Out-of-Plane Resistance of Quasi-Two Dimensional Metal
(BEDT-TTF)$_3$Cl(DFBIB) in Transverse Magnetic Fields

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We investigated the magneto-resistance effect on an organic conductor, (ET)$_3$Cl(DFBIB), which has a quasi-two-dimensional electron system. The out-of-plane resistivity in transverse magnetic fields was found to obey a simple formula that was derived by Schofield and Cooper [Phys. Rev. B 62 (2000) 10779]: $\rho_{\perp}(B) = \rho_{\perp}(0) \sqrt{1 + \beta B^2}$, where $\rho_{\perp}(B)$ is the magnetic field ($B$)-dependent resistivity and $\beta$ is a parameter that depends on the material and the temperature. To our knowledge, this is the first example of quasi-two dimensional metals, the magnetoresistance of which is explained by an analytical formula in wide ranges of temperature and magnetic field.

KEYWORDS: 2D metal, transverse magneto-resistance, out-of-2D-plane resistance, (ET)$_3$Cl(DFBIB)

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The development of conductors with layered crystal structures, such as high Tc superconductors or organic conductors, has stimulated the investigation of quasi-two dimensional (quasi-2D) electron systems. In those investigations, magnetoresistance measurement has been a powerful tool to clarifying the characteristics of the electron system. Magnetoresistance also serves as a sensitive detector of anomalies in the electron system. If we encounter an anomaly in the magnetoresistance effects, we expect an unconventional situation in the electron system which requires detailed investigation. To recognize an anomaly, however, we must know the normal magnetoresistance. Unfortunately, our knowledge on the magnetoresistance for quasi-2D metals is incomplete, and therefore, we sometimes cannot recognize anomalous magnetoresistance. The purpose of this work is to establish the “basic” or “standard” type of magnetoresistance for quasi-2D metals. In this paper, we focus on the transverse magnetoresistance for current in the direction normal to the 2D plane. Experiments were performed on an organic conductor, (ET)$_3$Cl(DFBIB)$^{1,2}$ This material was selected because it is a good metal with a quasi-2D electron system. The energy band is nearly isotropic in the 2D plane, and the effect of electron correlation is expected to be weak. Moreover, we can obtain crystals with low impurity density.

We also searched for a formula to express the magnetoresistance. What we are concerned with here are layered metals, the conductivity of which in the direction normal to the 2D plane is much less than that in the 2D plane. They have quasi-2D Fermi surfaces of a cylindrical shape that opens in the direction normal to the 2D plane. Hence, three dimensional pictures assuming parabolic energy bands with anisotropic masses are not applicable. For those systems, Schockley’s tube integral$^3$ is valid for calculating the conductivity tensor. Such a calculation was performed by Schofield and Cooper$^4$ and used to explain the magnetoresistance of Sr$_2$RuO$_4$. On a simplified model system, they succeeded in obtaining an analytical formula for the out-of-2D-plane resistivity in the transverse magnetic field. Experimental results of the out-of-2D-plane resistivity in (ET)$_3$Cl(DFBIB) are explained almost perfectly by this formula. We believe that this is the first example of quasi-2D metals which exhibit magnetoresistance that obeys the formula in wide ranges of temperature and magnetic field. We claim that this type of magnetoresistance is “basic” or “standard” for quasi-2D metals when the current is in the out-of-2D-plane.

Now, we follow the discussion in ref. 4 and derive a formula for the out-of-2D-plane resistivity of quasi-2D metals placed in a transverse magnetic field. We go into some details of the calculation to clarify the limit of applicability of the formula. Take an electron system with the energy spectrum

$$\epsilon = \frac{\hbar^2}{2m} k^2 - 2t \cos(bk_z),$$

where $\mathbf{k}$ is the wave vector in the 2D plane ($k_x,k_y$ plane), and $m$ is the mass of an electron which is assumed to be isotropic in the 2D plane. The second term on the right-hand side expresses the interlayer electron transfer. Here, $b$ is the lattice constant in the $z$ direction and $t$ is the interlayer transfer integral. Assuming $t$ to be much less than the Fermi energy $\epsilon_F$, this energy spectrum gives a quasi-2D Fermi surface with a small corrugation along the $k_z$-direction.

