Interaction induced quantum spin Hall ordered insulator state in the organic Dirac electron system $\alpha$-(BEDT-TSeF)$_2$I$_3$

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To elucidate the low-temperature ($T$) insulating state of $\alpha$-(BEDT-TSeF)$_2$I$_3$ ($\alpha$-(BETS)$_2$I$_3$), we constructed a two-dimensional extended Hubbard model with transfer integrals, spin-orbit coupling (SOC), and long-range Coulomb interactions, based on the ab initio many-body perturbation calculations. We investigated the low-$T$ electronic state using the Hartree-Fock approximation, and found that the interaction induced quantum spin Hall (QSH) ordered insulator state similar to the one proposed in the honeycomb lattice model is a candidate for the insulating state. We found that the phase modulations owing to the SOC alone and the interaction induced QSH order give the same spin Chern number. As a result, the energy gap owing to the SOC is continuously increased by the development of the interaction induced QSH order. The DC resistivity and the Onsager phase factor calculated in the interaction induced QSH ordered insulator state are consistent with recent experimental results. Such an interaction induced QSH order mechanism generally has the advantage of stabilizing a large-gap topological order in various systems with weak spin-orbit interactions.

I. INTRODUCTION

For the past few decades, Dirac electrons in solids such as graphene [1–2], bismuth [3–4], and several organic conductors [5–12] have attracted attention in both theoretical and experimental aspects. These materials exhibit various physical properties, including quantum conduction by the universal conductivity [13], large diamagnetism [14], and anomalous electron correlation effects. Furthermore, the quantum spin Hall (QSH) effect is found in the Dirac electron system with spin-orbit coupling (SOC) and attracting attention from the aspect of the application to spintronics [15–17].

The QSH effect in the Dirac electron system has been investigated theoretically in numerous early studies for the honeycomb lattice model such as graphene. In the calculation using a tight binding model [18], it was shown that the quantum Hall effect appears by considering the change in the imaginary part of the transfer integrals between the next-nearest-neighbor sites. Kane and Mele theoretically showed that the SOC causes a topological insulator (TI) state with the QSH effect [19–21]. Furthermore, even without the above contributions, Raghu et al. investigated the possibility of the QSH and quantum anomalous Hall (QAH) effects originating from only the Coulomb interaction contribution using analysis by the extended Hubbard model based on the mean-field theory, and found that the interaction induced QSH and QAH ordered insulator states were stabilized when the next-nearest-neighbor interaction is much stronger than the nearest-neighbor interaction [22]. Both the TI and interaction induced QSH ordered insulator states show approximately the same physical properties, such as the QSH effect. The TI state is a band insulator caused by the contribution of intrinsic SOC alone, whereas the interaction induced QSH ordered insulator state is an ordered state derived from spontaneous symmetry breaking caused by the Coulomb interaction contribution. Subsequently, additional analyses such as the density matrix renormalization group suggested that the interaction induced QSH ordered insulator state was not actually stabilized in the honeycomb lattice model [23–34]. However, it has recently been suggested that other materials such as digital transition metal oxide hetero structures can realize the interaction induced QSH ordered insulator state [23, 35–37]. It has been suggested that the TI state appears owing to strong SOC [19–21, 24]. If a QSH effect can be caused by the Coulomb interaction contribution, there is an advantage that the QSH effect can be observed with various different materials. In this study, we proposed the possibility that the interaction induced QSH ordered insulator state is realized in an organic Dirac electron system $\alpha$-(BEDT-TSeF)$_2$I$_3$ [BEDT-TSeF = bis(ethylenedithio)tetrastelenafulvalene] (BETS) at low temperature ($T$).

In organic Dirac electron systems such as $\alpha$-(BEDT-TTF)$_2$I$_3$ [BEDT-TTF = bis(ethylenedithio)tetrathiafulvalene] (ET) and $\alpha$-(BETS)$_2$I$_3$, which are the primary focus of this study, Dirac cones owing to accidentally degeneracy appear in the wavenumber space, and these Dirac points coincide with the Fermi energy [3, 12, 38–40]. The energy scale of the transfer integrals in them is as small as approximately 1/10 of those of other Dirac electron systems such as graphene. Furthermore, the Coulomb...
interaction between Dirac electrons is significant compared to the transfer integrals. Therefore, it is expected that electron correlation effects significantly contribute to the electronic state in both materials. The crystalline structure of α-(ET)$_2$I$_3$ (α-(BETS)$_2$I$_3$) has four ET (BETS) molecules in a unit cell and belong to the space group $P\bar{1}$, and inversion symmetry exists in the two-dimensional (2D) conduction plane composed of ET and BETS molecules [38].

The 2D massless Dirac electron system in α-(ET)$_2$I$_3$ transitions to the insulator phase with the horizontal stripe charge order [41–43] owing to inversion symmetry breaking caused by the nearest-neighbor Coulomb interactions. Mass generation because of the horizontal stripe charge order leads to unique transition to the insulator phase with the horizontal stripe charge order [41–43] owing to inversion symmetry breaking caused by the nearest-neighbor Coulomb interactions. Therefore, it is expected that the interaction between Dirac electrons is significant compared to the transfer integrals. Therefore, there is an insulating state at low-$T$ of α-(BETS)$_2$I$_3$, but its candidate ordered state has not been clearly elucidated. The above experimental results have suggested that this insulating state is unrelated to the charge, bond, and spin order.

Furthermore, a recent experiment reported that the temperature at which the insulating state appears decreases with the increase in hydrostatic pressure $P$ and the insulating state completely vanishes at $P \approx 5$ kbar [60–64]. Additionally, Shubnikov-de Haas oscillation measurements at the low-$T$ region also indicated that the Onsager phase factor switches from 1/2 to zero at the same pressure. These results suggest that not only SOC but also the electron correlation effect plays an important role in the α-(BETS)$_2$I$_3$ electronic state.

To elucidate the low-$T$ insulating state of α-(BETS)$_2$I$_3$, in our recent study [65], the low-$T$ electronic state of α-(BETS)$_2$I$_3$ has been calculated using a 2D Kane Mele Hubbard model with Hartree approximation; a spin ordered massive Dirac electron phase, which involves only time-reversal symmetry breaking, was obtained as the stable solution. However, signs of a spin order have not been observed in recent NMR experiments [62–63]. We needed to consider not only the effect of the onsite Coulomb interaction with the electronic state but also the contribution of long-range components because these interactions remain finite in the Dirac electron system, even when the screening effect is considered [60–64]. In addition, the treatment of SOC had been oversimplified because the coupling constant and spin vector were set as constants, which is not a good approximation in α-(BETS)$_2$I$_3$ [55].

In this study, to elucidate the low-$T$ insulating state of α-(BETS)$_2$I$_3$ in consideration of contributions of more long-range components of the Coulomb interactions and exact SOC values derived from the crystalline structure, we estimated the Coulomb interaction including the long-range component and SOC from the first-principles calculation, and constructed an extended Hubbard model. In particular, we focused on the effects of the nearest- and next-nearest-neighbor interactions to investigate the possibility that the interaction induced QSH ordered insulator state proposed in preceding studies for the honeycomb lattice model [22–23] is realized in α-(BETS)$_2$I$_3$ at low-$T$. As a result of the analysis of the Hartree-Fock approximation, it was found that the interaction induced QSH ordered insulator state is realized in both low-$T$ and high-$T$ regions. When the SOC is absent, both the interaction induced QAH and QSH ordered insulator states can appear and these states are degenerated. When the SOC exists, only the interaction induced QSH ordered insulator state is stabilized. The interaction induced QSH ordered insulator state has approximately the
same physical properties as the TI state caused only by the SOC [59] [I0] [I5] but has finite order parameters and a band gap corresponding with the interaction induced QSH order. As \( T \) is decreased, the band gap increases owing to the contribution of the next-nearest-neighbor Coulomb interaction. In other words, the SOC gap was enhanced by the long-range Coulomb interaction effect.

This tendency is well-consistent with the experimental results such as transport phenomena [59] [I4].

The remainder of this paper is organized as follows. In Section II, we performed first-principles calculation using X-ray data at 30 K under ambient pressure for constructing a 2D extended Hubbard model with SOC. The Coulomb interactions, including the long-range components, are calculated using constrained random phase approximation (cRPA). As \( \alpha-(BETS)_2I_3 \) has the Dirac cone near the Fermi energy and does not have Fermi surface, the long-range components of the Coulomb interaction are expected to survive even when the screening effect is considered [66] [68]. In fact, we confirmed the value of the next-nearest-neighbor interaction is approximately of the same order as the nearest-neighbor interaction in \( \alpha-(BETS)_2I_3 \). In Section III, the electronic state of \( \alpha-(BETS)_2I_3 \) at low-\( T \) is first calculated applying the Hartree-Fock approximation. It was found that the interaction induced QSH order is stabilized by the Coulomb interaction, which was obtained by the cRPA method. Next, the stability of the interaction induced QSH order owing to the presence of the nearest-neighbor and next-nearest-neighbor Coulomb interactions was investigated, and the phase modulation of the transfer integrals owing to the emergence of the interaction induced QSH order was calculated. We confirmed that the interaction induced QSH ordered insulator state suggested in the honeycomb lattice model [22] was stabilized in \( \alpha-(BETS)_2I_3 \) by the contribution of the nearest- and next-nearest-neighbor Coulomb interactions. Moreover, physical quantities, such as the Onsager phase factor and DC conductivity, were calculated using the semiclassical theory [69] [I74], Nakano-Kubo formula, and \( T \)-matrix approximation [15] [I74] [I77], and compared with a recent experiment [60]. We demonstrated that when the insulating state at low-\( T \) is assumed to be the interaction induced QSH ordered insulator state, experimental results [59] [I01] can be explained without any contradiction [59] [I01]. Finally, we investigated the effect of spin fluctuations on the electronic state of \( \alpha-(BETS)_2I_3 \) using random phase approximation (RPA) [7] [I49] [I52]. For a weak interaction case, when \( T \) decreased, only ferrimagnetic spin polarization appears owing to the characteristic wave function of \( \alpha-(BETS)_2I_3 \) [I8]. This behavior is similar to the ferrimagnetic spin polarization observed in \( \alpha-(ET)_2I_3 \) [I50].

