Monte Carlo Mean-Field Theory and Frustrated Systems in Two and Three Dimensions

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(Received 26 October 1990)

A new method, combining mean-field and Monte Carlo approaches, is applied to frustrated Ising systems in $d=2$ and 3, in zero and nonzero uniform fields. The method brings to mean-field theory the hard-spin condition and uses much less sampling than the Monte Carlo simulation. The phase diagram of the $d=2$ triangular antiferromagnet is easily obtained with remarkable global quantitative accuracy. The phase diagram of the $d=3$ stacked triangular antiferromagnet shows three ordered phases, in a new multicritical topology of lines of *XY*, Ising, and three-state Potts transitions, accessible to experiments with layered magnets.

PACS numbers: 75.25.+z, 05.50.+q, 64.60.Cn, 75.50.Lk

The antiferromagnetic Ising model on the triangular lattice is the simplest fully frustrated system and has been shown[1] to exhibit no ordering at temperatures $T>0$ and a critical point at $T=0$. The three-dimensional model formed by stacking such triangular antiferromagnetic Ising models is of interest due to experimental realizations[2] and novel phases,[3,4] partially ordered in accommodating the entropy dictated by frustration.

The Landau-Ginzburg-Wilson (LGW) theory[4] reveals two possible ordered phases, $(M,-M,0)$ and $(M,-M/2,-M/2)$ as labeled by the relative magnetizations of the three triangular sublattices. A Monte Carlo simulation[4] has indicated that, in $d=3$, these two phases are consecutively encountered as the temperature is lowered. However, the status of these phases and phase transitions is far from resolved: (i) LGW theory ignores the hard-spin condition,[4(b)] $s_i^2=1$ at each site i, whereas this condition is at the heart of the energy cancellation, always present at microscopic localities, defining frustration. (ii) LGW theory is self-consistent only for small M and thus can, at best, be suggestive for an explanation of the change of ordering at low temperature. (iii) Coppersmith[5] has raised the possibility that the ordering does not saturate at $T=0$ (i.e., $M<1$). This needs further exploration. (iv) Finally, renormalization-group analysis[4] has indicated that the transition from the disordered phase, if not first order, should have the critical exponents of the $d=3$ *XY* model. However, recent Monte Carlo simulations point to tricritical exponents.[6]

Accordingly, we have developed a new method, merging mean-field theory and Monte Carlo sampling, that addresses all of the above points and yields new information, the finite-field phase diagram. This "Monte Carlo mean-field theory" is of general applicability to statistical mechanics. It improves quantitatively the mean-field solutions of general spin models, while using a substantially smaller number of samplings than a conventional Monte Carlo simulation. It brings the hard-spin condition to mean-field theory, and therefore is appropriate for problems involving frustration. Indeed, we demonstrate here that the method correctly distinguishes the different consequences of frustration in different dimensions (giving, at zero fields, no $T>0$ phase transition in $d=2$ and actual $T>0$ phase transitions in $d=3$, for the frustrated triangular systems here). As such, the method is a qualitative improvement in the self-consistent treatment of frustrated systems.

For an Ising spin system with nearest-neighbor interactions, $\beta\mathcal{H}=J\sum_{\langle ij\rangle}s_is_j$, where $s_i=\pm1$ at each site i of the lattice and $\langle ij\rangle$ indicates summation over all nearest-neighbor pairs of sites, the conventional mean-field equations for individual spins are

$$\langle s_i\rangle=\tanh(H_i)\,, \tag{1}$$

with the effective field $H_i=-J\sum_j\langle s_j\rangle$, where the summation is over sites j nearest neighbor to site i. For triangular-lattice ($d=2$) antiferromagnets, $J>0$, these individual equations have the solution $\langle s_i\rangle_a=-\langle s_j\rangle_b=M$, $\langle s_k\rangle_c=0$, and all permutations, where (a,b,c) denote the three sublattices. The free energy is in fact minimized by this solution of the set of the coupled individual equations, with M becoming nonzero via a second-order phase transition at temperature $T\equiv J^{-1}=3$, clearly violating the exact result[1] of no phase transition for $J<\infty$ in $d=2$.

