the text. The solution is

$$1+(4N/3J\rho)=\tfrac{1}{2}\ln\left[\frac{(kT)^2+\Delta^2}{D^2+\Delta^2}\right]+(\Delta/kT)\tan^{-1}(kT/\Delta). \quad \text{(A3)}$$

For low temperatures, $kT\ll\Delta(T)$,

$$1+(4N/3J\rho)=\tfrac{1}{2}\ln\left\{\left(\frac{\Delta}{D}\right)^2\left[1+\left(\frac{kT}{\Delta}\right)^2\right]\right\}+1, \quad \text{(A4)}$$

$$(\Delta/D)[1+\tfrac{1}{2}(kT/\Delta)^2]=\exp(4N/3J\rho), \quad \text{(A5)}$$

$$\Delta(T)=[D\exp(4N/3J\rho)][1-\tfrac{1}{2}(kT/\Delta)^2]. \quad \text{(A6)}$$

For high temperatures, $kT\gg\Delta(T)$,

$$1+(4N/3J\rho)=\tfrac{1}{2}\ln\{(kT/D)^2[1+(\Delta/kT)^2]\}+(\Delta/kT)[\tfrac{1}{2}\pi-(\Delta/kT)] \quad \text{(A7)}$$

$$=\ln(kT/D)+\tfrac{1}{2}\pi(\Delta/kT), \quad \text{(A8)}$$

$$\Delta(T)=(2kT/\pi)[1+(4N/3J\rho)-\ln(kT/D)] \quad \text{(A9)}$$

$$=(2kT/\pi)\ln(T_K/T), \quad \text{(A10)}$$

where T_K is the critical temperature above which $\Delta(T)=0$.

$$T_K=2.7(D/k)\exp(4N/3J\rho). \quad \text{(A11)}$$

PHYSICAL REVIEW VOLUME 172, NUMBER 2 10 AUGUST 1968

Anomalous Hall Effect in Single-Crystal Dysprosium

J. J. RHYNE
U. S. Naval Ordnance Laboratory, White Oak, Silver Spring, Maryland
(Received 27 February 1968)

The "anomalous" or magnetization-dependent contribution to the Hall effect in single-crystal dysprosium has been investigated from 340 down to 4°K. In the paramagnetic region, with **H** applied along $\langle 11\bar{2}0\rangle$ directions, the Hall resistivity, defined by $\rho_H\equiv R_0B+R_s4\pi M_s$, is linear in H and exhibits a temperature dependence principally due to that of the magnetic susceptibility. The resulting anomalous coefficient $R_s=-18\times10^{-12}\,\Omega$ cm/G and normal coefficient $R_0=-0.3\times10^{-12}\,\Omega$ cm/G are both temperature-independent above 190°K. Below the Néel temperature (178°K), the anomalous coefficient changes sign, passes through a maximum near 120°K (where $R_s=+21\times10^{-12}\,\Omega$ cm/G), and decreases essentially to zero at 30°K. The temperature dependence of R_s below 105°K is well represented by the first power of the magnetic (spin disorder) resistivity, ρ_m. Such a dependence is in agreement with the f-spin–s-orbit interaction model due to Maranzana, considering only scattering from the thermally disordered spin system as calculated by Irkhin and Abelskii. The contributions to ρ_H predicted by other mechanisms are evaluated and discussed. The second independent component of ρ_H allowed in hcp symmetry and characterized by **H** applied along $\langle 0001\rangle$ was determined in the paramagnetic range. High-temperature values of R_s and R_0 are $-23\times10^{-12}\,\Omega$ cm/G and $-3.7\times10^{-12}\,\Omega$ cm/G, indicating appreciable anisotropy between c-axis and basal-plane components. Results could not be continued into the ordered temperature range, because of the enormous magnetic anisotropy.

I. INTRODUCTION

STUDIES of the Hall effect in ferromagnetic materials[1] have indicated the presence of a term dependent on the magnetization, the "anomalous Hall effect," as well as the ordinary term linear in the magnetic field. Initial attempts to explain the origin of the anomalous part by Karplus and Luttinger[2] were based on an intrinsic spin-orbit coupling of the magnetic electrons which are also charge carriers. In their model, this interaction led to an asymmetric scattering of the electrons and a resulting Hall-type transverse electric field. Such a model would not be expected to explain the anomalous effect in the rare earths due to the highly localized magnetic electrons which do not participate in conduction.

More recently, Kondo[3] has shown that in the second Born approximation an anomalous Hall effect can result from the s-d (or equivalent s-f) electron spin-spin interaction that in principle also describes the resistivity of the rare earths.[4] In this picture, the charge carriers are scattered by the thermal disorder of the magnetic spin system. To obtain the necessary transverse or "skew" scattering, Kondo invokes an intrinsic spin-orbit coupling of the f electrons which permits odd powers of the s-f interaction matrix elements to appear. This, together with an assumed anisotropy in the s-f interaction, leads to a transverse Hall current. As pointed out by Kondo, such a model depends explicitly on the angular momentum of the magnetic electrons remaining unquenched. Such is not the case in Gd, yet it has a very large anomalous Hall effect.

