PHYSICAL REVIEW B VOLUME 34, NUMBER 1 1 JULY 1986

Critical behavior of the magnetization of a $d=3$ random-field Ising system

W. Kleemann,* A. R. King, and V. Jaccarino
Department of Physics, University of California, Santa Barbara, California 93106
(Received 17 March 1986)

Optical Faraday rotation was used to measure the *uniform* "dc" magnetization M versus T in the random-field Ising model (RFIM) system $Fe_{0.47}Zn_{0.53}F_2$ in fields $0.2 \leq H \leq 1.9$ T. Both $(\delta M/\delta T)_H$ and $(\delta M/\delta H)_T$ diverge as $H^y \ln|t|$, as does the specific heat. Significantly different amplitude exponents were measured, $y \sim 0.6$ and 1.0, respectively, confirming scaling-theory predictions. The observed rounding of the phase transition below $|t| \sim 1\times10^{-3}$, at $H=1.0$ T, results from extreme slowing down of the RFIM critical fluctuations consistent with extrapolating $\chi'(\omega)$ studies to a measurement time scale $\tau=1/\omega\sim100$ s.

Since the lower critical dimension d_l of the random-field Ising model (RFIM) is now agreed to be $d_l=2$,[1] attention has shifted toward understanding the phase transition at $T_c(H)$ for $d>d_l$ (e.g., $d=3$). Both theory[2] and experiment[3] have begun to focus on the critical dynamics of the RFIM at $d=3$ because of the unusual nature of the slowing down of the critical fluctuations as $T\to T_c(H)$. The latter is almost certainly at the heart of the hysteresis and metastability observed in field-cycling studies.

Without exception, all experiments on RFIM systems have been performed on diluted uniaxial antiferromagnets (AF's) in a *uniform* field H parallel to the c axis, following the Fishman-Aharony prediction[4] of their equivalence to the Ising ferromagnet in a *random* field. The Faraday rotation (FR) measurements reported below were made on the diluted Ising AF $Fe_xZn_{1-x}F_2$, upon which there have been extensive birefringence (Δn),[5] capacitance,[6] neutron scattering,[7] NMR,[8] and ac susceptibility $[\chi'(\omega)]$ (Refs. 3 and 9) studies. It was from $\chi'(\omega)$-vs-ω studies[3] that the first experimental indication came of the extreme slowing down of the critical fluctuations as $T\to T_c(H)$. With FR measurements of $(\delta M/\delta H)_T$ vs T we have effectively increased the characteristic measurement time $(\tau=\omega^{-1})$ of the susceptibility by at least two orders of magnitude, relative to the lowest frequency (2 Hz) of the $\chi'(\omega)$ studies. The peak in $(\delta M/\delta H)_T$ appears considerably sharper and higher than that in $\chi'(\omega)$, but the effects of the unusual RFIM dynamics are still seen even at this extreme ω, at reduced temperatures $|t|\leq10^{-3}$ in $H=1.0$ T.

In addition, we have determined the field-scaling properties of the leading singularities of all second derivatives of the free energy with respect to T and H; $\partial^2F/\partial T^2$, $\partial^2F/\partial H\partial T$, and $\partial^2F/\partial H^2$ all diverge as $H^y|t|^{-\tilde{\alpha}}$ with the *same* $\tilde{\alpha}$, but *different* y. Previous experiments on $d(\Delta n)/dT$ (Ref. 5) and $\chi'(\omega)$ (Ref. 3) exhibit a $\ln|t|$ divergence (i.e., $\tilde{\alpha}\sim0$) for the RFIM. The FR measurements show this also to be the case for $(\partial M/\partial T)_H$ and $(\partial M/\partial H)_T$. Since $d(\Delta n)/dT$ is a linear combination of $\partial^2F/\partial T^2$ and $\partial^2F/\partial H\partial T$, it follows that the magnetic specific heat C_m, which as yet has not been directly measured in $Fe_xZn_{1-x}F_2$, must have a $\ln|t|$ divergence, too.

