Effect of anion potential on the zero-gap state in the
two-dimensional organic conductor $\alpha$-(BEDT-TTF)$_2$I$_3$

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Abstract. The organic conductor $\alpha$-(BEDT-TTF)$_2$I$_3$ exhibits the zero-gap state (ZGS) described by the massless Dirac fermions, and is in contrast to that of the single layer graphite, i.e. graphene since the former is robust against the site potential compared with the latter. However, the ZGS of the organic conductor is not fully clarified due to the complexity of several kinds of transfer energies arising from four kinds of donor molecules in a unit cell. In the present paper, we analyze the details of the ZGS by focusing on the role of the anion potential, which acts differently on respective site of donor molecules. We show that the anion potential with a small magnitude does not destroy the ZGS but has an effect of varying the location of the contact point between the conduction and valence bands as found in the case of pressure. Such a behavior can be understood by the tilted Weyl equation with a perturbation for the potential. Further, the ZGS is discussed by examining the relation between the anion potential and the charge disproportionation.

1. Introduction

Two-dimensional organic conductor, BEDT-TTF salt, and its derivative have been investigated extensively owing to the variety of electronic states [1]. Especially, $\alpha$-(BEDT-TTF)$_2$I$_3$, which consists of the unit cell containing four BEDT-TTF molecules (A, A’, B, and C site) and has the 3/4-filled band [2] is a peculiar salt. It has been shown that the insulating state occurs as a result of the charge ordering at low temperatures, and the superconducting state emerges under the uniaxial strain [3, 4, 5]. Further pressures result in the zero-gap state (ZGS), which exists around a contact point [6] between the conduction band and the valence band. These two bands degenerate at two momenta of the contact point, around which the ZGS displays massless fermions with a linear dispersion. Such a state can explain the experiment showing the strong temperature dependence of the Hall coefficient under the high pressure [4, 7].

The ZGS of $\alpha$-(BEDT-TTF)$_2$I$_3$ [6] exhibits following characteristic features compared with the graphene (single layer graphite) in which the ZGS has been also found [8, 9]. The location of the contact point in the momentum space moves with varying the parameters of transfer energies [6]. The linear dispersion is anisotropic and is described by the tilted Weyl equation [10], while the location stays at the symmetry point and the linear dispersion is isotropic for the graphene. There is the charge disproportionation in the real space even at high temperatures for the $\alpha$-(BEDT-TTF)$_2$I$_3$ salt. The pattern is given by $\langle n_C \rangle > \langle n_A \rangle = \langle n_A' \rangle > \langle n_B \rangle$, where $\langle n_\alpha \rangle$ denotes the quantity of the charge [11, 12, 13]. In addition to such a difference in the magnitude, the local magnetic susceptibilities also exhibit a difference given by $\chi_C > \chi_A = \chi_A' > \chi_B$ [14]. It is
shown theoretically that the difference in the local magnetic susceptibilities originates from the local density of states for respective sites [15]. It should be noted that the system has an inversion symmetry in the ZGS and the amount of the charge and magnetic susceptibility at A site are equal to those at A’ site. The role for the A and A’ sites is similar to that of the carbon atoms in graphene while that of B and C sites exhibits a peculiar property for \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) salt as seen from the behavior of the eigenvector of the Hamiltonian.

The \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) salt contains four donor molecules connected by several kinds of transfer energies. Since it is not straightforward to understand the ZGS due to the complicated structure in the unit cell, we have discussed it using a reduced model instead of the full model [6]. Recently, Kondo et al. [16] examined the effect of the local potential corresponding to the one from the anion by using transfer energies based on their X-ray experiment. In the present paper, we investigate the electronic state of the ZGS in the \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) by varying the magnitude of the anion potential, while the transfer energies obtained by the first principle calculation by Kino and Miyazaki [17] are utilized. The anion, I\(_{-1}\), which has a negative charge, gives rise to the repulsive potential for the electrons at BEDT-TTF molecules. We examine the effect of the anion on the ZGS by varying magnitudes of two kinds of anion potentials. The result of the calculation is analyzed by the perturbation in terms of the anion potential. Further, the relation between the anion potential and charge disproportionation is discussed.

