STATISTICAL PHYSICS OF

GEOMETRICALLY FRUSTRATED MAGNETS

Classical spin liquids, emergent gauge fields

and fractionalised excitations

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Outline

- **Geometrically frustrated magnets**

  Experimental signatures of frustration

- **Classical models**

  Degeneracy of under-constrained ground states

  Ground state selection: order from disorder

- **Low temperature correlations**

  Emergent degrees of freedom

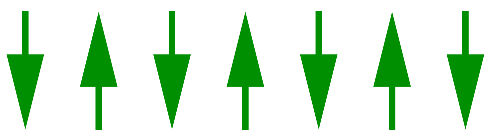
  Fractionalised excitations

Unfrustrated antiferromagnets: for contrast

Spin S Heisenberg antiferromagnet on simple cubic lattice

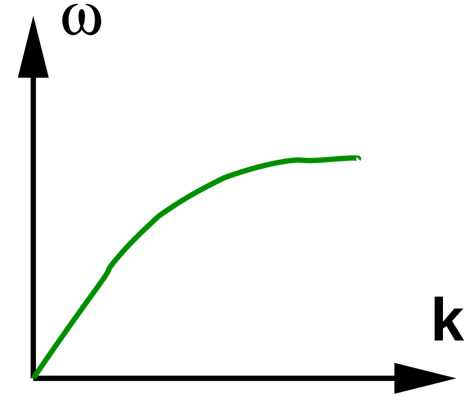
$$\mathcal{H} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

Classical ground state



unique up to symmetries

Excitation spectrum



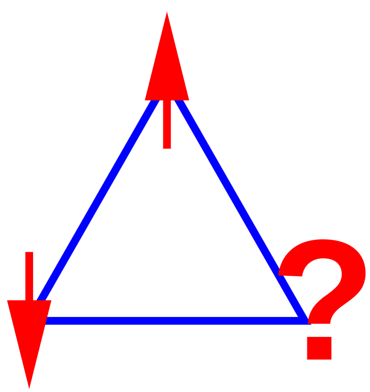
Sublattice magnetisation:

$$\langle S_i^z \rangle = S - \delta S \qquad \delta S \sim \sum_{\mathbf{k}} \frac{1}{\omega_{\mathbf{k}}} \left(\langle n_{\mathbf{k}} \rangle + \frac{1}{2} \right)$$

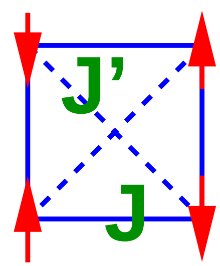
Types of frustration in magnetic systems

With quenched disorder

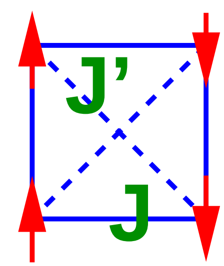
- in spin glasses



From competition



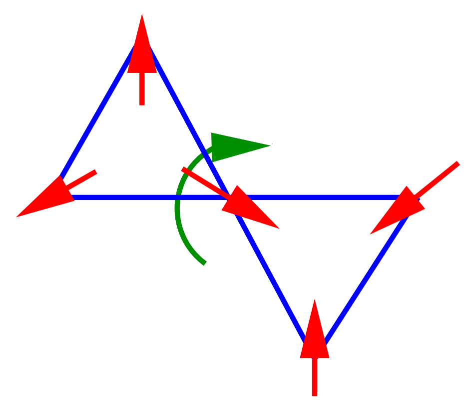
vs



From geometry

structure $\longrightarrow$ degeneracy

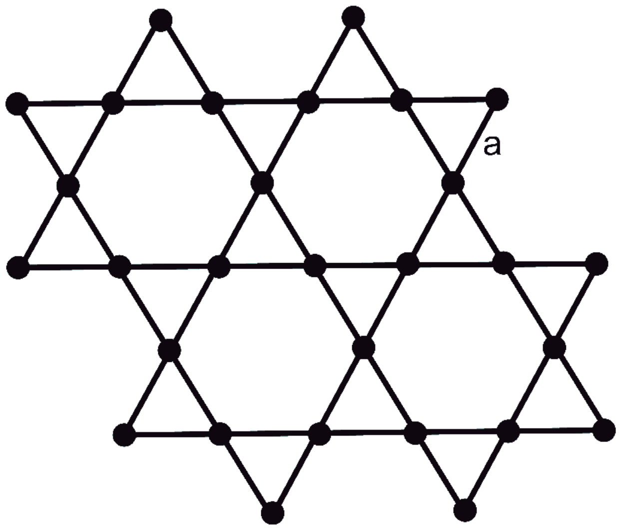
Anderson 1956, Villain 1977



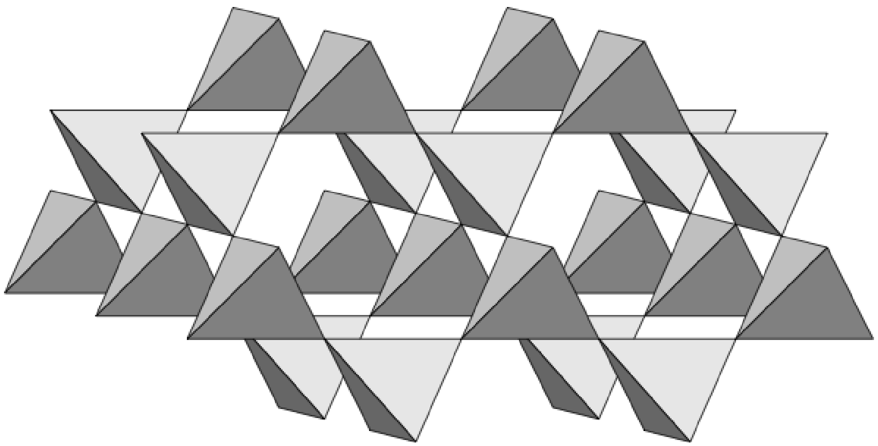
Examples of frustrated lattices

Building block: corner-sharing frustrated units

2D: kagome lattice



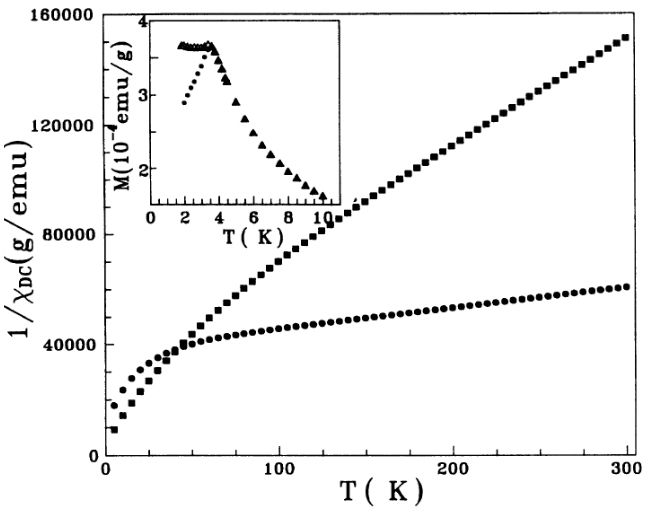
3D: pyrochlore lattice



Characteristics of geometrically frustrated antiferromagnets

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

Paramagnetic even for $T \ll |\Theta_{\mathrm{CW}}|$



χ^{-1} **vs** T

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

Mean field theory

$$\chi \propto \frac{1}{T-\Theta_{\mathrm{CW}}}$$

Selected examples of frustrated magnets

Layered materials

SCGO

pyrochlore slabs

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

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

Herbertsmithite

kagome layers

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

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

κ-ET

triangular layers

molecular $\quad S = 1/2$

$\Theta_{\mathrm{CW}} \sim -400\mathrm{K}$

Pyrochlore lattices

$ZnCr_2O_4$

pyrochlore Heisenberg

antiferromagnet

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

$\Theta_{\mathrm{CW}} \sim -390\mathrm{K} \quad T_{\mathrm{N}} \sim 12.5\mathrm{K}$

