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To cite this article: J - P Michenaud and J -P Issi 1972 *J. Phys. C: Solid State Phys.* **5** 3061

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Electron and hole transport in bismuth

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MS received 19 April 1972, in revised form 10 July 1972

Abstract. Measurements of the low field galvanomagnetic tensor components of bismuth from 77 to 300 K are reported. The eight magnetoresistivity coefficients A_{ij} are found to vary as T^{-2} up to nearly 120 K, then as $T^{-3.9}$ from 120 to 300 K. The electronic parameters are computed. The values of the electron ellipsoid tilt angle lie between $6^{\circ}40'$ and $8^{\circ}30'$ and the carrier density varies from 4.55×10^{17} to $24.5 \times 10^{17} \text{ cm}^{-3}$ at 77 K and 300 K respectively. All mobilities except μ_2 vary as T^{-2} in the lowest temperature range.

1. Introduction

1.1. General

Among the semimetals, bismuth is famous for its peculiar transport properties. We owe to this element the discovery of galvanomagnetic effects as well as their thermal equivalent, the thermomagnetic effects. The early observations were facilitated by the enormous magnetoresistance and Hall effect in this material, which stem directly from the very high carrier mobility of the two carrier species present. Transport effects in bismuth are more dramatic than in typical metals, or indeed in the other semimetals. To compute the band model parameters in a straightforward way from the galvanomagnetic data, one approach is to turn to low field measurements, that is fields for which $\mu B \ll 1$ where μ is some carrier mobility and B the magnetic induction, and in this case the effects are very small. Zitter (1962) was the first to study rigorously this low field condition in his investigation of the galvanomagnetic tensor of bismuth at 4.2 K. Hartman (1969) extended this work by exploring carefully the range 4.2 to 15.7 K. His results were consistent with the known Fermi surface (FS) and confirmed the puzzling T^{-2} dependence of the mobilities which was previously inferred by many authors (Chopra *et al* 1971).

Abeles and Meiboom (1956) and Okada (1957) explored a higher temperature range and derived expressions of the galvanomagnetic tensor consistent with the FS model of bismuth. The data of Abeles and Meiboom were taken at 80 and 300 K and those of Okada at four temperatures from 113 to 318 K and all data were relative to fields above 200 Oe. In the 80–300 K range, although the EMF measured are higher than in the liquid helium range, the problem of measuring them is still a delicate one (see § 2.2). The situation is further complicated by the fact that at higher temperatures bismuth has a relatively good thermoelectric figure of merit ($\sim 0.5 \times 10^{-3} \text{ K}^{-1}$) which means that, under usual experimental conditions, a given DC current flowing across the sample will give rise to an

appreciable thermoelectric voltage drop in addition to the ohmic voltage drop (Issi *et al* 1971).

Workers at Durham have used galvanomagnetic effects as a tool to probe the FS and the scattering mechanism in the two other group V semimetals, antimony (Oktü and Saunders 1967) and arsenic (Jeavons and Saunders 1969). They found interesting features of the FS and showed that carrier degeneracy was high and that its density remains temperature insensitive up to 300 K. For both semimetals an anomalous temperature variation of the mobilities was observed, a $T^{-1.4}$ for antimony and a $T^{-1.7}$ for arsenic instead of the expected T^{-1} variation.

The aim of the present work is to report the measured low field isothermal galvanomagnetic coefficients of bismuth from 77 to 300 K. From these data, and assuming that the usual relations between these coefficients and the microscopic parameters are valid, the temperature variations of the mobilities, carrier density and tilt angle are computed.

1.2. The Fermi surface of bismuth

Bismuth crystallizes in the rhombohedral A_7 structure of the point group $\bar{3}m$. Its unit cell consists of two atoms, and one can think of its lattice as being obtained by displacing slightly the atoms of a simple cubic lattice thus causing a small overlap of the fifth and sixth bands (Jones 1934). This explains the presence at 0 K of an equal small density of electrons and holes ($\sim 3 \times 10^{17} \text{ cm}^{-3}$).

