Topological Mott insulator phase and metal-insulator crossover in 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 construct a two-dimensional extended Hubbard model with transfer integrals including spin-orbit-coupling (SOC) and Coulomb interaction, based on first-principles calculation. We investigate the low-$T$ electronic state using Hartree Fock approximation, and found that the topological Mott insulator (TMI) state is a candidate for the insulating state. In this state, energy gap in the high-$T$ weak topological insulator (TI) state is strongly enhanced by next-nearest-neighbor interactions $V''$. With the increase in $T$, the TMI state continuously changes to a high-$T$ weak TI state. Calculations using random-phase-approximation also indicate that the fermi-magnetic spin fluctuation appears in the high-$T$ TI state, when weak onsite interaction is considered.

I. INTRODUCTION

Dirac electrons in solids such as graphene [1, 2], bismuth [3, 4], and several organic conductors [5–12] exhibit various physical properties, including quantum conduction [13], large diamagnetism [14], and anomalous electron correlation effects. Therefore, Dirac electron systems have attracted attention in both theoretical and experimental research. In organic Dirac electron systems such as $\alpha$-(BEDT-TTF)$_2$I$_3$ and $\alpha$-(BEDT-TSeF)$_2$I$_3$ ($\alpha$-(ET)$_2$I$_3$ and $\alpha$-(BETS)$_2$I$_3$), which are the main focus of this study, the Coulomb interaction between Dirac electrons is relatively large because of the small values of the transfer integrals, resulting in low group velocity.

The two-dimensional (2D) massless Dirac electron system in $\alpha$-(ET)$_2$I$_3$ transitions to the insulator phase with stripe charge order [15, 17] owing to nearest-neighbor (N.N.) Coulomb interaction. Mass generation because of the stripe charge order leads to unique temperature ($T$) and pressure ($P$) dependencies of the transport coefficients and spin gap [18, 22]. In the massless Dirac electron phase, fermi-magnetic spin fluctuation, velocity renormalization, and exotic spin fluctuation owing to electron correlation effects have been reported by theoretical studies as well as nuclear magnetic resonance (NMR) experiments under an in-plane magnetic field [23–28].

Organic conductor $\alpha$-(BETS)$_2$I$_3$ is similar to $\alpha$-(ET)$_2$I$_3$. Both have almost the same crystalline structure and 2D Dirac electron systems, obtained by first-principles calculation using high-accuracy X-ray diffraction data [29]. The transfer integrals and spin-orbit coupling (SOC) of $\alpha$-(BETS)$_2$I$_3$ are greater than those of $\alpha$-(ET)$_2$I$_3$ [30]. The slightly stronger SOC of $\alpha$-(BETS)$_2$I$_3$ compared to that of $\alpha$-(ET)$_2$I$_3$ is derived from the substitution of sulfur (S) atoms by selenium (Se) atoms, and its value is estimated to be approximately 2 meV using generalized gradient approximation (GGA) [31, 32]. This result suggests that $\alpha$-(BETS)$_2$I$_3$ becomes a topological insulator (TI) with a finite SOC gap at the Dirac cone. The energy band structure of $\alpha$-(BETS)$_2$I$_3$ has been extensively studied [33–35], and the appearance of a weak TI due to SOC has been reported [31, 32].

Interestingly, although $\alpha$-(BETS)$_2$I$_3$ has a TI state, a clearly different insulating state appears with the decrease in $T$. Quantum conduction has been observed by direct current (DC) resistivity measurements at $T > 50$ K (high-$T$) and $P > 5$ kbar [36, 37], and the DC resistivity increases sharply at $T < 50$ K (low-$T$) and $P < 5$ kbar. Shubnikov–de Haas oscillation measurements for a carrier-doped sample ($\sim 10$ K) indicate that the Onsager phase factor switches from 1/2 to zero at $P \approx 5$ kbar at which the insulating state vanishes [37, 38]. The results of a synchrotron X-ray diffraction experiment suggest that the insulating state of $\alpha$-(BETS)$_2$I$_3$ does not involve spatial inversion symmetry breaking and change in the bond length between N.N. BETS molecules [29]. Furthermore, NMR experiments show that the spin susceptibility and $1/T_1 T$ are proportional to $T$ and $T^2$, respectively, near the emergence of the insulating state, and no signs of a spin order are observed [39, 40]. Therefore, an insulating state unrelated to the charge, bond, and spin order is expected to appear at low-$T$ in $\alpha$-(BETS)$_2$I$_3$.

In our recent study [41], we had investigated the insulating state of $\alpha$-(BETS)$_2$I$_3$ using a 2D Hubbard model with Hartree approximation; a spin ordered massive Dirac electron (SMD) 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 [39, 40]. We need to consider not only the effect of 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 [42, 44]. In this study, we investigate the possibility of a topological Mott insulator (TMI) state [45, 50] as a candidate for the insulating state of α-(BETS)$_2$I$_3$. It is known that the TMI state is caused by the phase modulation in the transfer integrals [45], the contribution of SOC [46], and the next-nearest-neighbor (N.N.N.) Coulomb interaction [47].

The remainder of this paper is organized as follows. In Section II, we perform 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). In Section III, the electronic state of α-(BETS)$_2$I$_3$ at low-$T$ is firstly calculated applying Hartree Fock (HF) approximation. Next, the stability of the TMI state due to the presence of N.N. and N.N.N. Coulomb interactions is investigated, and the phase modulation of the transfer integrals due to the emergence of TMI is calculated. Moreover, physical quantities, such as the Onsager phase factor and DC conductivity, are calculated using $T$-matrix approximation [19, 51, 54], and compared with a recent experiment [57]. Finally, we investigate the effect of spin fluctuation on high-$T$ electronic state of α-(BETS)$_2$I$_3$ using random phase approximation (RPA) [7, 23, 26]. 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 α-(BETS)$_2$I$_3$ at 30K under ambient pressure [29]. As the exchange-correlation function, GGA was used in the Quantum espresso (QE) package [55, 56]. We used SG15 optimized norm-conserving Vanderbilt (ONCV) fully relativistic pseudopotentials to consider the effect of SOC in the first-principles calculation [57]. 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 first-principles calculation, to obtain the transfer integrals with SOC, maximally localized Wannier functions (MLWFs) were created using the Wannier90 code [58]. Eight bands near the Fermi energy were selected to construct the MLWFs, and the initial coordinates of MLWFs were set at the center of the BETS molecule in the unit cell.

