

Semiconductor Wannier equations: A real-time, real-space approach to the nonlinear optical response in crystals

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We develop the semiconductor Wannier equations (SWEs), a real-time, real-space formulation of ultrafast light-matter dynamics in crystals, by deriving the equations of motion for the electronic reduced density matrix in a localized Wannier basis. Working in real space removes the structure-gauge ambiguities that hinder reciprocal-space semiconductor Bloch equations. Electron-electron interactions are included at the time-dependent Hartree plus static screened-exchange level. Decoherence is modeled with three complementary channels: pure dephasing, population relaxation, and distance-dependent real-space dephasing, providing physically grounded damping for strong-field phenomena such as high-harmonic generation. Conceptually, the SWEs bridge real-space semiclassical intuition with many-body solid-state optics, offering a numerically robust and gauge-clean alternative to reciprocal-space approaches for nonlinear optical response and attosecond spectroscopy in solids.

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I. INTRODUCTION

In 2011, the discovery of high-harmonic generation (HHG) in solids [1] opened a frontier in attosecond science, extending concepts originally developed in atomic and molecular systems [2] to the condensed matter domain [3–6]. This breakthrough demonstrated that high-order harmonics could be generated directly from crystalline solids subjected to strong midinfrared laser fields, unveiling a regime of ultrafast light-matter interaction governed by the interplay of band structure [7], Bloch electron dynamics [1], and quantum coherence [8,9].

Since then, high-harmonic spectroscopy has emerged as a powerful tool to probe electronic and structural properties of solids on femtosecond and attosecond timescales [5]. Applications range from observation of Bloch oscillations [1], optical reconstruction of the band structure [7], electron-hole dynamics [10], probing of collective excitations [11], imaging of Van

Hove singularities [12], topological phase transitions [13], and dynamics in strongly correlated materials [14,15], to name a few.

The understanding of high-harmonic generation in atoms and molecules is deeply rooted in the semiclassical three-step model [16,17] and its quantum counterpart, the Lewenstein model [18]. These foundational works underpin the field of attosecond physics, offering essential physical insight and establishing a clear connection between HHG and classical electron trajectories [19,20]. *Ab initio* simulations, especially those solving the time-dependent Schrödinger equation in real space where electron trajectories can be directly visualized [21], were instrumental in identifying electron recollision as the primary mechanism behind high-harmonic emission in the early days of attoscience [17].

Several theoretical frameworks have been used to model HHG in solids, including semiclassical approaches [22,23], time-dependent density functional theory [24,25], and the semiconductor Bloch equations (SBEs) [26–31]. Among these approaches, the SBEs in the length gauge, where the coupling to a homogeneous electric field is described via the scalar potential $\phi(\mathbf{r}, t) = -\mathbf{r} \cdot \mathbf{E}(t)$, have proven particularly successful in capturing the underlying physics and providing valuable insight into the HHG process [29,32].

The advantages of the length gauge formulation of the SBEs when compared to the velocity gauge are numerous [29,32]. First, less bands are required for the convergence

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of numerical simulations when compared to velocity gauge [33,34]. This is also the case in atomic and molecular targets; i.e., when only bound-bound transitions are involved, the length gauge is preferable over the velocity gauge [35,36]. Second, the inclusion of dephasing is most transparently done in the length gauge [29]. Last, the decomposition of currents into inter- and intraband components is only natural when working in the length gauge [29].

Despite its advantages, working in the length gauge presents a significant challenge due to the fact that the position operator, which appears in the light-matter interaction term, is ill-defined in a crystal with periodic boundary conditions [37]. The standard solution is to define the position operator in the thermodynamic limit, which requires computing derivatives of the Bloch functions with respect to Bloch's crystal momentum [38]. The gauge freedom in defining the Bloch functions up to a random phase, which we will refer to as *structure gauge*, poses severe difficulties to the numerical solution of the SBEs [34,39]. Solutions to this technical problem have been proposed in the literature, such as the twisted parallel transport gauge [34], the gauge invariant SBEs [32,40], and the use of maximally localized Wannier functions to construct a smooth gauge in the reciprocal space [31].

In solid-state systems, the seminal work of Vampa *et al.* [41] introduced a semiclassical framework for HHG, emphasizing the role of electron-hole pair trajectories in the process. However, most standard theoretical tools available to describe electron dynamics in solids, such as the SBE, are usually formulated in reciprocal space. A theoretical toolbox formulated in real space is essential to bridge the gap between the real-space semiclassical picture and current theoretical methods.

In this work, we present an alternative formulation of the SBEs, the semiconductor Wannier equations (SWEs), where the dynamical object propagated in time is the one-electron reduced density matrix (1RDM), represented in a real-space basis of localized Wannier orbitals. We will show that with this approach, the problems that stem from the structure-gauge freedom are absent. Furthermore, real-space dephasing to model solid-state HHG makes the use of unphysically short dephasing times unnecessary [9,42]. Also, the inclusion of excitonic effects [43] is substantially easier than in the SBEs formulation. Finally, in the context of solid-state attoscience, we demonstrate that SWEs are significantly more efficient than SBEs, opening the way to simulate larger and complex systems.

From a conceptual standpoint, this work opens avenues for exploring theoretical quantities in real space to advance the interpretation of solid-state HHG. It also contributes to bridging the gap between the concepts developed in atomic and molecular attoscience and their counterparts in the solid-state regime.

II. THEORETICAL FORMALISM

In this section, we will derive the equations of motion that form the backbone of the semiconductor Wannier equations. In the following, we will use atomic units unless otherwise stated.

A. General formulation

1. Hamiltonian

We start by writing a general electronic Hamiltonian, $\hat{\mathcal{H}}(t)$, that is the sum of a single-particle time-dependent Hamiltonian, $\hat{h}(t) = \hat{h}^0 + \hat{U}(t)$, and a time-independent electron-electron interaction term, $\hat{V}$. In a second quantization formalism, we have that

$$\hat{\mathcal{H}}(t) = \underbrace{\sum_{ij} h_{ij}(t) c_i^\dagger c_j}_{\hat{h}(t)} + \underbrace{\frac{1}{2} \sum_{ijkl} V_{kl}^{ij} c_i^\dagger c_j^\dagger c_l c_k}_{\hat{V}}, \quad (1)$$

$$h_{ij}(t) = \int d\mathbf{r} \phi_i^*(\mathbf{r}) \hat{h}(t) \phi_j(\mathbf{r}), \quad (2)$$

$$V_{kl}^{ij} = \int d\mathbf{r} d\mathbf{r}' \phi_i^*(\mathbf{r}) \phi_j^*(\mathbf{r}') v_C(\mathbf{r}, \mathbf{r}') \phi_l(\mathbf{r}') \phi_k(\mathbf{r}), \quad (3)$$

where $c_i^\dagger$ (c_i) is the creation (annihilation) operator for an electron in state i with wavefunction $\phi_i(\mathbf{r})$ and $v_C(\mathbf{r}, \mathbf{r}')$ is the electron-electron interaction potential. Since the interaction potential is of the form $v_C(\mathbf{r}, \mathbf{r}') = v_C(\mathbf{r} - \mathbf{r}') = v_C(\mathbf{r}' - \mathbf{r})$, the tensor V_{kl}^{ij} has the following symmetries: $V_{kl}^{ij} = V_{lk}^{ji} = (V_{ij}^{kl})^*$.

In this work, the time-dependent perturbation, $\hat{U}(t)$, will be a light-matter interaction term. Under the dipole approximation and using the electromagnetic length gauge, $\hat{U}(t)$ can be written as

$$\hat{U}(t) = -e\hat{\mathbf{r}} \cdot \mathbf{E}(t), \quad (4)$$

where $e = -1$ is the electron charge, $\mathbf{E}(t)$ is the applied homogeneous electric field, and $\hat{\mathbf{r}}$ is the position operator.

2. Observables and the one-electron reduced density matrix

In most of the cases, we are interested in calculating observables that are single-particle operators, which have the general form $\hat{O} = \sum_{ij} O_{ij} c_i^\dagger(t) c_j(t)$ (from hereinafter we will be using the Heisenberg picture). The expectation value of a single-particle operator can be written as $\langle \hat{O}(t) \rangle = \sum_{ij} O_{ij} \rho_{ji}(t) = \text{tr}(O\rho(t))$, where

$$\rho_{ij}(t) \equiv \langle c_j^\dagger(t) c_i(t) \rangle \quad (5)$$

is the one-electron reduced density matrix. Note that the indexes are switched.

