Field-Induced Dispersion in the Amorphous Antiferromagnet

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A magnetic-field–induced exchange stiffness and dispersion have been observed in the spin-flip Raman spectra of localized donor electrons in n-type CdS, a model amorphous antiferromagnet. At high applied fields, there is a q-dependent shift in the peak position of the Stokes line. With increasing field, the peak and spectral weight of the excitations move to the low-energy side of the Zeeman energy as q increases. This negative dispersion is consistent with the antiferromagnetic coupling.

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The nature of the excitations in spatially random systems of interacting magnetic spins and the possible occurrence of long-wavelength modes are problems of considerable interest.[1] Bound shallow donors in a semiconductor form a typical example of a class of such systems, the amorphous anitferromagnets. Here the exchange interaction, $J_{ij}\vec{S}_i\cdot\vec{S}_j$, is short range and hydrogeniclike, with $J_{ij}>0$, i.e., antiferromagnetic, and given by[2]

$$J_{ij}(r_{ij})=1.636 r_{ij}^{5/2}\exp(-r_{ij}/a_0)(2R^*), \quad (1)$$

where a_0 is the donor Bohr radius (~25 Å in CdS), r_{ij} is the donor separation, and R^* is the donor Rydberg. The term amorphous is used because of the random donor position on lattice sites and the smallness of the lattice constant compared with a_0.

The tendency of any two donors to align antiparallel is "frustrated" by the random placement of neighboring donors so that any anticipated ordering in the system might be analogous to that of a spin-glass[3,4] with spins frozen in all orientations and a net moment of zero. In spin-glass systems, the hallmark of this ordering is an abrupt change in the susceptibility, χ,[4] at the ordering temperature T_g, which occurs in the vicinity of the median exchange.

Kummer and co-workers[5,6] have measured χ vs T for In:CdS samples with ~10^{17} In donors/cm^3, finding no peak or discontinuity in χ down to 35 mK. For such concentrations Walstedt *et al.*[5] estimate that the median J_{ij} of closest neighbors is ~10 K. They also showed that at the lowest temperatures there exists an infinite connected network of bonds with $J_{ij}\gg k_BT$. The absence of long-range order under these conditions may be due to the effective weakening of interaction between spins, already tightly coupled to other spins, or to the presence of large quantum fluctuations in $S=\frac{1}{2}$ systems.[5,7] Similar failure to order has been reported for P:Si.[8] In spite of the lack of long-range ordering, one is still led to ask about the nature of the excitations in this system at temperatures that are low compared to typical J_{ij}'s. To this end we have studied the dynamic response in n-type CdS by observing the scattering function $S(q,\omega)$ by spin-flip Raman light scattering (SFRS). Previously, SFRS was studied in n-CdS[9,10] but with an instrumental resolution that was far too low to see any effects of the exchange. Later, high-resolution SFRS in CdS revealed[11] for the first time for *bound* donors spin diffusion resulting from the exchange, but the fields used were still too low to observe any dispersive effects.

In SFRS, an incident photon at frequency Ω_i and wave vector $\vec{k}_i$ is scattered to a photon $\Omega_s,\vec{k}_s$ with an accompanying spin-flip excitation $\vec{q},\omega_0$ where $\vec{q}=\vec{k}_s-\vec{k}_i$ and $\omega_0=\Omega_i-\Omega_s$. In general, the intensity of the scattered light is proportional to the scattering function $S_\perp(q,\omega)=(1/2\pi)\int_{-\infty}^{\infty}\langle\sigma_q^-(t)\times\sigma_{-q}^+(0)\rangle e^{i\omega t}\,dt$, where σ_q is the qth Fourier component of the spin density. For noninteracting spins, the scattered light has two peaks, the Stokes and anti-Stokes lines at $\Omega_i\mp\omega_0$, where $\omega_0=g\mu_BH_0/\hbar$ and H_0 is the applied field. The linewidths would be determined by a spin lifetime T_1 or T_2 or an inhomogeneous broadening T_2^*, and would show no q dependence. With exchange present, the peak position may vary with q, as well as the linewidth.

