GEOMETRICALLY FRUSTRATED MAGNETS

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Outline

How are geometrically frustrated magnets special?

What they are not

Evading long range order

Degeneracy and fluctuations - models and experiment

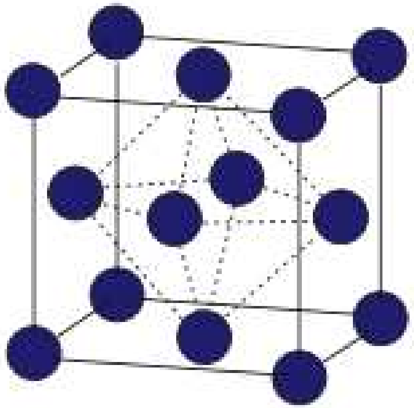
Statistical physics of underconstrained systems

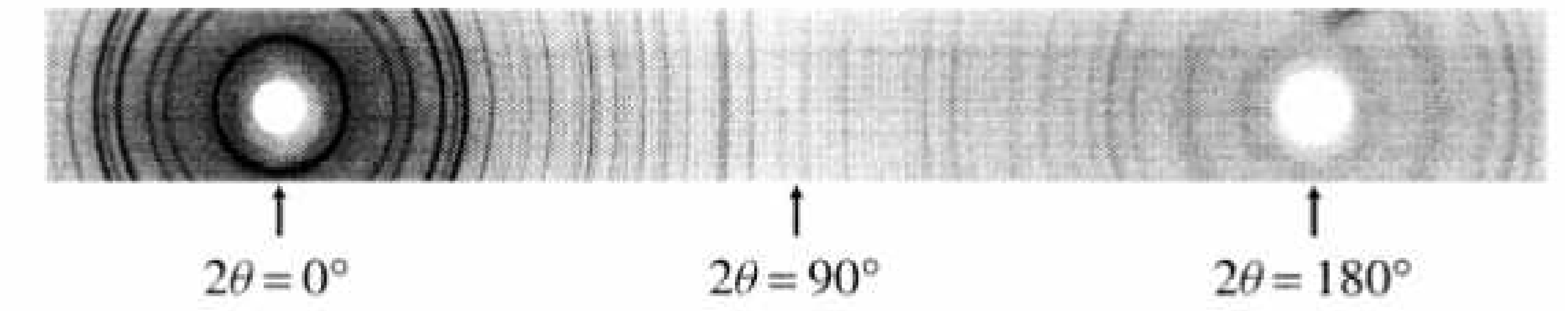
Emergent degrees of freedom and classical fractionalization

