First-order phase transition in the fcc Heisenberg antiferromagnet

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The nature of the phase transition of the nearest-neighbor Heisenberg antiferromagnet on a fcc lattice is studied by means of Monte Carlo simulations. Using a finite-size scaling, the transition is shown to be unambiguously of first order. Evidence for a first-order transition in the corresponding XY model is also presented.

Phase transitions of various antiferromagnets on a face-centered cubic (fcc) lattice have attracted both experimental and theoretical interest. Examples are type I antiferromagnet UO_2 (Ref. 1) and type II antiferromagnets MnO (Ref. 2), CeSe, and CeTe (Ref. 3): the type I ordering is specified by the three [1,0,0] wave vectors, while the type II ordering is characterized by the four $[\frac{1}{2},\frac{1}{2},\frac{1}{2}]$ wave vectors. Experimentally, UO_2 and MnO are known to undergo a first-order transition (Refs. 1 and 2, respectively). The first-order nature of the transition in these materials was explained by Bak, Krinsky, and Mukamel[4] and by Brazovskii, Dzyaloshinskii, and Kukharenko[5] as a fluctuation-induced first-order transition within the framework of the Ginzburg-Landau-Wilson Hamiltonian combined with the ϵ-expansion analysis. On the other hand, Ott, Kjems, and Hulliger[3] found by neutron scattering that the transition of CeSe and CeTe is continuous, in apparent contradiction with the ϵ-expansion prediction.

Extensive theoretical studies have also been made for localized spin models on a fcc lattice. In particular, the fcc antiferromagnetic Ising model with nearest-neighbor (NN) interaction was studied in detail by several authors.[6-8] There appears a consensus among the authors that the transition in the Ising case is of first order, the associated spin ordering being type I. In the case of classical Heisenberg spins on a fcc lattice, it has been shown[9] that the ground-state degeneracy is infinite due to the frustration just as in the case of the fully frustrated simple cubic lattice.[10] However, the nature of the transition in the fcc Heisenberg antiferromagnet remains less clear. Fernandez *et al.*[11] performed Monte Carlo (MC) simulations for the NN classical Heisenberg fcc antiferromagnet and concluded that the transition is continuous, in contrast with the Ising case. On the other hand, Henley[12] applied a spin-wave analysis to the same system and found that the ordered-state spin configuration at finite temperature is collinear with four-sublattice periodicity (ordering of type I). This collinear configuration is selected out of the ground-state infinite degeneracy by thermal fluctuations, a mechanism which was called ordering by disorder.[13] Henley[12] then suggested that the associated phase transition is of first order since the ordered state has a threefold degeneracy (Z_3) reminiscent of the three-state Potts model.

In this paper, we study by systematic MC simulations the NN classical Heisenberg fcc antiferromagnet with the aim of determining the order of the phase transition in an unambiguous manner. For this purpose, we consider a three-component ($n=3$) classical Heisenberg model with the Hamiltonian

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

where $\mathbf{S}_i$ is a three-component spin of unit length, J is negative, and the sum runs over all NN pairs. In the following, physical quantities are expressed in units of $|J|$.

Extensive MC simulations have been carried out for the lattice sizes $N=4L^3$ with $L=6$, 8, 10, 12, and 16. Periodic boundary conditions have been used. The MC procedure is the multiflipping method proposed by Creutz[14] in which several flipping trials are attempted at each spin under the same local field before moving to another spin. The convergence to equilibrium is faster than the standard single-spin flipping for a given CPU time. In each run, N_1 MC flipping trial steps (MCS's) per spin were discarded to equilibrium the system before averaging physical quantities over N_2 MCS/spin. In some cases, we used $N_1=10^5$ and $N_2=75\times10^3$, and in general $N_1=N_2=25\times10^3$ were employed. The initial spin configuration used in each run is the collinear ordered state of type I. We have also used the random initial states for comparison. We will return to this point later. Various physical quantities have been calculated, but the main results shown in the following are the internal energy per spin U and the specific heat per spin C calculated from thermal fluctuations of U. In order to pinpoint the transition temperature T_c, a very small interval of successive temperatures $\Delta T=0.0001$ was used.