[1] A review of some of the early work in this field is found in E. M. Pugh and N. Rostoker, Rev. Mod. Phys. **25**, 151 (1953).
[2] R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954).
[3] J. Kondo, Progr. Theoret. Phys. (Kyoto) **27**, 772 (1962).
[4] The well-known Ruderman-Kittel-Kasuya-Yosida interaction applied to the resistivity by T. Kasuya, Progr. Theoret. Phys. (Kyoto) **22**, 227 (1959).

Maranzana[5] has overcome this difficulty in a recent paper which gives a very thorough treatment of the Hall effect based on the Kondo model. In Maranzana's work, the necessary asymmetry is provided by a spin-orbit interaction between the spin of the localized moment and the orbital momentum of the charge carriers. Additional aspects concerned with both types of spin-orbit coupling are found in papers by Irkhin *et al.*,[6,7] Kagan and Maksimov,[8] Kondorskii *et al.*,[9] and others.

The qualitative picture of the crossed *s-f* mechanism, as given by Maranzana,[5] is that of a localized moment (e.g., 4*f* electron) taken at the origin which sets up a vector potential $\mathbf{A}=(\mathbf{M}\times\mathbf{r})/r^3$. The energy of interaction between this moment and the current of primary charge carriers is (using $\nabla\cdot\mathbf{A}=0$)

$$H=(-e/mc)(\mathbf{A}\cdot\mathbf{p})=(-e/mcr^3)(\mathbf{M}\times\mathbf{r}\cdot\mathbf{p})$$
$$=(-e/mcr^3)(\mathbf{M}\cdot\mathbf{L}), \quad (1)$$

where $\mathbf{L}$ is the angular momentum of a charge carrier about the site of the localized moment, e is the charge, and m is the mass. Such a spin-orbit coupling (M is proportional to S) clearly provides the necessary asymmetry mechanism for "skew scattering," since the Hamiltonian is odd under reflection of the position of the charge carrier in the plane containing $\mathbf{M}$ and the current direction (i.e., $\mathbf{r}\rightarrow-\mathbf{r}$). As shown by Maranzana this Hamiltonian, when combined with an isotropic *s-f* exchange Hamiltonian and the appropriate scattering mechanism (spin disorder[3,5,6,8] or phonon[7]), leads in the second Born approximation to a difference between scattering from quasifree electron states k to k' than from k' to k. Odd powers of matrix elements of the Hamiltonian (1) appearing in the transport equation thus produce the required transverse Hall field. Such a "mixed" *f*-spin–*s*-orbit coupling would not be expected to be a strong effect compared to an intrinsic *f*-spin–*f*-orbit coupling. However, even though the interaction itself is much weaker, the high mobility of the *s* charge carriers allows a considerable net impact on the Hall effect. The rare earths (as opposed to the 3*d* ferromagnets) are ideally suited for examining the anomalous Hall effect of the Kondo-Maranzana type due to the complete dichotomy between the highly localized 4*f* magnetic electrons and the outer-shell charge carriers.

This paper reports the results of Hall coefficient studies on single-crystal dysprosium from 4 to 340°K. Dysprosium is a hcp metal ($c/a=1.57$) with a 0°K magnetic moment of 10.3 μ_B.[10,11] Neutron diffraction studies[12] have shown a region of spiral antiferromagnetic order from 178 (Néel temperature) to 87°K. A giant crystal-field anisotropy of order 10^9 erg/cc[13,14] at $T=0$°K constrains the moment to the basal plane. Applied fields above a temperature-dependent critical magnitude[10] collapse the spiral and produce ferromagnetic alignment. Fanning of the moment about the field direction occurs before final alignment is achieved for temperatures close to T_N. Below 87°K (Curie temperature) dysprosium spontaneously orders into a ferromagnet. The a axis $[11\bar{2}0]$ is the easy magnetic direction with an in-plane anisotropy energy of order 10^6 ergs/cc.

Hall measurements on polycrystalline dysprosium have been reported by Kevane *et al.*[15] in the paramagnetic region and by Volkenshtein *et al.*[16] Single-crystal data on Dy were also reported by Volkenshtein *et al.*[17]; however, magnetic saturation could not be achieved with their available fields and choice of crystal direction. This precluded separation of the anomalous and normal parts of the Hall effect, which we have accomplished in the present work. Single-crystal measurements on Gd, the only other rare earth studied in single-crystal form, have appeared in papers by Lee and Legvold[18] and Volkenshtein *et al.*[19] Single crystals are required to obtain meaningful results on the galvanomagnetic properties of the rare earths because of their large magnetic and Fermi-surface anisotropies.[20,21]

