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LETTER TO THE EDITOR

Parity-broken ground state for the spin-1 pyrochlore antiferromagnet

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Abstract

The ground-state properties of the spin-1 pyrochlore antiferromagnet are studied by applying the VBS-like tetrahedron-unit decomposition to the original spin system. The symmetrization required on every vertex is taken into account by introducing a ferromagnetic coupling. The pairwise effective Hamiltonian between the adjacent tetrahedrons is obtained by considering the next nearest neighbour and the third neighbour exchange interactions. We find that the transverse component of the spin chirality exhibits a long-range order, breaking the parity symmetry of the tetrahedral group, while the chirality itself is not broken.

The pyrochlore lattice, the network of the corner sharing tetrahedrons or a fcc-array of tetrahedrons, is a typical example of a three-dimensional (3D) frustrated system which is found in a number of materials, such as spinels, pyrochlores and C15-type Laves phase. To study effects of the geometric frustrations and the resultant enhanced spin fluctuations, the pyrochlore spin systems have been investigated intensively. For the spin-1/2 antiferromagnetic (AF) Heisenberg model, the quantum spin-liquid ground state is proposed based on the series expansion at finite temperature [1], but concerning the properties of the low-lying spin-singlet states below the spin gap a consistent picture has not emerged yet [2–5].

For the spin-1 case, in order to obtain definitive results [4] for the spin-1/2 systems powerful numerical methods and an analytical approach are used. We proposed the idea of the tetrahedron-unit decomposition of the pyrochlore lattice, which is a natural generalization of the valence-bond-solid(VBS) state approach developed for the 1D spin-1 systems [7]. In this approach the ground states of the fundamental unit, corresponding to the valence bond for the VBS case is the tetrahedron spin singlets which form the two-dimensional E representation of the tetrahedral group (T_d). In consequence, the constructed VBS-type wavefunctions define the variational spin-singlets manifold with a macroscopic degeneracy, to which the ground state of the original problem may be continued adiabatically. In this situation, it is essential to

investigate how the ground state in the thermodynamic limit is stabilized from a collection of spin singlets by lifting the degeneracy.

In a previous paper [7], we examined the magneto-elastic couplings between the tetrahedron-singlets and the local lattice distortion of the E modes on the same tetrahedron as the source of lifting the degeneracy. As a result we have found that a Jahn-Teller mechanism driven by the non-magnetic spin degrees of freedom gives rise to the structural phase transition. The local lattice distortion is given by the Q_v mode of T_d. Actually the uniform Q_v distortion compressed along the c-axis is consistent with the cubic-to-tetragonal structural phase transition without any accompanying magnetic order observed in the spin-1 spinel-type antiferromagnets, ZnV_2O_4 and MgV_2O_4. This scenario can explain, at least qualitatively, the experimental results around the structural transition temperature.

In this letter we investigate another way of lifting the degeneracy when the inter-tetrahedron interactions, such as the next nearest and/or third neighbour spin–spin interactions, are relevant. In particular, we will discuss the properties of the order parameters of the obtained ground state. Since the tetrahedron-singlets may be labelled by the spin chirality, a chiral ordered state is an interesting possibility. However, as long as physically reasonable interactions, such as spin exchange interaction or the dipole interaction are considered, we find that it is not the chirality but its transverse component that shows the long-range order.

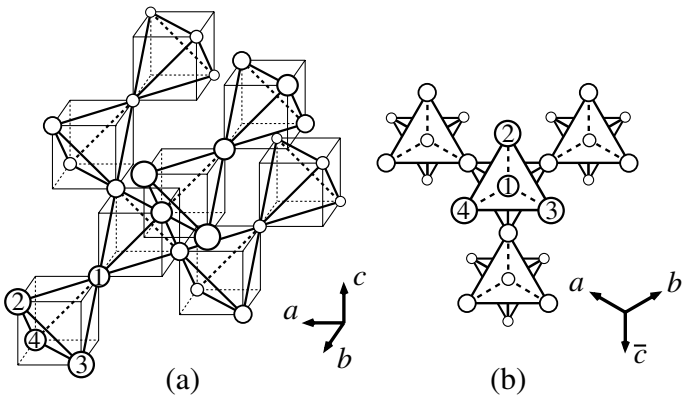

Figure 1. The primitive cell of the pyrochlore lattice (a) and its projected view from the $[11\bar{1}]$-axis (b). The small cubes in part (a) are a guide for the eye with one-sixteenth volume of the unit cell.

