Magnetic Resonance in Single-Crystal Dysprosium at 100 GHz^{*}

T. K. Wagner[†] and J. L. Stanford Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa 50010

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The microwave absorption of a single crystal of dysprosium at 100 GHz was studied as a function of temperature and magnetic field applied in the magnetically hard $\langle 10\overline{1}0 \rangle$ direction. A peak in the absorption is attributed to ferromagnetic resonance, and the temperature dependence of the field value at which the resonance occurs is compared with previous theories. The results provide what is believed to be the first unambiguous evidence that magnetoelastic effects are important in the spin-wave spectrum of dysprosium at high microwave frequencies. The best fit between experiment and theory requires a value of the c-axis anisotropy constant less than half that derived from static measurements: i.e., $-2P_2S=21.6$ K/atom, rather than 50.4 K/atom. The value of the magnetoelastic coupling constant that gave the best fit was D^{y} =1.4 K/atom, comparable to the static value of 1.1 K/atom.

INTRODUCTION

Ferromagnetic resonance in dysprosium single crystals has previously been studied by microwave absorption by Bagguley and Liesegang¹ at 10 GHz and by Rossol et al. at 38 GHz,² and was observed to follow the behavior predicted by Cooper and Elliott.³ Subsequent experiments by Wagner and Stanford at 100 GHz in terbium⁴ showed the necessity of incorporating magnetoelastic coupling effects as calculated by Cooper.⁵ In particular, the elastic strains were found to be "frozen" in their equilibrium configuration, rather than being free to follow the magnetic moment. This model was originally proposed by Turov and Shavrov, ⁶ and was one of the models used by Cooper. What this meant experimentally was that the resonance field did not increase at low temperatures, as would be expected from the increasing hexagonal anisotropy, but rather continued to decrease with decreasing temperature. Because of the similarities between Tb and Dy, we decided to extend our 100-GHz absorption studies to Dy.

EXPERIMENT

The 100-GHz microwave spectrometer has been described previously.^{4,7} The single crystal of Dy was grown by the strain-anneal process from 99.9% pure dysprosium. The high-purity Dy metal was obtained from Spedding of this Laboratory and contained total analyzed impurities of approximately 130 ppm, the largest impurity concentrations being 44-ppm O, 14-ppm C, and 12-ppm Fe. A circular disk approximately 5.4 mm in diameter and 0.69 mm thick was spark cut with the axis of the disk within $\frac{1}{2}^{\circ}$ of the *c* axis of the crystal. After slight mechanical lapping, the sample was electropolished to an optical smoothness. The final sample, a disk with rounded edges, was treated as an oblate

ellipsoid in the calculation of demagnetizing factors.

The temperature of the sample was monitored by a copper-constantan thermocouple embedded in the backing plate to which the sample was glued.

The magnetic field was oriented in the plane of the disk, along a magnetically hard $\langle 10\overline{1}0 \rangle$ direction in the crystal, and approximately perpendicular to the microwave magnetic field. The results of several temperature runs are compiled in Fig. 1. The points plotted are the average position of the peak for field increasing and field decreasing. Most of the scatter seems to be due to changes in the microwave-cavity resonant frequency due to magnetostriction in the specimen. This resulted in a change in coupling to the cavity with change in magnetic field.

THEORY

Curves A, B, and C in Fig. 1 were calculated using Cooper's⁵ expression for the frozen-lattice model with appropriate demagnetizing factors, i.e.,

$$(h\nu)^{2} = \left[-2P_{2} S(\hat{I}_{5/2}/\sigma) + 6P_{6}^{6} S^{5}(\hat{I}_{13/2}/\sigma) + D^{\gamma}(\hat{I}_{5/2}^{2}/\sigma) + g\mu_{B}(H + 4\pi M - D_{z}M)\right] \\ \times \left[36P_{6}^{6} S^{5}(\hat{I}_{13/2}/\sigma) + 2D^{\gamma}(\hat{I}_{5/2}^{2}/\sigma) + g\mu_{B}(H - D_{z}M)\right],$$

where *h* is Planck's constant, ν is the microwave frequency (99.6 GHz), $-2P_2 S$ is the *c*-axis anisotropy factor, $P_6^8 S^5$ is the hexagonal anisotropy factor, D^r describes the magnetoelastic coupling, g is the gyromagnetic ratio, and μ_B is the Bohr magneton ($g\mu_B = 0.0895$ K/atom kOe for Dy). D_z is the demagnetizing factor for an oblate spheroid in the