The In:CdS crystals and superconducting magnet were immersed in superfluid helium pumped to 2 K. Our experimental configuration would only allow a temperature variation between 1.8 and 2.1 K. The output of an argon-ion laser tuned to 4880 or 4965 Å was focused onto the crystal. Scattered light was frequency analyzed with an electronically stabilized triple-pass Fabry-Perot interferometer. The spectra were recorded on

a multichannel analyzer and then processed by computer. Figure 1(a) shows the raw data for the Stokes lines for a field of 52 kG at scattering angles of $\theta = 6°$ ($q \sim 0$ Å^{-1}) and $\theta = 153°$ ($q = 0.0075$ Å^{-1}) for a sample with donor concentration $n_d \sim 2.3\times10^{17}/\text{cm}^3$, at least a factor of 4 below any insulator-metal transition. Although our largest $q = 0.0075$ Å^{-1} is small compared with neutron probes, it samples a good part of the "zone," since at these concentrations the mean separation, $\langle r_{ij}\rangle$, between donors gives $q_{153°}\langle r_{ij}\rangle \simeq 0.4$. Figure 1(b) shows the $S(q,\omega)$ corresponding to Fig. 1(a), smoothed and corrected for the instrumental width. A difference, δ, in the peak positions of the line shapes, as well as a severe asymmetry of the $\theta = 153°$ line shape is evidence for dispersive effects in $S(q,\omega)$. The anti-Stokes line is a mirror image of the Stokes line about the position of the laser line for the same effective q,[12] but much reduced in intensity at the high fields. For the Fabry-Perot free spectral range used in Fig. 1(a), δ can be measured to an accuracy of 7.5 MHz (or $\sim 3\times10^{-4}$ meV!).

Figure 2 summarizes the contrasting behavior of $S(q,\omega)$ in high field. There is a residual linewidth at $q = 0$ which is even seen in the most dilute "pure" crystals where the donors are quite isolated and noninteracting. The data as presented include this intrinsic width, $1/T_2$. For $H_0 \to 0$ (the lower portion of Fig. 2) there is no perceptible shift of the line as q increases and it is

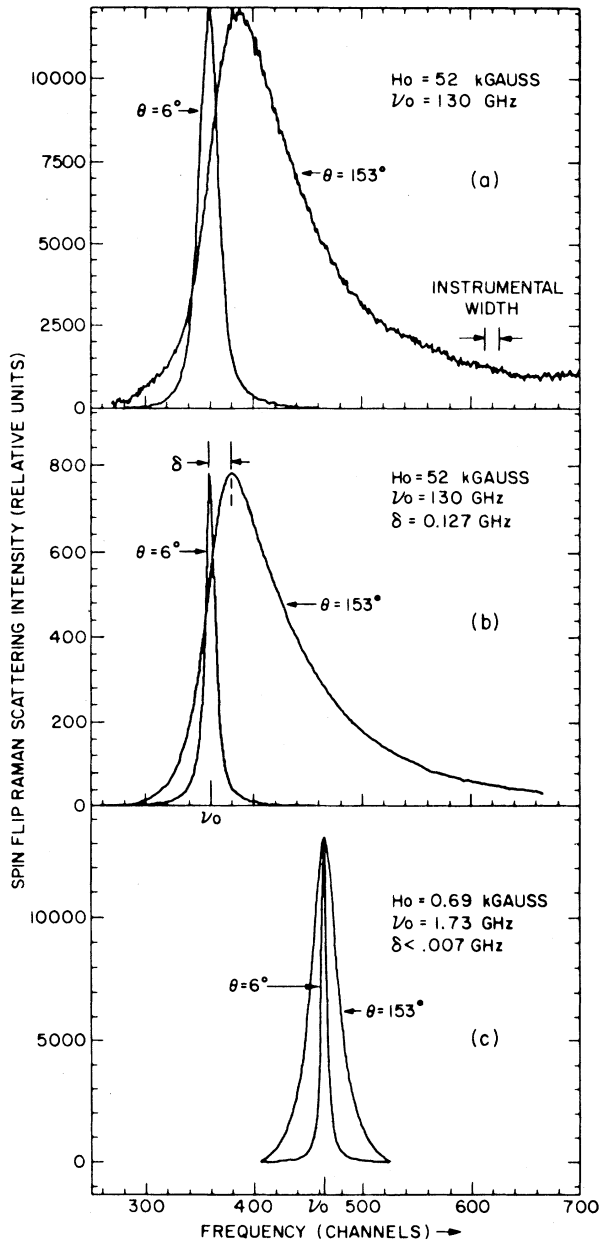

FIG. 1. SFRS line for n-CdS with concentration 2.3×10^{17} at 2 K. 1 channel = 6.7 MHz. (a) Raw data at high field for near-forward and backscattering angles: Stokes line. (b) Smoothed data with instrumental width removed. (c) Same as (b) except in low field.