Spin ices

$Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$

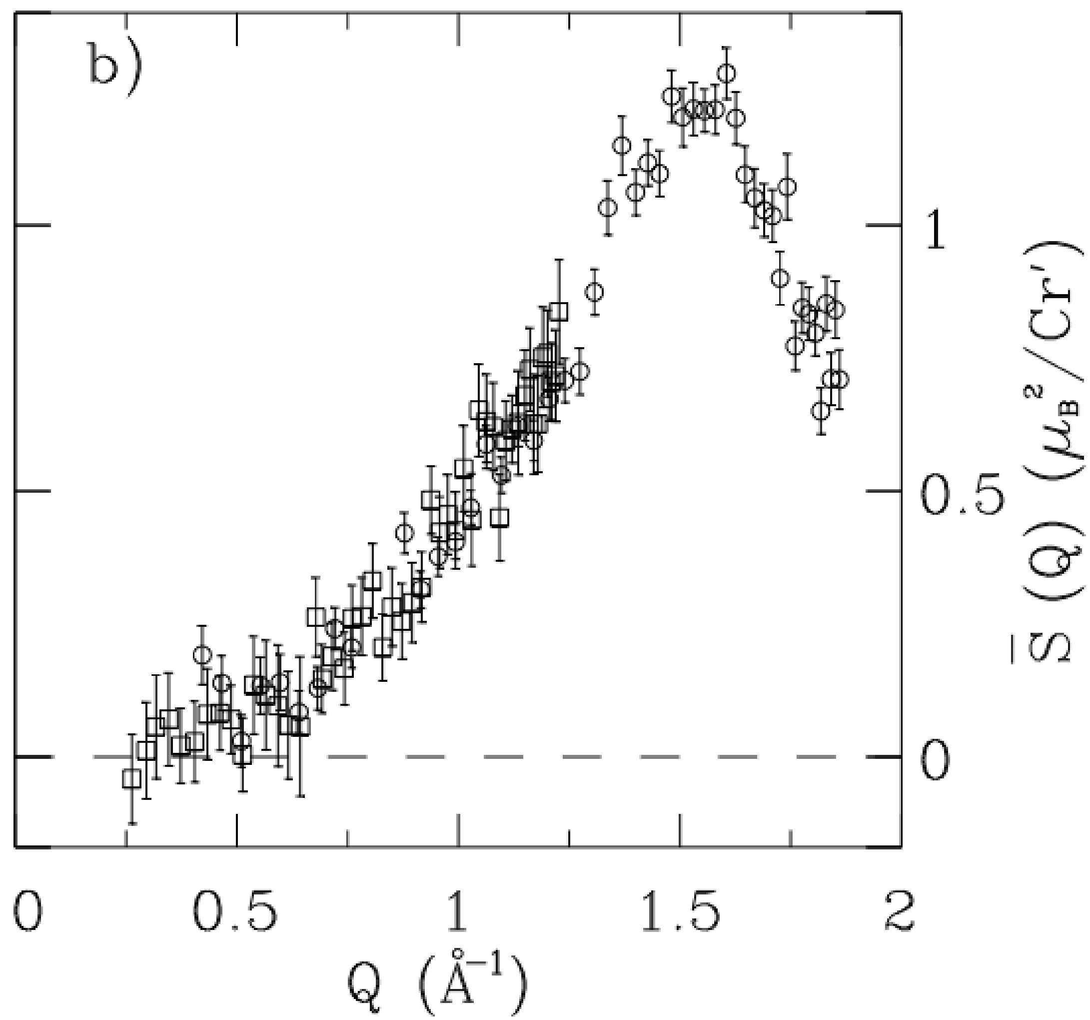
ferromagnets with single-ion anisotropy

— hence frustration

Effective description:
pyrochlore Ising antferromagnet

$J_{\mathrm{eff}} \sim +2\mathrm{K}$

Spin correlations: $S(Q)$ for SCGO (powder at 1.5 K)

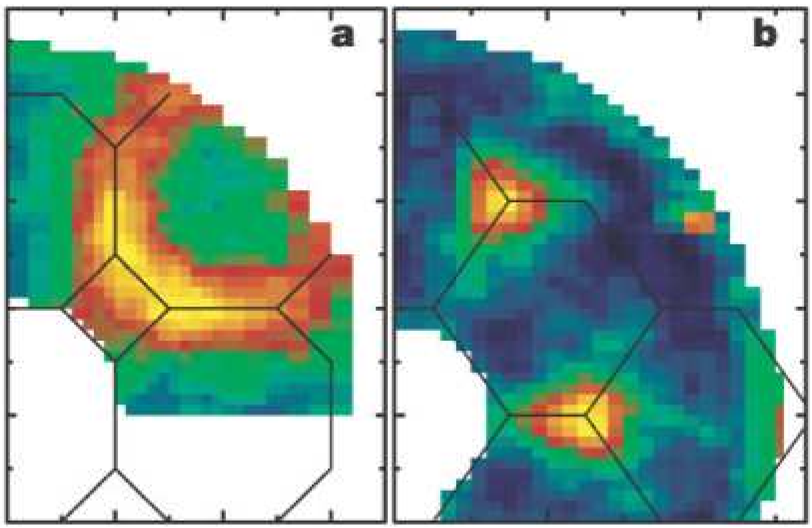


S-H Lee *et al.*, EPL 35, 127 (1996)

