Instability of Long-Range Order in a $d = 3$ Random-Field Ising Model System: $Fe_xZn_{1-x}F_2$

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Instabilities of long-range order are observed in samples of the d = 3 dilute uniaxial antiferromagnet $Fe_xZn_{1-x}F_2$ with $x = 0.56$ and 0.41, under strong random fields. The onset of instability, mapped in the (H, T) phase diagrams of the samples, reveals that the H, T and x dependence of the effective random field is in qualitative agreement with a mean-field expression predicted in the weak-field limit for site diluted antiferromagnets.

The influence of random impurities and random fields on the phases and phase transitions of Ising systems has attracted considerable attention, both from the theoretical and experimental sides [1]. More than two decades ago, energetic arguments of domain formation were used by Imry and Ma to demonstrate [2] that long-range order (LRO) is not destroyed by arbitrarily weak random fields in more than $d = 2$ dimensions in Ising systems. A rigorous proof of this latter assertion was established years later [3]. If the magnitude of the random field exceeds the field of neighboring spins, it is expected that the spins follow the random field, destroying LRO. Experimentalists have been attracted to the random field Ising model (RFIM) problem after the realization [4] that the application of a uniform magnetic field (H) in a diluted antiferromagnet generates local random staggered fields.

In this paper we explore the instability of LRO in samples of the highly anisotropic diluted antiferromagnet $Fe_xZn_{1-x}F_2$, with $x = 0.56$ and $x = 0.41$, due the presence of strong local random fields. The magnetization measurements have been performed in a wide range of magnetic fields $0 < H < 18T$, applied parallel to the easy magnetization direction, in the T range $2 < T < 50K$. Measurements were made using a vibrating sample magnetometer adapted to a 20 Tesla superconducting magnet at the National High Magnetic Field Laboratory, Los Alamos Facility. The $Fe_xZn_{1-x}F_2$ system is recognized as the best experimental realization of the random-field Ising model in three dimensions ($d = 3$). Neutron scattering studies performed in samples of $Fe_xZn_{1-x}F_2$ with Fe concentrations in the range 0.3×0.9, have shown that the AF LRO, effective at $H = 0$ for $T < T_N$ (T_N is the Néel temperature), is stable against the presence of *weak* random fields. In other words, when a *weak* external field is applied parallel to the easy axis of the compound, AF LRO is maintained. The instability of AF LRO, characterized here by the time dependence of the magnetization, occurs beyond a given magnitude of the local random fields. For both samples, the line marking onset of this instability in the (H, T) plane reveals that the magnitude of the effective random field is in qualitative agreement with Cardy's expression[5], obtained by a mean-field approach in a site- disordered Ising antiferromagnet, in the *weak-field* limit:

$$< h_{RF}^2 >_{av} = \frac{x(1-x)[T_N^{MF}(0)/T]^2(H/\kappa_B T)^2}{[1-\theta^{MF}(x)/T]^2}, \quad (1)$$

where x is the concentration of magnetic ions, κ_B is Boltzmann's constant, θ_N^{MF} is the mean-field Néel tem-

perature and MF is the mean-field Curie-Weiss parameter (see Ref.5).

