The microbalance measurements on post-bombardment heating of germanium and silicon have generally shown at most about 5×10^{14} trapped argon atoms/cm², or less than one atom per surface atom. There are, however, occasional exceptions (D-1), when as many as 10¹⁶ argon atoms appear to have been trapped during the sputtering process. It is difficult to explain these exceptions. It is possible that something other than argon, either in the form of bulk or surface impurities, is being removed by the heating.

V. CONCLUSION

The vacuum microbalance has been shown to be a very useful tool for investigations of the phenomena of sputtering with positive ions. The extreme sensitivity of the apparatus is especially valuable for studies in the threshold region. Bombardments at low current densities and short time intervals should provide considerable information concerning the nature of the target surface and also contribute to the understanding of the basic processes involved in sputtering.

In this work the microbalance studies have enabled us to establish criteria of the reproducibility and cleanliness of the ion-bombarded surfaces prior to subsequent adsorption or kinetic studies.

ACKNOWLEDGMENTS

The author is indebted to Dr. S. Aisenberg and Mr. A. Fowler for their helpful discussions and to Mr. L. Rubin for his assistance with instrumentation problems. The continued aid of Mrs. P. M. Rodriquez is also acknowledged.

PHYSICAL REVIEW

VOLUME 108, NUMBER 5

DECEMBER 1, 1957

Galvanomagnetic Effects in *n*-Type Indium Antimonide*

H. P. R. FREDERIKSE AND W. R. HOSLER National Bureau of Standards, Washington, D. C. (Received August 12, 1957)

The magnetic field dependence of the magnetoresistive effects and the Hall coefficient have been investigated at 78°K and at liquid helium temperatures. Results at very low magnetic field strength are in agreement with the assumption of an isotropic conduction band. Quantization of the electron orbits causes deviations from the conventional theory at large fields (of the order of a few thousand gauss and higher). Oscillations in the magnetoresistance observed at 4.2°K and lower are attributed to these quantum effects. The measured period of oscillation agrees reasonably well with theoretical predictions. An effective mass value of 0.01 m_0 is obtained from the field and temperature dependence of the amplitude of the oscillations. The magnitude of the magnetoresistive effects appears to depend considerably on the geometry and inhomogeneity of the sample. Strong magnetic fields also influence the distribution of "free" and bound electrons, causing a freeze-out effect at temperatures of liquid helium.

INTRODUCTION

INDIUM antimonide is by far the most extensively studied member of the group of III-V compounds. There are two reasons why this semiconductor has received so much attention. First, it is rather easy to prepare indium antimonide in single crystals of a purity approaching that of the purest germanium and silicon, and second, it has striking physical properties: small energy gap, high mobility and small effective mass of conduction electrons. A number of experiments^{1,2} have now established a value of 0.23 ev for the width of the energy gap at $T=0^{\circ}$ K. Fan and Gobeli³ reported an optical activation energy of 0.175 ev at room temperature involving direct transitions. The room temperature mobility of electrons is 65 000 cm²/volt-sec,⁴ while

mobilities as high as 5×10^5 cm²/volt-sec have been measured in very pure samples⁵ just below 78°K. Cyclotron resonance experiments have shown that the conduction band of this material has only one minimum (at $\mathbf{k}=0,0,0$) and a very small density of states corresponding to an effective electron mass of 0.013 $m_{0.6}$ The latter value is in close agreement with results from other experiments. Hall effect and conductivity data⁷ can be fitted with $m_e^* = 0.015 m_0$, while thermoelectric power measurements⁸ give $m_e^* = 0.014 m_0$. Burstein *et al.*⁹ have observed the cyclotron resonance at infrared frequencies in intrinsic InSb and found $m_e^*=0.015 m_0$. These values pertain only to the bottom of the conduction

^{*} Research sponsored by Office of Naval Research.

¹ R. G. Breckenridge *et al.*, Phys. Rev. 96, 576 (1954).
² V. Roberts and J. E. Quarrington, J. Elec. 1, 152 (1955).
³ H. Y. Fan and G. W. Gobeli, Bull. Am. Phys. Soc. Ser. II, 1, 111 (1956)

⁴ H. Welker, Physica 20, 893 (1954).

⁵ A. C. Beer (private communication). [See also Reports of the Battelle Memorial Institute (unpublished).] ⁶ Dresselhaus, Kip, Kittel, and Wagoner, Phys. Rev. 98, 556

^{(1955).} ⁷ Hrostowski, Morin, Geballe, and Wheatley, Phys. Rev. 100,

^{1672 (1955)} ⁸ H. P. R. Frederikse and Eugenie V. Mielczarek, Phys. Rev. 99,

^{1889 (1955)}

⁹ Burstein, Picus, and Gebbie, Phys. Rev. 103, 825 (1956).

band. There are several indications¹⁰⁻¹² that the effective mass increases higher up in the band. A number of workers^{13,14} who have made a theoretical study of the band structure of InSb come to the same conclusion: Kane states that the conduction band loses its parabolic nature at an energy (above $\mathbf{k}=0,0,0$) equal to "a fraction of the band gap energy."

