

# Efficient time-dependent Wannier functions for ultrafast dynamics

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Time-dependent Wannier functions were initially proposed as a means for calculating the polarization current in crystals driven by external fields. In this work, we present a simple gauge where Wannier states are defined based on the maximally localized functions at the initial time, and are propagated using the time-dependent Bloch states obtained from established first-principles calculations, avoiding the costly Wannierization at each time step. We show that this basis efficiently describes the time-dependent polarization of the laser-driven system through the analysis of the motion of Wannier centers. We use this technique to analyze highly nonlinear and nonperturbative responses such as high-harmonic generation in solids, using hexagonal boron nitride as an illustrative example, and we show how it provides an intuitive picture for the physical mechanisms.

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## I. INTRODUCTION

Wannier functions [1,2] form a single-particle orthonormal basis in solid-state crystals that are widely used as an alternative to Bloch orbitals. Especially, maximally localized Wannier functions (MLWFs) have proven to be of great practical use, because they give chemical intuition to first-principles numerical results of static and thermodynamic properties [2]. In this representation one can visualize properties typically studied in molecular systems, such as hybridization, covalency, and ionicity, in solid-state crystals [3–5], and provide the basis for the implementation of the modern theory of polarization [6–8]. Wannier functions also play an important role in the formulation and evaluation of response properties because of their close relation to polarization [9], Born effective charges [10], Berry phase and curvature [11], etc., making them particularly useful in describing various topology-related properties. Wannier functions have been expanded to perturbation theory [12], Floquet theory [13], and time propagation [14,15] in order to obtain localized basis representations in each of these domains. They have also been used as interpolators to extract electron-phonon coupling [16–19].

MLWFs are widely used as a convenient and well-defined orbital basis to derive tight-binding models of materials from first principles. These models are especially useful in analyzing

complex materials such as highly correlated systems [20], because they allow the reduction of the correlated problem to a set of few active atomic orbitals per site, for instance through methods like constrained random-phase approximation (RPA) [21]. To describe dynamical properties in such systems one normally propagates these lattice models built on top of a static MLWF basis set. This requires in principle the Wannierization of all unoccupied states into which the system evolves, relying on prior intuition of the dynamic processes involved and often entailing increased computational cost.

Another proposed approach is to make the Wannier functions themselves time dependent and calculate the observables directly in this basis [14]. Despite the early recognition of this application, Wannier function propagation has not been systematically pursued as a practical and efficient computational strategy and the precise definition has not yet been explored. The one notable case is an approach where the Wannierization is done with the constraint to maximally localize the states at each time step of the out-of-equilibrium driven system [15,22]. This approach showed good agreements between the electronic polarization and experimental absorption spectra [15], and it retained the intuitive chemical representation in that process. However, besides the high computational cost associated with maximally localizing the Wannier functions at each time step, there is also a conceptual issue in that this basis set can have discontinuities in time as the global minima of the MLWFs change, leading to discontinuous derived observables [15].

The most direct application of time-dependent Wannier functions is to study the dynamics of the dielectric polarizations and its current, particularly beyond the regime of response theory, for instance high-order harmonic generation (HHG) in solids which is not accessible via perturbation theory [23–25]. With the advances in ultrafast lasers, HHG in solid-state materials has been studied both experimentally and computationally. One common calculation method is using tight-binding models [26–34], but in order to fully capture the

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dynamics, particularly the interband dynamics, these models need to be expanded to include the unoccupied bands [35], increasing the computational cost. MLWFs have been shown to be especially useful for analyzing HHG, but the basis set here is static corresponding to the ground-state system [36–41], unlike other useful basis sets which evolve with the Hamiltonian, like Houston [25,42–45] or Floquet-Bloch states [46–52]. On the other hand, first-principles methods, like real-time time-dependent density-functional theory (TDDFT) [53] incorporate all of these dynamics within the occupied orbitals only [54–60].

Here we propose and numerically study a definition of the time-dependent Wannier functions, where the Wannierization step is done only once on the ground state (i.e., at  $t = 0$ ), and the time-dependent Wannier states are determined by the time-dependent Bloch states, which we take from established first-principles calculations. We use this definition to study HHG in both noninteracting model system and *ab initio* in hexagonal boron nitride (h-BN). We show that with this definition, the current can be described purely from the motion of Wannier centers in a wide range of laser-driving profiles, providing an intuitive analog for the dipole acceleration in finite systems but applied to periodic solids. Our work therefore shows that the time evolution of Wannier states is able to incorporate ultrafast electron dynamics within a far smaller basis set compared to standard schemes, which can be highly useful for obtaining mechanistic physical insight to nonlinear phenomena.

## II. TIME-DEPENDENT WANNIER FUNCTION DEFINITIONS

We consider time-dependent Wannier functions of the following form:

$$\Phi_{n\mathbf{R}}(\mathbf{r}, t) = \sum_i \int_{\text{BZ}} U_{i\mathbf{k}}^{n\mathbf{R}} f_{i\mathbf{k}}(t) \Psi_{i\mathbf{k}}(\mathbf{r}, t) d^3\mathbf{k}, \quad (1)$$

where  $\Phi$  are the time-dependent Wannier functions,  $\Psi$  are the time-dependent Bloch states,  $U$  is the Wannier transformation matrix, which is calculated only once at the ground state to give the MLWF, and  $f$  is a time-dependent phase-compensation function that we introduce in order to retain the localized nature of the Wannier functions. Note that one could also recompute  $U$  over time, but this would not be numerically efficient or guarantee a continuous evolution of the time-dependent Wannier states. Moreover, it is not clear that maximally localized states are an ideal basis set for analyzing out-of-equilibrium systems, where by definition electrons must hop between sites (i.e. have non-localized nature for finite times). Instead, we here express the time dependence of the Wannier functions via the phase factor  $f$  and time-dependent Bloch states  $\Psi$  only, which does not result in maximally localized functions, but does guarantee continuous evolution while still maintaining a high degree of localization. We explore three such phase-compensation functions:

$$f_{i\mathbf{k}}^{\text{none}}(t) = 1 \quad \forall i, \mathbf{k}, t, \quad (2)$$

$$f_{i\mathbf{k}}^{\text{static}}(t) = e^{i(1/\hbar)\epsilon_{i\mathbf{k}}^0 t}, \quad (3)$$

$$f_{i\mathbf{k}}^{\text{int}}(t) = e^{i(1/\hbar)\int_0^t \epsilon_{i\mathbf{k}}(\tau) d\tau}, \quad (4)$$

where we either do not perform any phase compensation [Eq. (2)], compensate for the static band energy  $\epsilon^0$  [Eq. (3)], or compensate for the instantaneous energy expectation value [Eq. (4)].

