Floquet flat-band engineering of twisted bilayer graphene

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Twisted Bilayer Graphene at the magic twist angle features flat energy bands, which lead to superconductivity and strong correlation physics. These unique properties are typically limited to an extremely narrow range of twist angles around the magic angle. Here we demonstrate that coherent optical irradiation could lead to emergence of flat isolated Floquet-Bloch bands at a wide range of small twist angles. We discuss the conditions under which these bands exhibit a non-zero Chern number. We show that the effect can be realized with relatively weak optical beams at the visible-infrared range, potentially allowing for realization of optically tuned flat bands.

Van der Waals heterostructures have become a prominent tool for discovery of new emergent phenomena in condensed-matter physics. These materials allow for a considerable degree of control in their physical structure, being formed by stacking of individual atomic layers [1–4]. Stacking different atomic layers with a relative angular twist has become a salient mechanism in structuring the energy bands of these materials [1–4]. This twist forms a slowly varying moiré pattern, which modulates the inter layer electronic potential. At certain twist angles, the bands near charge-neutrality point (CNP) can become flat and relatively isolated from other bands [8–10]. These flat bands have recently attracted considerable attention, with the discovery of superconductivity, correlated insulating states and ferromagnetism, which emerge at low temperatures [11,12].

Twisted bilayer graphene (TBG) exhibits isolated flat bands when twisted near the magic twist angle, \( \theta_m \approx 1.1^\circ \). At smaller twist angles \( \theta < \theta_m \), a larger moiré pattern is formed, eliminating the energy gap to distant energy levels and increasing the bandwidth of the bands near CNP [8,10]. Therefore, the discovery of strong correlated phenomena has been limited to a small range of twist angles near the magic angle, where the bandstructure features narrow gapped bands.

Floquet-engineering with optical fields is a valuable technique that could induce topological band structures and electronic correlations in various materials [17,31]. Floquet engineering of TBG at large twist angles has been considered as a technique to control the topology of the bands near CNP [32]. In this work, we focus on Floquet engineering at small angle TBG \( \theta \leq \theta_m \).

We consider small angle twisted bilayer graphene driven with optical fields, as shown in Fig. 1. We demonstrate that a driving laser could improve the flatness of the bands near the CNP and reduce their bandwidth, even at twist angles smaller than the magic angle. We further show that the driving field opens a gap between the emerging flat bands, and increases the gaps separating them from other bands. We also obtain conditions under which the drive induces a nontrivial Berry curvature in the flat bands. The presented effect is found robust for lattice relaxation and could potentially be implemented with relatively weak optical fields in the visible range.

We model the low energy band structure of TBG using a continuum model for a single valley and spin [8,10,34–36]. These models, accurately describe the Hamiltonian of TBG with a relatively small twist angle (\( \theta \lesssim 10^\circ \)) where intervalley processes are strongly suppressed. In the absence of a driving field, our model Hamiltonian is given by

\[
H = \begin{pmatrix}
\hbar(\theta/2, \mathbf{r}) & T(\mathbf{r}) \\
T^\dagger(\mathbf{r}) & \hbar(-\theta/2, \mathbf{r})
\end{pmatrix},
\]

which acts on the spinor \( \Psi(\mathbf{r}) = (\psi_{1A}, \psi_{1B}, \psi_{2A}, \psi_{2B})^T \). The subscripts 1,2 denote the top, bottom layer respectively and the A,B subscripts denote the sub-lattice isospin of a monolayer. The Hamiltonians of the two rotated monolayers of graphene are denoted by \( \hbar(\pm\theta/2, \mathbf{r}) \),

![Figure 1. Schematics of Floquet Twisted Bilayer Graphene.](image-url)
and feature a nearest neighbor coupling with a hopping amplitude $\tau$. The operator $T(r)$ denotes the periodic inter-layer moiré potential

$$T(r) = \sum_{n=1}^{3} \left[ w_0 \sigma_0 + w_1 (\sigma_x \cos n\phi + \sigma_y \sin n\phi) \right] e^{i(n\phi - q_n r)}.$$ 

Here we use the standard Bernal stacking for untwisted layers ($\theta = 0$) and $\phi = 2\pi/3$. The set of wavenumbers $q_1 = k_0(0, -1)$, $q_{2,3} = k_0(\pm \sqrt{3}, 1)/2$, represents the relative displacements of the Dirac cones between the layers where $k_0 = 4\pi\theta/(3\sqrt{3}a)$ is determined by the twist angle $\theta$ and $a = 1.42\,\text{Å}$. The $2 \times 2$ Pauli matrices and identity matrix are denoted by $\sigma$ and $\sigma_0$, respectively. We use the notation $w_0$ for the inter-layer coupling between the AA and BB domains and $w_1$ for the AB and BA inter-layer coupling. We note that our model uses the exact band-structure of monolayer graphene (generalizing the approach which uses the $k \cdot p$ approximation) for better modeling of the higher energy levels. In our model we use $\tau = 2.73\,\text{eV}$, $w_1 = 110\,\text{meV}$, and account for the effects of lattice relaxation by approximating $w_0 = 0.8w_1$. We consider a circularly polarized driving light field of frequency $\Omega$, represented by the electric field $\mathcal{E}(t) = \mathcal{E}(\cos(\Omega t)\hat{x} - \sin(\Omega t)\hat{y})$. We take the light field to be at normal incidence and to be uniform over the sample. We model the interaction with the driving field using a Peierls substitution for the intra-layer hopping parameters in the Hamiltonian, $\tau \rightarrow \tau \cdot \exp(-i\mathcal{E}a/\hbar\Omega)$. In the presence of the time-periodic drive, the solution of the Schrödinger equation can be indexed by the quasi-energies $\varepsilon$, which fall within a single “Floquet-Brillouin” zone $-\hbar\Omega/2 \leq \varepsilon < \hbar\Omega/2$, and can be written as

