Random fields and three-dimensional Ising models: $Co_xZn_{1-x}F_2$

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The phase transition and ordering of the $d=3$ diluted Ising antiferromagnet $Co_xZn_{1-x}F_2$ has been studied using two-axis neutron diffraction for the cases (i) where x is close to the percolation threshold, in zero applied magnetic field, and (ii) as a function of applied magnetic field for samples with $x=0.26$ and 0.35. The results of the percolation study show complicated behavior, probably due to concentration gradient problems. Nevertheless, there is strong evidence that the inverse correlation length decreases to zero at the onset of long-range order. The results of the magnetic field study are compared with the theoretical predictions for the $d=3$ Ising model in a random field. It is found that when the samples are cooled in even the smallest (nonzero) fields the long-range magnetic order is destroyed and that the structure factor is well described by the Lorentzian plus Lorentzian squared form. The inverse correlation length is found to have a power-law dependence on the applied magnetic field at low temperature with exponents $\nu_H=2.17\pm0.3$ for the $x=0.26$ sample and $\nu_H=3.63\pm0.3$ for the $x=0.35$ sample. This result is not consistent with the current theoretical predictions for the field dependence of the inverse correlation length in the $d=3$ Ising model in a random field. The measurements also show that the system is frozen at low temperatures and this freezing may be responsible for the discrepancy between theory and experiment.

I. INTRODUCTION

The effect of a random field on the ordering of Ising models has been studied theoretically in several recent papers. Imry and Ma[1] showed by comparing the random-field energy to the energy needed to produce a smooth domain wall that an Ising model is unstable against the breakup into domains for all dimensionalities less than $d_c=2$. In contrast, there have been ϵ-expansion[2,3] and supersymmetry arguments[4] which suggest that in the presence of random fields the behavior of a system is similar to that of the pure system in two less dimensions. Since $d_c=1$ for the Ising model this would suggest that $d_c=3$ in the presence of a random field.

It was the existence of these two results that led us to begin an experimental study of the effect of a random field on a three-dimensional Ising model. Fishman and Aharony[5] first pointed out that a uniform field applied to a random antiferromagnet produced a randomly directed staggered field. They initially considered a random bond antiferromagnet when the random field is proportional to the ferromagnetic susceptibility and the applied uniform field. In practice, site random antiferromagnets are more readily available, and then the random staggered field has two components: a part proportional to the ferromagnetic susceptibility as discussed by Fishman and Aharony; and another part, which is probably dominant in practice due to the randomness in the dipole moment from site to site. The strength of this latter term is directly proportional to the applied field and is independent of temperature. Our experiments were performed on the antiferromagnet CoF_2 diluted with the nonmagnetic material ZnF_2. CoF_2 is an antiferromagnet with exchange interactions between nearest and next-nearest neighbors; due to the crystal-field effects the exchange is very anisotropic.[6] CoF_2 and ZnF_2 form mixed crystals in which there is no tendency toward short-range chemical order of the Co and Zn. Both the excitations[7] and the phase transitions[8] of this system have been measured, so that it is quite well characterized.

When this work was begun we intended to study the phase transitions and the onset of long-range order close to the percolation threshold. This was intended to complement earlier work[9-11] on the percolation problem, by providing more reliable measurements of the thermal exponents for a three-dimensional Ising system and further information about the ordering of three-dimensional systems. In three-dimensional systems with close to Heisenberg interactions,[11] the inverse correlation length κ is not zero at the onset of long-range order and in a $KMn_xZn_{1-x}F_3$ crystal the long-range order was found

to decrease below 6.0 K. Experiments on a three-dimensional Ising system would clarify whether these unexpected features result from unexpected aspects of three dimensions or are present only in the systems with nearly Heisenberg-type interactions.

Our experiments were performed using neutron scattering techniques at the Brookhaven National Laboratory High Flux Beam Reactor and are described in detail in Sec. II. The results of the percolation experiments are described only briefly in the Appendix because they were not wholly successful. The effect of applying a uniform field to these samples is described in Sec. III, and these experiments yielded many interesting new results which are analyzed in Sec. IV; some of these have already been briefly described.[12]

Since we first began this experimental work there have been several theoretical papers on the effect of random fields on Ising models. Pytte *et al.*[13] have extended the work of Wallace and Zia[14] on capillary waves to systems with a random field. The model proposed by Pytte *et al.* included in the Hamiltonian the lowest-order analytic term introduced by the random field and then used the replica technique to perform the averaging over the disorder. In this model Pytte *et al.* found that, as a result of the critical dimension for the roughening transition being shifted from $d_R=3$ to 5 by the random field, the lower critical dimension for the model was 3 and not 2. A physical interpretation of this result, given by Binder *et al.*,[15] was that the shift in d_R meant that in $d=3$ the width of a domain wall increased at the same rate as the domain size and that the interface must be considered as rough on all length scales. In a recent paper, however, Grinstein and Ma[16] have argued that the random field introduces a non-analytic term into the Hamiltonian, rather than an analytic one. Their renormalization-group calculations with this Hamiltonian show that, although d_R shifts from 3 to 5, the variation of the domain wall width in $d=3$ is not as fast as the domain size, and as a consequence for large length scales the domain walls are effectively smooth. They then argue that $d_c=2$ as given by Imry and Ma. Similar results have been obtained by Villian.[17]

An alternative approach was used by Kogen and Wallace.[18] They extended the supersymmetry argument of Parisi and Sourlas[4] and showed within this framework that capillary waves destroyed the long-range order for $d<d_c=3$. In a recent paper Cardy[19] has argued that this is an exact result for $T=0$ and similar arguments have been given by Niemi.[19]

Clearly, in view of this theoretical controversy and activity, experiments must be performed to test these theories. As we shall describe in detail in the conclusion, a cursory glance at our experimental results suggests that all these theories are incorrect, but a more circumspect analysis suggests caution. There is no doubt that although aspects of these theories are correct, much work needs to be done before they will provide a complete description of our results.

Specifically, the theories described above are all for a ferromagnet with uniform interactions in the presence of a random field. Our experiments, on the other hand, are for an antiferromagnet *with vacancies* in a uniform applied field. It is believed that near six dimensions[16] these problems are equivalent. However, it is not at all obvious that the vacancies do not become relevant in $d=3$; certainly domain walls will preferentially be located near the vacancies and this could affect the scaling of the domain-wall width with length thus altering d_c.

II. EXPERIMENTAL TECHNIQUES

Crystals of $Co_xZn_{1-x}F_2$ were grown from very pure CoF_2 and ZnF_2 by using the Czochralski method of growth. The samples had a volume of several cm^3 and consisted of several large single-crystal grains. These crystals were cleaved to obtain single crystals with a volume of about 1 cm^3. Several of these crystals with x nominally 0.25 and hence close to the percolation concentration were studied in detail as a function of temperature in the hope of elucidating the percolation properties of this three-dimensional Ising system. The results are reported in the Appendix.

One of these crystals and another with a nominal concentration of $x=0.35$ were selected for studying in an applied magnetic field. It is unfortunately very difficult to determine accurately the concentrations, x, in these samples. Approximate values of x for these two samples can be obtained from the measured Néel temperatures of 6.70 ± 0.05 and 13.25 ± 0.01 K, respectively. As a rough estimate, if we assume that the Néel temperature is proportional to the number of spins in the infinitely connected network, and that the fraction of spins in the infinitely connected network is taken from the work of Kirkpatrick,[20] then the concentrations x are 0.26 and 0.35, respectively. The former concentration is very close to the percolation concentration of 0.24 for a bcc lattice.[21]

The neutron-diffraction measurements were performed at the Brookhaven National Laboratory High Flux Beam Reactor using a two-axis spectrometer. An incident neutron wave vector of 2.67 $Å^{-1}$ was obtained by reflection from the (002) planes of a pyrolitic graphite monochromator. In order to suppress neutrons reflected from higher-order planes two pyrolitic graphite filters were used. In the percolation experiments the samples were mounted with the [010] axis vertical in a variable temperature cryostat. The collimation used was 20′ before the monochromator, 20′ between the monochromator and sample, and 20′ between the sample and detector. The resolution function was then measured at the (100) reflection to be 0.015 $Å^{-1}$ full width at half maximum (FWHM) parallel to the wave-vector transfer and 0.005 $Å^{-1}$ perpendicular to the wave-vector transfer.

