Theoretical study of multiple high-order harmonic generation by intense ultrashort pulsed laser fields: A new generalized pseudospectral time-dependent method

Xiao-Min Tong, Shih-I Chu*
Department of Chemistry, University of Kansas and Kansas Center for Advanced Scientific Computing, Lawrence, KS 66045, USA

Received 5 December 1996

Abstract

We present a new time-dependent method for accurate and efficient non-perturbative treatment of multiple high-order harmonic generation (HHG) in intense laser fields. The time development of the wavefunction is obtained by a new split-operator time-propagation technique in the energy representation. The generalized pseudospectral technique is extended to perform an optimal spatial grid discretization, leading to significant improvement of the quality of the wavefunction over that obtained by the equal-spacing spatial discretization techniques. The accuracy of the method is demonstrated by several benchmark calculations including the excellent agreement of the HHG power spectra in length and acceleration forms. The method is applied to a detailed study of the coherent control of HHG spectra of atomic hydrogen in intense ultrashort pulsed laser fields with or without the chirp. Several novel chirp-dependent and pulse-duration-dependent phenomena are predicted. Comparison with available experimental observation is made. © 1997 Elsevier Science B.V.

1. Introduction

The study of the non-linear response of atoms and molecules to intense laser fields is a subject of much current interest and significance in atomic, molecular, and optical physics and in chemical physics [1]. In this article we focus on the theoretical study of one of such high-intensity processes of technological importance, namely, multiple high-order harmonic generation (HHG) in intense laser fields. The generation of harmonics of orders well in excess of 100 from noble gas targets has been demonstrated by several recent experiments using ultrahigh intensity pump lasers [2–4]. Experimental observations of high-order harmonic generation in molecular gases have been also recently reported [5,6]. In these atomic or molecular HHG processes, typically only the odd harmonics are observed and the harmonic spectrum is characterized by an initial decline in intensity with increasing harmonics of similar brightness, and finally a sharp cut-off beyond which no further harmonic emission is seen. A quasiclassical model [7,8] has been proposed to explain the qualitative nature of HHG processes. It is found [9] that for single atom (molecule) response, the cutoff occurs at the energy $I_p + 3.2U_p$, where $I_p$ is the ionization potential and $U_p = e^2F^2/4m_e\omega^2$ is the ponderomotive potential, i.e. the mean kinetic energy acquired by a free electron in a laser field of amplitude $F$ and frequency $\omega$. Such a prediction is expected to
hold better in the so-called tunneling regime for ionization. This regime is characterized by the Keldysh parameter \[ \gamma = \left( \frac{I_p}{2U_p} \right)^{1/2} \]. \( \gamma < 1 \) indicates the tunneling regime, whereas \( \gamma > 1 \) denotes the multiphoton regime. However, the location of the cut-off can be modified when collective effects (phase matching) become relevant \[11\]. Deviations from the cut-off law have been reported in several experiments.

The theoretical approaches of HHG can be divided into two general non-perturbative approaches: (a) the time-independent non-Hermitian Floquet Hamiltonian methods for one-laser \[12-14\] or two-laser \[14\] cases, and (b) direct numerical integration of the time-dependent Schrödinger equation \[9,15\]. The time-independent and time-dependent methods are complementary techniques. The advantages of the time-independent Floquet approaches lie in their numerical accuracy and computational efficiency. For example, in one of our recent Floquet studies of one- and two-color HHG of atomic hydrogen \[14\] using the complex-scaling generalized pseudospectral (CS-GSP) technique \[16,17\] for the discretization of the non-Hermitian Floquet Hamiltonian \[18-20\], it was demonstrated that excellent agreement of the HHG power spectra obtained from the length and the acceleration forms can be achieved, spanning at least 20 orders of magnitude. (Other recent applications of the time-independent non-Hermitian Floquet formalism using the CSGPS discretization techniques include high-intensity processes such as multiphoton detachment of \( H^- \) \[16\], angular distributions and partial rates in above-threshold multiphoton detachment of \( H^- \) in two-color laser fields \[21\], stabilization and ionization suppression of negative ions in intense laser fields \[22\], laser-induced molecular stabilization and chemical bond hardening in one-color \[23\], pulsed \[24\], and two-color \[25\] laser fields, and the energy spectra of above-threshold multiphoton dissociation of diatomic molecules \[26\], etc.) Such an accuracy is in general difficult to achieve by the time-dependent methods, partly due to the non-optimal spatial discretization and partly due to the numerical errors in the wavefunctions accumulated from the time propagation. For example, previous time-dependent calculations \[27,28\] showed that the length and acceleration power spectra for higher harmonics can be quite different and high-precision wavefunctions are required to obtain reliable high-order harmonic generation spectra. (Note that in the time-dependent approaches, the acceleration form of the induced dipole moment is found to be generally superior to the length form \[27-29\] because the dominant contributions to the HHG arise from the wavefunctions at short distances in the acceleration-form calculations.) Nevertheless, the time-dependent methods have the advantages that they are more readily applicable to any time-dependent Hamiltonians with arbitrarily shaped laser fields.

