Random-field critical behavior of a $d=3$ Ising system: Neutron scattering studies of $Fe_{0.6}Zn_{0.4}F_2$

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The effects of random fields on the critical scattering of neutrons in a $d=3$, diluted Ising antiferromagnet have been observed for the first time. The experiments were made on the same crystal of $Fe_{0.6}Zn_{0.4}F_2$ in which earlier birefringence (Δn) studies showed a sharp phase transition $T_c(H)$ to exist. Only the scattering in the region above $T_c(H)$ was studied, where hysteretic behavior is absent. In this equilibrium region, new random-field critical behavior is observed, indicative of an approach to a sharp, second-order phase transition at $T_c(H)$, unlike the $d=2$ case where rounding of the phase transition occurs within the same region. Hence the lower critical dimensionality of the *equilibrium* random-field Ising model (RFIM) is bounded by $2 \leq d_l < 3$. The critical scattering is well described by $S(q)=A/(\kappa^2+q^2)+B/(\kappa^2+q^2)^2$, with A scaling as $\kappa^{\overline{\eta}}$ and B as $\kappa^{2\overline{\eta}}$. A preliminary analysis yields $\overline{\nu}=1.0\pm0.15$, $\overline{\gamma}=1.75\pm0.20$, and $\overline{\eta}\simeq\frac{1}{4}$ for the random-*field* thermal correlation length (κ^{-1}), staggered susceptibility (χ_{st}), and correlation function ($\langle S_0^z S_r^z\rangle$) critical exponents, respectively. κ scales with the random field h_{RF} as $\kappa \propto h_{RF}^{2(\nu-\overline{\nu})/\phi}\,|t-t_c|^{\overline{\nu}}$ as expected, with $t-t_c=(T-T_c)/T_N$ and ϕ and ν the crossover and random-*exchange* thermal correlation-length exponents, respectively. When taken together with the previously measured specific-heat critical exponent $\overline{\alpha}=0.00\pm0.03$ and amplitude ratio $\overline{A}/\overline{A}'\simeq1$, the values of $\overline{\nu}$, $\overline{\gamma}$, and $\overline{\eta}$ are consistent with an effective dimensionality $\overline{d}\simeq2$ for a $d=3$ Ising system subject to an h_{RF}. Implicit in this is a modified hyperscaling relation $\overline{d}\,\overline{\nu}=2-\overline{\alpha}$ for the RFIM. The present neutron scattering results, including the striking field dependence of the phase transition, agree with and complement the interpretation previously given to the Δn critical-behavior measurements. The conclusion obtained from earlier neutron scattering experiments below T_c, that $d_l \geq 3$, must then result from an incorrect identification of the nonequilibrium, *field-cooled* configuration with the ground state of the RFIM.

I. INTRODUCTION

The intriguing question of what is the lower critical dimensionality d_l of the random-field Ising model (RFIM) has occupied the attention of both theorists and experimentalists alike, since Imry and Ma[1] first defined the problem and suggested an answer (namely, $d_l=2$) in 1975. A review of current theories as regards d_l may be found elsewhere.[2] Of equal interest is the prediction[3,4] that, at a dimension $d>d_l$, new critical behavior will occur with critical exponents characteristic of a lower dimensional Ising system, $\overline{d}<d$.

However, it was not until Fishman and Aharony[5] (FA) showed that the randomly diluted antiferromagnet in a uniform field H, applied collinearly with the direction of spontaneous ordering, maps directly into the RFIM for the ferromagnet, were any experimental tests of the theoretical predictions made. The experimental studies have been of two kinds: first, to attempt to determine the ground state (in practice, the nature of the low-temperature state) of the RFIM either by cooling in a field or by first zero-field cooling and then applying the field. All previous neutron scattering work[6–9] has concentrated on this aspect of the problem; second, to investigate the region near the phase boundary so as to ascertain whether or not a sharp phase transition occurs. If it does, one would obviously want to determine the critical behavior associated with the RFIM fixed point. In this latter category there have been susceptibility,[10] birefringence,[11–13] specific-heat,[14] and thermal-expansion[15] studies.

Of the many experiments that have probed the RFIM, only one, the linear birefringence (Δn) determination of the magnetic specific heat[12] of the randomly diluted, $d=3$ antiferromagnet $Fe_xZn_{1-x}F_2$, has given evidence of both a sharp phase transition and of critical behavior that is characteristic of an effective dimensionality $\overline{d}\simeq2$. Similar Δn studies on the $d=2$ diluted antiferromagnet $Rb_2Co_xMg_{1-x}F_4$ have revealed[13] a progressive rounding of the specific heat with increasing field. Thus from these two investigations, it has been established that $2\leq d_l<3$ for the RFIM. (We shall return to the interpretation of the Δn studies in Sec. VI.)

Our goals in the present experiment in the vicinity of the phase boundary were threefold: (1) to see if the sharp transition found in the $d=3$ system using the Δn method was also observable with the neutron scattering technique; (2) if so, to measure the exponents $\overline{\nu}$ and $\overline{\gamma}$ associated with the thermal correlation length (κ^{-1}) and staggered susceptibility (χ_{st}), respectively, and (3) to determine whether or not the exponents were consistent with $\bar{d} \simeq 2$ and with a modified hyperscaling ($\bar{d}\bar{\nu}=2-\bar{\alpha}$).

For this first study we have concentrated on the equilibrium region above $T_c(H)$. Equilibrium is assumed to prevail, at a given T and H, when identical results are obtained for any measured quantity independent of the field and temperature cycling procedure used to arrive at that point. The most common procedures are field cooling (FC) and zero-field cooling (ZFC). It has recently been established[16] that the equilibrium boundary $T_{eq}(H)$ lies very close to but *slightly* above $T_c(H)$ and that $T_{eq}(H)$ scales as $h_{RF}^{2/\phi}$, as does $T_c(H)$. Presumably, it is the *field cooling* through $T_{eq}(H)$ that results in the freezing-in of domains which is manifest below $T_c(H)$ as the broadened Bragg peaks seen in the early neutron scattering experiments. To avoid confusing this static source of broadening with that which arises from critical fluctuations in the vicinity to $T_c(H)$, we deliberately restrict ourselves to investigating the region $T > T_{eq}(H)$.

In Sec. II the scaling and crossover behavior and the shift of $T_c(H)$ in the presence of a random field are presented. Sections III and IV give the experimental details and experimental results, respectively. In Sec. V an interpretation of the results is developed on the basis of certain simplifying assumptions and in the last section a summary is provided in which the results of the present experiment and relevant earlier ones are compared.

