We investigate the topological properties of Floquet-engineered twisted bilayer graphene above the so-called magic angle driven by circularly polarized laser pulses. Employing a full Moiré-unit-cell tight-binding Hamiltonian based on first-principles electronic structure, we show that the band topology in the bilayer, at twisting angles above 1.05°, essentially corresponds to the one of single-layer graphene. However, the ability to open topologically trivial gaps in this system by a bias voltage between the layers enables the full topological phase diagram to be explored, which is not possible in single-layer graphene. Circularly polarized light induces a transition to a topologically nontrivial Floquet band structure with the Berry curvature analogous to a Chern insulator. Importantly, the twisting allows for tuning electronic energy scales, which implies that the electronic bandwidth can be tailored to match realistic driving frequencies in the ultraviolet or midinfrared photon-energy regimes. This implies that Moiré superlattices are an ideal playground for combining twistronics, Floquet engineering, and strongly interacting regimes out of thermal equilibrium.

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I. INTRODUCTION

Light-matter coupled systems are emerging as an important research frontier bridging condensed matter physics [1], quantum optics [2–6], and cold atoms in optical lattices [7–9]. In ultrafast materials science, intriguing phenomena have been explored, including but not limited to ultrafast switching between different phases of matter [10–12], light control of important couplings in solids [13–15], and light-induced superconductivity [16,17]. In cavities, spectacular effects have been observed or predicted, such as dramatically enhanced conductivity in polymers [18], cavity-modified materials properties [19–22], novel spectroscopies using the quantum nature of light [5], and light-controlled chemical reaction pathways [23]. Finally, in optical lattices, periodically driven quantum systems are investigated within the realm of Floquet engineering, in which the driving is used as a tool to generate effective Hamiltonians with tunable interactions [24–28], which has also been demonstrated in purely photonic systems [29].

In solids, key progress has been made, for instance, in observing Floquet-Bloch states in time-resolved photoemission spectroscopy [30] or with the recently demonstrated light-induced anomalous Hall effect in graphene under circularly polarized laser driving [31–33]. However, the concept of Floquet engineering of a material’s topological properties [34–43] has been limited to very few material systems so far. The reason for this is that the time and energy scales often cause problems in avoiding resonant excitations and accompanying heating effects that are usually detrimental for the observation of interesting light-induced states of matter, unless full thermalization is delayed [44,45] or unless the laser is only used as a means to break the symmetry for transient intermediate states, such as in the case of laser-controlled superconductors [12]. It is therefore an important task to identify material platforms in which crucial energy scales, such as the relevant effective electronic bandwidth, can be tuned with respect to the photon energy of the pump laser. This is exactly the case in twisted van der Waals heterostructures forming Moiré superlattices [46]. In particular, twisted bilayer graphene (TBG) has recently attracted considerable attention due to the discovery of superconductivity [47] along with correlated insulating phases in its vicinity [48]. In TBG, the Fermi velocity and electronic bandwidth of the Dirac bands can be tuned by changing the twisting angle, leading to flat bands at particular magic angles and opening the possibility of twistronics [49] and tunable energy absorption spectra [50]. In addition, this paves the way to tune the role of interactions in a materials setting [51], similar to the case of artificial lattices, in which more exotic Floquet Hamiltonians have been generated [52], and recently also a cold-atom realization of twisted bilayers has been proposed [53]. In the context of TBG, this might, for instance, open the possibility to study strongly correlated Floquet-engineered phases of matter such as the proposed fractional Floquet-Chern insulator [54].

In this work, we perform a full tight-binding model calculation for TBG for an intermediate rotation angle, θ = 7.34°, with a unit cell containing 244 sites, in order to investigate its electronic and topological properties in and out of equilibrium above the magic angle regime in a microscopic picture. This choice of the twist angle is motivated by the fact that the Dirac bandwidth is 2.23 eV, which corresponds to a realistic laser
angles, whose associated bandwidths lie within the midinfrared photon energy range. Thus, we expect a time-resolved Hall current measurement, in the electronic band energies but also for the system topology. Furthermore, we explicitly show that this is not only true for circularly polarized driving, we find a topologically nontrivial phase, which is tracked by the Berry curvature of Floquet time-reversal symmetry and induces a topologically nontrivial to single-layer graphene, to study the transition between the layers. This offers the unique opportunity, in contrast to single-layer graphene, to study the transition between the topologically trivial and nontrivial phases, as originally envisioned by Haldane in his seminal work on the quantum anomalous Hall effect [55]. Notably, this is unlike the case of time-reversal symmetry breaking by a magnetic field, where the large magnetic unit cell and corresponding small Brillouin zone cause dramatically different effects such as the emergence of a Hofstadter butterfly [56]. Our results confirm that for angles larger than the highest magic angle (134.05°) the interlayer interactions can be captured by perturbative treatment, which maintains the linear band dispersions while renormalizing the Fermi velocities, as stated in Refs. [57,58]. Furthermore, we explicitly show that this is not only true for the electronic band energies but also for the system topology. Thus, we expect a time-resolved Hall current measurement, in the time-reversal symmetry-broken state, to show a nonzero Hall conductance [34,55] approaching a quantized value of 4e²/h for completely filled valence bands and empty conduction bands, where the factor of 4 reflects two valence bands, two spin species, and two valleys.