$$\epsilon_{i\mathbf{k}}(t) = \langle \Psi_{i\mathbf{k}}(t) | \hat{H}(t) | \Psi_{i\mathbf{k}}(t) \rangle. \quad (5)$$

The reasoning behind these choices is to explore the scope and importance of such phase compensation, but the choice of the form of these functions is arbitrary and chosen out of physical intuition. We expect that other effective gauges exist, and currently we do not have a proof for why this choice should be preferred, other than intuition, and it upholding all of the proper physical limits. The  $f^{\text{static}}$  is the trivial initial state, or field-free, evolution, while  $f^{\text{int}}$  takes the driving field into account, via the expectation value of the time-dependent Hamiltonian  $\langle \hat{H}(t) \rangle$ , as well as changes in the character of the Bloch states during time evolution.

Numerically, we have computed Eq. (4) at each time step of the simulation using a simple Euler integration method:

$$f_{i\mathbf{k}}^{\text{int}}(t_n) = f_{i\mathbf{k}}^{\text{int}}(t_{n-1}) e^{i(1/\hbar)\epsilon_{i\mathbf{k}}(t_n)\delta t}. \quad (6)$$

The form of Eq. (1) guarantees that the Wannier functions remain unitary at each time step independent of the phase factor  $f$ .

To analyze the differences of these definitions, we look at the evolution of the Wannier spread  $\Omega(t)$ :

$$\Omega_n(t) = \int_V \mathbf{r}^2 |\Phi_{n\mathbf{R}}(\mathbf{r}, t)|^2 d^3\mathbf{r} - |\mathbf{r}_{n\mathbf{R}}(t)|^2, \quad (7)$$

$$\mathbf{r}_{n\mathbf{R}}(t) = \int_V \mathbf{r} |\Phi_{n\mathbf{R}}(\mathbf{r}, t)|^2 d^3\mathbf{r}, \quad (8)$$

where we denote here  $\mathbf{r}_{n\mathbf{R}}(t)$  to be the time-dependent Wannier center.

We investigate the different time-dependent Wannier functions first by considering a noninteracting 1D electron system under a Mathieu potential with or without an external laser interaction  $\mathbf{A}(t)$ :

$$\hat{H}(t) = \frac{1}{2m} [\hat{\mathbf{p}} - e\mathbf{A}(t)]^2 + \hat{V}, \quad (9)$$

$$V(x) = -V_0[1 + \cos(2\pi x/a)]. \quad (10)$$

Here, we choose well-studied parameters for the Mathieu potential  $V_0 = 0.37(E_h)$ ,  $a_0 = 8(a_0)$ , resulting in the static band structure shown in Fig. 1(a). In this model we take the first two bands to be fully occupied, and for this section we focus on the dynamics of these valence bands and the evolution of their corresponding Wannier function.

We first consider the stationary ground-state case

$$\mathbf{A}(t) = 0 \quad \forall t, \quad (11)$$

and let both the system and Eq. (1) evolve coherently in time. By definition, all observables are time independent and in the ground state, as the system evolves with only a pure global phase that cancels out when evaluating observables. But, the Wannier function basis from Eq. (1) is still time dependent as it is obtained by summing Bloch states that coherently interfere due to the different phase propagations of the band eigenvalues of the Bloch states [Fig. 1(a)]. This effect is evident in Fig. 1(b) when evaluating the Wannier

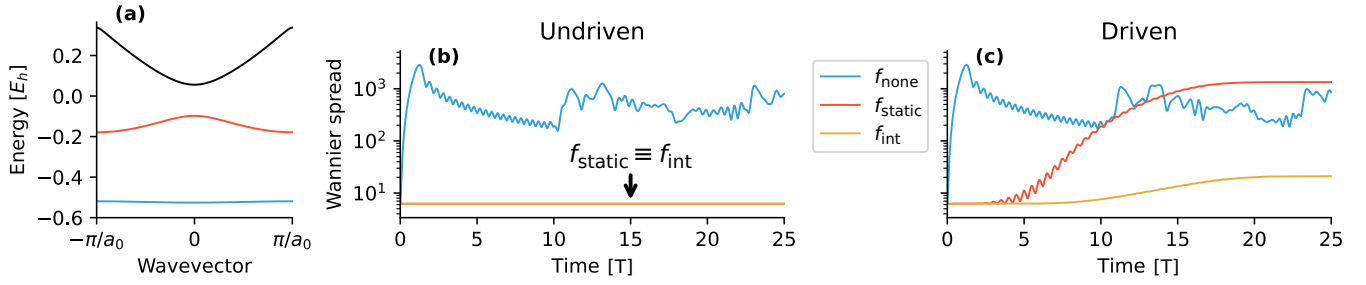


FIG. 1. Noninteracting 1D electron system under Mathieu potential. (a) Ground-state band structure. (b), (c) Wannier spread evolution without and with laser perturbation, respectively. The band gap in (b) measures to  $\approx 0.154(E_h)$ .

spread  $\Omega$  without external perturbation. Wannier functions  $f^{\text{none}}$  without phase compensation (blue line) lose localization very fast, with a timescale matching roughly the band energy bandwidth. This trivial dephasing effect is exactly compensated by the other two definitions (red and yellow lines) that show a spread that is independent of time. In Fig. 1(b), the red and yellow lines are on top of each other because by definition they are equivalent ( $f^{\text{static}} \equiv f^{\text{int}}$ ) when there is no time-dependent perturbation. This behavior is the main motivation for the introduction of the phase-compensation term  $f$  in our definition of the time-dependent Wannier functions [Eq. (1)].

