Hidden Order in a Frustrated System: Properties of the Heisenberg Kagomé Antiferromagnet

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We show that the classical Heisenberg antiferromagnet on a Kagomé lattice is an example of a spin nematic. By calculating the effects of thermal fluctuations around ground states, and by Monte Carlo simulations, we find entropically driven, local spin-nematic order at low temperature T with a correlation length which is divergent in the limit $T \to 0$. Dynamical correlations are also studied, within the harmonic approximation, in the nematically ordered states.

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Frustrated antiferromagnets are interesting particularly because of the scope they offer for novel low-temperature states, which may be magnetically disordered [1,2] or have only unconventional (non-Néel) order [3–5]. One example, the Heisenberg antiferromagnet on a Kagomé lattice, has been proposed as a model for two experimental systems: ^{3}He adsorbed on graphite (with spin $S=\frac{1}{2}$) [6], and the insulating, layered compound $SrCr_8Ga_4O_{19}$ (with $s=\frac{3}{2}$) [7,8].

In this Letter we discuss the properties of the Heisenberg Kagomé antiferromagnet in the classical limit ($S=\infty$), by low-temperature expansion, by Monte Carlo simulation, and via spin-wave theory. We find that the system displays "order from disorder" [9], the order being spin nematic [4,10,11]. Specifically, thermal fluctuations select, from the vicinity of the highly degenerate classical ground states, configurations in which spins are coplanar. We show that such nematic states support "disguised" spin waves, which have properties consistent with recent inelastic neutron scattering measurements on $SrCr_8Ga_4O_{19}$.

Our approach is complementary to recent treatments of the model with $S=\frac{1}{2}$ [12], and to the quantum-fluid description of Chandra and Coleman [4]. An interesting difference between our results (for $S=\infty$) and those (for S finite) of the latter authors is in the symmetry of the nematic state. Using the classification of Andreev and Grishchuk [10], we find (for $T \to 0$) n-type as opposed to [4] p-type order.

In brief, one expects the dominant classical, low-temperature configurations to be close to whichever ground states have the softest fluctuations [13,14]. The ground states in which spins are coplanar (so defining a nematic axis perpendicular to the plane) are unique in having an entire branch of soft modes. Consequently, the nematic correlation length is divergent in the low-temperature limit.

As a starting point, we need an understanding of the classical ground states of the Heisenberg Kagomé antiferromagnet. A picture can be built up as follows, with the Hamiltonian

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

where $\mathbf{S}_i$ is a classical, three-component unit vector, and the sum runs over pairs of sites on the Kagomé lattice (Fig. 1). The energy is minimized by any configuration for which the total spin of each elementary triangle on the lattice is zero. In such states, the spins of a given triangle lie in one plane, forming a rigid unit in spin space; the degeneracy stems from the many ways of fitting these units together. Consider first the subset of ground states having *all spins coplanar*. Each such planar state is highly constrained, in the sense that only three distinct spin orientations occur, but the subset as a whole retains a large degeneracy: It has an extensive entropy, and

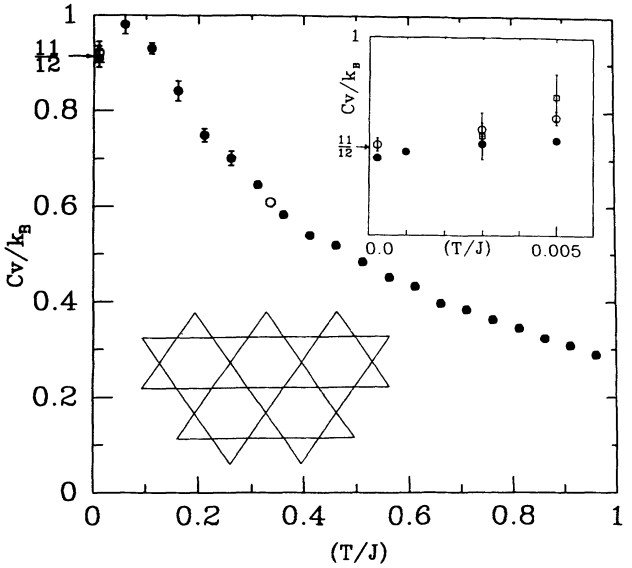

FIG. 1. Temperature dependence of specific heat. Upper inset: Enlargement of low-T region. Lower inset: Kagomé lattice. 108, 432, and 768 site systems: ●, ○, and □.

