function of energy. The effect of tilting the ion beam away from normal incidence, evidently, is more important at the lower range of energy than at the higher. This is attributed to increased penetration at higher energy. Under this condition, it can be conjectured that the initial momentum of the ion is dissipated throughout a larger lattice volume with the resultant distribution approaching that for normal incidence. Again, the importance of the depth of the collision below the surface is thought to be a primary factor in the emission of sputtered particles, whereas focused collision transport to the surface is thought to be a secondary consideration.

CONCLUSIONS

From the measured data, it is apparent that the sputtering yield and angular distribution can be markedly affected by the crystalline state of the target and its orientation relative to the incident ion beam. The yield is low for incidence along open crystallographic directions such as the [110] or [100] axis while it is increased quite substantially for incidence along the opaque [755] direction. The maximum in the yield as a function of ion energy has been noted in many experiments and has been attributed to the competition between higher-energy transfer to lattice particles and increased penetration of the incident particle as the energy is increased.

The angular distribution results, as interpreted by the nonlinear regression analysis fit of Eq. (1) to the data, suggest that the close-packed focusing contribution to sputtering in these measurements is a second-order effect. The major contribution (80% or more) is a result of the cosine term of Eq. (1) which could be representative of random scattering within the crystal lattice.

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Magnetic Resonance in Single-Crystal Terbium Metal at 100 GHz*

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The absorption of 100-GHz (3-mm) microwave radiation has been studied as a function of temperature and applied magnetic field in single-crystal terbium metal. For magnetic fields up to 26 kOe, three main features are observed in the absorption spectrum: (a) an absorption onset occurring at low fields and low temperatures, which is shown to be associated with the effects of domain rotation; (b) an absorption line occurring around 19 kOe. This line is essentially temperature-independent, although it greatly intensifies below the Curie temperature T_C; (c) a temperature-dependent absorption line which exhibits a dramatic shift as the sample is cooled through T_C, even in the presence of fields thought to be sufficiently strong to destroy the antiferromagnetic ordering and induce ferromagnetic alignment above T_C. The resonance linewidths are very large (~5 to 10 kOe). At low temperatures this line is in good agreement with the theory of Cooper and Elliott for ferromagnetic resonance in a material with large twofold magnetic anisotropy. Extrapolation to $T=0$ yields a value of the twofold-anisotropy constant K_2 for terbium of 5.3×10^8 erg/cm^3 $\pm7\%$. This is in excellent agreement with the latest static-torque data of Rhyne and Clark for terbium, substantiating their result as well as reducing the uncertainty involved in their technique by more than a factor of 3. The agreement with theory at temperatures near and above T_C is poorer although still qualitatively correct. Considering the approximations involved in the theoretical expression, the agreement is considered satisfactory throughout the temperature range investigated. The need is indicated for a theoretical calculation to higher order.

I. INTRODUCTION

THE theory of microwave magnetic resonance in the heavy rare-earth metals was first presented by Cooper *et al.*[1] in 1962, and was discussed in further detail by Cooper and Elliott[2] in 1963. (The latter reference will hereafter be referred to as CE). Since then, resonant microwave power absorption has been studied in dysprosium, terbium, and erbium at 9 and 35 GHz by Bagguley and Liesegang,[3] and in dysprosium at 38 GHz by Rossol *et al.*[4] The latter have observed ferromagnetic resonance in Dy with the aid of an appreciable effective hexagonal anisotropy field which occurs in Dy below about 110°K. The presence of this hexagonal anisotropy field allows the resonance frequency to be brought down, by application of an ex-

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[1] B. R. Cooper, R. J. Elliott, S. J. Nettel, and H. Suhl, Phys. Rev. **127**, 57 (1962).

[2] B. R. Cooper and R. J. Elliott, Phys. Rev. **131**, 1043 (1963).

[3] D. M. S. Bagguley and J. Liesegang, Phys. Letters **17**, 96 (1965); J. Appl. Phys. **37**, 1220 (1966).

[4] F. C. Rossol, B. R. Cooper, and R. V. Jones, J. Appl. Phys. **36**, 1209 (1965); F. C. Rossol and R. V. Jones, *ibid.* **37**, 1227 (1966).

ternal field, from over 100 GHz to within the range of their experimental apparatus.

