Transport Phenomenon of Multilayer Zero-Gap Conductor
in the Quantum Limit

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Mysterious transport phenomena observed in a zero gap conductor \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) were investigated. In particular, the electron system in the quantum limit where electrons in the Landau level with index 0 dominate the transport was of interest. In order to determine the magnetic field dependence of the electron transport, we utilized a conductivity tensor. We showed how each component of the conductivity tensor changes with the magnetic field. All the components were found to be expressed by simple functions of magnetic field. Interlayer Hall resistance (Hall effect observed in the interlayer current) was observed for the first time.

KEYWORDS: interlayer Hall effect, zero-gap conductor, \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\), quantum limit, conductivity tensor

Conductors in which electrons have energy dispersion with a cone like structure (refer to Fig. 1) is called zero gap conductors. Today, we have two intrinsic zero-gap conductors, graphene\(^1\) and \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\)\(^2\).\(^3\)\(^4\) \((\text{hereafter, abbreviated as } \alpha\text{-ETI}_3)\). Both have a Dirac cone-type energy spectrum with the Fermi energy at the contact points of cones so that at zero \(K\), the valence band (bottom cone) is fully occupied and the conduction band (top cone) is empty. The energy \(e\) spectrum of electrons on the Dirac cone is expressed as \(e = e_F \pm \hbar v_F |k|\), where \(e_F\) is the Fermi energy and \(k\) is the wave number measured from the contact point. It gives the linear energy dispersion passing the contact point. The Fermi velocity \(v_F\) is the parameter that determines the slope of the cone roof. The Dirac cones of \(\alpha\text{-ETI}_3\) are tilted and \(v_F\) depends strongly on the direction of \(k\).

In a magnetic field \(B\) normal to a two-dimensional (2D) layer, Landau levels are formed. The energy level with the Landau index \(N (N = 0, \pm 1, \ldots)\) is expressed as \(\epsilon_N = \epsilon_F + [\text{sign}(N) v_F \sqrt{2 \hbar B |N|}]\). Since the energy is proportional to \(\sqrt{B\!|N|}\), a weak magnetic field can open large energy gaps between Landau levels. For example, in a system with \(v_F = 10^5 \text{ m/s}\),\(^2\)\(^3\) a magnetic field of 1 T opens an energy gap between \(N = 1\) and 0 by about 20 K.

One of the important characteristics of a 2D zero-gap conductor in the magnetic field is the appearance of a zero-mode Landau level. This level (Landau index \(N = 0\)) is formed at the energy of contact points of Dirac cones irrespective of the magnetic field strength.

If the temperature is low or the magnetic field is high, we find electrons only in the zero-mode Landau level. Such a situation is called the quantum limit. The quantum limit is achieved when the energy gap between \(N = \pm 1\) and 0 is much larger than the thermal energy. In the quantum limit, electrons in the zero-mode Landau level dominate the transport. The purpose of this work is to clarify the transport properties of zero mode electrons in the quantum limit. In order to discuss the electric transport, we measured the resistivity tensor for samples in a magnetic field. An attempt to measure several components of the resistivity tensor simultaneously was successfully performed. In particular, interlayer Hall resistance (Hall resistance in the interlayer current and the transverse magnetic field) was observed for the first time. Using the resistivity tensor, we derived the conductivity tensor and compared it with a theoretical value. In the experiment, we used a zero-gap conductor \(\alpha\text{-ETI}_3\). This crystal consists of conductive and insulating layers stacked alternately.\(^5\)\(^6\) It changes to a zero-gap conductor when placed under pressures higher than 1.5 GPa.\(^2\)\(^4\)

In this work, we deal with electrons in the zero-mode Landau level, of which the density given by the Landau degeneracy is about \(2.4 \times 10^{10} \text{ cm}^{-2} \text{T}^{-1}\). For such a low-density system, the effect of spacial fluctuation of carrier density is sometimes not negligible, as is pointed out in the graphene case.\(^7\)\(^8\) In bulk single crystals of \(\alpha\text{-ETI}_3\), on the other hand, we found no indication of such troubles. Therefore we can be used in the investigation of zero-gap conductors with a low-carrier density.

