Predicted photoinduced topological phases in organic salt α -(BEDT-TTF)₂I₃

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The emergence of photoinduced topological phases and their phase transitions is theoretically predicted in organic salt α -(BEDT-TTF)₂I₃, which possesses inclined Dirac cones in its band structure. By analyzing a photodriven tight-binding model describing conduction electrons in the BEDT-TTF layer using the Floquet theorem, we demonstrate that irradiation with circularly polarized light opens a gap at the Dirac points, and the system eventually becomes a Chern insulator characterized by a quantized topological invariant. A rich phase diagram is obtained in the plane of amplitude and frequency of light, which contains Chern insulator, semimetal, and normal insulator phases. We find that the photoinduced Hall conductivity provides a sensitive means to detect the predicted phase evolutions experimentally. This work contributes towards developing the optical manipulation of electronic states in matter through broadening the range of target materials that manifest photoinduced topological phase transitions.

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I. INTRODUCTION

Photoinduced phase transitions are one of the central topics in recent condensed-matter physics [1–5]. A theoretical study using the Floquet theorem has predicted that the honeycomb lattice irradiated with circularly polarized light attains a topological band structure similar to the band structure originally proposed by Haldane [6] that exhibits a topological phase transition. This transition results in the photoinduced quantum Hall effect in graphene, even in the absence of an external magnetic field [7,8], and was indeed confirmed in a recent experiment [9]. Since this pioneering theoretical work, the Floquet theory has been applied to various electron systems and has revealed many interesting photoinduced topological phenomena [10–29]. The topic is now attracting enormous research interest from the viewpoint of both fundamental science and electronics applications.

However, most of the previous research has dealt with tight-binding models on simple lattices such as the honeycomb lattice [7,8,10], the Kagome lattice [14,23], the Lieb lattice [14], and the stacked graphene systems [15] or simple materials with a two-dimensional atomic layer such as graphene [7,8,10], silicene [16], black phosphorene [17] and transition-metal dichalcogenides [18,19]. There have been few studies based on realistic models for specific materials. In addition, the only successful experiment so far was done for graphene [9], which has a simple electronic structure described well by the tight-binding model on honeycomb lattice [6,30]. However, to further develop this promising

research field, widening the range of target materials is indispensable, and towards this objective, theoretical studies on actual materials with complex electronic and crystalline structures are highly desired. Moreover, we can expect richer material-specific photoinduced topological phenomena in studies on actual materials. One promising material is an organic salt α -(BEDT-TTF)₂I₃, where BEDT-TTF denotes bis(ethylenedithio)-tetrathiafulvalene [31]. This compound has inclined Dirac cones in its band structure [32–34], and many interesting topological properties and phenomena arising from these Dirac-cone bands have been theoretically investigated so far, e.g., the quantum Hall effect [32], the structures of Berry curvature in momentum space [35], and the flux-induced Chern insulator phases [36].

In this paper, we theoretically predict the emergence of photoinduced topological phases and their phase transitions in α -(BEDT-TTF)₂I₃. By applying the Floquet theory to the photodriven tight-binding model for conduction electrons in the BEDT-TTF layer, we demonstrate that the inclined Dirac cones become gapped at the Dirac points by irradiation with circularly polarized light [see Fig. 1(a)]. The system then becomes a topological insulator characterized by a quantized Chern number [37,38] and conductive chiral edge states [39]. We obtain a rich phase diagram in the plane of the amplitude and frequency of the light that contains phases corresponding to a Chern insulator, semimetal, and normal insulator. The calculated photoinduced Hall conductivity shows characteristic dependencies on the light amplitude and temperature in each phase, indicating that this quantity provides a sensitive experimental indicator in the detection and identification of these topological phases and their phase transitions. One advantage of using organic compounds is that an effective amplitude of light is an order of magnitude larger than graphene because their lattice constants are much larger (to be discussed later). They enhance the feasibility of the experiments. This work contributes to the development of

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FIG. 1. (a) Schematics of α -(BEDT-TTF)₂I₃ irradiated with circularly polarized light. (b) Unit cell (dashed rectangle) with four nonequivalent BEDT-TTF molecules (A, A', B, C) and transfer integrals in the BEDT-TTF layer.

this field by expanding the potential range of materials for research.

