Optical Imprinting of Superlattices in 2D Materials

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We propose a novel optical method to imprint superlattice structures in two-dimensional electronic systems. By changing the shape of the optical field, we synthesize various lattice structures with different spatial symmetry, periodicity, and strength. We find that the wide optical tunability allows one to tune different properties of the effective band structure, including Chern number, energy bandwidths, and band gaps. We consider the high-frequency regime where the electronic system can remain in the quasi-equilibrium phase for an extended amount of time. The spatiotemporal reconfigurability of our approach opens up new possibilities to control light-matter interaction to generate novel electronic states and optoelectronic devices.

Introduction.— Superlattice structure in 2D materials has opened a new way to engineer electronic bands, starting with investigating of honeycomb superlattice structure in monolayer graphene [1]. Recently, the moiré pattern in twisted bilayer van der Waals heterostructure has been immensely successful in generating a variety of band structures, including Hofstadter butterfly [2, 3] and flat bands [4–8]. These bands can induce intriguing strongly correlated phases such as fractional Chern insulator [3], anomalous Hall phase [9, 10], Mott insulating phase [8, 11–13], non-trivial magnetic phases [9, 14–16], and superconductivity [7, 13, 17–19]. Yet, this passive way of creating superlattice has been largely limited by the microscopic structure of the 2D materials and it is difficult to dynamically control the superlattice. Therefore, it is interesting to find alternative ways to synthesize spatiotemporal structure in 2D materials.

As the same time, the recent progress in beam shaping technique has enabled the generation of arbitrary beam patterns with high resolution comparable to the optical wavelengths [20–25], which already found remarkable successes in ultracold atom systems [26–30]. This wide tunability of light can be naturally applied to 2D electronic systems to imprint arbitrary superlattices, regardless of the underlying microscopic lattice structure. This is particularly interesting in the context of the “Floquet topological insulator”, where the illumination of circularly-polarized (CP) light can turn a trivial system into a topological insulator [31–35].

In this Letter, we propose a method to create superlattice structures in 2D material using continuous waves of light. We illustrate the idea with an example of monolayer graphene irradiated by a circularly-polarized beam with a superlattice structure, where the beam amplitude is spatially periodic. To demonstrate the tunability of this superlattice structure, we first study the case of a square lattice and explore the topological phase transition induced by varying the superlattice size. Then, we investigate the topological phase transitions, when the square lattice sheared to a stretched hexagonal one. In particular, we examine the relationship between this topological phase transition and the role of lattice geometry in creating complex tunneling phases. Also, we demonstrate the possibility of creating more exotic lattices by superposing multiple lattices, with an example of tuning between a hexagonal and a kagome lattice.
Graphene with spatially patterned light.— Let us consider a monolayer graphene with the inter-atomic distance $a$ and the tight-binding energy $t$ between the nearest neighbors. The low-energy description for this monolayer graphene under the electromagnetic field $A(r, t)$ is given by

$$H = v [p + eA(r, t)] \cdot (\tau_x \sigma_x \hat{x} + \sigma_y \hat{y})$$

(1)

where $\sigma_x, \sigma_y, \sigma_z$ are Pauli matrices acting on sublattice degrees of freedom, $v = (3/2)ta$ is the Fermi velocity at Dirac points, and $\tau_z = \pm 1$ is the valley index [36]. In particular, if we shine the CP beam with spatial amplitude pattern

$$A(r, t) = A_0(r) e^{i \omega t} (\hat{x} + i \hat{y}) + c.c.$$ (Fig. 1), the effective Floquet Hamiltonian to the first order of $\omega^{-1}$ becomes [31, 37–43]

$$H_{\text{eff}} = v (\tau_z p_x \sigma_x + p_y \sigma_y) + \tau_z \frac{4e^2v^2}{\omega} |A_0(r)|^2 \sigma_z.$$ (2)

We denote the peak amplitude of $A_0(r)$ as $A_0$. Then Eq. (2) becomes a valid description when frequency $\omega$ is high enough ($\omega \gg ev/A_0$) and the amplitude varies in length scale larger than $a$ ($A_0/\max \{ |\nabla A_0(r)| \} \gg a$). For brevity, we set $h = 1$ from here on.

