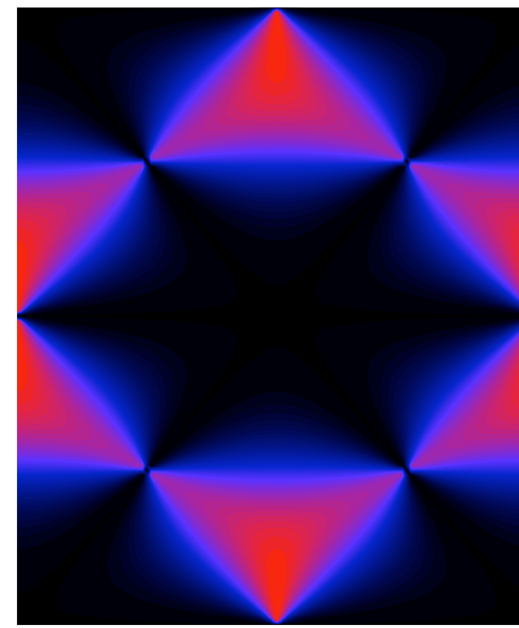
Tutorial on frustrated magnetism

Roderich Moessner



Overview

- Frustrated magnets
  - What are they? Why study them?
- Classical frustration – degeneracy and instability
- Order by disorder
- Quantum frustration
  - weak quantum fluctuation
  - strong quantum fluctuations, and the $S = 1/2$ kagome magnet
- The spinels: experimental model systems
  - magnetoelastics and heavy Fermions
- Outlook

Why study frustrated magnets

- Materials physics
  - because they exist (and may be useful)
- Conceptually important model systems – often tractable
  - strong correlations/fluctuations
  - coupled degrees of freedom
  - interesting (quantum) phases, including liquids
  - unconventional phase transitions

History

[Contribution from the Gates Chemical Laboratory, California Institute of Technology, No. 506]

The Structure and Entropy of Ice and of Other Crystals with Some Randomness of Atomic Arrangement

By Linus Pauling

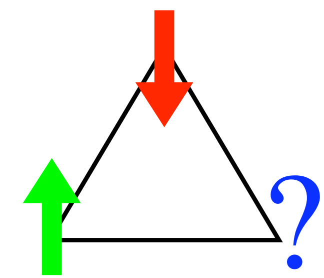
Investigations of the entropy of substances at low temperatures have produced very important information regarding the structure of crystals, covery of the hydrogen bond[4] that the unusual properties of water and ice (high melting and boiling points, low density, association, high di-

- First system: ice Pauling, JACS 1935
- 1950s: triangular Ising magnet Wannier+Houtappel; pyrochlore Ising magnet ('spin ice') Anderson
- 'cooperative paramagnets' Villain 1977
- Most complete bibliography (by Oleg Tchernyshyov) `http://www.pha.jhu.edu/~olegt/pyrochlore.html`
- Reviews: Misguich+Lhuillier cond-mat; H.T. Diep book; R.M.+Ramirez Phys. Today

Frustration leads to (classical) degeneracy

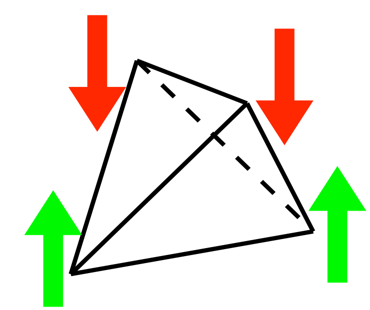
Consider Ising spins $\sigma_i = \pm 1$ with antiferromagnetic $J > 0$:

$$\mathcal{H} = J \sum_{\langle ij \rangle} \sigma_i \sigma_j$$



- Not all terms in $\mathcal{H}$ can simultaneously be minimised
- But we can rewrite $\mathcal{H}$:

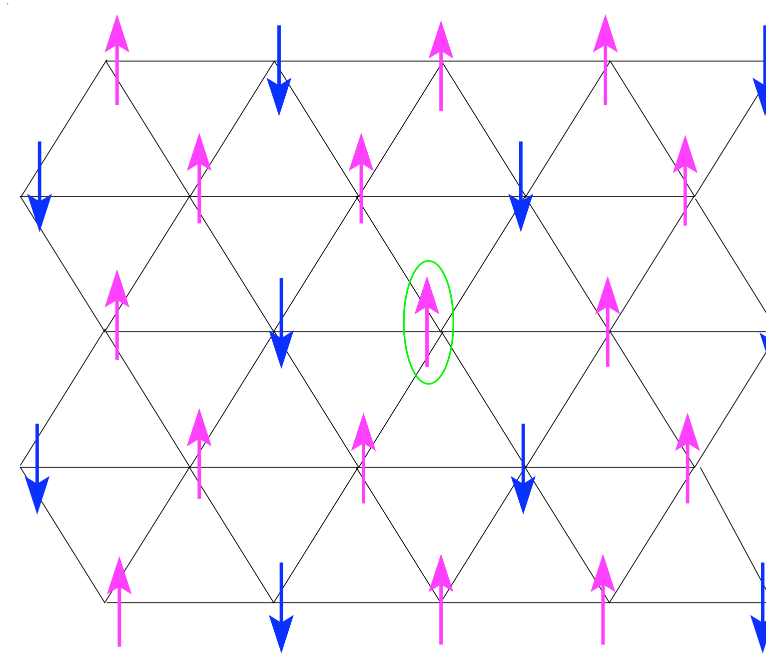
$$\mathcal{H} = \frac{J}{2} \left(\sum_{i=1}^{q} \sigma_i \right)^2 + const$$



- Number of ground states: $N_{gs} = \binom{4}{2} = 6$ for one tetrahedron
- Degeneracy is hallmark of frustration

Frustration $\Rightarrow$ degeneracy $\Rightarrow$ zero-point entropy

- ground-state condition: $\uparrow\uparrow\downarrow$ or $\uparrow\downarrow\downarrow$ for each triangle
- finite entropy in ground state: $S = 0.323 k_B$
- ‘flippable spins’ experience vanishing exchange field



