

# NATIONAL INSTITUTE FOR FUSION SCIENCE

# Quantum Nernst Effect in a Bismuth Single Crystal

M. Matsuo, A. Endo, N. Hatano, H. Nakamura, R. Shirasaki, K. Sugihara

(Received - June 24, 2009)

NIFS-954

July 09, 2009

RESEARCH REPORT NIFS Series

1.5. C.S.S.

TORI, JAPAN

This report was prepared as a preprint of work performed as a collaboration research of the National Institute for Fusion Science (NIFS) of Japan. The views presented here are solely those of the authors. This document is intended for information only and may be published in a journal after some rearrangement of its contents in the future.

Inquiries about copyright should be addressed to the Research Information Office, National Institute for Fusion Science, Oroshi-cho, Toki-shi, Gifu-ken 509-5292 Japan.

E-mail: bunken@nifs.ac.jp

### <Notice about photocopying>

In order to photocopy any work from this publication, you or your organization must obtain permission from the following organization which has been delegated for copyright for clearance by the copyright owner of this publication.

Except in the USA

Japan Academic Association for Copyright Clearance (JAACC) 6-41 Akasaka 9-chome, Minato-ku, Tokyo 107-0052 Japan Phone: 81-3-3475-5618 FAX: 81-3-3475-5619 E-mail: jaacc@mtd.biglobe.ne.jp

# In the USA

Copyright Clearance Center, Inc. 222 Rosewood Drive, Danvers, MA 01923 USA Phone: 1-978-750-8400 FAX: 1-978-646-8600

## Quantum Nernst Effect in a Bismuth Single Crystal

M. Matsuo<sup>1,3\*</sup>, A. Endo<sup>2</sup>, N. Hatano<sup>3</sup>, H. Nakamura<sup>4</sup>, R. Shirasaki<sup>5</sup>, K. Sugihara<sup>6</sup>

<sup>1</sup>Graduate School of Integrated Sciences, Ochanomizu University, Otsuka, Bunkyo, Tokyo 112-8610, Japan

<sup>2</sup>Institute for Solid State Physics, University of Tokyo, Kashiwanoha, Kashiwa, Chiba 277-8581, Japan

<sup>3</sup>Institute of Industrial Science, University of Tokyo, Komaba, Meguro, Tokyo 153-8505, Japan

<sup>4</sup>Department of Simulation Science, National Institute for Fusion Science, Oroshi-cho, Toki, Gifu 509-5292, Japan
 <sup>5</sup>Department of Physics, Yokohama National University, 79-5 Tokiwadai, Yokohama 240-8501, Japan

<sup>6</sup>1-40-6-506 Shibayama, Funabashi, Chiba 274-0816, Japan

(Dated: June 24, 2009)

We calculate the phonon-drag contribution to the transverse (Nernst) thermoelectric power  $S_{yx}$  in a bismuth single crystal subjected to a quantizing magnetic field. The calculated heights of the Nernst peaks originating from the hole Landau levels and their temperature dependence reproduce the right order of magnitude for those of the pronounced magneto-oscillations recently reported by Behnia *et al* [Phys. Rev. Lett. **98**, 166602 (2007)]. A striking experimental finding that  $S_{yx}$  is much larger than the longitudinal (Seebeck) thermoelectric power  $S_{xx}$  can be naturally explained as the effect of the phonon drag, combined with the well-known relation between the longitudinal and the Hall resistivity  $\rho_{xx} \gg |\rho_{yx}|$  in a semi-metal bismuth. The calculation that includes the contribution of both holes and electrons suggests that some of the hitherto unexplained minor peaks located roughly at the fractional filling of the hole Landau levels are attributable to the electron Landau levels.

Keywords: Bismuth; quantum Hall effect; quantum Nernst effect; phonon-drag effect.

#### I. INTRODUCTION

A semi-metal bismuth has been attracting longstanding interest in the solid-state physics owing to its fascinating properties. The extraordinarily low carrier densities ( $\sim 10^{-5}$  per atom) and small effective masses ( $\sim 10^{-2}m_0$  with  $m_0$  the free electron mass) combined with the availability of highquality single crystals with highly mobile carriers render it an archetypal material for investigating the phenomena originating from the Landau quantization. In fact, a plethora of magneto-oscillation phenomena, including the de Hass-van Alphen and the Shubnikov-de Hass effects, were first discovered in bismuth,<sup>1</sup> illustrating distinguished roles played by the material in the history of the solid-state physics. Bismuth remains to be a subject of intensive ongoing studies spurred by its intriguing properties such as multi-valley degeneracy of Dirac-like electrons,<sup>2</sup> enhanced spin-orbit interaction on the surface,<sup>3</sup> strong diamagnetism advantageous for the potential observation of the quantum spin-Hall effect.<sup>4,5</sup>

