

Pressure Dependence of the Magnetic Transition Temperatures in Terbium from Resistance Measurements*

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The pressure coefficient of the Curie and Néel temperatures, T_{cf} and T_N , are determined using electrical-resistance R measurements. The resistance of polycrystalline terbium at constant pressure is determined over the temperature range 200 to 400°K. Pressures to 14.22 kbar are employed. Resistance measurements were made using the standard four-probe method. The Curie and Néel temperatures are determined from (1) plots of R versus T and (2) plots of dR/dT versus T . The values of T_{cf} and T_N thus obtained agree to within $\pm 1^\circ\text{K}$. The pressure coefficient of the Néel temperature is found to be $-1.05^\circ\text{K}/\text{kbar}$; while the coefficient for the ferromagnetic Curie point is determined at $-1.24^\circ\text{K}/\text{kbar}$. The variation of the ratio of the Néel temperature to the de Gennes factor, $T_N(g-1)^2J(J+1)$, with pressure is discussed in terms of the variation of the shape of the Brillouin zone, the number of energy states per atom, and the density of states per energy level in the zone with pressure which is directly related to the variation of the c/a ratio with pressure.

INTRODUCTION

TERBIUM can exist in a paramagnetic, an antiferromagnetic, and a ferromagnetic state. It transforms from the paramagnetic state to the helical spin state at the Néel temperature, T_N , about 230°K. In this state the magnetic moments are in planes perpendicular to the c axis of the hexagonal crystal structure, namely the basal planes. In any one of these basal planes the atomic moments are parallel to each other. In the next adjacent plane however, the atomic moments are parallel to each other but make an angle ω with respect to the moments in the reference plane. This angle is called the interlayer turn angle. After passing through n such planes where $n\omega=2\pi$, the moments will have cancelled each other resulting in zero net magnetic moment. Terbium transforms from the helical spin state (antiferromagnetic state) to the ferromagnetic state at the Curie temperature T_{cf} about 220°K. In this state the atomic moments lie in the basal plane along the direction of the b axis (i.e., at 30° to the a axis). The turn angle varies from 20.5° at T_N to 360° at T_{cf} . Therefore, any factor that may bring about a change in the hexagonal structure of terbium or a change in the (c/a) ratio, etc., is certain to have some effects upon its magnetic properties.

Recently, using the transformer method, Robinson *et al.*¹ and McWhan *et al.*² determined the effect of hydrostatic pressure upon the magnetic transition temperatures in terbium. The results of Ref. 1 are

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¹ L. B. Robinson, S. Tan, and K. F. Sterrett, Phys. Rev. **141**, 548 (1966).

² D. B. McWhan and A. L. Stevens, Phys. Rev. **139**, A682 (1965).

presented schematically in Fig. 1. Robinson *et al.* suggested that the disappearance of one of the peaks in the curve of secondary voltage output versus temperature upon the application of pressure, curve II of Fig. 1, indicates that either T_{cf} has disappeared, that is, terbium does not possess antiferromagnetic properties any further, or that the sensitivity of the transformer method is not sufficient to detect T_{cf} upon the

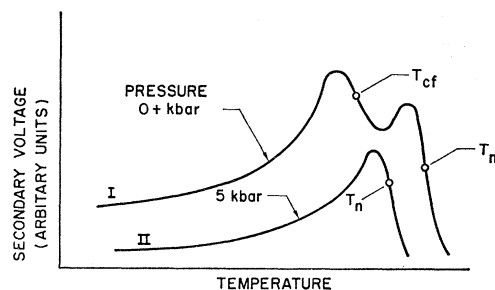


FIG. 1. Schematic of secondary voltage output for a terbium toroid as a function of temperature and pressure.

application of pressure. On the other hand, McWhan stated that the Curie point T_{cf} in terbium cannot be distinguished from the Néel point T_N in the presence of pressure because of the relatively small difference (about 8°K) between the two transition temperatures.

The results of Robinson *et al.* and McWhan *et al.* leave several questions pertaining to the magnetic properties of terbium under pressure unanswered. These are: (1) Does the antiferromagnetic region disappear with applied pressure? (2) If so, at what pressure does the antiferromagnetic region disappear? (3) If T_{cf} does not disappear with pressure, what is its pressure coefficient?

The neutron-diffraction results of Koehler *et al.*³ show that the ferromagnetic Curie temperature of terbium is about 220°K and the Néel temperature is about $230 \pm 1^\circ\text{K}$. These magnetic transitions in terbium

³ W. C. Koehler, H. R. Child, E. O. Wollan, and J. W. Cable, J. Appl. Phys. **34**, 1335 (1963).

are manifested in several of its physical properties. Jennings *et al.*⁴ found a *lambda* anomaly in the heat capacity of terbium at $227 \pm 0.05^\circ\text{K}$ and a further anomalous behavior in the $220\text{--}222^\circ\text{K}$ region. Thermal-conductivity measurements⁵ have shown an anomalous behavior at about 225°K . Magnetization and electrical resistivity studies of terbium single crystals by Hegland *et al.*⁶ resulted in $T_N = 229 \pm 1^\circ\text{K}$ and $T_{cf} = 221 \pm 2^\circ\text{K}$. Therefore, the effect of pressure upon the electrical resistivity, among many other properties, may be used to determine the effect of pressure upon the magnetic transition temperatures in terbium.

