Transverse Fluctuations in an Ising Spin-Glass: $Fe_{0.4}Mg_{0.6}Cl_2$

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ac-susceptibility and neutron-diffraction experiments show that $Fe_{0.4}Mg_{0.6}Cl_2$ is a three-dimensional Ising spin-glass with a transition temperature $T_{sg}=3.4$ K. Inelastic-scattering experiments show a sharp spin-wave peak at the zone center for all temperature $T \lesssim 2T_{sg}$. This is the first direct observation of long-wavelength excitation in a spin-glass system. We suggest that it is due to the precession of spin clusters around the easy axis.

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In magnetic systems with conventional ferromagnetic or antiferromagnetic order, spin fluctuations transverse to the ordering direction are well described by spin waves. In spin-glasses, however, the existence of these elementary excitations are unclear. Halperin and Saslow[1] have pointed out that a system of spins ordered in random directions can still have spin waves, but the presence of microscopic relaxation processes with comparable frequencies can lead to overdamping of the modes. The decrease of the exchange stiffness can also make the excitations empirically difficult to observe. Inelastic-neutron-scattering[2] and computer-simulation[3] studies of Heisenberg spin-glasses have thus far been unable to confirm the existence of spin waves. In systems with reentrant (ferromagnet to spin-glass) transitions, as the temperature is lowered towards the spin-glass phase, inelastic-neutron-scattering experiments[4] have shown that the spin-wave peaks at finite energies in the ferromagnetic phase collapse into a central peak at zero energy. At the same time, the stiffness constant decreases and the damping coefficient increases. These observations are consistent with Halperin and Saslow's arguments. The damping can be attributed to the fact that there is a wide range of slow relaxation processes in the spin-glass state[5] which correspond to the reorientation of spin clusters.

The situation in *Ising-type* spin-glasses can be quite different, however. In such systems, the slow relaxations are associated with the longitudinal spin component $\mathbf{S}_{\parallel}$ (parallel to the easy axis) and the spin waves are associated with the transverse component $\mathbf{S}_{\perp}$ (perpendicular to the easy axis). The damping due to disorder may not be too severe. Furthermore, the energies of the long-wavelength modes should be mostly determined by the anisotropy and not the exchange. A small stiffness constant would not hinder the observation of these modes. In this Letter, we report the first spin-wave study of an easy-axis spin-glass: $Fe_{0.4}Mg_{0.6}Cl_2$. Our results show clear evidence for long-wavelength spin waves. In fact, we find that they exist at temperatures well above the freezing temperature (T_{sg}). Some qualitative explanations of this new phenomenon are suggested.

The structural and magnetic properties of the $Fe_{1-x}Mg_xCl_2$ system have been described previously.[6-8] We recall that it is an insulating crystal with a layered structure. There are competing ferromagnetic and antiferromagnetic exchange interactions due to Fe ions at different neighboring distances. The easy axis is perpendicular to the layers. With increasing Mg concentration x, the low-temperature phase of the system changes from antiferromagnetic to spin-glass. There is a range of x ($\lesssim 0.5$) where the two types of order coexist.[6] For $x=0.6$, the system behaves like an almost ideal Ising spin-glass, as we demonstrate in the following.

In Fig. 1, we show the ac-susceptibility data obtained by the conventional mutual-inductance technique, with a driving field of 0.1 Oe. For measurements made parallel to the easy axis, the real part of the susceptibility ($\chi'_{\parallel}$), as a function of temperature, shows a cusp typical of spin-glass systems. The cusp temperature is strongly frequency dependent, shifting to lower T as the frequency is reduced. For the lowest frequency we used (11 Hz), it occurs at about 3.4 K, which we identify as the transition temperature T_{sg}. For temperatures near or below T_{sg},