Next, we consider the strongly driven nonperturbative case with a supersine laser envelope [61]:

$$A_x(t) = A_0 \sin\left(\frac{\pi t}{n_{\text{cyc}} T}\right)^{\frac{\pi}{\sigma}} \left| \frac{t}{n_{\text{cyc}} T} - \frac{1}{2} \right| \sin\left(\frac{2\pi t}{T}\right), \quad (12)$$

where we use the standard notations for the laser period  $T$ , and wavelength  $\lambda$ :

$$T = \frac{\lambda}{c}. \quad (13)$$

Here, we look at the case with a peak laser intensity of  $I = 10^{11}$  W/cm<sup>2</sup>, a wavelength of  $\lambda = 1500$  nm, corresponding to an energy of  $\sim 0.198$  times the direct band gap, and a pulse duration of  $n_{\text{cyc}} = 25$  cycles. The Wannier spread evolutions in this case are presented by Fig. 1(c). The uncompensated evolution of the Wannier function ( $f^{\text{none}}$ , blue line) shows no significant difference from the undriven case, and we can disregard it as a practical definition of time-dependent Wannier functions. Looking at the  $f^{\text{static}}$  Wannier function (red line), we see that compensating for the static energy dephasing alone is also insufficient to retain the localized nature, as the spread evolves over time to become as fully delocalized as the uncompensated case. This is particularly evident when compared to the performance of the  $f^{\text{int}}$  Wannier function (yellow line), which loses only some locality during this strong driving process. Note that the spreads in Figs. 1(b) and 1(c) are shown on a logarithmic scale. We observe that even with the phase compensation of Eq. (4), the Wannier functions still retain some dynamics, and in the Sec. III we show that these dynamics can carry physical significance.

The model system here is simple enough that we can explore the evolution of the orbitals themselves in Fig. 2, where we plot the propagation of the Wannier function envelope for the case of the strong driving. Here, we get a more intuitive

view of how the  $f^{\text{int}}$  Wannier functions retain their localized nature. We can see that although subtle, the Wannier function envelope is still evolving, with the appearance of small oscillations in the tails of the envelope. This indicates that there is some nontrivial dynamics encoded in the Wannier center and spread. The time-dependent Wannier centers allow us to generalize the theory of polarization to the time domain, as discussed in the Sec. III.

We note that this definition of time-dependent Wannier orbitals is closely related to other commonly used time-dependent single-particle basis sets, such as the Peierls basis

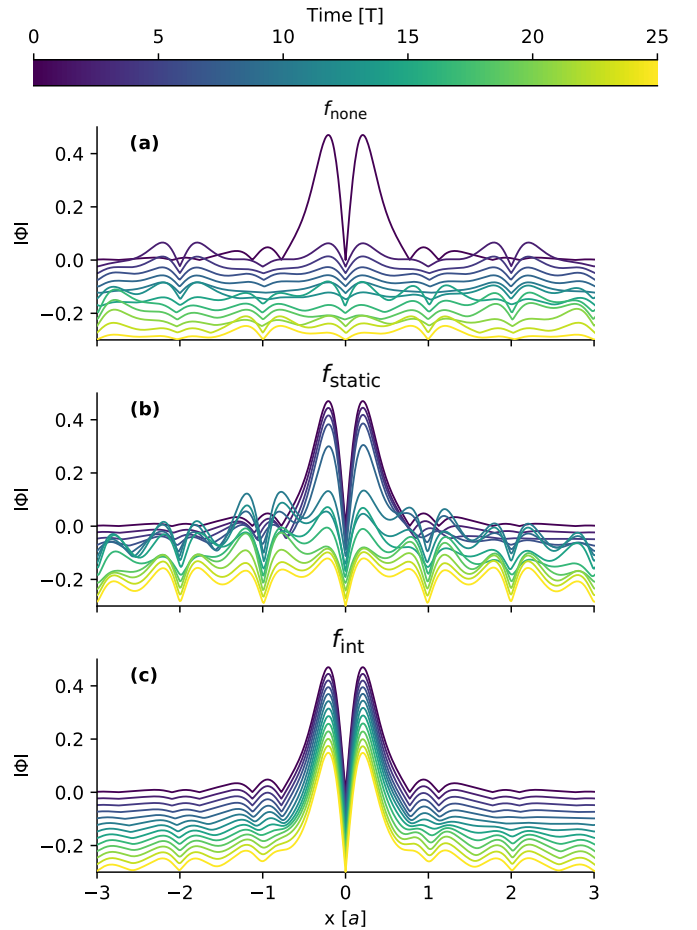


FIG. 2. Propagation of the Wannier function envelope in time. Each evaluated time step is color coded according to the color bar and shifted downwards.

and the Houston basis, which we detail in Appendix A. We especially note that the other bases do not change the shape of the single-particle wave-function envelope, and thus do not vary in their center and spread, as we have seen with the time-dependent Wannier functions given here. In Appendix A we also give a reasoning of why Eq. (4) is expected to work well for off-resonant excitation. Finally, we point out that here we do not discuss the generation of a time-dependent Wannier basis, but rather the construction of such states from a given time evolution of Bloch states. This means that the time dependence of the Bloch states is required to be available through an explicit time-propagation method and the Wannier functions discussed here can be thought of as a coevolving complement basis that ideally spans the same space, but with far fewer states.

### III. HIGH-HARMONICS GENERATION WITH TIME-DEPENDENT WANNIER FUNCTIONS

In order to showcase the capability of the time-dependent Wannier functions as a basis set, we investigate the dynamic theory of polarization derived from the time-dependent Wannier centers [14]. We calculate the time-dependent Wannier function for a model noninteracting 1D system and a real-space TDDFT calculation on h-BN. The calculations here were performed using octopus [62].

The main observable that we investigate is the polarization current derived from the electric dipole moment:

$$\mathbf{J}(t) = \frac{d\mathbf{P}(t)}{dt}, \quad (14)$$

where  $\mathbf{J}$  is the total electronic current and  $\mathbf{P}$  is the dynamic electronic polarization, which we relate to the time-dependent Wannier functions [14] via

$$\mathbf{P}(t) = -\frac{e}{V} \sum_n \mathbf{r}_{n0}(t). \quad (15)$$

We compare this current with the integrated current density calculated directly in the Bloch-state representation [62] taken to be the reference current.

