Magnetoresistance in dysprosium single crystals

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The magnetoresistance in the ferromagnetic phase of the Dy *a*-, *b*-, and *c*-axis crystals has been investigated in magnetic fields up to 75 kOe and in the temperature range between 1–70 K. It is anisotropic with respect to the crystallographic axes and to relative orientations of the field and the current. The longitudinal isotherms exhibit a minimum which increases in depth and moves to higher fields as the temperature increases. The transverse magnetoresistance in the basal-plane crystals for fields above 20 kOe increases with the field in the entire temperature range. But for the *c*-axis crystal ($\vec{I} \parallel \langle 0001 \rangle$ and $\vec{H} \parallel \langle 11\bar{2}0 \rangle$ or $\langle 10\bar{1}0 \rangle$) above 25 K, it decreases as field increases. These results have been discussed on the basis of scattering due to impurities, phonons, magnetic domains, and spin waves, and in terms of magnetic anisotropy of the metal. When the field is parallel to the *b* axis, the transverse-resistance isotherms around 20–40 K show a resistance maximum of about 0.2% at a field which gradually decreases as temperature increases. The critical field for the spin-wave energy gap around 40 K, as estimated from the resistance maximum is about 10 kOe.

INTRODUCTION

Dysprosium is antiferromagnetic in the temperature range 179–85 K and is ferromagnetic below 85 K. In the first state, the magnetic moments in each basal plane are ordered ferromagnetically, but the direction of magnetization rotates while going from one plane to another plane, and as a consequence the ordering is helical antiferromagnetic. This structure has been explained on the basis of the three-plane model in which the exchange interaction between the nearest-neighboring planes is opposite in sign to that between the next-nearest planes.[1] The helical-antiferromagnetic state of the metal has been studied using magnetoresistance measurements by Mackintosh and Spanel,[2] Wilding and Lee,[3] and recently by Akhavan *et al.*[4] The ferromagnetic state has a high magnetic anisotropy with an easy direction of magnetization parallel to the *a* crystal axis. The anisotropy is due to crystal-field and magnetostriction effects, and it introduces an energy gap Δ for excitation of magnons, since a finite energy is required to turn a spin against the anisotropy field. Around 4 K, $\Delta/k_B \approx 34$ K (k_B is the Boltzmann constant) and gradually decreases as temperature increases. The gap is isotropic, but the changes induced by an external field are anisotropic and have recently been calculated by Mackintosh and Møller.[5] The results show that if a field is applied parallel to the easy axis (*a* axis) the gap monotonically increases, but it passes through a minimum for fields along the hard direction (*b* axis) in the basal plane. Further, the minimum of the gap is lower than its zero-field value, i.e., $\Delta(H_C) < \Delta(0)$. The field at the minimum is known as the critical field (H_C) and decreases as the temperature approaches the Curie temperature. It has been suggested that the presence of a gap will modify various temperature-dependent properties of the metal such as magnetization, spin-wave resistivity, etc.[5] In the ferromagnetic state, magnetization measurements on single-crystal Dy have been reported by Behrendt *et al.*,[6] Jew and Legvold,[7] and Rhyne *et al.*[8]; resistivity results have been reported by Hall *et al.*[9] and many other workers. In this paper, we report magnetoresistance results in the ferromagnetic state of several Dy single crystals.

EXPERIMENTAL PROCEDURE

The single crystal (purity 99.8 wt%) of cylindrical form with its long axis parallel to the *c* crystallographic axis was obtained from Metals Research Ltd. The basal-plane-orientation samples were obtained from the cross section of this *c*-axis crystal. All the crystals were aligned using Laue back-reflection techniques and specimens were spark cut in the form of a rectangular parallelepiped. The typical thickness of the samples was about $\frac{1}{3}$mm, and their length varied from 3 to 11 mm. The samples, after spark cutting, were etched and annealed in vacuum at 600°C for 10 h. Four pieces of fine platinum wire were spot welded on the specimen for making electrical contacts. The specimen was mounted on a small copper block which was insulated on the surface by a thin layer of GE varnish. This block was then varnished on another copper plate which had the temperature sensors and a miniature heater for the temperature controller. The resistance ratio $R(300)/R(4)$ of the specimens lies between 20 and 24 and the resistivity at 4 K is about 4 $\mu\Omega$cm.

