Kagomé Antiferromagnet with Defects: Satisfaction, Frustration, and Spin Folding in a Random Spin System

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It is shown that site disorder induces noncoplanar states, competing with the thermal selection of coplanar states, in the nearest neighbor, classical Kagomé Heisenberg antiferromagnet. For weak disorder, it is found that the ground state energy is the sum of energies of separately satisfied triangles of spins. This implies that disorder does not induce conventional spin glass behavior. A transformation is presented, mapping ground state spin configurations onto a folded triangular sheet (a new kind of "spin origami") which has conformations similar to those of tethered membranes.

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It is well known that geometrical frustration in some nonbipartite lattices prevents long range magnetic order from being established and allows novel kinds of low temperature magnetic states to develop [1–3]. The Heisenberg Kagomé antiferromagnet with nearest neighbor couplings is one of the most interesting of such systems. The classical system exhibits a rich, nontrivial ground state degeneracy, with both coplanar and noncoplanar states in the degenerate manifold. For the coplanar states, linear spin-wave theory yields one zero-energy mode for every point in the Brillouin zone [4, 5]. All noncoplanar states have fewer zero modes, and, as a result, thermal effects select a nematiclike coplanar ground state [5], an example of the "order by disorder" effect [6, 7]. Numerical studies have confirmed the tendency for thermal selection of the nematiclike state [5, 8], and there is also evidence [4, 5, 8–10] for a tendency toward $\sqrt{3}\times\sqrt{3}$ ordering in the plane.

By far the best-studied experimental Kagomé system is the magnetoplumbite, $SrCr_{9p}Ga_{12-9p}O_{19}$ [11]. For $p=1$, this system contains dense Kagomé layers, separated by dilute triangular layers, of Cr. Although its Curie-Weiss temperature Θ_{CW} (for $p=1$) is over 500 K, no sublattice ordering is found down to helium temperature, where a spin glass, rather than an ordering transition is observed at a temperature T_f. The ratio Θ_{CW}/T_f is about 130, at least for $p>0.5$ [11–13]. T_f itself varies rapidly with doping [12, 13], having its *maximum* value of about 4 K near $p=1$, where one might expect structural disorder to be least important, and falling monotonically as p is reduced. These observations raise two questions: (1) Why is spin glass behavior, with a temperature scale of order J, not generated by nonmagnetic impurities at the 10% to 20% level and (2) what is the origin of the spin-glass-like behavior which is observed even for $p\approx 1$? It is the first question which is addressed in this Letter, while the second is discussed briefly in our conclusions. Our main results are as follows:

(1) Quite generally we find that disorder induces noncoplanarity in the ground state. At low temperatures, the nematiclike state, which is selected by thermal fluctuations, is overwhelmed by this tendency of disorder to induce noncoplanarity [14].

(2) For a large class of distributions of spins of random magnitude, including dilute distributions of vacancies, the ground state configuraton is such that the energy of each separate triangle is minimized. We call this the "rule of satisfied triangles." In the general case, not all triangles are satisfied, but we conjecture that the rule can be extended by replacing "triangles" by more complicated spin clusters.

(3) The rule of satisfied triangles implies that the energy of a collection of nonoverlapping defects is also independent of their spatial arrangement. Hence the system is not a spin glass, despite the change in the degree of frustration introduced by the disorder.

(4) For the uniform system and for moderate randomness, we introduce a mapping of the ground states of the Kagomé system onto a folded, close-packed sheet of triangles. The folding of this sheet of "spin triangles" constitutes a new kind of "spin origami," a term originally coined by Ritchey, Coleman, and Chandra [15] for the folding of spin planes in the Kagomé lattice. The properties of this folded sheet are related to those of "tethered surfaces" which have been studied extensively by Nelson and co-workers [16]. Folding is used to study a variety of ground state configurations for the pure and diluted cases.

In the remainder of this Letter, we justify and elaborate on the four points listed above.

