

**Ordering due to disorder and
gauge-like degeneracy in the
large- S antiferromagnet on the
highly frustrated pyrochlore lattice**

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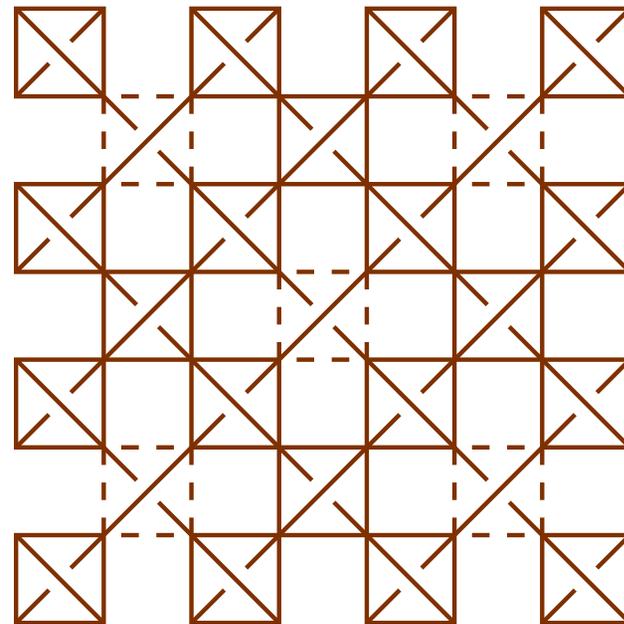
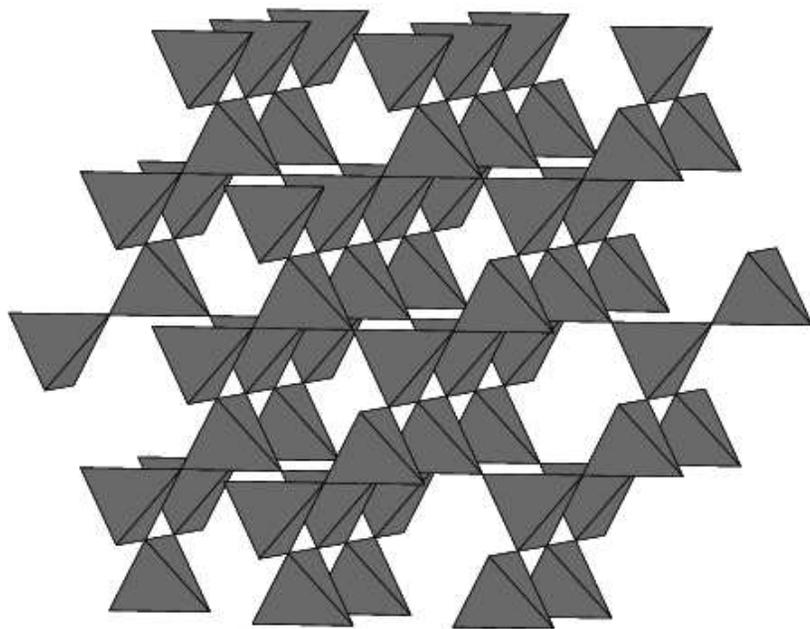
Effective Hamiltonians of the large-S pyrochlore antiferromagnet: outline

0. Introduction:
pyrochlore lattice (vs. Bethe lattice); effective Hamiltonians
1. Harmonic-order spinwaves
[CLH, PRL 96, 047201 (1006)]
[U. Hizi & CLH, PRB 73, 054403 (2006)]
- 2 . Trace expansion and loop effective Hamiltonian \mathcal{H}_{eff} .
- 3 . Gauge-like degeneracy of ground states
- [4.]. Quartic-order spinwaves and large-N expansion
(summary only)
5. Conclusions: (i) comparison to kagomé lattice
(ii) related systems (field plateaus)?
(iii) spin disordered states??

0. Introduction

Pyrochlore lattice

Corner sharing tetrahedra = bond midpoints of diamond lattice



Hamiltonian: Heisenberg Antiferromagnet

$$\mathcal{H} = \sum J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j = \frac{1}{2} \sum_{\substack{\text{tetra.} \\ \alpha}} |\mathbf{L}_\alpha|^2 + \text{const.}$$

$$\Rightarrow \mathbf{L}_\alpha \equiv \sum_{i \in \alpha} \mathbf{S}_i = 0 \quad (\text{tetrahedron sum in ground state})$$

Classically, any state with $\mathbf{L}_\alpha = 0$ is ground state.

