Critical behavior of the site random Ising antiferromagnet $Mn_{1-x}Zn_xF_2$

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Nuclear magnetic resonance has been used to measure β, the critical exponent of the order parameter, in the random Ising antiferromagnet $Mn_{0.864}Zn_{0.136}F_2$. A single-power-law fit to the measured hyperfine field of the form $H_{hf}=Bt^{\beta}$ yields $\beta=0.349(8)$ over the reduced temperature range of 5×10^{-4} to 2×10^{-2}. This differs from the value $\beta=0.335(5)$ obtained by Heller [P. Heller, Phys. Rev. 146, 403 (1966)] for pure MnF_2 and is consistent with the prediction that random disorder will cause a crossing to a new fixed point in the three-dimensional Ising system. The measured exponent can be explained in terms of recent predictions for the location and extent of the crossover region in reduced temperature.

I. INTRODUCTION

During the past few years a number of renormalization-group calculations have been made on the critical behavior of randomly disordered systems.[1–7] These hinge critically on the sign of the specific-heat exponent in the pure system, α_p. If $\alpha_p>0$, they predict convergence to a new fixed point with changed critical exponents; otherwise they hold that random disorder will not change the critical behavior.

So far few experimental results of critical-point measurements in random systems have been reported. Wertheim *et al.*[8] have studied the three-dimensional Ising antiferromagnet $Mn_{1-x}Fe_xF_2$ with $x=0.007$. They have observed the pure-system value for the order-parameter exponent β. Birgeneau *et al.*[9] have observed pure-system values of β, γ, and ν in the two-dimensional Ising system $Rb_2Mn_{0.5}Ni_{0.5}F_4$. More recently Chowdhury *et al.*[10] have observed the pure-system value of β in the three-dimensional Heisenberg ferromagnet $Fe_{1-x}Al_x$, with impurity concentrations $0.01<x<0.04$. In cases where a crossing to new exponents was not predicted, experiments seem to be consistent with theory. The measurements of Wertheim *et al.*,[8] however, do not show the predicted effects of random impurities on a system with $\alpha_p>0$. This makes additional experiments, especially in systems with $\alpha_p>0$, of interest.

Here we present the results of a measurement of the exponent β via an NMR measurement of the ^{19}F hyperfine field in MnF_2 doped with 13.6 at. % Zn in the Mn^{++} sites. The motivation for this study is as follows: (i) Pure MnF_2 has exponents appropriate for the $d=3$ Ising system; in particular, $\alpha_p\simeq+0.10$. (ii) The three-dimensional Ising system is the only physical system predicted to show a crossing to a new fixed point in the presence of random disorder. (iii) Pure MnF_2 has been well studied and there is a highly reliable experimental value of β for comparison.[11]

II. EXPERIMENTAL METHOD

Sample. Our sample was a 0.40-g single crystal of $Mn_{0.864}Zn_{0.136}F_2$, provided by V. Jaccarino of the University of California, Santa Barbara. Butler *et al.*[12] have done NMR studies of similar MnF_2 samples doped with lower Zn concentrations far below the critical temperature. In these samples they have observed various resonances for ^{19}F nuclei with three Mn^{++} nearest neighbors resulting from different second-nearest-neighbor configurations. Because of broadening in the critical region we observe only a single resonance which results from ^{19}F nuclei with three Mn^{++} nearest neighbors.

NMR technique. To measure the ^{19}F hyperfine field as a function of temperature we have used the method that Heller[11] applied to pure MnF_2. This involved sweeping small temperature regions with a fixed rf frequency. Our sample was oriented with its easy magnetic axis perpendicular to the applied rf field. A modulation field of about 150 G peak-to-peak at 75 Hz was applied parallel to the sample easy axis for the purpose of measuring the derivative of the magnetic-resonance signal. This helped to improve the signal-to-noise ratio of the system. The details of the probe and spectrometer have been described elsewhere.[13]