For $Fe_{0.56}Zn_{0.44}F_2$, the time dependencies of the magnetization, normalized by the initial magnetization $M_0 \equiv M(t=0)$, are displayed for increasing field (FI) in Figs. 1 and 2 for $T = 10K$ and $4K$, respectively. In the FI procedure, the sample is first zero-field cooled (ZFC) from the paramagnetic phase to a given $T < T_N$. Then the field is increased to the value at which the time dependence is measured. For $T = 10K$, our magnetization data indicates the stability of the AF LRO, at least for $H \leq 10T$. For higher values of H, the local random fields become stronger than the field produced by the neighboring spins, inducing a time dependent spin flipping, characterized as a glassy phase in earlier works [6-9]. The rate of flipping is strongly dependent on the magnitude of the random fields. This rate, however, decreases in the proximity of the paramagnetic phase, as illustrated in Fig. 1, by comparing the curve for $H = 17K$ with the ones at lower fields. The inset shows the field dependence of the initial magnetization M_0. A similar behavior is found in the M/M_0 versus t curves displayed in Fig. 2, for $T = 4K$. In this case, however, AF LRO is no longer stable at $H = 10T$, contrasting with the behavior at the same value of H for $T = 10K$. This reflects the higher magnitude of the local random fields at lower temperatures, for fixed x and T, as theoretically predicted by equation (1). The time dependencies studied in Figs. 1 and 2 are mapped by the asterisks in the phase diagram of Fig. 3. The critical, $T_c(H)$, and equilibrium, $T_{eq}(H)$, phase boundaries are represented by full and dashed lines, respectively. These boundaries have been obtained by M *vs* T and $M vs H$ cycles, as reported in Ref. 9. $T_c(H)$ was determined in the high-H experiments by the field position of the peak in the field derivative of M_{FI} (dM_{FI}/dH), for each value of T (see data with vertical error bars). In the low-H regime, due to the shape of the (H,T) phase diagram, it is convenient to determine $T_c(H)$ from the position of the peaks in dM_{FH}/dT, which appear at low H when the sample is heated in the presence of a fixed H, after it has been zero-field cooled from the paramagnetic phase to a given $T < T_N$ (see data with horizontal error bars). These curves are not shown here (see Refs. 8 and 9), but it is noteworthy that the positions of the dM_{FI}/dH peaks in the (H,T) phase diagrams of Fig. 3 correlate quite well with the customary peaks in dM_{FH}/dT. Similarly, the upper equilibrium boundary, $H^u_{eq}(T)$, is in good correlation with the previously defined $T_{eq}(H)$ for the low-H experiments. We would like to stress here the significance of the novel lower equilibrium boundary, $H^l_{eq}(T)$. For $H < H^l_{eq}$, $\Delta M = 0$ and the system recovers its AF minimum-energy configuration by decreasing H from the paramagnetic phase. The $H^l_{eq}(T)$ boundary tends to reach the $H = 0$ line at a temperature $T^*(x)$. For $T < T^*$, the $H = 0$ AF LRO is not recovered by FD in the time scale of the experiments. On the other hand by increasing T, the $H^l_{eq}(T)$ line approaches a singular region in the (H,T) phase diagram. A multicritical (or possibly end-critical) point separates a low-H line of phase transitions, governed by the universal REIM-RFIM crossover scaling, from a pseudo-critical boundary at high-H, where strong random fields induce a glassy behavior.[7]

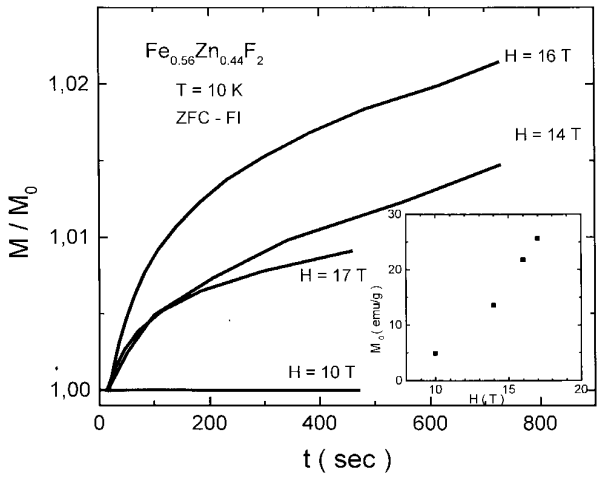

Figure 1. Time dependence of the magnetization $M(t)$ following the FI procedure (see text) at $T = 10K$ up to $H = 10T$, $14T$, 16 T and 17 T. The measurement begins as soon as the field reaches the above values. The magnetization is normalized by the initial value M_0, which is plotted versus H in the inset.

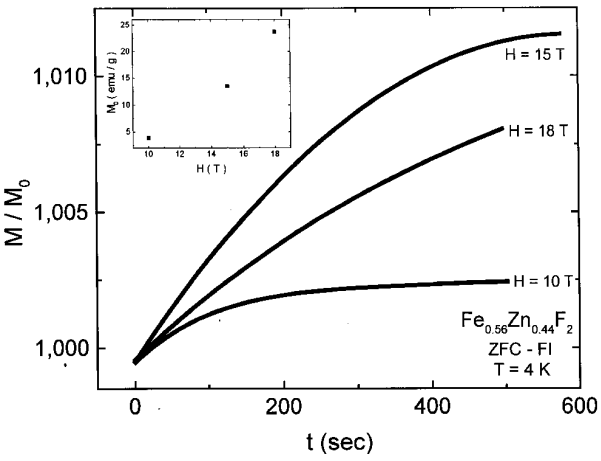

Figure 2. The same as in Fig. 1, but for $T = 4K$ and $H = 10T$, $15T$ and 18 T.