The sample resistance was measured by a four-

probe method using a photochopper technique developed for precise magnetoresistance measurements. Briefly, dc currents through the sample and reference resistors, which are electrically isolated, are chopped synchronously using a dual photochopper which is operated at 9 Hz. The sample voltage is compared with a variable voltage across a reference resistance which is at room temperature, using a lock-in amplifier as a null detector, and the resistance is given as $R=(I_r/I_s)$ R_r, where I_r/I_s is the ratio of the currents in the reference and sample circuts at null, and R_r is the reference resistor, which is constant. Thus, the current ratio is a measure of the variation of the sample resistance and was measured using two voltage-to-frequency converters and an electronic counter.[10] The sample temperature was measured using a calibrated germanium resistance thermometer, and a copper-constantan thermocouple. For the measurements below 1.5 K, a calibrated gold-iron (0.03%)–chromel thermocouple and a capacitance thermometer[11] were used. The sample temperature in the presence of a magnetic field was established using the field-insensitive capacitance sensor as a secondary thermometer. The errors in the temperature control are within the calibration error of the germanium sensor, which is less than 10 mK for $T<20$ K and gradually rises to about 100 mK for higher temperatures.

RESULTS AND DISCUSSIONS

We have measured the changes in the resistivity as a function of temperature of some Dy crystals from 1.2 to 50 K, with a precision of better than 1 part in 10^{-4}. The increase in the resistivity due to temperature can be expressed empirically as

$$[R(T)-R(1.2)]/R(1.2)\propto T^m, \qquad (1)$$

where R is the sample resistance. For the temperature range 6–25 K, a nonlinear least-squares fit gave $m\simeq 3.1$ for the a and b axes, and $m\approx 3.8$ for the c axis. At higher temperatures m gradually decreases. It should be emphasized that the resistivity data show a smooth and monotonically increasing temperature variation in the neighborhood of $T_0=\Delta/k_B$ for a-, b-, and c-axis crystals. There is no anomaly of the type reported by Sze *et al.*[12] for a Tb c-axis crystals. Also, our results on the Tb single crystals[13] do not show any anomalous contribution either. Thus, information about the gap cannot be directly obtained from the resistivity data around T_0. This is consistent with the theoretical expressions,[5,13] which predict a gradual variation in the spin-wave resistivity around T_0. For temperatures below 4 K, the resistivity results show an anomalous behavior which will be discussed later.

When the sample is first magnetized, its resistance is altered even though the applied field is reduced to zero. But after the first magnetization, the resistance in zero field ($H=0$) is reproducible on further field cycling provided its direction is unchanged. This change in the resistance depends on the crystal axis, the direction of the magnetizing field, and the direction of the current. When the sample is magnetized parallel to its length, the resistance decreases and at 1.2 K the change is 3.6% and 1.7% for the a- and b-axis crystals, respectively. For the transverse magnetization (H is parallel to the short dimension of the sample, and $\vec{I}\perp\vec{H}$), however, the change in resistance is relatively small. It increases by about 0.4% for the a and c axes but it decreases by almost the same amount for the b-axis crystal. This change in resistance, in our view, is associated with domain reorientation. In order to compare various isotherms, all the results reported here were taken after the samples had been magnetized. Further, the change in resistance is plotted in terms of current ratios (I_r/I_s), since precise measurements of the shape factors for our specimens are difficult.

Figure 1 shows some of the longitudinal magnetoresistance isotherms for the a-axis crystal. The general features of the isotherms are, first, as field increases the resistance initially decreases and finally passes through a minimum and, second, the depth of the resistance minimum $[R(H=0)-R(H_m)]$ and the field at the minimum increase with temperature. The isotherms around 1 K show a hysteresis effect, and typical results at 1.2 K are shown in the figure. If the direction of the field is the same as the previous direction of magnetization, then the isotherm shows a sharp change in the rate of decrease of the resistance at around 5 kOe; but for the reverse direction of the field, it exhibits a small maximum (0.4%) at the above field. The results at higher fields are independent of the direction of the field. As the temperature increases, the hysteresis in the magnetoresistance decreases and above 6 K it is negligible. At 1.2 K, the depth of the minimum is 2%, and it appears around 47 kOe.