In Sec. IV, the study is summarized and the relationship with preceding studies is discussed, and the scope for future research is presented.

**II. MODEL AND FORMULATION**

**A. Effective model based on first-principles calculation**

First-principles calculation was performed to derive an effective model using the X-ray crystal structural data of \( \alpha-(BETS)_2I_3 \) at 30K under ambient pressure [38]. As the exchange-correlation function, GGA was used in the Quantum espresso (QE) package [79] [I80]. We used SG15 optimized norm-conserving Vanderbilt fully relativistic pseudopotentials to consider the effect of SOC in the first-principles calculation [I31]. We set the cutoff energies of the wave functions and charge densities as 80 and 320 Ry, respectively, and the mesh of wave number \( k \) as \( 5 \times 5 \times 3 \). After the first-principles calculation, to obtain the transfer integrals with SOC, the maximally localized Wannier functions (MLWFs) were created using the Wannier90 code [I82]. Eight bands near the Fermi energy were selected to construct the MLWFs, and the initial MLWF coordinates were set at the center of the BETS molecule in the unit cell.

Figure 1(a) shows the crystal structure of the unit cell for \( \alpha-(BETS)_2I_3 \). The box drawn by the black line represents the unit cell, and there are four BETS molecules.

![Figure 1](image-url)
The Wannier interpolation performed using Wannier90. The real-space distribution of the MLWFs at each center of A and A' sites include inversion symmetric and C sites are non-equivalent. The B and C sites, and A' sites are crystallographically equivalent but the A, B and C sites are the unit cell. The A and sites labeled A, A’, B, and C in the unit cell. The A and A’ sites are crystallographically equivalent but the A, B and C sites are non-equivalent. The B and C sites, and the center of A and A’ sites include inversion symmetric points. The real-space distribution of the MLWFs at each unit cell site are also shown. Figure 1(b) shows the energy band structure with SOC calculated using QE and the Wannier interpolation performed using Wannier90. The energy origin is set at the Fermi energy.

We also calculated the long-range Coulomb interaction considering the screening effect using the eCPA method in the RESPACK code [34]. We set the energy cutoff of the dielectric function as 5.0 Ry.

Figure 2 (a) and (b) show the two-dimensional (2D) lattice structures of α-(BETS)$_2$I$_3$. The nearest-neighbor and next-nearest-neighbor components of the transfer integrals and the Coulomb interactions considered in our calculation are also illustrated. In our calculation, spin polarization was not considered. However, as the energy scale of the spin polarization is small compared to the Coulomb interactions, its effect on the Coulomb interactions may not be significant. The detailed values of transfer integrals and Coulomb interactions are shown in Appendix A.

Based on the above first-principles calculation results, we constructed a 2D extended Hubbard model [41]:

$$H = \sum_{R,\delta, \alpha, \beta} \sum_{n, \alpha, \sigma} t^{(\delta)SOC}_{\alpha, \beta} \phi_{n R, \alpha, \sigma}^\dagger \phi_{n R + \delta, \beta, \sigma} + \xi \sum_{R, \alpha} n_{R \alpha, \uparrow} n_{R \alpha, \downarrow} + \frac{e}{2} \sum_{R, \delta, \alpha, \beta} \sum_{n, \alpha, \sigma} V^{(\delta)}_{\alpha, \beta} n_{R \alpha, \sigma} n_{R + \delta, \beta, \sigma}, \quad (1)$$

where $R$ is the coordinate of the unit cell and $\delta = (\delta_x, \delta_y)$ is the relative lattice vector in the $a$-$b$ plane. $\alpha$ and $\beta$ are the site indices, and $\sigma_1$ and $\sigma_2$ are the spin indices ($\uparrow, \downarrow$). $t^{(\delta)SOC}_{\alpha, \beta, \sigma_1, \sigma_2}$ is the transfer integral including SOC between $(\alpha, \sigma_1)$ and $(\beta, \sigma_2)$ separated by $\delta$. Site potentials, which is defined as the onsite components of transfer integrals $(t^{(0)SOC}_{n R, \alpha, \sigma})$, $t^{(0)SOC}_{B}$ and $t^{(0)SOC}_{C}$ are 4.470 eV, 4.465 eV, and 4.477 eV are excluded in our model because their contribution to the energy band is negligible. $c^{\dagger}_{R, \alpha, \sigma}(c_{R, \alpha, \sigma})$ is the creation (annihilation) operator for the $\alpha$ site with spin $\sigma_1$ in the unit cell located at $R$. $U_\alpha$ and $V^{(\delta)}_{\alpha, \beta}$ are the onsite and the inter-site Coulomb interactions, respectively, given by the static effective direct integrals $W^{(\delta)}_{\alpha, \beta}$ calculated using RESPACK (see Appendix A). Values of $W^{(\delta)}_{\alpha, \beta}$ are a little too large to be used in mean-field calculation, although it is a value that considers the screening effect. Therefore, in the following calculations, $W^{(\delta)}_{\alpha, \beta}$ is multiplied by a constant $\xi (0 < \xi < 1)$ and the value is controlled. We also defined the number operator as $n_{R, \alpha, \sigma} = c^{\dagger}_{R, \alpha, \sigma} c_{R, \alpha, \sigma}$. In the following, the lattice constants, Boltzmann constant $k_B$, and the Planck constant $h$ are considered to be unity. Furthermore, the electronvolt (eV) is used as the unit of energy throughout this paper, unless otherwise stated.

**B. Electronic state using Hartree-Fock approximation**

We treated Eq. (1) within the Hartree-Fock approximation in the wave number space. Fourier inverse transform, $c_{R, \alpha, \sigma} = N^{-1/2} \sum_k c_{k, \alpha, \sigma} e^{i k R}$, was performed on Eq. (1). Here, $N_{\text{cell}}$ is the total number of unit cells and $k = (k_x, k_y)$ indicates the wave-number vector. The Hartree-Fock Hamiltonian is as follows:

$$H_{\text{HF}} = H_T + H_U + H_V$$

$$H_T = \sum_{k, \alpha, \beta} \sum_{\sigma_1, \sigma_2} t^{(\delta)SOC}_{\alpha, \beta} \phi_{k \alpha, \sigma_1}^\dagger \phi_{k \beta, \sigma_2} + e^{i k R} \delta^{\dagger} \phi_{k \alpha, \sigma_1} \phi_{k \beta, \sigma_2}$$

$$H_U = \xi \sum_k \sum_{\sigma_1} U_{\sigma} \left[ \sum_{\sigma_2} \left( n_{\alpha, \sigma_1} \right) c_{k, \alpha, \sigma_1}^\dagger c_{k, \alpha, \sigma_1} \right]$$

$$- \frac{1}{N_{\text{cell}}} \sum_{k', q} \left[ c_{k' + q, \alpha, \sigma_1}^\dagger c_{k', \alpha, \sigma_1} c_{k, \alpha, \sigma_1} \right]$$

$$H_V = \xi \sum_k \sum_{\sigma_1, \sigma_2} \sum_{\delta} V^{(\delta)}_{\alpha, \beta} \left[ 2 \left( n_{\beta, \sigma_2} \right) c_{k, \alpha, \sigma_1}^\dagger c_{k, \alpha, \sigma_1} \right]$$

$$- \frac{1}{N_{\text{cell}}} \sum_{k', q} e^{-i q R} \left[ c_{k' + q, \alpha, \sigma_1}^\dagger c_{k', \alpha, \sigma_1} c_{k, \alpha, \sigma_1} \right]$$

$$+ \left( c_{k' + q, \beta, \sigma_2}^\dagger c_{k', \beta, \sigma_2} c_{k, \beta, \sigma_2} \right),$$

where $\sigma_1 = -\sigma_2$, and $\langle n_{\alpha, \sigma_1} \rangle = \langle c_{\alpha, \sigma_1}^\dagger c_{\alpha, \sigma_1} \rangle$ is the charge density of site $\alpha$ and spin $\sigma_1$. $\langle \delta^{\dagger} c \rangle$ in Eq. (2) is
an order parameter in the Hartree-Fock approximation. When the calculation is performed using Eq. (2), orders such as charge/spin density waves, bond order wave, and the interaction induced QSH order can happen with various periodicities. We set $q = 0$ and $k' = k$ to exclude long-periodic orders, which have not been experimentally observed. With these simplifications, the Fourier transform of the order parameter in the Hartree-Fock approximation is given by

$$\langle c^\dagger_{k,\alpha,1} c_{k,\beta,2} \rangle = \sum_\delta \langle c^\dagger_{0,\alpha,1} c_{\delta,\beta,2} \rangle e^{-ik\cdot\delta},$$

$$\langle c^\dagger_{0,\alpha,1} c_{\delta,\beta,2} \rangle = \frac{1}{N_{\text{cell}}} \sum_{k,\nu,\sigma} d_{\alpha,\nu,\sigma}(k) d^\ast_{\beta,\nu,\sigma}(k) \langle E_{\nu,\sigma}(k)/T \rangle.$$  