Regarding optical response, two of the most crucial and important observables are the position and velocity operators. The position operator is just expressed as

$$\hat{\mathbf{r}} = \sum_{ij} \mathbf{r}_{ij} c_i^\dagger c_j, \quad (6)$$

$$\mathbf{r}_{ij} = \int d\mathbf{r}' \phi_i^*(\mathbf{r}') \mathbf{r}' \phi_j(\mathbf{r}'). \quad (7)$$

The velocity operator $\hat{\mathbf{v}}$ can be calculated using the Heisenberg equations of motion

$$\hat{\mathbf{v}} = -i[\hat{\mathbf{r}}, \hat{\mathcal{H}}(t)]. \quad (8)$$

The electron-electron interaction term, $\hat{V}$, is an operator that only depends on the position of the electrons; therefore, it

commutes with $\hat{r}$. Equivalently, this reflects the fact that internal forces do not modify the total current, in agreement with Newton's third law. Furthermore, if the time-dependent perturbation, $\hat{U}(t)$, also commutes with the position, as is the case when working under the dipole approximation in the length gauge, the velocity operator can be written as

$$\hat{v} = \sum_{ij} v_{ij} c_i^\dagger c_j = -i[\hat{r}, \hat{h}^0], \quad (9)$$

where $v_{ij} = -i \sum_k (\mathbf{r}_{ik} h_{kj}^0 - h_{ik}^0 \mathbf{r}_{kj})$ are the velocity matrix elements and $\langle \hat{v} \rangle = \text{tr}(\mathbf{v} \rho)$. It is important to emphasize that the calculation of the velocity operator is done without assuming any approximations. In the following, we will approximate the equations of motion by performing a mean-field decoupling, effectively adding a self-energy term, Σ , to $\hat{h}^0$. One might be tempted to include in the definition of the velocity operator this self-energy term, Σ . This would be incorrect; however, Eq. (9) was derived without any approximation, and it is the correct expression for $\hat{v}$ even in the presence of Σ . Recomputing the velocity as $-i[\hat{r}, \hat{h}^0 + \Sigma]$ would therefore amount to double counting the interaction effect: the interaction has already been accounted for once in the exact derivation of the current operator, and Σ is only introduced afterward as an approximate closure for the dynamics.

3. Equations of motion for the one-electron reduced density matrix

Using the Heisenberg equation, we will determine the equation of motion for the 1RDM, $\rho_{ij}(t) = \langle c_j^\dagger(t) c_i(t) \rangle$. We have used QUANTUMALGEBRA.JL [44,45] to deal with the quantum operator algebra. We have that (omitting the time argument for simplicity)

$$\begin{aligned} -i \frac{d(c_j^\dagger c_i)}{dt} &= [\hat{\mathcal{H}}(t), c_j^\dagger c_i], \quad (10) \\ -i \frac{d\rho_{ij}}{dt} &= \langle [\hat{\mathcal{H}}(t), c_j^\dagger c_i] \rangle = \sum_{\alpha} h_{\alpha j}(t) \rho_{i\alpha} - \sum_{\alpha} h_{i\alpha}(t) \rho_{\alpha j} \\ &\quad - \frac{1}{2} \sum_{\alpha\beta\gamma} V_{\gamma j}^{\alpha\beta} \langle c_{\alpha}^{\dagger} c_{\beta}^{\dagger} c_{\gamma} c_i \rangle \\ &\quad + \frac{1}{2} \sum_{\alpha\beta\gamma} V_{j\gamma}^{\alpha\beta} \langle c_{\alpha}^{\dagger} c_{\beta}^{\dagger} c_{\gamma} c_i \rangle \\ &\quad + \frac{1}{2} \sum_{\alpha\beta\gamma} V_{\beta\gamma}^{\alpha i} \langle c_{\alpha}^{\dagger} c_j^{\dagger} c_{\beta} c_{\gamma} \rangle \\ &\quad - \frac{1}{2} \sum_{\alpha\beta\gamma} V_{\beta\gamma}^{i\alpha} \langle c_{\alpha}^{\dagger} c_j^{\dagger} c_{\beta} c_{\gamma} \rangle. \quad (11) \end{aligned}$$

We arrive to an equation of motion for the 1RDM that involves the computation and evolution of the expectation value of four operators, $\langle c_i^\dagger c_j^\dagger c_l c_k \rangle$. If we proceed to compute the equation of motion for the mean value of four operators, we will get equations involving mean values of six operators. If we proceed further, we end up with an infinite hierarchy of operators, also known as Bogoliubov-Born-Green-Kirkwood-Ývon (BBGKY) hierarchy [27,28,44]. In the following, we will use a mean-field decoupling to obtain a close set of equations for the 1RDM.

4. Mean-field decoupling

To obtain a closed set of equations, we can apply a mean-field decoupling to the $\langle c_i^\dagger c_j^\dagger c_l c_k \rangle$ terms, equivalent to a Hartree-Fock approximation. Specifically, we will approximate [46,47]

$$\langle c_i^\dagger c_j^\dagger c_l c_k \rangle \approx \langle c_i^\dagger c_k \rangle \langle c_j^\dagger c_l \rangle - \langle c_i^\dagger c_l \rangle \langle c_j^\dagger c_k \rangle \approx \rho_{ki} \rho_{lj} - \rho_{li} \rho_{kj}. \quad (12)$$

Note that in this decoupling, we neglect anomalous terms, $\langle c_i^\dagger c_j^\dagger \rangle$ and $\langle c_i c_j \rangle$, that may be relevant to superconductivity [48]. Replacing this decoupling in the equation of motion for the 1RDM [Eq. (11)], we obtain the equation of motion for the 1RDM within the time-dependent Hartree-Fock approximation:

$$i \frac{d\rho_{ij}}{dt} = [h(t), \rho]_{ij} + [\Sigma^F[\rho], \rho]_{ij} + [\Sigma^H[\rho], \rho]_{ij}, \quad (13)$$

where Σ^F and Σ^H are the Fock and Hartree self-energies (mean-field potentials) and are defined as

$$\Sigma^F[\rho]_{ij} = - \sum_{kl} V_{kj}^{il} \rho_{kl}, \quad (14)$$

$$\Sigma^H[\rho]_{ij} = \sum_{kl} V_{jk}^{il} \rho_{kl}. \quad (15)$$

Up to this point, $\hat{h}^0$ has been treated as a purely noninteracting Hamiltonian. However, in practical calculations, $\hat{h}^0$, and therefore the equilibrium RDM [49], ρ^0 , are obtained from either tight-binding parametrizations of the experimental equilibrium dispersion or from different levels of *ab initio* calculations, such as *GW* [50] or hybrid density-functional theory calculations [51]. In both cases, the resulting $\hat{h}^0$ already includes some level of electron-electron interactions, in the form of a single-particle self-energy Σ_0 at equilibrium. Therefore, in the equation of motion [Eq. (13)], we must subtract the equilibrium ground-state self-energy, $\Sigma_0 = \Sigma^F[\rho_0] + \Sigma^H[\rho_0]$, to avoid double counting.

In the present formulation, $\hat{h}^0$ is used as an effective single-particle Hamiltonian for the active subspace. Thus, quasiparticle-level renormalizations of the band energies, including band-gap renormalization, are assumed to be incorporated, at least effectively, in the input band structure. The interaction terms treated explicitly below should therefore be understood as residual screened interactions within the reduced subspace.

Second, the Fock self-energy neglects important screening effects, which are usually taken into account within a random-phase approximation (RPA) [46,50]. As a result, the interaction potential V in Eq. (14) is often replaced with W , the RPA-screened interaction potential. Note that this argument does not apply to the Hartree self-energy since RPA screening would result in diagrams that are not single-particle irreducible, leading to double counting of screening. The screened interaction W is, in general, a dynamical quantity, $W(\omega)$, as it depends on the state of the system. However, in order to keep the equations of motion local in time, we will employ a common approximation, known as static exchange

(SEX), by taking the static limit, i.e., $W \approx W(\omega \rightarrow 0)$ [50]. Therefore, our final equation of motion will read

$$i \frac{d\rho_{ij}}{dt} = [h(t) + \Sigma^{\text{HSEX}}[\rho] - \Sigma_0, \rho]_{ij}, \quad (16)$$

$$\Sigma^{\text{SEX}}[\rho]_{ij} = - \sum_{kl} W_{kj}^{il} \rho_{kl}, \quad (17)$$

$$\Sigma^{\text{HSEX}}[\rho]_{ij} = \Sigma^{\text{SEX}}[\rho]_{ij} + \Sigma^H[\rho]_{ij}, \quad (18)$$

$$\Sigma_0 = \Sigma^{\text{HSEX}}[\rho^0]. \quad (19)$$

The Σ_0 term is chosen to be the full self-energy at equilibrium, regardless of the level of approximation used to compute h^0 and ρ^0 . This will ensure that ρ^0 , the equilibrium 1RDM, will remain a stationary solution of the equations of motion, in the absence of driving. This approach is sometimes known in the literature as time-dependent Bethe-Salpeter (TD-BSE) equation [52], time-dependent adiabatic GW (TD-aGW) [53], or time-dependent Hartree plus screened exchange (TD-HSEX) [54–56].

