Dynamical time-reversal and inversion symmetry breaking, dimensional crossover, and chiral anomaly in $\alpha$-(BEDT-TTF)$_2$I$_3$

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In most Dirac semimetals, time-reversal and inversion symmetries are believed to play a crucial role in their stability. We demonstrate that these symmetries are broken in Dirac fermions in the organic conductor $\alpha$-(BEDT-TTF)$_2$I$_3$ due to the strong electronic correlation. The system is a three-dimensional type-II Dirac semimetal in the coherent inter-layer tunneling regime. A chiral anomaly is predicted to be observed in the magnetoresistance when the magnetic field is tuned to the inter-layer tunneling direction. Our result suggests that $\alpha$-(BEDT-TTF)$_2$I$_3$ is a useful platform to explore interplay between the chiral anomaly and the strong correlation and/or dimensionality.

Recently, the topological Dirac and Weyl semimetals have attracted intense theoretical and experimental interest because of their intriguing topological and electronic properties [1, 2]. In nodal semimetals, the conduction and valence bands touch only at certain points in the Brillouin zone (BZ), and the low-energy excitations are described by a relativistic Dirac or Weyl equation, where the velocity of light is replaced by the Fermi velocity $v_F$ [3]. Two-dimensional (2D) Dirac semimetals are realized in graphene [4], as is clearly demonstrated by the Dirac nature of the electronic transport and the surface state of the three-dimensional (3D) topological insulators [5, 6], where spin-orbit coupling plays a significant role. Based on theoretical and experimental efforts, 3D Dirac semimetals are now realized experimentally, for instance, in Na$_3$Bi [7, 8] and Cd$_3$As$_2$ [9, 10]. Remarkably, the topological semimetals have deep connections with particle physics because they provide solid state analogues of relativistic chiral fermions [2, 11] and lattice realizations [12] of the chiral anomaly of the quantum field theory [13, 14]. Experimental evidence has been accumulated about the existence of Fermi arc surface states [15–18] and the novel responses of the chiral anomaly to applied electronic and magnetic fields [19–23].

Now it is well accepted that symmetries and spin-orbit coupling are the keys to realizing 3D Dirac semimetals in general. Spin-orbit coupling can create a linear energy dispersion and a 3D Dirac semimetal appears when the symmetry conditions are met. Since the net Chern numbers at each contact point are zero, we need additional crystal symmetries [7, 9, 24–26]. In such 3D Dirac semimetal systems, Dirac points are on the symmetry lines in the BZ. Our current understanding of the condition of the Dirac semimetal is mostly based on symmetry consideration and spin-orbit coupling, though the latter is not necessary for some exceptional cases [27]. Another important feature of Dirac and Weyl semimetals is due to their lack of fundamental Lorentz symmetry. In general, the energy dispersion around the contact points is described by $\varepsilon_{\pm}(k) = T(k) \pm U(k)$, where $U(k)$ describes the anisotropic cone and the term $T(k)$, which is a linear function of $k$, describes the cone tilt. In the case of a type-II semimetal, the chiral anomaly only appears in the directions where $T(k) > U(k)$ [28] in contrast to a type-I semimetal where $T(k) < U(k)$ in all directions.

In this Letter, we demonstrate that the quasi-2D organic conductor, $\alpha$-(BEDT-TTF)$_2$I$_3$, is an unprecedented type of Dirac semimetal that does not fit into our current understanding of Dirac semimetal conditions. We show that contrary to other Dirac semimetals, both the time-reversal symmetry (TRS) and inversion symmetry are broken and spin-orbit coupling plays no role. In the symmetry broken state, the energies of the Dirac points are shifted asymmetrically from the Fermi energy and their positions are not symmetrically located with respect to the origin of the BZ. We also show that the system exhibits a dimensional crossover from a type-I 2D Dirac semimetal to a type-II 3D Dirac semimetal upon entering the coherent inter-layer tunneling regime.