We specifically study the superlattice structure created by a spatially periodic amplitude $|A_0(r)| = |A_0(r + L_1)| = |A_0(r + L_2)|$. While the 2D material with spatially modulated beams has been studied in the different contexts [44–46], generation of superlattice with this has not been investigated yet. In particular, to make the beam experimentally relevant, we consider the superposition of CP Gaussian beams positioned on the superlattice,

$$A_0(r) = \sum_{n_1, n_2} A_0 \exp \left( -\frac{|r - n_1L_1 - n_2L_2|^2}{2w^2} \right),$$ (3)

where $w$ is the radius of each Gaussian beam. This beam configuration is achievable with recent progress in beam shaping technologies [20–25]. For the cases $|L_2|, |L_1| = l \gg a$, Brillouin zone folding occurs on a momentum scale $l/2$, so that the low-energy description is captured by Eq. (2). We obtain Bloch eigenstates $|\psi_{m, k}\rangle$ and eigenenergies $E_{m, k}$ where $m$ is the band index and $k$ is the crystal momentum within the Brillouin zone set by reciprocal lattice vectors of $L_1$ and $L_2$. Note that Eq. (2) preserves particle-hole symmetry $(\sigma_x H_{\text{eff}} \sigma_x = -H_{\text{eff}})$ and therefore the energy spectrum is symmetric with respect to the zero energy. Also, $\sigma_y H_{\text{eff}} \sigma_y = H_{\text{eff}}$, so the energy spectrum is invariant under a unitary operation, $\sigma_y$. This also ensures that both valleys have the same Chern number. For brevity, let us only consider $\tau_z = 1$ valley from now on.

Square lattice.— We first consider the simplest case of a square superlattice, $L_1 = l \hat{x}$ and $L_2 = l \hat{y}$. Before directly diagonalizing Eq. (2), we can make some speculations. First of all, the contribution from the spatial average of $|A_0(r)|$ opens up the gap around the zero energy ($\Delta_0$) as in the case of the graphene under the CP uniform light, where the lowest positive energy band has non-zero Chern number [31, 33, 44, 47, 48]. This regime of Floquet Chern insulator is pertained for small $l$ where the maximum kinetic energy within the Brillouin zone, which is of order $v/l$, is much larger than the spatial Fourier components of the $\sigma_z$ term in Eq. (2), which is of order $e^2v^2/\omega A_0^2$. On the other hand, as $l \rightarrow \infty$, the contribution of the kinetic term becomes negligible and therefore the bands become flat. Also, the Bloch wavefunctions look similar regardless of $k$ and therefore the bands become topologically trivial. Therefore, there must be a topological phase transition from a FCI to a normal insulator (NI) as we increase $l$. This topological transition would occur at superlattice size which makes the two energy scales $e^2v^2/\omega A_0^2$ and $v/l$ comparable to each other. For a succinct description of this phase transition, we use the rescaled superlattice size $\chi = (v/e^2 A_0^2) l$ so that the critical superlattice size $\chi_c$ to be $O(1)$.

To study the detail of this topological phase transition, we...
we numerically diagonalize Eq. (2) as shown in Fig. 2(a). Along with the energy spectrum, we present the Chern number $C$ of each band calculated based on Ref. [49]. Given parameters in Fig. 2(a), we can check that the topological phase transition is $\chi = 0.965$ which is close to 1. This topological transition accompanies the direct gap closing at $k = M$ and the band inversion between the first–second-lowest positive energy bands. To see this, we compare the particle and current density of the lowest positive energy band’s wavefunction at the direct gap closing point. Here, for the Bloch wavefunction of the $m$-th band $\psi(r) = \langle r|\psi_{m,k}\rangle$, the particle and current density are given by

$$n(r) = \psi^\dagger(r)\psi(r),$$

$$j(r) = -e\psi^\dagger(r)\frac{\partial H_{\text{eff}}}{\partial p}\psi(r) = -e\psi^\dagger(r)(\sigma_x\hat{x} + \sigma_y\hat{y})\psi(r).$$

