Spin Dynamics of a Model Singlet Ground-State System

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Inelastic neutron scattering has been used to study the spin dynamics of $LiTb_pY_{1-p}F_4$, an induced-moment ferromagnet in which the lowest crystal-field levels of the Tb^{3+} ions are two singlets. For $p=0.38$, magnetic excitons are observed which obey the dispersion relation given by mean-field theory. In addition, there is a resolution-limited peak at zero energy transfer which is not accounted for within conventional mean-field theory. For $p = 0.97$, this central peak is also observed, but no magnetic excitons are seen.

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One of the most important results of renormalization-group theory is that the critical behavior of pure uniaxial dipolar-coupled ferromagnets should obey mean-field theory with logarithmic corrections.[1] This was confirmed experimentally in studies of $LiTbF_4$.[2,3] More recently, there has been renewed interest in the behavior of dilute Ising dipolar-coupled systems, partly because of a conjectured spin-glass phase in the low-concentration limit.[4] (In a dipolar-coupled system, the percolation ideas[5] used to describe short-range–coupled systems are not applicable.) With this in mind, two groups[6] have performed studies of $LiTb_pY_{1-p}F_4$, in which Y acts as a random nonmagnetic dilutant in the magnetic Tb lattice. Unfortunately, a simple Ising description cannot be applied to $LiTb_pY_{1-p}F_4$. Firstly, the "Ising doublet" in this compound is actually a pair of nonmagnetic states split by 1.34 K,[7] so that magnetic moments must be induced by the interactions; the Hamiltonian is effectively an Ising Hamiltonian in a transverse field. Secondly, the hyperfine interactions are not negligible for small p. In this Letter, we present the results of a neutron scattering study of the dynamics of $LiTb_pY_{1-p}F_4$. We will show that although this compound is unsuitable for studying the critical properties of a simple Ising system, it is an unprecedentedly clean example of a singlet-singlet magnet.

Recall that the Hamiltonian for the Ising model in a transverse field (or equivalently, the singlet-singlet ground-state system) is

$$\mathcal{H} = -\Delta \sum S_i^{x} - 2 \sum_{i>j} \kappa_{ij} S_i^{z} S_j^{z}, \qquad (1)$$

where Δ corresponds to the splitting between the two singlets.[8] The ferromagnetic transition temperature T_c for this model is given within mean-field theory by the formula

$$\coth(\Delta/2kT_c) = \tilde{\kappa}(0)/\Delta = \eta, \qquad (2)$$

where $\tilde{\kappa}(\vec{Q}) = \sum_j \kappa_{ij} \exp i\vec{Q}\cdot(\vec{r}_i - \vec{r}_j)$ is the Fourier transform of the interaction κ_{ij} between spins. If we ignore the effects of randomness, $\tilde{\kappa}(0)$ for a dilute system scales linearly with the concentration p of magnetic ions, so that $\eta(p) = p\eta(1)$. Consequently, to determine T_c as a function of p, it is sufficient to know $T_c(p=1)$ and Δ. Using the values $\Delta = 1.34$ K, obtained for $LiTb_{0.01}Y_{0.99}F_4$ from ESR measurements,[7] and $T_c = 2.86$ K for pure $LiTbF_4$,[3] we obtain the solid curve shown in Fig. 1. The data points in the same figure represent the values of T_c given by susceptibility measurements. For p exceeding the threshold $p_c = 0.21$, these data are in good agreement with the curve derived from simple mean-field theory. However, ferromagnetism is still observed for $p < p_c$. This can be attributed to the large hyperfine interaction of the Tb^{3+} ion.[7,9] Here, we will not consider this point further; work in this area is in progress and will be discussed elsewhere.[10]

Apart from predicting the dependence of T_c on Δ and $\tilde{\kappa}(0)$, mean-field theory gives the excitation spectrum for the Hamiltonian (1).[8] For $T > T_c$, the magnetic excitons obey a dispersion relation

$$E(\vec{Q}) = \Delta(1 - \eta' \gamma_{\vec{Q}})^{1/2}, \qquad (3)$$

where $\gamma_{\vec{Q}} = \tilde{\kappa}(\vec{Q})/\tilde{\kappa}(0)$ and $\eta' = \eta \tanh(\Delta/2kT)$. On