The isotropy of the conduction band had already been concluded from one of the first experiments on InSb, the measurement of the magnetoresistance.¹⁵ The very small longitudinal magnetoresistive effect strongly suggested spherical energy surfaces. These measurements were, however, performed on polycrystalline samples and only at room temperature. A systematic study of this effect on pure oriented single crystals over a wide temperature range seemed therefore to be desirable.

Another aspect which makes the investigation of galvanomagnetic effects in *n*-type InSb of great interest is the high electron mobility. The Wilson-Sommerfeld theory¹⁶ of these effects considers only small magnetic fields. This treatment breaks down, however, when the magnetic field strength H becomes so large that

$$\omega\tau \text{ (or }\mu H, \text{ or } l/r) \gg 1, \tag{1}$$

where $\omega = eH/m^*c = cyclotron$ frequency, $\tau = collision$ time, $\mu = \text{mobility}$, l = electron mean free path, and r = radius of curvature of electron path in magnetic field. It is then necessary to take the quantization of the electron orbits¹⁷ into account. The energy spectrum of an electron in a magnetic field is equivalent to that of a harmonic oscillator of frequency equal to the cyclotron frequency plus that of a free one-dimensional motion along the field. Argyres and Adams¹⁸ have treated the case that all electrons are in the ground oscillator state (quantum limit). On lowering the magnetic field higher levels come into play, giving rise to an oscillatory behavior of transport properties¹⁹ and susceptibility (De Haas-van Alphen effect²⁰).

The high electron mobility of InSb of the order of 10⁵ cm²/volt-sec makes it possible to observe these effects in fields as low as several kilogauss. Our results show several oscillations in the magnetoresistance of relatively pure but degenerate *n*-type InSb at helium temperatures. The quantization effects manifest them-

selves in a different way at higher temperature when the samples are nondegenerate.

In the course of this investigation we have become aware of a number of "geometry effects," which complicate galvanomagnetic measurements, especially in materials with high carrier mobilities. It has become apparent that the Hall effect, and particularly the magnetoresistance, are strongly dependent on the shape of the sample, the configuration of the measuring probes, and the inhomogeneity of the specimen used. Several investigators have recently reported similar experiences.^{21,22} These effects are especially strong in large magnetic fields; it is still an open question whether it will be possible to separate experimentally the quantization effect from the geometry effects.

Although electrons in *n*-type InSb are excited into the conduction band at nearly all temperatures, one still has to consider the donor levels because of the fact that these states are affected by the presence of a magnetic field.23 Strong fields will reduce markedly the interaction between localized impurity states and increase the activation energy. As a result a de-ionization of donor impurities will take place. This "freeze-out" effect has been observed by us and by other workers²⁴ in measurements of the Hall coefficient as a function of magnetic field strength (at helium temperatures).

SAMPLES

The samples used in these experiments were cut from single crystals, prepared from extensively zone-melted material. The dimensions of most specimens were 10 mm \times 1 mm \times 1 mm; their length directions were taken perpendicular to the length axes of the original boules in order to minimize impurity gradients. Samples 6 and 7 were properly oriented, the largest dimension coinciding with the 100- and 110-axes respectively.

Characteristics of seven samples are given in Table I. Column 2 shows that the impurity gradients are rather small except for the last sample (7).

EXPERIMENTAL TECHNIQUES

The apparatus used for the determination of conductivity, the Hall coefficient and magnetoresistance has been described in earlier publications.²⁵ In general, the sample holder was mounted in a brass envelope, which could be filled with helium gas to establish heat contact with the refrigerant bath. In some cases measurements have been made with the sample directly immersed in the cooling liquid. The magnetic field strength varied between 100 and 7000 gauss; a few

¹⁰ Keyes, Zwerdling, Foner, Kohn, and Lax, Phys. Rev. **104**, 1804 (1956).

R. P. Chasmar and R. Stratton, Phys. Rev. 102, 1686 (1956).
 H. Weiss, Z. Naturforsch. 11a, 131 (1956).
 G. Dresselhaus, Phys. Rev. 100, 580 (1955).

 ¹⁵ G. Dresselhaus, Phys. Rev. 100, 580 (1955).
 ¹⁴ E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957).
 ¹⁵ G. L. Pearson and M. Tanenbaum, Phys. Rev. 90, 153 (1953).
 ¹⁶ E.g., A. H. Wilson, *The Theory of Metals* (University Press, Cambridge, 1954).
 ¹⁷ L. Landau, Z. Physik 64, 629 (1930).
 ¹⁸ P. N. Argyres and E. N. Adams, Phys. Rev. 104, 900 (1956).
 ¹⁹ B. Davydov and I. Pomeranchuk, J. Phys. U.S.S.R. 2, 147 (1940) (1940)

 ⁽¹⁾ FO.
 ²⁰ W. J. De Haas and P. M. Van Alphen, Commun. Kamerlingh Onnes Lab. Univ. Leiden No. 212a (1930).

