Dynamics of the antiferromagnetic random-exchange Ising system $Fe_xZn_{1-x}F_2$ near the percolation threshold

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Dynamic magnetic properties of the diluted Ising antiferromagnet, $Fe_{0.25}Zn_{0.75}F_2$, have been investigated by dc-magnetic relaxation and ac-susceptibility measurements. In similarity with spin glasses, a strongly frequency-dependent cusp in the dynamic susceptibility appears at low temperature. The corresponding slowing down of the relaxation times is however found to follow an Arrhenius law, i.e., critical dynamics indicating a phase transition is not observed. Also, there are no indications of an equilibrium Almeida-Thouless line in the H-T plane and the field dependence of the equilibrium susceptibility behaves noncritically. However, an ageing behavior, a common feature of frustrated disordered magnetic systems, is observed at low temperatures.
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I. INTRODUCTION

A para- to antiferromagnetic phase transition at a finite temperature, T_N, occurs in diluted $3d$ antiferromagnets with a concentration of magnetic ions larger than the percolation threshold x_p. For the tetragonal rutile Ising system $Fe_xZn_{1-x}F_2$ the percolation threshold is $x_p=0.25$.[1] The physical properties of $Fe_xZn_{1-x}F_2$ have been intensely investigated primarily in large applied magnetic fields to study the random-field problem[2,3] but also zero-field random exchange Ising properties have been comprehensively studied.[3] Investigations of samples near the percolation limit have shown expected and dramatic differences from the behavior of samples of slightly higher concentration. The peculiar spin-glass-like dynamics at the percolation concentration was discovered in early ac-susceptibility studies[4] and numerical simulations.[5] The system was then suggested to be a $3d$ Ising spin glass with a finite spin-glass temperature, T_g. Neutron-scattering results[6] have, on the other hand, been interpreted just to manifest a theoretically predicted slowing down of the dynamics at low temperature.[7] A magnetic system close to the percolation threshold is expected to be unstable with respect to small frustrating interactions and form a spin glass.[8] In this paper we discuss results from dc-magnetization and ac-susceptibility measurements on $Fe_{0.25}Zn_{0.75}F_2$. Dynamic scaling and a noncritical field dependence of the susceptibility firmly establishes that a spin-glass phase transition is not anticipated at any temperature in this system. However, an ageing behavior, characteristic of the logarithmically slow dynamics of disordered and frustrated magnetic systems such as spin glasses, is observed at low temperatures.

II. EXPERIMENTAL

The sample, a single crystal of $Fe_{0.25}Zn_{0.75}F_2$, was cut into the form of a parallelepiped of size $1.75\times1.9\times3.6$ mm^3 with its longest axis aligned with the crystalline c axis. The dynamic properties of the sample were studied by means of low-field time-dependent zero-field-cooled (ZFC) magnetization measurements, utilizing a noncommercial superconducting quantum interference device (SQUID) magnetometer, and by ac-susceptibility measurements in a Lake Shore 7225 ac susceptometer. The temperature dependence of the magnetization at different applied dc fields was measured in a Quantum Design MPMS5 SQUID magnetometer. The magnetic field was applied parallel to the c axis of the sample when not stated otherwise.

III. RESULTS AND DISCUSSION

A. Zero-field equilibrium dynamics

The low-field dynamic susceptibility is plotted vs temperature at different observation times in Fig. 1. The short-time data are measured by ac-susceptibility measurements in an ac field of 3 Oe at 15, 125, 10^3, and 10^4 Hz, the observation time is then given by $t\approx1/\omega$, where ω is the angular frequency. Figure 1(a) shows the in-phase component, $\chi'(\omega,T)$ and Fig. 1(b) the corresponding out-of-phase component, $\chi''(\omega,T)$. The data at longer times in Fig. 1(a) are obtained from measurements of the relaxation of the zero-field-cooled (ZFC) magnetization, $m_{\mathrm{ZFC}}(t)$, at different temperatures. In this experimental procedure the sample is cooled in zero field from a high temperature $T_{\mathrm{ref}}=21$ K to the measurement temperature T_m. After a wait time, $t_w=100$ s, a small field $h=3$ Oe is applied and the relaxation of the magnetization, $m_{\mathrm{ZFC}}(t)$, is recorded in the time range $0.3<t<10^4$ s after the application of the magnetic field.

