Title: Correlations, Fluctuations and Constraints: The Statistical Physics of Classical Models for Geometrically Frustrated Magnets

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Abstract: TBA

GEOMETRICALLY FRUSTRATED MAGNETS

Correlations and Excitations

in Classical Spin Liquids

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Physics Department, Oxford University

Outline

Geometrically frustrated magnets

- Experimental signatures of frustration

Classical models

- Degeneracy of under-constrained ground states
- Ground state correlations and Coulomb phases

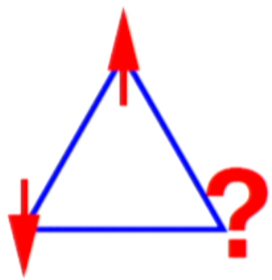
Order and disorder

- Transitions from the Coulomb phase
- Quenched disorder in geometrically frustrated magnets

Types of frustration in magnetic systems

With quenched disorder

- in spin glasses

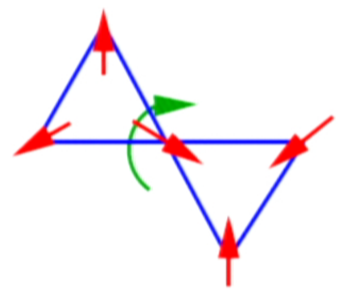


From competition

From geometry

structure ⟶ degeneracy

Anderson 1956, Villain 1977



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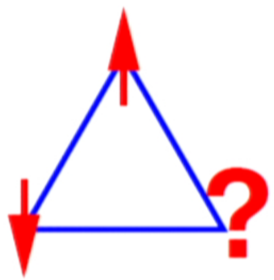

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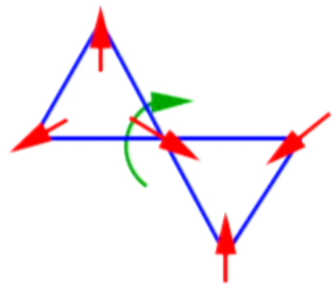
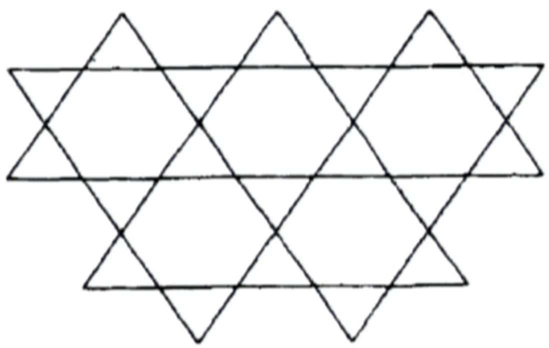


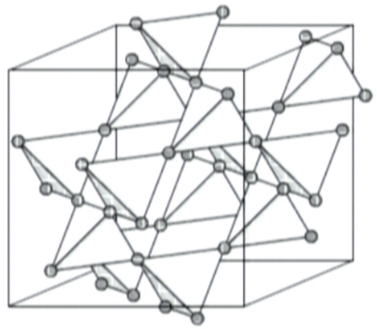
Examples of frustrated lattices

Building block: corner-sharing frustrated units

2D: kagome lattice



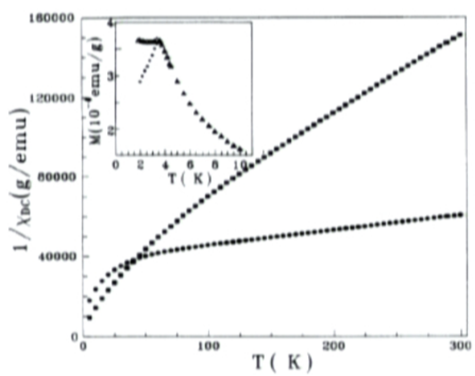
3D: pyrochlore lattice



Characteristics of geometrically frustrated antiferromagnets

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

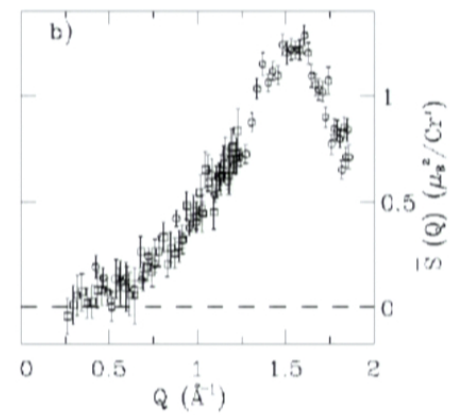
Paramagnetic even for $T \ll |\Theta_{CW}|$



χ^{-1} vs T

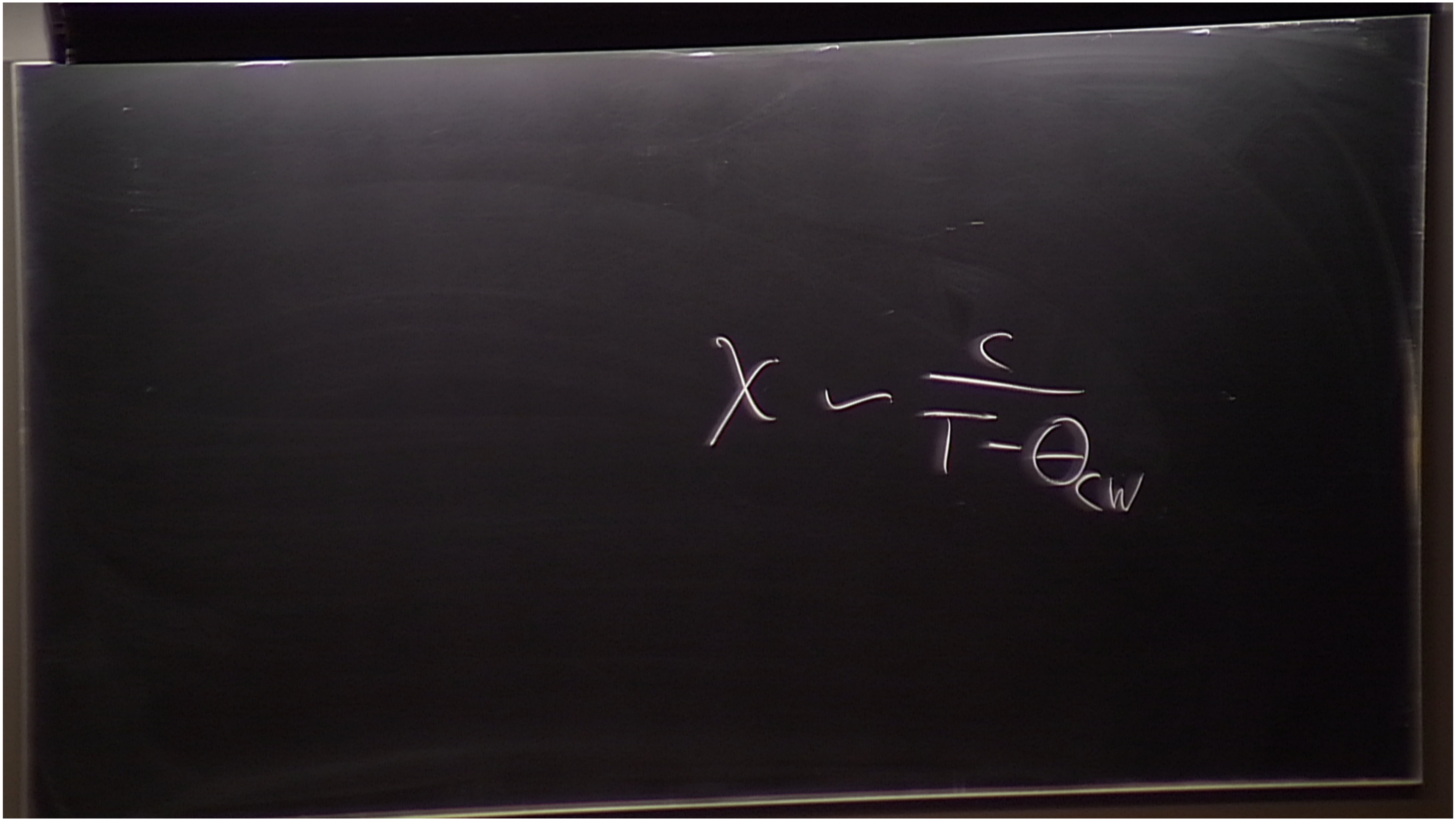
Martinez et al, PRB **46**, 10786 (1992)