Here, we analyze the HHG yield [54,57–59,63]:

$$\xi(t) = \frac{d\mathbf{J}(t)}{dt}, \quad (16)$$

which we calculate either directly from the integrated current density evaluated in the full TDDFT calculation by taking the time-dependent expectation value of the current operator, or from the polarization current that is in turn derived from the Wannier centers. We are primarily investigating the HHG yield, which we calculate from its Fourier transform:

$$I_{\text{HHG}}(E) = \int e^{-i(1/\hbar)Et} \xi(t) dt. \quad (17)$$

We apply a Gaussian filter along the time domain before evaluating its Fourier transform in order to clean up the residual oscillation at the tail of the laser pulse.

#### A. Noninteracting 1D system

For the noninteracting 1D system we take the Mathieu potential explored in Sec. II, and we explore a wider range

of laser parameters. The HHG in this system has been extensively studied using various calculation methods [35,64]. In Fig. 3 we see a good agreement of the HHG yield derived from the Wannier functions (dashed red line) to the reference calculation using the Bloch states (solid blue line), across a wide range of laser parameters.

It is worth emphasizing how good the agreement of these results is. We reiterate that the time-dependent Wannier function definition is not unique, so there is no guaranteed correlation between the current obtained from the Wannier functions through Eqs. (14) and (15) and the observable current calculated from the Bloch states. This was clear from Sec. II, but it is even more evident if we compare the HHG spectra from Fig. 4, where it is clear that only the definition using Eqs. (1) and (4) gives us a polarization current that matches the reference observable. In principle, we would still be able to use the other Wannier functions as a time-dependent basis set, but in order to calculate an observable on such a basis we would have to calculate the time-dependent occupation and/or expand the time-dependent Wannier basis set beyond the initially occupied ones in order to compensate for the difference in the Hilbert space. Here, we show that the time-dependent Wannier basis is an efficient basis set for capturing the local dynamics of the electrons in a more compact and localized representation. Importantly, this representation is able to capture the full range of nonlinear processes in HHG, as can be seen in the agreement of all the HHG spectra evaluated in Fig. 3.

One key usage of the Wannier functions is to gain chemical intuition of electronic structure, and is worthwhile exploring how much of an intuitive picture of the dynamic processes is provided by our time-dependent Wannier functions. We now show that the time-dependent Wannier functions indeed allow for an intuitive picture also for processes in the time domain and even highly nonlinear ones. First, we Wannierize each band in Fig. 1(a) separately, i.e., without performing the sum over bands in Eq. (1), and we propagate these states individually according to Eqs. (1) and (4). We plot the contribution to the HHG spectra of each of these Wannier functions in Fig. 5(a), where the independent bands caption refers to the initial Wannierization being performed individually for each band. We observe that effectively all of the HHG is contained in the dynamics of the Wannier function derived from the valence band [Wannier 2 in Fig. 5(a)], even as the Wannier function derived from the lower band [Wannier 1 in Fig. 5(a)] shows an equally strong linear response.

Next, we perform the Wannierization procedure on both bands together, as done in all examples above, allowing the Wannier functions to mix and converge to a more localized basis set, and we propagate these states individually as before. This is more indicative of the general procedure that we might encounter when the bands are not as well separated that we could neatly converge the individual bands. We represent this case in Fig. 5(b), where now the labeling of Wannier functions is simply in order of their spread. What is immediately intriguing about the result shown in Fig. 5(b) is that the total HHG remains unchanged, which can be explained by how the initial Wannier functions are related with each other by a unitary transformation. In this representation, the dynamics of the valence band is spread across both Wannier functions,

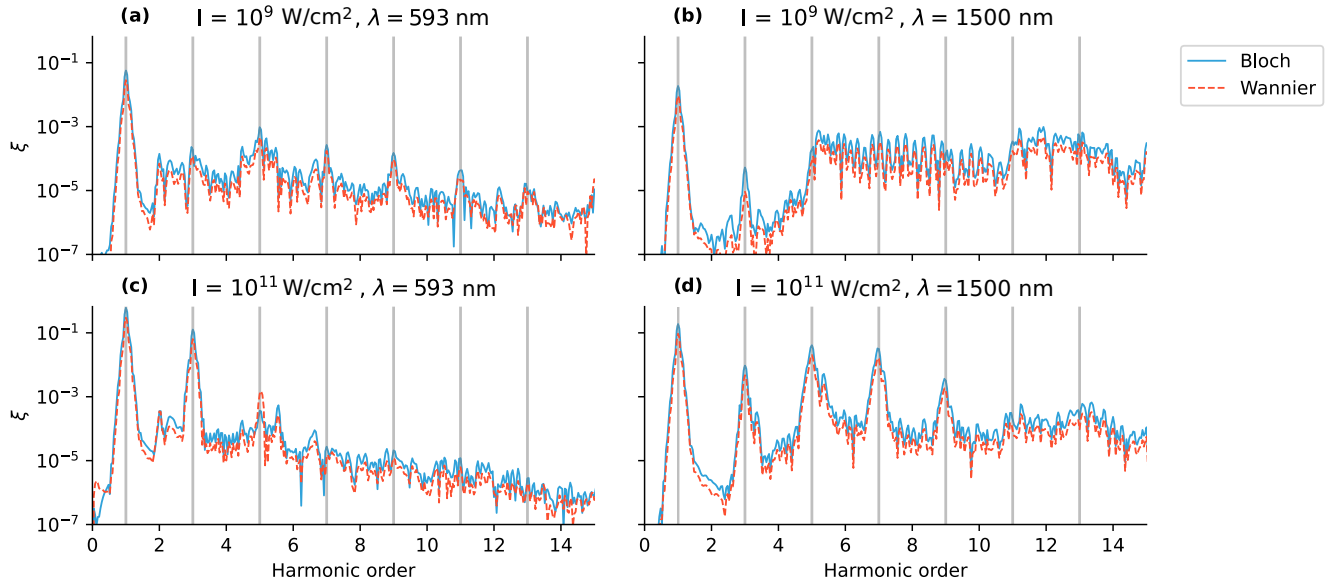


FIG. 3. HHG spectra of the noninteracting 1D system with varying laser interactions. The different lines correspond to the HHG yield calculated from the current calculated directly from the Bloch states or from the Wannier centers [Eq. (14)].

and therefore we cannot visualize the HHG as nicely separated contributions as before. However, we can still observe an important effect: the individual Wannier states propagate with even-order harmonics that cancel each other out due to the dynamical inversion symmetry in the model [65]. This shows that the individual trajectories of Wannier centers of mixed-band character can break the symmetry; however, the dynamics of the whole set of Wannier centers is restoring the symmetry. This can be thought of in analogy to the motion of individual atoms in phonons modes, which only collectively form the eigenmode of the lattice.

