Single attosecond pulse generation for a dielectric driven by mid-infrared laser field

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Abstract
We theoretically investigate the high-order-harmonic generation (HHG) from solids by simulating the dynamics of a single active electron in a dielectric. Interband tunnelling is investigated, by which the harmonic generation can be modeled based on the classical trajectory analysis of electron-hole pairs. The supercontinuum region of the fundamental HHG plateau, whose intensity is about seven-order magnitude enhancement as compared with that of the second plateau, is used in the generation of isolated attosecond pulses. Thus, it provides us a practical way to increase the conversion efficiency of HHG with laser intensity below the damage threshold.

1. Introduction

The process of high-order-harmonic generation (HHG), which services as a source of coherent radiation at high frequencies, has been studied extensively in the past several decades, and has become one of the most important research areas in ultrafast atomic and molecular physics [1–3]. HHG is a fundamental way to produce single attosecond pulses, which are important tools for studying and controlling the motion of electrons inside atoms until now [4–6]. However, the intensity of the generated harmonics is still too low due to the high nonlinearity of the HHG process, which restricts its applications [7]. Recently, HHG has been generated from bulk crystals experimentally [7–10]. Due to the high density of solid-state materials, it is possible to produce HHG with higher efficiency.

The harmonic generation process in atomic and molecular gases can be described by a three-step semiclassical model [11], i.e., an electron first tunnels out of the Coulomb potential barrier suppressed by an external laser field and moves away from the ion. Then the freed electron is driven back and accelerated when the laser field direction is reversed. Finally it recombines with the ion, upon which harmonic radiation is emitted. As a comparison, HHG in solids can originate from two channels: interband and intraband currents [12]. Intraband contributions are thought as a result of Bloch oscillations in the same band [9, 13]. And interband currents are due to polarization buildup between valence band (VB) and conduction band (CB) [10, 14–18]. Interband HHG is similar to atomic HHG. Saddle point integration in the Keldysh-Lewenstein limit reveals the mechanism responsible for interband HHG: after the creation of an electron-hole pair by tunnel ionization, electrons and holes are accelerated and driven apart by the laser field. When an electron and a hole reencounter each other, recombination can occur resulting in the emission of a harmonic photon [17]. Therewith interband HHG opens the possibility to apply atomic attosecond technology to the condensed matter phase.

Until now, two main theoretical models, the many-and single-electron models, have been adopted in the studying of electron dynamics in solids. The many-electron model, which is based on the semiconductor Bloch equation, has been successfully applied in solids and semiconductor physics, such as semiconductor excitons and many-body correlations [19, 20]. Nevertheless, due to the interactions between electrons in solids, it is difficult to obtain accurate solutions in many-electron systems. Besides, it needs a long computing time to solve the related problems. The single-electron model, which treats the motion of each electron in solids independently in an effective potential, exhibits its advantages in addressing electron dynamics in solids. For
ideal crystals, atoms are arranged periodically. The electron dynamic problems can be solved as long as the effective potential is determined [21].

In this work, solid HHG spectra induced by interband tunnelling of a dielectric to intense few-cycle mid-infrared (MIR) laser pulses have been theoretically studied. 15 energy bands are used in the calculation in order to achieve numerical convergence. There shows a supercontinuum region in the fundamental HHG plateau, which is induced by population transfer between the second and third nethermost CBs. The supercontinuum region is dependent on the intensity of the driving MIR laser pulse. When the intensity is too low, no electrons can reach the Brillouin zone (BZ) edge and be excited onto band 4. And when the intensity is too strong, more electrons are excited onto the higher CBs. A peak intensity between 1.3 \times 10^{12} and 7.0 \times 10^{12} W \ cm^{-2} is available for the generation of a single attosecond pulse in the simulation. With increasing CEP of the driving laser pulse, the HHG spectra are periodically distributed with a π interval. Nevertheless, the supercontinuum region of the fundamental plateau is always been there, with changing harmonic orders. Thus, a single attosecond pulse can be synthesized at any carrier-envelope phase (CEP) of the driving MIR laser pulse. By optimizing the peak intensity and CEP of the driving laser pulse, an isolated attosecond pulse with a full width at half maximum (FWHM) of 500 as can be produced.

