Pressure effects on Dirac fermions in $\alpha$-(BEDT-TTF)$_2$I$_3$

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Abstract

We investigate the pressure effect on the layered Dirac fermion system, which is realized in quasi-two-dimensional organic compound $\alpha$-(BEDT-TTF)$_2$I$_3$. The trajectory of the contact points is investigated using the tight-binding model with the transfer integrals determined by x-ray diffraction experiments. Vanishing of the Dirac fermion spectrum, opening of the gap, and pressure dependence of interlayer magnetoresistance are discussed.

1. Introduction

Layered organic conductors, BEDT-TTF [bis(ethylendithiolo)tetrathiofulvalene] salts, exhibit various electronic states due to electronic correlation, for example, superconductivity, Mott insulation, and charge ordering, with variation of pressure or temperature [1, 2].

Recently, Katayama et al have theoretically suggested that the 3/4-filled $\alpha$-(BEDT-TTF)$_2$I$_3$ salt is a zero-gap system under uniaxial pressure along the BEDT-TTF molecule stack axis (a-axis) [3, 4]. In the zero-gap state, the Fermi surface is reduced to two points, and the valence and conduction bands contact at two points ($\pm k_0$), which are not located in highly symmetric points in the two-dimensional (2D) Brillouin zone. Those points shift their positions toward the center of the Brillouin zone with increasing pressure [5–8]. In the vicinity of each contact point, two bands show linear dispersion with a tilted cone-like shape. In consequence of the cone-like dispersion, the low-lying excitation properties of the conduction electrons are described by a tilted massless Dirac equation [5]. First principles calculations support this Dirac cone structure [9, 10].

The massless Dirac electron system has also been found in graphene which is a single layer of graphite [11–13]. However, in contrast to graphene, $\alpha$-(BEDT-TTF)$_2$I$_3$ is not a 2D system but a multi-layered bulk material. In $\alpha$-(BEDT-TTF)$_2$I$_3$ crystal, conducting layers of BEDT-TTF molecules and insulating layers of I$_3$ anions stack alternately along the c-axis. Since the conductive layers are separated by the insulating layers, interlayer transfer energy is sufficiently small and thus this system has a strong 2D nature.

It has theoretically been explained by Osada [14] that the experimentally observed negative interlayer magnetoresistance [15] is due to the zero-mode Landau level of the massless Dirac fermion. In addition, we have proposed that the presence of a tilted and anisotropic Dirac cone can be verified using the interlayer magnetoresistance [16]. The interlayer magnetoresistance in $\alpha$-(BEDT-TTF)$_2$I$_3$ depends on the in-plane magnetic field direction because of tilt.

The hopping parameters of the $\alpha$-(BEDT-TTF)$_2$I$_3$ system are controlled by pressure. Two Dirac cone locations in the Brillouin zone also move by pressure, therefore the distance between valleys would be changed. This feature is very intriguing, because the inter-valley scattering effect would be affected by the change of Dirac cone locations.

In this paper, we investigate the pressure effect on the trajectory of the two Dirac points by changing transfer energies within the tight-binding model. We also calculate the pressure effect on the interlayer magnetoresistance. We use the parameters of the Weyl equation estimated by the tight-binding model [5, 4] with the transfer integrals determined by x-ray diffraction experiments [17], and discuss the Dirac fermion merging and the gap opening in the high pressure region.

The organization of this paper is as follows. In section 2, we calculate the pressure dependence of the Dirac cone parameters in $\alpha$-(BEDT-TTF)$_2$I$_3$ by the tight-binding model. In section 3, we discuss the merging behavior of Dirac points.
In section 4, we show the exact solution of the Landau level on the tilted Weyl equation. In section 5, we calculate the pressure dependence of the interlayer magnetoresistance by using the parameters estimated from the tight-binding model. Section 6 gives the conclusions of this work.

2. Pressure dependence

In figure 1, the basic model describing the electronic state in α-(BEDT-TTF)2I3 is shown [3, 18, 4, 5, 19]. The unit cell consists of four BEDT-TTF molecules named A, A', B, and C according to charge disproportionation. To consider the Coulomb interaction between molecules, we use the extended Hubbard model which is given by

\[ H = \sum_{(\alpha,\beta),\sigma} t_{\alpha\beta} n_{\alpha\sigma} n_{\beta\sigma} + \sum_{\alpha} U_{\alpha} n_{\alpha\uparrow} n_{\alpha\downarrow} + \sum_{\beta} V_{\beta} n_{\beta\uparrow} n_{\beta\downarrow} , \]