In contrast, the hexagonal anisotropy field in terbium is very weak for temperatures above 150°K,[5] where most of the data taken in our experiment were obtained. In this paper, we report what we believe is the first detailed study and comparison with theory of ferromagnetic resonance in a substance (single-crystal terbium) with an extremely large twofold magnetic anisotropy and essentially no hexagonal anisotropy to allow bringing down the resonance frequency with an external field. (The magnetic anisotropy in terbium is two orders of magnitude larger than that in the $3d$ transition elements.) The resonance occurs in the millimeter range for the usual laboratory fields ($\approx$10 kOe). The experimental frequency used was 100 GHz.

The magnetism of the rare-earth metals is attributed to an indirect exchange mechanism between localized electrons in the unfilled $4f$ shell, mediated by conduction electrons.[6] Terbium, in particular, may exhibit paramagnetism, antiferromagnetism, and ferromagnetism, depending on temperature and applied magnetic field.[7,8] In the absence of an applied field, terbium is paramagnetic above its Néel temperature, $T_N \cong 228°K$, antiferromagnetic between 228°K and its Curie temperature, $T_C \cong 219°K$, and ferromagnetic at lower temperatures. Uncertainties in the ordering temperatures are the result of sluggishness in the phase transitions, caused by impurities. In the antiferromagnetic region, neutron-diffraction experiments[8] have revealed a helical magnetic structure, with the axis of the helix lying along the hexagonal axis (c axis). (Terbium has hexagonal close-packed structure.) The atoms in a given plane perpendicular to the c axis (basal plane) experience a ferromagnetic spin alignment pointing in the basal plane. The spin alignment in the plane immediately above or below will be in a different direction, but still in the basal plane, so that the magnetization traces out a helix as successive planes of atoms are encountered in the c direction. For Tb, the magnetization completes one turn around the helix in approximately 20-layer spacings along the c axis.[9] The antiferromagnetic state may be destroyed by application of an external magnetic field, with the ferromagnetic state apparently being induced by a field of less than 1 kOe.[7] Below T_C, terbium is ferromagnetic with magnetization lying in the basal plane along a b axis, $(10\bar{1}0)$. The preference between this "easy" basal plane axis of magnetization, and the "hard" basal plane a axis $(11\bar{2}0)$, is small: on the order of 10^4 Oe or less, even at low temperatures. In contrast, the preference for spin alignment in the basal plane, as opposed to being along the c axis, is extraordinarily great. This is thought to be a result of the combined effects of crystal field anisotropy and spin-orbit coupling,[10] which lead to a large effective anisotropy field tending to keep the magnetization lying in the basal plane. In terbium, this effective anisotropy field is 600 kG at liquid-helium temperatures. This very large anisotropy causes the experimental ferromagnetic resonance spectrum in the rare-earth metals to be qualitatively different from that in the transition elements iron, cobalt, and nickel, where the anisotropy fields are on the order of the applied magnetic fields or smaller. In particular, the resonance frequency and field in terbium are not proportional to one another, even in an infinite medium, but obey a nonlinear relation which will now be discussed.

A brief discussion of the relevant theory is given in Sec. II, while Sec. III contains a description of the experimental procedures used. The experimental results are given and compared with theory in Sec. IV, and Sec. V contains a summary and conclusions.

II. THEORY

In the heavy rare-earth metals, the unpaired electrons in the incomplete $4f$ shell are localized close to the nucleus by the large effective nuclear charge. The effect of the anisotropic crystalline electric field is not as important as that of spin-orbit coupling in this case, so that the total angular momentum $\mathbf{J}=\mathbf{L}+\mathbf{S}$ may be regarded as a good quantum number even in the metal, to first approximation. For our purposes, metallic terbium may be represented by a Hamiltonian of the form[2]:

$$\mathcal{H} = -\sum_{i\neq j} \mathcal{J}_{ij}\mathbf{J}_i\cdot\mathbf{J}_j - K_2\sum_i (J_i)_\zeta^2 - g\mu_B H\sum_i (J_i)_\xi. \quad (1)$$

The first two terms account for exchange and twofold anisotropy, while the last term represents the energy incurred when an external magnetic field H is applied along the ξ direction in the basal plane. The ζ direction is that of the hexagonal axis (c axis). J_i is the total angular momentum of the atom located at the ith lattice site. Recent measurements by Rhyne and Clark[5] have shown that the fourth- and sixth-order anisotropy constants K_4 and K_6 are very small for terbium, and, consequently, these effects have not been included in the Hamiltonian. The effects of basal-plane anisotropy are also small in terbium and have been neglected in Eq. (1), as well as the effects of anisotropy in the exchange mechanism. It will be seen from (1)

