Galvanomagnetic Effects in Cadmium Sulfide*†

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(Received 1 October 1962)

The Hall and magnetoresistance effects were measured at 297, 196, and 77°K in single crystals of n-type CdS having a room-temperature resistivity of 4 Ω-cm. The results of the measurements were consistent with a simple single-valley model of the energy surfaces in the conduction band and can be summarized in terms of two Hall mobilities μ_{H1} and μ_{H3} and a magnetoresistance parameter $\xi=\Delta\rho/(\rho_0\mu_{H1}\mu_{Hi}B^2)$, where $\Delta\rho/\rho_0$ is the relative change in resistivity in a transverse magnetic field B, and i may be 1 or 3, depending on the directions of current and magnetic field with respect to the c axis. The measured Hall mobilities μ_{H1} were 390, 840, and 5600 cm²/V-sec, and the values of ξ were 0.077, 0.074, and 0.045 at the three respective temperatures. A two-terminal method of measuring the ratio μ_{H1}/μ_{H3} was devised, which yielded the values 1.014, 1.05, and 1.32 at the three temperatures. The magnitude of the mobility at the two upper temperatures is explained quite well on the basis of combined optical mode and piezoelectric scattering by assuming an effective mass $m^*=0.19m_e$, while at the lowest temperature impurity scattering also contributes. The temperature-dependent anisotropy is larger than predicted for piezoelectric scattering, possibly due to some form of anisotropic defect scattering.

INTRODUCTION

GALVANOMAGNETIC effects such as the Hall and magnetoresistance effects have been found to be useful in studying both the symmetry of the energy band structure and the mechanisms by which electrons and holes are scattered in crystalline solids. Both of these objectives are of interest in the case of CdS because of the possibility of verifying the band structure determined by optical studies[1,2] and of identifying the lattice scattering mechanism(s).[3,4] The purpose of the present study was to measure all of the independent components of the resistivity, Hall, and magnetoresistance tensors in single crystals of n-type CdS of sufficient purity and at high enough temperature that lattice scattering would predominate, and to interpret the measurements in the light of current models of band structure and lattice scattering in CdS.

PHENOMENOLOGY AND DESIGN OF THE EXPERIMENTS

In order to determine an efficient method of measuring galvanomagnetic effects, it is useful to consider them from a purely phenomenological viewpoint, invoking only Onsager's relations (which hold for all isothermal galvanomagnetic effects) and the requirement of the symmetry imposed by the crystal lattice. A complete phenomenological theory of isothermal galvanomagnetic effects has been given by Kao and Katz.[5] They expand the conductivity in powers of the magnetic field B in the following way (the notation differs somewhat from that of Kao and Katz):

$$\sigma_{ij}(B)=\sigma_{ij}+\sigma_{ijk}B_k+\sigma_{ijkl}B_kB_l+\cdots, \quad (1)$$

where the subscripts indicate components along a particular set of Cartesian coordinates in the crystal, and repeated indices are summed from 1 to 3. An expansion in powers of B is useful because it is found experimentally that such an expansion converges quite rapidly except, of course, in the presence of oscillatory behavior due to quantum effects. Kao and Katz have tabulated (for all the crystal classes) the number of independent coefficients, the coefficients which are required to be zero by crystal symmetry, and the relationships between nonzero coefficients. Equation (1) can then be inverted and the coefficients in it can be related to the coefficients in a similar expansion of the resistivity tensor:

$$\rho_{ij}(B)=\rho_{ij}+\rho_{ijk}B_k+\rho_{ijkl}B_kB_l+\cdots. \quad (2)$$

The coefficients in Eq. (2) are the coefficients measured in an actual experiment, i.e., the resistivity (zero-order terms); the Hall coefficients (negatives of the first-order terms); and the magnetoresistivity coefficients (the second-order terms).

By applying the results of Kao and Katz to the case of CdS (crystallographic point group[6] C_{6V}), one obtains the following results. There are two resistivity coefficients (ρ_{11} and ρ_{33}), two independent Hall coefficients ($-\rho_{123}$ and $-\rho_{231}$), and six independent magnetoresistivity coefficients (ρ_{1111}, ρ_{3333}, ρ_{1122}, ρ_{1133}, ρ_{3311}, and ρ_{2323}). In addition, the following coefficients may be nonzero:

$$\begin{aligned}&\rho_{22}=\rho_{11}, \quad -\rho_{312}=-\rho_{231}, \quad \rho_{2222}=\rho_{1111},\\ &\rho_{2211}=\rho_{1122}, \quad \rho_{2233}=\rho_{1133}, \quad \rho_{3322}=\rho_{3311},\\ &\rho_{1313}=\rho_{2323}=\rho_{3131}=\rho_{3232}, \quad \rho_{1212}=\rho_{2121}=\rho_{1111}-\rho_{1122}.\end{aligned} \quad (3)$$

* Based on a thesis submitted to the University of Wisconsin Physics Department by one of us (J. D. Z.) in partial fulfillment of the requirements for the Ph.D. degree.

† Supported by the Air Research and Development Command, U. S. Air Force.

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[1] J. J. Hopfield and D. G. Thomas, Phys. Rev. **122**, 35 (1961).
[2] W. W. Piper and D. F. T. Marple, J. Appl. Phys. **32**, 2292 (1961).
[3] H. Frohlich, in *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd, London, 1954), Vol. 3, p. 325.
[4] A. R. Hutson, J. Appl. Phys. **32**, 2286 (1961).
[5] L. P. Kao and E. Katz, J. Phys. Chem. Solids **6**, 223 (1958).
[6] M. L. Glasser, J. Phys. Chem. Solids **10**, 229 (1959).

One advantage of knowing the phenomenological theory is that one can check for spurious effects which may violate the theory, e.g., due to size, geometry, voltage probes, inhomogeneities, or nonisothermal conditions. Another advantage is that the experiment can be designed in a way such that all the possible galvanomagnetic constants can be measured with a minimum number of samples. In the present case, only two types of samples are needed, one with current parallel to the c axis, and one with current perpendicular to the c axis as illustrated in Fig. 1. The theory shows that measurements on either type of sample are independent of the orientation of the sample with respect to either of the other crystal axes.