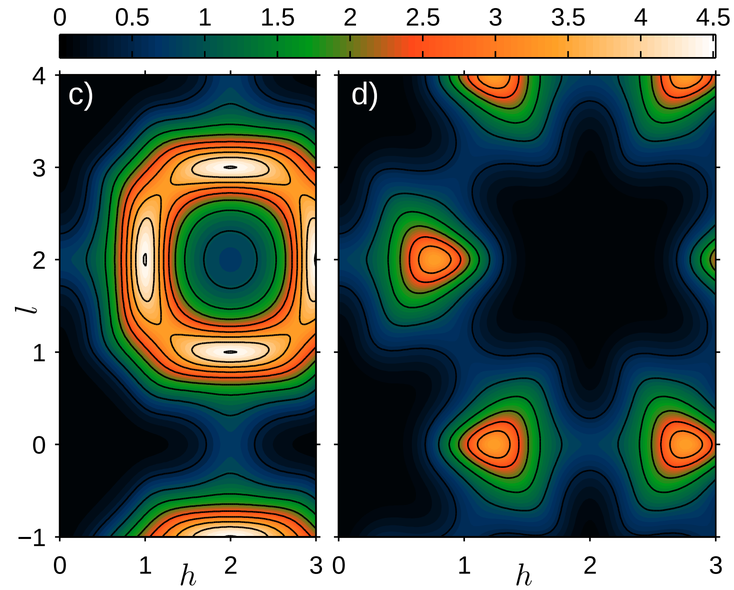
Low-T spin correlations in $ZnCr_2O_4$

$S(q)$ in $(h0l)$ and (hhl) scattering planes

Experiment



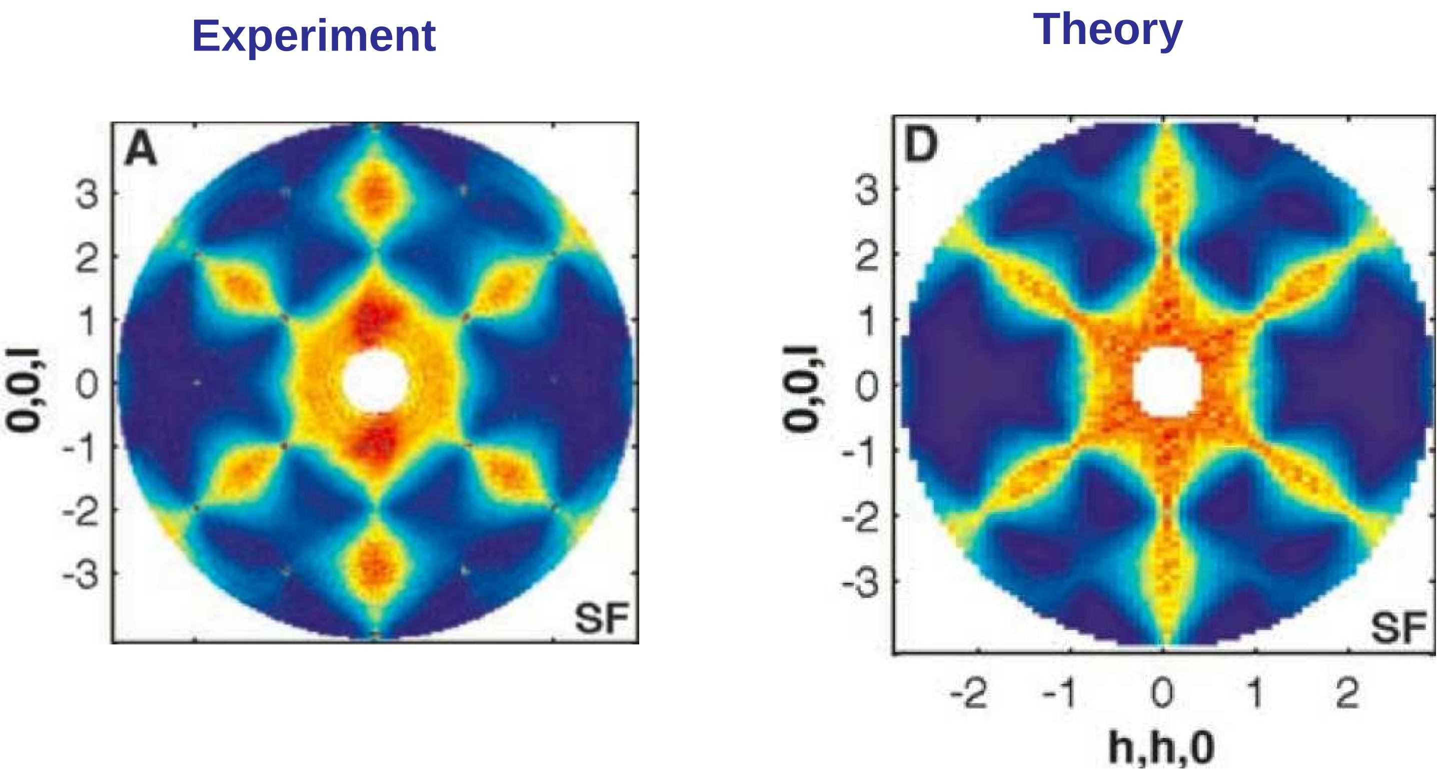
Theory



Lee *et al*, Nature (2002)

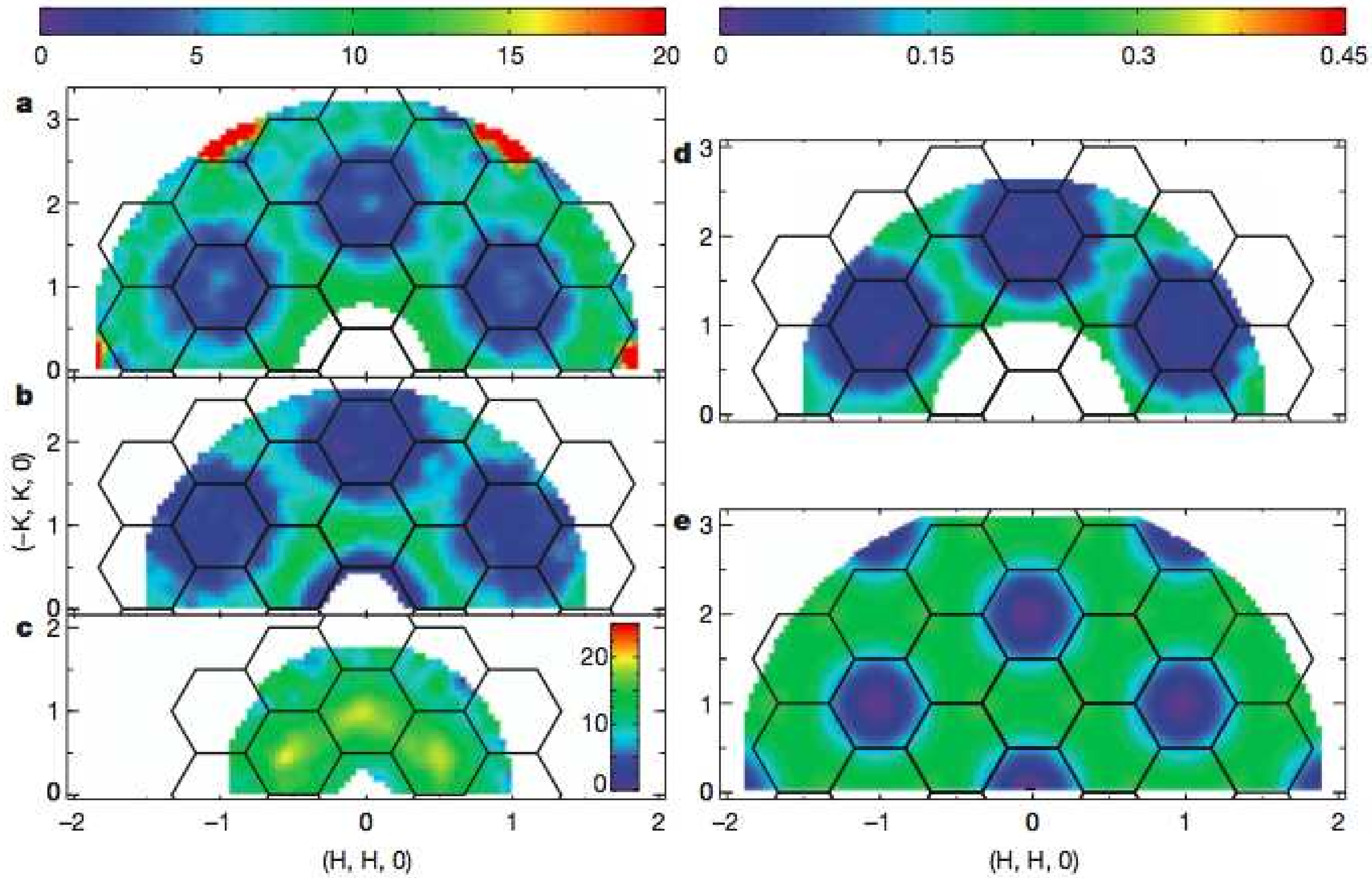
Conlon + JTC (2010)

