Transition to Long-Range Order in the Three-Dimensional Random-Field Ising Model

J. P. Hill,[1] T. R. Thurston,[2] R. W. Erwin,[3] M. J. Ramstad,[1] and R. J. Birgeneau[1]
[1]*Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139*
[2]*Sandia National Laboratory, Albuquerque, New Mexico 87185*
[3]*National Institute of Standards and Technology, Gaithersburg, Maryland 20899*
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We report both synchrotron-x-ray and neutron-diffraction studies of the magnetic ordering in a crystal of $Mn_{0.75}Zn_{0.25}F_2$ in a magnetic field. The neutron experiments reproduce previous results showing the establishment of a short-range domain state on cooling in a field. Remarkably, the magnetic x-ray scattering shows that within a few microns of the surface there is a second-order transition to true long-range order on field cooling. Implications of these results are discussed.

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The random-field Ising model (RFIM) is perhaps the simplest system in which both quenched disorder and competing interactions play a fundamental role.[1-3] In spite of the simplicity of the Hamiltonian, the resultant physics is remarkably rich and, as yet, not well understood. In the early stages of work on this problem, much attention was focused on the determination of the lower marginal dimensionality d_l, that is, the dimensionality above which there could be a transition to long-range order (LRO). It is now believed that $d_l=2$, largely because of Imbrie's rigorous proof that the RFIM exhibits true LRO at $T=0$ in $d=3$ dimensions.[4] However, no such transition in $d=3$ has ever been observed.[3] Instead, on cooling in the presence of a random field (FC), the system abruptly falls out of equilibrium and a long-lived domain state is established.[5] Heuristic ideas for the onset of metastability and the behavior in the metastable region have been developed based on the experiments but our theoretical understanding is still quite incomplete.[6]

In this paper, we report a high-resolution synchrotron magnetic x-ray scattering study of the ordering process in $Mn_{0.75}Zn_{0.25}F_2$ in a magnetic field. As discussed previously,[3,7] this system is in the universality class of the $d=3$ RFIM but, in addition, has low-lying spin-wave modes which facilitate the relaxation of the sublattice magnetization. Previous synchrotron-x-ray studies of MnF_2 and $Mn_{0.5}Zn_{0.5}F_2$ have shown that magnetic x-ray techniques are invaluable in characterizing antiferromagnetic transitions since the order-parameter scattering is extinction free and the real-space resolution may be of order microns.[8] Our original intent in these experiments was to study the behavior of the FC RFIM domain state at small fields and hence large domain sizes. Remarkably, as we shall discuss in detail below, we discovered that our x-ray experiments reveal a transition to LRO under conditions where previous neutron experiments[7] had shown the establishment of a domain state with correlation lengths as short as 500 Å. To rule out pathological effects, we repeated the neutron experiments on the x-ray sample, confirming earlier neutron results.[7] As we shall discuss below, this necessitates that LRO is established within about 2 μm of the surface but not in the bulk. This experiment thus provides direct experimental proof that $d_l<3$ in agreement with Imbrie.[4] Further, it enables us to study the properties of the $d=3$ RFIM at equilibrium, a regime previously inaccessible to experiment.[3,5] Our results clearly have important implications for the interpretation of previous measurements.[3,7,9]

The magnetic x-ray experiments were carried out primarily on the IBM-MIT X20-B beam line of the National Synchrotron Light Source (NSLS). This beam line, equipped with a single bent Si(111) monochromator and a flat reflecting gold-coated glass mirror, provided a beam of x rays with energy $E=17.4$ keV and negligible higher-order contamination. Scattering was in the horizontal plane and we used a flat Si(111) analyzer to achieve high angular resolution in the scattered beam. The consequent resolution was ~ 0.0015 Å^{-1} half width at half maximum (HWHM) longitudinally; the transverse in-plane resolution was controlled by the sample mosaic of $\sim 0.002°$ HWHM. The sample was mounted in an x-ray-compatible split-coil superconducting magnet kindly made available by P. M. Horn of IBM. A limited set of measurements to check the polarization of the magnetic scattering were carried out on IBM-MIT beam line X20-C with the sample mounted in a Displex cryostat. The neutron measurements were carried out using the triple-axis spectrometer BT-2 at the National Institute of Standards and Technology (NIST) Reactor.