The comparison of $n(r)$ and $j(r)$ before ($\chi = 0.8$) and after ($\chi = 4$) the transition point shows a drastic change in the wavefunction, which signifies that the band inversion has occurred in the phase transition. In the current density plot, one can also find that the circulation direction of electron flips as the band inversion occurs. This phenomenon can be also captured in the calculation of the $m$th band contribution to the orbital magnetization [50–52],

$$M_{\text{orb}} = \text{Im} \int \frac{d^2k}{(2\pi)^2} e^{\frac{\partial \langle u_{m,k}\rangle}{\partial k_x}}(H_k + E_{m,k})\frac{\partial |u_{m,k}\rangle}{\partial k_y}$$

where $|u_{m,k}\rangle = e^{-ikr}|\psi_{m,k}\rangle$ and $H_k = e^{-ikr}H_{\text{eff}}e^{ikr}$. In Fig. 2(b), one can see that $M_{\text{orb}}$ of the lowest positive band shows the sign flip at the phase transition point, agreeing with the observation in the current density plots.

This topological phase transition could be experimentally detected in several ways. In FCI regime, the finite size sample can have the chiral edge state and the evidence for topological Floquet bands can be revealed by transport experiments, similar to Ref.[35]. For the bulk property, one can measure the orbital magnetization, wherein the sudden jump would be observed at the phase transition Fig. 2(b).

As the superlattice size $\chi$ increases, the electrons become localized at local minima of $|A_0(r)|$. This provides an explanation for exponential suppression of the bandwidth of the lowest positive energy band ($\delta E$) in $\chi$ [Fig. 2(c)]. For well-localized electrons, the dynamics can effectively be described by a tight-binding model, and the tunneling energy of that model is approximately given by the WKB integrals. This integral decays exponentially with the distance between the superlattice sites, so the bandwidth decreases exponentially as well. The band gaps ($\Delta_0$, $\Delta_1$, $\delta_M$) decay as $O(\chi^{-1})$, where the details of this band gap scaling are explained in the supplemental material [43].

**Lattice shearing.**— To investigate the role of the superlattice geometry further, let us shear the square superlattice by angle $\theta$ so that $L_1 = l\hat{x}$ and $L_2 = l(tan(\theta)\hat{x} + \hat{y})$.

From the perspective of a FCI with uniform CP light, in large superlattice size limit where the tight-binding description is valid, we might interpret the electron tunneling between superlattice sites as the chiral currents around each Gaussian CP beam. If the system has reflection symmetry around the line connecting the two sites, this tunneling should be real. Otherwise, the tunneling can have a complex phase. For examples, the next-nearest neighbor tunnelings for $\theta = 0, \pi/4$ case and $\theta = \tan^{-1}(1/2)$ case are presented. (b) Chern number of the lowest positive energy band is shown as a phase diagram between the shearing angle $\theta$ and the superlattice size $\chi$. (c) Energy spectrums for $\chi = 2.4$ at selected angles are shown where the colors of lowest-lying bands represent the Chern numbers. Particle density in the unit of $t^{-2}$ is plotted for angles before and after the phase transition.

![FIG. 3. (a) We shear a square lattice by angle $\theta$. Tunneling between two sites can be understood as the flow of chiral edge currents, around each Gaussian CP beam. If the system has reflection symmetry around the line connecting the two sites, this tunneling should be real. Otherwise, the tunneling can have a complex phase. For examples, the next-nearest neighbor tunnelings for $\theta = 0, \pi/4$ case and $\theta = \tan^{-1}(1/2)$ case are presented. (b) Chern number of the lowest positive energy band is shown as a phase diagram between the shearing angle $\theta$ and the superlattice size $\chi$. (c) Energy spectrums for $\chi = 2.4$ at selected angles are shown where the colors of lowest-lying bands represent the Chern numbers. Particle density in the unit of $t^{-2}$ is plotted for angles before and after the phase transition.](image)
FIG. 4. Superposition of the triangular lattice beam, $A_{\text{tri}}(r)$, and the hexagonal lattice beam $rA_{\text{hex}}(r)$. As we increase the ratio $r$, we effectively change the superlattice type from the hexagonal lattice to the kagome lattice. Energy spectrums for $\chi = 5.4$ at selected values of $r$ are shown where the colors of low-lying bands represent the Chern numbers. By zooming in the spectrum, we can check the gaps in the two-band model and the three-band models in the lowest part of the spectrum.

neighbor tunneling makes the system a FCI at this angle. With these observations, we can predict NI-FCI-NI transitions as we increase $\theta$ from 0 to $\pi/4$.

