

## Critical behavior of a site-diluted three-dimensional Ising magnet

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The critical behavior of the site-diluted three-dimensional Ising antiferromagnet  $\text{Fe}_{1-x}\text{Zn}_x\text{F}_2$  has been studied with the use of neutron scattering and linear birefringence techniques. The neutron scattering measurements were performed on a crystal with  $x \simeq 0.5$  and  $T_N = 42.50$  K. The critical scattering has a Lorentzian profile both above and below  $T_N$ . The staggered susceptibility and correlation length exhibit power-law divergences over the reduced-temperature range  $10^{-1}$  to  $2 \times 10^{-3}$  with critical exponents  $\gamma = \gamma' = 1.44 \pm 0.06$  and  $\nu = \nu' = 0.73 \pm 0.03$ , respectively. From the linear birefringence  $\Delta n$ , the critical behavior of the magnetic heat capacity was determined on a crystal with  $x = 0.4$  and  $T_N = 47.05$  K.  $d(\Delta n)/dT$  clearly exhibits cusplike behavior rather than a power-law divergence over the reduced-temperature range  $2 \times 10^{-2}$  to  $10^{-3}$  with exponent  $\alpha = \alpha' = -0.09 \pm 0.03$ . The correlation-length and heat-capacity critical indices satisfy the hyperscaling relation  $3\nu + \alpha - 2 = 0$  to within the experimental errors. Since all measured exponents differ markedly from the pure Ising values, this strongly suggests that *random-exchange* Ising behavior has been observed. The results are consistent with recent theoretical predictions for the random-exchange Ising fixed point by Newman and Riedel.

## I. INTRODUCTION

The phase-transition behavior of random systems is a subject of continuing interest.<sup>1</sup> One of the earliest, and still most dramatic, predictions is due to Harris.<sup>2</sup> Using a heuristic criterion he predicted that for  $n$ -vector models, the phase-transition behavior would be unaltered by small randomness in the interactions provided that the heat-capacity exponent  $\alpha$  was less than zero. Renormalization-group calculations<sup>3</sup> have reinforced this prediction. For  $\alpha > 0$ , a crossover to new critical behavior<sup>4,5</sup> is expected. For  $n$ -vector models in three dimensions (3D) only the Ising model ( $n = 1$ ) has  $\alpha > 0$ . However, because the crossover behavior<sup>3-5</sup> is governed by  $(\Delta J/\bar{J})^{1/\alpha}$ , where  $\Delta J$  is the measure of the randomness in  $J$  and  $1/\alpha \simeq 9$ , it has been felt that this new random-exchange critical behavior for the 3D Ising model would be very difficult to observe.

As part of a study of random-field effects in Ising magnets, we have carried out systematic measurements<sup>6,7</sup> of the phase-transition behavior in the site-diluted Ising system  $\text{Fe}_{1-x}\text{Zn}_x\text{F}_2$ ; in those experiments the emphasis has been on the behavior in an external magnetic field  $H$ . However, in the course of that work we found that our samples were of suf-

ficiently high quality, both in terms of mosaicity and chemical homogeneity that accurate measurements of the critical behavior at  $H = 0$  could be performed at reduced temperatures of  $\sim 10^{-3}$ , both above and below  $T_N$ . In pure magnets one observes exponents close to the asymptotic values in the  $10^{-2}$  to  $10^{-3}$  reduced-temperature range. It seemed possible, therefore, that the influences of the random-exchange Ising fixed point might be observed for

$$|\tau| = \left| \frac{T}{T_N} - 1 \right| \sim 10^{-2}$$

in systems with very large randomness, that is,  $\Delta J/J \sim \frac{1}{2}$ . This would clearly be of interest in its own right. Further, in order to interpret the random-field experiments, one needs a proper understanding of the zero-field critical behavior.