The $Mn_{0.75}Zn_{0.25}F_2$ sample was a parallelepiped of dimensions $0.6\times0.6\times0.2$ cm^3 with the **c** axis along a 0.6-cm direction and the **a** axes along the 0.6- and 0.2-cm directions; the 0.6×0.6 cm^2 **a** face was polished to minimize stray scattering and an x-ray spot size of 0.1×0.45 cm^2 was used. This crystal was supplied by Y. Shapira and it was cut from the same boule as that used for magnetization, thermal-expansion,[10] and previous neutron studies.[5,7] The sample was of excellent crystallographic perfection and chemical homogeneity. The former was verified by our own high-resolution x-ray and neutron-diffraction measurements. The microscopic chemical homogeneity was best characterized by the $H=0$ Néel transition itself. We found that any rounding of the transition was much less than 100 mK as measured with both x rays and neutrons. Further, the transition temperatures in the outer 3.6 μm as measured with x rays

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and in the bulk as measured with neutrons agreed to within the respective thermometer absolute-temperature calibration uncertainties. For both the neutron and $H \neq 0$ x-ray studies the sample was mounted with the **c** axis vertical so that the magnetic field was along the **c** axis (Ising) direction and perpendicular to the scattering plane. For the polarization measurements, the **c** axis could be rotated continuously about the [100] direction into the scattering plane.

$Mn_{1-x}Zn_xF_2$ is a tetragonal antiferromagnet with a large isotropic exchange and a smaller Ising dipolar anisotropy which causes the spins to orient along the **c** axis.[3,7] At low fields, there is a transition into an Ising phase; at high fields the spins "flop" and there is a transition into an XY phase. These two regions are separated by the "spin-flop" boundary. The overall behavior in $Mn_{0.75}Zn_{0.25}F_2$ is discussed in detail in Ref. 7. In FC neutron experiments it is found that the XY phase has LRO, whereas in the Ising phase a domain state with exponentially decaying spin correlations is established. This difference in behaviors arises from the fact that the random field induced by the applied field H along **c** is only in the Ising direction.

Both the x-ray and neutron experiments were carried out around the (100) reciprocal-lattice position. Charge (nuclear) scattering, even when arising from magnetoelastic effects, is symmetry forbidden at the (100) position.[11] Thus in the x-ray experiments only magnetic scattering exactly analogous to that seen in neutron-scattering measurements may be observed. The salient result of this paper is illustrated in Fig. 1. Here we show x-ray and neutron scans deep into the Ising regime obtained after cooling in fixed fields of $H=6.6$ and 7.0 T, respectively, to temperature $T=30$ K [$T_N(7.0)\sim 40.8$ K] (the difference in fields is due to a recalibration of the x-ray magnet after the experiments were completed). As expected, the neutron scans reveal the Lorentzian-squared line-shape characteristic of the $d=3$ RFIM FC domain state.[5] The corresponding correlation length is ~ 800 Å in agreement with previous neutron results.[7] The x-ray scan, on the other hand, shows a resolution-limited peak. The x-ray HWHM is 0.7×10^{-4} Å^{-1}, which means that the domain size is well in excess of 1 μm and can be considered to correspond to LRO. In previous neutron experiments it was explicitly verified that the (200) transverse scans were always resolution limited in a field thus eliminating any possible pathological mosaicity effects. There is, therefore, an apparent contradiction between the neutron and x-ray experiments. Most significantly, x rays reveal a LRO antiferromagnetic state as expected at equilibrium according to Imbrie's proof.[4] The neutron data, on the other hand, correspond to the nonequilibrium metastable FC behavior which has been observed exclusively up to now.[3,5,7]

