Reentrant Spin-Glass Transition in a Dilute Magnet

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We perform a large-scale Monte Carlo simulation of a dilute classical Heisenberg model with ferromagnetic nearest neighbor and antiferromagnetic next-nearest neighbor interactions. We found that the model reproduces a reentrant spin-glass transition. That is, as the temperature is decreased, the magnetization increases rapidly below a certain temperature, reaches a maximum value, and then disappears at some lower temperature. The low temperature phase was suggested to be a spin-glass phase that is characterized by ferromagnetic clusters.

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Reentrant spin-glass (RSG) transition is a well-known phenomenon of spin glasses (SGs). The RSG transition is found near the phase boundary between the SG phase and the ferromagnetic phase [1,2]. As the temperature decreases from a higher temperature, magnetization once increases and then disappears at a lower temperature. Finally, the SG phase is realized. The phenomenon was first considered as a phase transition between a ''ferromagnetic phase'' and a ''SG phase'' [3]. However, neutron diffraction studies have revealed that the ''SG phase'' is characterized by ferromagnetic clusters [4–6]. Now the RSG transition is believed to be a reentry from a ferromagnetic phase to a frozen state with ferromagnetic clusters.

The mechanism responsible for this reentrant transition has not yet been resolved. Two ideas have been proposed for describing the RSG transition: (i) an infinite-range Ising bond model, and (ii) a phenomenological random field concept. Sherrington and Kirkpatrick solved the infinite-range Ising bond model using a replica technique [7]. They predicted the occurrence of the RSG phase transition before the RSG transition was observed experimentally [1]. However, successive studies of the model revealed that the RSG phase transition does not occur [8], even when the vector spins substitute for the Ising spins [9]. On the other hand, the random field idea was proposed to explain experimental observations of neutron scattering functions [4–6,10]. The essential point of that conception is that the system is decomposed into a ferromagnetic part (FM part) and a part with frustrated spins (SG part). At low temperatures, the spins of the SG part yield random effective fields to the spins of the FM part. The FM order, which grows at higher temperatures, vanishes because of a random field effect [11]. Later, it was pointed out that not random field but random anisotropy brings a RSG-like phenomenon [12,13]. Nevertheless, no theoretical evidence has yet been presented for this idea in a microscopic point of view. In the last two decades, computer simulations have been performed extensively to solve the RSG transition in various models such as short-range bond models [14–16], short-range site models [17,18], and a Ruderman-Kittel-Kasuya-Yoshida model [19,20]. However, no realistic model has been shown to reproduce the vanishing magnetization that grows at higher temperatures.

This Letter reports a dilute classical Heisenberg model that reproduces a RSG transition. That is, the magnetization increases rapidly below a certain temperature, then disappears at some lower temperature. The low temperature phase was suggested to be a SG phase, which is characterized by ferromagnetic clusters. It should be emphasized that no novel mechanism is necessary to reproduce the RSG transition. The model studied here is a natural one that was proposed experimentally [21] and investigated using a Monte Carlo (MC) method [17]. A large-scale MC simulation revealed the nature of the model. We believe that the system size is crucial for eliminating the magnetization, because the SG phase at low temperatures is composed of ferromagnetic clusters.

We start with a dilute Heisenberg model with competing nearest and next-nearest neighbor exchange interactions described by the Hamiltonian:

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

where $\boldsymbol{S}_i$ is the classical Heisenberg spin of $|\boldsymbol{S}_i| = 1$; $J_1(>0)$ and $J_2(>0)$, respectively, represent the nearest neighbor and the 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 $x(\equiv \langle x_i \rangle)$ is the concentration of a magnetic atom. Note that an experimental realization of this model is $Eu_xSr_{1-x}S$ [21], in which magnetic atoms (Eu) are located on the fcc lattice sites [22]. Here, for simplicity, we consider the model on a simple cubic lattice with $J_2 = 0.2J_1$ [23].