As was demonstrated by Yagi et al.$^5$ in their work on clarifying the origin of angular-dependent magnetoresistance oscillation (AMRO),$^7$ the transport properties of quasi-2D metals are well explained in terms of the semi-classical theory based on the Boltzmann equation.

For the current in the $z$-direction and the magnetic field in the $x$-direction, the $z$-component of the conductivity tensor is written as a tube integral,

$$\sigma_{zz} = \frac{\epsilon^2}{4\pi^2 \hbar^2} \int_{-k_x}^{k_x} \text{d}k_x \frac{m_1}{\omega H_1} \int_0^{2\pi} v_z(\phi) \text{d}\phi \times \int_0^{\infty} v_z(\phi - \phi') \text{exp} \left( -\frac{\phi'}{\omega H_1 \tau} \right) \text{d}\phi'.$$
Here, $v_z = (\partial \epsilon / \partial k_z)/\hbar$ is the group velocity of an electron. (Parameters $\omega_H$ and $m_H$ are defined later.) To perform this calculation, we must know the electron orbits in the $k$-space.

In a magnetic field in the $x$-direction, it is given as a line where a plane normal to the magnetic field (which is expressed as $k_z = k_{z0}$) cuts the Fermi surface. An example of the orbit is depicted in Fig. 1. In the present configuration of the magnetic field, most orbits are open in the $k_z$ direction. The integral in eq. (2) over $\phi$ and $\phi'$ is taken along those orbits. The variables $\phi$ and $\phi'$ are the phase parameters denoting the position of an electron on the orbit. It is defined as $\phi = 2\pi r^*/T^*$. Here, $T^*$ is the time taken by an electron to travel from a point on the orbit (for example, point a in Fig. 1) to travel across the Brillouin zone and return to the initial position. (For example, point d. Note that points a and d are identical in the reduced zone scheme.) $r^*$ is the time required for an electron starting from the origin on the orbit (for example, point a) to reach a particular point (for example, point c). Other parameters appearing in eq. (2) are defined as $\omega_H = 2\pi T^*/m_H$ and $m_H = eB/\omega_H$.

In the calculation of eq. (2), we meet two types of electron motions. One is the motion in the $k$-space, which is caused by the magnetic field applied in the $x$-direction. In the limit of $t = 0$, electrons move on straight lines along the $k_z$ direction with the velocity $dk_z/dt = eBv_z/h = eBk_z/m$. When $t$ is finite but small, the electron motion in the $k$-space is primarily determined by the 2D energy spectrum with no corrugation in the Fermi surface. Warping of the Fermi surface induces a small perturbation unless the space is primarily determined by the 2D energy spectrum $d$. The electron energy spectrum is simple. It has a quasi-2D material are as follows. 1) It is a good metal. As shown in Fig. 2, the resistivity drops by about four orders of magnitude as the system is cooled from 300 to 1.5 K. 2) The electron energy spectrum is simple. It has a quasi-2D

$T^*$ is known, $\omega_H$ and $m_H$ are obtained by putting $T^*$ into $\omega_H = eB/T^*$ and $m_H = eB/\omega_H$. Integration of $v_z = 2t\sin(bk_z)/\hbar$ over $\phi$ also becomes easy because, in this approximation, we can set $k_z = \phi/b$ which otherwise is a complex function of $\phi$.

In this approximation, the three-dimensional nature of the system which makes the conductivity along the $z$-direction finite, is brought into the calculation through $v_z$. By substituting $v_z = 2t\sin(bk_z)/\hbar = 2b\sin(\phi)/h$ and parameters $\omega_H$ and $m_H$ into eq. (2) and integrating, we obtain an expression for the $zz$-component of the conductivity tensor $\sigma_{zz}(B) = 1/m_2 \frac{2\pi e^2}{h} \tau \frac{(2b)^2}{\hbar} \frac{1}{\sqrt{1 + (bk_{z0} \tau)^2}}$ (3)

where $\omega_c = eB/m$. (The same result should be obtained from eq. (11) in ref. 6 by taking the limit, $\theta \to 90^\circ$.) To compare this theory with the experiment, we must know $\rho_{zz}$, which generally is not equal to the inverse of $\sigma_{zz}$ because of build up of Hall voltage. In the present case, however, the Hall voltage exerts only a negligible effect since the crystal is very thin. Hence, we have

$$\rho_{\perp}(B) = \frac{1}{\sigma_{zz}(B)} = \rho_{\perp}(0) \sqrt{1 + (bk_{z0} \tau)^2}$$