In the following, we discuss the transport phenomena of zero-mode Landau electrons in \(\alpha\text{-ETI}_3\). We are interested in the region where the system is in the quantum limit.

In our experiments, single crystals of \(\alpha\text{-ETI}_3\) synthesized electrochemically were used. The typical dimensions of the samples are \(0.85 \times 1.55 \times 0.06 \text{ mm}^3\). The sample was placed...
in a pressure bombe made of BeCu, with Daphene 7474 oil as the pressure medium. A quasi-hydrostatic pressure above 1.5 GPa was applied to the sample to make the system a zero-gap conductor. This experiment was performed at temperatures between 1.5 and 40 K and at magnetic fields up to 7 T. In this paper, we focus on the experiments performed at 4.2 K.

We measured 5 components of the resistivity tensor ($\rho_{xx}$, $\rho_{yy}$, $\rho_{zz}$, $\rho_{xy}$, and $\rho_{xz}$). In the suffix $\mu v$ ($\mu = x, y, z$, $\nu$ indicates the current direction and $\nu$, the direction in which we measure the voltage difference. We take the $x$- and $y$-axes parallel to the 2D plane and the $z$-axis normal to the layers. The transport phenomena of this material were found to be nearly isotropic in the 2D plane, so that we did not specify the direction of the $x$-axis in the 2D plane. The other four components, $\rho_{xx}$, $\rho_{yy}$, $\rho_{zz}$, and $\rho_{xy}$, were not measured. Instead, we replaced them with the experimental data obtained by assuming the relations $\rho_{a\beta} = \rho_{\beta a} = \rho_{a\alpha} = \rho_{a\alpha} = 0$, and $\rho_{xz} = 0$. The applicability of these relations was checked experimentally. For resistance measurements, a conventional method with the dc current was used. To measure four components of the resistivity tensor simultaneously, eight electrodes were attached to the sample, as shown in Fig. 2. The magnetic field with a fixed strength $B$ was rotated in the $yz$-plane and the resistances were measured as functions of the magnetic field direction $\theta$. Here, the angle $\theta$ was measured from the $y$-axis. The raw data is usually the sum of intrinsic and extrinsic signals due to other components of resistance tensor. The intrinsic signal was determined by using the data obtained at the magnetic fields $\pm B$. Resistance was translated to resistivity. Owing to the difficulty in measuring the distances between electrodes, the absolute values of data include ambiguity with a factor of 3.

In Fig. 3, an example of the experimental results of $\rho$ is presented. Here, we examine $\rho_{x\alpha}$ and $\rho_{y\alpha}$. This is the first experimental result in which the interlayer Hall effect in organic zero-gap conductors is detected successfully (where $\rho_{xz}$ is called the interlayer Hall resistivity). In measuring $\rho_{xz}$, the current is applied in the direction normal to the conductive layers. We realize that signals in Fig. 3 have peaks at around $\theta = 0$ with the same shape and inverse polarity. These data imply that the relation $\rho_{xz} = -\rho_{zx}$ holds. This fact includes important information about how carriers move in the interlayer direction. When the in-plane electric current flows, carriers move in the conductive layers. On the other hand, when the current is in the interlayer direction, electrons move across the insulating layer by the tunneling effect. This difference in the mechanism of electron motion generally gives rise to the different transport phenomena.

For the two signals in Fig. 3 to have the same $\theta$ dependence, the system should be in the dirty limit where the electrons are scattered frequently so that they lose the phase memory before jumping to a neighboring layer.