As is seen in Fig. 1(a), there is a strongly frequency-dependent maximum in the susceptibility [accompanied by the appearance of an out-of-phase component of the ac susceptibility, Fig. 1(b)]. The position of the maximum in $\chi(T,t)$ can be used to define a spin-freezing temperature T_f at the corresponding observation time, which also defines

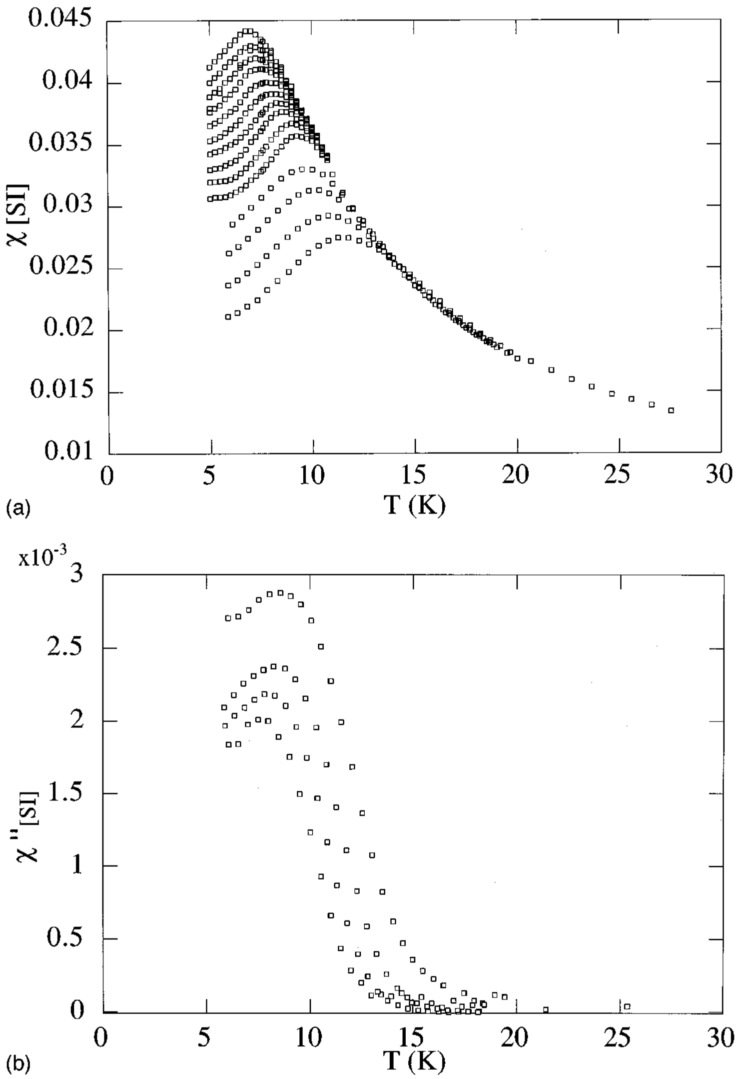

FIG. 1. Temperature dependence of the ac-susceptibility at different observation times, $t \approx 1/\omega$, in zero dc field. The short-time data are from ac-susceptibility measurements in an ac field of 3 Oe at 15, 125, 10^3, and 10^4 Hz and the longer times are from time-dependent ZFC measurements in a field of 3 Oe. (a) the in-phase component χ' at observation times (bottom to top) 1.6×10^{-5}, 1.6×10^{-4}, 1.3×10^{-3}, 0.01, 0.3, 1, 3, 10, 30, 100, 300, 10^3, 3×10^3, and 10^4 s. (b) the out-of-phase component χ'' at observation times (top to bottom) 1.6×10^{-5}, 1.6×10^{-4}, 1.3×10^{-3}, and 0.01 s.

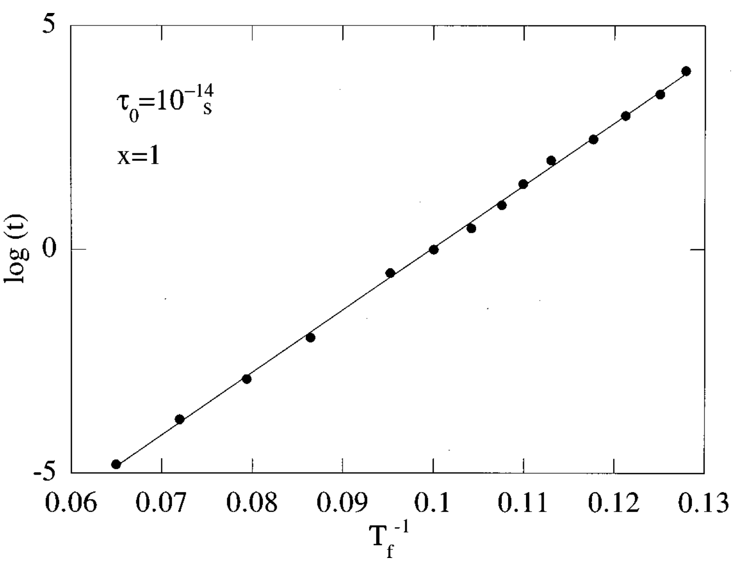

FIG. 2. The best fit of the relaxation times to activated dynamics: $\log t$ vs T_f^{-1}. Implying a pure Arrhenius behavior of the slowing down of the dynamics.

