Inverse freezing in a cluster Ising spin-glass model with antiferromagnetic interactions

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Inverse freezing is analyzed in a cluster spin-glass (SG) model that considers infinite-range disordered interactions between magnetic moments of different clusters (intercluster interaction) and short-range antiferromagnetic coupling J_1 between Ising spins of the same cluster (intracluster interaction). The intercluster disorder J is treated within a mean-field theory by using a framework of one-step replica symmetry breaking. The effective model obtained by this treatment is computed by means of an exact diagonalization method. With the results we build phase diagrams of temperature T/J versus J_1/J for several sizes of clusters n_s (number of spins in the cluster). The phase diagrams show a second-order transition from the paramagnetic phase to the SG order at the freezing temperature T_f when J_1/J is small. The increase in J_1/J can then destroy the SG phase. It decreases T_f/J and introduces a first-order transition. In addition, inverse freezing can arise at a certain range of J_1/J and large enough n_s . Therefore, the nontrivial frustration generated by disorder and short-range antiferromagnetic coupling can introduce inverse freezing spontaneously.

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I. INTRODUCTION

Inverse transitions (ITs) are reversible transformations from a more ordered phase (crystalline) to a less ordered one (liquid or disordered phase) as the temperature diminishes. This counterintuitive class of phase transition has recently been the subject of several theoretical and experimental studies. The main reasons for the increasing interest are recent experimental findings showing ITs in a great variety of physical systems such as gold nanoparticles [1], magnetic thin films [2], high- T_c superconductors [3], ferromagnetism in semiconductors [4], organic monolayers on a metal surface [5], polymers [6], and others (see Ref. [7] and references therein). From a theoretical point of view, the identification of mechanisms underlying ITs is a quite important issue and it is one of the points investigated in the present work.

To improve the theoretical understanding of ITs, it is useful to study models able to produce this phenomenon at least qualitatively. In this sense, some spin-1 ($s^z = -1, 0, 1$) particle models have been adopted, with interesting results. For instance, the Blume-Capel model [8] can describe an IT (inverse melting) from the ferromagnetic order to the paramagnetic (PM) phase as the temperature decreases since an entropic advantage of the interacting states ($s^z = 1, -1$) is assumed [7]. In other words, the IT arises if the $s^z = 0$ states controlled by the crystal lattice field D are energetically favored at the same time that an entropic advantage of interacting states is adopted. Another important contribution has been obtained from mean-field studies of the strongly disordered Ghatak-Sherrington model, in which the IT known as inverse freezing (IF) appears spontaneously. This means that a re-entrant transition from the spin-glass (SG) phase to the PM one is found in phase diagrams of the temperature versus D [7,9,10]. Particularly, the presence of frustration introduced by disorder has been suggested as a mechanism important to the existence of IF [7,9,10], but only disorder without frustration has not been indicated as essential for the spontaneous occurrence of ITs [11].

In addition, Monte Carlo investigations of the disordered Blume-Capel model in three dimensions have also found spontaneous IF for a certain range of D [12]. In this case, the authors indicate that the low-temperature PM phase presents a high number of $s^z = 0$ sites, in contrast to the higher temperature PM phase determined by completely disordered interacting spin states. These results suggest that IF can occur as a consequence of the simultaneous presence of frustration and noninteracting spin states. The same conclusions are also obtained by recent studies in fermionic SG models [13–15]. In the fermionic case the magnetic dilution caused by the presence of empty and double-occupied sites (nonmagnetic sites) play the role of $s^z = 0$ states.

Recently, numerical studies of a two-dimensional randombond Ising model have shown that weak disorder and frustration can spontaneously introduce an IT from the ferromagnetic to the PM phase [16]. However, in contrast to previous analyses, this result is obtained by considering Ising spins. Therefore, one can raise the following issues: Is a strongly disordered SG model with Ising spins able to exhibit IF? and Is the presence of $s^z = 0$ spin states really necessary for the existence of IF?

To answer these questions, the present work studies a strongly disordered Ising SG model within a cluster formulation, in which infinite-range intercluster disordered interactions are considered with short-range AF interactions

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 J_1 . The short-range interactions are only between Ising spins belonging to the same cluster (intracluster interaction). Therefore, the intercluster disorder can introduce frustration, while the intracluster AF interaction can give rise to a decrease in the effective magnetic moment of clusters without long-range AF order. In other words, the disordered interactions between the magnetic moments of clusters can be affected by the intracluster AF interaction. In particular, the increase in J_1 can favor a scenario in which the total magnetic moment of clusters become very small. As a consequence, it can result in a PM phase characterized by a large number of clusters with small intercluster interaction.