2. Simulation model

When comes to the strong-field regime, perturbation theory shows its shortcomings. And especially in the region where the Keldysh parameter \( \gamma \) is around 1, the polarization response of the chosen solid to the driving laser pulse is no longer linear and nonlinear [22, 23]. Therefore, we use the time-dependent Schrödinger equation (TDSE) to do the simulation. Our theoretical method is based on the single-active electron model, in which we describe the laser–crystal interaction in one-dimensional systems with laser polarization along the crystal plane. In the velocity gauge, the corresponding TDSE for an electron of quasimomentum \( \hbar k \) can be written as [21, 24]

\[
\frac{i\hbar}{\partial t} \psi_\hbar(x, t) = \left[ H_0 + \frac{e}{m_0} A(t) \mathbf{p}_k \right] \psi_\hbar(x, t).
\]

Where \( e \) and \( m_0 \) are the absolute values of the charge and mass of the electron. \( A(t) \) is the time-dependent vector potential of the driving MIR laser pulse. \( p_k = -i\hbar \partial / \partial x \) is the momentum operator. \( H_0 \) is the free-field crystal Hamiltonian, which can be expressed as \( H_0 = \frac{p_k^2}{2m_0} + U(x) \), and \( U(x) \) is our model potential. In order to obtain a bandgap close to that of SiO2, 8.95 eV [25], \( U(x) \) is defined as

\[
U(x) = -0.7 \left[ 1 + \tanh(x + 0.8) \right] \left[ 1 + \tanh(-x + 0.8) \right], \quad |x| \leq a_0 / 2. \tag{2}
\]

Here \( a_0 \) is the lattice constant, which is set as \( a_0 = 9.45 \) a.u..

The electron wave function \( \psi_\hbar(x, t) \) can be represented in the basis of Bloch states \( |\varphi_k^\alpha(x)\rangle \), which is evaluated by the following single-electron stationary Schrödinger equation

\[
H_0|\varphi_k^\alpha(x)\rangle = E_k^\alpha|\varphi_k^\alpha(x)\rangle. \tag{3}
\]

Here \( n \) is a band index. Ansatz \( |\varphi_k^\alpha(x)\rangle \) can be expressed as

\[
|\varphi_k^\alpha(x)\rangle = \sum_{j=1}^{N_{max}} C_{kj}^\alpha \exp[\I(k + K_j)x]. \tag{4}
\]

Here \( N_{max} \) is the plane waves number. And \( K_j \) denotes reciprocal lattice vectors, each of which can be written as \( K = mkb \), while \( m_k \) are integers and \( b = 2\pi/a_0 \) is the primitive vector of the reciprocal crystal lattice. The coefficients \( C_{kj}^\alpha \) in equation (4), as well as the field free energies \( E_k^\alpha \), are determined by diagonalizing the Hamiltonian \( H_0 \) in the basis of plane waves \( \exp(\I K_j x) \).

The general solution of equation (1) is written as

\[
|\psi_\hbar(x, t)\rangle = \sum_{n=1}^{N_{max}} \alpha_n^\alpha(t) |\varphi_k^\alpha(x)\rangle. \tag{5}
\]

By substituting ansatz (4) into equation (1), and since \( \langle \varphi_k^\alpha | \varphi_l^\beta \rangle = \delta_{\alpha\beta} \delta_{kl} \) the time-dependent coefficients \( \alpha_n^\alpha(t) \) are evaluated by solving the following system of equation

\[
\frac{i\hbar}{\partial t} \alpha_n^\alpha(t) = E_k^\alpha \alpha_n^\alpha(t) + \frac{e}{m_0} A(t) \sum_{l=1}^{N_{max}} R_k^{\alpha\beta} \alpha_l^\beta(t). \tag{6}
\]

Here \( R_k^{\alpha\beta} = \langle \varphi_k^\alpha | R_k | \varphi_l^\beta \rangle \).
The single-electron contributions to the current density over a unit cell can be expressed as

\[ j_k(t) = -\frac{e}{m_0} \Omega \int dx \{ \text{Re}\{\psi_k^*(x, t)p_k\psi_k(x, t)\} + eA(t) |\psi_k(x, t)|^2 \}. \quad (7) \]

Here \( \Omega \) is the volume of a unit cell. With the aid of equation (5), the single-electron current density can be presented as

\[ j_k(t) = -\frac{e}{m_0} \left\{ \text{Re}\left[ \sum_{q,p} (\alpha_{p,q}^k(t))^\dagger \alpha_p^q(t)p_k^\dagger \right] + eA(t) \right\}. \quad (8) \]

In laser fields, electrons oscillating in the VB have probabilities to tunnel to the CBs. Interband tunneling are related to the energy gap, only a small portion populated in the vicinity of \( k = 0 \) in band 2 can tunnel from the VB to the CBs with the laser parameters used in the current work. So the initial electron localization band is chosen as band 2 in the calculation, see figure 1, i.e., \( \alpha_2^k(t = 0.0) = 1.0 \) and \( \alpha_2^k(t = 0.0) = 0.0 \), here \( p = 3, 4, \ldots, N_{\text{max}} \). And the macroscopic electric current density can be written as \( j(t) = \int j_k(t)dk \).

