Observation of possible nonlinear anomalous Hall effect in organic two-dimensional Dirac fermion system

Andhika Kiswandhi* and Toshihito Osada

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

Received 28 September 2021, revised 17 November 2021
Accepted for publication 3 December 2021
Published 20 December 2021

Abstract
We report the observation of nonlinear anomalous Hall effect (NLAHE) in the multilayered organic conductor α-(BEDT-TTF)$_2$I$_3$ in the charge order (CO) insulating phase just under the critical pressure for transition into two-dimensional (2D) massless Dirac fermion (DF) phase. We successfully extracted the finite nonlinear Hall voltage proportional to square current at zero magnetic field. The observed NLAHE features, current direction dependence and correlation with CO, are consistent with the previous estimation assuming 2D massive DF with a pair of tilted Dirac cones. This is the first observation of topological transport in organic conductors, and also the first example of NLAHE in the electronic phase with spontaneous symmetry breaking.

Keywords: strongly correlated systems, topological material, high pressure, transport measurements

Supplementary material for this article is available online (Some figures may appear in colour only in the online journal)

1. Introduction
Nonlinear anomalous Hall effect (NLAHE) in zero magnetic field condition has attracted much interest since its prediction by Sodemann and Fu [1]. The phenomenon is induced by current flowing through the sample and originating from the nonequilibrium distribution of electrons (current-carrying state) on the integral of the Berry curvature. The model is assumed to be time-reversal invariant having Kramers pair states at \( \mathbf{k} \) and \( -\mathbf{k} \) points in the momentum space, whose Berry curvatures are related by \( \Omega(\mathbf{k}) = -\Omega(-\mathbf{k}) \), and requires broken inversion symmetry for nonzero Berry curvature. In such system, the Berry curvature summed over the occupied states must vanish in equilibrium condition, so anomalous velocity effectively cancels out. However, in nonequilibrium state by applying electric current, the electron distribution is shifted by \( \Delta \mathbf{k} \) in the \( \mathbf{k} \)-space. Therefore, the Kramers pair states at \( \mathbf{k} \) and \( -\mathbf{k} \) are not equally occupied, and the sum now can be finite. Generally, in the system with the time reversal symmetry, the sum of the Berry curvature over occupied states in the nonequilibrium current-carrying state is given by the inner product of \( \Delta \mathbf{k} \) and the Berry curvature dipole of the equilibrium state. Due to the finite Berry curvature, the electrons gain a net anomalous velocity \( \mathbf{v}_\perp \) perpendicular to the applied electric field, similar to Hall effect even in the absence of magnetic field. The anomalous current density, \( j_\perp \) is written as \( j_\perp = -e \int \delta f(\epsilon_k) v_\perp(\mathbf{k}) d\mathbf{k} \), where \( f(\epsilon_k) = f_0(\epsilon_k) + \delta f(\epsilon_k) \) is the nonequilibrium Boltzmann distribution function. Here, \( f_0(\epsilon_k) \) is the distribution at equilibrium and \( \delta f(\epsilon_k) \) is the correction to the first order in applied electric field \( E \). Since the anomalous velocity is also proportional to \( E \), the anomalous current density is proportional to \( E^2 \). Therefore, unlike the conventional Hall effect, the anom-
lous current exhibits a second-harmonic behavior, implying a rectification effect since its direction is unchanged even when the excitation current direction is reversed. The signal strength depends on the magnitude of the Berry curvature dipole, which is more prominent in systems with tilted Dirac or Weyl cones with a finite gap.

NLAHE has been observed in several materials, such as few-layer 3D Weyl fermion system WTe$_2$ [2, 3], bulk 3D Dirac semimetal Cd$_3$As$_2$ [4], and thin-film TaIrTe$_4$ [5]. Notably, in the latter, the rectification effect of radiofrequency attributable to NLAHE has been demonstrated. NLAHE has also been considered in p-doped trigonal Te crystal [6], as well as a probe for massive Dirac fermion (DF) state because Berry curvature is nonzero only in the gapped state [7].

