

Phonon Drag in *n*-Type InSb\*

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The electronic and phonon-drag contributions to the thermoelectric power of *n*-InSb have been investigated as a function of temperature and magnetic field. It is found that in zero field, the phonon-drag contribution is negligible except for the temperature region between 10 and 30°K. In the quantum limit, at high field and low temperatures, the phonon-drag contribution becomes quite large. In fact, it dominates the electronic contribution, which increases only logarithmically above its classical saturation value.

## I. INTRODUCTION

A LARGE contribution to the Seebeck coefficient due to a nonequilibrium phonon distribution<sup>1</sup> was first identified experimentally in *n*-Ge by Frederikse<sup>2</sup> and Geballe.<sup>3</sup> The phonon-drag effect,<sup>4</sup> as it is called has since been observed in many other semiconductors<sup>5</sup> as well as metals.<sup>6</sup> In semiconductors, the low-energy acoustic phonons tend to become more closely coupled with the conduction electrons as the temperature is lowered. The electrons also are scattered mostly by these phonons, provided the crystal is sufficiently pure. This means that the phonon distribution tends to follow any disturbance in the electron distribution. For example, an external electric field affects not only the electron distribution but also the phonon distribution. This gives rise to the additional term in the thermoelectric coefficients. Herring has given an extensive review of the theory<sup>7</sup> of the phonon-drag effect.

It is known that while the phonon-drag makes an appreciable contribution in case of *p*-type InSb samples, it is small or negligible in *n*-type samples.<sup>8</sup> The reason for this is that in *n*-type InSb, the scattering of electrons

on ionized impurities dominates the electron-phonon scattering by several orders of magnitude at temperatures where a large phonon drag is to be expected. The small effective mass of electrons emphasizes this effect.

We have measured the thermoelectric power  $Q$  for several samples of high-purity *n*-type InSb in the temperature range from 7 to 80°K in the presence of transverse magnetic fields up to 100 kG. It is found that in the classical region of magnetic fields,  $\hbar\omega/kT \ll 1$ , the phonon-drag contribution is small and lies within the uncertainty in measurements. In high magnetic fields, on the other hand, when the quantization of the cyclotron orbits alters significantly the behavior of transport properties, the phonon-drag contribution becomes measurable and even dominant over the electron contribution at the lower end of the temperature range. Here  $\omega$  is the cyclotron frequency and the other symbols have their usual meaning. Measurements of the dependence of  $Q$  on the cross section of the sample similar to those which demonstrated the phonon drag in Ge,<sup>9</sup> further confirm this conclusion.

As has been pointed out,<sup>7,10,11</sup> the analysis of the phonon-drag effect can provide valuable information about the electron-phonon and phonon-phonon scattering. In InSb the mobility is effectively determined by electron scattering from optical phonons<sup>12</sup> near room temperature and from ionized impurities at lower temperatures, scattering by acoustic phonons being weak throughout the temperature range. The present experiments were done to obtain electron coupling to acoustic phonons and the relaxation time of long-wavelength phonons. To obtain quantitative information the magnitude of the phonon-drag contribution to the thermoelectric power must be established precisely which, in turn, demands a sufficiently accurate knowledge of the electronic contribution in high magnetic fields. Recently there has been some confusion about the behavior of

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<sup>1</sup> A preliminary account of this work was reported by S. M. Puri, *Bull. Am. Phys. Soc.* **8**, 219 (1963).

<sup>2</sup> H. P. R. Frederikse, *Phys. Rev.* **91**, 491 (1953); **92**, 248 (1953).

<sup>3</sup> T. H. Geballe, *Phys. Rev.* **92**, 857 (1953).

<sup>4</sup> L. E. Gurevich, *Zh. Eksperim. i Teor. Fiz.* **16**, 193, 416 (1946).

<sup>5</sup> E. Mooser and W. Woods, *Phys. Rev.* **97**, 1721 (1954); R. Monsfield and S. A. Salam, *Proc. Phys. Soc. (London)* **B66**, 377 (1953); I. N. Tinchenko and S. S. Shalyt, *Fiz. Tverd. Tela* **1**, 1302 (1959); **4**, 934 (1962) [English transl.: *Soviet Phys.—Solid State* **1**, 1192 (1959); **4**, 685 (1962)]; see also Ref. 7.

<sup>6</sup> W. B. Pearson, *Fiz. Tverd. Tela* **3**, 1411 (1961) [English transl.: *Soviet Phys.—Solid State* **3**, 1024 (1961)]; D. K. D. MacDonald, W. B. Pearson and I. M. Templeton, *Proc. Roy. Soc. (London)* **A248**, 107 (1958).

<sup>7</sup> C. Herring, in *Halbleiter und Phosphore*, edited by M. Schon and H. Welker (Fredrick Veiweg und Sohn, Braunschweig, 1958), p. 184.

<sup>8</sup> H. P. R. Frederikse and E. Mielczarck, *Phys. Rev.* **99**, 1889 (1955); T. H. Geballe, *Bull. Am. Phys. Soc.* **2**, 56 (1957).

<sup>9</sup> T. H. Geballe and G. W. Hull, *Phys. Rev.* **94**, 1134 (1954).

<sup>10</sup> C. Herring, T. H. Geballe, and J. E. Kunzler, *Phys. Rev.* **111**, 36 (1958).

<sup>11</sup> D. K. C. MacDonald, *Thermoelectricity* (John Wiley & Sons, Inc., New York, 1962).

