Magnetic Exciton Dispersion in Praseodymium

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Measurements of the dispersion of magnetic excitons have been made in a single crystal of praseodymium metal using inelastic neutron scattering. A preliminary analysis of the data yields the first detailed information about the exchange interactions and the crystal field splittings in the light rare-earth metals.

The elementary magnetic excitations in the light rare earths are fundamentally different from the magnons in the heavy rare earths which have been extensively studied in recent years by inelastic neutron scattering.[1] For the heavy rare earths the exchange forces are considerably larger than the crystal field interactions, and the crystal field is manifested as a source of anisotropy which determines the spin direction in the ordered state and gives rise to the energy gap in the measured magnon spectra. In the light rare earths the effective exchange is weaker and is comparable in magnitude to the crystal field forces.[2] Consequently the ordered moment is often reduced from the free-ion value $g\mu_B J$, and indeed it is possible to have complete suppression of magnetic order. In this case the magnetic excitations have the nature of magnetic excitons in which a transition between single-ion crystal field states is propagated through the crystal by the exchange interactions between neighboring ions.[3-6] We report here the first measurements of the dispersion of these excitations in the light rare-earth metal praseodymium.

Praseodymium has the double hexagonal close-packed crystal structure in which there is a stacking sequence $ABAC$ along the crystal c axis. The immediate surroundings at the A sites have approximately cubic symmetry, while the B and C layers both have local hexagonal symmetry. Bleaney[7] made detailed calculations of the crystal field potential and proposed a level scheme which accounted reasonably well for the Schottky anomaly in the heat capacity.[8] The crystal field splittings at the cubic sites (A layers) are different from those at the hexagonal sites (B or C layers) because of the different symmetry of their surroundings. Recent susceptibility[9] and neutron diffraction[10] measurements on a single crystal of Pr suggest[11] that Bleaney's level scheme is incorrect since it predicts the wrong sign for the magnetic anisotropy. However, there is little doubt that the ground state at both sites is a nonmagnetic singlet state. In this case no magnetic order is possible at low temperatures unless the exchange interaction exceeds a certain critical value.[12] Although neutron diffraction measurements[13,14] have indicated that polycrystalline Pr may be ordered antiferromagnetically at 4.2°K, no trace of long-range magnetic order has been detected in recent experiments on single crystals at low temperatures.[10,14]

The present measurements were made by inelastic neutron scattering from a small single crystal of Pr using a triple-axis spectrometer situated at the DR3 reactor at Risö. The dispersion relations measured with the specimen held at 4.2°K are shown in Fig. 1(a). For each of the symmetry directions investigated these have three branches. The single branch lying between 8 and 9 meV (open squares) can be assigned to a mode propagating on the cubic sites since these lie on a simple hexagonal lattice with one atom per unit cell. The two lower branches (closed circles and squares) have a larger dispersion and correspond to a mode propagating on the hexagonal sites. There are two branches in this case because the hexagonal sites lie on a hexagonal close-packed (hcp) lattice. From the measured inelastic structure factors, the upper of these two branches (closed circles) has been identified as the "acoustic" mode, while the lower is the "optic" mode. This indicates that the exchange interactions between B and C layers, which give rise to the Davydov splitting between the modes, are antiferromagnetic. The optic exciton between Γ and K shows a splitting which might be due to an interaction with a transverse acoustic phonon. A splitting is also observed for the acoustic exciton mode between Γ and M. In addition there is an extra mode (open circles) lying below the optic exciton. The origin of these two features is not yet understood.

The optic exciton at Γ displays a soft-mode behavior in that its energy is considerably lower at 4.2°K than at 18°K [Fig. 1(a)]. This is in qualitative agreement with Wang and Cooper's predictions[5] concerning the temperature dependence of

