

Time-resolved ARPES in pumped excitonic systems: Floquet physics induced by excitonic fields

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We develop a theoretical framework based on the dynamical projective operatorial approach (DPOA) to study the time-resolved and angle-resolved photoemission spectroscopy (TR-ARPES) of pumped excitonic systems. Including Coulomb electron-electron interactions at the Hartree-Fock level, our formalism captures the formation of excitonic bound states under the application of pump pulses. Considering a prototypical two-dimensional two-band semiconductor, we analyze the equilibrium phase diagram, which shows the expected transition from a semiconducting to an excitonic-insulator phase as the Coulomb interaction strength or its range increases. Out of equilibrium, we find that when the pump frequency is resonant with an excitonic mode, coherent oscillations of the excitonic order parameter persist after the pump pulse subsides, and they give rise to clear Floquet sidebands in the TR-ARPES spectrum. These exciton-field-induced sidebands are distinct from those originating from the pump laser field. We also identify band-resonance-induced sidebands arising from residual coherences at momenta where the band gap is resonant with the pump frequency. Finally, we analyze the local Coulomb interaction limit. Our results corroborate recent experimental observations of exciton-field-induced Floquet-like sidebands and establish DPOA as an efficient and accurate method for simulating ultrafast phenomena in interacting electron systems.

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I. INTRODUCTION

The development of ultrafast pump-probe spectroscopy has opened a new frontier in exploring the dynamics of condensed-matter systems on femtosecond and even sub-femtosecond time scales [1–9]. By monitoring the temporal evolution of photoexcited carriers upon perturbation by ultrashort and intense electromagnetic pulses, these techniques provide direct access to the microscopic mechanisms governing electronic, spin, and lattice degrees of freedom out of equilibrium [10–14]. From a technological standpoint, such insight is essential for advancing next-generation ultrafast optoelectronic and spintronic devices. From a fundamental perspective, it enables probing pulse-induced symmetry breaking, coherence, and relaxation processes in real time [10,12,13]. One avenue to probe the response of a solid requires that the system is excited by an intense pump pulse, typically in the infrared regime with a duration ranging from a few to hundreds of femtoseconds, and subsequently analyzed using a positively or negatively delayed probe pulse. This analysis can measure either the transient change in optical properties [7–11,15–24] or the time-resolved angle-resolved photoemission spectroscopy (TR-ARPES) signal [14,25–32].

Angle-resolved photoemission spectroscopy probes the electronic band structure of materials by analyzing the energy and momentum of photoejected electrons [33–39]. In

pump-probe setups, TR-ARPES extends this capability to out-of-equilibrium systems by measuring the signal as a function of the pump-probe delay [39–43]. This technique reveals dynamical processes [39] of fundamental importance for understanding and engineering materials, providing access to bands above the Fermi energy beyond what equilibrium measurements can achieve [44–46]. TR-ARPES can also probe the dressing of bands and the emergence of pump-induced Floquet sidebands [29,47], as well as complex effects such as the perturbation of ordered states [14,25,26] and the dynamical excitation of collective modes [28,48,49].

Understanding these phenomena requires theoretical modeling and numerical simulation. The standard approach, time-dependent density functional theory (TD-DFT) [21,24,50–56], is computationally expensive and often limits physical insight [57]. Model-Hamiltonian approaches, which typically use parameters derived from equilibrium DFT calculations [58], offer a more flexible alternative, allowing controlled investigation of individual terms and their interplay, but they often suffer from oversimplification [57]. To address these challenges, we recently developed the dynamical projective operatorial approach (DPOA) [7,59–62], an efficient operator-based formalism for studying ultrafast phenomena in realistic multiband systems. DPOA enables real-time evolution of composite operators [63] under intense pump fields while maintaining access to all microscopic observables, including excitation populations [7,59,64,65] and multitime response functions such as TR-ARPES [59] and transient optical properties [9,61,66].

On the other hand, the pursuit of nontrivial phases of matter through optical driving has been a persistent objective

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in condensed-matter physics [67–69]. The oscillatory nature of the pump field itself provides a natural connection to the concept of Floquet engineering, where a time-periodic external drive is used to reshape the electronic band structure of a material [70–72]. By hybridizing electronic states with the photons of an intense laser field, one can create Floquet-Bloch states, leading to the emergence of novel phases of matter such as Floquet topological insulators or Weyl semimetals [29,73–75]. In essence, pump-probe experiments can be viewed as a realization of Floquet physics in a pulsed, rather than continuous-wave, regime, where the finite duration of the pump pulse introduces additional temporal structure while preserving the fundamental mechanism of light-induced band hybridization [59]. Despite the immense theoretical potential, the experimental realization of Floquet physics in solids faces significant hurdles; consequently, the observation of clear Floquet gaps and band hybridization has remained limited to a few specific systems and conditions [76,77].

Recent proposals have turned to bosonic excitations beyond photons as a means of generating time-periodic driving. Examples include phonon-driven Floquet matter [77], Floquet superconductivity [78], and exciton-driven Floquet physics [79,80]. In particular, the Coulomb interaction in low-dimensional semiconductors leads to the formation of excitons—bound states of an electron and a hole—with noticeable binding energies and transition dipole moments, which can provide a large internal field. In a recent work, Pareek *et al.* [81] pursued this direction. In a monolayer semiconductor, an exciton creates an oscillation of the self-energy: the excitonic field. Such a field can drive Floquet-like hybridization. Using TR-ARPES, they directly observed the modification of the electronic bands in a monolayer transition-metal dichalcogenide. Upon resonant excitation of an excitonic mode, the valence band develops a clear sideband, as well as the so called “Mexican-hat” dispersion shape.

Several theoretical studies of TR-ARPES signals in pumped excitonic systems have been performed [82–86]. A recently developed approach is the *ab initio* time-dependent GW (TD-aGW) approach [79,81]. In this approach, the dynamical equations of the single-particle density matrix are solved and then used to compute the two-time Green’s functions, which in turn enter the calculation of the TR-ARPES response. Such an approach is indeed quite resource-consuming, as it involves not only solving the dynamical equations but also computing the double integrals required for the TR-ARPES signal [87]. As we will demonstrate in this work, one can calculate the TR-ARPES signal of pumped excitonic systems using DPOA, which has a noticeably low computational cost.

Along these lines, in this work we extend the DPOA framework to investigate pumped interacting systems. We develop a theoretical description that incorporates Coulomb electron-electron interaction at the Hartree-Fock (HF) level, allowing for the formation of excitonic bound states both in equilibrium and under the influence of a pump pulse. We compute the transient electronic structure and analyze the emergence of Floquet sidebands in the TR-ARPES spectrum driven by the excitonic field. Our approach captures the essential physics of exciton formation and its dynamical evolution out of equilibrium. We find that when the pump frequency

matches an excitonic resonance, coherent oscillations of the excitonic order parameter persist after the pump pulse and give rise to Floquet sidebands in the TR-ARPES spectrum, which confirms the findings of Ref. [81]. These Floquet sidebands are distinct from those originating from light field, and provide a direct fingerprint of excitonic coherence. Moreover, our formalism allows for a detailed analysis of the pump-induced features in the presence of excitonic modes and, more generally, Coulomb electron-electron interaction. Our results demonstrate that TR-ARPES can serve as a powerful tool to detect and characterize excitonic dynamics in pumped semiconductors, and they establish DPOA as an efficient and accurate method for simulating ultrafast phenomena in interacting electronic systems.

The paper is organized as follows. In Sec. II, we present the theoretical formulation of the Coulomb electron-electron interaction Hamiltonian, its HF treatment, the time evolution under optical pumping, and the computation of the TR-ARPES signal within the DPOA framework. In Sec. III, we apply our formalism to a prototypical two-dimensional two-band (valence-conduction) semiconductor to investigate the two equilibrium phases (the semiconducting and the excitonic-insulator phases) and the out-of-equilibrium TR-ARPES response for both of them. We mainly consider a long-range Coulomb electron-electron interaction, but we will also consider the special case of the local Coulomb electron-electron interaction at the end of the same section. Finally, Sec. IV, we summarize our conclusions.

II. THEORY

A. Coulomb electron–electron interaction Hamiltonian

The Hamiltonian governing a noninteracting lattice system can be written as [59]

$$H_0 = \sum_{\mathbf{k}} c^\dagger(\mathbf{k}) \cdot \Xi^0(\mathbf{k}) \cdot c(\mathbf{k}), \quad (1)$$

where $c(\mathbf{k})$ is a vector whose components are the annihilation operators, $c_l(\mathbf{k})$, for an electron with the set of quantum numbers (such as band, spin, etc.) l and with momentum $\mathbf{k}$, $\cdot$ is used for matrix multiplication in such a space of quantum numbers (it will also be used as scalar contraction in Cartesian space), and $\Xi^0(\mathbf{k})$ is obtained by diagonalizing the hopping matrix expressed in the basis of localized Wannier functions, $\tilde{T}_{\mathbf{k}}$, via the unitary transformation given by the matrix $\Omega_{\mathbf{k}}$,

$$\Xi^0(\mathbf{k}) = \Omega_{\mathbf{k}}^\dagger \cdot \tilde{T}_{\mathbf{k}} \cdot \Omega_{\mathbf{k}}, \quad (2)$$

where $\Xi_{l,l'}^0(\mathbf{k}) = \delta_{l,l'} \varepsilon_l^0(\mathbf{k})$, in which $\delta_{l,l'}$ is the Kronecker delta and $\varepsilon_l^0(\mathbf{k})$ represent the noninteracting band energies.

