Semiclassical Ordering in the Large-$N$ Pyrochlore Antiferromagnet

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We study the semiclassical limit of the Sp($N$) generalization of the pyrochlore lattice Heisenberg antiferromagnet by expanding about the $N \rightarrow \infty$ saddlepoint in powers of a generalized inverse spin. To leading order, we write down an effective Hamiltonian as a series in loops on the lattice. Using this as a formula for calculating the energy of any classical ground state, we perform Monte Carlo simulations and find a unique collinear ground state. This state is not a ground state of linear spin-wave theory, and can therefore not be a physical ($N = 1$) semiclassical ground state.

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Geometrically frustrated antiferromagnets [1,2] have attracted interest because their large classical ground state degeneracy can allow a rich variety of correlated states, including (at $T = 0$) quantum spin liquids or complex ordered states. The simplest examples are nearest-neighbor, exchange-coupled antiferromagnets in which spins from triangles or tetrahedra share corners [3]: the kagomé, “checkerboard,” and SCGO ($\text{SrCr}_2\text{Ga}_{12-9\gamma}\text{O}_{19}$) lattices [1] in two dimensions, plus the garnet and pyrochlore lattices in three dimensions. The pyrochlore, in particular, consists of tetrahedra whose centers form a diamond lattice. The Hamiltonian is $H = \sum J_{ij} S_i \cdot S_j$, where $J_{ij} = 1$ for nearest neighbors ($ij$). In fact, additional terms—dipole interactions and anisotropies (as in $\text{Gd}_3\text{Ti}_2\text{O}_7$), and magnetoelastic couplings (as in $\text{ZnV}_2\text{O}_4$ and $\text{ZnCr}_2\text{O}_4$)—decide the order in most real materials [1,2]. Still, the case with pure Heisenberg exchange is worth understanding since (i) most simulations are done for this case; (ii) the more realistic systems emerge from it by the addition of perturbations; (iii) this has motivated experimentalists to search for model systems in which the aforementioned perturbations are small; (iv) quantum effects can be studied without being overshadowed by classical effects.

What is the ground state for large spin length $S$? In unfrustrated antiferromagnets, it is just the classical ground state dressed with zero-point fluctuations of harmonic spin waves, and in frustrated cases the spin-wave zero-point energy may lift the degeneracy of classical ground states [4]. In the pyrochlore case, though, a large degeneracy remains [5]; its resolution by higher-order (anharmonic) terms in the semiclassical (1/$S$) expansion requires arduous approximations [6].

An established alternative to the spin-wave approach is to generalize the Heisenberg spins [with $SU(2) \equiv \text{Sp}(1)$ symmetry] to Sp($N$) symmetry [7]; here $N$ is the number of flavors of Schwinger bosons whose bilinear form represents a generalized spin [7], with length $\kappa = 2S$. The resulting mean-field theory (valid in the $N \rightarrow \infty$ limit) is popular as an analytic approach to the $S = 1/2$ limit, since the small-$\kappa$ limit captures various disordered and exotic ground states [7,8]. The large-$N$ mean-field theory is also useful at large $\kappa$ for this Letter’s problem, since it gives a simple analytical prescription for ground state selection: unlike the spin-wave expansion, here all degeneracies are (typically) broken at the lowest-order [$O(1/\kappa)$] quantum correction [7,9].

However, on highly frustrated lattices this approach has the complication of a macroscopic (exponential) number of degenerate saddle points not related by symmetry, so it is unknown a priori which of these should be expanded around; this was handled till now by limiting the investigation to ordering patterns of high symmetry and small magnetic cells, or by enumerating all saddle points in a small finite system [9,10]. In this Letter, we develop an effective Hamiltonian [3] approach to this question. The pertinent saddle points are labeled by arrangements of valence bond variables, and we obtain a simple formula for the large-$N$ mean-field energy of any classical ground state, as a function of these variables. The effective Hamiltonian is constructed as an analytical real-space expansion of loops made of valence bonds. This allows us to systematically search for a collinear pyrochlore ground state, using Monte Carlo annealing, on quite large system sizes. However, we also find that the pyrochlore ground state does not agree with even the lowest-order term in the spin-wave expansion, and therefore cannot give the right answer for the physical ($N = 1$) ground state, in the large-$S$ limit, demonstrating a limitation of the large-$N$ approach for this case.

