Hybrid Monte Carlo spin-dynamics simulation of metallic spin-glass alloys

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(Received 29 June 1992)

We study a dilute system of the isotropic classical Heisenberg spins coupled via the Ruderman-Kittel-Kasuya-Yosida interaction using a hybrid Monte Carlo spin-dynamics method. We find strong evidence of the occurrence of a spin-glass phase at finite, nonzero temperatures, contrary to previous predictions. We suggest that the spin-glass phase of this system can be characterized by a power-law decay of spin correlations.

I. INTRODUCTION

Dilute magnetic alloys such as *Cu*Mn and *Au*Fe have been extensively investigated in the last decade. The experimental consensus has been that the spin-glass transition observed in these alloys is a thermodynamic phase transition.[1] To explain this, a dilute magnetic system in which the Heisenberg spins are coupled via the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction has been investigated both analytically and numerically. However, the low-temperature properties of the model are not yet understood. In particular, even whether or not the spin-glass phase occurs in the isotropic RKKY model has remained controversial. Theoretical studies[2,3] have predicted that the RKKY model is at its lower critical dimension $d_l=3$, and anisotropy, which always exists in real materials, induces the spin-glass transition at a finite, nonzero temperature. Although the prediction has been widely accepted, it is not very reliable because these studies were made based on the replica method.[4] Numerical studies have also been made by various authors: Walstedt and Walker[5] suggested that the model does not show any transition at finite temperatures without anisotropy. Their results cannot be considered conclusive, however, because they calculated only the spin-glass order parameter, which always vanishes in finite systems because of a uniform rotation of all the spins. Fernandez and Streit[6,7] calculated the spin-glass susceptibility of the model with up to 169 spins and suggested that a phase transition occurs at a finite, nonzero temperature even when the anisotropy is absent. Chakrabarti and Dasgupta[8,9] also calculated the spin-glass susceptibility of the same model with up to 312 spins and suggested that, from a finite-size scaling analysis, $T_c=0$ and the correlation length diverges as a power law. They also suggested that a weak anisotropy induces the spin-glass phase transition at a finite, nonzero temperature.[9,10] On the other hand, Reger and Young[11] studied a *RKKY-like* model with up to 4096 spins, where every lattice site is occupied by the Heisenberg spin and the strength of the interactions falls off with the inverse third power of the distance between the spins, and obtained a result which is not incompatible with the prediction of $d_l=3$; i.e., the correlation length diverges exponentially at $T_c=0$. However, their result also was not conclusive because they could not rule out the possibility of $T_c\neq 0$. Moreover, their model is different from the RKKY model.

It is the purpose of the present paper to discuss whether or not a spin-glass phase occurs in the RKKY model without anisotropy. We make a computer simulation of the model using a hybrid Monte Carlo spin-dynamics (HMCSD) method,[12] which was proved to be very effective for studying the low-temperature properties of $\pm J$ Heisenberg models.[13,14] The method is also very effective in this model and enables us to simulate it with up to 1600 spins within reasonable computer CPU time. We find from a finite-size scaling analysis that, *even when anisotropy is absent, the spin-glass susceptibility diverges at a finite, nonzero temperature* T_c, *contrary to the previous prediction.* The spin-glass phase realized below T_c is suggested to be characterized by a power-law decay of spin correlations, like that in the two-dimensional *XY* model.[15] A brief report of this result is given in Ref. 16.

In Sec. II the model and method are described. Results are presented in Sec. III. Section IV is devoted to conclusions and discussions.

II. MODEL AND METHOD

We start with the model described by the Hamiltonian

$$H=-J_0\sum_{i<j}[\cos(2k_F r_{ij})/r_{ij}^3]\mathbf{S}_i\cdot\mathbf{S}_j \ , \tag{1}$$

where the $\mathbf{S}_i$'s are classical Heisenberg spins of $|\mathbf{S}_i|=1$ randomly distributed on the sites of an $L\times L\times L$ fcc lattice with concentration c, J_0 is an energy constant, k_F is the Fermi wave vector of the host metal, and r_{ij} is the distance between the ith and jth spins. Parameters of the interaction are chosen representing *Cu*Mn, i.e., $k_F a_0=4.91$, where a_0 is the lattice constant. The temperature T is measured in units of $J_0/a_0^3 k_B=1$. Periodic boundary conditions are imposed. Since the exchange interaction decreases oscillatorily as a power of $(r_{ij})^{-3}$, each of the spins is not affected strongly from the individual spins separated far from it. Then we consider the interactions only between the spins for $|r_{ij}|<r_0$. Of course, r_0 should be chosen to be long enough for each spin being coupled by a large number of the spins, and the sum of

