Temporal Phase Transition in the Three-Dimensional Random-Field Ising Model

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(Received 4 February 1985)

High-resolution neutron-scattering measurements of the random-field Ising phase-transition behavior in $Mn_{0.75}Zn_{0.25}F_2$ are reported. An abrupt, apparently discontinuous, transition at $T_N(H)$ from history-independent to history-dependent behavior occurs; below $T_N(H)$ all states with length scales between the field-cooled value and infinity are stable for indefinite lengths of times; logarithmic decay of the domain size in time may be ruled out. No existing theory accounts for this transition behavior.

PACS numbers: 75.40.Dy, 75.10.Hk, 75.25.+z, 75.50.Ee

Despite a large amount of theoretical and experimental study, the behavior of the $d=3$ Ising model in a random field is still not properly understood.[1] There is now widespread acceptance of the theoretical arguments which give the equilibrium lower critical dimension $d_l=2$. However, experiments performed, using the suggestion of Fishman and Aharony[2] that a uniform field applied to a random antiferromagnet generates a random staggered field, have shown when $d=3$ Ising systems are cooled in a field, long-range order (LRO) is not achieved, and that at low temperatures the properties are history dependent.[1,3,4] Recently, theories have been developed[5] to reconcile the theoretical result that $d_l=2$ with the neutron-scattering observations by showing that there are large energy barriers to domain-wall motion in the presence of random fields, and that these will lead to very long relaxation times. In order to examine this problem in more detail we have performed high-resolution neutron-scattering measurements on the random antiferromagnet $Mn_{0.75}Zn_{0.25}F_2$. The results show the following: (i) The history dependence sets in at a well-defined phase boundary; (ii) any relaxation of the field-cooled state domain size just below that boundary is slower than logarithmic with time; (iii) when the sample is cooled in a field there appears to be a break in the slope of the correlation length against temperature at the phase boundary; and (iv) all states with length scales longer than the field-cooled value are stable indefinitely below $T_N(H)$; on heating these states undergo a discontinuous transition to the field-cooled state at the phase boundary. Results (i)–(iii) either cannot be explained or are not addressed by the current theories of the relaxation processes[5]; the data strongly suggest that there is a well-defined phase transition when the sample is field cooled and that at this transition the relaxation times become unmeasurably long.

The measurements were performed on a single crystal of $Mn_{0.75}Zn_{0.25}F_2$. $Mn_xZn_{1-x}F_2$ is a well-characterized diluted-antiferromagnet system in which the spins interact with predominantly Heisenberg exchange interactions but in which the dipolar forces break the continuous symmetry. The system therefore belongs to the Ising universality class but has very-low-energy spin-wave excitations which might be expected to relax the system to a state of thermodynamic equilibrium. Measurements on $Mn_xZn_{1-x}F_2$ in a field have already been reported[1,6,7] and at magnetic fields below the spin-flop boundary the behavior is very similar to that in the good Ising systems $Fe_xZn_{1-x}F_2$ and $Co_xZn_{1-x}F_2$. However, at low temperatures these latter systems are totally frozen so that they are not suitable for the study of relaxation characteristics, whereas the $Mn_xZn_{1-x}F_2$ system is not frozen. Specifically, if the samples are cooled in a field to a temperature well below $T_N(H)$ and the field removed, $Mn_xZn_{1-x}F_2$[6] recovers LRO whereas the good Ising systems[4] stay in the field-cooled state.

The present experiments are an extension of these earlier measurements to study the properties in the neighborhood of the phase boundary in detail. The excellent crystal was supplied by Shapira and was cut from the same boule as the one used for magnetization and thermal expansion measurements.[7] It has a mosaic spread of less than 0.02° and a spread in the concentration of the order of 0.15%. It was mounted with the c-axis vertical in a superconducting magnet and the temperature could be held constant to 0.02 K and reproduced with an accuracy of 0.07 K, while the magnetic field was varied between 0 and 7.8 T with an accuracy of 0.1 T. The neutron-scattering measurements were performed at the Brookhaven high-flux beam reactor. Most of the data were taken with a triple-axis spectrometer set for elastic scattering with 5-meV neutrons and horizontal collimation 20′, 5′, 10′, and 10′

from reactor to counter. As we shall discuss below, the consequent high-energy resolution of 0.02-meV half width at half maximum enabled us to isolate the static Lorentzian-squared component of $S(\mathbf{Q})$, and this greatly simplified the data analysis. The momentum resolution elements were 0.002, 0.0009, and 0.015 reciprocal-lattice units (half width at half maximum) along $\mathbf{Q}$, perpendicular in plane, and perpendicular out of plane, respectively.