This cluster SG model has initially been proposed by Soukoulis and Levin [17] to improve the thermodynamic description of strongly disordered SG models by introducing short-range interactions within a cluster mean-field treatment of quenched disorder. Their analyses were done within the replica symmetric approximation with both ferromagnetic [17,18] and antiferromagnetic short-range interactions, in which the behavior of the specific heat c_v and magnetic susceptibility χ were discussed. However, for the AF case, the authors presented results for very strong AF coupling between classical Heisenberg spins in rather specific cluster configurations, in which a small number of spins in each cluster n_s is considered ($n_s \leq 6$) [17]. In contrast, in this work, the cluster SG model is studied in the range of weak and medium AF coupling with Ising spin, which has not yet been explored. In addition, the mean-field treatment of the disorder is improved by adopting one-step replica symmetry breaking (1S-RSB). We also use a simple square lattice geometry for the spins in each cluster, which can be greater than 6 ($n_s \leq 20$). Furthermore, the results obtained are interpreted in the context of the IF problem.

II. GENERAL FORMULATION

Therefore, the present study considers an infinite-range cluster Ising SG model described by

$$H = -\sum_{\nu\lambda}^{N_{\rm cl}} J_{\nu\lambda} S_{\nu}^z S_{\lambda}^z - \sum_{\nu}^{N_{\rm cl}} \left(\sum_{ij}^{n_s} J_{ij}^0 S_{i\nu}^z S_{j\nu}^z \right), \qquad (1)$$

where N_{cl} and n_s represent the number of clusters and the number of spins in each cluster, respectively. In Eq. (1), S_{ν}^{z} corresponds to the magnetic moment of cluster ν ($S_{\nu}^{z} = \sum_{i=1}^{n_s} S_{i\nu}^{z}$), while $S_{i\nu}^{z}$ is the Ising spin of site *i* of cluster ν . The intercluster coupling $J_{\nu\lambda}$ is a random variable given by a Gaussian distribution with variance $16J^2/N_{cl}$ and mean 0. J_{ij}^0 is the intracluster exchange interaction between Ising spins belonging to the same cluster. We consider here a square lattice geometry and a nearest-neighbor antiferromagnetic coupling J_1 within the clusters.

The averaged free energy is obtained within the replica method formalism: $\beta f = \lim_{n\to 0} (\langle Z^n \rangle_{J_{\nu\lambda}} - 1)/n$, where $\langle \dots \rangle_{J_{\nu\lambda}}$ means the average over the quenched disorder of $J_{\nu\lambda}$, and Z^n is the replicated partition function. Performing the average over Z^n and using Hubbard-Stratonovich transformations, which introduce the replica matrix elements $\{Q\}$, the free energy per cluster is obtained as

$$\frac{\beta f}{N_{\rm cl}} = \lim_{n \to 0} \frac{1}{n} \Biggl\{ \frac{\beta^2 J^2}{2} \sum_{\alpha \gamma} Q_{\alpha \gamma}^2 - \ln \operatorname{Tr} \exp \Biggl[\beta \sum_{\nu} \\ \times \Biggl(\sum_{\alpha} \sum_{ij} J_{ij} S_{i\nu}^{z\alpha} S_{j\nu}^{z\alpha} + \sum_{\alpha \gamma} J Q_{\alpha \gamma} S_{\nu}^{z\alpha} S_{\nu}^{z\gamma} \Biggr) \Biggr] \Biggr\}, \quad (2)$$

where α and γ are replica indices. In the thermodynamic limit $(N_{\rm cl} \rightarrow \infty)$, the functional integrals over $Q_{\alpha\gamma}$ have been evaluated by the steepest descent method, which gives

$$Q_{\alpha\gamma} = \langle S^{\alpha}_{\nu} S^{\gamma}_{\nu} \rangle$$
 and $Q_{\alpha\alpha} = \langle S^{\alpha}_{\nu} S^{\alpha}_{\nu} \rangle$, (3)

where $\langle ... \rangle$ means the average over the effective model represented by Eq. (2). The parameter $Q_{\alpha\gamma}$ is related to the cluster SG order parameter and the diagonal replica $Q_{\alpha\alpha}$ is associated with the expectation value of the cluster magnetic moment magnitude [17].

