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ABSTRACT

The spin-wave dispersion relations for the [110] and [001] symmetry directions in ferromagnetic dysprosium metal (isotopically enriched 96.8% with ^{163}Dy) have been studied between 4.7° and 77°K. Throughout this temperature range the measured temperature dependence of the energy gap at zero wave vector ($q = 0$) is in fair agreement with that predicted theoretically for the frozen lattice model. However, the magnitude of the total planar anisotropy deduced from these gap measurements is larger by a factor of two than that obtained from macroscopic measurements. The increase of the magnon energies with decreasing temperature is much more pronounced at $q = 0$ than near the Brillouin zone boundaries. Thus the magnitude of the isotropic exchange interaction decreases substantially, about 25% in the [001] direction, between 77° and 4.7°K. Significant magnon-phonon interactions are observed, particularly in the [110] direction where the acoustic magnon branch is mixed with both the longitudinal and transverse acoustic phonon branches. The peak in the Fourier transform of the exchange interaction, $J(q)$, which at 77°K occurs near the wave vector of the helical-magnetic structure of Dy just above T_c , persists down to 4.7°K with shifts to smaller q as the temperature is decreased. At 77°K the optic and acoustic spin-wave branches are degenerate, to within 0.1 meV, at the K-symmetry point of the Brillouin zone. However at 4.7°K this degeneracy is lifted, with $\Delta E \approx 0.8$ meV, thereby indicating the existence of a temperature dependent long-range anisotropic magnetic interaction in Dy.

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INTRODUCTION

In previous publications [1,2] we have reported the results of a neutron scattering investigation of the magnon dispersion relation for the c-direction of dysprosium metal at 77° and 98°K; just below and above the Curie temperature of 85°K. One puzzling feature of those results is the large discrepancy (~40%) between the magnon energy gap (at zero wave vector) measured in the ferromagnetic phase and that calculated from published values of the crystal field and magnetoelastic anisotropy parameters. The effective planar anisotropy deduced from the energy gap measurement at 77°K is approximately a factor of two larger than that obtained from macroscopic measurements. To investigate this discrepancy further we have studied the temperature dependence of the energy gap between 4.7° and 77°K. In addition the spin-wave dispersion relation has been measured in the [001] direction at 4.7°K and in the [110] direction at 4.7° and 77°K. As before [1] the measurements were carried out on a sample enriched in the low-neutron-capturing isotope ^{163}Dy , and neutrons with energies in the range 10-16 meV were used.

RESULTS AND DISCUSSION

The measured temperature dependence of the spin-wave energy gap at zero wave vector $\hbar\omega(0)$ is shown in Fig. 1. Throughout the 4° to 77°K range, the measured gap is approximately 40% larger than that calculated from the generally accepted theoretical expression for the frozen lattice model [3,4], viz.,

$$\hbar\omega(0) = (J\sigma)^{-1} \{ 3K_2 [36K_6^6 + C_Y(\lambda_Y)^2 + 5C_Y\lambda_Y A] \}^{1/2} \quad (1)$$

and the measured values for the elastic constant $C_Y = 2(C_{11} - C_{12})$ [5]; the first and second order magnetoelastic constants λ_Y and A [6,7]; and the axial and hexagonal crystal field anisotropy constants K_2 and K_6^6 [8,9]. Here σ is the reduced magnetization [10] and J is the total angular momentum for each atom. As an example the calculated gap at 4.7°K is 2.2 meV compared with the measurement of 3.2 meV.