Thus, $H_{HF}$ can finally be expressed as follows:

$$H_{HF} = \sum_{k} \sum_{\alpha,\beta} H_{\alpha,\sigma_1;\beta,\sigma_2}(k) c^\dagger_{k,\alpha,1} c_{k,\beta,2}$$

$$H_{\alpha,\sigma_1;\beta,\sigma_2}(k) = \sum_\delta [\langle \delta \rangle_{\text{SOC}} f_{\alpha,\beta;1,2} e^{ik\cdot\delta},$$

$$-\xi V_{\alpha,\beta} \left( \sum_\delta \langle c^\dagger_{\delta,\beta,2} c_{1,\alpha,1} \rangle e^{-ik\cdot\delta} - \langle \hat{c}_{0,\alpha,1} c_{0,\alpha,1} \rangle \delta_{\sigma_1,1} \right)$$

$$+ \xi \delta_{\alpha,\beta} \left( n_{\alpha,\sigma_1} \delta_{\sigma_1,1} - \langle \hat{c}_{0,\alpha,1} c_{0,\alpha,1} \rangle \delta_{\sigma_1,1} \right)$$

$$+ \sum_{\gamma} \sum_{\sigma_3} \sum_\delta V_{\alpha,\gamma} \langle \hat{c}_{\gamma,\sigma_3} \rangle,$$

where the off-diagonal site component of the order parameter $\langle c^\dagger_{0,\alpha,1} c_{\delta,\beta,2} \rangle (\alpha \neq \beta)$, which is included as the Fock term in Eq. (3), modulates the transfer integrals. When the interaction induced QSH order occurs, the imaginary part of $\langle \hat{c}_{0,\alpha,1} c_{\delta,\beta,2} \rangle$ becomes finite and opens a gap in the energy band structure.

We numerically diagonalized $H_{\alpha,\sigma_1;\beta,\sigma_2}(k)$ using the eigenvector $d_{\alpha,\nu,\sigma}(k)$ and obtained the energy eigenvalues. For convenience, we define the eigenvalues $E_{\nu,\sigma}(k)$ as

$$E_{\nu,\sigma}(k) = \bar{E}_{\nu,\sigma}(k) - \mu,$$

$$\bar{E}_{\nu,\sigma}(k) = \sum_{\alpha,\beta} \sum_{\sigma_1,\sigma_2} d^\ast_{\nu,\sigma_1;\nu,\sigma_2}(k) H_{\alpha,\sigma_1;\beta,\sigma_2}(k) d_{\beta,\nu,\sigma_2}(k) - \mu,$$  

where $\mu$ is the chemical potential. $E_{\nu,\sigma}(k)$ are the eigenvalues with band index $\nu$ obtained by numerical diagonalization $\{\bar{E}_{1,\sigma}(k) > \bar{E}_{2,\sigma}(k) > \bar{E}_{3,\sigma}(k) > \bar{E}_{4,\sigma}(k)\}$. In the calculation, we ignored spin orders such as a spin ordered massive Dirac electron phase suggested in our preceding study because the spin order is not consistent with the results in a recent NMR experiment. Calculation results when spin orders are allowed as a stable solution are shown in Appendices B and C.

The Berry curvature $B_{\nu,\nu'}(k)$ and the Chern number for spin $\sigma$ $N_{\sigma}^{\text{Ch}}$ are defined as

$$B_{\nu,\nu'}(k) = \sum_{\nu' \neq \nu} \frac{\nu_{\nu',\nu}(k)\nu_{\nu',\nu}(k)}{|E_{\nu,\sigma}(k) - E_{\nu',\sigma}(k)|} + \text{c.c.},$$

$$N_{\sigma}^{\text{Ch}} = \frac{1}{2\pi} \int_{BZ} dkdB_{\nu,\sigma}(k),$$

where the Chern number $N_{\sigma}^{\text{Ch}}$ is calculated by $N_{\sigma}^{\text{Ch}} = \sum_{\nu} N_{\sigma}^{\text{Ch}}$. The velocity matrix $v_{\nu,\nu'}^{\eta}(k)$ along the $a$-axis ($\eta = a(y)$) and $b$-axis ($\eta = b(x)$) is given by

$$v_{\nu,\nu'}^{\eta}(k) = \sum_{\alpha,\beta} \sum_{\sigma_1,\sigma_2} d^\ast_{\alpha,\sigma_1;\nu,\sigma}(k)$$

$$\times \left[ \partial H_{\alpha,\sigma_1;\beta,\sigma_2}(k) / \partial k_{\eta} \right] d_{\beta,\sigma_2;\nu',\sigma}(k).$$

The spin Chern number $N_{\sigma}^{\text{Ch}} = \pm 1$ in the interaction induced QSH ordered insulator state $\{18, 19, 22, 23, 85\}$:

$$N_{\sigma}^{\text{Ch}} = \sum_{\sigma} \text{sgn}(\sigma) N_{\sigma}^{\text{Ch}}.$$

The Onsager phase factor $\gamma$ and the Berry phase $\phi_B$ were obtained based on the semiclassical theory $\{69, 73\}$:

$$\gamma \equiv 1 - \frac{\phi_B}{2\pi},$$

$$\phi_B = \int_{S_F} B_{\nu,\sigma}(k) dS,$$

where $S_F$ is the Fermi surface and $\int_{S_F}$ indicates surface integration on $S_F$. In the $\gamma$ calculation, the energy band which is shifted $0.005$ eV considering electron-doping is used to reproduce the carrier doping performed in the recent experiment. $\{60\}$.

C. Conductivity

The DC conductivity was calculated using the Nakano-Kubo formula $\{13, 74\}$. The longitudinal DC conductivity along $\eta$-axis ($\eta = b(x), a(y)$) direction is expressed by

$$\sigma_{\eta} = \int d\omega \left( \frac{df}{d\omega} \right) \Phi_{\eta}(\omega),$$

$$\Phi_{\eta}(\omega) = \frac{2e^2}{\pi N_{\text{cell}}} \sum_k \left[ \text{Tr} \left[ \hat{v}_{\eta}^\dagger(k) \hat{G}^R(k,\omega) \hat{v}_{\eta}^\dagger(k) \hat{G}^R(k,\omega) \right] \right],$$

where $\hat{G}^R(k,\omega)$ is the retarded Green’s function, which is obtained by the analytic continuation of the Matsubara frequency $i\varepsilon_n (\varepsilon_n = (2n + 1)\pi T)$ to a real frequency $\omega$. We treated the effect of elastic scattering between electrons and the impurities originating from the lack and
disorder of $\text{I}^-$ molecules, using $T$-matrix approximation. The impurity potential term $H_{\text{imp}}$ is defined by

$$H_{\text{imp}} = \frac{V_0}{N_{\text{cell}}} \sum_{\mathbf{k}, \mathbf{q}, \alpha, \sigma} \sum_{i} N_{\text{imp}} e^{-i \mathbf{q} \cdot \mathbf{r}_i} c_{\mathbf{k}+\mathbf{q}, \alpha, \sigma}^\dagger c_{\mathbf{k}, \alpha, \sigma},$$  \hspace{1cm} (13)

and we treated it using the perturbation theory for Green's function. Here, $V_0$ is the strength of the potential and $\mathbf{r}_i$ $(i = 1, \cdots, N_{\text{imp}})$ is the coordinate of the $i$-th impurity. We treated $H_{\text{imp}}$ within $T$-matrix approximation, assuming that the impurity density $c_{\text{imp}} = N_{\text{imp}}/N_{\text{cell}}$ is quite small ($c_{\text{imp}} \ll 1$) and impurities are uniformly distributed. We introduced the one body Green’s function in the Hartree-Fock approximation calculated using $H_{\text{HF}}$ (Eq. (4)) as

$$C_{\alpha, \sigma_1; \beta, \sigma_2}(\varepsilon_n, \mathbf{k}) = \sum_{\nu, \sigma} \frac{d_{\alpha, \sigma_1, \nu, \sigma}(\mathbf{k}) d_{\beta, \sigma_2, \nu, \sigma}(\mathbf{k})}{\varepsilon_n - E_{\nu, \sigma}(\mathbf{k})},$$  \hspace{1cm} (14)

The retarded self-energy in $T$-matrix approximation $\Sigma_{\nu, \nu'}(\mathbf{k}, \omega)$ is calculated using the perturbation theory for Green’s function. When the real part of $\Sigma^R_{\nu, \nu'}(\mathbf{k}, \omega)$ and inter-band components of $\Sigma^R_{\nu, \nu'}(\mathbf{k}, \omega)$ $(\nu \neq \nu')$ can be ignored, the damping constant $\gamma_{\nu, \sigma}(\mathbf{k}, \omega)$ in the $T$-matrix approximation is obtained as follows:

$$\gamma_{\nu, \sigma}(\mathbf{k}, \omega) = \frac{\hbar}{2 \tau_{\nu, \sigma}(\mathbf{k}, \omega)} = -\text{Im} \Sigma^R_{\nu, \sigma}(\mathbf{k}, \omega)$$

$$= c_{\text{imp}} \left| d_{\nu, \nu, \sigma}(\mathbf{k}) \right|^2 \left\{ \pi V_0^2 \rho_\sigma^E(\omega) \right\} - \frac{1}{1 + \left\{ \pi V_0^2 \rho_\sigma^E(\omega) \right\}^2},$$  \hspace{1cm} (15)

where $c_{\text{imp}} = N_{\text{imp}}/N_{\text{cell}} = 0.02 \ll 1$ is the impurity density and $\tau_{\nu, \sigma}(\mathbf{k}, \omega)$ is the relaxation time in $T$-matrix approximation. $\rho_\sigma^E(\omega)$ represents the density of states for $\sigma$:

$$\rho_\sigma^E(\omega) = \frac{1}{N_{\text{cell}}} \sum_{\mathbf{k}, \alpha, \sigma} \left| d_{\alpha, \sigma, \nu, \sigma}(\mathbf{k}) \right|^2 \delta(\hbar \omega - E_{\nu, \sigma}(\mathbf{k})).$$  \hspace{1cm} (16)

We ignored the real part of $\Sigma^R_{\nu, \nu'}(\mathbf{k}, \omega)$ because it only gives the constant shift of energy in the limit of $c_{\text{imp}} \ll 1$ \cite{77}. In this case, $\Phi_\nu(\omega)$ in Eq. (12) can be represented as follows:

$$\Phi_\nu(\omega) = \frac{2 e^2}{N_{\text{cell}}} \sum_{\mathbf{k}, \nu, \sigma} \left| V_{\nu, \sigma}(\mathbf{k}) \right|^2 \tau_{\nu, \sigma}(\mathbf{k}, \omega) \delta(\hbar \omega - E_{\nu, \sigma}(\mathbf{k})).$$

\hspace{1cm} (17)

In this study, the DC conductivity is normalized to the universal conductivity $\sigma_0 = 4e^2/\pi \hbar$.

