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Numerical study of the spin-glass transition in a dilute Ising model on a triangular lattice

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We study an Ising model with nearest-neighbor antiferromagnetic interactions. It is placed on a triangular lattice, where each site is occupied by a spin with x probability. There is no applied magnetic field. Randomness and frustration, two essential ingredients of spin-glasses, are present in this model. We study its critical properties here. The entropy is obtained by a transfer-matrix calculation as a function of x at low temperature ($T=0.3J/k_B$) for systems on a lattice of 10×20 sites. A fairly shallow minimum appears near $x\simeq0.9$, which suggests that this case is the one most likely to show a transition into an ordered state at low temperature. We study the cases $x=1$, 0.9, and 0.74, which is about half-way to the critical percolation. We simulate systems on lattices of 50×50 sites and 30×30 sites by the Monte Carlo method. The specific heat has a broad maximum at $T\simeq0.9$ for $x=0.74$ and 0.9. χ_{SG}, defined by $\chi_{SG}=N^{-1}\sum_{i,j}\langle\sigma_i\sigma_j\rangle^2$, and the relaxation time (τ) are obtained for $T\geq0.6J/k_B$. Both quantities, τ and χ_{SG}, turn out to be proportional to $\exp[A/(T-T_0)^c]$ and $0\leq T_0\lesssim0.4$; a fit with $T_0=0$ yields $c\approx1$ for $x=1$ but $c\approx2$ for $x=0.74$ and 0.9.

I. INTRODUCTION

Quenched randomness is an essential feature of spin-glasses.[1] It occurs in metallic spin-glasses,[2] such as CuMn or AuFe, where the magnetic atoms enter substitutionally at *random* into the nonmagnetic metal and occupy fixed lattice sites after the system is quenched. It also occurs in insulators or semiconductors, such as[3] $Cd_{1-x}Mn_xS$ (Zn and Hg instead of Cd may be used, and any element in the Periodic Table column below S may also be used instead of S). *Frustration* (the impossibility to minimize simultaneously all pairwise spin interactions in the system) is believed to be the other essential ingredient of spin-glasses.[4] It occurs in metallic spin-glasses because the *random* location of the local magnetic moments (spins) and the oscillatory in sign nature of the long-ranged Ruderman-Kittel-Kasuya-Yosida spin-spin interaction produce a random mesh of ferromagnetic and antiferromagnetic interactions which cannot be minimized simultaneously. Frustration occurs in the case of $Cd_{1-x}Mn_xS$ because (a) of the geometrical structure of the fcc or hcp lattice where the Mn atoms enter substituting Cd atoms, and (b) the magnetic moments carried by the Mn atoms interact *anti*ferromagnetically and only (approximately) if they are nearest neighbors.[5] Since two nearest-neighbor atoms may themselves be nearest neighbors in these lattices, it follows that not all interaction energies can be minimized simultaneously, and it is said that the lattice itself is frustrated. The triangular lattice is similarly frustrated.

A cusp in the magnetic susceptibility as a function of temperature at $T=T_0$ has been reported[3] for $Cd_{1-x}Mn_xTe$ and for $Cd_{1-x}Mn_xSe$, but extensive measurements to establish that these results are not time dependent [by, for instance, studying $T_0(\omega)$ as a function of the frequency (ω) of the applied magnetic field] have not been performed yet.

Whether randomness and frustration are sufficient to produce a true transition to the spin-glass state or whether some additional ingredient, such as some local anisotropy is necessary, is not yet established. The study of models with some or all of these ingredients is, therefore, of interest to establish a theory of spin-glasses.

Previous Monte Carlo (MC) simulations of dilute Ising models on fcc lattices, as well as on triangular lattices,[6] have shown that there is considerable

slowing down at low temperature T, which is typical of spin-glasses, but it did not indicate whether it occurs gradually as T decreases or whether there is a critical point.[7] An analytic solution exists only for $x=1$, and it shows that there is no phase transition in that case.[8]