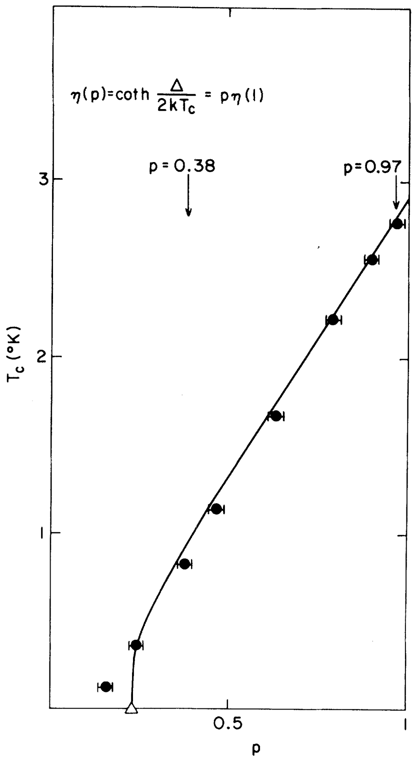

FIG. 1. Magnetic phase diagram of $LiTb_pY_{1-p}F_4$. Data (filled circles) are taken from Ref. 6.

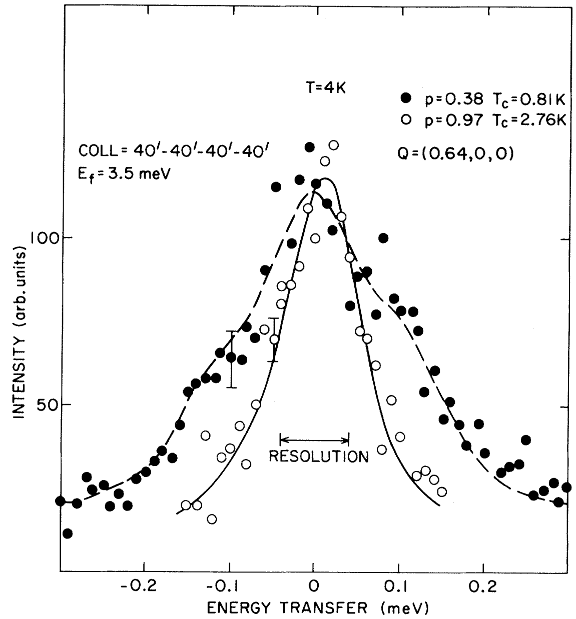

FIG. 2. Constant-$\vec{Q}$ energy scans for $p = 0.38$ and 0.97 in paramagnetic phase. Solid and dashed lines represent theoretical line shapes fitted to the data (see text).

the other hand, for $T \ll T_c$,

$$E(\vec{Q}) = \Delta(\eta^2 - \gamma_{\vec{Q}})^{1/2}. \tag{4}$$

Note that the excitation spectrum has a gap for $T \neq T_c$, while at $T = T_c$, $\eta' = 1$ and the system undergoes a soft-mode transition with $E(\vec{Q}) \sim |\vec{Q}|$ for small $\vec{Q}$.

Our neutron-scattering experiments were carried out at the cold source of the Brookhaven National Laboratory high-flux beam reactor. We performed inelastic scans by varying the incident neutron energy while keeping the final energy fixed at $E_f = 3.5$ meV. Single crystals of $LiTb_pY_{1-p}F_4$ with $p = 0.38 \pm 0.02$ and 0.97 ± 0.02 were mounted in pumped ^3He or pumped ^4He cryostats so that the (010) zone coincided with the scattering plane. The data to be described below were all collected for momentum transfers $\vec{Q} = (Q,0,0)$, where Q is expressed in units of $a^* = 2\pi/a$ and $a = 5.20$ Å is the in-plane (perpendicular to the Ising spin axis) lattice constant.

Figure 2 shows constant $\vec{Q}_0 = (0.64,0,0)$ inelastic spectra obtained at the same temperature for the two samples in their paramagnetic phases. We chose this particular value of $\vec{Q}$ because $\bar{\kappa}(\vec{Q})$ vanishes at $\vec{Q} \approx \vec{Q}_0$, which means that the excitonic energy $E(\vec{Q}_0)$ should provide a direct measure of the splitting Δ [see Eq. (3)]. Contrary to naive expectations of a spectrum with substantial intensity at energy transfers $\pm\Delta$, there is negligible true inelastic scattering for the nearly pure material. This is true not only of the spectrum shown, but also of data taken at other momentum transfers and temperatures, both above and below T_c. The elastic incoherent background accounts for less that 25% of the peak intensity in Fig. 2; most of the resolution-limited elastic scattering observed for $p = 0.97$ is therefore of magnetic origin.

