Singlet Excitations in Pyrochlore: A Study of Quantum Frustration

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We apply the contractor renormalization (CORE) method to the spin half Heisenberg antiferromagnet on the frustrated checkerboard and pyrochlore lattices. Their ground states are spin-gapped singlets which break lattice symmetry. Their effective Hamiltonians describe fluctuations of orthogonal singlet pairs on tetrahedral blocks, at an emergent low energy scale. We discuss low temperature thermodynamics and new interpretations of finite size numerical data. We argue that our results are common to many models of quantum frustration.

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Frustration in classical spin models often leads to a complex energy landscape. Certain models, such as the Heisenberg antiferromagnet on the pyrochlore lattice, have an extensively degenerate ground state manifold. This model, given by $H = J\sum_{\langle ij\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$, has spins $\mathbf{S}_i$ sitting on corner sharing tetrahedral units [see Fig. 1(a)]. In the *semiclassical* approximation [1–3], a large degeneracy survives the quantum fluctuations, and thus resists ground state selection by the "order from disorder" mechanism.

A pressing open question is what happens in the *strong* quantum limit, e.g., the spin half case. Series expansions [4] suggest rapid decay of spin correlations. Does this indicate a translationally invariant spin liquid or a lattice symmetry breaking valence bond solid? In the quantum case, is there an emerging low energy scale, in lieu of the classical ground state degeneracy?

The purpose of this Letter is to derive the low energy effective Hamiltonian starting from the Heisenberg model. As a warm-up to the pyrochlore lattice, we treat its two-dimensional reduction, the checkerboard lattice [Fig. 1(b)], which has recently received significant theoretical attention [5–9].

Our approach is the contractor renormalization (CORE) method [10]. The CORE is a real-space discrete renormalization transformation invented by Morningstar and Weinstein. It maps a lattice Hamiltonian to an effective Hamiltonian, with the same low energy spectrum, on a coarse grained lattice. CORE computes the effective interactions at all ranges using exact diagonalizations of finite connected clusters. Truncation of interactions beyond a finite range is an approximation, whose error can be estimated numerically from the next higher range terms. Although CORE involves clustering of the lattice to disjoint blocks, it is different from the perturbative techniques in Refs. [11–13] since it does not rely on weakening the interblock couplings. Instead, convergence of the cluster expansion is controlled by an emerging length scale (coherence length) associated with the effective degrees of freedom. CORE has been successfully applied to describe the spectra of Heisenberg models on chains and ladders [10,14]. Recently, it was applied to the square lattice Hubbard model to derive the plaquette boson-fermion model for cuprate superconductors [15]. We refer the reader to previous reviews [10,15] for the mathematical background and technical details.

For each of the Hamiltonians at hand, we define local operators from the lowest eigenstates of the elementary clusters, e.g., a tetrahedral unit in the pyrochlore lattice. We shall compute the effective interactions by CORE up to four clusters range, and estimate the truncation error.

Our key results are the following: *For the checkerboard,* we confirm the conclusions of Refs. [6,8,9], derived by other means, that the ground state is closely approximated by the product of uncrossed plaquette singlets. The effective pseudospin Hamiltonian allows us to interpret the numerical spectrum of low lying singlets [5,6] in terms of Ising spin flips. This yields the number of singlets as a function of lattice size: a numerically testable prediction. We also find a branch of weakly dispersive triplets at an energy scale slightly below the Heisenberg exchange, in agreement with numerical studies [6,16]. *For the pyrochlore,* we apply two CORE steps to arrive at an Ising-like model of local singlets on larger supertetrahedra blocks which form a cubic superlattice. At the mean field level, we obtain a singlet ground state which breaks lattice symmetry [see Fig. 3 (below)]. Here, too, the effective Hamiltonian describes Ising-like domain walls. We shall conclude that lattice symmetry breaking and

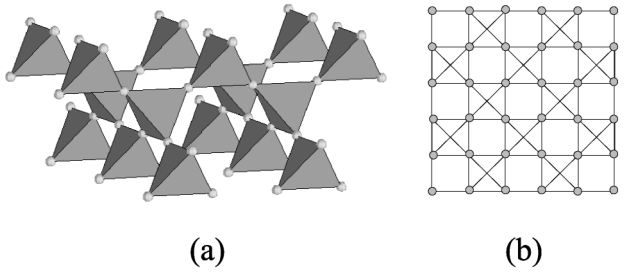

FIG. 1. The pyrochlore (a) and checkerboard (b) lattices.