II. MODEL AND METHOD

We employ a tight-binding model to describe the electronic structure of α -(BEDT-TTF)₂I₃ [32,33], which is given by

$$H = \sum_{i,j} \sum_{\alpha,\beta} t_{i\alpha,j\beta} c_{i\alpha}^\dagger c_{j\beta}. \quad (1)$$

The unit cell of the BEDT-TTF layer contains four molecular sites (A, A', B, C), and we consider transfer integrals $t_{i\alpha,j\beta}$ among them [Fig. 1(b)], where *i* and *j* label the unit cells, whereas α and β label the molecular sites. Under ambient pressure, this compound exhibits a charge-ordered ground state [31]. When a uniaxial pressure is applied along the *a* axis, this charge order melts and eventually inclined Dirac cones emerge within its peculiar band structure. In this study, we consider the latter situation by taking the transfer integrals evaluated theoretically at uniaxial pressure P_a of 4 kbar; specifically, $t_{a1} = -0.038$ eV, $t_{a2} = 0.080$ eV, $t_{a3} = -0.018$ eV, $t_{b1} = 0.123$ eV, $t_{b2} = 0.146$ eV, $t_{b3} = -0.070$ eV, and $t_{b4} = -0.025$ eV [40].

After the Fourier transformations with respect to the spatial coordinates, the tight-binding Hamiltonian is rewritten in the momentum space as

$$H = \sum_{\mathbf{k}} (c_{\mathbf{k}A}^\dagger \ c_{\mathbf{k}A'}^\dagger \ c_{\mathbf{k}B}^\dagger \ c_{\mathbf{k}C}^\dagger) \hat{H}(\mathbf{k}) \begin{pmatrix} c_{\mathbf{k}A} \\ c_{\mathbf{k}A'} \\ c_{\mathbf{k}B} \\ c_{\mathbf{k}C} \end{pmatrix}, \quad (2)$$

where

$$\hat{H}(\mathbf{k}) = \begin{pmatrix} 0 & A_2(\mathbf{k}) & B_2(\mathbf{k}) & B_1(\mathbf{k}) \\ A_2^*(\mathbf{k}) & 0 & B_2^*(\mathbf{k}) & B_1^*(\mathbf{k}) \\ B_2^*(\mathbf{k}) & B_2(\mathbf{k}) & 0 & A_1(\mathbf{k}) \\ B_1^*(\mathbf{k}) & B_1(\mathbf{k}) & A_1(\mathbf{k}) & 0 \end{pmatrix}, \quad (3)$$

with

$$A_1(\mathbf{k}) = 2t_{a1}(\mathbf{k}) \cos(k_y/2), \quad (4)$$

$$A_2(\mathbf{k}) = t_{a2}(\mathbf{k}) e^{ik_y/2} + t_{a3}(\mathbf{k}) e^{-ik_y/2}, \quad (5)$$

$$B_1(\mathbf{k}) = t_{b1}(\mathbf{k}) e^{i(k_x/2 + k_y/4)} + t_{b4}(\mathbf{k}) e^{-i(k_x/2 - k_y/4)}, \quad (6)$$

$$B_2(\mathbf{k}) = t_{b2}(\mathbf{k}) e^{i(k_x/2 - k_y/4)} + t_{b3}(\mathbf{k}) e^{-i(k_x/2 + k_y/4)}. \quad (7)$$

We then consider a situation in which this compound is irradiated with circularly polarized light applied perpendicular to the *ab* plane [Fig. 1(a)]. The applied circularly polarized light generates a vector potential $\mathbf{A}(\tau) = A(\cos \omega\tau, \sin \omega\tau)$, which corresponds to a time-dependent electric field:

$$\mathbf{E}(\tau) = -\frac{d\mathbf{A}}{d\tau} = A\omega(\sin \omega\tau, -\cos \omega\tau). \quad (8)$$

In the presence of this vector potential, the transfer integrals attain Peierls phases as,

$$\begin{aligned} t_{i\alpha,j\beta}(\tau) &= t_{i\alpha,j\beta} \exp \left[-i \frac{e}{\hbar} \mathbf{A}(\tau) \cdot (\mathbf{r}_{i\alpha} - \mathbf{r}_{j\beta}) \right] \\ &= t_{i\alpha,j\beta} \exp [i \{ \mathcal{A}_b(\tilde{x}_j - \tilde{x}_i) \cos \omega\tau + \mathcal{A}_a(\tilde{y}_j - \tilde{y}_i) \sin \omega\tau \}]. \end{aligned} \quad (9)$$

Here we introduce dimensionless quantities $\mathcal{A}_a = eAa/\hbar$ and $\mathcal{A}_b = eAb/\hbar$ and dimensionless coordinates $\mathbf{r}_{i\alpha} = (b\tilde{x}_{i\alpha}, a\tilde{y}_{i\alpha})$, with *a* and *b* being the lattice constants along the *y* and *x* axes, respectively [Fig. 1(b)]. We use experimental values $a = 0.9187$ nm and $b = 1.0793$ nm [41], which give a ratio $\mathcal{A}_b/\mathcal{A}_a = b/a = 1.175$. The amplitude of the ac electric field of light E^ω is given by $E^\omega = A\omega = \mathcal{A}_a \hbar \omega / ea$.

