Fluctuating clusters in a reentrant spin-glass system

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(Received 30 September 2006; revised manuscript received 22 December 2006; published 12 April 2007)

We theoretically examined the reentrant spin-glass transition in a dilute ferromagnetic (FM) system. Results show that *clusters of fluctuating spins exist in the FM phase*. As the temperature is decreased, the spins in those clusters freeze in conjunction with all other spins in the system. This freezing breaks up the FM order into ferromagnetic clusters. These results are compatible with those of a phenomenological random field model that was proposed for experimentation.

DOI: 10.1103/PhysRevB.75.144413 PACS number(s): 75.10.Nr, 75.10.Hk

I. INTRODUCTION

Reentrant spin-glass (RSG) transition is a well-known phenomenon of spin glasses (SGs). The RSG transition is found nearby, but it is on the ferromagnetic (FM) side of the phase boundary between the SG phase and the FM phase.[1,2] As the temperature is decreased from a higher temperature, the magnetization increases; it then ceases at a lower temperature. Finally, the SG phase, which is characterized by FM clusters, is realized. However, no theoretical model has yet been given that explains the RSG transition. A phenomenological random-field (RF) model was proposed to explain experimental observations of quasielastic neutron scattering of RSG systems.[3–5] The essential point of that conception is the decomposition of the system into a FM part and a part with frustrated spins (SG part). At low temperatures, the spins of the SG part freeze yielding random fields to the spins of the FM part, and the FM phase is destroyed due to the random-field effect.[6] Evidence of decomposition into two parts was given in various experiments of both quasielastic[3–5,7,8] and inelastic[7–11] neutron scatterings, of Mössbauer measurements,[12] and of time variations of the magnetization.[13] However, no evidence of the RF hypothesis has yet been given from a microscopic perspective.

The purpose of this paper is to examine the RF model of the RSG transition. A special attention is paid to whether the system is really decomposed into two parts or not and, if it is decomposed, whether it occurs due to a clustering effect of magnetic atoms or scattering, which inevitably occurs in random magnets. We theoretically examine the RSG transition in a dilute ferromagnet.[14,15] Results show that the system is, in fact, decomposed into a FM part and a SG part, together with an interface (IF) part between them. Majority spins belong to the FM part and form a network over the lattice. The SG part consists of isolated clusters that are located in domains with a lower spin concentration. As the temperature is decreased from a high temperature, the spins in the FM part order ferromagnetically, whereas the spins in the SG part are weakly magnetized under the influence of the FM order in the FM part. As the temperature is decreased further, the spins in the SG part freeze in cooperation with all other spins in the system; subsequently, the FM order breaks up into FM clusters. We conclude that the RF model is a concept that can explain the RSG transition. However, further studies are necessary to reveal the role of the SG part in the RSG transition. We emphasize that the appearance of clusters of fluctuating spins is essential in the RSG transition and that the scattering of local spin concentrations yields those clusters.

The paper is organized as follows. We present the model and the method in Secs. II and III, respectively. In Sec. IV, we will show that the system is decomposed into the FM, SG, and IF parts. In Sec. V, the temperature dependence of the magnetization and the site magnetization for those parts will be examined. Section VI is devoted to conclusions.

II. MODEL

We begin with a dilute FM Heisenberg system with antiferromagnetic next-nearest-neighbor interactions on a simple cubic lattice with periodic boundary conditions.[14,15] The Hamiltonian is

$$H = -\sum_{\langle ij\rangle}^{nn} J_1 x_i x_j \boldsymbol{S}_i \cdot \boldsymbol{S}_j - \sum_{\langle kl\rangle}^{nnn} J_2 x_k x_l \boldsymbol{S}_k \cdot \boldsymbol{S}_l, \tag{1}$$

where $\boldsymbol{S}_i$ is the classical Heisenberg spin of $|\boldsymbol{S}_i|=1$; $J_1(>0)$, and $J_2(<0)$ respectively represent the FM nearest-neighbor and antiferromagnetic next-nearest-neighbor exchange interactions; $x_i=1$ or 0 when the lattice site i is occupied respectively by a magnetic or nonmagnetic atom. The average number of $x(\equiv[x_i])$ is the concentration of a magnetic atom, where $[\cdots]$ indicates an average over the lattice. Note that an experimental realization of this dilute FM model is $Eu_xSr_{1-x}S$,[16] in which magnetic atoms (Eu) are located on the fcc lattice sites. Here, for simplicity, we consider the model with $J_2=-0.2J_1$ on a simple cubic lattice.