The system we are considering is an organic conductor $\alpha$-(BEDT-TTF)$_2$I$_3$, which is well recognized as a multilayer 2D massless DF system with a pair of tilted Dirac cones at high pressure [8]. At the ambient pressure, it shows a metallic conduction at high temperatures. Upon cooling it transitions into an insulating state due to the horizontal-stripe charge ordering (CO) at $T_{\text{CO}} = 135$ K, as schematically shown in figure 1. The metallic state belongs to $P1$ space group, whereas in its insulating state the CO breaks the inversion symmetry, resulting in $P1$ symmetry and appearance of two domains with different charge distributions [9, 10]. With increasing hydrostatic pressure, $T_{\text{CO}}$ decreases and, finally, above a critical pressure $P_c \approx 1.3$ GPa the system enters the massless DF state, where the transport shows a nearly temperature-independent, but metallic behavior.

The massless DF state at high pressure in $\alpha$-(BEDT-TTF)$_2$I$_3$ has been firmly established. Theoretical work [11–13] show gapless Dirac cones, which are tilted and form at general points $-k_0$ and $k_0$ of the 2D Brillouin zone due to its low crystal symmetry [14]. Experimental evidences strongly support the massless DF model. Interlayer magnetoresistance suggests the effect of zero mode Landau level characteristic to DF [15–17]. Specific heat exhibits the characteristic $C \propto T^3$ associated with linear density of states [18]. Furthermore, site-selective NMR supports the tilting of Dirac cones [19].

On the other hand, the details of the transition between the CO state and massless DF remain to be solved. Here, we refer the CO insulator region close to the critical pressure $P_c$ to the weak CO state. It is expected that in the weak CO state the CO is highly suppressed and the Dirac cones are narrowly gapped, realizing insulating massive DF state with finite Berry curvature. Experimentally, the massive DF manifests in the temperature dependent interlayer transport as a reentrant metallic behavior suggesting the edge transport in the insulating state [20]. As for the electronic structure of the weak CO state, however, other possibilities have been proposed. A particular phase diagram for this system suggests a semimetallic CO state between the two states [21]. Furthermore, optical study suggests DFs with a pseudogap behavior at intermediate pressures [22]. More recently, a strong evidence for the massive DF in the weak CO state was reported. It has been shown that the temperature-dependent interlayer magnetoresistance features a peak structure, whose magnetic field dependence can only be reproduced when the insulating massive DF picture is taken into consideration [23].

The 2D massive DF system with tilted Dirac cones is the most simple system with finite Berry curvature dipole. Therefore, the weak CO state in $\alpha$-(BEDT-TTF)$_2$I$_3$ provides an ideal platform to investigate Berry curvature dipole effects such as NLAHE. We have previously considered the NLAHE behaviors in the weak CO state of $\alpha$-(BEDT-TTF)$_2$I$_3$ and estimated its magnitude to be in the observable range [24, 25]. The present study is aimed to confirm these results experimentally.