II. RANDOM-FIELD CRITICAL BEHAVIOR AND SCALING RELATIONS

The application of site-random fields to a uniform Ising ferromagnet has been predicted to alter both the lower critical dimensionality and the critical exponents. Such site-random fields may be defined by their quenched configurational averages, $\langle H_i \rangle = 0$, $\langle H_i^2 \rangle \equiv H_{RF}^2 > 0$ (where RF denotes random field). Hardly any experimental interest was generated, however, until FA (Ref. 5) showed that an antiferromagnet with random exchange, placed in a *uniform* field, was a physical realization of a random-field system. Wong *et al.*[14,17] first suggested an extension of the FA argument to the actual experimental situation of site dilution. Cardy[18] demonstrated a mapping of the site-diluted Ising antiferromagnet in a uniform field onto the problem of a random-field ferromagnet and identified the effective random field in the weak-field limit. The equivalence is shown to be exact in the sense that both systems belong to the same universality class.

Amongst the results of FA is the prediction that new critical behavior will be observed within a crossover region

$$|t| < h_{RF}^{2/\phi} , \tag{1}$$

where

$$t \equiv (T - T_N + bH^2)/T_N \tag{2}$$

is the reduced temperature measured relative to the mean-field (MF) phase boundary, $T_N^{MF} = T_N - bH^2$, h_{RF} is the reduced rms random field, and ϕ is the crossover exponent predicted to be equal to the $h_{RF}=0$ susceptibility exponent γ.

The mean-square reduced random field for the site-diluted case[18,12] has been shown to be

$$h_{RF}^2 = \frac{x(1-x)[T_N^{MF}(1)/T]^2 (g\mu_B SH/k_B T)^2}{[1+\Theta^{MF}(x)/T]^2} , \tag{3}$$

where $T_N^{MF}(1)$ is the mean-field T_N in the pure system, and $\Theta^{MF}(x)$ is the mean-field Curie-Weiss parameter.

If a sharp phase transition exists, the new transition temperature is expected to be

$$T_c(H) = T_N - bH^2 - T_N(ch_{RF}^2)^{1/\phi} , \tag{4}$$

where c is a constant of order unity.

The free energy was shown[5] to have a scaling form which can be written as

$$F = |t|^{2-\alpha} f(th_{RF}^{-2/\phi}) , \tag{5}$$

which exhibits the leading $|t|^{2-\alpha}$ behavior in $h_{RF}=0$ and describes the crossover to new critical behavior when $h_{RF} \neq 0$. From (5) it follows that the specific heat has the form

$$C_m \sim |t|^{-\alpha} f(th_{RF}^{-2/\phi}) . \tag{6}$$

The staggered susceptibility χ_{st} is similarly expected to exhibit the behavior

$$\chi_{st} \sim |t|^{-\gamma} f'(th_{RF}^{-2/\phi}) , \tag{7}$$

where γ is the zero-field, i.e., $h_{RF}=0$, exponent. The random-*field* behavior can be seen more clearly if we rewrite (7) to suppress the leading zero-field, i.e., $h_{RF}=0$, behavior, as

$$\chi_{st} \sim h_{RF}^{-2\gamma/\phi} g(th_{RF}^{-2/\phi}) . \tag{8}$$

If a sharp phase transition occurs, $g(y)$ is expected to behave as $|y - y_c|^{-\bar{\gamma}}$, giving

$$\chi_{st} = \bar{\chi}_{st}^0 |t - t_c|^{-\bar{\gamma}} \sim h_{RF}^{-2(\gamma-\bar{\gamma})/\phi} |t - t_c|^{-\bar{\gamma}} . \tag{9}$$

where $t_c \equiv [T_c(H) - T_N + bH^2]/T_N = (ch_{RF}^2)^{1/\phi}$, so that $t - t_c$ represents the relative departure from the actual phase boundary in any given field. The new exponent $\bar{\gamma}$ is expected to be one characteristic of an Ising system of lower dimensionality $\bar{d}$. Likewise, the inverse correlation length κ is expected to behave according to

$$\kappa \sim |t|^{\nu} f''(th_{RF}^{-2/\phi}) , \tag{10}$$

where ν is the zero-field, i.e., $h_{RF}=0$, exponent. This may be rewritten, as for χ_{st}, as

$$\kappa = \bar{\kappa}_0 |t - t_c|^{\bar{\nu}} \sim h_{RF}^{2(\nu-\bar{\nu})/\phi} |t - t_c|^{\bar{\nu}} \tag{11}$$

if a sharp transition exists, where $\bar{\nu}$ is a new random-*field* exponent.

Previous neutron scattering and birefringence experiments[19] on $Fe_xZn_{1-x}F_2$ have determined the critical exponents in *zero field* to be the random-*exchange* exponents, which are $\gamma = 1.44 \pm 0.06$, $\nu = 0.73 \pm 0.03$, and $\alpha = -0.09 \pm 0.03$ in good agreement with theoretical predictions. Since it is the random-exchange exponents, not the "pure" $d=3$ Ising ones, which describe the observed behavior of the phase transition at $H=0$, it is these quantities, with $\phi = \gamma$, which should be used in Eqs. (4)—(11) as

TABLE I. The critical exponents of the Ising $d=3$ pure, random exchange and random field systems. Only the theoretical values are given for the pure system. It is clear the exponents differ for the three cases. The pure Ising $d=2$ case is also shown to emphasize the similarity between it and $d=3$ random field results. The specific-heat amplitude ratio A/A' is shown for only the $d=3$ random field and the pure $d=2$ cases, again to emphasize the same point. Question marks indicate no theory exists, as yet.

	Ising; $d=3$					Ising; $d=2$
Critical exponents	Pure (theory)[a]	Random exchange Expt.[b]	Random exchange Theory[c]	Random field Expt.[d]	Random field Theory	pure (theory)[e]
α; A/A'	0.11	$-0.09\ \pm0.03$	$(-0.04)\ -0.09$	0.00 ± 0.03; $\simeq 1$	?	0; 1
ν	0.63	$0.73\ \pm0.03$	(0.68) 0.70	$1.0\ \pm0.15$	?	1
γ	1.24	$1.44\ \pm0.06$	$(1.34)\ -1.39$	1.75 ± 0.20	?	$\frac{7}{4}$
β	0.325	0.349 ± 0.008	(0.35) 0.35		?	$\frac{1}{8}$
η[f]	0.02	$0.02\ \pm0.07$	(0.02) 0.01	$\sim\frac{1}{4}$	?	$\frac{1}{4}$

[a]J. C. LeGuillou and J. Zinn-Justin, Phys. Rev. B **13**, 3081 (1976) and other references therein.
[b]Reference 19 and R. A. Dunlap and A. M. Gottlieb, Phys. Rev. B **23**, 6106 (1981).
[c]K. E. Newman and E. K. Riedel, Phys. Rev. B **25**, 264 (1982); values in parentheses are from G. Jug, *ibid.* **25**, 609 (1983).
[d]Value of α from Ref. 12; all others from this work.
[e]L. Onsager, Phys. Rev. **65**, 117 (1944).
[f]η calculated from $\eta=2\nu-\gamma$ except for random-field value.

the $h_{RF}=0$ exponents. The experimental and theoretical values of the random-exchange exponents are collected together in Table I.