[5] J. J. Rhyne and A. E. Clark, J. Appl. Phys. (to be published).
[6] See, for example, K. Yosida, in *Progress in Low-Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Company, Amsterdam, 1964), Vol. IV, Chap. V.
[7] D. E. Hegland, S. Legvold, and F. H. Spedding, Phys. Rev. **131**, 158 (1963).
[8] W. C. Koehler, H. R. Child, E. O. Wollan, and J. W. Cable, J. Appl. Phys. **34**, 1335 (1963).
[9] W. C. Koehler, J. Appl. Phys. **36**, 1078 (1965).
[10] See, for example, C. Kittel, *Introduction to Solid State Physics* (John Wiley & Sons, Inc., New York, 1966), 3rd ed., p. 490.

that if the magnetization is to lie in the basal plane, K_2 is a negative number.

Starting from a Hamiltonian of the form of (1), CE find that spin-wave modes of frequency ω can be excited in the ferromagnetic spin configuration if the following condition is satisfied:

$$\hbar\omega=[(-2K_2\sigma(T)+g\mu_BH)g\mu_BH]^{1/2}, \quad (2)$$

where $\sigma(T)=\langle(J_i)_\xi\rangle$ and $\sigma(T=0)=J_{\max}=6\hbar$ for terbium. σ is not the maximum, but the equilibrium angular momentum per atom at T and H. Thus, Eq. (2) must be solved self-consistently if the correct value of H corresponding to resonance excitation of the ferromagnetic spin configuration is to be calculated for a specified excitation frequency ω.

The experimental values of the twofold anisotropy constant show a temperature dependence corresponding to the third power of the magnetization, as predicted by the semiclassical theory of Zener,[11] and the recent quantum-mechanical approach of Callen and Callen[12] for low temperatures. However, the quantity labeled K_2 in (2) is a temperature-*independent* constant. In the Appendix, it is shown that if the anisotropic crystalline electric field is mainly responsible for the magnetic anisotropy, when this effect is correctly taken into account, the quantity K_2 appearing in Eqs. (1) and (2) is temperature-independent. The experimental measurements yield an M^3 dependence because the static-torque technique utilized measures the anisotropy energy,

$$E_{\text{anis}}=\langle\mathcal{H}_{\text{anis}}\rangle=\langle-K_2\sum_i(J_i)_\xi^2\rangle. \quad (3)$$

Theory[11,12] predicts that at low temperatures Eq. (3) should be proportional to

$$M^3(H,T)P_2(\cos\theta), \quad (4)$$

where P_2 is the Legendre polynomial and θ is the angle between the c axis and the magnetization M. The experimentally measured twofold anisotropy constant is the coefficient of $P_2(\cos\theta)$, which explains the measured M^3 dependence. However, if the crystal field is mainly responsible for the anisotropy, then K_2 is a temperature-independent quantity, as is shown in the Appendix. In particular, the correct value of K_2 to be used is the $T=0$ value of the twofold anisotropy constant. This is found by Rhyne and Clark[5] to correspond to 5.5×10^8 erg/cm$^3\pm25\%$.

Note that σ is simply related to M by $\sigma=M/g\mu_Bn$, where M is the magnetization per volume and n is the number of atoms per volume. It will be seen from Eq. (2) that in the limit of large twofold magnetic anisotropy, $-2K_2\sigma\gg g\mu_BH$, the resonance field H will be proportional to the inverse of the magnetization, $M(H,T)$.

In general, (2) is true only for an infinitely large specimen. For finite specimens, (2) must be modified to include demagnetizing effects and then has the form[2]:

$$[\hbar\omega]^2=[-2K_2\sigma+g\mu_B(H-(N_\xi-N_\zeta)M)]\times[g\mu_B(H-(N_\xi-N_\eta)M)]. \quad (5)$$

Here ξ and ζ are mutually perpendicular directions in the basal plane, H being oriented along ξ. N_ξ and N_η are the usual demagnetizing coefficients. The specimen used in this investigation was in the form of a flat circular disk, whose normal coincided with the c axis. Thus, $N_\xi=N_\eta$ in our experiment.

