Correlation-driven organic 3D topological insulator with relativistic fermions

Exploring new topological phenomena and functionalities induced by strong electron correlation has been a central issue in modern condensed-matter physics. One example is a topological insulator (TI) state and its functionality driven by the Coulomb repulsion rather than a spin–orbit coupling. Here, we report a ‘correlation-driven’ TI state realized in an organic zero-gap system $\alpha$-(BETS)$_2$I$_3$. The topological surface state and chiral anomaly are observed in temperature and field dependences of resistance, indicating a three-dimensional TI state at low temperatures. Moreover, we observe a topological phase switching between the TI state and non-equilibrium Dirac semimetal state by a dc current, which is a unique functionality of a correlation-driven TI state. Our findings demonstrate that correlation-driven TIs are promising candidates not only for practical electronic devices but also as a field for discovering new topological phenomena and phases.

Topological insulator (TI) is a new class of materials that possess both bulk insulating and exotic surface/edge states. Realizing a TI state usually requires a strong spin–orbit coupling (SOC) of heavy elements, which induces a band inversion and an insulating bulk gap. Thus, it is generally challenging to observe a TI state and its unique physical features in materials composed of light atoms, such as carbon-based systems, because of their weak SOC. Although several materials, such as two-dimensional organometallic systems, have been theoretically proposed as candidates for TIs consisting of light atoms, no real system has been confirmed experimentally at present. Recent theoretical investigations propose another approach to realize a TI state using a strong electron correlation where the Coulomb interaction opens the bulk gap.

A correlation-driven TI is significant in science because it broadens the range of materials that can form TI states and has the potential to add new functions unique to correlated systems. Strongly correlated organic conductors with a topological band structure are promising candidates for this type of TIs. Here, we report a three-dimensional (3D) TI state realized in a quasi-two-dimensional (quasi-2D) organic conductor $\alpha$-(BETS)$_2$I$_3$ (Fig. 1a) and its anomalous transport properties, where BETS denotes bis(ethylenedithio)tetraselenafulvalene.
proposed to date, α-(BETS)$_3$I$_3$ is expected to behave as a stacked 2D-TI (weak 3D-TI) with a gapless state on the edge of the conducting plane (ab-plane) (Fig. 1c). However, experimental evidence of the 2D-TI state such as a quantum spin Hall effect has not been observed.

Results

We carefully performed electrical resistance measurements by the setups shown in Fig. 2a, b to verify the accurate low-temperature electronic state of α-(BETS)$_3$I$_3$. Figure 2c shows the temperature dependence of the in-plane ($I_{ab}$-plane) and out-of-plane ($I_{c}$-axis) resistances of α-(BETS)$_3$I$_3$. The in-plane resistance (Sample #1) slightly decreases with decreasing temperature and an exponential increase is observed at the MI transition temperature of 50 K. Furthermore, from 50 to 35 K, $R(T)$ can be fitted well using the Arrhenius equation $R(T) = R_0 \times e^{(\Delta E/k_B T)}$, with an estimated gap size of $\Delta \sim 30$ meV, which agrees with previous reports. Since the gapless state exists only on the small area of the sample edges and most parts of the crystal behave as a semiconducting insulator in the stacked 2D-TI state, it is natural that the resistance increases following the Arrhenius law. However, the slope of the in-plane resistance becomes smaller and saturates at lower temperatures, with a step-like anomaly observed between 35 and 10 K (orange region). Such saturation of resistance is often observed in a 3D-TI with a surface conduction, which is a different character from that expected for 2D-TIs. The out-of-plane resistance (Sample #2) is even more anomalous; an abrupt drop between 10 and 35 K and a temperature-independent behavior below 10 K are observed, while a sharp increase is observed at 50 K. Previous bulk measurements, such as the magnetic susceptibility, NMR, and lattice constant measurements, have not detected any anomalies and phase transitions near 35 K. Therefore, the anomaly in the in- and out-of-plane resistances suggests the emergence of surface conduction around 35 K. This indicates that a dimensional crossover from 2D to 3D occurs at this temperature and the actual low-temperature electronic state of α-(BETS)$_3$I$_3$ is a 3D-TI state (Fig. 1c) despite theoretical predictions. The dimensional crossover in TIs has been theoretically predicted, for example, in cold-atomic gases in optical lattices or quasi-2D layered systems, but this is the first experimental observation in solids.

