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Unusual electronic transport properties of a thin polycrystalline bismuth film

Ralph Rosenbaum[1] **and Jean Galibert**[2]

[1] Tel Aviv University, School of Physics and Astronomy, Raymond and Beverly Sackler Faculty of Exact Sciences, Ramat Aviv 69978, Israel
[2] Laboratoire National des Champs Magnétiques Pulsés, BP 14245, 143 Avenue de Rangueil, Toulouse Cedex 4, 31432, France

E-mail: rachel-r@zahav.net.il (Ralph Rosenbaum) and ralphr@post.tau.ac.il (Ralph Rosenbaum)

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Abstract

The electronic transport properties of a polycrystalline (3420 Å thick) bismuth film have been measured over a wide temperature interval (0.23 K $< T <$ 292 K) and a magnetic field range (0 T $< B <$ 25 T). The results for the polycrystalline film are very different and anomalous from those of an epitaxial thin bismuth film. The zero field resistance increases by a factor of five. The magnetoresistance (MR) values in perpendicular magnetic fields have the same magnitude at low temperatures as compared to the MR values at room temperature. The Hall coefficient data in perpendicular fields show oscillations at liquid helium temperatures; there should be no Shubnikov–de Haas oscillations in a polycrystalline bismuth film. The sign of the Hall coefficient at room temperature is positive in small fields and becomes negative in large fields. In contrast, the Hall coefficient is always negative in thick bismuth films. The magnetoresistances in parallel magnetic fields show maxima at intermediate fields followed by decreases at high field values; in theory there should be a small or no MR in the parallel field orientation. The most anomalous behaviours are large Hall voltages and Hall coefficients in parallel magnetic fields; the parallel Hall data also have oscillations at low temperatures. The magnetoresistance in transverse fields is anomalous and can be explained by strong diffused boundary scattering at the top and bottom surfaces of the film. Acceptable fits to most of the transport data are obtained using the two carrier expressions of Pippard and of Fawcett and using the Drude expression.

1. Introduction

The electronic transport properties of bismuth have been studied extensively both experimentally and theoretically during the past sixty years. As early as 1929, Kapitza [1] showed that the resistivity of pure crystalline bismuth samples depends strongly on magnetic field up to $B = 35$ T at 77 K $< T <$ 300 K. High-quality crystals of Bi were also shown to have extremely large magnetoresistances (MRs) at low temperature even in fields of several teslas, consistent with high carrier mobilities in this semi-metallic material, and equal number densities of electrons n and holes p. Good introductory review articles have been presented by Fawcett [2] and by Èdel'man [3]; and informative books on general transport properties in normal metals and semimetals have been written by Shoenberg [4], by Blatt [5] and by Pippard [6].

It was found that high-quality films could be obtained if the bismuth was deposited on heated mica substrates at 110 °C and the films were then annealed at 265 °C for 48 h [7], or if grown on a heated substrate at 165 °C [8], or fabricated by electrodeposition and then annealed [9, 10]. The variation of the zero field resistance of these annealed films with respect to temperature is similar to that of bulk Bi crystals, decreasing with temperature; and the carrier mobilities in such films were estimated to be greater than 35 m^2 V^{-1} s^{-1} at 4.2 K [7]. Large MR ratios $r = R(B)/R(0)$ of 4000 at 5 K were obtained for annealed films having thicknesses of 10–20 μm; in this case the magnetic field was oriented perpendicular to the film plane and to the current [9, 10].

Many authors claim that their Bi films are fully compensated at liquid helium temperatures; that is, the concentrations of electrons and holes are equal, namely $c = p/n \approx 1$, a property displayed approximately in high-quality bulk Bi crystals.

In this paper, we report magnetoresistances (MRs), the Hall effect and zero field resistivity data in a temperature range between 0.23 K $< T <$ 292 K and in fields up to 25 T on a polycrystalline 3420 Å bismuth film. This paper shows that the behaviours of these quantities are very anomalous compared to those of an epitaxial film. By using a simple two carrier model, we demonstrate that one can extract the mobilities of the electron and hole carriers as a function of temperature from the MR and Hall results, which in turn allows us to predict the zero field resistivity behaviour.

2. Theoretical background

2.1. Magnetoresistance expressions

In order to fit the bismuth magnetoresistance (MR) ratio data, $r = R(B)/R(0)$, taken in perpendicular, parallel and transverse fields, we have used a model involving the two independent carriers, the holes and electrons. Kaganov and Peschanski [11] were the first to propose an expression for the perpendicular magnetoresistance (MR), which was further refined by Fawcett [2]. Pippard elaborated on the theory and suggested that the MR ratio $r = \rho(B)/\rho(0) = R(B)/R(0)$ is given by the following expression (equation (1.55) in Pippard's book [6]):

$$r = [1 + (\mu_e B)(\mu_h B)(\mu_h + c_\perp \mu_e)/(\mu_e + c_\perp \mu_h)]/[1 + (1 - c_\perp)^2 \times (\mu_e B)(\mu_h B)\mu_e \mu_h/(\mu_e + c_\perp \mu_h)^2]; \quad (1a)$$

$$r \approx 1 + (\mu_{\mathrm{eff}} B)^2 = 1 + \{[c_\perp \mu_e \mu_h(\mu_e + \mu_h)^2]/[\mu_e + c_\perp \mu_h]^2\}B^2, \qquad \text{for } B \to 0; \quad (1b)$$

$$r \approx [1 + c_\perp(\mu_e/\mu_h + \mu_h/\mu_e) + c_\perp^2]/[1 - c_\perp]^2, \qquad \text{for } B \to \infty. \quad (1c)$$

Here $\mu_e = e\tau_e/m_e^*$ and $\mu_h = e\tau_h/m_h^*$ are the mobilities of the electrons and of the holes in perpendicular fields, and $c_\perp = p/n$ is the ratio of the hole p to electron n number densities. τ is the lifetime of the carrier, and m^* is its effective mass, whose magnitude depends upon the orientation of the magnetic field.

Assuming the approximation that the mobility of the holes is equal to that of the electrons, namely $\mu_h \approx \mu_e = \mu_{eff} = e\tau_{eff}/m^*_{eff}$, Pippard [6] obtained the following 'simplified' expression for equation (1*a*):

$$r_{simp} = R(B)/R(0) = \rho(B)/\rho(0) = [1 + (\mu_{eff} B)^2]/[1 + (1 - c_\perp)^2(\mu_{eff} B)^2/(1 + c_\perp)^2]. \quad (2)$$

This approximation gives us physical insight into the behaviour of equation (1*a*). For example, equation (2) exhibits symmetry about the value of $c_\perp = 1$; that is, replacing $c_\perp$ by $1/c_\perp$ has no effect at all. Thus, it is not possible to identify which carrier, the hole or the electron, is the majority carrier from the MR fits of an uncompensated sample, if the mobilities are similar. The majority carrier information comes from fits to the Hall coefficient data. For small fields, then $r \approx 1 + (\mu_{eff} B)^2$, exhibiting the well known quadratic field behaviour. For the compensated case where $c_\perp = 1$ or $p = n$, the MR follows a quadratic dependence in *all* the field range. For the uncompensated case where $c_\perp \neq 1$, the MR ratio *saturates* at high fields to the finite value of $r \approx (1+c_\perp)^2/(1-c_\perp)^2$. In the case of one type of carrier ($c_\perp$ is zero or infinity), the resistivity does not depend upon the field, and $R(B) = R(0)$. Hence as the carrier ratio $c_\perp$ decreases close to zero (or increases to infinity), then the saturation magnitudes of r approach values close to unity. The overall $r = R(B)/R(0)$ behaviour is illustrated nicely for different $c_\perp$ values in figure 1.16 in Pippard's book [6]. This model is also limited in that it cannot account for MR ratio values less than 1.

