

High order harmonic generation in semiconductors driven at near- and mid-IR wavelengths

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We study high order harmonics generation (HHG) in crystalline silicon and diamond subjected to near and mid-infrared laser pulses. We employ time-dependent density functional theory and solve the time-dependent Kohn-Sham equation in the single-cell geometry. We demonstrate that clear and clean HHG spectra can be generated with careful selection of the pulse duration. In addition, we simulate dephasing effects in a large silicon super-cell through a displacement of atomic positions prepared by a molecular dynamics simulation. We compare our results with the previous calculations by Floss et al. [Phys. Rev. A **97**, 011401(R) (2018)] on Diamond at 800 nm and by Tancogne-Dejean et al. [Phys. Rev. Lett. **118**, 087403 (2017)] on Si at 3000 nm.

I. INTRODUCTION

High order harmonics generation (HHG) in solids has attracted efforts of many experimental and theoretical groups. The ultimate goal of these efforts is producing a compact and tunable source of coherent XUV and soft X-rays on a chip that can be integrated into a microelectronic device. The recent advances of HHG in solids has been reviewed in the experiment [1] and theory [2]. Previous efforts on HHG in condensed matter have been driven by improved harmonic intensities offered by the higher atomic densities of solids compared to those offered by atomic gases. Experimental studies into HHG for thin films of bulk crystal targets [3–13] have shown that improved intensities are achievable as demonstrated theoretically [1]. Targets have also recently been expanded to structured metasurfaces [14, 15] and their 2D component layers like graphene [16–18].

Theoretical studies currently employ methods of varying quantitative power to model the interaction of solid state targets with the pulsed electric fields driving HHG. These methods include solution of the time-dependent Schrödinger equation [19–21], one-dimensional models [22–26], density matrix models [27–29] and ab initio descriptions like time-dependent density functional theory (TDDFT) [30–34]. Recently, theories have also been developed to describe HHG in more complex targets such as bulk surfaces [35–37] and metasurfaces [38]. Investigations using these methods have revealed the contribution of inter- and intraband mechanisms of the HHG process which are visualized in the energy band picture [1, 28]. Similarly, the impact of band structure, band selection and laser polarization have been studied at length theoretically [8, 10, 17].

Theoretical description of HHG in bulk crystalline tar-

gets requires dealing with light propagation effects. Indeed, in the experiment, a strong dephasing effect is often observed for condensed matter targets [5, 6, 10, 28, 39, 40]. This effect has been explored in theoretical studies [31, 32, 41] and found to be prominent in bulk solids due to the higher atomic density than found in atomic gases. However, ultra-fast dephasing times of the order of 1 fs that has been proposed in theory [42, 43] have posed a problem for the strong-field community. Recently, these dephasing times have been reconsidered through simulation of the linear response of the current density to broadband excitation [31]. This analysis proposed dephasing times of the order of 10 fs and rectified the experimental and theoretical spectra generated using TDDFT and semiconductor Bloch equations (SBE) through simulation of the laser focus propagation effect.

One of the major goals of theoretical investigations of HHG in solids is to find the optimum conditions to generate clear and strong harmonic signals. In the Maxwell-SBE calculations [31], it was shown that the propagation effect is essential to obtain a clear HHG signal of high order due to the dephasing effect. Using the single-scale Maxwell-TDDFT scheme [44], an effort was made to find the optimal thickness of a thin film that produces the most intense HHG signals. It was shown that a very thin film of thickness of 2 - 15 nm is the optimum choice to produce intense HHG signals. In the present work, we investigate yet another unexplored factor, a time duration of the pulse that produces clear and intense HHG signals of high order. For longer driving pulses, more cycles near the maximum field strength contribute to the HHG process with the matter current getting closer to a quasi-periodic behavior thus contributing to cleaner harmonic peaks. For the purpose of our investigation, we study HHG in crystalline diamond (Di) and Si utilizing the TDDFT implementation within the computational SALMON framework [45]. We limit ourselves with the single cell (SC) calculations, which solve the time-dependent Kohn-Sham (TDKS) equation for the electron

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dynamics in TDDFT. These calculations consider the interaction between the electrons inside the bulk crystal targets and a pre-specified pulsed electric field. Within this scheme, we also investigate a dephasing effect by considering a large supercell with thermally distorted atomic positions. Calculation of HHG signals from the supercell will clarify the dephasing effect coming from electron-phonon coupling.