The random-field critical behavior observed in the Δn experiments indicates a sharp phase transition with $\bar{\alpha}\simeq 0$, and amplitude ratio $\bar{A}/\bar{A}'\simeq 1$, which are the values expected and observed in a *pure* $d=2$ Ising system.[20] The field dependence of $T_c(H)$ is given by Eq. (4), with $\phi=1.40\pm0.05$ in agreement with the random-exchange value of γ, as expected. If the other exponents are also characteristic of $\bar{d}\simeq 2$, i.e., $\bar{\gamma}=1.75$, $\bar{\nu}=1.0$, and $\bar{\eta}=0.25$, this fact should be readily observable in a neutron scattering experiment. Implicit in this concept is the related assumption that a modified hyperscaling relation holds amongst the random-field exponents; i.e., $\bar{d}\bar{\nu}=2-\bar{\alpha}$. We shall return to the matter of the effective dimensionality and the current state of theoretical predictions of it in the summary, Sec. VI.

III. EXPERIMENTAL DETAILS

Since concentration gradients in mixed crystals are the single most important factor in establishing the sharpness of the phase transition, even in the absence of random-field effects (and more so in their presence), it was decided to employ the very same crystal of $Fe_{0.6}Zn_{0.4}F_2$ used in the Δn experiment, from which the best and most detailed results on the magnetic specific-heat critical behavior were obtained. Use of this crystal also allowed for a direct comparison of the striking field dependence of the transition temperature $T_c(H)$ as measured by two different techniques. This particular sample of $Fe_{0.6}Zn_{0.4}F_2$ is approximately $4\times5\times7$ mm, with the largest dimension parallel to the c axis. It is also the growth axis, which is the direction along which the concentration gradient is the largest. The gradient was measured by utilizing the difference in the optical birefringence of FeF_2 and ZnF_2 at ambient temperatures. The variation in Δn along the growth axis is shown in Fig. 1. The laser beam was oriented perpendicular to the scan direction, as indicated in the lower inset to Fig. 1. Although the average gradient, as determined from Δn measurements, is only 0.65%/cm, it does produce a variation of the transition temperature of approximately 0.35 K across the sample in zero applied field.

Two separate experimental arrangements were used to collect data. Initially, the crystal was mounted on an aluminum holder with nearly the whole of it exposed to the beam. The platinum thermometer and control heater were situated about $\frac{1}{4}$ m from the sample, outside the superconducting solenoid, to reduce the field effect on the temperature reading. We denote this arrangement as configuration A, for which data were obtained at $H=0$, 1.4, 2.0, and 5.5 T. Having confirmed that critical scattering was observable, the experimental arrangement was im-

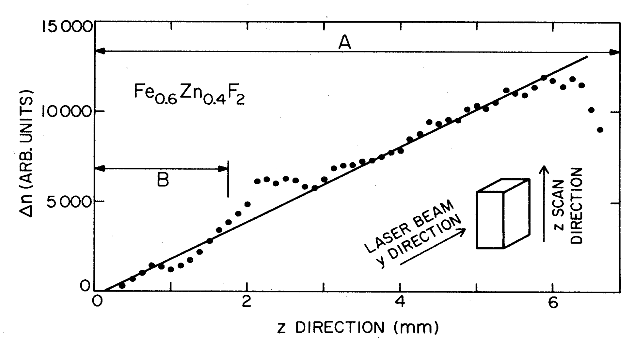

FIG. 1. Variation of the optical birefringence Δn at ambient temperature along growth (z) axis. The total variation corresponds to a variation in concentration $\delta x=0.45$ mol %. The entire crystal was exposed to the neutron beam in configuration A, while only about $\frac{1}{4}$ was exposed in configuration B, as indicated.

proved as follows: The sample was masked with cadmium for all but 1.5 mm of its length along the c axis. This results in a variation δT_N of only 0.08 K in T_N, as a consequence of the concentration gradient. We denote this arrangement as configuration B, for which data were taken at $H=0$, 1.4, and 2.0 T. (Unfortunately, as we shall see, time did not permit a run at 5.5 T.) Additionally, a carbon-glass thermometer, which has only a slight field dependence, was placed on a copper block about which a separate control heater was wound, permitting a temperature stability of 30 mK to be achieved. The temperature scale for the carbon-glass thermometer was adjusted so that the 2.0-T data agreed within 100 mK with that obtained using the calibrated platinum thermometer. The regions of the sample that were exposed to the neutron beam in configurations A and B are schematically indicated in Fig. 1.

The neutron scattering was performed with a two-axis spectrometer at the high flux beam reactor at the Brookhaven National Laboratory. The data are reported here and in part in the following paper by Yoshizawa *et al.*[21] The sample was oriented with [001] (c axis) vertical and the field applied in this direction. The lattice parameters for $Fe_{0.6}Zn_{0.4}F_2$ are $c=3.24$ Å and $a=b=4.71$ Å. The incident wave vector was $\kappa_i=2.67$ Å^{-1} and all collimations were 10′ of arc, giving a measured resolution half-width at half maximum (HWHM) of 0.000 75, 0.0025, and 0.025 reciprocal lattice units (r.l.u.) in the transverse, longitudinal, and vertical directions, respectively. Nearly all data were taken in transverse scans near the (100) magnetic lattice point. The range of q scanned was $|q| \leq 0.03$ at 5.5 T and $|q| \leq 0.1$ at 1.4 and 2.0 T.

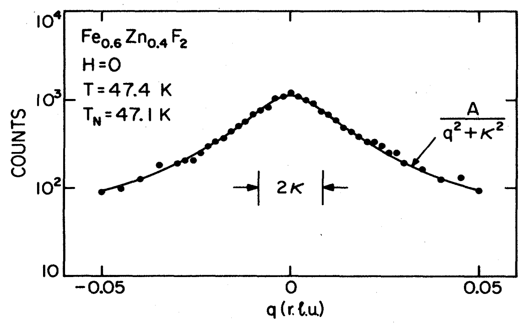

FIG. 3. The observed scattering $S(q)$ vs q for transverse scans at (001) in $Fe_{0.6}Zn_{0.4}F_2$ at $H=0$ close to, but slightly above, T_N. $S(q)$ is well fit by a Lorentzian line shape as is indicated by the solid line.