In order to explore this remarkable phenomenon further, and to eliminate all obvious caveats, we have carried out a wide variety of further measurements. Figure 2 shows a series of FC x-ray scans at $H=4.7$ T and a number of temperatures. It is evident that the peak is resolution limited at all temperatures. The intensities

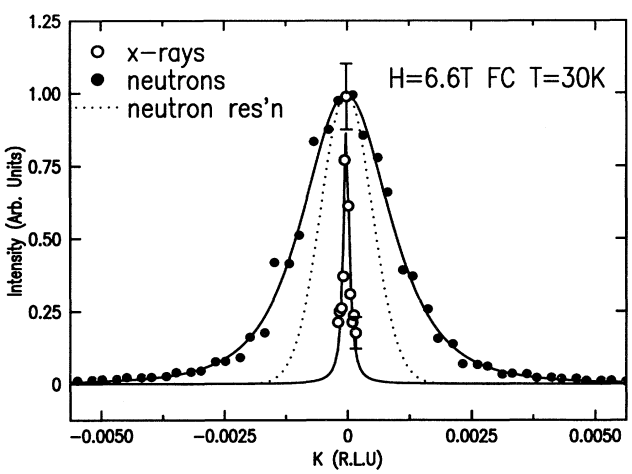

FIG. 1. X-ray and neutron transverse scans through the (100) magnetic peak. Data were taken on the same crystal in an $H=6.6$ and 7.0 T, $T=30$ K FC state for x rays and neutrons, respectively. Peak heights of 1.4 and 860 counts/sec, respectively, have been normalized to unity and a background of 0.1 count/sec has been subtracted from the x-ray data. The solid line through the x-ray data is a Lorentzian of width (HWHM) 7×10^{-5} r.l. (radiation length). The solid line through the neutron data is a Lorentzian squared of width 1×10^{-3} r.l. HWHM. The double-axis neutron resolution with a Gaussian of width 6.1×10^{-4} r.l. HWHM is shown for comparison purposes.

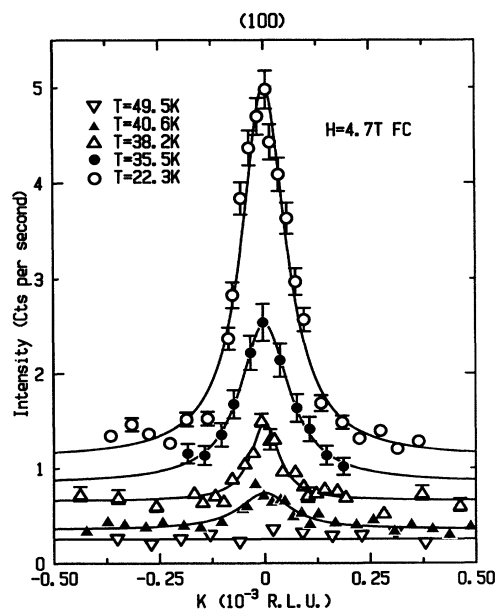

FIG. 2. Transverse x-ray scans through the (100) peak. The temperature is successively lowered at a fixed field of $H=4.7$ T. Data are offset by amounts of 0.125, 0.15, 0.6, and 1.0 counts/sec, respectively. The solid lines are Lorentzians of width 6×10^{-5} r.l. HWHM.

obtained on cooling (FC) and then on heating (FH) are shown in the top panel of Fig. 3. The data are clearly consistent with a conventional second-order magnetic transition to LRO. Further, there is no measurable hysteresis, in contrast to analogous neutron measurements.[7] The data at 5.6 T, shown in the bottom panel of Fig. 3, clearly exhibit rather different behavior. These turn out to provide an important clue as to the underlying cause of the apparent inconsistency between the x-ray and neutron measurements. However, before discussing these data we first describe briefly our x-ray polarization measurements.