To corroborate the presence of the metallic surface state associated with the 2D–3D crossover, the in-plane resistances were measured using three different configurations; i.e., the standard four-terminal, inverted, and non-local configurations (Fig. 2a). In the inverted configuration, where both the current and voltage terminals were separately attached to the top and bottom surfaces of the crystal, the current flow near the bottom surface is mostly attributed to the surface conduction. In the non-local configuration, where both the current and voltage terminals were attached on the top surface, the observed resistance is expected to exclude the bulk contribution and to be more sensitive to the surface state than the inverted configuration. Figure 2d shows the comparison of the temperature dependences of the in-plane resistance measured by the standard configuration for Sample #1 (blue), the inverted configuration for Sample #3 (green), and the non-local configuration for Sample #4 (orange). For the inverted and non-local configurations, flattened resistance curves are observed below 10 K, which is characteristic for...
3D-TIs where the conductivity is dominated by the surface contribution\[^{5,6}\], but not for 2D-TIs without surface conduction. In addition, the observed resistance in the non-local configuration monotonically increases around 35 K and becomes flattened below 10 K, whereas the resistance curves in the standard and inverted configurations show step-like anomalies in this temperature range. These results indicate the emergence of the surface contribution and are consistent with our interpretation that the 2D–3D crossover occurs between 35 and 10 K. The saturating tendency of resistance observed in the standard configuration below 10 K can be attributed to the combination of the exponential increase of the bulk state resistance and the constant value of the surface resistance. More detailed comparisons between the in-plane and out-of-plane resistance and discussions on the sample dependence are provided in Sections 1 and 2 of Supplementary Information.

Next, we performed magnetoresistance (MR) measurements up to high magnetic fields to reveal the topological character of $\alpha$-(BETS)$_2$I$_3$. If $\alpha$-(BETS)$_2$I$_3$ is a 3D-TI with a narrow band gap at low temperatures, the thermally excited carriers are expected to exhibit the unique properties of relativistic 3D Weyl fermions due to the Dirac cone type band dispersion of this compound\[^{21,22}\]. On the other hand, the unique properties of relativistic 3D Weyl fermions due to the Dirac–conical component owing to the slight misalignment of magnetic fields to reveal the topological character of $\alpha$-(BETS)$_2$I$_3$ is a 3D-TI with a narrow band gap at low temperatures, the thermally excited carriers are expected to exhibit the unique properties of relativistic 3D Weyl fermions due to the Dirac cone type band dispersion of this compound\[^{21,22}\].

![supplementary information](https://doi.org/10.1038/s41467-023-37293-3)

**Fig. 3 | Negative magnetoresistance (MR) in $\alpha$-(BETS)$_2$I$_3$.** a) Longitudinal MR of $\alpha$-(BETS)$_2$I$_3$ ($I//B$). The magnetic-field dependence of MR was measured at several temperatures. The quadratic negative magnetoresistance can be observed below 4 K. b) Schematic of the charge flow induced by CME. In a magnetic field, the positive magnetoconductance (MG) due to the chiral anomaly flows between two Weyl nodes with different chirality $x = \pm 1$ and MG at 1.6 K. The green dashed line denotes the fitting curve $MG = C_B B$ with $C_B = 4.92 \times 10^{-6}$. Below 15 T, MG follows $B^2$ dependence, indicating the CME-induced MG at 1.6 K. The hump-like positive MG can be observed below 5 T, implying weak localization.

