Random-field-crossover scaling in $Mn_{0.35}Zn_{0.65}F_2$

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The weakly anisotropic and highly diluted antiferromagnet $Mn_{0.35}Zn_{0.65}F_2$ has been studied by dc magnetization measurements under a uniform magnetic field and by ac susceptibility at zero field. Earlier experiments in $Mn_xZn_{1-x}F_2$ for $x>0.4$ have shown that under an external field applied parallel to the easy direction [001] the critical phase boundary $T_c(H)$ is governed by a random-exchange Ising model (REIM) to random-field Ising model (RFIM) crossover scaling. This crossover scaling stipulates that $T_N-T_c(H)\sim H^{2/\phi}$, where $\phi\cong 1.4$ is the universal REIM-RFIM crossover exponent. We present magnetization results which show that for $x=0.35$, $T_c(H)$ has a different curvature following the above scaling law but with $\phi\cong 3.4$ (similar to the de Almeida–Thouless line in spin glasses). Zero-field ac susceptibility (χ_{ac}) measurements performed on this sample reveal a freezing of spatial fluctuations at low temperatures. The magnetic features of $Mn_{0.35}Zn_{0.65}F_2$ are distinct in some aspects from the spin-glass behavior found in the isostructural but more anisotropic compound $Fe_xZn_{1-x}F_2$ close to the percolation threshold.

I. INTRODUCTION

Dilute uniaxial antiferromagnets (DAF's) have been extensively studied as a testing ground for some of the most challenging models of random magnetism.[1] In the absence of external fields ($H=0$) and provided the exchange interactions between nearest neighbors do not include frustrated bonds, anisotropic DAF's can be represented by the random-exchange Ising model (REIM). In three dimensions ($d=3$) the best example of a REIM system is the $Fe_xZn_{1-x}F_2$ compound, where the Ising character comes from the large and x-independent[2] single ion anisotropy. The isostructural $d=3$ system $Mn_xZn_{1-x}F_2$ possesses only a weak x-dependent anisotropy of dipolar origin. Pure FeF_2 and MnF_2 are body-centered-tetragonal antiferromagnets with the rutile structure, where the spins are oriented along the c axis. In the absence of exchange frustration, antiferromagnetic (AF) long-range order (LRO) in the infinite cluster is expected to occur for the entire range of x above the percolation concentration, i.e., the line of continuous phase transition terminates at $T=0$ for $x=x_p$. Because of the dominant[3,4] next-nearest-neighbor interactions J_2, x_p is estimated[5] to be close to 0.245 in both $Fe_xZn_{1-x}F_2$ and $Mn_xZn_{1-x}F_2$ compounds. However, recent neutron-scattering measurements[6] in two samples of $Fe_xZn_{1-x}F_2$, with $x=0.25$ and $x=0.27$ showed absence of AF LRO at $H=0$ upon cooling to $T=2$ K. This result is in agreement with previous magnetization[7,8] and ac susceptibility[9] studies in $Fe_{0.25}Zn_{0.75}F_2$ from which it was deduced a spin-glass (SG) behavior occurring at $H\geq 0$ for T below a freezing temperature $T_f\simeq 10$ K. These studies suggest an apparent shift of the percolation threshold concentration to a larger value in $Fe_xZn_{1-x}F_2$. In this system, AF LRO with a characteristic REIM critical behavior has been reported[10] at $H=0$ for Fe concentrations $x\geq 0.31$. In spite of the weak Ising character of $Mn_xZn_{1-x}F_2$, the asymptotic critical behavior at zero field, for $x>0.4$, is similar[11,12] to that found in $Fe_xZn_{1-x}F_2$,[13–15] in agreement with $d=3$ REIM theoretical predictions.[16–18] At $H=0$, AF LRO has been observed[19] to occur for Mn concentrations very close to the percolation threshold. However, the critical behavior has been, to the best of the authors' knowledge, explored only for $x>0.4$ in $Mn_xZn_{1-x}F_2$.

As well known, a uniform field applied parallel to the easy direction of a DAF compound produces an equivalence[20] to the random-field Ising model (RFIM) for the ferromagnet. But for the ferromagnet, there is a crossover from pure Ising to RFIM behavior when the random field is "turned on." In the case of the dilute antiferromagnet, which corresponds to a random-exchange system, the crossover is from REIM to RFIM.[21] In the presence of a uniform field parallel to the c axis, REIM–RFIM crossover behavior has been observed for all measured[22] samples with $x\geq 0.31$ in $Fe_xZn_{1-x}F_2$. However, if the random field becomes sufficiently strong, LRO is destroyed and a glassy phase is induced in the upper part of the (H,T) phase diagram, as recently observed[23,10] in $Fe_{0.31}Zn_{0.69}F_2$. The process evolves to an Ising spin-glass phase dominating the whole (H,T) diagram for values of x very close and also below the geometric percolation threshold $x_p=0.24$. In $Mn_xZn_{1-x}F_2$, good fits for the REIM–RFIM crossover scaling have also been obtained[24–26] for $x>0.4$, under *weak* random fields. In this later compound the situation for $x<0.4$ is still controversial. On one hand, Bazhan and Petrov[27] claim from magnetization and ac susceptibility studies, that in the concentration range $0.2\leq x\leq 0.33$ and $H=0$, $Mn_xZn_{1-x}F_2$ converts into a state of a time-varying spin glass at low temperatures. The magnetic moments of the Mn^{2+} ions are randomly distributed. Under an applied field, the system assumes a stationary state, but the magnetic moments remain randomly oriented with respect to the direction of the applied field. On the other hand,

neutron-scattering studies by Cowley *et al.*[19] sustains that at $H=0$ the termination of the line of the AF-paramagnetic (P) continuous phase transition occur at $T=0$ at $x=x_p$. In fact, the dipolar anisotropy present in $Mn_xZn_{1-x}F_2$ is expected to become random in strength and direction as x decreases. If this is the case, one is led to ask how the phase diagram of this compound evolves upon dilution, for strong randomness. It would be of considerable interest to compare the nature of phase diagrams in this system, much closer to the percolation threshold and for all ranges of x and H/J, with the corresponding ones[23,8] in the isostructural compound $Fe_xZn_{1-x}F_2$ where the single-ion anisotropy is x independent.

