

# Observation of Nonadiabatic Landau-Zener Tunneling among Floquet States

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Electromagnetic fields induce electronic transitions, generate currents, and fundamentally alter quantum states of matter through strong light-matter interactions. As one established route, Floquet engineering provides a powerful framework to dress electronic states with time-periodic fields, giving rise to quasistationary Floquet states. With increasing field strength, nonperturbative responses of the dressed states emerge, yet their nonlinear dynamics remain challenging to interpret. In this work, we explore the emergence of nonadiabatic Landau-Zener transitions among Floquet states in Cu(111) under intense optical fields. With increasing field strength, we reveal a transition from perturbative dressing to a regime where nonadiabatic tunneling among Floquet states dominate the electronic dynamics. These insights are obtained through interferometrically time-resolved multiphoton photoemission spectroscopy, which serves as a sensitive probe of transient Floquet state dynamics. Numerical simulations and the theory of instantaneous Floquet states allow us to directly examine real-time excitation pathways in this nonperturbative photoemission regime. Our results clarify different light-dressing regimes in materials and provide microscopic insight relevant to ultrafast light wave electronics.

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## I. INTRODUCTION

The study of high optical harmonic generation and the related research in attosecond electron physics motivated research on the light driven nonperturbative light-matter interactions [1–3]. The onset of nonperturbative responses of solids to optical fields is subtle due to their periodic crystalline structures and the dielectric screening. When applied to semiconductors and insulators, valence electrons can be promoted to and accelerated into conduction bands, with the possibility of rescattering with holes, leading to high harmonic generation. Such strong field driven

phenomena are central to the burgeoning field of light wave electronics where THz fields drive PHz frequency electronic responses [4–9]. For metals, the negative real part of the dielectric function,  $\text{Re}(\epsilon) < 0$  [10], couples the optical fields [11–13] with their free electron screening responses, resulting in nonlocal field intensification on attosecond timescales—manifested as the familiar phenomenon of mirror reflection. The enhanced surface fields can drive perturbative multiphoton quantum transitions evident in one or two color multiphoton photoemission (mPP) processes [14–19]. As the driven surface field strength approaches the level of electronic Coulomb potentials, the electronic structure is dressed on an attosecond timescale, offering novel approaches to tailor the electronic structure of matter with light.

The electronic dressing is naturally described within the framework of Floquet engineering, a paradigm in which periodic light fields dress matter and enable control over the electronic band structure [20–25]. In common applications, low frequency fields generate Floquet sidebands within

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band gaps of semiconductors and insulators [26–30], where they have been mapped in the energy-momentum space by angle-resolved photoemission spectroscopy [26,29,31,32]. Resonant matter polarization fields, such as excitons [33–35] or plasmons [36] can also spectroscopically impose multiquanta dressed Floquet states. Directly probing Floquet states is challenging due to their low intensity, adversarial scattering and decoherence, and competing effects such as laser-assisted photoemission effect (LAPE) [30–32,37]. Therefore, it is important to investigate how Floquet states and the associated electronic dynamics [23,28,30,38,39] manifest in spectroscopic experiments. In the strong-field regime, this connection remains elusive as photoemission becomes highly nonlinear—yet the understanding can be valuable for accessing momentum-resolved nonlinear polarization and population dynamics.

Here, we investigate the coherent dressing of Cu(111) surface states, which offers an ideal two-level platform to explore strong-field responses in the presence of many-body screening effects [40]. To this end, we employ

coherent interferometrically time-resolved multiphoton photoemission spectroscopy (ITR-mPP). At lower field strengths, this method has been used to study Floquet sidebands, where their Autler-Townes (AT) splitting becomes visible in the spectra [14] and coherent electron transmission through intermolecular quantum well states on Ag(111) [41]. We extend the study of Cu(111) to higher field strengths to explore the transition from the perturbative to nonperturbative responses with attosecond phase precision and field amplitude control. We show from experimental mPP data that the onset of additional structure at high field strengths can no longer be purely described with the Floquet dressing picture, but stands as a signature of nonadiabatic dynamics—particularly Landau-Zener (LZ) tunneling among the dressed states, as illustrated in Fig. 1(a). The analyzed Fourier components of the measured spectrum can be largely understood in terms of Floquet states and their nonadiabatic dynamics. Furthermore, our results show that the LZ tunneling among Floquet states can be actively controlled by tuning the time

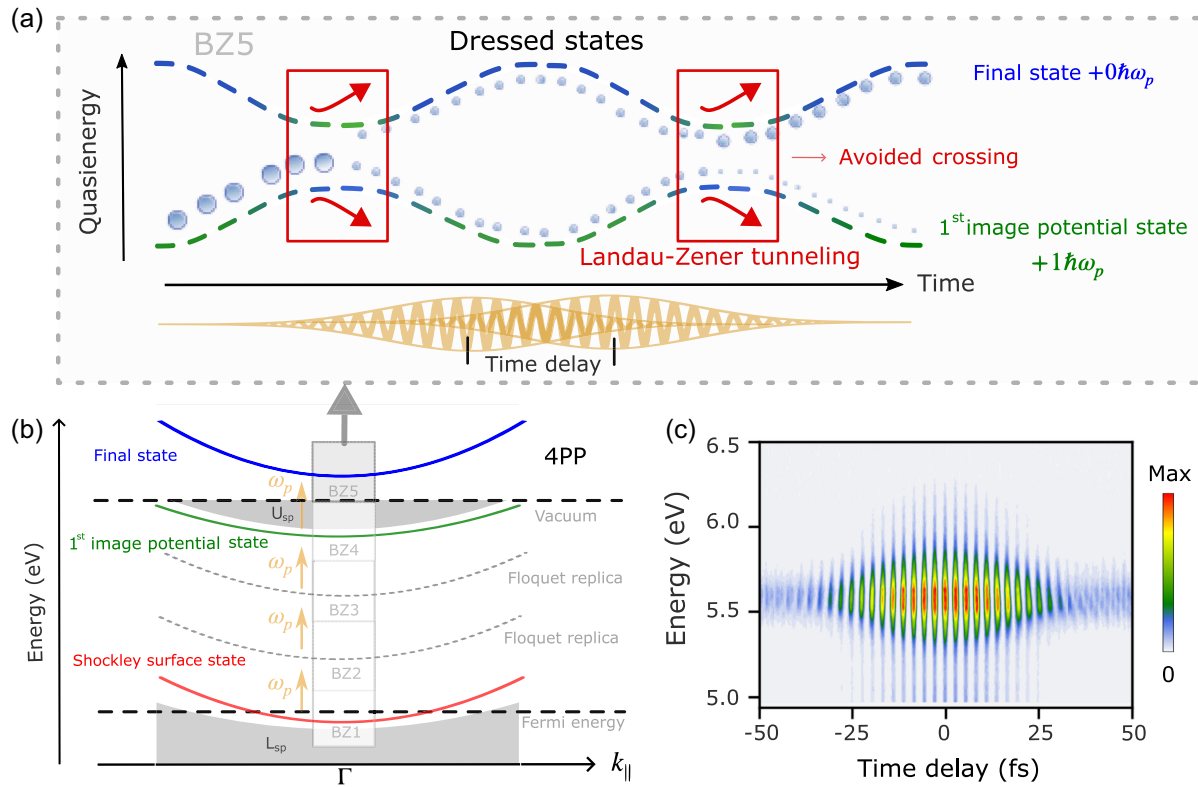


FIG. 1. Schematic of interferometric multiphoton photoemission process in strong field regime. (a) Landau-Zener mechanism among dressed states in photoemission. Following two identical pulses with a time delay (orange lines), the quasienergies of the dressed states evolve as a function of time. Dressed photoelectron final states can form avoided crossings (AC) with other Floquet states, where the dressed final states gain electron population via Landau-Zener tunneling. (b) Sketch of the band dispersion of the Shockley surface state, first image potential state, a photoelectron final state, and their Floquet replicas in different Floquet Brillouin zones (BZ). Electrons are photoemitted from the occupied SS through IP1 into the final state continuum, leading to 4-photon photoemission. SS and IP1 lie within the surface projected band gap between lower and upper  $sp$  band  $L_{sp}$  and  $U_{sp}$  (gray shading).  $\omega_p$  denotes the photon energy. (c) An example of the experimental interferometric-time-resolved 4PP data at  $\Gamma$  with field strength 2.2 V/nm, showing 3D photoelectron counts (color scale), as a function of energy with respect to Fermi level and time delay between two pulses.