The paper is organized as follows. In Sec. I, we explain the basic properties of our tight-binding model. In Sec. II, we investigate the electronic properties of our starting point, particularly the equilibrium topology of the system. Section III outlines the Floquet electronic band structure and its key differences, compared to equilibrium. In Sec. IV, we present our main result of the Floquet-engineered Berry curvature and explore the Floquet-topological phase space in dependence of driving amplitudes and different choices of local potentials. We finish with a brief conclusion in Sec. V.

II. TIGHT-BINDING MODEL

Our starting point are two A-A-stacked graphene layers [59]. Choosing the origin of the Cartesian coordinate system at an atomic site, we construct the bilayer by a rotation of the top layer by a twist angle, θ = 7.34°. This rotation amounts to a reflection of the bottom layer within the x-y plane along the supercell diagonal, as depicted in Fig. 1(a). Using the commensurability condition derived in Ref. [57], this results in a supercell of 244 carbon atoms, which has regions of A-A and A-B stacking, respectively. We use a general tight-binding hopping Hamiltonian of the form

\[ H = \sum_i \epsilon_i c_i^\dagger c_i + \sum_{i \neq j} t_{ij} c_i^\dagger c_j, \]  

(1)
where $c_i^\dagger$ ($c_i$) creates (annihilates) an electron in the $p_z$ orbital at the atomic position $r_i$. The variables $\epsilon_i$ and $t_{ij}$ denote the local potential and hopping matrix elements, respectively. The model parameters are given in Appendix A.

The equilibrium band structure along the symmetry path $\Gamma$-$K_1$-$M$ in the mini-Brillouin zone (mBZ) of the Moiré superlattice is presented in Fig. 1(b). It shows a linear Dirac-cone dispersion around one Dirac point $K_1$ with a renormalized Fermi velocity, $v_F/v_F^0 = 0.9$, compared to the one in single-layer graphene (dashed lines). The renormalized $\Gamma$-point bandwidth $\Delta_\Gamma$ and $M$-point energy difference $\Delta_M$ for the bottom conduction and top valence bands are indicated by colored arrows.

Envisioning periodic laser driving at specific photon energies with the goal to Floquet engineer a light-induced anomalous Hall effect [31,32], we now address the relevant electronic energy scales as a function of twisting angle. To this end, we show in Fig. 1(c) both $\Delta_\Gamma$ and $\Delta_M$ for different commensurate rotation angles above the magic-angle regime. As discussed in various references, such as Ref. [46], the low-energy Dirac bands can be rescaled to exhibit a desired bandwidth by tuning to particular twist angles. In the inset of Fig. 1(c), we focus on the range of angles for which the $\Delta$ values lie in the range attainable in an existing ultrafast transport setup [31]. This opens up the possibility of a Floquet-engineered band structure with a light-induced anomalous Hall effect if the laser breaks time-reversal symmetry (see below) with electronic energy scales tunable near the photon energy. This might, for instance, be possible for a twist angle of $1.7^\circ$ and photon energy of 200 meV, which matches roughly the low-energy bandwidth in this case.

III. EQUILIBRIUM TOPOLOGY

Before turning to Floquet-engineered bands and topology, we first discuss the equilibrium Berry curvature near the Dirac points when different onsite potentials are applied to TBG. This can be seen as a direct generalization of the Haldane model for single-layer graphene [55] with broken inversion symmetry and intact time-reversal symmetry.

