# Specific-heat study of random-field and competing-anisotropy effects in $Fe_{1-x}Co_xCl_2$

Po-zen Wong
*Schlumberger-Doll Research, Ridgefield, Connecticut 06877-4108*
(Received 19 December 1985)

We report a comprehensive specific-heat study of the mixed antiferromagnetic system $Fe_{1-x}Co_xCl_2$ with $x=0.286$, 0.366, and 0.604. Four different issues related to competing anisotropies and random fields are discussed. (i) In zero field, a single sharp peak at the Néel transition $T_N$ is seen in each sample. There is no evidence for additional transitions at lower temperatures. We suggest that there are differences between systems with competing Ising-$XY$ anisotropies and those with Ising-Ising anisotropies. Some new neutron scattering data are presented. The behavior of other competing anisotropy systems is discussed. (ii) When a small magnetic field **H** is applied along the easy axis ($c$ axis) in $Fe_{0.714}Co_{0.286}Cl_2$, it exhibits a random-field Ising model (RFIM) behavior similar to the diluted system $Fe_{0.682}Mg_{0.318}Cl_2$. The reduction of $T_N$ obeys the scaling prediction $T_N(H)=T_N(0)-AH^2-BH^{2/\phi}$, where the crossover exponent $\phi$ is found to be $1.24\pm0.09$. The shape of the specific-heat peak also changes continuously with increasing field. This is attributed to crossover effects in constant applied fields. Small thermal hystereses between the field-cooled and field-warmed data were detected in $Fe_{0.682}Mg_{0.318}Cl_2$, but not in $Fe_{0.714}Co_{0.286}Cl_2$. Some previously unpublished results on $Fe_{0.682}Mg_{0.318}Cl_2$ are presented. The measurement of the specific-heat critical exponent $\alpha$ is also discussed. We point out that the standard indirect methods, such as susceptibility and birefringence, may be invalid in finite fields. Our direct specific-heat data suggest that $\alpha$ is large and negative ($\approx -1$). This can be interpreted as a Fisher renormalization effect. (iii) In higher fields, $Fe_{0.714}Cl_{0.286}Cl_2$ has a spin-flop (SF) phase. Sharp cusps are observed at the transitions to the paramagnetic (PM) phase, but no anomalies are observed at the transitions to the low-field uniaxial antiferromagnetic (AF) phase. The AF-PM boundary is found to join smoothly to the SF-PM boundary at an inflection point and the peak near this point shows substantial rounding. Possible experimental and theoretical causes for these observations are discussed. (iv) In $Fe_{0.396}Co_{0.604}Cl_2$, the spins order perpendicular to the $c$ axis in zero field. For applied fields parallel to the $c$ axis, the system should correspond to the three-state Potts model in random fields. We find that the shape of the specific-heat peak changes with increasing field in a manner similar to the RFIM systems, becoming quite symmetric at 19.2 kOe. The phase boundary can be described either by the singular equation given in (ii) with an unusually small crossover exponent $\phi=0.42\pm0.03$, or by an analytic equation involving unusually large $H^4$ and $H^6$ terms. These results are not well understood.

## I. INTRODUCTION

$Fe_{1-x}Co_xCl_2$ is a hexagonal lattice antiferromagnet with competing anisotropies. The Fe spins have an easy direction parallel to the $c$ axis (Ising-like) and the Co spins have an easy plane perpendicular to it ($XY$-like). Susceptibility and neutron scattering experiments reported in a previous paper[1] (hereafter referred to as paper 1) have found a *tetracritical-like* magnetic phase diagram in zero field (Fig. 1). For high Fe concentrations, the spin component parallel to the $c$ axis ($\mathbf{S}_{\parallel}$) orders and, for high Co concentrations, the spin components perpendicular to the $c$ axis ($\mathbf{S}_{\perp}$) order. For intermediate concentrations near $x_m=0.31$, $\mathbf{S}_{\parallel}$ and $\mathbf{S}_{\perp}$ appear to order at two different temperatures. These qualitative features of Fig. 1 can be explained by Landau theory and mean-field calculations.[1-5] A renormalization-group theory by Fishman and Aharony[6] (FA) has predicted further that the ordering of the two components are asymptotically decoupled near the tetracritical point $M$ and the phase boundaries should cross smoothly, but these are not seen in most systems. For $Fe_{1-x}Co_xCl_2$, paper 1 showed that $\mathbf{S}_{\parallel}$ and $\mathbf{S}_{\perp}$ are strongly coupled, and the lower transitions, represented by the lines $CM$ and $DM$ in Fig. 1, are smeared, resembling the behavior of a ferromagnet in applied fields. These results were thought to be caused by random off-diagonal exchange interactions, which can generate ran-

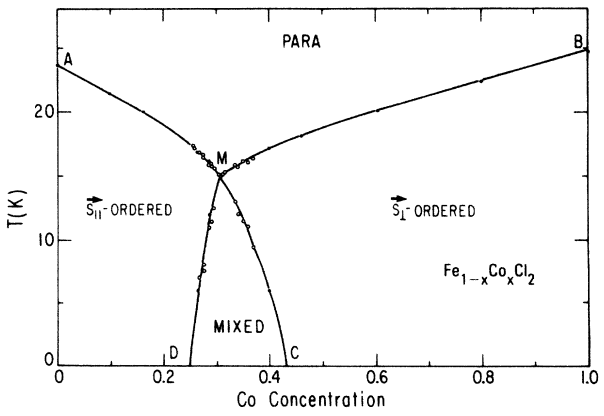


FIG. 1. Magnetic phase diagram, from paper 1.

 

dom molecular fields and destroy the lower transitions. Such interactions should exist because of the lack of local symmetry in a random system,[1,7–10] but they were not included in FA's theory. During the last few years, several other systems with competing Ising $XY$ anisotropies have been studied in detail and they all show similar evidence of $\mathbf{S}_{\|}$-$\mathbf{S}_{\perp}$ coupling.[11–16] In particular, when one component orders at the upper transition (line $AM$ or $BM$ in Fig. 1), it seems to induce some degree of ordering in the other component. In many cases, one of the lower phase boundaries could not even be observed. However, in $Fe_{1-x}Co_xCl_2 \cdot 2H_2O$, which has orthogonal Ising-Ising competing anisotropies, the predictions of FA do seem to be obeyed.[17–20] This was attributed to the quenching of the orbital moment in the $Fe^{2+}$ ion,[20–22] which should give very small off-diagonal terms.

Although the importance of the off-diagonal terms is clear, their effects are not well understood. In particular, when paper 1 was written, it was widely believed that random fields could destroy phase transitions for both Ising and $XY$ systems in three dimensions[23–25] (3D), and thus we interpreted the smearing of the lower transitions as a random-field effect resulting from the off-diagonal terms. However, more recent studies have shown that the random-field Ising model (RFIM) has a well-defined transition in 3D,[26–28] which is only smeared by nonequilibrium effects in the experiments when the system is cooled in the presence of random fields.[29,30] Since the lower transitions are similar to the field-cooled experiments, the argument in paper 1 may still be valid. On the other hand, there are other effects which cannot be explained by random fields. For example, $\mathbf{S}_{\perp}$ was found to have very long correlations well above $T_L$,[1] while random fields should suppress these correlations. In order to gain a better understanding, it would be best to make a more detailed comparison between the lower transitions and other genuine random-field transitions. For example, the transitions along the $AM$ line in Fig. 1 are Ising-like in zero field and they correspond to the RFIM when a field is applied along the easy axis.[31,32] These can be compared with the lower transitions on the $CM$ line. Similarly, the transitions along the upper line $BM$ are $XY$-like. An axial field induces a finite moment $\langle S_{\|}(i) \rangle$ at every site $i$, which produces random molecular fields on $\mathbf{S}_{\perp}$ through the off-diagonal exchange terms.[33] At the same time, Bazhan and Ul'yanov[34] have shown that the crystal symmetry of the system allows a fourth-order term in the Landau free energy of the form $uM_{\|}S_{\perp}^3$, where $M_{\|}$ is the uniform magnetization. This gives a three-state Potts anisotropy. Hence, the transitions should correspond to the three-state Potts model in random fields. According to Mukamel,[35] the transitions along the $DM$ line are also described by the same model, because a $wS_{\|}S_{\perp}^3$ term in the free energy is allowed by symmetry. Thus by studying the upper transitions in applied fields, one can gain some insights on the smeared lower transitions in zero field. Furthermore, if the coupling constant $u$ is large enough, one may be able to observe random-field effects for a three-state Potts system.[36,37]

In this work, we carried out a comprehensive specific-heat study on three $Fe_{1-x}Co_xCl_2$ samples with $x = 0.286$, 0.366, and 0.604. Our objectives are fourfold: $Fe_{0.714}Co_{0.286}Cl_2$ and $Fe_{0.634}Co_{0.366}Cl_2$ have concentrations on different sides of the multicritical point $M$ in Fig. 1. We can characterize the specific-heat behavior of the lower transitions and compare it to the $Fe_{1-x}Co_xCl_2 \cdot 2H_2O$ system which shows sharp lower transitions.[17–20] (ii) $Fe_{0.714}Co_{0.286}Cl_2$ has spins that order uniaxially near the upper transition. We can study its RFIM behavior and compare it with the lower transition in $Fe_{0.634}Co_{0.366}Cl_2$. In addition, because previous neutron scattering studies[29] on $Fe_{0.725}Co_{0.275}Cl_2$ and $Fe_{0.7}Mg_{0.3}Cl_2$ have shown some difference in the nonequilibrium behavior (hysteresis), it is of interest to compare the specific-heat of $Fe_{0.714}Co_{0.286}Cl_2$ to $Fe_{0.682}Mg_{0.318}Cl_2$, which had been studied previously.[32] Some unpublished data on $Fe_{0.682}Mg_{0.318}Cl_2$ are presented here for this purpose. (iii) From a neutron scattering study on $Fe_{0.725}Co_{0.275}Cl_2$,[33] we expect $Fe_{0.714}Co_{0.286}Cl_2$ to have an unusual spin-flop phase in high fields with no long-range order. A specific-heat study will further characterize the behavior of this phase. (iv) The $Fe_{0.396}Co_{0.604}Cl_2$ sample orders like an $XY$ system in zero field. An axial field should cause it to behave like a three-state Potts system in random fields. The results can be compared with both the lower transition of $Fe_{0.714}Co_{0.286}Cl_2$ in zero field, and its high-field transitions associated with the spin-flop phases.

The organization of this paper is as follows. In Sec. II, we describe the technical details of the experimental method. Section III gives the zero-field results of $Fe_{0.714}Co_{0.286}Cl_2$ and $Fe_{0.634}Co_{0.366}Cl_2$. Sections IV, V, and VI describe the effects of applied fields on $Fe_{0.714}Co_{0.286}Cl_2$, $Fe_{0.682}Mg_{0.318}Cl_2$, and $Fe_{0.396}Co_{0.604}Cl_2$, respectively. In each of Secs. III–VI, we also compare our results to other studies on similar systems. Section VII summarizes the current status of competing-anisotropy systems and random-field systems. Some unpublished neutron scattering data on $Fe_{0.725}Co_{0.275}Cl_2$ are presented for the discussion of competing-anisotropy systems. The issue of critical behavior in RFIM systems is discussed in detail. In particular, we point out the problems of using indirect specific-heat techniques, such as the linear birefringence, on random-field systems. Based on our direct specific-heat data, and similar data on $Mn_{0.45}Zn_{0.55}F_2$ by Ikeda and Kikuta,[38] we suggest that the specific-heat exponent $\alpha$ is large and negative ($\approx -1$). The possible theoretical explanations are discussed.[39–42]

## II. EXPERIMENTAL METHOD

Single-crystal boules were grown by the Bridgman method.[43] They have composition gradients typically about 0.01 at.%/cm along the growth direction. To minimize the smearing of the transition due to sample inhomogeneity, we use small samples in the shape of flat platelets, about $3 \times 3 \times 0.2$ mm$^3$ in dimension and 5 mg in weight. This limits the variation in $x$ ($\delta x$) to about 0.003 and the transitions are always sharper than $10^{-2}$ (sometimes as sharp as $10^{-3}$) in reduced temperature $\tau$. Although this is not sufficient for a reliable measurement of the exponent $\alpha$, much useful information can be obtained.

As in our previous work on $Fe_{1-x}Mg_xCl_x$,[32] we used a

thermal relaxation technique suitable for small samples. The large surface of the sample is the natural cleavage plane of the crystal, perpendicular to the $c$ axis. It is glued onto a flat bolometer by melting a small amount of Apiezon-$N$ grease ($\sim$0.2 mg) between the surfaces and pressing them together. The bolometer is made of a 76-$\mu$m-thick sapphire slide slightly larger than the sample. Separate Au-Ge alloy and Cr films were deposited on the back of the bolometer which serves as a resistance thermometer and a heater. Each of these films is attached to two Au—7 at. % Cu wires (76 $\mu$m in diameter and about 1 cm long) which are thermally anchored at a copper block. The block is held at a fixed baseline temperature $T_0$ by a temperature controller. The whole assembly is mounted inside a chamber that can either be filled with exchange gas or evacuated. An additional vacuum jacket isolates the chamber from the helium bath. A superconducting solenoid in the bath provides a vertical magnetic field up to 20 kOe. The gold wires attached to the bolometer serve to suspend it in a slightly flexible way such that the sample's orientation can be adjusted to have the $c$ axis parallel to the field. The wires are strong enough to resist possible magnetic forces (which are less than the sample's weight) due to field inhomogeneity and the sample's anisotropy. Each of the wires is soldered to two electrical leads at the copper block, so that quasi-four-terminal resistance measurements can be made. The resistance versus temperature curves for the films are determined against a calibrated Ge resistance thermometer in zero field. Magnetoresistance correction is negligible over our field range (e.g., at 4.2 K, the correction for 20 kOe is less than 0.05 K).