As previously noted, the value of K_2 is so large for terbium that it corresponds to an effective anisotropy field of 600 kG at low temperatures. On the other hand, the magnetization of terbium has a low-temperature value of less than 35 kG, and the experimental values of applied fields utilized in this investigation were under 26 kOe. Thus, the corrections for demagnetizing effects are negligible at low temperatures, and are only of second order even for temperatures at and above T_C. However, these corrections have been included in the theoretical calculations presented in Sec. IV.

For $T\ll T_c$, there is no difference between $M(T)$ and $M(H,T)$, once domain alignment has been achieved. However, for higher temperatures the magnetization is field-dependent. In fact, near T_c, even for a single domain, the magnetization is greatly enhanced by the application of an external field H. Since our data were obtained at temperatures near T_c, M must be considered a function of both T and H. The values of M used in Eq. (5) must be consistent with the field values obtained for H from the same equation. Thus, Eq. (5) must be solved in a self-consistent manner if the correct H-versus-T dependence is to be obtained for a specified ω.

III. EXPERIMENTAL PROCEDURES

Apparatus

A block diagram of the experimental apparatus is shown in Fig. 1. The 3-mm microwaves are generated by a Raytheon type QKK-1074 klystron with an output power in the range of tens of milliwatts. The power reflected from the specimen cavity is detected by means of a crystal detector whose output is fed into the Y posts of an X-Y recorder. The X posts of the recorder are driven by a voltage proportional to H, derived from a Rawson rotating-coil gaussmeter and passing through a low-pass filter to reduce the ac ripple.

The klystron is frequency-modulated by a small amplitude 70-kHz voltage applied to the reflector. This

[11] C. Zener, Phys. Rev. **96**, 1335 (1954).
[12] H. B. Callen and E. R. Callen, J. Phys. Chem. Solids **27**, 1271 (1966).

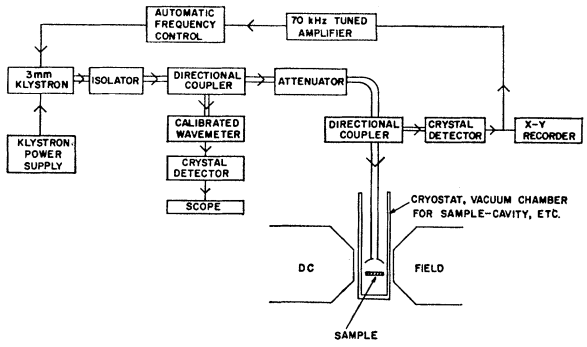

FIG. 1. Block diagram of the experimental apparatus.

produces a 70-kHz signal output from the crystal detector, which is fed into a 70-kHz tuned amplifier. The amplified signal goes into an automatic frequency control system which utilizes a 70-kHz phase-sensitive detector to lock the klystron frequency to that of the sample cavity. Thus, frequency shifts due to thermal and magnetic contractions of the sample and/or cavity are automatically accounted for, as well as the shifts encountered near resonance.

The sample-cavity is depicted in Fig. 2. Because of the spherical-cap image seen in the sample, the system approximates a confocal resonator.[13] The loaded Q of this system is sufficiently high ($\sim$2000) even at 3 mm for good operation of the microwave system.

The klystron frequency was calibrated by a flat-plate interferometer designed and constructed in this laboratory. The copper faces of the interferometer had a diameter of over 50 wavelengths (at 100 GHz) and were flat and parallel to about 1/400 and 1/40 wavelength, respectively. A fine screw mechanism allowed precision adjustment of the spacing between the faces. Utilizing this interferometer in conjunction with a secondary confocal resonator, a repeatable calibration curve of $\pm 0.2\%$ accuracy was obtained. The secondary confocal resonator consisted of two spherical caps, one attached to a calibrated micrometer head, and was kept in the microwave circuit as a calibrated 3-mm wavemeter.

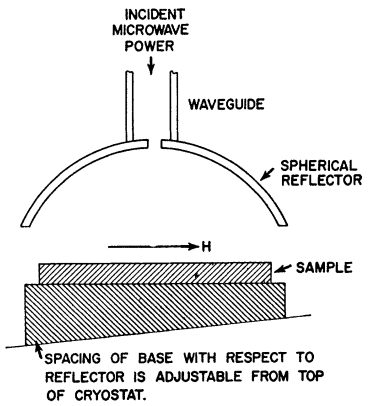

FIG. 2. Sample-cavity system. The normal to the sample is aligned along the (0001) direction. H is applied parallel to the sample along a $(11\bar{2}0)$ direction.

