Ordering (partway) by Disorder in Large–S Kagome and Pyrochlore Antiferromagnets

CHRISTOPHER L. HENLEY, CORNELL UNIV.

Kagome: ERNEST P.CHAN (Ph.D. thesis,1994);
CLH and E.P. Chan, J. Mag. Mag. Mater. 140–144,1693 (19
Pyrochlore harmonic: O. Tchernyshyov et al,
PRB 68, 144422 (2003); PRB 69, 212402 (2004).
Pyrochlore anharmonic: UZI HIZI (currrent).

Support: NSF DMR–0240953

Summary: approach to find true ground state

* spinwave zero–point energy breaks degeneracies
* start from discrete family of selected states
* discrete spins are coefficients in spinwave expansion
* obtain effective Hamiltonian in terms of discrete spins
* uncontrolled perturbation expansion
* defined on ANY discrete state (not just simple periodic ones)

Outline: review/preview
I. Kagome
II. Pyrochlore

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Frustration

Highly frustrated

classical states – continuous –
deg. of freedom ~ N

OR discrete states no. ~ exp(N)

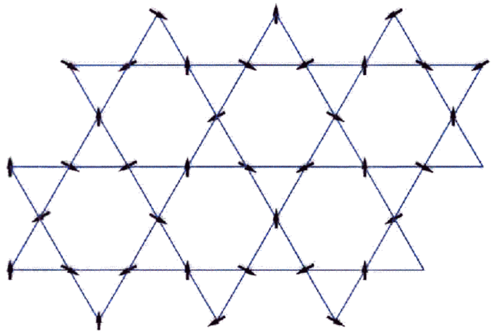
Frustration ≠ [satisfy some terms, but not all at once)

→ modern concept : large (near) degeneracy

∴ expect rich phase diagrams —
can make states w/ all sorts of order

(Compare METALS wh. also have large number of low E states)

Kagomé antiferromagnet



$$\mathcal{H} = \frac{1}{2} \sum_{\langle ij \rangle} |J| \vec{s}_i \cdot \vec{s}_j$$

nearest neighbor

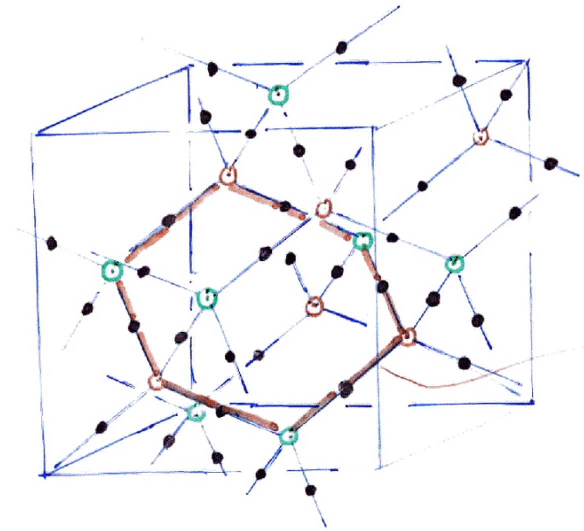
Heisenberg spins
(S_{ix}, S_{iy}, S_{iz})

$S \gg 1$ (semiclassical limit)

[Definitely not valid for $S=\frac{1}{2}$, $S=1$!]
C. Zeng & V. Elser; Lecheminant et al ...]

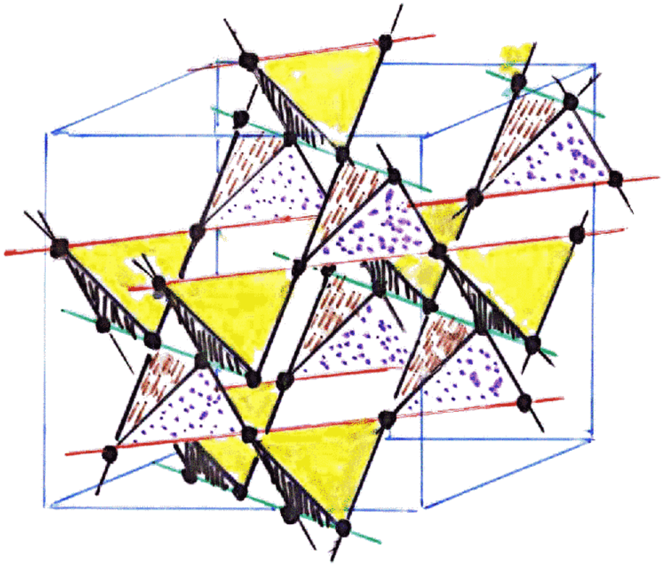
Expect ground state has Néel order —
but in which ordering pattern?
(How nearly degenerate the alternatives?)