(4)

where $\rho_{\perp}(0) = (e^4/2m^2\tau^2 h)$, and $\tau$ is the mean free time. This formula applies to quasi-2D metals that have the electron energy spectrum given by eq. (1) with a small $t$, i.e., $t \ll \epsilon_F$. It covers a wide range of magnetic fields from low magnetic fields where $bk_{z0} \tau \ll 1$ to higher fields where $bk_{z0} \tau \gg 1$. This formula loses its validity when the effect of the closed orbits that appear around $k_z = \pm k_F$ (refer to Fig. 1) is strong. Therefore, in the high-field limit where electrons on the closed orbits contribute dominantly to transport, the magneto-resistance will not obey eq. (4). Note that in this theory, the effect of temperature is introduced through the scattering lifetime $\tau$. The applicability of this assumption should be checked in the experiment.

In the following, we present experimental results and show how they are understood in terms of eq. (4). Single crystals of (BEDT-TTF)$_2$(DFBFI)$_2$ were prepared by electrochemical oxidation. Characteristic features of this material are as follows. 1) It is a good metal. As shown in Fig. 2, the resistivity drops by about four orders of magnitude as the system is cooled from 300 to 1.5 K. 2) The electron energy spectrum is simple. It has a quasi-2D
Fermi surface with an area 50% of the first Brillouin zone. According to the AMRO data, the Fermi surface is a slightly distorted ellipse with the ratio of the long axis to the short axis of 1.23. The carrier mass determined from the Schubnikov–de Haas oscillation and cyclotron resonance is about twice the free electron mass. The interlayer electron transfer is much less than the intralayer transfer, as evidenced by the fact that we can clearly observe AMRO.

We performed magnetoresistance measurements using single crystals of (BEDT-TTF)$_3$Cl(DFBIB). Parallelepiped samples with the typical dimensions of $0.5 \times 0.5 \times 0.1$ mm$^3$ were used. The resistance measurements were carried out using a conventional DC method with the electrical current in the $b$-crystal axis, which is normal to the 2D plane. Magnetic field was applied in the direction parallel to the 2D plane. We did not specify the direction of the field in the 2D plane because only a small difference in the magneto-resistance was observed when the field was rotated in the 2D plane.

Experiments were performed at several temperatures ranging from 4.2 to 50 K. In this region, the resistivity varies by about two orders of magnitude. With this large change in resistivity, $\rho_\perp$ vs $B$ curves exhibit a qualitative change in their appearance. At low temperatures below 12 K, the magnetic field dependence of curves changes with increasing field. In the low-field region, the increment of resistivity is proportional to $B^2$. In the high-field region, on the other hand, the curve becomes linear. As the temperature rises, the boundary separating low- and high-field regions moves to higher fields. Above 14 K, the boundary goes to a field beyond 5 T and we can see only the low-field behavior of resistivity. Equation (4) is used to explain these experimental results as follows. At low fields where $bk_F \tau \ll 1$, the resistivity is approximated as

$$\rho_\perp(B) \approx \rho_\perp(0) + \rho_\perp(0) \frac{1}{2} (bk_F \tau)^2.$$  \hspace{1cm} (5)

This gives a quadratically rising resistivity with increasing $B$. On the other hand, for high $B$ where $bk_F \tau \geq 1$, eq. (4) gives

$$\rho_\perp(B) \approx \rho_\perp(0) bk_F \tau = \frac{\pi \hbar^2 k_F}{2e m^* \tau} B = \alpha B.$$  \hspace{1cm} (6)

where $\alpha = (\pi \hbar^2 k_F)/(2e m^* \tau)$. According to this formula, $\rho_\perp$ in high fields traces a straight line that passes through the origin of the graph. Note that the expression of slope $\alpha$ does not contain scattering time $\tau$. Therefore, in sufficiently high fields, curves for different temperatures are expected to have a straight line as the asymptote. Actually, this is seen in Fig. 3 for curves below 12 K. The asymptote is shown by the broken line.