The paper is organized as follows: In Sec. II we describe the theoretical techniques for both time-dependent HHG and dephasing calculations. In Sec. III, we present numerical results and examine the HHG spectra. We conclude in Sec. IV by outlining the future of this method through extension to other condensed-matter systems.

II. METHODS

A. Single unit-cell method

The SC-TDDFT method considers the interaction of a bulk crystal unit cell with a spatially uniform electric field [46, 47]. This method has been used in the study of nonlinear and ultrafast dynamics [30–34, 48–52]. Utilizing the dipole approximation, the electronic motion can be described through the Bloch orbitals of the bulk, $u_{n\mathbf{k}}(\mathbf{r}, t)$, where the wave vector \mathbf{k} is contained within the three dimensional (3D) Brillouin zone of the unit cell. The TDKS equation for such orbitals is written as:

$$i\hbar \frac{\partial u_{n\mathbf{k}}(\mathbf{r}, t)}{\partial t} = \left\{ \frac{1}{2m} \left(-i\hbar \nabla + \hbar \mathbf{k} + \hat{\mathbf{x}} \frac{e}{c} A(t) \right)^2 - e\phi(\mathbf{r}, t) + \delta \hat{V}_{\text{ion}} + V_{xc}(\mathbf{r}, t) \right\} u_{n\mathbf{k}}(\mathbf{r}, t). \quad (1)$$

The applied electric field in this approach is defined by the vector potential of the incident pulse, $A(t)$. The electric potential $\phi(\mathbf{r}, t)$ includes the Hartree potential from the electrons and the local contribution of the ionic potential. The terms δV_{ion} and $V_{xc}(\mathbf{r}, t)$ describe the nonlocal part of the ionic pseudopotential [53] and the exchange-correlation potential [54], respectively. In our calculations, this exchange-correlation potential utilizes the adiabatic local-density approximation (LDA) [55]. Note that the LDA underestimates the bandgap of Si, as it does for diamond and other dielectrics. The experimental direct bandgap of Si is 3.4 eV while LDA gives 2.4 eV. We neglect the exchange-correlation term in the vector potential $A_{xc}(t)$ for simplicity and do not rigorously model the exchange-correlation effects of these types for infinite periodic systems [56, 57].

The averaged electric current density, used in generating HHG spectra, is calculated as follows:

$$\mathbf{J}(t) = -\frac{e}{m} \int_{\Omega} \frac{d\mathbf{r}}{\Omega} \sum_{n\mathbf{k}}^{\text{occ}} u_{n\mathbf{k}}(\mathbf{r}, t) \times \left(-i\hbar \nabla + \hbar \mathbf{k} + \hat{\mathbf{x}} \frac{e}{c} A(t) \right) u_{n\mathbf{k}}(\mathbf{r}, t) + \delta \mathbf{J}(t). \quad (2)$$

Here Ω is the volume of the unit cell and n is the band index restricted to occupied bands. The term $\delta \mathbf{J}(t)$ denotes the contribution to the current density from the nonlocal part of the pseudopotential [46] and is given as follows:

$$\delta \mathbf{J}(t) = -\frac{e}{m} \int_{\Omega} \frac{d\mathbf{r}}{\Omega} \sum_{n\mathbf{k}}^{\text{occ}} u_{n\mathbf{k}}^*(\mathbf{r}, t) e^{-i[\mathbf{k} + \hat{\mathbf{x}}(e/c)A(t)]\mathbf{r}} \times \frac{[\mathbf{r}, \delta \hat{V}_{\text{ion}}]}{i\hbar} e^{i[\mathbf{k} + \hat{\mathbf{x}} \frac{e}{c} A(t)]\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r}, t) \quad (3)$$

B. Simulation details

We utilize the open-source software package SALMON (Scalable Ab initio Light-Matter simulator for Optics and Nanoscience) [45]. In this code, a uniform 3D spatial grid is defined for the electron orbitals. The time evolution of the electron orbitals is carried out using the Taylor expansion method [58].