The Coulomb electron-electron interaction is taken into account through the following density-density term:

$$H_V = \frac{1}{2} \sum_{\mathbf{i}, \mathbf{i}'} \sum_{l, l'} V_{l, l'}(\mathbf{i} - \mathbf{i}') n_l(\mathbf{i}) n_{l'}(\mathbf{i}'), \quad (3)$$

where $V_{l, l'}(\mathbf{i} - \mathbf{i}')$ is the interaction potential matrix element, $\mathbf{i}$ and $\mathbf{i}'$ run over lattice sites, and $n_l(\mathbf{i}) = c_l^\dagger(\mathbf{i}) c_l(\mathbf{i})$. The real-space operators are linked to their momentum-space counterparts through $c_l(\mathbf{i}) = \frac{1}{\sqrt{M}} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}_i} c_l(\mathbf{k})$, with M the total number of lattice sites and $\mathbf{R}_i$ the position of site $\mathbf{i}$. A

direct Fourier transformation yields

$$H_V = \frac{1}{2M} \sum_{\mathbf{k}} \sum_{l,l'} V_{l,l'}(\mathbf{k}) \times \sum_{\mathbf{q},\mathbf{q}'} c_l^\dagger(\mathbf{q} - \mathbf{k}) c_l(\mathbf{q}) c_{l'}^\dagger(\mathbf{q}' + \mathbf{k}) c_{l'}(\mathbf{q}'), \quad (4)$$

in which we have introduced the Fourier transform of the interaction potential matrix element,

$$V_{l,l'}(\mathbf{k}) = \sum_{\mathbf{i}} V_{l,l'}(\mathbf{i}) e^{-i\mathbf{k} \cdot \mathbf{R}_i}. \quad (5)$$

B. Hartree-Fock approximation

We treat the Coulomb interaction at the mean-field level by employing the HF decoupling. Moreover, (i) we assume that the equilibrium expectation values conserve crystal momentum, i.e., $\langle c_l^\dagger(\mathbf{q}) c_{l'}(\mathbf{q}') \rangle \propto \delta_{\mathbf{q},\mathbf{q}'}$, and (ii) we remove the contributions that would be canceled by the ionic positively charged background [specifically, the terms proportional to $V_{l,l'}(\mathbf{k} = \mathbf{0})$]. After these steps, the HF-decoupled Coulomb interaction Hamiltonian acquires the form

$$H_V \simeq H_{\text{HF}} = \sum_{\mathbf{k}} c^\dagger(\mathbf{k}) \cdot \Xi^{\text{HF}}(\mathbf{k}) \cdot c(\mathbf{k}), \quad (6)$$

where

$$\Xi_{ll'}^{\text{HF}}(\mathbf{k}) = -\frac{1}{M} \sum_{\mathbf{q}} V_{ll'}(\mathbf{k} - \mathbf{q}) \rho_{ll'}(\mathbf{q}), \quad (7)$$

in which we have assumed $V_{ll'}(-\mathbf{k}) = V_{ll'}(\mathbf{k})$.

Here $\rho_{ll'}(\mathbf{k}) = \langle c_l^\dagger(\mathbf{k}) c_l(\mathbf{k}) \rangle$ is the single-particle density matrix (SPDM). It is worth noting that although the original Coulomb interaction H_V is repulsive, the subtraction of the background term makes the effective HF Coulomb interaction attractive, a crucial feature for the formation of excitonic bound states. Note that $\Xi^{\text{HF}}(\mathbf{k})$ is referred to as the HF self-energy.

Because the HF Hamiltonian depends on the SPDM, a self-consistent calculation is necessary to determine the equilibrium properties. The iterative procedure adopted in this manuscript for such a self-consistent loop is described in detail in Appendix A.

C. Time evolution

Once the self-consistent equilibrium solution has been obtained, we examine the system's response to a pump pulse. The coupling to the electric field is introduced within the standard minimal coupling framework, but now the Hamiltonian becomes time-dependent through both the pump field and the instantaneous HF potential. Consequently, the full Hamiltonian can be cast in the form

$$H(t) = \sum_{\mathbf{k}} c^\dagger(\mathbf{k}) \cdot \Xi(\mathbf{k}, t) \cdot c(\mathbf{k}), \quad (8)$$

with

$$\Xi(\mathbf{k}, t) = \Xi^0(\mathbf{k}) + \Xi^{\text{pu}}(\mathbf{k}, t) + \Xi^{\text{HF}}(\mathbf{k}, t), \quad (9)$$

where, following Refs. [59,61],

$$\Xi^0(\mathbf{k}) + \Xi^{\text{pu}}(\mathbf{k}, t) = T_k(t) + eE_{\text{pu}}(t) \cdot \mathbf{D}_k(t), \quad (10)$$

in which $\mathbf{D}_k$ corresponds to the dipole moment, and for operator $O \in \{T, \mathbf{D}\}$, the time-dependent matrix is obtained through a generalized Peierls substitution [59]:

$$O_{\mathbf{k}}(t) = \Omega_{\mathbf{k}}^\dagger \cdot \tilde{O}_{\mathbf{k} + \frac{e}{\hbar} \mathbf{A}_{\text{pu}}(t)} \cdot \Omega_{\mathbf{k}}, \quad (11)$$

where $\tilde{O}$ refers to these matrices in the localized Wannier basis, and $\mathbf{A}_{\text{pu}}(t)$ is the vector potential of the pump field, related to the pump electric field by $\mathbf{E}_{\text{pu}}(t) = -\partial_t \mathbf{A}_{\text{pu}}(t)$. We operate within the dipole approximation, assuming the pump wavelength is much larger than the lattice spacing.

The quadratic form of the time-dependent Hamiltonian, Eq. (8), suggests that the creation and annihilation operators are convenient choices for the eigenoperators within DPOA [59,62]. Therefore, in the Heisenberg picture we write $c(\mathbf{k}, t) = P(\mathbf{k}, t) \cdot c(\mathbf{k})$, where $c(\mathbf{k}) = c(\mathbf{k}, t_0)$ corresponds to the operators at an initial time $t_0 \rightarrow -\infty$, well before the pump is applied, i.e., in equilibrium. The evolution matrix $P(\mathbf{k}, t)$ satisfies the equation of motion [59]

$$i\hbar \partial_t P(\mathbf{k}, t) = \Xi(\mathbf{k}, t) \cdot P(\mathbf{k}, t), \quad (12)$$

subject to the initial condition $P(\mathbf{k}, t_0) = \mathbf{1}$. At each instant of time, $\Xi^{\text{HF}}(\mathbf{k}, t)$ depends on the instantaneous SPDM, $\rho(\mathbf{k}, t)$ [through Eq. (7)] by replacing $\rho(\mathbf{q}) \rightarrow \rho(\mathbf{q}, t)$, which itself is given by

$$\rho(\mathbf{k}, t) = P(\mathbf{k}, t) \cdot \rho^{\text{eq}}(\mathbf{k}) \cdot P^\dagger(\mathbf{k}, t), \quad (13)$$

where $\rho^{\text{eq}}(\mathbf{k}) = \rho(\mathbf{k}, t_0)$ is the SPDM obtained from the equilibrium self-consistent calculation (see Appendix A).

It is noteworthy that one must compute the dynamical version of Eq. (7) [by substituting $\rho_{ll'}(\mathbf{q}) \rightarrow \rho_{ll'}(\mathbf{q}, t)$] at each time step and for each point on the numerical momentum grid. As is well known, such direct calculations are extremely resource-consuming. To make the computation feasible, we employ a fast-Fourier-transform routine combined with the standard convolution approach, which significantly reduces the numerical cost. For the time evolution of the dynamical equations [in particular, Eq. (12)] we use a fourth-order Runge-Kutta approach.

It is worth noting that, in this work, the interaction matrix elements are defined in the band basis, while hopping and dipole matrices are given in the localized Wannier basis and then transformed into the band basis [Eq. (11)]. In such a case, to correctly compute the time-dependent HF self-energy, one has to smooth the phases of the transformation matrices, $\Omega_{\mathbf{k}}$, over the BZ in the numerical calculations. This is needed to assure that SPDMs at different crystal momenta have consistent phases [see Eq. (7)]. Instead, if the interaction matrix elements too are defined in the localized Wannier basis, such a phase smoothing is not required.

