Detecting light-induced Floquet band gaps of graphene via trARPES

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We propose a realistic regime to detect the light-induced topological band gap in graphene via time-resolved angle-resolved photoelectron spectroscopy (trARPES), which can be achieved with current technology. The direct observation of Floquet-Bloch bands in graphene is limited by low-mobility, Fourier-broadening, laser-assisted photoemission (LAPE), probe-pulse energy-resolution bounds, space-charge effects, and more. We characterize a regime of low driving frequency and high amplitude of the circularly polarized light that induces an effective band gap at the Dirac point that exceeds the Floquet zone. This circumvents limitations due to energy resolutions and band broadening. The electron distribution across the Floquet replicas in this limit allows for distinguishing LAPE replicas from Floquet replicas. We derive our results from a dissipative master equation approach that gives access to two-point correlation functions and the electron distribution relevant for trARPES measurements.

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I. INTRODUCTION

Floquet engineering constitutes a novel approach to control material properties via light [1–4]. A prominent example is the proposed light-induced topologically insulating state of monolayer graphene [5–7]. The resulting anomalous Hall effect in this system has been observed experimentally [8] and has been explained as a geometric-dissipative effect [9] in accordance with Floquet theory. Meanwhile, time-resolved angle-resolved photoemission spectroscopy (trARPES) has been established as the prime method for resolving dynamical changes in effective band structures of solid-state systems [10–16]. Experimental trARPES setups are constantly improving and being used for investigating the dynamical electronic processes in two-dimensional Dirac materials such as graphene [17–21], WSe₂ [20–24], and Bi₂Se₃ [25–28]. Approaches related to observing pseudospin textures in ARPES were discussed in Refs. [29–32]. In Bi₂Se₃, Floquet replicas of electronic bands were observed using trARPES setups [33]. However, the direct observation of both the replicas and the topological gap at the Dirac point has been met with intricate challenges in graphene and remains unachieved to date.

In this work we determine the regime of trARPES measurements for observing the topological band gap at the Dirac point of irradiated graphene. We propose to perform these measurements in the regime of low driving frequencies and high driving field strengths. In this previously unexplored

regime the dominant Floquet-Bloch band occupations are spaced farther apart than the driving frequency. They are therefore outside of the first Floquet zone. We propose to detect this light-induced energy gap beyond the Floquet zone in experiment because the different spectral features are well resolved in this regime. We discuss the dependence of our predictions on the system parameters, specifically how they affect the systematic limitations of the energy resolution of photoemission spectroscopy. These parameters include the driving frequency and field strength, which determine the Floquet-Bloch band structure, the dissipation coefficients that broaden the band signals, and the pulse lengths of drive and probe lasers. For the pulse lengths we point out a desirable regime with sufficient energy resolution and high enough repetition rates. These repetition rates are necessary to avoid undesired space-charge effects, where the photoemitted electrons interact and affect each other's trajectories [23,34–36]. In particular, it is possible to distinguish the laser-assisted photoemission (LAPE) replicas from the Floquet replicas within the gap at the Dirac point in our suggested regime.

We consider a single layer of graphene irradiated by a circularly polarized infrared laser from a perpendicular direction. We consider a laser pulse with a temporal Gaussian envelope of pulse length τ_d . The pulse length is assumed to be much longer than the driving period, so that it induces Floquet-Bloch states that vary with the envelope function of the pulse. The graphene sample is probed by a tunable extreme ultraviolet laser pulse from the same direction. It has a shorter pulse length and excites photoelectrons out of the driven graphene over a time span during which the driving intensity is approximately constant. This is necessary for resolving the time-dependent Floquet-Bloch bands, which are sensitive to the driving amplitude. This is considered the standard approach to trARPES experiments [37] and is illustrated