$$|\psi_\nu(t)\rangle = e^{-i\varepsilon_\nu t/\hbar} \sum_{m=-\infty}^{\infty} e^{-i\hbar\Omega t}|\psi^{(m)}_\nu\rangle,$$

where the index $\nu$ carries all other quantum numbers of the state. The set of modes $\sum_m |\psi^{(m)}_\nu\rangle$ are the eigenmodes of the Floquet Hamiltonian which we numerically solve in momentum-space, truncating both the number of Floquet blocks and the infinite representation of the matrix $T(k)$, see Supplementary Information.

Typical Floquet band structures of TBG are shown in Fig. 2(a)-(d), in the presence and absence of a driving field. The band structures are plotted along a contour in the first moiré Brillouin zone (mBz), which is a hexagon with size $k_0$, as shown in Fig. 1(c). For the undriven case of TBG with twist angle $\theta = 0.9^\circ$, the lower and upper bands near $E = 0$ experience level crossing with other bands at the $\Gamma$ point shown in Fig. 2(a). The relatively large bandwidth of the resulting connected group of bands manifests larger kinetic energy of the electrons which hinders the observation of strong correlation effects. Upon driving, an energy gap $\delta$ between the lower and upper bands is opened, as well as an energy gap $\Delta$ isolating these two bands from the rest of the spectrum. These gaps are shown in Fig. 2(b) for a drive with $\hbar\Omega = 1.5\,\text{eV}$ and peak electric field of $\mathcal{E} = 5.6\,\text{MV/cm}$. For $\theta = 0.7^\circ$, the undriven band-structure exhibits larger bandwidth and multiple level crossings, as shown in Fig. 2(c). In the presence of the driving field, the energy gaps $\delta$ and $\Delta$ are opened, and the bandwidth of the lower and upper bands decreases, flattening the bands as shown in Fig. 2(d). Interestingly, here the drive also flattens the next nearest bands to CNP.

To quantify the effect of the drive on the band structure, in Fig. 3(a-c) we plot the gaps and bandwidth of the upper Floquet-band at $\hbar\Omega = 1.5\,\text{eV}$ as a function of the twist angle $\theta$ and the quantity $P = (3r_{ea}\mathcal{E}/(4\hbar\Omega^3))$, which gives the photo-induced gap in monolayer graphene and is proportional to the intensity of the drive at a given frequency. In the absence of a driving field, the energy gap $\Delta$ between the lower and upper bands vanishes due to the symmetries of monolayer graphene, and the gap $\Delta$ isolating these two bands vanishes for $\theta \leq 0.9^\circ$, as shown in Fig. 3(a) and Fig. 3(b). Upon irradiation, for $\theta \leq 0.9^\circ$ the gaps $\Delta$ and $\delta$ obtain nonzero values and increase almost linearly with $P$, thus yielding isolated, narrow bands. In Fig. 3(c) we plot the...
The realistic parameter range of $0 \leq \theta \leq 1^\circ$, below the first magic angle. From Fig. 3(d) and the supplementary information, we present changes in the lattice relaxation parameter to verify the robustness of the presented mechanism to latter regions, yielding and thus effectively reduce the inter-layer coupling in the AB/BA domains at the expense of the AA/BB domains magic angle.

Conclude that light irradiation allows for emergence of the amplitude of the driving field is increased. We thus conclude that the important effects of the drive are robust with respect to variations in the lattice relaxation. Similar results are obtained for other twist angles below the magic angle. We therefore conclude that the important effects of the drive are robust with respect to variations in the lattice relaxation.

The Floquet bandstructure of TBG in the presence of the drive is a result of both on-resonant and off-resonant processes. It is insightful to demonstrate that the mechanism presented here is associated with an off-resonant process, and in particular, to identify the range of driving frequencies in which the hybridization of the resulting flat bands with high energy bands in the undriven bandstructure is suppressed. To this effect, we define the time-averaged density of states (DOS) defined as

$$\bar{\rho}_0(k) = \sum_{\nu} \sum_{m} A_{\nu}^{(m)}(k) \delta(\epsilon_{\nu} - m \hbar \Omega - E),$$

with $A_{\nu}^{(m)}(k) = |\psi_{\nu}^{(m)}(k)|^2$. The DOS in Eq. (3) is the imaginary part of the time-averaged Green’s function.