The organic conductor $\alpha$-(BEDT-TTF)$_2$I$_3$ with the space group $P\overline{T}$ is a 2D Dirac semimetal with a layered structure comprising of Dirac fermion layers and insulating layers [29]. The Dirac points are at non-high-symmetry points in the BZ similar to certain inorganic materials with the $P\overline{T}$ space group [27, 30]. In $\alpha$-(BEDT-TTF)$_2$I$_3$, the unit cell is composed of four BEDT-TTF molecules, where BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene, A, A', B, and C, in the conduction layer [31, 32], as shown in Fig. 1(a). The system is metallic above 135 K but undergoes a metal-insulator transition [33–35] at 135 K where a charge order stripe pattern forms as confirmed by $^{13}$C-NMR (nuclear magnetic resonance) measurement [36]. Here, the short-range inter-site Coulomb repulsion plays a key role [37, 38]. The 2D Dirac semimetal appears when the charge order is suppressed under high-pressure as revealed by the tight-binding model calculation [39] and confirmed by the first-principle calculations [40, 41]. It should be stressed that the band filling is fixed to 3/4 [32] and the Fermi energy is exactly at the Dirac point within these calculations. The inter-layer magnetoresistance, which is negative and is in inversely proportional
FIG. 1. (a) Configuration of BEDT-TTF molecules in a conducting plane of α-(BEDT-TTF)$_2$I$_3$. The rectangle shows a unit cell that contains four molecules, A, A', B, and C. The molecules stacked in the $a$ axis, which is taken as the $y$ axis and the $b$ axis is taken as the $x$ axis. (b) Conduction and valence bands are plotted as functions of $k_x$ and $k_y$ at $P = 0.8$. There are two Dirac points at different energies, which are encircled by dotted circles, and they are located at the generic wave vectors. The horizontal plane denotes the Fermi energy. Magnified views of the Dirac nodes at $k_D^{(1)}$ and $k_D^{(2)}$ are shown in (c) and (d), respectively. The Dirac point at $k_D^{(1)}$ is below the Fermi energy, while the Dirac point at $k_D^{(2)}$ is above the Fermi energy.

to the applied magnetic field, clearly demonstrates the presence of the zero-energy Landau level [42–45]. Furthermore, the phase of the Dirac fermions is confirmed by the Shubnikov-de Haas oscillation of the hole-doped sample, where the sample is placed on polyethylene naphthalate substrate [46].

Thus far, the presence of the massless Dirac fermion spectrum has been established in α-(BEDT-TTF)$_2$I$_3$ but the role of the strong electronic correlation is unclear: The charge order stripe pattern at ambient pressure is replaced by charge disproportionation [47, 48], where $n_A = n_{A'}$ and $n_B \neq n_C$ with charge density at molecule $\alpha$ denoted by $n_\alpha$. The on-site Coulomb repulsion plays a central role in the determination of the charge densities but the inter-site Coulomb interaction plays a minor role. The purpose of this study is to demonstrate that the inter-site Coulomb interaction plays a crucial role in stabilizing a non-trivial Dirac semimetal state.

The Hamiltonian of electrons within a conduction layer is given by $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}}$. The first term describes the hopping between the molecules and in the momentum space,

$$\mathcal{H}_0 = \sum_{k,\sigma = \uparrow, \downarrow} c_{k\sigma}^\dagger H_0^k c_{k\sigma},$$

where $c_{k\sigma}^\dagger = (c_{k1\sigma}^\dagger, c_{k2\sigma}^\dagger, c_{k3\sigma}^\dagger, c_{k4\sigma}^\dagger)$ is the four-component creation operator for an electron with momentum $k$ and spin component $\sigma$. The indices 1, 2, 3, and 4 represent molecules A, A', B, and C, respectively. The matrix elements of $H_0^k$ are given by $H_0^k = t_{\alpha\beta} e^{-ik \cdot d_{\alpha\beta}}$, where the displacement vectors, $d_1$, $d_2$, and $d_3$, are defined by $d_1 = (0, a/2)$, $d_2 = (b/2, -a/4)$, and $d_3 = (b/2, a/4)$ [29]. Hereafter, we measure the energies in units of eV and we set $a = 1$ and $b = 1$ for the lattice constants. The transfer energies are pressure dependent and given by [48] $t_0 = C_\alpha (1 + b_\alpha P)$. Here, the pressure $P$ is in units of GPa. The numerical coefficients $C_\alpha$ are $C_{a1} = -0.028$, $C_{a2} = 0.048$, and $C_{a3} = -0.020$ for the stacking direction and $C_{b1} = 0.123$, $C_{b2} = 0.140$, $C_{b3} = -0.002$, and $C_{b4} = -0.025$ for the other directions. The numerical constants $b_\alpha$ are $b_{a1} = 0.89$, $b_{a2} = 1.67$, $b_{a3} = -0.25$, $b_{b1} = 0$, $b_{b2} = 0.11$, $b_{b3} = 0.32$, and $b_{b4} = 0$.

