

Random-field-induced destruction of the phase transition of a diluted two-dimensional Ising antiferromagnet: $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$

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Optical birefringence has been used to study the effects of a uniform field H on the magnetic specific heat C_m near the phase transition of a randomly diluted, two-dimensional (2D) Ising antiferromagnet, $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$. For $H=0$, a well-defined transition is observed with a symmetric logarithmic divergence in C_m , corresponding to the critical exponent $\alpha=0$, as is predicted for the 2D Ising random-exchange case. With \vec{H} applied parallel to the c axis, a systematic rounding of the peak in C_m occurs which increases with increasing H . This demonstrates that induced random fields destroy the phase transition and that the lower critical dimensionality for the random-field problem $d_l \geq 2$. No rounding is found with $\vec{H} \perp c$, as expected. The peak in C_m with $\vec{H} \parallel c$ exhibits a decrease in amplitude, which varies as $\ln H$. This behavior, together with the $\ln |t|$ dependence of C_m at $H=0$, was used to determine the crossover exponent $\phi = 1.58 \pm 0.22$. This value is in essential agreement with the exactly known critical exponent of the staggered susceptibility γ for the 2D Ising problem, namely $\gamma = \frac{7}{4}$, as predicted theoretically. By suitable rescaling, the rounded peaks found for different values of H may all be collapsed to a single curve in the critical region, in accord with renormalization-group scaling relations. The shift and rounding (and hence the peak amplitude) are in quantitative agreement with the site-diluted, random-field predictions.

I. INTRODUCTION

Although there has been considerable theoretical and experimental interest in various aspects of the random-field problem, the one issue about which the most controversy exists is that of the lower critical dimensionality d_l . In the initial work on the subject, Imry and Ma¹ showed that the Ising model in a random field is unstable with respect to formation of domains with smooth walls for $d \leq d_l = 2$. Subsequently^{2,3} roughening of the walls was shown to result in $d_l = 3$ with a domain-wall width comparable with the domain size. More recent calculations⁴⁻⁶ showed that the effect of roughening was probably overestimated, and that d_l appears to be 2.

Although the ultimate experimental determination of d_l for Ising systems in a random field is of great importance, there appears to be no doubt that $d_l \geq 2$. Hence, experiments on two-dimensional (2D) Ising systems are not expected to resolve this question. However, a systematic study of a phase transition, which is unquestionably destroyed by a random field, would be a valuable comparison for deciding whether this does or does not occur in the three-dimensional (3D) case. Such a study would, of course, be extremely interesting in its own right.

Some of the most dramatic effects of random fields are those predicted by Fishman and Aharony (FA) (Ref. 7) on the critical behavior. These include a rapid lowering of T_c with H and, within a crossover region, a new critical behavior characteristic of a lower effective dimensionality. A physically realizable example of a random-field system

was shown to be a randomly diluted antiferromagnet in a uniform applied field. Although no sharp phase transition is expected in the 2D case, the scaling relations should describe the systematics of the field-induced rounding of the transition. Previous experimental work on 2D random-field systems include neutron scattering^{8,9} and susceptibility¹⁰ studies of $\text{Rb}_2\text{Co}_x\text{Mg}_{1-x}\text{F}_4$. Both gave indications of the destruction of long-range order by a random field. We will discuss these works in the summary section. However, no measurements, nor quantitative analysis in the critical region have been made on these systems.

In the present work, we present the first detailed experiments in the critical region of a 2D random-field Ising system, and show quantitatively the systematics of the destruction of the phase transition. The results are compared to, and are shown to be in agreement with, the theoretical predictions.

II. EXPERIMENTAL TECHNIQUES

The experiments utilized the optical linear magnetic birefringence (Δn) technique. The temperature derivative $d(\Delta n)/dT$ has been shown¹¹ to be proportional to the magnetic specific heat C_m . Experiments have been performed on a variety of materials¹²⁻¹⁴ which confirm this proportionality. Also, a recent study of the pure Rb_2CoF_4 (Ref. 15) has revealed an excellent agreement with the exact Onsager solution of the 2D Ising antiferromagnet. The Sénarmont method was employed with a He-Ne laser

and 50 kHz modulation, with a resolution in Δn of about 10^{-8} . The temperature was measured with a calibrated carbon-glass resistance thermometer using an ac resistance bridge operating at 17 Hz. The field dependence of the thermometer (about 20 mK at T_N at $H=20$ kOe) was measured and corrected for. Measurements of Δn were made by first stabilizing the temperature to within ~ 100 μ K, until thermal equilibrium was reached (approximately 3 min). Then $d(\Delta n)/dT$, which is proportional to C_m , was determined by dividing the difference in Δn between successive points by the temperature difference ΔT . That value of $\Delta n/\Delta T$ was then plotted at the mean T . In some cases the data were smoothed by taking a numerical derivative involving several measured data points.

The sample was an optically excellent single-crystal piece of $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$, approximately 5 mm thick, cut from a large boule grown by the zone-melting method. As in all experiments on these mixed crystals, the primary intrinsic mechanism limiting the sharpness of the observed transition was found to be the gradient of concentration along the growth direction of the crystal (perpendicular to the c axis). This effect was minimized by carefully aligning the narrow laser beam, defined by a pinhole of approximately 100 μm , perpendicular to this direction. This necessitated cutting and polishing the crystal perpendicular to the layers, which fortunately presented no particular problems. In an initial attempt, the laser beam was aligned parallel to the growth direction, with the result that the phase transition was almost completely obliterated. From this observed broadening we estimate the concentration gradient to be 1 mol %/cm.