PYR. AS MEDIAL LATTICE OF THE DIAMOND LATTICE.



smallest loop is 6

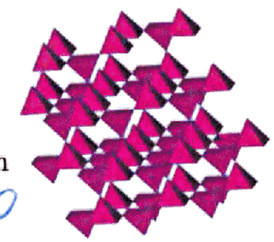
PYROCHLORE



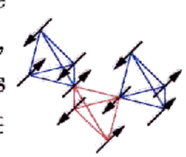
U. Hizi & C. L. H.
March 2004

Heisenberg Model on the Pyrochlore lattice

$$\mathcal{H} = J\sum_{\langle ij\rangle} \vec{S}_i \cdot \vec{S}_j = \sum_\alpha \Big|\underbrace{\sum_{i\in\alpha} \vec{S}_i}_{L_\alpha}\Big|^2 + \text{const.}$$



- Classically, all states with zero sum in each tetrahedron are degenerate. $\vec{L}_\alpha \equiv 0$
- Thermal fluctuations do not break the degeneracy enough to make LRO (*Reimers 1992, Moessner and Chalker, 1998*).
- In large S limit, quantum fluctuations choose a subset of the collinear ground states (*Henley, APS March Meeting 2001*). Ground state is characterized by Ising variables $\vec{S}_i = \eta_i \hat{z}$, $\eta_i \in \{\pm 1\}$, $\sum_{i\in\alpha} \eta_i = 0$



- Does the large S quantum model possess long range order?

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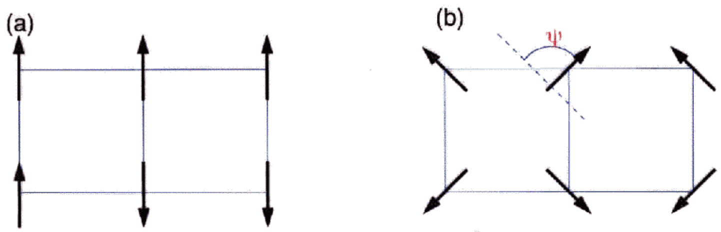
Review "order by disorder"

NON highly frustrated (just one global degeneracy parameter ψ)

resolve degeneracy by (harmonic) zero–point E.

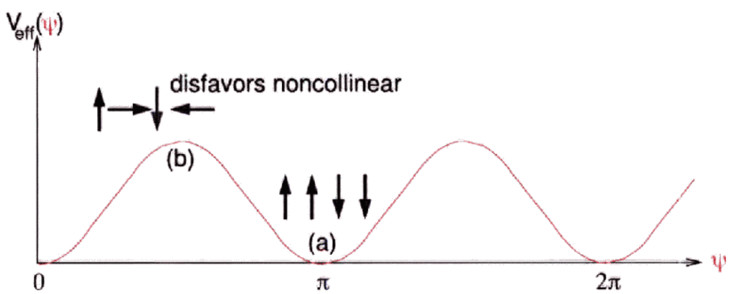


same classical energy ($O\, J(S^2)$)

different harmonic spinwave spectra

Define effective Hamiltonian

$$\boxed{V_{eff}(\psi) = \tfrac{1}{2}\sum \hbar\, \omega^{\psi}(k)}$$



favors collinear

Side remark: competing selection effects

	small parameter	favors
quantum [as above]	1/S	↑↑↓↓
thermal [classical spins]	T	↑↑↓↓
dilution [quenched]	δx	↑→↓←

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Spin wave expansion

... around a classical ground state $\{\vec{S}_i^{(0)}\}$

$$\boxed{\mathcal{H} \cong \mathcal{H}_{class} + \underbrace{\hat{\mathcal{H}}_2}_{O(a^\dagger a)} + \underbrace{\underbrace{\hat{\mathcal{H}}_3}_{O(a^\dagger a a)} + \underbrace{\hat{\mathcal{H}}_4}_{O(a^\dagger a a a^\dagger + \cdots)}}_{\text{ANHARMONIC}}}$$

$a_i\, a_i^\dagger \equiv$ Holstein - Primakoff boson operators

$\sigma_{ix} \pm i\sigma_{iy} \leftrightarrow a_i^\dagger, a_i^-$

Philosophy of this approach

Usual approach

construct 2 or so [high symm] candidates
evaluate E_{eff} as accurately as poss.

