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Spin-waves in the uniaxial spin-glass system $\text{Fe}_{1-x}\text{Mg}_x\text{Cl}_2$

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Abstract

We report an inelastic neutron scattering study of the uniaxial dilute antiferromagnets $\text{Fe}_{1-x}\text{Mg}_x\text{Cl}_2$, for $x=0.3$, 0.45 and 0.6 . The first two samples have long-range AF order. Their spin-wave peaks are broadened by the exchange fluctuations. The $x=0.6$ sample orders like an Ising spin-glass with a very short AF correlation length ($\xi \approx 10\text{\AA}$). For wavelength $\lambda \leq \xi$, the spin-wave peaks are similar to the dilute antiferromagnets, but for $\lambda \geq 2\xi$, the peak sharpens up and becomes resolution limited at the zone center. We give a simple explanation for this behavior.

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MASTER

The existence of spin-wave in spin-glasses has attracted much theoretical^{1,2} and experimental³⁻⁵ attention in recent years. Computer simulation² and inelastic neutron scattering³⁻⁵ studies have thus far been unable to observe these elementary excitations in systems with isotropic RKKY or Heisenberg interactions. In reentrant systems with successive ferromagnetic and spin-glass transitions, the spin-waves established in the ferromagnetic phase are known to disappear as the system approaches the spin-glass transition.⁴ This is either due to a sharp decrease of the stiffness constant which makes the spin-waves difficult to observe, or caused by a wide distribution of relaxation processes in the spin-glass phase which overdamp the excitations.⁶ In this paper, we report the first spin-wave study on an Ising-like dilute antiferromagnet $\text{Fe}_{1-x}\text{Mg}_x\text{Cl}_2$ which evolves into a spin-glass with increasing dilution. Three samples with $x=0.3$, 0.45 and 0.6 were studied. Our emphasis is to observe how the spin-waves are affected as the system change from antiferromagnetic at $x=0.3$ to spin-glass at $x=0.6$. Our results show a new novel feature. Namely, in the latter sample where the spin-spin correlation length ξ is only $10\text{\AA}$, a spin-wave peak still exists at the zone center ($Q=0$ and $\lambda=\infty$) and it is resolution limited. The spin-wave energy is roughly independent of Q for wavelength $\lambda \geq 2\xi$. We explain this phenomenon in terms of spin-cluster precession around the easy axis.

FeCl_2 is an extensively studied antiferromagnet (AF).⁷ It has ferromagnetic layers that stack antiferromagnetically along the hexagonal c-axis. The easy axis for the spins is perpendicular to the layer, parallel to the c-axis. Within each layer, the first-neighbor exchange is ferromagnetic and the second-neighbor exchange is antiferromagnetic. Substituting the Fe-ions randomly by the nonmagnetic Mg-ions

can cause the system to evolve from AF order to spin-glass order.^{8,9} During the last three years, we have performed a variety of experiments on three samples of representative Mg-concentrations: 30%, 45% and 60%. Our a.c. susceptibility and elastic neutron scattering data show that the 30%-Mg sample is a regular dilute AF, the 45%-Mg sample has both AF and spin-glass order, and the 60%-Mg sample has purely spin-glass order with a very short AF correlation length ($\xi \approx 10\text{\AA}$).^{8,10} In the following, we briefly describe the results obtained by inelastic neutron scattering on these three samples. A more complete description is given elsewhere.¹⁰

Our experiments were carried out at the High Flux Beam Reactor of the Brookhaven National Laboratory.⁸ Different spectrometers, incident energies and collimations were used depending on the intensity and resolution requirements. The samples were always oriented with their reciprocal a^* and c^* axes in the horizontal scattering plane, so that constant- Q scans can be performed for wavevectors in the $(h,0,l)$ plane. Because of the weak inter-layer exchange, the spin-wave energies are essentially independent of l .⁷ Most of our measurements were made for $Q=(h,0,3)$ with h between 0 and 0.5, or $Q=(h,0,\bar{1})$ with h between 0.5 and 1. These are equivalent Brillouin zones that center at $(0,0,3)$ and $(1,0,\bar{1})$, respectively.

In both the 30%-Mg and the 45%-Mg samples, we observed broad spin-wave peaks over the entire Brillouin zone, from $h=0$ to $h=0.5$. Fig. 1 shows the dispersion curves, along with that of pure FeCl_2 from Ref. 7. Due to the uniaxial anisotropy, there is always a gap at the zone center. The gap energy E_g is given by $g\mu_B H_A$, where H_A is the anisotropy field and $g\mu_B$ is the moment of the Fe^{2+} ion. According to Birgeneau et al.,⁷ H_A is due to both single-ion anisotropy and exchange anisotropy,

with approximately a 3:4 ratio. Hence E_g should be lower for increasing Mg-concentrations, which is what we observed. At the zone boundary ($h=0.5$), the spin-wave energy is also reduced with increasing dilution because it reflects the average exchange interaction in the system. The local fluctuations of the exchange cause the width of the spin-wave peak to increase for larger Q 's. This behavior is typical of all dilute magnetic systems¹¹ but it has never been completely understood. Since both the 30%-Mg and 45%-Mg samples have long-range AF order, these results are to be expected. We should point out, however, that the 45%-Mg sample has a Néel transition at 7.5 K and a spin-glass transition at 3 K. These two types of order coexist below 3 K.⁸ The data in Fig. 1 are taken at 1.45 K. It shows that the spin-waves are not destroyed by the presence of spin-glass ordering. This behavior is contrary to the reentrant systems where the spin-waves established in the ferromagnetic phase are always destroyed by the spin-glass ordering.^{4,5} The difference is probably due to the fact that the spin-glass behavior in the 45%-Mg sample is associated only with the longitudinal spin-component⁸ and it does not overdamp the spin-waves which are associated with the transverse components.