where \( i \) and \( j \) denote indices of the unit cell, and \( \alpha \) and \( \beta \) are indices of BEDT-TTF molecules in the unit cell. In the first term, \( a_{\alpha\sigma}^\dagger a_{\gamma\sigma} \) denotes the creation (annihilation) operator for the electron of spin \( \sigma \) \( (= \uparrow, \downarrow) \) at the \( i \)th site, and \( t_{\alpha\beta} \) is the transfer energy between the \((i, \alpha)\) and \((j, \beta)\) sites. The second and the last terms denote repulsive Coulomb interactions where \( U \) is the on-site interaction, and \( V_{\beta} \) the anisotropic nearest-neighbor interaction. Following [5, 4], we introduce the effect of the uniaxial pressure along the \( a \)-axis connecting the nearest-neighbor sites of the unit cell. The Hamiltonian (3) is diagonalized as

\[ \epsilon_{\alpha\beta}(k) = \xi_{\alpha\sigma}(k) \delta_{\alpha\sigma} + \sum_{\beta} \epsilon_{\beta\sigma}(k) \delta_{\beta\sigma} , \]

where \( \epsilon_{\alpha\beta}(k) \) is the corresponding eigenvector.

The averaged number of electrons \( n_{\alpha\sigma} \) is written as

\[ n_{\alpha\sigma} = \sum_{k} \sum_{\beta} \exp(\xi_{\alpha\beta}(k) - \mu)/k_B T + 1 , \]

where \( T \) is a temperature and \( k_B \) denotes the Boltzmann constant. The chemical potential \( \mu \) is determined by the condition \( \frac{1}{2} \sum n_{\alpha\sigma} \) because of 3/4-filling. The parameters \( U = 0.4 \) eV, \( V_c = 0.17 \) eV, and \( V_p = 0.05 \) eV are chosen [18, 5].

In this paper, we consider the zero-gap state. The conduction band \((r = 1)\) and the valence band \((r = 2)\) are degenerate at the two points \( k_0 \) and \( -k_0 \), and in the vicinity of the contact point \( k_0 \), the Hamiltonian is written as [5]

\[ H_z(k) = \tau \left( \sigma_0 + \tau \gamma \lambda \sigma_x + \gamma \lambda \sigma_y \right) , \]

where \( k = k_0, \sigma_x \), and \( \sigma_y \) are Pauli matrices and \( \sigma_0 \) is the identity matrix. \( \tau = \pm \) denotes the valley index which corresponds with \( k_0 \) and \( -k_0 \) respectively. These contact points occur in pairs and can be described by independent degrees of freedom, which leads to a twofold valley degeneracy. The valley degree of freedom is called the ‘valley spin’. We note that the \( k_+\) and \( k_-\) axes are not along the crystalline \( a^*\) - and \( b^*\) -axes, respectively (the superscript * means the reciprocal), because the system is rotated in order to remove complexity of the Hamiltonian. We define the angle made by \( k_z \) and \( b^* \) as \( \phi_0 \).
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For convenience, we define the Dirac cone parameters as

\[
\tilde{v}_0 e^{i\phi} = \frac{v_0^x}{v_x} + \frac{v_0^y}{v_y}, \quad \alpha = \sqrt{\frac{v_x}{v_y}}, \quad \gamma = \sqrt{1 - \frac{v_0^x}{v_0^y}}, \tag{7}
\]

where \( \tilde{v}_0 \) and \( \phi \) represent the tilting magnitude and the direction of the Dirac cone, respectively. The parameter \( \alpha \) represents the strength of anisotropy coming from the non-tilting effect. The parameter \( \gamma \) measures the strength of tilt of the Dirac cone which satisfies the relation \( 0 < \gamma \leq 1 \) (\( \gamma = 1 \) for the non-tilting case).

Figure 2 shows the pressure dependence of the Dirac cone parameters under the uniaxial pressure \( P_x \). Figure 2(a) shows the pressure dependence of the anisotropy \( \alpha \) coming from the non-tilting effect. At \( P_x \approx 8 \) kbar, the system is almost isotropic (\( \alpha \approx 1 \)) because the hopping parameter \( t_{22} \) takes almost the same value as \( t_{01} \) and \( t_{02} \). At \( P_x > 10 \) kbar, \( \alpha \) increases with pressure. This growth results in the increase of the interlayer magnetoresistance peak with respect to azimuthal angle dependence. In the region 4.5 kbar \( \leq P_x < 8 \) kbar, \( \alpha \) decreases with increasing pressure. Figure 2(b) shows the pressure dependence of the amplitude of Dirac cone tilting. At \( P_x \approx 5 \) kbar, the tilt of the Dirac cone is maximum.

In the region 5 kbar \( < P_x < 35 \) kbar, \( \gamma \) increases with increasing pressure, thus, the tilt of the Dirac cone decreases. At \( P_x > 35 \) kbar, the tilt of the Dirac cone increases again with pressure. Figure 2(c) shows the angle made by \( k_x \) and crystalline \( b^* \)-axis. In the high pressure region, \( P_x \geq 20 \) kbar, this angle becomes almost constant and the \( k_x \) axis is parallel to the crystalline \( b^* \)-axis. We recall that the pressure \( P_x \) is uniaxial. In the high pressure region, the transfer integrals \( t_{01} \) and \( t_{02} \) are enhanced by the uniaxial pressure, so the energy contour becomes elliptic and shrinks along the \( a^* \)-axis.