 ²¹ H. Weiss and H. Welker, Z. Physik 138, 122 (1954).
 ²² D. K. C. MacDonald, Phil. Mag. 2, 97 (1957).
 ²³ Yafet, Keyes, and Adams, J. Phys. Chem. Solids 1, 137 (1956).
 ²⁴ R. W. Keyes and R. J. Sladek, J. Phys. Chem. Solids 1, 143

^{(1956).} ²⁵ R. G. Breckenridge and W. R. Hosler, Phys. Rev. **91**, 793 W. B. Berg, Phys. Rev. **91**, 793 (1953); Blunt, Frederikse, and Hosler, Phys. Rev. 100, 663 (1955).

Sample	$T = 78^{\circ}$ K						
	Rª cm³/coul	$ohm^{-1}cm^{-1}$	$(R\sigma)$ cm [?] /v-sec	<i>R</i> cm³/coul	σ ohm ^{−1} cm ^{−1}	$(R\sigma)$ cm ² /v-sec	<i>n</i> cm ⁻³
1	946 862	166	1.57×10⁵	1016	54.6	5.55×104	7 ×10 ¹⁵
2	1215 1331	145	1.77×105	1182	82.2	9.70×10 ⁴	5.3 ×10 ¹⁵
3	2638 2663	86.9	2.29×10 ⁵	2610	40.3	10.5×10^{4}	2.38×10 ¹⁵
4	2709 2793	81.9	2.22×10 ⁵	3055	34.6	10.6×10^{4}	2.30×10 ¹⁵
5	4640	50.3	2.33×10 ⁵	4696	18.4	8.64×10 ⁴	1.35×1015
6(100)	5970 5465	57.8	3.45×10^{5} 3.16×10^{5}	5981	17.7	10.6 ×10 ⁴	1.0 ×10 ¹⁵
7(110)	6101 7672	49.3	3.00×10 ⁵ 3.78×10 ⁵	•••		•••	9 ×10 ¹⁴

TABLE I. Characteristics of *n*-type InSb samples.

^a The two values of the Hall coefficients were measured on two sets of Hall probes \sim 3.5 mm apart.

measurements were made in fields as great as 22 kilogauss.²⁶

Samples were mounted in a horizontal position in a sample holder that could be turned 360° . This arrangement made it possible to investigate the longitudinal and the transverse effects without remounting the sample.

EFFECTS OF GEOMETRY AND INHOMOGENEITY

The usual theory of the Hall effect and magnetoresistance assumes that the current flows in the length direction of the sample $(J=J_x)$, and hence transverse currents are zero $(J_y=0)$. In order to approximate these conditions experimentally the sample has to be long and narrow. Even then the above assumptions do not hold for the end regions of the specimen. The current probes usually cover the entire end surfaces; consequently the Hall field (E_y) is zero at x=0 and x=l(l= length of the specimen). The Hall voltage then reaches its full value at a certain distance from the ends. Calculations of E_y as a function of x for different length-to-width ratios have been made by Isenberg *et al.*²⁷ and by Volger.²⁸

A consequence of $J_y \neq 0$ is that the electrons will follow a longer path through the specimen, yielding a magnetoresistance which is larger than that given by



FIG. 1. Geometry of samples and probe configuration; (a) shape of samples 4-7; (b) shape of samples 1-3. the conventional expressions by an amount proportional to $\mu^2 H^2$ ($\mu = \text{mobility}$).^{5,21} This extra term might be small at low fields, but it will prevent saturation of the magnetoresistive effect at high magnetic fields.

It appears extremely difficult to suppress these "geometry" effects in a material, such as InSb, which has a high electron mobility. Even the smallest potential probes (indium solder, diameter 0.25 mm) will distort the current pattern and introduce nonzero transverse current components. As a result longitudinal effects will contribute to the transverse magnetoresistance and vice versa.

Another deviation from the conventional expressions is caused by inhomogeneities. Herring²⁹ has made an analysis of two particular cases. In the first case he assumes small, random, isotropic fluctuations of the impurity concentration n. It appears then that at low fields the conventional values of the longitudinal and transverse magnetoresistance have to be multiplied by certain constant factors. The other case is more serious. If the impurity concentration shows a gradient in the length direction of the sample [n=n(x)], the correction factors will be field-dependent. For high fields $(\Delta \rho / \rho_0)_{\text{transv}}$ will increase as H^2 , while the longitudinal effect might become negative in certain cases.

During this investigation we have made a detailed study of these geometry and inhomogeneity effects. A number of samples were measured with several different probe configurations. Results on dumbbell-shaped samples were compared with those from specimens having soldered contacts (see Fig. 1). Considerable attention was given to the inhomogeneity of the samples. We have been able to obtain reliable data for the transverse magnetoresistance at fields as large as 9 kilogauss. A proof of this statement is the very good agreement between the samples 3 and 4 both at 78°K (see Figs. 3 and 4) and at 1.7°K.

