We present a low-energy model describing the reconstruction of the electronic spectrum in twisted bilayers of honeycomb crystals with broken sublattice symmetry. The resulting moiré patterns can be classified into two families with different symmetry. In both cases, flat bands appear at relatively large angles, without any magic angle condition. These flat bands arise from midgap states spatially localized in regions of unnatural stacking, where ions of the same polarity lie on top of each other. Transitions between these bands give rise to sharp resonances in the optical absorption spectrum at frequencies well below the gap of the monolayer. The system displays circular dichroism – different absorption of left- and right-handed circularly-polarized light – due to the chiral symmetry of the twisted bilayer. In hexagonal boron nitride, we find that circular dichroism is maximized in one type of moiré structure, which can be linked to approximate symmetries not contained in the space group. Our calculation illustrates how the chiral response of moiré-patterned materials probes geometrical properties of the electronic wave function.

Twisting and sliding the layers of van der Waals materials creates superstructures with new emergent properties. The most singular case is that of twisted bilayer graphene, i.e., two graphene layers rotated with respect to each other an angle $\theta$ [1]. In the regime of small twist angles, the relative shift in the periodicities of each individual lattice creates a moiré beating pattern. Quantum interference of Dirac electrons in the associated superlattice potential reshapes the conical dispersion of the low-energy bands. As the twist approaches the magic angle condition $\theta \sim 1.1^\circ$ [2-3], the reconstructed Dirac cones evolve into flat bands, favoring the emergence of electronic correlated states [4].

In twisted bilayers whose components have a gap (e.g., semiconducting transition-metal dichalcogenides [5-6] or hexagonal boron nitride), flat bands appear without any special alignment condition due to the existence of a natural energy scale for electron confinement in the original spectrum. This is in stark contrast with the case of twisted bilayer graphene, which remains gapless after twisting, as the Dirac cones are preserved by the emergent symmetries of incommensurate superstructures [7]. The latter are captured by the continuum model [1, 3], which provides an accurate description of the electronic spectrum at small twist angles.

A common property of these systems is that twisting the layers break all the inversion planes/centers, reducing the symmetry to a purely rotational (i.e., chiral) group. Chirality gives rise to optical activity, and is usually measured through different responses to left- and right-handed circularly-polarized light [8]. This circular dichroism (CD) results from current counterflows in response to an electric field [9], which engenders an in-plane magnetization density that rotates the polarization of the incident light [10].

Here, we study the optical activity of twisted bilayers with broken sublattice symmetry (i.e., where each sublattice is occupied by different atomic species). We employ a low-energy continuum model and go beyond the single band approximation [5-11] by including coherences across the gap. Applying our model to twisted bilayers of hexagonal boron nitride (hBN), we find significant circular dichroism at frequencies that are resonant with the energy difference between the lowest flat bands. We show that, while band localization depends on the orbital hybridization in regions of unnatural stacking, optical activity is influenced by more subtle geometrical properties of the electronic wave function linked to approximate symmetries of the system.

There are two families of twisted structures characterized by different symmetry, as shown in Fig. 1. Any structure can be generated by a relative rotation of angle $\theta \in [−30^\circ, 30^\circ]$ taken along a common center of the hexagons, followed by a relative translation $\mathbf{u}$, starting from either AA’ or AA stacking configurations, see Fig. 1(b). In the first case, ions of opposite polarity lie on top of each other before the twist; these are moiré structures of type I. The other case, where ions of same polarity initially sit on top of each other, are moiré structures of type II. The shift in periodicity of the individual lattices define the primitive wavevectors $\mathbf{g}_i$ of the associated beating pattern, $\mathbf{G}_i = R(−\theta/2)\mathbf{g}_i − R(\theta/2)\mathbf{g}_i = 2\sin(\theta/2)\mathbf{g}_i × \hat{\mathbf{z}}$, where $\mathbf{g}_i$ are the primitive vectors of the reciprocal lattice prior to the rotation, and $R$ is the associated matrix. Within the moiré supercell, highlighted with dashed lines in Fig. 1(a), structures of type I explore the initial AA’ (black dots), AB’ (red dots) and BA’ (blue dots) registries. In type-II structures, we have instead AA, AB and BA registries.

At small twist angles, the low-energy electronic states around valley $\zeta$ can be generically described by a continuum Hamiltonian of the form

$$\mathcal{H}_\zeta(r) = \begin{bmatrix} \mathcal{H}^{(t)}_\zeta(r) & \hat{T}_\zeta^{(t)}(r) \\ \hat{T}_\zeta^{(t)}(r)^T & \mathcal{H}^{(l)}_\zeta(r) \end{bmatrix},$$

where blocks in the diagonal describe the top ($t$) and
FIG. 1. Moiré patterns and symmetries. (a) Moiré pattern resulting from a twist of θ = 5° and relative translation u = 0. Black, red and blue dots correspond to local high-symmetry stackings. (b) High-symmetry stacking configurations. Starting from either AA’ or AA configurations, a high-symmetry stacking is generated by sliding one layer with respect to the other a nearest-neighbor vector δi. (c) Point-group operations in both types of moiré structures, where open (filled) circles represent atoms on the top (bottom) layer. Dashed lines represent in-plane C2 rotation axes and the black point is the C3 rotation.

bottom (b) layers, and the off-diagonal blocks account for interlayer hopping processes. The Hamiltonian acts on a Hilbert space of envelope wave functions, which are smooth on the scale of the microscopic lattice spacing. a Next, we derive the model for hBN, where the edges of the π bands reside around the two inequivalent corners K± of the hexagonal Brillouin zone. The sublattice, layer, and valley (ζ = ±1 for K±) projections of the microscopic wave function are incorporated as internal quantum numbers. The blocks in the diagonal follow from the tight-binding Hamiltonian $\mathcal{H}(k) = t \text{Re}\{\Phi(k)\} \hat{\sigma}_x - t \text{Im}\{\Phi(k)\} \hat{\sigma}_y + \Delta/2 \hat{\sigma}_z$, where $\Phi(k) = \sum_{\delta} e^{i\delta \cdot k}$, and the sum is extended to the three vectors connecting first nearest neighbors. The Pauli matrices act on sublattice space; $t \approx 2.8$ eV [12] represents the intralayer hopping, while ∆ accounts for the energy difference between the two ions in the unit cell. In type-I structures, the atomic species on each sublattice are swapped in the bottom layer, so the sign of ∆ is inverted.

We now introduce the effect of the twist. The positions of the microscopic valleys are shifted to $K_\pm^{(t,\theta)} = R(\pm \theta/2) K_\pm$; similarly, the nearest-neighbors vectors on each layer read $\delta_i^{(t,\theta)} = R(\pm \theta/2) \delta_i$. Deviations of crystalline momentum from the valleys, $p = k - K_\pm^{(t,\theta)}$, represent the conjugate variable to the position of the envelope wave functions, $p \rightarrow -i \partial$. Changes in the atomic arrangement within the moiré period are incorporated as a modulation of the crystal field parameter, $\Delta(r)$. We parametrize the stacking configurations by a vector field $\mathbf{d}(r)$ defined up to a microscopic lattice translation, see Fig. 1. Thus, $\Delta(r)$ must be a functional of $\mathbf{d}(r)$ with the periodicity of the original Bravais lattice; a generic first-star expansion compatible with 3-fold symmetry reads

$$\Delta[\mathbf{d}] = \Delta_0 + \sum_{i=1}^{3} \{ \Delta_1 \cos(g_i \cdot \mathbf{d}) + \Delta_2 \sin(g_i \cdot \mathbf{d}) \}. \tag{2}$$

In the moiré pattern, the stacking configuration varies as $\mathbf{d}(r) = 2 \sin \frac{\theta}{2} \mathbf{z} \cdot \mathbf{r} + \mathbf{u}$. The modulation on the scale of the moiré pitch, $L_m = a/(2 \sin \frac{\theta}{2})$, follows from the identity $g_i \cdot \mathbf{d}(r) = G_i \cdot (\mathbf{r} - \mathbf{u})$, where $\mathbf{u} = 2 \times \mathbf{u} / (2 \sin \frac{\theta}{2})$ represents the center of the beating pattern corresponding to a AA’ or AA local stacking. The parameters $\Delta_{[0,1,2]}$ can be extracted from fittings to band structure calculations of hBN bilayers in high-symmetry stackings [13]; we take $\Delta_0 = 4.10$ eV, $\Delta_1 = -0.01$ eV, and $\Delta_2 = 0.08$ eV for type-I structures, and $\Delta_0 = 4.15$ eV, $\Delta_1 = -0.02$ eV, and $\Delta_2 = 0$ (due to symmetry, see Fig. 1) for type II.

