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Scaling of the equilibrium boundary of three-dimensional random-field Ising-model systems

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The onset of nonequilibrium behavior in three-dimensional random-field Ising-model (RFIM) systems ($Fe_xZn_{1-x}F_2$; $x=0.46$, 0.72) was studied via the capacitance method, using zero-field cooling, field cooling, and reverse-field cooling techniques, in fields $H \leq 100$ kOe. Equilibrium was found to occur only above a boundary $T_{eq}(H)$, which lies slightly *above* the sharp phase transition $T_c(H)$. Like $T_N - T_c(H)$, $T_N - T_{eq}(H) \propto H^{2/\phi}$, after mean-field corrections; $\phi = 1.40 \pm 0.05$ is the random-field crossover exponent. Thus, the onset of nonequilibrium is tied to RF critical behavior. The scaling property of $T_{eq}(H)$ follows directly from Villain's criterion for the breakdown of linear response in RFIM systems.

Nonequilibrium behavior (e.g., hysteresis and metastability) is an inherent feature of random-field Ising-model (RFIM) systems as is apparent from both experimental[1] and theoretical[2] studies. One of the most intriguing results obtained to date is the finding[3] that the metastability boundary $T_F(H)$, at the lower critical dimension d_l (Ref. 4) of the RFIM ($d_l = 2$), lies well *below* the destroyed phase transition "$T_c(H)$"; the latter determined from the rounded peak in the specific heat.[5] Moreover, it was observed that $T_N - T_F(H)$ scaled with the random field as $H_{RF}^{2/\phi}$, with $\phi = 1.74 \pm 0.02$, showing that RF crossover scaling governed the onset of metastability, since ϕ is predicted to be equal to $\gamma = \frac{7}{4}$, the $d = 2$ pure Ising susceptibility exponent.[3]

We define equilibrium to be established when the system no longer exhibits hysteretic behavior above a given $(T,H) \equiv T_{eq}(H)$, irrespective of the field-cycling procedure employed.

Where does equilibrium first establish itself for $d > d_l$ and, in particular, for $d = 3$, where a sharp phase transition has been observed?[6] Using the capacitance method, we provide here the first precise experimental answer to this question in the $d = 3$ RFIM systems $Fe_xZn_{1-x}F_2$ ($x = 0.46, 0.72$). Briefly stated, we find the equilibrium boundary $T_{eq}(H)$ to lie slightly *above* the sharp phase transition at $T_c(H)$ and that $T_N - T_{eq}(H) \propto H_{RF}^{2/\phi}$, as is $T_N - T_c(H)$, with $\phi = 1.40 \pm 0.05$, the $d = 3$ RFIM crossover exponent. We show the scaling property of $T_{eq}(H)$ follows directly from Villain's criterion for nonlinear response in RFIM systems.[2] Thus the nonequilibrium properties are intimately tied to RF critical behavior. The capacitance (C) method is an almost ideal technique for the measurement of $T_{eq}(H)$; it is simple, and exhibits pronounced hysteretic effects. Extremely small samples can be used, which circumvents, to a large degree, the problem arising from concentration gradients causing spurious rounding of the phase transition.[7] Since $T_{eq}(H)$ turns out to lie close to $T_c(H)$, the latter is an important consideration.

The capacitance method has been applied to magnetic critical behavior studies.[8] More recently,[9] it was used to measure the time dependence of the approach to equilibrium in a three-dimensional RFIM system which had been field-cooled into a metastable configuration. (Just below T_c such a state evolves logarithmically with time towards equilibrium.) At $H = 0$, C is proportional to the internal magnetic energy and hence dC/dT to the magnetic specific heat. For $H \neq 0$, there is, in addition, a sensitivity of C to terms both linear and quadratic in the external field H. Hence the behavior of C or dC/dT vs T in the region around $T_c(H)$ can be quite complicated and is not yet understood in detail.[10] Our sole concern here is with using dC/dT to establish $T_{eq}(H)$.