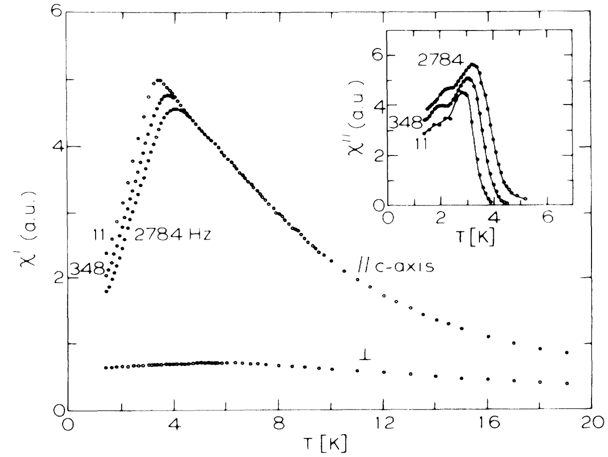

FIG. 1. Magnetic susceptibilities parallel and perpendicular to the easy axis.

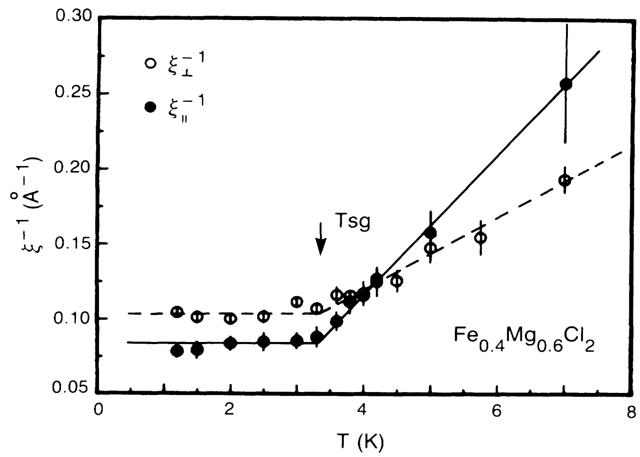

FIG. 2. Inverse of spin-spin correlation lengths parallel and perpendicular to the easy axis.

there is a rather large onset of the imaginary part $\chi''_{\parallel}$, which is also strongly frequency dependent. These results illustrate that there are slow relaxation processes associated with the freezing of $\mathbf{S}_{\parallel}$. Additionally, we show in Fig. 1 the transverse susceptibility $\chi'_{\perp}$ measured perpendicular to the easy axis. It has no measurable frequency dependence or out-of-phase response. The temperature dependence is very weak, showing only a broad maximum around 6 K. These results indicate that $\mathbf{S}_{\perp}$ does not have a freezing transition and its relaxation rate is much faster than the measurement frequencies. In other words, only $\mathbf{S}_{\parallel}$ is frozen and the system behaves like an Ising spin-glass. The specific heat of this system had also been measured before and it shows a broad maximum above T_{sg}, consistent with spin-glass behavior.[6]

The *instantaneous* magnetic correlations are measured by double-axis neutron scattering. The experimental conditions are similar to those described previously.[6,9] We find that there are no $\langle \mathbf{S}_{\perp}\mathbf{S}_{\perp}\rangle$ correlations at any temperature, either above or below T_{sg}. However, there are weak $\langle \mathbf{S}_{\parallel}\mathbf{S}_{\parallel}\rangle$ correlations, manifested by Lorentzian diffuse peaks at the superlattice points where pure $FeCl_2$ has antiferromagnetic Bragg peaks. This implies the existence of short-range $\mathbf{S}_{\parallel}$ correlations which are ferromagnetic in the layer and antiferromagnetic perpendicular to the layer.[8] Figure 2 shows how the in-plane and out-of-plane inverse correlation lengths ($\xi_{\perp}^{-1}$ and $\xi_{\parallel}^{-1}$) evolve as a function of temperature. Above T_{sg}, both of these decrease with decreasing temperature. Below T_{sg}, they are roughly constant, with $\xi_{\parallel} \approx 12$ Å and $\xi_{\perp} \approx 10$ Å. The former corresponds to twice the layer spacing and the latter is approximately 2.8 times the in-plane lattice constant a. Hence, the system is fully three dimensional in spite of the layered structure. The fact that similar $\mathbf{S}_{\perp}$ correlations are not observed also reconfirms the Ising character established by the susceptibility data.