D. TR-ARPES signal

To compute the TR-ARPES signal, we employ the formalism of nonequilibrium Green's functions. We define the standard retarded and lesser Green's functions as

$$G_{l,l'}^R(\mathbf{k}, t, t') = -i\theta(t - t') \langle \{c_l(\mathbf{k}, t), c_{l'}^\dagger(\mathbf{k}, t')\} \rangle, \quad (14)$$

$$G_{l,l'}^<(\mathbf{k}, t, t') = i\langle c_{l'}^\dagger(\mathbf{k}, t') c_l(\mathbf{k}, t) \rangle. \quad (15)$$

Within DPOA, one can demonstrate that these Green's functions can be written as [59]

$$G^R(\mathbf{k}, t, t') = -i\theta(t - t')P(\mathbf{k}, t) \cdot P^\dagger(\mathbf{k}, t'), \quad (16)$$

$$G^<(\mathbf{k}, t, t') = iP(\mathbf{k}, t) \cdot \rho^{\text{eq}}(\mathbf{k}) \cdot P^\dagger(\mathbf{k}, t'). \quad (17)$$

The experimentally accessible quantity is the photocurrent, which we refer to as the electron TR-ARPES signal, or simply the TR-ARPES signal. It is defined by convolving the lesser Green's function with the probe pulse envelope [59,87]:

$$I^e(\mathbf{k}, \omega, t_{\text{pr}}) = \frac{\tau_{\text{pr}}}{\sqrt{8\pi \ln 2}} \int_{-\infty}^{+\infty} dt \int_{-\infty}^{+\infty} dt' S_{\text{pr}}(t - t_{\text{pr}}) \times S_{\text{pr}}(t' - t_{\text{pr}}) \text{Im}[e^{i\omega(t-t')} \text{Tr}[G^<(\mathbf{k}, t, t')]], \quad (18)$$

where $S_{\text{pr}}(t) = \frac{2\sqrt{\ln 2}}{\sqrt{\pi} \tau_{\text{pr}}} e^{-4 \ln 2 t^2 / \tau_{\text{pr}}^2}$ is the probe-pulse Gaussian envelope of full width at half maximum (FWHM) τ_{pr} , and t_{pr} is the probe delay. Note that this definition is a measure of the actual TR-ARPES signal, but it does not include the matrix elements coupling the free electronic states in vacuum and the states in the material [59]. Accordingly, our I^e has the dimensions of time, and the overall coefficients have been chosen such that integrating over ω gives the total number of particles at each crystal momentum. I^e reflects the occupied electronic states, but to map out the full band structure (or the transient bands), one may instead use the retarded Green's function, defining the quantity [59]

$$I^R(\mathbf{k}, \omega, t_{\text{pr}}) = -\frac{\tau_{\text{pr}}}{\sqrt{2\pi \ln 2}} \int_{-\infty}^{+\infty} dt \int_{-\infty}^{+\infty} dt' S_{\text{pr}}(t - t_{\text{pr}}) \times S_{\text{pr}}(t' - t_{\text{pr}}) \text{Im}[e^{i\omega(t-t')} \text{Tr}[G^R(\mathbf{k}, t, t')]]. \quad (19)$$

Following analogous steps to those presented in Ref. [59], these expressions can be recast in a compact form:

$$I^e(\mathbf{k}, \omega, t_{\text{pr}}) = \frac{\tau_{\text{pr}}}{\sqrt{8\pi \ln 2}} \text{Tr}[Q(\mathbf{k}, \omega, t_{\text{pr}}) \rho^{\text{eq}}(\mathbf{k}) Q^\dagger(\mathbf{k}, \omega, t_{\text{pr}})], \quad (20)$$

$$I^R(\mathbf{k}, \omega, t_{\text{pr}}) = \frac{\tau_{\text{pr}}}{\sqrt{8\pi \ln 2}} \text{Tr}[Q(\mathbf{k}, \omega, t_{\text{pr}}) Q^\dagger(\mathbf{k}, \omega, t_{\text{pr}})], \quad (21)$$

where we have introduced the auxiliary matrix

$$Q(\mathbf{k}, \omega, t_{\text{pr}}) = \int_{-\infty}^{+\infty} dt e^{i\omega t} P(\mathbf{k}, t) S_{\text{pr}}(t - t_{\text{pr}}). \quad (22)$$

This procedure avoids directly computing many double integrals and noticeably speeds up numerical calculations.

Experimentally, only the electron signal, I^e , is directly measurable. Nevertheless, it is useful to define theoretically a hole signal as

$$I^h(\mathbf{k}, \omega, t_{\text{pr}}) = I^R(\mathbf{k}, \omega, t_{\text{pr}}) - I^e(\mathbf{k}, \omega, t_{\text{pr}}), \quad (23)$$

which provides complementary information about the unoccupied states.

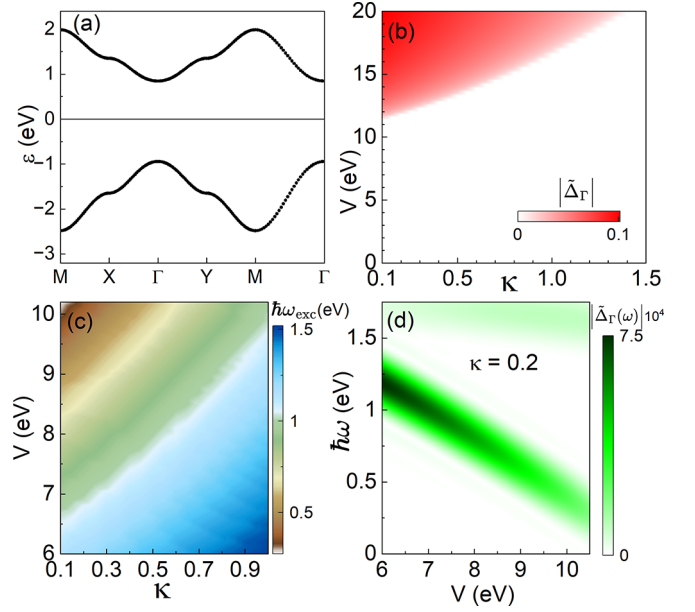


FIG. 1. Equilibrium properties of the two-band model. (a) Non-interacting band structure of the two-band model along a high-symmetry path in the BZ. (b) Equilibrium normalized off-diagonal HF self-energy at the Γ point, $\tilde{\Delta}_\Gamma$, as a function of the Coulomb interaction strength V and the screening factor κ . The boundary between the semiconducting phase ($\tilde{\Delta}_\Gamma = 0$) and the excitonic insulator phase ($\tilde{\Delta}_\Gamma \neq 0$) is visible. (c) Main equilibrium excitonic energy, $\hbar\omega_{\text{exc}}$, in the semiconducting phase as a function of V and κ . (d) Fourier-transformed excitonic field, $\tilde{\Delta}_\Gamma(\omega)$, as a function of V and $\hbar\omega$ for a fixed screening factor $\kappa = 0.2$, showing the main excitonic mode and a weaker higher-energy mode.

III. EXCITONIC DYNAMICS IN A PUMPED TWO-BAND SEMICONDUCTOR

A. System and equilibrium phases

In this section, we apply our formalism to a prototypical two-dimensional (2D) two-band (valence-conduction) model that mimics a semiconductor. The lattice constant is denoted by $a = 0.5$ nm, and for simplicity we consider spinless electrons. In the localized Wannier basis, the hopping matrix elements are specified as follows. The onsite energies are $\tilde{T}_{\mathbf{R}=0,1,1} = -1.65$ eV and $\tilde{T}_{\mathbf{R}=0,2,2} = 1.35$ eV for the two Wannier states indexed by 1 and 2. The nearest-neighbor diagonal hoppings are $\tilde{T}_{\mathbf{R}=\mathbf{a},1,1}^{\text{hop}} = 0.2$ eV and $\tilde{T}_{\mathbf{R}=\mathbf{a},2,2}^{\text{hop}} = -0.15$ eV, while the off-diagonal nearest-neighbor hopping is $\tilde{T}_{\mathbf{R}=\mathbf{a},2,1} = \tilde{T}_{\mathbf{R}=\mathbf{a},1,2} = -0.1$ eV. Here, $\tilde{T}_{\mathbf{R},\nu,\nu'}$ denotes the hopping matrix element between Wannier states ν and ν' centered on lattice sites separated by the vector $\mathbf{R}$, and $\mathbf{a} \in \{\pm a\hat{x}, \pm a\hat{y}\}$ runs over all nearest-neighbor displacement vectors. The Brillouin zone (BZ) is sampled by a uniform grid of $M = 64 \times 64$ $\mathbf{k}$ -points including the Γ point. Fourier transforming $\tilde{T}_{\mathbf{R}}$ yields the momentum-space hopping matrix $\tilde{T}_{\mathbf{k}}$ as described in Refs. [59,61]. All calculations are performed at zero temperature, and the chemical potential is set within the band gap. In Fig. 1(a), we show the noninteracting bands of this system in equilibrium along a high-symmetry path in the BZ.