delay in the interferometric setup, providing new insight into driving and probing nonadiabatic dynamics in strongly driven systems.

## II. RESULTS

### A. Experimental results

The discontinuity of a periodic crystal lattice at metal-vacuum interfaces supports partially occupied Shockley surface states (SS). Similarly, the Coulomb interaction between a vacuum electron and its screening image charge forms a Rydberg-like series of image potential states (IP) [42,43]. We explore imprinting of the nonadiabatic surface response in interferometric nonlinear four-photon photoemission (4PP) electron distributions when the photon energy is tuned to the three-photon resonance from the SS to the first image potential state (IP1) at  $\Gamma$  ( $k_{\parallel} = 0$ ). Because the decoupling of surface states from the bulk bands and the coherent two-level optical response of Cu(111) are well documented by ultrafast photoemission spectroscopy [44], it is an ideal platform for studying the onset of nonperturbative light-matter interaction with momentum resolution. A schematic of the surface electronic structure and the mPP process is shown in Fig. 1(b).

To study the dressing of the surface electronic structure, two identical 20 fs laser pulses excite the ITR-4PP signal by scanning their time delay with 50 attosecond precision [Fig. 1(c)]. The photon energy  $\hbar\omega_p = 1.54$  eV is set to one-third of the gap between SS and IP1 at the  $\Gamma$  point to enhance the four-photon excitation above the vacuum level by the IP1-SS three-photon resonance. The optical field is sufficiently intense to excite five photon above threshold photoemission (ATP) 5PP signal [45]. In Ref. [14], it is shown that as the laser field is strong enough, ITR-4PP spectra of Cu(111) reveal formation of a Floquet engineered band structure that is characterized by a momentum dependent AT splitting of the coupled bands. Figure 1(c) shows the ITR-4PP interferogram at  $\Gamma$  point with a moderate estimated experimental field strength of  $F_0^{\text{exp}} \sim 2.2$  V/nm. The signal shows nonlinear 4PP oscillation at the driving frequency ( $\omega_p$ ), with an oscillation period of 2.7 fs. The decay of the IP1-SS coherence is slower than the laser pulse with a reported decoherence time around 20 fs [46,47]. Therefore, the dressing of the surface states remains coherent and robust during the initial multiple cycles, which is the primary focus of our study.

Additional ITR-4PP interferograms at  $\Gamma$  recorded for increasing field strength is shown in Figs. 2(e)–2(g). We further apply Fourier analysis to extract their coherent dynamics (see the Appendix for experimental details). In multidimensional spectroscopies, Fourier components have been linked to coherent polarization responses [15,48], and they also reflect signatures of nonlinear effects such as Floquet splittings [14]. Here, we present the energy-, time-, and field-strength-resolved amplitudes of the nonresonant

$2\omega_p$  coherent component in Figs. 4(a), 4(b), 4(d), and 4(e), as the  $2\omega_p$  component is mostly sensitive to the dressing of electronic bands, such as Floquet splitting features [15] (see Supplemental Material [49] for the correspondence between the other Fourier components and ITR-4PP features). The amplitude of the  $2\omega_p$  harmonic signal at a peak field strength of  $F_0^{\text{exp}} \sim 2.2$  V/nm in Fig. 4(a) shows a clear twofold splitting of the Fourier amplitude, which we interpret as the AT splitting of the light-matter coupled Floquet states as the joint pulse field strength reaches its maximum at zero delay [14]. The interest here is when the optical field strength is further increased [Fig. 4(d)], the pulse delay dependent spectrum deviates from the twofold splitting and acquires a dominant central peak between the AT dressed bands. Examining the phase of the  $2\omega_p$ , the low field AT split responses oscillate in phase with each other and the driving field, but the high field response shows a distinct  $\pi$  phase shift relative to the AT side peaks [Fig. 4(e)].

### B. Nonequilibrium Green's function simulations

To simulate the field strength dependent ITR-mPP results, we calculate photocurrent within time-dependent nonequilibrium Green's function (td-NEGF) formalism. We use a one-dimensional real-space model Hamiltonian with Chulkov-type potential  $\hat{V}_c(z)$  [42,43] (See Supplemental Material [49]), in order to reproduce the surface state eigenenergies and band gaps. The light-matter interaction is described with the velocity gauge minimal coupling as

$$\hat{H}(z, t) = \frac{1}{2} \left[ -i \frac{\partial}{\partial z} - A(t) \right]^2 + \hat{V}_c(z), \quad (1)$$

where the vector potential  $A(t)$  describes the surface normal electric field. The photoemission intensity is calculated by integrating photocurrent over time, including entire duration of the pulses. This formalism captures nonperturbative responses including LAPE [56,57] (See the Appendix for details of the simulations).

To first illustrate the field-strength dependence of 4PP, we simulate the spectrum with a single Gaussian pulse, as a function of the peak field strength  $F_0^{\text{sim}}$  in Fig. 2(a). The 4PP spectrum develops observable twofold energy splitting structure at moderate field strengths of  $F_0^{\text{sim}} \leq 5$  V/nm and acquires additional structure as the field strength increases. Up to fields of  $F_0^{\text{sim}} \sim 7$  V/nm, the 4PP primarily probes the AT splitting of Floquet state quasienergies, as represented with the black dots in Fig. 2(a). As  $F_0^{\text{sim}}$  is increased, distinct central peaks start to emerge in a spectral range between the Floquet quasienergies. In Figs. 2(b)–2(d), we select three different peak field strengths  $F_0^{\text{sim}}$  [red dashed lines in Fig. 2(a)], to simulate different regimes of the corresponding ITR-4PP interferograms. Decoherence is not