In this paper we report results of neutron scattering and birefringence measurements at  $H = 0$  in  $\text{Fe}_{0.5}\text{Zn}_{0.5}\text{F}_2$  and  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2$  samples, respectively. Our measurements show that these crystals exhibit critical behavior which differs markedly from the 3D Ising model, both in terms of exponents and amplitude ratios. The experimental results are consistent with recent theoretical predictions by New-

man and Riedel for the random Ising model. The format of this paper is as follows. In Sec. II we discuss the crystallography, magnetism, and phase-transition behavior. Section III contains the neutron scattering results and analysis. In Sec. IV the linear birefringence measurements and analysis are presented. A general discussion summary and conclusions are given in Sec. V.

## II. BEHAVIOR IN PURE $\text{FeF}_2$

The properties of  $\text{FeF}_2$  have been discussed extensively by Hutchings, Schulhof, and Guggenheim.<sup>8</sup> The crystal structure of  $\text{FeF}_2$  is rutile with  $a = 4.697$  Å and  $c = 3.309$  Å at room temperature. The space group is  $D_{4h}^{14}-P4/mnm$ , and the cation sites, surrounded by distorted octahedra of  $\text{F}^-$  ions, have point symmetry  $D_{2h}$ . The magnetic structure is such that the spins align along the  $c$  axis, with the spin of the body-centered ions antiparallel to those at the corners of the cell. The magnetic and nuclear unit cells are therefore identical.

The spin-wave dispersion relations for  $\text{FeF}_2$ , investigated using neutron scattering,<sup>9</sup> show that the magnetic properties may be well described by a spin Hamiltonian of the form ( $S = 2$ )

$$\mathcal{H} = \sum_{i,j} J_2 \vec{S}_i \cdot \vec{S}_j + \sum_i DS_i^z{}^2 + \sum_j DS_j^z{}^2. \quad (1)$$

The summation in the first term is over all next-nearest-neighbor pairs (corner and body-centered ions), and the single-ion terms are summed over ions on both sublattices  $i$  and  $j$ . The other magnetic interactions are found to be less than 6% of  $J_2$ . The parameters thus determined are  $J_2 = 0.45$  meV and  $D = 0.84$  meV, so that the ratio of anisotropy field to exchange field acting on a  $\text{Fe}^{2+}$  ion is  $(2S - 1)D/16J_2 = 0.35$ . The single dominant inter-sublattice exchange interaction and well-defined relatively high anisotropy make  $\text{FeF}_2$  an ideal substance in which to investigate Ising critical behavior. Further, since  $\text{FeF}_2$  may be readily alloyed with  $\text{ZnF}_2$  it provides a model 3D random exchange Ising system.

These mixed crystals have been studied by several techniques outside of the critical region. We have measured the spin-wave gap  $E_0$  in the sample  $\text{Fe}_{0.5}\text{Zn}_{0.5}\text{F}_2$ , and find that  $E_0 = 4.2 \pm 0.2$  meV, which is to be compared with 6.5 meV in pure  $\text{FeF}_2$ . This result is consistent with light-scattering results<sup>10</sup> and coherent potential approximation (CPA) theory.<sup>11</sup> The low-temperature perpendicular susceptibility  $\chi_{\perp}$  has been measured and found to be in excellent agreement with a classical-spin computer simulation.<sup>12</sup> The dependence of  $T_N$  on  $x$  has been previously studied via the linear birefringence,<sup>13</sup> and has been found to be in agreement

TABLE I. Exponents and amplitude ratios for uniform and random three-dimensional Ising models.

	Pure Ising <sup>a</sup>	$\text{FeF}_2$	Random Ising <sup>c</sup>	$\text{Fe}_{1-x}\text{Zn}_x\text{F}_2$ $x = 0.4, 0.5^f$
$\gamma$	1.24	$1.38 \pm 0.08^b$	1.39 (1.34)	$1.44 \pm 0.06$
$\nu$	0.63	$0.67 \pm 0.04^b$	0.70 (0.68)	$0.73 \pm 0.03$
$\alpha$	0.11	$0.11 \pm 0.005^c$	-0.09 (-0.04)	$-0.09 \pm 0.03$
$\beta$	0.325	$0.325 \pm 0.005^d$	0.35 (0.35)	? <sup>g</sup>
$\chi/\chi'$	5.1	$6.1 \pm 1.0^b$	?	2.2
$A/A'$	0.51	$0.54 \pm 0.02^c$	?	$1.6 \pm 0.3$
$3\nu - 2 + \alpha$	0	$0.12 \pm 0.12$	0	$0.10 \pm 0.12$

<sup>a</sup>Reference 15; the amplitude ratios are from A. Aharony and P. C. Hohenberg, Phys. Rev. B **13**, 3081 (1976).