The shape of the FS at 0 K has been determined by the sharpest experimental tools available, mostly quantum effects (Boyle and Smith 1963, Dresselhaus 1971) and there is now good agreement for a model of the FS. The model consists of a set of ellipsoids located at the L and T points of the Brillouin zone, six half electron ellipsoids (L points) and two hole ellipsoids of revolution around the trigonal axis (T points). The prolate electron ellipsoids are tilted from the trigonal plane by an angle of about 6° . The electron band was found to be nonparabolic (Lax 1960) because of a lower lying hole band, located also at point L, separated by a small energy gap of 11 meV (Dresselhaus 1971). The Fermi energy for electrons is 26 meV and for holes 11 meV. The ellipsoids are usually described in a triad consisting of a binary (labelled 1), a bisectrix (2) and the trigonal (3) axes. Brown *et al* (1968) pointed out that a well defined sense for these axes must be given in order to avoid confusion when dealing with the sign of some galvanomagnetic coefficients. Their convention relative to the sign will be adopted here, that is a right handed triad in which the L point of the Brillouin zone lies in the first quadrant and the tilt angle is positive.

1.3. The galvanomagnetic tensor of bismuth

The phenomenological theory of the low field galvanomagnetic effects for the A_7 structure has been broadly described by many authors (Okada 1957, Juretschke 1955, Drabble and Wolfe 1956). The basic concepts are well summarized by Hartman (1969) and will not be dealt with in detail here. In short, the theory assumes that the magnetoresistivity tensor $\rho(\mathbf{B})$ may be expanded in a power series of $\mathbf{B}$, if $\mathbf{B}$ is low enough. Retaining only terms to the second order in $\mathbf{B}$, each component of the tensor may be written

$$\rho_{ij}(\mathbf{B}) = \rho_{ij}^0 + R_{ijk}B_k + A_{ijkl}B_kB_l \quad (1)$$

with the usual summation convention. The number of independent coefficients in the expansion (1) was calculated by Juretschke (1955) using Onsager's (1931a, b) reciprocity relations

$$\rho_{ij}(\mathbf{B}) = \rho_{ji}(-\mathbf{B}) \quad (2)$$

and point group symmetry arguments. Such an analysis shows that only twelve independent coefficients remain, two resistivities ρ_{11}^0 and ρ_{33}^0 , two Hall coefficients R_{123} and R_{231} and eight magnetoresistance coefficients A_{1111} (or A_{11} in shorthand notation), A_{3333} (A_{33}), A_{1123} (A_{14}), A_{2311} (A_{41}), A_{1122} (A_{12}), A_{1133} (A_{13}), A_{3311} (A_{31}) and A_{2323} (A_{44}).

Similarly, the conductivity tensor may be expanded as

$$\sigma_{ij}(\mathbf{B}) = \sigma_{ij}^0 - P_{ijk}B_k - S_{ijkl}B_kB_l \quad (3)$$

and the relationship between the coefficients in (1) and (3) may be readily obtained by solving the set of equations

$$\sigma_{ij}(\mathbf{B})\rho_{jk}(\mathbf{B}) = \delta_{ik} \quad (4)$$

where δ_{ik} is the Kronecker delta.

The magnetoconductivity coefficients can be calculated and expressed in terms of mobilities, carrier densities and tilt angles. The general procedure consists of expressing the current density for one ellipsoid using the Boltzmann equation and assuming an isotropic relaxation time (Abeles and Meiboom 1954). If intravalley scattering is also assumed then the total current density is obtained by summing the contribution of each ellipsoid.

Freedman and Juretschke (1961) gave a general expression for the conductivity coefficients considering a set of tilted ellipsoids for holes as well. The coefficients are then the sum of two terms:

$$\begin{aligned} \sigma_{ij}^0 &= \sigma_{ij,e} + \sigma_{ij,h} \\ P_{ijk} &= P_{ijk,e} + P_{ijk,h} \\ S_{ij} &= S_{ij,e} + S_{ij,h} \end{aligned}$$