Figure 2(d) shows the azimuthal angle of the tilting direction. In the high pressure region, \( P_x > 30 \) kbar, the tilt of the Dirac cone is almost along the \( k_y \)-axis.

3. Merging Dirac points

As pressure increases, the two contact points approach each other, and then they merge into the single point. After merging, the contact points vanish and the gap opens between the electron and hole bands. Montambaux et al have proposed the universal \( 2 \times 2 \) Hamiltonian to describe the motion and merging behavior of Dirac points, and they have obtained a semiclassical description of the Landau levels’ spectrum [6–8]. This model describes continuously the Landau level coupling between valleys associated with two Dirac points in the vicinity of the merging Dirac points.

Here we calculate the trajectory and the gap opening behavior of the Dirac points by the four-band tight-binding model described in section 2. Figure 3 shows the pressure dependence of the distance of the Dirac points for several values of \( U, V_c, \) and \( V_p \) scaled by an interaction parameter \( x \). The contact points exist over a wide pressure range. However, in some pressure region, they are not located at the Fermi level. Under low pressure regions, \( P_x < 3 \) kbar, the contact points exist but they are not located at the Fermi level because of the existence of the hole and electron pockets, which are denoted by dashed lines in figure 3. In the high pressure
region, $P_a > 68$ kbar, the contact points in the case of the interaction parameters $x = 0.8$ and 1.0, are not located at the Fermi level as denoted by the dashed line in figure 3. In the case of the interaction parameter $0 \leq x \leq 0.6$, the contact points are located just on the Fermi level in the vicinity of the critical pressure for the merging of the Dirac points.

After merging, the contact points vanish and the gap opens between the electron and hole bands. Figure 4 shows the pressure dependence of the gap between the two subbands. The bandgap depends on the pressure linearly. The critical pressure for the merging of the Dirac points increases with increasing the parameter $x$. This critical pressure increase is not general behavior for other interaction parameters. In this case, the interactions $U$, $V_c$, and $V_p$ are taken so that the charge disproportionation pattern becomes the stripe pattern which is consistent with the experiment. We do not understand the mechanism of this upward shift. But this upward shift suggests that the Dirac fermions are stabilized by increasing the interaction parameters. Probably this is associated with the enhancement of charge disproportionation. We would like to investigate this point in a future publication.

4. Exact solution of the Landau level on the tilted Weyl equation

As shown in the section 2, Dirac fermions in the $\alpha$-(BEDT-TTF)$_2$I$_3$ system are described by the tilted Weyl equation. Reflecting the tilt of the Dirac cone, the Landau level wavefunctions are anisotropic. In this section we derive the exact Landau level wavefunctions of those Dirac fermions in a magnetic field.

First, we rescale the system to remove the anisotropy coming from non-tilting effects: $v_i \pi_i \rightarrow v_it_i$, $v_i \pi_i \rightarrow v_it_i$ where $v = \sqrt{v_x v_y}$ and $\pi_i = -i\hbar d_{i\tau} + eA_t$ ($i = x, y$). Second, we rotate the system by the angle $\phi_t$ in the plane so that the tilting direction of the Dirac cone is along the rotated $k_t$ axis:

$$\begin{pmatrix} \pi_x \\ \pi_y \end{pmatrix} = \begin{pmatrix} \cos \phi_t & -\sin \phi_t \\ \sin \phi_t & \cos \phi_t \end{pmatrix} \begin{pmatrix} \pi'_x \\ \pi'_y \end{pmatrix}. \quad (8)$$

After these transformations, the tilted Weyl Hamiltonian is written as

$$H_t(p) = vU_t^i(x_i)[\tau \bar{v}_0 \pi'_x \sigma_0 + \tau \pi'_x \sigma_x + \pi'_y \sigma_y]U_t(\phi_t), \quad (9)$$

$$U_t(\phi_t) = \cos(\phi_t/2) \sigma_0 + i\tau \sin(\phi_t/2) \sigma_c. \quad (10)$$

We multiply both sides of the Schrödinger equation $H\psi = E\psi$ by the operator $U^i_t(x_i)[\tau \pi'_x \sigma_x + \pi'_y \sigma_y]U_t(\phi_t)$ from the left, and then after some algebra we obtain

$$\begin{aligned}
\nu^2 \left[ \left( 1 - v^2 \right) \left( \pi'_x + \frac{\nu}{v} \frac{E_0}{1 - v^2} \pi'_y \right) \right] \psi \\
= \begin{bmatrix} E^2 & - \nu^2 \frac{h^2}{tE} \pi'_x (\sigma_z + i\nu \sigma_y) U_x(\phi_t) \\
\nu^2 \frac{h^2}{tE} \pi'_x (\sigma_z + i\nu \sigma_y) U_x(\phi_t) & \pi'_y \end{bmatrix} \psi. \quad (11)
\end{aligned}$$