We briefly summarize the magnetic ordering of this model: the model exhibits a FM phase transition for $x\gtrsim 0.79$ at a finite temperature; it exhibits a SG phase transition for $x\lesssim 0.95$ at a lower temperature; and the magnetization at $T\sim 0$ has a finite value only for $x\gtrsim 0.84$. Then the model exhibits the RSG phenomenon for $0.79\lesssim x\lesssim 0.83$. Hereafter, we mainly consider a system with $x=0.80$ for which the transition temperatures are given as $T_C/J_1\sim 0.27$, $T_R/J_1\sim 0.12$, and $T_{SG}/J_1\sim 0.10$,[14] and where T_R is the temperature at which the FM phase disappears.

III. METHOD

We use a conventional heat-bath Monte Carlo (MC) method. We have examined, as well as the magnetization $\boldsymbol{M}$ of the system, a site magnetization $\boldsymbol{m}_i$ for each spin i:

$$\boldsymbol{m}_i(\equiv\langle \boldsymbol{S}_i\rangle) = \frac{1}{M_s}\sum_{t=1}^{M_s} \boldsymbol{S}_i(t), \qquad (2)$$

where M_s is the time window (MC steps in the measurement of $\boldsymbol{m}_i$). For calculating $\boldsymbol{m}_i$, we should pay special attention to the thermal drift (uniform rotation) of the system, which always occurs in a finite system without anisotropy. In a conventional MC simulation, even in the FM phase, $m_i(\equiv|\boldsymbol{m}_i|)\to 0$ when M_s becomes large. Of course, we want to know the value of m_i for a large M_s in an infinite system where the thermal drift is absent. In the present study, the simulation is made as follows. For each sample and at each temperature, we first prepare an equilibrium spin configuration $\{\boldsymbol{S}_i(0)\}$ using the conventional MC method. Then starting with this spin configuration, we update the spin configuration $\{\boldsymbol{S}_i(t)\}$ $(t>0)$ step by step using the MC method. For every $M_R=20$ MC steps,[17] to bring the system back to $\{\boldsymbol{S}_i(0)\}$, a uniform rotation $R(t)$ is applied to the system so that

$$\Delta S = \sum_i |R(t)\boldsymbol{S}_i(t) - \boldsymbol{S}_i(0)|^2 \qquad (3)$$

becomes minimum.[18] Figures 1(a) and 1(b) respectively show typical examples of M_s dependence of the distribution $P(m_i)$ of the site magnetization in the conventional MC method and the present MC method without the thermal drift. Indeed, the results in Fig. 1(b) imply that $P(m_i)$ converges to a nontrivial distribution as M_s increases, in contrast to $P(m_i)$ in the conventional MC method [Fig. 1(a)].

The simulation has been made under the following conditions. The lattice size is $L\times L\times L$ ($L=24$, 32, and 40). The MC steps are $M_s=1\times 10^5$, 2×10^5, and 4×10^5 for $L=24$, 32, and 40,[19] respectively, and for each L, the same MC steps are spent for preparing $\{\boldsymbol{S}_i(0)\}$. Numbers of samples with different distributions of the magnetic atoms are $N_s=16$ for each lattice size. We measure the temperature in units of $J_1 (k_B=1)$.

