Influence of surface recombination on the longitudinal Shubnikov-de Haas oscillation amplitude

N Satoh
Department of Electronics and Computer Science, Iwaki Meisei University, 5-5-1 Iino chuoudai, Iwaki, Fukushima 970-8551, Japan
E-mail: nsatoh@iwakimu.ac.jp

Abstract. Longitudinal magnetoresistance has been measured for various sizes of pure bismuth single crystals in magnetic fields up to 5 T at a temperature of 4.2K in order to investigate the sample size dependence of the longitudinal Shubnikov-de Haas (SdH) oscillation. The samples used were sticks with regular square cross section elongate along the bisectrix axis of bismuth. Dimensions of the samples were in the range of 0.80 – 3.8 mm thick and about 20 mm long. When the sample thickness $d \leq 2.5$ mm, the amplitude of the longitudinal SdH oscillation reduces rapidly, while $d \geq 2.5$ mm it tends to saturate. This can be qualitatively explained by assuming that the gradient of carrier concentrations, even in the case of longitudinal magnetoresistance, is established near at surface by a balance of the Lorentz force, the counter diffusion force and the generation and recombination of carriers. The surface recombination time obtained from our data was found to be $2.6 \times 10^{-7}$ sec. The fact that the non-oscillatory part of magnetoresistance became gradually less in magnitude with decreasing in sample size was also found.

1. Introduction
It is well known that when measurements of resistivity are made on thin plate or on wires of small diameter, there is almost always extra scattering of carriers by the surface, especially if the mean free path in the bulk material is comparable to the smallest dimension of the sample [1]. In the presence of a magnetic field, generally, the Lorentz force of the applied magnetic field tends to bring about a “pileup” of charge carriers at some surface, and hence the gradient of carrier concentrations is set up near the surface, the amount of which depends strongly on the surface recombination velocity of carriers. Such diffusion size effect was observed in the transverse magnetoresistance and Hall coefficient of semimetal [2]. Tanuma and Ishizawa [3] observed that the amplitude of transverse Shubnikov-de Haas (SdH) oscillation for bismuth single crystals became gradually smaller in magnitude with decreasing sample thickness. They estimated the recombination time of electrons and holes to be $1.5 \times 10^{-8}$ sec from their data of the oscillation amplitude. We also obtained the recombination time for antimony to be $2.5 \times 10^{-10}$ sec from its transverse SdH oscillation amplitude [4]. While, to the best of our knowledge, no investigations have been made concerning the size effect in the case of longitudinal SdH oscillation in semimetals or semiconductors. Therefore, it should be interesting to investigate how the longitudinal SdH oscillation patterns changes for bismuth single crystals when the sample size is reduced.
2. Experimental

Brief description of the sample preparation and our experimental configuration are as follows. A piece of commercially available pure bismuth ingot (6N grade) was purified further and crystallized by zone-melting method. We prepared finally the various sizes of bismuth single-crystal rods with regular square cross section elongate along the \( y \) (bisectrix) direction. Each sample size was in the range of 0.80–3.8 mm thick and about 20 mm long. The surface treatment of all samples was done in exactly the same manner, that is, every sample was etched for one minute in a mixture of nitric and acetic acids (1 : 2) at a temperature of 10\(^\circ\)C and was annealed isothermally at 245\(^\circ\)C for a week to remove strain. The configuration of measurements was such that both the direction of electric and magnetic fields were parallel to the bisectrix axis, and potential probes were soldered to the arris of the rectangular parallelepiped. Hereafter, we denote this longitudinal magnetoresistivity component as \( \rho_{yy}(y) \). To achieve the longitudinal condition, we adjusted the sample mount to minimize \( \rho_{yy}(y) \) by in-situ measurement at a magnetic field of 0.15 T in which the SdH oscillations did not yet appear. The samples were directly immersed in liquid helium.