there is evidence [8] for power-law decay with distance of the correlation function $\langle \mathbf{S}_0 \cdot \mathbf{S}_R \rangle$ averaged over the subset. *Nonplanar states* can be generated by continuous distortions of a planar state, without crossing energy barriers. To do this, line defects are introduced into the planar state by (i) finding a closed line of adjacent sites which has, as its nearest neighbors off the line, only spins of one orientation, $\mathbf{S}$, and (ii) rotating rigidly all spins on the line about the axis defined by $\mathbf{S}$. In fact [15], given suitable boundary conditions (spins in surface triangles all coplanar), *all ground states* can be generated by repeated introduction of defects into the different parent planar states.

The set of all ground states is disordered, in the sense that a zero-temperature average would presumably leave any correlation function short ranged. At low temperature T, however, the entropy of fluctuations selects configurations which are locally close to planar ground states, with a nematic correlation length that is divergent for $T \to 0$. Behavior at $T=0$ is hence discontinuous, which is the essence of Villain's "order from disorder" [9,13,14]. To show this, we start by calculating, in the harmonic approximation, the energy cost of small distortions from an arbitrary planar ground state. The result is remarkable in two ways. First, in a suitable coordinate system, it is identical for *all* planar states. That is, harmonic fluctuations around these states are oblivious to the underlying order or lack of order, and do not select between different planar configurations. Second, there is an entire branch of zero modes. For this branch, energy is independent of displacement to second order, at all wave vectors. Anharmonic forces, discussed below, stabilize the zero modes.

Distortions about nonplanar ground states are much harder to characterize, but it is fortunately possible to prove one essential fact responsible for nematic order: No nonplanar state has as many zero modes as the planar states. Details will be presented elsewhere [15].

To discuss fluctuations around planar states, a suitable coordinate system is one oriented with respect to local spin directions. At each site i we choose right-handed axes in spin space with $\hat{\mathbf{z}}_i$ parallel to $\mathbf{S}_i$ in the particular ground state, and all $\hat{\mathbf{y}}_i$ perpendicular to the ground-state spin plane and mutually parallel. With spin orientations parametrized by $\mathbf{S}_i = (\epsilon_i^x, \epsilon_i^y, 1-\alpha_i)$, with α_i determined from $|\mathbf{S}_i| = 1$, the Hamiltonian [Eq. (1)] becomes $H = H_0 + \sum_{n \geq 2} H_n$, where $H_n \sim O(\epsilon^n)$. Specifically,

$$H_2 = (J/2)[(3\delta_{ij} - M_{ij})\epsilon_i^x \epsilon_j^x + 2M_{ij}\epsilon_i^y \epsilon_j^y], \tag{2}$$

using the summation convention and defining the matrix $\mathbf{M}$ as $M_{ii} = 1$, $M_{ij} = \frac{1}{2}$ if i,j are nearest-neighbor sites, and $M_{ij} = 0$, otherwise. Independence of H_2 from the ground-state directions $\mathbf{S}_i$ is explicit in Eq. (2). The eigenvalues of $\mathbf{M}$, $\lambda_l(\mathbf{q})$, form three branches,

$$l = 0, +, -: \ \lambda_0(\mathbf{q}) = 0 \text{ and } \lambda_\pm(\mathbf{q}) = \tfrac{3}{2}(1 \pm \sqrt{1-A}) ;$$

