Monte Carlo study of the random-exchange–Ising-model behavior in a diluted antiferromagnet: $Fe_{0.48}Zn_{0.52}F_2$

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We present a microscopic Monte Carlo description of the site-diluted b.c.c. frustrated Ising antiferromagnet $Fe_xZn_{1-x}F_2$, with an emphasis on the random exchange Ising model (REIM) phase observed at high and intermediate magnetic concentrations, $0.3 \lesssim x < 1.0$, in a zero magnetic field. Thermodynamical, microscopic, and critical properties of the $x=0.48$ system are found to be in good agreement with both zero-field experimental measurements and previous Monte Carlo results on the site-diluted s.c. nonfrustrated ferromagnetic Ising model. In particular, we contrast the results of the REIM behavior at $x=0.48$ with those of the zero-field spin-glass phase observed at $x=0.25$.

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The site-diluted b.c.c. frustrated Ising antiferromagnet (AF) $Fe_xZn_{1-x}F_2$ has remained for over two decades as one of the most extensively studied disordered magnetic compounds with respect to thermodynamical, dynamical, and critical properties.[1] In fact, the richness of its phase diagram includes a regime with random exchange Ising model (REIM) behavior at zero magnetic field H and high and intermediate magnetic concentrations, $0.3 \lesssim x < 1.0$, with a crossover to a random-field Ising model (RFIM) behavior for fields up to a lower equilibrium boundary, $H_{\rm low}(T,x)$, where T is the temperature.[2–6] In these not too strongly disordered regimes the AF long-range order (LRO) is sustained at low T. On the other hand, glassy phases with no AF LRO emerge either at intermediate concentrations, $0.3 \lesssim x \lesssim 0.6$, and high fields between the lower and upper equilibrium boundaries, $H_{\rm low}(T,x) < H < H_{\rm eq}(T,x)$ [for $H > H_{\rm eq}(T,x)$ the system is paramagnetic],[7] or just above the percolation threshold, $x_p = 0.24$, at $H=0$.[8–12] In particular, the thermodynamical, dynamical, aging, and critical properties of the strongly disordered spin-glass (SG) phase at $x=0.25$ and $H=0$ has been recently discussed[12] in great detail through heat-bath Monte Carlo (MC) simulations of the system Hamiltonian, with the presence of its actual b.c.c. geometry structure and its short-range exchange interactions. Moreover, we have identified[12] the microscopic mechanism underlying this zero-field SG phase, namely, the effective local random-field distribution associated with the presence of correlated AF fractal domains, independently of the presence of *ab initio* competing interactions.

In this work we present a MC investigation of the $H=0$ thermodynamical, critical, and microscopic properties of the REIM phase of the diluted Ising AF $Fe_xZn_{1-x}F_2$, with an emphasis on the $x=0.48$ system. This description extends our previous studies[6,9] using the local mean-field approach, in which thermal fluctuations are neglected. All the microscopic features of the compound $Fe_xZn_{1-x}F_2$ at the zero magnetic field are present in the Hamiltonian,

$$\mathcal{H} = \sum_{\langle i,i+\delta \rangle} J_{i,i+\delta}\, \varepsilon_i \varepsilon_{i+\delta} S_i S_{i+\delta}, \qquad (1)$$

where $\varepsilon_i = 0$ or 1 denotes the occupation index of site i of a b.c.c. lattice, with average $\bar{\varepsilon}_i = x$, Ising spins are considered, $S_i = \pm 1$, and $\delta = 1,2,3$ concerns the three nearest types of exchange couplings:[1,13] $J_1/J_2 = -0.013$, $J_3/J_2 = 0.053$, and J_2 is fixed so as to obtain the Néel temperature, $T_N(x=1.0) = 78.4$ K; J_2 is the dominant AF interaction, whereas J_3 is a small frustrated planar coupling. Configurational averaging (denoted by $[\cdots]$) is performed over 32 randomly quenched disordered lattices, with independent samples initialized in a paramagnetic spin state at a very high T, from which they are cooled at rates $\Delta T = 1.275$ K (we use $\Delta T = 0.319$ K close to the transition). For thermodynamical measurements, samples with $N = 2L^3 x$, $L = 32$, magnetic sites are considered. For critical properties and scaling analysis purposes, we take $8 \leqslant L \leqslant 32$. At each T, τ MC steps per spin (MCS) (thermal equilibration time) are taken before thermal averaging (denoted by $\langle \cdots \rangle$) during the subsequent τ_m MCS; τ and τ_m values were chosen in order to guarantee the stability of the data for each concentration. For instance, we take $\tau = \tau_m = 10^5$ MCS for $x = 0.25$ and $\tau = \tau_m = 5 \times 10^3$ MCS for $x = 0.48$. Moreover, for the critical analysis at $x = 0.48$ we have considered, e.g., $\tau = \tau_m = 4 \times 10^5$ for $L = 8$ and $\tau = \tau_m = 6 \times 10^4$ for $L = 32$. Periodic boundary conditions are used.