In the momentum representation, the effects of Peierls phases are taken into account by replacing the momenta k_x and k_y with $k_x + \mathcal{A}_b$ and $k_y + \mathcal{A}_a$, respectively. Then we obtain the time-dependent Hamiltonian for the photoirradiated α -(BEDT-TTF)₂I₃ as

$$\hat{H}(\mathbf{k}, \tau) = \begin{pmatrix} 0 & A_2(\mathbf{k}, \tau) & B_2(\mathbf{k}, \tau) & B_1(\mathbf{k}, \tau) \\ A_2^*(\mathbf{k}, \tau) & 0 & B_2^*(\mathbf{k}, \tau) & B_1^*(\mathbf{k}, \tau) \\ B_2^*(\mathbf{k}, \tau) & B_2(\mathbf{k}, \tau) & 0 & A_1(\mathbf{k}, \tau) \\ B_1^*(\mathbf{k}, \tau) & B_1(\mathbf{k}, \tau) & A_1(\mathbf{k}, \tau) & 0 \end{pmatrix}, \quad (10)$$

with

$$A_1(\mathbf{k}, \tau) = 2t_{a1} \cos \left(\frac{k_y}{2} + \frac{\mathcal{A}_a}{2} \sin \omega\tau \right), \quad (11)$$

$$\begin{aligned} A_2(\mathbf{k}, \tau) &= t_{a2} \exp \left[i \left(\frac{k_y}{2} + \frac{\mathcal{A}_a}{2} \sin \omega\tau \right) \right] \\ &\quad + t_{a3} \exp \left[-i \left(\frac{k_y}{2} + \frac{\mathcal{A}_a}{2} \sin \omega\tau \right) \right], \end{aligned} \quad (12)$$

$$\begin{aligned} B_1(\mathbf{k}, \tau) &= t_{b1} \exp \left[i \left(\frac{k_x}{2} + \frac{k_y}{4} \right) \right] \exp [i\mathcal{A} \sin(\omega\tau + \theta)] \\ &\quad + t_{b4} \exp \left[-i \left(\frac{k_x}{2} - \frac{k_y}{4} \right) \right] \exp [i\mathcal{A} \sin(\omega\tau - \theta)], \end{aligned} \quad (13)$$

$$\begin{aligned} B_2(\mathbf{k}, \tau) &= t_{b2} \exp \left[i \left(\frac{k_x}{2} - \frac{k_y}{4} \right) \right] \exp [-i\mathcal{A} \sin(\omega\tau - \theta)] \\ &\quad + t_{b3} \exp \left[-i \left(\frac{k_x}{2} + \frac{k_y}{4} \right) \right] \exp [-i\mathcal{A} \sin(\omega\tau + \theta)], \end{aligned} \quad (14)$$

where

$$\mathcal{A} = \frac{1}{4} \sqrt{4\mathcal{A}_b^2 + \mathcal{A}_a^2}, \quad \theta = \tan^{-1} \left(\frac{2\mathcal{A}_b}{\mathcal{A}_a} \right). \quad (15)$$

We analyze this time-periodic tight-binding Hamiltonian using the Floquet theory. The time-dependent Schrödinger equation is given by

$$i\hbar \frac{\partial}{\partial \tau} |\Psi(\mathbf{k}, \tau)\rangle = H(\mathbf{k}, \tau) |\Psi(\mathbf{k}, \tau)\rangle. \quad (16)$$

According to the Floquet theorem, the wave function $|\Psi(\tau)\rangle$ is written in the form

$$|\Psi(\mathbf{k}, \tau)\rangle = e^{-i\varepsilon\tau/\hbar} |\Phi(\mathbf{k}, \tau)\rangle, \quad (17)$$

where $|\Phi(\mathbf{k}, \tau)\rangle = |\Phi(\mathbf{k}, \tau + T)\rangle$. Here $T (= 2\pi/\omega)$ is the temporal period of the ac light field, and ε is the quasienergy. This equation is rewritten in the form

$$\sum_m \hat{\mathcal{H}}_{n,m}(\mathbf{k}) |\Phi_v^m(\mathbf{k})\rangle = \varepsilon_v^n(\mathbf{k}) |\Phi_v^n(\mathbf{k})\rangle \quad (18)$$