2. Experimental

A single crystal with a dimension of approximately 0.56 $\times$ 0.48 $\times$ 0.04 mm$^3$ was used. Eight contacts were attached with carbon paint (figure 2(a)), with contact numbering shown in figure 2(b). Since a whole crystal was used, the crystallographic axes can be determined from its shape [26]. The line connecting the corners with 99° angle is the direction of the crystallographic $b$-axis. The $a$-axis direction is then almost parallel to a line joining contacts 4–5. The sample was enclosed in a Be–Cu piston-cylinder type pressure cell with Daphne 7373 as the pressure medium. The pressure at room temperature was determined by measuring the resistance of a manganin wire enclosed with the sample. All measurements were performed in the dc limit by using Keithley 2182 nanovoltmeter and Yokogawa 7651 dc current source.
determined the majority carrier as holes, with a density of the carrier density with conventional Hall measurement and state. Since NLAHE requires carrier imbalance, we checked the sample is in the desired range of pressure for the weak CO pressure (figure 1). At 1.25 GPa, the peak is broad and center, we performed temperature dependent resistance measurements as shown in figure 2(c). The in-plane resistivity data at 1.25 GPa and resistivity upturn at \( T_{\text{min}} = 37 \) K are consistent with known result [27]. In the CO state, \( T_{\text{CO}} \) can be evaluated by finding the peak of \(- (\ln \rho_{zz}) / \Delta T\) of the interlayer resistivity. As the weak CO state is approached, the CO transition becomes less abrupt and more second-order like. We evaluated \( T_{\text{CO}} \) by finding the peak in \( \ln \rho / (\ln T) \) at each pressure (figure 1). At 1.25 GPa, the peak is broad and centered at \( T \approx 20 \) K. These results justify that at \( P = 1.25 \) GPa, the sample is in the desired range of pressure for the weak CO state. Since NLAHE requires carrier imbalance, we checked the carrier density with conventional Hall measurement and determined the majority carrier as holes, with a density of about \( n = 10^{17} \) cm\(^{-3}\). We note that the carrier density is one order of magnitude higher than that used in the calculation \( (n \approx 10^{16} \text{ cm}^{-3}) \) [24, 25].

We measured the current dependence of the Hall voltage (the \( V-I \) curve) from \(-1\) mA to \(1\) mA for four possible current directions at zero magnetic field. To eliminate the mixing of the linear longitudinal resistance due to unexpected but unavoidable misalignment the Hall contacts, we extracted the nonlinear Hall signal by reversing the current direction.

The symmetric Hall signal, \( V_{\perp}^S \), from dc \( V-I \) data can be obtained as

\[
V_{\perp}^S = \frac{V_\perp(I \geq 0) + V_\perp(I \leq 0)}{2},
\]

where \( V_\perp \) is the raw signal taken with Hall electrode configuration. However, \( V_{\perp}^S \) is still contaminated by the nonlinear longitudinal resistance component \( V_{\parallel}^L \) due to contact offset. To obtain the NLAHE component, \( V_{\text{NLAHE}} \), we measured simultaneously signals in Hall (\( V_{\perp} \)) and longitudinal (\( V_{\parallel} \)) configurations. The \( V-I \) signals were then separated into components symmetric and antisymmetric with respect to current reversal as the following

\[
V_{\parallel} = V_{\parallel}^{S} + V_{\parallel}^{A},
\]

where the labels S and A denote the symmetric and antisymmetric components, respectively. At zero magnetic field, there should be no voltage component perpendicular to the applied current. Additionally, scattering does not give perpendicular component, except in the case of broken time-reversal symmetry, such as in anomalous Hall effect. Therefore, the voltage which appears as \( V_{\perp} \) must come only from the Hall contact offset and NLAHE if it exists. We then estimated the amount of nonlinear longitudinal voltage component across the Hall electrodes as \( kV_{\parallel} \), where \( k \) is a proportionality factor. With this, the voltage appearing in the Hall configuration can be written as

\[
V_{\parallel} = kV_{\parallel}^S + (kV_{\parallel}^S + V_{\text{NLAHE}}) = V_{\parallel}^S + V_{\text{NLAHE}}.
\]

Here, \( V_{\parallel}^S \) is zero when the Hall contacts are perfectly aligned. We obtained \( V_{\parallel}^S \) following equation (1) and the factor \( k = V_{\parallel}^S / V_{\parallel}^A \) following equation (3). NLAHE voltage then was obtained by subtracting the longitudinal voltage contribution \( kV_{\parallel}^S \) from \( V_{\parallel}^S \). The details are provided in supplementary information ([https://stacks.iop.org/JPCM/34/105602/mmedia](https://stacks.iop.org/JPCM/34/105602/mmedia)) [28]. From this point, NLAHE signal refers to the symmetrized Hall voltage after the background subtraction.

