# Order from disorder in the Pyrochlore lattice Heisenberg Antiferromagnet

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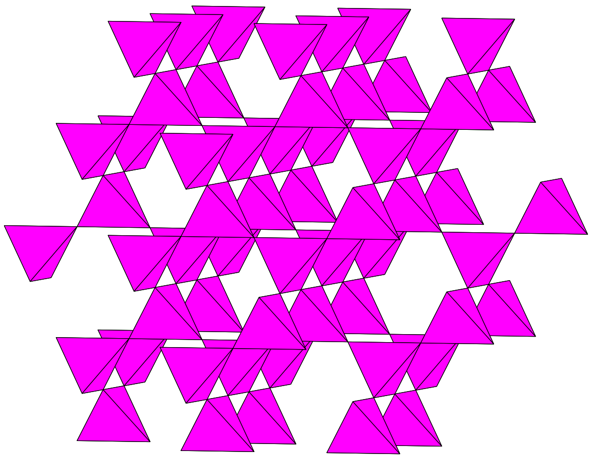
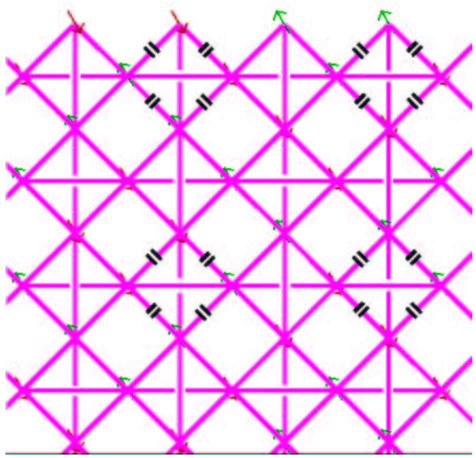
## Abstract

We study the Heisenberg model on the pyrochlore lattice using anharmonic spin-waves. We have done a variational calculation of the large $S$ quartic energy and find the all harmonic degeneracies are broken. We perform this calculation on a large set of computer generated harmonic ground states to find the unique ground state. We present an effective Hamiltonian for this model. We also apply a large $N$ calculation on the same system, and find that the degeneracy is not broken at first order in $1/\kappa$.

## The Pyrochlore lattice

- 3-dimensional, cubic symmetry.
- Composed of corner sharing tetrahedra.
- Appears in many compounds, most notably B sites in $A_2B_2O_7$ oxides.

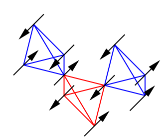




## The Pyrochlore Heisenberg Antiferromagnet

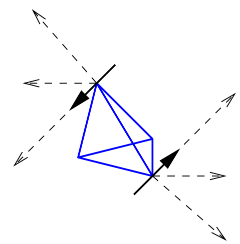
$$\mathcal{H} = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j = \sum_{\substack{tetra. \\ \alpha}} \left| \sum_{i \in \alpha} \vec{S}_i \right|^2 + \text{const.}$$

- Classically, all states with zero sum in each tetrahedron (tetrahedron rule) are degenerate.
- In large $S$ limit, quantum fluctuations (or classical thermal fluctuations) choose a subset of the collinear ground states . Ground state is characterized by Ising variables $\vec{S}_i = \eta_i \hat{z}$, $\eta_i \in \{\pm 1\}$, $\sum_{i \in \alpha} \eta_i = 0$



- Ground state manifold is massively degenerate.
- Does the large $S$ quantum model possess long range order?

