

Degeneracy and strong fluctuation-induced first-order phase transition in the dipolar pyrochlore antiferromagnet

O. Cépas,¹ A. P. Young,² and B. S. Shastry²

¹Institut Laue Langevin, Boîte Postale 156, F-38042 Grenoble Cedex 9, France

²Department of Physics, University of California, Santa Cruz, California 95064, USA

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We show that a continuous set of degenerate critical soft modes strongly enhances the first-order character of a fluctuation-induced first-order transition in the pyrochlore dipolar Heisenberg antiferromagnet. Such a degeneracy seems essential to explain the strong first-order transition recently observed in $\text{Gd}_2\text{Sn}_2\text{O}_7$. We present some evidence from Monte Carlo simulations and a perturbative renormalization group expansion.

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I. INTRODUCTION

In this paper we study phase transitions in Heisenberg magnets on the pyrochlore lattice, which consists of corner sharing tetrahedra.^{1,2} The motivation is partly theoretical, to understand the behavior of highly frustrated systems when there is a degeneracy, or near degeneracy, between different ordered states. There is also experimental motivation since experiments on $\text{Gd}_2\text{Sn}_2\text{O}_7$ and $\text{Gd}_2\text{Ti}_2\text{O}_7$ have shown a rich behavior,^{3,4} including multiple field transitions^{5,6} which we would like to understand.

For the family of rare earth pyrochlore systems it is well known³ that dipole-dipole interactions are important, since the angular momentum is large ($S=7/2$ for Gd). If one adds nearest neighbor exchange to dipole-dipole interactions, the Fourier transform of the total interaction $J(\mathbf{q})$ is virtually independent⁷ of \mathbf{q} (and takes its minimum value) along the (1,1,1) directions of the reciprocal space.^{3,8} This means that the magnetic ordering wave vector could, potentially, lie anywhere along these lines.

It turns out that the phases at the endpoints, $\mathbf{q}=\mathbf{0}$ (denoted, following Ref. 10, by *A*) and $\mathbf{q}=(\pi, \pi, \pi)$ (denoted by $\boldsymbol{\pi}$ or *B*) are particularly important. The ordering expected at $\mathbf{q}=\mathbf{0}$ (*A* type) has been discussed by several authors^{3,5,8,10} and is shown in Fig. 1. A possible ordering at $\mathbf{q}=\boldsymbol{\pi}$ (*B* type) has been proposed in Ref. 10.

Although one would imagine that $\text{Gd}_2\text{Sn}_2\text{O}_7$ and $\text{Gd}_2\text{Ti}_2\text{O}_7$ should be quite similar, since the crystal structures are the same (apart from a very small difference in the lattice constant), it is found that $\text{Gd}_2\text{Sn}_2\text{O}_7$ has a *strong* first-order transition,⁴ while $\text{Gd}_2\text{Ti}_2\text{O}_7$ has a second-order transition.¹² Furthermore, $\text{Gd}_2\text{Ti}_2\text{O}_7$ orders at $\mathbf{q}=\boldsymbol{\pi}$ ^{12,13} while $\text{Gd}_2\text{Sn}_2\text{O}_7$ appears more compatible with the *A* phase.¹⁴ While the small change in lattice parameter between $\text{Gd}_2\text{Sn}_2\text{O}_7$ and $\text{Gd}_2\text{Ti}_2\text{O}_7$ could change somewhat the exchange constants, it seems remarkable that the nature of the ordering changes so dramatically. We would like to understand such a delicate dependence of ordering on exchange constants.

The first-order nature of the transition observed in $\text{Gd}_2\text{Sn}_2\text{O}_7$ is at variance with mean-field theory which predicts¹⁰ a second-order transition. In order to clarify the order of the transition, and to see whether it is affected by longer range exchange interactions which lift the degeneracy

of $J(\mathbf{q})$, we have performed Monte Carlo simulations of the classical dipolar Heisenberg model on the pyrochlore lattice. These show that the apparent order of the transition is indeed very sensitive to the exchange constants.

We have supplemented the numerics by a perturbative renormalization group (RG) analysis. It is known that “fluctuation-induced first-order transitions” occur when there is no stable fixed point in a perturbative RG calculation. This frequently occurs when the number of components of the order parameter n is larger than 4.^{15–18} In general, such transitions are expected to be only *weakly* first order. Another microscopic scenario for a first-order transition is the proposal of Brazovskii¹⁹ that the existence of a continuous set of degenerate soft modes could change the order of the transition. It was later shown that the RG analysis of models with soft modes along special directions lacks stable fixed points and the models are indeed likely to undergo first-order transitions.^{20,21} This scenario is, for instance, relevant to the description of the liquid crystal transition where the degeneracy naturally comes from the isotropy of the liquid.²⁰ It is also particularly relevant in frustrated magnets where frustra-

