broad, diffuse excitations consistent with spinons

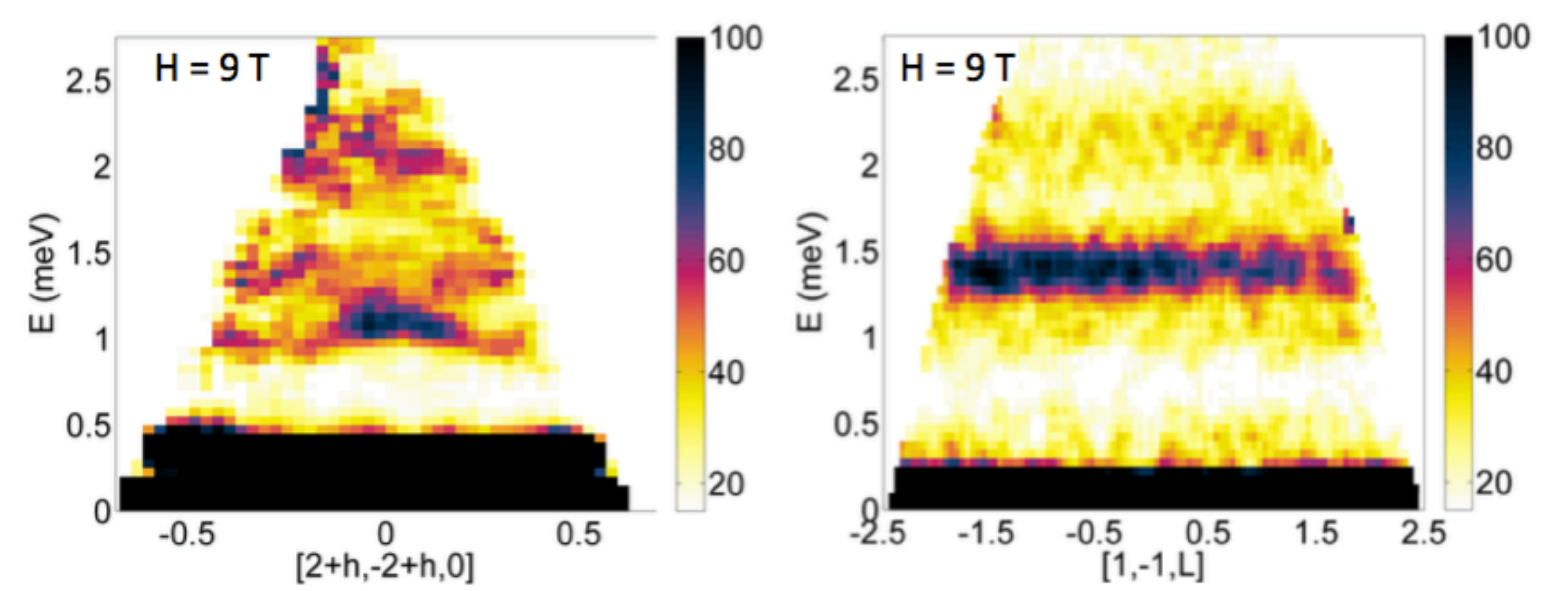
Inelastic Neutron Scattering at H =0



Spin Waves: INS at H = 11T, T = 90 mK



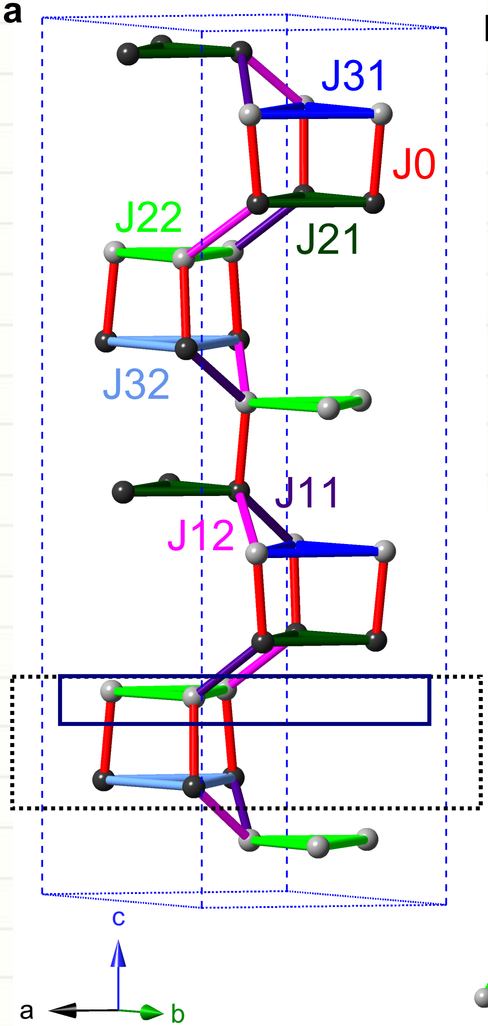
Spin Waves: INS at H = 9T, T = 2K



- Sharp spin-wave dispersion within Kagome bilayers
- Absence of dispersion along (0,0,L) direction (perpendicular to Kagome bilayers) indicates 2D magnetic interactions