with

$$\hat{\mathcal{H}}_{n,m}(\mathbf{k}) \equiv \hat{H}_{n-m}(\mathbf{k}) - m\omega\delta_{n,m}\hat{1}_{4 \times 4}, \quad (19)$$

where v labels the eigenstates, and n corresponds to the number of photons. The four-component vector $|\Phi_v^n(\mathbf{k})\rangle$ represents the v th Floquet state ($v = 1, 2, 3, 4$) in the n -photon subspace. We introduce the following Fourier coefficients:

$$\begin{aligned} |\Phi_v^n(\mathbf{k})\rangle &= \frac{1}{T} \int_0^T |\Phi_v(\mathbf{k}, \tau)\rangle e^{in\omega\tau} d\tau \\ \hat{H}_n(\mathbf{k}) &= \frac{1}{T} \int_0^T \hat{H}(\mathbf{k}, \tau) e^{in\omega\tau} d\tau \\ &= \begin{pmatrix} 0 & A_{2,n}(\mathbf{k}) & B_{2,n}(\mathbf{k}) & B_{1,n}(\mathbf{k}) \\ A_{2,-n}^*(\mathbf{k}) & 0 & B_{2,-n}^*(\mathbf{k}) & B_{1,-n}^*(\mathbf{k}) \\ B_{2,-n}^*(\mathbf{k}) & B_{2,n}(\mathbf{k}) & 0 & A_{1,n}(\mathbf{k}) \\ B_{1,-n}^*(\mathbf{k}) & B_{1,n}(\mathbf{k}) & A_{1,n}(\mathbf{k}) & 0 \end{pmatrix}, \end{aligned} \quad (20)$$

with

$$A_{1,n}(\mathbf{k}) = t_{a1} e^{ik_y/2} J_{-n}(\mathcal{A}_a/2) + t_{a1} e^{-ik_y/2} J_n(\mathcal{A}_a/2), \quad (22)$$

$$A_{2,n}(\mathbf{k}) = t_{a2} e^{ik_y/2} J_{-n}(\mathcal{A}_a/2) + t_{a3} e^{-ik_y/2} J_n(\mathcal{A}_a/2), \quad (23)$$

$$\begin{aligned} B_{1,n}(\mathbf{k}) &= t_{b1} e^{i(k_x/2 + k_y/4)} J_{-n}(\mathcal{A}) e^{-in\theta} \\ &\quad + t_{b4} e^{-i(k_x/2 - k_y/4)} J_{-n}(\mathcal{A}) e^{+in\theta}, \end{aligned} \quad (24)$$

$$\begin{aligned} B_{2,n}(\mathbf{k}) &= t_{b2} e^{i(k_x/2 - k_y/4)} J_n(\mathcal{A}) e^{+in\theta} \\ &\quad + t_{b3} e^{-i(k_x/2 + k_y/4)} J_n(\mathcal{A}) e^{-in\theta}. \end{aligned} \quad (25)$$

We solve the eigenequation (18) by restricting the number of photons to $|m| \leq M_{\max}$ ($M_{\max} = 16$ throughout the present work). Consequently, the Floquet-Hamiltonian matrix $\hat{\mathcal{H}}(\mathbf{k})$ is composed of $(2M_{\max} + 1) \times (2M_{\max} + 1)$ block matrices $\hat{\mathcal{H}}_{n,m}(\mathbf{k}) \equiv \hat{H}_{n-m}(\mathbf{k}) - m\omega\delta_{n,m}\hat{1}_{4 \times 4}$. The total size of $\hat{\mathcal{H}}(\mathbf{k})$ is $(8M_{\max} + 4) \times (8M_{\max} + 4)$ (132×132 in the present work), because the size of each block matrix is 4×4 . Note that as the frequency ω is reduced, a Floquet matrix of larger size $|m|$ is required, typically of order $W/\hbar\omega$, where W is the

bandwidth. Having adopted $|m| \leq 16$ for α -(BEDT-TTF)₂I₃ with $W \sim 0.8$ eV, the obtained results are sufficiently accurate for $\hbar\omega \gtrsim 0.05$ eV.