Massive degeneracy \Rightarrow **rich phase diagrams** of correlated states

Our question: what is $S \gg 1$ ground state? expect spin order!

- disordered for classical [Moessner & Chalker 1998]
- ... or for $S = 1/2$ [Canals & Lacroix 1998]

Experimental pyrochlore systems?

Many $A_2B_2O_7$ oxides (pyrochlore structure) or AB_2O_4 (spinels):

- $Gd_2Ti_2O_7$: noncollinear (dipolar interaction)
[Champion *et al*, PRL 2001]
- $ZnCr_2O_4$: $S = 3/2$; Heisenberg behavior but distortion at lower T (magnetoelastic interaction) [S. H. Lee *et al*, PRL 2000, Nature 2002; see Tchernyshyov PRL 2002, 2004]
- $CdCr_2O_4$: Magnetization plateau; complicated helical ordering (Dzyaloshinskii-Moriya) [J.-H. Chung *et al*, PRL 2006]

Future clean realizations in cold gases?

[e.g. L. Santos *et al* 2004, theory kagomé optical lattice]

Effective Hamiltonian notion

Expect perturbation energies E' to split the degeneracies.

- Usual approach: (i) guess 2 or 3 simple/high symmetry states (ii) compute E' for each (iii) compare
- Our approach: define a function \mathcal{H}^{eff} for **every** state.

Advantages and disadvantages

- + If true ground state isn't one of those you guessed!?
- + Building block: (i) $T > 0$ simulation (ii) \mathcal{H}^{eff} + disorder, anisotropy, or tunneling among discrete states
- Usually uncontrolled expansion, crude (truncated) form.

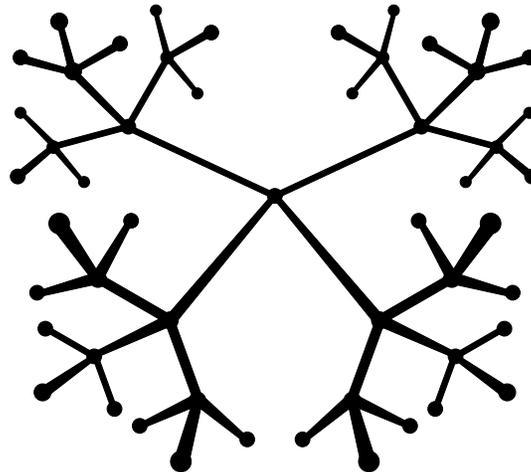
Want: (i) analytic form (ii) energy scale
(iii) ground states (iv) their degeneracy

Remark: Bethe lattice versus loops

We'll see the important states are discrete, like Ising model.

On Bethe lattice, **all** Ising states are symmetry equivalent
(hence exactly degenerate)

In pyrochlore lattice, \Rightarrow effective Hamiltonian \mathcal{H}^{eff} **must** depend
on loops.



1. Harmonic zero-point energy

Break degeneracy with zero-point spin wave energy:

$$E_{\text{harm}}(\{\hat{\mathbf{n}}_i\}) \equiv \sum_m \frac{1}{2} \hbar \omega_m = O(JS) \ll E_{\text{class}} = O(JS^2)$$

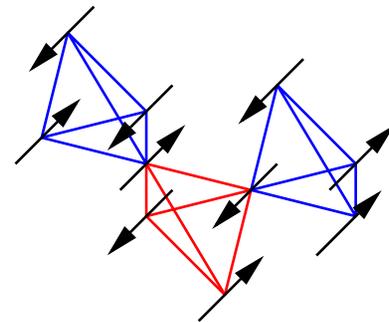
- Implicitly function of classical directions $\{\hat{\mathbf{n}}_i\}$
- Splits states because different magnon spectra

Collinear ground states $\hat{\mathbf{n}}_i = \eta_i \hat{z}$

avored by E_{harm} .

\Rightarrow Assume minimum E_{harm} is collinear state.