In this paper we investigate the low-temperature phase and critical behavior of the strong dilute AF compound $Mn_{0.35}Zn_{0.65}F_2$ by magnetization ($H\neq 0$) and ac susceptibility ($H=0$) techniques. The measurements have been made in a comprehensive range of applied fields and temperatures, such that a large part of the (H,T) diagram can be accessed. The present study establishes an unusual shape for the critical phase boundary and indicates a number of magnetic features which distinguishes the $Fe_xZn_{1-x}F_2$ and $Mn_xZn_{1-x}F_2$ compounds close to the percolation threshold.

The paper is arranged as follows. The theoretical predictions and experimental verifications related to the REIM–RFIM crossover behavior in dilute antiferromagnets are discussed in Sec. II. The experimental method is described in Sec. III. The experimental results and discussion are presented in Sec. IV. Finally, in Sec. V we give an interpretation of the present results and make a comparison with experimental work performed in $Fe_xZn_{1-x}F_2$.

II. RANDOM-EXCHANGE TO RANDOM-FIELD CROSSOVER BEHAVIOR

The REIM–RFIM crossover behavior stipulates that if a sharp phase transition exists, the new transition temperature T_c is expected to occur at[20]

$$T_c(H)=T_N-bH^2-cT_n h_{\rm RF}^{2/\phi} , \tag{1}$$

where T_N is the Néel temperature, $h_{\rm RF}$ is the reduced rms random field, and c is a constant of order unity. For the site-diluted case the mean-square-reduced random field, in the limit $h_{\rm RF}^2 \ll 1$, is given by[28]

$$h_{\rm RF}^2=\frac{x(1-x)[T_N^{\rm MF}(1)/T]^2(g\mu_B SH/k_B T)^2}{[1+\Theta^{\rm MF}(x)/T]^2} . \tag{2}$$

In Eq. (2) $T_N^{\rm MF}(1)$ is the mean-field Néel temperature in the pure system, and $\Theta^{\rm MF}(x)$ is the mean-field Curie-Weiss parameter. So, in DAF systems, for a given x the magnitude of the random field can be controlled by the strength of the applied field. Combining Eqs. (1) and (2), one obtains that the variation of the transition temperature in the (H,T) phase diagram is given by

$$\Delta T_c(H)=T_N-bH^2-T_c(H)=cT_N h_{\rm RF}^{2/\phi}=CH^{2/\phi} , \tag{3}$$

where the value of the crossover exponent is given by[21] $\phi\approx 1.1\gamma$, with γ being the REIM staggered susceptibility exponent. All experiments on both $Fe_xZn_{1-x}F_2$ (Ref. 22) and $Mn_xZn_{1-x}F_2$ (Refs. 22, 24, and 26) systems under *weak* random fields and $x>0.4$ have in common a critical phase boundary in the (H,T) diagram governed by the same $\phi=1.42\pm 0.03$ REIM to RFIM crossover scaling exponent. Dramatic slow dynamics appear[29–32] close to the transition temperature. These effects are manifested in two distinct ways depending on the prior history of the sample. If the system is cooled in zero field (ZFC) to the long-range ordered (LRO) antiferromagnetic state, then a uniform field is applied and the system is heated approaching $T_c(H)$, the critical behavior is dynamically rounded by the extreme critical slowing down inherent to the RFIM problem. On the other hand, if the transition temperature is approached cooling first the sample in a field (FC), the system cannot achieve AF long-range order and freezes in a nonequilibrium domain state in the laboratory time scale. This behavior has been observed both in $Fe_xZn_{1-x}F_2$ and $Mn_xZn_{1-x}F_2$ compounds with $x>0.4$. The equilibrium is lost at a field-dependent temperature boundary $T_{\rm eq}(H)$ which lies just above the transition boundary $T_c(H)$ obtained from ZFC procedure. $T_{\rm eq}(H)$ is the temperature above which different field cycling procedures give the same results in all measurements made in the same time scale. It has been observed[33] that $T_{\rm eq}(H)$ scales with the field obeying a RFIM crossover scaling similar to that given in Eq. (3) for $T_c(H)$, i.e.,

$$\Delta T_{\rm eq}(H)=T_N-bH^2-T_{\rm eq}(H)=C_{\rm eq}H^{2/\phi} . \tag{4}$$

Good fits have been obtained[22,23,33] from $T_{\rm eq}$ data in $Fe_xZn_{1-x}F_2$ for $x=0.73$, 0.40, and 0.31, using $\phi\approx 1.42$. In $Mn_xZn_{1-x}F_2$, a precise determination of ϕ has been not obtained yet from the equilibrium phase boundary.

The above described critical and equilibrium behavior occurs within the *weak* random-field limit, which depends on both the magnitude of the applied field H and the concentration of magnetic ions x, as can be seen from Eq. (2). Above this *weak* limit deviations from the REIM–RFIM scaling behavior, given by Eq. (3) with $\phi=1.42$, are theoretically expected[34–38] and were actually observed[23,39] to occur in diluted antiferromagnets. As x decreases, the AF LRO state becomes unstable[10] against random fields of strong magnitudes, giving place to a "glassy phase" in the upper part of the (H,T) diagram. A striking reversal of the curvature of the equilibrium boundary $T_{\rm eq}(H)$ is observed,[23] now scaling with H as the de Almeida–Thouless line in spin glasses, i.e., Eq. (4) is obeyed with $\phi\cong 3.4$. The value $\phi=3.4$ governs[7,8] the whole equilibrium phase boundary, for values of x very close to and also below the percolation concentration $x_p=0.24$ in $Fe_xZn_{1-x}F_2$. A major purpose of the present study is to verify how the (H,T) phase diagram evolves on approaching x_p from above in $Mn_xZn_{1-x}F_2$.