Following the 1D VBS approach, we break up the original spin-1 objects (denoted by $\vec{S}_i$) into two spin-1/2 ones ($\vec{s}_{ia}$ and $\vec{s}_{ib}$) and introduce ferromagnetic Hund couplings between them ($-J_F$), which serve as the symmetrization. To project out the singlet sector of the composite spins completely it is necessary to take the $J_F \to \infty$ limit, however, in nature a strong but finite J_F is sufficient. The pyrochlore lattice may be decomposed into A and B type bonds, which form the upward and downward tetrahedrons as shown in figure 1(b). Because of the symmetrization we can consider that the 1/2-spins with a (b) index construct the network of the A (B) bonds without loss of generality. In this decomposite-spin representation, the original AF Heisenberg model can be rewritten in the strong ferromagnetic coupling limit ($J_F \to \infty$) as follows

$$\mathcal{H}_{dec} = 4J \sum_{\langle i,j\rangle \in A} \vec{s}_{ia} \cdot \vec{s}_{ja} + 4J \sum_{\langle i,j\rangle \in B} \vec{s}_{ib} \cdot \vec{s}_{jb} - J_F \sum_i \vec{s}_{ia} \cdot \vec{s}_{ib}. \tag{1}$$

In the $J_F = 0$ limit, the ground states of equation (1) are given by;

$$|\Psi_0\rangle = \prod_{k=1}^{N/2} (\alpha_k |u\rangle_k + \beta_k |v\rangle_k) \tag{2}$$

with arbitrary complex parameters α_k and β_k keeping $|\alpha_k|^2 + |\beta_k|^2 = 1$, where N is the number of sites and k specifies a tetrahedron. Here the orthonormal basis, $\{|u\rangle, |v\rangle\}$ defined for a single tetrahedron are the total spin-singlet states with $\vec{s}_{1x} + \vec{s}_{2x} = \vec{s}_{3x} + \vec{s}_{4x} = 0$ or 1, respectively, $(x = a, b)$ (see figure 2). These tetrahedron singlets are the non-magnetic doublets belonging to the E representation of T_d and the $2^{N/2}$-fold degenerate manifold defined by equation (2) is expected to be adiabatically continued to the low-energy sector of the original model. When representing the symmetry operation of T_d in this singlet subspace, $\{|u\rangle, |v\rangle\}$ real-basis diagonalizes the parity operations with respect to the bonds vertical to the c-axis. On the other hand, the chirality basis [7], which is defined by $|R\rangle = (|u\rangle - i|v\rangle)/\sqrt{2}$ and $|L\rangle = |R\rangle^*$, diagonalizes the four distinct C_3 operations with the eigenvalues of $\omega = (-1 + \sqrt{3}i)/2$ and its complex conjugate ω^*, respectively.

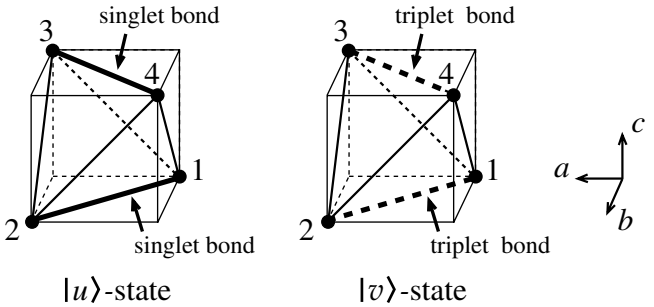

Figure 2. The schematic representations of the tetrahedron-singlet states, $|u\rangle$ and $|v\rangle$.

We proceed to the next step of lifting the $2^{N/2}$-fold degeneracy of equation (2). When considering the pure two-tetrahedron problem, we can easily see that the ferromagnetic coupling J_F alone does not lift the degeneracy. Therefore the pairwise perturbation between the two adjacent tetrahedrons caused by J_F never fixes the original 2×2-fold degeneracy. From a geometry viewpoint, this means that the relative rotation of two tetrahedrons is not fixed only by J_F. The degeneracy is partly lifted by considering three tetrahedrons and eventually the degeneracy will be lifted for the entire lattice. However energy scales of the lifting of degeneracy are expected to be small and hierarchical [5]. This problem is interesting but more academic since in nature there are other couplings to lift the degeneracy. In [7] we have considered the local coupling with lattice distortion which leads to the structural phase transition. As another source of lifting the degeneracy we can introduce longer-range interactions which are relevant to lift the degeneracy for a pair of tetrahedrons.