To quantify the sharpness of $\bar{\rho}_0(k, E)$, we consider the total intensity computed from the DOS in an interval $\Delta E = 40$ meV around $E = 0$, yielding

$$I(k) = \int_{-\Delta E/2}^{\Delta E/2} dE \bar{\rho}_0(k, E).$$

We then consider this quantity as a sum of contribution coming from different Floquet states, $I(k) = \sum_{\nu} I_{\nu}(k)$ and plot the histogram characterizing the distribution of $I_{\nu}(k)$ averaged along the contour shown in Fig. 1(c). A comparison of the histograms of $I_{\nu}(k)$ for different driving frequencies at $\theta = 0.9^\circ$ is shown in Fig. 1(a)-(c). To maintain $\Delta(\Omega)$ and $\delta(\Omega)$ constant throughout the three panels, we keep the photo-induced gap constant, with $P = 33$ meV.

For drive frequencies in the UV-visible range, the dis-
distribution of \( I_\nu \) has a sharp peak at \( I = 1 \), and the two bands correspond to the upper and lower flat-bands. This is shown in Fig. 4(a), plotted for a driving field oscillating at \( h\Omega = 3 \text{ eV} \) (blue light). For a near-infrared driving field with \( h\Omega = 0.94 \text{ eV} \), the number distribution of \( I_\nu \) is broad, with many Floquet eigenstates corresponding to \( I_\nu < 1 \), as shown in Fig. 4(b). This is a result of the drive inducing strong mixtures of states near \( E = 0 \) with high energy states at \( E + n\hbar\Omega \). This mixing leads to a reduction of the spectral weight corresponding to the flat bands, along with a background spectral weight and band crossings. These effects are also directly seen in a plot of \( \tilde{\rho}_0(\mathbf{k}, E) \) at \( h\Omega = 0.94 \text{ eV} \) shown in Fig. 4(d), which can be compared with the sharply defined bands in the DOS obtained at \( h\Omega = 1.5 \text{ eV} \), see Fig. 2. At even lower driving frequencies, \( I_\nu \) approaches a Poisson distribution, implying that the density of states is dominated by rapidly oscillating Floquet-bands with low spectral weight at the \( |E| \leq \Delta E/2 \) spectral window, as can be seen for \( h\Omega = 0.3 \text{ eV} \) in Fig. 4(c).

To estimate the range of frequencies for which the DOS at energies \( |E| \leq \Delta E/2 \) is predominately sharp, we consider the quantity \( S_n = \frac{\phi(\sum_{\nu} |I_\nu(\mathbf{k})|^2) / \sum_{\nu} |I_\nu(\mathbf{k})|d|k| \text{ along the contour in Fig. 4(c).} \) For \( n \geq 2 \), \( S_n \) approaches unity for a fully sharp DOS, i.e., when the distributions \( I_\nu(\mathbf{k}) \) are bi-modal and peaked at \( I = 0 \) or \( I = 1 \) at all \( \mathbf{k}'s \). Conversely, \( S_n \) with \( n \geq 2 \) becomes vanishingly small when \( I_\nu(\mathbf{k}) \) takes the Poisson form peaked at \( I = 0 \), as in Fig. 4(c). In Fig. 4(e) we plot \( S_4(\Omega) \), finding that the DOS remains sharp for driving frequencies \( \Omega \gtrsim 1.2 \text{ eV} \), the sharpness parameter \( S_4 \) approaches unity, indicating that the interaction with the drive is predominately off-resonant.

To gain an intuitive understanding of the threshold frequency \( \Omega^* \), we consider the interaction of TBG with the drive in the limit of weak interlayer coupling, \( \alpha = (w_0 + w_1)/(3\hbar k_0) \ll 1 \). For a uniform drive field and absence of interlayer coupling, the interaction with light is resonant only between Bloch states at a given momentum whose energy difference matches the light frequency [see Fig. 4(b)]. Since states near \( E = 0 \) and states near \( E = n\hbar\Omega \), for integer \( n \), correspond to different momenta, for \( \alpha = 0 \) the matrix elements of the drive between such states vanishes. For nonzero interlayer coupling, the Bloch wavefunctions are a superposition with nonzero amplitudes of many momentum states (in the extended mBz). As a result, non-zero matrix elements of the drive are obtained between large-momentum components of the states near \( E = 0 \) and the states near \( E = n\hbar\Omega \). We now estimate the amplitudes of these large-momentum components of the Bloch wavefunctions near \( E = 0 \).

