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 (2006)]
   [U. Hizi & CLH, PRB 73, 054403 (2006)]

2. Trace expansion and loop effective Hamiltonian $\mathcal{H}_{\text{eff}}$.

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

\[ \mathcal{H} = \sum J_{ij} S_i \cdot S_j = \frac{1}{2} \sum_{\text{tetra.}} |L_\alpha|^2 + \text{const.} \]

\[ \Rightarrow \quad L_\alpha \equiv \sum_{i \in \alpha} S_i = 0 \quad \text{(tetrahedron sum in ground state)} \]

Classically, any state with \( L_\alpha = 0 \) 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 $H^\text{eff}$ for every state.

Advantages and disadvantages

+ If true ground state isn’t one of those you guessed!?

+ Building block: (i) $T > 0$ simulation (ii) $H^\text{eff}$ + 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 $\mathcal{H}^\text{eff}$ must depend on loops.
1. Harmonic zero-point energy

Break degeneracy with zero-point spin wave energy:

\[ E_{\text{harm}}(\{\hat{n}_i\}) = \sum_{m} \frac{1}{2} \hbar \omega_m = O(JS) \ll E_{\text{class}} = O(JS^2) \]

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

Collinear ground states \( \hat{n}_i = \eta_i \hat{z} \)
favored by \( E_{\text{harm}} \).

\[ \Rightarrow \text{Assume minimum } E_{\text{harm}} \text{ is collinear state.} \]
\[ \Rightarrow E_{\text{harm}}(\{\eta_i\}) \text{ will give } \mathcal{H}_{\text{harm}}^{\text{eff}}. \]
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 $120^\circ$ 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 $\mathcal{H}_{\text{eff}}$

Spinwave equation of motion

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

Reduces to one for tetrahedron spin $L_\alpha$:

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

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

$[i(\alpha, \beta) \equiv \text{pyrochlore site between diamond lattice sites } \alpha \text{ and } \beta]$.

The dynamical matrix $\mu$ is the classical configuration $\{\eta_i\}$.
(And its eigenvalues are $\omega_m$.)
Trace expansion for $E_{\text{harm}}$

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

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

Formally Taylor-expand square root about constant matrix $A1$:

$$E_{\text{harm}} = S\text{Tr} \left[ A1 + \left( \frac{\mu^2}{4} - A1 \right) \right]^{1/2} \quad (3)$$

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

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

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

Each term of $E_{\text{harm}}$ is $\text{Tr}(\mu^{2k})$: 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 $\eta_i^2 \to 1$ (“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:]

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

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

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

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

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 \( \alpha \)).

Matrix \( \mu' = \tau \mu \tau^{-1} \), is similarity transformation. Same eigenvalues \( \Rightarrow \) same \( E_{\text{harm}} \).

- 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 $\prod_{i \in \bigcirc} \eta_i = -1$ around every hexagon is a degenerate ground state.

- Called “$\pi$-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

- 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:

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

where \( \mathcal{P}_{2l} = \text{no. AFM } (\uparrow\downarrow\uparrow\downarrow \ldots ) \text{ loops of length } 2l; \]
\( C_{2l}(S) = O(\ln S^2). \)
Ground states for large $N$?

Monte Carlo simulate $\mathcal{H}_{\text{eff}}^{Sp(N)}$ ($\leq 3456$ 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 “$\pi$-flux” state.

$\Rightarrow$ ground states of $\mathcal{H}_{\text{eff}}^{Sp(N)}$ are not $\pi$-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** \([\text{(matrix)}^2]\)

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

- Effective Hamiltonian as loop sum

    ![Graphs (a), (b), and (c)]

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

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

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

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

- 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 $\mathcal{H}_{\text{quart}}^{\text{eff}}$:

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

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

- 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. $\mathcal{H}^{\text{eff}}$) $\ll$ 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:
  \[ a_i = \left( \eta_i \sigma_i^x + i \sigma_i^y \right) / \sqrt{2S}. \]

- Hamiltonian: classical + harmonic + quartic
  \[
  \mathcal{H} = -J N_s S^2 \\
  + J \left( 1 + \frac{1}{2S} \right) \sum_i |\vec{\sigma}_i|^2 \\
  + J \left( 1 + \frac{1}{4S} \right)^2 \sum_{\langle ij \rangle} \vec{\sigma}_i \cdot \vec{\sigma}_j \\
  + \frac{J}{4S^2} \sum_{\langle ij \rangle} \left( \eta_i \eta_j |\vec{\sigma}_i|^2 |\vec{\sigma}_j|^2 - \frac{1}{2} \vec{\sigma}_i \cdot \vec{\sigma}_j \left( |\vec{\sigma}_i|^2 + |\vec{\sigma}_j|^2 \right) \right) + \mathcal{O}(S^{-1}).
  \]

(Harmonic term – which gave \( E_{\text{harm}} \) –)
depends on \( \{\eta_i\} \) via \([\sigma^x_i, \sigma^y_j] = i\eta_i S \delta_{ij}\)
Self-consistent decoupling (standard)

“Mean-field” Hamiltonian: as if modify bonds

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

Numerically: dominated by \textbf{divergent} modes (with \( \hbar \omega_m = 0 \)).

Consider only harmonic ground (“\( \pi \)-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:

\[ \mathcal{H}_{\text{quart}}^{\text{eff}} = C_6(S)P_6 + C_8(S)P_8 + \ldots, \]

where \( P_{2l} = \text{no. AFM (↑↓↑↓... loops of length } 2l; \)

- Obtained from empirical fit (first!)
- Lead term \( (P_6) \) derived (later!) by inserting \( \delta J_{ij} = (\ldots)\epsilon \eta_i \eta_j \)
  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 $N \to \infty$ exactly. [Read & Sachdev, 1989]

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

- Usual: small $\kappa$ to approximate $S = 1/2$

- Ours: large $\kappa$ – 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 \(m = 1, \ldots, N\)]

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 \(\lambda_i \to 4\kappa\) (every \(i\)): enforce \(\kappa\) 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

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

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:

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

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

Numerical check
with general
collinear ground states