$\Rightarrow E_{\text{harm}}(\{\eta_i\})$ will give $\mathcal{H}_{\text{harm}}^{\text{eff}}$.



An aside about collinear selection

Shender 1982 (later CLH 1989): zero point energy favors states in which spins are collinear. (Why: this gives maximum coupling between fluctuations of different spins.) Like the entire spin wave energy it is down by $1/S$ relative to classical energy; coefficient is small.

In Kagome lattice, of corner sharing triangles, each triangle has 120° spin state; collinearity impossible, coplanarity favored instead. In Kagome, all the (many) coplanar states have *identical* spin wave spectra hence degenerate at harmonic order (extensive entropy of such states).

That was a consequence of neighbor spin angles $\theta_{ij} = \pm 2\pi/3$ everywhere; pyrochlore has $\theta_{ij} = 0, \pi$. So should split even in harmonic order!

2. Trace expansion and loop \mathcal{H}_{eff}

Spinwave equation of motion

Linearized eqn. of motion for “ordinary” spinwave modes?
[the other modes have $\omega = 0$]

Reduces to one for tetrahedron spin \mathbf{L}_α :

$$\delta \dot{\mathbf{L}}_\alpha = -SJ \sum_{\beta} \mu_{\alpha\beta} \hat{\mathbf{z}} \times \delta \mathbf{L}_\beta, \quad (1)$$

$$\mu_{\alpha\beta} \equiv \eta_{i(\alpha\beta)} \quad (2)$$

[$i(\alpha, \beta) \equiv$ pyrochlore site between diamond lattice sites α and β].

The dynamical matrix $\underline{\mu}$ is the classical configuration $\{\eta_i\}$.
(And its eigenvalues are ω_m .)

Trace expansion for E_{harm}

Eigenvalues of matrix μ^2 are $(\hbar\omega_m)^2$.

$$E_{\text{harm}}(\{\eta_i\}) \equiv \frac{1}{2} \sum_m \hbar\omega_m = JS\text{Tr} \left(\sqrt{\frac{1}{4}\mu^2} \right)$$

Formally Taylor-expand square root about constant matrix $A\mathbb{1}$:

$$E_{\text{harm}} = S\text{Tr} \left[A\mathbb{1} + \left(\frac{\mu^2}{4} - A\mathbb{1} \right) \right]^{1/2} \quad (3)$$

$$= S \sum_{k=0}^{\infty} c_{2k} A^{-(k-1/2)} \text{Tr} (\mu^{2k}) \quad (4)$$

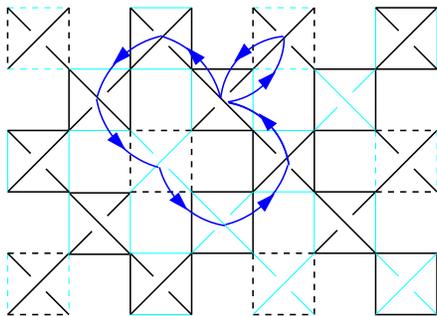
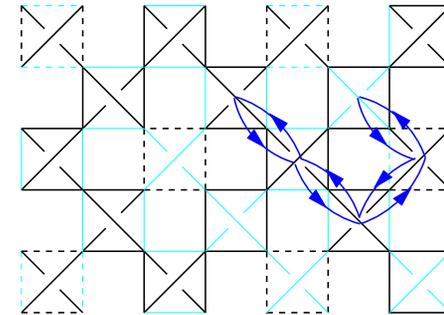
$\{c_{2l}\}$ have closed expressions]

[Result converges to A -indep. result, as long as $A > 1.4$].

Loop expansion

Each term of E_{harm} is $\text{Tr}(\boldsymbol{\mu}^{2k})$: a sum over products of $2k$ $\mu_{\alpha\beta}$'s:
 i.e., $\prod \eta_i$ (Ising spins) on all possible closed paths of $2k$ steps.

Retraced portions: trivial factors
 $\eta_i^2 \rightarrow 1$ (“decorations”). To resum,
 approximate (well) by Bethe lattice
 paths.