The specific form of V_{kl}^{ij} and W_{kl}^{ij} can be simplified when working with a localized basis, as occurs in a tight-binding formulation. Specifically, assuming that the orbitals $\phi_i^*(\mathbf{r})$ are strongly localized around the center of the orbital, $\boldsymbol{\tau}_i = \langle i | \mathbf{r} | i \rangle$, we can approximate

$$\begin{aligned} V_{kl}^{ij} &= \int d\mathbf{r} d\mathbf{r}' \phi_i^*(\mathbf{r}) \phi_j^*(\mathbf{r}') V(\mathbf{r} - \mathbf{r}') \phi_l(\mathbf{r}') \phi_k(\mathbf{r}) \\ &\approx V(\boldsymbol{\tau}_i - \boldsymbol{\tau}_j) \int d\mathbf{r} d\mathbf{r}' \phi_i^*(\mathbf{r}) \phi_j^*(\mathbf{r}') \phi_l(\mathbf{r}') \phi_k(\mathbf{r}), \end{aligned} \quad (20)$$

which is a valid approximation, provided that $V(\mathbf{r} - \mathbf{r}')$ does not vary strongly within typical size of an orbital. If we further recall the orthogonality of Wannier orbitals, $\int d\mathbf{r} \phi_i^*(\mathbf{r}) \phi_k(\mathbf{r}) = \delta_{ik}$, we obtain

$$V_{kl}^{ij} \approx \delta_{ik} \delta_{jl} V(\boldsymbol{\tau}_i - \boldsymbol{\tau}_j), \quad (21)$$

where $\boldsymbol{\tau}_i$, $\boldsymbol{\tau}_j$ are the centers of the orbitals i and j . In the same way,

$$W_{kl}^{ij} \approx \delta_{ik} \delta_{jl} W(\boldsymbol{\tau}_i - \boldsymbol{\tau}_j). \quad (22)$$

We refer to this as the ultralocalized orbital approximation [47]. Within this approximation, we can then calculate $\Sigma^{\text{SEX}}[\rho]$ and $\Sigma^H[\rho]$ as

$$\Sigma^{\text{SEX}}[\rho]_{ij} \approx -W(\boldsymbol{\tau}_i - \boldsymbol{\tau}_j) \rho_{ij}, \quad (23)$$

$$\Sigma^H[\rho]_{ij} \approx \delta_{ij} \sum_k \rho_{kk} V(\boldsymbol{\tau}_i - \boldsymbol{\tau}_k). \quad (24)$$

When working with spin-degenerate models, we must multiply the Hartree term, $\Sigma^H[\rho]$, by 2 since it depends on the total charge density. Therefore,

$$\Sigma^H[\rho]_{ij} \approx \delta_{ij} \sum_k s_k \rho_{kk} V(\boldsymbol{\tau}_i - \boldsymbol{\tau}_k), \quad (25)$$

where s_k takes into account the spin degeneracy of the k orbital.

We emphasize that the ultralocalized orbital approximation is not an essential component of the SWEs formalism, but an additional approximation adopted here to simplify the evaluation of the two-electron integrals entering the Σ^{HSEX}

self-energy. The SWEs can be formulated using the full interaction matrix elements, V_{kl}^{ij} and W_{kl}^{ij} , although at a higher computational cost. The approximation is expected to be reasonable when the Wannier orbitals are sufficiently localized and the interaction potentials do not vary significantly over their spatial extent.

5. Decoherence

The equation of motion derived in the previous subsection [Eq. (16)] is purely coherent, with no dissipation effects. However, decoherence plays a major role in the modelization of electronic dynamics in solids under intense lasers, particularly in high-harmonic generation [8,9,41,42]. To that purpose, we must include decoherence in our framework. We will include three forms of decoherence: pure dephasing, relaxation, and real-space dephasing.

In the seminal work by Vampa *et al.* [41], it was found that HHG spectrum obtained by solving the SBEs in ZnO was very noisy and agreement with experiments was only achieved if ultrashort dephasing times of few femtoseconds, $T_2 \equiv \gamma_D^{-1}$, were introduced in the simulations. Pure dephasing is introduced in the basis that diagonalizes the unperturbed Hamiltonian, $\hat{h}^0 |\varepsilon_i\rangle = \varepsilon_i |\varepsilon_i\rangle$. The pure dephasing term in the equation of motion, in the eigenbasis of $\hat{h}^0$, reads

$$\mathcal{L}_D[\rho]_{ij} = \gamma_D (\delta_{ij} - 1) \rho_{ij}. \quad (26)$$

Despite pure dephasing being commonly used in numerical simulations of HHG in solids, it does not account for population relaxation. To that purpose, within the relaxation time approximation and assuming that the relaxation rate is much smaller than the driving frequency, we include relaxation to thermal equilibrium, ρ^0 [57–59]. The relaxation term reads as

$$\mathcal{L}_r[\rho] = -\gamma_r (\rho - \rho_0), \quad (27)$$

where $\gamma_r \equiv (2T_r)^{-1}$ is the relaxation rate.

Recently, Brown *et al.* [9,42] introduced an additional term that implements real-space dephasing where the rate of dephasing depends on the distance between orbitals. In this specific case, we must work in a basis where the single-particle orbitals are well localized. Having $\boldsymbol{\tau}_i$ as the center of state i , we have that the real-space dephasing term reads as

$$\mathcal{L}_{\text{rs}}[\rho]_{ij} = -\gamma_{\text{rs}}(|\boldsymbol{\tau}_i - \boldsymbol{\tau}_j|) \cdot (\rho - \rho^0)_{ij}, \quad (28)$$

where $\gamma_{\text{rs}}(|\boldsymbol{\tau}_i - \boldsymbol{\tau}_j|)$ is a dephasing rate that depends on the distance between orbitals i and j and is zero at the origin, $\gamma_{\text{rs}}(0) = 0$. The functional form that will be used for γ_{rs} is a polynomial function that only acts on coherences between states whose distance is greater than $R_{\text{rs,cut}}$, i.e.,

$$\gamma_{\text{rs}}(r) = \begin{cases} 0, & r < R_{\text{rs,cut}}, \\ \beta_{\text{rs}}(r - R_{\text{rs,cut}})^{\alpha_{\text{rs}}}, & r \geq R_{\text{rs,cut}}, \end{cases} \quad (29)$$

where α_{rs} is the degree of the polynomial. This form of real-space dephasing is similar to the one used in Ref. [9] and resembles the complex absorbing potentials used in atomic and molecular strong-field calculations [60–63].

B. Crystalline systems

We are now in position to lay down the equations of motion for the electronic RDM in a crystalline solid in real space. Let

we consider an infinite crystalline solid with the Bravais lattice

$$\mathbf{R} = \sum_{i=1}^D n_i \mathbf{a}_i, \quad n_i \in \mathbb{Z}, \quad (30)$$

where D is the dimension of the system, $\mathbf{a}_i$ are the primitive lattice vectors, and $\mathbf{b}_i$ denotes the primitive reciprocal lattice vectors such that $\mathbf{a}_i \cdot \mathbf{b}_j = 2\pi\delta_{ij}$. Let us consider a set of M localized Wannier spin orbitals in each cell $w_\alpha(\mathbf{r} - \mathbf{R}) = \langle \mathbf{r} | \mathbf{R} \alpha \rangle$. These Wannier orbitals are characterized by two indexes $(\mathbf{R}, \alpha)$, with $\mathbf{R}$ the center of the orbital and α the orbital type, and form an orthonormal basis, i.e., $\langle \mathbf{R}' \beta | \mathbf{R} \alpha \rangle = \delta_{\alpha\beta} \delta_{\mathbf{R}', \mathbf{R}}$. Throughout this work, we use the terms “Wannier orbitals” and “localized orbitals” interchangeably. We will denote $\rho_{\alpha\beta}(\mathbf{R}', \mathbf{R}) \equiv \langle c_{\mathbf{R}\beta}^\dagger c_{\mathbf{R}'\alpha} \rangle$ and $\rho_{\alpha\beta}(\mathbf{R}) \equiv \rho_{\alpha\beta}(\mathbf{0}, \mathbf{R})$. As a consequence of the periodicity of our system, the equilibrium state, ρ^0 , obeys

$$\rho_{\alpha\beta}^0(\mathbf{R} - \mathbf{R}') = \rho_{\alpha\beta}^0(\mathbf{R}', \mathbf{R}). \quad (31)$$

This is a consequence of Bloch’s theorem, which in reciprocal space only allows for nonvanishing coherences between states with the same $\mathbf{k}$.