II. PHENOMENOLOGICAL HALL COEFFICIENTS

The general expression for the electric current in a ferromagnetic material can be written in the following form:

$$j_i=\sigma_{ij}E_j+(\sigma_H)_{ijk}B_jE_k+(\sigma_H')_{ijk}M_jE_k, \quad (2)$$

where the first term on the right is from the normal

[5] F. E. Maranzana, Phys. Rev. **160**, 421 (1967).
[6] Yu. P. Irkhin and Sh. Sh. Abelskii, Fiz. Tverd. Tela **6**, 1635 (1964) [English transl.: Soviet Phys.—Solid State **6**, 1283 (1964)].
[7] Yu. P. Irkhin and V. G. Shavrov, Zh. Eksperim. i Teor. Fiz. **42**, 1233 (1962) [English transl.: Soviet Phys.—JETP **15**, 854 (1962)].
[8] Yu. Kagan and L. A. Maksimov, Fiz. Tverd. Tela **7**, 530 (1965) [English transl.: Soviet Phys.—Solid State **7**, 422 (1965)].
[9] E. I. Kondorskii, A. V. Cheremushkina, and N. Kurbaniyazov, Fiz. Tverd. Tela **6**, 539 (1964) [English transl.: Soviet Phys.—Solid State **6**, 422 (1964)].
[10] D. R. Behrendt, S. Legvold, and F. H. Spedding, Phys. Rev. **109**, 1544 (1958).
[11] T. T. Jew and S. Legvold, U.S. Atomic Energy Commission Research and Development Report, Ames Laboratory, IS-867, 1963 (unpublished).
[12] M. K. Wilkinson, W. C. Koehler, E. O. Wollan, and J. W. Cable, J. Appl. Phys. **32**, 48S (1961).
[13] S. H. Liu, D. R. Behrendt, S. Legvold, and R. H. Good, Jr., Phys. Rev. **116**, 1464 (1959).
[14] J. J. Rhyne and A. E. Clark, J. Appl. Phys. **38**, 1379 (1967).
[15] C. J. Kevane, S. Legvold, and F. H. Spedding, Phys. Rev. **91**, 1372 (1953).
[16] N. V. Volkenshtein and G. V. Fedorov, Zh. Eksperim. i Teor. Fiz. **44**, 825 (1963) [English transl.: Soviet Phys.—JETP **17**, 560 (1963)].
[17] N. V. Volkenshtein, I. K. Grigorova, and G. V. Fedorov, Zh. Eksperim. i Teor. Fiz. **51**, 780 (1966) [English transl.: Soviet Phys.—JETP **24**, 519 (1967)].
[18] R. S. Lee and S. Legvold, Phys. Rev. **162**, 431 (1967).
[19] N. V. Volkenshtein, I. K. Grigorova, and G. V. Fedorov, Zh. Eksperim. i Teor. Fiz. **50**, 1505 (1966) [English transl.: Soviet Phys.—JETP **50**, 1003 (1966)].
[20] J. O. Dimmock and A. J. Freeman, Phys. Rev. Letters **13**, 750 (1964); A. J. Freeman, R. E. Watson, and J. O. Dimmock, Phys. Rev. **167**, 497 (1968).
[21] S. C. Keeton and T. L. Loucks, Phys. Rev. **168**, 672 (1968).

(diagonal) conductivity, the second term originates from the normal Hall effect, and the third term arises from the anomalous Hall effect peculiar to magnetic materials. This term was postulated empirically by Pugh.[1]

The conductivity tensors must remain invariant under the operations of the symmetry group of the crystal in question, and in addition the Onsager relations must be satisfied. For dysprosium which is of hcp ($6/mmm$) symmetry, this leaves only two independent components for the Hall conductivity, one characterized by the magnetic field (and magnetization) in the basal plane and one by the field along the c axis.

Imposing the requirement that the transverse current be zero leads to the following phenomenological expression for the Hall resistivity ρ_H (sometimes designated by e_H):

$$\rho_H = R_0 B + R_s 4\pi M. \tag{3}$$

R_0 is the normal Hall coefficient and R_s is the "anomalous" or "spontaneous" Hall coefficient. Corresponding to the Hall conductivity there are two components of ρ_H and two sets of coefficients R_0 and R_s for hexagonal symmetry. The Hall resistivity ρ_H is the transverse Hall electric field per unit longitudinal current density. It is related to the Hall conductivity appearing in Eq. (2) by the square of the ordinary resistivity [e.g.,

$$(\rho_H)_{yx} \equiv \frac{E_y}{j_x} = \frac{(\sigma_H)_{xyz} B_z + (\sigma_H')_{xyz} M_z}{-\sigma_{xx}\sigma_{yy}}$$

to first order in σ_H].

III. EXPERIMENTAL PROCEDURE

The rare-earth single crystals used in this study were prepared by arc-melting distilled metal, followed by a controlled high-temperature anneal in argon. Two platelike specimens were prepared by spark cutting and were designated "a" and "c" according to the direction of the magnetic field applied normal to the plate. For both samples the primary current was along the b ([10$\bar{1}$0]) direction. The dimensions of the specimens were: "a", 11.3×2.7×1.6 mm, and "c", 9.0×1.6× 0.98 mm. The resistivity ratio $\rho(300)/\rho(4.2)$ of sample "a" was 16.