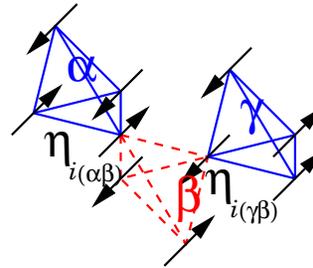
Path around a **loop**: contributes
 configuration-dependent factor $\prod \eta_i$
 around that loop.

Loop expansion 2

[Recall: Zero-point energy was:]

$$E_{\text{harm}} = JS \text{Tr} \left(\sqrt{\frac{1}{4} \mu^2} \right), \quad \mu_{\alpha\beta} \equiv \eta_{i(\alpha\beta)}.$$

Example of two factors in a term:



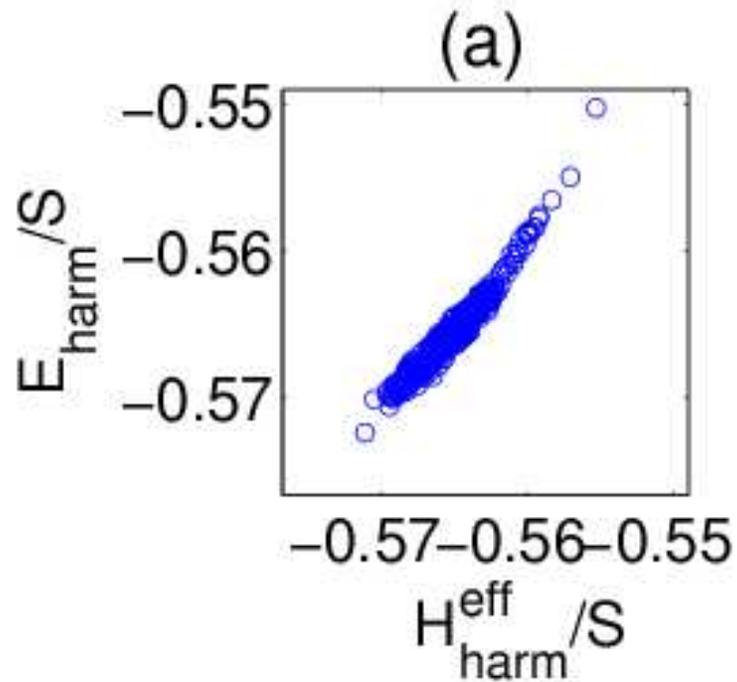
$$(\mu^2)_{\alpha\gamma} = \begin{cases} 4 & \alpha = \gamma \\ \eta_{i(\alpha\beta)} \eta_{i(\beta\gamma)} & \alpha, \gamma \text{ next-nearest neighbors} \\ 0 & \text{otherwise} \end{cases}$$

Result: Loop effective Hamiltonian

$$\mathcal{H}_{\text{harm}}^{\text{eff}} = -0.5640N_{\text{spins}} + 0.0136\Phi_6 - 0.0033\Phi_8 + \dots$$

(Here $\Phi_{2l} \equiv \sum \prod_{k=1}^{2l} \eta_{i_k}$.)

Numerical check (from database of numerous ground states.)



3. Gauge-like symmetry

Consider: take Ising configuration $\{\eta_i\}$, and

$$\eta'_{i(\alpha,\beta)} \equiv \tau_\alpha \tau_\beta \eta_{i(\alpha,\beta)}$$

with $\tau_\alpha = \pm 1$ arbitrarily (for every diamond site α .)

Matrix $\mu' = \tau \mu \tau^{-1}$, is similarity transformation.

Same eigenvalues \Rightarrow same E_{harm} .

