Orderings of a stacked frustrated triangular system in three dimensions

Daniel Blankschtein, M. Ma, and A. Nihat Berker
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

Gary S. Grest and C. M. Soukoulis
Corporate Research Science Laboratories, Exxon Research and Engineering Company, Annandale, New Jersey 08801

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An Ising system is constructed by stacking frustrated antiferromagnetic triangular lattices. Landau-Ginzburg-Wilson and Monte Carlo analyses suggest two ordered phases. In the low-temperature phase, one sublattice is fully ordered and, oppositely, two sublattices are partially ordered. In the intermediate phase, two sublattices are fully and oppositely ordered, and one is disordered. The transition to the paramagnetic phase is in XY universality. The transition between the ordered phases is due to sixfold symmetry-breaking flop.

The Ising model with antiferromagnetic nearest-neighbor interactions on the triangular lattice is the simplest spin system which is totally frustrated.[1] In each elementary triangle of this lattice, the three antiferromagnetic bonds cannot be simultaneously satisfied and the energy is minimized with any one of the bonds violated. Thus, the entire system has infinitely many ground states, connected by single spin flips made possible by mutually cancelling antiferromagnetic bonds, and remains disordered at all finite temperatures.[2] However, it is rigorously known[2,3] that algebraic order is attained at a zero-temperature critical point, where correlations decay with an inverse power of distance, as opposed to the rapid exponential decay of a truly disordered phase. The many ground states play the same statistical role as the critical fluctuations at an ordinary critical point.

To study the possible finite-temperature ordering of a highly degenerate, yet simply formulated system, we considered antiferromagnetic triangular Ising systems stacked and nearest-neighbor coupled along the z direction. Possible experimental realizations[4] will be discussed below. The Hamiltonian is

$$\mathscr{H} = J\sum_{\langle ij\rangle}^{xy} s_i s_j - J'\sum_{\langle ij\rangle}^{z} s_i s_j \ ,$$

where $J,J'>0$, $s_i=\pm 1$, and $\langle ij\rangle$ indicates summation over nearest-neighbor pairs in the xy plane or along the z direction. This system is fully frustrated in the xy planes, but has no competing interactions along the z direction. Each ground state of the two-dimensional ($d=2$) model corresponds to a ground state of the stacked $d=3$ model, with spins aligned along the z direction, so that the ground-state degeneracy[2] is $\exp(0.323N^{2/3})$, where $N\rightarrow\infty$ is the number of spins. At zero temperature, the magnitude of the spin-spin correlation function in the xy plane is bounded below by that of the $d=2$ model. If we hypothesize ordering, the average energy price of introducing a domain wall at a given xy plane is at least

$$2J\int_0^{N^{1/3}}\frac{2\pi r\,dr}{r^{1/2}}\sim N^{1/2} \ ,$$

using the known $d=2$ power-law decay.[3] This domain wall can occur at any one of $N^{1/3}$ positions along the z direction, so that the entropy gain is $(1/3)\ln N$. Thus, one naively concludes true long-range order at zero temperature, which can be presumed to extend to finite temperatures.

This expectation is indeed fulfilled, as seen in Fig. 1. Specific-heat curves from Monte Carlo simulation of $15\times 15\times L$ systems are shown. The rounded Schottky peak of the $L=1$ system, due to the onset of short-range order in $d=2$, sharpens as L is increased and is consistent with the phase transition cusp discussed below.

The possible onsets of ordering can be deduced from Landau-Ginzburg-Wilson (LGW) theory.[5,6] The Hamiltonian is Fourier transformed:

$$\mathscr{H} = \sum_{\vec{q}} J(\vec{q})s(\vec{q})s(-\vec{q}) \ ,$$

with

$$J(\vec{q}) = J[\cos(q_x)+2\cos(q_x/2)\cos(\sqrt{3}q_y/2)] - J'\cos(q_z) \ ,$$

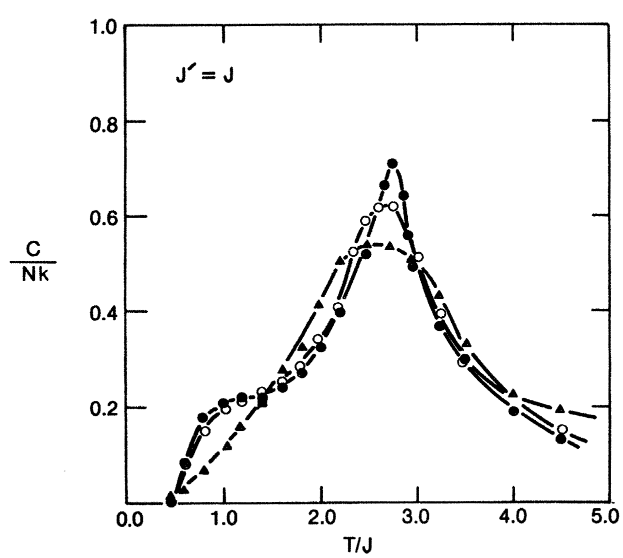

FIG. 1. Specific heats from Monte Carlo simulation, for $J'=J$ and $15\times 15\times L$ systems. The number of layers L is 4(▲), 8(○), and 12(●).

where the sum is over a hexagonal Brillouin zone. The mode(s) with the lowest energy $J(\vec{q})$ is predicted to become critical as the temperature is lowered from the disordered phase. In our case, $J(\vec{q})$ is minimized at the six corners of the Brillouin zone.[6] These correspond to two degenerate modes $\vec{Q}_{\pm}=(\pm 4\pi/3,0,0)$, the other corners being related by reciprocal lattice vectors. A two-component ($n=2$) order parameter is thus revealed. The LGW Hamiltonian is constructed in terms of the critical and nearby modes $\psi_{1,2}(\vec{q})=s(\vec{Q}_{\pm}\pm\vec{q})$, $|\vec{q}|<<1$, by noting all possible invariants under the symmetries of the system, at each consecutive order:

$$\bar{\mathscr{H}}=\frac{1}{2}\sum_{\vec{q}}(r+\vec{q}^2)\psi_1(\vec{q})\psi_2(\vec{q})+u_4\sum_4(\psi_1\psi_2)^2 +u_6\sum_6(\psi_1\psi_2)^3+v_6\sum_6(\psi_1^6+\psi_2^6)\ ,$$

where $\sum_p$ signifies summation over p momentum arguments which add to zero. Since s_i is real, $\psi_{1,2}=m\exp(\pm i\phi)$, yielding

$$\bar{\mathscr{H}}=\frac{1}{2}\sum_{\vec{q}}(r+\vec{q}^2)m^2+u_4\sum_4 m^4 +u_6\sum_6 m^6+v_6\sum_6 m^6\cos(6\phi)\ ,$$

the LGW Hamiltonian of the XY ($n=2$) model with sixfold symmetry-breaking appearing at sixth order. Simple power counting indicates that the sixth-order terms are marginal at $d=3$. Moreover, $d=4-\epsilon$ expansion gives v_6 irrelevant at criticality, with rescaling behavior

$$v_6(l)=v_6(0)e^{-(2+\epsilon)l}\ .$$

Thus, quantitatively, the isotropic XY critical behavior should not be modified much, if at all. Indeed, the specific heat at the transition from the disordered phase is consistent with the cusp of the XY exponent[7] $\alpha\simeq-0.02$.

The microscopic configuration of the ordered phases can be investigated by Fourier transforming the thermal average of the critical modes, $\langle s_i\rangle \sim M\cos(\vec{Q}_+\cdot\vec{r}_i+\Phi)$, where $m=M$ and $\phi=\Phi$ minimize $\bar{\mathscr{H}}$. Accordingly, the ordered phases are characterized in terms of the three sublattices of the triangular xy planes, with translational symmetry along the z direction. For $v_6<0$, $\Phi=0$ and the magnetizations $(M,-M/2,-M/2)$ are assigned to the three sublattices (Fig. 2). For $v_6>0$, $\Phi=\pi/6$, dictating sublattice magnetizations[8] $(\sqrt{3}M/2,-\sqrt{3}M/2,0)$ shown in Fig. 2. The Edwards-Anderson[9] order parameter $Q=N^{-1}\sum_i\langle s_i\rangle^2$ equals $M^2/2$ in either ordered phase. Each of these two ordered phases is sixfold degenerate, as seen by up-down symmetry $M\rightarrow -M$ and the three ways of singling out one sublattice.