Magnetic Interactions : Novel Frustration Mechanism

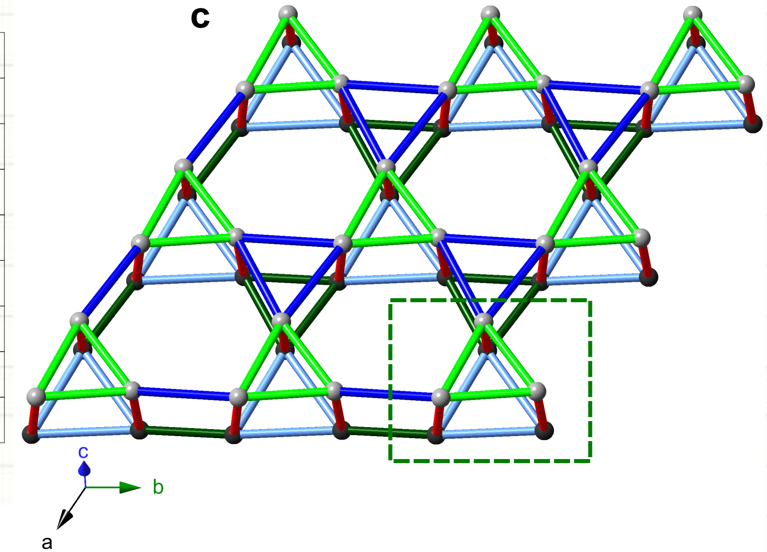
a



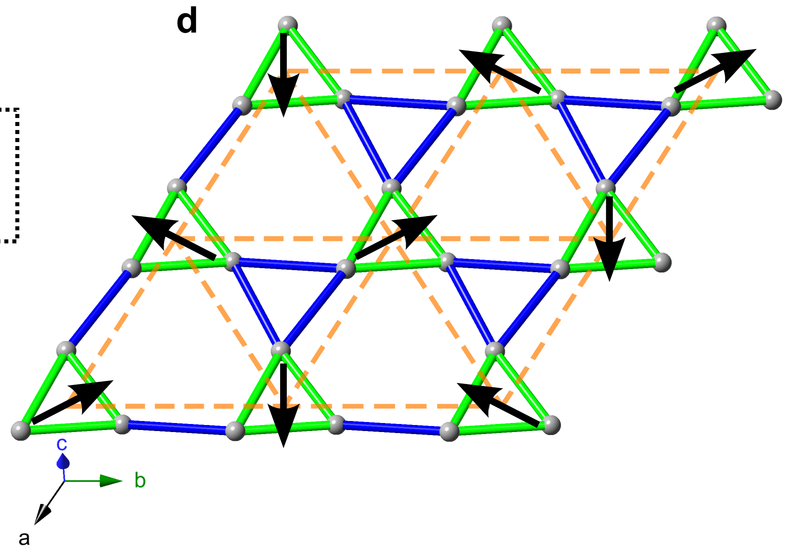
b

exchange	coupling [meV]	type
J0	-0.08(4)	FM
J11	0	
J12	0	
J21	-0.76(5)	FM
J22	-0.27(3)	FM
J31	0.09(2)	AFM
J32	0.11(3)	AFM
ΣJ	-0.91(17)	

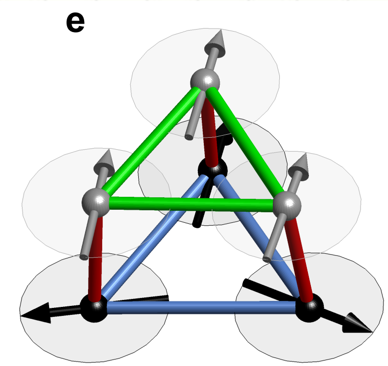
c



d



e



Summary

- A new QSL candidate discovered in the complex oxide $Ca_{10}Cr_7O_{28}$ with a bilayer Kagome lattice of S = ½ Cr^{5+} moments
- No LRO or freezing down to 19 mK
- Gapless, dispersionless, diffuse magnetic excitations: spinons
- Unusual combination of magnetic interactions with dominant FM interactions leads to frustratation
- PFFRG for the extracted Hamiltonian confirms absence of LRO

C. Balz, et al. arxiv:1606.06463,
Nature Physics (2016)

Herbertsmithite