

provided the temperature is low enough ( $\kappa$  is small enough), we obtain a lower bound of the form (4), independent of  $N$ .

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#### APPENDIX

Peierls gives an expression (3) in his original paper<sup>4</sup>:

$$(4\lambda)^L(1-4\lambda) \quad (\text{A1})$$

$$[\lambda = e^{-2J/kT} < \frac{1}{4}],$$

which is supposed to be an upper bound on the numbers of borders of length  $L$  passing through a given point. The reasoning leading to this result is, unfortunately, rather obscure; the result itself is incorrect, at least near  $\lambda = \frac{1}{4}$ . Since no border in a square containing  $N$  spins may have a length exceeding  $4N$ , it is clear that when  $\lambda$  is sufficiently close to  $\frac{1}{4}$ , (A1) implies that the probability of *any* border passing through a point is arbitrarily small. This cannot be correct.

The derivation of a similar expression at the top of p. 106 of Wannier's book<sup>4</sup> is unclear and the expression incorrect. When the temperature is sufficiently high the denominator diverges, and the probability of finding any border of finite length goes to zero.

## Thermal Conductivity and Electrical Resistivity of Terbium Between 5 and 300°K

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The thermal conductivity  $\lambda$  of polycrystalline terbium has been studied as a function of temperature  $T$  between 5 and 300°K. The  $\lambda$ -versus- $T$  curve exhibits a maximum of 0.205 W cm<sup>-1</sup> °K<sup>-1</sup> at 23°K. The antiferromagnetic-paramagnetic transition,  $T_{A-P}$ , causes an anomaly in the thermal conductivity at about 225°K. The ferromagnetic-antiferromagnetic transformation,  $T_{F-A}$ , because of the narrow antiferromagnetic region, is not observable from the  $\lambda$  versus- $T$  curve. According to the electrical resistivity data,  $T_{F-A} = 219 \pm 1^\circ\text{K}$  and  $T_{A-P} = 230 \pm 1^\circ\text{K}$ . The Lorenz function, calculated from the measured thermal conductivity and electrical resistivity values on the same sample, indicates that heat is transported mainly by electrons, with possible additional transport by phonons and magnons. The intrinsic electrical resistivity between 5 and 20°K is proportional to  $T^{4.19 \pm 0.06}$ .

#### INTRODUCTION

RECENTLY, we have initiated thermal conductivity measurements on the rare-earth metals from about 5 to 300°K in order to enlarge the knowledge of heat transport in substances exhibiting various magnetic states. Up to the present time such studies have been completed on dysprosium<sup>1</sup> and gadolinium.<sup>2</sup> In this paper we present our measurements on terbium with a discussion of their significance.

#### EXPERIMENTAL CONSIDERATIONS

The initial stock of terbium was obtained from Research Chemicals. This material was arc-melted for about 12 min. The partial analysis of the original terbium, provided by the supplier, is summarized in Table I. The electrical resistivity at 4.2°K before arc-melting was found to be 7.01  $\mu\Omega$  cm. After the melting a rod of diameter 0.572 cm and length about 8 cm was cut from the ingot. This rod was swaged to a diameter of 0.476 cm. A section of this material, about 6 cm long,

was wrapped in a tantalum foil, sealed into a silica capsule evacuated to  $10^{-5}$  mm Hg, and heat treated at 790°K for 40 h. After this procedure, the specimen was allowed to cool to room temperature in about 3 h. The electrical resistivity at 4.2°K of this specimen was 4.85  $\mu\Omega$  cm.

The thermal conductivity measurements, obtained with increasing temperatures from 5°K, were made using the apparatus described in detail elsewhere.<sup>1</sup> The electrical resistivities on the same sample with the thermal contacts used as potential contacts were made with the equipment briefly discussed before.<sup>3</sup>

TABLE I. Partial analysis of the initial terbium stock.

Impurities	Amount (weight %)
O <sub>2</sub>	0.08
Y	0.06
Ca	0.01
Si	0.01
Mg	0.003

\* Deceased 26 March 1964.

<sup>1</sup> R. V. Colvin and S. Arajs, Phys. Rev. **133**, A1076 (1964).

<sup>2</sup> S. Arajs and R. V. Colvin, J. Appl. Phys. **35**, 1043 (1964).

<sup>3</sup> S. Arajs, R. V. Colvin, and M. J. Marcinkowski, J. Less-Common Metals **4**, 46 (1962).

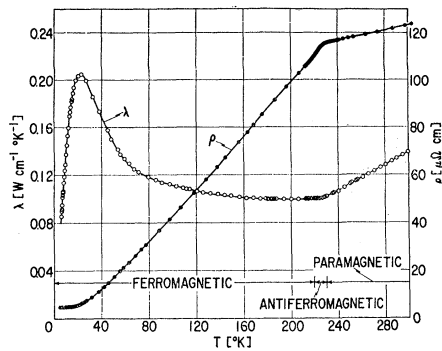


FIG. 1. Total thermal conductivity and electrical resistivity of terbium as a function of temperature.