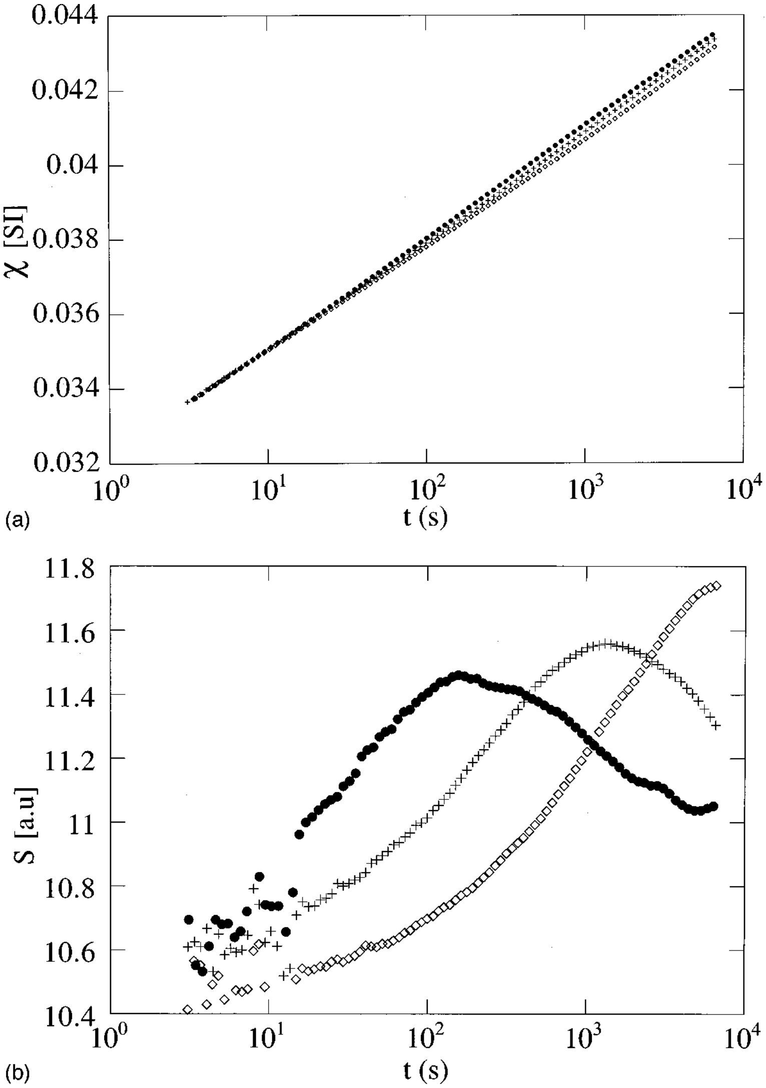

FIG. 3. $\chi = m_{\mathrm{ZFC}}(t)/h$ vs $\log t$ (a), and relaxation rate $S = 1/h\ dm/d\ \log t$ vs $\log t$ (b), for $t_w = 100$ (●), 1000 (+), 10 000 (◇) s, at $T_m = 6$ K and $h = 3$ Oe.

the maximum relaxation time $\tau_{\max}$ of the system at that temperature. A time window ranging over nine decades from 10^{-5} to 10^4 s is covered by our data. To characterize the slowing down of the relaxation times of the system with temperature we use pairs of T_f and $\tau_{\max}$ deduced from Fig. 1(a). A $3d$ spin glass should exhibit conventional critical slowing down according to[9]

$$\frac{t}{\tau_0} \propto \left(\frac{T_f - T_g}{T_g} \right)^{-z\nu}, \tag{1}$$

where τ_0 is a microscopic relaxation time of order 10^{-13} s, T_g is the spin-glass temperature, and $z\nu$ is a dynamic critical exponent. It is not possible to fit our data to this expression with reasonable values of the parameters involved. Alternatively, activated dynamics could govern the dynamics of the system, yielding a slowing down of the relaxation times according to

$$\ln\left(\frac{t}{\tau_0} \right) \propto \frac{1}{T_f} \left(\frac{T_f - T_g}{T_g} \right)^{-\psi\nu}, \tag{2}$$

where $\psi\nu$ is a critical exponent.[10] Fitting the data to this equation suggets $T_g \approx 0$, which implies that the slowing down is described by only a generalized Arrhenius law:

$$\log\left(\frac{t}{\tau_0} \right) \propto T_f^{-x}. \tag{3}$$

Figure 2 shows the best fit to this expression, which yields $\log t$ vs T_f^{-1}, i.e., $x = 1$, and $\tau_0 = 10^{-14}$ s. In other words the slowing down of the relaxation time obeys a pure Arrhenius behavior, implying that thermal activation over constant barrier heights governs the relaxational behavior, i.e., there is no indication of a critical behavior and a spin-glass phase transition is neither anticipated at a finite nor at zero temperature.