the contributions from the other spins being almost offset, even when all the spins point the same direction. To determine r_0, we calculate the sum $R(r)$,

$$R(r) = -\sum_{|r_{0i}|\geq r} J(r_{0i}) . \tag{2}$$

where $J(r_{ij})$ is the coupling constant between the spins on the ith and jth lattice sites. As seen in Fig. 1, $R(r)$ almost vanishes for $r/a_0 = 3.0, 3.7, 4.2, \ldots$. Since already for $r/a_0 = 3.0$ the number of the lattice sites for $|r_{ij}| < r_0$ is about 400, we chose $r_0 = 3.0a_0$ for simplicity. We believe this will not affect the conclusion given here that a spin-glass phase transition occurs in this model.

The simulation is made using the HMCSD method[12] with $t_0 = 0.2$ measured in units of $\hbar a_0^3/J_0 = 1$. Special attention is paid to whether or not the system reaches its thermal equilibrium. To examine it, we consider the spin-glass susceptibility χ_{SG} defined by

$$\chi_{SG} = \frac{1}{N}\sum_{i,j}\langle\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle_T^2\rangle_c , \tag{3}$$

where $\langle\cdots\rangle_T$ is the thermal average and $\langle\cdots\rangle_c$ means the sample average. We calculate it in two different ways as follows.[17] After K_i Monte Carlo steps per spin (MCS) are discarded, data of the next K MCS were used to calculate the average. But the average is taken first over the former $K/2$ MCS, and then it is taken over the latter $K/2$ MCS, which are denoted as $\langle\cdots\rangle_{T1}$ and $\langle\cdots\rangle_{T2}$, respectively. We first estimate χ_{SG} from χ_{SG}^u, where

$$\chi_{SG}^u = \frac{1}{N}\sum_{i,j}\langle\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle_{T1}\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle_{T2}\rangle_c . \tag{4}$$

The other estimation is made as follows. We prepare two lattices A and B with the same distribution of spins. The simulation is made starting from different initial spin configurations. After K_i MCS are discarded, the averages are taken independently over K MCS, which are denoted as $\langle\cdots\rangle_T^A$ and $\langle\cdots\rangle_T^B$, respectively. Second, we estimate χ_{SG} from χ_{SG}^l, where

$$\chi_{SG}^l = \frac{1}{N}\sum_{i,j}\langle\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle_T^A\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle_T^B\rangle_c . \tag{5}$$

When K is not large enough, χ_{SG}^u will be larger than χ_{SG} because some correlation still remains between the two averages of $\langle\cdots\rangle_{T1}$ and $\langle\cdots\rangle_{T2}$. When K_i is not large enough, χ_{SG}^l is smaller than χ_{SG} because the systems do not reach their thermal equilibrium, which will be the same. Only when χ_{SG}^l and χ_{SG}^u coincide with each other can we regard them as the equilibrium value. In Fig. 2 a typical example of these quantities is presented together with those calculated by using the conventional MC method. The thermal equilibrium is already reached for $K_i \sim 1000$. We need only several thousands of MCS to get the equilibrium value. In contrast to this, in the MC method, we could not equilibrate the system within reasonable MCS.

The simulation is made in the case of $c = 5.0$ at. %. The linear sizes of the lattice are $L = 6-20$; i.e., the numbers of the spins are about $N = 43-1600$. The numbers of the samples prepared in this simulation are 47, 36, 36, 16, 10, and 6 for $L = 6$, 8, 10, 12, 16, and 20, respectively. We use the set of (K_i, K) as (2000,4000) for $L \leq 10$, (4000,8000) for $L = 12$, (5000,10 000) for $L = 16$, and (6000,15 000) for $L = 20$.

We calculate such physical quantities as the energy, specific heat, magnetization, and susceptibility as well as the spin-glass susceptibility to study equilibrium properties of the model. We also calculate the spin-glass order parameter $Q(K)$,[18,19]

$$Q(K) = \frac{1}{N}\sum_{i}\langle\langle \mathbf{S}_i\rangle_T^2\rangle_c , \tag{6}$$

to study time-dependent properties of the model.