The purpose of this paper is to determine, by means of a Monte Carlo simulation, whether a critical point exists separating the paramagnetic and spin-glass phases of a dilute antiferromagnetic Ising model on a two-dimensional triangular lattice, or whether the system freezes gradually as the temperature decreases. We study systems with the following values of the fraction x of sites occupied: $x=0.74$, 0.9, and 1, but concentrate on the case of $x=0.9$ for reasons that follow. We are interested in studying the system most likely to exhibit a sharp transition, which we expect to be the one with the lowest entropy S per spin in the $T\rightarrow 0$ limit. By means of a transfer-matrix computation,[9] we first determine S as a function of x for a system of 10×20 sites at $T=0.3J/k_B$. The results, exhibited in Fig. 1(a), show that the minimum value of S occurs for $x\simeq 0.9$. Figure 1(b) shows S vs T for $x=0.74$, 0.9, and 1. The transfer-matrix method is used, instead of an MC simulation, because long relaxation times at low temperatures (see Sec. II) turn

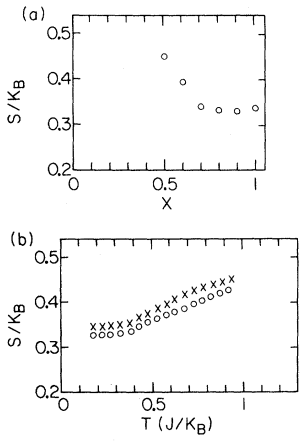

FIG. 1. (a) Entropy per spin as a function of x is shown for $T=0.3J/k_B$ for systems on 10×20 sites. It is obtained by the transfer-matrix method. The value for $x=1$ agrees with the exact result of Wannier. (b) The entropy (in units of k_B) is shown as a function of T (in units of k_B) for systems of atomic concentrations $x=0.74$, 0.9, and 1, all on 10×20 sites. The transfer-matrix method was used to obtain these values. o is for $x=1$ and for $x=0.9$, and $\times$ is for $x=0.74$.

MC simulations impractical. Furthermore, MC simulations do not yield S directly; the specific heat C must first be obtained as a function of T, and only then can S be obtained by means of

$$S=k_B\ln 2-\int_T^\infty (C/T')dT' ,$$

where k_B is Boltzmann's constant.

If there is a critical point at some temperature T_0 separating the paramagnetic and spin-glass phases, the relaxation time τ ought to diverge as $T\rightarrow T_0$ from above, as Kirkpatrick[10] has stressed. To calculate τ we first obtain

$$q(t)=N^1\sum\langle\sigma_i(0)\sigma_i(t)\rangle , \tag{1.1}$$

where N is the number of spins in the system, $\sigma_i(t)$ is the ith spin at time t, and $\langle\ \rangle$ denote time averages, i.e.,

$$\langle\sigma_i(0)\sigma_i(t)\rangle=\frac{1}{m}\sum_{n=1}^{m}\sigma_i(t_n)\sigma_i(t_n+t) . \tag{1.2}$$

Incidently, if the system is mixing[11]—which spin-glasses may not be—then the time average defined in Eq. (1.2) becomes equal to an ensemble average in the $m\rightarrow\infty$ limit. We let

$$\tau=\int_0^\infty q(t)dt , \tag{1.3}$$

since $q(0)=1$.

It is appropriate at this point to remark on the relevance of the dependence of $q(t)$, which we obtain by the Monte Carlo method, to its *experimental* behavior. $q(t)$ evolves experimentally, due to spin-phonon and dipolar interactions. However, the MC algorithm does *not* simulate the effect of these interactions. Nevertheless, we believe that the *qualitative* behavior of $q(t)$ we obtain is the correct one for the reason that follows: The Arrhenius law which governs the relaxation of a metastable state is easily shown (at least for a three-level system) to follow from the master equation *regardless* of the details of the dynamics of the system. The numbers depend on the details of the dynamic, but the qualitative (exponential decay) behavior depends only on detailed balance being obeyed. Analogously, we expect that the qualitative aspects of the relaxation exhibited by spin-glasses (SG) depend on detailed balance being satisfied, which is the case for the MC algorithm, and not on the details of the perturbations.