[13] G. D. Boyd and J. P. Gordan, Bell System Tech. J. **40**, 489 (1961); G. D. Boyd and H. Kogelnik, *ibid.* **41**, 1347 (1962).

Specimen

The specimen used in this investigation was kindly lent to us by Professor Sam Legvold of the Ames Laboratory. It was grown as a single crystal from an arc-melted button of pure metal by the thermal-strain-anneal method described by Nigh.[14] The purity of the specimen is about 99.5%, with the largest impurity concentrations being $\frac{1}{4}\%$ tantalum and $\frac{1}{10}\%$ oxygen.

The heavy rare-earth metals oxidize, although rather slowly. The specimen was acid-etched with a solution of 60% acetic and 40% nitric acid to remove the accumulated surface oxide layer. A methanol rinse was used to stop the etching process. A subsequent Laue back-scatter x-ray examination of the sample revealed a good crystal structure.

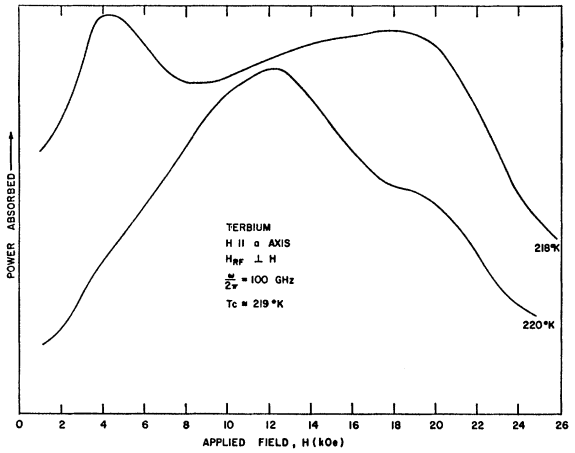

FIG. 3. Tracing of experimental X-Y recorder curves for temperatures near T_C.

The specimen was in the shape of a flat disk approximately $\frac{3}{8}$ in. in diameter and having a diameter-to-thickness ratio of 4.2. Thus, ordinarily the demagnetizing effects would be important. However, as discussed in Sec. II, the condition for ferromagnetic resonance in a substance of large magnetic anisotropy is such that the extremely large values of K_2 overshadow demagnetizing effects. Even above T_C, where the magnetic anisotropy decreases more rapidly than the demagnetizing effects, the corrections are still of second order. These effects have nevertheless been included in the theoretical calculations.

After some initial data had been taken, the sample was electropolished by a technique involving a perchloric acid solution at -70°C.[15] This noticeably

[14] H. E. Nigh, J. Appl. Phys. **34**, 3323 (1963).

[15] E. N. Hopkins, D. T. Peterson, and H. H. Baker, U.S. Atomic Energy Commission Report No. IS-1184 (unpublished).

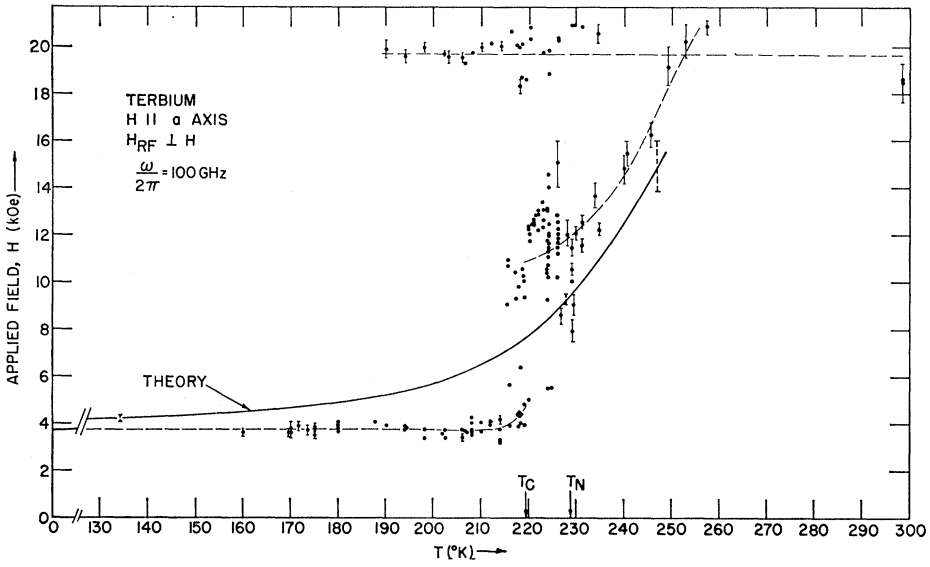

FIG. 4. Values of applied field for which power absorption maxima occur, plotted versus temperature. To reduce congestion, only representative error bars are included. The scatter of the data points is generally greater than experimental error and reflects magnetic hysteresis effects. The theoretical curve was obtained by a self-consistent calculation of Cooper and Elliott's expression for ferromagnetic resonance in a material with twofold magnetic anisotropy. The sample was disk-shaped, with normal along (0001) and H applied in the plane of the disk.