The Fawcett expression, equation (58) in [2], is extremely similar to that of equation (1*a*) of Pippard's [6]; namely

$$r = R(B)/R(0) = 1 + [c_\perp(\mu_e + \mu_h)^2\mu_e\mu_h B^2]/[(\mu_e + c_\perp\mu_h)^2 + (1 - c_\perp)^2(\mu_e^2\mu_h^2 B)^2], \quad (3)$$

where $c_\perp$ is again defined as $c_\perp = p/n$. Equation (3) exhibits all the properties of the Pippard expression equation (1*a*); and it predicts values for the MR ratio r to within a few per cent of those of Pippard's expression, equation (1*a*).

When the magnetic field is oriented parallel to the current flow, the Lorentz forces acting on the carriers should be very small in most metals, and therefore there should be a very small or negligible magnetoresistance. Hence $c_{eff} \approx 0$ or $R(B) \approx R(0)$ for all fields. For high-quality epitaxial films, the parallel magnetoresistance is about two orders of magnitude smaller than the perpendicular magnetoresistance at liquid helium temperatures [9, 10]; and this prediction works well. In contrast for polycrystalline films, the parallel magnetoresistances are slightly smaller than the magnitudes of the perpendicular magnetoresistances, arising from the strong diffused and specular scattering of the carriers off the rough surfaces of the polycrystallites. Kao and McGill discuss this scattering mechanism [12, 13]. Thus, there will be Lorentz forces acting on some of the carriers, resulting in sizable parallel MR ratios values and parallel Hall voltage magnitudes. The c_{eff} values extracted from the parallel MR ratio data are approximately one-half the values of those from the perpendicular MR ratio data. For the parallel field case c_{eff} is no longer equal to p/n but should be corrected to a smaller magnitude by a boundary scattering correction factor f_s; we propose that $c_{eff} = (p/n) f_s$ where $f_s < 1$. In addition, the mobilities extracted from the parallel field orientation data are larger than those of the perpendicular field orientation, owing to smaller effective masses m^*s in this field orientation.

2.2. Hall coefficient expressions

Fawcett and Pippard have derived identical expressions for the Hall coefficient: $R_{Hall} = V_{xy} t_{film}/IB = [V_{xy}(B) - V_{xy}(-B)]/2IB$, where t_{film} is the film thickness, I is the current, V_{xy} is the voltage measured across the two slightly misaligned Hall probes, and B and $-B$ are the forward and reversed perpendicular fields. Referring to equation (61) in Fawcett's review article [2] or to equation (1.55) in Pippard's book [6], the two carrier model for the Hall constant R_{Hall} of an isotropic material is

$$R_{\mathrm{Hall}} = (-1/|e|)[n\mu_{\mathrm{e}}^2 - p\mu_{\mathrm{h}}^2 + (n-p)\mu_{\mathrm{e}}^2\mu_{\mathrm{h}}^2B^2]/[(n\mu_{\mathrm{e}} + p\mu_n)^2 + (n-p)^2\mu_{\mathrm{e}}^2\mu_{\mathrm{h}}^2B^2];$$

or

$$R_{\mathrm{Hall}} = (-1/n|e|)[\mu_{\mathrm{e}}^2B^2 - c_\perp\mu_{\mathrm{h}}^2B^2 + (1-c_\perp)\mu_{\mathrm{e}}^2B^2\mu_{\mathrm{h}}^2B^2] \times [(\mu_{\mathrm{e}}B + c_\perp\mu_{\mathrm{h}}B)^2 + (1-c_\perp)^2\mu_{\mathrm{e}}^2B^2\mu_{\mathrm{h}}^2B^2]^{-1}, \quad (4a)$$

where $c_\perp = p/n$. In the limit of vanishing perpendicular fields for $B \to 0$:

$$R_{\mathrm{Hall}} \approx (-1/|e|)[n\mu_{\mathrm{e}}^2 - p\mu_{\mathrm{h}}^2]/[(n\mu_{\mathrm{e}} + p\mu_{\mathrm{h}})^2] = (-1/n|e|)[\mu_{\mathrm{e}}^2 - c_\perp\mu_{\mathrm{h}}^2]/[\mu_{\mathrm{e}} + c_\perp\mu_{\mathrm{h}}]^2; \quad (4b)$$

and in the limit of large perpendicular fields for $B \to \infty$:

$$R_{\mathrm{Hall}} \approx (-1/|e|)/(n-p) = (-1/n|e|)/(1-c_\perp). \quad (4c)$$

All three above expressions yield interesting predictions. Equation (4*a*) predicts that the Hall coefficient has a significant magnetic field dependence, increasing to more negative magnitudes by a factor of approximately three. This result is very different from the single carrier prediction in that the Hall coefficient is a *constant*, independent of field. Equation (4*c*) predicts that the Hall coefficient saturates to a negative value in high fields. Equation (4*b*), the low field limit, is particularly interesting in that the sign of the Hall coefficient depends upon the numerator term, namely upon $[\mu_{\mathrm{e}}^2 - c_\perp\mu_{\mathrm{h}}^2]$. If the mobility of the holes μ_{h} is considerably larger than the electron mobility μ_{e}, then this term is negative, thus producing a *positive* Hall coefficient value. As we will see, positive R_{Hall} values are observed at room temperature in bismuth films thinner than about 7000 Å [14–16]. Thus we have the surprising conclusion that for thin bismuth films, $\mu_{\mathrm{h}} > \mu_{\mathrm{e}}$.

Mobilities and $c_\perp$ were extracted from both the perpendicular MR data and Hall coefficients; the same mobilities and $c_\perp$ were used in fitting both sets of data, thus maintaining self-consistency. We also applied this same fitting procedure to the parallel MR data and Hall coefficient data, extracting different mobilities and a smaller value for c_{eff}.

In fitting the perpendicular Hall coefficient data using equation (4*a*), we observed that the value for the prefactor $n = n_{\mathrm{Hall}}$, the electron concentration, had to be treated as a 'free' fitting parameter and had to be scaled up by one order of magnitude greater than the value $n = n_{\mathrm{cond}}$ found from the Drude expression using the zero field conductivity data $\sigma(T)$. We have no theoretical explanation for this surprising result. We believe that the correct magnitude for n is extracted from the conductivity data. We also had to scale up $n = n_{\mathrm{Hall}}$ in fitting the parallel Hall coefficient data.

Owing to the diffused and specular scattering of the carriers at the boundaries of the crystallites [12, 13], the parallel Hall coefficients are extremely large, being approximately one-quarter of the magnitudes of the perpendicular Hall coefficients. This is a special and anomalous property of the polycrystalline nature of the bismuth films and has gone unnoticed over the past fifty years of investigating these films.

2.3. Zero field resistivity expression

The resistivity $\rho(T)$ is determined by inverting the conductivity $\sigma(T)$ calculated from the classical two carrier Drude model. Since the three electron Fermi pockets and the one hole Fermi pocket are in different momentum space locations, the carriers do not annihilate one another but both contribute to the total conductivity:

$$\sigma(T) = 1/\rho(T) = en_{\mathrm{cond}}\mu_{\mathrm{e,eff}} + ep_{\mathrm{cond}}\mu_{\mathrm{h,eff}} = en_{\mathrm{cond}}(\mu_{\mathrm{e,eff}} + c_\perp\mu_{\mathrm{h,eff}}). \quad (5)$$

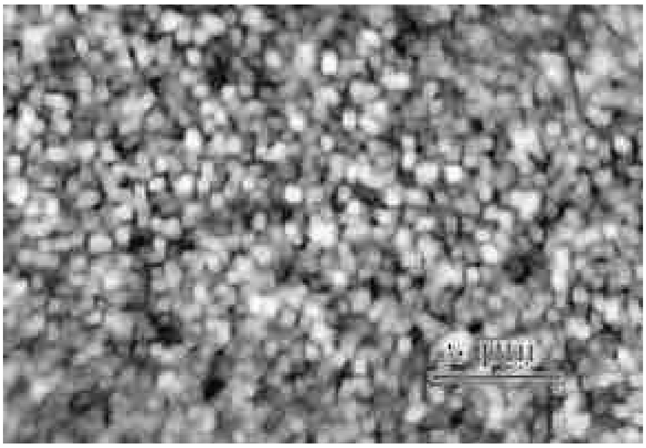

Figure 1. Optical microscope image of the surface of a 3235 Å polycrystalline bismuth film. The 'ruler line symbol' represents a length scale of 5 μm. Notice the large number of grains having typical sizes smaller than 0.4 μm. This image was supplied by Dr Florence Lecouturier.

