The random field critical concentration in dilute antiferromagnets

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Monte Carlo techniques are used to investigate the equilibrium threshold concentration, $x_e$, in the dilute anisotropic antiferromagnet $Fe_xZn_{1-x}F_2$ in an applied magnetic field, considered to be an ideal random-field Ising model system. Above $x_e$ equilibrium behavior is observed whereas below $x_e$ metastability and domain formation dominate. Monte Carlo results agree very well with experimental data obtained using this system.

The dilute antiferromagnet (AF) $Fe_xZn_{1-x}F_2$ in an applied field is a realization of the random-field Ising model (RFIM). Many studies have been done on this system for $x < 0.75$. In such cases, metastability and domain formation mask the equilibrium critical behavior, particularly in scattering measurements that are by nature dominated by long-range antiferromagnetic correlations. The specific heat, $C_m$, on the other hand, is not as greatly affected by domain formation except very close to the transition, $T_c(H)$, since it is primarily sensitive to short range correlations. Only recently has it been discovered that equilibrium scattering behavior can be observed for $x = 0.93$ with no evidence of domain formation. Domain walls form with little energy cost when vacancies are so numerous that magnetic bonds can be largely avoided. At high magnetic concentrations domain walls must cut a large number of magnetic bonds and long-range order (LRO) is stable, as in the Imry-Ma domain wall energy arguments. Hence, the RFIM can be studied in equilibrium for $x = 0.93$ and a transition to LRO is observed, consistent with theory. The question remains as to the nature of the disappearance of the domain walls as the magnetic concentration increases. Do they disappear gradually or is there a critical concentration above which they do not form?

We have performed Monte Carlo (MC) simulations of the RFIM modeled as closely as possible
after Fe\(_{1-x}\)Zn\(_x\)F\(_2\). We provide evidence that there is a critical equilibrium threshold concentration, \(x_c\), above which domain formation does not occur. We estimate this concentration to be \(0.75 < x_c < 0.80\). The simulation results also shed light on the experimentally observed formation of domain walls upon approach to \(T_c(H)\) for \(x < x_c\) after cooling in zero field and subsequently heating in a field (ZFC). Finally we demonstrate that cooling in a field (FC) for \(x < x_c\) results in a metastable state at low \(T\).

The staggered magnetization (\(M_s\)) and correlation functions for the first three nearest-neighbor (NN) pairs have been calculated using MC simulations. The magnetic lattice corresponding to the body-centered-tetragonal Fe\(_{1-x}\)Zn\(_x\)F\(_2\) lattice is described as two cubic sub-lattices of size \(L \times L \times L\) each, delineated as one dimensional arrays bit coded to accommodate large lattice sizes. Most of the results reported here were obtained with \(L = 64\), corresponding to more than \(5.2 \times 10^5\) sites magnetically occupied with probability \(x\). Periodic boundary conditions are used. At each temperature magnetic sites are visited randomly with each site visited an average of once per MC step. Upon each visit, a spin is flipped with a probability given by the metropolis algorithm. The temperature was changed in steps of 0.01 K after \(N\) MC steps. \(N\) was increased until the results of the simulation were largely independent of \(N\) for \(H = 0\). Unless otherwise stated, we used \(N = 500\) for the results reported here. Using \(N = 1000\) gave essentially identical results for \(H = 0\). The temperature scanning procedures correspond to the experimental ones, i.e. cooling and heating for \(H = 0\), and FC and ZFC for \(H = 13\) T. Fe\(_{1-x}\)Zn\(_x\)F\(_2\) is well represented by the Hamiltonian

\[
H = \sum_{<ij>} J_{ij} \vec{S}_i \cdot \vec{S}_j - D \sum_i \vec{S}_i^2 - \vec{H} \cdot \sum_i \vec{S}_i .
\]  

(1)

In the simulations we use the Hamiltonian

\[
H = \sum_{<ij>} J_{ij} S_i^z S_j^z - H \sum_i S_i^z ,
\]  

(2)

which corresponds to the Ising limit \(D \to \infty\). The first three NN exchange interaction strengths are taken from spin-wave dispersion measurements on FeF\(_2\): \(J_1 = -0.069\) K; \(J_2 = 5.278\) K; and \(J_3 = 0.279\) K. All other interactions are negligible and are not included in the simulations. The transition temperature increases with anisotropy in this system. Hence, the infinite anisotropy
of the model results in a transition temperature much higher than in the real system. As is done elsewhere [11], the included interactions were all scaled by a factor 2/3 to make the transition temperatures more closely correspond to the ones observed [12] in Fe$_{1-x}$Zn$_x$F$_2$.