## Large S expansion



- Holstein Primakoff transformation:

  - Change to local coordinates: $\mathsf{x} = \eta_i x$, $\mathsf{y} = y$, $\mathsf{z} = \eta_i z$.
  - Express spin components in terms of bosons $\{a_i\}$, $\{a_i^\dagger\}$:

$$
\begin{aligned}
S_i^{\mathsf{z}} &= S - a_i^\dagger a_i\,, \\
S_i^{+} &\equiv S^{\mathsf{x}} + iS^{\mathsf{y}} = \sqrt{2S - a_i^\dagger a_i}\; a_i \approx \sqrt{2S}\left(1 - \frac{a_i^\dagger a_i}{4S}\right) a_i\,, \\
S_i^{-} &\equiv S^{\mathsf{x}} - iS^{\mathsf{y}} = a_i^\dagger\sqrt{2S - a_i^\dagger a_i} \approx \sqrt{2S}a_i^\dagger\left(1 - \frac{a_i^\dagger a_i}{4S}\right).
\end{aligned}
$$

  - Spin deviations $\sigma_i^x \equiv \eta_i\sqrt{\frac{S}{2}}(a_i + a_i^\dagger)$, $\sigma_i^y \equiv -i\sqrt{\frac{S}{2}}(a_i - a_i^\dagger)$, with commutation $[\sigma_i^x, \sigma_j^y] = i\eta_i S\delta_{ij}$.

- Hamiltonian:

$$
\begin{aligned}
\mathcal{H} &= -JN\left(S + \frac{1}{2}\right)^2 \\
&\quad + J\left(1 + \frac{1}{2S}\right)\sum_i |\vec{\sigma}_i|^2 + J\left(1 + \frac{1}{4S}\right)^2 \sum_{\langle ij\rangle} \vec{\sigma}_i\cdot\vec{\sigma}_j \\
&\quad + \frac{J}{4S^2}\sum_{\langle ij\rangle}\left(\eta_i\eta_j|\vec{\sigma}_i|^2|\vec{\sigma}_j|^2 - \frac{1}{2}\vec{\sigma}_i\cdot\vec{\sigma}_j\left(|\vec{\sigma}_i|^2 + |\vec{\sigma}_j|^2\right)\right) + \mathcal{O}(S^{-3})\,.
\end{aligned}
$$

## Harmonic Hamiltonian

$$\mathcal{H} = (\boldsymbol{\sigma}^x)^T \, \mathbf{H} \boldsymbol{\sigma}^x + (\boldsymbol{\sigma}^y)^T \, \mathbf{H} \boldsymbol{\sigma}^y \, .$$

- This should be diagonalized to obtain the spin waves modes $\{\mathbf{v}_p\}$, corresponding to frequencies $\{\omega_p\}$.
- An important feature of the harmonic term in the Hamiltonian, is that if $\sum_{i\in\alpha} \mathbf{v}_p^i = 0$ for all tetrahedra $\alpha$, then $\mathbf{H}\mathbf{v}_p = 0$. Such a *zero mode* does not contribute to the harmonic zero-point energy $E_2 = \frac{\hbar}{2} \sum_p \omega_p$.
- This allows us map all *non-zero modes* to tetrahedron variables $\mathbf{L}^{x/y} = \sum_{i\in\alpha} \boldsymbol{\sigma}^{x/y}$.

## Harmonic conclusions

Writing things in terms of tetrahedra variables, one obtains (*Henley 2001*):

- Harmonic zero point energy is invariant under a "gauge" transformation in which $L_\alpha \to -L_\alpha$. This is a gauge freedom that arises from the tetrahedron zero sum constraint.
- Effective harmonic Hamiltonian is sum over products of spins around loops

$$E_2^{eff} = Const. + A \sum_{\hexagon} \prod_{i\in\hexagon} \eta_i - B \sum_{\bigcirc} \prod_{i\in\bigcirc} \eta_i + \ldots \, .$$

- All classical ground states with negative spin product around all hexagons are degenerate ground states of the harmonic Hamiltonian. The number of degenerate ground states is reduced from $e^{Const.\times L^3}$ to $e^{Const.\times L}$.