The principle of the technique is similar to that described by Forgan and Nedjat.[44] The sample and bolometer are assumed to be in thermal equilibrium with each other, i.e., the internal and boundary relaxation times are short compared to the measuring time. The sample chamber is evacuated so that the gold wires and thermal radiation define a fixed thermal link between the bolometer and the copper block. If the sample temperature $T$ is above the baseline temperature $T_0$ at the copper block, it loses heat at a rate $W(T,T_0)$. This heat flux has a negligible effect on the temperature of the copper block because of its much larger thermal mass. If there is a constant heating power $P$ in the Cr film, the sample temperature will change at a rate

$$\frac{dT}{dt}=\frac{P-W(T,T_0)}{C(T)}\ , \tag{1}$$

where $C(T)$ is the total heat capacity of the sample and the bolometer at temperature $T$. The heat-loss function $W(T,T_0)$ can be calibrated by determining the equilibrium temperature $T_p$ for different $P$, because $W(T_p,T_0)=P$ when $dT/dt=0$. If $W(T,T_0)$ is known, the heat capacity $C(T)$ can be determined by measuring $dT/dt$ as a function of $T$ for some given $P$ and substituting the results into Eq. (1). The measurements were made in two different ways in our experiments. First, we let the sample equilibrate at $T_0$ with no power input, a constant power was then turned on at time $t=0$ and the change in the Au-Ge film conductance $G(t)$ was recorded digitally

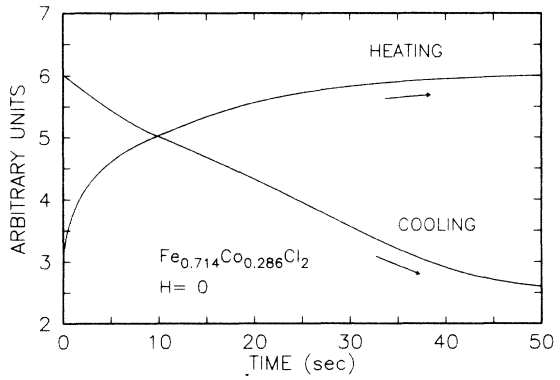


FIG. 2. Conductance of the Au-Ge resistance thermometer recorded upon warming and cooling the sample.

by a computer. After the sample reached its final temperature $T_p$, the power was turned off and $G(t)$ was again recorded. A set of such data for $Fe_{0.714}Co_{0.286}Cl_2$ in zero field is shown in Fig. 2. These traces are digitized at a rate of ten points per second and each trace consists of several hundred points. The sample temperature $T(t)$ at each point is determined from the bolometer calibration and a piecewise cubic fit gives $dT/dt$ versus $T$. The heat capacity $C(T)$ is then determined from Eq. (1). If the absolute specific heat is of interest, the heat capacity of the bolometer and the grease can be measured and subtracted. The contribution from the grease can also be estimated from literature data.[45,46] We have found empirically that these are small and smooth contributions which have little effect on observations related to the phase transitions. Hence, all the data presented in this paper are the total heat capacity of the sample plus the addenda, without any background subtraction or normalization to the sample's mass. The validity of the technique was established by measuring pure $FeCl_2$.[32]

Figure 3 shows the $C$ versus $T$ curves obtained from the traces in Fig. 2. The warming curve is shifted up for clarity. We note that the warming curve is truncated at

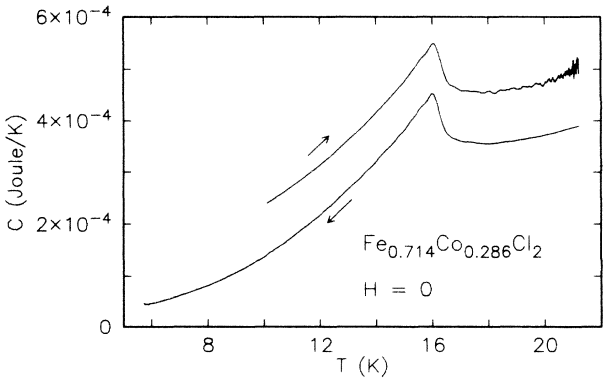


FIG. 3. Heat capacity of the $Fe_{0.714}Co_{0.286}Cl_2$ sample in zero field. The two curves are obtained from the two traces shown in Fig. 2. The warming curve is shifted up by one vertical division for clarity. There is no evidence of a lower transition in the 10—12 K range.

low temperature and more noisy at high temperature. The reason is that the heat capacity is small at low temperatures. When a constant power $P$ is applied, the samples heat up very rapidly (as manifested by the steep initial slope in Fig. 2) and $dT/dt$ cannot be obtained accurately. Near the maximum temperature $dT/dt$ is very small and, hence, less accurate. In principle, these problems can be overcome by using a heater power that increases with time. In practice, we find that the heating curve matches very well the cooling curve (see Fig. 3) and the latter alone is sufficient for most purposes. Nevertheless, heating data were always obtained to provide a comparison. For example, the peak positions of the two curves in Fig. 3 actually differ by about 0.04 K (at $\sim 16$ K). This indicates a slight thermal lag between the bolometer and the sample, due to the thermal resistance of the thin layer of $N$ grease.[47] By averaging the peak temperatures of the heating and cooling data, this systematic error can be reduced and $T_N$ can be determined with a 0.01-K resolution.

## III. $Fe_{0.714}Co_{0.286}Cl_2$ AND $Fe_{0.634}Co_{0.366}Cl_2$ IN ZERO FIELD: COMPETING ANISOTROPIES

Our first effort was to study the zero-field behavior of $Fe_{0.714}Co_{0.286}Cl_2$ and $Fe_{0.634}Co_{0.366}Cl_2$. According to the susceptibility and neutron data in paper 1, $Fe_{0.714}Co_{0.286}Cl_2$ has an upper transition at $T_N \approx 16.1$ K and a lower transition at $T_L \approx 11.5$ K. For $Fe_{0.634}Co_{0.366}Cl_2$, $T_N \approx 16.3$ K and $T_L \approx 10.0$ K. The specific-heat data for these two samples are shown in Figs. 3 and 4. Cusplike second-order transition peaks are observed at 16.05 and 16.3 K, respectively, in good agreement with the values of $T_N$ obtained previously. However, there are no anomalies of any kind in the 10–12-K temperature range for the lower transitions. Very recently, Nitsche and Kleemann[48] have used refractive index measurements to deduce the magnetic specific heat in samples with $x$ between 0.20 and 0.38. There was also no evidence for any lower transition. Although these results are consistent with the smeared lower transitions observed in the earlier experiments, we shall see in Secs. IV and V that these are not characteristics of random-field systems.

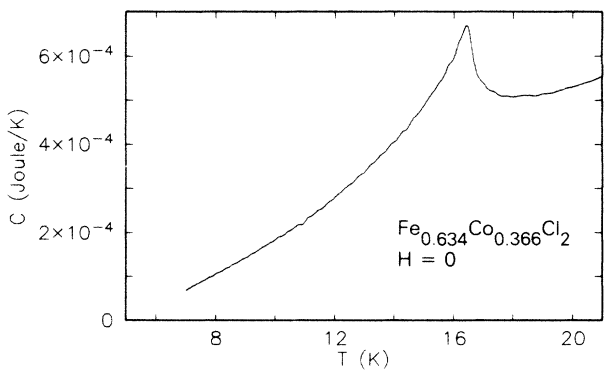


FIG. 4. Heat capacity of the $Fe_{0.634}Co_{0.366}Cl_2$ sample in zero field obtained from cooling data. There is also no evidence for a lower transition.

Among the other competing-anisotropy systems, the hydrated $Fe_{1-x}Co_xCl_2 \cdot 2H_2O$ is the only one that had been studied by specific-heat measurements.[17] In that study, two successive peaks were clearly seen over a wide range of concentrations ($0.45 < x < 0.65$), even when $T_L$ is more than a factor of 2 lower than $T_N$. Such behavior is in sharp contrast with our data in Figs. 3 and 4, which show no lower peak for $x$ very close to $x_m$ and much smaller separations of $T_N$ and $T_L$. This difference between the two systems is consistent with how the spin correlations develop between $T_L$ and $T_N$. In the hydrated system, a recent neutron scattering by Katsumata *et al.*[20] has shown that the correlations of the spin component associated with the lower transition grew in a small temperature range above $T_L$, resulting in a sharp critical scattering peak at $T_L$, characteristic of a second-order transition. For the anhydrous system, however, paper 1 shows that the $S_\perp$-correlation length becomes very long when $S_\parallel$ orders. One would thus expect a much smaller entropy change at $T_L$ and possibly no detectable anomaly in the specific heat. In Sec. VII, we address the question of whether $S_\perp$ has true long-range order immediately below the upper transition.

As mentioned in Sec. I, the hydrated $Fe_{1-x}Co_xCl_2 \cdot 2H_2O$ system is the only one studied to date which has competing Ising-Ising anisotropies, and it seems to obey all of FA's predictions.[17–20] In addition to the sharp lower transitions, it appears to have smoothly crossing phase boundaries. Katsumata *et al.*[20–22] have suggested a logical explanation for this behavior, namely, that the orbital moment of the $Fe^{2+}$ ion is nearly quenched (it has $g = 2.23$ and $S = 2$) and this should result in small off-diagonal exchange terms. Unfortunately, several recent studies provide counter examples of this idea. In $Fe_{1-x}Ni_xCl_2$, the $Ni^{2+}$ ion has $g = 2.24$ and $S = 1$,[49] but the lower $XY$ line (like $DM$ in Fig. 1) was not observed and $S_\parallel$ seems to order at temperatures much above the $CM$ line;[12] in $Mn_{1-x}Fe_xCO_3$, the $Mn^{2+}$ ion ($g = 2$, $S = \frac{5}{2}$) has only dipolar anisotropy, but a Mössbauer study has found smeared lower transitions;[16] in another Mn compound, $K_2Mn_{1-x}Fe_xF_4$, no critical scattering peaks were observed at $T_L$ and very long two-dimensional (2D) correlations were believed to exist well above $T_L$.[50] These three systems all have negligible orbital moments but their behavior is very different from $Fe_{1-x}Co_xCl_2 \cdot 2H_2O$. Instead, they are more like the anhydrous $Fe_{1-x}Co_xCl_2$ which has large orbital moments ($g = 4$ for the $Fe^{2+}$ ion and $g = 6$ for the $Co^{2+}$ ion).[51] We note that strong departures from the FA predictions have also been seen in several other systems with unquenched orbital moments. In $Fe_{1-x}Co_xTiO_3$ and $K_2Fe_{1-x}Co_xF_4$, a lower Ising line (like $CM$ in Fig. 1) was not observed.[11,13,14] In $Fe_{1-x}Co_xBr_2$, a lower $XY$ line was not observed.[15] Not only do these systems have different $g$ factors, they also have lattice structures that range from rhombohedral to tetragonal. The latter suggests that the weakness of the FA theory is not limited to the absence of Mukamel's $wS_\parallel S_\perp^3$ term,[35] because such a term is only applicable to systems with three-fold symmetry. The most obvious common feature for all the systems, besides the hydrated $Fe_{1-x}Co_xCl_2 \cdot 2H_2O$, is that they have com-

peting Ising-$XY$ anisotropies. It is thus tempting to infer that these systems are fundamentally different from the ones with Ising-Ising anisotropies,[52] but we have no good arguments to support this conjecture. (In fact, there was an early study of another Ising-Ising system $Ni_xCo_{1-x}Cl_2 \cdot 2H_2O$ which has two competing easy axes 115° apart,[53] and no evidence of any lower transition was observed. Unfortunately, only polycrystalline samples were studied.)

## IV. $Fe_{0.714}Co_{0.286}Cl_2$ IN APPLIED FIELDS: A MIXED ISING SYSTEM IN RANDOM FIELDS

As mentioned in Sec. I, the upper transition of $Fe_{0.714}Co_{0.286}Cl_2$ should correspond to the RFIM when a field is applied along the $c$ axis.[31,32] Figure 5(a) shows the data in six different fields. We observe that there is a continuous change in the shape of the peak with increasing field, a behavior similar to the $Fe_{1-x}Mg_xCl_2$ system.[32] We note that all the data shown in Fig. 5(a) are taken by field cooling the sample from the paramagnetic phase and that the peak at 3.51 kOe has a rounding of about 0.1 K, limited by the concentration gradient. Figure 5(b) shows that there is no significant difference between the field-cooled and field-warmed data in that field. We show in Sec. V that a slightly bigger difference can be observed in the diluted $Fe_{0.682}Mg_{0.318}Cl_2$ system. For fields above 4 kOe, the peak in $Fe_{0.714}Co_{0.286}Cl_2$ broadens rapidly. The 4.49-kOe data in Fig. 5(a), for example, show a rounding of order 1 K. In higher fields (e.g., 9.60 kOe), the peak evolves into a sharper cusp.