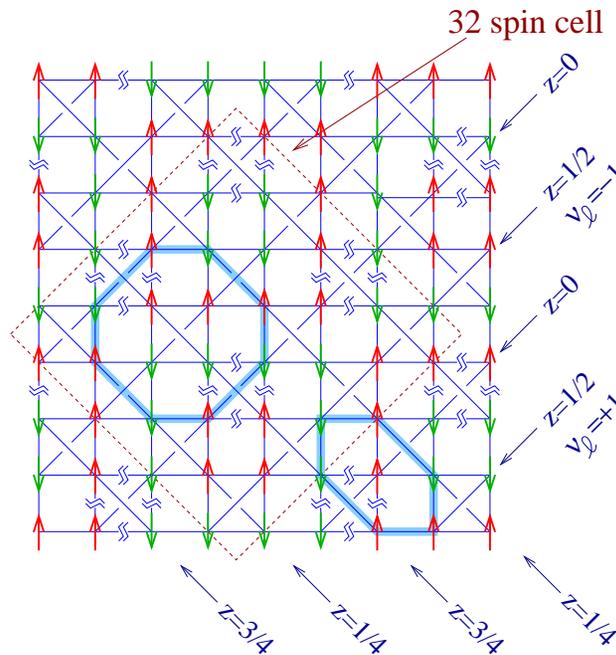
- Explains why $\mathcal{H}_{\text{harm}}^{\text{eff}}$ had form of gauge model.
- Large $[\exp(\text{const}L)]$ degeneracy!
- Gauge-like only: still must satisfy ground-state condition

$$\sum_{i \in \alpha} \eta_i = 0, \quad (\text{each tetrahedron } \uparrow\uparrow\downarrow\downarrow).$$

Ground-state degeneracy

Any collinear classical ground state with spin product $\prod_{i \in \hexagon} \eta_i = -1$ around **every** hexagon is a degenerate ground state.

- Called “ π -flux” state
- The smallest magnetic unit cell has 16 spins.
- The residual entropy is at least $O(L)$ and at most $O(L \ln L)$



4. Beyond harmonic spin-waves

Two routes to further break degeneracy

- Quartic-order spinwaves

[U. Hizi & CLH, J. Phys. Condens. Matt. 19, 145268 (2007);
cond-mat 0811.0395 [to appear PRB, 2009] see U. Hizi Ph. D.
thesis, 2006]

- Large-N [Sp(N)] mean-field theory

[U. Hizi, P. Sharma, and C. L. Henley PRL 95, 167203 (2005)]

These also lead to loop effective Hamiltonians

Effective Hamiltonian that splits harmonic states:

$$\mathcal{H}^{\text{eff}}_{\text{quart}} = C_6(S)\mathcal{P}_6 + C_8(S)\mathcal{P}_8 + \dots ,$$

where $\mathcal{P}_{2l} =$ no. AFM ($\uparrow\downarrow\uparrow\downarrow \dots$) loops of length $2l$;

$$C_{2l}(S) = O(\ln S^2).$$

Ground states for large N?

Monte Carlo simulate $\mathcal{H}^{\text{eff}}_{Sp(N)}$ (≤ 3456 sites): hunt ground states.

- nearly degenerate stacks of layers (alternate $a/4$, $a/2$ thick)
- Splitting $\sim 10^{-7}$ per spin (from length $2l = 16$ loops) \Rightarrow unique ground state (96 site cell).

Oops! $1/N$ expansion gave wrong answer!

- Harmonic term of $1/S$ expansion **must** dominate at $S \gg 1$
 \Rightarrow Physical ($SU(2)$) ground state **must** be “ π -flux” state.
 \Rightarrow ground states of $\mathcal{H}^{\text{eff}}_{Sp(N)}$ are **not** π -flux states.

Spin waves at quartic order give a similar (but not identical) stacked state with even longer period, which we think is right.

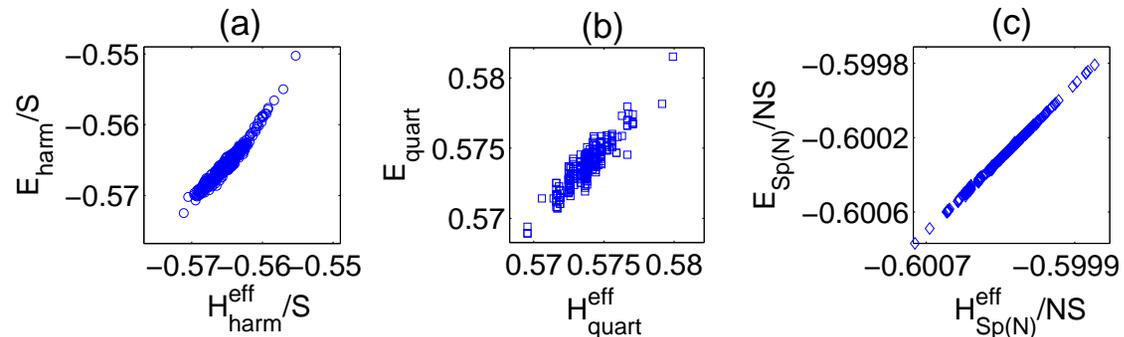
5. Conclusions: summary/discussion

Key tricks used:

- Get configuration labels as **coefficients** (in matrix)
- Zero-point energy in terms of **trace** $[(\mathbf{matrix})^2]$

Common features between harmonic/quartic/large-N calculations:

- Effective Hamiltonian as loop sum



- Degenerate (or nearly) states, entropy $O(L)$ (non extensive).

