INELASTIC SCATTERING OF NEUTRONS BY SPIN WAVES IN TERBIUM

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This Letter describes measurements of the spin-wave dispersion relations for magnons propagating in symmetry directions in ferromagnetic Tb. It is the first experiment to give detailed information on the magnetic excitations in the heavy rare earths. Tb was chosen for these measurements because it is one of the few rare-earth metals which does not have a very high thermal-neutron capture cross section, so that inelastic neutron scattering experiments can give satisfactory information on the magnon dispersion relations.

The measurements were carried out on the triple-axis spectrometer situated at the DR-3 reactor, using the constant- \overline{q} method. The sample was a single crystal of Tb, 5 cm long and 6 mm in diameter. It was mounted in a cryostat, the temperature of which could be varied between 90°K and room temperature. The measured magnon dispersion relations for the *a*, *b*, and *c* directions are shown in Fig. 1. Most of the neutron groups observed were well

defined with a peak intensity of typically 10 counts/min and a background of 2 counts/min. From the accuracy in determining the center of the neutron groups and the spread of these, obtained in different scans, the uncertainty of the measured magnon energies is estimated to be ± 0.1 meV. In some cases it was verified that the neutron groups were due to magnon scattering by applying a magnetic field in the direction of the scattering vector; since this increased the intensity of the neutron groups, they could only be due to magnon-scattered neutrons.

An interesting feature of the *c*-axis dispersion curve is the rapid change of magnon energy with wave vector near the q value corresponding to the periodicity of the helically ordered magnetic phase of Tb. This periodicity is believed to be determined by a prominent Fermi-surface dimension¹ and the kink in our results may be a magnon Kohn anomaly of the type discussed by Woll and Nettel.² Using a



FIG. 1. Experimental magnon dispersion curve at 90°K.

free-electron model, they predict anomalies of only a few percent, but the actual form of the Fermi surface is probably such as to enhance the effect.

There are no other obvious anomalies in our results. From measurements of the sound velocity³ in Tb, we know that the magnon- and phonon-dispersion curves cross each other. The magnon-phonon interaction, which has recently been observed in uranium dioxide,⁴ must therefore be so small in Tb that its effect on the magnon dispersion curve is less than a few percent.

The measured magnon dispersion curves can be analyzed in terms of the Fourier-inverted exchange parameters

$$J(\mathbf{\hat{q}}) = \sum_{\mathbf{\hat{R}}_{j}} J(\mathbf{\hat{R}}_{j}) \exp(i\mathbf{\hat{q}} \cdot \mathbf{\hat{R}}_{j}),$$
$$J'(\mathbf{\hat{q}}) = \sum_{\mathbf{\hat{R}}_{j'}} J(\mathbf{\hat{R}}_{j'}) \exp(i\mathbf{\hat{q}} \cdot \mathbf{\hat{R}}_{j'}), \qquad (1)$$

if it is assumed that the Hamiltonian

$$H = -\sum_{i < j} J(\vec{R}_{j} - \vec{R}_{i})\vec{S}_{i} \cdot \vec{S}_{j} + \sum_{j} \{BS_{zj}^{2} + \frac{1}{2}G[(S_{xj} + iS_{yj})^{6} + (S_{xj} - iS_{yj})^{6}]\}$$
(2)

is sufficient to describe the magnetism of ferromagnetic Tb. The sums in the first term of the Hamiltonian are over all pairs of spin *i*, *j*, whereas the sums in (1) are over all vectors from one atom to the other atoms in the same sublattice $(\vec{\mathbf{R}}_j)$ and over all vectors to atoms in the other sublattice $(\vec{\mathbf{R}}_j)$, respectively.

Under the above assumption, the magnon dispersion relation becomes⁵

$$\hbar\omega_{j} = S\{f_{j}^{2} + 2(B + 21GS^{4})f_{j} + 72GS^{4}(B + 3GS^{4})\}^{1/2}, \quad j = 1, 2,$$
(3)

where

$$f_{j} = J(0) - J(\mathbf{\bar{q}}) + J'(0) + (-1)^{j} |J'(\mathbf{\bar{q}})|.$$
(4)

The values 1 and 2 of the subscript j refer to the acoustical and optical branch, respectively.