In order to perform the random-field experiments the samples were aligned with the [001] axis vertical and placed in the variable temperature insert of a superconducting magnet which produced a vertical magnetic field of up to 7.5 T. The measurements on the $x=0.26$ sample were performed with the same collimations as the zero-field experiments. However, for the measurements on the $x=0.35$ sample the 20′ collimators were replaced by 10′ collimators throughout. The resolution function in the scattering plane was then measured as 0.009 $Å^{-1}$

(FWHM) parallel to the wave-vector transfer and 0.0024 Å^{-1} perpendicular to the wave-vector transfer.

III. EXPERIMENTAL RESULTS

As discussed in the Introduction a uniform field applied to a random antiferromagnet produces a randomly directed staggered field. We have therefore performed experiments by applying a uniform field to the $Co_xZn_{1-x}F_2$ system. In the sample for which $x=0.26$ x is close to the percolation point, and the applied field is relatively large so that the results are undoubtedly strongly influenced by the proximity of the percolation threshold. Consequently, we also performed experiments on a more concentrated system with $x=0.35$.

One of the difficulties of working with Ising systems at low temperatures is the problem of ensuring that the relaxation times for the establishment of thermodynamic equilibrium are always shorter than the time of the experiment. Because of this problem the experiments were mostly performed by changing the magnetic field while the sample was at a temperature above T_N, and then cooling the sample while keeping the magnetic field fixed. Although there is no guarantee that this procedure gives the low-temperature ground state, it is accepted as giving the best approximation to the ground state in spin-glasses. We measured the scattering in the neighborhood of the (1,0,0) lattice point for several different magnetic fields as shown in Fig. 1 for $x=0.26$ and in Fig. 2 for $x=0.35$. In the former case the peak intensity at the (1,0,0) lattice point monotonically decreased with increasing field as also shown in more detail in Fig. 3. This decrease is similar to that observed for the scattering in the absence of a field as a function of temperature (see Fig. 16, sample *A*) suggesting that there might be a phase transition at about 1.0 T. The behavior is, however, quite different as can be seen

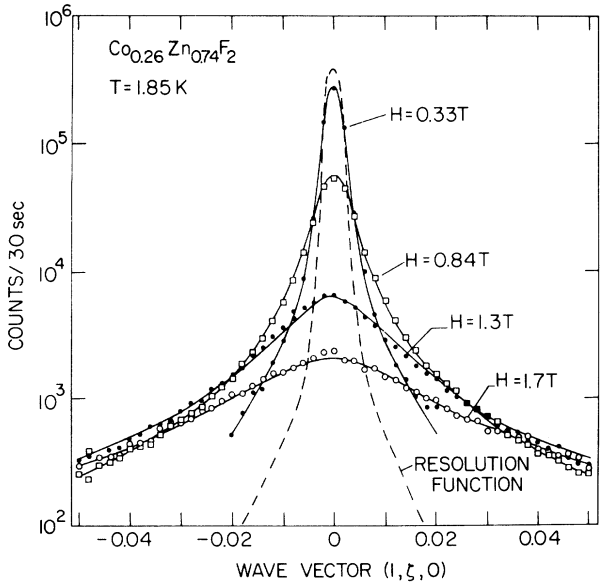

FIG. 1. Scattering for wave-vector transfers along [1ζ0] at 1.85 K from $Co_{0.26}Zn_{0.74}F_2$. The lines are fits to the sum of a Lorentzian and a Lorentzian squared as detailed in the text.

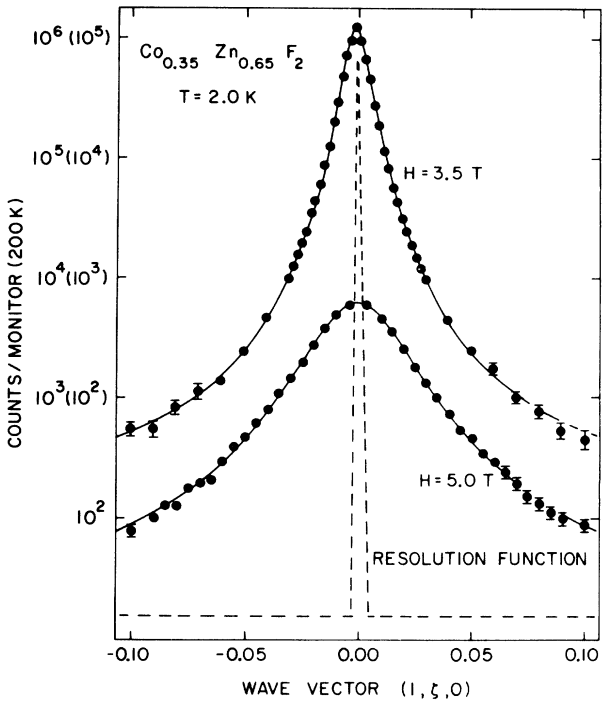

FIG. 2. Scattering for wave-vector transfers along [1ζ0] at 2.0 K from $Co_{0.35}Zn_{0.65}F_2$. The solid lines are fits to the sum of a Lorentzian and a Lorentzian squared as described in the text.

from inspection of Figs. 1 and 2. The width of the scattering as a function of the wave vector is steadily increasing with increasing magnetic field, unlike the case of decreasing temperature. Since the width of the (2,0,0) nuclear Bragg peak is unchanged on applying the field, this increase in the width means that the long-range antiferromagnetic order has been destroyed for magnetic fields much less than 1.0 T. The behavior of the width in the sample with $x=0.35$ is very similar, as shown in Fig. 2, although the increase in the width is much smaller for a given magnetic field strength than in the more dilute sample. The peak intensity in the $x=0.35$ sample initially increases as a function of magnetic field; the peak intensity is a maximum for a field of about 1.5 T, and at larger

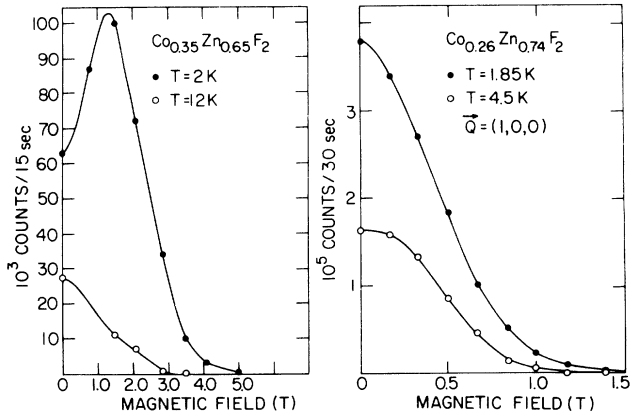

FIG. 3. Peak intensity of the scattering with $\vec{Q}=(1,0,0)$ in $Co_xZn_{1-x}F_2$ as a function of magnetic field.

fields the peak intensity falls uniformly as a function of magnetic field in a similar way to that for $x=0.26$. We believe that this "anomalous" increase for small fields can be explained by considering the effect of extinction. This crystal, even though it is site disordered, has a sufficiently small mosaic spread so that in the zero field the intensity of the (1,0,0) reflection is *extinction limited.* When the sample is cooled in an applied field the magnetic order within the mosaic blocks is broken up into domains by the random-field effect, and as a consequence the Darwin angle of the magnetic domains within the mosaic block will be increased allowing a greater proportion of the incident beam to be scattered. The effect of the random field on the peak intensity is thus manifested in two ways; one reduces the scattering power of a magnetic domain by the destruction of long-range order while the other increases the proportion of the beam that each mosaic block is able to scatter. The initial effect of a random field might therefore increase the magnetic peak intensity by lifting the extinction present in the (1,0,0) reflection. This behavior makes it very difficult to make precise statements about the small field measurements in this sample. It should be emphasized that this behavior occurs only for the magnetic scattering and that the nuclear Bragg peaks are unaffected.