As noted above, group-theory arguments[11] demonstrate that there can be no measurable charge scattering at the (100) position. It is, nevertheless, important to verify directly that the scattering is magnetic in origin. In MnF_2 the magnetic scattering at the (100) Bragg peak position is such that the component of the spin perpendicular to the scattering plane produces the dominant contribution to the signal but does not alter the polarization of the x-ray beam, while the component in the plane scatters a horizontally polarized (σ) initial state into a vertically polarized (π) final state and vice versa.[12] The intensity contribution from these off-diagonal elements, however, is reduced by a factor of $\sin^2\theta$ which makes the experiment technically much more difficult. The essence of our experiment was to rotate the **c** axis into the scattering plane and measure the degree of linear polarization of the scattered beam as a function of this rotation angle. Simultaneous measurements of the charge (200) peak provided a consistency check on this procedure.

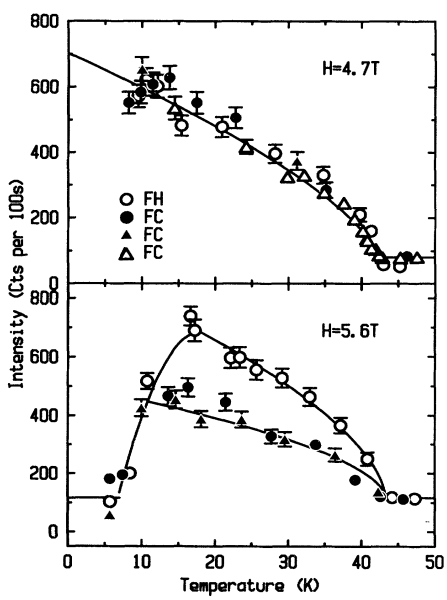

FIG. 3. X-ray peak intensity at two fixed fields. Data were taken by cooling in a fixed field (FC) and heating at fixed field (FH). The upper panel shows data obtained at $H=4.7$ T for which there is no observable hysteresis. The lower panel shows that at $H=5.6$ T a large jump in intensity is observed when warming up from the XY phase. Data from three different runs are shown. Implications of this behavior are discussed in the text.

This test was carried out on the IBM-MIT beam line X20-C at NSLS. This beam line has a vertically and horizontally focusing mirror upstream of a double-bounce Si(111) monochromator. The degree of linear polarization, $P'=(I'_\sigma-I'_\pi)/(I'_\sigma+I'_\pi)$ (where I' is the integrated intensity), was measured by selecting each of the two components independently by use of an analyzer with a 90° reflection.[12] Briefly, with the analyzer set to diffract in the scattering plane, the horizontal polarization σ was detected, while the vertical polarization π was measured with the analyzer diffracting at 90° to the scattering plane. Both a graphite (008) crystal with an x-ray energy of $E=10.45$ keV and a Ge(440) crystal with an energy of $E=8.766$ keV were used. We found that for **c** perpendicular to the scattering plane $P'=0.9\pm0.1$ for both the (100) and (200) reflections, whereas for **c** in the scattering plane $P'=0.9\pm0.1$ for (200) while $P'=-0.85\pm0.15$ for (100). This implies that at $H=0$ T the scattering at the (100) position, which first appears at $T_N=46.4$ K, is primarily if not completely of magnetic origin.

For measurements in the superconducting magnet where the **c** axis is always vertical, we find that the magnetic x-ray scattering intensity decreases gradually with increasing field, consistent with a local canting of the Mn^{2+} spins due to the applied field. Thus the scattering at nonzero fields must also be magnetic in origin as, of course, the group-theoretical arguments[11] require.