Field-warmed and field-cooled data were taken in 25 different fields. The average peak temperature for each field is chosen as the transition temperature and shown as the solid circles in Fig. 6. They form a smooth line with an inflection point near 5 kOe. To understand this, we show, in the same figure, the $H$-$T$ phase diagram obtained previously for $Fe_{0.725}Co_{0.275}Cl_2$ by neutron scattering[33] (represented by the open circles and dashed lines). The latter is known to have a uniaxial antiferromagnetic (AF) phase in low fields and a disordered spin-flop (SF) phase in higher fields. This SF phase consists of large domains ($\sim 10^3$ Å) of spins ordered perpendicular to the $c$ axis and this is believed to be caused by the random fields due to the off-diagonal terms. Comparing the two sets of data in Fig. 6, we observe that $Fe_{0.714}Co_{0.286}Cl_2$ has a slightly lower $T_N$ in zero field, consistent with its higher Co concentration. Consequently, we also expect this sample to have a SF phase in lower fields and, crudely speaking, the inflection point near 5 kOe might be regarded as a "psuedo-bicritical-point." The specific-heat peaks below this field correspond to transitions between the AF phase and the paramagnetic (PM) phase, and those above it correspond to SF-PM transitions. The AF-SF transition is not observed in the present experiment. This may be due to the fact that the AF-SF line is almost parallel to the $T$ axis. On the other hand, we note that this transition is a spin-reorientation transition like the lower transition in zero field. The absence of a specific-heat anomaly may also be related to the existence of long $S_\perp$ correlations in the AF phase.

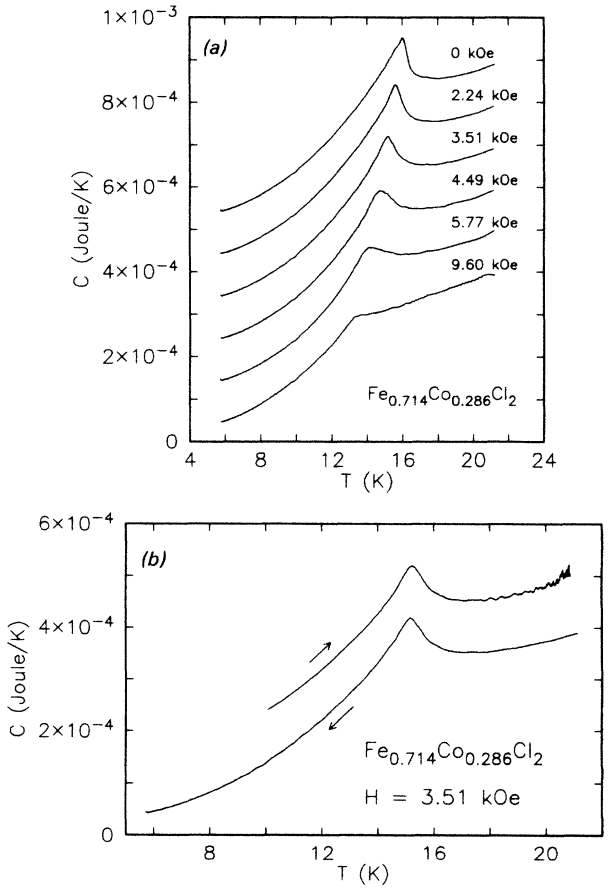


FIG. 5. (a) Heat capacity of the $Fe_{0.714}Co_{0.286}Cl_2$ sample in different fields. The vertical scale applies to the bottom curve. Other curves are displaced successively upward by one division. All of the data are obtained by field cooling the sample. Rounding of the peak is seen around 5 kOe. (b) A comparison of the cooling and warming data at 3.51 kOe shows no significant difference.

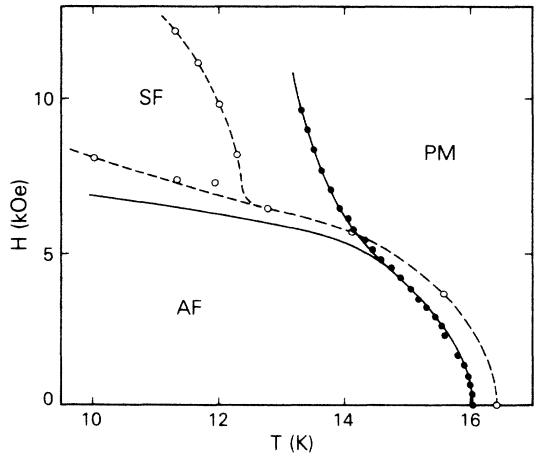


FIG. 6. A comparison of the $T$-$H$ phase diagram of the $Fe_{0.714}Co_{0.286}Cl_2$ sample obtained in this work (solid circles) and that of $Fe_{0.725}Co_{0.275}Cl_2$ from Ref. 33 (open circles).

It is interesting to observe that the AF-PM and SF-PM boundaries in Fig. 6 join smoothly and the specific-heat peak is severely rounded near 5 kOe. Similar rounding (but less severe) had also been observed in $Mn_{0.75}Zn_{0.25}F_2$ by Shapira *et al.*[30] These are in contrast to the bicritical behavior in pure antiferromagnets.[54] There are many possible explanations, e.g., sample misalignment,[54] demagnetization effect,[55] impurity smearing of the first-order AF-SF line,[56] and nonequilibrium effect due to the random fields.[29,30,39] We should also point out that the intrinsic phase diagram of $Fe_{0.714}Co_{0.286}Cl_2$ may not be bicritical even in the absence of all these effects. As noted in Sec. I, the applied field induces three-fold in-plane anisotropy and the SF phase should behave like a three-state Potts system in random fields. According to recent theories, such systems should have first-order transitions with or without random fields.[36,37] Hence, the phase diagram cannot be bicritical, because the bicritical point is by definition the meeting point of two *second-order* boundaries. In other words, there should be a totally different description for spin-flop phenomena in Potts systems. Consequently, how we should interpret the specific-heat cusp at the SF-PM boundary is unclear. This is an interesting problem that needs more careful study.

The shape of the AF-PM boundary in low fields can be analyzed as a random-field crossover effect. According to FA,[31]

$$\Delta T_N \equiv T_N(H) - T_N(0) = -AH^2 - BH^{2/\phi} . \quad (2)$$

This prediction is based on the assumption that the random fields are proportional to the applied field. Since the random fields are partly due to the uniform magnetization, it would require $M_c(H) \propto H$, where $M_c$ denotes the magnetization at the transition. We have found this to be a good approximation in our previous study of the $Fe_{1-x}Mg_xCl_2$ system.[32] Independent work by Gelard *et al.* also confirmed this approximation.[57] Hence, we shall assume Eq. (2) to be true for $Fe_{0.714}Co_{0.286}Cl_2$ in our analyses. We note that the analytic $AH^2$ term represents the simple fact that the applied field favors a uniform magnetization over a staggered magnetization, an effect that can be explained by mean-field theory.[58] The singular term $BH^{2/\phi}$ is due to random-field effects and $\phi$, the crossover exponent, is predicted to be the susceptibility exponent $\gamma$ in zero random field. Since the exchange interactions are site random, one might expect $\phi$ to be the random-Ising exponent $\gamma_{\text{random}}$. However, the crossover from pure-Ising to random-Ising behavior is very slow and it is unclear what exponent should be observed. Moreover, $\gamma_{\text{pure}}$ is well known to be 1.24 theoretically[59] and the best estimate for $\gamma_{\text{random}}$ is $1.34 \pm 0.05$.[60] This difference is too small to be distinguished in most experiments but, since the singular term's exponent $2/\phi$ is less than 2 for any $\phi > 1$, its effect can be detected by a plot of $H^2$ versus $T_N$.

In Fig. 7, we show an $H^2$ versus $T$ plot of the phase boundary of pure $FeCl_2$ up to 8 kOe.[32] The line represents a least-squares fit of the data to the analytic $AH^2$ term with two adjustable parameters, $T_N(0)$ and $A$. The standard deviation $\sigma$ of the fit is 11.5 mK, about the same as the spacing between data points in our $C$ versus $T$ curves ($\sim 10$ mK). $T_N(0)$ was found to be $23.55 \pm 0.01$ K, in excellent agreement with the literature value for $FeCl_2$.[61] The prefactor $A$ was found to be 0.0149 K/kOe$^2$ with negligible statistical error. These results demonstrate that, in the absence of random fields, the analytic term describes the phase boundary extremely well. [We note that

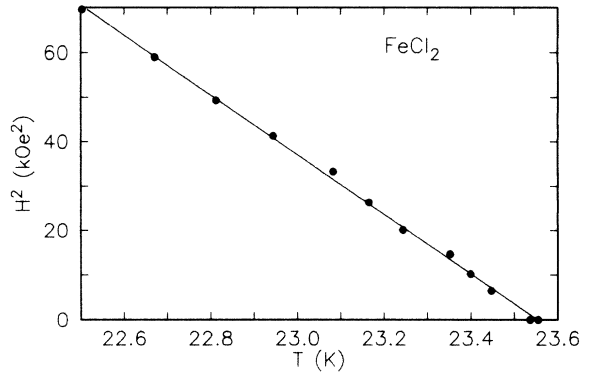


FIG. 7. The low-field phase boundary of pure $FeCl_2$ (from Ref. 32). The straight line is a least-squares fit of the data points, which shows that $\Delta T_N(H) \propto -H^2$.

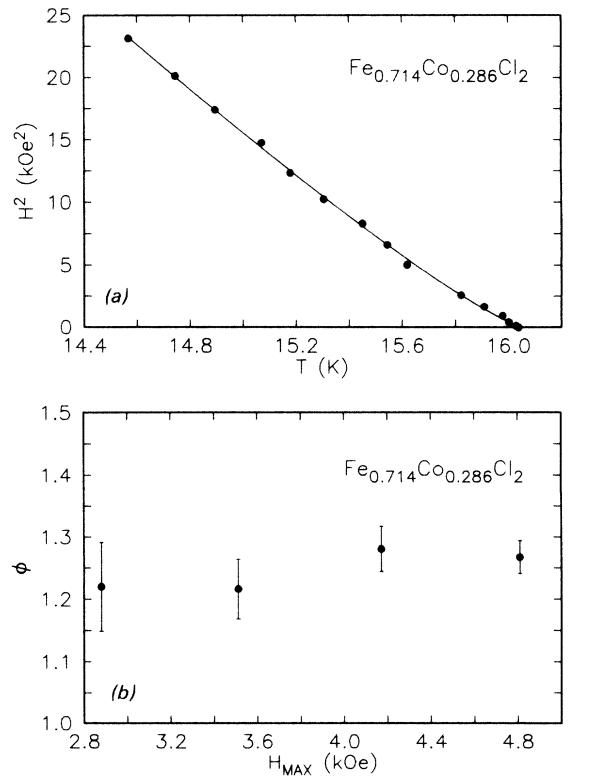


FIG. 8. (a) The low-field phase boundary of $Fe_{0.714}Co_{0.286}Cl_2$ shows a concave curvature in an $H^2$ versus $T$ plot. The line is a fit of all the points to Eq. (2), which gives $\phi = 1.27 \pm 0.03$. (b) Results for $\phi$ obtained by varying the maximum field in the fit. $\sigma$ for the four fits are, with increasing $H_{\max}$, 13.8, 12.8, 13.2, and 12.7 mK.

a mean-field theory calculation, using Eq. (2) from Ref. 58 and the exchange parameters given in Ref. 51, gives $A=0.065$ K/kOe$^2$, comparable to the measured value. The difference can be *partly* attributed to demagnetization effect: the susceptibility of $FeCl_2$ is about 0.02 emu/cm$^3$ near the transition, which can cause a 25% difference between the internal field and the applied field.] In contrast, Fig. 8(a) shows a similar plot of the phase boundary of $Fe_{0.714}Co_{0.286}Cl_2$ up to 4.8 kOe. It clearly deviates from a straight line. The concave curvature is indicative of the $BH^{2/\phi}$ term in Eq. (2) with $\phi>1$.

