Monte Carlo studies of an fcc Ising antiferromagnet with nearest- and next-nearest-neighbor interactions

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We have investigated, via Monte Carlo computations, the phase diagram of an ordering binary alloy—equivalent to an Ising spin system—on an fcc lattice with nearest- and next-nearest-neighbor pair interactions $H = J \sum_{nn} \sigma_i \sigma_j - \alpha J \sum_{nnn} \sigma_i \sigma_j$, $\sigma_i = \pm 1$, $J > 0$. Our studies indicate that this system undergoes a first-order transition; i.e., there is a discontinuity in the energy and order parameters as a function of temperature, for values $-1 \lesssim \alpha \lesssim 0.25$. For larger values of $|\alpha|$ the transition appears to be continuous, without any metastable states. Our results are in good agreement with Kikucki's cluster variation method at the two values of α at which it has been applied, namely, 0 and -0.25. For $\alpha \lesssim -0.5$ renormalization-group arguments strongly indicate that the transition is first order. If this is so, then our results indicate that the discontinuities for $\alpha < -1$ must be very small. The nature of the ground states changes at $\alpha = 0$ and -0.5. At these values of α the ground states are infinitely degenerate. The structure of the low-temperature phases, at all values of α, is discussed.

I. INTRODUCTION

We describe here the results of Monte Carlo studies of the phase diagram of an ordering binary alloy on a rigid fcc lattice, i.e., coherent phase diagram, with nearest- and next-nearest-neighbor pair interactions. As is well known there is a complete isomorphism between configuration of an AB alloy and that of an Ising spin system—achieved simply be identifying "spin up" ("spin down") at site i, $\sigma_i = 1(-1)$, with an $A(B)$ atom at that site. The Hamiltonian for the system may be conveniently written in spin language as

$$H = J \sum_{nn} \sigma_i \sigma_j - \alpha J \sum_{nnn} \sigma_i \sigma_j - h \sum \sigma_i \ , \qquad (1.1)$$

where the sums will always be understood to go over the $N = 4L^3$ sites of an fcc lattice with periodic boundary condition. The spin-spin interaction between sites i and j in Eq. (1.1) is equal to $\frac{1}{4}(v_{ij}^{AA} + v_{ij}^{BB} - 2v_{ij}^{AB})$ where v_{ij}^{AA} is the interaction between atoms of type A on sites i and j, etc. The equivalent external magnetic field is given by $h = \frac{1}{2}(v_{ij}^{AA} - v_{ij}^{BB}) + \frac{1}{2}(\mu_A - \mu_B)$, where μ_A and μ_B are the chemical potentials of the A and B atoms. The term $h \sum \sigma_i$ in Eq. (1.1) is relevant only when the magnetization m [or fractional concentration of A atoms $c = \frac{1}{2}(m + 1)$] is not precisely fixed since otherwise it is independent of the configuration. In the present paper we shall consider only the case $h = 0$ corresponding to an average concentration of 0.5. Preliminary results for $\alpha > 0$ were reported earlier,[1] and results for $c \neq 0.5$ will be presented in a separate note.

The phase diagram of this system for $J > 0$, a model of an alloy which orders at low temperatures is a problem of considerable theoretical and practical interest. The latter arises from the fact that many important alloys with an fcc structure can be reasonably well described by the Hamiltonian (1.1).[2,3] At the same time, different approximation schemes give greatly varying results for this system. Thus, for only nn interaction, $\alpha = 0$, mean-field[4] predicts a second-order transition at $T = 4J/k$, the quasichemical approximation[5] predicts a first-order transition at $T \approx (0.36)(4J/k)$, while the pair approximation[6] gives no transition at all. These methods do not seem capable of dealing with the complexity of the frustrated structure of the phases here. As we shall see later, however, our computations indicate that Kikucki's cluster variation method[7] does offer a reliable approximation scheme for this system. Using tetrahedral basic clusters, it gives[2,3] for $\alpha = 0$ a first-order transition at $T \approx 0.4733(4J/k)$. Other methods which may in principle yield accurate information about the phase diagram are low-temperature series expansions and renormalization-group calculations. (High-temperature series do not seem to be useful for the study of the transition.)

A low-temperature expansion has been carried out, for $\alpha = 0$, to five terms by Betts and Elliott[8] extending the three-term series by Danielian.[9] The coeffi-

cients in the series (beginning with the third) depend on the ground state about which the expansion is made. Since there are, for $\alpha=0$, an infinite number of ground states, Betts and Elliot performed an average over all of them. As was shown by Slawny, however, the six most symmetric ground states dominate all others at $\alpha=0$ and only they should be used in the expansion.[10] The extension and analysis of such an expansion is therefore an interesting open problem.

Renormalization-group calculations have so far been limited to predictions about the order of transition.[11] This will be discussed in Sec. II, where the structure of the ground states and low-temperature phases, at $h=0$, are described. We also present there in summary form our proposed $T-\alpha$ phase diagram for $h=0$ (equal concentrations). The Monte Carlo computations forming the basis of our phase diagram are explained in Sec. III and details of the results are given in Sec. IV.

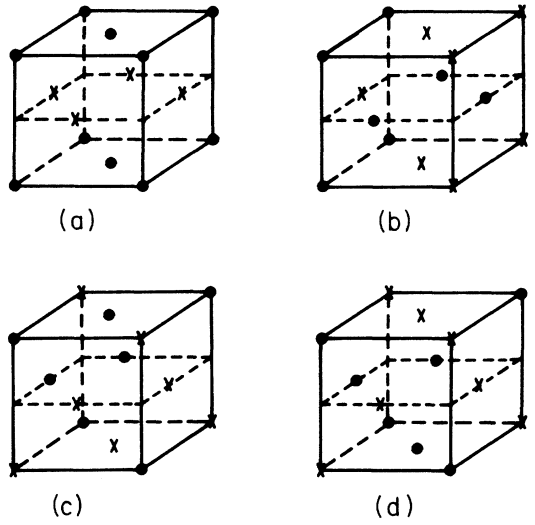

FIG. 1. One cube of the fcc lattice shown for the four possible structures (a) type-I antiferromagnetic (AB) structure, (b) type-III antiferromagnetic (A_2B_2) structure, (c) type-II antiferromagnetic, and (d) type-IIb antiferromagnetic structure.