For the onsite potentials, we choose from three different options, detailed in Appendix A: (i) a back-gate voltage which corresponds to an energetic difference $V_{BG}$ between the top and bottom layers; (ii) an asymmetric $A$-$B$ potential with energetic difference $V_{AB}^{\text{sym}}$ between $A$ and $B$ sublattice in one layer and $-V_{AB}^{\text{sym}}$ in the other layer; and (iii) a symmetric $A$-$B$ potential with energetic difference $V_{AB}^{\text{sym}}$ between $A$ and $B$ sublattice in both layers. As we will see, different combinations of these potentials (i) and (ii) and (i) and (iii), respectively, lead to different band structures and different Berry curvatures within the bands. We notice that for the real TBG device, $V_{AB}^{\text{sym}} = 0$ and $V_{AB}^{\text{sym}} = 0$. These onsite potentials should be seen as fictitious, primarily unphysical potentials that illustrate how the topology changes for different ways in which inversion symmetry can be broken, similar in spirit to the Haldane model [55]. Notice, though, that encapsulation of TBG between hexagonal boron nitride layers might lead to an induced $A$-$B$ potential even in TBG. Moreover, other hexagonal two-dimensional materials, such as transition-metal dichalcogenides, have an effective $A$-$B$ potential. Finally, the cold-atom realization of twisted bilayers paves the way to implement such potentials in artificial TBG [53]. On the other hand, the back-gate potential can be readily applied experimentally, as has been demonstrated for conventional $A$-$B$-stacked bilayer graphene [60,61]. Unlike the case of $A$-$B$-stacked bilayer graphene, which has a parabolic energy dispersion, TBG does have a Dirac-like dispersion, which offers the unique opportunity to experimentally study the competition between broken time-reversal and broken inversion symmetries.

Figure 2 shows the evolution of band structures and Berry curvatures (see Appendix C) near both Dirac points $K_1$ and $K_2$ in the mBZ for twist angle $\theta = 7.34^\circ$. First, we focus on infinitesimally small $V_{AB}^{\text{sym}} = V_{BG} = 0.0001$ eV [Figs. 2(a) and 2(b)]. Both the valence and conduction bands are still almost twofold degenerate and a small energy gap of order 0.0001 eV opens at $K_1$ and $K_2$, inducing a nonvanishing Berry curvature. Figure 2(b) shows the Berry curvature in the two valence bands. For one valence band (green solid line), the Berry curvature is negative at $K_1$ and positive at $K_2$, while it is exactly the opposite for the other valence band (purple dashed line). Thus, the Berry curvature integrated within each valley ($K_1$ and $K_2$) vanishes. The same qualitative behavior is observed for increased values of the asymmetric potential $V_{AB}^{\text{sym}} = 0.01$ eV larger than the back-gate potential $V_{BG} = 0.001$ eV [Figs. 2(e) and 2(f)] as well as asymmetric potential $V_{AB}^{\text{sym}} = 0.01$ eV smaller than the back-gate potential $V_{BG} = 0.05$ eV [Figs. 2(i) and 2(j)]. In these cases, the Berry curvature is spread out a bit more in momentum space. In the latter case with larger back-gate potential, one can observe additional band crossings to the left and right of the Dirac points [Fig. 2(i)] with vanishing Berry curvature [Fig. 2(j)].

We now study what happens for the case of a symmetric $A$-$B$ potential $V_{AB}^{\text{sym}}$. For infinitesimally small potentials $V_{AB} = V_{BG} = 0.0001$ eV [Figs. 2(c) and 2(d)], both valence bands show the same Berry curvature being positive near $K_1$ and negative near $K_2$. Thus, in the symmetric case, as opposed to the asymmetric case, there is a nonvanishing total valley Berry curvature but vanishing overall Berry curvature due to cancellation between both valleys in the mBZ. The same qualitative behavior is observed for increased values of the symmetric potential $V_{AB}^{\text{sym}} = 0.01$ eV larger than the back-gate potential $V_{BG} = 0.001$ eV [Figs. 2(g) and 2(h)]. This behavior is switched when the back-gate potential $V_{BG} = 0.05$ eV is larger than the symmetric potential $V_{AB}^{\text{sym}} = 0.01$ eV [Figs. 2(k) and 2(l)]. In this case, the Berry curvature is analogous to the one for the asymmetric case with vanishing valley Berry curvature for each valley individually. This finding is particularly interesting as it opens up the possibility of starting from a topologically trivial insulating phase for $V_{BG} \neq 0$ and $V_{AB} = 0$, which is the realistic scenario for TBG. Below, we will show that by applying circularly polarized light, one can light induce the transition from topologically trivial to topologically nontrivial insulating band structures at a nonvanishing critical field strength, which is not possible for single-layer graphene.