IV. FM PART AND SG PART

We first consider the site magnetizations of individual spins. Figure 2 shows the distribution functions $P(m_i)$ for $x=0.80$ at $T/J_1=0.19$, together with those for $x=0.75$ and 0.85. The shapes of $P(m_i)$ for $x=0.75$ and 0.85 are approvable; at $T/J_1=0.19$, the system is in the paramagnetic (PM) phase for $x=0.75$ and in the FM phase for $x=0.85$.[15] In contrast, for $x=0.80$, $P(m_i)$ is strange: it has a large but rather broad peak at $m_i\sim 0.5$ and a hump at $m_i\sim 0$, which suggest that different kinds of spins coexist in the system. Some behave like those in the FM phase, whereas others fluctuate like those in the PM phase. Figures 3(a)–3(f) show $P(m_i)$ for $x=0.80$ at different temperatures. The hump is apparent for $T/J_1\gtrsim 0.16$. As the temperature is decreased, it diminishes and disappears at $T/J_1\sim 0.10$. Below this temperature, the peak becomes sharper and its position moves toward the large m_i side. These clearly reveal that two kinds of spins behave in different manner at intermediate temperatures of $T_C>T\gtrsim 0.13J_1$ and that both freeze at lower temperatures.

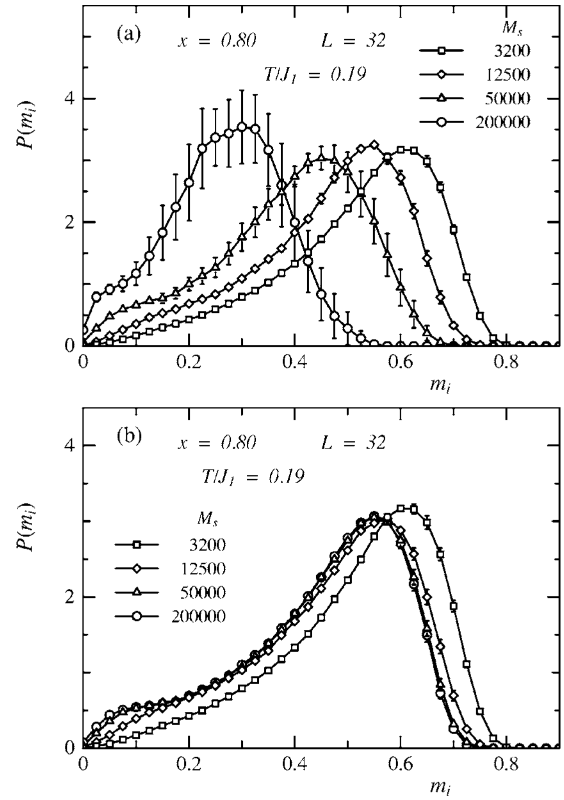

FIG. 1. Distributions of the site magnetizations $m_i(\equiv|\boldsymbol{m}_i|)$ calculated in various time windows M_s in the system with $x=0.80$: (a) a conventional MC method and (b) the present MC method which removes the thermal drift of the system. Note that drift effect becomes prominent for $M_s\gtrsim 10\,000$.

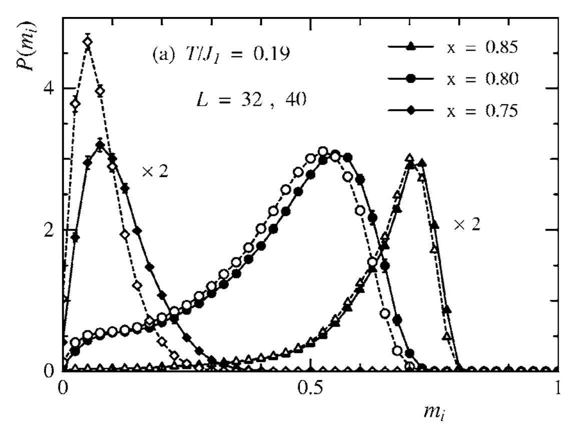

FIG. 2. Distributions of the site magnetizations $m_i(\equiv|\boldsymbol{m}_i|)$ for various spin concentrations x. Filled symbols are $P(m_i)$ for $L=32$; open symbols are those for $L=40$.

