Critical behavior of a frustrated Ising system

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Finite-size-scaling analysis of a simulation of the critical behavior of a nonrandom three-dimensional frustrated system yields a tricritical-like behavior not predicted by a simple Landau-Ginzburg-Wilson theory. We argue that, because of the frustration, the free energy of many phases is close to that of the observed ordered phase and that a complete theory must take into account interactions between fluctuations of order parameters of all these phases. We suggest that these properties are generic to frustrated systems. Related transitions in the model are also discussed.

Frustrated systems are statistical systems in which it is geometrically impossible for all interactions to be in minimum-energy states. There are two distinct types of systems in which frustration is thought to be important: random frustrated systems, such as spin glasses,[1] and nonrandom frustrated systems. Nonrandom frustrated models are of interest both as simple models containing one of the important features of spin glasses and as models of physical systems, for example, certain magnetic systems,[2] or helical polymers which exhibit crystalline phases with hexagonal packing, such as "smectic" isotactic polypropylene (iPP) or polytetrafluoroethylene (PTFE). In these polymers, the interchain energy between chains of the opposite handedness is different from this energy between chains of the same handedness,[3,4] leading to frustration in a hexagonal packing. Here we will discuss and analyze Monte Carlo results for a simple frustrated three-dimensional model. Based on finite-size-scaling estimates of the critical exponents and a Landau-Ginzburg-Wilson (LGW) argument involving competing order parameters, we show that the frustration appreciably changes the critical behavior (except possibly inaccessibly close to the critical point) from that which would be expected from a simpler LGW treatment, which accounts for frustration only through determination of the critical order parameters.

A classical example of a nonrandom frustrated sytem is the two-dimensional (2D) triangular Ising antiferromagnet (TIAF). This model was solved exactly by Wannier,[5] who showed that it is disordered at all finite temperatures, but has a critical point at $T=0$. Most (but not all) of the configurations present in the state at $T=0$ have three sublattices in which spins on two sublattices point in opposite directions, but the spins on the third sublattice point up or down at random. We will call these configurations "2D Wannier configurations." The entropy of the system is finite at $T=0$, and the Wannier configurations contribute most of this entropy. Whereas a simple LGW analysis suggests that this model should belong to the 2D XY-universality class, the critical behavior is in fact found to be completely different from that of the 2D XY model.[6,7] Not only is the transition at zero temperature, which is not expected from the LGW treatment, the spin-spin correlation function at the transition decays algebraically with distance with an exponent $\nu=\frac{1}{2}$, in contrast to the exponent $\nu=\frac{1}{4}$ obtained for the 2D XY model. It should also be remarked that applying any field which removes the frustration, e.g., infinitesimally weakening any one bond in each elementary triangle, results in a definite ground state. Thus $T=0$, which is the critical point for this system, is in fact an (infinite) multicritical point.

A three-dimensional (3D) analog of this system consists of stacking infinitely many such 2D TIAF's on top of each other and with the spins coupled ferromagnetically in chains along the direction perpendicular to the 2D TIAF's. We take the stacking direction to be the $\hat{\mathbf{z}}$ axis. The Hamiltonian is then taken to be

$$H_0=\frac{1}{2}J_\perp \sum_{\langle i,i'\rangle ,j} \sigma_{i,j}\sigma_{i',j} - J_z \sum_{i,j} \sigma_{i,j}\sigma_{i,j+1} ,$$

where $\sigma_{i,j}=\pm 1$ is the jth "spin," which for helical polymers specifies one of two molecular orientations, along the z axis on the ith chain and the sum over $\langle i,i'\rangle$ runs over nearest neighbors in the xy plane. This is a simple model of, for example, iPP or PTFE. There has recently been interest in this model.[2,8–12] Blankschtein *et al.*[9,10] have performed Monte Carlo simulations of H_0 which show two peaks in the specific heat at temperatures T_{c_1} and T_{c_2} ($T_{c_1} > T_{c_2} > 0$) and they argue from the point of a LGW theory that the system exhibits two ordered phases. They conclude that both phases have a triangular $\sqrt{3}\times\sqrt{3}$ superlattice structure in the xy plane, but the intermediate-temperature (IM) phase ($T_{c_2} < T < T_{c_1}$) has a sublattice magnetization of $(\sqrt{3}M/2, -\sqrt{3}M/2, 0)$, corresponding to critical wave vectors $\mathbf{q}_K=\pm(4\pi/3)\hat{\mathbf{y}}$ at the points K on the corners of the Brillouin zone (or any wave vectors connected to these by reciprocal lattice vectors). The order parameter M thus belongs to the 3D XY-universality class. In contrast, they argue that the sublattice magnetization of the low-temperature (LT) phase ($T < T_{c_2}$) is $(M, -M/2, -M/2)$. Later, Coppersmith[12] showed that Landau-type arguments are unreliable for the low-temperature behavior, and argued from entropy considerations that the LT phase of this system should be the 3D analog of the 2D Wannier phase, with a three-sublattice structure in which all spin chains are fully ordered—two sublattices of spin chains are fully ordered in opposite directions, but the third sublattice has its spin chains ordered in random directions. We will refer to this phase as the 3D Wannier phase.