III. EXPERIMENTAL METHOD

The single-crystal of $Mn_{0.35}Zn_{0.65}F_2$ used in the present experiments has its concentration determined by density

measurements. The crystal has the dimensions $4.62\times4.53\times4.88$ mm^3 with the easy direction [001] perpendicular to one of the 4.62×4.53 mm^2 faces. We have measured the magnetization (M) in $Mn_{0.35}Zn_{0.65}F_2$ with a vibrating sample magnetometer adapted to work in a split coil superconducting magnet. The temperature and applied field were varied in the ranges $4.2<T\leq30$ K and $0<H\leq30$ kOe, respectively. The sample holder geometry and temperature controller used in the experiments are capable of a temperature control within 1 mK using a carbon glass thermometer. H was applied along the easy direction [001]. The field (H) and temperature (T) dependences of M were measured using temperature cycling (H fixed) and field cycling measurements (T fixed). The temperature cycling was made in the usual zero-field-cooling (ZFC) and field-cooling (FC) procedures. In ZFC, the crystal is cooled in zero field to a given temperature, then the field is raised and finally the temperature is increased. Data were obtained for consecutive T steps, stabilizing T before each reading. In the procedure FC, data are obtained upon cooling in a field, with the temperature stabilized before each reading. In the field cycling measurements the sample is first zero-field cooled from the paramagnetic phase to a certain temperature. Then the field is increased (FI) and finally decreased (FD), while the corresponding M and h values are stored in a computer. The field was swept at a rate of 2 kOe/min. The ac susceptibility (χ_{ac}) in $Mn_{0.35}Zn_{0.65}F_2$ was measured, from 4.2 to 30.0 K with no dc applied magnetic field, with an automated mutual inductance bridge. The frequency of the ac modulating field ranged from 47.3 to 1733.3 Hz, while its amplitude was maintained at $H_{ac}\sim1.0$ Oe.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Finite field magnetization results

Temperature cycling of the susceptibility (M/H) is presented in Fig. 1 for applied fields in the interval $3.5\leq H\leq10$ kOe, using the ZFC and FC procedures described above. Similar plots are shown for lower values of H in Fig. 2. For $0<H<7.5$ kOe ZFC curves of Figs. 1 and 2 are those typical of a predominant AF order taking place at low temperatures. However, no previous neutron-scattering data are available to confirm AF LRO in $Mn_xZn_{1-x}F_2$ under uniform field for $x<0.4$. For $H>8$ kOe ZFC data are no longer compatible with AF ordering. A spin-flop (SF) phase would be expected to occur for large H, as previously observed and characterized[26] by M measurements in $Mn_{0.5}Zn_{0.5}F_2$. It has been shown[26] that if an AF-SF phase boundary is crossed by varying T, the appearance of a minimum in M indicates the change from AF to SF arrangement of spins at a temperature $T_{sf}(H)$. No clear signature of this phase transition is seen in the $(M/H)\times T$ curves of Fig. 1. So, in the present case, two possibilities may be conjectured. (1) AF-SF phase boundary exists but is somewhat horizontal in shape; (2) there is a gradual evolution to a mixed AF-SF configuration in the H and T ranges of the present experiments. These aspects will be discussed in more detail later on.

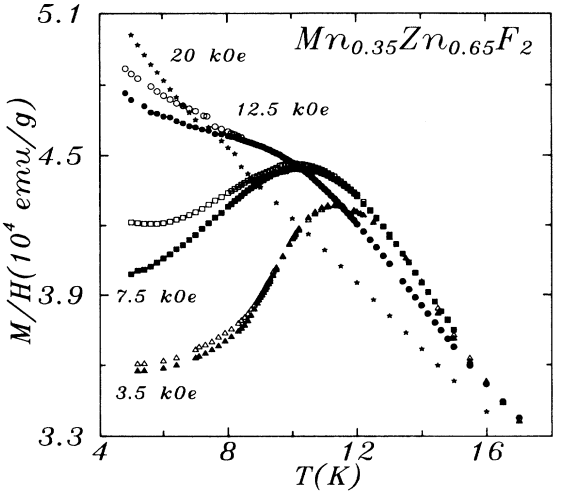

FIG. 1. M/H versus T measured on $Mn_{0.35}Zn_{0.65}F_2$ for applied fields in the range $3.5\leq H\leq20.0$ kOe, comprising the intermediate- and high-field regimes (see text), using ZFC (full symbols) and FC (open symbols) procedures.

Another low-temperature feature evidenced in Figs. 1 and 2 is the presence of an excess of magnetization ($\Delta M_H\equiv M_{FC}-M_{ZFC}$) in the FC curves as compared with the ZFC ones, in the field range $0.5\leq H\leq12.5$ kOe. A better understanding of the irreversible behavior of the magnetization in $Mn_{0.35}Zn_{0.65}F_2$ can be attained if we separate the field interval of measurements in three field regimes: low-field regime ($0<H\leq2$ kOe); intermediate-field regime ($2\leq H\leq10$ kOe); and high-field regime ($10\leq H\leq20$ kOe). The irreversible behavior found at the intermediate-field regime is similar to the behavior commonly observed in samples with large x on both $Mn_xZn_{1-x}F_2$ and $Fe_xZn_{1-x}F_2$. ΔM_H increases with increasing H (see Fig. 1), as expected[40–42] from the nu-