Three distinct field-cycling procedures were used in the present experiments: zero-field cooling (ZFC), field cooling (FC), and reverse-field cooling (RFC). ZFC and FC have been defined and employed previously[1,6] in connection with the RFIM problem but RFC has not, to our knowledge. In RFC, the system is cooled through $T_c(H)$ in a given field H to a temperature $T < T_c(H)$ and then the field is lowered through $H = 0$ and reversed in direction until some desired magnitude of field $|H'|$ is achieved. The configurations resulting from FC and RFC are metastable and *both* are observed to decay towards the ground state (ZFC configuration) as T approaches $T_c(H)$ from below.[9]

Measurements of C vs T were made on two crystals: $Fe_{0.46}Zn_{0.54}F_2$ and $Fe_{0.72}Zn_{0.28}F_2$. The former was cut from the most gradient-free mixed crystal that has been grown at the University of California at Santa Barbara (UCSB). From a birefringence scan of the entire crystal, we have determined the concentration gradient in the capacitance sample (disk shaped; diameter=0.9 cm, thickness=0.25 mm) to be less than 0.5% cm. This translates into a variation in T_N of $\delta T_N \leq 2.5 \times 10^{-4} T_N$. Although the $Fe_{0.72}Zn_{0.28}F_2$ crystal was not of comparable quality, a capacitor was made from it with its central section thinned to 0.05

mm in the direction of the concentration gradient. In this way a variation in $\delta T_N < 10^{-3} T_N$ was achieved in the thinned section as judged by dC/dT measurements at $H=0$ and in small fields.

In Fig. 1 we show dC/dT vs T in $Fe_{0.46}Zn_{0.54}F_2$ in the vicinity of $T_c(H)$ at a field of 15 kOe using the ZFC, FC, and RFC procedures. $T_c(H)$ is identified as that point at which dC/dT is a maximum after ZFC. It exactly coincides with the peak in $d(\Delta n)/dT$ that is observed upon ZFC in a sample cut from the same boule (Δn = birefringence). The peak seen upon RFC also appears sharp but is reversed relative to the ZFC peak. We presume the latter effect arises from the reversed moment, frozen in during the RFC procedure. Note that FC appears to dramatically broaden the transition.

Of primary importance is that only above a temperature $T_{eq}(H)$, which lies slightly *above* $T_c(H)$, are the results of the ZFC, FC, and RFC procedures identical for dC/dT vs T. Both $T_{eq}(H)$ and $T_c(H)$ vs H were measured in the field range $0 \leqslant H \leqslant 19$ kOe. A mean-field correction (bH^2) was applied to each for the nonrandom part of the shift in $T_c(H)$.[11] It has already been demonstrated from earlier birefringence and capacitance studies[6] that $T_N - T_c(H) - bH^2 \propto H^{2/\phi}$, with $\phi = 1.40 \pm 0.05$, the $d=3$ Ising random-field crossover exponent. Hence, in Fig. 2, we have plotted $T_c(H) + bH^2$ vs $H^{2/\phi}$, where for consistency we have chosen $\phi \equiv 1.40$. (The linearity of the fit is improved by a slight variation of ϕ, yet remaining within the errors quoted above.) Likewise, $T_{eq}(H) + bH^2$ vs $H^{2/\phi}$ is plotted in Fig. 2; its linearity shows that the equilibrium boundary also exhibits random-field crossover behavior and thus must be tied to random-field critical phenomena.

$T_c(H)$ and $T_{eq}(H)$ have also been determined in the crystal $Fe_{0.72}Zn_{0.28}F_2$ in the field range $0 \leqslant H \leqslant 100$ kOe.

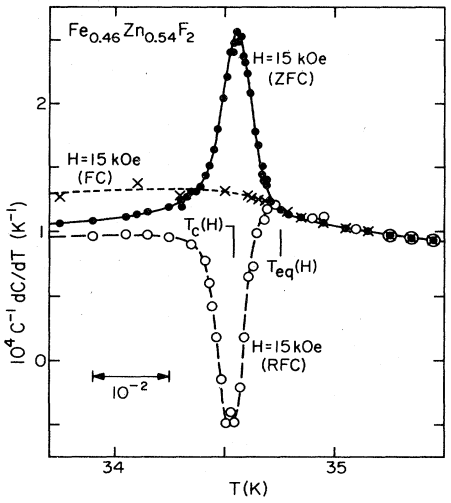

FIG. 1. Temperature derivative of the capacitance dC/dT vs T, in $Fe_{0.46}Zn_{0.54}F_2$ in ZFC, FC, and RFC field-cycling procedures, in a field of 15 kOe with $H \| c$ axis. Note that the transition at $T_c(H)$ appears sharp in ZFC and RFC but is clearly broadened during FC. $T_{eq}(H)$ is the temperature at which dC/dT, as obtained from a RFC and/or FC procedure, first becomes identical with the ZFC result. The concentration gradient in this C sample would result in a rounding of the transition at $H=0$ $\delta T_N \sim 2.5 \times 10^{-4} T_N \simeq 9$ mK. A reduced temperature interval $\Delta t = 10^{-2}$ is indicated.

