Spin Freezing in Geometrically Frustrated Antiferromagnets with Weak Disorder

T. E. Saunders and J. T. Chalker

Theoretical Physics, University of Oxford, 1 Keble Road, Oxford, OX1 3NP, United Kingdom

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We investigate the consequences for geometrically frustrated antiferromagnets of weak disorder in the strength of exchange interactions. Taking as a model the classical Heisenberg antiferromagnet with nearest neighbor exchange on the pyrochlore lattice, we examine low-temperature behavior. We show that spatial modulation of exchange generates long-range effective interactions within the extensively degenerate ground states of the clean system. Using Monte Carlo simulations, we find a spin glass transition at a temperature set by the disorder strength. Disorder of this type, which is generated by random strains in the presence of magnetoelastic coupling, may account for the spin freezing observed in many geometrically frustrated magnets.

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Geometrically frustrated magnets—materials in which magnetic ions form a lattice consisting of frustrated units such as triangles and tetrahedra—characteristically remain in the paramagnetic phase even at temperatures that are small on the scale set by exchange interactions. Nevertheless, at sufficiently low temperature most examples exhibit spin freezing [1]. Indications of freezing include a difference between field-cooled and zero-field-cooled susceptibilities [2–7], a suppression of inelastic magnetic neutron scattering [4,5,8,9], and in some cases a divergence in the nonlinear susceptibility on approaching T_F, as in conventional spin glasses [10]. Some well-studied systems are $SrCr_{8-x}Ga_{4+x}O_{19}$ (with a layered structure consisting of slabs from the pyrochlore lattice) [2–4,8], the kagomé antiferromagnet hydronium jarosite [5,6], and the pyrochlore $Y_2Mo_2O_7$ [7,9].

The origin of this spin freezing has long been puzzling. On the theoretical side, it is established that freezing is absent from some simple models without disorder, including the classical Heisenberg antiferromagnet with nearest neighbor interactions on the pyrochlore lattice [11]. While a more realistic treatment should take account of disorder and various residual interactions, in some of these materials there appears to be little structural disorder [5], and in others [3] T_F is rather insensitive to the identified form of disorder, dilution at magnetic sites.

In this context, recent experiments which show the importance of random strains in two pyrochlore materials are particularly interesting, since via magnetoelastic coupling such strains will lead to local variations in the strength of exchange interactions. One material is $Y_2Mo_2O_7$, in which disorder in Mo-Mo distances has been revealed using x-ray absorption fine structure [12]. The other is $Zn_{1-x}Cd_xCr_2O_4$. Unusually for a geometrically frustrated magnet, in pure form ($x = 0$) this has a first-order transition to a low-temperature phase in which magnetic degeneracy is lifted by a lattice distortion and there is Néel order [13]. This phase has a striking sensitivity to substitution of Cd for Zn: $x = 0.03$ is sufficient to suppress Néel order completely, with spin freezing taking its place [14]. The effect is argued [14] to arise from strains around sites, generated because Cd^{2+} has a larger ionic radius than Zn^{2+}. In remarkable contrast, Néel order is much less sensitive to magnetic dilution by substitution of Gd for Cr: it survives with up to 25% of magnetic ions removed [14].

Taking the background outlined above as motivation, our aim in this Letter is to discuss the effects of weak exchange randomness in geometrically frustrated antiferromagnets. We focus on the classical Heisenberg model with nearest neighbor interactions on the pyrochlore lattice, because of the large body of experimental work on pyrochlore antiferromagnets, and because its behavior without disorder is well understood [11]. In particular, the disorder-free model is known to have an extensively degenerate, connected ground-state manifold [11], and a dipolar form for spin correlations in the limit of low temperature T [15,16]. In the presence of exchange disorder one expects two regimes according to its amplitude. Characterizing interactions by their average J and a magnitude Δ of fluctuations, for strong disorder ($\Delta \gtrsim J$) the model is simply a conventional example of a spin glass. Our interest lies instead with the weak disorder limit ($\Delta \ll J$) in which exchange randomness acts as a perturbation lifting the ground-state degeneracy of the clean system. In the following we show that projection of fluctuations in nearest-neighbor exchange interactions into the ground-state manifold of the clean system generates long-range effective interactions. In addition, using Monte Carlo simulations with parallel tempering, we show for $\Delta \ll J$ that the model has a transition at a temperature $T_F \propto \Delta$. As the transition is approached from above, the spin glass susceptibility diverges, and for $T < T_F$ there is long-range spin glass order.