Although this crystal is of excellent quality, the single most important limitation in this and all other experiments on phase transitions in random materials has been the quality of the crystal used. Experiments which sample a minimum volume (e.g., by aligning a narrow laser beam, or by using a thin slice of material perpendicular to the concentration gradient) are able to produce better data from a given sample than possible otherwise, but any major improvement in the sharpness of the transitions seen will depend upon the availability of still more homogeneous samples.

III. EXPERIMENTAL RESULTS

In Fig. 1 we present the results for $d(\Delta n)/dT$ for various fields, $\vec{H}||c$, and $0 \leq H \leq 20$ kOe. The $H=0$ data exhibit a sharp, symmetric divergence, as is the case in the pure Rb_2CoF_4 .¹⁵ Far above T_N , a broad shoulder appears, which we attribute to increased short-range order not present in the pure material. With increasing H , the peak is seen to dramatically round and decrease in amplitude; only a small shift of the peak temperature is apparent, which is less than the rounding. All of these effects differ markedly from those seen in the 3D random-field case.^{16,17} Outside the crossover region, which increases with H as does the rounding, the data for all fields are indistinguishable from those at $H=0$. This is illustrated in Fig. 2, where the $H=0$ and 20 kOe data are superimposed.

Measurements were also made with $H=20$ kOe perpen-

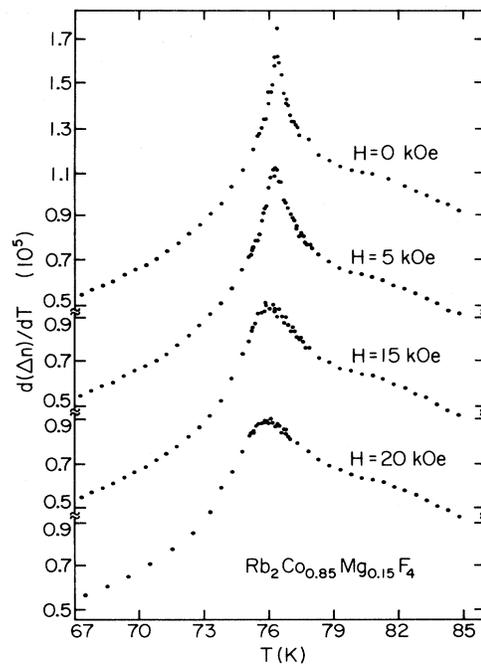


FIG. 1. $d(\Delta n)/dT$ vs T of $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$ in applied fields $H=0, 5, 15, 20$ kOe, with $\vec{H}||c$ axis. Points represent the differences between successive data points Δn , divided by the temperature interval ΔT , and are plotted at the average temperature.

dicular to c . No change in the shape of the transition peak from that at $H=0$ was observed, and ΔT_c is only 0.02 K, consistent with value expected from mean-field theory.¹⁷ That there are no effects attributable to random fields with $\vec{H}||c$ agrees with both theory and other measurements since transverse fields, either uniform or random, do not couple to the antiferromagnetic (AF) order parameter.

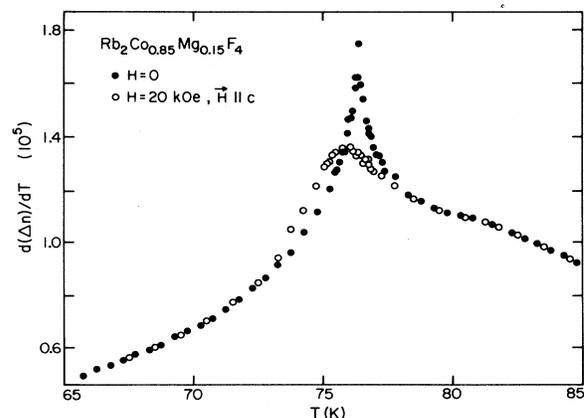


FIG. 2. $d(\Delta n)/dT$ vs T of $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$ in applied fields $H=0$ and 20 kOe. Peak value of $d(\Delta n)/dT$ shifts from $T_N=76.35$ K at $H=0$ to $T_c=75.97$ at $H=20$ kOe. Points were determined as indicated in caption of Fig. 1.

IV. THEORETICAL BACKGROUND

A. Random exchange

The effects of randomness in the exchange interaction on the critical exponents of 3D and 2D Ising systems have been extensively studied.¹⁸⁻²⁷ In the 3D case a crossover from "pure" Ising exponents ($\alpha = +0.11$) to "random" Ising ones ($\alpha = -0.09$) (Ref. 24) is predicted at a reduced temperature $t_c \sim (\Delta J/J)^{1/\alpha}$, where $\Delta J/J$ is a measure of the randomness in the exchange $\Delta J/J \simeq x(1-x)$. This random behavior is expected to be observable only for highly diluted materials, and then only for very small $|t|$. Similar, though less pronounced, changes occur in the other exponents (e.g., γ, ν, \dots). Recent experiments²⁸ have generally confirmed the theoretical predictions in this case.

