Magnetic field effect on the fractal cluster spin-glass phase of an Ising antiferromagnet near the first-neighbor percolation threshold: $Fe_{0.25}Zn_{0.75}F_2$

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The effect of a magnetic field on the fractal cluster spin glass phase of the three-dimensional dilute antiferromagnet Ising compound $Fe_xZn_{1-x}F_2$, $x = 0.25$, near the first-neighbor percolation threshold, $x_p \approx 0.24$, is investigated via Monte Carlo and local-mean-field simulations. Here we consider the actual short-range spin couplings taken from the referred compound, rather than from stochastic distributions. We study the dynamics of the correlation function, thermoremanent (TRM) and isothermoremanent (IRM) magnetizations, and the thermodynamics of hysteresis cycles and zero-field-cooled (ZFC) and field-cooled (FC) magnetizations as well as the associated microscopic configuration of spins. Results are obtained for the first-neighbor percolating cluster and the whole sample. At very low temperature, the behavior of the system is dominated by the competition between the antiferromagnetic couplings and the field. As the temperature rises, our findings are consistent with a vitreous or metastable spin-glass-like thermodynamic state in the context of a modified droplets picture of thermal activation of fractal domains over logarithmic energy barriers.

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I. INTRODUCTION

The effect of a magnetic field on the spin glass (SG) phase is still an unsettled question despite over three decades of debate.[1] On one hand, replica symmetry breaking (RSB) ideas in the mean-field context of the infinite-range Sherrington-Kirkpatrick model[1–3] predict an equilibrium SG phase in fields up to the Almeida-Thouless (AT) line.[4] This fact is suggested[1] to be also valid for short-range SGs. On the other hand, scaling concepts based on thermal activation over free-energy barriers in short-range models—the "droplets" picture[5]—lead to the breakdown of the SG state in a field. Beyond mean field, early renormalization-group calculations[1,6–8] have not been able to find any stable fixed point in a field below the upper critical dimension $d_c = 6$, unless in a theory[1,7] qualitatively distinct from Parisi's ultrametric solution. Nevertheless, a more recent approach[9] to a generic replica symmetric field theoretical model have successfully probed the existence of the AT line below d_c.

In the droplets scenario, a paramagnetic (PM) state in a magnetic field H emerges from the zero-field SG phase below the freezing temperature, T_f, and beyond some crossover length scale, which diverges as $H \to 0$. As a consequence, experimental and numerical studies on SGs in H are greatly hindered, since their relaxation times are very large and equilibrium properties, if any, are difficult to characterize. On the experimental side, measurements of magnetization and susceptibility under distinct thermodynamical cycles,[10] AT line and critical properties,[11,12] including also dynamic scaling,[13,14] have characterized the SG behavior of the Ising compounds $Fe_{0.25}Zn_{0.75}F_2$[10,11,13,14] and $Fe_xMn_{1-x}TiO_3$,[12] $0.41 \leqslant x \leqslant 0.57$. The crossover from the random-field phase to a SG-like phase in a field was also investigated in detail.[15] In addition, dynamical studies on the susceptibility[16–18] and AT line[16,18] on the above-mentioned materials concluded that their relaxation times in H are finite. This finding suggests the presence of an H-induced vitreous or metastable SG-like thermodynamic state through a dynamical crossover consistent with the droplets picture.[8,17,18]

The above scenario finds support in Monte Carlo (MC) aging results[19] on the autocorrelation function and field-cooled magnetization of the three-dimensional (3D) short-range Edwards-Anderson (EA) Ising model. Although a number of MC studies in H have supported[20] the RSB scenario, other numerical results on the 3D EA model provide[19,21,22] evidence for the H-induced breakdown of the SG phase. We also mention that MC simulations on 1D Ising models, with spin-spin interactions falling off with a power of the distance, have been inconclusive in favoring[23] or not[24,25] the existence of a stable AT line. The relevance of these results relies in the fact that, through mapping arguments, they are connected with *short-range* Ising models in $d < d_c$. Further, by numerically probing the field effect on the SG phase through local low-energy excitations,[26–28] the ground-state critical fields $H_c \approx 0.4$[28] and 0.65[27] (in units of the variance of the Gaussian distribution of exchange couplings) have been estimated to be considerably smaller than the mean-field AT value ($H_{\rm AT}^{\rm MF} \approx 1.86$[29]). This result supports the droplets scenario, though the spin clusters look fractal, rather than compact. In spite of this, arguments favoring the RSB picture cannot be disregarded as well.[27]

In this work we perform MC and local-mean-field (LMF) simulations to investigate how the presence of a uniform field H affects the zero-field SG phase[10,11,13,14] of the bcc site-disordered Ising antiferromagnet (AF) $Fe_xZn_{1-x}F_2$ at $x = 0.25$. We remark that, both at $x = 0.25$ and 0.27, low-T neutron scattering experiments in $H = 0$ found[30] no Bragg peak associated with AF ordering, while Mössbauer measurements indicate[14] a competition between AF and SG order below 21 K and freezing temperature around 10 K. Recent experimental studies also show[31] that for $x \gtrsim 0.24$ the AF sharp peak in the specific heat data disappears and the rounded bump becomes

the only observed feature. Since the first-neighbor exchange coupling is the dominant interaction to set the AF order, then it is gratifying that the above experimental results are in agreement with both the best estimate for the first-neighbor site-percolation threshold of a bcc lattice, $x_p \approx 0.246$,[32] and low-T numerical findings[33] for the concentration thresholds associated with both AF and SG orderings. Moreover, although the inclusion of next-nearest-neighbor contributions reduces the value of x_p to ≈ 0.175,[34] the referred experimental and numerical results strongly suggest that this effect is not relevant to the magnetic properties of the present Ising system, since its next-nearest exchange couplings are very small (see next section).

We emphasize that our numerical study is based on a microscopic model system, with the values of the three nearest types of *short-range* spin couplings (J_1, J_2, and J_3; see below) taken from the *real material*. In fact, our zero-field MC studies[35–37] indicate that the low-T fractal domain structure with slow dynamics is the underlying physical picture of the SG phase at $x = 0.25$. Such structure actually inhibits the occurrence of the expected fragile AF long-range order in the vicinity of $x_p \approx 0.24$. Indeed, at $x = 0.25$, the sublattices lose their identity, such that the average sublattice magnetizations of the pristine AF structure are virtually zero, though the EA order parameter remains finite.[35] In this context, the very small planar frustrated interaction J_3 has been shown to play a noticeable role only in temperatures $T \lesssim 1$ K, as in the zero-field behavior of the specific heat.[37,38]

Our analysis includes the (off-equilibrium) dynamics of the correlation function, thermoremanent (TRM) and isothermoremanent (IRM) magnetizations in H-shift protocols, and the thermodynamics of low-T hysteresis cycles and ZFC and FC magnetizations, as well as the associated microscopic configuration of spins. Results are obtained for the first-neighbor percolating cluster and the whole sample. We conclude that the behavior of the $Fe_{0.25}Zn_{0.75}F_2$ system at very low T is dominated by the competition between the AF interactions and the applied field. As the temperature raises, our findings are consistent with a vitreous or metastable SG-like thermodynamic state in the context of a modified droplets picture of thermal activation of fractal correlated domains over logarithmic energy barriers.