We redefine the momentum operator as

$$\pi_x = \sqrt{v} \left( \pi'_x + \frac{\nu}{v} \frac{E_0}{1 - v^2} \pi'_y \right), \quad \pi_y = \frac{1}{\sqrt{v}} \pi'_y. \quad (12)$$

Both $\pi_x$ and $\pi_y$ satisfy the commutation relation $[\pi_x, \pi_y] = -i\hbar \nu E$, where $l_c = \sqrt{\hbar/eB}$ is the magnetic length. We rewrite equation (11) by the redefined momentum operator

$$\begin{aligned}
\left[ \pi_x^2 + \pi_y^2 \right] \psi = \frac{1}{\gamma} \frac{\hbar^2}{l_c^2} \begin{pmatrix} \pi_x^2 & -\tau \nu \bar{v}_0 e^{i\nu \phi_t} \\
-\tau \nu \bar{v}_0 e^{-i\nu \phi_t} & \pi_y^2 \end{pmatrix} \psi, \quad (13)
\end{aligned}$$

where $E = \frac{B}{\gamma} v\nu$. We define the ladder operator

$$\tilde{a} = l_c \left( \pi_x - i\nu \pi_y \right), \quad (14)$$

which satisfies the commutation relation $[\tilde{a}, \tilde{a}^\dagger] = 1$. In addition, we define the number operator $\tilde{N} = \tilde{a}^\dagger \tilde{a} = \frac{\pi_x^2 + \pi_y^2}{2\hbar^2} - \frac{1}{2}$. We take the eigenstate of the number operator, $\tilde{N}\phi_n = n\phi_n$. The eigenstate of Hamiltonian (6) is denoted by $\phi_n$.

Now we comment on the difference between tilted and non-tilted Dirac cones. If the Dirac cone is not tilting, the right-hand side of equation (13) becomes diagonal. In the tilted case, the off-diagonal part does not vanish, hence the wavefunctions are linear combinations of $\phi_n$ and $\phi_{n+1}$. We write the wavefunction as $\psi = (u, v)^T \phi_n$, and substitute this into equation (13), then we get the relation

$$\begin{pmatrix} \pi_x^2 - \gamma^2 \tau - \gamma^3 (2n + 1) & -\nu \bar{v}_0 e^{i\nu \phi_t} \pi_x \\
\nu \bar{v}_0 e^{-i\nu \phi_t} & \pi_y^2 \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} = 0. \quad (15)$$

For this equation to have a solution, the determinant of the left-hand side matrix must be equal to zero, so the eigenenergy $\epsilon$ becomes

$$\epsilon = \pm \sqrt{\gamma^3 (2n + 1 \pm 1)}. \quad (16)$$
When $\varepsilon = \pm \sqrt{2}\gamma^3n$, the wavefunctions are given by
\[
\psi_n^T = \frac{1}{\sqrt{2(1 + \gamma^2)}} \begin{pmatrix} -\tau \nu_0 e^{-i\tau \phi} \\ \tau + \gamma \end{pmatrix} \phi_n. \quad (17)
\]
When $\varepsilon = \pm \sqrt{2}\gamma^3(n + 1)$, the wavefunctions are given by
\[
\psi_n^T = \frac{1}{\sqrt{2(1 + \gamma^2)}} \begin{pmatrix} \tau + \gamma \\ -\tau \nu_0 e^{i\tau \phi} \end{pmatrix} \phi_n. \quad (18)
\]
Then, the wavefunction $\psi_n$ which has the eigenenergy $\varepsilon = \text{sgn}(n)\sqrt{2}\gamma^3|n|$ reads
\[
\psi_n = \frac{A_n}{\sqrt{2(1 + \gamma^2)}} \begin{pmatrix} -\tau \nu_0 e^{-i\tau \phi} \\ \tau + \gamma \end{pmatrix} \phi_{|n|} + \frac{B_n}{\sqrt{2(1 + \gamma^2)}} \begin{pmatrix} \tau + \gamma \\ -\tau \nu_0 e^{i\tau \phi} \end{pmatrix} \phi_{|n| - 1}. \quad (19)
\]
The coefficients $A_n$ and $B_n$ satisfy the relation $A_n e^{-i\phi} = \text{sgn}(n)B_n$, which is determined from the Schrödinger equation $H\psi_n = E_n\psi_n$.