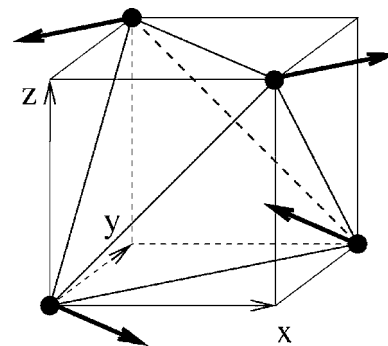


FIG. 1. The *A* state ($\mathbf{q}=\mathbf{0}$) of the pyrochlore lattice stabilized at low temperatures by the dipole-dipole interactions (Refs. 3, 5, 8, and 10). Since the ordering is at $\mathbf{q}=\mathbf{0}$, all tetrahedra have the same spin configuration as the one shown. In the figure, all the spins lie onto the (xy) plane and form pairs of antiparallel spins that are parallel to the opposite edge of the tetrahedron they belong to. There are equivalent (xz) and (yz) states. The magnetic order is therefore characterized by a $n=3$ -component order parameter, ψ with (xy) corresponding to $\psi=(1,0,0)$ at $T=0$.

tion may, precisely, provide a large number of quasidegenerate soft modes, though with different geometrical structures. We shall consider a similar RG analysis, applicable for the symmetry of the dipolar pyrochlores which have lines of degeneracy, in this paper.

In this work, we study the order of the transition of the classical Heisenberg model on the pyrochlore lattice with long-range dipole-dipole interactions and exchange interactions. We show, by means of Monte Carlo simulations, that the transition is *strongly* first order when a continuous set of soft modes is present in $J(\mathbf{q})$, and becomes *weakly* first order when that degeneracy is removed by including further neighbor interactions (Sec. II). In Sec. III, we present a Landau-Ginzburg-Wilson simplified model and a perturbative renormalization group analysis of the transition that predicts a first-order transition, in agreement with the numerical results in Sec. II. We summarize our conclusions in Sec. IV.

II. MONTE CARLO SIMULATIONS

We present in this section some results of Monte Carlo simulations, using the parallel tempering approach,²² on the Heisenberg model with dipolar and exchange interactions on the pyrochlore lattice.

The classical Heisenberg Hamiltonian is

$$\mathcal{H} = \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + (g\mu_B)^2 \sum_{\langle i,j \rangle} \left(\frac{\mathbf{S}_i \cdot \mathbf{S}_j}{r_{ij}^3} - 3 \frac{(\mathbf{r}_{ij} \cdot \mathbf{S}_i)(\mathbf{r}_{ij} \cdot \mathbf{S}_j)}{r_{ij}^5} \right), \quad (1)$$

where \mathbf{S}_i is a classical spin vector of length $S=7/2$ (for Gd^{3+}) on site i , and J_{ij} is the Heisenberg exchange between the neighbors; we will consider the first (J), second (J_2), and third neighbor (J_3) couplings.¹⁰

The number of spins is $N=16L^3$ ($L \leq 4$) and periodic boundary conditions are applied. The factor of 16 arises because the pyrochlore lattice consists of a fcc lattice of tetrahedra, each tetrahedron has four spins, and there are four sites of the fcc lattice in the conventional cubic cell. To incorporate the B phase with periodic boundary conditions, we need L to be even, so most of our results are for $L=2$ and 4. Often the long-range dipolar interactions are cut off beyond a couple of neighbors²³ to speed up the simulations. However, here we have kept a large number of neighbors (practically infinite) in order to reproduce accurately the structure of the degenerate states. If the dipole-dipole interaction is cut off, ripples appear in the degenerate lines of soft modes.^{3,10,11} We constructed periodic repetitions of the Monte Carlo clusters and included the contributions of many blocks in performing the dipolar sums. Because there is no cutoff in the dipole interactions, the simulation becomes slow for large sizes, so we are limited to $L \leq 4$ ($N \leq 1024$).

We investigate ordering at $\mathbf{q}=0$ (called A , see Fig. 1), and at $\mathbf{q}=\boldsymbol{\pi}$ (called B , see Ref. 10). With A ordering, the order parameter has $n=3$ components and with B ordering it has $n=4$ components, as detailed in Sec. III. The n -component order parameters are given by

$$\boldsymbol{\psi} = (\psi_1, \dots, \psi_n), \quad (2)$$

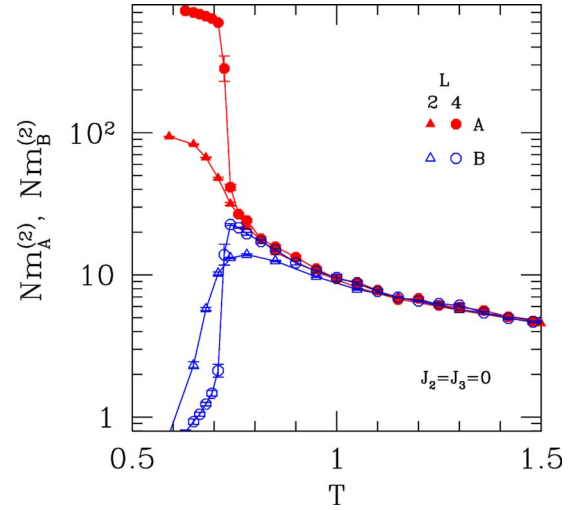


FIG. 2. (Color online). Order parameter squared ($\times N$) for the A phase, shown in Fig. 1, and the B phase, for $J_2=J_3=0$ as function of temperature T in a Monte Carlo simulation of the dipolar pyrochlore Heisenberg antiferromagnet. We see a large jump in the order parameter for the largest system size $L=4$ signaling the onset of a strong first-order transition. The number of spins is given by $N=16L^3$.