We obtain the phase diagram for this topological transition numerically [Fig. 3(b)]. Here, we calculate the Chern number of the lowest positive energy band for different values of $\chi$ and $\theta$. As we predicted, we can observe NI-FCI-NI transition at $\chi$ larger than a certain value, which corresponds to the phase transition point described in Fig. 2. One can also find that the system remains as FCI at the angle $\theta = \tan^{-1}(1/2)$ regardless of $\chi$, while the range of angle which makes the system FCI shrinks as $\chi$ increases. This can be explained by combining the fact that the size of tunneling strengths decreases exponentially in the distance between the superlattice points and the Dirac cones can disappear and the topologically trivial gap opens in the extreme strain [43]. We can also see that the topological phase transition also accompanies the gap closing and the band inversion as shown in the particle density plots [Fig. 3(c)].

Hexagonal lattice to kagome lattice.—To engineer favorable features such as flatter bands, we can create even more complicated superlattice by superposing different kinds of lattices. For instance, we consider the superposition of the triangular lattice beam $A_{\text{tri}}(r)$ and the hexagonal lattice beam $rA_{\text{hex}}(r)$ where $r$ is the amplitude ratio of the two lattices (Fig. 4). When the contribution from the hexagonal lattice beam is negligible, the localized electrons form a hexagonal superlattice and the lowest part of the positive energy spectrum can be explained by a two-band model. As $r$ increases, electrons are confined to a kagome superlattice and the lowest part of the positive energy spectrum can be explained by a three-band model including a flat band. Note that slight gaps are observed in both of the two-band model for the hexagonal superlattice and the three-band model for the kagome superlattice. The gap in the two-band model can be explained with the Haldane model with complex phases in the next-nearest neighbor tunneling as shown in Fig. 3(a). The gap in the kagome lattice comes from the complex phase in the nearest neighbor tunneling [42, 56]. At $r = 0$, we can see that the third band is nearly flat while it is gapped well from the other bands. This flat band can be potentially used to stabilize strongly-correlated phases.

Experimental feasibility.—For numerical calculation, we set $A_0 = 0.006(\omega a)^{-1}$, $\omega = 0.06t$, and $w/l = 0.3$ for Fig. 2 and Fig. 3. Provided $t = 3$ eV and $a = 0.142$ nm, these values correspond to the field amplitude $7.6 \times 10^6$ V/m, the beam frequency 43.5 THz, and beam spot size 0.1 $\mu$m (FWHM). This is the similar beam frequency in the recent experiment [35] while the peak intensity is about 4% of the beam used in the same experiment. With these parameters, typical size of gap ($\Delta_b$ in Fig. 2) is 4 meV. Fig. 4 uses $A_0 = 0.0015(\omega a)^{-1}$ and $\omega = 0.06t$ while $w/l = 0.3$ and $w/l = 0.15$ for $A_{\text{tri}}(r)$ and $A_{\text{hex}}(r)$, respectively. Finally, we remark that due to the injection of photons into the system, heating effects could eventually destroy the non-trivial topological behavior that is initially formed. Therefore, we only consider the prethermal regime where electron-electron and electron-phonon scatterings can be ignored. In the past few years, the existence of this transient regime has been convincingly demonstrated in several pump-probe experiments [34, 35, 57].

Outlook.—By considering Coulomb interaction in our nearly flat and topologically non-trivial bands, one could potentially induce strongly correlated phases such as fractional Chern insulators [3, 58–60], superconductors [7, 13, 17–19, 61, 62], or magnetic phases [9, 14–16]. Moreover, by irradiating with frequencies comparable to the bare tunneling strength, instead of the high-frequency regime considered here, higher-order terms become relevant [42], and therefore, one can induce a wider class of structures. While we focus on the Dirac semimetal system in this paper, our scheme can be also applied to other 2D materials such as semiconductors [63]. Our approach can be combined with other methods, such as surface acoustic waves in solid-state platform [64], for trapping, cooling, and controlling charged particles, and simulation of quantum many-body systems. Finally, these ideas could be used to engineer a new class of dielectric materials for potential applications in optical devices [65].