In the 2D Ising case, where Onsager's exact results exist for the pure system, the effects of randomness in the exchange are much less drastic. Pure Ising behavior, $C_m \propto \ln |t|$, is expected for $t_c \ll |t| \ll 1$, and a new random behavior²⁶ $C_m \propto \ln \ln |t|$ for $|t| \ll t_c \ll 1$, where $t_c \sim \exp[-\text{const}/(1-x)]$. Thus for the modest dilution ($1-x=0.15$) in the present experiment, t_c is expected to be extremely small, and only pure 2D Ising behavior should be observed. In a neutron scattering experiment,²⁹ in $\text{Rb}_2\text{Co}_x\text{Mg}_{1-x}\text{F}_4$, the exponents β, ν, γ , and η were found to be unchanged from those in the pure Rb_2CoF_4 , within experimental error.

B. Random fields

The effects of a random field are predicted to alter both the lower critical dimensionality and critical exponents of Ising systems. Site-random magnetic fields H_i may be defined by their configurational averages, $\langle H_i \rangle = 0$, $\langle H_i^2 \rangle \equiv H_{\text{RF}}^2 \neq 0$. However, not until FA (Ref. 7) showed that an antiferromagnet with random exchange, when placed in a uniform field, was a physical realization of the random-field (RF) problem, was there much experimental interest evidenced. Amongst the results FA obtained was a scaling relation for the specific heat

$$C = |t_H|^{-\alpha} f(h_{\text{RF}}^2 |t_H|^{-\phi}). \quad (1)$$

In the above equation, $t_H = (T - T_N + bH^2)/T_N$ is the reduced temperature relative to the mean-field phase boundary $T_N^{\text{MF}} = T_N - bH^2$, and

$$h_{\text{RF}}^2 = (g\mu_B S/kT)^2 H_{\text{RF}}^2$$

is the reduced mean-square random field. For convenience, we define $h_{\text{RF}} \equiv (h_{\text{RF}}^2)^{1/2}$. The crossover exponent ϕ is predicted³⁰ to be equal to the susceptibility exponent $\gamma = \frac{7}{4}$ for both the pure and random-exchange 2D Ising model. Equation (1) exhibits the leading random-exchange singularity $|t|^{-\alpha}$ when $h_{\text{RF}}^2 = 0$, and describes the crossover, characterized by the exponent ϕ , to random-field behavior when $h_{\text{RF}}^2 \neq 0$. Crossover to new critical behavior is expected to occur for temperatures

$$t_H < h_{\text{RF}}^{2/\phi}. \quad (2)$$

When a sharp transition does occur, $f(x)$ will exhibit a singularity at a new transition temperature given by

$$T_c = T_N - bH^2 - T_N (ch_{\text{RF}}^2)^{1/\phi}, \quad (3)$$

where c is a constant of order unity.

In the case where the thermal eigenvalue (in this case, $\nu^{-1} = 1$) divides the dimensionality (in this case, $d = 2$), the crossover behavior in the presence of an additional relevant field (h_{RF}) is more complicated and contains additional logarithmic terms which cannot simply be obtained from Eq. (1). This case has been treated by Niemeijer and van Leeuwen³¹ with the result that the singular part of the free-energy scales according to

$$F_s(t_H, h_{\text{RF}}) = -\frac{1}{2} A^* t_H^2 \ln h_{\text{RF}} + h_{\text{RF}}^{2d\nu/\phi} \Psi(t_H h_{\text{RF}}^{-2/\phi}), \quad (4)$$

where $\Psi(x)$ is a scaling function. Differentiating twice with respect to t_H gives

$$C_m = g(t_H h_{\text{RF}}^{-2/\phi}) - A^* \ln h_{\text{RF}} + D(t_H, H), \quad (5)$$

where $g(x)$ is a scaling function, and $D(t_H, H)$ represents some nonsingular, but possibly H - and t_H -dependent background.

Since it is expected that $C_m \propto \ln |t|$ when $H = 0$, we must have $g(x) \sim -A \ln |x|$ for $|x| \gg 1$. It follows that as $h_{\text{RF}}^2 \rightarrow 0$,

$$C_m = -A \ln |t| + \frac{2}{\phi} A \ln h_{\text{RF}}^2 - A^* \ln h_{\text{RF}}^2 + D(t, 0). \quad (6)$$

The $\ln h_{\text{RF}}^2$ terms in Eq. (6) diverge as $h_{\text{RF}}^2 \rightarrow 0$, unless

$$A^* = \frac{2}{\phi} A, \quad (7)$$

so that

$$C_m = -A \ln |t| + D(t, 0), \quad (8)$$

when $H = 0$. Despite the fact that no sharp transition occurs when $H > 0$, Eq. (5) still holds, but now $g(x)$ describes the rounding of the transition in the applied field.

It has recently been shown^{17,32} that the mean-field (MF) shift is

$$bH^2 = T_N^{\text{MF}} (g\mu_B S H/x\bar{J})^2 / (1-x\bar{J}/kT_N^{\text{MF}})^2, \quad (9)$$

and the effective random field for the site-dilution problem is

$$h_{\text{RF}}^2 = (\bar{J}/kT)^2 x(1-x)h^2 / (1-x\bar{J}/kT)^2, \quad (10)$$

where $\bar{J} \equiv \sum_r (-1)^r J(r)$, $\tilde{J} \equiv \sum_r J(r)$, and $h \equiv g\mu_B S H/kT$. (In the molecular-field approximation $kT_N^{\text{MF}} = x\bar{J}$ and $k\Theta^{\text{MF}} = x|\tilde{J}|$.) In the first sum, $(-1)^r$ alternates on opposite sublattices.