The target of the present paper is the thermoelectric response of bismuth in a quantizing magnetic field. In a magnetic field B applied perpendicular to the temperature gradient  $\nabla T$ , the thermopower tensor contains not only the longitudinal (Seebeck effect,  $S_{xx}$ ) but also the transverse (Nernst effect,  $S_{yx}$ ) components, where we set the direction of  $\nabla T$ and B as the x and z directions, respectively. It is worth mentioning that the Nernst effect was also originally discovered in bismuth.<sup>6</sup> Magneto-oscillations of  $S_{xx}$  and  $S_{yx}$  due to the Landau quantization have been extensively studied in two-dimensional electron gases (2DEGs).<sup>7</sup> The effect of the Landau quantization is expected to be less easily observed in three-dimensional (3D) materials. Nevertheless, the initial ob-

servation of the magneto-oscillation in the thermoelectric coefficients of bismuth dates back to several decades ago.<sup>8-10</sup> The thermopower of bismuth has attracted renewed interest since the publication of recent experimental works by Behnia et al. They extended the measurement to lower temperatures  $(\sim 0.3 \text{ K})$  and higher magnetic fields  $(\sim 30 \text{ T})$  and reported prominent magneto-oscillations that rather appear as a series of discrete peaks <sup>11</sup> and further, small features in the ultraquantum limit that possibly signals the fractional quantization in three dimensions.<sup>12</sup> The oscillations in the thermopower were much more pronounced than the oscillations in the resistivity (the Shubnikov-de Haas oscillations). Interestingly, the Nernst signal  $S_{yx}$  was found to be much larger than the Seebeck signal  $S_{xx}$  in bismuth, in marked contrast to the case in 2DEGs, where generally  $S_{xx} > |S_{yx}|$ . Moreover, the lineshape of  $S_{yx}$  in bismuth was quite unlike that in 2DEGs: the former takes a peak when the chemical potential crosses a Landau level (as is the case in  $S_{xx}$  for 2DEGs), while in 2DEGs,  $S_{yx}$  changes sign.<sup>7</sup> The amplitudes of the peaks were large (~mV/K), and the peak heights rapidly increased with temperature. These findings, as well as the origin of small peaks located between the main peaks attributable to the Landau levels of the holes, remain unexplained. In an initial attempt toward the understanding, the present authors extended to 3D the theory for 2DEGs by Nakamura et al.13 that invokes the edge-current picture.<sup>14</sup> Although the calculation qualitatively reproduced the main peaks of the experimental traces, the amplitudes were found to be orders of magnitude smaller (~ 10  $\mu$ V/K). Furthermore, the theory failed to reproduce the strong temperature dependence. Note that the thermopower originating from the edge current corresponds to the contribution of the carrier diffusion in the clean limit in a quantizing magnetic field.<sup>13,15,16</sup> Inclusion of disorders was shown to further reduce the magnitude.<sup>15,17</sup> Therefore, the experimentally observed large-amplitude oscillation is not attributable to the diffusion contribution.

<sup>\*</sup>matsuo@iis.u-tokyo.ac.jp

In the present paper, we show that the large amplitude, the temperature dependence, and the dominance of  $S_{yx}$  over  $S_{xx}$  can be consistently explained as the effect of the phonon drag in the system containing both holes and electrons as carriers. Note that the phonon-drag contribution is known to play a dominant role also in 2DEGs.<sup>7</sup> Preliminary results of the phonon-drag contribution that consider only holes as carriers were already presented in Ref. 14. Here we describe more refined calculation that takes account of contributions of electrons, the charge neutral condition, and the Zeeman splitting neglected in Ref. 14. The calculation suggests that the minor peaks that appear at locations where fractional numbers of the hole Landau levels are filled actually originate from electron Landau levels.

#### II. PHONON-DRAG CONTRIBUTION TO TRANSVERSE THERMOPOWER

The Hamiltonian of the system with a magnetic field B and a small electric field  $F_y$  applied in the z (trigonal axis of bismuth) and y direction, repectively, is given by

$$\mathcal{H}_{a} = \frac{1}{2} (\boldsymbol{p} - e_{a} \boldsymbol{A}) \mathbb{M}_{a}^{-1} (\boldsymbol{p} - e_{a} \boldsymbol{A}) + \sigma g_{a} \mu_{B} B - e_{a} F_{y} y, \quad (1)$$

where  $\mathbf{A} = (-By, 0, 0)$  denotes the vector potential and  $\sigma = \pm 1/2$  the spin. The suffix a is used throughout the paper to indicate the quantity either of a hole (a = h) or of an electron (a = e), with  $e_h = e$  and  $e_e = -e$  (e > 0). The effective mass tensors for holes and electrons are

$$\mathbb{M}_{\rm h} = \begin{pmatrix} m_{\rm hx} & 0 & 0\\ 0 & m_{\rm hx} & 0\\ 0 & 0 & m_{\rm hz} \end{pmatrix}$$
(2)

