Out-of-Plane Resistance under Transverse Magnetic Field in Quasi-One-Dimensional Layered Metals

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(Received February 15, 2006; accepted March 24, 2006; published May 10, 2006)

We succeeded in deriving an analytical expression that yields the electrical conductivity for quasi-one-dimensional electron systems, when the current is in the least conductive direction and the magnetic field is perpendicular to it. This formula is valid for the magnetic field along an arbitrary direction in the conducting layers and the field strength from zero to higher fields, where \( \omega t \approx 1 \). We verified the applicability of the formula using the experimental results for an organic quasi-one-dimensional layered metal (TMTSF)\(_2\)ClO\(_4\). The formula was found to reproduce the experimental results fairly well.

KEYWORDS: layered material, quasi-one-dimensional metal, transverse magnetoresistance, out-of-plane resistance, (TMTSF)\(_2\)PF\(_6\), (TMTSF)\(_2\)ClO\(_4\)

DOI: 10.1143/JPSJ.75.053704

In the recent decades, conductors with a layered structure such as high \( T_c \) superconductors and organic conductors have provided a good basis for investigating low-dimensional electron systems. Many studies that clarified the effect of the dimensionality on the character of electrons were accumulated. Investigating the mechanism of transport phenomena has been one of the main issues in these investigations. It is also important because it contains information on the Fermi surface, which is inevitable in the discussion on the electronic property. Shubnikov–de Haas phenomena has been one of the main issues in these discussions on the electronic property. Shubnikov–de Haas oscillation\(^{1-4})\) are examples of this property. Recently, we showed that out-of-plane resistance in the transverse magnetic fields for quasi-two-dimensional electron systems with closed Fermi surfaces can be expressed by an analytical formula.\(^5)\) Since it is expressed analytically, it provides a method to obtain information on Fermi surfaces.

In this study, we extended the study of the magnetoresistance effect to quasi-one-dimensional electron systems such as (TMTSF)\(_2\)PF\(_6\) and (TMTSF)\(_2\)ClO\(_4\). In these materials, the electron energy spectrum is approximated by the following formula:

\[
\epsilon(k) = -2t_x \cos(ak_x) - 2t_y \cos(ak_y) - 2t_z \cos(ak_z) - \epsilon_F, \tag{1}
\]

where \( x, y, \) and \( z \) axes are perpendicular to each other. Here, \( a, b, \) and \( c \) are lattice constants along the \( x, y, \) and \( z \)-directions, respectively, \( t_x \) and \( t_y \) are the transfer energies along the \( x \)- and \( y \)-directions, respectively, and \( t_z \) is the transfer energy along the \( z \)-direction. \( \epsilon_F \) is the Fermi energy. They are assumed to satisfy the following relations: \( t_x \gg t_y \gg t_z \) and \(-2t_x + 2t_y + 2t_z < \epsilon_F < 0 \). It exhibits a pair of sheet-like Fermi surfaces parallel to the \( k_x \)- and \( k_y \)-directions (Fig. 1).

Osada et al.\(^6)\) discussed the angular-dependent magneto-resistance in (TMTSF)\(_2\)ClO\(_4\). Danner and Chaikin\(^7)\) also discussed the magneto-resistance for quasi-one-dimensional electron systems. Lebed and Bagmet\(^8)\) treated the "third magic angle effect"\(^9)\) theoretically and showed that the resistance in the magnetic field along this direction obeys the relation \( \rho \propto \sqrt{B} \). In most of these studies, discussions are limited to the topological aspect of the phenomena. However, quantitative discussions are required for fully understanding the magneto-resistance phenomenon. See Fig. 2 for example. In this figure, a strongly anisotropic magnetoresistance is observed for the field direction \( \theta = \pi/2 \), the resistance in the magnetic field of 7 T is approximately 60 times the zero field value. For \( \theta = 0 \), on the other hand, it is only 2.5 times the zero field value. Lee and Naughton\(^10)\) and Yoshino and Murata\(^11)\) independently discussed this strong anisotropy quantitatively by means of a numerical calculation. The numerical result reproduces the anisotropy in magnitude fairly well; however, it cannot explain the field or angular dependence of the magneto-resistance in detail. On the other hand, Lebed and Naughton\(^12)\) and, recently, Cooper and Yakovenko\(^13)\) derived an analytical formula for the anisotropic magnetoresistance. However, the formula is a complicated function expressed as the sum of infinite terms of Bessel function such that the approximate form of the formula is required to fit the data. In this study, we propose a new formula expressed by an elementary function, which allows the discussion of the out-of-plane resistance in a transverse magnetic field quantitatively. A semiclassical calculation based on the Boltzmann equation was performed, and we succeeded in deriving a "basic" formula for the first time in a quasi-one-dimensional system.

