Frustrated Heisenberg antiferromagnet on the pyrochlore lattice

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We investigate quantum phase transitions for the $s=\frac{1}{2}$ antiferromagnetic Heisenberg model on the pyrochlore lattice. By means of the series expansion method starting from isolated tetrahedra, the ground-state phase diagram is determined. When the ratio of the two competing exchange couplings is varied, a first-order (second-order) quantum phase transition occurs between two spin gap phases (the spin-gap and the antiferromagnetic phases). We also discuss some properties expected for the $s=1$ pyrochlore spin system.

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I. INTRODUCTION

Geometrically frustrated magnetic materials have been the subject of considerable interest recently. In particular, compounds with pyrochlore structure, i.e., a tetrahedral network of magnetic ions, show a wide variety of different phenomena caused by strong frustration. For instance, for typical Ising-type pyrochlore magnets $Ho_2Ti_2O_7$ (Ref. 1) and $Dy_2Ti_2O_7$,[2] an anomalous peak structure in the specific heat was observed at low temperatures. This anomaly, called the ''spin ice'' behavior, is caused by the macroscopically large ground-sate degeneracy induced by geometrical frustration. On the other hand, the metallic compound LiV_2O_4 (Ref. 3) exhibits heavy-fermion behavior, such as the exceptionally large specific-heat coefficient at low temperatures. It was claimed that frustration caused by a tetrahedral network of vanadium ions is important to understand the heavy-fermion behavior in this compound.[4] Such frustration effects should be more important for $Y(Sc)Mn_2$,[5] for which the quantum spin-liquid behavior induced by frustration was indeed observed experimentally. Magnetic property of this compound may be described by the $s=\frac{1}{2}$ antiferromagnetic Heisenberg model on the pyrochlore lattice. Furthermore, the possibility of a ''topological spin glass'' was pointed out for $Y_2Mo_2O_7$,[6] making this class of frustrated quantum spin systems more attractive and challenging.

Theoretical studies[7–9] of the $s=\frac{1}{2}$ quantum spin model on the pyrochlore lattice were first done by Harris *et al.*,[7] who pointed out the possibility of the dimerized ground state by exploiting a field theoretical approach. Canals and Lacroix[8] clarified that the ground state of the model is a spin-liquid state with a spin gap. They found that the neutron diffraction data of $Y(Sc)Mn_2$ (Ref. 5) are in fairly good agreement with their results.[8] However, by using a bond-operator approach, Isoda and Mori[9] suggested that the ground state may be described by a RVB-like (plaquette) singlet state, which is different from the dimer-singlet state known so far.

In this paper, we investigate the $s=\frac{1}{2}$ quantum spin model on the pyrochlore lattice with competing antiferromagnetic interactions shown in Fig. 1(a). Our system may describe pyrochlore-lattice compounds such as $Y(Sc)Mn_2$ (Ref. 5) as well as $GeCu_2O_4$ (Ref. 10) found recently. In Sec. II, we study the ground-state phase diagram to discuss the role of geometrical frustration. For this purpose, we investigate quantum phase transitions by means of series expansion techniques.[11] It is shown that when the ratio of the competing exchange couplings is varied, the system undergoes a first-order (second-order) quantum phase transition between the two spin gap phases (the spin-gap and the antiferromagnetic phases). In Sec. III, we also discuss the $s=1$ case briefly. A summary is given in Sec. IV.

II. QUANTUM PHASE TRANSITIONS

Let us first consider the $s=\frac{1}{2}$ spin model on the pyrochlore lattice, which is described by the following Hamiltonian:

$$H=J\sum_{(i,j)}\mathbf{S}_i\cdot\mathbf{S}_j+J'\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j, \tag{1}$$

where (i,j) denotes a pair of two adjacent sites connected by the thick bond in Fig. 1(a), whereas $\langle i,j\rangle$ those connected by the thin bond. Both of the exchange couplings J and J' are assumed to be antiferromagnetic. For the compound $Y(Sc)Mn_2$, we may take $J/J'\sim 1$,[5] while for $GeCu_2O_4$, $J/J'\sim 6$.[10]

