

the average number of secondaries per primary displacement. The observed agreement was interpreted by Cooper, Koehler, and Marx as an indication that both the basic theory and the specific assumptions outlined above were essentially correct. In view of the present work, we feel that these conclusions need to be modified.

The present situation is summarized in Table I where the predicted ratios are tabulated for several different assumptions on the thresholds and resistivities. Following Seitz and Koehler the resistivity change per incident deuteron is taken proportional to $\bar{\nu}Z^2 \cdot \Delta\rho_f/T_a M$ where Z and M are, respectively, the atomic number and atomic weight of the bombarded metal. If assump-

tions (b) and (c) above are retained, the present thresholds predict values in substantial disagreement with experiment. However, if instead of (b) we assume either $\Delta\rho_f=C$ or $\Delta\rho_f=CA_0$, agreement is to some extent restored. While the agreement is satisfactory for Ag, the situation for Au is much worse. We therefore conclude that while the basic theory predicts the proper general trend, the noble metals are not identical as regards the damage process. It is necessary to consider each metal separately.

ACKNOWLEDGMENTS

We wish to acknowledge valuable discussions with W. A. Harrison.

PHYSICAL REVIEW

VOLUME 117, NUMBER 4

FEBRUARY 15, 1960

Electrical Resistivity of Dysprosium Single Crystals*

P. M. HALL, S. LEGVOLD, AND F. H. SPEDDING

Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa

(Received September 8, 1959)

Resistivity measurements are reported on two single crystals of hcp dysprosium metal in the temperature range 1.3°K to 400°K. The two magnetic transitions at 90°K and 175°K are very evident in the resistivity. Significant anisotropy is observed only above 175°K, which is the paramagnetic range; at 400°K, $\rho_{\perp}/\rho_{\parallel}=1.5$. A prediction for the resistivity of polycrystalline dysprosium based on these measurements is seen to be in good agreement with the resistivity of a polycrystalline sample.

INTRODUCTION

THE electrical resistivity of a hexagonal single crystal sample, in which the current flows at an angle ϕ with the c axis, is given by¹

$$\rho(\phi) = \rho_{\parallel} \cos^2\phi + \rho_{\perp} \sin^2\phi. \quad (1)$$

One can estimate the resistivity of a polycrystalline sample by averaging $\rho(\phi)$ over all solid angles. The result is

$$\bar{\rho} = (2\rho_{\perp} + \rho_{\parallel})/3. \quad (2)$$

Now for dysprosium, we assume the additivity of three contributions to the resistivity,

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

This assumption has been made by Kasuya² and by de Gennes and Friedel,³ and has been verified by Anderson and Legvold⁴ and Kondorsky et al.⁵ The first term in

Eq. (3) is assumed temperature independent. It is the resistivity extrapolated to 0°K. The second term is due to phonon scattering and theoretically is essentially linear in temperature when $T > 0.5\theta$, where θ is the Debye temperature. The third term is due to magnetic, or "magnon" scattering. It increases with the magnetic entropy, which is temperature independent above the highest magnetic ordering temperature.

Part of the interest in dysprosium (and in many of the other rare earths) is due to its three magnetic states.⁶ Below 90°K (the Curie point), it is ferromagnetic. Between 90°K and 175°K it is antiferromagnetic, and above 175°K (the Néel point), it is paramagnetic. However, it is felt that in the region 90–175°K the application of a magnetic field can cause a spin flip from antiferromagnetic to ferromagnetic alignment. Also, it has been found that dysprosium is magnetically very anisotropic in all three magnetic states, the hard direction being the c axis. Below 110°K there is anisotropy in the basal plane also, it being easier to magnetize along an a axis than along a direction 30° away from an a axis and in the basal plane. It was of interest, then, to see what anisotropy would occur in the magnetic resistivity, and how it would change as the sample goes from one magnetic state to another.

* Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission.

¹ W. Boas and J. K. Mackenzie, *Progr. in Metal Phys.* **2**, 90 (1950).

² T. Kasuya, *Progr. Theoret. Phys. Kyoto* **16**, 58 (1956).

³ P. G. de Gennes and J. Friedel, *J. Phys. Chem. Solids* **4**, 71 (1958).

⁴ G. S. Anderson and S. Legvold, *Phys. Rev. Letters* **1**, 322 (1958).

⁵ E. Kondorsky, O. S. Galkina, and L. A. Tchernikova, *J. Appl. Phys.* **29**, 243 (1958).

⁶ D. R. Behrendt, S. Legvold, and F. H. Spedding, *Phys. Rev.* **109**, 1544 (1958).