The behavior of the scattering as a function of temperature at fixed field is illustrated in Fig. 4 for $x=0.26$ and in Fig. 5 for $x=0.35$. At low temperatures the width of the scattering is almost independent of temperature but then increases with increasing temperature, while the intensity generally decreases with increasing temperature.

At the beginning of this section we discussed the difficulty of knowing whether the system is in the thermodynamic equilibrium state at low temperatures. In both samples the state of the system is frozen at low temperatures below about 4.0 K but in the case of $x=0.26$ only for fields below 1.2 T. This is illustrated in Fig. 6 where we compare the scattering observed when the system is cooled in zero field, with that resulting from cooling in a

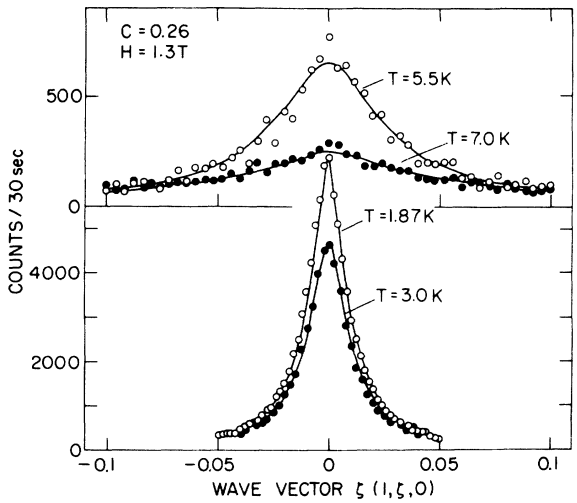

FIG. 4. Scattering for wave-vector transfers $\vec{Q}=(1,\zeta,0)$ with $H=1.3$ T at various temperatures for $Co_{0.26}Zn_{0.74}F_2$. The solid lines are Lorentzian fits to the measurements.

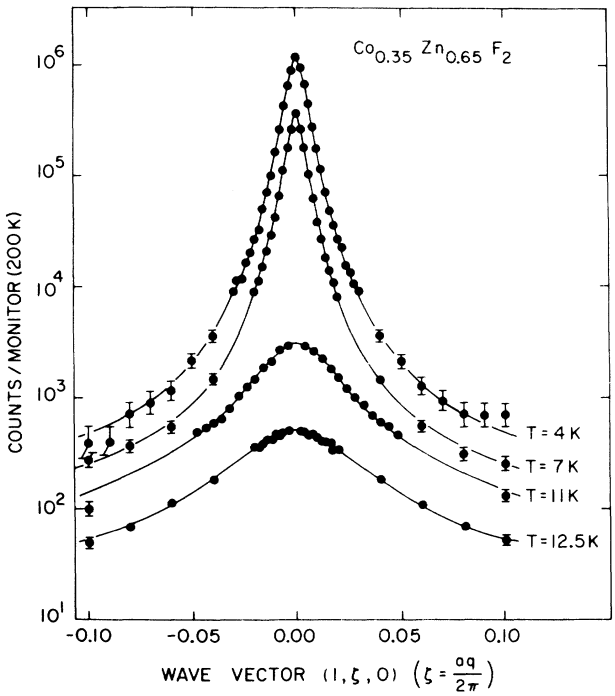

FIG. 5. Scattering for wave-vector transfers $\vec{Q}=(1,\zeta,0)$ with $H=3.5$ T at various temperatures for $Co_{0.35}Zn_{0.65}F_2$. The solid lines are Lorentzian plus Lorentzian-squared fits to the measurements for temperatures below 11 K and Lorentzian fits for 11 K and 12.5 K.

field of 2.5 T and then reducing the field to zero. In the latter case the scattering is fairly similar to that observed when the sample is cooled in a field of 0.8 T. We do not understand the origin of the small shift in peak position in Fig. 6, especially as the nuclear Bragg reflections were not changed. Clearly one expects strong magnetoelastic effects accompanying the domain wall formation and such effects presumably account for the distortions evidenced in Fig. 6. This result demonstrates that at low temperatures and fields the scattering is dependent on the immediate prior history of the sample. Similar results were obtained even at the largest fields used in the $x=0.35$ sample. We cannot therefore be certain that the results obtained by cooling in a field, shown in Figs. 1–5 and discussed in the rest of this paper, are characteristic of the thermodynamic equilibrium state at low temperatures. They are characteristic of the state obtained by cooling in a field and the scattering is then constant over periods of several days.

IV. ANALYSIS OF THE RESULTS

The scattering, shown in Figs. 1 and 2, was initially analyzed by fitting it to a Lorentzian profile, which has proved to be so successful at describing the scattering observed in many other circumstances. The intensity was fitted to the form

$$I(\vec{Q})=|f(\vec{Q})|^2\frac{A}{\kappa^2+(\vec{q}^{\,*})^2}\ , \tag{1}$$

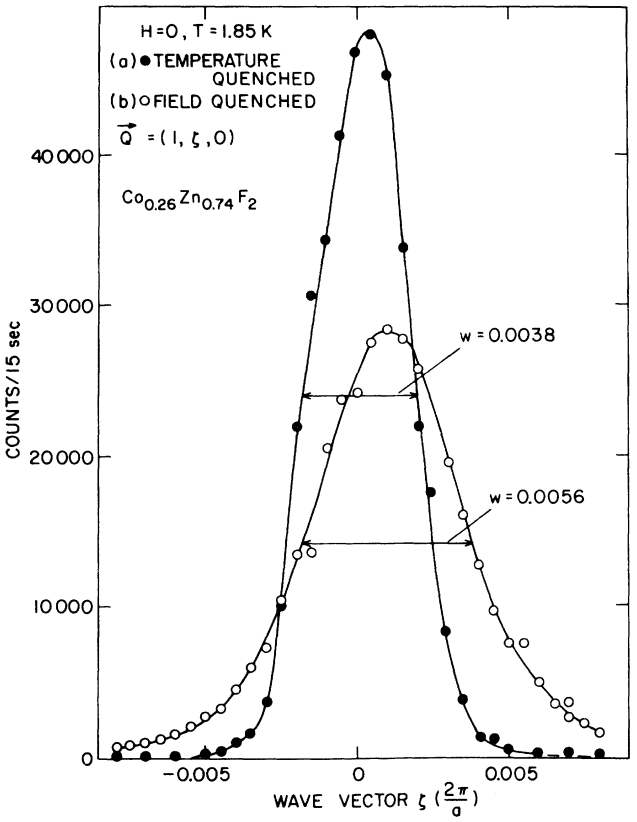

FIG. 6. Scattering observed for wave vectors along the direction [1ζ0] at 1.85 K and $H=0$ obtained by cooling the sample when $H=0$ and in a field of 2.5 T and then reducing the field. The difference in the results show that the spins are frozen at low temperatures and fields.