## Divergent fluctuations

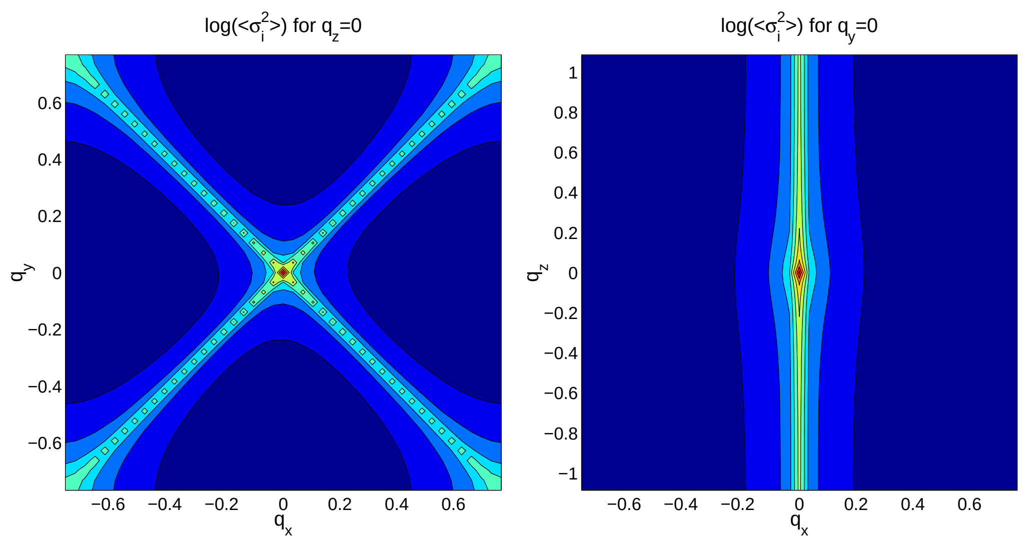
- The equations of motion:

$$\boldsymbol{\eta} \equiv \begin{pmatrix} \eta_1 & 0 & 0 & \cdots & 0 \\ 0 & \eta_2 & 0 & \cdots & 0 \\ 0 & 0 & \eta_3 & \cdots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \cdots & \eta_N \end{pmatrix}$$

$$\begin{aligned} \dot{\boldsymbol{\sigma}}^x &= -\frac{i}{\hbar}[\boldsymbol{\sigma}^x, \mathcal{H}] = \frac{2S}{\hbar}\boldsymbol{\eta}\mathbf{H}\boldsymbol{\sigma}^y, \\ \dot{\boldsymbol{\sigma}}^y &= -\frac{i}{\hbar}[\boldsymbol{\sigma}^y, \mathcal{H}] = -\frac{2S}{\hbar}\boldsymbol{\eta}\mathbf{H}\boldsymbol{\sigma}^x. \end{aligned}$$

- The spin waves are eigenmodes of $(\boldsymbol{\eta}\mathbf{H})^2$. These are also eigenvectors of $\boldsymbol{\eta}\mathbf{H}$, if it is diagonalizable. However, if $\boldsymbol{\eta}\mathbf{H}$ cannot be diagonalized, we obtain pairs of conjugate modes zero modes satisfying $\boldsymbol{\eta}\mathbf{H}\mathbf{w}^y \sim \mathbf{v}^x$, $\boldsymbol{\eta}\mathbf{H}\mathbf{v}^x = 0$, resulting in divergent fluctuations.

- In $\vec{q}$ space, the divergent modes live on one-dimensional lines. These lines are the same for gauge equivalent states.



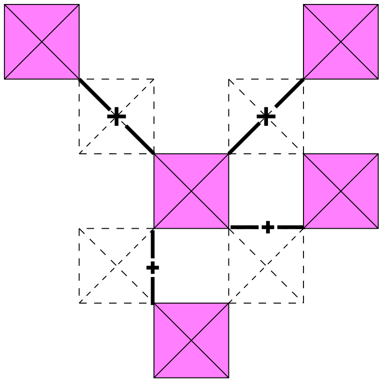

- After Fourier transform back to real space, one obtains logarithmically divergent fluctuations.