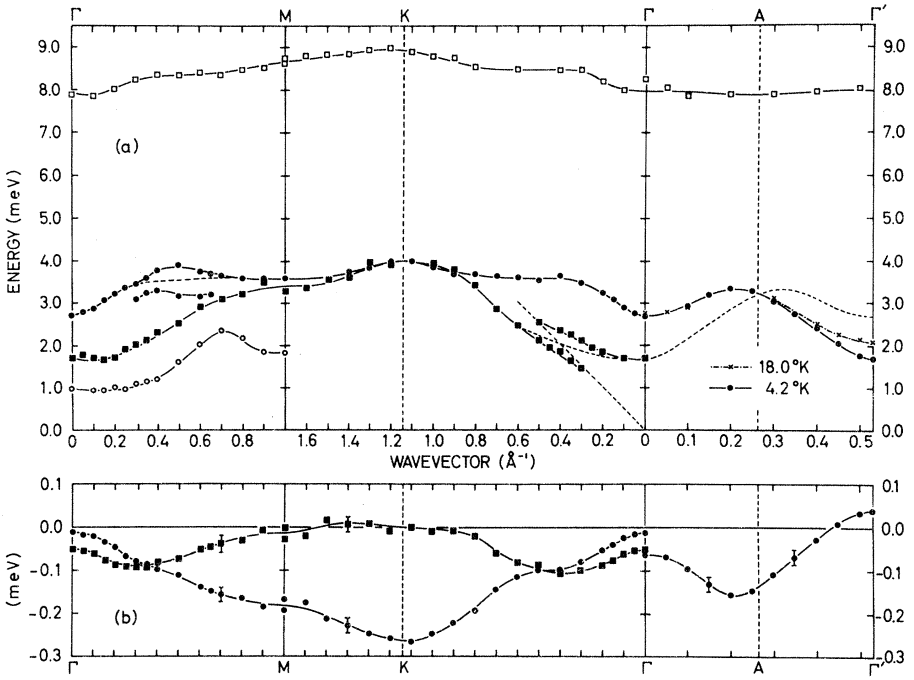

FIG. 1. (a) Dispersion relations for magnetic excitons in praseodymium at 4.2°K (Ref. 15). Measurements of the c-axis dispersion of the lower mode at 18°K are also shown. (b) $\mathcal{J}(q)$ (closed circles) and $\mathcal{J}'(q)$ (closed squares) for the hexagonal sites, deduced from Eq. (2) using the value of Δ derived from the data in Fig. 2.

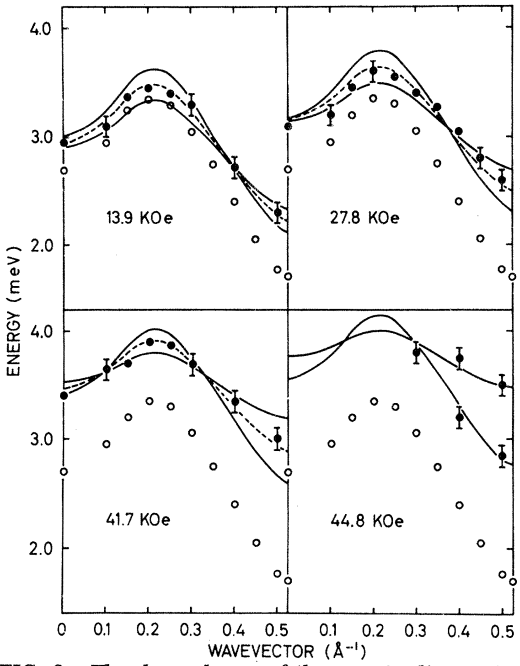

FIG. 2. The dependence of the c-axis dispersion of the lower mode on external magnetic field applied normal to the c axis. The solid curves are fits of the dispersion relations given in Eqs. (3) and (4). Where the splitting of the mode has not been resolved, the mean of the two energies (dashed curves) has been fitted. The zero-field data are included for comparison (open circles).

the exciton dispersion in induced-moment systems. The instability of this mode would lead to an antiferromagnetically ordered structure with a modulation vector close to a (001) reciprocal lattice vector. It is interesting to note that the measurements on polycrystalline Pr suggested magnetic ordering with this periodicity.

Although the neutron groups measured at 18°K were perfectly well defined, preliminary measurements at 30°K suggest that the lifetime of the excitations is very short at this temperature. The observed energy broadening of the excitons could originate either in exciton-exciton interactions or in spin-lattice or conduction-electron relaxation. Further measurements are planned to examine these possibilities.

We have also investigated the effect of a magnetic field on the c-axis dispersion of the lower mode. The field was applied perpendicular to the c axis along a b direction. The measurements (Fig. 2) show an upward shift of the mode and a pronounced change in the shape of the dispersion relations with increasing field. A splitting of the mode is just resolvable at the highest applied field.

In the light of a recent analysis[11] of the single-crystal susceptibility results we can assign the wave functions involved in the transition on the

hexagonal sites as

$|g\rangle = |m_J = 0\rangle$, ground state (singlet);

$|e\rangle = |m_J = \pm 1\rangle$, excited state (doublet).