This approach ("effective Hamiltonians")

find $\mathcal{H}_{eff}(\cdots)$ defined for every state
[in a subspace defined by a previous level of selection]

Disadvantages

crude form of $\mathcal{H}_{eff}$, uncontrolled deriv.

Advantages

Maybe the right state is one you didn't think of?

Insert $\mathcal{H}_{eff}$ in a thermal ensemble

[or, augment by tunneling "flip" terms, to make a discrete quantum model w/ the possibility of a disordered state.]

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Preview of main results

Kagomé

discrete var. $\eta_\alpha = \pm 1$ on triangles
"chirality" of classical config. $\vec{S}_i \times \vec{S}_j \propto \eta_\alpha \hat{z}$

- harmonic $\mathcal{H}^{eff} = 0$
- anharmonic $\boxed{\mathcal{H}_{eff} = \sum_{\alpha\beta} J_{\alpha\beta}\, \eta_\alpha \eta_\beta}$ $J' > 0$

$\Rightarrow \sqrt{3}\times\sqrt{3}$ LRO [AS EVERYONE EXPECTED]

Pyrochlore

$\eta_i = \pm 1$ actual spins ($\vec{S}_i \propto \eta_i \hat{z}$)

- harmonic $\boxed{\mathcal{H}_{eff} = K \sum \prod_{i \in \hexagon} \eta_i}$ $K > 0$

Z_2 gauge-like form

$\Rightarrow \prod \eta_i = -1$ (each $\hexagon$) ← state selected

[UNEXPECTED - LARGE UNIT CELL]

Remaining $O(e^L) \sim O(e^{N^{1/3}})$ degeneracy

- anharmonic: in progress

I. KAGOMÉ CASE : classical gr. states

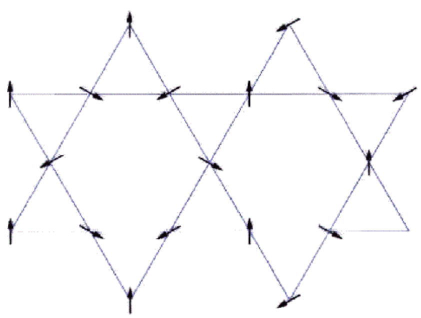
Ground state of one triangle: $\Sigma S_i = 0$

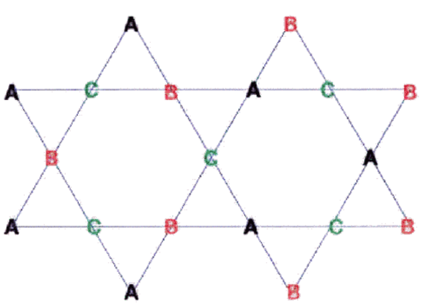
i.e. 120 state... we can do this on all trnagles

subclass of "coplanar" states

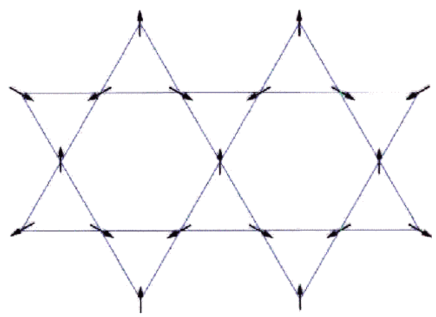
discrete 3-state Potts spins

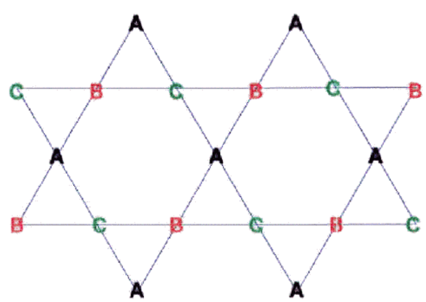
"$\sqrt{3} \times \sqrt{3}$"





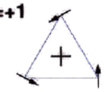
"Q=0" state





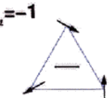
"Chiralities" [defined on each triangle]