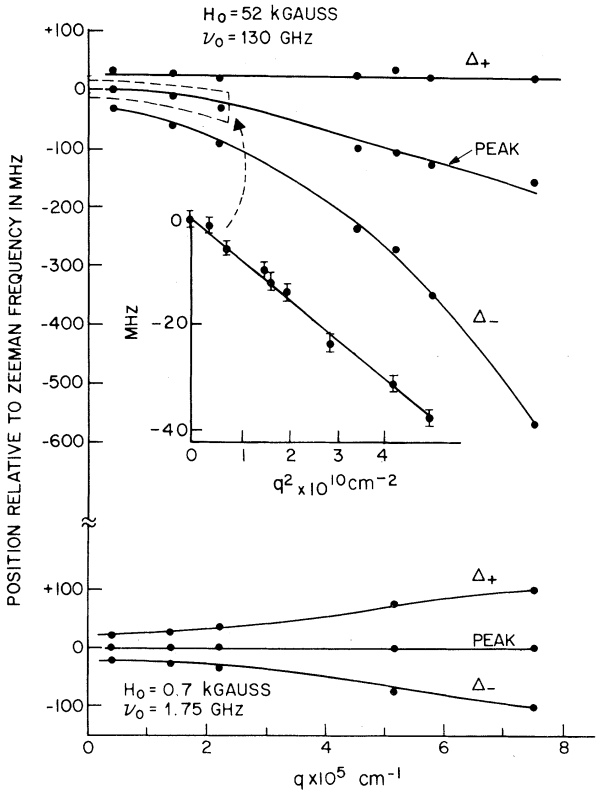

FIG. 2. Peak position and half-intensity points Δ_+ (upper) and Δ_- (lower) of the Stokes line for the sample in Fig. 1. Inset: low q, $H_0 = 52$ kG.

Lorentzian. The width varies as (Dq^2+1/T_2), where D is a spin-diffusion constant due to the exchange as described elsewhere.[11] If there is any dispersion of the excitation spectrum in this case it must be very small (< 7 MHz) over the full range of $\vec{q}$. In high fields, on the other hand, the whole spectral weight of the spin excitations moves towards *lower* energy (corresponding to increasing Stokes frequency) as $|\vec{q}|$ increases. At the same time the line broadens and becomes asymmetric. The broadening is predominantly on the low-energy side and the spectral weight above ω_0 becomes steadily less. The shift varies initially as $|\vec{q}|^2$.

Figure 3 is a plot of $S(q,\omega)$ for fixed q ($\theta = 153°$) as a function of H_0 and shows the increased stiffness and dispersion with increasing H_0. These effects are consistent with the assumption that, in high fields, when the individual spins have acquired an average moment along H_0, these induced moments interact antiferromagnetically. Turning a spin over in H_0 costs magnetic energy $\hbar\omega_0$, but since the reversed spin is now oppositely oriented to the average direction of the other spins, the exchange energy is lowered as a result of the antiferromagnetic coupling. As q and the field-induced moment increase, the antiferromagnetic interactions become more effective, increasingly shifting the excitations below ω_0. If the spins were ferromagnetically coupled, the dispersion would rise above ω_0. The system resembles a regular antiferromagnet in a field in its paramagnetic phase [which occurs at any field above its ordering temperature, T_N, or above some critical field $H_c(T)$ below T_N] which has a dispersion relation with ω decreasing as q increases.[13] However, because of the randomness in our system, a given $\vec{q}$ will correspond to a spectrum of excitations at different frequencies with some component at $\vec{q}$. As $\vec{q}$ increases, the spread in frequency of these excitations increases and most of them will be below their most probable value, resulting in the observed increased width and asymmetry.