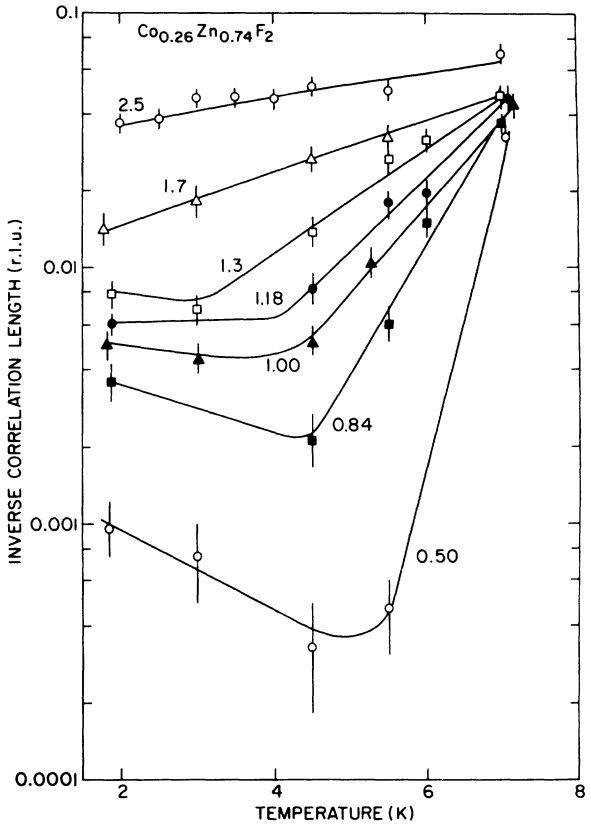

FIG. 7. Inverse correlation length κ (reciprocal-lattice units) deduced for $Co_{0.26}Zn_{0.74}F_2$ as a function of temperature for various applied fields. The solid lines are guides to the eye.

where $f(\vec{Q})$ is the form factor of the Co^{2+} ion[22] and $\vec{q}^*$ is the difference between $\vec{Q}$ and the reciprocal-lattice vector (1,0,0) expressed in reciprocal-lattice units. The analysis was performed by convoluting Eq. (1) with the measured experimental resolution and fitting the parameters A and κ to the experimental results. Equation (1) gave a good description of the experimental results for temperatures above T_N and in the $x=0.26$ sample for fields above 1.2 T. At lower temperatures and fields, the results could not be described by Eq. (1). We attempted to fit these results by the Lorentzian form [Eq. (1)], and an additional central Gaussian to represent an increased mosaic spread for the magnetic structure. This combination of functions failed to describe many of these results, because the scattering falls off more slowly than a Gaussian for small q^*, but more rapidly than a Lorentzian in the wings.

Several different functional forms have been tried to describe the results for low temperatures. Before the recent theoretical advances described in the Introduction, fits to the $x=0.26$ data were made assuming that

$$I(\vec{Q})=|f(\vec{Q})|^2\frac{A}{|\vec{q}^*|^{2-\eta}}\ . \tag{2}$$

This form gives a good description[12] of the $x=0.26$ experimental results when convoluted with the experimental resolution function. The exponent η is close to zero for

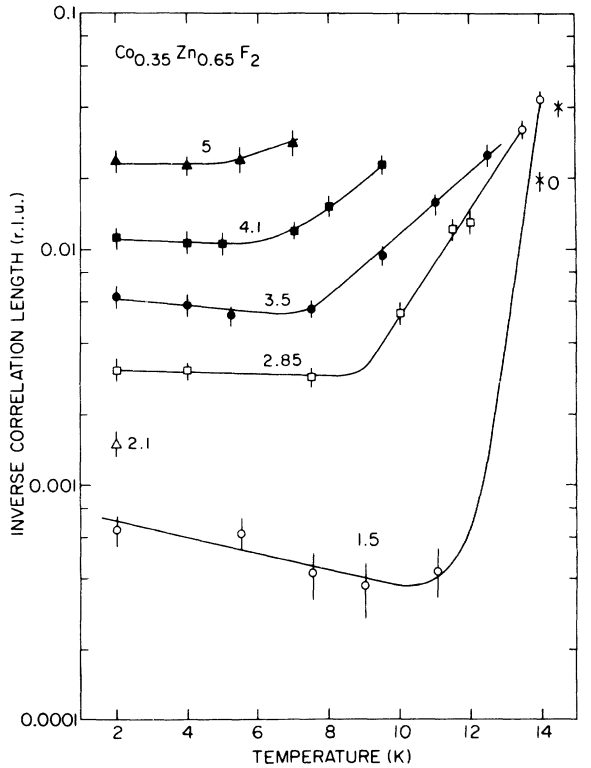

FIG. 8. Inverse correlation length κ deduced for $Co_{0.35}Zn_{0.65}F_2$ as a function of temperature for various applied fields.

$H=1.2$ T and at a temperature of 2.0 K and then steadily decreases with decreasing field and approaches -1 when $H=0$. The exponent is given approximately by $\eta=-1+(H/H_c)^2$ when $H_c\approx 1.1$ T, and the amplitude A depends at least approximately on the applied field such as H^{-2}.

Although this form gives a reasonably accurate description of the experimental results, recent theoretical work[13,18] suggests that a more appropriate form is

$$I(\vec{Q})=|f(\vec{Q})|^2\left[\frac{A}{\kappa^2+|\vec{q}^{\,*}|^2}+\frac{B}{(\kappa^2+|\vec{q}^{\,*}|^2)^2}\right]. \tag{3}$$

As discussed in the paper[23] on $Rb_2Co_xMg_{1-x}F_4$, the Lorentzian-squared term continuously evolves into a Bragg peak as $H\to 0$, provided that $B\sim\langle S_z\rangle^2\kappa^{4-d}$, where d is the dimensionality of the system. We find that Eq. (3) does give a very satisfactory description of the results, so that the results can be described either by Eqs. (2) or (3). A similar conclusion was found[23] in the two-dimensional system $Rb_2Co_xMg_{1-x}F_4$. However, since current theory strongly favors the Lorentzian plus Lorentzian-squared form, most of our efforts have concentrated on fitting the parameters in Eq. (3), A, B, and κ, to the experimental results.

As shown in Figs. 1, 2, 4, and 5, Eq. (3) gives a good description of our experimental results. The results for the temperature dependence of the inverse correlation length κ for various different fields are shown in Fig. 7 for $x=0.26$ and in Fig. 8 for $x=0.35$. In both cases, on cooling from high temperatures κ sharply decreases with decreasing temperature and is then constant or even slightly increases on further cooling.

As noted previously, the random staggered field contains both a direct Zeeman contribution due to the randomness in the moment and an induced contribution due to the randomness in the interaction. The former, which is dominant, is temperature independent whereas the latter peaks near T_N. These two fields both are generated by the dilution and further they *oppose* each other locally. The diminution of the bond-randomness term with decreasing temperature below T_N may account for the observed decrease in the correlation length as $T\to 0$.

In Fig. 9 we show the low-temperature behavior of B/κ and A as a function of field for the two samples. For $x=0.35$ B/κ is nearly constant except at the lowest fields where the effect of the extinction is to reduce its value. For $x=0.26$ B/κ decreases to zero at about 1.2 T. The amplitude of the Lorentzian A is almost constant for fields above 1.2 T in the $x=0.26$ sample and decreases as the field is reduced. In the $x=0.35$ sample the amplitude A increases with increasing field in a similar way to the $x=0.26$ sample, but does not become constant even at the highest fields. These results are, we believe, characteristic of both high- and low-field behavior respectively. At low fields $A\to 0$ and the scattering is almost purely Lorentzian squared with B/κ a constant. The scattering then develops smoothly into a Bragg peak as $\kappa\to 0$. At relatively large fields the amplitude of the Lorentzian squared decreases and the system becomes a normal paramagnet.