For TDDFT calculations without dephasing, we define a cubic unit cell of the diamond structure containing eight atoms with varying side lengths depending on the specific target. For Di and Si calculations, the unit cell dimensions are defined as $a_{\text{Di}} = 0.357$ nm and $a_{\text{Si}} = 0.543$ nm. A spatial grid of 16^3 points is used in Si and 24^3 in Di calculations to enable an accurate description of the Bloch orbitals. Convergence over spatial (r-point) grids is observed at these grid sizes. Similarly, the 3D Brillouin zone is sampled by a grid made up of 32^3 k -points. Note that convergence over the Brillouin zone is not observed at this grid size for diamond although qualitatively this size appears reasonable where we restrict ourselves to below the 50th order harmonic. Otake [30] finds similar grid parameters for diamond but with convergence observed over the Brillouin zone. The timestep is set for stability of the calculation to be 2.5×10^{-3} fs for Si and 1×10^{-3} fs for Di. Note that tests for convergence are important in these calculations due to the discretization of the spatial, k -space and time parameters discussed here.

For the Si calculation with dephasing, a supercell of size 4^3 describing a 512 atom system is used. The Brillouin zone is sampled by a grid made up of 8^3 k -points. Atomic positions are shifted from the equilibrium position with the diamond structure while are frozen during the time evolution of electron orbitals. They are generated from molecular dynamics simulations. The detail of the preparation will be described below.

C. Generating HHG spectra

In our SC-TDDFT calculations, we utilize the following incident pulse profile:

$$A(t) = -\frac{cE_0}{\omega} \sin(\omega t) \cos^6 \left[\frac{\pi}{T} \left(t - \frac{T}{2} \right) \right], \quad 0 \leq t \leq T. \quad (4)$$

Here E_0 is the peak amplitude of the electric field, ω is the carrier frequency and T is the full duration of the pulse. For the \cos^6 envelope, the FWHM duration T_{FWHM} is related to the full duration T by $T_{\text{FWHM}} \simeq 0.3T$. In our calculations, we consider the photon energies $\hbar\omega = 0.43$ eV ($\lambda = 3000$ nm), $\hbar\omega = 0.62$ eV ($\lambda = 2000$ nm) or $\hbar\omega = 1.55$ eV ($\lambda = 800$ nm). We select typical pulse durations $T \gtrsim 100$ fs which are considerably larger than in previous HHG studies (~ 32 fs in [31, 32] and 30 fs in [44]).

In the SC-TDDFT formalism, the HHG spectrum is calculated as a Fourier transform of the matter current

$$\text{HHG}(\omega) = \omega^2 |J(\omega)|^2, \quad (5)$$

$$J(\omega) = \int dt e^{i\omega t} J(t) w(t), \quad (6)$$

$$w(t) = 1 - \left(\frac{t}{T} \right)^2 + 2 \times \left(\frac{t}{T} \right)^3, \quad (7)$$

where $w(t)$ is the window smoothing function applied to remove spurious peaks in the spectra. A Gaussian convolution of the form

$$\text{HHG}_{\text{av}}(\omega) = \int d\omega' (\pi\epsilon^2)^{-1/2} \exp \left[-\left(\frac{\omega - \omega'}{\epsilon^2} \right)^2 \right] \text{HHG}(\omega'), \quad (8)$$

is applied to carry out frequency averaging as often the case in experimental HHG measurements.

III. RESULTS

A. Bulk Si at mid-IR wavelengths

We first consider bulk Si driven by a 2000 nm laser pulse with 200 fs full duration with a peak intensity of 5×10^{11} W/cm². In Fig. 1, the time profile of the vector potential is shown in the upper panel, and the induced matter current is shown in the lower panel. Here the simulated matter current displays large noise contributions that become particularly prominent after around 110 fs. Note also that the current is very asymmetric compared to the symmetric driving pulse centered around 100 fs. We thus find that the SC-TDDFT method assuming a diamond structure without any distortion effects records unphysically oscillating current towards the end of the specified pulse length. In Sec. III.C, we will show that the oscillating current mostly disappears once we introduce the dephasing effect caused by the thermal motion of atoms.

Fig. 2 displays the corresponding HHG spectrum (in green) generated from the matter current shown in Fig.