$$\psi_\alpha = \frac{1}{N} \sum_{i=1}^N \mathbf{S}_i \cdot \mathbf{e}_i^{(\alpha)}, \quad (3)$$

where $\mathbf{e}_i^{(\alpha)}$ is the unit vector of spin i assuming the system is fully ordered in component α of ordering type A with the (xy) state [resp. $(xz), (yz)$] corresponding to $\boldsymbol{\psi}=(1,0,0)$ [resp. $\boldsymbol{\psi}=(0,1,0), \boldsymbol{\psi}=(0,0,1)$; see, e.g., the arrows in Fig. 1] or B . In the course of the simulation of a finite system, the spin configuration can fluctuate between different, equivalent ordered states. We therefore compute the invariant quantities

$$m^{(2)} = \sum_{\alpha=1}^n \langle \psi_\alpha^2 \rangle; \quad m^{(4)} = \left\langle \left(\sum_{\alpha=1}^n \psi_\alpha^2 \right)^2 \right\rangle. \quad (4)$$

It is convenient to also compute the dimensionless Binder ratio

$$g = \frac{1}{2} \left[(n+2) - n \frac{m^{(4)}}{(m^{(2)})^2} \right], \quad (5)$$

for both A and B orderings, which has the property that it tends to 0 at high temperature and to 1 in an ordered state. (Remember that $n=3$ for A -type ordering and $n=4$ for B -type ordering.)

Firstly we consider the dipolar model with only nearest neighbor interactions. Results for the order parameters are shown in Fig. 2. The parameter J is taken to be the same as in $\text{Gd}_2\text{Ti}_2\text{O}_7$, i.e., from the high temperature susceptibility $J=0.4$ K, and the dipole-dipole interactions are fixed by the inter ion distances.^{3,4} The order parameter associated with the A state jumps rapidly at $T \approx 0.7$ K, indicating a strong first-order transition at that temperature, whereas that of the B state goes to zero at the same temperature. These results

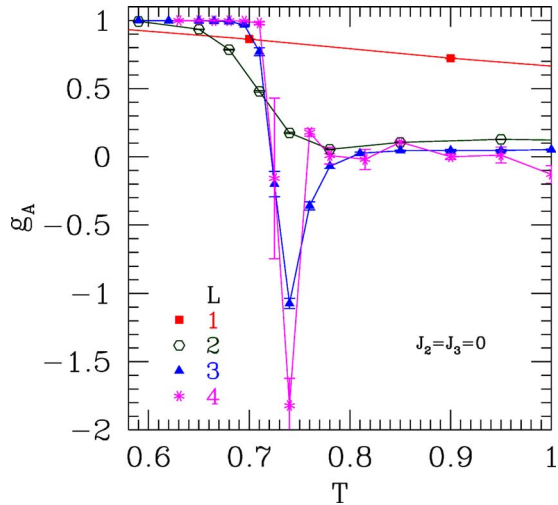


FIG. 3. (Color online). Binder ratio for the A phase for different system sizes L for $J_2=J_3=0$.

give a much lower value of the transition temperature T_N than the mean-field theory value of 5.3 K, though it is a bit smaller than the experimental value of 1 K.

The Binder ratio shown in Fig. 3 is also strongly discontinuous. In the vicinity of T_c the Binder ratio gets negative, as expected for a first-order phase transition.²⁴ Given the results of Figs. 2 and 3, we conclude that the transition is *strongly* first order for $J_2=J_3=0$. The very small preference for the B phase^{9–11} indicated by the small minimum of $J(\mathbf{q})$ at $\mathbf{q}=\pi$, which would be relevant at a second-order transition, is unimportant here because the transition is so strongly first order.

We now include further neighbor interactions J_2 and J_3 that lift the degeneracy of the dipolar model and select other states, as studied in detail in Ref. 10. The lowest part of the spectrum of $J(\mathbf{q})$ is shown in Fig. 4 for \mathbf{q} along the (1, 1, 1) direction. We see that spectrum is almost precisely degenerate for $J_2=J_3=0$ but that $\mathbf{q}=0$ is preferred if J_2 and J_3 are negative, while $\mathbf{q}=\pi$ is preferred if J_2 and J_3 are positive. For simplicity we restrict ourselves to $J_2=J_3$ and study how

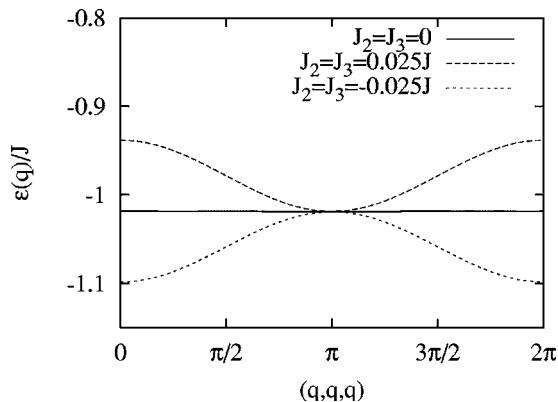


FIG. 4. The lowest part of the spectrum of $J(\mathbf{q})$ (from Ref. 10). One sees that the degeneracy of the critical soft modes is lifted by second and third neighbor couplings. A positive (resp. negative) $J_2=J_3$ favors $\mathbf{q}=\pi$ (resp. $\mathbf{q}=0$).