Strong short-range correlations



Elastic neutron scattering

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


$\chi \sim \frac{C}{T-\Theta_{CW}}$

Selected examples of frustrated magnets

Layered materials

SCGO

pyrochlore slabs

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

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

hydromium iron jarosite

kagome layers

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

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

Herbertsmithite

kagome layers

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

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

Pyrochlore antiferromagnets

$Y_2Mo_2O_7$

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

$\Theta_{\rm CW} \sim 200{\rm K} \qquad T_{\rm F} \sim 22{\rm K}$

Spin ice materials

$Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$

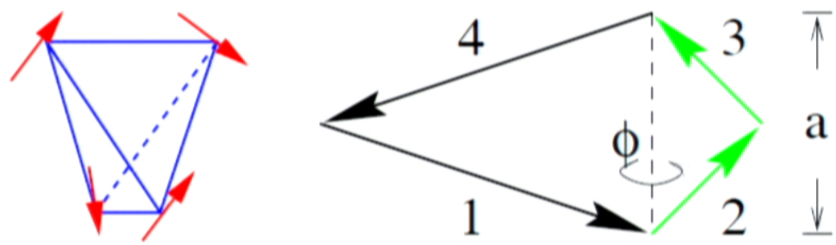
ferromagnets with single-ion anisotropy
– hence frustration

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

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 HFMs

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: $F = 2 \times (\text{number of spins})$

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

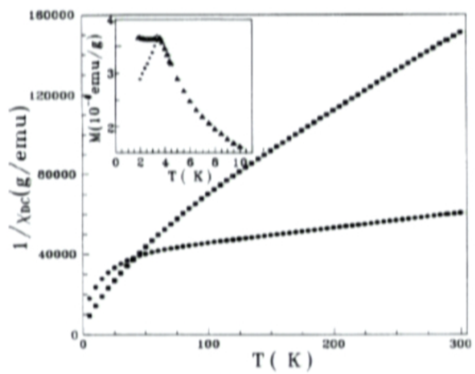
Ground state dimension:

D=F-K

Geometric Frustration $\longrightarrow$ **Macroscopic** D

Consequences of degeneracy: SCGO

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



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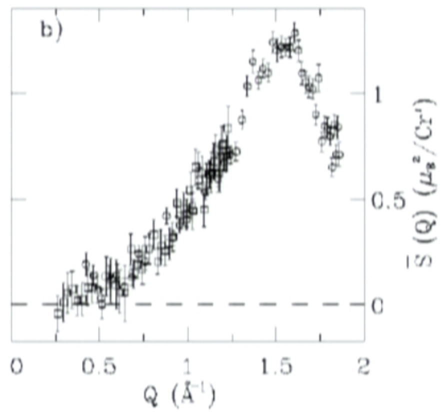

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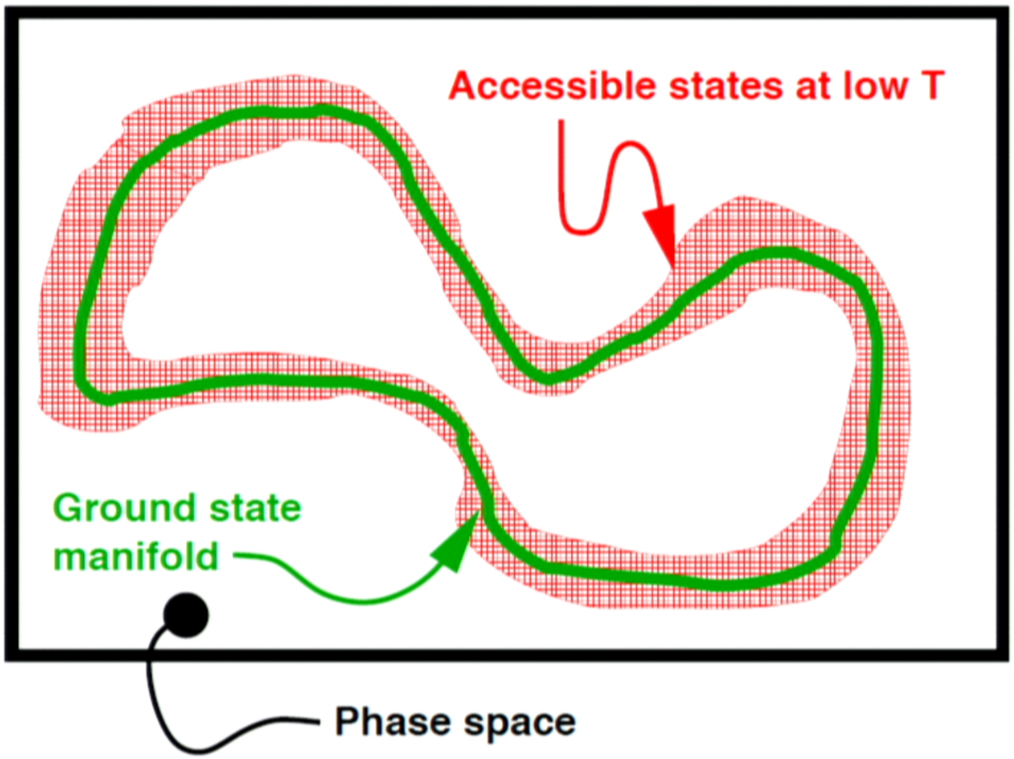
Schematics of behaviour at low temperature