Figure 1(a) shows the real-space distribution of the MLWFs for α-(BETS)$_2$I$_3$ under ambient pressure at 30 K in a unit cell drawn by VESTA [59]. (b) Energy band structure with SOC effect obtained by first-principles calculation.

FIG. 1. (Color online) (a) Crystal structure and real-space distribution of the MLWFs for α-(BETS)$_2$I$_3$ under ambient pressure, (b) Energy band structure with SOC calculated using QE and the Wannier interpolation performed using Wannier90. The energy origin is set at the Fermi energy.

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.

Next, we calculated the long-range Coulomb interaction considering the screening effect using the constrained random phase approximation (cRPA) method in the RESPACK code [60]. We set the energy cutoff of the dielectric function as 5.0 Ry.

Figure 2 (a) and (b) show the two-dimensional (2D)
TABLE I. List of transfer integrals without (with) SOC $t_{\alpha,\beta,\sigma}^{(\delta, \text{SOC})}$, and effective Coulomb interactions $W_{\alpha,\beta}^{(\delta)}$ (units of meV). Here, $\text{sgn}(\sigma)$ is $+1 (\sigma = \uparrow)$ or $-1 (\sigma = \downarrow)$.

| $t_{\alpha,\beta,\sigma}$ | $t_{\alpha,\sigma,\beta,\sigma}^{\text{SOC}}$ | $\text{Im}[t_{\alpha,\beta,\sigma}^{(\delta, \text{SOC})}]$ | $\text{Im}[t_{\alpha,\sigma,\beta,\sigma}^{(\delta, \text{SOC})}]$ |
|-------------------------|---------------------------------|---------------------------------|---------------------------------|
| $t_{\alpha,1}$          | -10.12                          | -9.345                          | $\text{sgn}(\sigma) \times 1.365$ |
| $t_{\alpha,2}$          | -16.31                          | -16.80                          | $\text{sgn}(\sigma) \times 0.206$ |
| $t_{\alpha,3}$          | 51.08                           | 50.22                           | $\text{sgn}(\sigma) \times 0.614$ |
| $t_{\alpha,4}$          | 138.1                           | 136.5                           | $\text{sgn}(\sigma) \times 12.06$ |
| $t_{\alpha,5}$          | 158.7                           | 154.1                           | $\text{sgn}(\sigma) \times 19.46$ |
| $t_{\alpha,6}$          | 65.84                           | 63.77                           | $\text{sgn}(\sigma) \times 8.866$ |
| $t_{\alpha,7}$          | 18.65                           | 17.92                           | $\text{sgn}(\sigma) \times 4.205$ |
| $t_{\alpha,8}$          | 14.09                           | 13.88                           | $\text{sgn}(\sigma) \times 0.064$ |
| $t_{\alpha,9}$          | 4.527                           | 4.425                           | 0.0                              |
| $t_{\alpha,10}$         | 21.89                           | 21.68                           | 0.0                              |
| $t_{\alpha,11}$         | -1.289                          | -1.276                          | $\text{sgn}(\sigma) \times (-0.186)$ |
| $t_{\alpha,12}$         | -1.280                          | -1.161                          | 0.0                              |
| $t_{\alpha,13}$         | 2.494                           | 2.636                           | 0.0                              |

The values of the transfer integrals and Coulomb interactions are listed in Table I. The first column of the Table I presents the notations of the inter-molecular transfer integrals and interactions shown in Figs. 2 (a) and (b). The values of the real part of the transfer integrals without (with) SOC, $\text{Re}[t_{\alpha,\beta,\sigma}^{(\delta, \text{SOC})}]$ ($\text{Re}[t_{\alpha,\sigma,\beta,\sigma}^{(\delta, \text{SOC})}]$), are listed in the second (third) column. The forth column shows the imaginary part of the transfer integrals with SOC $\text{Im}[t_{\alpha,\beta,\sigma}^{(\delta, \text{SOC})}]$. Inter-plane transfer integrals are not considered in this study because they are considerably smaller than those in the intra-plane (approximately 1/1000 order of the intra-plane components). The bottom of the second order of the values of the real part of the effective direct integral $\text{Re}[W_{\alpha,\beta}]$, calculated using RESPACK. Here, $\delta = (\delta_b, \delta_a)$ is the relative lattice vector in the $a$-$b$ plane, and $\alpha$ and $\beta$ are the indices of the molecules in the unit cell (A, A', B, C). The onsite components $U_\alpha = \text{Re}[W_{\alpha,\alpha}^{(0)}]$ and the average values of the N.N. and N.N.N. components are $V_{a} = \frac{1}{3} \sum_{n=1}^{3} V_{an} = 581.1 \text{ meV}$, $V_{b} = \frac{1}{4} \sum_{n=1}^{4} V_{bn} = 561.9 \text{ meV}$, $V_{\alpha} = \frac{1}{3} \sum_{n=1}^{3} V_{\alpha n} = 329.0 \text{ meV}$, and $V_{b} = \frac{1}{4} \sum_{n=1}^{4} V_{b n} = 327.7 \text{ meV}$. As $V_{b}/V_{a} = 0.967 \simeq 1$ and $V_{\alpha}^{b}/V_{\alpha}^{a} = 0.996 \simeq 1$, there is charge geometrical frustration in $\alpha$-(BETS)$_2$I$_3$. This charge geometrical frustration effect is one of characteristic features of organic conductors [61].

Figure 3 (a) shows the range (|R|)-dependence of the bare direct integral $V_{\alpha}(R)$ and static effective direct integral $W_{\alpha}(R)$ at the A site ($\alpha = \text{A}$) for (a) $\alpha$-(BETS)$_2$I$_3$ at 30 K, and (b) result of aluminium (Al) as an example.
effective Coulomb interaction is considerably decreased by the screening effect, and even the N.N. component becomes zero.