<u>What happens at low T?</u>

Frustration $\Rightarrow$ degeneracy $\Rightarrow$ zero-point entropy

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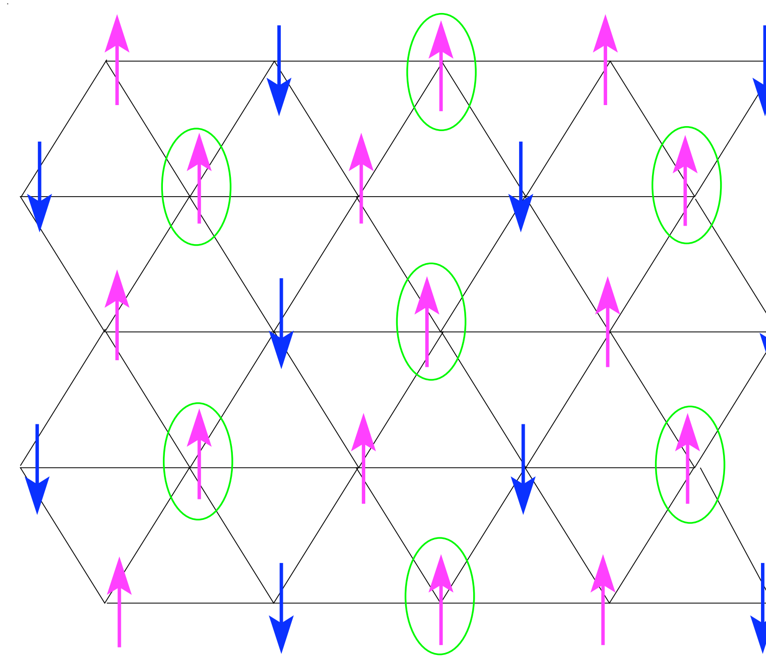


$\Rightarrow$ lower bound on entropy

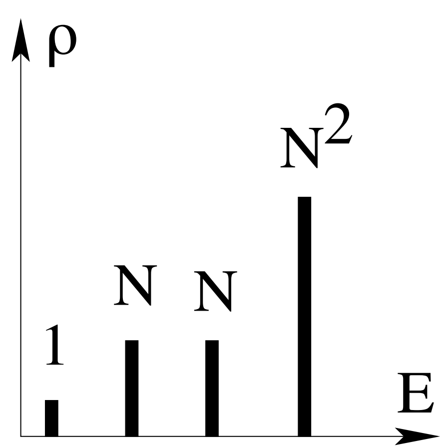
$$\mathcal{S} \geq (k_B/3) \ln 2$$

- Important: <u>local</u> d.o.f.

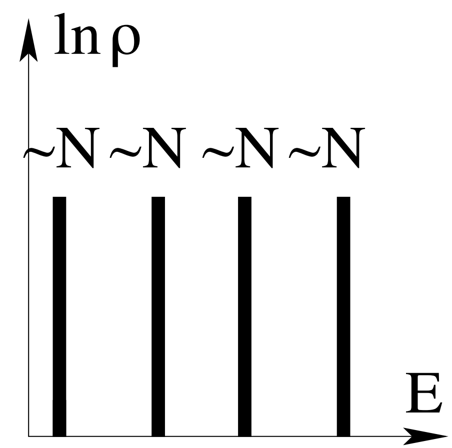
<u>What happens at low T?</u>

Why degenerate systems are special

d.o.s – unfrustrated magnet



d.o.s – frustrated magnet

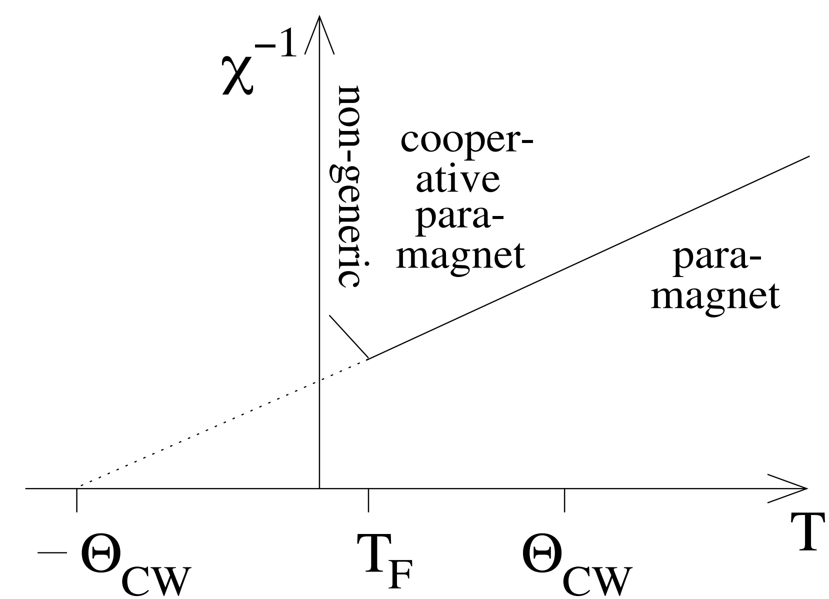


- Ground states can exhibit subtle correlations (seen at low T)
- Degenerate ground states provide no energy scale
  $\Rightarrow$ all perturbations are strong $\Rightarrow$ many instabilities
- Very rich behaviour (theory+experiment) – but also hard
- Cf. quantum Hall physics (degenerate Landau levels)

The cooperative paramagnetic regime Villain

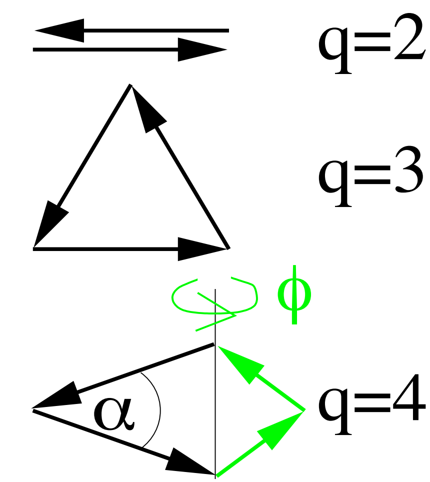
- Definition: regime at low temperature $T \ll J$ which is continuously connected to high-temperature paragmagnetic phase
- Properties: correlations short-ranged in space and time (?)
- Experiments: phase transitions occur much below the Curie-Weiss temperature: $T_F \ll \Theta_{CW}$ Ramirez