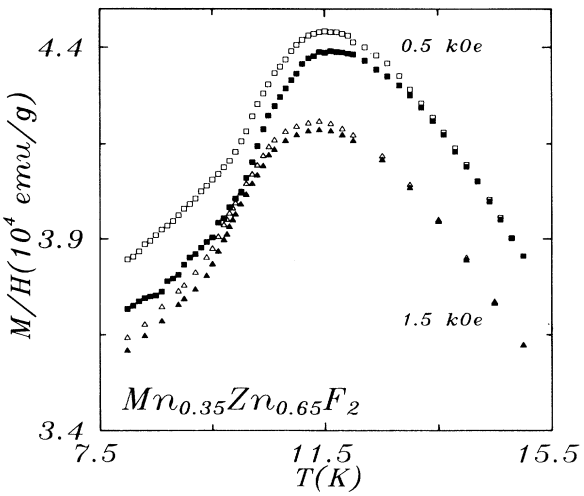

FIG. 2. M/H versus T measured on $Mn_{0.35}Zn_{0.65}F_2$ in the low-field regime ($0.5\leq H\leq1.5$ kOe), following ZFC (full symbols) and FC (open symbols) protocols.

cleation of field aligned domains on cooling diluted uniaxial antiferromagnets from the paramagnetic (P) phase under an applied field parallel to the easy axis. These FC-induced metastable domains are also found to exhibit[43,44] low-temperature relaxation. However, no significant time dependence has been observed in the time-scale of acquisition of M_{FC} data (typically 5 min per point). Dynamic effects were not explored here in a long-time basis. The most surprising irreversible feature is found in the low-field regime, where it has been found that the excess of magnetization ΔM_H reverses its field dependence as compared with the behavior seen at intermediate fields (compare the ZFC-FC irreversibilities of Fig. 1 with the corresponding ones in Fig. 2). The origin of this irreversible behavior at lower fields is not clear. However, the evolution of the low-H irreversibilities with decreasing H could even raise suspicion about the existence of a REIM behavior at $H=0$ in this sample.[27]

It has been proposed[40,41] that for DAF systems, in a given time scale (τ) the excess of magnetization will have the dependence on the uniform field H

$$\Delta M_H \propto H^{\nu_H} . \tag{5}$$

Faraday rotation experiments[42] in $Fe_{0.7}Zn_{0.3}F_2$ yield $\nu_H=2.0\pm0.1$. This result indicates that the excess of magnetization arises from surface contributions of the frozen-in domains. However, a value $\nu_H=3.05$ has been measured[45] in $Fe_{0.48}Zn_{0.52}F_2$, indicating a volume contribution ($\nu_H=3.0$) for ΔM_H. Of course, none of the above possibilities can explain the low-field dependence of the irreversible behavior in $Mn_{0.35}Zn_{0.65}F_2$ (where ν_H was found to be negative). The reversal in the field dependence of ΔM_H is accompanied by a corresponding inversion of the H dependence of both the ZFC and FC amplitudes of the dc susceptibility (M/H), as can be seen comparing Figs. 1 and 2. As H increases, the decreasing rates of both ΔM_H and the amplitude (M/H) become smaller and stabilizes around $H=2$ kOe. On further increasing H above 2 kOe, ΔM_H, and (M/H) have their H dependences reversed, so that both quantities increase with H, which is compatible with Eq. (5) within the field limits of the above defined intermediate-field regime. On increasing H even more, the sample reaches the high-field regime where the maximum in (M/H) disappears and ΔM_H decreases with increasing H. For $H\geq20$ kOe the ZFC-FC irreversibilities are no longer seen and a typically paramagnetic behavior appears.

The temperature derivative of the parallel susceptibility has been largely employed to determine the AF-P critical phase boundaries $T_c(H)$ in pure[46] and dilute[23,26,42,47,48] AF's. In the vicinity of $T_c(H)$, $d(M/H)/dT$ is proportional[49] to the asymptotic critical behavior of the magnetic specific heat C_m, which has a peak at $T_c(H)$. A plot of the ZFC temperature dependence of $d(M/H)/dT$ is presented in Fig. 3 for values of H in the range $0.5\leq H\leq7.5$ kOe. The positions of $d(M/H)/dT$ peaks shift to lower temperatures and become drastically rounded as H increases. The rounding of the peaks in the present case is possibly not entirely due to concentration gradients and RFIM critical slowing

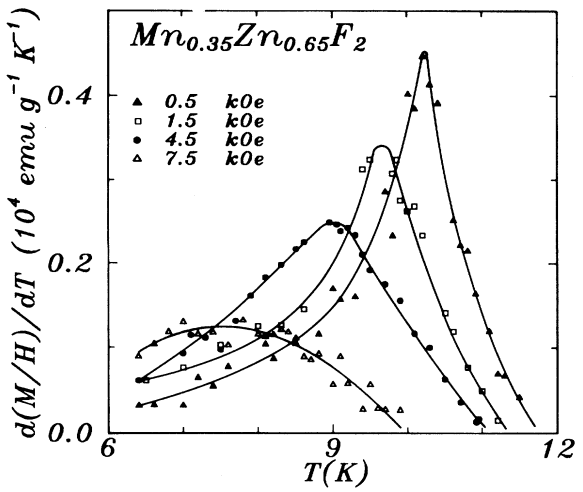

FIG. 3. ZFC temperature dependence of $d(M/H)/dT$ on $Mn_{0.35}Zn_{0.65}F_2$, for $0.5\leq H\leq7.5$ kOe. The solid lines are guides to the eye.