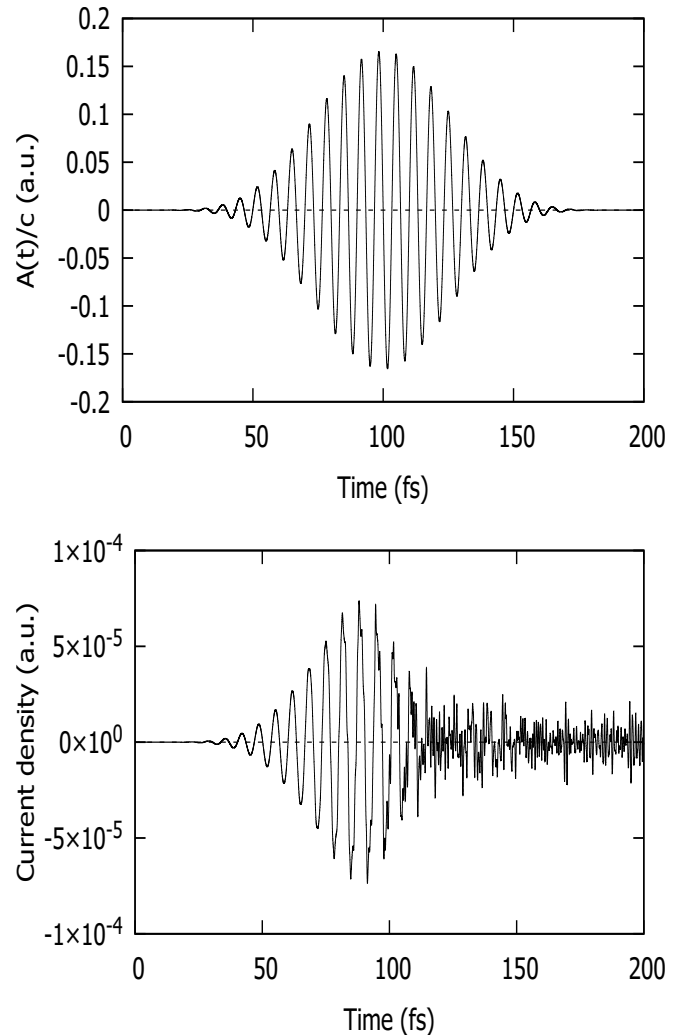


FIG. 1: The applied vector potential (top) of a pulse with a full duration of 200 fs at $\lambda = 2000$ nm and the peak intensity 5×10^{11} W/cm² and the matter current (bottom) in Si from a SC-TDDFT simulation.

1 using Eqs. (5-7). We confirmed the simulation's convergence over spatial (r-point) and momentum (k-point) grids up to 51st order harmonic for Si at 2000 nm. Nevertheless we show the spectrum up to 99th order to demonstrate both the presence of, and our methods application to, higher order harmonics. Although HHG signals can be identified in the unaltered spectrum (green) even around the highest order shown in the figure, the harmonic structure appears noisy and some harmonics cannot be clearly seen in this spectrum. Gaussian convolution using Eq. (8) overcomes this problem. Figure 2 shows the spectrum with the Gaussian filter (in red).

Fig. 3 demonstrates the major effect that pulse duration has on the clarity and extent of HHG spectra, considering a 3000 nm pulse interacting with Si. The 200 fs calculation (in green) shows clear harmonic peaks

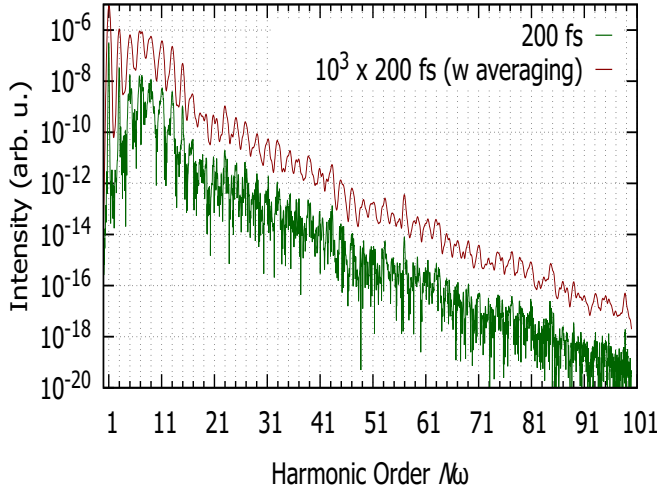


FIG. 2: HHG spectra for Si are generated with a Fourier transform (green) and then Gaussian convolution (red) using Eq. 8 with $\epsilon = 0.136$ eV. The spectra are the result of SC-TDDFT calculation considering interaction with a pulse with a full duration of 200 fs at $\lambda = 2000$ nm and peak intensity 5×10^{11} W/cm².