The interaction term $\mathcal{H}_{\text{int}}$ describes a strong electronic correlation that is given by

$$\mathcal{H}_{\text{int}} = U \sum_{j,\sigma} n_{j\alpha\uparrow} n_{j\alpha\downarrow} + \sum_{i,j,\alpha,\beta} V_{\alpha\beta} n_{i\alpha} n_{j\beta}.$$  

Here, the charge density of the electrons with spin $\sigma$ at molecule $\alpha$ in site $j$ is denoted by $n_{j\alpha}$, and we define $n_{j\sigma} = n_{j\alpha\uparrow} + n_{j\alpha\downarrow}$. The first term in the right-hand side describes the on-site Coulomb interaction while the second term describes the Coulomb interaction between nearest neighbor molecules. We set $V_{11,12} = V_{13,14} = V_c$ along the stacking direction of the BEDT-TTF molecules (the $a$-axis) and $V_{i\alpha,j\beta} = V_p$ otherwise. These interactions lead to an insulating state, which is a charge ordered state [37, 38], under ambient pressure and lead to dynamical TRS and inversion symmetry breaking in the Dirac semimetal phase as we illustrate below.

It has not yet been considered explicitly, except for in limited cases [49, 50], but the significant electronic correlation in the Dirac semimetal state, in the presence of the inter-molecule interaction $V_{\alpha\beta}$ is the bond correlation described by

$$\chi_{\alpha\sigma,\beta\sigma',\pm} = \frac{1}{N} \sum_k e^{-i k \cdot d_{\alpha\beta}^{(\pm)}} \langle c_{k\alpha\sigma}^\dagger c_{k\beta\sigma'} \rangle,$$

where $N$ is the number of BZ points and $d_{13}^{(\pm)} = d_2$, $d_{13}^{(\mp)} = d_3$, etc. Non-zero values of $\chi_{\alpha\sigma,\beta\sigma',\pm}$ can break
the TRS and inversion symmetry. We also include the site order defined as
\[ n_{\alpha\sigma} = \frac{1}{N} \sum_k \langle c^\dagger_{k\alpha\sigma} c_{k\alpha\sigma} \rangle, \quad \ldots \]
where \( n_{\alpha\sigma} \) is the site order, and \( n_{\alpha\sigma} \) is defined as
\[ n_{\alpha\sigma} = \frac{1}{N} \sum_k \langle c^\dagger_{k\alpha\sigma} c_{k\alpha\sigma} \rangle. \]

The resulting mean field Hamiltonian is denoted by
\[ H_{\text{mf}} = \sum_{k,\sigma,\sigma'} c^\dagger_{k\sigma} [H(k)]_{\sigma\sigma', \sigma} c_{k\sigma'}. \]

The matrix \( H(k) \) is \( 8 \times 8 \). We solve the self-consistent equations for \( x_{\alpha\beta\gamma, \beta\sigma, \tau} \) and \( n_{\alpha\sigma} \).

Now we describe symmetries of the system. To describe the symmetry operations, we define
\[ X_{\mu\nu\lambda} = \sigma_\mu \otimes \sigma_\nu \otimes \sigma_\lambda, \]
where \( \mu, \nu, \lambda = 0, 1, 2, 3 \). Here, \( \sigma_1, \sigma_2, \) and \( \sigma_3 \) denote the Pauli matrices and \( \sigma_0 \) is the \( 2 \times 2 \) unit matrix. In the definition of \( X_{\mu\nu\lambda} \), the first two Pauli matrices act on the four molecule indices and the last Pauli matrix acts on the spin index. The TRS is
\[ X_{002} H^*( -k ) X_{002} = H(k). \]

The system has an inversion center \([32, 51, 52]\) between A and A'. The symmetry operation associated with this inversion is
\[ iH( -k ) i = H(k), \]
where \( i = i ( x_{103} + x_{133} + x_{003} - x_{033} ) / 2 \). In the absence of the interactions, we see that the system is invariant under these symmetry operations. The situation does not change if we include the on-site Coulomb repulsion. If we restrict the self-consistent calculation to the Hartree level, these symmetries are unbroken but the exchange correlation associated with the inter-site Coulomb repulsion leads to breaking of both symmetries.