Figure 3a shows the normalized in-plane MR of $\alpha$-(BETS)$_2$I$_3$ (Sample #1) in the configuration of $I//B$, which results in a negligible effect of the Lorentz force on MR. In the high-temperature region ($T > 10$ K), a positive MR monotonically increases with $B$. However, a dip-like anomaly is observed below 25 T with a further decrease in the temperature ($T < 10$ K). Finally, the dip structure converts into a negative MR below 4 K. At the lowest temperature of the measurement ($T = 1.6$ K), the negative MR reaches $-14\%$ at $B = 25$ T. Although a large negative MR is often observed in magnetic molecular compounds owing to electron–spin scattering\[^{10}\], $\alpha$-(BETS)$_2$I$_3$ is non-magnetic. Thus, the negative MR in $\alpha$-(BETS)$_2$I$_3$ can be attributed to CME. In the case of Dirac band systems, CME induces a charge flow in the direction of $I$ (and $B$) to compensate for the chirality imbalance between the two Weyl nodes with distinct chirality $x = \pm 1$ and $x = -1$ (Fig. 3b). In theories of CME\[^{19,20}\], the positive magnetoconductance (MG) in weak magnetic fields is expressed as $MG = -C_B B$ with $C_B$ being a fitting parameter. Figure 3c shows the magnetic-field dependence of MG and the results of the quadratic fitting. The consistency of the fitting in a wide magnetic field range ($B < 10$ T) suggests that the positive MR (negative MR) in $\alpha$-(BETS)$_2$I$_3$ is induced by CME. Above 25 T, MG exhibits a downturn behavior. Because the magnetic field was applied parallel to the 2D conduction layers, the effects of the Landau level splitting and quantum limit are considered to be small\[^{18,19}\]. Thus, the downturn observed in the high-field region is probably caused by the transverse MR or Hall component owing to the slight misalignment of $B$. Moreover, an additional positive MR of less than $1\%$ in weak magnetic fields ($B < 5$ T) was observed, as shown in the inset of Fig. 3c. Owing to its extremely small value and tendency to appear only at low temperatures and weak magnetic fields, it is considered to be a contribution of the weak localization effect due to the impurities\[^{7}\]. These results demonstrate that the conduction electrons in $\alpha$-(BETS)$_2$I$_3$ behave as 3D Weyl fermions. The original band structure in this compound has a Dirac-cone-type linear dispersion, where the small band gap of $\Delta$ - $30$ meV is induced by electron correlations. Therefore, the thermally excited electrons can hold a character of the chiral fermions\[^{7}\]. A detailed
Another characteristic magnetotransport property of Dirac systems is a large positive MR effect in the $I \perp B$ configuration\textsuperscript{34–37}. Figure 4a shows the magnetic field dependence of MR of Sample #1 when $B$ is perpendicular to the $ab$-plane. In contrast to the $I \parallel B$ configuration, a large and non-saturating positive MR is observed. The positive MR increases with decreasing temperature and exceeds 400\% at 2.3 K and 50 T. In addition, it shows $B^2$-dependence at lower magnetic fields ($B < 20$ T) and a sublinear dependence at higher magnetic fields ($B > 20$ T). Such a non-saturating MR is characteristic of compensated metals, such as Dirac semimetals, wherein holes and electrons compensate each other, thereby resulting in a positive MR effect by the Lorentz force that continues to appear in high magnetic fields. Indeed, the positive MR can be qualitatively reproduced using the two-carrier model as well as other topological semimetals\textsuperscript{38–40} (see Supplementary Information). Figure 4b shows the MR of Sample #5 at 4.3 K measured at several angles $\theta$. The angle $\theta$ is defined by the angle between $I$ and $B$. c Angle dependence of MR of Sample #4 at 30 T. The orange dashed line is the fitting line of $\sin^2 \theta$.

Comparison of MRs of Sample #1 of $I \perp B$ and $I \parallel B$ at 2.3 and 100 K, respectively. The strong anisotropy of MR can be observed only at low temperatures.

e Temperature dependence of the magnitude of the transverse MR of Sample #1 and the out-of-plane resistance of Sample #2. Below 35 K, the positive MR begins to increase, which agrees with the appearance of the surface conduction on the $ab$-plane.

