Order induced by dipolar interactions in a geometrically frustrated antiferromagnet

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We study the classical Heisenberg model for spins on a pyrochlore lattice interacting via long-range dipole-dipole forces and nearest-neighbor exchange. Antiferromagnetic exchange alone is known not to induce ordering in this system. We analyze low-temperature order resulting from the combined interactions, both by using a mean-field approach and by examining the energy cost of fluctuations about an ordered state. We discuss behavior as a function of the ratio of the dipolar and exchange interaction strengths and find two types of ordered phase. Below a certain value of this ratio, we find that the system orders in a four-sublattice Néel state. For interaction strengths above this critical ratio, the system orders with an incommensurate wave vector. We relate our results to the recent experimental work and reproduce and extend the theoretical calculations on the pyrochlore compound, $Gd_2Ti_2O_7$, by N. P. Raju, M. Dion, M. J. P. Gingras, T. E. Mason, and J. E. Greedan, Phys. Rev. B **59**, 14 489 (1999).

INTRODUCTION

Geometrically frustrated magnetic systems have received a great deal of attention in recent years from both experimentalists and theorists.[1] Such systems are typically composed of corner- or edge-sharing frustrated units, usually triangles or tetrahedra. For example, a two-dimensional network of corner-sharing triangles forms the *kagomé* lattice, while the three-dimensional pyrochlore structure is composed of corner-sharing tetrahedra. Magnetic systems with such structure exhibit unusual low-temperature properties, which are not completely understood. These materials typically remain disordered to a freezing temperature, T_g, much less than the magnitude of the Curie-Weiss constant $|\theta_{CW}|$.[1,2] Indeed, the depression of the transition temperature below that expected from the measured value of $|\theta_{CW}|$ is often used to gauge the degree of frustration in a magnetic system.

The reluctance of geometrically frustrated antiferromagnets to order can be understood by considering classical models, many examples of which yield macroscopically degenerate ground states. For instance, it has been shown that the Heisenberg model for spins on a pyrochlore lattice interacting via nearest-neighbor antiferromagnetic exchange has an extensive number of degrees of freedom in the ground state.[3–6] The degeneracy of the ground-state manifold is not lifted in this model by thermal fluctuations, and the system has no finite-temperature ordering transition.[5,6] Several realizations of pyrochlore magnets have been studied experimentally.[2,7–9]

Classical models that include nearest-neighbor exchange interactions alone may not be sufficient for explaining all the low-temperature properties of some frustrated antiferromagnets. For instance, the origin of the freezing transition must involve additional aspects of the system. In real materials, several other types of interactions are present. Further neighbor exchange may be relevant[4] as may crystal-field effects,[10] magnetic dipole interactions,[9] or the effect of quenched disorder.[3,11]

In this paper, we focus on how the inclusion of dipolar forces affects the properties of the pyrochlore antiferromagnet. Because of the ground-state degeneracy of this system with only nearest-neighbor exchange, dipolar interactions are important in establishing order even if they are weak. Moreover, the influence of dipole interactions in a pyrochlore antiferromagnet has been probed in the compound $Gd_2Ti_2O_7$,[12] which should be well represented by an isotropic Heisenberg model.

The properties of $Gd_2Ti_2O_7$ have been studied both experimentally and theoretically in a recent paper by Raju *et al.*, in which it was shown that $Gd_2Ti_2O_7$ undergoes a transition to long-range order at a temperature of about 1 K.[12] Their measurement of the high-temperature susceptibility gives a negative Curie-Weiss constant, $\theta_{CW} \simeq -9.8$ K.[12] Measurements on a magnetically dilute sample show a reduction in $|\theta_{CW}|$, indicating that this value is predominantly due to antiferromagnetic exchange.[12] The transition temperature is much lower than $|\theta_{CW}|$, indicating that the system is frustrated. The theoretical work of these authors involves mean-field calculations expressed as a Landau expansion of the free energy and taken to quadratic order. They examine the ordering instabilities that occur as the temperature is lowered. With nearest-neighbor exchange and long-range dipolar interactions, they find that order parameter fluctuations on entire branches in q space along the star of the (111) direction become unstable simultaneously at the mean-field critical temperature T_c.[12] Degeneracies of this kind are often broken by thermal or quantum fluctuations, a phenomenon known as order-by-disorder.[13] Raju *et al.* suggest that this mechanism may operate to induce order in the model they study.[12]