The resistivity and magnetoresistance measurements were taken by the four-terminal method and the Hall and planar Hall coefficients (such as ρ_{2323}) were taken by a five-terminal method as illustrated in Fig. 1(a). By choosing the proper direction of the magnetic field with respect to the current and voltage probes, each galvanomagnetic coefficient can be determined independently. Furthermore, some of the coefficients can be measured more than once, and this redundancy provides a check that the measurements do indeed obey the phenomenological theory. For example, the Hall coefficient $-\rho_{123}$ would be measured on voltage probes 1, 2, and 3, and $-\rho_{231}$ would be measured on probes 4, 5, and 6, but either probes 1 and 2 or probes 4 and 5 could be used to measure ρ_{11}, ρ_{1111}, ρ_{1122}, and ρ_{1133}, and measuring them on both sets of probes would give a check for spurious effects. Also, the Hall coefficient $-\rho_{231}$ can be measured on both types of samples. However, since there may be some variation from sample to sample in the carrier density n, one might expect discrepancies when comparing data from sample to sample. Since the present samples were extrinsic, only one band was involved with the conduction processes, each of the galvanomagnetic coefficients contains n simply as a factor, and the ratios of coefficients (such as the Hall mobility $-\rho_{231}/\rho_{33}$) should be most useful in comparing data taken in different samples.

EXPERIMENTAL PROCEDURE

All the CdS crystals on which measurements were taken were obtained from the Aeronautical Research Laboratory at the Wright-Patterson Air Force Base, Dayton, Ohio, and were grown from the vapor phase[7] under the direction of D. C. Reynolds. The samples were not intentionally doped but contained small amounts of uncompensated donors giving resistivities in the range from 3 to 50 Ω-cm at room temperature. The samples came in form of cleaved chunks and were cut into rectangular bars with a jeweler's saw, then lapped to the final dimensions which were typically 2×2×8 mm. The direction of the c axis in a crystal could easily be determined by looking at a sample while it was between a pair of crossed polaroids. Whenever possible, the cleaved chunks were cut into layers containing the c axis, and alternate layers were then cut into bars having their lengths parallel and perpendicular to the c axis. By using a two-terminal method, the resistances of the bars were approximately measured at room temperature and in liquid nitrogen to see if systematic variations existed from sample to sample due to inhomogeneities. Samples were then selected for low inhomogeneities and high ratios of room temperature resistance to liquid nitrogen resistance (which implied high mobilities at the lower temperature since preliminary measurements showed that in most of the crystals the Hall coefficients do not change appreciably between the two temperatures).

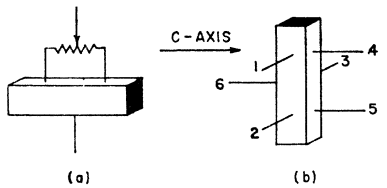

FIG. 1. The two types of samples and the voltage probes needed to give a complete set of data. (a) Current is parallel to the c axis and the sides are at an arbitrary orientation with respect to the other axes. (b) Current is perpendicular to the c axis but otherwise arbitrary and two sides are perpendicular to the c axis. Probes 1, 2, and 3 measure transverse electric fields perpendicular to the c axis and probes 4, 5, and 6 measure transverse fields parallel to the c axis by the five-terminal method illustrated in (a).

The samples were etched with hot 9% HCl and electrical contacts were made using indium solder agitated ultrasonically with a Sonobond[8] soldering apparatus. The current contacts covered the ends of the sample and usually six voltage contacts were used as shown in Fig. 1. The voltage contacts were between 0.2 and 0.3 mm in diameter, and had resistances which were usually about twice as large as the sample resistance. The voltage contacts still did not approximate point contacts because effects due to shorting of the Hall voltages by the contacts were clearly evident in the magnetoresistance data as will be discussed below. A lower limit to the size and resistance of the contacts was set by the requirement of low noise. The sample was etched again after all the leads had been attached and tested, so as to remove any surface layers that might have been formed when the sample was heated for soldering, and that might contribute a spurious surface conductivity[9] to the sample.

The sample holder and Dewar system were designed so that the sample could be in contact with a stirred temperature bath at room temperature (297°K), dry ice temperature (196°K), and at liquid nitrogen temperature (77°K). The sample holder was designed so

[7] L. C. Green, D. C. Reynolds, S. J. Czyzak, and W. M. Baker, J. Chem. Phys. **29**, 1375 (1958).

[8] Manufactured by Aeroprojects, Inc., West Chester, Pennsylvania.

[9] C. Herring, T. H. Geballe, and J. E. Kunzler, Bell System Tech. J. **38**, 657 (1959).

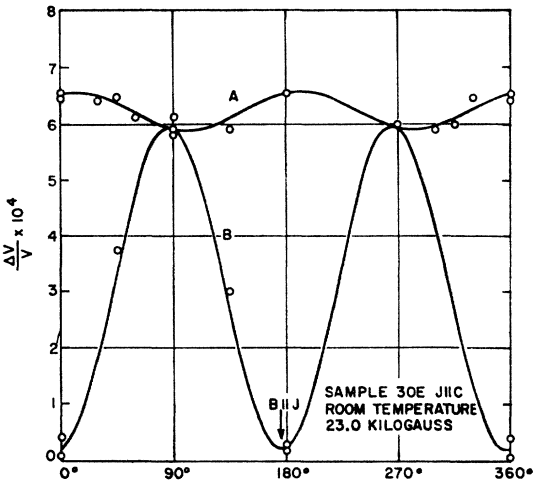

FIG. 2. Typical transverse and longitudinal magnetoresistance. The relative change in voltage which is equal to the relative change in resistivity is plotted as a function of angle as the sample is rotated with respect to the magnetic field. Curve A shows the angular dependence of the transverse magnetoresistance and curve B shows the variation in magnetoresistance as the magnetic field varies in direction from parallel to the current at the minima to perpendicular to the current at the maxima.

that the sample could be rotated continuously around an axis perpendicular to the magnetic field, and so that the sample axis could be changed externally from a direction parallel to the rotation axis to perpendicular to the rotation axis in order to make purely transverse or combined longitudinal and transverse magnetoresistance measurements without removing the sample from the temperature bath. The sample holder accommodated samples up to $\frac{1}{2}$ in. in length and the complete assembly, including the Dewar, fit inside the 1-in. gap of the magnet.

The magnet was a 12-in. Varian electromagnet with tapered polefaces which could give fields up to 24 kG. The magnetic fields were measured with a Rawson rotating-coil fluxmeter which was accurate to 1%. The voltage measurements were made with a Leeds and Northrop potentiometer (type K-3) and galvanometer. The entire circuit (except for resistors) was copper and stray thermal emf's were found to be negligible. Magnetoresistance and Hall measurements were made in the usual way, by averaging the readings obtained by reversing the magnetic field. No changes in the Hall or magnetoresistance voltages were observed when the current through the samples was reversed, so that reversing the current was not part of the measurement procedure except as an occasional check.

Results

Preliminary measurements on several samples (ranging in room temperature resistivity from 50 to 0.1 Ω-cm) at the three temperatures showed considerable variation in the galvanomagnetic constants from sample to sample. Firstly, a large range of Hall mobilities was found, especially at 77°K where the mobilities ranged from 260 to 5500 cm²/V-sec. Such measurements are consistent with the data of Piper and Halstead[10] who measured resistivity and Hall effect in CdS over a wide range of temperatures and impurity concentrations. Indeed, the mobilities and resistivities in most of the preliminary samples agreed almost exactly with the curves of Piper and Halstead. Their experiments show that the wide range of mobilities could be explained by the varying amounts of ionized impurity scattering in different samples.