It is worth mentioning that the tendency of the ZFC susceptibility curves in Fig. 1(a) to flatten out, and even turn upward at lower temperatures (see below, inset of Fig. 5), indicates that some of the Fe ions behave as being isolated spins showing a Curie-like behavior at low temperatures. A similar behavior has been observed in some dilute semimagnetic semiconductors, such as $Hg_{0.7}Mn_{0.3}Te$, at low temperatures. These systems, on the other hand, exhibit characteristic $3d$ spin-glass transitions and a critical slowing down at a finite temperature.[11]

B. Ageing

One property that reveals correlated dynamics and frustration in a slowly relaxing system at low temperature is the

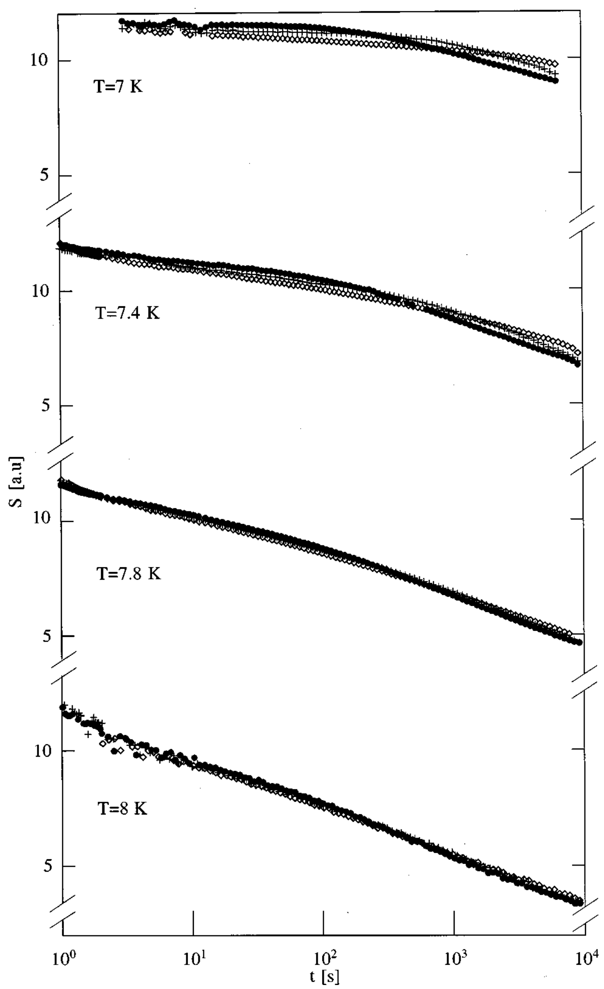

FIG. 4. Relaxation rate $S=1/h\ dm/d \log t$ vs $\log t$, for $t_w = 100$ (●), 1000 (+), 10 000 (◇) s at the different measurement temperatures (top to bottom) $T_m = 7$, 7.4, 7.8, and 8 K.

existence of magnetic ageing. Magnetic ageing implies that the system at constant temperature (and constant field) is subjected to a continuous rearrangement process of the spin structure continuing to time scales of the order of the maximum relaxation time of the system. The ageing phenomenon has been amply studied in $3d$ spin glasses,[12] but is also an intrinsic property of $2d$ spin glasses[13] (where there is no phase transition at finite temperature), and other disordered and frustrated magnetic systems.[14] To reveal a possible ageing phenomenon of the low temperature slowly relaxing ''phase'' we performed time-dependent ZFC magnetization measurements. The sample was cooled from the reference temperature $T_{ref}=21$ K to the measurement temperature (T_m) and kept there a wait time (t_w). Thereafter a weak field (h) was applied and the magnetization (m) was recorded as a function of time. In Fig. 3(a), m is plotted vs $\log t$ at $T_m = 6$ K, $h=3$ Oe, and $t_w = 100$, 1000, and 10 000 s. The corresponding relaxation rate ($S=1/h dm/d \log t$) is plotted in Fig. 3(b). A wait time dependence of the response is observed. The m vs $\log t$ curves have an inflection point at an observation time nearly equal to t_w and the relaxation rate attains a corresponding maximum. It is worth mentioning that the influence of ageing on the magnetic relaxation of $Fe_{0.25}Zn_{0.75}F_2$ is much weaker than in a $3d$ spin glass[15] and also a lot weaker than at a corresponding temperature in a $2d$ spin glass.[13]

In Fig. 4, S vs $\log t$ is plotted for different t_w at four different measurement temperatures, $T_m = 7$, 7.4, 7.8, and 8 K. A strong temperature dependence is observed. The wait time dependence of the magnetic relaxation decreases rapidly with increasing temperature to practically vanish at $T_m = 8$ K. The maximum relaxation time of the system decreases with increasing temperature and has at 8 K reached the order of 10^4 to 10^5 s. When the minimum effective ageing time (time at constant temperature+cooling rate time scale) becomes of the order of the maximum relaxation time of the system at the measurement temperature (see also Fig. 1), observable ageing disappears. A similar fading away of observable ageing when τ_{max} approaches the minimum experimental effective wait time is also seen in $2d$ spin glasses[13] and in $3d$ spin glasses at temperatures above T_g.[15]