and

$$\mathbb{M}_{e} = \begin{pmatrix} m_{ex} & 0 & m_{exz} \\ 0 & m_{ey} & 0 \\ m_{exz} & 0 & m_{ez} \end{pmatrix},$$
(3)

respectively.<sup>18</sup> The values of the components are listed in Table I. The eigenenergy of the Hamiltonian (1) in first order of  $F_y$  reads

$$E_a(n,k_z,\sigma) = \hbar\omega_a \left(n + \frac{1}{2}\right) + \frac{\hbar^2 k_z^2}{2m_{az}} + \sigma g_a \mu_B B - e_a F_y Y_{0a}$$
<sup>(4)</sup>

with the cyclotron frequency  $\omega_a \equiv eB/m_a$ , where the cyclotron mass  $m_a$  is given by  $m_{\rm h} = m_{\rm hx}$  and  $m_{\rm e} = \sqrt{\det \mathbb{M}_{\rm e}/m_{\rm ez}}$ .<sup>19,20</sup> The corresponding eigenfunction is  $\psi_a(y - Y_{0a}; n, k_x, k_z) = \phi_a(y - Y_{0a}; n) \exp[i(k_x x + k_z z)]$ , where  $Y_{0\rm h} = \hbar k_x/(eB)$ ,  $Y_{0\rm e} = -\hbar (k_x - k_z m_{\rm exz}/m_{\rm ez})/(eB)$ , and

$$\phi_a(y;n) \equiv \left(2^n n! \sqrt{\pi} l_a\right)^{-1/2} e^{-y^2/(2l_a^2)} H_n(y/l_a), \quad (5)$$

with the magnetic length  $l_a = \sqrt{\hbar/(m_{ay}\omega_a)}$  represented as  $l_{\rm h} = \sqrt{\hbar/(eB)}$  and  $l_{\rm e} = \sqrt{m_{\rm e}/m_{\rm ey}}\sqrt{\hbar/(eB)}$ .

We now describe our calculation of the phonon-drag effect. The phonon-drag thermopower in a magnetic field was studied for bismuth by Sugihara <sup>21,22</sup> and for a GaAs/AlGaAs 2DEG by Kubakaddi et al.23 We here closely follow Sugihara's calculation. The difference from his calculation is that we treat the Fermi and Bose distributions exactly and evaluate the magnetic-field dependence numerically. For the calculation of the thermopower, there are two equivalent approaches. In the Q approach, we calculate the electric current under a temperature gradient, while in the  $\Pi$  approach, we calculate the heat current under an electric field. The two approaches are related through the Kelvin-Onsager relation <sup>24</sup>;  $S_{yx}(B) = \prod_{xy}(-B)/T$ , where  $\prod_{xy}$  is the Peltier coefficient. Here we follow the  $\Pi$  approach. Carriers accelerated by the electric field  $F_y$  "drag" phonons because of carrier-phonon interaction and thus generate the heat current of phonons. The heat currents of holes and electrons are negligibly smaller than that of phonons. Then the Peltier coefficient is given by  $\Pi_{xy} = Q_x \rho_{xx} / F_y$ , where  $Q_x$  denotes the heat current of phonons in the x direction and we used the relation  $\rho_{xx} \gg |\rho_{yx}|$  characteristic of the systems that contain both holes and electrons as carriers, where  $\rho_{xx}$  and  $\rho_{yx}$  denote the longitudinal and the Hall resistivities, respectively.

At low temperatures we may neglect all lattice excitations except acoustic phonons with the energy  $\hbar\omega_q$  and the wave vector q, which are generated through deformation coupling. The heat current of phonons in the x direction is then given by

$$Q_x = \int \frac{d\boldsymbol{q}}{(2\pi)^3} \hbar \omega_q v_s \frac{q_x}{q} g(\boldsymbol{q}), \tag{6}$$

where  $\omega_q = v_s q$ ,  $v_s$  is the group velocity of the phonons and  $g(q) = N_q - N_q^{(0)}$  represents the displacement of the phonon distribution  $N_q$  from its equilibrium Bose distribution  $N_q^{(0)}$ . In order to estimate the displacement, we use the Boltzmann equation in the steady state;

$$\left(\frac{\partial N\boldsymbol{q}}{\partial t}\right)_{\text{carrier}} + \left(\frac{\partial N\boldsymbol{q}}{\partial t}\right)_{\text{relaxation}} = 0. \tag{7}$$