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



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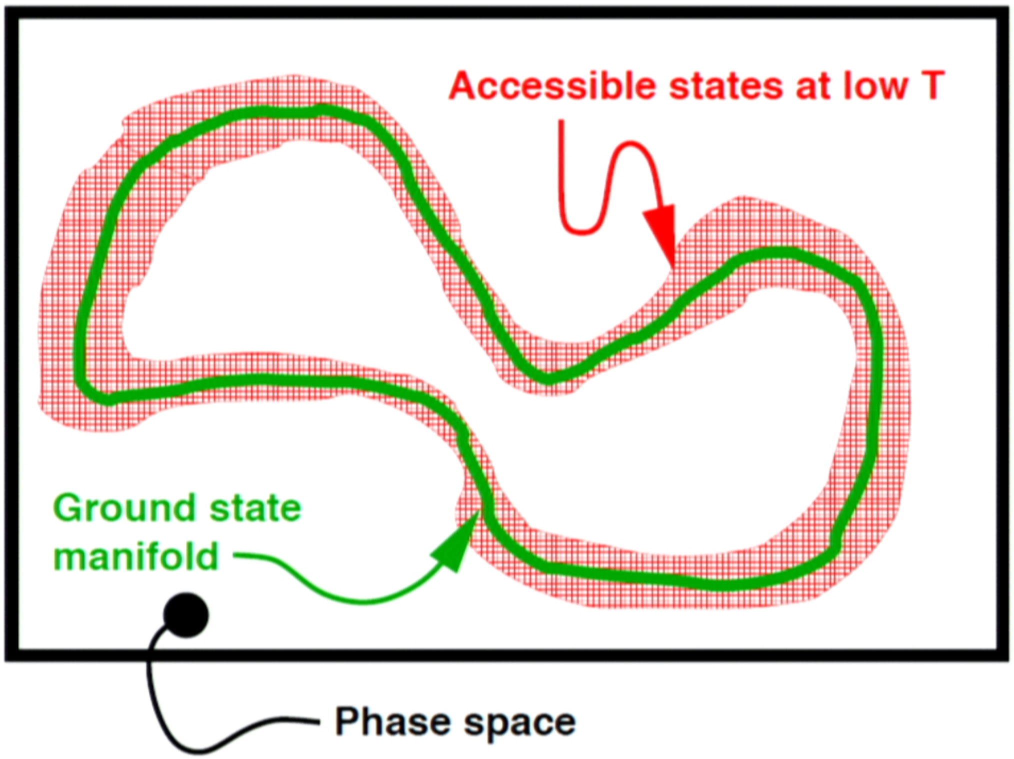


Spin Ice and Ising pyrochlore AFMs

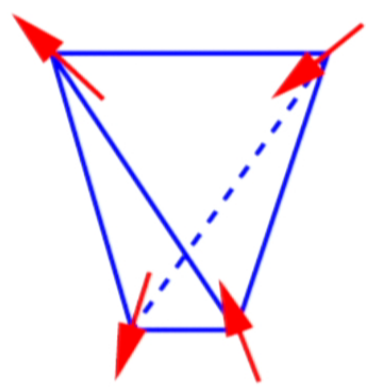
Pyrochlore ferromagnet with single-ion anisotropy

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

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

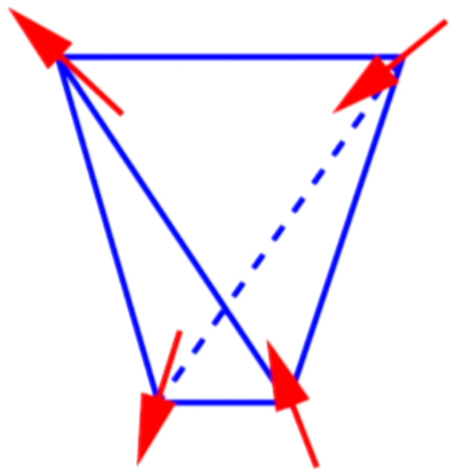
$$\mathcal{H} = J_{\text{eff}} \sum_{\langle ij\rangle} \sigma_i \sigma_j - \sum_i h_i^{\text{eff}} \sigma_i$$

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

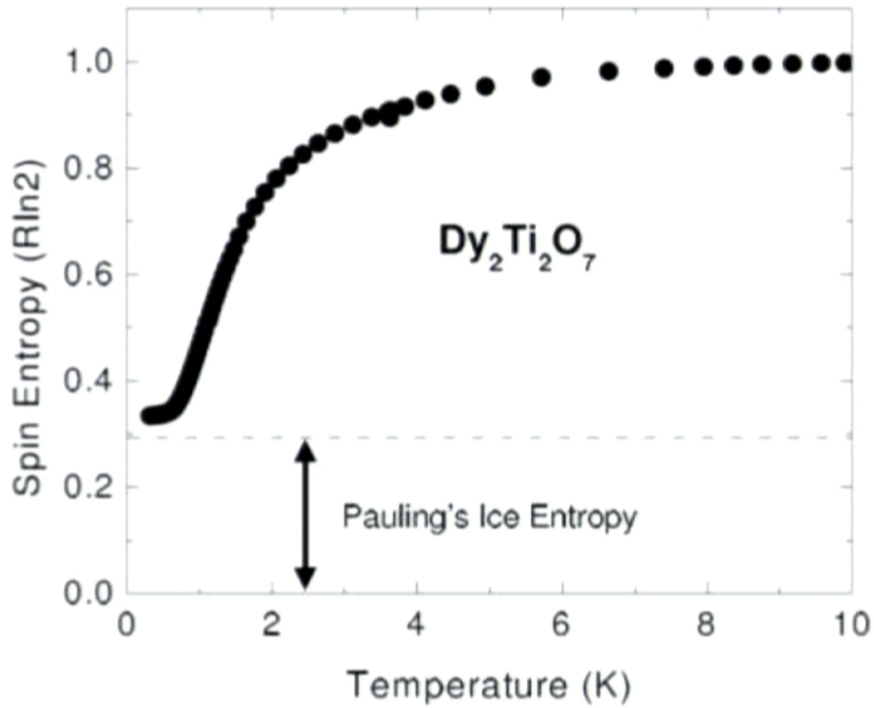
Fraction that are ground states: $\frac{6}{16}$