Spin correlations: spin ice single crystal



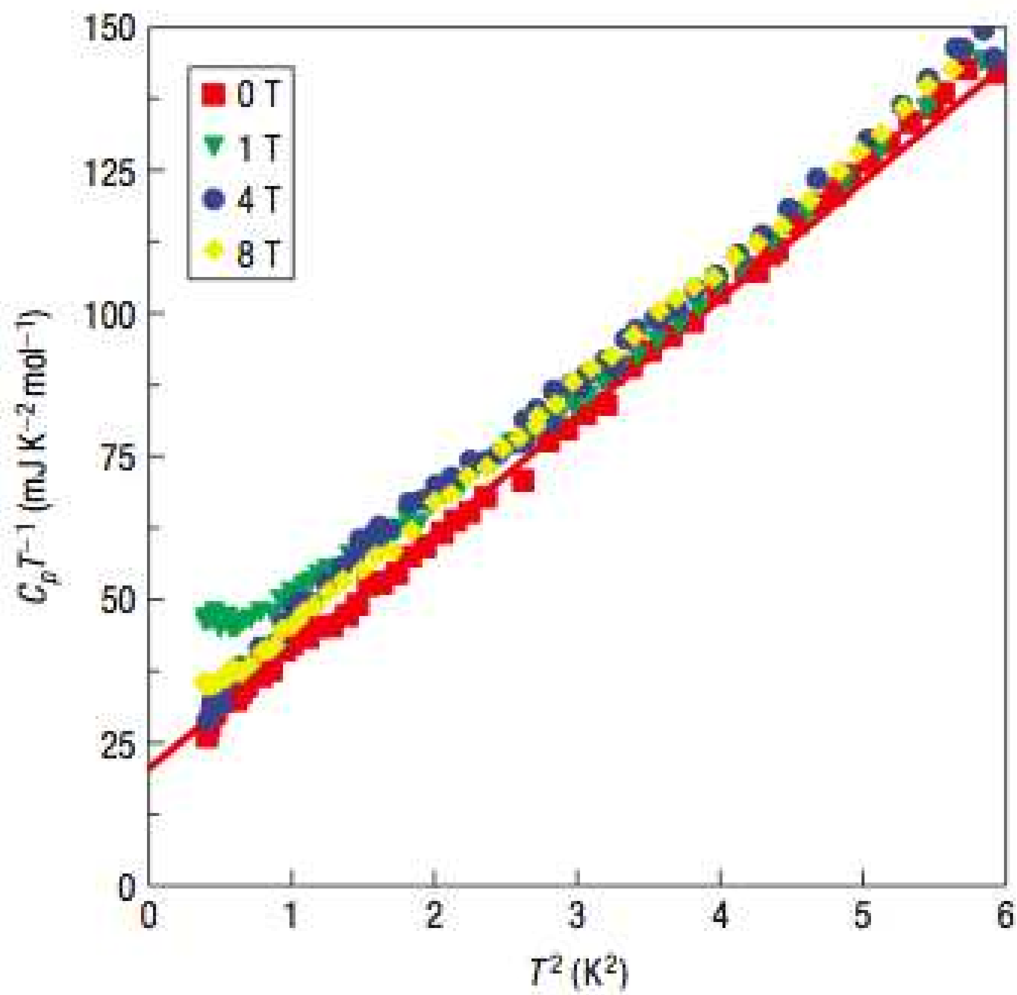
Fennell et al, Science 326 415 (2009)

Excitations: inelastic neutron scattering from herbertsmithite



Han *et al.*, Nature 492, 406 (2012)

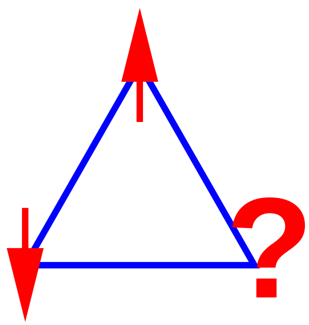
Excitations: heat capacity of κ(BEDT-TTF)$_2$Cu$_2$(CN)$_3$



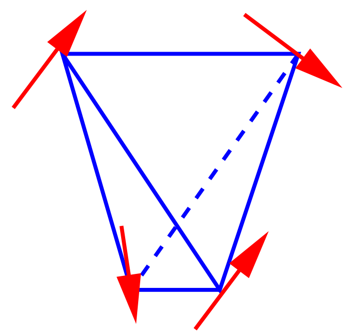
C/T vs T^2. Yamashita *et al.*, Nature Phys. 4, 459 (2008).

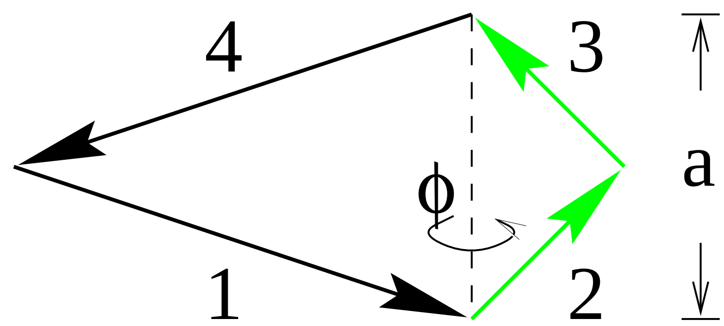
Antiferromagnetic spin clusters
- frustration and degeneracy

Ising triangle



Heisenberg tetrahedron





General problem: simplex of q spins, each with n components

$$\mathcal{H} = J \sum_{\text{pairs}} \mathbf{S}_i \cdot \mathbf{S}_j \equiv \frac{J}{2} |\mathbf{L}|^2 + c \qquad \text{with} \qquad \mathbf{L} = \sum_{i=1}^{q} \mathbf{S}_i$$