'Susceptibility fingerprint' of frustration



Constraint counting as a measure of frustration

Units of q
Heisenberg spins



$$H = J \sum_{ij} S_i S_j \simeq (J/2)(\sum_{i=1}^{q} S_i)^2$$

gives ground state degeneracy:

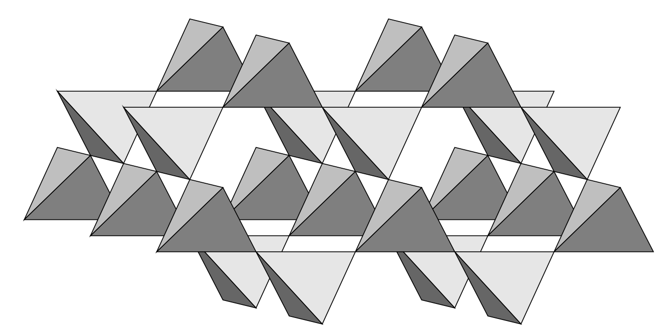
$L \equiv \sum_i S_i$ to be minimised.

degeneracy grows with q

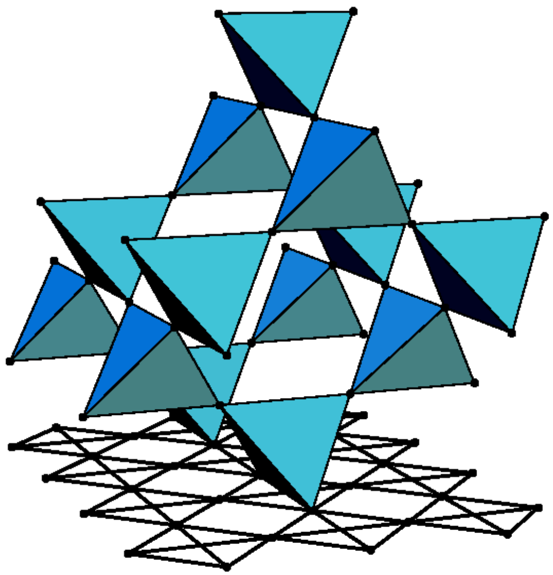
Constraint counting: $D = F - K$

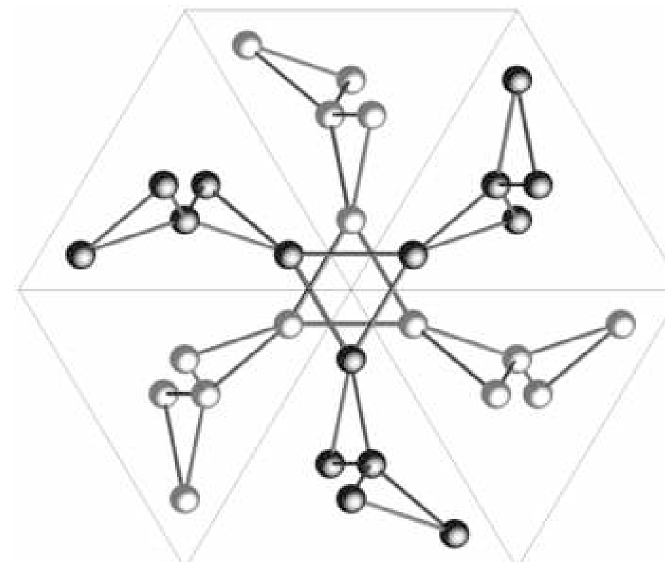
- ground-state degeneracy D
- total d.o.f. F
- ground-state constraint K

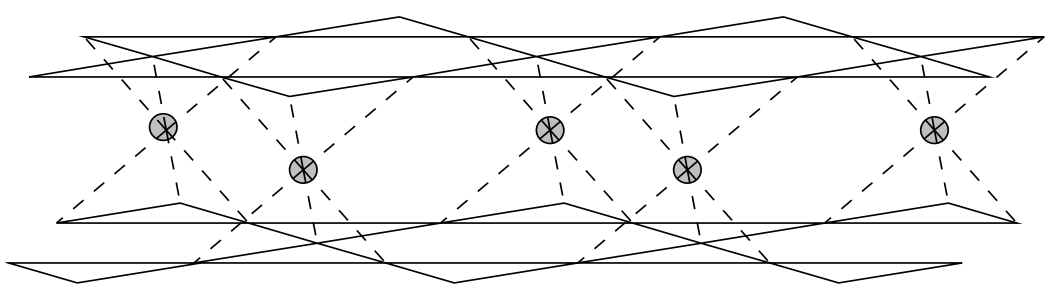
Pyrochlore antiferromagnets are particularly frustrated