II. MODEL AND METHODS

We consider the microscopic Hamiltonian of the site-diluted AF Ising system $Fe_xZn_{1-x}F_2$:[39]

$$\mathcal{H} = \sum_{\langle i,i+\delta\rangle} J_{i,i+\delta}\mathcal{E}_i\mathcal{E}_{i+\delta}S_iS_{i+\delta} - \sum_i \mathcal{E}_iS_iH, \qquad (1)$$

where $S_i = \pm 1$ represent Ising spin variables and $\mathcal{E}_i = 0$ or 1 is the random occupation index of the site i of a bcc lattice, with average $\overline{\mathcal{E}_i} = x = 0.25$ in this work. In our simulations, we take into account the three nearest types of short-range exchange couplings of the compound $Fe_xZn_{1-x}F_2$:[39,40] $J_1/J_2 = -0.013$, $J_3/J_2 = 0.053$, and J_2 fixed to obtain the Néel temperature, $T_N(x = 1.0) = 78.4$ K. Notice that J_2 is the dominant AF interaction, whereas J_3 is a small frustrated planar coupling. Indeed, by considering the coordination numbers, $z_1 = 2$, $z_2 = 8$, and $z_3 = 4$, relative to neighbor spins interacting respectively via J_1, J_2, and J_3, one has[39,40] $z_2J_2/(z_1J_1) \approx 307.7$ and $z_2J_2/(z_3J_3) \approx 37.7$. The Zeeman term in $\mathcal{H}$ accounts for the interaction of spins with an external uniform magnetic field H (in units of $g\mu_B$). We consider samples with $N = 2L^3x$ magnetic (Fe^{+2}) sites, $L = 32$, and apply periodic boundary conditions.

In the MC simulation, we perform thermal ($\langle \dots \rangle$) and configurational (disorder) ($[\dots]$) averages over 32 independent samples. The simulation protocols involve cooling and heating processes with either quenched or slow procedures. In the former, the system is taken from the high-T PM phase, at $T = 1.5T_f$, to a low-T configuration, with no intermediate T steps, where $T_f = 9.5 \pm 0.5$ K is the MC freezing temperature in $H = 0$.[35–37] In the latter, the temperature is varied through small $\Delta T = 0.013T_f$ intervals, with 3×10^4 MC steps per spin (MCS) at each T. The mentioned MC estimate for T_f from the critical behavior of the Binder cumulant[37] should be compared with the experimental value, 10.0 ± 0.2 K, obtained from the critical noninear and dc susceptibilities.[11]

The LMF method[33,41–45] is based on the iterative solution of the set of self-consistent equations involving the thermally averaged spins,

$$m_i = \langle S_i\rangle = \tanh[h_i/(k_BT)], \quad \mathcal{E}_i = 1,$$

in which the effective local field of the spin S_i is

$$h_i = -\sum_{i+\delta} J_{i,i+\delta}\mathcal{E}_{i+\delta}m_{i+\delta} + H.$$

Since the LMF states represent local minima in the free-energy surface, the general concept of the method consists of following the evolution of the system among these minima upon changes in T or H. Although thermal fluctuations are not considered in this approach, we observe that the local character of the iterative mean-field equations allows a much deeper description of the system's properties, in comparison with the so-called virtual crystal mean field.[33] Indeed, in the latter, one has that $m_i = m$ for all spins homogeneously distributed in the sample. As in the MC procedure, in the LMF simulation the system is initially cooled from the PM phase at $T = 1.5T_f$, through small T steps of $\Delta T = 0.013T_f$. A change in T is only accomplished after the following convergence criterion[33,41–44] is fulfilled:

$$\frac{\sum_i[(m_i)_n - (m_i)_{n+1}]^2}{\sum_i[(m_i)_n]^2} \leqslant 10^{-6},$$

where the index n denotes the nth iteraction. When the lowest T is reached, the system is heated by the same ΔT, and measurements are done, with disorder average taken over 32 independent samples.

III. MONTE CARLO SIMULATIONS OF DYNAMICS IN A FIELD

Experimental evidence[16] on the compound $Fe_{0.25}Zn_{0.75}F_2$ has suggested that the observed SG behavior might not be thermodynamically stable in a field with decay of the metastable AT line for long times. Therefore, in order to analyze the influence of H on the zero-field SG phase, we start by studying, through MC simulation, the off-equilibrium dynamics of

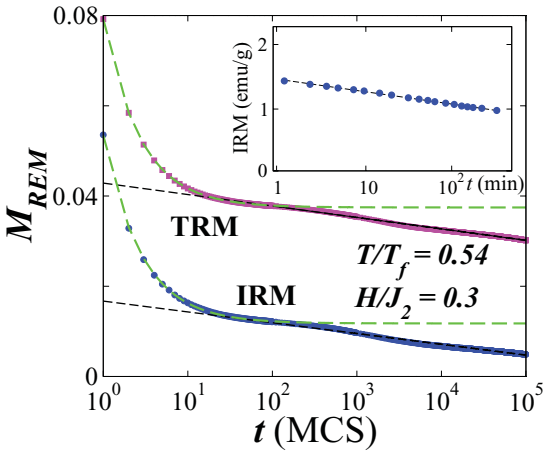

FIG. 1. (Color online) Dynamics of thermoremanent (TRM) and isothermoremanent (IRM) magnetizations of the whole sample after switching off the field $H/J_2 = 0.3$ at $T/T_f = 0.38$. Dashed lines in green and black indicate, respectively, short ($M_{\rm REM} \sim t^{-\beta} +$ constant) and long ($M_{\rm REM} \sim -\gamma \ln t +$ constant) time behaviors. Inset shows the logarithmic decay of the experimental IRM of the compound $Fe_{0.25}Zn_{0.75}F_2$ taken from Ref. 11.

the thermoremanent (TRM) and isothermoremanent (IRM) magnetizations and the spin autocorrelation function.