The dc Hall voltages were determined by standard two-probe techniques. Potentials were measured with a Rubicon model 2772 potentiometer and Keithley 148 nanovoltmeter. The output of the nanovoltmeter was coupled to an x-y plotter through an isolation amplifier and active low-pass filter. Static values of Hall emf and magnetic field were plotted as points on the recorder with an over-all sensitivity better than 10^{-9} V.

Current leads were ultrasonically soldered to the samples and sharpened pressure contacts were used for the Hall emf probes. The specimens were mounted on a copper plate and sample holder to minimize thermal gradients and associated thermoelectric emf's. Hall-field results were found to be insensitive to reversal of the primary current direction.

The sample and holder were contained in a helium exchange gas environment. An automatic temperature control provided ±0.1° stability from 4 to 350°K. Temperature was measured with an AuCo thermocouple above 70°K and with a Ge resistance thermometer below 70°K. Absolute accuracy is ±1°K.

Spurious magnetoresistance contributions (of even parity in **H**) due to slight Hall-probe misalignment were eliminated by 180° magnetic field reversal achieved by rotating the sample. To eliminate the effects of the large anisotropy torques on the specimen and holder when the field was rotated out of the easy magnetic direction, the data were taken sequentially from zero to full field for the initial and then for the 180° reversed field direction. This method gave no difference in resulting Hall resistivities over the more conventional procedure of 180° reversal at each field value. The Hall resistivity at a field H was determined from the expression

$$\rho_H(H) = \tfrac{1}{2}[V_H(0°) - V_H(180°)](t/I), \tag{4}$$

where $V_H(0°)$ and $V_H(180°)$ were the voltages (referenced to $H=0$) appearing across the Hall probes for the two field directions. I and t were the sample current and thickness.

IV. HALL-RESISTIVITY RESULTS

A. Crystal "a" (H || ⟨11$\bar{2}$0⟩)

Hall-resistivity data as a function of applied field for the "a" crystal are shown in Fig. 1 in both paramagnetic and magnetically ordered temperature regions. Curves taken below T_C (87°K), the spontaneous ferromagnetic ordering temperature [Fig. 1(a)], show a constant slope up to the demagnetizing field (approximate demagnetizing factor $N \approx 0.6$ from experiment and also as calculated by the method of Joseph and Schlömann).[22] The Hall emf shows a sharp change in slope at magnetic saturation, which occurs a few kOe below our maximum attainable field. The normal Hall effect becomes evident only above moment saturation. In the temperature region below T_C, the Hall emf is principally positive and increasing with temperature. Negligible hysteresis or residual emf's were found in the Hall data in any temperature region in contrast to other results[17] reported on Dy. This is possibly a result of less strained crystals.

Figure 1(b) demonstrates the behavior for temperatures in the spiral region. In the lower-temperature part, essentially zero Hall voltage is found up to the

[22] R. I. Joseph and E. Schlömann, J. Appl. Phys. **36**, 1579 (1965).

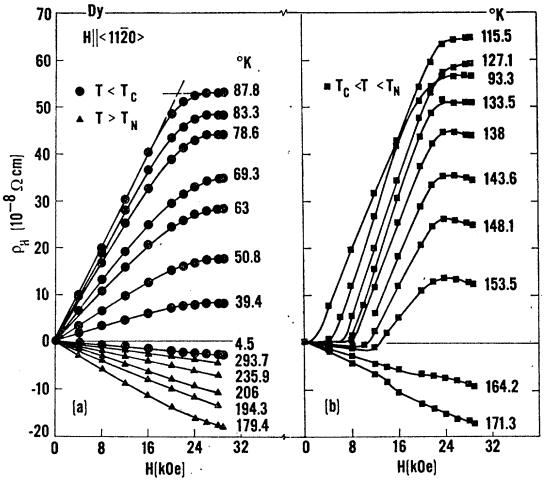

FIG. 1. Hall resistivity versus applied magnetic field for the "a" crystal ($\mathbf{H} \parallel \langle 11\bar{2}0 \rangle$). Data are shown in both paramagnetic and magnetically ordered temperature regions. The intersection of the dashed lines on the 87.8°K curve indicates the magnitude of the anomalous contribution to the Hall resistivity at this temperature.

critical field H_c, with a behavior characteristic of the ferromagnet above H_c. The saturation Hall emf is positive with a maximum near 120°K. A fairly sharp decrease occurs above this temperature with the sign changing to negative near 160°K. Effects of fanning of the magnetic moment and lack of complete saturation are evident near T_N (178°K).

Hall emf curves above T_N [Fig. 1(a)] are linear in field with a fairly strong temperature dependence (see Sec. V). Some curvature at the highest fields is noted close to T_N because of short-range ordering. The paramagnetic data for the field dependence of the Hall resistivity are in reasonable agreement with those of Volkenshtein *et al.*[17] Below T_N these authors were not able to achieve saturation of the Hall voltage in the available fields. Their basal-plane measurements were made on a crystal with $\mathbf{H}$ applied along the $\langle 10\bar{1}0 \rangle$ direction (intermediate hard axis), which precluded attaining saturation below T_C in normal laboratory fields. Thus, no meaningful comparison of the two sets of results can be made in the magnetically ordered temperature range except to note the agreement of sign and order of magnitude.