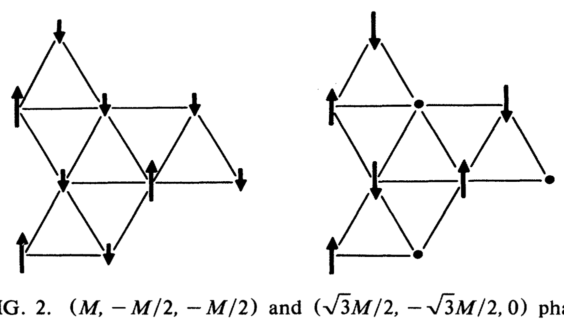

FIG. 2. $(M,-M/2,-M/2)$ and $(\sqrt{3}M/2,-\sqrt{3}M/2,0)$ phases, respectively occurring at low and intermediate temperatures.

The orderings suggested by LGW analysis are indeed revealed by Monte Carlo simulation. These were carried out on $15\times15\times L$ lattices, where $L=4,8,12$ is the number of layers, with periodic boundary conditions. We have performed a number of runs starting from widely different initial configurations to verify final equilibrium. A single-flip sequence of Metropolis dynamics was used, with the trial spin chosen randomly. Typically, 1500–2500 Monte Carlo steps per spin (MCS) were taken and the first 300–500 MCS were discarded. Longer runs were made near the transitions.

Specific-heat curves obtained by cooling the system from a disordered high-temperature configuration are shown in Fig. 1 for $J'=J$. As mentioned above, these data are consistent with a second-order phase transition at $T_c\simeq2.8J$ with a slightly negative exponent. The sublattice magnetizations below T_c indicate the $(\sqrt{3}M/2,-\sqrt{3}M/2,0)$ phase. However, the system moved readily between the six degenerate ordered phases [for example, to $(\sqrt{3}M/2, 0,-\sqrt{3}M/2)$], such interchanges occurring at the time scale of a few hundred MCS. This special feature is explained in terms of the sixfold symmetry breaking of the $d=3$ XY Hamiltonian and the finiteness of the system. If the sixfold symmetry breaking did not occur ($v_6=0$), the system would be equivalent to an XY model. Then the ordered system would indeed be expected to shift between the degenerate phases, corresponding to a global drift in the direction of XY magnetization. This is related to the infinite transverse susceptibility. In fact, this interpretation has been recently given[10] to a similarly shifting $(M,-M,0)$ phase of the $d=2$ triangular Ising model with nearest-neighbor antiferromagnetic and next-nearest-neighbor ferromagnetic interactions. This is applicable with no further caveat in $d=2$, since v_6 is irrelevant for a range of temperatures below the transition.[11] This means that the effective value of v_6 decreases as larger length scales are probed, being zero at the macroscopic scales. The situation is different in our $d=3$ case, where v_6 is nonzero (our simulation indicates $v_6>0$ below T_c) and presumably eventually grows under rescaling inside the ordered phase, dictating the nature of the ordering. However, this should occur only after many rescalings, by continuity of renormalization-group flows, since ϵ expansion suggests an irrelevant v_6 at criticality. Thus, large length scales must be probed to have an effective v_6 strong enough to pin the system to one phase. In sum, our observed shifts can be explained by weak crossover from isotropic XY order in a finite system.

The ordering into the $(\sqrt{3}M/2,-\sqrt{3}M/2,0)$ phase was confirmed by application of a small conjugate field, namely, $H_s=(H,-H,0)$ on the three sublattices. This field rounds and moves the specific-heat peak to higher temperature. The staggered magnetization $M_s=(\langle s_i^{(1)}\rangle-\langle s_j^{(2)}\rangle)/2$, between sublattices (1) and (2), and the Edwards-Anderson order parameter Q clearly exhibit (Fig. 3) the expected behavior in the temperature range $J'<T<2.7J$. The fact that Q levels at 2/3 as M_s approaches 1 shows that sublattice 3 does not freeze.