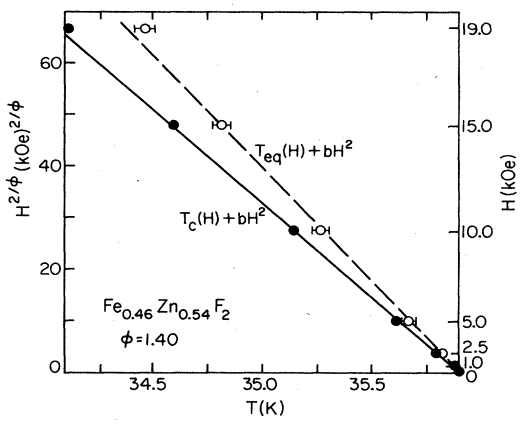

FIG. 2. Part of the "phase diagram" of the $Fe_{0.46}Zn_{0.54}F_2$ RFIM system. $T_c(H)$ and $T_{eq}(H)$ are plotted linearly vs $H^{2/\phi}$, after a mean-field correction is made to each. The value used for the random-exchange crossover exponent $\phi = 1.40$ was the one that had been determined in early birefringence studies (Ref. 6). The fields at which the measurements were made are shown on the right-hand-side ordinate axis.

dC/dT vs T at $H=80$ kOe is shown in Fig. 3 for the ZFC and RFC procedures and the points $T_c(H)$ and $T_{eq}(H)$ identified. Note that the ZFC procedure results in a sharp *dip* in dC/dT at $T_c(H)$ while the RFC one in this sample gives a peak, accompanied by a considerable time dependence below $T_c(H)$.[9,12]

Figure 4 shows $T_c(H) + bH^2$ and $T_{eq}(H) + bH^2$ plotted versus $H^{2/\phi}$, with $\phi = 1.40$. (As was the case above, a very slight variation in the choice of ϕ improves the linearity of the fit.) Again scaling is seen to be obeyed for both quantities.

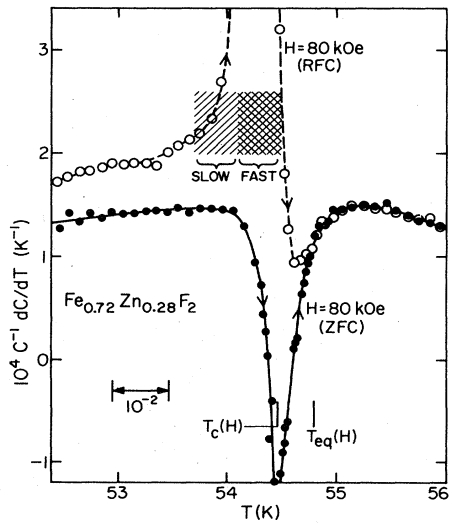

FIG. 3. dC/dT vs T in $Fe_{0.72}Zn_{0.28}F_2$, in ZFC and RFC field-cycling procedures, in a field of 80 kOe with $H \| c$ axis. The regions in temperature indicated as "slow" and "fast" qualitatively characterize the observable time dependence in C following RFC. (See discussion in Ref. 9.) A reduced temperature interval $\Delta t = 10^{-2}$ is indicated.