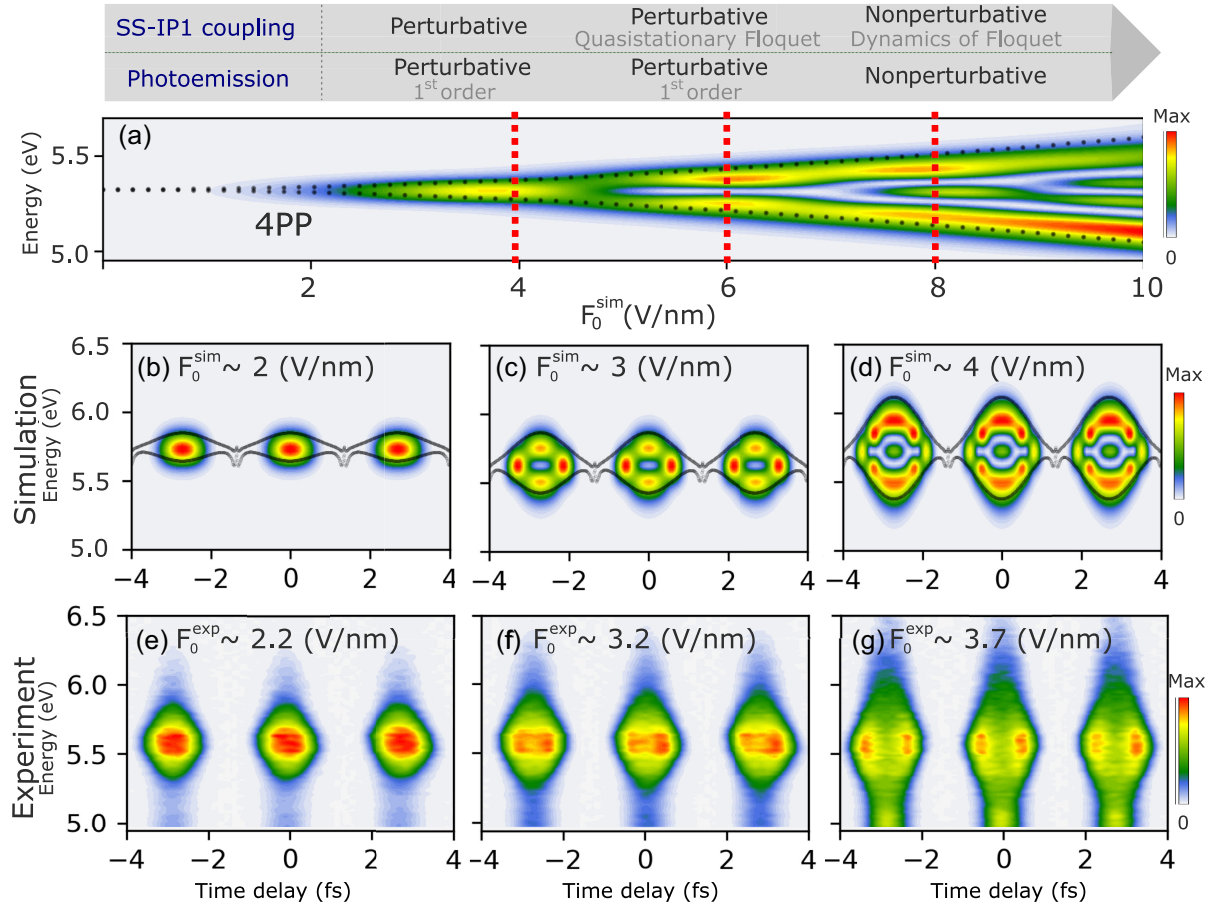


FIG. 2. Field strength dependent NEGF simulation and experiment. (a) Simulated field dependent spectrum (color coding) and the calculated Floquet quasienergy (black dots). The Floquet quasienergies are calculated with basis of SS and IP1, using a continuous sine wave field (see the Appendix), which can be justified if the Gaussian pulse is sufficiently long. (b)–(d) ITR-4PP NEGF simulation with peak field strength  $F_0^{\text{sim}} \sim 2, 3$ , and  $4$  V/nm and the corresponding Floquet quasienergies calculated with effective field parameters averaging over two pulses (black dots). The definition of effective field parameters can be found in Supplemental Material [49]. (e)–(f) Experimental ITR-4PP data with estimated field strength  $F_0^{\text{exp}} \sim 2.2, 3.2$ , and  $3.7$  V/nm. Here we use  $F_0^{\text{sim}}$  and  $F_0^{\text{exp}}$  to denote theoretical and experimental field strength. The energy axis is then referenced to the Fermi energy, with a theoretical work function of  $4.9$  eV in the simulation.

included in the simulation since we focus on the coherent dressing dynamics. Remarkably, the field dependence demonstrates the evolution from twofold to multifold splitting, in accordance to the behavior observed in the experimental  $2\omega_p$  component. At comparable field strengths, the NEGF simulation reproduces our main experimental finding: the emergence of the central  $\pi$  phase shifted  $2\omega_p$  spectral component of ITR-4PP data shown in Figs. 4(c) and 4(f).

When two pulses are in phase, the ITR-4PP signal exhibits a large energy splitting due to enhanced field strength by constructive interference. In contrast, when the pulses are close to being out of phase, the signal vanishes due to destructive interference. This transition between constructive and destructive interference is closely related to the modulation of splitting features. If the pulses are long and time delay is small, the system is approximately dressed by an effective continuous sine wave, as illustrated

in Fig. 3(a). The resulting effective field strength and photon energy—calculated via Fourier analysis—vary with time delay, as plotted in Figs. 3(b) and 3(c) (see Supplemental Material [49]). For Gaussian pulses with peak field strength  $3$  V/nm and photon energy  $\omega_p$ , the effective field strength peaks at zero delay and drops to zero at  $\pm 0.5$  period. The effective photon energy stays close to  $\omega_p$  and shows slight modulation near destructive interference. The corresponding Floquet quasienergies, shown as black dots in Figs. 2(b)–2(d), closely match the main ITR-4PP features (see Supplemental Material [49]). This highlights how the ITR-4PP splitting structure is shaped by the evolution of effective field parameters and their quasienergy trajectories.

From the comparison of the splitting structure and Floquet quasienergies in Figs. 2(a)–2(d), we identify three distinct regimes of the photoemission: (i) For weak fields

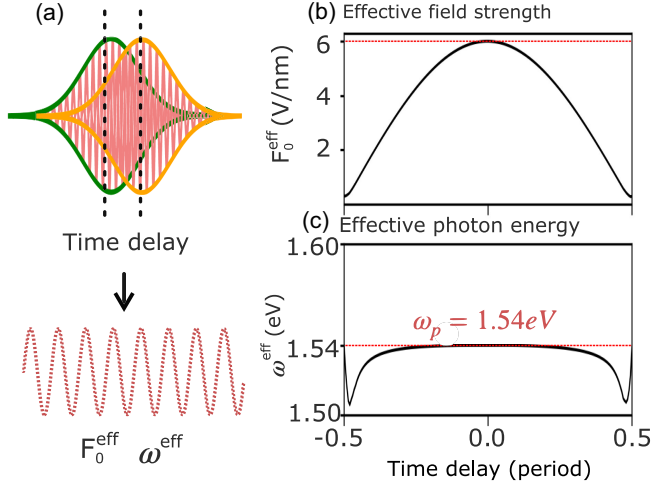


FIG. 3. Illustration of the effective pulse parameters. (a) Dressing with two pulses separated by a small time delay can be approximated as dressing with a continuous sine wave, characterized by an effective field strength  $F_0^{\text{eff}}$  and effective photon energy  $\omega^{\text{eff}}$ , both determined by Fourier analysis. Details are provided in the Supplemental Material [49]. For two Gaussian pulses with a peak field strength of 3 V/nm and photon energy  $\omega_p = 1.54$  eV, the calculated (b) effective field strength and (c) effective photon energy are shown.