The instability of coplanar states against perturbations to the magnitudes of the spins is easily demonstrated by the case of a single defect spin, S', in an initially coplanar ground state configuration of spins, S. Consider the spins in the two hexagons which include S' [cf. Fig. 1(a)]. Rotations by small angles $\pm\theta$ into or out of the plane,

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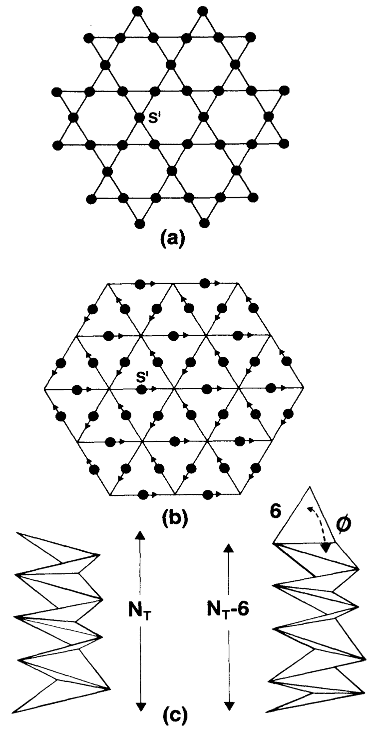

FIG. 1. (a) A Kagomé lattice of spins (dots) with nearest neighbor interactions (solid lines). S' labels a defect spin. (b) The same Kagomé lattice of dots, superimposed on a triangular lattice. The lengths of the sides of each triangle represent the lengths of spins, and the arrows represent their directions. For simplicity, $S = S'$ here. Making S' longer or shorter would require folding the sheet of triangles. As shown, the figure represents the perfect **q**=0 ground state. (c) Left: a triangular sheet of N_T triangles folded into a stack. When flattened into a single triangle, this stack represents the $\sqrt{3} \times \sqrt{3}$ ground state. Right: the "weather vane" defect, a stack of $N_T - 6$ triangles with six stacked triangles (a folded hexagon) protruding. The dihedral angle, ϕ, between the two stacks is arbitrary.

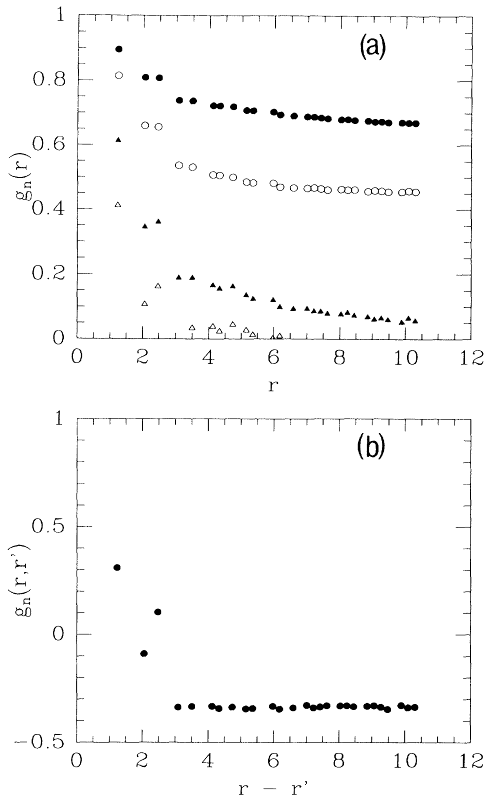

FIG. 2. (a) Nematic correlation function, as defined in Ref. [5], for $T/J = 0.000\,25$ and 432 lattice sites. Solid dots are for no vacancies ($x = 0$); the open dots are for 3 vacancies ($x = 0.007$); the solid triangles are for 8 vacancies ($x = 0.02$); and the open triangles are for 20 vacancies ($x = 0.05$). (b) Nematic correlation function $g_n(\mathbf{r}, \mathbf{r}')$ for a system of 431 spins plus one vacancy, with $\mathbf{r}'$ located in a triangle adjacent to the vacancy and $T/J = 0.000\,25$.

which alternate in sign around each hexagon, may have two possible relative phases. If $\delta S = S' - S < 0$, then the mode which is odd under inversion through the site of spin S' has a negative energy, $\delta E = JS\delta S\theta^2$, and a node at the site of S'. If $\delta S > 0$, then the symmetric mode lowers the energy by $\delta E = -3JS\delta S\theta^2$, and S' is rotated by 2θ. The instability arises because there is no quadratic restoring force for such modes in the perfect system. For a finite density of defects, out-of-plane distortions will compete with thermal excitations favoring coplanarity. If the spatially averaged value of $|\delta S|/S$ is of order 1, then thermal selection cannot suppress out-of-plane canting. If it is small compared to 1, then the canting grows up at low temperatures, $T/JS^2 << 1$.