down, as experimentally observed[24–26] for $x>0.4$ in $Mn_xZn_{1-x}F_2$. For weakly diluted samples, the critical behavior is governed by the REIM–RFIM crossover scaling given in Eq. (3), with a REIM–RFIM crossover exponent $\phi\simeq1.4$. In contrast to the critical behavior found[23,24,36,47,50] in RFIM systems, the amplitude of the $d(M/H)/dT$peaks decreases as H increases in $Mn_{0.35}Zn_{0.65}F_2$. This H dependence of the magnetization is incompatible with a REIM–RFIM crossover scaling. A critical-point analysis of the scaling form[20] of the RFIM free energy predicts that the leading singularity of the temperature derivative of M at a fixed H is given by[47]

$$(\partial M/\partial T)_H \sim H^y|t|^{-\tilde{\alpha}} , \tag{6}$$

where y and $\tilde{\alpha}$ are critical exponents and $t=(T-T_c)/T_N$. Assuming a REIM–RFIM crossover scaling, a value $y=0.56$ is predicted[47] and experimentally confirmed[47,50] in $Fe_{0.47}Zn_{0.53}F_2$ and $Fe_{0.7}Mg_{0.3}F_2$. It can be noted by inspection of data in Fig. 3 that the use of Eq. (6) implies that $y<0$, in disagreement with the prediction for a REIM–RFIM crossover behavior. The REIM–RFIM crossover breaking in $Mn_{0.35}Zn_{0.65}F_2$ is shown directly by the own shape of the $T_c(H)$ "critical phase boundary," measured from the position of peaks in $d(M/H)/dT$ curves of Fig. 3. The resulting (H,T) phase diagram is presented in Fig. 4, for $0<H\leq5$ kOe. By using Eq. (3) to fit the data of Fig. 4, we obtain $\phi=3.4\pm0.2$, similar to the de Almeida–Thouless line in spin glasses. $T_N=10.8\pm0.1$ K was obtained from peaks in the ac susceptibility measured at zero field in the same sample used in the present work (see Fig. 9). The position of T_N coincides[51] with the extrapolation of the $T_c(H)$ data to $H\rightarrow0$. As no frequency dependence was found in the close vicinity of T_N in the ac susceptibility data and the temperature sensors used in both experiments have been previously calibrated, we can conclude that the convex "$\phi\cong3.4$ regime" extends to $H\rightarrow0$ in $Mn_{0.35}Zn_{0.65}F_2$. The $T_c(H)$ "critical phase boundary" is also shown in the

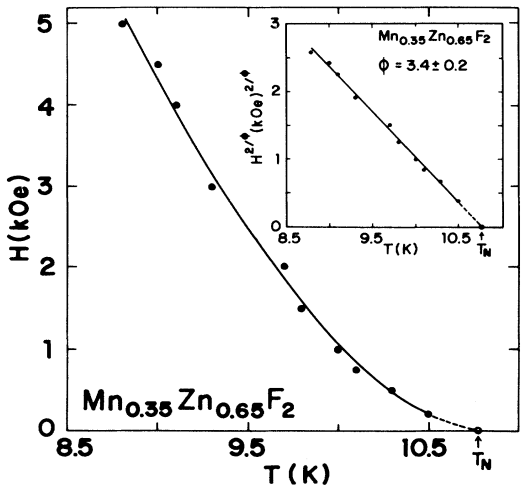

FIG. 4. The critical phase boundary $T_c(H)$ in $Mn_{0.35}Zn_{0.65}F_2$. The inset shows the same data in a $H^{2/\phi}$ vs T plot. The solid lines are best fitted to the data using the scaling law $T_N - T_c(H) \sim H^{2/\phi}$ with $\phi = 3.4 \pm 0.2$. T_N was obtained from ac susceptibility measurements at $H = 0$ (see Fig. 9) and coincides with the extrapolation of the $T_c(H)$ data to $H = 0$.

inset of Fig. 4, but plotted as $H^{2/\phi}$ versus T, with $\phi \cong 3.4$. This value of ϕ is in contrast with the standard $\phi \cong 1.4$ REIM–RFIM crossover behavior, found at weak applied fields in all measured samples of $Mn_xZn_{1-x}F_2$ for $x \geq 0.4$.[24,26] These results suggest a REIM–RFIM crossover breaking occurring somewhere in the interval $0.35 < x < 0.4$.

At low H the irreversible behavior ends far above $T_c(H)$ in $Mn_{0.35}Zn_{0.65}F_2$ (see Figs. 1 and 2) and $T_{eq}(H)$ is difficult to specify. More data concerning the irreversible behavior of the magnetization in $Mn_{0.35}Zn_{0.65}F_2$ can be obtained from field cycling measurements. In Fig. 5 we show the positive H part of M versus H cycles for $T = 4.8$ K. A similar plot is shown for $T = 12.0$ K in Fig. 6. For

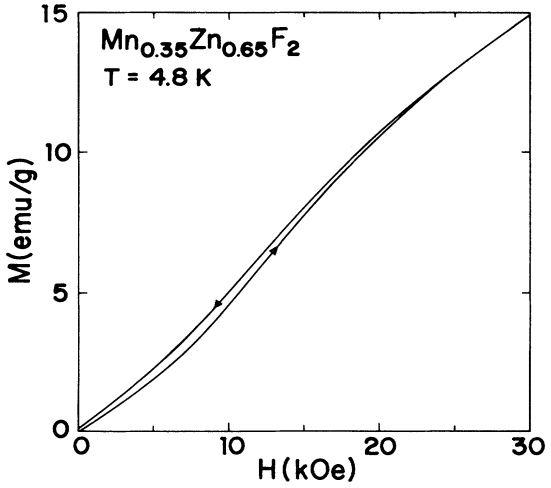

FIG. 5. The positive H part of M versus H cycles for $Mn_{0.35}Zn_{0.65}F_2$ at $T = 4.8$ K obtained after ZFC procedure.