Néel temperature. The Néel temperature was determined by a method independent of the temperature dependence of the order parameter. This distinguishes our experiment from others in which T_N is a fitted parameter. Our method consists of making temperature sweeps through the phase transition with

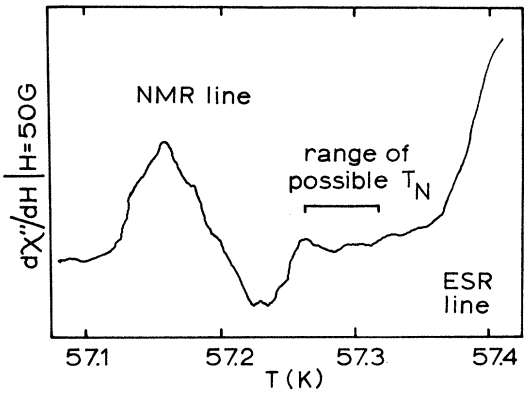

FIG. 1. Resonant absorption in $Mn_{0.864}Zn_{0.136}F_2$ as a function of temperature at 21.60 MHz. From this and similar temperature sweeps we have deduced a critical temperature of $T_N = 57.285(10)$ K.

a small (~ 50 G) field applied parallel to the magnetic easy axis. An example of such a temperature sweep is illustrated in Fig. 1. Above T_N the ESR line results from the electronic absorption and below T_N the NMR line occurs when the spontaneous field satisfies the resonance condition. Thus the possible range of critical temperatures is bracketed by the ESR line above and the NMR line below. By decreasing the rf frequency, this range of temperatures is decreased. From this type of analysis we have deduced a Néel temperature of $T_N = 57.285(10)$ K. This is consistent with the Néel point studies of Zn-doped MnF_2 of Belanger *et al.*[14]

Temperature control. Temperature control was achieved by a pumped nitrogen bath with the vapor pressure regulated by means of a Cartesian diver manostat. The temperature servo system consisted of a carbon thermistor placed in one leg of a dc bridge. A Beckman 833 power op-amp was used to drive a servo heater wound around a boron nitride sample holder. Temperatures were swept by placing a linearly varying resistance in another leg of the bridge. Temperatures had to be swept slowly enough to allow for the thermal response of the system. For this experiment we used a temperature sweep rate of about 0.03 mK/sec. Temperatures were measured by a calibrated platinum resistance thermometer which provided measurements accurate to 3 mK.

III. RESULTS

Values of the mean ^{19}F hyperfine field as a function of temperature are given in Table I (also see Fig. 2). Values of the reduced temperature, $t = 1 - T/T_N$, were obtained using the independently measured value of $T_N = 57.285$ K. Also shown in Table I are values of the linewidth $\Delta H/H$. Values of the mean hyperfine field were fitted to the power-law expression for the order parameter;

$$H_{\rm hf} = Bt^{\beta_{\rm eff}} . \tag{1}$$

Since the observed linewidths are, in general, larger by about a factor of 3 than those observed in the pure system by Heller[15] for uncertain reasons, there is some ambiguity in the assumption that the mean hyperfine field can be taken as a measure of the order parameter. We have assumed that Eq. (1) is reasonable[10] if $\Delta H/H < 1$. Table I shows that this is

TABLE I. Mean ^{19}F hyperfine-field values for $Mn_{0.864}Zn_{0.136}F_2$. To obtain values of the reduced temperature the independently measured value of $T_N = 57.285$ K was used.

T (K)	t	H (kG)	$\Delta H/H$
56.122	0.020 3	12.605	0.026
56.420	0.015 1	11.136	0.030
56.602	0.011 9	10.641	0.025
56.794	0.008 57	9.179	0.040
57.047	0.004 15	7.134	0.10
57.169	0.002 02	5.639	0.096
57.190	0.001 66	5.440	0.18
57.186	0.001 73	5.391	0.21
57.205	0.001 40	4.950	0.20
57.210	0.001 31	4.849	0.22
57.220	0.001 13	4.637	0.31
57.237	0.000 838	4.443	0.31
57.244	0.000 716	4.222	0.31
57.258	0.000 471	3.514	0.43

TABLE II. Comparison of single-power-law fits for the order parameter for pure and random MnF_2.