Based on the above first-principles calculation results, we constructed a 2D extended Hubbard model [15]:

$$
H = \sum_{\mathbf{R}, \delta} \sum_{\alpha, \beta} \sum_{\sigma_1, \sigma_2} t^{(\delta)\text{SOC}}_{\alpha, \beta; \sigma_1, \sigma_2} c_{\mathbf{R}, \alpha, \sigma_1}^\dagger c_{\mathbf{R} + \delta, \beta, \sigma_2} + \lambda \sum_{\mathbf{R}, \alpha} U_\alpha n_{\mathbf{R}, \alpha, \uparrow} n_{\mathbf{R}, \alpha, \downarrow} + \frac{\lambda}{2} \sum_{\mathbf{R}, \delta} \sum_{\alpha, \beta} \sum_{\sigma_1, \sigma_2} V^{(\delta)}_{\alpha, \beta} n_{\mathbf{R}, \alpha, \sigma_1} n_{\mathbf{R} + \delta, \beta, \sigma_2},
$$

(1)

where \( \mathbf{R} \) is the coordinate of the unit cell, \( \alpha \) and \( \beta \) are the site indices, and \( \sigma_1 \) and \( \sigma_2 \) are the spin indices (\( \uparrow, \downarrow \)). \( t^{(\delta)\text{SOC}}_{\alpha, \beta; \sigma_1, \sigma_2} \) is the transfer integral between \( (\alpha, \sigma_1) \) and \( (\beta, \sigma_2) \) separated by \( \delta \). Site potentials \( t^{(0)}_{\alpha} = t^{(0)}_{\beta} = 4.470 \) eV, \( t^{(0)}_{\text{B}} = 4.465 \) eV, and \( t^{(0)}_{\text{P}} = 4.477 \) eV are excluded in our model because their contribution to the energy band is negligible. \( c_{\mathbf{R}, \sigma_1} (c_{\mathbf{R}, \sigma_1}^\dagger) \) is the creation (annihilation) operator for the \( \alpha \) site with spin \( \sigma_1 \) in the unit cell located at \( \mathbf{R} \). \( U_\alpha \) and \( V^{(\delta)}_{\alpha, \beta} \) are the onsite and inter-site Coulomb interactions, respectively, given by the static effective direct integrals \( W^{(\delta)}_{\alpha, \beta} \) calculated using RESPACK.

\( \lambda \) (0 < \( \lambda < 1 \)) is multiplied with \( W^{(\delta)}_{\alpha, \beta} \) in our model to control the strength of \( W^{(\delta)}_{\alpha, \beta} \). We also define the number operator as \( n_{\mathbf{R}, \alpha, \sigma} = c_{\mathbf{R}, \alpha, \sigma}^\dagger c_{\mathbf{R}, \alpha, \sigma} \). In the following, the lattice constants, Boltzmann constant \( k_B \), and the Planck constant \( h \) are considered to be unity. Electron-volt (eV) is used as the unit of energy throughout this paper.

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

We treat Eq. (1) within the Hartree-Fock (HF) approximation in the wave number space. Fourier inverse transform, \( c_{\mathbf{R}, \sigma_1} = N^{-1/2} \sum_{\mathbf{k}} c_{\mathbf{k}, \alpha, \sigma_1} e^{i \mathbf{k} \cdot \mathbf{R}} \), was performed on Eq. (1). Here, \( N_{\text{cell}} \) is the total number of unit cells and \( \mathbf{k} = (k_x, k_y) \) indicates the wave-number vector. The Hartree-Fock Hamiltonian is as follows:

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

$$
H_T = \sum_{\mathbf{k}} \sum_{\alpha, \beta} \sum_{\sigma_1, \sigma_2} t^{(\delta)\text{SOC}}_{\alpha, \beta; \sigma_1, \sigma_2} c^\dagger_{\mathbf{k}, \alpha, \sigma_1} c_{\mathbf{k}, \beta, \sigma_2} e^{i \mathbf{k} \cdot \mathbf{R}}
$$

$$
H_U = \lambda \sum_{\mathbf{k}} \sum_{\alpha} U_\alpha \sum_{\sigma_1} \left( \frac{1}{N_{\text{cell}}} \sum_{\mathbf{q}} \left( (c_{\mathbf{k}, \alpha, \sigma_1} c_{\mathbf{k}, \alpha, \sigma_1}^\dagger) c_{\mathbf{k} + \mathbf{q}, \alpha, \sigma_1} c_{\mathbf{k}, \alpha, \sigma_1}^\dagger + (c_{\mathbf{k}, \alpha, \sigma_1}^\dagger c_{\mathbf{k} + \mathbf{q}, \alpha, \sigma_1}^\dagger) c_{\mathbf{k}, \alpha, \sigma_1} c_{\mathbf{k} + \mathbf{q}, \alpha, \sigma_1} \right) \right)
$$

(2)

$$
H_V = \frac{\lambda}{2} \sum_{\mathbf{k}} \sum_{\alpha, \beta} \sum_{\sigma_1, \sigma_2} \sum_{\mathbf{q}} V^{(\delta)}_{\alpha, \beta} \left( \frac{2}{N_{\text{cell}}} \sum_{\mathbf{k}} \left( c_{\mathbf{k}-\mathbf{q}, \alpha, \sigma_1} c_{\mathbf{k}, \beta, \sigma_2} c_{\mathbf{k} + \mathbf{q}, \sigma_2} c_{\mathbf{k}, \alpha, \sigma_1} + (c_{\mathbf{k} + \mathbf{q}, \alpha, \sigma_1}^\dagger c_{\mathbf{k}, \beta, \sigma_2} c_{\mathbf{k}, \alpha, \sigma_1} c_{\mathbf{k} + \mathbf{q}, \sigma_2}^\dagger) \right) \right)
$$