In view of the large magnitude of this discrepancy, it is essentially impossible, with the data presently available, to determine which of the constants in Eq. (1) is in error, or whether some other important interaction has been omitted from the theory. The term $36K_6^6$ is approximately a factor of ten larger at 4.7°K than the other contributions to the square bracketed factor in Eq. (1). Thus to bring the calculated gap into agreement with the neutron measurements, one must increase either K_6^6 (or K_2) by about a factor of two, λ_Y by a factor of 3, A by a factor of 10, or make some combination of these changes. To obtain the calculated curve shown in Fig. 1, we changed only K_6^6 to fit Eq. (1) to the 4.7°K neutron results. We then assumed that each constant varied with temperature as given by the theory of Callen and Callen [11] viz.,

$$K_2 \propto \sigma^3, \quad K_6^6 \propto \sigma^{21}, \quad \lambda_Y \propto \sigma^3, \quad \text{and} \quad A \propto \sigma^{10}.$$

The agreement between the calculation and the neutron data would be difficult to improve, but it is likely fortuitous since other choices for the constants involved are possible. In addition according to Brooks [12] there is theoretical evidence that the temperature dependences assumed here may not be correct. Assuming that K_2 is well determined by existing macroscopic data, about all we can conclude at present from these neutron measurements is that the dominant contribution (whatever its source) to the square brackets in Eq. (1) varies between 4° and 80°K approximately as σ^n with $n \sim 20$. Thus, for example, if K_6^6 dominates as we have assumed, the σ^{36} variation of this constant deduced by Brooks [12] would not be satisfactory.

The data obtained for the [001] and the [110] directions are shown in Fig. 2 for 4.7°K and in Fig. 3 for 77°K. At 4.7°K evidence is seen for magnon-phonon interactions for both of the crystallographic directions studied. In the [001] direction the results are qualitatively similar to those reported for Tb [13], except that for Tb the optical magnon branch lies above the optical phonon branch. In the [110] direction the magnon acoustic branch (MA in Fig. 1) appears to interact with both the longitudinal and transverse acoustic phonon branches. Such interactions were not possible in Tb where the acoustic magnon branch lies entirely above the acoustic phonon branch in this symmetry direction. There is no evidence in these data of an interaction between the magnon optic branch (MO) and the acoustic phonon branches or between the magnon acoustic and the phonon optic branches.

Calculations based on Jensen's [14] theory for magnon-phonon interactions in the rare-earth metals indicate that an energy splitting of approximately 1.0 meV could be expected at the nominal crossing point of the acoustic magnon and the transverse acoustic phonon branches in the [110] direction. This is in good agreement with the splitting of approximately 0.8 meV observed at both 4.7° and 77°K. In making this comparison we have assumed, as indicated in Figs. 2 and 3, that the interaction is between the magnon and the TA_{11} phonon. The theoretical prediction for the splitting associated with the magnon interaction with the [110]-longitudinal mode at 4.7°K is about 0.4 meV again in reasonable agreement with the experimental results. The theory also predicts a linear dependence of the splitting on the q corresponding to the point of crossing. Thus the absence of an observable magnon-phonon interaction associated with either the [110]-longitudinal and the [001]-transverse modes at 77°K is probably due to the fact that the corresponding crossing points have changed with increasing temperature so as to occur at a q that is too small to result in a splitting which could be resolved in these experiments.

The absence of a degeneracy between the acoustic and optic spin-wave branches at the K-symmetry point in the [110] direction at 4.7°K indicates the existence of a significant anisotropic magnetic interaction in Dy in addition to those mentioned above. Similar results found in Tb have been explained by introducing a dipolar interaction into the spin-wave theory [15]. We have not yet carried out similar calculations for Dy. Also in the vicinity of K at 4.7°K, neutron scattering by both acoustic and optic modes was observed at locations in reciprocal space where the neutron scattering

structure factor for only one mode was expected to be significant. These observations are indicated in Fig. 1 by showing both open circles (acoustic scans) and triangles (optic scans) on both branches near K. The data indicate that these branches do not cross at all. These results are temperature dependent, and at 77°K the optic and acoustic branches are degenerate at K as shown in Fig. 2.