$\eta_\alpha = +1$



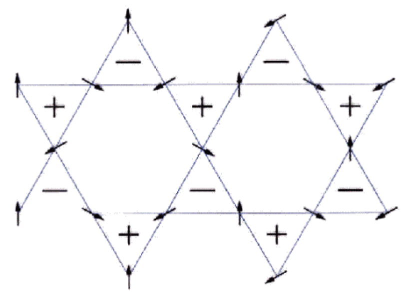
turn through +360
[Potts spins A,B,C]

$\eta_\alpha = -1$



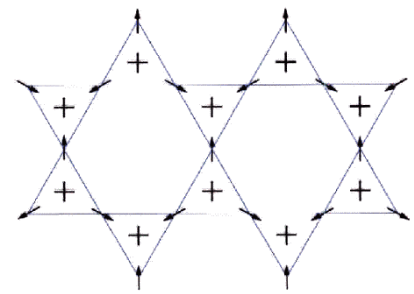
turn through −360
[Potts spins A,C,B]

"$\sqrt{3} \times \sqrt{3}$"



...is "antiferromagnetic"

"Q=0" state



...is "ferromagnetic"

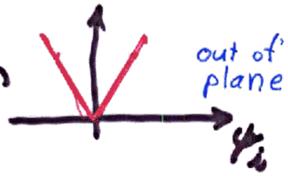
... in terms of chiralities.

Harmonic selection in Kagomé case

Coplanar states yes

[Ritchey, Chandra, Coleman c. 1992]

$E^{eff}_{(harm)}$



But: to harmonic order, $\omega(k)$ [hence $E^{eff}_{harm.} = \sum \frac{1}{2}\hbar\omega(k)$] is same for all coplanar ground states.

[see spinwave expansion]

$\therefore$ to get $\mathcal{H}^{eff}$ which selects between these (discrete) states, need odd anharmonic terms $\hat{\mathcal{H}}_3$.

$$\mathcal{H}_{(2)} = J^* \left\{ \sum_i (\sigma_{ix}^2 + \sigma_{iy}^2) + \sum_{\langle ij\rangle} \left(\overbrace{-\tfrac{1}{2}}^{\cos(\theta_i-\theta_j)} \sigma_{ix}\sigma_{jx} + \sigma_{iy}\sigma_{jx} \right) \right\}$$

$J^* \equiv J \cdot \sqrt{\frac{S+1}{S}}$

$$\mathcal{H}_{(3)} = \frac{J^*}{S} \sum_{\langle ij\rangle} (\hat{x}_i \cdot \hat{z}_j) \left\{ -\tfrac{1}{2}(\sigma_{ix}\sigma_{jy}^2 - \sigma_{iy}^2\sigma_{jx}) - \tfrac{1}{6}(\sigma_{ix} - \sigma_{jx})^3 \right\}$$

$\sin(\theta_i - \theta_j) = \pm\frac{\sqrt{3}}{2} \equiv \eta_\alpha \cdot \frac{\sqrt{3}}{2}$ $\{\eta_\alpha$ = chirality of triangle α

$$\mathcal{H}_{(4)} = \frac{J^*}{S^2} \sum_{\langle ij\rangle} \Big\{ \tfrac{1}{16}(\sigma_{ix}\sigma \;\; -2\sigma_{ix}^2\sigma_{jx}^2 + \sigma_{ix}^3\sigma_{jx})$$
$$+ \tfrac{1}{16}(\sigma_{ix}\sigma_{jx}\sigma_{jy}^2 + \sigma_{jx}\sigma_{jx}\sigma_{iy}^2 - 2\sigma_{iy}\sigma_{jy}\sigma_{jx}^2 - 2\sigma_{iy}\sigma_{jy}\sigma_{ix}^2$$
$$- 2\sigma_{ix}^2\sigma_{jy}^2 - 2\sigma_{jx}^2\sigma_{iy}^2)$$
$$- \tfrac{1}{8}(\sigma_{iy}\sigma_{jy}^3 + \sigma_{iy}^3\sigma_{jy}^2 + \sigma_{iy}^3\sigma_{jy}) \Big\}$$

E.P. CHAN (Thesis 1994)

note powers $1, \frac{1}{S}, \frac{1}{S^2}$

$\sigma_{iy} \perp$ plane of spins

σ_{ix} (in layer) relative to $\hat{z}_i$; conjugate to σ_{iz}

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Indeed

$$\boxed{\mathcal{H}_3 = \sum_\alpha \eta_\alpha \hat{\mathcal{H}}_\alpha}$$

triangles

chiralities as coeff's (±1)

operator only involves the 3 spins in $\Delta\ \alpha$
[does NOT depend which coplanar state!]