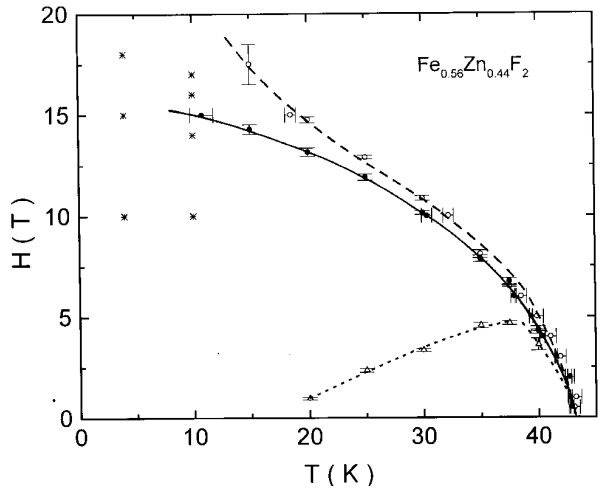

Figure 3. Critical and irreversibility phase boundaries in $Fe_{0.56}Zn_{0.40}F_2$. Full symbols, with horizontal (or vertical) error bars, represent $T_c(H)$ originating in the position of the peaks in the $MvsH$ (or $MvsT$) plots (see Ref. 9) for zero-field-cooled $Fe_{0.56}Zn_{0.44}F_2$. The asterisks represent the positions where time dependence of the magnetization were presented in Figs. 1 and 2.

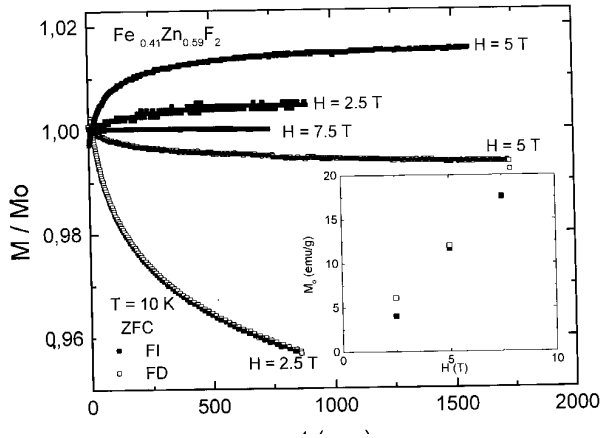

Figure 4. Time dependence of the magnetization $M(t)$ following the FI procedure in $T = 10.0K$ up to $H = 2.5T$, 5.0 T and 7.5 T and by field decreasing (FD) the field from the paramagnetic phase to $H = 5.0T$ and 2.5 T. The magnetization is normalized by the initial value M_0, which is plotted versus H in the inset.

From equation (1), the magnitude of the random field increases as x decreases, for fixed T and H. So, it is expected that for $Fe_{0.41}Zn_{0.59}F_2$ the random-field induced spin flipping occurs for lower H, for the same values of T used in the study of $Fe_{0.56}Zn_{0.44}F_2$. This corresponds, indeed, to the scenario of the time dependencies of $Fe_{0.41}Zn_{0.59}F_2$, depicted in Fig. 4 and 5, for $T = 10K$ and $4K$, respectively. The field dependence of the initial magnetization M_0 is plotted in the inset of Fig. 4, for $T = 10K$ and in Fig. 6, for $T = 4K$. For $T = 10K$, the time dependence of M is effective

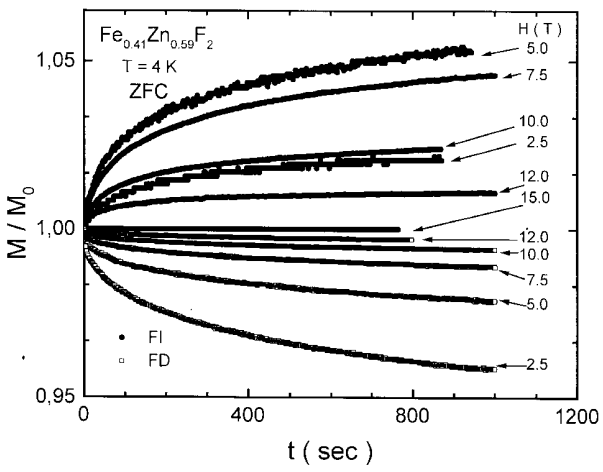

Figure 5. The same as in Fig. 4 but for $T = 4.0K$ and several values of H, both FI and FD procedures.