## 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 |
| Fluctuations | Power law in $S$ | Logarithmic in $S$ |
| Anharmonic selection | $\mathcal{H}_3$ (*Chubukov 1992, Henley, Chan 1995*) | $\mathcal{H}_3$=0<br>$\mathcal{H}_4$? |

## Harmonic ground state generation

- Gauge transformations involve flipping the Ising spins in entire tetrahedra. However these transformations are not local since they must be done in a way that conserves the tetrahedron rule on neighboring tetrahedra.
- Even and odd gauge transformations commute and can be applied separately.
- A valid even (or odd) transformation is a network on the FCC lattice, where each flipped tetrahedron is connected to four others by satisfied bonds, each belonging to one of the neighboring odd (even) tetrahedra .



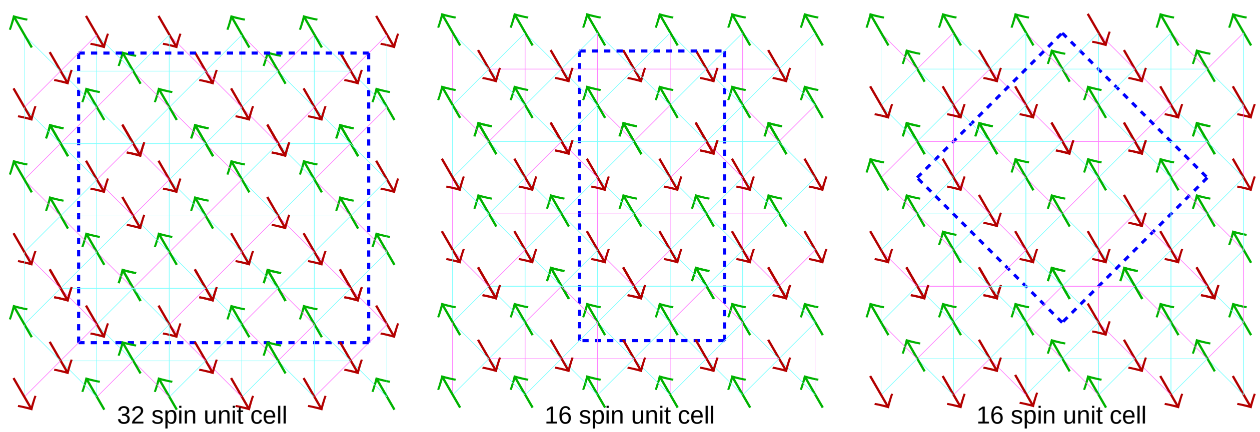
- In a 128 site unit cell (composed of two layers along each direction), we generated random gauge transformations and found approximately 140 harmonic ground states.

## Quartic large S

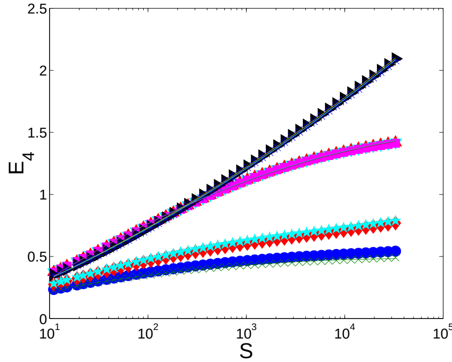
- Problem: Quartic energy corrections would be calculated by decoupling the quartic interaction term, and plugging in the harmonic fluctuations. However, some of the modes have divergent fluctuations!
- On the other hand, since there are no cubic terms in the Hamiltonian, we could still consider Gaussian modes, i.e. use a harmonic variational Hamiltonian. Simplest choice:

$$\mathbf{H}_{var} = \mathbf{H}_2 + \delta \boldsymbol{\eta} \mathbf{H}_2 \boldsymbol{\eta} + \varepsilon I \ .$$