Thus, \( H_{\text{HF}} \) for each \( \mathbf{k} \) can be finally expressed as follows:

$$
H_{\alpha, \sigma_1; \beta, \sigma_2} (\mathbf{k}) = \sum_{\delta} t^{(\delta)\text{SOC}}_{\alpha, \beta; \sigma_1, \sigma_2} c^\dagger_{\mathbf{k}, \alpha, \sigma_1} c_{\mathbf{k}, \beta, \sigma_2} e^{i \mathbf{k} \cdot \mathbf{R}} + \lambda U_\alpha \delta_{\alpha, \beta} \sum_{\sigma_1, \sigma_2} \left( c_{\mathbf{k}, \alpha, \sigma_1}^\dagger c_{\mathbf{k}, \alpha, \sigma_1} - \delta^{(0)}_{\alpha, \sigma_1; \alpha, \sigma_1} c_{\mathbf{k}, \alpha, \sigma_1}^\dagger c_{\mathbf{k}, \alpha, \sigma_1} \right)
$$

$$
+ \frac{\lambda}{2} \sum_{\delta} V^{(\delta)}_{\alpha, \beta} \left( \frac{2}{N_{\text{cell}}} \sum_{\mathbf{k}} \left( c_{\mathbf{k}, \alpha, \sigma_1}^\dagger c_{\mathbf{k}, \alpha, \sigma_1} c_{\mathbf{k}, \alpha, \sigma_1} + h.c. \right) \right) + \delta^{(\delta')}_{\alpha, \beta; \sigma_1, \sigma_2} e^{-i \mathbf{k} \cdot \mathbf{R}} c_{\mathbf{k}, \beta, \sigma_2} c_{\mathbf{k}, \alpha, \sigma_1} + h.c.
$$

(4)

where the off-diagonal site component of the order parameter, \( \delta^{(\delta')}_{\alpha, \beta; \sigma_1, \sigma_2} \equiv \langle c_{\mathbf{k}, \alpha, \sigma_1}^\dagger c_{\mathbf{k}, \beta, \sigma_2} \rangle (\alpha \neq \beta) \), modulates the transfer integrals.

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

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

$$= \left\langle \sum_{\alpha,\beta} \sum_{\sigma_1,\sigma_2} d_{\alpha,\sigma_1;\nu,\sigma}(k)H_{\alpha,\sigma_1;\beta,\sigma_2}(k)d_{\beta,\sigma_2;\nu,\sigma}(k) \right\rangle - \mu,$$

where $\mu$ is the chemical potential calculated to satisfy $3/4$-filling. $\tilde{E}_{\nu,\sigma}(k)$ are the eigenvalues with band index $\nu$ obtained by numerical diagonalization \[ |E_{1,\sigma}(k) > |E_{2,\sigma}(k) > |E_{3,\sigma}(k) > |E_{4,\sigma}(k) |. \]

The Berry curvature $B_{\nu,\sigma}(k)$ is calculated as

$$B_{\nu,\sigma}(k) = \sum_{\nu' \neq \nu} \frac{\nu_{\nu',\nu}(k)\nu_{\nu',\nu}(k)}{i [E_{\nu,\sigma}(k) - E_{\nu',\sigma}(k)]^2} + c.c.,$$

where the velocity matrix $\nu_{\nu',\nu}(k)$ along the $a$-axis ($\eta = a(y)$) and $b$-axis ($\eta = b(x)$) is given by

$$\nu_{\nu',\nu}(k) = \sum_{\alpha,\beta} \sum_{\sigma_1,\sigma_2} d_{\alpha,\sigma_1;\nu,\sigma}(k)$$

$$\times \frac{\partial H_{\alpha,\sigma_1;\beta,\sigma_2}(k)}{\partial k_\eta} d_{\beta,\sigma_2;\nu',\sigma}(k).$$

The Onsager phase factor $\gamma$ and Berry phase $\phi_B$ were obtained based on the semiclassical theory [34 58]:

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

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

$\int_{S_F}$ indicates surface integration on the Fermi surface. In $\gamma$ calculation, we used an electron-doped band of approximately 0.005, which is of the same order as the carrier doping value in the experiment [27].

C. Conductivity

The DC conductivity was calculated using the Nakano-Kubo formula [13 51 52]:

$$\sigma_\eta(\omega) = \frac{1}{i\omega} \left[ Q^R_\eta(\omega) - Q^R_\eta(0) \right],$$

$$Q^R_\eta(\omega) = \frac{e^2}{N_{cell}} \sum_{k,\nu',\nu,\sigma} |\nu^\eta_{\nu',\nu}(k)|^2 \chi^{0}_{\nu',\nu}(k,\omega),$$

$$\chi^{0}_{\nu',\nu}(k,\omega) = -\frac{f(E_{\nu,\sigma}(k)) - f(E_{\nu',\sigma}(k))}{E_{\nu,\sigma}(k) - E_{\nu',\sigma}(k) + i\omega + i0^+},$$

where $\eta = b(x)$ and $a(y)$ are the axes and $\omega$ is the frequency. $0^+$ is set as $0^+ = 5 \times 10^{-4}$, in this study. In the limit of $\omega \rightarrow 0$, we can obtain the longitudinal conductivity along the $a$-axis ($\eta = a(y)$) and $b$-axis ($\eta = b(x)$):

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

$$\Phi_\eta(\omega) = \frac{2e^2}{N_{cell}} \sum_{k,\nu,\sigma} |\nu^\eta_{\nu,\sigma}(k)|^2 \tau_{\nu,\sigma}(\omega, k)\delta(h\omega - E_{\nu,\sigma}(k)).$$

We treat the effect of elastic scattering between electrons and the impurities originating from the lack and disorder of $I^3$-molecules, using $T$-matrix approximation. The impurity potential term $H_{imp}$ is defined by

$$H_{imp} = \frac{V_0}{N_{cell}} \sum_{k,\nu,\sigma} N_{imp} \sum_{i} e^{-i\mathbf{q}_i \cdot \mathbf{r}_i} c^\dagger_{k+\mathbf{q}_i,\alpha,\sigma} c_{k,\alpha,\sigma},$$

and we treat it using the perturbation theory for Green’s function. Here, $V_0$ is the strength of the potential and $r_i$ ($i = 1, \cdots, N_{imp}$) is the coordinate of the $i$-th impurity. The damping constant $\gamma_{\nu,\sigma}(\omega, k)$ is obtained by calculation as follows:

$$\gamma_{\nu,\sigma}(\omega, k) = \frac{\hbar}{2^\nu_{\nu,\sigma}(\omega, k)} = \frac{\text{Im} \Sigma^R_{\nu,\sigma}(\omega, k)}{1 + \{\pi V_0^2 \rho^E_{\sigma}(\omega)\}^2},$$

where $c_{imp} = \frac{N_{imp}}{N_{cell}} = 0.02 \ll 1$ is impurity density and $\rho^E_{\sigma}(\omega)$ represents the density of state for spin $\sigma$:

$$\rho^E_{\sigma}(\omega) = \sum_{k,\alpha,\sigma,\nu} |d_{\alpha,\sigma_1;\nu,\sigma}(k)|^2 \delta(h\omega - E_{\nu,\sigma}(k))$$

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

D. Calculation of the spin fluctuation using RPA

We investigated the effect of spin fluctuation on the NMR properties, e.g., the Knight shift and $1/T_1T$ in the high-$T$ Dirac electron phase in RPA using eq. [11 23 25].