We now return to the data shown in the bottom of Fig. 3. In FC experiments at $H=5.6$ T there is a transition to LRO at ~41.3 K. However, as observed previously by Cowley *et al.*,[7] at ~10 K there is a first-order spin-flop transition to an XY phase. In our experimental geometry, the XY magnetic scattering is a factor of $\sin^2\theta$ (~100) times weaker than the Ising scattering and hence is unobservable. Indeed, this provides further evidence that the x-ray scattering in a field is entirely magnetic in origin since no such dramatic diminution in intensity would be expected for charge scattering. In Ref. 7 it is shown that the XY phase has LRO. Further, when one heats back up from the XY phase into the Ising region the entire crystal retains antiferromagnetic LRO. For our magnetic x-ray measurements this means that the entire illuminated volume must have LRO. Strikingly, in contrast to the data obtained at 4.7 T, there is now significant hysteresis with the FH peaks about 60% more intense than the FC peaks. We have observed this effect a number of times under different field and temperature-cycling conditions. The most reasonable explanation of these results is that only about 60% of the volume illuminated by the x rays *achieves LRO on field cooling into the Ising regime.* This would correspond to a negligible part of the total volume of the sample and it is for

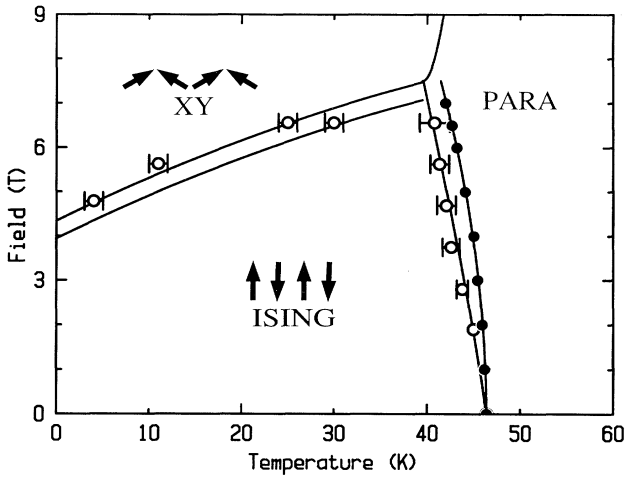

FIG. 4. $Mn_{0.75}Zn_{0.25}F_2$ phase diagram as determined by x rays and neutrons. In the Ising phase the spins are along the **c** axis, parallel to the external field. In the *XY* phase the spins "flop" into the plane perpendicular to the field. The open circles are LRO boundaries as determined by x rays; the solid circles represent the metastability boundary as determined by neutrons.

this reason that the neutrons do not observe the LRO component. The simplest assumption is that the FC LRO region corresponds to the outer 2 μm of the sample, although more complicated geometries are also possible.

From a series of measurements such as that shown in Fig. 3 we can obtain the overall magnetic phase diagram. The results so obtained are shown in Fig. 4. We include, in addition, the location of the Ising metastability boundary measured with neutrons. The phase boundaries overall agree well with those reported in Ref. 7. We note in Fig. 4 that the Ising metastability temperatures $T_M(H)$ measured with neutrons at NIST are consistently higher than the LRO temperatures $T_N(H)$ measured with x rays at NSLS; both, of course, have been measured on this same sample. Because of uncertainties in the magnetoresistance corrections of the resistance thermometer as well as the stability of the x-ray magnet temperature control system, the errors on the LRO $T_N(H)$ values are relatively large. This will be ameliorated in future experiments. Nevertheless, our preliminary results suggest that the onset of LRO occurs slightly below the metastability boundary with a temperature difference which increases progressively with increasing field. This is exactly the effect anticipated in Ref. 7 and by certain RFIM simulation studies.[13]

These experiments have opened up the possibility of studies of the $d=3$ RFIM in equilibrium. With various technical improvements it should be possible to perform experiments of critical-phenomena quality. Clearly, it is essential to understand why the relaxation to the LRO state occurs in the near-surface region of the sample but is entirely inhibited in the bulk. It is possible that surface imperfections nucleate the equilibrium state. Synchrotron-x-ray studies of $Mn_xZn_{1-x}F_2$ samples with varying surface morphologies as well as other systems such as $Fe_{1-x}Zn_xF_2$ are planned to explore these ideas. Studies of thin films should also provide valuable insights into this mysterious and entirely unexpected phenomenon.

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