### D. Calculation of the spin fluctuations using RPA

We investigated the effect of spin fluctuations on the NMR properties, e.g., the Knight shift and $1/T_1 T$ in the high-$T$ Dirac electron phase in RPA using eq. (4) \cite{77} \cite{49} \cite{61}. In the linear response theory, the irreducible susceptibility $\chi_{0, \alpha, \beta}(\mathbf{q}, \omega_m)$ can be calculated using the one body Green’s function $\tilde{G}^0$ (Eq. (14)) as follows:

$$\chi_{0, \alpha, \beta}(\mathbf{q}, \omega_m)$$

$$= -\frac{T}{N_{\text{cell}}} \sum_{\mathbf{k}, \mathbf{n}} \chi_{0, \alpha, \beta, 1}(\mathbf{k} + \mathbf{q}, \varepsilon_n + \omega_m) G_{0, \beta, 2}(\mathbf{k}, \varepsilon_n)$$

$$= \frac{1}{N_{\text{cell}}} \sum_{\mathbf{k}, \sigma, \sigma'} \sum_{\nu, \nu'} F_{\alpha, \sigma_1; \beta, \sigma_2}(\mathbf{k}, \mathbf{q}) \chi_{0, \nu, \sigma_1; \sigma_2}(\mathbf{q}, \omega_m),$$

\hspace{1cm} (18)

$$\chi_{0, \nu, \sigma_1; \sigma_2}(\mathbf{q}, \omega_m) = -\frac{f(E_{\nu, \sigma}(\mathbf{k} + \mathbf{q})) - f(E_{\nu, \sigma'}(\mathbf{k}))}{E_{\nu, \sigma}(\mathbf{k} + \mathbf{q}) - E_{\nu, \sigma'}(\mathbf{k}) + i\omega_m},$$  \hspace{1cm} (19)

where $N_{\text{cell}}$ is the system size and $\omega_m = 2m\pi T$. $f(E_{\nu, \sigma}(\mathbf{k})) = [1 + \exp(E_{\nu, \sigma}(\mathbf{k})/T)]^{-1}$ represents the Fermi distribution function. $F(\mathbf{k}, \mathbf{q})$ indicates the form factor represented by

$$F_{\alpha, \sigma_1; \beta, \sigma_2}(\mathbf{k}, \mathbf{q}) = d_{\alpha, \sigma_1, \nu, \sigma}(\mathbf{k} + \mathbf{q}) d_{\beta, \sigma_2, \nu, \sigma}(\mathbf{k} + \mathbf{q})$$

$$\times d_{\beta, \sigma_2, \nu, \sigma}(\mathbf{k}) d_{\alpha, \sigma_1, \nu, \sigma}(\mathbf{k}).$$  \hspace{1cm} (20)

In RPA, the spin susceptibility $\hat{\chi}^S$ and the transverse spin susceptibility $\hat{\chi}^S_{\perp}$ in the absence of an external field and in presence of spin symmetry are calculated as follows:

$$\hat{\chi}^S(\mathbf{q}, \omega) = \chi^S(\mathbf{q}, \omega) = \left( \mathbf{I} - \chi^0(\mathbf{q}, \omega) \right)^{-1} \chi^0(\mathbf{q}, \omega),$$  \hspace{1cm} (21)

where $\mathbf{I}$ is the unit matrix and $\hat{\chi}^0(\mathbf{q}, \omega)$ represents the estimation of the intra- and inter-band components to the spin fluctuations, we divided the irreducible susceptibility into two components \cite{61}:

$$\chi_{0, \text{Intra}}^{\text{Intra}}(\mathbf{q}, \omega) = \frac{1}{N_{\text{cell}}} \sum_{\mathbf{k}} \sum_{\sigma, \sigma'} \sum_{\nu, \nu'} F_{\alpha, \sigma_1; \beta, \sigma_2}(\mathbf{k}, \mathbf{q})$$

$$\times \chi_{0, \nu, \sigma_1; \sigma_2}(\mathbf{q}, \omega),$$

\hspace{1cm} (22)

$$\chi_{0, \text{Inter}}^{\text{Inter}}(\mathbf{q}, \omega) = \frac{1}{N_{\text{cell}}} \sum_{\mathbf{k}} \sum_{\sigma, \sigma'} \sum_{\nu, \nu'} F_{\alpha, \sigma_1; \beta, \sigma_2}(\mathbf{k}, \mathbf{q})$$

$$\times \chi_{0, \nu, \sigma_1; \sigma_2}(\mathbf{q}, \omega),$$

\hspace{1cm} (23)

where $\chi_{0, \text{Intra}}^{\text{Intra}}$ and $\chi_{0, \text{Inter}}^{\text{Inter}}$ are the intra- and inter-band components of the irreducible susceptibility $\chi^0$. Thus, the intra-band component of the spin susceptibility is calculated as follows:

$$\hat{\chi}^S_{\text{Intra}} = \left( \mathbf{I} - \chi_{0, \text{Intra}}^{\text{Intra}} \right)^{-1} \chi_{0, \text{Intra}}^{\text{Intra}}.$$  \hspace{1cm} (24)

The inter-band component $\hat{\chi}^S_{\text{Inter}}$ is also obtained based on the definition $\hat{\chi}^S_{\text{Inter}} = \hat{\chi}^S - \hat{\chi}^S_{\text{Intra}}$. Applying the analytical continuation $i\omega_m \to \hbar \omega + i0^+$, the site-resolved
Knight shift $K_\alpha$ and $1/T_1T$ in RPA are obtained as follows:

$$K_\alpha = \sum_\beta \text{Re} \left[ \chi_{\alpha,\beta}^S(q = 0, \omega = 0) \right],$$  \hspace{1cm} (25)

and

$$1/T_1T = \sum_q \sum_\alpha \frac{\text{Im} \left[ \chi_{\alpha,\alpha}^\pm(q, \omega_0) \right]}{\omega_0},$$  \hspace{1cm} (26)

where the frequency $\omega_0$ is infinitely close to zero and is set as $\omega_0 = 0.001$ in this study.

III. NUMERICAL RESULTS

A. Electronic state at low-$T$ when a static effective direct integral is used

In this subsection, to investigate more realistic orders which are possible caused in $\alpha$-(BETS)$_2$I$_3$ at low-$T$ region, the numerical results using the transfer integrals with SOC $t_{\alpha,\beta,\sigma_1,\sigma_2}^{(d)\text{SOC}}$ and the static effective direct integral $W_{\alpha,\beta}^{(d)}$ obtained based on the first-principles calculation are shown. We set the initial states of the mean-field calculation randomly and investigated the electronic state with the lowest energy, other than the charge, spin, and bond orders. Throughout this subsection, $T = 1 \times 10^{-4}$ and we sweep the control parameter $\xi$ which is multiplied to $W_{\alpha,\beta}^{(d)}$ from 0 (non-interacting case) to 1 to investigate the effect of the interaction to the electronic state. As a result, we found that the interaction induced QSH order proposed in previous studies for the honeycomb lattice model $[22, 23, 25, 26]$ is the most stable order at low-$T$ region in $\alpha$-(BETS)$_2$I$_3$.

Figure 3(a) shows the $\xi$-dependence of the energy gap $\Delta$ at the Dirac point $\Delta$. In $\xi = 0$ (non-interacting case), the electronic system becomes a topological insulator (TI) and a small energy gap $\Delta \simeq 0.002$ opens owing to the contribution of only the SOC term in the transfer integrals $t_{\alpha,\beta,\sigma_1,\sigma_2}^{(d)\text{SOC}}$. With an increase in $\xi$, $\Delta$ increases continuously by the contribution of the interaction $W_{\alpha,\beta}^{(d)}$. In Fig. 3(b), the $\xi$-dependence of the imaginary part of the order parameters $\text{Im}[\langle c_\sigma^\dagger c_\sigma \rangle_{b1}]$ and $\text{Im}[\langle c_\sigma^\dagger c_\sigma \rangle_{b4}]$ plotted. $\text{Im}[\langle c_{\uparrow}^\dagger c_{\uparrow} \rangle_{b1}]$ (Im[\langle c_{\downarrow}^\dagger c_{\downarrow} \rangle_{b4}]) contributes to the phase modulation of the next-nearest-neighbor (nearest-neighbor) component of the transfer integral $t_{b1,\sigma}^{(d)}$ ($t_{b4,\sigma}$) illustrated in Fig. 2. The signs of them are inverted according to the degrees-of-freedom of the spin $\sigma$ in the interaction induced QSH ordered insulator state, as plotted in Fig. 3(b). In $\xi > 0$, the imaginary part of the order parameter such as $\text{Im}[\langle c_{\uparrow}^\dagger c_{\uparrow} \rangle_{b1}]$ and $\text{Im}[\langle c_{\downarrow}^\dagger c_{\downarrow} \rangle_{b4}]$ which is the same as the interaction induced QSH order proposed in previous studies for the honeycomb lattice model $[22, 23, 24, 26]$ becomes finite and is enhanced with the increase in $\xi$. On the other hand, in the TI state ($\xi = 0$), $\text{Im}[\langle c_{\uparrow}^\dagger c_{\uparrow} \rangle_{b1}]$ and $\text{Im}[\langle c_{\downarrow}^\dagger c_{\downarrow} \rangle_{b4}]$ are zero.

