Loss of Long Range Order in the 3D Random Field Ising Model

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We report a synchrotron magnetic x-ray scattering study of the temperature evolution of the magnetic long range order (LRO) of $Mn_{0.75}Zn_{0.25}F_2$ in a magnetic field with emphasis on the behavior after zero field cooling (ZFC). We show that with increasing temperature the metastable ZFC LRO vanishes continuously at a temperature well above the equilibrium T_N; the LRO follows a rounded power law with exponent $\beta_{ZFC}=0.20\pm0.02$ and a transition width which scales as $\sim H^2$ but with no divergent critical fluctuations. We argue that this behavior is generic to the random field Ising model.

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The problem of quenched disorder and competing interactions is of fundamental importance in condensed matter systems. The essential physics is most simply encapsulated in the random field Ising model (RFIM) [1-3], which has applications for systems as diverse as hydrogen in metals, commensurate phases on surfaces, order-disorder structural phase transitions, and fluid phase transitions in porous media. The canonical prototype for the RFIM has been diluted antiferromagnets in an applied field [1-3]. Initially much of the work on this problem was focused on the lower marginal dimensionality of the model, d_l, the dimensionality at which the phase transition is destroyed in equilibrium. There has been a gradual theoretical consensus [1] that $d_l=2$ and in the past year a transition to long range order (LRO) on field cooling was seen experimentally in a three-dimensional RFIM system for the first time [4]. However, the physics of the RFIM is dominated by metastability effects which are, at present, not well understood. Thermodynamic measurements of bulk properties at low fields show anomalies after zero field cooling and subsequent heating in a field which mimic the critical behavior of an equilibrium phase transition [5-8]. However, it has not been possible to reconcile this apparent critical behavior with the fact that the system is manifestly not in equilibrium, and with the absence of a divergent magnetic correlation length [9].

An essential component of our empirical description of the RFIM transition which has been missing to date is a quantitative measure of the behavior of the order parameter following various cooling and heating protocols. Neutron measurements of the LRO are severely affected by extinction [9] while other probes are hampered by the large disorder. Magnetic x-ray scattering suffers from none of these disadvantages [4,10]. The scattering is extinction free because of the very weak magnetic x-ray scattering cross section and high spatial resolution is routine. The inherent eV energy resolution ensures correct integration over all relevant thermal fluctuations.

The metastability is characterized by a history dependence of the magnetic state. Specifically, on cooling in the presence of a random field (FC) the bulk of the system abruptly falls out of equilibrium in a narrow temperature range and forms a long-lived metastable domain state [9,11]. Alternatively, if LRO is first established by cooling in zero field and a field is subsequently applied (ZFC) then LRO is retained on warming until the metastability temperature, $T_M(H)$, is reached. As previously reported [4], in the near surface region LRO has also been seen by x rays on FC. This allows us to determine the equilibrium Néel temperature, $T_N(H)$, directly. In this paper we focus on the unusual metastability which lies at the heart of the RFIM. The salient result of this work is that the ZFC LRO in the metastable temperature region above $T_N(H)$ vanishes continuously with increasing temperature with a universal, rounded power-law behavior. This observation clarifies empirically many apparently contradictory results in the literature [4-9,12]. Previous neutron experiments on $Mn_{1-x}Zn_xF_2$ [9] suggest that the continuous shedding of LRO in this metastable system arises from the flipping of domains of progressively larger size as the metastability boundary is approached. However, a quantitative theory of these effects is lacking.

The experiments were performed on the IBM-MIT X20A beam line at the National Synchrotron Light Source. The x-ray beam was vertically and horizontally focused by a platinum coated silicon mirror. X rays of energy $E=8.35$ keV were selected with a double bounce Ge(111) monochromator. The focused 1 mm×1 mm spot incident on the sample was horizontally scattered onto a flat Si(111) analyzing crystal. The resulting longitudinal resolution was 4.8×10^{-4} Å^{-1} half width at half maximum (HWHM). The transverse resolution of 5.7×10^{-5} Å^{-1} HWHM was controlled by the sample mosaic and by beam inhomogeneities caused by imperfections in the mirror. This x-ray resolution is more than an order of magnitude better than that used in the neutron experiments [9,11,12]. The sample was mounted in a split-pair superconducting magnet with the $\hat{c}$ axis aligned