To diagnose whether a critical point exists, it is also of interest to calculate

$$\chi_{\mathrm{SG}}=N^{-1}\sum_{i,j}\langle\sigma_i\sigma_j\rangle^2 , \tag{1.4}$$

which must diverge at T_0 if the associated correlation length ξ diverges there. To make the relation between χ_{SG} and ξ more explicit, let

$$g(\vec{r}_{ij})=N^{-1}\sum_{i,j}{}' \langle \sigma_i\sigma_j\rangle^2 , \tag{1.5}$$

where the primed sum is over all i and j such that $\vec{r}_{ij}$ is fixed. Now, assuming $g(r)\sim r^{-u}f(r/\xi)$,

$$\chi_{SG}=c\int_0^\infty g(\vec{r})d\vec{r} \tag{1.6}$$

yields

$$\chi_{SG}\sim\xi^{d-u} , \tag{1.7}$$

where d is the dimensionality of the system.

The plan of the paper is described next. In Sec. II the systems studied are described in detail, the method of calculation is described (including how we calculate χ_{SG}), and the results obtained, which indicate that both τ and χ_{SG} only diverge as $T\to T_0$ and that $0\leq T_0\lesssim 0.4$, are presented. Finally, the results are discussed in Sec. III.

II. PROCEDURE AND RESULTS

We simulate, by means of a Monte Carlo computation in its METROPOLIS[12] version, an Ising system on a triangular lattice. Each site is occupied with a spin with probability x, each spin interacts with its nearest neighbors only, and it does so antiferromagnetically. The external field is null throughout this paper.

We have studied systems on lattices of 50×50 sites with three values of x: (i) $x=1$, (ii) $x=0.9$ (more precisely, 2240 spins), and (iii) $x=0.74$ (1862 spins). The same computations were repeated for all three values of x for systems on lattices of 30×30 sites to check for size effects.

The lattice is placed on a cylinder, that is, periodic (free) boundary conditions are used in the horizontal (vertical) direction.

The temperature is expressed in units of J/k_B throughout, where J is the magnitude of the exchange constant and k_B is Boltzmann's constant. The time t variable is expressed in units of Monte Carlo steps per spin (MCS/spin).

We next describe how we performed the MC simulations. Let $T_1,T_2,\ldots,T_i$ be the temperatures at which runs were made. In one sequence of runs, all spins are up in the initial state at the highest temperature T_i. The final configuration obtained at T_i was then used as the initial one for T_{i-1}. This procedure was repeated down to the lowest temperature. To test for initial condition effects, runs were also made at all temperatures starting from random-spin orientations. Results obtained from different initial conditions are displayed in the figures as distinct points. At each temperature, we let the system evolve for an equilibration time t_1 before we take note of the value of any quantity. The values of t_1 used are one order of magnitude larger than τ, defined by Eq. (1.3). These values had to be guessed, of course, and checked *a posteriori*. Values of $t_1=3\times 10^5$ MCS/spin were used in some cases at low temperatures ($T\lesssim 0.7$), but $t_1=10^3$ MCS/spin was a typical value used at high ($T\gtrsim 1$) temperatures. MC runs go on to yield average values, after the system becomes equilibrated, for a time approximately equal to t_1.

To compute $q(t)$, defined in Eq. (1), we used Eq. (1.2). Note that the first term in the sum corresponds to $t_n=t_1$ and all other terms are evaluated at later times. Thus $q(t)$ is an *equilibrium* correlation function. Its long time behavior is exhibited in Fig. 2 for a system with $x=0.9$ at several temperatures.

To obtain χ_{SG} it is not practical to find the value of *each* average $\langle\sigma_i\sigma_j\rangle$ in Eq. (1.4) since there are $N(N-1)/2$ of them, which are too many. Instead, we first compute the equilibrium correlation func-

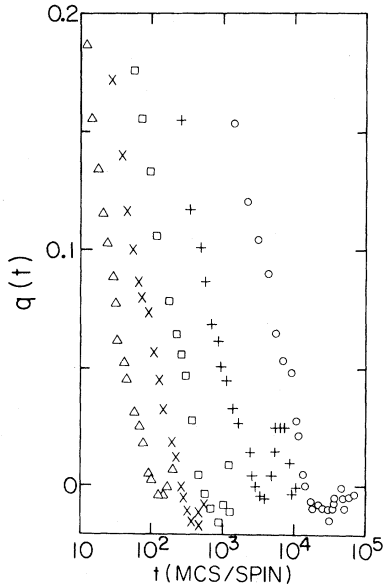

FIG. 2. Long-time behavior of the equilibrium correlation function, $q(t)$, defined in Eq. (1.1) is shown as a function of t for various temperatures for a system with $x=0.9$. ○ is for $T=0.7J/k_B$, + is for $T=0.8J/k_B$, □ is for $T=0.9J/k_B$, × is for $T=J/k_B$, and △ is for $T=1.2J/k_B$.