The measurements were made on a section of an extremely homogeneous boule, other parts of which have been used in previous investigations.[3,7-9] From the Néel temperature $T_N\equiv T_c(H=0)=36.47$ K, obtained from $\Delta n(T)$, we have determined the average composition to be $Fe_{0.47}Zn_{0.53}F_2$. Since the narrow laser beam (diameter 175 μm) is directed along the c axis (thickness 3.6 mm), which is *perpendicular* to the crystal growth direction, we believe the concentration gradient causes a rounding of the transition no larger than $\delta t=2\times10^{-4}$. Temperatures were stabilized to within 1 mK. A resolution of $\delta\Theta=0.002°$ was obtained for the FR angle Θ using a sensitive modulation method[10] in conjunction with a feedback-controlled rotating analyzer for compensation. It was found that Ar^+ ion laser light at $\lambda=457.9$ nm gave a considerable dispersion enhancement of the FR at short wavelengths[11] relative to that of the He-Ne laser ($\lambda=632.8$ nm). The initial FR data on FeF_2 were taken with the latter.

The temperature dependence of the FR in both FeF_2 and $Fe_{0.47}Zn_{0.53}F_2$ is shown in Fig. 1. The FR is strictly proportional to the uniform magnetization M in FeF_2. This can be seen in that it varies as does $\chi_\parallel(T)$ with T,[12] exhibiting Curie-Weiss behavior at high T, peaking slight-

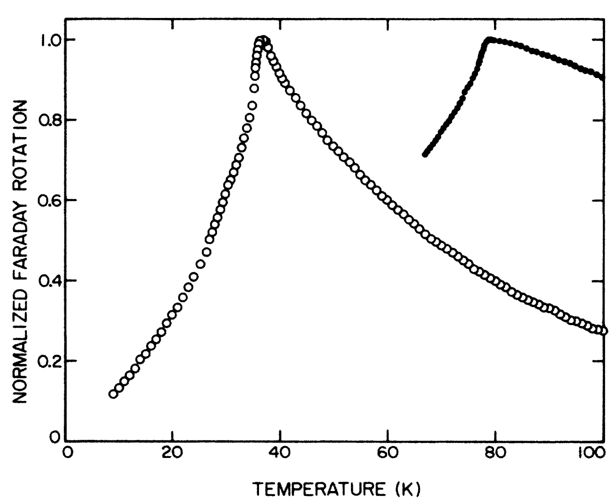

FIG. 1. Temperature dependence of the Faraday rotation at 1.0 T normalized to its peak value Θ_m in FeF_2 ($\lambda=633$ nm, $\Theta_m=49.0$ deg/cm at 79 K; full circles) and in $Fe_{0.47}Zn_{0.53}F_2$ ($\lambda=458$ nm, $\Theta_m=11.0$ deg/cm at 37 K; open circles).

 © 1986 The American Physical Society

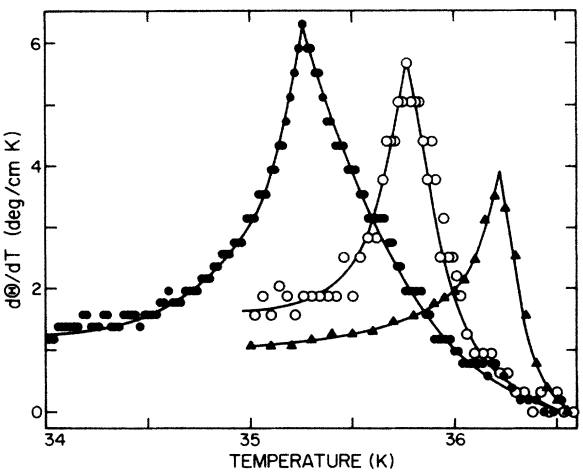

FIG. 2. $(\delta\Theta/\delta T)_H$ vs T for zero-field-cooled $Fe_{0.47}Zn_{0.53}F_2$ measured at $H=0.5$ T (triangles), 1.0 T (open circles), and 1.5 T (full circles), respectively. The solid lines are guides for the eye.