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parallel to the vertical magnetic field. A face perpendicular to the $\hat{a}$ axis was polished for the x-ray work. The penetration depth of the x rays was ~ 1.5 μm compared with 3.6 μm in Ref. [4]. The field inhomogeneity was $< 0.5\%$ over 10 mm. This translates into a potential smearing of the transition of less than 20 mK at $H = 6.0$ T. The temperature was controlled to better than 10 mK over the course of a typical scan. Temperature measurements were performed via a field-insensitive carbon-glass sensor.

$Mn_{1-x}Zn_xF_2$ is a weakly anisotropic Ising antiferromagnet. It has the tetragonal rutile structure with a large isotropic exchange and a smaller dipolar anisotropy [2]. At low temperatures and low fields, the spins form an Ising phase and the Hamiltonian is isomorphic with the RFIM [1–3]. At high fields the spins "flop" into the plane perpendicular to the applied field, as discussed in Refs. [6] and [9]. In this phase the random field, which is directed along the $\hat{c}$ axis, plays no role and LRO is established. It is known that heating at a fixed field from the XY phase (FHXY) into the Ising phase does not destroy LRO [9]. This provides an experimentally more convenient technique for preparing the system in a LRO state. It is equivalent to ZFC and was used for fields $H \geq 6.0$ T.

Data from two samples of $Mn_{0.75}Zn_{0.25}F_2$ are reported here. The first, labeled No. 1, which was used in our previous work [4], exhibits a transition to LRO in the near surface region on FC. The second, labeled No. 2, was cut from the first, but underwent a more extensive polishing process which produced a much smoother surface. Interestingly, it does not show LRO on FC but rather forms a domain state consistent with that seen with neutron diffraction [2,9]. This lends credence to the idea that line defects (scratches) on the surface of sample No. 1 play an important role in the nucleation of the LRO; specifically, a subtle imbalance in the random fields in the neighborhood of line defects could initiate the transition. This will be discussed in more detail in a future publication. For the purpose of this current work on hysteresis, however, both samples exhibit identical ZFC behavior thus confirming that the behavior we observe is intrinsic. Both are of excellent crystallographic perfection and chemical homogeneity. This is best evidenced by the sharp x-ray peaks, and the rounding in the $H = 0$ Néel transition which is less than 10 mK. Both samples are from the same boule used by Shapira, Oliveira, and Foner [6]; they exhibit identical $H = 0$ and ZFC transition temperatures to within the experimental errors. This eliminates the possibility of chemical or physical differences such as differing sample surface temperatures affecting the results. For all of the results reported in this paper both field and chemical inhomogeneity effects are negligible.

We now discuss the essential experimental results with emphasis on the ZFC (or equivalently FHXY) behavior. As observed previously [4], sample No. 1 shows a transition to LRO on cooling in a field. The FC LRO transition temperature $T_N(H)$ is systematically less than the metastability temperature $T_M(H)$. By contrast, on field cooling sample No. 2 shows Lorentzian squared (L^2) scattering with widths $\kappa(H,T)$ which agree quantitatively with those observed previously in the bulk using neutron scattering techniques [9]. This is an important result both from the point of view of establishing the technique of x-ray magnetic scattering for probing two spin correlation functions in disordered magnetic systems and for making contact with previously observed behavior. In brief then, the FC behavior in the near surface region (~ 1.5 μm) of sample No. 2 is identical to that in the bulk. For sample No. 1 a quantitative analysis of the scattering shows that approximately 60% of the illuminated volume exhibits a transition to LRO while the other 40% condenses into the FC metastable domain state [4]. We will discuss the ordering process and the FC L^2 results in more detail in a future publication. Our emphasis in this paper will be on the ZFC behavior which is identical in both samples.