The spin dynamics of the system is studied by triple-axis inelastic neutron scattering. We find that the longitudinal fluctuations are too slow to be effectively studied with our limited instrumental resolution (100 μeV), but the transverse fluctuations exhibit a new and interesting behavior. By performing energy scans with constant wave vector Q, where $Q=(h,0,l)$, we observe clear spin-wave peaks over much of the Brillouin zone (see below). All the experimental details will be described in future publications.[9] Here, we only summarize our main results:

(1) The spin-wave energies are essentially independent of l, the out-of-plane component of the wave vector. This is readily explained by the relatively weak interlayer exchange, as in pure $FeCl_2$.

(2) Figure 3 shows the in-plane spin-wave dispersion along $(h,0,3)$. For comparison, we include similar data on pure $FeCl_2$ from Ref. 8. The new data are taken at $T=1.70$ K$=0.5T_{sg}$ and the old data were obtained at $T=5$ K$=0.2T_N$, where T_N is the Néel temperature. There are some important similarities and differences between the two. Both systems show a sharp energy gap E_g at the zone center ($h=0$). In $FeCl_2$, $E_g=2.2$ meV and it is known to be due to both the single-ion anisotropy and the exchange anisotropy, with approximately a 3:4 ratio. In $Fe_{0.4}Mg_{0.6}Cl_2$, $E_g=1.05\pm0.05$ meV. This reduced value is probably due exclusively to the single-ion anisotropy.

(3) For wave vectors with h up to about 0.15, the spin-wave peaks in $Fe_{0.4}Mg_{0.6}Cl_2$ remain resolution limited[9] and their energies are roughly the same as E_g, as shown in Fig. 3. Compared with the $FeCl_2$ data, it is evident that the stiffness constant is much smaller. In the wave vector range $0.15<h<0.3$, the spin-wave peaks are relatively ill defined and difficult to observe. The two data points in Fig. 3 in this range have large uncertain-

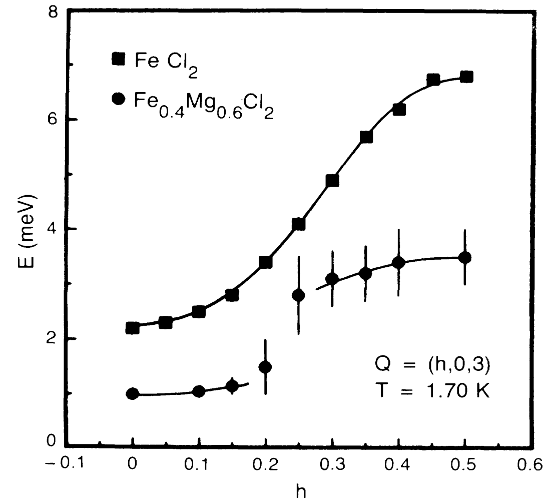

FIG. 3. In-plane spin-wave dispersions for $Fe_{0.4}Mg_{0.6}Cl_2$ and $FeCl_2$.

ties. Since the in-plane wavelength is given by $\lambda_\perp = 2\pi/ha^*$ (where $a^* = 2.01$ Å^{-1} is the reciprocal-lattice vector), this range of h corresponds to $\lambda_\perp$ between 10 and 20 Å, i.e., $\xi_\perp \lesssim \lambda_\perp \lesssim 2\xi_\perp$. It is possible that the spin waves are heavily damped for these wavelengths.