II. GROUND STATES AND LOW TEMPERATURE PHASES FOR $h=0$

A. Ground states

The ground states of the Hamiltonian (1.1) on an fcc lattice with L cells on a side, L^3 cells, $N=4L^3$ sites, with periodic boundary condition were analyzed by Cahn and Allen.[12] The nature of the ground states changes at $\alpha=0$ and -0.5. Considering the fcc lattice as made up of four simple-cubic lattices, the ground state for $\alpha>0$ consist of three pairs of configurations in which two sublattices are occupied by A particles (spin pointing up) and the other two by B particles (spin pointing down) as in Fig. 1(a). The ground-state energy per site in units of J is $E_0=-2-3\alpha$. (The order parameter of this system is a three-component vector, $n=3$, with cubical symmetry; cf. Mukamel and Krinsky.[13])

For $-0.5<\alpha<0$ there are again six ground states (three pairs) consisting of A_2B_2-type configurations (type-III antiferromagnet) as shown in Fig. 1(b) with $E_0=-2-\alpha$. The order parameter now corresponds[13] to $n=6$.

For $\alpha<-0.5$ the ground state is either the CuPt structure (type-II antiferromagnet) or the "type-IIb" structure ($Fd3m$). The two arrangements of the unit cell which are not related by symmetry are shown in Figs. 1(c) and 1(d). In the structure of Fig. 1(c) each nearest-neighbor tetrahedron consists of an odd number of each of the two species (A or B) which gives rise to ferromagnetically ordered (111) planes. This is distinct from the arrangement in Fig. 1(d) where the tetrahedra have an even number of the species and no ordered (111) planes exist. In both structures each of the four sublattices is antiferromagnetically ordered. (With a two-component order parameter to each sublattice, we have $n=4$ for this system.) To get from one structure to the other it is necessary to flip the spins on one of the sublattices. The degeneracy of each of these sets of states is 8. The ground-state energy is $E_0=-3\alpha$. At $\alpha=0$ and -0.5 the degeneracy of the ground state goes to infinity[14] as b^L, $b>1$, when $L\to\infty$. The entropy per site therefore goes to zero as L^{-2} when $L\to\infty$.

B. Low-temperature phases

There exists a general theory due to Pirogov and Sinai,[15] extended by Holtzinski and Slawny[16] that describes the low-temperature phases of very general lattice systems, e.g., with many-body interactions as well as many-component alloys without any symmetry. The theory starts with the periodic ground states of the system and its main technical restriction at the present time is that these be finite. It therefore applies directly in our case only for $\alpha\neq 0$ or -0.5. It follows from this theory that for $\alpha>0$ and for $-0.5<\alpha<0$ there are six (three pairs) phases at sufficiently low temperatures. For $\alpha<-0.5$, where there are two types of ground states, Slawny has shown that the type-IIb structure dominates at low temperatures[17]; i.e., a macroscopic system will have eight (four pairs) equilibrium phases at low temperatures corresponding to Fig. 1(d). This dominance is however quite weak—it is only in sixth order that the

low-temperature expansions for the free energy based on the two types of ground states differ. The type-IIb structure gives lower values. Hence, we might expect to see both kinds of states in our computer simulations of small systems, but the type IIb will dominate as the size increases.

For $\alpha=0$ and -0.5 the theory does not apply directly. Arguments by Slawny,[10] however, strongly suggest that at $\alpha=0$ the six symmetric states given in Fig. 1(a) (the ground state for $\alpha>0$) dominate at low temperatures while for $\alpha=-0.5$ the states of Fig. 1(b) (ground states for $-0.5<\alpha<0$) dominate.

These considerations lead to the part of the phase diagram appropriate to very low temperatures given in Fig. 2. Note there the leftward curvature of the dashed lines starting at $\alpha=0$ and -0.5 in accordance with the suggested dominance at these values of α. This is unfortunately all that rigorous theory can say at the present time about the phase diagram except that at sufficiently high temperatures there is for all α a single disordered phase. There is nothing that can be said rigorously about the nature of the transition, whether second or first order, between the low-temperature ordered phases and the high-temperature disordered one. It is therefore necessary to analyze this question by approximate methods and by experiment. Computer simulations constitute one type of such experiment. They have the advantages of an exactly known Hamiltonian and the disadvantages of being limited to small systems and finite computations.

We have already mentioned the conflicting results of some approximation schemes and shall now describe briefly the results, as far as we understand them, of renormalization-group considerations. (We refer the reader to Refs. 11, 13, and 19–23 for details.) For $\alpha>0$, we have an $n=3$ order parameter with cubic symmetry and the Landau-symmetry criteria[18] permit either a first- or second-order transition. The renormalization group, to first order in ϵ, finds a stable fixed point for the appropriate Landau-Ginzburg Hamiltonian with an uncertain domain of attraction. Hence, it also permits the transition to be either first or second order. In particular it can switch from one kind to the other kind, as suggested by our phase diagram, Fig. 2. Such a changeover has also been found both from renormalization-group and Monte Carlo computations for a system like ours on a bcc lattice[19] and for systems with more complicated interactions[20] on a simple-cubic lattice. For $\alpha<-0.5$ the Ginzburg-Landau Hamiltonian for a type-II antiferromagnet corresponds to a system with an $n=4$ order component. Such an iteration shows no stable fixed point to second order in an ϵ expansion.[21] This result suggests strongly that the transition should be first order but that conclusion has been questioned on the basis of experiments on cerium monochalcogenides.[22] A possible explanation lies in the existence of a very weak first-order transition.[23] We were therefore particularly interested in investigating this question by Monte Carlo methods. As we shall see however, later, the computer experiments, while strongly supporting the existence of a first-order transition for $-1\lesssim\alpha\lesssim-0.5$, are ambiguous for $\alpha<-1$.

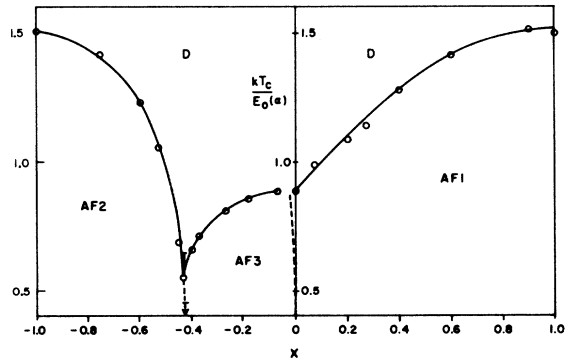

FIG. 2. The $T\alpha$ phase diagram for the fcc lattice, $x=3\alpha/(2+3|\alpha|)$. The circles are Monte Carlo data for $L=8$ except for the point at $x=,(\alpha=-2.0)$. Points at $x=\pm1$ are the series of expansion results (Ref. 27). The dashed lines are the proposed boundaries separating the different phases. Close to $\alpha=0$ and $\alpha=-0.5$, the points are triple points, exact location of which cannot be determined.