Symmetrized voltages after background subtraction obtained at \( P = 1.25 \) GPa and \( T = 4.2 \) K as a function of \( I^2 \) for various current directions are shown in figure 3(a). The symmetrized signals show second order \( V \propto I^2 \) behavior. Hereafter, we will denote the resistance and voltage as \( R_{ijkl} \) and \( V_{ijkl} \), where the indices \( i, j, k, \) and \( l \) indicate the positive current, negative current, positive voltage, and negative voltage electrodes, respectively. We also maintain the relative position of the electrodes as shown in the inset of figure 3(a) for all measurements. The configuration \( V_{45,27} \), where the current is directed almost parallel to the \( a \)-axis gives the strongest nonlinear signal. The signal weakens as the current direction is rotated towards the \( V_{72,45} \) configuration, corresponding to the current direction almost parallel to the \( b \)-axis. For \( V_{45,27}, \) \( V_{81,36} \), and \( V_{72,45} \) the resulting nonlinear voltage is positive. Therefore, the electric fields of the nonlinear voltages for those configurations are directed from the positive voltage probe to the negative voltage probe, as indicated by the solid arrow.
However, the nonlinear voltage for the $V_{36,18}$ configuration is negative. Therefore, its nonlinear electric field is directed from the negative to positive voltage probe. NLAHE signal is the strongest when the current is biased parallel to the Dirac cone tilting axis direction. For a 2D system, the optimal current bias direction is constrained only when a mirror symmetry axis is present. In such a case, the largest signal is produced for current bias perpendicular to the mirror axis [1]. The absence of mirror axis in the CO state of $\alpha$-(BEDT-TTF)$_2$I$_3$ means the Dirac cone tilting direction cannot be determined simply from its crystallographic axis.

The nonlinear current due to the NLAHE in the weak CO state in $\alpha$-(BEDT-TTF)$_2$I$_3$ is represented by $j^{(2)} = \left(\chi_{xy}^2 E_x E_y \right) n_x + \left(\chi_{yxx} E_y^2 \right) n_y$ [24, 25]. Here, the $xy$-plane is parallel to the conducting layer, and the tilting direction of Dirac cones is chosen as the $x$-axis. $E = (E_x, E_y)$ is the in-plane electric field, and $n_x$ and $n_y$ are unit vectors in the $x$- and $y$-directions, respectively. The elements $\chi_{xyz}$ and $\chi_{yxx} = - \chi_{xxx}$ are finite nonlinear Hall conductivity elements, which are represented by the Berry curvature dipole of the system. In the current-carrying state, the normal linear transport $j^{(1)} = \sigma_{xx} E$ coexists with the NLAHE, where $\sigma_{xx}$ is the scalar longitudinal conductivity. The total current is given by $j = j^{(1)} + j^{(2)}$. Assuming $\left(\chi_{yxx}/\sigma_{xx}^2\right) j^{(2)} \ll 1$, we can obtain the following asymptotic formula for the anomalous Hall field as a function of electric current directed at an angle $\alpha$ away from the $x$-axis (Dirac cone tilting axis) in the presence of normal linear transport

$$E_{xy} = \frac{\chi_{yxx} \cos \alpha}{\sigma_{xx}} j^2. \quad (4)$$

Therefore, in the case of simple massive DF systems, symmetrized voltage with a current dependence of the form $V^2 \propto \cos \alpha$ is expected, although it does not hold for general systems with more complicated band dispersion such as WTe$_2$ [29]. In $\alpha$-(BEDT-TTF)$_2$I$_3$, the in-plane resistivity anisotropy is typically within a factor of two [30], so the current directions giving the strongest and the weakest signals should remain perpendicular to each other. The appearance of the strongest signal for the current direction close to the $a$-axis is qualitatively consistent with previous report that the $x$-axis in the weak CO state is rotated by approximately $\theta = -30^\circ$ away from the $a$-axis towards the $b$-axis [20].