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We start from the Hamiltonian

$$\mathcal{H} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j, \tag{1}$$

in which classical spins $\mathbf{S}_i$ are three-component unit vectors at the sites i of a pyrochlore lattice, and exchange interactions J_{ij} are nonzero only between neighboring pairs of sites. We begin with a qualitative discussion of the special features of this model at weak disorder.

As a first step, consider a single tetrahedron taken from this lattice, with spins $\mathbf{S}_1, \ldots, \mathbf{S}_4$ at the vertices. Its ground states are the configurations for which $\sum_i \mathbf{S}_i = \mathbf{0}$. With all J_{ij} equal, spin stiffness is zero in this toy problem in the sense that, within the set of ground states, the orientations of a pair of spins can be chosen arbitrarily. The consequences of small variations in J_{ij} with amplitude Δ have been set out in Ref. [17]: generically, a unique ground state is selected (up to global spin rotations) in which all four spins are collinear and the spin pairs linked by the strongest interactions are arranged antiparallel. Variations in J_{ij} hence induce a ground-state stiffness, since changes in the relative orientation of a pair of spins cost an energy $\mathcal{O}(\Delta)$.

Moving to the full, pyrochlore lattice problem, we next argue that small variations in exchange interactions generate long-range effective couplings. The logic is as follows. Without disorder J_{ij} can be block diagonalized by Fourier transform. Its spectrum has four branches in the Brillouin zone. The lowest two branches are degenerate and independent of wave vector, mirroring the ground-state degeneracy of the model. In the limit $\Delta \ll J$ it is natural to project the matrix J_{ij} onto this degenerate subspace. The matrix elements P_{ij} of the projection operator have a dipolar form [18]: they decrease as $|\mathbf{r}_i - \mathbf{r}_j|^{-3}$ for large $|\mathbf{r}_i - \mathbf{r}_j|$. Hence so does the projected interaction matrix. It is partly the long range of these effective interactions that distinguishes the system at $\Delta \ll J$ from conventional spin glasses. Although dipolar interactions in a conventional spin glass have been shown not to be sufficiently long ranged to generate different critical behavior from that with short-range interactions [19], it is not clear at present whether this conclusion carries over to the problem we are concerned with.

The interaction matrix and, in the limit $\Delta \ll J$, its projected version, enter directly into a calculation in which the fixed spin length $|\mathbf{S}_i|^2 = 1$ is treated within the spherical approximation $\sum_i |\mathbf{S}_i|^2 = N_s$ (where the sum is over N_s spins in the lattice). In the absence of disorder such an approach is exact for a model in which the number of spin components $n \to \infty$ [20], and gives an excellent treatment of low T correlations at all n [15]. Applying it and the replica method, with a Gaussian distribution for J_{ij} of variance Δ^2, we find [21] for $\Delta \ll J$ that there is a spin glass transition at a temperature independent of J and proportional to Δ:

$$k_B T_F = \sqrt{8/3}\Delta. \tag{2}$$

A difficulty in proceeding further with a conventional replica treatment lies in restricting spin configurations to the manifold of ground states for the model without disorder, as is necessary for $\Delta, T \ll J$. A natural and elegant way of building in that constraint is to parametrize the ground states in terms of a gauge field, as set out in Refs. [15,16]. We next examine how the effects of weak exchange disorder can be introduced into that formulation. The essence of the gauge field parametrization for the model without disorder can be summarized as follows. A set of vector fields $\mathbf{B}^a(\mathbf{r})$ is introduced to represent spin configurations, with one field for each spin component. The mapping between a spin configuration and vector fields (see [15,16] for details) is made in such a way that the condition for a configuration to be a ground state translates into the condition $\nabla \cdot \mathbf{B}(\mathbf{r}) = 0$. After coarse graining, the fluxes $\mathbf{B}^a(\mathbf{r})$ are treated as continuous, divergence-free fields, with a statistical weight e^{-S_0} (before normalization) given by

$$S_0 = \frac{\kappa}{2} \int d^3\mathbf{r} \sum_a |\mathbf{B}^a(\mathbf{r})|^2, \tag{3}$$