V. ANALYSIS OF THE DATA

A. Random exchange

In Fig. 3 we show a semilogarithmic plot of the $H = 0$ data versus $|t|$. The data for both $T > T_N$ and $T < T_N$ are seen to fall on a single straight line for $10^{-3} \leq |t| \leq 10^{-2}$, indicating a symmetric, logarithmic divergence, as was seen in the pure 2D Ising case.¹⁵ For

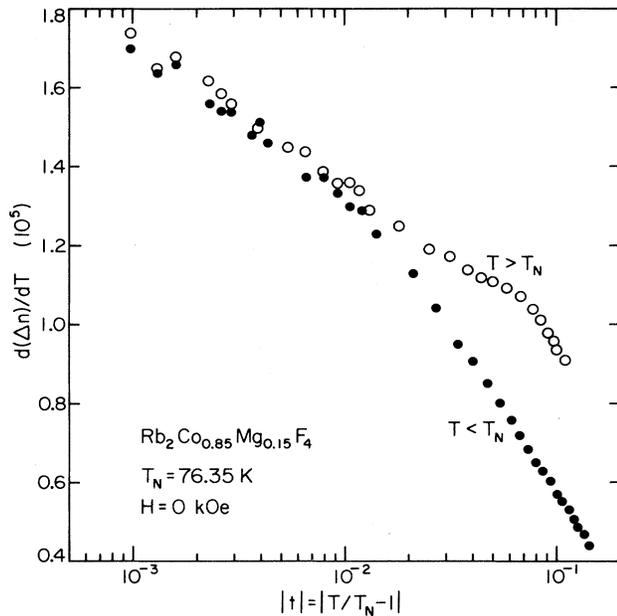


FIG. 3. $d(\Delta n)/dT$ of $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$ vs $\log_{10}H$ of the reduced temperature in $H=0$. Points were determined as indicated in caption of Fig. 1. Data for both $T > T_N$ and $T < T_N$ are seen to fall on a single straight line for $10^{-3} \leq |t| \leq 10^{-2}$, indicating a symmetric, logarithmic divergence, as was seen in the pure 2D Ising case.

$|t| > 10^{-2}$ the data deviate from this behavior, with those for $t < 0$ having a steeper slope than those for $t > 0$. It is expected²⁶ that C_m for the 2D Ising random-exchange system has a logarithmic behavior in the experimentally accessible critical region. Therefore, the data within the range $5 \times 10^{-4} \leq |t| \leq 10^{-2}$ were fitted to the temperature integral of the expression Eq. (8) with $D(t,0) = B + Et$,

$$C_m \propto \frac{d(\Delta n)}{dT} = -\tilde{A} \ln |t| + \tilde{B} + \tilde{E}t, \quad t > 0 \quad (11)$$

TABLE I. Parameters obtained from fitting Δn data for $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$ with $H=0$ to the temperature integral of the equation

$$C_m \propto \frac{d(\Delta n)}{dT} = -\tilde{A} \ln |t| + \tilde{B} + \tilde{E}t, \quad t > 0$$

and the same expression with amplitude A' for $t < 0$. σ is the standard deviation of the fit. \tilde{A}/\tilde{A}' is treated as a variable in the first column and is fixed in the second column.

σ	2.01	2.30
T_N (K)	76.35 ^a	76.35 ^a
\tilde{A}/\tilde{A}'	0.95 ± 0.10	1.00 ^a
\tilde{A}' (K ⁻¹)	(1.24 ± 0.14) × 10 ⁻⁶	(1.23 ± 0.14) × 10 ⁻⁶
\tilde{B} (K ⁻¹)	(7.83 ± 0.13) × 10 ⁻⁶	(7.68 ± 0.14) × 10 ⁻⁶
\tilde{E} (K ⁻¹)	(5.4 ± 1.1) × 10 ⁻⁵	(1.91 ± 0.36) × 10 ⁻⁵

^aFixed.

and to the same expression with amplitude \tilde{A}' for $t < 0$. The results are shown in Table I. Since the ratio of \tilde{A}/\tilde{A}' is close to the theoretical value of 1.0, it was fixed at this value, and the fitting repeated. The quality of the fit was substantially unchanged. The data were also fitted with $\tilde{E}=0$ for the same range of reduced temperature t , both with \tilde{A}/\tilde{A}' fixed to be 1.00 and \tilde{A}/\tilde{A}' variable. For both cases the values obtained for \tilde{A}' and \tilde{B} are within 5% of those shown above. A value of $\tilde{A}/\tilde{A}' = 1.02 \pm 0.01$ was obtained when it was treated as an adjustable parameter. The quality of the fits was slightly worse ($\sigma = 3.10$ and 2.64, respectively) than those found when $\tilde{E} \neq 0$. For all of the fits, T_N was fixed at the value 76.35 K determined from the peak in the $d(\Delta n)/dT$ plot. This could be accomplished with an accuracy of $\sim \pm 20$ mK ($t \sim 2.6 \times 10^{-4}$) since the peak is sharp and symmetric. When fits were attempted with T_N variable, the program would not converge, presumably since $\ln |t|$ is such a slowly-varying function.

