Random-field and glassy dynamics in a diluted Ising antiferromagnet: $Fe_{0.42}Zn_{0.58}F_2$

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Random-field and glassy dynamics in a diluted Ising antiferromagnet: $\mathrm{Fe_{0.42}Zn_{0.58}F_2}$

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Abstract. – The random-field Ising model (RFIM) system $\mathrm{Fe_{0.42}Zn_{0.58}F_2}$ is studied by magnetization and dynamic susceptibility measurements, under finite dc applied fields (H). For $H < 20\,\mathrm{kOe}$, the magnetic behaviour is compatible with a long-range ordered (LRO) antiferromagnetic (AF) ground state, which undergoes a phase transition (PT) at a critical temperature $T_\mathrm{c}(H)$. For higher H, the LRO configuration becomes unstable and the PT is destroyed. A glassy dynamics emerges in the upper part of the (H, T) phase diagram. Our results reconcile earlier concepts associated with the weak RFIM problem with recent experimental data in $\mathrm{Fe}_x\mathrm{Zn}_{1-x}\mathrm{F}_2$.

There is now theoretical and experimental consensus that the ground state of the random field Ising model (RFIM) problem has long-range order (LRO) in $d = 3$ [1, 2]. This settles earlier controversies concerning the lower critical dimension (d_l) of this problem. However, the nature of the phase transition (PT) and the non-equilibrium behaviour of $d = 3$ RFIM systems are still a subject of intense debate [2–6]. Random-field models have been applied to a wide variety of physical systems, such as type-II superconductors, two-component fluids in random media, magnetic multilayers, etc., leading to a general interest in this problem. As first pointed out by Fishman and Aharony [7], a uniaxial diluted antiferromagnet (DAF) under a weak uniform field (H) applied parallel to its easy direction presents the critical behaviour of the RFIM. The magnitude of the local random field h was shown to be proportional to H, and h increases under larger dilution [8]. The uniaxial DAF $\mathrm{Fe}_x\mathrm{Zn}_{1-x}\mathrm{F}_2$ under an external magnetic field is probably the most representative experimental realization of the RFIM in $d = 3$. Above the percolation threshold (x_p) and in the absence of exchange frustration, the condensed phase of a $d = 3$ DAF is expected to exhibit a ground state with LRO at $H = 0$. The PT as well as the critical dynamics near the Néel temperature (T_N) in $\mathrm{Fe}_x\mathrm{Zn}_{1-x}\mathrm{F}_2$ are observed experimentally to be well described by the random-exchange Ising model (REIM) universality class, for all measured samples with $x > 0.30$ [9–11]. If the sample is zero-field cooled (ZFC) from the

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paramagnetic (P) phase through T_N, and then a *weak* H is applied, LRO is maintained below a critical temperature $T_\mathrm{c}(H)$. Earlier experimental data, obtained from birefringence, neutron scattering, Faraday rotation, ac susceptibility and capacitance techniques [12–16], support a PT taking place at $T_\mathrm{c}(H)$. The PT is accompanied by an extreme critical slowing-down inherent in the RFIM problem [14, 15]. In RFIM systems, hysteresis is commonly observed between ZFC and field-cooled (FC) data, below an equilibrium temperature $T_\mathrm{eq}(H)$. For low H, $T_\mathrm{eq}(H)$ lies just above $T_\mathrm{c}(H)$ [16]. In the FC procedure finite-size metastable clusters are frozen just below $T_\mathrm{eq}(H)$.

New interpretations concerning the nature of the phase transition and also the dynamics of a diluted antiferromagnet in a field (DAFF) have appeared, based on experiments performed in samples of $\mathrm{Fe}_x\mathrm{Zn}_{1-x}\mathrm{F}_2$, with $x \sim 0.50$. Birgeneau *et al.* [2] explored the staggered magnetization (M_s) of $\mathrm{Fe}_{0.50}\mathrm{Zn}_{0.50}\mathrm{F}_2$ by magnetic X-ray and neutron scattering, and its relation to the uniform magnetization (M) and magnetic specific heat (C_m) to conclude that the peaks observed [13, 14] at $T_\mathrm{c}(H)$ in C_m and $(\mathrm{d}M/\mathrm{d}T)_H$ reflect only the effects of a non-equilibrium ZFC LRO. This scenario was based primarily on the fact that the intensity of the ZFC Bragg peaks goes to zero with increasing field at progressively lower temperatures in an increasingly rounded fashion. This rounding was observed [2] to increase as $\sim H^2$. On the other hand, Binek *et al.* [6] observed that the ZFC peak found at low H in the real part of the parallel ac susceptibility (χ'_ac) in $\mathrm{Fe}_{0.47}\mathrm{Zn}_{0.53}\mathrm{F}_2$ splits, for higher values of H, into a narrow peak at $T_\mathrm{c}(H)$ and a broad peaking at $T_\mathrm{p} > T_\mathrm{c}(H)$. They interpreted their results in the context of a Griffiths-type pseudocriticallity [17] of the system, contrasting with earlier attribution [15] of the low-H peak to the dynamics inherent in the RFIM criticality.