The results for $p = 0.38$ are qualitively different from those obtained for $p = 0.97$: Here there are obvious excitonic sidebands in addition to a quasielastic central peak (see Fig. 2). To analyze our data, we assume a theoretical cross section similar to that used for soft-mode problems connected with structural phase transitions[11]: the sum of a Lorentzian central peak and a damped harmonic oscillator term,

$$\frac{d^2\sigma}{d\Omega\, d\epsilon} = \frac{\kappa_f}{\kappa_i}(1 - e^{-\beta\epsilon})^{-1}\left\{A_H \frac{1}{\pi}\frac{\epsilon\Gamma}{[\epsilon^2 - (\hbar\omega)^2]^2 + (\epsilon\Gamma)^2} + A_L \frac{1}{\pi}\frac{\gamma\epsilon}{\epsilon^2 + \gamma^2}\right\}. \tag{5}$$

Expression (5) is convolved with the experimental resolution function and the parameters $\hbar\omega_0$, Γ, A_L, and A_H are varied to minimize the deviation from the experimental data. The Lorentzian half-width γ is fixed at 0.01 meV, a value significantly below the experimental resolution of 0.04 meV (half width at half maximum). For $p = 0.97$, we find that the Lorentzian term by itself (solid line in Fig. 2) gives an excellent account of the data. For $p = 0.38$, our fitting procedure gives an excitonic energy $\hbar\omega_0 = 1.5 \pm 0.1$ K, in reasonable agreement with the $\Delta = 1.34$ K obtained from ESR,[7] and a substantial damping coefficient $\Gamma = 1.3 \pm 0.3$ K.

Figure 3 shows spectra collected for the $p = 0.38$ material at a temperature $T = 0.35$ K, well below its Curie point $T_C = 0.8$ K. Comparison with Fig. 2 shows that the excitonic contribution here is sharper than in the paramagnetic phase at $T = 4$ K. However, there is still a large quasi-elastic component, which grows considerably as $\vec{Q}$ is reduced. Following the procedure described above, we have performed fits to the data; the solid curves in Fig. 3 represent the cross section calculated with the final values of the parameters. The resulting energies $\hbar\omega_0$ are consistent with the theoretical form (4), evaluated at $T = 0$ with the dipolar $\kappa(\vec{Q})$ given by Holmes, Als-Nielsen, and Guggenheim,[3] and $\eta = \coth(\Delta/2kT_c)$ (see inset in Fig. 3). As also predicted by the effective-boson theory,[8] the amplitude factor A_H in Eq. (5) is constant to within experimental error.

Mean-field theory and its generalizations[12] suggest that the dynamic structure factor $S(\vec{Q}, \epsilon)$ for singlet ground-state systems is dominated by (possibly overdamped) excitonic peaks centered at finite energies. This is clearly not what we see in $LiTb_pY_{1-p}F_4$; for $p = 0.97$ ($\eta = 3.7$), excitonic sidebands are absent, and for $p = 0.38$, they co-