A computer simulation was performed using a conventional heat bath MC method. The system was cooled gradually from a high temperature (cooling simulation). We calculated the magnetization M defined as $M = [\langle M(s) \rangle]$, where $M(s)(\equiv |\Sigma_i x_i \boldsymbol{S}_i|)$ is the magnetization at

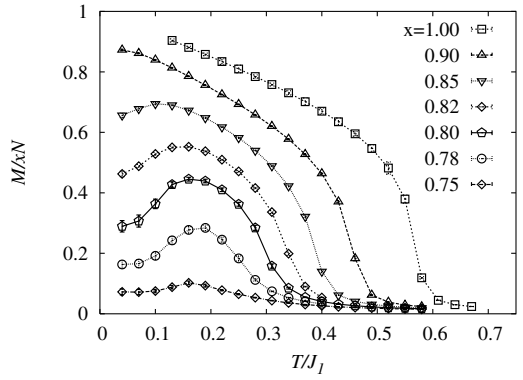

FIG. 1. Magnetizations M in the $32 \times 32 \times 32$ lattice for various spin concentrations x.

the sth MC step, and $\langle \cdots \rangle$ represents a MC average and $[\cdots]$ a sample average. Here, for larger lattices, 200 000 MC steps (MCS) were allowed for relaxation; data of successive 200 000 MCS were used to calculate average values. We show later that these MCS are sufficient for studying equilibrium properties of the model at a temperature range within which the RSG behavior is found. We made a simulation for $x > 0.70$. We treated lattices of $L \times L \times L$ ($L = 8$–48) with periodic boundary conditions. Numbers N_s of samples with different spin distributions are $N_s = 1000$ for $L \leq 12$, $N_s = 600$ for $L = 16$, $N_s = 200$ for $L = 24$ and 32, and $N_s = 80$ for $L = 48$. We measured the temperature in the unit of J_1 ($k_B = 1$).

Figure 1 shows temperature dependencies of the magnetization per spin M/xN for various x, where $N(\equiv L^3)$ is the number of lattice sites. For $x = 1$, as the temperature decreases, M increases rapidly below a temperature revealing the occurrence of a ferromagnetic phase. As x decreases, M exhibits an interesting behavior. In the range of $0.78 \lesssim x \lesssim 0.85$, M once increases, reaches a maximum value, and then decreases. Such behavior of M is reminiscent of the occurrence of the RSG transition.

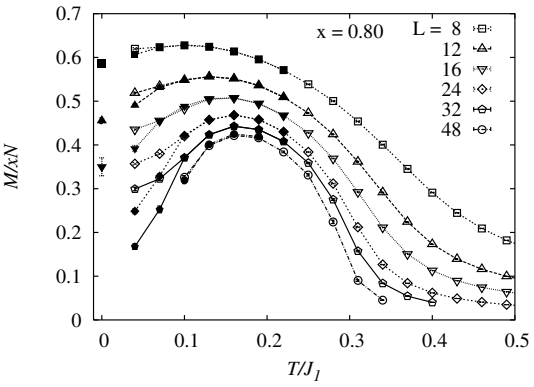

FIG. 2. Magnetizations M for $x = 0.80$ in the $L \times L \times L$ lattice. Open symbols indicate M in the cooling simulation and filled symbols that in the heating simulation. Data at $T = 0$ indicate those in the ground state.

To examine this phenomenon, we made detailed studies in the case of $x = 0.80$. First, we note that we performed a complementary simulation. That is, starting with a random spin configuration at a low temperature, the system is heated gradually (heating simulation). In addition, we investigated M in the ground state for smaller lattices ($L \leq 16$) having used a hybrid genetic algorithm [24]. Figure 2 shows temperature dependencies of M in both cooling and heating simulations for various L together with the value of M in the ground state. For $T \gtrsim 0.1J_1$, data of the two simulations almost coincide mutually, even for large L. We thereby infer that M for $T \gtrsim 0.1J_1$ are of thermal equilibrium. On the other hand, for $T < 0.1J_1$, a great difference in M is seen between the two simulations; estimation of the equilibrium value is difficult. We speculate that the heating simulation gives a value of M that is similar to that in the equilibrium state because the data in the heating simulation seem to connect to those in the ground state.

Figure 2 shows that the lattice size dependence of M is remarkable. For smaller L, as the temperature decreases, M decreases slightly at very low temperatures. The decrease is enhanced as L increases. Consequently, a strong size dependence of M is indicated for $T \lesssim 0.1J_1$. Particularly in the ground state, M apparently decreases rapidly as L increases. These facts imply that M disappears at low temperatures as well as at high temperatures.