In this letter we present magnetization and ac susceptibility data in $\mathrm{Fe}_{0.42}\mathrm{Zn}_{0.58}\mathrm{F}_2$, which indicate that at low H, here called the *weak* RFIM regime, the magnetic behaviour is compatible with the maintenance of the AF LRO ground state, occurring for $T < T_\mathrm{c}(H)$, once the ZFC protocol is followed. In this regime, the ZFC LRO undergoes a PT at $T_\mathrm{c}(H)$, accompanied by an extreme critical slowing-down, as reported [12–15] in earlier studies of $\mathrm{Fe}_x\mathrm{Zn}_{1-x}\mathrm{F}_2$. For large H, here denoted as the *strong* RFIM regime, intense random fields make at least part of the ZFC LRO configuration unstable, separating flipping domains from the AF backbone, even for $T < T_\mathrm{c}(H)$. The PT is destroyed, and $T_\mathrm{c}(H)$ represents only a pseudocritical boundary. A glassy phase sets in for $T_\mathrm{c}(H) < T < T_\mathrm{eq}(H)$. In addition, our results explain the nature of two peaks in ZFC χ'_ac, which appear [6] in the strong RFIM regime, for intermediate x. The glassy behaviour induced by strong random fields extends (at least) along the interval $0.31 \le x \le 0.60$ [18–20]. The lack of stability of the AF LRO [21] and the glassy behaviour induced by strong random fields [22, 23], are in agreement with theoretical studies in DAFs submitted to strong applied fields.

The ZFC and FC results for $\mathrm{d}(M/H)/\mathrm{d}T$ *vs.* T and χ'_ac *vs.* T in $\mathrm{Fe}_{0.42}\mathrm{Zn}_{0.58}\mathrm{F}_2$ are displayed in fig. 1a and b, for comparison. At low H, $\mathrm{d}(M/H)/\mathrm{d}T$ shows a sharp and symmetric ZFC peak at the critical temperature $T_\mathrm{c}(H)$. The peak shifts to lower temperatures as H increases, following the expected REIM to RFIM crossover scaling [1]. A small ZFC-FC hysteresis, limited to the vicinity of $T_\mathrm{c}(H)$, is observed below the equilibrium temperature $T_\mathrm{eq}(H)$. This hysteresis corresponds to an excess in the FC magnetization, which relaxes towards the ZFC ground state just below $T_\mathrm{c}(H)$. In fig. 1b, we see that ZFC χ'_ac shows a maximum, rounded by the extreme critical slowing-down, which coincides in temperature with the ZFC $\mathrm{d}(M/H)/\mathrm{d}T$ peak. χ'_ac exhibits negligible hysteresis with respect to the ZFC and FC procedures. The low-H features described above have been fairly used [14, 15] as evidence that a phase transition occurs in the presence of weak random fields in $\mathrm{Fe}_x\mathrm{Zn}_{1-x}\mathrm{F}_2$, for samples with $x = 0.46$ and 0.47. In the strong RFIM regime ($H > 20\,\mathrm{kOe}$) the ZFC $\mathrm{d}(M/H)/\mathrm{d}T$ peaks of fig. 1a show a visible rounding, which increases as $\sim H^2$. The ZFC-