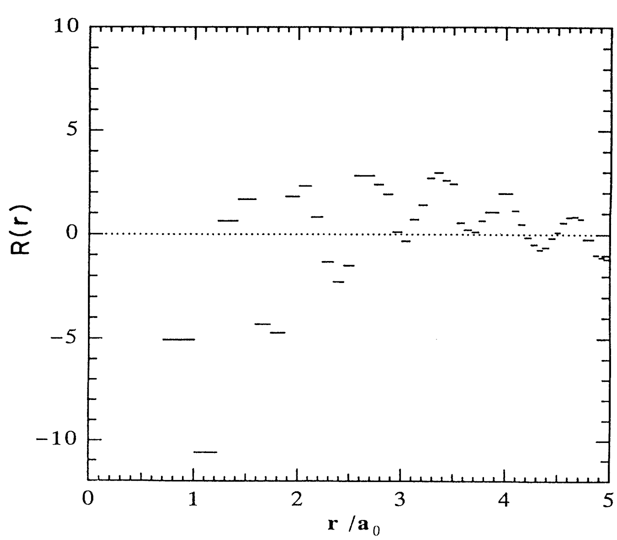

FIG. 1. Sum of the maximum contribution $R(r)$ from the spins ignored in the simulation.

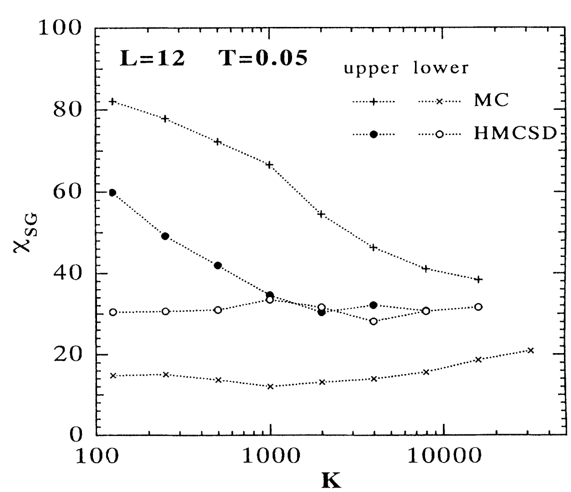

FIG. 2. Upper and lower bounds of the spin-glass susceptibility.

III. RESULTS

A. Energy and specific heat

We first present results of the thermodynamic quantities. In Fig. 3 the energy is shown for different sizes of the lattice. The energy for smaller lattices exhibits a large sample dependence. This is because the energy depends largely on the number of nearest-neighbor spin pairs because the nearest-neighbor coupling is extremely larger than the other couplings. The deviation of the number is proportional to $\sqrt{N}$, which leads to a deviation of the energy per spin of the order of $J_0/\sqrt{N}$, where N is the number of spins. The specific heat is shown in Fig. 4. It exhibits a broad maximum around $T \sim 0.07$. Although the height of the maximum increases a little with increasing lattice size, its shape remains broad.

B. Magnetization and susceptibility

Since the average of the magnetization $\mathbf{M}=\sum_i \mathbf{S}_i$ vanishes as K increases because of a uniform rotation of all the spins, we calculate the average of its absolute value $|\mathbf{M}|$. The results for different sizes of the lattice are shown in Fig. 5. These are almost constant in the temperature and decrease in proportion to $1/\sqrt{N}$ as the size of the lattice increases. This clearly reveals the absence of the ferromagnetic phase. Moreover, the above fact indicates that the spin directions are uniformly distributed. We also calculate the susceptibility. Because of a very slow fluctuation of the magnetization, we could not get any equilibrium value at low temperatures within the MCS performed in this simulation. However, we may expect that the susceptibility exhibits a size-independent Curie law of

$$\chi=\frac{1}{3NT}\langle\langle \mathbf{M}^2\rangle_T\rangle_c$$
$$\propto \frac{1}{3T}(\langle\langle|\mathbf{M}|\rangle_T\rangle_c/\sqrt{N})^2 , \quad (7)$$

because $\langle \mathbf{M}\rangle_T=0$ will hold for $K\to\infty$. To confirm this, we calculate the susceptibility assuming $\langle \mathbf{M}\rangle_T=0$ for $L=10$ and present it in Fig. 6 together with that given by Eq. (7) with $\langle\langle|\mathbf{M}|\rangle_T\rangle_c/\sqrt{N}\sim 1.85$, which is estimated from the results in Fig. 5. The agreement is fairly good. It should be mentioned that this fact never means the absence of the phase transition because the Curie law also

