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**A new type of spin ordering in a random magnet
with competing anisotropies**

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The result of an elastic neutron scattering study of the phase transition in the random magnet with competing anisotropies $\text{Fe}_{0.75}\text{Co}_{0.25}\text{Br}_2$ is reported. We found that the ordering of the spin component parallel to the crystalline c axis at the transition from the paramagnetic to antiferromagnetic phase induces an ordering of the spin component perpendicular to it. Thus, there is no boundary separating the higher temperature antiferromagnetic and lower temperature mixed ordering phase. We interpret this new type of spin ordering as arising from a coupling between the orthogonal spin components.

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I. INTRODUCTION

The magnetic phase transition in random systems with competing spin anisotropies has been extensively studied both theoretically and experimentally. The mean-field theories¹⁻³ have predicted the presence of three ordered phases, bounded by four critical lines, in the concentration-versus-transition-temperature (x - T) phase diagram of such a random magnet. These four second-order transition lines meet at a tetracritical point. Katsumata et al.⁴⁻⁶ have reported that the random magnet $\text{Fe}_{1-x}\text{Co}_x\text{Cl}_2 \cdot 2\text{H}_2\text{O}$ exhibits a tetracritical phase diagram as shown in Fig. 1. They have observed sharp anomalies at the four phase boundaries in both the susceptibility and heat capacity measurements.⁴ A neutron scattering study⁶ on a sample close to the tetracritical concentration ($\text{Fe}_{0.4}\text{Co}_{0.6}\text{Cl}_2 \cdot 2\text{H}_2\text{O}$) showed that the higher and the lower transition temperatures are equally sharp and that the orderings of the spin components occur independently in accordance with the theoretical prediction by Aharony and Fishman.^{7,8}

A quite different behavior from this idealized tetracritical phase diagram has been observed in the anhydrous random magnet with competing anisotropies $\text{Fe}_{1-x}\text{Co}_x\text{Cl}_2$ [Refs. 9-12]. Although the higher temperature phase boundaries in $\text{Fe}_{1-x}\text{Co}_x\text{Cl}_2$ corresponding to the lines L_1 and L_2 in Fig. 1 are sharply defined, the two lower temperature ones corresponding to the lines L_3 and L_4 in Fig. 1 are broad.

Another kind of phase diagram has been found in a random magnet with competing spin anisotropies $\text{Fe}_{1-x}\text{Co}_x\text{Br}_2$ (Ref. 13). Figure 2 shows the x - T phase diagram of $\text{Fe}_{1-x}\text{Co}_x\text{Br}_2$ obtained from the susceptibility and magnetization measurements.¹³ The

lines L_1 and L_4 indicate the ordering of the spin component parallel to the c axis($S_{//}$) , while the line L_2 represents the ordering of the spin components in the c plane($S_{\perp}$) . The phase transition at L_1 is sharp; the susceptibility along the c axis is sharply peaked at the transition temperature. The transition at L_2 is also well defined except in the vicinity of the point at which the four lines meet. On the other hand, the transition at L_4 is broad as in $\text{Fe}_{1-x}\text{Co}_x\text{Cl}_2$ [Refs. 9-12]. No detectable anomaly was observed at L_3 in the magnetic measurements.¹³

In order to clarify the nature of the phase transition in $\text{Fe}_{1-x}\text{Co}_x\text{Br}_2$, especially that at L_3 , we have performed a neutron scattering study on a single crystal sample of $\text{Fe}_{0.75}\text{Co}_{0.25}\text{Br}_2$.

II. PRELIMINARY DETAILS

Both FeBr_2 and CoBr_2 have the same hexagonal structure($P\bar{3}m$). The lattice parameters are, $a=3.772$ Å and $c=6.223$ Å for FeBr_2 , and $a=3.728$ Å and $c=6.169$ Å for CoBr_2 . From the neutron scattering study,¹⁴ the two compounds are antiferromagnetic at low temperatures. The spin structures of FeBr_2 and CoBr_2 are such that spins in a c plane are coupled ferromagnetically, while they are coupled antiferromagnetically with the spins in the adjacent c planes. Spins in FeBr_2 point parallel to the c axis, while they are confined in the c plane in CoBr_2 , the anisotropy in this plane being rather small. Thus the alloy system $\text{Fe}_{1-x}\text{Co}_x\text{Br}_2$ provides an archetypal example of a random magnet with competing orthogonal Ising and XY anisotropies.

The neutron scattering experiments were carried out using a triple-axis spectrometer at the High Flux Beam Reactor of the Brookhaven National Laboratory. The single crystal of

$\text{Fe}_{0.75}\text{Co}_{0.25}\text{Br}_2$ used in this experiment was cut from a larger boule grown by S. Legrand in Saclay. The dimensions of the sample were about $10 \times 20 \times 3 \text{ mm}^3$. The mosaic spread of the crystal was approximately 0.2° full width at half maximum measured about the $(0,0,l)$ direction. The sample was mounted with the a^*-c^* axes in the horizontal scattering plane.