Hole	Electron
$m_{\rm hx} = 0.06289$	$m_{\rm ex} = 0.26$
	$m_{\rm ey} = 0.00113$
$m_{\rm hz} = 0.6667$	$m_{\rm ez} = 0.00443$
—	$m_{\mathrm exz} = -0.0195$
$2.16 \hbar \omega_{\rm h} {}^{26}$	$0.5849  \hbar \omega_{\mathrm{e}}^{-19}$
$D_{\rm h} = 1.2~{\rm eV}$	$D_{\rm e} = 2.2 \; {\rm eV}$
Band gap at L point $E_g = 15.3$ meV <sup>25</sup>	
Band overlap $\mu_0 = 38.5$ meV <sup>25</sup>	
Group velocity of phonons $v_s = 2 \times 10^3$ m/s $^{27}$	
Density $\rho = 9.75 \times 10^3  \mathrm{kg/m^3}$	
Size of the sample $W = 2.2$ mm, $L = 4.0$ mm $^{11}$	
	$\frac{m_{hx} = 0.06289}{m_{hz} = 0.6667}$ $\frac{m_{hz} = 0.6667}{D_{h} = 1.2 \text{ eV}}$ $\frac{p_{hz} = 15.3}{p_{hz} = 15.3}$ $\ln \mu_{0} = 38.5 \text{ me}$ $p_{hz} = 0.6667$ $\frac{p_{hz} = 0.6667}{p_{hz} = 15.3}$

TABLE I: Parameter values used in our calculation, taken from Refs. 25–27. We calculated the Zeeman energy of electrons by Smith's method <sup>19</sup> using the effective masses in Ref. 25.

The first term of the left-hand side represents the change of the phonon distribution due to interaction with carriers and the second term represents that due to other interactions such as boundary scattering, phonon-phonon interaction and impurity scattering. These two terms are balanced in the steady state.

We estimate the quantity  $(\partial N q / \partial t)_{\text{carrier}}$  in the Born approximation as

$$\left(\frac{\partial N\boldsymbol{q}}{\partial t}\right)_{\text{carrier}} = \sum_{\alpha,\alpha'} \left[ W^{(\text{em})}(\alpha',\alpha) f_{\alpha'}(1-f_{\alpha}) - W^{(\text{ab})}(\alpha,\alpha') f_{\alpha}(1-f_{\alpha'}) \right], (8)$$

where  $f_{\alpha} = f(E(\alpha))$  is the Fermi distribution of carriers in a state  $\alpha$ . Each  $\alpha$  represents the set of three quantum numbers  $(n, k_x, k_z)$ , and  $W^{(\text{em})}(\alpha', \alpha)$  and  $W^{(\text{ab})}(\alpha, \alpha')$  are the transition probabilities from a state  $\alpha$  to a state  $\alpha'$  by emitting or absorbing a phonon, respectively, given by Fermi's golden rule,

$$\frac{W^{(\text{em})} = N\boldsymbol{q} + 1}{W^{(\text{ab})} = N\boldsymbol{q}} \left\{ \times \frac{2\pi |V_q|^2}{\hbar} \left| \langle \psi_{\alpha'} | e^{\pm i \mathbf{q} \cdot \mathbf{r}} | \psi_{\alpha} \rangle \right|^2 \times \delta \left( E_a(\alpha') - E_a(\alpha) - \hbar \omega_q \right) \quad (9)$$

with  $|V_q|^2 = D_a^2 \hbar q / (2\rho V v_s)$ , where  $\rho, V$ , and  $D_a$  are the bismuth density, the sample volume, and the deformation potential of carriers, respectively. Expanding Eq. (8) in  $O(F_y)$ we have the first term in Eq. (7). In the second term, we use the relaxation-time approximation;  $(\partial N_q / \partial t)_{\text{relaxation}} = -g(q)/\tau_r(q)$ . The carrier-phonon interaction changes the phonon distribution, but other interactions make the nonequilibrium distribution relax back to the equilibrium one in time  $\tau_r$ . Solving Eq. (7) with respect to g(q), we obtain

$$\left(\frac{\partial N\boldsymbol{q}}{\partial t}\right)_{\text{carrier}} = -\frac{1}{\tau_{\text{c}}(\boldsymbol{q})} \left(\frac{\hbar F_{y} \tilde{q}_{a}}{k_{B} T B} N_{q}^{(0)} (N_{q}^{(0)} + 1) + g(\boldsymbol{q})\right), (10)$$

$$g(\boldsymbol{q}) = -\frac{\hbar F_y \tilde{q}_a}{k_B T B} N_q^{(0)} (N_q^{(0)} + 1) \frac{\tau_{\text{tot}}(\boldsymbol{q})}{\tau_{\text{c}}(\boldsymbol{q})}, \qquad (11)$$

where

$$\frac{1}{\tau_{\rm c}(\boldsymbol{q})} \equiv \frac{2\pi}{\hbar} |V_q|^2 \sum_{\alpha,\alpha'} |\langle \psi_a(\alpha')|e^{-i\mathbf{q}\cdot\mathbf{r}}|\psi_a(\alpha)\rangle|^2 \\ \times \frac{f_\alpha(1-f_{\alpha'})}{N_q^{(0)}+1} \delta(E_a(\alpha')-E_a(\alpha)-\hbar\omega_q), \quad (12)$$