III. MONTE CARLO METHOD

The microscopic state x of the system of N sites can be specified by giving the value of $\sigma_i=\pm1$ at each site

$$x=(\sigma_1,\sigma_2,\ldots,\sigma_N)\ . \tag{3.1}$$

The variable x can take on 2^N values. We then consider an ensemble of these systems such that the probability that a system picked from the ensemble at random will be in the microscopic state x, is $P(x)$. When the system is in equilibrium at a temperature T then

$$\begin{aligned} &P_{\rm eq}(x)=Z^{-1}\exp[-\beta H(x)],\\ &Z=\sum_x\exp[-\beta H(x)]\ , \end{aligned} \tag{3.2}$$

where $\beta=(kT)^{-1}$, k is the Boltzmann constant, and $H(x)$ is the configurational part of the Hamiltonian given by Eq. (1.1). In our model we consider an fcc lattice with periodic boundary conditions. When the system is started at some initial time $t=0$, with some nonequilibrium probability distribution $P(x,0)$, $P(x,t)$ will evolve according to a master equation

$$\frac{\partial P(x,t)}{\partial t}=\sum_{x'}[K(x',x)P(x',t)-K(x,x')P(x,t)]\ . \tag{3.3}$$

In the usual Monte Carlo calculations, the transition probability $K(x,x')$ from a state x to x', is constructed so as to satisfy the detailed-balance condition

$$K(x',x)\exp[-\beta H(x')] = K(x,x')\exp[-\beta H(x)] \ , \tag{3.4}$$

which is sufficient but not necessary to ensure that the equilibrium distribution is a stationary solution of the master equation and is approached for long times, $t \to \infty$, from any initial state. Thus at long times, the Monte Carlo provides configurations drawn from the distribution $P_{eq}(x)$. Any required averages may be computed from these.

In our simulations at a fixed magnetic field $h = 0$ we used a transition probability corresponding to a "spin-flip" operation, i.e., Glauber dynamics. A site is picked at random and the energy change ΔE that would result from the flip is calculated. The spin is in fact reversed if $\exp(-\Delta E/kT)$ is greater than a random number ξ chosen uniformly between 0 and 1. The number of attempted "flips" per site gives a time scale for the calculation regarded as a stochastic simulation of Eq. (3.3).

In carrying out our computer simulation at a particular temperature the system was started either (a) from a random state, (b) from an ordered state, or (c) from an "equilibrium configuration" at a nearby temperature. Quantities such as the internal energy E and sublattice magnetization m were calculated directly from the averages of these quantities over many different configurations after an initial equilibration period. The fluctuations in the energy were also determined. This yields an independent but sometimes unreliable determination of the specific heat at temperature T

$$\frac{C(N,T)}{k} = \left(\frac{1}{k^2T^2}\right)(\langle H^2\rangle - \langle H\rangle^2), \quad \langle H\rangle = NE \ . \tag{3.5}$$

The results of these computations will be described in the Appendix.

The use of Monte Carlo techniques to study phase transitions, particularly continuous (second-order) ones, has been discussed by many authors.[24,25] We shall therefore limit our discussion here to first-order transitions. At a first-order transition the infinite system exhibits a discontinuity in the energy and/or in other order parameters. While there is no discontinuity in a finite system, a graph of energy versus temperature formed by connecting points obtained from Monte Carlo studies of small systems at different temperatures, may reveal the discontinuity present in the infinite systems. This becomes more difficult, however, as the infinite system discontinuity, ΔE, becomes smaller. For sufficiently small ΔE it may be entirely impossible. There are thus several sources of difficulty in deciding the nature of the transition on the basis of Monte Carlo results: (a) the intrinsic round-off of the average energy-versus-temperature curve $E(T,N)$ for fixed N, (b) the limited number of temperatures at which we carry out the calculation, and (c) the uncertainty in the values of $E(T,N)$ obtained from one (or a few) computer runs at a given T—this is particularly troublesome in the vicinity of the transition temperature. There are different but comparable problems in a real experiment when ΔE is small.

To understand the nature of these problems we note for T in the vicinity of $\hat{T}$, the first-order transition temperature in the infinite system, the probability distribution for the energy H will have two peaks, centered roughly about the average energy in each of the two coexisting phases. In the limit $N \to \infty$ one of the peaks dominates for $T \neq \hat{T}$ with ratio of areas under each peak behaving asymptotically as $\exp[\phi(T)N]$, $\phi(\hat{T}) = 0$. In a Monte Carlo simulation the number of attempts per site required to sample both peaks adequately will grow with N going to infinity as $N \to \infty$. It is thus only by taking a very long sample that one would get very accurate values of $E(T,N)$. Even more important, extremely long runs are necessary to obtain the specific heat $C(T,N)$ from computations of $\langle(\Delta H)^2\rangle$. The divergent contributions to $C(T,N)$ in Eq. (3.5) at $T = \hat{T}$ come from jumps between the peaks and not from fluctuations inside the peaks. These are clearly distinguished, however, only when N is large in which case we cannot wait long enough to see the jumps between the peaks (cf. Appendix).

The existence of these two peaks manifests itself in practice by giving rise, for the reasons described above, to metastable states when our system is heated or cooled across $\hat{T}$. The length of time spent in these metastable states should increase with the size of the system and decrease, for given N, as $-\Delta E/kT$ decreases. The existence or absence of metastable states, becoming more pronounced as N is increased, is then a guide in determining the order of the transition. The expected relaxations of the energy as it is cooled across the transition temperature is shown in Figs. 3(a) and 3(b) for a first-order and a second-order transition: At a second-order transition there is a monotone relaxation of energy while at a first-order transition long-lived metastable states are evident.

Most of our simulations were carried out on an fcc lattice with $L = 8$ ($N = 2048$sites). Some computations were, however, also carried out for $L = 5$, 10, and 14 size lattices. All results unless otherwise indicated will be understood to refer to the $L = 8$ lattice. The energy and sublattice magnetization were averaged over time intervals between 500 and 2500 attempted interchanges per site. For a given N we denote by $T_c(N)$ the temperature at which a smoothed curve of $E(T,N)$ vs T appears to have its maximum slope.