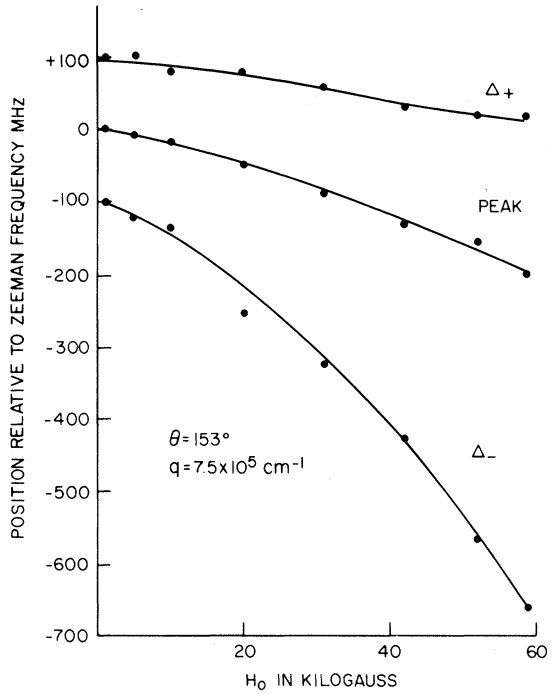

FIG. 3. Peak position and half-intensity points Δ_+ and Δ_- as a function of field for fixed q. Sample and temperature are the same as in Figs. 1 and 2.

In addition to the data presented above for a sample with $n_d \sim 2.3\times10^{17}$ data were also taken on samples with $n_d \sim 8\times10^{16}$ and $n_d \sim 8\times10^{17}$. The shifts were approximately 3 times smaller in the more dilute sample and 10 times greater in the more concentrated sample, consistent with a very rapid increase in the J's with concentration.

In a regular antiferromagnet or ferromagnet in its paramagnetic region in a field H_0, one might anticipate that the field-induced dispersion would go as $\Delta\nu \sim J\langle S_z\rangle a^2q^2$, where a is the distance between spins and $\langle S_z\rangle$ is the expectation value of the spin along H_0. A similar expression should apply to a random system with appropriate values which reflect the distributions of J_{ij}, $\langle S_{iz}\rangle$, and r_{ij}. Calculation of $S_\perp(q,\omega)$ for this random system is obviously more difficult than that of a static quantity such as the susceptibility. Even for the latter, progress has only been made by computer simulation.[5,8] Decoupling, at various levels, of the relevant Green's function equations leaves an intractable linear problem with matrix elements involving unknown static averages. It is, however, clear that the shift and broadening predicted will indeed go as q^2. One approximate solution, based on a first-order decoupling, is equivalent to treating the system as one of fixed, field-induced moments interacting as $J_{ij}\langle S_i{}^z\rangle\langle S_j{}^z\rangle$. The resulting expression may then be evaluated in the nearest-neighbor pair approximation. Even though this procedure eliminates a large number of spins bound in singlets, it gives a coefficient for q^2 which is about 10 times greater than the observed value of $\sim 9\times10^{-4}$ Hz cm^2. This overestimate may reflect the neglect of fluctuations in the $S_i{}^z$ as well as effective reduction of these antiferromagnetic exchange couplings as they are

renormalized by the interaction among more strongly interacting spins.[5,8]

In the reentrant spin-glass Fe:Cr, Shapiro *et al.*[14] may have observed the analog of the field-induced dispersion reported here for ferromagnetic coupling.

In conclusion, the amorphous antiferromagnet shows no exchange stiffness in the absence of a field but does show increasing exchange stiffness and resultant dispersion as magnetization is induced with increasing field. The high resolution of SFRS should find application in probing the dispersion of the excitations of other random systems including a variety of spin-glasses.

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Nonlinear Mixing of Bulk and Surface Magnetostatic Spin Waves

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The first discussion of the nonlinear mixing of bulk and surface magnetostatic spin waves is presented. The unusual property of these waves—that the frequency is dependent on the direction of propagation and not the magnitude of the wave vector—allows resonant generation of a new wave if the propagation directions of the two incident waves are properly chosen. In addition to the mixing of two bulk (or surface) waves to produce a new bulk (or surface) wave, the mixing of two bulk waves to produce a resonantly enhanced surface wave is discussed.

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Nonlinear mixing of bulk waves and of surface waves of various types has received a great deal of attention. For instance, the mixing of acoustic waves and the mixing of polaritons has a considerable literature.[1-3] In contrast, the topic discussed here—the nonlinear mixing of magnetostatic bulk and surface spin waves—has not been treated up to the present time, and shows some very interesting results.

Magnetostatic spin waves are long-wavelength modes which propagate in magnetic systems. The properties of these modes are governed, not by the short-range exchange interaction, but by macroscopic dipole fields set up by the motion of the spins precessing around the magnetic field. These dipole fields are calculated through the use of the magnetostatic form of Maxwell's equations. The properties of these magnetostatic spin waves have recently been extensively studied through the use of Brillouin scattering from ferro-