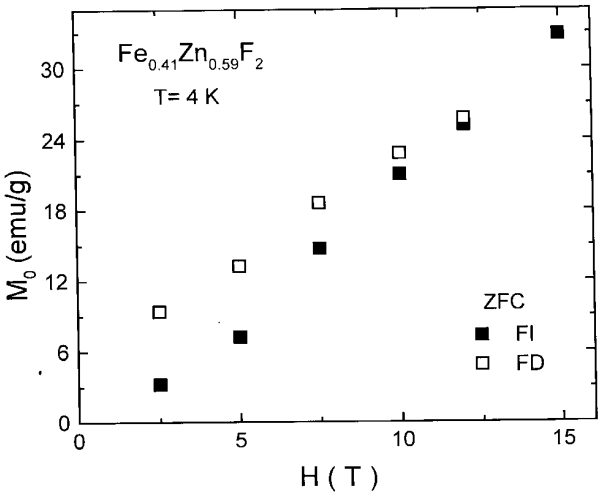

Figure 6. The initial magnetization M_0 plotted versus H for $T = 4.0K$ in the FI and FD procedures.

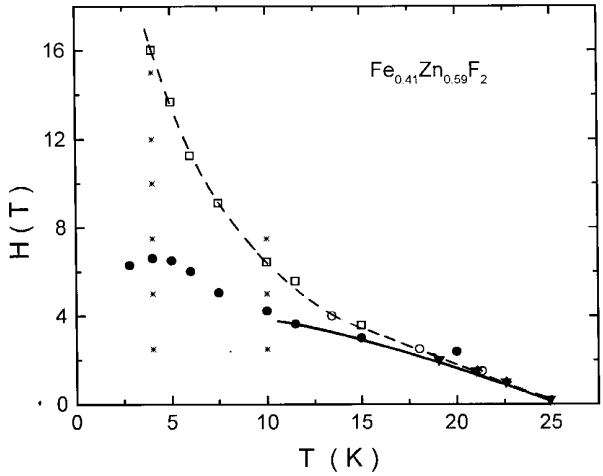

Figure 7. Critical (full symbols) and irreversibility (open symbols) phase boundaries in $Fe_{0.41}Zn_{0.59}F_2$. The asterisks represent the positions where time dependence of the magnetization are presented in Figs. 4 and 5.

for field as low as $H = 2.5T$, in the FI procedure. The relaxation rate increases for $H = 5T$ for the same T. The (H, T) position of the latter point lies a little above the virtual line of the destroyed AF-paramagnetic transition $T_c(H)$, as seen in the phase diagram of Fig. 7. For $H = 7.5T$ the system is stable in the paramagnetic phase. The time dependencies of M when the field is decreased (FD) from the paramagnetic phase are also presented in Fig. 4, for the same T values. In the latter procedure the relaxation rates are dramatically different from the ones observed in the FI protocol, as easily seen comparing, for instance, the FI and FD curves for $Fe_{0.41}Zn_{0.59}F_2$ at $H = 2.5T$ in Fig. 4.

In conclusion, we presented magnetization measurements on the diluted antiferromagnets $Fe_{0.41}Zn_{0.59}F_2$ and $Fe_{0.56}Zn_{0.44}F_2$, which show that the instability of AF LRO, observed in the upper part of the phase diagrams of these samples, shows relaxation effects strongly dependent on the magnitude of the local random fields, and also on the field and temperature cycling protocols. To the best of the our knowledge, a theoretical study of the dynamics in diluted antiferromagnets submitted to strong local random fields is still lacking. So, a comparison of the relaxation effects presented in this work with theoretical results is not feasible at the moment. We hope that the presented results motivate further theoretical and experimental efforts on this subject.

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