In Fig. 1 we display (a) the $H=0$ thermal dependence of the staggered (sublattice) magnetization $M_S = [\langle M_S(t) \rangle] = [\langle (2/N) \Sigma_i S_i(t) \rangle]$, where the summation is over one of the sublattices at each MC instant t; (b) the staggered linear magnetic susceptibility $\chi_S = N\{[\langle M_S(t)^2 \rangle] - [\langle M_S(t) \rangle^2]\}/(k_B T)^2$, and in (c) the magnetic specific heat $C_m = N\{[\langle \mathcal{H}(t)^2 \rangle] - [\langle \mathcal{H}(t) \rangle^2]\}/(k_B T)^2$, with k_B denoting the Boltzmann constant, for magnetic concentrations $x = 1.0, 0.75, 0.48, 0.40, 0.35, 0.25$. For all x values the total

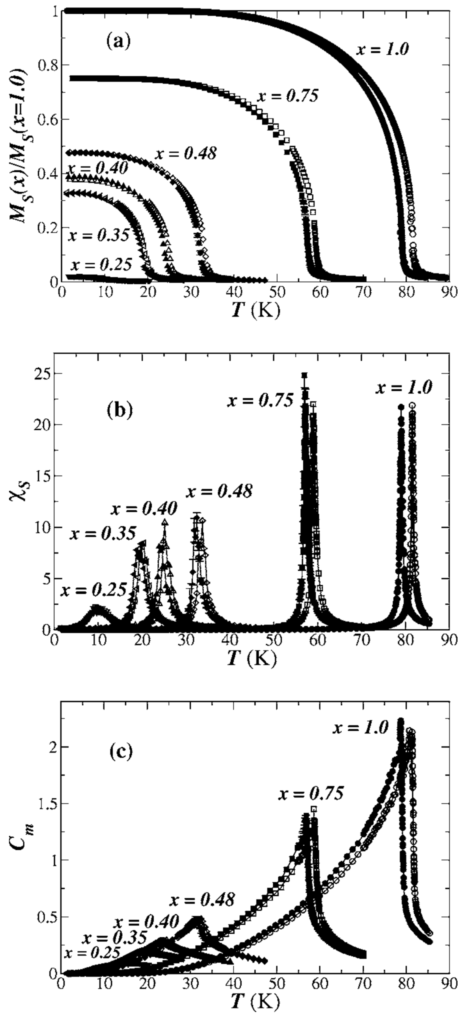

FIG. 1. T dependence of the (a) relative (staggered) sublattice magnetization M_S, (b) staggered linear magnetic susceptibility χ_S, and (c) magnetic specific heat C_m for $H=0$ and $x=1.0, 0.75, 0.48, 0.40, 0.35, 0.25$. Full (open) symbols depict data obtained with $J_3/J_2=0.053$ ($J_3/J_2=0$).

magnetization is zero (within numerical accuracy) at any T. For $x>0.3$ we notice a sharp peak in χ_S and C_m at the Néel temperature $T_N(x)$, as an indicative for $T<T_N(x)$ of the stability of the AF LRO against the site disorder, in agreement with experimental results.[1,3,4,7] In this regime, the AF order parameter, M_S, becomes nonzero for $T<T_N(x)$ and saturates at about $M_S=xM_S(x=1.0)$ at the LRO AF ground state, as also found using a local mean-field approach.[6] However, the magnitudes of the peaks in χ_S and C_m are very sensitive to the choice of the temperature rate ΔT. On the other hand, in the strongly diluted regime close to the percolation threshold, $0.24<x<0.3$, one observes that $M_S\approx 0$ at low T, the linear susceptibility peak rounds considerably and the maximum in C_m becomes a Schottky-like one at a temperature much higher than the critical (freezing) temperature, as typical of SG phases.[1,8,12] In this case, the AF LRO breaks down and a fractal structure of correlated finite domains displaying AF arrangements sets in.[10,12] Interestingly, such behaviors shows little influence of the small frustrated coupling J_3 for $T \gtrsim 1$ K, as can be inferred from Fig. 1. In such cases, the nonfrustrated systems, with $J_3=0$, present slightly higher Néel temperatures in comparison with the frustrated ones, as expected. However, J_3 has been shown to play a distinctive role only in the very low-T specific heat behavior of the SG phase close to the percolation threshold.[11,12]