Large $N$ mean-field theory.—We begin by discussing the mean-field Hamiltonian derived from the Sp($N$) generalization of $H$. For the $N = 1$ case we can write the spin interaction in terms of Schwinger boson operators as $\hat{S}_i \cdot \hat{S}_j = b^\dagger_{i\sigma} b_{i\sigma} b^\dagger_{j\sigma'} b_{j\sigma'}$, where $\sigma$ and $\sigma'$ (that take values $\uparrow, \downarrow$) is implied. The Hilbert space of the spin model is obtained by constraining the number of bosons on each site $b^\dagger_{i\sigma} b_{i\sigma} = 2S$. We can rewrite the interaction in terms of valence bonds created by the operator $\mathbf{e}_{\sigma\sigma'} b^\dagger_{i\sigma} b^\dagger_{j\sigma'}$, where $\mathbf{e}_{\parallel} = -\mathbf{e}_{\perp} = 1$. An arbi-
trary singlet state can be written in terms of some arrangement of these bonds with at most 2$^3$ bonds emanating from any lattice site. Generalizing these bond operators to $N$ flavors allows us to put a large number of bonds on a link. Since the Hamiltonian acting on a state changes at most two bonds per link, the relative change in the number of bonds goes like $1/NS$. In the large-$N$ limit, their fluctuations are quenched. Therefore, we factorize the interaction in terms of valence bonds $Q_{ij} = (\epsilon_{x,y} b_{x,m}^\dagger b_{y,m}^\dagger)/N$, where the flavor index $m = 1, 2, \ldots, N$. We treat $Q_{ij}$ as classical quantities, to obtain the mean-field Hamiltonian

$$H_{\text{MF}} = \frac{1}{2} \sum_{ij} \left[ N |Q_{ij}|^2 + (\epsilon_{x,y} x_{i,m} x_{j,m} Q_{ij} + \text{H.c.}) \right] + \sum_i \lambda_i (b_{i,m}^\dagger b_{i,m} - N\kappa). \quad (1)$$

Here Lagrange multipliers $\lambda_i$ have been introduced to enforce the constraint on boson number $N\kappa$ at every site $i$, defining the generalized spin length $\kappa = 2S$. In what follows, we shall take $\lambda_i$ to be spatially uniform. We are interested in large enough values of $\kappa$ to condense a flavor mode of the itinerant bosons. $\langle b_{i,m} \rangle = \sqrt{N}\delta_{i,m} x_{i,m}$ for long-range order to develop. The mean-field ground state energy (per flavor) is obtained by diagonalizing (1) by a canonical Bogoliubov transformation:

$$E_{\text{MF}} N = \frac{1}{2} \sum_{ij} \left[ |Q_{ij}|^2 + (\epsilon_{x,y} x_{i,m} x_{j,m} Q_{ij} + \text{c.c.}) \right] + \sum_i \lambda_i (x_{i,m}^2 - \kappa) + \frac{1}{2} \text{Tr} \left[ \Delta^2 1 - Q^\dagger Q - N, \lambda \right] \quad (2a)$$

Here $N_s$ is the number of lattice sites, and (2b) is the zero-point energy contribution of the bosons. The exact mean-field ground state is obtained by a constrained minimization of the above expression. It can be systematically approached as an expansion in powers of $1/\kappa$. The leading contribution to the energy (of order $\kappa^2$) comes from terms in (2a), whose minimization simply relates the valence bonds to the condensate configuration in the classical ground state(s) of the Heisenberg Hamiltonian $H$ with spin size $\kappa/2$. We will denote this configuration of bond variables with a superscript $c$: $\{Q_{ij}^c\}$. The quantum correction (of order $\kappa$) is provided by terms in (2b) for these bond configurations.

The ground states of the classical Hamiltonian (2a) consist of all spin configurations in which the spin vectors sum to zero in every tetrahedron. On general grounds we expect quantum corrections to select collinear ground states from the classical manifold [3,4,11]. We therefore restrict our attention to such states, in which each spin can be denoted by an Ising variable $\eta_i \in \{\pm 1\}$. Collinearity implies that, up to an arbitrary gauge transformation, $Q_{ij} = \kappa (\eta_i - \eta_j)/2$ and thus the bond variables are $\pm \kappa$ for every satisfied, antiferromagnetic (AFM) bond, and zero otherwise. Also, $\lambda^c = 4\kappa$ for all pyrochlore lattice classical ground states.

Loop expansion and effective Hamiltonian.—Next, we recast the first quantum correction to the mean-field energy, Eq. (2b), for a given collinear classical ground state, into an effective Hamiltonian form where only some of the degrees of freedom remain [3]. Equation (2b) can formally be Taylor-expanded

$$E_q N = -\frac{1}{2} \sum_{m=1}^{\infty} \frac{(2m + 1)!}{\lambda^{2m-1} m!} \text{Tr} (Q^\dagger Q)^m. \quad (3)$$

Since $|Q_{ij}/\kappa| = 1$ for AFM bonds, and zero otherwise, $\text{Tr} (Q^\dagger Q)/\kappa^2$ equals to the number of closed paths of length $2m$, composed of AFM bonds. All terms in Eq. (3) depend solely on the structure of the network formed by AFM bonds. Note that since this network is bipartite, each nonzero element of $Q^\dagger Q$ is $\kappa^2$.