²⁶ We want to thank the Cryogenic Physics Section of the National Bureau of Standards for the use of an Arthur D. Little electromagnet.

 ²⁷ Isenberg, Russell, and Greene, Rev. Sci. Instr. 19, 685 (1948).
 ²⁸ J. Volger, Phys. Rev. 79, 1023 (1950).

²⁹ C. Herring (private communication). We want to thank Dr. Herring for showing us his calculations before publication.

An exact determination of the longitudinal effect is, however, much more difficult. On the basis of the conventional theory, the longitudinal effect should be zero assuming that the conduction band of InSb is spherically symmetric. Recently it has been shown that this is not true when quantization of the electron orbits has to be taken into account.¹⁸ It is clear, however, from the above considerations that at least part of the measured longitudinal magnetoresistance might be due to transverse contributions caused by geometry and inhomogeneity effects. A big question is how to determine the best experimental procedure for aligning the sample parallel to the magnetic field. Originally we considered the optimum position as that for which $(\Delta \rho / \rho)_{\text{long}}$ was a minimum. The angular dependence of the magnetoresistance-examples of which are given in Fig. 2-shows, however, that this criterion is not applicable. Another method is to find the position where the transverse voltage across the Hall probes becomes zero. This procedure has been used in the experiments presented in this paper. The position of the sample determined by this method differed slightly, however, from that of minimum $(\Delta \rho / \rho)$. Moreover, both the angular dependence and the position where the transverse voltage was zero depended on the magnetic field strength. The absolute magnitude of our values for $(\Delta \rho / \rho)_{\text{long}}$ are therefore not very significant. We believe, however, that our data give some indication of the order of magnitude of the longitudinal magnetoresistive effect and show clearly the oscillatory behavior at low temperatures.

Finally we mention another inhomogeneity effect. Measuring the emf between points A and B (see Fig. 1) in transverse magnetic field, we noticed that the results were not reproduced when the field was reversed. This can be explained if the sample is inhomogeneous; then the Hall fields at A and B would have different values. The difference between the Hall potentials at A and B

FIG. 2. Angular dependence of magnetoresistance, (a) for sample 4; (b) for sample 3. (ϕ is the angle between I and H.)





FIG. 3. Magnetoresistance at 78°K (sample 3).

will be added in one case and subtracted in a reverse field. The normal averaging procedure will eliminate this effect.³⁰

MAGNETORESISTANCE AND HALL EFFECT AT 78°K

The resistance of the samples listed in Table I was determined as a function of the magnetic field. Typical results obtained on four of these specimens are shown in Figs. 3, 4, 5, and 6. Samples 1 and 2 are degenerate at 78°K, all others are in the nondegenerate range.

We shall first discuss the transverse magnetoresistance. The low field region in which the magnetoresistance depends quadratically on the magnetic field $(\mu H < 10^8)$ is limited to fields less than 300-400 gauss. The measurements represented in Figs. 3-6 show indeed that the transverse effect is approximately proportional to H^2 for fields of 100-200 gauss, but give evidence that at higher fields saturation effects appear (2 to 3 kilogauss). At still higher fields a quadratic dependence on H is again approached.

It has been shown that the conventional theory is valid as long as $\hbar\omega \ll kT$. When the magnetic field becomes so strong, however, that $\hbar\omega \gg kT$, quantization

 $^{^{30}\,\}mathrm{We}$ are grateful to Dr. Glicksman for bringing this point to our attention.



FIG. 4. Magnetoresistance at 78°K (sample 4).

effects become important. The field dependence of the magnetoresistance is then in general different from that predicted by the conventional expressions. This is due to the fact that both the density of states and the collision time become functions of the magnetic field strength. Consequently the results will depend on the statistics (degenerate or nondegenerate) and the scattering mechanism (lattice or impurity scattering).

The quantization range can be divided into two regions. As long as $\zeta \gg \hbar \omega_0$, many quantized levels will be involved; when the field is so large that $\hbar\omega_0 \gg \zeta$, all electrons will be in the lowest quantum level ("quantum limit"). A number of investigators have studied the transverse and longitudinal magnetoresistance of metals and semiconductors under these conditions.^{18,19,31-36} We shall not discuss these papers at this point, but quote only the results which are relevant to our measurements. (For *n*-type InSb at 78°K, the condition $\hbar\omega > kT$ is fulfilled for H > 7500 gauss.) Titeica³¹ has analyzed the

³¹ S. Titeica, Ann. Physik (5) 22, 129 (1935).
 ³² J. Appel, Z. Naturforsch. 11a, 689, 892 (1956).
 ³³ A. Sommerfeld and B. W. Bartlett, Physik. Z. 36, 894 (1935).
 ³⁴ I. M. Lifschitz, J. Exptl. Theoret. Phys. (U.S.S.R.) 30, 814 (1956) [translation: Soviet Phys. JETP 3, 774 (1956)].
 ³⁵ P. N. Argyres, Westinghouse Research Report 6-94760-2-P9 (unpublished)