where

$$\begin{aligned} \sigma_{11,e}^0 &= \frac{1}{2}Ne(\mu_1 + C_e\mu_2 + S_e\mu_3) \\ \sigma_{33,e}^0 &= Ne(S_e^2\mu_2 + C_e^2\mu_3) \\ P_{231,e} &= \frac{1}{2}Ne\{\mu_2\mu_3 + \mu_1(S_e^2\mu_2 + C_e^2\mu_3)\} \\ P_{123,e} &= Ne\{\mu_1(C_e^2\mu_2 + S_e^2\mu_3)\} \\ S_{13,e} &= \frac{1}{2}Ne(\mu_1 + C_e^2\mu_2 + S_e^2\mu_3)\{\mu_1(C_e^2\mu_2 + S_e^2\mu_3)\} \\ S_{31,e} &= \frac{1}{2}Ne(S_e^2\mu_2 + C_e^2\mu_3)\{\mu_2\mu_3 + \mu_1(S_e^2\mu_2 + C_e^2\mu_3)\} \\ -S_{44,e} &= \frac{1}{2}Ne(S_e^2\mu_2 + C_e^2\mu_3)\{\mu_1(C_e^2\mu_2 + S_e^2\mu_3)\} \\ S_{11,e} &= \frac{1}{8}Ne\{S_e^2\mu_2(\mu_1 - \mu_3)^2 + C_e^2\mu_3(\mu_1 - \mu_2)^2 + 3C_e^2S_e^2\mu_1(\mu_2 - \mu_3)^2\} \\ S_{12,e} &= \frac{1}{8}Ne\{3S_e^2\mu_2(\mu_1^2 + \mu_3^2) + 3C_e^2\mu_3(\mu_1^2 + \mu_2^2) + C_e^2S_e^2\mu_1(\mu_2 - \mu_3)^2 + 2\mu_1\mu_2\mu_3\} \\ S_{33,e} &= NeC_e^2S_e^2\mu_1(\mu_2 - \mu_3)^2 \\ S_{14,e} &= -\frac{1}{4}NeC_eS_e\mu_1(\mu_2 - \mu_3)(-\mu_1 + C_e^2\mu_2 + S_e^2\mu_3) \\ S_{41,e} &= -\frac{1}{4}NeC_eS_e(\mu_2 - \mu_3)\{\mu_2\mu_3 - \mu_1(S_e^2\mu_2 + C_e^2\mu_3)\} \end{aligned} \quad (5)$$

Here, N is the total density of electrons, $S_e \equiv \sin \varphi_e$, $C_e \equiv \cos \varphi_e$, where φ_e is the tilt

angle of electrons and μ_1, μ_2, μ_3 , are the mobilities along the principal axes of the ellipsoids. The hole coefficients are obtained by replacing N by P , the density of holes, μ_i by ν_i , the hole mobilities, and ϕ_e by ϕ_h , the tilt angle for the hole ellipsoids. The corresponding expressions for bismuth are obtained by making $C_h = 1$, $S_h = 0$ and $\nu_1 = \nu_2$.

2. Experimental

2.1. Samples and cryostat

A spherical single crystal was grown by a method described elsewhere (Issi and Moureau 1970) from bismuth of 6N nominal purity purchased from the Métallurgie Hoboken-Olen. X ray photographs revealed that the crystal was free from strains. The main crystallographic directions were determined by means of the (111) main cleavage plane (trigonal plane) and the slip lines parallel to the binary axes. All samples studied were cut from the same sphere. They were of rectangular cross section and their thinnest dimension was large enough to avoid diffusion size effects at 77 K. Approximate dimensions of each sample are indicated in table 1. While in principle only two samples are needed to

Table 1. Approximate dimensions of samples and directions of current, magnetic induction and measured EMF with respect to the main crystallographic directions

Sample number	Dimension (mm)†			Direction of			Coefficient measured
	<i>a</i>	<i>b</i>	<i>l</i>	<i>J</i>	<i>B</i>	<i>E</i>	
1	4.2 [1]	3.9 [2]	13 [3]	3		3	ρ_{33}^0
				3	1	3	A_{33}
				3	3	3	A_{31}
				3	2	1	R_{231}
				1		1	ρ_{11}^0
2	3.7 [2]	2.4 [3]	13 [1]	1	1	1	A_{11}
				1	2	1	A_{12}
				1	3	1	A_{13}
				1	2.3 plane	1	A_{14}
3	2.7 [3]	2.7 [1.2 plane]	13 [1.2 plane]	1.2 plane	J.3 plane	3	A_{41}
				1.2 plane	J.3 plane	3	A_{44}
4	0.65 [3]	2.0 [1]	12 [2]	2	3	1	R_{123}

† The direction 1,2 or 3, is indicated in brackets.

measure all the resistivity tensor components we have, for practical reasons, used four of them. For example, one separate thin sample was cut and used to measure the small Hall coefficient R_{123} .