The Chern number of the v th band N_{Ch}^v ($v = 1, 2, 3, 4$) is related to the Berry curvature $B_z^{nv}(\mathbf{k})$,

$$N_{\text{Ch}}^v = \frac{1}{2\pi} \iint_{\text{BZ}} B_z^{nv}(\mathbf{k}) dk_x dk_y, \quad (26)$$

where the Berry curvature $B_z^{nv}(\mathbf{k})$ of the v th n -photon Floquet band at each $\mathbf{k}$ point is given by

$$\begin{aligned} B_z^{nv}(\mathbf{k}) &= \\ i \sum_{(m,\mu)} \frac{\langle \Phi_v^n(\mathbf{k}) | \frac{\partial \mathcal{H}}{\partial k_x} | \Phi_\mu^m(\mathbf{k}) \rangle \langle \Phi_\mu^m(\mathbf{k}) | \frac{\partial \mathcal{H}}{\partial k_y} | \Phi_v^n(\mathbf{k}) \rangle - \text{c.c.}}{[\varepsilon_\mu^m(\mathbf{k}) - \varepsilon_v^n(\mathbf{k})]^2}. \end{aligned} \quad (27)$$

Here $\mathcal{H}$ denotes the matrix of the Floquet Hamiltonian, and $\varepsilon_v^n(\mathbf{k})$ and $|\Phi_v^n(\mathbf{k})\rangle$ the eigenenergy and eigenvector of Eq. (18), with $v = 1, 2, 3, 4$ and $|n| \leq 16$. The summation is taken over m and μ , where $(m, \mu) \neq (n, v)$; “c.c.” denotes the complex conjugate of the first term of the numerator. In this work, the Chern numbers are calculated using a numerical technique proposed by Fukui *et al.* in Ref. [42].

III. RESULTS

We first discuss the photoinduced variation of band structures and their topological properties. Figure 2(a) shows the band dispersions for the third and fourth bands, $E_3(\mathbf{k})$ and $E_4(\mathbf{k})$, in the absence of photoirradiation. These two bands make contact at two individual points in momentum space to form a pair of inclined Dirac cones. Note that the Dirac points are located at the Fermi level because this compound has a 3/4 electron filling with three fully occupied lower bands and an unoccupied fourth band. Figures 2(b)–2(d) show plots of $E_3(\mathbf{k})$ and $E_4(\mathbf{k})$ for photoirradiated systems with various E^ω and ω of light. Once the system is irradiated with circularly polarized light, a gap opens at the Dirac points. These three band structures correspond to three different photoinduced phases characterized by the Chern number N_{Ch} and the band gap E_{gap} , that is, (b) Chern insulator, (c) semimetal, and (d) normal insulator phases, respectively.

Here the band gap is defined by $E_{\text{gap}} = \min[E_4(\mathbf{k})] - \max[E_3(\mathbf{k})]$, where $\min[E_4(\mathbf{k})]$ and $\max[E_3(\mathbf{k})]$ are the minimum energy of the fourth band and the maximum energy of the third band, respectively. A positive E_{gap} means that the bulk is insulating, for which, in the whole momentum space, the fourth band is located above the Fermi level, whereas the other three bands are located below the Fermi level. In contrast, a negative E_{gap} means that the bulk is semimetallic, for which the third band is located above the Fermi level whereas the fourth band is located below the Fermi level in some portion of the momentum space. Importantly, when the electron filling is 3/4 with three fully occupied lower bands, a sum of the Chern numbers over three bands below the Fermi level, $N_{\text{Ch}} = \sum_{v=1}^3 N_{\text{Ch}}^v$, coincides with $-N_{\text{Ch}}^4$, because conservation of invariance requires the sum of the Chern numbers over all four bands to be zero.

The band structure in Fig. 2(b), characterized by $E_{\text{gap}} > 0$ and $N_{\text{Ch}} = -N_{\text{Ch}}^4 = +1$, is assigned to the Chern insulator

FIG. 2. (a) Band dispersions of the third and fourth bands, $E_3(\mathbf{k})$ and $E_4(\mathbf{k})$, before light irradiation. (b)–(d) Those for the photoinduced phases under irradiation with circularly polarized light, i.e., (b) Chern insulator, (c) semimetal, and (d) normal insulator phases. (e, f) Berry curvatures of the fourth band $B_z^{04}(\mathbf{k})$ in momentum space for (e) the photoinduced Chern insulator phase with $N_{\text{Ch}}^4 = -1$ and (f) the photoinduced normal insulator phase with $N_{\text{Ch}}^4 = 0$.

phase. In contrast, the band structure in Fig. 2(c) is characterized by $E_{\text{gap}} < 0$, for which the fourth band around $\mathbf{k} = (\pm\pi, 0)$ is lower in energy than the third band around the Dirac points. This band structure is assigned to the semimetal phase. Interestingly, the band structure in Fig. 2(d) has $E_{\text{gap}} > 0$ and resembles the band structure for the Chern insulator [Fig. 2(b)]. However, the Chern number $N_{\text{Ch}} = -N_{\text{Ch}}^4$ is zero in this case, indicating that the system lies in a topologically trivial insulating state. Therefore, this band structure is assigned to the normal insulator phase. Note that these phases appear as nonequilibrium steady states under the continuous application of circularly polarized light, and, in this sense, they are distinct from thermodynamically equilibrium phases.