Since terbium is hexagonal, its single-crystal resistivity exhibits anisotropic behavior. The resistivity measured in the *c* direction ρ_c is different from the resistivity measured in the basal plane ρ_a . Therefore, the resistivity in a direction making an angle θ with the *c* axis is given by⁷

$$\rho(\theta) = \rho_c \cos^2\theta + \rho_a \sin^2\theta. \quad (1)$$

Polycrystalline resistivity ρ may be estimated from single-crystal data by averaging expression (1) over all solid angles. In a polycrystal where the grains are randomly oriented and where the grain size is small compared to the sample size, this averaging process results in the following expression for ρ :

$$\rho = \frac{1}{3}(2\rho_a + \rho_c). \quad (2)$$

Hegland verified the applicability of this equation to terbium.

According to Kasuya,⁸ the resistivity of a rare-earth metal such as terbium may be expressed by

$$\rho = \rho_{\text{res}} + \rho_{\text{ph}} + \rho_{\text{mag}}, \quad (3)$$

where ρ_{res} is the resistivity at $T = 0^\circ\text{K}$, referred to as the residual resistivity, ρ_{ph} is the resistivity due to phonon-scattering, and ρ_{mag} is the resistivity arising from magnetic disorder or spin-disorder scattering. The residual resistivity results from impurity and structural imperfection scattering. In applying Eq. (3) to experimental values, ρ_{res} is commonly assumed independent of temperature. The term ρ_{ph} , however, is a function of temperature and is assumed to vary with temperature in the same manner as the resistivity of a nonmagnetic hexagonal metal. It is further assumed that its variation with temperature is continuous throughout the transition region. The term ρ_{mag} is also assumed to vary with temperature below T_N ; however, it is regarded as constant in the paramagnetic region, $T > T_N$. The magnitude of ρ_{mag} in the paramagnetic region is found by extrapolating the almost linear region above T_N to $T = 0^\circ\text{K}$ and subtracting ρ_{res} . A typical

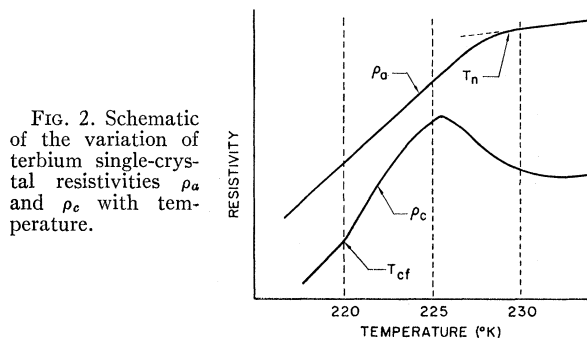


FIG. 2. Schematic of the variation of terbium single-crystal resistivities ρ_a and ρ_c with temperature.

value of ρ_{mag} for terbium is $88 \mu\Omega \text{ cm}$ at 0°K (extrapolated).

At low temperatures the change of ρ results mainly from the variation in ρ_{mag} with temperature. Attempts that have been made to analyze this resistivity by a spin-wave model⁹ have led to a T^2 dependence. Arajs has found that between 5 and 20°K ρ can be represented by $\rho = BT^n$, where $B = e^{-12.5}$ and $n = 4.19 \pm 0.06$; from 40 to 210°K ρ varies linearly with temperature. Hegland *et al.* measured both ρ_a and ρ_c as a function of temperature. These results are presented schematically in Fig. 2. The resistivity ρ_a shows a well-defined change in slope at $229 \pm 1^\circ\text{K}$; the change in slope begins at about 226°K . On the other hand, the resistivity ρ_c shows a sharp increase in slope at $220 \pm 1^\circ\text{K}$. The change in slope around 229°K is continuous.

In view of the above discussion, a plot of the polycrystalline resistivity of terbium as a function of temperature should exhibit changes in slope at about 220 and 229°K . Whereas the change in slope of ρ versus T at about 220°K is related to the discontinuity in the slope of ρ_c , the change in slope of ρ at 230°K is directly related to the change in slope of ρ_a . Therefore, the magnetic transition temperatures in terbium may be determined from the variation of its polycrystalline resistivity with temperature. Furthermore, the effect of pressure upon the magnetic transition temperatures may also be determined from a study of the effect of pressure upon the polycrystalline resistivity over the appropriate temperature range as is done in the present experiment.

EXPERIMENTAL PROCEDURE

Terbium of 99.9% purity was supplied by the Research Chemicals Company in the form of a disk 3.75 cm in diameter and 0.25-cm thick. Samples of about 1.75 cm in length with cross-sectional area varying between 0.01 and 0.05 cm^2 were cut from the disk. Variations in cross-sectional area were maintained below 0.0025 cm^2 over the entire length of the sample. The samples were annealed in vacuum for approximately 6 h at 800°C then slowly cooled to room temperature.

⁹ P. G. de Gennes and J. Friedel, *J. Phys. Chem. Solids* **4**, 71 (1958).

⁴ L. D. Jennings, R. M. Stanton, and F. H. Spedding, *J. Chem. Phys.* **27**, 909 (1957).

⁵ A. Arajs and R. V. Colvin, *Phys. Rev.* **136**, A439 (1964).

⁶ D. E. Hegland, S. Legvold, and F. H. Spedding, *Phys. Rev.* **131**, 158 (1963).

⁷ W. Boas and F. K. MacKenzie, *Progr. Metal Phys.* **2**, 90 (1950).

⁸ T. Kasuya, *Progr. Theoret. Phys. (Kyoto)* **16**, 58 (1956).

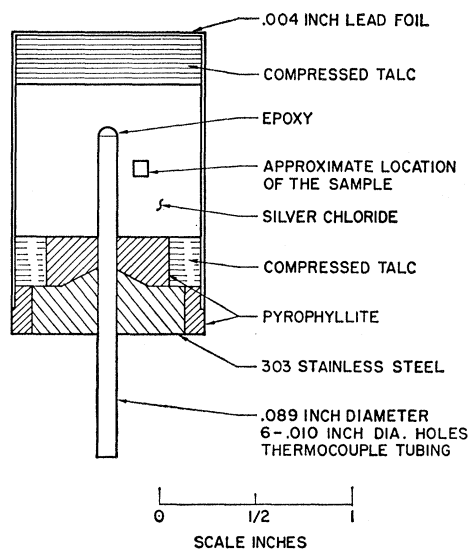


FIG. 3. Cross section of pressure capsule.