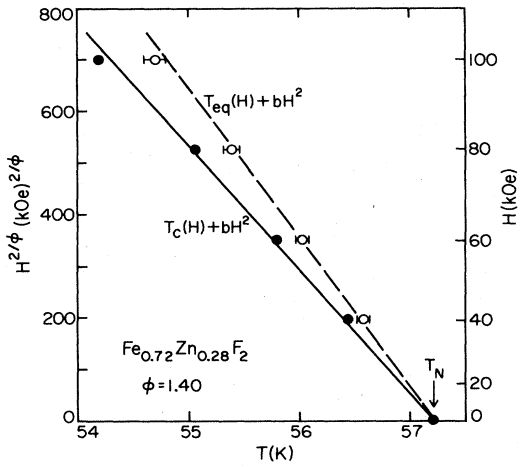

FIG. 4. Part of the "phase diagram" of the $Fe_{0.72}Zn_{0.28}F_2$ RFIM system. As in Fig. 2, $T_c(H)$ and $T_{eq}(H)$ vs $H^{2/\phi}$ is shown, after the mean-field correction is made to each. Again the value of $\phi=1.40$ was chosen to make the fit, as indicated above, for the $Fe_{0.46}Zn_{0.54}F_2$ crystal. The fields at which the measurements were made are shown on the right-hand-side ordinate axis.

One may calculate the quantity $R\equiv[T_{eq}(H)-T_c(H)]/[T_N-T_c(H)-bH^2]$ for the $x=0.46$ and $x=0.72$ RFIM systems. In both cases $R=0.16\pm0.03$. Because $R\ll 1$, $T_{eq}(H)$ lies well within the random-field crossover region and thus must be tied to RFIM critical behavior.

None of the theories referred to earlier[2] explicitly predicted that nonequilibrium behavior in RFIM systems would be governed by crossover scaling. However, we now show this to be implicit in Villain's model in which he estimates the metastable domain size at the phase boundary.

Villain[2] has argued that there is a "maximum value of the correlation length $1/\kappa$ *above* T_c beyond which linear-response theory cannot be applied," as T approaches $T_c(H)$. He shows this value of $1/\kappa$ to be of the same size as the linear dimension R_{mo} of the smallest metastable domain as $T\rightarrow T_c(H)$ from below, namely, $\kappa R_{mo}\sim 1$, with R_{mo} scaling with h_{RF} along the phase boundary as $R_{mo}\simeq h_{RF}^{-2\nu/\gamma}$, with h_{RF} the magnitude of the reduced rms random field. In the randomly diluted antiferromagnet (AF), $h_{RF}\propto H$ applied collinearly to the AF ordering.[11] Now κ has the general scaling form $\kappa\sim t^{\nu}f(th_{RF}^{-2/\phi})$, where $t\equiv(T-T_N+bH^2)/T_N$. If one suppresses the leading zero-field behavior this may be rewritten as[7]

$$\kappa\propto h_{RF}^{2\nu/\phi}g\left(\frac{t-t_c}{h_{RF}^{2/\phi}}\right) , \tag{1}$$

with ν the correlation length and $\phi=\gamma$ the susceptibility exponent of the random-exchange Ising model and where $t-t_c\equiv[T-T_c(H)]/T_N$ represents the relative departure from the actual phase boundary in any given field. It follows that the condition $\kappa R_{mo}\sim 1$ implies that $g((t_{eq}-t_c)/h_{RF}^{2/\gamma})\sim 1$, which can only be true if

$$t_{eq}-t_c\sim h_{RF}^{2/\gamma} . \tag{2}$$

But we know $T_N-T_c(H)-bH^2\sim H^{2/\gamma}$; hence it follows from (2) that

$$T_N-T_{eq}(H)-bH^2\propto H^{2/\phi} , \tag{3}$$

as we have observed.

Thus from this study and the recent one[3] on the $d=2$ RFIM system, the scaling properties of the nonequilibrium behavior of RFIM systems, at and above d_l, have been firmly established. In one case ($d=d_l=2$) the boundary which signals the onset of equilibrium lies *below* the destroyed transition "$T_c(H)$," while in the other ($d=3$) it lies *above* the sharp phase transition. At $d=2$, $T_F(H)$ denotes a fairly narrow region separating completely frozen metastable behavior below $T_F(H)$ (i.e., independent of changes in H or T and time) from equilibrium behavior above. Time dependence (log t behavior) is seen in the vicinity of $T_F(H)$. Since "$T_c(H)$" is above $T_F(H)$, the former can be studied in equilibrium. At $d=3$ the situation is more complex. If the system is FC, there is a frozen (nonequilibrium) region $T\ll T_c(H)$, a region of irreversible (but time-independent) behavior $T<T_c(H)$, a region of observable time dependence $T\leq T_c(H)$, and finally a boundary $T_{eq}(H)>T_c(H)$ above which no hysteretic behavior is observed.[13]