As observed previously in both neutron [9,11] and x-ray [4] experiments for both ZFC and FHXY procedures the system exhibits LRO. We show in Fig. 1 the LRO intensity in sample No. 1 on field cooling (FC), reheating from within the Ising phase (FHI), and reheating from the XY phase (FHXY) for $H = 6.0$ T. The LRO on FC first appears at $T_N(6.0) = 41.4 \pm 0.2$ K; the LRO vanishes in an identical fashion on heating for both FHI and FHXY procedures at $T_M(6.0) = 42.63 \pm 0.05$ K in good agreement with the results of Ref. [9]. The difference

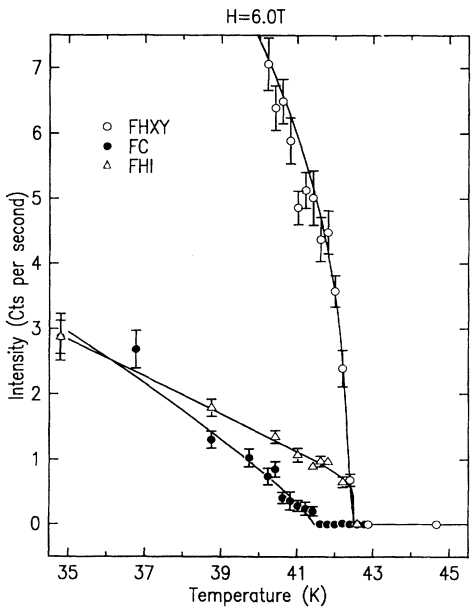

FIG. 1. (100) integrated LRO intensity vs temperature after field cooling (FC) and field heating from the Ising (FHI) and XY (FHXY) phases in sample No. 1. The solid lines are guides to the eye.

$T_M(6.0) - T_N(6.0) = 1.2 \pm 0.2$ K should be compared with the value $T_M(6.0) - T_N(6.0) = 0.9 \pm 0.2$ estimated by Cowley *et al.* [9] based on power-law fits to the measured inverse correlation length versus temperature in the equilibrium region above T_M. From x-ray measurements at $H = 5.0$ and 3.5 T we observe behavior identical to that shown in Fig. 1 with $T_M(H) - T_N(H) = 0.5 \pm 0.2$ K and 0.3 ± 0.2 K, respectively. These should be compared with the predictions of Cowley *et al.* [9] of $T_M(5.0) - T_N(5.0) = 0.5 \pm 0.2$ K and $T_M(3.5) - T_N(3.5) = 0.25 \pm 0.1$ K. This excellent agreement between our measurements and the predictions of Ref. [9] is most gratifying. This consistency validates their analysis which yields for the equilibrium correlation length exponent $\nu = 1.4 \pm 0.3$ and it also provides additional strong evidence that $T_N(H)$ measured in sample No. 1 represents the true equilibrium phase transition temperature to within the quoted errors.

We now discuss the observed FHXY and ZFC behaviors. We show in Fig. 2 the ZFC-FHXY LRO intensities as a function of temperature for both samples at a series of fields. Qualitatively, the intensities appear to exhibit power-law-like behavior as a function of temperature as one would expect at a normal second order phase transition. This is in spite of the fact that, as may be seen from Fig. 1 and from previous work [2,6,9,11], the ZFC LRO is only one of many possible metastable LRO states. On closer examination (see Fig. 3, especially the inset) it is evident that each of the transitions for $H > 0$ is rounded as suggested by earlier neutron experiments [9,11,12]. This is in contrast to the behavior at $H = 0$ where the intensity near T_N is well described by a simple power law $I \sim [T_N(0) - T]^{2\beta}$ with $\beta = 0.35 \pm 0.03$ and with a fitted rounding of < 0.01 K.