Before proceeding with the analysis, we note that, in some limiting cases, the spin system is reduced to the simple models whose properties can be well understood. For $J'=0$, it is equivalent to the $s=\frac{1}{2}$ massless Heisenberg spin

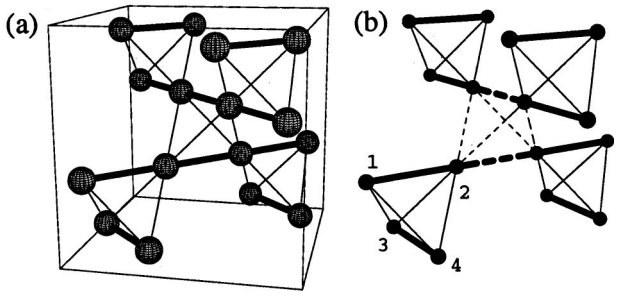

FIG. 1. (a) Frustrated antiferromagnetic spin model on the pyrochlore lattice. Thick and thin bonds represent the exchange couplings J and J', respectively. (b) Initial configuration for the series expansion: Broken lines represent the bonds with λJ and $\lambda J'$ (see text).

TABLE I. Eigenvalues of the $s=1/2$ spin system on an isolated tetrahedron.

S_{12}	0	1	0	1	1	1
S_{34}	0	0	1	1	1	1
S_{total}	0	1	1	0	1	2
E/J'	$-\frac{3}{2}j$	$-\frac{1}{2}j$	$-\frac{1}{2}j$	$-2+\frac{1}{2}j$	$-1+\frac{1}{2}j$	$1+\frac{1}{2}j$

chain, while for $J=0$ an antiferromagnetically ordered state is stabilized since the model has a three-dimensional structure without frustration in this case. To determine the phase diagram, we study what kind of quantum phase transition occurs when the competing interactions are varied. For this purpose, we use the series expansion method,[11] which has the advantage of dealing with frustrated spin systems in higher dimensions. In fact, it was successfully applied to frustrated spin systems such as the J_1-J_2 model,[12] the plaquette system,[13] and the orthogonal-dimer system.[14] To apply the series expansion method to the pyrochlore-lattice system, we first divide the original Hamiltonian Eq. (1) into two parts as $H=H_0+\lambda H_1$ by introducing an auxiliary parameter λ, where $H_0(H_1)$ represents the unperturbative (perturbative) Hamiltonian. The system is reduced to the original model at $\lambda=1$. We here choose a tetrahedron composed of four spins as a starting configuration (H_0),[7,8] and then connect the tetrahedra via the bonds labeled by λJ and $\lambda J'$ [see Fig. 1(b)]. The Hamiltonian h for an isolated tetrahedron in H_0 is given by

$$h=J(\mathbf{S}_1\cdot\mathbf{S}_2+\mathbf{S}_3\cdot\mathbf{S}_4)+J'(\mathbf{S}_1+\mathbf{S}_2)\cdot(\mathbf{S}_3+\mathbf{S}_4). \qquad (2)$$

The energy eigenvalues E of the tetrahedron for a given $j=J/J'$ are listed in Table I, where $S_{12}(S_{34})$ represents the combined spin $S_1+S_2(S_3+S_4)$, and S_{total} the total spin. It is seen in this table that for $0<j<1$, the isolated tetrahedron has a plaquette-singlet ground state with $S_{12}=S_{34}=1$ and $S_{\text{total}}=0$. On the other hand, for $j>1$ we have the dimer ground state with $S_{12}=S_{34}=S_{\text{total}}=0$. The phases specified by these singlets are referred to as the plaquette and dimer phases, respectively.

