Anomalous amplitude of the quantum oscillations in the longitudinal magneto-thermoelectric power

N Satoh
Department of Electronics and Computer Science, Iwaki Maisie University, 5-5-1 Iino chuoudai, Iwaki, Fukushima 970-8551, Japan
E-mail: nsatoh10015@cg8.so-net.ne.jp

Abstract. Longitudinal magneto-thermoelectric power $S_y(y)$ of a pure bismuth single crystal was measured in magnetic fields up to 8T at several fixed temperatures between 1.4 and 15 K to investigate the magneto-phonon effect in the longitudinal magneto-thermoelectric power (MTP). The oscillation patterns of the longitudinal MTP was similar to that of the longitudinal Shubnikov-de Haas (SdH) effect, expectedly. However, the observed amplitude of oscillations showed a curious temperature dependence. That is, in the range of temperature $T > 4.2$ K, the amplitude has a maximum around 9K, which is well described by considering the inter-Landau level scattering of electrons. On the contrary, in the range of $T < 4.2$K, the observed amplitude is enhanced markedly although that of the longitudinal SdH oscillations becomes less pronounced by lowering temperature. This discrepancy may be attributed to the effect of the surface (wrapping) current and to the energy dependence of the electron relaxation time.

1. Introduction
It is well known that bismuth has frequently played an important pioneer-role in the history of solid state physics. Fundamental phenomena such as de Haas-Alphen effect, Shubnikov-de Haas (SdH) effect, giant magnetoresistance and oscillatory magnetostriction as well as Nernst effect were all first observed in bismuth [1]. Bismuth is the archetypical semimetal and is still attracting new attention owing to its unique properties, following the development of study on the Dirac Fermions in bismuth-based of topological insulators [2]. Nevertheless, there is no doubt that several fundamental problems concerning the transport phenomena in bulk bismuth remain still unsolved. One of them, for instance, is such that what brings about so large amplitude in the SdH oscillations. Also, we do not know exactly the reason why magnetic field can markedly enhance the resistivity by a factor of $10^6$ without saturation. We have been investigating the longitudinal SdH oscillations of semimetal, especially diffusion size effect, a small amount of charged (neutral) impurity effect [3-5] and surface recombination effect on the SdH oscillation amplitudes [6] to find a key to the unsolved problems.

We present here our study on the longitudinal magneto-thermoelectric power (MTP) $S_y(y)$ in bismuth as an extension of our investigation. Although studies on the ordinary Seebeck effect of bismuth in strong magnetic fields were previously performed decades ago [7], but our experiment would serve as a good testing ground to investigate how the inter- and intra-Landau level scatterings, the so-called “magneto-phonon effect”, can play a role in the longitudinal MTP oscillations.
2. Experimental
Brief description of our sample preparation is as follows. A piece of commercially available pure bismuth ingot (6N grade) was purified further by zone-melting, and was finally single crystallized. The obtained single crystal was cut with an electric discharge cutting machine along the bisectrix (y) axis of bismuth into a rectangle shape (3.0 × 3.0 × 18 mm³) after the crystal orientation was checked by X-ray diffraction analysis. The sample was annealed isothermally at 265°C for two weeks to remove strain. In our experimental configuration, both the temperature gradient ΨT and the magnetic field B were parallel to the bisectrix axis of the sample. Thermoelectric voltages were detected in a conventional way using a thermocouple (0.07 at % Fe-Au verse Ag) that had extremely low magnetic-field-induced-error (less than 0.1% at 9T). Temperature was measured by a calibrated ceramic thin film resistance thermometer.

3. Result and discussions
Figure 1 shows our typical results of the oscillatory longitudinal MTP for bismuth single crystal along bisectrix axis as a function of inverse magnetic field at several fixed temperatures of 1.4K, 4.2K, 5.6K and 7.5K, respectively. Originally, the thermoelectric power $S_{yy}(y)$ is defined in terms of the thermal diffusion coefficient $A$ and the conductivity tensor $\sigma_{yy}$ as $S_{yy} = A/\sigma_{yy}$. Then, as would be expected, SdH-like oscillations certainly appeared in our measurements. In fact, it is clear by visual inspection that the oscillation patterns in figure 1 are the same as our previous work [3-5]. For instance, a series of pronounced oscillation peaks were observed at the same magnetic fields of 1.25, 0.625, 0.42 T.