$F_0^{\text{sim}} < 5$  V/nm, the 4PP spectrum follows SS to IP1 resonant excitation under Fermi's golden rule. (ii) Intermediate field strengths  $5 \text{ V/nm} < F_0^{\text{sim}} < 7 \text{ V/nm}$  induce significant perturbative dressing of SS and IP1, giving rise to twofold split Floquet states. (iii) In the strong-field regime  $F_0^{\text{sim}} > 7 \text{ V/nm}$ , however, a multifold splitting of bands appears that cannot alone be explained by Floquet physics. Clearly, at high field strengths, nonlinear responses emerge beyond probing of the quasistationary light-dressed states.

### C. Instantaneous Floquet excitation pathway

The agreement between experiments and td-NEGF simulation indicates that the photoemission dynamics can be captured within the manifold of SS, IP1, and photoelectron final states. Nevertheless, the physical origin of the strong field multifold splitting is still unclear.

Now we investigate how the dynamics of Floquet states produce the additional structure within the splitting features and the  $\pi$  phase shift. To get a physical picture of the mPP process, we need to interpret the excitation pathways in terms of Floquet states, and understand *how electrons are promoted from the initial SS via Floquet states to the photoemission final states in real time*. This population dynamics in strongly driven systems can be understood in the language of instantaneous Floquet states (IFS). For a single pulse with sufficiently many cycles, following the envelope of the pulse, we can define Floquet states at the instantaneous field strength. For a sufficiently strong

instantaneous field, the Floquet states form avoided crossings that lead to nonadiabatic dynamics [58]. As we will show, the extra splitting branches in the  $2\omega_p$  harmonics are signatures of such nonadiabatic dynamics, as shown in the sketch in Fig. 1(a).

Any driven state  $|\Phi(t_r)\rangle$  can be expanded with a set of IFS at time  $t_r$  as  $|\Phi(t_r)\rangle = \sum_{\alpha m} c_{\alpha m}(t_r) |\phi_{\alpha, m}(t_r)\rangle$ , where  $|\phi_{\alpha, m}(t_r)\rangle$  is the  $m$ th mode of IFS  $\alpha$  (see the Appendix). Here the photoemission dynamics is modeled with a set of IFS originating from the three relevant states:  $\alpha = \text{SS, IP1,}$  and a photoelectron final state with energy  $E_{\text{final}}$ . The Floquet states excitation pathway is described by the population dynamics  $|c_{\alpha m}(t_r)|^2$  of the IFS, following interaction with two identical Gaussian pulses with a phase (time) delay. We take the final states to be time-reversed low-energy electron diffraction (LEED) states (see Supplemental Material [49]). The mPP signal at energy  $E_{\text{final}}$  is given by the corresponding final state population  $\sum_m |c_{\text{final}, m}(t_r = t_{\text{end}})|^2$  at the end of the pulses. The IFS can form avoided crossings, where the nonadiabatic dynamics can be described by LZ tunneling.

To illustrate, we show two distinct cases of the IFS dynamics leading to photoemission labeled by a star [Figs. 5(a)–5(c)] and a circle [Figs. 5(d)–5(f)]. They are simulated with different final state energies  $E_{\text{final}}$  as the time delay between two Gaussian pulses tunes the states to possible avoided crossings where the LZ mechanism occurs (circle) or not (star). For the star (circle) case, the Gaussian pulses with time delay 0.3 period (0.1 period) are illustrated in Figs. 5(a) and 5(d). Here, the instantaneous photon energy is approximated to be constant  $\omega(t_r) = \omega_p$  over time, and instantaneous field strength  $F_0^{\text{sim}}(t_r)$  follows the envelope for the addition of two pulses. The final states have energy  $E_{\text{final}} \sim 5.05$  eV for the star case and  $E_{\text{final}} \sim 5.0$  eV for the circle case, respectively.

Figures 5(b) and 5(e) show the corresponding real time ( $t_r$ ) dependent IFS quasienergy spectrum within one Floquet Brillouin zone, with the color coding representing the projection of each IFS onto the three relevant states. At the beginning of the excitation ( $t_r = 0$  fs), IFS1, IFS2, and IFS3 are the exact copies of SS, IP1 and the final state, because mixing among them has yet to occur, and IFS1 and IFS2 are degenerate under three photon resonant condition. As the field strength gradually increases following the pulse envelope overlap, the splitting of IFS1 and IFS2 ensues. At  $t_r \sim 50$  fs, the peak field strength causes IFS3 to acquire some SS/IP1 character.

In the star case, the population dynamics is entirely simulated with a finite-space time-dependent Schrödinger equation (TDSE) (see the Appendix). Whereas in the circle case, IFS2 and IFS3 form avoided crossings at time  $t_r \sim 40$  fs (AC1) and time  $t_r \sim 60$  fs (AC2). We categorize the dynamics into type I and II, as labeled in Fig. 5(e). For type I, the dynamics are simulated with a finite-space TDSE as in the star case. At the beginning of the pulse, only

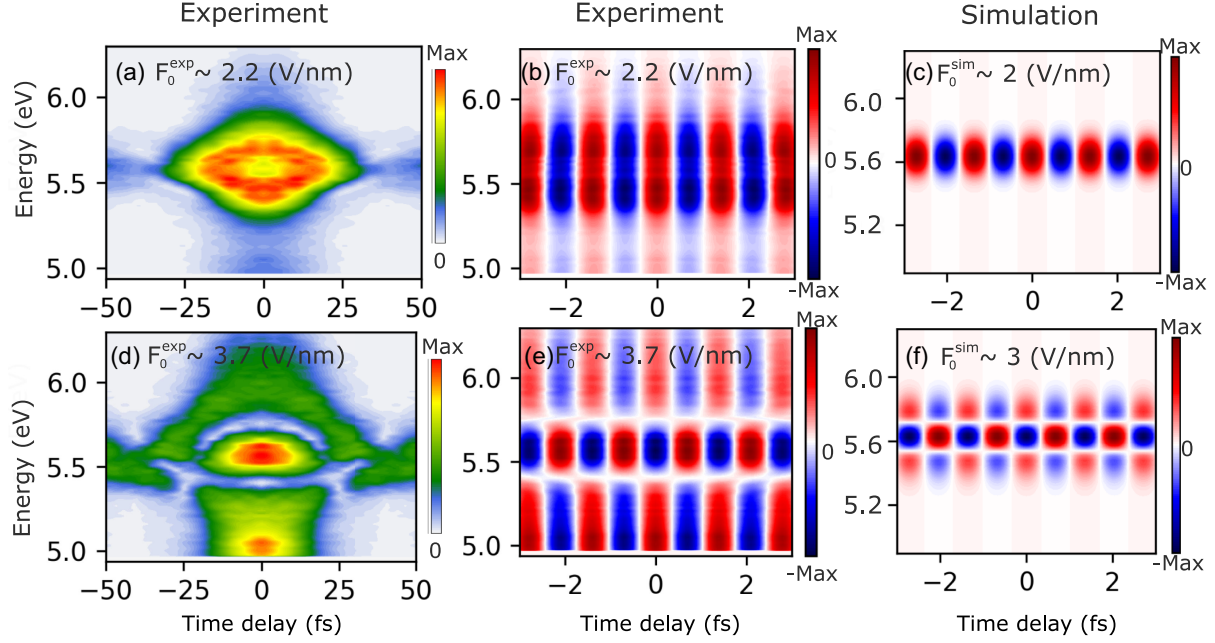


FIG. 4. Comparison of the  $2\omega_p$  coherent polarization fields in experiment and simulation. The  $2\omega_p$  harmonics is obtained by moving the window forward-backward Fourier transformation and inverse Fourier transformation with a Gaussian profile centered at  $2\omega_p$ . (a) The envelope function and the (b)  $2\omega_p$  oscillation at  $F_0^{\text{exp}} \sim 2.2$  V/nm shows twofold in phase splitting. (d) The envelope function and the (e)  $2\omega_p$  oscillation at  $F_0^{\text{exp}} \sim 3.7$  V/nm shows threefold splitting with a  $\pi$  phase shift in the central peak. (c),(f) The simulated  $2\omega_p$  oscillation at  $F_0^{\text{sim}} \sim 2$  and  $F_0^{\text{sim}} \sim 3$  V/nm. The energy axis is referenced to the Fermi energy, with a theoretical work function of 4.9 eV in the simulation.