An analysis for $\phi$ cannot be performed by directly fitting the data to Eq. (2), because the $BH^{2/\phi}$ and $AH^2$ terms are highly correlated. Instead, we estimate the value of $A$ and fix it in the fit. In any theory, Eq. (2) should be written in a dimensionless form which has all the temperatures and fields normalized with respect to the exchange $J$. This implies that the prefactor $A$, as written in Eq. (2), should be inversely proportional to $J$, and hence $A\propto 1/T_N(0)$. Since $A$ and $T_N(0)$ are known for pure $FeCl_2$, we can estimate $A$ for $Fe_{0.714}Co_{0.286}Cl_2$ from its zero-field transition temperature ($\sim 16.05$ K):

$$A=0.0149\times(23.55/16.05)$$
$$=0.0219 \text{ K/kOe}^2 .$$

Although $A$ is sensitive to demagnetization, we do not make such corrections, because the susceptibility of $Fe_{0.714}Co_{0.286}Cl_2$ at the transition is nearly the same as pure $FeCl_2$ ($\approx 0.02$ emu/cm$^3$).[1] When the data ($T$ in units of K and $H$ in units of kOe) are fitted to Eq. (2) with three adjustable parameters, we obtain

$$\phi=1.27(3),\quad B=0.081(4),\quad T_N(0)=16.05(1) \text{ K} .$$

The solid line in Fig. 8(a) is the result of the fit. Since Eq. (2) is valid only in the $H\rightarrow 0$ limit, we also fitted the data for different maximum fields ($H_{\max}$), above which the data is excluded. We find $\phi$ ranges from 1.22 to 1.28 for $H_{\max}$ between 2.9 and 4.8 kOe. The amplitude $B$ ranges from 0.076 to 0.083. The quality of these fits is practically the same, with $\sigma$ between 12.7 and 13.8 mK, comparable to the experimental resolution. These results indicate the insensitivity to the choice of $H_{\max}$. Figure 8(b) shows the fitted values of $\phi$ for different $H_{\max}$. The maximum variation of $\phi$, including the statistical errors, is between 1.15 and 1.32. Using these as upper and lower bounds, we estimate

$$\phi=1.24\pm 0.09 .$$

This result is in agreement with $\gamma_{\text{pure}}$. However, because there are additional uncertainties involved in estimating $A$, it is not inconsistent with $\gamma_{\text{random}}$. In the next section, we show that a nearly identical result ($\phi=1.25\pm 0.11$) is obtained in $Fe_{0.682}Mg_{0.318}Cl_2$. In other systems, Shapira *et al.* obtained $\phi=1.25\pm 0.07$ in $Mn_{0.75}Zn_{0.25}F_2$,[30] also in agreement with our results. However, somewhat higher values were found in $Fe_{1-x}Zn_xF_2$ (1.4 and 1.5) and these had been suggested as evidence for $\gamma_{\text{random}}$.[62,63] This is not unreasonable because $FeF_2$ is a more ideal 3D Ising system with $\alpha>0$.[64] $MnF_2$ has smaller anisotropy and $FeCl_2$ is less three dimensional; they have effective exponents $\alpha\lesssim 0$ for reduced temperatures $\tau\gtrsim 10^{-3}$.[64,65] Thus it is possible that the crossover from pure to random Ising behavior is more difficult to detect in these systems. On the other hand, one cannot be certain of this interpretation for many reasons. First, the difference between $\gamma_{\text{random}}$ and $\gamma_{\text{pure}}$ is very small. Second, the estimate for the analytic $AH^2$ is an uncontrolled approximation. Third, Hutchings *et al.*[66] have found $\gamma_{\text{pure}}=1.38\pm 0.08$ in pure $FeF_2$ and Birgeneau *et al.*[67] have found $\gamma_{\text{random}}=1.44\pm 0.06$ in $Fe_{0.5}Zn_{0.5}F_2$. These results are indistinguishable, but they differ from pure $MnF_2$, which has $\gamma_{\text{pure}}=1.27\pm 0.02$.[68] The cause is not understood. Finally, Kaufman and Kardar have found that $\phi=1.5$ in mean-field theory.[69] This is not inconsistent with the $Fe_{1-x}Zn_xF_2$ results.

## V. $Fe_{0.682}Mg_{0.318}Cl_2$ IN APPLIED FIELDS: A DILUTE ISING SYSTEM IN RANDOM FIELDS

During our earlier specific-heat study of $Fe_{1-x}Mg_xCl_2$,[32] little was known about the nonequilibrium effects in random-field systems. For each applied field, data were collected by first warming the sample and then cooling it, after which the field was increased while the sample was at its lowest temperature and the warming-cooling sequence repeated. Although the peak was found to be slightly more rounded in the field-cooled data, the difference was considered too small to be significant. The $Fe_{1-x}Co_xCl_2$ data in this paper were obtained shortly thereafter using the same procedure. The fact that the field-cooled peak in $Fe_{0.714}Co_{0.286}Cl_2$ is as sharp in 3.51 kOe as in zero field first suggested that there might be nonequilibrium effects that are more severe in a diluted system (Mg doped) than in a undiluted mixed system (Co doped). Subsequent neutron scattering studies[29] of $Fe_{0.7}Mg_{0.3}Cl_2$ and $Fe_{0.725}Co_{0.275}Cl_2$ have confirmed that the hysteresis in the Mg-doped system is indeed stronger. To make the comparison between the two systems more complete, we present here some previously unpublished results of $Fe_{0.682}Mg_{0.318}Cl_2$.

Figures 9(a)—9(d) show the field-cooled and field-warmed curves for four different applied fields. The 0.964-kOe data show that, within the experimental resolution, the two curves are indistinguishable. As the field increases, the field-cooled peak becomes slightly more rounded than the field-warmed peak. The difference is barely noticeable in the 2.88-kOe data, but becomes more apparent in the 3.26- and 4.80-kOe data. The data we previously published in Ref. 32 were all field cooled except those for $Fe_{0.682}Mg_{0.318}Cl_2$ at 3.52 and 4.48 kOe. Figures 9(c)—9(d) show the small difference that exists in this field range. If one attempts to integrate these curves to obtain the entropy or the energy, the difference will clearly be below the noise level. (A difference should exist because the field was always increased at the lowest temperature.) We note that other studies using indirect methods, such as thermal expansion and birefringence,[30,62] to measure the specific heat have found much stronger hysteresis. The birefringence work has also suggested $\alpha=0$. A possible interpretation for these experiments is given in Sec. VII.

Aside from the subtle difference in hysteresis, the behavior of the diluted $Fe_{0.682}Mg_{0.318}Cl_2$ is very similar to the nondiluted $Fe_{0.714}Co_{0.286}Cl_2$. Most notably, Figs. 5 and 9 show a very similar change in the peak shape. We have suggested before that this can be caused by *crossover* effects in a constant applied field,[32] because the random field $H_R$ contains contributions from both the applied field $H$ and the uniform magnetization $M$, and the latter is strongly temperature dependent in a constant applied field. This effect is illustrated in Fig. 10, where we show that the path $P$ taken by the sample in the $T$-$H_R$ plane is *not* a straight line parallel to the $T$ axis. This effect should be more pronounced for $Fe_{0.714}Co_{0.286}Cl_2$ and $Fe_{0.682}Mg_{0.318}Cl_2$ than for the 3D fluorides,[30,38,62] because the former are metamagnets which have large magnetizations in small applied fields. In Sec. VII, we discuss how this might affect the critical behavior.

The shape of the $T$-$H$ phase boundary in $Fe_{0.682}Mg_{0.318}Cl_2$ is also similar to $Fe_{0.714}Co_{0.286}Cl_2$. Although an analysis of this had been reported before,[32] we present the results here in a form that can be compared more readily to $Fe_{0.714}Co_{0.286}Cl_2$. Figure 11(a) shows the phase boundary in an $H^2$ versus $T$ plot. The data points represent the average peak temperatures. A slightly concave curvature similar to Fig. 7(a) is observed. Our method of analysis is the same as that described above for $Fe_{0.714}Co_{0.286}Cl_2$. Since the zero-field transition occurs at $T_N(0)=12.78$ K, the analytic term in Eq. (2) is held fixed at $A=0.0149\times(23.55/12.78)=0.0275$ K/kOe$^2$. (We do not make a demagnetization correction on $A$ because the susceptibility of $Fe_{0.682}Mg_{0.318}Cl_2$ at $T_N$ is nearly the same as $FeCl_2$).[57] A three-parameter fit of Eq. (2) to the data in Fig. 11(a) gives

$$T_N(0)=12.78(1)\text{ K},\quad B=0.120(7),\quad \phi=1.26(4)\ .$$

Varying $H_{\max}$ for the fit from 2.2 to 4.3 kOe gives $\phi$ between 1.20 and 1.32, as shown in Fig. 11(b). $\sigma$ for these fits ranges from 10.1 to 11.5 mK, comparable to the experimental resolution. Including the statistical error bars in Fig. 11(b), $\phi$ ranges from 1.13 to 1.36. Using these numbers as upper and lower limits, we estimate

$$\phi=1.25\pm0.11\ ,$$

in agreement with $Fe_{0.714}Co_{0.286}Cl_2$. We also verified that this result is quite insensitive to the chosen value of $A$. Setting $A=0$ for the data in Fig. 11(a), for example, gives $\phi=1.20$. The change is less than the estimated error of $\pm0.11$. For the prefactor $B$, it varies from 0.113 to 0.126 over the same range of $H_{\max}$. We cannot estimate the error in $B$ because it is strongly dependent on the assumed value of $A$. However, it is interesting to note that the value of $B$ in $Fe_{0.682}Mg_{0.318}Cl_2$ is comparable to that in $Fe_{0.714}Co_{0.286}Cl_2$, which implies that the magnitudes of the random fields in the two systems are comparable for the same applied fields. This is also consistent with the observation that the two systems have a similar degree of change in the peak shape (Figs. 5 and 9).

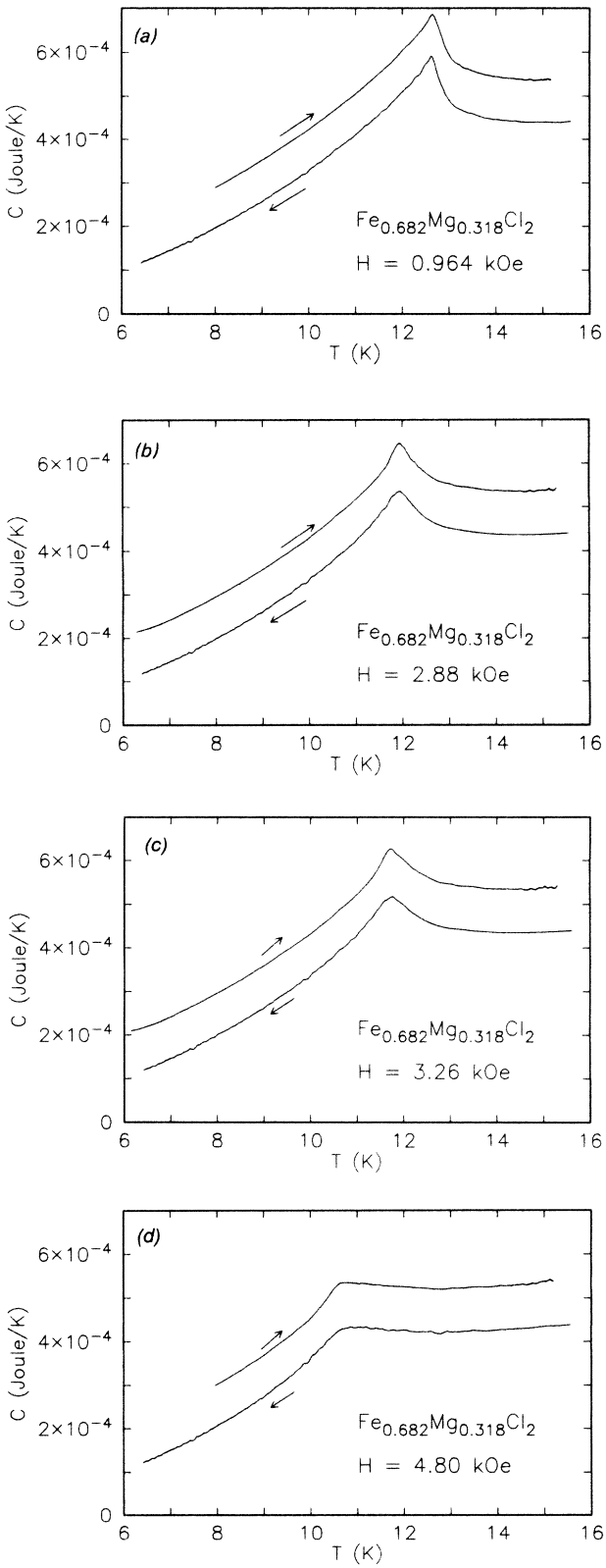


FIG. 9. Comparison of field-warmed and field-cooled data in $Fe_{0.682}Mg_{0.318}Cl_2$ shows that there is a barely detectable difference in the sharpness of the peak as the field increased. The field-cooled peak is slightly more rounded. These small differences are in contrast with the strong hystereses observed in the indirect specific-heat measurements. The reasons are discussed in Sec. VII B.

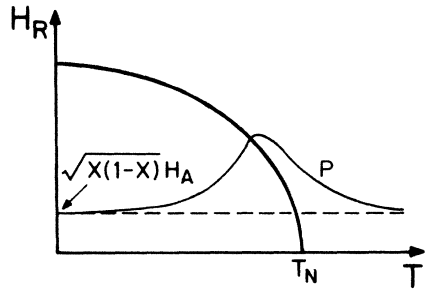


FIG. 10. For a constant applied field, the random fields in the sample are temperature dependent, because they are related to the magnetization. The path taken by the sample in the $T$-$H_R$ plane is thus not a straight line parallel to the $T$ axis. This effect can qualitatively explain the change of peak shape seen in Figs. 5, 9, and 11. It can also lead to Fisher renormalization of the critical exponents (see Sec. VII B).

## VI. $Fe_{0.396}Co_{0.604}Cl_2$ IN APPLIED FIELDS: A POSSIBLE REALIZATION OF THE THREE-STATE POTTS MODEL IN RANDOM FIELDS

In zero field, the spins in $Fe_{0.396}Co_{0.604}Cl_2$ order in the $a$-$b$ plane with negligible in-plane anisotropy. Previous measurements on other $Fe_{1-x}Co_xCl_2$ samples have found that the order-parameter exponent has a value of $\beta \approx 0.33$ along the $BM$ line in Fig. 1, consistent with 3D $XY$ behavior.[1] When a field is applied along the $c$ axis, out-of-plane magnetic moments are induced and, similar to the SF phase in $Fe_{0.714}Co_{0.286}Cl_2$, the system should cross over to the behavior of the three-state Potts model in random fields. Therefore, one would expect to see drastic changes in the transition.