Secondly, the preliminary measurements showed that there were large differences in the anisotropies of the transverse magnetoresistance and in the ratio of longitudinal to transverse magnetoresistance. Masumi[11,12] measured magnetoresistance in CdS using a two-terminal method and found a sizable longitudinal magnetoresistance for current along the c axis but negligible longitudinal magnetoresistance for current perpendicular to the c axis. Some of the preliminary samples in the present study had appreciable longitudinal magnetoresistance for both current directions, but others had negligible longitudinal magnetoresistance. Since longitudinal magnetoresistance can arise due to side contacts[9,13] or to inhomogeneities,[14] it was apparent that it is important to select samples which were as homogeneous as possible, and so the samples on which detailed measurements were to be made were selected

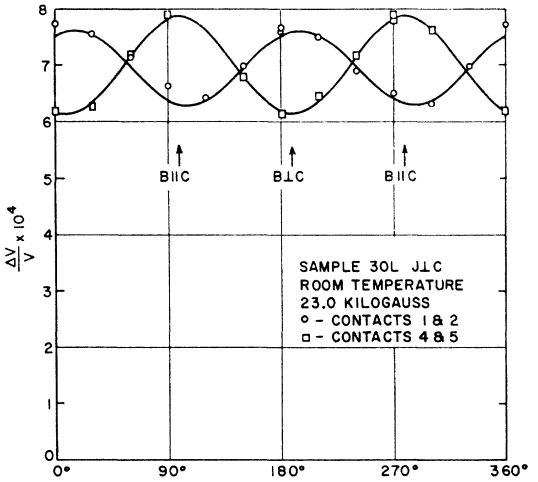

FIG. 3. Typical transverse magnetoresistance measured by two sets of voltage contacts on the same sample. The ordinate gives the relative change in voltage on the two sets of contacts which are labeled as shown in Fig. 1.

[10] W. W. Piper and R. E. Halstead, in *Proceedings of the International Conference on Semiconductor Physics, Prague, 1960* (Academic Press Inc., New York, 1961), p. 1046.
[11] T. Masumi, J. Phys. Soc. Japan **14**, 47 (1959).
[12] Y. Suge, S. Tanaka, and T. Masumi, in *Solid State Physics in Electronics and Telecommunications (Proceedings of the International Conference held in Brussels, June 2–7, 1958)*, edited by M. Desirant and J. L. Michiels (Academic Press Inc., New York, 1960), Vol. 2, p. 872.
[13] M. Glicksman, J. Phys. Chem. Solids **8**, 511 (1959).
[14] C. Herring, J. Appl. Phys. **31**, 1939 (1960).

TABLE I. Galvanomagnetic constants at room temperature (297°K).[a]

Sample number	ρ_{11} $1/ne\mu_1$	ρ_{33} $1/ne\mu_3$	ρ_{123} ρ_{11} $-a\mu_1$	ρ_{231} ρ_{11} $-a\mu_1$	ρ_{231} ρ_{33} $-a\mu_3$	$\left(\frac{\rho_{1111}}{\rho_{11}}\right)\times10^{-4}$ 0	$\left(\frac{\rho_{3333}}{\rho_{33}}\right)\times10^{-4}$ 0	$\left(\frac{\rho_{1122}}{\rho_{11}}\right)\times10^{-4}$ $b\mu_1\mu_3$	$\left(\frac{\rho_{1133}}{\rho_{11}}\right)\times10^{-4}$ $b\mu_1^2$	$\left(\frac{\rho_{3311}}{\rho_{33}}\right)\times10^{-4}$ $b\mu_1\mu_3$	$\left(\frac{\rho_{2323}}{\rho_{11}}\right)\times10^{-4}$ $b\mu_1^2$	$\left(\frac{\rho_{2323}}{\rho_{33}}\right)\times10^{-4}$ $b\mu_1\mu_3$
37A	3.90		382	388		0.07		1.14	1.10			
37C		4.35			350, 354		0.08			1.13, 1.10		
30A	3.124		410	413		0.11			1.19			
30B	3.56		364	364		0.05		1.33	1.31			
30G	3.196		438	427		0.06		1.05	1.06			
30L	3.70		418	390		0.11		1.16	1.18		1.45	
30N	3.98		354	396		0.13		1.23	1.16		1.26	
30E		3.75			369, 387		0.05			1.11, 1.07		1.19
30J		3.64			386, 378		0.01			1.15, 1.09		

[a] The resistivities are in Ω-cm, the mobilities are in cm²/V-sec, and the magnetoresistance coefficients divided by the resistivities are in cm⁴/V²-sec².

for low inhomogeneity and high mobilities as described above.

The Hall effect measurements in the selected samples showed that the samples were n type and the Hall voltages always exhibited the correct dependence on the angle of the magnetic field with respect to the sample. The magnetoresistance measurements, however, showed appreciable disagreement with the phenomenological theory as illustrated for typical samples in Figs. 2, 3, and 4. Similar curves were obtained for all the samples at each of the three temperatures, and in each case it was found that $\Delta V/V$ varied sinusoidally (to within the accuracy of the measurements) as the sample was rotated about an axis perpendicular to the magnetic field.

Figure 2 illustrates measurements of $\Delta V/V$ for a sample having current parallel to the c axis. According to the phenomenological theory $\Delta V/V$ should be constant when the magnetic field is transverse, but in all such cases a sinusoidal variation was observed (curve A). The longitudinal magnetoresistance (minimum of curve B) was quite small compared to the transverse magnetoresistance as measured on two sets of contacts on a sample with current perpendicular to the c axis. The fact that the curves do not coincide for both sets of contacts is a violation of the phenomenological theory. Figure 4 illustrates the planar Hall effect measurements in the same sample. In this case, the fact that the curve is not centered about zero is a violation of the phenomenological theory.

The violations cited above are undoubtedly due to the asymmetry introduced by the voltage contacts, as is clearly shown from the fact that the minimum for a particular transverse magnetoresistance curve occurs roughly at an angle such that the magnetic field is in the plane of the surface on which the measuring contacts are located. When the magnetic field is perpendicular to the side on which the contacts are located, the contacts partially short out the Hall electric field, distorting the current and thus giving rise to additional magnetoresistance. In such cases it is correct[9] (at least to a first approximation) to take the minimum of each curve as the measure of the true magnetoresistance (in the limit of point contacts). It is not clear how one should estimate the true planar Hall effect from curves such as Fig. 4, so for lack of a better procedure, the average electric fields with magnetic field 45° from the current direction were taken as the measure of the planar Hall effect.