FIG. 1. An illustration of the trARPES principle. A circularly polarized infrared drive pulse (red) hits the graphene target from a perpendicular direction, exciting a transient state in the illuminated region. After a time delay of Δt , an ultraviolet probe pulse (blue) hits the target and excites photoelectrons. A given electron leaves the target with a kinetic energy E_{kin} at an inclination θ to the graphene target and at an azimuthal angle ϕ . The momentum component $\vec{k}_{||}$ parallel to the target is that of the preprobe transient state of the electron bound in the graphene layer.

in Fig. 1. The emitted photoelectrons corresponding to a probe frequency ω_p have the kinetic energy

$$E_{\text{kin}} = \hbar\omega_p - E_b - \Phi, \quad (1)$$

where E_b is the binding energy and Φ is the work function of the material, which is the energy required to remove the electron from the graphene. In addition, a photoelectron has momentum $\vec{k}$ with components

$$\vec{k}_{||} = \sqrt{2mE_{\text{kin}}}\hbar^{-1} \sin(\theta) (\cos(\phi), \sin(\phi))^T \quad (2)$$

parallel to the graphene layer. m is the electron mass, and θ and ϕ are the inclination and azimuthal angles of the momentum with respect to the graphene layer. Measuring the photoelectron counts at these angles and energies gives access to the time-resolved Floquet-Bloch bands. The momentum $\vec{k}_{||}$ is the electron momentum prior to the excitation process. For simplicity this is denoted as $\mathbf{k}$ in the following.

II. PREDICTIONS FOR trARPES MEASUREMENTS

We describe the electron dynamics in graphene with the Hamiltonian

$$H(t) = \sum_{\mathbf{k}} c_{\mathbf{k}}^\dagger h(\mathbf{k}, t) c_{\mathbf{k}}, \quad (3)$$

where $c_{\mathbf{k}} = (c_{\mathbf{k},A}, c_{\mathbf{k},B})^T$. Here $c_{\mathbf{k},i}^{(\dagger)}$, with $i \in \{A, B\}$, are the annihilation (creation) operators of the graphene sublattices.

The single-particle Hamiltonian is given by

$$h(\mathbf{k}, t) = \hbar v_F [q_x(t) \sigma_x + q_y(t) \sigma_y], \quad (4)$$

with

$$q_x(t) = k_x + \frac{e}{\hbar} A_d(t) \sin(\omega_d t), \quad (5)$$

$$q_y(t) = k_y + \frac{e}{\hbar} A_d(t) \cos(\omega_d t), \quad (6)$$

where $v_F \approx 10^6 \text{ ms}^{-1}$ is the Fermi velocity and $k_x = |\mathbf{k}| \cos(\phi)$ and $k_y = |\mathbf{k}| \sin(\phi)$ are the momentum components. σ_i are the Pauli matrices. The pulsed vector potential amplitude is given by

$$A_d(t) = \frac{E_d}{\omega_d} \exp \left\{ -t^2 \tau_d^{-2} 4 \ln(2) \right\}, \quad (7)$$

where τ_d is the driving pulse full width at half maximum (FWHM). For the pulse length we use the value $\tau_d = 500 \text{ fs}$ as a realistic value for driving frequencies in the range of tens to hundreds of terahertz. Note that the specific choice of a temporal Gaussian envelope is not crucial for the results. However, more quickly varying temporal envelopes may induce transient behavior that potentially obscure band signals.