tion,

$$\chi_{SG}(t)=N^{-1}\sum_{i,j}\langle[\sigma_i(0)\sigma_j(0)] \times[\sigma_i(t)\sigma_j(t)]\rangle , \quad (2.1)$$

where the average $\langle B(0)A(t)\rangle$ is defined by

$$\langle B(0)A(t)\rangle=\frac{1}{m}\sum_{n=1}^{m}B(t_n)A(t_n+t) , \quad (2.2)$$

and t_m+t corresponds to the last MCS/spin taken in the run. Now, since $\langle\ \rangle$ stands for a *time* average in Eq. (1.4), it follows that

$$\chi_{SG}=(2/t_f^2)\int_0^{t_f}(t_f-t)\chi_{SG}(t)dt , \quad (2.3)$$

where t_f is the time interval between t_1 and the end of the run. To obtain $\chi_{SG}(t)$, let

$$A(t_n,t)=\sum_i\sigma_i(t_n)\sigma_i(t_n+t) . \quad (2.4)$$

Then

$$\chi_{SG}(t)=N^{-1}m^{-1}\sum_{n=1}^{m}[A(t_n,t)]^2 , \quad t_m=t_f-t . \quad (2.5)$$

Thus storing m spin configurations (200 in our case), which occur at m equally spaced times throughout the run after t_1, allows $A(t_n,t)$ to be computed.

To speed the convergence of χ_{SG}, as given by Eq. (2.3), to the $t_f\to\infty$ limit, we use

$$\chi'_{SG}=2/(t_f-t')^2\int_{t'}^{t_f}(t_f-t)\chi_{SG}(t)dt , \quad (2.6)$$

where t' is chosen to eliminate the contribution of the nonasymptotic part of $\chi_{SG}(t)$ from the integral. Note that $\chi'_{SG}=\chi_{SG}$ in the $t_f\to\infty$ limit since t' is finite.

Figure 3 exhibits the long-time behavior of $\chi_{SG}(t)$ at various temperatures for a system of 50×50 sites with 90% of them occupied. It shows that $\chi_{SG}(t)$ relaxes to its asymptotic value somewhat faster than $q(t)$.

Figure 4 exhibits the specific heat C per spin (in units of Boltzmann's constant k_B) versus temperature for $x=0.74$ and 0.90. The position of the maxima of C ($T_m\simeq0.9$ in both cases) serves as a reference point for a temperature scale. One set of data points was obtained by numerical differentiation, with respect to temperature, of the mean values of the energy per spin obtained. The other set of points was obtained by applying the formula

$$C=(Nk_BT^2)^{-1}\langle(\delta E)^2\rangle , \quad (2.7)$$

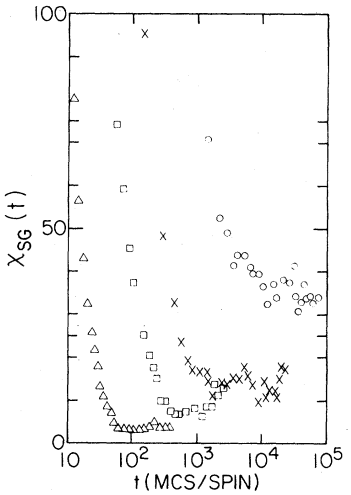

FIG. 3. Long-time behavior of the equilibrium correlation function $\chi_{SG}(t)$, defined in Eq. (1.4), is shown as a function of t (on a logarithmic scale) for various temperatures for a system with $x=0.9$. ○ is for $T=0.7J/k_B$, + is for $T=0.8J/k_B$, □ is for $T=0.9J/k_B$, and △ is for $T=1.2J/k_B$.

where $\delta E=E-\langle E\rangle$. Some scattering in the points shown is observed, since the two methods are only exactly equal for infinite time averages. Values obtained for systems with $x=0.74$ and 0.9 on a smaller lattice of 30×30 sites agreed with the values shown within statistical errors.