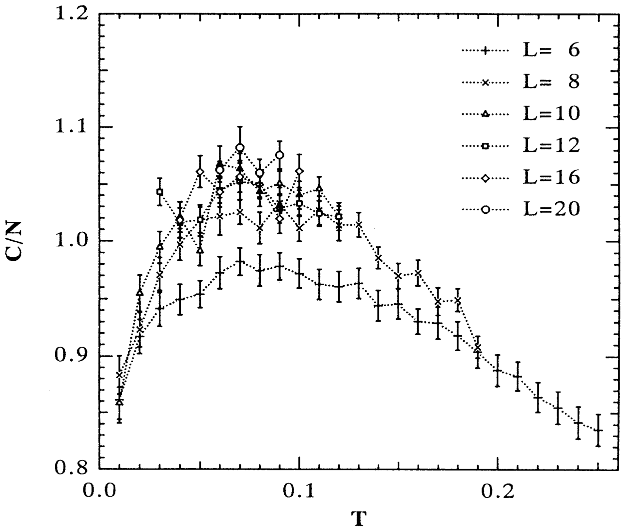

FIG. 4. Temperature dependences to the specific heat for different sizes of the lattice. Error bars indicate probable errors of averaged values.

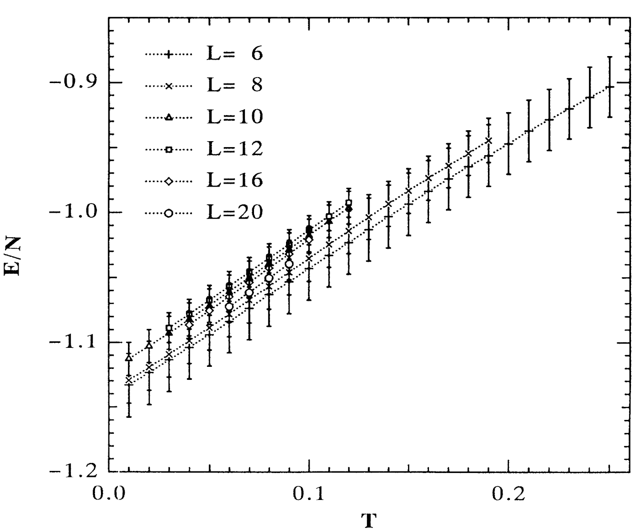

FIG. 3. Temperature dependences of the energy for different sizes of the lattice. Error bars indicate probable errors of averaged values.

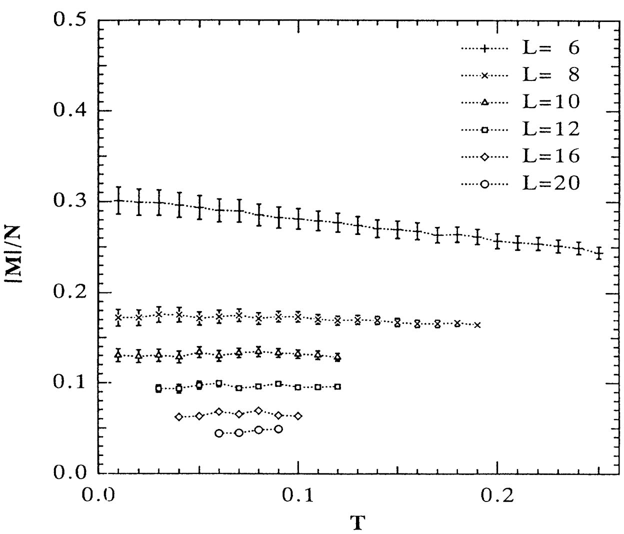

FIG. 5. Temperature dependences to the absolute magnetization for different sizes of the lattice. Error bars indicate probable errors of averaged values.

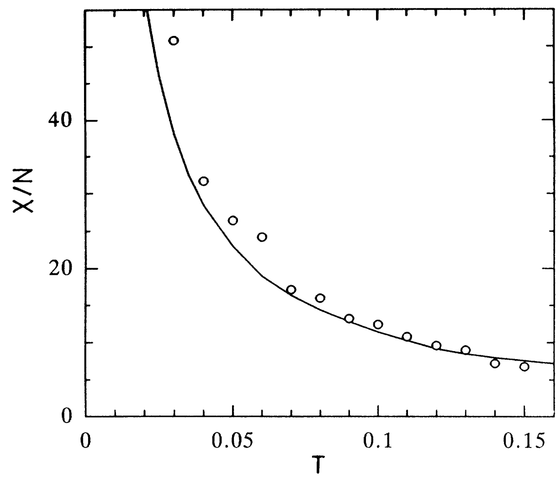

FIG. 6. Temperature dependence of the susceptibility for $L = 10$. The solid curve represents the result of Eq. (7).