Ground state degeneracy in Heisenberg AFM

Maxwellian constraint-counting

Example: Heisenberg pyrochlore antiferromagnet

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

Total number of degrees of freedom: $F = 2 \times \left(\text{number of spins}\right)$

Constraints satisfied in ground state: $K = 3 \times \left(\text{number of units}\right)$

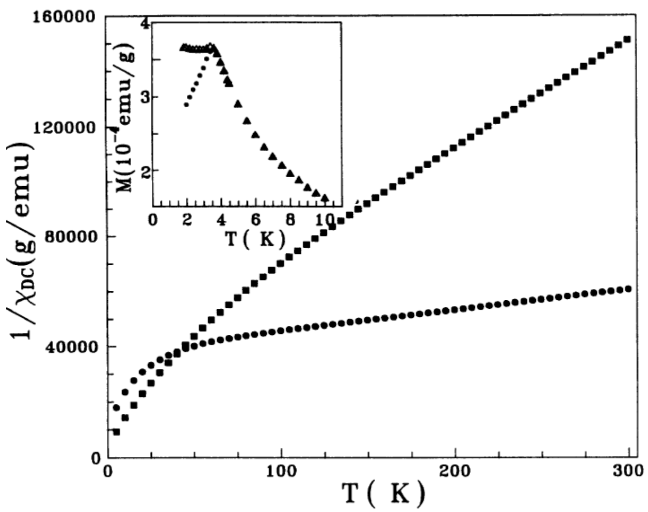
Ground state dimension:

D=F-K

Geometric Frustration $\rightarrow$ Macroscopic D

Consequences of degeneracy: SCGO

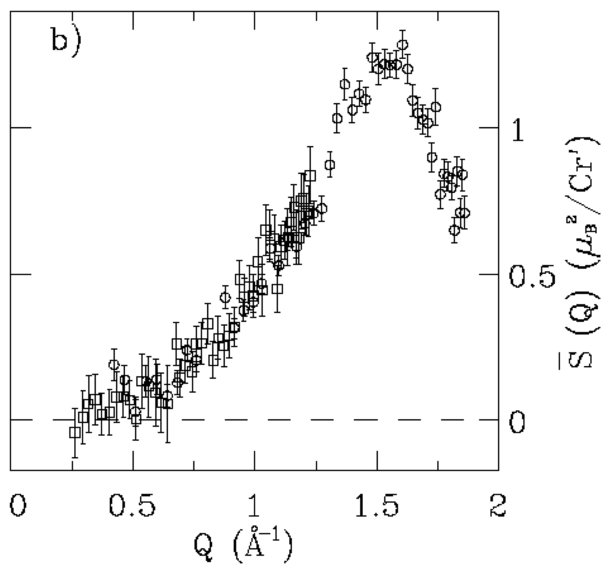
Paramagnetic even for $T \ll |\Theta_{CW}|$



χ^{-1} vs T

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

Strong short-range correlations

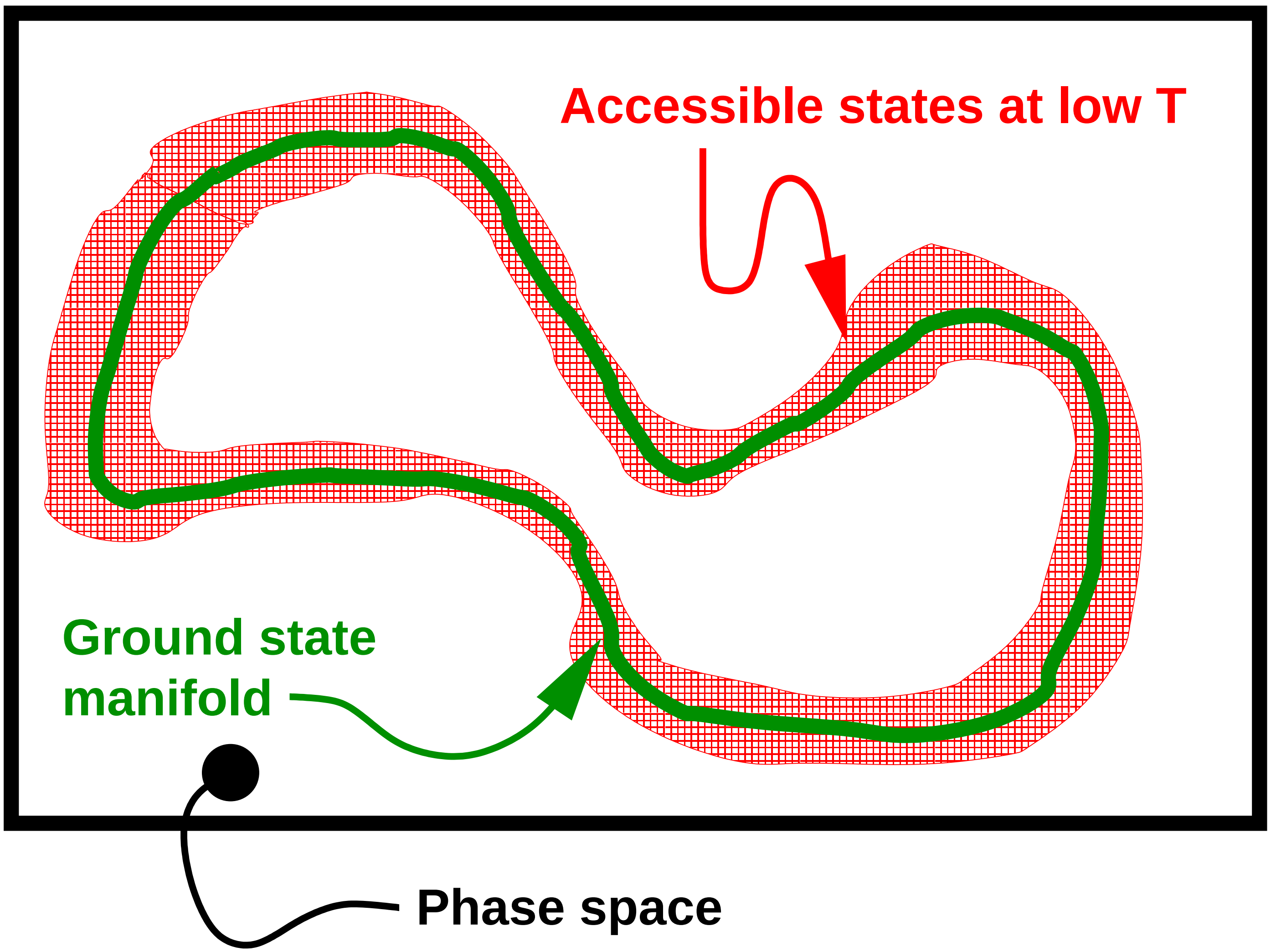


Elastic neutron scattering

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

Schematics of behaviour at low temperature

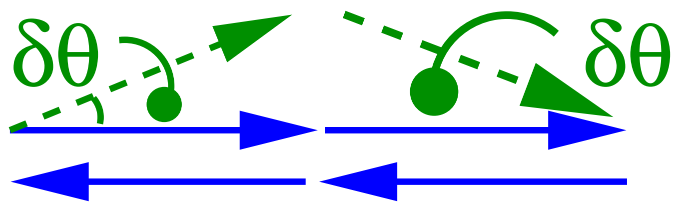
Classical cooperative paramagnet: $JS \ll k_{\rm B}T \ll JS^2$



Ground state selection by fluctuations?

