Equilibrium Random-Field Ising Critical Scattering in the Antiferromagnet $Fe_{0.93}Zn_{0.07}F_2$

Z. Slanič and D. P. Belanger

Department of Physics, University of California, Santa Cruz, California 95064

J. A. Fernandez-Baca

Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393
(Received 1 September 1998)

It has long been believed that equilibrium random-field Ising model (RFIM) critical scattering studies are not feasible in dilute antiferromagnets close to and below $T_c(H)$ because of severe nonequilibrium effects. The high magnetic concentration Ising antiferromagnet $Fe_{0.93}Zn_{0.07}F_2$, however, does provide equilibrium behavior. We have employed scaling techniques to extract the universal equilibrium scattering line shape, critical exponents $\nu = 0.87 \pm 0.07$ and $\eta = 0.20 \pm 0.05$, and amplitude ratios of this RFIM system. [S0031-9007(98)08150-2]

Models for the statistical physics of phase transitions can be experimentally tested by measuring the universal critical parameters and comparing them with theoretical predictions. This is crucial for the understanding of difficult problems such as the random-field Ising model (RFIM). Although it is well known that a phase transition occurs [1] for the $d = 3$ RFIM, the characterization of the equilibrium critical behavior in its experimental realization, anisotropic randomly dilute antiferromagnets such as $Fe_xZn_{1-x}F_2$ in applied uniform fields [2], has not been possible near to and below the transition, $T_c(H)$, despite years of intense experimental investigation. This is primarily a result of the severe hysteresis in the scattering line shapes near to and below $T_c(H)$. Although long-range order is observed after cooling in zero field (ZFC) and heating through the transition, upon cooling through the transition in the field (FC) long-range antiferromagnetic order is never achieved, even at low temperatures. In either case, the line shapes are difficult to interpret. It was widely thought for many years that these nonequilibrium effects were unavoidable.

Since random-field effects increase with the dilution, as well as the strength of the applied magnetic field [3], most early studies focused on magnetic concentrations $x < 0.8$ in order to achieve, for reasonable H, a suitable range of reduced temperature, $t = [T - T_c(H)]/T_c(H)$, over which asymptotic critical behavior might be observed. However, nonequilibrium effects exist below the equilibrium temperature [4] $T_{eq}(H)$ which lies just above $T_c(H)$. Much effort was directed to understanding the behavior of the domains, both near to and below $T_c(H)$. Neutron scattering studies of the Bragg intensity in the bulk crystal [5] $Fe_{0.46}Zn_{0.54}F_2$ and the epitaxial film [6] of $Fe_{0.52}Zn_{0.48}F_2$ showed anomalous behavior even under ZFC caused by domain formation. It was shown that, at low T, RFIM dynamics are dominated by domains [7,8]. The dominance of domain effects near $T_c(H)$ was evident in ac susceptibility measurements [9]. From these studies over several years along with Monte Carlo (MC) simulations [10,11] it became clear that the severe nonequilibrium effects were primarily due to the large number of vacancies that are well connected and thus allow the formation of domain walls with insignificant energy cost.

Recently, we demonstrated [12] that the metastability problem can, in fact, be overcome by employing a magnetically concentrated crystal, $Fe_{0.93}Zn_{0.07}F_2$, in which the domain walls must break many magnetic bonds to form since the vacancies are well separated [13]. As a consequence, domains do not form under either FC or ZFC and the line shapes from both processes are found to be identical for all T and, hence, represent equilibrium behavior. Of course, the small dilution necessitates the use of large fields to probe the RFIM critical regime, so we used the highest field available with the present apparatus, $H = 7$ T. Attempts to analyze the equilibrium critical scattering in a preliminary study [12] failed because the sample had a concentration gradient [14] that limited analysis to $|t| > 10^{-3}$. Hence, we were able only to attempt an analysis using mean-field (MF) line shapes that yield values for the inverse correlation, κ, and staggered susceptibility, χ_s, that do not, however, follow power-law behavior in $|t|$ below $T_c(H)$. We have overcome these difficulties by using a thin crystal of thickness 0.44 mm, less than $1/10$ the original thickness, cut with its plane perpendicular to the concentration gradient. This has made it possible to extend the scattering measurements to $|t| > 10^{-4}$, greatly increasing the accuracy of the critical behavior analysis and, by making a few simple assumptions about the scaling properties of the scattering line shape as described below, we have been able to go beyond the inaccurate MF line shapes traditionally used in this type of analysis. With the use of the scaling techniques and the additional data from the thin crystal, we have finally achieved a consistent analysis of the critical behavior above *and below* $T_c(H)$ for the RFIM.