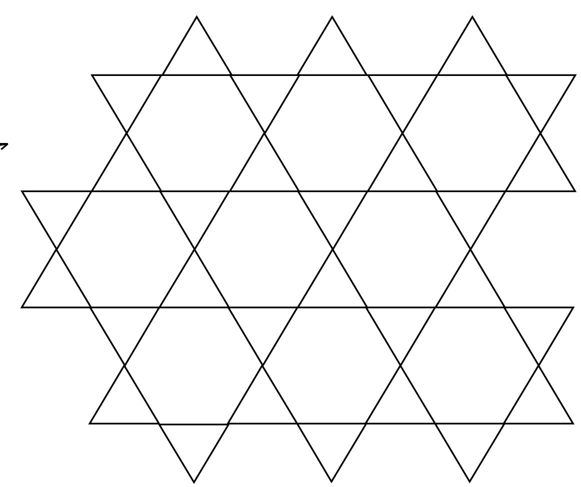


Highly frustrated (corner-sharing) lattices



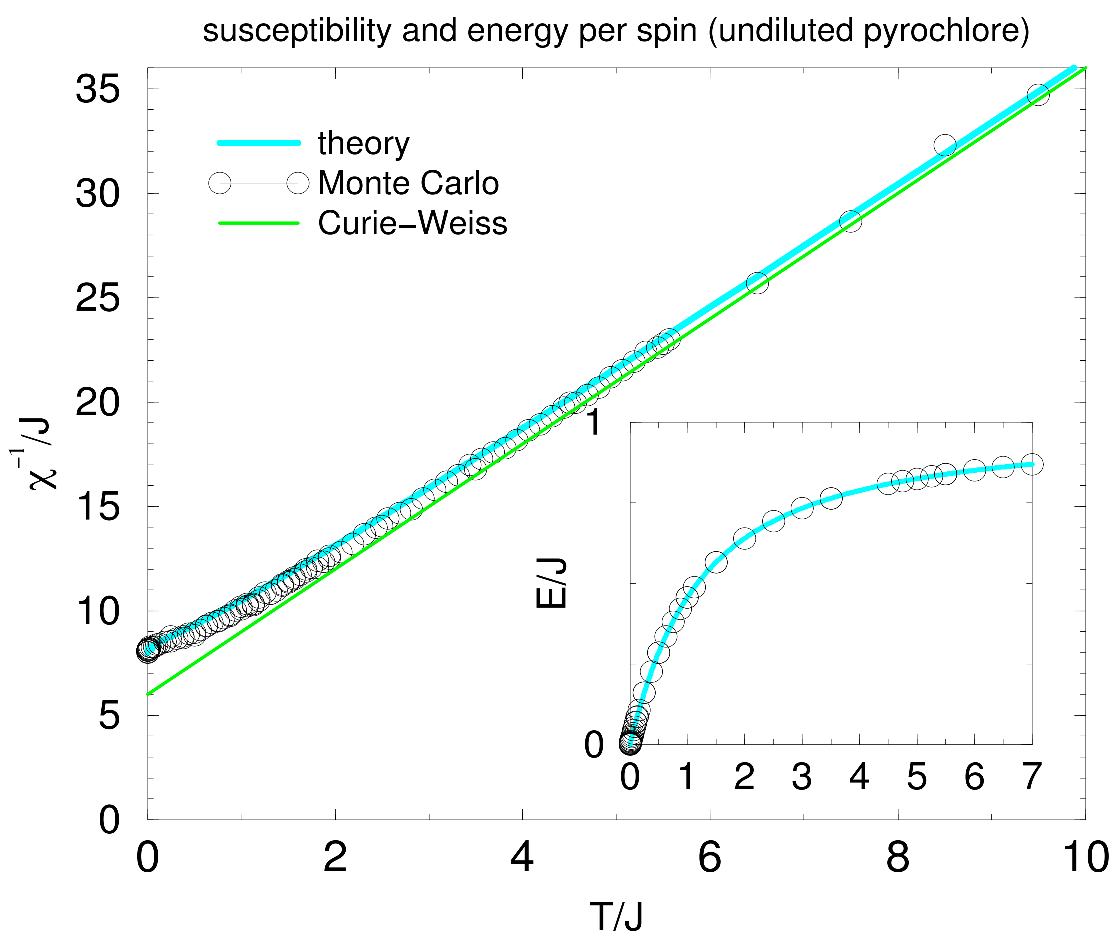






Thermodynamics: the single-unit approximation

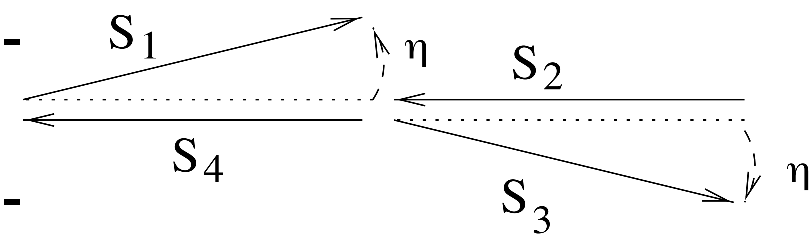
$\chi^{-1}(T)$ and $E(T)$ for Heisenberg pyrochlore

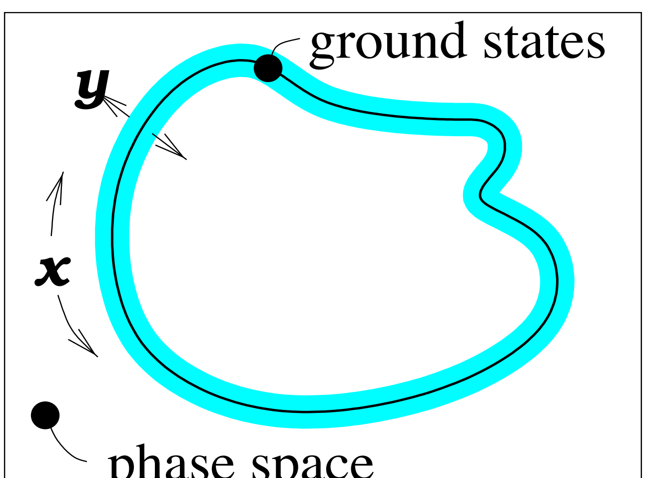


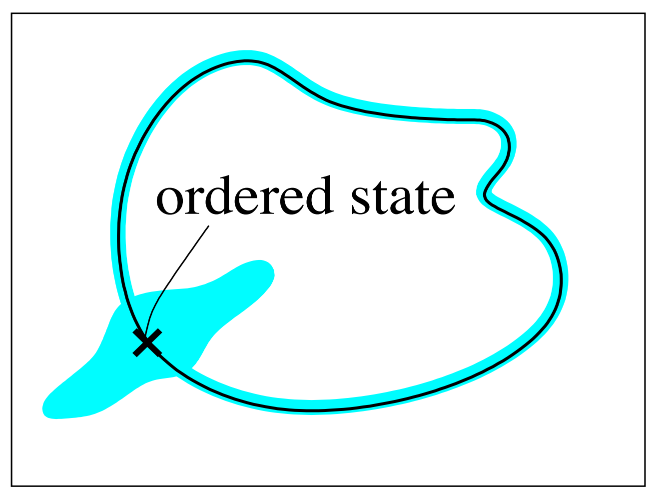
- ‘Natural’ d.o.f.: single tetrahedron spin $\mathbf{L} = \sum_i \mathbf{S}_i$, with $L \propto \sqrt{T}$ and $L \to 2$ at low (high) T.
- Solve ‘single unit’ (single tetrahedron) exactly
- Works rather well, despite neglect of *all* correlations beyond nearest neighbour.