The results for fields up to 19.2 kOe, the maximum in our apparatus, are shown in Fig. 12(a). These data were all obtained by field cooling. Figure 12(b) illustrates that there is no significant difference in the field-warmed data. We observe that the peak shape changes in a manner similar to $Fe_{0.714}Co_{0.286}Cl_2$ and $Fe_{0.682}Mg_{0.318}Cl_2$ (Figs. 5 and 9). At 19.2 kOe, for example, the peak is quite symmetric. This is suggestive of the random-field crossover effect depicted in Fig. 10. We note that there is no significant smearing of the peak even at the highest field. This is consistent with the existence of the Potts anisotropy, because if the system were purely $XY$-like, the random fields should have destroyed the transition com-

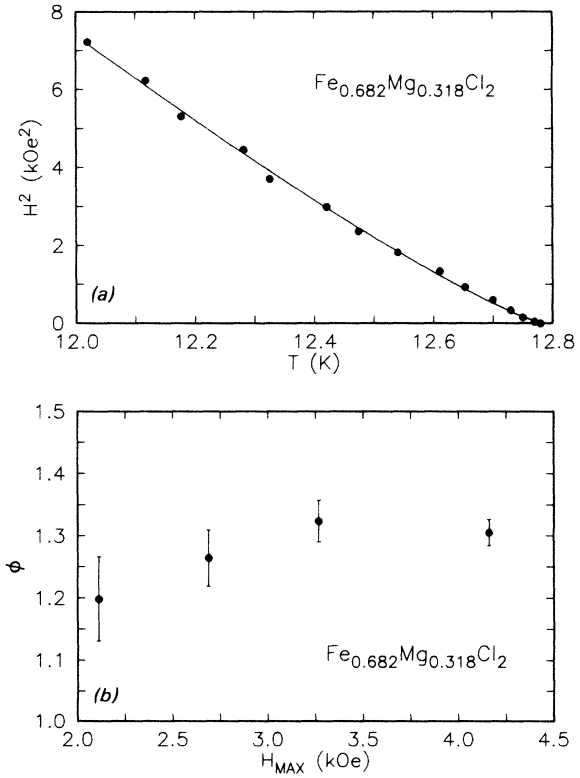


FIG. 11. (a) The low-field phase boundary of $Fe_{0.682}Mg_{0.318}Cl_2$ is very similar to that of $Fe_{0.714}Co_{0.286}Cl_2$, showing a weak concave curvature. The line is a fit of the data points to Eq. (2), which gives $\phi = 1.26 \pm 0.04$. (b) Variation of $\phi$ with $H_{max}$ is insignificant. $\sigma$ for the four fits are, with increasing $H_{max}$, 10.1, 10.3, 11.2, and 11.3 mK.

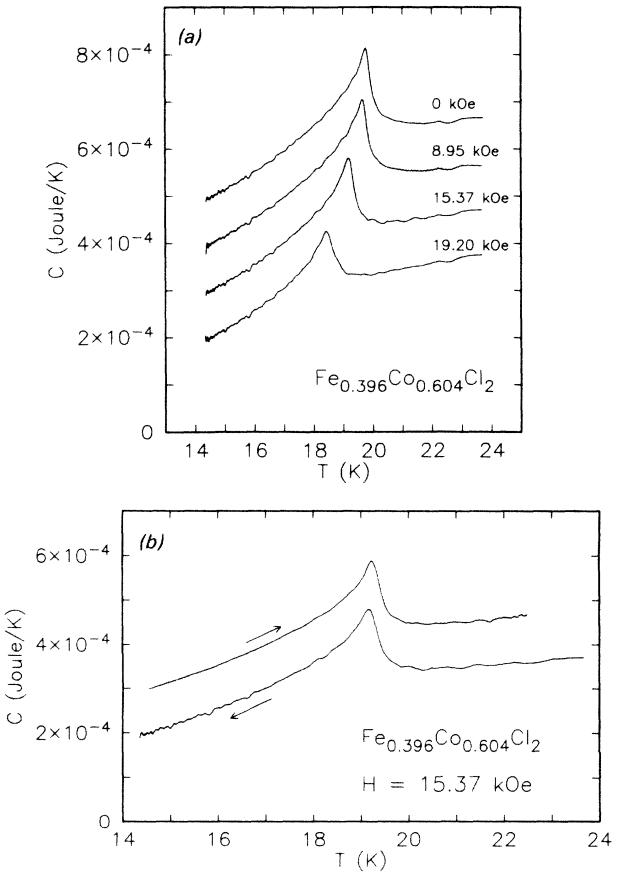


FIG. 12. (a) Heat capacity of the $Fe_{0.396}Co_{0.604}Cl_2$ sample in different applied fields. The vertical scale refers to the bottom curve. Other curves are displaced upward successively by one division. The change in peak shape with increasing field is very similar to Figs. 5 and 9, suggestive of random-field effects. (b) A comparison of the cooling and warming data at 15.37 kOe, which shows no significant difference.

pletely. Most interestingly, we find that a plot of the average peak temperatures on an $H^2$ versus $T$ scale (Fig. 13) shows a large *convex* curvature, in contrast to the *concave* curvatures seen in Figs. 8 and 11. This implies that a fit to Eq. (2) will give $\phi<1$, as opposed to $\phi>1$ in $Fe_{0.714}Co_{0.286}Cl_2$ and $Fe_{0.682}Mg_{0.318}Cl_2$.

Before discussing these results further, we have to first rule out the possibility that they are caused by a misalignment of the field with respect to the $c$ axis. We estimate that the accuracy of our alignment is not worse than 5°, which means that even for $H=19.2$ kOe, the in-plane component is less than 1.7 kOe. According to a specific-heat study of pure $CoCl_2$ by Moses *et al.*,[70] a 15-kOe in-plane field reduces its $T_N$ by about 1 K. Compared to the value of $T_N$, this reduction is about 4%. Assuming $\Delta T_N \propto H^2$ from mean-field theory, a 1.7-kOe in-plane field should reduce $T_N$ by about 0.05% in $CoCl_2$. Although the reduction should be somewhat larger in $Fe_{0.396}Co_{0.604}Cl_2$ due to its weaker interactions, the data in Fig. 13 gave a $\Delta T_N/T_N(0)$ of about 7% at 19.2 kOe. This hundredfold increase cannot be the result of a small in-plane field, but it could be a random-field effect, e.g., the $Fe_{0.714}Co_{0.286}Cl_2$ data in Fig. 8(a) show a 9% change. We also note that the specific-heat peak of pure $CoCl_2$ at 15 kOe has an asymmetric shape similar to that in zero field.[70] This is in contrast to the symmetric peak observed in $Fe_{0.396}Co_{0.604}Cl_2$ at 19.2 kOe (Fig. 12). Hence, it is highly unlikely that the unusual behavior observed in $Fe_{0.396}Co_{0.604}Cl_2$ is an artifact of misalignment.

The phase boundary in Fig. 13 was first analyzed by fitting to Eq. (2). Because the curvature is strong, it was possible to perform the fit without fixing $A$. We obtained

$$T_N(0)=19.79(1)\ \text{K},\quad A=1.2(2)\times10^{-3}\ \text{K/kOe}^2,$$
$$B=7.2(6.6)\times10^{-5},\quad \phi=0.42(3).$$

$\sigma$ for the fit is 10.0 mK, comparable to the experimental resolution. The line in Fig. 13 is the fitted curve. We note that the value of $A$ for this sample is about an order of magnitude smaller than those $Fe_{0.714}Co_{0.286}Cl_2$ and $Fe_{0.682}Mg_{0.318}Cl_2$. This is to be expected because the mean-field effect should be much smaller for a field perpendicular to the spins. (Sharpira and Foner[58] have found this to be true in $MnF_2$.) The very large error bar for $B$ is due to its strong correlation with $A$. The most striking result is that $\phi$ is very small and completely inconsistent with the susceptibility exponent of the pure 3D $XY$ model ($\gamma=1.32$) (Ref. 59) which is expected to govern the zero-field behavior of the system. A possible explanation is that the zero-field transition is not simply $XY$-like. According to Mukamel and Grinstein,[8] the off-diagonal exchange, along with the hexagonal anisotropy, can cause the transition to become first order. If this is true, one would expect the transition temperature to decrease analytically in the presence of random fields, i.e.,

$$\Delta T_N \equiv T_N(H)-T_N(0)=-AH^2-CH^4-DH^6-\cdots. \tag{3}$$

We find that fitting the data up to $H^4$ (with $H$ in units of kOe) gives

$$T_N(0)=19.78(1)\ \text{K},\quad A=6.7(9)\times10^{-4},$$
$$C=7.8(3)\times10^{-6}.$$

$\sigma$ for the fit is 12.4 mK, not much worse than the fit to Eq. (2) with four parameters. If the $H^6$ term is included, $\sigma$ reduces to 10.1 mK and the fitted line is indistinguishable from that in Fig. 13. The parameters obtained are

$$T_N(0)=19.79(1)\ \text{K},\quad A=1.1(2)\times10^{-3},$$
$$C=4.6(1.2)\times10^{-6},\quad D=6.1(2.2)\times10^{-9}.$$

In both of these fits, the value of $A$ is comparable to that obtained by using Eq. (2), but the higher-order terms are unusually large. For example, in a field as small as 10 kOe, the $H^4$ and $H^6$ contributions are as large as the $H^2$ contribution. Compared to the similar experiment on pure $MnF_2$,[58] the $H^4$ term there is an order of magnitude smaller than the $H^2$ term even at 100 kOe. Therefore, our results cannot be explained as a normal mean-field effect. As mentioned in Sec. I, the applied field induces both random fields and three-fold Potts anisotropy in the plane. We discuss the possible effects in the following.

We note that Goldschmidt and Xu have calculated the phase boundary for the Potts model in random fields.[37] Their unpublished numerical data show a convex phase boundary in an $H^2$ versus $T$ plot, in qualitative agreement with Fig. 13. However, the high-order terms there are quite small. Specifically, for the first 4% reduction of $T_N$, 90% of the effect comes from the $H^2$ term. In Fig. 13, we estimate that only half of the first 4% change comes from the $H^2$ term. This discrepancy is not surprising, because $Fe_{0.396}Co_{0.604}Cl_2$ is not a Potts system in zero field. Both the Potts anisotropy and the random fields are turned on by the applied field, i.e., there are two simultaneous crossovers. Hence, it is unclear how the data should be compared to the theory. If we assume that the zero-field transition is $XY$-like, the crossover to three-state Potts behavior is governed by an exponent $\psi_3$ which, according to Wallace,[71] is 0.46 in 3D, to order $\epsilon^2$. This

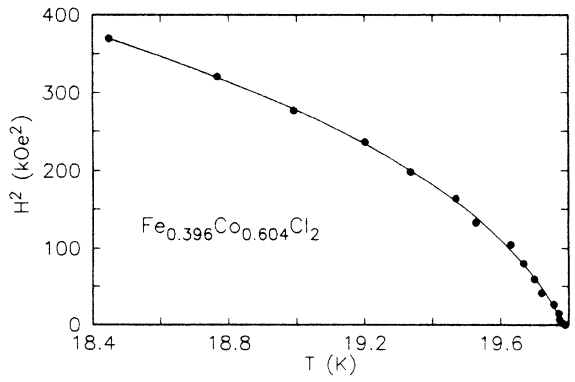


FIG. 13. The phase boundary of $Fe_{0.396}Co_{0.604}Cl_2$ shows a large convex curvature, in contrast with Figs. 8 and 10. This behavior *may be* related to a three-state Potts model in random fields. The line represents both the fits to Eqs. (2) and (3). The two fits are indistinguishable (see Sec. VI for a discussion).

should give rise to a reduction in $T_N$ proportional to $H^{1/\psi_3}$. Since $1/\psi_3 \approx 2$, this effect is difficult to identify. Furthermore, we note that there is another hexagonal symmetry $XY$ antiferromagnet that shows a similar phase boundary in axial field: Shapira *et al.* have studied $CsMnF_3$ and found large corrections to the $H^2$ term, even for the first 1% change of $T_N$.[72] These authors attributed the convexity of the phase boundary to the existence of a virtual bicritical point. The crystal symmetry of that system also allows the $M_{||}S_1^3$ term and the results may be affected by an $XY$ to Potts crossover. In comparison with $Fe_{0.396}Co_{0.604}Cl_2$, we note that there was no change in the shape of the specific-heat peak in $CsMnF_3$. This may be due to the fact that it is a pure system without random fields, but we believe a more detailed theoretical consideration is needed to fully understand the problem.

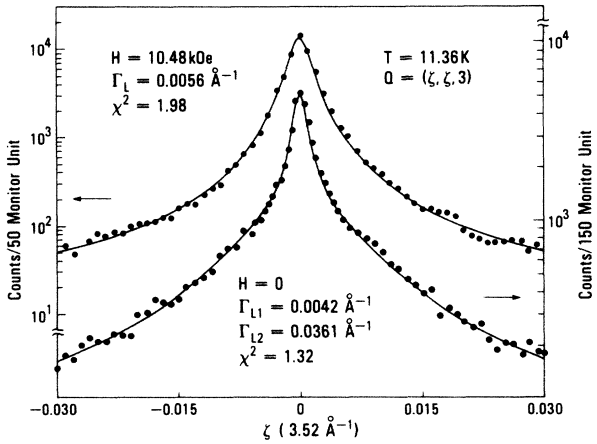


FIG. 14. Transverse scans of the (0,0,3) magnetic peak in $Fe_{0.725}Co_{0.275}Cl_2$. See Sec. VII A for details.