In order to fix the relative rotation of the two adjacent tetrahedrons, we include the next nearest neighbour (J_1) and the third neighbour interactions (J_2). In spinels, J_2 is expected to be important because of the existence of a superexchange-path through the oxygens on a single plane. The arrangement of these interactions are shown in figure 3. By the second-order perturbation in $-J_F$, J_1, and J_2 within the degenerate subspace spanned by equation (2), we obtain the pairwise effective Hamiltonian between the tetrahedron-singlet states on the

adjacent tetrahedrons as follows

$$\mathcal{H}_{eff} = \frac{1}{2}\left(J_2 - J_1\right)\left(J_F - 4J_2\right) \sum_{\langle k_a, k_b\rangle} \left(\tau_{k_a}^{+}\tau_{k_b}^{-} + \tau_{k_a}^{-}\tau_{k_b}^{+}\right) + C_1 \tag{3}$$

where the coupling is the same for the four distinct directions of pairs. In equation (3),

$$C_1 = -\frac{N}{64}\left\{ \left(J_F - 4J_2\right)^2 + 2\left(J_F - 4J_1\right)^2 + 10\left(4J_2 - 4J_1\right)^2 \right\} \tag{4}$$

and the Pauli matrix $\vec{\tau}_k$, describing the two-dimensional spin-singlet space on the kth tetrahedron, is defined by using the chirality basis as,

$$\tau^z = |R\rangle\langle R| - |L\rangle\langle L|$$
$$\tau^+ = |R\rangle\langle L| \qquad \tau^- = |L\rangle\langle R|.$$

In fact, we can show that the universality class of the effective Hamiltonian is XXZ type by using a symmetry consideration as follows. First, the $\pm 2\pi/3$ rotation, C_3 and C_3^{-1}, around the axis joining the centres of two tetrahedrons leads to the conservation of their total chirality. Second, the $|R\rangle$ and $|L\rangle$ states are mutually conjugate by the time-reversal symmetry. Therefore we can conclude that the derived effective interaction has the XXZ-type symmetry of the pseudo $\vec{\tau}$-spins and that the Ising term must vanish in the even-order perturbations. It should be noted that the second-order perturbation gives the leading terms, since we start from the non-magnetic zeroth order states.

In order to estimate the higher-order perturbation terms concerning J_F within the pairwise treatment, we consider the infinite J_F limit by considering the original spin-1 object on the sharing top vertex of the two tetrahedrons. In this case, the effective Hamiltonian obtained by the first-order perturbation is given by,

$$h_{k_a,k_b} = \frac{1}{2.25}\left(J_2 - J_1\right)\left(\tau_{k_a}^{+}\tau_{k_b}^{-} + \tau_{k_a}^{-}\tau_{k_b}^{+}\right) + C_2 \tag{5}$$

with $C_2 = \left(J_2 + 2J_1\right)/9$, which is just $8/9$ of the coefficient of the first order term of equation (3) in J_F. The $\vec{\tau}$ matrixes here are defined concerning the bottom triangles, which are related to the $\vec{\tau}$ matrix in equation (3) through the relations like $|R\rangle = \left(|\uparrow_R\rangle|\downarrow_V\rangle - |\downarrow_R\rangle|\uparrow_V\rangle\right)/\sqrt{2}$, where $|\uparrow_R\rangle$ is the doublet state with R chirality about the bottom triangle and $|\uparrow_V\rangle$ represents the spin state on the top vertex. It should be noted that such decompositions cannot be extended consistently over the whole lattice. Here we have shown it to illustrate the generic form of the pairwise effective interaction in the strong J_F limit.

Comparison of equation (3) and equation (5) suggests that higher-order effects of J_F only renormalize the strength of the pairwise interaction. Therefore, we assume that the effective Hamiltonian which describes the low-energy part of the original Heisenberg model with next nearest neighbour (J_1) and third neighbour interactions (J_2) is given as follows, by using the spin-$1/2$ pseudospin operator, $\vec{\tau}_k$, defined for the kth tetrahedron

$$\mathcal{H}_{eff} = cJ' \sum_{\langle k_a, k_b\rangle} \left(\tau_{k_a}^{+}\tau_{k_b}^{-} + \tau_{k_a}^{-}\tau_{k_b}^{+}\right) \tag{6}$$

where $J' = J_2 - J_1$ with a positive c. Since both of the upward (labelled by k_a) and downward (k_b)tetrahedrons form the distinct fcc lattice structure, (6) is the spin-$1/2$ XY model on ZnS type bipartite lattice with coordination number $z = 4$.