In this paper, we present results from Monte Carlo simulations combined with finite-size-scaling analysis of the 3D stacked TIAF system. In order to explore the effects of frustration we have also considered the model with a *third* nearest neighbor (3NN) interaction in the xy plane so that

$$H=H_0-J_3\sum_{i,i';j}{}'\sigma_{ij}\sigma_{i'j} ,$$

with the prime indicating that the sum runs over i,i' 3NN. Although we find the same IM phase for $J_3=0$ as Blankschtein and co-workers,[9,10] the critical exponents are very different from those of the 3D XY-universality class, and are close to those of a tricritical point. We show that, because of the frustration, there are many wave vectors at which the inverse susceptibility is close to its value at the critical wave vector $\mathbf{q}_K$, and that there are many phases close in free energy to the frustrated IM phase. We argue that these two facts, which would seem to be generic both to nonrandom frustrated systems and to random frustrated systems, give rise to tricritical behavior. Furthermore, our results for the low-temperature phase support the conjecture of Coppersmith[12] that the ordered phase is the 3D Wannier phase.

The Monte Carlo simulations were performed on systems with $N=L^2L_z=6^3$, 12^3, 18^3, and 24^3 spins with periodic boundary conditions. These sizes are consistent with a variety of low-energy phases. The simulations at constant J_3 were started in random initial configurations at a high temperature and slowly cooled, with a few thousand Monte Carlo steps (MCS's) per spin at each temperature. The first thousand MCS's were discarded in calculating thermodynamic quantities. We have also run the simulations with independent random initial configurations at each temperature, in which case the only difference from the cooldown was a slight instability at the very lowest temperatures. Finally, we also performed a simulation for 1.5×10^4 MCS's to ensure that the system was stable and had equilibrated after 1000 MCS's at $T=2.7J_z$ ($J_\perp=J_z$) on a system of 18^3 spins. We found no indication that this was not the case. To analyze the system, we define the following three order parameters: first

$$M=\frac{\sqrt{6}}{3}(m_1^2+m_2^2+m_3^2)^{1/2} , \tag{1}$$

where

$$m_1=\tfrac{1}{2}[M_1-\tfrac{1}{2}(M_2+M_3)]$$

(and similarly for m_2 and m_3) and M_α is the sublattice magnetization

$$M_\alpha=\frac{3}{N}\sum_{ij}^{\alpha}\sigma_{ij} ,$$

where $\sum^\alpha$ indicates a sum over sites in the α sublattice ($\alpha=1,2,3$) of a $\sqrt{3}\times\sqrt{3}$ superlattice; second

$$\overline{m}=\frac{1}{\sqrt{3}}(\overline{m}_1+\overline{m}_2+\overline{m}_3)^{1/2} , \tag{2}$$

where

$$\overline{m}_\alpha=\frac{3}{N}\sum_i{}^{\alpha}\left|\sum_j\sigma_{ij}\right| ,$$

and third

$$\tilde{m}=\sum_l\left[\tfrac{1}{2}\sum_k\tilde{m}_{kl}^2\right]^{1/2} , \tag{3}$$

with

$$\tilde{m}_{kl}=\frac{2}{N}\sum_{i,j}{}^{k}\sigma_{ij} ,$$

$l=1,2,3$, where the index k runs over the two sublattices of a 2×1 superlattice parallel to a nearest-neighbor direction of the triangular lattice and the index l runs over the three possible such superlattices. The order parameter M is the relevant one for the IM phase, in which M is bounded by unity. The order parameter $\overline{m}$, which is a measure of the degree of ordering of each individual column of spins, is bounded by $1/\sqrt{2}$ in the IM phase. In the LT phase proposed by Blankschtein *et al.*,[9,10] M is bounded by $\sqrt{102}/12\approx0.84$, whereas $\overline{m}$ is bounded by $1/\sqrt{2}$, and, finally, in the Wannier phase, M is bounded by $\sqrt{3}/2$ and $\overline{m}$ is bounded by unity. As the 3NN coupling J_3 increases, we expect the ordered phase of the system to be a two-sublattice system, with spins in alternating rows along a symmetry direction in the xy plane of the lattice aligned in the same direction. The three critical wave vectors $\mathbf{q}_M$ for this are $\mathbf{q}_M=\pm(2\pi/\sqrt{3})\hat{\mathbf{x}}$ at the M points of the Brillouin zone and wave vectors related to $\mathbf{q}_M$ by rotations. This phase, which we will refer to as the J_3 phase, then has a different symmetry than the IM phase, and the order parameter corresponding to this phase is $\tilde{m}$.