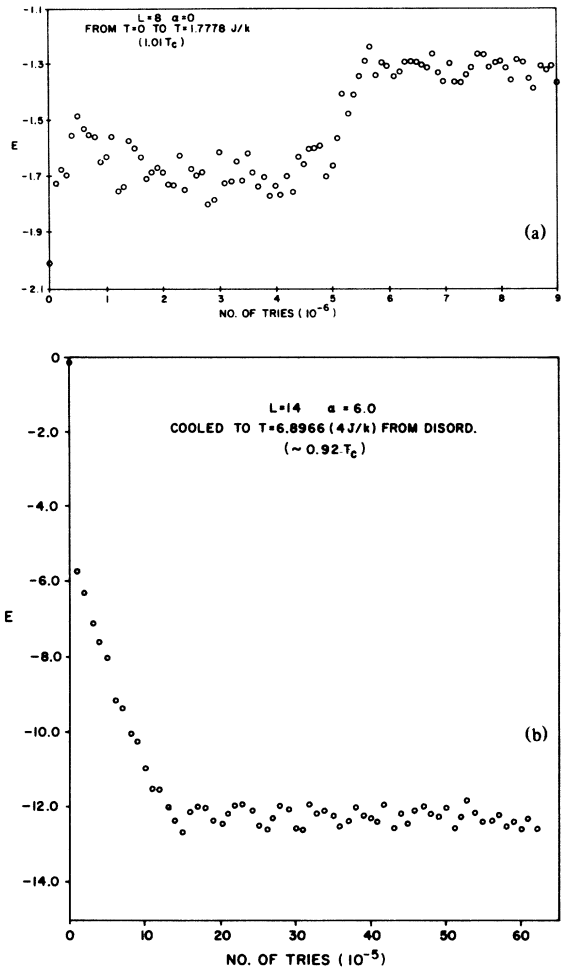

FIG. 3. (a) Time variation of energy per site for $\alpha=0$ as it is heated up from $T=0$ [perfectly ordered (AB)] configuration to $T=0.4445(4J/k)$ [above $T_c(N)$] for the $L=8$ lattice. Existence of the metastable states shows a first-order transition. (b) Time variation of energy per site for $\alpha=6.0$ as it is cooled across T_c from a disordered state to $T=6.8966(4J/K)$ for $L=14$. There does not exist any metastable states indicating a second-order transition.

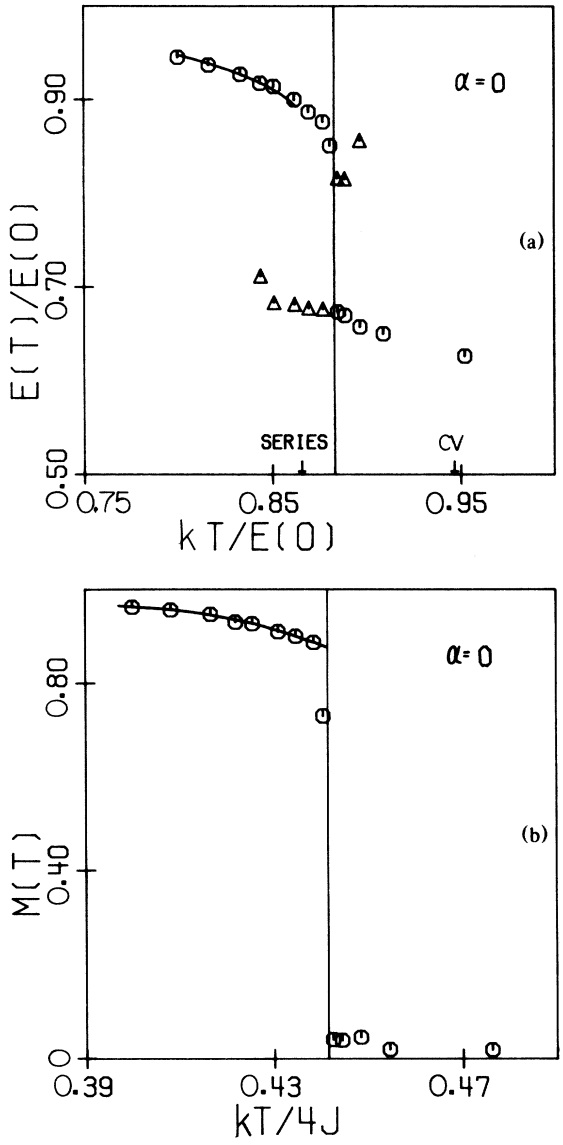

FIG. 4. (a) The energy $E(T)/E(0)$ for $\alpha=0$ is shown by circles. The triangles are the metastable points obtained by heating and cooling across T_c shown by the vertical line. The solid curve is given by the (2,2) Padé approximate (Ref. 8). The transition temperatures given by both series-expansion and cluster-variation methods are shown. (b) Sublattice magnetization vs $kT/4J$ for $\alpha=0$. The solid line gives the position of the transition temperature.

IV. RESULTS

a. $\alpha=0$. Figure 4(a) shows the plot of energy $E(T)/E(0)$, for $\alpha=0$, together with the metastable points. The existence of such metastable points can be seen from a typical plot of energy with time in Fig. 4(a) when the system was heated from a perfectly ordered state to a temperature $T=0.4445(4J/k)$. These observations suggest the existence of a first-order transition at temperature $T_c\cong 0.4415(4J/k)$ as compared to $T_c=0.4733(4J/k)$ predicted by the cluster variation (CV) method.[2,3] The change in entropy $\Delta S=\Delta E/kT_c$ at the observed transition is ~ 0.20. This is in good agreement with $\Delta S\sim 0.25$ obtained by the CV method.

The large degeneracy of the ground state[9] (of order 2^L) posed a problem in getting stable values of the sublattice magnetization in a reasonable time. To obtain such values the simulation had to be started from an ordered state. A plot of sublattice magneti-

zation versus T is given in Fig. 4(b).

b. $\alpha > 0$. Simulations at $\alpha = 0.05$ and 0.167 showed behavior similar to that observed at $\alpha = 0$. The discontinuities ΔE and m^* at the transition temperature $T_c(\alpha)$ kept on decreasing, however, and for $\alpha = 0.25$, 0.445, 1.0, and 6.0 were not observable at all. Figure 5 shows plots of the average energy $E(T, \alpha)$ divided by $E(0, \alpha)$ vs $kT/E(0, \alpha)$, where $E(0, \alpha) = -2 - 3\alpha$ is the ground-state energy. Plots of average sublattice magnetization versus $T/T_c(\alpha)$ are shown in Fig. 6.