The results of the measurements on the preselected samples are shown in Tables I and II. The resistivities are in Ω-cm, mobilities are in cm²/V-sec, and the magnetoresistance coefficients divided by the resistivities are in cm⁴/V²-sec². In some of the measurements of ρ_{231}/ρ_{33} and ρ_{3311}/ρ_{33} two numbers are given which were measured on two different sets of voltage probes on the same sample, and the variations in these quantities as well as the variations from sample to sample are indicative of the errors in the measurements due to contacts, inhomogeneities, etc. The magnetoresistance coeffi-

TABLE II. Galvanomagnetic constants at 196 and 77°K.[a]

Sample number	ρ_{11} $1/ne\mu_1$	ρ_{33} $1/ne\mu_3$	ρ_{123} ρ_{11} $-a\mu_1$	ρ_{231} ρ_{11} $-a\mu_1$	ρ_{231} ρ_{33} $-a\mu_3$	$\left(\frac{\rho_{1111}}{\rho_{11}}\right)\times10^{-4}$ 0	$\left(\frac{\rho_{3333}}{\rho_{33}}\right)\times10^{-4}$ 0	$\left(\frac{\rho_{1122}}{\rho_{11}}\right)\times10^{-4}$ $b\mu_1\mu_3$	$\left(\frac{\rho_{1133}}{\rho_{11}}\right)\times10^{-4}$ $b\mu_1^2$	$\left(\frac{\rho_{3311}}{\rho_{33}}\right)\times10^{-4}$ $b\mu_1\mu_3$	$\left(\frac{\rho_{2323}}{\rho_{11}}\right)\times10^{-4}$ $b\mu_1^2$	$\left(\frac{\rho_{2323}}{\rho_{33}}\right)\times10^{-4}$ $b\mu_1\mu_3$
30L, 196°K	1.99		785	851		0.20		4.91	4.79		7.52	
30N, 196°K	2.10		785	833		0.36		5.30	5.35		5.79	
30E, 196°K		1.99			792, 862		0.19			4.63, 4.67		4.56
30J, 196°K		1.89			819, 817		0.31			4.96, 5.09		7.36
37C, 77°K	0.740				4270, 4280	8				112		
37A, 77°K		0.634	5330	5240		4		101	114			
30L, 77°K	0.578		5800			1		125	133		232	
30N, 77°K	0.661		5330	5670		2		121	128		160	
30E, 77°K		0.769			4550		2			107		111
30J, 77°K		0.749			4420, 4320		4			117, 122		137

[a] The resistivities are in Ω-cm, the mobilities are in cm²/V-sec, and the magnetoresistance coefficients divided by the resistivities are n cm⁴/V²-sec².

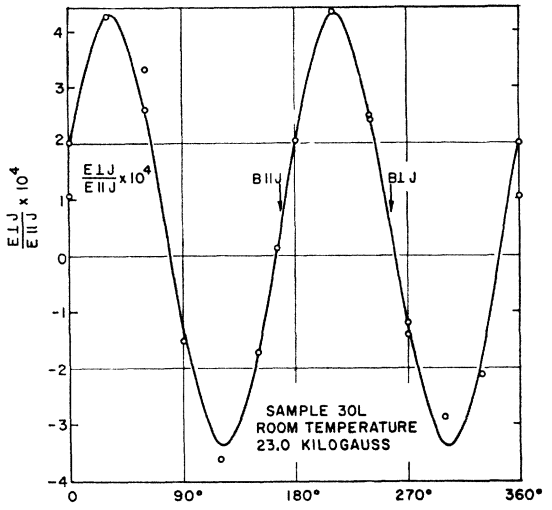

FIG. 4. Typical planar Hall effect. The ordinate has been normalized by dividing the planar Hall field by the electric field parallel to the current.

cients at 77°K were obtained by plotting $\Delta V/B^2V(B)$ as a function of magnetic field squared and extrapolating to zero field as illustrated in Fig. 5, since at this temperature the mobility was high enough that higher order galvanomagnetic constants were becoming important.

The expressions in the second rows of the headings of the two tables give the measured quantities in terms of a simple single-valley model of the energy surfaces in CdS, and can be obtained by inverting the magnetoconductivity coefficients obtained by Abeles and Meiboom.[15] The model has the parameters n (the electron density), μ_1 and μ_3 (the average drift mobilities parallel and perpendicular to the c axis), and the parameters a and b which depend on the energy dependence of mobility:

$$a=\frac{\langle\mu^2\rangle}{\langle\mu\rangle^2} \quad \text{and} \quad b=\frac{(\langle\mu^3\rangle\langle\mu\rangle-\langle\mu^2\rangle^2)}{\langle\mu\rangle^4}, \tag{4}$$

where μ is the mobility in any particular direction and the averages are Maxwellian averages with an additional weighting factor of the energy.[16,17] The parameter a is the ratio of the Hall mobility to the average drift mobility and is a number close to unity. Neither a nor b can be determined from low-field measurements directly, but the quantity

$$\xi=b/a^2=(\langle\mu^3\rangle\langle\mu\rangle/\langle\mu^2\rangle^2)-1 \tag{5}$$

can be determined by dividing the magnetoresistance coefficients by the resistivity and by the appropriate Hall mobilities.

[15] B. Abeles and S. Meiboom, Phys. Rev. **95**, 31 (1954).
[16] H. Brooks, in *Advances in Electronics and Electron Physics*, edited by L. Martin (Academic Press Inc., New York, 1955), Vol. 7, p. 85.
[17] C. Herring and E. Vogt, Phys. Rev. **101**, 944 (1956).

Measurements in Two-Terminal Samples

A comparison of the measurements and corresponding theoretical expressions in the two tables shows that the data fit the single-valley model within experimental error (see the next section). An obvious characteristic of the data is that the two Hall mobilities $\mu_{H1}=a\mu_1$ and $\mu_{H3}=a\mu_3$ are very nearly equal especially at the two highest temperatures. The mobility ratio is difficult to determine accurately from the resistivity or Hall data because of the variations from sample to sample or from transverse magnetoresistance measurements because of the errors introduced by the finite size of the voltage contacts.