3. Results and discussion

Figure 1 shows our experimental results of the longitudinal magnetoresistivity component \( \rho_{yy}(y) \) for the various thicknesses of pure bismuth single crystals as a function of inverse magnetic field \( B^{-1} \). Quantum number of the peaks indicated by arrows at a magnetic field of 1.25 T in the figure is due to electron Landau levels \( e(0+, 1-) \) [5]. First, as shown in Fig. 1, the amplitude of the SdH oscillations becomes smaller in magnitude with decreasing sample sizes. Second, the decrease in the non-oscillatory part of \( \rho_{yy}(y) \) due to sample size reduction is also found. To visualize the size dependence of oscillation amplitude more clearly, we show the oscillation amplitude \( \rho_{osc} \) as a function of sample thickness \( d \) in Fig. 2, for the peaks indicated by arrows at magnetic fields of 0.82 and 1.25 T, respectively. Inset of Fig. 2 illustrates the definition of the \( \rho_{osc} \). As shown in Fig. 2, when \( d \leq 2.5 \) mm the amplitude reduces rapidly; however, when \( d \geq 2.5 \) mm, the amplitude has a tendency to saturate with increasing sample thickness.

![Figure 1](image1.png)  
**Figure 1.** \( \rho_{yy}(y) \) vs \( B^{-1} \). Inset denotes sample thickness. The values of magnetic field for the peaks indicated by arrows are 1.25 T and 0.82 T, respectively.

![Figure 2](image2.png)  
**Figure 2.** The amplitude \( \rho_{osc} \) at fields of 1.25 and 0.82 T as a function of sample thickness \( d \). Inset depicts the definition of \( \rho_{osc} \). Solid line is given by Eq. (1).
In the present paper, we are confine ourselves to this size dependence of the oscillation amplitude. From our measured data at a magnetic field of 1.25 T in Fig. 2, when $d \leq 2.5$ mm holds, the solid line obtained from a least square fitting of data can be written as follows:

$$\ln A = \ln(7.0 \times 10^{-9}) + 0.95d,$$

(1)

where $d$ is measured in millimeter. Suppose, both the electric and magnetic fields are parallel to the $y$-direction of a sample, the classical path of carriers is helices towards the $y$-direction. At the same time, transverse carrier flow appears along the $x$ (binary) axis direction, and hence the density gradient of carriers sets up near at surface of the sample. The magnitude of this carrier flow is determined by a balance of the Lorentz force, the counter diffusion force and the generation and recombination of electrons and holes in and on surface at the both ends of $x$-direction.

Now, according to Tanuma and Ishizawa [3], the quantity $q$ can be defined as follows:

$$q = [n(x) - n_0]/n_0 = [p(x) - p_0]/p_0,$$

(2)

where $n_0$ and $p_0 (=n_0)$ are the electron and hole number densities in thermal equilibrium, respectively, and $n(x) = \bar{n}(x)$ is the actual carrier density that deviates from $n_0 (= p_0)$ along the $x$-axis. In a spherical Fermi surface, for simplicity, Fermi energy holds, the solid line obtained from a least square fitting of data can be written as follows:

$$| \epsilon_F(x) - \epsilon_F | = (2/3)\epsilon_F q = k_B T^*.$$

(3)

$T^*$ is regarded as a kind of “Dingle Temperature”. Based on the theory of Roth and Argyres [6], and taken $T^*$ into account, the amplitude of SdH oscillation $A$ is expressed as $A \propto \exp[-2\pi^2k_B(T^* + T)/\hbar\omega_c]$, where $T$ denotes a real temperature of the sample, and $\omega_c$ the cyclotron frequency along the bisectrix axis. Using Eq. (3), we can rewrite the oscillation amplitude $A$ as:

$$\ln A = R - (4\pi^2\epsilon_F/3\hbar\omega_c)q,$$

(4)

where $R$ is the constant including $T$. As reported by Tanuma and Ishizawa [3], when the surface recombination is small, $q$ can be roughly replaced by

$$q = 2[1 - (Y/L^2)d],$$

(5)

where

$$Y = \frac{2(\epsilon_F^e + \epsilon_F^h)}{3e(\mu_e - \mu_h)BE}, \quad L = \left(\frac{\mu_eD_h + \mu_hD_e}{\mu_e + \mu_h}\tau_{eh}\right)^{1/2}.$$  