Both carrier concentrations $n_{\rm cond}(T)$ and $p_{\rm cond}(T)$ have strong temperature dependences, *decreasing* by a factor of about ten upon cooling from room temperature to liquid helium temperatures. We used the quadratic temperature dependences for the concentrations as suggested by Franket and Chu [17]; namely

$$\begin{aligned} n_{\rm cond}(T) &= n_{\rm cond}(10\ {\rm K}) + s_{\rm e}(T - 10\ {\rm K})^2, \\ p_{\rm cond}(T) &= p_{\rm cond}(10\ {\rm K}) + s_{\rm h}(T - 10\ {\rm K})^2, \end{aligned} \tag{6}$$

where $s_{\rm e}$ and $s_{\rm h}$ are prefactors or slopes derived from the calculated carrier concentrations found from using equation (5) at 10 and 292 K.

The effective mobilities, $\mu_{\rm e,eff}(T)$ and $\mu_{\rm h,eff}(T)$, are small in magnitude, limited mainly by the strong dominating boundary scattering at the crystallite surfaces; the mobilities increase weakly by only a factor of three upon cooling to liquid helium temperature, owing to the freezing out of phonon scattering within the grains. Since hole and electron mobility values are extracted both from the MR and $R_{\rm Hall}$ data taken in parallel fields and two additional values are obtained from the perpendicular field data at each temperature, we take the effective mobility value at each temperature to be $\mu_{\rm eff} \approx (2/3)\mu_\parallel + (1/3)\mu_\perp$. We also give a simple linear temperature dependence to each effective mobility to describe its weak increase with decreasing temperatures. As we will show, the fit to the resistivity data is very acceptable, using the above rough approximations and no adjustable fitting parameters.

3. Film preparation, characterization and experimental details

Measurements were made on a polycrystalline thick bismuth film of average thickness of 3420 Å fabricated by evaporating high-purity bismuth through a mask onto a cleaved mica sheet, maintained at room temperature. The average thickness, measured using a thickness monitor, is known to an accuracy of $\pm 5\%$. The evaporation source was a tantalum boat coated with silicon dioxide, and the evaporation rate varied between 40 and 50 Å s^{-1} in a rather poor vacuum of 4×10^{-6} Torr.

The unannealed sample was composed of very small grains. In figure 1, an optical microscope image shows the typical 'sandy' structure of the surface of a similar polycrystalline

3325 Å film. The scale length in figure 1 is 5 μm, indicated by the 'line ruler marking'. This image has been 'filtered' and greatly contrasted using Adobe Photoshop in order to clarify the size dimensions of the different grains. From this figure, we roughly estimate that about 60% of the volume was composed of grains having diameters between 0.15 and 0.3 μm. Another 35% of the volume had grains of diameters ranging from 0.3 to 0.4 μm. The remaining 5% had very few but larger diameter grains up to 1.5 μm. SEM images gave complementary results (not shown). Our granular structure is very different from that reported in the literature by Garcia *et al*, where their typical grain diameter of 1 μm was observed [18]. The majority of our grains had their $C3$ trigonal axis oriented perpendicular to the mica substrate, similar to that observed by Garcia *et al* [18].

Surface roughness measurements were made using the Alpha Step 200 stylus surface profiler of Tencor Instruments[3]. We were worried that the probing needle would 'dig' channels into the soft bismuth surface, but this was not the case. The roughness of the cleaved mica sheet was ± 50 Å. The average roughness of the film surface, including the mica substrate, was ± 115 Å. A few sections of the film were extremely smooth, displaying the same roughness of the mica sheet. A few other sections were extremely rough, displaying deviations of ± 390 Å (including the mica substrate roughness).

The resistance of the 3420 Å film was measured using a standard four wire technique with a 100 μA dc current. Electrical contacts to the film were made by attaching small copper wires using pressed indium contacts onto the patterned bismuth film. The width of the film was 1.57 mm. The indium contact pads at each end of the film were made wide to achieve homogeneous current injection and to avoid the anomalous longitudinal magnetoresistance contribution which Yoshida discusses [19]. The current tabs were located 3 mm away from each voltage tab. This film had a geometric factor $f_g = 13.4 \times 10^{-6}$ cm, used to convert resistances to resistivities. The room temperature resistivity was 390 $\mu\Omega$ cm, much higher than the $\approx$100–140 $\mu\Omega$ cm values for high-quality epitaxial films [7].

Measurements on the 3420 Å film were carried out either in a top loading Janis He^3 refrigerator, model HE-3-TLSL, equipped with a 17.5 T superconducting magnet, or in a 25 T Bitter resistive magnet at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee, Florida. Complementary data on other bismuth films were obtained at the pulsed magnet laboratory LNCMP in Toulouse, France.

4. Transport data

The MR and Hall voltage were measured:

(1) in the perpendicular $\perp$ geometry where the magnetic field is applied perpendicular to the mica substrate plane (parallel to the trigonal $C3$ axis and hence perpendicular to the current);
(2) in the parallel $\|$ geometry where the magnetic field is applied parallel to the substrate and also parallel to the current;
(3) in the transverse ($T_{\|,\perp}$) geometry where the field is applied parallel to the substrate but perpendicular to the current.

4.1. Experimental results in perpendicular fields

The behaviour of our polycrystalline 3420 Å bismuth film shows positive but small MR ratios, $r = R(B)/R(0)$, of the order of only a five-fold increase for fields oriented perpendicular ($\perp$)

[3] KLA-Tencor Instruments, 160 Rio Robles, San Jose, CA 95134, USA.

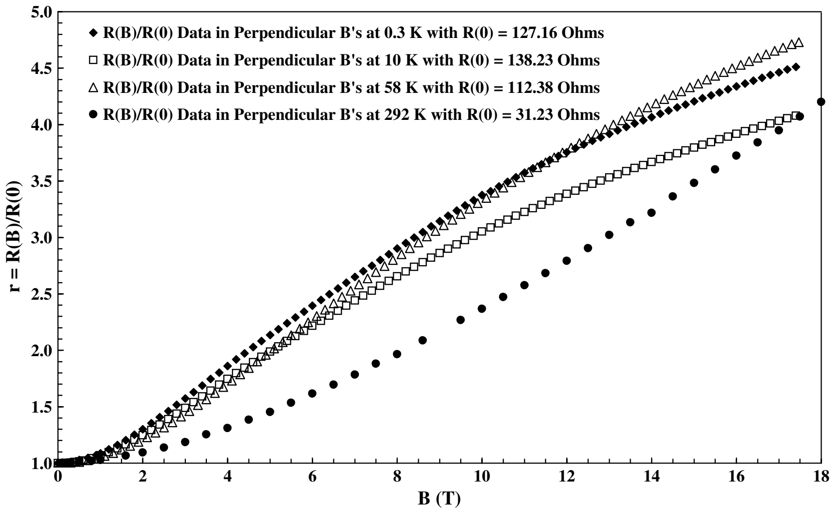

Figure 2. Magnetoresistance (MR) ratios in perpendicular fields at different temperatures for a 3420 Å polycrystalline bismuth film. These data are anomalous at high fields, displaying *small* MR ratios at low temperatures and no quadratic field dependences.

to the substrate, as illustrated in figure 2. There are no indications of Shubnikov–de Haas (SdH) oscillations in the 0.3 K data. Peschanskii and Sinolitskii point out that in a polycrystalline sample whose thickness is of the order of the dimensions of the crystallites, then the Shubnikov–de Haas oscillation amplitudes are much smaller owing to the averaging over the random orientations of all of the crystallites [20].