 $\tau_{\rm tot}(\boldsymbol{q})^{-1} = \tau_r(\boldsymbol{q})^{-1} + \tau_c(\boldsymbol{q})^{-1}$ , and  $\tilde{q}_{\rm h} = q_x$ ,  $\tilde{q}_{\rm e} = q_x - q_z m_{exz}/m_{ez}$ . At low temperatures, the phonons in a bismuth single crystal are known to be ballistic and the boundary scattering is dominant,<sup>10,21,28</sup> and therefore we set  $\tau_{\rm tot}(\boldsymbol{q})^{-1} \cong v_s/L$  where L is the length in the x direction.

By plugging Eq. (11) into Eq. (6), we have

$$\frac{Q_{xa}}{F_y} = -\frac{1}{(2\pi)^4} \frac{e\hbar L D_a^2}{2k_B T \rho} \sum_{n,n'} \int d\mathbf{q} \, q_x \tilde{q}_a \, q \, N_q^{(0)} I_{an,n'}(\mathbf{q}) \\
\times \int dk_z f_\alpha (1 - f_{\alpha'}) \delta \left( E_a(\alpha') - E_a(\alpha) - \hbar \omega_q \right), (13)$$

where

$$I_{an,n'}(\boldsymbol{q}) = \left| \int_{-\infty}^{\infty} \phi_a(y - \hbar \tilde{q}_a/(e_a B); n') e^{-iq_y y} \phi_a(y; n) \mathrm{d}y \right|^2 (14)$$

We thus arrive at  $S_{yx} = -Q_x \rho_{xx}/(F_y T)$  by adding Eq. (13) up over the spin degree of freedom and also over electrons and holes. The integration with respect to  $k_z$  can be done analytically, and we obtain

$$S_{yx} = -\frac{1}{(2\pi)^4} \frac{e\rho_{xx}L}{2k_B T^2 \rho \hbar} \sum_{a=h,e} D_a^2 m_{az}^* \sum_{\sigma} \sum_{n,n'} \\ \times \int d\mathbf{q} \, \frac{q_x \tilde{q}_a \, q}{q_z} N_q^{(0)} I_{an,n'}(\mathbf{q}) \\ \times f(E_a(n,k_{z0a},\sigma)) [1 - f(E_a(n',k_{z0a}+q_z,\sigma))]$$
(15)

with

$$k_{z0a} = \frac{m_{az}}{\hbar q_z} [\omega_a (n - n') + \omega_q] - \frac{1}{2} q_z.$$
(16)

Finally the integration with respect to q is performed numerically. For the values of  $\rho_{xx}$ , we made use of the experimental data (at 0.25 K) by Behnia.<sup>29</sup>

#### III. RESULTS OF CALCULATION AND COMPARISON WITH EXPERIMENT

We first consider only holes as carriers; holes produce a greater contribution than electrons because the effective mass in the direction of the magnetic field, hence the density-of-states peak at a Landau level, is larger for holes than for electrons. In Fig. 1 we compare the theoretical and experimental results at T = 0.28 K. In the calculation we used the parameter values given in Table I and the constant chemical potential for holes  $\mu_{\rm h} = 11.4$  meV.<sup>30,31</sup> The calculated locations and heights of the peaks are in reasonable agreement with the experiment (except for the location of  $1\uparrow$ , whose agreement is improved by the use of *B*-dependent chemical potential, see below). The good agreement infers that the phonon-drag is the dominant mechanism for the observed Nernst effect. We stress that our theory here has *no adjustable fitting parameters*.

Next we further include the contribution of electrons. Instead of using a fixed value, we now use a *B*-dependent chemical potential satisfying the charge neutral condition; that is, we determine it such that the number of electrons and holes are equal. (Note that  $\mu_e + \mu_h = \mu_0$ , where  $\mu_e$  and  $\mu_h$  represent chemical potentials for electrons and holes, respectively, and



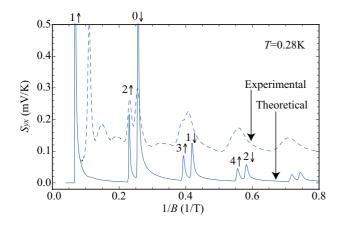


FIG. 1: (Color online). The transverse thermopower  $S_{yx}$  of holes at T=0.28 K against the inverse magnetic field 1/B. The solid lines are our theoretical results and the broken lines are the experimental results by Behnia *et al.*<sup>11</sup> The peaks are labeled by (n, spin).