![Figure 1. Typical magneto-thermopower $S_{yy}$ as a function of inverse magnetic field $B^{-1}$ at 1.4, 4.2, 5.6 and 7.5 K. Quantum number of the peaks indicated by arrows are electron levels assigned by Smith, et.al. [8]](image)

Secondly, the temperature dependence of the oscillation amplitude (figure 1) seems to be unusual. To visualize this anomaly more clearly, we have picked up oscillation amplitudes $A_{osc}$ at a magnetic field of 1.25T and shown them in figure 2 as a function of temperature. Inset (b) of figure 2 illustrates the definition of the $A_{osc}$; this arises only from a piece of electron pockets located at point $L$ in the trigonal-bisectrix plane of the Brillouin zone. Our method of extracting $A_{osc}$ from those peaks is the same as that of Tanuma and Inada [9] and our previous works. Figure 2 certainly shows that the observed $A_{osc}$ becomes larger in magnitude and has a maximum around 9 K above 4.2 K. But on the other hand,
below 4.2 K, the $A_{osc}$ enhances markedly although the amplitude of the longitudinal SdH oscillation become less pronounced by lowering temperature [3]. In the present paper, attention will be paid mainly to this anomalous temperature dependence of the amplitude.

Quantum oscillations in the MTP are treated differently in the literature. According to Young [10] and Fletcher [11], the oscillatory MTP depends on the derivative of the density of states at the Fermi level. Also, Pantsulaya and Varlamov [12] calculated the oscillatory longitudinal MTP considering the energy dependence of the electron relaxation time. The amplitude $A_{osc}$ of oscillation is given by the derivative of the Lifshiz-Kosevich formula as follows,

$$A_{osc}(T) \propto (X \coth X - 1)/\sinh X,$$

where

$$X = 2\pi^2 r k_B T/\hbar \omega_c.$$  

In equation (1), $\omega_c (=eB/m_e)$ is the cyclotron frequency, $m_e$ the cyclotron effective mass, $r$ the number of harmonics. It is difficult to describe the whole temperature dependence (figure 2) only by equation (1). To understand this curious temperature dependence, we separate, for convenience, the observed data in figure 2 into two regions of temperature. One is upper branch ($T > 4.2$ K) and the other is lower branch ($T < 4.2$ K).

3.1. Upper branch ($T > 4.2$ K)

We shall, for the moment, confine ourselves to the amplitude $A_{osc}$ at a magnetic field of 1.25 T above 4.2 K. It is established that the longitudinal MTP oscillation is mostly explained by inter-Landau level scattering of electrons just as the longitudinal SdH oscillation. That is, the thermally excited electrons diffusing along bisectrix axis are scattered by excited phonons, which cause the large amplitude. Such inter-Landau level scattering of electrons is assisted by longitudinal acoustic phonons illustrated an arrow $q$ in figure 3(a), because of the momentum conservation law. The magnitude of wave number $q$ of phonons is given by a distance between two Landau tubes, i.e., $(0,-)$ and $(0,1,-)$, which remain in a piece of electron pocket at point $L$. Those phonons increase in number with rising temperature. Therefore, as pointed out by Tanuma and Inada [9], we need to put a proportional factor on equation (1): the excitation $\omega_q^2 N_q$ of phonons only having the wave number $q$ where $N_q$ is Planck’s function, $\omega_q$ the frequency of phonons. Also, the Dingle factors $e^{-\delta}$ is put to express Landau level broadening. Then, the amplitude $A_{osc}$ should be given as follows,