In figure 2, the surprising behaviour occurs in the high perpendicular field regime where the values of the MR ratios at liquid helium temperatures are slightly larger than those at room temperature. These 0.3 K results are in contrast to those measured on epitaxial films where the perpendicular magnetoresistances at 4.2 K are at least several orders of magnitude greater than those measured at room temperature [9, 10]. Perpendicular MR ratios of the order of 10 000 at 1.2 K have been reported by Babiskin in oriented single crystals, as seen in figure 3 of [21]. Even more striking are the perpendicular MR ratios of 200 000 observed at liquid helium temperature in single crystals of bismuth by Brandt *et al* [22]; these authors observed either saturation at 35 T or a 'maximum' followed by a decrease, a behaviour very similar to our results in parallel fields as discussed below.

Only at small fields below 2 T is a quadratic B law observed for the MR ratios r, as illustrated in figure 3; it appears that this film is uncompensated with $c_{\perp} \approx 0.36$. But at low fields the MR behaviour appears 'normal' since larger MR values are associated with lower measuring temperatures; this is due to the larger mobilities at the lowest temperatures that arise from longer scattering times (less carrier scattering from phonons). We are aware of only one theoretical paper by Franket and Chu who predict non-compensation or carrier ratios $c \neq 1$ [17]. These authors predict a *hole-majority* condition where $c > 1$; in contrast, we observe an *electron-majority* condition where $c < 1$.

The Hall coefficient data R_{Hall} in perpendicular fields are summarized in figure 4. Note that, at room temperature, the sign of R_{Hall} is positive in small fields. With increasing fields R_{Hall} changes signs and tends to saturate to a negative value at highest fields, as shown in figure 4. In thicker films, R_{Hall} is always negative [14, 15]. The positive sign can be achieved if the hole mobility is considerably larger than the electron mobility; this is a surprising results

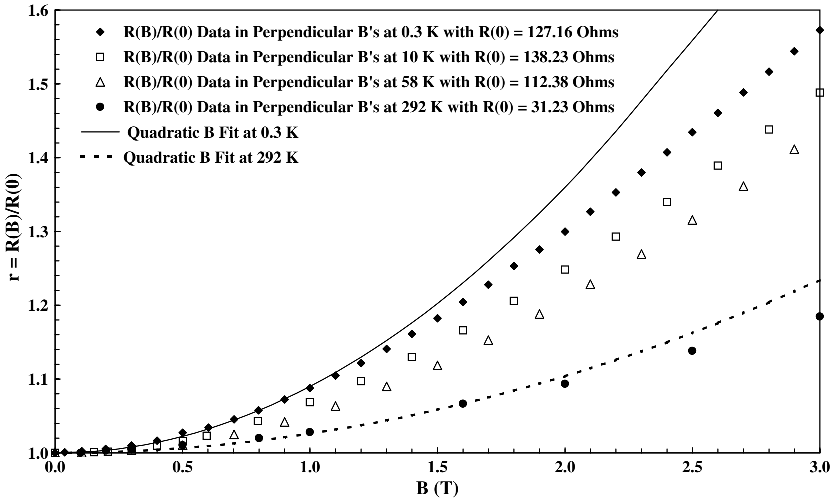

Figure 3. Low field MR ratios in perpendicular fields illustrating the very limited field regime where the MR ratios exhibit quadratic field dependences (solid and dashed curves). This behaviour suggests that this 3420 Å film is not compensated or $p/n \neq 1$.

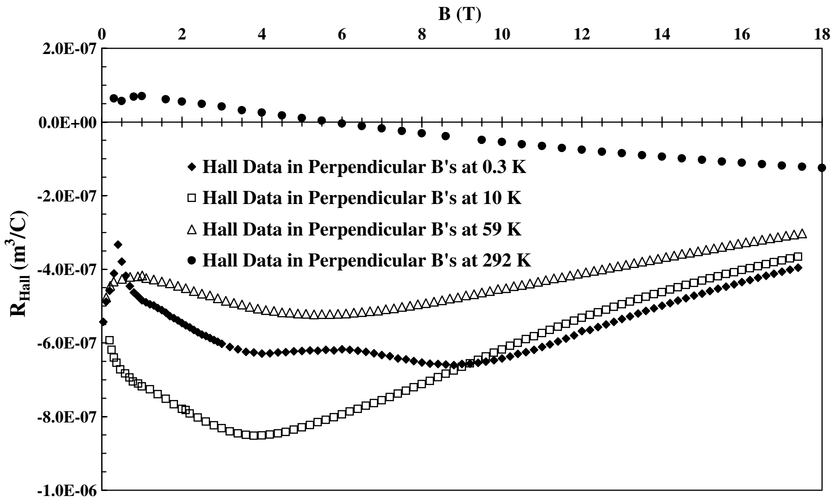

Figure 4. Hall coefficients R_{Hall} in perpendicular B's at different temperatures for the 3420 Å polycrystalline bismuth film. Note that the sign of the R_{Hall} is *positive* for small fields at room temperature. At lower temperatures R_{Hall} is negative, exhibiting negative maxima at intermediate fields, and decreases toward zero at high fields. There are also oscillations superimposed upon the 0.3, 10 and 59 K data.

since it is well known that, in much thicker films, the hole mobility is smaller than that of the electron owing to the smaller electron effective mass [17]. Good fits to the Hall coefficient data were possible only if $c_{\perp} < 1$. For $c_{\perp} > 1$, the fits exhibited the wrong field dependence, increasing to more positive values with the field, in contradiction to the behaviour of the data. At room temperature, R_{Hall} tends to saturate at high fields, in accordance to the Pippard–Fawcett prediction. Note that, at $B = 0$ T, the magnitude of R_{Hall} increases by almost one order of

Table 1. Fitting parameters for the *perpendicular* MR and R_{Hall} data using equations (1*a*) and (4*a*). Parameters are known to ±50%.

Temperature, T (K)	Electron mobility, μ_e ($m^2\ V^{-1}\ s^{-1}$)	Hole mobility, μ_h ($m^2\ V^{-1}\ s^{-1}$)	Carrier density ratio, $c_\perp = p/n$	Hall prefactor, $n = n_{Hall}$ (electrons m^{-3})
292	0.09	0.25	0.42	4.15×10^{25}
10	0.48	0.25	0.36	6.0×10^{24}

magnitude to more negative values as the temperature is lowered; this increase is caused by the decreases of the carrier numbers by about factors of ten upon cooling from room temperature to liquid helium temperatures. These decreases are also responsible for the increase of the zero field resistivity upon cooling to liquid helium temperatures.

In figure 4, the R_{Hall} data at 0.3 K shows some oscillations which are not periodic in B^{-1}, a signature of Shubnikov–de Haas oscillations [24, 25]. No oscillations were observed in the perpendicular magnetoresistance (MR) data at 0.3 K in figure 2.

The low field Hall coefficient data (below 2 T) in figure 4 exhibit an anomalous oscillatory behaviour at the relatively high temperatures of 10 and 59 K. This behaviour might be associated with the *high temperature oscillations* (HTOs) first observed by Bogod *et al* [26] in a bismuth crystal and further elaborated experimentally and theoretically by Krasovitsky [27]. Krasovitsky suggests that the oscillations result from electron–hole transitions between the Landau levels close to the Fermi level via the interaction with acoustic phonons [27]. At temperatures below 10 K, these oscillations become negligible owing to the freezing out of acoustic phonons. These oscillations are also periodic in B^{-1} [26].

Shubnikov–de Haas oscillations have been observed by Partin *et al* superimposed upon perpendicular Hall voltages in their epitaxial films [28]. The SdH oscillations also have been observed in high-quality 500 Å Bi films, and the results have been interpreted in terms of an increase of the density carrier [29].