Overall, we note that TBG obviously offers more possibilities for combinations of Berry curvature than single-layer graphene. In particular, the Berry curvature can vanish per valley when summed over all valence bands, while being nonzero...
for each band separately within a valley, for broken inversion
symmetry due to an asymmetric A-B potential or due to a
symmetric A-B potential when the back-gate voltage exceeds
this A-B potential. This scenario obviously does not exist for
single-layer graphene, which has only one valence band (per
spin, which is suppressed here) in the Dirac bands. In all cases
discussed here, the total Berry curvature and thus the valence
band Chern number vanishes when time-reversal symmetry is
intact, like in single-layer graphene or the Haldane model with
time-reversal symmetry. Importantly, we do not make a claim
about the topology of the regime at and below the highest
magic angle around $1.05^\circ$, for which there have been several
predictions for the effective Berryology [62–65].

IV. FLOQUET BAND STRUCTURE

We now turn to the case of laser-driven TBG with circularly
polarized light in which Floquet bands are formed. Focusing
again on a twist angle $\Theta = 7.34^\circ$, used here and in all that
follows, we choose a photon frequency $\Omega = 2.23$ eV tuned to
the bandwidth $\Delta \Gamma$ for this particular twist angle. This choice
is motivated by the fact that Floquet sidebands overlapping
with the original bands are not created in this case within the
low-energy Dirac bands but only with higher lying bands. The
laser field is described by a time-dependent, spatially homoge-
nous vector potential $A(t) = A_{\text{max}}[e_x \sin(\Omega t) + e_y \cos(\Omega t)]$,
and is coupled to the electrons in TBG via Peierls substitution,

$$t_{ij} \rightarrow \tilde{t}_{ij}(t) = t_{ij} e^{A_{\text{max}} B r_{ij}}. \tag{2}$$

From the resulting time-dependent Hamiltonian, we compute
the Floquet bands by expanding in Floquet harmonics [66–68]
and diagonalizing the Floquet Hamiltonian with a cutoff in the
Floquet index and checked convergence in this cutoff. In order
to plot the resulting bands and their topology, we choose to
project the Floquet bands onto the original energy eigenstates
of the bare Hamiltonian, i.e., to compute the square of the
wave function overlap and color the resulting Floquet bands
with a color scale according to this overlap (see Appendix B).

In Fig. 3, we show the original equilibrium bands (dashed lines) and the projected Floquet bands for a relatively strong

![FIG. 2. Equilibrium band structure and corresponding valence-band Berry curvatures in close proximity to the Dirac points of the mBZ
along $\Gamma$-$K_1$-$M$-$K_2$-$\Gamma$ (interval of length 0.022 Å around $K_1$ and $K_2$) for different onsite potentials $V_{\text{AB}}$ and backgate voltages $V_{\text{BG}}$. [(a), (c),
(e)] Antisymmetric $\pm/\mp$ sublattice potential within the layers. [(g), (i), (l)] Symmetric $\pm/\pm$ sublattice potentials. [(b), (f), (j), (l)] Local and
valley band curvature shows opposite sign, respectively. [(d), (h)] Nonvanishing valley Berry curvature but zero integrated Berry curvature
due to sign flip between valleys. We use the following values for the potentials: [(a), (b)] $V_{\text{AB,asym}} = 0.0001$ eV, $V_{\text{BG}} = 0.0001$ eV, [(c), (d)]
$V_{\text{AB,sym}} = 0.0001$ eV, $V_{\text{BG}} = 0.0001$ eV, [(e), (f)] $V_{\text{AB,asym}} = 0.001$ eV, $V_{\text{BG}} = 0.001$ eV, [(g), (h)] $V_{\text{AB}} = 0.001$ eV, $V_{\text{BG}} = 0.001$ eV, [(i), (j)]
$V_{\text{AB,asym}} = 0.01$ eV, $V_{\text{BG}} = 0.05$ eV, and [(k), (l)] $V_{\text{AB}} = 0.01$ eV, $V_{\text{BG}} = 0.05$ eV.