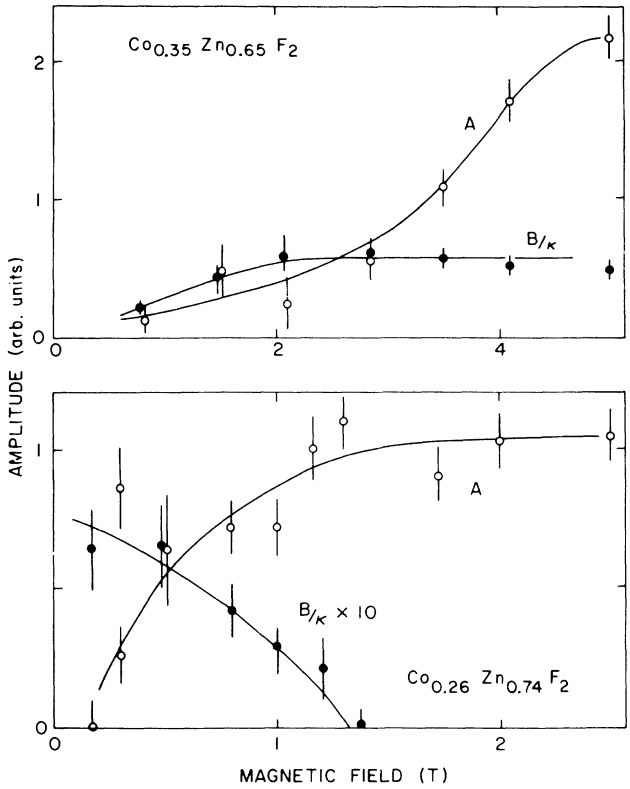

FIG. 9. Amplitude of the Lorentzian A and Lorentzian squared B divided by κ deduced for $Co_{0.26}Zn_{0.74}F_2$ at 1.85 K and $Co_{0.35}Zn_{0.65}F_2$ at 2.0 K.

The behavior of B/κ is very similar to that of the square of an order parameter. In Fig. 10 we show its behavior as a function of temperature for various fields and the data collapses, at least approximately, to a single curve which is very similar to that of the Bragg peak intensity (see Fig. 16, sample A). The behavior is more complex in the $x=0.26$ sample because the amplitude of the Lorentzian-squared term is nonzero only within a circle described approximately by

$$\left[\frac{T}{T_c}\right]^2+\left[\frac{H}{H_c}\right]^2=R^2=1\ ,$$

where $T_c=6.7$ K and $H_c=1.2$ T. In Fig. 11 we have therefore shown B/κ plotted against R. Although the errors are considerable the behavior is not inconsistent with that of the order parameter (see Fig. 16, sample A). This behavior is thus consistent with our heuristic prediction that $B\sim\langle S_z\rangle^2\kappa$ in three dimensions.

As described in the Introduction there is considerable interest in the behavior of the three-dimensional Ising model in small random fields and at low temperatures. Our results show that the scattering is broader than a Bragg peak and evolves smoothly into a Bragg peak as the field is reduced. Although we cannot determine κ with any accuracy at the smallest fields, there is a marked increase in the intensity of the wings of the Bragg peak for fields as small as 0.17 T for $x=0.26$ and 0.8 T for

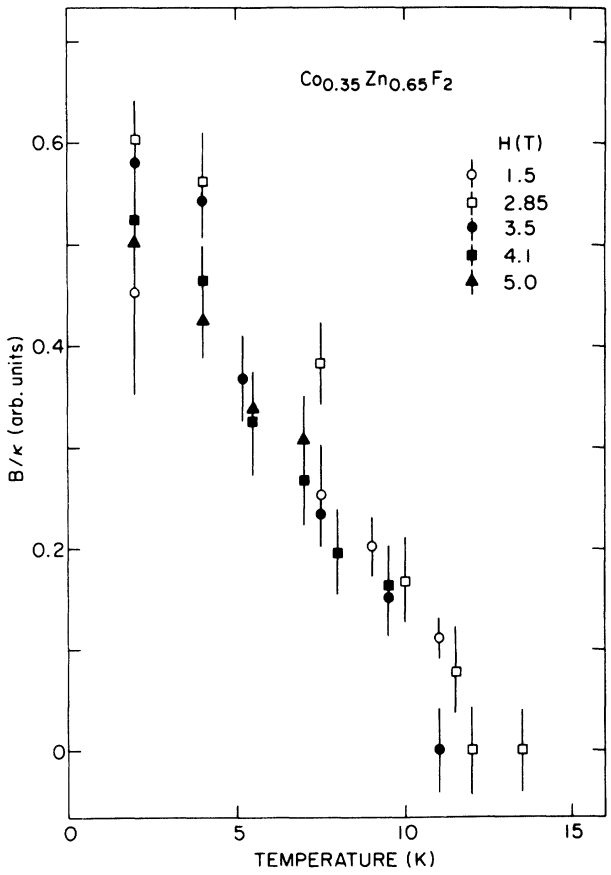

FIG. 10. Amplitude of the Lorentzian squared plotted against temperature for $Co_{0.35}Zn_{0.65}F_2$ for various fields.

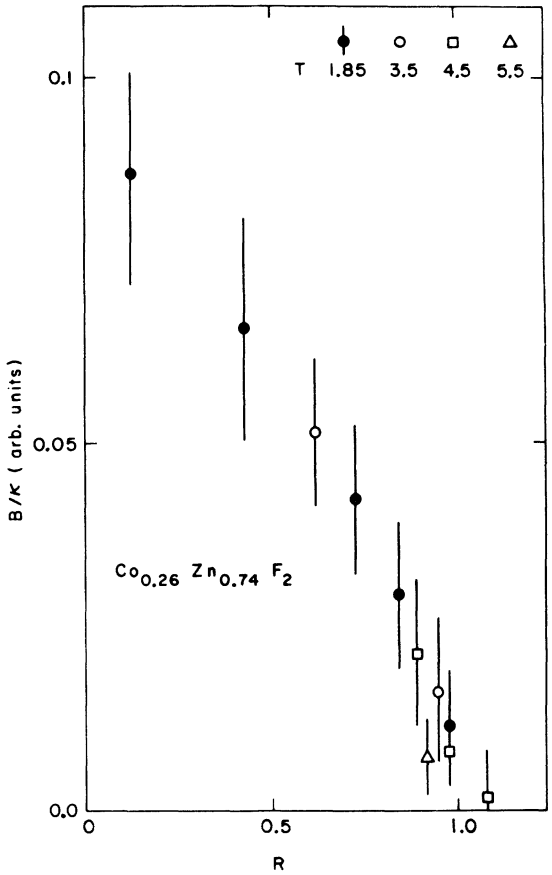

FIG. 11. Amplitude of the Lorentzian squared B/κ plotted against $R=[(T/T_c)^2+(H/H_c)^2]^{1/2}$ for $Co_{0.26}Zn_{0.74}F_2$.