A. First-order transition

Keeping the above properties in mind, we now discuss how the plaquette and the dimer states compete with each other when the intertetrahedron couplings λJ and $\lambda J'$ are introduced. We calculate the ground state energy up to the sixth order in λ for several values of j. We show the obtained energy in Fig. 2, for which the first-order inhomogeneous differential method[15] is applied to the finite-order series calculated above. As mentioned above, for $\lambda\to 0$, the first-order quantum phase transition occurs between the plaquette and the dimer phases at the critical point $j_c=1$. It is seen in Fig. 2 that the critical value j_c for the phase transition is not sensitive to the change in λ. In fact, the energy up to the second order in λ is the same for both states near $j=1$, as pointed out by Harris *et al.*[7] For small λ, the first-order transition point is given by $j_c\sim 1-0.021\lambda^3$. Remarkably enough, the first-order transition point is estimated as j_c

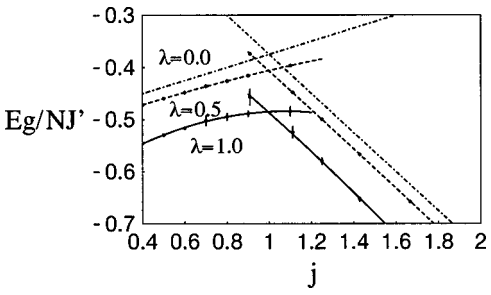

FIG. 2. Ground state energy for the dimer (right-side) and the plaquette (left-side) phases for various values of λ. The energy for $\lambda=0.0$, 0.5, and 1.0 is shown as the dash-dotted, dashed, and solid lines, respectively.

~ 1.0 even for $\lambda=1$. Therefore, we arrive at the interesting conclusion that the isotropic spin system ($\lambda=j=1$) with pyrochlore structure is located quite closely to the phase boundary of the first-order quantum phase transition, although it is difficult to definitely say within our accuracy which phase the ground state actually belongs to. This fact explains the reason why Harris *et al.* and Isoda *et al.* have drawn different conclusions about the nature of the ground state for the same $j=1$ model: the former (latter) claimed that the ground state is a dimer singlet (plaquette singlet). As mentioned above, the energy for two phases is very close to each other, so that the mean-field type treatment may not correctly specify the ground state. Furthermore, there even remains the possibility that the system is just on the boundary, and thus the ground state could be degenerate at $j=1$. In any case, it is instructive to notice that unusual dual properties reflecting both natures of the plaquette and dimer states should appear around $j=1$ in various physical quantities such as the excitation spectrum, etc.

B. Second-order transition

The results obtained above do not necessarily imply that the disordered ground state is always realized in the whole range of j. We should study how the disordered phases compete with possible antiferromagnetic phases stabilized by the three-dimensional (3D) exchange couplings. We first recall that for $j=0$ and $\lambda=1$, the system should have an antiferromagnetic order, as mentioned before. On the other hand, in the case $j\to\infty(J'\to 0)$ the spin system is reduced to the $s=\frac{1}{2}$ massless Heisenberg chain characterized as a Tomonaga-Luttinger liquid phase.[16] In the parameter regime $(j>1)$, a different type of the magnetic order was observed experimentally for $GeCu_2O_4$ $(j\sim 6)$.[10] Therefore, we have to carefully check whether the above two different antiferromagnetic orders are indeed stabilized in our model.

We first study the magnetically ordered phase in the region $0<j<1$. To this end, we compute the staggered susceptibility and the triplet-excitation energy up to the fourth order in λ for various values of j. To observe the second-order transition to the magnetically ordered phase, we study the spin gap at $\mathbf{k}=0$ in the Brillouin zone, which should vanish at the phase transition point. By applying Padé approximants to the computed series,[15] we obtain the spin gap and the

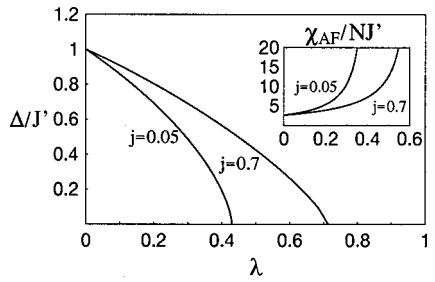

FIG. 3. The energy gap $\Delta = E(0,0,0)$ (inset: the staggered susceptibility χ_{AF}) as a function of λ with $j = 0.05$ and $j = 0.7$ obtained by the Dlog $[\frac{1}{2}]$ Padé approximants.[15]