Any statistical mechanical treatment of these frustrated systems that predicts ordering in $d=3$, to be seriously taken, must conversely yield no ordering in $d=2$. As seen above, conventional mean-field theory fails this criterion. The reason is that in the actual system, the spin s_i, the thermal average of which is evaluated in Eq. (1), never actually feels the field H_i constructed with *the average values* of the nearest-neighbor spins. These spins s_j always have *unit magnitude* and it is their sum that enters the actual field on s_i,

$$H_i=-J\sum_j s_j,\quad s_j=\pm1\,. \tag{2}$$

For antiferromagnetic local correlations, H_i can be strictly zero no matter how large J is, so that s_i would be disordered. Many such frustrated local configurations are lost in conventional mean-field theory (for example,

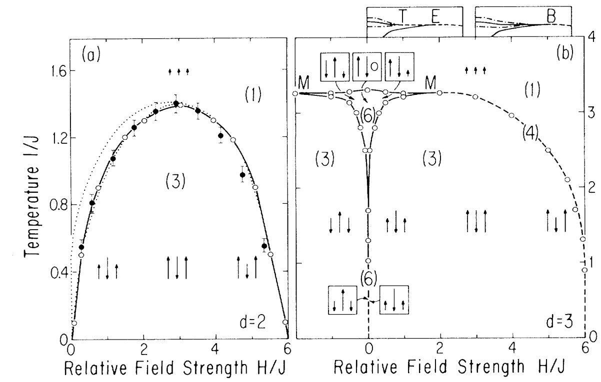

FIG. 1. Phase diagrams: (a) the $d=2$ triangular antiferromagnetic Ising model. The open circles and solid curves are Monte Carlo mean-field theory, using up to 24×24 spins and 50–300 MCS after discarding 150–300 MCS. The entire $T>0$ phase boundary is second order. The solid circles are the conventional Monte Carlo simulation results with 99×99 spins and 2000 MCS after discarding 50 MCS [Ref. 7(a)]. The dotted curves are renormalization-group results [Ref. 7(b)]. (b) The $d=3$ stacked-triangular antiferromagnetic Ising model. The open circles and solid (second-order phase boundary) and dashed (first-order) curves are Monte Carlo mean-field theory, using up to $24\times24\times6$ spins and 50–500 MCS after discarding 100–550 MCS. Given in parentheses are the numbers of coexisting phases. Insets: Possible structures at the multicritical region M; (left) a tricritical point T with a critical end point E, or (right) a bicritical point B. Outside the crossover boundaries (shown schematically by the dash-dotted curves), tricritical or bicritical exponents will be observed.

when $M=0$ is assigned to one sublattice).

We have introduced the Monte Carlo mean-field theory in order to satisfy this basic requirement. The thermal average of each spin s_i is calculated individually, by Eq. (1), in its nearest-neighbor field H_i given by Eq. (2). The nearest-neighbor field is constructed, as specified in Eq. (2), with unit-length spins $s_j=\pm1$, the sign of which is determined by stochastic sampling, namely, by the sign of $\langle s_j\rangle - r$, where r is a random number in the interval $[-1,1]$.

Systems with periodic boundary conditions with sizes up to 24×24 and $24\times24\times6$ were used. The results were insensitive to sequential or parallel updating where, respectively, a randomly chosen spin is updated one at a time or all the spins are updated simultaneously. The former was adopted for computational efficiency. The self-consistency in Eqs. (1) and (2) was typically achieved after 50 samplings per spin (MCS), up to noise due to the random number entering our procedure. Values then needed averaging typically over 100 MCS.

Our calculated phase diagram is given in Fig. 1(a) for the $d=2$ triangular Ising antiferromagnet, also including the possibility of a uniform external field H so that Eq. (2) is supplemented as $H_i=-J\sum_j s_j + H$. Previous results, from a Monte Carlo simulation[7(a)] and renormalization-group calculations,[7(b)] are also shown. It is seen that the Monte Carlo mean-field approach yields the zero-temperature phase transition of $H=0$ and, over the entire phase diagram, excellent agreement with the previous works. Figure 2(a) shows one of the constant-temperature scans used in determining the phase diagram. No hysteresis was observed. The transitions appear to be second order (and are known[7(b)] to be so).