Neutron scattering measurements were made at the Oak Ridge National Laboratory High Flux Isotope Reactor using a two-axis spectrometer configuration. We used the (0 0 2) reflection of pyrolytic graphite (PG) at an energy of 14.7 meV to monochromate the beam. We mainly employed two different collimation configurations. The lower resolution, primarily used for the large sample, is with 70 arc min before the monochromator, 20 before the sample and 20 after the sample. Primarily for the thin sample, we made scans with 10 arc min before and after the sample. PG filters were used to eliminate higher-order scattering. The carbon thermometry scale was calibrated to agree with recent specific heat results [15] for the $H = 0$ transition. The field dependence of the thermometry was also calibrated. All scans used in this report are transverse about the (1 0 0) antiferromagnetic Bragg point.

The observed line shape is given by $S(q)$ convoluted with the instrumental resolution. In MF [1]

$$S(q) = \frac{A}{q^2 + \kappa^2} + \frac{B}{(q^2 + \kappa^2)^2} + M_s^2 \delta(q), \quad (1)$$

where $\kappa = \kappa_o^{\pm} |t|^{\nu}$, $+$ and $-$ are for $t > 0$ and $t < 0$, respectively, and M_s is the staggered magnetization. The first term represents $\chi_s(q)$ and the second two the disconnected susceptibility, $\chi_s^{\mathrm{dis}}(q)$. In principle, the amplitudes A and B are constant in T and the same above and below $T_c(H)$, although in practice this is not true since real systems do not follow MF behavior. For translationally invariant systems, $B = 0$. For real systems MF is inadequate since we must have the scaling behavior $\chi_s(q) \propto \kappa^{\eta-2} f(q/\kappa)$ with the limits $\chi_s(q) \propto \kappa^{\eta-2}/(1 + q^2/\kappa^2)$ for $|q| \ll \kappa$ and $\chi_s(q) \propto q^{\eta-2}$ for $|q| \gg \kappa$. For the $d = 3$ pure transition, $\eta \approx 0.03$. Hence, the MF Lorentzian line shape is adequate except very close to $T_c(H)$, as explicitly shown [16] for FeF$_2$ where the corrections to MF are important for $|t| < 10^{-3}$, particularly for $T < T_c(H)$. For smaller $|t|$, the $t < 0$ Tarko-Fisher [17] (TF),

$$f(q/\kappa) \propto \frac{(1 + \phi'^2 q^2/\kappa^2)^{\sigma+\eta/2}}{(1 + \psi' q^2/\kappa^2)(1 + \phi''^2 q^2/\kappa^2)^{\sigma}}, \quad (2)$$

and the $t > 0$ Fisher-Burford [18] (FB),

$$f(q/\kappa) \propto \frac{(1 + \phi^2 q^2/\kappa^2)^{\eta/2}}{1 + \psi q^2/\kappa^2}, \quad (3)$$

approximants, where ϕ, ϕ', ϕ'', and σ are fixed, and $\psi = 1 + 1/2\eta\phi^2$ and $\psi' = 1 + 1/2\eta\phi'^2 + \sigma(\phi'^2 - \phi''^2)$, were found to be excellent for the analysis and yielded exponents in superb agreement with theory [1].

The dilute case with $H = 0$ corresponds to the random-exchange Ising model for which the value of η is similarly small and the MF line shape proved adequate for $|t| > 10^{-3}$. For $x = 0.46$, the scattering results [19] $\nu = 0.69 \pm 0.03$ and $\gamma = 1.33 \pm 0.02$ agree remarkably well with recent MC results [20] $\nu = 0.684 \pm 0.005$ and $\gamma = 1.34 \pm 0.01$.