∴ idea: 2nd order pert. theory

spinwave ground state

excited states

$$\delta \mathcal{E}^{pert} \approx \boxed{-\sum_n \frac{|\langle 0|\hat{\mathcal{H}}_3|n\rangle|^2}{\tilde{\mathcal{E}}_n}}$$

$$= -\sum_{\alpha\beta} \underbrace{\left(\sum_n \frac{\langle 0|\mathcal{H}^\Delta_\alpha|n\rangle\langle n|\mathcal{H}^\Delta_\beta|0\rangle}{\tilde{\mathcal{E}}_n}\right)}_{\equiv \frac{1}{2} J_{\alpha\beta}} \eta_\alpha \eta_\beta$$

$$\boxed{\mathcal{E}_{Ising} = -\frac{1}{2}\sum_{\alpha\beta} J_{\alpha\beta}\eta_\alpha\eta_\beta}$$

effective Ising Hamiltonian in terms of chiralities! But...

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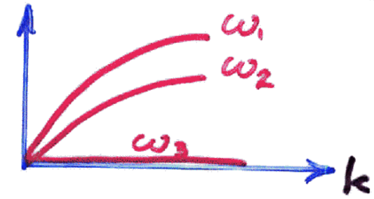
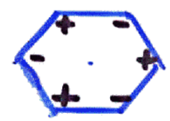
Problem: can't use $|0\rangle$ and $\tilde{\mathcal{E}}_n$ from bare spinwave $\hat{\mathcal{H}}_2$ — zero denominators!

branches of $\omega(k)$:



Interpretation: local "hexagon modes"



σ_y [out of plane] deviation costs zero to $\mathcal{O}(\sigma_y^2)$

[since $\delta \vec{L}_\alpha = 0$, where $L_\alpha \equiv \sum_{i\in\alpha} \vec{s}_i$ triangle's spin]

Why this is bad in $\delta\mathcal{E}^{pert}$:

- $\tilde{\mathcal{E}}_n = 0$
- $\langle \sigma_y^2 \rangle = 0$ due to soft branch zero-point fluct's

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Fix it: need anharmonic terms [even and odd] to make self-consistent $|0\rangle$ and $\tilde{\varepsilon}_n$ [Chubukov, 1992].

Soft modes acquire

$$\hbar\omega \approx \mathcal{O}(\mathcal{H}_4) - \mathcal{O}\left(\frac{\hat{\mathcal{H}}_3^2}{\tilde{\varepsilon}_n}\right) \sim S^{2/3}$$

Many other approxn's [Chan & Henley '95]

$\tilde{\varepsilon}_n \approx$ indep. of $\vec{K}_n$ [total mom. of exc state] $\approx \varepsilon_0$

$\mathcal{O}(S^{2/3})$

$$\mathcal{J}_{\alpha\beta} \approx \frac{1}{\tilde{\varepsilon}_0}\sum_n \langle 0|\mathcal{H}^\Delta_\alpha|n\rangle\langle n|\mathcal{H}^\Delta_\beta|0\rangle = \boxed{\frac{1}{\varepsilon_0}\langle \mathcal{H}^\Delta_\alpha \mathcal{H}^\Delta_\beta\rangle_0 < 0}$$

for nearest nbrs

$\mathcal{J}_{\alpha\beta} < 0 \Leftrightarrow$ antiferro. $\{\eta_{\alpha\beta}\} \Leftrightarrow$ $\boxed{\sqrt{3}\times\sqrt{3} \text{ state is lowest}}$

II. PYROCHLORE LARGE-S ANTIFERROMAGNET

Comparison to Kagome lattice

	Kagome	Pyrochlore
Spin order	Coplanar	Collinear
$\mathcal{H}_2$ breaks degeneracy?	No	Partially †
Symmetry between deviation components	No (in-plane and out-of-plane)	Yes (x and y)
Divergent modes in $\vec{q}$ space	An entire zone	Along lines
Correlations	Power law in S	Logarithmic in S
Anharmonic selection	$\mathcal{H}_3$ (*Chubukov 1992, Henley, Chan 1995*)	$\mathcal{H}_3=0$ $\mathcal{H}_4$?

† Entropy not extensive, Gauge equivalence between ground harmonic ground states (*Henley 2001*).