IV. EXPERIMENTAL RESULTS

In Fig. 2 a plot is shown of the uncorrected HWHM of the scattering function versus temperature for $H=0$, 1.4, and 2.0 T. These data were all taken in configuration A. In a field the HWHM vs T is seen to be linear whereas at $H=0$ it curves downward as T approaches T_N. The general behavior of the latter is in agreement with a previous study of the $H=0$ random-exchange problem in $Fe_{0.5}Zn_{0.5}F_2$.[19]

One obvious conclusion that can be drawn from Fig. 2, within the limits set by instrumental resolution, is that the HWHM in a field appears to approach zero linearly as the temperature decreases *without any evidence of rounding.* (One must recognize that at these low fields the contribution to the HWHM that would arise from the field-cooled domain state below $T_{eq}(H)$ is less than the resolution limit.) Moreover, the values of temperature for which the HWHM extrapolates to zero at $H=1.4$ and 2.0 T agree

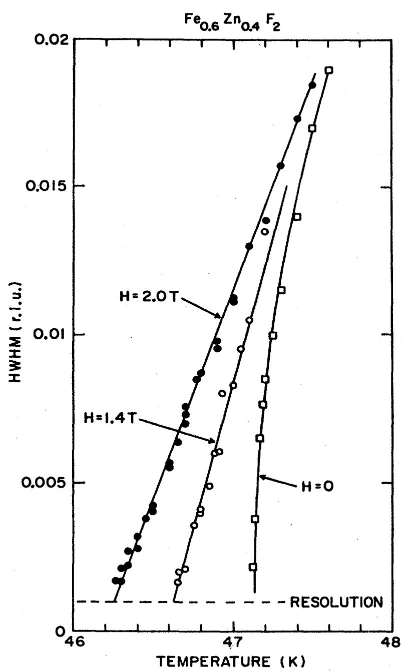

FIG. 2. The uncorrected half-width at half maximum (HWHM) of $S(q)$ vs temperature at $H=0$, 1.4, and 2.0 T. Over the range of temperatures measured, it is clear that the HWHM vs T is linear, within experimental error, for $H\neq0$. The $H=0$ results are in accord with earlier studies of the random-exchange problem (Ref. 19). All of the data shown in the figure were collected using configuration A.

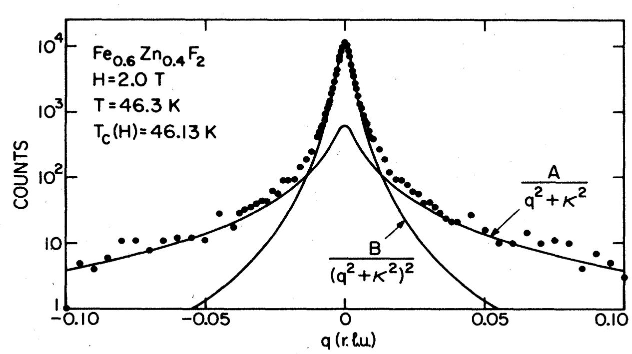

FIG. 4. The observed scattering $S(q)$ vs q, now at $H=2.0$ T and close to, but slightly above, $T_c(H)$. $S(q)$ is well described by Eq. (15). The separate contributions from the Lorentzian and Lorentzian-squared terms that provide the best fit are given by the solid lines. The values of κ, A, and B determined at this field and temperature were then used in the analyses of the critical behavior.

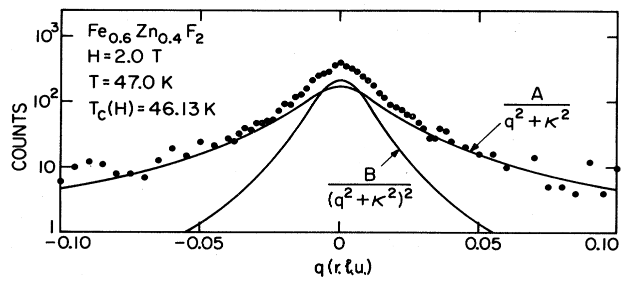

FIG. 5. The observed scattering $S(q)$ vs q under the same conditions as in Fig. 4 except for $T-T_c(H)$ being slightly larger. Note that the relative size of the Lorentzian and Lorentzian-squared contributions change as $T-T_c(H)$ varies as can be seen from a comparison of the indicated best-fit solid lines for the two terms in this figure with the corresponding ones in Fig. 4.

with those at which the peak in $d\Delta n/dT$ vs T occurs, within the accuracy of the present measurements. Thus there is strong evidence from the neutron scattering experiments of an approach to a sharp, second-order phase transition, opening the possibility of determining the critical behavior associated with the RFIM fixed point. This we next do below.

The scattering line shapes for $H=0$ at $T=47.4$ K and for $H=2.0$ T at $T=46.3$ K and $T=47.0$ K, when folded with an appropriate instrumental resolution function and fitted to the data, are shown in Figs. 3, 4, and 5, respectively. The $H=0$ data are well fit by a Lorentzian line shape

$$S(q)=\frac{A}{q^2+\kappa^2} \tag{12}$$

for reduced wave vectors $0.005\leq|q|\leq0.05$. However, the scattering in a field is clearly *not* Lorentzian but it may be quite adequately represented by a Lorentzian plus a Lorentzian-square line shape

$$S(q)=\frac{A}{q^2+\kappa^2}+\frac{B}{(q^2+\kappa^2)^2} \tag{13}$$

for all values of T and $H>0$ with $\chi^2\simeq1$. We discuss the choice of the fitting functions in Sec. V, where the interpretation of the configuration B data at 1.40 and 2.0 T and the configuration A data at 5.5 T is given.

V. INTERPRETATION

Since no theory exists for the exact form of the structure factor $S(q)$ in the presence of random fields, some assumptions have to be made in order to analyze the data. The wave-vector-dependent susceptibility $\chi(q)$ is assumed to have the Ornstein-Zernike form

$$\chi(q)\propto\frac{1}{\kappa^2+q^2}\ , \tag{14}$$

which has the usual Lorentzian q dependence. Its amplitude is required by scaling to be proportional to κ^η.

Second, in the presence of a random field $S(q)$ is expected, on the most general grounds,[22] to have, in addition to the Lorentzian contribution, a Lorentzian-squared term

$$S(q)=\frac{A}{\kappa^2+q^2}+\frac{B}{(\kappa^2+q^2)^2}\ , \tag{15}$$

where scaling requires that the coefficient $A\sim\kappa^\eta$. At present, no theory exists for the behavior of B in the critical region. However, the form of the Lorentzian-squared term suggests that it might scale as $\chi^2(q)$, apart from any other dependence on H. We assume this to be the case. If it did scale as simply as this, then one might expect $B\sim\kappa^{2\eta}$ along with an additional dependence on H. In both the pure and random exchange $d=3$ cases, η is very small and is usually ignored. However, for the $d=3$ Ising random-field case, $\bar{\eta}$ might be as large as 0.25, as was discussed in Sec. II, and its effect on A and B cannot be overlooked. (It should be noted that in the analysis to be made here everything that has been observed is related to critical scattering and κ^{-1} is the true thermal correlation length. On the other hand, the Lorentzian-squared term that appears in the low-temperature analyses[6,7] of the *field-cooled*-induced domain structure is associated with Bragg scattering from an inhomogeneous medium.[23] The meaning of κ^{-1} in that instance is some length characteristic of the mean size of the domain structure.)