For $J_3=0$ and $J_z=J_\perp$, our results are in agreement with those of Blankschtein *et al.*[9,10] in that we find a phase transition at $T_{c_i}\approx2.88J_z$ from a disordered paramagnetic phase to an ordered phase, identical to the IM phase already described. In order to examine the universality class of this transition, we performed finite-size-scaling analyses on the order parameter M, its corresponding susceptibility χ, and the specific heat C. We find the analysis in accordance with the scaling hypothesis (see Fig. 1), and that the critical exponents satisfy the scaling relation $\gamma+\alpha+2\beta=2$. The best values for the critical exponents that we obtained are $\gamma=1.15\pm0.05$, $\alpha=0.5\pm0.1$, and $\beta=0.19\pm0.1$, in disagreement with the exponents for the XY-universality class but very close to the exponents at a tricritical point. Note that as the divergent part of the specific heat above the transition is comparable to that below the transition, the term ($\propto M^6$) in a tricritical free energy would have to be large. In consequence, large logarithmic corrections would be expected, and the differences between these exponents and the (exactly known) three-dimensional tricritical exponents is not surprising. We also verified that the values of the exponents did not vary appreciably for different ratios of the coupling constants, $J_\perp/J_z=\frac{1}{2}$ and $J_\perp/J_z=2$, so that this behavior appears to be universal. Below T_{c_1} and for $J_3=0$, we observed large fluctuations in the order parameter $\tilde{m}$, suggesting that the J_3 phase is close to the IM phase in free energy.

In an attempt to understand this behavior, we calculated the structure factor $S(\mathbf{q})$ at $J_3=0$ and at a temperature slightly above T_{c_1} (Fig. 2). In Fig. 2, we see the two rather narrow, sharp peaks of the IM phase at the corners of the Brillouin zone and a high flat plateau along the zone edge. This line of scattering as a function of wave vector suggests quasi-two-dimensional behavior, as do the critical exponents which are in the correct range for two-dimensional critical exponents. However, such behavior is inconsistent with the fact that the lines of large $S(\mathbf{q})$ are in three different directions in the xy plane. Furthermore, we have for $J_\perp=J_z$ calculated ν by superimposing the finite-size-scaling analyses for different sizes. The result $\nu=1/2.11$ is consistent with hyperscaling ($d\nu=2-\alpha$) only in three dimensions. We note that the large amount of scattering close to the critical wave vectors could simply imply that the actual critical behavior can be observed only in very large systems and only very near the critical point. In addition, there are a number of Umklapp contributions to fourth-order interactions between order parameter fluctuations close to the

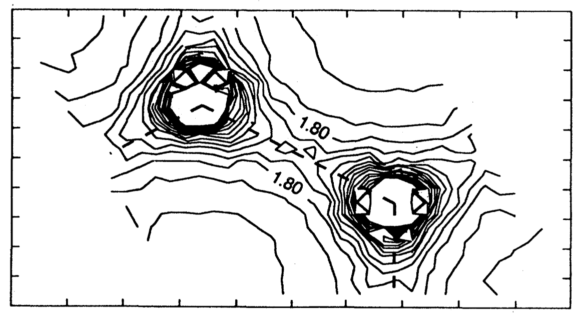

FIG. 2. Contour of $S(\mathbf{q})$ at $q_z=0$ at an increment of 0.6 between the contour lines. The peaks at $\mathbf{q}_K$ are truncated at $S(\mathbf{q})=100$. The dashed line indicates the boundary of the first Brillouin zone.

critical wave vector [at *inter alia* $(\frac{3}{4})\mathbf{q}_K$] which are not possible at $\mathbf{q}_K$. However, we see no appreciable variation in the exponents even for our largest simulation.