To study the behavior at the lowest temperatures, we consider small interlayer couplings (Fig. 4), for reasons to be apparent shortly. Another phase transition is now indicated by the peak in the specific heat at temperature T_f of the order of J', below which the system orders into the

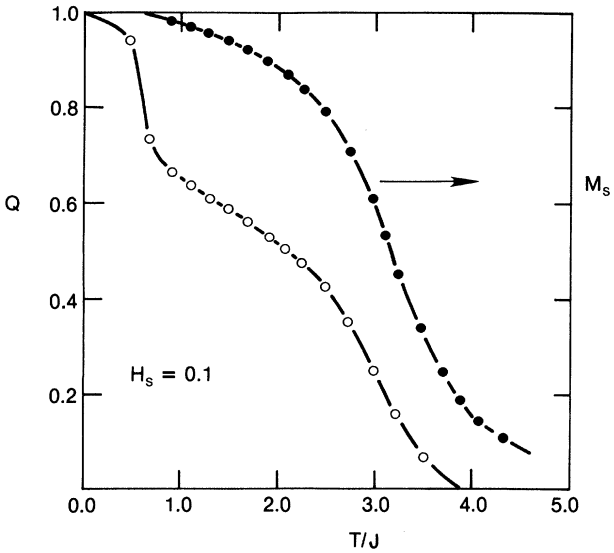

FIG. 3. Staggered magnetization M_s and Edwards-Anderson order parameter Q in presence of the conjugate field $H_s = 0.1$, for $J' = J$ and a $15 \times 15 \times 12$ lattice subjected to 2000 MCS.

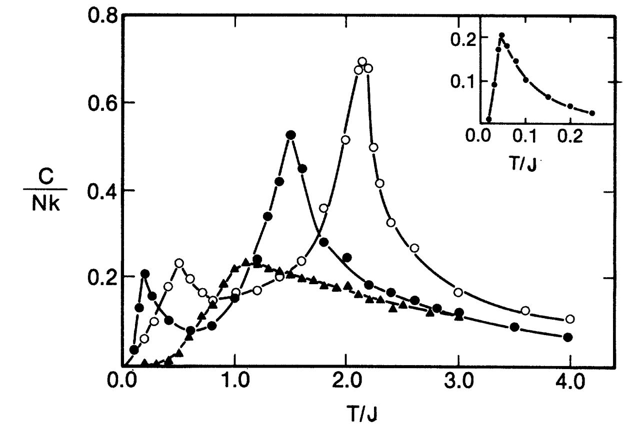

FIG. 4. Specific heats from Monte Carlo simulation of the $15 \times 15 \times 12$ lattice, with $J' = 0(\blacktriangle)$, $0.2(\bullet)$, $0.5(\circ)$, and 0.05 (inset).

$(M, -M/2, -M/2)$ phase. One possible explanation is that the fluctuation renormalized v_6 changes sign as a function of temperature. (This is also consistent with the weak crossover used to explain the finite-system phase shifting at intermediate temperatures.) In that case, the transition is crossing the isotropic, ordered XY phase along a symmetry-breaking direction, so that a first-order transition with a delta-function specific-heat singularity and an infinite transverse susceptibility would be predicted for the ideal system. The above results may be relevant to the experimental system VI_2, which is a stacked frustrated triangular system with weak interplanar coupling.[4] Two consecutive transitions into partially ordered phases have been reported[4] recently for this compound, with the low-temperature transition first order.

As our Monte Carlo runs were taken to near zero temperature, the sublattice magnetizations fell short of saturating to $(1, -\frac{1}{2}, -\frac{1}{2})$. We interpret this as a kinetic effect. Indeed, the low-temperature ordering, which sets at the temperature scale of the interlayer coupling, involves a strong enhancement of the interlayer correlations. (For $T < T_c$, the intralayer energy remains essentially constant at its minimum value of $-J$ per spin.) Strongly correlated columns of spins are sluggish under the single-flip dynamics. This should explain the shoulder and the absence of the low-temperature order in Fig. 1.

To conclude, the large ground-state degeneracy of the stacked triangular antiferromagnetic Ising model is reflected by the intrinsic entropy of the ordered phases: The intermediate-temperature phase has one sublattice totally disordered,[8] and the low-temperature phase has two sublattices half-disordered. This entropy occurs factorized into local units, which are the partially or fully disordered single spins, periodically permeating long-range order. (A similar behavior was seen[12] for antiferromagnetic Potts models, another system with large ground-state degeneracy.) The disordering frustration of the xy planes and the stabilizing third spatial direction conspire to produce here two distinct ordered phases related to the XY model, but intrinsically accommodating a significant amount of local disorder.

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