where the stiffness κ is determined by microscopic details of the model. From the example of a single tetrahedron (and the details of the mapping between spins and vector fields), we know that the effect of small variations in J_{ij} is to favor a specific axis for flux. This axis turns out to be one of the cubic crystal axes: the particular one selected depends on the values of J_{ij} and varies randomly from one tetrahedron to another. We therefore propose an effective theory for a geometrically frustrated antiferromagnet with weak exhange disorder, taking $S_{\text{eff}} = S_0 + S_{\text{dis}} + S_{\text{int}}$ with

$$S_{\text{dis}} = -\beta\Delta \int d^3\mathbf{r} \sum_a [\mathbf{B}^a(\mathbf{r}) \cdot \mathbf{n}(\mathbf{r})]^2, \tag{4}$$

$$S_{\text{int}} = \beta u \int d^3\mathbf{r} \left[\sum_a |\mathbf{B}^a(\mathbf{r})|^2 \right]^2. \tag{5}$$

Here, the preferred local axis for flux is defined by the random field $\mathbf{n}(\mathbf{r})$, which has average $[n_i(\mathbf{r})]_{\text{av}} = 0$ and variance $[n_i(\mathbf{r}) n_j(\mathbf{r}')]_{\text{av}} = \delta_{ij}\delta(\mathbf{r} - \mathbf{r}')$. Inverse temperature is denoted by β: S_{dis} dominates over S_0 at low temperature, since the first is an energetic contribution while the second is entropic. Microscopics imply an upper bound to $|\mathbf{B}^a(\mathbf{r})|$, imposed here by S_{int} with phenomenological coefficient u. Note that S_{eff}, like $\mathcal{H}$, is invariant under global spin rotations, which translate to rotations between the different $\mathbf{B}^a(\mathbf{r})$. In the langauge of this effective theory, spin freezing is condensation of flux into a specific arrangement favored by disorder.

To search for such freezing, we turn to Monte Carlo simulations. While we believe that the formulation of Eqs. (3)–(5) most clearly embodies the special features

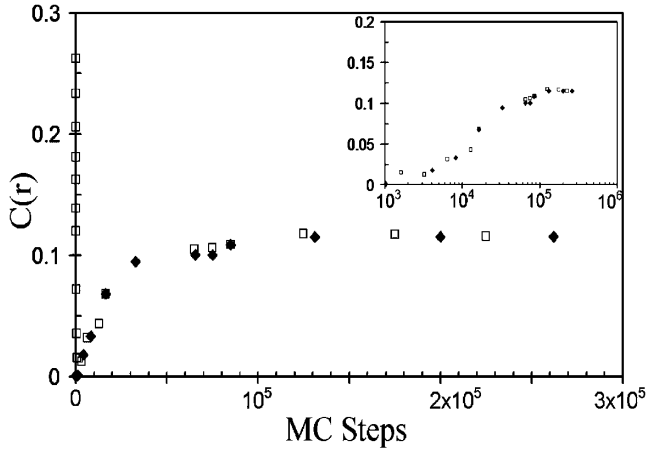

FIG. 1. Evolution of $C(\mathbf{r})$ with Monte Carlo time for $T/J = 0.01$, $\Delta/J = 0.1$, $L = 7$, and maximum r; open and filled symbols are from runs with different initial states, as described in the main text. Inset: Same data on logarithmic time scale.

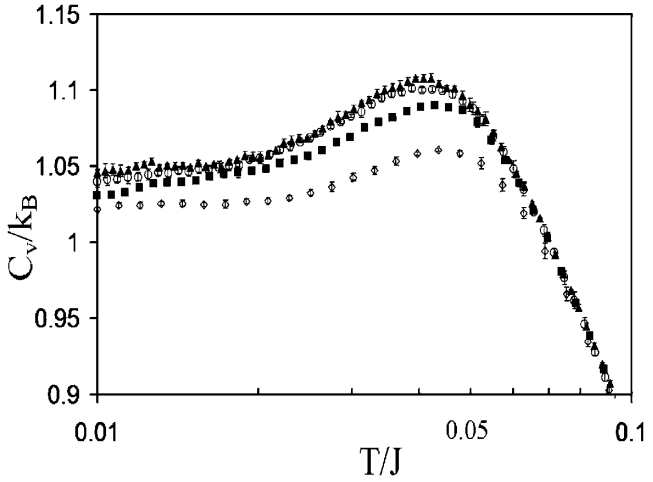

FIG. 2. C_v (in units of k_B) vs T/J for $\Delta/J = 0.1$ and (from top to bottom) $L = 6$, 5, 4, and 3.