<sup>12</sup> H. Ehrenreich, *Phys. Chem. Solids* **2**, 131 (1957); **9**, 129 (1959).

the electronic contribution in the quantum limit.<sup>1,13</sup> It is our purpose in this paper to clarify this point and present some supporting experimental evidence. A complete quantitative analysis of the data requires taking into account the complicated, though well established band structure of InSb and also the effects of quantization of Landau levels on the electron-phonon scattering. This program is being carried out and the results will be published elsewhere.<sup>14</sup>

In Sec. II, we review briefly the expected behavior of the electronic contribution to  $Q$  in high magnetic fields. It will be seen that in contrast to the zero-field case, the mechanism of electron scattering does not influence the value of electronic contribution to  $Q$  in high transverse magnetic fields which can, therefore, be calculated precisely when the band structure is known. The high-field measurements thus can provide a more accurate estimate of phonon-drag  $Q$  than the zero-field results. In Sec. IV we present the experimental results and give a brief discussion of these.

We shall confine our discussion to situations where the electronic and the phonon-drag contributions to the thermoelectric phenomena are independent of each other and can be discussed separately.<sup>15</sup> Physically, this demands that the shift in the phonon distribution does not affect significantly the electron-phonon scattering, i.e., the distortion of the phonon spectrum just adds an additional independent term to the Peltier heat flux, leaving unaltered the electronic contribution. Experimentally, this requires the charge carrier concentration to be small so that the "saturation effect" of Herring<sup>7</sup> can be neglected. These conditions are satisfied by the present experiments.

## II. ELECTRONIC PART OF $Q$

The effect of magnetic field on the electronic part of  $Q$  has been discussed by several authors.<sup>16</sup> Most of these treatments adopt the Boltzmann equation or some variation of the classical transport theory. As such, the results obtained therein are valid only as long as the spacing of the cyclotron levels (Landau levels) is small compared to the average kinetic energy of the electron. For semiconductors this requires  $\hbar\omega/kT \ll 1$ . A quantum-mechanical treatment of thermomagnetic effects has been given by Anselm and Askerov,<sup>13</sup> taking into account the quantization of the electron energy levels. They find that in the case of extreme quantum limit,  $\hbar\omega/kT \gg 1$ ,  $Q_e$  varies linearly with the magnetic field and is independent of electron scattering. As far as we can see, the first of these conclusions is not correct. The source of error seems to stem from the incorrect use of

Onsager's reciprocal relations.<sup>17</sup> As a result a term is missed which has important consequences in the transverse field case in the quantum limit where, in the first approximation, the velocity parallel to the electric field is zero. In longitudinal fields or in the classical region, this correction is not very important. We shall not follow the rigorous approach of the density-matrix technique used in Ref. 13 but obtain the results from simple but sound arguments. The same results are obtained from the more rigorous approach if the procedure indicated in Ref. 18 is followed.<sup>19</sup>

The Peltier coefficient  $\pi$ , is given, in general, in terms of the electrochemical potential  $\mu$ , the band-edge energy  $\epsilon_b$ , and the energy of the electron  $\epsilon$ , measured from the band edge.

$$e\pi_e = |\epsilon_b - \mu| + \langle v\epsilon \rangle / \langle v \rangle, \quad (1)$$

where  $v$  is the velocity of the electron and the angular brackets denote average over the steady-state distribution. In quantum transport theories, a similar relation can be written down but the quantities inside the angular brackets must be considered as operators and the averages are replaced by the trace of the product of the density matrix with the respective operator. Equation (1) is nothing more than the definition of  $\pi$  using the definition of heat flux given by Callen.<sup>18</sup> The Seebeck coefficient can be written from the reciprocal relation

$$Q_e = - \left\{ \frac{k}{e} \left| \frac{\epsilon_b - \mu}{kT} \right| + \frac{1}{kT} \frac{\langle v\epsilon \rangle}{\langle v \rangle} \right\}. \quad (2)$$

The first term is determined by the band structure alone while the second term depends on electron scattering as well. In the classical region of magnetic fields,  $|\epsilon_b - \mu|$  is not affected by the magnetic field and the total change in  $Q_e$  arises from the variation of the second term. In the high field approximation, the electron distribution approaches a limiting value.<sup>20</sup> The second term does likewise, and therefore  $Q_e$  does also. If the magnetic field is transverse to the temperature gradient, the

<sup>13</sup> In order to clarify this point, first consider the definition of heat flux which is used to obtain the Peltier coefficient. Callen (Ref. 18) has shown that, in principle, there is a certain flexibility, and slightly different definitions of the thermoelectric coefficients can be given. The Onsager relations will be valid within each set of coefficients defined in a consistent manner. He has also shown that the quantities which are experimentally significant are obtained if in writing the reciprocal relations (a) the generalized forces are derived from the electrochemical potential  $\mu$  rather than the chemical potential, and (b) the heat flux is defined by the difference of the total energy current density  $W$  (in the notation of Ref. 16, the total energy current density is  $U$ ) and the potential energy current density  $\mu J$ . There is, of course, no ambiguity in the definition of the electric current density  $eJ$  with  $e$  as the electronic charge. In this respect, the set of equations (1.3) and (1.4) of Anselm and Askerov (Ref. 13) is inconsistent with the set (1.1) and (1.2), and therefore Eq. (1.6) of their paper is not valid. Equation (1.6) will be valid if from the left-hand sides of Eqs. (1.3) and (1.4) we subtract a term  $\varphi J_x$  and  $\varphi J_y$ , respectively, in the notation of Ref. 13.

<sup>18</sup> H. B. Callen, Phys. Rev. **85**, 16 (1952); **73**, 1349 (1948).

<sup>19</sup> S. M. Puri, Ph.D. thesis, Columbia University, New York, 1964 (unpublished).

<sup>20</sup> J. A. Swanson, Phys. Rev. **99**, 1799 (1955).

<sup>13</sup> A. I. Anselm and B. M. Askerov, Fiz. Tverd. Tela **3**, 3668 (1961) [English transl.: Soviet Phys.—Solid State **3**, 2665 (1962)].

<sup>14</sup> S. M. Puri (to be published).