Although the assumed form of the structure factor seems reasonable, based on pure system behavior and simple modification due to randomness, it is *not* exact, and it should be recognized that all subsequent analysis hinges on this fact and must be qualified accordingly.

As regards the $H=0$ data, no analysis was made other than to verify the Lorentzian form to $S(q)$ vs q at all temperatures, since this problem (i.e., random exchange) has been previously studied.[19] For $H\neq0$, $S(q)$, as given by Eq. (15), was used to fit all of the data at $H=1.4$, 2.0, and 5.5 T to obtain κ, A, and B as a function of field and temperature. The range of q over which the data was analyzed was $0.005\leq|q|\leq0.1$ at 1.4 and 2.0 T and $0.005\leq|q|\leq0.03$ at 5.5 T. The quality of the fits can be judged from the fact that $\chi^2\simeq1$ for nearly all of the scans.

Figure 6 shows plots of κ vs T as obtained from the analysis of the configuration-B data at $H=1.4$ and 2.0 T, and the 5.5-T configuration-A data. It is immediately evident that κ is linear in temperature at 1.4 and 2.0 T, just as is the HWHM (see Fig. 2). It appears that there is a sharp phase transition and new critical behavior characteristic of a new fixed point. In view of the previous evidence for the supposed destruction of the phase transition at $d=3$ because of the formation of a domain state in a FC procedure,[6,7] it might appear surprising to see evidence for a sharp transition. However, at these lower fields, the freezing into a metastable domain state at $T_{eq}(H)$ during FC occurs so close to $T_c(H)$ that it is within the temperature resolution of the data. The domain size it would produce results in an effective κ well below the instrumental resolution limit. The rounding that would be expected from the smearing $\delta T_c(H)$ of $T_c(H)$ in the B configuration would also lie within the temperature resolution. Both of these features are illustrated in Fig. 6 for the $H=1.4$ at 2.0 T data.

Although the κ vs T plot at 5.5 T exhibits an extended linear region, it is apparent that rounding does occur as

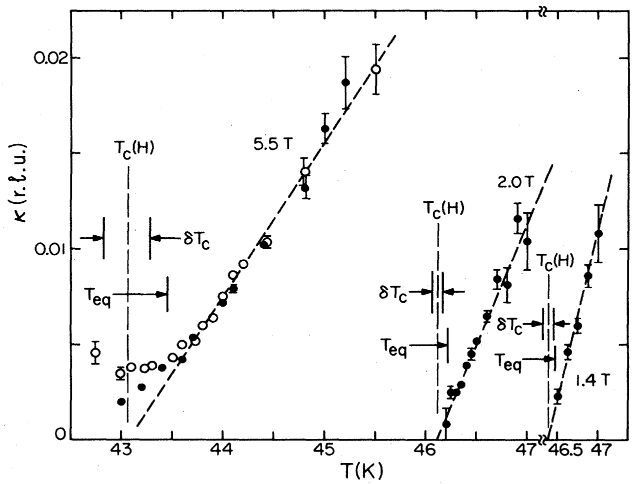

FIG. 6. Temperature dependence of κ as determined from the best fits of $S(q)$ to Eq. (15) at 1.4, 2.0, and 5.5 T. The straight lines drawn through the data are discussed in the text. The data at 5.5 T includes both field-cooled (FC) (●) and zero-field-cooled (ZFC) (○) points. The rounding in the ZFC data at 5.5 T is attributed to the indicated variation δT_c in $T_c(H)$ arising from the concentration gradient, as discussed in the text. The rounding in the FC data at 5.5 T is due to a combination of δT_c and freezing at $T_{\rm eq}(H)$. Both of these effects are negligible in the lower-field configuration-B data.

$T_c(H)$ is approached. However, two points have to be considered before one concludes anything from the apparent rounding. The first is that the *entire* crystal was exposed to the beam at this higher field which causes a smearing of the transition temperature $\delta T_c \simeq 0.35$ K even at $H=0$. The second is that additional rounding occurs in a field because of the dependence of the random-field-induced shift ΔT_c of $T_c(H)$ on the concentration x, at a given H. From Eq. (4) one may show

$$\Delta T_c \propto T_N[x^{-3}(1-x)H^2]^{1/\phi} \qquad (16)$$

and that the variation in ΔT_c that occurs at 5.5 T amounts to an additional smearing of $\delta T_c \simeq 0.10$ K. Thus in total a rounding of the transition of $\delta T_c \simeq 0.45$ K is expected. This is indicated in Fig. 6 along with the position of $T_{\rm eq}(H)$ relative to $T_c(H)$ at 5.5 T.

What is the effect that the smearing of $T_c(H)$ has on $S(q)$ and the observed κ vs T, in particular within the critical region? If there were no smearing of $T_c(H)$ then, in the ZFC case, the contribution to κ from critical scattering would vanish as T approaches $T_c(H)$ from above or below. In the presence of a concentration gradient, there is always a part of the crystal with $\kappa \neq 0$ in the region around $T_c(H)$. The experimentally determined $\kappa(T)$ therefore represents an average over the distribution of κ's in the region. This explains why κ appears not to vanish and has a minimum near $T_c(H)$. In addition, when $T < T_c(H)$, for some part of the sample, a resolution-limited Bragg peak appears as well. Since the entire peak is contained in the range $|q| < 0.005$ r.l.u. excluded in the analysis of the data, it has no effect on the analysis of κ arising from the critical scattering.

Even if there were no smearing of $T_c(H)$, then in the FC case below $T_{\rm eq}(H)$ the scattering would consist of a broadened contribution from the "elastic" part arising from the frozen domain structure and possibly some inelastic one. As in the ZFC case, we have excluded the region $|q| < 0.005$ r.l.u. However, this is not sufficient to eliminate all of the tails of the elastic scattering. (There is no obvious $|q|$ at which the two may be differentiated.) Despite the fact that elastic scattering in the FC case is broader than the Bragg peak when ZFC, it is still *narrower* than the minimum κ's derived in the ZFC case. This explains the fact that the apparent FC κ's are smaller than the corresponding ZFC ones. The smearing of $T_c(H)$ only acts to broaden the region over which the freezing occurs.