Absent the drive, the wavefunctions of the TBG Hamiltonian in Eq. (11) are given by \( |\psi_\nu(\mathbf{k})\rangle = \sum_{l,m,n,p} c_{\nu,lmnp} |\psi_l(\mathbf{k}+\mathbf{k}_{mnp})\rangle \) where \( \mathbf{k} \) is in the first mBz,
the index \( l \) denotes the sublattice of the top and bottom layer (1A,1B,2A,2B) and the indices \( m,n,p \) correspond to \( k_{mnp} = mg_1 + ng_2 + pg_3 \), which denotes the hexagon reciprocal grid in the extended zone. This grid is defined by the \( \text{Moiré} \) reciprocal lattice vectors \( g_1 \) and \( g_2 \), the integers \( m,n \), and the basis index \( p = \pm 1 \) (see SI). We define the hopping number of each spinor component with the index \( r = [m] + [n] + [m + n - p] \), which indicates how many applications of the interlayer coupling are needed in order to mix states with momenta \( k \) and \( k_{mnp} \). For states near \( E = 0 \), the coefficients \( c_{mnp} \) decrease exponentially as \( |c_{mnp}| \leq \alpha^r \) for \( r \geq 1 \). We approximate the characteristic momenta \( q_{mnp} \) in which the interaction with light is on-resonance by the points satisfying \( r(m,n,p) \approx h\Omega/(3tk_0) \).

At large driving frequencies, we can estimate \( S_4 \approx \int |I_0(k)|^2 d|k| \) where \( I_0(k) \) corresponds to Floquet states that can be traced back to Bloch states with momentum \( k \) and \( |E| < \Delta E \) in the absence of the drive. At the same time, the contributions to \( I_0(k) \) arise from momentum components different from the characteristic momenta \( q_{mnp} \). Therefore we can roughly estimate \( I_0 \approx \langle 1 + 3\alpha^2 \rangle^{-1} \), where the factor \( 3\alpha \) arises due to the number of terms with hopping number \( r \) in the hexagonal grid. In practice for \( \theta = 0.9^\circ \) we get \( \alpha = 0.64 \), and the condition \( S_4 \approx |I_0|^2 \geq 0.5 \) yields a threshold frequency of \( h\Omega^* \approx 1.4 \text{eV} \), which can be compared with the numerically calculated value \( h\Omega^* = 1.2 \text{eV} \) as shown in Fig. 4(e).

As shown above, for high drive frequencies \( h\Omega \gtrsim 1.2 \text{eV} \), a direct coupling between the bands near CNP and higher levels in the mBZ is suppressed. Thus, the leading order effect of the interaction with the drive can be approximated with the effective static Hamiltonian \( H_{\text{eff}} \approx H + |H_{\text{HI}},H_{\text{I0}}|/\Omega \), describing an off-resonant process which is second order in the drive. The effect of a weak drive \( E \ll 2\hbar\Omega^2/(3\ell) \), can be approximated with an addition of a photo-induced Haldane mass term to the Hamiltonian describing each monolayer. Thus in this limit, the Hamiltonians \( \hbar \pm(k) \) describing a monolayer in reciprocal space near the \( K(+) \) and \( K'(-) \) points acquire an additional term \( \eta P\sigma_z \), where \( \eta = \pm 1 \) respectively. Within this picture, the band structure of the driven system can be described as the result of the interlayer hybridization between gapped Dirac-cones of the two layers.

The Haldane mass term breaks time-reversal symmetry and therefore the bands may exhibit non-zero Chern numbers. In undriven TBG, the lower and upper bands cross and thus a Chern number cannot be defined for each of them separately. Upon driving the system, a nonzero gap \( \delta \) between the lower and upper bands is opened. We calculate the Chern number of the lower and upper bands near \( E = 0 \) resulting from the addition of Haldane mass-term near the \( K \) point, by integrating the Berry curvature of each band in the entire mBZ see Refs. 37, 38 and SI. At angles \( \theta \geq \theta^* \), a nonzero Chern number is obtained in the limit of weak driving, as shown in Fig. 5 for \( \omega_3/\omega_1 = 0.8 \), which gives \( \theta^* \approx 1^\circ \). However, for these angles the gap \( \Delta \) decreases as the drive strength is increased from zero [cf. Fig. 4(a) at \( \theta = 1^\circ \)]. At a critical drive strength the bands cross with the nearest remote bands above and below the lower and upper bands. For larger drive strength than this critical value, the Chern numbers are trivial. The Berry curvature of the upper and lower Floquet bands at intermediate driving frequencies and above \( (\gtrsim 3\text{eV}) \) is nearly identical to those exhibited by the static model used to obtain Fig. 5 see Supplementary Information.

In conclusion we show that driving TBG with circularly polarized visible to near-infrared light can lead to appearance of flat bands in the Floquet spectrum. The effect persists at a wide range of twist angles, giving a mechanism for engineering flat bands without a need of accurate tuning of the relative twist angle between the graphene layers. Thus Floquet engineering of flat bands may play a particular important role for twisted van der Waals heterostructures which typically exhibit long wavelength non-uniformity in the twist angle.