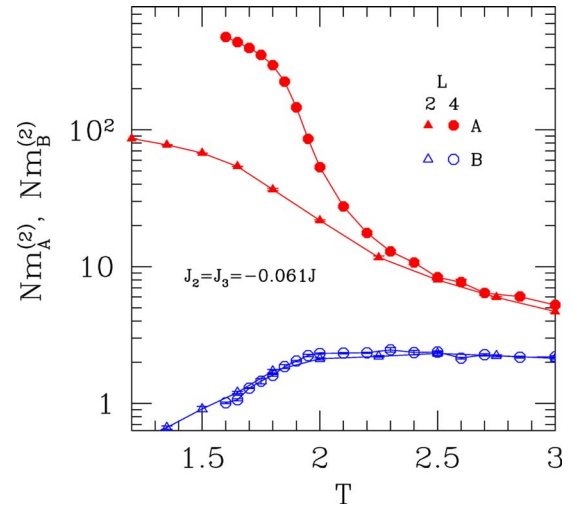


FIG. 5. (Color online). Order parameters squared ($\times N$) for $J_2=J_3=-0.061J$. A transition occurs to the A phase ($\mathbf{q}=0$) which is more gradual than for the degenerate case with $J_2=J_3=0$ shown in Fig. 2.

the character of the transition is modified relative to the case $J_2=J_3=0$.

For $J_2=J_3<0$, we find the same A state as for $J_2=J_3=0$, but the transition temperature shifts to higher temperatures; see Figs. 5 and 6. This is expected from Fig. 4 since, with $J_2=J_3<0$, $J(\mathbf{q})$ acquires a well-defined minimum at $\mathbf{q}=0$ which gets deeper with increasing J_2 and J_3 .

Even with couplings as small as $J_2=J_3=-0.061J$, the order parameter and the Binder ratio, shown in Fig. 6, vary in a much more gradual way than for the degenerate case $J_2=J_3=0$. Although only finite-size scaling on a bigger range of sizes could say whether the transition is first or second order, it is clear that removing the degeneracy makes the transition less first-order compared with the degenerate case. These results are consistent with earlier simulations on model without dipole-dipole interactions (where larger clus-

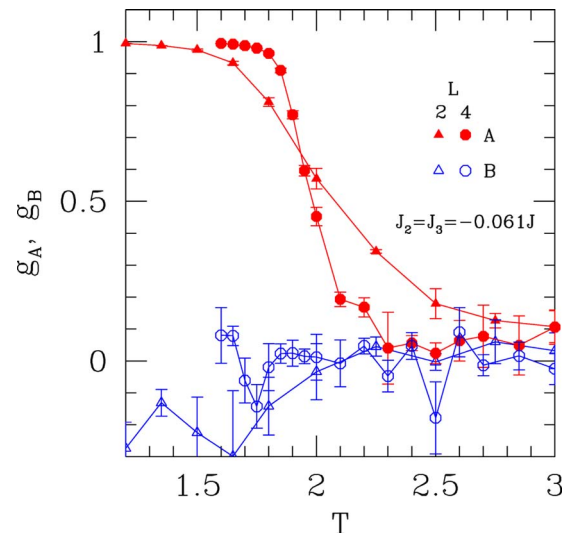


FIG. 6. (Color online). The Binder ratio for the A and B phases for different system sizes L for $J_2=J_3=-0.061J$.

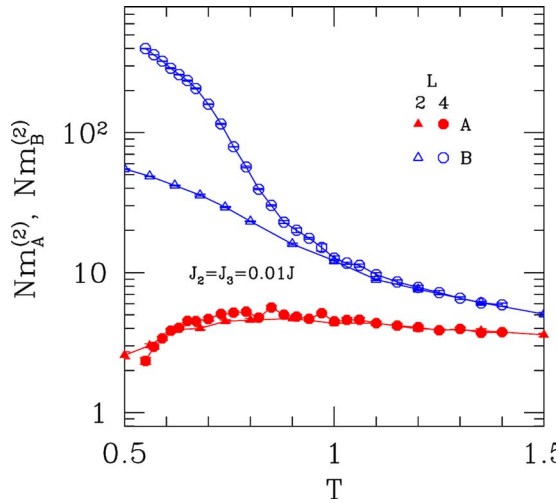


FIG. 7. (Color online). Order parameters obtained by Monte Carlo simulation for $J_2=J_3=0.01J$. The transition gives rise to the B -like state ($\mathbf{q}=\boldsymbol{\pi}$) and is more gradual than for the case of $J_2=J_3=0$ shown in Fig. 2.

ters could be considered) which pointed out a continuous²⁵ or *weakly* first-order²⁶ transition for the pyrochlore lattice with $J_3 < 0$.