Since the sign of the coupling constant of the XY model on a bipartite lattice can be converted by the π-rotation around the z-axis concerning one of the two sublattices in the following we discuss the case with the negative interaction. For the spin-$1/2$ case with N sites, it is shown that the ground state is unique with $S_{tot}^z = 0$ by using the Perron-Frobenius

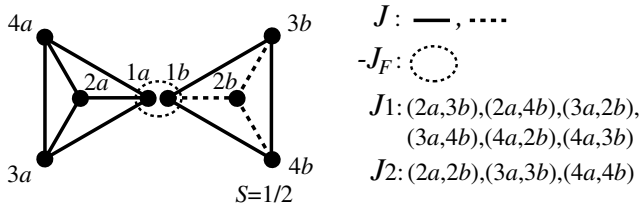

Figure 3. The decomposite-spin representation of the Heisenberg model (J and infinite J_F) and the next-nearest (J_1) and third-neighboring interactions (J_2) are shown for the pairwise (k_a, k_b) tetrahedrons.

theorem [10]. As one can naively imagine, the state with $S_{tot} = N/2$ and $S^z_{tot} = 0$ is a good zeroth-order approximation. In fact, the Gutzwiller-type wavefunction constructed from this state is shown to be an extremely good variational state [11, 12]. When applying their results for our $z = 4$ case, the square of the long-range order (LRO) and the short-range magnetic correlation are given by;

$$\langle M^{x2}\rangle/N^2 = 2^{15}\cdot 3^4\cdot 7^{-8} = 0.4604\cdots \tag{7}$$

$$\langle \tau^z_k \cdot \tau^z_{k+\delta}\rangle \;=\; -1/7 \tag{8}$$

where $\vec{M} = \sum_{k=1}^{N}\vec{\tau}_k$ and $\vec{\tau}_{k+\delta}$ means the nearest neighbour pseudospin next to the $\vec{\tau}_k$ pseudospin. In the XY model, the Hamiltonian has the U(1) symmetry about the rotation around the z axis in the spin space. Therefore the direction of the order parameter in the τ^x-τ^y plane cannot be determined even for the infinite system. The U(1) symmetric general order parameters, $\vec{M}(\theta)$, are defined by the ground-state expectation value of the following operator;

$$\hat{M}\,(\theta) = \left(M^x\cos\theta,\, M^y\sin\theta,\, 0\right) \tag{9}$$

where $\langle M^x\rangle = \langle M^y\rangle = M > 0$. Concerning every local tetrahedron, $\hat{M}(\theta)$ belongs to the E representation of T_d.

Although the effective Hamiltonian, equation (6), has the continuous U(1) symmetry as represented by equation (9), the original model possesses only the finite group (T_d) symmetry. The U(1) symmetry is fictitious, valid only in the lowest-order treatments with the pseudo-spin representation. Suppose that we take account of the higher-order interactions like biquadratic term and others, then this spurious U(1) symmetry should be broken. In order to understand how this lowering of the symmetry acts, we make use of the Ginzburg–Landau type argument by expanding the free energy as a function of the order parameter $\vec{M}(\theta)$, by its magnitude M and phase θ around the second-order transition point. The U(1) symmetry, obtained by the perturbation, corresponds to the second-order expansion of the free energy by $\vec{M}(\theta)$. This is because, on every tetrahedron, the product representation $E \times E$ $(= A_1 + A_2 + E)$ contains the unique totally symmetric one (A_1), which is a constant independent of θ. Now we proceed to higher order corrections. Since under the time-reversal symmetry only τ^z changes the sign, while both τ^x and τ^y remain unchanged, the third-order expansion gives the next leading term, introducing the anisotropy to fix θ. By calculating the third order invariant, which is obtained by reducing the product representation $E \times E \times E$, we find that the anisotropy is proportional to $\cos 3\theta$. Thus we get a set of the stable states, $\{\vec{M}(\pi), \vec{M}(\pm\pi/3)\}$ or $\{\vec{M}(0), \vec{M}(\pm 2\pi/3)\}$, depending on the sign of the M^3 term. These two sets only differ in the overall sign and are essentially the same.