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[from U. Hizi, March 2004 talk]

pyh-4

Zero-point energy

linearize dynamics: $\boxed{\delta \dot{\vec{s}}_i = (\cdots)\, \delta \vec{s}_j}$

(details later)

$\Rightarrow$ spin wave (normal modes) $\{\omega\}$

[Note: some directions $\{\delta\vec{s}_i\}$ keep you in the ground-state manifold (though not symmetry). They have no "restoring force" $\Rightarrow \omega = 0$]

Quantize as harmonic oscillators

$\Rightarrow$ magnons (analog of phonons)

(In fact $\hbar\omega \sim |J| s \ll |E_0| \sim |J| s^2$: $\frac{1}{s}$ is the small parameter)

$$\boxed{E_{harm} = \sum \tfrac{1}{2}\hbar\omega}$$

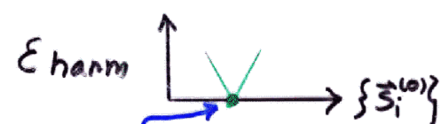
Different classical ground states $\{\vec{s}_i^{\,0}\}$ have different E_{harm} $\therefore$

$\therefore$ consider $\boxed{E_{harm}(\{\vec{s}_i^{\,(0)}\})}$ as an effective Hamiltonian (defined only on class. ground states)

True ground state has E_{harm} = minimum

"Order by disorder" (by fluctuations)

pyh-5



every collinear state is local minimum
[coplanar spin state on Kagomé]

collinear: $\vec{s}_i^{\,(0)} = s\, m_i \hat{z}$ ($m_i = \pm 1$)

Mapping: consider only collinear subspace (discrete)

(But still many: n° discrete gr. states $\sim e^{\text{const}\, N}$)

Consider $\mathcal{E}_{harm}(\{m_i\})$ as effective Hamiltonian on the discrete subspace

a) Kagomé case: all $e^{0.1N}$ coplanar states have same spectrum $\{\omega\} \Rightarrow$ same $\mathcal{E}_{harm}$!

$\Rightarrow$ need anharmonic terms to get a final unique gr. state

(E.P. Chan thesis, 1994 (with CLH))

b) Pyrochlore case: this calculation

preview of answer: collinear states have different $\mathcal{E}_{harm}$, but there are still $e^{(\text{const}\, L)}$ of minimum energy ($L \sim N^{1/3}$) DEGENERATE pyh-6

Dynamics (to get ω's): [semi] classical

$$\boxed{\hbar \dot{\vec{s}}_i = \vec{s}_i \times \vec{h}_i}$$

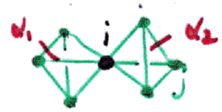
precess around this "local force field"

$$\boxed{\vec{h}_i = -\frac{\delta \mathcal{H}}{\delta \vec{s}_i} = -|J| \sum_{\substack{\text{nearest}\\ \text{neighbor}\\ \text{of } i}} \vec{s}_j}$$

Moessner-Chalker eq. of motion (for simplex spins $\vec{L}_\alpha$)

(1998)

$$\vec{h}_i \equiv |J| \left[(\vec{L}_{\alpha_1} - \vec{s}_i) + (\vec{L}_{\alpha_2} - \vec{s}_i) \right]$$



$\vec{s}_i \times \vec{s}_i = 0$

$$\Rightarrow \hbar \dot{\vec{s}}_i = -\vec{s}_i \times |J| \sum_{\alpha: i\in\alpha} \vec{L}_\alpha$$

neighbor connected by i

$$\boxed{\hbar \dot{\vec{L}}_\alpha \equiv \hbar \sum_{i\in\alpha} \dot{\vec{s}}_i = |J| \left(\sum_{i\in\alpha} \vec{s}_i \times (\vec{L}_\alpha + \vec{L}_{\beta(i)}) \right) = |J| \left(\underbrace{\cancel{\vec{L}_\alpha \times \vec{L}_\alpha}}_{0} + \sum_{\beta \text{ nbr. to } \alpha} \vec{s}_{i(\alpha\beta)} \times \vec{L}_\beta \right)}$$

site connecting simplex α to β

linearize

$$\boxed{\hbar\, \delta \dot{\vec{L}}_\alpha = |J| \sum_\beta \vec{s}^{(0)}_{i(\alpha\beta)} \times \delta \vec{L}_\beta}$$