If we keep the model perfectly centrosymmetric, in the semiconducting phase the excitons would remain *dark*,

because, for instance, the first-order light-matter coupling at Γ would vanish. To break this symmetry, and make the excitons *bright*, we introduce a local dipole moment: $\tilde{\mathbf{D}}_{\mathbf{R}=0,1,2} = \tilde{\mathbf{D}}_{\mathbf{R}=0,2,1} = D\hat{x}$ with $D = 0.1$ nm. It is worth noting that a similar effect could have been achieved also by applying a left-right asymmetry in the hopping matrix elements, or considering a model like SSH where the selection rules allow for the nonzero coupling elements at Γ .

The expression of the Coulomb electron-electron interaction potential, $V(\mathbf{k})$, has been obtained by solving the discrete Poisson equation on a three-dimensional (3D) lattice [88–91], embedding our 2D one. This latter resides at $z = 0$ within the embedding 3D lattice, and its actual expression for $V(\mathbf{k})$ has been computed integrating out k_z from the corresponding 3D expression [88]. Accordingly, we have

$$V(\mathbf{k}) = \frac{V}{\sqrt{(\tilde{k}^2 + \kappa^2 + 2)^2 - 4}}, \quad (24)$$

where $\tilde{k}^2 = 2(2 - \cos k_x a - \cos k_y a)$ and κ controls a Yukawa-like screening mimicking the effects on the dielectric function of a small, but finite, carrier mobility. Here, we have assumed that the 3D lattice is cubic ($a_x = a_y = a_z = a$) and that the system exhibits an isotropic dielectric function. This long-range interaction will be considered for most of the results we analyze. At the end of the results section, we will also consider the local interaction case and, therefore, a momentum-independent Coulomb electron-electron interaction potential.

We choose the Coulomb interaction to act only between the conduction and the valence bands (the CBs and the VBs, respectively), because our main focus in this study is the electron-hole bound states. Thus, the only nonzero matrix elements are $V_{v,c}(\mathbf{k}) = V_{c,v}(\mathbf{k}) = V(\mathbf{k})$, where v and c refer to the VB and the CB, respectively, and V is dubbed the Coulomb interaction strength. As such, the only relevant term in the HF self-energy is the off-diagonal one. To analyze the excitonic field, we consider the value of the off-diagonal HF self-energy divided by the Coulomb interaction strength V and denote it, in equilibrium, by

$$\tilde{\Delta}_{\mathbf{k}} = \frac{\Xi_{c,v}^{\text{HF}}(\mathbf{k})}{V}. \quad (25)$$

As a representative quantity that captures the excitonic order parameter at the center of the BZ, we focus mainly on $\tilde{\Delta}_{\Gamma} = \tilde{\Delta}_{\mathbf{k}=0}$. Out of equilibrium, this quantity becomes a function of time and we denote it by $\tilde{\Delta}_{\Gamma}(t)$. Performing the Fourier transform with respect to time, we obtain it as a function of the angular frequency, ω , and denote it by $\tilde{\Delta}_{\Gamma}(\omega)$.

To characterize the equilibrium phases, we study $\tilde{\Delta}_{\Gamma}$ in equilibrium. In Fig. 1(b), we show $\tilde{\Delta}_{\Gamma}$ as a function of the Coulomb interaction strength V and the screening factor κ . For each given κ , there is a critical value V below which $\tilde{\Delta}_{\Gamma} = 0$ and the system has no excitonic bound state in its ground state; we refer to this regime as the semiconducting phase. In this phase, the ground-state SPDM is the same as that of the noninteracting system, which makes the HF self-energy vanish, and hence the equilibrium bands also coincide with the noninteracting ones. Above this critical V , the equilibrium value of $\tilde{\Delta}_{\Gamma}$ becomes finite, indicating the spontaneous

formation of excitonic bound states in the ground state. We will refer to such a phase as the excitonic insulator phase. It is noteworthy that the critical V increases as κ increases. This is because a larger κ means a more localized (shorter-range) Coulomb interaction, and reducing the range of the interaction also decreases its effective strength, thus requiring a larger bare V to reach the excitonic instability.

B. Equilibrium excitonic frequency for the semiconducting phase

In the semiconducting phase, there is no excitonic bound state in the ground state. However, one may pump the system, and if the pump frequency is in a specific range, excitonic bound states form. Such frequency ranges determine what we call hereafter the excitonic frequencies. In such a scenario, after the application of the pump pulse, the excitonic self-energy oscillates at the excitonic frequency [79,81]. This excitonic frequency depends on the pump-pulse parameters (amplitude and duration) if the pump pulse is intense and long enough, but, for weak pump intensities, it is an intrinsic property of the system, to which we refer as the equilibrium excitonic frequency.

To determine the equilibrium excitonic frequency, we have devised the following protocol. We apply a very weak impulsive pump pulse with a Dirac-like δ function shape in time (i.e., a very narrow Gaussian), resulting in a wide frequency content. Then, we compute $\tilde{\Delta}_{\Gamma}(\omega)$ and find the frequency ω_{exc} for which $|\tilde{\Delta}_{\Gamma}(\omega_{\text{exc}})|$ is maximum among all positive $\hbar\omega$ smaller than the energy band gap at Γ . The quantity $\hbar\omega_{\text{exc}}$ is the main equilibrium excitonic energy of the system. It is worth noting that, in general, $|\tilde{\Delta}_{\Gamma}(\omega)|$ can have several local maxima, all of which would determine the equilibrium excitonic frequencies corresponding to different excitonic states.

To numerically implement such an impulsive pump pulse, we consider the following vector potential:

$$A_{\text{imp}}(t) = A_{0,\text{imp}} e^{-(4 \ln 2)t^2/\tau_{\text{imp}}^2}, \quad (26)$$

with $\tau_{\text{imp}} = 0.03$ fs and $A_{0,\text{imp}} = 0.05$ V fs/nm (the robustness of the results with respect to reasonable variations of these parameters has been verified). In Fig. 1(c), we report the main equilibrium excitonic energy, $\hbar\omega_{\text{exc}}$, as a function of V and κ for ranges that fall into the semiconducting phase. As it can be seen, by increasing the Coulomb interaction strength V , or by making it more long-range (decreasing κ), the main equilibrium excitonic energy decreases. This behavior occurs because a stronger or more long-range interaction leads to a more strongly bound electron-hole pair, which requires less energy to be excited. Approaching the critical Coulomb interaction parameters (the boundary of the semiconducting phase), the equilibrium excitonic energy approaches zero, meaning that one can have excitonic bound states with a negligibly small photon energy.

The full dependence of $|\tilde{\Delta}_{\Gamma}(\omega)|$ for the specific case of $\kappa = 0.2$ is shown in Fig. 1(d) as a function of V and $\hbar\omega$. It is clear that increasing V reduces the excitonic energy, consistently with the trend observed in panel (c). Moreover, we observe another weak excitonic mode at a higher energy, which corresponds to another excitonic state. The time interval for these Fourier transformations is from 2 to 32 fs.

Moreover, Hann windowing has been applied to all Fourier transformation of the manuscript to reduce noise.

C. Out-of-equilibrium: Semiconducting phase

To study the out-of-equilibrium dynamics, we apply a linearly polarized pump pulse with vector potential $A_{\text{pu}}(t) = A_0 e^{-t^2/\tau_{\text{pu}}^2} \sin(\omega_{\text{pu}} t)$, where

$$A_{\text{pu}}(t) = A_0 e^{-(4 \ln 2) t^2 / \tau_{\text{pu}}^2} \sin(\omega_{\text{pu}} t). \quad (27)$$

The pulse has a Gaussian envelope with full width at half maximum (FWHM) $\tau_{\text{pu}} = 10$ fs. Its amplitude is A_0 , and ω_{pu} is the central pump-pulse frequency, or simply, the pump-pulse frequency ($\hbar\omega_{\text{pu}}$ is dubbed the pump-pulse photon energy). For the TR-ARPES calculations, the FWHM of the probe pulse is set to $\tau_{\text{pr}} = 10$ fs, equal to that of the pump pulse.

We focus on the post-pump regime (time delay $t_{\text{pr}} = 40$ fs) for two fundamental reasons. First, from an experimental perspective, measuring at large positive delays is easier because it requires less effort to time-resolve the pump and probe envelopes, reducing the need for ultraprecise synchronization. Second, and more importantly, Floquet sidebands in the TR-ARPES spectrum require a time-periodic driving term in $\Xi(\mathbf{k}, t)$ [see Eq. (9)]. After the pump pulse has subsided, the pump contribution to the self-energy vanishes [$\Xi^{\text{pu}}(\mathbf{k}, t) = 0$] and the time dependence in the Hamiltonian cannot be attributed to the light field: it must originate from the only other time-dependent term, namely the HF self-energy, $\Xi^{\text{HF}}(\mathbf{k}, t)$, which indicates an oscillating excitonic order parameter that persists after the pump is gone. Consequently, the Floquet sidebands observed in the post-pump TR-ARPES spectrum are uniquely due to the excitonic field, providing a clean signature of coherent excitonic dynamics [81].