Sample resistance under pressure was measured using the standard four-probe method. A known dc current is passed through the sample and the voltage drop across the sample is measured. The resistance of the sample is determined using Ohm's law. The resistivity of the annealed sample was of the order of $7 \mu\Omega \text{ cm}$ at 4.2°K .

In order to determine the resistivity of the sample under pressure, the sample was embedded in a pressure capsule; a cross section of the capsule is shown in Fig. 3. Solid silver chloride serves as a quasi-hydrostatic-pressure transmitting media.

Six wires (two current leads, two voltage leads, and two thermocouple leads) were led out through a six-hole ceramic insulating tube. Pressures up to 15 kbar were obtained using a high-pressure vessel of the piston-cylinder type (Fig. 4). The sample was cooled by bringing the pressure capsule containing the sample in contact with liquid nitrogen. The sample was then allowed to warm up into the paramagnetic range.

Resistance measurements were made during this warm-up period. The warm-up rate was always maintained below $1^\circ\text{K}/\text{min}$ in the transition-temperature region. Resistance-measurements were made at 5-deg intervals above and below the transition region. Within the transition region, measurements were made at 1- and frequently at $\frac{1}{2}$ -deg intervals. Each run was made at a constant pressure within the range 0 to 16 kbar. The sample was always allowed to warm up above 275°K before the pressure was increased to the next value.

RESULTS AND DISCUSSION

The resistance as a function of pressure over the temperature range 200 to 240°K is presented in Fig. 5. The resistance at both ends of this temperature range is seen to vary linearly with temperature. The lower temperature at which the resistance begins to deviate from the linear behavior is taken as T_{ef} , whereas the upper temperature is taken to be T_N as illustrated in Fig. 6. The validity of this method of determining T_{ef} and T_N is demonstrated when noting that the present values of T_{ef} and T_N at 0 kbar are 223 and 231°K in good agreement with the values of Koehler *et al.* ($T_{ef} \sim 220^\circ\text{K}$ and $T_N = 230 \pm 1^\circ\text{K}$) and the values of Hegland *et al.* ($T_{ef} = 221 \pm 1^\circ\text{K}$ and $T_N = 229 \pm 1^\circ\text{K}$) obtained using other techniques. In any event, the primary point of interest here is the pressure coefficient of the Curie and Néel temperature rather than their absolute values. The variation of T_{ef} and T_N (obtained as outlined above) with pressure is presented in Fig. 7. T_{ef} and T_N appear to vary almost linearly with pressure. The average pressure coefficient of T_{ef} is -1.24 ± 0.1 and that of the Néel temperature is $-1.05 \pm 0.1^\circ\text{K}/\text{kbar}$.

A second method of determining T_{ef} and T_N from the resistance data was also used. The method is based on the fact that a change in the variation of resistance with temperature at the magnetic transition tempera-

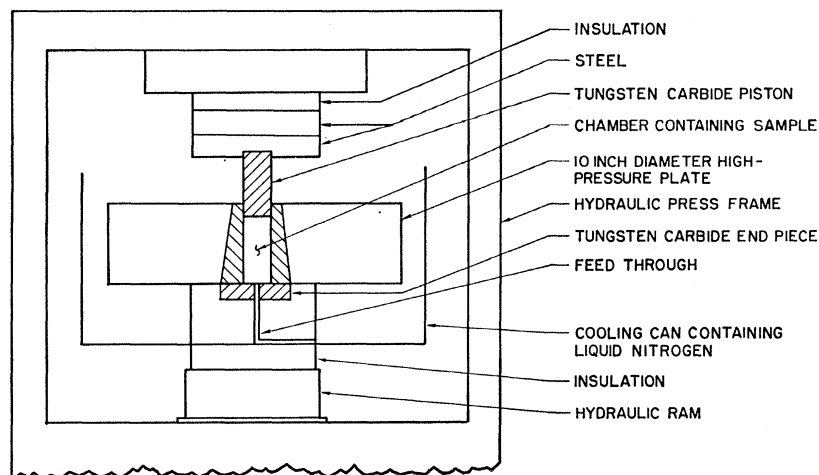
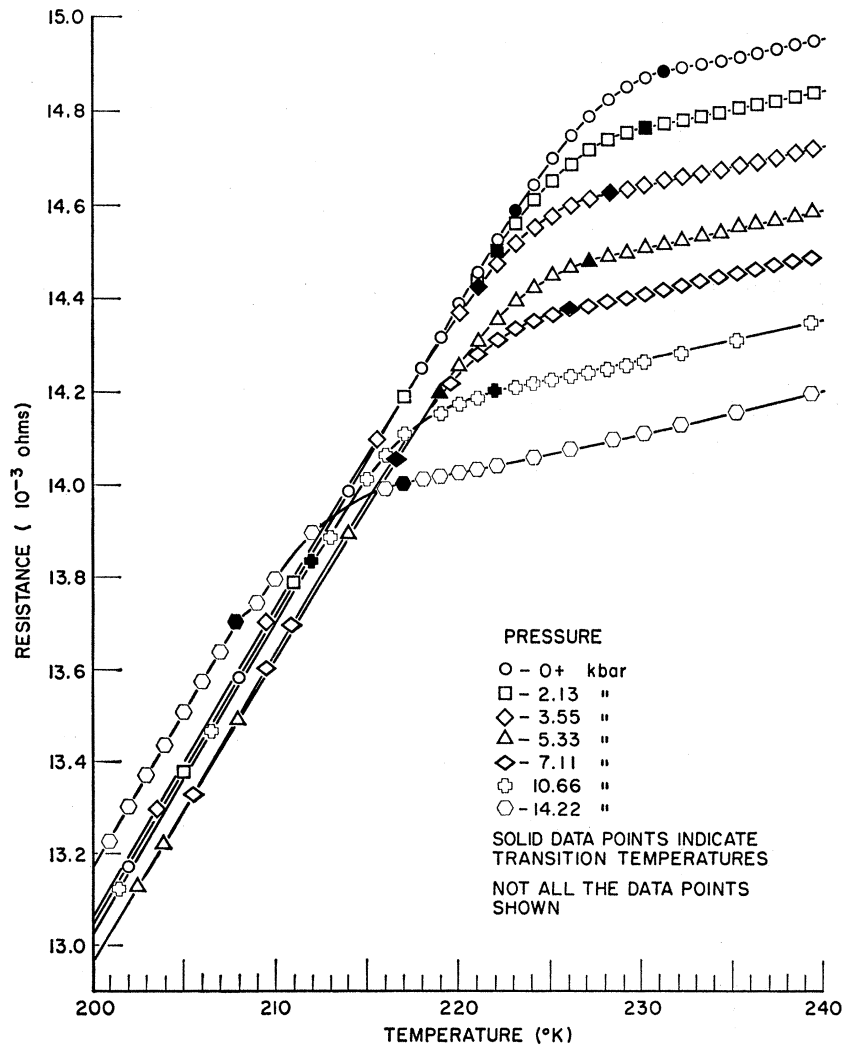