The next topic to be discussed concerns with the $\theta$ dependence of $\rho_{xz}$. Examining data of $\rho_{xz}$, we found that the data are expressed as $\rho_{xz}(\theta) = a \cot \theta$, where $a$ is independent of the magnetic field. This is demonstrated in Fig. 4. Here, $\rho_{xz}$ is plotted against $\cot \theta$. In the region where $\cot \theta$ is small (for example, in the region $\cot \theta < 5$ for $B = 7$ T), $\rho_{xz}$ is on a straight line that passes the origin. This line expressed as $\rho_{xz}(\theta) = a \cot \theta$ is the asymptote of all the curves in the figure. As $\cot \theta$ increases and exceeds a certain value, the data curve bends downward. In this region, $\theta$ is small and thus $B_{z}$ is small. This gives two effects. First, the energy gap between $N = \pm 1$ and 0 decreases. Secondly, the density of zero-mode electrons decreases. As a result, the system cannot be in the quantum limit. The deviation of the data curve from the straight line is expected to occur in this region.

Now, we proceed to the discussion of transport phenomena based on the conductivity tensor. By measuring $\rho_{xz}$, we obtain all the components of the resistivity tensor, and using them, we derive the conductivity tensor. In Fig. 5, the components of the conductivity tensor are shown as functions of angle $\theta$. In the following, we examine each component of the conductivity tensor.
1) In Fig. 5(a), $\sigma_{xx}$ is plotted as a function of $\theta$. The same data are plotted in Fig. 6 against $B_{\perp}$ ($= B \sin \theta$). In this plot, all the data are on a single curve. This indicates that $\sigma_{xx}$ is a function of $B_{\perp}$ and is almost independent of the parallel component $B_{||}$ ($= B \cos \theta$). The highest conductivity is obtained at the field $B_{||} = 0$. As mentioned above, the field component $B_{||}$ negligibly affects $\sigma_{xx}$. Therefore, the conductivity at the peak is expected to be equal to the value obtained in the absence of a magnetic field. According to Tajima et al.,\textsuperscript{2)} it is close to the quantum conductivity $e^2/(\epsilon_c \hbar) = 2.2 \times 10^{-4}$ S/cm, where $\epsilon_c$ (= 1.75 nm) is the lattice constant in the $z$-direction. The result in Fig. 5(a) is about 1/10 of this value. As the magnetic field $B_{\perp}$ increases, $\sigma_{xx}$ decreases rapidly and saturates to a value that is about 1/8 of the zero-field value. The saturation occurs at the field of around $B_{\perp} = 1$ T. Above 1 T, $\sigma_{xx}$ remains constant up to 7 T. It is reasonable to expect that the system will be in the quantum limit in the region $B_{\perp} > 1$ T. The quantum limit is achieved when the energy gap between the $N = 0$ and 1 Landau levels is comparable to the thermal energy. Substituting $T$ with 4.2 K we obtain $k_B T = v_F \sqrt{2e\hbar B}$, and substituting $B$ with 1 T, we obtain $v_F \sim 2.4 \times 10^5$ m/s.

2) The data of $\sigma_{xy}$ is shown in Fig. 5(b). Except for the data for $B = 0.2$ T, sharp peak structures are seen at $\theta = 0$ and 180°. Between these two peaks, there is a concave region. (When the system is far from the quantum limit, we observe a sinusoidal curve as shown for $B = 0.2$ T.) Figure 7 shows $\sigma_{xy}$ as a function of $B_{\perp}$. In this plot, the data are on a single curve indicating that $\sigma_{xy}$ depends only on $B_{\perp}$ and is independent of $B_{||}$. The signal is inversely proportional to $B_{\perp}$. Hence, we obtain the relation $\sigma_{xy} \propto 1/B_{\perp}$.

3) $\sigma_{zz}$ is difficult to analyze, because it depends both on $B_{\perp}$ and $B_{||}$. Thus, the magnetic field dependence of the signal is complex. In Fig. 5(c), the curve has sharp peaks at $\theta = 0$ and 180°, where the field is parallel to the 2D layer. Referring to the expression $\rho_{zz} = a \cot \theta$, we examined the data and found that the shape of the signal is written as $\sigma_{zz}(B_{\perp}, B_{||}) = b B_{||}/B_{\perp}^2$, where $b$ is independent of magnetic field. Figure 8 shows the data of $\sigma_{zz}$ against $B_{||}/B_{\perp}^2$. The $\sigma_{zz}$ data satisfying $B_{\perp} > 1$ T were selected to plot only the data in the quantum limit. Figure 8 indicates that $\sigma_{zz} \propto B_{||}/B_{\perp}^2$.