We investigated the Binder parameter g_L,

$$g_L = \left(5 - 3\frac{[\langle M(s)^4 \rangle]}{[\langle M(s)^2 \rangle]^2}\right)\Big/2, \tag{2}$$

to examine whether or not the ferromagnetic phase actually occurs within an intermediate temperature range. Figure 3 shows g_L's for various L [25]. We can see that g_L's for different L cross at two temperatures T_C and T_R ($< T_C$). The cross at $T_C/J_1 = 0.265 \pm 0.010$ is a usual one that is found in the ferromagnetic phase transition [26]. That is, for $T > T_C$, g_L for a larger size is smaller than that for a smaller size; for $T < T_C$, this size dependence in g_L is reversed. On the other hand, the cross at T_R is strange. For

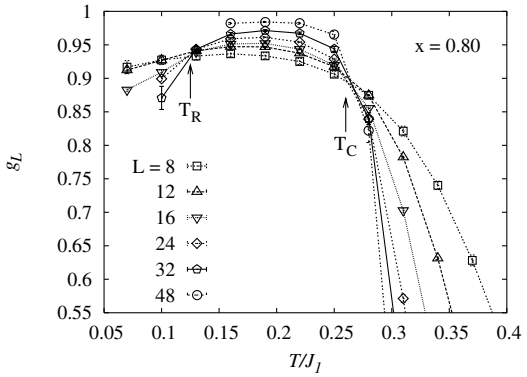

FIG. 3. Binder parameters g_L for $x = 0.80$.

$T < T_R$, g_L for a larger size again becomes smaller than that for a smaller size. Interestingly, the cross for different g_L occurs at almost the same temperature of $T_R/J_1 = 0.125 \pm 0.005$ [26]. These facts suggest that, as the temperature is decreased beyond T_R, the ferromagnetic phase, which occurs below T_C, disappears.

Does the spin correlation function below T_R decay rapidly as the distance between two spins increases? If it decays according to the power law, then $[\langle M(s)^2\rangle]/xN \propto L^{2-\eta}$ with η being the decay exponent of the spin correlation function. We plot, in Fig. 4, $[\langle M(s)^2\rangle]/xN$ as a function of L in a log-log form at various temperatures. In fact, at a temperature just below T_R ($T/J_1 = 0.10$), data seem to lie on a straight line; the spin correlation function will decay algebraically. Different L dependences are found above and below this temperature. At higher temperatures ($T > T_R$), as L is increased, data deviate upward from the straight line being compatible with the fact that the ferromagnetic long range order occurs at these temperatures. On the other hand, data deviate downward at lower temperatures. That is, the spin correlation function decays exponentially, similar to that in the SG phase.

Is the SG phase realized at low temperatures? A convincing way of examining the SG phase transition is a finite size scaling analysis of the correlation length, ξ_L, in samples of different sizes L [27,28]. Data for the dimensionless ratio ξ_L/L are expected to intersect at $T = T_{SG}$. Here we consider the correlation length of the SG component of the spin, i.e., $\tilde{\boldsymbol{S}}_i (\equiv \boldsymbol{S}_i - \boldsymbol{m})$ with $\boldsymbol{m}$ being the ferromagnetic component of $\boldsymbol{m} = \Sigma_i x_i \boldsymbol{S}_i/(xN)$. We performed a cooling simulation of a two-replica system with $\{\boldsymbol{S}_i\}$ and $\{\boldsymbol{T}_i\}$ [29]. The SG order parameter, generalized to wave vector $\boldsymbol{k}$, $q^{\mu\nu}(\boldsymbol{k})$, is defined as

$$q^{\mu\nu}(\boldsymbol{k}) = \frac{1}{xN}\sum_i \tilde{S}_i^{\mu}\tilde{T}_i^{\nu} e^{i\boldsymbol{k}\boldsymbol{R}_i}, \tag{3}$$

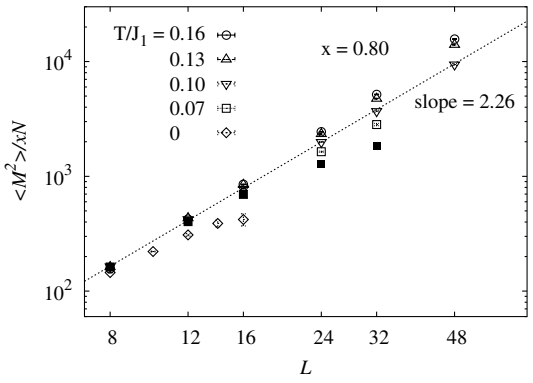

FIG. 4. The square of the magnetization $[\langle M(s)^2\rangle]/xN$ vs L. The straight line at $T = 0.1J_1$ is obtained in the least square for smaller lattices of $L \leq 16$. For $T = 0.07J_1$, open symbols indicate those in the cooling simulation and filled symbols represent those in the heating simulation. Data in both simulations deviate downward from the straight line.