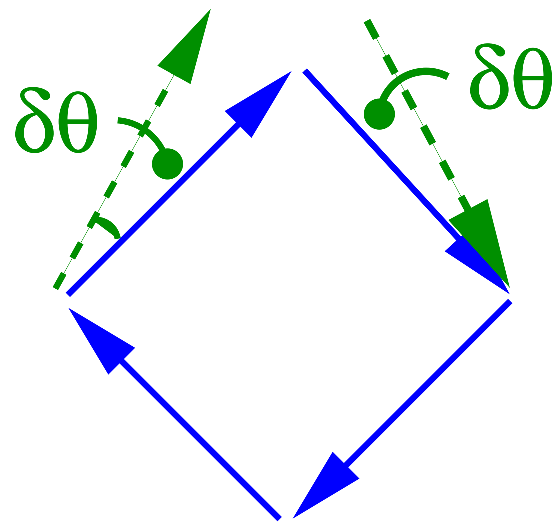
'Order by disorder' **Villain (1980), Shender (1982)**

Some states have soft modes



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

Others don't



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

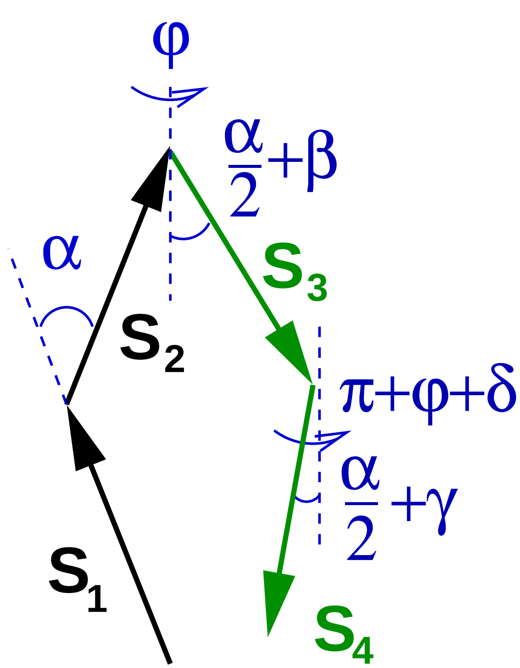
Order by disorder: four XY or Heisenberg spins

Thermal distribution of $S_1 \cdot S_2$?

Integrate out S_3 and S_4

$$P(\alpha) \propto \left\{ \begin{array}{c} \sin\alpha \\ 1 \end{array} \right\} \mathrm{d}\alpha\, \mathcal{Z}(\alpha)$$

$$\mathcal{Z}(\alpha) = \int \mathrm{d}\vec{S}_3\, \mathrm{d}\vec{S}_4\, \exp(-\tfrac{\beta J}{2}|\vec{S}_3 + \vec{S}_4 + 2\hat{z}\cos(\alpha/2)|^2)$$



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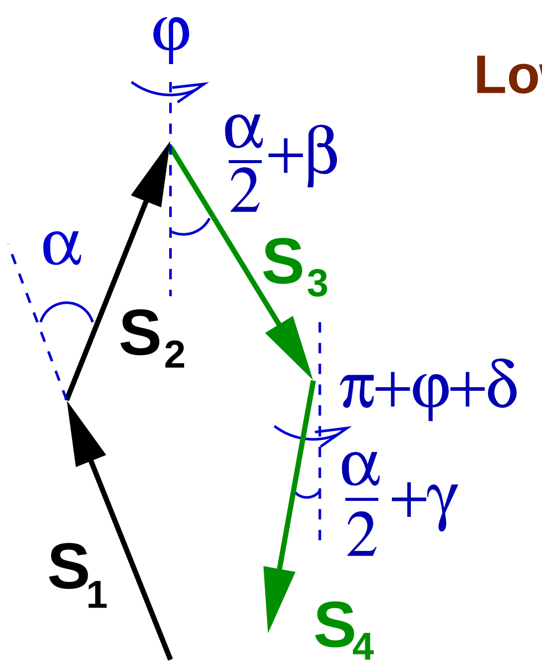


Low temperature limit:

Heisenberg $P(\alpha) \propto \sin(\alpha/2)$

— no order

XY $P(\alpha) \propto 1/\sin(\alpha)$

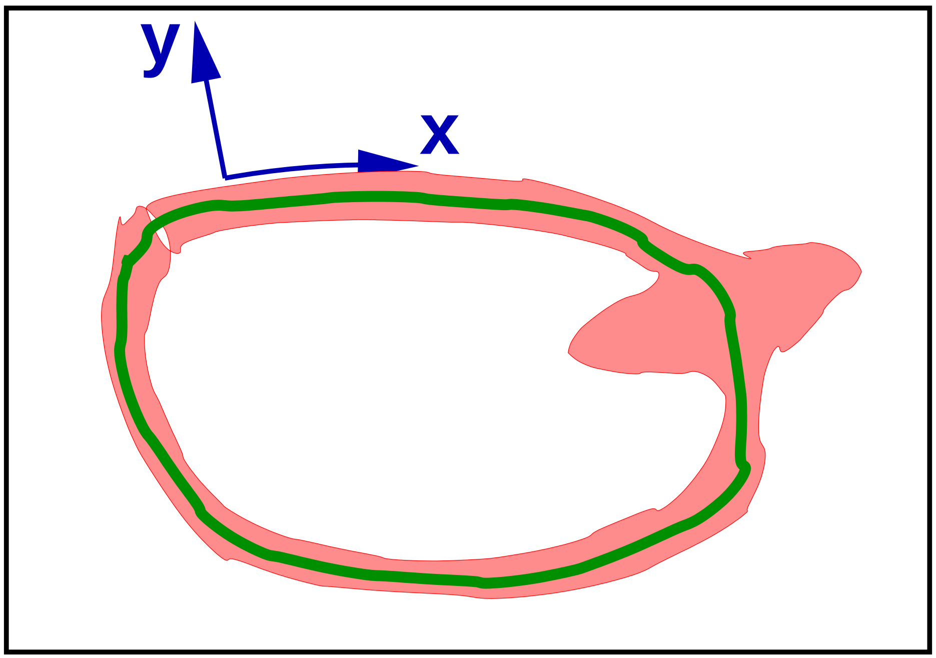
— collinear order

Ground state selection in general?