An inspection of Figs. 5 and 6 indicates that at small values of α there is a discontinuity in the energy and sublattice magnetization which is absent for large values of α. This suggests a changeover from a first- to a second-order transition at about $\alpha = 0.25$ with the likelihood of a tricritical point. Such an interpretation is further strengthened by the absence of metastable states for larger values of α. The time variation of the energy at $\alpha = 6.0$ for a lattice with $L = 14$, when the system was cooled from a random state to $T = 6.8966(4J/k)$ is shown in Fig. 3(b). There does not appear to be any metastable state and

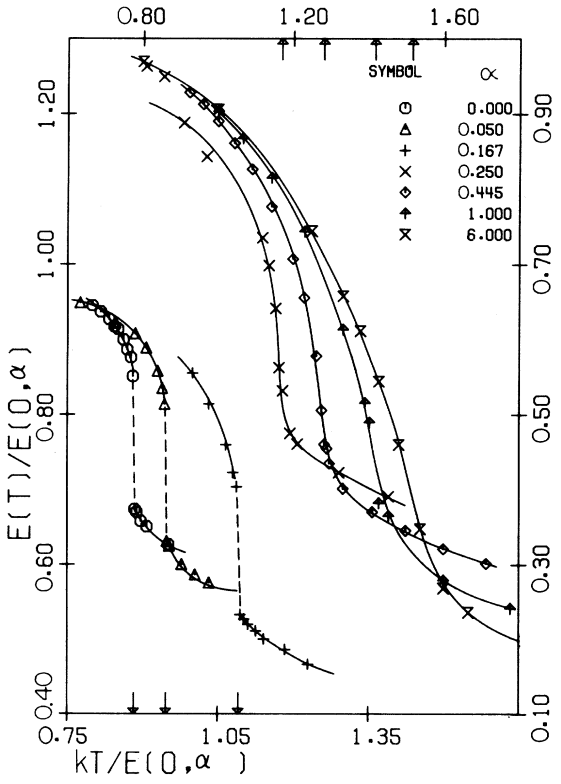

FIG. 5. $E(T, \alpha)/E(0, \alpha)$ vs $kT/E(0, \alpha)$ for positive values of α are shown. The left bottom scales are for the three smaller values of α where the discontinuities are indicated. The top and right scales are for $\alpha \geq 0.25$ where the transition is continuous. The transition points are indicated by arrows.

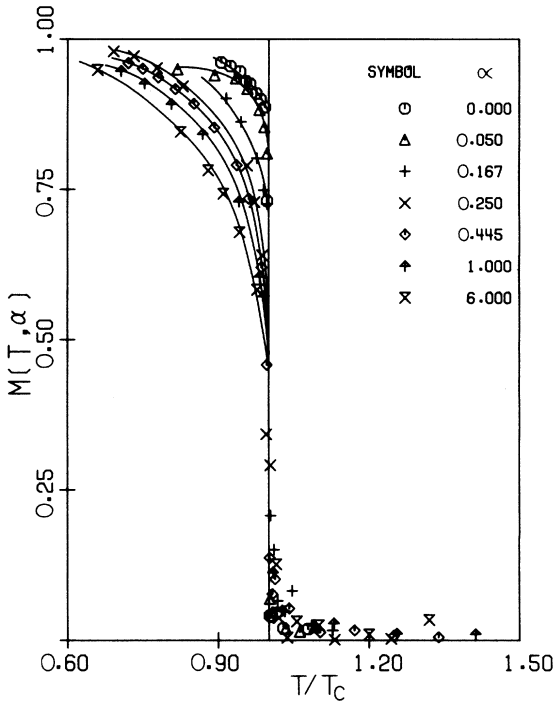

FIG. 6. Plots of the sublattice magnetization vs T/T_c are all α.

so we judge the transition at $\alpha = 6$ to be a continuous one.

We also carried out a finite-size scaling analysis at $\alpha = 6.0$ where we might expect to be outside the tricritical region. Using lattices of size $L = 5$, 8, and 10 and the scaling relation[26]

$$T_c(N) = T_c(\infty)(1 - a/L^{\lambda}); \quad \lambda = 1/\nu = 1/0.64 \qquad (4.1)$$

we estimate $kT_c(\infty)/4J = 7.8$ and the exponent β for the sublattice magnetization to be in the range 0.21–0.36. The specific-heat behavior can be represented by the relation

$$C(T,N)/Nk = A[1 - T/T_c(\infty)]^{-\alpha} + b \qquad (4.2)$$

for $T < T_c(N)$ with $\alpha = \frac{1}{8}$, $A \simeq 1.4$, and $b = -2.0$. This is consistent with the behavior at a second-order transition (Ising or Heisenberg) but is certainly no proof.

In Fig. 7 we plot our values of the transition temperatures $kT_c(\alpha)/E(0, \alpha)$ versus a variable $x = 3\alpha/(2 + 3|\alpha|)$ the fraction of the ground-state energy due to next-nearest-neighbor interactions to obtain a phase diagram of our system in the Tx plane for $\alpha > 0$. For $x = 1$ ($\alpha = \infty$), the system splits up into four independent sublattices with nn interactions. Extrapolation of our plot to $x = 1$ gives a value of the transition temperature within a few percent of

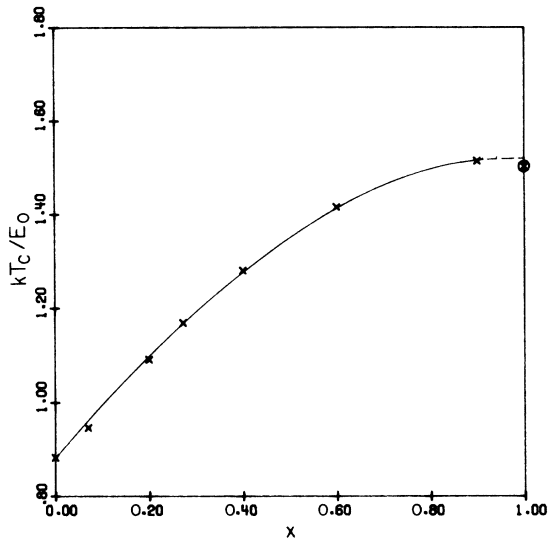

FIG. 7. Plot of $kT_c(\alpha)/E_0(\alpha)$ vs $x=3\alpha/(2+3\alpha)$ for $\alpha>0$. The point at $x=1$ is the series-expansion values.

that computed from an extended series expansion.[27] It is generally accepted that the transition for this system at $x=1$ is of second order.

c. $-0.5<\alpha<0$. Computations for $-0.5<\alpha<0$ where the ground state is A_2B_2 or type-III antiferromagnet indicate a first-order transition with strong metastable states and pronounced discontinuity in energy. The sublattice magnetization is no longer a suitable order parameter here as it equals zero even in the ordered state. Calculations at $\alpha=-0.05$, -0.15, -0.25, -0.40, and -0.45 show that the transition temperature gradually decreases as one goes from $\alpha=0$ to $-\frac{1}{2}$, while the entropy discontinuity $\Delta S=\Delta E/kT_c$ increases, see Figs. 8(a), 8(b), and Table I.