Pyrochlore lattice

Estimate for number of ground states:

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

Pauling entropy in experiment



$Dy_2Ti_2O_7$, **Ramirez *et al*, Nature 399, 333 (1999).**

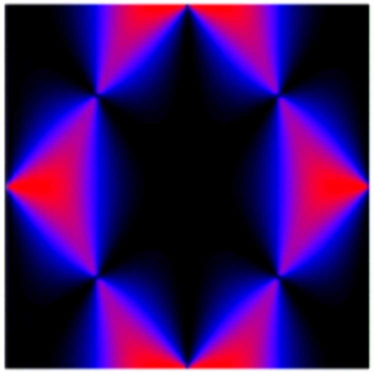
Correlations induced by ground state constraints

Local constraints

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

Long range correlations

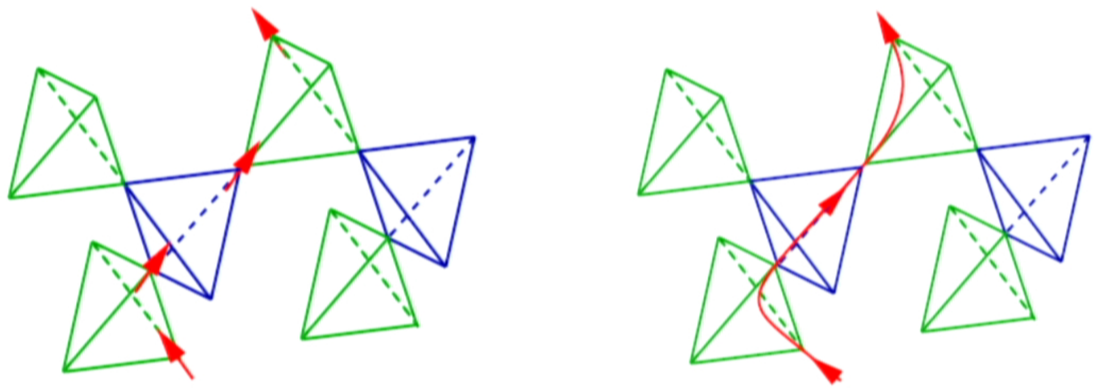
Sharp structure in $\langle \mathbf{S}_{-\mathbf{q}} \cdot \mathbf{S}_{\mathbf{q}} \rangle$



Gauge theory of ground state correlations

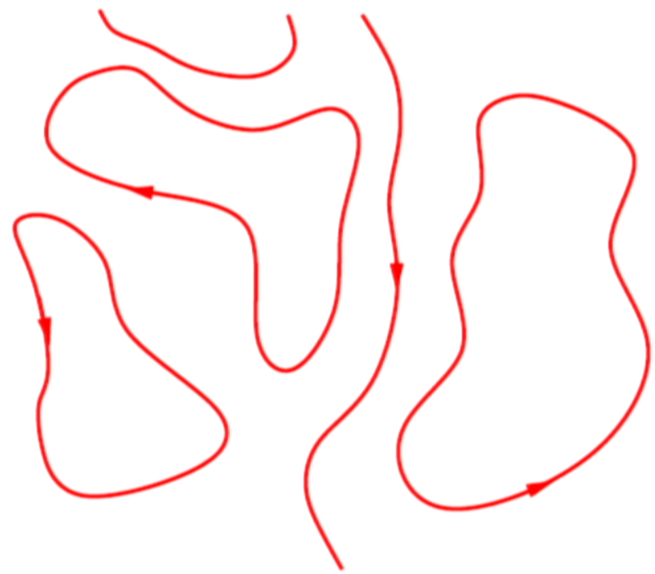
Youngblood *et al* (1980), Huse *et al* (2003), Henley (2004)

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



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

Ground states as flux loops

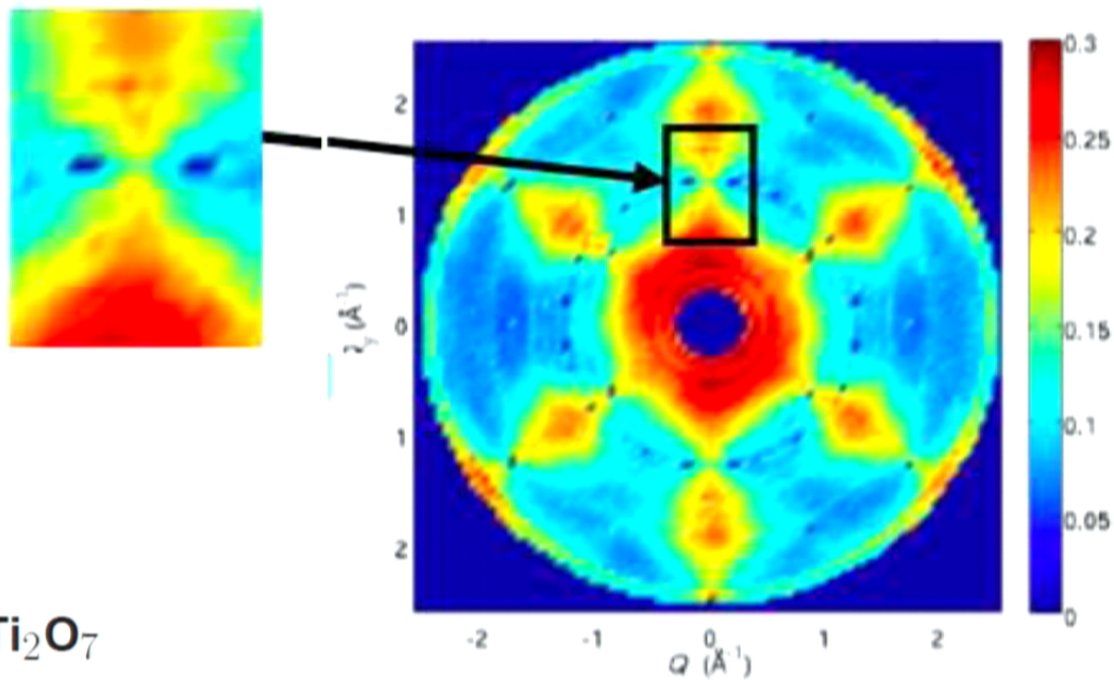


Entropic distribution: $P[\mathbf{B}(\mathbf{r})] \propto \exp(-\kappa \int \mathbf{B}^2(\mathbf{r}) \mathrm{d}^3\mathbf{r})$