It has been suggested[15,16] that in a first-order transition, the maximum of the specific heat, $C_{\max}$, is proportional to the system volume, i.e., L^3. However, to our knowledge, except for the works by Challa *et al.*[15] and Binder and Landau,[16] there has not been another numeri-

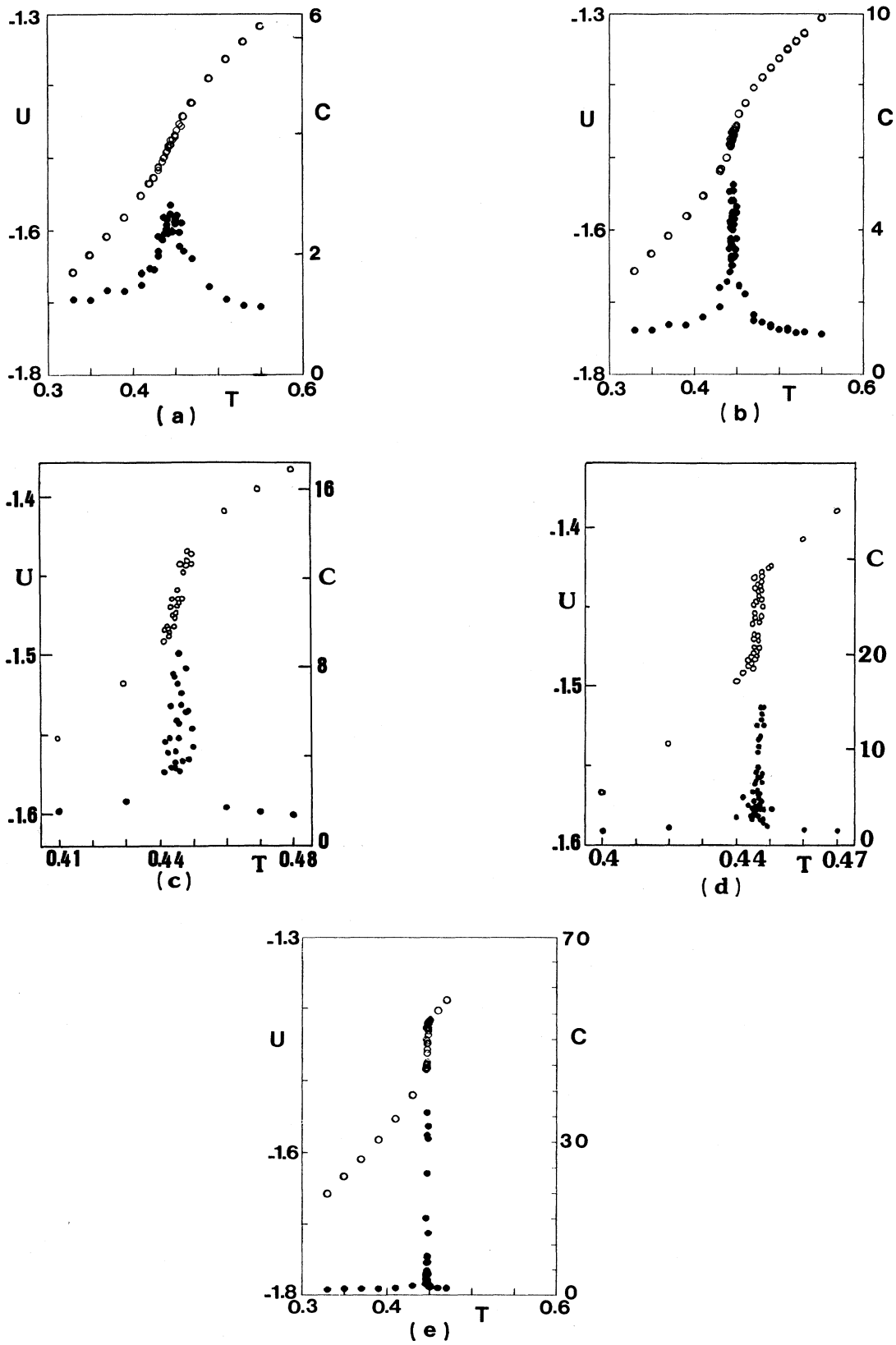

FIG. 1. Internal energy per spin U (open circles, left-hand side) and specific heat per spin C (solid circles, right-hand side) vs temperature T near the transition point, in units of $|J|=1$ and k_B (Boltzmann constant)$=1$, for various lattice sizes L: (a) $L=6$, (b) $L=8$, (c) $L=10$, (d) $L=12$, and (e) $L=16$. Raw data obtained from several independent runs are shown together in each figure. Note the difference in the scales in each figure.

cal scaling study for a first-order transition. Indeed, time-consuming calculations are required (long runs and very small ΔT) if one wishes to get an accurate finte-size scaling. This work thus provides another example of finite-size scaling for a first-order transition.

Figure 1 shows C and U as a function of temperature T for various L. For some sizes, up to 64 temperatures near T_c have been studied. We have made several independent runs with different sequences of random numbers to check the stability of the results. No appreciable hysteresis has been observed through heating and cooling procedures (for cooling, random initial spin configurations were used). For larger sizes, C is impressively δ-function-like and U is almost discontinuous.