### RESULTS AND DISCUSSION

Figure 1 shows the total thermal conductivity  $\lambda$  and electrical resistivity  $\rho$  of terbium as a function of temperature. The thermal conductivity first rapidly increases with increasing temperatures, reaches a maximum of  $0.205 \text{ W cm}^{-1} \text{ }^\circ\text{K}^{-1}$  at about  $23^\circ\text{K}$ , gradually decreases becoming practically independent of temperature between  $170$  and  $225^\circ\text{K}$ , and then increases with temperature above  $225^\circ\text{K}$ . The anomalous behavior at about  $225^\circ\text{K}$  is associated with the magnetic transformations. According to magnetization, electrical resistivity,<sup>4</sup> and neutron diffraction studies<sup>5</sup> on terbium single crystals, this metal is ferromagnetic below  $221^\circ\text{K}$ , antiferromagnetic between  $221$  and  $229^\circ\text{K}$ , and paramagnetic above  $229^\circ\text{K}$ . In the antiferromagnetic region terbium has a helical magnetic structure with the magnetic moments parallel to the basal plane and the

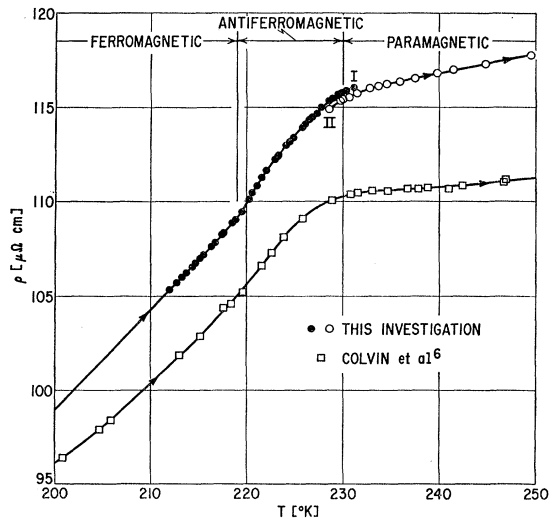


FIG. 2. Total electrical resistivity of terbium in the neighborhood of the magnetic transformations.

<sup>4</sup> D. E. Hegland, S. Legvold, and F. H. Spedding, Phys. Rev. **131**, 158 (1963).

<sup>5</sup> W. C. Koehler, H. R. Child, E. O. Wollan, and J. W. Cable, J. Appl. Phys. **34**, 1335 (1963).

$c$  axis as the screw axis. Due to the narrow antiferromagnetic range and the nature of the ferromagnetic-antiferromagnetic transition the  $\lambda$ -versus- $T$  curve instead of exhibiting two anomalies as in dysprosium shows only one irregularity centered at  $225^\circ\text{K}$ . It is believed that the increase in the thermal conductivity above this temperature is due to the change from the antiferromagnetic to the paramagnetic state. The ferromagnetic-antiferromagnetic transformation in polycrystalline terbium does not show a sharp anomaly in the electrical resistivity as can be seen in Fig. 2. Thus it can be expected that the thermal conductivity would also not alter significantly due to this particular change in the magnetic ordering.

According to the electrical resistivity data shown in Fig. 2, the magnetic transformations in terbium occur at  $219 \pm 1^\circ\text{K}$  and  $230 \pm 1^\circ\text{K}$ , respectively. The small shift in the upper resistivity curve resulted from an accidental warming of the sample at the end of the run (point I) to about  $295^\circ\text{K}$  and then cooling it to

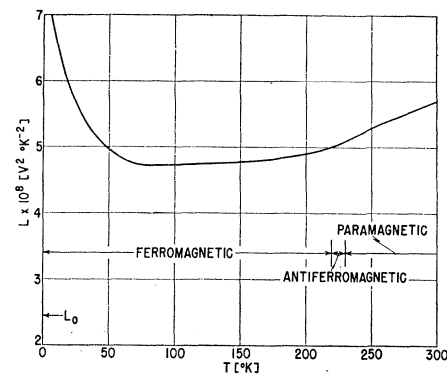


FIG. 3. Temperature dependence of the Lorenz function of terbium.

$228.5^\circ\text{K}$  (point II). Hysteresis effects of this type, associated with magnetic transformations, have been observed before. The lower resistivity curve is due to an earlier study<sup>6</sup> of another terbium sample. One can observe a great similarity between these two curves.