Figure 2 presents the post-pump TR-ARPES signal for the semiconducting phase, with parameters $V = 6$ eV and $\kappa = 0.2$. Panels (a), (c), and (e) show the post-pump TR-ARPES signal at the Γ point ($\mathbf{k} = \mathbf{0}$) as a function of the pump frequency $\hbar\omega_{\text{pu}}$ for different pump intensities, identified by the amplitudes $A_0 = 0.1, 0.25$, and 0.5 V fs/nm, respectively. In these panels, the horizontal dashed lines indicate the CB and VB energies at Γ . Panels (b), (d), and (f) show the amplitude of the Fourier transform of the normalized off-diagonal element of the Hartree-Fock self-energy, $|\tilde{\Delta}_{\Gamma}(\omega)|$, as a function of ω_{pu} and ω , computed with the same pump-pulse parameters as their companion panels (panels in the same row correspond to identical pump-pulse parameters). The range of pump-pulse frequencies shown in these panels is narrower than in the TR-ARPES panels because we are primarily interested in the excitonic resonance frequencies that lie inside the band gap. In these panels, the horizontal gray line indicates the equilibrium excitonic energy, computed in Fig. 1(c). In all panels of this figure, the vertical short-dashed lines indicate the pump-pulse frequency that is in resonance with the equilibrium band gap at Γ : $\hbar\omega_{\text{pu}} = \varepsilon_c(\Gamma) - \varepsilon_v(\Gamma)$. The time interval for all of the post-pump Fourier transformations of this manuscript is from 25 to 55 fs, and the Hann windowing is used to reduce the noise.

We now analyze each pair of panels (row) in detail.

Panels (a) and (b): Weak pump intensity ($A_0 = 0.1$ V fs/nm). In Fig. 2(a), a small exciton-field-induced Floquet sideband

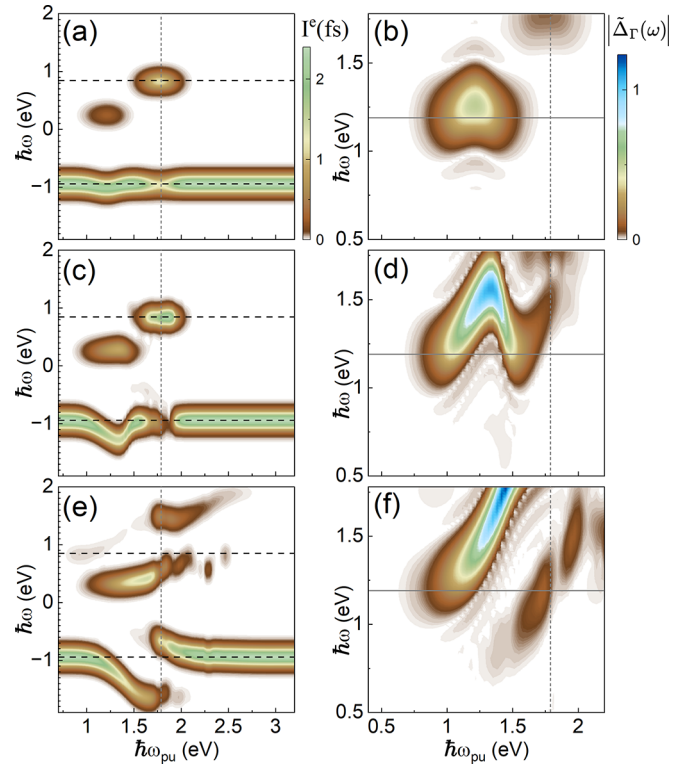


FIG. 2. Results for $V = 6$ eV, semiconducting phase. (a), (c), (e) Post-pump TR-ARPES signal at the Γ point as a function of pump frequency for pump amplitudes $A_0 = 0.1, 0.25$, and 0.5 V fs/nm, respectively. Horizontal dashed lines mark CB and VB energies at Γ ; vertical short-dashed lines indicate the pump frequency resonant with the equilibrium band gap at Γ . (b), (d), (f) Corresponding amplitude $|\tilde{\Delta}_{\Gamma}(\omega)|$ as a function of ω_{pu} and ω . Horizontal gray lines indicate the equilibrium excitonic energy from Fig. 1(c), and vertical short-dashed lines are similar to the other panels.

appears, reaching its maximum intensity for $\hbar\omega_{\text{pu}} = 1.22$ eV. This value is very close to 1.19 eV, the corresponding (same V and same κ) equilibrium excitonic frequency obtained from Fig. 1(c). For pump-pulse frequencies near resonance with the equilibrium band gap, we observe residual electron populations in the CB, that is, some electrons are excited across the gap and remain there after the pump pulse has subsided. For pump-pulse frequencies higher than the band-gap resonance, no noticeable feature at the Γ point emerges. Turning to Fig. 2(b), $|\tilde{\Delta}_{\Gamma}(\omega)|$ exhibits its maximum at $\hbar\omega = 1.26$ eV, which is close to the equilibrium excitonic frequency, and the corresponding pump-pulse frequency is also almost in resonance with it, as indicated in the previous panel. This demonstrates that at this weak pump intensity, the excitonic frequency is not noticeably shifted from its equilibrium value. Lowering the pump intensity further causes the post-pump excitonic frequency and the equilibrium one to match (not shown).

Panels (c) and (d): Intermediate pump intensity ($A_0 = 0.25$ V fs/nm). In Fig. 2(c), we see that the pump-pulse frequency that yields the strongest exciton-field-induced Floquet sideband has increased to $\hbar\omega_{\text{pu}} = 1.33$ eV. Moreover, at this excitonic resonance, the VB is shifted downward in energy, a manifestation of the so-called Mexican-hat effect

[81,92–96]. This effect arises from the coupling between the excitonic order parameter and the single-particle band structure, which renormalizes the VB dispersion near Γ . Importantly, the excitonic energy—defined as the energy difference between the exciton-field-induced Floquet sideband and the post-pump VB energy (the local maxima of the corresponding TR-ARPES signal)—exceeds the pump-pulse photon energy $\hbar\omega_{\text{pu}}$ more than in the previous case ($A_0 = 0.1$ V fs/nm). This observation is clarified in Fig. 2(d): the maximum of $|\tilde{\Delta}_\Gamma(\omega)|$ occurs for $\hbar\omega_{\text{pu}} = 1.33$ eV but at $\hbar\omega = 1.56$ eV. Note that the latter value is larger than 1.19 eV, the equilibrium excitonic frequency computed in Fig. 1(c). This shift demonstrates the effect of an intense pump pulse on the excitonic frequency. Because the system is interacting and the Hamiltonian depends on the electronic state through the HF self-energy, the pump-induced changes in the electron distribution modify the effective attraction between electrons and holes, thereby altering the excitonic resonance condition.

Panels (e) and (f): Strong pump intensity ($A_0 = 0.5$ V fs/nm). Increasing the intensity further magnifies all of the effects observed in the previous panels. The pump-pulse frequency that gives the strongest exciton-field-induced Floquet sideband increases further to $\hbar\omega_{\text{pu}} = 1.45$ eV. The Mexican-hat effect becomes noticeably stronger, as evidenced by the more pronounced downward shift of the VB. This downward shift and the increase of the exciton frequency goes on even if we increase $\hbar\omega_{\text{pu}}$ to be almost in resonance with the equilibrium band-gap energy. Interestingly, this has a counterintuitive effect: for this very intense pump pulse and for a pump photon energy in resonance with the equilibrium band gap, the residual excitation is smaller than in the case of $A_0 = 0.25$ V fs/nm. This happens because, by the application of the pump pulse, the Mexican hat effect, which now occurs also at this pump frequency, dynamically increases the effective band gap and pushes it out of resonance. Panel (f) shows that the maximum of $|\tilde{\Delta}_\Gamma(\omega)|$ occurs for $\hbar\omega_{\text{pu}} = 1.45$ eV but at a value of $\hbar\omega$ that reaches almost the band-gap energy, representing a very large shift between the two and with respect to the equilibrium excitonic frequency.

To better clarify the behavior of the exciton-field oscillations, we consider the high-intensity case ($A_0 = 0.5$ V fs/nm) and set the pump frequency to $\hbar\omega_{\text{pu}} = 1.35$ eV, which, as identified in Fig. 2(e), is in the frequency range that is capable of yielding an exciton-field-induced Floquet sideband, while it is far from the band-gap resonance. In Fig. 3(a), we plot $\tilde{\Delta}_\Gamma(t)$ as a function of time. The vertical dashed lines mark $t = \pm\tau_{\text{pu}} = \pm 10$ fs, indicating the temporal region where the pump pulse envelope is appreciably nonzero, thereby clarifying when the pump pulse is turned on and off. Both the real and imaginary parts of $\tilde{\Delta}_\Gamma(t)$ oscillate after the application of the pump pulse, while before its application they are zero, as the system is initially in the semiconducting phase with no preexisting excitonic order. To reveal the frequency content of these post-pump oscillations, in Fig. 3(b) we plot the post-pump amplitude $|\tilde{\Delta}_\Gamma(\omega)|$ as a function of frequency. We clearly see the main peak, which determines the dominant exciton-field oscillation frequency.