The temperature T_c corresponding to the maximum of C is rather insensitive to the lattice size, as expected in first-order transitions. One has $T_c=0.443\pm0.002$ ($L=6$), 0.444 ± 0.0020 ($L=8$), 0.446 ± 0.001 ($L=10$), 0.047 ± 0.001 ($L=12$), and 0.4470 ± 0.0005 ($L=16$), where the errors have been estimated from the dispersion of T_c obtained by independent runs.

In order to make a finite-size scaling analysis, we plot $\ln(C_{\max})$ versus $\ln(L)$ in Fig. 2. The slope of the straight line drawn by least-squares fit for $L \geq 8$ is 2.84, which is not far from 3. Therefore, within statistical errors, our results indicate that the transition is of first order.

We have estimated the latent heat per spin ΔU by plotting the jumps of U at the transition as a function of L for $L=10$, 12, 16, and 20 (the last size has been used for this purpose): they are 0.055, 0.06, 0.071, and 0.074 for these sizes, respectively. A straightforward extrapolation to $L=\infty$ yields $\Delta U \simeq 0.08$. Note that when we fitted our data with Eq. (23) of Ref. 15, where L^d is replaced by $4L^d$, the resulting ΔU is about 0.042, which is smaller than the extrapolated value. The discrepancy between the two methods in estimating ΔU may be due to two factors: the uncertainty in reading the jumps of U which, strictly speaking, are continuous, and the approximation used to derive Eq. (23) of Ref. 15.

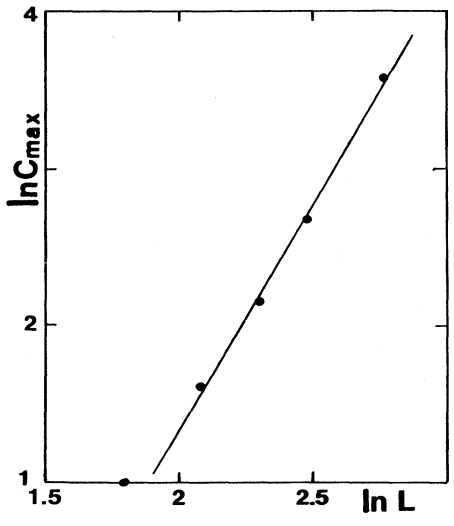

FIG. 2. Log-log plot of specific heat maximum $C_{\max}$ vs L. The slope of the solid straight line, obtained from a least-squares fit for $L > 8$, is 2.84.