For simplicity, we retain only interlayer scattering events between atoms sitting on top of each other conserving crystalline momentum (i.e., umklapp scattering is neglected). The modulation on the moiré period comes from the shift in the positions of the microscopic valleys. For each type of moiré structure, we need to consider three different parameters characterizing interlayer hoppings in boron-boron, nitrogen-nitrogen, and boron-nitrogen dimers. The interlayer tunneling matrix acquires the following general structure:

$$\hat{T}_\zeta(r) = \sum_{n=0}^{\infty} \hat{T}^{(n)}_\zeta e^{i\kappa_n \cdot (r - \mathbf{u})}, \text{ with } \hat{T}^{(n)}_\zeta = \hat{T}_0 e^{\frac{i\pi}{2} \kappa_n \cdot \sigma_z}, \tag{3b}$$

and $\kappa_n = R(n2\pi/3)K_\pm$ with $K_\pm = K_\pm' - K_\pm''$. For type-I structures, we obtain $\hat{T}_0 = w_{AA'} \hat{1} + w_{AB'} \hat{\sigma}_+ + w_{BA} \hat{\sigma}_-$, with $\hat{\sigma}_\pm = (\hat{\sigma}_x \pm i \hat{\sigma}_y) / 2$ and $w_{AA'} = 100$ meV, $w_{AB'} = 300$ meV, and $w_{BA} = 50$ meV extracted from band structure calculations [13]. In the case of type-II structures, we have $\hat{T}_0 = w_{BB'} (1 + \hat{\sigma}_z) / 2 + w_{NN} (1 - \hat{\sigma}_z) / 2 + w_{AB} \hat{\sigma}_x$, where we take $w_{BB'} = w_{AB'}$, $w_{NN} = w_{BA'}$, and $w_{AB} = 200$ meV.

The valley and superlattice translation symmetry allows us to diagonalize the Hamiltonian in a basis of (quasi-)Bloch states of the form

$$\psi_{\zeta, q}(r) = \frac{e^{i(q - \kappa_{\pm}) \cdot (r - \mathbf{u})}}{\sqrt{A}} \sum_{G} \psi_{\zeta, G}(q) e^{iG \cdot (r - \mathbf{u})}, \tag{4}$$
where $A$ is the total area of the system and $\alpha$ ($\mu$) are the internal sublattice (layer) degrees of freedom. Note that this gauge choice explicitly removes the dependence of the Hamiltonian on the beating pattern coordinate $\mathbf{u}$, which represents a soft (phason) mode of incommensurate structures [14]. The model also incorporates the $D_3$ symmetry of such structures, as shown in Fig. 1(c). To avoid redundancies, quasi-momenta $\mathbf{q}$ must be restricted to the Brillouin zone of the moiré superlattice defined by the primitive beating pattern wavevectors, so that superpositions with $\mathbf{q}$ and $\mathbf{q} + \mathbf{G}$ represent the same electronic state; we impose periodic boundary conditions, $u_{\gamma,\mathbf{L}}^{\alpha,\mu}(\mathbf{q} + \mathbf{G}) = u_{\gamma,\mathbf{L}}^{\alpha,\mu}(\mathbf{q})$. In this scheme, microscopic valleys are folded into the moiré Brillouin zone corners, $\mathbf{K}^{t}_{\pm} \equiv \pm \kappa_t = \pm (2\mathbf{G}_2 + \mathbf{G}_1)/3$ and $\mathbf{K}^{b}_{\pm} \equiv \pm \kappa_b = \pm (2\mathbf{G}_1 + \mathbf{G}_2)/3$. In practice, the sum in Eq. (4) is truncated to a finite number of harmonics, $n_G$. The coefficients $w_{\gamma,\mathbf{L}}^{\alpha,\mu}(\mathbf{q})$ can be arranged in a column vector, $|u_{\gamma,\mathbf{q}}\rangle$, defining a $4 n_G \times 4 n_G$ matrix to diagonalize for each value of $\mathbf{q}$.

Narrow bands are formed in a broad range of twist angles, see Fig. 2. We have chosen commensurate angles such that the moiré translation symmetry is exact and our results can be directly compared to microscopic calculations [15, 16]. For twist angles of $3^\circ$ or smaller, the bandwidth of the lowest conduction and valence bands is only of a few meVs. In our calculations, we have included the full tight-binding dispersion of monolayer hBN. Linearizing $\phi(\mathbf{k})$ and neglecting the rotation of the sublattice basis does not change appreciably the bands. In that situation, the spectrum of twisted bilayer graphene recovers the electron-hole symmetry of the Dirac Hamiltonian [17]. For hBN, however, this symmetry is absent due to the physical inequivalence of the two sublattices.

Panels (c) and (d) show the total amplitude (summed on sublattice and layer) of the envelope wave functions of the lowest-energy flat bands at the $\gamma$ point within a region containing four supercells [see Fig. 1(a1) for a comparison]. Profiles of similar symmetry are obtained at $\kappa_{t,b}$ points. In type-I structures, the first conduction band is localized on boron sites of AB'-stacked regions, while the wave function of the first valence band is mostly on nitrogen sites of BA' stackings. In type-II structures, however, the wave functions are both localized in regions of AA stacking. Just like in type-I structures, the conduction band is mostly localized on boron sites, while the valence band is on the nitrogen atoms. In both cases the wave function have equal weight in the top and bottom layer, as prescribed by $C_2$ symmetry.

We can see from these results that the emergence of flat bands in twisted bilayers of hBN and graphene responds to two different mechanisms of electron confinement. In hBN, the lowest flat bands result from resonant tunneling in stacking regions where atoms of the same polarity lie on top of each other. For graphene, however, a more subtle quantum interference process takes place at certain

![FIG. 2. Band structure and envelope wave functions of twisted hBN bilayers.](image-url)
(magic) twist angles. The most dramatic manifestation of the different nature of confinement takes place if we set the couplings $w_{AB} = w_{BA} = 0$: the lowest-energy flat bands of hBN disappear, whereas the analogous simplification in twisted bilayer graphene gives rise to completely flat bands at the magic angle [19].

We turn now to the calculation of the chiral response of the system. Light (of frequency $\omega$ and at normal incidence) induces in-plane electrical currents in the material, which can be decomposed in layer-resolved components $\hat{j}^{e,b}(\omega)$. The conjugate forces are the electric fields at the top and bottom layers, $E^{e,b}(\omega)$. Following Ref. [13], we express currents and fields in terms of total and counterflow components, specifically, $\hat{j} = \hat{j}^{e} + \hat{j}^{b}$ and $E^{(e,b)} = (E^{(e)} - E^{(b)})/2$ for the currents, and $E = (E^{(e)} + E^{(b)})/2$ and $E^{(e,b)} = E^{(e)} - E^{(b)}$ for the fields. Within this framework, the most general constitutive relations allowed by $D_3$ and time-reversal symmetries are

$$\begin{pmatrix} \sigma_0(\omega) & \sigma_\varepsilon(\omega) \\ -\sigma_\varepsilon(\omega) & \sigma_{\varepsilon\varepsilon}(\omega) \end{pmatrix} = \begin{pmatrix} \mathbf{E} \\ -\mathbf{E}^{(e)} \end{pmatrix}.$$

(5)

Here $\sigma_{(e,b)}(\omega)$ is the total (counterflow) conductivity, characterizing the longitudinal response of the system; and $\sigma_\varepsilon$ is the chiral conductivity, accounting for the transverse flow of charge in one layer generated by the electric field in the other layer. The latter response arises from the chirality of the structure and governs the optical activity.