Order by disorder *Villain, Shender*

- basic idea: fluctuations lift degeneracy
- thermal obdo: $F = U - TS$
- Ising spins: no low-energy fluctuations
- continuous spins: gapless excitations possible – some soft: $E \propto \eta^4$



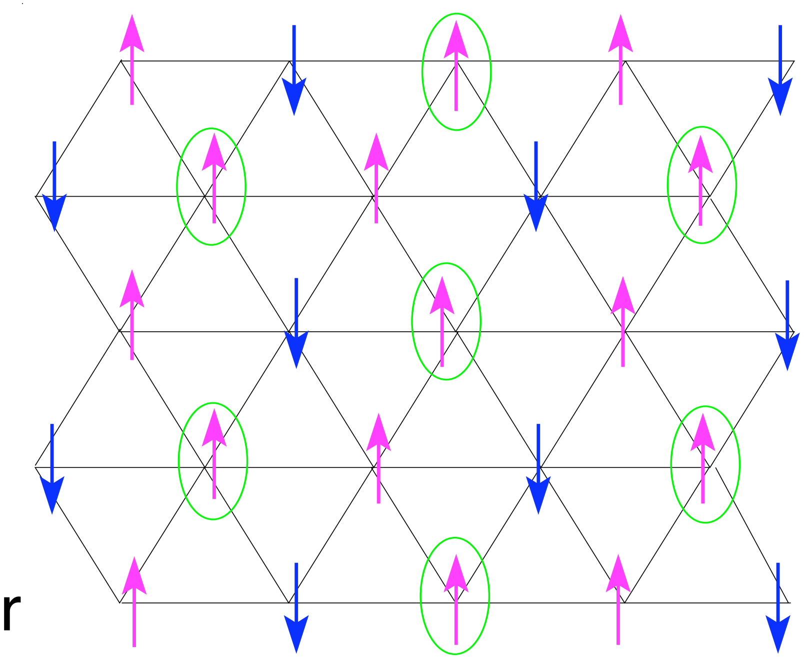




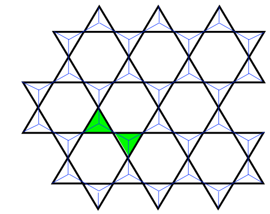
Where is weight concentrated?

Quantum frustration

- used to describe many (very different) situations
- simplest starting point think of transverse field Ising model
  - Hilbert space spanned by class. (discrete) ground states
  - quantum dynamics: as local as possible
- quantum obdo
  - 'maximally flippable' (triangle)
  - recent work on supersolids
  - 3d XY transition
- disorder by disorder (kagome)

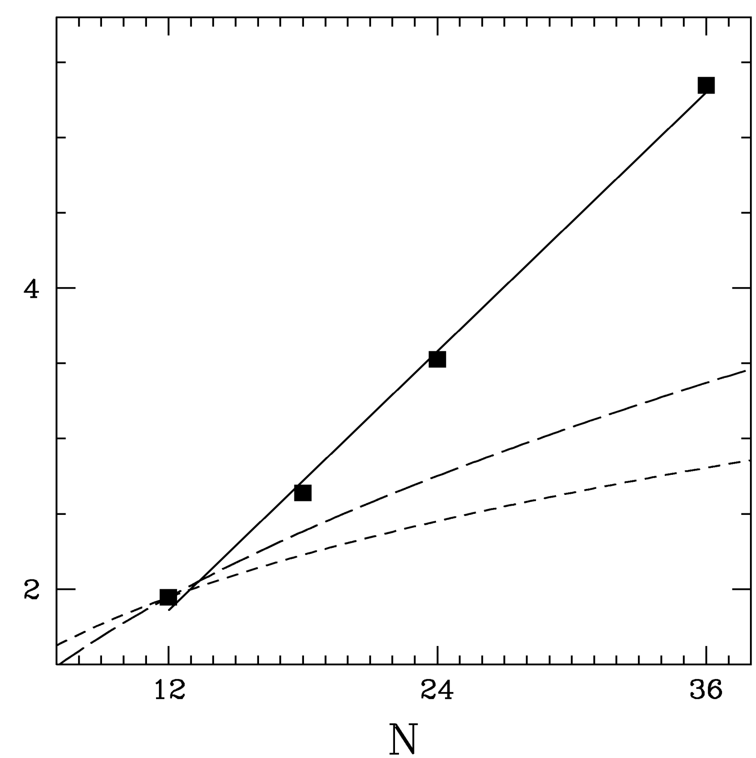


‘The holy grail’: $S = 1/2$ kagome



- kagome lattice has played important role historically
  - first experimental on SCGO (with kagome motif) Obradors
  - kagome $S = 1/2$ remains a mystery

- apparently no order at all
- spin gap Δ
- small singlet gap (if any)
- **many** singlet states with $E < \Delta$
- **even more** theories



The ‘simple’ spinel oxides AB_2O_4 (after Takagi)

$d^{0.5}$ $LiTi_2O_4$ BCS SC	$d^{1.5}$ LiV_2O_4 heavy Fermion	$d^{2.5}$ AlV_2O_4 charge-ordered	$d^{3.5}$ $LiMn_2O_4$
d^1 $MgTi_2O_4$ valence bond solid	d^2 $\{Zn,Mg,Cd\}V_2O_4$ spin+orbital ordering	d^3 $\{Zn,Mg,Cd\}Cr_2O_4$ spin+structural phase transition	d^4 $ZnMn_2O_4$

- ions on B-sublattice form pyrochlore lattice
- properties tunable by varying ions on A, B sublattices
- many more compounds exist
- LiV_2O_4: non-integer nominal valence; orbital d.o.f.; spin
  - many sources of entropy at low T
  - whence heavy Fermion behaviour?