We now derive a formula for an out-of-plane resistance when quasi-one-dimensional layered metal is placed in a transverse magnetic field. We consider the electron system with the dispersion relation given by eq. (1). Within the linear approximation, it is expressed as

\[
\epsilon(k) = \pm \hbar v_F (k_x \mp k_y) - 2t_z \cos(ak_z) - 2t_{\perp} \cos(ak_z), \tag{2}
\]

Here, we introduce the Fermi wave number \( k_F \) defined as \(-2t_z \cos(ak_z) = \epsilon_F \) and the Fermi velocity as \( v_F = 2t_{\perp} \sin(ak_z)/\hbar \). This equation describes the fairly anisotropic planar metals with weak coupling along the out-of-plane direction; Fermi surfaces for which the weakly
modulated sheets open to the $B$-field. Two types of orbits are expected in our field geometry (see Fig. 1). One is the orbit open in the $k_z$-direction (open orbit). The electrons in these orbits undergo periodical motion across the first Brillouin zone. The other is the small closed orbit (SCO), appearing only when $\theta \sim 0$. In the calculation of the electron motion in the $k$-space, we neglect the $z$-component of the group velocity in eq. (4); therefore,

$$ \mathbf{v} = \left( v_F, \frac{2t_z b}{\hbar} \sin(bk_y), 0 \right) . $$

Electron motion is given by solving the semiclassical equation of motion:

$$ \mathbf{k} = -\frac{e}{\hbar} \mathbf{v} \times \mathbf{B} . $$

with $\mathbf{v}$ given by eq. (6). The result is $k_z(t) = k_z(0)$, $k_y(t) = k_y(0)$ and

$$ k_x(t) = k_x(0) - \frac{\omega_t \sin \theta - \eta \omega_t \cos \theta \sin(bk_y(0))}{c} , $$

where $\omega$ is the cyclotron frequency defined by

$$ \omega = \frac{e v_F B_c}{\hbar} , $$

and $\eta$ is the anisotropy parameter of the velocity between the $x$- and $y$-directions expressed as

$$ \eta = \frac{2t_z b}{\hbar v_F} . $$

This equation suggests that in this approximation, all the orbits are straight lines along the $k_z$-direction and no SCOs appear. Substituting eq. (8) into eq. (3), we perform the integration over $t$. Note that eq. (3) includes $v_z(0)$ and $v_z(t)$. For these equations, we apply the finite values given by eq. (5). Substituting $k_z$ in eq. (5) by eq. (8), we have the time evolution of the $z$-component of the velocity as

$$ v_z(t) = \frac{2t_z c}{\hbar} \sin(ck_z(0)) \tau - \Omega \tau . $$

Here, $\Omega = \omega \sin(\theta) - \eta \omega \cos(\theta) \sin(bk_y(0))$. Using

$$ \int_0^\infty dt \cos(\Omega t) \exp \left( -\frac{t}{\tau} \right) = \frac{\tau}{\tau + (\Omega \tau)^2} , $$

$$ \int_0^\infty dt \sin(\Omega t) \exp \left( -\frac{t}{\tau} \right) = \frac{\Omega \tau}{\tau + (\Omega \tau)^2} , $$

we obtain

$$ \int_0^\infty dt \frac{v_z(t) \exp \left( -\frac{t}{\tau} \right)}{\tau} = \frac{2t_z c}{\hbar} \sin(ck_z(0)) \tau - \Omega \tau \cos(ck_z(0)) \frac{1}{\tau + (\Omega \tau)^2} . $$