The bare Green’s function on the site representation is defined as

$$G_{\alpha,\sigma_1;\beta,\sigma_2}(k, \varepsilon_n) = \sum_{\nu,\sigma} d_{\alpha,\sigma_1;\nu,\sigma}(k)d^*_{\beta,\sigma_2;\nu,\sigma}(k) \frac{1}{i\varepsilon_n - E_{\nu,\sigma}(k)},$$

where $\varepsilon_n = (2n+1)\pi T$ is the Matsubara frequency. In the linear response theory, the bare susceptibility
The intra-band component of the spin susceptibility can be calculated as follows:

\[
\chi^0_{\alpha,\beta}(q,\omega_m) = \frac{-T}{N_{\text{cell}}} \sum_{k,n} G_{\alpha,\sigma_1,\beta,\sigma_2}(k+q,\varepsilon_n+\omega_m)G_{\beta,\sigma_2,\alpha,\sigma_2}(k,\varepsilon_n) 
\]

\[
= \frac{1}{N_{\text{cell}}} \sum_{k} \sum_{\sigma,\sigma',\nu,\nu'} ^4 F_{\nu,\sigma_1,\nu',\sigma_1'}(k,q) \chi^0_{\nu,\sigma_1,\nu',\sigma_1'}(q,\omega_m),
\]

\[
\chi^0_{\nu,\sigma_1,\nu',\sigma_1'}(q,\omega_m) = -\frac{f(E_{\nu,\sigma}(k+q)) - f(E_{\nu',\sigma'}(k))}{E_{\nu,\sigma}(k+q) - E_{\nu',\sigma'}(k) + i\omega_m},
\]

where \( N_{\text{cell}} \) is the system size and \( \omega_m = 2m\pi T \). \( F(k,q) \) indicates the form factor represented by

\[
F_{\alpha,\sigma_1;\beta,\sigma_2}(k, q) = d_{\alpha,\sigma_1,\nu,\sigma}(k+q) d^*_{\beta,\sigma_2,\nu',\sigma'}(k+q) 
\times d_{\beta,\sigma_2,\nu',\sigma'}(k) d^*_{\alpha,\sigma_1,\nu,\sigma}(k).
\]

In RPA, the spin susceptibility \( \hat{\chi}^S \) and the transverse spin susceptibility \( \hat{\chi}^T \) are calculated as follows:

\[
\hat{\chi}^S(q,\omega) = \hat{\chi}^T(q,\omega) = \left( \hat{1} - \hat{\chi}^0(q,\omega) \hat{U} \right)^{-1} \hat{\chi}^0(q,\omega),
\]

where \( \hat{1} \) is the unit matrix and \( \hat{U}_{\alpha} = U_{\alpha}(\lambda U)\delta_{\alpha,\beta} \). Moreover, to estimate the contribution of the intra- and inter-band components to the spin fluctuation, we divide the bare susceptibility into two components \( \hat{\chi}^0 \) as follows:

\[
\chi^0,\text{Intra}_{\alpha,\sigma_1;\beta,\sigma_2}(q,\omega) = \frac{1}{N_{\text{cell}}} \sum_{k} \sum_{\sigma,\sigma',\nu,\nu'} F_{\nu,\sigma_1,\nu',\sigma_1'}(k,q) 
\times \chi^0_{\nu,\sigma_1,\nu',\sigma_1'}(q,\omega),
\]

\[
\chi^0,\text{Inter}_{\alpha,\sigma_1;\beta,\sigma_2}(q,\omega) = \frac{1}{N_{\text{cell}}} \sum_{k} \sum_{\sigma,\sigma',\nu,\nu'} F_{\nu,\sigma_1,\nu',\sigma_1'}(k,q) 
\times \chi^0_{\nu,\sigma_1,\nu',\sigma_1'}(q,\omega),
\]

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

\[
\hat{\chi}^{S,\text{Intra}} = \left( \hat{1} - \chi^0,\text{Intra} \hat{U} \right)^{-1} \hat{\chi}^0,\text{Intra}.
\]

The inter-band component \( \chi^{S,\text{Inter}} \) is also obtained based on definition \( \chi^{S,\text{Inter}} = \chi^S - \chi^{S,\text{Intra}} \). Applying analytical continuation \( i\omega_m \rightarrow \omega \rightarrow i\omega^+ \), the site-resolved Knight shift \( K_{\alpha} \) and \( 1/T_1 \) in RPA are obtained as follows:

\[
K_{\alpha} = \sum_{\beta} \text{Re} \left[ \chi^{S,\alpha,\beta}(q = 0, \omega = 0) \right],
\]

\[
1/T_1 = \sum_q \sum_{\alpha} \frac{\text{Im} \left[ \chi^{S,\alpha,\beta}(q, \omega_0) \right]}{\omega_0},
\]

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 temperature

In this subsection, the numerical results for the electronic state with HF approximation are shown. We set the initial states randomly and investigated the electronic state with the lowest energy, other than the charge, spin, and bond orders, with HF approximation. Throughout this subsection, \( T = 1 \times 10^{-4} \).