FIG. 4. Cross section of pressure die containing the pressure capsule.

FIG. 5. Electrical resistance as a function of temperature and pressure.



tures must clearly manifest itself in a well-defined change in the slope of the R -versus- T curve at these transitions. Therefore, plots of (dR/dT) versus temperature at several pressures were prepared and are presented in Fig. 8. A plot of (dR/dT) versus temperature is found to be well approximated by three linear segments as shown in Fig. 9. The transition temperatures T_{ef} and T_N were defined as the temperatures at the points of intersection of the three linear segments, Fig. 9. The validity of this method of evaluating T_{ef} and T_N is again demonstrated by noting that at 0 kbar this method again results in a value of $T_{ef}=222^\circ$ and $T_N=231^\circ\text{K}$. The variation of T_{ef} and T_N (evaluated from curves of dR/dT versus T) with pressure is presented in Fig. 10. T_{ef} and T_N appear to decrease linearly with increasing pressure up to a pressure of 14.22 kbar. According to this method of evaluating T_{ef} and T_N the pressure coefficient of the Curie temperature is $(-1.08 \pm 0.1)^\circ\text{K}/\text{kbar}$, and that of the Néel temperature is $(-1.14 \pm 0.1)^\circ\text{K}/\text{kbar}$. The

pressure coefficient of T_{ef} and T_N as determined from both methods is summarized in Table I. Results of other workers are included for comparison. The results of Figs. 5 and 8 indicate that the antiferromagnetic region is present to at least 14.22 kbar. Based upon the present results, it is evident that Robinson *et al.* and McWhan *et al.* measured the shift of T_N with pressure and that their experimental method was not sufficiently accurate to differentiate between T_{ef} and T_N .

It is to be recognized that the resistivity curves presented in Fig. 5 are also of the type associated with a single magnetic transition temperature. Therefore the above results may all be pertinent to a single transition temperature, perhaps T_N . This possibility, however, is quite remote. This is because, as shown in Fig. 9, the transition here occurs over an interval of temperature approximately equal to the difference between T_{ef} and T_N , nearly 7°K ; a transition associated with a single magnetic order-disorder would be

TABLE I. Pressure coefficients of the Curie and Néel temperatures of terbium.

Reference	Method	$^{\circ}\text{K}/\text{kbar}$ dT_{cf}/dP	$^{\circ}\text{K}/\text{kbar}$ dT_N/dP
Robinson <i>et al.</i>	Transformer		-1.0 ± 0.1
McWhan <i>et al.</i>	Transformer		-1.07 ± 0.03
Present	Resistivity: R versus T	-1.24 ± 0.10	-1.05 ± 0.1
	Resistivity: (dR/dT) versus T	-1.08 ± 0.10	-1.14 ± 0.1
Monfort <i>et al.</i> ^a	Compressibility	-0.7 ± 0.3	-0.7 ± 0.3
Bloch <i>et al.</i> ^b	Susceptibility		-0.82

^a C. E. Monfort and C. A. Swenson, *J. Phys. Chem. Solids* **26**, 623 (1965).