We present the energy dispersion of the conduction and valence bands in Fig. 1(b), (c), and (d). The conduction and valence bands are both spin degenerate. Contrary to the naive expectation, there is no mass gap in the Dirac fermion spectrum. We find that the two Dirac points are at \( k^{(1)}_D \) = (0.9250, -0.7978) where energy \( \varepsilon^{(1)}_D = 1.2788 \) and at \( k^{(2)}_D \) = (-0.8988, 0.7631) where energy \( \varepsilon^{(2)}_D = 1.2822 \). Here, the Fermi energy is \( \varepsilon_F = 1.2807 \) and we note that \( k^{(1)}_D \neq -k^{(2)}_D \) and \( \varepsilon^{(1)}_D < \varepsilon_F < \varepsilon^{(2)}_D \). By changing the pressure, the Dirac points move in the BZ and the electronic correlation changes as well [53]. Hereafter, we take \( U = 0.4, V_c = 0.17, V_p = 0.05, \) and \( P = 0.8 \). This set of interaction parameters reproduces the experimentally observed stripe pattern in the insulating state [48].

Although the renormalized hopping parameters break the inversion symmetry, the charges do not. In fact, we find that \( n_A = n_{A'} = 1.4554 \) where \( n_B = 1.2204 \) and \( n_C = 1.8696 \). This symmetry is protected by the strong correlation associated with \( U \), whereas \( V_c \) favors breaking this symmetry. The system undergoes a quantum phase transition as we increase the value of \( V_c \), merging the two Dirac points [54].

Since \( \alpha-(\text{BEDT-TTF})_2I_3 \) has a layered structure and high-mobility \([29] \sim 10^6\text{cm}^2\text{V}^{-1}\text{s}^{-1} \), the system undergoes a dimensional crossover from the 2D electronic state to the 3D electronic state. When the interlayer tunneling is incoherent, the electronic structure is 2D and when the interlayer tunneling becomes coherent \([55] \), the electronic structure is 3D. Due to the high-mobility value of \( \alpha-(\text{BEDT-TTF})_2I_3 \), the crossover temperature is in the order of the inter-layer tunneling amplitude. We describe the inter-layer tunneling between adjacent layers with the matrix \( t_1 A_{000} + t_2 A_{010} \), where the first term is the tunneling between the same molecules. The second term is the tunneling between A and \( A' \) and between B and C, where these pairs of molecules are aligned in the stacking direction. We note that these terms do not break the TRS or inversion symmetry, \( I \). Therefore, the contact points are stable against them. Because of the mirror reflection about the \( a-b \) plane, two copies of each Dirac point appear at \( k_z = \pm \pi/2 \). Since interlayer hopping parameters \( t_1 \) and \( t_2 \) are much smaller than the intralayer parameters, we include their effects based on the 2D result. We observe that the Dirac cone is tilted along the \( k_z \) direction by \( t_1 \) as shown in Fig. 2. For \( t_1/t_2 > \eta_1 \) (Fig. 2(c) and (d)), both Dirac cones are type-II \([28] \), which is to be applied to \( \alpha-(\text{BEDT-TTF})_2I_3 \), while for \( t_1/t_2 < \eta_2 \) (Fig. 2(a) and (b)), both Dirac cones are type-I. Here, \( \eta_1 = 0.6867 \) and \( \eta_2 = 0.6827 \). Interestingly, for \( \eta_2 < t_1/t_2 < \eta_1 \), the Dirac cones at \( k^{(1)}_D \) with \( k_z = \pm \pi/2 \) are type-I and the Dirac cones at \( k^{(2)}_D \) with \( k_z = \pm \pi/2 \) are type-II, where we can expect the partial chiral anomaly effect to be associated with the type-I Dirac cones.

Now we consider the chiral anomaly in this system. When a magnetic field \( B_z \) is applied along the \( z \)-direction, the spectrum of the Landau levels is given by
\[ \varepsilon_{n,k_z}^{\pm}\tau = -2t_1 \cos k_z \pm \sqrt{\varepsilon_{n\tau}^2 + 4n^2/2^2 \cos^2 k_z}, \]
where \( \varepsilon_{n\tau} \) is the 2D Landau level at the Dirac point \( k^{(\tau)}_D \) \((\tau = 1, 2) \) given by \([44, 45] \)
\[ \varepsilon_{n\tau} = \left(1 - \lambda^2_\tau \right)^{3/4} \frac{h^2}{\ell_z} \left(2|n|\right). \]

Here, \( n \) is an integer, \( \lambda_\tau \) is the tilt parameter of the Dirac cone, \( \nu^2_{2D} \) is the averaged Fermi velocity, and \( \ell_z = \sqrt{\hbar/|eB_z|} \) is the magnetic length, where \( \hbar \) is the reduced Planck constant and \( e \) is the electron charge. In \( \alpha-(\text{BEDT-TTF})_2I_3 \), \( \lambda_\tau \) and \( \nu^2_{2D} \) can be estimated exper-
mentally from the analysis of the interlayer magnetoresistance. It is found that \( (1 - \lambda_z^2)^{3/4} v_z^{2D} \approx 5 \times 10^{-4} \text{ m/s} \) \[56\], where \( \sqrt{1 - \lambda_z^2} \approx 0.05 \) \[57\].