Figure 2(a) shows the results of a Monte Carlo calculation of the low temperature nematic correlation function, $g(r)$, which is defined in Ref. [5], for various concentrations of vacancies between $x = 0$ and $x = 0.05$. This figure shows that even a low density of vacancies is sufficient to suppress nematic order, and suggests that nematic order should not be observable in samples such as those discussed in Ref. [11]. This large effect results from the fact, derived below, that each isolated vacancy locks at least ten neighboring spins into a noncoplanar configuration.

Next we consider ground state configurations of the Kagomé system, with disorder in the magnitudes of the spins and/or dilution with vacancies, for which the nearest neighbor Heisenberg Hamiltonian may be written as a sum over all triangles, Δ,

$$E = (J/2) \sum_{\Delta} \left[\left(\sum_{m=1}^{3} \mathbf{S}_{\Delta,m} \right)^2 - \sum_{m=1}^{3} \mathbf{S}^2_{\Delta,m} \right], \qquad (1)$$

where m is summed over the three spins in a triangle. This means that a lower bound for the ground state energy is obtained by minimizing the total spin of each triangle separately. A particularly simple case is the one in which the spins in every triangle add to zero, i.e., where they form closed triangles, as happens, for example, in ground states of the perfect system.

A useful device for describing Kagomé spin ground states is spin origami, which is most easily visualized for the case of no disorder. Then in the ground state, each Kagomé triangle of spins forms a closed equilateral triangle in spin space which we call a "spin triangle." Each spin forms an edge shared by two neighboring spin triangles [see Fig. 1(b)], and hence a ground state may be represented by a folded sheet, fashioned by joining spin triangles together along common edges. The $\mathbf{q}$=0 state corresponds to a flat sheet [Fig. 1(b)]. The $\sqrt{3}\times\sqrt{3}$ state is generated by folding the entire sheet into a single triangle, as shown on the left in Fig. 1(c). The so-called "weather vane defect" of the $\sqrt{3}\times\sqrt{3}$ state corresponds to a single protruding triangle, six layers thick, which can make any angle with the rest of the spin triangles as shown on the right in Fig. 1(c).

Introducing defects in the magnitudes of spins can be thought of as shrinking or lengthening sides of spin triangles. In general, this will lead to buckling of the sheet. A vacancy is generated by shrinking a bond shared by two triangles to zero length. The remaining sides of the triangles, which then abut, represent antiparallel spins. To construct the ground state for a single vacancy by folding, a rhombus is cut out of the sheet which is then folded so as to join the cut edges. The resulting sheet can be made coplanar (or even folded into a single triangle) except for two triangles, each four layers thick and involving the four cut edges, which protrude out of the plane, forming two sides of a tetrahedron (faces CEF and ADF of the tetrahedron in Fig. 3). A third face is the plane (ABC), defined by the rest of the spin triangles. The fourth face (BDE) is empty. In this way, noncoplanarity can be localized to the ten spins in the three edges of the two protruding triangles. (These are the ten spins, connected by dashed lines, in the two hexagons which share the vacancy in Fig. 3.) Of course there can also be a larger or even infinite number of noncoplanar spins because of the degeneracy of folding, but only the ten spins around the vacancy are forced to be noncoplanar. Similar arguments apply to any isolated defect spin S', where $0 < S' < 2S$, so that the surrounding spin triangles can close. For $S' > 2S$, the spins in the two triangles involving S' are colinear and the ground state energy is equal to the lower bound in which the sum of the spins in the two triangles involving S' is $S' - 2S$. The usual entropy argument [5] implies that spins away from a vacancy will be coplanar. Figure 2(b) shows the low temperature nematic correlation function, taking the origin in a triangle next to the vacancy. The correlation function decays in a few lattice spacings to a value $-1/3$, which is consistent with the spins being coplanar outside the tetrahedral arrangement of Fig. 3.