$x=0.35$. These fields correspond to temperatures $g\mu_B H/k_B$ of 0.48 and 2.3 K, respectively, which are much smaller than the transition temperatures of 6.7 and 13.7 K and of the energy needed to turn one Co^{2+} ion in the presence of only one of its antiferromagnet neighbors: 11.1 K.[6,7] These results show that cooling the samples in these fields which are considerably smaller than the other fields in the system results in the crystals being in a magnetic state with no long-range order at low temperatures. If $d_c=3$ then it would be expected[13,18] that at low temperatures $\kappa \propto \exp[-(H_0/H)^2]$, where H_0 is some constant. In Fig. 12 we show $\ln\kappa$ plotted against $1/H^2$ for the $x=0.35$ sample, and the results clearly do not give a straight line. We also test a power-law description in Fig. 13 and this gives a much better description of the results apart from the result at lowest fields which may be influenced by the extinction problem discussed above. A least-squares fit omitting this point gives

$$\kappa=\kappa_0 H^{\nu_H}$$

with $\nu_H=3.63\pm0.12$, $\kappa_0=0.000\,069\pm0.000\,012$, and χ^2 is 2.62, where κ is in reciprocal-lattice units and the magnetic fields in tesla. A similar analysis has been performed on the results for the $x=0.25$ sample and gives $\nu_H=2.17\pm0.16$ while $\kappa_0=0.0047\pm0.0004$, while χ^2 is 1.12; the fit is shown in Fig. 14. It is not too surprising

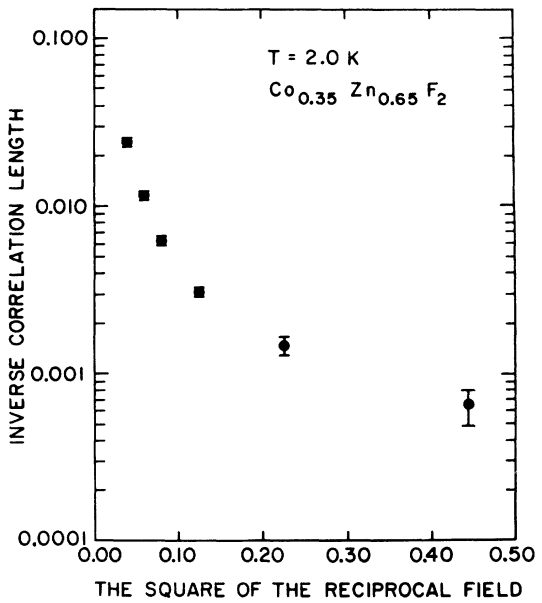

FIG. 12. Logarithm of κ at 2.0 K plotted against $1/H^2$. A straight line is to be expected if the lower critical dimension is 3.

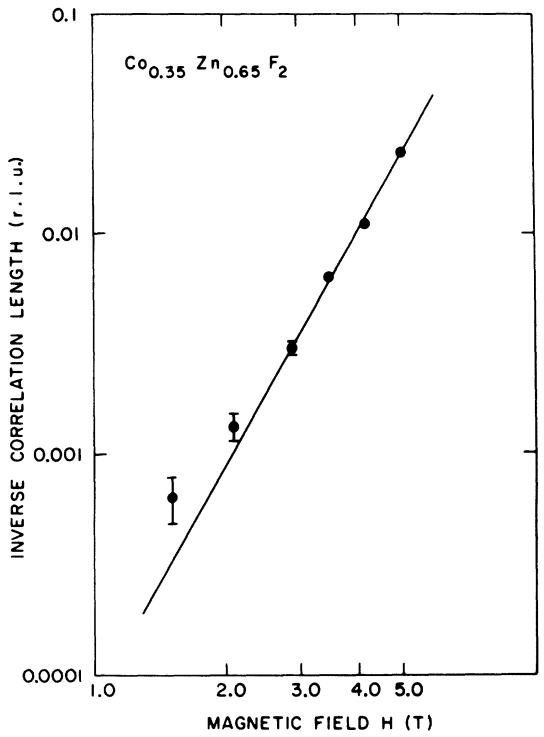

FIG. 13. κ at 2.0 K plotted against the applied field for $Co_{0.35}Zn_{0.65}F_2$. The straight line is a fit which gives the exponent.

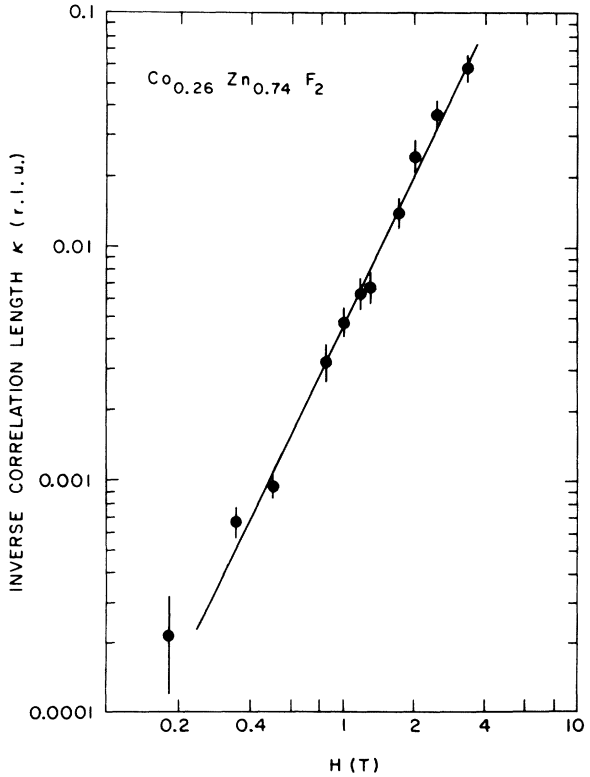

FIG. 14. κ at 1.85 K for $Co_{0.26}Zn_{0.74}F_2$ plotted against H. The straight line is a fit to the results.

that the results for ν_H for the two concentrations are different because all the results are in the Lorentzian-squared quasiordered phase for the $x=0.35$ sample whereas for $x=0.26$ the fit was performed over the whole range of H. It is more likely therefore that $\nu_H=3.63$ is the appropriate "random-field exponent" for this system. There is also a very large difference in the values of κ_0 by nearly 2 orders of magnitude. Such a large change cannot be accounted for by statistical factors such as x or $1-x$. The effect of the random field must increase very rapidly as the percolation point is approached. As discussed by Fähnle,[24] this probably reflects the highly ramified and hence quasi-one-dimensional nature of the infinite network near percolation.

Finally in Fig. 15 we show the low-temperature structure factor $I(1,0,0)$; that is, the intensity for $\vec{Q}=(1,0,0)$ corrected for the finite experimental resolution as a function of magnetic field for both systems. The results for $x=0.35$ are approximately linear apart from the point at the lowest fields. When this is omitted a least-squares fit to the form

$$I=I_0H^{-\gamma_H}$$

gives $\gamma_H=10.9\pm0.6$. In the case of $x=0.26$ the three points at the largest fields clearly are not part of the same straight line and omitting these points gives $\gamma_H=8.0\pm0.6$.

V. DISCUSSION AND CONCLUSIONS

The main conclusion of the experiment is that the application of a magnetic field whose energy is much smaller than the exchange energy or kT_N has a drastic effect upon

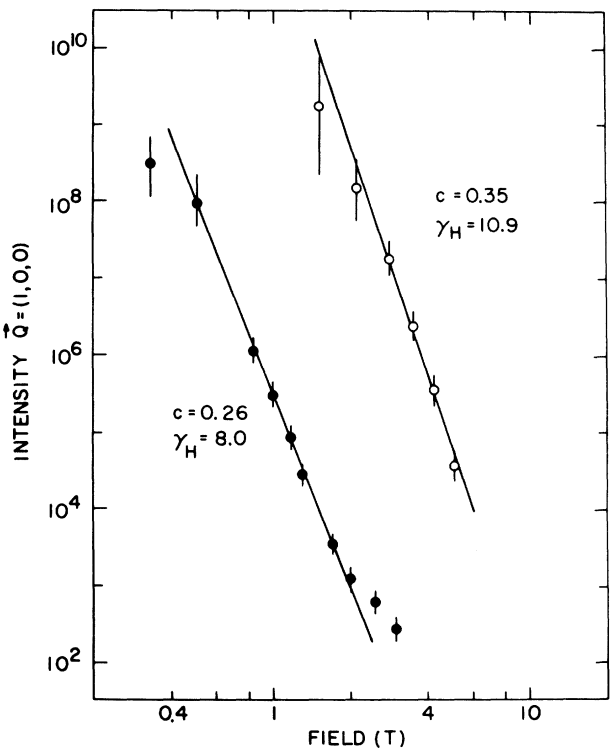

FIG. 15. Scattering for wave vector $\vec{Q}=(1,0,0)$ corrected for the resolution as a function of the magnetic field.

