Crossover from random-exchange to random-field critical behavior in $Fe_xZn_{1-x}F_2$

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Aharony's prediction of the inequivalence of the random-exchange to random-field crossover exponent ϕ to the random-exchange susceptibility exponent γ has been tested experimentally with a precise neutron scattering measurement of γ in $Fe_{0.46}Zn_{0.54}F_2$. We find $\phi/\gamma = 1.08 \pm 0.05$, which is in keeping with his estimate of $1.05 \leq \phi/\gamma \leq 1.1$.

Recently Aharony[1] has shown that the exponent ϕ which governs the crossover from random-exchange to random-field critical behavior should *not* be identical to the random-exchange susceptibility critical exponent γ, as would be the case for nonrandom systems, where the corresponding exponents *are* identical. In particular, for the $d=3$ random-field Ising model (RFIM), his rough renormalization-group calculations yield a ratio $\phi/\gamma = 1.05$, although he believes it might well be as large as 1.1.

We report here on a new, precise measurement of γ obtained from zero-field neutron scattering studies performed on the $d=3$ anitferromagnet $Fe_{0.46}Zn_{0.54}F_2$. This is the same "super" crystal upon which recent capacitance,[2] birefringence,[3] and susceptibility[4] RFIM studies have been made. From all of the studies of the shift in T_c with H, in this and other $Fe_xZn_{1-x}F_2$ crystals, a value of $\phi = 1.42 \pm 0.03$ is obtained. Combining this with the new value of $\gamma = 1.31 \pm 0.03$, one finds $\phi/\gamma = 1.08 \pm 0.05$, which is in keeping with Aharony's prediction.

The RFIM (i.e., the pure ferromagnet in a random field), first introduced by Imry and Ma,[5] remained a theoretical curiosity until Fishman and Aharony[6] demonstrated that one could generate random fields in a randomly diluted anitferromagnet (AF) by applying a *uniform* field H. All subsequent experimental studies[7-9] have been performed on the diluted AF. Cardy[10] showed that the site-diluted AF, in the weak-H limit, could be mapped onto the RFIM; and hence the asymptotic critical behavior of the two systems must belong to the same universality class.

However, the pure Ising ferromagnet with *no* random field and the diluted Ising AF at $H=0$ have different fixed points and therefore different critical exponents [i.e., the $d=3$ Ising model and the $d=3$ random-exchange Ising model (REIM) in the respective cases]. Because of this, the crossover from pure Ising to RFIM behavior and from REIM to RFIM behavior will be governed by *different* crossover exponents. For the pure Ising model in a random field h it has been shown[6] that $\phi \equiv \gamma$, where γ is the $h=0$ susceptibility critical exponent defined by

$$\chi \sim |t|^{-\gamma}. \tag{1}$$

Until recently, it was generally assumed that a similar equality would obtain if random exchange dominated the $H=0$ critical behavior, i.e., $\phi=\gamma$, and early experiments seemed to bear this out.[11,12] However, Aharony[1] has just shown that in the presence of random exchange the inequality

$$\phi > \gamma \tag{2}$$

holds, and he has estimated that $1.05 \leq \phi/\gamma \leq 1.1$.

A test of the Aharony inequality requires a precision measurement of ϕ and γ. The value of ϕ was obtained from the measured shift in T_c with H:

$$T_c(H) - T_N = -AH^{2/\phi}, \tag{3}$$

after correcting for a small mean-field shift and the very slight field dependence of the thermometer, on several samples of $Fe_xZn_{1-x}F_2$ ($x = 0.46, 0.60, 0.72$). Within experimental error, the capacitance (C), birefringence (Δn), and susceptibility measurements,[2-4] all yield the same value for ϕ, showing ϕ to be independent of x in the measured range of x. The value of $\phi = 1.42 \pm 0.03$ was obtained from averaging the best measurements made with the C with Δn techniques on the three crystals. The measurement of γ is obtained from quasielastic neutron scattering as described below.