We find a clear difference between the Chern insulator phase and the normal insulator phase in the Berry curvature $B_z^{04}(\mathbf{k})$. The Berry curvature in the Chern insulator phase has two negative peaks around the gapped Dirac points [Fig. 2(e)], corresponding to a nonzero quantized Chern number N_{Ch}^4 of -1 , whereas that for the normal insulator phase has additional positive peaks as well as two negative peaks around the gapped Dirac points [Fig. 2(f)] that cancel the opposite

FIG. 3. (a) Phase diagram of photodriven α -(BEDT-TTF) $_2$ I $_3$ in the plane of the amplitude E^ω and frequency ω of the applied circularly polarized light. (b, c) Color maps of (b) the Chern number of the highest (fourth) band N_{Ch}^4 and (c) the band gap E_{gap} in the plane of E^ω and ω .

contributions, resulting in $N_{\text{Ch}}^4 = 0$. Figure 3(a) shows the phase diagram of photodriven α -(BEDT-TTF) $_2$ I $_3$ in the plane of the amplitude E^ω and frequency ω of an applied circularly polarized light. Three phases are present, namely, the Chern insulator, semimetal, and normal insulator phases. This phase diagram is produced by calculating the Chern number of the (highest) fourth band N_{Ch}^4 [Fig. 3(b)] and the band gap E_{gap} [Fig. 3(c)]. We assign the area with $E_{\text{gap}} > 0$ and $N_{\text{Ch}}^4 \neq 0$ to the Chern insulator phase, and the area with $E_{\text{gap}} > 0$ and $N_{\text{Ch}}^4 = 0$ to the normal insulator phase. The area with $E_{\text{gap}} < 0$ is assigned to the semimetal phase. According to the obtained phase diagram, we expect that the usage of high-frequency light with $\hbar\omega > 0.75$ eV is preferable to observe the photoinduced Chern insulator phase in the low- E^ω range, whereas the usage of low-frequency light with $\hbar\omega < 0.7$ eV is suitable to observe the rich phase transitions upon the variation of E^ω .

Finally, we discuss the Hall conductivity in the three different photoinduced electronic phases. This physical quantity is closely related to the topological nature of the electronic states and can be exploited to identify the predicted topological phases and to detect their phase transitions. We plot the calculated Chern number $-N_{\text{Ch}}^4$ and band gap E_{gap} [Fig. 4(a)] as functions of the amplitude E^ω of the applied circularly polarized light setting $\hbar\omega = 0.5$ eV, for which the three photoinduced phases, semimetal, normal insulator, and Chern insulator phases, emerge successively as E^ω increases. Recall that the relation $N_{\text{Ch}} = -N_{\text{Ch}}^4$ holds when the electron filling

FIG. 4. (a) Chern number $-N_{\text{Ch}}^4$ and band gap E_{gap} plotted as a function of the light amplitude E^ω when $\hbar\omega = 0.5$ eV, which manifests successive emergence of three photoinduced electronic phases with increasing E^ω . (b) E^ω profiles of the Hall conductivity σ_{xy} for various temperatures when $\hbar\omega = 0.5$ eV.

is $3/4$. We also plot the calculated E^ω profiles of the Hall conductivity σ_{xy} for various temperatures when $\hbar\omega = 0.5$ eV [Fig. 4(b)]. The Hall conductivity σ_{xy} is calculated using the relation

$$\sigma_{xy} = \frac{2e^2}{h} \iint_{\text{BZ}} \frac{dk_x dk_y}{2\pi} \sum_{n,v} n_{nv}(\mathbf{k}) B_z^{nv}(\mathbf{k}), \quad (28)$$

where the factor 2 accounts for the spin degeneracy. Here $n_{nv}(\mathbf{k})$ is the nonequilibrium distribution function, which describes the electron occupations of Floquet bands in the photodriven nonequilibrium steady states.