Fits to both the perpendicular MR ratio data and R_{Hall} data at room temperature are shown in figure 5 and its insert using the same fitting parameters, namely $\mu_{e,\perp}$, $\mu_{h,\perp}$, and $c_\perp$. The fitting parameters are summarized in table 1. Only $n = n_{Hall}$ took on the anomalously high value of 4.2×10^{25} electrons m^{-3} compared to the value of $n = n_{cond} = 6.5 \times 10^{24}$ electrons m^{-3} extracted from the conductivity magnitude at room temperature. Partin *et al* found $n = n_{cond} \approx 4.0 \times 10^{24}$ electrons m^{-3} at room temperature for a 5000 Å film [28], a value similar to our magnitude. At 4.2 K, they found $n \sim p \approx (5 \pm 1) \times 10^{23}$ carriers m^{-3} at 4.2 K [28], again similar to our findings.

There is one unique behaviour of the Hall coefficient data at low temperatures and high fields. Note that in figure 4 there are maxima in the negative values around 6 T followed by decreasing R_{Hall} values at higher fields. The decrease is also seen in the 0.3 K data but the oscillations obscure the exact position of the maximum. The decrease at high fields is certainly not predicted by the Pippard–Fawcett model. This mechanism is probably responsible also for the small MR ratio magnitudes above 10 T as seen in figure 1 at 0.3 K. We will return to this topic in the next section.

4.2. Experimental results in parallel fields

The parallel MR ratios r are shown in figure 6 and are characterized by maxima or peaks, first reported by Andrievskii *et al* [30–32]. The magnitudes of the parallel MR are approximately one half the magnitudes of those in perpendicular fields; and hence the classical prediction of

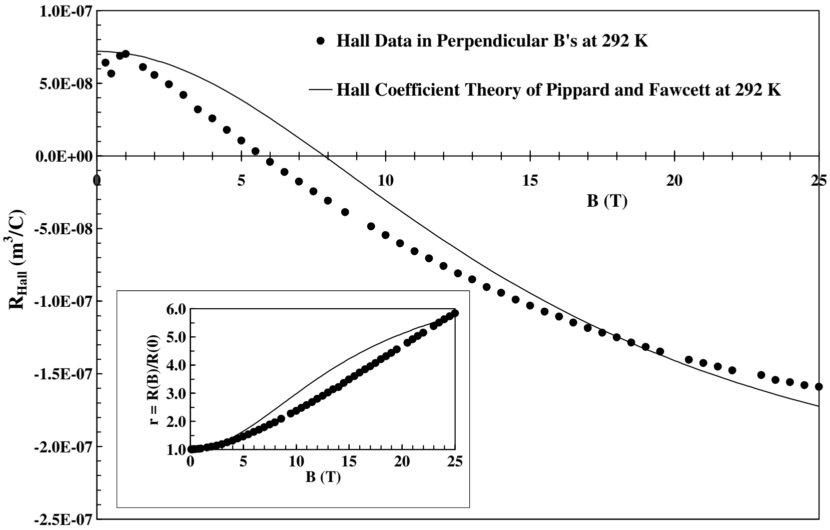

Figure 5. Fits using the Pippard–Fawcett two carrier expressions to the Hall coefficient data and to the MR ratio data (inset) taken in perpendicular B's at 292 K. The fits (solid curves) use the same hole and electron mobilities and $c_{\perp} = p/n$ values. The surprising result from the fits is that the hole mobility must be considerably larger than the electron mobility at 292 K in order to yield positive values for the Hall coefficient at small fields.

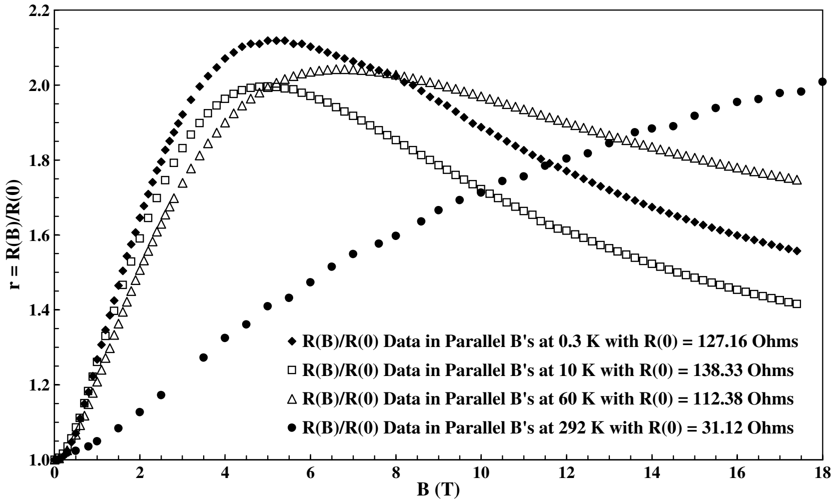

Figure 6. Magnetoresistance (MR) ratios in parallel fields at different temperatures for the 3420 Å polycrystalline bismuth film. These data are anomalous since classical theory predicts very small or no magnetoresistance. At intermediate fields, the polycrystalline bismuth film displays maxima followed by decreases. The room temperature data extrapolate to a maximum around 45 T.

no magnetoresistance in parallel fields fails badly for the polycrystalline samples. Babiskin observed parallel MR ratios in oriented single crystals whose magnitudes and behaviours with field (figures 4 and 5 in [21]) are very similar to the results in our figure 6. In complete contrast, no maximum was observed in the parallel MR of an epitaxial film by the Johns Hopkins University group [9, 10]. As seen in figure 6, the field where the 'maximum' occurs in the MR data, $B_{\parallel,\max}$, moves to larger values as the temperature is increased. If at room

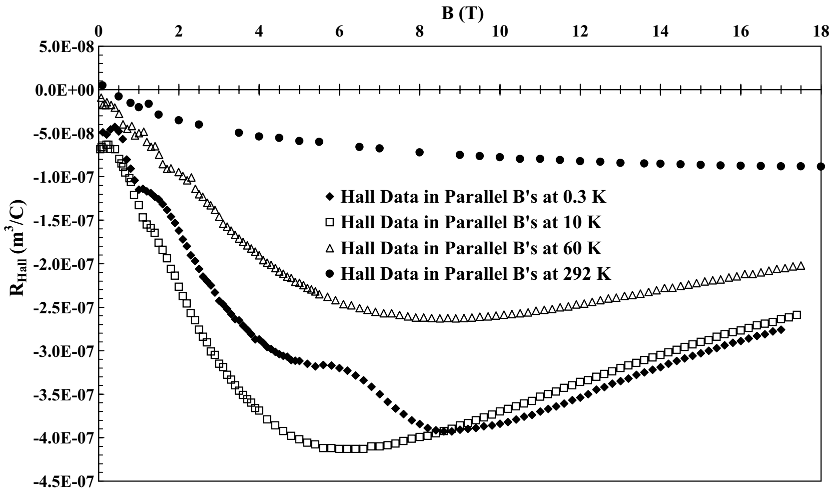

Figure 7. Hall coefficients R_{Hall} in parallel fields at different temperatures for the 3420 Å polycrystalline bismuth film. These data are anomalous since classical theory predicts no Hall voltages in parallel fields. Diffused and specular scattering of the carriers at the surfaces of the crystallites are most likely the mechanisms responsible for these large Hall voltage magnitudes. Oscillations are superimposed upon the 0.3, 10 and 60 K data. Note the strong decreases of R_{Hall} at fields greater than $B \approx 8$ T.

temperature a very weak maximum is observed in the 35–45 T interval, then at liquid helium temperatures, the maximum typically occurs between 4 and 6 T. The decrease of the ratio values above the maximum follows nicely a $B^{-1/2}$ law. There are very weak oscillations superimposed upon the parallel MR ratio data measured at 0.3 K.