$$A_{osc}(T) = RB^{1/2}[(X \coth X - 1)/\sinh X] \omega_q^2 N_q e^{-\delta},$$

where

$$N_q = \{\exp(h \omega_q/k_B T) - 1\}^{-1},$$

$$\delta = \frac{2\pi^2 r k_B}{\hbar \omega_c} (T_D + T).$$

In equation (2), $R$ is an adjustable constant and $T_D$ denotes the Dingle Temperature. The wave number $q$ (figure 3(a)), the corresponding values of the frequency $\omega_q$ and the energy $h \omega_q$ of phonons are given to be $8.46 \times 10^4$ m$^{-1}$, $2.17 \times 10^{12}$ s$^{-1}$ and 1.43 meV, respectively [9]. By fitting equation (2) on the experimentally observed amplitudes ($T > 4.2$K in figure 2) using the Levenberg-Marquardt algorithm, the cyclotron frequency $\omega_c$ is obtained to be $1.21 \times 10^{13}$ s$^{-1}$. Then, the cyclotron effective mass $m_c$ is to be 0.0180 $m_0$ where $m_0$ is the electron rest mass. This value is almost 9.1% larger than the value of 0.0165 $m_0$, which is the cyclotron effective mass of heavy bisectrix and binary electrons [13]. This large mass can be understood as a measure of electron-phonon interaction (phonon drag) in the concerned range of temperature. Dingle temperature $T_D$ obtained from the so-called “Dingle plot” is to be 1.2 K. Then, the relaxation time $\tau (= \hbar/2\pi k_B T_B$) is to be $6.7 \times 10^{-11}$ s. The fact that the observed
amplitudes agree with equation (2) excellently is conclusive evidence that inter-level scatterings by excited phonons play a dominant role in the oscillatory longitudinal MTP above 4.2 K (figure 2).

![Figure 3](image)

**Figure 3.** (a) Electron Fermi surface at 1.25 T being parallel to bisectrix axis. \(q\) and \(q'\) are wave number vector for inter- and intra-level transition, respectively. (b) Schematic drawing of the cyclotron orbits of carriers, of which centre migrate toward the sample boundary by means of scattering.

3.2. Lower branch (\(T < 4.2\) K)

To the best of our knowledge, no reports have been made such enhanced data as our amplitude in the oscillatory longitudinal MTP below 4.2 K, especially in bismuth. To begin with, the MTP must vanish at \(T = 0\) K, because of entropy flow by carriers. Then, a local maximum of the amplitude will surely appear at a certain temperature \(T_{\text{max}}\) below our lowest data (1.4 K). Equation (1) will show a maximum for \(X = 1.62\) [10], i.e., at

\[
T_{\text{max}} = 0.110 B / rm_c. \tag{3}
\]

We have fitted equation (1) on the observed amplitudes to determine the cyclotron effective mass for the lower branch \((T < 4.2 \) K). The obtained mass \(m_c\) is to be 0.187\(m_0\), which is an order of magnitude heavier than that of the upper branch. A heavy mass may suggest the metallic surface state \((\approx 0.2m_0)\) of bismuth [14]. If we employ the following values: \(B = 1.25\) T, \(r = 1\), and \(m_c = 0.187m_0\) in equation (3), the value of \(T_{\text{max}}\) is to be 0.74 K, which is a reasonable one as shown in figure 2.

To carry out numerical simulation for the \(A_{\text{osc}}\) in full range of temperature, we have apparently obtained the following expression of \(A_{\text{osc}}(T)\) from combing equation (1) with (2) as

\[
A_{\text{osc}}(T) = RB^{1/2}[(X\coth X - 1)/\sinh X][1 + a_0^2 N q e^{-\delta}] \tag{4}
\]

The resultant simulated curve is drawn in the inset (a) of figure 2. In calculation, a weighted effective mass \(pm_0\) (\(p\): parameter) was assumed; that is, electron-phonon interaction changes only the mass.

Although the temperature dependencies of the experimentally observed amplitudes are in good agreement with equation (1) and (2) in each temperature range, but we do not fully understand yet what is essence. Some other process may play a role below 4.2 K. So, we will consider the motion of electrons semi-classically as follows.