Since the system is a 3D Dirac semimetal, the \( n = 0 \) Landau level has a chiral mode, \( \tau_{0,\pm \pi/2 + \delta k_z} = \pm h v_z^\tau \delta k_z \), such that
\[
v_z^\tau = \frac{2 a_z}{\hbar} (t_1 - \eta_2 t_2),
\]
where the lattice constant \( a_z \) is explicitly shown. The plus (minus) sign is for the Dirac cone at \( k_z = \pi/2 (\pm \pi/2) \). Because of the current flow between the two Dirac nodes, the negative magnetoresistance is observed when the magnetic field is tuned, within the angle \( \delta \theta \), to the direction of the inter-layer hopping, which is taken as the \( z \)-axis here for simplicity. We emphasize that the effect is limited to the magnetic field directions close to the inter-layer tunneling direction because of the type-II nature of the chiral anomaly \[28\]. We note that \( \delta \theta \) is approximately proportional to \( 1/t_1 \), and we find \( \delta \theta \approx 0.36^\circ \) when \( t_1 = 0.001 \).

The calculation above can be extended to include the effect of spin-orbit coupling \[58, 59\]. Due to the configuration of the molecules in the unit cell, there is no spin-orbit coupling along the molecule stacking direction. Although there is some ambiguity in the choice of the spin-orbit coupling parameters, a clear conclusion can be reached that the spin-orbit coupling does not create any mass gap at the Dirac points. Here, the exchange correlation effect also plays a crucial role \[60\].

In conclusion, we have demonstrated that \( \alpha-(\text{BEDT-TTF})_2\text{I}_3 \) is a new type of Dirac semimetal with remarkable features. Contrary to other Dirac semimetals, both the TRS and inversion are broken in a non-trivial way. This result clearly extends our current understanding of the symmetry condition for Dirac semimetals. In particular, our new Dirac semimetal can be used to deepen our understanding of chiral anomaly; we expect chiral anomaly to exist in 3D but not in 2D and the transition between them can be investigated in \( \alpha-(\text{BEDT-TTF})_2\text{I}_3 \) through the negative magnetoresistance in the direction of the inter-layer tunneling. One limitation is that the sample must be in a pressure cell, though the pressure is useful to control the electronic correlation of the system and investigate the interplay between chiral anomaly and other electronic states.

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Supplementary Material

SYMMETRIES OF THE HAMILTONIAN AT THE DIRAC POINTS

As discussed in the main text, the exchange correlation breaks both the TRS and the inversion symmetry, \( \mathcal{I} \). However, some components of the Hamiltonians at the Dirac points retain the inversion symmetry, \( \mathcal{I} \). To clarify this point, we represent the Hamiltonian at each Dirac point in terms of \( \lambda_{\mu \nu \lambda} \), where the coefficients are as shown in Table S1. In case of \( U = 0.4 \) and \( V_c = V_p = 0 \), the Dirac points are located at \( k = k_D, -k_D \) and both the TRS and the inversion symmetry, \( \mathcal{I} \), are unbroken. Under these symmetries, the Hamiltonian, in general, has the form

\[
\mathcal{H}(k) = \begin{pmatrix}
  a & x - iy & p & q \\
  x + iy & a & p^* & q^* \\
p^* & p & -a + b & c \\
q^* & q & c & -a - b
\end{pmatrix}
\]

(S1)

where \( a, b, c, x, \) and \( y \) are real functions of \( k \) and \( p \) and \( q \) are complex functions of \( k \). The terms \( y, \text{Im} \, p, \) and \( \text{Im} \, q \) are odd, while the others are even. Based on this form, we see that some of the coefficients are the same, such as \( \lambda_{020} \) and \( \lambda_{320} \), and some of the coefficients have opposite signs, such as \( \lambda_{200} \) and \( \lambda_{320} \).