4) $\sigma_{zz}$ is an exception and also appears to be a function of $B_{\perp}$. As shown in Fig. 9, in the region $B_{\perp} > 1$ T, where the system is in the quantum limit, $\sigma_{zz}$ is a slowly increasing function of $B_{\perp}$.

In the following, we compare the above results with the theoretical calculation by Osada.\textsuperscript{9)} He calculated the conductivity tensor of a zero gap conductor using the Kubo formula. The anisotropy in the Dirac cone was neglected. The Zeeman effect and electron correlation were not considered. The results are applied to the zero-gap conductors in the quantum limit.

According to Osada, the components of the conductivity tensor ($\sigma_{xx}$,\textsuperscript{10} $\sigma_{yy}$, $\sigma_{zz}$, and $\sigma_{xy}$\textsuperscript{11}) are expressed as

![Graph](image-url)
Here, the function $f(x)$ is nearly equal to 1 except for a very narrow region, where $x$ is close to 0. Thus, we neglected this term. In this calculation, a small amount of carrier doping is assumed. $n_{(2D)}/c_z$ in the equations is the density of doped carriers. $\alpha$ is the factor of the order of 1. $t_e$ is the interlayer transfer energy. $\tau$ is the scattering life time of carriers in the 2D plane. $v_F$ is the Fermi velocity of carriers on the Dirac cone.

These theoretical results agree qualitatively with the experimental findings indicating that when the system is in the quantum limit, 1) $\sigma_{yz}$ does not depend on the magnetic field, 2) $\sigma_{xy} \propto 1/B_{\perp}$, 3) $\sigma_{xx} \propto B_{\perp}^1$. However, this result fails to explain the behavior of $\sigma_{zz}$. The theoretically predicted $\sigma_{zz}$ at a high magnetic field is proportional to $B_{\perp}$. In the experiment, we observed the saturation of $\sigma_{zz}$ at high magnetic fields.

We determined some of the parameters to obtain the quantitative agreement between the experimental results and eqs. (1)–(4). We obtained $\alpha \sim 0.02$, $n_{(2D)} \sim 2 \times 10^{11}$ m$^{-2}$/layers, $t_e/v_F \sim 9 \times 10^{-27}$ J-s/m, and $t_e \tau/h \sim 0.1$, (which was obtained from the slope of the broken line in Fig. 9). $t_e \ll h/\tau$ indicates that the system is in the dirty limit. If we use $v_F \sim 2 \times 10^4$ m/s, we can separately determine $t_e \sim 1$ meV and $\tau \sim 0.05$ ps.

Finally, we discuss the Zeeman effect and the effect due to the tilted Dirac cones. The Zeeman effect works to split the electron energy level and thus reduces the density of zero-mode electrons. This effect is large for the magnetic field parallel to the 2D plane. The dips in Fig. 3 may be due to the Zeeman effect. This effect has been investigated by Tajima and coworkers.\cite{12-16} Tilted Dirac cones can also affect the transport. Morinari et al.\cite{15} however, showed that at a certain magnetic field, the velocity of an electron are averaged so that the electron appears to be an electron on an isotropic cone.

Summarizing this work, we investigated the behavior of zero mode Landau electrons in a zero gap conductor $\sigma$-ET$_3$I$_3$. We performed a simultaneous measurement of 5 components of the resistivity tensor on one sample. Among them is the interlayer Hall resistivity $\rho_{xy}$. This is the first experiment conducted to observe the interlayer Hall resistance in multilayer zero-gap conductors. The resistivity tensor is converted to a conductivity tensor so that we can compare the experimental and theoretical results. The experimental results are well reproduced by the theoretical results obtained by Osada.\cite{9,11} Components of conductivity tensor are found to be expressed by simple functions of the magnetic field. Some material parameters were determined.

In conclusion, we have succeeded in clarifying the magnetic-field-dependent transport of electrons in a zero-gap conductor in the quantum limit.

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