(4) For $h > 0.3$, the spin-wave peaks become pronounced again, but their widths are much broader than the instrumental resolution. These are represented by the large error bars in the upper branch of Fig. 3. This behavior is common in dilute magnets and it reflects the local fluctuations of the exchange interactions.[10] The reduction of the zone-boundary energy, from 6.8 meV in $FeCl_2$ to about 3.5 meV in $Fe_{0.4}Mg_{0.6}Cl_2$, is consistent with the reduction of the average exchange due to dilution. It is interesting to note that our data[9] in this Q range are remarkably similar to those obtained recently by Uemura and Birgeneau[11] on another dilute antiferromagnet $Mn_{0.5}Zn_{0.5}F_2$. These authors noted that when the wavelengths are shorter than the percolation correlation length, the excitations might be considered as quasilocalized *fractons* instead of propagating spin waves. In our sample, as a result of the dominance of the in-plane ferromagnetic exchange, the relevant percolation threshold is $x_c = 0.5$ (2D triangular lattice). The concentration $x = 0.6$ corresponds to a reduced value $\delta x = x/x_c - 1 = 0.2$. Using the correlation length exponent $\nu_p = \frac{4}{3}$,[12] we estimate the percolation correlation length to be $\xi_p \sim \delta x^{-\nu_p} a = 8.5a$, which is significantly larger than the in-plane magnetic correlation length $\xi_\perp = 2.8a$. Since for $h > 0.3$, the wavelength $\lambda_\perp$ is less than $\xi_\perp$, the excitations are insensitive to whether the system has long-range order and they could be affected by the percolation geometry.

(5) To better understand the zone-center excitations, we have studied the temperature dependence of the in-

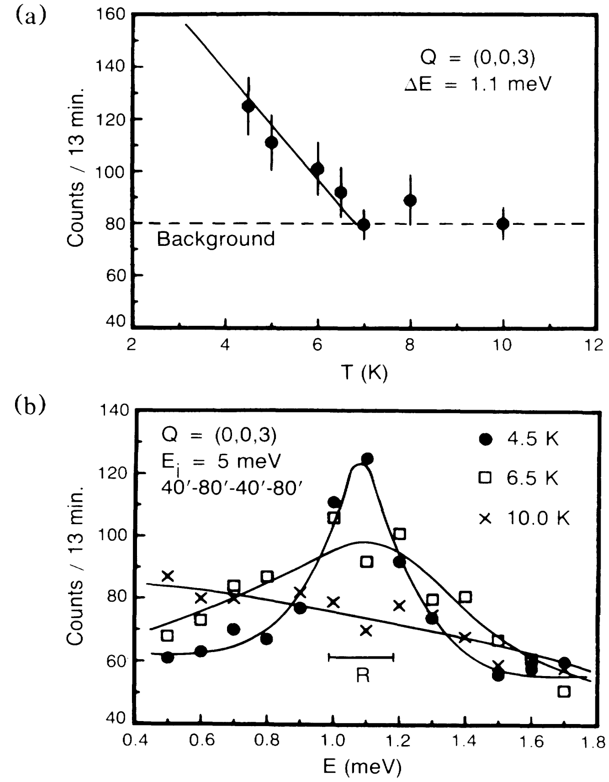

FIG. 4. (a) Temperature dependence of the zone-center spin-wave peak intensity. The background value includes some magnetic diffuse scattering. (b) Inelastic scans of the spin-wave peak above and below $2T_{sg}$.

elastic peak at $Q = (0,0,3)$. Figure 4(a) shows that this peak rises sharply below 7.0 K, which is approximately $2T_{sg}$. Figure 4(b) shows three scans at representative temperatures: 10 K where there is no peak, 6.5 K where there is a broad peak, and 4.5 K where the peak has become resolution limited. We note that the last temperature is significantly above the transition ($T_{sg} = 3.4$ K).