Figure 1 and its inset show typical behavior for $x = 0.60 < x_c$ for three cases: $H = 0$, which is free of hysteresis; FC with $H = 13$ T; and ZFC with $H = 13$ T. In the inset, the behavior of $C_m = dE/dT$ vs. $T$ is shown. For $H = 0$ random-exchange behavior is observed [3] with slight rounding from finite sample size effects. Taking the rounding into consideration [13], we estimate the transition temperature $T_N \approx 45.0$ K. For the ZFC and FC procedures, the specific heat is depressed in $T$ as expected. Away from the shifted transition, the two behaviors are essentially identical with a shape more symmetric than for $H = 0$, as is observed in experiments [4]. Since the ZFC and FC behaviors are the same outside the distorted regions, they clearly share the same $T_c(H)$. On a plot of the specific heat versus the logarithm of the reduced temperature, $T_c(H)$ appears close to the peak of the ZFC data, $\approx 40.5$ K. From the main figure, we see that for $H = 0$, $M_s$ falls to zero very near the $T_N$ taken from $C_m$. Upon FC, however, $M_s$ remains unusually small quite far below the $T_c(H)$ determined from $C_m$. Upon further cooling, the system begins to rapidly evolve toward AF LRO with a curvature opposite to that for $H = 0$. Finally, at a rather well defined temperature ($T_1$) the $M_s$ curve abruptly changes curvature. Upon further cooling, $M_s$ increases smoothly and slowly, much like the $H = 0$ case but shifted to lower $T$. Upon ZFC, $M_s$ vs. $T$ follows a much smoother evolution but nevertheless approaches $T_c(H)$ with a curvature opposite to the $H = 0$ case. The unusually rapid disappearance of AF LRO well below $T_c(H)$ is attributed to the formation of domains and has been observed experimentally [8]. The FC behavior in experimental systems, on the other hand, is quite different from that shown in Fig. 1. In the experiments, the domain formation prevents significant $M_s$ down to low $T$. Hence, we examined this behavior further using the MC techniques. We found that by slowing the temperature scans by using $N = 1000$ with $L = 64$, the domains that form for $x < x_c$ remain stable to much lower temperatures. We also did a simulation with $L = 128$ and $N = 500$ and again the domains were stable to lower temperatures. Extrapolating these two trends to the much larger experimental systems that are cooled much more
slowly relative to the spin flipping time, it is not surprising that the experimental systems show metastable domains that are very stable at low temperatures.

Figure 2 shows the ZFC and FC behavior for $x = 0.60, 0.70, 0.80$ and $0.90$ at $H = 13$ T. It is clear that the hysteresis decreases as $x$ increases and is absent for the cases $x \geq 0.80$. It thus appears that the systems at larger $x$ are in equilibrium. To demonstrate that there is a critical threshold concentration above which equilibrium behavior, such as that observed for $x \geq 0.80$, prevails, we plot in Fig. 3 the temperature $\Delta T = T_2(H, x) - T_1(H, x)$, where $T_2(H, x)$ is the temperature at which the ZFC joins the FC one just below $T_c(H)$. Note that the ZFC and FC curves separate at $T_1(H) < T_2(H)$. It is clear that $\Delta T$ becomes zero somewhat above $x = 0.70$ and below $x = 0.80$, strongly indicating a critical concentration $0.75 < x_c < 0.80$, which is much larger than the magnetic percolation threshold $x_p = 0.25$. The field dependence of $x_c$ is weak. Within the accuracy of the present MC results, we find that for $H = 7$ T and $H = 13$ T, $0.75 < x_c < 0.8$. This suggests that the critical concentration is primarily geometric in origin. Domain formation in the ZFC procedure indicates that for $H > 0$, the free energy must favor domain formation close to $T_c(H)$ for $x < x_c$. Such behavior is indicated in local-mean-field simulations [14]. However, there is no evidence for a latent heat associated with the temperature at which the free energy difference for the domain state and AF state switches sign.

Finally, we demonstrate in the inset of Fig. 3 that the ZFC peak in $d(M_s^2)/dT$ (the Bragg scattering) vs. $T$ for $x = 0.60$ is at a lower temperature by more than 1 K than $T_c(H)$ as determined from the $C_m$. This behavior is in agreement with neutron scattering results [8] but in contradiction to the so-called ‘trompe l’oeil’ phenomenology [15].

In conclusion, we have presented MC evidence for the existence of a critical concentration $x_c$ above which equilibrium RFIM behavior prevails and below which domains form close to the transition. The numerous vacancies for $x < x_c$ allow domain formation without a great energy cost. We note that the vacancies themselves percolate at $x = 0.75$ and surely $x_c$ is related to this percolation threshold, although this may not be the entire story. Domain walls take advantage of surfaces of vacancies, not just the filamentary percolation threshold structures. Hence, it would not be
surprising if \( x_e \) were somewhat different from the vacancy percolation threshold. Further simulations are currently under way at \( L = 128 \) and should help to determine \( x_e \) more precisely as well as determine the influence of the finite sample size on the present results. Certainly, critical behavior studies should be performed in the real systems only for \( x > x_e \).

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FIG. 1. $M_s$ vs. $T$ for $x = 0.60$ at $H = 0$, and for ZFC and FC at $H = 13$ T. The transition temperatures are determined from the specific heat behaviors shown in the inset where the units are expressed in terms of two magnetic sites per unit cell. In both the main figure and inset, curves are, from left to right, for the FC, ZFC and $H = 0$ procedures.
FIG. 2. The ZFC and FC $M_s$ vs. $T$ for $x = 0.60$ (a), 0.70 (b), 0.80 (c) and 0.90 (d). The upper curves are ZFC in each case except $x = 0.80$ and 0.90, where the ZFC and FC curves coincide at all temperatures.
FIG. 3. The temperature range over which hysteresis occurs at $H = 13 \, T$, $\Delta T = T_2(H, x) - T_1(H, x)$ vs. $x$. The inset shows $d(M_s^2)/dT$ (arbitrary units) and $C_m/R$ vs. $T$ for $x = 0.60$. Note that the peak in $d(M_s^2)/dT$ vs. $T$ is significantly lower than the corresponding peak in $C_m/R$. 