Condensed matter at low temperature

Symmetry breaking as the norm

Crystalline solids

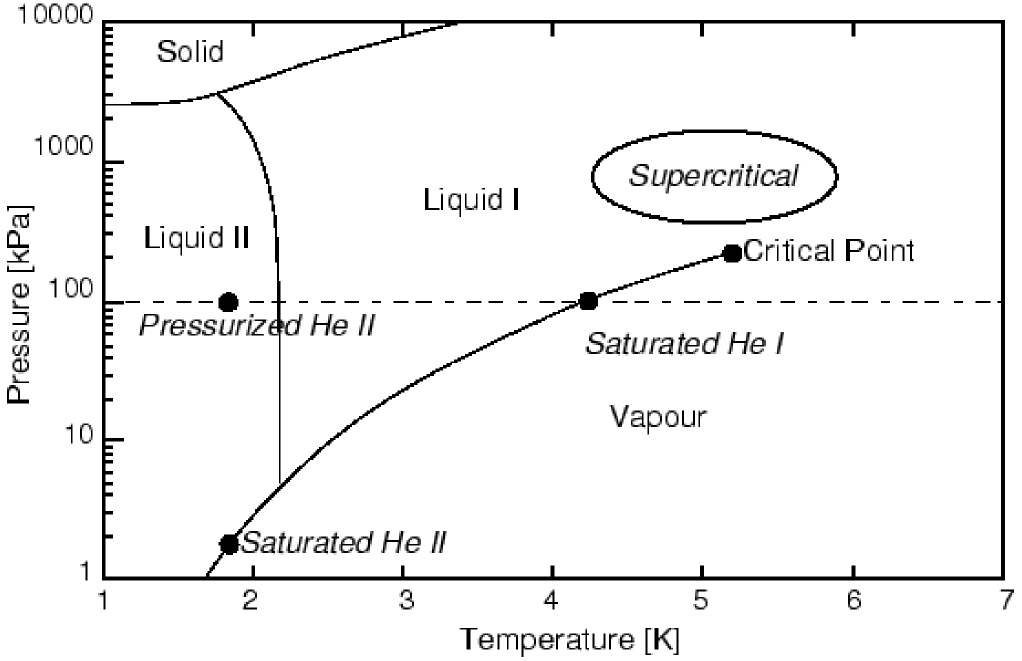


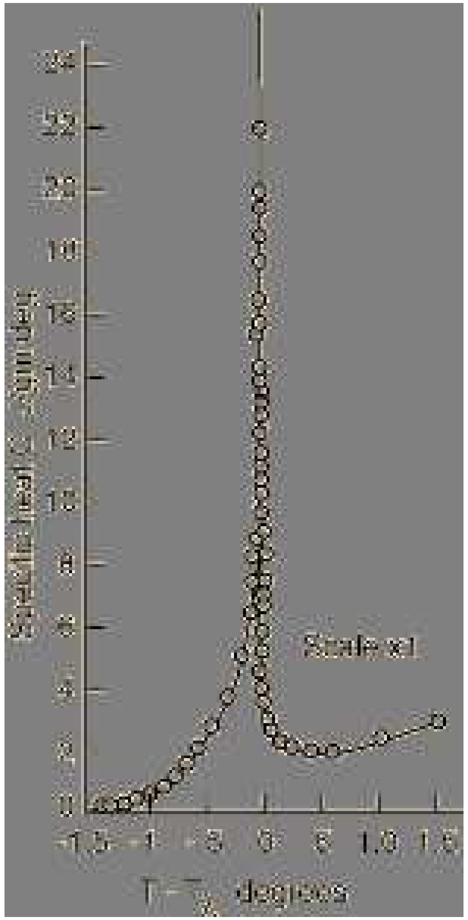


Broken symmetry in Bose liquids

Quantum fluctuations suppress crystalisation

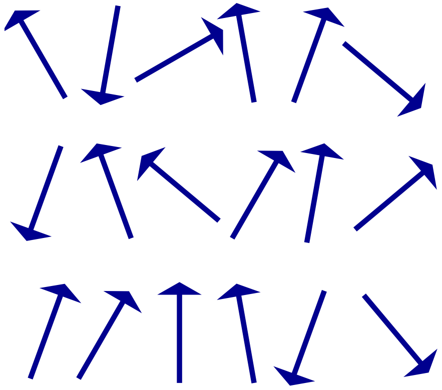
^{4}He phase diagram

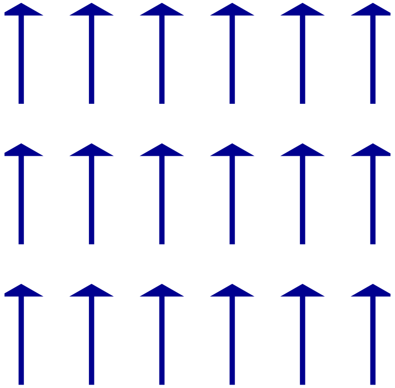




Ordering in ferromagnets

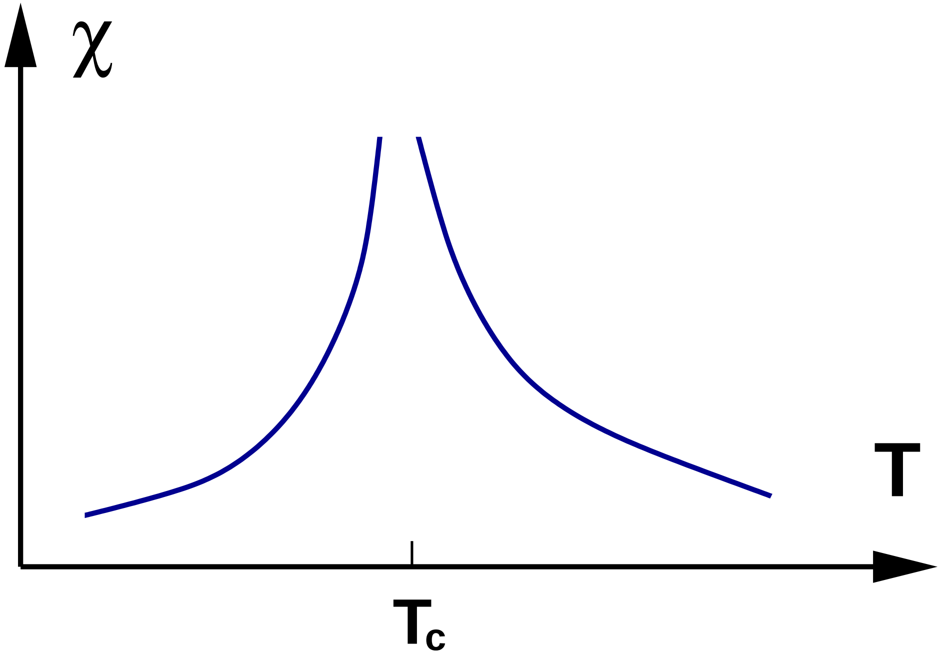
High temperature





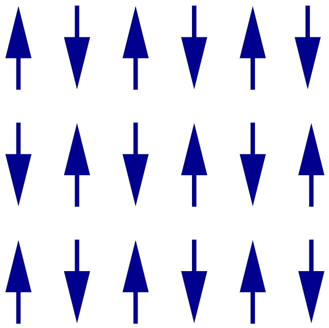
Ground state

Susceptibility

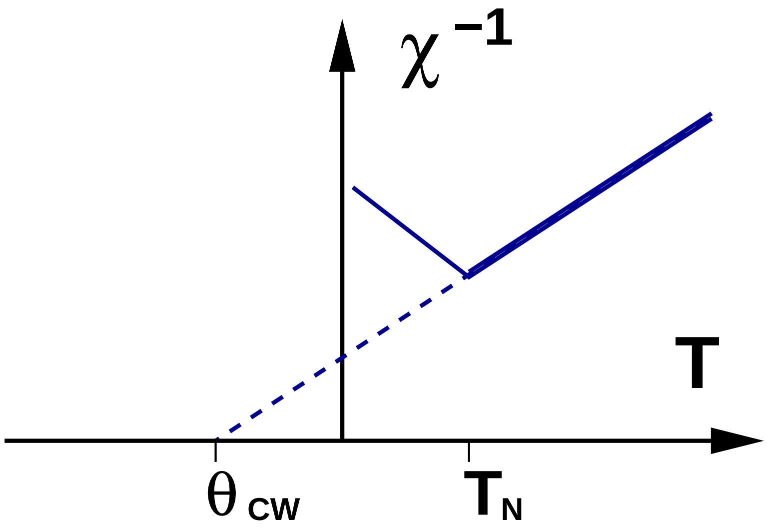


Unfrustrated antiferromagnetic order

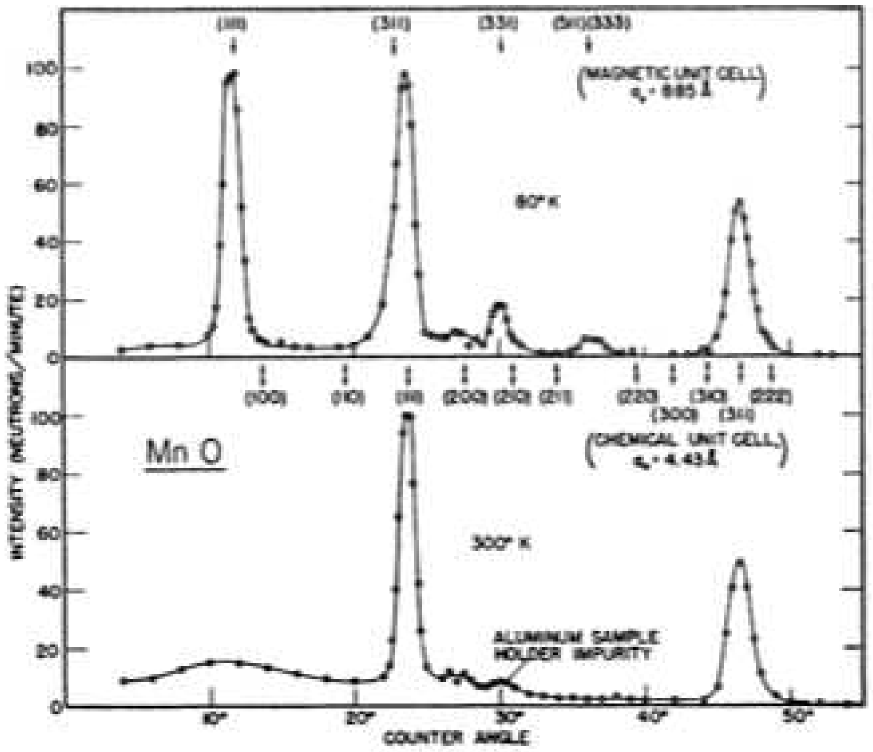
Néel order



Inverse susceptibility



Neutron diffraction

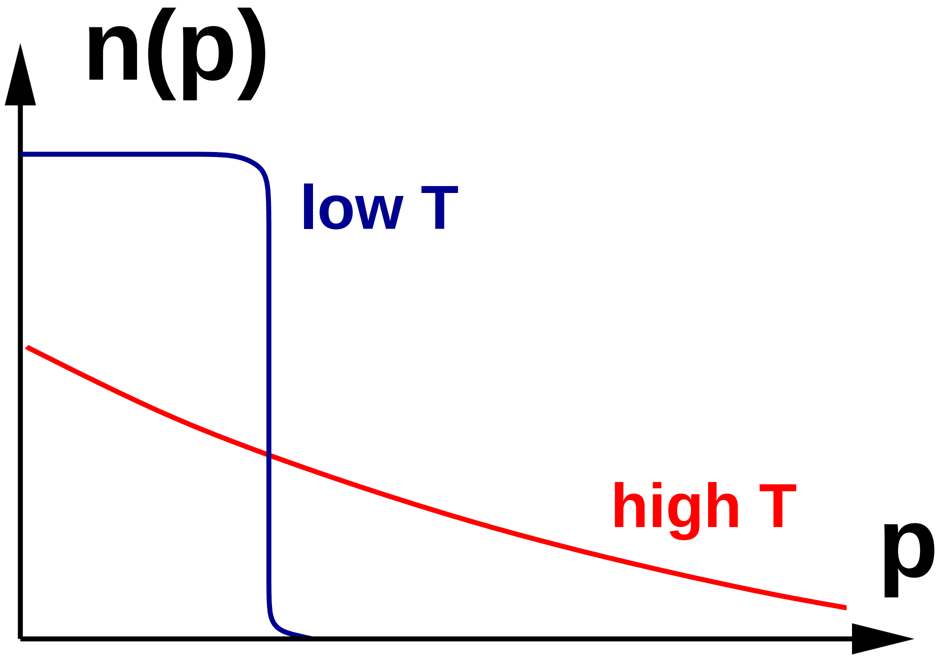


Shull and Smart (1949)