2. Band and Model

2.1. Energy Band

The unit cell of the \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) consists of four donor molecules, BEDT-TTF, and two anions, I\(_{-1}\) as shown in Fig. 1 [18]. The bonds \(c1, c2, c3, p1, p2, p3, \) and \(p4\) correspond to \(a2, a3, a1, b3, b4, b1, \) and \(b2\) in Ref. [18], and \(c1’, c3’, c4’\) corresponds to \(a1’, a3’, \) and \(a4’\) in Ref. [17], respectively. Electrons move among BEDT-TTF molecules (A, A’, B, and C sites) connected with several kinds of transfer energies on the \(ab\) plane. The locations of anions existing between two conducting layers can be projected on the C site and the midpoint between

\[x(b)\]

\[y(a)\]

**Figure 1.** Conducting plane of the \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) with the unit cell (gray square). Indices \((c1, \cdots, c3, p1, \cdots, p4)\), and \((c1’, c3’, c4’)\) denote nearest neighbor bonds and next nearest neighbor ones, respectively, which give the transfer energies, \(t_{c1}, \cdots, t_{c4’}\). The red diamonds represent the anion, I\(_{-1}\), located above and below the \(ab\) plane.
A and A’ sites in the ab plane. Such a system is described by the extended Hubbard model:

\[ H = \sum_{\langle i,j \rangle, \sigma} t_{i\alpha,j\beta} c_{i\alpha}^{\dagger} c_{j\beta} + \sum_{i,\alpha} \epsilon_i c_{i\alpha}^{\dagger} c_{i\alpha} + \sum_{\langle i,j \rangle, \sigma, \sigma'} v_{i\alpha,j\beta} c_{i\alpha}^{\dagger} c_{j\beta} c_{i\sigma}^{\dagger} c_{j\sigma}, \]

where \( i \) and \( j \) are indices of the unit cell and \( \alpha, \beta = (1, 2, 3, 4) \) are sites in the unit cell, i.e., A, A’, B, and C sites. The operator \( c_{i\alpha}^{\dagger} \) annihilates the electron at \( (i, \alpha) \) site and with \( \sigma = (\uparrow, \downarrow) \) spin. The transfer energy between \( (j, \beta) \) and \( (i, \alpha) \) site is given by \( t_{i\alpha,j\beta} \) with \( t_{c1}, t_{c2}, t_{c3}, t_{p1}, t_{p2}, t_{p3}, t_{p4}, t_{c1}', t_{c\beta}', \) and \( t_{c4}' \). The coefficients of the remained terms are the magnitudes for the anion potential, \( I_{\alpha} \), the on-site Coulomb interaction \( U \), and the off-site one \( V_{i\alpha,j\beta} \), where \( V_{i\alpha,j\beta} = V_c \) for c1, c2, and c3 bonds and \( V_{i\alpha,j\beta} = V_p \) for p1, p2, p3, and p4 bonds. The lattice constant is taken as unity. Applying the mean-field approximation and Fourier transformation to eq. (1), the \( 4 \times 4 \) matrix is obtained as follows:

\[ t_{\alpha\beta}(k) = \left( \begin{array}{cccc}
2t_{c1}\cos ky + C_1 & tc_{1} + t_{c2}e^{-iku} & tp_{1} + t_{p4}e^{iku} & tp_{2} + t_{p3}e^{iku} \\
tc_{1} + t_{c2}e^{iku} & 2t_{c1}\cos ky + C_2 & tp_{1}e^{iku} + tp_{2}e^{iku} & tp_{3} + tp_{2}e^{iku} \\
tp_{1} + t_{p4}e^{-iku} & tp_{1}e^{-iku} + tp_{2}e^{-iku} & tc_{3} + tc_{3}e^{-iku} & tc_{3} + tc_{3}e^{-iku} \\
tp_{2} + t_{p3}e^{-iku} & tp_{2}e^{-iku} & tc_{3} + tc_{3}e^{-iku} & 2t_{c1}\cos ky + C_4
\end{array} \right), \]

where \( C_1 = \frac{1}{2} U \langle n_1 \rangle + 2V_c \langle n_2 \rangle + 2V_p \langle n_3 \rangle + 3V_p \langle n_4 \rangle + I_1, \)