Next we consider $J_2=J_3 > 0$ which, from Fig. 4, is expected to favor the B -like states ($\mathbf{q}=\boldsymbol{\pi}$), and indeed this is the case as shown by Figs. 7 and 8. The minimum of $J(\mathbf{q})$ at $\mathbf{q}=\boldsymbol{\pi}$ shown in Fig. 4 is independent of J_2 and J_3 which implies that the mean-field transition temperature is also independent of J_2 and J_3 . Although we do not have enough system sizes to attempt a serious estimate of T_N using finite-size scaling, it seems that the transition temperature is indeed quite similar to that for $J_2=J_3=0$

As was also found for $J_2=J_3 < 0$, the transition is much more gradual than for the degenerate case, showing that removing the degeneracy reduces the first-order character of the transition. The sensitive dependence of both the order of

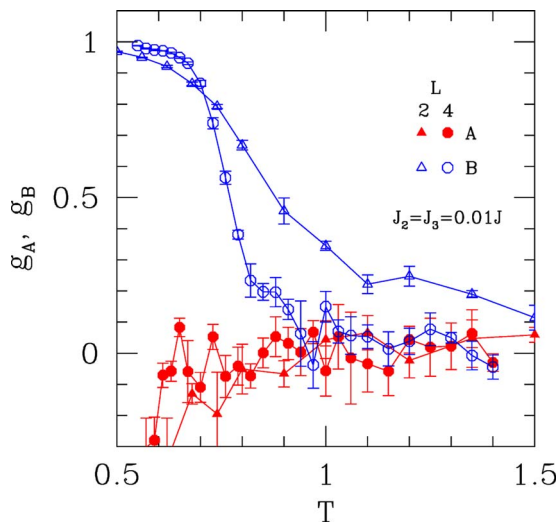


FIG. 8. (Color online). Binder ratio for the A and B phases for different system sizes L for $J_2=J_3=0.01J$.

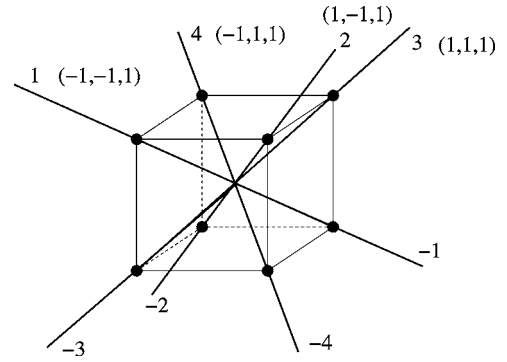


FIG. 9. Lines of minimum energy in reciprocal space, given by the four equivalent $(1,1,1)$ directions (the cube is drawn for convenience). The (lack of) dispersion along the $(1,1,1)$ lines is shown in Fig. 4.

the transition and the nature of the ordered phase on J_2 and J_3 points to the possible relevance of these terms in explaining the difference between $\text{Gd}_2\text{Sn}_2\text{O}_7$ and $\text{Gd}_2\text{Ti}_2\text{O}_7$.

While the data for $J_2=J_3=0$ clearly indicate a strong first-order transition, our results for J_2 and J_3 nonzero (with either sign) are not conclusive as to the order of the transition. The smooth behavior of the Binder ratios shown in Figs. 6 and 8 is typical for a second-order transition. However, we cannot rule out the possibility of a *weak* first-order transition where the correlation length at the critical point, ξ_c , is large. In this case, if $L < \xi_c$ the behavior will look like that of a second-order transition. Only for sizes where $L > \xi_c$ can one see a crossover to behavior expected at a first-order transition. Hence for J_2 and J_3 nonzero, where the degeneracy is removed, the transition is either second order or weakly first order. However, comparing Figs. 6 and 8 with the corresponding figure for $J_2=J_3=0$, Fig. 3, we see that behavior for J_2 and J_3 nonzero is very different from the strong first-order behavior found in the degenerate case.

III. RG ANALYSIS OF THE $(1,1,1)$ MODEL

We now study a Landau-Ginzburg-Wilson (LGW) model by means of the renormalization group analysis. Although the method is usually aimed to study second-order phase transitions, the lack of stable fixed points is often considered as an indication for a first-order kind of transition.

Given the degeneracies of the soft modes with \mathbf{q} along the four equivalent $(1,1,1)$ directions when $J_2=J_3=0$ (see Fig. 4), the fluctuations of all these modes must be taken into account simultaneously. For this reason, we consider a model with an infinite-component order parameter (extended to dimension d) and the fluctuations with wave vectors close to these soft-mode directions. The quadratic part of the Hamiltonian is written

$$H^{(2)} = \int \frac{d^d \mathbf{q}}{(2\pi)^d} \sum_{i=1}^4 \mathcal{G}_{0i}^{-1}(\mathbf{q}) \psi_{i\mathbf{q}} \bar{\psi}_{i\mathbf{q}}, \quad (6)$$

$$\mathcal{G}_{0i}^{-1}(\mathbf{q}) = (r + \mathbf{q}_{\perp,i}^2 + aq_{\parallel,i}^{2m}), \quad (7)$$

$$\mathbf{q}_{\perp,i} = \mathbf{q} - (\hat{v}_i \cdot \hat{\mathbf{q}})\mathbf{q}, \quad (8)$$

where the \hat{v}_i are of norm 1 and represent the $i=1, \dots, 4$ (1,1,1) directions given in Fig. 9. We have eight fields, $\psi_{i\mathbf{q}}$ and $\bar{\psi}_{i\mathbf{q}}$ ($i=1, \dots, 4$), with $\bar{\psi}_{i\mathbf{q}} = \psi_{-i,-\mathbf{q}} = \psi_{i\mathbf{q}}^*$. If we ignore the $aq_{\parallel,i}^{2m}$ term, then, when $r=0$, all the modes with $\mathbf{q}_{\perp,i}=0$ become simultaneously unstable. However, as in previous works,^{20,21} we include the small dispersion along the (1,1,1) lines, $aq_{\parallel,i}^{2m}$, to make the calculation well defined at intermediate stages. To study the degenerate case, it will be eliminated at the end of the calculation by taking $m \rightarrow +\infty$.