We note that the signs of $V_{45,27}$, $V_{81,36}$, and $V_{72,45}$ are consistent with rectification effect since the nonlinear voltages are polarized only in a fixed direction relative to the applied current. For $V_{36,18}$, the direction is opposite, so it appears to violate the rectification effect. This can be seen in figure 3(b), where $V_{36,18}$ and $V_{63,81}$ deviate from the cosine curve. Although the voltage is second-order so that the reverse current configuration $V_{63,81}$ will have the same voltage as $V_{36,18}$ and there will be no issue, as mentioned later, we consider that the measurement of $V_{36,18}$ has some accidental problem owing to geometrically non-ideal electrode. The angular dependence of the NLAHE voltage can be visualized as figure 3(c).

Next, data taken at a higher pressure 1.35 GPa represented by $V_{45,27}$ and $V_{81,36}$ are shown in figures 3(d) and (e), respectively. The signal drops significantly, almost vanishes. Here, we consider two possibilities. First, relative to the conductivity at 1.25 GPa, there are conductivity increases by about 3 times at 1.35 GPa and by about 5 times at 1.78 GPa (figure 2(c)). If we assume a constant Berry curvature dipole at those pressures, this corresponds to one and two order of NLAHE signal magnitude decrease at 1.35 GPa and 1.78 GPa, respectively according to equation (4). If this scenario is true, inversion symmetry must be broken at those pressures in order...
for NLAHE to exist. In the second scenario, the system has entered the massless DF state at 1.35 GPa. In the massless DF state, the CO transition is completely suppressed; thus inversion symmetry is preserved and second-order conduction is prohibited. This second scenario is more likely considering the data taken at $P = 1.78$ GPa, where the system is known to be in the massless DF state, and consistent with previous reports on the disappearance of $T_{\text{CO}}$ in the massless DF state and the interlayer transport [20, 23, 27, 31]. The behavior under pressure thus suggests that the system undergoes a transition into massless DF state at 1.25 GPa $< P_{\text{C}} < 1.35$ GPa.

For $P = 1.25$ GPa, the nonlinear Hall coefficient $\chi_{xxx}$ can be estimated using equation (4). Here, we consider $V_{34,36}$ data because they show a large response, while both current density and longitudinal conductivity can be evaluated with a standard geometry. Further, we limit our analysis for data taken at 4.2 K with an applied current range of 0.5–1 mA, where the uncertainty is small. The conductivity at 4.2 K was estimated to be $\sigma_{xx} = 19.9$ S cm$^{-1}$. The NLAHE electric field $E_{xx}$ was taken as the ratio of the symmetrized voltage to the distance between the voltage electrodes (0.41 mm). The current density $j_x$, was estimated from the applied current and the sample geometry. Using the Dirac cone tilting axis direction from reference [20], we estimated $\chi_{xxx} = 0.033 \pm 0.004$ A V$^{-2}$ at 4.2 K. As a comparison, following reference [24] and inputting the experimentally obtained carrier density $n = 10^{17}$ cm$^{-3}$ and the CO gap $\Delta_{\text{CO}} = k_B T_{\text{CO}}$ with $T_{\text{CO}} = 20$ K, the theoretically predicted value is $\chi_{xxx} = 0.36$ A V$^{-2}$. The experimental value, therefore, is smaller by one order of magnitude. This discrepancy is discussed again later.

The temperature dependence of the NLAHE signal for $V_{45,27}$ configuration is shown in figure 4. In general, the signal increases with decreasing temperature, particularly at $T < 20$ K, below $T_{\text{CO}}$ since the normal metallic state is inversion symmetric, so second-order behavior is not allowed. The temperature dependent conductivity as a factor $1/\sigma_{xx}$, in addition, causes a rapid decrease due to its activated form. The NLAHE signal falls with temperature more rapidly than $1/\sigma_{xx}$, supporting the appearance of the signal at $T < 20$ K.