Alternative to symmetry breaking # 1

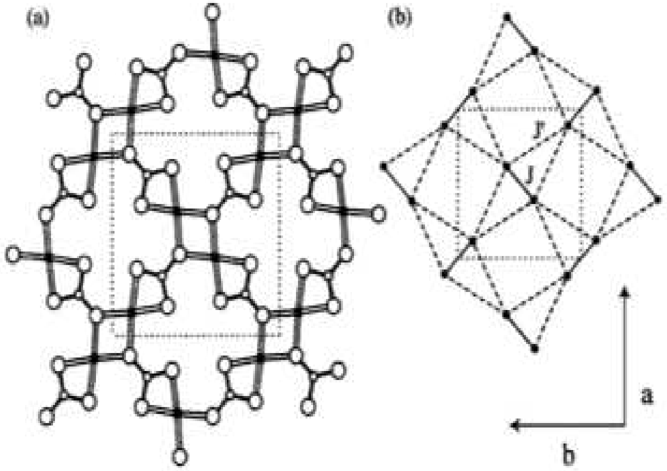
. . . a unique ground state

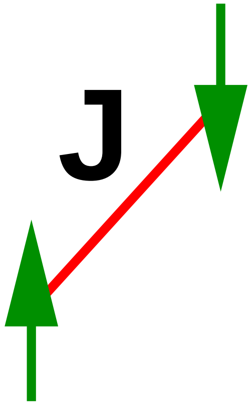
In the Fermi gas



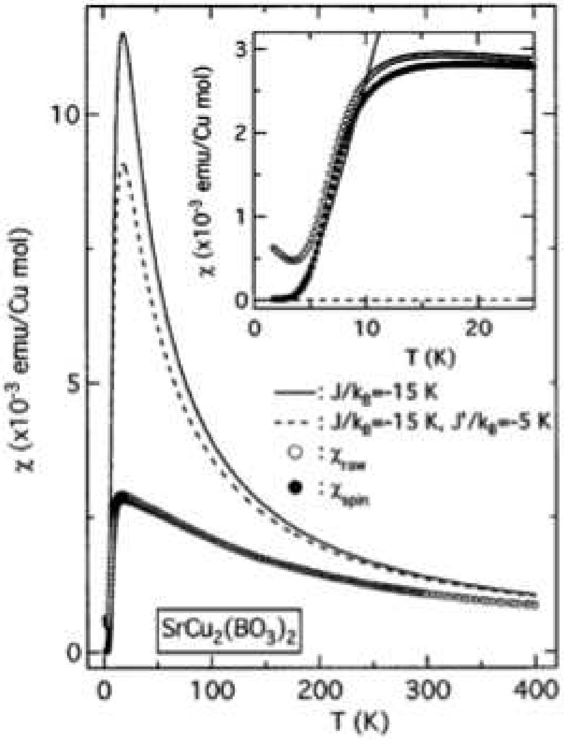
Momentum distribution

Spin system with unique ground state





Weakly coupled singlet pairs



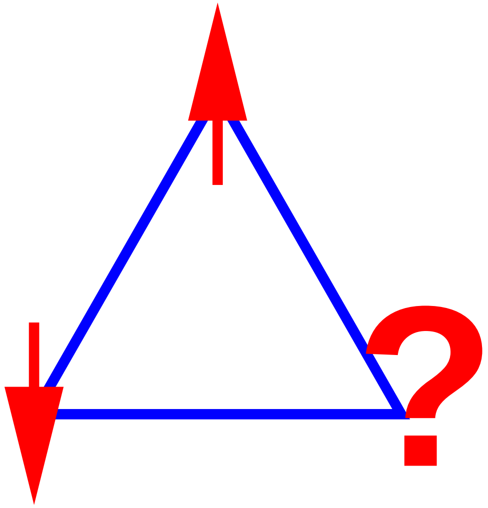
χ **vs** T

$SrCu_2(BO_3)_2$

Alternative to symmetry breaking # 2

. . . strong fluctuations

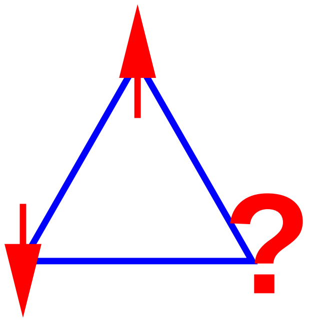
Frustration and degeneracy



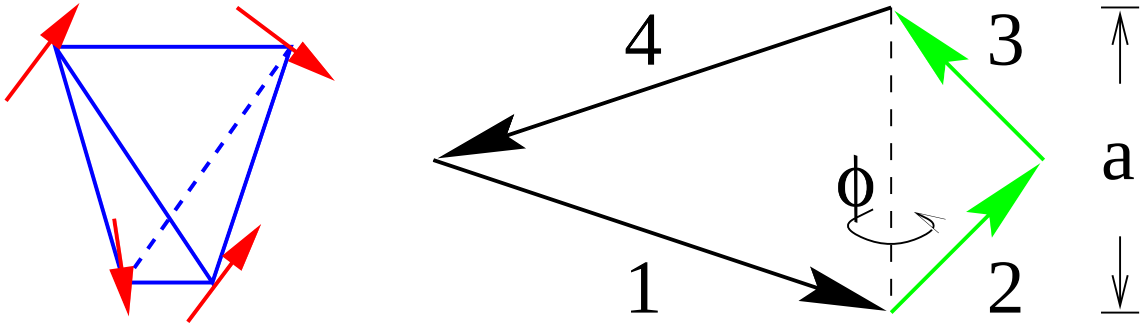
Anderson 1956, Villain 1979

Antiferromagnetic spin clusters
- frustration and degeneracy

Ising triangle



Heisenberg tetrahedron



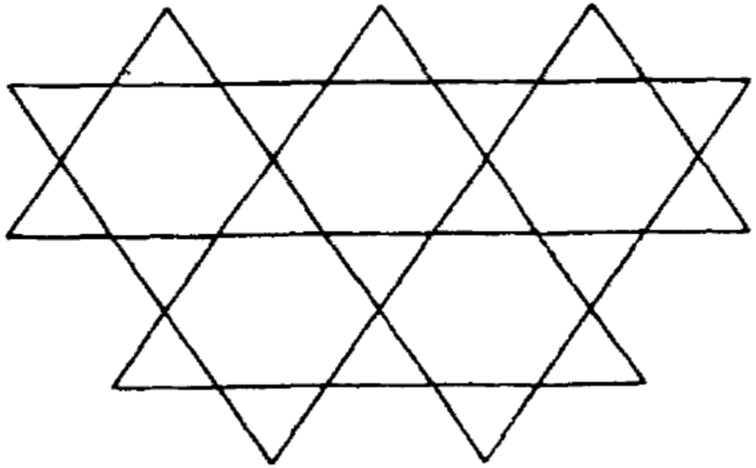
Ground states: **cluster spin** $\mathbf{L} \equiv \sum_i \mathbf{S}_i$ **minimised**

$$\mathcal{H} = J \sum_{\text{pairs}} \mathbf{S}_i \cdot \mathbf{S}_j \equiv \frac{J}{2} |\mathbf{L}|^2 + c$$

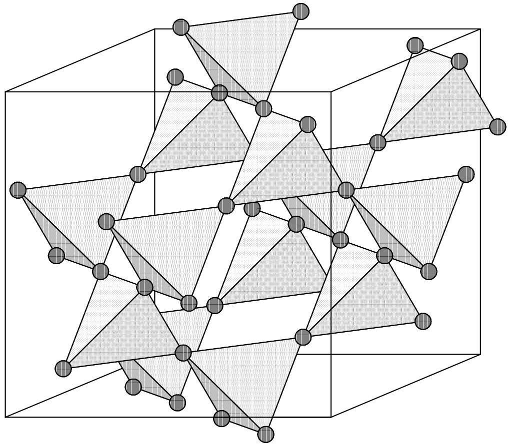
Examples of frustrated lattices

Building block: corner-sharing frustrated units

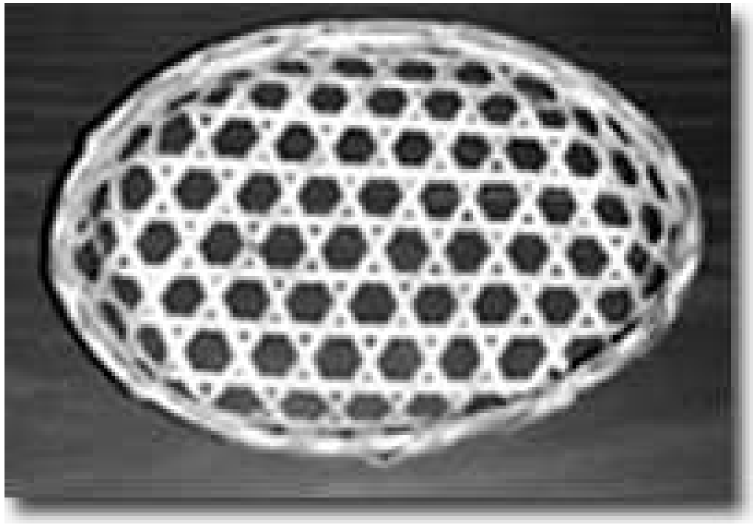
2D: kagome lattice



3D: pyrochlore lattice



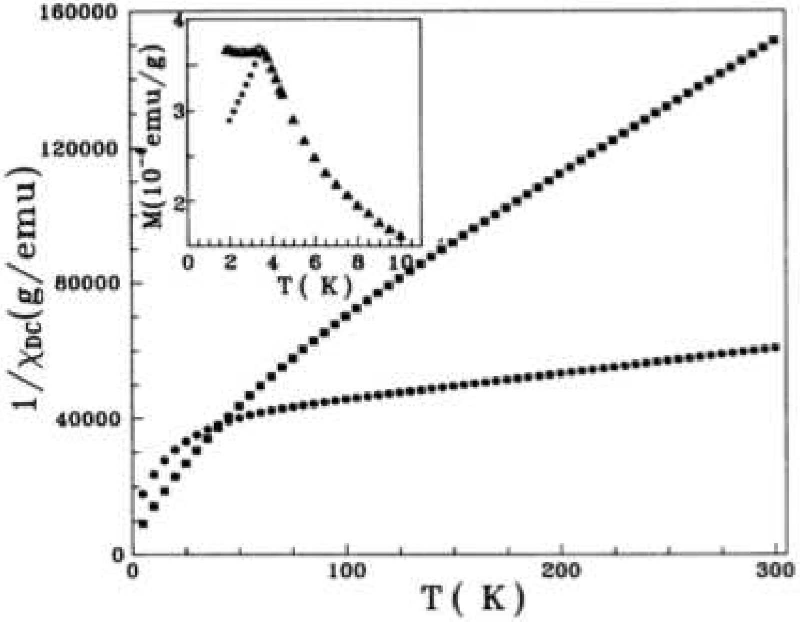
Frustrated lattices beyond physics . . .



Characteristics of frustrated magnets

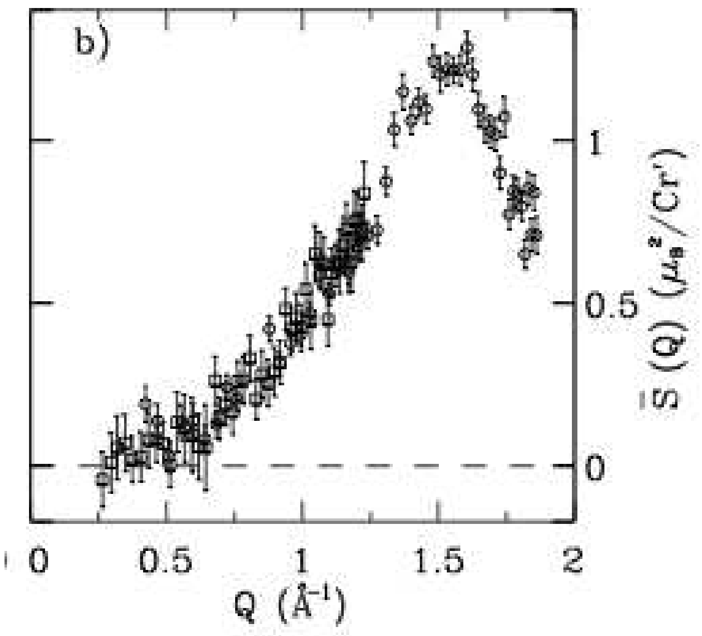
$\mathrm{SrGa_{12-x}Cr_xO_{19}}$ **(SCGO) as an example**

Paramagnetic even for $T \ll |\Theta_{\mathrm{CW}}|$ **Strong short-range correlations**



χ^{-1} **vs T**

Martinez et al, PRB **46**, 10786 (1992)



Elastic neutron scattering

S.H. Lee et al, Europhys Lett **35**, 127 (1996)

Selected examples of frustrated magnets

Layered materials

SCGO

pyrochlore slabs

$$Cr^{3+} \qquad S = 3/2$$

$$\Theta_{\mathrm{CW}} \sim 500\mathrm{K} \qquad T_{\mathrm{F}} \sim 4\mathrm{K}$$

hydromium iron jarosite

kagome layers

$$Fe^{3+} \qquad S = 5/2$$

$$\Theta_{\mathrm{CW}} \sim 700\mathrm{K} \qquad T_{\mathrm{F}} \sim 14\mathrm{K}$$