The nonequilibrium distribution function $n_{nv}(\mathbf{k})$ is calculated using the Floquet-Keldysh formalism [5,43], which is formulated by combining the Keldysh Green's function technique [44,45] with the Floquet theory. The Dyson equation for the Green's function matrices is given by

$$\begin{pmatrix} \hat{G}^R(\mathbf{k}, \varepsilon) & \hat{G}^K(\mathbf{k}, \varepsilon) \\ 0 & \hat{G}^A(\mathbf{k}, \varepsilon) \end{pmatrix}^{-1} = \begin{pmatrix} [\hat{G}^{R0}(\mathbf{k}, \varepsilon)]^{-1} & 0 \\ 0 & [\hat{G}^{A0}(\mathbf{k}, \varepsilon)]^{-1} \end{pmatrix} - \begin{pmatrix} \hat{\Sigma}^R & \hat{\Sigma}^K(\varepsilon) \\ 0 & \hat{\Sigma}^A \end{pmatrix}. \quad (29)$$

Here $\hat{G}^R$, $\hat{G}^A$, and $\hat{G}^K$ ($\hat{\Sigma}^R$, $\hat{\Sigma}^A$ and $\hat{\Sigma}^K$) are matrices of the retarded, advanced, and Keldysh Green's functions (self-energies), respectively, each of which is composed of $(2M_{\text{max}} + 1) \times (2M_{\text{max}} + 1)$ block matrices where the size of each block matrix is 4×4 . The matrix components of $\hat{G}^{R0}$, $\hat{G}^{A0}$, $\hat{\Sigma}^R$, $\hat{\Sigma}^A$, and $\hat{\Sigma}^K$ are given, respectively, by

$$[\hat{G}^{R0}(\mathbf{k}, \varepsilon)]_{nv,m\mu}^{-1} = \varepsilon \delta_{n,m} \delta_{v,\mu} - \mathcal{H}_{nv,m\mu}(\mathbf{k}), \quad (30)$$

$$[\hat{G}^{A0}(\mathbf{k}, \varepsilon)]_{nv,m\mu}^{-1} = \varepsilon \delta_{n,m} \delta_{v,\mu} - \mathcal{H}_{nv,m\mu}(\mathbf{k}), \quad (31)$$

$$[\hat{\Sigma}^R]_{nv,m\mu} = -i\Gamma \delta_{n,m} \delta_{v,\mu}, \quad (32)$$

$$[\hat{\Sigma}^A]_{nv,m\mu} = i\Gamma \delta_{n,m} \delta_{v,\mu}, \quad (33)$$

$$[\hat{\Sigma}^K(\varepsilon)]_{nv,m\mu} = -2i\Gamma \tanh \left[\frac{\varepsilon - \mu + m\omega}{2k_B T} \right] \delta_{n,m} \delta_{v,\mu}, \quad (34)$$

where the symbol $\hat{M}_{nv,m\mu}$ denotes the (v, μ) th component of the (m, n) th block matrix $\hat{M}_{nm}$ (4×4), and the block matrix $\hat{\mathcal{H}}_{n,m}$ constituting the Floquet Hamiltonian is given by Eq. (19). We consider a situation where the system is coupled to a heat reservoir at temperature T with a dissipation coefficient Γ where we set $\Gamma = 0.1$ eV for the calculations. The lesser Green's function $\hat{G}^<$ and the lesser self-energy $\hat{\Sigma}^<$ are calculated, respectively, by

$$\hat{G}^<(\mathbf{k}, \varepsilon) = \hat{G}^R(\mathbf{k}, \varepsilon) \hat{\Sigma}^<(\varepsilon) \hat{G}^A(\mathbf{k}, \varepsilon), \quad (35)$$

$$\hat{\Sigma}^<(\varepsilon) = (\hat{\Sigma}^A + \hat{\Sigma}^K(\varepsilon) - \hat{\Sigma}^R)/2. \quad (36)$$

The matrix components of the lesser self-energy $\hat{\Sigma}^<$ read

$$[\hat{\Sigma}^<(\varepsilon)]_{nv,m\mu} = i\Gamma \left(1 - \tanh \left[\frac{\varepsilon - \mu + m\omega}{2k_B T} \right] \right) \delta_{n,m} \delta_{v,\mu}. \quad (37)$$

The nonequilibrium distribution function $n_{nv}(\mathbf{k})$ for the v th Floquet band in the n -photon subspace is given by

$$n_{nv}(\mathbf{k}) = \frac{\langle \Phi_v^n(\mathbf{k}) | \hat{N}_k(\varepsilon_v^n(\mathbf{k})) | \Phi_v^n(\mathbf{k}) \rangle}{\langle \Phi_v^n(\mathbf{k}) | \hat{A}_k(\varepsilon_v^n(\mathbf{k})) | \Phi_v^n(\mathbf{k}) \rangle}, \quad (38)$$

where

$$\hat{A}_k(\varepsilon) = i(\hat{G}^R(\mathbf{k}, \varepsilon) - \hat{G}^A(\mathbf{k}, \varepsilon))/2\pi, \quad (39)$$

$$\hat{N}_k(\varepsilon) = -i\hat{G}^<(\mathbf{k}, \varepsilon)/2\pi, \quad (40)$$

and $|\Phi_v^n(\mathbf{k})\rangle$ is the Floquet eigenstates obtained by solving Eq. (18).