<sup>15</sup> C. Herring, Phys. Rev. **96**, 1163 (1954); see also Ref. 7.

<sup>16</sup> M. Kohler, Ann. Physik **6**, 18 (1949); M. Rodot, Ann. Phys. (Paris) **5**, 1085 (1960); G. I. Guseva and I. M. Tsidil'kovskii, Fiz. Tverd. Tela **4**, 2490 (1962) [English transl.: Soviet Phys.—Solid State **4**, 1824 (1963)].

limiting value of the second term is  $\frac{5}{2}$  irrespective of the electron-scattering mechanisms.<sup>10</sup> In longitudinal fields, the saturation value is the same as if the magnetic field were absent.<sup>10</sup> Therefore in transverse magnetic fields, the saturation value of  $Q_e$  can be calculated accurately from a knowledge of the electron concentration and the band structure.

In the quantum region  $\epsilon_b$  is raised by an amount  $\frac{1}{2}\hbar\omega$ , which is the energy of the lowest Landau level. If nothing else is changed and the density of charge carriers stays constant,  $\mu$  will also be raised by exactly  $\frac{1}{2}\hbar\omega$ , and thus  $|\epsilon_b - \mu|$  will remain constant. But due to the degeneracy of the quantized levels, the density of states near the band edge is increased. If all or most of the electrons are in the lowest Landau level, the altered density of states increases  $|\epsilon_b - \mu|$  as is necessary to keep the total number of occupied levels the same. It is shown in the Appendix that for a fixed value of carrier concentration,  $|\epsilon_b - \mu|$  increases under the above conditions as  $kT \ln(\hbar\omega/kT)$ , provided the spin splitting of the Landau levels is small compared to  $kT$ . If the charge carrier concentration also changes with the field, we must add a term  $kT \ln(n_0/n_B)$  where  $n_B$  and  $n_0$  are, respectively, the carrier concentrations in field  $B$  and in zero field. So far as the second term of Eq. (2) is concerned, we note that in transverse fields, the Hall velocity  $v_y$  which is perpendicular to the external fields is independent of electron scattering and much greater than the velocity parallel to the electric field  $v_x$ . To a first approximation, the effect of collisions can be neglected, in which case  $v_x = 0$  and  $v_y = cE/B$ . The second term is then reduced to

$$\langle \epsilon \rangle = \frac{1}{2}\hbar\omega \coth(\hbar\omega/2kT) - kT \ln\{2 \sinh(\hbar\omega/2kT)\} + \frac{1}{2}kT + kT.$$

The first and second terms together represent the contribution due to electron motion in the  $x-y$  plane which is quantized. The second term is subtracted because of a shift in the zero of energy. The third term is due to electron motion parallel to the magnetic field. The last term is due to the pressure (see Appendix) exerted by the electron gas which is independent of the electron-energy spectrum and therefore remains unaltered by the quantization. Therefore, in the transverse magnetic field,

$$Q_e(B) - Q_e(\text{saturation}) = -\frac{kT}{e} \left[ \ln\left(\frac{\hbar\omega}{kT}\right) + \ln\frac{n_0}{n_B} + \frac{\hbar\omega}{2kT} \coth\frac{\hbar\omega}{2kT} - \ln\left\{2 \sinh\left(\frac{\hbar\omega}{2kT}\right)\right\} - 1 \right]. \quad (3)$$

In the extreme quantum limit, the motion in the  $x-y$  plane is completely frozen and does not contribute to  $\langle \epsilon \rangle$ . Figure 1 demonstrates qualitatively the effect of magnetic field on the Peltier coefficient. The effect of collisions is to add terms of the order of  $(v_x/v_y)^2$  to (3). This correction is very small in practical cases. In the extreme quantum limit, the first term of Eq. (3) domi-

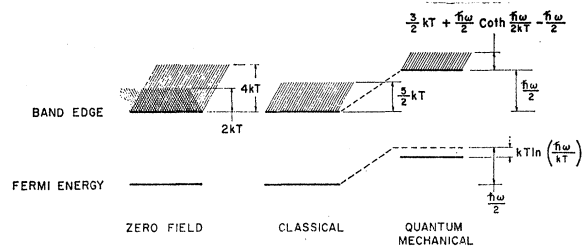


Fig. 1. Effect of transverse magnetic field on the different contributions to the Peltier coefficient. On the extreme left is the case corresponding to zero field and on the extreme right, the situation corresponding to the extreme quantum limit. The center portion represents the picture for high field in the classical region.

nates and  $Q_e$  increases as  $k/e \ln(\hbar\omega/kT)$ . In longitudinal fields, the variation of the second term of Eq. (2) will be more complicated but the contribution from it to  $\Delta Q_e$  is small in any case. In the above discussion we have neglected the splitting of the Landau levels due to spin.

### III. PHONON-DRAG PART OF $Q$

Herring<sup>7,15</sup> has derived expressions for the phonon-drag contribution  $Q_p$  to the Seebeck coefficient. Neglecting the saturation effect,  $Q_p$  is given by the following expressions for a semiconductor with a nondegenerate electron distribution:

$$Q_p = (k/e)\gamma(ms^2/kT)\bar{\tau}_q/\langle\tau_e\rangle \quad (4a)$$

$$= (1/T)\gamma nes^2\bar{\tau}_q\rho. \quad (4b)$$

In these expressions  $m$  is the effective mass of the electron,  $s$  is the sound velocity in the crystal,  $\tau_e$  is the total relaxation time of electrons,  $n$  is the electron concentration and  $\rho$  is the electrical resistivity of the crystal;  $\bar{\tau}_q$  is some average relaxation time of phonons taking part in phonon drag, and  $\gamma$ , a number lying between zero and one, is a measure of the crystal momentum delivered to acoustic phonons compared to the total crystal momentum dissipated by the electrons. For pure acoustic scattering  $\gamma = 1$ . In general,  $\gamma$  is given, in the relaxation time approximation,<sup>15</sup> by

$$\gamma = \frac{\langle K/\tau_{ep} \rangle}{\langle K/\tau_e \rangle},$$

where  $K$  is the electron wave vector, and  $\tau_{ep}$  is the electron relaxation time due to scattering on acoustic phonons. The angular brackets denote average over steady distribution of electrons.