In Fig. 4, we plot the $\mathbf{k}$ -resolved post-pump TR-ARPES signal along the high-symmetry path in the BZ for selected pump-pulse frequencies, all computed with the large pump

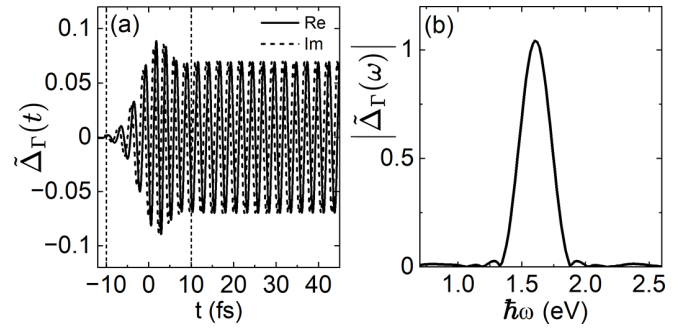


FIG. 3. Results for $V = 6$ eV, semiconducting phase. (a) Time evolution of the real and imaginary parts of $\tilde{\Delta}_\Gamma(t)$ for the high-intensity case ($A_0 = 0.5$ V fs/nm) with pump frequency $\hbar\omega_{\text{pu}} = 1.35$ eV. Vertical dashed lines indicate $t = \pm\tau_{\text{pu}} = \pm 10$ fs, marking the pump-pulse duration. (b) Corresponding post-pump amplitude $|\tilde{\Delta}_\Gamma(\omega)|$ as a function of frequency, showing the main exciton-field oscillation peak.

amplitude $A_0 = 0.5$ V fs/nm. These cuts correspond to vertical slices through the color plots shown in Fig. 2(e), allowing us to examine the momentum-resolved structure of the post-pump spectral features.

Panel (a): $\hbar\omega_{\text{pu}} = 1.35$ eV. Here, we observe a sideband induced by the excitonic field that appears almost everywhere parallel to the VB, which is a characteristic signature of a Floquet sideband. However, this sideband is stronger near Γ

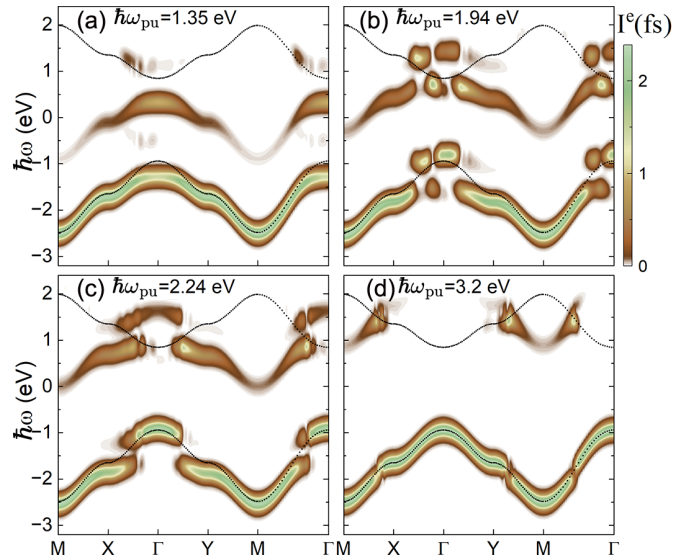


FIG. 4. Results for $V = 6$ eV, semiconducting phase. $\mathbf{k}$ -resolved post-pump TR-ARPES signal along the high-symmetry path in the BZ for the large pump amplitude $A_0 = 0.5$ V fs/nm, corresponding to vertical cuts in Fig. 2(e). (a) $\hbar\omega_{\text{pu}} = 1.35$ eV, showing a strong exciton-field-induced Floquet sideband parallel to the VB. (b) $\hbar\omega_{\text{pu}} = 1.94$ eV, slightly above the band gap at Γ , showing band-resonance-induced sidebands and level splitting near Γ . (c) $\hbar\omega_{\text{pu}} = 2.24$ eV, with resonant $\mathbf{k}$ -points farther away from Γ , yielding weaker band-resonance-induced sidebands. (d) $\hbar\omega_{\text{pu}} = 3.2$ eV, with resonant $\mathbf{k}$ -points very far from Γ , producing very weak band-resonance-induced sidebands. The equilibrium band energies are shown by dotted lines.

and becomes significantly weaker away from the zone center. This momentum dependence has two origins. First, the HF self-energy itself turns out to be larger in magnitude at Γ than at other $\mathbf{k}$ -points, leading to a stronger effective driving. Second, and more importantly, the oscillation frequency of the HF self-energy at Γ is close to the local equilibrium band-gap energy at that $\mathbf{k}$ -point; this small-detuning condition enhances the sideband intensity. The latter factor turns out to be much more important; we will clarify this point further when we analyze the local Coulomb interaction case. We note that there are also small residual electronic excitations at $\mathbf{k}$ -points that satisfy a two-photon resonance condition with the pump pulse.

Panel (b): $\hbar\omega_{\text{pu}} = 1.94 \text{ eV}$. This pump frequency is slightly above the direct band-gap resonance at Γ . In this case, we observe residual excitations at the resonant $\mathbf{k}$ -points, which give rise to some Floquet sidebands that have a different origin from those of the previous panel. For pump-pulse photon energies larger than the band gap, and with an intense enough pump pulse, we observe a Floquet sideband located approximately at an energy corresponding to $\hbar\omega_{\text{pu}}$ above the main band energies. Whenever the pump-photon energy exceeds the energy gap, there exist some $\mathbf{k}$ -points in the BZ whose band gap is in resonance with the pump frequency. Consequently, a residual excitation remains in the system after the pump pulse has subsided. These residual excitations possess coherence terms that oscillate with their own energy gap, which is equal to $\hbar\omega_{\text{pu}}$. These coherences contribute to the HF self-energy and provide the oscillatory Hamiltonian term which is necessary for the emergence of Floquet sidebands. As such, these sidebands are not strictly induced by excitons; we refer to them as band-resonance-induced sidebands to distinguish them from the genuine exciton-field-induced Floquet features.

It should be noted that this pump-photon energy also excites some excitonic coherences, and therefore the induced sidebands have a mixed origin, partly from the excitonic field and partly from the band-gap resonance. This hybrid nature can be verified by a detailed analysis of $\tilde{\Delta}_{\Gamma}(\omega)$ for this case (not shown). Finally, the resonant driving condition causes a level splitting near Γ , an effect that was also visible in Fig. 2(e).

Panel (c): $\hbar\omega_{\text{pu}} = 2.24 \text{ eV}$. Here, the resonant $\mathbf{k}$ -points are located farther away from Γ than in the previous case. The band-resonance-induced Floquet sidebands emerge clearly, and as explained earlier, they appear at an energy approximately $\hbar\omega_{\text{pu}}$ above the VB. These sidebands are weaker than those in the preceding panel.

Panel (d): $\hbar\omega_{\text{pu}} = 3.2 \text{ eV}$. At this high pump frequency, the resonant $\mathbf{k}$ -points are very far from Γ , and the corresponding band-resonance-induced Floquet sidebands are very weak. When the pump is tuned far away from the excitonic resonance, the induced sidebands become progressively weaker and eventually fade away.

D. Out-of-equilibrium: Excitonic-insulator phase

We now turn to the excitonic-insulator phase. We choose parameters $V = 15 \text{ eV}$ and $\kappa = 0.2$, which place the system well inside the excitonic-insulator phase as established in Fig. 1(b).