The IRM (TRM) protocol involves slow cooling from the PM state in (without) a field, down to a low-T phase at $T = 0.38T_f \approx 3.6$ K, in a procedure that closely resembles the experimental protocol.[1,11] In the sequence, a field H is applied during a waiting time $t_w = 5 \times 10^4$ MCS and switched off ($t = 0$). After that, the remanent magnetization,

$$M_{\rm REM}(t,t_w) = \frac{1}{N}\sum_{i=1}^{N}[S_i(t+t_w)],$$

is recorded as the system slowly evolves toward the zero-field SG state. In the dynamics studies, the average is taken only over configurational disorder.

Figure 1 displays the time evolution of IRM and TRM for $H = 0.3J_2 \approx 2.85$ T. A crossover behavior from a rapid power-law decay $M_{\rm REM} \sim t^{-\beta} +$ constant, $\beta \approx 1$, at short times $t \lesssim 10^2$ MCS to a slow logarithmic decay $M_{\rm REM} \sim -\gamma \ln t +$ constant, $\gamma \approx 0.001$, for long times $10^3 \lesssim t \leqslant 10^5$ MCS can be identified. This long-term signature has been also observed experimentally in IRM measurements of the $Fe_{0.25}Zn_{0.75}F_2$ compound,[11] after times typically larger than one minute, as shown in the inset of Fig. 1. This result indicates that a low-T metastable glassy thermodynamic state has set in through this field procedure.

We next consider in Fig. 2 the dynamics of the autocorrelation function,[1,46,47]

$$C(t,t_w) = \frac{1}{N}\sum_{i=1}^{N}[S_i(t_w)S_i(t+t_w)],$$

after a zero-field quench from the PM state to the low-T phase at $T = 0.54T_f \approx 5.1$ K, followed by the application of a field during $t_w = 10^a$ MCS, $a = 1,2,\ldots,5$. After the subsequent turning off of the field ($t = 0$), measurements were performed for times up to $t = 5 \times 10^5$ MCS.

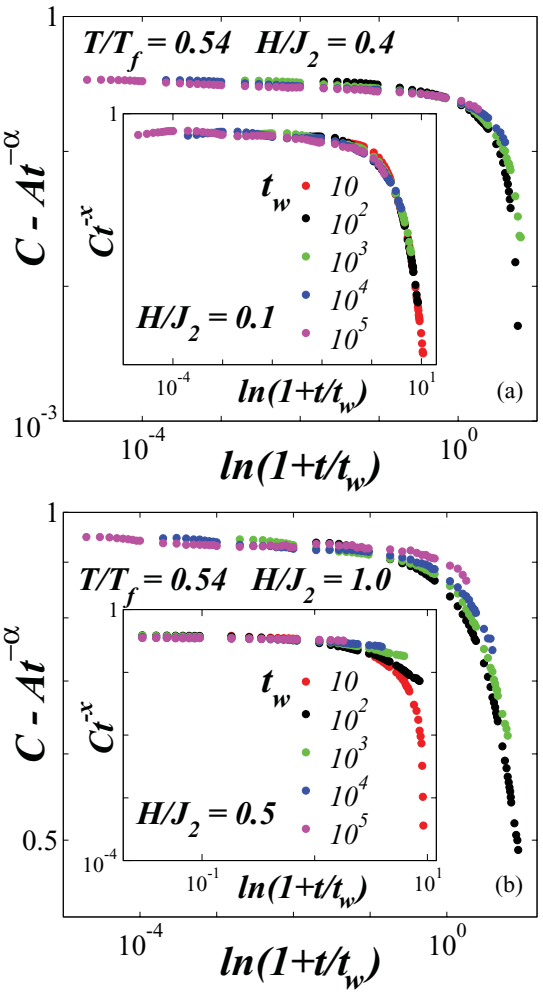

FIG. 2. (Color online) (a) Data collapse of the autocorrelation function of the whole sample, for $t_w = 10^a$ MCS, $a = 1,2,\ldots,5$, through additive ($H/J_2 = 0.4$) and multiplicative ($H/J_2 = 0.1$, inset) scalings at $T/T_f = 0.54$. (b) Absence of data collapse through additive ($H/J_2 = 1.0$) and multiplicative ($H/J_2 = 0.5$, inset) scalings at $T/T_f = 0.54$. Notice that in the additive case, t_w starts at 10^2 MCS.

As previously observed in the MC study of the zero-field SG phase,[35–37] we identify, for a broad range of fields, the slow (quasiequilibrium) and fast (aging) regimes characterized by the asymptotic power-law time decays:[1,46,47] $C \sim t^{-x}$, $t \ll t_w$, and $x(T,H) \approx 0.03$–0.04 and $C \sim t^{-\lambda}$, $t \gg t_w$, and $\lambda(T,H,t_w) \approx 0.06$–$0.15$, respectively. Such behavior leads to a nice collapse of the curves with distinct t_w in the low-H regime,[48] through both multiplicative and additive scalings,[46,47,49,50] respectively:

$$C \sim (t/t_w)^{-x}\Phi_m(t/t_w)$$

and

$$C \sim (t/t_w)^{-\alpha} + \Phi_a(t/t_w),$$

with Φ_m and Φ_a denoting scaling functions [see Fig. 2(a)]. This fact has been regarded[46,51,52] as indicative of a vitreous or metastable SG-like thermodynamic state according to a modified droplets picture of thermal activation over logarithmic energy barriers (see below), which in the present case

is justified by the proximity of the first-neighbor percolation threshold.[53] The exponent obtained in the additive scaling $\alpha \approx 0.01$ is close to the one ($\alpha = 0.02$) reported for both the 3D EA model[49] and the SG compound $AgMn_{2.6}$.[50] In addition, we also observe that the additive scaling leads to a non-null autocorrelation function even for arbitrarily large times, as expected in SGs.[1,46] Nevertheless, as shown in Fig. 2(b), it is remarkable that the data collapse through additive scaling breaks down for fields approaching $H \approx J_2 \approx 9.5$ T, a value that is nearly the AT field for this temperature $T = 5.1$ K (see next section). For the multiplicative scaling, the collapse already starts to fail for fields below this value, as indicated in the inset of Fig. 2(b). As higher fields are considered, the superposition of scaled curves with distinct t_w becomes even less apparent.