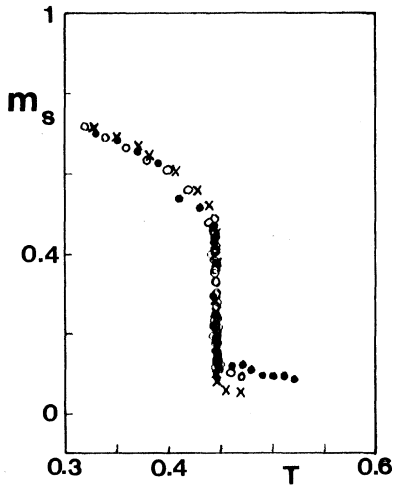

FIG. 3. Sublattice magnetization m_s vs T for $L=10$, 12, and 16 (filled circles, open circles, and crosses, respectively). m_s was averaged over the four sublattices as $m_s=[\sum_i <|m_i|>]/4$, where m_i is the sublattice magnetization per spin for the sublattice i ($i=1-4$) and $\langle \cdots \rangle$ denotes a thermal average.

Another interesting question is the nature of the ordering below T_c. Is it the collinear type I as predicted by Henley?[12] To answer this question, we have examined the associated sublattice magnetizations, and the results for $L=10$, 12, and 16 are displayed in Fig. 3. Indeed, the spin ordering realized below T_c is found to be that of collinear type I. We note that if a random initial spin configuration is used, the same type I ordering is observed just below T_c, but the stacking faults set in at lower temperatures ($T<0.4$), at least with our present simulation time, causing smaller sublattice magnetizations as compared to the case of ordered initial condition.

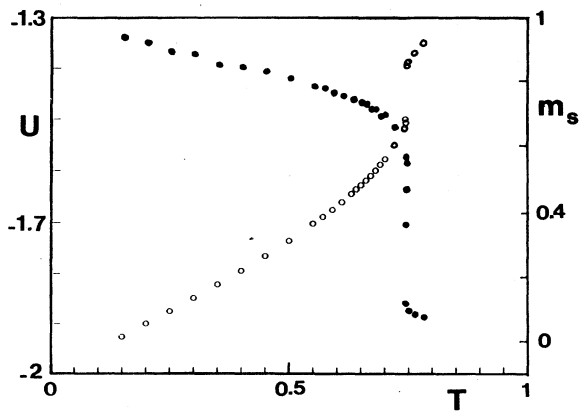

FIG. 4. XY case: internal energy per spin U (open circles, left-hand side) and sublattice magnetization m_s (solid circles, right-hand side) vs temperature T for $L=12$.

On the other hand, the use of random initial spin configurations does not change the behavior of sublattice magnetizations near the transition point ($T > 0.40$). The stacking faults are probably caused by the infinite ground-state degeneracy resulting from the frustration, as discussed earlier.

The first-order character of the transition found in this work, while in agreement with theoretical predictions,[4,5,12] is in contradiction with early MC results by Fernandez *et al.*[11] Their conclusion of a continuous transition may be due to rather small lattice sizes (up to $L = 8$), large ΔT, and rather short runs, which altogether prevented a detailed data analysis.

In the experimental connection, the collinear spin configuration of type I observed in our simulations suggests that the NN Heisenberg model may be appropriate for UO_2 among several materials mentioned above. Thus, experimental observation[1] of a first-order transition in UO_2 is consistent with our result.

As a concluding remark, let us touch upon the corresponding XY (plane rotator) model. We have also made a Monte Carlo simulation for the nearest-neighbor XY antiferromagnet on a fcc lattice. The collinear type I ordering is observed at low temperatures, accompanied with an "almost discontinuous" gap in the internal energy as in the case of Heisenberg model (see Fig. 4). Although we did not attempt a systematic finite-size scaling for the XY case, it appears to be clear that the transition is of first order due to the strong similarities between the Heisenberg and XY cases found in the Monte Carlo data as well as the ordered-state degeneracy discussed above.

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