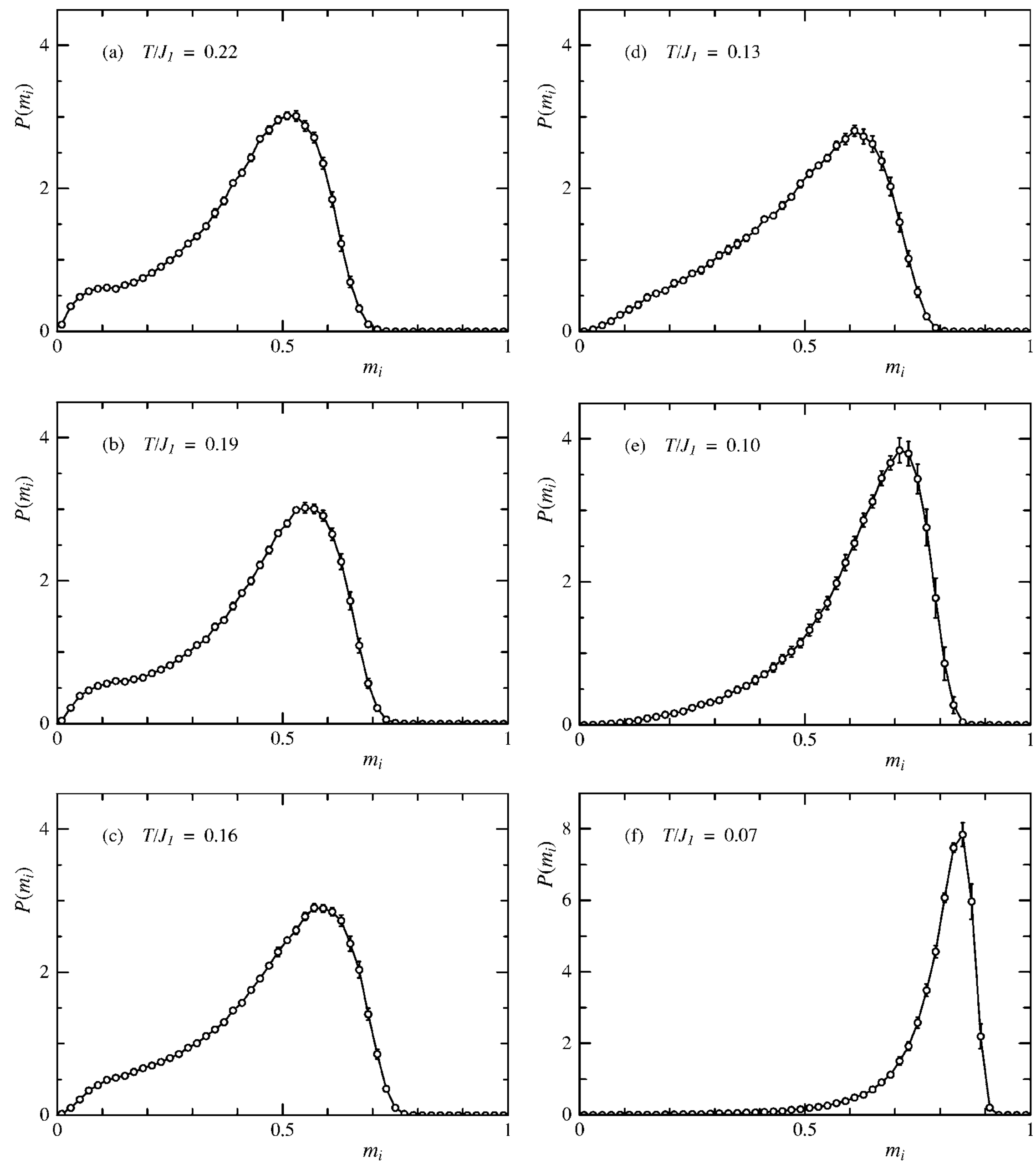

FIG. 3. Distributions of the site magnetizations $m_i(\equiv|\boldsymbol{m}_i|)$ for various temperatures for $x=0.80$ in the system with $L=32$.