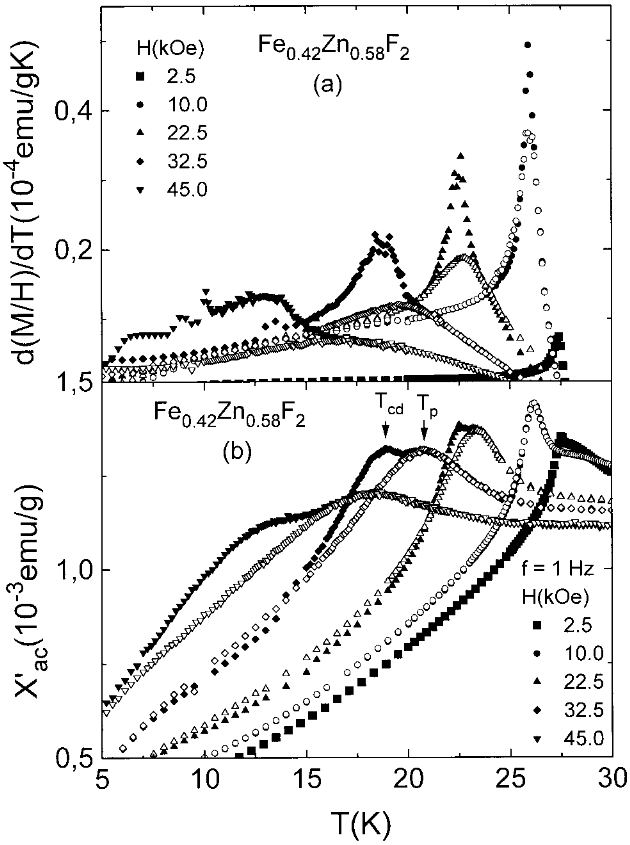

Fig. 1 – (a) $\mathrm{d}(M/H)/\mathrm{d}T$ *vs.* T for applied fields $H = 2.5$, 10, 22.5, 32.5 and 45 kOe. Filled symbols are ZFC data and open symbols are FC. (b) χ'_{ac} *vs.* T measured at $f = 1\,\mathrm{Hz}$, for the same H as in panel (a). Filled symbols are ZFC data and open symbols are FC. The amplitude of the ac field is $h_{\mathrm{ac}} = 4\,\mathrm{Oe}$.

FC hysteresis extends to lower temperatures. The amplitude of these peaks decreases with increasing H, contrasting with the behaviour observed in the *weak* RFIM regime [14]. This behaviour is consistent with the field dependence of the amplitudes of the ZFC peaks seen in C_{m} *vs.* T and also in the temperature derivative of the linear birefringence $\mathrm{d}(\Delta n)/\mathrm{d}T$ *vs.* T, for $Fe_{0.46}Zn_{0.54}F_2$, and $Fe_{0.60}Zn_{0.40}F_2$, respectively [13, 24]. In this regime, the ZFC χ'_{ac} peak splits into a small low-T peak at $T = T_{\mathrm{cd}}(H)$ and a second broad maximum at $T = T_{\mathrm{p}}(H)$, with $T_{\mathrm{cd}}(H) < T_{\mathrm{p}}(H)$, as shown in fig. 1b. A pronounced ZFC-FC hysteresis is found, close and below T_{cd}. The χ'_{ac} peaks at T_{cd} and the one at T_{p} become excessively rounded and, as occurs with the ZFC $\mathrm{d}(M/H)/\mathrm{d}T$ peaks, decrease in amplitude with increasing H. At low frequencies ($f = \omega/2\pi \sim 1\,\mathrm{Hz}$), the second peak at $T_{\mathrm{p}}(H)$ coincides quite well with $T_{\mathrm{eq}}(H)$, as obtained from the ZFC-FC irreversibility of the magnetization, in the time scale of the experiments (see fig. 1). Heat capacity measurements, recently reported [25] by Satooka *et al.* in $Fe_{0.58}Zn_{0.42}F_2$, presented a visible rounding in the width of the magnetic heat capacity peak, which also increases as $\sim H^2$, for $H > 4\,\mathrm{T}$. This result is in agreement with the evolution of the $\mathrm{d}(M/H)/\mathrm{d}T$ peaks with H, as shown in fig. 1a, and also with the magnetic phase diagram of $Fe_{0.56}Zn_{0.44}F_2$ and $Fe_{0.60}Zn_{0.40}F_2$, reported by ref. [20]. In ref. [25], the absence of hysteresis between ZFC and FC protocols for $H < 3.0\,\mathrm{T}$ is also compatible with