In longitudinal magnetic fields, because the classical Boltzmann-type equation remains valid<sup>21</sup> even in the quantum region, Eq. (4b) can be used without any modifications. Even in transverse fields, Eq. (4b) is valid<sup>19,22,23</sup> in the quantum region, provided  $\gamma$  is equated

<sup>21</sup> P. N. Argyres and E. N. Adams, Phys. Rev. **104**, 900 (1958).

<sup>22</sup> L. E. Gurevich and G. M. Nedlin, Fiz. Tverd. Tela **3**, 2779 (1961) [English transl.: Soviet Phys.—Solid State **3**, 2029 (1962)].

<sup>23</sup> T. Ohta, J. Phys. Soc. Japan **18**, 909 (1963); **19**, 769 (1964).

to unity and  $\rho$  is replaced by  $\rho_{xx}^p$ ;  $\rho_{xx}^p$  is the transverse resistivity caused by scattering on acoustic phonons only. A detailed justification for using Herring's results in the quantum region will be given in our next paper<sup>14</sup> where we shall also give quantitative formulas for a complete analysis of the experimental results. We shall be content here with these skeleton formulas which will suffice for the qualitative discussion of  $Q_p$  presented here.

#### IV. EXPERIMENT AND DISCUSSION

The cryostat used for making these measurements is described in Ref. 19. It is similar in design to that used previously.<sup>10</sup> Normally only the change in the Seebeck voltage is measured as a function of field. In order to translate voltage into the ratio of the Seebeck coefficient in field  $B$  to that in zero field  $Q_B/Q_0$ , it is necessary that, for constant-heat input to the sample, the temperature gradient as well as the mean temperature of the sample are not affected by the presence of the magnetic field. This is reasonable to assume because the electronic contribution to thermal conductivity is negligible below 100°K in InSb.<sup>24</sup> Also care was taken to insure that there were no appreciable magnetothermal resistances in the thermal-links from the sample to the bath. Various direct and indirect checks substantiate the validity of this assumption within 1%. The absolute magnitude of  $Q_0$  was measured using Ge resistance thermometers or copper/constantan thermocouples.

The conventional bridge-shaped samples were cut from the single-crystal specimens such that the magnetic field is along the [111] when it is applied in the transverse direction. The Hall mobility of the samples has a maximum value near 60°K of 5 to  $7 \times 10^5$  cm<sup>2</sup>/V sec. A list of samples together with their carrier concentration determined from the Hall effect data is given in Table I.

Figure 2 shows the magnitude of  $Q_0$  for sample 584 as a function of temperature between 5 and 80°K. The value of  $Q_0$  decreases monotonically with temperature. Similar behavior led Frederikse and Mielczarek<sup>8</sup> to conclude that there is no phonon drag in  $n$ -type InSb. In this figure we have also shown the value of  $Q_e$  calcu-

TABLE I. Samples of  $n$ -InSb used in the experiment.

Sample	Carrier density at 77°K
584	$4.0 \times 10^{14}$ /cc
801	$1.0 \times 10^{14}$ /cc
808	$3.0 \times 10^{18}$ /cc
Sample A <sup>a</sup>	
Sample B <sup>a</sup>	

<sup>a</sup> Samples A and B were obtained from the adjoining parts of a slice cut from the same crystal as 808. The carrier concentration of A and B was not measured but is expected to be roughly the same as 808.

<sup>24</sup> D. Kh. Amirkhanov and R. I. Bashirov, *Fiz. Tverd. Tela* **2**, 1597 (1960) [English transl.: *Soviet Phys.—Solid State* **2**, 1447 (1960)].

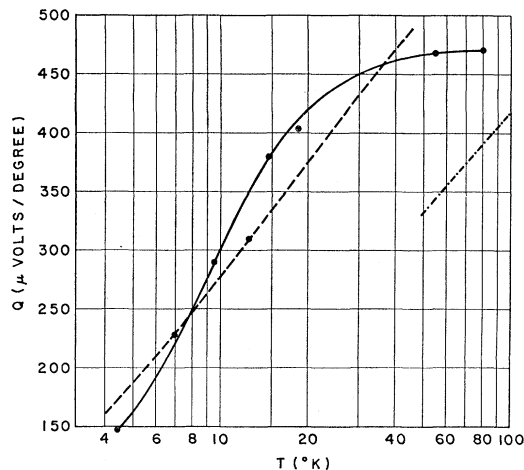


FIG. 2. Thermoelectric power of an  $n$ -type InSb sample (584) with carrier density  $4 \times 10^{14}$ /cc. The solid line represents a smooth curve through the experimental points marked by  $\bullet$ . The other two curves represent the theoretical values of the electronic contribution for different assumptions about the electron scattering: (i) dashed curve (---) for scattering on ionized impurities; (ii) dot-dash curve (- · -) for scattering on acoustic phonons.