In magnetic metals, the usual resistivity contributions are due to impurities and lattice imperfections, magnetic domains, phonons, and spin waves. The first two contributions are weakly temperature dependent and can account for most of the total resistivity around 1 K. When a field is applied, the domain tends to realign and as a result this resistivity rapidly decreases at low fields but approaches some field-independent value at higher fields. The above realignment is also

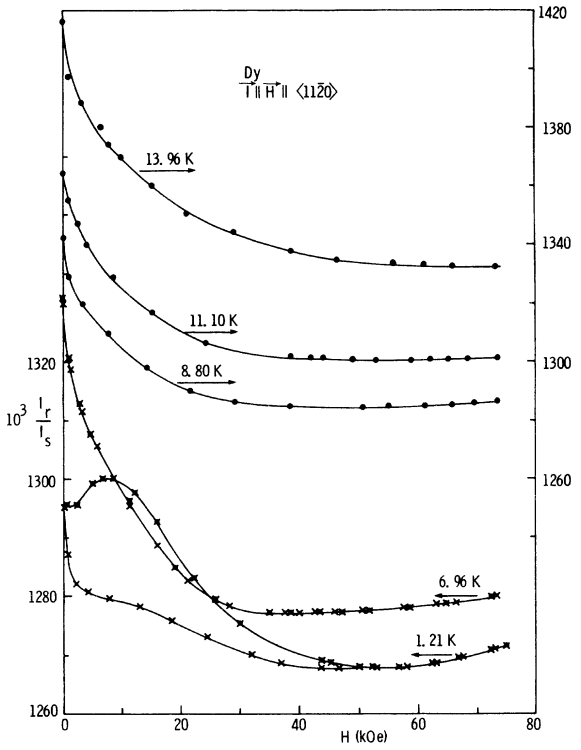

FIG. 1. Changes in the longitudinal magnetoresistance vs applied field for the ⟨11$\bar{2}$0⟩ crystal. I_r/I_s is the current ratio in the sample and reference circuits at null and is proportional to the sample resistance. The bottom isotherm shows the hysteresis in the magnetoresistance; the × represent data for the field parallel to the previous direction of magnetization and the crossed full circles are for the antiparallel direction. Above 6 K, the hysteresis is negligible.

responsible for a rapid increase in magnetization at low fields.[6,7] On the other hand, the nonmagnetic scattering in the presence of a magnetic field gives an increase in the resistivity due to the Lorentz force on conduction electrons. This increase is usually referred to as the normal magnetoresistance. Thus, the resistance minimum around 1 K is a result of the changes in domain resistivity and the increase in the normal magnetoresistance.

At higher temperatures, scattering of electrons by phonons and by spin waves gives rise to the temperature-dependent part of the resistivity. When a field is applied, the electron-magnon scattering is reduced since most of the low-energy magnons are suppressed by the field. This additional decrease in the resistivity seems to be responsible for both the increase in the depth of the resistance minimum, and the increase in the field at the minimum. Around 14 K, the depth of the minimum is 6% of the resistance in zero field ($H=0$) and it appears around 75 kOe.

Figure 2 shows some of the longitudinal magnetoresistance isotherms for the b-axis crystal. In contrast to the a-axis results, the hysteresis effect around 1 K for the b axis is very small ($\simeq 0.05\%$). The qualitative nature of the b-axis isotherms is similar to those shown in Fig. 1; however, the depth of the minimum is relatively smaller and occurs at lower fields. For example, at 1.2 K, the depth of the minimum in this case is 0.14% and it appears around 9 kOe. This implies that either the preferred direction for domain alignment is along the b axis and so its resistivity contribution reaches a field-independent value at a much lower field, or the normal magnetoresistance for this orientation has a stronger field dependence; or perhaps a combination of both. The domain structure in a few dysprosium crystals has recently been reported, which seems to suggest the preferred direction of the alignment is along the b axis.[14] However, this is inconsistent with magnetization measurements, which imply an easy direction along the a axis.[6] Further work on the domain structure in the presence of the magnetic field is desirable to resolve this point. In the temperature range 6–18 K, the depth of the resistance minimum rises as $T^{2.9}$.