Figures 4(a) and 4(b) respectively show the structures of $\boldsymbol{m}_i$ for $x=0.80$ at $T/J_1=0.19$ and 0.04. In fact, at $T/J_1=0.19$, the FM correlation of the spins extends over the lattice; clusters of fluctuating spins also exist (ones with smaller values of m_i). At a lower temperature of $T/J_1=0.04$, those fluctuating spins are frozen and the FM order breaks up to FM clusters.

To proceed with the consideration of the spin ordering process, let us classify the spins. For each sample, we classify them according to the magnitudes of m_i at $T/J_1=0.19$ (Ref. 20): a FM spin for $m_i>\tilde{r}_{\mathrm{FM}}$, a fluctuating (FL) spin for $m_i<\tilde{r}_{\mathrm{SG}}$, and a border (BD) spin for $\tilde{r}_{\mathrm{SG}}<m_i<\tilde{r}_{\mathrm{FM}}$, where $\tilde{r}_{\mathrm{FM}}\equiv r_{\mathrm{FM}}[m_i]$ and $\tilde{r}_{\mathrm{SG}}\equiv r_{\mathrm{SG}}[m_i]$. For this illustration, we choose $r_{\mathrm{FM}}=1$ and $r_{\mathrm{SG}}=0.6$.[21] We designate the part composed of FM spins as a FM part, the part composed of FL spins as a SG part, and the part composed of BD spins as an interface (IF) part. Note that the name of *SG part* comes from the RF model,[5] in which it is suggested that the FL spins freeze randomly at a low temperature, which triggers the RSG transition. We denote the numbers of the spins in the FM, SG, and IF parts, respectively, as N_{FM}, N_{SG}, and N_{IF}, and their ratios to the total lattice site N as x_{FM}, x_{SG}, and x_{IF}. Majority spins belong to the FM part and a fraction of the spins to the SG part, e.g., for a lattice with $L=32$, $x_{\mathrm{FM}}=0.468(2)$, $x_{\mathrm{IF}}=0.214(2)$, and $x_{\mathrm{SG}}=0.118(2)$. However, x_{SG} increases slightly with lattice size L,[22] implying that, in the thermodynamic limit, a finite ratio of the spins actually belongs to the SG part.

The appearance of SG parts will arise from scattering of the spin concentration in domains. To explore this conjec-

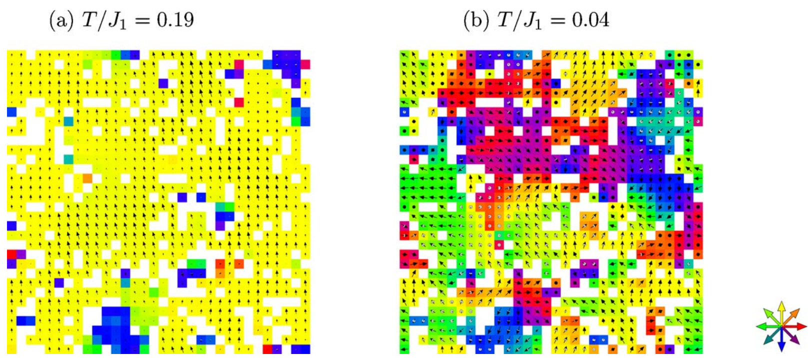

FIG. 4. (Color online) Structures of $\boldsymbol{m}_i$ in the system for $x=0.80$ at (a) $T/J_1=0.19$ and (b) $T/J_1=0.04$ on the same plane of the $32\times 32\times 32$ lattice. Sizes of the arrows indicate the magnitudes of $\boldsymbol{m}_i$ and colors of the cells indicate their directions. Positions of the nonmagnetic atoms are represented in white.

ture, we calculate for each lattice site j the average value of m_i, S_j, and the average spin concentration of $\tilde{x}_j$: $S_j=[1/n_j(R_1)]\Sigma_{i\in\Omega_j(R_1)} m_i$ and $\tilde{x}_j=n_j(R_2)/N(R_2)$ with R_2 ($\geqslant R_1$), where $\Omega_j(R)$ is a sphere of radius R centered at j, and $N(R)$ and $n_j(R)$, respectively, indicate the number of lattice sites and the number of spins inside the sphere. Because $\tilde{x}_j$ has discrete values x_s, for each of those values x_s, we calculate the average value of S_j $[S(x_s)]$. Figure 5 shows $[S(x_s)]$ as function of x_s for $R_1=2$ and for various R_2 in a typical sample with $L=32$. In fact, considerable dependence of $[S(x_s)]$ on x_s is apparent. In particular, when $R_2>R_1$, this dependence becomes strong.