<sup>b</sup>Reference 8.

<sup>c</sup>Reference 17.

<sup>d</sup>Reference 14.

<sup>e</sup>Reference 5; the values in parentheses are the results of Jug.

<sup>f</sup>The exponents represent averages of those obtained above and below  $T_N$ .

<sup>g</sup>It is interesting to note that in  $\text{Mn}_{0.86}\text{Zn}_{0.14}\text{F}_2$ , R. A. Dunlap and A. M. Gottlieb [Phys. Rev. B **23**, 6106 (1981)] find  $\beta = 0.349 \pm 0.008$  in agreement with the random Ising theory.

with the theory for Ising magnets.

A variety of measurements has been performed on the phase-transition behavior of  $\text{FeF}_2$ . In 1967, Wertheim and Buchanan<sup>14</sup> measured the order parameter using the Mössbauer effect. They found  $M(T)/M_0 = 1.36 |\tau|^{+0.325 \pm 0.005}$  over the reduced-temperature range  $10^{-3} < |\tau| < 10^{-1}$ . As shown in Table I the exponent  $\beta = 0.325 \pm 0.005$  is in exact agreement with the current best theoretical value<sup>15</sup> of  $0.325 \pm 0.002$  for the 3D Ising model. It should be noted that there was no precise theoretical prediction for  $\beta$  at the time of the Wertheim-Buchanan experiment. The critical heat-capacity behavior has been determined both by direct measurement<sup>16</sup> and indirectly<sup>17</sup> via the temperature derivative of the linear birefringence. These measurements yield  $\alpha = \alpha' = 0.113 \pm 0.007$  and an amplitude ratio  $A/A' = 0.54 \pm 0.02$ . With a small correction to scaling term these exponents describe the results over the range  $10^{-4} \leq |\tau| \leq 5 \times 10^{-2}$ . As shown in Table I, both the exponent and amplitude ratio are in good agreement with the theoretical values.

Hutchings *et al.*<sup>8</sup> studied the static and dynamic behavior of the wave-vector dependent susceptibility. They found that only the Ising component  $\chi^{||}(\vec{q})$  had appreciable weight near  $T_N$ , thereby directly verifying the Ising character of the phase transition. They determined that the correlation-length exponent  $\nu = 0.67 \pm 0.04$ , which agrees, within the errors, with the theoretical value of 0.63. The staggered susceptibility was observed to diverge with an exponent  $\gamma = 1.38 \pm 0.08$ , somewhat larger than the Ising value of 1.24. However, we do not regard this discrepancy as serious since only three of the data points were in the region  $\tau < 8 \times 10^{-3}$  and one of these was used to fix  $T_N$  in the correlation length fits. In addition, the median value of  $\gamma$  was determined to be larger than the median value of  $2\nu$ , a result inconsistent with the scaling relation  $\gamma = (2 - \eta)\nu$  with  $\eta > 0$ . The ratio of the susceptibilities above and below  $T_N$  was in satisfactory agreement with theory. Unfortunately the data below  $T_N$  were not accurate enough to determine  $\nu'$  and  $\gamma'$ .