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Supplementary Information

1. Fourier representation of the Hamiltonian

In this section, we describe the Fourier representation of the Floquet Hamiltonian. The set of modes \(|\psi^{(m)}_\nu\rangle\) used in the main text are the eigenmodes of the Floquet hamiltonian satisfying the time independent eigenvalue equation

\[
\sum_{n=-\infty}^{\infty} \mathcal{H}^{mn}|\psi^{(n)}_\nu\rangle = \varepsilon_\nu|\psi^{(m)}_\nu\rangle, \tag{S1}
\]

where \(\mathcal{H}^{mn}\) represents a block of the infinite Floquet Hamiltonian in the extended zone, given by

\[
\mathcal{H}^{mn} = m\hbar\Omega\delta_{mn} + \frac{\Omega}{2\pi} \int_0^{2\pi} dt e^{-i(m-n)\Omega t} H(t). \tag{S2}
\]

We start with the representation of the Hamiltonian of the undriven system.

\[
H^{(1)}_{\perp}(\theta/2, k') = -\tau f(\theta/2, k') \sigma_{1+} + \text{h.c.} \tag{S3}
\]

where \(\sigma_{1+} = |k', 1A\rangle \langle k', 1B|\) is the Pauli matrix using the iso-spin basis, 1 denotes the top layer (while 2 denotes the bottom layer) and \(f(\theta/2, k') = \sum_{i=1}^3 e^{ik'\delta_i}\) is the graphene dispersion relation with \(\delta_1 = (0, a)\) and \(\delta_2, 3 = a(\mp \sqrt{3}, 1)/2\). Here and throughout this paper, primed variable are correspond to the top layer, whereas \(R\) ensures crystal

\[
h_{2}(-\theta/2, k'') = -\tau f(-\theta/2, k'') \sigma_{2+} + \text{h.c.} \tag{S4}
\]

where \(\sigma_{2+} = |k'', 2A\rangle \langle k''', 2B|\) and \(k'' = R_z(-\theta/2)k\) and \(f(-\theta/2, k'') = \sum_{i=1}^3 e^{ik''\delta_i'}\).

The interlayer interaction operator \(T(k)\) can be represented by an infinite matrix using the plane wave expansion, with elements \(T^{\alpha\beta}_{k', p'} \equiv \langle k', 1\alpha|H_{\perp}|p'\rangle \langle 1\beta|\). The superscripts \(\alpha, \beta \in \{A, B\}\) denote the iso-spin of the monolayer and the indices 1, 2 indicate the top, bottom layer respectively. Note that \(k'\) and \(p'\) are defined in the repeated reciprocal zone and measured from the center of the Brillouin zone (and not relative to the Dirac point). The interlayer interaction hamiltonian \(H_{\perp}\) can be represented using a tight binding in real space

\[
H_{\perp} = \sum_{ij} t_{\perp ij} \hat{c}_{1i}^\dagger \hat{c}_{2j} + \text{h.c.}, \tag{S5}
\]

where \(\hat{c}_{1i}\) and \(\hat{c}_{2j}\) are the fermionic annihilation operators of the top and bottom layers respectively, the indices \(i, j\) denote the real space lattice point, and \(t_{\perp ij}\) denote the interlayer hopping parameters. We invoke the two-center approximation, assuming that the hopping parameters depend only on the relative distance between the different lattice points \(t_{\perp ij} = t_{\perp}(|R_i - R_j|)\) where \(R_i\) denotes the position of the \(i\)th site. Representing \(H_{\perp}\) in the reciprocal Fourier space yields the interlayer coupling coefficients \([88, 84]\),

\[
T^{\alpha\beta}_{k', p'} = \sum_{G_1', G_2'} \frac{t_{\perp}(k' + \mathbf{G}_1')}{A_{uc}} e^{iG_1'\delta_1' - G_2'(\delta_3' - a) - R_z(-\theta)\mathbf{G}_2'} \delta_{k' + \mathbf{G}_1', p' + R_z(-\theta)\mathbf{G}_2'}. \tag{S6}
\]

Here we sum over the reciprocal lattice vectors \(G_1'\) and \(G_2'\) of the two layers, \(A_{uc}\) is the unit cell area, \(d\) is the spacing between the two layers. The parameters \(\delta_1'\) and \(\delta_3'\) indicate the relative position of the \(A\) and \(B\) atoms within a unit cell in the top layer, with \(\delta_A = 0\) and \(\delta_B = (0, a)\). The Kronecker delta \(\delta_{k' + \mathbf{G}_1', p' + R_z(-\theta)\mathbf{G}_2'}\) ensures crystal
momentum conservation by the tunneling process. To simplify the expression of $T^{\alpha \beta}_{k',p''}$ further, it is plausible to assume that the interlayer tunneling is slowly varying in space due to the large interlayer separation $d > a$, such that $t_{11}(k)$ falls rapidly to zero at high momenta (cf. [S8, S30]). We can then truncate the sum over all reciprocal lattice vectors to a sum over the three smallest reciprocal lattice vectors (which are of the same magnitude) yielding

$$T^{\alpha \beta}_{k',p''} \approx \sum_{i=1}^{3} \delta_{k' + q_i, p''} T_i^{\alpha \beta}.$$  \hspace{1cm} (S7)

where the momenta $q_i$ are given in the main text. The $2 \times 2$ matrices $T_i$ in the iso-spin basis which are given in the main text include lattice relaxation effects. Eq. (S7) can be considered as a nearest-neighbors hopping Hamiltonian in the reciprocal moiré lattice.