1. *Hall Coefficients above the Néel Temperature*

For temperatures above T_N, the spontaneous moment contribution to the Hall effect vanishes. This does not imply the absence of a significant anomalous part in the Hall emf. The effect of the paramagnetic moment induced by the applied field is strongly apparent in dysprosium because of its relatively large susceptibility as measured by Behrendt *et al.*[10]

In the paramagnetic region $M=\chi H$, and Eq. (3) becomes

$$\rho_H/H_{\text{appl}}=R_0+[R_s+R_0(1-N)]4\pi\chi/(1+4\pi N\chi). \quad (5)$$

The total effect is linear in H, but is divided between the normal part and a part depending on the magnetic susceptibility χ. Plotting the slope of the Hall-resistivity data in Fig. 1 (i.e., ρ_H/H_{appl}) versus the effective susceptibility χ^* defined by

$$\chi^*\equiv\chi/(1+4\pi N\chi), \quad (6)$$

one finds the results shown in Fig. 2. The most striking feature of this is the linear behavior from almost the Néel temperature to above 340°K. Such a relationship implies temperature independence of the Hall coefficients. The intercept on the ordinate axis ($\chi=0$, corresponding to infinite temperature) is the normal coefficient $R_0=-0.3\times10^{-12}$ Ω cm/G. The anomalous coefficient $R_s=-18\times10^{-12}$ Ω cm/G is found from the slope of the line.[23] The temperature dependence of the observed Hall resistivity results entirely from the magnetic susceptibility in agreement with the *s-f* interaction model as calculated by Kondo[3] (see Sec. V).

2. *Anomalous Coefficient below the Néel Temperature*

Below T_N the expression (3) for the Hall resistivity measured at magnetic saturation is

$$\rho_H=R_0B+R_s4\pi M=R_0[H_{\text{appl}}+(1-N)4\pi M_s]+R_s4\pi M_s. \quad (7)$$

The magnitude of the Hall resistivity extrapolated back to an applied field corresponding to the demagnetizing field (i.e., $H_{\text{appl}}=4\pi NM_s$) as shown by the

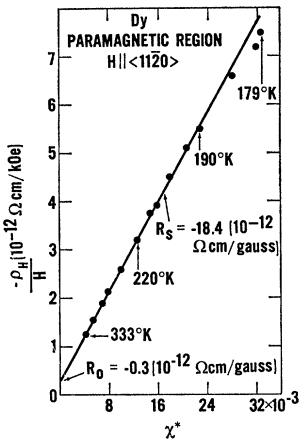

FIG. 2. Slope of the Hall resistivity versus applied field curves in the paramagnetic region plotted against the effective magnetic susceptibility (see text). The susceptibility accounts for the total temperature dependence of the Hall effect. The normal and anomalous coefficients have the constant values indicated.

[23] In the paramagnetic region with H along the b $\langle 10\bar{1}0 \rangle$ direction, Volkenshtein *et al.* (Ref. 17) gave values of $R_0=-1\times10^{-12}$ Ω cm/G and $R_s=-32.6\times10^{-12}$ Ω cm/G, significantly at variance with the present results.

dashed lines on the 87.8°K curve in Fig. 1(a) is

$$(\rho_H)_{H_{\text{demag}}}=(R_0+R_s)4\pi M_s\approx R_s 4\pi M_s \qquad (8)$$

for $R_0\ll R_s$. In cases where the demagnetizing field is a large fraction of the maximum field as in the present investigation, this method of determining R_s from the intercept at the demagnetizing field is considerably more accurate than the conventional extrapolation back to $H_{\text{appl}}=0$. This procedure also makes clear what value of M is to be used in Eq. (8), namely, the spontaneous moment M_s corresponding to $H_{\text{int}}=0$ (or $H_{\text{int}}=H_{\text{crit}}$ for $T>T_C$). In interpreting the dysprosium results, the moment data of Behrendt *et al.*[10] and Jew *et al.*[11] were used.

The slope of the Hall-resistivity curve, $\partial\rho_H/\partial H_{\text{appl}}$, above the "knee" is

$$\partial\rho_H/\partial H_{\text{appl}}=R_0+4\pi[R_s+R_0(1-N)](\partial M_s/\partial H_{\text{appl}})$$
$$\approx R_0+4\pi R_s(\partial M_s/\partial H_{\text{appl}}) \qquad (9)$$

for $R_0(1-N)\ll R_s$. The factor $\partial M_s/\partial H_{\text{appl}}$ is the high-field susceptibility, which when multiplied by the large value of R_s, can be an appreciable fraction of R_0, especially near T_N. Equations (8) and (9) would allow for the determination of both normal and anomalous coefficients, except that in these experiments on Dy the enormous demagnetizing field ($\approx$23 kOe at 4°K) made an accurate determination of the high-field slope of the Hall resistivity impossible in the 29 kOe maximum available field. Thus, meaningful results on the normal coefficient could not be determined.