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

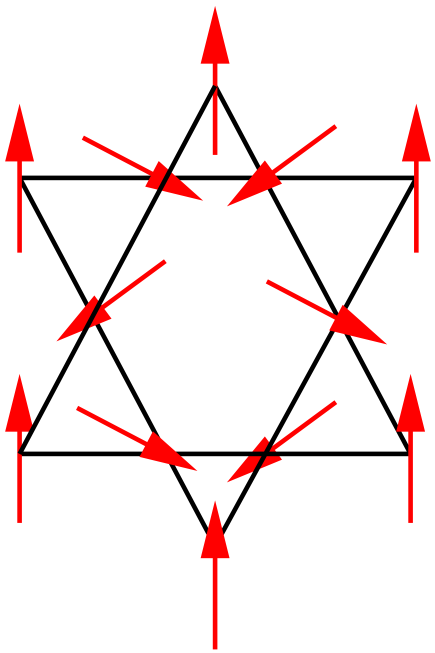
kagome $\rightarrow$ coplanar

pyrochlore $\rightarrow$ disordered

Order by disorder in the kagome Heisenberg model

Coplanar spin configurations have soft modes

Coplanar states



soft modes

Generic states

Constraint counting

$$\begin{aligned} F &= 2(\#\textbf{spins}) \\ &= 2 \times \tfrac{3}{2}(\#\textbf{triangles}) \\ K &= 3(\#\textbf{triangles}) \\ D &= F - K = 0 \end{aligned}$$

– no soft modes

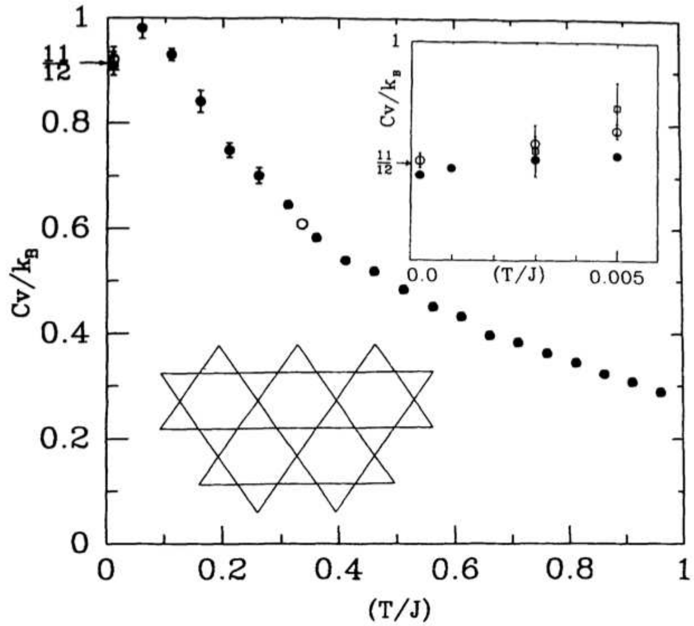
Coplanar states selected

Soft modes in MC simulations

Equipartition: mode with $E \propto y^2$ contributes $\frac{k_B}{2}$ to heat capacity

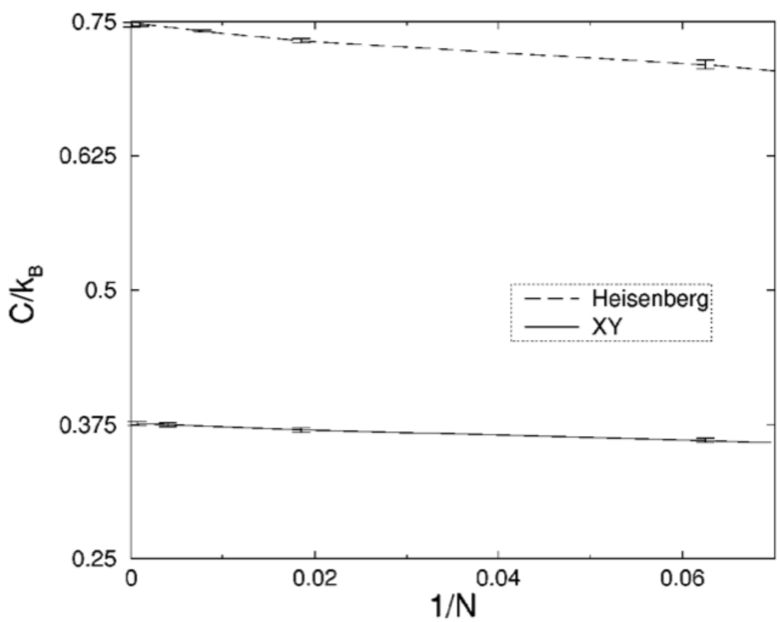
mode with $E \propto y^4$ contributes $\frac{k_B}{4}$

Kagome Heisenberg model



Co-planar states: $1/6$ of modes are quartic

Pyrochlore Heisenberg & XY models



Heisenberg: $1/4$ of modes cost zero energy

XY: $1/4$ of modes are quartic

Ground state selection?

Quantum fluctuations

Zero-point energy of stiff modes $\Rightarrow$ Effective Hamiltonian for soft degrees of freedom

$$\mathcal{H}_{\rm eff}(\mathbf{x}) = \frac{1}{2}\sum_l \hbar\omega_l(\mathbf{x})$$

Expected consequences:

- **Large** S

Minimise zero-point energy in ordered (collinear/coplanar) state

- **Small** S

Delocalisation over classical ground state manifold $\Rightarrow$ spin liquid

Spin Ice and Ising pyrochlore AFMs

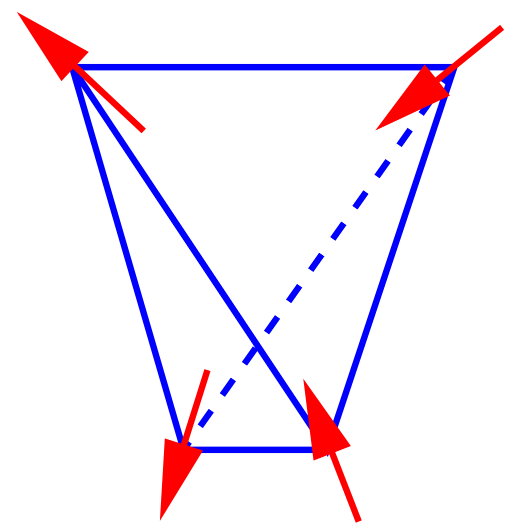
Pyrochlore ferromagnet with single-ion anisotropy

$$\mathcal{H} = -J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - D \sum_i (\hat{\mathbf{n}}_i \cdot \mathbf{S}_i)^2 - \mathbf{h} \cdot \sum_i \mathbf{S}_i$$

Large D: $\mathbf{S}_i = \sigma_i \hat{\mathbf{n}}_\mathbf{i}$ $\sigma_i = \pm 1$ $J_{\rm eff} = -J \hat{\mathbf{n}}_\mathbf{i} \cdot \hat{\mathbf{n}}_j$ $h_i^{\rm eff} = \mathbf{h} \cdot \hat{\mathbf{n}}_i$