beyond the 27th harmonic order where the 70 fs calculation generally shows unclear harmonics until the 15th order. In addition to this, we note that the harmonic cutoff, comparing the simulations for the 200 fs and 70 fs pulses, appears dependent on pulse duration. We further examined this dependence through the application of an inverse Gabor transform focused to reveal the underlying current contributions associated with the highest order harmonics. Our analysis demonstrates, as expected, the importance of multi-cycle current accumulation in the formation of harmonic structure, where we observe that a 70 fs, 3000 nm pulse possesses too few cycles to capture the higher order returns.

Our Si results at 2000 nm (Fig. 2) and 3000 nm (Fig. 3) need to be contrasted with the earlier calculation on Si at 3000 nm presented by Tancogne-Dejean et al. [34]. In particular, using equivalent parameters to those reported in [34], a direct comparison is achieved for 3000 nm in Fig. 3. Tancogne-Dejean et al. [34] recorded noisy harmonics of the lower order while their higher order harmonics appeared clean. They related this effect with the joint density of states (JDOS) as a measure of the overlap between the valence and conduction bands. A large overlap and high JDOS would promote the recombination and inter-band HHG. Somewhat counter-intuitively, Tancogne-Dejean et al. [34] discovered that a high JDOS was in fact detrimental to HHG emission making the corresponding harmonic peaks noisy. Conversely, those harmonics falling into a low JDOS region of the HHG spectrum stayed clean. This observation led the authors to conclude that it was the intra-band mechanism that was largely responsible for HHG in silicon driven at MIR.

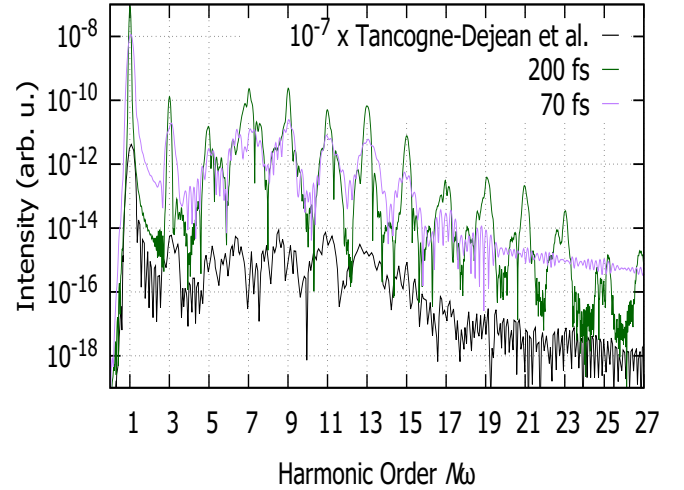


FIG. 3: HHG spectra for Si are generated with a Fourier transform (green) and compared with calculation (purple) similar to Tancogne-Dejean et al. [34] (black). The spectra are the result of SC-TDDFT calculation considering interaction with a pulse with full durations of 200 fs and 70 fs respectively, at $\lambda = 3000$ nm and peak intensity 1×10^{11} W/cm². Note that a \cos^6 envelope is applied here (for the pink calculation) instead of the \cos^2 carrier used by Tancogne-Dejean et al. [34].

However, our HHG spectra at both 2000 nm (Fig. 2) and 3000 nm (Fig. 3) at 200 fs total duration are both entirely clean even without averaging. Note that we clearly observe that the general JDOS effect is not present in our calculations at both 2000 nm and 3000 nm.