C. Temperature and field dependence of the magnetization

Figure 5 shows the temperature dependence of the field-cooled (FC), $M_{FC}(T)/H$, and zero-field-cooled (ZFC),

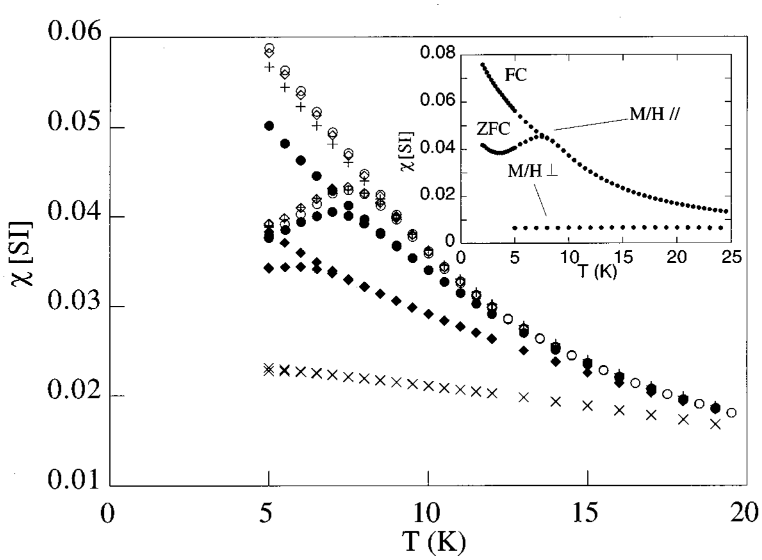

FIG. 5. Temperature dependence of the dc susceptibility for zero-field-cooled (ZFC) and field-cooled (FC) procedures in different fields, 10^{-3} T (○), 10^{-2} T (◇), 0.1 T (+), 0.3 T (●), 1 T (♦), and 3 T (×) vs temperature. The inset shows the result for $H=10^{-2}$ T (100 Oe) parallel and perpendicular to the easy axes.

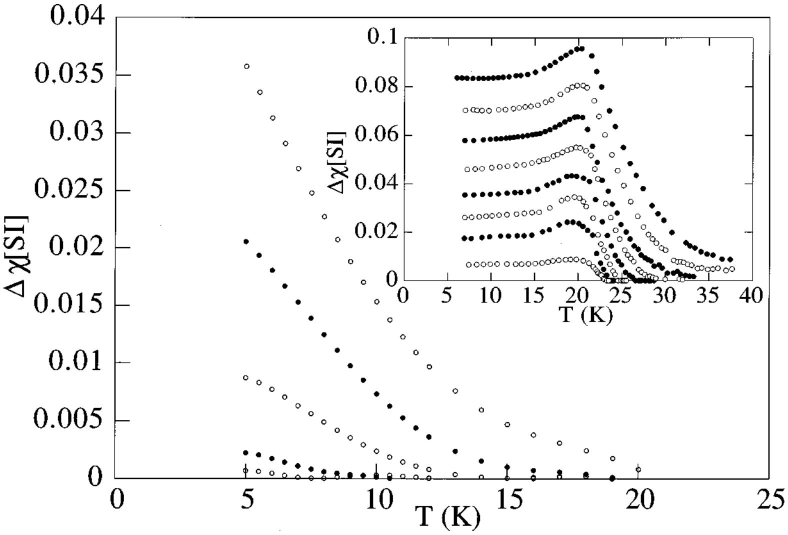

FIG. 6. $\Delta\chi = [M_{FC}(H\to 0) - M_{FC}(H)]/H$ vs temperature for different fields (top to bottom) $H = 3$, 1, 0.3, 0.1, and 0.01 T. In the inset the corresponding quantity for the $3d$ Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$ at the fields (top to bottom) $H = 1.28$, 0.64, 0.32, 0.16, 0.08, 0.04, 0.02, and 0.004 T is shown. Data from Ref. 17.

$M_{ZFC}(T)/H$, susceptibility[16] at different applied fields. The general behavior is somewhat reminiscent of the field dependence of the susceptibility of a spin glass. The inset includes a plot of the susceptibility when the field is applied perpendicular to the c axes. The perpendicular susceptibility ($H = 10^{-2}$ T) shows an almost temperature-independent behavior and a weak irreversibility at low temperature (not resolved in the figure) that can be assigned to a slight misalignment of the sample yielding a small contribution from the c-axis susceptibility.