^b Daniel Bloch and Rene Pauthenet, in *Proceedings of the International*

Conference on Magnetism, Nottingham, 1964 (Institute of Physics and The Physical Society, London, 1965), p. 255-259.

more likely to occur over a smaller temperature interval of about 1 to 2 $^{\circ}\text{K}$. Second, and more important, let us recall that according to Eq. (2) we write

$$\frac{d\rho}{dT} = \frac{1}{3}(d\rho_c/dT + 2d\rho_a/dT).$$

If $d\rho_c/dT$ and $d\rho_a/dT$ are evaluated from the results of Ref. 6 on terbium, which are schematically presented here in Fig. 2, one obtains the curve shown in Fig. 11. This curve except for the discontinuity at T_{cf} is of the same general shape as the curves obtained by us and presented in Fig. 8. The absence of the discontinuity near T_{cf} is not surprising. This is because to detect a discontinuity of such magnitude requires very accurate and very detailed (with respect to temperature increments) measurements in its immediate neighborhood. In view of the above discussion it is felt that the present method of identifying T_{cf} and T_n is quite

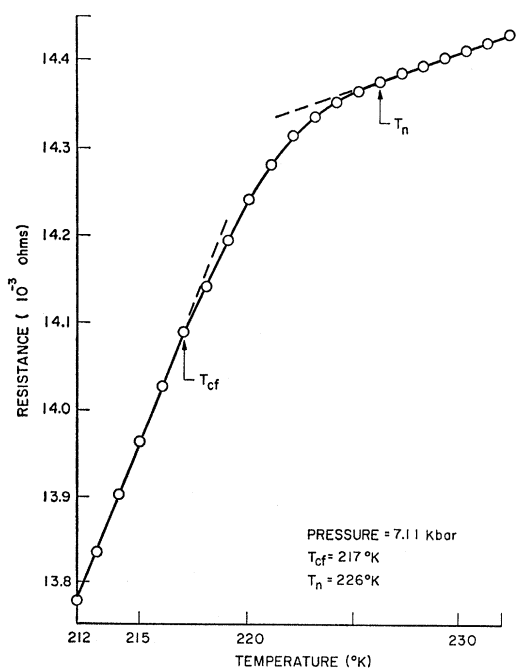


FIG. 6. Electrical resistance of a polycrystalline terbium sample as a function of temperature at a pressure of 7.11 kbar.

appropriate for the present analysis. This conclusion is further strengthened by noting that the values thus obtained for T_{cf} and T_n at 0 kbar are in good agreement with neutron-diffraction measurements.

The effect of hydrostatic pressure upon the resistivity of metals may be attributed to (1) a change in the interaction between the electrons and the lattice waves caused by a stiffening of the lattice, (2) a change in the lattice parameter and the associated change in the band structure and Fermi energy, and (3) crystallographic modifications and the accompanying changes in the band structure. These factors have an effect upon the magnetic transition temperature which in

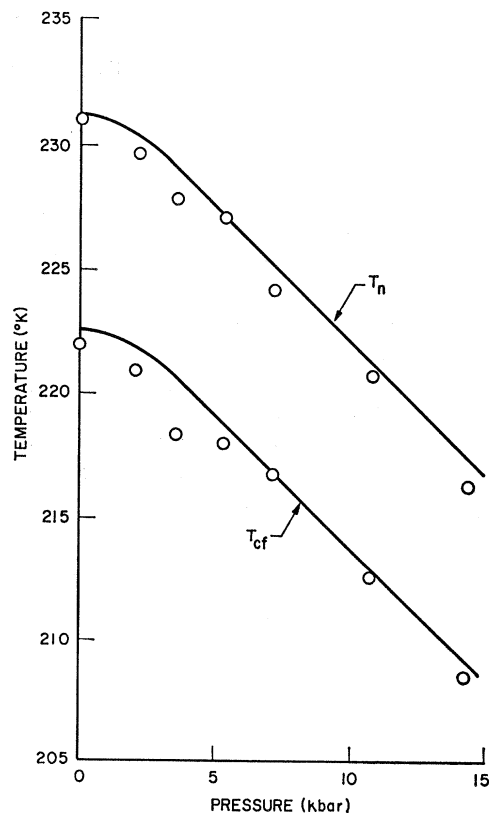


FIG. 7. The variation of T_{cf} and T_N with pressure (from R -versus- T plots).

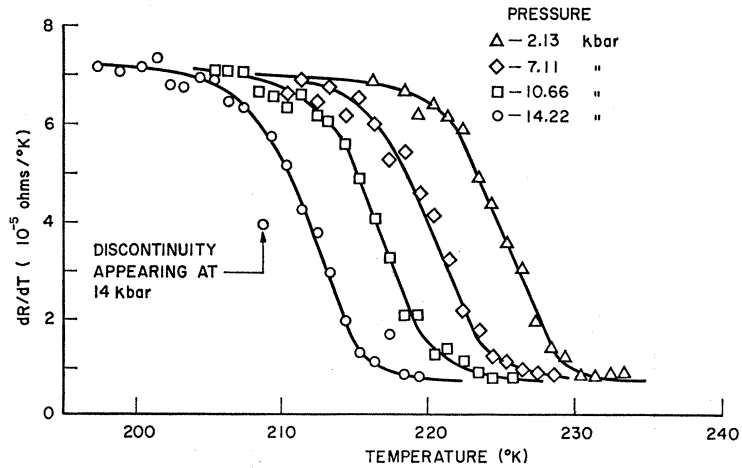


FIG. 8. Plots of dR/dT versus temperature at various pressures.

the rare-earth metals may be concisely stated by writing¹⁰

$$\frac{T'}{(g-1)^2 J(J+1)} = \frac{Z\pi}{k} \frac{V^2}{E_F} \sum_R \Phi(2k_F R), \quad (4)$$

where T' refers to either T_p or T_N , Z is the number of conduction electrons, k is Boltzmann's constant, E_F is the Fermi energy, k_F is the Fermi wave vector, V is an intra-atomic exchange integral similar to the atomic $6s-4f$ and $5d-4f$ exchange integrals, and the function Φ is of the form

$$\Phi(y) = (y \cos y - \sin y)/y^4.$$

An exact analysis of the effect of pressure upon expression (4) is a rather formidable task. Therefore, the effect of pressure upon the magnetic transition temperatures is carried out in the following simple manner.