In Fig. 6 we have arbitrarily drawn straight lines (i.e., $\bar{\nu}=1.0$) through the κ-vs-T data which intersect the T axis at the values of $T_c(H)$ measured by the birefringence experiments at $H=1.4$ and 2.0 T and that determined by extrapolation of Eq. (4) to $H=5.5$ T. It is immediately apparent that the assumption of $\bar{\nu}=1.0$ results in a quite adequate fit to the data, and that the field dependence of $T_c(H)$ as measured by neutron scattering is consistent with the Δn measurements. This is also the value of ν for the pure $d=2$ Ising system. However, we regard the data as preliminary and not accurate enough to justify a determination of $\bar{\nu}$ via a least-squares analysis. Instead, we have roughly estimated the limits of the possible variation of $\bar{\nu}$ to be ± 0.15 and observe that the data are consistent with $\bar{\nu}=1.0\pm 0.15$.

The field dependence of $\bar{\kappa}_0$ is predicted by the scaling relation Eq. (11). Using the value of the random-exchange exponent $\nu=0.73\pm 0.03$ from Table I and

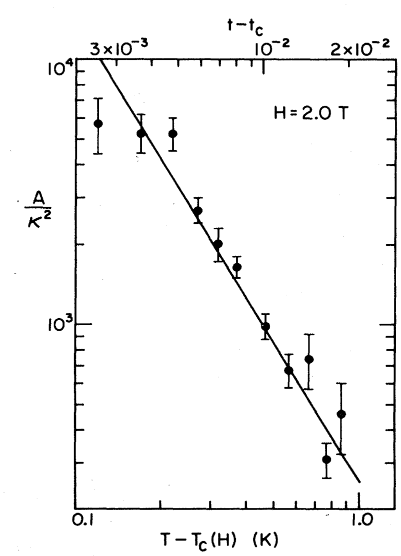

FIG. 7. $\chi_{\rm st} \propto A/\kappa^2$ vs $T-T_c(H)$ at $H=2.0$ T. The critical divergence of $\chi_{\rm st} \propto |t-t_c|^{-\bar{\gamma}}$; the straight line through the data has a slope corresponding to $\bar{\gamma}=1.75$. The reduced temperature scale is shown above.

choosing the values $\bar{\nu}=1.0\pm0.15$ and $\phi=1.40\pm0.05$, we find $\bar{\kappa}_0 \propto H^{0.4\pm0.2}$. This is to be compared with the approximate $H^{0.6\pm0.2}$ dependence of the slopes of the lines through the data in Fig. 6. The agreement is satisfactory considering the errors in the various exponents.

Since χ_{st} is given by Eq. (15) with $q=0$, we may determine its temperature dependence from that of A/κ^2 since $A/\kappa^2 \propto \chi_{st}=\bar{\chi}^0_{st}|t-t_c|^{-\bar{\gamma}}$. A log-log plot of A/κ^2 vs $|t-t_c|$ is shown in Fig. 7, for the $H=2.0$ T data, with the value of T_c chosen to be the same as the extrapolation of κ to zero in Fig. 6. The straight line corresponds to a value of $\bar{\gamma}=1.75$. This is also the value of γ for the pure $d=2$ Ising system. Again, we have not attempted a least-squares fitting but have estimated the errors to be ±0.20.

The temperature dependence of the amplitude of the Lorentzian-squared component of the scattering profile may be found from a log-log plot of B/κ^4 vs $|t-t_c|$, as shown in Fig. 8, for the $H=2.0$ T data using the same value of T_c as above. We find a very reasonable fit is obtained with a line which has a slope equal to 3.50. This is just twice the value of γ for the pure $d=2$ Ising system. This result lends strong support to the identification of the Lorentzian-squared term in Eq. (15) with χ^2_{st}. The estimated error for the slope is ±0.30. The measured coefficients A and B of Eq. (15) are shown in Figs. 9 and 10. We observe a definite temperature dependence in all the data, but which is more obvious in the data for B than for A, and is stronger at higher fields. The variation in the 5.5-T data for B is particularly striking! This temperature dependence implies that $\bar{\eta}$ is not negligible. Since scaling requires $A \sim \kappa^{\bar{\eta}}$ and having found $B/\kappa^4 \propto |t-t_c|^{-2\bar{\gamma}}$, and hence to χ^2_{st}, scaling requires that $B \propto \kappa^{2\bar{\eta}}$ and thus the quantities $A\kappa^{-\bar{\eta}}$ and $B\kappa^{-2\bar{\eta}}$ should be temperature independent.

It is possible to make this correction without refitting the data. Since the errors in the data are too great to justify attempting a determination of $\bar{\eta}$, we choose a value that is consistent with $\bar{d}\simeq2$, as already deduced from the measured $\bar{\alpha}\simeq0$, $\bar{A}/\bar{A}'\simeq1$, $\bar{\nu}\simeq1$, and $\bar{\gamma}\simeq\frac{7}{4}$, i.e., $\bar{\eta}=\frac{1}{4}$. The scaled quantities are plotted in Figs. 9 and 10 and show a noticeably reduced temperature dependence, although at the expense of increased error bars, due to the relative inaccuracy of κ close to $T_c(H)$. Finally, since we have determined $\kappa \propto |t-t_c|^{\bar{\nu}}$, we plot in Figs. 9 and 10 the quantities $A(t-t_c)^{-\bar{\nu}\bar{\eta}}=A(t-t_c)^{-1/4}$ and $B(t-t_c)^{-2\bar{\nu}\bar{\eta}}=B(t-t_c)^{-1/2}$. This results in smaller error bars but with as good or better temperature independence.

This lack of temperature dependence to the scaled A indicates that indeed $\bar{\eta}$ must be close to $\frac{1}{4}$. The fact that the scaled B is also temperature independent supports the contention that $B/\kappa^4 \propto \chi^2_{st}$. Note again that whatever the effect of $\bar{\eta}$ is on A and B it does not change the Lorentzian plus Lorentzian-squared dependence to $S(q)$. The rough constancy of $A|t-t_c|^{-1/4}$ and $B|t-t_c|^{-1/2}$ with T is indicated by the dashed lines in Figs. 9 and 10 which we designate as A'_0 and B'_0, respectively.

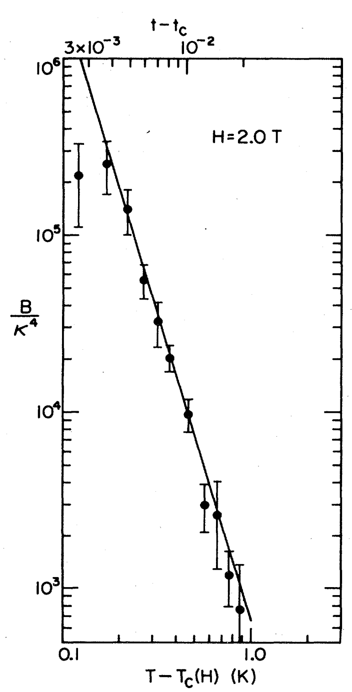

FIG. 8. The amplitude B of the Lorentzian-squared contribution to $S(q)$ divided by κ^4 vs $T-T_c(H)$. If B/κ^4 were proportional to χ^2_{st}, then one would expect its critical divergence to be proportional to $|t-t_c|^{-2\bar{\gamma}}$. The straight line through the data has a slope of 3.50 which is just equal to $2\bar{\gamma}$.