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laser field ($E_{\text{max}} = 13.7$ MV/cm). We choose this large field strength to make the Floquet sidebands and corresponding level crossings clearly visible. We investigate two cases, the one of infinitesimal potentials $V_{\text{sym}}^{AB} = V_{\text{BG}} = 0.0001$ eV [Fig. 3(a), for which the Floquet bands are essentially those of pristine TBG] and the one of vanishing $A$-$B$ potential but nonvanishing back-gate potential $V_{\text{BG}} = 0.05$ eV [Fig. 3(b)]. The circularly polarized driving field opens a sizable energy gap at both Dirac points $K_1$ and $K_2$ and creates a variety of Floquet bands, most of them with relatively small overlap with the equilibrium bands. Nevertheless, a number of avoided band crossings are visible in the chosen energy window, leading to a more complicated structure than for single-layer graphene [40]. Similarly, for the case in Fig. 3(b), the main feature induced by the laser driving is a band gap opening at $K_1$ and $K_2$ but now with visibly split Floquet valence and conduction bands due to the sizable back-gate potential. Interestingly, in both cases the relatively strong field leads to a suppression of band overlap between Floquet bands and bare bands at the top of the conduction bands at the $M$ point, because there are many higher and lower lying bands whose Floquet replicas happen to match the bare bands at this point and hybridize with them. Together these results show that relatively complex Floquet bands can in principle be induced in TBG. Recent combined experimental-theoretical studies of the light-induced anomalous Hall effect in single-layer graphene have demonstrated the importance of avoided band crossings for signatures of Hall currents as a function of chemical potential [31,33], which is why our results presented here serve as a guide to the understanding of potential future transport measurements on laser-driven TBG.

V. FLOQUET-ENGINEERED TOPOLOGY

Finally, the obvious follow-up question to address is the one of the light-induced Berry curvature and corresponding Floquet-band topology. To this end, we focus on the low-energy Floquet-Dirac bands near $K_1$ and $K_2$ and color these bands according to their respective Berry curvatures. The resulting Floquet-engineered Berry curvatures are presented in Fig. 4. Figures 4(a)–4(d) shows the evolution for infinitesimal potentials $V_{\text{sym}}^{AB} = V_{\text{BG}} = 0.0001$ eV as a function of increasing driving field strength $E_{\text{max}}$. Here, in contrast to Fig. 3, we choose much weaker driving field strengths in order to keep the Floquet band structures simpler and the light-induced Berry curvature well localized near the Dirac points. They are initially in equilibrium [Fig. 4(a)], and one observes a nonzero Berry curvature in each valley but with flipped sign between valleys [cf. Fig. 2(d)]. This immediately changes when a driving field is applied [Fig. 4(b)], which leads to a band inversion and sign change of the Berry curvature around $K_1$ and a nonvanishing total Berry curvature in the valence bands (conduction bands) due to broken time-reversal symmetry exceeding the breaking of inversion symmetry.

We remark here that the complexity of the Floquet band structure, with replicas and avoided crossings as discussed above, renders the discussion of a Chern number of Floquet bands somewhat meaningless. The important physical ingredient to understand the consequences of local Berry curvature is the effective winding number that emerges around the Dirac points, and this is locally analogous to a Chern insulator with Chern number $C = 4$. However, it is well known that this does not necessarily imply a fully quantized Hall conductance in the driven state, as discussed by some of us in Ref. [32]. Nevertheless, if a sweet spot for driving the system such that heating is largely avoided could be identified, our results would imply a light-induced anomalous Hall effect similar to single-layer graphene, but with an additional double degeneracy due to the bilayer instead of single-layer structure ($4e^2/h$ instead of $2e^2/h$). We stress here that this quantization is strictly only valid in the limit where the Berry curvature is highly localized in the two valence and two conduction bands very close to the Dirac points. In fact, this is only strictly true in the weakest-field case [Fig. 4(a)] and becomes less clear when some Berry curvature also develops at avoided band crossings. The observed band gap and spreading of the Berry curvature in momentum space become more pronounced as the field strength increases [Figs. 4(c) and 4(d)]. We now turn to the case of vanishing $A$-$B$ potential and nonzero backgate potential $V_{\text{BG}} = 0.05$ eV [Figs. 4(e)–4(h)]. Initially, the Berry curvature vanishes identically everywhere [Fig. 4(e)]. Turning on the laser field, a nonvanishing Berry curvature is induced.
but with compensation between the two valence bands (conduction bands) for a moderate field [Fig. 4(f)]. Increasing the field strength more, one reaches a critical field [Fig. 4(g)] at which the top valence band and bottom conduction band invert and the net valence band Berry curvature as well as the net nonzero valley Berry curvature [Fig. 4(m)], qualitatively equivalently to the case of the single-layer Haldane model. In this case, a moderate driving field creates asymmetric energy gaps at $K_1$ versus $K_2$ [Fig. 4(n)], where one of the gaps at $K_2$ increases due to the laser while the other one at $K_1$ decreases. A gap closing and band inversion are then observed at $K_1$ for a critical field strength [Fig. 4(o)] with a completed transition to a topologically nontrivial Floquet band structure above the critical field strength [(Fig. 4(p)].