In a previous scattering work [12], we found two important results using Eq. (1) to fit the data for Fe$_{0.93}$Zn$_{0.07}$F$_2$ for $H = 7$ T. First, the results for κ and χ_s are independent of whether the FC or ZFC procedures were used; i.e., the line shapes are equilibrium ones at all T. Second, the values for κ and χ_s obtained from the line shape analysis could be fit to a power law in $|t|$ for $T > T_c(H)$, but not $T < T_c(H)$. For $T > T_c(H)$, $\nu = 0.90 \pm 0.01$, $\gamma = 1.72 \pm 0.02$, and $\bar{\gamma} = 3.0 \pm 0.1$ were obtained. The lack of power-law behavior for κ and χ_s below $T_c(H)$ indicates that the MF line shapes are inadequate, particularly for $T < T_c(H)$. This is not surprising, since the predicted $d = 3$ RFIM exponent $\eta \approx 0.5$ is quite large [1], and a large deviation from MF behavior is therefore to be expected, particularly below $T_c(H)$, at $|t|$ much larger than in the pure FeF$_2$ case.

Since we lack a theoretically predicted line shape for data analysis, we instead invoke the scaling form [1]

$$S(q) = A^{\pm}\kappa^{\eta-2} f(q/\kappa) + B^{\pm} A^{\pm 2}\kappa^{\bar{\eta}-4} g(q/\kappa), \quad (4)$$

for $|q| > 0$, where $\gamma = \nu(2 - \eta)$ and $\bar{\gamma} = \nu(4 - \bar{\eta})$. This expression is still too complicated to use for scaling of the data, particularly since the scaling in this unusual case involves two perhaps independent functions. To proceed, we make two assumptions strongly motivated by results of Monte Carlo simulations [21,22], high temperature expansions [23], and previous experiments [24] at $x = 0.6$, namely, $\bar{\eta} = 2\eta$ and $g(q/\kappa) = f^2(q/\kappa)$. With these assumptions, we have

$$S(q) = A^{\pm}\kappa^{\eta-2} f(q/\kappa)[1 + B^{\pm} A^{\pm}\kappa^{\eta-2} f(q/\kappa)], \quad (5)$$

for $|q| > 0$. Finally, we employ the TF and FB approximants for $f(q/\kappa)$ except that we let ϕ, ϕ', ϕ'', and σ be fitting variables. The line shape analysis includes a small fixed constant term and one that is linear in q to account for background counts.

For $H = 0$, the random-exchange Ising model case, we set $B = 0$. Folding the resolution corrections into the scaling line shape, we fit all of the data simultaneously over the range $1.14 \times 10^{-4} < |t| < 10^{-2}$ for the small sample and $1.15 \times 10^{-3} < |t| < 10^{-2}$ for the large one. We obtain the critical parameters $\nu = 0.70 \pm 0.02$ and $\gamma = 1.34 \pm 0.06$, which are in good agreement with previous results using the MF equation cited above.

The same procedure is followed for $H = 7$ T, the RFIM case, where we now let B vary. The fitted parameters are given in Table I for $|t| < 10^{-2}$ and $|t| < 3 \times 10^{-3}$. The lower limits for $|t|$ are the same as for $H = 0$. In Fig. 1 we plot, versus q/κ, the intensity data after subtracting the background, deconvoluting with the instrumental resolution, and dividing by $A^{\pm}\kappa^{2-\eta}[1 + B^{\pm} A^{\pm}\kappa^{2-\eta} f(q/\kappa)]$. The collapse of the data onto $f(q/\kappa)$ is excellent. We find no evidence of systematic deviations of the data from the scaling function in any of the scans used in the analysis. The collapse

TABLE I. Values found from fits at $H = 7$ T. Data for the large sample were fit for $|t| > 1.15 \times 10^{-3}$ and for the small sample were fit for $|t| > 1.14 \times 10^{-4}$. κ is given in reciprocal lattice units (rlu).

