Frustrated Heisenberg antiferromagnet on a pyrochlore lattice

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

Geometrically frustrated magnetic materials have been the subject of considerable interest recently. A typical example is a class of transition-metal oxides with pyrochlore structure. In particular, it was reported that the specific-heat coefficient is exceptionally large for a metallic compound LiV$_2$O$_4$. It was recently pointed out that the frustration caused by a tetrahedral network of vanadium ions may be important to understand the heavy-fermion behavior in this compound, stimulating further intensive studies on the related systems with pyrochlore structure. Such frustration effects should be much more prominent for quantum spin systems. Theoretical studies for the $s = 1/2$ quantum spin model on a pyrochlore lattice were first done by Harris et al. who pointed out the possibility of the dimerized ground state by exploiting a field theoretical approach. Canals and Lacroix clarified that the ground state of the model is a spin-liquid state with the spin gap. They found that the neutron diffraction cross section observed in Y(Sc)Mn$_2$ is in fairly good agreement with their results. Isoda and Morii, however, used a bond-operator approach to suggest that the ground state may be described by a RVB-like tetrahedral (plaquette) singlet state, which is different from the dimer-singlet state known so far. Furthermore, the possibility of the "topological spin glass" was pointed out in Y$_2$Mo$_2$O$_7$, making this issue more attractive and challenging.

In this paper, we investigate the $s = 1/2$ quantum spin model on a pyrochlore lattice with competing antiferromagnetic interactions shown in Fig. 1(a). Our system may describe some pyrochlore-lattice compounds such as Y(Sc)Mn$_2$ as well as GeCu$_2$O$_4$ found recently. We study the ground-state phase diagram and clarify the role of the geometrical frustration by studying quantum phase transitions by means of series expansion techniques. We also discuss the $s = 1$ case briefly.

Let us first consider the $s = 1/2$ spin model on a pyrochlore lattice, which is described by the following Hamiltonian

$$H = J \sum_{<i,j>} S_i \cdot S_j + J' \sum_{<i,j>} S_i \cdot S_j,$$  \hspace{1cm} (1)

where $(i,j)$ denotes a pair of two adjacent sites connected by the thick bond in Fig. 1(a), whereas $<i,j>$ is that for 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 $J/J' \sim 1$ while for GeCu$_2$O$_4$, it is to be $J/J' \sim 6$.

Before proceeding with the analysis, we note that, in some limiting cases, the spin system is reduced to simple models whose nature can be easily understood. For $J' = 0$, it is equivalent to the $s = 1/2$ massless Heisenberg spin chain, whereas an antiferromagnetically ordered state is stabilized for $J = 0$ 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 which has an advantage to deal with frustrated spin systems in higher dimensions. In fact, it was successfully applied to frustrated spin systems such as $J_1-J_2$ model plaquette system, orthogonal-dimer system. 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 $\lambda$, where $H_0(H_1)$ represents the unperturbed (perturbed) Hamiltonian. Note that the system is reduced to the original model for $\lambda = 1$. We here choose a tetrahedron composed of four spins as a starting configuration $(H_0)$ and then connect each tetrahedron via tetrahedral bonds labeled by $\lambda J$ and $\lambda J'$ [see Fig. 1(b)]. The Hamiltonian $h$ for an isolated tetrahedron in $H_0$ is given

![FIG. 1: (a) Frustrated antiferromagnetic spin model on a pyrochlore lattice. Bold and thin-solid lines represent the perturbed bonds, $\lambda J$ and $\lambda J'$ (see text).](image-url)
by
\[ h = J(S_1 \cdot S_2 + S_3 \cdot S_4) + J'(S_1 + S_2) \cdot (S_3 + S_4) \] (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 \( S_{\text{total}} \) is the total spin. It is seen in this table that for \( 0 < j < 1 \),

|                | \( S_{12} \) | \( S_{34} \) | \( S_{\text{total}} \) | \( E/J' \) |
|----------------|-------------|-------------|-----------------|------------|
|                | 0           | 1           | 0               | \( -\frac{1}{2} \) |
|                | 0           | 0           | 1               | \( -\frac{1}{2} \) |
|                | 0           | 1           | 0               | \( -\frac{1}{2} \) |
|                | 1           | 0           | 1               | \( 0 \) |
|                | 0           | 0           | 1               | \( \frac{1}{2} \) |
|                | 1           | 0           | 2               | \( 1 \) |

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.

Keeping the above properties in mind, we now discuss how the plaquette and the dimer states compete with each other when the inter-tetrahedron couplings \( \lambda J \) and \( \lambda J' \) are introduced. We expand the ground state energy up to the sixth order in \( \lambda \) for several values of \( j \). We show the obtained energy in Fig. 2 for which the first-order inhomogeneous differential method is applied to the bare series. As mentioned above, for \( \lambda \to 0 \), the first-order quantum phase transition occurs between the plaquette and the dimer phase at the critical point \( j_c = 1 \). It is seen in the Fig. 2 that the critical value for the phase transition is little changed even if we increase \( \lambda \). In fact, the energy up to the second order in \( \lambda \) is same for both states near \( j = 1 \), as pointed out by Harris et al. For small \( \lambda \), 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 \( j_c \sim 1.0 \) even for \( \lambda = 1 \), where our generalized model is reduced to the original one. Therefore, we arrive at a quite interesting conclusion that the homogeneous 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 conclude which phase the ground state really belongs to within our accuracy. This fact clarifies the reason why Harris et al. and Ishida et al. had different conclusions on the nature of the ground state for the same \( j = 1 \) model, where 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 emerge around \( j = 1 \) in various physical quantities such as the excitation spectrum, etc.