$$A = \tfrac{8}{9}\{1 - \cos(\pi q_1)\cos(\pi q_2)\cos(\pi[q_1 - q_2])\},$$

where $\mathbf{q} = q_1\mathbf{G}_1 + q_2\mathbf{G}_2$ with $\mathbf{G}_1$ and $\mathbf{G}_2$ basis vectors for the reciprocal lattice. The zero modes correspond to distortions out of the ground-state spin plane; a physically transparent, complete, but nonorthogonal set of eigenvectors for these modes can be defined locally. Each eigenvector is associated with one elementary hexagon of the lattice, having amplitude $\pm 1/\sqrt{6}$ at alternate sites around this hexagon and amplitude zero at other sites. It represents rotations of the spins in each triangle [rigid to $O(\epsilon)$] about axes defined by the spins at the vertices lying off the hexagon. The defect lines discussed above are not, in general, in one-to-one correspondence with these zero modes: While each defect line, in the small amplitude limit, is equivalent to a linear combination of zero modes, there are, in a typical planar state, fewer defect lines than zero modes.

Anharmonic forces play a central role. At low temperatures, it is necessary only to retain the leading terms in H. Anticipating the result, $\epsilon^x \sim O(T^{1/2})$ and $\epsilon^y \sim O(T^{1/4})$, we require from H_3 only terms of order $\epsilon^x(\epsilon^y)^2$ [terms $O((\epsilon^y)^3)$ being absent] and, from H_4, terms of order $(\epsilon^y)^4$. To this accuracy,

$$H_3 = (J/2)\sum_{\langle i,j \rangle} \hat{\mathbf{x}}_i \cdot \hat{\mathbf{z}}_j (\epsilon_i^{y2}\epsilon_j^x - \epsilon_j^{y2}\epsilon_i^x) \tag{3}$$

and

$$H_4 = (J/16)\sum_{\langle i,j \rangle} (\epsilon_i^{y2} - \epsilon_j^{y2})^2 . \tag{4}$$

In calculations of the partition function Z or correlation functions, integration over $\{\epsilon_i^x\}$ leaves a purely quartic effective potential $\tilde{H}_4$ for the zero modes. Each such mode contributes a factor of $T^{1/4}$ to Z, while quadratic modes contribute $T^{1/2}$, so that behavior is dominated at low T by fluctuations around ground states with the largest number of zero modes: the planar states. In particular, the zero modes stabilize the planar ground states against the introduction of line defects, which, when present, remove one zero mode for every hexagon of the Kagomé lattice they share a site with. The coefficients appearing in $\tilde{H}_4$ are different for each planar state and, as a result, the $T \to 0$ Boltzmann probabilities of these states are not all the same. A consequence of this could be additional (e.g., Néel) order.

Our treatment leads to two predictions that can be checked against Monte Carlo simulations. First, the specific heat C_V is sensitive to the presence of zero modes, since, while a quadratic mode contributes $\frac{1}{2}k_BT$ to the internal energy, a zero mode contributes only $\frac{1}{4}k_BT$. Hence we expect $C_V = \frac{11}{12}k_B$ per spin. Second, and more directly, appropriate correlation functions should reveal nematic short-range order. To specify the spin plane of each triangle α, we introduce normals defined by

$$n_\alpha = (2/3\sqrt{3})(\mathbf{S}_1 \times \mathbf{S}_2 + \mathbf{S}_2 \times \mathbf{S}_3 + \mathbf{S}_3 \times \mathbf{S}_1) ,$$

where $\mathbf{S}_1$, $\mathbf{S}_2$, and $\mathbf{S}_3$ are the spins at successive vertices of the triangle α; in any ground state, $|\mathbf{n}_\alpha| = 1$. Nematic or-