The connection between the observed power-law time decay of $C(t,t_w)$ and the presence of logarithmic energy barriers near x_p can be understood as follows. From a microscopic point of view, spins are locally arranged in AF fractal domains with reversed orientation regarding their neighbor clusters (see Sec. V). The distribution $\mathcal{N}(v,t)$ of fractal domains of volume v at a time t is given by $\mathcal{N} \sim v^{-\delta}$, a result valid for the zero-field SG phase at $x = 0.25$,[35–37] as well as for the glassy high-H states of a diluted AF model at $x = 0.50$.[54] Denoting by $c_v(t,t_w)$ the autocorrelation function of domains of volume v at time t, one has in the continuum limit,[37,46]

$$C(t,t_w) = \int_1^{v_{\max}(t)} \mathcal{N}(v,t) c_v(t,t_w) dv.$$

In addition, by considering the Arrehnius dynamics of thermal activation,[46] the typical time

$$t \sim \exp(F_L/k_B T)$$

for a domain of size L to overcome a logarithmic energy barrier close to the percolation threshold[53]

$$F_L \sim \Delta \ln(L/L_0),$$

implies

$$L \sim L_0 t^{k_B T/\Delta},$$

where L_0 is a microscopic length scale and $\Delta(T)$ is the free-energy scale at temperature T. This result, combined with the integral above and the fractal scaling relation $v \sim L^{d_f}$ leads to the observed power-law decay with time of $C(t,t_w)$.

We also mention that the standard droplets assumption[1,5,46] for the energy barriers $F_L \sim \Delta(L/L_0)^{\psi}$ leads to the autocorrelation function dependent on the dynamical variable $\ln t/\ln t_w$ and a logarithmic asymptotic decay of $C(t,t_w)$ with time. In this sense, our attempts of data collapse as function of $\ln t/\ln t_w$ have failed. Moreover, the successful use of a single scaling variable t/t_w is neither consistent with ideas of dynamic ultrametricity in the RSB scenario, involving a complex pattern of time scales organized in a hierarchical way.[1,46]

IV. THERMODYNAMICS AND HYSTERESIS

A. MC Simulations

We generate hysteresis cycles in MC simulations by first slowly cooling the system in zero field from the PM state down to the temperature of interest, which is then kept fixed as the field is varied (increased or decreased) through $\Delta H = 0.05 J_2 \approx 0.48$ T, with thermalization along 5×10^3 MCS at each H. We measure the magnetization of both the whole sample and the first-neighbor percolating cluster (PC) of each sample. In the whole sample, a large number of small clusters nucleate,[35–37] which are more sensitive to the effect of a magnetic field. In contrast, the zero-field SG configuration of the PC, with typically ≈25% of all spins, is expected to be more robust to the influence of the applied field.

Figure 3(a) shows the MC hysteresis loops at a low temperature, $T = 0.013 T_f$. A structure of plateaus of stable magnetization emerges for both the PC and the whole sample (inset), related to discrete flips of Ising spins induced by the field.[55] In such a low-T phase, thermal disorder plays a negligible role, and the behavior of the system is essentially governed by the competition between the AF interactions and the applied field in the highly diluted regime. In this sense, the sequence of plateaus coincides with the number of neighbor spins interacting through the dominant AF coupling J_2 (up to $z = 8$ in the dilute bcc lattice), which flip as the result of these competing factors. The very-low field behavior is shown in Fig. 3(b), for both the whole sample and the PC. The dashed lines are mean-field-like fittings, since the spins giving rise to these low-H magnetizations are either loose spins (whole sample) or spins at domain interfaces (PC) behaving as such, thereby resulting in tiny field-dependent magnetization values before the first plateau of the PC is reached. Regarding the influence of J_3 on the pattern of plateaus in the hysteresis curve, we mention that the competition between the Zeeman and J_3-mediated exchange energies is defined in favor of the former already in the low-H regime, i.e., typically for $H/J_2 \gtrsim H_3/J_2 \equiv x z_3 S J_3/J_2 \approx 0.11$. We have indeed found that the effect of J_3 in the hysteresis curve is present for very low fields, $H/J_2 \lesssim 0.1$, and is manifested in the form of small irregularities in M (due to resolution, however, it is not possible to claim that they actually constitute hyperfine plateaus). These very low-H irregularities are consistently smoothed when $J_3 = 0$. This result also evidences that the role of J_3 is relevant only in the regime of very-low T and H. The plateaus can be also identified with the low-T peaks of the H- and T-dependent susceptibility, dM/dH, shown in Fig. 3(c). We notice that the transition region between the nth and $(n+1)$th plateaus occurs at $H/J_2 = n$, a feature valid up to the saturation limit (not shown), in which all spins align with the field. At $H/J_2 \approx H_{AT}(T \to 0)/J_2 \approx zx = 2$, the hysteresis cycles close, marking the onset of the PM phase at this temperature. In addition, the irreversibility regime becomes evident as the magnetization measurements differ for increasing or decreasing fields.

We also observe in Fig. 3(a) a remarkable difference regarding the hysteresis data of the whole sample and the PC. In the latter the first plateau presents *null* magnetization, indicating that, whereas the PC remains essentially unperturbed up to fields $H/J_2 \approx 1$ at low T, the smaller clusters in the whole