The expansion of $T^{\alpha \beta}_{k',p''}$ couples the wavefunction $|\psi_1(k')\rangle$ in the first layer with the wavefunctions of the other layer $|\psi_2(p'')\rangle$ for $p'' = k' + q_1$, $p'' = k' + q_2$ and $p'' = k' + q_3$. In general, this matrix couples points on a hexagonal lattice in reciprocal space connected by reciprocal lattice vectors of the moiré lattice. To numerically solve this infinite expansion, we pose a further (numerical) truncation by setting a maximal hopping number $r_{\text{max}}$, where the total number of sites considered is $\frac{3}{2}r_{\text{max}}^2 + \frac{3}{2}r_{\text{max}} + 1$. As an example, the Hamiltonian for $r_{\text{max}} = 13$ is given by

$$H^{(1)}_{k}(\theta/2,k') = \begin{pmatrix}
    h_1(\theta/2,k') & T_1 & T_2 \\
    T_1^\dagger & h_2(-\theta/2,k' + q_1) & T_3 \\
    T_2^\dagger & T_3^\dagger \\
\end{pmatrix}. \hspace{1cm} (S8)

A diagrammatic representation of the Hamiltonian in the reciprocal space is shown in Fig. S1 for $r_{\text{max}} = 13$. In this representation, the central node has a momentum $k$. Each node represents a $2 \times 2$ Hamiltonian of monolayer graphene, whereas black nodes represent the matrices $h_1(\theta/2,k' + k_{\text{mnp}})$ while gold-colored node represent $h_2(-\theta/2,k' + k_{\text{mnp}})$. Here $k_{\text{mnp}} = mg_1 + ng_2 + pq_1$, denotes the hexagon reciprocal grid in the extended zone. This grid is defined by the moiré reciprocal lattice vectors $g_1 = q_1 - q_3$, and $g_2 = q_2 - q_1$, the integers $m, n$, and the basis index $p = \pm 1$. A nonzero transition matrix connecting layer 1 to 2 is represented by an edge, where the transition matrix $T_1$ is indicated by a blue edge, a transition matrix $T_2$ is indicated by a red edge and a transition matrix $T_3$ by a green edge. Note that the coupling between layer 2 to 1 is represented in a similar way but with the Hermitian conjugated matrices $T_1^\dagger$, $T_2^\dagger$ and $T_3^\dagger$. The numbers near each point indicate the index of the vector basis in the numerical matrix (not including the iso-spin degree of freedom). It is useful to write the general static Hamiltonian in reciprocal space with

$$H^{(r_{\text{max}})}(k) = T(k) + \sum_{p=0}^{1} \sum_{m,n}(m,n,p)h_{m,n,p}(\theta/2,k)\langle m,n,p |,$$  \hspace{1cm} (S9)

where the sum is over all integers $m, n$ satisfying $r(m,n,p) = |m| + |n| + |m + n - p| \leq r_{\text{max}}$. The hopping number $r$ counts the number of edges connecting that $k_{m,n,p}$ grid point to the momentum $k$. For even values of $r(m,n,p)$ we identify $h_{m,n,p} = h_1(\theta/2,k' + k_{\text{mnp}})$ while for odd values of $r(m,n,p)$ we have $h_{m,n,p} = h_2(-\theta/2,k' + k_{\text{mnp}})$. In general $r_{\text{max}}$ is determined upon convergence of the result of the low energy (we verify that the error is less than 1% in the low energy spectrum of $|E| \leq 50$ meV), and we typically use $13 \leq r_{\text{max}} \leq 22$ in our calculations.

2. Floquet Hamiltonian

We construct the time dependent driven Hamiltonian by applying the Peierls substitution in reciprocal space $\mathbf{k} \rightarrow \mathbf{k} - e\mathbf{A}(t)/\hbar$, where $\mathbf{A} = \hat{E}/\Omega$ is the vector potential. While the interlayer interaction is independent of the normally incident drive field, the monolayer Hamiltonian is transformed via $h_1(\theta/2,k') \rightarrow h_1(\theta/2,k' - e\mathbf{A}'(t)/\hbar)$ for the first layer and $h_2(-\theta/2,k'') \rightarrow h_2(-\theta/2,k'' - e\mathbf{A}''(t)/\hbar)$ for the second layer. We expand the Hamiltonian in a harmonic expansion,

$$h_1(\theta/2,k' - e\mathbf{A}'(t)/\hbar) = -\tau \sum_{n=-\infty}^{\infty} (g_+(\theta/2,k',n)\sigma_{1+} + g_-(\theta/2,k',n)\sigma_{1-}) e^{i\Omega t}.$$  \hspace{1cm} (S10)
Similarly for the second layer we use

\[ g_{\pm}(\theta/2, k', n) = i^n e^{i n(\phi + \theta/2)} \left( e^{\pm ik'\delta_1} J_n\left(\frac{e\alpha}{\hbar} A\right) + e^{\pm ik'\delta_2} e^{-i n\psi} J_n(\pm \Gamma+) + e^{\pm ik'\delta_2} e^{-i n\psi} J_n(\pm \Gamma-) \right), \]  