We suggest, hence, that for $x\sim 0.80$, the system is decomposed into the FM part and the SG part together with the IF part between them, and the SG part is composed of clusters of the FL spins which appear inside domains with a lower spin concentration.

V. PARTIAL MAGNETIZATIONS AND PARTIAL SITE MAGNETIZATIONS

We consider roles of respective parts on the spin ordering process of the system. We define for the FM, SG, and IF parts, respectively, the partial magnetizations $M_{\rm FM}$, $M_{\rm SG}$, and $M_{\rm IF}$, and the partial site magnetizations $m_{\rm FM}$, $m_{\rm SG}$, and $m_{\rm IF}$, as follows:

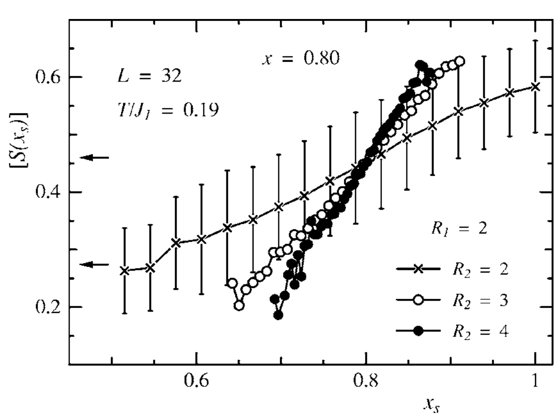

FIG. 5. The relationship between the $[S(x_s)]$ and x_s for $R_1=2$ and for various R_2 in a typical sample. Two arrows indicate from above $\tilde{r}_{\rm FM}(\equiv[m_i])$ and $\tilde{r}_{\rm SG}$. The deviations of $S(x_s)$ for these R_2 are of the same order and we present them only for $R_2=R_1$.

$$M_C=\left[\frac{1}{N_C}\left\langle\left|\sum_{i\in C}\boldsymbol{S}_i\right|\right\rangle\right]_s, \tag{4}$$

$$m_C=\left[\frac{1}{N_C}\sum_{i\in C} m_i\right]_s, \tag{5}$$

where C=FM, SG, and IF, and $[\cdots]_s$ denotes the sample mean. In the PM and FM phases, $m_C\sim M_C(\geqslant 0)$. On the other hand, in the SG phase, $m_C>0$ and $M_C\sim 0$. Then the partial SG order parameter may be defined as

$$O_C=m_C-M_C. \tag{6}$$

Figure 6 shows temperature dependences of M_C and m_C for the FM and SG parts for different sizes of the lattice. We first note that qualitative behaviors of those quantities depend little on the lattice size L.[23] Two remarkable points are apparent. One is that $M_{\rm FM}$ is much larger than $M_{\rm SG}$ and exhibits a temperature dependence that is similar to that of the magnetization of the system.[14,15] That is, the spins in the FM part mainly assume the magnetization of the system. The other is that $m_{\rm FM}$ and $m_{\rm SG}$ begin to increase at $T\sim 0.12J_1$ ($\sim T_{\rm R}$), implying that spin freezing occurs at lower temperatures. Figure 7 shows temperature dependences of the SG order parameters $O_{\rm SG}$ and $O_{\rm FM}$ for different sizes. In fact, they increase rapidly around $T\sim 0.10J_1$ ($\sim T_{\rm SG}$). In particu-