IFS1 is populated since SS is the only occupied state. The time-dependent populations [Figs. 5(c) and 5(f)] show clear coherent oscillation between IFS1 and IFS2 with the amplitude proportional to the field envelope. The contribution of coherent oscillation to IFS3 population is minimal, because the matrix elements between the final state and two surface states are much smaller than the matrix element between SS and IP1. Consequently, the population dynamics of IFS3 in type I region remains adiabatic. The time-dependent wave-function coefficient  $c_{am}(t_r)$  projected onto the manifold of IFS1 and IFS2 can be visualized on a Bloch sphere in Fig. 5(k), where the wave-function trajectory evolves with a constant radius, indicating that the coherent populations dynamics evolve mostly between the IFS1 and IFS2.

In the circle case, however, upon crossing AC1, the type II dynamics becomes nonadiabatic and is described by an LZ transfer matrix [58]

$$\hat{\mathbf{T}}_{ii'} = \begin{pmatrix} \sqrt{1 - P_{ii'}} e^{-i\phi_{ii'}^s} & -\sqrt{P_{ii'}} \\ \sqrt{P_{ii'}} & \sqrt{1 - P_{ii'}} e^{+i\phi_{ii'}^s} \end{pmatrix}, \quad (2)$$

where  $i = \{am\}$  denotes the  $m$ th Floquet replica of IFS  $\alpha$ . The Stokes phase  $\phi_{ii'}^s$  and  $P_{ii'}$  are defined by details of the avoided crossing gaps (see the Appendix). Because of

the finite off-diagonal elements in Eq. (2), part of the population exchange between IFS2 and IFS3 at both AC1 and AC2. On the Bloch sphere formed within the space of IFS2 and IFS3 in Fig. 5(l), the Landau-Zener mechanism significantly alters the orbits before and after AC2. Because the dynamics of IFS3 remains adiabatic in the type-I part, after crossing AC2, most of the population at IFS3 persist until the end of the pulse, leading to a finite photoemission signal. By contrast, the absence of LZ tunneling in the star case does not enhance the 4PP signal. In this picture, we are able to demonstrate how exactly the mPP process occurs in real time: the population oscillates between SS and the intermediate IP1 coherently, and “jumps” to the final state via a nonadiabatic LZ mechanism.

To elucidate the role of LZ tunneling, we simulate the interferometric 4PP by recording the population of IFS3 for the entire range of  $E_{\text{final}}$  and varying time delay between the two pulses over one cycle. The simulation in Figs. 5(g)–5(j) is performed for two peak field strengths ( $F_0^{\text{sim}} = 1.8$  and  $F_0^{\text{sim}} = 3.5$  V/nm), in order to show the impact of the nonadiabatic dynamics on ITR-4PP. The IFS approach remarkably reproduces the field strength dependent experimental 4PP features—the emergence  $\pi$  phase shift in strong field in Fig. 5(j).

The observed  $\pi$  phase shift originates from the overall structure of the ITR-4PP features in the  $E_{\text{final}}$ -time delay plane. In Fig. 5(i), the finite ITR-4PP with a stronger field

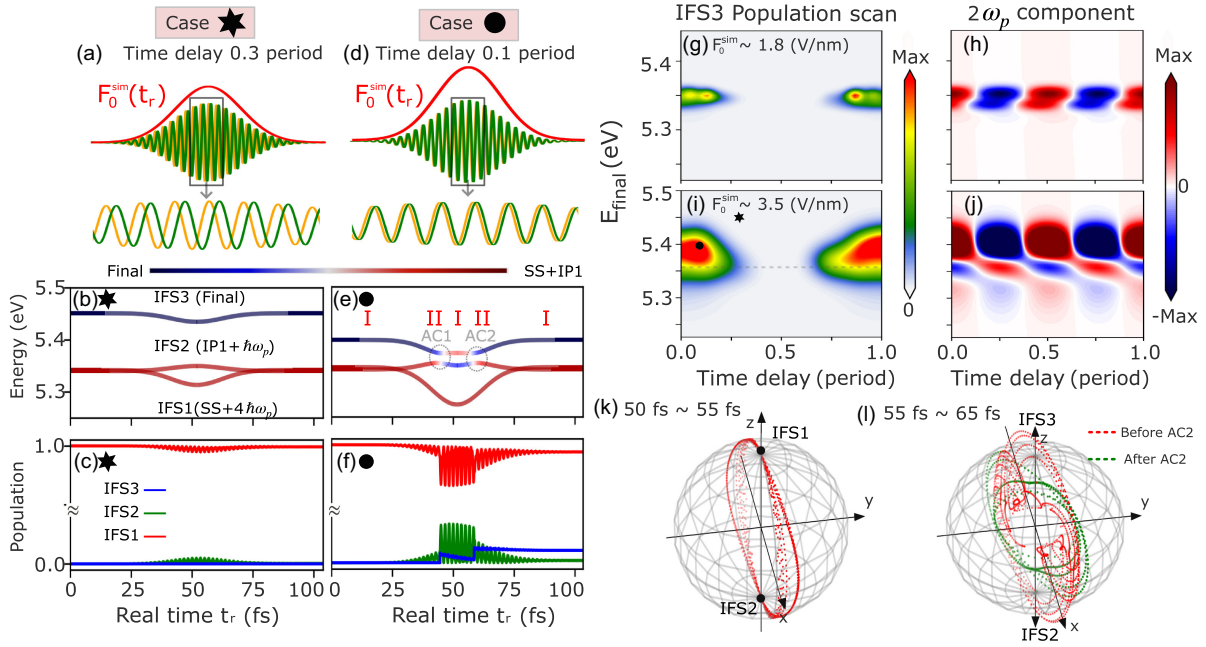


FIG. 5. Dynamics of instantaneous Floquet states. (a)–(f) Two example cases of the IFS dynamics. Two identical Gaussian pulses with time delay (a) 0.3 period and (d) 0.1 period are used to calculate the corresponding real-time ( $t_r$ ) dependent IFS quasienergy spectrum with final state energy (b)  $E_{\text{final}} \sim 5.45$  and (e)  $E_{\text{final}} \sim 5.4$  eV. The color coding shows the character of each IFS, i.e., projection of each IFS onto three relevant states (SS, IP1, and final state). (c), (f) The corresponding time-dependent population of three IFS, where the population of IFS3 is multiplied by 5 for visualization. IFS2 and IFS3 form avoided crossing points (AC1, AC2) in (e), and the Landau-Zener mechanism leads the population jump in (f). (g), (i) Population of IFS3 at the end of the pulse with varying final state energy ( $E_{\text{final}}$ ) and time delay, where the shaded circle and star in i correspond to two cases in (a)–(f). The gray dashed line in i labels the energy for  $\pi$  phase shift in the  $2\omega_p$  component. (h), (j) The corresponding  $2\omega_p$  harmonics. (k), (l) Coherent oscillation of the driven wave function in the case circle calculation [(d)–(f)] visualized with Bloch sphere for (k) IFS1 and IFS2 between 50 and 55 fs and (l) IFS2 and IFS3 between 55 and 65 fs. Radius for both spheres is normalized for better visualization. The energy axes and  $E_{\text{final}}$  are referenced to the theoretical Fermi energy with a theoretical work function of 4.9 eV.