The motivations of this paper are twofold: (a) Firstly we shall present a new time-dependent method for numerical integration of the Schrödinger equation which is computationally efficient and capable of providing more accurate wavefunctions for reliable HHG calculations. The significant improvement of the time-dependent wavefunctions is achieved both by the use of a generalized pseudospectral method for a more optimal discretization of the spatial coordinates and by a new time propagation method presented. (b) Secondly, we shall extend the new method for the exploration of the dynamics and coherent control of HHG by intense ultrashort laser fields. This is prompted by a recent experimental observation on the enhancement and tunability of HHG processes by means of ultrashort chirped laser fields \[30\].

In the next section (Section 2), we shall present the details of the method. The accuracy and computational advantages of the procedure are examined in Section 3. Then we extend the procedure for a detailed investigation of HHG spectrum in intense ultrashort pulsed laser fields with or without the chirp. This is followed by a conclusion in Section 4.

2. The method

In this section we shall consider the numerical solution of the time-dependent Schrödinger equation for atomic systems in intense laser fields. While the discussion below is limited to one-electron systems, the methodology presented is general and may be extended to more complex systems as well. The Schrödinger equation for atomic hydrogen in the presence of linearly polarized fields \( (F \parallel \mathbf{z}) \) can be written as (in atomic units)

\[
i \frac{\partial}{\partial t} \psi(r, t) = \hat{H} \psi(r, t) = [\hat{H}_0 + \hat{V}] \psi(r, t).
\]
Here $\hat{H}_0$ is the unperturbed H-atom Hamiltonian and $\hat{V}$ is the atom–field interaction:

\[ \hat{H}_0(r) = -\frac{1}{2} \frac{d^2}{dr^2} + \frac{\hat{L}^2}{2r^2} - \frac{1}{r}, \]
\[ \hat{V}(r, t) = -F \cdot r f(t) = -F \varepsilon f(t), \]

where $F$ is the electric field amplitude and $f(t)$ the pulse shape of the laser field. We shall extend the second-order split-operator technique in spherical coordinates \[28,31\] for the time propagation of the Schrödinger equation:

\[ \psi(r, t + \Delta t) \approx \exp \left( -i \hat{H}_0 \Delta t / 2 \right) \]
\[ \times \exp \left( -i \hat{V}(r, \theta, t + \Delta t / 2) \Delta t \right) \]
\[ \times \exp \left( -i \hat{H}_0 \Delta t / 2 \right) \psi(r, t) + O(\Delta t^3). \]  

(4)

Such an expression is different from the traditional split-operator techniques \[28,31,32\], where $\hat{H}_0$ is usually chosen to be the radial kinetic energy operator and $\hat{V}$ the remaining Hamiltonian depending on the spatial coordinates only. The advantage of using the energy-representation expression in Eq. (4) will be made clearer in the following discussion.

Eq. (4) shows that the time propagation of the wavefunction from $t$ to $t + \Delta t$ is achieved by three steps: (i) First the wavefunction $\psi(r, t)$ is propagated for a half-time step $\Delta t / 2$ in the energy space spanned by $\hat{H}_0$:

\[ \exp \left( -i \hat{H}_0 \Delta t / 2 \right) \psi(r, t) \equiv \psi_1(r, t). \]  

(5)

(ii) Then the wavefunction $\psi_1(r, t)$ is transformed back to the coordinate space and propagated for a time step $\Delta t$ under the influence of the atom–field coupling:

\[ \exp \left( -i \hat{V}(r, \theta, t + \Delta t / 2) \Delta t \right) \psi_1(r, t) \equiv \psi_2(r, t). \]  

(6)

(iii) Finally, the wavefunction $\psi_2(r, t)$ is transformed back to the energy space spanned by $\hat{H}_0$ and propagated for another half-time step $\Delta t / 2$:

\[ \exp \left( -i \hat{H}_0 \Delta t / 2 \right) \psi_2(r, t) = \psi(r, t + \Delta t). \]  

(7)

To pursue the time propagation, we expand the total wavefunction $\psi(r, t)$ in Legendre polynomials \[28,31\],

\[ \psi(r_i, \theta_j, t) = \sum_{l=0}^{l_{\text{max}}} g_l(r_i) P_l(\cos \theta_j), \]

(8)

where the $P_l$’s are the normalized Legendre polynomials. $g_l(r, t)$ can be determined accurately by the Gauss–Legendre quadrature

\[ g_l(r_i) = \sum_{k=1}^{L+1} w_k P_l(\cos \theta_k) \psi(r_i, \theta_k, t), \]

(9)

where $\{\cos \theta_k\}$ are the $L + 1$ zeros of the Legendre polynomial $P_{L+1}(\cos \theta_k)$ and $\{w_k\}$ are the corresponding quadrature weights. The propagation in $\hat{H}_0$ energy space (step (i) above) can now be expressed as

\[ \exp \left( -i \hat{H}_0 \Delta t / 2 \right) \psi(r_i, \theta_j, t) \]
\[ = \sum_{l=0}^{l_{\text{max}}} \left[ \exp \left( -i H_0^0 \Delta t / 2 \right) g_l(r_i, t) \right] P_l(\cos \theta_j), \]

(10)

with

\[ H_0^0 = -\frac{1}{2} \frac{d^2}{dr^2} + \frac{l(l + 1)}{2r^2} - \frac{1}{r}. \]

Note that in Eq. (10), each partial-wave wavefunction component $g_l$ is propagated independently under individual $\hat{H}_0$ energy space, leading to efficient propagation of the total wavefunction in step (i).