Most of the scans were performed by variation of the wave vector transversely through the (100) magnetic lattice point; we shall discuss here primarily results for $H=5.0$ T although closely analogous data were obtained at other fields. We also shall limit our discussion to the behavior in the immediate vicinity of the paramagnetic-Ising boundary. Scans were performed primarily by cooling the sample in a field (FC) or by cooling in zero field (ZFC) to 8 K, raising the field, and then heating. Representative scans are shown in Fig. 1. It is evident that at 44.0 K the FC and ZFC procedures give identical results whereas at 43.4 K the profiles are drastically different. We shall present a quantitative analysis of these profiles below.

In Fig. 2 the intensity at the peak position $\mathbf{Q}=(1,0,0)$ and at the wing position $\mathbf{Q}=(1,-0.004,0)$ are shown as functions of temperature for the FC and ZFC procedures. The FC data show that on decreasing the temperature the (100) peak intensity increases monotonically while the $\mathbf{Q}=(1,-0.004,0)$ wing scattering has a broad maximum at ~ 43.7 K [$T_N(0)=46.0$ K] and is still quite large at 42.0 K. We note that for $H=5.0$ T the width remains broader than the resolution function down to 8 K. The ZFC data are quite different. The ZFC scans at low temperature show only a narrow resolution-limited peak corresponding to magnetic LRO; indeed, the profile is identical to that found in the $H=0$ Néel state. As found previously[1,4] in $Co_xZn_{1-x}F_2$ and $Fe_xZn_{1-x}F_2$, the amplitude of the ZFC Bragg peak is much less than that of the broader FC peak because the crystal is so perfect that extinction greatly reduces the intensity of the scattering when there is LRO. Between 8 and 43.0 K the ZFC peak intensity decreases slightly but the profile is essentially unchanged. Above 43.0 K diffuse tails appear in the ZFC profiles; this is evident in the ZFC scan at 43.4 K shown in Fig. 1. Between about 43.5 and 43.75 K the ZFC profiles change drastically; the peak intensity approximately doubles while the wing intensity rises linearly to the FC value. All ZFC and FC scans at and above 43.8 K are identical to within the errors.

It is evident that the loss of LRO from the ZFC state occurs very abruptly and that within our temperature resolution and concentration spread the phase transition temperature coincides with the temperature above which the FC and ZFC procedures give the same results. We have repeated these measurements several times using different pathways in the H-T plane; we find that the temperature above which there

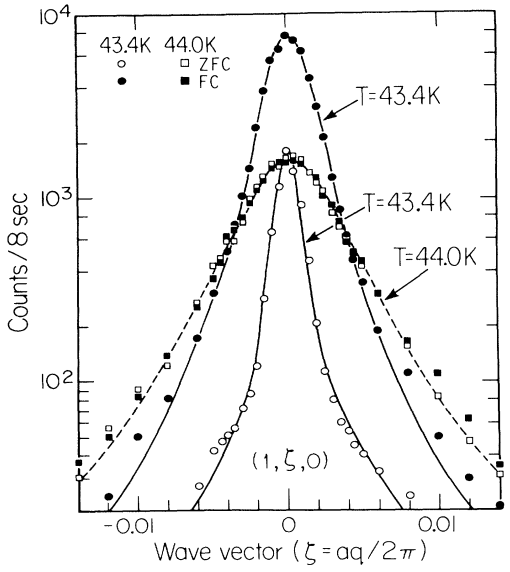

FIG. 1. Transverse scans at $H=5.0$ T, $T=44.0$ K following the FC and ZFC procedures. The solid lines are the results of least-squares fits to a Lorentzian-squared profile (plus Gaussian for the 43.4-K ZFC scan) as discussed in the text.

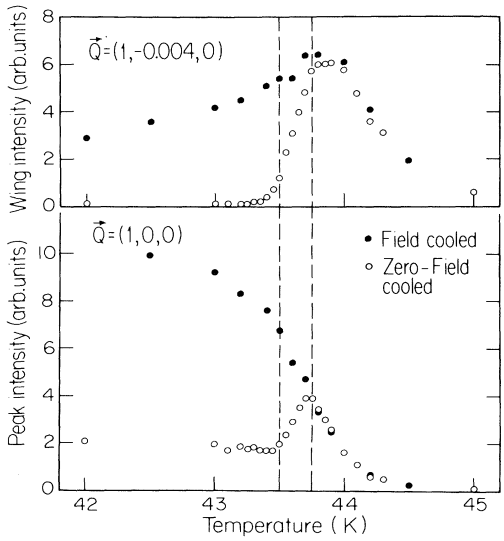

FIG. 2. Peak intensity and wing intensity for $H=5.0$ T for the FC and ZFC procedures in the neighborhood of the transition. The vertical dashed lines indicate the transition region.