The result suggests that NLAHE may exist in $P = 1.25$ GPa, in the weak CO state of $\alpha$-(BEDT-TTF)$_2$I$_3$. However, there are several issues still remaining in this NLAHE measurement. So far in other NLAHE experiments [2–5] second order longitudinal voltage is not active, whereas it is present in our experiment. In contrast to them, the present NLAHE was observed in a slightly-doped CO insulator. Considering that the CO state of this system could show nonlinear longitudinal transport [32], the finite second-order longitudinal voltage might be reasonable.

In addition, Hall contact misalignment and their finite size and shapes introduce some ambiguity because it is possible to underestimate or overestimate the correction, which can be the cause of voltage sign problem in $V_{36,18}$. In this sense, the present result does not necessarily confirm but strongly support the existence of NLAHE in weak CO state in $\alpha$-(BEDT-TTF)$_2$I$_3$.

It should be noted that in a perfect $\alpha$-(BEDT-TTF)$_2$I$_3$, the Fermi energy is located exactly at the Dirac point due to its 3/4 band filling originating from charge transfer between (BEDT-TTF)$^{+1/2}$ and (I$^-$)$_3$. The chemical potential thus falls inside the mass gap in the massive DF state. On the other hand, the theory [1, 7] was actually formulated assuming metallic state with Fermi energy, which is the chemical potential at zero temperature, located just outside the gap. In the previous work, on the other hand, we discussed the possible NLAHE in the insulating massive DF system simulating $\alpha$-(BEDT-TTF)$_2$I$_3$ as a function of chemical potential or carrier density imbalance quantitatively [24, 25]. When there exist finite carrier imbalance, the thermally excited carriers cause finite NLAHE at finite temperature even if the chemical potential is located in the CO gap [24]. Experimentally it has been shown that although the low temperature resistivity shows a typical insulating behavior ($d\rho/dT < 0$), infrared measurement results indicates Drude conductivity increase at low frequencies, indicating existence of thermally excited free carriers [22].

Finally, we address the discrepancy between $\chi_{xxx}$ obtained in this experiment and the theory. Here, the data were obtained under zero-field cooling, whereas the theory predicted the necessity of the current-field cooling technique [24, 25]. The transition into the CO state breaks the inversion symmetry, resulting in two domains. The two domains are randomly aligned and opposite in sign for NLAHE; thus, under zero field cooling, the signal should vanish. The proposed current-field cooling was expected to align the domains by exploiting dc current-induced magnetization (orbital Edelstein effect), which is also a Berry curvature dipole effect, as the sample is being cooled in a constant magnetic field. As described above, the experimentally determined $\chi_{xxx}$ is an order of magnitude smaller than the predicted value. The discrepancy can be understood since the theoretical prediction assumes a single-domain system, so it gives only the upper limit of $\chi_{xxx}$. The existence of the two types of domains has been investigated by Yamamoto et al using infrared second-harmonic spectroscopy.
[33]. The domains were found to be mobile and macroscopic in size. Difference in proportion of the domains has been reported by Kakiuchi et al [9], so it is likely that one type of domain already dominates across the sample. Therefore, we assert that the nonzero signal observed in this study resulted from asymmetry in the domain formation.

4. Conclusions

In conclusion, we have successfully observed the NLAHE in the weak CO state of \( \alpha-(\text{BEDT-TTF})_2I_3 \). The observed NLAHE features, the current direction dependence and the correlation with the CO relation, are consistent with our previous estimation assuming the 2D massive DF with a pair of tilted Dirac cones. This is the first observation of topological transport in organic conductors, and also the first example of NLAHE in the electronic state with spontaneous symmetry breaking. The rather small value of the observed NLAHE can be explained by the cancellation between two types of CO domains.