The edges of samples 1, 2 and 4 (table 1) were parallel to the main crystallographic directions while sample 3 had its axis in the (1, 2) plane inclined at an angle θ with respect to the 1 direction. This sample was used to measure the coefficients A_{44} and A_{41} . If we take ϕ as the angle between the magnetic induction B and the 3-axis then:

$$E_3 = JB^2(A_{41} \sin 3\theta \sin^2 \phi - A_{44} \sin 2\phi) \quad (6)$$

The angle θ was taken equal to 15° . Its sign was determined by means of an etch pit analysis following the convention of Brown *et al* (1968), in order to obtain the true sign

of A_{41} . The etchant consisted of 6 parts of fuming nitric acid, 6 parts of glacial acetic acid and 1 part of water.

The upper parts of samples 1, 2 and 4 were each fixed to a cylindrical copper plate by means of Wood's alloy and the plate with an intermediate indium foil was screwed on the copper sink of a vertical cryostat which is described in detail elsewhere (Issi *et al* 1971).

The essential feature of the entire experimental system is that it was designed to ensure strict isothermal conditions for the samples. The main trouble comes from the Peltier effect which accompanies current flow, resulting in an appreciable thermoelectric voltage drop which reverses with current reversal. This voltage drop, for the most favourable experimental conditions, introduces errors larger than 10% in the estimation of the electrical resistance.

2.2. Measurement technique

Exploratory measurements indicated that the low field limits in the investigated temperature range correspond to a magnetoresistance $\Delta\rho/\rho_0$ of roughly 2% whatever the temperature or direction. If we take for example the situation at 77 K, where it was shown (Issi *et al* 1966) that the low field limit lies around 100 Oe, then in order to check the linearity of each coefficient with respect to B^2 we need to start with lower fields (sometimes 20 Oe). This could be realized by achieving an overall stability of 2 parts in 10^4 in our system and a resolution of the order of 10^{-8} V in EMF measurements for a sample current of 1 A. Further details about the experimental set up may be found elsewhere (Michenaud 1970).

For the Hall coefficient R_{231} , although for the sample used the EMF measured are of the order of 10^{-6} V at low fields, requirements about stability are less stringent since we do not have to deal with differences of two nearly equal numbers (as is the case for magnetoresistance). However, the R_{123} coefficient is about two orders of magnitude smaller than R_{231} and thus for a sample of the same thickness and for the same longitudinal current an EMF of the order of 10^{-8} V should be measured. For this reason we have used a special thin sample to measure R_{231} , the error which could be introduced from diffusion size effects being here much less than that introduced if a thicker sample is used.

Before measuring any coefficient, we made use of the thermocouples attached on the sample and the small carbon resistor to reduce to a few millidegrees the temperature gradient due mainly to the Peltier effect. In an ambient helium gas this operation took only a few minutes (Issi *et al* 1971). For each magnetoresistance coefficient at each temperature at least three low field values were taken, each value corresponding to the mean of a set of five to ten readings. The low field limit was found to be anisotropic as expected, on account of the different mobility contributions in the component measured. At 77 K, for example, it was around 40 Oe for A_{12} and 140 Oe for A_{13} . All measurements were taken at eight selected temperatures from 77 to 300 K.