The total resistivity is usually taken as the sum

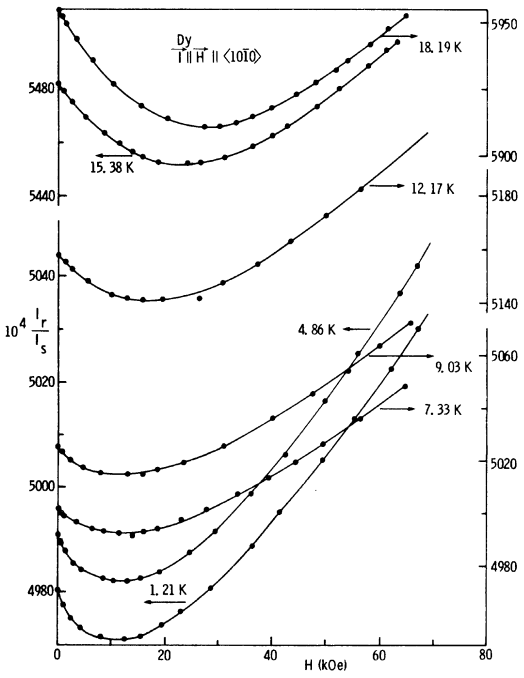

FIG. 2. Changes in the longitudinal magnetoresistance vs applied field for the ⟨10$\bar{1}$0⟩ crystal. The increase in the depth of the resistance minimum is due to a reduction in the inelastic electron-magnon scattering.

of its components and can be expressed as

$$\rho = \rho_i + \rho_d + \rho_p + \rho_s, \tag{2}$$

where successive terms are due to scattering of conduction electrons by impurities, domains, phonons, and spin waves. As mentioned earlier, around 1 K, the contributions due to phonons and spin waves are negligible compared to other scattering and changes in the resistance due to the field are attributed to the impurities and domains.

It has been recognized for some time that the domain structure affects the resistivity of a ferromagnetic metal, but its mechanism is not well understood at the present time. Two important sources are the magnetoresistance due to the field within the domain[15] and the scattering by domain boundaries.[16] The first effect is due to the fact that even in the absence of any external field, electrons are moving in the effective field within the domain. This local field, in a multidomain state, would be oriented at different angles with respect to the current and as a consequence the magnetoresistance would be the sum of longitudinal and transverse components. When the sample is magnetized parallel to the current, the resistance decreases because the transverse component reduces to the longitudinal contribution. Further, this implies that the domain resistivity would increase if the sample is magnetized normal to the current direction. The second mechanism is due to diffuse scattering of conduction electrons at the domain boundaries, which can act as internal surfaces for the crystal. For the present crystals, the ratio of domain size to the electron mean free path is of the order of 10^2 and so the contribution due to boundary scattering is expected to be much smaller. Beside the above mechanisms, there may be some contribution due to spontaneous anisotropy of the resistivity, which is usually taken as a difference between the longitudinal and transverse resistivities extrapolated to $B=0$. This arises because of anisotropic scattering of the conduction electrons by the localized spins, but the exact mechanism is still debatable.[17,18] The domain structure is sensitive to magnetic anisotropy, demagnetizing field, strains, etc., and experimentally it is difficult to isolate the above contributions.

The increase in the normal magnetoresistance can be described by a semiempirical relation known as the Kohler's rule,[19] which states that

$$\Delta\rho/\rho = F(B/P), \tag{3}$$

where $\Delta\rho/\rho$ is the fractional increase in the resistance due to a field B $(=H+4\pi M)$. The function F depends on the electronic properties of the metal, relative orientations of the field, the current direction, and the crystal axis. It should be remarked that application of Kohler's rule in a ferromagnetic metal is not straightforward because the resistance in zero field is experimentally inaccessible. For the purposes of the qualitative discussions, however, the magnetoresistance can be expressed as $\Delta R = aB^n$. Thus, the resistance as a function of field around 1.2 K can be expressed in a dimensionless form as

$$R(H)/R_0 = R_d(H)/R_0 + (1/R_0)(R_e + aB^n), \tag{4}$$

where R_d is the field-dependent part of the domain resistance, and R_e is the sum of the contributions due to nonmagnetic scattering and the residual part of the domain resistance, which at higher fields is almost a constant. The normalization constant R_0 is the total resistance at 1.2 K and for zero applied field. For $H>20$ kOe, the magnetization approaches technical saturation[7] and the specimen is driven into a single domain. Thus, at higher fields changes in the resistance due to domain realignment would be negligible, and R_e can be obtained by extrapolating the high-field part of the isotherm to the zero-field value. The normal magnetoresistance above 20 kOe is then given by $R(H)-R_e$, which we find can be empirically expressed as

$$[R(H)-R_e]/R_0 = \alpha B^{2.5}, \tag{5}$$

where $\alpha = a/R_0$ is a constant. The magnetization data of Jew and Legvold[7] have been used for calculating the B field in the crystal.