The intensity of the harmonic emission at the \( N \)th harmonic frequency \( N\omega \) is dependent essentially on the absolute square of the Fourier transform of the single-electron current density

\[ S(N\omega) = |I(N\omega)|^2. \quad (9) \]

And an ultrashort pulse train can be obtained by superposing several orders \( (N_1 \leq N \leq N_2) \) of the harmonics

\[ I(t) = \left| \sum_{N=N_1}^{N_2} J(N\omega) \exp(iN\omega t) \right|^2. \quad (10) \]

Here \( J(N\omega) = N\omega \int_{-\infty}^{\infty} j(t) \exp(-iN\omega t)dt \). Before the Fourier transform, we multiply \( j(t) \) by a time-dependent Hanning window function \( \sin^2(\pi t/T) \) in order to increase the signal-noise ratio. Here \( T \) is the entire pulse duration of the external interaction laser pulse.

3. Simulation results and discussion

The vector potential of the linearly polarized MIR laser pulse is defined as

\[ A(t) = -E_0/\omega_0 \sin(\pi t/T_0)^4 \cos(\omega_0 t + \varphi_{\text{CEP}}). \quad (11) \]

Here \( E_0 \) is the amplitude of the electric field, \( \omega_0 \) and \( \varphi_{\text{CEP}} \) are the central frequency and CEP of the laser pulse, respectively. The external electric field acting on electrons can be definite as \( E(t) = -A' \).

The HHG spectra calculated by Bloch-state basis are presented in figure 2(a). The central wavelength, electric field amplitude, CEP, and total pulse duration of the driving laser pulse are 2.0 \( \mu \text{m} \), 0.008 a.u., 0.0, and 20 fs (3 optical cycles), respectively. 15 energy bands are used in the calculation in order to achieve numerical
convergence in solving the TDSE. In order to investigate the contributions of population transfer between different bands on HHG spectra, one CB is removed in the calculation (this can be done by setting the coefficient of \( \alpha_n^0 \) to zero in equation (6) at each evolution time step in solving the TDSE, and band \( n \) is removed). From this figure one can find two HHG plateaus when 15 energy bands are considered in the simulation. A second plateau emerges followed by another exponential drop (see the solid gray curve). The intensity of the first plateau is about five-order magnitude decrease when band 3 is dynamically excluded in the calculation, see the solid blue curve. We recover the known result that the fundamental HHG plateau is mainly determined by interband transitions between the nethermost CB (band 3) and the uppermost VB (band 2) [15, 26, 27]. The 'staircase' structure of HHG in our simulation comes from the contributions of the dynamics to the higher CBs, predominantly due to population transitions between CBs 4, 5, 6, and 7, as can be seen from the dash green, dash black, dash-dot red, and dot purple curves. When the plane wave is \( n \geq 7 \), the HHG spectra are stable. Electrons of the uppermost VB can not be excited onto a higher CB under such a situation.

There shows an abnormal enhanced supercontinuum region of the fundamental HHG plateau from the 32 to the 42 order when 15 energy bands are considered in the calculation, see the solid gray curve in figure 2(a). And the supercontinuum region can be used to synthesize an isolated attosecond pulse, the FWHM of which lasts about 560 as, as shown in figure 2(b). The intensity of the fundamental HHG plateau is about seven orders magnitude enhancement as compared with that of the second plateau. Thus, it provides us a practical way to increase the conversion efficiency of HHG with laser intensity below the damage threshold.

In order to further investigate the physical mechanisms behind these HHG spectra, we perform a time-profile analysis of the harmonics by using the Morlet wavelet [28], as shown in figure 3. Bands 4, 5, 6, and 7 are removed in the simulation for figures 3(a)–(d), respectively. By comparing the previous three figures, one can find that the supercontinuum region from the 32 to the 42 order of the first HHG plateau is mainly from the contributing of the short trajectory, which is induced by population transfer between bands 4 and 5. Each electron is tunnel ionized to an upper band predominantly at the minimum band gap, e.g., from band 2 to 3 at \( k = 0.0 \). And population transfer between bands 3 to 4 is mainly at the vicinity of the BZ edge [29–31]. For populations of C2, they move toward the top of the energy band when the vector potential is decreasing, for the negative correlation between \( E^c_k \) and \( k \) within the first BZ. When the vector potential is equal to zero, most populations can reach the top of C2 and then transit to C3 regardless of the laser field [29, 30]. Population transition between bands 4 and 5 just can happen after the inversion of the driving electric field, which contributes the short trajectory predominantly, as can be seen from figure 3(c). For population transition between the uppermost VB and the lowest CB, the spectral intensity of the long trajectories becomes stronger with increasing dephasing times [17]. Dephasing is lacked, e.g., the dephasing time is \( \tau = \infty \), in our model. Population transfer between bands 2 and 3 contributes the long trajectory predominantly, see figure 3(a).