\( C_2 = \frac{1}{2} U \langle n_2 \rangle + 2V_c \langle n_1 \rangle + 2V_p \langle n_3 \rangle + 3V_p \langle n_4 \rangle + I_2, \)

\( C_3 = \frac{1}{2} U \langle n_3 \rangle + 2V_c \langle n_4 \rangle + 2V_p \langle n_1 \rangle + 3V_p \langle n_2 \rangle + I_3, \) and

\( C_4 = \frac{1}{2} U \langle n_4 \rangle + 2V_c \langle n_3 \rangle + 2V_p \langle n_1 \rangle + 3V_p \langle n_2 \rangle + I_4. \)

The quantity \( \langle n_{\alpha} \rangle \) denotes the amount of the charge at \( \alpha \)-th site. Considering the 3/4-filled band, the chemical potential, \( \mu \), and \( \langle n_{\alpha} \rangle \) can be calculated self-consistently by the equations shown below:

\[ \sum_{\beta} (t_{\alpha\beta}(k) - \mu \delta_{\alpha\beta})d_{\beta\gamma}(k) = (\xi_{\gamma}(k) - \mu)\delta_{\alpha\gamma}(k), \]

\[ \langle n_{\alpha} \rangle = \frac{1}{N} \sum_{k\gamma} |d_{\alpha\gamma}(k)|^2 \frac{1}{\exp[(\xi_{\gamma}(k) - \mu)/T] + 1}. \]

\[ \sum_{\alpha} \langle n_{\alpha} \rangle = 6, \]

where \( \xi_{\gamma}(k) (\xi_1(k) > \xi_2(k) > \xi_3(k) > \xi_4(k)) \) is the eigenvalue of eq. (2), and \( d_{\alpha\gamma}(k) \) represents the \( \alpha \)-th component of the eigenvector for \( \xi_{\gamma}(k) \). The computational and theoretical work for the band (valence) band. We note that, in the ZGS, there is no magnetic moment and \( \langle n_{\alpha} \rangle \) must satisfy the condition, \( \langle n_1 \rangle = \langle n_2 \rangle \), due to the inversion symmetry. The temperature, \( T \), in eq. (3) is set to \( T = 0 \), and thus the Fermi distribution function \( \exp(\xi_{\gamma}(k) - \mu)/T + 1 \) is replaced by the step function, \( \theta(\xi_{\gamma}(k) - \mu) \) with \( \varepsilon_F \) being the Fermi energy. After the calculation of \( \varepsilon_F \) and \( \langle n_{\alpha} \rangle \), the eigenvalue, \( \xi_{\gamma}(k) \) and eigenvector, \( d_{\alpha\gamma}(k) \), can be obtained by diagonalizing the matrix, eq. (2). Hereafter, the unit of energy is taken as eV.

For the numerical calculation, we utilize the data of \( t_{i\alpha,j\beta} \) estimated from the first principle calculation at 8 K under the ambient pressure [17], where the data are given as \( t_{c1} = -0.0158, t_{c2} = 0.0029, t_{c3} = -0.0173, t_{p1} = 0.1154, t_{p2} = 0.1075, t_{p3} = 0.0085, t_{p4} = 0.0559, t_{c1'} = 0.0085, t_{c3'} = 0.0072, \) and \( t_{c4'} = -0.0012. \) The on-site Coulomb interaction is set to \( U = 0.4, \) and the off-site ones are set to \( V_p = 0.05 \) and \( V_c = 0.17. \) Note that such a choice of parameters of \( t_{i\alpha,j\beta}, U, \) and \( V_{i\alpha,j\beta} \) leads to the ZGS. We can take generally the anion potential at A and A’ as \( I_1(= I_A) = 0, I_2(= I_A') = 0, \) which corresponds to a choice of the origin of the energy. Thus, the electronic states are studied by considering \( I_3(= I_B) \) and \( I_4(= I_C) \) as parameters.
2.2. Tilted Weyl equation