EXPERIMENTAL

Two crystals were grown under argon pressure in a slightly modified Bridgman technique such as that used by Behrendt, *et al.*⁶ A few grams of redistilled dysprosium were melted in a 16 cm long, 3-mm diameter tantalum crucible, which was then steadily lowered out the bottom of the heated portion of a furnace at one cm an hour. Two crystals were grown in this manner. They were shaped into rods with rectangular cross sections and dimensions (in mm) of $2.0 \times 2.3 \times 20.3$ and $1.5 \times 2.1 \times 14.9$.

The resistivities of these samples were measured from 1.3°K to 400°K in a cryostat built by Colvin of this laboratory.⁷ A four-probe system was used to measure the resistivity, the potential contacts being 9.9 mm apart for the longer sample, and 6.8 mm for the shorter one. Also measured was a rod of polycrystalline dysprosium, cast from the same batch of metal as were the single crystals. The rod was 4.8 mm in diameter and 5.1 cm long, and its potential contacts were 2.5 cm apart. Some measurements were made in a transverse magnetic field also, but the effect of a magnetic field was detectable only in the antiferromagnetic range, so these measurements were made only between 81°K and 181°K.

The probable errors are estimated to be 0.1 microhm-cm at very low temperatures, and 1.5% of the resistivity at room temperature. The temperature was controlled and measured to within one half of one degree at all times. The estimated purity from spectrographic analysis was 98.7%, the major contaminant being about 1% of tantalum, which was dissolved from the crucible in growing the crystals. The remaining impurities were mainly O₂, C, and N₂, the amounts reported being 130, 100, and 150 ppm, respectively. Metallic impurities (including other rare earths) were not detected spectrographically.

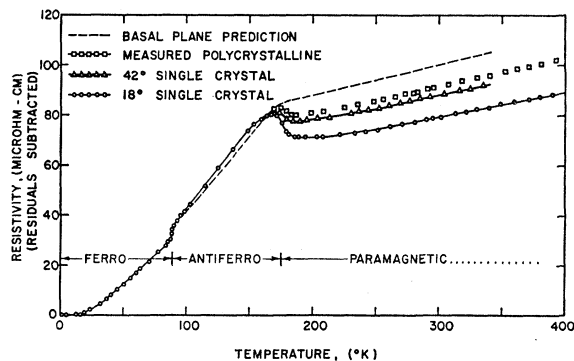


FIG. 1. Electrical resistivity of dysprosium. The lowest curve represents the crystal oriented 18.2° from the *c* axis. The next lowest is for a crystal 42.0° from the *c* axis. The squares are the measured resistivity of a polycrystalline sample. The dashed line is a prediction for the basal plane resistivity.

⁷ R. Colvin, thesis, Iowa State University, Ames, Iowa, 1958 (unpublished).

RESULTS

The residual resistivities were 2.4, 3.1, and 5.1 microhm-cm for the polycrystalline sample, the crystal oriented 18.2° from the *c* axis, and the crystal oriented 42.0° from the *c* axis, respectively. These residual resistivities were subtracted from all the data to obtain the results shown in Fig. 1. The circles are the experimental points for the 18.2° sample. The triangles represent the 42.0° sample, and the squares correspond to the polycrystalline sample. The dashes are a prediction for ρ_1 . They represent an average of two predictions, one using Eq. (1) and the data from the two crystals, the other using Eqs. (1) and (2), the polycrystalline data and the 18.2° crystal data. For purposes of clarity, all but two of the curves have been omitted below 160°K; below 90°K, only the 18.2° crystal data are shown, because all the curves would be nearly identical. Table I is a list of all the measured and predicted resistivities at 300°K. The predicted polycrystalline curve and the calculated *c*-axis curve have been omitted from Fig. 1, but the table will show where these curves come in the paramagnetic region.

TABLE I. Resistivities of dysprosium samples.

300°K (microhm-cm, residual subtracted)	
1. 42.0° crystal	88.2
2. 18.2° crystal	79.2
3. Predicted polycrystalline from 1 and 2	93.9
4. Measured polycrystalline	91.2
5. Predicted ρ_1 from 1 and 2	102.4
6. Predicted ρ_1 from 2 and 4	98.2
7. Average ρ_1 from 5 and 6	100.3
8. Calculated ρ_{11} from 2 and 4	77.4

The curves show clearly the three regions of magnetic behavior. At about 89°K there is an abrupt increase in the resistivity with increasing temperature, marking the transition from ferromagnetic to antiferromagnetic ordering. An unusual feature of the data is the anomalous behavior associated with the Néel point. The effect is particularly pronounced in the case of the 18° crystal, whose resistivity decreases by 11% when the temperature is raised from 167°K to 193°K. Above 200°K, the resistivity is essentially linear with temperature, as would be expected for a paramagnetic metal. It is to be noted that the only really significant anisotropy is in the paramagnetic region. Also, it should be noted that the curves are practically parallel in this region. If they are extrapolated to 0°K, they do not meet each other, as might be expected if the anisotropy were due to the phonon part of the resistivity.