If the thermal conductivity is due to electrons only and if the relaxation time is the same for both the electric and thermal transport, then the Lorenz function

$$L = \rho\lambda/T \quad (1)$$

is a constant

$$L_0 = \pi^2 k^2 / 3e^2, \quad (2)$$

where  $k$  is the Boltzmann constant and  $e$  is the electronic charge. This should occur at low temperatures ( $T \ll \theta$ , where  $\theta$  is the Debye temperature) for the scattering of electrons by impurities and at high temperatures ( $T > \theta$ ) for the electron-phonon scattering. For intermediate temperatures the interpretation of  $L$  is more

<sup>6</sup> R. V. Colvin, S. Legvold, and F. H. Spedding, Phys. Rev. **120**, 741 (1960).

complicated because of the nonequality of the relaxation times for different transport processes. Figure 3 shows the temperature variation of the Lorenz function. The following observations can be made. First, the quantity  $L$  at low temperatures is higher than the value of  $L_0$  expected for pure electronic thermal conductivity. Second, the general behavior of the Lorenz function with temperature is anomalous in comparison to that of metals in which only electrons are responsible for the heat transport. Figure 4 presents

$$\lambda' = \lambda - (L_0 T / \rho) \quad (3)$$

at low temperatures for gadolinium,<sup>2</sup> terbium, and scandium.<sup>7</sup> The additional thermal conductivity above the electronic contribution is suspected to be due to phonons and possibly magnons, except for scandium which is not ordered magnetically at low temperatures. At the present time it is difficult to separate the magnon contribution from the lattice thermal conductivity. The temperature dependences of  $\lambda'$  for these elements, indicated in Fig. 4, are such that more than one scattering mechanism must be operating at any one temperature. The usual phonon-electron interaction gives  $T^2$  dependence in the lattice thermal conductivity at low temperatures. Phonon scattering by external and grain boundaries gives  $T^3$  dependence, stacking faults  $T$ , and point defects  $T^{-1}$  behavior. The theoretical understanding of the magnon scattering processes is very limited at the present time, especially in metallic ferromagnets. This makes further analysis of the conductivity  $\lambda'$  difficult.

Recently, Mackintosh<sup>8</sup> has suggested that the mag-

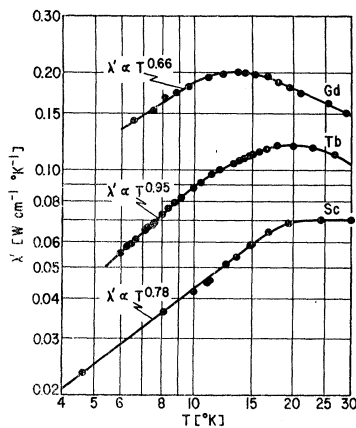


FIG. 4.  $\lambda'$  for gadolinium, terbium, and scandium at low temperatures.

<sup>7</sup> S. Araj and R. V. Colvin, in Proceedings of the Fourth Rare-Earth Conference, Phoenix, Arizona, 22-25 April 1964 (to be published).

<sup>8</sup> A. R. Mackintosh, Phys. Letters 4, 140 (1963).

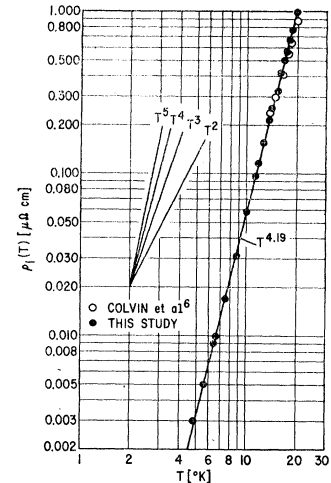


FIG. 5. Temperature dependence of the intrinsic electrical resistivity of terbium at low temperatures.

netic electrical resistivity of terbium at low temperatures should be

$$\rho_m = AT^2 \exp(-\delta/T), \quad (4)$$

where  $A$  is a constant and  $\delta = \Delta/K = 20^\circ\text{K}$ ,  $\Delta$  being the energy gap in the spin-wave spectrum. However, we found that our data of  $\rho_i$  between 5 and  $20^\circ\text{K}$  do not fit this equation if the electron-phonon and electron-electron resistivities are neglected. Empirically the intrinsic electrical resistivity of terbium in the temperature range 5 to  $20^\circ\text{K}$  can be represented by  $\rho_i = BT^n$ , where, according to a least-square determination,  $B = e^{-12.5}$  and  $n = 4.19 \pm 0.06$  (Fig. 5). The quantity  $n = 4$  is the prediction of Mackintosh for the magnetic resistivity of a ferromagnet with a linear spin-wave dispersion law. In order to test Eq. (4) fairly one should know more about the other electrical resistivity contributions in terbium at low temperatures. One could approximately estimate the electron-phonon resistivity for terbium from the intrinsic electrical resistivity of lutetium by using the Bloch-Grüneisen formulation. However, the presently available data<sup>6,9</sup> on lutetium are not of sufficient accuracy at low temperatures for this purpose.

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<sup>9</sup> S. Araj and R. V. Colvin, J. Less-Common Metals 4, 572 (1962).