Finally, the energy and eigenstate are written as
\[
E_n = \text{sgn}(n)\sqrt{2h|v_x|e|Bz\gamma^3|n|}, \quad (20)
\]
\[
\psi_n^T = \frac{1}{2\sqrt{1 + \gamma^2}} \begin{pmatrix} -\tau \nu_0 e^{-i\tau \phi} \\ (\tau + \gamma) e^{i\tau \phi} \end{pmatrix} \phi_{|n|} + \begin{pmatrix} \tau + \gamma \\ -\tau \nu_0 e^{i\tau \phi} \end{pmatrix} \phi_{|n| - 1} \quad (n \neq 0), \quad (21)
\]
and
\[
\psi_0^T = \frac{1}{\sqrt{2(1 + \gamma^2)}} \begin{pmatrix} -\tau \nu_0 e^{-i\tau \phi} \\ \tau + \gamma \end{pmatrix} \phi_0. \quad (22)
\]
The explicit form of the Landau level wavefunctions depends on the choice of the gauge. In order to see anisotropy of the wavefunction, it is convenient to take a symmetric gauge. On the other hand, for the calculation of the interlayer magnetoresistance, it is convenient to take the Landau gauge. Below we show both cases separately.

4.1. Symmetric gauge case

As we shall see later, the interlayer magnetoresistance in $\alpha$-(BEDT-TTF)$_2$I$_3$ depends on the in-plane magnetic field direction because of anisotropy in the Landau level wavefunction. In order to get a clear picture, we solve the tilted Weyl equation with the symmetric gauge $\mathbf{A} = \frac{1}{2} \mathbf{B} \times \mathbf{r}$.

The presence of the in-plane magnetic field is taken into account by a gauge transformation
\[
\psi = \exp \left[ -i \left( \frac{x}{L_x} - \frac{y}{L_y} \right) \frac{\hbar}{2} \right] \psi', \quad (23)
\]
with the magnetic length $L_x = \sqrt{\hbar/eB_x}$ ($\mu = x, y$). After this transformation, the vector potential is given by $\pi_x = -i\hbar\partial_x + \frac{1}{2}eB_y$ and $\pi_y = -i\hbar\partial_y - \frac{1}{2}eB_x$. We rescale and rotate the system as
\[
\begin{align*}
\nu_x^{-1}x &\to vx, \\
\nu_y^{-1}y &\to vy,
\end{align*}
\]
\[
\begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} \cos \phi & -\sin \phi \\ \sin \phi & \cos \phi \end{pmatrix} \begin{pmatrix} \nu_x^{-1}x \\ \nu_y^{-1}y \end{pmatrix}, \quad (24)
\]
respectively. The transformation of equation (12) is equivalent to $\tilde{x} = \frac{1}{\sqrt{\nu}}x$ and $\tilde{y} = \nu^\frac{1}{2}(y + \frac{\nu_x}{\nu_y^2}x)$. The center coordinate of the cyclotron motion $\tilde{X}, \tilde{Y}$ reads
\[
\tilde{X} = \tilde{x} - \frac{J^2}{\hbar} \tilde{\pi}_y, \quad \tilde{Y} = \tilde{y} + \frac{J^2}{\hbar} \tilde{\pi}_x. \quad (25)
\]
This satisfies the non-vanishing commutation relation as $[\tilde{X}, \tilde{Y}] = i\tilde{L}_z$, and wavefunctions cannot be simultaneously eigenfunctions of both of them. We choose to use the operator $\tilde{X}^2 + \tilde{Y}^2$ which also commutes with the Hamiltonian. In the symmetric gauge case, this operator is given by
\[
\tilde{X}^2 + \tilde{Y}^2 = 2\frac{L_z^2}{\hbar^2}(\tilde{N} - \tilde{L}_z + \frac{1}{2}) \quad (26)
\]
with the number operator $\tilde{N} = \tilde{a}^\dagger \tilde{a}$ and the angular momentum $\tilde{L}_z = \frac{1}{2}(\tilde{x}\tilde{p}_x - \tilde{y}\tilde{p}_y) = \hbar(\tilde{a}^\dagger \tilde{a} - \tilde{b}^\dagger \tilde{b})$. Here, we define the ladder operator $\tilde{b}$ as
\[
\tilde{b} = \frac{1}{\sqrt{2\ell_z}} (\tilde{X} + i\tilde{Y}). \quad (27)
\]
$\tilde{a}$ and $\tilde{b}$ satisfy commutation relations: $[\tilde{a}, \tilde{b}^\dagger] = 1$, $[\tilde{a}, \tilde{b}] = [\tilde{a}, \tilde{b}^\dagger] = 0$. We can rewrite these expressions by introducing complex coordinates $\tilde{\chi} = \frac{\tilde{x} + i\tilde{y}}{\ell_z}$, $\tilde{\chi}' = \frac{\tilde{x}' + i\tilde{y}'}{\ell_z}$.
\[
\tilde{a} = \frac{1}{\sqrt{2}} \left( -i\tilde{\chi}' + i\tilde{\chi} \right), \quad \tilde{b} = \frac{1}{\sqrt{2}} \left( \tilde{\chi}' + \frac{1}{2} \tilde{\chi} \right). \quad (28)
\]
Using these operators, we find that the eigenstates are denoted by a ket vector $n, m \ (n \geq 0, m \leq -n)$, where $\tilde{a}^\dagger \tilde{a} n = mn$ and $\tilde{b}^\dagger \tilde{b} m = (m - n)n, m$. The eigenvalue of $\tilde{L}_z$ is $nm$.