This scattering also suggests a reason for the transition to be tricritical. It is well known[13] that if the square of an order parameter φ can couple linearly to a field ρ, then the transition will be more tricritical, i.e., the free energy density

$$F=r\varphi^2+u\varphi^4+w\rho^2+g\rho\varphi^2$$

is just $r\varphi^2+u'\varphi^4$ with $u'=u-g^2/4w$, when ρ has been eliminated from the free energy. The large value of $S(\mathbf{q})$ near the M points $\mathbf{q}_M$ implies that if ρ is taken to be the square of $\sigma(\mathbf{q})$ with $\mathbf{q}$ near $\mathbf{q}_M$, then u, g, and w are all expected to be of order unity so that u' might well be small, leading to tricritical behavior. Similar effects have been hypothesized in a variety of other systems. In order to test this hypothesis we applied a finite 3NN coupling J_3 and performed simulations at a constant temperature while increasing J_3. For each value of J_3, the simulations were started with random initial configurations. We also performed simulations at a constant J_3 while decreasing the temperature. Positive J_3 reduces the frustration and increases the energy of states with finite $S(\mathbf{q}_K)$ and decreases those with finite $S(\mathbf{q}_M)$, and thus is expected to further increase w and further decrease u', leading to a first-order transition. Note that this also relieves the frustration. Simulations show that for J_3 near zero, the transition temperature T_{c_1} decreases with increasing J_3, and for J_3 small and positive ($J_3\approx0.08$ for $J_\perp/J_z=1$), there is an apparent first-order transition to the J_3 phase. The transition temperature to this phase depends only weakly on J_3 for larger J_3 and there is an additional (first-order) transition between the two ordered phases which, close to the transition to the disordered state, is essentially at constant J_3. Since the critical value of J_3 is essentially independent of system size, the $J_3=0$ critical point is not a multicritical point for which any interaction reducing the frustration and yielding a unique ground state is a relevant operator, unlike the 2D case. This behavior is consistent with the above-mentioned argument for tricriticality only if there is a critical endpoint

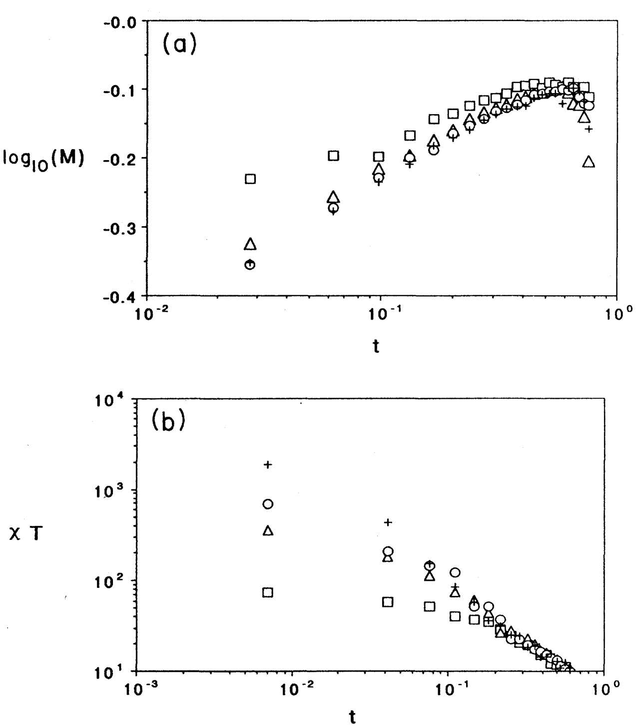

FIG. 1. Order parameter M [part (a)] and χT [part (b)] vs reduced temperature t for $N=6^3$ (□), $N=12^3$ (△), $N=18^3$ (○), and $N=24^3$ (+).

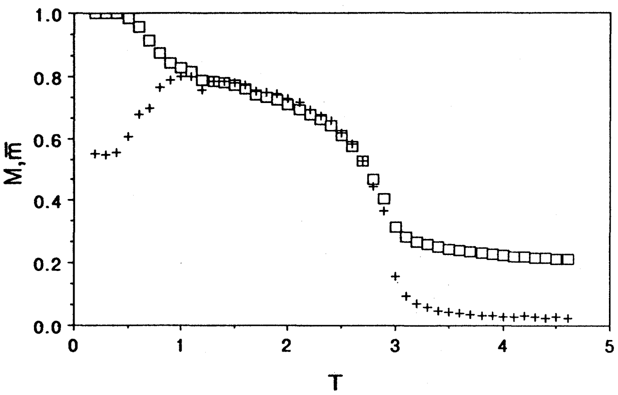

FIG. 3. Order parameters M (+) and $\overline{m}$ (□) vs T ($N=24^3$).