3. Results and discussion

3.1. The low field resistivity tensor

Figure 1 represents the two components of the zero field isothermal resistivity ρ_{11} and ρ_{33} against temperature. The experimental data of Okada (1957) are also shown for comparison. The broken line represents lower temperature data given by Friedman

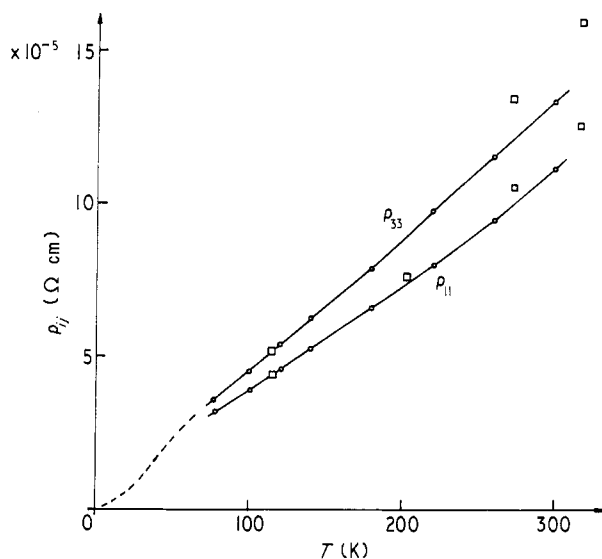


Figure 1. The two components of the zero field isothermal resistivity ρ_{11} and ρ_{33} against temperature. $\circ$ Our data; $\square$ Okada (1957).

(1967). The resistivity obeys a T^2 law at low temperatures while in the highest temperature range the behaviour is very close to linear. This almost linear behaviour is pure accident and is due, as will be seen later, to the combined effect of a peculiar type of scattering and a temperature variation of the carrier density.

Figure 2 represents the two low field Hall coefficients against temperature. It should

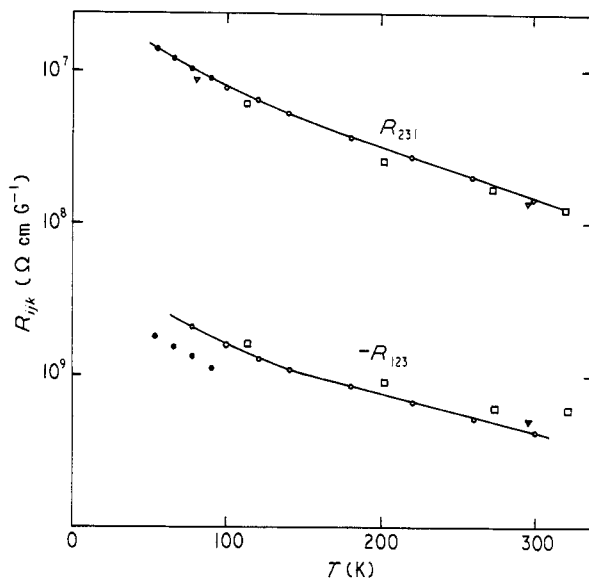


Figure 2. Low field Hall coefficients against temperature. $\circ$ Our data; $\square$ Okada (1957); ∇ Abeles and Meiboom (1956); $\bullet$ Jain *et al* (1968).

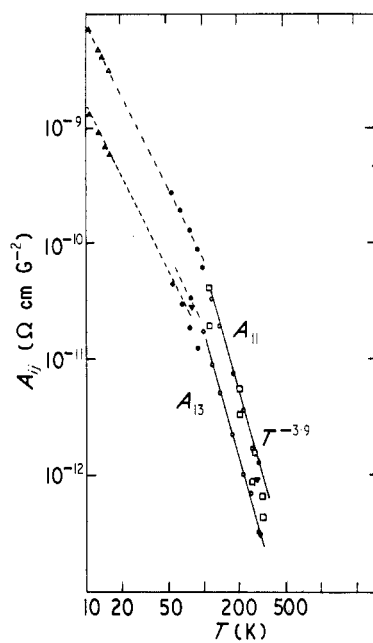


Figure 3. The A_{11} and A_{13} coefficients against temperature. Δ Hartman (1969).

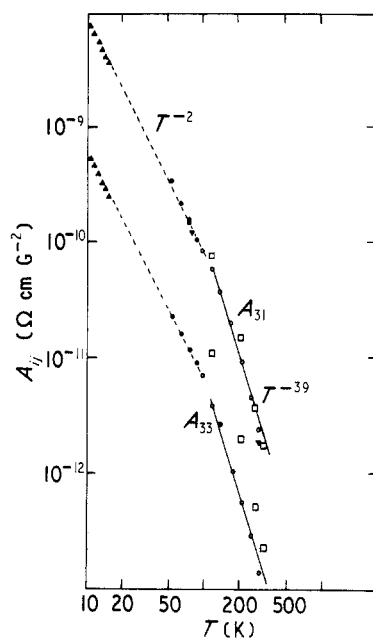


Figure 4. The A_{31} and A_{33} coefficients against temperature.