The effect of pressure upon the Fermi energy or the Fermi wave vector k_F may be computed for the free-electron model. Freeman *et al.*,¹¹ however, recently

computed the Fermi surface for Tm and found it extremely complex and different from the free-electron model. In view of this difficulty the effect of pressure on k_F may be examined (as suggested by Milstein¹²) by studying the variation of k_F from one element to the other in the Gd-Tm rare-earth series. Yosida,¹³ based upon the Kohn anomaly, experimentally determined k_F in this series. The ratios of the actual value of the Fermi vector k_F to its value based on a free-electron model k_{F_0} as determined by Yosida for the Tm-Gd series, are given in Table II. It is noted that starting with Tm the Fermi vector k_F increases slightly, then becomes constant for Tb and Gd. Therefore, recalling that the c/a ratio increases in the series Tm-Gd, Table III, one may conclude that starting with Tm the Fermi wave vector increases slightly with the c/a ratio, becoming, however, independent of it in Tb and Gd. Furthermore, since the c/a ratio in Ho, Dy, Tb and Gd increases with pressure² we may, in view of the above statement, assume that starting with Tm the

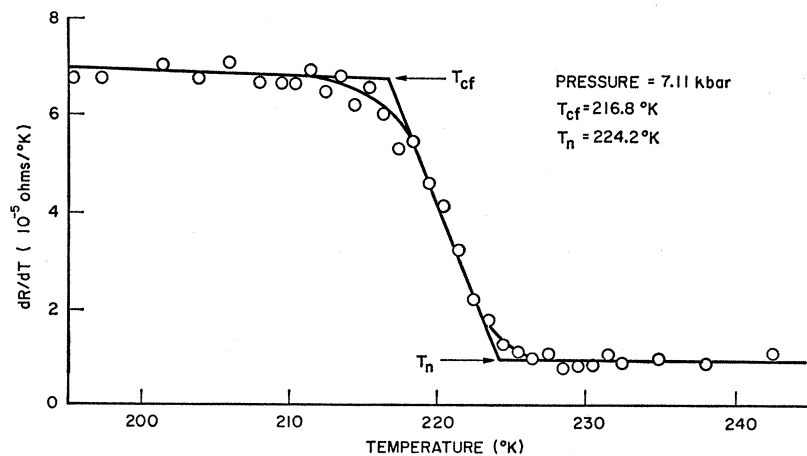


FIG. 9. Plot of dR/dT versus temperature at a pressure of 7.11 kbar.

¹⁰ R. J. Elliott, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1965), Chap. 7.

¹¹ A. J. Freeman, J. O. Dimmock, and R. E. Watson, *Phys. Rev. Letters* **16**, 94 (1966).

¹² F. Milstein, Ph.D. thesis, Department of Engineering, University of California, Los Angeles, 1966 (unpublished).

¹³ K. Yosida, in *Progress in Low-Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Company, Amsterdam 1964), p. 291.

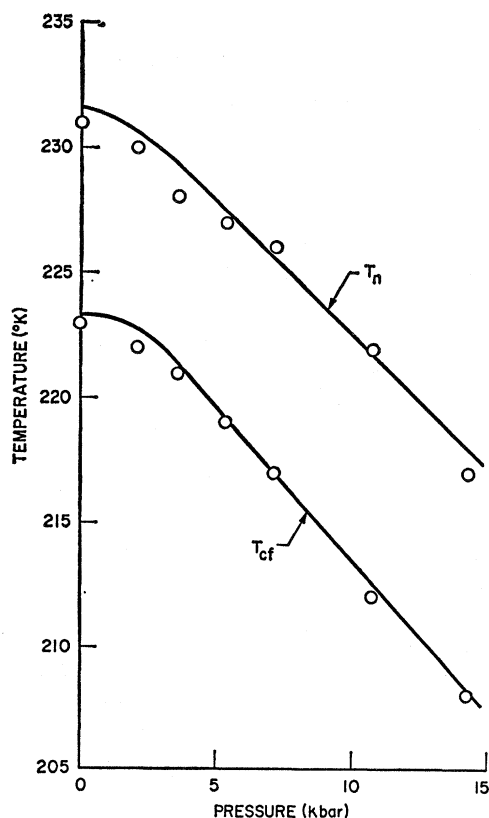


FIG. 10. The variation of T_{cf} and T_N with pressure (from dR/dT -versus- T plots).

Fermi wave vector increases with pressure, becoming independent of pressure in Tb and Gd. The same may also be said of the Fermi energy E_F , since $E_F = \hbar^2 k_F^2 / 2m$, where \hbar and m are constants.

The variation of the intra-atomic exchange integral V with pressure in Gd, Dy and Gd-Dy alloys has been studied by Milstein. Using a $4f$ - $6s$ type of exchange integral, he finds V to be relatively independent of pressure.

The function Φ that appears in Eq. (4) is a function of the Fermi wave vector k_F and R . According to Yosida (Table II), the value of k_F in terbium deviates only slightly from the value of the free-electron Fermi wave-vector k_{F0} . Therefore, the magnitude of k_F is assumed to be proportional to R^{-1} . Hence, the product $2k_F R$ which is the argument of Φ appears to be relatively

TABLE II. Fermi wave vector in the Gd-Tm rare-earth series after Yosida.^a

Element	Gd	Tb	Dy	Ho	Er	Tm
k_F/k_{F0}	1.045	1.034	1.021	1.006	0.966	0.948
k_F (\AA^{-1})	1.45	1.45	1.44	1.43	1.42	1.41

^a Kei Yosida, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Company, Amsterdam, 1964), p. 291.

TABLE III. Room-temperature lattice constants of Gd through Tm after Daane^a and Gschneider^b.