The field-dependent part of the domain resistance is given as the total resistance minus the normal magnetoresistance as given by the extrapolated part of the curve; this can be expressed empirically as

$$R_d(H)/R_0 = [R_d(0)/R_0]e^{-AH}, \tag{6}$$

where $R_d(0)/R_0 = 1 - R_e/R_0 = 4.2\times10^{-3}$, and a best fit to the data gives $A = 0.152$ kOe^{-1}. An attempt to describe this domain contribution using the B field was less satisfactory, which seems to suggest that domains respond to the external field rather than the total field. It should be noted that besides affecting various resistivity components, the field also induces a strain in the specimen which changes at low fields but is almost constant above 10 kOe (the magnetic strain $\Delta l/l \approx 4\times10^{-3}$ has been reported by Legvold *et al.*[20]). This is expected to give a few percent correction in the value of A which has been neglected in the analysis.

The transverse magnetoresistance isotherms for $\vec{I} \parallel \langle 10\bar{1}0\rangle$ and $\vec{H} \parallel \langle 11\bar{2}0\rangle$ are shown in Fig. 3. The magnetoresistance around 4 K is positive. This is due to the domain realignments in this case giving a positive contribution, since the spontane-

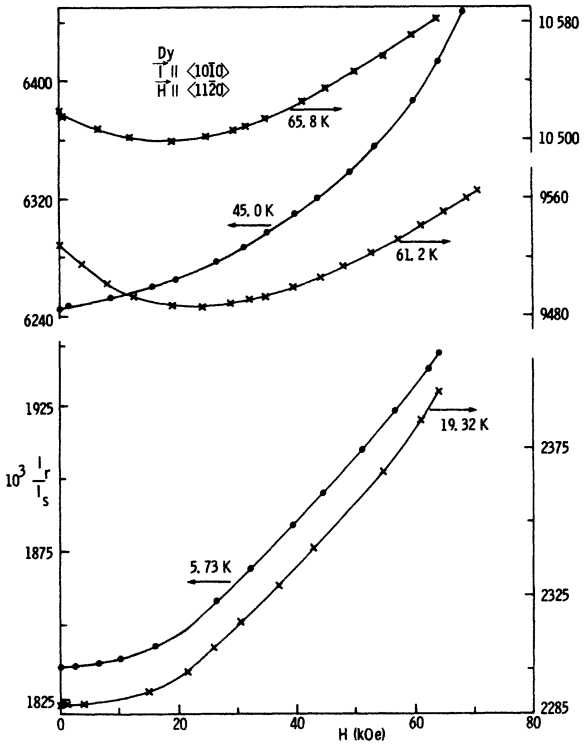

FIG. 3. Changes in transverse magnetoresistance ($\vec{I} \perp \vec{H}$) vs applied field for $\vec{I} \parallel \langle 10\bar{1}0 \rangle$ and $\vec{H} \parallel \langle 11\bar{2}0 \rangle$.