All experiments on the critical behavior of random systems are extremely sensitive to the sample quality. Neutron scattering studies are particularly so because of the relatively large sizes required, typically a few millimeters in each dimension. Since for $Fe_xZn_{1-x}F_2$ $T_N(x) \simeq xT_N(1)$ for $x > 0.4$, gradients in x cause variations of T_N through the sample, thereby smearing the intrinsically sharp transition. The $Fe_{0.46}Zn_{0.54}F_2$ crystal is the result of an intensive effort to improve and characterize sample quality by the authors and co-workers; it has only a 2×10^{-4} variation in concentration over the entire volume. This "super" crystal has allowed critical-behavior scattering studies to be made down to $|t| = 10^{-3}$, free from concentration-gradient effects.

The neutron scattering data were obtained using a two-axis spectrometer configuration at the High Flux Isotope Reactor of the Oak Ridge National Laboratory. The $Fe_{0.46}Zn_{0.54}F_2$ crystal is a 2-mm-thick disk 8 mm in diameter with the c axis in the plane of the disk. The crystal was mounted with the c axis vertical on a copper holder

masked with Gd paint to eliminate scattering from the copper. A carbon thermometer and control heater were mounted on the copper holder. The sample and holder were in vacuum, thermally shielded by an aluminum can. Sample temperature was controlled to 0.01 K near $T_N \simeq 35.7$ K. The (002) reflection of Be was used to monochromatize the beam at 2.35 Å (14.8 meV). A pyrolytic graphite filter reduced higher-order reflections. Horizontal collimations were 20′ of arc before the monochromator, 10′ between the monochromator and the sample, and 10′ between the sample and the detector. The lattice parameters of $Fe_{0.46}Zn_{0.54}F_2$ are $a=b=4.70$ Å and $c=3.24$ Å. The resulting spectrometer resolution was determined by scanning the magnetic (100) Bragg reflection at $T=32.1$ K in the transverse, longitudinal, and vertical directions. Because the crystal is of high quality, the scans were well described by Gaussian line shapes, with half widths at half maximum of 0.00075, 0.003, and 0.02 reciprocal-lattice units (rlu), respectively.

For the pure Ising AF FeF_2 the scattering line shape is well described,[13] for $10^{-3} < |t| < 10^{-1}$, by the mean-field expression for the structure factor

$$S(\mathbf{q}) = \frac{A^+}{q^2+\kappa^2} \text{ for } T > T_N,$$
$$S(\mathbf{q}) = \frac{A^-}{q^2+\kappa^2} + m_s^2\delta(q) \text{ for } T < T_N, \tag{4}$$

where m_s is the staggered magnetization. The Lorentzian terms represent the susceptibility $\chi(\mathbf{q})$ and the δ function represents the magnetic Bragg reflection which may be removed from consideration by excluding data with very small $|\mathbf{q}|$.

For the REIM, which applies to $Fe_{0.46}Zn_{0.54}F_2$ for $H=0$, $S(\mathbf{q})$ is expected[14] to differ from the Lorentzian form in Eq. (4) for $T < T_N$ since the lack of translational invariance destroys the strict proportionality between $S(\mathbf{q})$ and $\chi(\mathbf{q})$ for $\mathbf{q} \neq 0$. No satisfactory theory has yet been developed to provide a suitable replacement for that given in Eq. (4). Since we observe no direct evidence for non-Lorentzian scattering, we have analyzed the data using Eq. (4) with the spectrometer resolution properly folded into it. Pelcovits and Aharony[14] suggest that the susceptibility amplitude ratio χ_0^+/χ_0^- determined in this manner might be too small because of additional scattering below T_N. In any case, the determination of critical exponents is much less dependent on the precise line shape used in the data analysis.