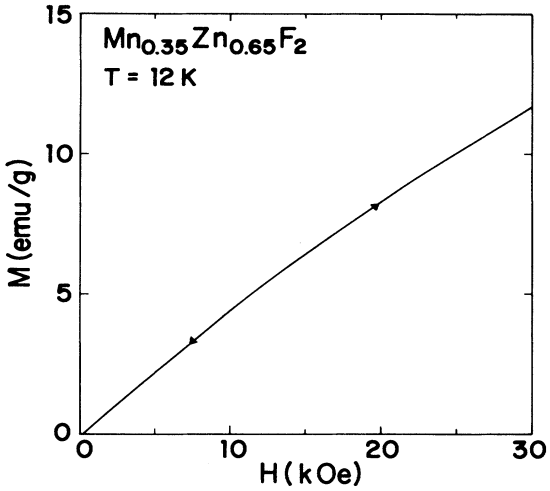

FIG. 6. The positive H part of M versus H cycle for $Mn_{0.35}Zn_{0.65}F_2$ at $T = 12$ K obtained after ZFC procedure.

$T < T_N$, an excess of magnetization $\Delta M_H \equiv M_{FD} - M_{FI}$, appears as the field is decreased (FD) from the paramagnetic phase, when compared with the field increasing (FI) data, as seen in Fig. 5. The hysteresis cycles shrink as T increases. In addition, for $T < T_N$ a remanent magnetization is left when the field is decreased to zero from the paramagnetic phase (see Fig. 5). Based on the presence of this remanent magnetization we conclude that if the system is led to an ordered AF configuration following ZFC, this state is not recovered by increasing H up to the paramagnetic phase and then decreasing it down to zero again, in the time scale of the experiment. As obvious, no hysteresis appears for field cycling performed at temperatures $T > T_N$ (see Fig. 6). However, a nonlinear magnetization $M_{NL}(H) \equiv \chi_0 H - M(H)$ appears above T_N. Here $\chi_0 \equiv (dM/dH)_{H \to 0}$ is the initial dc susceptibility measured along the easy axis. The temperature dependence of the inverse susceptibility $(M/H)^{-1}$ is plotted in Fig. 7 for two values of H at the two limits of the field range of present experiment. A salient feature of the curves in Fig. 7 is the fact that a departure from the Curie-Weiss law is observed below a "clustering temperature" $T_{cl} \cong 22$ K. The nonlinear susceptibility regime emerges for $T < T_{cl}$, where the M/H versus T curves become notably field dependent (see Fig. 1). This is the region where presumably the formation of short-range-ordered clusters initiate in $Mn_{0.35}Zn_{0.65}F_2$. There is a considerable temperature difference between T_{cl} and T_N, which has been not observed[26] before in samples with higher concentrations of Mn. However, the premature formation of clusters is a common feature[7,8] in spin-glass-like compounds. By extrapolating the linear part of $(M/H)^{-1}$ with the T axis, we obtain a value $\Theta \cong 30$ K for the mean-field Curie-Weiss parameter, so a ratio $\Theta/T_N \cong 2.8$ is obtained for this sample.

For large values of H, $Mn_xZn_{1-x}F_2$ comprises a low-temperature spin-flop phase, as already observed for samples with $x > 0.4$. Clear signatures of a first-order AF-SF transition line has been recently reported[26] by magnetiza-

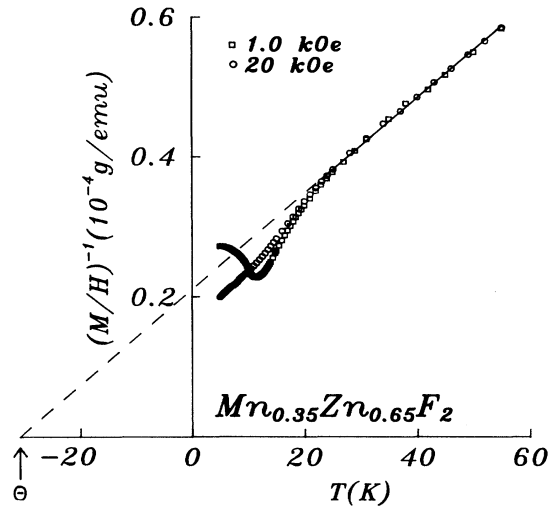

FIG. 7. Temperature dependence of the inverse dc susceptibility $(M/H)^{-1}$ of $Mn_{0.35}Zn_{0.65}F_2$ for two values of H obtained after ZFC procedure.

tion measurements in $Mn_{0.5}Zn_{0.5}F_2$. The spin-flop critical field H_c^{sf} can be determined by the position of the (dM/dH)peak. In Fig. 8 we show the field derivative (dM/dH) vs H for $T=4.8$ K in this strongly diluted $Mn_{0.35}Zn_{0.65}F_2$ sample. The broad maximum found is not a sufficient feature for an unambiguous determination of H_c^{sf}. For higher values of T the (dM/dH) peaks are even broader than the one shown in Fig. 8. Based on Figs. 1, 5, and 8, one could think in terms of a gradual transition from AF to paramagnetic phase in this sample, crossing a region of a mixed or intermediate phase composed of AF and SF clustering. However, there is also the possibility that the broadness of the dM/dH peaks is an artifact of concentration gradients. As H_c^{sf} depend on x, concentration gradients tend to blur out the AF-SF transition in a similar way to that found in the AF-P transition in diluted antiferromagnets.