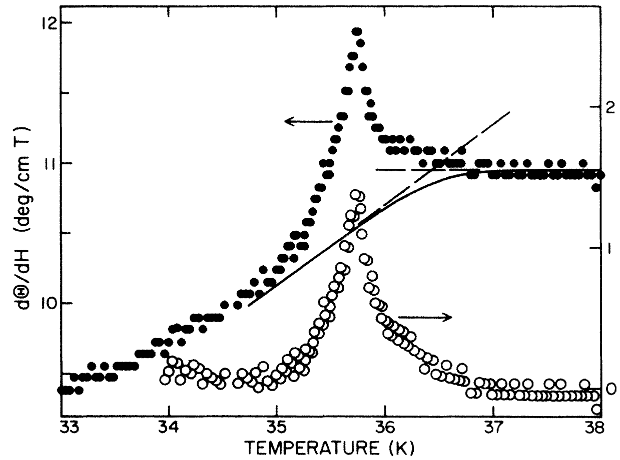

FIG. 4. $(\delta\Theta/\delta H)_T$ vs T for zero-field-cooled $Fe_{0.47}Zn_{0.53}F_2$ measured at $H=1$ T with $\delta H=0.03$ T (full circles, left-hand scale), and its singularity (open circles, right-hand scale) after subtracting a standard background function (solid line, see text).

ly above $T_N=78.4$ K, and monotonically vanishing as $T\rightarrow 0$.[13] The peak in the FR near T_N sharpens with dilution and decreases in temperature. Its amplitude decreases (at constant λ) by one order of magnitude. The variation of the FR with T appears to be identical with $\chi'(\omega)$ (Refs. 3 and 9) except in the region *very* close to $T_c(H)$.

Figure 2 shows $(\delta\Theta/\delta T)_H$ vs T in the vicinity of $T_c(H)$ for $H=0.5$, 1.0, and 1.5 T, respectively. Critical behavior is evident from the appearance of sharp peaks which, upon increasing H, become more symmetric and shift to lower T. A plot of $(\delta\Theta/\delta T)_H$ vs $\log_{10}|t|$ at $H=1.5$ T, as shown in Fig. 3, suggests that it diverges logarithmically as does $d(\Delta n)/dT$.[5] Furthermore, the peak position at $T_c(H)$ does obey crossover scaling [i.e., $T_N-T_c(H)\propto H^{2/\phi}$, after mean-field correction[4]]. In close agreement with previous results[5,6] we find $\phi=1.44\pm0.04$ from measurements within $0.2\text{ T}\leq H\leq 1.9$ T.

Very similar critical behavior appears in the "dc" susceptibility χ, as is manifest in $(\delta\Theta/\delta H)_T$ vs T for $H=1.0$ T in Fig. 4. It is obtained from individual $\Theta(T)$ vs T curves taken at $H\pm\delta H/2$, with $\delta H=0.03$ T. A symmetric peak appears on top of a large background, which is qualitatively similar to $\chi'(\omega)$.[3] Virtually no change of the peak shape and a mere 0.1% increase of its height is expected in the limit $\delta H\rightarrow 0$; hence, the identification of $(\delta M/\delta M)_T$ with χ. The singular part of $(\delta\Theta/\delta H)_T$ is plotted versus $\log_{10}|t|$ in Fig. 5. These data were obtained by subtraction of a smooth background, which would correspond to $\chi'(\omega)$ in the limit $\omega\rightarrow\infty$, as was done