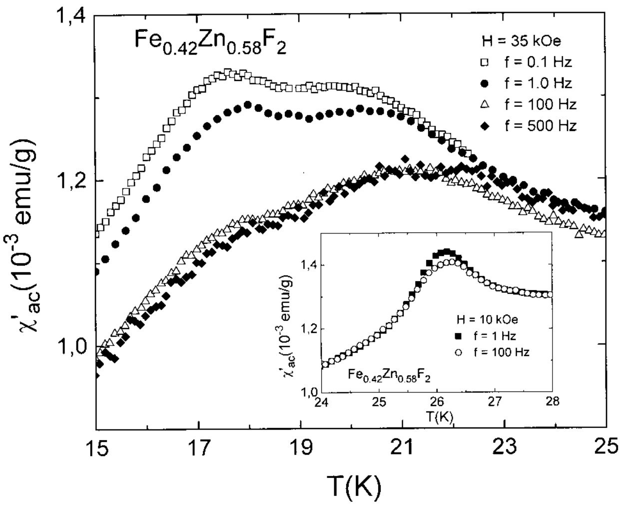

Fig. 2 – Frequency dependence of the ZFC χ'_{ac} in the strong RFIM regime ($H = 35\,\mathrm{kOe}$) for $f = 0.1$, 1, 100 and 500 Hz and $h_{\mathrm{ac}} = 4\,\mathrm{Oe}$. The inset shows the behaviour in the weak RFIM regime ($H = 10\,\mathrm{kOe}$) at two frequencies, for comparison.

the magnetic behaviour presented in this work. Except in the close vicinity of $T_{\mathrm{c}}(H)$, where a small hysteresis between these procedures is visible at low H. The single low H ZFC χ'_{ac} peak has been associated with the RFIM criticality in previous studies [15]. Our magnetization and ac susceptibility data at low-H are compatible with this interpretation, *i.e.*, $T_{\mathrm{cd}}(H)$ corresponds to the critical temperature $T_{\mathrm{c}}(H)$, where the cooperative PT takes place. The critical dynamics acts only very close to $T_{\mathrm{c}}(H)$ in the weak RFIM regime. At higher H, however, the broader peaks centered at $T_{\mathrm{cd}}(H) = T_{\mathrm{c}}(H)$ are, instead, associated with a gradual transition from a partially ordered AF phase to a glassy phase, which dominates the region $T_{\mathrm{c}}(H) < T < T_{\mathrm{eq}}(H)$ [18–20]. This will be clarified after the presentation of the phase diagram and explanation of the time dependence of M, respectively in figs. 3 and 4, below. It is worth noting that the Griffith singularities argument [6] may be quite valid and its relation with the glassy behavior observed at large H deserves further investigations.

In fig. 2, the frequency dependence of the ZFC χ'_{ac} is displayed in the weak and strong RFIM regimes, for $f = \omega/2\pi$ ranging from 0.1 to 500 Hz. For $0 < H < 20\,\mathrm{kOe}$, where the only peak found is associated with the PT at $T_{\mathrm{c}}(H)$, we note the sharpening of the peak and the increasing in its amplitude with decreasing f (see inset of fig. 2). No observable shift in the peak temperature occurs as a function of f. Our results in the weak RFIM regime of $\mathrm{Fe_{0.42}Zn_{0.58}F_2}$ are in agreement with those previously observed in $\mathrm{Fe_{0.46}Zn_{0.54}F_2}$ by King *et al.* [15], confirming the low-frequency nature of the critical fluctuations in RFIM systems. For $H > 20\,\mathrm{kOe}$, the second broad peaking appears at $T_{\mathrm{p}}(H)$. The amplitude of both peaks decreases, and $T_{\mathrm{p}}(H)$ shifts to higher temperatures, with increasing f. At higher frequencies, however, the narrow low-T peak at $T_{\mathrm{cd}}(H)$ disappears and the broad peaking at $T_{\mathrm{p}}(H)$ becomes the only feature in the χ'_{ac} *vs.* T curves. These latter characteristics reflect the glassy dynamics, which occur at the strong RFIM regime.

