Scaling of the Metastability Boundary of a $d = 2$ Random-Field Ising System

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The H and T dependence of the magnetic Bragg peak intensity has revealed a relatively sharp metastability boundary $T_F(H)$ in a $d=2$ random-field (RF) Ising system: $Rb_2Co_{0.85}Mg_{0.15}F_4$. Remarkably, $T_F(H)$ scales as $T_N - T_F(H) \propto H^{2/\phi}$, with the RF crossover exponent $\phi = 1.74 \pm 0.02$, and lies just inside the RF crossover region. Freezing must therefore be tied to RF critical behavior. The approach of the system to equilibrium at $T_F(H)$ proceeds *logarithmically* with time.

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It is widely believed that metastability is an inherent feature of the random-field Ising model (RFIM).[1] This is in keeping with pronounced time-dependent effects that have been observed in recent studies of RFIM systems.[2] However, current theories provide little guidance as to how the metastability *region* depends if at all, on dimensionality d, and how it relates to the scaling behavior observed at[3] or above[4] d_l, the lower-critical dimensionality of the RFIM[5] (now generally accepted to be $d_l = 2$). We report here on a neutron scattering study of a $d=2$ RFIM system: $Rb_2Co_{0.85}Mg_{0.15}F_4$, a diluted antiferromagnet (AF) in which a uniform field H generates a random field H_{RF} proportional to H. We have located, for the first time, the metastability boundary and found that it is a fairly narrow and well-defined region whose center $T_F(H)$ shifts with H relative to T_N as

$$T_N - T_F(H) \propto H^{2/\phi} \propto H_{RF}^{2/\phi}$$

with $\phi = 1.74 \pm 0.02$. This could only be the case if random-field crossover scaling governed the onset of metastability just as it does the shift in T_c, the crossover region, and, at $d=2$, the rounding of the phase transition. We have found that the approach to equilibrium is logarithmic in time, as most theories predict.[1] The crossover exponent ϕ is in excellent agreement with the pure $d=2$ Ising susceptibility exponent $\gamma = \frac{7}{4}$.

Metastability should accompany the hysteresis that is observable in neutron scattering experiments on $d=2$ or $d=3$ diluted AF.[6] It is found that domains are frozen in at low T in field-cooling (FC) but *not* in zero-field–cooling (ZFC) experiments. The domain structure, with a characteristic length varying roughly as H^{-2}, irrespective of d, manifests itself in the broadening of the AF Bragg peak which is otherwise resolution limited.

Here we focus on locating the metastability boundary and determining its dependence on H_{RF}. To accomplish this the system was initially prepared in a nonequilibrium state at low T. Then, as T was slowly increased, we monitored the evolution of the system. The point where equilibrium first occurs was established by noting the absence of hysteretic behavior above it.

Neutron scattering measurements were performed at the Brookhaven National Laboratory. We used the identical sample upon which birefringence (Δn) measurements[3] had established the destruction of the phase transition by H_{RF} at $d=2$; $Rb_2Co_{0.85}Mg_{0.15}F_4$. It is $4\times4\times6$ mm with the largest dimension along [001]. It was masked with cadmium, and exposed for only that portion ($\sim\frac{1}{2}$) for which Δn measurements had shown that the concentration was uniform to within 0.1% or better. The structure factor $S(q)$ was studied in the vicinity of the $(\frac{1}{2},\frac{1}{2},0)$ magnetic reflection and will be reported elsewhere. Here we are solely concerned with how the relative *intensity* $I(T,H)$ of the scattering at the Bragg peak depends upon the procedures used to arrive at a given T and H. $I(T,H)$ is a sensitive indicator of the presence of domains, since these both broaden $S(q)$ and decrease $I(T,H)$.

The sample was mounted on a copper block with a carbon-glass resistor to effect accurate and relatively field-independent temperature measurements with a stability of better than 10 mK. Overshoots of the final temperature setting could be kept to less than 20 mK. The c axis [001] was aligned parallel to H and data were taken at $H=0$, 1.00, 1.74, 2.65, 4.38, and 6.50 T.

In the AF state $I(T)$ is proportional to the square of the sublattice magnetization $M_0(T)$, except very close to T_N, as is shown in Fig. 1 by the curve labeled $H=0$. Very close to T_N ($T_N = 76.35$ K) scattering from critical fluctuations contributes to $I(T)$, both above and below T_N. This obscures the sharpness of the phase transition as monitored by $I(T)$.