In summary,  $\text{FeF}_2$  exhibits 3D Ising critical behavior for  $|\tau| < 10^{-1}$  with, in general, both exponents and amplitude ratios in satisfactory agreement with current theoretical predictions. As noted above, by diluting  $\text{FeF}_2$  with the nonmagnetic isomorph  $\text{ZnF}_2$  it is possible to prepare site-random alloys which should be ideal realizations of the 3D random-exchange Ising model. The only restriction is that the  $\text{Fe}^{2+}$  concentration should be much larger than  $x_p$ , the percolation concentration. For  $\text{FeF}_2$   $x_p \simeq 0.75$ , and our samples have  $x = 0.5$  and  $0.4$ , so that there should be almost no effect from the percolation multicritical point.

### III. NEUTRON SCATTERING RESULTS AND ANALYSIS

The neutron scattering measurements were carried out using a two-axis spectrometer at the Brookhaven High Flux Beam Reactor. The sample, which had dimensions  $10 \times 8 \times 8 \text{ mm}^3$ , had a nominal concentration of  $\text{Fe}_{0.5}\text{Zn}_{0.5}\text{F}_2$ . In fact, this sample had a macroscopic concentration gradient along the  $c$  axis with a consequent spread of Néel temperatures of nearly 3 K about a median of 41 K. The value of 41 K is in reasonable agreement with the value  $0.5 \times T_N(x=0) = 39.2 \text{ K}$ , suggesting that the average Zn concentration was indeed quite close to 50%. Accordingly, the sample was masked with cadmium so that only the top 2 mm were illuminated with neutrons. By monitoring both the Bragg scattering and the critical scattering we found that this section was quite homogeneous with  $T_N = 42.50 \text{ K}$  and a spread of only 0.05 K halfwidth at half maximum (HWHM). This masked sample allowed studies of the critical behavior to be performed down to about  $2 \times 10^{-3}$  in reduced temperature.

The sample was mounted with the  $c$  axis vertical in a standard flow cryostat; relative temperatures could be controlled to about 0.01 K. An incident neutron wave vector of  $2.67 \text{ \AA}^{-1}$  was used with two pyrolytic graphite filters to eliminate higher-order neutrons. The collimation was  $10'$  throughout, resulting in an instrumental resolution function of  $0.05 \text{ \AA}^{-1}$  HWHM vertically,  $0.005 \text{ \AA}^{-1}$  HWHM in the longitudinal direction, and  $0.0014 \text{ \AA}^{-1}$  HWHM transverse to the wave vector. Measurements were performed exclusively around the  $(1,0,0)$  antiferromagnetic lattice point. The room-temperature lattice constants of our sample were  $a = 4.707 \text{ \AA}$  and  $c = 3.219 \text{ \AA}$ .

As will be discussed elsewhere,<sup>7</sup> the sample was of such high quality that the magnetic Bragg scattering suffered from severe extinction. This in turn, vitiated any possible study of the critical behavior of the order parameter. However, the very small mosaicity of the sample did mean that the transverse resolution function was accurately Gaussian with no extended tails. This allowed a detailed study of the critical scattering below the Néel temperature to be made. In the following, we shall use as notation  $(Q_x, Q_y, Q_z)$  where  $Q_x$  and  $Q_y$  are measured in units of  $2\pi/a$  and  $Q_z$  in units of  $2\pi/c$ . By monitoring the scattering at  $(1, -0.005, 0)$  we could measure the strength of the critical fluctuations. The intensity at  $(1, -0.005, 0)$  showed a well-defined peak at  $42.55 \pm 0.05 \text{ K}$ , which should be close to, albeit slightly higher than,<sup>18</sup> the Néel temperature.

We show in Fig. 1 a series of transverse scans through the critical scattering both above and below

the Néel temperature. The data are clearly of comparable quality for  $T > T_N$  and  $T < T_N$ . In the quasielastic approximation, which should be well satisfied here,<sup>8</sup> the measured cross section is proportional to  $\chi^{||}(\vec{q})$  convoluted with the instrumental resolution function. Because  $\chi^{||}(\vec{q})$  is both very weak and flat over the  $q$  range of interest, it is simply absorbed into the background. In order to deconvolve the data we assume a cross section,