A similar 'maximum' behaviour is observed in the parallel Hall coefficient data, shown in figure 7. Notice that the negative maxima in the Hall data occur at slightly higher field values (between 6 and 9 T) as compared to those of the MR maxima in the MR data of figure 5. As the temperature is increased, these maxima also occur at higher fields. Note that the actual Hall voltages in parallel fields are extremely large, of the order of one millivolt using small currents of 100 μA at 5 T; thus R_{Hall} is easily measured even in small fields. Our results are contrasted by the small parallel Hall voltages of an oriented single crystal; Babiskin observed voltages of $-30\ \mu$V, using a large current of 2 A at 5 T, as seen in figure 6 of [21]. Unfortunately he could not reverse the field direction to eliminate the voltage contribution from misalignment of his Hall probes. We estimate his Hall coefficient value to be about $R_{\mathrm{Hall}} \approx -1.5 \times 10^{-8}$ m^3 C^{-1} at 4.2 K, a factor 30 times smaller than our observed values in figure 7. Thus, the polycrystalline nature of our film plays the major role in creating the large magnitudes of the parallel Hall voltages.

Both the parallel Hall coefficient data and MR ratio data taken at room temperature can be fitted consistently using the Pippard–Fawcett expressions as shown in figure 8 and its inset. The fitting parameters are given in table 2. Again it was necessary to make $n = n_{\mathrm{Hall}}$ a free fitting parameter, scaling it to the large value of 7.5×10^{25} electrons m^{-3}. The fits are acceptable. Note the small value for c_{eff} of 0.18.

The parallel Hall coefficient data at 0.3 K are superimposed with oscillations, as seen in figure 7; this is a surprising result. To ascertain if the oscillations are Shubnikov–de Haas (SdH) oscillations, we plot the oscillation contribution versus B^{-1}. We have subtracted off the monotonic background arising from the two carrier Hall contribution by using the

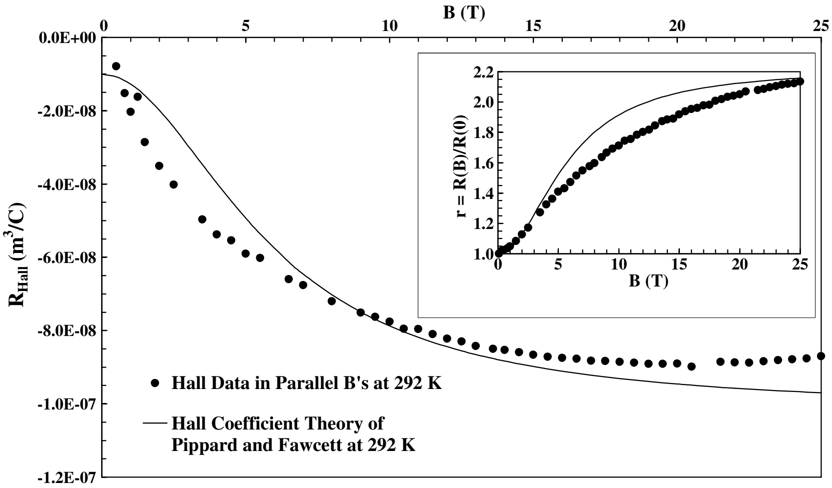

Figure 8. Fits using the Pippard–Fawcett two carrier expressions to the parallel Hall coefficient data and to the MR ratio data (inset) at 292 K. The fits (solid curves) use the same hole and electron mobilities and c_{eff} parameter. The surprising result from the fits is that the hole mobility is considerably larger than the electron mobility, giving rise to the very small magnitudes of the Hall coefficient at small fields.

Table 2. Fitting parameters for the *parallel* MR and R_{Hall} data using equations (1*a*) and (4*a*). Parameters are known to ±50%.

Temperature, T (K)	Electron mobility, μ_{e} ($\mathrm{m^2\,V^{-1}\,s^{-1}}$)	Hole mobility, μ_{h} ($\mathrm{m^2\,V^{-1}\,s^{-1}}$)	Carrier density ratio, $c_{\mathrm{eff}} = (p/n)f_{\mathrm{s}}$	Hall prefactor, $n = n_{\mathrm{Hall}}$ (electrons $\mathrm{m^{-3}}$)
292	0.14	0.29	0.18	7.5×10^{25}
10	0.45	0.90	0.18	1.75×10^{25}

Pippard–Fawcett equation (4*a*). The results of the subtraction procedure are shown in figure 9, where clearly the oscillations are not periodic in B^{-1}. However, if the oscillations are SdH, then they should be difficult to observe at temperatures greater than the Dingle temperature T_{D} owing to 'smearing'. Here $T_{\mathrm{D}} = h/(2\pi k_{\mathrm{B}}\tau_{\mathrm{e}}) \approx 15$ K [25], where τ_{e} is the electron mean-free time, estimated to be 8×10^{-14} s using the electron mobility $\mu_{\mathrm{e}} \approx 0.7\ \mathrm{m^2\,V^{-1}\,s^{-1}}$ and $m^*_{\mathrm{eff}} \approx 0.02m_{\mathrm{e}}$. This behaviour is confirmed by comparing the Hall data at 0.3 and 10 K in figure 7. We speculate that there is some alignment correlation between neighbouring crystallites, tending to cause the alignment orientations of the three electron pockets in one grain to be similar to the alignments in its nearest neighbours. Thus we suggest that there are clusters of similarly aligned grains in our films. The oscillations of figure 9 might then be the combined contributions of SdH patterns from numerous different clusters in the film; otherwise no oscillations should have been observed according to Peschanskii and Sinolitskii for completely random orientations [20]. It will be useful to extend the parallel Hall data to lower temperatures to observe if the magnitudes of the oscillations continue to increase.

4.3. Experimental results in transverse fields

There are very few publications devoted to the magnetoresistance measured in the transverse field orientation in which the magnetic field is aligned perpendicular to the current flow but

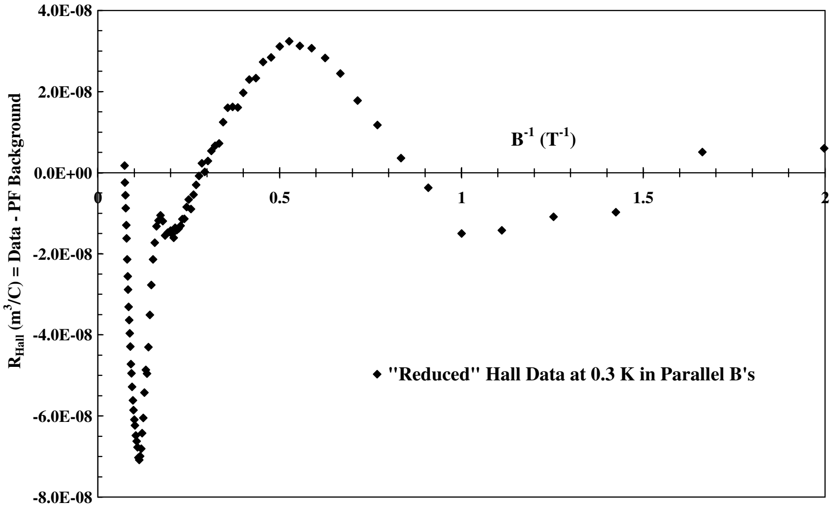

Figure 9. 'Reduced' parallel Hall coefficient data at 0.3 K plotted versus B^{-1}. The monotonic background has been subtracted off (using the Pippard–Fawcett expression) to better clarify the oscillatory contribution. The oscillations are not periodic in B^{-1}, as would be anticipated for Shubnikov–de Haas oscillations.

parallel to the film surface (hence, parallel to the direction of the two oppositely patterned Hall voltage tabs). The Johns Hopkins University group has shown that the MR in this transverse orientation has a field dependence very similar to the perpendicular MR, except that the magnitudes of their transverse MRs are about four times smaller in value [24]. Their measurements were made on thick, high-quality epitaxial films. In this transverse case, the Lorentz force directs the carriers either towards the bismuth–air interface, namely to the top surface of the film, or towards the bismuth–mica interface, namely to the bottom surface of the film.