Although all of the theories[2] of metastability indicate nonequilibrium behavior should be observed in RFIM systems, it only becomes clear from the present experiments just how this comes about at $d=3$ and where the departure from equilibrium is first manifest. Since we have established $T_{eq}(H)>T_c(H)$, FC must, of necessity, trap the system into some nonequilibrium (metastable) domain configuration before the expected transition to long-range order would otherwise occur upon further cooling. Thus the FC procedure, as a means to access the ground state of the three-dimensional RFIM system, cannot work. The interpretation of the early neutron studies[13] mistook the nonvanishing width of the observed scattering arising from the finite size of the static, *nonequilibrium* configuration, beginning at $T\sim T_{eq}(H)$, for the failure of the critical scattering correlation length to diverge as would happen if the system were in *equilibrium* and the phase transition were truly destroyed (as is manifest at $d=d_l=2$, Ref. 5). By following the FC procedure Birgeneau, Cowley, Shirane, and Yoshizawa[14] concluded that $d_l\geq 3$, which we have shown to be incorrect.

We wish to thank J. L. Cardy for suggesting a general argument for relating Villain's condition on κR_{mo} to the scaling properties of κ along $T_c(H)$, and L. Schimmele for discussions on nonlinear response in RFIM systems. N. Nighman grew the crystals used in these experiments. Research at the University of California at Santa Barbara was supported in part by the National Science Foundation Grant No. DMR80-17582.

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[1]See, for example, Y. Shapira and N. F. Olivera, Jr., Phys. Rev. B **27**, 4336 (1983); P. Wong, J. Cable, and P. Dimon, J. Appl. Phys. **55**, 2377 (1984); H. J. Ikeda, J. Phys. C **16**, L21 (1983), and earlier references contained therein.

[2]J. Villain, Phys. Rev. Lett. **52**, 1543 (1984); R. Bruinsma and G. Aeppli, *ibid.* **52**, 1547 (1984); G. Grinstein and J. F. Fernandez, Phys. Rev. B **29**, 6839 (1984).

[3]D. P. Belanger, A. R. King, and V. Jaccarino, Phys. Rev. Lett. **54**, 577 (1985).

[4]J. Z. Imbrie, Phys. Rev. Lett. **53**, 1747 (1984).

[5]I. Ferreira, A. R. King, V. Jaccarino, and J. L. Cardy, Phys. Rev. B **28**, 5192 (1983).

[6]D. P. Belanger, A. R. King, V. Jaccarino, and J. L. Cardy, Phys. Rev. B **28**, 2522 (1983).

[7]D. P. Belanger, A. R. King, and V. Jaccarino, Phys. Rev. B **31**, 4538 (1985).

[8]A. R. King, D. P. Belanger, P. Nordblad, and V. Jaccarino, J. Appl. Phys. **55**, 2410 (1984).

[9]D. P. Belanger, S. M. Rezende, A. R. King, and V. Jaccarino, J. Appl. Phys. **57**, 3294 (1985).

[10]S. M. Rezende, A. R. King, and V. Jaccarino, J. Appl. Phys. **55**, 2413 (1984).

[11]S. Fishman and A. Aharony, J. Phys. C **12**, L729 (1979).

[12]The reversal in sign of the ZFC and RFC dC/dT peaks in this sample relative to that found in the $Fe_{0.46}Zn_{0.54}F_2$ crystal is not understood. We are currently investigating the dependence of dC/dT on Fe concentration, crystallographic orientation, and its relation to direct dilation measurements in connection with the RFIM problem.

[13]V. Jaccarino, A. R. King, and D. P. Belanger, J. Appl. Phys. **57**, 3291 (1985); see table and figures therein.

[14]See R. J. Birgeneau, R. A. Cowley, G. Shirane, and H. Yoshizawa, J. Stat. Phys. **34**, 817 (1984) for a review of the FC neutron-scattering studies; H. Yoshizawa, R. A. Cowley, G. Shirane, R. J. Birgeneau, H. J. Guggenheim, and H. Ikeda, Phys. Rev. Lett. **48**, 438 (1982), wherein it was established that the FC "result alone demonstrates unambiguously that $d_l \geq 3$."