is no history dependence is independent of the path of approach and agrees with the measurements of the thermodynamic phase boundary of Shapira, Oliveira, and Foner.[7] As is evident in Fig. 2, the width of the transition region at $H=5.0$ T is ~ 0.25 K. This is consistent with the width of ~ 0.1 K at $H=0$ measured by Shapira, Oliveira, and Foner[7] in their sample which was ~ 3 times smaller in volume than ours. We conclude, therefore, that within the spread due to the composition gradient, the loss of LRO occurs discontinuously and that above this boundary the sample is always in thermal equilibrium. Our previous results in other three-dimensional Ising systems[1,4] are consistent with this behavior, but because of experimental limitations it had not been possible to establish the nature of the transition clearly.

We have studied the behavior of the history-dependent states in some detail; we summarize here only the essential results. Briefly, as noted above, the FC results show that the width continues to narrow on cooling but the system never attains LRO. At all temperatures below $T_N(H)$ if the field is reduced after field cooling, the domains expand to the FC size at the terminal field, ultimately achieving LRO for $H \rightarrow 0$. However, if the field is increased at temperatures below $T_N(H)$, the profiles are unaltered; that is, for all temperatures below $T_N(H)$, the domains can expand but they do not then contract. Exactly this irreversibility is predicted by the dynamic theories of Ref. 5. As discussed in Ref. 4, the dynamic theories also account successfully for the empirical H^{-2} dependence of the FC domain size in two and three dimensions. An additional prediction of the dynamic theories of Villain[5] and of Grinstein and Fernandez[5] is that the domain size should increase logarithmically with time, that is, $K^{-1} \sim \ln(t/\tau)$, where τ is a microscopic time of order $\hbar/J \sim 10^{-11}$ sec. Careful scans, as a function of time at $H=7.0$ T for $T_N(7.0)-T=0.4$ K, were reported in Ref. 6. Least-squares fits of those data to the profile

$$S(\mathbf{Q}) = \frac{A}{K^2+q^2} + \frac{BK}{(K^2+q^2)^2}, \quad \mathbf{q}=\mathbf{Q}-\tau, \qquad (1)$$

where τ is the (100) magnetic Bragg reflection, give the results listed in Table I; this table makes quantitative the qualitative statements made in Ref. 6. The peak shapes are independent of time whereas the peak intensity increases by a few percent over many hours (cf. Ref. 3 where increases in peak intensity are assumed to imply a concomitant increase in domain size). The ratio of domain sizes at 5.4×10^4 and 6.0×10^2 sec is 1.01 ± 0.03 compared with a minimum ratio of 1.14 predicted by the logarithmic law. *Thus in $Mn_xZn_{1-x}F_2$ logarithmic expansion of the domain size with time is excluded.* As emphasized above, this is the most favorable case for relaxation compared with more anisotropic systems such as $Fe_xZn_{1-x}F_2$ where, away from the phase boundary, little or no relaxation occurs even when the field is removed.[4]

TABLE I. Fits to field-cooled data at $H=7.0$ T, $T=40.6$ K [$T_N(7.0)=41.0$ K].

Time (seconds)	B[a] (arbitrary units)	K[a] (10^{-3} reciprocal lattice units)
1.4×10^2	3.50 ± 0.12	1.46 ± 0.07
6.0×10^2	3.62 ± 0.04	1.39 ± 0.02
1.3×10^3	3.65 ± 0.04	1.37 ± 0.02
4.3×10^3	3.64 ± 0.04	1.35 ± 0.02
5.4×10^4	3.72 ± 0.04	1.38 ± 0.02

[a]The error bars are 1 standard deviation.

In order to discuss the transition region at $H=5.0$ T quantitatively, it is necessary to carry out detailed fits to the profiles. The form shown as Eq. (1) has successfully described the experimental results in $Co_xZn_{1-x}F_2$ and $Fe_xZn_{1-x}F_2$. Above $T_N(H)$, the Lorentzian term corresponds to the dynamic susceptibility fluctuations whereas the Lorentzian-squared term originates from the static ordered moments induced by the random field. We had anticipated that the 20-μeV resolution used in these experiments would serve to eliminate the Lorentzian term even well above $T_N(H)$, and this is indeed the case. Specifically, we find that for temperatures up to 47.0 K the FC profiles are well described by a pure Lorentzian-squared line shape ($\chi^2 \sim 1$) with an amplitude that varies smoothly through $T_N(H)$, thus confirming the above physical picture for the two terms. For the ZFC data below $T_N(H)$, the profiles were fitted to a resolution-limited Gaussian peak together with a broader Lorentzian-squared peak. Representative fits are shown as the solid lines in Fig. 1.