Sample	T_N (K)	B (kG)	β	χ^2	Range of t	Reference
MnF_2	67.336(3)	47.6(1)	0.335(5)	2.6	5×10^{-5}–2×10^{-1}	11
$Mn_{0.864}Zn_{0.136}F_2$	57.285(10)	48.9(3)	0.349(8)	4.1	5×10^{-4}–2×10^{-2}	This work

the case for all of our data; however, our measurements come close to the minimum reduced temperature which meets this linewidth criteria for this sample. The results of a least-square fit to Eq. (1) for both our data and that of Heller[11] for pure MnF_2 are given in Table II. Our result for β_{eff} differs from that obtained in the pure system by about two standard deviations and is suggestive of the predicted crossing o a new fixed point. We note that, although the critical temperature is lower in the impure sample, the coefficient B is not significantly different. The χ^2 analysis indicates that both fits are of comparable quality.

In order to determine the reliability of our measured exponent, we have performed a range-of-fit analysis.[16] Least-squares fits to Eq. (1) were performed while successively eliminating data points from the top of the reduced temperature range. The results of this analysis for β_{eff} as a function of $t_{\max}$ for our data as well as that of Heller[11] for pure MnF_2 are shown in Fig. 3. This analysis could not be performed for β in $Mn_{0.993}Fe_{0.007}F_2$ as the original data were not available. The range-of-fit analysis shows that reliable values of β can be deduced from both Heller's measurements[11] in pure MnF_2 and our measurements in the disordered system. Figure 3 provides $\beta=0.335(5)$ for pure MnF_2 and $\beta=0.352(10)$ for $Mn_{0.864}Zn_{0.136}F_2$.

Values of the linewidths in Table I are plotted in Fig. 4. A least-squares fit to these data shows that this linewidth diverges with an exponent of $-0.82(6)$. This is consistent with the value of -0.83 found for the pure system by Heller.[15] In principle the dynamic exponent z can be determined from linewidth measurements in the critical region if the line broadening is a result of critical fluctuations.[17,18] However, since we cannot be certain that broadening in the random system can be attributed only to relaxation effects, we cannot assume that the spin-autocorrelation time $\tau_c \propto \Delta H$.

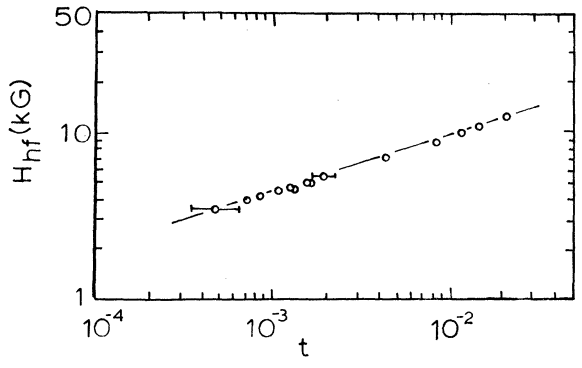

FIG. 2. Measured values of the ^{19}F hyperfine field as a function of reduced temperature. The independently measured value of T_N was used.