Supplementary (lattice) d.o.f. in the Cr spinels

- nominal valence of Cr: d^3 (half-filled t_{2g} orbitals)
  $\Rightarrow$ isotropic $S = 3/2$ on pyrochlore lattice
- Q: Interplay of elastic degrees of freedom and frustration?
- magnetoelatic Hamiltonian $\mathcal{H}_{tot} = \mathcal{H}_m + \mathcal{H}_{me} + \mathcal{H}_e$
  - magnetic exchange $\mathcal{H}_m = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$
  - magnetoelastic coupling (x_a ... displacements)

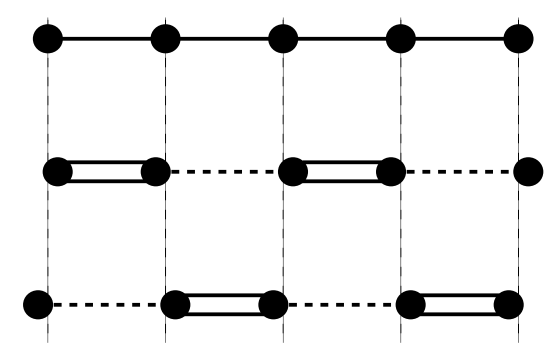
$$\mathcal{H}_{me} = \sum_{aij} \frac{dJ_{ij}}{dx_a} \left(\mathbf{S}_i \cdot \mathbf{S}_j \right) x_a$$

  - elastic energy $\mathcal{H}_e = \sum_{ab} k_{ab} x_a x_b$ (k_{ab} ... elastic constants)

Unfrustrated magnetoelastics: chain in $d = 1$

- $\mathbf{S}_i \cdot \mathbf{S}_j = c_{nn}$ is uniform for nearest neighbours
- Simplest case: $dJ_{ij}/dx_a = J'\delta_{a,i}$:

  $\mathcal{H}_{me} + \mathcal{H}_e = \sum_a J' c_{nn} x_a + k x_a^2$ minimised by $x_a = -J' c_{nn}/(2k)$
  $\Longrightarrow E_{min} = -(J' c_{nn})^2/(4k)$ grows with $|c_{nn}|$
- $\mathcal{H}_m$ minimised by extremal $c_{nn} = \mathbf{S}_i \cdot \mathbf{S}_j = -S^2$
- global minimum of $\mathcal{H}_{tot}$: only uniform contraction!
- quantum $S = 1/2$ chain:
  - $\mathbf{S}_i \cdot \mathbf{S}_j$ cannot independently extremised
  - modulated $\mathbf{S}_i \cdot \mathbf{S}_j \Rightarrow$ modulated distortion $\Rightarrow$ dimerisation



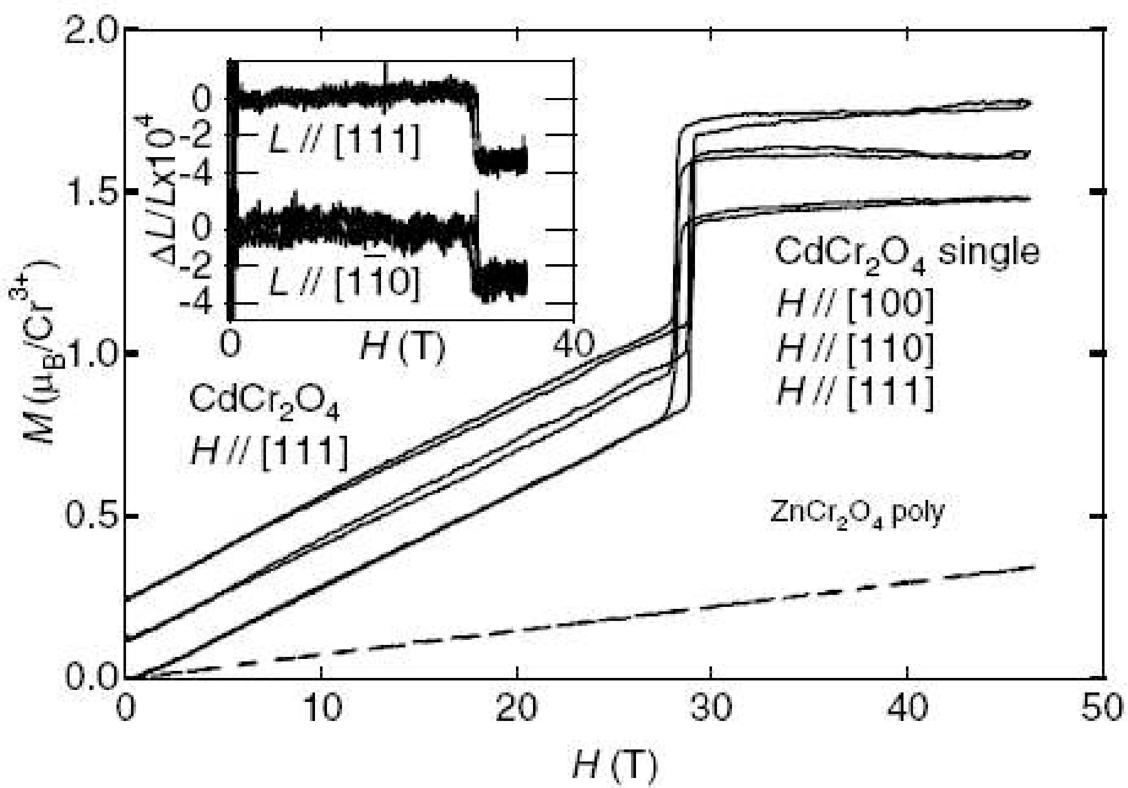
Frustrated magnetoelastics in a nutshell

- Frustration $\longrightarrow$ degeneracy of ground states
- Degenerate states *not* symmetry equivalent

  $\Rightarrow$ $\mathbf{S}_i \cdot \mathbf{S}_j$ can be non-uniform
- Distortions (strengthen)weaken (un)frustrated bonds
- Energy balance: distortions generally present at low T
  - magnetic energy: linear gain $(\mathbf{S}_i \cdot \mathbf{S}_j) \times x$
  - elastic energy: quadratic cost kx^2
- Basically: $x \sim \mathbf{S}_i \cdot \mathbf{S}_j \Rightarrow$ eff. biquadratic exchange $(\mathbf{S}_i \cdot \mathbf{S}_j)^2$

$\Rightarrow$ favours collinear states (not always seen!)