### B. Hexagonal-BN

To show the generality of this approach we now consider a realistic system in 3D, h-BN, which we drive with similarly strong laser profiles. Although the system itself is a 2D lattice, and we only present the nontrivial results that are observed along the lattice plane, the calculations are performed in the full 3D real-space grid. The simulation is done with Hartwigsen-Goedecker-Hutter pseudopotentials [66] and corresponding local-density approximation exchange-correlation functionals, and lattice dimension of  $a = 4.76(a_0)$ . For the

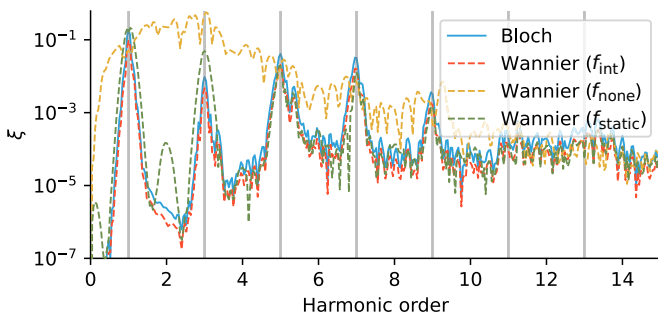


FIG. 4. HHG spectra of the noninteracting 1D system of the different time-dependent Wannier function definitions from Eqs. (2)–(4).

laser driving we take only  $n_{\text{cyc}} = 8$  and focus on the results with a peak laser intensity of  $I = 10^{11} \text{ W/cm}^2$  and wavelength of  $\lambda = 800 \text{ nm}$ . The simulation was done in a real-space grid with spacing  $\Delta a = 0.38(a_0)$ , a  $k$ -space grid of  $27 \times 27 \times 1$ , and a time step of  $\Delta t = 0.29(\hbar/E_h)$ . In this simulation we only sample the time-dependent Wannier functions every 20 time steps.

The HHG spectra of this system have previously been investigated using first-principles methods and model calculations alike [36,67,68].

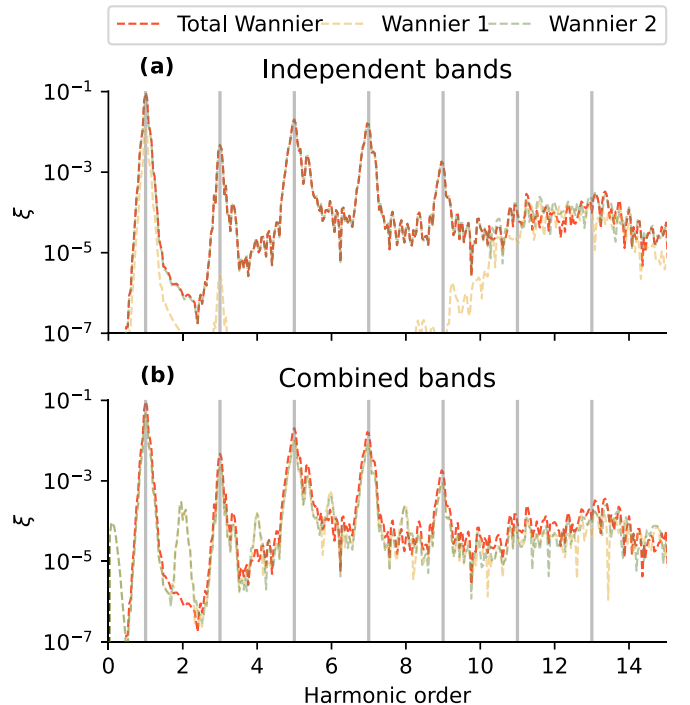


FIG. 5. HHG spectra of the noninteracting 1D system with different initial Wannier functions.

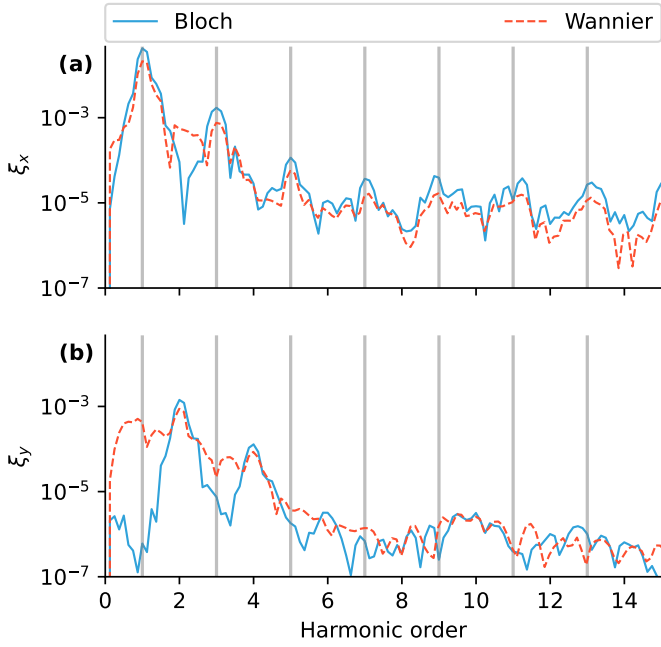


FIG. 6. HHG spectra of the h-BN system along the  $x$ - and  $y$  axes derived from the current calculated directly from the Bloch states or from the Wannier centers [Eq. (14)].