shows enhanced photoemission signal between the fringe maxima, compared to the weaker field case in Fig. 5(g). Specifically, at  $E_{\text{final}} = 5.36$  eV (gray dashed line), the boundary of the ITR-4PP in Fig. 5(i) extends toward the out-of-phase time delays, resulting in significant intensity at delays of 0.25 and 0.75 period. This extension of the 4PP intensity into the out-of-phase time delays results in a  $\pi$  phase shift in the extracted  $2\omega_p$  component. A similar effect is observed in the experimental ITR-4PP data, where the fringes under the strong field extend toward out-of-phase time delays, leading to the  $\pi$  phase shift. (See Supplemental Material [49] for details on the harmonic analysis).

Interestingly, the overall ITR-4PP features associated with the harmonic structure can be interpreted as trajectories of effective quasienergies in the  $E_{\text{final}}$ -time delay plane. Figures 2(b)–2(d) show that the simulated ITR-4PP features are indeed roughly outlined by these effective quasienergies. A stronger field increases the Floquet quasienergy splitting, thereby broadening the range of 4PP features. In the Supplemental Material [49], we further demonstrate that the field dependence of the  $2\omega_p$  harmonic components can be well approximated by the behavior of effective quasienergies. From the IFS perspective, the

boundary of the finite 4PP intensity region is approximately defined by the “touching” points between IFS3 and IFS1/IFS2. When IFS3 intersects with another IFS and forms an avoided crossing, Landau-Zener tunneling can occur, contributing to photoemission. Analogous to the effective Floquet quasienergy picture, increasing the field strength enhances the splitting between IFS1 and IFS2, thereby expanding the energy range over which ITR-4PP features appear (see Supplemental Material [49] for further discussion on touching points among IFS).

### III. DISCUSSION

We have systematically explored different regimes of light-matter interaction on the Cu(111) surface. For moderate field strength, the 4PP spectrum shows evidence of Floquet splitting as the field strength increases [14], which is explained with light dressing of the SS-IP1 two level system. Identifying such light-induced hybridization phenomena in conventional pump-probe optical and photoemission experiments has been precluded by the overlap of multiple effects in frequency and time. In particular, the presence of decoherence and LAPE can complicate the

identification of Floquet states [28–32,37]. By contrast, the Fourier harmonic components extracted via the interferometric technique isolate coherent processes that oscillate at integer multiples of the photon energy. These components effectively allow better resolution of the splitting Floquet states with intermediate field strength, as shown in Fig. 4(b). (A more detailed discussion is presented in the Supplemental Material [49].)

For even stronger driving strengths, both the light-matter coupling and the photoemission process itself become nonperturbative. This largely unexplored territory of strong-field photoemission exhibits features beyond the adiabatic switching of photon dressing. From both experimental observations and theoretical analysis, we identify a fracturing of the photoemission harmonic components into multiple branches in the strong-field regime. These features suggest that photocurrents are generated via nonadiabatic LZ tunneling into dressed photoelectron states. In particular, the multibranch structure and the relative phase shifts in the  $2\omega_p$  component reveal the details of the nonadiabatic excitation dynamics.

The LZ optical field induced nonadiabatic coupling lies at the heart of strong field physics. For instance, in HHG from solids [5,59], the interplay between complex band dispersion or topology [60,61] often lead to nonperturbative and nonlinear spectra at higher order. In particular, the nonlinear dynamic Bloch oscillation in Dirac cones gives rise to efficient HHG [62,63]. The LZ mechanism [64,65] and its connection to quantum geometry of the Bloch electrons [60,66] also play key roles in these processes.

Furthermore, light wave currents in strongly driven graphene can be understood within the framework of Landau-Zener-Stückelberg-Majorana (LZSM) interference [67,68], where the conduction-band population exhibits complex interference patterns governed by the phase accumulated between successive LZ transitions. These interference patterns, and the resulting nonlinear population dynamics, are highly sensitive to field parameters [69–72], including the field strength, the phase delay between pulses, or the carrier-envelope phase (see Supplemental Material [49] for examples of population control in a driven two-level system and in graphene). In analogy, ITR-4PP can be modeled by a driven three-level system, where the time delay—controlled with attosecond precision—tunes the population of the final states, giving rise to the complex features observed in Fig. 2. Momentum-resolved ITR-4PP could directly map the momentum-dependent excited-state populations, thereby providing new insights into the microscopic origins of light wave currents and other nonperturbative strong-field phenomena [73–75].

The framework of IFS offers deeper insight into strongly driven systems. In two-level systems, population transfer can be described by LZSM interferometry within the IFS picture [58,72] (see Supplemental Material [49] for IFS analysis of a driven two-level system). In ITR-4PP, the time

delay tunes LZ tunneling among IFS, leading to the observed  $\pi$  phase shift, while the overall features trace the trajectories of the effective Floquet quasienergies. These results underscore the value of the dressed-state perspective for understanding strong-field processes.

A direct view of the microscopic processes underlying strong-field phenomena has remained elusive, since the inherent momentum- and time-integration in optical or transport experiments masks the nonadiabatic transitions. Moreover, photoemission experiments directly monitor electron currents [76], which provide information about the nonadiabatic dynamics, whereas optical measurements detect photons whose generation involves a convolution of both electron and hole dynamics. In principle, the connection among ITR-4PP, LZSM interferometry, and the Floquet picture applies broadly across material classes. Our work paves a way for a deeper understanding of strong-field dynamics and can inform the future development of ultrafast light wave electronics.

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## DATA AVAILABILITY

The data that support the findings of this article are openly available [55], embargo periods may apply.

## APPENDIX

### 1. Experiments

The interferometrically time-resolved multiphoton photoemission experiments (ITR-mPP) have been performed in the in-house laboratory for time-resolved photoemission spectroscopy at the University of Pittsburgh. The optical and the ultrahigh-vacuum setups are described in Refs. [15,77]. In addition, Ref. [14] provides a detailed discussion on data handling and Fourier analysis of the ITR-mPP experiments.

The single-crystal Cu(111) surface is prepared by repeated cycles of sputtering and annealing under ultrahigh vacuum conditions ( $< 10^{-10}$  mbar). The quality of the surface is judged by the observation of sharp and dominant photoemission signatures from the SS and the IP state in static mPP experiments. 20 fs infrared laser pulses are generated with a noncollinear optical parametric amplifier

(NOPA) pumped by a fiber laser oscillator-amplifier system operating at 1 MHz pulse rate (Clark MXR Impulse). The pulse duration is evaluated by interferometric autocorrelation mPP measurements on the polycrystalline tantalum sample holder. The vacuum electric field strength  $F_0^{\text{exc}}$  is on the order of 0.1–4 V/nm and can be adjusted by tuning the distance between the focusing lens and the sample while keeping the average power constant. The phase-locked pulse-pair for the ITR-mPP excitation emerges from a passively stable, home-built Mach-Zehnder interferometer [78], which is calibrated by recording the interference fringes at the center excitation laser wavelength. The  $p$ -polarized pulse-pair light illuminates the sample at an angle of incidence of  $45^\circ$ . Photoelectron energy-momentum spectra are recorded at each 100 as an increment of pump-probe time delay with a hemispherical energy analyzer (SPECS Phoibos 100).