The results for the inverse correlation length K are shown in Fig. 3. We have included here data taken with $E=14$ meV neutrons with much coarser energy resolution; the analysis of those profiles included both terms in Eq. (1). The agreement between the K's determined with the two approaches is gratifying. Only data at temperatures below 45.5 K are explicitly displayed. We find that between 47.0 and ~ 44.5 K, K varies linearly with $T-T_0$ with $T_0 \sim 43.8$ K; the dashed line in Fig. 3 is the empirical linear law. Below 44.5 K, the points lie significantly above the linear line. Similar behavior has been observed previously[4] in $Fe_xZn_{1-x}F_2$. As expected, the FC and ZFC values for K agree above 43.75 K while below they differ. In the FC data, the slope of K vs T changes drastically

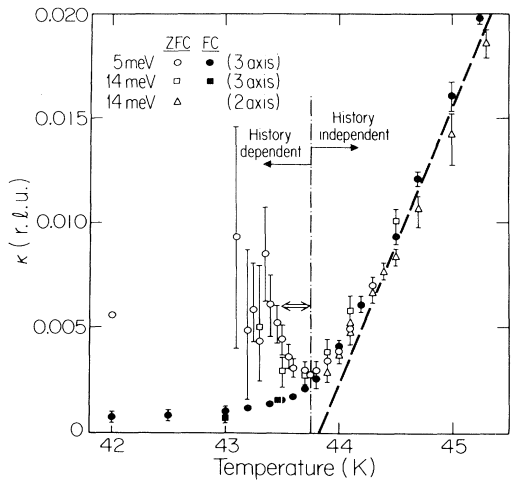

FIG. 3. Inverse correlation length K vs temperature at $H = 5.0$ T for FC and ZFC scans. The triple-axis values correspond to pure Lorentzian-squared profiles whereas the double-axis values result from fits to Eq. (1). The dashed line is a straight line through the data from 47.0 to 44.5 K. The double bar gives the size of the presumed composition gradient rounding.

from 43.75 to 43.5 K suggestive of a discontinuity in the slope. We note that below $T_N(H)$, K determined from the broad scattering in the ZFC scans decreases as the phase boundary is approached and reaches the FC value in the transition region. It is tempting to conclude that the abrupt transition out of the ZFC state is precipitated by critical fluctuations whose length scale is comparable to the FC domain size. Ideas along this line are contained in Villain's[5] heuristic discussion.

These experiments show that the physics of the random-field problem is more subtle than can be explained by current theories. The results in Fig. 3 suggest that, on cooling, the system is developing towards a long-range ordered state,[8] but that this evolution is preempted by a well-defined phase transition to a state with extremely long relaxation times and many metastable states. The long relaxation times would seem to require barriers much larger than those estimated in Ref. 5. This behavior is reminiscent of that in ordinary glasses and in spin-glasses. An alternative possibility is that the equilibrium three-dimensional random-field Ising transition in weak random fields is intrinsically first order but with the novel feature that the low-temperature state has anomalous lifetime characteristics. Clearly, continued experimental and theoretical work is required before we will obtain a proper understanding of the random-field Ising model. Full details of this work and of our results close to the spin-flop boundary will be published later.

We should like to acknowledge stimulating discussions with G. Aeppli, G. Grinstein, P. M. Horn, P. A. Lee, and Y. Shapira. We are grateful to A. Mak for his assistance with the analysis of the $H = 7.0$ T data. We are also grateful to Y. Shapira for providing us with the excellent crystal of $Mn_{0.75}Zn_{0.25}F_2$ and for communicating his results prior to publication. This work at Brookhaven was supported by the Division of Materials Sciences, U.S. Department of Energy, under Contract No. DE-AC02-76CH00016 and that at MIT by the National Science Foundation, Low Temperature Physics, under Grant No. DMR85-01856.

Note added.—After submitting this paper for publication we received a very interesting preprint from Young and Nauenberg[9] reporting Monte Carlo simulations of the random-field Ising model on lattices up to 64^3 in size. They also observe a discontinuous transition with pretransitional behavior above T_N similar to that reported here and in Ref. 4. There are some differences from our experiments which may originate either in finite-size effects in the simulations or from differences in metastability between the metamagnet and true random-field Ising-model problems.

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