When \( U = 0.4, V_c = 0.17, \) and \( V_p = 0.05 \), both the TRS and the inversion symmetry, \( \mathcal{I} \), are broken but the symmetries in the coefficients are preserved. The symmetries are broken by the functions multiplied to \( \lambda_{0\mu 0} \) that are neither even nor odd with respect to \( k \). Despite the presence of such symmetry breaking factors, part of the inversion symmetry, \( \mathcal{I} \), is still preserved. In fact, we find

\[
\text{tr} \left[ \lambda_{0\mu 0} \mathcal{H}(k_D^{(1,2)}) \right] = -\text{tr} \left[ \lambda_{330} \mathcal{H}(k_D^{(1,2)}) \right].
\]

(S2)

This is indispensable because \( \lambda_{0\mu 0} - \lambda_{330} \) and \( \lambda_{300} \) combined with the identity matrix lead to degenerate and other separated levels. We note that these generators are diagonal matrices and that diagonal components arise from the charge at each molecule. Therefore, they are associated with the inversion symmetry between \( n_A \) and \( n_{A'} \).

This inversion symmetry in the molecule charges is protected by the strong correlation. In Fig. S1, we show the interaction dependence of the mass gap in the Dirac points. The charge values \( n_A \) and \( n_{A'} \) are primarily determined by \( U \), while \( V_c \) favors \( n_A \neq n_{A'} \). By increasing \( V_c \), the system undergoes a quantum phase transition from the Dirac semimetal state to the massive Dirac fermion state. This quantum phase transition occurs due to the merging of the two Dirac points [1] at the time-reversal invariant momentum \((0, \pi)\).

![FIG. S1. Interaction dependence of the Dirac mass, \( m_D \), at the contact points. Here, we set \( U = 0.4, V_c = 0.17\zeta, \) and \( V_p = 0.05\zeta \). The mass gap opens for \( \zeta \geq 1.66 \).](image)

TABLE S1. Decomposition of the Hamiltonian at each Dirac point into \( \lambda_{\mu \nu \lambda} \). For the case of \( U = 0.4 \) and \( V_c = V_p = 0 \), both the TRS and inversion symmetry are unbroken. The Dirac points are at \( k = k_D, -k_D \), with \( k_D = (1.5516, -0.8044) \). Meanwhile, for the case of \( U = 0.4, V_c = 0.17, \) and \( V_p = 0.05 \), these symmetries are broken, and the Dirac points are at \( k = k_1^{(1)} k_2^{(1)} \), with \( k_1^{(1)} \neq k_2^{(1)} \), \( k_2^{(1)} = (0.9296, -0.8009) \), and \( k_2^{(2)} = (-0.9018, 0.7618) \). The breaking of the TRS and the inversion symmetry, \( \mathcal{I} \), is clearly illustrated through the comparison of these two cases.

| \( \lambda_{\mu \nu \lambda} \) | \( k_D \) | \( -k_D \) | \( k_D^{(1)} \) | \( k_D^{(2)} \) |
|-----------------|---------|---------|--------|--------|
| \( \lambda_{010} \) | 0.0000387 | 0.0000387 | 0.0097423 | -0.0092377 |
| \( \lambda_{020} \) | 0.0610626 | -0.0610626 | 0.0617559 | -0.0603957 |
| \( \lambda_{030} \) | -0.0102378 | -0.0102378 | 0.0227222 | 0.0227222 |
| \( \lambda_{100} \) | -0.0687389 | -0.0687389 | -0.0237383 | -0.0105359 |
| \( \lambda_{110} \) | -0.0687389 | -0.0687389 | -0.0237383 | -0.0105359 |
| \( \lambda_{120} \) | -0.141172 | 0.141172 | -0.1511936 | 0.1524187 |
| \( \lambda_{230} \) | -0.0650720 | -0.0650720 | -0.1106209 | -0.1070677 |
| \( \lambda_{200} \) | 0.0173256 | -0.0173256 | -0.0398026 | 0.048009 |
| \( \lambda_{101} \) | -0.0173256 | -0.0173256 | 0.0398026 | -0.048009 |
| \( \lambda_{122} \) | -0.0650720 | 0.0650720 | -0.1106209 | -0.1070677 |
| \( \lambda_{230} \) | -0.141172 | 0.141172 | -0.1511936 | 0.1524187 |
| \( \lambda_{000} \) | 0.0160446 | 0.0160446 | -0.0152960 | -0.0152960 |
| \( \lambda_{010} \) | 0.0290397 | 0.0290397 | 0.0265949 | 0.0379016 |
| \( \lambda_{020} \) | 0.0610626 | -0.0610626 | 0.0617559 | -0.0603957 |
| \( \lambda_{030} \) | 0.0102378 | 0.0102378 | -0.0227222 | -0.0227222 |
FIG. S2. Dirac point motion in 2D BZ. The two Dirac points are denoted by \( k_D^{(1)} \) and \( -k_D^{(2)} \). The pressure range is \( 0.50 \leq P \leq 2.0 \). The interaction parameters are \( U = 0.4, V_c = 0.17, \) and \( V_p = 0.05 \). As we increase the pressure, the two Dirac points move in the BZ. Here, we show \( k_D^{(1)} \) and \( -k_D^{(2)} \).