In the following, we extend this mean-field description of the system with only nearest-neighbor exchange and long-range dipolar interactions to find the ordering pattern below T_c. We show that the quartic term in the free-energy expansion lifts the degeneracy of the critical modes. The ordering pattern obtained for the ratio of dipolar to exchange interaction strengths appropriate for $Gd_2Ti_2O_7$ is a four-sublattice Néel state. In addition, we analyze the low-temperature fluctuations away from the four-sublattice ground state. We show that all distortions have a positive-energy cost. Order-

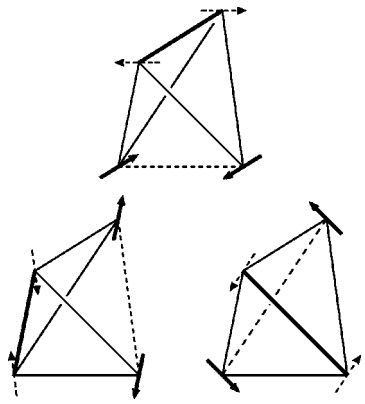

FIG. 1. Three degenerate ground states for spins on a single tetrahedron. The spins in each configuration are parallel to certain edges of the tetrahedron. These edges and the spins parallel to them are drawn with the same type of line, either bold or dashed. There are three other ground states, obtained from these by reversing all spins.

ing in this model, therefore, has an energetic origin and is not an example of fluctuation-induced order.

I. SPINS ON A SINGLE TETRAHEDRON

It is instructive first to consider spins on a single tetrahedron. Labeling the spins by i,j, the interaction energy for the tetrahedron is

$$U_{int}=\frac{J_{ex}}{2}\sum_{i\neq j}\mathbf{S}_i\cdot\mathbf{S}_j+\frac{J_{dd}}{2}\sum_{i\neq j}[\mathbf{S}_i\cdot\mathbf{S}_j-3(\mathbf{S}_i\cdot\hat{\mathbf{r}}_{ij})(\mathbf{S}_j\cdot\hat{\mathbf{r}}_{ij})], \tag{1.1}$$

where J_{ex} and J_{dd} are the relative interaction strengths of the exchange and dipolar terms, $J_{dd}\propto\mu^2/r_{nn}^3$, r_{nn} is the edge length of the tetrahedron (the nearest-neighbor distance between spins), and μ is the dipole moment. We minimize this energy using a standard numerical search and find all the possible ground states. These are shown in Fig. 1. In each of the ground-state configurations, the spins are coplanar and are tangent to the sphere circumscribing the tetrahedron. It is worthwhile to note that these states are also ground states for spins interacting via nearest-neighbor antiferromagnetic exchange only. Without dipolar interactions, the condition for a configuration to be a ground state is that the vector sum of the spins on each tetrahedron is zero, leaving two internal degrees of freedom for the configuration.[4,6] Dipolar interactions fix these two degrees of freedom and those arising from the $O(3)$ symmetry.

Since the interaction energy for a single tetrahedron involves only nearest-neighbor dipolar contributions, one might not expect the spin configurations in Fig. 1 to be a useful guide to behavior on the full pyrochlore lattice. In fact, and somewhat surprisingly, the ground state for the full lattice that we find in Sec. III turns out to be a periodic repetition of that for a single tetrahedron, provided the ratio of dipolar to exchange interactions does not exceed a critical value.

II. MEAN-FIELD THEORY FOR INTERACTIONS ON THE PYROCHLORE LATTICE

We consider a system of spins interacting via the Hamiltonian

$$\mathcal{H}=\frac{1}{2}\sum_{i\neq j,\alpha\beta}S_i^\alpha J_{ij}^{\alpha\beta}S_j^\beta,$$

with

$$J_{ij}^{\alpha\beta}=\delta_{\alpha\beta}\delta_{nn}+\frac{C}{r_{ij}^3}\left(\delta_{\alpha\beta}-\frac{3(r_{ij}^\alpha)(r_{ij}^\beta)}{r_{ij}^2}\right), \tag{2.1}$$

where $C=r_{nn}^3J_{dd}/J_{ex}$. The exchange interaction is between nearest-neighbors only, as indicated by δ_{nn}. The labels i and j refer to sites in the lattice and α and β label components of the spin or spatial vectors. A mean-field treatment for Hamiltonians of this kind has been developed by Reimers, Berlinsky, and Shi.[4] For completeness, we briefly summarize their approach. Order parameters for the system are defined by

$$\mathrm{Tr}(\rho_i\mathbf{S}_i)=\mathbf{B}_i, \tag{2.2}$$

where ρ_i is the local density matrix and is constrained by $\mathrm{Tr}(\rho_i)=1$. Expanding the free energy in a power series in $\mathbf{B}_i$ yields

$$F=\mathrm{const}+\frac{1}{2}\sum_{i\neq j,\alpha\beta}B_i^\alpha(3T\delta_{\alpha\beta}\delta_{ij}+J_{ij}^{\alpha\beta})B_j^\beta$$
$$+\frac{9}{20}T\sum_{i,\alpha\beta}(B_i^\alpha B_i^\alpha)(B_i^\beta B_i^\beta)+O(B^6). \tag{2.3}$$