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

Low T correlations from neutron diffraction

Fennell, Bramwell and collaborators (2009)

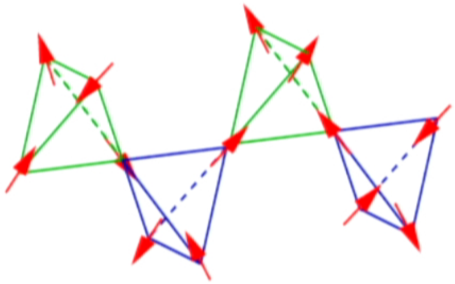


$Ho_2Ti_2O_7$

Monopoles in spin ice

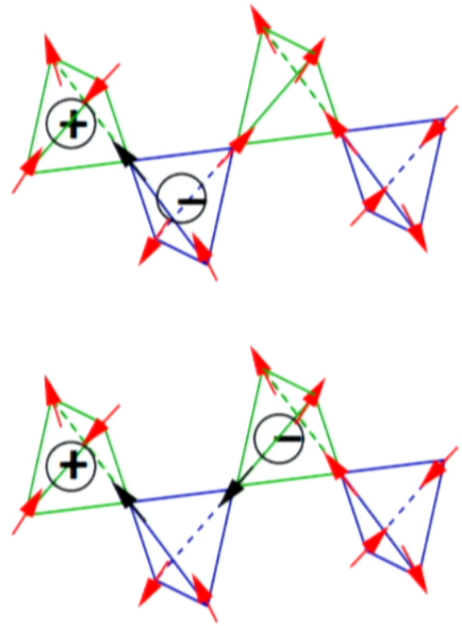
Monopole excitations

Ground state



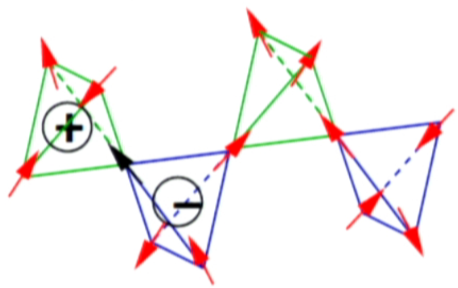
Castelnovo, Moessner and Sondhi (2008)