Herbertsmithite

kagome layers

$$Cu^{2+} \qquad S = 1/2$$

$$\Theta_{\mathrm{CW}} \sim 300\mathrm{K}$$

Pyrochlore antiferromagnets

$Y_2Mo_2O_7$

$$Mo^{4+} \qquad S = 1$$

$$\Theta_{\mathrm{CW}} \sim 200\mathrm{K} \quad T_{\mathrm{F}} \sim 22\mathrm{K}$$

Cs Ni Cr F_6

$$Ni^{2+}\, S = 1 \quad Cr^{3+}\, S = 3/2$$

$$\Theta_{\mathrm{CW}} \sim 70\mathrm{K} \qquad T_{\mathrm{F}} \sim 2.3\mathrm{K}$$

Spin ice materials

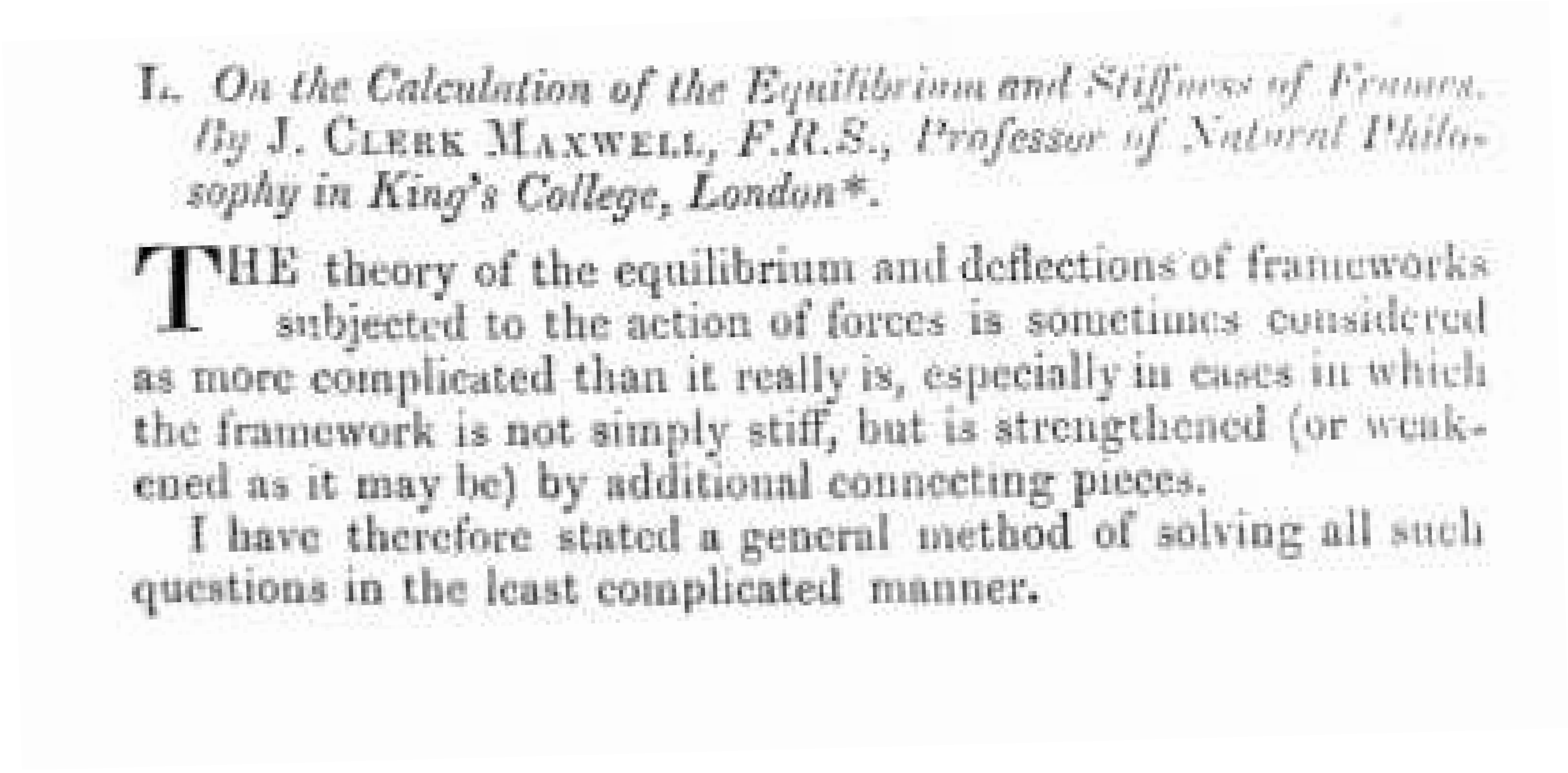
$Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$

ferromagnets with single-ion anisotropy
— hence frustration

$$J_{\mathrm{eff}} \sim 1\mathrm{K} - 2\mathrm{K}$$

Ground state degeneracy in classical Heisenberg models

Maxwellian constraint-counting



I. *On the Calculation of the Equilibrium and Stiffness of Frames.* *By* J. Clerk Maxwell, *F.R.S., Professor of Natural Philosophy in King's College, London**.

THE theory of the equilibrium and deflections of frameworks subjected to the action of forces is sometimes considered as more complicated than it really is, especially in cases in which the framework is not simply stiff, but is strengthened (or weakened as it may be) by additional connecting pieces.

I have therefore stated a general method of solving all such questions in the least complicated manner.

Ground state degeneracy in Heisenberg pyrochlore AFM

$$\mathcal{H} = J \sum_{\text{bonds}} \mathbf{S}_i \cdot \mathbf{S}_j \equiv \frac{J}{2} \sum_{\text{units}} |\mathbf{L}_\alpha|^2 + c$$

Total number of degrees of freedom: $F = 2 \times \big($number of spins$\big)$

Constraints satisfied in ground state: $K = 3 \times \big($number of units$\big)$

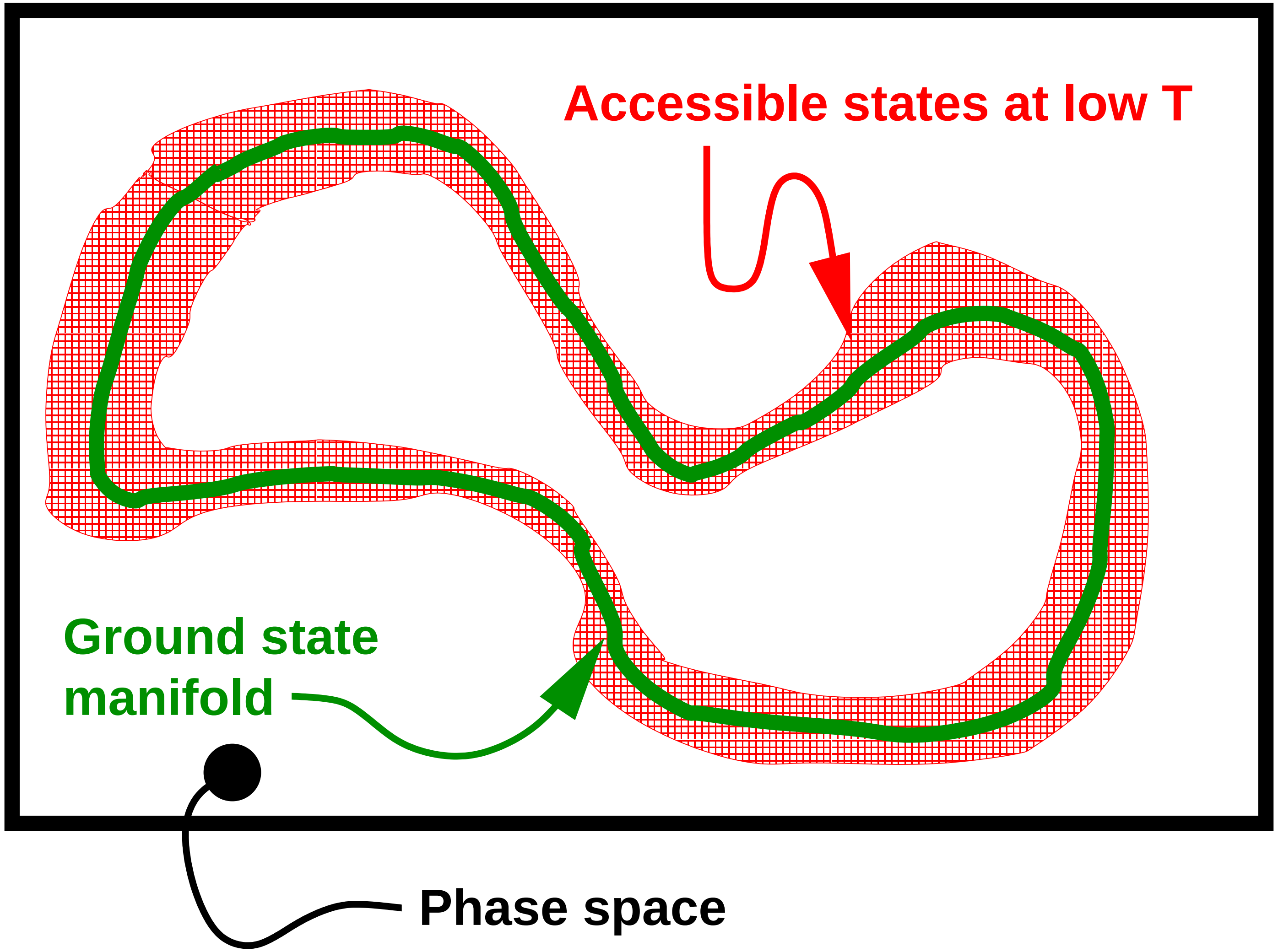
Ground state dimension:

$\boxed{\text{D=F-K}}$

Geometric Frustration $\longrightarrow$ Macroscopic D

Schematics of behaviour at low temperature

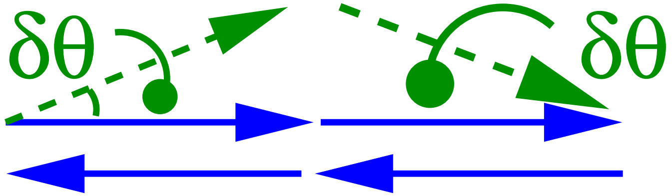
Classical cooperative paramagnet: $JS \ll k_{\mathrm{B}}T \ll JS^2$



Ground state selection by fluctuations?