close to the tricritical point, as this first-order transition is to a completely different state than that found for $J_3=0$. Thus we have not achieved a direct test of our tricritical hypothesis. However, the sharp bend in the transition line to the J_3 phase, which coincides within our precision with this critical endpoint, and the fact that it occurs for rather small J_3 clearly confirm the existence of strongly interacting and competing order parameters. It is in fact reasonable to expect that we are close to two tricritical points, one for the disordered IM phase transition made tricritical by interactions with the order parameter $\overline{m}$, and one for the disordered J_3 phase transition made nearly first order by interactions with the order parameter M. While we have not explored this possibility, these Monte Carlo results clearly establish that, while the interactions in 3D between order parameters do not seem, as in two dimensions, to change the critical point into a multicritical point, they do strongly affect the behavior near the critical point and lead to nearby multicritical points.

In order to establish the nature of the LT phase, we performed simulations to temperatures as low as $T=0.2J_z$. In Fig. 3 the order parameters M and $\overline{m}$ are depicted. In this figure, we see that the values of these order parameters are incompatible with the bounds given above for the LT phase proposed by Blankschtein *et al.*[9,10] We also see that the order parameter $\overline{m}$ saturates to unity at $T\approx 0.4J_z$, which means that all spin chains are fully aligned. These facts and inspection of the obtained phase are consistent with the LT phase being the Wannier phase. Simulations at finite uniform magnetic fields, both at fixed fields and at fixed temperature, also indicate that the same LT phase persists for small magnetic fields at the lowest temperatures. Thus our simulations support the conjecture by Coppersmith[12] that the LT phase is the Wannier phase. This phase transition then has the unusual property that it does not involve a change in symmetry, but only a change in structure, since what happens is that the disordered spin chains on one sublattice align, but in random directions. Of course, the simulations are not conclusive evidence that indeed there is a real phase transition at T_{c_2}, but the results from the finite-field simulations indicate that the LT phase does occupy a finite region in the phase diagram, consistent with a real phase transition.

In conclusion, we first find that although a simple LGW theory does predict the correct structure of the intermediate-temperature phase, the critical exponents that we obtain from finite-size scaling are not in agreement with those of the XY-universality class predicted from the LGW theory. Analysis of the structure factor $S(\mathbf{q})$ suggests that *all* wave vectors on the Brillouin zone edge have direct importance to a Landau-type theory, except very close to the transition. In addition, we have argued that these nearly critical modes will have the effect of a strongly fluctuating background field, inducing tricritical-like phenomena. Given this reasoning for the complex critical behavior of the present system, we expect complex critical behavior in frustrated systems in general. In such systems, by relieving the frustration it is always possible to find phases close in free energy to the frustrated phase, for example, the J_3 phase in the present system. The critical behavior of the actual order parameter is then expected to be strongly influenced by its interactions with the large fluctuations in various other order parameters. So although a LGW analysis may yield the correct order parameter for the frustrated phase, the critical behavior is not determined by this order parameter alone. Additionally, the results of our simulations at the lowest temperatures are consistent with the LT Wannier phase suggested by Coppersmith.[12]

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[1]For a comprehensive review, see K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1986).
[2]See, for example, K. Hirakawa, H. Kadowaki, and U. Ubukoshi, J. Phys. Soc. Jpn. **52**, 1814 (1983).
[3]R. A. DeFazio, P. L. Taylor, and N. F. Wright, Bull. Am. Phys. Soc. **33**, 248 (1988).
[4]B. L. Farmer and R. K. Eby, Polymer **26**, 1944 (1985).
[5]G. H. Wannier, Phys. Rev. **79**, 357 (1950).
[6]J. Stephenson, J. Math. Phys. **11**, 420 (1970).
[7]For a review of frustrated Ising lattices, see R. Liebman, *Statistical Mechanics of Periodic Frustrated Systems* (Springer, New York, 1986).
[8]F. Matsubara and S. Ikeda, Phys. Rev. B **28**, 4064 (1983).
[9]D. Blankschtein *et al.*, Phys. Rev. B **29**, 5250 (1984).
[10]A. N. Berker *et al.*, J. Appl. Phys. **55**, 2416 (1984).
[11]E. Dubois-Violette, R. Julien, and J. P. Carton, J. Phys. (Paris) **45**, 1297 (1984).
[12]S. N. Coppersmith, Phys. Rev. B **32**, 1584 (1984).
[13]For example, the nematic–smectic-*A* phase transition in liquid crystals. For a recent review, see L. Longa, J. Chem. Phys. **85**, 2974 (1986).