We compute the total and chiral conductivities from the Kubo formula. The contributions from interband transitions are given by the regular part of current-current correlation functions with suitable combinations of coordinate and layer indices in each case, see [18] for details. The final result (including spin degeneracy) reads

$$\sigma_i(\omega) = \sum_{n,m} \int \frac{d\mathbf{q}}{(2\pi)^2} \frac{4e^2 c}{\hbar \omega + \epsilon_{n+q}^+ - q^+_{n+q}} \bar{\epsilon}_{n+q,a}(f_{n+q}^a - f_{n+q}^a),$$

(6a)

with

$$\bar{\epsilon}_{n+q,a}(\mathbf{q}) = \frac{\hbar^2}{(2\pi)^2} \langle |u_{n,\mathbf{q}}^{\ast}|, |v_{n,\mathbf{q}}^{\ast}|, |u_{n+q,\mathbf{q}}^{\ast}|, |v_{n+q,\mathbf{q}}^{\ast}| \rangle^2 / \left( \epsilon_{n+q,a}^+ - \epsilon_{n+q,a}^- \right)^2,$$

(6b)

$$\bar{\epsilon}_{c,n,m}(\mathbf{q}) = \frac{\hbar^2}{(2\pi)^2} \langle |u_{n,\mathbf{q}}^{\ast}|, |v_{n,\mathbf{q}}^{\ast}|, |u_{m,\mathbf{q}}^{\ast}|, |v_{m,\mathbf{q}}^{\ast}| \rangle^2 / \left( \epsilon_{c,n,m}^+ - \epsilon_{c,n,m}^- \right)^2,$$

(6c)

In these expression $f_{n+q}^{a,b}$ are the Fermi-Dirac occupation numbers for (quasi-)Bloch electrons in band $n$ with quasimomentum $\mathbf{q}$ within valley $\zeta = \pm 1$ and $\epsilon_{n+q,a}^{\pm}$ is the corresponding band energy. We have also introduced oscillator strengths containing matrix elements of the total velocity, $\hat{\mathbf{v}}_\zeta \equiv (-i/\hbar)[\mathbf{f}, \hat{\mathbf{F}}_\zeta]$, and layer-resolved velocity operators, $\hat{\mathbf{v}}_{\zeta,m} \equiv \hat{P}_m \hat{\mathbf{v}}_\zeta \hat{P}_m$, where $\hat{P}_m$ are projectors on layer subspace labeled by $\mu$. The sum is on band indices and the integration is over the moiré Brillouin zone.

Sharp resonances in the real part of the total conductivity appear within the nominal gap of monolayer hBN, as shown in Fig. 3(a). They are associated with optical transitions between the low-energy flat bands. Remarkably, the chiral conductivity in type-II structures is comparable (and even larger for small twist angles) to the total conductivity. This is manifested in the CD, which is defined as [21]

$$\text{CD} = \frac{\sigma^- - \sigma^+}{(\sigma^+ + \sigma^-)/2} \approx \frac{\omega_{20} \delta \sigma}{c} \text{Re} \sigma_\varepsilon,$$

(7)

where $\sigma^\pm = \sigma_{\varepsilon\varepsilon} \pm \sigma_\varepsilon^2/\sigma_{\varepsilon\varepsilon}^2$ is the absorbance of right- (left-) circularly polarized light [18]. In the above expression $\omega_{20} \approx 3.6$ Å is the interlayer distance and $c$ is the speed of light. Figure 3 (b) shows the CD of chiral partners with twist angles $\theta = 7.34^\circ$. In both types of structures, the CD is resonant with the transition between the lowest-energy flat bands. The CD can be made to be larger by decreasing the twist angle, but at the expense of the overall absorption signal.

Surprisingly, the two types of moirés give rise to significantly different chiral responses. The circular dichroism of the type-I structure is of the same order as in twisted bilayer graphene (≈ 20 meV at the first resonance) [8, 9], but in type II is one order of magnitude larger. This difference lies in the existence of some approximate symmetries not contained in the space group. In twisted bilayer graphene, the approximate electron-hole symmetry suppresses the optical transitions between the saddle
points of the lowest-energy bands \[17\] , where most of the density of states is concentrated; similarly, when the symmetry is enforced by neglecting the rotation of the sublattice basis, the interband contribution to the chiral conductivity cancels exactly \[9\]. As we mentioned before, the electron-hole symmetry is never restored in the case of twisted hBN. However, we have found approximate pseudo-inversion symmetries that become exact when the interlayer hopping between atoms of different polarity, which is larger in type-II structures, is neglected \[18\]. Enforcing the symmetries does not change much the lowest bands but leads to a cancellation of the chiral conductivity \[18\]. The approximate pseudo-inversion symmetry in type-II structures also suppresses the optical transitions at the \( m \) point, diminishing the total conductivity, while the same is not true in type-I structures \[15\]. All these factors conspire to give a CD signal systematically larger in type-II structures.

In conclusion, twisted bilayers of polar materials such as hBN host flat bands without the requirement of a special magic-angle alignment. These bands are built from in-gap resonant states at regions of unnatural stacking, where atoms of the same species lie on top of each other. Their localization is therefore controlled by the interlayer tunneling rate between atoms of the same polarity. In the case of hBN, these flat bands are manifested as sharp resonances in the optical absorption spectra deep inside the original gap. Owing to their chiral structure, these materials absorb left- and right-handed circularly-polarized light differently. This chiral optical response is engendered by current counterflows controlled by interlayer tunneling between atoms of opposite polarity. We believe that our predictions for hBN are within experimental reach, provided that the formation of flat bands contribute to a 25%-reduction of the optical gap. In particular, the CD response in the ultraviolet can be used not only to identify the magnitude and sign of the twist angle, but also to distinguish between the two types of moiré structures with different symmetry. At lower frequencies, the response will be dominated by phonons, excitons and other hybrid quasiparticles strongly influenced by the moiré superlattice. Their contribution to the chiral optical response is an interesting subject for future research.

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Supplemental Information: Flat bands and chiral optical response of twisted bilayer insulators

We derive Eqs. (6) of the main text from the Kubo formula and the leading contribution to the circular dichroism in Eq. (7). We analyze the presence of approximate pseudo-inversion symmetries in the Hamiltonian, leading to cancellations in the circular dichroism.

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Calculation of the optical conductivity

Hamiltonian

The Hamiltonian can be written as \( \hat{H} = \hat{H}_0 + \hat{V}(t) \), where \( \hat{H}_0 \) describes the stationary electronic spectrum and \( \hat{V}(t) \) represents the coupling with the external field. The goal is to compute the response of the system to linear order in the perturbation \( \hat{V}(t) \).

In the continuum model, the first term can be written as

\[
\hat{H}_0 = \sum_{\zeta = \pm 1} \int dr \, \hat{\psi}^\dagger_\zeta(r) \hat{\mathcal{H}}_\zeta(r) \hat{\psi}_\zeta(r),
\]

where \( \hat{\psi}_\zeta(r) \) is the electron field operator in valley \( \zeta \). Note that \( \hat{\psi}^\dagger_\zeta(r) \) is in fact a 4-column vector containing fermion operators that create an electron in position \( r \) of sublattice \( \alpha \) and layer \( \mu \). In the continuum model, these operators are defined by a \( \mathbf{k} \cdot \mathbf{p} \) expansion of the true, microscopic field operators around valleys \( \zeta \),

\[
\hat{\Psi}^{\alpha,\mu}(\mathbf{r}) = \sum_{\zeta = \pm 1} f^{\alpha,\mu}_\zeta(\mathbf{r}) \hat{\psi}^{\alpha,\mu}_\zeta(\mathbf{r}),
\]

where \( f^{\alpha,\mu}_\zeta(\mathbf{r}) \) represents the fast (on the atomic scale) oscillating factors of the electronic wave function. In our tight-biding description of hBN, these fast oscillating factors are nothing but the Bloch wave functions at \( \mathbf{K}_\zeta^{(\mu)} \) points,

\[
f^{\alpha,\mu}_\zeta(\mathbf{r}) = \frac{1}{\sqrt{N}} \sum_i \Phi^{\alpha,\mu}_\zeta(\mathbf{r} - \mathbf{R}_i^{\alpha,\mu}),
\]

where \( \mathbf{R}_i^{\alpha,\mu} \) represent the positions of the atoms, the sum is extended to \( N \) microscopic cells, and \( \Phi^{\alpha,\mu}(\mathbf{r}) \) are Wannier functions of \( \pi \) orbitals in \( \alpha \) sublattice of layer \( \mu \).
The continuum Hamiltonian in the main text is written in a gauge of envelope wave functions such that the translational invariance is defined only up to a unitary rotation, namely,

$$\hat{H}_\zeta (r + R) = \hat{U}_\zeta (R) \hat{H}_\zeta (r) \hat{U}_\zeta^\dagger (R), \text{ with } \hat{U} (R) = \begin{bmatrix} e^{-i\zeta \kappa_\mu R} & 0 \\ 0 & e^{-i\zeta \kappa}\end{bmatrix}. \tag{11}$$