In order to obtain more accurate values of the mobility ratio by magnetoresistance methods, transverse magnetoresistance measurements on two-terminal samples were made at the three temperatures. In such measurements the contact resistances are not eliminated directly, but can be subtracted out if the resistivity of the sample is known. Also, the measured magnetoresistance includes an apparent magnetoresistance due to the shorting of the Hall voltage by the end contacts. The theory of the effect of the geometrical shape of a two-terminal sample on the measured magnetoresistance has been given by various authors[18–20] for the case of cubic symmetry and is generalized in the appendix for the present case. The change in voltage

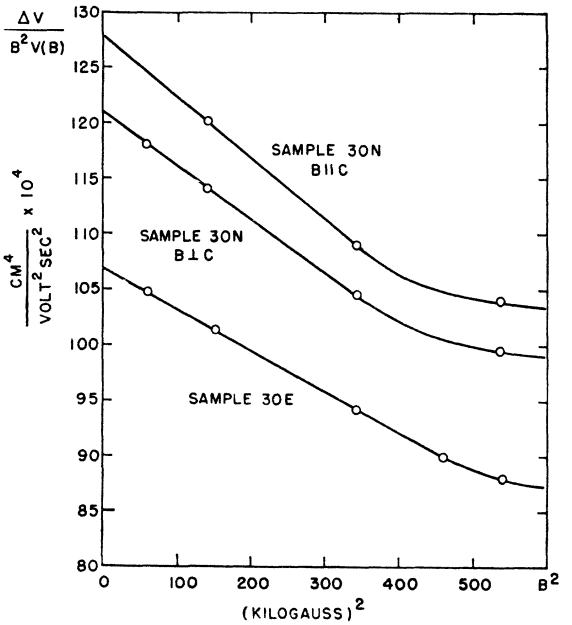

FIG. 5. Typical field dependences of magnetoresistance at 77°K illustrating the method used for extrapolating to zero field. $\Delta V/B^2V(B)$ was found to give a straight line plot vs B^2 over a wider range of B^2 than $\Delta V/B^2V(0)$.

[18] J. R. Drabble and R. Wolfe, J. Elec. Cont. **3**, 259 (1959).
[19] H. J. Lippman and F. Kuhrt, Z. Naturforsch. **A13**, 462 (1958).
[20] Y. I. Gorkun, Soviet Phys.—Solid State **3**, 173 (1961).

TABLE III. Galvanomagnetic effects in two-terminal samples.

Sample number	297°K $\Delta V_{\|\|}$[a] (calc)	297°K $\Delta V_{\|\|}$ (meas)	297°K μ_1/μ_3	196°K $\Delta V_{\|\|}$ (calc)	196°K $\Delta V_{\|\|}$ (meas)	196°K μ_1/μ_3	77°K $\Delta V_{\|\|}$ (calc)	77°K $\Delta V_{\|\|}$ (meas)	77°K μ_1/μ_3
30B	0.211	0.200	1.022	0.194	0.199	1.029	23.0	20.8	1.425
30D	0.215	0.222	1.140	1.140	1.032	1.072	14.70	11.53	1.303
30H	0.224	0.252	1.073	1.083	1.040	1.062	14.37	11.55	1.230
30M1	0.491	0.494	1.009	1.092	1.143	1.018	16.08	16.12	1.418
30M2	0.489	0.511	1.012	1.086	1.150	1.067	15.39	16.39	1.238

[a] The values of $\Delta V_{\|\|}$ are in millivolts.

across the sample is given by the following formulas:

$$\Delta V_{\|\|}=\rho_{11}J\mu_{H1}^2B^2[\xi+0.5428W_{\|\|}/L] \quad (6)$$

for $B\|\|C\perp J$, and

$$\Delta V_{\perp}=\rho_{11}J\mu_{H1}\mu_{H3}B^2[\xi+0.5428(W_{\perp}/L)(\mu_1/\mu_3)^{1/2}] \quad (7)$$

for $B\perp C\perp J$, where L is the length of the sample and $W_{\perp}$ and $W_{\|\|}$ are the widths of the sides of the sample through which the magnetic field passes in each case. By measuring $\Delta V_{\|\|}$ one can test one's knowledge of ρ_{11}, μ, and ξ in the sample, and by measuring $\Delta V_{\|\|}/\Delta V_{\perp}$ one can solve a quadratic equation for the ratio of mobilities μ_1/μ_3. The value of μ_1/μ_3 determined in this way does not depend on the values of ρ_{11} or μ_{H1} but only on the dimensions of the sample and the value of ξ (and, in fact, small changes in ξ result in much smaller changes in μ_1/μ_3) so that this method should give values of μ_1/μ_3 which are considerably more accurate than measurements which involve comparisons between samples or between different pairs of voltage contacts on the same sample.

The results of the two-terminal measurements on five different samples at each of the three temperatures are given in Table III. The values of $\Delta V_{\|\|}$ are in millivolts at a magnetic field of 23.0 kG for the particular value of current used in each measurement. The agreement between the calculated and measured values is fairly good, and the differences indicate variations of ρ_{11} or μ, from sample to sample. The ratios μ_1/μ_3 obtained in different samples do indeed show a smaller spread than the rest of the data. Samples 30D and 30H showed significant differences from the other samples at room temperature, so their contacts were removed and it was discovered that the solder had not covered the ends uniformly. New contacts were used for the measurements at the other temperatures and the results agreed with those of the other samples, so their room temperature ratios were not included in the averages shown in Table IV.

Table IV summarizes the results of all the experiments by giving values of the parameters in the single-valley model which best fit the data at each of the three temperatures. The average ratios obtained from the two-terminal measurements were used in calculating the other averages; e.g., the Hall mobilities μ_{H3} were multiplied by the appropriate value of μ_1/μ_3 at each temperature and were averaged in with the measurements of μ_{H1}. The planar Hall coefficients were not included in the averages because they include larger errors due to the other side contacts than did the transverse magnetoresistance coefficients. The errors given in the table include only the spread of the measurements from sample to sample and do not include any estimate of the systematic errors which might exist.

DISCUSSION

Single Ellipsoid Model

A comparison of the data given in Tables I and II with the theoretical expressions near the heads of the columns shows that the data fit the single-valley model within experimental error. Firstly, the longitudinal magnetoresistance for both current directions is much smaller than the transverse magnetoresistance. The fact that a nonzero longitudinal magnetoresistance is observed could be partly due to misalignment, and partly due to the fact that the current was not parallel at all points within the sample due to inhomogeneities and to the side contacts.

Secondly, the two Hall coefficients are equal to within experimental error (as was also observed by Piper and Halsted[10]); and the transverse magnetoresistance coefficients are also consistent with the theoretical expressions. The degree to which the data fit the model can be judged from the fact that the spread in the values of the parameters listed in Table IV is not appreciably larger than the spread in the values of the galvanomagnetic constants.

Another reason, however, for believing from this experiment that the single-valley model applies to CdS is that the data strongly disagree with other proposed models such as those illustrated in Fig. 6. Several authors[21–25] have discussed the shapes of energy surfaces

TABLE IV. Average parameter values.