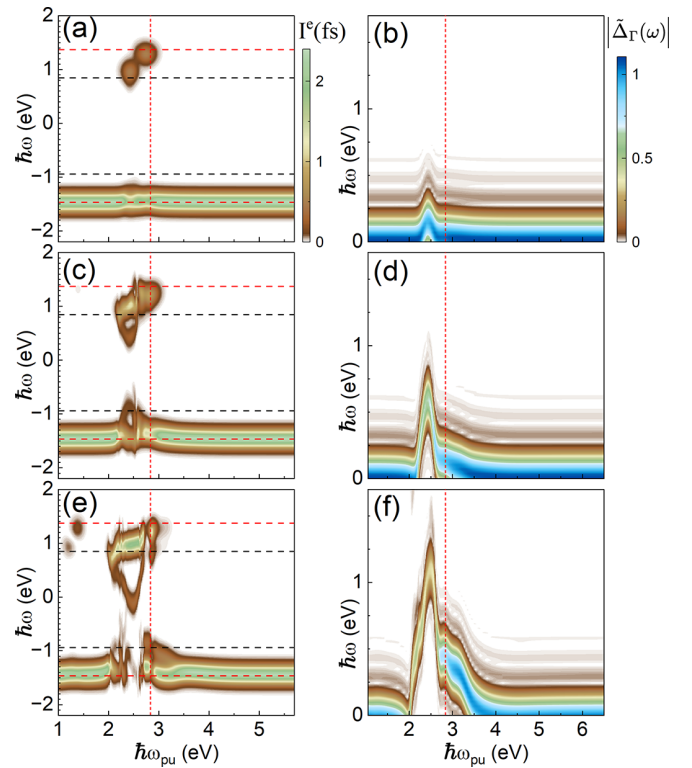


FIG. 5. Results for $V = 15 \text{ eV}$, excitonic-insulator phase. (a), (c), (e) Post-pump TR-ARPES signal at the Γ point as a function of pump frequency $\hbar\omega_{\text{pu}}$ for the excitonic-insulator phase ($V = 15 \text{ eV}$, $\kappa = 0.2$) and pump amplitudes $A_0 = 0.1, 0.25$, and 0.5 V fs/nm , respectively. Black horizontal dashed lines mark the noninteracting CB and VB energies at Γ ; red horizontal dashed lines mark the *interacting* band energies. (b), (d), (f) Corresponding amplitude $|\tilde{\Delta}_{\Gamma}(\omega)|$ as a function of ω_{pu} , and ω . Vertical red dashed lines indicate the pump frequency resonant with the equilibrium band gap of the interacting system.

Figure 5 is the counterpart of Fig. 2 for the excitonic-insulator phase. As in the semiconducting case, panels (a), (c), and (e) show the post-pump TR-ARPES signal at the Γ point as a function of the pump frequency $\hbar\omega_{\text{pu}}$ for three different pump intensities $A_0 = 0.1, 0.25$, and 0.5 V fs/nm , respectively. Panels (b), (d), and (f) show the corresponding amplitude $|\tilde{\Delta}_{\Gamma}(\omega)|$ as a function of ω_{pu} , and ω , computed with the same pump-pulse parameters as their companion panels. In panels (a), (c), and (e), the black horizontal dashed lines indicate the CB and VB energies of the noninteracting system, while the red horizontal dashed lines indicate those of the interacting system. Because of exciton formation in equilibrium, the band energies must be computed by diagonalizing the full interacting Hamiltonian, and these red lines mark the *interacting* band positions. The vertical red dashed line indicates the equilibrium resonance condition using the *interacting* bands.

We now analyze each pair of panels in detail.

Panels (a) and (b): Weak pump intensity ($A_0 = 0.1 \text{ V fs/nm}$). Well before reaching the resonance condition, no noticeable features appear in the TR-ARPES signal. As the pump frequency approaches the resonance, we observe some residual electrons appearing in the CB. Just before hitting the

resonance with the interacting band gap, which results in its own residual excitations, some electrons are excited to the noninteracting equilibrium CB. This happens because the pump photon energy gets in resonance with the energy gap between the interacting VB and the noninteracting CB. This indicates that the pump pulse has dissociated some of the equilibrium excitons, causing the bands to shift slightly and move toward the noninteracting band positions. This dissociation of the equilibrium excitons induces some low-energy coherent oscillations in the self-energy, which are not strong enough to induce notable exciton-field-induced Floquet sidebands. Panel (b) confirms this observation: there is only a very low-frequency component in $|\tilde{\Delta}_\Gamma(\omega)|$ for pump-pulse frequencies near the resonance at Γ , indicating that no noticeable coherent high-frequency excitonic oscillation persists after the pump.

Panels (c) and (d): Intermediate pump intensity ($A_0 = 0.25$ Vfs/nm). Again, far away from the resonance, no features are visible. Near the resonance to the energy gap between the interacting VB and the noninteracting CB, we obtain residual electrons in the CB, with a larger population than in the weak-intensity case. An exciton-field-induced sideband appears, running parallel to both the VB and the occupied portions of the CB. This effect emerges because of the coherent oscillations induced by dissociating the preexisting excitons. Panel (d) confirms that for pump-pulse photon energies near the resonance, there exists a well-defined excitonic field frequency, manifested as a local maximum in $|\tilde{\Delta}_\Gamma(\omega)|$. Note that for large pump photon energies exceeding the overall bandwidth, no resonance occurs, and consequently no features are observed.

Panels (e) and (f): Strong pump intensity ($A_0 = 0.5$ Vfs/nm). The behavior is qualitatively similar to the intermediate-intensity case, but the effects are generally stronger. The bending of the bands toward the noninteracting positions near the resonance is more pronounced. Additionally, a residual signal appears when the pump-pulse photon energy is approximately half the band gap, arising from a two-photon resonance process. These panels clearly demonstrate how strongly the exciton-field-induced sideband depends on the pump-pulse intensity.

Figure 6 presents momentum-resolved results for the excitonic-insulator phase.

In Fig. 6(a), we show the equilibrium $\mathbf{k}$ -resolved ARPES signal along the high-symmetry path in the BZ (no pump). The signal is centered around the lower *interacting* energy band. The *interacting* bands are indicated by the red dotted lines, and represent the eigenvalues of the full Hamiltonian after the self-consistent procedure described in Appendix A. For comparison, the noninteracting bands are indicated by the black dotted lines. The renormalization of the band structure due to exciton formation is clearly visible.

In Fig. 6(b), we show the $\mathbf{k}$ -resolved post-pump TR-ARPES signal along the high-symmetry path for pump frequency $\hbar\omega_{pu} = 2.5$ eV and pump amplitude $A_0 = 0.5$ Vfs/nm. There are $\mathbf{k}$ -points near Γ that are in resonance with this pump-pulse frequency: the electrons can be excited from the interacting VB to the noninteracting CB. The broken excitonic bonds induce coherent oscillations, giving rise to exciton-field-induced sidebands that run parallel to both the occupied CB and the VB. Additionally, we observe slightly

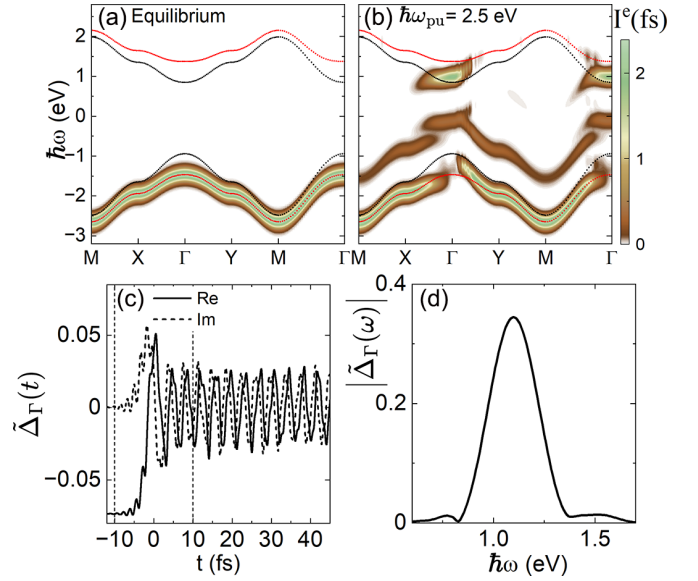


FIG. 6. Results for $V = 15$ eV, excitonic-insulator phase. (a) Equilibrium $\mathbf{k}$ -resolved ARPES signal along the high-symmetry path in the BZ for the excitonic-insulator phase (no pump). (b) Post-pump $\mathbf{k}$ -resolved TR-ARPES signal for $\hbar\omega_{pu} = 2.5$ eV and $A_0 = 0.5$ V fs/nm, showing exciton-field-induced sidebands parallel to the occupied CB and VB. In panels (a) and (b), red dotted lines indicate the interacting band structure from the self-consistent procedure (Appendix A); black dotted lines indicate the noninteracting bands. (c) Time evolution of $\tilde{\Delta}_\Gamma(t)$ for the same parameters as panel (b), showing reduction and oscillations upon pumping. (d) Post-pump Fourier transform $|\tilde{\Delta}_\Gamma(\omega)|$, revealing the main excitonic oscillation frequency.

occupied CB states at $\mathbf{k}$ -points that are not in direct resonance with the pump frequency. This occurs because during the application of the pump pulse, the HF self-energy is finite and oscillating, which can generate additional resonances involving the sum or difference of these oscillation frequencies with the pump frequency.