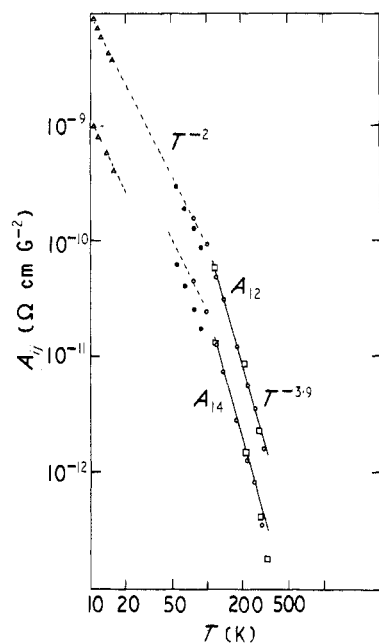


Figure 5. The A_{12} and A_{14} coefficients against temperature.

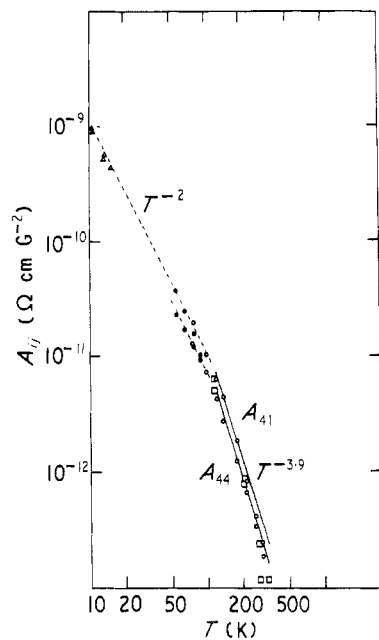


Figure 6. The A_{41} and A_{44} coefficients against temperature.

be noted that the coefficient R_{123} is negative and is about two orders of magnitude smaller than R_{231} , which is positive. Our data are compared with those of Abeles and Meiboom (1956), Okada (1957) and Jain *et al* (1968).

Figures 3 to 6 show the magnetoresistance coefficients. The data are also compared with those of other authors and the discrepancy with those of Okada, especially for A_{33} and A_{13} , should be noted.

In the present work one remarkable feature is shown which could not be suspected from the results of previous studies: for all the A_{ij} coefficients there is a T^{-2} variation at low temperatures, which breaks at some temperature near θ_D to an almost T^{-4} law. Also, except for A_{13} and A_{14} , if we extrapolate Hartman's (1969) results to higher temperatures, the agreement with the present results is striking. Further, we have measured the A_{ij} coefficients with one of Hartman's crystals (sample axis $\parallel$ 3 direction) at 77 K and found that the results were in agreement with ours to within 10%. Taking Hartman's and the present data together it can be reasonably inferred that the general behaviour of the A_{ij} coefficients from 8 to 300 K is a T^{-2} variation at low temperatures, then an abrupt change occurs around 120 K and a $T^{-3.9}$ variation is observed at higher temperatures. The values of the various coefficients are reported in table 2.