In any collinear classical ground state, each tetrahedron has two up spins and two down spins, and four AFM bonds forming a closed loop [see Figs. 1(a)–1(d)]. This means that the local connectivity of the AFM network is identical for all states, and many closed paths only contribute state-independent terms to Eq. (3). For example, $\text{Tr} Q^\dagger Q = 4N, \kappa^2$ for any classical ground state since the only paths of length 2 involve going to and fro on the same bond, and each site has four neighbors which have the opposite spin [see Fig. 1(a)]. Similarly $\text{Tr} (Q^\dagger Q)^2 = (16 + 12 + 4)N, \kappa^4$, where the three terms correspond to the paths shown in Figs. 1(b)–1(d), respectively. All paths that do not involve loops [e.g., those in Figs. 1(a)–1(c)] can be viewed as paths on a Bethe lattice of coordination 4 and would contribute a constant term to the energy for all collinear classical ground states. The same is true for paths involving only trivial loops, in addition to the Bethe lattice path, as in

![Diagram](image-url)
evaluate the coefficients to any accuracy in Eq. (4), using

\[ \frac{E^{\text{eff}}}{N(k/2)} = K_0 + K_6 P_6 + K_8 P_8 + K_{10} P_{10} + \cdots, \]  

where \( \{K_m\} \) are numerical coefficients, and \( P_m \) is the number of nontrivial AFM loops of length \( 2m \), per site.

To evaluate the coefficients \( \{K_m\} \), we need to calculate two types of terms: (i) The number \( F(2m) \) of closed paths of total length \( 2m \) on a decorated (with trivial 4 loops) coordination-4 Bethe lattice [Fig. 1(e)]. (ii) The number \( G(2l,2m) \) of closed paths of length \( 2m + l \), involving a particular loop of length \( 2l \) with decorated Bethe lattice paths emanating from each site along the loop [Fig. 1(f)]. Calculating these terms is a matter of tedious but tractable combinatorics. We find that the functions \( F(2m) \), \( G(2l,2m) \) decay rapidly with \( m \), allowing us to sum them in order to evaluate the coefficients to any accuracy in Eq. (4), using

\[ K_0 = \sum_{m=0}^{\infty} F(2m), \quad K_{2l} = \sum_{m=0}^{\infty} G(2l,2m). \]  

We show the first five coefficients in Table I. Thus we have obtained an effective Hamiltonian that is parametrized solely by the number of AFM loops of various sizes. Note that the coefficients decay rapidly \( K_{2l+2}/K_{2l} \approx 1/10 \), which leads us to expect short loops to be the dominant terms in the expansion. This allows us, in principle, to calculate the energy, to any accuracy, for any member of an infinite ensemble of classical ground states. This represents a significant improvement over previous calculations that were always limited to small system sizes [9,10].

Although we derived the effective Hamiltonian for collinear states, it turns out that, in fact, the classical tetrahedron zero sum rule implies that Eq. (4), with the coefficients in Table I, is valid for any noncollinear classical ground state, as well, with the generalized loop variables expressed as sums over nontrivial loops

\[ P_{2l} = \frac{1}{k^{2l}} \sum_{(i_1,\ldots,i_{2l})} \text{Re}(Q_{i_1 i_2}^\dagger Q_{i_3 i_4}^\dagger \cdots Q_{i_{2l-1} i_{2l}}^\dagger Q_{i_1 i_2}). \]  

Unlike the collinear case, where the elements of \( Q^\dagger Q \) could only take the values 0 or \( k^2 \), and thus each loop would contribute 0 or 1 to the sum (6), in the general case, the matrix elements of \( Q^\dagger Q \) are complex.

Numerical results.—To verify the validity of the effective Hamiltonian (4), we calculated the energy for a large number of collinear classical ground states, as well as linear spin-wave ground states, obtained by a random flipping algorithm described elsewhere [11]. We find that the energies are remarkably well described by \( E^{\text{eff}} \), even when we cut the expansion (4) off at \( 2l = 8 \), as shown in Fig. 2. We used the coefficient values of Table I, but had to adjust the constant term \( K_0 \) separately for each choice of cutoff, in order to get a good fit [12]. In practice, this means that the effective Hamiltonian is extremely useful for comparing energies of various states, even with a small cutoff, but requires many terms in order to accurately determine the energy. An independent 5-parameter numerical fit, to Eq. (4), up to \( 2l = 12 \), gives the values shown in the right-hand column of Table I.

