Magnetotransport of Massless Dirac Fermions in Multilayer Organic Conductors

T Osada, K Uchida, and T Konoike
Institute for Solid State Physics, University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan
E-mail: osada@issp.u-tokyo.ac.jp

Abstract. Magnetotransport has been considered in multilayer massless Dirac fermion systems, in which two-dimensional (2D) layers with a pair of Dirac-cone dispersion stack with weak interlayer coupling. The Fermi level is assumed to be fixed at the Dirac point resulting in the zero-gap conductor. At the high-field quantum limit, where only the $n=0$ Landau level at the Dirac point is partially occupied, all elements of conductivity tensor have been evaluated by the lowest order contribution of interlayer coupling. We have deduced saturation of in-plane resistance, local maxima of in-plane Hall resistance, negative interlayer magnetoresistance, and $\cot\theta$-type unusual angle-dependence of interlayer Hall resistance. These results propose good explanations for mysterious transport features observed in organic zero-gap conductors, $\alpha$-(BEDT-TTF)$_2$I$_3$ and its related compounds.

1. Introduction
The massless Dirac fermion system in solids has attracted a great deal of attention since monolayer graphene devices were realized and the half-integer quantum Hall effect was demonstrated using them [1, 2]. The enormous amount of research has been done on magnetotransport of two-dimensional (2D) massless Dirac fermions in graphene [3]. On the other hand, just around the same time, another massless Dirac fermion system was discovered in layered organic conductors $\alpha$-(BEDT-TTF)$_2$I$_3$ and its related compounds, where BEDT-TTF denotes bis(ethylenedithio)-tetrathiafulvalene. In $\alpha$-(BEDT-TTF)$_2$I$_3$ crystals, BEDT-TTF molecules and I$_3$ anions form conducting layers and insulating layers, respectively, and they stack alternately. Since the interlayer coupling is very weak (the interlayer transfer energy $t_c$ is much smaller than 1 meV), the electron system is regarded as a quasi-2D system with no coherent interlayer coupling down to low-temperature. Based on the band calculation using pressure-dependent band parameters, it was theoretically predicted that these compounds become anisotropic massless Dirac fermion systems under high pressures [4]. The Fermi level is always located at the band contact point (Dirac point) of 2D Dirac cone dispersion. In other words, these systems are zero-gap conductors [5]. In fact, the charge ordering transition at $T=135$ K observed at ambient pressure is suppressed at high pressures above 15 kbar, suggesting the change to the zero gap conductor with very small carrier density.

Before the discovery of Dirac fermion, it has been known that $\alpha$-(BEDT-TTF)$_2$I$_3$ and its related compounds exhibit anomalous magnetotransport features at low temperatures. We set the z-axis along the stacking direction normal to the conducting plane ($xy$-plane). The in-plane diagonal resistivity $\rho_{xx}$ exhibits saturation behavior in high magnetic fields [5], whereas the in-plane Hall resistivity $\rho_{yx}$ deviates from $B$-linear behavior and shows a local maxima at high fields [6]. The interlayer diagonal
resistivity \( \rho_{zz} \) shows remarkable decrease with increasing magnetic fields [7]. Namely, interlayer transport shows negative magnetoresistance at high magnetic fields. In addition, recently, Kajita and Sato et al. have reported another remarkable behavior of interlayer Hall resistance \( (\rho_{zz}) \) [8]. They have measured the off-diagonal resistivity \( (\rho_{zz}=E_{zz}/J_z) \) of \( \alpha-(BEDT-TTF)_2I_3 \) as a function of strength and direction of the magnetic field. Under sufficiently strong magnetic fields, \( \rho_{zz} \) seems to depend not on magnetic field strength but only on magnetic field direction. It shows unusual angle-dependence proportional to \( \cot\theta=B_z/B_x \) \( (B_x > 0) \), where \( \theta \) denotes the elevation angle from the \( y \)-axis (Hall configuration) to the \( z \)-axis. The origins of above mysterious magnetotransport features have been unsolved questions for a long time.