The key step in the time propagation of Eq. (10) is to construct the evolution operator

\[ \exp \left( -i H_0^0 \Delta t / 2 \right) \equiv S(l), \]

(11)

through an accurate representation of $\hat{H}_0^0$. Thus we first seek an optimal grid discretization method for accurate and efficient solution of the eigenproblem:

\[ \hat{H}_0^0(r) \chi_l(r) = \epsilon_l \chi_l(r). \]

(12)

For atomic structure calculations involving the Coulomb potential, one typical problem associated with commonly used equal-spacing grid methods is the Coulomb singularity at $r = 0$ and the long-range nature of the potential. Generally one truncates the semi-infinite $(0, \infty)$ domain into finite domain $[r_{\text{min}}, r_{\text{max}}]$ to avoid the Coulomb singularity at the origin and the infinite domain. For this purpose, $r_{\text{min}}$
must be chosen sufficiently small and $r_{\text{max}}$ sufficiently large. This results in the need of a large number of grid points. Further, extreme care must be exercised to ensure the wavefunctions obtained from such discretization is of sufficient accuracy for performing reliable high-order harmonic generation calculations. Here we extend a generalized pseudospectral (GPS) method \cite{16,17} for optimal discretization of the radial coordinates to overcome some of the above-mentioned problems. The GPS method has been recently applied to the time-independent non-Hermitian Floquet studies of HHG \cite{14}. The results show excellent agreement of the length and acceleration power spectra spanning over 20 orders of magnitude.

In the following we briefly describe the essence of the GPS method and its extension to the current HHG studies. The first step is to map the semi-infinite domain $[0, \infty)$ or $[0, r_{\text{max}}]$ into the finite domain $[-1, 1]$ using a non-linear mapping $r = r(x)$, followed by the Legendre or Chebyshev pseudospectral discretization \cite{33}. This allows for denser grids near the origin, leading to more accurate eigenvalues and eigenfunctions and the use of a considerably smaller number of grid points than those of the equal-spacing grid methods. A suitable algebraic mapping for atomic structure calculations is provided by the following form \cite{16,17,33}:

$$r = r(x) = \frac{L}{1 - x^2} + x,$$  

(13)

where $L$ and $\alpha = 2 \times L/r_{\text{max}}$ are mapping parameters. The introduction of non-linear mapping usually leads to either an asymmetric or a generalized eigenvalue problem. Such undesirable features may be removed by the use of a symmetrization procedure recently introduced \cite{16,17}. Thus by defining

$$\phi_i(r) = \sqrt{r^2(x)} \chi_i(r(x)),$$  

(14)

one finds the transformed Hamiltonian possesses the following symmetrized form:

$$\hat{H}_{\text{I}}^0(x) = -\frac{1}{2} \frac{d^2}{dx^2} \frac{1}{r'(x)} + V_i(r(x)),$$  

(15)

where $V_i = \frac{l(l+1)}{2r^2} - \frac{1}{r}$, leading Eq. (15) to a symmetric eigenvalue problem. In the Legendre pseudospectral method, the collocation points $\{x_i\}$ are the roots of the polynomials $P_{N+1}^l(x)$, where $N$ is the total number of grid points used in the discretization. In such a discretized scheme, the Hamiltonian $\hat{H}_{\text{I}}^0(x)$, Eq. (15), can be represented by the following matrix form:

$$[\hat{H}_{\text{I}}^0]_{ij} = (D_2)_{ij} + V_i(x_i) \delta_{ij},$$  

(16)

with

$$(D_2)_{ij} = \frac{1}{r'(x)} \frac{(N + 1)(N + 2)}{6(1 - x_i^2)} \frac{1}{r'(x)}, \quad i = j,$$  

(17)

$$(D_2)_{ij} = \frac{1}{r'(x)} \frac{1}{(x_i - x_j)^2} \frac{1}{r'(x)}, \quad i \neq j.$$  

(18)

The eigenvalues and eigenfunctions of $\hat{H}_{\text{I}}^0$ will be denoted by $\{\epsilon_k(l)\}$ and $\{\chi_k(l)\}$, respectively. The propagation of a given partial-wave function $g_l(r,t)$ under $\hat{H}_{\text{I}}^0$ can now be expressed as

$$[\exp(-i\hat{H}_{\text{I}}^0 \Delta t/2) g_l(r)]_i = \sum_{j=1}^{N} S_{ij}(l) g_l(r_j),$$  

(19)

where

$$S_{ij}(l) = \sum_k \chi_{k\ell}(l) \chi_{kj}(l) \exp(-i\epsilon_k(l) \Delta t/2).$$  

(20)

Note that $S(l)$ is a complex symmetric matrix and it needs to be computed only once. The time propagation is therefore reduced to the matrix-vector product which can be performed efficiently using the BLAS (basic linear algebra subroutines).