In Figs. 6(c) and 6(d), we examine the excitonic order parameter dynamics. Panel (c) shows $\tilde{\Delta}_\Gamma(t)$ as a function of time for the same parameters. Initially, in equilibrium, $\tilde{\Delta}_\Gamma$ is finite, reflecting the presence of the excitonic order parameter in the excitonic-insulator phase. Upon application of the pump pulse, its absolute value is reduced, and both the real and imaginary parts exhibit oscillations, indicating that the pump has partially suppressed but not completely destroyed the excitonic order, leaving a coherently oscillating component. Panel (d) displays the amplitude of the post-pump Fourier transform, $|\tilde{\Delta}_\Gamma(\omega)|$, which reveals a main peak at a characteristic excitonic frequency. This confirms that coherent excitonic oscillations persist after the pump pulse in the excitonic-insulator phase, with a reduced amplitude compared to the equilibrium value of $\tilde{\Delta}_\Gamma$.

E. Local Coulomb interaction limit

In this section, we analyze the limit of local interaction. This limit is obtained by setting both $V \rightarrow \infty$ and $\kappa \rightarrow \infty$ in Eq. (24), such that $V(\mathbf{k}) \rightarrow V_{loc} = V/\kappa^2$ remains finite

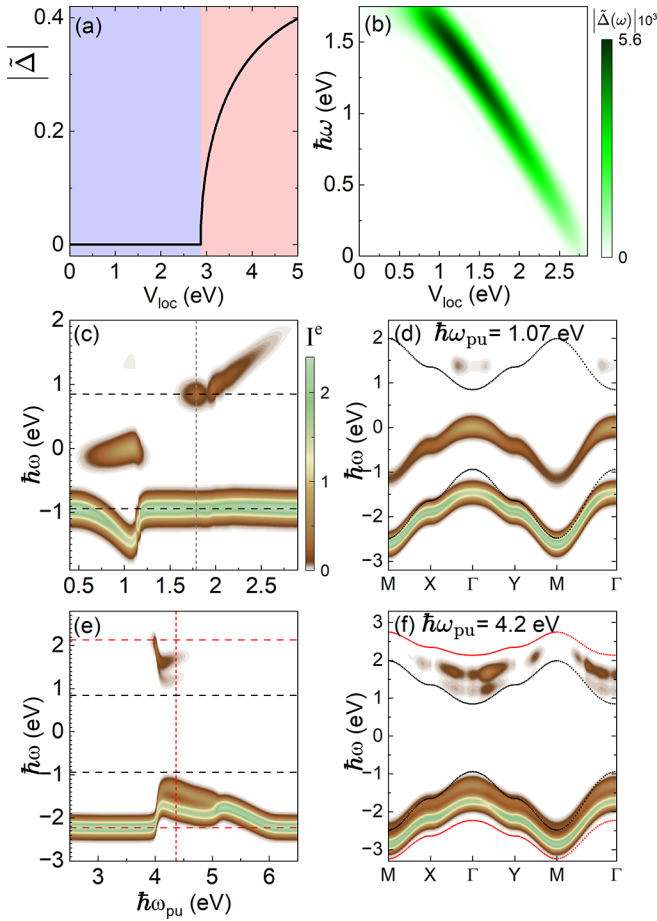


FIG. 7. Results for the local interaction. (a) Equilibrium $\tilde{\Delta}$ as a function of the local Coulomb interaction strength V_{loc} , showing the transition between semiconducting and excitonic-insulator phases at $V_{\text{loc}} \approx 2.87$ eV. (b) $|\tilde{\Delta}(\omega)|$ as a function of V_{loc} and frequency, giving the equilibrium excitonic frequency in the local interaction limit, for the same protocol as in Figs. 1(c) and 1(d). (c) Post-pump TR-ARPES signal at Γ for the semiconducting phase ($V_{\text{loc}} = 2$ eV) with $A_0 = 0.5$ V fs/nm, counterpart of Fig. 2(e). (d) $\mathbf{k}$ -resolved post-pump TR-ARPES signal for $\hbar\omega_{\text{pu}} = 1.07$ eV and $A_0 = 0.5$ V fs/nm in the semiconducting phase, counterpart of Fig. 4(a). (e) Post-pump TR-ARPES signal at Γ for the excitonic-insulator phase ($V_{\text{loc}} = 5$ eV) with $A_0 = 0.5$ V fs/nm, counterpart of Fig. 5(e). (f) $\mathbf{k}$ -resolved post-pump TR-ARPES signal for $\hbar\omega_{\text{pu}} = 4.2$ eV and $A_0 = 0.5$ V fs/nm in the excitonic-insulator phase, counterpart of Fig. 6(b).

and becomes momentum-independent. In this limit, the HF self-energy becomes $\mathbf{k}$ -independent, which simplifies the numerical calculations.

In Fig. 7(a), we study the equilibrium phase diagram by plotting the equilibrium value of $\tilde{\Delta}_{\Gamma} = \tilde{\Delta}$ as a function of V_{loc} . There is a critical local Coulomb interaction strength of approximately 2.87 eV. Below this critical value, the system is in the semiconducting phase; above it, the system enters the excitonic-insulator phase. Applying the same protocol as in Figs. 1(c) and 1(d), we plot $|\tilde{\Delta}(\omega)|$ as a function of V_{loc} and frequency in Fig. 7(b), thereby obtaining the equilibrium excitonic frequency in this local interaction regime.

In Figs. 7(c) and 7(d), we consider $V_{\text{loc}} = 2$ eV, which corresponds to the semiconducting phase. Panel (c) shows

the post-pump TR-ARPES signal at the Γ point ($\mathbf{k} = \mathbf{0}$) as a function of the pump frequency $\hbar\omega_{\text{pu}}$ for the amplitude $A_0 = 0.5$ V fs/nm. This figure is the counterpart of Fig. 2(e). The qualitative behavior is similar, but quantitatively it reflects the $\mathbf{k}$ -independent nature of the interaction. Panel (d) displays the $\mathbf{k}$ -resolved post-pump TR-ARPES signal along the high-symmetry path for pump frequency $\hbar\omega_{\text{pu}} = 1.07$ eV and pump amplitude $A_0 = 0.5$ V fs/nm. This figure is the counterpart of Fig. 4(a). Again, the qualitative behavior is similar, while the quantitative details differ. Also in this case, the excitonic-field-induced Floquet sideband is stronger near the Γ point than at other momenta. This behavior clarifies what we have already stated for the $\mathbf{k}$ -dependent interaction: it is the proximity of the excitonic frequency to the energy band gap at Γ (the small-detuning condition) that causes a stronger sideband close to Γ and not the $\mathbf{k}$ -dependence of the interaction.

In Figs. 7(e) and 7(f), we consider $V_{\text{loc}} = 5$ eV, which corresponds to the excitonic-insulator phase. Panel (e) shows the post-pump TR-ARPES signal at the Γ point ($\mathbf{k} = \mathbf{0}$) as a function of the pump frequency $\hbar\omega_{\text{pu}}$ for the amplitude $A_0 = 0.5$ V fs/nm. This figure is the counterpart of Fig. 5(e). Panel (f) shows the $\mathbf{k}$ -resolved post-pump TR-ARPES signal along the high-symmetry path for pump frequency $\hbar\omega_{\text{pu}} = 4.2$ eV and pump amplitude $A_0 = 0.5$ V fs/nm. This figure is the counterpart of Fig. 6(b). Once more, the qualitative behavior is similar, but quantitatively it is different: the pumped exciton frequency in this case is so low that the sidebands are very close in energy to the main bands.

IV. SUMMARY AND CONCLUSIONS

In this work, we have developed a theoretical framework based on the dynamical projective operatorial approach (DPOA) to investigate the time-resolved ARPES response of pumped excitonic systems. By incorporating Coulomb electron-electron interactions at the Hartree-Fock level, our formalism captures the formation of excitonic bound states both in equilibrium and under the influence of an external pump pulse. We have applied this framework to a prototypical two-dimensional two-band semiconductor model, systematically exploring the equilibrium phase diagram and the out-of-equilibrium dynamics for both the semiconducting and the excitonic-insulator phases.

Our results demonstrate that pump-induced coherent oscillations of the excitonic order parameter persist after the pump pulse subsides and give rise to clear excitonic-field-driven Floquet sidebands in the TR-ARPES signal. These Floquet sidebands are distinct from those originating from the pump laser field itself and provide a direct fingerprint of excitonic coherence.

For the semiconducting phase, where no excitons exist in equilibrium, we showed that resonant pumping at the excitonic frequency generates coherent excitonic oscillations that manifest as Floquet sidebands parallel to the VB in the post-pump TR-ARPES spectrum. The Mexican-hat effect emerges for sufficiently strong pump intensities, and we distinguished genuine exciton-field-induced sidebands from band-resonance-induced sidebands, which arise from residual coherences at momentum points resonant with the pump frequency.

Figure 8(b) is the counterpart of Fig. 5(e) for $D = 0$, corresponding to the excitonic-insulator phase. In contrast to the semiconducting case, the main excitonic sideband persists even when $D = 0$. However, it becomes much weaker and exhibits different features compared to the case with finite D .

This indicates that while excitonic order exists in equilibrium in the excitonic-insulator phase, the dipole moment still plays an important role in mediating the coupling between the pump pulse and the excitonic field, thereby influencing the post-pump spectral response.

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