- Spin rotation symmetry requires $\varepsilon + 4\delta = 0$, so there is only one parameter.
- Diagonalize equations of motion obtained from $\mathbf{H}_{var}$ and calculate (Gaussian) fluctuations. Plug into decoupled $\mathcal{H}$. Minimize the total energy to get self-consistent energy.
- We applied this calculation to the randomly generated harmonic ground states, as well as to various other classical ground states.
- Examples of some of the harmonic ground states with small single layer unit cells:



## Effective Hamiltonian (1): Gauge invariant




- The main contribution to the energy from the quartic terms is gauge invariant.
- This term only depends on the number of divergent modes. The effective Hamiltonian has the form

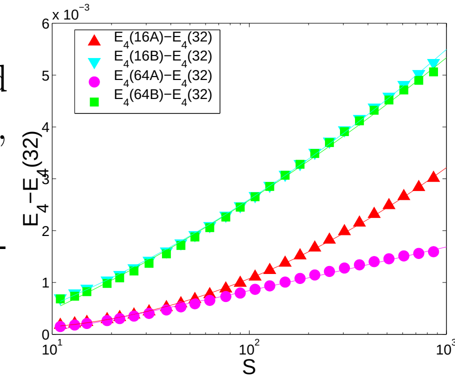
$$E_4^{eff}(\text{gauge inv.}) = A_0(lnS) + A_1(lnS)\sum_{\hexagon}\prod_{i\in\hexagon}\eta_i + A_2(lnS)\sum_{\octagon}\prod_{i\in\octagon}\eta_i + \ldots .$$

where

$$\begin{aligned} A_0(lnS) &= 0.0357 + 0.1009lnS + 0.0048(lnS)^2\,, \\ A_1(lnS) &= 0.0497 - 0.0441lnS - 0.0016(lnS)^2\,, \\ A_1(lnS) &= 0.0119 - 0.0091lnS + 0.0053(lnS)^2\,, \end{aligned}$$

- The gauge invariant Hamiltonian has the same form as the effective harmonic Hamiltonian, but with opposite signs.
- The quartic energy scales as a second order polynomial in $\ln S$, as expected.

## Effective Hamiltonian (2): Gauge dependent



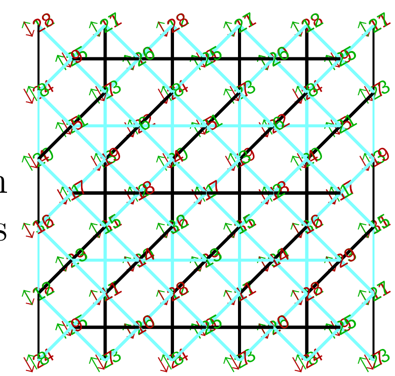

- Although the leading order quartic energy is the same for all harmonic ground states, there is a quartic energy difference between gauge equivalent states, 2-3 orders of magnitude less than the gauge invariant term.
- The leading order in the effective gauge dependent Hamiltonian can be written in terms of 2-spin 2nd and 3rd neighbor Ising terms.

$$E_4^{eff}(\text{gauge dep.}) = B_0(lnS) \sum_{\substack{ij \;\; \text{2nd neigh.} \\ |\vec{r}_i - \vec{r}_j| = 2}} \eta_i \eta_j + B_1(lnS) \sum_{\substack{ij \;\; \text{3nd neigh.} \\ |\vec{r}_i - \vec{r}_j| = 2}} \eta_i \eta_j + \ldots .$$

- These are the simplest non-trivial Ising terms, however they are not enough to explain the choice of a unique state.

## Ground state selection

- The state that we found to have the lowest energy is (in (001) projection:
- This is an example of "order from disorder": quantum fluctuations, which normally work against ordering, lift the ground state degeneracy and thus restore order.



## Large N Calculation

- In addition to the spin wave calculation, we also performed a large $N$ calculation to first order in $1/\kappa$. Similar studies on other systems have found that all degeneracies are broken at the lowest order (*Sachdev 1992*).