A typical transverse scan $(1,q,0)$ is shown in Fig. 1 for $T < T_N$. Data were taken for $0 \leq |\mathbf{q}| \leq 0.45$ rlu but are shown for $0.005 \text{ rlu} \leq |\mathbf{q}| \leq 0.14$ rlu, and the Bragg peak therefore does not appear. The solid curve is the best fit to the Lorentzian line shape corrected for spectrometer resolution. The inverse correlation length κ was determined from the widths of the Lorentzian fits as a function of temperature, and the correlation length exponent ν was obtained from power-law fits to $\kappa_0^{\pm}|t|^{\nu}$. Extrapolating to $q=0$, we evaluate the staggered susceptibility from

$$\chi(0) = \chi_0^{\pm}|t|^{-\gamma} + B = A^{\pm}\kappa^{-2}.$$

The values of $A^{\pm}\kappa^{-2}$ vs $|t|$ are shown in Fig. 2 along

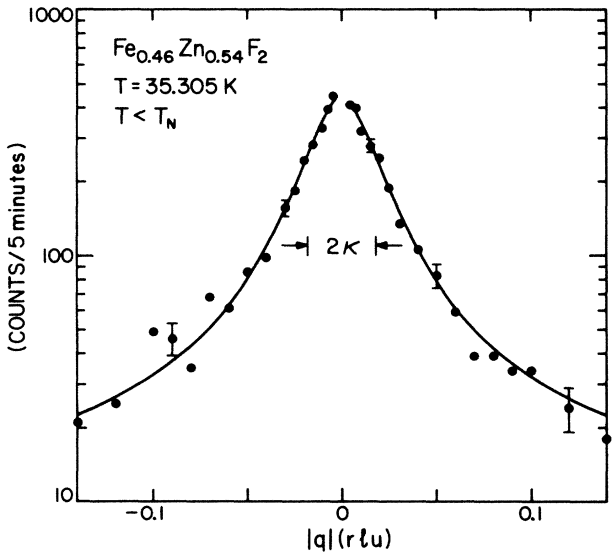

FIG. 1. Transverse scan along $(1,q,0)$ below T_N. The elastic scattering of the Bragg peak ($|q| < 0.005$) has been removed. The solid line is a Lorentzian fit to the critical scattering.

with the best fit to $\chi(0)$ that includes a nonsingular background term B. The resulting parameters for $1.5\times10^{-3} \leq |t| \leq 10^{-1}$ for ν, γ, κ_0^+/κ_0^- and χ_0^+/χ_0^- are shown for $Fe_{0.46}Zn_{0.54}F_2$ in Table I along with theoretical predictions. These new results are in much better agreement with theoretical predictions than those obtained in earlier studies[12] (the amplitude ratios are not as well determined theoretically as are the exponents). The exponents and amplitude ratios obtained with more restrict-

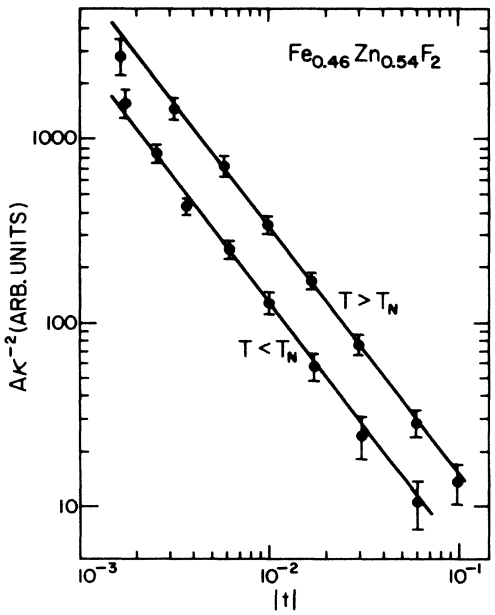

FIG. 2. $\text{Log}_{10}A\kappa^{-2}$ vs $\log_{10}|t|$ for $T \lesssim T_N$. From the amplitudes and slope the amplitude ratio A^+/A^- and the staggered susceptibility critical exponent are obtained.

TABLE I. Comparison of neutron scattering exponents and amplitude ratios with theory for the random-exchange Ising model. The errors in the experimental values for the critical exponents ν and γ represent *two* standard deviations but do not take into account possible systematic errors.