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Supplemental Material: Optical Imprinting of Superlattices in 2D Materials

I. FLOQUET EFFECTIVE HAMILTONIAN IN HIGH FREQUENCY REGIME

Let us consider the Hamiltonian Eq. (1) with $\mathbf{A}(\mathbf{r}, t) = A_0(\mathbf{r})e^{i\omega t}(\mathbf{\hat{x}} + i\mathbf{\hat{y}}) + c.c.$ Then we can write the time-dependent Hamiltonian as

$$H(t) = v(\tau_2 p_x \sigma_x + p_y \sigma_y) + 2ev \tau_2 A_0(\mathbf{r}) \exp(i\tau_2 \omega t) \sigma_+ + 2ev \tau_2 A_0(\mathbf{r}) \exp(-i\tau_2 \omega t) \sigma_-$$

where $\sigma_\pm = (\sigma_x \pm i\sigma_y)/2$. For this Hamiltonian, the non-zero temporal Fourier components $H_q = (\omega/2\pi) \int_{0}^{2\pi/\omega} H(t) e^{-iq\omega t} d\tau$ are $H_0 = v(\tau_2 p_x \sigma_x + p_y \sigma_y)$ and $H_{\pm} = 2ev \tau_2 A_0(\mathbf{r}) \sigma_{\pm}$. Then the effective Hamiltonian in high frequency regime is [31, 37–42]

$$H_{\text{eff}} = H_0 + \sum_{q>0} \frac{[H_q, H_{-q}]}{q\omega} + O(\omega^{-2}) = H_0 + \frac{H_1 H_{-1} - H_{-1} H_1}{\omega} + O(\omega^{-2})$$

$$= v(\tau_2 p_x \sigma_x + p_y \sigma_y) + \tau_2 \frac{4e^2 v^2}{\omega} |A_0(\mathbf{r})|^2 \sigma_x + O(\omega^{-2}).$$

II. BAND GAP SCALING IN SUPERLATTICE SIZE

We consider the eigenvalue problem of the effective Hamiltonian in Eq. (2),

$$\begin{pmatrix} \alpha(\mathbf{r}) & -i \nu(\partial_x - i \partial_y) \\ -i \nu(\partial_x + i \partial_y) & -\alpha(\mathbf{r}) \end{pmatrix} \begin{pmatrix} u_A \\ u_B \end{pmatrix} = E \begin{pmatrix} u_A \\ u_B \end{pmatrix} \Leftrightarrow -i \nu(\partial_x - i \partial_y) u_B = |E - \alpha(\mathbf{r})| u_A, \quad -i \nu(\partial_x + i \partial_y) u_A = |E + \alpha(\mathbf{r})| u_B,$$

where $\alpha(\mathbf{r}) = (4e^2 v^2/\omega)|A_0(\mathbf{r})|^2$. This can lead to

$$\nabla^2 u_B + \frac{1}{\nu^2} |E^2 - \alpha(\mathbf{r})|^2 |u_B| + \frac{[(\partial_x + i \partial_y) \alpha(\mathbf{r})][(\partial_x - i \partial_y) u_B]}{E - \alpha(\mathbf{r})} = 0.$$  

In the vicinity of minima of $\alpha(\mathbf{r})$, we can approximate this function as an harmonic potential with rotational symmetry. This is valid for the square lattice of Gaussian beam with the fixed ratio $c = w/l$,

$$\alpha(\mathbf{r}) = \sum_{m_1, m_2, n_1, n_2} \frac{4e^2 v^2 A_0^2}{\omega} \exp\left[-\frac{(x - m_1 l)^2 + (x - n_1 l)^2 + (y - m_2 l)^2 + (y - n_2 l)^2}{2c^2 l^2}\right].$$