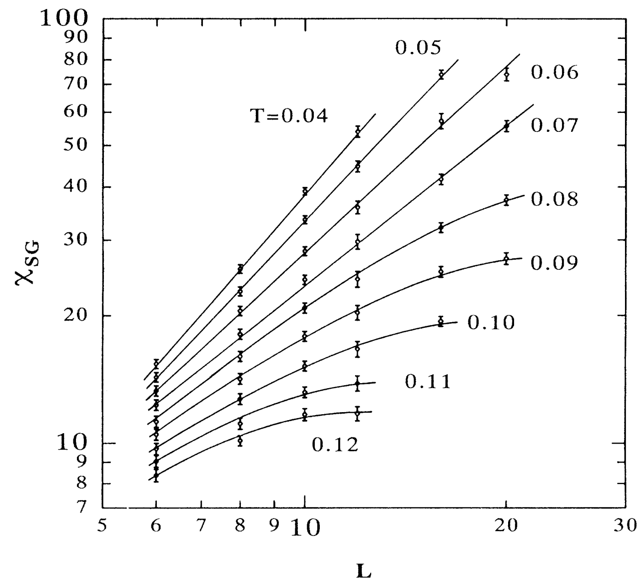

FIG. 8. Lattice size dependences of χ_{SG} at different temperatures. Error bars indicate probable errors of averaged values, and lines are guides for the eye. The slope of the line at $T=0.07$ is about 1.25, which leads to $\eta \sim 2-1.25=0.75$.

results from a fluctuation of the total magnetization. To see whether or not the phase transition occurs, we calculate the magnetization in a small magnetic field. This is planned to be discussed in a separate paper.

C. Spin-glass susceptibility

In Fig. 7 we present the temperature dependence of χ_{SG} for different sizes of the lattice. At low temperatures, χ_{SG} exhibits a marked size dependence. In Fig. 8 the data are plotted as a function of L in a log-log form for different temperatures. For $T \geq 0.08$, the results reveal an exponential decay of the pair-spin correlations $\langle\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle_T^2 \rangle_c$ with increasing r_{ij}. On the other hand, for $T \leq 0.07$, the data seem to lie on a straight line, suggesting a power-law decay of the correlations. We expect that a phase transition takes place at $T \sim 0.07$.

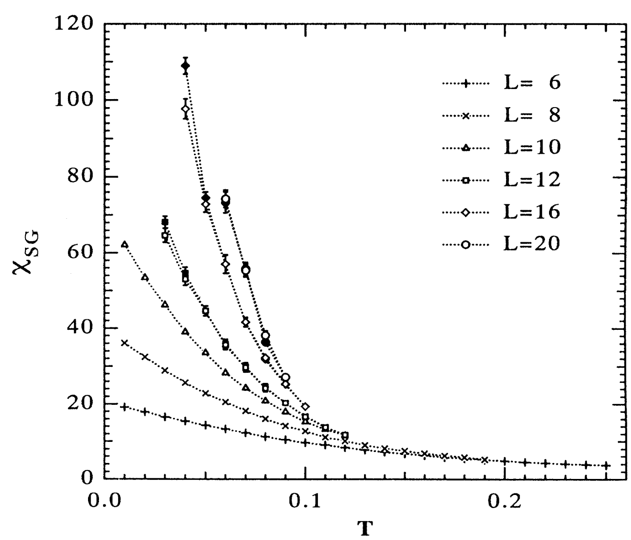

FIG. 7. Temperature dependence of χ_{SG} for different sizes of the lattice. Open and solid symbols represent lower and upper bounds of χ_{SG}, respectively. Error bars indicate probable errors of averaged values.