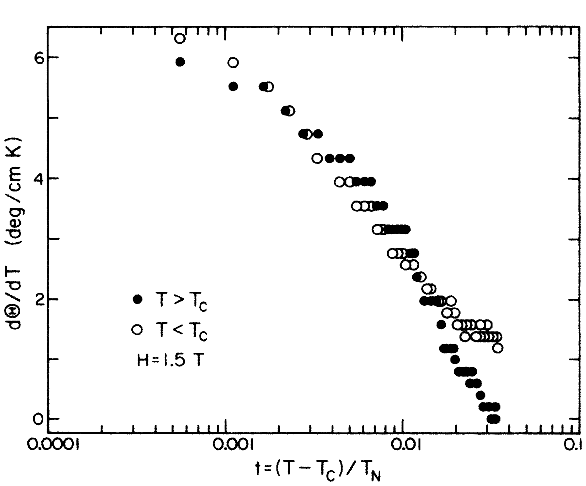

FIG. 3. $(\delta\Theta/\delta T)_H$ vs $\log_{10}|t|$ for $H=1.5$ T (cf. Fig. 2), where $t=(T-T_c)/T_N$ with $T_c=35.28$ K and $T_N=36.47$ K. Open circles: $T<T_c$, full circles: $T>T_c$.

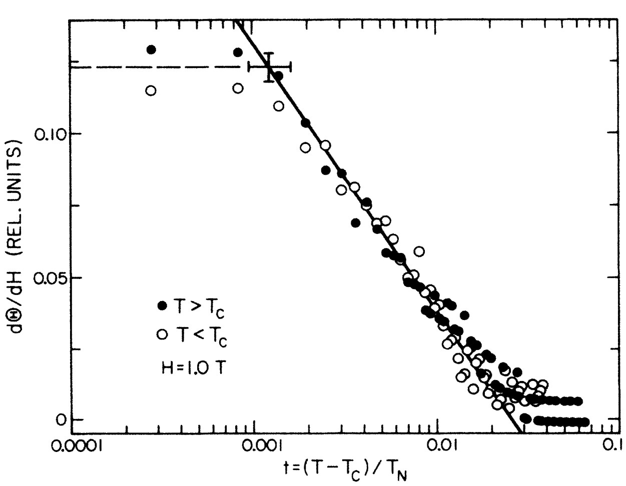

FIG. 5 Singularity of $(\delta\Theta/\delta H)_T$ vs $\log_{10}|t|$ normalized to $(\delta\Theta/\delta H)(37\text{ K})=10.95$ deg/cm T (Fig. 4). The horizontal line through the peak height and the $\log_{10}|t|$ line intersect at $t^*=1.3\times10^{-3}$ (see text). Open circles: $T<T_c$, full circles: $T>T_c=35.73$ K.

in the analysis of $\chi'(\omega)$.[3] It is hyperbola shaped with asymptotes tangent to $(\delta\Theta/\delta H)_T$ at $|t|>0.03$ and symmetrizes the singularity within $0.001<|t|<0.03$ (see Fig. 4). Again, within errors, a $\ln|t|$ behavior emerges as shown by the solid line best fitted to the data within $0.003 \leq |t| \leq 0.02$ (Fig. 5). The departure from $\ln|t|$ behavior below $|t| \lesssim 10^{-3}$ in both $(\delta\theta/\delta T)_H$ and $(\delta\Theta/\delta H)_T$ will be discussed below.

A critical-point analysis of both $(\partial M/\partial T)_H$ and $(\partial M/\partial H)_T$ may be obtained from the scaling form of the free energy,[4]

$$F(T,H)=H^{2(2-\alpha)/\phi}g(|t_H|H^{-2/\phi}) \ , \qquad (1)$$

in which the leading random-exchange behavior has been suppressed.[5] Here, $t_H=(T-T_N+bH^2)/T_N$, and α is the exponent at $H=0$. Assuming a sharp phase transition at $T_c(H)$, this transforms into

$$F(T,H) \propto H^{2(\tilde{\alpha}-\alpha)/\phi}|t|^{2-\tilde{\alpha}} \ , \qquad (2)$$