**DIRAC POINT MOTION IN THE BZ AND THE CHANGE OF THE ELECTRONIC CORRELATION**

As discussed in the main text, the Dirac points are not symmetrically located with respect to their origin in the 2D Dirac semimetal state. The positions of the Dirac points move by changing the pressure, as shown in Fig. S2. There is an accidental TRS point, where \( k_x = 1.06 \) and \( k_y = -0.501 \) at \( P = 1.25 \).

As we change the pressure, the electronic correlation changes as well. We can investigate the change of the electron correlation by the slave-rotor theory [2]. A phase degree of freedom \( \theta_j \), which is conjugate to the total charge at site \( j \), is introduced. In the simplest mean field approximation, where \( \langle e^{-i(\theta_j - \theta_j')} \rangle \approx \langle \cos \theta \rangle^2 = Z \), the hopping parameters \( t_\alpha \) are replaced by \( Z t_\alpha \). The physical meaning of \( Z \) is the quasiparticle weight. The stronger the electronic correlation is, the smaller the value of \( Z \).

In the Mott insulator, \( Z = 0 \), while \( Z \neq 0 \) in a metallic state. Assuming an effective on-site Coulomb interaction, \( U_{\text{eff}} = 0.32 \) for all molecules, we carry out the self-consistent calculation without the exchange correlation for simplicity. The result is shown in Fig. S3. The system exhibits the Mott-Dirac transition at \( P = P_c \) with \( P_c \approx 0.669 \). For \( P > P_c \), the system is a Dirac semimetal. In this regime, the electronic correlation decreases as we increase \( P \).

**SPIN-ORBIT COUPLING EFFECT ON DIRAC FERMION SPECTRUM**

In the presence of the spin-orbit coupling [3, 4], the \( \mathcal{H}_k \) in the main text is replaced by

\[
\begin{align*}
\mathcal{H}_{k}^{0} &\quad = \left( t_{a3} e^{-ik d_1} + t_{a2} e^{ik d_1} \right) \sigma_0, \quad (S3) \\
\mathcal{H}_{k}^{10} &\quad = t_{b3} \left( 1 + i \lambda_3 \cdot \sigma \right) e^{-ik d_3} \\
&\quad \quad + t_{b2} \left( 1 + i \lambda_2 \cdot \sigma \right) e^{ik d_2}, \quad (S4) \\
\mathcal{H}_{k}^{11} &\quad = t_{b4} \left( 1 + i \lambda_4 \cdot \sigma \right) e^{-ik d_2} \\
&\quad \quad + t_{b1} \left( 1 + i \lambda_1 \cdot \sigma \right) e^{ik d_3}, \quad (S5) \\
\mathcal{H}_{k}^{23} &\quad = t_{b2} \left( 1 + i \lambda_2 \cdot \sigma \right) e^{-ik d_2} \\
&\quad \quad + t_{b3} \left( 1 + i \lambda_3 \cdot \sigma \right) e^{ik d_3}, \quad (S6) \\
\mathcal{H}_{k}^{24} &\quad = t_{b4} \left( 1 + i \lambda_4 \cdot \sigma \right) e^{-ik d_2} \\
&\quad \quad + t_{b1} \left( 1 + i \lambda_1 \cdot \sigma \right) e^{ik d_3}, \quad (S7) \\
\mathcal{H}_{k}^{34} &\quad = \left( t_{a1} e^{ik d_1} + t_{a2} e^{-ik d_1} \right) \sigma_0. \quad (S8)
\end{align*}
\]