We note that these Floquet-engineered Berry curvatures could be measured in a momentum-, energy-, and time-resolved fashion using circularly polarized time- and angle-resolved photoemission spectroscopy [69]. Finally, we
summarize our findings for the Floquet-engineered band gaps and topology in Fig. 5. The red curve in Fig. 5(a) shows the Dirac point band gap at zero onsite potentials (pristine TBG) as a function of laser driving field strength, or as a function of peak vector potential, respectively. The blue (black) line shows $2(v_F A_{\text{max}})^2/\Omega$, respectively, with $v_F$ the renormalized Fermi velocity of TBG and $v_F^0$ the bare Fermi velocity of single-layer graphene. Interestingly, the light-induced band gap scales essentially with the bare Fermi velocity of single-layer graphene as opposed to the renormalized one in TBG (see Appendix D for an extended discussion). For small twist angles, however, deviations from scaling with $v_F^0$ appear. The origin of this scaling behavior presumably lies in the fact that the renormalization of the Fermi velocity stems from interlayer hoppings, which are predominantly perpendicular to the plane of the individual graphene layers. In contrast, the light field is chosen to have in-plane polarization, thus not coupling to the out-of-plane currents. Therefore, the single-layer Fermi velocity is the key ingredient for the light-induced band gap.

Figure 5(b) shows the Floquet band gap at the Dirac points for vanishing onsite potentials at different intermediate twist angles. The peak electric field strength, $E_{\text{max}} = 9.1 \text{ MV/cm}$, and the driving frequency, $\Omega = 2.23 \text{ eV}$, are kept constant. The gap increases as a function of twist angle at small twist angles and then reaches a plateau approaching the gap value for single-layer graphene. Clearly, for $7.34^\circ$ the size of the gap is very close to the single-layer value [70].

Figure 5(c) shows the Floquet phase diagram with the trivial phase (vanishing valence band integrated Berry curvature) and nontrivial phases (nonzero integrated Berry curvature) as a function of field strength for varying symmetric $A$-$B$ potential and a small back-gate voltage. Because of the additional back-gate voltage, the topologically nontrivial phase always requires nonzero driving amplitude. (d) With back-gate voltage only. Throughout, a photon frequency, $\Omega = 2.23 \text{ eV}$, is assumed.
VI. CONCLUSION AND OUTLOOK

We have shown how the Berry curvature in twisted bilayer graphene is affected by different types of inversion-symmetry breaking. The effects of circularly polarized light on the full tight-binding model of twisted bilayer graphene are found to be essentially as expected for two copies of single-layer graphene. Importantly, the light-induced energy gap is not strongly affected by interlayer coupling at twist angles much larger than the largest magic angle. Moreover, the opportunity to break inversion symmetry by back-gating allows us to tune the phase transition between topologically trivial and nontrivial states, in distinct contrast with single-layer graphene. This opens up the realistic prospect of a finite-field topological transition in solid-state experiments. A future task is to study theoretically the ultrafast light-induced transport properties of TBG for realistic driving pulses especially in the midinfrared and for smaller twisting angles near 1.7°, including heating effects by fully taking into account excitation and dissipation in real time, which is a much more formidable task for TBG compared to a single branch of Dirac fermions [32,33]. In particular, real-time signatures of tunneling into topological edge states might provide additional insights [71]. An intriguing opportunity opens up for small twist angles, at which the distance between both Dirac points in the mini-Brillouin zone becomes very small. In this case, the laser field strength could realistically lead to a peak vector potential that exceeds the distance |K₂ − K₁|. In this case, the band gap induced initially by a moderate field amplitude could close again, leading to a second topological phase transition, which should lead to a signature in the Hall transport experiment. Moreover, it is also interesting to address the question of light-induced topology without classical driving fields by encapsulating TBG in a cavity, as recently proposed for single-layer graphene [72].