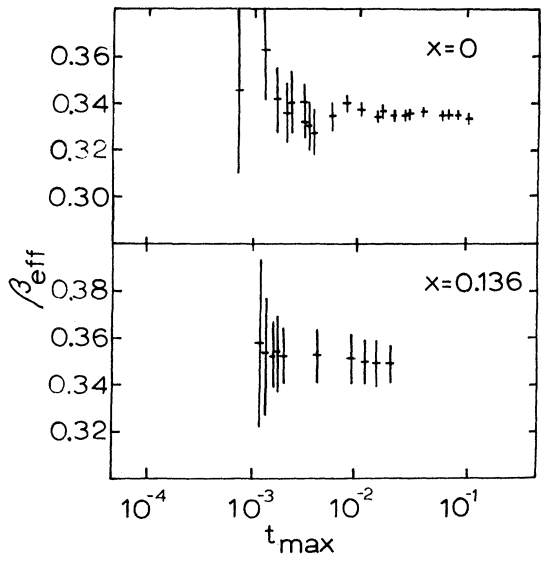

FIG. 3. Range-of-fit analyses for the values of β in pure MnF_2 from Ref. 11 and in 13.6 at. % Zn-doped MnF_2 from this work. Power-law fits were made while successively eliminating data points from the top of the reduced temperature range. For pure MnF_2, T_N was left as a free parameter and for $Mn_{0.864}Zn_{0.136}F_2$, T_N was fixed to the independently measured value, $T_N=57.285$ K. Power-law fits to our data with T_N as a free parameter have yielded consistent results.

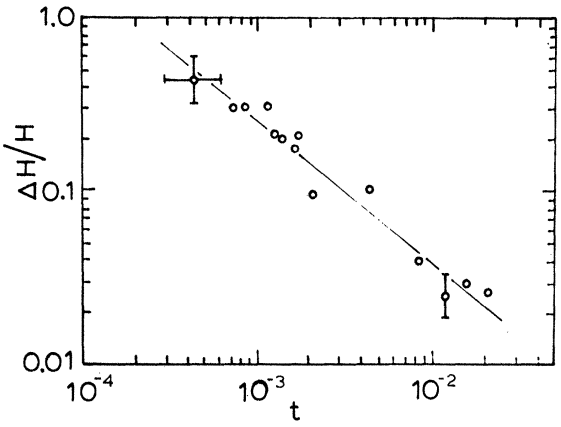

FIG. 4. Relative linewidths, $\Delta H/H$, as a function of reduced temperature for $Mn_{0.864}Zn_{0.136}F_2$. A power-law fit to the data shows that this quantity diverges with an exponent of $-0.82(6)$.

IV. DISCUSSION

Qualitatively the behavior of the order parameter in a random three-dimensional Ising system may be described by Fig. 5. For reduced temperatures larger than some value t_p the system may be described by the pure system exponent. For reduced temperatures less than t_r the system may be described by a single power law with the new random-system exponent. The crossover region covers some reduced temperature range $t_r < t < t_p$. Although our data appear to obey a single power law based on our range-of-fit analysis, we must question whether our measured value of β in fact represents the asymptotic random value.

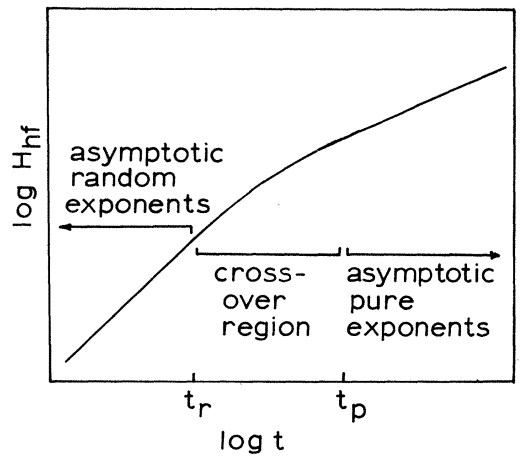

FIG. 5. Predicted effects of impurities on the critical behavior of the order parameter in the three-dimensional Ising system. This behavior is defined by a single-power law with the pure-system exponent for reduced temperatures greater than t_p and by a single-power law with the random exponent for reduced temperatures less than t_r.

Recent calculations by Fishman and Aharony[19] have predicted that random behavior should be observed for the random three-dimensional Ising system for reduced temperatures less than

$$t < t_r = x^{|1/\alpha_a|} , \tag{2}$$

where x is the impurity concentration. With $x = 0.136$ and the theoretical value for the specific-heat exponent in the three-dimensional Ising system,[20] $\alpha_p = 0.11$, Eq. (2) yields $t_r = 1.3 \times 10^{-8}$.