The results obtained above do not necessarily imply that a disordered ground state is always realized in the whole range of \( j \). It is needed to study how the disordered phases compete with possible antiferromagnetic phases driven 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 \( s = 1/2 \) massless Heisenberg chain characterized by the Tomonaga-Luttinger liquid phase. In the parameter regime \( (j > 1) \), a different type of the magnetic order was observed experimentally for GeCu2O4 \((j \sim 0) \). Therefore, we have to carefully check whether the above 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 susceptibility for a staggered field and the triplet excitation energy up to the fourth order in \( \lambda \) for various values of \( j \). To observe the second-order transition to the magnetically ordered phase, we study the spin gap at \( k = 0 \) in the Brillouin zone, which should vanish at the phase transition point. By applying Padé approximants to computed series, we obtain the phase boundaries shown in Fig. 3. When \( (j, \lambda) = (0, 0) \), the system is reduced to an assembly of isolated tetrahedra. With the increase of \( \lambda \), the 3D network develops, enhancing the antiferromagnetic correlation. At last, 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 \( j = 0 \)). The increase of \( j \) suppresses the magnetic correlation due to strong frustration, and thus favors the plaquette phase. The critical value is estimated as \( j_c \sim 0.9 \) for \( \lambda = 1 \), where our generalized model is reduced to the original system. 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 desired to perform a higher-order calculation to determine the phase boundary more precisely, its essential feature is certainly given by the present calculation; e.g., the magnetic phase is not dominant for \( j = 1 \), but its phase boundary is rather close to \( j = 1 \).

We next examine another possibility of the antiferromagnetic order observed for GeCu_{2}O_{4} in the region \( j > 1 \). For this purpose, we calculate the susceptibility for the corresponding staggered field up to the third order in \( \lambda \). As a result, we find that the divergent singularity around \( \lambda = 1 \) is gradually suppressed, as \( j \) is decreased from the value \( (j = \infty) \) for the isolated spin chain. This tendency implies that in the region \( j > 1 \), the system does not enter the antiferromagnetically ordered phase, but always stays in the dimer phase with spin gap. Therefore, it is seen from the above analysis that the magnetic order observed for the compound GeCu_{2}O_{4} \( (j \sim 0) \) may not be simply explained in terms of the isotropic Heisenberg model employed here. This in turn suggests that anisotropic interactions may be important to realize an ordered state in this compound.

Let us now turn to the \( s = 1 \) system on a pyrochlore lattice. Although the series-expansion calculation becomes 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 an isolated \( s = 1 \) tetrahedron, whose eigenvalues are listed in Table \( \text{I} \). It is seen that the \( s = 1 \) plaquette-singlet with \( S_{1} + S_{2} = S_{3} + S_{4} = 2 \) and \( S_{\text{total}} = 0 \) is the ground state for \( 0 < j < 1 \), whereas the dimer singlet state with \( S_{1} + S_{2} = S_{3} + S_{4} = 0 \) and \( S_{\text{total}} = 0 \) is the ground state for \( j > 1 \). In contrast to the \( s = 1/2 \) model, an isolated tetrahedron in the homogeneous point \( (j = 1) \) has three-fold degenerate ground states, which are composed of the above-mentioned singlet states together with another singlet state with \( S_{1} + S_{2} = S_{3} + S_{4} = 1 \) and \( S_{\text{total}} = 0 \), which may be regarded as a \( s = 1/2 \) plaquette-singlet state (see Table \( \text{II} \)). By turning on the inter-tetrahedron coupling, we observe how the above three-fold singlet states evolve on a pyrochlore lattice. To investigate the first-order quantum phase transitions among three phases, we estimate the ground state energy up to the fourth order in \( \lambda \). This expansion claims that in contrast to the \( s = 1/2 \) case, there exists an intermediate \( (s = 1/2 \) plaquette) phase between the \( s = 1 \) plaquette phase and the dimer phase in small \( \lambda \): 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 bold dashed lines in Fig. 3. Although we do not have a definite answer to the question whether this intermediate state can survive as the ground state even for \( \lambda = 1 \), it may be a possible candidate which competes with other states at 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 \( s = 1/2 \) case. By applying the Padé approximants to the third-order results, we deduce the phase boundary shown as the solid line in Fig. 4. It is seen that the area of the magnetically ordered phase is more extensive than the \( s = 1/2 \) case. By recalling the phase diagram for the \( s = 1/2 \) case, it is thus expected that the magnetically ordered phase may be more dominant around the homogeneous 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 $\lambda_c = 0.6$ \cite{4}. Although it is difficult to estimate the phase boundary between these spin-gap states in the presence of interchain coupling, it is naively expected that the Haldane phase may disappear when $j$ is decreased down to $j = 1$.

In conclusion, we have discussed the ground-state phase diagram for the $s = 1/2$ 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 $\text{Y(Sc)}\text{Mn}_2\text{O}_4$ may be located. Also, the antiferromagnetic phase has been shown to be extended rather closely to the phase boundary. Concerning $\text{GeCu}_2\text{O}_4$\cite{8} for which $j \sim 6$, it has turned out that the present model may not describe its magnetic order, suggesting that some other mechanism should be considered for the magnetism. For the $s = 1$ system, we have not been able to deduce the definite conclusion on the phase diagram, but have checked that the magnetically ordered phase may be more dominant around $j = 1$ in comparison with the $s = 1/2$ case. Also, besides 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 has been based on the calculation for small $\lambda$, it is desired to confirm whether this spin-gap state really takes part in the strong competition around $j = 1$, which is now under consideration.

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