Element	c (\AA)	a (\AA)	c/a
Gd	5.7826 ± 6	3.6360 ± 9	1.5904 ± 4
Tb	5.6936 ± 2	3.6010 ± 3	1.5811 ± 2
Dy	5.6475 ± 2	3.5903 ± 1	1.5730 ± 1
Ho	5.6158 ± 2	3.5773 ± 1	1.5706 ± 1
Er	5.5874 ± 3	3.5588 ± 3	1.5700 ± 2
Tm	5.5546 ± 4	3.5375 ± 4	1.5702 ± 3

^a A. H. Danne, in *Physical Properties of the Rare-Earth Metals*, edited by F. H. Spedding and A. H. Daane (John Wiley & Sons, Inc., New York, 1961), Chap. 13.

^b Gschneider, Karl A., Jr., in *Crystallography of the Rare-Earth Metals*, edited by F. H. Spedding and A. H. Daane (John Wiley & Sons, Inc., New York, 1961), Chap. 14.

independent of the lattice parameter and thus Φ should be relatively independent of pressure.

Thus far we have arrived at a position where, on the basis of Eq. (4), one would conclude that the magnetic transition temperature T' is insensitive to pressure, in contradiction with the experimental measurements. This difficulty, in view of the credibility of the experimental results, must lie in the inadequacy of the treatment given (Eq. 4) above.

The variation of T' with pressure is reexamined in terms of phenomenological arguments. The exchange interaction that is currently accepted as underlying the theory of magnetism in the rare earths and that led to Eq. (4) is the well-known Ruderman-Kittel interaction. This interaction is essentially an indirect interaction between two rare-earth ions via a conduction electron. This interaction must be affected by the application of pressure because (1) the interionic separation and (2) the trajectories of the conduction electrons under pressure are certain to change.

The latter effect must be suspect of being overwhelmingly responsible for the change in the indirect exchange interaction with pressure; after all, the direct exchange interaction which is dependent upon the interionic separation is of minor consequence in the rare earths. In metals with hexagonal close-packed lattice the number of energy states per atom in the B.Z.

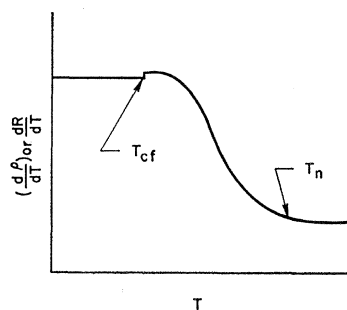


FIG. 11. Schematic of $d\rho/dT$ versus T for polycrystalline terbium.

TABLE IV. The ratio of the Néel temperature T_N to the de Gennes factor $(g-1)^2J(J+1)$ for the rare-earth elements Tb through Tm.

Element	$(g-1)^2J(J+1)$	T_N (°K)	$T_N/(g-1)^2J(J+1)$ (°K)
Tb	10.5	230	22.0
Dy	7.083	179	25.3
Ho	4.5	133	29.6
Er	2.55	80	31.4
Tm	1.065	56	52.6

(Brillouin zone) is a function of the c/a ratio¹⁴

$$n = 2 - \frac{3}{4}(a/c)^2 \left\{ 1 - \frac{1}{4}(a/c)^2 \right\}.$$

Furthermore in cases where the change in the c/a ratio is not accompanied by a phase change (i.e., the number of atoms per unit cell remain unchanged), the change in the number of states n is necessarily accompanied by a change in the electron occupancy (the density of states per energy level) within the B.Z. Thus, during an isotropic compression of a solid where the c/a ratio remains constant, the B.Z. simply expands leaving its shape unchanged. Under pressure, however, hexagonal metals such as terbium generally undergo anisotropic deformations, whereby the c/a ratio is changed. These anisotropic deformations are, as discussed above, accompanied by a change in the shape of the B.Z., a change in the number of states, and a change in the density of states per energy level within the B.Z. These changes combined essentially bring about a change in the energy states of the free electron. Hence, one can argue that since the indirect exchange-interaction is dependent upon the energy of free electron (magnitude and distribution) it must be a function of the c/a ratio. The variation of the indirect

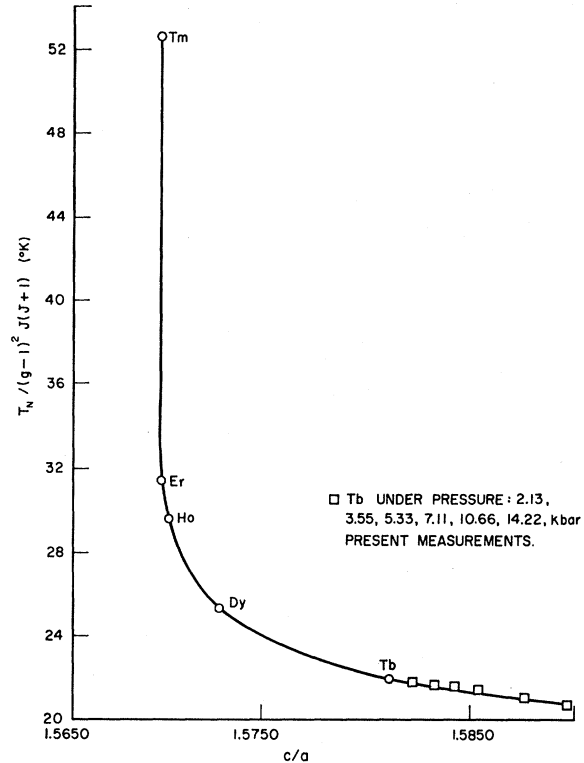


Fig. 12. Plot of $T_N/(g-1)^2J(J+1)$ as a function of the c/a ratio for the elements Tb-Tm.

exchange interaction or simply the $T_N/(g-1)^2J(J+1)$ ratio with pressure must therefore be largely due to the variation of the c/a ratio with pressure. The dependence of $T_N/(g-1)^2J(J+1)$ on the c/a ratio in a given phase, e.g., the antiferromagnetic phase in the rare earths, may be observed by noting the variation of

TABLE V. The Curie and Néel temperature of terbium as a function of pressure.