Looking at the HHG spectra of this system in Fig. 6, we find a weaker but still relatively good agreement between the Wannier function-derived spectra and the reference one along the driving  $x$  axis. The fact that the agreement is not as good as in the previous model case is to be expected because we are performing various conceptual leaps as we apply TDDFT functional approximations; use a coarser time, space, and  $k$ -space grid; evaluate a shorter laser driving; and evaluate the time-dependent Wannier functions less frequently. It is natural that more numerical errors accumulate, and in light of those factors, the fact that the HHG spectra still agree that much, however, is promising, especially considering again that the two spectra are calculated independently, one from the exact quantum-mechanical expression and the other from the intuitive expression based on the Wannier states' centers of

mass. We highlight that these two expressions are not identical and we could not prove their connection analytically, although intuitively we expect this connection to exist, at least in a cleverly chosen gauge. Further theory connecting the expressions should motivate future work.

We next consider the HHG spectra along the  $y$  axis, where we see a stronger disagreement. To understand the cause of this deviation, we look at the raw time-dependent currents, which we show in Fig. 7 along with the driving-laser profile. Here, we do not apply the Gaussian filtering, and we scale the laser driving, the reference current, and the total Wannier current to their highest absolute value for plotting visibility. Here, we want to point out how each Wannier function contributes to the total Wannier current. Along both the  $x$  and  $y$  axes we have significant cancellation of currents as we have similarly seen in the noninteracting model in Fig. 5, but in this case, the canceling oscillations exceed the total current, especially along the  $y$  axis where the difference is more than one order of magnitude. This, combined with how the reference HHG spectra along the  $y$  axis are two orders of magnitude smaller, and the limitations of the simulations that we previously mentioned, makes it acceptable to find such deviations due to the numerical limitations.

In Appendix B we evaluate the propagator of the noninteracting system to find one source of deviation due to resonant excitation. We expect that with further refinement of the time-dependent Wannier function definition, we will find a more optimal single-particle basis set to represent the dynamics of the many-body interacting system.

#### IV. CONCLUSION

The main result of this work is the definition of the time-dependent Wannier functions that can be efficiently performed on top of well-established first-principles calculation methods, primarily the real-time TDDFT. In comparison with other proposed definitions for the time-dependent Wannier functions, the method developed here avoids the most expensive part of maximally localizing the basis set. The dynamical polarization model is shown to give quite good agreements of the current-density calculations if we compensate for the dephasing factor of the Wannier states.

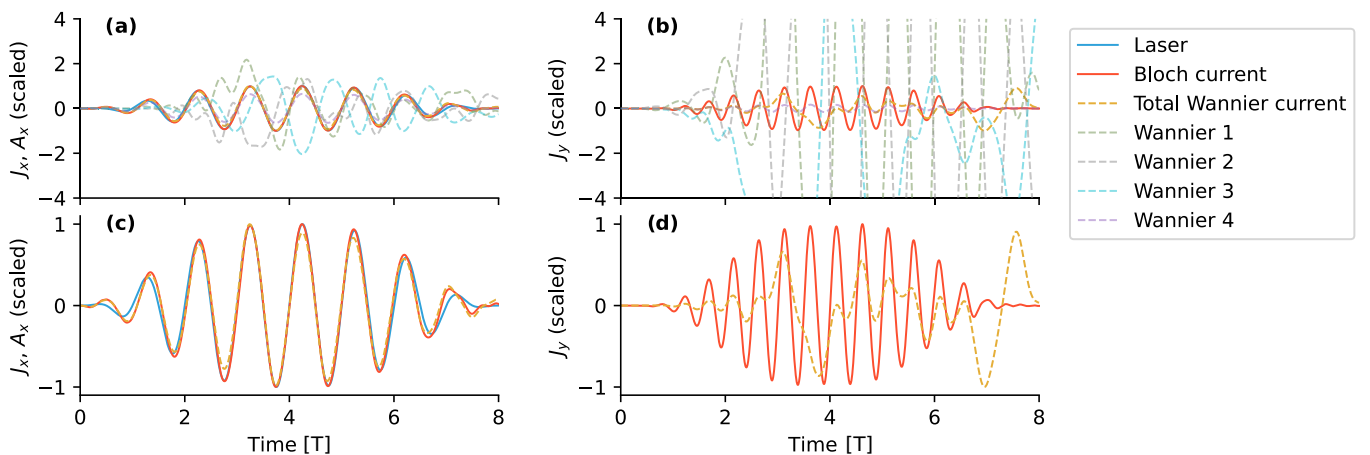


FIG. 7. Applied laser and currents of the h-BN system calculated directly from the Bloch states or from the Wannier centers [Eq. (14)].

While other time-dependent lattice basis, such as the Peierls basis set, require an extensive basis including the initial unoccupied bands and various lattice sites [35,64,69], with the time-dependent Wannier functions we are able to limit the necessary basis set to a few of the occupied bands and a single lattice site. This results in a much simpler lattice model that can also be used to study strongly correlated materials, besides the applications demonstrated here.

The choice of the phase-compensation function that we studied here is somewhat arbitrary and clearly nonuniversal. In our preliminary study, we have found that the relative good agreement we have shown in this study breaks down at strong and resonant drivings, which should motivate future work exploring the reasons of this failure and other potential gauge choices for time-dependent Wannier states.

While we here have only explored the applicability of the time-dependent Wannier functions for extracting some physical intuition of the underlying dynamics and as a tool for analyzing nonlinear electronic dynamics, there is a vast range of further applications to explore, from improving the TDDFT calculations to visualizing and more efficiently calculating the dynamic observables. Looking forward, we believe that time-dependent Wannier functions will prove useful in the evolving field of ultrafast dynamics in solids, especially moving towards strongly correlated materials [60,70–75], phonon dynamics [55,63,76,77], excitonic effects [78–84], superconductivity [85,86], etc.