In this paper, we clarify magnetotransport behaviors of multilayer massless Dirac fermion systems in the high-field quantum limit where only the \( n=0 \) Landau level exists at the Fermi level. Particularly, we concentrate on the interlayer transport which is additional characteristic of multilayer systems to those of monolayer systems like graphene. We employ the tunneling picture which treats interlayer coupling as a perturbation, and we discuss its lowest order contribution to conductivity. We show that almost all of magnetotransport features observed in \( \alpha-(BEDT-TTF)_2I_3 \) can be explained as those of multilayer Dirac fermion systems at the quantum limit.

2. Tunneling picture for magnetotransport in multilayer massless Dirac fermion system

We consider the multilayer massless Dirac fermion system of which dispersion around the Fermi level \( E_F=0 \) is given by \( H(\mathbf{k})=\hbar v(\mathbf{k},\sigma_1+k_0\sigma_2)-2t_c\cos k_0\sigma_0 \) [9, 10]. Here, \( \mathbf{k}=(k_x, k_z) \) is the 2D wave number measured from the band-contact point (Dirac point) \( \mathbf{k}=\mathbf{k}_0 \) (or \( \mathbf{k}=-\mathbf{k}_0 \)). \( v \) is the in-plane group velocity of isotropic Dirac cones, and \( t_c \) and \( \sigma_0 \) denote interlayer transfer energy and interlayer distance, respectively. \( \sigma_1 \) and \( (\sigma_0, \sigma_x, \sigma_y) \) are the unit matrix and Pauli spin matrices. Under the general magnetic field \( \mathbf{B}=(B_x, B_y, B_z) \), the effective mass Hamiltonian \( H_{\text{eff}}=H_0+H' \) is given by

\[
H_0 = \hbar v \begin{pmatrix}
0 & \left(-i\partial_x + eB_x/z/h\right) & \left(-i\partial_y + eB_y/z/h\right) \\
\left(-i\partial_y + eB_y/z/h\right) & 0 & \left(-i\partial_z + eB_y/z/h\right) \\
\left(-i\partial_z + eB_y/z/h\right) & \left(-i\partial_y + eB_y/z/h\right) & 0
\end{pmatrix},
\]

\[
H' = -2t_c \cos(-ic\partial_z) \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}. \tag{1}
\]

Here, we have chosen a gauge as \( \mathbf{A}=(B_z, B_x, -B_z, 0) \). We assume \( B_z > 0 \) without any loss of generality. In the present model, spin splitting due to Zeeman effect is not taken into account. The unperturbed Hamiltonian \( H_0 \) gives the free Landau levels of 2D Dirac fermion system on each layer located at \( z=z_i \). The quantum numbers of these unperturbed states are the Landau index \( n \), the center coordinate \( x_0 \), and the layer position \( z_l \). Their eigen energies are given by

\[
E_{n,x_0,z_l} = \text{sgn}(n)\sqrt{2\hbar v/\ell}\sqrt{|n|}. \tag{2}
\]

Here, \( \ell = \sqrt{\hbar/eB_z} \) is the magnetic length. We also assume the existence of two Dirac cones at \( \mathbf{k}_0 \) and \( -\mathbf{k}_0 \) so as to satisfy time-reversal symmetry. So, each of Landau states has two-fold spin degeneracy and two-fold valley degeneracy. To reproduce the finite Hall effect, we consider very small but finite carrier imbalance \( n^{(2D)} \) due to extrinsic origin such as charged impurities. The sheet carrier density per single layer \( n^{(2D)} \) is defined positive for electrons \( (E_F>0) \) and negative for holes \( (E_F<0) \). When \( |n^{(2D)}| \) becomes smaller than one half of the Landau level degeneracy \( 4/2\pi^2 \) (including spin and valley degeneracy), the system reaches the quantum limit where the Fermi level exists in the \( n=0 \) Landau level and only the \( n=0 \) Landau level is partially occupied. Note that in the present Dirac fermion
system, the quantum limit is easily realized even in low magnetic fields because of large Landau level spacing between \( n=0 \) and \( n=1 \) (or \( -1 \)) levels and small carrier density \( |n^{(2D)}| \).