Once the first step of time propagation, Eq. (5), is accomplished to obtain $\psi_1(r,t)$, we can perform the second step of time propagation by first transforming $\psi_1(r,t)$ to the $(r, \theta)$ coordinate representation and propagate Eq. (6) by a simple multiplication to obtain $\psi_2(r,t)$. Finally, the last step of time propagation, Eq. (7), is performed by repeating step (1). This completes the time propagation of the total wavefunction from $t$ to $t + \Delta t$.

Once the time-dependent wavefunction is determined, one can compute the induced dipole moment in either the length or the acceleration form:

$$d(t) = \langle \psi(r,t) | z | \psi(r,t) \rangle,$$  

(21)
\[ d_A(t) = \langle \psi(r,t) \mid \frac{d^2}{dt^2} \psi(r,t) \rangle \]

\[ = \langle \psi(r,t) \mid -\frac{z}{r^3} + F_f(t) \mid \psi(r,t) \rangle. \]  

(22)

And the corresponding power spectrum can be obtained by the Fourier transformation of the time-dependent dipole moments:

\[ P(\omega) = \left| \frac{1}{t_f - t_i} \int_{t_i}^{t_f} d(t) e^{-i\omega t} \right|^2, \]  

(23)

or

\[ P_A(\omega) = \left| \frac{1}{t_f - t_i} \frac{1}{\omega^2} \int_{t_i}^{t_f} d_A(t) e^{-i\omega t} \right|^2. \]  

(24)

The power spectra \( P(\omega) \) and \( P_A(\omega) \) should be the same if the wavefunction \( \psi(r,t) \) is fully converged. In the presence of either periodic fields or continuous-wave (cw) lasers, one can further obtain the time-independent \( n \)-th order harmonic generation emission rate by means of the expression

\[ \Gamma_n = \frac{4\omega_0^3 n^3}{3c^3} P(n\omega_0), \]  

(25)

or

\[ \Gamma_n = \frac{4\omega_0^3 n^3}{3c^3} P_A(n\omega_0), \]  

(26)

where \( \omega_0 \) is the fundamental frequency of the laser field.

### 3. Results and discussions

In this section we shall apply the time-dependent method to the study of multiple high-order harmonic generation (HHG) in intense laser fields. First we shall test the accuracy of the wavefunctions by performing (a) multiphoton ionization of atomic H, and (b) high-order harmonic generation of atomic H, both in monochromatic fields, and compare the results with available time-independent Floquet calculations. Then we will provide a detailed discussion of the HHG study in intense ultrashort pulsed laser fields. Comparison with experimental observations will be made.

#### 3.1. Multiphoton ionization rates

As the first test, we calculate the multiphoton ionization of atomic H in monochromatic laser fields for a wide range of laser frequencies and intensities and compare the results with the time-independent non-Hermitian Floquet calculations [34]. For the present time-dependent calculations, the laser field is ramped on according to

\[ E(t) = F_f(t) = \begin{cases} F \sin^2 \left( \frac{\pi t}{20T} \right) \sin \omega_0 t, & t \leq 10T, \\ F \sin \omega_0 t, & t \geq 10T, \end{cases} \]  

(27)

where \( T = 2\pi/\omega_0 \). The total ionization rate is evaluated from the decay of the time-dependent correlation function or the survival probability

\[ C(t) = \langle \psi(r,0) \mid \psi(r,t) \rangle. \]  

(28)

Table 1 shows some typical results for the total ionization rates for one-photon dominant (\( \omega = 0.60 \) a.u.),

<table>
<thead>
<tr>
<th>( \omega_0 ) (a.u.)</th>
<th>Total Ionization Rate (in a.u.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.6</td>
<td>3.14 × 10^-3</td>
</tr>
<tr>
<td>0.3</td>
<td>1.63 × 10^-3</td>
</tr>
<tr>
<td>0.2</td>
<td>3.50 × 10^-3</td>
</tr>
</tbody>
</table>

\( \omega_0 = 0.6 \) a.u., \( \omega_0 = 0.3 \) a.u., \( \omega_0 = 0.2 \) a.u.
two-photon dominant ($\omega = 0.30$ a.u.), and three-photon dominant ($\omega = 0.2$ a.u.) ionization processes for the case of laser field strength $F_{\text{rms}} = 0.05$ a.u. (1 atomic unit of $F_{\text{rms}}$ is equivalent to $7 \times 10^{16}$ W/cm$^2$ laser intensity.) It is seen that the present time-dependent results are nearly identical to the Floquet results [34].