Now that we have an approximate formula for \( E^{\text{eff}} \), for any collinear classical ground state, we can systematically search these states, with large magnetic unit cells, to find a ground state. We conducted Monte Carlo simulations using a Metropolis loop flipping algorithm and the effective energy of Eq. (4), for various orthorhombic unit cells of sizes ranging from 128 to 3456 sites, with periodic boundary conditions. We find a minimum energy of \( E^{\text{eff}}/(Nk/2) = -0.60077N_s \) for a family of nearly degenerate states. They are composed of layers, that can each be in one of four arrangements, resulting in \( \sim e^{L/2} \) states, where \( L \) is the system size, and \( c \) is a constant. Each of these states has \( P_6 = N_s/3 \), which is the maximum value that we find (but is not unique to these states), and \( P_8 = 23N_s/6 \). Upon closer investigation, however, we find that a unique ground state (depicted in Fig. 3) is selected. The energy difference

![FIG. 2 (color online). Calculated energies \( E^{\text{eff}} \) of 50 sample classical ground states (open symbols), 16 of which are harmonic spin-wave ground states (squares), along with \( E^{\text{eff}} \), with \( 2l \leq 6 \) (dashed line) and \( 2l \leq 8 \) (solid line). The constant term \( K_0 \) was numerically fitted (see main text). The inset shows the linear spin-wave energy for the same states. Although the spin-wave energy tends to be lower for states with lower \( E^{\text{eff}} \), the large-\( N \) ground state need not be a spin-wave ground state.](image)
to nearby states is of order $10^{-7}N$, corresponding to the $2l = 16$ term.

Discussion.—It was noted by one of us [5] (see also Ref. [13]) that, in the pyrochlore, the degeneracy of ground states of the spin-wave quantum Hamiltonian, at the lowest order in $1/S$, is associated with a "gaugelike" symmetry. This symmetry characterizes the degenerate submanifold of collinear ground states by the condition

$$\prod_{i \in \Omega} \eta_i = -1,$$

(7)

for all nontrivial hexagons. Since the spin-wave theory is expected to be exact in the limit of infinite $S$, the physical ground state must satisfy Eq. (7). The state depicted in Fig. 3, however, does not. Looking at the inset in Fig. 2, we find that states with negative hexagon products tend to have lower large-$N$ energy than other states, since they tend to have more AFM loops, but this is not a strict rule. Thus it would seem that the $N \rightarrow \infty$, large-$\kappa$ ground state cannot be the physical ($N = 1$) large-$S$ ground state.

Nevertheless, if we restrict the large-$N$ calculation to harmonic spin-wave ground states only, we find that the ordering of energies for various states is similar to preliminary anharmonic spin-wave results [6], and does predict the same ground state. As shown in Fig. 4, in both cases, the lowest energy among harmonic spin-wave ground states belongs to a state with the most AFM hexagons.

The effective Hamiltonian approach that we have outlined here can easily be applied to other lattices. In the checkerboard lattice, the energy is lowest for states that have the most AFM (square) nontrivial plaquettes. Thus, the nondegenerate ground state is clearly the $(\pi, \pi)$ state in which all plaquettes are AFM [10]. In the kagomé case, all classical ground states are noncollinear. However, if we limit ourselves to coplanar arrangements, we find that $Q_{ij}$ has the same absolute value for all of the lattice bonds, but the signs differ depending on the chirality of the triangle to which the bond $(i, j)$ belongs. Therefore, the effective Hamiltonian (4), with the generalized variables (6), prefers classical ground states with negative product of triangle chiralities around all hexagons. One can thus conclude that the ground state is the $\sqrt{3} \times \sqrt{3}$ state, as large-$N$ calculations have indeed found [9]. Let us also remark that our method can be generalized to long-range Heisenberg interactions which are relevant in the context of real materials like Tb$_2$Ti$_2$O$_7$ [12].

Finally, it has been suggested that the disordered (small-$\kappa$) limit of the large-$N$ approximation for the pyrochlore lattice also has a massive multiplicity of saddle points [14]; an effective Hamiltonian similar to this Letter’s could organize the handling of this family.


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[12] We could get a cutoff-independent constant term $\vec{K}_0$ if we replaced $P_{ij}$ by $\bar{P}_{ij} = P_{ij} - \langle P_{ij} \rangle$, where $\langle P_{ij} \rangle$ is the ensemble average of $P_{ij}$, and took $\vec{K}_0 = \vec{K}_0 + \sum \vec{K}_j (P_{ij})$.


FIG. 3 (color online). The ground state of our large-$N$ theory, as viewed in a (001) projection. Here, light (dark) bonds represent AFM (ferromagnetic) bonds (unlike in Fig. 1). The shown pattern is repeated along x and y directions, as well as in adjacent z slices. This state has a 48 site magnetic unit cell.

FIG. 4 (color online). Per site large-$N$ energies $E_N$ calculated for various harmonic spin-wave ground states (bottom), compared to the per site spin-wave energy obtained from an anharmonic calculation for $S = 1500$ (top). In both cases the lowest energy is for a state that maximizes the number of AFM hexagons.