![FIG. 3. (Color online) $\xi$-dependence of the (a) energy gap $\Delta$, (b) the imaginary part of order parameters which modulate transfer integrals $t_{b1,\sigma}$ and $t_{b4,\sigma}$: $\text{Im}[\langle c_{\uparrow}^\dagger c_{\uparrow} \rangle_{b1}]$ and $\text{Im}[\langle c_{\downarrow}^\dagger c_{\downarrow} \rangle_{b4}]$, respectively.](image_url)

Figures 4(a) and (b) show the energy eigenvalues $E_{\nu,\sigma}^{+,-}\nu(k)$ near the Fermi energy ($\nu = 1, 2$) in the TI ($\xi = 0$) and the interaction induced QSH ordered ($\xi = 0.5$) states, respectively. The energy gap in the TI state is approximately 2 meV as shown in the inset of Fig. 4(a). The Berry curvature $B_{\nu,\sigma}(k)$ in the interaction induced QSH ordered insulator state ($\xi = 0.5$) is plotted in Figure 4(c) for each spin. $B_{\nu,\sigma}(k)$ has two peaks originating from two massive Dirac cones in the Brillouin zone. The spin Chern number $N_{\text{Ch}}$ (see Eq. (9)) becomes $-1$ in the interaction induced QSH ordered insulator state because the two peaks of $B_{\nu,\sigma}(k)$ have the same sign, which invert according to the spin index $\sigma$. These wavenumber and spin dependencies on $B_{\nu,\sigma}(k)$ are the approximately the same as those in the interaction induced QSH order of the honeycomb lattice model $[22, 23]$ and TI state of $\alpha$-(BETS)$_2$I$_3$ $[25]$. Therefore, the spin Chern number $N_{\text{Ch}}$ defined in Eq. (9) becomes $\pm 1$ in both the TI and interaction induced QSH ordered insulator states. In other words, the edge spin current in both states is quantized by the spin Chern number $N_{\text{Ch}}$ owing to the bulk-edge correspondence. The difference between the TI state $[39, 40, 55]$ and the interaction induced QSH ordered insulator state $[22, 23]$ is the energy gap values and order parameters: The TI state has a slight gap of approximately 2 meV owing to the SOC contribution alone $[39, 40, 55]$ and is not an ordered state because any corresponding order parameter, $\langle c_{\downarrow} \rangle$, does not exist. Conversely, in the interaction induced QSH ordered insulator state, the energy gap value depends on the magnitude of the imaginary part of the
order parameter $\text{Im}[\langle c^\dagger c \rangle]$, as shown in Figs. 3 (a) and (b). The TI state is a band insulator caused by the contribution of intrinsic SOC alone, whereas the interaction induced QSH ordered insulator state is an ordered state derived from spontaneous symmetry breaking caused by the contribution of the Coulomb interaction.

**B. Stability of the interaction induced QSH ordered insulator state in the presence of the Coulomb interaction**

Next, to investigate the relationship between the stability of the interaction induced QSH order at low-$T$ in $\alpha$-(BETS)$_2$I$_3$ and the values of the nearest-neighbor and next-nearest-neighbor interactions, we represent the nearest-neighbor and next-nearest-neighbor Coulomb interactions by parameters $V$ and $V'$, respectively, and draw the phase diagram for these parameters. We also calculated the phase modulation caused by the order parameter of the interaction induced QSH order

$$\varphi = \tan^{-1} \frac{\text{Im}[\langle c^\dagger c \rangle]}{\text{Re}[\langle c^\dagger c \rangle]}$$

for several closed loops in the unit cell and investigate the effect of a local magnetic flux owing to the interaction induced QSH order. In this section, we show the calculation results for the following two cases to investigate the effect of the SOC term in transfer integrals: (1) when transfer integrals with SOC, $t^{(\delta)\text{SOC}}_{\alpha,\beta,\sigma}$, are used, and (2) when transfer integrals without SOC, $t^{(\delta)}_{\alpha,\beta,\sigma}$, are used (see Table I in Appendix A). Throughout this subsection, the onsite Coulomb interaction $U$ and $T$ are fixed at $(U, T) = (0.5, 1 \times 10^{-4}$), unless otherwise stated.

1. $V$-$V'$ phase diagram when SOC exists

We first draw the $V$-$V'$ phase diagram with SOC. The calculation result is shown in Fig. 5 (a). When SOC is considered, the interaction induced QSH order is stabilized at $V' > 0$ and $V < 0.12$. This indicates that $V'$ plays a significant role in stabilizing the interaction induced QSH ordered insulator state in $\alpha$-(BETS)$_2$I$_3$, as already suggested in studies for the honeycomb lattice model [22, 23]. As indicated in the list of static effective direct integrals evaluated by RESPACK [84] in Table I, $V'/V$ is expected to be large in $\alpha$-(BETS)$_2$I$_3$. This tendency favors the realization of the interaction induced QSH ordered insulator state. The horizontal stripe charge ordered insulator state [41] appears in $V > 0.12$ and $V' < V^{\text{CO}}$ (upper-left region). However, the charge ordered insulator state is not realistic in $\alpha$-(BETS)$_2$I$_3$ because inversion symmetry breaking has not been observed by the synchrotron X-ray diffraction experiment [38]. The TI state caused only by the SOC contribution appears at $V' = 0$ and $V < 0.12$.

Next, we set $V = 0.1$ and calculate the amount of phase modulation caused by the order parameter $\varphi$ defined as Eq. (27) in the unit cell and investigated the presence or absence of a local magnetic flux. Figures 5(b) and (c) show the schematic of the unit cell of $\alpha$-(BETS)$_2$I$_3$ and the loop patterns to calculate the summation of $\varphi$. These loops include only the nearest-neighbor and next-nearest-neighbor bonds. The $V'$-dependence of the summation of phases in each loop of the unit cell $\varphi_n$ ($n = 1, \cdots, 10$) at $V = 0.1$ are plotted in Figs. 5(d) and (e). Signs of $\varphi_n$ depend on the spin degrees-of-freedom $\sigma$ in the interaction induced QSH ordered insulator state. The sum of $\varphi_n$ in the unit cell becomes zero because cancellation occurs: $\varphi_1 = -\varphi_4$, $\varphi_2 = -\varphi_5$, $\varphi_3 = -\varphi_6$ (loops

FIG. 4. (Color online) The energy band structure $E_{\nu,\sigma}(k)$ in the (a) TI ($\xi = 0$) and (b) interaction induced QSH ordered ($\xi = 0.5$) states near the Fermi energy ($\nu = 1, 2$) for $\sigma = \uparrow$. $E_{\nu,\uparrow}(k)$ and $E_{\nu,\downarrow}(k)$ are degenerated. The inset in (a) is the magnified view of $E_{\nu,\sigma}(k)$ in the TI state in the $-0.02 < E_{\nu,\sigma}(k) < 0.02$ energy range. (c) The Berry curvature $B_{\nu,\sigma}(k)$ in the interaction induced QSH ordered insulator state for $\nu = 1$. 

FIG. 3. (Color online) A comparison of the energy bands of $\alpha$-(BETS)$_2$I$_3$ for (a) $V = 0$ and (b) $V = 0.1$. The energy bands for the TI state are drawn with black, whereas the energy bands for the interaction induced QSH ordered insulator state are drawn with red. The differences between the two states are shown in (a) $V = 0$ and (b) $V = 0.1$. The inset shows the magnified view of the energy bands near the Fermi energy. The energy bands for the TI state are drawn with black, whereas the energy bands for the interaction induced QSH ordered insulator state are drawn with red. The differences between the two states are shown in (a) $V = 0$ and (b) $V = 0.1$. The inset shows the magnified view of the energy bands near the Fermi energy. The energy bands for the TI state are drawn with black, whereas the energy bands for the interaction induced QSH ordered insulator state are drawn with red. The differences between the two states are shown in (a) $V = 0$ and (b) $V = 0.1$. The inset shows the magnified view of the energy bands near the Fermi energy.
including only nearest-neighbor bonds) and \( \varphi_7 = -\varphi_{10} \), \( \varphi_8 = -\varphi_9 \) (loops including nearest-neighbor and next-nearest-neighbor bonds). Therefore, no total magnetic flux in the unit cell exists in the interaction induced QSH ordered insulator state. \( \varphi_n \) is zero at only \( V' = 0 \) and continuously increased as \( V' \) is increased.