### 3.2. HHG of atomic H in monochromatic fields

As another test of the accuracy of the present method, we perform the time-dependent HHG calculations in monochromatic fields. This provides a more stringent test on the quality of the wavefunctions. The field is turning on using a 10-optical-cycle sine-square ramp similar to that of Eq. (27) and then the wavefunction is propagated for another 25 optical cycles under a constant peak field strength. The last 10 optical cycles are used to calculate the power spectrum. Table 2 lists the HHG results from the present time-dependent calculations using both the length and the acceleration forms of the induced dipole moment. Also shown in Table 2 are the results from the time-independent non-Hermitian Floquet calculations [14] for comparison. The laser wavelength and intensity used are 775 nm and $3 \times 10^{13}$ W/cm$^2$, respectively. It is seen that the agreement of the length-form power spectrum with the acceleration-form power spectrum from the time-dependent calculations are very good, well within a few percents, demonstrating the accuracy of the time-dependent wavefunctions. To our knowledge, this is the first time-dependent HHG calculations of atomic systems that have achieved such a quantitative agreement. It is also gratifying to see that the time-dependent results agree well with the time-independent Floquet results, considering very different numerical procedures are used in these calculations.

### 3.3. Comparison with other split-operator time-propagation methods

It is instructive to discuss here the comparison of the present time-dependent split-operator technique (using the pseudospectral radial discretization and energy representation), denoted by SOPS, with the conventional split-operator method (using equal-spacing spatial discretization and fast Fourier transformation (FFT) techniques) [28,31], denoted by SOFFT. In terms of the computational speed, the FFT algorithm allows in principle faster time-step propagation than the present matrix-vector product algorithm in the SOPS calculations. However, we found that the overall computational speed in the SOPS method is still faster to considerably faster than that of the SOFFT method. This can be attributed to the special features of the proposed SOPS method: (a) each partial-wave wavefunction is propagated separately in the first step (i), Eq. (10), (b) the number of grid points needed is considerably smaller than that required for the SOFFT method, and (c) the time step $\Delta t$ used in the SOPS method can be at least an order of magnitude larger than that used in the SOFFT method. The substantial increment of the time step is made possible in the SOPS method since we can eliminate those (extreme) high-energy eigenvalues of each partial-wave Hamiltonian in Eq. (12) before performing the time propagation. Such (extreme) high-energy components contribute negligibly to the overall dynamics but the inclusion of them can impose severe time step constraints. As an example, in a test calculation of HHG in monochromatic fields using 20 partial waves, each partial-wave wavefunction is discretized with $N = 200$ grid points in the SOPS calculations, and $N = 512$ in the SOFFT calculations. The time step $\Delta t = 0.01$ a.u. is required in the SOFFT method to achieve numerical convergence, while the SOPS method allows the use of $\Delta t = 0.1$ a.u. The overall computational speed of such a SOPS calculation is found to be about a factor of 5 faster than that of the SOFFT calculation. Another major advantage of
Table 3
Comparison of the power spectra calculated from the length and acceleration forms in $\sin^2$ pulse for the case of $I = 5 \times 10^{13}$ W/cm$^2$, and $\lambda = 1064$ nm. The total pulse length is 60 optical cycles. $n$ is the order of harmonic generation peak.

<table>
<thead>
<tr>
<th>$n$</th>
<th>$P(n\omega_0)$, in a.u.</th>
<th>$P_A(n\omega_0)$, in a.u.</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>9.85[-07]</td>
<td>9.89[-07]</td>
</tr>
<tr>
<td>5</td>
<td>2.32[-08]</td>
<td>2.41[-08]</td>
</tr>
<tr>
<td>7</td>
<td>2.06[-09]</td>
<td>1.98[-09]</td>
</tr>
<tr>
<td>9</td>
<td>1.05[-08]</td>
<td>1.03[-08]</td>
</tr>
<tr>
<td>11</td>
<td>8.94[-09]</td>
<td>9.02[-09]</td>
</tr>
<tr>
<td>13</td>
<td>8.70[-10]</td>
<td>8.71[-10]</td>
</tr>
<tr>
<td>15</td>
<td>6.94[-10]</td>
<td>6.95[-10]</td>
</tr>
<tr>
<td>17</td>
<td>2.97[-10]</td>
<td>2.97[-10]</td>
</tr>
<tr>
<td>19</td>
<td>1.08[-10]</td>
<td>1.08[-10]</td>
</tr>
<tr>
<td>21</td>
<td>5.21[-11]</td>
<td>5.23[-11]</td>
</tr>
<tr>
<td>23</td>
<td>5.74[-11]</td>
<td>5.74[-11]</td>
</tr>
<tr>
<td>27</td>
<td>1.64[-11]</td>
<td>1.65[-11]</td>
</tr>
<tr>
<td>29</td>
<td>5.10[-12]</td>
<td>5.11[-12]</td>
</tr>
<tr>
<td>31</td>
<td>7.04[-13]</td>
<td>7.02[-13]</td>
</tr>
<tr>
<td>33</td>
<td>5.58[-14]</td>
<td>5.40[-14]</td>
</tr>
</tbody>
</table>


the SOPS method lies in its capability of providing more accurate time-dependent wavefunctions for performing reliable higher-order harmonic generation calculations, a main goal of this paper.