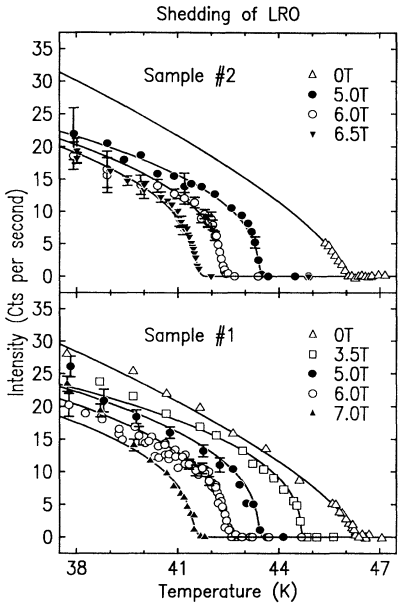

FIG. 2. Order parameter squared vs temperature after ZFC ($H = 3.5$ and 5.0 T) and FHXY ($H = 6.0$, 6.5, and 7.0 T) in samples No. 1 and No. 2. The solid lines through the $H = 0$ data are the results of power-law fits $I \sim (T_N - T)^{2\beta}$ with $2\beta = 0.70$. The solid lines through the $H \neq 0$ data are the results of fits to a Gaussian rounded power-law equation (1) with $2\beta_{ZFC} = 0.40$ and $\sigma_{ZFC}(H) = 0.0034H^2(\mathrm{K/T^2})$.

There is, at present, no theory for this shedding of the metastable LRO. In order to characterize the data empirically we assume a power-law description with a Gaussian distribution of transition temperatures, that is,

$$I(T,H) = \frac{A}{\sqrt{\pi}\sigma_{ZFC}(H)} \int \left[\frac{t_C - T}{t_C}\right]^{2\beta_{ZFC}} \times \exp\left[-\left[\frac{t_C - T_C(H)}{\sigma_{ZFC}(H)}\right]^2\right] dt_C . \quad (1)$$

The Gaussian approximation is for computational convenience alone. Initial fits gave $\beta_{ZFC} \sim 0.2$ for both samples at all fields, that is, for $H = 3.5$, 5.0, 6.0, 6.5, and 7.0 T. Further, the fitted widths, $\sigma_{ZFC}(H)$, were found to scale approximately as H^2; specifically we found $\sigma_{ZFC}(H) = 0.0034H^2(\mathrm{K/T^2})$. Here the exponent 2 is uncertain at a level of at least ± 0.5. The solid lines in Fig. 2 are calculated using Eq. (1) with $\beta_{ZFC} = 0.20$ and the above expression for $\sigma_{ZFC}(H)$. Only the overall amplitude and $T_C(H)$ are adjustable for each separate field. Clearly, this universal form describes the ZFC intensity data for both samples at each field remarkably well. As a practical matter the ZFC LRO component is unobservable for $T - T_C(H) \gtrsim 1.5\sigma_{ZFC}(H)$; this therefore serves to define $T_M(H)$. Thus we have $T_M(H) - T_C(H) \simeq 0.005H^2(\mathrm{K/T^2})$. Finally, we should note that in $Mn_{0.5}Zn_{0.5}F_2$ at $H = 0.34$ T, Thurston *et al.* [10] also found $\beta = 0.2$.

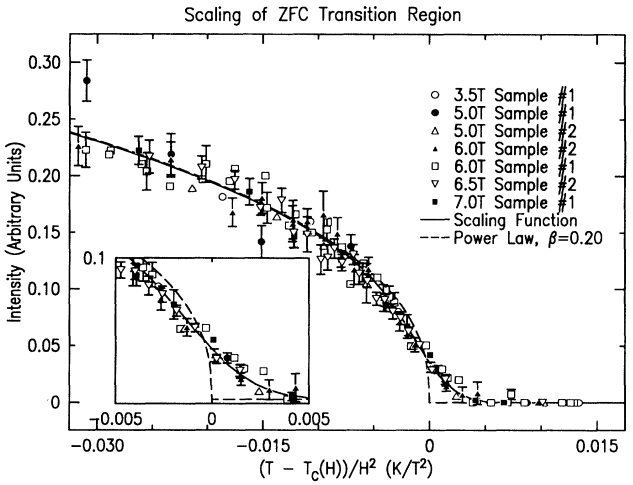

FIG. 3. Scaling plot showing the normalized integrated intensity for all data in Fig. 2 vs the scaled reduced temperature $[T - T_C(H)]/H^2$. The solid line is Eq. (1) with $\beta_{ZFC} = 0.20$ and $\sigma_{ZFC} = 0.0034H^2$. The dashed line is a single power law with $\beta = 0.20$ and no rounding.