Thus, we were surprised to observe 'kinks' in the transverse magnetoresistance data, occurring at low fields on this *thin* 3420 Å bismuth film, shown in figure 10. The behaviour is much different from the MR behaviour in the perpendicular orientation and also in the parallel orientation, as contrasted in figures 2 and 6. Firstly, the 'kinks' occur at the small field B_k of about 2.7 T, compared to the larger field value of 4.8 T associated with the 'maximum' in the parallel MR at 4.2 K (see figure 6). Secondly, this field B_k is almost temperature independent, increasing weakly at high temperatures as seen in figure 10; this is in contrast to the strong temperature dependence of $B_{\|,\max}$ in the parallel orientation.

We speculate that the carriers undergo strong diffused scattering at both surfaces. The 'kink' behaviour is reminiscent of the decrease observed by Huber's group on bismuth wires; refer to their figure 4 in [33]. These authors contribute the decrease to a decreasing cyclotron orbital radius of the carrier, resulting from an increasing field. At a sufficiently strong field B_k, the carriers will no longer collide with the surface interface of the wire; thus, there is less scattering of the carriers and fewer losses; and hence the resistance decreases according to the theories of Chambers [34], Kao [12], and Way and Kao [35]. We refer to this explanation as the 'cyclotron radius boundary scattering' model. Using the data of figure 10, we have fitted the low field MR below the 'kink' by using only the Pippard–Fawcett formalism; and at fields greater than B_k above the kink, we have included an additional conductivity ratio term. The fitting details of our phenomenological model are given in [36], where acceptable

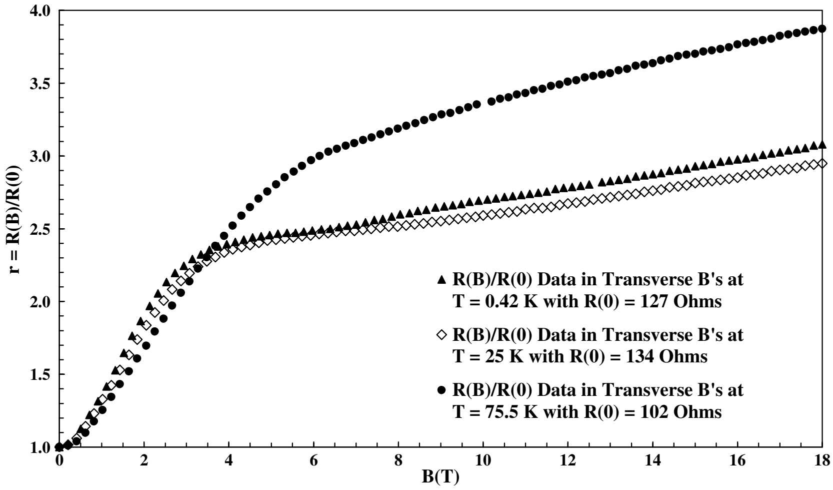

Figure 10. Magnetoresistance (MR) ratios in transverse fields at different temperatures for the 3420 Å polycrystalline bismuth film. These data are anomalous, exhibiting 'kinks' at low fields. The 'kinks' most likely arise from decreasing diffused boundary scattering and hence decreasing losses, as the cyclotron radii of the carriers are reduced with increasing field.

Table 3. Fitting parameters used in predicting the zero field resistivity data (solid curve in figure 10) using equations (5) and (6).

T (K)	σ $(1/\Omega\,\mathrm{m})$	$n = n_{\mathrm{cond}}$ (electrons m^{-3})	$p = p_{\mathrm{cond}}$ (holes m^{-3})	$\mu_{\mathrm{h,ave}}$ ($\mathrm{m}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$)	$\mu_{\mathrm{e,ave}}$ ($\mathrm{m}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$)	c
292	248 760	6.50×10^{24}	2.73×10^{24}	0.12	0.28	0.42
10	58 080	5.17×10^{23}	1.84×10^{23}	0.46	0.68	0.36

fits are obtained. Additional transverse MR data for films having different thicknesses are also included in [36].

We believe that this boundary scattering scenario explains nicely the unusual behaviour observed in the transverse MR data.

5. Zero field resistivity data

Figure 11 shows the resistivity data for the 3420 Å film, where the resistivity increases by a factor of four upon cooling to liquid helium temperatures. This is a typical behaviour for a polycrystalline bismuth film, where the strong decreases in the electron and hole carrier numbers dominate over the small increases in their mobilities. In contrast, the resistivity decreases for an epitaxial film upon cooling [7]. The solid curve in figure 11 is a fit using equations (5) and (6) and the fitting parameters of table 3. The fit is good.

6. Unanswered questions and conclusions

The two carrier model of Pippard and Fawcett and the Drude expression give acceptable fits to most of the transport data. However, the fitting results raise numerous unexplained questions relating to the physical processes in these very thin films. Here are some of the questions that need theoretical explanations and answers:

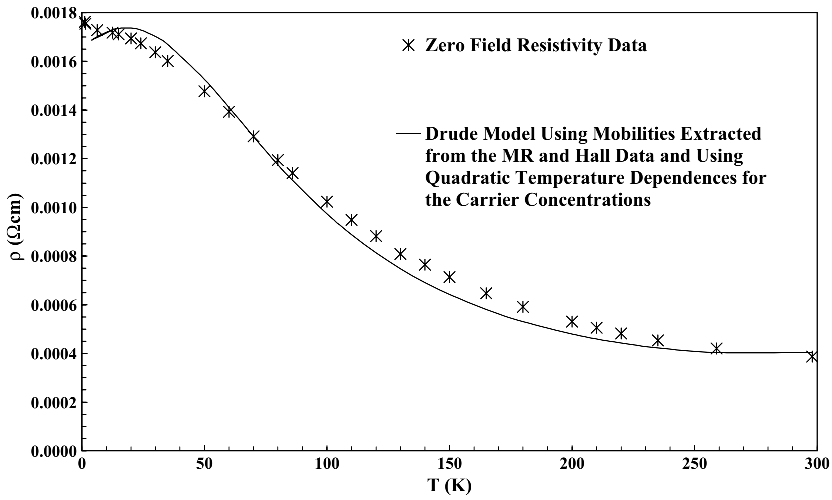

Figure 11. Zero field resistivity ρ of the 3420 Å polycrystalline bismuth film. The four-fold increase of ρ between room temperature and liquid helium temperature is typical for a polycrystalline bismuth film; this increase arises mainly from ten-fold decreases of the electron and hole number concentrations upon cooling to liquid helium temperatures. The solid curve is a fit using the Drude expression.

(a) What is a simple argument that will explain the ten-fold *decreases* of the carrier numbers in bismuth upon cooling to liquid helium temperatures?
(b) What are the forms of the Fermi surfaces of the single hole pocket and of the three electron pockets in these thin films (at least thinner than 7000 Å) that would give rise to hole mobilities that are larger than the electron mobilities?
(c) Why are these thin films uncompensated?
(d) What is the origin of the oscillations that are superimposed upon the low temperature Hall coefficient data?
(e) What is a satisfactory model for explaining the decreasing MR ratios and Hall coefficients observed in high *parallel* fields and at low temperatures?
(f) Why must one scale up the prefactor appearing in front of the Hall coefficient equation of Pippard and Fawcett, namely the electron concentration $n = n_{\mathrm{Hall}}$, by more that one order of magnitude compared to the magnitude for $n = n_{\mathrm{cond}}$ extracted from the Drude conductivity expression?
(g) What is the reason for the very small magnetoresistance values in high perpendicular fields at low temperatures?

We summarize the experimental findings of the MR and Hall coefficient data on this 3420 Å polycrystalline bismuth film.