Fig. 4 | Positive and non-saturating MR in $\alpha$-(BETS)$_3$I$\textsubscript{3}$. a Transverse MR of $\alpha$-(BETS)$_3$I$\textsubscript{3} (I$\perp$B). The magnetic-field dependence of MR of Sample #1 was measured at several temperatures. a Large positive and non-saturating MR can be observed. b The magnetic field dependence of MR of Sample #5 at 4.3 K measured at several angles $\theta$. The angle $\theta$ is defined by the angle between $I$ and $B$. c Angle dependence of MR of Sample #4 at 30 T. The orange dashed line is the fitting line of $\sin^2 \theta$. d Comparison of MRs of Sample #1 of $I \perp B$ and $I \parallel B$ at 2.3 and 100 K, respectively. The strong anisotropy of MR can be observed only at low temperatures.

e Temperature dependence of the magnitude of the transverse MR of Sample #1 and the out-of-plane resistance of Sample #2. Below 35 K, the positive MR begins to increase, which agrees with the appearance of the surface conduction on the $ab$-plane.

Discussion on the negative MR is in Section 3 of Supplementary Information.

Another characteristic magnetotransport property of Dirac systems is a large positive MR effect in the $I \perp B$ configuration\textsuperscript{34–37}. Figure 4a shows the magnetic field dependence of MR of Sample #1 when $B$ is perpendicular to the $ab$-plane. In contrast to the $I \parallel B$ configuration, a large and non-saturating positive MR is observed. The positive MR increases with decreasing temperature and exceeds 400\% at 2.3 K and 50 T. In addition, it shows $B^2$-dependence at lower magnetic fields ($B < 20$ T) and a sublinear dependence at higher magnetic fields ($B > 20$ T). Such a non-saturating MR is characteristic of compensated metals, such as Dirac semimetals, wherein holes and electrons compensate each other, thereby resulting in a positive MR effect by the Lorentz force that continues to appear in high magnetic fields. Indeed, the positive MR can be qualitatively reproduced using the two-carrier model as well as other topological semimetals\textsuperscript{38–40} (see Supplementary Information). Figure 4b shows the MR of Sample #5 at 4.3 K measured at several angles $\theta$. The angle $\theta$ is defined by the angle between $I$ and $B$. c Angle dependence of MR of Sample #4 at 30 T. The orange dashed line is the fitting line of $\sin^2 \theta$.
The characteristic is reminiscent of those of a tunnel diode or correlation-driven TI. A significant difference from conventional TIs is that the bulk insulating state is sustained by the electron correlation. It is known that the conventional insulating states derived from the electron correlation, such as Mott insulators and charge-ordered (CO) insulators, can be electrically controlled using a dc current. Similarly, a correlation-driven TI state is expected to be controlled by a dc current and be changed into another topological state. Such electrical controllability of topological matters would further broaden practical applications for electronic devices. Here, we challenge to control the correlation-driven TI state of α-(BETS)$_3$I$_3$ by an external dc current.

Figure 5a, b show the current–voltage ($I$–$V$) characteristic and current dependence of the resistance of Sample #6 under constant current conditions, respectively. These measurements were performed under the exchange gas condition using a pulsed current for $<$10 ms to avoid the influence of Joule heating. We observe a giant nonlinear conduction effect at temperatures below the MI transition (Fig. 5a). The sample resistance ($R/I$) decreases with increasing current, and a drastic reduction of approximately four orders of magnitude in resistance is observed at 10 K (Fig. 5b). Such an anomalous $I$–$V$ characteristic is reminiscent of those of a tunnel diode or correlation-driven insulators. The $I$–$V$ curves show a local peak at some voltages ($V_{\text{peak}}$); for instance, $V_{\text{peak}}$ is 0.072 V (=7.2 V/cm) at 40 K (Fig. S5a). Note that the value of $V_{\text{peak}}$ almost corresponds to the threshold voltage $E_{\text{th}}$ of nonlinear conduction. Based on the Zener breakdown model, which is a classical mechanism for nonlinear conduction, $E_{\text{th}}$ is estimated as $E_{\text{th}} = -\Delta/ea$, where $\Delta$ is the gap size, $e$ is the elementary charge, and $a$ is a lattice parameter. Using $a \sim$ 1 nm from crystallographic data and $\Delta = 30$ meV, $E_{\text{th}}$ is estimated to be $\sim$0.3 MV/cm, which is clearly smaller than the estimated $E_{\text{th}}$ value for this compound. Consequently, our $I$–$V$ findings demonstrate that correlation-driven TIs hold the potential to be a new playground for the science of topology. This unique feature has not been observed in conventional TIs driven by SOC. Topological materials, whose physical properties can be electrically controlled, are desirable not only as a practical electronic device but also as a system for seeking a novel topological phase and phenomenon. Consequently, our findings demonstrate that correlation-driven TIs hold the potential to be a new playground for the science of topology.