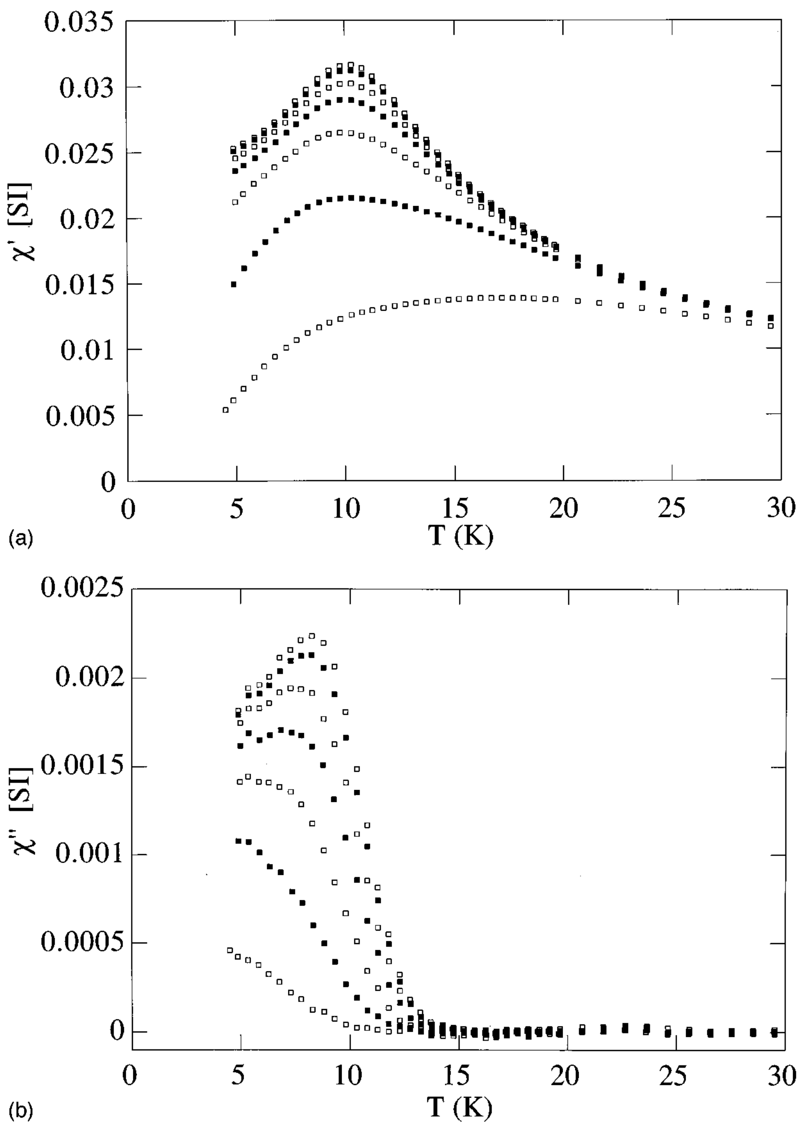

FIG. 7. The ac susceptibility $\chi'(\omega)$ (a) and $\chi''(\omega)$ (b) at frequency $\omega/2\pi = 125$ Hz with amplitude $h_{ac} = 3$ Oe and different superposed dc fields: (top to bottom) 0, 0.1, 0.2, 0.3, 0.5, 1, and 3 T.