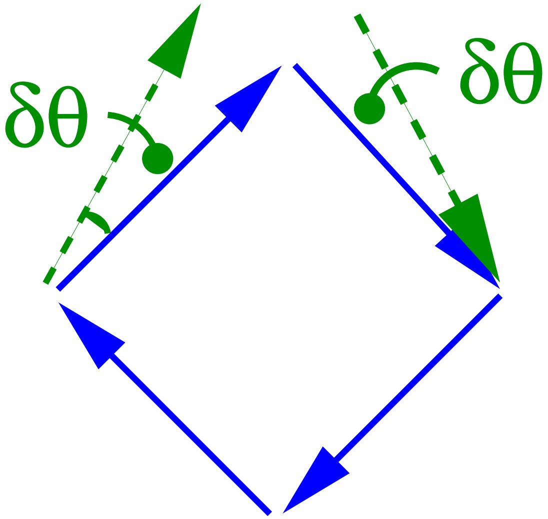
‘Order by disorder’   Villain (1980), Shender (1982)

Some states have soft modes



$$E = \frac{J}{2}|\mathbf{L}|^2 \propto (\delta\theta)^4$$

Others don’t



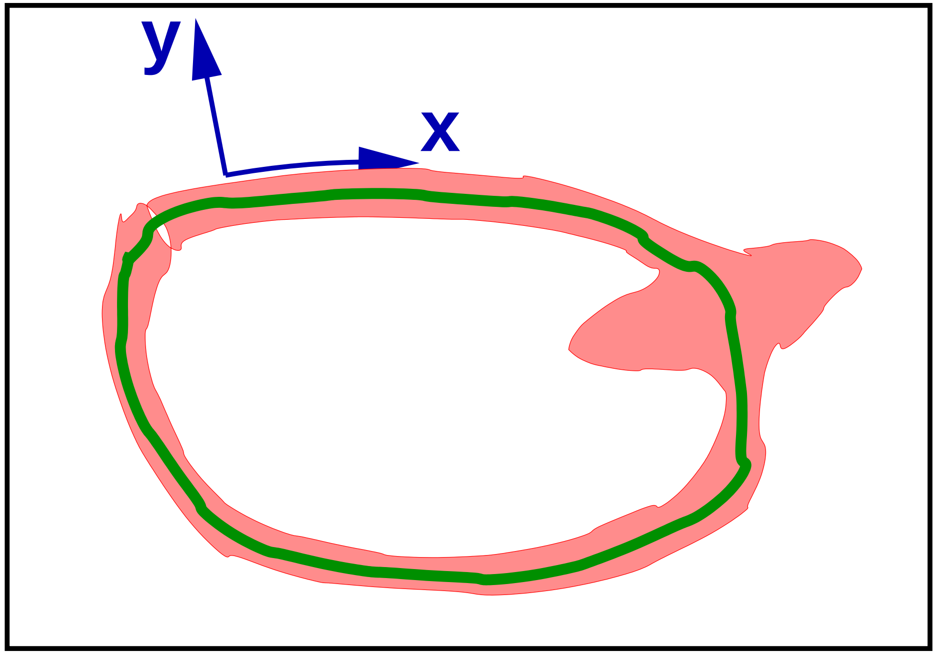
$$E = \frac{J}{2}|\mathbf{L}|^2 \propto (\delta\theta)^2$$

Ground state selection?

Thermal fluctuations

Probability distribution on ground states

$$\int \mathrm{d}y\ \mathrm{e}^{-\omega y^2/k_\mathrm{B}T} \propto \sqrt{\frac{k_\mathrm{B}T}{\omega}}$$



$$P(\mathbf{x}) \propto \prod_l \left(\frac{k_\mathrm{B}T}{\omega_l(\mathbf{x})}\right)$$

Thermal fluctuations

kagome → coplanar

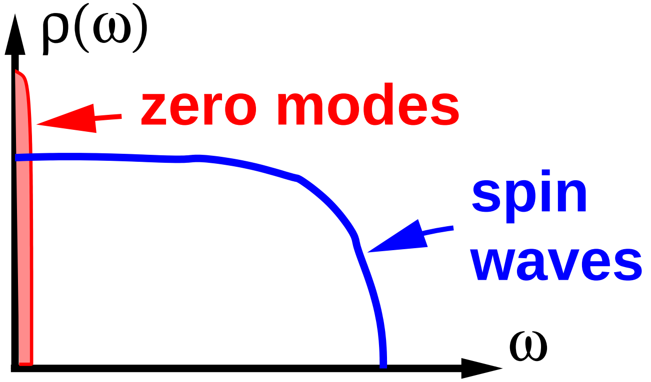
pyrochlore → disordered

Dynamics of Heisenberg systems

How does system explore ground state manifold?

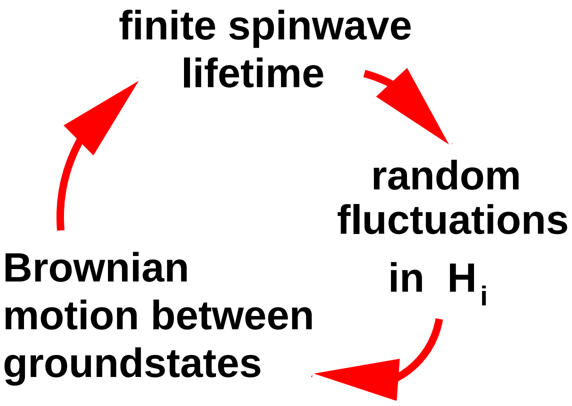
Equation of motion: $$\hbar\frac{d\mathbf{S}_i}{dt} = \mathbf{S}_i \times \mathbf{H}_i = -J\mathbf{S}_i \times \sum_j \mathbf{S}_j$$

Harmonic approximation



Normal mode frequencies ω

Anharmonic interactions



finite spinwave lifetime

random fluctuations in $\mathbf{H}_i$

Brownian motion between groundstates

Langevin: $d\mathbf{S}/dt(0) = \mathbf{S} \times \mathbf{h}(t)$

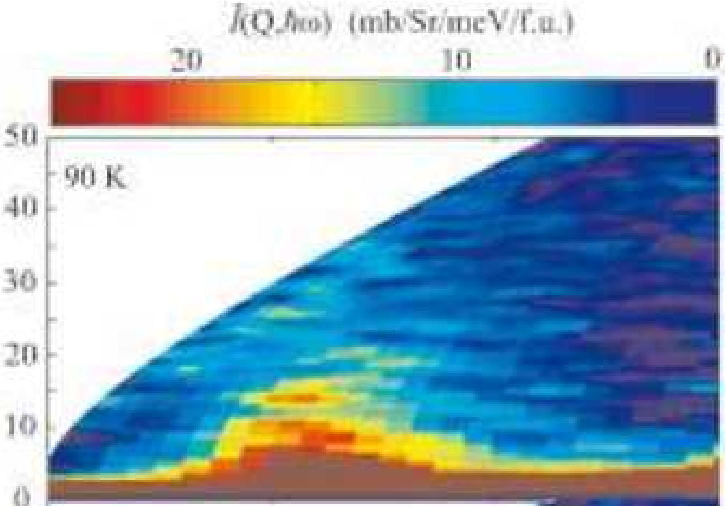
$$\langle \mathbf{S}(0) \cdot \mathbf{S}(t) \rangle \approx \exp(-ck_{\mathrm{B}}Tt/\hbar)$$

Quasielastic neutron scattering in $Y_2Ru_2O_7$

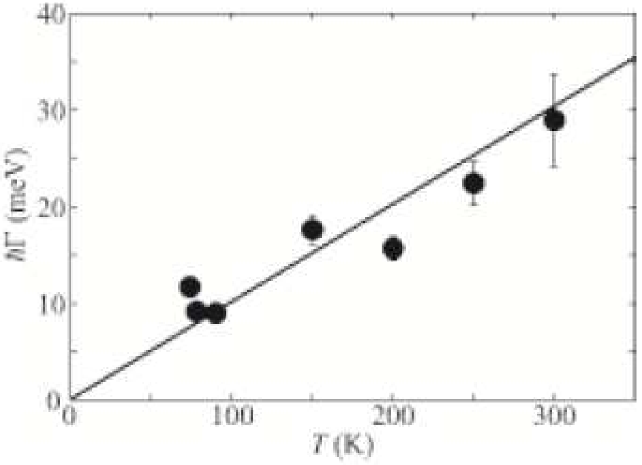
$$\Theta_{\mathrm{CW}} = -1100\mathrm{K}$$