The vectors \( \lambda_j \) \((j = 1, 2, 3, 4)\) arise from the spin-orbit coupling [3]. For \( \alpha\)-(BEDT-TTF)\(_2\)I\(_3\), it is estimated that \( |\lambda_j| \sim 0.01 \) [3]. We note that the spin-orbit coupling effect between molecules along the \( y \)-axis is negligible; the orbitals of the molecules are almost parallel along this direction, and thus, the spin-orbit coupling effect is negligible [5]. Due to the spatial variation of the molecular wave functions, the vectors \( \lambda_j \) are almost perpendicular to the \( x\)-\( y \) plane. In the following, we assume \( \lambda_j = s_j(0, 0, \lambda) \) with \( s_j = \pm 1 \). From the diagonalization of the Hamiltonian, \( \mathcal{H}_k^0 \), we see that there is no gap opening for the case of \( s_1 = s_2 = s_3 = s_4 = 1 \). Meanwhile, a small mass gap, which is on the order of \( \lambda \), is created when we take, for instance, \( s_1 = s_2 = 1 \) and \( s_3 = s_4 = -1 \) [4].

Applying the mean field approximation to the interac-
The on-site term is given by
\[
H_{\text{on-site}} = \sum k, \alpha, \beta, \sigma, \sigma' \left( [H_{k}^{(\pm)}]_{\alpha, \beta, \sigma, \sigma'} e^{ik \cdot d_{\alpha, \beta}^{(\pm)}} + [H_{k}^{(-)}]_{\alpha, \beta, \sigma, \sigma'} e^{-ik \cdot d_{\alpha, \beta}^{(-)}} + [H_{k}^{(c)}]_{\alpha, \beta} \delta_{\alpha, \beta}^{(\sigma, \sigma')} c_{\kappa, \alpha, \sigma}^{\dagger} c_{\kappa, \beta, \sigma'} \right),
\] (S9)

The matrix components are given by
\[
[H_{k}^{(\pm)}]_{\alpha, \beta, \sigma, \sigma'} = \frac{1}{2} \left[ \delta_{\sigma, \sigma'} + i \lambda_{\alpha, \beta}^{(\pm)} \right] - V_{\alpha, \beta} \lambda_{\alpha, \beta, \sigma, \sigma'}^{*},
\] (S10)

where \( t_{13}^{(\pm)} = t_{13}^{(-)} = t_{12}, \lambda_{13}^{(+) = \lambda_{2}, \lambda_{13}^{(-)} = \lambda_{3}, \text{etc.} \)

The on-site term is given by
\[
[H_{k}^{(c)}]_{\alpha, \beta} = 2 \sum_{\gamma} V_{\alpha, \gamma} n_{\gamma} + U \left< n_{\alpha, \sigma} \right>,
\] (S11)

where \( \sigma \) is flipped to \( \sigma' \).

Based on the self-consistent calculation described in the main text with including the spin-orbit coupling terms, we find that there are significant differences in the resulting electronic structures depending on the choice of signs, \( s_{j} \). The choice of \( s_{1} = s_{2} = s_{3} = s_{4} = 1 \) is distinct from other choices and the corresponding energy dispersion of the conduction and valence bands is shown in Fig. S4. For this choice of signs, both the upper and lower bands are spin degenerate. Here, we set \( \lambda = 0.01 \) and \( U = 0.4, V_{c} = 0.17, \) and \( V_{p} = 0.05 \) for the interactions [6]. Contrary to the naive expectation, there is no mass gap in the Dirac fermion spectrum; however, there is a possibility of taking different sets of the spin-orbit coupling [4]. For the other choices of \( s_{j} \), we find that the spin-orbit coupling opens a huge gap in the spectrum. The gap is two orders of magnitude larger than that created by the spin-orbit coupling itself due to the enhancement of its effect by the exchange correlation. In this case, the mass gap is not scaled by the spin-orbit coupling interaction but by the interaction parameters, \( V_{c} \) and \( V_{p} \). This huge gap is inconsistent with the transport experiments [6-8]. The situation is the same for the other choices of spin-orbit coupling. Remarkably, there is no such enhancement for the case of \( s_{1} = s_{2} = s_{3} = s_{4} = 1 \). Of course, even in this case, the gap opens for extremely large values of \( \lambda \). We examined the \( \lambda \) dependence of the Dirac fermion spectrum and found that the mass gap opens for \( \lambda > 0.641 \). This critical value is much larger than that expected in \( \alpha-(BEDT-TTF)_{2}I_{3} \).

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