2. \( V-V' \) phase diagram when SOC is absence

Next, we used transfer integrals without the SOC term, \( t_{\alpha,\beta,\sigma}^{(\delta)} \), in the calculation and draw the \( V-V' \) phase diagram without SOC. The calculation result is shown in
ordered insulator states can appear at \( V' > V'^{\text{IC}} \) and \( V' > V'^{\text{CO}} \) and these states are degenerated. In the interaction induced QAH ordered insulator phase, the sign of the order parameter \( \langle c^\dagger c \rangle \) does not depend on the degrees-of-freedom of the spin \( \sigma \) and the Chern number \( N_{\text{Ch}}^{\nu,\sigma} = \sum_{\nu} N_{\text{Ch}}^{\nu} \) defined by Eq. 6 becomes \( \pm 1 \). With the decrease in \( V \) and \( V' \), the massless Dirac electron phase appears owing to phase transition at \( V' < V'^{\text{CO}} \) and \( V' < V'^{\text{IC}} \).

The \( V' \)-dependence of the summation of phases on several loops as shown Figs. 6 (b) and (c), \( \varphi_n \) \((n = 1, \ldots, 10)\), at \( V = 0.1 \) are plotted in Figs. 6(b) and (c). The sum of \( \varphi_n \) in the unit cell cancelled each other out: \( \varphi_1 = -\varphi_4, \varphi_2 = -\varphi_5, \varphi_3 = -\varphi_6 \) (loops including only the nearest-neighbor bonds) and \( \varphi_7 = -\varphi_{10}, \varphi_8 = -\varphi_9 \) (loops including the nearest-neighbor and next-nearest-neighbor bonds). \( \varphi_n \) vanishes suddenly at \( V' = V'^{\text{IC}} \) because the QAH order to the massless Dirac electron phase transition occurs.

It can be considered that the contribution of SOC cannot be ignored in real material, so the QAH to massless Dirac electron phase transition does not occur in \( \alpha-(\text{BETS})_2\text{I}_3 \). The difference of the interaction induced QSH order with SOC and QAH order without SOC in our results is analogous to a ferromagnet with and without an external magnetic field. When SOC is considered, as presented in the previous subsection, the interaction induced QSH ordered insulator state at \( V' > 0 \) continuously changes to the TI state at \( V' = 0 \) with a decrease in \( V' \).

### C. Switching of Onsager phase factor by contribution of the interaction

In this subsection, we calculated the Onsager phase factor \( \gamma \) under the interaction induced QSH ordered insulator state using Eq. 10 and discuss the consistency with experimental results [60]. It has been reported by Shubnikov-de Haas oscillation measurements that the Onsager phase factor \( \gamma \) clearly switches 1/2 to zero at the hydrostatic pressure \( P = 0.5 \)GPa at which the insulator state of \( \alpha-(\text{BETS})_2\text{I}_3 \) vanishes in the entire T-region [60]. This result suggests that electron correlation effects cannot be negligible in \( \alpha-(\text{BETS})_2\text{I}_3 \) similar to \( \alpha-(\text{ET})_2\text{I}_3 \) under hydrostatic pressure [44–54]. In this subsection, we considered the \( P \)-dependence as the change in \( V' \) associated with the change in \( P \) and calculate \( V' \)-dependence of \( \gamma \) at low-\( T \) to explain the experimental results. Throughout this subsection, \( T = 1 \times 10^{-4}, U = 0.5, \) and \( V = 0.1 \) to investigate the contribution of the interaction induced QSH order to \( \gamma \).

The \( V' \)-dependence of the absolute value of the Onsager phase factor \( |\gamma| \) at \( (U, V, T) = (0.5, 0.1, 1 \times 10^{-4}) \) is plotted in Fig. 7. The value of \( |\gamma| \) becomes zero as \( V' \) decreases, and increases to 0.5 when \( V' \) is sufficiently large.

![FIG. 7. (Color online) (a) \( V' \)-dependence of the absolute value of the Onsager phase factor \( \gamma \) at \( (U, V, T) = (0.5, 0.1, 1 \times 10^{-4}) \). (b) and (c) Illustration of the Berry curvature \( B_{\nu,\sigma}(k) \) and the energy band \( E_{\nu,\sigma}(k) \) near the Dirac point for two \( \Delta \) values: \( \Delta \ll |E_F| \) and \( \Delta \gg |E_F| \).](image-url)
state, the peak of $B_{\alpha\sigma}^z(k)$ decreases with the increase in the energy gap $\Delta$, and $B_{\alpha\sigma}^z(k)$ widens and spreads in the Brillouin zone as shown in Fig. 4 (b). Therefore, when the spread of $B_{\alpha\sigma}^z(k)$ becomes sufficiently larger than the integral range $S_F$, the peak of $\sigma^z_{\alpha\sigma}$ is lowered and becomes zero ($|\gamma|$ becomes 0.5).

**D. Temperature dependence of DC resistivity under ambient pressure**

Next, we calculated the $T$-dependence of the electronic state with the Hartree-Fock approximation and investigate temperature $T$ effects for the interaction induced QSH order. It has been reported that the DC resistivity of $\alpha$-(BETS)$_2$I$_3$ obtained experimentally [59–61] was nearly constant at $T > 50 \times 10^{-4}$ and sharply increases below $T = 50 \times 10^{-4}$. This experimental fact suggests that the electronic state changes around $T = 50 \times 10^{-4}$ and a large band gap opens below this temperature. To demonstrate the experimental results described above, we sweep $T$ as a parameter and investigate the $T$-dependence on DC conductivity using $T$-matrix approximation. Throughout this subsection, $U = 0.5$, $V = 0.1$, and $V' = 0.16$.

Figure 8 (a) shows the $T$-dependence of the energy gap $\Delta$ at $(U, V, V') = (0.5, 0.1, 0.16)$ with and without SOC. When SOC is absent, the QAH ordered phase in which the order parameter $\text{Im}[\langle c^\dagger c \rangle]$ and the Chern number $N^{Ch}$ becomes $\pm 1$, and $\Delta$ without SOC increases sharply below the critical temperature $T_C$ owing to the phase transition between the massless Dirac electron and QAH ordered phases. Conversely, when SOC is considered, $\Delta \sim 0.01$ at $T = 0.03$, which is approximately five times that of the case without the Coulomb interaction. On decreasing $T$, $\Delta$ with SOC gradually increases toward low-$T$ owing to the next-nearest-neighbor interaction induced QSH order and has a constant value $\Delta > 0.038$, which is twice that of the case without SOC. This result indicates that the contribution of SOC and $V'$ renders the interaction induced QSH ordered insulator state more stable. In Figs. 8 (b), the $T$-dependence of the DC resistivity along the $a$-axis direction $\rho_a(T)/\rho_0 \equiv \sigma^z_{\alpha\sigma} = (4e^2/\pi h)^{-1}$ is plotted for two cases with and without SOC. $\rho_a(T)/\rho_0$ without SOC is nearly constant at $T > T_C$ and sharply increases at $T < T_C$. Alternatively, $\rho_a(T)/\rho_0$ with SOC increases continuously near $T = T_C$ with the decrease in $T$ reflecting the gentle $T$-dependence on the energy gap $\Delta$. Therefore, the sharp increase in DC resistivity below $T = 50 \times 10^{-4}$ observed in experiments [59–61] can be explained by considering the interaction induced QSH order, and it is difficult to explain if such an order does not exist. As mentioned in section III.B, the difference with and without SOC is analogous to a ferromagnet with and without an external magnetic field.

To summarize this subsection, we presented the $T$-dependence of the electronic state obtained with the Hartree-Fock approximation (Fig. 9). In this study, the interaction induced QSH ordered insulator state stably appears in the entire $T$-region. As $T$ is decreased, the order parameter of the interaction induced QSH order which is primarily caused by the nearest- and next-nearest-neighbor interaction and SOC increases and the energy gap is gradually enhanced. The interaction induced QSH ordered insulator state is consistent with experimental results such as X-ray diffraction, DC resistivity, and Shubnikov-de Haas oscillation [59–61] and is a strong candidate for the insulating state on $\alpha$-(BETS)$_2$I$_3$ at low-$T$. 

**FIG. 8.** (Color online) $T$-dependence of (a) the energy gap $\Delta$ at $(U, V, V') = (0.5, 0.1, 0.16)$, and (b) the DC resistivity along the $a$-axis direction $\rho_a(T)/\rho_0$ in units of the universal conductivity reciprocal $\rho_0 \equiv \sigma^z_{\alpha\sigma} = (4e^2/\pi h)^{-1}$ at $(U, V, V') = (0.5, 0.1, 0.16)$.

**FIG. 9.** (Color online) Illustration of the $T$-dependence of the electronic state obtained with the Hartree-Fock approximation, with onsite $U$, nearest-neighbor sites $V$, and next-nearest-neighbor sites $V'$. 

Large gap opens by interaction
QSH ordered insulator state
SOC gap $\sim 2$meV

Low- $T$

High-$T$
E. Spin fluctuations in the high-$T$ Dirac electron phase

In this subsection, to investigate the effects of the Coulomb interaction on spin fluctuations, we calculated the spin susceptibility using RPA and discuss the relationship with NMR experiments of $\alpha$-(BETS)$_2$I$_3$ [62, 63, 78]. Previous studies on $\alpha$-(ET)$_2$I$_3$ [49, 51] have shown that the ferrimagnetic (FM) spin polarization observed in the site-resolved Knight shift [50] is induced by $U$, where only the site-resolved Knight shift at B site $K_B$, defined in Eq. (25), becomes negative with the increase in $U$, and the other components always remain positive. It has been reported that this behavior is caused by the interband electron-hole excitation enhanced by $U$ [49, 51]. In this subsection, we also investigated the possibility of FM spin polarization in $\alpha$-(BETS)$_2$I$_3$ [78].