3.4. HHG in intense ultrashort chirped laser pulses

Having established the validity and accuracy of the new time-dependent method, we now consider the application of the technique to the study of HHG driven by ultra-short intense chirped laser fields. The electric field of the chirped pulses used in the investigation has the following time-dependent form:

$$E(t) = Ff(t) = F \sin^2 \left( \frac{\pi t}{nT} + \frac{\pi}{2} \right) \sin(\omega_0 t + c_r t^2),$$

$$-\frac{1}{2}nT \leq t \leq \frac{1}{2}nT,$$

(29)

where $n$ is the pulse length measured in optical cycle ($T = 2\pi/\omega_0$), and $c_r$ is the chirp parameter. The factor $\pi/2$ is introduced to shift the pulse center position to $t = 0$. Given below are some results for the case of linearly polarized pulsed fields with intensity $5 \times 10^{13}$ W/cm$^2$ and wavelength 1064 nm. The pulse length used is 60 optical cycles.

Fig. 1 shows the time-dependent induced dipole moments in length and acceleration forms, $d(t)$ and $d_A(t)$, respectively, for the case of $c_r = 0.0$. The power spectrum is obtained by performing the Fourier transformation of the respective time-dependent dipole moment according to Eqs. (23)–(24). The results are shown in Fig. 2. It is seen that the length-form power spectrum $P(\omega)$ (dotted line) and the acceleration-form power spectrum $P_A(\omega)$ (solid line) are nearly identical for all the fine details of line shapes for each harmonic peak and the agreement extends all the way to the point of cut-off. Beyond that point, the length-form power spectrum levels off and the acceleration-form power spectrum provides more accurate declining background. The excellent agreement of the length-form and acceleration-form results can
be seen more clearly by a comparison of the intensity of each individual harmonic peak shown in Table 3. This justifies further the accuracy and computational advantages of the SOPS method.

We now discuss the effects on the HHG spectrum by introducing the chirp into the laser pulses. Figs 3(a)–3(d) show the length-form power spectra corresponding to the chirps $c_r = 0.0, -1.0, -2.0, \text{and} -3.0$ (in units of $4.86 \times 10^{-8}$ a.u.), respectively. The cases of positive chirps are shown in Figs 4(a)–4(d) for the cases of $c_r = 4.0, 3.0, 2.0, \text{and} 1.0$ (same units as in Fig. 3), respectively. Generally speaking, we see that the higher-order peaks for negative chirps are broader than those of positive chirps. Among the negative chirp cases, the higher-order peaks become broader as the chirp amount is increased. However, for the positive chirp cases, the higher-order peaks become sharper as the amount of chirp is increased from 1.0 (Fig. 4(d)) to 3.0 unit (Fig. 4(b)), beyond which the peaks seem to become broader (Fig. 4(a)) again. To see more details of the subtlety of the effects of introducing the chirp, we give in Table 4 the ratio of individual harmonic peak intensity (for various $c_r$) to that of the case without the chirp ($c_r = 0$). A striking trend is revealed in Table 4: the positive-chirp laser pulses tend to suppress the lower-order harmonic intensities but enhance those of the higher-order harmonics. The enhancement of the higher-order harmonics reaches a maximum at $c_r = 2.0$ unit. For the negative-chirp pulsed laser, there is significant suppression of the higher-order harmonics. Some of our prediction has been also reported in a recent experimental study of HHG of rare gas atoms using ultrashort chirped laser fields [30].

3.5. The effect of varying the pulse length on HHG spectrum

It is also instructive to study the effect of varying the pulse length on the HHG spectrum. Figs 5(a)–5(d) show the HHG power spectra driven by a $\sin^2$ pulse (of the form Eq. (29) but without the chirp ($c_r = 0$)) for the pulse length $\tau$ equal to 40, 60, 80, and 100 optical cycles.
Table 4
The ratio of the intensity of individual harmonic generation peak for the pulsed laser fields with the chirp \( c_r \neq 0 \) versus the case without the chirp \( c_r = 0 \). All the pulses have the same pulse length (60 optical cycles), laser intensity \( 5 \times 10^{13} \text{ W/cm}^2 \), and fundamental wavelength (1064 nm). The chirp \( c_r \) is in units of \( 4.86 \times 10^{-8} \text{ a.u.} \)