For this case, one can show that

$$\partial_x \alpha(\mathbf{r})|_{x = y = l/2} = \partial_y \alpha(\mathbf{r})|_{x = y = l/2} = \partial_x \partial_y \alpha(\mathbf{r})|_{x = y = l/2} = \partial_y \partial_x \alpha(\mathbf{r})|_{x = y = l/2} = 0,$$

$$0 = \alpha(\mathbf{r})|_{x = y = l/2} > 0, \quad \alpha_1 = \frac{\nu^2}{2} \partial_x^2 \alpha(\mathbf{r})|_{x = y = l/2} = \frac{\nu^2}{2} \partial_y^2 \alpha(\mathbf{r})|_{x = y = l/2} > 0, \quad \partial \alpha_0 / \partial l = \partial \alpha_1 / \partial l = 0.$$  

Then we can write $\alpha(\mathbf{r}) = \alpha_0 + \alpha_1 (r/l)^2$, where $r$ is the distance from the minima of $\alpha(\mathbf{r})$. Now we can use polar coordinates $(r, \phi)$, with $\partial_x \pm i \partial_y = -1/r \partial_r + i r^{-1} \partial_\phi$. Due to the rotational symmetry, we can impose $u_B(\mathbf{r}) = \beta(r) e^{i\rho \phi}$. Then

$$\frac{1}{r} \partial_r (r \partial_r \beta) - \frac{m^2}{r^2} \beta + \frac{1}{2} \left[E^2 - \alpha_0^2 - 2\alpha_0 \alpha_1 \left(\frac{r}{l}\right)^2 - \alpha_1^2 \left(\frac{r}{l}\right)^4\right] \beta + \frac{2\alpha_1(r \partial_r + m) \beta}{(E - \alpha_0)^2 - \alpha_1^2 r^2} = 0.$$  

Note that $l \rightarrow \infty$ limit corresponds to $\nabla^2 u_A + v^2 (E^2 - \alpha_0^2) = 0$. The positive spectrum in this limit is $[\alpha_0, \infty)$ with no gap in between. To study the behavior of the positive spectrum for large $l$, we may define $\delta E = E - \alpha_0$. For the low-lying spectrum, we can only consider the limit where $\delta E \ll \alpha_0$. Then we can simplify Eq. (S7) into

$$\frac{1}{r} \partial_r (r \partial_r \beta) - \frac{m^2}{r^2} \beta + \frac{1}{2} \left[2\alpha_0 \delta E - 2\alpha_0 \alpha_1 \left(\frac{r}{l}\right)^2\right] \beta + \frac{2(r \partial_r + m) \beta}{r^2 E/\alpha_1} = 0$$

up to the correction terms of order $O(\delta E^2), O(\eta^{-4})$. Now the rescaling $r = (vl)^{1/2}(\alpha_0 \alpha_1)^{-1/4} \xi$ and $\delta E = (\alpha_1/\alpha_0)^{1/2}(v/l)(\delta \epsilon)$ gives

$$\frac{1}{\xi} \partial_\xi (\xi \partial_\xi \beta) - \frac{m^2}{\xi^2} \beta + 2 (\delta \epsilon - \xi^2) \beta + \sqrt{\alpha_0 \alpha_1} \frac{2(r \partial_r + m) \beta}{v \delta \epsilon} = 0.$$  

(S9)
and this equation is independent of $l$. Then the spectrum of $\delta \epsilon$ is independent of $l$, Therefore $\delta E - \alpha_0$ should scale as $l^{-1}$. This means that $\Delta_h - 2\alpha_0$ and $\Delta_l$ should be proportional to $l^{-1}$. This explains the inverse proportionality of band gaps in $\chi = (\nu e^2 \mathcal{A}_0^2 / \omega) l$ shown in Fig. 2(b).

### III. Tight-Binding Model for Hexagonal Lattice Under a Uniform Strain

![Figure S1](image)

FIG. S1. Tight-binding model for the hexagonal lattice under a uniform strain, in the vicinity of angle $\theta = \tan^{-1} 0.5$ in the sheared lattice.