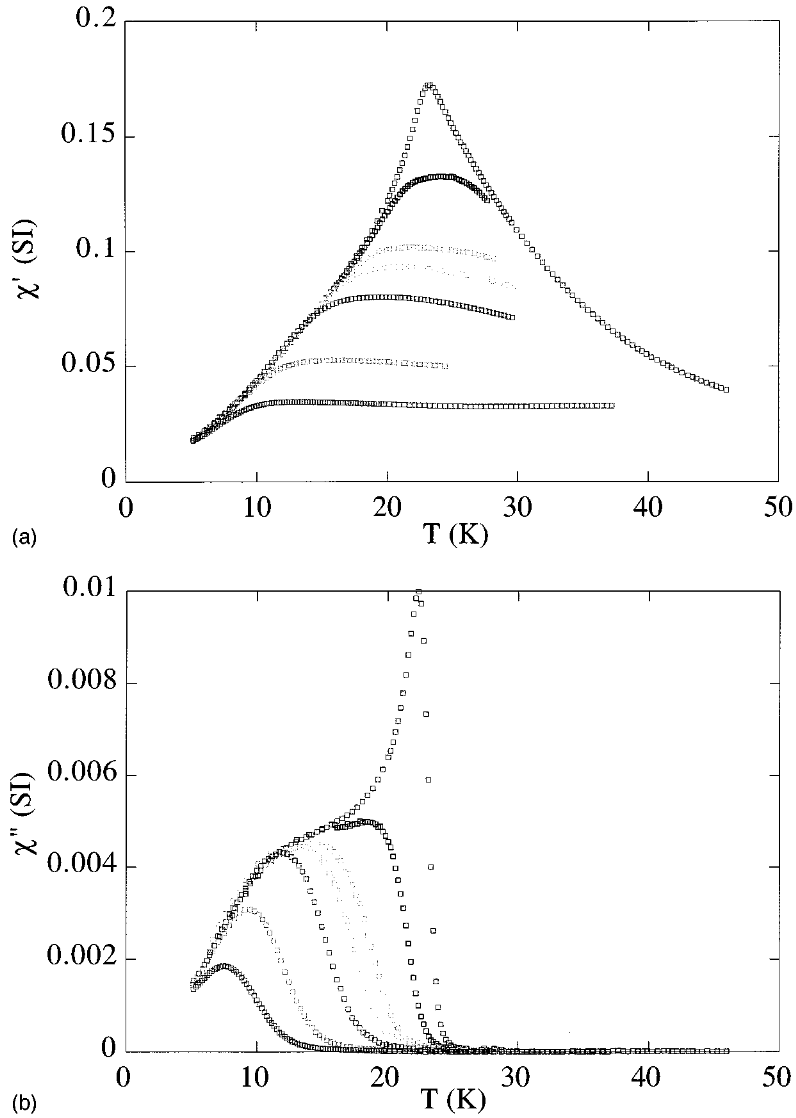

FIG. 8. The ac susceptibility $\chi'(\omega)$ (a) and $\chi''(\omega)$ (b) of $Fe_{0.5}Mn_{0.5}TiO_3$ at frequency $\omega/2\pi = 125$ Hz and different superposed dc fields: (top to bottom) 0, 0.1, 0.4, 0.6, 1, 2, and 3 T. Data from Ref. 20.

There is a significant field dependence of the FC susceptibility. In Fig. 6 the quantity $\Delta\chi = [M_{\mathrm{FC}}(H\rightarrow 0) - M_{\mathrm{FC}}(H)]/H$ is plotted vs temperature. This quantity reflects the field and temperature dependence of the nonlinear susceptibility. As can be seen from the figure, there is a continuous increase of $\Delta\chi(H,T)$ with decreasing temperature. There is no indication of a critical divergence in $\Delta\chi(H,T)$ in the measured temperature range. For comparison and contrast we have in the inset plotted $\Delta\chi$ vs T as derived from measurements on the model $3d$ Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$, (data from Ref. 17). This sample has a finite $T_g \approx 20.5$ K, and thus also an inherent critical divergence in the nonlinear susceptibility at T_g, here seen as a maximum in $\Delta\chi$ at $T \approx T_g(0)$ at all fields.

At temperatures below the maximum in the ZFC susceptibility and at low and intermediate fields χ_{ZFC} becomes somewhat enhanced by an increasing field, which is also a characteristic of spin glasses.[17] This behavior is associated with an S-shaped M_{ZFC} vs H curve at low temperatures. It is also worth noting that at lower fields ($H \leqslant 0.1$ T), the measured thermoremanent magnetization M_{TRM},[18] adequately fulfills the relation: $M_{\mathrm{TRM}}(T,H) \approx M_{\mathrm{FC}}(T,H) - M_{\mathrm{ZFC}}(T,H)$.

D. Almeida-Thouless line or just dynamics?

The temperature where the $M_{\mathrm{FC}}(T)$ and $M_{\mathrm{ZFC}}(T)$ curves merge can be used to define an irreversibility temperature (T_{ir}). The irreversibility temperature remains closely constant at lower fields $H \leqslant 0.1$ T. The effect of higher magnetic fields is to shift the irreversibility temperature to lower temperatures. The irreversibility signals that the sample falls out of equilibrium and is thus directly related to the increase of the maximum relaxation time of the system with decreasing temperature. The time scale for $T_{\mathrm{ir}}(H)$ is related to the heating rate when measuring $M_{\mathrm{FC}}(T)$. The position of this line in an H-T diagram (see below Fig. 9) is governed by the observation time of the experiment. In spin-glass literature, empirical irreversibility lines are sometimes interpreted to mirror the mean-field derived in-field spin-glass phase-transition line: the Almeida-Thouless line (AT line).[19]

The dynamic irreversibility lines discussed above can be measured on well-defined time scales by ac-susceptibility experiments in different superposed dc fields. In Figs. 7(a) and 7(b) $\chi'(\omega)$ and $\chi''(\omega)$ are plotted vs temperature in different superposed dc fields. In the measurement $\omega/2\pi = 125$ Hz and $h_{\mathrm{ac}} = 3$ Oe. Corresponding curves for the $3d$ Ising spin glass