der will be evident in the correlation function, $g(\mathbf{r}_{\alpha\beta}) = [\frac{3}{2}\langle(\mathbf{n}_\alpha\cdot\mathbf{n}_\beta)^2\rangle - \frac{1}{2}]$, which takes the value 1 in planar ground states. Expanding around a planar state, $\delta\mathbf{n}_\alpha = -\frac{2}{3}(\epsilon_1^\nu\hat{\mathbf{z}}_1+\epsilon_2^\nu\hat{\mathbf{z}}_2+\epsilon_3^\nu\hat{\mathbf{z}}_2)+O(\epsilon^2)$. We expect, therefore, a characteristic temperature dependence, $1-g(r) \propto \langle\epsilon_i^\nu\epsilon_j^\nu\rangle \sim T^{1/2}$ as $T\to 0$, which serves to emphasize that nematic short-range order will appear only at rather low temperatures $[(T/J)^{1/2}\ll 1]$. It is probable that the nematic correlation length diverges only in the limit $T\to 0$: While a two-dimensional nematic supports topological defects, renormalization group analysis [16] suggests that, in models with non-Abelian continuous symmetry, interactions between small-amplitude fluctuations are sufficient to generate a finite correlation length, preempting a defect-unbinding transition.

To test these ideas, we have performed Monte Carlo simulations of the model defined by Eq. (1). The most interesting behavior is at low temperatures ($T/J < 10^{-2}$), where results are in striking accord with theory. The specific heat, Fig. 1, varies smoothly with temperature. There is no indication of a phase transition at finite temperature. Its low-temperature limit agrees rather precisely with the theoretical value $C_V = [\frac{11}{12} - (5/4N)]k_B$ in a system of N spins. We estimate (via the entropy loss on cooling from $T=\infty$) that, for $S=\frac{3}{2}$, quantum aspects would dominate below $T/J\simeq 0.2$. Short-range nematic correlations, Fig. 2, are large at low temperatures, have the predicted temperature dependence, and extrapolate to complete order as $T\to 0$. Nematic correlations fall with distance, Fig. 3, initially as a power. Antiferromagnetic correlations are also present. We find from a detailed analysis (not shown) that they have the structure of the $\sqrt{3}\times\sqrt{3}$, three-sublattice Néel state [Ref. [8], Fig. 1(a)]. In Fig. 3 (inset) we compare the nematic correlation function $g(r)$ with the spin correlation function, $g_s(r)=\langle\mathbf{S}(0)\cdot\mathbf{S}(r)\rangle$, for spins belonging to the same sublattice of the $\sqrt{3}\times\sqrt{3}$ state. In contrast to the nematic correlations, antiferromagnetic correlations do not saturate even at very low temperatures. While it is clear that the predominant order is nematic, it remains an open and delicate question whether the system supports long-range antiferromagnetic order as $T\to 0$. *If* such order exists in the model studied, it is likely to be sensitive to quantum fluctuations, impurities, or glassy freezing.

The influence of finite system size on these results is indicated in Figs. 1 and 3: Its effects on C_V are very small, and on $g(r)$ are significant only for r approaching the lattice half-width. The influence of finite simulation time (10^6–10^7 Monte Carlo steps per spin at each temperature) requires more discussion. We have been able to reproduce the correlation function data in Fig. 3 (inset) starting from both a high-temperature configuration and the $\sqrt{3}\times\sqrt{3}$ antiferromagnetic state. There are, however, degrees of freedom that do not relax in the course of the simulations. Specifically, we have been unable to equilibrate the $q=0$, three-sublattice, planar antiferromagnetic ground state (which has three spins per magnetic unit cell). This is illustrated in Fig. 2 by the small differences in behavior between samples heated from this state and those cooled from high temperature. We believe the important distinction in this context between the $\sqrt{3}\times\sqrt{3}$ and $q=0$ antiferromagnetic states is that escape from the former is possible via the introduction of short line defects, while escape from the latter involves line defects which span the system.