the phase transition and ordering of $Co_xZn_{1-x}F_2$. When the system is cooled in a uniform magnetic field, the state reached at low temperatures is not one of long-range antiferromagnetic order. The scattering at low fields is dominated by a Lorentzian squared with a width which increases with increasing field. This Lorentzian-squared term represents, we believe, a quasiordered random-field state. Firstly, this is because the intensity of the scattering at low fields is the same as that of the Bragg reflection describing the long-range order in the absence of a magnetic field. This result holds not only at low temperatures but at all temperatures below T_N. Secondly, at large fields, 1.2 T when $x=0.26$, the intensity of the Lorentzian-squared term decreases and the system becomes a normal paramagnet. This critical field corresponds to a single-site field energy of only about $\frac{1}{2}T_N$ or 0.31 of the nearest-neighbor exchange energy.

The detailed behavior of the inverse correlation length as a function of field is undoubtedly complex. In the $x=0.35$ sample $\kappa \propto H^{\nu_H}$ with $\nu_H=3.63\pm0.12$, at low temperatures. A different power 2.17 ± 0.16 was obtained for the $x=0.26$ sample, but this latter result is dominated by the results in the high-field paramagnetic region, $H>1.2$ T, and the results at low fields were of insufficient accuracy to determine the limiting low-field behavior.

These results appear to be inconsistent with current theories of the $d=3$ Ising model in a random field. Those theories for which $d_c=2$ predict long-range order at low temperatures while the theories which give $d_c=3$ suggest that $\kappa \sim \exp[-(H_0/H)^2]$. Our results, if simply interpreted, would suggest that $d_c>3$.

There may be several reasons for the discrepancy other than errors in the theories or the experiments. As mentioned in the Introduction, it may be that a uniform field applied to a random antiferromagnet does not give the same behavior as a random field applied to a uniform ferromagnet. The physics of random-field systems has been found to be unexpectedly subtle and the effect of the random exchange interactions and, specifically, vacancies which provide a soft path for the domain walls may be more severe than currently thought.

Another difficulty may arise because of the difficulty of establishing the thermodynamic ground state at low temperatures. We know that this system is frozen at 2.0 K and so, if long-range order is the equilibrium state only for temperatures somewhat below the freezing temperature, then this state could not be sampled in the experiment. The theories might then be correct but, so far as experimental work is concerned, they are irrelevant to the behavior in real experiments.

Birefringence measurements[25] show a sharp peak at low fields for these three-dimensional antiferromagnets. Indeed the peak in $d(\Delta n)/dT$, which is proportional to the heat capacity, appears to be sharper in a field than for $H=0$, reflecting a crossover in the heat-capacity exponent α from -0.09 to ~ 0.0. In addition, the temperature of the peak as a function of field follows the theoretically predicted changes in T_N ($\Delta T_N \sim H^{2/\gamma}$) quite well. In $Fe_xZn_{1-x}F_2$ a comparison of the neutron scattering results with the birefringence results shows that the peak occurs at a temperature just above that at which κ reaches its minimum value. Our results indicate that the peak is not associated with the development of true long-range antiferromagnetic order. Current theories describe the breakup of long-range order in a random field as due to the presence of domains walls. Most likely these walls are sufficiently far apart in the region probed by the birefringence measurements that the ordering within the antiferromagnetic domains gives a peak in the heat capacity with negligible rounding. The change in α suggests that new critical behavior is being observed but, presumably, this critical behavior is cut off by entry into the domain-wall state. Extension of the birefringence measurements to higher fields, especially in more dilute samples, would test this latter idea.

Clearly these experiments show that random fields drastically change the properties of systems close to phase transitions and prevent the establishment of long-range order. The random fields may have an energy which is very much less than kT_N and yet produce a very large effect. Since impurities in crystals can frequently produce random fields, their effects may be more important than hitherto considered.

Finally, these experiments present a challenge to theory. The theory should be extended to include the effect of temperature when our experiments show unambiguously that large effects occur close to T_N for the $d=3$ Ising model. The theory should then be extended to consider the establishment of the thermodynamic equilibrium state at lower temperatures. Random fields produce large and dramatic effects on the ordering and phase transitions of $d=3$ Ising models and we hope this paper will lead to further experimental and theoretical work on this difficult problem.

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APPENDIX: PERCOLATION IN $Co_xZn_{1-x}F_2$

1. Experimental results

The neutron scattering from the magnetic fluctuations in $Co_xZn_{1-x}F_2$ was studied for six samples with x nominally equal to 0.25. In two of these samples the magnetic scattering observed for wave vectors close to the (1,0,0) lattice point was weak and only slowly varying with the

wave vector even at 1.2 K. This presumably meant that the concentrations of these samples were so much less than the percolation threshold that all the magnetic spins were in relatively small independent clusters.

In each of the other four samples the results were qualitatively similar although different in detail and so we describe the results for only two of the samples labeled *A* and *B*. In each of these samples there was intense magnetic scattering at the (1,0,0) reciprocal-lattice point at low temperatures, the width of which was determined by the experimental resolution. This scattering is indicative of long-range order at low temperatures and its temperature dependence is shown in Fig. 16. The intensity of the scattering is closely proportional to $T_N - T$ for each sample except that close to T_N, where there is evidence of rounding, presumably due to concentration fluctuations. The onset of long-range order occurred at a similar temperature between 5.5 and 7.0 K in each sample.

The diffuse scattering observed for a wave-vector transfer $\vec{Q}=(1,0,-0.008)$ is shown in Fig. 17 for samples *A* and *B*. It increases rapidly as the phase transition is approached and has a maximum at a temperature somewhat below the onset of long-range order. On further cooling the amplitude of the critical scattering decreases slightly for sample *A* but at temperatures below 2.5 K increases again. This increase in the critical scattering intensity at low temperatures is unexpected. It also occurs in the $x=0.35$ sample in the applied magnetic field. This suggests that the low-temperature behavior for sample *A* may arise from random fields possibly arising from impurities or from dipolar interactions.

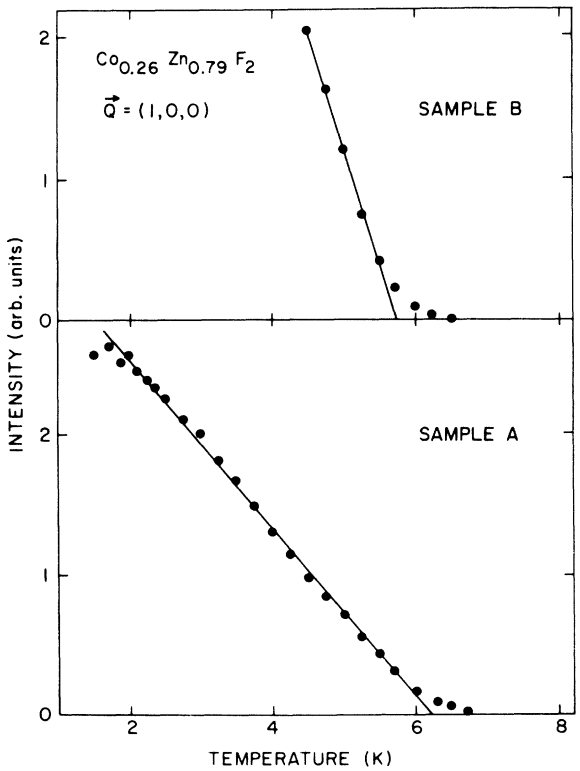

FIG. 16. Integrated Bragg intensity of the (100) magnetic lattice point for two samples of $Co_xZn_{1-x}F_2$ with $x \approx 0.26$.