Here $R$ is a vector of the moiré superlattice such that $e^{iG \cdot R} = 1$. The unitary operator in layer space contains momentum boosts associated with the shift of the positions of the microscopic valleys. Bloch wave functions in this gauge satisfy

$$\psi^{\alpha,\mu}_{\zeta, q} (r + R) = e^{i(q - \zeta \kappa_\mu) \cdot R} \psi^{\alpha,\mu}_{\zeta, q} (r), \tag{12}$$

with $q$ restricted to the first Brillouin zone of the moiré superlattice. This form of Bloch theorem leads to the ansatz in Eq. (4) of the main text. The extra phase factor may look odd at first glance, but note that it is just a reminiscence of the fast oscillating factor of the microscopic wave function since $f^{\alpha,\mu}_\zeta (r + R) \approx e^{iK^{(\mu)}_\zeta \cdot R} f^{\alpha,\mu}_\zeta (r)$, where the identity is exhausted only when the moiré superlattice symmetry is microscopically exact. In that case, the folding scheme imposing $K_\zeta^{(\mu)} \equiv \zeta \kappa_\mu$ is exact and the microscopic wave function $\Psi^{\alpha,\mu}_{\zeta, q} (r) = \sum \zeta f^{\alpha,\mu}_\zeta (r) \times \psi^{\alpha,\mu}_{\zeta, q} (r)$ satisfies the Bloch theorem in the usual form, $\Psi^{\alpha,\mu}_{\zeta, q} (r + R) = e^{iQ_\alpha \cdot R} \Psi^{\alpha,\mu}_{\zeta, q} (r)$.

The translation of this discussion to second quantization is that the band Hamiltonian is diagonalized in a basis of operators of the form

$$\hat{c}_\zeta^{\alpha,\mu} (k) = \frac{1}{\sqrt{A}} \int dr e^{-i(k - i\zeta \kappa_\mu) \cdot r} \psi^{\alpha,\mu}_{\zeta, q} (r). \tag{13}$$

In this definition, momentum $k$ is not restricted to the first Brillouin zone of the moiré superlattice, but it is convenient to introduce such restriction in the expression of the Hamiltonian. In order to do so, let us introduce the following ket notation:

$$|q\rangle \otimes |\zeta, \alpha, \mu, G\rangle \equiv \left[ \hat{c}_\zeta^{\alpha,\mu} (q + G) \right]^\dagger |0\rangle, \tag{14a}$$

such that $\langle r | q \rangle = e^{iq \cdot r}$, and

$$\langle r | \zeta, \alpha, \mu, G \rangle = e^{i(G - \zeta \kappa_\mu) \cdot r} \sqrt{N_m}, \tag{14b}$$

$$\langle r | \zeta, \alpha, \mu, G \rangle = e^{i(G - \zeta \kappa_\mu) \cdot r} \sqrt{A_m}, \tag{14c}$$

where $N_m$ is the number of moiré supercells and $A_m$ is their area. Here we have just written momentum $k$ as $k = q + G$, with $q$ restricted to the first moiré Brillouin zone (defining the usual Bloch factor), and the Fourier replica in the reciprocal lattice $G$ has been incorporated as a new internal quantum number. The Hamiltonian reads then

$$\hat{H}_0 = \sum_{\zeta = \frac{\pi}{2} \pm} \sum_{q \in \text{mBZ}} \hat{H}_\zeta (q) \otimes |q\rangle \langle q|, \tag{15}$$

where $\hat{H}_\zeta (q)$ is a $4n_G \times 4n_G$ matrix of the form (in the case of type-II structures)

$$\hat{H}_\zeta (q) = \sum_{\alpha, \beta} \sum_{\{G\}} \sum_\mu \left[ t \Re \{ \phi_\mu (q - \zeta \kappa_\mu + G) \} \hat{\sigma}_z + t \Im \{ \phi_\mu (q - \zeta \kappa_\mu + G) \} \hat{\sigma}_y + \frac{\Delta_0}{2} \hat{\sigma}_z \right] |\zeta, \alpha, \mu, G\rangle \langle \zeta, \beta, \mu, G|$$

$$+ \frac{\Delta_1 - i\Delta_2}{4} \sum_{\alpha, \beta} \sum_{\{G\}} \sum_\mu \sum_{i=1,2,3} \hat{\sigma}_z |\zeta, \alpha, \mu, G + G_i\rangle \langle \zeta, \beta, \mu, G| + \text{h.c.} \tag{16}$$

$$+ \sum_{\alpha, \beta} \sum_{\{G\}} \sum_{n=0,1,2,3} \sum_\mu T^{(n)}_{\alpha, \beta} |\zeta, \alpha, t, G + \zeta Q_n\rangle \langle \zeta, \beta, t, G| + \text{h.c.},$$

with $Q_0 = 0, Q_1 = G_2, Q_2 = -G_1$ and

$$\phi_\mu (p) \equiv \sum_{i=1,2,3} e^{i\delta^{(\mu)} \cdot (p + K^{(\mu)}_\zeta)} \tag{17}.$$
In the case of type-I structures, note that the parameters $\Delta_i$ enter with opposite sign in the bottom layer sector. The solution of the diagonalization problem of this matrix defines the band energies $\varepsilon_{\zeta}^n$, and the coefficients $u_{n,\zeta,\mathbf{G}}(\mathbf{q})$ in the ansatz of Eq. (4) of the main text, which are just the $4n_{\mathbf{G}}$ components of the corresponding eigenvector:

$$
\begin{equation}
|u_{n,\zeta}(\mathbf{q})\rangle = \sum_{\alpha} \sum_{\mu} \sum_{\{\mathbf{G}\}} u_{n,\zeta,\mathbf{G}}^{\alpha,\mu}(\mathbf{q}) |\zeta,\alpha,\mu,\mathbf{G}\rangle. 
\end{equation}
$$

In second quantization, this last relation amounts to

$$
\hat{c}_{\zeta}^{\alpha,\mu}(\mathbf{q} + \mathbf{G}) = \sum_{n} u_{n,\zeta,\mathbf{G}}^{\alpha,\mu}(\mathbf{q}) \hat{c}_{n,\zeta}(\mathbf{q}),
$$

where the summation is in band indices. Here $\hat{c}_{n,\zeta}(\mathbf{q})$ are fermion operators associated with excitations in band $n$ of valley $\zeta$ with quasi-momentum $\mathbf{q}$,

$$
\hat{c}_{\zeta,n}^{\dagger}(\mathbf{q}) |0\rangle = |\mathbf{q}\rangle \otimes |u_{n,\zeta}(\mathbf{q})\rangle.
$$

The relation between operators in Eq. (19) is univocal as long as we define boundary conditions in reciprocal space. Note that if a finite number of Fourier replica $u_{\mathbf{G}}$ is included, then the matrix Hamiltonian is periodic in the momentum space up to a unitary transformation,

$$
\hat{\mathcal{H}}_{\zeta}(\mathbf{q} + \mathbf{G}_i) = \hat{U}_{\mathbf{G}_i} \hat{\mathcal{H}}_{\zeta}(\mathbf{q}) \hat{U}_{\mathbf{G}_i}^{\dagger}.
$$