## 2. Floquet quasienergy calculation

For a time periodic Hamiltonian  $\hat{H}(t+T) = \hat{H}(t)$  with frequency  $\omega = 2\pi/T$ , the solutions to the time-dependent Schrödinger equation (TDSE) are Floquet states  $|\psi_\alpha(t)\rangle$ , which are labeled with quasienergy  $\epsilon_\alpha$  as

$$|\psi_\alpha(t)\rangle = e^{-i\epsilon_\alpha t} |\Phi_\alpha(t)\rangle = e^{-i\epsilon_\alpha t} \sum_m e^{-im\omega t} |\phi_\alpha^m\rangle, \quad (\text{A1})$$

where  $|\phi_\alpha^m\rangle$  is the  $m$ th harmonic of the time periodic orbital  $|\Phi_\alpha(t)\rangle = |\Phi_\alpha(t+T)\rangle$ .

We use the time-dependent Hamiltonian Eq. (1) and projected onto the subspace spanned by the basis states  $i, j = \text{SS, IP1}$ . The pulse  $A(t)$  is replaced with a continuous sine wave  $A_c(t)$  to restore time periodicity as

$$h^{\text{sub}}(t) = \begin{pmatrix} E_{\text{SS}} & O_{\text{SS,IP1}}(t) \\ O_{\text{IP1,SS}}(t) & E_{\text{IP1}} \end{pmatrix}, \quad (\text{A2})$$

with the off-diagonal matrix elements

$$O_{ij}(t) = A_c(t) \langle i | \hat{p} | j \rangle. \quad (\text{A3})$$

$E_i$  is the energy of state  $i = \text{SS, IP1}$  solved from the one dimensional model Hamiltonian with Chulkov potential (see Supplemental Material [49]). The Floquet quasienergy  $\epsilon_\alpha$  can be solved from the time-independent Floquet Hamiltonian as

$$H_{mn} = \frac{1}{T} \int_0^T dt h^{\text{sub}}(t) e^{i(m-n)\omega t}, \quad (\text{A4})$$

$$\sum_m (H_{mn} - m\omega \delta_{mn}) |\phi_\alpha^m\rangle = \epsilon_\alpha |\phi_\alpha^n\rangle. \quad (\text{A5})$$

We use a sufficiently large index of Floquet Brillouin zone  $m, n = -12 \dots +12$ , for the Floquet quasienergies to

converge. In Fig. 2(a), the black dots are the calculated Floquet quasienergies as a function of field strength.

## 3. Nonequilibrium Green's function simulation

We build upon the embedding time-resolved photoemission theory from Ref. [79] and extended it to the non-perturbative regime. To this end, we construct the extended Hamiltonian  $\hat{H}^{\text{PE}}$  including SS and IP1 to a set of photoelectron final states with momentum  $p$ :

$$\hat{H}^{\text{PE}}(t) = \hat{H}^{\text{sub}}(t) + \sum_p \epsilon_p(t) \hat{d}_p^\dagger \hat{d}_p - \left[ qA(t) \sum_{i,p} M_i(E) \hat{d}_p^\dagger \hat{c}_i + \text{H.c.} \right]. \quad (\text{A6})$$

Here,  $\hat{H}^{\text{sub}}(t) = \sum_{ij} h_{ij}^{\text{sub}}(t) \hat{c}_i^\dagger \hat{c}_j$  denotes the minimal-coupling Hamiltonian with  $h_{ij}^{\text{sub}}(t)$  defined in Eq. (A2). The operators  $\hat{c}_i^\dagger$  ( $\hat{c}_i$ ) denote the creation (annihilation) operator for state  $i = \text{SS, IP1}$ , while  $\hat{d}_p^\dagger$  ( $\hat{d}_p$ ) stand for a photoelectron state with out-of-plane momentum  $p$  and energy  $E = p^2/2$ . The vector  $A(t)$  describes the Gaussian profile of the pulse. The dressing of the photoelectrons in the laser field is incorporated by the time-dependent energy  $\epsilon_p(t) = [p - qA(t)]^2/2$ . In what follows we assume the continuum photoelectrons has been discretized and use the final state energy  $E$  as a discrete quantum number. The photoemission matrix elements are calculated in the velocity gauge as  $M_i(E) = \langle \chi_E | \hat{p} | i \rangle$ , where  $|\chi_E\rangle$  denotes the scattering state with outgoing boundary conditions calculated from the Chulkov potential (see Supplemental Material [49] for details).

We define the one-particle Green's function in the subspace of surface states by  $G_{ij}(t, t') = -i \langle T_C \hat{c}_i(t) \hat{c}_j^\dagger(t') \rangle$ . The Green's function is obtained as the solution to the Kadanoff-Baym equation (KBE):

$$\left[ i \frac{d}{dt} - \mathbf{h}^{\text{sub}}(t) \right] \mathbf{G}(t, t') = \delta_C(t, t') + \int_C d\bar{t} \mathbf{\Sigma}(t, \bar{t}) \mathbf{G}(\bar{t}, t'), \quad (\text{A7})$$

where the bold symbol  $\mathbf{G}$  denotes the two-by-two matrix-valued Green's function. The matrix elements of embedding self energy  $\mathbf{\Sigma}$  are calculated as

$$\Sigma_{ij}(t, t') \equiv \sum_E \Sigma_{ij}(E; t, t') = \sum_E M_i^*(E) g_E(t, t') M_j(E), \quad (\text{A8})$$

where  $g_E(t, t')$  is the Volkov Green's function. Finally, the current flowing into the photoelectron subsystem  $\dot{N}_E(t)$  is computed from the transient Meir-Wingreen formula [80]. The photoelectron intensity is then given by

$I(E) = \int_0^\infty dt \dot{N}_E(t)$ . We thus obtain the following expression:

$$I(E) \propto \text{ReTr} \int_0^\infty dt \int_0^t dt' \mathbf{\Sigma}^R(E; t, t') \mathbf{G}^<(t', t). \quad (\text{A9})$$

For the simulation in Figs. 2 and 4, we use a work function of 4.9 eV, and the energy axis is referenced to as the Fermi energy.

#### 4. Instantaneous Floquet states (IFS) dynamics

Within IFS theory, time dependence is introduced to Floquet states by following the pulse profile as a function of real time ( $t_r$ ) and constructing instantaneous Floquet states. At time  $t_r$ , the Floquet states with respect to instantaneous photon energy  $\omega(t_r)$  and the pulse envelope  $F_0(t_r)$  form a complete set [Note that in the main text we use  $F_0^{\text{sim}}(t_r)$  to clarify this is a field strength for simulation]. An IFS  $\alpha$  at real time  $t_r$  is defined as

$$|\psi_\alpha(F_0(t_r), \omega(t_r), t)\rangle = e^{-ie_\alpha(t_r)t} \sum_m e^{-im\omega(t_r)t} |\phi_{\alpha,m}(t_r)\rangle, \quad (\text{A10})$$

where  $|\phi_{\alpha,m}(t_r)\rangle$  denotes  $m$ th instantaneous Floquet mode and  $\epsilon_\alpha(t_r) + m\omega(t_r)$  is the instantaneous Floquet quasienergy displaced into the  $m$ th Floquet Brillouin zone. The photoemission process can be modeled as a driven three states system with SS, IP1, and a final photoelectron state. Here we focus on the dynamics within the first Floquet Brillouin zone above vacuum [BZ5 in Fig. 1(a) and 1(b)], and the dynamics are categorized into two types based on IFS quasienergy spectrum.