sharpened the resonances, but otherwise no major differences were noted. Figure 3 was obtained after electropolishing.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

Representative tracings of experimental power-absorption curves versus applied magnetic field are shown in Fig. 3. A plot of the field values for which absorption maxima occur versus sample temperature is given in Fig. 4. The data show several distinct features:

(a) A low-field absorption onset is found to occur at low temperatures (Fig. 5). Apparently similar effects have been observed in dysprosium at lower frequencies.[16]

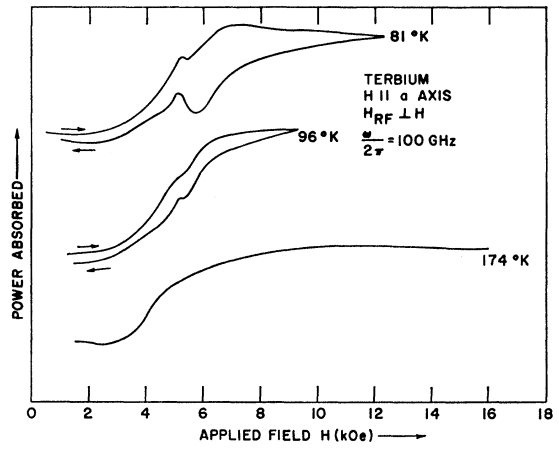

FIG. 5. Microwave power absorption onset that occurs at lower temperatures. As discussed in the text, these are associated with domain-orientation effects.

[16] H. A. Blackstead and P. L. Donoho, Bull. Am. Phys. Soc. **11**, 760 (1966).

We believe that this absorption onset is associated with domain-orientation effects. The magnetization within a given domain is the equilibrium saturation magnetization $M_s(H, T)$. However, the total magnetization of the sample is the vector sum of the individual domain magnetizations. When the specimen is placed in an applied field, the total sample magnetization approaches $M_s(H, T)$ for large values of H, when all the domains find it energetically more favorable to align along H. For this reason, the sample undergoes magnetostriction, which occurs with an onset behavior as H is increased from zero. Rhyne and Legvold[17] have published the results of extensive magnetostriction measurements in terbium, and in fact, for the same sample as was used in the present microwave experiment. Plots of the onset of magnetostriction and the onset of microwave power absorption are found to lie on the same curve, as a function of sample temperature. We thus conclude that the low-temperature absorption onsets are due to domain orientation effects.

(b) An absorption line at about 19 kOe which appears to be almost temperature-independent is observed to occur from room temperature down to temperatures at least as low as 190°K, the lowest temperature for which high-field data were obtained. This line would correspond to a g value near 3.7, whereas for Tb, a g value of 1.5 is expected [and confirmed for the other resonance line to be discussed in part (c)]. If the 19-kOe line were due to paramagnetic resonance in an oxide coating on the sample surface, any demagnetizing effects would shift the resonance field as the temperature is lowered below T_C. Within data scatter, the line remains temperature-independent both above and below T_C. However, paramagnetic resonance in an oxide coating, not being in the bulk metal, might not

[17] J. J. Rhyne and S. Legvold, Phys. Rev. **138**, A507 (1965).

experience an appreciable fraction of the bulk demagnetizing fields. The absorption of the line greatly intensifies for $T<T_C$ (by nearly an order of magnitude in strength), suggesting that it is connected with the terbium sample and not some extraneous property of the cavity system. The power absorption is on the order of 50% for $T<T_C$. (Contrast the 19-kOe lines in the two curves shown in Fig. 3.) Terbium oxides are known to have coherent magnetic properties, but with very low ordering temperatures, in the liquid-helium range.[18] At present we do not have a satisfactory explanation for this line.