The fourth-order invariants are similar to that introduced to describe the nematic smectic-*C* transitions²⁰ and are given by

$$H^{(4)} = \int \frac{d^d \mathbf{q}_1}{(2\pi)^d} \frac{d^d \mathbf{q}_2}{(2\pi)^d} \frac{d^d \mathbf{q}_3}{(2\pi)^d} \frac{d^d \mathbf{q}_4}{(2\pi)^d} \delta_{\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3, \mathbf{q}_4} \mathcal{H}_4, \quad (9)$$

where $\delta_{\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3, \mathbf{q}_4}$ ensures that the total momentum of the four ψ is zero. For instance, we can cancel the momentum by choosing pairs of momenta along the same (1,1,1) line, i.e., by combining ψ_{i,\mathbf{q}_1} with $\psi_{-i,\mathbf{q}_2=-\mathbf{q}_1}$, and similarly with \mathbf{q}_3 and \mathbf{q}_4 . That gives a fourth-order term $u_p \psi_i \bar{\psi}_i \psi_{i+p} \bar{\psi}_{i+p}$ term, where $i=1, \dots, 4$ and $(i+p)$ is meant for $(i+p-4)$ if $(i+p) > 4$. Note that given the C_4 symmetry, u_p does not depend upon i , but only upon $p=0, 1, 2$. In addition, we could choose the first two \mathbf{q} 's along 1 and 3 for instance (see Fig. 9) and the other two along -2 and -4 , which gives $\psi_1 \bar{\psi}_2 \psi_3 \bar{\psi}_4$. Another simplifying feature that we have adopted consists of neglecting the wave vector dependence of the coefficients u_p . Omitting to write the $\mathbf{q}_{1,2,3,4}$ wave vectors, the only fourth-order invariants are given by

$$\mathcal{H}_4 = \sum_{p=0}^2 u_p \sum_{i=1}^4 \psi_i \bar{\psi}_i \psi_{i+p} \bar{\psi}_{i+p} + \frac{1}{2} u_3 (\psi_1 \bar{\psi}_2 \psi_3 \bar{\psi}_4 + \bar{\psi}_1 \psi_2 \bar{\psi}_3 \psi_4), \quad (10)$$

where $\bar{\psi}_i = \psi_{-i}$. We call $H^{(2)} + H^{(4)}$ the (1,1,1) model. First, we consider the Hartree correction¹⁹ to the gap r (self-energy):

$$\mathcal{G}_i^{-1} \equiv r + \mathbf{q}_{\perp,i}^2 + \Sigma_i(r), \quad (11)$$

$$\Sigma_i(r) = \frac{1}{6} (3u_0 + 2u_1 + u_2) \int \frac{d^d \mathbf{q}}{(2\pi)^d} \mathcal{G}_{0i}(\mathbf{q}). \quad (12)$$

In $d=3$, if we introduce momentum cutoffs, the new gap $r'(a \rightarrow 0)$ is given by

$$r' = r + \alpha \Lambda' \ln \left(1 + \frac{\Lambda^2}{r'} \right), \quad (13)$$

where α is a proportionality coefficient and Λ and Λ' the cutoffs. Due to the strong singularity of the right-hand side, the gap does not vanish anymore. It suggests that the paramagnetic phase remains locally stable below the transition, together with other more stable phases. From the existence of other phases (at least at the mean-field level), the transition is expected to be first order.¹⁹

However, the cutoffs Λ and Λ' enter explicitly the equation and a more controlled result can be obtained by

renormalization group analysis by restricting the integration to a shell of momentum $\Lambda/b < q < \Lambda$ with $b > 1$.^{20,21} For this we introduce new real order parameters ($i=1, \dots, 4$),

$$\psi_i = \phi_i + i\bar{\phi}_i \quad \bar{\psi}_i = \phi_i - i\bar{\phi}_i, \quad (14)$$

and the quadratic and quartic terms become

$$\begin{aligned} \mathcal{H} = & \sum_{i=1}^4 \mathcal{G}_{0i}^{-1} (\phi_i^2 + \bar{\phi}_i^2) + u_0 \sum_{i=1}^4 (\phi_i^2 + \bar{\phi}_i^2)^2 + u_1 [(\phi_1^2 + \bar{\phi}_1^2) + (\phi_3^2 \\ & + \bar{\phi}_3^2)] [(\phi_2^2 + \bar{\phi}_2^2) + (\phi_4^2 + \bar{\phi}_4^2)] + u_2 [(\phi_1^2 + \bar{\phi}_1^2)(\phi_3^2 + \bar{\phi}_3^2) \\ & + (\phi_2^2 + \bar{\phi}_2^2)(\phi_4^2 + \bar{\phi}_4^2)] + u_3 (\phi_1 \phi_2 \phi_3 \phi_4 + \bar{\phi}_1 \bar{\phi}_2 \bar{\phi}_3 \bar{\phi}_4) \\ & - u_3 (\phi_1 \bar{\phi}_2 \phi_3 \bar{\phi}_4 + \bar{\phi}_1 \phi_2 \bar{\phi}_3 \phi_4) + u_3 (\phi_1 \bar{\phi}_3 + \bar{\phi}_1 \phi_3)(\phi_2 \bar{\phi}_4 \\ & + \bar{\phi}_2 \phi_4). \end{aligned} \quad (15)$$