We consider a product state $\rho = \prod_{\mathbf{k}} \rho_{\mathbf{k}}$ and evolve the system using a Lindblad–von Neumann master equation that includes dissipation. The dissipation channels amount to dephasing, decay, and electron exchange with a back gate. The corresponding coefficients are chosen to be $\gamma_z = 22.5 \text{ THz} \approx (44.4 \text{ fs})^{-1}$, $\gamma_{\pm} = 10 \text{ THz} = (100 \text{ fs})^{-1}$ and $\gamma_{\text{bg}} = 25 \text{ THz} = (40 \text{ fs})^{-1}$. These are the values that were demonstrated to describe the experimental realization of Ref. [8] in Ref. [9]. Hence, this approach is well suited to describing the dynamics in realistic solid-state samples, despite the potential intricacies of material defects [38–41]. These values for the dissipation coefficients also agree with the relaxation times of 20 to 40 fs found in Refs. [42–45] and the electron-phonon channel relaxation estimated to be on the order of 100 fs [44–46]. This also gives the timescale of the system to form a steady state with an effective Floquet spectrum. We include a nonzero temperature in the system by giving corresponding Boltzmann factors to complementary dissipation coefficients so that the equilibrium states realize the desired temperature T . Throughout this work we use room temperature $T = 300 \text{ K}$. For details of this approach, see Ref. [9] or Appendix A.

Our predictions for the trARPES measurements are based on the momentum- and energy-resolved electron distribution calculated as [47]

$$n(\mathbf{k}, \omega) = \int_{-t_0}^{t_0} \int_{-t_0}^{t_0} s(t_1) s(t_2) \mathcal{G}(\mathbf{k}, t_2, t_1) \frac{e^{i\omega(t_2-t_1)}}{4t_0^2} dt_2 dt_1 \quad (8)$$

with the correlator [48]

$$\mathcal{G}(\mathbf{k}, t_2, t_1) = \sum_{i \in \{A, B\}} \langle c_{\mathbf{k},i}^\dagger(t_2) c_{\mathbf{k},i}(t_1) \rangle \quad (9)$$

and the probe pulse envelope

$$s(t) = \exp \left\{ -(t - \Delta t)^2 \tau_p^{-2} 4 \ln(2) \right\}, \quad (10)$$

where τ_p is the probe pulse length (FWHM) and Δt is the delay time between the incidence of the drive and probe pulses. For the probe length we use the value $\tau_p = 100 \text{ fs}$. t_0 is the

FIG. 2. The electron distribution at the Dirac point $n(\mathbf{k} = 0, \omega)$ for zero delay ($\Delta t = 0$) (a) as a function of the driving field strength at the driving frequency $\omega_d = 2\pi \times 29.8 \text{ THz} \approx 123 \text{ meV} \hbar^{-1}$ and (b) as a function of the driving frequency at driving field strength $E_d = 20 \text{ MV m}^{-1}$. The solid lines show the expected Floquet energies $\pm\sqrt{(ev_F E_d/\omega_d)^2 + (\hbar\omega_d/2)^2} \pm \hbar\omega_d/2$. The dotted lines show the Floquet energies confined within the first Floquet zone of width $\hbar\omega_d$. The dashed lines indicate the field strength at which $\Delta_0 = \hbar\omega_d$ in (a) and the driving frequencies $\Omega_{m=1,2,3}$ in (b). The frequencies $\Omega_{m=1,2,3}$ are defined in Eq. (14). The examples shown in Figs. 3 and 4 also use these frequencies.

temporal integration range, for which we choose $t_0 = 3\tau_d$ to support the probe pulse sufficiently well.

We choose the pulse lengths of the drive and the pump pulse to fulfill two requirements. First, the probe pulse length is chosen to be short compared to the drive pulse length, so that the drive-induced dynamics is resolved. Second, the probe pulse length is chosen to be large compared to the driving period. These conditions are expressed as

$$\tau_d \gg \tau_p \gg \frac{2\pi}{\omega_d}. \quad (11)$$

When the probe pulse length τ_p and the driving period $2\pi/\omega_d$ are comparable, Eq. (8) no longer resolves Floquet-Bloch bands but rather resolves subdriving period electron occupations. We note that increasing the pulse lengths requires increasing the drive pulse energies, which are experimentally limited. This also leads to a reduced repetition rate, which results in undesirable space-charge effects that greatly decrease the resolution due to electron scattering [34,35,49]. We do not include this effect in our numerics but acknowledge that it necessitates a compromise in the pulse lengths, which is reached with the given values of τ_p and τ_d .