Two basic quantities in the problem of light-matter interaction are the unperturbed Hamiltonian, $\hat{h}^0$, and the position operator, $\hat{\mathbf{r}}$. Their matrix elements can be either calculated from electronic structure calculations followed by a wannierization procedure [64] or be defined in the context of a phenomenological tight-binding model. For the description of light-matter interaction under the dipole approximation, it is necessary to have the knowledge of their matrix elements

$$h_{\alpha\beta}^0(\mathbf{R}) \equiv \langle \mathbf{0} \alpha | \hat{h}^0 | \mathbf{R} \beta \rangle, \quad (32)$$

$$\mathbf{r}_{\alpha\beta}(\mathbf{R}) \equiv \langle \mathbf{0} \alpha | \hat{\mathbf{r}} | \mathbf{R} \beta \rangle. \quad (33)$$

The unperturbed Hamiltonian is invariant under lattice translations, i.e., $\langle \mathbf{0} \alpha | \hat{h}^0 | (\mathbf{R} - \mathbf{R}') \beta \rangle = \langle \mathbf{R}' \alpha | \hat{h}^0 | \mathbf{R} \beta \rangle$.

On the other hand, the position operator is not invariant under lattice translations and transforms as

$$\langle \mathbf{R}' \alpha | \hat{\mathbf{r}} | \mathbf{R} \beta \rangle = \mathbf{r}_{\alpha\beta}(\mathbf{R} - \mathbf{R}') + \delta_{\mathbf{R}', \mathbf{R}} \delta_{\alpha\beta} \mathbf{R}. \quad (34)$$

At first glance, the second term in Eq. (34) might seem problematic as it breaks the periodicity of our problem. However, under the dipole approximation, the reduced density matrix in real space is an object that remains invariant under lattice translations, i.e., $\rho_{\alpha\beta}(\mathbf{R} - \mathbf{R}') = \rho_{\alpha\beta}(\mathbf{R}', \mathbf{R})$. Let us take a closer look at the equation of motion for the RDM by only taking into account the problematic $\delta_{\mathbf{R}', \mathbf{R}} \delta_{\alpha\beta} \mathbf{R}$ term

$$i \frac{d\rho_{\alpha\beta}(\mathbf{R}', \mathbf{R})}{dt} = -E(t) \cdot (\mathbf{R} - \mathbf{R}') \rho_{\alpha\beta}(\mathbf{R}', \mathbf{R}). \quad (35)$$

Taking into account that in our initial state, $\rho_{\alpha\beta}^0(\mathbf{R}', \mathbf{R}) = \rho_{\alpha\beta}^0(\mathbf{0}, \mathbf{R} - \mathbf{R}')$, we can see that the previous term in the equation of motion will maintain the lattice invariance of the 1RDM. In the following, we will always assume implicitly this property of the 1RDM, i.e.,

$$\rho_{\alpha\beta}(\mathbf{R}) = \rho_{\alpha\beta}(\mathbf{R}', \mathbf{R} + \mathbf{R}'), \quad (36)$$

and it is this quantity that will be our central dynamical object.

Both self-energy terms, Σ^H and Σ^{SEX} , will also be invariant under lattice translations. In the following, we employ the

ultralocalized orbital approximation introduced in Eqs. (21) and (22). Under this approximation, the SEX self-energy term takes the form

$$\Sigma^{\text{SEX}}[\rho]_{\alpha\beta}(\mathbf{R}) = -W(\boldsymbol{\tau}_\alpha - (\boldsymbol{\tau}_\beta + \mathbf{R})) \rho_{\alpha\beta}(\mathbf{R}), \quad (37)$$

where $\boldsymbol{\tau}_\alpha = \langle \mathbf{0} \alpha | \hat{\mathbf{r}} | \mathbf{0} \alpha \rangle$ is the center of the α Wannier function at the $\mathbf{0}$ unit cell. The Hartree self-energy can be written as

$$\Sigma^H[\rho]_{\alpha\beta}(\mathbf{R}) = \delta_{\alpha\beta} \delta_{\mathbf{0}, \mathbf{R}} \sum_\gamma \rho_{\gamma\gamma}(\mathbf{0}) X_{\gamma\alpha}, \quad (38)$$

$$X_{\gamma\alpha} \equiv \sum_{\mathbf{R}'} s_\gamma V(\boldsymbol{\tau}_\alpha - (\boldsymbol{\tau}_\gamma + \mathbf{R}')), \quad (39)$$

where s_γ takes into account the spin degeneracy of the orbital γ .

1. Equations of motion in Wannier basis

We are now in a position to derive the equations of motion for $\rho_{\alpha\beta}^L(\mathbf{R})$, where the upperscript L stands for length gauge, as defined by Eq. (4). In the following, we will omit the indexes for the Wannier orbitals implying that we are referring to an $M \times M$ matrix. The resulting equation of motion is

$$\begin{aligned} i\partial_t \rho^L(\mathbf{R}) = & \sum_{\mathbf{R}'} [h^0(\mathbf{R}'), \rho^L(\mathbf{R} - \mathbf{R}')] \\ & + E(t) \cdot \sum_{\mathbf{R}'} [\mathbf{r}(\mathbf{R}'), \rho^L(\mathbf{R} - \mathbf{R}')] - E(t) \cdot \mathbf{R} \rho^L(\mathbf{R}) \\ & + \sum_{\mathbf{R}'} [\Sigma^{\text{HSEX}}[\rho^L](\mathbf{R}'), \rho^L(\mathbf{R} - \mathbf{R}')] \\ & - \sum_{\mathbf{R}'} [\Sigma_0(\mathbf{R}'), \rho^L(\mathbf{R} - \mathbf{R}')] + i\mathcal{L}_{\text{incoh}}[\rho^L](\mathbf{R}), \end{aligned} \quad (40)$$

where we group all incoherent terms, discussed in Sec. II A 5, into

$$\mathcal{L}_{\text{incoh}} = \mathcal{L}_r + \mathcal{L}_{\text{rs}} + \mathcal{L}_D. \quad (41)$$

The explicit formulation of the incoherent terms in the case of a periodic crystal will be shown later.

2. Length gauge and Peierls gauge

The term $E(t) \cdot \mathbf{R} \rho^L(\mathbf{R})$ in Eq. (40) clearly diverges in a crystal and this poses severe numerical difficulties. These difficulties might be solved by working with sufficiently bigger lattices, since it should be expected that $\rho(\mathbf{R}) \rightarrow 0$ when $|\mathbf{R}| \rightarrow \infty$. At the same time, dealing with the $E(t) \cdot \mathbf{R}$ term for big $\mathbf{R}$ will require a very fine temporal resolution. However, one might find a more elegant solution to this problem. In a reciprocal-space formulation, the position operator is expressed as a gradient in k space [37,38,65]. One might numerically discretize the gradient, as done in previous works [43,66]. An alternative way to deal with the gradient of the density matrix in the reciprocal space is to move to the accelerated basis [29]. In our real-space approach, a time-dependent translation in reciprocal space is represented as a time-dependent phase shift in real space. At the level of the reduced density matrix, this is the real-space counterpart of the Houston-basis transformation [29] in reciprocal-space formulations. Therefore, one might define $\rho^P(\mathbf{R})$, the real-space

1RDM in the Peierls gauge, as

$$\rho^P(\mathbf{R}) = \rho^L(\mathbf{R})\phi_P(t, -\mathbf{R}), \quad (42)$$

where $\phi_P(t, \mathbf{R}) = \exp(-i\mathbf{A}_L(t) \cdot \mathbf{R})$ is the Peierls phase between unit cells and $\mathbf{A}_L(t) = -\int^t \mathbf{E}(t')dt'$ is the laser vector potential. We might define $\mathcal{G}^{P \rightarrow L}$ as the transformation from the length gauge to the Peierls gauge as

$$\mathcal{G}^{L \rightarrow P}[\rho^L(\mathbf{R})] \equiv \rho^P(\mathbf{R}) = \rho^L(\mathbf{R})\phi_P(t, -\mathbf{R}) \quad (43)$$

and the corresponding inverse transformation, $\mathcal{G}^{P \rightarrow L}$. The equation of motion for $\rho^P(\mathbf{R})$ is

$$\begin{aligned} i\partial_t \rho^P(\mathbf{R}) = & \sum_{\mathbf{R}'} \phi_P(t, -\mathbf{R}') [h^0(\mathbf{R}') + \mathbf{E}(t) \cdot \mathbf{r}(\mathbf{R}'), \rho^P(\mathbf{R} - \mathbf{R}')] \\ & + \sum_{\mathbf{R}'} \phi_P(t, -\mathbf{R}') [\Sigma^{\text{HSEX}}[\rho^L](\mathbf{R}') \\ & - \Sigma_0(\mathbf{R}'), \rho^P(\mathbf{R} - \mathbf{R}')] \\ & + i\phi_P(t, -\mathbf{R}) \mathcal{L}_{\text{incoh}}[\rho^L](\mathbf{R}), \end{aligned} \quad (44)$$

where we can see that the term $\mathbf{E}(t) \cdot \mathbf{R}\rho^L(\mathbf{R})$ is no longer involved in the propagation, as it is encoded in the time-dependent Peierls phase, ϕ_P . Equation (44) is what we call the semiconductor Wannier equations, a real-space equivalent of the semiconductor Bloch equations. This is our preferred formulation of the equations of motion. Up to this point, the problematic transition dipole moments in the reciprocal-space states did not appear in any of our equations and therefore we avoid the problem of the structure-gauge freedom. This is one of key features that makes the SWEs more suitable than the SBEs

