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Hysteretic behavior of the diluted random-field Ising system $Fe_{0.70}Mg_{0.30}Cl_2$

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We report a systematic neutron scattering study of the hysteretic behavior of the randomly diluted metamagnet $Fe_{0.70}Mg_{0.30}Cl_2$ in magnetic fields, which is presumed to correspond to the three-dimensional Ising model in random fields. In general, we find that the low-temperature state of the system is disorderd if it is approached from the disordered paramagnetic phase, but ordered if it is approached from the zero-field-ordered phase. In contrast to typical spin-glass systems, time dependence is observed for the field-cooled states. Plausible explanations for these phenomena and further experiments are suggested.

The ordering of uniaxial random antiferromagnets in uniform applied fields has attracted considerable interest in recent years. Fishman and Aharony[1] and Wong, von Molnar, and Dimon[2] have independently pointed out two different mechanisms by which the applied field generates random staggered fields in these systems. Because of this, the presence or absence of long-range order (LRO) in these systems provides a test for the lower critical dimension d_c (the dimension below which no LRO can occur) of the random-field Ising model, which is currently a rather controversial issue. Different theories predict that d_c is equal to either two or three.[3] In principle, these predictions can be tested easily by neutron scattering experiments on three-dimensional (3D) systems. The peak shapes of the magnetic reflections in such experiments give direct information on the range of order and the form of the spin-spin correlation function. To this date, however, at least four different 3D systems have been studied by neutron scattering and the answer is still unclear. In $Co_{0.3}Zn_{0.7}F_2$ (Ref. 4) and $Fe_{0.5}Zn_{0.5}F_2$,[5] there is evidence for disorder. In $Mn_{0.78}Zn_{0.22}F_2$ (Ref. 6) and $Fe_{0.725}Co_{0.275}Cl_2$,[7] there is evidence for LRO. The conflict is mainly due to the fact that there is complicated hysteretic behavior in each of these sytems, and it occurs in a *different* region of the T-H plane for a *different* system. Inside that region, whether the system is ordered or disordered depends on its history. Clearly, before a definitive conclusion for d_c can be reached, these history-dependent phenomena must be first understood.

In this Communication, we report a neutron scattering study of a fifth system, $Fe_{0.70}Mg_{0.30}Cl_2$, with emphasis on its hysteretic behavior. This system is chosen for the study because it has interesting similarities and differences as compared with the four systems mentioned above. For example, similar to $Fe_{1-x}Co_xCl_2$,[7] it is a layered metamagnet in which very large random fields can be generated by moderate applied fields.[2] On the other hand, unlike $Fe_{1-x}Co_xCl_2$, it is a *diluted* system with nonmagnetic impurity ions (Mg), which makes it similar to the three fluoride systems that had been studied.[4–6] In particular, it is similar to $Fe_{1-x}Zn_xF_2$ and $Co_{1-x}Zn_xF_2$ in having strong anisotropy, while $Mn_{1-x}Zn_xF_2$ and $Fe_{1-x}Co_xCl_2$ have weak anisotropy. As we shall show, the hysteretic behavior of this system indeed makes an interesting comparison with the other systems and leads us to suggest some plausible explanations for these behaviors.

The neutron scattering experiments were performed with the applied field (H) parallel to the spins' easy exis (the c axis of the crystal). The experimental details are identical to the $Fe_{1-x}Co_xCl_2$ experiment.[7] The magnetic reflection of interest is $(1, 0, \bar{1})$. Its peak intensity measures the square of the sublattice magnetization $M_{\parallel}^{s}$ and its width measures the range of order. The Néel temperature T_N of the sample is 13.5(1) K in zero field.