Comparison with CV results of de Fontaine and Sanchez[28] at $\alpha=-0.25$ give excellent agreement for the transition temperature and the energy change. Figure 9 shows the plot of energy versus kT/J as obtained from the CV[29] and Monte Carlo methods. Renormalization-group analysis for the $n=6$ component system indicates the existence of a stable fixed point which is however unreachable for a pure type-III structure; hence the transition should be first order.[30]

d. $\alpha=-0.5$. As α changes from $\alpha=0$ to -0.45 the entropy discontinuity $\Delta E/kT_c$ appears to go towards a limiting value close to $\sim\frac{1}{2}\ln 2(\simeq 0.36)$ (see Table I). This would suggest that the system at $\alpha=-\frac{1}{2}$ is so highly degenerate as to have a finite entropy at $T=0$. It can be shown,[14] however, that this is not the case. The ground state at $\alpha=-\frac{1}{2}$ has zero entropy per site (in the infinite system) and the thermodynamic entropy must go to zero as $T\to 0$.

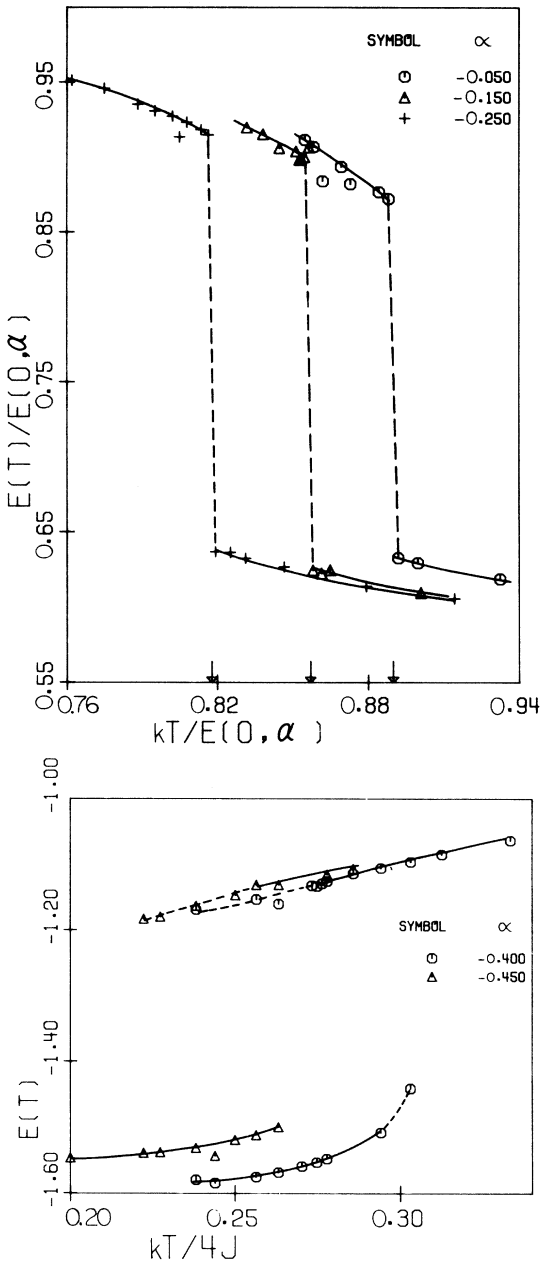

FIG. 8. (a) Plots of energy per site vs $kT/4J$ for $\alpha=-0.05$, -0.15, and -0.25 where the transitions are first order. The discontinuities are shown by dotted lines and the position of the transition shown by arrows. (b) Energy per site vs $kT/4J$ for $\alpha=-0.4$ and -0.45 with the respective metastable states.

It required very extensive computations to see a transition to the ordered structure which was a majority A_2B_2 (type III) plus some type II. Since the transition takes place at very low temperature, the metastable states are very long-lived, and we could not get

TABLE I. Monte Carlo estimates of the transition temperatures and energy discontinuities for different values of α. All data for $L=8$ lattice unless mentioned otherwise.

α	$kT_c/4J$	$kT_c/\lvert E_0\rvert$	$\Delta E_t/kT_c$
6.0	7.5758 + 0.1420	1.5151 ± 0.0284	0
1.0	1.7699 ± 0.0150	1.4159 ± 0.0120	0
0.445	1.0672 ± 0.0029	1.2806 ± 0.0035	0
0.25	0.8036 ± 0.0032	1.1406 ± 0.0047	0
0.167	0.6821 ± 0.0023	1.0913 ± 0.0037	0.09
0.05	0.5089 ± 0.0013	0.9468 ± 0.0024	0.19
0.0	0.4415 ± 0.001	0.8830 ± 0.002	0.20
0.0[a]	0.4733	0.9466	0.26
−0.05	0.4338 ± 0.0019	0.8898 ± 0.0039	0.27
−0.15	0.3964 ± 0.0025	0.8571 ± 0.0035	0.33
−0.25	0.3578 ± 0.0008	0.8178 ± 0.0018	0.34
−0.25[b]	0.3750	0.8571	0.29
−0.40	0.2857 ± 0.01	0.7163 ± 0.025	~0.34
−0.45	0.2560 ± 0.0125	0.6606 ± 0.0323	~0.36
−0.50	0.2084 ± 0.04	0.55 ± 0.107	~0.33
−0.55	0.2857 ± 0.021	0.6926 ± 0.051	~0.39
−0.75	0.5952 ± 0.008	1.0582 ± 0.0142	...
−1.0	0.9259 ± 0.0128	1.2345 ± 0.017	...
−2.0[c]	2.1277 ± 0.0453	1.4185 ± 0.0302	0
± ∞[d]		1.5036	

[a]References 2 and 3.
[b]References 28 and 29.
[c]Data for $L=14$ lattice.
[d]Reference 27.

a very accurate estimate of T_c. Our best guess is $T_c=(0.21\pm0.04)(4J/k)$. A plot of energy versus temperature is given in Fig. 10.

e. $\alpha<-0.5$. In this region there are two types of ground states, type-II and type-IIb antiferromagnets with the latter structure expected to dominate weakly at low temperatures. We carried out simulations at $\alpha=-0.55$, -0.75, -1, and -2. The ordered states we obtained by cooling from above T_c were about evenly divided between the two structures.