Finally, the possibility to not only tune single-particle bandwidth and Berry curvature but also interaction effects [51,54] could pave the way for solid-state platforms with exotic nonequilibrium phases of matter analogous to Floquet-engineered interaction phases in artificial lattices [52]. Moreover, the concept of twistronics can also be extended to other two-dimensional materials, such as hexagonal boron nitride [73] and GeSe [74], and it is an intriguing subject for future study to investigate Floquet engineering in these materials.

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APPENDIX A: MODEL

The tight-binding Hamiltonian is of the general form

\[ H = \sum_i \epsilon_i c_i^\dagger c_i + \sum_{i \neq j} t_{ij} c_i^\dagger c_j, \]  

(A1)

FIG. 6. Tight-binding hopping elements. Only \( p_z \) orbitals are taken into account. (a) Intralayer hopping is restricted to \( pp_p \) terms. There are two channels for interlayer hopping: Vertical hopping is dominated by the \( pp_p \) term (b). For nonvertical transitions, also \( pp_p \) terms (c) contribute.

where \( c_i^\dagger (c_i) \) creates (annihilates) an electron in the \( p_z \) orbital at the atomic position \( r_i \). We use hopping matrix elements as introduced in Ref. [58]:

\[ t_{ij} = n^2 V_{pp_p}(r_{ij}) + (1 - n^2)V_{pp_p}(r_{ij}). \]  

(A2)

In the above equation, \( r_{ij} = |r_i - r_j| \) refers to the distance between two sites at positions \( r_i \) and \( r_j \). The real number \( n = z_{ij}/r_{ij} \) defines the ratio between the distance along the \( z \) direction and the total distance. \( V_{pp_p} \) and \( V_{pp_p} \) denote the \( \pi \) and \( \sigma \)-orbital-related Slater and Koster parameters, respectively. For intralayer hopping, only the \( pp_p \) term contributes, as \( z_{ij} = 0 \). For interlayer hopping, both the \( pp_p \) and the \( pp_p \) terms are nonzero \( (z_{ij} = a_1) \). A detailed illustration is shown in Fig. 6. Both hopping contributions are assumed to decay exponentially as a function of distance:

\[ V_{pp_p} = \gamma_0 \exp\left[ q_x (1 - r_{ij}/a)\right], \]  

\[ V_{pp_p} = \gamma_1 \exp\left[ q_x (1 - r_{ij}/a_1)\right]. \]  

(A3)

Here, \( a = 1.418 \) Å is the intralayer nearest-neighbor distance, and \( a_1 = 3.364 \) Å is the interlayer distance. The monolayer lattice constant of the triangular Bravais lattice is \( a_0 = 2.445 \) Å. For the nearest-neighbor hopping, \( \gamma_0 = -3.24 eV \), we add 20% to the DFT-fitted value in order to compensate for many-body effects beyond the employed DFT functional and in order to fit the van Hove singularity in the tunneling density of states reported in Ref. [75]. For the \( \sigma \)-related hopping, we find \( \gamma_1 = 0.55 eV \). For the exponential decay, we find fitting parameters, \( q_x = 3.15 \) and \( q_0 = q_0 \). For the fictitious local potentials on the \( A \) and \( B \) sublattices that break inversion symmetry, \( \epsilon_i \), we assume different symmetry configurations, as shown in Fig. 7. We assume periodic Born–von Kármán boundary conditions within in the \( x-y \) plane. The Hamiltonian in momentum space is then calculated by a Fourier transforms, \( c^\dagger (k) = \sqrt{V_{BZ}} \sum_i c^\dagger_i \exp(ikr_i) \) and \( c(k) = \sqrt{V_{BZ}} \sum_i c_i \exp(-ikr_i) \), where \( V_{BZ} \) is the volume of the two-dimensional Brillouin zone.

APPENDIX B: FLOQUET DYNAMICS

A circularly polarized laser driving field is included by coupling to a time-dependent external gauge field, \( A(t) = A_{\max} [e_y \sin(\Omega t) + e_x \cos(\Omega t)] \), where \( e_y \) and \( e_x \) denote the unit vectors in the spatial in-plane directions, via Peierls
substitution of the form
\[ t_{ij} \rightarrow \tilde{t}_{ij}(t) = t_{ij}e^{i\mathcal{A}(\mathbf{k}_i)}. \]  

The main effect of the circularly polarized field is the breaking of time-reversal symmetry, which induces a mass term to the Dirac fermions, as has been investigated for single-layer graphene [34,40]. The peak electric field strength is calculated numerically with the trapezoidal rule. We ensure convergence of the Berry curvature is sufficiently converged.