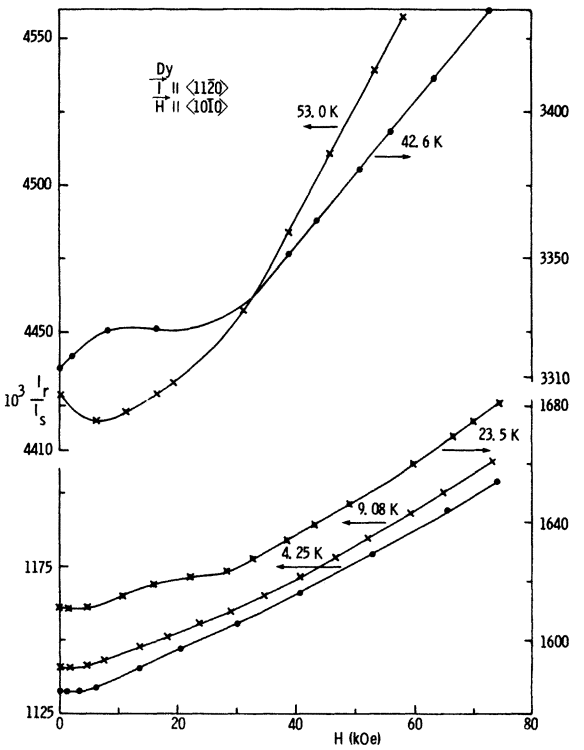

FIG. 4. Changes in transverse magnetoresistance vs applied field for $\vec{I} \parallel \langle 11\bar{2}0 \rangle$ and $\vec{H} \parallel \langle 10\bar{1}0 \rangle$. The maximum in the isotherms at 23.5 and 42.5 K has been suggested to be due to the minimum of the spin-wave energy gap.

ous magnetization is now aligned normal to the current. Further, the normal magnetoresistance for a transverse field is usually higher than for the longitudinal field. These contributions seem to dominate any decrease in the resistance due to removal of domain walls and reduction in the electron-magnon scattering. The positive magnetoresistance persists even at temperatures close to the Curie point, with an exception of a shallow minimum around 20 kOe. This is in contrast to the Tb basal-plane results, where the transverse magnetoresistance around 4 K is positive but gradually decreases as temperature increases; for $T > 25$ K and for the field above 30 kOe, the resistance isotherms exhibited a negative slope.[21] This is surprising in view of the remarkable similarities in the magnetic properties of the two metals.

Figure 4 shows typical transverse magnetoresistance isotherms for $\vec{I} \parallel \langle 11\bar{2}0 \rangle$ and $\vec{H} \parallel \langle 10\bar{1}0 \rangle$. The qualitative features of these isotherms are very similar to those shown in Fig. 3 except for the presence of a shallow maximum in some of the isotherms. This appears around 16 and 10 kOe in the 23.5- and 42.6-K isotherms, respectively. If a smooth curve is drawn through the data points on either side of the maximum, the height of the maximum is about 0.2% of the total resistance at zero field ($H = 0$) in both the isotherms. It should be noted that the scatter in the data points is much less than the estimated height of the maximum. In an attempt to further investigate the basal-plane anisotropy the following experiment was done.

A sample in the form of a rectangular parallelepiped was aligned such that its length was parallel to the c axis with the sides of its cross section parallel to the basal-plane axes. The error in the latter alignment was within 3°. The magnetoresistance was measured for two different orientations of the field by rotating the copper block through 90°. In both measurements, the direction of the current was parallel to the c axis and was thus always perpendicular to the applied field. The results are shown in Figs. 5 and 6; it is obvious that the maximum in the isotherms appears only when the field is parallel to the b axis. The source of this maximum is not certain at the present time, but may plausibly be attributed to the minimum energy gap as a function of the applied field. If the field-induced minimum value of the gap is lower than the gap in zero field,[5] then the energy gap decreases as the field increases and

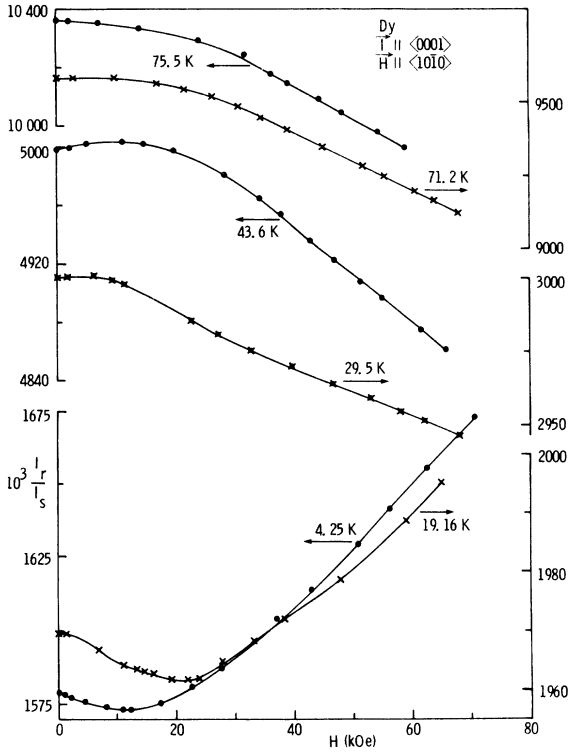

FIG. 5. Changes in transverse magnetoresistance vs applied field for $\vec{I} \parallel \langle 0001 \rangle$ and $\vec{H} \parallel \langle 10\bar{1}0 \rangle$. The decrease in the resistance at 4.25 K is mostly due to elimination of domain-boundary scattering.