of the system at $\Delta \ll J$, we choose instead to simulate the Hamiltonian of Eq. (1) because it is closer to being a realistic microscopic model. Our work builds on the initial investigation of Ref. [22], but is much more detailed. In common with a recent study of the spin glass transition in the three-dimensional Edwards-Anderson Heisenberg model [23], we use parallel tempering [24] to reach equilibrium at low temperature. Some details are as follows. We take J_{ij} independently and uniformly distributed on the interval $[J - \Delta, J + \Delta]$ with $0.05 \leq \Delta/J \leq 0.2$, and study the temperature range $10^{-2} \leq T/J \leq 1$. System sizes, specified by the linear dimension L in units of the lattice constant and by the number of spins $N_s = 4L^3$, are $2 \leq L \leq 7$ and $32 \leq N_s \leq 1372$. Run lengths vary from 5×10^3 Monte Carlo steps per spin (MCS) for $L = 2$ to 2×10^5 for $L = 7$. Results are averaged over a number of disorder realizations varying from 10^3 for $L = 2$ to 200 for $L = 7$.

We present data for three quantities: the heat capacity per spin C_v, the spin glass correlation function $C(\mathbf{r})$, and the spin glass susceptibility χ. The last two are defined in terms of the behavior of two copies of a system with identical disorder. Labeling the copies with $l = 1, 2$ denoting thermal averages in each copy by $\langle \cdots \rangle_l$ and a disorder average by $[\cdots]_{\text{av}}$, we have

$$C(\mathbf{r}) = [\langle \mathbf{S}(0) \cdot \mathbf{S}(\mathbf{r}) \rangle_1 \langle \mathbf{S}(0) \cdot \mathbf{S}(\mathbf{r}) \rangle_2]_{\text{av}} \qquad (6)$$

and $\chi = \sum_{\mathbf{r}} C(\mathbf{r})$.

First, as a test for equilibration, we examine $C(\mathbf{r})$ as a function of simulation time, comparing initial conditions for which $C(\mathbf{r}) = 1$ (both copies initially in the same Néel ground state of the diorder-free model) with ones for which $C(\mathbf{r}) = 0$ (each copy initially an independent random spin configuration). Results are shown in Fig. 1 for the most demanding case (maximum L and r, minimum T). On this basis, for $L = 7$ we collect data after discarding an initial 1×10^5 MCS. Equilibration is much more rapid at smaller L or larger T.

The heat capacity, shown in Fig. 2, varies smoothly with T, as expected in the absence of a Néel ordering transition. The consequences of exchange randomness are revealed in the limiting value of C_v at small T: without disorder this is [11] $3k_B/4$ for large L, reflecting the zero modes (1/4 of all degrees of freedom) of the ground states, but with disorder it rises to k_B, because all zero modes are removed, except for the three arising from global spin rotations.

Next, we present the central result of our simulations, the behavior of the spin glass correlation function $C(\mathbf{r})$, shown in Fig. 3. We believe this provides clear evidence that $C(\mathbf{r})$ is nonzero at large r below a transition temperature T_F. From simple inspection of this figure $0.04 < T_F < 0.02$ at $\Delta/J = 0.1$. In an effort to determine T_F more precisely, and to illustrate finite-size effects, we turn to the susceptibility χ, shown in Fig. 4. The rapid increase in

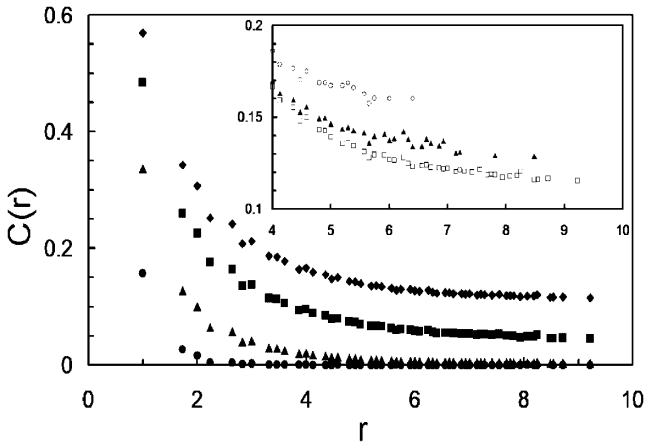

FIG. 3. $C(\mathbf{r})$ vs r for $L = 7$ and $\Delta/J = 0.1$ at (from top to bottom) $T/J = 0.01$, 0.02, 0.04, and 0.1. Inset: Dependence on system size at $T/J = 0.01$, $L = 5$, 6, 7.