The above results are corroborated by the analysis of the local effective field distribution $P(h_i)$,[10,12] where

$$h_i=\sum_{i+\delta} J_{i,i+\delta}\varepsilon_{i+\delta}S_{i+\delta}/J_2, \tag{2}$$

for $\varepsilon_i=1$, displayed in Fig. 2 for only one of the sublattices at a temperature close to the ground state ($T=2.6$ K). In fact, whereas for $x=0.48$ a Gaussian-like AF distribution is seen in Fig. 2(a), indicating that all spins in the sublattice point parallel, for $x=0.25$ the fractal structure of finite AF domains, reversed with respect to each other, implies in the

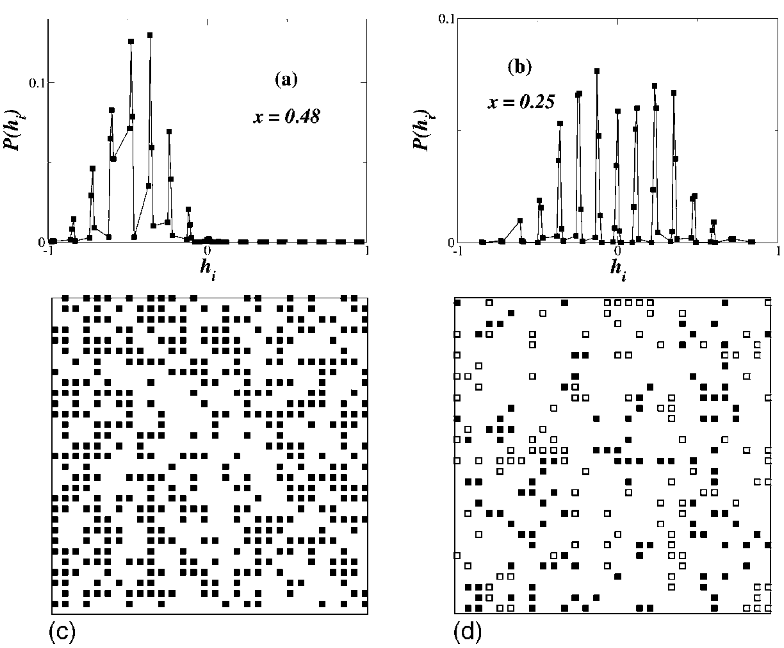

FIG. 2. Distribution at $T=2.6$ K of the $H=0$ sublattice local field h_i at (a) $x=0.48$ and (b) $x=0.25$, with a snapshot of one layer of the respective sublattice spin configuration shown in (c), AF phase (left), and (d), SG phase (right).

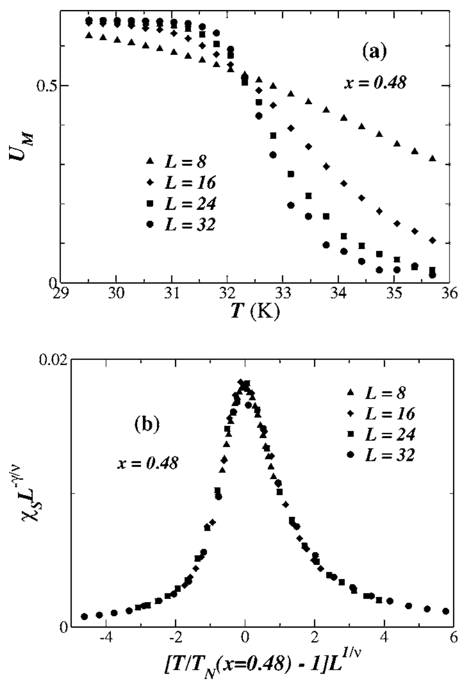

FIG. 3. (a) T dependence of the $H=0$ Binder cumulant U_M for $L=8,16,24,32$. (b) Best collapse of the $\chi_S L^{-\gamma/\nu}$ data versus $L^{1/\nu}[T/T_N(x=0.48)-1]$ for $H=0$ and $L=8,16,24,32$, obtained for $\beta=0.34$, $\nu=0.71$, $\gamma=1.46$, and $T_N(x=0.48)=32.3$ K.

presence of coexisting negative and positive local fields in the same sublattice, together with a relatively large number of losing spins with $h_i \approx 0$, as observed in Fig. 2(b). Indeed, these results are also visualized through the associated one-layer sublattice spin configurations shown in Figs. 2(c) and 2(d): at $x=0.48$ [Fig. 2(c)] all spins in a sublattice point parallel (full squares), thus indicating the presence of a perfect AF LRO; on the other hand, a fractal AF domain structure emerges at $x=0.25$ [Fig. 2(d)], with up (full) and down (empty squares) spins occurring in the same sublattice, implying $M_S \approx 0$ in this zero-field SG phase.