We have not yet attempted an analysis of these data in terms of a complete Hamiltonian which includes magnon-phonon interactions. However, to estimate the magnitude of the exchange interaction along c, we have ignored the magnon-phonon splittings at 4.7°K by joining "smoothly" the magnon acoustic and optic branches in order to obtain the Fourier transform of the exchange, $J(\vec{q}) - J(0)$, from the data as described previously [1,2]. The result is compared to that obtained at 77°K in Fig. 4. Because the magnon energy for the acoustic branch at zero wave vector increased much more than that for the optic branch, as is illustrated in Fig. 2, the overall magnitude of $J(\vec{q}) - J(0)$ along c decreased between 78° and 4.7°K. If these results are interpreted in terms of interplanar exchange constants, one finds that the dominant constant, that between adjacent planes, decreases 25% in this temperature interval. It is interesting to note that the magnitude of the peak in $J(\vec{q}) - J(0)$ at small $\vec{q}$ is essentially temperature independent in the ferromagnetic phase, and its position shifts to smaller q as the temperature is decreased.

REFERENCES

- [1] NICKLOW, R. M., WAKABAYASHI, N., WILKINSON, M. K., REED, R. E., *Phys. Rev. Letters* 26 (1971) 140.
- [2] NICKLOW, R. M., *J. Appl. Phys.* 42 (1971) 1672.
- [3] NIELSEN, M., BJERRUM MØLLER, H., LINDGARD, P. A., MACKINTOSH, A. R., *Phys. Rev. Letters* 25 (1970) 1451.
- [4] GOODINGS, D. A., SOUTHERN, B. W., *Can. J. Phys.* 49 (1971) 1137.
- [5] ROSEN, M., KLIMKER, H., *Phys. Rev. B* 1 (1970) 3748.
- [6] CLARK, A. E., DeSAVAGE, B. F., BOZORTH, R., *Phys. Rev.* 138 (1965) A216.
- [7] RHYNE, J. J., in *Magnetic Properties of Rare Earth Metals*, to be published by Plenum Press (1972).
- [8] RHYNE, J. J., CLARK, A. E., *J. Appl. Phys.* 38 (1967) 1379.
- [9] LIU, S. H., BEHRENDT, D. R., LEGVOLD, S., GOOD, JR., R. H., *Phys. Rev.* 116 (1959) 1464.
- [10] BEHRENDT, D. R., LEGVOLD, S., SPEDDING, F. H., *Phys. Rev.* 109 (1958) 1544.
- [11] CALLEN, H. B., CALLEN, E., *J. Phys. Chem. Solids* 27 (1966) 1271.
- [12] BROOKS, M. S. S., *Phys. Rev. B* 1 (1970) 2257.
- [13] NIELSEN, M., BJERRUM MØLLER, H., MACKINTOSH, A. R., *J. Appl. Phys.* 41 (1970) 1174.
- [14] JENSEN, J., *Intern. J. Magnetism* (to be published).
- [15] LINDGARD, P. A., HOUMANN, JENS GYLDEN, *Proceedings of Conference on Rare Earths and Actinides* (Institute of Physics, London, 1971) 192.

FIGURE CAPTIONS

- Fig. 1. Temperature dependence of the spin-wave energy gap for ferromagnetic dysprosium.
- Fig. 2. Spin-wave dispersion relation for ferromagnetic dysprosium at 4.7°K.
- Fig. 3. Spin-wave dispersion relation for ferromagnetic dysprosium at 77°K.
- Fig. 4. Temperature dependence of the exchange interaction along the c-direction of ferromagnetic dysprosium.

Temperature Dependence of Spin-Wave Energy Gap
for Ferromagnetic Dysprosium.

Spin-Wave Dispersion Relation for Ferromagnetic Dysprosium at 4.7°K.

Spin-Wave Dispersion Relation for Ferromagnetic Dysprosium at 77°K.

Temperature Dependence of Exchange Interaction Along the C-Direction of Ferromagnetic Dysprosium.