Riedel and Wegner[21] have predicted that the crossover region in an annealed system with $\alpha_p > 0$ is very slow in reduced temperature and extends over approximately five decades. More recently, Newman and Riedel[6] have suggested that this crossover is even slower in a quenched system, perhaps over eight decades of reduced temperature. If it is assumed that this extent of the crossover region is not a function of impurity concentration, then for data taken over our reduced temperature range of 5×10^{-4} to 2×10^{-2} we expect, on the basis of Eq. (2) to find the pure exponent, β_p, for impurity concentrations $x < 0.057$ and the random exponent, β_r, for impurity concentrations $x > 0.65$. This suggests that our data for $x = 0.136$ are in the crossover region and our value for β_{eff} is not the asymptotic random value. The appearance that our data are described by a single-power law in the range-of-fit analysis results from the fact that the crossing to the new fixed point occurs so slowly in reduced temperature. Our argument also explains why Wertheim *et al.*[8] have observed the pure exponent in MnF_2 doped with 0.7 at.% Fe.

The recent calculations of Newman and Riedel[6] have yielded $\beta_p = 0.323$ for the pure three-dimensional Ising system, consistent with renormalization-group calculations in 4-ϵ dimensions[20] which yield $\beta_p = 0.325$. For the random three-dimensional Ising system Newman and Riedel[6] have obtained $\beta_r = 0.354$. These results are summarized in Fig. 6. The effective value of the exponent is the pure value for $x < 0.057$ and the random value for $x > 0.65$. The effective value of the exponent in the crossover region depends on the shape of the order parameter curve in this region. Figure 6 shows values of β_{eff} for our data along with values for pure MnF_2 from Heller[11] and for $x = 0.007$ from Wertheim *et al.*[8] In order to make the three sets of data more comparable we would like to use values of the exponent obtained from the range-of-fit analyses with the same value of

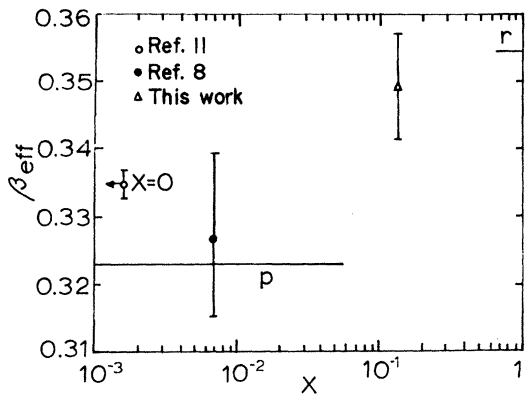

FIG. 6. Calculated values of the effective exponent, β_{eff}, over the reduced temperature range of 5×10^{-4} to 2×10^{-2}. Values of the pure and random exponent are from Ref. 6. We have assumed that the crossover region covers eight decades of reduced temperature and that $t_r = x^{|1/\alpha_p|}$. The region of pure behavior is indicated p and that of random behavior is indicated r. Values of β_{eff} from Ref. 11, Ref. 8, and this work are shown.

t_{max} for each set of data. The values of β_{eff} shown in Fig. 6 for pure MnF_2 and for $Mn_{0.864}Zn_{0.136}F_2$ are obtained from Fig. 3 with $t_{max} \simeq 0.02$. Since a range-of-fit analysis could not be performed for the $Mn_{0.993}Fe_{0.007}F_2$ data, the value of the exponent shown in Fig. 6 for this experiment is for the entire temperature range with $t_{max} = 0.06$.

In conclusion we feel that the exponent we have obtained is consistent with the predicted crossing to a new fixed point for the three-dimensional Ising system with random impurities and that this exponent represents an effective value for some part of the crossover region. A systematic study of β_{eff} for some given reduced temperature range as a function of impurity concentration in order to fill in data points on Fig. 6 could provide important information on the shape of the crossover region. Unfortunately line broadening, as illustrated in Fig. 4, will probably prohibit critical-point measurements in systems with impurity concentrations greater than about 20 at.%.

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