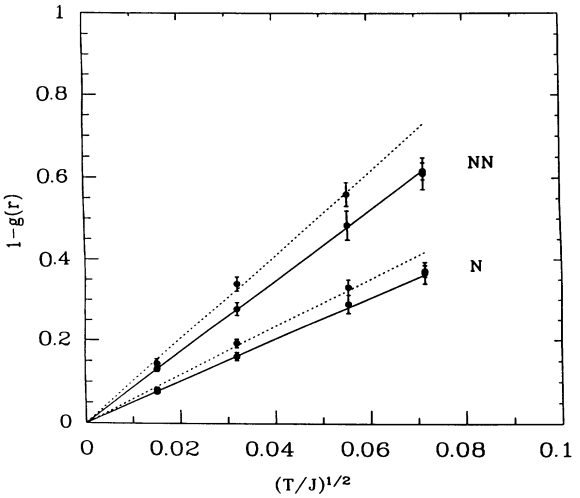

FIG. 2. Temperature dependence of nematic correlation function $g(\mathbf{r}_{\alpha,\beta})$ for α,β nearest-neighbor (N) and next-nearest-neighbor (NN) triangles. 108 site system. The data were obtained by heating the $q=0$, antiferromagnetic ground state (solid lines), and by cooling a high-temperature configuration (dashed lines).

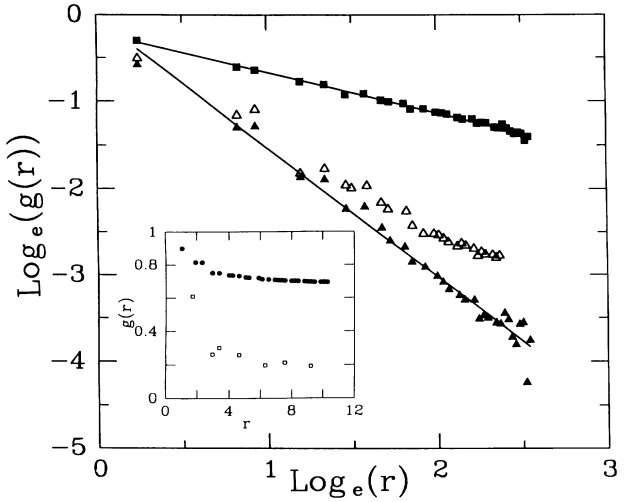

FIG. 3. Distance dependence of nematic correlation function. $T/J=5\times10^{-3}$, 432 and 768 sites: △, ▲. $T/J=3\times10^{-3}$, 768 sites: ■. Inset: Nematic (●) and spin (□) correlation functions $g(r)$ and $g_s(r)$ at $T/J=2.5\times10^{-4}$, 432 sites.

Finally, we consider dynamical correlations within harmonic spin-wave theory. We choose local spin quantization axes as in Eq. (2) and obtain the same spectrum for all planar states, with energies $\omega_l(\mathbf{q}) = JS[2\lambda(3-\lambda)]^{1/2}$, where $\lambda \equiv \lambda_l(\mathbf{q})$, defined above. The branch $l=0$ has zero energy for all $\mathbf{q}$: If anharmonic interactions simply generate a stiffness, it is expected [17] to remain a Goldstone mode. At low energy, the two other branches, $l=+,-$, have linear dispersion and are, respectively, polarized mainly within ($l=+$) and out of ($l=-$) the ground-state spin plane. The two polarizations make qualitatively different contributions to $\chi''(\mathbf{Q},\Omega)$, the imaginary part of the dynamic susceptibility. In-plane polarization is sensitive to disorder in the static correlation function $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ and gives a term broad in Q. Out-of-plane polarization generates a term proportional to $\delta(\mathbf{Q}-\mathbf{q})$, with a form factor which vanishes as $Q \rightarrow 0$ but which is large near other Bragg points.

The possibility of excitations sharp in $\mathbf{Q}$ and Ω around disordered ground states is striking. At present, it is unclear whether this feature survives anharmonic interactions with the zero modes. Results of inelastic neutron scattering on single crystals of $SrCr_8Ga_4O_{18}$ would be especially interesting in that context.

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Note added.—Since submitting this work, we have received three related preprints [18].

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