Finally, we perform the integration over the Fermi surface. The area element $dS$ is written as $dS = dk_x dk_z$, where $dk_z$ is the line element along the Fermi surface on the $k_z$-$k_x$-plane. Using eq. (2),

$$ dk_z = \sqrt{(dk_x)^2 + (dk_y)^2} = dk_x \sqrt{1 + [\eta \sin(bk_y)]^2} . $$

In addition, using eq. (6), $|v| = v_F \sqrt{1 + [\eta \sin(bk_y)]^2}$. Therefore,

$$ \frac{dS(0)}{\hbar |v(0)|} = \frac{dk_y(0) dk_z(0)}{\hbar v_F} . $$
Combining eqs. (5), (12), (13), and eq. (3) and performing integration over $k_z(0)$ in the first Brillouin zone ($0 \sim 2\pi/c$), we obtain
\[
\sigma_{zz} = 2 \frac{e^2 \tau}{4\pi^2 \hbar v_F} \left( \frac{2\hbar c}{\hbar} \right)^2 \int_0^{2\pi} \frac{d\phi}{1 + (\omega t \sin \theta - \eta \omega t \cos \theta \sin \phi)^2},
\]
where $\phi = \beta \gamma(0)$. Factor 2 arises from the two Fermi surfaces ($\pm$). By integrating over $\phi$, we obtain an analytical expression for the out-of-plane conductivity of the quasi-one-dimensional metal that is placed in a transverse magnetic field as
\[
\sigma_{zz}(B, \theta) = \sigma_{zz}(0) \frac{\sqrt{1 - (\omega t)^2 (\sin^2 \theta - \eta^2 \cos^2 \theta) + \sqrt{1 - (\omega t)^2 (\sin^2 \theta - \eta^2 \cos^2 \theta)^2 + (2\omega t \sin \theta)^2}}}{\sqrt{2} \sqrt{1 - (\omega t)^2 (\sin^2 \theta - \eta^2 \cos^2 \theta)^2 + (2\omega t \sin \theta)^2}}.
\]
Equation (17) is the main result of this study. It expresses the direction in the formula is interesting and important since this single formula and independent of $\eta$. Another special angle is $\theta = \theta_{\text{c}}$ (third magic angle, TMA\cite{Sugawara:2001}), where TMA is defined by
\[
\tan \theta_{\text{c}} = \eta.
\]
Substituting this angle into eq. (17), we have
\[
\rho_\perp(B, \theta) = \rho_\perp(0) \frac{\sqrt{2} \sqrt{1 - (\omega t)^2 (\sin^2 \theta - \eta^2 \cos^2 \theta)^2 + (2\omega t \sin \theta)^2}}{\sqrt{1 + \sqrt{1 + (2\omega t \sin \theta)^2}}},
\]
where $\rho_\perp$ is proportional to $B^{1/2}$ in the high field region. This was pointed out by Lebed and Bagmet in their study\cite{Lebed:1996}.

Hereafter, we compare the experimental results with the theory and discuss the applicability of eq. (17). We prepared needle-shaped single crystals of (TMTSF)$_2$ClO$_4$ with a typical size of $1 \times 0.35 \times 0.3$ mm$^3$ grown using an electrochemical method. The magnetoresistance of the sample was measured at 1.5 K in the relaxed state.

Due to the anion ordering\cite{Sugawara:2001} that occurs below 24 K, the second term of eq. (1) for the relaxed sample is modified as follows:\cite{Sugawara:2001}
\[
\epsilon_j(k_j) = 2t_i \cos k_j \to \pm \sqrt{(2t_i \cos k_j)^2 + \epsilon_{\text{g}}^2},
\]
where $\epsilon_{\text{g}}$ is the anion ordering gap. Therefore, our result from eq. (17) should also be modified. If we assume that the resultant energy band is approximately expressed as
\[
|\epsilon_j(k_j)| \sim \tilde{\epsilon} + 2t_i \cos(2\beta k_j) + \cdots,
\]
in the first two terms of Fourier expansion of eq. (24), we essentially obtain the same result with eq. (17), except for the following differences: $t_i \to \tilde{t}_i$ and $b \to \tilde{b} = 2b$. In the following discussion, we use $\tilde{t}_i$ in the place of $t_i$ and $b$ in the place of $\tilde{b}$.