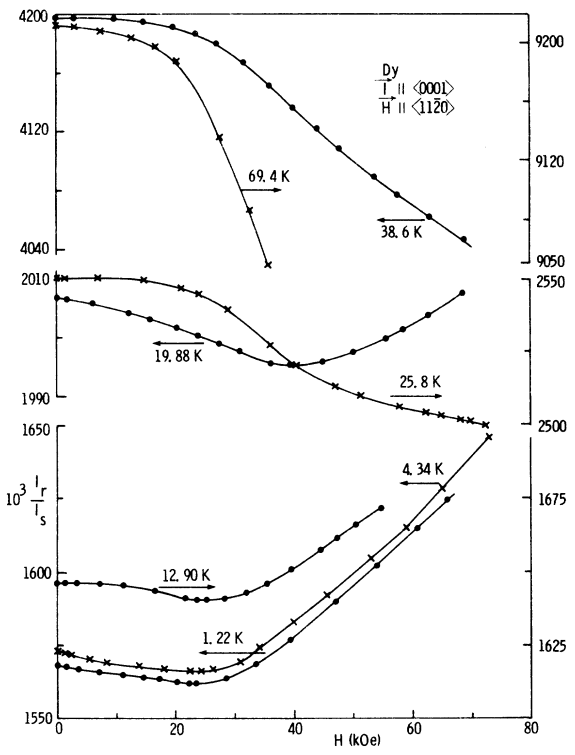

FIG. 6. Changes in transverse magnetoresistance vs applied field for $\vec{I} \parallel \langle 0001 \rangle$ and $\vec{H} \parallel \langle 11\bar{2}0 \rangle$.

consequently more magnons may be excited. This gives rise to an initial increase in the spin-wave resistivity instead of the usual decrease. On the other hand, for fields higher than the critical field, the gap monotonically increases and as a result the spin-wave resistivity decreases in a way similar to the a-axis results. These results would suggest that around 40 K, the critical field is about 10 kOe, which is much lower than the value of order 140 kOe calculated by Mackintosh and Møller.[5] Some other interesting features of these results are the following.

First of all, because of the magnetic anisotropy, the spontaneous magnetization within domains is confined to the basal plane even in the absence of an external field. Thus, the magnetization is normal to the current direction in the present case. When a field is applied in the basal plane, the changes in the resistance due to reorientation of domain magnetization would consequently be very small and the decrease in the resistance around 4 K is attributed mostly to the displacement of domain boundaries. This contribution is not apparent in the transverse magnetoresistance of the basal-plane crystals (Figs. 3 and 4) since contributions due to reorientation of the domain magnetization are more important in that case. At 4.25 K, the normal magnetoresistance for $\vec{H} \parallel \langle 10\bar{1}0 \rangle$ can be evaluated by extrapolating the results above 20 kOe to zero field. The increase in the resistance $[R(H) - R_e]$ at higher fields ($H > 20$ kOe) can be expressed empirically as $\Delta R \propto B^{2.2}$. The qualitative nature of the isotherms around 4 K for $\vec{H} \parallel \langle 11\bar{2}0 \rangle$ is very similar to $\vec{H} \parallel \langle 10\bar{1}0 \rangle$ except for the fact that the minimum appears at higher fields and the increase in resistance above the minimum is relatively weak as a function of the field. Second, for both directions of the field and for temperatures above 25 K, the resistance continuously decreases in the available field range. A comparison of these results with those shown in Figs. 3 and 4 suggests that the spin-wave scattering in the c-axis crystal is more sensitive to the field since the decrease in the resistance, for any given field, is greater.