T (°K)	$a\mu_1=\mu_{H1}$	μ_1/μ_3	$\xi=a/b^2$	n/a (Sample 30)
297	-388 ± 24	1.014 ± 0.005	0.077 ± 0.008	4.51×10^{15}
196	-837 ± 35	$1.05\ \pm0.02$	0.074 ± 0.006	3.78×10^{15}
77	-5630 ± 240	$1.32\ \pm0.09$	0.045 ± 0.005	1.81×10^{15}

[21] R. C. Casella, Phys. Rev. **114**, 1514 (1959); Phys. Rev. Let-

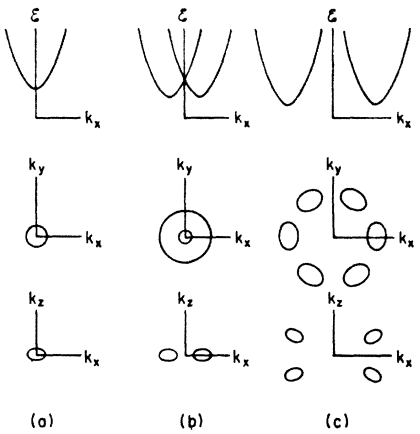

FIG. 6. Possible band structure in wurtzite-type crystals. (a) single ellipsoid. (b) toroid. (c) simple many valley. The upper figures show the relationship between energy and wave number; the middle and lower figures show cross sections of surfaces of constant energy.

which are possible (according to group theory) in wurtzite-type crystals. They show that in the absence of spin-orbit splitting the conduction band should have a minimum at $\mathbf{k}=0$ [Fig. 6(a)] but that spin-orbit coupling may lead to toroidal energy surfaces[21,22] [Fig. 6(b)] or to a many-valley type of band structure[23] [Fig. 6(c)]. Figure 6 illustrates the three distinct types, but it is certainly possible (in principle) to have all three and also the intermediate cases in the same crystal as the temperature (or Fermi energy) is changed relative to the size of the spin-orbit coupling. Magnetoresistance in the three different types should be quite different. In the single-valley model magnetoresistance arises only from the spread in mobilities due to the dependence of scattering time on energy. Any other type of band structure gives rise to transverse currents (whose sum must be zero) in the absence of a magnetic field, and in the presence of a magnetic field such a band structure exhibits longitudinal magnetoresistance and much larger transverse magnetoresistance than the single-valley model. For example, in the simple many-valley model the parameter ξ is given by

$$\xi=(\langle\mu\rangle\langle\mu^3\rangle/\langle\mu^2\rangle^2)F(k)-1, \tag{8}$$

where $F(k)$ is a function of the anisotropy of the valleys and the orientation of the current and magnetic field with respect to the valleys.[16] Thus in semiconductors with the many-valley band structure (such as Ge and Si) ξ is approximately equal to unity; but in CdS the largest value of ξ was only 0.08, an order of magnitude smaller. Casella[21] has calculated galvanomagnetic coefficients for the toroidal model and finds appreciable longitudinal magnetoresistance as well as a strong temperature dependence in the anisotropy of the Hall coefficients, neither of which were found in the present data. The present data do not exclude the possibility, of course, that effects due to toroidal surfaces are present at temperatures lower than 77°K.

There have been several other recent experiments which also point to a single valley model. Piper and Halsted[10] measured the temperature dependence of the Hall coefficient in the region where it was changing rapidly with temperature, and interpreted the results in terms of a simple hydrogen-like model of the donor level, thus arriving at an electron effective mass $m^*=0.19m_e$. They also fitted their Hall mobility measurements to theoretical expressions which included polar optical mode and piezoelectric scattering, thus obtaining a value of $m^*=0.16m_e$. This second value relied on an early estimate of the piezoelectric scattering by Hutson,[26] and an improved estimate by Hutson[4] would increase Piper and Halsted's value of the effective mass. Hopfield and Thomas[1,27] have studied exciton lines in the reflection and absorption spectra of CdS, and concluded that the single-valley model applied to CdS. From studies of the Zeeman effect[1] of the exciton lines they were able to obtain values of the effective masses of both electrons and holes. They concluded the electronic effective mass is given by $m^*=(0.204\pm0.010)m_e$, and is isotropic within experimental error, although they did find a small anisotropy:

$$m_1^*/m_3^*=0.209/0.198=1.05.$$

Hopfield[28] has also been able to place an upper limit on the value of $(d\epsilon/dk)_{k=0}$, i.e., an upper limit on the temperature at which toroidal effects might become important. This upper limit was much less than one degree Kelvin, so it is improbable that such effects will be observed in any experiment on CdS.

Piper and Marple[2] studied free electron absorption in the infrared spectrum of doped CdS and agreed with Hopfield and Thomas about the size and anisotropy of the free electron mass, finding $m^*=(0.22\pm0.01)m_e$ and $m_1^*/m_3^*=1.08\pm0.05$.

Thus the structure of the conduction band minima in CdS is well established as being characterized by an almost isotropic mass $m^*=0.21m_e$. The only data that have not been explained with this model are the blue emission lines seen by Pedrotti and Reynolds,[29] but it

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may be possible to explain these lines in terms of excitons bound to impurities as suggested by Thomas and Hopfield.[30] The anisotropy in the mobility measured in the present experiments is opposite to the mass anisotropy found in the optical studies, i.e., $\mu_1/\mu_3=(\tau_1/\tau_3)\times(m_3/m_1)>1$, but $m_3/m_1<1$. The differences can be explained if the scattering is anisotropic ($\tau_1/\tau_3>1$). The fact that the mobility ratio is temperature dependent also supports the hypothesis of anisotropy in the scattering, since the relative importance of the different scattering modes can change with temperature.

Lattice Scattering

In a partially ionic lattice such as CdS, one would expect polar optical mode scattering to be the main mechanism for lattice scattering. The theory of polar optical mode scattering has been reviewed by Frölich[3] who showed that a perturbation theory is applicable for values of α (a coupling constant) small compared with unity. Since α is equal to 0.3 for CdS, one might expect the theory to apply reasonably well. The formula for mobility given by Fröhlich can be written as

$$\mu_0=(16.91/\Theta^{1/2})(1/n^2-1/\kappa)^{-1}(m_e/m^*)^{3/2}\times(e^{\Theta/T}-1)\ \text{cm}^2/\text{V-sec}, \quad (9)$$

where κ is the static dielectric constant, n is the optical index of refraction, and Θ is the characteristic temperature of longitudinal optical phonons.