Figure 4(a) shows the \( \lambda \)-dependence of the energy gap \( \Delta \) at the Dirac point \( \Delta \) with and without SOC. In the absence of SOC, \( \Delta \) opens at \( \lambda > \lambda_C \), and phase transition from the zero gap Dirac electronic state (ZGS) to the topological Mott insulator (TMI) state occurs. On the other hand, in the presence of SOC, the electronic system becomes a topological insulator (TI) at \( \lambda = 0 \). With the increase in \( \lambda \), \( \Delta \) increases continuously. In Fig. 4(b), the \( \lambda \)-dependence of the imaginary part of the order parameter is shown.
parameter \( \text{Im}[\delta b_{b1,\sigma}] \) and \( \text{Im}[\delta b_{b4,\sigma}] \) with SOC are plotted. Due to the contribution of SOC, \( \delta b'_{b1,\pm} \) \((\delta b_{b4,\pm})\) and \( \delta b''_{b1,\pm} \) \((\delta b''_{b4,\pm})\) have opposite signs. With the increase in \( \lambda \), \( \text{Im}[\delta b] \) gradually increases above \( \lambda > 0 \). In Fig. 4(c), the \( \lambda \)-dependence of the charge density \( \langle n_{\alpha} \rangle \) is shown. At \( \lambda = 0 \), charge disproportion due to the chemical potential is observed, and as \( \lambda \) increases, the charge densities continuously become uniform. It is to be noted that when SOC is considered, the TMI state caused by the contribution of Coulomb interaction and SOC [12, 50] appears in the region above \( \lambda > 0 \). The difference between the TI and TMI states is summarized as follows: The TI state has a slight gap of approximately 2 meV due to the contribution of the SOC alone [32, 41]. The gap in the TI state is enhanced by the contribution of Coulomb interaction when \( \lambda > 0 \), and we refer to this state as the TMI state with SOC. In the TMI state with SOC, phase modulation of the transfer integrals occurs, as shown in Fig. 4(b). On the other hand, the TMI state without SOC at \( \lambda > \lambda_C \) is induced by the contribution of Coulomb interaction alone [12, 50]. In this state, the sign of order parameter \( \delta b \) does not depend on the degrees-of-freedom of the spin. The TMI state with and without SOC is analogous to a ferromagnet with and without an external magnetic field.

Figures 3(a) and (b) show the energy eigenvalues \( E_{\nu,\sigma}(k) \) near the Fermi energy \( \nu = 1, 2 \) in the TI \( (\lambda = 0) \) and TMI \( (\lambda = 0.5) \) states, respectively, with SOC. The energy gap in the TI state is approximately 2 meV as shown in the inset of Fig. 3(a). The Berry curvature \( B_{\nu,\sigma}(k) \) in the TMI state with SOC \( \lambda = 0.5 \) is plotted in Figure 5(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 becomes finite 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 of \( B_{\nu,\sigma}(k) \) are the almost same as those in the TI state [41].

**B. Stability of the TMI state in the presence of Coulomb interaction**

Next, to investigate the relationship between the stability of the TMI state in \( \alpha-\text{(BETS)}_2\text{I}_3 \) and the values of the N.N. and N.N.N. interactions, we consider the N.N. and N.N.N. Coulomb interactions by parameter \( V \) and \( V' \), respectively, and draw the phase diagram. Throughout this subsection, the onsite Coulomb interaction \( U \) and \( T \) are fixed at \((U,T) = (0.5, 1 \times 10^{-4})\), unless otherwise stated.

We first draw the \( V-V' \) phase diagram. The calculation result is shown in Fig. 6. \( V'C \) indicates the value of \( V' \) at which ZGS to TMI phase transition occurs when SOC is absent. \( V'^{D} \) is defined as the value of \( V' \) at which the Onsager phase factor \( \gamma \) without SOC becomes 0.25, as described below. In Fig. 6, it can be observed that the CO state appears in \( V > 0.12 \) and \( V' < V'^{CO} \) (upper-left region). However, with the increase in \( V' \), the TMI state is stabilized above \( V' > V'^{CO} \) and \( V' > V'^{C} \) or \( V'^{D} \). This indicates that \( V' \) plays a significant role in stabilizing the TMI state. As indicated in the list of Coulomb interactions in Fig. 2, \( V'/V \) is expected to be large in \( \alpha-\text{(BETS)}_2\text{I}_3 \). This tendency favors the realization of the TMI state.

Next, we calculate the phase of order parameter \( \varphi = \tan^{-1} \text{Im}[\delta b] / \text{Re}[\delta b] \) in the unit cell and investigate the possibility of a local magnetic field caused by the TMI state. Figures 7(a) and (b) show the schematic of the unit cell of \( \alpha-\text{(BETS)}_2\text{I}_3 \) and the loop patterns to calculate the summation of \( \varphi \). These loops include only N.N. and N.N.N. bonds. The \( V' \)-dependence of the summation of phases in each loop of the unit cell \( \varphi_n \) \((n = 1, \cdots, 10)\) at \( V = 0.18 \).
are plotted in Figs. 3(c) and (d). The sign of $\varphi_n$ depends on the spin degrees-of-freedom when SOC is considered. 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 including only N.N. bonds) and $\varphi_7 = -\varphi_{10}$, $\varphi_8 = -\varphi_9$ (loops including N.N. and N.N.N. bonds). Therefore, the TMI state due to $V'$ and SOC in this study differs from a current state suggested in cuprates superconductors (TMI to TMI) in which the summation of the phases in the unit cell is finite and there is a net current.

The $V'$-dependence of the absolute value of the Onsager phase factor $|\gamma|$ at $V = 0.1$ is plotted in Fig. 8. Here, $V'^D$ is defined as the value at which $|\gamma|$ without SOC becomes 0.25. The value of $|\gamma|$ becomes zero as $V'$ decreases, and increases to 0.5 when $V'$ is sufficiently large. This behavior is consistent with previously reported experimental results in which the phase factor changes from 0.5 to zero as the pressure $P$ decreases, where the $P$-dependence can be considered as the change in $\gamma$ associated with the change in $P$. $|\gamma|$ does not become zero even when the system is in the TMI phase because of the following. The Berry phase $\phi_B$ is calculated by the surface integral of the Berry curvature on Fermi surface $S_F$ as shown in Eq. (8). In a massive Dirac electron system such a TMI, the peak of the Berry curvature $B_{\nu,\sigma}(k)$ decreases with the increase in the energy gap $\Delta$, and $B_{\nu,\sigma}(k)$ widens and spreads in the Brillouin zone as shown in Fig. 5(b). Therefore, when the spread of $B_{\nu,\sigma}(k)$ becomes sufficiently larger than the integral range $S_F$ with the increase in $\Delta$ due to $V'$ (see Figs. 5(b) and (c)), $\phi_B$ decreases and becomes zero ($|\gamma|$ becomes 0.5).