3. Periodic boundary conditions

The equations of motion for ρ^P [Eq. (44)] are defined for an infinite crystal and for a practical implementation one must truncate our sums over $\mathbf{R}$ to a finite set of lattice vectors. The most common choice is to work in a supercell approach. For that, we can define the supercell Bravais lattice as

$$\mathbf{R}_S = \sum_{i=1}^D d_i \mathbf{a}_{S,i}, \quad d_i \in \mathbb{Z}, \quad (45)$$

where $\mathbf{a}_{S,i} = N_i \mathbf{a}_i$ are the primitive lattice vectors of the supercell and $N = \prod_{i=1}^D N_i$ is the number of unit cells in the supercell. Furthermore, we impose Born-von Karman periodic boundary conditions, i.e., $c_{\mathbf{R}\alpha}^\dagger \equiv c_{(\mathbf{R}+\mathbf{R}_S)\alpha}^\dagger$. The Bravais lattice vectors enclosed into the origin supercell are given by

$$\mathbf{R} = \sum_{i=1}^D n_i \mathbf{a}_i, \quad n_i \in [-N_i/2, N_i/2 - 1]. \quad (46)$$

From now on, the infinite sums in $\mathbf{R}$ will be replaced by a sum of Bravais vectors in a single supercell, according to Eq. (46). The choice of periodic boundary conditions leads to a discretization of our Brillouin zone, i.e.,

$$\mathbf{k} = \sum_{i=1}^D \frac{m_i}{N_i} \mathbf{b}_i, \quad m_i \in [0, N_i - 1], \quad (47)$$

which defines a Monkhorst-Pack reciprocal-space grid [67]. In the following, all the sums over $\mathbf{k}$ will be assumed to be done on this reciprocal-space grid.

We can define the Bloch states associated with the localized Wannier orbitals as

$$|\psi_{\mathbf{k}\alpha}^W\rangle = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}} |\mathbf{R}\alpha\rangle, \quad (48)$$

$$c_{\mathbf{k}\alpha}^{\dagger, W} = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}} c_{\mathbf{R}\alpha}^\dagger, \quad (49)$$

$$c_{\mathbf{R}\alpha}^\dagger = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}} c_{\mathbf{k}\alpha}^{\dagger, W}, \quad (50)$$

which are constructed from the localized Wannier orbitals. The superscript W refers to the Wannier structure gauge [31,64,68]. It is important to distinguish between two distinct contexts in which the term gauge is used: the *laser gauge*, referring to the choice between velocity and length gauges in the light-matter interaction, and the structure gauge, associated with the inherent freedom in selecting a Bloch basis for the crystal Hamiltonian. In the W gauge, we can also define the reduced density matrix in the reciprocal space, $\rho_{\alpha\beta}^W(\mathbf{k}) = \langle c_{\mathbf{k}\beta}^{\dagger, W} c_{\mathbf{k}\alpha}^W \rangle$. This RDM is related with $\rho^L(\mathbf{R})$ by a simple discrete Fourier transform

$$\rho^W(\mathbf{k}) = \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}} \rho^L(\mathbf{R}), \quad (51)$$

$$\rho^L(\mathbf{R}) = \frac{1}{N} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}} \rho^W(\mathbf{k}). \quad (52)$$

It is useful to define

$$\mathcal{F}^{L \rightarrow W}[\rho^L(\mathbf{R})] \equiv \rho^W(\mathbf{k}) \quad (53)$$

$$= \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}} \rho^L(\mathbf{R}) \quad (54)$$

and the corresponding inverse transformation, $\mathcal{F}^{W \rightarrow L}$.

4. Initial state

In order to obtain the initial state, we must diagonalize $\hat{h}^0$. In the basis, $|\psi_{\mathbf{k}\alpha}^W\rangle$, $\hat{h}^0$ is expressed as

$$\hat{h}^0 = \sum_{\alpha\beta} h_{\alpha\beta}^{0,W}(\mathbf{k}) c_{\alpha\mathbf{k}}^{\dagger, W} c_{\beta\mathbf{k}}^W, \quad (55)$$

$$h_{\alpha\beta}^{0,W}(\mathbf{k}) = \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}} h_{\alpha\beta}^0(\mathbf{R}), \quad (56)$$

and by diagonalizing $h_{\alpha\beta}^{0,W}(\mathbf{k})$, we have

$$\hat{h}^0 = \sum_{kn} \varepsilon_n(\mathbf{k}) c_{n\mathbf{k}}^{\dagger, H} c_{n\mathbf{k}}^H, \quad (57)$$

$$h_{nm}^{0,H}(\mathbf{k}) = \delta_{nm} \varepsilon_n(\mathbf{k}), \quad (58)$$

$$h^{0,H}(\mathbf{k}) = U^\dagger(\mathbf{k}) h^{0,W}(\mathbf{k}) U(\mathbf{k}), \quad (59)$$

where $U(\mathbf{k})$ is the unitary matrix that diagonalizes $h^{0,W}(\mathbf{k})$, the superscript H refers to the Hamiltonian structure gauge, and $\varepsilon_n(\mathbf{k})$ is the energy of band n . The equilibrium RDM in the Hamiltonian gauge is just

$$\rho_{nm}^{0,H}(\mathbf{k}) = \delta_{nm} F_{\mu,T}(\varepsilon_n(\mathbf{k})), \quad (60)$$

where $F_{\mu,T}(\epsilon) = (e^{\beta(\epsilon-\mu)} + 1)^{-1}$ is the Fermi-Dirac function for a chemical potential μ and inverse temperature β . It is useful to define

$$\mathcal{U}^{W \rightarrow H}[\rho^W(\mathbf{k})] \equiv \rho^H(\mathbf{k}) \quad (61)$$

$$= U^\dagger(\mathbf{k})\rho^W(\mathbf{k})U(\mathbf{k}) \quad (62)$$

and the corresponding inverse transformation, $\mathcal{U}^{H \rightarrow W}$. Note that the choice of $U(\mathbf{k})$ is not unique. This fact stems from the structure-gauge freedom to choose the eigenstates, i.e., $|\psi_{kn}^W\rangle \rightarrow e^{i\theta_n(\mathbf{k})}|\psi_{kn}^W\rangle$. However, one must notice that $\rho^{0,W}(\mathbf{k})$ remains unaltered by this phase transformation. The $\mathcal{U}^{W \rightarrow H}$ transformation will allow to have access to populations of bands and to compute terms that are inherent to the H gauge, such as, interband and intraband currents, and the inclusion of the pure dephasing term, $\mathcal{L}_D$.

5. Observables

The velocity operator, in contrast to the position operator, is invariant under lattice translations. For a general invariant operator under lattice translations, $\hat{O}$, its mean value normalized to the unit cell can be expressed as

$$\begin{aligned} \frac{\langle O \rangle}{N} &= \sum_{\alpha\beta\mathbf{R}} O_{\alpha\beta}(\mathbf{R})\rho_{\beta\alpha}(-\mathbf{R}) \\ &= \sum_{\alpha\beta\mathbf{R}} O_{\alpha\beta}(\mathbf{R})(\rho_{\alpha\beta}(\mathbf{R}))^*. \end{aligned} \quad (63)$$

The explicit expression for the velocity operator matrix elements in real space is

$$\mathbf{v}(\mathbf{R}) = i \left(\mathbf{R}h^0(\mathbf{R}) - \sum_{\mathbf{R}'} [\mathbf{r}(\mathbf{R}'), h^0(\mathbf{R} - \mathbf{R}')] \right). \quad (64)$$

We can recognize in the first (second) term of the velocity operator the intraband (interband) term expressed in a real-space basis [31]. We note that Berry-curvature effects are not neglected in the present formulation. Although Eq. (64) is written in real space, its Fourier transform yields the standard multiband velocity operator in reciprocal space [31]. The expression for the total current is, according to Eq. (63),

$$\mathbf{j}(t) = e \frac{\langle \mathbf{v} \rangle}{N} = e \sum_{\alpha\beta\mathbf{R}} \mathbf{v}_{\alpha\beta}(\mathbf{R})(\rho_{\alpha\beta}(\mathbf{R}))^*. \quad (65)$$

We must emphasize that observables must be calculated with ρ^L , by performing the transformation defined in Eq. (42), as the corresponding operators do not take into account the Peierls phase.