The diffuse scattering was measured in detail using scans with wave vectors along the direction $[\zeta 00]$ and $[00\eta]$ through the (1,0,0) lattice point. The diffuse scattering was then analyzed assuming that it could be described by a Lorentzian profile, Eq. (1). The results are displayed in Figs. 18 and 19. The inverse correlation length decreased as the temperature was reduced from 10 K and would appear to be heading towards zero at T_N. Somewhat above T_N the inverse correlation length ceases to decrease and is then almost independent of temperature until possibly at low temperatures, below 2 K, when it decreases again. The amplitude *A* decreases with decreasing temperature.

2. Discussion of results

As mentioned in the Introduction the initial objective of these experiments was to perform a detailed study of the percolation in a three-dimensional Ising system similar to that performed for other systems. Unfortunately, the crystal-growth problems made a detailed study impossible. The results described above and shown in Figs. 16–19 are in some respects unexpected but can probably be understood if there is a considerable spread in T_N in different parts of the crystal. Indeed, in an Ising system it is expected that the transition temperature increases very rapidly with concentration close to the percolation point, because the temperature is determined by the one-

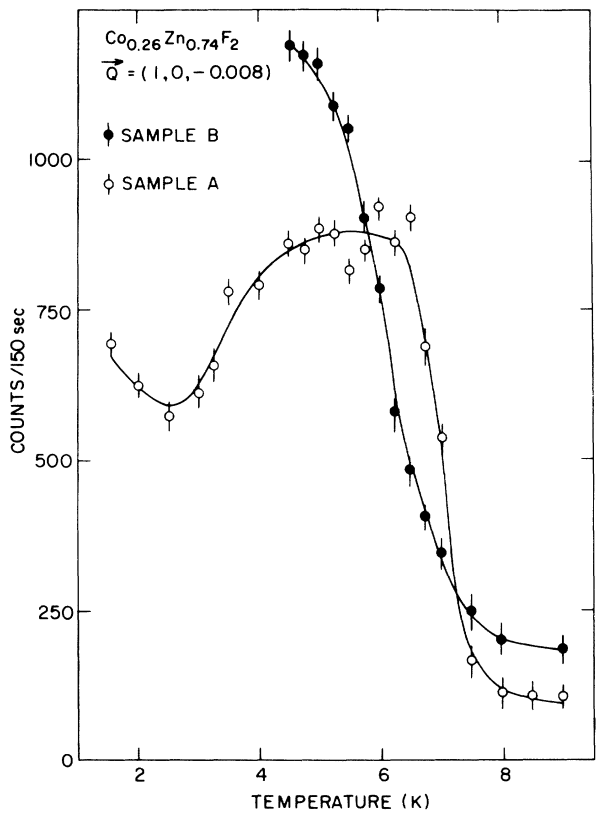

FIG. 17. Scattering observed for a wave-vector transfer of $\vec{Q}=(1,0,-0.008)$ from two samples of $Co_xZn_{1-x}F_2$. The wave-vector transfer was chosen to be close to the (1,0,0) reciprocal-lattice point but not so close that the scattering was contaminated by the Bragg reflection.

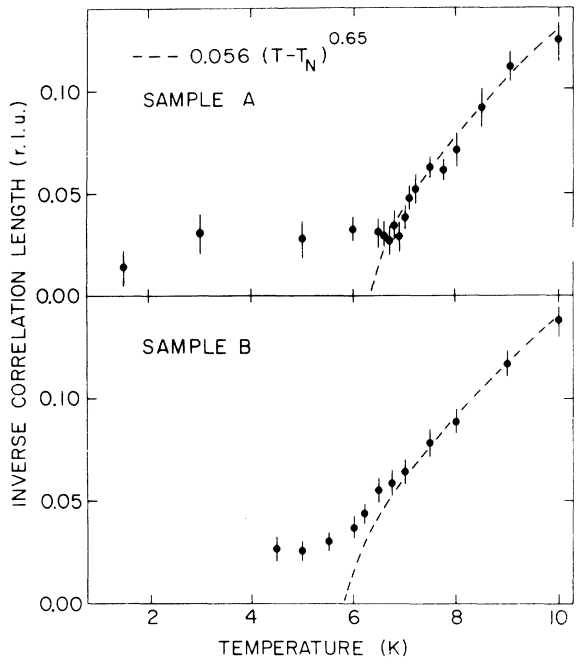

FIG. 18. Inverse correlation length κ as a function of temperature for two samples of $Co_xZn_{1-x}F_2$ with $x \approx 0.26$.

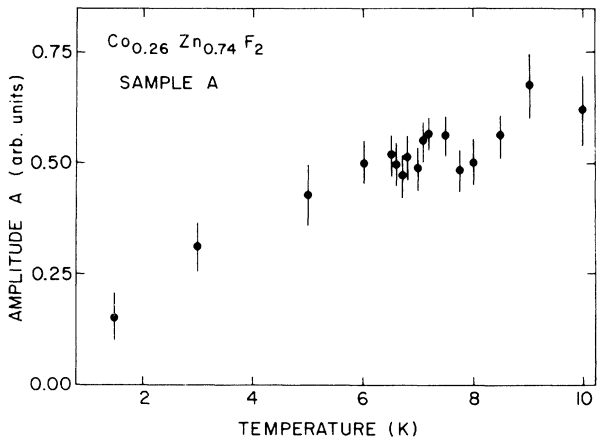

FIG. 19. Amplitude A of the Lorentzian critical scattering for $Co_xZn_{1-x}F_2$ with $x \approx 0.26$.

dimensional weak links[26] which order as $\exp(-J/k_BT)$. Specifically then $T_N \sim \ln(c - c_p)$ so that a small spread in concentration will produce a very broad distribution of Néel temperatures; it is interesting to note that because of the singular dependence of T_N on concentrations the peak in the T_N distribution will come near the temperature appropriate to the large concentration limit. This may explain why all samples which ordered had similar apparent Néel temperatures. For a system with a distribution of Néel temperatures it is known that an analysis of the critical scattering, such as that described above, gives a nonzero value of κ at T_N as we indeed observe. The mean-field-like behavior of the intensity at the (1,0,0) reflection has two possible origins. First, the spread in Néel temperatures tends to linearize the behavior near the mean T_N. Second, for systems near the percolation threshold the infinite network is highly ramified with many dangling ends connected by one-dimensional links. These dangling ends will come into registry with the backbone only gradually with decreasing temperature. This could produce the observed linear behavior. As shown in Fig. 18, between 10 and 7 K the inverse correlation length exhibits behavior consistent with that expected[27] for a three-dimensional Ising model. We have argued above that κ does not go to zero at $\langle T_N \rangle \sim 6.4$ K because of the distribution of Néel temperatures. The diffuse scattering below $\langle T_N \rangle$ presumably arises from scattering from each of finite clusters, dangling ends on the infinite network, and critical fluctuations in regions with a reduced T_N.

In conclusion, we believe that these results strongly suggest that the inverse correlation length would decrease to zero at T_N in a homogeneous Ising system, unlike the behavior found in three-dimensional Heisenberg systems.[11] We believe that the unusual results found in the Heisenberg systems are a consequence of the importance of dipolar forces in these systems.

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