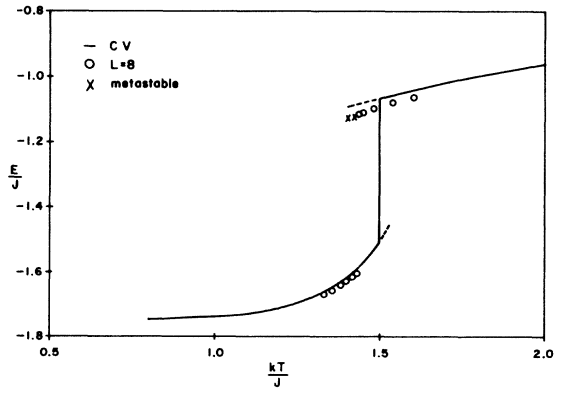

FIG. 9. Comparison plot of energy/site vs kT/J at $\alpha=-0.25$ by Monte Carlo date (circles) and cluster-variation data (solid line) (Ref. 29). The dashed lines indicate the metastable regions.

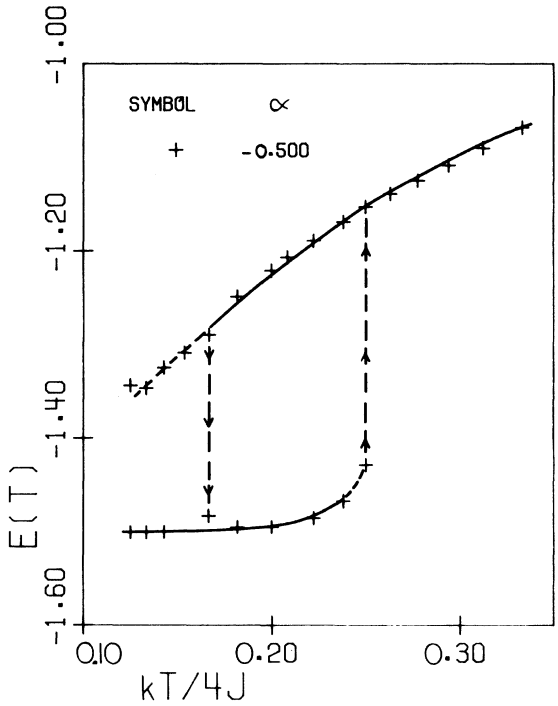

FIG. 10. Energy per site vs $kT/4J$ for $\alpha=-0.5$. Cooling from high temperature; only at one point did the system order after long computation (shown by dashed arrow); also the point where it went to disorder by heating from ordered state is shown by similar dashed lines. The transition temperature lies between the two dashed vertical lines.

As mentioned in Sec. II renormalization-group analysis predicts that for all α's less than -0.5 the transition should be first order[23,31] and only in the limit $\alpha \to -\infty$ should it go over to second order: Experiments[22] on cerium monochalcogenides, which are type-II antiferromagnets, on the other hand, show a continuous transition.

Our simulations give a strong first-order transition at $\alpha = -0.55$. As α becomes more negative, however, the transition temperature increases, and the discontinuity in the energy becomes less and less pronounced (see Figs. 11 and 12). Using an $L = 8$ lattice we did not observe any metastable states at $\alpha \leq -0.75$.

We then carried out computations at $\alpha = -0.75$, -1.0, and -2.0 on larger lattices $L = 10$ ($N = 4000$) and $L = 14$ ($N = 10\,976$). While the energy versus temperature curve does not change much with size both larger lattices exhibit metastable states for $\alpha = -0.75$ (Fig. 12) and $\alpha = -1.0$ (Fig. 13) indicating a first-order transition. For $\alpha = -2.0$, however, even the $L = 14$ system did not exhibit any metastable states (Fig. 14). While this suggests a changeover from a first- to a second-order transition, one must be cautious. The smooth appearance of the E vs T curve and the absence of metastable states may be due to the decrease in $\Delta E/kT_c$ which permits the system to go back and forth between the two phases across T_c when the system is small. It is impossible to decide unambiguously on the basis of computer calculations whether for $\alpha < -1$ the transition actually goes over from a first- to a second-order one. Such an ambiguity might also exist for $\alpha > 0$ where we had reported such a changeover. Here too we could not detect any metastable states at $\alpha = 6.0$ even for the $L = 14$ lattice [Fig. 3(b)]. The question therefore remains open.

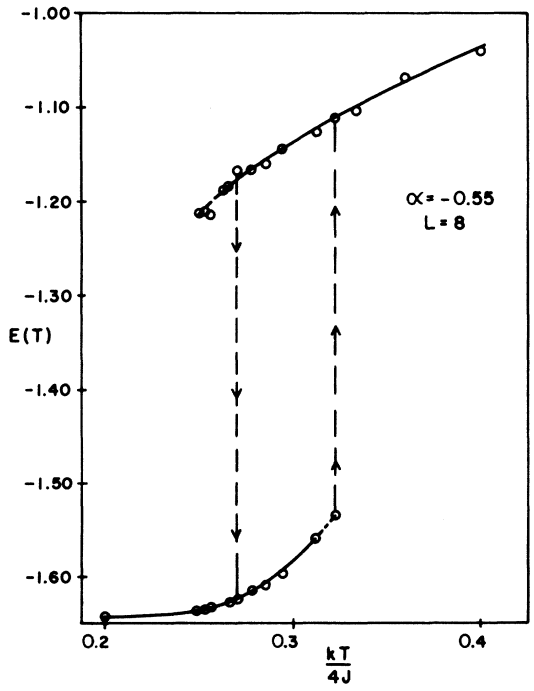

FIG. 11. Plot of energy per site vs $kT/4J$ for $\alpha = -0.55$. The metastable regions are shown.

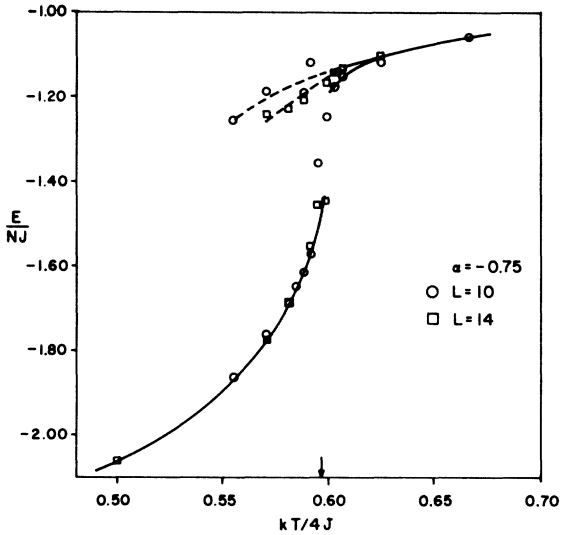

FIG. 12. Energy per site vs $kT/4J$ for $\alpha = -0.75$ for $L = 10$ and 14 lattices.