The wavefunction for zero-mode eigenfunction 0, 0 is obtained by solving $\tilde{a}0, 0 = \tilde{b}0, 0 = 0$. In the coordinate representation $\phi_{0,0}(r) = r(0, 0)$,
\[
\phi_{0,0}(r) = \frac{1}{2\pi \ell_z} e^{-i\frac{r^2}{4}} = \frac{1}{2\pi \ell_z} \exp \left(-\frac{x^2 + y^2}{4\ell_z^2} \right). \quad (29)
\]
Higher Landau level wavefunctions are derived as $n, m = \frac{(\nu_x)^{\ell_z} (\nu_y)^{\ell_z-m}}{\sqrt{n! m!}}0, 0$. Thus, the coordinate representation of wavefunctions is given by
\[
\phi_{n,m}(r) = N_{n,m} \text{exp} \left(-\frac{|\tilde{x}|^2}{4} \right) \tilde{x}^{|m|} \tilde{L}_z^{|m|} \left( \frac{|\tilde{x}|^2}{2} \right), \quad (30)
\]
where $N_{n,m} = \frac{(-1)^m}{\sqrt{2\pi \ell_z}} \frac{n!}{\sqrt{2\pi \ell_z}^m (n-m)!}$ is a normalization constant and $\tilde{L}_z^{|m|}$ is the Laguerre polynomial $L_{|m|}^{|m|}(\tilde{r}) = \frac{1}{\pi} e^{-|r|} |d^{|m|} (e^{-|r|^2} r^{|m|})$. 

(continued)
4.2. Landau gauge case

Lowest Landau level wavefunctions are isotropic in real space (not shown). We show \( |\phi_{0,0}(\vec{r})|^2 \) and \( |\phi_{0,m}(\vec{r})|^2 \) in figure 5. In the presence of Dirac cone tilting, the energy contour of the cone becomes elliptic. From the uncertainty principle, we expect that the wavefunction shrinks in the tilt direction. In fact, for a tilted Dirac cone, the zero energy Landau level wavefunction is anisotropic and shrinks in the tilt direction as shown in figure 5. For a non-tilted Dirac cone, the zero energy Landau level wavefunction is isotropic in real space (not shown).

\[
\psi = \exp \left[ -i\frac{\hbar}{2} \left( \begin{array}{c} \frac{\partial}{\partial \alpha} - \frac{\partial}{\partial \beta} \\ \frac{\partial}{\partial \beta} + \frac{\partial}{\partial \alpha} \end{array} \right) \right] \psi'.
\]  

Then we obtain
\[
\pi_x = -i\hbar \partial_x - eA^{(c)}_x, \quad \pi_y = -i\hbar \partial_y - eA^{(c)}_y. \tag{33}
\]

We rescale and rotate the system as equation (24). After transformation, we take \( A^{(c)}_x = -B_y \gamma' \) and \( A^{(c)}_y = 0 \). The transformation of equation (12) is equivalent to \( \tilde{x} = \frac{1}{\sqrt{\gamma'}} x - eB_y \alpha, \quad \tilde{y} = \sqrt{\gamma'} (y - \frac{eB_x}{\hbar} \gamma'). \) We choose the operator \( \hat{Y} \) to also commute with the Hamiltonian. In this gauge, the operator \( \hat{Y} = \frac{\hbar}{2} \hat{\gamma}/\hbar \) the momentum in the \( \hat{\alpha} \) direction is conserved in this gauge. Thus, the wavefunction is given by
\[
\phi(\tilde{x}, \tilde{y}) = \frac{1}{\sqrt{L_x}} \phi(\tilde{y}) e^{i\tilde{x} \tilde{y}} = \frac{1}{\sqrt{\gamma' L_x}} \phi(\tilde{y}) e^{i\tilde{x} \tilde{y}}, \tag{34}
\]

where \( L_x \) is the length of the system in the \( x \) direction. The ladder operator \( \hat{a} \) is given by
\[
\hat{a} = -\frac{1}{\sqrt{2L_y}} (\tilde{\eta} + \tilde{\eta} \hat{\gamma} \gamma), \tag{35}
\]

where
\[
\tilde{\eta} = \sqrt{\gamma'} (y' - \tilde{r} \tilde{\gamma}) - \sqrt{2} \gamma \tilde{\gamma} \tilde{\gamma} \gamma. \tag{36}
\]

The wavefunction for zero-mode eigenfunction \( \phi_0(y) \) is obtained by solving \( \hat{a} \phi_0(y) = 0 \). The eigenfunction is given by
\[
\phi_n(\tilde{\eta}) = \frac{(-1)^n}{\pi^{n + \frac{1}{2}}} \sqrt{\frac{1}{\gamma' L_x}} \exp \left[ -\frac{\hbar^2}{2L_y} \gamma \right] H_n \left( \frac{\tilde{\eta}}{L_x} \right), \tag{37}
\]

where the Hermite polynomials are given by \( H_n(x) = (-1)^n e^{-x^2} \frac{d^n}{dx^n} e^{-x^2} \).