Acknowledgments

The author thanks Dr T Taen, Dr K Uchida, Ms A Mori, Mr K Yoshimura, and Dr M Sato for valuable discussions and support. This work was supported by JSPS KAKENHI Grant Numbers JP20H01860 and JP21K18594.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

ORCID iDs

Andhika Kiswandhi https://orcid.org/0000-0002-9099-0049

Toshihito Osada https://orcid.org/0000-0001-5779-8824

References

[1] Sodemann I and Fu L 2015 Quantum nonlinear Hall effect induced by Berry curvature dipole in time-reversal invariant materials Phys. Rev. Lett. 115 216806

[2] Ma Q et al 2019 Observation of the nonlinear Hall effect under time-reversal-symmetric conditions Nature 565 337–42

[3] Kang K, Li T, Sohn E, Shan J and Mak K F 2019 Nonlinear anomalous Hall effect in few-layer WTe_2 Nat. Mater. 18 324–8

[4] Shvetsov O O, Esin V D, Timonina A V, Kolesnikov N N and Deviatov E V 2019 Nonlinear Hall effect in three-dimensional Weyl and Dirac semimetals JETP Lett. 109 715–21

[5] Kumar D, Hsu C-H, Sharma R, Chang T-R, Yu P, Wang J, Eda G, Liang G and Yang H 2021 Room-temperature nonlinear Hall effect and wireless radiofrequency rectification in Weyl semimetal TaIrTe_4 Nat. Nanotechnol. 16 421–5

[6] Tsirkin S S, Puente P A and Souza I 2018 Gyrotropic effects in trigonal tellurium studied from first principles Phys. Rev. B 97 035158

[7] Rostami H and Jurić V 2020 Probing quantum criticality using nonlinear Hall effect in a metallic Dirac system Phys. Rev. B 2 013069

[8] Kajita K, Nishio Y, Tajima N, Suzumura Y and Kobayashi A 2014 Molecular Dirac fermion systems—theoretical and experimental approaches J. Phys. Soc. Japan 83 072002

[9] Kakiuchi T, Wakabayashi Y, Sawa H, Takahashi T and Nakamura T 2007 Charge ordering in \( \alpha-(\text{BEDT-TTF})_2I_3 \) by synchrotron x-ray diffraction J. Phys. Soc. Japan 76 113702

[10] Seo H 2000 Charge ordering in organic ET compounds J. Phys. Soc. Japan 69 805–20

[11] Katayama S, Kobayashi A and Suzumura Y 2006 Pressure-induced zero-gap semiconducting state in organic conductor \( \alpha-(\text{BEDT-TTF})_2I_3 \) salt J. Phys. Soc. Japan 75 054705

[12] Kobayashi A, Katayama S, Suzumura Y and Fukuyama H 2007 Massless fermions in organic conductor J. Phys. Soc. Japan 76 034711

[13] Suzumura Y and Kobayashi A 2013 Zero-gap state in \( \alpha-(\text{BEDT-TTF})_2I_3 \) under hydrostatic pressure J. Phys. Soc. Japan 82 044709

[14] Goerbig M O, Fuchs J-N, Montambaux G and Pi echon F 2008 Tilted anisotropic Dirac cones in quinoid-type graphene and \( \alpha-(\text{BEDT-TTF})_2I_3 \) Phys. Rev. B 78 045415

[15] Osada T 2008 Negative interlayer magnetoresistance and zero-mode Landau level in multilayer Dirac electron systems J. Phys. Soc. Japan 77 084711

[16] Tajima N, Sugawara S, Kato R, Nishio Y and Kajita K 2009 Effect of the zero-mode Landau level on interlayer magnetoresistance in multilayer massless Dirac fermion systems Phys. Rev. Lett. 102 176403