Let us now present a simple interpretation of the long-wavelength spin waves in $Fe_{0.4}Mg_{0.6}Cl_2$. The value of E_g suggests that these excitations exist because of the uniaxial single-ion anisotropy. In the spin Hamiltonian,[6] this is represented by a $-DS_\parallel^2$ term, which can also be written as $-\mathbf{H}_A \cdot \mathbf{S}_\parallel$, where $\mathbf{H}_A \equiv D\mathbf{S}_\parallel$ is the anisotropy field. If $\mathbf{S}_\parallel$ flips very slowly compared to the transverse fluctuations, then $\mathbf{H}_A$ is effectively static and the $\mathbf{S}_\perp$ at every site can precess around it with a Larmor frequency $\omega_L = g\mu_B H_A/\hbar$, where $g\mu_B$ is the longitudinal moment of the Fe^{2+} ion. For a normal ferromagnet, the local field at every site points in the same direction and all the spins can precess in phase in the long-wavelength limit. Here, however, because $\mathbf{H}_A$ points up and down randomly according to the direction of $\mathbf{S}_\parallel$ and has a correlation length ξ, we expect the $\mathbf{S}_\perp$ precessions to be also correlated over a distance ξ. In other words, we can view the excitations in real space as random precessions of spin clusters around the easy axis. Although the cluster size varies, the average size is ξ. Since there is no spatial periodicity in these cluster precessions, they correspond to a branch of spin-wave modes in the reciprocal space with wave vector in the range $0 \leq Q \lesssim \pi/\xi$ and the frequencies of which are all given by ω_L, as the lower branch in Fig. 3 shows.[13] The stiffness constant is small because the average exchange between spin clusters is close to zero. We should also remark that the randomness in the phase of the precession is due to the fact that we are making observations on the two-spin correlation functions, such as $\langle S_x(0,0)S_x(r,t)\rangle$, which are inappropriate for describing the fluctuations of the spin-glass order parameter. It is possible that phase coherence of the excitations exists in the four-spin correlation functions, such as $\langle S_x^2(0,0)S_x^2(r,t)\rangle$.

From the above discussion, we can identify two main criteria for the existence of long-wavelength spin waves. First, $\mathbf{S}_{\parallel}$ must fluctuate very slowly compared to $\mathbf{S}_{\perp}$. According to a recent theoretical study by Ogielski,[14] the longitudinal relaxation time for a three-dimensional Ising spin-glass diverges as $\tau \sim (T/T_{sg}-1)^{-z\nu}$ near the transition, where z is the dynamic exponent and ν is the correlation length exponent. The best estimate for $z\nu$ is 7.9 ± 1. Hence, τ increases extremely rapidly when the reduced temperature is less than unity, i.e., when $T < 2T_{sg}$. This explains why the zone-center spin wave sets in below $2T_{sg}$ in our experiment. Another important criterion is that the transverse spin component should not be frozen; otherwise the excitations might be overdamped. Most recently, Bray[15] has analyzed this problem using the infinite-range model with uniaxial anisotropy and he reached the same conclusion. Evidently, this requirement is satisfied in $Fe_{0.4}Mg_{0.6}Cl_2$ because the $\chi'_{\perp}$ data in Fig. 1 show no frequency dependence. However, we note that a recent specific-heat study on $Eu_xSr_{1-x}As_3$ (another uniaxial spin-glass system) has suggested the existence of an energy gap in the density of states,[16] consistent with the present work, but the susceptibility data on that system also indicated separate freezing transitions for $\mathbf{S}_{\parallel}$ and $\mathbf{S}_{\perp}$. Therefore, the effects of frozen transverse moments on the spin waves should require further studies.

In summary, we have found that long-wavelength ($\lambda > 2\xi$) spin waves can exist in a spin-glass system with strong uniaxial anisotropy. We have given a simple explanation, namely, the precession of spin clusters, to account for the temperature and wave-vector dependences of these excitations. We have also shown that for short wavelengths ($\lambda < \xi$) the excitations are similar to other dilute magnets and these might be affected by the percolation geometry. (Whether these should be described as fractons is unclear because the wavelengths involved are not much larger than the lattice constant.) In the intermediate-wavelength regime, $\xi < \lambda < 2\xi$, there appears to be an abrupt crossover which cannot be described in simple terms.

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