We treat the interlayer coupling \( H' \) in (1) as a perturbation. We call this method the tunneling picture. This picture is effective when interlayer coupling is weaker than in-plane scattering resulting in incoherent interlayer coupling. The interlayer coupling is responsible for interlayer transport and it causes broadening of unperturbed Landau levels together with scattering. At the quantum limit, the perturbation matrix elements relating to the \( n=0 \) Landau level are important. They can be easily obtained using eigen states of \( H_0 \). They give the following selection rule of interlayer tunneling:

Tunneling is only allowed between neighboring layers \((z_i'=z_i\pm c)\), and the center coordinate of the final state on the layer at \( z=z_i \pm c \) satisfies \( x_0'=x_0\pm(\frac{B_x}{B_z})c \), where \( x_0 \) is the center coordinate of initial state on the layer at \( z=z_i \). So, the direction connecting the center coordinates of the initial and final states is parallel to the projected magnetic field on the \( xz \)-plane. The tunneling amplitude is proportional to the overlap integral of the initial and final states. The matrix elements of interlayer current \( j_z = (-e)(1/\hbar)[z, H'] \) are proportional to those of interlayer tunneling \( H' \).

Each element of the conductivity tensor can be calculated using Kubo formula. In the tunneling picture, each conductivity element is expanded into a power series of interlayer coupling. In the case of weak interlayer coupling \((t_c<<\hbar/\tau \) : the dirty limit), the lowest order contribution gives a good approximation of the conductivity. So, we have evaluated all conductivity elements by the lowest order contribution, \( i.e., \) the leading term of expansion at the quantum limit where only the \( n=0 \) Landau level is located at the Fermi level. Figure 1 shows the leading terms of interlayer diagonal conductivity \( \sigma_z \) and off-diagonal conductivity \( \sigma_{xz} \). Note that interlayer current \( j_z \) changes the layer \( z_i \) but in-plane current \( j_x \) and \( j_y \) do not change the layer. They give the following analytic formulae for d.c. interlayer conductivity elements \((B_z>0)\) [9, 10].

\[
\sigma_z(0) = \frac{8e^2c e^{1/2} \tau_z^2}{\pi^2 \hbar^2} B_z e^{\frac{1}{2\hbar} \beta_z^1 B_z^2} \tag{3}
\]

\[
\sigma_{xz}(0) = -\frac{t_z^2 c e^{1/2} \tau_x^2}{\hbar^2 \tau_z^2} B_z e^{\frac{1}{2\hbar} \beta_x^1 B_z^2} \tag{4}
\]

Here, we assumed low temperature \((k_B T<<\hbar/\tau) \) and the dirty limit \((t_c<<\hbar/\tau) \). The exponential factor results from the overlap of \( n=0 \) wave functions. Except this factor, \( \sigma_{xz} \) is proportional to the normal magnetic field \( B_z \), reflecting the degeneracy of the \( n=0 \) Landau level. The factor \( B_z/B_z^2 \) in \( \sigma_{xz} \) also results from the overlap between \( n=0 \) and \( n=\pm 1 \) wave functions.
As for the in-plane conductivity elements $\sigma_{xx}$ and $\sigma_{xy}$, their leading terms are the 0-th order of interlayer coupling. They have already been discussed for graphene using self-consistent Born approximation by Shon and Ando [11]. According to their results, 2D diagonal conductivity $\sigma_{xx}^{(2D)}$ takes a constant value $2e^2/\pi^2$ independent on temperature and magnetic field. This is called the universal conductance of graphene. So, using their results, we can set $\sigma_{xx} = 2\alpha e^2/\pi^2$ and $\sigma_{xy} = -en^{(2D)}/cB_z$, where $\alpha (~1)$ is an empirical correction factor for multilayers.

