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Random-Field Effects in Two- and Three-Dimensional Ising Antiferromagnets

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Neutron scattering experiments have been performed in the diluted antiferromagnets $Rb_2Co_{0.7}Mg_{0.3}F_4$ and $Co_{0.3}Zn_{0.7}F_2$ in a uniform magnetic field. These systems are isomorphous to, respectively, two- and three-dimensional Ising ferromagnets in a site-random magnetic field. It is shown that small random magnetic fields destroy the long-range order at all temperatures, consistent with three as the lower marginal dimensionality; the structure factor in the disordered state is predominantly a squared Lorentzian.

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It is by now well known that small randomness in the exchange interactions has little, if any, effect on magnetic phase transitions.[1] On the other hand, it is expected that site-random magnetic fields will have drastic consequences. Specifically, in continuous-symmetry systems, the random field is believed to lower the effective spatial dimensionality (d) by two.[2] For Ising systems, the results are more controversial.[2-4] For $d > 4$ it is believed that one will also have a shift in d by 2. However, arguments based on sharp domain walls[2] suggest a lower marginal dimensionality, that is, the dimensionality below which long-range order cannot occur at finite temperatures, of $d_l = 2$ for the Ising random field problem rather than $d_l = 3$ which would follow from the $d \to d - 2$ algorithm. Recent theoretical[3] and experimental[5] results, including those discussed here, have precipitated a reconsideration of the domain-wall arguments.[6-8] These new theories suggest that domain-wall roughening[8] may indeed raise d_l from 2 to 3 thus making the $d \to d - 2$ law universal. At the same time, very little is understood about the magnetic correlations in the proposed random field disordered state.

In this paper, we report a neutron-scattering study of the magnetic correlations in the prototypical two- and three-dimensional (2D and 3D) diluted Ising antiferromagnets, $Rb_2Co_{0.7}Mg_{0.3}F_4$ (Ref. 9) and $Co_{0.3}Zn_{0.7}F_2$ (Ref. 10), in an applied magnetic field. Fishman and Aharony[4] have shown that in random-exchange Ising antiferromagnets (AF) the bulk magnetic field H, acting through the random component of the exchange interaction, generates a random staggered magnetic field H_{RF}. In the small-field limit, H_{RF} varies

linearly with H. Thus these materials provide very convenient systems for studying random-field effects in a systematic, quantitative fashion. Our principal results are that (a) at low temperatures and low fields the antiferromagnetic order is destroyed by the applied field in both 2D and 3D thus establishing $d_l \geq 3$; (b) the structure factor $S(\vec{Q})$ for small random fields has the usual form $A/(\kappa^2+q^2)^2+B/(\kappa^2+q^2)$ with $\vec{q}=\vec{Q}-\vec{G}$ ($\vec{G}$ is a reciprocal-lattice vector) as suggested by recent theory[6,7]; (c) in 2D both κ and $S(\vec{G})$ vary as simple powers of H.

The experiments were performed with a two-axis spectrometer at the Brookhaven National Laboratory high-flux reactor. The samples were mounted in a superconducting magnet with the tetragonal (001) axis vertical so that the field was applied along the Ising axis.[9,10] An incident neutron wave vector of 2.67 $Å^{-1}$ was used with two pyrolytic graphite filters to eliminate higher-order neutrons. For both samples measurements were performed primarily around the (100) antiferromagnetic lattice point. The instrumental resolution in both experiments was approximately 0.04 $Å^{-1}$ half-width at half maximum (HWHM) vertically and 0.002 $Å^{-1}$ HWHM transverse to the wave-vector transfer. The longitudinal resolution was 0.005 $Å^{-1}$ and 0.008 $Å^{-1}$ HWHM in the $Rb_2Co_{0.7}Mg_{0.3}F_4$ and $Co_{0.3}Zn_{0.7}F_2$ experiments, respectively.

A number of measurements were carried out to characterize the zero-field properties of both samples. First, we emphasize that no evidence was found for any chemical ordering in either of the samples. Pure Rb_2CoF_4 orders at 102.1 K while the 70% sample orders with the same square lattice AF structure at 42.5 ± 0.5 K, the uncertainty reflecting a rounding of T_N due to a concentration gradient. This smearing of T_N has no effect on the measurements to be discussed here. CoF_2 orders antiferromagnetically with a body-centered tetragonal AF structure at 38.0 K while the crystal of $Co_{0.3}Zn_{0.7}F_2$ ordered with the same structure at 6.7 ± 0.1 K.