Excited states

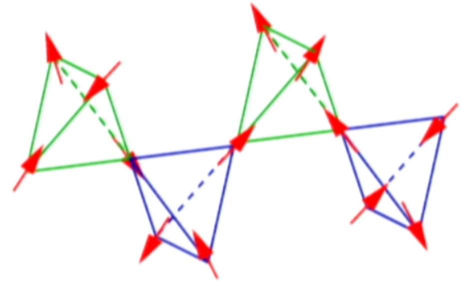


Interactions between monopoles

View spins as extended dipoles

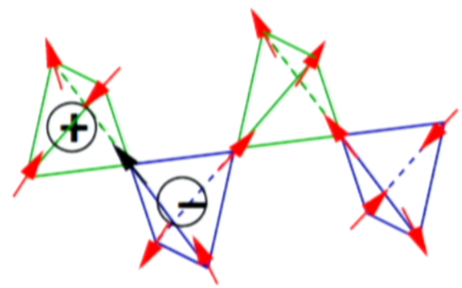


Two-in two-out

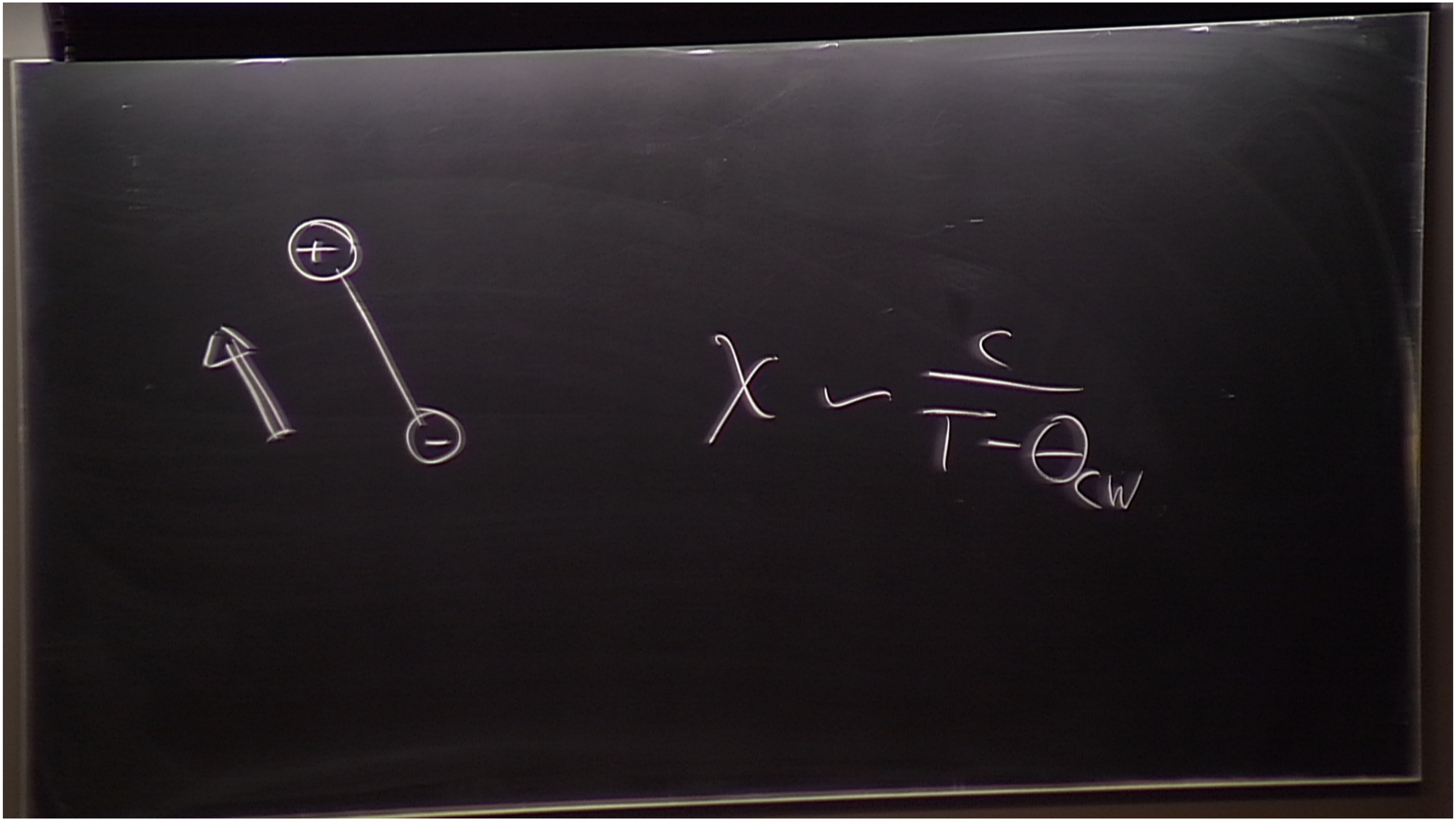


zero net charge

Three-in one-out

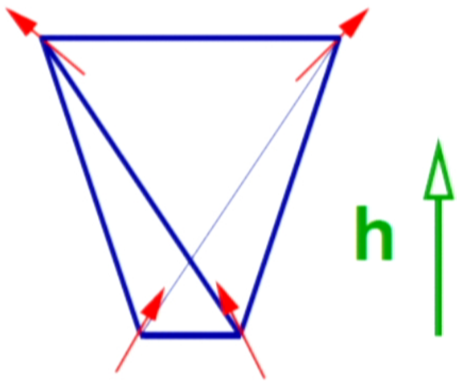


non-zero net charge


+
−
χ ~ C / (T − Θ_CW)

Engineering transitions in spin ice

Select ordered state with Zeeman field



Magnetisation

vs h^{eff}/T

for $h^{\mathrm{eff}}, T \ll J$

Uniform Zeeman field

– staggered coupling to Ising pseudospins

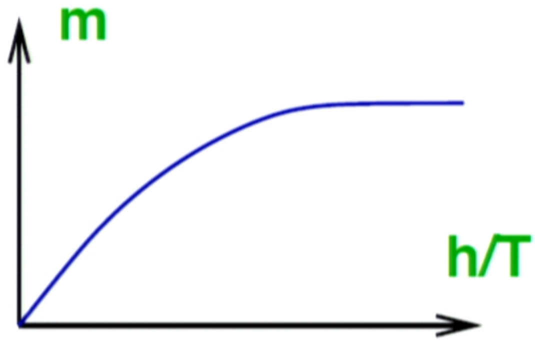
Jaubert, JTC, Holdsworth + Moessner, (2008)

A Kasteleyn transition

Magnetisation induced by applied field

Magnetisation vs temperature

In a paramagnet



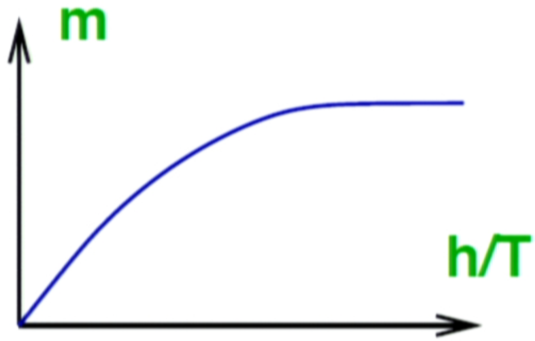
No transition

A Kasteleyn transition

Magnetisation induced by applied field

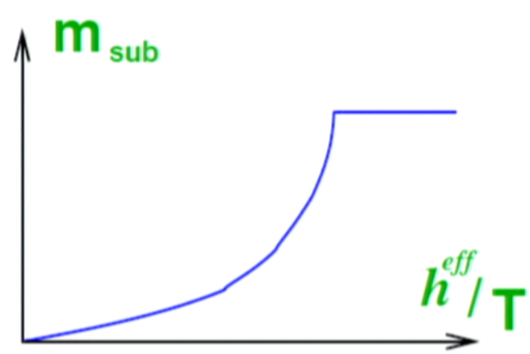
Magnetisation vs temperature

In a paramagnet



No transition

From the Coulomb phase

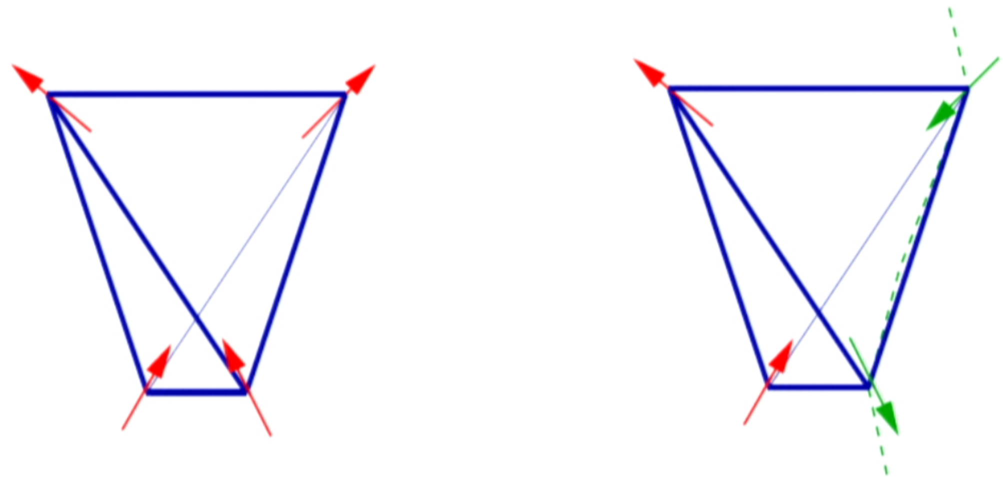


One-sided transition

- Continuous from low-field side
- First-order from high-field side

Description of the transition

Reference state: fully polarised — Excitations: spin reversals



‘Vacuum’ — String excitation

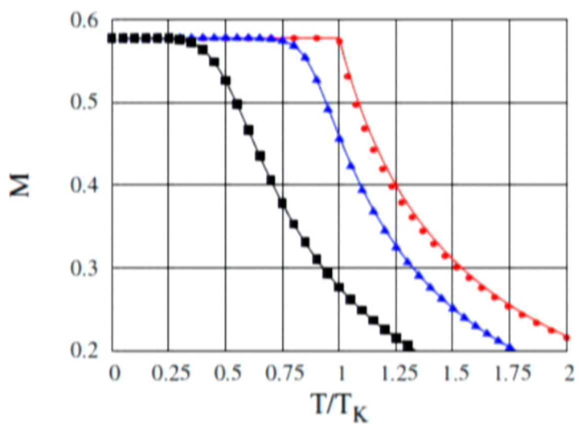
Thermodynamics of isolated string, length L:

Energy $L \cdot h$ Entropy $L \cdot k_{\mathrm{B}} \ln(2)$ Free energy $L \cdot [h - k_{\mathrm{B}} T \ln(2)]$

String density: finite for $h/k_{\mathrm{B}}T < \ln(2)$ zero for $h/k_{\mathrm{B}}T > \ln(2)$