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local singlet excitations are general features of highly frustrated quantum antiferromagnets.

The checkerboard.—The lattice depicted in Fig. 1(b) contains crossed plaquettes (equivalent to tetrahedra) connected by uncrossed plaquettes. The first step is to choose elementary clusters which cover the lattice. Two different options for plaquettes are the *crossed* and the *uncrossed plaquettes*. While the two clustering choices may appear to yield different ground states and excitations, we shall see that they are in fact consistent with each other, and yield complementary information.

Clustering with uncrossed plaquettes.—From the spectrum of a single uncrossed plaquette, we retain the singlet ground state as a vacuum state $|\Omega\rangle_i$ and the lowest triplet as a hard core boson $t^\dagger_{\alpha i}|\Omega\rangle_i$. $\alpha = x, y, z$ is a Cartesian index of the triplet.

The effective Hamiltonian in the uncrossed plaquettes basis is (in units of $J = 1$)

$$\mathcal{H}_{\rm eff} = \epsilon'_t \sum_i t^\dagger_{\alpha i} t_{\alpha i} + K \sum_{\langle i\,j\rangle} \mathbf{L}_i \cdot \mathbf{L}_j + \sum_{\langle i\,j\rangle \alpha\beta} \{-\Delta t^\dagger_{\alpha i} t^\dagger_{\alpha j} t_{\beta i} t_{\beta j} + b t^\dagger_{\alpha i} t_{\alpha i} t^\dagger_{\beta j} t_{\beta j}\}, \quad (1)$$

where $\mathbf{L}_j = \sum_{\alpha\beta} t^\dagger_{\alpha j} \vec{\mathcal{L}}_{\alpha\beta} t_{\beta j}$ and $\vec{\mathcal{L}}_{\alpha\beta}$ are 3×3 spin-1 matrices in a Cartesian basis.

The parameters calculated by CORE up to range 2 are $\epsilon'_t = 0.5940$, $K = 0.2985$, $\Delta = 0.1656$, and $b = 0.0776$. The truncation errors from up to range 4 are less than 2%, and will be ignored [17].

Note that $\mathcal{H}_{\rm eff}$ in (1) commutes with the number of triplets since it has no anomalous pair creation terms, as appear, e.g., for the square lattice [15]. Thus, at this level of truncation, the plaquette vacua product $|\Psi_0\rangle = \prod_i |\Omega_i\rangle$, *is an exact ground state of the effective Hamiltonian* (1). This result agrees with Moessner *et al.* [8], who argued for a plaquettized singlet ground state based on an effective quantum dimer model, and with Fouet *et al.* [6], who proposed this state based on a numerical study.

We are also able to obtain the triplet gap for, since $t^\dagger_{\alpha,i}|0\rangle$ is an approximate eigenstate of (1), its energy (spin gap) can be read from $\epsilon'_t = 0.5940$. This compares well with the value of 0.6–0.7 estimated by exact diagonalizations of finite systems [6]. We have found very weak hopping terms (of magnitude $0.01J$) due to CORE interactions of range 4, which will give the triplets a weak dispersion in the full lattice.

Clustering with crossed plaquettes.—The isolated crossed plaquette has twofold degenerate singlet ground states, which we can represent by a pseudospin-$\frac{1}{2}$ doublet. The quantization axis for the pseudospin operators is chosen as in Ref. [13], with the $+z$ ($-z$) directions representing states with positive (negative) chirality. The planar angles 0, $\pi/3$, and $2\pi/3$ represent the three (nonorthogonal) dimer configurations of the tetrahedron.