- Using Schwinger bosons, the Heisenberg Hamiltonian can be written:

$$\mathcal{H} = \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j = \sum_{\langle ij \rangle} (: B_{ij}^{\dagger} B_{ij} : - A_{ij}^{\dagger} A_{ij}) \,, + \sum_i \lambda_i (b_{i\uparrow}^{\dagger} b_{i\uparrow} + b_{i\downarrow}^{\dagger} b_{i\downarrow} - 2S)$$

  where the last term is a constraint on each site

$$B_{ij} = \frac{1}{2}(b_{i\uparrow}^{\dagger} b_{j\uparrow} + b_{i\downarrow}^{\dagger} b_{j\downarrow}) \,, \qquad A_{ij} = \frac{1}{2}(b_{i\uparrow} b_{j\downarrow} - b_{i\downarrow} b_{j\uparrow}) \,.$$

- The generalization to large $N$ involves adding more flavors of Schwinger bosons:

$$B_{ij} = \frac{1}{2}\sum_m (b_{im\uparrow}^{\dagger} b_{jm\uparrow} + b_{im\downarrow}^{\dagger} b_{jm\downarrow}) \,, \qquad A_{ij} = \frac{1}{2}\sum_m (b_{im\uparrow} b_{jm\downarrow} - b_{im\downarrow} b_{jm\uparrow}) \,.$$

- The constraint can now be replaced by a constraint for every boson flavor $m$ $\sum_\sigma b_{im\sigma}^{\dagger} b_{im\sigma} = \kappa$ (*Sachdev and Read 1991*) or an average constraint $\sum_{m\sigma} b_{im\sigma}^{\dagger} b_{im\sigma} = \kappa N$ (*Ceccatto, Gazza, and Trumper 1993*). Here $\kappa$ takes the place of $2S$.

## Large N calculation: first order in $1/\kappa$

- The leading order term correction to the classical energy, in $1/\kappa$ is obtained by taking the classical expectation values $\langle A_{ij}\rangle$, $\langle B_{ij}\rangle$, and plugging them into a decoupled quartic Hamiltonian.
- On then obtains a quadratic Hamiltonian of the form

$$\frac{1}{N}\mathcal{H}_q = \kappa \mathbf{\Psi}^\dagger \mathbf{D} \mathbf{\Psi}\,,$$

where

$$\mathbf{\Psi} = \begin{pmatrix} \tilde{\mathbf{b}}_\uparrow \\ \tilde{\mathbf{b}}^\dagger_\downarrow \end{pmatrix}\,,$$

and

$$D = \begin{pmatrix} \lambda + \mathbf{R}_{ij} & -\mathbf{Q}_{ij} \\ \mathbf{Q}^*{}_{ij} & \lambda + \mathbf{R}_{ij} \end{pmatrix}\,, \qquad Q_{ij} = \sin\frac{\theta_i - \theta_j}{2}\,, \qquad R_{ij} = \cos\frac{\theta_i - \theta_j}{2}$$

- This Hamiltonian can be diagonalized to obtain eigenvalues $\{\Omega_p\}$, and the quantum correction to the energy is $\sum_p \Omega_p$.

## Large N results

- If we apply the large $N$ calculation to first order in $1/\kappa$, using the constraint on every Schwinger boson flavor we find that the ground states are not among the large-S harmonic ground states.
- If we apply the calculation with an average constraint on the Schwinger bosons, we do find that the lowest energy is for the quartic spin-wave ground state.
- However, we find that there are various states that are degenerate to the numerical precision we have been able to achieve.

## Conclusions and future work

- Using the self-consistent calculation, we break the degeneracy between various harmonic ground states, and find a unique ground state.
- We obtain an effective Hamiltonian in terms of the Ising spins, with both gauge dependent, and gauge invariant contributions. Further work is needed to understand the origin of the ground state selection and obtaining an effective Hamiltonian that has a unique ground state.
- Using a large $N$ calculation the classical degeneracies are not completely broken, to the accuracy we can obtain. We are working on calculating the next order in the $1/\kappa$ expansion, as well as trying to understand the origin of these degeneracies.