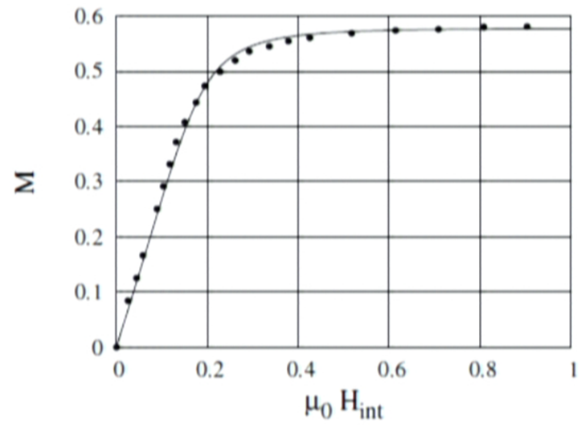
Kasteleyn: Simulation and Experiment

Magnetisation vs T



$h/J = 0.58,\ 0.13,\ 10^{-3}$

Magnetisation vs H



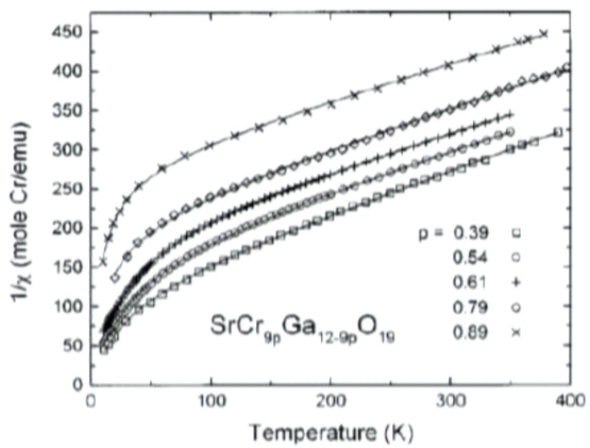
Data for $Dy_2Ti_2O_7$ at 1.8K

(Fukazawa *et al.* 2002)

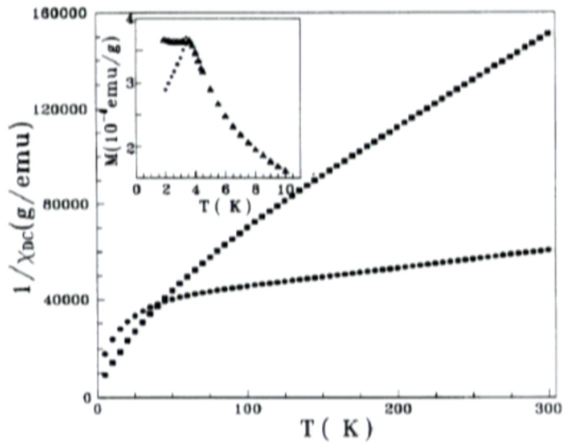
$k_B T \simeq 1.6 J_{\rm eff}$

Quenched disorder

Free moments and spin freezing



Curie tail in χ



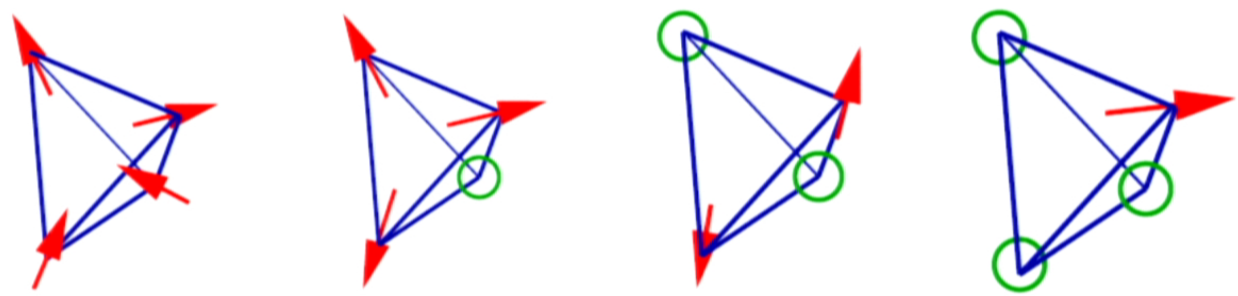
FC vs ZFC χ

Free moments as orphan spins

Separate susceptibility into two parts: $\chi(T) = \frac{c_1}{T - \theta_{\rm CW}} + \frac{c_2}{T}$

Schiffer and Daruka (1997)

Only orphan spins are uncompensated



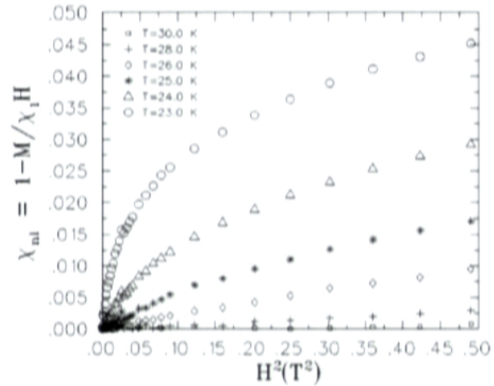
Developed in: Sen, Damle and Moessner (2009)

Spin freezing in geometrically frustrated magnets

Examples

	$\lvert\Theta_{\rm CW}\rvert$	$T_{\rm F}$
SCGO	**500K**	**4K**
Hydronium iron jarosite	**700K**	**14K**
$Y_2Mo_2O_7$	**200K**	**22K**
$CsNiCrF_6$	**70K**	**2.3K**

Evidence for SG transition



$Y_2Mo_2O_7$: $\chi_{\rm nl}$ **vs** H^2

Gingras ***et al.*** **(1997)**

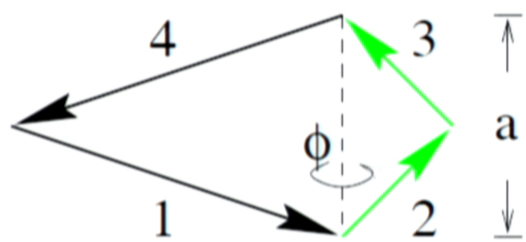
Freezing at low T, sometimes despite low levels of structural disorder