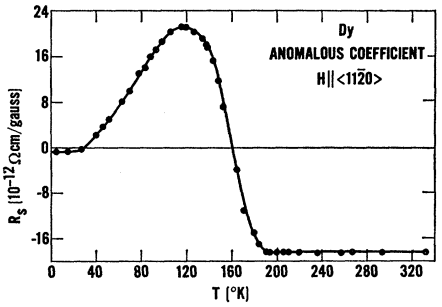

FIG. 3. The anomalous Hall coefficient for the "a" crystal as a function of temperature. Below T_N (178.5°K) the values shown were derived under the assumption that $R_0\ll R_s$ as given in the text. Note the reversal in sign just below the ordering temperature and the broad maximum near 120°K.

The anomalous coefficient obtained from the data of Fig. 1 is shown in Fig. 3. Above 190°K, R_s is constant as shown in Fig. 2. (Just above T_N, R_s was found by drawing tangents to the data plotted versus χ^* as in Fig. 2.) For temperatures significantly below T_N, the constant was evaluated from Eq. (8). The neglect of R_0 compared to R_s is not considered to appreciably affect the results except at low temperatures, where R_s approaches 0. The magnitude of R_0 would not be expected to differ greatly over the entire temperature range from the value -0.3×10^{-12} Ω cm/G found above T_N. The small negative value plotted for the anomalous constant at the lowest temperatures presumably reflects the neglected contribution of the normal coefficient dominant at this temperature.

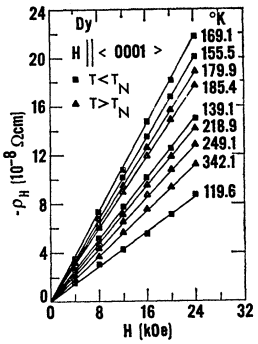

FIG. 4. Isotherms for the Hall resistivity versus applied field for the "c" crystal (H || ⟨0001⟩). Below 178.5°K the moment structure is a spiral in the basal plane with a small perpendicular susceptibility in the field direction resulting in a linear ρ_H versus H relationship.

At temperatures between about 130° and T_N significant fanning of the moment can occur above the spiral critical field. Due to this possible lack of saturation, values of R_s in this temperature range must be considered to be only semiquantitative.

B. Crystal "c" (H || ⟨0001⟩)

Figure 4 presents Hall data on the "c" crystal in the paramagnetic and spiral regions. Data are linear with field above and also below T_N since no intrinsic order is developed in the c-axis direction. For fields applied in this direction below T_N the moment structure remains spiral effectively in the basal plane with a small perpendicular susceptibility.[10]

The temperature dependence of the slope of the Hall-resistivity curves is shown in Fig. 5. Included are the paramagnetic data from the "a" crystal to illustrate the significant anisotropy. The "c" crystal shows a typical behavior for an antiferromagnet except that the peak in the Hall data occurs considerably below the Néel temperature in contrast to the susceptibility peak which occurs at T_N. The Hall-resistivity slope for the

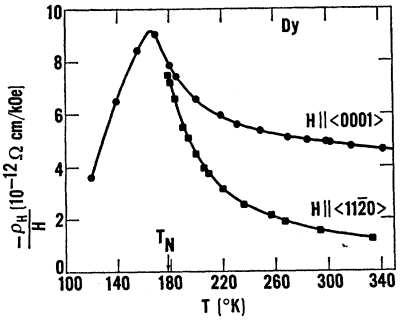

FIG. 5. The temperature dependence of the slope of the Hall resistivity-versus-H curves for the "a" crystal [Fig. 1(a)] and "c" crystal (Fig. 4) in the temperature range where ρ_H is linear in H.

"c" crystal is linear in the effective susceptibility χ^* [defined in Eq. (6)] only above 225°K. Below this temperature departures occur which, along with the peak below T_N, may be ascribed to a temperature dependence of the Hall coefficients. From the data linear in χ^* above 225°K, a value of $R_s=-23\times10^{-12}$ Ω cm/G and of $R_0=-3.7\times10^{-12}$ Ω cm/G were found. These are compared to values of -25×10^{-12} Ω cm/G and -5.7×10^{-12} Ω cm/G for R_s and R_0, respectively, obtained by Volkenshtein *et al.*[17] The "c" crystal constants are appreciable larger in magnitude than those for the "a" crystal.

Hall data with $\mathbf{H}\parallel\langle0001\rangle$ were not extended below 120°K as field components in the basal plane, produced by minute sample misorientation, begin to distort the spiral structure introducing extraneous effects.