lated using Eq. (2). The contribution from the second term of (2), which is sensitive to electron scattering, is relatively large in the case of  $n$ -type InSb because the small effective mass of the electrons makes the first term small. In the figure we have plotted the computed values assuming (i) scattering on ionized impurities, (ii) scattering by acoustic phonons. The assumption of a relaxation time is made in both cases and an effective mass of  $0.013 \times 9.1 \times 10^{-28}$  g is used. The experimental values agree fairly well with the first curve. At the upper end of the temperature scale, a small admixture of acoustic scattering improves the agreement between the calculated and the measured values. At temperatures below 10°K, as the sample becomes degenerate, electron screening of the charged impurity potential, which has been neglected, would, if included, decrease the calculated value. The difference in the observed and calculated values between 10 and 30°K which amounts to less than 50  $\mu$ V/deg, is larger than the estimated uncertainty of the measurements. This implies that a phonon-drag contribution of less than 50  $\mu$ V/deg exists between 10 and 30°K and is negligible outside this temperature range. This result is not totally unexpected, since  $Q_p$  can be estimated by comparison with Ge by a correspondence argument. The observed electron mobility in InSb has a value of  $\sim 2 \times 10^5$  cm<sup>2</sup>/V sec at 20°K. Extrapolating Ehrenreich's results,<sup>12</sup> the mobility due to scattering on acoustic phonons alone has a value  $\sim 10^8$  units at 20°K. This gives, as a crude estimate, the value of  $2 \times 10^{-3}$  for  $\gamma$  to be used in Eq. (4a). At the same temperature,  $Q_p$  for  $n$ -Ge has a value of  $\sim 10$  000  $\mu$ V/deg, with a  $\gamma \approx 1$  and an electron mobility of  $\sim 4 \times 10^5$  units. Upon using this data and (4a), one

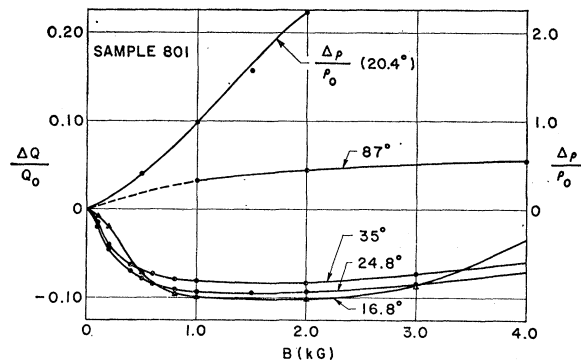


FIG. 3. Relative change of thermoelectric power  $\Delta Q/Q_0$  for sample 801 in transverse magnetic field in the classical region. The negative value of  $\Delta Q/Q_0$  observed near the liquid-hydrogen temperature range indicates that the phonon-drag contribution is small. The relative magnetoresistance at 20.4°K is also shown (vertical scale on the right).

obtains for the ratio of  $Q_p$  in InSb and in  $n$ -Ge as

$$Q_p^{\text{InSb}}/Q_p^{\text{Ge}} \approx (\gamma^{\text{InSb}}/\gamma^{\text{Ge}})(\mu_e^{\text{Ge}}/\mu_e^{\text{InSb}}) \approx 4 \times 10^{-3},$$

or a value of 40  $\mu\text{V}/\text{deg}$  for  $Q_p$  in InSb. The estimate is much too crude to take the exact agreement with experiment seriously; nevertheless, the order of magnitude should be correct. The above argument depends upon the  $\bar{\tau}(q)$  for Ge and InSb being similar which is reasonable as judged from thermal-conductivity data.

The above results find further confirmation from the change in  $Q$  in the classical region of magnetic fields. The data for sample 801 are shown in Fig. 3. The expected change in the electronic part  $\Delta Q_e$  is an increase with a saturation value of 43  $\mu\text{V}/\text{deg}$  for pure acoustic scattering and is a decrease with a saturation value of 129  $\mu\text{V}/\text{deg}$  for scattering on ionized impurities. On the other hand, the change in the phonon-drag part,  $\Delta Q_p/Q_p(0)$  is always positive and  $\approx \Delta\rho/\rho_0^{10}$ ;  $\Delta\rho/\rho_0$  is the magnetoresistance ratio. The observed  $Q$  increases by 20  $\mu\text{V}/\text{deg}$  at 87°K and decreases by 40  $\mu\text{V}/\text{deg}$  near liquid-hydrogen temperatures. It is fairly reasonable to assume that the result at 87°K can be explained by an admixture of impurity scattering and acoustic scattering. Assuming, on the other hand, that acoustic scattering is completely negligible at 35°K and below, from the observed  $\Delta Q = \Delta Q_e + \Delta Q_p$ , we get a value of 90  $\mu\text{V}/\text{deg}$  for  $\Delta Q_p$ . The presence of acoustic scattering lowers this estimate. Accepting the experimental results on  $\Delta\rho/\rho_0$ , also plotted in the figure, at its face value,<sup>25</sup> we get  $Q_p(0) \sim 45 \mu\text{V}/\text{deg}$ . This value is in agreement with our previous conclusion at 20°K but is somewhat higher than expected for 35°K.

Next we turn our attention to  $\Delta Q$  in the quantum region. From the point of view of comparison with

<sup>25</sup> In contrast to  $\Delta Q$  and, in contradiction to theory, the magnetoresistance does not saturate in the classical region. The reason for this is not quite clear; it may be due to inhomogeneities in the carrier concentration of the sample. See Ref. 19 and C. Herring, *J. Appl. Phys.* **31**, 1939 (1961); R. T. Bate and A. C. Beer, *ibid.* **32**, 800 (1961); **32**, 806 (1961).

theory, it is better to measure the change in  $Q$  from the classical saturation value where the electronic contribution is insensitive to electron scattering. We define  $\Delta Q_c = Q_B - Q_{\text{saturation}}$ .  $Q_{\text{saturation}}$  is the observed saturation value in the classical region.  $\Delta Q_c$  has contributions from the electron-diffusion as well as phonon drag which we want to separate.