It is not clear at this time whether the existence of the two types of ground states, with the dominance of type IIb can affect the nature of the transition. Renormalization-group analysis does not distinguish between these two structures.

We should note here that as α decreases below -0.5 the slope of E vs T sharply rises as $T \to T_c$ from below. It is conceivable that, while the transition remains first order for $-\infty < \alpha < -0.5$, the specific heat (and staggered susceptibility) might diverge for sufficiently negative α as $T \to T_c$.

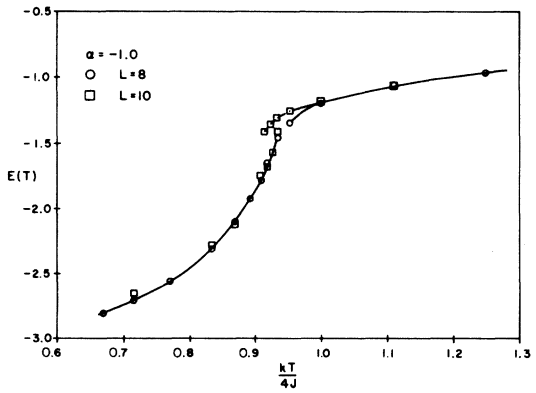

FIG. 13. Energy per site vs $kT/4J$ for $\alpha = -1.0$ for $L = 8$ and 10 lattices where the existence of metastable states could still be detected for the latter although the energy discontinuity is not very much evident.

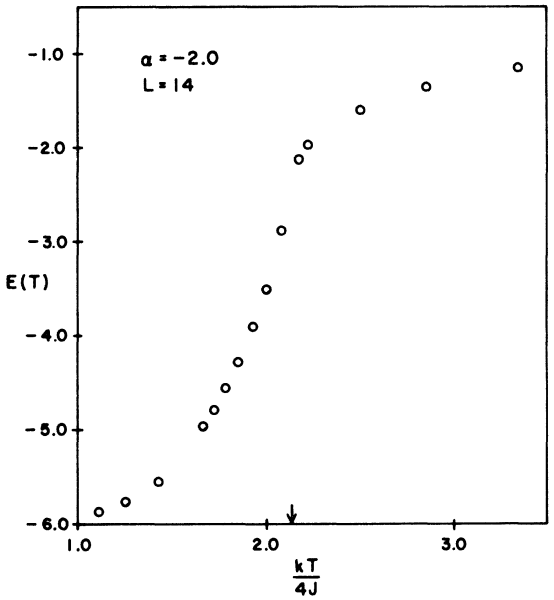

FIG. 14. Energy per site vs $kT/4J$ for $\alpha = -2.0$ for $L = 14$ lattice. The continuous variation of energy and nonexistence of metastable states strongly suggests a second-order transition.

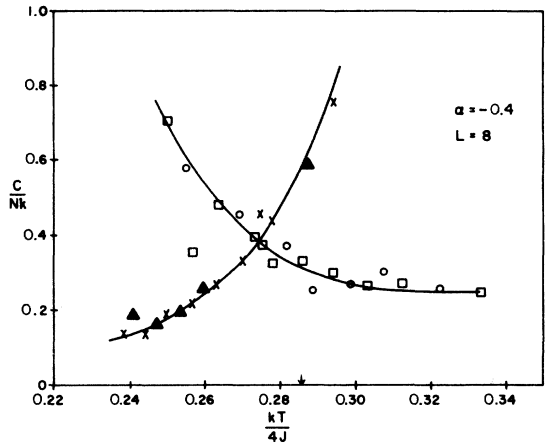

FIG. 16. Plot of specific heat with temperature for $L = 8$ at $\alpha = -0.4$ where the transition is strong first order. The squares and crosses are points obtained from energy fluctuations as one comes from high and low temperatures, respectively. The circles and the triangles are the corresponding points from energy differentiation.

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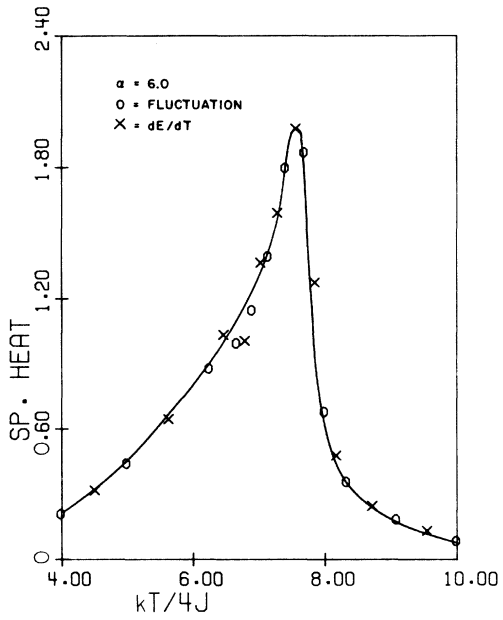

FIG. 15. Specific heat C/k vs $kT/4J$ at $\alpha = 6.0$ where the transition is continuous for $L = 8$ lattice as obtained from fluctuations in energy (circles) and from differentiation of energy (crosses).

APPENDIX

As mentioned in the text, we monitored the variations in the energy $H(t)$ as a function of time t following thermalization. The fluctuations of H computed over a period of time τ then approach as $\tau \to \infty$ the specific heat C given by Eq. (3.5). The C computed this way should of course coincide with that obtained from differentiation of the energy $E(T)$. We found, that for the values of τ we used, there was indeed good agreement between C computed from the fluctuations and the numerical differentiation of the E vs T curve. The agreement continued to be reasonable even in the vicinity of a second-order transition (Fig. 15). In the neighborhood of a first-order transition, however, we expect for the reasons discussed in the text, that the energy over the period τ will be limited to the range of one of the peaks; i.e., it would not jump from one peak to another. Hence C given by Eq. (3.2) should be close to the specific heat in each of the phases missing the jump in energy corresponding to a delta function singularity in the specific heat due to the transition (cf. Fig. 16). In that figure the specific heat is continued into the metastable region and appears to approach an infinite value at some spinodal line.

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