 $\mu_0$  the band overlap.) We evaluated the chemical potentials by Smith *et al.*'s method.<sup>19</sup> Smith *et al.* used a model proposed by Lax *et al.*,<sup>20,32</sup> where the conduction band becomes non-parabolic under the influence of a filled band just below it. The energy of electron in the Lax model is represented by

$$E_{\rm e}\left(1+\frac{E_{\rm e}}{E_g}\right) = \hbar\omega_{\rm e}\left(n+\frac{1}{2}\right) + \frac{\hbar^2k_z^2}{2m_{\rm ez}} + \sigma g_{\rm e}\mu_B B, (17)$$

where  $E_g$  is the band gap between the conduction band and the filled band. According to the model, the chemical potential for electrons (holes) increases (decreases) as the magnetic field is increased. In the actual calculation of Eq. (15), we linearized the Lax model (17) around  $E_e = \mu_e$  for simplicity, noting that only the energy level in the immediate vicinity of the chemical potential is relevant at low temperatures.

The result is shown in Fig. 2. We again used the parameter values shown in Table I. The peaks originating from holes remain basically unchanged from Fig. 1, but we now have additional peaks resulting from electrons. Our result suggests the possibility that the minor peaks between the major ones observed in the experiment are due to the electron contribution, rather than to the fractional quantization as implied in Ref. 12. The peak labeled as  $e1\uparrow$  may correspond to the peak at 14 T shown in Fig. 1 of Ref. 12 (not shown in Fig. 2). The calculated values of  $S_{yx}$  at the peaks due to electrons, as well as those of holes at low magnetic-field regime, are substantially smaller than those of the experiment. However, the height of the peaks, if we disregard the smooth background that considerably differs between the calculation and the experiment, are in rough agreement. The origin of the smooth background observed in the experiment is not known at present but is presumably related to the presence of disorders completely neglected in our calculation. Further discussion on the role of the disorder will be given below. In the calculation of electron contribution, we considered one of the three equivalent electron pockets rotated by  $120^{\circ}$  to each other,<sup>1</sup> one with the long axis parallel to the heat current, and simply multiplied the re-

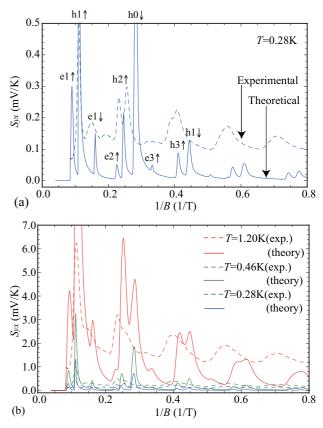


FIG. 2: (Color online). (a) The transverse thermopower  $S_{yx}$  of holes and electrons at T=0.28 K against the inverse magnetic field 1/B. (b) The data at T=0.28, 0.46 K, 1.20 K (from bottom to top). The solid lines are our theoretical results and the broken lines are the experimental results by Behnia *et al.*<sup>11</sup>. Note that the peak labeled  $e2\downarrow$  is overlapping with  $h0\downarrow$ . The peaks are labeled by (a, n, spin).

sult by three, neglecting the anisotropy. We estimate that the peak heights would become slightly smaller than those shown in Fig. 2 due to the anisotropy, although it is difficult to take full account of the anisotropy in the calculation. Compared with Fig. 1, the peak h1<sup>↑</sup> shifted to lower magnetic field side owing to decrease in  $\mu_h$  with increasing B, and coincide better with the experimental peak, while agreement of the positions of other major peaks slightly worsen. The slight inconsistency of the peak locations may be attributable to the minute discrepancy between values of the effective masses, the g-factor, and the band parameters in the literature and those of the sample used in the experiment. (Very recently, it has been pointed out that slight misalignment in the direction of the magnetic field from the trigonal axis can also cause small shift in the peak positions.<sup>33</sup>) In Fig. 2(b) it can be seen that the strong temperature dependence of the experimental peak heights is reproduced well in the calculation.