FIG. 5. Magnetoresistance at 78°K (sample 6).

transverse (and longitudinal) magnetoresistance of metals (degenerate statistics) for low fields ($\hbar\omega_0 \ll kT$) and for very high fields $(\hbar\omega_0 > \zeta)$ when the scattering is due to lattice vibrations. He predicts a quadratic field dependence in the low field range and a linear behavior in the quantum limit. Argyres³⁵ also studied the quantum limit for both degenerate and nondegenerate statistics (lattice scattering only). His results indicate that $(\Delta \rho / \rho)_{\text{transv}}$ should be a quadratic function of H in the former and a linear function of the field in the latter case. A paper of Lifschitz³⁴ seems to predict a quite different behavior $\left[(\Delta \rho / \rho)_{\text{transv}} \sim H^{-1} \right]$ for a metal with predominant impurity scattering.

All these theories contain a number of assumptions and approximations which make the results somewhat doubtful. At the same time we have stressed already the uncertainty of our experimental data at high magnetic field strength. Hence a comparison of theory and experiment does not seem very fruitful at this point. Our measurements should be extended to higher fields while the theory should be worked out for those cases where impurity scattering is predominant. It is quite possible that the accelerated increase of the transverse magnetoresistance between 3 and 9 kilogauss (see Figs. 3-6) is due to geometry or inhomogeneity effects.

⁽unpublished).

A. H. Kahn (to be published).

In the low-field region it should be possible to compare our results with theoretical predictions concerning the magnitude of the transverse and longitudinal effects. The expression for the transverse magnetoresistance has been calculated by Seitz³⁷:

$$\Delta \rho / \rho = a \mu^2 H^2. \tag{2}$$

The constant a equals 0.38×10^{-16} in the case of pure lattice scattering; it decreases sharply for small amounts of additional impurity scattering and increases again when scattering by ionized impurities becomes predominant. The factor a also depends on the magnetic field, or rather on the product μH . The formulas for different values of $\beta \left[= 6(\mu_{\text{latt}}/\mu_{\text{imp}}) \right]$ and $\gamma \left[= (9/16)\pi\mu^2 H^2 \right]$ have been worked out by several authors.³⁸ We have calculated the mobilities from our magnetoresistive data on four samples at fields of 100 or 200 gauss, using a curve of $a vs \beta$. The results are the following:

> sample 7: μ_{78} =3.6 ×10⁵ cm²/volt-sec, sample 6: $\mu_{78} = 4.1 \times 10^5 \text{ cm}^2/\text{volt-sec}$, sample 4: $\mu_{78} = 1.55 \times 10^5$ cm²/volt-sec, sample 3: $\mu_{78} = 1.5 \times 10^5 \text{ cm}^2/\text{volt-sec.}$



FIG. 6. Magnetoresistance at 78°K (sample 7).

³⁷ F. Seitz, Phys. Rev. **79**, 372 (1950). ³⁸ V. A. Johnson and W. J. Whitesell, Phys. Rev. **89**, 941 (1953); O. Madelung, Z. Naturforsch. 9a, 667 (1954); J. Appel, Z. Natur-



FIG. 7. Hall coefficient as a function of magnetic field strength at 78°K.

These figures have to be compared with the $(R\sigma)_{78}$ values of Table I. Considering the uncertainty with respect to the amount of ionized impurity scattering, the discrepancy in the mobilities is actually rather small. It seems that the magnitude of the magnetoresistance is consistent with an isotropic one-carrier model.

Turning now to the longitudinal magnetoresistance, we find that this effect is considerably smaller than the transverse effect for all samples. The field dependence is, however, vastly different for different specimens. Some of these effects might be due to quantization of the electron orbits. For instance, Argyres and Adams¹⁸ have shown that the longitudinal magnetoresistance can become negative in nondegenerate samples when impurity scattering is predominant.

The completely different behavior of the basically similar samples 3 and 4 (see Table I), however, strongly suggests that geometry and inhomogeneity effects are responsible for this discrepancy. A slight misalignment might introduce a small transverse contribution, wiping out a negative longitudinal effect, or a larger inhomogeneity in one sample than in the other might produce a negative effect at high fields.

In recent years several authors $^{\rm 39,\,40}$ have studied the relation between the magnetoresistive effects and the shape of the energy surfaces, assuming a number of anisotropic models. It seems rather senseless to compare our results with their predictions. One can state, however, that the very small longitudinal magnetoresistance

forsch. 9a, 167 (1954); R. K. Willardson and A. C. Beer, Bull. Am. Phys. Soc. Ser. II, 2, 142 (1957); J. H. Becker, Bull. Am. Phys. Soc. Ser. II, 1, 57 (1957).

³⁹ B. Abeles and S. Meiboom, Phys. Rev. 95, 31 (1954).

at very low fields (about an order of magnitude smaller than the transverse effect) is not in contradiction with the assumption of spherical energy surfaces.