Now that we have described how $q(t)$ and $\chi_{SG}(t)$ were computed and the position of the maximum in

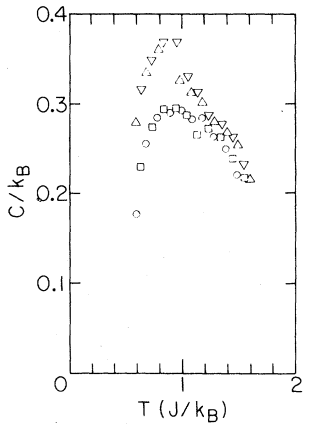

FIG. 4. Specific heat (in units of k_B) is shown as a function of T (in units of J/k_B) for systems of atomic concentration $x=0.74$ and 0.9 on 50×50 sites. The MC method was used to obtain these values. △ and ○ are computed for $x=0.74$ and 0.9, respectively, using Eq. (2.7). ▽ and □ are computed for $x=0.74$ and 0.9, respectively, using $C=\Delta\langle E\rangle/\Delta T$.

the specific heat has been established to serve as a reference point in temperature, we turn our attention to our results obtained for (a) the relaxation times, defined in Eq. (1.3), and for (b) χ_{SG}, defined in Eq. (1.4).

Figures 5, 6, and 7 exhibit $\ln[\ln(\tau)]$ vs T on a logarithmic scale, for $x=0.74$, 0.9, and 1, respectively. These results, obtained for systems on 50×50 sites, are fitted with the rule[13] (Fulcher's law)

$$\tau=\exp[A_t/(T-T_0)^{C_t}] . \tag{2.8}$$

The dotted (dashed) lines are least-squares fits with $T_0=0$ ($T_0=0.4$), and the corresponding values of A_t and C_t are given in Table I. While the value of $T_0=0$ seems to give better fits for all three values of x, the value $T_0=0.4$ cannot be ruled out entirely. A value of $T_0=0.5$ gives significantly worse fits for all three values of x. Results obtained for systems on 30×30 sites are also shown for $x=0.74$ and 0.9.

Figures 8, 9, and 10 exhibit $\ln(\chi_{SG})$ vs T for $x=0.74$, 0.9, and 1, respectively. These results, obtained for systems on 50×50 sites, are fitted with the rule

$$\chi_{SG}=\exp[A_x/(T-T_0)^{C_x}] . \tag{2.9}$$

The dotted (dashed) lines are least-squares fits with $T_0=0$ ($T_0=0.4$), and the corresponding values of A_x and C_x are given in Table I. The value $T_0=0$ gives a better fit for $x=0.74$, but $T_0=0$ and $T_0=0.4$ seem to give equally good fits for $x=0.9$ and 1 in this case. On the other hand, a value of $T_0=0.5$ gives significantly worse fits for all three values of x. Results obtained for systems on 30×30 sites are also shown for all concentrations.

Figures 8 and 9 show that χ_{SG} becomes size dependent for $\ln\chi_{SG}\gtrsim3$ both for $x=0.74$ and for $x=0.9$, which was to be expected since ξ becomes comparable to the linear dimensions of the system then. This effect produces the size dependence of

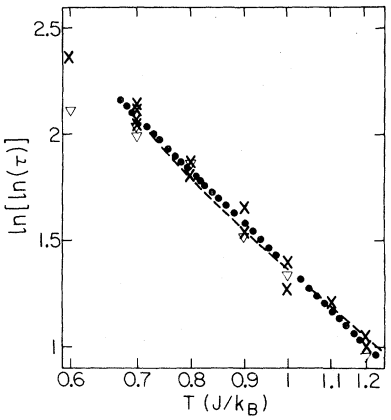

FIG. 6. Quantity $\ln[\ln(\tau)]$ is shown vs temperature, on a logarithmic scale for systems with $x=0.9$ on 50×50 sites ($\times$), and on 30×30 sites (∇). The relaxation τ is defined in Eq. (1.3). The dotted (dashed) line is a least-squares fit with Eq. (2.8) and $T_0=0$ ($T_0=0.4$). The point corresponding to $T=0.6$ was not taken into account when finding the best fit because there is a significant size dependence then.