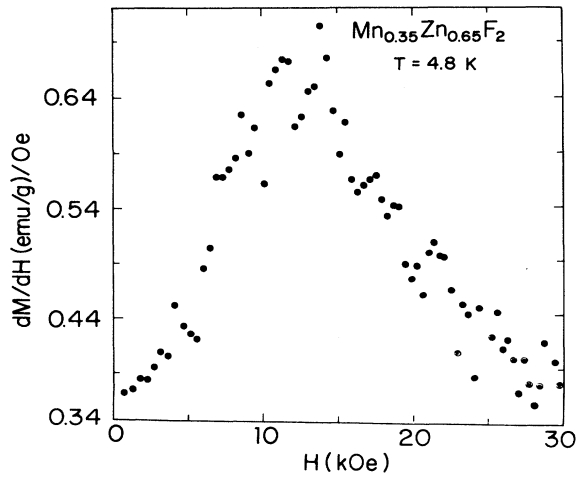

FIG. 8. dM/dH versus H measured on $Mn_{0.35}Zn_{0.65}F_2$ at $T=4.8$ K after ZFC procedure.

B. Zero-field ac susceptibility results

The low-temperature behavior of the in-phase component of the ac susceptibility χ_{ac} at $H=0$ is presented in Fig. 9 for three frequency values of the modulating field, applied parallel to [001]. The temperature of the χ_{ac} peaks determined from Fig. 9 do not shown noticeable frequency dependence and coincides with the Néel temperature $T_N=10.8\pm0.1$ determined from extrapolation of magnetization measurements at the same sample, as shown in Fig. 4. However, a strong frequency-dependent behavior of χ_{ac} appears at lower temperatures, indicating that frozen clusters coexist with AF ordering along the [001] direction. The absence of frequency-dependent behavior close to the peak temperature points in favor of a gradual predominance of the AF arrangement of spins as T increases, approaching T_N.

A comparison between ac susceptibility of $Mn_{0.35}Zn_{0.65}F_2$ for the modulating field positioned parallel $(\chi_\parallel)$ and perpendicular $(\chi_\perp)$ to [001] is shown in Fig. 10 for a frequency value of 1733 Hz. The curves depart one from the other below $T_N\cong10$ K. Similar behavior has been observed for other values of frequency of the modulating field. In $Mn_xZn_{1-x}F_2$, the dipolar anisotropy is expected to become random in direction as x decreases. However, data from Fig. 10 show that at least down to $x=0.35$ the dipolar anisotropy is still effective to induce a preferential alignment of spins parallel to [001]. The freezing process occurs only along this longitudinal direction, as shown in Fig. 9. When the field is perpendicular to [001], data from all curves collapse at the same $\chi_\perp$ curve, shown in Fig. 10. In Fig. 11, $\chi_\perp$ is plotted against $1/T$, for a frequency value of the modulating field of 1733 Hz. $\chi_\perp$ data departs from the Curie-Weiss law for $T\leq8$ K, approximately the same temperature below which the freezing process arises in $\chi_\parallel$ (see Fig. 9).

The effect of frozen clusters in this sample is also noted

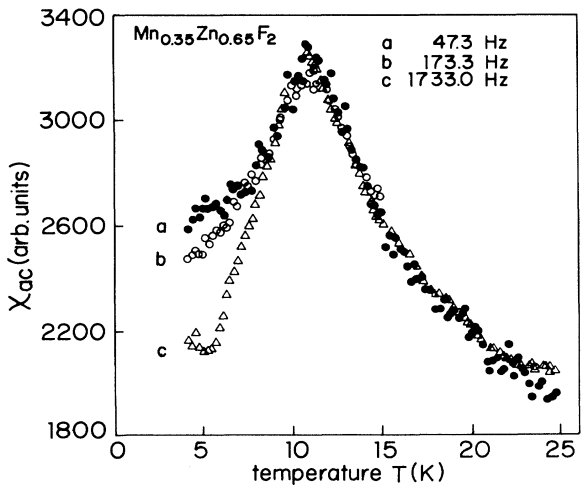

FIG. 9. Temperature dependence of the in-phase component of the parallel ac susceptibility $(\chi_\parallel)$ measured on $Mn_{0.35}Zn_{0.65}F_2$ at $H=0$, for $f=47.3$ Hz (curve a), 173.3 Hz (curve b), and 1733.0 Hz (curve c).

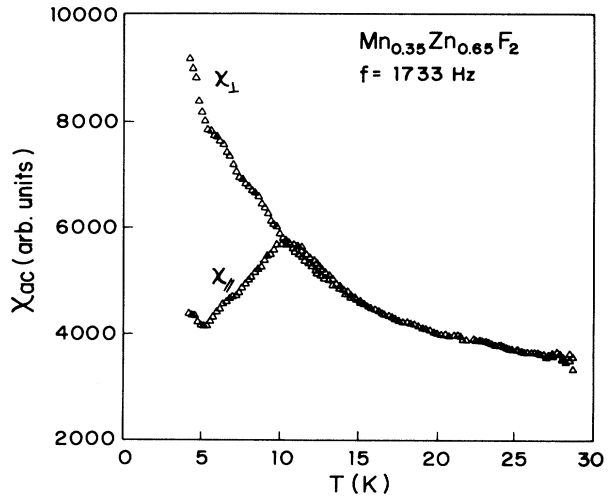

FIG. 10. Temperature dependence of the ac susceptibility measured on $Mn_{0.35}Zn_{0.65}F_2$ at $H=0$ for a driven field of 1.0 Oe applied parallel ($\chi_\parallel$) to the easy direction [001] and perpendicular ($\chi_\perp$) to it, using a frequency $f=177.0$ Hz.

in the low-field dc magnetization. The excess of magnetization $\Delta M_H = M_{FC} - M_{ZFC}$, shown in Figs. 1 and 2, reveals a striking reversal in its field dependence as H decreases below ≈ 2 kOe, when ΔM_H starts to increase as H decreases. This low-field behavior is not to be expected[40–42] to occur in a nonfrustrated diluted antiferromagnet under a uniform field H, where a monotonic increasing in ΔM_H is predicted as H increases [see Eq. (5)].