(S11)

are the coefficients of the \( n \)th harmonic order for the first layer. The parameters \( \Gamma \pm \) and \( \psi \pm \) will be defined below. Similarly for the second layer we use

\[ h_2(-\theta/2, k'', n) = -\tau \sum_{n=-\infty}^{\infty} \left( g_+(-\theta/2, k'', n) \sigma_{2+} + g_-(\theta/2, k'', n) \sigma_{2-} \right) e^{in\Omega t} \]  

(S12)

where the coefficients of the \( n \)th harmonic order of the second layer are given by

\[ g_{\pm}(\theta/2, k'', n) = i^n e^{i n(\phi - \theta/2)} \left( e^{\pm ik''\delta_1} J_n\left(\frac{e\alpha}{\hbar} A\right) + e^{\pm ik''\delta_2} e^{-i n\psi} J_n(\pm \Gamma+) + e^{\pm ik''\delta_2} e^{-i n\psi} J_n(\pm \Gamma-) \right), \]  

(S13)

The above relations are derived using the identities \[ S41 \]

\[ e^{iz \cos \phi} = \sum_{n=-\infty}^{\infty} i^n J_n(z) e^{in\phi}, \]  

(S14)

\[ e^{-i q z} J_q(1) = \sum_{n=-\infty}^{\infty} J_{n+q}(\alpha) J_n(\beta) e^{-i n\phi}, \]  

(S15)

where \( J_n(z) \) is the \( n \)th order Bessel function and \( \phi \) is the phase retardence between \( A_x \) and \( A_y \). The parameters \( \Gamma \) and \( \psi \) in Eqs. [S14] and [S15] can be written by two parameters \( \alpha \) and \( \beta \) as

\[ \Gamma = \sqrt{\alpha^2 + \beta^2 - 2\alpha \beta \cos(\phi)}, \]  

(S16)
\[ \psi = \text{atan} \left( \frac{\beta \sin(\phi)}{\alpha - \beta \cos(\phi)} \right). \]  

(S17)

In our case we find that \( \Gamma_\pm \) and \( \psi_\pm \) in Eqs. (S10)-(S12) are given by

\[ \Gamma_\pm = \frac{\alpha}{2} \sqrt{A_y^2 + 3A_y^2 \pm 2\sqrt{3}A_y \cos(\phi)}, \]  

(S18)

and

\[ \psi_\pm = \text{atan} \left( \frac{\sqrt{3}A_y \sin(\phi)}{\pm A_y + \sqrt{3}A_y \cos(\phi)} \right). \]  

(S19)

For the special case of circularly polarized light considered here we use \( \phi = \pi/2 \) and obtain the simple relations \( \Gamma_\pm = eaA/\hbar \) and \( \psi_\pm = \pm \pi/3 \). We can use the expansion in Eq. (S10), with a truncated sum on integers from \(-N_F \) to \( N_F \), in order to construct the time independent Floquet Hamiltonian \( \mathcal{H}_{mn} \) with \( -N_F \leq m \leq N_F \), yielding

\[ \mathcal{H}_{m,m+n}(k) = \left( m\Omega \sigma_0 + T(k) \right) \delta_{mn} + \frac{1}{2} \sum_{p'=0}^1 \sum_{m',n'} |m',n',p'| \tilde{h}_{m',n',p'}(\theta/2, k, n)(m',n',p'|. \]  

(S20)

where for even values of \( r(m',n',p') \) we define

\[ \tilde{h}_{m',n',p'}(\theta/2, k, n) = -\tau (g_+(\theta/2, k' + k_{m',n',p'}, n)\sigma_{1+} + g_-(\theta/2, k' + k_{m',n',p'}, n)\sigma_{1-}), \]  

(S21)

and for odd values of \( r(m',n',p') \) we define

\[ \tilde{h}_{m',n',p'}(\theta/2, k', n) = -\tau (g_+(\theta/2, k' + k_{m',n',p'}, n)\sigma_{2+} + g_-(\theta/2, k' + k_{m',n',p'}, n)\sigma_{2-}). \]  

(S22)

In practice, we observe convergence of numerical calculations when setting a cutoff of \( N_F = 2 \) or 3. The dimension of the Floquet matrix is then \( 2(N_F + 1) \cdot (3r_{\max}^2 + 3r_{\max} + 2) \).

The functions \( g_\pm(\pm \theta/2, k, n) \) obtains a simple form in the low power regime \( eaA/\hbar \). In this regime the zeroth order describes the undriven Hamiltonian with \( g_+(\pm \theta/2, k, 0) = f(\pm \theta/2, k) \) and \( g_-(\pm \theta/2, k, 0) = f^*(\pm \theta/2, k) \). Furthermore near the K point in the top layer we find that \( g_+(\theta/2, k, -1) = 0 \) and \( g_+(\theta/2, k, 1) \approx 3eaAe^{-\theta/2}/(2\hbar) \), and near the K Dirac point of the bottom layer we have \( g_+(\theta/2, k, -1) = 0 \) and \( g_+(\theta/2, k, 1) \approx 3eaAe^{-\theta/2}/(2\hbar) \). We also find that the contribution of terms with higher orders of \( n \) are smaller by powers of \( (eaA/\hbar)^n \). Importantly, for \( \theta \ll 1 \) we find that the effective Hamiltonian in leading powers of the small parameter \( [(3reaA)/(2\hbar^2\Omega)] \) at these conditions is given by

\[ H_{\text{eff}} \approx H + [\mathcal{H}_{01}, \mathcal{H}_{10}] / \Omega \]  