III. EXPERIMENTAL RESULT

We first measured the line shape along the $(h,0,0.5)$ direction. Here the $(0,0,0.5)$ represents the antiferromagnetic Bragg point. We found that the line is composed of two parts; a sharp Bragg peak whose width is limited by the neutron spectrometer resolution indicating a long range ordering and broad tails. These tails presumably come from short-range ordered spins.

Figure 3 shows the measured peak intensities at $(0,0,0.5)$ and $(2,0,0.5)$ as a function of temperature. From the relation,¹⁵ $I \propto \sin^2 \alpha$, between the intensity(I) of scattered neutrons and the angle(α) that the scattering vector makes with the spin direction in the sample, we see that the intensity measured at the $(0,0,0.5)$ magnetic Bragg point depends only on $S_\perp$. On the contrary the scattering from the $(2,0,0.5)$ magnetic Bragg point contains contributions from both $S_\parallel$ and $S_\perp$. It is evident in Fig. 3 that the dominant $S_\parallel$ component orders at the higher transition temperature and it induces an ordering of the $S_\perp$ component. In order to eliminate any double-Bragg contamination, we have varied the incident energy(E_i). We found that $E_i=13.5 \text{ meV}$ minimized the double scattering processes and still allowed a simultaneous study of the two Bragg peaks. In order to be absolutely sure that the scattering at $(0,0,0.5)$ is not contaminated by double scattering,

measurements were made at $E_i=5.0$ meV. There was no indication of double scattering at this energy and the temperature variation of the peak intensity at (0,0,0.5) is similar to that observed in Fig. 3.

IV. DISCUSSION

Wong et al. have proposed a model to explain the broadening of the lower temperature phase boundaries in $\text{Fe}_{1-x}\text{Co}_x\text{Cl}_2$ based on a random field effect.¹² Suppose, coupling exists between the orthogonal spin components $S_{//}$ and $S_{\perp}$ of the form $J S_{//} \cdot S_{\perp}$, where J is site random. As a consequence if, $S_{//}$ component establishes a long-range ordering at the corresponding higher temperature phase boundary, this interaction produces a site random field $J \langle S_{//} \rangle$ to the $S_{\perp}$ component, where $\langle \dots \rangle$ means the thermal average. This random field causes the lower temperature phase transition to be smeared. In the case when the random field is not completely random but has a finite value after being averaged over all sites, we expect that the ordering of $S_{//}$ component induces an ordering of $S_{\perp}$ when the system is cooled from the paramagnetic phase. This explains what we have observed. Wong^{16,17} has suggested from a neutron scattering experiment on $\text{Fe}_{0.725}\text{Co}_{0.275}\text{Cl}_2$ in applied magnetic fields that the sample has moments in zero field tilted slightly from the c axis at temperatures between the higher and lower phase boundary. In the present sample, we have found that the ordering of $S_{//}$ at the transition from the paramagnetic to antiferromagnetic phase induces an ordering of $S_{\perp}$. This kind of ordering of orthogonal spin components has not been observed so far.

Our observation of the simultaneous ordering of the orthogonal spin components is consistent with the x-T phase diagram shown in Fig. 2. If there is no coupling between $S_{//}$ and $S_{\perp}$, only the $S_{//}$ component is ordered in the higher temperature phase between L_1 and L_3 and both of the $S_{//}$ and $S_{\perp}$ components exhibit an ordering at the lower temperature phase below L_3 . In the present case, since the ordering of $S_{//}$ induces an ordering of $S_{\perp}$ at L_1 , there is no distinction between the higher and lower temperature phase.

Finally, it should be noted that the mechanism of producing a coupling between the $S_{//}$ and $S_{\perp}$ components discussed here is different from that proposed by Matsubara and Inawashiro(MI).¹⁸ MI studied theoretically the phase diagram of a random mixture of two antiferromagnets whose easy axes are not orthogonal and found that the coupling between the $S_{//}$ and $S_{\perp}$ components existed in the Hamiltonian.¹⁸ The MI theory explains the absence of the lower temperature phase boundaries in $\text{Ni}_x\text{Co}_{1-x}\text{Cl}_2 \cdot 6\text{H}_2\text{O}$ [Ref.19] and the simultaneous ordering of the $S_{//}$ and $S_{\perp}$ spin components in $\text{Fe}_x\text{Co}_{1-x}\text{TiO}_3$ [Ref.20]. In these systems, the easy axes of the constituent magnetic ions are not orthogonal.^{19,21}

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Figure Captions

- Fig. 1. Concentration-versus-transition-temperature phase diagram of $\text{Fe}_{1-x}\text{Co}_x\text{Cl}_2 \cdot 2\text{H}_2\text{O}$ obtained from susceptibility and heat capacity measurements. Figure from Ref. 4.
- Fig. 2. Concentration-versus-transition-temperature phase diagram of $\text{Fe}_{1-x}\text{Co}_x\text{Br}_2$ obtained from magnetic measurements. Figure from Ref. 13.
- Fig. 3. Peak intensities at (0,0,0.5) and (2,0,0.5) versus temperature for the $\text{Fe}_{0.75}\text{Co}_{0.25}\text{Br}_2$ crystal.

Fig. 1

$\text{Fe}_{0.75}\text{Co}_{0.25}\text{Br}_2$ Fig. 3

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