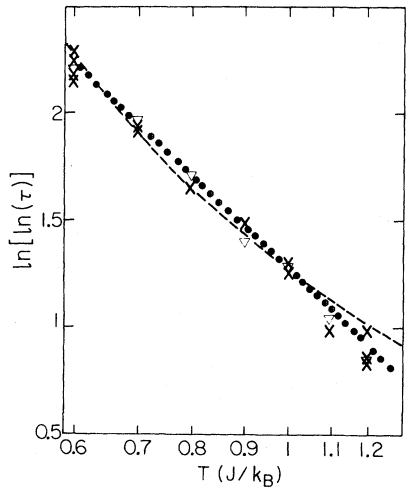

FIG. 5. Quantity $\ln[\ln(\tau)]$ is shown vs temperature on a logarithmic scale for systems with $x=0.74$ on 50×50 sites ($\times$), and on 30×30 sites (∇). The relaxation τ is defined in Eq. (1.3). The dotted (dashed) line is a least-squares fit with Eq. (2.8) and $T_0=0$ ($T_0=0.4$).

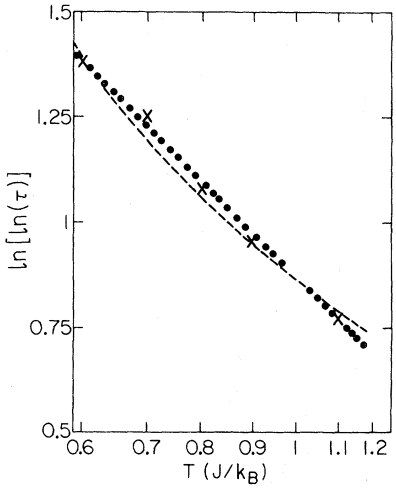

FIG. 7. Quantity $\ln[\ln(\tau)]$ is shown vs temperature (in units of J/k_B) on a logarithmic scale, for a system with $x=1$ on 50×50 sites. The relaxation τ is defined in Eq. (1.3). The dotted (dashed) line is a least-squares fit with Eq. (2.8) and $T_0=0$ ($T_0=0.4$).

TABLE I. Values of constants, defined in Eqs. (2.8) and (2.9), corresponding to $T_0=0$.

x	Number of sites	C_x	C_t	A_x	A_t
1.0	50×50	1.03	1.01	1.37	2.37
0.75	50×50	1.84	1.95	1.44	3.52
0.75	30×30	1.57	1.93	1.50	3.51
0.9	50×50	2.26	2.02	1.58	3.95
0.9	30×30	1.78	1.98	1.41	3.83

C_x shown in Table I. It becomes quite large in the case of $x=0.9$, for $T\lesssim 0.6J/k_B$, as Fig. 9 shows; accordingly, the points for $T=0.6$, $x=0.9$ were not taken into account when finding the best fits.

III. SUMMARY AND DISCUSSION

We have studied Ising systems on a triangular lattice with atomic concentrations given by $x=0.74$, 0.9, and 1. The case of $x=0.9$ is interesting because, as Fig. 1(a) shows, the very low-temperature ($T=0.3J/k_B$) entropy as a function of x has its minimum value near $x=0.9$, which suggests that if there is a transition into an ordered state for some range of values of x, $x=0.9$ should be within that range.

The results obtained for the relaxation time τ and for the generalized susceptibility χ_{SG} are *consistent* with a vanishing critical temperature T_0 for $x=0.74$, 0.9, and 1. Both τ vs T and χ_{SG} vs T can be fitted by exponential functions, given by Eqs. (2.8) and (2.9) (setting $T_0=0$), respectively, for all values of x studied. The values of C_t and C_x, defined by these two equations, corresponding to $x=0.74$ and 0.9, are about twice as large as their values obtained for $x=1$, as shown in Table I. This is the only remarkable difference we see between the case of $x=1$ and the dilute case. Grest and Gabl[6] had already reported that these dilute systems become very slow for $T\lesssim 0.7$. Equation (1.7) implies that the correlation length (ξ) diverges with the same index C_x as χ_{SG}, and the value shown in Table I for $x=1$ agrees approximately with the known value.[14]

A nonvanishing T_0 cannot be entirely ruled out since the values obtained for τ and for χ_{SG} can also be fitted by Eqs. (2.8) and (2.9) for any value of T_0