This unitary transformation reads

$$
\hat{U}_{\mathbf{G}_i} = \sum_{\alpha} \sum_{\mu} \sum_{\{\mathbf{G}\}} |\zeta,\alpha,\mu,\mathbf{G} - \mathbf{G}_i\rangle \langle \zeta,\beta,\mu,\mathbf{G}|.
$$

It follows that if $|u_{n,\zeta}(\mathbf{q})\rangle$ is an eigenvector of $\hat{\mathcal{H}}_{\zeta}(\mathbf{q})$ with eigenvalue $\varepsilon_{\zeta}^n$, then $\hat{U}_{\mathbf{G}_i} |u_{n,\zeta}(\mathbf{q})\rangle$ is an eigenvector of $\hat{\mathcal{H}}_{\zeta}(\mathbf{q} + \mathbf{G}_i)$ with the same eigenvalue. Periodic boundary conditions amounts to the identification

$$
|u_{n,\zeta}(\mathbf{q} + \mathbf{G}_i)\rangle \equiv \hat{U}_{\mathbf{G}_i} |u_{n,\zeta}(\mathbf{q})\rangle \mapsto u_{n,\zeta,\mathbf{G}_i}^{\alpha,\mu}(\mathbf{q} + \mathbf{G}_i) = u_{n,\zeta,\mathbf{G} + \mathbf{G}_i}^{\alpha,\mu}(\mathbf{q}), \hat{c}_{n,\zeta}(\mathbf{q} + \mathbf{G}_i) = \hat{c}_{n,\zeta}(\mathbf{q}).
$$

Finally, as the continuum model is derived from a tight-binding Hamiltonian, it is natural to introduce the coupling with external fields via a Peierls substitution, $\hbar \mathbf{q} \rightarrow \hbar \mathbf{q} - e \mathbf{A}(t, \mathbf{r}, z)$. Before writing down the perturbation, some remarks are in order. First, in our model, the $z$ coordinate only enters through the internal layer number, so instead of dealing with a function of $z$, we should consider the values of the external field in the top and bottom layer,

$$
\mathbf{A}^{(t,b)}(t, \mathbf{r}) = \mathbf{A}(t, \mathbf{r}, \pm \frac{z}{2}).
$$

Second, as the model only retains interlayer hopping processes between atoms sitting on top of each other and these processes do not carry lateral currents, the total microscopic currents can be also decomposed into two layer-resolved components. To the leading order in the external field, the perturbation reads

$$
\hat{\mathcal{V}}(t) = -\sum_{\mu} \int d\mathbf{r} \hat{j}_{p}^{(\mu)}(\mathbf{r}) \cdot \mathbf{A}^{(\mu)}(t, \mathbf{r}) = -\sum_{\mu} \sum_{\mathbf{k}} \hat{j}_{p}^{(\mu)}(\mathbf{k}) \cdot \mathbf{A}_{k}^{(\mu)}(t),
$$

where $\mathbf{A}_{k}^{(\mu)}(t)$ are the Fourier components of the external field,

$$
\mathbf{A}_{k}^{(\mu)}(t) = \frac{1}{\sqrt{A}} \int d\mathbf{r} \mathbf{A}^{(\mu)}(t, \mathbf{r}) e^{-i\mathbf{k} \cdot \mathbf{r}},
$$

and $\hat{j}_{p}^{(\mu)}(\mathbf{k})$ is the paramagnetic term of the layer-resolved current operator in momentum representation,

$$
\hat{j}_{p}^{(\mu)}(\mathbf{k}) = \frac{e}{\sqrt{A}} \sum_{n_1,n_2} \sum_{\zeta} \sum_{\mathbf{q} \in \text{mBZ}} \langle u_{n_1,\zeta}(\mathbf{k} + \mathbf{q}) | \hat{\theta}_{\zeta,\mu} | u_{n_2,\zeta}(\mathbf{q}) \rangle \hat{c}_{n_1,\zeta}(\mathbf{k} + \mathbf{q}) \hat{c}_{n_2,\zeta}(\mathbf{q}).
$$

In this last expression we have used the relation in Eq. (19) along with the representation in momentum space of the (layer-resolved) velocity operator, which is just

$$
\hat{\theta}_{\zeta} = \frac{1}{\hbar} \tilde{\mathbf{p}}_{\zeta} \hat{\mathcal{H}}_{\zeta}(\mathbf{q}).
$$

Next, we elaborate a bit more on the structure of the current operators before addressing the calculation in the last subsection.
Current operators in the continuum model

The layer-resolved density operators can be defined as usual in terms of field operators,

\[ \hat{\rho}^{(\mu)}(r) = e \sum_{\zeta} \sum_{\alpha} \left[ \hat{\psi}^{(\alpha,\mu)}_{\zeta}(r) \right]^{\dagger} \hat{\psi}^{(\alpha,\mu)}_{\zeta}(r) = \frac{1}{\sqrt{A}} \sum_{k} e^{-ikr} \hat{\rho}^{(\mu)}(k), \]

where

\[ \hat{\rho}^{(\mu)}(k) = \frac{e}{\sqrt{A}} \sum_{\zeta} \sum_{\alpha} \sum_{p} \left[ \hat{c}^{(\alpha,\mu)}_{\zeta}(k + p) \right]^{\dagger} \hat{c}^{(\alpha,\mu)}_{\zeta}(p). \]

Note that the summations in momentum are not restricted in either of the last two expressions. Introducing the relations in Eq. (19) and the ket notation discussed before, we arrive at the compact expression

\[ \hat{\rho}^{(\mu)}(k) = \frac{e}{\sqrt{A}} \sum_{n_{1},n_{2}} \sum_{\zeta} \sum_{q \in \text{mBZ}} \langle u_{n_{1},\zeta}(k + q)| \hat{p}_{\mu} |u_{n_{2},\zeta}(q) \rangle \hat{c}_{n_{1},\zeta}^{\dagger}(k + q) \hat{c}_{n_{2},\zeta}(q). \]

Charge continuity in momentum space implies

\[ \sum_{\mu} \hat{j}^{(\mu)}(k) = \frac{i}{\hbar} \left[ \hat{H}, \sum_{\mu} \hat{\rho}^{(\mu)}(k) \right] = i \sum_{\mu} k \cdot \hat{j}^{(\mu)}(k), \]

where \( \hat{j}^{(\mu)} \) the total (paramagnetic plus diamagnetic) layer-resolved current. The commutator reads (expanding the band Hamiltonian up to first order)

\[ \frac{i}{\hbar} \left[ \hat{H}, \sum_{\mu} \hat{\rho}^{(\mu)}(k) \right] = i \sum_{\mu} k \cdot \hat{j}^{(\mu)}(k) - \frac{i}{\hbar} \sum_{\mu,\nu} \sum_{p} \hat{A}^{(\nu)}(t) \cdot \left[ \hat{j}^{(\nu)}(p), \hat{\rho}^{(\mu)}(k) \right]. \]

The commutator in the second term of the right-hand side of the last equation reads (up to the same order in external momentum)

\[ \left[ \hat{j}^{(\nu)}(p), \hat{\rho}^{(\mu)}(k) \right] = \frac{e^{2} \hbar k_{i} \hat{e}_{i}}{A} \sum_{n_{1},n_{2}} \sum_{\zeta} \sum_{q \in \text{mBZ}} \langle u_{n_{1},\zeta}(p + k + q)| \hat{p}_{\mu} \hat{\Theta}_{\zeta}^{i,j} \hat{p}_{\mu} |u_{n_{2},\zeta}(q) \rangle \hat{c}_{n_{1},\zeta}^{\dagger}(p + k + q) \hat{c}_{n_{2},\zeta}(q), \]

where summation over spatial (latin) indices is assumed hereafter and we have introduce the inverse effective-mass tensor,

\[ \hat{\Theta}_{\zeta}^{i,j} = \frac{1}{\hbar^{2}} \partial_{q_{i}} \partial_{q_{j}} \hat{F}_{\zeta}(q). \]