Table 2. The measured low field galvanomagnetic tensor components

$T(K)$	ρ_{11}	ρ_{33}	R_{231}	R_{123}	A_{11}	A_{12}	A_{13}	A_{33}	A_{31}	A_{14}	A_{41}	A_{44}
77	3.2	3.6	10.6	-0.21	133	160	35.5	12.2	160	45	20	-13.0
100	3.9	4.5	7.8	-0.16	65	94	18	7.1	87.0	24	10.7	-7.5
120	4.6	5.4	6.5	-0.13	34	49	9.2	3.9	60.6	12.6	6.5	-4.3
140	5.3	6.3	5.3	-0.11	20.3	31.5	5.25	2.7	38.0	7.0	4.6	-2.8
180	6.6	7.9	3.66	-0.086	7.7	12.5	2.34	1.03	20.6	2.8	1.9	-1.24
220	8.0	9.8	2.66	-0.067	3.8	5.5	1.04	0.56	9.4	1.2	0.84	-0.68
260	9.5	11.6	2.0	-0.053	1.73	3.56	0.55	0.29	4.7	0.83	0.42	-0.35
300	11.2	13.4	1.47	-0.043	0.7	1.6	0.33	0.14	2.4	0.35	0.25	-0.19
$10^{-5} \Omega \text{ cm}$ $10^{-8} \Omega \text{ cm G}^{-1}$ $10^{-12} \Omega \text{ cm G}^{-2}$												

3.2. The electronic parameters

A least squares method was used to compute the carrier density $N = P$, the mobilities μ_i and ν_i and the tilt angle φ_e of electrons from the relations (5).

The program was run on the IBM 360/67 computer of the University of Durham (England). We used the program written by Jeavons and Saunders (1969) for arsenic. The mathematical principle of this calculation is described by Jeavons (1969). The values of six out of the seven parameters μ_i , ν_i , N and φ_e are given in table 3. Since the contribution of holes to the conductivity in the trigonal direction is very small the function to minimize Q is almost insensitive to ν_3 , and we could not obtain the values of this mobility.

The value of the angle of tilt for electrons varies from $6^\circ 40'$ at 77 K to $8^\circ 30'$ at 300 K (table 3). However, this temperature variation does not seem to be significant. This is due to the fact that the computer gives the value of the cosine of the angle, and is therefore very insensitive to the angle for small values. The values given here lie in the range found by various authors at 4.2 K.

Figure 7 shows the variation of the carrier density with temperature from 4.2 to 300 K. At 4.2 K we took Barghava's (1967) value of $3 \times 10^{17} \text{ cm}^{-3}$ as determined from the

Table 3. The computed electronic parameters fitting the galvanomagnetic data

$T(K)$	φ_e	$N = P$ (10^{17} cm^{-3})	μ_1 (10^4 cm^2 $\text{V}^{-1} \text{ s}^{-1}$)	μ_2 (10^2 cm^2 $\text{V}^{-1} \text{ s}^{-1}$)	μ_3 (10^4 cm^2 $\text{V}^{-1} \text{ s}^{-1}$)	$v_1 = v_2$ (10^4 cm^2 $\text{V}^{-1} \text{ s}^{-1}$)
77	6°40'	4.55	64	60	39	10
100	7°40'	5.67	41	41	24	6.7
120	7°10'	7.03	27	31	17	4.6
140	7°50'	8.47	20	21	12	3.5
180	7°50'	11.9	11	16	7.0	2.0
220	7°50'	15.6	7.1	10.0	4.1	1.2
260	8°30'	20.0	4.6	6.5	2.6	0.95
300	8°30'	24.5	3.2	4.4	1.8	0.6

de Haas–van Alphen effect. The full line represents the values computed from the present work from 77 to 300 K, where the carrier density varies from 4.55×10^{17} to $24.5 \times 10^{17} \text{ cm}^{-3}$. The carrier density thus changes by nearly an order of magnitude from 4.2 to 300 K, a situation which is quite different from the case of the two other group V semimetals in this temperature range (Oktü and Saunders 1967, Jeavons and Saunders 1969).

The mobilities calculated from the present work are represented in figure 8 together with those of Hartman (1969). Above 140 K they follow a $T^{-2.5}$ law. Hartman's results above 10 K follow a T^{-2} law for μ_1 , μ_3 and v_1 . Between 77 and 120 K an almost T^{-2} dependence is also observed. However, there is a gap in the experimental data between the highest temperature investigated by Hartman (15.7 K) and the lowest reported here (77 K).