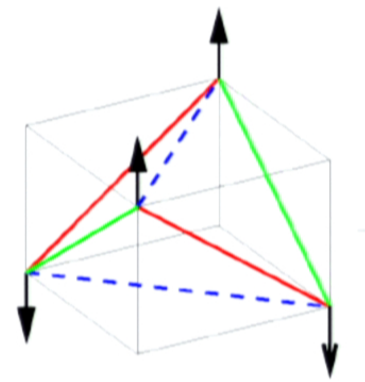
Exchange randomness: one tetrahedron

Bellier-Castella *et al.* (2001)

$$J \longrightarrow J + \delta J_{ij}$$

Collinear spin states selected





Ground states in clean system

Ferromagnetic pairs where exchange is weakest

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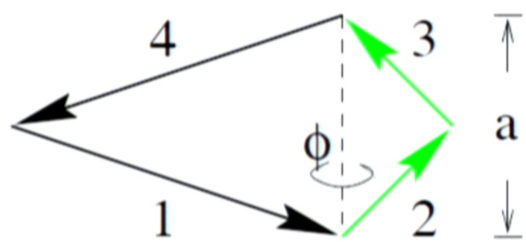

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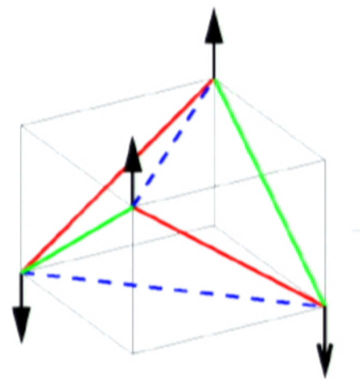
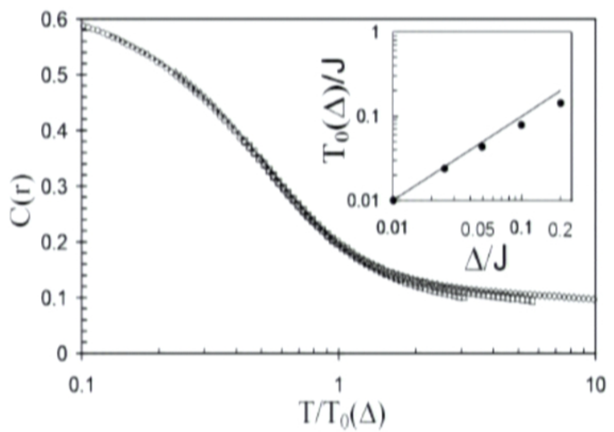


Ferromagnetic pairs where exchange is weakest

Spin glass transition in simulations

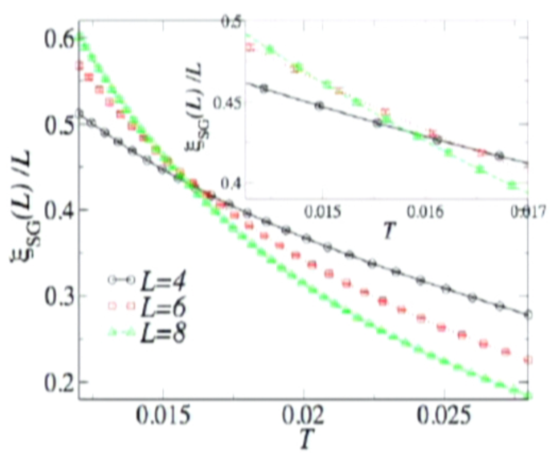
$$J \to J + \delta J_{ij} \text{ with } \langle \delta J_{ij}^2 \rangle \equiv \Delta^2 \text{ and } \Delta \ll J$$

Scaling with disorder strength



Saunders + JTC (2007)

Transition is cooperative



Tam, Hitchcock & Gingras (2010)

Summary

Geometric frustration

leads to macroscopic classical degeneracies

— long-range order avoided

Coulomb phases

ground state constraints generate long range correlations

— emergent gauge field & fractionalized excitations

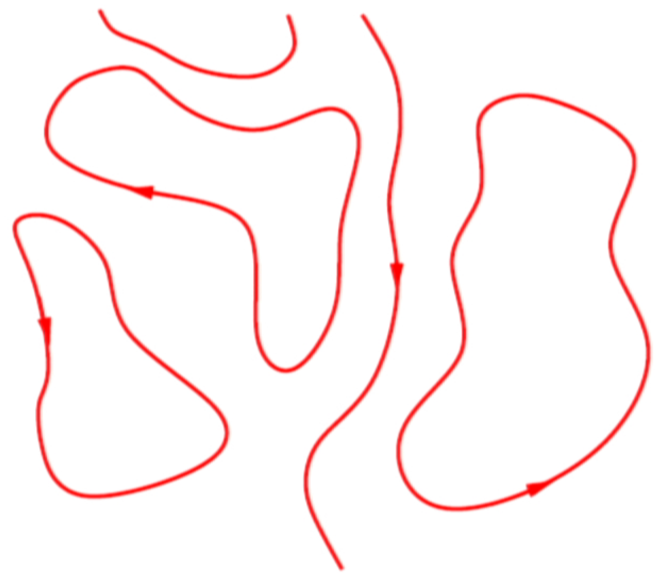
Order and disorder

exotic phase transitions

robust to some forms of disorder, highly sensitive to others

Further reading: "Highly Frustrated Magnetism" Eds. C. Lacroix, P. Mendels, F. Mila

Ground states as flux loops



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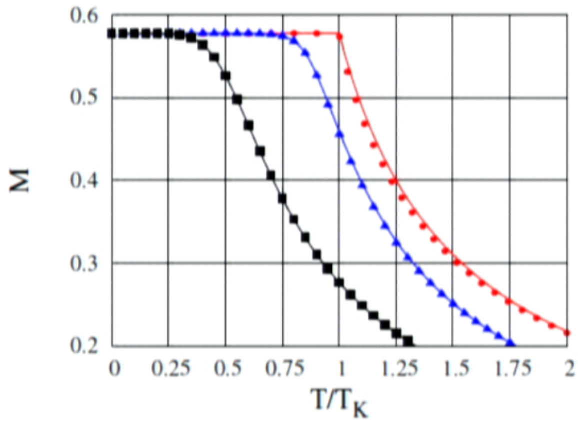

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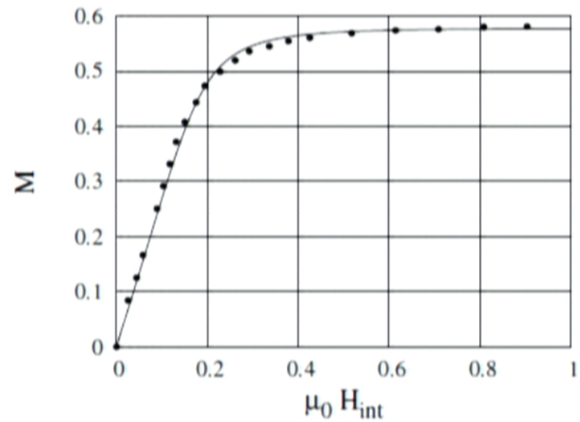

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