$$\mathcal{S}(\vec{q}) = AT/(K^2 + q^2), \quad (2)$$

where  $q^2 = q_x^2 + q_y^2 + q_z^2$  with the  $q_\alpha$  measured in reciprocal-lattice units,  $T$  is the temperature and  $\vec{q} = \vec{Q} - (1, 0, 0)$  where  $\vec{Q}$  is the momentum transfer. The solid lines in Fig. 1 represent the results of least-squares fits to the data, excluding the points  $|q_y| \leq 0.003$  for  $T \leq T_N$ , of Eq. (2) convoluted with the instrumental resolution function. Clearly the Lorentzian form, Eq. (2), describes the measured profiles very well at all temperatures. The  $\chi^2$  goodness of fit parameter typically varied between 0.9 and 1.3.

The results for the inverse correlation length  $K$  and the inverse staggered susceptibility  $\chi^{-1}(\vec{0})$  are shown in Fig. 2. In order to obtain optimal values for the critical exponents we first fit all four sets of data separately to power laws with the amplitude, exponent, and Néel temperature allowed to

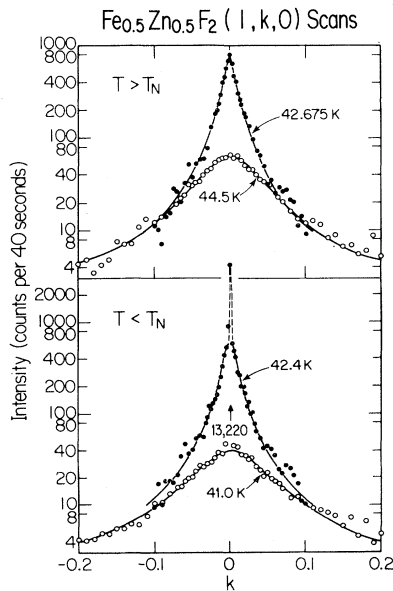


FIG. 1. Transverse scans through the (1,0,0) Bragg peak position in  $\text{Fe}_{0.5}\text{Zn}_{0.5}\text{F}_2$  above and below the Néel temperature  $T_N = 42.50$  K. The solid lines are the results of fits of the critical scattering to a Lorentzian as discussed in the text.

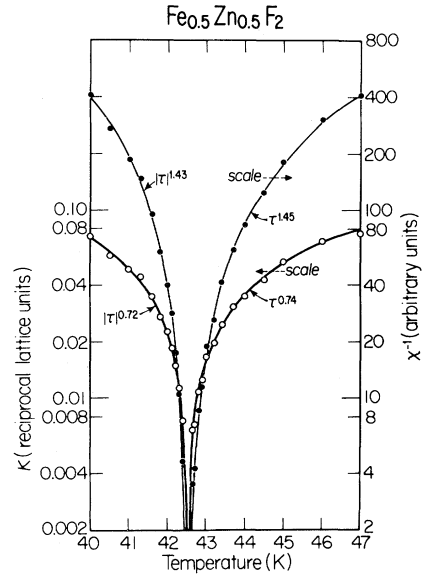


FIG. 2. Inverse correlation length and inverse staggered susceptibility in  $\text{Fe}_{0.5}\text{Zn}_{0.5}\text{F}_2$  above and below  $T_N = 42.50$  K. The solid lines are the results of fits to single power laws as discussed in the text.

vary freely. Fits to  $K(T > T_N)$ ,  $\chi(T > T_N)$ ,  $K(T < T_N)$ , and  $\chi(T < T_N)$  then yield  $T_N = 42.49$ ,  $42.48$ ,  $42.53$ , and  $42.53$  K, respectively; these all agree reasonably with the estimate from the wing critical scattering of  $42.55 \text{ K} \pm 0.05 \text{ K}$ . With  $T_N$  fixed at the median value of  $42.505 \text{ K}$ , the fits then yield

$$\begin{aligned} K(T > T_N) &= 0.0264 \pm 0.0003 \tau^{0.74 \pm 0.01}, \\ K(T < T_N) &= 0.0364 \pm 0.0005 |\tau|^{0.72 \pm 0.01}, \\ \chi^{-1}(T > T_N) &= 0.472 \pm 0.008 \tau^{1.45 \pm 0.02}, \\ \chi^{-1}(T < T_N) &= 1.046 \pm 0.02 |\tau|^{1.43 \pm 0.02}. \end{aligned} \quad (3)$$