## VII. DISCUSSIONS AND CONCLUSIONS

### A. Competing-anisotropy systems

We have summarized in Sec. III most of the recent experiments on competing-anisotropy systems. We have particularly emphasized that there are unexplained differences between Ising-Ising and Ising-$XY$ systems. The issues center around the seemingly random absence or presence of the lower transitions among different systems, and how the spin correlations evolve below the upper transition. It is quite likely that the random off-diagonal terms play an important role in the physics, but their effects are not fully understood. Mukamel and Grinstein[8] and Oku and Igarashi[10] have analyzed the problem by the renormalization-group method and found no stable fixed points. Hence, little can be said about the nature of the mixed phase and the lower transitions. What is clear, however, is that the decoupled tetracritical behavior predicted by FA (Ref. 6) rarely occurs. In part, this is due to the fact that FA did not include crystal-symmetry-related terms in the free energy, which can be important in real systems. One example is the $wS_{||}S_1^3$ term pointed out by Mukamel[35] for $Fe_{1-x}Co_xCl_2$, but this cannot account for the similarities between tetragonal and rhombohedral systems. It is interesting to note that Inawashiro *et al.* have performed a Monte Carlo simulation of an Ising-Ising system in 2D without off-diagonal terms and they found behavior in excellent agreement with FA's prediction.[53] Hence, we believe a similar study of an Ising-$XY$ system, with and without the off-diagonal terms, will be most illuminating.

In paper 1, we have interpreted some of the features associated with the lower transition as random-field effects. The present study shows that this is not entirely satisfactory. Most notably, the lower transitions do not behave in any way like the random-field transitions, i.e., the upper transitions in applied fields. The data in Figs. 3 and 4 show no anomaly at all at the lower transition. Paper 1 also show very long $\mathbf{S}_\perp$ correlations in Fe-rich samples immediately below the upper transition, which is very difficult to understand in terms of random fields. Whether these correlations correspond to long-range order is unclear. The data presented in paper 1 show that the superlattice peak is only slightly broader than the instrumental resolution [$\sim$0.0055 Å$^{-1}$ half-width at half maximum (HWHM)] above $T_L$. Since then, we have made some new measurements in $Fe_{0.725}Co_{0.275}Cl_2$ with a better resolution ($\sim$0.0042 Å$^{-1}$ HWHM). We find that by fitting the data to a composite peak shape, a resolution-limited component is obtained.[73] This result is depicted in Fig. 14, which shows scans of the (0,0,3) superlattice peak at 11.36 K, well above $T_L$ ($\approx$8 K). The scattering at this peak is simply proportional to the $\langle \mathbf{S}_\perp \cdot \mathbf{S}_\perp \rangle$ correlation. Because the sample mosaic is intrinsically Lorentzian (i.e., a Bragg peak would have a Lorentzian shape), the zero-field data are fitted to a sum of two Lorentzians. We observe that there is a resolution-limited component ($\Gamma_{L1}$=0.0042 Å$^{-1}$) superimposed on a broad diffuse component ($\Gamma_{L2}$=0.0361 Å$^{-1}$). The field dependence of the widths and amplitudes of these two components are shown in Fig. 15. With increasing field, $\Gamma_{L1}$ remains resolution limited while $\Gamma_{L2}$ narrows. The two merge together at the spin-flop phase boundary shown in Fig. 6. In higher fields, the system is in the disordered spin-flop phase and the data can be fitted by a single Lorentzian with a width wider than resolution. This is illustrated by the 10.48-kOe data in Fig. 14. These results suggest that the zero-field state of $Fe_{0.725}Co_{0.275}Cl_2$ has moments tilted very slightly from the $c$ axis and $\langle \mathbf{S}_\perp \rangle$ has long-range order. When the system approaches $T_N$ from below, the tilt angle approaches zero. We note that an independent study by Ito *et al.*[11] on a similar system, $Fe_{1-x}Co_xTiO_3$, has also found an apparently resolution-limited peak well above $T_L$. The width is extremely small (0.0013 Å$^{-1}$ HWHM) and it is temperature independent, consistent with long-range order. This would explain the absence of specific-heat anomalies at $T_L$. However, this explanation implies that both $\mathbf{S}_\perp$ and $S_{||}$ are ordered at the upper transition. This is inconsistent with the fact that critical fluctuations due to $\mathbf{S}_\perp$ have not been observed at the upper transition.

Finally, we mention two interesting recent studies on competing-anisotropy systems. First, Nitsche and

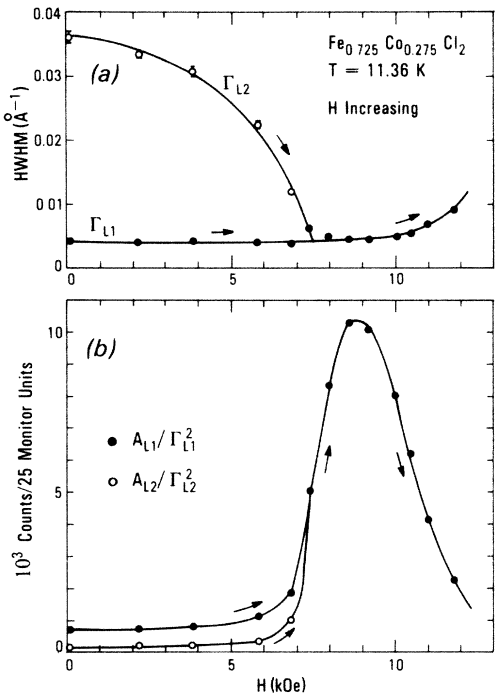


FIG. 15. Field dependence of the fitted widths and amplitudes of the $(\zeta,\zeta,3)$ scans shown in Fig. 14.

Kleemann[48] have studied $S_{\|}$-$S_{\perp}$ correlations in $Fe_{1-x}Co_xCl_2$ by birefringence technique. They found finite off-diagonal $\langle S_{\|}S_{\perp}\rangle$ correlations below the upper transition. This indicates that the off-diagonal terms are indeed relevant in competing-anisotropy systems. Their results are also consistent with the idea that both components order at $T_N$. In the second study, Endoh *et al.*[74] have found evidence of a Lorentzian-squared peak shape in $Fe_{1-x}Co_xTiO_3$. They also observed different in-plane and out-of-plane correlations. These results suggest that more high-resolution experiments are needed to understand the competing-anisotropy systems.

### B. Random-field Ising systems

#### 1. *Metastability of the zero-field-cooled state*

The understanding of the RFIM problem has advanced considerably in the last few years. Earlier controversies related to the lower critical dimension ($d_c$) are largely resolved. Theoretically, it has been shown that the roughening of domain walls does not affect the original Imry and Ma prediction of $d_c=2$ and there is now even a rigorous mathematical proof of this result.[26–28] Experimentally, the observation of a domain state in the field-cooled experiment is also understood to be due to nonequilibrium effects.[29,30,39,63] The present work, as well as previous neutron scattering studies,[29] suggests that such effects are more severe in systems with nonmagnetic impurities. There had been some doubts that such dilute systems might have a different behavior, even in equilibrium, due to percolation effects.[29,75] Specifically, if one considers a cluster with $N_1$ spins up and $N_2$ spins down, and if there are $Q$ bonds linking the cluster to the infinite one, then this cluster will have to flip when the field $H$ satisfies $H>QJ/n$, where $n=|N_1-N_2|$. The flipping of such clusters may break up the infinite cluster and change $d_c$. Quite recently, Kim and Harris[76] have pointed out that this "cluster-flip" mechanism is ineffective in the weak-field limit, because the probability of finding such a cluster is exponentially small:

$$P_n \propto \exp[-kn^{1-d^{-1}}] \propto \exp[-k(QJ/H)^{1-d^{-1}}] , \quad (4)$$

where $k$ is a constant. Thus $d_c$ for dilute systems should be unchanged. These authors also argued that, in finite fields, the zero-field-cooled states are *not* in equilibrium because, for any nonzero value of $P_n$, some clusters have to be flipped and this is expected to be an extremely slow process. Presumably, this is why a time dependence has not been observed for the zero-field-cooled state,[29] except for temperatures within 1% of the transition.[38] It is important to remember that the energy difference between the "frozen" zero-field-cooled state and the true equilibrium state should be negligible, not only because $P_n$ is vanishingly small, but because the two configurations of the clusters are nearly degenerate. For the same reason, the field-cooled and field-warmed states should have very little difference in energy (and hence specific heat) at any temperature. This is what the data in Figs. 5 and 9 show.

#### 2. *Validity of indirect specific-heat measurements*

In many ways, the specific-heat studies of different RFIM systems show a remarkable consistency. For example, a crossover exponent $\phi \approx \gamma_{\text{pure}} \approx \gamma_{\text{random}}$ is found in all the systems.[30,32,62] The change in the shape of the specific-heat peak with increasing field is seen in at least three different systems, $Fe_{1-x}Mg_xCl_2$, $Fe_{1-x}Co_xCl_2$, and $Mn_{1-x}Zn_xF_2$.[30,38] As we have discussed in Sec. V, this can be explained by the crossover effect depicted in Fig. 10. However, the critical behavior of the transition is still an issue being debated. Our direct specific-heat data in Figs. 5 and 9 suggest a large and negative $\alpha$, but the nonequilibrium nature of the system and the slight inhomogeneities of the samples preclude a precise analysis. The specific-heat data of Ikeda and Kukuta on $Mn_{0.45}Zn_{0.55}F_2$ also show the same behavior.[38] On the other hand, several *indirect* specific-heat studies have shown quite different behavior. Most notably, Belanger *et al.* have studied the $Fe_{1-x}Zn_xF_2$ system by birefringence ($\Delta n$) measurements and they found that the $d(\Delta n)/dT$ peak becomes sharper in finite field than in zero field.[62] Shapira *et al.*[30] have studied $Mn_{0.75}Zn_{0.25}F_2$ by thermal expansion ($dl/dT$). They found a sharp peak similar to the $d(\Delta n)/dT$ data in low fields, but it disappears in high fields. More recently, King *et al.*[77] have studied $Fe_{1-x}Zn_xF_2$ by a capacitance technique, extending the measurements to higher fields and more dilute samples. These data are related to both $dl/dT$ and $d\epsilon/dT$, where $\epsilon$ is the dielectric constant. The results are very similar to the $d(\Delta n)/dT$ data, showing a sharp and symmetric peak, consistent with $\alpha=0$. The apparent discrepancies among these several studies have

been puzzling for some time. In the following, we suggest two possible explanations.

First, we consider the Ising Hamiltonian for an antiferromagnet:

$$\mathcal{H}=\sum_{i,j} J_{ij}S_iS_j+H\sum_i S_i \ , \tag{5}$$

where $J_{ij}=J>0$ for nearest-neighbor pairs of magnetic ions and $J_{ij}=0$ otherwise. Taking thermal average of Eq. (5) shows that the total magnetic energy ($E_{\rm tot}=\langle\mathcal{H}\rangle$) of the system consists of two parts, the exchange energy

$$E_J\propto J\langle S_0S_1\rangle$$

and the field energy

$$E_H=H\sum_i\langle S_i\rangle=-HM(T,H)=-H\int_0^H\chi_0(T,h)dh \ . \tag{6}$$

Here, $\langle S_0S_1\rangle$ denotes the nearest-neighbor spin-spin correlation, $M(T,H)$ is the uniform magnetization, and $\chi_0(T,H)$ is the uniform susceptibility. For a simple two-sublattice antiferromagnet, such as $FeF_2$ and $MnF_2$, Fisher[78] has shown that $\langle S_0S_1\rangle\approx G_0-1$ near a phase transition, where $G_0\equiv N^{-1}\sum_{i,j}\langle S_iS_j\rangle$ is the zero wave-vector ($q=0$) spin-spin correlation function and $N$ is the number of spins. In zero field, $E_{\rm tot}=E_J\propto G_0-1$ and, hence, the specific-heat singularity is given by

$$C_{\rm sing}\propto\frac{dG_0}{dT} \ . \tag{7}$$

The indirect specific-heat measurements on antiferromagnets are essentially all based on this principle. The use of $d(T\chi_0)/dT$ was first pointed out by Fisher.[78] The use of $d(\Delta n)/dT$ and $d\epsilon/dT$ was suggested by Gehring.[79] The proportionality between $C_{\rm sing}$ and $dl/dT$ is more general, not limited to antiferromagnets. It comes from magnetoelastic couplings (i.e., the competition between exchange energy and elastic energy).[80] The validity of these techniques is well established for pure systems. It is easy to show that these techniques are also valid for random systems in zero field. One can simply follow Fisher's original derivation[78] and add configuration averages (denoted by $[\ ]_c$) for the randomness. This gives

$$E_J=fJ[\langle S_0S_1\rangle]_c \ ,$$

where $f$ is a constant, and

$$G_0=N^{-1}\sum_{i,j}[\langle S_iS_j\rangle]_c=[\langle S_0S_1\rangle]_c+1 \ . \tag{8}$$

Hence, Eq. (7) is unchanged. On the other hand, applying the indirect techniques in finite fields is questionable. This is related to the field energy term in Eq. (6). Since

$$k_BT\chi_0=G_0-G_0^{\rm dis} \tag{9}$$

(where $G_0^{\rm dis}\equiv N^{-1}\sum_{i,j}[\langle S_i\rangle\langle S_j\rangle]_c$ is the disconnected part of the correlation function), we can write

$$E_{\rm tot}=E_J+E_H=fJ[G_0(T,H)-1]-\frac{H}{k_BT}\int_0^H[G_0(T,h)-G_0^{\rm dis}(T,h)]dh \ . \tag{10}$$

The presence of the field energy integral has nontrivial consequences. Fisher has argued that even for pure systems, $G_0$ should contain a term directly proportional to the specific heat.[78] This would invalidate the indirect specific-heat techniques. For random systems, the situation is even more complicated because $G_0^{\rm dis}$ is nonzero in zero field. FA have suggested that it has a $t^{2\beta}$ singularity.[31] This implies that the $G_0$ terms in Eq. (10) contain a similar singularity to cancel with $G_0^{\rm dis}$, so that $E_{\rm tot}$ has only a $t^{1-\alpha}$ singularity. How this occurs is unclear. At present, we can only note that all the indirect measurements are related to $dG_0/dT$ and Eq. (10) suggests that they acquire a new singularity in finite field. This would explain why a new sharp peak arises with increasing field in the indirect specific-heat experiments, but not in the true specific-heat, but we emphasize that a detailed analysis of Eq. (10) is needed before we can reach a firm conclusion. [Strictly speaking, Eq. (10) is not entirely correct because we have used Eq. (8), which is valid in zero field, for the first term in Eq. (10), which is intended for finite fields. This neglects a term of order $M^2$, where $M=\int_0^H\chi_0dh$. The inclusion of this higher-order term will further complicate the analysis.]