The $d=3$ stacked-triangular antiferromagnetic Ising model in a field has the Hamiltonian

$$\beta\mathcal{H} = J\sum_{\langle ij\rangle} s_i s_j - J'\sum_{\langle ij\rangle}{}' s_i s_j - H\sum_i s_i ,$$

where $J,J'>0$, the first and second sums are over nearest-neighbor pairs in the x-y planes and along the z direction, respectively. This system is fully frustrated in the x-y planes, but with no competing interactions along the z direction. An energy-versus-entropy argument[4(a)] indicates that long-range order should occur at $T>0$. Monte Carlo mean-field theory is done with $H_i = -J\sum_j s_j + J'\sum_j' s_j + H$. Results for $J'=J$ will be reported here.

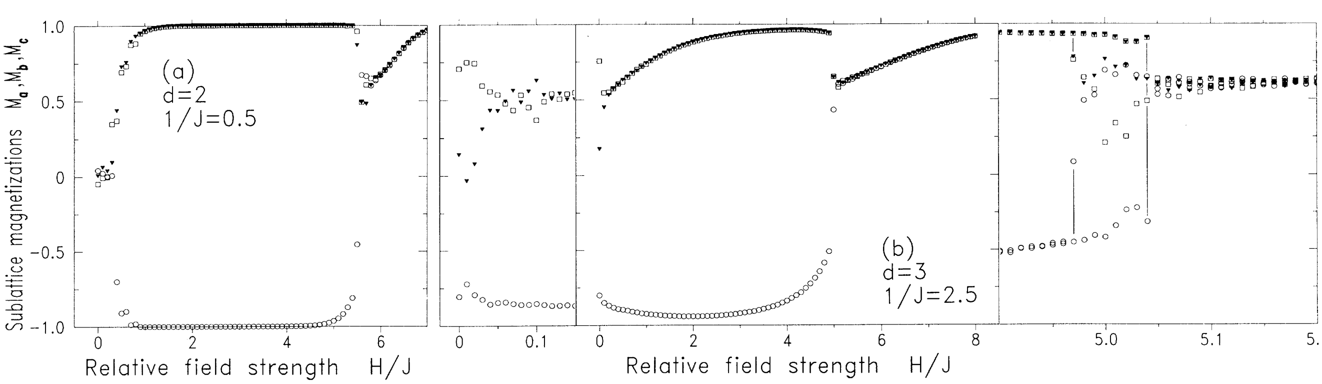

FIG. 2. Monte Carlo mean-field scans: (a) With 24×24 spins and 100 MCS after discarding 200 MCS. (b) With $24\times24\times6$ spins, 100 MCS after discarding 200 MCS (central and left panels), and 200 MCS after discarding 400 MCS (right panel). The right panel is the superposition of two scans in opposite directions, and shows hysteresis, signaling a first-order transition.

Our calculated phase diagram for this $d=3$ system is shown in Fig. 1(b). First, we discuss the $H=0$ line. Two different ordered phases occur, as seen from the sublattice magnetization data in Fig. 3(a). The intermediate-temperature ordered phase has sublattice magnetizations[3,4] $(M,-M,0)$. This phase is sixfold degenerate,[4] as obtained from the sublattice permutations. Note in Fig. 3(a) switching between the degenerate phases (e.g., at $J^{-1}\approx 2$). This switching has been explained[4(b)] by renormalization-group trajectories which, in a finite system, stop before reaching asymptotic limits. The fact that, here, switching is seen at the higher temperatures supports this previous argument. The low-temperature ordered phase has sublattice magnetizations $[M_a,M_b,-(M_a+M_b)]$, with $M_b\approx M_a$, also sixfold degenerate[4] if the equality holds. This phase kinetically freezes when the chains along the z direction freeze predictably at $J^{-1}\sim 2/\ln(N_sN_z)$, where N_s is the MCS number and N_z is the number of spins along z. The histograms at the left of Fig. 3(a) show the sublattice magnetizations at freezing for twenty separate calculations. Thus, the possibility of unsaturated magnetization[5] $(M_a,M_b\neq\frac{1}{2},\ M_c<1)$ remains open. The upper phase transition occurs at temperature $J^{-1}\approx 3.3$, somewhat higher than the extensive Monte Carlo[4(b)] transition temperature of $J^{-1}\approx 3.0$, but considerably less than the conventional-mean-field transition temperature of $J^{-1}=5$. This is because Monte Carlo mean-field theory properly accounts for the bonds violated due to frustration. The upper phase transition in Fig. 3(a) is clearly second order and should be[4] in the XY universality class. We tentatively interpret the lower phase transition in Fig. 3(a) as first order (while another possibility is a narrow transition region between the two ordered phases, which would constitute yet another ordered phase). Figure 3(b) shows the $H=0$ specific heat, calculated from $C=k\langle(\beta\mathcal{H}-\langle\beta\mathcal{H}\rangle)^2\rangle$ by Monte Carlo mean-field theory, with $24\times24\times6$ spins and 500 MCS after discarding 200 MCS. Also shown are the results of the conventional Monte Carlo simulation,[4(a)] with $15\times15\times12$ spins and 1500–2500 MCS after discarding 300–500 MCS. Agreement is satisfactory.