Here the  $K$ 's are in reciprocal-lattice units while  $\chi^{-1}$  is in arbitrary units. The solid lines in Fig. 2 are calculated from Eq. (3). All but the datum point closest to  $T_N$  are fit by Eq. (3) to within the errors. Thus the single power laws work well over the reduced-temperature range  $10^{-1} > |\tau| > 2 \times 10^{-3}$ . We conclude from Eq. (3) that, including the uncertainty in  $T_N$  as well as statistical errors,

$$\gamma = \gamma' = 1.44 \pm 0.06, \quad \chi/\chi' = 2.2 \pm 0.1, \quad (4)$$

$$\nu = \nu' = 0.73 \pm 0.03, \quad K/K' = 0.73 \pm 0.02,$$

for  $10^{-1} > |\tau| > 2 \times 10^{-3}$ . It is clear from Table I that both the exponents and the amplitude ratios in  $\text{Fe}_{0.5}\text{Zn}_{0.5}\text{F}_2$  differ significantly from those of the

pure Ising model. We shall discuss these results further in Sec. V.

#### IV. BIREFRINGENCE RESULTS

In the transition-metal difluorides it is well established both experimentally<sup>13</sup> and theoretically<sup>19</sup> that the temperature derivative of the linear birefringence  $\Delta n$  is proportional to the magnetic specific heat in the critical region  $|\tau| < 10^{-1}$ . The birefringence method has the salient advantage over conventional specific-heat techniques of being largely insensitive to the lattice specific-heat background. In addition, the laser beam may be oriented perpendicular to the concentration gradient in mixed alloys, thereby reducing the effects of such gradients on the critical behavior measurements.

The birefringence measurements were carried out on a crystal of  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2$ . The melt was stirred during the growth process, which resulted in a significantly reduced concentration gradient along the growth axis. The experimental configuration and techniques were identical to those described previously.<sup>20</sup> From the peak in  $d(\Delta n)/dT$  the Néel temperature was determined to be  $T_N = 47.04 \pm 0.01$  K. The Fe concentration was estimated to be 0.60 by scaling the observed  $T_N$  by the value of  $T_N = 78.39$  K for  $\text{FeF}_2$ . We show in Fig. 3  $d(\Delta n)/dT$  over the reduced-temperature range  $4 \times 10^{-4} < |\tau| < 1.0$ . The differences between these data and those of pure  $\text{FeF}_2$  are immediately obvious in this figure. Whereas in  $\text{FeF}_2$  the data show positive curvature everywhere in the critical region ( $|\tau| < 0.1$ ), which indicates that  $\alpha > 0$ , the present data show *negative* curvature for  $|\tau| < 0.1$ , which indicates  $\alpha < 0$ . The  $\Delta n$  anomaly at  $T_N$  is largely suppressed, owing to

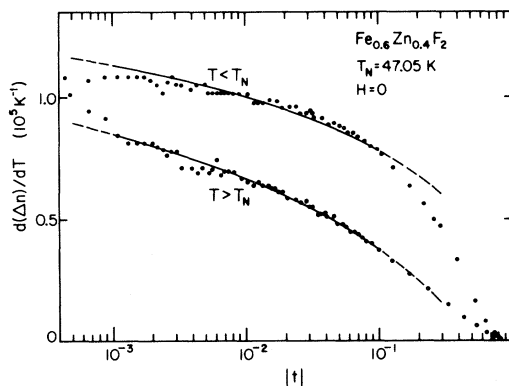


FIG. 3. Temperature derivative,  $d(\Delta n)/dT$ , of the linear birefringence in  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2$  above and below Néel temperature  $T_N = 47.04$  K. The solid lines are the results of fits to the scaling form for the magnetic heat capacity, Eq. (5), as discussed in the text.

the change from divergent ( $\alpha > 0$ ) to cusplike ( $\alpha < 0$ ) behavior. Outside the critical region ( $|\tau| > 0.1$ ),  $d(\Delta n)/dT$  is only slightly reduced from that of  $\text{FeF}_2$ , owing to the magnetic dilution. Data for  $|\tau| < 10^{-3}$  show definite evidence of rounding, presumably because of a small residual concentration gradient.