5. Interlayer magnetoresistance

Now we compute the interlayer magnetoresistance and discuss pressure effects on it. We represent the magnetic field as \( \mathbf{B} = (B_x, B_y, B_z) = B(\cos \theta \cos \phi, \cos \theta \sin \phi, \sin \theta) \).

The interlayer tunneling between the \( j \)th plane and the \((j + 1)\)th plane is described by
\[
H_c = -t_c \sum_{j_c, \sigma = \uparrow, \downarrow} \int d^2 r \tilde{\psi}_{j_c, \sigma}(r) \tilde{\psi}_{j_c + 1, \sigma}(r) + \text{h.c.}, \tag{38}
\]

where \( t_c \) is interlayer transfer energy. In the Landau gauge, the momentum in the \( \tilde{\gamma} \) direction is conserved because the central coordinate \( \tilde{X} \) is conserved, hence the operator \( \hat{\psi} \) is written as
\[
\hat{\psi}_{j_c}(r) = \sum_{n, X} \psi_{n, X}(x, y, j_c) \hat{c}_{j_c}. \tag{39}
\]

Using equation (32), the Landau wavefunction is given by
\[
\psi_{n, X}(x, y, j_c) = \frac{1}{\sqrt{L_x}} \exp \left[ i \frac{\hbar}{\gamma'} K \right] \psi_{n, X}(y, z) e^{i\tilde{x} \tilde{y}}, \tag{40}
\]

where \( K \) represents a phase factor defined by
\[
K(x, y, \theta, \phi, \varphi) = [x (\sin \phi \cos \varphi - \alpha^2 \cos \phi \sin \varphi) - y (\alpha^2 \sin \phi \sin \varphi + \cos \phi \cos \varphi)] \frac{e}{\hbar} B \cos \theta. \tag{41}
\]
The current operator is written as

\[ J_z = \frac{1}{L_z} \int \int \sum_{n,k,k'} \left[ \psi_{+ n,k}^{*} \psi_{+ n,k'}^{*} + \psi_{- n,k} \psi_{- n,k'} \right] e^{i k_\parallel \cdot \rho} K(x,y,\theta,\phi) \times \psi_{+ n,k}^{\dagger} \psi_{+ n,k'}^{\dagger} \times \tilde{c}_{n,k,\perp} \tilde{c}_{n,k',\perp} + \text{h.c.} \],

(42)

where the x-integration gives \( k' = k + \delta k \). Hence, the center of mass \( \tilde{Y} \) is written as \( \tilde{Y} = \tilde{Y} + l_\parallel^2 \alpha \sqrt{T} \delta k \), where

\[ \delta k = \frac{eB}{\hbar} \cos \theta \sin \phi \cos \phi - \alpha^{-2} \sin \phi \cos \phi \text{d}c. \]

(43)

The current operator \( J_z \) becomes

\[ J_z = \frac{1}{L_z} \sum_{n,k,k'} \left[ \int \text{d}y \exp \left\{ -i \delta_c \frac{eB}{\hbar} \cos \theta \times (\alpha^2 \sin \phi \sin \phi + \cos \phi \cos \phi \text{d}y) \right\} \times \phi_{+ n,k,+\delta k} \phi_{+ n,k',+\delta k} \times \tilde{c}_{n,k,\perp} \tilde{c}_{n,k',\perp} + \text{h.c.} \right]. \]

(44)

The matrix element \( n, k, j_1, j_2 | n', j_1', j_2' \) is written as

\[ n, Y, j_1, j_2 | n', Y', j_1', j_2' = \text{det} \left( \delta_{j_1+1,j_1'} - \delta_{j_1,j_1'} \right) \delta_{Y,Y'} + \delta_{Y,Y} \delta_{j_1,j_1'} \]

\[ \times \int \text{d}y \exp \left\{ -i \frac{eB}{\hbar} \cos \theta \left( \alpha^2 \sin \phi \sin \phi + \cos \phi \cos \phi \right) \text{d}y \right\} \times \phi_{n,j_1} \left( \frac{\alpha \sqrt{y}}{l_z} \right) \phi_{n,j_1'} \left( \frac{\alpha \sqrt{y}}{l_z} \right) \left( y - \delta B k \right) \delta_{j_1,j_1'} \text{d}y. \]

(45)