(c) The most interesting feature of the data is a resonance line which starts at low fields (~3.8 kOe) for low temperatures, makes an abrupt jump at T_C, and finally rises steeply for $T>T_N$. The absorption peaks have intensities characteristic of collective phenomena (30–50% power absorption). This line we attribute to ferromagnetic resonance, and the peculiar magnetic anisotropy of terbium accounts for this line quantitatively at low temperatures, and qualitative at temperatures around and above T_C. Using published experimental values of $M(H, T)$ for Tb, we have solved Eq. (5), CE's expression for ferromagnetic resonance in a material with twofold magnetic anisotropy. The magnetization data of Hegland *et al.*[7] was used to obtain the necessary values of $M(H, T)$. The calculated values for the resonance field versus temperature are shown as the solid line in Fig. 4. The error bars shown as dashed lines are for the theoretical curve and are a consequence of uncertainties in $M(H, T)$ values, while experimental error bars are shown as solid lines. To reduce congestion, only representative experimental error bars are shown. Measurement of the low-field line was not carried out below 160°K because the line had flattened out well above this temperature and more important, at low temperatures the field strength required for domain alignment in our sample exceeds that of the low-field line.

Because of the strong spin-orbit coupling in terbium, a g value of 1.5 is expected. Aside from using $g=1.5$, the only adjustable parameter used in the theoretical curve is K_2. Allowing the theoretical curve to coincide at $T=0$ with the extrapolated experimental value of $3.7_7\pm0.2$ kOe, we obtain a value of K_2 corresponding to

$$K_2=5.3\times10^8 \text{ erg/cm}^3\pm7\%. \quad (6)$$

This is in excellent agreement with the latest $T=0$ value of K_2 obtained from the static-torque method by Rhyne and Clark,[5] who find $K_2=5.5\times10^8$ erg/cm$^3\pm25\%$. With the microwave-resonance technique we are thus able to substantiate their result, as well as reduce the uncertainty by more than a factor of 3.

The theory and the experimental data are only in qualitative agreement at finite temperatures. This is to be expected because the approximations involved in the theoretical expression render it exact only in the limit of very low temperatures. Considering the approximations involved, the agreement over the whole temperature range is considered reasonable. However, the need is demonstrated for a theoretical calculation to higher order.

Above T_N, the H-versus-T curve rises steeply, exceeding the range of our magnet for $T>260$°K. Presumably, as T increases to a value far above the ordering temperature, the temperature-independent paramagnetic resonance condition should be approached:

$$\hbar\omega=g\mu_B H. \quad (7)$$

This follows from Eq. (5) in the high-temperature limit where the coherent magnetic effects disappear, i.e., when $M(T)\to0$. For $g=1.5$ and 100 GHz, Eq. (7) yields $H=47.6$ kOe, which is beyond the range of our apparatus.

From the magnetization curves in a magnetic field,[7] it appears likely that all or almost all of the antiferromagnetic phase of terbium is destroyed by application of a field as low as 1 kOe. For fields ~10 kOe, the magnetic state of Tb for $T_C<T<T_N$ is expected to be ferromagnetic, and certainly only very smooth changes are seen in the magnetization between 220 and 218°K.[7] In contrast to this, Fig. 3 reveals a dramatic shift in the microwave power absorption spectrum as the sample is cooled through T_C, even in the presence of fields on the order of 10 kOe. Evidently, the mechanism responsible for the coherent magnetism senses that something "ought to happen" at T_C, even though the applied field is apparently large enough to suppress the antiferromagnetic phase and force the system ferromagnetic above T_C.

(d) Finally, the absorption peaks exhibit very large linewidths, on the order of 5 to 10 kOe. Interference between the two absorption lines makes it difficult to obtain meaningful linewidth values. However, it can be seen that the linewidth of the temperature-dependent line is much smaller below T_C, jumping abruptly from a value of 2 to 3 kOe below T_C to around 10 kOe as the temperature is increased through the Curie point. It appears that the temperature-independent line has just the opposite behavior. Before electropolishing the sample, the linewidths were larger but by less than an order of magnitude. Anomalously large linewidths, of the same magnitude, have also been reported in dysprosium metal at 38 GHz by Rossol and Jones.[4] Linewidths too large to measure in applied fields up to 10 kOe were reported several years ago by Kip *et al.*[19] in a study of 9- and 24-GHz power absorption in the ferromagnetic

[18] J. B. MacChesney, H. J. Williams, R. C. Sherwood, and J. F. Potter, J. Appl. Phys. **37**, 1435 (1966); B. C. Gerstein, J. F. Jelinek, and F. H. Spedding, Phys. Rev. Letters **8**, 425 (1962).