In Fig. 1(a), we show the peak intensity $I(1, 0, \bar{1})$ as a function of H for five different temperatures below T_N. The increasing field data were taken with the sample cooled in zero field. The decreasing field data were taken with the sample initially in the high-field paramagnetic phase. These two procedures will be called low-field approach (LFA) and high-field approach (HFA), respectively. We observe that at 9.24 K, there is a strong hysteresis which persists down to zero field; while at 12.98 K, there is no obvious hysteresis. Between these two temperatures, the hysteretic effect decreases with increasing T. In Fig. 1(b), we show $I(1, 0, \bar{1})$ as a function of T for $H = 4.55$ kOe. The increasing T data (solid circles) were taken with the sample zero-field cooled. The decreasing T data were taken with the sample field cooled. These will be referred to as low-temperature approach (LTA) and high-temperature approach (HTA), respectively. Here we observe that the hysteresis persists up to the approximate transition temperature 11.7 K. From Fig. 1(a), we *estimated* $I(1, 0, \bar{1})$ for LFA and HFA at 9.24, 10.01, and 11.08 K for $H = 4.55$ kOe. The results are shown in Fig. 1(b) by the solid and open triangles. We note that the LFA points fall near the LTA curve and the HFA points fall near the HTA curve. This comparison suggests that the two states which evolve from the zero-field-ordered states (LFA and LTA) are similar, and so are the two states which evolve from the paramagnetic phase (HFA and HTA).

To investigate which states are ordered or disordered, systematic peak shape measurements were made as a function of T and H using the four approaches. These measurements were similar to those performed for $Fe_{0.725}Co_{0.275}Cl_2$ previously.[7] The intensity profiles were fitted to a Gaussian plus a Lorentzian to represent the Bragg and diffuse scattering. The Gaussian width (Γ_G) reflects the range of order.

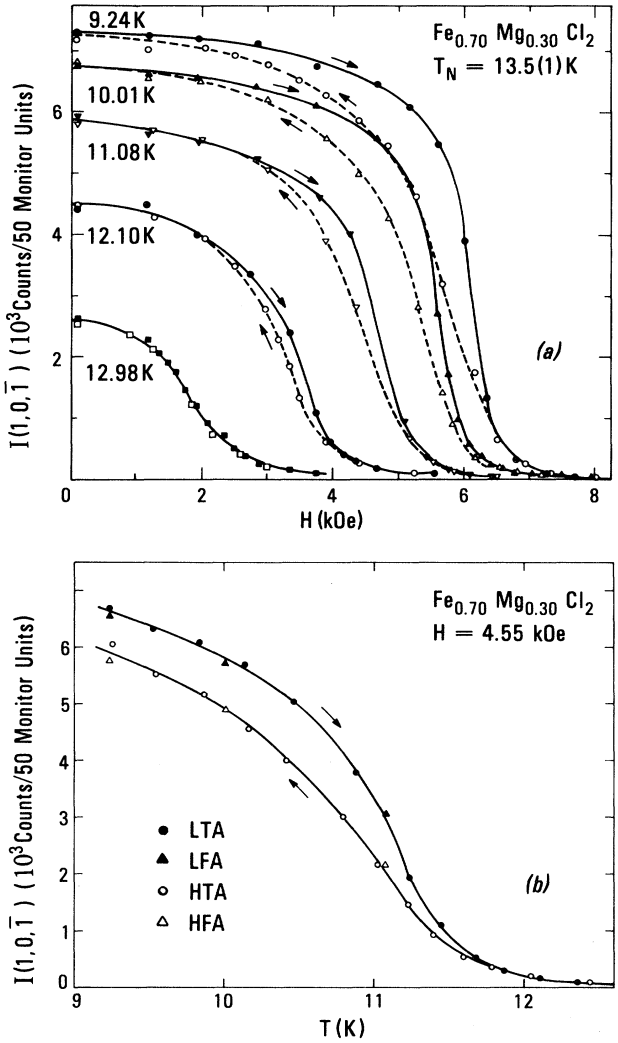

FIG. 1. (a) Field hystereses of the peak intensity $I(1, 0, \bar{1})$ for five different temperatures below T_N. (b) Comparison of the four approaches for $H = 4.55$ kOe (see text).