Critical exponents were extracted directly from the measured  $\Delta n$  rather than its temperature derivative. Specifically, the  $\Delta n$  data were analyzed by fitting to the temperature integral of the specific-heat scaling function

$$C \sim \frac{d(\Delta n)}{dT} = \frac{A}{\alpha} |\tau|^{-\alpha(1+D|\tau|^X)+B} \quad (5)$$

for  $\tau > 0$  and the same function with  $A$  replaced by  $A'$  and  $D$  by  $D'$  for  $\tau < 0$ . Data were fit to Eq. (5) over the reduced temperature range  $10^{-3} < |\tau| < 10^{-1}$ ; initially  $X$  was fixed at 0.5.

When all the parameters were allowed to vary simultaneously, a fit was obtained with  $\alpha = -0.115$ , a large ratio  $A/A'$ , and very large  $D$  and  $D'$  terms. We felt that this fit was physically unreasonable, and concluded that the data were not good enough to distinguish between the exponents in the asymptotic and crossover regions. In order to separate these regions, we attempted to determine the asymptotic behavior by making several fits for small  $|\tau|$  with *no* correction terms, and found  $\alpha$ ,  $A$ , and  $A'$  to be relatively independent of the limits of  $|\tau|$  in the range  $10^{-3}$  to  $2 \times 10^{-2}$ . The values of these parameters were held fixed and the fit repeated over the range  $10^{-3} \leq |\tau| \leq 10^{-1}$  letting  $D$  and  $D'$  vary. This resulted in reasonable fits out to  $|\tau| \sim 0.1$ , at the expense of a noticeable worsening of the fits near  $|\tau| \sim 10^{-2}$ . The parameters obtained are as follows:

$$\begin{aligned} \alpha &= -0.09 \pm 0.03, \quad T_N = 47.05 \pm 0.1, \\ A/A' &= 1.6 \pm 0.3, \quad A' = (7.5 \pm 0.7) \times 10^{-7}, \\ D/D' &= 0.6 \pm 0.3, \quad D' = 0.5 \pm 0.3, \\ B &= (1.6 \pm 0.2) \times 10^{-5}, \end{aligned} \quad (6)$$

with  $T_N$  expressed in K and  $A'$ ,  $D'$ , and  $B$  in  $\text{K}^{-1}$ .

In addition, the parameter<sup>21</sup>  $P = (1 - A/A')/\alpha$ , which is usually observed<sup>22</sup> to be close to 5, is found to have the value 7. The error bars are intended to reflect not only the errors in fitting Eq. (5), but also the attendant theoretical uncertainties. Indeed, after all of this analysis was complete we became aware of the theoretical work by Newman and Riedel.<sup>5</sup> Those authors suggest a correction to scaling exponent closer to 0.3. If this value is used in Eq. (5), it is found the fits are somewhat less satisfactory. It appears therefore that  $\alpha = \alpha' = -0.09 \pm 0.03$  should

be properly regarded as the *effective exponent* describing the divergence of the heat capacity in the reduced-temperature range  $10^{-3} \leq |\tau| \leq 2 \times 10^{-2}$  but that the correction terms remain ill determined. As found in the neutron measurements, both the exponent  $\alpha$  and amplitude ratio  $A/A'$  differ markedly from the pure 3D Ising values of 0.11 and 0.51, respectively.