Let us illustrate the symmetry property of the order parameter by taking $\vec{M}(0) = \sum_{k=1}^{N}\langle\tau^x_k\rangle = \sum_{k=1}^{N}\langle|u_k\rangle\langle u_k| - |v_k\rangle\langle v_k|\rangle$ as an example. The non-vanishing long-range

correlation between τ_k^x and $\tau_{k'}^x$ results in the different population of the $|u\rangle$ and $|v\rangle$ states on a tetrahedron. Therefore the parity symmetry concerning the bonds vertical to the c-axis is broken in the ground state, since the $|u\rangle$ and $|v\rangle$ states are characterized by the odd and even parity concerning these bonds respectively. Three different values of θ in each set of order parameters correspond to the three equivalent choices of the cubic principal axis; $\vec{M}(2\pi/3)$ and $\vec{M}(-2\pi/3)$ characterizes the break-down of the parity symmetry concerning the bonds vertical to the b and c axes, respectively. These three states form a domain structure in a real system.

Now we extend the present results for positive J_F and also summarize the ground-state properties of equation (6) by taking c-axis as a principal axis ($\theta = 0, \pi$). Depending on the sign of $J' = J_2 - J_1$, the uniform ($J' < 0$) or the staggered signed ($J' > 0$) summation of the $\langle\tau_k^x\rangle$ over all the tetrahedrons defines the order parameter. In the classical picture, for negative J' the ground state shows the ferromagnetic LRO of the transverse components of the spin chirality $|u\rangle$ ($|v\rangle$) when $\theta = 0$ (π), while for positive J' the antiferromagnetic LRO of the $|u\rangle$ and $|v\rangle$ states. For a local tetrahedron problem the compressed and elongated Q_v mode along the c-axis stabilize the $|u\rangle$ and $|v\rangle$ states, respectively [7]. Therefore, for the ferromagnetic J' model the order parameter, $\vec{M}(0)$ ($\vec{M}(\pi)$), induces the uniform Q_v lattice distortion with $c/a < 1$ (> 1). On the other hand, for the AF J' the staggered Q_v distortion is induced. Accordingly, under the assumption that the inter-tetrahedron interactions are more important than the local magneto-elastic couplings, the inter-tetrahedron couplings (J_1 and J_2) lift the many-fold degeneracy of the spin-singlet manifold by the second-order phase transition. However, introduction of the magneto-elastic coupling, even for a weak case, will induce a small first order structural distortion, leading eventually to a weak first-order phase transition.

In conclusion, we have studied the ground-state properties of the spin-1 pyrochlore antiferromagnet by using the tetrahedron-unit decomposition of the pyrochlore lattice. In this approach, we assume that the spin-singlet manifold spanned by the VBS-type wavefunction well describes the low-energy physics well and the essential point is how the degeneracy is lifted. We have investigated the case where this degeneracy is lifted by the inter-tetrahedron interactions caused by the next nearest neighbour and the third neighbour interaction, which produce the pairwise effective Hamiltonian of the XY type between the pseudospin operators describing the tetrahedron singlets. To break the spurious U(1) symmetry of the effective model, we have considered the higher-order anisotropic term based on the symmetry property (E) of the order parameter. It has turned out that the parity-broken ground state emerges through the second-order phase transition.

Lastly we briefly comment on the relation between the present results and the numerical results for the pure spin-1 Heisenberg model [4]. The singlet ground states of a single spin-1 tetrahedron problem are three-dimensional with $A_1 + E$ irreducible representations. When we define these spin-singlet states, with $\vec{S}_1 + \vec{S}_2 = \vec{S}_3 + \vec{S}_4 = 0$, 1, and 2, by $|a\rangle$, $|b\rangle$, and $|c\rangle$, respectively, the orthonormal basis of the E representation are given by $\{|U\rangle, |V\rangle\} = \{|b\rangle, (-2|a\rangle + \sqrt{5}|c\rangle)/3\}$. Considering the fact that the symmetrization on every vertex does not affect the local symmetry properties, the parity-broken symmetry of the ground state, obtained by using the decomposite-spin representation, may correspond to the $|U\rangle$ and $|V\rangle$ states in the spin-1 picture. Since Koga *et. al* suggested the possibility of the new spin-gap ground state characterized by the $|U\rangle$ state around the isotropic Heisenberg limit [4], our result seems to be consistent with their result.

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