A more interesting and impressive plot of the data is shown in Fig. 4. In this figure where $\rho^2_\perp(B)$ is plotted against $B^2$, we see that all the curves are straight lines over the entire region of $B^2$ from 0 to 25 T$^2$. Moreover, the slopes of the lines appear almost independent of temperature. This is verified by detailed analysis of the data, as shown in Fig. 5. Between 4.2 and 50 K, the slope changes only by a factor of 1.5. This interesting characteristic of $\rho_\perp$ is understood in terms of eq. (4). Squaring both sides of eq. (4), we obtain...
values of \(m, b\) and \(k_F\), and hence we can estimate the transfer integral \(t\) and the scattering lifetime \(\tau\). The transfer integral \(t\) is determined from the slope of the line in Fig. 4. It is written as \(\alpha^2 = (\pi h k_F)^2/(2m e^2 t^2)\). Using the values of \(k_F\) and \(m\), we obtain \(\tau \approx 9.08 \times 10^{-4} \text{eV} = 12.5 \text{K}\).  

Another parameter we can estimate is the scattering time \(\tau\). From eq. (4), we obtain \((\rho_{\perp}(B)/\rho_{\perp}(0))^2 = 1 + (bk_F M/ m^2 B^2)\). The scattering time \(\tau\) is estimated from the slope of \((\rho_{\perp}(B)/\rho_{\perp}(0))^2\) vs \(B^2\) curves. For example, \(\tau\) at 4.2 K is estimated to be \(6.06 \times 10^{-12} \text{s}\). With increasing temperature, it becomes short. At 50 K, it is about \(8.4 \times 10^{-14} \text{s}\). Note that the energy corresponding to this scattering time \((h/\tau)\) is about 7.9 meV (= 91.2 K). It is much larger than the interlayer transfer integral \(t\) (= 12.5 K). In such a situation, the concept based on the band structure will not be applicable and the interlayer transport will be incoherent. Nevertheless, the experimental results obey eq. (4) with the scattering time \(8.4 \times 10^{-14} \text{s}\). Concerning this problem, we can refer to the work of McKenzie and Moses.  

They performed theoretical calculation on the out-of-2D-plane conductivity assuming the incoherent situation and showed that the results of the calculation based on the Boltzmann equation can apply to the interlayer magnetoresistance not only in the coherent region but also in the weakly incoherent region.  

To summarize our work, we found that the resistivity of quasi-two-dimensional metals for the out-of-2D-plane current in the transverse magnetic field is expressed by a simple formula, \(\rho_{\perp}(B) = \rho_{\perp}(0)\sqrt{1 + (bk_F M/ m^2 B^2)}\). Experimental results are perfectly explained by this formula. Therefore, we claim that this is the basic type of magnetoresistance that we sought. Deviation of experimental results from this expression must be a signal of some anomaly, which may simply be due to a too large transfer integral, or a result of a complex Fermi surface. Otherwise, electron correlation may be responsible for the anomaly. In any case, in such a situation, detailed investigations are required.  

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16) $m = 2.1m_e$; $b = 1.502\,\text{nm}$. $k_F$ is estimated from the carrier density given by the energy band structure. Assuming the band is isotropic in the 2D plane, we have $k_F = 2.67 \times 10^7 \,\text{cm}^{-1}$.

17) This experiment can be used to estimate the interlayer transfer integral $t$. A method of determining $t$ different from the present method was proposed by Hanasaki et al. It utilizes the small closed orbit (SCO) effect. Although Hanasaki et al.’s method is powerful, it requires a high magnetic field or clean samples so that carriers can complete a circular motion along the SCO. Our method, on the other hand, can be applied to samples with low carrier mobility.