According to the temperature dependence of the resistance, this material is a clean metal. The resistance decreases by almost three orders of magnitude when cooled from room temperature to 1.5 K. Even below 20 K, it changes by one order of magnitude. Two sets of experiments were conducted. In the first experiment, a magnetic field with a fixed strength was rotated in the layer ($x$-$y$-plane) and the angular dependence of the magnetoresistance was measured. The
results are shown in Fig. 2, where we observe that the resistance strongly depends on the magnetic field direction. To compare the results with eq. (17), we calculated $\rho_\|$, using two fitting parameters, $\omega_r$ and $\eta$. We used $\eta = 0.265$ and $\omega_r = 7.65$ for a field of 7 T. They were selected in order to provide the best fit for the data under the field of 7 T. As observed in the left panel of Fig. 3, the theory reproduces the resistance curves over the entire region of $\theta$ and in a wide region of the field strength (3 to 7 T). The reproducibility is also fairly good in the region $\theta \sim 0$, as demonstrated in the expanded figure (right panel of Fig. 3). Note that eq. (17) reconstructs the “third angular effect” when $\omega_r$ is sufficiently large.

In the second experiment, the magnetic field direction is fixed at the three angles discussed previously, and the field dependence of the resistance was measured. The results depicted in Fig. 4 indicate that the agreement between theory and experiment is almost perfect.

Here, we emphasize that the analytically expressed eqs. (20) and (21) allow us to evaluate the anisotropic parameters $\eta$ and $\zeta$ of this material. This, in turn, provides us information on the transfer energies $t_\parallel, t_\|$, and $t_\perp$. Rewriting eqs. (20) and (21) as $\rho_\|(B, \pi/2)/\rho_\|(0) - 1 = \beta_1 B, \sqrt{\rho_\|(B, 0)/\rho_\|(0) - 1} = \beta_2 B$, respectively, $\eta$ is given by the ratio $\beta_2/\beta_1$. We estimated $\beta_1$ and $\beta_2$ from Fig. 4 and obtained $\eta = 0.221$. This value is comparable with the fitting parameter $\eta = 0.265$ used in Fig. 3. Using $\beta_3$ defined by $\rho_\|(B, 0) = \rho_\|(0) + \beta_3 B^2$, $\zeta$ can be expressed as $\zeta = \sqrt{\pi b^2 c^2 \eta/e\sqrt{\beta_3}}$. By substituting $b = 7.638$ Å, $c = 13.123$ Å, $\beta_3$ determined from Fig. 4, and $\eta = 0.221$ into this equation, we get $\zeta = 0.00788$. By combining eq. (10) and eq. (19), we get $t_\parallel/t_\perp = (\eta/\zeta)(c/b)$. Using $\eta = 0.221$ and $\zeta = 0.00788$, we obtain $t_\parallel/t_\perp = 48.2$. Further, we evaluate $v_F$. Considering that the energy band of (TMTSF)$_2$ClO$_4$ is quarter filled, we obtain $v_F = \sqrt{2t_\parallel a}/h$. Replacing $v_F$ in eq. (10) and using $\eta = 0.221$, we obtain $t_\parallel/t_\perp = (a/b)\eta/\sqrt{2} = 0.0725$. Finally, we obtain $t_\parallel = 0.0725t_\perp = 0.265$ eV and $t_\perp = 1.5$ K. The theoretical curves are indicated by broken lines. The values of the parameters are the same as those used in Fig. 3.
In conclusion, we found an analytical formula that expresses the out-of-plane resistance in the transverse magnetic field for quasi-one-dimensional electron systems. It is applicable for the magnetic field along an arbitrary direction in the layer. The theory was verified by comparison with the experimental results for the organic conductor (TMTSF)$_2$ClO$_4$. The agreement of theory and experiment was found to be satisfactory. We emphasize that the formula given by eq. (17) represents the standard type of magneto-resistance for quasi-one-dimensional electron systems. It will be applied to several quasi-one-dimensional layered metals such as TMTTF- or DMET-based organic conductors.

Acknowledgment

This work is partially supported by a Grant-in-Aid for Scientific Research on the priority areas of molecular conductors (No. 15073222) from the Ministry of Education, Culture, Sports, Science and Technology, Japan.