where $t=[T-T_c(H)]/T_N$ and $\tilde{\alpha}$ refer to the RFIM limit. A straightforward calculation shows that the leading singularities for $(\partial M/\partial T)_H$ and $(\partial M/\partial H)_T$ have $H^y \times |t|^{-\tilde{\alpha}}$ dependences with $y=(2/\phi)(1+\tilde{\alpha}-\alpha-\phi/2)=0.56$ and $y=(2/\phi)(2+\tilde{\alpha}-\alpha-\phi)=0.99$, respectively. The numerical values are obtained using $\tilde{\alpha}=0$ (in accordance with logarithmic divergence), $\phi=1.40$ (Refs. 5 and 7) and $\alpha=-0.09$.[14] They are satisfactorily confirmed by the experiments on $(\delta\Theta/\delta T)_H$ ($y=0.60\pm0.10$) and $\chi'(\omega)$ [$y\sim0.97$ (Ref. 9)]. C_m also scales as $H^y|t|^{-\alpha}$ with $y=(2/\phi)(\tilde{\alpha}-\alpha)=0.13$. In this case, direct experimental verification of y is lacking, but rather small values, $y\sim0.1$, have been inferred from the studies of $d(\Delta n)/dT$ vs H.[5] Since C_m involves both two-spin correlation functions and Zeeman terms, whereas $d(\Delta n)/dT$ involves only the former,[15,16] $d(\Delta n)/dT$ may be thought of as the *difference* between two quantities with identical thermal exponents but very different field-scaling exponents, $y=0.13$ and 1.56, respectively. Thus, $d(\Delta n)/dT$ would not have a pure power-law field-dependent amplitude. However, in the small-field limit ($H \ll H_{ex}$) it would scale with H as does C_m (i.e., $y=0.13$), as seems to be the case experimentally.[5]

The leveling off of both $(\delta\Theta/\delta T)_H$ and $(\delta\Theta/\delta H)_T$, which occurs at $|t|\sim10^{-3}$ (see Figs. 3 and 5), is well above where sample inhomogeneity would result in a rounding of the phase transition. We believe an explanation is to be found in the critical dynamics of RFIM systems, first seen in $\chi'(\omega)$ studies.[3] It might at first seem surprising that a "dc" experiment would be influenced by the dynamics that governs critical phenomena. However, a "dc" measurement is made on a time scale $\tau\sim10^2-10^4$ s. Since the RFIM is characterized by extraordinary slowing down of the critical fluctuations, effects of the dynamics manifest themselves even at these extremely long-time scales, i.e., at low frequencies $\omega=\tau^{-1}$. All of our data were taken approximately 100 s after any change of T or H; hence, $\tau\sim100$ s.

Following the interpretation given to the rounding of $\chi'(\omega)$,[3] we characterize the departure from the expected $\ln|t|$ divergence in terms of a "dynamic rounding temperature" $t^*(\tau)$. It is defined by the intersection of the asymptotic $\ln|t|$ behavior of the singularity with a horizontal line through the peak height at $|t|=0$, in a semilogarithmic plot. Analyzing the susceptibility (Fig. 5) in this way we obtain $t^*(100\text{ s})=(1.3\pm0.4)\times10^{-3}$. Adopting a conventional dynamical scaling approach, one finds $t^*(\tau)\propto\tau^{-1/\tilde{z}\tilde{\nu}}$, with $\tilde{z}$ the dynamical and $\tilde{\nu}$ the correlation-length RFIM critical exponents. The ω-dependent rounding of $\chi'(\omega)$ between $2\leq\omega/2\pi\leq1000$ Hz was fit[3] with $\tilde{z}\tilde{\nu}\sim14$. Extrapolating this to $\tau=100$ s at $H=1$ T yields $t^*=1.35\times10^{-3}$, which agrees well with our observation. In the same approach, the peak height of $\chi'(\omega)$ is expected to vary as $\ln\tau$. A similar extrapolation of the ac results to $\tau=100$ s yields a value of 0.132 ± 0.03 (relative to that of the flat plateau at 37 K), in essential agreement with our value of 0.124 ± 0.005 (Fig. 5), corrected for the finite δH effect.