Finally, we investigated the $T$-dependence of the electronic state with HF approximation and calculated the DC conductivity using $T$-matrix approximation. Figure 9(a) shows the $T$-dependence of the energy gap $\Delta$ at $(V, V') = (0.1, 0.16)$. With SOC and without Coulomb interaction, $\Delta$ is constant, ($\Delta \approx 0.002$) for $0 < T < 0.03$. This small gap is considered as the energy gap in the TMI state of $\alpha$-(BETS)$_2$I$_3$ [32]. On the other hand, when considering $U$, $V$, and $V'$ in the Hamiltonian without SOC, the TMI state in which order parameter $\text{Im}[\delta]$ has a finite value appears, and $\Delta$ increases sharply below the critical temperature $T_c$ due to ZGS to TMI phase transition. When both SOC and Coulomb interaction are considered, $\Delta \sim 0.01$ at $T = 0.03$, which is approximately five times that of the case without Coulomb interaction. On decreasing $T$, $\Delta$ gradually increases toward $T_c$ due to the effect of TI to TMI crossover and has a constant value $\Delta \sim 0.038$, which is twice that of the case without SOC. This result indicates that the contribution of SOC and $V'$ renders the TI state as well as TMI states more stable.
FIG. 8. (Color online) (a) $V'$-dependence of the absolute value of the Onsager phase factor $|\gamma|$ at $(U,V,T) = (0.5,0.1,10^{-4})$. $V'^D$ is defined as $V'$ at which the absolute value of $|\gamma|$ without SOC becomes 0.25. (b) and (c) Illustration of the Berry curvature $B_{\nu\sigma}(k)$ and energy band $E_{\nu\sigma}(k)$ near the Dirac point for two $\Delta$ values: $\Delta \ll |E_F|$ and $\Delta > |E_F|$. In Figs. 8 (b) and (c), the $T$-dependence of the DC conductivity $\sigma_{\eta}/\sigma_0$ in units of the universal conductivity $\sigma_0 = 4e^2/\pi h$ at $(U,V,V') = (0.5,0.1,0.16)$ when (b) SOC is not considered and (c) SOC is considered.

FIG. 9. (Color online) (a) $T$-dependence of the energy gap $\Delta$ at $(U,V,V') = (0.5,0.1,0.16)$. $T$-dependence of the DC conductivity $\sigma/\sigma_0$ in units of the universal conductivity $\sigma_0 = 4e^2/\pi h$ at $(U,V,V') = (0.5,0.1,0.16)$ when (b) SOC is not considered and (c) SOC is considered.

FIG. 10. (Color online) Illustration of the $T$-dependence of the electronic state obtained by calculation with HF approximation, with onsite $U$, N.N. sites $V$, and N.N.N. sites $V'$. with the decrease in $T$ reflecting the gentle $T$-dependence of the energy gap due to crossover.

To summarize this subsection, we presented the $T$-dependence of the electronic state obtained with HF approximation (Fig. 10). As $T$ is increased, TMI to TI crossover occurs, and a TI state with small gap due to SOC appears at high-$T$. The TMI state, which is mainly caused by the N.N. interaction $V'$ and SOC, is a strong candidate of the insulating state on $\alpha$-(BETS)$_2$I$_3$ at low-$T$. 
C. Spin fluctuations in the high-$T$ Dirac electron phase

In this subsection, we calculate the spin susceptibility using RPA and discuss the relationship with NMR in the high-$T$ TI state. Previous studies on $\alpha$-(ET)$_2$I$_3$ [23–25] have shown that the ferrimagnetic (FM) spin fluctuation observed in the site-resolved Knight shift [24] is induced by $U$, where only the site-resolved Knight shift at B site $K_B$, defined in Eq. (24), 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 inter-band electron-hole excitation enhanced by $U$ [23–24]. In this study, we investigate the possibility of FM spin fluctuation in $\alpha$-(BETS)$_2$I$_3$ [21].

Figures 11(a)-(c) show the $T$-dependence of $K_\alpha$ at the $\alpha = A, B$, and $C$ sites obtained using RPA at $U = 0.13$ (thick line) and $U = 0$ (thin line) as an example. As RPA overestimates the magnitude of the Coulomb interaction, we consider the $U$ value, which is smaller than those used in previous subsections ($U < 0.5$) for comparison with the experimental results. As shown 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 $\chi^{S}_{A,\beta}$ and $\chi^{S}_{C,\beta}$ [$\beta = A, A', B$, 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 $\chi^{S}_{B,\text{Intra}}$, $\chi^{S}_{B,\text{Inter}}$, and $\chi^{S}_{B}$ are also plotted in the inset of Fig. 11(c). It is indicated that $\chi^{S}_{B,\text{Inter}}$ becomes negative for $0 < T < 0.01$, and causes $\chi^{S}_{B}$ to become negative. This behavior is qualitatively similar to that observed in $\alpha$-(ET)$_2$I$_3$ [24]. This is because $\alpha$-(BETS)$_2$I$_3$ at high-$T$ has a characteristic wavenumber dependence of the square of the absolute value of the eigenvector such a zero line for B and C sites, which is similar to that of $\alpha$-(BEDT-TTF)$_2$I$_3$ under high pressure.

Next, we calculated spin susceptibility considering stronger interaction and investigated the kind of spin susceptibility that was enhanced. Figures 12(a) and (b) show the momentum $q$-dependence of the spin susceptibility $\chi^{S}_{A,B}$ at $\omega = 0$ for $\beta = A, A'$ with $(U, T) = (0.46, 0.007)$. In the strong interaction case, $\chi^{S}_{B}$ exhibits a peak at $q = 0$ reflecting the Fermi point in the Dirac electron system.
We considered the AF spin fluctuation occurring at strong Coulomb interaction. It can be observed that the FM spin fluctuation is dominant, and the AF spin fluctuation occurs at weak Coulomb interaction. For a case weak interaction, when $U$ decreased, only ferrimagnetic spin fluctuation appeared owing to the characteristic wave function of $\alpha$-(BETS)$_2$I$_3$. This behavior is similar to the ferrimagnetic spin fluctuation observed in $\alpha$-(BEDT-TTF)$_2$I$_3$. While for a case strong interaction, the antiferromagnetic spin fluctuation is enhanced. In this case, the components of sites A-A and A-A' of the spin susceptibility at $q = 0$ diverged positively and negatively with the decrease in $T$. This divergence corresponds to the emergence of the SMD phase as suggested in a previous study [11], but is not consistent with the results of NMR experiments [39, 40].