The effective Hamiltonian in the crossed plaquettes basis is an Ising-like model:

$$H_{\rm eff} = -J_I \sum_{\langle ij\rangle} (\mathbf{S}_i \cdot \mathbf{e}_{ij})(\mathbf{S}_j \cdot \mathbf{e}_{ij}) - h \sum_i S^x_i. \quad (2)$$

where $\mathbf{e}_{ij}$ are directors on the x-y plane pointing $\pi/3$ ($-\pi/3$) away from the x axis for horizontal (vertical) bonds. At range 2, we obtain $J_I = 0.5277$, and $h = 0.2362$. Corrections from range-3 and range-4 CORE were computed [17] and found to be unimportant for the symmetry and correlations of the low excitations.

We solve (2) in mean field theory. The energy exhibits two minima, where the pseudospins describe, to a good approximation, vertical or horizontal dimers. This is consistent with the result of the uncrossed plaquette scheme. Indeed, the two columnar states correspond, respectively, to projections of the two equivalent plaquette ground states onto the truncated Hilbert space of the crossed plaquettes. While the ground state energy is not as well converged at range 2 as that of the uncrossed plaquettes, (2) treats the two symmetry breaking ground states in an unbiased fashion. It therefore provides a simple description of the low energy singlet excitations as pseudospin flips. The ordering transition is governed by dynamics of Ising-like domain walls between the two ground states.

The energy gap and dispersion of the low energy singlets is calculated using a spinwave expansion about the mean field ground states of (2). We find a gap $\Delta_0 \approx J_I$ for single spin flips in (2) and bandwidth $W \approx 0.78 J_I$. Figure 2 demonstrates the relation between a pseudospin-flip and 2-triplet bound states in the uncrossed basis. Recent results from series expansions [16] indicate that 4-magnon bound states may be found below the 2-magnon. To see this will require computing CORE interactions beyond the simple range-2 model (2).

Thus, we see that the two methods elucidate complementary aspects of the checkerboard. The approach using uncrossed plaquettes gives a very accurate description of

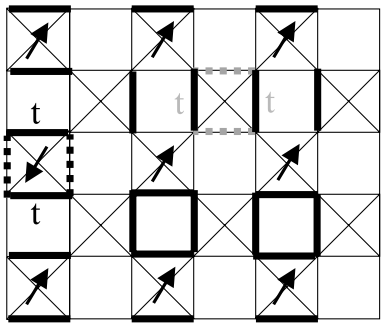

FIG. 2. A low energy singlet excitation which is a bound state of two vertical triplets. This state is represented by a single pseudospin flip in the crossed plaquette approach. In contrast, the horizontal triplet pair shown in gray cannot be described by a simple spin flip in this crossed plaquettes basis.

one ground state, which serves as its vacuum, and correctly counts the 2-magnon bound states above it. However, the second ground state (the vacuum of the alternative uncrossed plaquette clustering) is a complicated multimagnon bound state in this representation. The crossed plaquettes on the other hand, provide an unbiased treatment of the two symmetry breaking ground states, but can capture only half of the 2-magnon bound states in terms of simple pseudospin flips (see Fig. 2).

The number of singlet states below the triplet gap is expected to grow as *power laws* with the size of the system N. For example, the lowest lying single spin flips grow as N, while higher spin flip pairs grow as N^2, and so on. An Ising-like phase transition is expected between the broken and unbroken translational symmetry phases at a temperature $T_c \simeq 0.3J_I \simeq 0.15J$, with an associated logarithmic divergence of the heat capacity at T_c.

The pyrochlore.—Depicted in Fig. 1(a) is a three-dimensional network of corner sharing tetrahedra. Like the checkerboard, it has a macroscopically degenerate (exponential in lattice size) classical ground state manifold. For the quantum $S = 1/2$ case, local pseudospins can be defined by the degenerate singlets of disjoint tetrahedra. These cover all sites of the pyrochlore and form an fcc superlattice. The effective Hamiltonian on this fcc lattice was calculated by CORE. The first nontrivial intertetrahedra coupling are obtained at range-3 connected tetrahedra, which yield

$$H_{\text{eff}}^{(3)} = \sum_{\langle ijk \rangle} \Big[J_2 (\mathbf{S}_i \cdot \mathbf{e}_{ijk}^{(i)})(\mathbf{S}_j \cdot \mathbf{e}_{ijk}^{(j)}) - J_3 \Big(\frac{1}{2} - \mathbf{S}_i \cdot \mathbf{e}_{ijk}^{(i)}\Big) \times \Big(\frac{1}{2} - \mathbf{S}_j \cdot \mathbf{e}_{ijk}^{(j)}\Big)\Big(\frac{1}{2} - \mathbf{S}_k \cdot \mathbf{e}_{ijk}^{(k)}\Big)\Big]. \quad (3)$$