III. NONEQUILIBRIUM ELECTRON DISTRIBUTION NEAR THE DIRAC POINT

In Fig. 2(a), we show the electron distribution $n(\mathbf{k} = 0, \omega)$ at the Dirac point at zero delay, i.e., $\Delta t = 0$. We choose the driving frequency $\omega_d = 2\pi \times 29.8 \text{ THz}$ and display the electron distribution as a function of the driving field strength

E_d . We refer to the energy difference of the two distribution maxima that emerge at $\mathbf{k} = 0$ as the energy gap Δ_0 . We see that this gap Δ_0 grows monotonously as a function of E_d rather than being confined within the first Floquet zone of width ω_d , as also discussed in Ref. [50]. We derive the Floquet energy gap at the Dirac point from $h(\mathbf{k} = 0, t)$ with a fixed vector potential amplitude $A_d = E_d/\omega_d$. Using the Rabi solution, we obtain the energy (see Appendix B)

$$\Delta_0/2 = \sqrt{(ev_F E_d/\omega_d)^2 + (\hbar\omega_d/2)^2} - \hbar\omega_d/2. \quad (12)$$

In the following, we point out the most promising regime in which this gap can be detected. As we display in Fig. 2(a), the gap Δ_0 grows with increasing driving strength E_d ; in particular it grows beyond the Floquet zone boundary at $\omega_d/2$. We propose to detect the energy gap Δ_0 in this strongly driven regime in which $\hbar\omega_d \approx \Delta_0$. While the Floquet quasienergies are confined to the Floquet zone, the maxima of the electron distribution continue to be shifted to higher frequencies with increasing E_d so that they can be resolved despite broadening effects and energy resolution limitations. For very large field strengths with $\Delta_0 \gg \hbar\omega_d$, the electrons will predominantly populate the lower bands at the Dirac point. Therefore, intermediate values of E_d are desirable, such that $\Delta_0 \approx \hbar\omega_d$, as we discuss throughout this paper.

In Fig. 2(b), we show the electron distribution $n(\mathbf{k} = 0, \omega)$ at zero delay, i.e., $\Delta t = 0$, at the Dirac point for driving field strength $E_d = 20 \text{ MV m}^{-1}$ as a function of the driving frequency ω_d . We display the energies $\pm\sqrt{(ev_F E_d/\omega_d)^2 + (\hbar\omega_d/2)^2} \pm \hbar\omega_d/2$, which reproduce the

maxima of the electron distribution. We see the expected scaling behavior of the gap at the Dirac point Δ_0 of Eq. (12) as well as the spacing between the nearest Floquet replica $\Delta_0 + \hbar\omega_d$. We propose to measure the electron distribution in the regime that is given by $\omega_d > \gamma$ and $\Delta_0 > \hbar\gamma$. γ is given by $\gamma = \gamma_{\pm}/2 + 2\gamma_z + \gamma_{bg} = 75 \text{ THz} \approx 50 \text{ meV}/\hbar \approx (13.3 \text{ fs})^{-1}$ as an overall metric for the decay rate. This value for γ is comparable to the coherence times of 22 fs found in Ref. [51]. With increasing ω_d and for fixed E_d , Δ_0 decreases. This dependence is predicted by Eq. (12). If Δ_0 is smaller than $\hbar\gamma$, the two maxima of the electron distribution are not resolved and are not detectable via trARPES. With decreasing ω_d , Δ_0 increases and becomes easier to resolve. However, the driving period $2\pi/\omega_d$ needs to be shorter than the characteristic timescale of the dissipative processes, i.e., $\omega_d > \gamma$. Otherwise, the picture of a close to adiabatically stirred Dirac cone in equilibrium is more appropriate than that of emerging Floquet-Bloch bands. Long scattering times have also been connected to the visibility of Δ_0 in Ref. [52]. The range of feasible driving frequencies given by these two conditions decreases for increasing γ but increases for increasing E_d .