A further characterization of the REIM phase for $x=0.48$ and $H=0$, including the study of its universality class, which differs from that of the pure ($x=1.0$) AF Ising model, requires the scaling analysis near the critical point. In this sense, the severe slowing down of the REIM critical fluctuations, if compared with the transition to the AF state at $x=1.0$, demands a considerable increase in the equilibration and total simulation times, together with a much more detailed sweeping in T in the critical region. The MC Néel temperature, $T_N(x=0.48)=32.2\pm0.3$ K, is determined from the crossing points of the Binder cumulant, $U_M=1-[\langle M_S^4(t)\rangle]/3[\langle M_S^2(t)\rangle^2]$, for $L=8,16,24,32$, as shown in Fig. 3(a). This value should be compared with the mean-field-like experimental[3] and numerical[6,9] finding $T_N(x)=xT_N(x=1.0)\simeq 37.6$ K. The above results contradict the fact that the experimental $T_N(x)$ should exhibit a reduction due to thermal fluctuations, as observed in our MC simulation. This point is claimed for further detailed experimental studies.

The scaling analysis of the MC data for the b.c.c. structure with the small frustrated coupling J_3, using $U_M=f_1(\zeta)$, $M_S=L^{-\beta/\nu}f_2(\zeta)$, and $\chi_S=L^{\gamma/\nu}f_3(\zeta)$ [Fig. 3(b)], with $\zeta \equiv L^{1/\nu}[T-T_N(x=0.48)]$ and f_i representing scaling functions, leads to $\beta=0.34\pm0.02$, $\nu=0.71\pm0.03$, and $\gamma=1.46\pm0.07$, with the goodness of the fitting estimated to be $\chi^2/\mathrm{dof} \approx 0.15$. The exponents obtained independently thus satisfy the scaling relation $2\beta=d\nu-\gamma$. We note that this set of exponents differs from that of the pure Ising model[15] ($\beta=0.3250\pm0.0015$, $\nu=0.6300\pm0.0015$, and $\gamma=1.241\pm0.002$), therefore implying that randomly diluted and pure $x=1.0$ AF Ising systems actually belong to distinct universality classes. In addition, our $x=0.48$ MC results compare well with the experimental β ($=0.350\pm0.009$) and ν ($=0.69\pm0.01$) exponents at $x=0.9$[5] and $x=0.46$[2], respectively. A less better agreement is found for the γ exponent ($\gamma=1.33\pm0.02$ from the $x=0.46$ compound[2] and $\gamma=1.34\pm0.06$ from the $x=0.93$ compound[4]), which might be related to the larger sample-to-sample variation in the susceptibility MC measurements, although the scaling relation remains valid. In addition, our MC exponents also present relatively good agreement with those of the site-diluted simple-cubic (s.c.) nonfrustrated ferromagnetic Ising model obtained from an extensive MC study[16] ($\beta=0.3546\pm0.0018$, $\nu=0.684\pm0.002$, and $\gamma=1.342\pm0.005$), as well as from several analytical results[17] ($\beta=0.343-0.354$, $\nu=0.668-0.697$, and $\gamma=1.306-1.336$), which point to the universal critical behavior of the REIM.[16]

For the sake of coherence, it is also worthwhile to contrast the above MC results for the $x=0.48$ REIM system with those of the SG phase at $x=0.25$ and $H=0$:[12] $\nu=1.4\pm0.1$, $\beta=0.5\pm0.1$, and $\gamma=3.2\pm0.1$, calculated using the same microscopic model in Eq. (1). We add that this SG set of exponents is also in relatively good agreement[12] with that of a short-range SG with binary coupling distribution[18] ($\nu=1.3\pm0.1$, $\beta=0.5$, and $\gamma=2.9\pm0.3$), as well as with those of experimental SG compounds: $Fe_{0.25}Zn_{0.75}F_2$[8] ($\nu=1.4\pm0.2$, $\beta=1.0\pm0.2$, and $\gamma=2.3\pm0.3$) and $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_{13}$ (Ref. 19) ($\nu=1.39$, $\beta=0.38$, and $\gamma=3.4$).

In summary, we have presented a microscopic MC description of the site-diluted b.c.c. frustrated Ising AF $Fe_xZn_{1-x}F_2$, with an emphasis on the REIM phase observed at $x=0.48$ and $H=0$. Thermodynamical, microscopic, and critical properties are found to be in good agreement with both zero-field experimental measurements[2–5] on the $Fe_xZn_{1-x}F_2$ compound for $0.3 \lesssim x < 1.0$ and with previous MC results on the site-diluted s.c. nonfrustrated Ising ferromagnetic model,[14,16] thus pointing to the universal critical behavior of the REIM.

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