B. Random fields

As can be seen from Eq. (5), the amplitude of the peak in C_m (C_{\max}) is expected to decrease with H as

$$C_{\max} \propto \left[\frac{d(\Delta n)}{dT} \right]_{\max} = \tilde{g}_{\max} - \tilde{A}^* \ln_{\text{RF}} + \tilde{D}(t_H, H), \quad (12)$$

where \tilde{g}_{\max} is the value of $\tilde{g}(x)$ at its peak. Since the data for all values of H are indistinguishable outside the crossover region, we will assume $D(t_H, H)$ to be H independent and $D(t_H, H) = D(t, 0) = B + Et$ as in Eq. (11).

Thus Eq. (12) can be written as

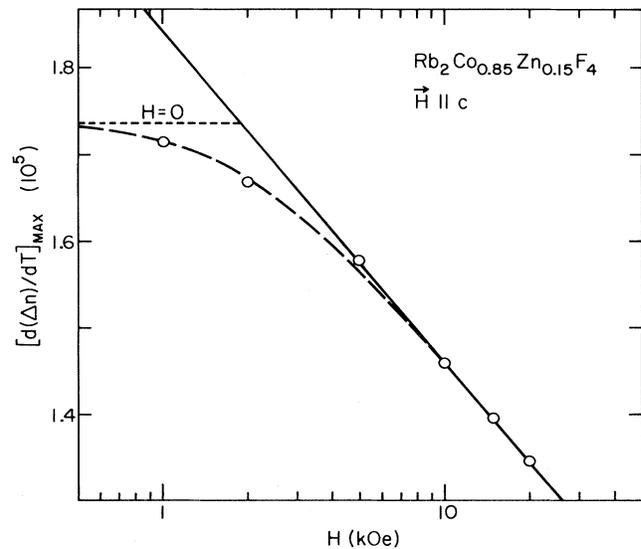


FIG. 4. $[d(\Delta n)/dT]_{\max}$ vs $\log_{10}H$ of $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$ in applied fields $H=0, 1, 2, 5, 10, 15,$ and 20 kOe. Points represent the amplitude of the peak in $d(\Delta n)/dT$ vs T . Data for $H > 2$ kOe are seen to fall on a straight line with slope $-\tilde{A}^*/\log_{10}e$ [Eq. (13)]. Long dashes represent the effective rounding field $H^* = (H^2 + 1.9^2)^{1/2}$ kOe.

$$C_{\max} \propto \left[\frac{d(\Delta n)}{dT} \right]_{\max} = -\tilde{A}^* \ln h_{\text{RF}} + \tilde{B}^*, \quad (13)$$

where $\tilde{B}^* = \tilde{g}_{\max} + \tilde{B}$. In Fig. 4 the peak amplitude $[d(\Delta n)/dT]_{\max}$ for all H is plotted versus $\log_{10} H$. For $H > 2$ kOe the points fall on a straight line; its slope A^* was found by fitting the data for $H > 2$ kOe to Eq. (13), with the result $\tilde{A}^* = (1.56 \pm 0.13) \times 10^{-6}$, $\tilde{B}^* = (6.37 \pm 0.49) 10^{-6}$, or $\tilde{g}_{\max} = (-1.31 \pm 0.51) \times 10^{-6}$. The use of this value of \tilde{A}^* and $\tilde{A} = (1.23 \pm 0.14) \times 10^{-6}$ from Table I allows us to check Eq. (7). We find $\tilde{A}/\tilde{A}^* = 0.79 \pm 0.11$, which is to be compared with $\phi/2 = 0.875$ from Eq. (7) with $\phi = \gamma$. This agreement is quite satisfactory, and encourages us to further study Eq. (5).

For $H < 2$ kOe, $[d(\Delta n)/dT]_{\max}$ approaches a maximum value corresponding to the $H = 0$ peak height. Projecting this value on to the high-field $\log_{10} H$ behavior we find the $H = 0$ peak height is limited by some other mechanism (almost certainly the concentration gradient) to a value equivalent to that which would be produced by a field $H \sim 1.9$ kOe. The peak height at any H can be described by an effective rounding field H^* through the empirical relation $H^* = (H^2 + 1.9^2)^{1/2}$ kOe, as is shown by the dashed line in Fig. 4.

The coefficient c of the random-field shift in T_c that is contained in Eq. (3) is predicted to be of order unity. It was determined using the experimental results, Eqs. (3), (9), and (10) as follows: From Fig. 2 it was found that the peak value of $d(\Delta n)/dT$ shifts from $T_N = 76.35$ K at $H = 0$ to $T_c = 75.97$ at $H = 20$ kOe; corrections were made for the mean-field shift bH^2 [Eq. (9)], which is 38 mK at $H = 20$ kOe. The reduced field h at $H = 20$ kOe and $T = T_N$ is found to be $h = 0.0618$ using $g_{\parallel} = 7.02$ and $S = \frac{1}{2}$.³³ The mean-square random field h_{RF}^2 , using Eq. (10), $kT_N/\bar{J} = 1.11$, and $T_N/\Theta = 0.56$ for Rb_2CoF_4 ,³³ is $h_{\text{RF}}^2 = 0.0233h^2$ at $T = T_N$. From Eq. (3) we obtain

$$c = [(T_N - T_c - bH^2)/T_N]^\phi / h_{\text{RF}}^2, \quad (14)$$

and using $\phi = \gamma = 1.75$, a value of $c = 0.9_{-0.4}^{+0.5}$ is found (see Table II).