In the electronic contribution to  $\Delta Q_c$ , the magnetic field and the temperature always occur together as the factor  $(\hbar\omega/kT)$ . Therefore, if only the electronic contribution to  $\Delta Q_c$  were important, the measurements at different temperatures will scale as  $B/T$ . The phonon-drag contribution has the extra temperature dependence due to  $\tau_q$  and will not scale as  $B/T$ . In Fig. 4, we have plotted the experimental values of  $\Delta Q_c$  as a function of  $B/T$  at three different temperatures. The three curves almost exactly coincide for  $B/T \leq 0.6 \text{ kG}/\text{deg}$ , indicating that the phonon drag is negligible. For higher values of the magnetic field, the three curves diverge from each other, showing that a measurable amount of  $Q_p$  is present. In Fig. 5 we compare the experimental values of  $\Delta Q$  at 82°K with the calculated electronic contribution. Curve II shows the value of  $\Delta Q_e$  calculated from Eq. (3.7) of Ref. 13. The calculated values in this case are much higher than the observed values of  $\Delta Q$  and it is not possible to explain this difference. The values calculated on the basis of Eq. (3) (Curve I) agree well with the experimental results below  $B/T = 0.6 \text{ kG}/\text{deg}$  which indicates there is little or no phonon-drag contribution up to this value. For higher values of  $B/T$ , the experimental values are higher than the calculated ones and the difference may be regarded as the phonon-drag contribution. We have used a value of  $0.013 \times 9.1 \times 10^{-28} \text{ g}$  for the effective mass of electrons in these calculations. However, we have not taken into account the effect of the spin splitting of the Landau levels. The effect of spin splitting is to raise the electrochemical potential, and

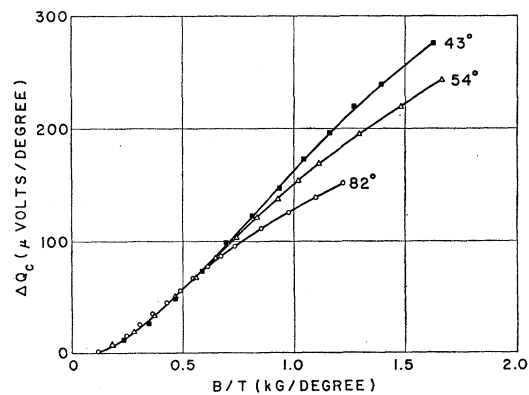


FIG. 4. Change of thermoelectric power in transverse magnetic field in the quantum region. On the y axis is plotted the increase in  $Q$  over the saturation value observed in the classical region (Fig. 3). The overlapping of curves for small values of  $B/T$  indicates negligible contribution from phonon drag. The divergence of the three curves for large  $B/T$  indicates that a measurable amount of phonon-drag contribution is realized.

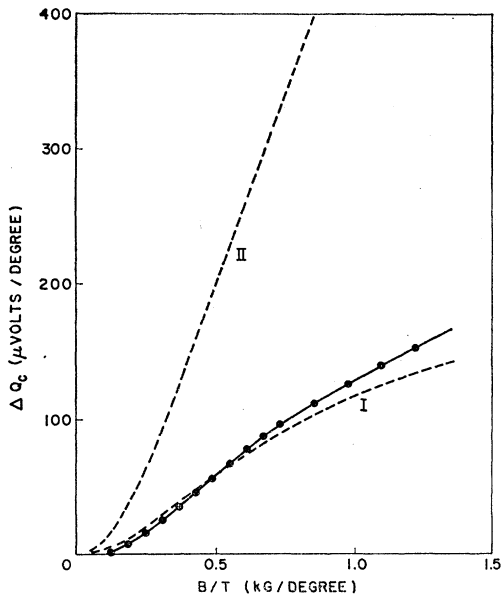


FIG. 5. Comparison of calculated value of electronic contribution with the experimentally observed  $\Delta Q_e$  at 82°K. The solid line shows the experimental curve, the curves marked I and II are, respectively, the calculated values using Eq. (3) and the results of Ref. 16.

hence reduce the thermoelectric power, due to a smaller value of the Boltzmann factor for the higher of the energy states. The neglected term approaches a value of  $(k/e) \ln 2 \approx 60 \mu\text{V}/\text{deg}$  as the splitting of the Landau level becomes much larger than  $kT$ . On account of a large  $g$  factor for electrons in InSb, the effect of spin-splitting should ideally have its maximum value in the highest fields even at 82°K. This would however spoil the good agreement obtained at this temperature between the observed and the calculated values in a region of magnetic field where phonon-drag is expected to be negligible (Fig. 4). It might be that the splitting is washed out by some additional mechanism at liquid-nitrogen temperatures.

Figure 6 presents data similar to those of Fig. 4 at lower temperatures. Even for small values of  $B/T$ , the curves are shifted up as the temperature is reduced, indicating as discussed above the presence of small phonon-drag component even in zero field. It may be pointed out that at still lower temperatures, e.g., around 10°K, where phonon drag in zero field again is negligible, values of  $\Delta Q_e$  fall closer to 82°K curve for values of  $B/T$  up to  $\leq 0.4$  kG/deg. As the field is increased, the observed value of  $\Delta Q_e$  become much greater than the calculated electron contribution. We ascribe the difference between the observed and calculated values as  $\Delta Q_p$ , which becomes of the order of several thousand  $\mu\text{V}/\text{deg}$  in high magnetic fields. In Fig. 7 we have plotted  $\Delta Q_p$  obtained from the difference between the observed and the calculated values from Eq. (3) as a function of temperature for a fixed field of 50 kG. A sharp change in the temperature dependence

of  $Q_p$  as the temperature is reduced so that low-energy acoustic phonons are scattered mostly from the walls of the specimen, is to be noted.