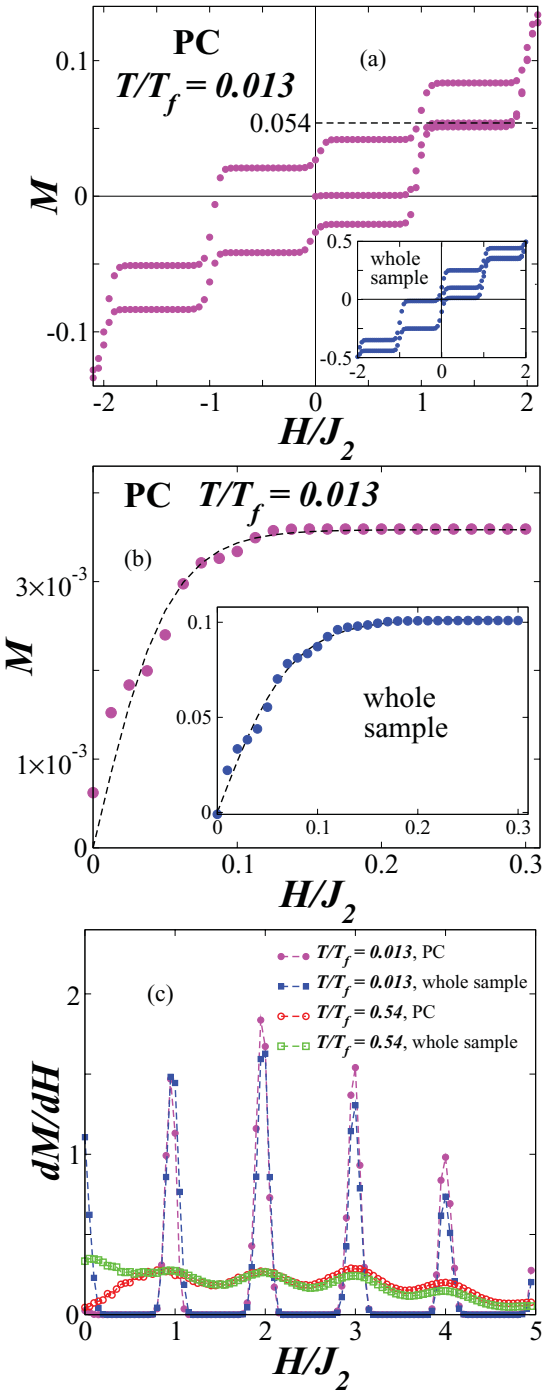

FIG. 3. (Color online) (a) Hysteresis cycles via MC simulation of the PC and whole sample (inset), at $T/T_f = 0.013$, displaying a structure of plateaus. (b) Very low-H behavior of data shown in (a), before the first plateau is reached. Dashed lines are mean-field-like fittings: $M = a\tanh(bH)$. (c) H- and T-dependent susceptibility at $T/T_f = 0.013$ [from data in (a)] and $T/T_f = 0.54$.

sample are susceptible to the influence of rather small fields. At higher temperatures, the hysteresis curves get smoother, with fading of plateaus as an open continuous symmetrical cycle sets in (see also the LMF results for the hysteresis cycles below). Indeed, the acute peaks of dM/dH, shown in Fig. 3(c), are broadened as T increases, and essentially disappear as

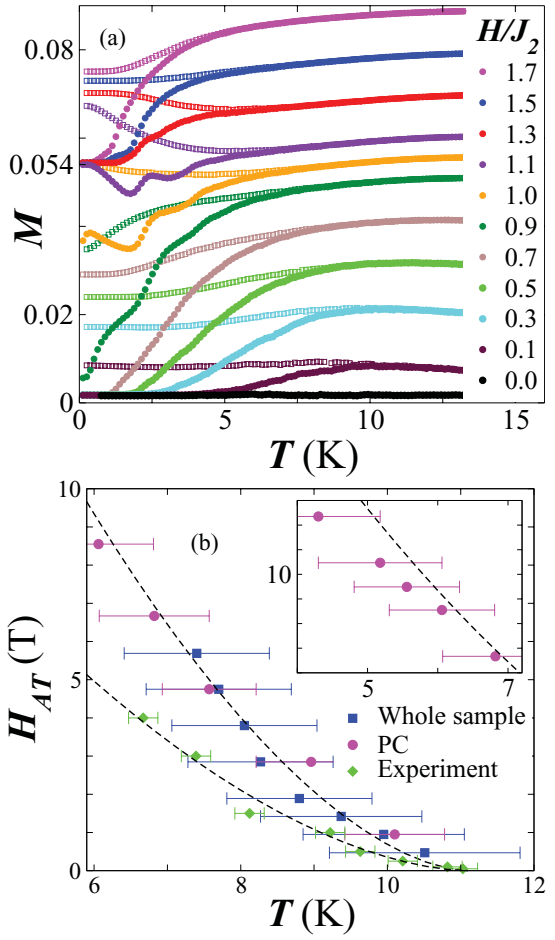

FIG. 4. (Color online) (a) T dependence of the ZFC (full circles) and FC (open squares) magnetizations of the PC for several H, via MC simulation. The low-T $M_{\rm ZFC}$ collapses either at the first ($H/J_2 < 1$) or second ($1 < H/J_2 < 2$) plateaus in (a), i.e., $M = 0$ and 0.054, respectively. (b) AT line for the PC [from data of (a)] and the whole sample, via MC simulation. Experimental data on $Fe_{0.25}Zn_{0.75}F_2$ were taken from Ref. 11. Dashed lines are best fittings for both the experimental and numerical data, leading to the same value of the AT exponent, $\phi = 3.4$. Inset details the region around $T = 0.54T_f \approx 5.1$ K for which $H_{AT} = 1.1J_2 \approx 10.5$ T.

the hysteresis cycle becomes a smooth, continuous curve [see Fig. 5(b) below].

The above findings are corroborated by the thermal dependence of the ZFC and FC magnetization curves of the PC, $M_{\rm ZFC}$ and $M_{\rm FC}$, displayed in Fig. 4(a) for various H, through MC simulation. Similarly to the experimental procedure, in the FC (ZFC) protocol the system is slowly cooled in (without) a field from the PM state down to $T = 0.013T_f$, from which it is slowly heated in a constant field, with measurement of magnetization. We notice that the $T \to 0$ collapses of the $M_{\rm ZFC}$ curves coincide with the respective magnetization value of each plateau in the hysteresis cycle of Fig. 3(a). For instance, the low-T magnetization of the PC is null both in the first plateau of the hysteresis cycle of Fig. 3(a) and in the lowest data collapse of $M_{\rm ZFC}$ as $T \to 0$, shown in Fig. 4(a). Moreover, the magnetization reads 0.054 in the second plateau of the hysteresis cycle, as well as in the second lowest data collapse of the $M_{\rm ZFC}$ curves. For fields in the transition regime between

consecutive plateaus, the low-T $M_{\rm ZFC}$ assumes intermediate values with respect to the stable magnetizations, as seen in Fig. 4(a) for $H/J_2 = 1$.

The AT line establishing the boundary between the vitreous or metastable SG-like thermodynamic state and the PM phase is determined as the H-dependent temperature below which the $M_{\rm ZFC}$ and $M_{\rm FC}$ values start to differ.[11,39] Results are shown in Fig. 4(b) for the whole sample and the PC, as well as for the actual compound $Fe_{0.25}Zn_{0.75}F_2$.[11] In particular, we notice in the inset that $H_{AT}(T = 5.1\ {\rm K}) = 1.1 J_2 \approx 10.5$ T, a value that nearly marks the onset of the regime in which the dynamical additive scaling of the correlation function breaks down, with emergence of the PM phase, as discussed in the previous section. The numerical AT exponent, determined from the best fitting of the scaling relation $T_0 - T \propto H_{AT}^{2/\phi}$, reads $\phi = 3.4 \pm 0.6$. This value is in agreement with the experimental measurement,[11] $\phi = 3.4 \pm 0.2$, and is somewhat larger than the mean-field prediction, $\phi = 3$.[4] However, it might be possible that this result only reflects a vitreous or metastable SG-like thermodynamic state,[19,21,46] as empirically evidenced[16] in the compound $Fe_{0.25}Zn_{0.75}F_2$ through a time-decaying AT line to zero as large times are considered. In this sense, our findings of metastable glassy behavior in H might be related to the finite typical time scale of the MC simulation, a result consistent with the droplets scenario (see discussion below).