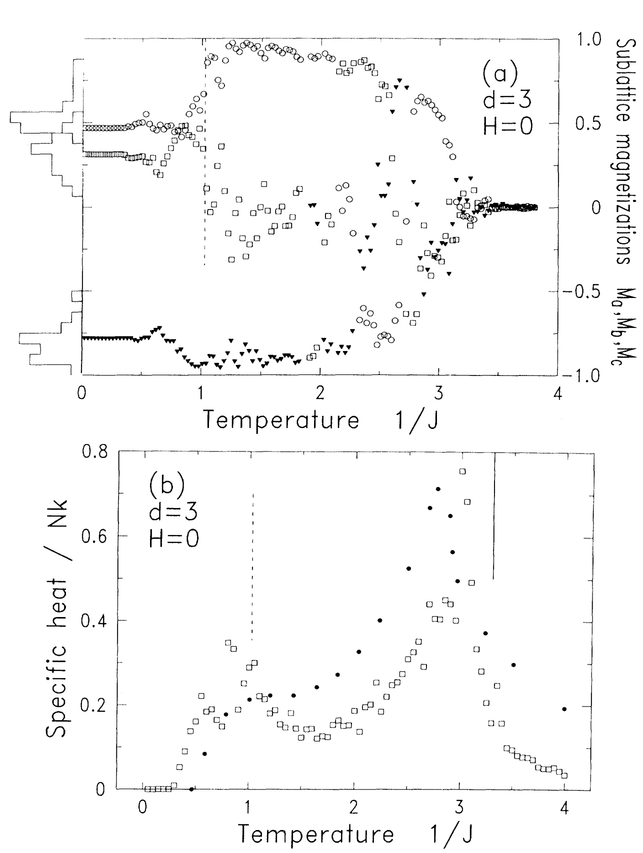

FIG. 3. (a) Sublattice magnetizations by Monte Carlo mean-field theory with $H=0$, using $24\times24\times6$ spins and 500 MCS after discarding 100 MCS. The dashed line indicates the surmised first-order transition between the ordered phases, affected by nearby tricritical points. (b) Specific heat by Monte Carlo mean-field theory (squares) with $H=0$, using $24\times24\times6$ spins and 500 MCS after discarding 200 MCS. The lines indicate the phase transitions deduced from the sublattice magnetizations in (a). The circles are the conventional Monte Carlo simulation results with $15\times15\times12$ spins and 1500–2500 MCS after discarding 300–500 MCS [Ref. 4(a)].