<table>
<thead>
<tr>
<th>( n )</th>
<th>( c_r = -1.0 )</th>
<th>( c_r = -2.0 )</th>
<th>( c_r = -3.0 )</th>
<th>( c_r = 1.0 )</th>
<th>( c_r = 2.0 )</th>
<th>( c_r = 3.0 )</th>
<th>( c_r = 4.0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>0.97</td>
<td>0.89</td>
<td>0.78</td>
<td>0.95</td>
<td>0.85</td>
<td>0.74</td>
<td>0.63</td>
</tr>
<tr>
<td>5</td>
<td>0.99</td>
<td>0.91</td>
<td>0.79</td>
<td>0.92</td>
<td>0.80</td>
<td>0.70</td>
<td>0.61</td>
</tr>
<tr>
<td>7</td>
<td>1.23</td>
<td>1.41</td>
<td>1.42</td>
<td>0.78</td>
<td>0.59</td>
<td>0.40</td>
<td>0.28</td>
</tr>
<tr>
<td>9</td>
<td>1.21</td>
<td>1.14</td>
<td>0.90</td>
<td>0.74</td>
<td>0.56</td>
<td>0.44</td>
<td>0.36</td>
</tr>
<tr>
<td>11</td>
<td>1.01</td>
<td>0.82</td>
<td>0.60</td>
<td>0.82</td>
<td>0.63</td>
<td>0.49</td>
<td>0.40</td>
</tr>
<tr>
<td>13</td>
<td>1.21</td>
<td>0.96</td>
<td>0.72</td>
<td>0.66</td>
<td>0.46</td>
<td>0.53</td>
<td>0.48</td>
</tr>
<tr>
<td>15</td>
<td>0.86</td>
<td>0.53</td>
<td>0.31</td>
<td>0.72</td>
<td>0.56</td>
<td>0.49</td>
<td>0.40</td>
</tr>
<tr>
<td>17</td>
<td>1.03</td>
<td>0.77</td>
<td>0.61</td>
<td>0.40</td>
<td>0.66</td>
<td>0.87</td>
<td>0.79</td>
</tr>
<tr>
<td>19</td>
<td>0.50</td>
<td>0.28</td>
<td>0.19</td>
<td>1.08</td>
<td>0.50</td>
<td>0.44</td>
<td>0.39</td>
</tr>
<tr>
<td>21</td>
<td>0.43</td>
<td>0.40</td>
<td>0.36</td>
<td>1.80</td>
<td>1.21</td>
<td>0.64</td>
<td>0.88</td>
</tr>
<tr>
<td>23</td>
<td>0.64</td>
<td>0.45</td>
<td>0.30</td>
<td>1.45</td>
<td>0.96</td>
<td>0.81</td>
<td>0.66</td>
</tr>
<tr>
<td>25</td>
<td>0.58</td>
<td>0.40</td>
<td>0.30</td>
<td>1.80</td>
<td>2.27</td>
<td>1.53</td>
<td>1.11</td>
</tr>
<tr>
<td>27</td>
<td>0.69</td>
<td>0.52</td>
<td>0.41</td>
<td>1.56</td>
<td>2.06</td>
<td>1.87</td>
<td>1.34</td>
</tr>
<tr>
<td>29</td>
<td>0.75</td>
<td>0.59</td>
<td>0.48</td>
<td>1.40</td>
<td>1.73</td>
<td>1.63</td>
<td>1.22</td>
</tr>
<tr>
<td>31</td>
<td>0.74</td>
<td>0.57</td>
<td>0.46</td>
<td>1.40</td>
<td>1.71</td>
<td>1.62</td>
<td>1.21</td>
</tr>
<tr>
<td>33</td>
<td>0.74</td>
<td>0.59</td>
<td>0.48</td>
<td>1.24</td>
<td>2.36</td>
<td>2.18</td>
<td>1.46</td>
</tr>
</tbody>
</table>

Table 5
The ratio of the intensity of each individual harmonic generation peak for laser fields with various pulse lengths (40, 60, and 80 optical cycles) versus the one with 100 optical cycles. All the laser pulses have the same laser intensity \( I = 5 \times 10^{13} \text{ W/cm}^2 \) and fundamental wavelength \( \lambda = 1064 \text{ nm} \). The last column \( P_{100}(n_{no}) \) shows the power spectrum (in a.u.) for the case with the pulse length equal to 100 optical cycles