Next we consider nonequilibrium problems associated with the indirect techniques. Because these are all related to the spin-spin correlation function $G_0$, they should be quite sensitive to the spin configuration. A large number of neutron scattering studies have shown that there are strong hysteresis effects in the spin configuration.[29,33,63,81] The largest difference is that between the field-cooled and zero-field-cooled states. The former is a domain state and the latter has long-range order. As a result, one should expect strong hysteresis in the indirect specific-heat measurements. This is indeed the case for the birefringence,[62] thermal-expansion,[30] susceptibility,[38] and dielectric[77] experiments. The sharpest peak is always observed in the zero-field-cooled experiments and the most rounded peak in the field-cooled experiments.[30] As we have argued above in Sec. VII B 1, the zero-field-cooled state can only reach equilibrium by flipping clusters of spins, which may be possible only very close to the transition. This implies an abrupt change in $G_0$ at the transition and a very sharp peak in $dG_0/dT$. Indeed, various neutron experiments have found an abrupt change of $G(q)$ for wave vector $q$ near the antiferromagnetic peak, and the indirect specific-heat measurements have shown sharp peaks in $dG(q)/dT$ at $q=0$. We believe these should not be used to determine whether the transition is first order or second order.[81,82] In addition, we also note that there is a difference in the sharpness of the peak among different systems, e.g., a sharp peak is seen in high fields for the zero-field-cooled states of $Fe_{1-x}Zn_xF_2$,[77] but not for $Mn_{0.75}Zn_{0.25}F_2$.[30] This may be related to the fact that the Fe system has a much stronger anisotropy and is more difficult to equilibrate. (This could also be due to the fact that the thermal-expansion measurements probe the spin

configuration through the magneto-elastic strain, which is equilibrated by the phonons.)

### 3. *Critical exponents and Fisher renormalization*

In spite of the ambiguities associated with the indirect specific-heat techniques and the nonequilibrium phenomena there continues to be arguments about the equilibrium critical behavior of RFIM systems. Belanger *et al.* have reported 2D Ising exponents for 3D RFIM systems, and they suggested a rigorous dimension reduction of $d \rightarrow d' = d - 1$.[62,63] There are several problems with their interpretation of the data. First, their claim of $\eta = \frac{1}{4}$ violates the Schwartz inequality, which requires $\eta \geq \frac{1}{2}$ in 3D.[41] Second, their value for $\nu$ $(=1)$ in low field seems to increase in higher fields $(\nu \approx 1.5)$.[81] Third, their value for $\alpha$ $(=0)$ is based on the birefringence data in the hysteretic region[62] which, from the preceding discussion, probably does not measure the true specific heat. Here, we offer an alternative viewpoint of the problem.

We note that direct specific-heat measurements are least sensitive to nonequilibrium effects. Based on the original Imry and Ma argument,[26] we know that the physics of the random-field system is in the competition between the exchange energy $E_J$ and the field energy $E_H$. Metastability arises because there are many different states with nearly the same $E_{\rm tot}$, but have very different spin configurations.[39,40] As a result, a true energy or specific-heat measurement should show very little history dependence. This is indeed what our field-cooled and field-warmed data in Figs. 5 and 9 show. Recent Monte Carlo simulation studies have also found similar results.[40] Had we performed zero-field-cooled experiments, perhaps a larger difference could be observed. However, since the zero-field-cooled state is not in equilibrium, this would still not provide a definitive answer for $\alpha$. Nevertheless, some useful information can be gained from the existing data. We note that the only true specific-heat data currently available are those in Figs. 5 and 9, and those obtained by Ikeda and Kikuta on $Mn_{0.45}Zn_{0.55}F_2$.[38] These three sets of results on three different systems are entirely consistent. They all show a peak shape that changes continuously with increasing field and becomes a broad cusp in sufficiently large fields. This essential feature has been reproduced by computer simulation in at least four independent studies.[40] There are no signs for any latent heat to support the idea of a first-order transition (although one cannot rule out the possibility of a very weak one).[81,82] One possible explanation is that this result is simply due to crossover[32] or nonequilibrium effects.[39] Otherwise, the broadening of the cusp with increasing field would imply that the value of $\alpha$ is more negative in finite field than in zero field. Recent calculations by Schwartz and Soffer have actually reached such a conclusion.[41] On the other hand, we note that there are also calculations which found large and positive values for $\alpha$.[42] Specifically, a scaling theory by Bray and Moore[42] has found that in Grinstein's modified hyperscaling relation[23] $2-\alpha=(d-y)\nu$, the exponents are given by $y=1+\varepsilon/2$ and $\nu=1/\varepsilon$, where $\varepsilon=d-2$. For 3D systems, these give $\nu=1$, $y=\frac{3}{2}$, and $\alpha=\frac{1}{2}$. An independent domain-wall renormalization-group calculation by Cheung[42] has also found $y=1.5\pm0.2$, $\nu=1.15\pm0.10$, and $\alpha=0.35\pm0.03$, in approximate agreement with Bray and Moore. At a first glance, these values for $\alpha$ contradict the experimental results. This is not true, however, if one takes into account the effect of Fisher renormalization.[84] The reason is that the theoretical model has constant random fields, whereas the experiments were done in constant applied fields. The latter lead to the temperature-dependent random fields depicted in Fig. 10. This acts as a constraint on the system, which can renormalize a positive $\alpha$ to an effective value $\alpha_{\rm eff}=-\alpha/(1-\alpha)$. Thus, for $\alpha \approx \frac{1}{2}$ in the theory, one can observe $\alpha_{\rm eff} \approx -1$ in the experiment. The renormalization effect is usually difficult to observe because the crossover exponent is $\alpha$, which is either small or negative in most circumstances. However, if $\alpha$ is as large as $\frac{1}{2}$, this could explain the data in Figs. 5 and 9. Given the many uncertainties in the experiments (sample inhomogeneity, hysteresis, crossover effects, etc.), we must emphasize that this is only a possibility and we cannot quote a value for $\alpha$ with any reliable precision. The situation of the theories is somewhat similar. For example, Schwartz and Soffer[41] have found $y=1.43$ in 3D, consistent with the results of Bray and Moore and Cheung. By assuming a dimension reduction of $d \rightarrow d' \equiv d-y$, these authors obtained $\nu \approx 1.4$ and $\alpha \approx -0.2$. Thus it appears that the small variations of $y$ and $\nu$ among the different calculations are giving a large uncertainty in $\alpha$, to the extent that there is no consensus as to whether $\alpha$ is positive or negative. Therefore, we believe the correct interpretation of the experiments is still an open issue. However, since all calculations give $y \approx \frac{3}{2}$, the possibility of 2D Ising critical behavior is highly unlikely. We should also remark that, if $\alpha$ is large and positive, Fisher renormalization can alter the other exponents, e.g., $\nu \rightarrow \nu_{\rm eff}=\nu/(1-\alpha)$ and $\gamma \rightarrow \gamma_{\rm eff}=\gamma/(1-\alpha)$. These might explain why $\nu$ and $\gamma$ in several experiments appear to increase rapidly with increasing field.[63,81,83]

In Secs. IV and VI, we have suggested that the high-field behavior of $Fe_{0.714}Co_{0.286}Cl_2$ and $Fe_{0.396}Co_{0.604}Cl_2$ be described by the three-state Potts model in random fields. That the peak shape in $Fe_{0.396}Co_{0.604}Cl_2$ changes with increasing field (Fig. 12) is consistent with the random-field crossover effect illustrated in Fig. 10. The cusplike anomalies of the SF-PM transitions in $Fe_{0.714}Co_{0.286}Cl_2$ (Fig. 5) also appear to be an extension of the same behavior in higher random fields. These results support the idea that there are random off-diagonal terms resulting in random-field effects, but a detailed understanding of these effects is still lacking. In particular, the random-field Potts transition is expected to be first order in 3D[36,37] and this has not been observed. This is an interesting problem for further studies, both theoretically and experimentally.

### Notes added

(1) A recent paper by Aharony[85] shows that, in systems dominated by random exchange, the random-field cross-

over exponent $\phi$ is not necessarily equal to the zero-field susceptibility exponent $\gamma_{\text{random}}$. Instead, he finds an inequality $\phi \geq \gamma_{\text{random}}$. This further prevents one from making an unambiguous interpretation on the value of $\phi$.

(2) Most recently, Ikeda has refined his ac specific-heat measurements on $Mn_{0.45}Zn_{0.55}F_2$.[86] A sharp cusp was observed for $H=0$ but it becomes progressively more rounded for $H=5$, 10, and 16 kOe. Comparisons of field-cooled, field-warmed, and zero-field-cooled data show no significant difference. Above the spin-flop field, the cusp sharpens again. These results are substantially consistent with those reported here.

## ACKNOWLEDGMENTS

The specific-heat experiments described in this paper were performed during my frequent visits to the IBM Thomas J. Watson Research Center (Yorktown Heights, NY). I have enjoyed the gracious hospitality of Judy and Paul Horn, and many illuminating conversations with G. Grinstein. I am indebted to S. von Molnar, P. Dimon, and A. Torressen for their invaluable help during all phases of the work. Special thanks are due to S. von Molnar for sponsoring this work and many suggestions for the manuscript. The neutron scattering data presented in Sec. VII were obtained at the Oak Ridge National Laboratory in collaboration with J. W. Cable. I am grateful to him for many important contributions. I wish to thank Y. Y. Goldschmidt, A. Aharony, H. Ikeda, M. Schwartz, and Y. Endoh, for communicating their results prior to publication. I have benefited from discussions with many distinguished scientists besides the ones named above, especially R. J. Birgeneau, R. Caflisch, S. Galam, A. B. Harris, D. Mukamel, Y. Shapira, and H. Yoshizawa.

[1]P.-z. Wong, P. M. Horn, R. J. Birgeneau, and G. Shirane, Phys. Rev. B **27**, 428 (1983); and Phys. Rev. Lett. **45**, 1974 (1980).

[2]F. Matsubara and S. Inawashiro, J. Phys. Soc. Jpn. **42**, 1529 (1977); **46**, 1740 (1979); **47**, 1102 (1979).

[3]P.-A. Lindgard, Phys. Rev. B **16**, 2168 (1977).

[4]T. Oguchi and T. Ishikawa, J. Phys. Soc. Jpn. **45**, 1213 (1978).

[5]Y. Someya, J. Phys. Soc. Jpn. **50**, 3897 (1981).

[6]S. Fishman and A. Aharony, Phys. Rev. B **18**, 3507 (1978).

[7]R. J. Elliot and M. F. Thorpe, J. Appl. Phys. **39**, 802 (1968).

[8]D. Mukamel and G. Grinstein, Phys. Rev. B **25**, 381 (1982).

[9]A. Aharony, Solid State Commun. **28**, 667 (1978).

[10]M. Oku and H. Igarashi, Prog. Theor. Phys. **76**, 1493 (1983).

[11]A. Ito, S. Morimoto, Y. Someya, Y. Syono, and H. Takei, J. Phys. Soc. Jpn. **51**, 3173 (1982).

[12]A. Ito, T. Tamaki, Y. Someya, and H. Ikeda, Physica **120B**, 207 (1983).

[13]W. A. H. M. Vlak, E. Frikkee, A. F. M. Arts, and H. W. de Wijn, J. Phys. C **16**, L1015 (1983); Phys. Rev. B **33**, 6470 (1986); W. A. H. M. Vlak, B. J. Dikken, A. F. M. Arts, and H. W. de Wijn, Phys. Rev. B **31**, 4496 (1985).

[14]S. A. Higgins, R. A. Cowley, M. Hagen, J. K. Kjems, U. Durr, and K. Fendler, J. Phys. C **17**, 3235 (1984).

[15]K. Katsumata, J. Tuchendler, and S. Legrand, Phys. Rev. B **30**, 1377 (1984); K. Katsumata and J. Tuchendler (unpublished).

[16]M. C. K. Wiltshire and D. C. Price, Solid State Commun. **51**, 531 (1984).

[17]M. Kobayashi, K. Katsumata, T. Sato, and Y. Miyako, J. Phys. Soc. Jpn. **46**, 1467 (1979); Phys. Rev. B **19**, 2700 (1979).

[18]K. Katsumata, M. Kobayashi, and H. Yoshizawa, Phys. Rev. Lett. **43**, 960 (1979).