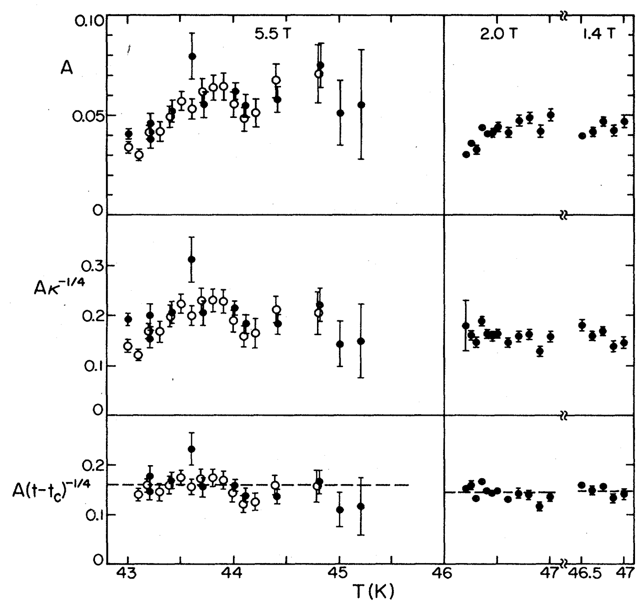

FIG. 9. The amplitude A, $A\kappa^{-1/4}$, and $A|t-t_c|^{-1/4}$ vs temperature for the scattering at $H=1.4$, 2.0, and 5.5 T. The rescaling of A by either $\kappa^{-1/4}$ or $|t-t_c|^{-1/4}$ results in much less temperature dependence to A. The data at 5.5 T includes both field-cooled (●) and zero-field-cooled (○) points. The dashed line in the plot of $A|t-t_c|^{-1/4}$ vs T for each of the three fields is a rough measure of the constancy of this quantity with T, at a given H, and is designated by A'_0 in Fig. 11.

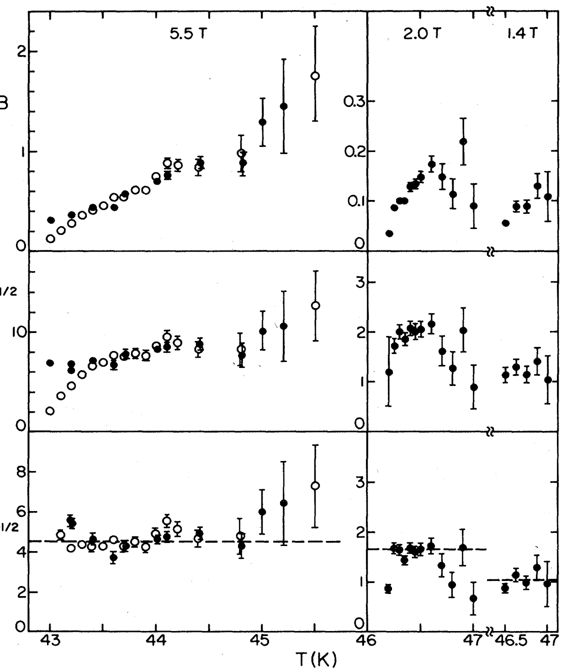

FIG. 10. The amplitude B, $B\kappa^{-1/2}$, and $B\,|t-t_c|^{-1/2}$ vs temperature for the scattering at $H=1.4$, 2.0, and 5.5 T. The rescaling of B by either $\kappa^{-1/2}$ or $|t-t_c|^{-1/2}$ results in much less temperature dependence to B. The data at 5.5 T includes both field-cooled (●) and zero-field-cooled (○) points. The dashed line in the plot of $B\,|t-t_c|^{-1/2}$ vs T for each of the three fields is a rough measure of the constancy of this quantity with T, at a given H, and is designated by B'_0 in Fig. 11.

In principle the field dependence of A and B could be checked against the scaling relations, as was accomplished for κ. Because the data at different fields was taken with different configurations, this was not possible to do. However, the ratio B/A can be studied as a function of H.

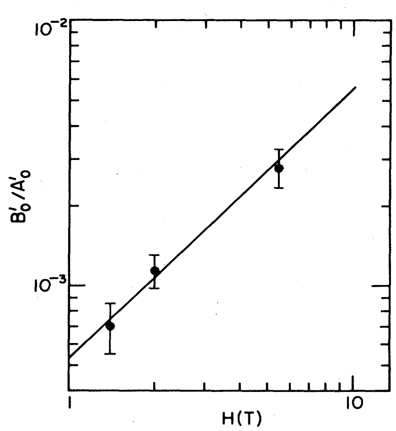

FIG. 11. The ratio B'_0/A'_0, as determined from Figs. 9 and 10, vs H. The solid line drawn through the data has a slope of 1.0±0.3. A discussion of this is given in the text.

Using Eqs. (9) and (11) and knowing $A \propto \chi_{st}\kappa^2$, one can show that

$$A \propto h_{RF}^{2(\eta-\bar{\eta})/\phi}\,|t-t_c|^{\bar{\eta}} \propto H^{-0.31}\,|t-t_c|^{1/4}\,, \qquad (17)$$

where we have used the values of the exponents from Table I and have chosen $\bar{\eta}=\frac{1}{4}$. Likewise, assuming $B \propto \chi_{st}^2\kappa^4$, it then follows that

$$B \propto h_{RF}^{4(\eta-\bar{\eta})/\phi}\,|t-t_c|^{2\bar{\eta}} \propto H^{-0.62}\,|t-t_c|^{1/2}\,. \qquad (18)$$

From Eqs. (17) and (18), it follows that

$$B\,|t-t_c|^{-1/2}/A\,|t-t|^{-1/4} \sim H^{-0.31}\,. \qquad (19)$$

In Fig. 11 we plot the quantity B'_0/A'_0 vs H, where A'_0 and B'_0 are the best-fit values to the plots of $A(t-t_c)^{-1/4}$ and $B(t-t_c)^{-1/2}$ vs T of Figs. 9 and 10, respectively, and is shown by the dashed line in each case. The slope of B'_0/A'_0 vs H in Fig. 11 has the value 1.0±0.3 and corresponds to an additional field dependence to B of $H^{1.3\pm0.3}$. From a simple mean-field picture, one would expect this dependence to be $B \propto H^2$. However, without a theory for the ratio B/A, perhaps one should not be alarmed by an apparent disagreement with mean-field theory.