The behavior of the 60%-Mg sample is even more intriguing. Fig. 2 show several constant- Q scans with $Q=(h,0,3)$. For $h \geq 0.3$, there are broad spin-wave peaks similar to the other two samples. The reason is quite simple: the reciprocal lattice vectors are $a^*=2.007\text{\AA}^{-1}$ and $c^*=0.179\text{\AA}^{-1}$, so for $Q \geq 0.3a^*$ the wavelength $\lambda \leq 10\text{\AA} \approx \xi$. In this short wavelength limit, the spin-waves are insensitive to whether the system has true long-range AF order. This is similar to spin-waves in one-dimensional systems¹² or other insulating ferromagnets above the Curie temperature.¹³

For longer wavelengths ($\xi \leq \lambda \leq 2\xi$ or $0.15 \leq h \leq 0.3$), we find that the spin-wave peaks are not as well defined and more difficult to observe. However, for still longer wavelengths ($\lambda > 2\xi$ or $h < 0.15$), the spin-wave peaks sharpen up and become resolution limited (0.1 meV half-width-half-maximum) at the zone center ($\lambda \rightarrow \infty$ and $h \rightarrow 0$).

To understand the unusual behavior of the 60%-Mg sample, we show the complete energy versus Q dispersion curve in Fig. 3. We note that the energy gap at the zone center is about 1 meV and it remains essentially unchanged for h up to 0.15. This energy is consistent with the estimated single-ion anisotropy in FeCl_2 ⁷ and it gives an anisotropy field H_A independent of the exchange interactions. The spins can precess around this field with a Larmor frequency $\omega = g\mu_B H_A / \hbar$. The direction of H_A (up or down) at a site is the same as the ordered moment at that site. For a usual ferromagnet, H_A at all the sites point in the same direction and the spins can all precess in phase in the $\lambda \rightarrow \infty$ limit. Here, however, the frozen moments are only correlated over a distance ξ , so the phase of the precessions can only be coherent over the same distance. For spins farther apart than ξ , the relative phase angle should appear random. On average, we might expect the phase to change by π over a distance of 2ξ . In this picture, we can view the excitations as precessions of spin-clusters of average size ξ around the anisotropy axis. From the uncertainty principle ($\Delta k \Delta x \geq 1/2$), such excitations are equivalent to a spectrum of spin-waves with the same energy over a range of wavevectors $0 \leq Q \leq 1/2\xi$. This is what the lower branch in Fig. 3 shows. As mentioned above, for these excitations to be well-defined or long-lived, the system should not have slow relaxations associated with the transverse

Fig. 1

Fig. 2a,b

Fig. 2c,d

Fig. 2e,f

Fig. 3

spin-component. We have verified this by other experiments. Our a.c. susceptibility measurements perpendicular to the easy axis show completely reversible behavior and elastic neutron scattering also show that there are no frozen transverse moments at low temperatures.¹⁰ Finally, we should add that the apparently uncorrelated precessions of spin-clusters may be due to the fact that we are making observations on the two-spin correlation function. This is not the appropriate quantity to characterize the fluctuations of the spin-glass order parameter. It is possible that long-range phase coherence can be restored by considering four-spin correlation functions such as $\langle S_x^2(0,0) S_x^2(r,t) \rangle$. (If $S_x(0)$ and $S_x(r)$ are out of phase by π , then $S_x^2(0)$ and $S_x^2(r)$ are exactly in phase).

In summary, our results have revealed a new behavior for spin-waves in uniaxial spin-glasses. For $\lambda < \xi$, the spin-waves are similar to any other dilute magnets. For $\lambda > 2\xi$, there are sharp spin-wave modes corresponding to the anisotropy energy. These can be explained by the precession of spin-clusters around the easy axis. Between these two regimes, there is an abrupt crossover which cannot be described in simple terms.

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therein.

Figure Captions

Fig. 1: Spin-wave dispersions along the $(h,0,0)$ direction for three samples with long-range AF order. The FeCl_2 data are from Ref. 7.

Fig. 2: Constant- Q scans in the spin-glass phase of $\text{Fe}_{0.4}\text{Mg}_{0.6}\text{Cl}_2$ show a well-defined peak for large and small Q 's. The lines are guides to the eye. The bars marked "R" represent the instrumental resolution.

Fig. 3: Spin-wave dispersion along the $(h,0,0)$ direction in $\text{Fe}_{0.4}\text{Mg}_{0.6}\text{Cl}_2$. There is large uncertainty in the spin-wave energy for $0.15 < h < 0.3$.

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