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FIG. 1. Resonance field vs temperature for our *c*-axisnormal Dy disk. H_c is the critical field (including the demagnetizing factor for our sample) which causes a magnetic transition from the spiral antiferromagnetic to the ferromagnetic phase. A, B, C, D are theoretical curves: A, Cooper's theory with $-2P_2S = 50.4$ K/atom (the value determined from the static magnetostriction data of Rhyne and Clark); B, Cooper's theory with $36P_6^6S^5 = -8.3$ K/atom and fit to our data with values $-2P_2S = 21.6$ K/atom and $D^r = 1.4$ K/atom; C, Cooper's theory with $-2P_2S = 50.4$ K/atom and $D^r = 0$. D, Vigren and Liu's theory with $-2P_2S = 50.4$ K/atom, $\Delta_{\star} = 2.03$ K/atom, $\Delta_{\bullet} = 0.6$ K/atom.

direction of a major axis ($D_z = 1.08$ for our sample). (The demagnetizing factors in the microwave field are considered equal to zero for the major axes and 4π for the minor axis, because of the small depth of penetration of the microwave field.) $\hat{I}_{5/2}$ and $\hat{I}_{13/2}$ are the ratios of the hyperbolic Bessel functions of order $\frac{5}{2}$ and $\frac{13}{2}$ to that of order $\frac{1}{2}$, the argument of which is the inverse Langevin function of the reduced magnetization $\sigma = M/M_{\rm sat}$. These functions appear in the theory of Callen and Callen.⁸

Curve A, which lies considerably below the experimental points, was calculated using parameters derived from static measurements, i.e., $-2P_2S = 50.4$ K/atom, $36P_6^8S^5 = -8.3$ K/atom, $D^{\nu} = 1.14$ K/atom. The values of P_2 and P_6^8 were obtained from magnetostriction data.⁹ Cooper calculated the value of D^{ν} , but omitted a factor of $\frac{2}{3}$, which has been included here.

*Work performed in the Ames Laboratory of the U. S. Atomic Energy Commission, Contribution No. 3111. [†]Present address: Physics Department, East StroudsIt should also be noted that these values of field vs temperature for curve A fall close to the knee of the magnetization curves, given by curve H_c , which is the critical field necessary to induce the transition from the spiral antiferromagnetic to the ferromagnetic state.

Curve B has parameters chosen to fit the experimental data at 110 and 150 K. In this temperature region the resonance field is not strongly dependent on the value of $36P_6^6S^5$, so this was taken to be the static value - 8.3 K/atom. The two fitted parameters are $2P_2S = -21.6$ K/atom, D = 1.4 K/atom.

Curve C with the magnetoelastic parameter $D^{r} = 0$ clearly cannot represent the experimental data. This shows that magnetoelastic effects are important in Dy at high microwave frequencies.

Recently, Vigren and Liu¹⁰ have carried out a more complete calculation of the magnetoelastic interaction and its effect on the ferromagnetic resonance. Their expression for the frozen-lattice case differs from that of Cooper only in that D^{r} in the first bracket is replaced by Δ_{\perp} , and $2D^{r}$ in the second bracket is replaced by Δ_{\perp} . For the frozenlattice case, they calculate $\Delta_{\perp} = 0.6$ K/atom and $\Delta_{\perp} = 2.03$ K/atom. The results of carrying out the computation of the resonance field using these modifications is shown by curve D. It can be seen that in this temperature region there is little change from Cooper's results.

CONCLUSION

Ferromagnetic resonance in Dy at 100 GHz is seen to be affected by the magnetoelastic interaction, treated in the frozen-lattice approximation. The *c*-axis anisotropy parameter is apparently reduced by more than a factor of 2 over that derived from static measurements. The experimental results on Dy are qualitatively similar to our earlier 100-GHz results on Tb, ⁴ even to the reduction in $P_2 S$.