B. Bulk diamond at near IR wavelengths

In Fig. 4 we display the HHG spectra of Di produced by the SC-TDDFT. The primary calculation is driven by an 800 nm incident pulse of a 200 fs full duration and the peak intensity of 1×10^{13} W/cm². The Gaussian convoluted spectrum again reveals clear harmonics up to 41st order of the fundamental frequency ω , shown in the top panel of Fig. 4. The lower order harmonics of this spectrum are zoomed in on the bottom panel. The HHG spectrum is compared with an analogous calculation of Floss et al. [32]. The three sets of the SC-TDDFT calculations displayed in Fig. 4 are clearly differentiated by the pulse duration.

These authors employed the SC-TDDFT driven by a 2×10^{13} W/cm² pulse with a total duration of 32 fs [31]. Such a calculation did not produce a particularly clear HHG spectrum as is seen in Fig. 4. It is only the Maxwell propagation technique coupled with the SBE that allowed Floss et al. [31] to generate a clear spectrum. We thus conclude that, with refined parameters and a longer pulse duration, one can obtain distinguish-

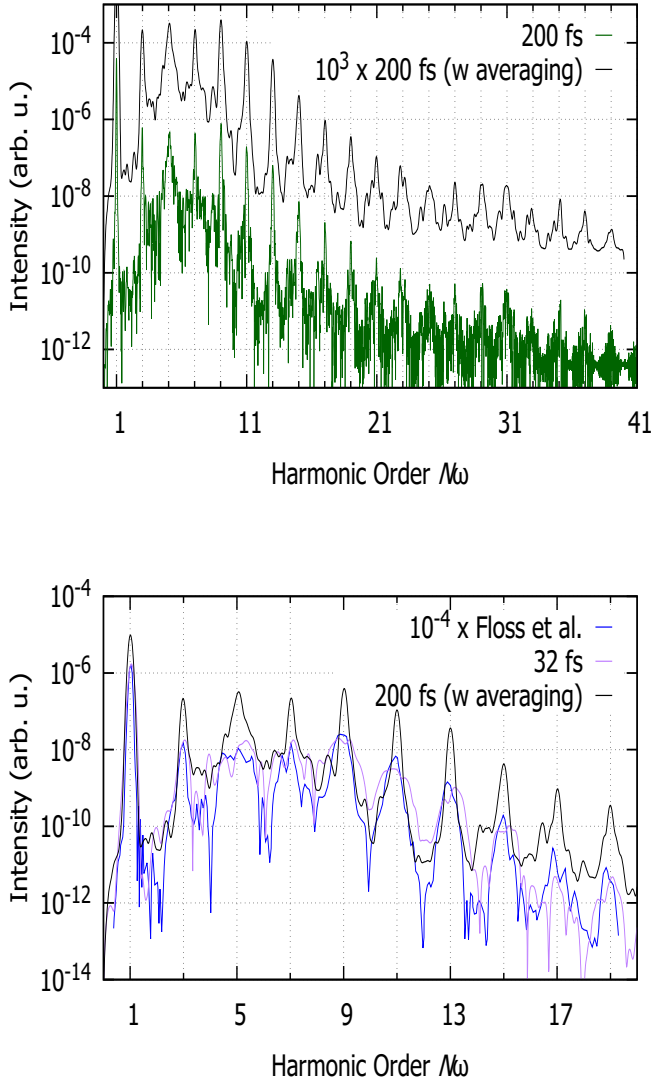


FIG. 4: Top: The HHG spectrum of Di (shown in green) and Gaussian convoluted spectrum (in black), using Eq. 8 with $\epsilon = 0.136$ eV, are generated with SC-TDDFT driven by a 800 nm pulse with a full duration of 200 fs and the peak intensity of 1×10^{13} W/cm². Bottom: The analogous spectrum of Floss et al. [32] with dephasing is displayed in blue and an SC-TDDFT calculation for a 32 fs pulse is shown in purple. The top and bottom panels show the expanded and zoomed in low order HHG spectra, respectively.

able high harmonics within the SC-TDDFT alone without the need for the Maxwell propagation or the explicit dephasing approach adopted in [32].