##### a. Type I. Adiabatic evolution for final state

When the final state energy is far away from the Floquet replicas of SS and IP1 in BZ5 ( $E_{\text{SS}} + 4\omega_p = E_{\text{IP1}} + 1\omega_p$ ), dynamics of the final state is mostly adiabatic. At the beginning and end of the pulse, the resonant condition between SS and IP1 makes IFS1/IFS2 degenerate. We consider a finite space-time evolution with light-matter interaction. A driven state  $|\Psi(t_r)\rangle = \sum_{i \in \{\text{SS}, \text{IP1}, \text{final}\}} c_i(t_r) |i\rangle$  follows TDSE as

$$i \frac{d}{dt_r} c_i(t_r) = \sum_j H_{ij}(t_r) c_j(t_r), \quad (\text{A11})$$

with the Hamiltonian

$$H(t_r) = \begin{pmatrix} E_{\text{SS}} & O_{\text{SS}, \text{IP1}}(t_r) & O_{\text{SS}, \text{final}}(t_r) \\ O_{\text{IP1}, \text{SS}}(t_r) & E_{\text{IP1}} & O_{\text{IP1}, \text{final}}(t_r) \\ O_{\text{final}, \text{SS}}(t_r) & O_{\text{final}, \text{IP1}}(t_r) & E_{\text{final}} \end{pmatrix}. \quad (\text{A12})$$

The final state energy  $E_{\text{final}}$  and the corresponding matrix elements are solved from Chulkov model Hamiltonian with suitable boundary condition (see Supplemental Material [49]). The off-diagonal matrix elements  $O_{ij}(t)$  are defined as in Eq. (A3).

Similarly, the driven state  $|\Psi(t_r)\rangle$  can also be expanded with IFS basis in Eq. (A10) as

$$|\Psi(t_r)\rangle = \sum_{\alpha m} c_{\alpha m}(t_r) |\phi_{\alpha,m}[F_0(t_r), \omega(t_r)]\rangle. \quad (\text{A13})$$

Because of the energy periodicity of the Floquet states and unitarity, we have  $c_{\alpha m}(t_r) = \sum_i V_{ai}(t_r) c_i(t_r) / \sqrt{N_f}$ , where  $N_f = 25$  denotes the truncated number of Floquet Brillouin zone and we approximate the transformation between real states ( $i = \text{SS}, \text{IP1}, \text{final}$ ) and the IFS ( $\alpha = \text{IFS1}, \text{IFS2}, \text{IFS3}$ ) as a three-by-three identity matrix  $\mathbf{V}(t_r) = \mathbf{I}$  due to the adiabaticity. Such approximation is justified by the fact that the character of each IFS does not evolve significantly during type I dynamics, as shown in Figs. 5(b) and 5(e). In a Floquet Brillouin zone  $m_0$ , we denote  $c_{\alpha m_0}(t_r)$  as a column vector  $\mathbf{c}_{m_0}(t_r)$ . The evolution between time  $t_1$  and  $t_2$  can then be expressed as a unitary evolution  $\hat{\mathbf{U}}_{m_0}(t_2, t_1) \mathbf{c}_{m_0}(t_1) = \mathbf{c}_{m_0}(t_2)$ , following the Hamiltonian in Eq. (A12).

##### b. Type II. Landau-Zener mechanism among IFS

When the final state energy is close to the Floquet replicas of the surface states in BZ5 ( $E_{\text{SS}} + 4\omega_p = E_{\text{IP1}} + 1\omega_p$ ), avoided crossings among IFS are formed and the dynamics becomes nonadiabatic. In the vicinity of the avoided-crossing points (AC1/AC2) formed by the IFS ( $\alpha, m$ ) and ( $\alpha', m'$ ), the population transfer can be described by a Landau-Zener transfer matrix in Eq. (2). The parameters related to the details of the avoided crossing are defined as

$$\begin{aligned} \delta_{\alpha m \alpha' m'} &= \frac{\Delta_{\alpha m \alpha' m'}^2}{4}, \\ P_{\alpha m \alpha' m'} &= \exp(-2\pi \delta_{\alpha m \alpha' m'}), \\ \phi_{\alpha m \alpha' m'}^s &= \frac{\pi}{4} + \delta_{\alpha m \alpha' m'} \ln(\delta_{\alpha m \alpha' m'} - 1) \\ &\quad + \arg \Gamma(1 - i\delta_{\alpha m \alpha' m'}). \end{aligned} \quad (\text{A14})$$

Here,  $\Delta_{\alpha m \alpha' m'}$  and  $v_{\alpha m \alpha' m'}$  are the gap and passing speed of the avoided crossings between IFS ( $\alpha, m$ ) and ( $\alpha', m'$ ).  $\phi^s$  denotes the Stokes phase, and  $\Gamma(z)$  is the gamma function. The original Landau-Zener transfer matrix approach requires time-independent coupling. Its generalization to time-periodic driving [68,81] has extended the applicability to the IFS problem [58].

In Fig. 5(e), the dynamics is separated into  $\text{I} \rightarrow \text{II} \rightarrow \text{I}$  based on instantaneous quasienergies. All the relevant avoided crossing points are formed by the IFS in the same Floquet Brillouin zone ( $m_0 = \text{BZ5}$  as defined in

Fig. 1). The time evolution of  $\mathbf{c}_{m_0}(t)$  vectors is then calculated as

$$\mathbf{c}_{m_0}(t_r = t_{\text{fin}}) = [\hat{\mathbf{U}}_{m_0}(t_{\text{fin}}, t_{\text{AC2}}) \hat{\mathbf{T}}_{\text{IP1}, m_0, \text{TL}, m_0}^{\text{AC2}} \hat{\mathbf{U}}_{m_0}(t_{\text{AC2}}, t_{\text{AC1}}) \times \hat{\mathbf{T}}_{\text{IP1}, m_0, \text{TL}, m_0}^{\text{AC1}} \hat{\mathbf{U}}_{m_0}(t_{\text{AC1}}, t_{\text{ini}})] \mathbf{c}_{m_0}(t_r = t_{\text{ini}}),$$

where  $t_{\text{ini}}$  and  $t_{\text{fin}}$  denote the starting and the ending time of the pump + probe pulse. At  $t = t_{\text{ini}}$ , only SS is occupied, so we set  $c_{\text{IFS1}, m_0}(t_r = t_{\text{ini}}) = 1/\sqrt{N_f}$ . The decoherence effect is not considered in our IFS simulation due to the reported long decoherence time [46], which would give an effective energy broadening. Therefore we employ an effective energy Gaussian smearing in the time delay and  $E_{\text{final}}$  scan in Figs. 5(d)–5(i), to generate a similar broadening as observed in the experiment.

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