[19] A. F. Kip, C. Kittel, A. M. Portis, R. Barton, and F. H. Spedding, Phys. Rev. **89**, 518 (1953); A. F. Kip, Rev. Mod. Phys. **25**, 229 (1953).

phase of polycrystalline gadolinium metal. The microwave fields in our cavity are concentrated near the center of the sample, so that the effect of demagnetizing fields on the linewidth is minimized. Colquitt[20] has recently calculated the effect of the microwave skin depth on the linewidth for the transition metals. It may be that an extension of his calculation to the rare-earth metals will account for the large linewidths.

In view of the data scatter from magnetic hysteresis and the approximations involved in the theory, we have made no attempt in the above analysis to correct for any shifts in the position of the resonance line caused by the microwave skin depth being much smaller than the sample thickness.

V. SUMMARY AND CONCLUSIONS

Microwave power absorption at 100 GHz has been studied in single-crystal terbium metal as a function of applied magnetic field and temperature. Two distinct resonance lines are observed, as well as a low-field absorption onset occurring at lower temperatures. The latter is shown to be connected with domain-orientation effects. The resonance lines have large linewidths (~5 to 10 kOe), and their intensities are characteristic of that of collective phenomena (~50% absorption). One line occurs around 19 kOe and is essentially temperature-independent although it greatly intensifies below T_C. The explanation for this line is at present unknown. The second line is temperature-dependent and is accounted for by the theory of Cooper and Elliott for ferromagnetic resonance in a material with large twofold magnetic anisotropy. This line has an abrupt shift as the sample is cooled through T_C, even in the presence of applied fields thought to be sufficiently strong to induce ferromagnetic ordering above T_C. Extrapolating to $T=0$, a value of the twofold anisotropy constant is found which is in good agreement with the latest static-torque data for terbium. Considering the approximations involved in the theoretical expression of Cooper and Elliott, the agreement between theory and experiment is considered to be satisfactory throughout the temperature range investigated. The need is indicated for a theoretical calculation to higher order.

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APPENDIX

We wish to prove that if the effects of the anisotropic crystalline electric field are correctly taken into account, the twofold anisotropy constant K_2 appearing in Eqs. (2) and (5) is temperature- and field-independent.[21] Physically, the macroscopic, phenomenological anisotropy constant K_2 arises from the combined effects of the anisotropic crystalline electric field seen by a $4f$ electron at a lattice site and spin-orbit interactions which allow the electron's magnetic moment to be influenced by its spacial orientation.

Let the crystal field potential term in the Hamiltonian be represented by the form

$$\mathcal{H}_{\text{crystal field}}=V_2\sum_i(z_i/r_i)^2. \tag{A1}$$

Terms of fourth, sixth, and higher order are found experimentally to be very small for terbium, as was discussed in the text. In (A1), the z axis is along the c axis of the crystal, and r_i is the distance between the ith lattice site and the lattice site at which the potential is being calculated.

The Wigner–Eckart[22] theorem allows $(z_i/r_i)^2$ to be identified with a constant C times $(J_i)_z^2$. Thus, (A1) may be rewritten as

$$V_2\sum_i C(J_i)_z^2=CV_2\sum_i(J_i)_z^2. \tag{A2}$$

The constant C is not a function of which particular lattice site is chosen since all the sites are equivalent. Therefore, C can be taken outside the summation. C depends only on the atomic wave functions of the $4f$ electrons, and indirectly on the lattice structure through its effect on the same atomic wave functions. Thus, aside from the effects of lattice thermal expansion and magnetostriction, C (and also V_2) is independent of temperature and applied magnetic field. From Eq. (1), the constant K_2 is just the coefficient of $\sum_i(J_i)_z^2$. K_2 is therefore seen to be independent of temperature and applied magnetic field. The effects of thermal expansion and magnetostriction are of second order compared with those of finite T and H on the statistical average of $\sum_i(J_i)_z^2$, which lead to an $M^3(H, T)$ dependence.

[20] L. Colquitt (to be published).

[21] We wish to thank Professor S. H. Liu for several enlightening discussions concerning this point.

[22] See, for example, M. E. Rose, *Elementary Theory of Angular Momentum* (John Wiley & Sons, Inc., New York, 1957), p. 85.