$$T_{\mathrm{N}} = 77\mathrm{K}$$

Scattering vs Q & ω



Linewidth vs temperature



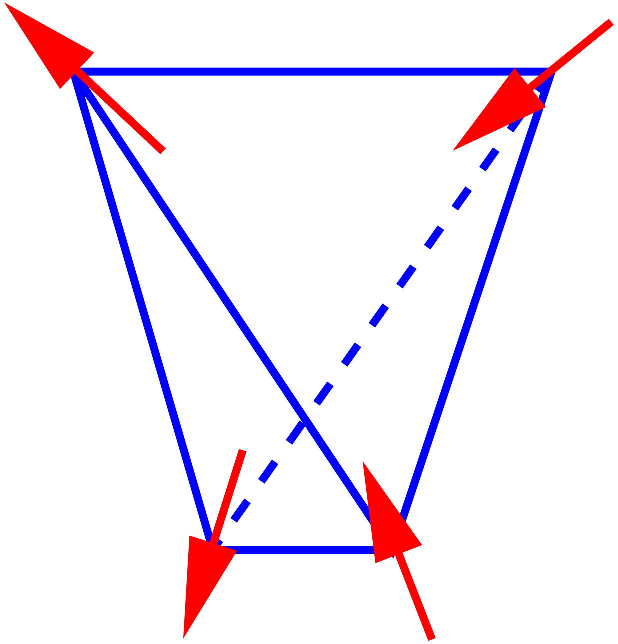
$$\hbar\Gamma = \mathcal{C} k_{\mathrm{B}} T$$

$$\mathcal{C} = 1.17$$

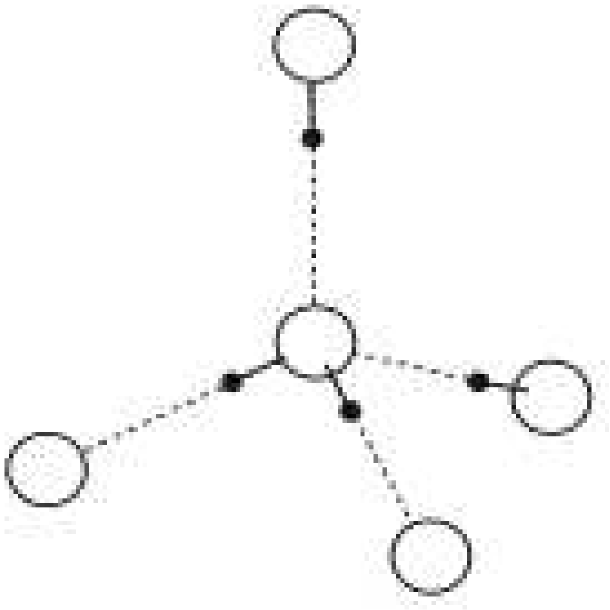
van Dujin *et al* (2008)

Frustration and residual entropy

Spin ice



Water ice



Anisotropy +

ferromagnetic exchange

Pauling 1935

Ground states: ‘two-in, two-out’

Pauling’s entropy estimate

One tetrahedron

Total number of states: 16

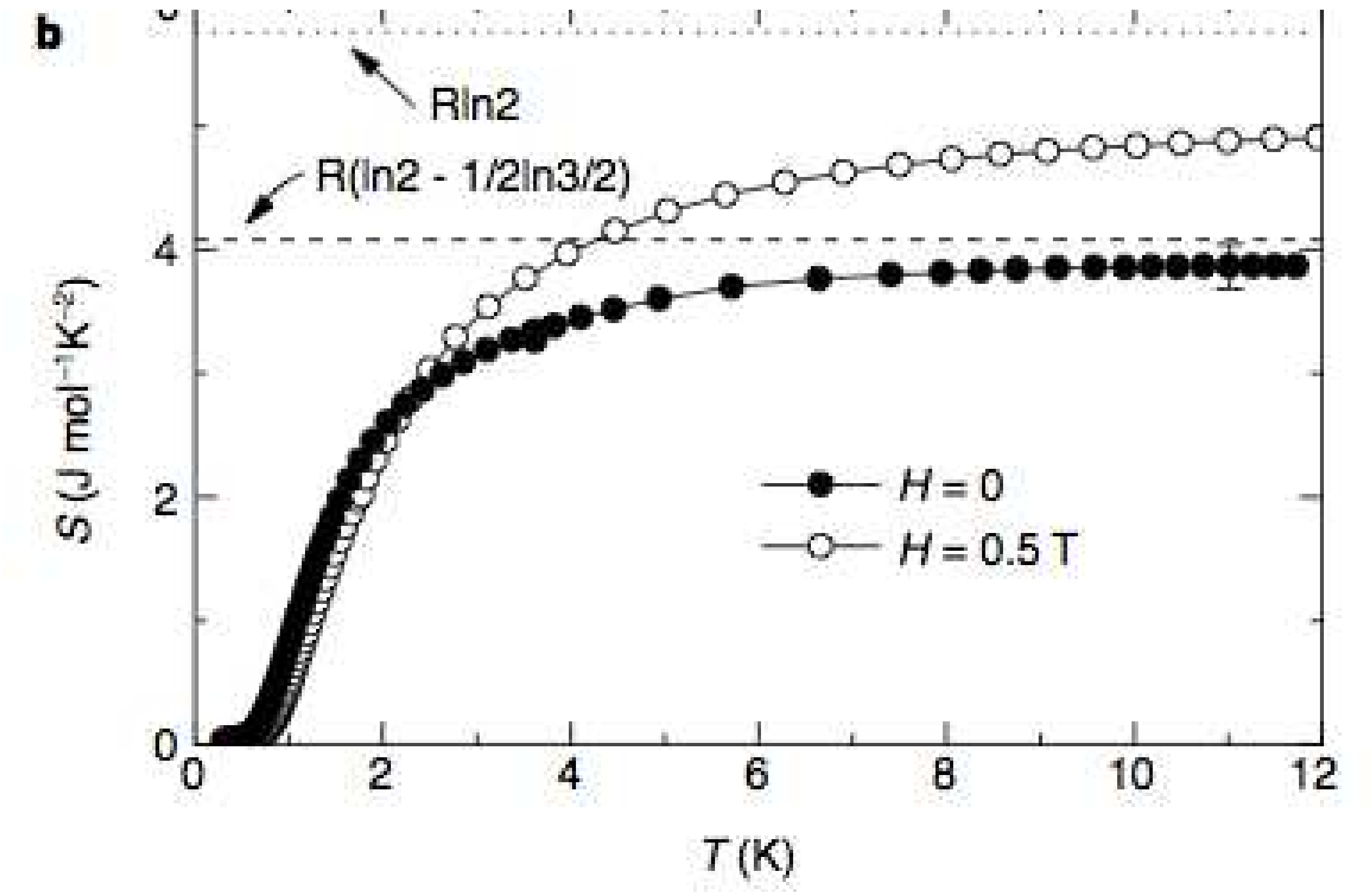
Fraction that are ground states: $\frac{6}{16}$

Pyrochlore lattice

Estimate for number of ground states:

$$(total\,\#\,states) \times \left(\frac{6}{16}\right)^{(\#\,\text{tetrahedra})} = \left(\frac{3}{2}\right)^{(\#\,\text{spins}/2)}$$