Practically: Energy differences too small to be seen experimentally.

Discussion

Details surprisingly unlike Kagomé case [E. P. Chan & CLH, 1994]

- Kagomé: always $\hat{\mathbf{n}}_i \cdot \hat{\mathbf{n}}_j = -1/2 \Rightarrow$ **all** coplanar states degenerate (**extensive** entropy) at harmonic order.
- Kagomé: zonefull of divergent modes $\Rightarrow E_{\text{anh}} = O(S^{2/3})$

Applicable to other systems

- checkerboard (planar pyrochlore) – confirm known answers
- Field plateau states (e.g. pyrochlore collinear $\uparrow\uparrow\uparrow\downarrow$)
(harmonic: **opposite sign** for loop product, $\prod \eta_i = +1$.)

Will addition of tunneling give spin-disordered superposition?

- No! $O(L)$ ground state entropy \Rightarrow must flip $O(L^2)$ spins \Rightarrow tunnel amplitude exponentially small.
- **Will** work in kagomé case. [See von Delft & Henley PRB 1993]

Disc. details 1: Comparison to Kagomé case

	Kagomé	Pyrochlore
1. Spin order	Coplanar	Collinear
2. Symmetry between deviation components	\Rightarrow No (in-plane and out-of-plane)	\Rightarrow Yes (x and y)
3. Divergent modes in \vec{q} space	\Rightarrow entire zone	\Rightarrow Along lines
4. Anharm. E scale	$S^{2/3}$	$(\ln S)^2$
5. Neighbor spin angles	$2\pi/3$	0 or π
6. $\mathcal{H}_{\text{harm}}$ gr. st. degen.?	\Rightarrow all	$\Rightarrow O(L)$ entropy
7. Anharmonic terms	$\mathcal{H}_{\text{cubic}} + \mathcal{H}_{\text{quart}}$	$\mathcal{H}_{\text{cubic}}=0$
8. \mathcal{H}_{eff} interactions	\Rightarrow nonlocal	\Rightarrow local

Discussion details 2: Other systems

(1) Harmonic $\mathcal{H}^{\text{eff}}_{\text{harm}}$:

- checkerboard (planar pyrochlore) lattice
[Done by Tchernyshyov-Starykh-Moessner-Abanov PRB 2003]
- field plateaus (kagomé $\uparrow\uparrow\downarrow$, pyrochlore $\uparrow\uparrow\uparrow\downarrow$)
(Opposite sign for loop product of Ising spins!)

(2) Quartic $\mathcal{H}^{\text{eff}}_{\text{quart}}$:

- checkerboard lattice [But lattice already breaks symmetry.]
- field plateaus (see list above): **undone**

(3) Large- N $\mathcal{H}^{\text{eff}}_{Sp(N)}$:

- checkerboard lattice [same answer as Bernier *et al* PRB 2004]
- Kagomé lattice [same answer as Sachdev PRB 1992]

Disc. details 3: disordered superpositions??

Just add to effective Hamiltonian the “off-diagonal” terms, representing tunneling between collinear states?

[See von Delft & Henley PRB 1993, kagomé; see Bergman *et al*, nearly-Ising pyrochlore at mag. plateau]

- Expect, if diagonal terms (i.e. \mathcal{H}^{eff}) \ll tunneling amplitudes.
- The extra terms are ring-exchanges [like Hermele et al, 2004]
- Mixture would be quantum spin nematic.

But **no!**:

- $O(L)$ ground state entropy \Rightarrow need to flip $O(L^2)$ spins \Rightarrow tunnel amplitude **exponentially small**.
- Try kagomé lattice (or garnet lattice – 3D!) built from triangles: there, harmonic-order ground states with **extensive** entropy.