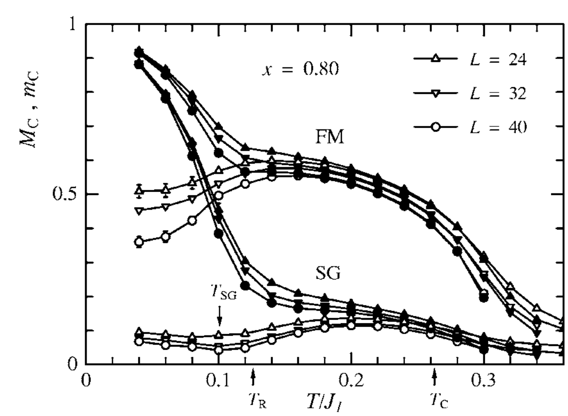

FIG. 6. The magnetizations $M_{\rm FM}$ and $M_{\rm SG}$ (open symbols) and the site magnetizations $m_{\rm FM}$ and $m_{\rm SG}$ (filled symbols) for the FM and SG parts, respectively, in the $L\times L\times L$ lattice. Note that we present those values only for FM and SG parts because those for the IF part have intermediate values.

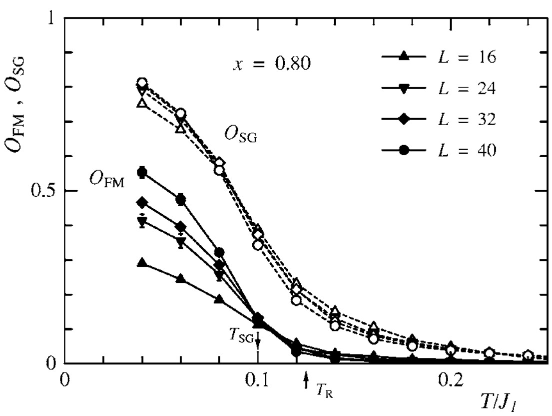

FIG. 7. Temperature dependences of the order parameters O_{SG} and O_{FM} in the $L \times L \times L$ lattice.

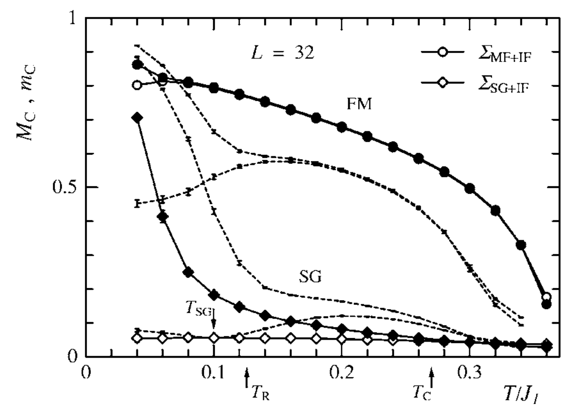

FIG. 8. The magnetizations (open symbols) and the site magnetizations (filled symbols) in systems Σ_{FM+IF} and Σ_{SG+IF} that are described in the text. Dotted curves are those in the original system $\Sigma(\equiv\Sigma_{FM+SG+IF})$ shown in Fig. 6.

lar, for $T < T_{SG}$, O_{FM} of a larger size becomes larger than that of a smaller size, in contrast to that for $T > T_{SG}$; the spin freezing will really occur for $T < T_{SG}$. This result is compatible with that of a previous study.[14] It is noteworthy that the increase of m_{FM} below T_R involves the decrease of the magnetization M_{FM}. That is, *the spins freeze and thereby break the FM order*.