As a final check, we have made measurements on two samples cut from the adjoining parts of a crystal, but of different cross-sectional area. The two samples with their respective heaters were mounted in the sample holder side by side. The current in each heater is adjusted till the thermoelectric voltage on each sample is the same in zero field. The values of  $\Delta Q$  on the two samples are then measured simultaneously as a function of  $B$ . This method is a crude version of the method used<sup>26</sup> to measure size effect on thermal conductivity but is adequate for thermoelectric measurements. The ratio of  $\Delta Q_e$  for the two samples in a field of 80 kG is plotted as a function of temperature in Fig. 8. The great sensitivity of  $Q$  to the size of the specimen at liquid-hydrogen temperatures indicates the presence of a large contribution from phonon drag. The low-temperature ratio of  $Q$  for the two samples is not so large as the ratio of their sizes. The samples are of nearly square cross section, being 0.15 cm (sample A) and 0.06 cm (sample B) on the side. It may be, as has been pointed out by Herring for the case in Ge, that there is an additional scattering mechanism present for phonons which gives a relaxation length comparable to the size of the specimen.

The results of this investigation can be summarized as follows:

- (i) In the absence of a magnetic field, the phonon-

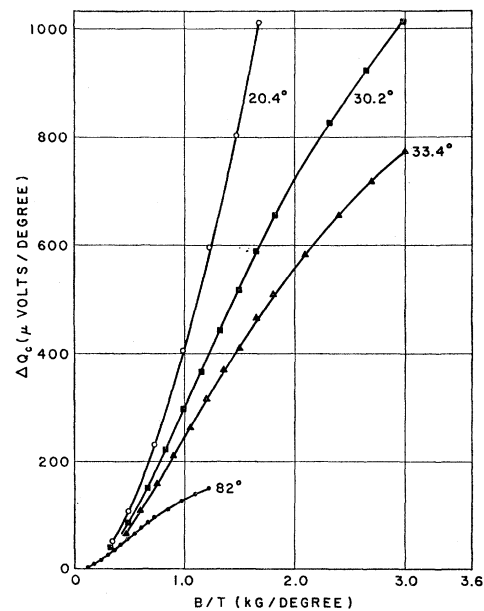


FIG. 6. Change of  $Q$  in the quantum region at lower temperatures. The rapidly diverging curves indicate a large contribution from phonon drag.

<sup>26</sup> T. H. Geballe and G. W. Hull, in Conférence de Physique des Basses Températures, Paris, September 2-8, 1955 (unpublished); see also Ref. 7.

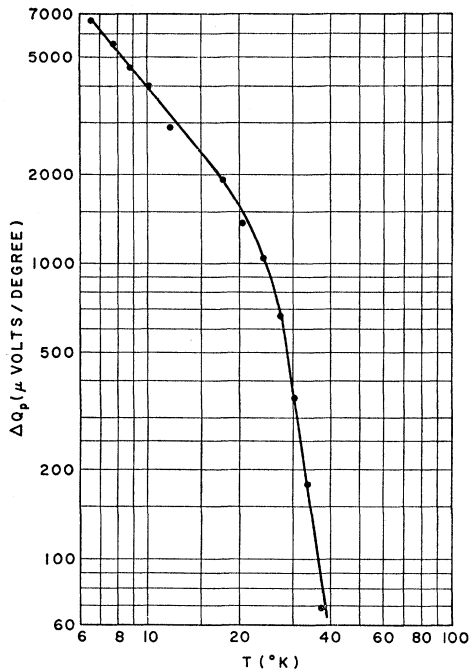


FIG. 7. Estimated values of phonon-drag contribution  $Q_p$  as a function of temperature for fixed value of magnetic field, 50 kG.

drag contribution to  $Q$  is small. The maximum value of  $Q_p$  is less than  $50 \mu\text{V}/\text{deg}$  and is obtained near  $20^\circ\text{K}$ . For temperatures outside the range between 10 and  $30^\circ\text{K}$ ,  $Q_p$  is completely negligible.

(ii) In the extreme quantum region, the electronic contribution to  $Q$  above the classical saturation value is given by Eq. (3).

(iii) In very high magnetic fields and at low temperatures, the observed change in  $Q$  cannot be explained by the change in the electronic contribution only. The difference between the observed and calculated values, which amounts to several  $\text{mV}/\text{deg}$ , is due to the phonon-drag effect.

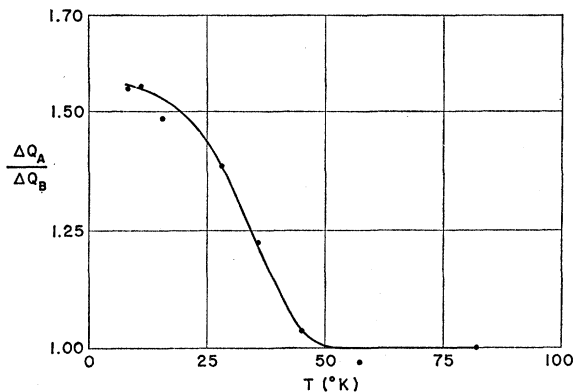


FIG. 8. Ratio of the thermoelectric power of two samples of different cross section area for a fixed field of 80 kG plotted as a function of temperature.

#### ACKNOWLEDGMENTS

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#### APPENDIX

Consider a gas of free electrons of density  $n/\text{cc}$ , in crossed electric and magnetic fields. The constant fields  $E$  and  $B$  are directed along the  $x$  and  $z$  axis, respectively. In the one-electron approximation, the system can be described by a one-particle Hamiltonian which is the same for all electrons of the system and is given by

$$\mathcal{H} = (1/2m)\{P_x^2 + (P_y + m\omega x)^2 + P_z^2\} + eEx, \quad (\text{A1})$$

where  $\omega = eB/mc$ . In what follows we neglect the energy of the electron spins.