In general, the m -photon gaps Δ_m open up at momenta of $v_F|\mathbf{k}| = m\omega_d/2$, with $m > 0$, for small field strengths E_d . These gaps move inwards towards the Dirac point for increasing field strengths. Thus, increasing E_d increases the gap at the Dirac point but at the same time decreases the distance in momentum space to higher-order gaps. For the driving field strength [50]

$$E_d = \frac{\hbar\omega_d^2}{ev_F} \sqrt{\frac{m^2}{4} + \frac{m}{2}} \quad (13)$$

the m th gap is located at $\mathbf{k} = 0$ and merges with the gap Δ_0 . The next gap Δ_{m+1} is then the gap closest to the Dirac point with its distance maximized with respect to E_d . This further enhances the visibility of the gap at the Dirac point and makes this relation between driving field strength and frequency desirable. We rewrite Eq. (13) to find the driving frequency that is necessary for a given field strength E_d to have the gap Δ_0 be equal to m times the driving frequency ω_d . It is

$$\Omega_m = \left(\frac{m^2}{4} + \frac{m}{2} \right)^{-\frac{1}{4}} \sqrt{ev_F \hbar^{-1} E_d}. \quad (14)$$

The driving frequencies $\omega_d = \Omega_m$ have the highest distinguishability and are indicated in Fig. 2(b) as vertical dashed lines. Additionally, at the frequency $\omega_d = \Omega_1 = (\frac{4}{3})^{\frac{1}{4}} \sqrt{ev_F \hbar^{-1} E_d}$, the energy of the first Floquet replica at $\Delta_0/2 + \hbar\omega_d$ is minimized. This point denotes a regime that is well suited for trARPES probing, and the conditions for resolvability simplify to $\gamma < (\frac{4}{3})^{\frac{1}{4}} \sqrt{ev_F \hbar^{-1} E_d}$. For $\gamma = 75 \text{ THz}$ and $E_d \approx 20 \text{ MV m}^{-1}$, this suggests a driving frequency close to $\omega_d = \Omega_1 \approx 2\pi \times 29.8 \text{ THz} \approx 123 \text{ meV}\hbar^{-1}$ or $\omega_d = \Omega_2 \approx 2\pi \times 23 \text{ THz} \approx 96 \text{ meV}\hbar^{-1}$.

To demonstrate the steady state that emerges for driving at frequencies $\omega_d = \Omega_1$, Ω_2 , and Ω_3 , we show the electron distribution $n(\mathbf{k}, \omega)$ in Fig. 3. This expands on the steady-state behavior of the electron distribution $n(\mathbf{k} = 0, \omega)$ that we displayed in Fig. 2. We choose driving field strength $E_d = 20 \text{ MV m}^{-1}$. In the vicinity of the Dirac point, band

FIG. 3. The electron distribution $n(\mathbf{k}, \omega)$ for zero delay ($\Delta t = 0$) and driving field strength $E_d = 20 \text{ MV m}^{-1}$. The driving frequencies are (a) $\omega_d = \Omega_1$, (b) $\omega_d = \Omega_2$, and (c) $\omega_d = \Omega_3$. The dashed lines indicate an effective gap Δ_0 of size $\hbar\omega_d$ in (a), $2\hbar\omega_d$ in (b), and $3\hbar\omega_d$ in (c). The solid lines indicate the instantaneous spectra obtained from the Floquet Hamiltonian.

occupations of the lower Floquet replicas are suppressed. The Floquet replicas with sizable electron occupation are two upper and two lower effective bands at the Dirac point, which are the four bands shown in Fig. 2. As the gap Δ_0 increases, the population is predominantly distributed among the two lower bands. Away from the Dirac point, for nonzero $\mathbf{k}$, the additional Floquet bands have sizable electron occupation and are visible in Fig. 3. The finite values of the electron distribution inside the Floquet gaps is a consequence of the broadening that occurs due to dissipative processes.