TABLE II. (a) Parameters (h , h_{RF} , and ΔT^{MF}) from Eqs. (9) and (10) and the "rounding" temperature t^* , from Eq. (17), at $H = 2$ and 20 kOe. (b) Parameters (\tilde{A}^* , \tilde{B}^* , and \tilde{g}_{\max}) determined by fitting $[d(\Delta n)/dT]_{\max}$ to Eq. (13), ϕ from Eq. (7), c from Eq. (14), and c^* from Eq. (16).

	2 kOe	20 kOe
(a)		
h	0.0062	0.062
h_{RF}	0.00095	0.0095
$\Delta T^{\text{MF}} = bH^2$	0.00038	0.038
t^*	0.0009	0.013
(b)		
$\tilde{A}^* = (1.56 \pm 0.13) \times 10^{-6} (\text{K}^{-1})$		$\phi = 1.58 \pm 0.22$
$\tilde{B}^* = (6.37 \pm 0.49) \times 10^{-6} (\text{K}^{-1})$		$c = 0.9_{-0.4}^{+0.5}$
$\tilde{g}_{\max} = (-1.31 \pm 0.51) \times 10^{-6} (\text{K}^{-1})$		$c^* = 5.4_{-1.8}^{+2.6}$

Similarly, a coefficient c^* of the random-field rounding can be defined in the following way. Using Eq. (7), we can rewrite Eq. (13) as

$$\left[\frac{d(\Delta n)}{dT} \right]_{\max} = -\tilde{A} \ln(c^* h_{\text{RF}}^2)^{1/\phi} + (\tilde{A}^*/\phi) \ln c^* + \tilde{B}^*, \quad (15)$$

and define c^* by $(\tilde{A}^*/2) \ln c^* + \tilde{B}^* = \tilde{B}$, or

$$-(A^*/2) \ln c^* = \tilde{g}_{\max}. \quad (16)$$

Noting that the right-hand sides of Eqs. (11) and (15) are of the same form, we can describe the field dependence of the peak height by an effective "rounding temperature" t^* , defined to be

$$t^* = (c^* h_{\text{RF}}^2)^{1/\phi}. \quad (17)$$

Using the values of $\tilde{g}_{\max} = (-1.31 \pm 0.51) \times 10^{-6}$ and $\tilde{A}^* = (1.56 \pm 0.13) \times 10^{-6}$ in Eq. (16), we obtain $c^* = 5.4_{-1.8}^{+2.6}$. In order to check this result, the temperature t^* , for which rounding is expected, was calculated from Eq. (17) for several magnetic field strengths. The results $t^* = 0.013$ at 20 kOe and $t^* = 0.0009$ at 2 kOe agree quite well with the onset of rounding in the two cases. Likewise, the rounding t^* due to $H^* = 1.9$ kOe is found to be $t^* = 0.0009$, in good agreement with the rounding in $H = 0$ (Fig. 3).

With the use of the renormalization-group scaling relation it is evident that it should be possible to collapse the peaks for all $H \neq 0$ on to a single curve, the scaling function $g(x)$ in Eq. (5). This prediction was tested in Fig. 5 by plotting \tilde{G}_m vs $t_H h^{-2/\phi}$ for all $H > 0$ where, in analogy with $g(x)$ in Eq. (5),

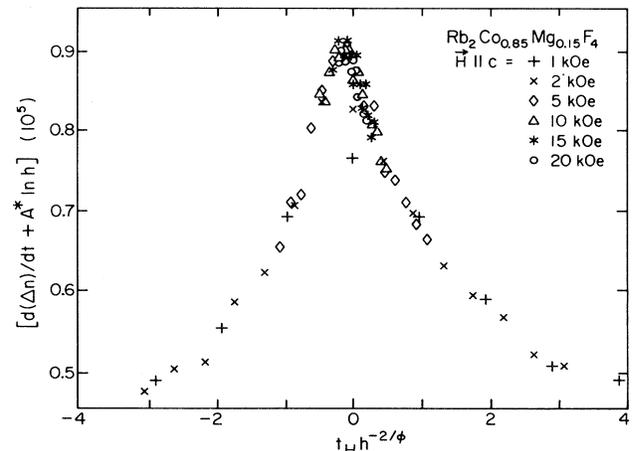


FIG. 5. $d(\Delta n)/dT + \tilde{A}^* \ln h$ vs $t_H h^{-2/\phi}$ of $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$ in applied fields $H = 1, 2, 5, 10, 15,$ and 20 kOe, and $10^{-3} \leq |t| \leq 10^{-2}$. $\tilde{A}^* = 1.64 \times 10^{-6}$, $\phi = 1.75$, and $h = g\mu_B SH/kT$. Data for all H collapse on a single curve: the scaling function $g(x)$ from Eq. (5). A few points near the peaks of the Δn data for $H \leq 2$ kOe do not collapse on the others because the rounding at low fields probably results from concentration gradients.

$$\tilde{G}_m \equiv \frac{d(\Delta n)}{dT} + \tilde{A}^* \ln h. \quad (18)$$

Only data within the range $10^{-3} \leq |t| \leq 10^{-2}$ were plotted, which is the range for which the $H=0$ data behave logarithmically. The value of $\tilde{A}^* = 1.56 \times 10^{-6}$ was initially tried, which is the one obtained from the fit of C_{\max} to Eq. (13). The data collapsed better, however, with the choice of $\tilde{A}^* = 1.64 \times 10^{-6}$, which is within the error in the above determination of \tilde{A}^* , and is the value used in Fig. 5.