To confirm this, we make a finite-size scaling plot.[20] If $d_l < 3$ and the phase transition occurs at some finite temperature T_c, the pair-spin correlation functions may be expressed using two critical exponents η and ν as

$$\langle\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle_T^2 \rangle_c \sim \frac{\exp(-r_{ij}/\xi)}{r_{ij}^{d-2+\eta}} \quad (T \gtrsim T_c) , \tag{8}$$

where $d=3$ and the correlation length ξ is given by

$$\xi \sim (T-T_c)^{-\nu} . \tag{9}$$

The finite-size scaling hypothesis predicts that χ_{SG} will behave as

$$\chi_{SG} = L^{2-\eta} \chi(L^{1/\nu}(T-T_c)) . \tag{10}$$

The scaling plot is shown in Fig. 9. When we put $T_c = 0.068 \pm 0.008$, we can scale all the data well. Values of the exponents determined in this analysis are $\eta = 0.69 \pm 0.20$ and $\nu = 0.74 \pm 0.05$. T_c obtained here is compatible with that suggested from the size dependence of χ_{SG}, and the value of $\eta \sim 0.7$ is also compatible with $\eta \sim 0.75$ estimated from the slope of the plot in Fig. 8. Note that we also examined the possibility of $T_c = 0$ and found that the data cannot be scaled well even when different types of the scaling functions are assumed.[16] We may conclude, hence, that the spin-glass phase transition occurs at $T_c \sim 0.068$.

It is desirable to know the nature of the spin-glass phase realized in this model. Our results presented in Fig. 8 strongly suggest that for $T < T_c$ long-range order is absent, but the pair-spin correlations decay according to the power law, i.e., $\xi = \infty$ and $\eta > -1$ in Eq. (8), like

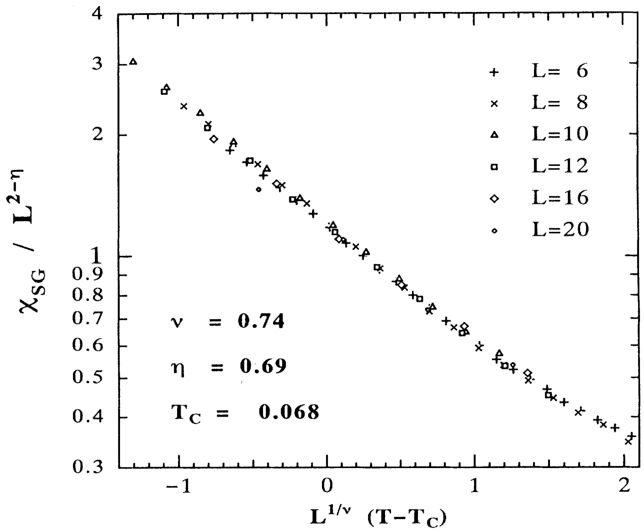

FIG. 9. Finite-size scaling of χ_{SG}.

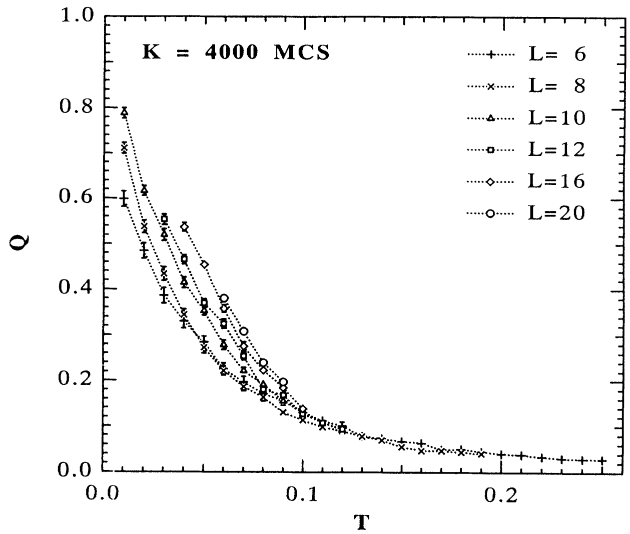

FIG. 11. Temperature dependences of the order parameter $Q(K)$ for different sizes of the lattice. Error bars indicate probable errors of averaged values.

those in the Kosterlitz-Thouless phase[15] in the two-dimensional xy model. We roughly estimate values of η for $T<T_c$ from the slopes of the lines and present them in Fig. 10. Note that η decreases almost linearly with temperature and seems to reach a certain value greater than -1 at $T=0$. This suggests that the ground state of the model is degenerate, as predicted by Walker and Walstedt.[21]

D. Order parameter $Q(K)$

Next, we consider the Edwards-Anderson order parameter $Q(K)$. In finite systems, this quantity will vanish for large enough MCS as a result of the uniform rotation even when some long-range order is realized. Here we consider its K dependence to study temporal properties of the model. In Fig. 11 we present the temperature dependence of $Q(K)$ for different sizes of the lattice. As the temperature is decreased, $Q(K)$ increases, indicating a slowing down of the fluctuation of the spin structure. For $T<0.09$, the size dependence becomes considerable. This is because the spin correlations develop at low temperatures, as seen in Sec. III C. In Figs. 12(a) and 12(b), $Q(K)$ are plotted as functions of K at both higher and lower temperatures of T_c, respectively. Even at $T<T_c$, $Q(K)$ decreases as K increases. At low temperatures, a