Having pointed out the regime that we propose to use to detect the energy gap at the Dirac point in terms of the driving field strength and the driving frequency, we now present the time-resolved response of the system. Figure 4 shows the electron distribution at the Dirac point $n(\mathbf{k} = 0, \omega)$ as a function of the pulse delay time Δt for the same driving field strength and driving frequencies as in Fig. 3, i.e., Ω_1 , Ω_2 , and Ω_3 for $E_d = 20 \text{ MV m}^{-1}$. This gives an estimate of the time-resolved Floquet-Bloch band occupations at the Dirac point. The dashed lines indicate the corresponding Floquet energies expected from static driving field strengths given by the drive pulse at the delay time Δt , i.e.,

$$\epsilon(\Delta t) = \pm \sqrt{\exp\left\{-\frac{\Delta t^2}{\tau_d^2} 8 \ln(2)\right\} \left(\frac{ev_F E_d}{\omega_d}\right)^2 + \left(\frac{\hbar\omega_d}{2}\right)^2} \pm \frac{\hbar\omega_d}{2}. \quad (15)$$

The electron distributions that we show in Fig. 4 are close to the instantaneous steady-state distribution for this value of γ . Deviations from the instantaneous steady-state distribution manifest themselves as features that are asymmetric during the pulse rise and pulse decay. For this choice of γ and of the pulse lengths, these features are small.

IV. LIMITATIONS OF RESOLUTION

One common phenomenon that obscures the results of trARPES is LAPE [53]. The essentially free photoelectrons emitted in a trARPES experiment respond to the drive pulse with driving frequency ω_d . This may result in the photoelectron energy being shifted by one unit of the photon energy $\pm\hbar\omega_d$. These energy shifts are detected in trARPES measurements as band replicas, whose similarities to Floquet replicas might hinder identifying the signatures of Floquet physics unambiguously. However, in contrast to Floquet replicas, these LAPE replicas are not related to band gaps [37]. The magnitude of the light-induced Floquet band gaps is tunable via the field strength E_d [see Eq. (12)]. We propose to use this tunability to distinguish the LAPE and Floquet replicas. More specifically, the Floquet replicas at the Dirac point are at $\pm\Delta_0/2$ and $\pm(\Delta_0/2 + \hbar\omega_d)$, as we show in Fig. 3. The monotonous behavior of the Dirac gap Δ_0 makes it possible to distinguish between LAPE and Floquet replicas.

The Floquet-Bloch bands resolved in $n(\mathbf{k}, \omega)$ are broadened due to dissipation. In addition they are Fourier broadened with the probe pulse length. The combined result is a Voigt

FIG. 4. The electron distribution $n(\mathbf{k} = 0, \omega)$ at the Dirac point as a function of the delay time Δt for a peak driving field strength of $E_d = 20 \text{ MV m}^{-1}$. The driving frequencies are (a) $\omega_d = \Omega_1$, (b) $\omega_d = \Omega_2$, and (c) $\omega_d = \Omega_3$. The dashed lines indicate the static Floquet energies corresponding to the driving field strengths at the center of the probe pulse.

profile of approximate width

$$\Gamma \approx \frac{\gamma}{2} + \sqrt{\frac{\gamma^2}{4} + \frac{4}{\tau_p^2}}, \quad (16)$$

with $\gamma = \gamma_{\pm}/2 + 2\gamma_z + \gamma_{bg}$. For our specified values this is $\Gamma = 80 \text{ THz} = (12.5 \text{ fs})^{-1} \approx 53 \text{ meV}\hbar^{-1}$. In order to successfully resolve the effective bands in $n(\mathbf{k}, \omega)$ it is crucial that the bands gaps are large compared to Γ . For probe pulses long enough that their contribution to Γ can be neglected, the broadening is γ due to intrinsic dissipation. Reducing γ can be achieved by using cleaner graphene samples with higher mobility, which is technologically challenging.