To examine the roles of the FM and SG parts further, we performed additional simulations in two systems Σ_{FM+IF} and Σ_{SG+IF}: the former is constructed from the original system $\Sigma(\equiv\Sigma_{FM+SG+IF})$ by removing the SG part, and the latter by removing the FM part. Figure 8 shows results of those simulations. In Σ_{FM+IF}, as the temperature is decreased, M_{FM} increases rapidly at $T_C'/J_1 \sim 0.34$ $(>T_C/J_1)$ and continues to increase down to a very low temperature. Moreover, M_{FM} is much larger than that of Σ. These results clearly reveal that the SG part plays no role in the FM order, but disturbs it. That is, a competition occurs between the FM part and the SG part in the occurrence of the FM phase. In Σ_{SG+IF}, m_{SG} increases rapidly at around a lower temperature of T_{SG}'/J_1 ~ 0.05 $(<T_{SG}/J_1)$.[24] This result reveals that the SG part brings the spin freezing of the system. It should be noted, however, that the spins of the SG part will freeze in conjunction with all other spins of Σ, because T_{SG} of Σ is higher than T_{SG}'.[25] This speculation is, of course, compatible with the result given in Fig. 7: O_{SG} and O_{FM} increase together at low temperatures. These results seem to be incompatible with the sequential scenario of the RSG transition in the RF model, i.e., the spins of the SG part freeze first, then they yield random effective fields to the spins of the FM part and the FM order breaks down, realizing the SG phase. However, if a feedback from the FM part to the SG part exists, the spin freezings in both parts would occur simultaneously. Further studies are necessary to elucidate the mechanism of the RSG transition.

VI. CONCLUSIONS

We have examined the spin ordering of a RSG system. We have found that the system is decomposed into a FM part and a SG part, together with an interface (IF) part between them. The FM part mainly assumes the magnetization of the system and the SG part brings the spin freezing at low temperatures. Thus, we conclude that the RF model is a concept that can explain the RSG transition. However, further studies are necessary to reveal the role of the SG part in the RSG transition.

ACKNOWLEDGMENTS

The authors would like to thank S. Abiko, T. Shirakura, and M. Sasaki for their valuable discussions. They also would like to thank K. Sasaki for his useful suggestions and critical reading of the paper. This work was financed by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture.

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[16] H. Maletta and W. Felsch, Phys. Rev. B **20**, 1245 (1979).
[17] We have tested the effect of the rotation by choosing several intervals ranging from $M_R=5$ to $M_R=40$ and found that the difference in $P(m_i)$ is negligible between those intervals.
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[19] These MC steps M_s are determined so that the difference between the distribution function $P(m_i)$ for M_s MC steps and that for $M_s/4$ MC steps becomes less than 2%.
[20] In the same sample, we have made several runs by changing seeds of the random number and examining the temperature dependence of m_i for several spins. We found that m_i is almost independent of the runs; the spins with smaller values of m_i at $T/J_1=0.19$ have smaller values at all temperatures for $T_R<T<T_C$ and the same is true for the spins with larger values. That is, we may distinguish the spins by considering the magnitude of m_i at an appropriate temperature for $T_R<T<T_C$.
[21] We have examined the classification of the spins using different values of $r_{SG}(=0.5, 0.6,$ and $0.8)$ and fixing $r_{FM}=1$ to confirm that the choice of r_{SG} is irrelevant to our argument. For those values, the properties of quantities described by using those classified spins do not change markedly, but they become more prominent as r_{SG} is decreased.
[22] Fractions x_{FM}, x_{IF}, and x_{SG} are obtained as follows: for $L=24$, $x_{FM}=0.469(2)$, $x_{IF}=0.218(2)$, and $x_{SG}=0.113(2)$; for $L=32$, $x_{FM}=0.468(2)$, $x_{IF}=0.214(2)$, and $x_{SG}=0.118(2)$; and for $L=40$, $x_{FM}=0.467(1)$, $x_{IF}=0.211(1)$, and $x_{SG}=0.122(1)$.
[23] In the RSG system, magnetic quantities exhibit a considerable lattice size dependence in a finite system (Ref. 14).
[24] We have examined the SG transition through the application of the same technique as that used in a previous paper (Ref. 14) and found that the SG transition occurs at a lower temperature of $T/J_1\sim 0.02(\ll T_{SG}/J_1)$.
[25] We performed similar simulations by enlarging the IF part (a larger value of $\tilde{r}_{FM}$ but fixed $\tilde{r}_{SG}$) and found that T'_{SG} increases with $\tilde{r}_{FM}$ and reaches T_{SG}.