V. DISCUSSION

Maranzana[5] gives the following result for the anomalous Hall resistivity based on the localized-spin–conduction-electron-orbit interaction model utilizing spin disorder scattering:

$$\rho_H'=\frac{3N}{ecV}\frac{\sin k_Fa}{E_Fa}\frac{m}{\hbar^2}\mathcal{J}^2g\mu_B\mathfrak{M}_3. \tag{10}$$

The quantity[24] $\mathfrak{M}_3$ comes from a three-spin correlation function and has the form of the following spin moment fluctuation expression:

$$\mathfrak{M}_3=N\langle(m-\langle m\rangle)^3\rangle=N(\langle m^3\rangle-3\langle m\rangle\langle m^2\rangle+2\langle m\rangle^3). \tag{11}$$

E_F and k_F are the Fermi energy and wave vector; a is a lattice constant; and $\mathcal{J}$, g, and μ_B are, respectively, an exchange constant, Landé splitting factor, and the Bohr magneton. $\mathfrak{M}_3$ contains the primary temperature dependence of the Hall effect. As T approaches 0°K the quantity $\langle(M-\langle M\rangle)^3\rangle$ vanishes and the anomalous part of the Hall effect disappears as observed. In the paramagnetic region Kondo[3] gives the following result for $\mathfrak{M}_3$:

$$(\mathfrak{M}_3)_{\text{para}}=-N(2J^2+2J+1)(\chi H/5g\mu_B), \tag{12}$$

where J is the angular momentum. The Hall resistivity above T_N is rigorously proportional to the magnetic susceptibility χ as was assumed on phenomenological grounds from Eq. (3). Expression (12) thus predicts that the anomalous coefficient will be temperature-independent above T_N as confirmed by the data in Fig. 2.

Irkhin *et al.* have calculated the anomalous Hall effect considering both the intrinsic spin-orbit coupling of the magnetic electrons and the s-d "mixed" coupling. For phonon scattering[7] they obtain the result that the anomalous Hall coefficient should depend on the square of the phonon resistivity, $R_s\propto\rho_{\text{ph}}^2$. In the case of spin disorder scattering,[6] Irkhin and Abelskii obtain for both spin-orbit mechanisms a result for R_s proportional to the magnetic spin disorder resistivity ρ_m:

$$R_s=\pm\tfrac{3}{16}(\lambda^{\text{eff}}/E_F)[\rho_m/M_s(0)], \tag{13}$$

where λ^{eff} is the effective spin-orbit coupling constant and $M_s(0)$ is the zero-deg magnetization. In obtaining this result they have used the spin-resistivity calculations of Kasuya[4] and have factored the magnetization out of the three-spin corrolation functions by a molecular-field approximation. The principal temperature dependence of the Hall coefficient lies in the dominant magnetic contribution to the total resistivity.

Although the quantity ρ_m is not directly observable, a reasonable separation of it from the total resistivity may be effected by the following procedure. The total resistivity is assumed to be made up of residual (impurity), phonon, and magnetic contributions: $\rho_t=\rho_0+\rho_{\text{ph}}+\rho_m$. The residual resistivity ρ_0 is a constant for a given crystal and makes a temperature-independent contribution to R_s.[25] In the paramagnetic region the slope of the resistivity versus T curve is dominated by the phonon contribution (the contribution from the spin system is small as given by Kasuya[26]). By the use of the phonon part obtained in the high-temperature region and the Grüneisen function to extrapolate back into the magnetically ordered region, a subtraction of the approximate phonon resistivity can be made. The Debye temperature used in the calculation was 170°K as obtained from sound velocity measurements.[27] This method ignores the interband scattering part[28] of the resistivity, but the temperature dependence of ρ_m is

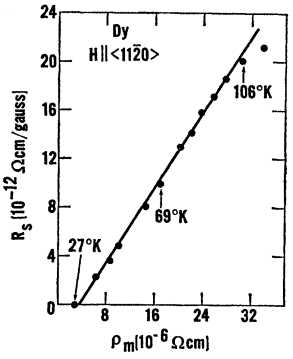

FIG. 6. The anomalous Hall coefficient for the "a" crystal plotted as a function of the derived spin disorder resistivity. Data shown are from the low-temperature side of the peak in the Hall coefficient (see Fig. 3).

[24] Kondo (Ref. 3) obtains a somewhat different expression for ρ_H' but which still contains the factor $\mathfrak{M}_3$.

[25] J. M. Luttinger, Phys. Rev. **112**, 195 (1958).

[26] T. Kasuya, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1966), Vol. IIB, p. 254.

[27] F. H. Spedding, S. Legvold, A. H. Daane, and L. D. Jennings, Progr. Low Temp. Phys. **2**, 368 (1957).

[28] D. A. Goodings, Phys. Rev. **132**, 542 (1963).

nevertheless expected to be qualitatively correct in the region covered. The Dy resistivity data of Spanel and Mackintosh[29] were analyzed for the magnetic contribution, which amounted to 73% of the total resistivity at 100°K. Their data taken at magnetic saturation (to eliminate the effects of magnetic superzones in the spiral region) show that the resistivity is isotropic in the ordered state and nearly linear with temperature above 40°K. In the paramagnetic region the former is not the case, and the polycrystalline average $(\frac{1}{3}\rho_c+\frac{2}{3}\rho_a)$ resistivity was used in extracting the phonon portion.