Figure 2 is merely an expanded view of the region near the Curie point. It gives an idea of the sharpness of the transition and of the amount of scatter in the data. Also, it shows that the Curie point is slightly different for each of the three samples. This could very well be due to differences in the impurities in the samples, since the

sample with the lowest Curie point had the highest residual resistivity, and vice versa. Thoburn *et al.*⁸ have shown that it does not take much nonmagnetic impurity to lower the Curie point. Also shown on Fig. 2 (closed circles) is the resistivity of the 18.2° crystal with a constant transverse external magnetic field of 18.9 kilogauss, showing how the anomaly can be completely removed. The magnetoresistance effect thus produced exhibited a behavior much like that of the magnetic susceptibility in that it took a certain critical field to get any effect, after which the resistivity decreased essentially linearly with the field, until (when the transition was complete) the effect became saturated. The temperature dependence of the magnetoresistance was also similar to that of the magnetic susceptibility in the region from 90° to 180°K.

DISCUSSION

The abrupt increase in the resistivity as the sample is moved from the ferromagnetic region into the antiferromagnetic region must be due to an increase in the magnetic scattering, and, it is believed, an increase in the magnetic entropy. This would mean that the magnetic transition is a first order transition. Now the specific heat data⁹ show a peak at the Curie point, but it is not a delta function, as would be expected for a first order transition. It is approximately symmetrical, but about four degrees wide at the half height. However, if there were a statistical spread of Curie points due to inhomogeneities, then the result would be a spread out first-order transition such as is observed. The shift in entropy associated with the area under the specific heat peak at this point is about 0.1 cal (g atom)⁻¹(°K)⁻¹, or about 5% of the total magnetic entropy at this point. In the polycrystalline sample, a shift of about 8% is observed in the resistivity. It is felt, then, that the change from ferromagnetic to antiferromagnetic ordering causes a decrease in the magnetic order, which must mean a decrease in the sublattice magnetization. However, this order is completely regained when the critical magnetic field is applied. It is also felt that the increased entropy stays with the sample until the neighborhood of the Néel point is reached, because the shift in resistivity is observed all the way up to 175°K. It is felt that with sufficiently high magnetic fields, the peak could be completely eliminated, and the curve would

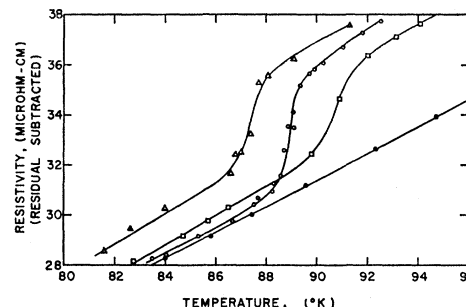


FIG. 2. Electrical resistivity of dysprosium near the Curie point. The open circles are the 18.2° crystal. The squares are the measured polycrystalline sample. The lowest curve is the 18.2° sample with a magnetic field applied, and the triangles correspond to the 42.0° crystal.

look like an ordinary ferromagnetic resistivity curve. This is not surprising, since then there would be only the two magnetic states possible, the ferromagnetic and the paramagnetic.

It still remains to account for the anisotropy observed. It is known⁶ that the magnetic susceptibility is anisotropic above the Néel point. In fact, the Curie plots of the paramagnetic susceptibility for the *c* axis and basal plane are parallel, but have different paramagnetic Curie points. θ_c is 121°K for the parallel, and 169°K for the perpendicular susceptibility. This was explained by Behrendt¹⁰ on the basis of anisotropic crystalline fields which would act, in effect, like an "anisotropic" exchange integral. And Kasuya² has shown that the magnetic part of the resistivity in the paramagnetic range should be proportional to the square of the effective exchange integral. If we apply this to an anisotropic exchange energy, we might expect the magnetic resistivity in the different directions to be proportional to the square of the paramagnetic Curie temperature. That is,

$$\rho_{11}/\rho_{\perp} = (\theta_{c11}/\theta_{c\perp})^2. \quad (4)$$

In fact, the right-hand side of Eq. (4) is 0.52, while the left-hand side is about 0.8. This is not very good agreement, but at least it is in the right direction, which is all one could expect from so naive an approach.

ACKNOWLEDGMENTS

The authors are indebted to Dr. A. H. Daane for supplying the distilled dysprosium, and to Mr. Richard Colvin for the polycrystalline resistivity data.

⁸ W. C. Thoburn, S. Legvold, and F. H. Spedding, Phys. Rev. **110**, 1298 (1958).

⁹ M. Griffel, R. E. Skotchdopole, and F. H. Spedding, J. Chem. Phys. **25**, 75 (1956).

¹⁰ D. R. Behrendt, thesis, Iowa State University, Ames, Iowa, 1956 (unpublished).