We show in Fig. 1 transverse scans through the (100) peak at low temperatures in both samples. At zero field, narrow intense AF peaks are observed with widths given by the instrumental resolution and with line profiles which are essentially Gaussian in character thus confirming that the magnets have long-range order. The behavior in finite field is quite different. If the crystals are cooled in the presence of a magnetic field, the width of the scattering is larger and the peak

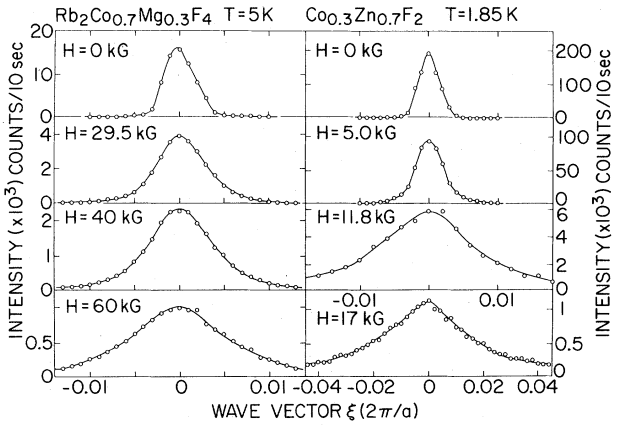

FIG. 1. Transverse scans through the (1,0,0) peaks as functions of magnetic field in $Rb_2Co_{0.7}Mg_{0.3}F_4$ and $Co_{0.3}Zn_{0.7}F_2$ at $T = 5$ and 1.85 K, respectively. In all cases the sample was cooled from high temperatures in the field. The solid lines at $H = 0$ kG give the transverse resolution function. The solid lines for finite H are the results of fits by Eq. (1).

intensity is decreased compared with the zero-field results. Further, it is clear from the scans shown in Fig. 1 that the line shapes are significantly affected by the field. We have confirmed that there is no corresponding change in the nuclear Bragg profiles. Thus we may conclude that the long-range order has been destroyed by the application of a small uniform field. No such effect is observed in the pure systems. We conclude, therefore, that the Fishman-Aharony mechanism is operative, that is, the uniform field has generated a random staggered field H_{RF}, and H_{RF} has destroyed the long-range order. This simple result alone demonstrates unambiguously that $d_l \geq 3$.

We have carried out an exhaustive study of the spin correlations in both samples as a function of magnetic field and temperature. In this Letter we discuss primarily the low-temperature results; the complete study will be presented elsewhere. It is evident from Fig. 1 that the peak intensity decreases and the width increases with increasing applied field H. For the scans in $Co_{0.3}Zn_{0.7}F_2$ below 10 kG and for all of the scans up to 70 kG in $Rb_2Co_{0.7}Mg_{0.3}F_4$ the line shapes are quite unusual. Specifically, from visual examination alone, it is clear that although the wings in the structure factor are quite extended, they are much too weak to be described by simple Lorentzian profiles. Lorentzian structure factors are normally obtained when temperature is the principal source of disorder. Recent theo-

ry[6,7] has suggested that in the presence of small random fields the structure factor should have the form[11]

$$S(\vec{Q}) = \frac{A}{(\kappa^2+q^2)^2} + \frac{B}{(\kappa^2+q^2)} \quad (1)$$

with $\vec{q} = \vec{Q} - \vec{G}$; here κ is the inverse correlation length; in three dimensions and possibly two, it is expected[6,7] that at low temperatures (T) $A/B \sim \langle H_{RF}^2 \rangle$.

The scattering is shown for selected scans on a semilogarithmic scale in Fig. 2. We have fitted these profiles with a variety of analytic forms including a Lorentzian, a Lorentzian raised to an arbitrary power $x/2$, and Eq. (1). As shown in Fig. 2, the profile is distinctly non-Lorentzian but may be described equally well by Eq. (1) or by the $(\kappa^2+q^2)^{-x/2}$ form. We cannot differentiate between these two. However, since Eq. (1) is strongly preferred by current theory we shall limit our discussion to results obtained from the analysis using Eq. (1). Results from the general power-law form will be given in a future publication. For $Rb_2Co_{0.7}Mg_{0.3}F_4$, Eq. (1) describes the data well at all fields and temperatures. At $T = 5$ K the Lorentzian term makes a constant fractional contribution of about 8% to the peak intensity at $q = 0$, that is $A/B\kappa^2 = 0.08$, over the complete field range from 18.5 to 70 kG. Both the inverse correlation length κ and the peak intensity $S(1, 0, 0)$ at fixed applied field are independent of temperature between 25 and 5 K. We note that the temperature scaling variable[9] is $\mu = e^{-2.8J/kT} \cong e^{-235/T}$ so that μ will be extremely small at $T = 5$ K. We conclude, therefore, that the system will be disordered at $T = 0$ K, provided that the ground state is approached by decreasing temperature in the presence of the applied field. We show in Fig. 3 on a log-log scale κ and S versus applied field at 10 K in the 2D system $Rb_2Co_{0.7}Mg_{0.3}F_4$. Both follow simple power laws: $\kappa \sim H^{1.6}$ and $S(1,0,0) \sim H^{-3.2}$. We note that at d_l, theory predicts $\kappa \sim \exp(-c/H_{RF}^2)$ whereas below d_l one expects a simple algebraic dependence of κ on the random field. Clearly we observe the latter behavior in our 2D AF. These results therefore necessitate $d_l > 2$ and are consistent with $d_l = 3$. The simplest theoretical ideas[11] suggest $\kappa \sim H_{RF}^2$. By direct measurement we find that in Rb_2CoF_4 for H between 0 and 80 kG, $M \sim H^{0.8}$ to a very good approximation. From Fishman and Aharony[4] we expect $H_{RF} \sim M$. Thus $\kappa \sim H_{RF}^{2.0}$, in agreement with theory. There is currently no theory for the dependence of the total structure factor $S(\vec{Q})$ on