From this last result we can identify the total layer-resolved current operator \( \hat{j}^{\mu} = \hat{j}^{\mu}_{p} + \hat{j}^{\mu}_{d} \), where the diamagnetic term reads

\[ \hat{j}^{\mu}_{d}(k) = -\frac{e^{2} \hbar k_{i} \hat{e}_{i}}{A} \sum_{n_{1},n_{2}} \sum_{\zeta} \sum_{p} \sum_{q \in \text{mBZ}} \left( \hat{e}_{j} \cdot \hat{A}^{(\mu)}(t) \right) \langle u_{n_{1},\zeta}(k + p + q)| \hat{\Theta}_{\zeta}^{i,j} |u_{n_{2},\zeta}(q) \rangle \hat{c}_{n_{1},\zeta}^{\dagger}(k + p + q) \hat{c}_{n_{2},\zeta}(q), \]

where we have used \( \hat{P}_{\mu} \hat{\Theta}_{\zeta}^{i,j} \hat{P}_{\nu} = \hat{P}_{\mu} \hat{\Theta}_{\zeta}^{i,j} \hat{P}_{\nu} \delta_{\mu,\nu} \) since the operator is diagonal in layer indices and \( \hat{\Theta}_{\zeta}^{i,j} \equiv \hat{P}_{\mu} \hat{\Theta}_{\zeta}^{i,j} \hat{P}_{\mu} \). Note, however, that the diamagnetic term only enters in the intraband contribution to the total conductivity. If we retained linear terms in \( q \) only, the diamagnetic term would be exactly zero.

Kubo formulas

We proceed now to the perturbative calculation. Let us consider normal incidence and assume that the light spot is large compared to the moiré pitch. We work in the Weyl gauge and assume the dipolar approximation for the
incident field, $A(t, \mathbf{r})^{(\mu)} = A^{(\mu)}(t)$, such that $E^{(\mu)}(t) = -\dot{A}^{(\mu)}(t)$. As customary, we will replace the incident fields by the total fields at the end of the calculation, neglecting local field corrections generated by induced currents.

Under these assumptions, the perturbation contains only the $k = 0$ component of the paramagnetic current. In frequency domain, we just have

$$
\tilde{V}(\omega) = \frac{i e}{\omega} \sum_{\mu} \sum_{n_1, n_2} \sum_\zeta \sum_\mathbf{q} \langle u_{n_1, \zeta} | E^{(\mu)}(\omega) \cdot \hat{n}_{\zeta, \mu} | u_{n_2, \zeta} \rangle \hat{c}_{n_1, \zeta}^\dagger \hat{c}_{n_2, \zeta}(\mathbf{q}).
$$

(37)

We want to compute the linear response in the total current, provided that the latter is also uniform in space. In the following expressions, we are going to employ the following notation for the matrix elements evaluated at the same point of the Brillouin zone,

$$
\langle u_{n_1, \zeta} | \hat{n}_{\zeta, \mu} | u_{n_2, \zeta} \rangle \equiv \delta^{(n_1, n_2)}.
$$

(38)

Similarly, all the summations in $\mathbf{q}$ from this point on are restricted to the moiré Brillouin zone.

For the layer-resolved current, first-order perturbation theory in the external fields gives (recovering the spin degeneracy)

$$
J^{(\mu)}_i(\omega) = \frac{i}{\omega} \sum_\nu x^{i,j}_{\mu,\nu}(\omega) E^{(\nu)}_j(\omega) + \frac{2ie^2}{\omega A} \sum_n \sum_\zeta \sum_\mathbf{q} \sum_\mu \sum_\nu D^{i,j;\mu,\nu}_\zeta(n_1, n_2) f_{\zeta,\mu,\mathbf{q}} E^{(\mu)}_j(\omega),
$$

(39)

where we have introduced the retarded (paramagnetic) current-current correlation function given by

$$
\chi^{i,j}_{\mu,\nu}(\omega) = \frac{2e^2}{A} \sum_{n_1, n_2} \sum_\zeta \sum_\mathbf{q} \left( f_{\zeta,\mu,\mathbf{q}}^{n_1} - f_{\zeta,\nu,\mathbf{q}}^{n_2} \right) \frac{v_{\zeta,\mu,\mathbf{q}}^{i(n_1, n_2)} v_{\zeta,\nu,\mathbf{q}}^{j(n_1, n_2)}}{\hbar \omega + i0^+ + \varepsilon_{\zeta,\mu,\mathbf{q}}^{n_1} - \varepsilon_{\zeta,\nu,\mathbf{q}}^{n_2}}.
$$

(40)

$D_3$ and time-reversal symmetries impose the following relations:

$$
\begin{align}
\chi^{x,x}_{t,t}(\omega) &= \chi^{y,y}_{b,b}(\omega) = \chi^{x,x}_{b,b}(\omega) = \chi^{y,y}_{b,b}(\omega), \\
\chi^{x,y}_{t,t}(\omega) &= \chi^{y,x}_{b,b}(\omega) = \chi^{y,x}_{b,t}(\omega) = 0, \\
\chi^{x,y}_{b,b}(\omega) &= \chi^{y,x}_{b,b}(\omega) = 0, \\
\chi^{x,y}_{t,b}(\omega) &= -\chi^{x,y}_{b,t}(\omega).
\end{align}
$$

(41a-41d)

Comparing the result in Eq. [39] with the constitutive relations in the main text allows us to write down Kubo formulas for the total and chiral conductivities,

$$
\begin{align}
\sigma_0(\omega) &= \frac{i}{\omega} \sum_{\mu,\nu} \chi^{x,x}_{\mu,\nu}(\omega) + \frac{2ie^2}{\omega A} \sum_n \sum_\zeta \sum_\mathbf{q} \sum_\mu \sum_\nu D^{x,x;\mu,\nu}_\zeta(n_1, n_2) f_{\zeta,\mu,\mathbf{q}}^n, \\
\sigma_c(\omega) &= \frac{i}{\omega} \chi^{x,y}_{t,b}(\omega).
\end{align}
$$

(42a-42b)

In order to arrive at the final formulas in Eqs. (6) of the main text, we just need to isolate the interband contributions. The kernel in the correlator can be decomposed in simple fractions as

$$
\frac{1}{\hbar \omega + i0^+ + \varepsilon_{\zeta,\mu,\mathbf{q}}^{n_1} - \varepsilon_{\zeta,\nu,\mathbf{q}}^{n_2}} = \frac{1}{\varepsilon_{\zeta,\mu,\mathbf{q}}^{n_1} - \varepsilon_{\zeta,\nu,\mathbf{q}}^{n_2}} + \frac{\hbar \omega}{(\varepsilon_{\zeta,\mu,\mathbf{q}}^{n_1} - \varepsilon_{\zeta,\nu,\mathbf{q}}^{n_2})^2} + \frac{\hbar^2 \omega^2}{(\hbar \omega + i0^+ + \varepsilon_{\zeta,\mu,\mathbf{q}}^{n_1} - \varepsilon_{\zeta,\nu,\mathbf{q}}^{n_2})^2}.
$$

(43)

The first term (along with the diamagnetic term in the case of the total conductivity) gives rise to the contribution due to intraband electron motion. The integration of the second term cancels, which can be demonstrated using the following identities imposed by time-reversal symmetry:

$$
\begin{align}
\varepsilon_{\zeta,\mu,\mathbf{q}}^{n_1} &= \varepsilon_{\zeta,-\mu,-\mathbf{q}}^{n_1}, \\
\varepsilon_{\zeta,\mu,\mathbf{q}}^{n_1, n_2} &= -\varepsilon_{-\zeta,\mu,-\mathbf{q}}^{n_2, n_1}.
\end{align}
$$

(44a-44b)

The third term leads to the final result in Eqs. (6), where we have used time-reversal symmetry in order to simplify the sum in valley index.
Electromagnetic response

Constitutive relations

Here, we follow Ref. [10] and rewrite the constitutive relations in the main text in terms of local quantities, as we can assume \( z_0 \ll c/\omega \) for the relevant frequencies in the experiment. As in the beginning of the previous section, we write the macroscopic electric field as a function of the vertical coordinate, i.e., \( E(z, \omega) \). Expanding in series up to first order in \( z_0 \), we have

\[
E^{(t,b)}(\omega) \equiv E\left(\pm \frac{z_0}{2}, \omega\right) \approx E(0, \omega) \pm \frac{z_0}{2} \partial_z E(z, \omega) \big|_{z=0}. \tag{45}
\]