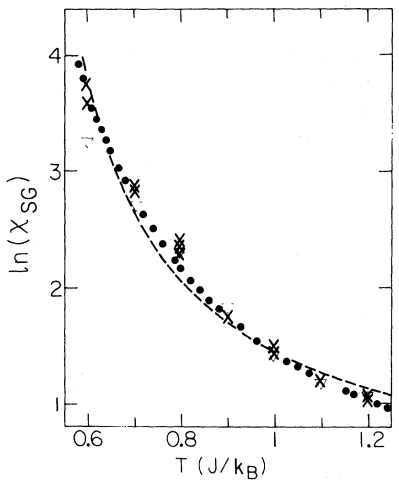

FIG. 8. Quantity $\ln(\chi_{SG})$ is shown vs temperature for a system with $x=0.74$ on 50×50 sites (×), and on 30×30 sites (∇). χ_{SG} is defined in Eq. (1.4). The dotted (dashed) line is a least-squares fit with Eq. (2.9) and $T_0=0$ ($T_0=0.4$).

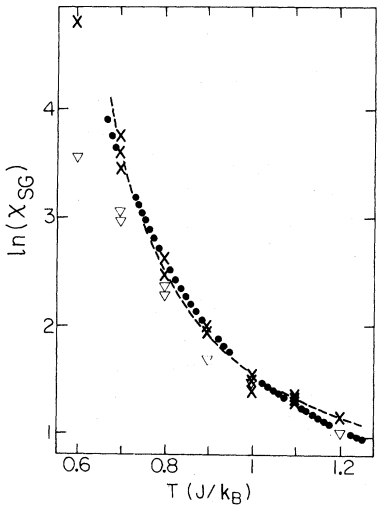

FIG. 9. Quantity $\ln(\chi_{SG})$ is shown vs temperature for a system with $x=0.9$ on 50×50 sites (×), and on 30×30 sites (∇). χ_{SG} is defined in Eq. (1.4). The dotted (dashed) line is a least-squares fit with Eq. (2.9) and $T_0=0$ ($T_0=0.4$).

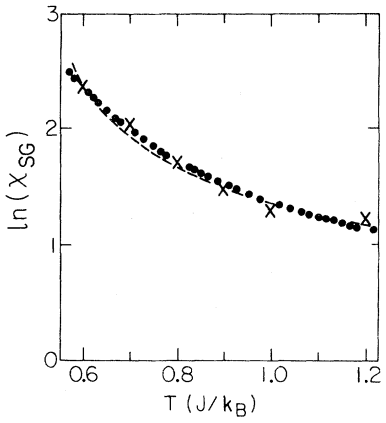

FIG. 10. Quantity $\ln(\chi_{SG})$ is shown vs temperature (in units of J/k_B) for a system with $x=1$ on 50×50 sites. χ_{SG} is defined in Eq. (1.4). The dotted (dashed) line is a least-squares fit with Eq. (2.9) and $T_0=0$ ($T_0=0.4$).

satisfying $T_0 \lesssim 0.4J/k_B$. Note, however, that $0.4J/k_B$ is a rather small temperature, since (a) it is less than a half of the value of the temperature where the maximum in C occurs (see Fig. 4), and (b) that C is very small [see Fig. 1(b) for $T \lesssim 0.4$]. We next remark on why it is difficult to rule out, by MC simulations, the existence of a transition at some T_0 in the range $0 < T \lesssim 0.4$ for $x \leq 1$. As Figs. (5) and (6) show, τ increases extremely rapidly as T decreases. Extrapolation yields $\tau \approx 10^7$ at $T=0.5$ for $x=0.9$, and the numbers of MCS/spin taken must be larger than τ to get meaningful results. Clearly, it is impractical to do MC simulations for $T \lesssim 0.5$. There is one additional reason why it is unlikely that *any numerical* method can rule out the possibility of a transition at any $T \lesssim 0.3$. As Fig. (1) shows, $C \approx 0$ for such low temperatures, which makes it difficult to distinguish numerically $T=0.2$, for example, from $T=0$. This is in marked contrast with other spin-glass models, in which $T^{-1}C$ does not vanish as $T \to 0$.

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