6. Decoherence

In Eq. (44), we did not show the explicit form of the incoherent terms that were grouped in $\mathcal{L}_{\text{incoh}}[\rho^L]$. The first thing to notice is that we should move to the L gauge. In our work, as discussed in Sec. II A 5, we include three forms of decoherence. The pure dephasing term, $\mathcal{L}_D$, is naturally expressed in the H gauge. By applying $\mathcal{F}^{L \rightarrow W}$ and $\mathcal{U}^{W \rightarrow H}$, one moves ρ to the H gauge, where $\mathcal{L}_D$ is simply

$$\mathcal{L}_D[\rho^H]_{ij}(\mathbf{k}) = \gamma_D(\delta_{ij} - 1)\rho_{ij}^H(\mathbf{k}), \quad (66)$$

and after this apply the corresponding inverse transformations, $\mathcal{U}^{H \rightarrow W}$ and $\mathcal{F}^{W \rightarrow L}$.

The relaxation term, $\mathcal{L}_r$, is simply expressed as

$$\mathcal{L}_r[\rho^L](\mathbf{R}) = \gamma_r(\rho^{0,L}(\mathbf{R}) - \rho^L(\mathbf{R})). \quad (67)$$

Regarding the real-space dephasing term,

$$\mathcal{L}_{\text{rs}}[\rho^L](\mathbf{R}) = \gamma_{\text{rs}}(d(\mathbf{R})) \odot (\rho^{0,L}(\mathbf{R}) - \rho^L(\mathbf{R})), \quad (68)$$

where $d_{\alpha\beta}(\mathbf{R}) = |\boldsymbol{\tau}_\alpha - \boldsymbol{\tau}_\beta - \mathbf{R}|$, $\boldsymbol{\tau}_\alpha$ is the center of the α Wannier function in the center unit cell, and $\odot$ denotes the Hadamard product. The functional form of γ_{rs} is given by Eq. (29). Effectively, with this term, we are suppressing coherences between orbitals that are separated by distances greater than $R_{\text{rs,cut}}$ and can therefore suppress trajectories that acquire a phase larger than 2π and destructively interfere in the far-field [9,69]. Having this analogy in mind, it will be useful to express $R_{\text{rs,cut}}$ in units of the excursion of an electron that acquires roughly a 2π phase, i.e., $R_{\text{rs},0} = 4\pi\omega_L/(|e|E_0)$, where E_0 and ω_L are, respectively, the amplitude and frequency of the laser field.

III. RESULTS

In this section, we show numerical results that were obtained with the SWE formalism. We first show the behavior of the equilibrium reduced density matrix and its decay. We also calculate the linear optical conductivity for monolayer hBN and monolayer MoS₂, the convergence of the SWEs with respect to the formulation of the SBEs in Ref. [31], the impact of pure and real-space dephasing in the HHG spectrum, and finally the HHG spectrum for MoS₂.

A. Decay of equilibrium density matrix

The basic object in the SWEs is the electronic reduced density matrix expressed in a basis of Wannier localized orbitals. Despite working with extended periodic systems, where Bloch states are delocalized, the coherences of the electronic density matrix decay rapidly with the distance between orbitals. *Nearsightedness* of electronic matter [70,71] is actually the core assumption for linear scaling electronic structure methods [72,73].

In Fig. 1, we show the decay of coherences in the density matrix for graphene, at zero and finite temperature ($T = 1000$ K), and monolayer hBN. For both cases, we model graphene and monolayer hBN with a tight-binding model [74], with a nearest neighbor hopping $t_0 = 2.8$ eV and a distance between neighboring atoms of 1.4457 Å. The gap energy in the monolayer hBN is set to be 4.52 eV. As expected, hBN (a gapped insulator) and graphene at finite temperature (a finite temperature semimetal) exhibit an exponential decay of the coherences, whereas graphene at zero temperature displays an algebraic decay [72,75]. It is the nearsightedness of electrons, depicted in Fig. 1, that make a real-space approach a compelling alternative to the reciprocal-space methods.

B. Linear optical conductivity

To validate the SWE approach, we have calculated the linear optical conductivity of hBN and MoS₂. In both cases, we have a point group symmetry D_{3h} and for symmetry

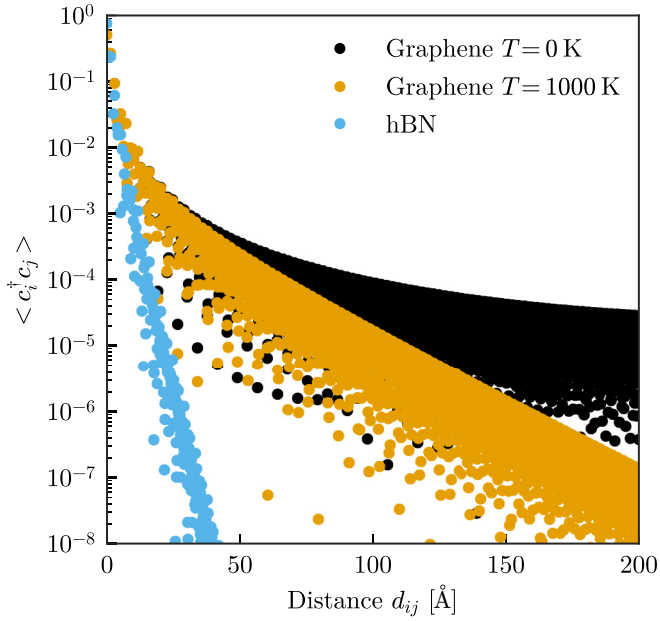


FIG. 1. Scatter plot of the coherences between localized orbitals for monolayer hBN (blue circles) and graphene, at zero (black circles) and finite temperature (orange circles).

reasons, $\sigma_{xy} = 0$ and $\sigma_{xx} = \sigma_{yy}$ [37]. We calculate the linear optical conductivity by applying a very short pulse,

with a field strength in the linear response regime, $E_0 = 2$ kV/m, and an FWHM in electric field of 0.157 fs for a $\cos^2$ envelope. The linear optical conductivity can be obtained by using Ohm's law, $\sigma(\omega) = \tilde{j}(\omega)/\tilde{E}(\omega)A_{UC}$, where $\tilde{j}(\omega) = \int_{-\infty}^{+\infty} dt \exp(-i\omega t)j(t)$ and A_{UC} is the unit-cell area. To dampen the signal, we introduce a relaxation parameter of $\hbar\gamma_r = 0.1$ eV. To model the screening potential, we used the Rytova-Keldysh potential (see Appendix), and used for the monolayer hBN (MoS₂) a screening length $r_0 = 10$ Å ($r_0 = 13.55$ Å) and set the dielectric constants as $(\epsilon_1 + \epsilon_2)/2 = 1$ ($(\epsilon_1 + \epsilon_2)/2 = 2.5$). The Rytova-Keldysh screening potential used for both hBN and MoS₂ is taken from the literature [76]. In both cases, we used a supercell with $N_1 = N_2 = 192$. The band structure of MoS₂ was obtained by performing an *ab initio* calculation using the HSE06 functional [77], including spin-orbital coupling, in a 16×16 Monkhorst-Pack grid using the QUANTUMESPRESSO code [78]. We perform a projection on the *p* orbitals of S and *d* orbitals of Mo, and a Wannierization procedure to obtain the Hamiltonian and dipole couplings using the WANNIER90 software [79], obtaining 22 bands to model MoS₂.