(1) Perpendicular field orientation.

The high field MR is anomalous and exhibits a small MR ratio at low temperatures; no SdH oscillations are observed superimposed upon the MR data.

R_{Hall} is positive for vanishing fields but negative at high fields at 292 K; the conclusion from fitting the data is that $\mu_{\mathrm{h}} > \mu_{\mathrm{e}}$. Oscillations are observed in the low temperature R_{Hall} data. R_{Hall} exhibits negative maxima at intermediate fields.

(2) Parallel field orientation.

Both the MR and Hall voltage magnitudes are anomalously large at all temperatures. The conclusion is that the surfaces of the crystallites act as scattering barriers. Maxima are observed in both the parallel MR and R_{Hall} data at low temperatures and intermediate fields. Oscillations are observed in the low temperature R_{Hall} and MR data. Fitting the data suggests again that $\mu_{\mathrm{h}} > \mu_{\mathrm{e}}$.

(3) Transverse field orientation.

The MR data exhibit 'kinks' at low B_{s}. The presence of such 'kinks' is a signature of a decrease of the boundary scattering due to the shrinkage of the cyclotron radii of the carriers. This behaviour again suggests that the surfaces of the crystallites act as scattering barriers.

Hence, there is still much theoretical work and experimental work remaining to clarify the physics of this complicated and challenging system. For example, it might be advisable to make experiments of the conductivity σ using the Corbino disc geometry, with B either perpendicular or parallel to the disc; in this case there is no Hall voltage contribution to the resistance voltage.

The appendix covers a discussion of several phenomenological models that might shed some light on the physical processes involved.

Acknowledgments

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Appendix. Phenomenological models that might explain the anomalous transport behaviours at high fields

Is there an explanation for the decreases of the parallel MR ratio values and Hall coefficient magnitudes that are observed in large parallel and perpendicular fields at low temperatures? We now consider several phenomenological models.

In an important experimental paper on the properties of epitaxial bismuth films, Partin *et al* demonstrated Shubnikov–de Haas oscillations [28]; and they mention: 'At higher magnetic fields, the oscillations no longer are periodic in $1/B$, because the carrier density changes, becoming, in fact, a linear function of field when the electrons are all on the lowest Landau level' [28, 37]. Unfortunately reference [37] gives only indirect details on the derivation of

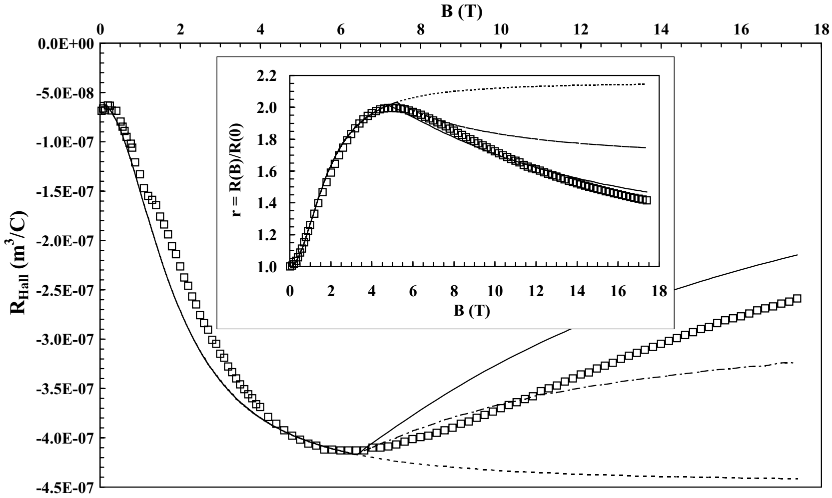

Figure A.1. Phenomenological fits to the parallel Hall coefficient and MR ratio data (inset) at 10 K. The dotted curves are derived using the Pippard–Fawcett expressions where the mobilities and carrier numbers are kept field independent and constant. The dashed–dotted curves are based using mobilities that have an additional B^{-1} term associated with a field dependent effective mass; the carrier numbers are kept constant. The solid curves are based upon an electron concentration that increases linearly with magnetic field, causing decreases in c_{eff}; the mobilities are kept constant.

the linear field dependence of n. Likewise, Smith *et al* [38] argue that 'as the field increases leaving only one Landau beneath the Fermi surface, the number of electrons which fit into this single Landau level increases and this increase tends to pull the Fermi level down and changes the number of carriers in the various electron and hole pockets'. In figure 10 of Smith's article [38], the hole and the electron densities do not increase linearly with B but more closely as $B^{1/2}$, similarly to the $B^{-1/2}$ dependence observed in the decreasing region of our parallel MR behaviours. But we will now adapt Partin's claim of a linear field dependence on n [28], and we will assume that p is field independent. We assign the following simple field dependence to the electron number concentration:

$$n(B) = n(0)[1 + a(B - B_{\text{max}})], \qquad \text{for } B > B_{\text{max}}, \tag{7}$$

where a is an adjustable fitting parameter. This dependence will have important consequences on the fits since $c = p/n = p/n(B)$ will decrease at high fields; in addition, recall that the Hall coefficient also has the $n = n_{\text{Hall}}(B)$ prefactor in the denominator of equation (4a), thus producing a significant decrease of R_{Hall} in high fields. The fits to the parallel Hall coefficient and MR data at 10 K are shown in figure A.1 and in its inset by the solid curves. The fits are surprisingly good. The adjustable fitting parameter, a, was set to 0.08 in both fits; both fits use the same parallel mobilities and c_{eff} values, given in table 2.

There is an alternative model. We recall that the Pippard–Fawcett expressions all involve mobilities that scale inversely with the effective mass of the carrier, where $\mu = (e\tau_{\text{eff}}/m^*_{\text{eff}})$. In an important experimental paper, Takano and Kawamura measured the interferences and the damping of Alfen waves in single crystal ingot of bismuth and extracted the *mass densities* for different field orientations and electric field polarizations [39]. These investigators defined the mass density as the product of the carrier number and effective mass, nm^*_{e} or pm^*_{h}. Their significant observation was that, for fields greater than 3 T, the effective mass density increased linearly and monotonically at high fields; in some cases, the increase was ten-fold. These changes are ascribed to the quantum variation of the Fermi energy at high magnetic fields. For fields less than 3 T, the mass density was a constant [39].

We use the following phenomenological model based upon Takano and Kawamura's results [39]. We make the extreme assumption that the electron number concentration n is now *constant* (in contrast to the first model); and now the masses of the electrons and holes both have strong magnetic field dependences. For fields below the 'maxima' in the Hall and MR data, we keep the effective masses constant; and hence the effective mobilities will be constant, independent of field. We use the mobility values found from the fits to the parallel low field MR and Hall data, which are listed in table 2. At fields above the 'maxima' we add a B^{-1} contribution to reflect the linear increases of the masses and hence the decreases of the mobilities; namely

$$\mu_{\text{eff}}(B) = \mu_{\text{eff}}(0)/[1 + s(B - B_{\|,\max})], \qquad \text{for } B > B_{\|,\max}, \tag{8}$$

where s is a fitting parameter. The fits to the parallel 10 K Hall and MR data using the Pippard–Fawcett model with the field dependent mobilities are shown by the dashed–dotted curves in figure A.1 and its inset, using $s = 0.5$ (T^{-1}); the fits are acceptable.

Fits excluding any field dependences of either the mobilities or of the electron number concentration are also shown by the dotted curves in figure A.1 and its inset. There are no decreases in this case. The fitting parameters are summarized in table 2.

The fitting results of these models suggest that the magnetic field dependence of both the carrier numbers and masses should be considered in explaining the decreases; also, the 'cyclotron radius boundary scattering' model should be included. A more rigorous transport model in the quantum limit is certainly needed to put these 'decreases' on firmer theoretical grounds. It is likely that the same mechanism(s) are responsible for the anomalously small perpendicular MR ratios observed at low temperatures in figure 2.

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