The ZGS of \( \alpha \)-(BEDT-TTF)\(_2\)I\(_3\) in the vicinity of the contact point can be described by the tilted Weyl equation. The equation with an effective Hamiltonian can be written in the following form:

\[
H_{\text{eff}}(k) x = E(k) x \\
H_{\text{eff}}(k) = \sum_{\rho = 0}^{3} k \cdot v_\rho = k \cdot \begin{pmatrix}
    v_0 + v_3 & v_1 - iv_2 \\
    v_1 + iv_2 & v_0 - v_3
\end{pmatrix}
\]

(4)

where \( k \) denotes the momentum measured from the contact point \((k_0)\) and \( x \) represents the two-component eigenvector. The velocity, \( v_\rho \), is defined as

\[
v_0 = \frac{1}{2}(\tilde{v}_{11} + \tilde{v}_{22}) \\
v_1 = \text{Re}(\tilde{v}_{12}) \\
v_2 = \text{Im}(\tilde{v}_{12}) \\
v_3 = \frac{1}{2}(\tilde{v}_{11} - \tilde{v}_{22})
\]

\[
\tilde{v}_{\gamma\gamma'} = \sum_{\alpha\beta} d_{\alpha\gamma}(k_0') \frac{\partial \tilde{t}_{\alpha\beta}}{\partial k} d_{\beta\gamma'}(k_0'),
\]

(5)

where eigenvector, \( d_{\beta\gamma}(k'_0) \), is calculated at \( k'_0 = k_0 + \delta k \) due to the degeneracy. The vector \( \delta k \), which is an infinitesimally small vector, is chosen to have only the \( x \)-component as taken previously [15]. In such a case, by defining \( u_\gamma \) as the four-component vector corresponding to \( d_{\alpha\gamma} \) with \( \gamma = 1, \ldots, 4 \), the bases are obtained as

\[
\begin{align*}
    u_1 &= \begin{pmatrix} 0.5194 \\ -0.4429 + 0.2713i \\ 0.0058 + 0.0071i \\ 0.6623 + 0.1473i \end{pmatrix}, \\
    u_2 &= \begin{pmatrix} 0.4419 \\ -0.3703 - 0.2411i \\ 0.6125 - 0.4841i \\ -0.0002 + 0.0010i \end{pmatrix}, \\
    u_3 &= \begin{pmatrix} 0.5154 \\ -0.1944 - 0.4773i \\ -0.0944 + 0.4392i \\ -0.2410 - 0.4571i \end{pmatrix}, \\
    u_4 &= \begin{pmatrix} 0.5190 \\ 0.3211 + 0.4077i \\ -0.4332 - 0.0311i \\ -0.4233 + 0.3057i \end{pmatrix}.
\end{align*}
\]

(6)

The velocities are approximately given as

\[
v_0 = (v_{0x}, v_{0y}), v_1 = (0, 0), v_2 = (0, v'), v_3 = (v, 0),
\]

(7)

where \( v_{0x} = -0.0389, v_{0y} = -0.0048, v' = 0.0439, v = 0.0515 \).

The eigenvalue of eq. (4) is obtained as

\[
E(k) = k \cdot v_0 \pm \sqrt{\sum_{\rho = 1}^{3} [k \cdot v_\rho]^2} \simeq k \cdot v_0 \pm \sqrt{(v k_x)^2 + (v' k_y)^2}.
\]

(8)

Note that \( d_{\beta\gamma}(k'_0) \), \( v_1 \), and \( v_2 \) depends on \( k'_0 \) but \( E(k) \) is independent of the choice of \( k'_0 \). By defining \( \theta \) as the angle between \( k_0 \)-axis and \( \delta k \), it has been shown [15] that \( \theta \)-dependence of \( d_{11}(\theta) (= d_{21}(\theta)) \) is weak while \( d_{31}(\theta) \) and \( d_{41}(\theta) \) have a node at \( \theta \simeq 0 \) and \( \theta \simeq \pi \), respectively. On the other hand, the \( \theta \)-dependence of \( d_{\alpha2}(\theta) \) is given by \( d_{\alpha2}(\theta) = d_{\alpha1}(\theta + \pi) \).
Figure 2. Phase diagram on the plane of $I_B$ and $I_C$. The shaded region in the ZGS phase satisfies a condition, $\langle n_4 \rangle > \langle n_1 \rangle = \langle n_2 \rangle > \langle n_3 \rangle$, which corresponds to the result observed in the experiment [13].