Figures 10(a)-(c) show the $T$-dependence of the site-resolved Knight shift $K_\alpha \equiv \sum_\beta \text{Re} \left[ \chi_{\alpha,\beta}^S (0,0) \right]$ at $U = 0.13$ (thick line) and $U = 0$ (thin line) for (a) $\alpha = A$, (b) $\alpha = C$, and (c) $\alpha = B$. The inset in (c) is the $T$-dependence of $K_B$ divided into two components: intra-band (dotted line) and inter-band (one-dot chain line).

![Figure 10](image1)

**FIG. 10.** (Color online) $T$-dependence of the site-resolved Knight shift $K_\alpha \equiv \sum_\beta \text{Re} \left[ \chi_{\alpha,\beta}^S (0,0) \right]$ at $U = 0.13$ (thick line) and $U = 0$ (thin line) for (a) $\alpha = A$, (b) $\alpha = C$, and (c) $\alpha = B$. The inset in (c) is the $T$-dependence of $K_B$ divided into two components: intra-band (dotted line) and inter-band (one-dot chain line).

Resuming the results of the previous subsections, we also investigated the possibility of FM spin polarization at $\alpha = B$. The inset in Figs. 11(a) and (b), $K_A$ and $K_C$ are enhanced when $U$ is considered, and become zero with the decrease in $T$ owing to the cancellation of each component of $\text{Re} \left[ \chi_{A,B}^S \right]$ and $\text{Re} \left[ \chi_{C,B}^S \right]$ (B = A, A’, B, and C). In contrast, $K_B$ decreases and becomes negative below $T \sim 0.0075$ as shown in Fig. 11(c). The $T$-dependencies of $\text{Re} \left[ \chi_{A,A'}^{S,\text{intra}} \right]$, $\text{Re} \left[ \chi_{A,A'}^{S,\text{inter}} \right]$, and $\text{Re} \left[ \chi_{B,B}^S \right]$ are also plotted in the inset of Fig. 11(c). It is indicated that $\text{Re} \left[ \chi_{B,B}^{S,\text{inter}} \right]$ becomes negative for $0 < T < 0.01$, and causes $\text{Re} \left[ \chi_{B,B}^S \right]$ to become negative. This behavior is qualitatively similar to that observed in $\alpha$-(ET)$_2$I$_3$ [60]. This is because $\alpha$-(BETS)$_2$I$_3$ at high-$T$ has a characteristic wavenumber dependence on the square of the absolute value of the eigenvector such as a zero line for B and C sites, which is similar to that of $\alpha$-(ET)$_2$I$_3$ under high pressure.

Next, we calculated spin susceptibility considering stronger interaction and investigated the type of spin susceptibility that was enhanced. Figures 11(a) and (b) show the momentum $q$-dependence of the spin suscepti-
FIG. 12. (Color online) $T$-dependence of $1/T_1T$ at $U = 0$, $U = 0.13$, and $U = 0.46$.

ability $\text{Re} \left[ \chi_{\alpha,\beta}^S \right]$ at $\omega = 0$ for $\beta = A, A'$ with $(U, T) = (0.46, 0.007)$. In the strong interaction case, $\text{Re} \left[ \chi_{AA}^S \right]$ and $\text{Re} \left[ \chi_{AA'}^S \right]$ exhibit a peak at $q = 0$ reflecting the Fermi point in the Dirac electron system. $\text{Re} \left[ \chi_{AA}^S \right]$ and $\text{Re} \left[ \chi_{AA'}^S \right]$ at $q = 0$ have approximately the same absolute values with opposite signs. Here, a negative peak in the site off-diagonal component $\text{Re} \left[ \chi_{AA}(0, \omega = 0) \right]$ indicates that there is spin fluctuation that aligns the spins between the A and A’ sites in the unit cell to the opposite directions. Furthermore, we plotted the $T$-dependence of $\text{Re} \left[ \chi_{AA}(q = 0, \omega = 0) \right]$ and $\text{Re} \left[ \chi_{AA'}(q = 0, \omega = 0) \right]$ (Fig. 11(c)). With the decrease in $T$, they are enhanced, and diverge positively and negatively toward $T \sim 0.006$. This result indicates that the spin fluctuation inducing the antiferromagnetic (AF) spin order between A and A’ sites in the unit cell is enhanced as $T$ decreases, when a strong Coulomb interaction is considered. This AF spin ordered insulator state corresponds to the spin ordered massive Dirac electron state mentioned in our previous study [65].

Finally, the $T$-dependence of $1/T_1T = \sum_q \sum_\alpha \text{Im} \left[ \chi_{\alpha,\alpha}^S(q, \omega_0) / \omega_0 \right]$ at $\omega_0 = 0.001$ is displayed in Fig. 12 for $U = 0, U = 0.13$ where FM spin fluctuation is dominant, and $U = 0.46$ where AF spin fluctuation is dominant. The FM spin polarization occurring at weak $U (\approx 0.13)$ causes only a slight increase in the $1/T_1T$ value and has no significant contribution to $1/T_1T$. However, the AF spin polarization causes a clear change in $1/T_1T$. When a strong $U (\approx 0.46)$ is considered and AF spin polarization occurs, $1/T_1T$ tends to increase and diverges as $T$ decreases.

IV. SUMMARY AND DISCUSSION

In this study, to elucidate the low-$T$ insulating state of $\alpha$-(BETS)$_2$I$_3$, a first-principles calculation was first performed to construct an extended Hubbard model with transfer integrals considering the SOC, onsite, nearest-neighbor site, and next-nearest-neighbor site Coulomb interactions. As a result of numerical calculation using the Hartree-Fock approximation, it was found that an interaction induced QSH ordered insulator state similar to the one proposed in the honeycomb lattice [22, 23] stably appears in $\alpha$-(BETS)$_2$I$_3$ by the contribution of the nearest- and next-nearest-neighbor interactions $V$ and $V'$. When the SOC is absent, both the interaction induced QAH and QSH ordered insulator states can appear but has a band gap depending on the order parameters by the interaction induced QSH order. As temperature $T (V$ and $V')$ is decreased (increased), the band gap with the interaction induced QSH order increases continuously. This tendency is well-consistent with the experimental results such as the Onsager phase factor and DC resistivity [59, 61].

In previous studies of the honeycomb lattice model, it was observed that the next-nearest-neighbor interaction $V'$ must be larger than the nearest-neighbor interaction $V$ to cause the interaction induced QSH and QAH orders in the honeycomb lattice model [22]. Furthermore, the interaction induced QSH and QAH ordered insulator states were not found in numerous subsequent studies using other calculation methods such as a density matrix renormalization group [23, 54]. Conversely, recent studies have reported that the interaction induced QSH order appears even in real materials such as digital transition metal oxide hetero structures [23, 35, 37]. It has been suggested that the TI state appears owing to a strong SOC [15, 17, 21, 23]. If a QSH effect can be caused by the Coulomb interaction contribution, there is an advantage in that the QSH effect can be observed with various different materials. In $\alpha$-(BETS)$_2$I$_3$, the long-range Coulomb interaction survives as shown in the results based on the first-principles calculation, and the interaction induced QSH order can be stably realized even at $V' < V$. Of course, it is possible that the interaction induced QSH order in $\alpha$-(BETS)$_2$I$_3$ does not occur as well as the honeycomb lattice model case when other calculation methods and electron correlation effects are taken into account. However, it can be considered that the interaction induced QSH ordered insulator state is one of the candidates that can explain numerous experiments such as X-ray diffraction, DC resistivity, and Shubnikov-de Haas oscillation without any contradiction. Recent studies for the Weyl model have shown a possible ordered state in Dirac electron systems with a long-range
Coulomb interaction \[68\], and according to this study, only a QSH ordered state can occur in \(\alpha-(\text{BETS})_2\text{I}_3\) because other charge, spin, and bond orders have not been observed in the experiment. Therefore, it is strongly expected that the interaction induced QSH ordered insulator state can be realized in an \(\alpha-(\text{BETS})_2\text{I}_3\) at low-\(T\).

Furthermore, we investigated the effect of spin fluctuations on the electronic state of \(\alpha-(\text{BETS})_2\text{I}_3\) using RPA \[71\] \[49\] \[52\]. For a weak interaction case, when \(T\) decreased, only ferrimagnetic spin polarization appears owing to the characteristic wave function of \(\alpha-(\text{BETS})_2\text{I}_3\) \[73\]. This behavior is similar to the ferrimagnetic spin polarization observed in \(\alpha-(\text{ET})_2\text{I}_3\) \[50\].

A recent NMR experiment reported that time reversal symmetry breaking was not observed, and \(1/T_1T\) was proportional to the power of \(T\) and varied continuously near \(T = 50 \times 10^{-4}\) \[63\]. In our calculation using RPA, ferrimagnetic spin polarization has no significant effect on the \(T\)-dependence of \(1/T_1T\) and this is consistent with the experimental results. The ferrimagnetic spin polarization has been observed by a site-resolved NMR experiment in \(\alpha-(\text{ET})_2\text{I}_3\) \[50\].