Remark: only involves $\{\vec{L}_\alpha\}$ which live on a diamond lattice (simpler). The degrees of freedom we threw away all have $\omega \equiv 0$ $\therefore$ don't matter for E_{harm}.

pyh-7

Collinear case $\quad \vec{s}_i^{(0)} = \eta_i \hat{z}$

$$\boxed{\begin{cases} \hbar\, \delta \dot{L}_{\alpha x} = s|J| \sum_\beta \eta_{\alpha\beta}\, \delta L_{\beta y} \\ \hbar\, \delta \dot{L}_{\alpha y} = -s|J| \sum_\beta \eta_{\alpha\beta}\, \delta L_{\alpha x} \end{cases}}$$

matrix elements $\eta_{\alpha\beta} \equiv \eta_{i(\alpha\beta)}$

eigenvalue eq^n for frequencies

matrix square

$$\boxed{(\hbar\omega)^2 \delta L_{\alpha x} = -\hbar^2 \delta \ddot{L}_{\alpha x} = (s|J|)^2 (\eta^2)_{\alpha\gamma}\, \delta L_{\gamma x}}$$

$$\hbar\omega \equiv s|J| \cdot (\text{eigenvalue of } \eta^2)^{1/2}$$

$$\boxed{E_{harm}(\{\eta_i\}) = \tfrac{1}{2} \sum \hbar\omega = \tfrac{1}{2} \operatorname{Tr}((\eta^2)^{1/2})}$$

(the matrix *is* the Ising spin configuration)

Remarks

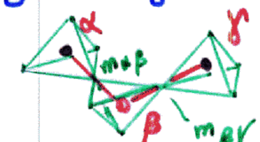
1. η^2 only connects even sites α, γ of diamond
   $\Rightarrow$ acts on fcc lattice ($\approx$ Bravais lattice)
   EASIER technically
2. This is gauge-invariant.
   Let $\theta_\alpha = \pm 1$ on each diamond site
   set $\eta'_{\alpha\beta} \equiv \theta_\alpha \theta_\beta \eta_{\alpha\beta}$
   ($\eta' = \Theta \eta \Theta^{-1}$ orthog. matrix Θ)

$\left.\begin{array}{l} \text{Same eigenvalues} \\ \text{Same } E_{harm}(\{\eta_i\}) \end{array}\right\} \Rightarrow$ DEGENERACY pyh. 8

write $\underline{\eta}^2 = 4\,\underline{I} + \underline{w}$

off diagonal terms connecting fcc neighbors

$W_{\alpha\gamma} \equiv \eta_{\alpha\beta}\,\eta_{\beta\gamma} = \pm 1$

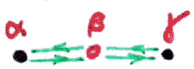


$E_{harm} = \frac{1}{2}|J|s\,\mathrm{Tr}(4+\underline{w})^{1/2}$

$$\boxed{E_{harm} = \tfrac{1}{2}|J|s\,\mathrm{Tr}\left\{4^{1/2}\left(1+\frac{w}{8}-\frac{w^2}{2^7}+\frac{w^3}{2^{10}}-\frac{5}{2^{15}}w^4+\dots\right)\right\}}$$

Note diag. terms in $\underline{\eta}$ independent of config.
since $(\eta^2)_{\alpha\alpha} = \sum_\beta (\eta_{\alpha\beta})^2 = \sum_{\text{neighbors}} 1$

Similarly terms in w^2, e.g.

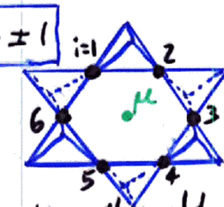


$\eta_{\alpha\beta}\,\eta_{\beta\gamma}\cdot\eta_{\beta\gamma}\,\eta_{\alpha\beta}$

The 1st nontrivial term comes in η^6 or w^3, since the smallest loop in diamond lattice is a hexagon
or pyrochlore
[There are also rings of length 8]

pyh-9

Mapping: let $\boxed{\tau_\mu \equiv \prod_{i\in \text{Hexagon}\ \mu} \eta_i = \pm 1}$



site of μ at center of hexagon
It turns out $\{\mu\}$ is a new pyrochlore lattice interpenetrating the old one!

plug in ⇒

$$\boxed{E_{harm} \cong |J|s\left\{1-\frac{1}{2^7}\cdot 12+\frac{1}{2^{10}}\cdot 48\langle\tau_\mu\rangle - \frac{5}{2^{15}}\cdot 12\left[25+12\langle\tau_\mu\rangle+4\langle\tau_\mu\tau_\nu\rangle\right]\right\}}$$