The eigenfunctions and eigenvalues correct up to terms linear in the electric field are given by

$$\psi_{l, k_y, k_z} = \exp(ik_y y + ik_z z) \varphi_l(x - x_0), \quad (\text{A2})$$

$$\mathcal{E}_{l, k_y, k_z} = (l + \frac{1}{2})\hbar\omega + (\hbar^2 k_z^2 / 2m) + eEx_0, \quad (\text{A3})$$

where

$$x_0 = -(\hbar/m\omega)(k_y + eE/\hbar\omega). \quad (\text{A4})$$

The  $\varphi_l$  are simple harmonic oscillator wave functions. As can be easily verified, a constant current  $ncE/B$  flows in the  $y$  direction in this system. To be in equilibrium, the system must exchange particles as well as energy with the "outside world" and therefore forms an "open system" in the thermodynamic sense. The energy current for a "closed system" which does not exchange particles with the outside world is given by the average of the product of the local velocity times the energy density and is given by

$$W_i^c = \frac{1}{2} \text{Tr}[(v_i \mathcal{H} + \mathcal{H} v_i) \rho], \quad (\text{A5})$$

where  $\rho$  is the density matrix and  $v_i$  is the velocity operator along the  $i$  axis. But to get energy current for an "open system" like the one under discussion, the energy density must be replaced by the enthalpy density,<sup>27</sup> the two differing from each other by the pressure of the electron gas  $P$ . This difference relates to the work done by the exchanged particles against the pressure of the electron gas,

$$W_i^0 = W_i^c + P \langle v_i \rangle. \quad (\text{A6})$$

The pressure  $P$  is independent of the energy spectrum of the electrons and is given by  $nkT$  where  $T$  is the temperature of the electron gas.

The density matrix  $\rho$  is diagonal with respect to the quantum numbers  $l, k_y, k_z$  and is just the Fermi dis-

<sup>27</sup> I. Prigogine, *Introduction to Thermodynamics of Irreversible Processes* (Interscience Publishers, Inc., New York), 2nd ed., Chap. 2; L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (W. A. Benjamin, Inc., New York, 1962), p. 135.

tribution<sup>28</sup> which in the limit of low density is given by

$$\rho_{\nu\nu} = \exp(\mu/kT) \exp(-\epsilon_\nu/kT), \quad (\text{A7})$$

where  $\epsilon_\nu$  is that part of  $\mathcal{E}_\nu$  which is independent of the electric field and  $\mu$ , the electrochemical potential, is determined by

$$\begin{aligned} n &= 2 \sum_\nu \exp(\mu - \epsilon_\nu)/kT \\ &= 2 \left(\frac{1}{2\pi}\right)^2 \frac{m\omega}{\hbar} \left(\frac{2\pi m kT}{\hbar^2}\right)^{1/2} \frac{1}{2 \sinh(\hbar\omega/2kT)} \exp\left(\frac{\mu}{kT}\right), \end{aligned}$$

which gives

$$\begin{aligned} \frac{\mu}{kT} &= \ln \left\{ \frac{n}{2} (2\pi)^3 \left(\frac{\hbar^2}{2\pi m kT}\right)^{3/2} \right\} \\ &\quad + \ln \left\{ 2 \sinh\left(\frac{\hbar\omega}{2kT}\right) \right\} - \ln \frac{\hbar\omega}{kT}. \quad (\text{A8}) \end{aligned}$$

From (A5), the energy current in  $y$  direction for the closed system  $W_y^c$  is given by

$$W_y^c = \sum \mathcal{E}_\nu \rho_{\nu\nu} v_{\nu y} + (\hbar\omega/2i) \text{Tr}(v_p^x). \quad (\text{A9})$$

The last term is due to noncommutability of  $v$  and  $\mathcal{H}$ . The diagonal matrix elements of  $v$  have a nonzero component only along the  $y$  direction which equals  $cE/B$  independent of  $\nu$ . Carrying out the summation over  $\nu$ ,

<sup>28</sup> A. H. Kahn and H. P. R. Frederikse, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1959), Vol. 9, p. 271.

we get, keeping only the lowest order terms in  $E$ ,

$$W_y^c = (cE/B)n \left[ \frac{1}{2} \hbar\omega \coth(\hbar\omega/2kT) + \frac{1}{2} kT \right]. \quad (\text{A10})$$

The energy current in the "open system,"  $W_y^0$ , is given by

$$W_y^0 = (cE/B)n \left[ \frac{1}{2} \hbar\omega \coth(\hbar\omega/2kT) + \frac{3}{2} kT \right]. \quad (\text{A11})$$

The above results can be more simply obtained by calculating the partition function  $Z$  and the enthalpy density  $h$ . The energy current is then  $(cE/B)h$ . The calculation can be done using either (i), the energy spectrum given by (A3) or (ii) the classical energy levels given by  $\epsilon = \hbar^2 k^2/2m$ . The result of calculation in case (ii) agree with that obtained using Boltzmann equation. Had we used energy density in place of enthalpy density to calculate energy current, the result using the thermodynamic approach would not agree with that obtained from the Boltzmann equation.

The heat flux  $F_y$  is given by

$$F_y = W_y^0 - n v_y \mu.$$

The thermoelectric power  $Q = (1/T)(F_y/J_y)$  has the following expression

$$\begin{aligned} Q &= \frac{k}{e} \left[ \frac{\hbar\omega}{2kT} \coth\left(\frac{\hbar\omega}{2kT}\right) + \frac{3}{2} + \ln\left(\frac{\hbar\omega}{kT}\right) \right. \\ &\quad \left. - \ln\left(2 \sinh\left(\frac{\hbar\omega}{2kT}\right)\right) - \frac{\mu_0}{kT} \right], \quad (\text{A12}) \end{aligned}$$

where

$$\frac{\mu_0}{kT} = \ln \left\{ \frac{n}{2} (2\pi)^3 \left(\frac{\hbar^2}{2\pi m kT}\right)^{3/2} \right\}.$$