In Fig. 2, we show in panels (a) and (b) the band structure and density of states (DOS) for hBN and MoS₂. In panels (c) and (d), we plot the linear optical conductivity for hBN and MoS₂ in the independent-particle approximation (IPA), i.e., setting $\Sigma^{HSEX} = 0$, and in the TD-HSEX approximation. First, our results match very well previous studies [43,76],

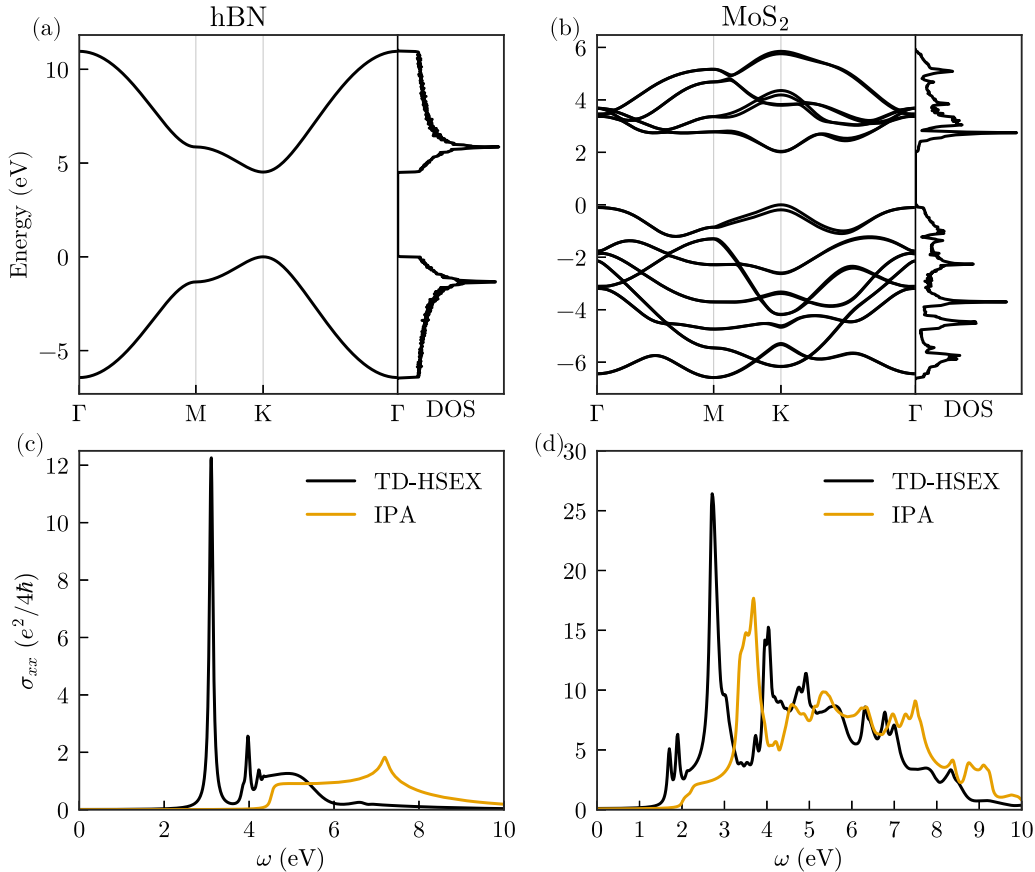


FIG. 2. Band structure and total DOS of monolayer hBN [panel (a)] and of monolayer 1H-MoS₂ [panel (b)]. In panels (c) and (d), we show the linear optical conductivity of monolayer hBN and 1H-MoS₂, respectively, in the IPA (orange lines) and TD-HSEX (black lines) approximations.

both in the IPA and TD-HSEX approximations. Furthermore, the TD-HSEX is able to capture correctly optical excitons in hBN and MoS₂.

C. Convergence: SWEs versus SBEs

We benchmarked the numerical convergence of the SBEs in momentum space, using the formalism of Ref. [31], against that of the SWEs in real space by calculating the current generated in monolayer hBN under illumination by a midinfrared laser pulse. The laser field is modeled as

$$\mathbf{E}(t) = E_0 f(t) \sin(\omega t + \phi) \hat{\mathbf{e}}, \quad (69)$$

where $E_0 = 40$ MV/cm polarized in the Γ -M direction (along the N-B bond), with a central wavelength of 3 μm and a carrier envelope phase $\phi = \pi/2$. The pulse envelope $f(t)$ is taken to be a $\cos^2$ profile with an FWHM in electric field of 47.1 fs. We introduce in the calculations pure dephasing with $T_2 = 5$ fs. For both cases, we integrate the equations of motion using a Runge-Kutta 4 propagator with a time step of $dt = 0.1$ a.u.

In Fig. 3, we show the calculated current in the SWE formalism [panels (a) and (b)] and in the SBE formalism [panels (c) and (d)]. For the SWEs, we calculate the current using different supercells, where $N_1 = N_2 = N_R$, and we take as reference a calculation with $N_R = 600$. In the SBEs, we discretize the reciprocal space in a $N_k \times N_k$ Monkhorst-Pack grid and we take as reference a calculation with $N_k = 1200$. We can see in Fig. 3(b) that the relative error decreases quite fast with a moderate N_R , noticing that with $N_R = 400$ we reach numerical precision accuracy. On the other hand, the SBEs converge much slower when compared with the SWEs [see Figs. 3(b) and 3(d)]. To achieve a relative error of around 10^{-3} , a supercell with $N_R = 50$ is sufficient. However, in the SBE case, we need to increase the k -space grid to $N_k = 600$. The slower convergence of the SBE calculations originates from the finite-difference discretization of the k derivative [31], which is avoided in the SWE formulation. One additional advantage of the SWE over the SBE approach is numerical: Convergence is achieved significantly faster, enabling the exploration of more complex materials that would otherwise be computationally prohibitive within the SBE framework.

D. Real-space dephasing

The requirement for ultrashort dephasing times to match experimental high-harmonic spectra in solids remains an open question. A recently proposed method seeks to address this issue by introducing real-space dephasing [9], which arises naturally within the SWE framework.

Figure 4 compares the two approaches by calculating the high-harmonic spectrum in hBN while scanning $R_{\text{rs,cut}}$ for the real-space dephasing case and T_2 for the pure-dephasing case. For real-space dephasing, we set $\alpha_{\text{rs}} = 2$ and $\beta_{\text{rs}} = 6.7736 \times 10^{-5}$ a.u. The laser field is the same as in Fig. 3. In these calculations, we used a supercell with $N_1 = N_2 = 100$ and propagate the SWEs with time step $dt = 0.5$ a.u. using a Dormand-Prince's 5/4 Runge-Kutta method. The harmonic spectrum is calculated as the square of the Fourier transform of the dipole acceleration [29], $\mathbf{a}(t) = \frac{d\mathbf{j}(t)}{dt}$. To avoid artificial

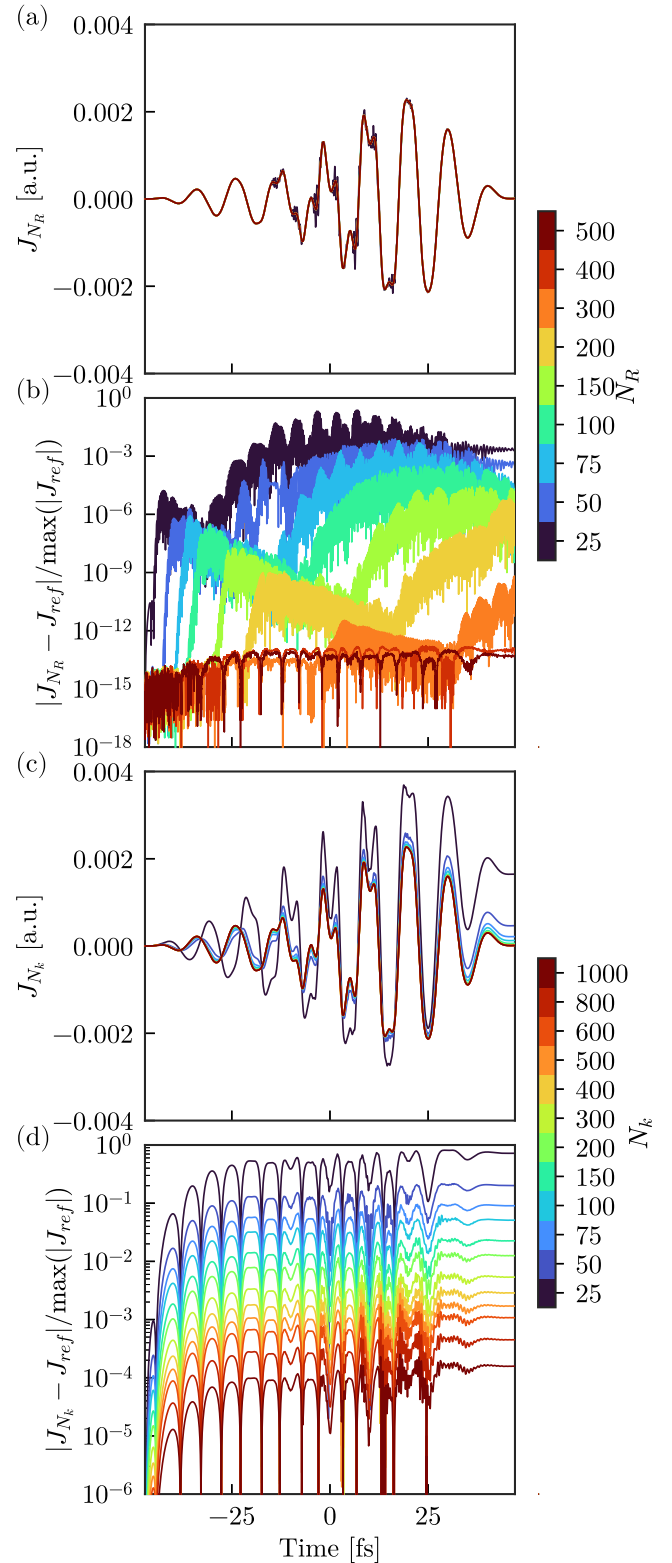


FIG. 3. Calculated current [panel (a)] and the relative error of the current [panel (b)] using the SWE approach for different N_R . (c), (d) The same for calculations using the SBE approach.

spectral features caused by abrupt discontinuities at the end of the pulse, a window function is applied to the current prior to the Fourier transform [80].