In making the interpretation that was given above, we are painfully aware of both the preliminary nature of the experimental results and the absence of a first-principles theory of $S(q)$ in the presence of a random field. Nevertheless, we regard the results obtained as being so strongly suggestive of $\bar{d}\simeq 2$ as to encourage us to make a more thorough and detailed neutron scattering study in the near future.

VI. SUMMARY AND RELATION TO OTHER STUDIES

The primary results of this experiment are as follows. (1) An approach to a sharp phase transition is observed in critical scattering experiments in a $d=3$ RFIM system with new exponents characteristic of the random-field fixed point, corresponding to an effective dimensionality $\bar{d}\simeq 2$. (2) No experimental evidence for the rounding of the phase transition is observed *in the equilibrium region* $T>T_{eq}(H)$ above $T_c(H)$; hence, by implication, the lower critical dimension of the RFIM $d_l<3$. (3) The critical scattering above $T_c(H)$ in a field is *not* Lorentzian but may be quite adequately described by the sum of a Lorentzian plus a Lorentzian-squared term. (4) Preliminary analysis of the critical scattering yields $\bar{\nu}=1.0\pm0.15$, $\bar{\gamma}=1.75\pm0.20$, and requires $\bar{\eta}\simeq\frac{1}{4}$ for the random-field thermal correlation length, staggered susceptibility and correlation function critical exponents, respectively. κ scales with the random field h_{RF} as $\kappa \propto h_{RF}^{2(\nu-\bar{\nu})/\phi}\,|t-t_c|^{\bar{\nu}}$ with ϕ and ν the crossover and random-exchange thermal correlation length exponents, respectively.

The earlier birefringence studied on the same crystal[12] showed that (1) a sharp phase transition in a field is apparent in the magnetic specific heat; thus $d_l<3$. (2) The critical divergence of the specific heat $C_m=\bar{A}\,|t-t_c|^{-\bar{\alpha}}$ for $T>T_c$ and $C_m=\bar{A}'\,|t-t_c|^{-\bar{\alpha}}$ for $T<T_c$ yielded $\bar{A}/\bar{A}'\simeq 1$ and $\bar{\alpha}=0.00\pm0.03$. From this came the first suggestion[11,12] that the new effective dimensionality of a

$d=3$ Ising system subject to a random field was $\bar{d}\simeq2$. (3) Crossover from random-exchange to random-field behavior was observed in the reduced temperature region $|t|<h_{RF}^{2/\phi}$ as in Eq. (1) above. The shift in $T_c(H)$ with H agrees with Eq. (4) with $\phi=\gamma$, the random-exchange susceptibility exponent, as predicted by FA.[5]

The critical behavior observed in both experiments is consistent with the Fishman-Aharony-Cardy mapping of the site-diluted antiferromagnet in a uniform field on to the ferromagnet in a random field.[5,18] From the combined results of the two experiments, it appears that all of the measured exponents ($\bar{\alpha}$, $\bar{\gamma}$, $\bar{\nu}$, and $\bar{\eta}$) and the specific-heat amplitude ratio $\bar{A}/\bar{A}'$ are consistent with an effective dimensionality $\bar{d}\simeq2$ and a modified hyperscaling relation $\bar{d}\bar{\nu}=2-\bar{\alpha}$.

The two experiments establish that $d_l<3$ for the equilibrium RFIM. Taken together with the complementary birefringence experiment[13] on the $d=2$ randomly diluted Ising antiferromagnet $Rb_2Co_{0.85}Mg_{0.15}F_4$, in which a progressive rounding of the transition was observed with increasing field beginning at a value of h_{RF} very much smaller than those generated in the present experiment, one can establish the following bounds on d_l:

$$2\leq d_l<3\;. \qquad (20)$$

Since the present experiments were completed and this paper first submitted for publication, several related studies have been made on the same $d=2$ and $d=3$ RFIM systems.[16,24,25] They have had as their focus the determination of the relative stability of the FC and ZFC states at both dimensions. At $d=2$ the metastability boundary $T_F(H)$ is found to lie well below the region of the destroyed phase transition and the ZFC (metastable) state is observed to decay logarithmically with time toward the equilibrium domain state in the region around $T_F(H)$.[16,24] Just the opposite is observed at $d=3$. The ZFC state is stable for *all* T and H below $T_c(H)$, whereas the FC (metastable) state shows irreversible behavior with respect to variations in T or H below $T_c(H)$ (Refs. 16 and 25) and close to $T_c(H)$ decays logarithmically with time towards the equilibrium antiferromagnetic ZFC state.[16]

Furthermore, as mentioned earlier, it has been demonstrated that $T_{eq}(H)$ lies slightly above $T_c(H)$ in contrast with what occurs for the $d=d_l=2$ RFIM system. This fact explains how the FC procedure always results in a metastable domain state, even for $d>d_l$. Indeed, $T_{eq}(H)$ lying above $T_c(H)$ is a necessary condition for the freezing of a metastable state above a sharp phase transition at $T_c(H)$. Since the antiferromagnetic state is the ground state, however, one can use the ZFC route and presumably see sharp critical behavior below $T_c(H)$ as well.

Thus, with regard to which is the lower energy state, the evidence appears to be overwhelming that at $d=3$ the ordered antiferromagntic state lies lowest and all domain states are metastable with respect to it below $T_c(H)$. It is also clear now why the interpretation given to the earlier neutron scattering studies[6] on $d=3$ RFIM systems that the existence of a domain state upon FC "demonstrates unambiguously that $d_l\geq3$" is incorrect; namely, FC results in the creation of domain configurations that are metastable.

Recent Monte Carlo studies[26] have shown that domains are formed in the RFIM when FC through $T_c(H)$, regardless of d, and that they are extremely long-lived. The stability of such quenched in domains has also been investigated theoretically.[27–29] It is found that domains larger than a certain size are metastable, while smaller ones decay with a characteristic $\log t$ behavior, such as has recently been observed.[16]

As to the current theoretical beliefs with respect to d_l, there appears to be a consensus[2] that $d_l=2$; in fact, Imbrie[30] claims to have a rigorous proof of this at $T=0$ K.

The situation with respect to $\bar{d}$ is less clear. As yet, there is no formalism which directly yields $\bar{d}$ nor any of the critical exponents for the $d=3$ RFIM. However, it has recently been suggested[31] if $\bar{\alpha}\leq0$ that $\bar{d}=(d+1/\bar{\nu})/2$, which agrees both with our results at $d=3$ and the prediction of $d_l=2$. An attempt has been made to calculate the critical exponents of the $d=3$ RFIM using high-temperature series-expansion methods.[32] Clearly, much remains to be done from the theoretical side in understanding $\bar{d}$ at accessible dimensions.

From the experimental side, it would be desirable to have more accurate critical behavior studies both below and above $T_c(H)$. These would require having available crystals with yet smaller concentration gradients than was the case in the present study.

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