\[ \mathcal{H}^{\text{Floquet}} = \frac{1}{T} \int_{0}^{T} dt e^{i(m-n)\Omega t} H(t) + \delta_{mn}m\Omega \mathbb{1}. \] (B2)

The integers \( m \) and \( n \) arise from the Floquet expansion of the time-dependent solutions of the Schrödinger equation,

\[ |\Psi(t)\rangle = \sum_{m=-\infty}^{+\infty} \exp[-i(\epsilon + m\Omega t)] |u_m\rangle, \] (B3)

where \( \epsilon \) is the Floquet quasienergy and \( |u_m\rangle \) is the corresponding Floquet eigenfunction of the Floquet Hamiltonian. For the results shown in the main text, we truncate the expansion after order \( |m| = 2 \) and use 200 sampling points to perform the integral over one period \( T = 2\pi / \Omega \) in Eq. (B2) numerically with the trapezoidal rule. We ensure convergence in the Floquet cutoff by means of the energy gap at the Dirac points with error smaller than \( 10^{-5} \text{eV} \). In order to highlight the bands which overlap with the bare bands of the undriven system, in contrast to Floquet sidebands, we calculate the squared overlap, \( \sum_{nm} |\langle \alpha |e_i\rangle|^2 \), of the Floquet energy eigenstates, \( |e_i\rangle \), with the energy eigenstates of the bare Hamiltonian, \( H(\alpha) = E_\alpha|\alpha\rangle \). Throughout this work, we use units where \( e = \hbar = c = 1 \).

**APPENDIX C: BERRY CURVATURE**

The Berry curvature at a fixed momentum point can be defined by the Berry flux of an infinitesimally small loop around that point, divided by the enclosed area [76],

\[ F^{(n)}(\mathbf{k}) = \lim_{\Delta k_\perp, \Delta k_z \to 0} \frac{F^{(n)}(\mathbf{k})}{\Delta k_\perp, \Delta k_z}. \] (C1)

where \( n \) refers to the corresponding band. On a discretized grid, the flux can be written in the form [77]

\[ F^{(n)} \approx \text{Im} \left( \ln \prod_{j} (u_{n,k_j} |u_{n,k_{j+1}}\rangle \langle u_{n,k_{j+1}} |u_{n,k_j}\rangle) \right). \] (C2)

Here, \( u_{n,k_i} \) refers to the \( n \)-th energy eigenstate of the bare Hamiltonian and the Floquet Hamiltonian, respectively, at quasimomentum \( k_i \), where \( i \in \{1, 2, 3, 4\} \), as depicted in Fig. 8. We use a grid spacing of \( \Delta k = 10^{-5} \text{Å}^{-1} \) for which the Berry curvature is sufficiently converged.

\[ \mathcal{A}(a^{-1}) \]

\[ A_{\text{max}}(a^{-1}) \]

\[ \Theta = 7.34^\circ \]

\[ \Theta = 5.09^\circ \]

\[ \Theta = 3.15^\circ \]

\[ V_{\text{BG}} = 0, V_{\text{AB}} = 0 \]

\[ E_{\text{max}}(\text{MV/cm}) \]

\[ 0.02 \]

\[ 0.04 \]

\[ 0.06 \]

\[ 0.08 \]

\[ 0.1 \]

\[ 0 \]

\[ 0.02 \]

\[ 0.04 \]

\[ 0.06 \]

\[ 0.08 \]

\[ 0.1 \]
APPENDIX D: LIGHT-INDUCED ENERGY GAPS

In Fig. 9, we show the light-induced energy gaps at the Dirac point without inversion-symmetry breaking fields for different twist angles as indicated. In all three cases, we choose as the photon frequency the $\Gamma$-point low-energy bandwidth, $\Delta_{\Gamma} = 2.23$ eV, which we extract from the equilibrium band structure for $\Theta = 7.34^\circ$. Importantly, we find almost perfect scaling behavior with the bare Fermi velocity of single-layer graphene, as opposed to the actual Fermi velocity of the bilayer, for the largest twist angles of 7.34°. For smaller twist angles, we find a deviation and the actual gap lies in between the one expected for single-layer graphene and the one for the bilayer.


[70] The decreasing size of the light-induced gap due to increasing band flatness (smaller twist angles) is confirmed in Fig. 9 of the Appendix D, in which we show the gap dependency on the driving amplitude for two smaller angles.