B. LMF Results

The MC structure of plateaus seen in Fig. 3(a) is also manifested through the LMF technique [see Fig. 5(a)]. At higher temperatures, the hysteresis cycle gets smoother, as illustrated in Fig. 5(b), becoming completely closed in the fully reversible high-T PM phase, as experimentally observed in the compound $Fe_{0.25}Zn_{0.75}F_2$.[11,39] A continuous hysteresis loop has been also reported for the 3D EA model and the Ising SG compound $Fe_{0.5}Mn_{0.5}TiO_3$, at $T \approx 0.31 T_f$.[56] These findings are also evidenced from the broadening with increasing temperature of the low-T peaks of the H- and T-dependent LMF susceptibility, dM/dH, shown in Fig. 5(c) [compare with the MC data of Fig. 3(c)].

In the LMF context, it is also interesting to probe the stability of the SG phase with respect to the small frustrated interaction, J_3, and the magnetic field. For some time, a debate was settled[33,41,57] on whether such incipient frustration could be the mechanism responsible for the SG phase in the compound $Fe_{0.25}Zn_{0.75}F_2$. Both zero-field measurements[33,41] of the sublattice magnetization and EA order parameter,[58]

$$q = \frac{1}{N}\sum_{i=1}^{N} m_i^2,$$

resulted virtually null for $J_3 = 0$, indicating that, according to the LMF approach characterized by the lack of thermal fluctuation effects, a SG phase in $H = 0$ occurs *only* if $J_3 \neq 0$.

Notwithstanding, a remarkable distinct picture emerges in the presence of a field. As Fig. 6(a) shows, a qualitative change takes place in q even for very low fields, after an FC protocol. In this case, the LMF technique shows a metastable glassy-like thermodynamic state in H either with or without the presence

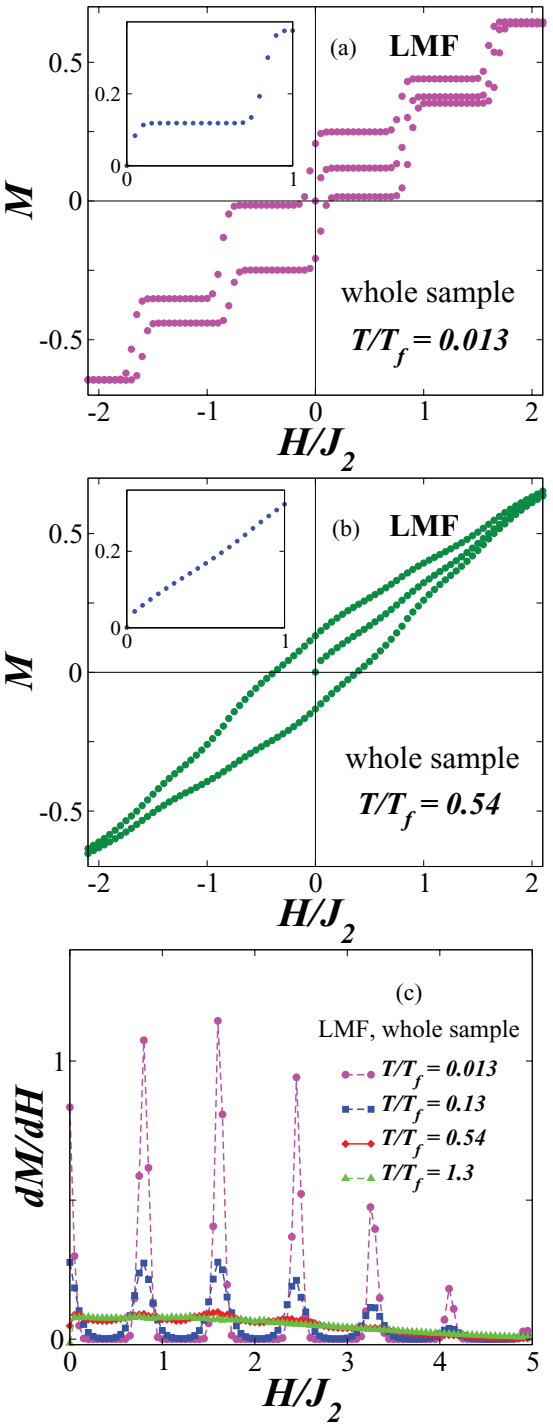

FIG. 5. (Color online) Hysteresis cycles of the whole sample, via LMF simulation: (a) $T/T_f = 0.013$ and (b) $T/T_f = 0.54$. As T increases, the smoothing of the structure of plateaus is evidenced. Insets detail the $H/J_2 \leqslant 1$ regime. (c) H- and T-dependent susceptibility of the whole sample at $T/T_f = 0.013$ [from data in (a)], 0.13, 0.54 [from data in (b)], and 1.3.

of J_3, including similar AT lines.[41] Indeed, both q_{J_3} and q_0 are non-null in $H \neq 0$, with the great stability of $q_{J_3} - q_0$ observed up to fields $H \approx J_2 \approx 10$ T. In this measurement, the negative value of $q_{J_3} - q_0$ expresses the role of the frustrated interaction in decreasing the sublattice magnetization as well as the EA

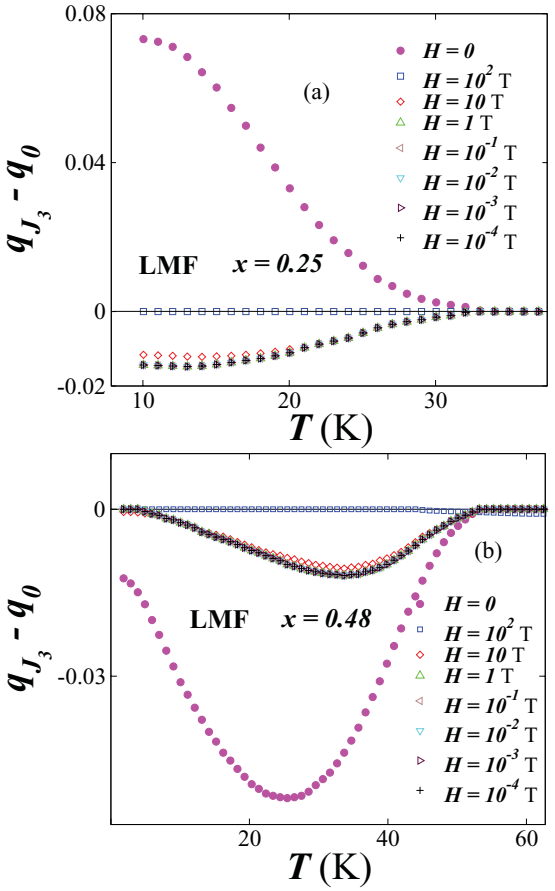

FIG. 6. (Color online) Stability of the EA order parameter of the whole sample regarding the small frustrated interaction J_3 and the field after a FC procedure, through LMF simulation. (a) Zero-field SG and $H \neq 0$ glassy behaviors at $x = 0.25$. (b) Zero-field REIM and $H \neq 0$ RFIM behaviors at $x = 0.48$ (data taken from Ref. 42).