Parameter	$|t| < 10^{-2}$	$|t| < 3 \times 10^{-3}$
T_c (fixed)	$70.61 K$	$70.61 K$
η	0.20 ± 0.05	0.16 ± 0.06
ν	0.88 ± 0.05	0.87 ± 0.07
A^+	10.0 ± 0.2	9.21 ± 0.3
A^-	6.15 ± 0.14	4.45 ± 0.15
κ_0^+	1.13 ± 0.04 rlu	0.95 ± 0.17 rlu
κ_0^-	3.24 ± 0.11 rlu	2.78 ± 0.5 rlu
B^+	$(4.7 \pm 0.1) \times 10^{-5}$	$(3.00 \pm 0.13) \times 10^{-5}$
B^-	$(4.0 \pm 0.3) \times 10^{-5}$	$(8.0 \pm 1.0) \times 10^{-5}$
σ	0.67 ± 0.5	0.86 ± 0.6
ϕ	0.16 ± 0.04	0.08 ± 0.01
ϕ'	0.39 ± 0.25	0.36 ± 0.3
ϕ''	0.31 ± 0.25	0.26 ± 0.2
χ^2	3.07	2.3
No. pts.	2444	1000

of the data onto a single scaling function can occur only if the critical exponents are chosen appropriately. By the quality of the scaling, we have demonstrated that by moving beyond the simple MF line shape, it is possible to fit the RFIM data for high magnetic concentration to obtain
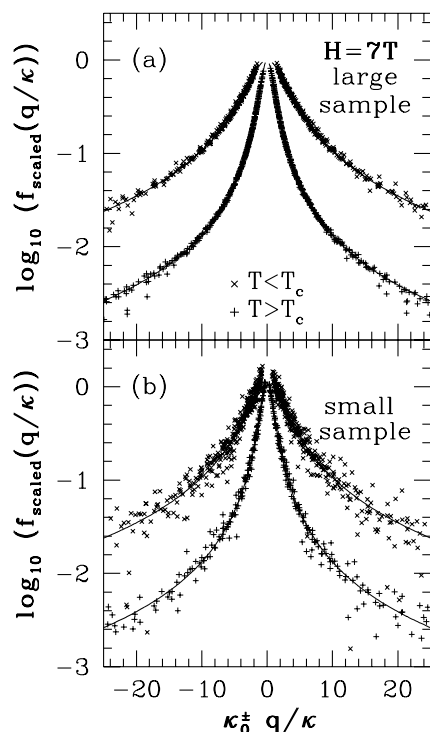

FIG. 1. Scaled neutron scattering data taken at different temperatures at $H = 7$ T collapsed onto the universal function $f(q/\kappa)$. The scatter in the small sample data is larger due to a smaller number of counts obtained in the thin sample. The fit was made for $|t| < 0.01$.

the critical parameters and a good representation of the actual scattering line shape. Note that these values, obtained for data above and *below* $T_c(H)$, agree rather well with the exponents obtained with the MF expression for this system above $T_c(H)$. The curves in Fig. 1, given by the TF and FB parameters in Table I, represent the experimental RFIM scaling functions that can be used to test future theoretical and simulation results.

Numerous simulation and series expansion studies yield critical exponents for the RFIM. The recent Monte Carlo results of Rieger [21] are $\nu = 1.1 \pm 0.2$, $\gamma = 1.7 \pm 0.2$, $\eta = 0.5 \pm 0.05$, $\bar{\gamma} = 3.3 \pm 0.6$, $\bar{\eta} = 1.03 \pm 0.05$, $\beta = 0.0 \pm 0.005$, and $\alpha = -0.5 \pm 0.2$. Similarly, Hartmann and Nowak [22], using exact ground state simulation techniques, find $\nu = 1.14 \pm 0.10$, $\beta = 0.02 \pm 0.01$, and $\bar{\gamma} = 3.4 \pm 0.4$. Note that the values for ν and γ agree fairly well with the results of the neutron scattering experiments [using $\gamma = \nu(2 - \eta)$]. However, η is larger than the value from the neutron study. The value of β has not been accurately determined experimentally. The largest discrepancy [25] is with α which appears to be close to zero since the experiments at all concentrations exhibit symmetric, nearly logarithmic peaks in the specific heat [1]. This disagreement has not yet been fully explained [22].