(S23)

2. Lattice relaxation

In Fig. S2 we calculate the dependence of the bandwidth and the gaps \( \Delta, \delta \) on the parameter \( w_0/w_1 \) for \( \theta = 0.7^\circ, 0.9^\circ \) using \( \hbar\Omega = 1.5 \text{eV} \) and \( \mathcal{E} = 4 \text{MV/cm} \) (\( P = 33 \text{meV} \)). These results demonstrate that the effect of the driving weakly depends on the exact lattice relaxation parameter.

3. Calculation of the Chern number

We calculate the Chern number of the lower and upper bands of TBG following the procedure presented in Refs. S37, S38. We discretize the reciprocal space using a rectangular grid \( k = k_0(m\hat{x} + n\hat{y})/N_k \) for integer \( -N_k \leq m, n \leq N_k \) and typically use \( N_k = 200 \). We consider only the grid points within the first mBz hexagon. Our goal is to diagonalize the static Hamiltonian in Eq. (1) including a Haldane photo-induced mass term of size \( P \).

Recall that the static Hamiltonian Eq. (1) is valley and spin degenerate. The two valley degenerate bands arise from Bloch wavefunctions which are superpositions of momenta near the \( K \) or \( K' \) points of both layers. Here, we calculate the Bloch wavefunctions in the mBz corresponding to the bands whose momenta \( k \) are always near the \( K \) points of both layers, and far from \( K' \) points of both layers, since \( r_{\max} k_\theta \ll |\mathbf{K} - K'| \). Therefore, to take the Haldane
Figure S2. The increased gaps $\Delta, \delta$ and low bandwidth of driven TBG are robust with variations of lattice relaxation effects. Calculation for $\theta = 0.7^\circ$ (left) and $\theta = 0.9^\circ$ (right) using $\hbar \Omega = 1.5 \text{eV}$ and $E = 4 \text{MV/cm}$.

Figure S3. Berry curvature of the upper band in the mBz for $\theta = 1.2^\circ$ and $P = 10 \text{meV}$. The effective static Hamiltonian and the full Floquet analysis yield the same result.

mass term into account, it is sufficient to add to the Hamiltonian $h(k)$ appearing in Eq. (S9) a mass term $P\sigma_z$, which represents the Haldane mass term correctly near the $K$ point.

We diagonalize the Hamiltonian in Eq. (1) including the term $P\sigma_z \otimes \sigma$, and find the wavefunctions $|\psi_l(k)\rangle$ of the lower and upper bands denoted with $l \in \{v, c\}$. We then calculate the local Berry curvature with

$$B_l(k) = \left( \frac{N_k}{k_{\theta}} \right)^2 \arg \left( U_{l,x}(k)U_{l,y}(k + \hat{x}k_{\theta}/N_k)U_{l,x}^*(k)U_{l,y}^*(k) \right),$$

(S24)

where the potentials $U_{l,x}(k)$ and $U_{l,y}(k)$ are given by

$$U_{l,x}^{v,c}(k) = \langle \psi_{v,c}(k + \hat{x}k_{\theta}/N_k)|\psi_{v,c}(k)\rangle,$$

(S25)

and

$$U_{l,y}^{v,c}(k) = \langle \psi_{v,c}(k + \hat{y}k_{\theta}/N_k)|\psi_{v,c}(k)\rangle.$$

(S26)

We calculate the Chern number of the lower and upper bands in Fig. 5 by summing the local Berry curvature over the mBz

$$C_l = \frac{1}{2\pi} \left( \frac{k_{\theta}}{N_k} \right)^2 \sum_k B_l(k).$$

(S27)
An example of the Berry curvature of the upper band of TBG with $\theta = 1.2^\circ$ driven by $P = 10$ meV is shown in Fig. S3 for the static Hamiltonian.

At relatively high drive frequencies, as shown in the main text, the Floquet bands closely resemble the bands of the static Hamiltonian, as mixing and level crossings are suppressed. Therefore we expect that the Berry curvature of the Floquet bands should closely resemble the Berry curvature of the static calculation with the Haldane mass term, up to isolated level crossings which effect the Berry curvature in very narrow regions in the reciprocal space. To demonstrate this we calculate the Berry curvature of the Floquet bands with the full time dependent Hamiltonian with $\hbar \Omega = 3$ eV , in Fig. S3(b). To perform this calculation we chose the wavefunctions of the upper and lower bands as the two wavefunctions whose spectral weight $A_\nu(k)$ in the energy interval $|E| < \Delta E$ is maximal, c.f. Eq. (3) (at high drive frequencies, these bands have $A_\nu(k) \approx 1$). This example demonstrates that indeed the Berry curvature of the static Hamiltonian captures the one of the Floquet bands at relatively high drive frequencies.