Pressure (kbar)	c/a	T_{ef} (°K)	$T_{ef}/(g-1)^2J(J+1)$	T_N (°K)	$T_N/(g-1)^2J(J+1)$ (°K)
From plots of R versus T					
0+	1.5811	223	21.24	231	22.00
2.13	1.5823	222	21.14	230	21.90
3.55	1.5833	221	21.05	228	21.71
5.33	1.5843	219	20.86	227	21.62
7.11	1.5854	217	20.67	226	21.52
10.66	1.5876	212	20.19	222	21.14
14.22	1.5897	208	19.80	217	20.67
From plots of (dR/dT) versus T					
0+	1.5811	222.0	21.14	231.0	22.00
2.13	1.5823	220.9	21.04	229.6	21.87
3.55	1.5833	218.3	20.79	227.8	21.70
5.33	1.5843	217.9	20.75	227.1	21.63
7.11	1.5854	216.8	20.65	224.2	21.35
10.66	1.5876	212.6	20.25	220.8	21.03
14.22	1.5897	208.6	19.87	216.4	20.61

¹⁴N. F. Mott and H. Jones, *The Theory of the Properties of Metals and Alloys* (Dover Publications, Inc., New York, 1958).

$T_N/(g-1)^2J(J+1)$ with the c/a ratio in the rare-earth elements that possess antiferromagnetic behavior.

Table IV presents the value of $T_N/(g-1)^2J(J+1)$ versus c/a ratio for the rare-earth elements Tb through Tm. These results are also presented in Fig. 12 versus c/a ratio. It is seen from Fig. 11 that if the c/a ratio in terbium is increased, the function $T_N/(g-1)^2J(J+1)$ decreases. McWhan *et al.* reported that the c/a ratio in terbium increases approximately 2.3% at a pressure of about 60 kbars. Jamieson¹⁵ also reports that the volume and the lattice constants of some rare-earth elements vary linearly with pressure to about 100 kbar. Therefore, in view of the lack of detailed data, we shall assume at present that the c/a ratio in terbium also varies linearly with pressure. The increase in c/a in terbium at a pressure P kbar is thus estimated at $(P/60)$ 2.3%. This expression is used to determine the c/a ratio in terbium at the various pressures employed in the present experiment. The results are included in Table V. The values of $T_N/(g-1)^2J(J+1)$ in terbium at the various pressures, that is as a function of c/a ratio, are also included in Fig. 11. The results are seen to be in excellent agreement with the variation of $T_N/(g-1)^2J(J+1)$ with c/a for the elements Tm through Tb. Therefore, it is not unreasonable at this

¹⁵ J. C. Jamieson, *Science* **145**, 572 (1964).

time to assume that the variation of the Rudermann-Kittel interaction or $T_N/(g-1)^2J(J+1)$ in the rare earths with pressure is due to the variation of the number of states and the density of states per energy level in the B.Z. with pressure, which is directly related to the variation of the c/a ratio with pressure.

CONCLUSIONS

1. The antiferromagnetic region in terbium appears to be present to at least 14.22 kbar.

2. The present results indicate that Robinson *et al.* and McWhan *et al.* measured the pressure-coefficient of the Néel temperature and not the Curie temperature.

3. The present results on the pressure-coefficient of the Néel temperature are in good agreement, within the accuracy of the experimental technique, with the results obtained using the transformer method.

4. As with the Néel temperature, the Curie temperature decreases with increasing pressure.

5. The function $T_N/(g-1)^2J(J+1)$ is a function of the c/a ratio.

6. The effect of the pressure upon the Rudermann-Kittel interaction or upon $T_N/(g-1)^2J(J+1)$ appears to be largely due to the effect of pressure upon the number of states and the density of states per energy level in the B.Z. which is directly related to the effect of pressure upon the c/a ratio.

Susceptibility-Derived Sublattice Magnetization in Antiferromagnets*

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A technique for deriving the sublattice magnetization in antiferromagnets from parallel-magnetic-susceptibility measurements is proposed. The relation between these quantities is derived by means of a molecular-field formalism. However, it is pointed out that the results should have a greater range of validity than the molecular-field theory itself. The theory is applied to the antiferromagnets MnF_2 , $CuCl_2 \cdot 2H_2O$, and FeF_2 and the results are compared to those available for these materials from nuclear-magnetic-resonance and Mössbauer-effect measurements as well as the usual molecular-field results. It is found that the susceptibility-derived results are in excellent agreement with those obtained by direct measurements. A reduction of an order of magnitude in the error between calculated and observed magnetization curves is obtained by using the susceptibility-derived results instead of the usual molecular-field results.

INTRODUCTION

IN order to calculate such statistical quantities as anisotropy, magnetostriction, magnetoelectric susceptibility, etc., in antiferromagnets, it is generally necessary to know the sublattice magnetization as a function of temperature. While by means of such

techniques as the Mössbauer effect, nuclear magnetic resonance (NMR), and neutron diffraction, a direct measurement of the sublattice magnetization may be made, the first two methods are useful only for special types of materials, while the latter, aside from requiring special equipment not commonly available, does not have high precision.

We here present a method for deriving the sublattice magnetization of a two-sublattice antiferromagnet from the experimental results for χ_{11} , the susceptibility parallel to the material's antiferromagnetic axis. In doing this, we make use of the suggestion of Callen and

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