In Fig. 4(b) we find that the Hall conductivity σ_{xy} is almost zero in the semimetal phase when E^ω is small ($E^\omega \lesssim 4$ MV/cm) but starts increasing with increasing E^ω at around $E^\omega \sim 4$ MV/cm towards the phase boundary to the normal insulator phase. It reaches a quantized value of $\sim 2e^2/h$ at the phase boundary and is kept constant in the normal insulator phase, although it is slightly decreased as temperature increases. This nearly constant behavior does not change even when the system enters the Chern insulator phase, in which the value of σ_{xy} is again nearly constant to be $\sim 2e^2/h$. The quantized Hall conductivity of $\sigma_{xy} \sim 2e^2/h$ in the Chern insulator phase is naturally understood from the quantized Chern number of $N_{\text{Ch}} = -N_{\text{Ch}}^4 = 1$. On the other hand, the finite σ_{xy} in the normal insulator phase is rather nontrivial because the Chern number vanishes ($N_{\text{Ch}} = 0$) in this phase. It is attributable to the formation and the electron occupations of Floquet subbands (a series of replicas of the original bands with an energy spacing of $\hbar\omega$) in the present photodriven system. Namely, when the light frequency is not large enough, as in the present case with $\hbar\omega = 0.5$ eV as compared to the bandwidth W (~ 0.8 eV in the present α -type organic salt), the hybridization and the anticrossing of Floquet bands

with different photon numbers occur. As a result, the highest Floquet band in the $m = -1$ subspace and the lowest Floquet band in the $m = +1$ subspace are partially occupied by electrons, and the Hall conductivity captures the Chern numbers of the Floquet bands in these finite-photon subspaces. This phenomenon cannot be expected in the usual normal insulator phase in equilibrium.

Because both the normal insulator phase and the Chern insulator phase exhibit nearly the same behaviors of the Hall conductivity, it may be difficult to distinguish these phases by the Hall conductivity measurements. However, the temperature dependence is more pronounced in the normal insulator phase. Hence, measurements of the precise temperature profiles of σ_{xy} may be exploited for identification of the phases. It should be mentioned that in this α -type organic salt, the Dirac points appear at low temperatures when the charge order melts. One possible option to realize such a situation is application of uniaxial pressure P_a , which is known to melt the charge ordering in the ground state. Thus, the predicted topological phase transitions may be observed under application of the pressure. The measurements must be performed using diamond anvil pressure cells, which are transparent for laser light. Another option is to perform the experiments above the charge ordering temperature $T_{CO}(\sim 40)$ K, which is much easier than the first option. Its feasibility is supported by our calculated Hall conductivity data, which shows that the quantized Hall conductivity is observed even at $T = 100$ K.

IV. CONCLUSION

To summarize, we theoretically predicted the emergence of rich photoinduced topological phases as nonequilibrium

steady states in organic salt α -(BEDT-TTF) $_2$ I $_3$ with inclined Dirac-cone bands under continuous application of circularly polarized light. The predicted topological electronic phases and their transitions upon the laser-parameter variations are expected to be observed in future experiments by measuring the photoinduced Hall effect. A crucial advantage of the usage of organic compounds is that the effective amplitude of the laser is enhanced significantly due to the large molecule sizes and resulting larger lattice constants because the dimensionless laser amplitude $\mathcal{A}_a = eaE^\omega/\hbar\omega$ and $\mathcal{A}_b = ebE^\omega/\hbar\omega$ are proportional to the lattice constants a and b . We expect the effective vector potential $\mathcal{A}$ in α -(BEDT-TTF) $_2$ I $_3$ to be nearly an order of magnitude larger than that for graphene, and hence the feasibility of experimental realization of the predicted photoinduced phenomena is expected to be increased. The field of photoinduced topological phase transitions is now rapidly growing, but target materials for research are still limited to a few toy models and simple atomic-layered materials. Our work may contribute by advancing this research field through broadening the range of target materials.

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