Contributions of population transfer of the higher CB (\( n = 6 \)), which are mainly on the second HHG plateau, are weak enough to be ignored, as shown in figure 2(a). The time-frequency analysis profiles are almost consistent without and with band 6, see figures 3(c) and (d).

When the amplitude of the external electric field is \( E_0 = 0.004 \) a.u., population transfer between the uppermost VB and the three nethermost CBs is too low to generate a supercontinuum region at the fundamental
HHG plateau, see figure 4(a1). The synthesized attosecond pulses by the harmonics of the fundamental plateau is of weak intensity, as can be seen from figure 4(a2). When $E_0/\omega < \pi/\alpha_0$, no electrons can reach the BZ edge. They oscillate in the first CB without a further excitation [29]. And when $E_0 = 0.016$ a.u., more electrons are excited onto the higher CBs ($n \geq 5$). The increased long path contributions induce the disappearance of the supercontinuum region of the first plateau, see figure 4(b1). And there show many other peaks around the main peak in the temporal profile of the attosecond pulses by synthesizing the harmonics of the fundamental plateau, as shown in figure 4(b2). A driving laser pulse with a strength between 0.006 a.u. (the corresponding peak intensity is $1.3 \times 10^{12}$ W cm$^{-2}$) and 0.014 a.u. (the corresponding peak intensity is $7.0 \times 10^{12}$ W cm$^{-2}$) is suitable for the supercontinuum region generation in the simulation.

When the driving pulse is short and intense, significant CEP effects, which can be interpreted as interference between pathways requiring different numbers of photons, would be produced in molecule-laser interactions [32, 33]. In figure 5(a), we show the HHG spectra with different CEPs of the driving laser pulse. 15 energy bands are used in the calculation. The central wavelength and amplitude of the interaction laser pulse are 2.0 $\mu$m and 0.008 a.u., respectively. From this figure one can find that, with increasing CEP, the laser-dielectric interaction HHG spectra are periodically distributed with an interval of $\pi$. Nevertheless, the supercontinuum region of the fundamental plateau is always there, with changed harmonic orders. Therefore, a single attosecond pulse can be synthesized at any CEP of the driving MIR pulse. For instance, when the CEP is $\varphi_{\text{CEP}} = 0.5\pi$, the supercontinuum region of the first plateau lasts among the 23 and 35 orders. And an isolated attosecond, whose FWHM lasts about 500 as, can be synthesized with this supercontinuum region, see figure 5(b).

### 4. Summary

In conclusion, solid HHG spectrum induced by interband tunnelling of a dielectric to intense few-cycle MIR laser pulses is theoretically studied with TDSE in the Bloch Basis. Interband tunnelling is investigated, by which the HHG spectra can be modeled based on the classical trajectory analysis of electron-hole pairs. The simulation results show that there shows a supercontinuum region in the fundamental HHG plateau, which can be used to synthesize an isolated attosecond pulse. The supercontinuum region, which is predominantly induced by...
population transfer between bands 4 and 5, is dependent on the intensity of the driving MIR laser pulse. When the peak intensity is too low, no electrons can reach the BZ edge and be excited onto band 4. And when the intensity is too strong, more electrons are excited onto the higher CBs. A peak intensity between $1.3 \times 10^{12}$ and $7.0 \times 10^{12}$ W cm$^{-2}$ is available for the generation of a single attosecond pulse in the simulation. With increasing CEP of the driving pulse, the HHG spectra show periodic distributions with a $\pi$ interval. Nevertheless, the supercontinuum region of the fundamental plateau is always been there, with changing harmonic orders. Thus, a single attosecond pulse can be synthesized at any CEP of the driving MIR laser pulse.

**Figure 4.** Upper panels: high-order-harmonic spectra induced by laser-dielectric interactions. Lower panels: temporal profiles of the attosecond pulses by synthesizing the harmonics of the fundamental plateaus of the upper panels. 15 energy bands are used in the calculation. The central wavelength and CEP of the driving laser pulse are 2.0 $\mu$m and 0.0, respectively. And the amplitudes of the electric field are 0.004 a.u. for (a1) and (a2), and 0.015 a.u. for (b1) and (b2), respectively.

**Figure 5.** (a) High-order-harmonic spectra with different CEPs. (b) Temporal profile of the attosecond pulse by synthesizing the harmonics with orders 23–35. The CEP of the driving pulse is $\pi/2$. 15 energy bands are used in the calculation. The central wavelength and amplitude of the interaction laser pulse are 2.0 $\mu$m and 0.008 a.u., respectively.
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