[17] Tajima N and Morinari T 2018 Tilted Dirac cone effect on interlayer magnetoresistance in \( \alpha-(\text{BEDT-TTF})_2I_3 \) J. Phys. Soc. Japan 87 045002

[18] Konoike T, Uchida K and Osada T 2012 Specific heat of the multilayered massless Dirac fermion system J. Phys. Soc. Japan 81 044601

[19] Hirata M, Ishikawa K, Miyagawa K, Tamura M, Berthier C, Basko D, Kobayashi A, Matsuno G and Kanoda K 2016 Observation of an anisotropic Dirac cone reshaping and ferromagnetic spin polarization in an organic conductor Nat. Commun. 7 12666

[20] Mori A, Sato M, Tajima N, Konoike T, Uchida K and Osada T 2019 Anisotropy of Dirac cones and Van Hove singularity in an organic Dirac fermion system Phys. Rev. B 99 035106

[21] Kobayashi A, Suzumura Y, Pi echon F and Montambaux G 2011 Emergence of Dirac electron pair in the charge-ordered state of the organic conductor \( \alpha-(\text{BEDT-TTF})_2I_3 \) Phys. Rev. B 84 075450

[22] Uykur E, Li W, Kuntscher C A and Dressel M 2019 Optical signatures of energy gap in correlated Dirac fermions npj Quantum Mater. 4 119

[23] Yoshimura K, Sato M and Osada T 2021 Experimental confirmation of massive Dirac fermions in weak charge-ordering state in \( \alpha-(\text{BEDT-TTF})_2I_3 \) J. Phys. Soc. Japan 90 035701

[24] Osada T and Kiswandhi A 2020 Possible current-induced phenomena and domain control in an organic Dirac fermion system with weak charge ordering J. Phys. Soc. Japan 89 103701

[25] Osada T and Kiswandhi A 2020 Erratum: possible current-induced phenomena and domain control in an organic Dirac fermion system with weak charge ordering J. Phys. Soc. Japan 89 103701

[26] Sugano T, Saito G and Kinoshita M 1986 Conduction-electron-spin resonance in organic conductors: \( \alpha \) and \( \beta \) phases of \( d|b(isoethylenedithiolo)tetrahydrofulvalene|tritriodide \) \( [\text{BEDT-TTF}_2]I_3 \) Phys. Rev. B 34 117–25
[27] Liu D, Ishikawa K, Takehara R, Miyagawa K, Tamura M and Kanoda K 2016 Insulating nature of strongly correlated massless Dirac fermions in an organic crystal Phys. Rev. Lett. 116 226401

[28] (Supplemental material) the details are provided online.

[29] Du Z Z, Wang C M, Lu H-Z and Xie X C 2018 Band signatures for strong nonlinear Hall effect in bilayer WTe$_2$ Phys. Rev. Lett. 121 266601

[30] Iimori S, Kajita K, Nishio Y and Iye Y 1995 Anomalous metal-nonmetal transition in $\alpha$-(BEDT-TTF)$_2$I$_3$ under high pressure Synth. Met. 71 1905–6

[31] Wojciechowski R, Yamamoto K, Yakushi K, Inokuchi M and Kawamoto A 2003 High-pressure Raman study of the charge ordering in $\alpha$-(BEDT-TTF)$_2$I$_3$ Phys. Rev. B 67 224105

[32] Kodama K et al 2012 Charge transport in charge-ordered states of two-dimensional organic conductors, $\alpha$-(BEDT-TTF)$_2$I$_3$ and $\alpha'$-(BEDT-TTF)$_2$IBr$_2$ J. Phys. Soc. Japan 81 044703

[33] Yamamoto K, Kowalska A A and Yakushi K 2010 Direct observation of ferroelectric domains created by Wigner crystallization of electrons in $\alpha$-[bis(ethylenedithio)-tetrathiafulvalene]$_2$I$_3$ Appl. Phys. Lett. 96 12290