3. Comparison with experiments on organic Dirac fermion systems
Since we have obtained all elements of conductivity tensor, we can discuss magnetotransport of the multilayer Dirac fermion system in any configuration. Figure 2 shows the calculated field-dependence of in-plane resistivity. The in-plane diagonal resistivity $\rho_{xx}$ shows saturation to the constant value of $1/\sigma_{xx}$. The amplitude of the in-plane Hall resistivity $\rho_{yx}$ exhibits the local maximum corresponding to the crossover of $\sigma_{xx}$ and $\sigma_{xy}$. On the other hand, as shown in Fig. 3, the interlayer diagonal resistivity $\rho_{zz}$ shows remarkable negative magnetoresistance. This feature was already reported in our previous paper [9], and calculation details are described there.

One of the most remarkable magnetotransport features in the multilayer Dirac fermion system is anomalous dependence of interlayer off-diagonal resistivity $\rho_{zx}$ on the magnetic field strength and orientation. $\rho_{zx}$ at the quantum limit is analytically written as

$$\rho_{zx} = -\alpha \frac{h}{4e^2 \sqrt{n^{(2D)}}} \left( \frac{B_z}{B_y} \right) \frac{1}{1 + \left(2\alpha eB_z / \pi^2 h n^{(2D)}\right)^2}.$$

Here, we assumed $B_z > 0$ and that the magnetic field is rotated in the $yz$-plane normal to the current. When the normal field component $B_z$ is less than $\pi^2 h n^{(2D)}/2ae$, which is the lower bound of quantum limit region, the last factor in (5) can be regarded as 1, so that $\rho_{zx}$ is almost proportional to $B_y/B_z = \cot \theta$, where $\theta$ is the elevation angle of magnetic field from the conducting plane. This means that $\rho_{zx}$ depends only on the field direction $\theta$ and it has no dependence on the field strength. In the field configuration parallel to the $y$-axis ($\theta=0$), $\rho_{zx}$ gives the interlayer Hall effect. So the absence of field-strength-dependence is very anomalous behavior. The $\cot \theta$-type angle-dependence is also anomalous

**Figure 2.** Calculated field-dependence of in-plane diagonal resistivity $\rho_{xx}$ and Hall resistivity $\rho_{yx}$ at the quantum limit. Dotted lines indicates the inverse of $\sigma_{xx}$ and $\sigma_{xy}$.

**Figure 3.** Calculated field-dependence of interlayer diagonal resistivity $\rho_{zx}$ at the quantum limit. The interlayer spacing $c=1.57$nm is assumed. (after [9])
Figure 4. Calculated angle-dependence of interlayer off-diagonal resistivity $\rho_{zx}$ at the quantum limit. (a) Angle-dependence of $\rho_{zx}$ at several field values. Inset shows the configuration of the system. (b) cot$\theta$-dependence of $\rho_{zx}$ at several field values.

since semiclassically cos$\theta$-type dependence is expected. In higher fields in the quantum limit region, $\rho_{zx}$ shows field-strength dependence. Figure 4(a) illustrates the angle-dependence of $\rho_{zx}$ for several field values. We can see that $\rho_{zx}$ deviates from the exact cot$\theta$-dependence (limit of $B_{z}=0$) in higher fields. However, the deviation is not large, so that the angle-dependence is approximately cot$\theta$-like. This feature is more clearly seen in Fig. 4(b), in which $\rho_{zx}$ is plotted as a function of cot$\theta$.

The above results obtained in this work explain all of mysterious magnetotransport behaviors observed in $\alpha$-(BEDT-TTF)$_2$I$_3$, which were mentioned in the first part, qualitatively very well. They are the characteristic phenomena of multilayer Dirac fermion systems, since the $n=0$ Landau level plays an important role for their appearance. Therefore, conversely, the observation of these magnetotransport phenomena gives strong evidences of realization of massless Dirac fermions in organic conductors, $\alpha$-(BEDT-TTF)$_2$I$_3$ and its related compounds.

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