$\mathrm{Tr}\,w^2$ $\mathrm{Tr}\,w^3$ $\mathrm{Tr}\,w^4$

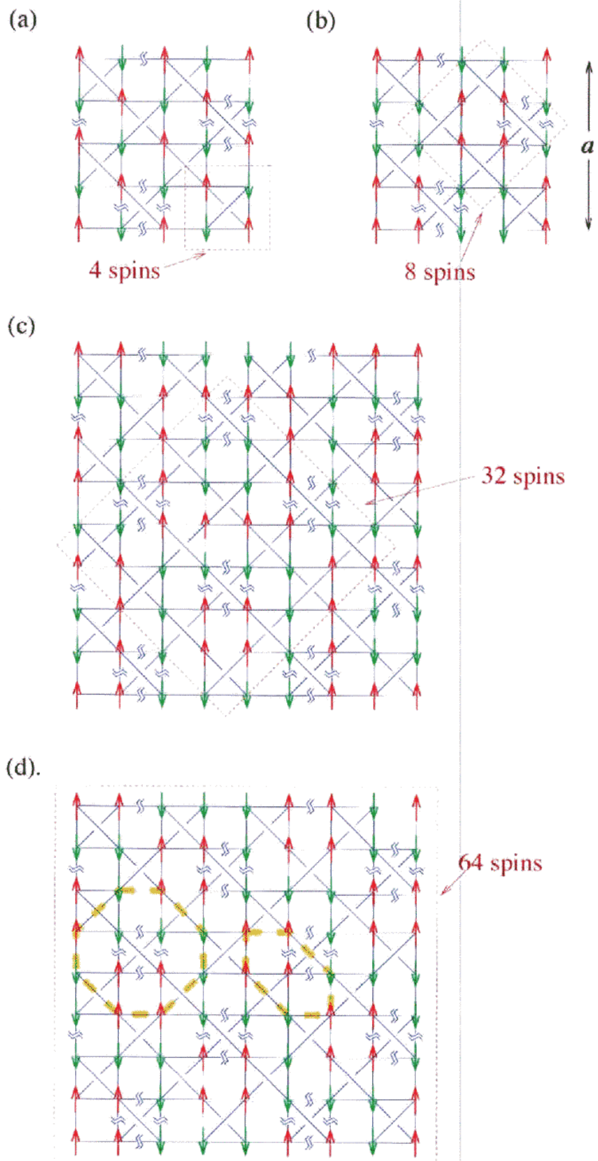
(here $\langle\dots\rangle$ means average over space)

i.e. $\boxed{E_{harm} \approx \text{const} + \tilde{h}\sum \tau_\mu - \tilde{J}\sum_{\langle\mu\nu\rangle}\tau_\mu\tau_\nu}$

It looks just like ferromagnetic Ising spins $\{\tau_\mu\}$ in an external field $\tilde{h}$.
∴ ground state is $\boxed{\tau_\mu \equiv -1}$

Indeed, we can find a set of $\{m_i\text{'s}\}$ that realize this, while still keeping $\sum_{i\in\alpha}\eta_i = 0$ (condition of classical ground state)

This is a surprisingly complicated ordering pattern. (Some simpler states all have $\tau_\mu \equiv +1$).



Concluding remarks : pyrochlore

- O. Tchernyshyov et al — "2D pyro."
  - a) thermal ensemble [CHECKERBOARD LATTICE]
  - b) on capped Kagomé lattice, smallest loops DON'T give right ans

- N.B. Hermele + MPA Fisher [Pyro. Ising model w/ ring exchange]

  ? might realize it for intermediate S

  $$e^{-\text{const}\, S} > K \sim (\text{small coeff.})\, S$$

  Must check that tunneling of the 6 spins isn't made incoherent by spin wave bath
  Compare von Delft + Henley (1992)

- Hexagon centers form a pyrochlore —

  exoticists, can you concoct an exactly self dual model?

Remarks - Kagomé

CLH has unpublished extensions to

a) 2-layer Kagomé sandwich lattice that models the experimental $S=3/2$ system Sr Cr Ga O

b). classical kagomé Heisenberg antiferromagnet

N.B. One can also use eff. Hamilton approach in a purely empirical fashion (fit a functional form to numerical results for a data base).