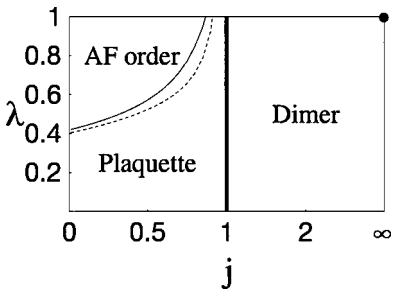

FIG. 4. Phase diagram for the $s = \frac{1}{2}$ quantum Heisenberg model with pyrochlore structure. The bold line represents the phase boundary which separates the dimer and the plaquette phases. A solid (dashed) line indicates the phase boundary between the plaquette and the magnetically ordered phases, which is determined by the spin gap (staggered susceptibility). The filled circle represents the location of the isolated $s = \frac{1}{2}$ spin chain.

staggered susceptibility as a function of λ (Fig. 3). When $\lambda = 0$, the system is reduced to an assembly of isolated tetrahedra with the spin gap $\Delta = J'$. With increasing λ, the 3D network develops, enhancing the antiferromagnetic correlation. Therefore, the spin gap decreases and the staggered susceptibility increases, and finally the second-order quantum phase transition occurs to the magnetically ordered phase (e.g., the critical value is given by $\lambda_c \sim 0.4$ for $j = 0.05$). From the critical points obtained for various values of j, we end up with the phase boundaries shown in Fig. 4. The increase of j suppresses the magnetic correlation due to strong frustration, and thus the system favors the plaquette phase. The critical value is estimated as $j_c \sim 0.9$ for $\lambda = 1$. As seen in this figure, the boundaries determined in two distinct ways slightly differ from each other, which may be due to the lower-order (fourth) series expansion done here. Although it is desirable to perform a higher-order calculation to determine the phase boundary more precisely, the essential features inherent in the $s = \frac{1}{2}$ pyrochlore system may be given by the present calculation; e.g., the magnetic phase is not stabilized at $j = 1$, but the phase boundary between the spin-gap phase and the magnetic phase is located rather closely to $j = 1$.

We next examine another possibility of the antiferromagnetic order observed for $GeCu_2O_4$ (Ref. 10) in the region $j > 1$. For this purpose, we calculate the susceptibility for the corresponding staggered field up to the third order in λ. As a result, we find that the divergence in the susceptibility around $\lambda = 1$ is suppressed, as j decreases from the value ($j = \infty$) for the isolated spin chain. This tendency implies that in the region $j > 1$, the system does not stabilize the antiferromagnetically ordered phase, but always stays in the dimer phase with spin gap. Therefore, we conclude from the above analysis that the magnetic order observed for the compound $GeCu_2O_4$ (Ref. 10) ($j \sim 6$) may not be explained simply in terms of the isotropic Heisenberg model employed here. This in turn suggests that the anisotropic interactions may be taken into account in order to discuss the ordered state in this compound.

III. $S = 1$ PYROCHLORE SYSTEM

Let us now turn to the $s = 1$ system on the pyrochlore lattice.[17,18] Although the series-expansion calculation becomes much more difficult in this case, we can still deduce some instructive comments on the $s = 1$ pyrochlore lattice. In order to use series expansion techniques, we again start with a configuration of isolated tetrahedra. The energy eigenvalues in this case are listed in Table II. It is seen that the $s = 1$ plaquette singlet with $S_1 + S_2 = S_3 + S_4 = 2$ and $S_{\mathrm{total}} = 0$ is the ground state for $0 < j < 1$, while the dimer singlet state with $S_1 + S_2 = S_3 + S_4 = 0$ and $S_{\mathrm{total}} = 0$ is the ground state for $j > 1$. In contrast to the $s = \frac{1}{2}$ model, an isolated tetrahedron in the isotropic point ($j = 1$) has threefold degenerate ground states, which include the abovementioned singlet states together with another singlet state with $S_1 + S_2 = S_3 + S_4 = 1$ and $S_{\mathrm{total}} = 0$, which may be regarded as a $s = \frac{1}{2}$ plaquette-singlet state (see Table I). By introducing the intertetrahedron coupling, we observe how the above threefold singlet states are changed. To investigate the first-order quantum phase transitions among three phases, we estimate the ground state energy up to the fourth order in λ. This expansion shows that, in contrast to the $s = \frac{1}{2}$ case, there exists an intermediate ($s = \frac{1}{2}$ plaquette) phase between the $s = 1$ plaquette phase and the dimer phase for small values of λ: two phase boundaries are estimated as $j_c = 1 - 0.42\lambda^3$ and $j_c = 1 + 0.084\lambda^3$, which are shown as the thick-dashed lines in Fig. 5. Although we do not have a definite answer to the