At $d=2$ the AF state is the ground state only at $H=0$, but a metastable AF configuration was achieved

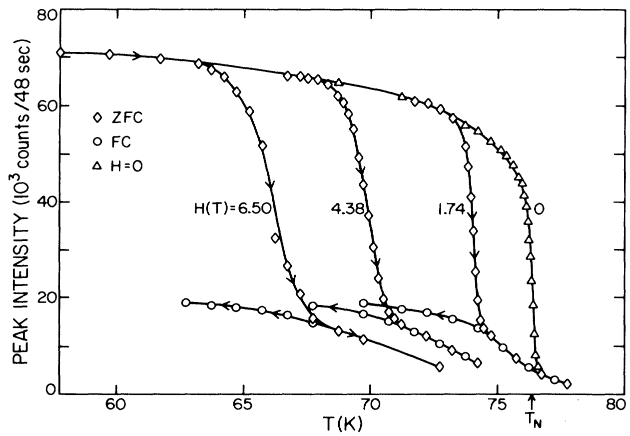

FIG. 1. AF Bragg peak intensity $I(T)$ vs T at $H=0$, and for zero-field cooling (ZFC) and field cooling (FC) at $H=1.74$, 4.38, and 6.50 T. The arrows indicate the direction in which data were taken.

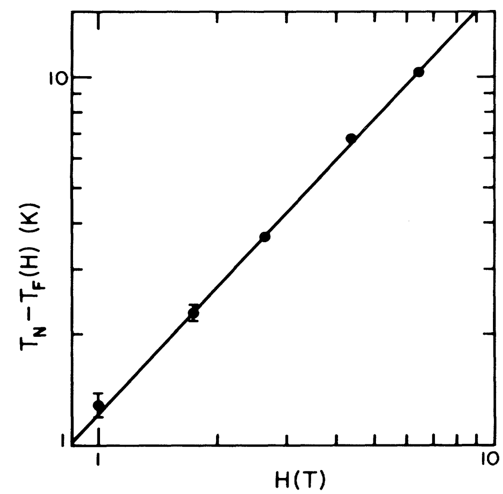

FIG. 2. Scaling behavior of $T_N - T_F(H) \propto H^{2/\phi}$. The metastability boundary $T_F(H)$ is defined as the point where $(-)dI(T)/dT$ is a maximum in the ZFC procedure. The slope $2/\phi$ yields the crossover exponent $\phi = 1.74 \pm 0.02$.

as follows: The sample was subjected to ZFC to a temperature T_i well below T_N. Keeping T_i fixed, the field was raised but *no change* was observed in $I(T_i)$, i.e., $I(T_i,H) = I(T_i,0)$, indicating that nonequilibrium AF conditions prevailed. As T was slowly increased (at constant H), $I(T)$ was seen to follow its $H=0$ dependence on T up to a rather well-defined T, whereupon $I(T)$ began to fall dramatically with increasing T. This continued until it joined (again at a rather well-defined T) the curve for $I(T)$ vs T that was obtained from a FC procedure at the *same* field H (see $H \neq 0$ curves in Fig. 1). Equilibrium prevails when the FC and ZFC routes to a given (T_a,H_a) result in identical values for $I(T_a,H_a)$. The FC curves of $I(T)$ exhibit a slow but monotonic increase with decreasing T with no sharp features and with $I(T,H)$ always well below $I(T,0)$, indicating a domain configuration. Over the range of T shown, the FC curves for $I(T)$ exhibited no apparent hysteresis. The ZFC and FC procedures were performed at each of five fields; curves for $I(T)$ vs T for only three of them are shown in Fig. 1. The width in T of the region over which the system evolves from metastability to equilibrium is relatively narrow and increases with increasing H.

An interesting feature of the ZFC $I(T)$ vs T curves is that the metastability boundary appears to approach T_N as H decreases. Hence one is led to examine its shift relative to T_N. We define a temperature $T_F(H)$ to be the point of maximum $-dI(T)/dT$ in a ZFC experiment. The difference $T_N - T_F(H)$ vs H is shown in a log-log plot in Fig. 2 for all five fields. Quite remarkably $T_N - T_F(H)$ not only exhibits power-law behavior but

$$T_N - T_F(H) = CH^{2/\phi}, \tag{1}$$

with $\phi = 1.74 \pm 0.02$.[7] This is precisely the form one would expect if RF crossover scaling governed the onset of metastability, just as it does the shift in T_c, the crossover region, and, at $d=2$, the rounding of the phase transition. If this is the case, ϕ should be equal to the susceptibility exponent $\gamma = \frac{7}{4}$ of the pure $d=2$ Ising system—and this is exactly what is observed! Both the temperature at which the ZFC and $H=0$ data first deviate from each other and the one at which the ZFC and FC data first coincide obey exactly the same scaling behavior as $T_F(H)$, which implies that the *width* of the metastability boundary also scales as indicated in Eq. (1).