The dispersion curves for the *c* direction can be regarded as a single acoustic branch extending twice as far as the first Brillouin zone if the points of the optical branch are moved from \overline{q} to $[(2\pi/c)-\overline{q}]$. If this rearrangement is carried out, we get

$$J(0) - J^{C}(q) = 2 \sum_{m=1}^{\infty} J_{m}^{C} (1 - \cos\frac{1}{2}mcq) \quad \left(-\frac{2\pi}{c} \le q \le \frac{2\pi}{c}\right), \tag{5}$$

so by Fourier inversion of $J(0)-J^{C}(q)$ it is possible to determine the interplanar exchange parameter J_{m}^{c} . (m odd gives interplanar exchange parameters between planes in different sublattices.) For the a and b directions we have

$$J(0) - J^{a}(q) = 2 \sum_{m=1}^{\infty} J_{m}^{a} (1 - \cos \frac{1}{2}maq) \quad \left(-\frac{2\pi}{a} \le q \le \frac{2\pi}{a}\right),$$

$$J'(0) - J'^{a}(q) = 2 \sum_{m=1}^{\infty} J_{m}^{a} (1 - \cos \frac{1}{2}maq) \quad \left(-\frac{2\pi}{a} \le q \le \frac{2\pi}{a}\right),$$
 (6)

and

$$J(0) - J^{b}(q) = 2 \sum_{m=1}^{\infty} J_{m}^{b} \left(1 - \cos\frac{\sqrt{3}}{2}maq\right) \quad \left(-\frac{2\pi}{\sqrt{3}a} \le q \le \frac{2\pi}{\sqrt{3}a}\right),$$
$$[J'(0)]^{2} - |J'^{b}(q)|^{2} = \sum_{p=1}^{\infty} J_{p}^{'b} \left(1 - \cos\frac{\sqrt{3}}{2}maq\right) \quad \left(-\frac{2\pi}{\sqrt{3}a} \le q \le \frac{2\pi}{\sqrt{3}a}\right), \tag{7}$$

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FIG. 2. Fourier-inverted exchange parameters and interplanar exchange parameters at 90°K.

where

$$J_{p'}^{b} = \sum_{m-\infty}^{\infty} J_{m'}^{b} J_{m-p'}^{b}$$

in an obvious notation.

Figure 2 shows the Fourier-inverted exchange parameters. The points are calculated from average values of the magnon energies obtained in different scans. The accuracy of the points on the figure is estimated to be better than ± 0.02 meV.

The figure also shows a least-squares fit of expressions (5), (6), and (7) to the data, and the interplanar exchange parameters thus obtained are given in Table I. The number of Fourier components used is what is necessary to fit the data within the experimental accuracy.

The anisotropy constants used for this analysis were obtained from susceptibility and magnetization measurements⁶ ($BS^2 \sim 9.5 \text{ meV}$, $GS^6 \sim 0.4 \text{ meV}$). This choice of anisotropy parameters gives an acoustical energy gap of 0.9 meV, which is consistent with the measured dispersion relations. There is a considerable uncertainty in the determination of the anisotropy constants, but this is not very important, since the results of the analysis are rather insensitive to changes of the anisotropy constants.

The determination of the interplanar exchange forces for the *c* direction is of particular interest. The theories⁷ for the stability of the spiral structure around 220°K assume "competition" of positive and negative exchange between neighboring and alternate planes, respectively. We found that the sign of the first two interplanar exchange parameters are both posi-

Table I. Interplanar exchange parameters. All are given in meV except the J' for the b direction, which are in meV². Accuracy ± 0.005 meV.

	a direction	b direction	c direction
J_1	0.200	0.240	0.305
$\overline{J_2}$	0.120	0.040	0.075
J_3	0.045	0.010	0.005
J_4	0.020	0.005	-0.035
J_5	0.005		
J_1'	0.195	0.050	
J_2'	-0.005	0.050	
$\bar{J_{3}'}$	0.050	0.010	
J_4'	0.015	0.010	
J_5'	0.010		

tive. Even if allowance is made for two further interplanar exchange constants, the condition for the stability of the spiral structure is far from fulfillment.

Preliminary measurements have been made in the *c* direction at various temperatures up to 171° K. The acoustical energy gap at q = 0and the interplanar exchange parameters seem to decrease with increasing temperature in qualitative agreement with the theory of Bogolyubov and Tyablikov.⁸ Also the relative magnitude of the interplanar exchange parameters seems to change with temperature.

More precise measurements at temperatures above 90°K are being carried out, and we plan soon to extend these measurements into the spiral region.

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