In the present work, the problem is analyzed within Parisi's scheme of 1S-RSB [19], in which the replica matrix is parametrized as $R = Q_{\alpha\alpha}$ and

$$Q_{\alpha,\gamma} = \begin{cases} Q_1 & \text{if } I(\alpha/a) = I(\gamma/a), \\ Q_0 & \text{if } I(\alpha/a) \neq I(\gamma/a), \end{cases}$$
(4)

where I(x) gives the smallest integer which is $\ge x$. The parameter *a* represents the size of diagonal blocks of the 1S-RSB solution. In this approximation, the cluster SG phase occurs when $Q_0 \ne Q1$. Therefore, the 1S-RSB free energy is obtained as

$$\frac{\beta f}{N_{\rm cl}} = \frac{\beta^2 J^2}{4} \left[R^2 + a \left(Q_1^2 - Q_0^2 \right) - Q_1^2 \right] \\ - \frac{1}{a} \int Dz \ln \int Dv [K(z,v)]^a, \tag{5}$$

where $K(z,v) = \int D\xi \operatorname{Tr} e^{-\beta H_{\text{ef}}}$ and

$$H_{\rm ef} = -\sum_{ij} J_{ij}^0 S_{i\nu}^z S_{j\nu}^z - h S_{\nu}^z \tag{6}$$

represents the effective one-cluster model with

$$h = J\sqrt{(Q_1 - Q_0)}v + J\sqrt{(R - Q_1)}\xi + J\sqrt{Q_0}z, \quad (7)$$

and $\int Dx = \int_{-\infty}^{\infty} dx \frac{e^{x^2/2}}{\sqrt{2\pi}}$ ($x = z, v, \text{ or } \xi$). The parameters Q_1, Q_0, R , and *a* are obtained by minimizing the free energy given by Eq. (5) (see the Appendix). The magnetic susceptibility χ and the entropy *s* are also derived from Eq. (5): $\chi = \beta(R - Q_1 + a(Q_1 - Q_0))$ and $s = -\frac{\partial f}{\partial T}$.

III. RESULTS AND DISCUSSION

Numerical results are obtained by solving the effective one-cluster problem [Eqs. (5)–(7)]. We consider clusters of n_s Ising spins on a square lattice. The intracluster interaction J_{ij}^0 is antiferromagnetic ($J_{ij}^0 = -J_1$) and only between nearest neighbors. The intercluster disordered interaction is adjusted by J, where the temperature T and J_1 are given in units of J. Particularly, the SG order is characterized by the RSB solution ($Q_1 - Q_0 > 0$). This means that T_f is located when $Q_1 - Q_0$ becomes different from 0. The PM phase appears in the region where $Q_0 = Q_1$. For instance, Fig. 1 exhibits



FIG. 1. Order parameters and susceptibility as functions of (a) temperature (for low J_1/J) and (b) short-range antiferromagnetic interaction (for low T/J). Results are for $n_s = 8$. Inset in (a): Order parameters for large J_1/J .

the normalized order parameter behavior $(q_0 = Q_0/n_s^2, q_1 = Q_1/n_s^2, r = R/n_s^2)$ for a typical case with $n_s = 8$. In Fig. 1(a),

the SG phase is found below T_f within the RSB region at a low value of J_1/J , but only the PM phase occurs at higher values of J_1/J [see inset in Fig. 1(a)]. The effects of increasing J_1 can be better analyzed in Fig. 1(b), in which the order parameters present a discontinuous jump at the SG/PM phase transition (SG/PM first-order transition) at lower temperatures. Here it is worthwhile discussing the 1S-RSB approach. It locates the PM/SG second-order transition correctly. The 1S-RSB ansatz is also suitable for locating the PM/SG first-order boundary. In particular, there is a very small difference between results obtained with replica symmetry and those obtained with the 1S-RSB concerning the location of the first-order boundary. This result is already known from other works that use the replica method [7,9,11]. Therefore the 1S-RSB can give reliable results for our purpose, which is to build phase diagrams.

The parameter *r* is also very important in the present study. It can be interpreted as the average of total magnetic moment of clusters. As one can see in Fig 1, *r* depends on T/J and J_1/J . *r* decreases when J_1/J increases and it is very small within the PM phase. This behavior is discussed in more detail in Fig. 4. The magnetic susceptibility χ is also illustrated in Fig. 1. It presents a cusp at T_f and is weakly dependent on J_1/J and T/J in the whole RSB phase.