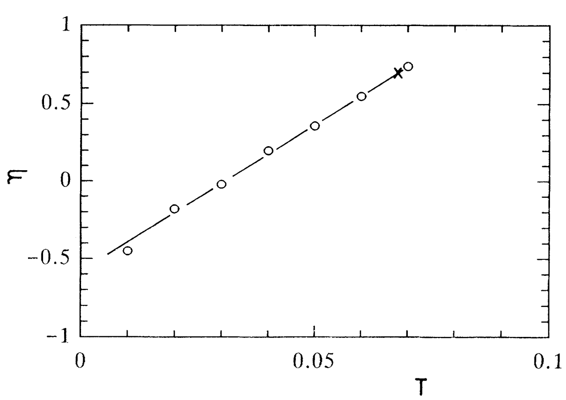

FIG. 10. Value of η estimated from the slope of the line in Fig. 8. The symbol $\times$ is the value obtained from the scaling analysis in Fig. 9.

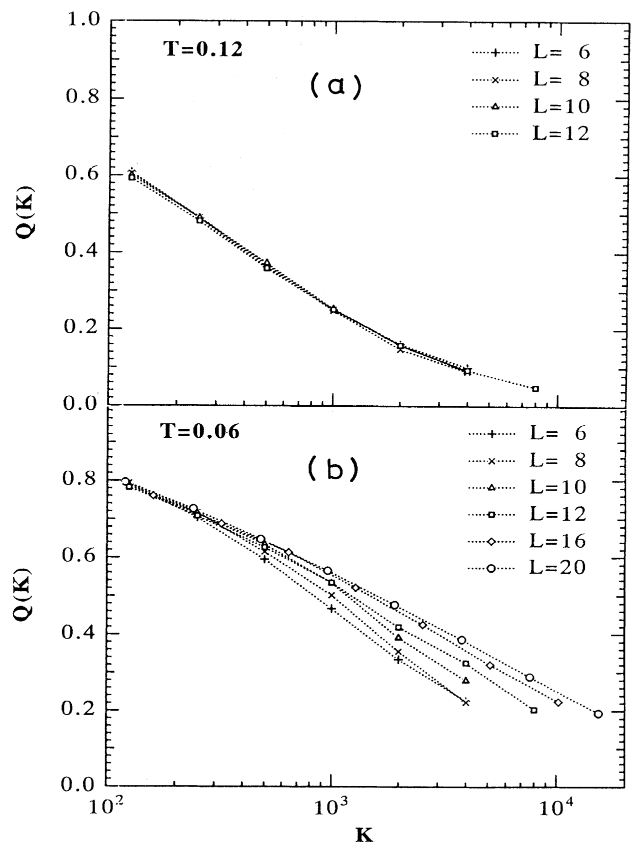

FIG. 12. K dependences of $Q(K)$ for different sizes of the lattice: (a) those in the paramagnetic phase and (b) those in the spin-glass phase.