In summary, we revealed that α-(BETS)$_3$I$_3$ is a correlation-driven 3D-TI with relativistic fermions at low temperatures. In addition, we...
demonstrated the topological phase switching phenomena between the Dirac semimetal and TI by an external current. These findings provide a new direction for the development of topological materials such as TIs and Dirac/Weyl semimetals and for the exploration of their functionality concerning device applications. The origin of the 2D–3D crossover near 35 K and the detailed mechanism of bulk gap closure by applying a dc current are still open questions. The theoretical investigation of the topological characters of $\alpha$-(BETS)$_2$I$_3$ will be presented in a future study.

Methods

Single crystals of $\alpha$-(BETS)$_2$I$_3$ were grown using a standard electrochemical method. The in-plane and out-of-plane resistances as shown in Fig. 2 were measured using the four-terminal method in a physical property measurement system (PPMS, Quantum Design). To perform the standard in-plane resistance and magnetoresistance (MR) measurements, four gold wires were attached to the conducting plane (ab-plane) using a carbon paste. In the case of the in-plane resistance measurement for Sample #3 with the inserted configuration, two gold wires were connected to the current electrodes on the top surface and the other two wires were connected to the voltage electrodes on the bottom surface. In the case of the non-local resistance measurement for Sample #4, both two gold wires for the current electrodes and the other two wires for the voltage electrodes are on the top surface. Dimensions of measured Samples #1–#7 are summarized in Table S1. Because the crystals were very thin, there is considerable ambiguity in thickness $t$. In the case of out-of-plane measurements, two gold wires were attached to the top of the ab-plane, whereas the other two were attached to the back.

The MR measurements in pulsed magnetic fields were performed using the standard ac four-terminal method with an excitation voltage at a frequency of 2–5 kHz. The amplitude of excitation voltage was set within the ohmic conduction range of the samples. Regarding the low-noise ac resistance measurements, the voltage signal was amplified using pre-amplifiers (Model SR560, Stanford Research Systems). Furthermore, the angle of $B$ was calculated as the ratio of induced voltages of multiple pickup coils, and the temperature was measured using a calibrated Cernox thermometer and a temperature controller (LakeShore model 335).

The current–voltage $(I–V)$ characteristics were measured using a short pulse current of 10 ms produced by a current source (NI-9265, National Instruments) and a multimeter (Model 2000, Keithley). To avoid the self-heating effect, this measurement was performed under exchange gas conditions. We also measured the temperature dependence of resistance with five different excitation current values (1, 4, 8, 12, and 16 mA) to estimate the size of the band gap in each condition and to discuss the current-induced electronic state in detail (see Section 5 in Supplementary Information).

The current dependence of the longitudinal MR was measured using the four-terminal method with an excitation voltage and a dc offset current. The dc offset current ($I_{\text{dc}}$) was produced by the current source NI-9265. The amplitude of the ac current was set to -10% of the magnitude of $I_{\text{dc}}$ to satisfactorily avoid the change of electronic state by an ac component. Although the application of the excitation ac voltage induced a mismatch between the actual sample resistance and the estimated value from the $I$–$V$ characteristic, a combination of dc and ac did not have extrinsic effects on the MR properties. Therefore, the effect of the current-induced electronic state on the magnetic response could be investigated.

To confirm that the current dependence of MR is intrinsic, we also measured it in steady magnetic fields by using PPMS (see Section 5 in Supplementary Information). In this measurement, the MR was measured by the standard dc four-terminal method by applying a short pulse of excitation current for 10 ms.

Data availability

Source data are provided with this paper. All other data that support the findings of this study are available from the corresponding author upon request. Source data are provided with this paper.

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Author contributions
T.N., S.I., and Y.K. performed the magnetotransport measurements, data analysis, and preparation of the manuscript. T.N., H.A., and Y.N. synthesized the single crystals of α-(BETS)$_2$I$_3$. All authors contributed to the writing of the manuscript.

Competing interests
The authors declare no competing interests.

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