The equilibrium exponents obtained in the present experiments can be compared with previous results at lower concentrations. Belanger, King, and Jaccarino [24] obtained $\nu = 1.0 \pm 0.15$, $\gamma = 1.75 \pm 0.20$, $\bar{\gamma} = 3.5 \pm 0.3$, and $\eta \approx 1/4$ using a sample of $Fe_{0.6}Zn_{0.4}F_3$ and the MF equation. Only data above the equilibrium boundary [4], $T_{eq}(H)$, were used. These results are in good agreement with those obtained using the MF line shapes in $Fe_{0.93}Zn_{0.07}F_2$. This agreement indicates that the corrections to MF are not very significant above $T_c(H)$ but are very large for $T < T_c(H)$.

In contrast, Feng *et al.* [26] obtain $\nu = 1.5 \pm 0.3$, $\gamma = 2.6 \pm 0.5$, and $\bar{\gamma} = 5.7 \pm 1.0$ for $Fe_{0.5}Zn_{0.5}F_3$. This discrepancy, however, may be the result of the analysis of data which may lie below $T_{eq}(H)$, where hysteresis prevents a study of the equilibrium critical behavior. This seems apparent [27] when comparing these authors' results with previous studies [32] of $Fe_xZn_{1-x}F_2$. The present study at high magnetic concentration does away with metastability effects entirely for the critical scattering line shape analyses. This has the particular advantage of allowing fits to the data on both sides of the transition and much closer to the transition, greatly narrowing possible interpretations of the data and yielding much more reliable critical behavior parameters.

In summary, we have characterized the critical scattering below $T_c(H)$ in the high magnetic concentration crystal $Fe_{0.93}Zn_{0.07}Zn_2$. This has not been possible until now because of the domain formation present in the more diluted systems previously studied. We have shown that the line shapes are adequately described by using simple

scaling hypotheses. The possibility still exists that the exponents obtained in $Fe_{0.97}Zn_{0.07}F_2$ are effective ones that may change somewhat with fields much larger than $H = 7$ T. Future efforts will be directed to scattering measurements to determine ν, γ, and β and birefringence and Faraday rotation measurements to determine the specific heat critical behavior for $H \gg 7$ T at high magnetic concentration. To avoid extinction effects, the Bragg scattering exponent determination requires future experiments either using neutron scattering on a film [6] or using magnetic x-ray surface scattering.

[1] For reviews, see D.P. Belanger and A.P. Young, J. Magn. Magn. Mater. **100**, 272 (1991); D.P. Belanger, *Spin Glasses and Random Fields,* edited by A.P. Young (World Scientific, Singapore, 1998), p. 251; T. Nattermann, *Spin Glasses and Random Fields,* edited by A.P. Young (World Scientific, Singapore, 1998), p. 277.

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

[3] J.L. Cardy, Phys. Rev. B **29**, 505 (1984).

[4] A.R. King, V. Jaccarino, D.P. Belanger, and S.M. Rezende, Phys. Rev. B **32**, 503 (1985).

[5] D.P. Belanger, V. Jaccarino, A.R. King, and R.M. Nicklow, Phys. Rev. Lett. **59**, 930 (1987).

[6] D.P. Belanger, J. Wang, Z. Slanič, S.-J. Han, R.M. Nicklow, M. Lui, C.A. Ramos, and D. Lederman, Phys. Rev. B **54**, 3420 (1996).

[7] S.-J. Han, D.P. Belanger, W. Kleemann, and U. Nowak, Phys. Rev. B **45**, 9728 (1992).

[8] M. Lederman, J.V. Selinger, R. Bruinsma, R. Orbach, and J. Hammann, Phys. Rev. B **48**, 3810 (1993).

[9] Ch. Binek, S. Kuttler, and W. Kleemann, Phys. Rev. Lett. **75**, 2412 (1995).

[10] M. Staats, U. Nowak, and K.D. Usadel, Phase Transit. **65**, 159 (1998).