Although the crossover from pure Ising to random Ising behavior is predicted to occur around  $|\tau| \sim (\Delta J/J)^{1/\alpha} \sim 2 \times 10^{-3}$ , we find no region in  $|\tau|$  which can be described by "pure" Ising exponents, or any evidence for crossover from pure to random exponents. We conclude that the crossover is either outside the critical region, or is so slow that only "effective" exponents are observed. The crossover from positive to negative  $\alpha$  means that the random system indeed satisfies the Harris criterion<sup>2</sup> and hence may exhibit a sharp phase transition as is observed experimentally. Unlike the neutron results, corrections to scaling are essential to fit the data beyond  $|\tau| = 2 \times 10^{-2}$ . Best results are obtained using the same form as in FeF<sub>2</sub>, that is  $X = 0.5$  rather than the value 0.3 suggested by Newman and Riedel.

## V. DISCUSSION AND CONCLUSIONS

A number of important qualitative and quantitative conclusions emerge from this work. In Fe<sub>1-x</sub>Zn<sub>x</sub>F<sub>2</sub> with  $x \sim 0.5$  the HWHM of the distribution of exchange fields seen by the Fe spins is about one-half of the median value, that is,  $(\Delta J/J) \sim 0.5$ . Thus we are in the limit of large randomness. In spite of this, the crystals exhibit phase transitions which are sharp on the scale of  $|\tau| \sim 10^{-3}$ ; the residual rounding is probably not intrinsic but is caused by concentration gradients.

The neutron scattering experiments have demonstrated that the critical scattering is Lorentzian in character both above and below  $T_N$ . Because of the high quality of the sample it was possible to carry out measurements of comparable quality for  $\tau \gtrsim 0$ . In fact, there are remarkably few measurements reported in the literature which allow a test of the scaling requirement of equality of the correlation length exponents above and below  $T_N$ . As noted in Secs. III and IV we find  $\gamma = \gamma'$ ,  $\nu = \nu'$ , and  $\alpha = \alpha'$  well within the errors. As discussed above, the Harris<sup>2</sup> argument suggests that the transition will be rounded if  $\alpha > 0$ . It has been assumed, therefore, that the random-exchange Ising fixed point<sup>4</sup> would be characterized by an  $\alpha < 0$  in order to allow a sharp transition. Our birefringence measurements provide strong experimental proof that this is, in fact, the case.

The heat capacity and correlation-length exponents should be related by the hyperscaling relation

$$d\nu = 2 - \alpha. \quad (7)$$

The left-hand and right-hand sides of Eq. (7) are, respectively,  $2.19 \pm 0.09$  and  $2.09 \pm 0.03$ . This agreement between the two quite independent measurements on different samples is clearly very encouraging.

Our quantitative results are summarized in Table I. The measured exponents differ significantly from those of the 3D Ising model. Given the general good agreement with the Ising model manifested by pure FeF<sub>2</sub> over the  $10^{-1} - 10^{-3}$  reduced-temperature range, we can only conclude that we are indeed observing random Ising critical behavior. As noted above, crossover from pure to random behavior should occur at  $\tau \sim (\Delta J/J)^{1/\alpha} \sim 2 \times 10^{-3}$ ; however, this estimate is certainly only valid to within an order of magnitude so that the observability of random behavior for  $\tau \sim 10^{-2}$  should not be considered surprising.

As noted in the Introduction, theoretical estimates for the 3D random-exchange Ising exponents have been given by Newman and Riedel and by Jug.<sup>5</sup> Their results are listed in Table I. Clearly, both theoretical predictions show the same trend as our measured values. Indeed, the values calculated by Newman and Riedel agree with our experimental values for each of  $\alpha$ ,  $\gamma$ , and  $\nu$  to within the errors. The theoretical numbers, however, correspond to asymptotes, whereas the experiments correspond to the temperature range  $10^{-1} - 10^{-3}$  so that this excellent quantitative agreement may be partly coincidental. Finally, we note that the amplitude ratios  $\chi/\chi'$  and  $A/A'$  both differ significantly from the pure system values. As far as we know, there are currently no theoretical estimates of amplitude ratios for the 3D random-exchange Ising model. Clearly, such calculations would be of considerable value.

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