Now, figure 3(b) shows a projection of the sample (x-z plane) schematically. In a strong magnetic field, as is well known, the centre of cyclotron orbits of each carrier migrates toward sample boundary after colliding with scatterers such as phonons or impurities. Such a scattering is depicted by an arrow \(q'\) in figure 3(a). Consequently, there will be a density and a steady-state electric potential difference along the bisectrix axis and the other axes as well, because no current can flow. Moreover, near the boundaries the trajectories of electrons are skipping orbits by means of elastic reflection from the surface: there exists surface (wrapping) current. The increased spatial confinement at the boundary has to entail a strong enhancement of the energy of the Landau levels at the surface: metallic surface state.
In our case of extremely pure bismuth, the magnitude of wave number \( q' \) (figure 3(a)) and the frequency \( \omega_q' \) of phonons must satisfy the inequalities of \( q' < 1.07 \times 10^6 \ \text{cm}^{-1} \) and \( \omega_q' < 2.9 \times 10^{11} \ \text{s}^{-1} \), respectively. Those values are obtained from the average radii of an electron pocket at point \( L \) in the Brillouin zone [15] and the average sound velocities as \( 2.4 \times 10^5 \ \text{cm/s} \). Therefore, a high temperature approximation \( \hbar \omega_q' < k_B T \) holds at temperature below 4.2 K in our experiment. Then, according to the work of Roth and Argyres [16], a relaxation time \( \tau(\varepsilon) \) for acoustic phonon scattering is expressed as:

\[
\frac{1}{\tau(\varepsilon)} = (2\pi E^2 k_B T / \hbar \mu v^2) D(\varepsilon),
\]  

(5)

where \( E \) is the deformation potential, \( \mu \) the mass density, \( v \) the sound velocity, \( D(\varepsilon) \) the density of state in the presence of magnetic field. Since electrons exhibit long relaxation time by lowering temperature (equation (5)), there would arise an increase in diamagnetic surface (wrapping) current that gives a correction to the thermal diffusion coefficient \( A \) in a quantizing magnetic field [17]. Also, the fact that the observed amplitude data are in good agreement with equation (1) provides strong evidence that the energy dependence of the electron relaxation time brings about giant oscillations of the longitudinal MTP below 4.2 K [12]. But, the energy dependence of \( \tau(\varepsilon) \) is not well understood [18].

In conclusion, the observed curious temperature dependence of amplitude in the longitudinal MTP may be explained by assuming that inter-level scatterings by phonons play a dominant role above 4.2 K, and that both the diamagnetism based on surface current and the energy dependence of the electron relaxation time give rise to the pronounced oscillations of the longitudinal MTP below 4.2 K.

References

[1] Edelman V S 1976 Adv. Phys. 25 555
[2] Fu L and Kane C L 2007 Phys. Rev. B 76 045302
[3] Satoh N 2003 Physica B 336 290
[4] Satoh N, Kitamura Y and Takenaka H 2007 Physica B 391 244
[5] Satoh N, Takenaka H 2008 Physica B 403 3705
[6] Satoh N 2009 J. Phys.: Conf. Ser. 150 022072
[7] Mangez J H, Issi J P and Heremans J 1976 Phys. Rev. B 14 4381
[8] Smith G E, Baraff G A and Rowell J M 1964 Phys. Rev. 135 A1118
[9] Tanuma S and Inada R 1975 J. Phys.: Cond. Mat. 19 95
[10] Young R C 1973 J. Phys. F: Met. Phys. 3 721
[11] Fletcher R 1981 J. Phys. F: Met. Phys. 11 1093
[12] Pantsulaya A V and Varlamov A A 1989 Phys. Lett. A 136 317
[13] Dinger R J and Lawson A W 1973 Phys. Rev. B 7 5215
[14] Hofmann Ph 2006 Prog. Sur. Sci. 81 191
[15] Brown R N, Hartman R and Koenig S H 1968 Phys. Rev. 172 598
[16] Roth L M and Argyres P N 1966 Semiconductors and Semimetals vol 1, ed R K Willardson and A C Beer (San Diego: Academic Press) chapter 6
[17] Obraztsov Yu N 1965 Sovi. Phys. Solid State 7 455
[18] Ashcroft N W and Mermin N D 1976 Solid State Physics (Philadelphia: Saunders College) Chap 13 p 258

Acknowledgements

The author would like to thank Professor Y Ishizawa for valuable discussions and encouragement.