Results on the Hall coefficient R_H as a function of magnetic field strength are plotted in Fig. 7 for the three samples 2, 4, and 6. At low fields—in this case of the order of a few hundred gauss— $R_H = (3\pi/8)(1/nec)$, (n = carrier concentration); for large fields the coefficient of 1/nec approaches the value 1.0. This explains the higher values of the Hall coefficient below 1000 gauss. The change of R_H between 1000 and 7000 gauss is at most 5%. The sinuous behavior in this range is as yet unexplained. It is perhaps no coincidence that the maximum occurs at about the same field $(H \approx 3000$ gauss) as the inflection point in the transverse magnetoresistance.

GALVANOMAGNETIC EFFECTS AT HELIUM TEMPERATURE

n-type InSb is completely degenerate at helium temperatures. The condition $\omega \tau > 1$ will be fulfilled for H>1000 gauss. The Fermi energy ζ_0 of relatively pure samples $(n=10^{15}-10^{16}/\text{cc})$ will be of the same order as $\hbar\omega$ for fields of a few thousand gauss. Hence we are in the quantization range for all magnetic fields above ~ 1000 gauss. Under these conditions theory predicts an oscillatory field dependence of the diamagnetic susceptibility (DeHaas-van Alphen effect) and of all transport phenomena. Such a behavior has been observed in Bi^{41,42} and in several other metals.

The theory of the quantization was first given by Landau¹⁷ and applied to the susceptibility (this treatment can be found in several textbooks^{16,43}). An analysis of the oscillatory transverse magnetoresistance observed in Bi at low temperatures was made by Davydov and Pomeranchuk.¹⁹ Argyres^{18,35} and Kahn³⁶ have recently derived expressions for the magnetoresistive effects assuming conditions which pertain to a degenerate extrinsic semiconductor like InSb.

A very brief description of the theory of these effects is as follows. In a high magnetic field the Fermi distribution of the electrons will change in such a way that the density becomes semidiscrete with discontinuities at energy values (Landau levels) corresponding to $(n+\frac{1}{2})\hbar\omega$ $(n=0,1,2,3\cdots)$. When the Fermi level is just above the nth Landau level, only a few electrons will be left in the *n*th state. The collision time of these particular electrons is much smaller than that of the bulk of the electrons in the lower magnetic state. The result is that the current will show maxima when the Fermi energy $\zeta_H = (n + \frac{1}{2})\hbar\omega$. The index H indicates that this is the value of ζ in the presence of a magnetic field. The difference between ζ_H and ζ_0 will be small in low

fields, but can be appreciable for large magnetic field strength. In the first place, the bottom of the Fermi sea will be raised by $\frac{1}{2}\hbar\omega$, and secondly, as H increases, the Fermi level will drop to the neighborhood of the next lower level whenever $\zeta_H \sim (n+\frac{1}{2})\hbar\omega$. Argures and Adams¹⁸ have shown that in the quantum limit the relation between the Fermi energy with and without field is given by

$$\zeta_H = (4/9) \left(\zeta_0 / \hbar \omega \right)^2 \zeta_0, \tag{3}$$

where ζ_H is measured relative to the $\frac{1}{2}\hbar\omega$ level. This formula is valid if no freeze-out (see below) takes place. Omitting the term $\frac{1}{2}\hbar\omega$, one finds³⁶ that the discontinuities will take place

for
$$n=1$$
, when $\zeta_0 = 1.31\hbar\omega$,
for $n=2$, when $\zeta_0 = 2.36\hbar\omega$,
for $n=3$, when $\zeta_0 = 3.38\hbar\omega$,
for $n=4$, when $\zeta_0 = 4.40\hbar\omega$, etc.

The periodicity is rapidly approached beyond the first level; it is clear, however, that a phase shift has to be taken into account.

The amplitude of the oscillations decreases rapidly with decreasing magnetic field strength. Landau¹⁷ has shown that this damping is expressed by a factor $1/\sinh(2\pi^2 kT/\beta^*H)$, where β^* is the effective double Bohr magneton $(=e\hbar/m^*c)$. If $2\pi^2 kT \gtrsim \beta^*H$, one can replace the hyperbolic sine by the exponential; the



FIG. 8. Magnetoresistance and Hall coefficient at 1.7°K (sample 6).

⁴¹ D. Shoenberg, Trans. Roy. Soc. (London) 245, 1 (1952). ⁴² L. Shubnikow and W. J. DeHaas, Commun. Kamerlingh Onnes Lab. Univ. Leiden No. 207d (1930); M. C. Steele and J. Babiskin, Phys. Rev. 98, 359 (1955). ⁴³ F. Seitz, *Modern Theory of Solids* (McGraw-Hill Book Com-pany, Inc., New York, 1940).

following expression then holds for the amplitude A^{44} :

$$A \sim TH^{\frac{1}{2}} \exp(-2\pi^2 k T / \beta^* H).$$
 (4)

Dingle⁴⁵ has pointed out that the damping is further influenced by collision broadening. He has shown that this can be taken into account if one replaces the ambient temperature T in expression (4) by a somewhat higher temperature (T+T'). It is clear then that one can determine β^* (and consequently the effective mass m_e^*) from the field and temperature dependence of the amplitude.