Plots of \tilde{G}_m vs $t_H h^{-2/\phi}$ were then made with different values of ϕ ($1.5 \leq \phi \leq 2.0$) and the data were found to collapse best for $\phi = 1.75 \pm 0.2$, confirming the prediction that $\phi = \gamma$. Examining Fig. 5 we see that a few points near the peaks of the $H \leq 2$ kOe data do not collapse on to the others. This is not a surprising result, since $[d(\Delta n)/dT]_{\max}$ for these fields did not fit the $\log_{10} H$ behavior in Fig. 4, for reasons already explained.

VI. SUMMARY AND DISCUSSION

The results obtained in this birefringence study of random exchange ($H=0$) and random-field ($H \neq 0$) 2D Ising critical behavior are to be compared with those previously found^{17,28} for 3D Ising systems. In the latter, a change in sign of the specific-heat exponent α was found in going from the pure ($\alpha = +0.11$) to random-exchange ($\alpha = -0.09$) 3D Ising case.²⁸ In the present 2D Ising study both pure and random-exchange exponents are the same ($\alpha = 0$). For both the 2D and 3D cases random exchange causes *no* rounding of the transition.

The difference between 2D and 3D Ising systems is more pronounced when the effects of random fields on the critical behavior are considered. In the 3D case a crossover was observed from random exchange to new random-field critical behavior, with a value for $\tilde{\alpha} \simeq 0$, characteristic of a lower effective dimensionality; namely, $\tilde{d} \simeq 2$. Of particular significance is that the transition remains sharp to the extent that no rounding is observed for values of $h \leq 0.13$ ($H \leq 20$ kOe) for $|t| \geq 10^{-3}$. T_c was observed to shift with field as $h_{\text{RF}}^{2/\phi}$, in agreement with the predicted scaling relation with $\phi = \gamma$, within experimental error, as predicted. The amplitude of the shift was found to be in good quantitative agreement with that predicted for the site-dilution case. By way of contrast, the present experiments reveal an almost immediate rounding of the transition which occurs for $h \leq 0.003$ ($H \leq 1$ kOe). Despite the absence of a sharp transition, our study reveals that crossover from 2D random exchange to a rounded transition does occur with a crossover exponent ϕ approximately equal to the predicted value; namely, that of the 2D susceptibility exponent $\gamma = \frac{7}{4}$.

It is found that the broadening, peak height, and shift can be described by the scaling prediction, and that, as in the 3D case, the amplitudes of these quantities are in good quantitative agreement with predictions for the site-dilution case. Indeed, the data for all $H > 0$ has been successfully collapsed on a single curve, the scaling function.

Thus it is clear that small random fields ($h_{\text{RF}} \ll 1$) do not destroy the sharp transition in 3D Ising systems for $x \gg x_p$, the percolation concentration, and the predicted crossover to new critical behavior characteristic of a lower effective dimensionality occurs, whereas in the 2D Ising case the transition immediately rounds in the presence of very small random fields.

In another study of the transition region in a random 2D Ising antiferromagnet in an applied field,¹⁰ the uniform parallel susceptibility χ was measured for different concentrations ($x = 1.0, 0.8, 0.7$, and 0.6) on the same system as was investigated in the present experiments, $\text{Rb}_2\text{Co}_x\text{Mg}_{1-x}\text{F}_2$. No attempt was made to determine critical behavior, but rather qualitative judgments were made about the rounding of the transition from the slope of the χ -vs- T plots. In low fields and $x \leq 0.7$, a peak in $\chi(T)$ was observed, which is seen to decrease in amplitude and broaden with increasing H , possibly accompanied by a small shift. This is in contrast with the pure antiferromagnet ($x = 1$) where $d(\chi T)/dT$ diverges with the specific heat exponent. FA have predicted a peak in χT in $h = 0$, expected to be smeared away when $h \neq 0$, which might explain what is seen, in part. However, to properly judge whether the transition is rounded and the manner in which crossover affects the changes that are observed in χ with increasing H , one should compare the measured results with the appropriate scaling function, namely, d^2F/dH^2 . Since this has not been done, it is difficult to compare the χ results with those we have obtained.

Neutron studies^{8,9} of the shape and width of the magnetic Bragg peaks have been made on $\text{Rb}_2\text{Co}_{0.7}\text{Mg}_{0.3}\text{F}_4$. From these measurements the field and temperature dependence of the inverse correlation length κ have been obtained. The results indicate that the random fields destroy long-range order (κ increases with increasing H_0). On the face of it, this would seemingly indicate complete agreement with our study and the measurement of χ as to what transpires in a 2D Ising system in random fields. However, a field dependence to κ was also found in the neutron studies of some 3D Ising systems ($\text{Fe}_{0.35}\text{Zn}_{0.65}\text{F}_2$ and $\text{Co}_{0.35}\text{Zn}_{0.65}\text{F}_2$) but not others ($\text{Mn}_{0.65}\text{Zn}_{0.35}\text{F}_2$), whereas our previous birefringence studies^{16,17} of the $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$ and $\text{Fe}_x\text{Zn}_{1-x}\text{F}_2$ indicate no destruction of the sharp phase transition occurs in random fields. The resolution of this dilemma is not yet at hand.