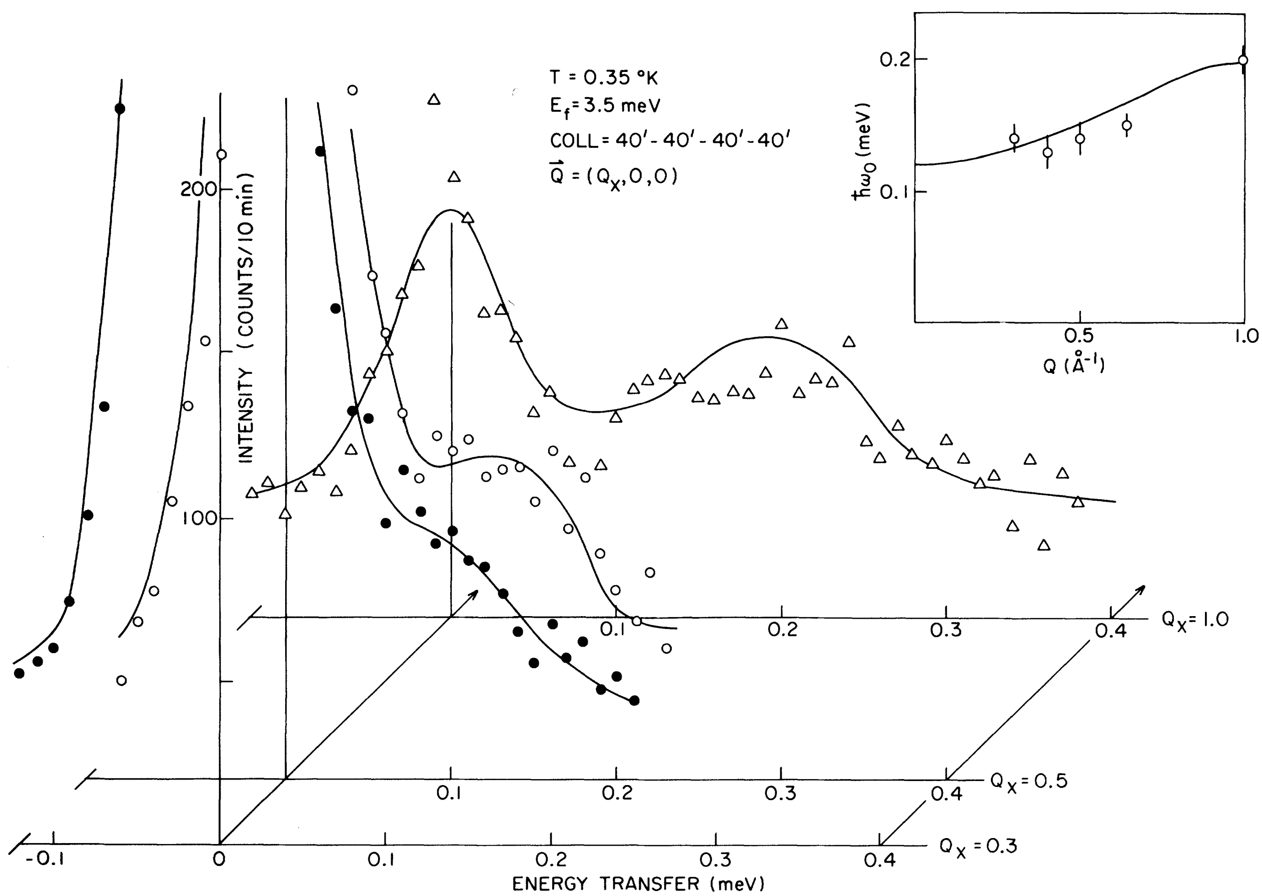

FIG. 3. Constant-$\vec{Q}$ energy scans for $p = 0.38$ in ferromagnetic phase. Solid lines represent theoretical line shapes fitted to the data (see text). Inset compares theoretical dispersion curve for excitons (solid line) and fitted values of the exciton energies.

exist with a substantial resolution-limited peak at $E=0$. Similar effects have been seen in other singlet ground-state systems, but could usually be attributed to excitations within higher-lying crystal-field multiplets.[8,13] Such an explanation cannot hold for $LiTb_pY_{1-p}F_4$, because the next crystal-field level beyond the two singlets is at ~ 155 K,[14] which is much larger than Δ, T_c, or the temperatures T at which our experiments were performed. Therefore, it is reasonable to surmise that the central peak in $LiTb_pY_{1-p}F_4$ arises from long-lived clusters of ferromagnetically aligned spins. Indeed, this is what one expects as $\Delta/\tilde{\kappa}(0) \to 0$, when the model is a pure Ising Hamiltonian. Correspondingly, as p is increased, we see a crossover between two different dynamical regimes. At small p $[\Delta > \tilde{\kappa}(0)]$ the spin fluctuations are predominantly the excitons of standard singlet ground-state theory, while at larger p $[\tilde{\kappa}(0) \gg \Delta]$ they correspond to ferromagnetic microdomains which might be particularly stable in this uniaxial dipolar-coupled material.

The dilute insulating series $LiTb_pY_{1-p}F_4$ is a nearly ideal singlet-singlet system, which means that it is *not* ideal for studying the effects of randomness on dipolar-coupled Ising systems. Indeed, mean-field theory for singlet-singlet magnets adequately describes the dependence of T_c on p, and the excitation spectrum for $p=0.38$. However, further work is required to understand the crossover from a dynamical response dominated by magnetic excitons to one dominated by long-lived ferromagnetic clusters.

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