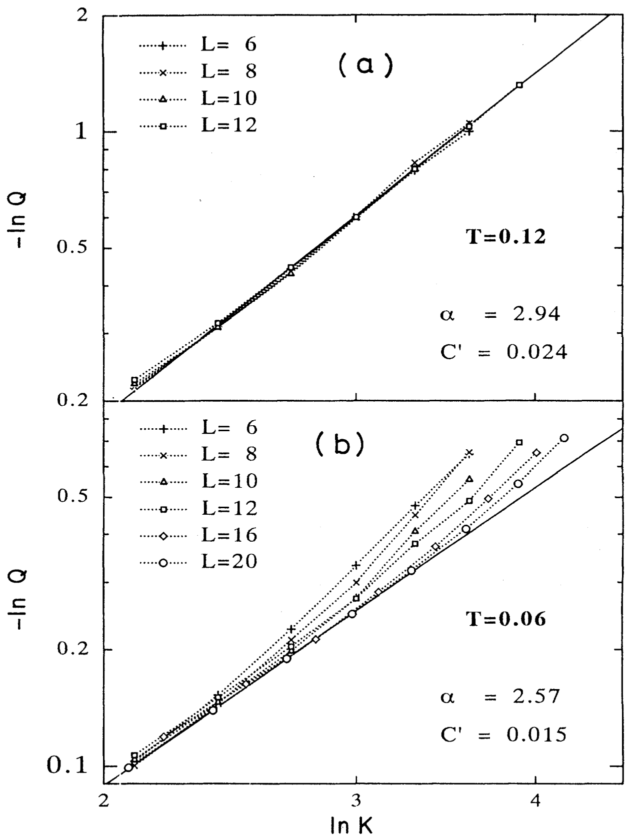

FIG. 13. K dependences of $Q(K)$ plotted in different scales. The data in (a) and (b) are the same as those in (a) and (b) in Fig. 12, respectively. Here $C' = C(\ln 10)^{\alpha-1}$.

size dependence is seen even for larger lattices, but not very large. This size dependence supports our suggestion that the correlations decay according to the power law. Because if some long-range order occurs, the system would exhibit a large size dependence for some characteristic time scale. And if the spin correlations decay exponentially, the size dependence would vanish rapidly as the lattice size is increased.

To see the K dependence quantitatively, we examine several functions of K and find that the data at high temperatures can be fitted well by the function

$$Q(K) \sim \exp[-C(\ln K)^{\alpha}] . \tag{11}$$

which is shown in Fig. 13(a). At low temperatures, as the size of the lattice is increased, the data seem to approach to the same function with different values of C and α, as seen in Fig. 13(b). We suggest, hence, that the relaxation of $Q(K)$ is described by Eq. (11) for both higher and lower temperatures of T_c. Of course, further studies are necessary to confirm this suggestion.

IV. CONCLUSIONS AND DISCUSSIONS

We have studied the equilibrium behavior in a dilute model of the classical Heisenberg spins coupled via the RKKY interaction using the HMCSD method. We have found that, *even when anisotropy is absent, the model exhibits a spin-glass phase transition at a finite, nonzero temperature.* We have suggested that the spin-glass phase of this model is characterized by a power-law decay of the spin correlations, like that in the Kosterlitz-Thouless phase in the two-dimensional xy model. We have also studied time-dependent behaviors of the model, calculating the order parameter $Q(K)$, and found that the spin structure slowly changes even below the transition temperature.

Our finding is incompatible with the idea widely accepted that finite-range Heisenberg models do not exhibit any spin-glass phase at finite, nonzero temperatures without anisotropy. Heisenberg spin-glass models extensively studied so far are, however, the *short-range bond models,*[13,14,22–24] in which different bonds are randomly distributed between neighboring spins. In contrast with those models, the RKKY model is a *long-range site model* in which the coupling constant between the spins is uniquely determined by their relative positions. The spin-glass phase will be stabilized either by the long-range nature of the interactions or by the effect of the site dilution. One has thought that, if a difference between the RKKY model and short-range spin-glass model exists, it would come from the difference in the interaction range.[2,3] In fact, Reger and Young[11] studied a bond model with long-range interactions and found that the model is in a different universality class from short-range models. They suggested, however, that the spin-glass phase does not appear at finite, nonzero temperatures. If their suggestion is true, the spin-glass phase would be stabilized by the effect of the site dilution. We think this is very probable because the distribution of bonds around each of the spins depends on the distribution of the magnetic atoms, and hence frustration is partly relieved.[25] Especially, in the RKKY model, different clusters of strongly coupled spins are formed everywhere in the lattice. Frustration will occur between the spin arrangements of those clusters. Hence frustration in the RKKY model will be much weaker than that in the bond models. To confirm this, it is desirable to study site models with short-range interactions. It is to be noted that in this paper we have examined the spin ordering in a rather dense RKKY model, i.e., $c=5.0$ at. %, in which the clustering effect will be enhanced. It is interesting to examine whether or not the spin-glass phase also occurs in more dilute RKKY models. This problem is currently under study, and the results are planned to be reported elsewhere.

Finally, we must emphasize that many spin-glass materials are well described by site models and the present study has predicted that a prototype of the spin-glass models exhibits the spin-glass phase at finite, nonzero temperatures. It is very interesting to study also different realistic spin-glass models such as a dilute Heisenberg model with only the ferromagnetic nearest-neighbor and antiferromagnetic next-nearest-neighbor interactions, i.e., a proposed model of $Eu_xSr_{1-x}S$.[26]

ACKNOWLEDGMENTS

This work was supported by a Grant-in-Aid Scientific Research on Priority Areas. "Computational Physics as a New Frontier in Condensed Matter Research," from the Ministry of Education, Science and Culture, Japan.

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