	$Fe_{0.46}Zn_{0.54}F_2$ $1.5\times10^{-3}<\lvert t\rvert<10^{-1}$	Theory
$T_n(K)$	35.667 ± 0.010	...
ν	0.69 ± 0.01	0.70[a](0.68)[b]
κ_0^+/κ_0^-	0.69 ± 0.02	0.83[c]
κ_0 (rlu)	0.46 ± 0.03	...
γ	1.31 ± 0.03	1.3[a](1.34)[b]
χ_0^+/χ_0^-	2.8 ± 0.2	1.7[c]
χ_0^-[d]	0.28 ± 0.05	...
B	-2.2 ± 2.0	...

[a]Arbitrary units.
[b]Reference 17(a).
[c]Reference 17(b).
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ed reduced temperature ranges were consistent with the errors given in Table I. We believe the values of the parameters we have obtained are considerably more reliable than those previously reported for $Fe_{0.5}Zn_{0.5}F_2$ (Ref. 12) because of the much higher quality of the $Fe_{0.46}Zn_{0.54}F_2$ crystal. Further details of line-shape analysis, resolution corrections, and exponent and amplitude ratio determination will be given elsewhere.

Comparing the random-exchange susceptibility exponent $\gamma=1.31\pm0.03$ with the crossover exponent $\phi=1.42\pm0.03$, we find the ratio $\phi/\gamma=1.08\pm0.05$, which supports Aharony's prediction.[1] Regarding the new value of correlation-length exponent $\nu=0.69\pm0.01$ it is interesting to combine it with the specific-heat exponent $\alpha=-0.09\pm0.03$, obtained from birefringence studies,[12] to check the hyperscaling relation $2-\nu d-\alpha=0$. Combining the experimental values with $d=3$, we find $2-\nu d-\alpha=0.02\pm0.06$, indicating very satisfactory consistency with hyperscaling. Neutron scattering studies of the RFIM critical behavior in this same crystal are planned in the near future.

Although our work is in keeping with his inequality, Aharony[1] had sought to explain other measurements[9,15] of ϕ in diluted antiferromagnets as being consistent with "weak randomness in the exchange coefficients," because they seemingly gave values close to the pure $d=3$ Ising one, $\phi=1.25$. In one particular system to which he refers ($Mn_xZn_{1-x}F_2$) there have been other studies[7,16] which strongly suggest that asymptotic REIM, and not pure Ising, critical behavior is observed at dilutions such that $x\leq0.9$. Furthermore, the exchange couplings are virtually identical in the isostructural FeF_2 and MnF_2 crystals as are the normalized shifts in $T_c(H)$ at the same fields and dilutions.

How then does one explain the *apparent* differences in ϕ for diluted MnF_2 and FeF_2? We believe the answer lies in the determination of T_N in randomly diluted crystals which contain a concentration gradient. Experiment[12] and theory[17] for the $d=3$ REIM show the magnetic specific heat (C_m) exponent α to be negative ($\alpha=-0.09\pm0.03$), and the experiment reveals the amplitude ratio $A^+/A^-=1.6\pm0.3$.[18] An elementary simulation[19] of $C_m=A^{\pm}\lvert t\rvert^{-\alpha}$, with an assumed variation of T_N through the sample, produces a rounding of the predicted cusp and with the average $\bar{T}_N$ *shifted so that it lies higher in temperature than does the rounded peak in* C_m.[20] When a field is applied and crossover to the $d=3$ RFIM occurs, C_m diverges logarithmically with $A^+=A^-$.[11] In that case the average $\bar{T}_c(H)$ does occur at the rounded peak. Thus the difference $\bar{T}_c(H)-\bar{T}_N$ is incorrectly given if one just takes the difference in the C_m *peak* temperatures $T_c^p(H)-T_N^p$. We have estimated the correction to $\bar{T}_N$ for the published $Mn_{0.75}Zn_{0.25}F_2$ differential thermal expansion data[15] to be at least $+0.07$ K. By making that correction we find ϕ changes from 1.25 to 1.39. A further confirmation of our conjecture awaits a precision measurement of both ϕ and γ in a homogeneously random crystal of $Mn_xZn_{1-x}F_2$, but it would seem there is doubtful evidence for "weaker randomness in the exchange coefficients" in this system.

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