From the Kubo formula, the interlayer magnetoresistance \( \sigma_{zz} \) is given by

\[ \sigma_{zz}(\omega) = \frac{i}{\hbar} \sum_{n,k,j} \left[ \frac{f(E_n') - f(E_n)}{E_n' - E_n} \right] \times \left[ \frac{\alpha^2 \sin \phi \sin \phi + \cos \phi \cos \phi}{2\pi} \right], \]

(46)

where the summations with respect to the layer index \( j \) and the wave number \( k \) yield the number of layer \( N_{\text{layer}} \) and the Landau level degeneracy \( \frac{1}{2\pi l_z^2} \), respectively. The interlayer magnetoresistance \( \rho_{zz} \) takes the form \[ \sigma_{zz}(\omega) \]

\[ \frac{\rho_{zz}(B = 0)}{\rho_{zz}(B \neq 0)} = \frac{B_0}{B_0 + B \sin \theta \text{ exp} \left\{ -\frac{1}{2} \left( \frac{\alpha^2 \sin \phi \sin \phi}{2\pi} \right) \right\}} \]

(47)

with

\[ I(\phi, \alpha, \phi_i, \gamma) = \gamma \left( \alpha \sin \phi \cos \phi_i \right)^2 \]

\[ + \frac{1}{\gamma} \left( \frac{\sin \phi \sin \phi_i + \frac{1}{\alpha} \cos \phi \cos \phi_i}{\alpha \cos \phi \cos \phi_i} \right)^2, \]

(48)

where \( B_0 \) is the resistance in the absence of a magnetic field. This formula is derived by using the zero-mode Landau level wavefunction. To justify this approximation, the magnetic field \( B_z \) should be large enough or the temperature low enough to satisfy the relation \( E_1 > k_B T \).

The anisotropy of the Landau level wavefunction shown in figure 5 leads to anisotropy in the interlayer magnetoresistance. Figure 6 shows the physical picture of the dependence of interlayer magnetoresistance on the in-plane magnetic field direction. The in-plane magnetic fields, \( B_x \) and \( B_y \), are treated by the gauge transformation \( (32) \), which gives rise to the phase factor when the electron hops between one layer to the adjacent layer. Figure 6 shows the case where the in-plane magnetic field is parallel to the \( x \)-axis. In this case, the phase factor is written as \( \exp \left( i \frac{eB}{\hbar} \right) \).

The wavefunction oscillates in real space along the direction perpendicular to the in-plane magnetic field because of the phase factor. As a consequence, the matrix element \( (45) \) is reduced when the in-plane magnetic field is perpendicular to the direction in which the wavefunction is extended. Reflecting the real space anisotropy in the wavefunction, the matrix element depends on the direction of the in-plane magnetic field, \( \phi \). The interlayer magnetoresistance, thus, depends on \( \phi \).

Figure 7 shows the in-plane magnetic field direction dependent interlayer magnetoresistance for different pressures. When the pressure increases, the minimum of the magnetoresistance moves to 90° and the peak of interlayer magnetoresistance increases. In the high pressure region, the parameter \( \alpha \) increases as shown in figure 2(a), so the effect from the anisotropy coming from the non-tilting effect becomes dominant. This growth results in the increase of the interlayer magnetoresistance peak. In this case, the energy contour shrinks along the \( a^* \)-axis by the uniaxial pressure, so the interlayer magnetoresistance is a minimum when the in-plane magnetic field is parallel to the \( a^* \)-axis, i.e. 90°.

6. Summary

In the present study, we examined pressure effects on Dirac fermions in \( \alpha-(BEDT-TTF)_{2}I \) within the tight-binding model. The electron and valence bands are degenerate at two contact points \( k_0 \) and \(-k_0 \) in the Brillouin zone. They are
located at the Fermi level over a wide pressure range. The pressure dependence of the distance between contact points in the Brillouin zone also depends on the interaction parameters. In the vicinity of the merging, the ‘valley spin’ picture would breakdown because the coupling between two valleys, which is usually neglected in graphene, becomes strong. This merging behavior may be observed in the pressure range $40 \text{kbar} \leq P_a < 70 \text{kbar}$, and the interaction parameter $x \leq 0.6$. Around that pressure, we expect a rapid increase of the interlayer resistivity coming from the opening of an energy gap. This suggests that this system is useful for investigating the valley splitting effect that is still controversial in graphene.

We show the exact solution of the Landau level on the tilted Weyl equation by using the symmetric and Landau gauges. Because of the tilt, the Landau level wavefunctions become anisotropic and shrink in the tilt direction in real space. We calculate the pressure dependence of the interlayer magnetoresistance by using the parameter estimated from the tight-binding model. In the high pressure region $P_a > 10 \text{kbar}$, anisotropy increases with pressure. This increase results in the increase of the interlayer magnetoresistance peak.

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Figure 7. The dependence of azimuthal angle magnetic field direction on the interlayer magnetoresistance for various values of uniaxial pressure.