To leading order in \( z_0 \), we thus write

\[
E(\omega) \approx E(0, \omega), \tag{46a}
\]

\[
E^{(cf)}(\omega) \approx z_0 \partial_z E(z, \omega) \big|_{z=0}. \tag{46b}
\]

The macroscopic field is a solution of the Maxwell equations; in particular, from Faraday’s law

\[
\hat{z} \times \partial_z E(z, \omega) = i\omega B(z, \omega). \tag{47}
\]

We can thus relate \( E^{(cf)} \) with the averaged magnetic field \( B(\omega) \equiv B(0, \omega) \),

\[
E^{(cf)}(\omega) \approx i\omega z_0 B(\omega) \times \hat{z}. \tag{48}
\]

In this approximation, we can substitute the bilayer system by a purely 2D system placed at \( z = 0 \), where \( j(\omega) \) expresses the total charge current or, equivalently, the polarization density,

\[
P(\omega) = \frac{i}{\omega} j(\omega), \tag{49}
\]

while the counter-flow current can be used to coarse grain an orbital magnetization density, i.e.,

\[
M(\omega) = \frac{1}{2} z_0 \times j^{(t)}(\omega) - \frac{1}{2} z_0 \times j^{(b)}(\omega) = z_0 \times j^{(cf)}(\omega). \tag{50}
\]

The constitutive relations to leading order in \( z_0 \omega/c \) are readily found to be [10]

\[
P(\omega) = \frac{i}{\omega} \sigma_0(\omega) E(\omega) - z_0 \sigma_c(\omega) B(\omega), \tag{51a}
\]

\[
M(\omega) = i\omega z_0^2 \sigma_{cf}(\omega) B(\omega) + \sigma_c(\omega) z_0 E(\omega). \tag{51b}
\]

This kind of response is allowed by the chiral (purely rotational) symmetry of the moiré patterned material, which allows us to identify vectors \( (P, E) \) with pseudo-vectors \( (M, B) \). Systems resulting from a twist of opposite sign can be envisioned as partners of opposite chirality connected by the inversion/reflection symmetry broken by the relative rotation. In type-I structures, this symmetry is inversion \( i \) along the center of the axis connecting a common center of the hexagons. The total and counter-flow currents transform as

\[
i : j \rightarrow -j, \tag{52a}
\]

\[
i : j^{(cf)} \rightarrow j^{(cf)}. \tag{52b}
\]

The same applies to the conjugate forces. In type-II structures, the pertinent symmetry is mirror reflection, \( h \), then

\[
h : j \rightarrow j, \tag{53a}
\]

\[
h : j^{(cf)} \rightarrow -j^{(cf)}. \tag{53b}
\]

In both cases, it follows that

\[
i, h : \sigma, \sigma_{cf} \rightarrow \sigma, \sigma_{cf}, \tag{54a}
\]

\[
i, h : \sigma_c \rightarrow -\sigma_c. \tag{54b}
\]

Therefore, chiral partners present opposite CD.
FIG. 4. **Scheme for the derivation of the Fresnel coefficients.** The incident field is partially reflected and transmitted (and absorbed) by the bilayer interface. The bilayer (blue-red) is understood as a sheet (dashed black) with a coarse-grained chiral response.

**Fresnel coefficients**

We now derive the Fresnel coefficients that describe transmission and reflection upon incidence of a normal electromagnetic field on the twisted bilayer, as shown in Fig. 4. In the previous section, we have described how the twisted bilayer can be described as a coarse-grained 2D system with a chiral coupling resulting from the Hall drag counter-flow. We employ this insight to model the material system as an interface between two dielectric regions. From now on, we assume that the two regions are vacuum, but it is a trivial extension to consider an embedding medium or substrate.

The reflected and transmitted fields are related to the incident field as

\[ E_t = \hat{t} E_{in}, \quad E_r = \hat{r} E_{in}, \] (55)

where the transmission and reflection matrices read

\[ \hat{t} = \begin{bmatrix} t & t_{xy} \\ -t_{xy} & t \end{bmatrix}, \quad \hat{r} = \begin{bmatrix} r & r_{xy} \\ -r_{xy} & r \end{bmatrix}, \] (56)

and \( t, r, r_{xy}, t_{xy} \) are the Fresnel coefficients. Note that the structure of the matrices is imposed by \( D_3 \) and time reversal symmetries. We find the values of the Fresnel coefficients from the constitutive equations (51) together with the following boundary conditions at the interface:

\[ \hat{z} \times (H_2 - H_1) = -i\omega P, \] (57a)
\[ \hat{z} \times (E_2 - E_1) = i\mu_0 \omega M, \] (57b)

where the subscripts \( \{1, 2\} \) refer to the regions depicted in Fig. 4. The electromagnetic fields in each region are

\[ E_1 = E_{in} + E_r, \quad E_2 = E_t, \] (58a)
\[ H_1 = \frac{1}{\mu_0 c} \hat{z} \times (E_{in} - E_r), \quad H_2 = \frac{1}{\mu_0 c} \hat{z} \times E_t, \] (58b)

where the propagation direction of the incident field is assumed to be normal to the interface. Note that we take both the polarization and magnetization densities to depend only on the incident fields. After some algebra, the Fresnel coefficients are readily found to be

\[ r = -\sigma_0 \frac{\epsilon_0 c}{2\epsilon_0 c}, \quad t = 1 - \sigma_0 \frac{\epsilon_0 c}{2\epsilon_0 c}, \quad r_{xy} = 0, \quad t_{xy} = \frac{i\omega \sigma_0 \epsilon_0 c}{\epsilon_0 c^2}, \] (59)

where we have dropped the explicit dependence of the conductivity on the frequency for the sake of clarity.

**Circular dichroism**

The circular dichroism (CD) can be easily derived from the Fresnel coefficients. For a circularly polarized incident field \( E_{in}^\pm = E_{in}(\hat{x} \pm i\hat{y})/\sqrt{2} \), the transmitted and reflected fields are readily found by simply operating with the transmission and reflection matrices,

\[ E_t^\pm = t_\pm E_{in}^\pm, \quad E_r^\pm = r_\pm E_{in}^\pm, \] (60)
with
\[
\begin{align}
t_{\pm} &= t \pm it_{xy} = 1 - \frac{\sigma_0}{2\epsilon_0 c} \mp \frac{\omega_{z_0}}{\epsilon_0 c^2} \sigma_c, \\
r_{\pm} &= r \pm ir_{xy} = -\frac{\sigma_0}{2\epsilon_0 c}.
\end{align}
\]
Note that the reflection coefficient does not depend on the chiral conductivity. This implies that circular dichroism cannot be detected by reflectance-only measurements. One instead has to measure either transmission (i.e., ellipticity) or the absorption of the chiral samples. We focus on the latter to define the circular dichroism as a difference between the absorption of left- and right-circularly polarized light, i.e.,
\[
\text{CD} = \frac{\mathcal{A}_+ - \mathcal{A}_-}{(\mathcal{A}_+ + \mathcal{A}_+)/2},
\]
where \(\mathcal{A}_\pm = 1 - |r_\pm|^2 - |t_\pm|^2\) is the absorbance of the bilayer, which reads
\[
\mathcal{A}_\pm = \frac{1}{\epsilon_0 c} \text{Re} \left\{ \sigma \pm \frac{2\omega_{z_0}}{c} \sigma_c \mp \frac{\omega_{z_0}}{2\epsilon_0 c^2} \sigma_c^* \right\}.
\]
To first order in \(\omega_{z_0}/c\), the circular dichroism is found to be
\[
\text{CD} \approx \frac{\omega_{z_0}}{c} \frac{4 \text{Re} \sigma_c}{\text{Re} \sigma_0} - \frac{\omega_{z_0}}{\epsilon_0 c^2} \frac{\text{Re} \sigma_c^*}{\text{Re} \sigma_0}.
\]
Since \(|\sigma_0|/\epsilon_0 c \ll 1\), we drop the second term above and find the expression in the main text.