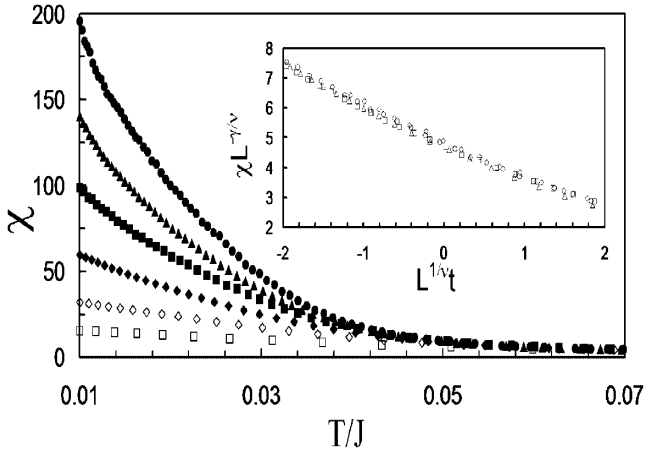

FIG. 4. χ vs T/J for $\Delta = 0.1$ J and system sizes from $L = 2$ to $L = 7$. Inset: Scaling collapse.

χ with L at low T is clear. Close to T_F one expects the finite-size scaling behavior

$$\chi(T, L) = L^{\gamma/\nu} f(L^{1/\nu} t), \tag{7}$$

where $t = (T - T_c)/T_c$, and ν and γ are the standard critical exponents for the correlation length and susceptibility. Scaling collapse of the data is shown in the inset of Fig. 4, with $T_F = 0.023$, $\nu = 1$, and $\gamma = 1.45$. Uncertainties in these parameters are hard to quantify because finite-size effects are large for $L = 2$ and $L = 3$ ($N_s = 32$ and $N_s = 108$); omitting these sizes, for $\Delta/J = 0.1$ collapse is obtained with $0.020 \leq T_F \leq 0.32$, $0.9 \leq \nu \leq 1.2$, and $1 \leq \gamma \leq 1.6$.

Finally, we examine the dependence of T_F on Δ. On dimensional grounds, one has $T_F/\Delta = g(\Delta/J)$, and from our discussion of spin stiffness in a single tetrahedron we expect $g(x)$ to be finite in the weak disorder limit $x \to 0$. We have evaluated $C(\mathbf{r})$ as a function of T for $\Delta/J = 0.2$, 0.1, 0.75, and 0.05, considering only $L = 5$ ($N_s = 500$) because of limitations on computational resources. As shown in Fig. 5, the dependence of data for $C(\mathbf{r})$ at fixed r on T and Δ can be reduced to a single scaled variable $T/T_0(\Delta)$, and $T_0(\Delta) \propto \Delta$ for small Δ.

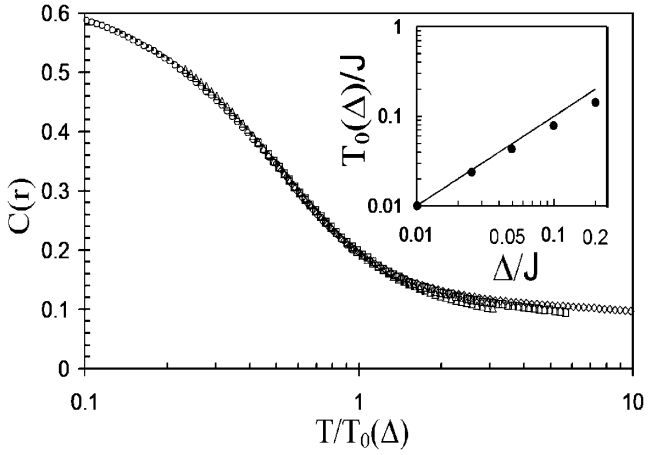

FIG. 5. Dependence on disorder strength Δ: $C(\mathbf{r})$ for $r = 4$ as a function of $T/T_0(\Delta)$. Inset: $T_0(\Delta)/J$ vs Δ/J; line is a guide to the eye.

In summary, we have shown that weak exchange randomness in the classical Heisenberg antiferromagnet on the pyrochlore lattice generates long-range effective interactions, and that these are responsible for a spin glass transition at a temperature set by the disorder strength. We suggest that this may account for spin freezing observed in many geometrically frustrated magnets.

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