The discussion above can also be applied to phase diagrams of T/J versus J_1/J . For this purpose, Fig. 2 shows phase diagrams for a set of cluster sizes n_s , in which n_s is even. They exhibit a general characteristic: the PM phase at high temperatures suffers a continuous transition to the SG order at T_f for low intensities of short-range antiferromagnetic coupling. As J_1/J increases, T_f/J gradually decreases until a tricritical point, where the transition becomes first order. Near the tricritical point, the behavior of the phase transition changes with increasing value of n_s : An SG-PM re-entrant transition can appear for high enough values of n_s (see, for example, results with $n_s \ge 8$). The SG/PM re-entrance is observed as the temperature decreases for a certain range of J_1 . This



FIG. 2. Phase diagrams of T/J versus J_1/J for several cluster sizes n_s with even values of n_s . Solid and dashed lines represent secondand first-order transitions, respectively. (a) Shapes of the clusters, where $n_s = 4$, 8, 12, 16, and 20 are presented from top left to bottom right. Dotted lines correspond to the temperatures where χ presents maximum values. (b–d) Details of the first-order transition, where dot-dashed lines are the PM and SG spinodals. Inset in (a): Entropy as a function of temperature for $J_1/J = 0.803$.



FIG. 3. Phase diagrams of T/J versus J_1/J for odd n_s . The same line convention as in Fig. 2 is used. (a) Shapes of the clusters with 5, 7, 9, and 15 sites; n_s increases from top left to bottom right.

re-entrance is related to the IF as shown by the entropy results in the lower inset in Fig. 2(a), which shows that the entropy of the PM phase at low temperatures is lower than the SG one. Particularly, the re-entrance becomes more pronounced as n_s increases. Furthermore, the difference between the transition lines of two successive n_s becomes smaller for larger n_s . Therefore, one can expect consistency of these results when increasing the cluster size. In addition, the same qualitative results were also obtained for other shapes of clusters such as those presented in Refs. [17,18]. In other words, the presence of short-range antiferromagnetic interaction in a cluster SG problem can diminish T_f , introducing a first-order re-entrant SG/PM transition.

The behavior of transition lines described above is also observed for an odd number of spins in each cluster (see Fig. 3) at small and intermediate values of J_1/J . However, there is an important difference in the phase diagrams with odd versus even n_s at strong antiferromagnetic intracluster interactions. For the case with odd n_s , the ground state is



FIG. 4. Magnetic susceptibility χ and order parameter *r* as a function of T/J for higher values of J_1/J .

SG for larger J_1/J . Nevertheless, the T_f at higher J_1/J goes towards lower temperatures as n_s increases. This suggests that the transition lines for even and odd n_s converge at the same location in phase diagrams for clusters of large enough size.

Figure 4 exhibits the cluster magnetic moment r and magnetic susceptibility χ , which allow one to discuss the low-temperature cluster PM phase at high values of J_1/J . For instance, the intensity of r decreases as J_1/J increases [see Fig. 4(a)]. At the same time the long-range intercluster coupling is weakened when r decreases, which can prevent the occurrence of the SG phase (see the Appendix) at high J_1/J values. In addition, the increase in J_1/J is able to introduce a low-temperature cluster PM phase with a very low magnetic moment, in which a high number of nonmagnetic clusters with total moment $S^{z} = 0$ is found. This means that the intracluster spins can freeze into perfect AF zero-moment states. As a consequence, no interaction between net cluster moments (no intercluster interactions) will give any long-range ordered states. Therefore, it is the source of the entropy decrease in the cluster PM phase compared to the high temperature one. In fact the model is not spin-1, but for strong J_1 , the existence of nonmagnetic clusters leads to effective nonmagnetic clusters, and the model is similar to a spin-1 model, but at the cluster level. The increase in cluster size also favors the AF short-range coupling, reducing the order parameter r as shown in Fig. 4(b). This behavior can also strongly affect T_f/J (see the Appendix). However, for n_s odd, the total moment is never compensated inside the clusters, even at high values of J_1/J ; in this case, the SG phase can occur at very low temperatures even with high values of J_1/J .