Pauling entropy in experiment

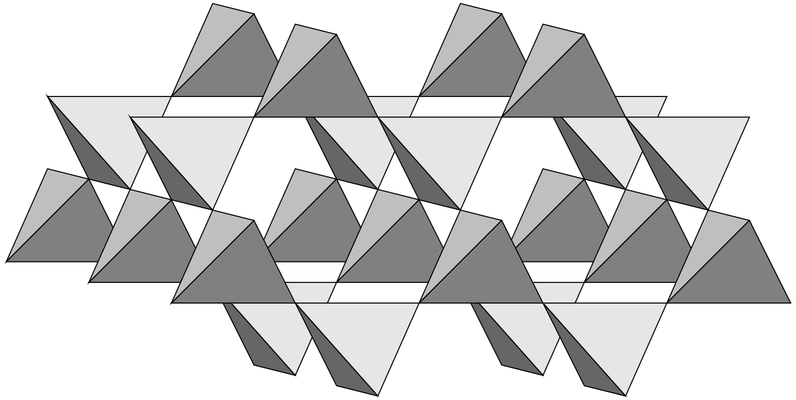


$\mathrm{Dy_2Ti_2O_7}$, **Ramirez *et al*, Nature 399, 333 (1999).**

Correlations induced by ground state constraints

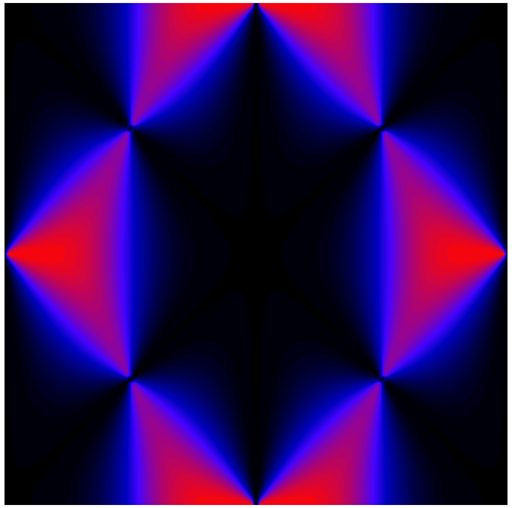
Local constraints

$$\sum_{tet} \mathbf{S}_i = 0$$



Long range correlations

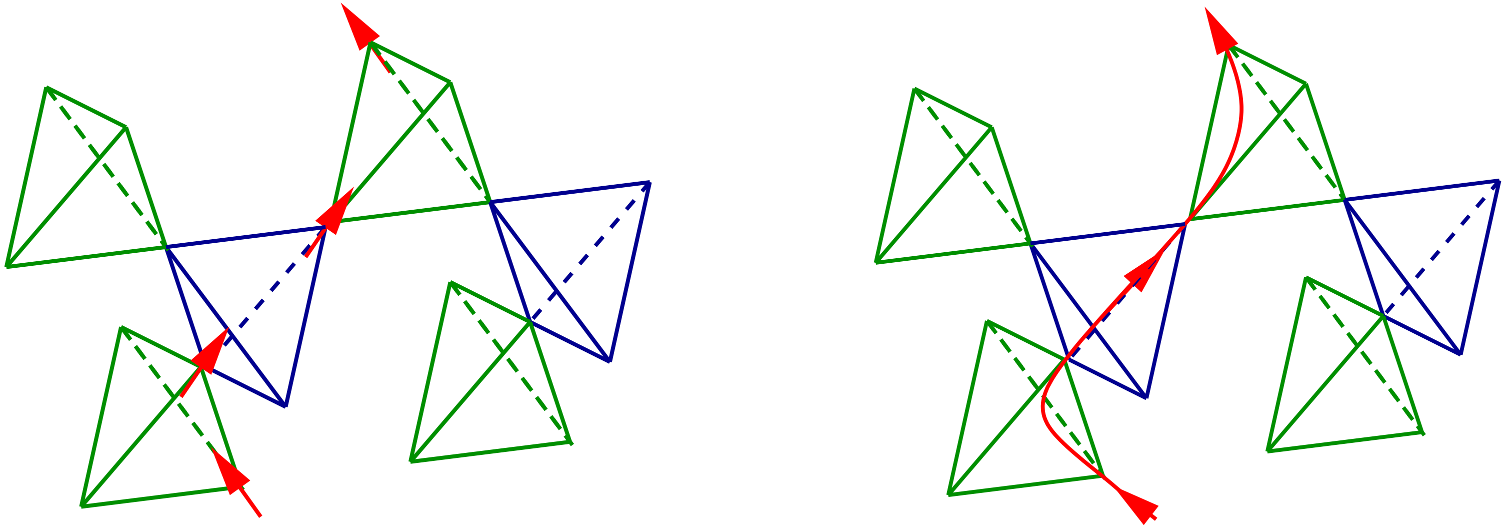
Sharp structure in
$$\langle \mathbf{S}_{-\mathbf{q}} \cdot \mathbf{S}_{\mathbf{q}} \rangle$$



Gauge theory of ground state correlations

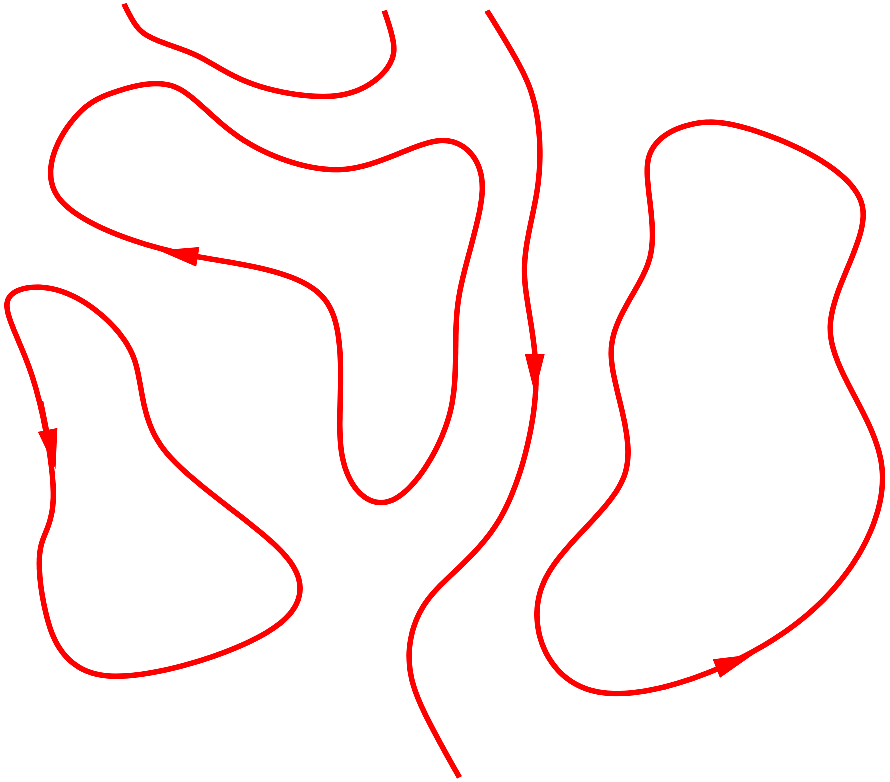
Youngblood *et al* (1980), Huse *et al* (2003), Henley (2004)

Map spin configurations . . . **. . . to vector fields** $\mathrm{B}(\mathbf{r})$



'two-in two out' groundstates . . . **. . . map to divergenceless** $\mathrm{B}(\mathbf{r})$

Ground states as flux loops

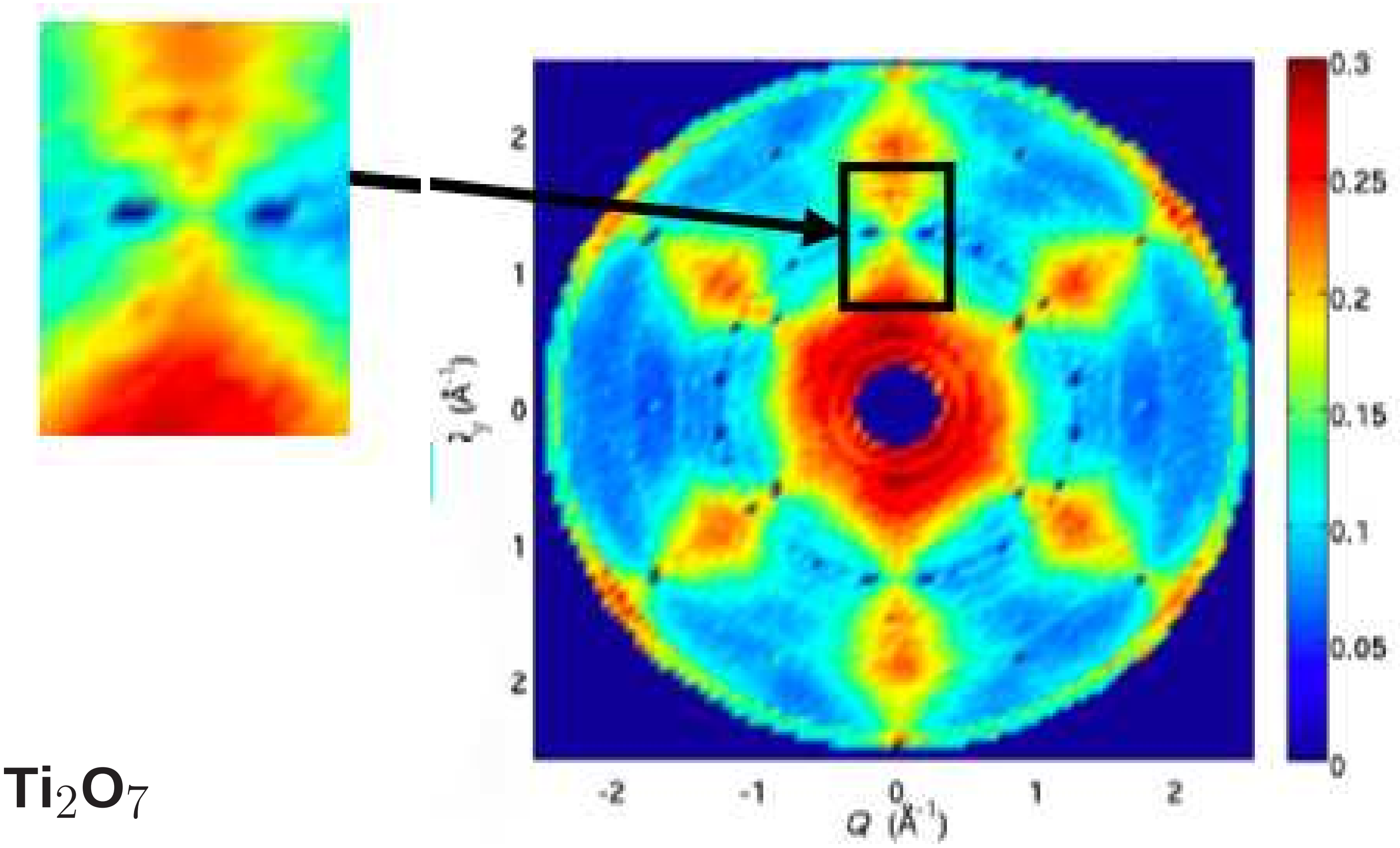


Entropic distribution: $P[\mathbf{B}(\mathbf{r})] \propto \exp(-\kappa \int \mathbf{B}^2(\mathbf{r})\mathrm{d}^3\mathbf{r})$

Power-law correlations: $\langle B_i(\mathbf{r}) B_j(\mathbf{0}) \rangle \propto r^{-3}$