We turn now to a comparison of our experimental results with the theory presented above. The magnetoresistive effects have been measured on the samples 1-6 as a function of the magnetic field at 4.2°K and at 1.7 or 1.8°K. Some of the results are shown in Figs. 8, 9, and 10. The oscillatory behavior is quite obvious from these graphs; the amplitude decreases rapidly towards lower fields as predicted by the theory. It seems that the oscillations are superimposed on a curve which varies smoothly with H. Experiments with different probe configurations have shown that this part of the curve depends strongly on geometrical factors. The values of H for which the maxima and minima occur are, however, practically independent of the geometry



FIG. 9. Magnetoresistance and Hall coefficient at 1.7°K (sample 4).

⁴⁴ Reynolds, Hemstreet, Leinhardt, and Triantos, Phys. Rev. 96, 1203 (1954)





FIG. 10. Magnetoresistance and Hall coefficient at 1.7°K (sample 2).

except for the first minimum in the longitudinal effect. That sample 2 shows negative values of $\Delta \rho / \rho$ (both transverse and longitudinal) over the measured range of magnetic fields can perhaps be attributed to geometry or inhomogeneity effects.

Table II presents a comparison of calculated and observed values of $1/H_i$ and the period of oscillation $[\Delta(1/H)]_{ij}$. The Fermi energy ζ_0 has been computed from the expression $\zeta_0 = (\hbar^2/2m^*)(3n/8\pi)^{\frac{2}{3}}$. In doing so, we have assumed that the impurity band is completely merged with the conduction band; this is fully justifiable at these impurity concentrations if we accept the hydrogenic model for the donor states.⁴⁶ It is worth noting that the effective mass m^* is eliminated in calculating $1/H_i = a(\beta^*/\zeta_0)$ (a=1.31, 2.36, etc.; see above).

The data of Table II show a reasonable agreement between experiment and theory. The results are somewhat better for the transverse than for the longitudinal effect.

A value for the effective electron mass has been deduced from the magnetic field and temperature dependence of the amplitude. It follows from expression (4) that a plot of $\ln(AH^{-\frac{1}{2}})$ vs 1/H should yield a straight line with slope $-2\pi^2 k(T+T')/\beta^*$. T' and β^* can then be calculated from results of measurements at two temperatures. This procedure has been applied to the magnetoresistance data of samples 2 and 3. Considerable scatter of the experimental points greatly reduced the accuracy of the results; a value of $(0.010 \pm 0.004) m_0$ was obtained for the electron effective mass.

Results of Hall effect measurements at 1.7°K or 46 See, e.g., F. Stern and R. M. Talley, Phys. Rev. 100, 1638 (1955).

Sample	n	1/H: (10 ⁻⁴ ga min lo	$\left(\Delta \frac{1}{H}\right)_{i,i+1}$ uss ⁻¹) ng.	$1/H_i$ (10 ⁻⁴ gat min tra	$\begin{pmatrix} 1\\ \Delta_{\overline{H}} \end{pmatrix}_{i,i+1}$ $\underset{\text{nsv.}}{\overset{\text{uss}^{-1}}{\overset{\text{nsv.}}{\overset{nsv.}}{\overset{nsv.}}{\overset{nsv.}}}}}}}}}}$	$aeta^*/\zeta_0$ $(10^{-4} ext{ gauss}^{-1})$	$ \begin{pmatrix} 1 \\ \Delta_{\overline{H}} \end{pmatrix}_{i,i+1} $ $ \begin{pmatrix} 10^{-4} \text{ gauss}^{-1} \\ \text{calculated} \end{pmatrix} $
1	1 2 3	2.06 3.30	1.24			1.13 2.03 2.91	0.90 0.88
2	1 2 3 4 5	2.18 3.50 4.42 5.02(?)	1.32 0.92 0.60(?)	2.37 3.19	0.82	1.44 2.60 3.72 4.82 5.90	$1.16 \\ 1.12 \\ 1.10 \\ 1.08$
3	1 2 3 4	1.56 3.69 5.72	2.13 2.03	1.98 3.73 5.69 7.41	1.75 1.96 1.72	$2.33 \\ 4.20 \\ 6.02 \\ 7.82$	1.87 1.82 1.80
4	1 2 3 4	1.47 3.53 5.56 7.52	2.06 2.03 1.96	1.82 3.55 5.56 7.52	1.73 2.01 1.96	2.38 4.30 6.16 8.00	$1.92 \\ 1.86 \\ 1.84$
5	1 2 3	2.35 5.50	3.15	2.97 5.72	2.75	3.37 6.08 8.70	2.71 2.62
6	1 2	2.63 6.45	3.82	1.67(?) 3.33 6.67	3.34	4.14 7.46	3.32

TABLE II. Minima and periods of oscillations in the transverse and longitudinal magnetoresistance at T=1.7 or 1.8° K (experimental). The last two columns show the theoretical values based on the carrier concentration of each sample.