During the course of this investigation, it was found that the resistance-versus-temperature curve shows a negative curvature below 4 K, whereas in most metals it is positive down to the millidegree range. In the virgin state of the specimens, the decrease in the resistance from 4 to 1.2 K is about 0.3% and is almost isotropic. Tindall and Jericho[22] reported a change of about 1.3%

between 4 K and 60 mK. But this anomalous contribution in the presence of the field is anisotropic and typical longitudinal isofield curves for the basal-plane crystals are shown in Fig. 7. It is seen from Fig. 7 that the field has little effect on the temperature-dependent component for the b-axis crystal although the total resistivity changes owing to variations in other components. The curve at 32 kOe is higher than at 8 kOe owing to the increase in the normal magnetoresistance. The qualitative behavior of the c-axis results is very similar to the b-axis crystal. By contrast, the temperature-dependent component for the a-axis crystal gradually decreases as the field increases and consequently the curves at higher fields show a weaker temperature dependence. The curve at 32 kOe shows a positive curvature; this implies that the above field is adequate to suppress the anomalous resistivity component. The source of this resistivity is not certain at the present time. However, the fact that it can be suppressed by a magnetic field in certain crystallographic directions does seem to suggest that it might be due to the magnetic ordering of some impurities. A relative maximum in the specific heat of this metal around 2–3 K has been reported by earlier workers and has been attributed to oxide impurities[23]; it may well be the source of the resistivity anomaly. Magnetoresistance measurements in the subhelium temperatures, with controlled amounts of different impurities, should throw more light on this phenomenon.

In summary, magnetoresistance results on several Dy single crystals have been reported. These have been discussed on the basis of the various scattering mechanisms and the magnetic anisotropy of the metal. Domain resistivity around 1 K, as estimated using the resistance minimum measured from resistance in virgin state, is about 0.2 and 0.08 $\mu\Omega$cm for the a- and b-axis crystals, respectively. In the longitudinal magnetoresistance the depth of the minimum increases at higher temperatures, which is attributed to the reduction of the inelastic electron-magnon scattering. The normal longitudinal magnetoresistance of the b-axis crystal shows a stronger field dependence than that of the a-axis. The transverse magnetoresistance of the basal-plane crystals ($\vec{I} \parallel \langle 10\bar{1}0 \rangle$, $\vec{H} \parallel \langle 11\bar{2}0 \rangle$ and vice versa) at temperatures close to the Curie point and for fields above 20 kOe, instead of the usual decrease as expected in the ferromagnets, increases with the field, which we cannot explain at present. For a field applied along the b axis, transverse magnetoresistance isotherms around 20 to 40 K show a small maximum which may be due to the field-induced minimum of the spin-wave energy gap. If so, the critical field for the minimum is much smaller than the theoretically predicted value.

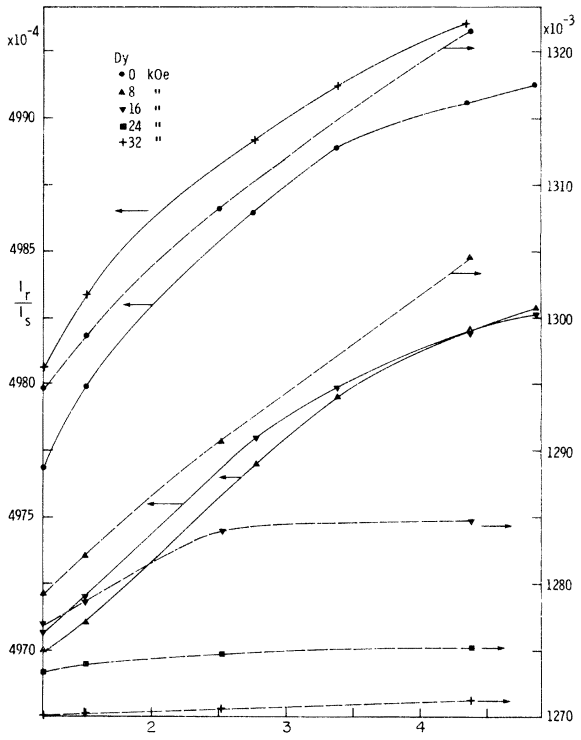

FIG. 7. Changes in longitudinal magnetoresistance isofield curves vs temperature for the $\langle 10\bar{1}0 \rangle$ and $\langle 11\bar{2}0 \rangle$ crystals. The curves pointing towards the left-hand side of the scale are for the b axis and those towards the right-hand side are for the a axis. Data have been obtained from the isothermal curves, a few of which are shown in Figs. 1 and 2. The resistivity component which gives rise to a negative curvature at $H=0$ is almost field independent for the b axis, but it gradually decreases as the field increases along the a axis.

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