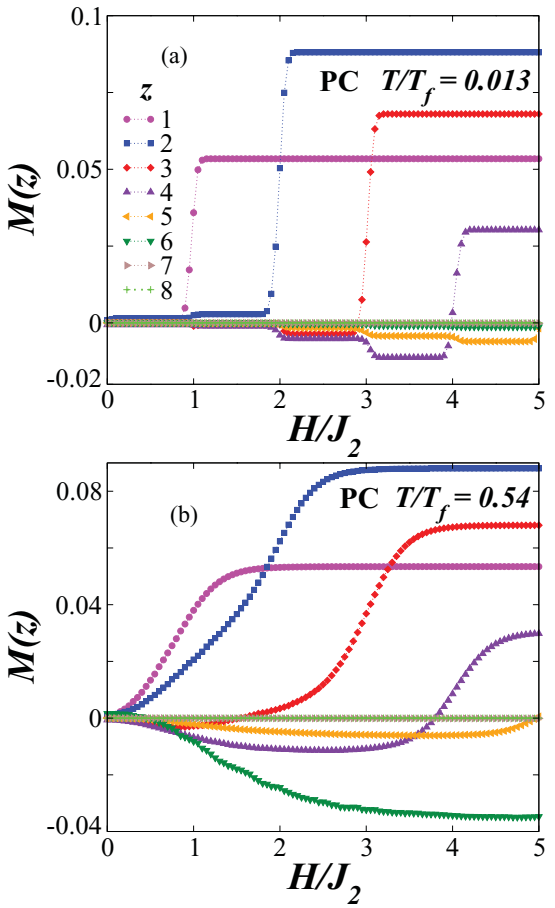

FIG. 7. (Color online) Partial magnetization $M(z)$ of spins with $z \leqslant 8$ neighbors interacting via J_2 in the PC, as function of H, through MC simulation: (a) $T/T_f = 0.013$ and (b) $T/T_f = 0.54$. The successive low-T plateaus in Fig. 3(a) correspond to the onset of the saturation of $M(z)$ for each z.

order parameter. At the saturation limit in very high fields, J_3 plays no role.

We compare the above results with the LMF measurement of $q_{J_3} - q_0$ in the $H = 0$ random exchange and $H \neq 0$ random-field Ising model-like phases (REIM and RFIM) of the system $Fe_xZn_{1-x}F_2$ at a moderate dilution, $x = 0.48$.[39,59] Data shown in Fig. 6(b) were taken from Ref. 42. First, we notice that in zero field, $q_{J_3} - q_0$ is positive (negative) at $x = 0.25$ ($x = 0.48$). The negative value at $x = 0.48$ results from the fact that in the LMF simulation the frustrated coupling reverses a few spin clusters in the established long-range AF order at this moderate dilution. In addition, in both cases even a very small field, e.g., $H = 10^{-4}$ T, disturbs the $H = 0$ states, which in turn behave in a similar way, i.e., exhibiting RFIM-like behavior for increasing field values up to saturation.

V. MICROSCOPIC ANALYSIS

In this section, we present MC results that allow us to provide a microscopic analysis of the magnetization and spin configuration of the system under a magnetic field. In order to better understand the competing mechanisms leading to the above findings, we first analyze the separate contributions, $M(z)$, from spins with $z \leqslant 8$ neighbors interacting through J_2, to the total magnetization, $M = \sum_{z=0}^{8} M(z)$. We display in Fig. 7 MC results for the PC for which the contribution from loose spins ($z = 0$) is absent at $T = 0.013T_f$ and $0.54T_f$. As Fig. 7(a) indicates, the low-T plateaus are successively induced as spins with $z = 1$ (first non-null plateau), $z = 2$ (second), etc., flip at fields $H/J_2 = z$, as discussed. This sequence of plateaus is in one-to-one correspondence with the sequence of low-T peaks of the susceptibility shown in Figs. 3(c) and 5(c). We observe that a nontrivial behavior emerges for spins with large connectivity. Indeed, as the neighbors of a largely connected spin ($z \geqslant 4$) flip with H, they might force the latter to flip reverse to H, due to the strong AF local coupling J_2. As a consequence, a negative contribution, $M(z) < 0$, arises for such spins in a broad range of fields. This effect is amplified when thermal disorder is present, as seen in Fig. 7(b). In addition, by comparing Figs. 3(a) and 7(a) one is led to the conclusion that locally isolated loose spins, with $z = 0$, are essentially responsible for the excess magnetization in the first plateau of the whole sample, for $H/J_2 \lesssim 1$ and $T = 0.013T_f$. On the other hand, at a higher temperature, $T = 0.54T_f$, Fig. 7(b) evidences the smoothing of plateaus, leading to the typical continuous hysteresis cycle

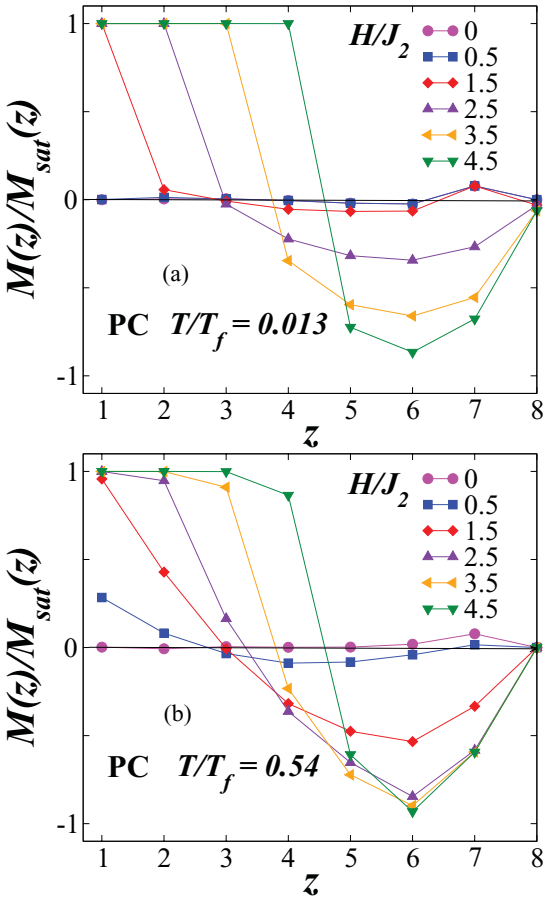

FIG. 8. (Color online) Partial magnetization normalized by its saturation value, $M(z)/M_{\rm sat}(z)$, of spins with $z \leqslant 8$ neighbors interacting via J_2 in the PC, as function of z for several H, through MC simulation: (a) $T/T_f = 0.013$ and (b) $T/T_f = 0.54$. A negative $M(z)$ arises as the neighbors of a largely connected spin flip with H, forcing the latter to flip reverse to H due to the strong AF coupling J_2.