## ACKNOWLEDGMENTS

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## APPENDIX A: RELATION TO OTHER BASIS SETS

In order to examine the relation of the time-dependent Wannier functions with other commonly used bases like the Peierls basis or the Houston basis, we look at the time-dependent noninteracting electron system, for which we can express the time-dependent Schrödinger equation as

$$\psi_{\mathbf{n}\mathbf{k}}^{\mathbf{A}}(\mathbf{r}, t) = \langle \mathbf{r} | \psi_{\mathbf{n}\mathbf{k}}^{\mathbf{A}}(t) \rangle = e^{-i(1/\hbar)E_{\mathbf{n}\mathbf{k}}^0 t} e^{i(\mathbf{k} + (e/\hbar)\mathbf{A}(t))\mathbf{r}} u_{\mathbf{n}\mathbf{k}}(\mathbf{r}), \quad (\text{A1})$$

$$C_{\mathbf{n}\mathbf{k}}^i(t) = \langle \psi_{\mathbf{n}\mathbf{k}}^{\mathbf{A}}(t) | \Psi_{\mathbf{i}\mathbf{k}}(t) \rangle, \quad (\text{A2})$$

$$\partial_t C_{\mathbf{n}\mathbf{k}}^i(t) = -e \frac{\partial \mathbf{A}(t)}{\partial t} \sum_m \mathbf{D}_{\mathbf{n}\mathbf{m}}^{\mathbf{k}}(t) C_{\mathbf{m}\mathbf{k}}^i(t), \quad (\text{A3})$$

where  $\psi$  is an auxiliary basis set which we use here for decomposing the dynamics of the propagator,  $C_{\mathbf{n}\mathbf{k}}^i$  is the time-dependent projection of the Bloch wave function  $\Psi_{\mathbf{i}\mathbf{k}}$  on the time-dependent wave function  $\psi_{\mathbf{n}\mathbf{k}}$ , and  $\mathbf{D}_{\mathbf{n}\mathbf{m}}^{\mathbf{k}}(t)$  is constructed

from the transition dipole moment  $\mathbf{D}_{\mathbf{n}\mathbf{m}}^{\mathbf{k}}$ :

$$\mathbf{D}_{\mathbf{n}\mathbf{m}}^{\mathbf{k}}(t) = e^{-i(1/\hbar)(E_{\mathbf{n}\mathbf{k}}^0 - E_{\mathbf{m}\mathbf{k}}^0)t} \mathbf{D}_{\mathbf{n}\mathbf{m}}^{\mathbf{k}}, \quad (\text{A4})$$

$$\mathbf{D}_{\mathbf{n}\mathbf{m}}^{\mathbf{k}} = \int d\mathbf{r} u_{\mathbf{n}\mathbf{k}}^*(\mathbf{r}) \mathbf{r} u_{\mathbf{m}\mathbf{k}}(\mathbf{r}). \quad (\text{A5})$$

More details on this derivation are provided in Appendix B. In this derivation we are assuming the dipole approximation. Here, we point out that outside the resonance regime, Eq. (A4) oscillates rapidly such that Eq. (A3) approximates to a constant, and we can take Eq. (A1) to approximate the time-dependent Bloch states.

We apply the basis equation (A1) to Eq. (1):

$$\Phi_{\mathbf{n}\mathbf{R}}(\mathbf{r}, t) \approx \sum_i \int_{\text{BZ}} U_{\mathbf{i}\mathbf{k}}^{\mathbf{R}} f_{\mathbf{i}\mathbf{k}}(t) \psi_{\mathbf{i}\mathbf{k}}^{\mathbf{A}}(\mathbf{r}, t) d^3\mathbf{k}, \quad (\text{A6})$$

$$\approx P^{\text{AC}}(\mathbf{r}, t) \sum_i \int_{\text{BZ}} P_{\mathbf{i}\mathbf{k}}^{\text{D}}(t) U_{\mathbf{i}\mathbf{k}}^{\mathbf{R}} \Psi'_{\mathbf{i}\mathbf{k}}(\mathbf{r}, t) d^3\mathbf{k}, \quad (\text{A7})$$

where we group the time-dependent effects into a phase acceleration  $P^{\text{AC}}$ , common for all Wannier functions and dependent only on the laser profile, the main dephasing contribution  $P^{\text{D}}$  that we investigated in Sec. II, and the remaining Bloch-state dynamics  $\Psi'$  that are covered in Eq. (A3):

$$P^{\text{AC}}(\mathbf{r}, t) = e^{i \frac{e}{\hbar} \mathbf{A}(t) \mathbf{r}}, \quad (\text{A8})$$

$$P_{\mathbf{i}\mathbf{k}}^{\text{D}}(t) \approx e^{-i(1/\hbar)E_{\mathbf{n}\mathbf{k}}^0 t} f_{\mathbf{i}\mathbf{k}}(t), \quad (\text{A9})$$

$$\Psi'_{\mathbf{i}\mathbf{k}}(\mathbf{r}, t) \approx \sum_j C_{\mathbf{i}\mathbf{k}}^j(t) e^{i\mathbf{k}\mathbf{r}} u_{j\mathbf{k}}(\mathbf{r}). \quad (\text{A10})$$

$P^{\text{AC}}$  does not contribute to the dynamics of Wannier centers or Wannier spread,  $P^{\text{D}}$  is close to unity in the Wannier functions  $f^{\text{int}}$ , and the only remaining effects come from the excitation dynamics in Eq. (A3) through  $\Psi'$ . The dynamics in  $\Psi'$  are the main dynamics that we see in Secs. II and III.

Comparing this expression with Peierls basis  $\Phi^P$  and the Houston basis  $\Psi^H$ ,

$$\Phi_{\mathbf{n}\mathbf{R}}^P(\mathbf{r}, t) = e^{i \frac{e}{\hbar} \mathbf{A}(t) \mathbf{r}} \Phi_{\mathbf{n}\mathbf{R}}(\mathbf{r}, 0), \quad (\text{A11})$$

$$\Psi_{\mathbf{i}\mathbf{k}}^H(\mathbf{r}, t) = e^{i \frac{e}{\hbar} \mathbf{A}(t) \mathbf{r}} \Psi_{\mathbf{i}\mathbf{k}+\mathbf{A}}(\mathbf{r}, 0), \quad (\text{A12})$$

we can see how all of these basis sets merely account for the  $P^{\text{AC}}$  contribution, while the remaining dynamics have to be included by expanding the basis set into the unoccupied states and/or the many-body coefficients. When we consider that the Peierls substitution is often performed on a single-band basis for computational efficiency, we can see how the dynamics in Eq. (A3) can be completely overlooked.