The experimentally determined basal-plane anomalous Hall coefficient is plotted against the derived magnetic spin disorder resistivity in Fig. 6. The relation (13) is well obeyed from low temperatures where the anomalous coefficient essentially vanishes up to within 15° of the peak in the Hall term (see Fig. 3).

From the above results it is evident that the ρ_i^2 relationship found by Karplus and Luttinger[2] from the band model and also the ρ_{ph}^2 relationship derived for *s-d* interaction with phonon scattering[7] do not represent the mechanism responsible for the anomalous Hall effect in Dy. The dominant source is the spin disorder scattering.

Direct evaluation of the three-spin correlation functions which appear in the transition probabilities for the Hall effect without the use of the molecular-field approximation is highly desirable, especially at low temperatures. This has been attempted in the spin-wave approximation by Kagan and Maksimov,[8] who calculated a T^4 dependence for R_s at low temperatures with a transition to a T^2 relationship at higher temperatures. The transition to T^2 behavior is realized at lower temperatures in metals with complex Fermi surfaces such as in the rare earths. The anomalous Hall coefficient for Dy is plotted versus T^2 in Fig. 7(a). Reasonable agreement is evident up to about 80°K. The omission of the sizeable anisotropy energy gap[30,31] from the magnon spectrum in Kagan and Maksinov's calculations may have an appreciable effect on the temperature dependence.

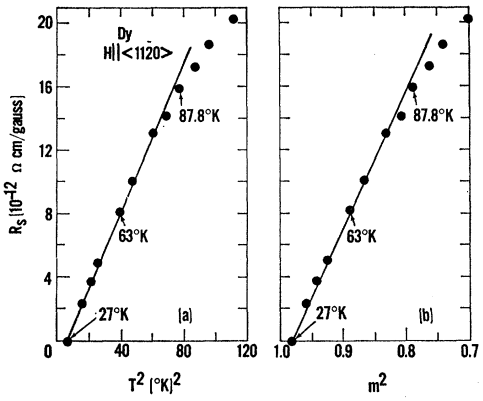

FIG. 7. (a) The dependence of the low-temperature anomalous Hall coefficient on T^2 as predicted for a spin-wave approximation (see text), and (b) on the square of the reduced magnetization $M(T)/M(0)$ as found by a molecular-field approximation.

A dependence of R_s on the square of the magnetization is calculated by Kagan and Maksimov and others using the molecular-field approximation, which should be most valid at high temperatures near T_N. It is noted, as shown in Fig. 7(b), that the m^2 dependence is obeyed in Dy for almost the same low-temperature range as the T^2 dependence of Fig. 7(a) found from spin-wave theory.

The appearance of the peak in the Hall coefficient near 120°K ($0.7T_N$) is at present not adequately explained. The quantity $\mathfrak{M}_3$ obtained by Kondo and Maranzana, when evaluated in molecular-field theory, is equivalent to the curvature of the Brillouin function. $\mathfrak{M}_3$ is small up to about $0.2T_C$, passes through a maximum at about $0.85T_C$, and becomes zero at T_C. The occurrence of the maximum significantly below this point for dysprosium and even more important the subsequent reversal in sign below T_N is puzzling. The corresponding anomalous coefficient in the *S*-state ion Gd[18,19] is negative both in the paramagnetic and ordered magnetic regions, which suggests the presence of a competing mechanism in Dy that is driving the Hall coefficient positive in the ordered state. Barring the occurrence of major Fermi-surface distortion, this leads one to associate the sign reversal in Dy with the contribution of the intrinsic spin-orbit coupling of the 4*f* electrons[3] in addition to and opposing the effect of the mixed *f*-spin–*s*-orbit mechanism[5] that is operable in both Gd and Dy. The possibility that the reversal in sign in Dy is produced by this or some other interaction dependent on the nonzero orbital momentum of the ground state is to be explored.

ACKNOWLEDGMENTS

These experiments were made possible by the kind cooperation of the Solid State Physics Group[32] at the University of Maryland in the use of their high-field electromagnet. The assistance of J. B. Boyce in the construction of the experimental apparatus and in the initial stages of the experiment is gratefully acknowledged, as is the work of R. E. Jones in preparing the arc-melted buttons. It is also a pleasure to acknowledge many valuable discussions and contributions from Dr. Earl Callen and Dr. James R. Cullen.

[29] A. R. Mackintosh and L. E. Spanel, Solid State Commun. **2**, 383 (1964); L. E. Spanel, Ph.D. thesis, Iowa State University, Ames, Iowa, 1964 (unpublished).

[30] K. Niira, Phys. Rev. **117**, 129 (1960).

[31] A. R. Mackintosh, Phys. Letters **4**, 140 (1963).

[32] Materials Sciences Center supported by the Advanced Research Projects Agency, Grant No. SD-101.