1.8°K are shown at the bottom of Figs. 8, 9, and 10. The general shape of the curve is surprisingly similar to that at 78°K. In some of the samples one can detect a somewhat oscillatory behavior; a correlation with the oscillations observed in the magnetoresistive effects is, however, not obvious. There are a few noteworthy coincidences. The high-field minimum (H=6500 gauss)for sample 4; H=3800 gauss for sample 6) occurs at precisely the same field as that of the longitudinal magnetoresistance. There is a slight suggestion of a dip at 4 and a hump at 3 kilogauss in the curve of sample 4. One might suggest then that for this sample and for sample 6, the maxima (minima) of the Hall coefficient correspond with the minima (maxima) of the transverse magnetoresistance. Although such a correlation seems logical, a full comparison of Hall effect and magnetoresistive effects should await further experimental and theoretical work.

"FREEZE-OUT" AND RE-EXCITATION

The energy states of impurity atoms in semiconductors can often be treated on the basis of a hydrogenic model.^{46,47} A donor with one electron then has energy levels which are similar to those of the hydrogen atom modified by the effective mass m^* and the dielectric constant κ . If m^* is small and κ is large, the wave functions are spread over a considerable distance, corresponding to a large effective Bohr radius $r_{\rm eff}$. In the case of donors in InSb, $r_{\rm eff} \approx 600$ A.

Yafet, Keyes, and Adams²³ have studied the effects of very large magnetic fields on such an impurity semiconductor at low temperatures. They have shown that if the magnetic field strength is much larger than the Coulomb forces, the wave functions will change considerably. As a result the effective Bohr radius will decrease (although to a different extent perpendicular and parallel to the field) and the ionization energy will increase. Consequently, carriers will drop back from the conduction band into the bound states and a freezeout occurs.

The above analysis was made assuming a separation of impurity band and conduction band in zero field. Keyes and Sladek²⁴ have measured the conductivity of InSb samples with 10¹⁴ impurities per cc, for which this assumption is valid, and have confirmed the prediction of a freeze-out effect.

In our case the samples 6 and 7 have slightly more than 10^{15} impurities per cc. A calculation of the width of the impurity band on the basis of the hydrogenic model shows that this band will overlap the conduction band to some extent. At high magnetic fields, however, the activation energy will become positive and a redistribution of electrons will take place with a preference for the bound states. It seems, therefore, that we can attribute the sudden rise of the Hall coefficient above 11 000 gauss (see Fig. 11) to the same freeze-out effect.

A confirmation of this explanation can be found in the experimental results of Fig. 12. At 22 000 gauss the electric field across sample 7 was increased from several millivolt per cm to 200 mv/cm. The Hall coefficient decreased by about 14%, indicating a re-excitation of carriers into the conduction band.

We have also investigated this effect in a rather

⁴⁷ W. Baltensperger, Phil. Mag. 44, 1355 (1953).



FIG. 11. Hall coefficient as a function of magnetic field strength at 4.2°K.

impure sample (8) $(1.55 \times 10^{16} \text{ electrons/cc})$. This specimen does not show the freeze-out effect or the re-excitation (see Figs. 11 and 12). The overlap is very large in this case and the magnetic field does not influence the activation energy enough to cause separation of impurity and conduction band.

SUMMARY

Measurements of the magnetoresistive effects (both transverse and longitudinal) and the Hall coefficient at 78°K and at helium temperatures have led to the following conclusions:

1. The magnitude of the transverse and longitudinal magnetoresistance in the region of low magnetic fields is consistent with an isotropic, one-carrier model.

2. In semiconductors with a large carrier mobility $(\sim 10^5 \text{ cm}^2/\text{volt-sec})$, the high-field region is reached at fields of a few thousand gauss. The magnetoresistive effects are extremely sensitive to the scattering mechanism and the statistics, as well as to the geometry and inhomogeneity of the sample.

3. The galvanomagnetic effects in InSb with electron concentrations between 10^{15} and 10^{16} electrons/cc show an oscillatory behavior at helium temperatures. The



FIG. 12. Hall coefficient as a function of electric field strength.

period of oscillation is in reasonable agreement with theoretical predictions. The field dependence of the amplitude of oscillation yields an effective electron mass of $0.01 \ m_0$.

4. The field dependence of the Hall effect agrees generally with theory. The detailed variations are, however, as yet unexplained.

5. Donor states will be strongly affected by large magnetic fields if the effective mass of the conduction band is small. Fields of more than 10 000 gauss are able to produce a condensation of electrons in *n*-type InSb at helium temperatures ("freeze-out"). These electrons can be re-excited by large electric fields of the order of 0.1-1.0 volt/cm.

ACKNOWLEDGMENTS

The authors wish to thank Dr. A. H. Kahn for many helpful discussions. They are indebted to Mr. D. E. Roberts who prepared the pure, single crystals used in this investigation.