Finally, the effect of spin fluctuations on the NMR properties were investigated using the transfer integrals with SOC obtained by first-principles calculations and RPA with $U$. For a case weak interaction, when $T$ decreased, only ferrimagnetic spin fluctuation appeared owing to the characteristic wave function of $\alpha$-(BETS)$_2$I$_3$. This behavior is similar to the ferrimagnetic spin fluctuation observed in $\alpha$-(BEDT-TTF)$_2$I$_3$. While for a case strong interaction, the antiferromagnetic spin fluctuation is enhanced. In this case, the components of sites A-A and A-A' of the spin susceptibility at $q = 0$ diverged positively and negatively with the decrease in $T$. This divergence corresponds to the emergence of the SMD phase as suggested in a previous study [11], but is not consistent with the results of NMR experiments [39, 40].

In our calculations, the contribution of $V'$ to the emergence of the TMI state was significant. Recent studies have reported that the TMI state induced by Coulomb interaction appears even in real materials such as digital transition metal oxide hetero structures [73–76]. $\alpha$-(BETS)$_2$I$_3$ is also considered to be a system in which the TMI state due to $V'$ and SOC appears by the calculation with HF approximation. In the TMI state, the total phase shift in the unit cell becomes zero; hence, this state is different from a current state suggested in cuprates superconductors [69, 70] in which the summation of the

IV. SUMMARY AND DISCUSSION

In this study, first-principles calculation was first performed to construct an extended Hubbard model with transfer integrals considering SOC, and onsite, N.N. site, and N.N.N. site Coulomb interactions ($U, V, V'$). As $\alpha$-(BETS)$_2$I$_3$ has the Dirac cone near the Fermi energy and does not have Fermi surface, the long-range components of Coulomb interaction are expected to survive even when the screening effect is considered [42, 44]. In fact, we confirmed the value of $V'$ is the almost the same order as $V$ in $\alpha$-(BETS)$_2$I$_3$. Next, the electronic state in the low-$T$ insulating state was investigated using HF approximation with Coulomb interaction obtained by RESPACK, and the TMI state was found as one of the candidate states without charge, spin, and bond orders. Furthermore, for investigating the stability of the TMI state against the change in $V$ and $V'$, we draw the $V$-$V'$ phase diagram and found that the TMI state was stabilized by the contribution of $V'$ and that the energy gap due to the TMI state was enhanced by the contribution of $V'$ and SOC. Moreover, the Onsager phase factor was calculated based on the semiclassical theory [63, 68] and compared with a recent experiment [37]. Considering the pressure dependence as the change in $V'$ associated with the change in pressure, we showed that the Onsager phase factor changed from 0.5 to zero at low-$T$ due to the spread of the Berry curvature in the wavenumber space in a massive Dirac electron system and slight shift of the Fermi energy due to carrier doping. This behavior is consistent with the recent Shubnikov–de Haas oscillation experiment [37]. In addition, using $T$-matrix approximation and the Nakano-Kubo formula, we showed that when the insulating state at low-$T$ is assumed to be the TMI state, the experimental result [36, 37, 72] where the DC conductivity decreases sharply at $T < 50 K$ can be understood without any contradiction.

Finally, the effect of spin fluctuations on the NMR properties were investigated using the transfer integrals with SOC obtained by first-principles calculations and RPA with $U$. For a case weak interaction, when $T$ decreased, only ferrimagnetic spin fluctuation appeared owing to the characteristic wave function of $\alpha$-(BETS)$_2$I$_3$. This behavior is similar to the ferrimagnetic spin fluctuation observed in $\alpha$-(BEDT-TTF)$_2$I$_3$. While for a case strong interaction, the antiferromagnetic spin fluctuation is enhanced. In this case, the components of sites A-A and A-A' of the spin susceptibility at $q = 0$ diverged positively and negatively with the decrease in $T$. This divergence corresponds to the emergence of the SMD phase as suggested in a previous study [11], but is not consistent with the results of NMR experiments [39, 40].

In our calculations, the contribution of $V'$ to the emergence of the TMI state was significant. Recent studies have reported that the TMI state induced by Coulomb interaction appears even in real materials such as digital transition metal oxide hetero structures [73–76]. $\alpha$-(BETS)$_2$I$_3$ is also considered to be a system in which the TMI state due to $V'$ and SOC appears by the calculation with HF approximation. In the TMI state, the total phase shift in the unit cell becomes zero; hence, this state is different from a current state suggested in cuprates superconductors [69, 70] in which the summation of the
phases in the unit cell is finite and there is a net current. A recent NMR experiment reported that time reversal symmetry breaking was not observed, and $1/T_1 T$ was proportional to the power of $T$ and varied continuously near 50 K \cite{10}. In our calculation using RPA, ferrimagnetic spin fluctuations have no significant effect on the $T$ near 50 K \cite{40}. In our calculation using RPA, ferrimagnetic spin fluctuations have no significant effect on the $T$ near 50 K \cite{40}. The ferrimagnetic spin polarization has been observed by a site-resolved NMR experiment in $\alpha$-(BEDT-TTF)$_2$I$_3$ \cite{24} and $\alpha$-(BETS)$_2$I$_3$ \cite{71}.

In future research, the effect of the TMI state 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 TI state is affected by orbital current and that the power of $T$ changes \cite{71, 78}. It would be interesting to investigate the presence or absence of similar effects in the TMI state of $\alpha$-(BETS)$_2$I$_3$. In recent studies on the TI state, an exotic state localized only at the intersection of the edges of the TI state has been reported, and such materials are called higher-order topological Mott insulators \cite{73, 79, 86, 80, 81}. Future research should investigate whether the TMI state suggested in this study is a higher-order topological Mott insulator, 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 \cite{72}. Finally, there are many unclear points on the low-$T$ insulating state of $\alpha$-(BETS)$_2$I$_3$; hence, it is possible that mean-field approximation is not a sufficient calculation method. Calculations considering electron correlation effects using vertex correction \cite{87}, variational Monte Carlo method \cite{88}, and functional renormalization group theory \cite{69, 70} will also be addressed in future.

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