The resolution limit for Γ_G was 0.0045 $Å^{-1}$ (half-width at half maximum).[8] In Fig. 2, we show the Gaussian amplitude A_G and width Γ_G as a function of T for $H = 4.68$ kOe, using LTA and HTA. We observe that the hysteresis in A_G is similar to the peak intensity data in Fig. 1(b). More importantly, Γ_G is always broader than resolution for the HTA states, but resolution limited for the LTA states. In Fig. 3, we show A_G and Γ_G as a function H for $T = 9.24$ K, using LFA and HFA. Here we find Γ_G broader than resolution for the HFA states, but resolution limited for the LFA states. The hysteresis in A_G is similar to the peak intensity data in Fig. 1(a). Together, these results suggest that the field-cooled states (HTA and HFA) are always disorderd and the zero-field-cooled states (LTA and LFA) are, within our instrumental resolution,[8] consistent with LRO. It is interesting to note, however, that in both Figs. 2 and 3, very close to the transition, Γ_G is above resolution for the zero-field-cooled states. Whether this is merely an artifact due to the sample's composition gradient or other experimental

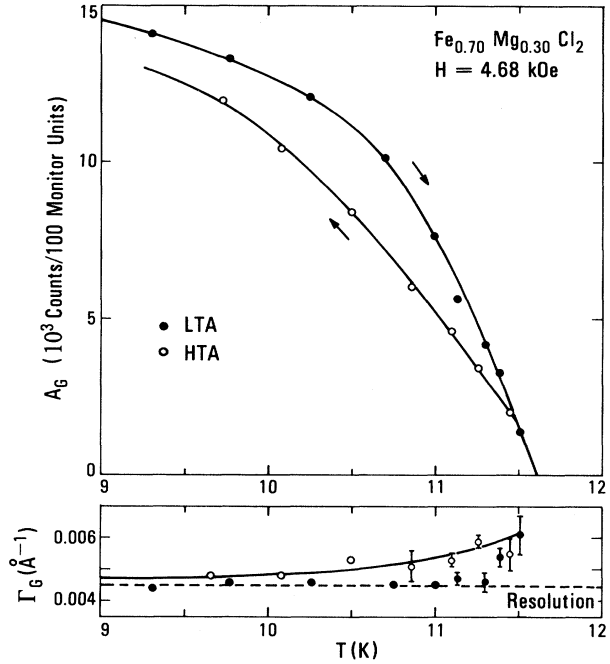

FIG. 2. Gaussian peak height A_G and peak width Γ_G as a function of T for $H = 4.68$ kOe. ● for LTA and ○ for HTA.

factors is uncertain. We will speculate below that this may be a true effect.

To test the stability of the field-cooled and zero-field-cooled states, we monitored $I(1, 0, \bar{1})$ as a function of time. Figure 4 shows a comparison of LFA and HTA states at $T = 10.15$ K and $H = 4.55$ kOe. Over six hours, there is a

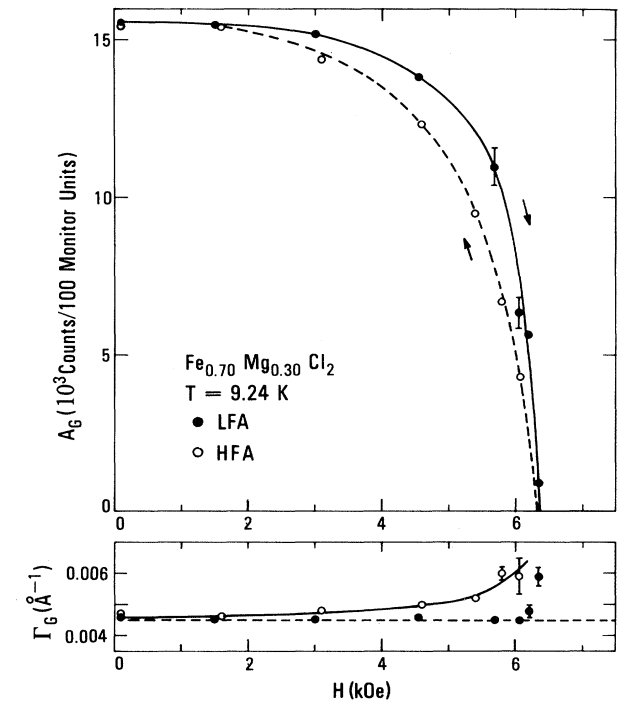

FIG. 3. Gaussian peak height A_G and peak width Γ_G as a function of H for $T = 9.24$ K. ● for LFA and ○ for HFA.