The next step is to diagonalize the quadratic term. In systems with n atoms per unit cell, it is convenient to divide the site label i into two parts: l labels the unit cell and a labels spins within the unit cell. Making the Fourier transforms

$$B_{la}^\alpha=\sum_{\mathbf{q}}B_{\mathbf{q}}^{a\alpha}e^{i\mathbf{q}\cdot\mathbf{r}_l}, \tag{2.4}$$

$$J_{lmab}^{\alpha\beta}=\frac{1}{N}\sum_{\mathbf{q}}J_{\mathbf{q}}^{a\alpha b\beta}e^{-i\mathbf{q}\cdot\mathbf{r}_{lm}} \tag{2.5}$$

and substituting these expressions into the free energy, one arrives at the expression for the quadratic part of $f=F/N$, the free energy per unit cell, used in Ref. 12:

$$f^{(2)}=\frac{1}{2}\sum_{ab,\alpha\beta}\sum_{\mathbf{q}}B_{\mathbf{q}}^{a\alpha}(3T\delta_{ab}\delta_{\alpha\beta}+J_{\mathbf{q}}^{a\alpha b\beta})B_{-\mathbf{q}}^{b\beta}. \tag{2.6}$$

Consider diagonalization of the $3n\times3n$ matrix, $J_{\mathbf{q}}^{a\alpha b\beta}$. Denote the eigenvalues by $\lambda_{\mathbf{q}}^i$ and eigenvectors by $\mathbf{U}_{\mathbf{q}}^i$ for $i=1,\ldots,3n$. Expanding the order parameters in the basis of eigenvectors

$$B_{\mathbf{q}}^{a\alpha}=\sum_i U_{\mathbf{q}}^{a,\alpha i}\phi_{\mathbf{q}}^i, \tag{2.7}$$

one obtains

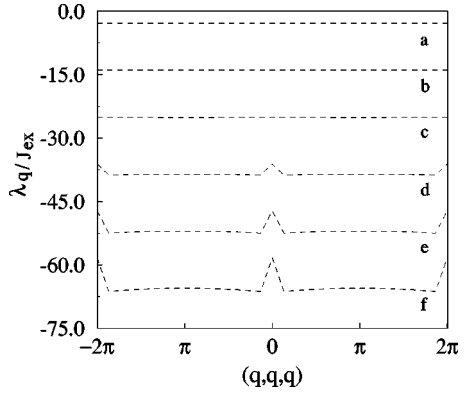

FIG. 2. Minimal eigenvalues $\lambda_{\mathbf{q}}/J_{ex}$ along the (111) line in q space for various values of the dipole-dipole interaction strength. Curve a corresponds to $J_{dd}/J_{ex}=0.2$ as in Ref. 12. Curves b through f have $J_{dd}/J_{ex}=2.92$, 5.63, 8.35, 11.06, and 13.78, respectively, (Ref. 14).

$$f^{(2)}=\frac{1}{2}\sum_{\mathbf{q}}\sum_{i}(3T+\lambda_{\mathbf{q}}^{i})\phi_{\mathbf{q}}^{i}\phi_{-\mathbf{q}}^{i}. \tag{2.8}$$

To find the minimum of the quadratic term, it is necessary to determine the minimum $\lambda_{\mathbf{q}}^{i}$ for all i and $\mathbf{q}$.

Specializing to the pyrochlore lattice, we follow previous workers in using a rhombohedral unit cell containing four magnetic ions $(3n=12)$.[4,12] Primitive basis vectors are $(a/2)(\hat{j}+\hat{k})$, $(a/2)(\hat{\imath}+\hat{k})$, and $(a/2)(\hat{\imath}+\hat{j})$, where a is the edge length of the cubic unit cell. Also, the four magnetic ions within the cubic unit cell are located at $(x,y,z)=(0,0,0)$, $(0,\frac{1}{4},\frac{1}{4})$, $(\frac{1}{4},0,\frac{1}{4})$, and $(\frac{1}{4},\frac{1}{4},0)$.