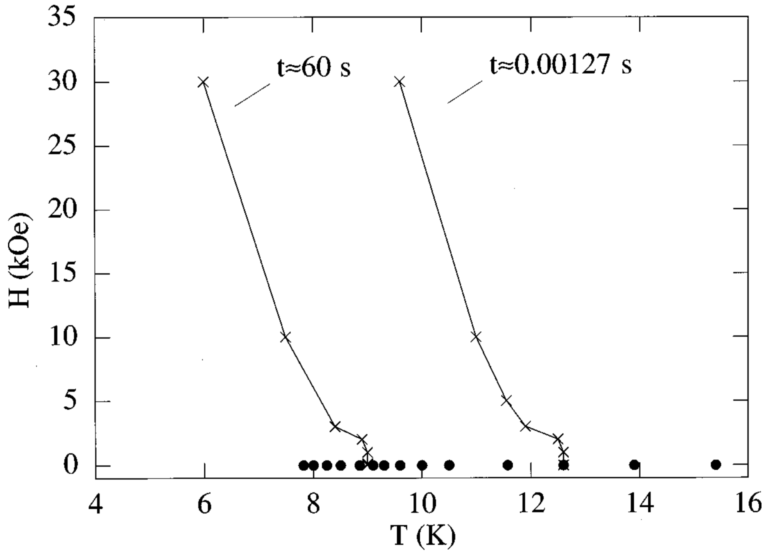

FIG. 9. H-T diagram of freezing temperatures at different observation times. The continuous left line represents the irreversibility temperature deduced from the FC-ZFC measurement of Fig. 5 and corresponds to an observation time $t \approx 60$ s. The line to the right represents the temperature where the out-of-phase component becomes nonzero in the ac-susceptibility measurement [from Fig. 7(b)] at the observation time $t \approx 0.0013$ s. Also plotted are the freezing temperatures at $H = 0$ (●) for different observation times (from left to right): 10 000, 3000, 1000, 300, 100, 30, 10, 3, 1, 0.3, 0.01, 1.3×10^{-3}, 1.6×10^{-4}, and 1.6×10^{-5} s.

$Fe_{0.5}Mn_{0.5}TiO_3$ are shown in Fig. 8 (from Ref. 20). There are significant differences between the effect that the superposed dc field has on the susceptibility of the two samples. In $Fe_{0.5}Mn_{0.5}TiO_3$, the applied field does not affect $\chi'(T,\omega,H)$ or $\chi''(T,\omega,H)$ at low temperatures, at higher temperatures where $\chi''(T,\omega,H)=0$; $\chi'(T,\omega,H)$ becomes flattened, strongly field-dependent, and equal to the equilibrium susceptibility $\chi'(T,0,H)$ in the same dc field. A similar behavior is characteristic for other $3d$ spin glasses.[21] $Fe_{0.25}Zn_{0.75}F_2$ shows a strikingly different behavior in larger superposed dc fields, as is seen from Fig. 7. $\chi'(T,\omega,H)$ is strongly suppressed by the dc field at all temperatures, there is no tendency for the curves at different dc fields to merge at low temperatures. However, the appearance of, and the maximum in $\chi''(T,\omega,H)$ vs T are shifted towards lower temperatures with increasing field, in an apparent similarity with spin-glass behavior.

The appearance of the out-of-phase component can be used to define the frequency and field-dependent freezing temperature, $T_f(\omega,H)$. At this temperature the maximum relaxation time of the system has just reached the order of the observation time, $\tau_{max} \approx 1/\omega$. In Fig. 9 we have plotted $T_f(\omega,H)$ at 125 Hz (or $t \approx 0.0013$ s) in an H-T diagram together with the above discussed irreversibility temperature; at $H=0$ we have also included the data from magnetic relaxation measurements discussed in Sec. III A. As can be seen from the figure, the effect of an applied field is to push the constant relaxation time contours at two significantly different observation times towards lower temperature. There is no visual crossover of the dynamic behavior in zero compared to a finite field, i.e., the slowing down of the relaxation time should obey an Arrhenius law also in a magnetic field.

The AT line scales with field as $T_g(H) - T_g(0) \propto H^{2/\Phi}$, where $\Phi = 3$ is the mean-field exponent.[19] The frequency-dependent freezing lines that we have deduced above do not converge toward an AT line. It is in this context also worth mentioning that there is experimental[20] and theoretical[10] work that convincingly suggest that there is no AT line and thus no finite-temperature phase transition in a magnetic field in $3d$ Ising spin glasses.

IV. CONCLUSIONS

Static and dynamic magnetic properties of the diluted antiferromagnet $Fe_{0.25}Zn_{0.75}F_2$ have been studied. The dynamic susceptibility shows some apparent similarities to spin-glass behavior, but the slowing down of the dynamics at low temperatures is not critical and well described by a pure Arrhenius law. Hence, there is no spin-glass phase transition at any temperature. In a magnetic field the maximum relaxation time of the system at a specific temperature is shortened but the slowing down of the dynamics remains Arrhenius like. In addition to strong disorder there is also weak frustration leading to an ageing phenomenon at low temperatures.

ACKNOWLEDGMENTS

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