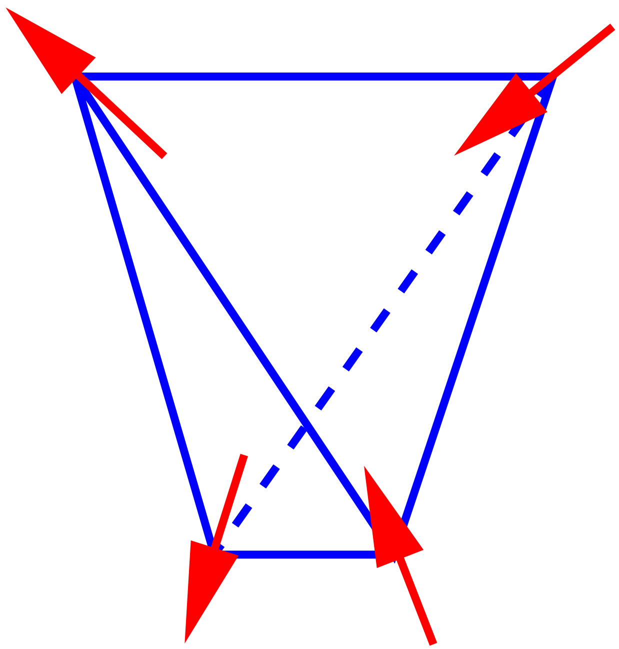
$$\mathcal{H} = J_{\rm eff} \sum_{\langle ij \rangle} \sigma_i \sigma_j - \sum_i h_i^{\rm eff} \sigma_i$$

Effective Ising system



Frustration and residual entropy

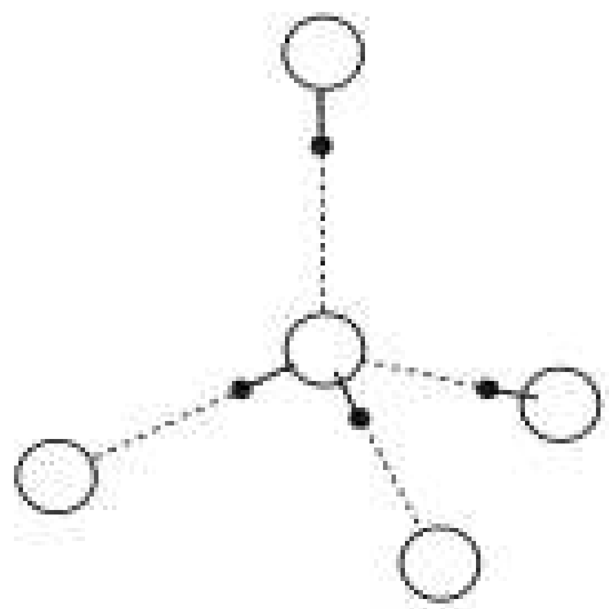
Spin ice



Anisotropy +

ferromagnetic exchange

Water ice



Pauling 1935

Ground states: 'two-in, two-out'

Pauling's entropy estimate

One tetrahedron

Total number of states: 16

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

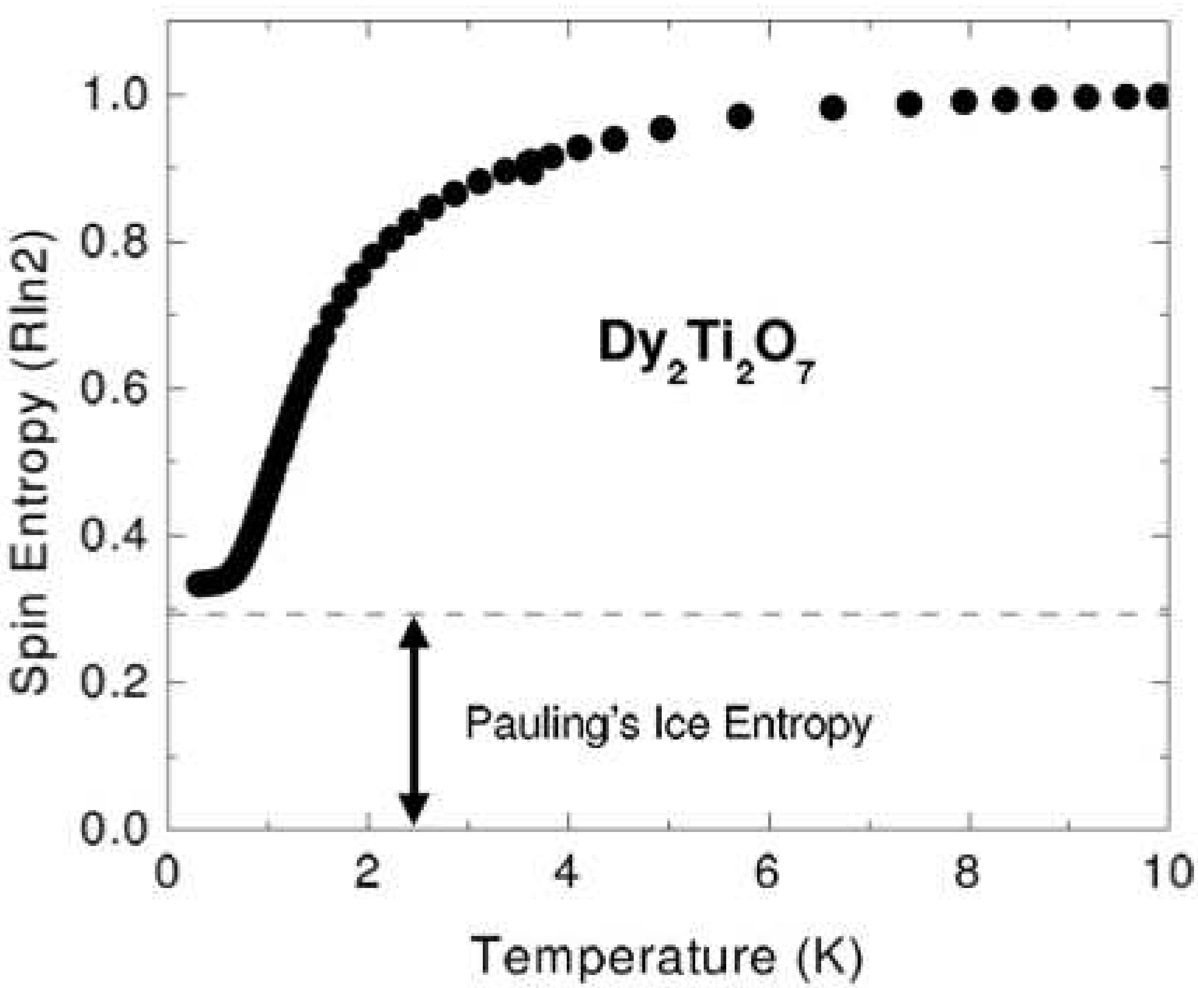
Pyrochlore lattice

Estimate for number of ground states:

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

$$= \left(\frac{3}{2}\right)^{(\#\,\text{spins}/2)}$$

Pauling entropy in experiment



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

Summary

Geometric frustration

leads to macroscopic classical ground state degeneracy

possibility of order-by-disorder

. . . **but long-range order may be avoided**

At low T: strong correlations + large fluctuations