C. Dephasing Effect

We expand the SC-TDDFT method to consider the atomic system at a finite temperature by allowing the atomic positions to be displaced from equilibrium. For

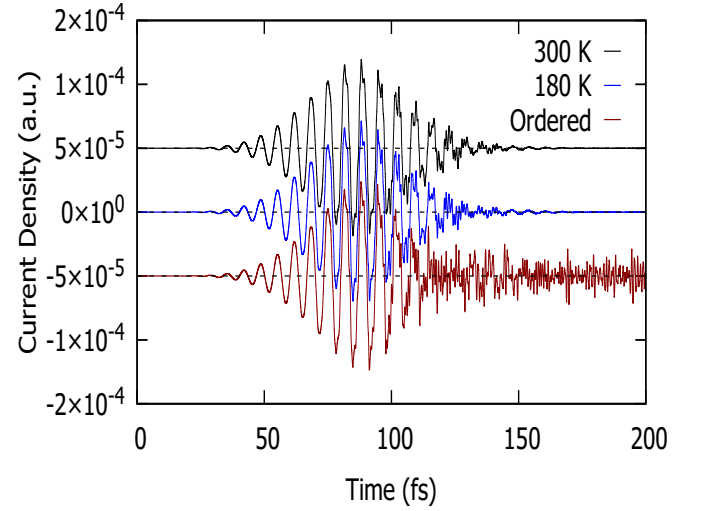


FIG. 5: Examples of the dephasing effect on the matter current in Si that arises from the application of a mean atomic position distortion. An effective temperature of 180 K (300 K) is used in simulating the current density shown in blue (black). These examples consider a 200 fs full duration pulse impinging on bulk Si and are compared to current from a similar TDDFT calculation without distortion of atomic positions (red). A shift of 5×10^{-5} is applied to help distinguish these results.

this purpose, we consider bulk Si and first carry out a molecular dynamics simulation with an empirical force field at a specified temperature using a thermostat. We use LAMMPS software [59] to carry out the molecular dynamics simulation where a three-body Stillinger-Weber potential [60] is used. We pick up several atomic configurations with sufficiently long intervals, and use these configurations in the SC-TDDFT. This represents a first-principles approach introducing the dephasing effect into TDDFT for the first time without relying on phenomenological parameter T_2 . This is a convenient approach where we consider that previously ultrafast dephasing times $T_2 \sim 1$ fs had to be introduced to reconcile theoretical treatments and experiment. During the time evolution stage in the SC-TDDFT, the atomic positions are fixed. We carry out calculations using supercells of different sizes, and have found that results of different configurations are quite similar to each other if we choose sufficiently large supercell. We will show below the calculation results using a supercell containing 512 atoms. Employing 8^3 k -points, the total number of atoms is equal to 262,144, that is equal to the total number of atoms in a cell containing 8 atoms with the 32^3 k -points that we showed in Fig. 2. In fact, we numerically confirmed that calculations without any distortion coincide with each other accurately between the two unit cells, 8 atoms with 32^3 k -points and 512 atoms with 8^3 k -points.

We show electron current density in bulk Si under the applied electric field in Fig. 5, caused by the same applied