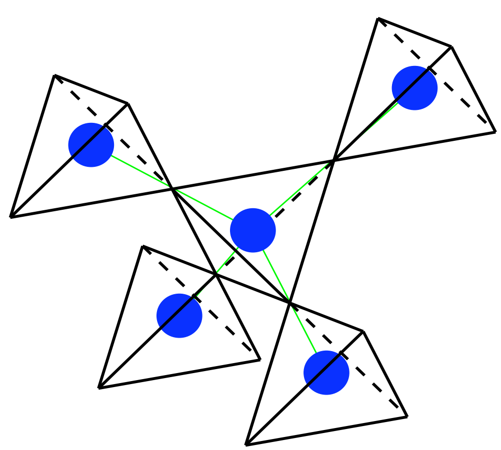
Collinear order by distortion in $CdCr_2O_4$ *Ueda et al.*

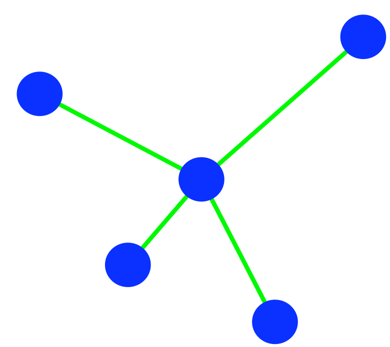


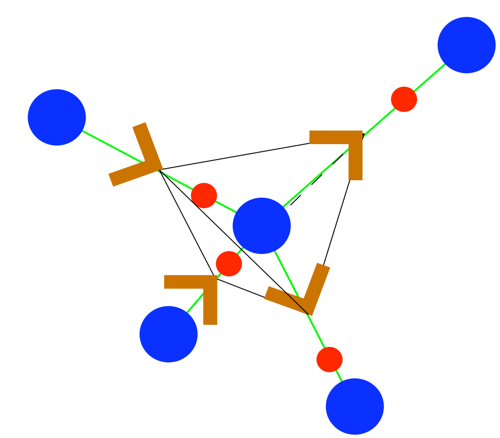
- at plateau centre, collinear ↑↑↑↓ among ground states
- eff. biquadratic exchange leads to plateau formation
- details to be worked out

Emergent gauge structure: from spins to fluxes

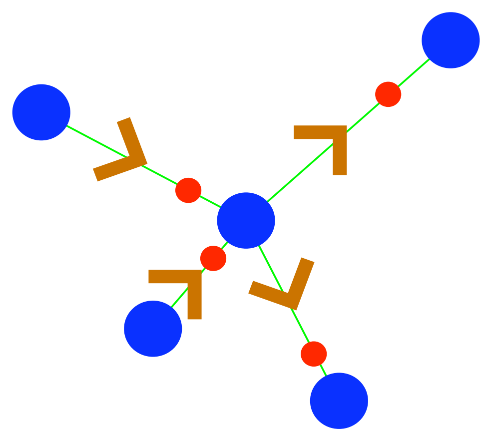
- Think of spins as living on links of dual lattice
- Easiest for Ising spins = 1 unit of flux
- Experimental realisation: spin ice compounds







Local constraint $\longrightarrow$ conservation law

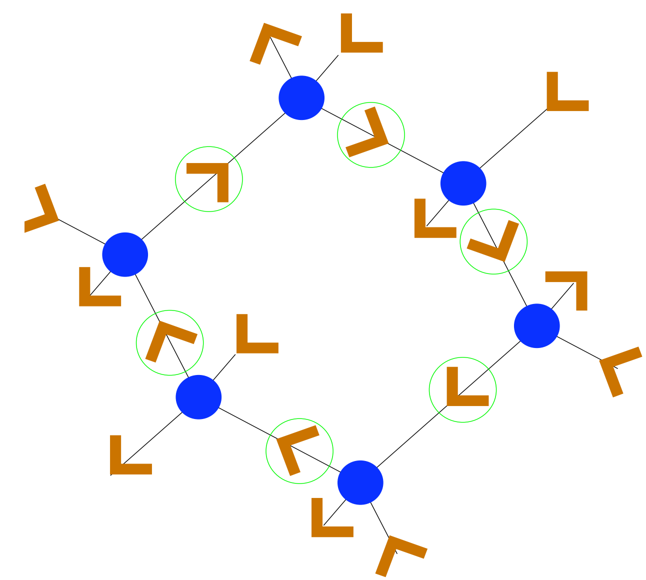


- Define 'flux' vector field on *links* of the ice lattice: $\mathbf{B}_i$
- Local constraint (ice rules) becomes conservation law (as in Kirchoff's laws)

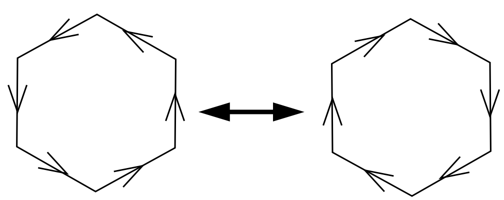
  $\Rightarrow$ gauge theory

$$\nabla \cdot \mathbf{B} = 0 \Longrightarrow \mathbf{B} = \nabla \times \mathbf{A}$$

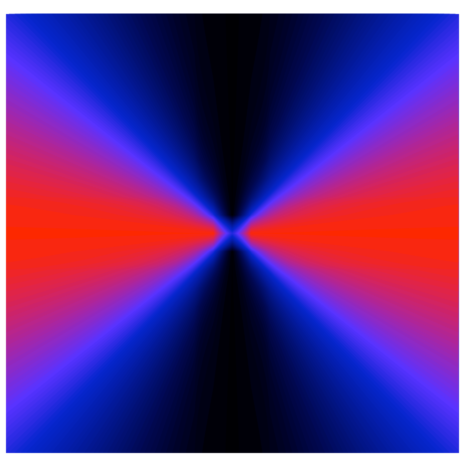
- Ice configurations differ by rearranging protons on a loop
- Amounts to reversing closed loop of flux $\mathbf{B}$
- Smallest loop: hexagon (six links)