The magnetic susceptibility also reflects the different behaviors of the cluster PM phase at low and high temperatures. For example, χ presents a maximum that is displaced to higher temperatures as J_1/J increases [see Fig. 4(c)]. The same effect is observed with increasing cluster size as shown in Fig. 4(d). However, for odd values of n_s the ground state can be SG, which explains the increase in χ at very low temperatures. The location of the χ maximum is also displayed in the phase diagrams in Figs. 2(a) and 3(a) by dotted lines. In this particular case, one can see that the effects of short-range AF interactions on the cluster PM phase are enhanced with increasing J_1/J or n_s . In other words, the AF character within the clusters can be favored by J_1/J and mainly by n_s . Particularly, for n_s in the thermodynamic limit, one can hope that the system presents the pure AF order without disorder, as expected.

IV. CONCLUSIONS

To summarize, in the present work, we have studied the IF transition by adopting an Ising cluster SG model with antiferromagnetic short-range interactions J_1 and infiniterange disordered interactions J. Within the 1S-RSB, the results indicate IF in a range of J_1/J for a large enough cluster size. Particularly, the low-temperature PM phase is characterized by clusters with a low magnitude of magnetic moment, which are obtained when short-range AF interactions between Ising spins within the clusters are large. Therefore, frustration introduced by disorder and a PM phase with a low cluster magnetic moment are key elements to produce IF spontaneously in the present study. These elements can be produced by strongly disordered clusters of Ising spins with short-range antiferromagnetic interactions. There is another important issue to be studied with this disordered cluster formalism. It is related to the presence of intracluster geometric frustration introduced by considering next-neighbor AF interactions such as the so-called J1-J2 model (see, e.g., Ref. [20] and references therein). There is clear evidence that trivial randomness alone (unable to generate frustration) [21] is not enough to create the necessary conditions to produce IF [11]. This specific point is currently under investigation.

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APPENDIX: ORDER PARAMETERS

The 1S-RSB order parameters are obtained from the free energy, Eq. (5), using the saddle point condition:

$$Q_0 = \int Dz \left[\frac{\int Dv[K(z,v)]^{a-1} \int D\xi \langle S_v^z \rangle_{H_{\text{eff}}}}{\int Dv[K(z,v)]^a} \right]^2, \quad (A1)$$
$$Q_1 = \int Dz \frac{\int Dv[K(z,v)]^{a-2} \left(\int D\xi \langle S_v^z \rangle_{H_{\text{eff}}} \right)^2}{\int Dv[K(z,v)]^a}, \quad (A2)$$

 $\int Dv[K(z,v)]^a$

with

$$R = \int Dz \frac{\int Dv [K(z,v)]^{a-1} \int D\xi \langle S_{\nu}^{z} S_{\nu}^{z} \rangle_{H_{ef}}}{\int Dv [K(z,v)]^{a}}, \quad (A3)$$
$$a^{2} \frac{\beta^{2} J^{2}}{4} (Q_{1}^{2} - Q_{0}^{2}) = -\int Dz \ln \int Dv [K(z,v)]^{a} + a \int Dz \frac{\int Dv [K(z,v)]^{a} \ln K(z,v)}{\int Dv [K(z,v)]^{a}}$$
(A4)

and $\langle \ldots \rangle_{H_{\text{ef}}} = \text{Tr} \ldots \exp(-\beta H_{\text{ef}}).$

In order to locate the second-order PM/SG phase transition, we can expand Eqs. (A1), (A2), and (A3) in powers of Q_0 and Q_1 . This procedure presents lengthy calculations that allow us to express the RSB order parameter $\delta = Q_1 - Q_0$ and R (for T close to T_f) as

$$\delta \approx \frac{\beta^2 J^2}{K_0^2} \bigg(\int d\xi \big\langle S_{\nu}^z S_{\nu}^z \big\rangle_{H^0_{\text{ef}}} \bigg)^2 \delta + O(\delta^2), \qquad (A5)$$

$$R(J_1/J, n_s) \approx rac{\int D\xi \langle S_{\nu}^z S_{\nu}^z \rangle_{H^0_{ ext{ef}}}}{K_0} + O(\delta^2),$$
 (A6)

where $K_0 = \int D\xi \exp(-\beta H_{ef}^0)$ and H_{ef}^0 is obtained from Eq. (6) with $Q_0 = Q_1 = 0$. Therefore, T_f can be located using Eqs. (A5) and (A6): $T_f/J = R(J_1/J, n_s)$. This indicates that the location of T_f/J also depends on the intensity of the total magnetic moment of clusters R.

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