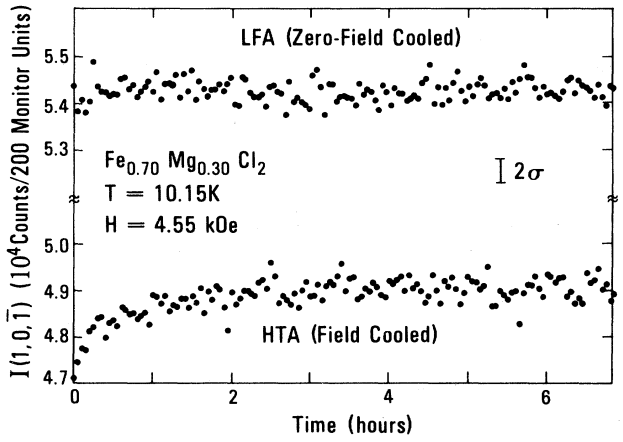

FIG. 4. Time dependence of $I(1,0,\bar{1})$ for the LFA and HTA states in the strongly hysteretic region.

clear indication that the HTA state drifts towards higher peak intensity, i.e., a more ordered state, but no change can be detected in the LFA state. The observed time dependence makes clear that, in spite of the favorable comparisons shown in Fig. 1(b), one should not regard the HTA and HFA states as *identical*. A similar caution should also apply to the LTA and LFA states. Although no time dependence is observed for these states, we cannot rule out the possibility that they are metastable states with very long relaxation times.

The hysteretic behavior described above is very similar to that found in the nondiluted system $Fe_{0.725}Co_{0.275}Cl_2$,[7] with one notable exception. Namely, field hysteresis is observed for all $H > 0$ in $Fe_{0.70}Mg_{0.30}Cl_2$ at low temperature, but only for $H \gtrsim 4$ kOe in $Fe_{0.725}Co_{0.275}Cl_2$. The simplest explanation is that there is a minimum exchange $J_{\min}$ in each system, when the applied field H satisfies $H \gtrsim J_{\min}S/g\mu_B$, domain walls formed across the weak bonds are energetically favorable and locally stable, which can lead to hysteresis. In $Fe_{0.70}Mg_{0.30}Cl_2$, $J_{\min}$ is zero. In $Fe_{0.725}Co_{0.275}Cl_2$, $J_{\min}$ is unknown, but $J_{\min}S/g\mu_B \lesssim 4$ kOe is a reasonable value since pure $FeCl_2$ has an intersublattice exchange that corresponds to about 11 kOe. That the system is able to find the favorable places to form domain walls in the field-cooled states can perhaps be argued as follows. When a field is applied in the paramagnetic phase, all $\langle \vec{S}_i \rangle$'s are in the same direction. With respect to the antiferromagnetically ordered state, this is equivalent to having domain walls everywhere. When the system crosses the phase boundary by either HTA or HFA, the ordering (annihilation of walls) nucleates from the most highly connected clusters. The walls that annihilate last are those situated across many weak (or broken) bonds. They can have long relaxation times since their annihilation involves the overturning of large domains. This dynamical process automatically places the domain walls across the weak bonds in the system. As a result, one might expect the wall configuration in the HFA and HTA states to be quite similar. The comparison of the two states in Fig. 1(b) is indeed consistent with this notion. For the zero-field cooled states (LTA or LFA), the system has LRO initially. If the ground state is disordered and consists of large domains, the system does not have an easy mechanism to create such domains (i.e., large energy barrier) and the relaxation time is likely to be much longer. The lack of time dependence of these states should therefore not be regarded as proof that they are the true equilibrium state.