Furthermore, trARPES experiments are, in general, limited by a Gaussian pulse energy resolution of the order [37]

$$\Delta E \approx \tau_p^{-1} 1825 \text{ meV fs}. \quad (17)$$

The energy resolution of the measurement has to exceed the band gap Δ_0 , the Floquet replica spacing ω_d , and the Floquet-Bloch band Voigt width Γ . These requirements are realistically achieved in the proposed regime of $\Delta_0 > \hbar\omega_d$. For instance, fulfilling the resolvability conditions $\Gamma \ll \omega_d$ and $\Delta E \ll \hbar\omega_d$ is not a necessity for identifying signatures of Floquet physics for this regime.

To determine the minimal probe length that is necessary to achieve an energy resolution equal to the gap, we insert the expression for the gap Δ_0 at the Dirac point into Eq. (17). It is

$$\tau_p^{\min} = \frac{1825 \text{ meV fs}}{\sqrt{4e^2 v_F^2 E_d^2 / \omega_d^2 + \hbar^2 \omega_d^2 - \hbar\omega_d}}. \quad (18)$$

An energy resolution several times better is necessary to clearly identify the Floquet-Bloch bands, which corresponds to probe pulse lengths several times larger than the minimal length, e.g., $\tau_p \approx 10\tau_p^{\min}$.

V. CONCLUSION

In conclusion, we have pointed out a realistic regime for the detection of the light-induced topological gap in graphene via time- and angle-resolved photoelectron spectroscopy. Our proposed regime addresses the limitations of band broadening, energy and momentum resolution, and intrinsic limitations of the detection method for realistic estimates of dissipative processes. We find that these limitations are overcome by increasing the driving field strength and decreasing the driving frequency so that the energy difference between finitely occupied Floquet-Bloch bands is larger than the Floquet zone. The timescales associated with the dissipative processes set the limits of this regime. On the one hand, the driving frequency has to be large enough that many driving cycles occur during one characteristic timescale of the dissipation. On the other, decreasing the driving frequency increases the gap size at the Dirac point, which has to exceed the band broadening. As the gap becomes larger than multiples of the driving frequency, limitations such as band broadening and inherent energy resolutions no longer obstruct the identifica-

tion of signatures of Floquet physics. This regime also allows undesired laser-assisted photoemission replicas in trARPES measurements to be unambiguously identified as such at the Dirac point and to be clearly distinguished from the Floquet replicas. The detection of Floquet bands via trARPES would constitute a profound insight in light-driven solids, which would complement the measurements of transport reported in Ref. [8] and thereby advance the field of optical control of solids.

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APPENDIX A: MASTER EQUATION

We base our numerics on the Lindblad–von Neumann master equation

$$\dot{\rho} = i[\rho, H] + \sum_j c_j (L_j \rho L_j^\dagger - \frac{1}{2} \{L_j^\dagger L_j, \rho\}), \quad (A1)$$

where $j \in \{z, +, -, \pm 1, \pm 2, \pm 3, \pm 4\}$. We consider a product state in momentum space $\rho = \prod_k \rho_k$ and the Hamiltonian in Eq. (3) in the extended basis spanned by the states $|0\rangle$, $c_{\mathbf{k},A}^\dagger |0\rangle$, $c_{\mathbf{k},B}^\dagger |0\rangle$, and $c_{\mathbf{k},B}^\dagger c_{\mathbf{k},A}^\dagger |0\rangle$. The transformation V into the instantaneous eigenbasis diagonalizes the Hamiltonian at any given time and determines the momentum-dependent Lindblad operators as

$$L_z = V(c_{\mathbf{k},A}^\dagger c_{\mathbf{k},A} - c_{\mathbf{k},B}^\dagger c_{\mathbf{k},B})V^\dagger, \quad (A2)$$