The critical, $T_{\mathrm{c}}(H)$, and equilibrium, $T_{\mathrm{eq}}(H)$, boundaries, measured from the magnetization data, are mapped in the (H, T) dynamic phase diagram illustrated in fig. 3. We include, for comparison, the location of the temperatures $T_{\mathrm{cd}}(H)$ and $T_{\mathrm{p}}(H)$ obtained from the ZFC χ'_{ac} data at frequencies $f = 1$ and 100 Hz. In the weak RFIM regime, both $T_{\mathrm{c}}(H)$ and $T_{\mathrm{eq}}(H)$ follow the expected REIM-RFIM crossover scaling: $\Delta T_{\mathrm{c(eq)}} = T_{\mathrm{N}} - bH^2 - T_{\mathrm{c(eq)}}(H) =$

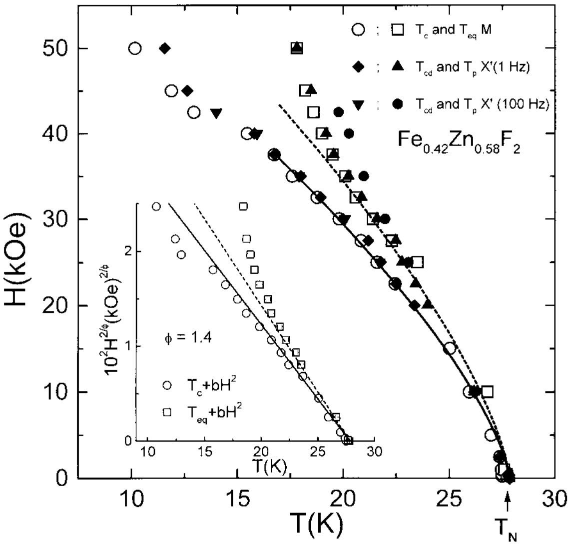

Fig. 3 – Part of the dynamic phase diagram of $Fe_{0.42}Zn_{0.58}F_2$. The critical, $T_c(H)$, and equilibrium, $T_{eq}(H)$, boundaries (open symbols) were obtained from magnetization data. $T_{cd}(H)$ and $T_p(H)$ (filled symbols) were obtained from the ZFC χ'_{ac} measurements, at frequencies $f = 1$ and 100 Hz. The solid and dashed lines are fits using REIM-RFIM crossover scaling for $T_c(H)$ and $T_{eq}(H)$, respectively, using $\phi = 1.42$ (see text). $T_c(H)$ and $T_{eq}(H)$ are plotted linearly *vs.* $H^{2/\phi}$ in the inset, after a mean-field correction is made to each.

$C_{c(eq)}H^{2/\phi}$, where $\phi = 1.42 \pm 0.08$ is the universal REIM-RFIM crossover exponent, and b, C_c and C_{eq} are constants. Good fits for $T_c(H)$ and $T_{eq}(H)$, using the above law with $\phi = 1.42$, are shown in the main figure and inset. The extrapolation of $T_c(H)$ to $H \to 0$ gives the Néel temperature $T_N = 27.85 \pm 0.15\,K$. The second ZFC χ'_{ac} broad peak at $T_p(H)$ is perceived only for $H > 20\,kOe$. $T_p(H)$ coincides quite well at low frequencies with the equilibrium temperature $T_{eq}(H)$, defined in terms of the ZFC-FC irreversibility of the magnetization. In the strong RFIM regime, however, $T_p(H)$ follows a convex de Almeida-Thouless line ($\phi > 2$), which shifts to lower temperatures as f decreases, as observed in spin glasses. In fact, $T_p(H) = T_{eq}(H)$ only when $1/f$ coincides with the time scale of the "dc" magnetization measurements (1 to 100 s, typically). The frequency dependence of the position of the low-T peak at $T_{cd}(H)$ is restricted to the strong random field regime, where the amplitude of the ZFC χ'_{ac} peaks decreases as f increases, as shown in fig. 2. These latter features are associated to the gradual changing from a partially ordered AF state, for $T < T_c(H)$, to the glassy state, which dominates the interval $T_c(H) < T < T_{eq}(H)$ for $H > 35\,kOe$. An intermediate regime occurs for $20 < H < 35\,kOe$, where $T_{eq}(H)$ changes from concave to convex shape.