Let us consider the effective lattice model for the sheared lattice in the vicinity of angle $\theta = \tan^{-1} 0.5$. By considering the terms up to the next-nearest neighbors, we can build a tight-binding model similar to Haldane model,

$$
H_{SH} = \sum_{m,n} \left( -t_1 c_{m,n}^{(B)} + t_2 c_{m-1,n}^{(B)} - t_3 c_{m,n-1}^{(B)} \right) c_{m,n}^{(A)}
$$

(S10)

$$
+ \left( s_1 c_{m+1,n}^{(A)} + s_2 c_{m,n-1}^{(A)} + s_3 c_{m-1,n+1}^{(A)} \right) c_{m,n}^{(A)}
$$

$$
+ \left( s_1 c_{m-1,n}^{(B)} + s_2 c_{m,n+1}^{(B)} + s_3 c_{m+1,n-1}^{(B)} \right) c_{m,n}^{(B)} + \text{H.c.}
$$

Here, $c_{m,n}^{(A/B)}$ creates an electron in the sublattice A/B at the unit cell $(m, n)$ and $t_{i=1,2,3}$ $(s_{i=1,2,3})$ is the nearest (the next-nearest) neighbor tunneling amplitude as shown in Fig. S1. This model can be thought of a hexagonal lattice under a uniform strain. By considering the inversion symmetry of the corresponding pairs of lattice sites, we can find that $\text{Im}(t_1) = \text{Im}(t_2) = \text{Im}(t_3) = 0$. Now we can write down the Bloch Hamiltonian of this tight-binding model as

$$
\mathcal{H}(k) = \begin{pmatrix}
  s_1 e^{-ik(2L_2 - L_1)} + s_2 e^{-ikL_2} + s_3 e^{-ikL_1} + \text{c.c.} & -e^{-ik(L_1 + L_2)}/3 (t_1 + t_2 e^{ikL_1} + t_3 e^{ikL_2}) \\
  -e^{ik(L_1 + L_2)}/3 (t_1 + t_2 e^{-ikL_1} + t_3 e^{-ikL_2}) & s_1 e^{ik(2L_2 - L_1)} + s_2 e^{ikL_2} + s_3 e^{ikL_1} + \text{c.c.}
\end{pmatrix}
$$

$$
= V(k) + h_x(k) \sigma_x + h_y(k) \sigma_y + h_z(k) \sigma_z,
$$

(S11)

where the $\sigma_z = \pm 1$ corresponds to the sublattice $A/B$. In the absence of the next-nearest neighbor tunnelings, $V = h_z = 0$ and the location of Dirac points are determined by $h_x(k) = h_y(k) = 0$. Then the two different Dirac points are given by the form of $k = \pm K'$. Since $h_x(-K') = -h_z(K')$, the sign of this mass term is opposite at the different Dirac points as in the Haldane model. Therefore, if the Dirac points exist in the absence of the next-nearest neighbor tunneling, the system becomes an FCI when the next-nearest neighbor tunneling is turned on. Regarding this condition, the equation $h_z(k) = h_y(k) = 0$ has two solutions as long as $|t_1 - t_j| < |t_k|$ for every $i \neq j \neq k \neq i$. Since the tunneling strength decreases exponentially in the inter-site distance, $t_1$, $t_2$, and $t_3$ become very different as the superlattice size gets larger. Then there is no Dirac point after some value of $\chi$ as shown in the square lattice case. Yet, at angle $\theta = \tan^{-1} 0.5$, $t_1 = t_2$ so that $|t_j - t_i| < |t_k|$ is satisfied as long as $t_3 \neq 0$, and therefore the system can remain as FCI at this angle.
We want to numerically calculate orbital magnetization of the nth band expressed in Eq. (5),

\[ M_{\text{orb}} = \text{Im} \int \frac{d^2k}{(2\pi)^2} e^{i \langle u_{m,k} \mid H_k + E_{m,k} \rangle} \frac{\partial |u_{m,k}\rangle}{\partial k_x} \]

\[ = \text{Im} \int \frac{d^2k}{(2\pi)^2} e^{i \langle u_{m,k} \mid H_k \rangle} \frac{\partial |u_{m,k}\rangle}{\partial k_y} + \frac{1}{2} \int \frac{d^2k}{(2\pi)^2} E_{m,k} A_{m,k}. \]  