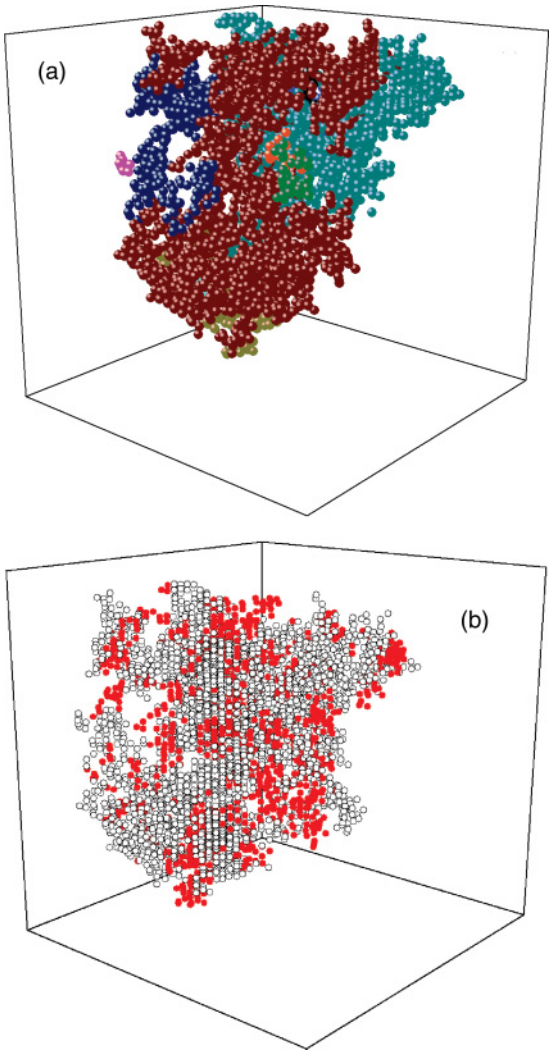

FIG. 9. (Color online) (a) MC microscopic spin configuration of the PC of a SG sample at $T/T_f = 0.013$ and $H = 0$. Symbols with distinct colors depict AF domains of locally connected spins, with reverse orientation regarding their neighbor clusters. (b) *Droplets* of reversed spins (in red) with respect to the configuration in (a), induced by the thermal disorder and magnetic field, at $T/T_f = 0.54$ and $H/J_2 = 1$.

observed in Fig. 5(b) and also experimentally in the compound $Fe_{0.25}Zn_{0.75}F_2$.[11,39]

The above results can be attested in Fig. 8 as well in which $M(z)$, normalized by its saturation value $M_{\rm sat}(z)$, is plotted versus the coordination number z at $T = 0.013T_f$ and $0.54T_f$ for several fields. Two effects can be noticed, consistently with the discussion above. First, at low temperature, $T = 0.013T_f$, and fields $H/J_2 = (2z+1)/2$ at the center of the zth non-null plateau, saturation is achieved for contributions to the magnetization from spins with connectivity $\leqslant z$. Further, the competing mechanism of spin flip reverse to the field is also observed for highly connected spins with $z \geqslant 4$. These effects are softened when thermal disorder comes into play, as seen in Fig. 8(b) at $T = 0.54T_f$. The findings of Figs. 7 and 8 are also corroborated by LMF simulation.

At last, we turn to the analysis of the global effect of the magnetic field on the microscopic spin configuration of the PC. Figure 9(a) displays the zero-field SG configuration of the PC of a sample at low temperature $T = 0.013T_f$ through MC simulation. Noticeably, as shown in Fig. 9(b) for $H/J_2 = 1$ and $T = 0.54T_f$, the combined effect of H and thermal disorder, promotes the collective reversion of clusters of spins with respect to the zero-field configuration of Fig. 9(a), as expected in the "droplets" picture.

VI. DISCUSSION AND CONCLUSIONS

The low-T behavior of the disordered AF Ising compound $Fe_{0.25}Zn_{0.75}F_2$ has been well documented both in $H = 0$ and in a magnetic field.[10,11,13–16,31,39] While the $H = 0$ data imply a quite clear SG phase, the experimental decay[16] of the AT line for long times suggests an H-induced metastable vitreous thermodynamic state, in agreement with the predictions of the droplets scenario.[5,8,17–19,21,46]

Our MC and LMF studies have been conducted by considering a microscopic Hamiltonian with presence of the short-range spin couplings[39,40] taken from the real $Fe_{0.25}Zn_{0.75}F_2$ compound. As previously suggested by a comprehensive

MC study[35–37] of thermodynamical, dynamical, critical, and microscopic properties of this system in $H = 0$, the proximity of the first-neighbor percolation threshold, $x_p \approx 0.24$, induces the emergence of the SG phase characterized by the thermal activation of fractal domains over logarithmic energy barriers.[53] This contrasts with the assumption of power-law energy barriers and compact domains in the original droplets model.[5] The logarithmic barriers also lead to the physical justification,[1,46,54] e.g., the power-law time decay of the autocorrelation function (see Sec. III). In particular, successful data collapse of scaled curves of $C(t,t_w)$ with distinct t_w are found for fields $H/J_2 \lesssim 1$.

In connection with the empirical finding of the AT line decay,[16] the metastable glassy thermodynamic state in H might be related to the finite typical time scale of our simulation results. Interestingly, an unstable glassy phase in H was also reported[60] in the diluted AF Ising model in a bcc lattice as $x \to x_p$, through $T = 0$ exact optimization methods. Thus it is conceivable[19,21,46] that a larger amount of time is required for the breakdown of the SG-like state by the field.

In conclusion, our results on the randomly diluted Ising $Fe_{0.25}Zn_{0.75}F_2$ system in a field indicate the following: (i) at very low temperatures, the behavior of the system is dominated by the competition between the AF interactions and the field, and (ii) as T rises, the vitreous or metastable SG-like thermodynamic state in H is consistent with a modified droplets picture of thermal activation of fractal clusters over logarithmic energy barriers near the first-neighbor percolation threshold.

ACKNOWLEDGMENTS

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