Long-wavelength analysis: coarse-graining



- Coarse-grain $\mathbf{B} \rightarrow \tilde{\mathbf{B}}$ with $\nabla \cdot \tilde{\mathbf{B}} = 0$
- 'Flippable' loops have zero average flux:
  low average flux $\Leftrightarrow$ many microstates
- Ansatz: upon coarse-graining, obtain energy functional of entropic origin:

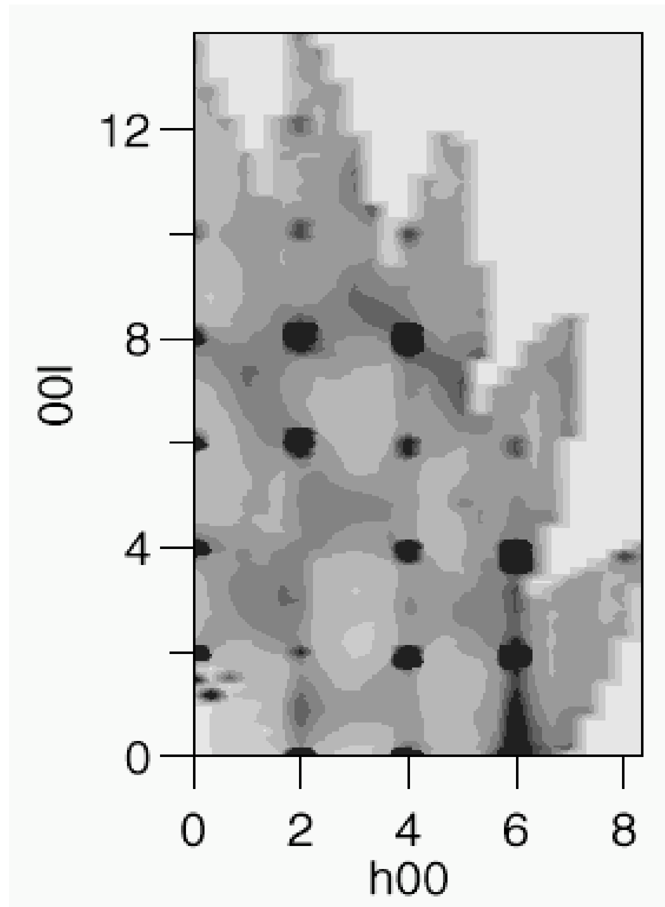


$$Z = \sum_{\mathbf{B}} \delta_{\nabla.\mathbf{B},0} \rightarrow \int \mathcal{D}\tilde{\mathbf{B}}\, \delta(\nabla \cdot \tilde{\mathbf{B}})\ \exp[-\frac{K}{2}\tilde{\mathbf{B}}^2]$$

- Artificial magnetostatics!
- Resulting correlators are transverse and algebraic (but not critical!): e.g.

$$\langle \tilde{B}_z(q)\tilde{B}_z(-q)\rangle \propto q_\perp^2/q^2 \leftrightarrow (3\cos^2\theta - 1)/r^3.$$

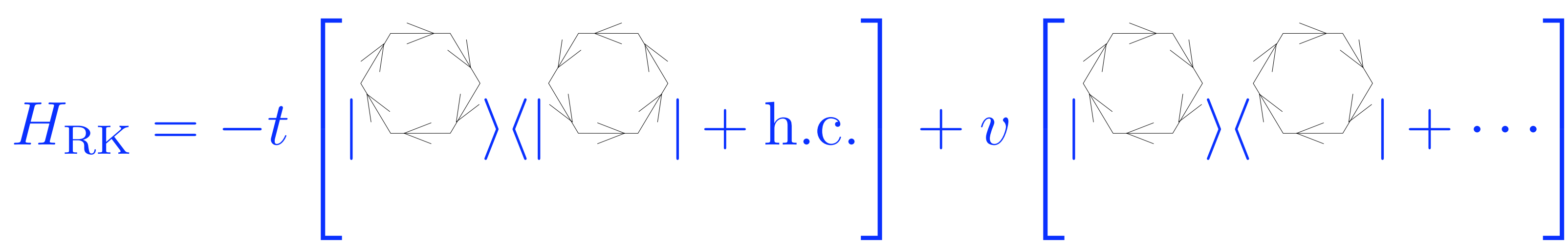
Bow-ties in the structure factor of ice



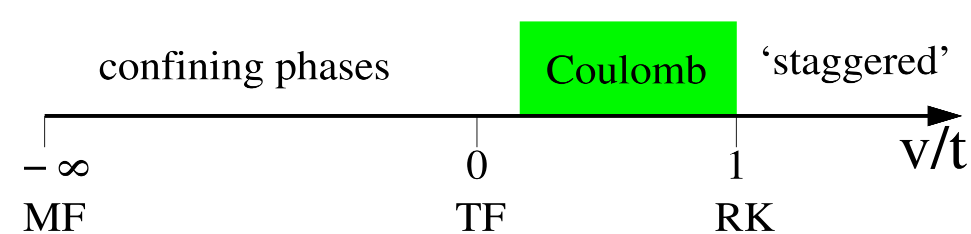
proton distribution in water ice, I_c Li *et al.*

‘Quantum ice’: artificial electrodynamics

- Hilbert space: (classical) ice configurations
- Add coherent quantum dynamics for hexagonal loop:



- Effective long-wavelength theory $\mathcal{H}_q = \int \tilde{\mathbf{E}}^2 + c^2 \tilde{\mathbf{B}}^2$ Maxwell
- This describes the Coulomb phase of a $U(1)$ gauge theory:
  - gapless photons, speed of light $c^2 \propto v - t$
  - deconfinement
  - microscopic model!



- Artificial electrodynamics with ice as ‘ether’ Wen’s noodle soup

Summary

- frustration $\Rightarrow$ degeneracy $\Rightarrow$ strong fluctuations
  - new phases/phase transitions/dynamics $\cdots$
- simple model systems
- many realisations
  - materials physics
  - nanotechnology
  - cold atoms