#### **IV. DISCUSSION**

We now comment on several characteristics of bismuth and/or the phonon-drag effect that are operative in yielding the sharp and large-amplitude oscillation of  $S_{yx}$ . (i) The small carrier density in bismuth is advantageous to the phonon-drag effect, since carriers with small Fermi momentum readily interact with phonons. (ii) The conservation of energy and momentum in the carrier-phonon interaction,  $\hbar^2 \left(k_z^{\prime 2} - k_z^2\right)/2m_{az} = \hbar v_s q$  and  $k_z^{\prime} = k_z + q_z$ , leads to  $k_z = O(q)$  [see Eq. (16)]. Here we consider only the intra-Landau-level scattering (n = n'); the inter-Landau-level scattering is practically prohibited in a quantizing magnetic field since  $\hbar\omega_a \gg \hbar\omega_q$ . Since only phonons having small q are available at low temperatures, only carriers with small  $k_z$  are involved in the phonon-drag events, resulting in sharp peaks where Landau levels cross the chemical potential [see Eqs. (4) and (15)]. (iii) The dominance of  $S_{yx}$  over  $S_{xx}$  is ascribable to the relation  $\rho_{xx} \gg |\rho_{yx}|$  in bismuth, which contains both holes and electrons as carriers. The longitudinal and transverse thermopowers  $S_{xx}$  and  $S_{yx}$  are given by  $S_{xx} = \rho_{xx}\epsilon_{xx} - \rho_{yx}\epsilon_{yx}$  and  $S_{yx} = \rho_{yx}\epsilon_{xx} + \rho_{xx}\epsilon_{yx}$ , respectively, where  $\epsilon$  is the thermoelectric tensor. For the phonondrag effect, it has been shown that  $|\epsilon_{yx}| \gg |\epsilon_{xx}| \sim 0,^{34}$  resulting in  $|S_{yx}| \gg |S_{xx}|$  for bismuth, or for ambipolar conductor in general (and  $|S_{yx}| \ll |S_{xx}|$  for 2DEGs or generally for systems with  $\rho_{xx} \ll |\rho_{yx}|$ ; roughly speaking  $S_{yx}$  in bismuth corresponds to  $S_{xx}$  in 2DEGs. The relation  $|\epsilon_{xx}| \sim 0$  also allows us to evaluate  $S_{xx} \simeq -\rho_{yx}\epsilon_{yx}$  simply by replacing  $\rho_{xx}$  in Eq. (15) with  $\rho_{yx}$ . Using the experimentally obtained  $\rho_{yx}$ <sup>29</sup> the calculation yields  $S_{xx}$  having the peaks at roughly the same positions as in  $S_{yx}$  but  $\sim 1/20$  in magnitude. The relation between  $S_{yx}$  and  $S_{xx}$  is in rough agreement with the experimental result shown in Fig. 1 of Ref. 11.

In our calculation, we neglected disorders in bismuth altogether. Although the effect of disorders is expected to be rather small in a high-quality bismuth single crystal, we believe that it constitutes the main source of the remnant discrepancy between the theoretical and the experimental traces. Inclusion of disorders introduces a width in the energy of Landau levels represented by the first term in Eq. (4). The delta function in Eq. (13) denoting the energy conservation is then replaced by a peak function having the width acquired by the Landau levels, thereby making the peaks in  $S_{yx}$  broader <sup>17,23</sup> with concomitant decrease in the peak heights. The narrower peak width in the theoretical curves that allows some of the peaks not well-resolved in the experiment to be resolved is thus attributable to the neglect of the disorders in our calculation. The width in the Landau levels will also affect the kinetics involved in the carrier-phonon interaction. In the energy and momentum conservation mentioned above, only the kinetic energy in the z direction,  $\hbar^2 k_z^2/2m_{az}$ , was allowed to vary, since the kinetic energy in the x-y plane was strictly fixed to the Landau levels. Introduction of the width into the Landau levels alters the situation; the phonons can now also impart their energy to the in-plane kinetic energy of the carriers without affecting  $\hbar^2 k_z^2/2m_{az}$ . The restriction on the extent of  $k_z$  mentioned above is thus removed, enabling the carrier-phonon scattering to take place regardless of the value of  $k_z$ . This may partly be responsible for the smooth background observed in the experiment.

#### V. CONCLUSIONS

We have calculated the transverse thermopower  $S_{yx}$  due to the phonon-drag effect, taking both holes and electrons into account as carriers. A series of large (~ mV/K) peaks originating from holes, with smaller peaks deriving from electrons in between, are obtained. The heights as well as the positions of the peaks are close to those recently observed experimentally by Behnia *et al*,<sup>11</sup> in stark contrast with the calculation based on the edge-current picture, corresponding to the diffusion contribution, in which the peak heights are orders of magnitude smaller. This strongly suggests that the phonondrag is the dominant mechanism in the experimentally observed prominent magneto-oscillations in the Nernst coefficient. Rather broad width of the peaks and the smooth background not reproduced in our calculation are attributable to the disorders neglected in our calculation.

#### Acknowledgments

We thank Y. Hasegawa for constructive comments, and K. Behnia for providing the experimental data. The work is supported partly by the Thermal & Electric Energy Technology Foundation, Foundation for Promotion of Material Science and Technology of Japan, and the Iketani Foundation as well as by NINS' Creating Innovative Research Fields Project (No. NIFS08KEIN0091) and Grants-in-Aid for Scientific Research (Nos. 17340115 and 20340101) from The Ministry of Education, Culture, Sports, Science and Technology (MEXT). The computation was partly done using the facilities of Supercomputer Center, Institute for Solid State Physics, University of Tokyo, Japan.