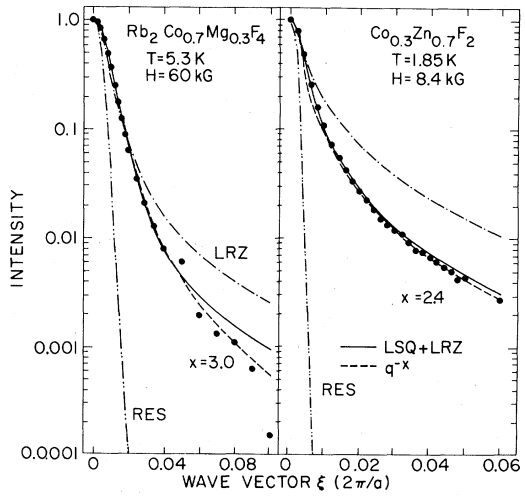

FIG. 2. Detailed line shapes with background subtracted for scans along $(1+\xi, 0, 0)$ in $Rb_2Co_{0.7}Mg_{0.3}F_4$ and $(1, \xi, 0)$ in $Co_{0.3}Zn_{0.7}F_2$. The successive lines represent the results of fits to a pure Lorentzian (LRZ), Lorentzian plus squared Lorentzian (LSQ + LRZ), and the power-law form $A/(\kappa^2+q^2)^{x/2}$. The dot-dashed lines labeled RES give the measured instrumental resolution function.

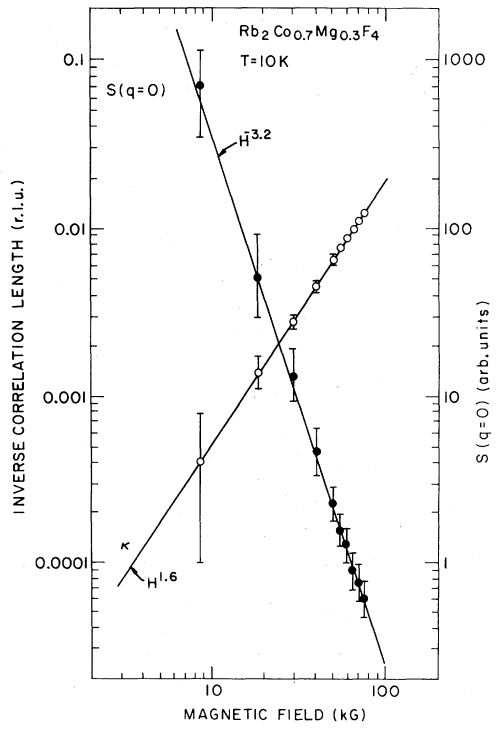

FIG. 3. Fitted inverse correlation length κ, and peak intensity $S(1, 0, 0)$ as a function of magnetic field in $Rb_2Co_{0.7}Mg_{0.3}F_4$.

H_{RF} in two dimensions. Our results should therefore stimulate the development of such a theory.

The results for $Co_{0.3}Zn_{0.7}F_2$ are somewhat more complicated. For $H < 10$ kG the scattering is well described by Eq. (1) with κ less than or comparable with the resolution width and with the squared Lorentzian term dominant. As stated above this is consistent with $d_t = 3$ and the predicted form of the line shape; furthermore, κ decreases as the field decreases as predicted by the theory. For $H > 12$ kG, $S(\vec{Q})$ has a Lorentzian line shape as also observed for $T > 6.7$ K at low fields. We believe that this reflects a changeover from the unusual random-field–induced type of disorder to conventional paramagnetic disorder. This is probably because the local-field energy $g\mu_B H$ for 12 kG is comparable with the transition-temperature energy kT_N. We note that in $Rb_2Co_{0.7}Mg_{0.3}F_4$ the local-field energy is always much smaller than kT_N so that in that case we are always in the small H limit. It is not therefore too surprising that we find results for κ, A, and B as a function of applied field which differ markedly from the theory which is applicable only to small H. We shall therefore discuss these results in detail elsewhere. A theory which takes into account both H and H_{RF} and which predicts $S(\vec{Q})$ explicitly would be most valuable.

Our experiments have shown that a small random field destroys long-range order in both 2D and 3D Ising magnets and that $S(\vec{Q})$ is consistent with the predicted and unusual form of correlation function $A/(\kappa^2+Q^2)^2$ and $\kappa \to 0$ as $H \to 0$.

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