### Approximate symmetries and cancellations

In the context of twisted bilayer graphene, it is common to consider, instead of the full tight-binding dispersion, the linearized theory resulting from the expansion of \(\phi_{\mu}\) in Eq. (17) up to first order,
\[
\phi_{(t,b)}(p) = \sum_i e^{i\delta_{(t,b)}(p_{\mu} + K_{(t,b)})} \approx \sum_i \left( \delta_{(t,b)}(p) \right) e^{i\delta_{(t,b)}(p_{\mu})} K_{(t,b)} = \frac{\sqrt{3} a e^{\pm i\xi^2/2}}{2} (-\zeta p_x + ip_y).
\]
Furthermore, it is also common to neglect the extra phase \(e^{\pm i\xi^2/2}\), which only introduces a subleading correction \(\sim \theta^2\) in the bands. Figure 5(a) shows the band structures of hBN bilayers for the largest twist angle considered in the main text imposing these approximations. The band dispersion does not change much, particularly in the case of the lowest-energy bands. The agreement improves as the twist angle decreases.

These approximations have a more subtle effect on the electronic wave functions and the optical response of twisted bilayer graphene due to an electron-hole symmetry [17]. For the following discussion, let us simplify the notation by introducing Pauli matrices \(\hat{\mu}_i\) acting on layer degree of freedom such that, for example, layer projectors read just \(\hat{P}_l = (\hat{1} + \hat{\mu}_z)/2\), \(\hat{P}_b = (\hat{1} - \hat{\mu}_z)/2\). In this notation, the electron-hole symmetry, \(\{\hat{U}_{e-h}, \mathcal{H}\} = 0\), is implemented by the following anti-unitary operator,
\[
\hat{U}_{e-h} = i \hat{\mu}_y \otimes \hat{\sigma}_x \mathbb{K},
\]
where \(\mathbb{K}\) represents complex conjugation. This approximate symmetry, when enforced, suppresses the optical transitions between bands of opposite energies at the \(m\) point [17] and, in combination with \(C_2\) symmetries, can be used to prove that contributions from opposite points of the moiré Brillouin zone to the interband chiral conductivity cancel out [9].

As mentioned in the main text, the electron-hole symmetry is not present in twisted hBN due to the physical inequivalence of the two sublattices. In particular, in the case of type-I moiré structures, this is just due to the opposite sign of \(\Delta(r)\) in the top and bottom layers; in the case of type-II structures, due to the fact that \(w_{BB} \neq w_{NN}\). However, as we discuss next, there are approximate pseudo-inversion symmetries that give rise to cancellations of the chiral response and, in the case of type-II structures, to additional selection rules that suppress optical transitions at certain high-symmetry points of the moiré Brillouin zone.
FIG. 5. **Bands of twisted hBN at $\theta = 7.34^\circ$ in a simplified model.** (a) Band structures obtained by taking Eq. (65), neglecting also the twist-angle related phase. (b) The same by taking now $w_{AA'} = 0$ (type I) and $w_{AB} = 0$ (type II).

**Type I**

In the limit of small twist angles, the Hamiltonian of type-I structures can be simplified to

$$
\hat{\mathcal{H}}_\zeta (r) = \frac{i \sqrt{3} \mathcal{A} t}{2} \hat{\sigma}_\zeta \cdot \partial + \frac{\Delta (r)}{2} \hat{\mu}_z \otimes \hat{\sigma}_z + \frac{1}{2} \hat{\mu}_x \otimes \left( \hat{T}_\zeta (r) + \hat{T}_\zeta^\dagger (r) \right) + \frac{i}{2} \hat{\mu}_y \otimes \left( \hat{T}_\zeta (r) - \hat{T}_\zeta^\dagger (r) \right),
$$

where $\hat{\sigma}_\zeta = (\hat{\sigma}_x, \hat{\sigma}_y)$. The bands shown in Fig. 5(a) (left panel) are deduced from this Hamiltonian.

If we take now $w_{AA'} = 0$, the band dispersion of the lowest-energy bands do not change much, but as shown in the calculation of Fig. 5(b) (left panel), the spectrum becomes degenerate in valley over the whole moiré Brillouin zone. This is due to a new *pseudo-inversion* symmetry that relates the Hamiltonians of the two valley sectors. Specifically, if $w_{AA'} = 0$, then the Hamiltonian in Eq. (67) satisfies

$$
\hat{U} \hat{\mathcal{H}}_\zeta (r) \hat{U}^\dagger = \hat{\mathcal{H}}_{-\zeta} (r),
$$

with \( \hat{U} = \hat{\mu}_y \otimes \hat{\sigma}_y \). \hspace{1cm} (68a)

When the symmetry is enforced, the chiral response is suppressed. This result follows from the following property of the velocity operators:

$$
\hat{U} \hat{\partial}_{\zeta,b} \hat{U}^\dagger = \hat{\partial}_{-\zeta,b},
$$

which can be used, along with $C_2$ symmetries, to prove that the contributions to $\chi^{xy}_{b} (\omega)$ from the two valleys cancel each other.

**Type II**

In the case of type-II structures, the Hamiltonian can be simplified to

$$
\hat{\mathcal{H}}_\zeta (r) = \frac{i \sqrt{3} \mathcal{A} t}{2} \hat{\sigma}_\zeta \cdot \partial + \frac{\Delta (r)}{2} \hat{\mu}_z \otimes \hat{\sigma}_z + \frac{1}{2} \hat{\mu}_x \otimes \left( \hat{T}_\zeta (r) + \hat{T}_\zeta^\dagger (r) \right) + \frac{i}{2} \hat{\mu}_y \otimes \left( \hat{T}_\zeta (r) - \hat{T}_\zeta^\dagger (r) \right),
$$

The bands in Fig. 5(a) (right panel) are deduced from this Hamiltonian.

If we take now $w_{AB} = 0$, just as in the previous case the dispersion of the lowest-energy bands does not change significantly, but the spectrum becomes also valley degenerate, see Fig. 5(b) (right panel). Again, this is due to a new *pseudo-inversion* symmetry. Specifically, if $w_{AB} = 0$, then the Hamiltonian in Eq. (70) satisfies

$$
\hat{U} \hat{\mathcal{H}}_\zeta (r) \hat{U}^\dagger = \hat{\mathcal{H}}_{-\zeta} (-r),
$$

with \( \hat{U} = \hat{\mu}_x \otimes \hat{\sigma}_z \). \hspace{1cm} (71a)
Note that, contrary to the previous case, the symmetry is nonlocal in space but imposes relations between the eigenvalues and eigenstates of the same valley. In particular, it implies that $\varepsilon_{n,\zeta}^n = \varepsilon_{\zeta,-\zeta}^n$. This, along with time-reversal symmetry, implies that $\varepsilon_{\zeta}^n = \varepsilon_{-\zeta}^n$.

When the symmetry is enforced, the chiral response is also suppressed. This result follows from the following property of the velocity operators:

$$\hat{\mathbf{u}} \hat{v}_{\zeta,t} \hat{\mathbf{v}}^\dagger = -\hat{v}_{\zeta,b},$$

which can be used to prove that the contributions to $\chi_{x,y}^{x,y}(\omega)$ from opposite $q$ points within the same valley cancel to each other.

The relation in Eq. (72) can also be used to demonstrate that some of the optical transitions at $\gamma$ and $m$ points (which remain invariant under the symmetry operation) are forbidden when the symmetry is enforced. In particular, since the lowest conduction and valence band states have the same parity under this symmetry, namely,

$$\hat{\mathbf{u}} \left| u_{n,\zeta}(q) \right\rangle = -\left| u_{n,\zeta}(q) \right\rangle \quad \text{for} \quad q = \gamma, m, \quad \text{and} \quad n = c, v,$$

it can be easily seen that $\mathbf{v}_{\zeta}^{(c,v)} = 0$ for $q = \gamma, m$. This gives rise to a partial suppression of the total conductivity of type-II structures (more evident as the twist angle decreases), which contribute to enhance the circular dichroism with respect to type-I structures.