4a. Quartic (self-cons.) spin-waves: details

Holstein-Primakoff transformation/expansion

- Relate standard bosons to spin deviation operators:

$$a_i = (\eta_i \sigma_i^x + i \sigma_i^y) / \sqrt{2S}.$$

- Hamiltonian: classical + harmonic + quartic

$$\begin{aligned} \mathcal{H} &= -JN_s S^2 \\ &+ J \left(1 + \frac{1}{2S}\right) \sum_i |\vec{\sigma}_i|^2 + J \left(1 + \frac{1}{4S}\right)^2 \sum_{\langle ij \rangle} \vec{\sigma}_i \cdot \vec{\sigma}_j \\ &+ \frac{J}{4S^2} \sum_{\langle ij \rangle} \left(\eta_i \eta_j |\vec{\sigma}_i|^2 |\vec{\sigma}_j|^2 - \frac{1}{2} \vec{\sigma}_i \cdot \vec{\sigma}_j (|\vec{\sigma}_i|^2 + |\vec{\sigma}_j|^2) \right) + \mathcal{O}(S^{-1}). \end{aligned}$$

(Harmonic term – which gave E_{harm} –

depends on $\{\eta_i\}$ via $[\sigma_i^x, \sigma_j^y] = i\eta_i S \delta_{ij}$)

Self-consistent decoupling (standard)

“Mean-field” Hamiltonian: as if modify bonds

$$\delta J_{ij} = -\frac{1}{2S^2} \left[\frac{1}{2} (\langle \sigma_i^2 \rangle + \langle \sigma_j^2 \rangle) - \eta_i \eta_j \langle \sigma_i \cdot \sigma_j \rangle \right]$$

Numerically: dominated by **divergent** modes (with $\hbar\omega_m = 0$).

Consider only harmonic ground (“ π -flux”) states: totally uniform in space (for harmonic=gauge-invariant properties).

Symmetry $\Rightarrow \langle \sigma_i \cdot \sigma_j \rangle$ depends only on $\eta_i \eta_j \Rightarrow$

$$\delta J_{ij} \cong -\frac{\epsilon}{8} \eta_i \eta_j$$

with $\epsilon = O(\ln S/S)$ [from integrate across divergence in B.Z.]

Effective Hamiltonian for quartic energy

Most of anharmonic energy is actually “gauge”-invariant

Effective Hamiltonian that splits harmonic states:

$$\mathcal{H}^{\text{eff}}_{\text{quart}} = C_6(S)\mathcal{P}_6 + C_8(S)\mathcal{P}_8 + \dots ,$$

where \mathcal{P}_{2l} = no. AFM ($\uparrow\downarrow\uparrow\downarrow \dots$) loops of length $2l$;

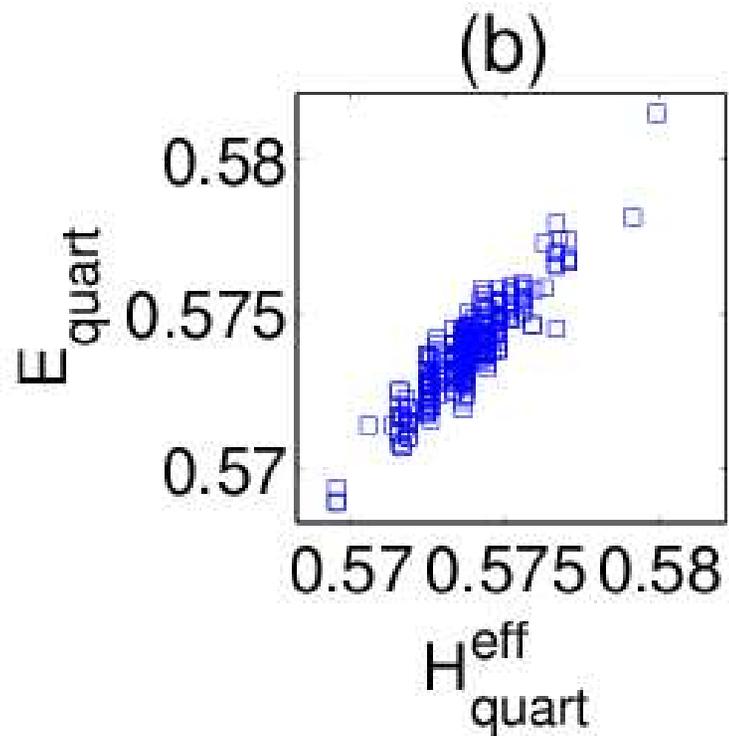
- Obtained from empirical fit (first!)
- Lead term (\mathcal{P}_6) derived (later!) by inserting $\delta J_{ij} = (\dots)\epsilon\eta_i\eta_j$ into loop expansion from harmonic calculation.
- Energy scale $C_{2l}(S) = O(\ln S^2)$.