[11] S.-J. Han and D.P. Belanger, Phys. Rev. B **46**, 2926 (1992).

[12] Z. Slanič, D.P. Belanger, and J.A. Fernandez-Baca, J. Magn. Magn. Mater. **177–181**, 171 (1998).

[13] E.P. Raposo and M.D. Coutinho-Filho, Phys. Rev. B **57**, 3495 (1998).

[14] The sample is the same one used in a previous specific heat study [15].

[15] Z. Slanič and D.P. Belanger, J. Magn. Magn. Mater. **186**, 65 (1998).

[16] D.P. Belanger and H. Yoshizawa, Phys. Rev. B **35**, 4823 (1987).

[17] H.B. Tarko and M.E. Fisher, Phys. Rev. B **11**, 1217 (1975).

[18] M.E. Fisher and R.J. Burford, Phys. Rev. **156**, 583 (1967).

[19] D.P. Belanger, A.R. King, and V. Jaccarino, Phys. Rev. B **34**, 452 (1986).

[20] H.G. Ballesteros, L.A. Fernandez, V. Martin-Mayor, A.M. Sudupe, G. Parisi, and J.J. Ruiz-Lorenzo, Phys. Rev. B **58**, 2740 (1998).

[21] H. Rieger, Phys. Rev. B **52**, 6659 (1995).

[22] A.K. Hartmann and U. Nowak (to be published).

[23] M. Gofman, J. Adler, A. Aharony, A.B. Harris, and M. Schwartz, Phys. Rev. B **53**, 6362 (1996).

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

[25] U. Nowak, K.D. Usadel, and J. Esser, Physica (Amsterdam) **250A**, 1 (1998).

[26] Q. Feng, Q.J. Harris, R.J. Birgeneau, and J.P. Hill, Phys. Rev. B **55**, 370 (1997).

[27] Feng *et al.* adopt two transition temperatures. One is the so-called [28,29] "trompe l'oeil pseudocritical temperature," a phenomenological parameter not considered relevant to the critical behavior. These authors obtain what they consider to be the true transition, "$T_N(H)$," from fits to data above another temperature, "$T_M(H)$." For $H = 0$, $T_N = 37.6$ K, indicating [30] $x \approx 0.48$. From previous studies [5] at $x = 0.46$ for $H \leq 3$ T, we can extrapolate that $T_N - T_{eq}(H) \approx 6.2$ K for $H = 5$ T (taking into account the small MF shift). This should be slightly smaller for $x = 0.48$ and, thus, $T_{eq}(H) > 31.4$ K. The three data points below $T_{eq}(H)$ should be excluded from the fit, leaving only two above that lie within the critical region $|t| < 0.1$. However, data down to "$T_M(H)$"= 30.5 K were included. The resulting fit yielded "$T_N(H)$" $\approx$ 27.2 K. The difference between the fitted "$T_N(H)$" and "$T_M(H)$" is 3.4 K [31]. This difference should be less than $T_{eq}(H) - T_c(H) = 1.1$ K obtained for $x = 0.46$ [5]. By fitting data too low in T, "$T_N(H)$" is found much too low which forces ν, for example, to be too large. At larger H more of the critical region is excluded from equilibrium, making critical behavior studies difficult. Previous studies [24] conducted at low H are consistent with our present results.

[28] R.J. Birgeneau, Q. Feng, Q.J. Harris, J.P. Hill, A.P. Ramirez, and T.R. Thurston, Phys. Rev. Lett. **75**, 1198 (1995).

[29] D.P. Belanger, W. Kleemann, and F.C. Montenegro, Phys. Rev. Lett. **77**, 2341 (1996).

[30] C.B. de Araujo, Phys. Rev. B **22**, 266 (1980).

[31] Note that if the difference between the transition and equilibrium temperatures were, in fact, 3.4 K, this would essentially cover the entire critical region $|t| < 0.1$ since $T_N(0) = 37.6$ K, and, hence, no equilibrium critical analysis would be possible.

[32] I.B. Ferreira, A.R. King, and V. Jaccarino, Phys. Rev. B **43**, 10797 (1991).