The transition between these states is then the only allowed transition on the hexagonal sites at low temperatures. The assignments for the cubic sites are less certain at present. Following Grover's[4] approach we can calculate the dispersion relations for the excitations using the Bogoliubov approximation. This is valid in the limit of low temperatures and small dispersion. For the hexagonal sites the Hamiltonian may be written

$$\mathcal{H} = V_c - \sum_{i>i'} \mathcal{J}_{ii'} \vec{J}_i \cdot \vec{J}_{i'} - \sum_{j>j'} \mathcal{J}_{jj'} \vec{J}_j \cdot \vec{J}_{j'} - \sum_{i>j} \mathcal{J}_{ij}' \vec{J}_i \cdot \vec{J}_j, \quad (1)$$

where V_c is the crystal field potential and the indices i and j refer to atoms on the two sublattices of the hcp lattice. We find two doubly degenerate branches with energies

$$E_q = \{\Delta^2 - \alpha^2 \Delta[\mathcal{J}(q) \pm \mathcal{J}'(q)]\}^{1/2}, \quad (2)$$

where $\mathcal{J}(q)$ and $\mathcal{J}'(q)$ are the Fourier transforms of the exchange integrals $\mathcal{J}_{mn}$ and $\mathcal{J}_{mn}'$, respectively. Δ is the crystal field splitting and α is the matrix element of J^+ or J^- between $|g\rangle$ and $|e\rangle$. Note that Δ cannot be directly determined from the experimental data. However, the measurements in an applied field give additional information. For a field H applied perpendicular to the c axis it can be shown[11] that the splitting of the mode is small up to moderately large fields in agreement with observation. The dispersion relations for an *integral* field $H_{\rm int}$ are then (using the double-zone scheme for the c-axis dispersion)

$$E_q^{(1)}(H) = \left[\frac{W + \alpha^2 h^2 \Delta \lambda(0)}{W^2}\right]\left[\frac{1 - \Delta^3 \lambda(q)}{W^3 + \alpha^2 h^2 \Delta \lambda(0)}\right]^{1/2} \quad (3)$$

and

$$E_q^{(2)}(H) = \tfrac{1}{2}(\Delta + W)\left\{1 - \frac{\Delta}{W}\lambda(q)\left[1 + \frac{2}{\alpha^2}\frac{W - \Delta}{W + \Delta}\right]\right\}^{1/2}, \quad (4)$$

where

$$h = g\mu_B H_{\rm int}, \quad W = (\Delta^2 + 2\alpha^2 h^2)^{1/2},$$

and

$$\lambda(q) = 1 - E_q^2(0)/\Delta^2.$$

$E_q(0)$ is the measured energy in zero field at wave vector q. The solid lines in Fig. 2 represent the best fit of these relations to the experimental data with Δ and $H_{\rm int}$ as disposable parameters for each value of the external field. The value of Δ corresponding to the best fit is 2.14 $\pm$ 0.10 meV, while the values obtained for the internal field are reasonably consistent with those estimated from the measured single-crystal susceptibility.[9] The fitted curves are seen to give a good account of both the change in shape of the dispersion relations with field and of the splitting between the modes where this has been resolved. The value for Δ, obtained in this way, has been used together with Eq. (2) to determine the Fourier-transformed exchange parameters $\mathcal{J}(q)$, $\mathcal{J}'(q)$ for the hexagonal sites; these are shown in Fig. 1(b). The observed splittings of two of the branches of the dispersion relations, referred to above, have been ignored in this calculation. The calculated Fourier-transformed exchange parameters show several interesting features. The maximum at Γ' indicates the tendency towards antiferromagnetic ordering, as discussed above. Further, the shape of $J(q)$ in the directions $\Gamma K M \Gamma$ closely reflects the shape of the dispersion relations of the upper mode [open squares, Fig. 1(a)] which propagates on the cubic sites. The results in Fig. 1(b) will be analyzed to yield interplanar exchange parameters.

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[1]For a recent review of these experiments on heavy rare earths see A. R. Mackintosh and H. Bjerrum Møller, in "Magnetic Properties of Rare Earth Metals," edited by R. J. Elliott (to be published), Chap. 5.

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[15]The notation $\Gamma A \Gamma'$ refers to the Brillouin zone (BZ) for the hexagonal sites; for the cubic sites the dimension of the BZ in the c direction is doubled, so that the point labeled Γ' corresponds to A of the BZ for the cubic sites.