Piezoelectric scattering is also expected in CdS and the theory has been applied to CdS by Hutson.[4] Piezoelectric scattering can occur only in crystals showing a piezoelectric effect, and is due to the electric fields produced by acoustic phonons. Hutson[4] writes the piezoelectric mobility as

$$\mu_p=1.44\kappa(m/m^*)^{3/2}(300/T)^{1/2}\{\sum_{\text{modes}}(K^2)_{\text{av}}\}^{-1}, \quad (10)$$

where K (the electromechanical coupling factor) is a dimensionless quantity given by $K^2=e^2/\kappa\epsilon_0 c$, where e is an appropriate piezoelectric constant and c is the appropriate stiffness coefficient. Hutson[4] has also given the formulas for averaging the squares of the piezoelectric constants for the longitudinal and transverse modes for the case of wurzite symmetry.

Equations (9) and (10) can be combined (by adding reciprocals of mobilities) to give an approximate theoretical formula for lattice scattering. The constants which appear in the expression have all been recently measured: the piezoelectric constants,[26] the dielectric constant[31] [$\bar{\kappa}=(1/3)(2\kappa_1+\kappa_3)=9.19$], the index of refraction[32] ($n^2=5.24$) and the optical mode frequency[33] ($\Theta=440°$K). One can then use the effective mass as the only adjustable parameter and can solve for it at each temperature using the measured mobility. At room temperature the average mobility is $\bar{\mu}_H=(1/3)\times(2\mu_{H1}+\mu_{H3})=385$ cm^2/V-sec which yields an effective mass $m^*=0.185m_e$; similarly, the dry ice value of average mobility yields $m^*=0.192m_e$, so that the simple theory gives the correct temperature dependence to within experimental error and yields a value of effective mass which is in quite reasonable agreement with the optical experiments. Actually there are several sources of error in the above procedure. Firstly, we have assumed that $a=1$ (that drift mobility equals Hall mobility), and this assumption tends to lower the deduced effective mass. Approximate values of a for pure optical mode scattering have been given by Lewis and Sondheimer,[34] but the value of a could be changed due to the relatively small amount of piezoelectric scattering. Secondly, an error is incurred from the use of (9) which is only correct in the lower temperature limit. Howarth and Sondheimer[35] have calculated the corrections using a variational technique. Below the Debye temperature the use of (9) instead of Howarth and Sondheimer's numerical results gives too high an estimate of the effective mass. Thirdly, there is an error due to adding reciprocal mobilities. A rigorous derivation of the lattice mobility in CdS would require the use of variational techniques for calculating mobility due to combined scattering mechanisms,[36] and in such calculations much numerical computation is required to achieve accuracies of even 10%. Such an effort is not justified at present because of uncertainty in the relevant constants for CdS as well as the spread in the mobility data. In summarizing the above considerations, we could say that the mobility data point to an electron effective mass of $m^*=(0.19\pm0.02)$ which includes a somewhat conservative estimate of the errors incurred by using the simple theory.

Ionized Impurity Scattering

The mobility measured at 77°K is considerably lower than the value predicted for pure lattice scattering (7200 cm^2/V-sec assuming $m^*=0.19m_e$). Since the discrepancy is probably due to ionized impurity scattering, it is of interest to see how much compensation (if any) is required to explain the magnitude of the measured mobility. The Brooks-Herring formula for ionized impurity scattering has been discussed by Long and Myers.[37] The conditions for applicability of the theory are met quite well in CdS. To apply the formula to the present case we again assume that $a=1$ and that

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reciprocals of mobility are additive. At room temperature all the shallow donor sites are ionized, so we can assume that the electron density (4.51×10^{15} cm^{-3} in sample 30) is equal to the density of shallow donors minus the density of acceptors. Using a value of $m^*=0.19m_e$ we can then calculate the ratio of donor density to acceptor density needed to explain the mobility at 77°K. The result is that $N_D/N_A=1.19$ which means that there was considerable compensation. The amount of compensation is somewhat surprising in view of the degree of homogeneity of the samples as reflected in the spread in mobility values.

Anisotropic Scattering

A significant feature of the anisotropy data is that the anisotropy increases as the relative amounts of piezoelectric scattering and impurity scattering increase. Ionized impurity scattering can be anisotropic due to either mass anisotropy[38] or to dielectric anisotropy.[39] The dielectric anisotropy of CdS is not agreed on [31,40] so that it is not possible to estimate the anisotropy of the ionized impurity scattering. The temperature dependence of the anisotropy fits quite well with what would be expected if the piezoelectric scattering were anisotropic. To show that this is true, let us assume that both the optical mode scattering and impurity scattering are isotropic so that we can combine them (at any given temperature) and the combined mobility μ_c will exhibit only the anisotropy due to the effective mass. Now if we define the average mobilities as $\bar{\mu}=(2\mu_1+\mu_3)/3$ and calculate $1/\bar{\mu}_c=1/\bar{\mu}_m-1/\bar{\mu}_p$, where $\bar{\mu}_p$ is given by (10) with $m^*=0.19m_e$, $\bar{\mu}_c$ is the combined mobility due to all other types of scattering and $\bar{\mu}_m$ is the average measured mobility, then, by trial and error, the anisotropies of μ_c and μ_p can be adjusted (keeping $\bar{\mu}_m$ and $\bar{\mu}_p$ fixed) so that a best fit to the experimental values of μ_1/μ_3 are obtained at the three temperatures. The result of such a procedure gives

$$\mu_{p1}/\mu_{p3}=1.55\pm0.12 \quad \text{and} \quad \mu_{c3}/\mu_{c1}=1.03\pm0.02,$$

where the error is due to the error in the mobility ratios. The two parameters fit the three experimental ratios quite well, and the above value of μ_{c3}/μ_{c1} agrees quite well with the mass ratios determined in the optical experiments.

Actually, the theory of piezoelectric scattering in wurtzite-type crystals predicts that the mobility will, in general, be anisotropic.[41] If one has spherical energy surfaces and dielectric and elastic isotropy, the anisotropy is given by

$$\frac{\mu_{p1}}{\mu_{p3}}=\frac{126+60r+14r^2}{147+54r+11r^2}, \tag{11}$$

where $r=(e_{33}-e_{31}-2e_{15})/e_{15}$, and e_{15}, e_{31}, and e_{33} are the piezoelectric constants. Equation (11) includes only the scattering due to the shear modes, but this is a good approximation since the velocity of longitudinal waves is considerably larger than transverse waves, thus leading to values of $(K^2)_{av}$ which are an order of magnitude smaller. For CdS, $r=-5.5$ (using the piezoelectric constants measured by Hutson[4]) and so Eq. (11) predicts $\mu_{p1}/\mu_{p3}=1.20$. Inclusion of the longitudinal modes[41] gives $\mu_{p1}/\mu_{p3}=1.21$. Thus, the data indicate a larger anisotropy than predicted. In fact, the anisotropy at 77°K is larger than would be expected even if the scattering mechanism were purely piezoelectric. It is unlikely that the theoretical value as given by Eq. (11) would be changed significantly if the effects of mass, dielectric and elastic anisotropy were taken into account, and so it seems likely that the impurity scattering is also anisotropic in the same direction as the piezoelectric scattering. If the anisotropy is due to ionized impurity scattering, the dielectric anisotropy would have to have the direction and approximate magnitude given by Masumi,[40] rather than the direction given by Berlincourt *et al.*[31] It is also possible that the anisotropy is due to some other form of defect scattering, such as scattering from dislocations or local inhomogeneities.[42]

CONCLUSION

The results of the measurements of Hall mobility and magnetoresistance at the three temperatures used in this experiment agree quite well with the band structure deduced from optical studies. The theories of scattering in polar semiconductors explain the magnitude and temperature dependence of the electron mobility on the assumption that $m^*=0.19m_e$. The temperature-dependent anisotropy of the mobility is in the direction predicted for piezoelectric scattering, but is larger than predicted. In view of the possibility of anisotropic defect scattering, the disagreement is not serious. Further experiments on crystals of higher purity would clarify the source of the anisotropy and the nature of the defect scattering.