Figure 3. Trajectory of the contact point, where the parameters are chosen as $-0.280 < I_C < 0.125$, $I_B = 0$ (closed circle), $-0.260 < I_B < 0.075$, $I_C = 0$ (open circle), and $-0.2 < I_B = I_C < 0.157$ (gray circle). For the case of $I_B = I_C = 0$, the contact point is located at the open square ($k_0 = (0.686\pi, -0.443\pi)$). The points $\Gamma$, $X$, $Y$, and $M$ represents $(0,0)$, $(\pi,0)$, $(0,-\pi)$, and $(\pi,-\pi)$, respectively.

3. Role of Anion Potential

Figure 2 shows the phase diagram of the ZGS and the insulating state as the function of $I_B$ and $I_C$ for $U = 0.4$, $V_p = 0.05$, $V_c = 0.17$. The ZGS is retained for small $I_B$ and $I_C$. The insulating state appears for large $I_B$ and/or $I_C$, although the inversion symmetry is kept even in the insulating phase.

We note that the dispersion including the anion potential has been studied by Kondo et al., recently [16] who reexamined the X-ray experiment for the $\alpha$-(BEDT-TTF)$_2$I$_3$ salt at the room temperature. Their analysis shows Fermi surfaces in the absence of the anion potential, where the contact point exists below the Fermi energy. The Fermi surface diminishes to the points for the large attractive potential at B and C sites, and the insulating phase emerges for the large repulsive potential at B or C site. On the other hand, the ZGS is always obtained in the present calculation when the contact point exists. Such a distinction around the Fermi energy comes mainly from the choice of the transfer energy, i.e., the difference between Ref. [16] and Ref. [17], although the global feature of Fig. 2 is qualitatively similar to that obtained by Kondo et al.

Figure 3 denotes the trajectory of the contact point given by the change of the anion potentials where the open square represents the location of the contact point $k_0 = (0.686\pi, -0.443\pi)$ for $I_B = 0$ and $I_C = 0$. For the range of $-0.260 < I_B < 0.075$, $I_C = 0$ ($-0.280 < I_C < 0.125$, $I_B = 0$), the point moves from $Y$($M$) point to $M$($Y$) point through the open square. Compared with Fig. 2, it is seen that the ZGS with increasing $I_B$ disappears at $Y$ on the boundary line connecting $I_B = -0.260$, $I_C = 0$ and $I_B = 0.0$, $I_C = 0.125$, and that the ZGS with increasing $I_C$ disappears at $M$ on the boundary line connecting $I_B = 0.075$, $I_C = 0$ and $I_B = 0.0$, $I_C = -0.280$. Two contact points $k_0$ and $-k_0$ merge at the inversion center ($\Gamma$, $X$, $Y$, and $M$) and the band insulator is obtained at the phase boundary as shown in Ref. [6].
trajectory for $-0.280 < I_C < 0.125$, $I_B = 0$ is the same as that for $-0.260 < I_B < 0.075$, $I_C = 0$. On the other hand, the trajectory for $I_B = I_C$ (gray circles) deviates instead of going to Y point, since the ZGS is stable for $I_B = I_C < 0$.