The derivation of the RG equations for the coupling constants is then similar to that of Ref. 20 except that we have to keep track of the field labels, given the anisotropy of \mathcal{G}_{0i} in Eq. (15). By integrating over a shell of momentum $\Lambda/b < q < \Lambda$, we find the recursion relations for the new coupling constants

$$u'_0 = b^\epsilon [u_0 - (40u_0^2 I_0 + 4u_1^2 I_0 + 2u_2^2 I_0)]$$

$$u'_1 = b^\epsilon [u_1 - (8u_1^2 I_1 + u_3^2 I_1 + 32u_0 u_1 I_0 + 8u_1 u_2 I_0)]$$

$$u'_2 = b^\epsilon [u_2 - (8u_1^2 I_0 + 8u_2^2 I_2 + u_3^2 I_2 + 32u_0 u_2 I_0)]$$

$$u'_3 = b^\epsilon [u_3 - (8u_2 u_3 I_2 + 8u_1 u_3 I_1)]$$

$$a' = b^{-2(m-1)} a. \quad (16)$$

where $\epsilon=4-d$ and the integrals are defined by

$$I_p = \int_{\Lambda/b}^{\Lambda} \frac{d^d \mathbf{q}}{(2\pi)^d} \mathcal{G}_{0i}(\mathbf{q}) \mathcal{G}_{0i+p}(\mathbf{q}). \quad (17)$$

A. Degenerate (1,1,1) model ($a \rightarrow 0$)

The relation (16), together with the divergences of the integrals I_p for $a \rightarrow 0$ (at $r=0$, finite b) implies that a is a dangerous irrelevant variable for $m > 1$. I_0 diverges indeed as $a^{(d-5)/2}$, and $I_{1,2}$ as $a^{(d-3)/2}$. To take into account these divergences, we have to introduce rescaled constants $\tilde{u}_i = u_i a^{(d-5)/2}$ (Refs. 20 and 21 and references therein). With $a^{(5-d)/2} I_{1,2} \rightarrow 0$, the recursion relations become (we introduce $\mathcal{I} \equiv \lim_{a \rightarrow 0} a^{(5-d)/2} I_0$)

$$\tilde{u}'_0 = b^{m(5-d-1/m)} [\tilde{u}_0 - (40\tilde{u}_0^2 + 4\tilde{u}_1^2 + 2\tilde{u}_2^2) \mathcal{I}],$$

$$\tilde{u}'_1 = b^{m(5-d-1/m)} [\tilde{u}_1 - (32\tilde{u}_0 \tilde{u}_1 + 8\tilde{u}_1 \tilde{u}_2) \mathcal{I}],$$

$$\tilde{u}'_2 = b^{m(5-d-1/m)} [\tilde{u}_2 - (8\tilde{u}_1^2 + 32\tilde{u}_0 \tilde{u}_2) \mathcal{I}],$$

$$\tilde{u}'_3 = b^{m(5-d-1/m)} \tilde{u}_3. \quad (18)$$

All the fixed points are unstable for $d < 5 - 1/m$ (the upper critical dimension is 5 for $m \rightarrow \infty$) since $\tilde{u}'_3 = b^{m(5-d-1/m)} \tilde{u}_3$.

Although strictly speaking new fixed points could occur at order ϵ^2 , the present calculation at order ϵ is compatible with the first-order transition observed in the Monte Carlo simulations.

B. No degeneracy

We now remove the degeneracy, e.g., by including further neighbor couplings in the microscopic Hamiltonian.

1. $\mathbf{q}=0$

First of all we assume that the ordering is at $\mathbf{q}=0$. It is simplest to go back to the order parameter shown in Fig. 1, realize that there are three components $\psi_i, i=1, 2, 3$, and that the symmetry is cubic. The Hamiltonian of this cubic model is therefore given by

$$\mathcal{H} = \sum_{i=1}^3 [(r + \mathbf{q}^2)\psi_i^2 + u_0\psi_i^4] + u_1 \sum_{i,j} \psi_i^2 \psi_j^2. \quad (19)$$

There has been a controversy regarding whether the stable fixed point of the cubic model is the Heisenberg or the cubic fixed point. A recent six-loop expansion has shown that for $n > 2.89$, the stable fixed point is the cubic one.²⁷ Depending on the initial values for the coupling constants, the transition could be either first order or continuous. However, to stabilize the *collinear* states (with ψ either (1, 0, 0), (0, 1, 0), or (0, 0, 1) and $\mathbf{q}=0$), the set of initial coupling constants leads to a first-order transition.²⁸