It is worthwhile to mention that although some spin-glass-like features have been detected in the magnetic behavior of $Mn_{0.35}Zn_{0.65}F_2$, they are not sufficient to characterize a true spin-glass phase in this sample. First of all, the stability of the AF LRO needs to be studied under a field in the (H,T) diagram. Moreover, static[8] and dynamic[9] scaling analysis of the critical behavior are necessary to ultimately characterize the nature of the cooperative behavior which takes place close to T_N in this system.

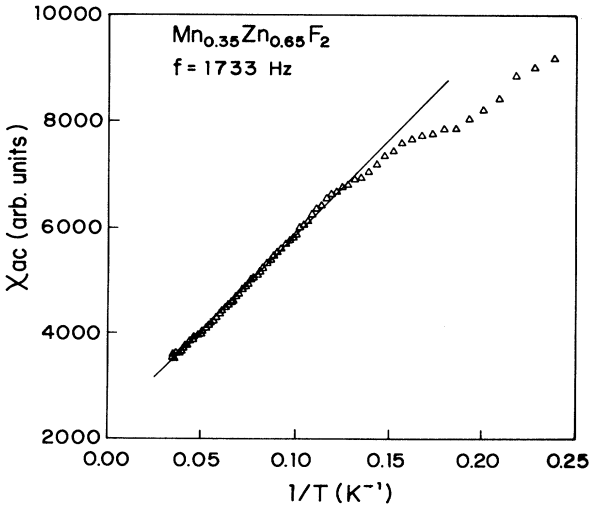

FIG. 11. $\chi_\perp$ versus $1/T$ measured on $Mn_{0.35}Zn_{0.65}F_2$ at $H=0$ for a driven field of 1.0 Oe applied perpendicular to the easy direction [001], using a frequency $f=1733.0$ Hz.

V. INTERPRETATION

Using dc magnetization and ac susceptibility techniques we have presented features on the magnetic behavior of the dilute antiferromagnet $Mn_xZn_{1-x}F_2$, for $x_p < x < 0.4$. The most intriguing feature here is the fact that the crossover exponent for the *critical* phase boundary assumes a value $\phi \cong 3.4$, in a striking departure from the standard REIM–RFIM crossover behavior value $\phi \cong 1.4$, found[25,26] at higher concentrations of Mn. The value of ϕ also suffer an identical change when the percolation threshold is approached in $Fe_xZn_{1-x}F_2$. However, a distinction is to be made between the magnetic behavior of these compounds in this strong dilution regime. The value $\phi = 3.4$ is obtained[23] from the curvature of the *equilibrium* phase boundary in $Fe_{0.31}Zn_{0.69}F_2$, when a glassy phase starts to nucleate in place of the original AF ordering. This spin-glass-like behavior evolves to be the sole feature in the close vicinity of x_p when a pure Ising spin-glass behavior takes place, in the sense that all thermodynamic and critical properties of canonical spin glasses are observed.[7–9] The analysis of the stability of the long-range AF ground state achieved by ZFC procedure is decisive to establish the crossover from AF to the spin-glass-like state in $Fe_xZn_{1-x}F_2$. In the latter regime the AF configuration is no longer the minimum in the free energy of the system, as becomes clear from the observed time dependence in the ZFC staggered magnetization[10] as well as in the uniform magnetization,[23] when the system is led to the glassy phase from the AF state. In $Mn_{0.35}Zn_{0.65}F_2$ the ZFC magnetization did not show any detectable time dependence in the H and T ranges of the present experiment. However, the results from zero-field ac susceptibility measurements presented here for the same sample indicates that a spin-glass-like clustering is present at low temperatures. The presence of a time-varying spin-glass state in $Mn_xZn_{1-x}F_2$ for a $0.2 \leq x \leq 0.33$ at low temperatures has been reported[27] by Bazhan and Petrov. The influence of these frozen SG clusters on the stability of the AF LRO in $Mn_xZn_{1-x}F_2$ for $x \rightarrow x_p$ is not well known. However, this fact and the excessive rounding[52] of $d(M/H)/dT$ peaks shown in Fig. 3 make uncertain the existence and nature of the equilibrium phase transition at $T_c(H)$ for strong diluted samples of $Mn_xZn_{1-x}F_2$. If AF LRO is unstable, $T_c(H)$ as shown in Fig. 4, represents only a virtual line of destroyed (rounded) phase transitions.

The origin of the freezing mechanism of $\chi_\parallel$ in $Mn_{0.35}Zn_{0.65}F_2$ at zero field is not well understood. One possibility comes from the magnetic-dipole interaction $J_{ij} = \mu_i\mu_j(1-3\cos^2\theta_{ij})/r^3$. Even though the spin correlations are mainly restricted to the [001] direction (see Fig. 10), the dipolar interaction can be either ferromagnetic or antiferromagnetic, depending on the relative orientations of each dipole. This competition, together with the effect

of strong dilution weakening the exchange interactions, can lead the system to a spin-glass-like behavior.[53–55] Another possible mechanism is the eventual presence of frustration in the exchange interactions of $Mn_{0.35}Zn_{0.65}F_2$. The pure MnF_2 compound presents a small frustration[4] in the next-nearest neighbor interaction J_3. Recent results from mean-field simulations at $H=0$ reveal[56] that a weak frustration plays no effect in the REIM properties of a dilute Ising antiferromagnet for weak dilution ($0.6<x<1$). However, in the strong dilution regime ($x<0.5$) it plays a dramatic role both on reducing the magnitude of the antiferromagnetic order parameter and inducing the appearance of a nonzero spin-glass order parameter close to the percolation concentration. Finally, the low-T behavior of $\chi_{\parallel}$ at $H=0$ could be also related to the extremely slow dynamics[57] of Ising spins close to the percolation threshold, where the surface of an incipient infinite cluster behaves like a ramified fractal lattice.

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