The instability of the paramagnetic phase is analyzed by considering the sign of $(3T+\lambda_{\mathbf{q}}^{i})$. In the paramagnetic phase, all are positive. At the mean-field critical temperature, the smallest becomes negative. Using the ratio $J_{dd}/J_{ex}=0.2$, Raju *et al.* showed that such an approach does not completely determine the ordering pattern.[12] At T_c, a star of modes simultaneously becomes unstable. For the same value of J_{dd}/J_{ex}, we recover these results. By contrast, for values of $J_{dd}/J_{ex}>R_c\simeq 5.7$, we find that a discrete set of isolated modes (related by the lattice symmetry) become unstable.

To illustrate this point, we plot in Fig. 2 the minimum eigenvalue of the $J_{\mathbf{q}}$ matrix as a function of $\mathbf{q}$ for various values of J_{dd}/J_{ex} ranging from that used by Raju *et al.*[12] (based on the measured values of $|\theta_{CW}|$ and the lattice spacing and the calculated value of the magnetic moment of Gd^{3+}) to a much larger value. For $J_{dd}/J_{ex}<R_c$, the minimum eigenvalue is independent of $\mathbf{q}$ along the (111) direction. For $J_{dd}/J_{ex}>R_c$, there are isolated minima located close to (but not at) $\mathbf{q}=\mathbf{0}$. These are the individual incommensurate modes that become unstable at T_c in the case $J_{dd}/J_{ex}>R_c$. From an examination of the eigenvectors associated with the minimum eigenvalues of the $J_{\mathbf{q}}$ matrix at these points, we determine that the ordering is not coplanar.

We next investigate the ordering pattern below T_c for $J_{dd}/J_{ex}<R_c$. The degeneracy of the soft modes is lifted when we include contributions to the free energy from the fourth-order term. At temperatures below T_c, the order parameters acquire finite magnitude and one is faced with the problem of simultaneously minimizing the quadratic and quartic parts of the free energy. Consider first the quartic term in isolation. In terms of the real space order parameters, B_{la}^{α}, this is

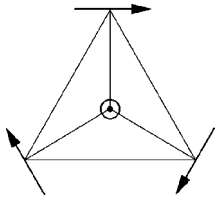

FIG. 3. An illustration of the eigenvectors for the soft modes along the star of the (111) directions in $\mathbf{q}$ space. The vector $\mathbf{q}$ is perpendicular to the base of the tetrahedron and is shown coming out of the page. The three spins are coplanar, have equal magnitude, and are tangent to the circle that circumscribes the base of the tetrahedron. The spin on the fourth site has zero average value.

$$F^{(4)}=\frac{9T}{20}\sum_{la,\alpha\beta}B_{la}^{\alpha}B_{la}^{\alpha}B_{la}^{\beta}B_{la}^{\beta}. \tag{2.9}$$

Its value depends on the magnitude and direction of $\mathbf{B}_{la}$. For fixed magnitude of $\Sigma_{la\alpha}(B_{la}^{\alpha})^2$, the quartic term is minimized by a state with all $|\mathbf{B}_{la}|$ equal. Fortunately, and apparently fortuitously, a state satisfying this condition can be constructed that also minimizes the quadratic term. In detail, we proceed as follows. The eigenvector associated with one of the modes that becomes unstable at T_c is illustrated in Fig. 3. Taking the most general combination of this eigenvector and its three partners, and imposing the condition that the values of $|\mathbf{B}_{la}|$ are the same at four sites of a tetrahedron, we generate the configurations of Fig. 1 and no others. Tiling the lattice with these configurations, we obtain only states with ordering wave vector $\mathbf{q}=\mathbf{0}$. We conclude that the ordering pattern for T just below T_c is as shown in Fig. 4.

III. STATIC DISTORTIONS AWAY FROM THE ORDERED STATE

Finally, we investigate whether a ground state with this ordering pattern is stable against thermal fluctuations at low-temperatures. To describe this calculation using the notation introduced above, we impose the constraint $|\mathbf{B}_{la}|=1$ for every l and a. We denote the components of $\mathbf{B}_{la}$ in the ground

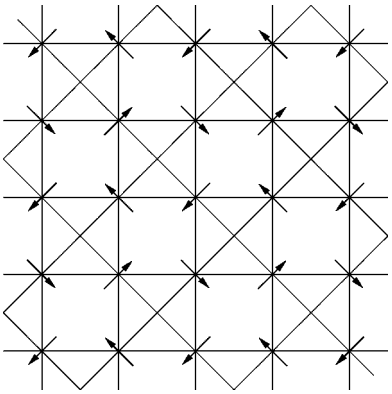

FIG. 4. A projection of the $\mathbf{q}=\mathbf{0}$ ordering pattern found for $J_{dd}/J_{ex}<R_c$ onto the xy plane of the cubic lattice. The spins are coplanar and form a four-sublattice Néel state.

state with the ordering pattern shown in Fig. 4 by $B^0_{la\alpha}$. We expand

$$B_{la\alpha}=B^0_{la\alpha}+\delta B_{la\alpha}-\frac{1}{2}B^0_{la\alpha}|\delta\mathbf{B}_{la}|^2+O(\delta\mathbf{B}^4). \quad (3.1)$$

Since $\delta\mathbf{B}_{la}$ (when small) is orthogonal to $\mathbf{B}_{la}$, it has only two independent components.