Ground state(s) – our grand answer – but complicated/unclear:

- Independent slabs, 2 choices/slab, alternately $a/2$ or $a/4$ thick
- Degenerate (?) \Rightarrow entropy = $O(L)$? I think not, but shortest loop that **might** split them is length 26(!)

Numerical check

Using database of many “ π -flux” (harmonic ground states)



5b. Large- N approach to large- S limit

with: Dr. Prashant Sharma

Motivation for large N approach

[U. Hizi, P. Sharma, and C. L. Henley, PRL 2005]

Another systematic approach beyond classical/harmonic level

- Generalized spins with $\text{Sp}(N)$ symmetry
[Physical: $\text{Sp}(1) = \text{SU}(2)$].
- Can solve $N \rightarrow \infty$ exactly. [Read & Sachdev, 1989]

N flavors of boson; representation labeled by κ (generalizes $2S$).

- Usual: **small** κ to approximate $S = 1/2$
- Ours: **large** κ – breaks degeneracies at lowest nontrivial order
(first nontrivial term in $1/\kappa$ expansion)

Set-up: Large-N mean field theory

Spin operators: bilinear in boson operators $b_{i\sigma m}, b_{i\sigma m}^\dagger$

[$\sigma = \uparrow, \downarrow$; flavor $m = 1, \dots, N$]

Exchange interaction: bilinear in “valence bond” operators

$$\hat{Q}_{ij} \equiv b_{i\uparrow,m}^\dagger b_{j\downarrow,m}^\dagger - b_{i\downarrow,m}^\dagger b_{j\uparrow,m}^\dagger.$$

Decoupling gives

$$\mathcal{H}_{Sp(N)} = \frac{1}{2} \sum_{\langle ij \rangle} \left(N |Q_{ij}|^2 + Q_{ij} \hat{Q}_{ij} + H.c. \right) + \sum_i \lambda_i \left(\hat{N}_i^b - N\kappa \right)$$

with classical coeff. $Q_{ij} \equiv \langle \hat{Q}_{ij} \rangle / N$ (self-consistent).

Lagrange multipliers $\lambda_i \rightarrow 4\kappa$ (every i): enforce κ bosons, each site

Ordered state: has $\langle b_{i\sigma,m} \rangle \neq 0$.

Trace expansion for large-N case

Bogoliubov diagonalization \Rightarrow zero-point energy is

$$E_{Sp(N)}(\{Q_{ij}\}) = \frac{N}{2} [\text{Tr} \sqrt{\lambda^2 \mathbb{1} - \mathbf{Q}^\dagger \mathbf{Q}} - N_s \lambda]$$

If a **collinear** type ordered state:

$$Q_{ij} = \kappa(\eta_i - \eta_j)/2 \quad [= 0 \text{ or } \pm \kappa].$$

Expand trace as previous cases!

- Different matrix (only connects on satisfied bonds)
- Different lattice (pyrochlore \Rightarrow trivial loops on 1 tetrahedron)

Effective Hamiltonian for large N case

Real-space expansion in *loops* of valence bonds:

$$\mathcal{H}_{Sp(N)}^{\text{eff}} = \frac{N\kappa}{2} (0.59684N_{\text{spins}} - 0.003482 \mathcal{P}_6 - 0.0000344 \mathcal{P}_8 + \dots)$$

[As in quartic. $\mathcal{P}_{2l} \equiv$ no. AFM loops ($\uparrow\downarrow\uparrow\downarrow \dots$) of length $2l$.]

Numerical check
with general
collinear ground states