That the scaling result of Eq. (1) describes the data so well leads to the surprising prediction that $T_F(H)$ exactly coincides with T_N when $H=0$. To test this idea, we cooled the sample in a field $H=6.5$ T to $T < T_F(6.5\ \text{T}) = 66.1$ K. $I(T)$ changed little upon lowering the field to zero (see Fig. 3), indicating that a *metastable* domain state was indeed frozen in. As T was then slowly increased, $I(T)$ decreased, remaining roughly proportional to $I(T)$ when cooled at $H=0$, but with its amplitude reduced by a factor of ~ 3.5. Only within a 0.5 K of T_N was any departure from this behavior observed. $I(T)$ then *increased* sharply, joining the $H=0$ curve essentially at T_N as shown in Fig. 3. Further increases in T resulted in $I(T)$ exactly duplicating the $H=0$ result. This confirms that $T_F(H=0)$ and T_N essentially coincide.

All measurements shown in Figs. 1 and 3 were made during the same length of time, about 3 min/point. In the ZFC studies we noticed that $I(T)$ exhibited some time dependence, if we attempted to repeat measurements in the vicinity of $T_F(H)$ where the slope of $I(T)$ vs T is large. No time dependence was seen ei-

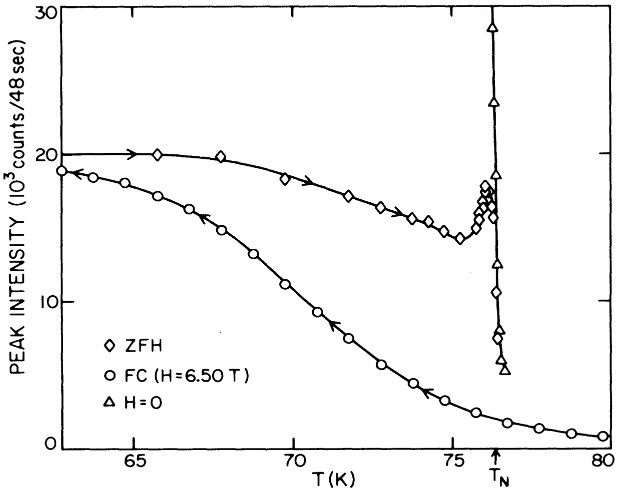

FIG. 3. $I(T)$ vs T, when first field cooled at 6.50 T, H then reduced to zero, and T subsequently increased (ZFH). The nearly vertical line is $I(T)$ vs T close to T_N with $H=0$.

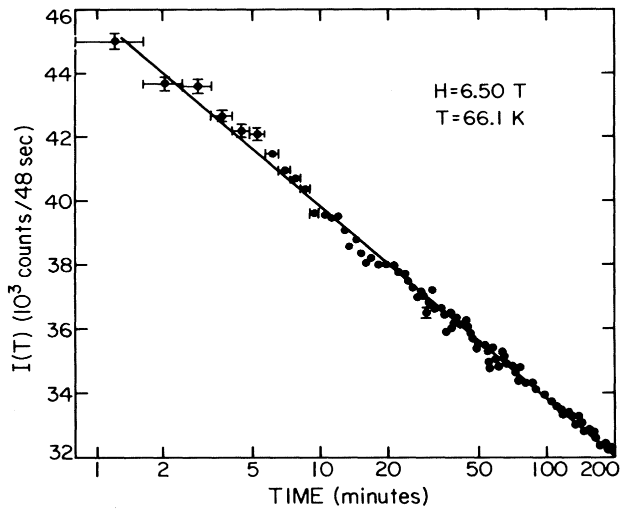

FIG. 4. Semilog plot of $I(T)$, at $T=66.1$ K and $H=6.50$ T, as a function of time, after ZFC. The horizontal bars represent the length of the counting interval.

ther well above or below this region. To study this time dependence at $T_F(H)$, the sample was subjected to ZFC to $T=66.1\ \mathrm{K}=T_F(6.50\ \mathrm{T})$. Then H was raised to 6.50 T as quickly as possible[8] and $I(T)$ was monitored repeatedly at 48-s intervals during a period of 200 min. The integrated $I(T)$ for each interval is shown in a semilog plot in Fig. 4 as a function of elapsed time, measured to the center of each interval. The horizontal bars on the points at early times are simply the 48-s interval widths. Points at later times represent averages over more than one interval. $I(T)$ is seen to decrease *logarithmically* with time over more than two decades. Logarithmic time dependence in the approach to equilibrium is characteristic of many current theories of nonequilibrium behavior in RFIM systems.[1]