The trajectory for weak $I_B$ and $I_C$ can be understood as follows by using the tilted Weyl equation. The anion potential represented by $4 \times 4$ matrix is given by

$$\hat{I} = \left( \begin{array}{cccc} I_A & 0 & 0 & 0 \\ 0 & I_A & 0 & 0 \\ 0 & 0 & I_B & 0 \\ 0 & 0 & 0 & I_C \end{array} \right).$$

(9)

Such a potential for the reduced model with $2 \times 2$ matrix representation is written as $\langle \gamma | \hat{I} | \gamma' \rangle$ ($\gamma, \gamma' = 1, 2$), where $| \gamma \rangle$ is given by $u_\gamma$ in eq. (6). Thus, the total Hamiltonian is expressed as $H_{eff} + A_0 \sigma_0 + A_3 \sigma_3$. Using the velocities of eq. (7), the eigenvalue of $H_{eff} + \langle \gamma | \hat{I} | \gamma' \rangle$ is calculated as

$$E = v_0 \cdot k + A_0 \pm \sqrt{(v'k_y)^2 + (vk_x + A_3)^2},$$

(10)

where

$$A_0 = 0.4651I_A + 0.2302I_C + 0.3047I_B,$$

$$A_3 = 0.0745I_A + 0.2302I_C - 0.3047I_B.$$

(11)

Thus, the ZGS is retained by shifting the contact point to satisfy $k_x = -A_3/v$. It is emphasized, within the first order of perturbation, that any anion potential modifies the coefficient of $\sigma_3$ in eq. (4). The contact point moves only $k_x$-direction due to $v_{3y} \approx 0$. Actually, the difference between the contact point for $I_B = I_C = 0$ and that for $I_B = I_C = 0.001$ is given by $0.0015\pi$, while the value estimated from eq. (10) is given by $-A_3/v = 0.0009\pi$. For anion potential increasing from small value, the trajectory in Fig. 3 could be explained by using the perturbation of the higher order.

![Figure 4](image)

Figure 4. The charge, $\langle n_a \rangle$, at the four kinds of BEDT-TTF molecules, A, A’, B, and C. as the function of $I_B$ with $I_C = 0$ (a), and $I_C$ with $I_B = 0$ (b), where the ZGS exists in $I_B < 0.075$ (a) and $I_C < 0.125$ (b), respectively.

Here we discuss the anion potential and charge disproportionation. Figure 4 (a) and (b) shows $\langle n_a \rangle$ as functions of $I_B$ and $I_C$, respectively. The value of $\langle n_B \rangle$ ($\langle n_C \rangle$) decreases when $I_B$ ($I_C$) increases due to the positive potential energy ($I_\alpha > 0$) for electrons. The relation, $\langle n_C \rangle > \langle n_A \rangle = \langle n_A' \rangle > \langle n_B \rangle$, which is observed in the experiment [13], is satisfied in the shaded region of Fig. 2. We expect $I_B < 0$ and $I_C > 0$ in the actual system, since the site C (B) is close to (far away from) an anion, and the anion potential at A and A’ sites are taken the origin, i.e. $I_A = I_{A'} = 0$. The parameter, which satisfies the experimental result of the charge disproportionation, is limited to a narrow region. Although the ZGS phase is large, the effect of the anion potential must be small in order to explain the experiment [13].
4. Conclusion

We studied the electronic states of the two-dimensional organic conductor $\alpha$-(BEDT-TTF)$_2$I$_3$ salt by considering the effect of the anion potential. Our result of the phase diagram on the plane of $I_C$ and $I_B$ obtained from the first principle calculation [17] resembles that of Kondo et al [16]. However, the present calculation shows the robust ZGS in the presence of the contact point, while the formation of the Fermi surface is also possible in Ref. [16]. For the strong potential, the charge disproportionation deviates from the result of the experiment and the ZGS disappears with the inversion symmetry being kept. The small potential is needed to explain the experimental result of the charge disproportionation.

The trajectory of the contact point, which is obtained in Fig. 3, can be understood analytically for the case of the weak potential. Actually, the anion potential has an effect of modifying the coefficient of the $\sigma_3$ in the tilted Weyl equation, and then the ZGS is retained by determining the location of the contact point so as to keep the zero value of the coefficient. However, it remains as an open problem to calculate the higher order with respect to the anion potential for understanding the whole trajectory of the contact point in Fig. 3.

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