We are now led to ask how the magnitude of the random fields affects the AF LRO ground state and, consequently, modifies the dynamics in the critical region. In fig. 4 we show the time dependence of the ZFC magnetization in the strong random field regime at $T = 10$ and 4.5 K. M_{ZFC} displays no time dependence at low H, as expected due to the stability of the AF LRO ground state in the *weak* $d = 3$ RFIM problem. For higher H, however, strong random fields make the AF configuration unstable. Close to, but below $T_c(H)$ some of the spins separated from the infinite cluster may relax back to the AF configuration, as shown in fig. 4a, for $H = 37\,kOe$. In the vicinity of and above $T_c(H)$, however, the system evolves to form clusters aligned with H, as shown in fig. 4b, for $H = 45\,kOe$. This magnetically viscous regime was

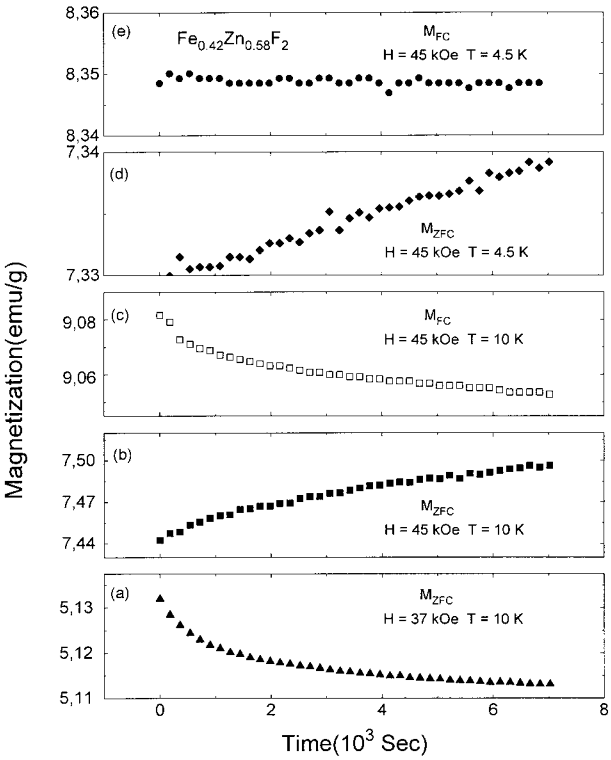

Fig. 4 – Time dependence of M_{ZFC} at $T = 10\,\mathrm{K}$, for $H = 37\,\mathrm{kOe}$ (a) and 45 kOe (b). The time dependence of M_{FC} is displayed in (c) for the same T, after cooling the sample from the paramagnetic phase at $H = 45\,\mathrm{kOe}$. Similar measurements of M_{ZFC} and M_{FC} are displayed in (d) and (e), respectively, for $T = 4.5\,\mathrm{K}$.

earlier observed by neutron scattering experiments in $\mathrm{Fe_{0.31}Zn_{0.69}F_2}$ under strong random fields [11]: the ZFC staggered magnetization decreases with time for $T_{\mathrm{c}}(H) < T < T_{\mathrm{eq}}(H)$. If T is decreased from the paramagnetic phase to the glassy phase, the magnetization gains an excess relative to the previously mentioned ZFC protocol, followed by field increasing procedure. The system now relaxes toward one of the multi-degenerate lower-energy configurations, with an intermediate magnetization between the FC and ZFC states. This is easily seen by comparing figs. 4c and b.

In figs. 4d and e the time dependencies of the ZFC and FC magnetization are compared for the same value $H = 45\,\mathrm{kOe}$, but at a lower $T = 4.5\,\mathrm{K}$. M_{ZFC} increases with time as it does for $T = 10\,\mathrm{K}$. However, M_{FC} is now frozen, displaying no noticeable time dependence.

In conclusion, we have made a comparative study of the magnetization and dynamic susceptibility in $\mathrm{Fe_{0.42}Zn_{0.58}F_2}$, which distinguishes the features associates with the *weak* and *strong* RFIM regimes in this compound. At low H, our ZFC results are compatible with a stable AF LRO ground state, which undergoes a PT at $T_{\mathrm{c}}(H)$, governed by the REIM-RFIM crossover scaling. An extreme RFIM critical slowing-down rounds the PT, in agreement with earlier studies [13–15]. At large H, however, AF LRO becomes unstable and the PT is destroyed. The extreme rounding of the ZFC $\mathrm{d}(TM)/\mathrm{d}T$ peaks found [2] at high H, and the existence of two ZFC χ'_{ac} peaks [6] in samples of $\mathrm{Fe}_x\mathrm{Zn}_{1-x}\mathrm{F}_2$, with $x \sim 0.50$, are not in the scope of the Imry-Ma [26] arguments, valid only for the *weak* RFIM problem. These features, instead, are associated to the effects of a random-field–induced glassy dynamics which emerge at high H, in samples of $Fe_xZn_{1-x}F_2$ with $0.31 < x < 0.60$ [18–20].

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