- <sup>1</sup> V. S. Edelman, Adv. In Phys. **25**, 555 (1976).
- <sup>2</sup> Lu Li, J. G. Checkelsky, Y. S. Hor, C. Uher, A. F. Hebard, R. J. Cava, and N. P. Ong, Science **321**, 547 (2008).
- <sup>3</sup> T. Hirahara, T. Nagao, I. Matsuda, G. Bihlmayer, E. V. Chulkov, Yu. M. Koroteev, P. M. Echenique, M. Saito, and S. Hasegawa, Phys. Rev. Lett. **97**, 146803 (2006).
- <sup>4</sup> L. Fu, C. L. Kane, and E. J. Mele, Phys. Rev. Lett. **98**, 106803 (2007).
- <sup>5</sup> S. Murakami, Phys. Rev. Lett. **97**, 236805 (2006).
- <sup>6</sup> A. V. Ettingshausen and W. Nernst, Annal. Phys. Chem. **265**, 343 (1886).
- <sup>7</sup> R. Fletcher, Semicond. Sci. Technol. **14**, R1 (1999).

- <sup>8</sup> M. C. Steele and J. Babiskin, Phys. Rev. **98**, 359 (1955).
- <sup>9</sup> J. H. Mangez, J. P. Issi, and J. Hermans, Phys. Rev. B 14, 4381 (1976).
- <sup>10</sup> B. Farag and S. Tanuma, Technical Report of ISSP Ser. B No. 18 (1976).
- <sup>11</sup> K. Behnia, M. A. Méasson, and Y. Kopelevich, Phys. Rev. Lett. 98, 166602 (2007).
- <sup>12</sup> K. Behnia, L. Balicas, and Y. Kopelevich, Science **317**, 1729 (2007).
- <sup>13</sup> H. Nakamura, N. Hatano and R. Shirasaki, Solid State Comm. 135, 510 (2005).
- <sup>14</sup> M. Matsuo, A. Endo, N. Hatano, H. Nakamura, R. Shirasaki, and K. Sugihara, *Proceedings of ISQM-TOKYO 08*, eds. S. Ishioka and K. Fujikawa (World Scientific, Singapore).
- <sup>15</sup> M. Jonson and S. M. Girvin, Phys. Rev. B **29**, 1939 (1984).
- <sup>16</sup> H. Oji, Phys. Rev. B **29**, 3148 (1984).
- <sup>17</sup> R. Shirasaki, H. Nakamura, and N. Hatano, e-J. Surf. Sci. Nanotech. 3, 518 (2005).
- <sup>18</sup> Here we interchanged the x and y axes from the standard notation for the convenience in the later calculation.
- <sup>19</sup> E. Smith, G. A. Baraff, and J. M. Rowell, Phys. Rev. **135**, A1118 (1964).
- <sup>20</sup> B. Lax, J. G. Mavroides, H. J. Zeiger, and R. J. Keyes, Phys. Rev. Lett. 5, 241 (1960).

- <sup>21</sup> K. Sugihara, J. Phys. Soc. Jpn. 27, 356 (1969).
- <sup>22</sup> K. Sugihara, J. Phys. Soc. Jpn. **27**, 362 (1969).
- <sup>23</sup> S. S. Kubakaddi, P. N. Butcher, and B. G. Mulimani, Phys. Rev. B 40, 1377 (1989).
- <sup>24</sup> C. Herring, Phys. Rev. **96**, 1163 (1954).
- <sup>25</sup> The Japan Institute of Metals (ed), *Handoutai to Hankinzoku (in Japanese)* (Agne Gijutsu Center, Tokyo, 1990).
- <sup>26</sup> S. G. Bompadre, C. Biagini, D. Maslov, and A.F. Hebard, Phys. Rev. B 64, 073103 (2001).
- <sup>27</sup> K. Walther, Phys. Rev. **174**, 782 (1968).
- <sup>28</sup> K. Behnia, M. A. Méasson, and Y. Kopelevich, Phys. Rev. Lett. 98, 076603 (2007).
- <sup>29</sup> K. Behnia, private communication.
- <sup>30</sup> H. Kawamura, *Kotai Purazuma (in Japanese)* (Asakura, Tokyo, 1972).
- <sup>31</sup> Slight variation between literatures is found on the value of  $\mu_{\rm h}$ . For example,  $\mu_{\rm h}$  (meV) = 38.5 – 27.6 = 10.9 is given in Ref. 19.
- <sup>32</sup> B. Lax, Rev. Mod. Phys. **30**, 122 (1958).
- <sup>33</sup> Y. V. Sharlai and G. P. Mikitik, Phys. Rev. B 79, 081102(R) (2009).
- <sup>34</sup> T. M. Fromhold, P. N. Butcher, G. Qin, B. G. Mulimani, J. P. Oxley, and B. L. Gallagher, Phys. Rev. B 48, 5326 (1993).