If we attempt to join the mobilities computed by Hartman by extrapolating our lowest temperature data the curves lie above the corresponding values of Hartman, contrary to what happens with most of the A_{ij} coefficients. Thus the behaviour of the

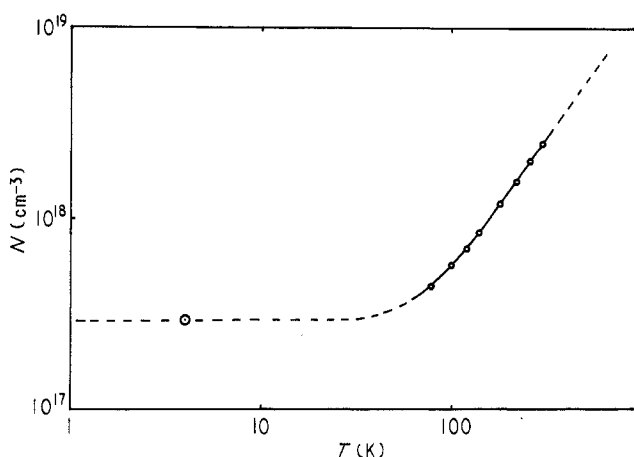


Figure 7. Variation of the carrier density with temperature. The value at 4.2 is taken from Bhargava (1967).

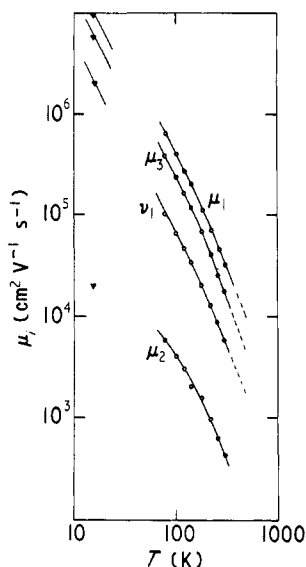


Figure 8. Electron and hole mobilities against temperature. Below 77 K the straight lines are relative to Hartman's (1969) data.

mobilities in the unexplored region probably does not obey a simple law. However from our knowledge of the resistivity variation in this range and from the partial results on the mobilities (μ_1 and ν_1) obtained by Friedman (1967) one can form a reasonable estimate about the mobility variation from 15.7 to 77 K. Friedman finds that the mobilities μ_1 and ν_1 follow a T^{-2} law up to about 50 K and 20 K respectively, then vary in a slower way.

Since the A_{ij} , which are roughly expressed as a ratio (mobility/carrier density), vary as T^{-2} in this range, this means that the density probably begins to increase in the same range. This fact suggests also that a part of the scattering mechanism may be intimately related to carrier excitation. As pointed out by Lopez (1968), carrier generation from the main valence to the main conduction band requires a phonon temperature of 43 ± 4 K. The increase in the carrier concentration must then start around this temperature, as indicated by a portion of the broken curve, figure 7. In this connection, the ingenious qualitative model proposed by Lopez (1968) about the relaxation of carriers in bismuth could be used to explain the mobility behaviour: that is, on increasing the temperature, the 'recombination scattering' should dominate and this type of scattering could at last rule the 43 K phonon scattering. Since the relaxation time of such phonons is expected to vary between T^{-2} and T^{-1} , it could possibly explain the change in slope of the curve in the sense observed. Ordinary intervalley scattering would act in the opposite sense and yield a steeper variation with T .

The variation of μ_2 , although similar to that of the other mobilities at high temperatures, is rather peculiar in the lowest temperature range. If one represents Hartman's results on a log-log plot, he finds a variation closer to the T^{-5} typical metallic behaviour, rather than a T^{-2} law. This may be explained by the fact that the electron Fermi ellipsoids are extremely elongated in K space in the 2 direction. In the 1 and 3 directions low energy phonons scatter electrons through large angles even at 4 K, while in the 2 direction at low temperatures scattering will be less effective since, as is the case for ordinary metals, it will be of the small angle type.

Acknowledgments

The authors are indebted to Professor A Luyckx for the impetus he gave to the present research. They enjoyed the friendly collaboration of Dr G A Saunders at every stage of this work and benefited much from the many fruitful discussions and suggestions and acknowledge his critical reading of the manuscript. They are also thankful to Mr A Moureau for his help in making some of the experimental measurements. They are grateful to Mr Paul Coopmans for his invaluable technical help concerning the cryostat and many delicate mountings. One of the authors (JPI) is indebted to Dr S H Koenig and R D Brown for sending a batch of bismuth single crystals, among which were those of Hartman.

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