Low T correlations from neutron diffraction

Bramwell and Harris, unpublished

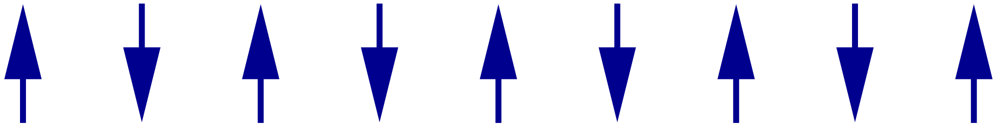


$Ho_2Ti_2O_7$

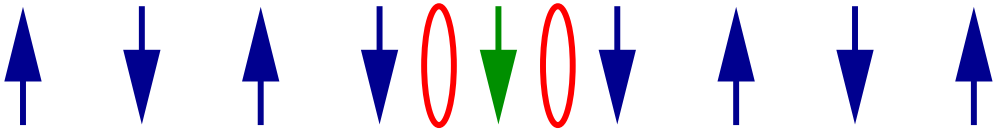
Classical fractionalised excitations

Fractionalisation in one dimension

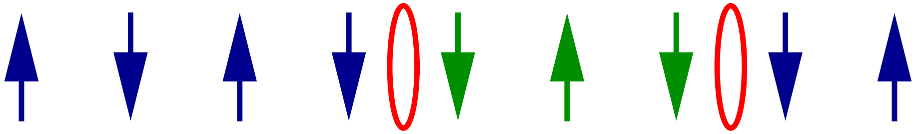
Ground state



An excited state



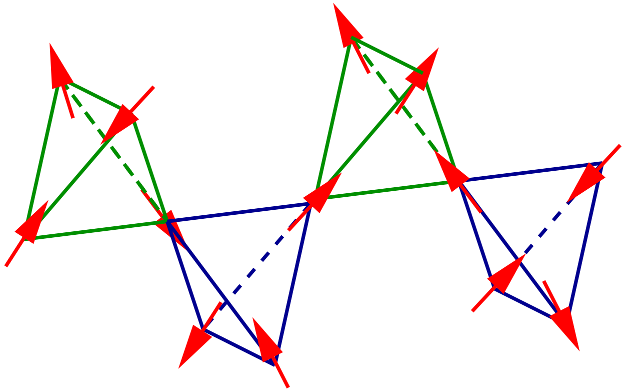
. . . two separated excitations



Fractionalisation in spin ice

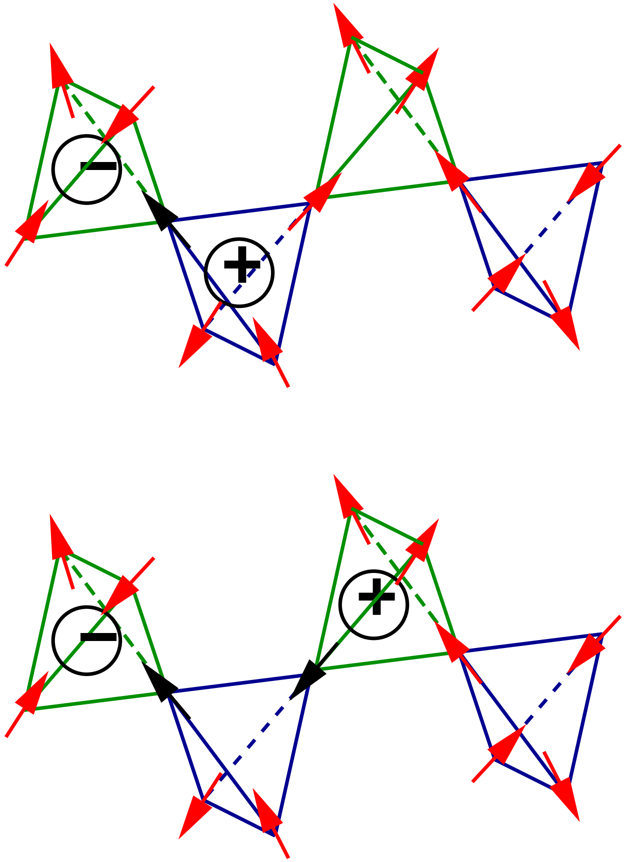
Monopole excitations

Ground state



Castelnovo, Moessner and Sondhi (2008)

Excited states

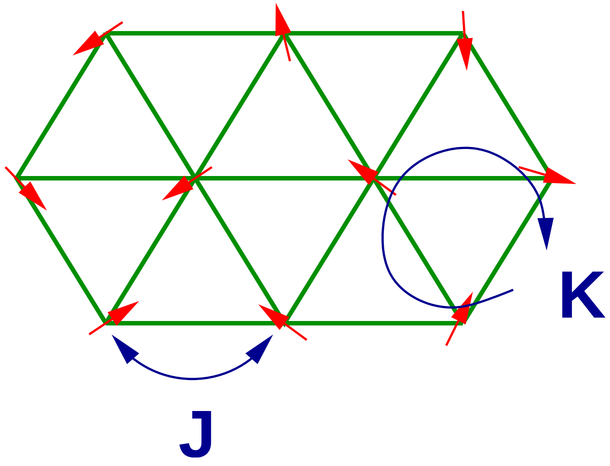


Candidate quantum Spin Liquids

κ-(ET)$_2$Cu$_2$(CN)$_3$

Interaction scale $J \sim 250$ **K**

No order to $T = 30$ **mK**

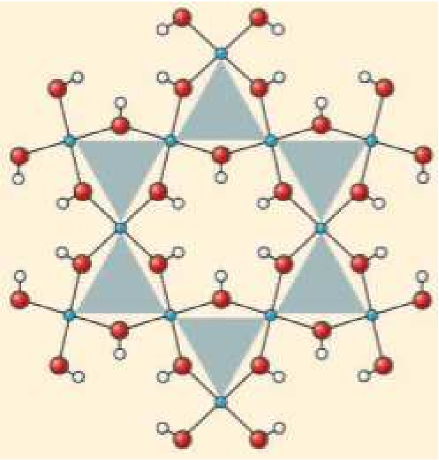


Herbertsmithite

ZnCu$_3$(OH)$_6$Cl$_2$

Interaction scale $J \sim 200$ **K**

No order to $T = 50$ **mK**

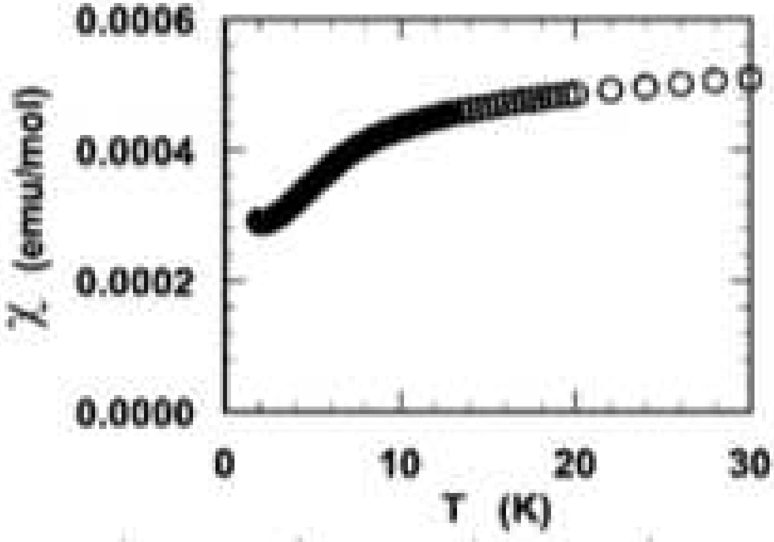


Metallic characteristics in an insulator

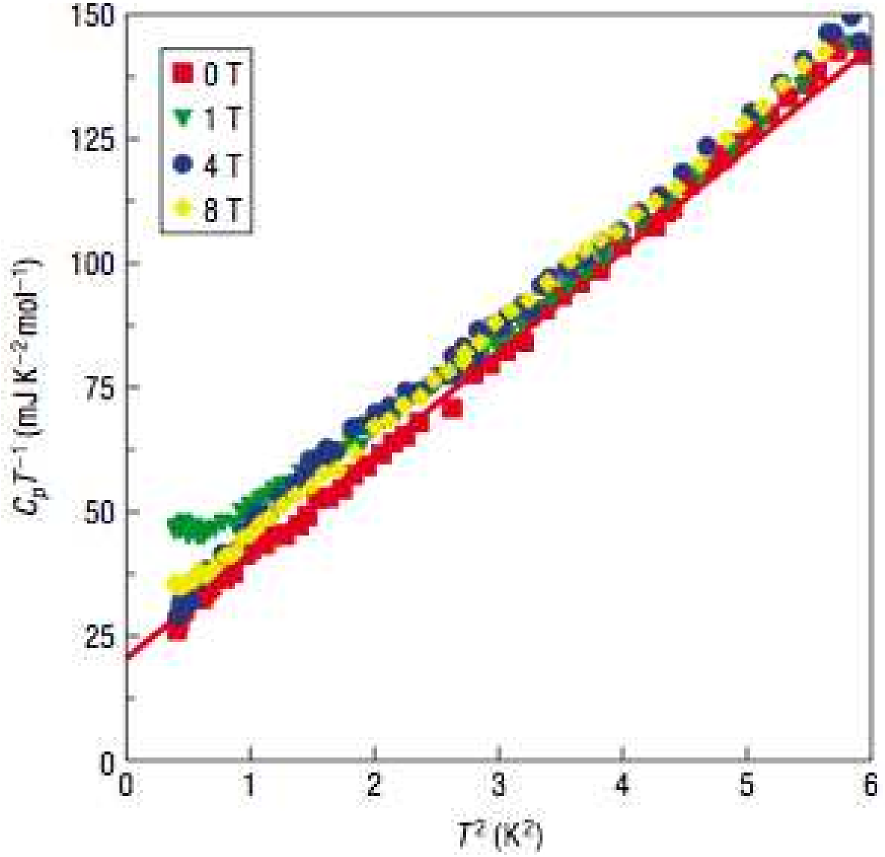
κ-**(ET)$_2$Cu$_2$(CN)$_3$**

Interaction scale $J \sim 250$ **K**

No order to $T = 30$ **mK**



Finite low-T susceptibility



Heat capacity $\sim aT + bT^3$

Summary

Geometric frustration

macroscopic classical degeneracies

long-range order avoided

Frustrated magnets at low T

soft modes and slow dynamics

emergent degrees of freedom

exotic excitations

Collaborators

M. J. Bhaseen

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P.C.W. Holdsworth

L. D. C. Jaubert

R. Moessner

T. S. Pickles

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E. F. Shender

S. E. Palmer

S. Powell

M. Y. Veillette