[19]A. Ito, Y. Someya, and K. Katsumata, Solid State Commun. **36**, 681 (1980); J. Phys. Soc. Jpn. **52**, 254 (1983).

[20]K. Katsumata, H. Yoshizawa, G. Shirane, and R. J. Birgeneau, Phys. Rev. B **31**, 316 (1985).

[21]K. A. Hay and J. B. Torrance, Jr., Phys. Rev. B **2**, 746 (1970).

[22]K. Katsumata, C. Uyeda, M. Motokawa, and M. Date, J. Phys. Soc. Jpn. **54**, 1244 (1985).

[23]G. Grinstein, Phys. Rev. Lett. **37**, 944 (1976); A. Aharony, Y. Imry, and S.-k. Ma, Phys. Rev. Lett. 37, 1367 (1976); A. P. Young, J. Phys. C **10**, L257 (1977); G. Parisi and N. Sourlas, Phys. Rev. Lett. **43**, 744 (1979).

[24]E. Pytte, Y. Imry, and D. Mukamel, Phys. Rev. Lett. **46**, 1173 (1981); K. Binder, Y. Imry, and E. Pytte, Phys. Rev. B **24**, 6736 (1981); D. Mukamel and E. Pytte, *ibid.* **25**, 4779 (1982).

[25]H. S. Kogon and D. J. Wallace, J. Phys. A **14**, L527 (1981).

[26]Y. Imry and S.-k. Ma, Phys. Rev. Lett. **35**, 1399 (1975); G. Grinstein and S.-k. Ma, Phys. Rev. B **28**, 2588 (1983).

[27]A. Aharony and E. Pytte, Phys. Rev. B **27**, 5872 (1983); J. F. Fernandez, G. Grinstein, Y. Imry, and S. Kirkpatrick, Phys. Rev. Lett. **51**, 203 (1983); D. Andelman, H. Orland, and L. C. R. Wijewardhana, *ibid.* **52**, 145 (1984); A. N. Berker, Phys. Rev. B **29**, 5243 (1984); M. Schwartz, J. Phys. C **18**, 135 (1985); Y. Shapir, Phys. Rev. Lett. **54**, 154 (1985).

[28]J. Z. Imbrie, Phys. Rev. Lett. **53**, 1747 (1984).

[29]P.-z. Wong and J. W. Cable, Phys. Rev. B **28**, 5361 (1983); Solid State Commun. **51**, 545 (1984); P.-z. Wong, J. W. Cable, and P. Dimon, J. Appl. Phys. **55**, 2377 (1984).

[30]Y. Shapira, N. F. Oliveira, and S. Foner, Phys. Rev. B **30**, 6639 (1984); Y. Shapira and N. F. Oliveira, *ibid.* **27**, 4336 (1983); Y. Shapira, J. Appl. Phys. **53**, 1931 (1982); Y. Shapira (private communication).

[31]S. Fishman and A. Aharony, J. Phys. C **12**, L729 (1979).

[32]P.-z. Wong, S. von Molnar, and P. Dimon, J. Appl. Phys. **53**, 7954 (1982); Solid State Commun. **48**, 573 (1983).

[33]P.-z. Wong and J. W. Cable, Phys. Rev. B **30**, 485 (1984).

[34]A. N. Bazhan and V. A. Ul'yanov, Zh. Eksp. Teor. Fiz. **79**, 186 (1980) [Sov. Phys.—JETP **52**, 94 (1980)].

[35]D. Mukamel, Phys. Rev. Lett. **46**, 845 (1981).

[36]D. Blankschtein, Y. Shapir, and A. Aharony, Phys. Rev. B **29**, 1263 (1984); H. Nishimori, *ibid.* **28**, 4011 (1983).

[37]Y. Y. Goldschmidt and G. Xu, Phys. Rev. B **32**, 1876 (1985); Nucl. Phys. B **265**, 1 (1986); Y. Y. Goldschmidt (private communication).

[38]H. Ikeda and K. Kikuta, J. Phys. C **17**, 1221 (1984).

[39]The metastability of RFIM systems has been demonstrated by mean-field simulations: C. Ro, G. Grest, C. M. Soukoulis, and K. Levin, Phys. Rev. B **31**, 1682 (1985); H. Yoshizawa and D. P. Belanger, *ibid.* **30**, 5220 (1984). We note that both the field-cooled and zero-field-cooled states show very slow relaxations in the Monte Carlo study by D. Stauffer, C. Hartzstein, K. Binder, and A. Aharony, Z. Phys. B **55**, 352 (1984). The dynamics near the transition has recently been

analyzed by J. Villain, J. Phys. (Paris) **46**, 1843 (1985); D. S. Fisher, Phys. Rev. Lett. **56**, 416 (1986). See also references therein.

[40]Broadening of the specific-heat cusp in finite field has recently been observed by Monte Carlo simulations: G. S. Grest, C. M. Soukoulis, and K. Levin, Phys. Rev. B **33**, 7659 (1986); A. T. Ogielski and D. A. Huse, Phys. Rev. Lett. **56**, 1298 (1986); and private communication. Grest *et al.* also found very small hysteresis in the specific heat. Similar, but less complete results had previously been obtained by Andelman *et al.* in Ref. 27, and by D. P. Landau, H. H. Lee, and W. Kao, J. Appl. Phys. **49**, 1356 (1978).

[41]M. Schwartz and A. Soffer, Phys. Rev. Lett. **55**, 2499 (1985); Phys. Rev. B **33**, 2059 (1986).

[42]A. J. Bray and M. A. Moore, J. Phys. C **18**, L927 (1985); H.-F. Cheung, Phys. Rev. B **33**, 6191 (1986).

[43]P.-z. Wong, J. Cryst. Growth **58**, 609 (1982).

[44]E. M. Forgan and S. Nedjat, Rev. Sci. Instrum. **51**, 411 (1980).

[45]M. Wun and N. E. Phillips, Cryogenics **15**, 36 (1975).

[46]E. F. Westrum, Jr., C. Chow, D. W. Osborne, and H. E. Flotow, Cryogenics **7**, 43 (1967).

[47]A. C. Anderson and R. B. Rauch, Rev. Sci. Instrum. **41**, 649 (1970).

[48]W. Nitsche and W. Kleemann, Proceedings of the International Conference on Magnetism, 1985 [J. Magn. Magn. Mater. **54-57**, 37 (1986)].

[49]R. J. Birgeneau, L. W. Rupp, Jr., H. J. Guggenheim, P. A. Lindgard, and D. L. Huber, Phys. Rev. Lett. **30**, 1252 (1973); P. A. Lindgard, R. J. Birgeneau, J. Als-Nielsen, and H. J. Guggenheim, J. Phys. C **8**, 1059 (1975).

[50]L. Bevaart, E. Frikkee, J. V. Lebesque, and L. J. de Jongh, Phys. Rev. B **18**, 3376 (1978); L. Bevaart, E. Frikkee, and L. J. de Jongh, *ibid.* **19**, 4741 (1979).

[51]L. J. de Jongh and A. R. Miedema, Adv. Phys. **23**, 1 (1974).

[52]K. Takeda, M. Matsuura, and T. Haseda, J. Phys. Soc. Jpn. **29**, 885 (1970).

[53]S. Inawashiro, T. Togawa, and R. Kurasaka, J. Phys. C. **12**, 2351 (1979).

[54]See, e.g, H. Rohrer, Phys. Rev. Lett. **34**, 26 (1975); Y. Shapira and C. C. Becerra, Phys. Lett. **57A**, 483 (1976). The effects of sample misalignment are discussed in K. W. Blazey, H. Rohrer, and R. Webster, Phys. Rev. B **4**, 2287 (1971). See also, S. Galam and A. Aharony, J. Phys. C **14**, 3603 (1981).

[55]H.-T. Shang and M. B. Salamon, Phys. Rev. B **22**, 4401 (1980); H.-T. Shang, C.-C. Huang, and M. B. Salamon, J. Appl. Phys. **49**, 1366 (1978); J. F. Dillon, Jr., E. Yi Chen, and H. J. Guggenheim, Phys. Rev. B **18**, 377 (1978).

[56]Y. Imry and M. Wortis, Phys. Rev. B **19**, 3580 (1979).

[57]J. Gelard, F. Bensamka, D. Bertrand, A. R. Fert, J. P. Redoules, and S. Legrand, J. Phys. C **16**, L939 (1983).

[58]Y. Shapira and S. Foner, Phys. Rev. B **1**, 3083 (1970).

[59]L. C. Le Guillou and J. Zinn-Justin, Phys. Rev. B **21**, 3976 (1980).

[60]G. Jug, Phys. Rev. B **27**, 609 (1983); see also K. E. Newman and E. K. Riedel, *ibid.* **25**, 264 (1982).

[61]W. B. Yelon and R. J. Birgeneau, Phys. Rev. B **5**, 2615 (1972).

[62]D. P. Belanger, A. R. King, V. Jaccarino, and J. L. Cardy, Phys. Rev. B **28**, 2522 (1983); Phys. Rev. Lett. **48**, 1050 (1982). We note that hystereses in the birefringence data have recently been reported in A. R. King and D. P. Belanger, J. Magn. Magn. Mater. **54-57**, 19 (1986).

[63]R. A. Cowley, H. Yoshizawa, G. Shirane, and R. J. Birgeneau, Z. Phys. B **58**, 15 (1984); Phys. Rev. B **31**, 4548 (1985); R. J. Birgeneau, Y. Shapira, G. Shirane, R. A. Cowley, and H. Yoshizawa, Physica **37B&C**, 83 (1986).

[64]M. Chirwa, L. Lundgran, P. Nordblad, and O. Beckman, J. Magn. Magn. Mater. **15-18**, 457 (1980); D. P. Belanger, P. Nordblad, A. R. King, V. Jaccarino, L. Lundgren, and O. Beckman, *ibid.* **31-34**, 1095 (1983).

[65]M. C. Lanusse, P. Carrara, A. R. Fert, G. Mischler, and J. P. Redoules, J. Phys. (Paris) **33**, 429 (1972).

[66]M. T. Hutchings, M. P. Schulhof, and H. J. Guggenheim, Phys. Rev. B **5**, 154 (1972).

[67]R. J. Birgeneau, R. A. Cowley, G. Shirane, H. Yoshizawa, D. P. Belanger, A. R. King, and V. Jaccarino, Phys. Rev. B **27**, 6747 (1983).

[68]M. P. Schulhof, R. Nathans, P. Heller, and A. Linz, Phys. Rev. B **4**, 2254 (1971).

[69]M. Kaufman and M. Kardar, Phys. Rev. B **31**, 2913 (1985).

[70]D. Moses, J. E. Kardontchik, R. Brener, and H. Shechter, J. Phys. C **13**, 3903 (1980).

[71]D. J. Wallace, in *Phase Transition and Critical Phenomena,* edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. 6, p. 351.

[72]Y. Shapira, N. F. Oliveira, Jr., and T. S. Chang, Phys. Rev. B **21**, 1271 (1980); J. Appl. Phys. **50**, 1790 (1979). The crystal symmetry of $CsMnF_3$ is described in E. A. Turov, *Physical Properties of Magnetically Ordered Crystals* (Academic, New York, 1965). I thank Y. Shapira for bringing my attention to this work.

[73]P.-z. Wong and J. W. Cable (unpublished).

[74]Y. Endoh, I. Hirosawa, J. Newsam, and Y. Noda, J. Phys. Soc. Jpn. (to be published); see also J. Phys. C **19**, 1273 (1986).

[75]M. Fähnle, Phys. Rev. B **27**, 5821 (1983).

[76]Y. Kim and A. B. Harris, Phys. Rev. B **32**, 4676 (1985).

[77]A. R. King, V. Jaccarino, D. P. Belanger, and S. M. Rezende, Phys. Rev. B **32**, 503 (1985).

[78]M. E. Fisher, Philos. Mag. **7**, 1731 (1962).

[79]G. A. Gehring, J. Phys. C **10**, 531 (1977).

[80]For a change of $\delta l$ in sample length, the elastic energy is proportional to $\delta l^2$ and the change in exchange energy is proportional to $\delta l (dJ/dl) \sum_{i,j} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$. Minimizing the sum of the two energies gives $\delta l \propto E_J \propto G_0$. (I thank Y. Shapira for pointing this out.) The extension of this argument to dilute systems is unclear, because the strain is nonuniform and it is correlated with the spin configuration, i.e., the change in $E_J$ is given by $\sum_{i,j} (dJ_{ij}/dl_{ij}) \langle \delta l_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \rangle$, with the constraint that $\sum_i \delta l_{i,i+1} = \delta l$ along a straight line across the sample.

[81]R. J. Birgeneau, R. A. Cowley, G. Shirane, and H. Yoshizawa, Phys. Rev. Lett. **54**, 2147 (1985). See also, R. A. Cowley, H. Yoshizawa, G. Shirane, and R. J. Birgeneau, Phys. Rev. B **30**, 6650 (1984).

[82]A. P. Young and M. Nauenberg, Phys. Rev. Lett. **54**, 2429 (1985); S. R. Anderson and G. F. Mazenko, Phys. Rev. B **33**, 2007 (1986).

[83]D. P. Belanger, A. R. King, and V. Jaccarino, Phys. Rev. B **31**, 4538 (1985).

[84]M. E. Fisher, Phys. Rev. **176**, 257 (1968).

[85]A. Aharony, Phys. Rev. B (to be published).

[86]H. Ikdeda (private communication).