An alternative activated dynamic scaling approach has been introduced by Fisher[2] for the RFIM, which predicts $t^*(\tau)\propto|\ln\tau|^{-1/\theta\tilde{\nu}}$, with $1\leq\theta\tilde{\nu}\leq2$. Making a corresponding extrapolation of the ac $t^*(\omega)$ data[3] using this expression yields a value $1.7\times10^{-3}\leq t^*(100\text{ s})\leq1.9\times10^{-3}$. Thus our data are also compatible with this approach, providing $\theta\tilde{\nu}\sim1$. Although our measurement at $\tau=100$ s extends the frequency range of $t^*(\tau)$ to six decades, the alternate scaling functions vary so slowly and are so similar, that the data cannot yet distinguish between them and other possible forms.[17] If this is to be done, better data, over a possibly even greater frequency range, will be required.

The existence of a dynamical rounding with $t^*(\tau)\sim10^{-3}$ for experimentally realizable measuring times ($\tau<10^4$ s) may render moot the whole question of what are the asymptotic static critical exponents[2] of the RFIM problem.

Useful discussions with D. P. Belanger, J. L. Cardy, M. Nauenberg, and A. P. Young are gratefully acknowledged. G. A. Gehring stimulated our interest in FR studies of the RFIM problem. This work was supported by National Science Foundation Grant No. DMR80-17582. W.K. would like to thank the Deutsche Forschungsgemeinschaft for research support.

*Permanent address: Angewandte Physik, Universität Duisburg, D-4100 Duisburg 1, Federal Republic of Germany.

[1]J. Z. Imbrie, Phys. Rev. Lett. **53**, 1747 (1985).
[2]D. S. Fisher, Phys. Rev. Lett. **56**, 416 (1986); A. T. Ogielski and D. A. Huse, *ibid.* **56**, 1298 (1986).
[3]A. R. King *et al.*, Phys. Rev. Lett. **56**, 2525 (1986).
[4]S. Fishman and A. Aharony, J. Phys. C **12**, L729 (1979).
[5]D. P. Belanger *et al.*, Phys. Rev. B **28**, 2522 (1983).
[6]A. R. King *et al.*, Phys. Rev. B **32**, 503 (1985).
[7]D. P. Belanger *et al.*, Phys. Rev. B **31**, 4538 (1985).
[8]C. Magon *et al.*, J. Magn. Magn. Mater. **54–57**, 49 (1986).
[9]J. A. Mydosh *et al.*, J. Magn. Magn. Mater. **54–57**, 47 (1986).

[10]F. A. Modine and R. W. Major, Appl. Opt. **14**, 761 (1975).

[11]A brief review on the theory of the FR focused onto AF is given by J. Pommier *et al.*, J. Phys. C **17**, 5621 (1984).

[12]S. Foner, in *Proceedings of the International Conference on Magnetism, Nottingham, England, 1964* (The Institute of Physics and The Physical Society, London, 1965), p. 438.

[13]The proportionality of the FR with M is confirmed by dispersion data on FeF_2 at 458 nm $< \lambda < 633$ nm, which suggest a typical (Ref. 11) $\lambda^{-2}/(\lambda_0^{-2}-\lambda^{-2})$ behavior with $\lambda_0 \sim 150$ nm.

[14]R. J. Birgeneau *et al.*, Phys. Rev. B **27**, 6747 (1983).

[15]G. A. Gehring (private communication).

[16]J. Ferré and G. A. Gehring, Rep. Prog. Phys. **47**, 513 (1984).

[17]Unfortunately, the peak height analysis, although, in principle, more precise than that of $t^*(\tau)$, is of little help, since activated dynamic scaling predicts (Ref. 2) a $\ln[\ln(\tau/\tau_0)]$ dependence without specifying a value for τ_0. Reasonable agreement with all data, ac (Ref. 3) *and* "dc" (this work), is found, if τ_0 is of order 10^{-8}–10^{-14} s.