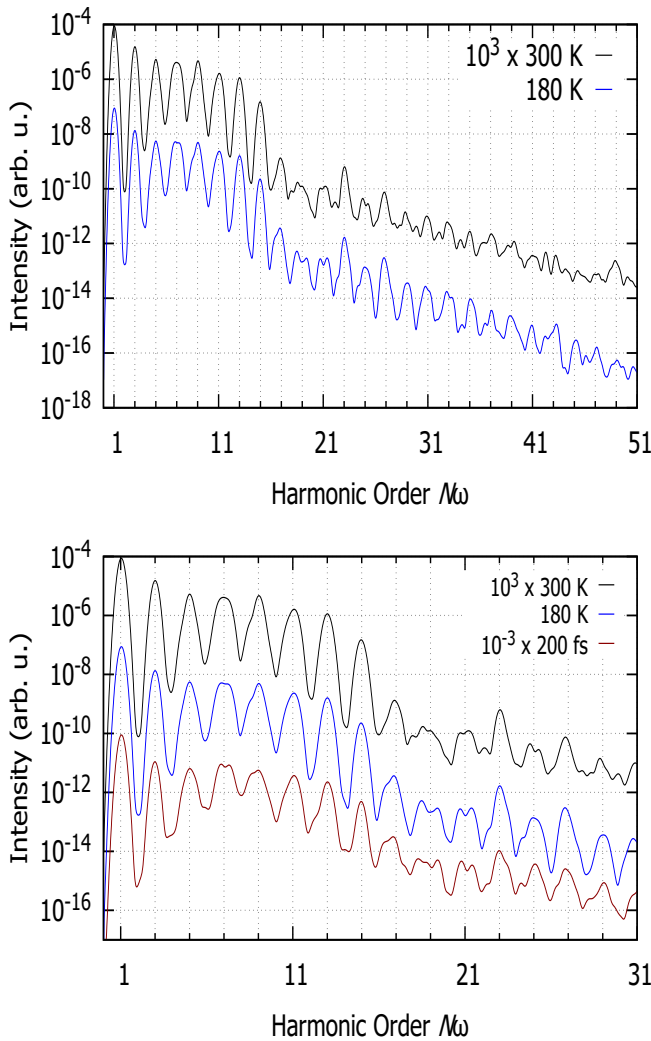


FIG. 6: Comparison of convoluted HHG spectra from bulk Si with dephasing effect by an atomic displacement (top), together with its equivalent single-cell calculation (red, bottom) with Gaussian convolution applied using Eq. 8 with $\epsilon = 0.136$ eV. Blue and black calculations here show the convoluted spectrum with atomic displacement by a thermal motion at 180 K (300 K) in classical molecular dynamics calculation.

pulse as shown in Fig. 1. The black (blue) calculation displays the current under the atomic configuration at a temperature of 300K (180K). The matter current in disordered Si with distortion exhibits the desired dephasing after the pulse has ceased interacting. With dephasing present, signals beyond 150 fs appear significantly suppressed and the noise contributions seen in Fig. 1 (red in Fig. 5) are removed.

Figure 6 demonstrates the effect of incorporating dephasing into the HHG calculations in Si where Gaussian convolutions are also applied. The black calculation shows the HHG spectrum using atomic positions from a molecular dynamics calculation at 300K. While signals

up to 100th order can be seen in Fig. 2 without atomic distortion, here we can see HHG signals up to 31st order, and higher order signals cannot be identified. We also note that the HHG signals themselves become deeper between peaks and cleaner than those without atomic distortion, even when not incorporating a Gaussian convolution. Therefore, the dephasing effect modified the HHG spectrum in two ways, removing signals of HHG signals higher than 31st order, and increasing peak-to-peak depth for those signals that survive.

In Fig. 6, the blue calculation shows the HHG spectrum using atomic positions from a molecular dynamics simulation at 180 K. Now we find HHG signals up to the 49th order, where higher order signals are suppressed. This result at 180 K clearly indicates that it is important to keep the material cold in measuring HHG to observe higher order signals. We however note that the distortion of atomic positions is generated by the classical molecular dynamics calculation. At very low temperatures, quantum zero-point motion will contribute significantly and should be included in discussing the HHG spectra.

The results with the effective thermally-induced dephasing effect, incorporated through atomic positions generated by molecular dynamics simulations, suggest that sample cooling could be used to greatly improve the high-order harmonic signal intensity observed in experiment. In principle, this should not pose a problem to current experiment as liquid nitrogen or helium cooling is commonly utilized in a variety of condensed matter physics contexts.

IV. CONCLUDING REMARKS

We have presented here clear HHG spectra for diamond-like semiconductors interacting with near- and mid-IR pulsed electric fields. We obtain clear harmonic spectra for sufficiently long pulses with durations $T_{FWHM} \gtrsim 70$ fs and these spectra are further clarified through Gaussian convolution. We therefore review the suggestions made in previous literature considering diamond [31, 32] and silicon [34]. In crystalline Si, we do not observe the expected joint density of states effect but do demonstrate a prominent dephasing effect on the HHG spectra. Using a first-principles approach introducing dephasing into TDDFT we observe that higher-order harmonics are strongly suppressed by the decoherence effects in the target whilst lower order harmonics appear clearer in the spectra. On the basis of the strength of the decoherence effects for Si at 800 nm laser wavelength, we suggest that helium or liquid nitrogen cooling of the target will improve higher order harmonic intensity returns.

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