In future research, the effect of the interaction induced QSH order on \(T_1\) can be calculated using the Weyl model. Theoretical studies with the Dirac Hamiltonian as well as NMR experiments have shown that \(T_1\) in the interaction induced QSH ordered insulator state is affected by orbital current and that the power of \(T\) changes \[80\] \[87\]. It would be interesting to investigate the presence or absence of similar effects in the interaction induced QSH ordered insulator state of \(\alpha-(\text{BETS})_2\text{I}_3\). In recent studies on the interaction induced QSH ordered insulator state, an exotic state localized only at the intersection of the edges of the interaction induced QSH ordered insulator state, has been reported \[23\] \[88\] \[95\]. Future research should investigate whether the interaction induced QSH ordered insulator state suggested in this study has the same properties suggested in preceding studies, using models such as a cylindrical system that considers the real space structure. Moreover, it is necessary to calculate physical quantities such as the Seebeck and Nernst coefficients \[81\]. Finally, there are many unclear points on the low-\(T\) insulating state of \(\alpha-(\text{BETS})_2\text{I}_3\); hence, it is possible that mean-field approximation is not a sufficient calculation method. Calculations considering electron correlation effects using vertex correction \[98\], variational Monte Carlo method \[97\], and functional renormalization group theory \[98\] \[99\] will also be addressed in the future to confirm the stability of the interaction induced QSH ordered insulator state to electron correlation effects and compare the results with previous studies \[23\] \[54\].

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### Appendix A: Values of transfer integrals and Coulomb interactions

In this appendix, we show the values of transfer integrals and Coulomb interactions considering the screening effect using the cRPA method in the RESPACK code \[83\]. Throughout the interaction calculation, we set the energy cutoff of the dielectric function as 5.0 Ry.

The values of the transfer integrals and Coulomb interactions are listed in Table I. The first column of the Table

| \(t_{\alpha,\beta,\sigma}^{(\delta)}\) | \(t_{\alpha,\beta,\sigma}^{(\delta)}\) SOC | Im \(t_{\alpha,\beta,\sigma}^{(\delta)}\) SOC |
|---|---|---|
| \(t_{a1}^{(\delta)}\) | -10.12 | -9.345 | sgn(\(\sigma\)) × 1.365 |
| \(t_{a2}^{(\delta)}\) | -16.31 | -16.80 | sgn(\(\sigma\)) × 0.206 |
| \(t_{a3}^{(\delta)}\) | 51.08 | 50.22 | sgn(\(\sigma\)) × 0.614 |
| \(t_{b1}^{(\delta)}\) | 138.1 | 136.5 | sgn(\(\sigma\)) × 12.06 |
| \(t_{b2}^{(\delta)}\) | 158.7 | 154.1 | sgn(\(\sigma\)) × 19.46 |
| \(t_{b3}^{(\delta)}\) | 65.84 | 63.77 | sgn(\(\sigma\)) × 8.866 |
| \(t_{b4}^{(\delta)}\) | 18.65 | 17.92 | sgn(\(\sigma\)) × 4.205 |
| \(t_{c1}^{(\delta)}\) | 14.09 | 13.88 | sgn(\(\sigma\)) × 0.064 |
| \(t_{c3}^{(\delta)}\) | 4.527 | 4.425 | 0.0 |
| \(t_{c4}^{(\delta)}\) | 21.89 | 21.68 | 0.0 |
| \(t_{d1}^{(\delta)}\) | -1.289 | -1.276 | sgn(\(\sigma\)) × (-0.186) |
| \(t_{d3}^{(\delta)}\) | -1.280 | -1.161 | 0.0 |
| \(t_{d4}^{(\delta)}\) | 2.494 | 2.636 | 0.0 |

| \(W_{\alpha,\beta}^{(0)}\) | \(W_{\alpha,\beta}^{(\delta)}\) |
|---|---|
| \(U_A\) | 1383 | 580.5 |
| \(U_{A'}\) | 1383 | 596.2 |
| \(U_B\) | 1396 | 566.7 |
| \(U_C\) | 1359 | 579.9 |
| | \(V_{a1}\) | 572.9 |
| | \(V_{a2}\) | 537.8 |
| | \(V_{a3}\) | 556.9 |
| | \(V_{a4}\) | 329.8 |
| | \(V_{b1}\) | 328.1 |
| | \(V_{b3}\) | 329.1 |
| | \(V_{b4}\) | 329.9 |
| | \(V_{b5}\) | 323.9 |
| | \(V_{b6}\) | 332.4 |
Coulomb interaction \[eV\]

| Value | \(\alpha\) is charge geometrical frustration in \(V\) | \(W\) are the molecule indices in the unit cell (A, A', B, C) |
|--------|------------------------------------------------------|-------------------------------------------------------------|
| -1     | \(0\) \(1\) \(2\) \(3\) \(4\) \(5\) \(6\) \(7\) \(8\) \(9\) \(10\) | \(329\) \(97\) \(96\) \(95\) \(94\) \(93\) \(92\) \(91\) \(90\) \(89\) |

The values of the real part of the transfer integrals \(t^{(\delta)}\) and \(t^{(\delta)}\) (SOC), are listed in the second (third) column. The fourth column shows the imaginary part of the transfer integrals \(\text{Re} t^{(\delta)}\). The bottom of the second column lists the values of the real part of the effective direct integral \(t^{(\delta)}\), calculated using RESPACK. Here, \(\delta = (\delta_a, \delta_b)\) is the relative lattice vector in the \(a-b\) plane, and \(\alpha\) and \(\beta\) are the molecule indices in the unit cell (A, A', B, C). \(W^{(\delta)}\) is an interaction taking into account the screening effect based on the first-principles calculations [84]. The average values of the nearest-neighbor and next-nearest-neighbor components are \(V_a = \frac{1}{3} \sum_{\alpha=1}^{2} V_{an} = 581.1\) meV, \(V_b = \frac{1}{4} \sum_{n=1}^{4} V_{bn} = 561.9\) meV, \(V_a' = \frac{1}{3} \sum_{\alpha=1}^{2} V'_{an} = 329.0\) meV, and \(V_b' = \frac{1}{3} \sum_{n=1}^{4} V'_{bn} = 327.7\) meV. As \(V_b/V_a = 0.967 \simeq 1\) and \(V_b'/V_a' = 0.996 \simeq 1\), there is charge geometrical frustration in \(\alpha\)-(BETS)\(2\)I\(3\). This charge geometrical frustration effect is one of characteristic features of organic conductors [100].

FIG. A.1. (Color online) Range \(|R|\)-dependence of the bare direct integral \(V_a(R)\) and the static effective direct integral \(W_a(R)\) at the A site (\(\alpha = A\)) for (a) \(\alpha\)-(BETS)\(2\)I\(3\) at 30 K, and (b) result of aluminum (Al) as an example.

In this appendix, we calculated the electronic state in the Hartree-Fock approximation and drew the phase diagram as a function of the strength of spin-orbit coupling and the onsite Coulomb interaction \(U\) to compare with the results from preceding studies for other models such as the honeycomb lattice model [23, 103, 104]. To change the strength of SOC as a parameter, the SOC values obtained by the first-principles calculation in this study is multiplied by \(\lambda_{SOC}\). Furthermore, to investigate the contribution of \(U\) to the electronic state in the presence of SOC, calculation was performed considering the spin order as a stable solution, which was prohibited in the main text. It was also confirmed that the relation between parameters \((U, \lambda_{SOC})\) and spin ordered massive Dirac electron (SMD) phase proposed in our preceding study [63] which is caused by \(U\) and associated with time-reversal symmetry breaking.

The calculation result is shown in Fig. B.1. As \(U\) is increased and \(U > U_{SMD}\), spin ordered massive Dirac electron phase associated with antiferromagnetism between the A and A' sites in the unit cell occurs [65]. In the spin ordered massive Dirac electron phase, the time-reversal symmetry is broken owing to antiferromagnetism between the A and A' sites in the unit cell. It is considered that this spin ordered phase corresponds to the AF insulator phase (AFI) in preceding studies for the honeycomb lattice model [23, 103, 104]. In \(U < U_{SMD}\), the topological insulator (TI) phase appears owing to the contribution of SOC alone, as in the previous study for the honeycomb lattice model [23, 103, 104].
Appendix C: Relation of spin ordered massive Dirac electron phase and the interaction induced QSH ordered insulator state

Next, to investigate the relationship between the spin ordered massive Dirac electron phase in a previous study and the interaction induced QSH ordered insulator state and charge-ordered phases in $\alpha$-(BETS)$_2$I$_3$, calculation using the Hartree-Fock approximation with $U$, $V$, and $V'$ was performed by considering the spin order as a stable solution as in the previous appendix. In previous studies for $\alpha$-(ET)$_2$I$_3$, a $U$-$V$ phase diagram is drawn and it has been shown that the spin order is stabilized when $U \gg 0$ and $V$ is small, and the horizontal stripe charge order is stabilized when $V \gg 0$ and $U$ is small. In the following, we fixed $\lambda_{SOC}$, $V$, and $T$ at $(\lambda_{SOC}, V, T) = (1, 0.1, 1 \times 10^{-4})$ and draw a $U$-$V'$ phase diagram.

Figure C.1 represents the $U$-$V'$ phase diagram calculation result. When both $U$ and $V'$ are small such as $U < U^{CO}$, the horizontal strip charge order phase owing to the contribution of $V$ alone appears. The spin ordered massive Dirac electron phase is stabilized in $U > U^{SMD}$, and the interaction induced QSH ordered insulator state appears during the transition between the spin ordered massive Dirac electron and horizontal strip charge order phases, $U^{CO} < U < U^{SMD}$. This indicates that the interaction induced QSH ordered insulator state is not energetically stable under physically unrealistic values of $V$ and $V'$ such as in the case of $V' \gg V$ when the spin order is allowed as a stable solution in $\alpha$-(BETS)$_2$I$_3$, and not only $V'$ but also $V$ significantly contributes to the emergence of the interaction induced QSH ordered insulator state.

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