where $\mu, \nu = x, y, z$. From this, we determine the wave vector dependent SG susceptibility $\chi_{SG}(\boldsymbol{k})$ by

$$\chi_{SG}(\boldsymbol{k}) = xN\sum_{\mu,\nu}[\langle |q^{\mu\nu}(\boldsymbol{k})|^2\rangle]. \tag{4}$$

The SG correlation length is then determined from

$$\xi_L = \frac{1}{2\sin(k_{min}/2)}\left(\frac{\chi_{SG}(0)}{\chi_{SG}(\boldsymbol{k}_{min})} - 1\right)^{1/2}, \tag{5}$$

where $\boldsymbol{k}_{min} = (2\pi/L, 0, 0)$.

Figure 5 shows the temperature dependence of ξ_L/L for various L. In fact, ξ_L/L for different L intersect at $T \sim 0.10J_1$. In particular, data for $L \geq 12$ are scalable on the assumption that $T_{SG}/J_1 = 0.105 \pm 0.003$. On the basis of this fact together with the rapid decay of the spin correlation function, we inferred that the SG phase is realized at low temperatures. The SG transition temperature T_{SG} estimated here is slightly lower than T_R [30]. However, the possibility of $T_R = T_{SG}$ cannot be ruled out, because the treated lattices of $L \leq 20$ for estimating T_{SG} are not large enough.

We considered the spin structure. Figures 6(a) and 6(b) show typical results for it. In the ferromagnetic phase [Fig. 6(a)], although the spin arrangement is considerably modulated, a ferromagnetic spin correlation extends over the lattice. On the other hand, in the SG phase [Fig. 6(b)], we can see that the system breaks up to yield ferromagnetic clusters with a linear size of $l_c \sim 7$ [31]. This result is compatible with the size dependence of M shown in Fig. 2. In the small lattice with $L = 8$, M does not exhibit a marked decrease at $T < T_{SG}$. The decrease becomes drastic when L is increased. This fact is further evidence that the system is divided into ferromagnetic clusters. We suggest that the SG phase of the model is characterized by ferromagnetic clusters. This concept is compatible with experimental observations [4–6].

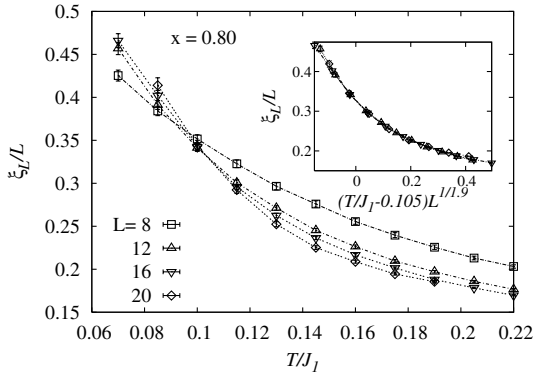

FIG. 5. The SG correlation length ξ_L divided by L. The data intersect at $T \sim 0.10J_1$, implying that there is a SG transition at this temperature. The inset shows a typical example of the scaling plot for $L \geq 12$.

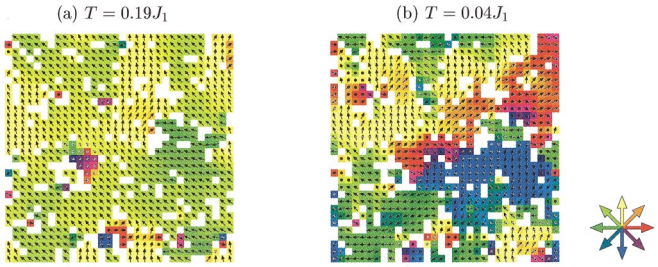

FIG. 6 (color). Spin structures of the model for $x = 0.80$ on a plane of the $32 \times 32 \times 32$ lattice in (a) the ferromagnetic phase and (b) the spin-glass phase. Spins represented here are those averaged over 10 000 MCS. The positions of the nonmagnetic atoms are represented in white.

In summary, we found a model that settles the most important issue of the RSG transition. Other important issues remain unresolved. Why does the ferromagnetic phase disappear at low temperatures? Does this dilute model exhibit the same behavior as that found in the bond SG model [28,32]? Does the chiral glass phase transition simultaneously occur at T_{SG} [33]? We intend the present model as one means to solve those and other remaining problems.

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