The coupling parameters (in units of J) are $J_2 = 0.1049$, $J_3 = 0.4215$, and $\mathbf{e}_{123}^{(i)}$; $i = 1$, 2, and 3 are three unit vectors in the x-y plane whose angles $\alpha_{123}^{(i)}$ depend on the particular plane defined by the triangle of tetrahedral units 123 as given in Table I of [13]. The effective Hamiltonian (3) resembles the terms obtained by second order perturbation theory (in intertetrahedra couplings) [12,13]. The classical mean field ground state of (3) is identical to that found in Refs. [12,13]: Three of the four fcc sublattices are ordered in the directions $\mathbf{e}(0)$, $\mathbf{e}(2\pi/3)$, $\mathbf{e}(-2\pi/3)$, while the direction of the fourth is completely degenerate. Therefore, classical mean field approximation for (3) is insufficient to remove the ground state degeneracy. Tsunetsugu [13] was able to lift the degeneracy by including spinwave fluctuation effects which produce ordering at a new low energy scale.

Here we avoid the *a priori* symmetry breaking needed for semiclassical spinwave theory, by treating (3) fully quantum mechanically. This entails a second CORE transformation which involves choosing the "*supertetrahedron*" as a basic cluster of four tetrahedra.

TABLE I. The values of the vectors $\mathbf{f}_{ij}$ in Eq. (4), depending on the vector $\mathbf{r}_{ij}$ separating the sites i and j.

$\mathbf{r}_{ij}$	$\mathbf{f}_{ij}$
$(\pm 1, 0, 0)$	
	(1,0,0)
$(0, \pm 1, \pm 1)$	
$(0, \pm 1, 0)$	
	$(-\frac{1}{2}, \frac{\sqrt{3}}{2}, 0)$
$(\pm 1, 0, \pm 1)$	
$(0, 0, \pm 1)$	
	$(-\frac{1}{2}, -\frac{\sqrt{3}}{2}, 0)$
$(\pm 1, \pm 1, 0)$	

Our new pseudospins $\boldsymbol{\tau}_i$ are defined by the two degenerate singlet ground states of the supertetrahedron. [This degeneracy is found for the Heisenberg model on the original lattice as well as for the effective model (3).] These states transform as the E irreducible representation of the tetrahedron (T_d) symmetry group, similar to the singlet ground states of a single tetrahedron.

The supertetrahedra form a cubic lattice, shown in Fig. 3. The effective Hamiltonian (3) and the lattice geometry imply that nontrivial effective interactions appear only at the range of three supertetrahedra and higher. Range-3 effective interactions include two and three pseudospin interactions, which are dominated by

$$\mathcal{H}_{\text{eff}} = J_1 \sum_{\langle ij \rangle} (\boldsymbol{\tau}_i \cdot \mathbf{f}_{ij})(\boldsymbol{\tau}_j \cdot \mathbf{f}_{ij}) + J_2^{(a)} \sum_{\langle\langle ij \rangle\rangle} (\boldsymbol{\tau}_i \cdot \mathbf{f}_{ij})(\boldsymbol{\tau}_j \cdot \mathbf{f}_{ij}) + J_2^{(b)} \sum_{\langle\langle ij \rangle\rangle} [\boldsymbol{\tau}_i \cdot (\mathbf{f}_{ij} \times \hat{\mathbf{z}})][\boldsymbol{\tau}_j \cdot (\mathbf{f}_{ij} \times \hat{\mathbf{z}})]. \quad (4)$$