(6)

In the above equations, the electric field $E$ is the electric field, $B$ the magnetic field, $\mu_e$ and $\mu_h$ are the electron and hole mobilities, $D_e(=2\mu_ek_B\epsilon_F^e/3e)$ and $D_h(=2\mu_hk_B\epsilon_F^h/3e)$ the diffusion constants, $\epsilon_F^e$ and $\epsilon_F^h$ the Fermi energies for electron and hole, respectively. Also, the symbol $\tau_{eh}$ expresses the recombination time of electrons and holes, and $L$ means diffusion length. Then, the following relation can be obtained by substituting Eqs. (5) into Eq. (4), and by comparing Eq. (4) with Eq. (1) as:

$$8\pi^2\epsilon_F Y/3\hbar\omega_cL^2 = 0.95.$$

(7)

Substituting the values of $Y$ and $L$ into Eq. (7) and putting $\epsilon_F = \epsilon_F^e$, we estimated the recombination time $\tau_{eh}$ to be $2.6 \times 10^{-7}$ sec from Eq. (7). In evaluating $\tau_{eh}$, we employed the
following values for the $e_F^2$, $h_F^2$, $\mu_e$, $\mu_h$, the cyclotron mass of electron along bisectrix axis $m^*$ and the fields $B, E$: $e_F^2 = 27.6$ meV, $h_F^2 = 10.9$ meV [5, 7], $\mu_e = 5.7 \times 10^7$ cm$^2$/Vsec, $\mu_h = 2.2 \times 10^7$ cm$^2$/Vsec [8], $m^* = 0.0083 m_0$ [9], $B = 1.25$ T and $E = 2.5$ mV/cm for all samples. The resulting value of $\tau_{eh} = 2.6 \times 10^{-7}$ sec is one order of larger than that of Tanuma and Ishizawa [3]. This suggests the fact that a low rate of surface recombination and long diffusion length are realized in the case of longitudinal magnetoresistance. As a matter of fact, the diffusion length $L$ obtained from Eq. (6) is found to be 3.2 mm long. This value of 3.2 mm is nearly equal to the sample thickness $d$ at which the magnitude of amplitude $\rho_{osc}$ begins to saturate as shown in Fig. 2. In conclusion, above value of $\tau_{eh}$ estimated is one order of longer than that of one obtained from the transverse SdH oscillations [3]. This implies that specular reflection is predominant at bismuth surface, especially in the case of longitudinal magnetoresistance, and thereby a large gradient of carrier concentrations is established in the sample. We conclude that a long surface recombination time causes the decrease in the longitudinal SdH oscillation amplitude when the sample size is reduced.

Acknowledgments
The author would like to thank Professor Dr. Y. Ishizawa for discussion and encouragement.

4. References
[1] Friedman A N and Koenig S H 1960 IBM Journal Research Develop 4 158-162
[2] Hattori T 1967 J. Phys. Soc. Japan 23 19-34
[3] Tanuma S and Ishizawa Y 1967 J. low Temp. Phys. 3 341-45
[4] Satoh N 1999 Jpn. J. Appl. Phys. 38 2033-38
[5] Smith G E, Baraff G A and Rowell J M 1964 Phys. Rev. 135 A1118-24
[6] 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 pp 159-202
[7] Golin S 1968 Phys. Rev. 166 643
[8] Hartman R 1969 Phys. Rev. 181 1070-86
[9] Dinger R J and Lawson A W 1973 Phys. Rev. B 7 5215-27