Since $T_F(H)$ is found to exhibit the scaling properties associated with the RFIM critical behavior, the metastability boundary must be intimately connected with, and may be an integral part of, the RFIM critical behavior. With regard to the RFIM, Fishman and Aharony[9] showed that new critical behavior is expected within a crossover region

$$|t| < (ch_{\mathrm{RF}}^2)^{1/\phi}, \tag{2}$$

where $t=(T-T_\mathrm{N}+bH^2)/T_\mathrm{N}$ is the reduced temperature measured from the mean-field phase boundary $T_c^{\mathrm{MF}}(H)=T_\mathrm{N}-bH^2$ and c is a constant of order of unity. h_{RF}^2 is the reduced mean square random field; expressions for $h_{\mathrm{RF}} \propto H$ are given elsewhere.[3,4,10] In the birefringence study of the critical region[3] the coefficients corresponding to the shift of T_c and the rounding of the transition with H have been found to be $c=0.9^{+0.5}_{-0.4}$ and $c^*=5.4^{+2.6}_{-1.8}$, respectively. Thus the rounding of the transition is considerably larger than its shift. Using a similar analysis, we find that the lower (T_{cr}^-) and upper (T_{cr}^+) crossover boundaries, at which the $H\neq 0$ Δn data first deviate from those at $H=0$, yield coefficients $c_{\mathrm{cr}}^- = 45\pm 5$ and $c_{\mathrm{cr}}^+ = 30\pm 5$, respectively. However, $T_F(H)$ is found to scale with coefficient $c_F=29\pm 1$. Hence for the $d=2$ RFIM, $T_F(H)$ occurs slightly inside the random-field crossover region, whereas all of the "critical" behavior of the (destroyed) phase transition (i.e., the shift and rounding) occur well above $T_F(H)$, and thus take place in a region where thermal equilibrium is well established! The scaling of the various quantities is illustrated in Fig. 5, which is the most appropriate representation of a phase diagram for a RFIM system.

Since "freezing" occurs just below T_N (but well above $T=0$ K), neither by ZFC nor by FC can one access the Imry-Ma[11] $T=0$ K ground state of a $d=2$ RFIM system. It follows that one would not expect to see a field dependence of the domain size that is characteristic of the Imry-Ma state. Rather, the domains that one sees are governed by critical behavior. They have a size, for $T<T_F(H)$, characteristic of the equilibrium configuration at or near $T_F(H)$, and do not change significantly with decreasing T.

Lastly, it is worthwhile to contrast the behavior seen here at $d=2=d_l$ and that found at $d=3>d_l$ as regards freezing and the phase boundary. It has been shown[4] that a sharp phase transition $T_c(H)$ occurs in a $d=3$ RFIM system with critical behavior characteristic of lower effective dimensionality $\bar{d}\simeq 2$. If one accesses the transition region via FC, domains begin to *freeze at* T_F which is just above but very close to

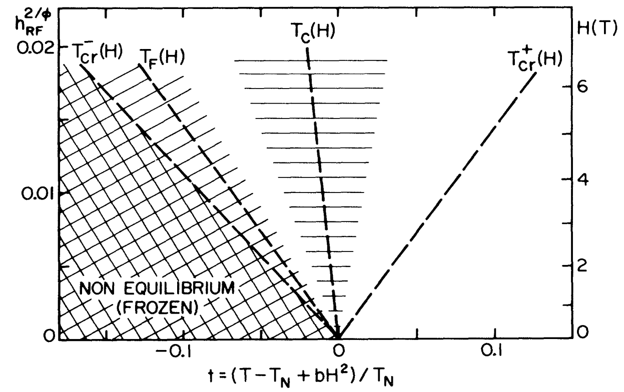

FIG. 5. New $d = 2$ RFIM "phase diagram." Shown are the scaling behavior of the (1) location $T_F(H)$ and width of the metastability boundary; (2) location $T_c(H)$ and width (indicated by shading) of the destroyed phase transition; and (3) random-exchange–random-field crossover boundaries $T_{cr}^+(H)$ and $T_{cr}^-(H)$. (2) and (3) are obtained from Δn measurements of the magnetic specific heat (Ref. 3).

$T_c(H)$. Thus a sharp transition at $T_C(H)$ may be seen only after ZFC. There is preliminary evidence that $T_F - T_N$ itself scales as $H^{2/\phi}$ and that below $T_c(H)$ the AF state is stable but the FC one is not.[12]

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7The bars appearing in Fig. 2 represent the errors in determining the centers of the region over which $I(T)$ changes abruptly, with an appropriate 90% confidence level. Drawing reasonable lines through these bars results in the quoted error in ϕ with an approximate $\frac{2}{3}$ confidence.

8Since a few minutes elapsed between the time when the time-dependent region was first entered, and the arrival at $H = 6.50$ T, the zero of time is uncertain. At most this would introduce *curvature* into the initial part of the log$t$ curve. However, none is seen in the data.

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