## APPENDIX B: NONINTERACTING PROPAGATOR

In Appendix A we have referenced the exact propagator of noninteracting systems, and in this appendix we will elaborate on how we have derived it for the sake of completeness. We start from the time-independent Hamiltonian:

$$\hat{H}^0 = \frac{1}{2m} \hat{\mathbf{p}}^2 + \hat{V}, \quad (\text{B1})$$

where  $\mathbf{p}$  and  $V$  are the momentum and spatially periodic potential, respectively. Let us consider we have solved the

time-independent Schrödinger equation to get the eigenstates and eigenvalues

$$\hat{H}^0 |\Psi_{\mathbf{k}}^0\rangle = E_{\mathbf{k}}^0 |\Psi_{\mathbf{k}}^0\rangle, \quad (\text{B2})$$

$$\Psi_{\mathbf{k}}^0(\mathbf{r}) = \langle \mathbf{r} | \Psi_{\mathbf{k}}^0 \rangle = e^{i\mathbf{k}\mathbf{r}} u_{\mathbf{k}}(\mathbf{r}). \quad (\text{B3})$$

For the laser interaction, we take the velocity gauge Hamiltonian:

$$\hat{H}(t) = \frac{1}{2m} (\hat{\mathbf{p}} - e\mathbf{A}(t))^2 + \hat{V}, \quad (\text{B4})$$

where  $e$  is the charge of the electron and  $\mathbf{A}$  is the applied laser field. For simplicity, we are taking the dipole approximation, ignoring the spatial variation of  $\mathbf{A}$ . We solve the

time-dependent Schrödinger equation using the basis defined in Eq. (A1). This basis is an instantaneous eigenbasis of the time-dependent Hamiltonian:

$$\langle \psi_{m\mathbf{k}'}^{\mathbf{A}}(t) | \hat{H}(t) | \psi_{n\mathbf{k}}^{\mathbf{A}}(t) \rangle = \delta_{mn} \delta(\mathbf{k}' - \mathbf{k}) E_{n\mathbf{k}}^0. \quad (\text{B5})$$

We solve the time-dependent Schrödinger equation using the overlap

$$C_{n\mathbf{k}}^{\mathbf{k}'}(t) = \langle \psi_{n\mathbf{k}'}^{\mathbf{A}}(t) | \Psi_{\mathbf{k}}(t) \rangle, \quad (\text{B6})$$

where  $\Psi_{\mathbf{k}}(t)$  is the time-dependent wave function of the non-interacting electron with the initial condition

$$|\Psi_{\mathbf{k}}(0)\rangle = |\Psi_{\mathbf{k}}^0\rangle. \quad (\text{B7})$$

Expanding the time-dependent Schrödinger equation on this basis we get

$$\langle \psi_{n\mathbf{k}'}^{\mathbf{A}}(t) | i\hbar \partial_t | \Psi_{\mathbf{k}}(t) \rangle = \langle \psi_{n\mathbf{k}'}^{\mathbf{A}}(t) | \hat{H}(t) | \Psi_{\mathbf{k}}(t) \rangle, \quad (\text{B8})$$

$$\sum_m \int d\langle \psi_{n\mathbf{k}'}^{\mathbf{A}}(t) | i\hbar \partial_t | \psi_{m\mathbf{k}''}^{\mathbf{A}}(t) \rangle \langle \psi_{m\mathbf{k}''}^{\mathbf{A}}(t) | \Psi_{\mathbf{k}}(t) \rangle = E_{n\mathbf{k}}^0 C_{n\mathbf{k}}^{\mathbf{k}'}(t), \quad (\text{B9})$$

$$\sum_m \int d \left[ -e \frac{\partial \mathbf{A}(t)}{\partial t} \int d\langle \psi_{n\mathbf{k}'}^{\mathbf{A}}(t) | \hat{\mathbf{r}} | \psi_{m\mathbf{k}''}^{\mathbf{A}}(t) \rangle + \delta_{mn} \delta(\mathbf{k}' - \mathbf{k}'') (i\hbar \partial_t + E_{m\mathbf{k}''}^0) \right] C_{m\mathbf{k}''}^{\mathbf{k}'}(t) = E_{n\mathbf{k}}^0 C_{n\mathbf{k}}^{\mathbf{k}'}(t). \quad (\text{B10})$$

We denote  $\mathbf{D}_{nm}^{\mathbf{k}'\mathbf{k}''}(t)$  constructed from the transition-dipole moment  $\mathbf{D}_{nm}^{\mathbf{k}'\mathbf{k}''}$  as

$$\mathbf{D}_{nm}^{\mathbf{k}'\mathbf{k}''}(t) = \langle \psi_{n\mathbf{k}'}^{\mathbf{A}}(t) | \hat{\mathbf{r}} | \psi_{m\mathbf{k}''}^{\mathbf{A}}(t) \rangle, \quad (\text{B11})$$

$$= e^{-i(1/\hbar)(E_{m\mathbf{k}''}^0 - E_{n\mathbf{k}'}^0)t} \mathbf{D}_{nm}^{\mathbf{k}'\mathbf{k}''}, \quad (\text{B12})$$

$$\mathbf{D}_{nm}^{\mathbf{k}'\mathbf{k}''} = \delta(\mathbf{k}' - \mathbf{k}'') \int d\mathbf{r} u_{n\mathbf{k}}^*(\mathbf{r}) \mathbf{r} u_{m\mathbf{k}}(\mathbf{r}). \quad (\text{B13})$$

Substituting this into Eq. (B10) and simplifying the Dirac deltas, we get

$$\partial_t C_{n\mathbf{k}}^{\mathbf{k}'}(t) = -e \frac{\partial \mathbf{A}(t)}{\partial t} \sum_m \mathbf{D}_{nm}^{\mathbf{k}'}(t) C_{m\mathbf{k}}^{\mathbf{k}'}(t), \quad (\text{B14})$$

or considering the initial condition

$$C_{n\mathbf{k}}^{\mathbf{k}'}(0) = \delta_{in} \delta(\mathbf{k} - \mathbf{k}'), \quad (\text{B15})$$

we can simplify the equation of motion by ignoring  $\mathbf{k}' \neq \mathbf{k}$  and we get the form in Eq. (A3).

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