Here, $\langle\,\rangle$ and $\langle\langle\,\rangle\rangle$ indicate summation over nearest- and next-nearest neighbors, respectively. The coupling constants are found to be relatively small: $J_1 = 0.048J$, $J_2^{(a)} = -0.006J$, and $J_2^{(b)} = 0.018J$. The vectors $\mathbf{f}_{ij}$

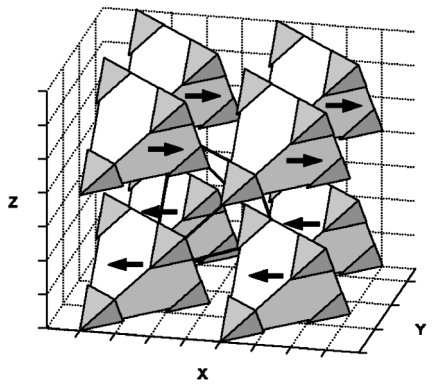

FIG. 3. The pyrochlore viewed as a cubic lattice of supertetrahedra. The arrows show the direction of the supertetrahedra's pseudospin in the mean field ground state.

depend on the vector $\mathbf{r}_{ij}$ connecting the two sites, and their values are presented in Table I.

We performed classical Monte Carlo simulations using the classical (large spin) approximation to (4). The ground state was found to choose an antiferromagnetic axis, and to be ferromagnetic in the planes as depicted in Fig. 3. It differs from the semiclassical ground state [12,13]. The latter involves condensation of high energy states of the supertetrahedron in the thermodynamic ground state. Since on a supertetrahedra we find a much larger gap to these states than intersite coupling, we believe they cannot condense to yield the semiclassical ground state symmetry breaking.

To estimate the truncation error, we calculated the contribution of range-4 interactions in both stages of CORE leading to (3) and (4). Evidently, these terms are small ($< 30\%$) and, most importantly, including them does not alter the mean field solution.

Discussion.—The CORE technique enabled us to derive an effective Hamiltonian for highly frustrated antiferromagnets, written in terms of low energy, local degrees of freedom. For both checkerboard and pyrochlore systems, we found lattice symmetry breaking ground states which are essentially products of local singlets. The spin gap to the lowest triplet excitation is large (of order J), and seems to survive interplaquette interactions. The low energy excitations are singlets, which are local pseudospin flips. The ordering energy scale is of order $J/100$ for the pyrochlore.

This picture seems to be consistent with existing numerical data for the density of low energy singlets on the checkerboard [6]. The pyrochlore lattice is currently not amenable to exact diagonalization. However, it would be instructive to test our results against large scale numerical diagonalization of the fcc effective Hamiltonian (3). Experimentally, lattice symmetry breaking could drive a static lattice distortion, which would be observable by additional Bragg peaks in neutron and x-ray scattering. For example, the antiferromagnetic order between planes of supertetrahedra would correspond to a lattice distortion with wavelength of four tetrahedra.

It is interesting to compare our results with another popular candidate for the ground state. In a recent paper, Lee *et al.* [18] studied the spin-3/2 pyrochlore $ZnCr_2O_4$, by means of neutron scattering. They reported results which are consistent with singlets residing on disjoint hexagons. It may be speculated that resonating singlets on disjoint hexagons are also a good candidate ground state for the spin-1/2 case. Hence, we compare the variational energy of the spin-1/2 Heisenberg Hamiltonian in the hexagon state and in a state with disjoint supertetrahedra in their singlet ground states. The result, $E_{\mathrm{var}} = -0.415$/spin for the hexagons and $E_{\mathrm{var}} = -0.444$/spin for the supertetrahedra, is a strong support for the ground state we propose in the spin-1/2 case. Of course, the additional correlations we find between supertetrahedra would further reduce the variational energy of this state.

How general are these results?—Formation of local singlets is a natural way to relieve the frustration in quantum antiferromagnets that can be written as a sum over clusters $\sum_c (\sum_i \mathbf{S}_{ic})^2$. On each even cluster, the ground state is a singlet with a gap to a local triplet. Frustration suppresses hopping of these triplets and could inhibit their condensation into a spin ordered ground state. Thus, lattice symmetry breaking singlet ground states are expected as a typical feature of frustrated quantum antiferromagnets [19].

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