Aside from relaxational effects, there are other interesting effects that should be considered in diluted antiferromagnets. For example, Fahnle[9] pointed out that near the percolation threshold, the random fields can destroy LRO by flipping one-dimensional clusters. King *et al.*[10] have also pointed out that an individual spin can flip when the applied field H overcomes the total molecular field acting on it from its neighboring spins. These ideas can be generalized and unified to a single effect which we shall call "cluster flip." Consider a small part of the magnetic infinite cluster which is connected to the main body by Q bonds and consists of N_1 and N_2 spins on sublattices 1 and 2, respectively. When the applied field H satisfies

$$Hg\mu_B(N_1 - N_2) > QJ\langle S \rangle \ , \tag{1}$$

it overcomes the molecular fields that bond the small cluster to the infinite cluster and the small cluster can flip as an entity according to the direction of H. Such an effect would reduce the number of spins on the infinite cluster which participate in the cooperative LRO and effectively pushes the system towards the percolation threshold. It is conceivable, therefore, that in high fields or at high temperatures (where $\langle S \rangle$ is small) a sufficiently large number of spins can be uncoupled from the infinite cluster to cause a "percolation transition" before the system becomes paramagnetic. In other words, it should be possible to have a spin-glass-like domain phase intervening between the paramagnetic phase and the antiferromagnetic phase if, in fact, $d_c = 2$.

We note that the arguments by Fahnle[9] and King *et al.*[10] correspond to the special cases of $Q = 2$ and $N_1 - N_2 = 1$ in Eq. (1), respectively. The generalization proposed above allows similar effects to take place in samples far above the percolation threshold and for fields small compared with J. It is interesting to see that cluster flip leads to hystereses naturally: At any temperature or field, there are many clusters that satisfy Eq. (1) and there are overlaps among them. When one cluster flips, it changes the configuration of the infinite cluster, and hence can prevent some other clusters from flipping. As a result, which cluster actually flips will depend on chance and the history of the system. Evidence for this effect has already been observed in the diluted system $Fe_{1-x}Zn_xF_2$ by King *et al.*[10] They found that for $T \gtrsim 0$, the χ vs H isotherms exhibit a series of peaks at fields that corresponds to integer values of $Q/(N_1 - N_2)$ in Eq. (1). Furthermore, they found that there is a crossover field H_{cr} below which hysteresis is very pronounced. Above this field, hysteresis is very weak, if it exists at all, and yet the system is not simply paramagnetic (it has peaks in the χ vs H curve). More recently, Birgeneau and co-workers[5] have investigated $Fe_{0.35}Zn_{0.65}F_2$ by neutron scattering. They found that in high fields there is a range of temperature below the apparent transition in which the system is disordered regardless of how it is cooled. Both of these observations are consistent with the existence of an intermediate domain phase. In principle, one would expect similar behavior in $Fe_{0.70}Mg_{0.30}Cl_2$. However, since this sample is more concentrated and H is much smaller in the present experiment, the domain phase, if it exists, probably occurs only in a very narrow region just below the phase boundary

where $\langle S \rangle$ is small. The absence of hysteresis in $I(1, 0, \bar{1})$ at 12.98 K in Fig. 1(a) and the broadening of Γ_G for the zero-field-cooled states near the transitions in Figs. 2–3 are consistent with the conjecture. In addition, Wong, von Molnar, and Dimon[2] have found that the specific-heat anomaly in $Fe_{0.682}Mg_{0.318}Cl_2$ disappears rapidly with increasing field, which may also be attributed to the domain phase.

To summarize, we have characterized the hysteretic behavior of $Fe_{0.70}Mg_{0.30}Cl_2$ systematically and suggested plausible explanations for its origin. Although these explanations are somewhat speculative, they suggest two new directions for further experiments. First, in nondiluted systems (those with two kinds of magnetic ions), the hystereses may not exist for $H \lesssim J_{\min}S/g\mu_B$. If true, one would be able to determine d_c unambiguously in systems with large $J_{\min}$, e.g., those with impurity ions which have a stronger exchange than the host ion. Second, in diluted systems, there is a possibility for the existence of an intermediate domain phase. To test this hypothesis, one can measure the phase boundaries in a given sample by different experiments and compare them carefully, e.g., the specific-heat peak may occur at a temperature different from the true transition temperature (which is known to happen in other disordered systems with domains, such as granular superconductors[11]). Such a study will be easier to carry out in the more diluted samples since the domain phase, if it exists, should be more expanded in those samples.

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