In principle, one could also compare experiment and theory by explaining the magnitude of the magnetoresistance parameter ξ in terms of the proposed band structure and scattering mechanisms. Such a procedure is impossible at present, however, due to the lack of a theory of low-field magnetoresistance in the presence of optical mode scattering at temperatures below Θ.[34]

ACKNOWLEDGMENTS

The authors wish to express their appreciation to Dr. R. C. Reynolds and co-workers at the Wright-Patterson Aeronautical Research Center for the generous supply of high quality crystals, and valuable in-

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formation on the preparation of samples and electrical contacts; to D. H. Hensler for help in calibrating the magnetic field; and to the Honeywell Research Center for providing encouragement in developing the theory of piezoelectric scattering anisotropy and for assistance in preparing the manuscript for publication.

APPENDIX

Effect of Sample Geometry on Two-Terminal Magnetoresistance Measurements

Gorkun's method[20] is most easily adapted to the present purposes, i.e., of generalizing the theory to an anisotropic material. Gorkun assumes a sample in the shape of a right-angle parallelepiped with length $2l$, width $2w$, and thickness $2t$. The current electrodes cover the ends of the sample, current flows along the x direction, and the magnetic field is in the z direction. Let us also assume that the current and magnetic field are both perpendicular to the c axis. Then we can write the current density in terms of the electric and magnetic fields by using Eq. (1):

$$\begin{aligned} J_x&=\sigma_{11}E_x-\sigma_{231}BE_y+\sigma_{1122}B^2E_x,\\ J_y&=\sigma_{32}E_y+\sigma_{231}BE_x+\sigma_{3311}B^2E_y,\\ J_z&=0. \end{aligned} \tag{A1}$$

If we now let $\mathbf{E}=-\nabla\varphi$ and substitute Eq. (A1) into $\nabla\cdot\mathbf{J}=0$, we obtain:

$$(\sigma_{11}+\sigma_{1122}B^2)\partial^2\varphi/\partial x^2+(\sigma_{33}+\sigma_{3311}B^2)\partial^2\varphi/\partial y^2=0. \tag{A2}$$

The terms containing the Hall coefficients cancel, so that the potential satisfies Laplace's equation if we make the transformations:

$$x'=x\left(\frac{\bar{\sigma}}{\sigma_{11}+\sigma_{1122}B^2}\right)^{1/2},\quad y'=y\left(\frac{\bar{\sigma}}{\sigma_{33}+\sigma_{3311}B^2}\right)^{1/2}, \tag{A3}$$

where $\bar{\sigma}$ is a constant.

Since the variables x' and y' satisfy Laplace's equation the anisotropic problem has been transformed into an isotropic problem by a linear transformation. The isotropic problem is solved by Gorkun, and we can use his results for the anisotropic case by regarding them as being in the primed coordinate system, and simply transforming back to the unprimed system using (A3). From Gorkun's Eq. (26) we have, for two-terminal samples:

$$\frac{1}{B^2}\left(\frac{\Delta\rho_{11}}{\rho_{11}}\right)_m=\frac{\rho_{1122}}{\rho_{11}}+\rho_{231}^2\sigma_{11}\sigma_{33}\left[1+\beta_1\left(\frac{l'}{w'}\right)\right], \tag{A4}$$

where $(\Delta\rho_{11}/\rho_{11})_m$ is the measured relative change in resistivity of the sample and $\beta_1(l'/w')$ is a function of the length to width ratio in the primed coordinate system. The quantities ρ_{1122}, ρ_{231}, ρ_{11}, σ_{11}, and σ_{33} do not appear in Gorkun's equation, of course; they must be substituted into the derivation to replace the cubic galvanomagnetic constants used by Gorkun.

To make sure that the proper substitutions have been made, we can check the limiting cases. For the case of infinitely long samples the function β_1 approaches unity, and the measured change in resistivity approaches the true magnetoresistivity ρ_{1122}/ρ_{11}. At the limit of infinitely short samples, the function β_1 approaches zero, and the measured change in resistivity approaches $\rho_{1122}/\rho_{11}+\rho_{231}^2\sigma_{11}\sigma_{33}$. Because of the relationship between the coefficients in Eqs. (1) and (2), however, this expression exactly equals the magnetoconductivity $\sigma_{1122}/\sigma_{11}$, so that again, the proper limit is approached.

The exact expression for β_1 is given by

$$\beta_1\left(\frac{l'}{w'}\right)=\frac{16}{\pi^3}\frac{l'}{w'}\sum_{n=0}^{\infty}\tanh\left(\frac{2n+1}{2}\pi\frac{w'}{l'}\right)\Big/(2n+1)^3. \tag{A5}$$

Since this expression is tedious to evaluate, approximate forms can be used in the limits of long and short samples. By comparing with the paper by Lippman and Kuhrt,[19] the limiting forms can be seen to be

$$\beta_1(l'/w')=1-kw'/l' \quad \text{as} \quad l'/w'\to\infty,$$

and

$$\beta_1(l'/w')=kl'/w' \quad \text{as} \quad l'/w'\to 0,$$

where

$$k=\frac{16}{\pi^3}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^3}=0.5428. \tag{A6}$$

By numerical evaluation of Eq. (A5) it can be shown that for $l'/w'>1.5$, the error incurred by using the approximation is less than 1%.

Equation (7) is obtained from (A4) by using the approximation for long samples and the expressions for the galvanomagnetic constants in the single valley model. When the magnetic field is along the c axis there is isotropy in the (x,y) plane, so that Eq. (6) can be derived by replacing μ_{H3} by μ_{H1} and μ_3 by μ_1.

The paper by Drabble and Wolfe[18] disagrees with the results of Gorkun and Lippman and Kuhrt because they obtain the constant $(2/\pi)^3=0.258$ instead of the constant 0.5428.