We have seen that a first-order transition is obtained both in the degenerate case and also when there is a well-defined minimum at $\mathbf{q}=0$. Is, then, the degeneracy of soft modes important or not? We note that, in the absence of degeneracy, the transition may be only *weakly* first order. The problem was studied some years ago in the context of the pyrochlore FeF_3 . For this compound, the $\mathbf{q}=0$ state found by neutron scattering was characterized by a three-component order parameter,²⁵ similar to the one we have here. Monte Carlo simulations have shown that the collinear structures with $\psi = (1, 0, 0); (0, 1, 0); (0, 0, 1)$ are preferred, but the transition first appeared to be second order, with unusual critical exponents²⁵ contrary to the RG argument given above. We can reconcile these results by suggesting that the transition may be *weakly* first order, so that the correlation length would exceed the size of the Monte Carlo cluster and the transition would appear second order in the simulation. This is also confirmed by a reexamination of the Monte Carlo results, which suggested that the transition is more likely to be indeed weakly first order.²⁶

2. $\mathbf{q}=\pi$

We now assume that the degeneracy is lifted in such a way that one of the four π wave vectors is selected. Since π and $-\pi$ are related by a reciprocal lattice vector, we have to take into account the fluctuations of four fields only, with \mathbf{q} close to any of the π wave vectors, $\psi_i, i=1, \dots, 4$. The critical model is given by

$$\mathcal{H} = \sum_{i=1}^4 [(r + \mathbf{q}^2)\psi_i^2 + u_0\psi_i^4] + u_1 \sum_{i,j} \psi_i^2 \psi_j^2 + u_3 \psi_1 \psi_2 \psi_3 \psi_4. \quad (20)$$

This model is known to possess unstable fixed points at order ϵ^2 .¹⁸ Therefore the transition to the π phases is also expected to be first order.

On the basis of the LGW models alone, we would conclude that the phase transitions in the dipolar pyrochlore are *all* first order in character. Such a simple analysis does not say whether the transition is *strongly* or *weakly* first order, which is quite a relevant question when one comes to compare with experiments. Nonetheless, the results presented in this section are compatible with the Monte Carlo simulations of Sec. II. The latter are important, precisely to say whether the transitions are weakly or strongly first order.

IV. CONCLUSION

We have considered the dipolar Heisenberg model on a pyrochlore lattice with nearest neighbor interactions and a small amount of second and third neighbor interactions (J_2 and J_3). For $J_2=J_3=0$ the system is highly degenerate (see Fig. 4) and fluctuation effects pick out ordering at $\mathbf{q}=0$ (A type). Monte Carlo simulations show that the transition is very strongly first order in this case, in contrast to mean-field theory which predicts a second-order transition. A first-order transition is also predicted by a renormalization group analysis. When the degeneracy is removed by including J_2 and J_3 the transition is more gradual, showing that the degeneracy is necessary to get a *strong* first-order transition. Given the limited range of sizes in the Monte Carlo simulations, we cannot say from the simulations whether the transition is second order or weakly first order for $J_2=J_3 \neq 0$. However, according to a renormalization group analysis for the nondegenerate case, both A and B type orderings have no stable fixed points, indicating, presumably, a fluctuation-induced first-order transition. Usually this type of transition is only weakly first order, and this seems to be consistent with our numerical data.

Because of the degeneracy for $J_2=J_3=0$, a small amount of second and third neighbor coupling can also change the nature of the ground state. We find that for $J_2=J_3 < 0$ the A phase is retained but for $J_2=J_3 > 0$ we obtain a $\mathbf{q}=\pi$ (B type) ordering. In future work we will study in more detail the nature of this B-type phase, and also consider other possible phases that occur when $J_2 \neq J_3$. It is possible that anisotropic interactions, suggested on the basis of high-temperature electron spin resonance (ESR)²⁹ and by electron paramagnetic resonance (EPR) on diluted samples,^{30,31} may be needed to explain the experimentally observed phases in detail.

Our results provide a natural explanation for $\text{Gd}_2\text{Sn}_2\text{O}_7$ having a strong first-order transition,⁴ while $\text{Gd}_2\text{Ti}_2\text{O}_7$ has a second-order transition¹² (though a weak first-order transition is not ruled out experimentally); namely, second and third neighbor interactions are very weak in $\text{Gd}_2\text{Sn}_2\text{O}_7$, but they are stronger and positive for $\text{Gd}_2\text{Ti}_2\text{O}_7$. In this respect, *ab initio* calculations could give some estimate of the

strength of the couplings. This picture is also consistent with the observations that $\text{Gd}_2\text{Ti}_2\text{O}_7$ orders^{12,13} at $\mathbf{q}=\boldsymbol{\pi}$ while $\text{Gd}_2\text{Sn}_2\text{O}_7$ should be *A* type with equivalent sites and moments perpendicular to the local (1,1,1) directions.¹⁴

In the presence of a magnetic-field,^{5,10} $\text{Gd}_2\text{Ti}_2\text{O}_7$ has a rich phase diagram. For the future, it would also be interesting to perform a study of field-induced transitions in $\text{Gd}_2\text{Sn}_2\text{O}_7$, since this starts off with a quite different state in zero field. In addition to the multiple phase transitions expected on the basis of mean-field theory, the field reduces the fluctuations and hence should reduce the strong first-order character of the transition.

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