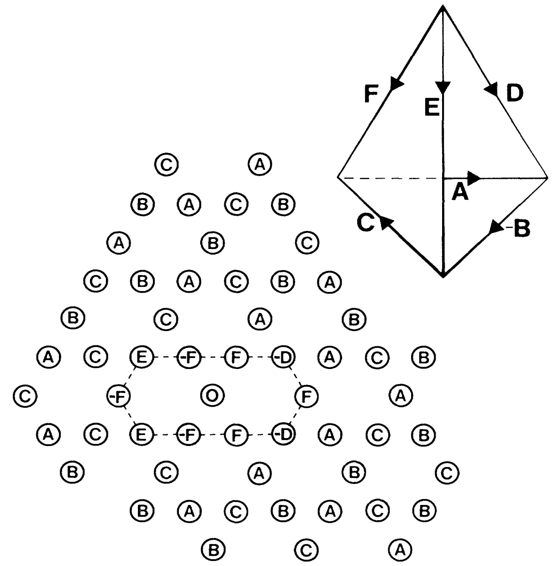

FIG. 3. Spin map of the ground state configuration around a vacancy. A, B, C, ±D, ±E, and ±F denote spin orientations in terms of the directions of labeled edges of the tetrahedron. The tetrahedron is the spin origami representation of the same state. The base ABC contains $N_T - 8$ triangles; the faces CEF and ADF each contain 4 triangles; and the face BDE is empty.

The fact that the effect of an isolated defect is localized to a very small area implies that, for a small concentration of defects, the ground state energy of the disordered system is equal to the sum of energies of independently satisfied triangles. In fact this statement also applies to cases where pairs of defects are close together or even nearest neighbors. In the case of divacancies, the triangle containing the two defects cannot close. Nevertheless, the ground state obeys the rule of satisfied triangles. It can be represented by spin origami by making a cut (a kind of dislocation) along a line of bonds emanating from the divacancy and then overlapping the two rows of triangles along the cut. Furthermore, we have checked that neighboring pairs of divacancies also satisfy the rule of satisfied triangles. Thus it appears that the rule applies to quite a wide range of defect densities and configurations.

Our results for vacancies are consistent with the recent work of Huber and co-workers [17] who studied the diluted Kagomé antiferromagnet and obtained the striking result that the distribution of local fields is discrete with a small number of values: $2JS$, JS, and 0. The rule of satisfied triangles explains this result immediately. Spins on all triangles with no vacancies or one vacancy feel the largest local field, and spins with two neighboring vacancies experience a field JS. The fraction of spins with this local field is proportional to x^2 where x is the fraction of vacancies. Spins with zero local field arise from more complicated configurations or from boundary effects.

It is not the case, however, that the rule of satisfied triangles is valid for all random distributions of spins. We have found a number of situations in which the rule fails, all of them involving rather strong disorder, i.e., clusters of defects. This suggests a generalization of the rule of satisfied triangles to a rule of satisfied clusters. The rule is simply that the ground state energy is the sum of energies of independently satisfied triangles and clusters. Clusters can be identified by calculating the local "triangle frustration energy" which is the difference between the energy of a triangle in a ground state configuration and the minimum possible energy for that individual triangle. Clusters are isolated regions in which the local triangle frustration energy is nonzero.

The rule of satisfied triangles/clusters implies that the ground state energy does not depend on the relative positions of defects or on their degenerate degrees of freedom (such as up or down). This is different from the usual situation in disordered spin systems. Two defects, which introduce frustration into a ferromagnet or antiferromagnet, induce overlapping spin distortions, resulting in an indirect interaction between the defect degrees of freedom and, ultimately, in spin glass behavior [18]. In our system, a defect induces strong local perturbations, but the system is so soft that this perturbation does not generate effective pair interactions between defects. Conventional spin glass behavior cannot arise in this situation. For a low density of vacancies, the ground state degeneracy remains infinite because there are still an infinite number of ways to fold the remaining spins. We note that similar arguments have been made by Villain [18] for the case of the pyrochlore antiferromagnet which he described as a "cooperative paramagnet." The rule of satisfied clusters breaks down when the disorder is so strong that the clusters merge into an infinite cluster. In this situation, spin glass behavior may occur [18]. We also note that weaker interactions which are not included in our model, such as magnetic anisotropy, further neighbor, interlayer, and dipole couplings as well as quantum effects [19], will lead to violations of the rule of satisfied triangles and may be the source of spin glass behavior with a very low T_f. It is also possible that the apparently nonergodic behavior, which is observed experimentally, is simply a property of the pure system at low T, as was proposed in Ref. [17]. If this is the case, then the new type of spin origami described above will be a useful tool for understanding the character of this low temperature state. Conversely we note that the the spin origami mapping seems to contain the essential ingredients for formulating a spin model of tethered membranes of the type studied by Nelson and co-workers [16], thus connecting the Kagomé spin system to a much broader range of physical problems.

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