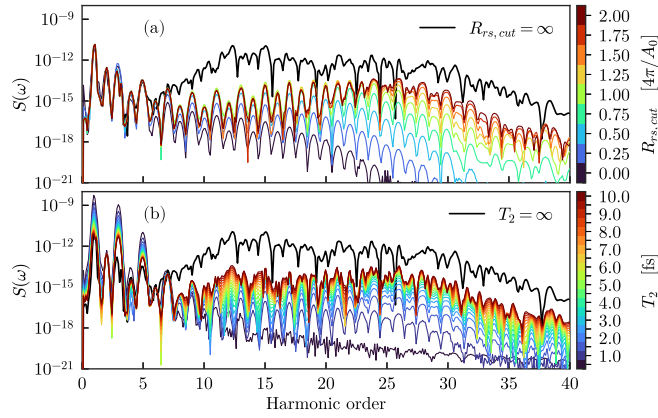


FIG. 4. High-harmonic spectrum of monolayer hBN with different dephasing mechanisms: (a) real-space dephasing and (b) pure dephasing.

Both real-space and pure dephasing reduce interferences in the high-harmonic signal. However, real-space dephasing preserves the intensity of low-order harmonics, matching the purely coherent case. In contrast, pure dephasing strongly enhances low-order harmonics, an artifact of the extremely short dephasing times, primarily caused by dephasing-induced ionization [81]. Conventional T_2 -type dephasing enhances the conduction-band population through dephasing-induced excitation, thereby increasing the low-order harmonics, whereas real-space dephasing largely preserves the short trajectories responsible for this spectral region [82]. For these reasons, the real-space dephasing approach is a more reliable and physically consistent alternative to simple pure-dephasing models.

E. HHG in MoS₂

We have calculated the HHG spectra in MoS₂. All parameters of the laser pulse are the same as in Fig. 3, apart from the peak field, which is $E_0 = 30$ MV/cm. We introduce a pure-dephasing term, $T_2 = 4$ fs. In these calculations, we used a supercell with $N_1 = N_2 = 100$ and propagate the SWEs with time step $dt = 0.5$ a.u. using a Dormand-Prince's 5/4 Runge-Kutta method.

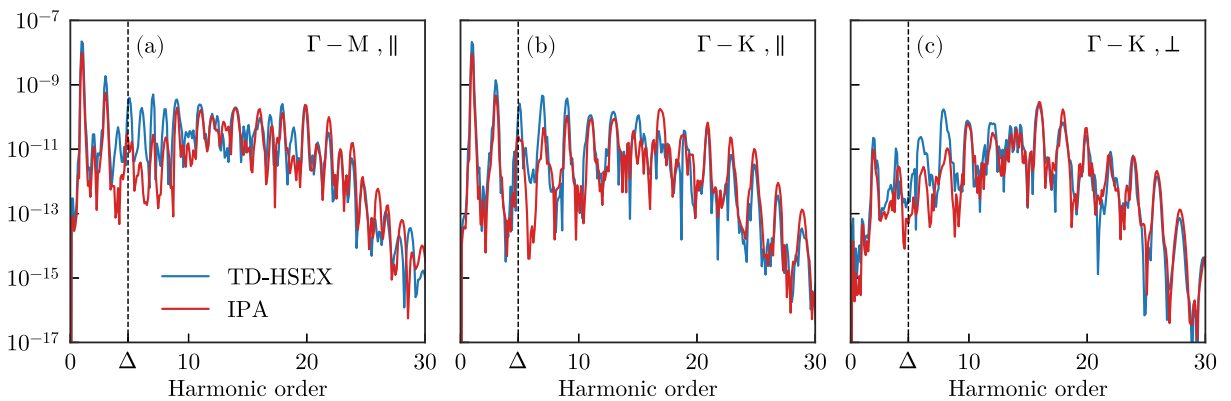


FIG. 5. High-harmonic spectrum at the IPA and TD-HSEX level in MoS₂. (a) HHG spectrum for a laser polarized along the Γ -M direction in the parallel direction. (b), (c) Same as in panel (a) for a laser along the Γ -K in the parallel and perpendicular directions, respectively. The dashed vertical line corresponds to the band gap of MoS₂.

Figure 5 shows the harmonic spectra for different laser polarization and emission directions. Symmetry requires that for a laser pulse oriented along the Γ -M direction, the perpendicular current vanishes and is therefore not displayed. In contrast, for emission parallel to the pulse in this direction, both even and odd harmonics are allowed because inversion symmetry is broken. When the laser pulse is aligned along the Γ -K direction, symmetry constrains the harmonic emission: only odd (even) harmonics appear for parallel (perpendicular) emission [83]. We observe that when excitonic effects are taken into account (TD-HSEX), we see an enhancement of the harmonic signal in harmonics below the band gap (Δ), which is consistent with previous findings in the literature [11].

IV. CONCLUSIONS

We have introduced the semiconductor Wannier equations, a real-time, real-space framework for describing ultrafast light-matter interaction and nonlinear optical response in crystalline solids. By expressing the electronic reduced density matrix in a localized Wannier basis, the SWEs provide a gauge-clean alternative to reciprocal-space SBEs, avoiding the numerical instabilities caused by the structure-gauge freedom of Bloch states. The method naturally incorporates electron-electron interactions at the TD-HSEX level and includes physically motivated decoherence mechanisms, pure dephasing, population relaxation, and distance-dependent real-space dephasing, offering a robust modeling of strong-field and high-harmonic generation dynamics.

We have shown that the SWEs reproduce key optical properties, such as the linear optical conductivity and excitonic features of monolayer hBN and MoS₂, in agreement with established approaches. Benchmarking against the SBE formalism demonstrated that the SWEs achieve significantly faster numerical convergence, enabling efficient simulations with moderate supercell sizes. Importantly, the real-space dephasing model introduced here provides a physically grounded alternative to the ultrashort pure-dephasing times often used to fit experimental HHG spectra, avoiding artificial enhancement of low-order harmonics while preserving the coherent features of the signal.

Conceptually, the SWEs bridge semiclassical real-space intuition, central to attosecond physics, with many-body solid-state optics, opening avenues for interpreting HHG and other strong-field phenomena in complex materials. Their computational efficiency, robustness to gauge ambiguities, and compatibility with *ab initio* Hamiltonians make the SWEs a promising platform for exploring nonlinear optical response, excitonic dynamics, and attosecond spectroscopy in emerging quantum materials.

Future developments may include the incorporation of fully dynamical screening, coupling to phonons and other bosonic modes, and extensions to strongly correlated systems beyond mean-field approximations. These advances will further strengthen the SWEs as a versatile and predictive tool for next-generation ultrafast solid-state optics.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

APPENDIX: RYTOVA-KELDYSH POTENTIAL

As we are dealing with two-dimensional materials, the Rytova-Keldysh potential [84–86] is our choice to model screening effects in the self-energy term, Σ^{SEX} . The Rytova-Keldysh potential reads

$$W_{\text{RK}}(r) = \frac{e^2}{4\epsilon_0(\epsilon_1 + \epsilon_2)r_0} \left[H_0\left(\frac{r}{r_0}\right) - Y_0\left(\frac{r}{r_0}\right) \right], \quad (\text{A1})$$

where ϵ_1 and ϵ_2 are the dielectric constants of the top and bottom medium, r_0 is the screening length, and H_0 , Y_0 are the zero-order Struve and Neumann special functions. In order to avoid the divergence of the potential at $r = 0$ [87,88], we renormalize $r \rightarrow \sqrt{r^2 + r_{\text{min}}^2}$. Furthermore, for large distances, i.e., $r > r_{\text{RK,cut}}$, we employ a radial cutoff and take W_{RK} to be zero. In practice, to avoid artifacts from periodic images in the calculation, we will set $r_{\text{RK,cut}}$ to be of the radius of the largest sphere that fits inside the supercell. The renormalization and radial cutoff are also applied to the bare Coulomb potential in the Hartree term, Σ^H .

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