The intermediate-temperature ordered phase $(M,-M,0)$ smoothly continues from $H=0$ to $H\neq0$, for example, as $(0<M_a,M_b<-M_a,0<M_c<M_a)$ for $H>0$. Conclusions of previous LGW and ($\epsilon=4-d$ expansion) renormalization-group theory[4] extend to the boundary between this phase and the high-temperature disordered phase $(M_a=M_b=M_c)$, indicating the XY universality class. As H is increased, the (sixfold-degenerate) intermediate-temperature phase undergoes a second-order transition, as seen in Fig. 2(b), to the threefold-degenerate $(0<M_a,M_b\neq M_a,M_c=M_a)$ phase. Thus, this phase transition involves the breaking of the $M_a=M_c$ symmetry (as H is lowered) and therefore is in the Ising universality class. Based on our tentative identification as first order of the low-temperature $H=0$ phase transition, this $H\neq0$ Ising transition line should be converted from second order to first order by a tricritical point, located close to $H=0$ in order to give the fluctuations observed at the low-temperature $H=0$ transition in Fig. 3(a). The $(0<M_a,M_b\neq M_a,M_c=M_a)$ phase joins along the low-temperature segment of $H=0$ with its $H<0$ counterpart $(M_a<0,M_b\neq M_a,M_c=M_a)$. This segment is thus a first-order phase boundary with sixfold coexistence, in agreement with the $H=0$ discussion above.

Figure 2(b) shows a constant-temperature scan in $d=3$. The high-field transition is first order, as seen from the hysteresis shown in the detailed right panel. This is as expected, since it involves the breaking of the $M_a=M_b=M_c$ symmetry, thereby being in the three-state Potts universality class. [In Fig. 1(a), the entire

$T>0$ phase boundary is in the three-state Potts universality class, which exhibits a second-order transition in $d=2$ but not in $d=3$.] This suggests that the intersection M of the phase boundaries is either a bicritical point or a tricritical-point, critical-end-point combination [Fig. 1(b), insets]. By general renormalization-group theory, phase boundaries and crossover boundaries join at a multicritical point along the same direction corresponding to the smallest relevant scaling exponent. Furthermore, by analyticity of the phase boundary (i.e., analyticity along an irrelevant direction) and by up-down symmetry, both of these boundaries are perpendicular to the $H=0$ line. Thus, the expected crossover lines are as shown in Fig. 1(b), insets. Accordingly, the proximity of this multicritical region to $H=0$ explains the exponents in recent Monte Carlo simulations:[6] The XY exponents[8(a)] ($\nu \approx \frac{2}{3}$, $\alpha \lesssim 0$, $\beta \approx \frac{1}{3}$, and $\gamma \approx \frac{4}{3}$) should be observable only inside the narrow region bounded by the crossover lines, whereas multicritical exponents are to be observed outside the crossover lines. A bicritical point corresponds to crossing a Heisenberg critical point along the anisotropy field direction,[9(a)] so that combining the Heisenberg exponents[8(b)] with the anisotropy crossover exponent[9(b)] $\phi \approx 1.25$ gives $\nu \approx 0.57$, $\alpha \approx 0.29$, $\beta \approx 0.29$, and $\gamma \approx 1.12$. A tricritical point has $\nu = \frac{1}{2}$, $\alpha = \frac{1}{2}$, $\beta = \frac{1}{4}$, and $\gamma = 1$. The latter exponents appear somewhat more consistent with the measured[6] values of $\nu \approx 0.47$, $\alpha = 0.5 \pm 0.1$, $\beta = 0.19 \pm 0.1$, and $\gamma = 1.15 \pm 0.05$. These authors[6] have also explained their data by the proximity of a tricritical point, but by extending the phase diagram toward an ordered phase which is entirely different from ours and not accessible to experiment.

The stacked triangular system is a possible model for magnetic halides. The $H=0$ orderings appear consistent with experiments on $CsCoBr_3$ [Ref. 2(a)], $CsCoCl_3$ [Ref. 2(b)], and VCl_2, VBr_2, VI_2 [Ref. 2(c)]. The present work indicates a new multicritical phase diagram at $H \neq 0$. This should be accessible to experiments, by application of an external field to previously studied materials. Furthermore, in samples with a threshold amount of random bonds due to doping or quenched vacancies, the $d=3$ three-state Potts transitions would show novel tricritical and critical phenomena.[10]

Support by an Evangelisches Studienwerk Fellowship is gratefully acknowledged by R.R.N. This research was supported by NSF Grant No. DMR-87-19217 and by Joint Services Electronics Program Contract No. DAAL 03-89-C0001.

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