In Tb, the ratio of the 100-GHz ferromagneticresonance value for $P_2 S$ to that from the static measurements was 0.57. The corresponding value for Dy reported here is 0.43. The uncertainty in the static $P_2 S$ values is fairly large, so that the ratios for Tb and Dy are the same to within experimental error. A satisfactory explanation of this reduction is lacking at present.

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Low-Temperature Heat Capacity of LaPt₂ and CePt₂ and the Magnetic Susceptibility of CePt₂⁺

R. R. Joseph, * K. A. Gschneidner, Jr., and R. E. Hungsberg[‡] Department of Metallurgy and Institute for Atomic Research, Iowa State University, Ames, Iowa 50010 (Received 30 September 1971)

The heat capacity of LaPt₂ and CePt₂ has been measured from 1.3 to 9 °K and the magnetic susceptibility of CePt₂ from 1.3 to 80 °K. LaPt₂ behaves as a normal metal with $\gamma = 1.17 \text{ mJ/}$ g atom °K² and $\Theta_D = 236$ °K. CePt₂ orders antiferromagnetically at (1.6 ± 0.1) °K. The magnetic-susceptibility behavior and the entropy of ordering can be explained on the basis of a crystal-field splitting of the $J = \frac{5}{2}$ multiplet into a ground-state doublet and an excited quartet lying 216 °K above the doublet.

INTRODUCTION

The Laves-phase compounds comprise one of the largest groups of intermetallic compounds known. This is primarily due to the formation of these compounds by the rare-earth group elements with 13 other metals. Rare-earth Laves-phase compounds, RM_2 , where R is a rare-earth metal, are formed with Mg, Al, Mn, Fe, Co, Ni, Tc, Ru, Rh, Re, Os, Ir, and Pt. In an effort to better understand these materials, we have begun a study of some of their physical properties, since about the only information known is their crystal structures and in some instances their magnetic properties.

The cerium Laves phases are of particular interest since the valence of cerium is known to vary from three to four in these compounds.¹ CePt₂ was chosen since cerium is trivalent in this compound.¹⁻⁴ Magnetic studies²⁻⁴ from 35 to 470 °K have confirmed this, and furthermore they indicate that CePt₂ might order antiferromagnetically near 0 °K [the Curie-Weiss parameters are $p_{eff} = 2.33 \mu_B$ and $\theta = 4$ °K (Ref. 2); $p_{eff} = (2.57 \pm 0.07) \mu_B$ and θ = (-26 ± 5) °K (Ref. 3); $p_{eff} = 2.50 \mu_B$ and θ was not given (Ref. 4), but a Curie temperature of 7 °K was reported].

The LaPt₂ Laves phase was chosen to be studied because the outer electronic configurations of $LaPt_2$ and $CePt_2$ are expected to be nearly the same. And it would be useful to compare the low-tempera-

ture heat capacities of these two compounds, especially if the 4*f* electron of the cerium atom causes some unusual low-temperature behavior.

A little information is available on the La-Pt and Ce-Pt phase diagrams. In addition to the Lavesphase compounds the RPt^5 and RPt_5^{6} compounds (where R is La or Ce) are also known to exist. There is some doubt⁷ whether RPt_3 exists or is the terminus of a solid-solution region extending from RPt_2 .^{8,9} No information exists on the melting modes and melting temperatures of these compounds.

EXPERIMENTAL

Sample Preparation

Chemical analysis of the component metals used to prepare the LaPt₂ and CePt₂ compounds is listed in Table I. Weighed amounts of the two components corresponding to the *R*Pt₂ stoichiometry were arc melted. The arc-melted samples of CePt₂ or LaPt₂ were not single phase, thus indicating that these compounds form peritectically. Homogenization of these compounds was accomplished by placing them in a water-cooled trough¹⁰ inside a continuously evacuated quartz tube and inductively heating them to (1350 ± 50) °C for 15 min. They were then cooled at the rate of 10 deg/min until a temperature of 700 °C was reached. At this temperature the power was shut off and the sample cooled rapidly to room temperature. Metallographic analysis showed that