$$L_+ = V c_{\mathbf{k},B}^\dagger c_{\mathbf{k},A} V^\dagger, \quad (A3)$$

$$L_- = V c_{\mathbf{k},A}^\dagger c_{\mathbf{k},B} V^\dagger, \quad (A4)$$

$$L_l = V \begin{pmatrix} 0 & \delta_{-1,l} & \delta_{-2,l} & 0 \\ \delta_{1,l} & 0 & 0 & \delta_{-3,l} \\ \delta_{2,l} & 0 & 0 & \delta_{-4,l} \\ 0 & \delta_{3,l} & \delta_{4,l} & 0 \end{pmatrix} V^\dagger. \quad (A5)$$

The coefficients c_j are

$$c_z = \gamma_z, \quad (A6)$$

$$c_+ + c_- = \gamma_{\pm}, \quad c_+ = c_- e^{-2\epsilon\beta}, \quad (A7)$$

$$c_j + c_{-j} = \gamma_{bg}, \quad c_{-1,2,3,-4} = c_{1,-2,-3,4} e^{\epsilon\beta}, \quad (A8)$$

with $\beta = (k_B T)^{-1}$ and the instantaneous level spacing ϵ . This four-dimensional description makes two-point correlation functions and therefore the electron distributions via Eqs. (8) and (9) accessible.

APPENDIX B: FLOQUET ENERGIES

At the Dirac point, the graphene Hamiltonian in Eq. (3) with constant amplitude simplifies to

$$h(\mathbf{k} = 0, t) = \frac{ev_F}{\hbar} A_d [\sin(\omega_d t) \sigma_x + \cos(\omega_d t) \sigma_y], \quad (\text{B1})$$

which is the Rabi-Hamiltonian

$$H_R = \hbar \begin{pmatrix} \omega_0/2 & \Omega e^{-i\omega_d t} \\ \Omega e^{+i\omega_d t} & -\omega_0/2 \end{pmatrix}, \quad (\text{B2})$$

with $\omega_0 = 0$. In the rotating frame given by $\exp\{-i\sigma_z \omega_d t/2\}$, the corresponding dynamics are governed by the static Hamiltonian

$$H'_R = \hbar \begin{pmatrix} -\omega_d/2 & \Omega \\ \Omega & +\omega_d/2 \end{pmatrix} \quad (\text{B3})$$

with eigenenergies

$$\epsilon = \hbar \sqrt{\Omega^2 + \omega_d^2/4}. \quad (\text{B4})$$

In the original frame of Eq. (B2) the solutions to the Schrödinger equation then rotate with the energies

$$\epsilon = \pm \hbar (\sqrt{\Omega^2 + \omega_d^2/4} - \omega_d/2), \quad (\text{B5})$$

which gives Eq. (12) for $\Omega = ev_F \hbar^{-1} E_d / \omega_d$.

This expression increases monotonously as a function of E_d , and the Floquet energies are given by this expression modulo the driving frequency ω_d . Put differently, $\epsilon \pm m\hbar\omega_d$ gives the various Floquet replicas at the Dirac point. Whenever $\epsilon = m\hbar\omega_d/2$, the expression of ϵ in Eq. (B5) crosses a Floquet zone boundary. We solve this condition for E_d to find the driving field strengths at which this occurs for the m th Floquet zone boundary. This gives us Eq. (13).

In the case of a time-dependent driving amplitude such as in Eq. (7) and under the assumption that the Floquet states form instantly, we insert the driving amplitude envelope at a given point in time Δt expressed relative to the amplitude peak, such that

$$\Omega(\Delta t) = \frac{ev_F}{\hbar} \frac{E_d}{\omega_d} \exp\{- (\Delta t)^2 \tau_d^{-2} 4 \ln(2)\}. \quad (\text{B6})$$

This leads to the expression $\epsilon(\Delta t)$ in Eq. (15).

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