Although our experiments are in accord with some of the theoretical predictions, it is clear that further study of the random-field problem is needed. To that end we intend to explore the H and x dependence of the birefringence in $\text{Rb}_2\text{Co}_x\text{Mg}_{1-x}\text{F}_4$ values of $x_p < x < 1.0$. It would also be extremely valuable to make detailed comparison of the correlation length with the birefringence in the critical region, to understand the role domains play in the destruction (rounding) of the phase transition.

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- ¹Y. Imry and S.-k. Ma, *Phys. Rev. Lett.* **35**, 1399 (1975).
- ²E. Pytte, Y. Imry, and D. Mukamel, *Phys. Rev. Lett.* **46**, 1173 (1981).
- ³K. Binder, Y. Imry, and E. Pytte, *Phys. Rev. B* **24**, 6736 (1981).
- ⁴G. Grinstein and S.-k. Ma, *Phys. Rev. Lett.* **49**, 684 (1982).
- ⁵J. Villain, *J. Phys. (Paris)* **43**, 6551 (1982).
- ⁶G. Grinstein and S.-k. Ma (unpublished).
- ⁷S. Fishman and A. Aharony, *J. Phys. C* **12**, L729 (1979).
- ⁸H. Yoshizawa, R. A. Cowley, G. Shirane, R. J. Birgeneau, H. J. Guggenheim, and H. Ikeda, *Phys. Rev. Lett.* **48**, 438 (1982).
- ⁹R. A. Cowley, R. J. Birgeneau, G. Shirane, and H. Yoshizawa, *Phys. Rev. B* **28**, 2588 (1983); R. J. Birgeneau, H. Yoshizawa, R. A. Cowley, G. Shirane, and H. Ikeda, *ibid.* **28**, 1438 (1983).
- ¹⁰H. Ikeda, *J. Phys. C* **16**, L21 (1983).
- ¹¹G. A. Gehring, *J. Phys. C* **10**, 531 (1977).
- ¹²I. R. Jahn, *Phys. Status Solidi* **57**, 681 (1973).
- ¹³D. P. Belanger, P. Nordblad, A. R. King, V. Jaccarino, L. Lundgrin, and O. Beckman, *J. Magn. Magn. Mater.* **31-34**, 1095 (1983).
- ¹⁴K. Iio, M. Sakatani, and K. Nagata, *J. Phys. Soc. Jpn.* **45**, 1567 (1978).
- ¹⁵P. Nordblad, D. P. Belanger, A. R. King, V. Jaccarino, and H. Ikeda, *Phys. Rev. B* **28**, 278 (1983).
- ¹⁶D. P. Belanger, A. R. King, and V. Jaccarino, *Phys. Rev. Lett.* **48**, 1050 (1982).
- ¹⁷D. P. Belanger, A. R. King, V. Jaccarino, and J. L. Cardy, *Phys. Rev. B* **28**, 2522 (1983).
- ¹⁸A. B. Harris, *J. Phys. C* **7**, 1671 (1974).
- ¹⁹A. B. Harris and T. C. Lubensky, *Phys. Rev. Lett.* **33**, 1540 (1974).
- ²⁰T. C. Lubensky, *Phys. Rev. B* **11**, 3580 (1975).
- ²¹G. Grinstein and A. Luther, *Phys. Rev. B* **13**, 1329 (1976).
- ²²D. E. Khmel'nitskii, *Zh. Eksp. Teor. Fiz.* **68**, 1960 (1975) [*Sov. Phys.—JETP* **41**, 981 (1976)].
- ²³C. Jayaprakash and H. J. Katz, *Phys. Rev. B* **16**, 3987 (1977).
- ²⁴K. E. Newman and E. K. Riedel, *Phys. Rev. B* **25**, 264 (1982).
- ²⁵G. Jug, *Phys. Rev. B* **27**, 609 (1983).
- ²⁶V. S. Dotsenko and V. S. Dotsenko, *J. Phys. C* **15**, 495 (1982).
- ²⁷G. Jug, *Phys. Rev. B* **27**, 4518 (1983).
- ²⁸R. J. Birgeneau, R. A. Cowley, G. Shirane, H. Yoshizawa, D. P. Belanger, A. R. King, and V. Jaccarino, *Phys. Rev. B* **27**, 6747 (1983).
- ²⁹H. Ikeda, M. Suzuki, and M. Hutchings, *J. Phys. Soc. Jpn.* **46**, 1153 (1979).
- ³⁰A. Aharony, *Phys. Rev. B* **18**, 3318 (1978).
- ³¹T. Niemeijer and J. M. J. van Leeuwen, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. 6, p. 446.
- ³²J. L. Cardy (unpublished).
- ³³D. J. Breed, K. Gilijamse, and A. R. Miedema, *Physica (Utrecht)* **45**, 205 (1969).