(S12)

where \( A_{m,k} = 2\text{Im} \left( \partial_{k_x} \langle u_{m,k} \mid \right) \partial_{k_y} |u_{m,k}\rangle \) is the Berry curvature. To numerically calculate it, we first need to discretize the Brillouin zone and calculate Bloch state \(|\psi_{m,k}\rangle\). Although the orbital magnetization is gauge-independent, we need local gauge fixing to make \(|\psi_{m,k}\rangle\) differentiable. While this local gauge fixing works well for smooth \( A_{m,k} \), it can work badly for the system in the vicinity of topological phase transition. To avoid this subtlety, let us find a way to calculate this quantity in a gauge-independent way. For Berry curvature \( A_{m,k} \), a method for gauge-independent calculation is known [49]. Similar to this method, we can calculate the first integral of Eq. (S12). For this, let us consider a square patch whose four corners are \( q_{ij} \equiv k_i + \frac{\delta k}{2}(s_j \hat{x} + w_j \hat{y}) \) where \((s_1, w_1) = (-1, -1), (s_2, w_2) = (1, -1), (s_3, w_3) = (1, 1)\), and \((s_4, w_4) = (-1, 1)\). Now

\[ |u_{m,q_{ij}}\rangle = |u_{m,k}\rangle + \frac{\delta k}{2} \left( s_j \partial_{k_x} |u_{m,k}\rangle + w_j \partial_{k_y} |u_{m,k}\rangle \right) + \frac{\delta k^2}{8} \left( \partial_{k_x}^2 |u_{m,k}\rangle + 2 s_j w_j \partial_{k_x} \partial_{k_y} |u_{m,k}\rangle + \partial_{k_y}^2 |u_{m,k}\rangle \right) + O(\delta k^3), \]

\[ = 1 + \frac{\delta k}{2} \sum_{j=1}^{4} \left( s_j \left\langle u_{m,k} \mid \partial_{k_x} |u_{m,k}\rangle \right\rangle + w_j \left\langle u_{m,k} \mid \partial_{k_y} |u_{m,k}\rangle \right\rangle \right) + \frac{\delta k^2}{8} \sum_{j=1}^{4} \left( \left\langle u_{m,k} \mid \nabla_{k_x}^2 |u_{m,k}\rangle \right\rangle + \frac{s_j w_j}{2} \left\langle u_{m,k} \mid \partial_{k_x} \partial_{k_y} |u_{m,k}\rangle \right\rangle \right) \]

+ \frac{\delta k^2}{8E_{m,k}} \sum_{j=1}^{4} \left( s_j \left\langle u_{m,k} \mid H_k \partial_{k_x} |u_{m,k}\rangle \right\rangle + w_j \left\langle u_{m,k} \mid H_k \partial_{k_y} |u_{m,k}\rangle \right\rangle \right) \]

\[ + \frac{\delta k^2}{8E_{m,k}} \sum_{j=1}^{4} \left( s_j \left\langle u_{m,k} \mid H_k \partial_{k_x} |u_{m,k}\rangle \right\rangle + w_j \left\langle u_{m,k} \mid H_k \partial_{k_y} |u_{m,k}\rangle \right\rangle \right) \]

\[ = 1 + 2\delta k^2 \text{Re} \left\langle u_{m,k} \mid \nabla_{k_x}^2 |u_{m,k}\rangle \right\rangle + i \frac{2\delta k^2}{E_{m,k}} \text{Im} \left( \partial_{k_x} \left\langle u_{m,k} \mid H_k \partial_{k_y} |u_{m,k}\rangle \right\rangle \right) + O(\delta k^3), \]

(S13)

therefore

\[ \frac{eE_{m,k}}{8\pi^2} \text{Arg} \left( \prod_{j=1}^{4} \left\langle u_{m,q_{ij}} \mid H_k \right\rangle \right) = \frac{e}{4\pi^2} \text{Im} \left( \partial_{k_x} \left\langle u_{m,k} \mid H_k \partial_{k_y} |u_{m,k}\rangle \right\rangle \right) \delta k^2 + O(\delta k^3) \]  

(S14)

and this corresponds to the first integral of Eq. (S12) over the square patch we considered. One can easily check that this expression is invariant under any gauge transformation, \(|u_{m,k}\rangle \rightarrow \exp[i\lambda(k)] |u_{m,k}\rangle, \forall \lambda(k)|, and does not require any local gauge fixing.