TABLE II. Eigenvalues of an isolated $s = 1$ tetrahedron: $E_D(E_P)$ is the energy for the first (second) part in Eq. (2).

S_{12}	0	0	0	1	1	1	1	1	1	1	2	2	2	2	2	2	2	2	2
S_{34}	0	1	2	0	1	1	1	2	2	2	0	1	1	1	2	2	2	2	2
S_{total}	0	1	2	0	0	1	2	1	2	3	2	1	2	3	0	1	2	3	4
E_D/J	−4	−3	−1	−3	−2	−2	−2	0	0	0	−1	0	0	0	2	2	2	2	2
E_P/J'	0	0	0	0	−2	−1	1	−3	−1	2	0	−3	−1	2	−6	−5	−3	0	4

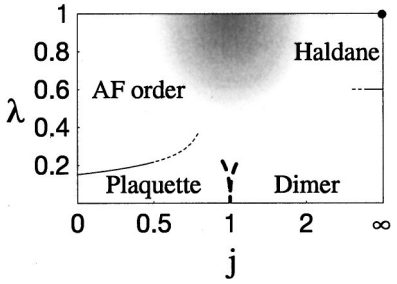

FIG. 5. Schematic phase diagram for the $s=1$ antiferromagnetic Heisenberg model with pyrochlore structure. The filled circle represents the location of the isolated $s=1$ spin chain.

question whether this intermediate state can be the ground state even at $\lambda=1$, it may compete with the other states at the fully frustrated point $j=1$ with $\lambda=1$. We next calculate the spin gap to see how stable the magnetically ordered phase for the $s=1$ case is in comparison with the $s=\frac{1}{2}$ case. By applying the Padé approximants to the third-order results,[15] we obtain the phase boundary shown as the solid line in Fig. 5. It is seen that the area of the magnetically ordered phase is more extensive in comparison with the $s=\frac{1}{2}$ case. Thus it is expected that the magnetically ordered phase for the $s=1$ case may be more dominant around the isotropic point $j=1$. Finally, we make a brief comment on the small J' (large j) case. For $J'=0$, the system is reduced to the $s=1$ spin chain with bond alternation, where the dimer phase and the Haldane phase are separated at the critical point $\lambda_c=0.6$.[19] Although it is difficult to estimate the phase boundary between these spin-gap states in the presence of the interchain coupling, it is naively expected that the Haldane phase may disappear when j decreases down to $j=1$.

IV. SUMMARY

In conclusion, we have discussed the ground-state phase diagram for the $s=\frac{1}{2}$ Heisenberg model with pyrochlore structure by means of the series expansion method. In particular, it has been found that the two different spin-gap states strongly compete with each other around $j=1$, where the compound $Y(Sc)Mn_2$ (Ref. 5) may be located. Also, the antiferromagnetic phase has been shown to be extended rather closely to the phase boundary. Concerning $GeCu_2O_4$,[10] for which $j\sim 6$, it has been found that the present model may not describe its magnetic order, suggesting that some other mechanism should be considered for the magnetism. Although we have not been able to reach a definite conclusion about the phase diagram for the $s=1$ system, we have checked that the magnetically ordered phase may be more dominant around $j=1$ in comparison with the $s=\frac{1}{2}$ case. Also, in addition to the known states such as dimer, plaquette, and magnetically ordered states, another intermediate spin-gap state may also be a candidate for the ground state around $j=1$. Since this argument is based on the calculation for small λ, it is desired to confirm whether this spin-gap state really plays an important role around $j=1$, which is now under consideration.

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