We expand the energy to quadratic order in the distortion variables obtaining

$$\mathcal{H}=E_0+\frac{1}{2}\sum_{lmab,\alpha\beta}(J^{\alpha\beta}_{lmab})[\,\delta B_{la\alpha}\delta B_{mb\beta}-\tfrac{1}{2}(|\delta\mathbf{B}_{la}|^2 + |\delta\mathbf{B}_{mb}|^2)\hat{B}^0_{la\alpha}\hat{B}^0_{mb\beta}], \quad (3.2)$$

where E_0 is the energy of the ground state. We then diagonalize the quadratic term, which involves an 8×8 matrix for each wave vector, **q**. We find the minimum eigenvalues $\gamma_{\mathbf{q}}$ of this matrix as a function of **q**. As expected, we recover a flat branch of zero-energy modes when $J_{dd}=0$.[4] For $J_{dd}>0$, we find that all fluctuations away from an ordered state are associated with a positive energy cost (even in the long-wavelength limit, since dipolar interactions break global rotational symmetry). As an illustration, we plot the minimum at each **q** along the (001) direction in q space for various values of the dipole-dipole interaction in Fig. 5. The energetic barriers around our proposed ground state at $T=0$ mean that there is not a degenerate, connected manifold. The ordering obtained is due to energetic selection and does not occur via an order-by-disorder mechanism.

One failure of the mean-field calculation presented in Sec. II is that it predicts a finite ordering temperature even for the case $J_{dd}=0$, where it is known that the system remains disordered to zero temperature.[5,6] In the face of this difficulty, we can arrive at a crude estimate of the ordering temperature from the size of the gap to static distortions at $T=0$. Using estimated values for J_{ex} and J_{dd} for $Gd_2Ti_2O_7$ from Raju *et al.*,[12] we obtain $T_c\sim 1.0$ K. The remarkable agreement of this result with the actual ordering temperature obtained experimentally, 0.97 K,[12] is presumably partly coincidental.

IV. SUMMARY

We have considered the influence of dipolar interactions in a nearest-neighbor Heisenberg model of a geometrically frustrated system, reproducing and extending earlier work by Raju *et al.*[12] In particular, we determine the ordering pattern, using a mean-field treatment and by examining the stability of an ordered state to fluctuations. We believe that the ordering we find for $J_{dd}/J_{ex}<R_c$ should be that associated with the transition that is observed experimentally at around 1 K in $Gd_2Ti_2O_7$.[12] This ordered state, shown in Fig. 4, is a four-sublattice Néel state. For systems in which the value of the ratio of dipolar to exchange interactions is above the critical ratio R_c, we expect that the ordering will occur with an incommensurate wave vector.

It will be interesting to compare these theoretical predictions with the results of neutron scattering from $^{160}Gd_2Ti_2O_7$, currently in progress.[15]

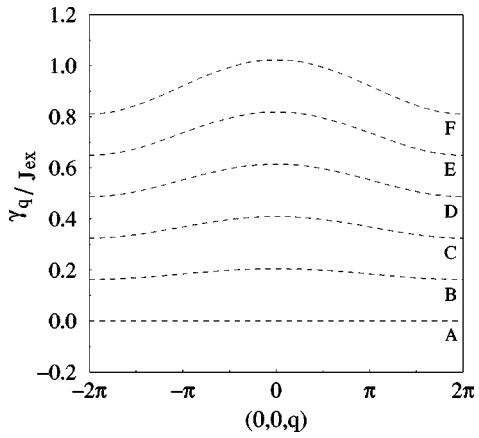

FIG. 5. The minimum eigenvalues, $\gamma_{\mathbf{q}}/J_{ex}$ for distortions away from the ground state in the (001) direction in q space. The various curves correspond to different values of the relative strength of the dipole-dipole interaction J_{dd}/J_{ex}. Curves A through E have $J_{dd}/J_{ex}=0.0$, 0.04, 0.08, 0.12, and 0.16, respectively. Curve F has $J_{dd}/J_{ex}=0.2$ as in Ref. 12.

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