In order to display the results in a fashion which illustrates the universal scaling behavior we plot in Fig. 3 the normalized measured intensity versus $[T-T_C(H)]/H^2$. We emphasize that all of the data shown in Fig. 3 are for temperatures in the metastable temperature region above $T_N(H)$. Figure 3 is the salient result of this paper. The observed behavior simulates equilibrium critical phenomena but, as shown in the original neutron studies [6,9] and as verified by these x-ray magnetic scattering measurements, there is no divergent magnetic correlation length at $T_C(H)$ and, of course, all of the data are in the metastable temperature region between $T_N(H)$ and $T_M(H)$. We label this new behavior *trompe l'oeil critical behavior.* These observations enable one to understand straightforwardly a variety of RFIM bulk property measurements including especially thermal expansion [6] and birefringence $[d(\Delta n)/dT]$ results [6–8] as well as previous neutron experiments [2,9,11,12]. For both the thermal expansion and birefringence in $Mn_{1-x}Zn_xF_2$ and $Fe_{1-x}Zn_xF_2$ one observes at low fields sharp, symmetric peaks with the peak temperature $T_C(H)$ just below $T_M(H)$. Both the widths of the peaks and the temperature differences, $T_M(H)-T_C(H)$, appear to scale approximately as $H^{2/\phi}$ with $\phi\sim 1.3$ to 1.4. Quantitatively, the measured widths and temperature differences, $T_M(H)-T_C(H)$, in both $Mn_{1-x}Zn_xF_2$ and $Fe_{1-x}Zn_xF_2$ correspond well to those predicted by Fig. 3 for comparable values of x. The difference between the inferred $H^{2/\phi}$ scaling for the transition width for $d(\Delta n)/dt$ from the birefringence measurements and our observed H^2 behavior for the ZFC LRO is not significant given the combined experimental uncertainties. Our own more recent measurements on the ZFC behavior in $Fe_{0.5}Zn_{0.5}F_2$ also give an H^2 scaling of the transition width [13]. We emphasize that $T_M(H)-T_C(H)$ is much smaller than $T_M(H)-T_N(H)$; for example, at 6.0 T, $T_M(6.0)-T_C(6.0)\simeq 0.2$ K while $T_M(6.0)-T_N(6.0)=1.2\pm 0.2$ K. In brief, both the ZFC thermal expansion [6] and $d(\Delta n)/dT$ [5,7,8] data exhibit peaks at a temperature $T_C(H)$ which almost certainly corresponds to the point at which the temperature derivative of the ZFC magnetic x-ray Bragg intensity, $|dI/dT|$, is a maximum rather than at the equilibrium phase transition temperature $T_N(H)$. Accordingly, the peaks observed in these experiments reflect the continuous shedding of metastable long range order and are unrelated to the equilibrium critical behavior of the RFIM [5].

As noted above, there is currently no theory for the vanishing of the LRO of the ZFC metastable state. Some indications of the underlying physical behavior are adumbrated in the neutron studies [9]. As may be seen in Fig. 11 of Ref. [9] for $H=6.5$ T, ZFC, as $T_M(H)$ is approached, diffuse scattering appears at a temperature above $T_N(H)$ but well below $T_M(H)$ with a width which decreases with increasing temperature and equals the FC width at $T_M(H)$. Similar behavior is found in $Fe_{0.5}Zn_{0.5}F_2$ [13]. This suggests that in the metastable temperature region domains of progressively larger size flip over with increasing temperature hence diminishing the LRO. This process goes to completion at $T_M(H)$ when the "flipped" domain size equals the equilibrium FC domain size. Qualitatively, then, the rounding may be understood as a finite-size effect; simple finite-size scaling arguments seem capable of giving the observed H^2 behavior for the width. Of course, this heuristic argument is unable to explain the effective exponents $\beta_{ZFC}=0.20$ and $\alpha_{ZFC}\sim 0.0$ (from the birefringence studies). Clearly, a proper theory for the metastability of the RFIM is required. We hope that our results on the order parameter will provide both an appropriate stimulus for the construction of such a theory and a guide for the form it must take.

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