<table>
<thead>
<tr>
<th>( n )</th>
<th>( R_{40/100} )</th>
<th>( R_{60/100} )</th>
<th>( R_{80/100} )</th>
<th>( P_{100}(n_{no}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>1.44</td>
<td>1.17</td>
<td>1.06</td>
<td>8.43 \times 10^{-7} \a</td>
</tr>
<tr>
<td>5</td>
<td>1.44</td>
<td>1.18</td>
<td>1.06</td>
<td>1.97 \times 10^{-8}</td>
</tr>
<tr>
<td>7</td>
<td>1.90</td>
<td>1.12</td>
<td>1.07</td>
<td>1.84 \times 10^{-9}</td>
</tr>
<tr>
<td>9</td>
<td>1.29</td>
<td>1.19</td>
<td>0.98</td>
<td>8.84 \times 10^{-9}</td>
</tr>
<tr>
<td>11</td>
<td>1.19</td>
<td>1.24</td>
<td>1.00</td>
<td>7.20 \times 10^{-9}</td>
</tr>
<tr>
<td>13</td>
<td>1.73</td>
<td>0.97</td>
<td>1.07</td>
<td>9.01 \times 10^{-10}</td>
</tr>
<tr>
<td>15</td>
<td>1.24</td>
<td>1.17</td>
<td>1.01</td>
<td>5.95 \times 10^{-10}</td>
</tr>
<tr>
<td>17</td>
<td>1.38</td>
<td>1.26</td>
<td>1.05</td>
<td>2.35 \times 10^{-10}</td>
</tr>
<tr>
<td>19</td>
<td>1.51</td>
<td>1.13</td>
<td>1.07</td>
<td>9.59 \times 10^{-11}</td>
</tr>
<tr>
<td>21</td>
<td>1.50</td>
<td>1.17</td>
<td>1.05</td>
<td>4.46 \times 10^{-11}</td>
</tr>
<tr>
<td>23</td>
<td>1.42</td>
<td>1.17</td>
<td>1.06</td>
<td>4.90 \times 10^{-11}</td>
</tr>
<tr>
<td>25</td>
<td>1.36</td>
<td>1.15</td>
<td>1.06</td>
<td>1.72 \times 10^{-11}</td>
</tr>
<tr>
<td>27</td>
<td>1.34</td>
<td>1.15</td>
<td>1.06</td>
<td>1.43 \times 10^{-11}</td>
</tr>
<tr>
<td>29</td>
<td>1.45</td>
<td>1.15</td>
<td>1.05</td>
<td>4.41 \times 10^{-12}</td>
</tr>
<tr>
<td>31</td>
<td>1.63</td>
<td>1.17</td>
<td>1.05</td>
<td>6.00 \times 10^{-13}</td>
</tr>
<tr>
<td>33</td>
<td>1.82</td>
<td>1.23</td>
<td>1.06</td>
<td>4.53 \times 10^{-14}</td>
</tr>
</tbody>
</table>

\a 8.43 \times 10^{-7} = 8.43 \times 10^{-7}.

Fig. 6 shows the corresponding HHG spectrum in the limit of monochromatic laser fields (infinite pulse length). As the pulse length becomes longer, each individual harmonic peak becomes sharper. However, the range of the plateau and the position of the cut-off \( (n = 35) \) is independent of the duration of the pulse. This is consistent with a recent experimental observation [35]. To see more details of the effect of varying the pulse length on the HHG spectrum, we list in Table 5 the ratio of the intensity of individual harmonic generation peak from a given pulse length (40, 60, or 80 optical cycles) to that of the longer pulse (100 optical cycles). The last column shows the power spectrum \( P_{100}(n_{no}) \) (in a.u.) for the case of 100-optical-cycle pulse length. It is seen that as the pulse length becomes shorter, all the harmonic peaks show various degrees of enhancement. This indicates that for a given laser pulse energy, the shorter laser pulses produce the most harmonic signal, with the harmonic efficiency scaling inversely with pulse duration. This prediction is in accord with the observations obtained in several recent experiments [30,35]. Finally, we note that the predicted cut-off position \( (n = 35) \) for the present cases (1064 nm, \( 5 \times 10^{13} \text{ W/cm}^2 \)) is...
larger than that predicted by the quasiclassical model \((n = 27)\). This is consistent with the fact that the Keldysh parameter for this case is \(\gamma = 1.13\), indicating that HHG is likely to be produced by the multiphoton excitation mechanism rather than by the tunneling mechanism.

3.6. The effect of laser intensity on the HHG spectrum

Finally, we examine the effect of varying the laser intensity on the HHG spectrum. Fig. 7 shows the HHG power spectra for both length and acceleration forms for the case of linearly polarized \(\sin^2\) pulsed fields with intensity \(10^{14}\) W/cm\(^2\) and wavelength 1064 nm. The pulse length is 40 optical cycles. At this higher intensity, significant ionization has occurred and the HHG plateau is significantly extended. In this calculation we used 300 grid points and \(r_{\text{max}} = 150-250\) a.u. Adjustment of the absorber position or parameters has little effect on the power spectrum before the cut-off position. Fig. 7 shows the general good agreement of the length and acceleration power spectra for the whole range of harmonics until the cut-off regime \((n \approx 45-51)\), beyond which the length-form spectrum levels off but the acceleration form spectrum continues to decline. The Keldysh parameter for this case is \(\gamma = 0.80\), indicating the tunneling regime is reached. The cut-off position at \(n \approx 45\) is consistent with that predicted by...
3. Conclusion

In this paper, we introduce a new split-operator method in energy representation along with a generalized pseudospectral technique for optimal spatial grid discretization for the numerical solution of time-dependent Schrödinger equation and non-perturbative treatment of multiphoton ionization and high-order harmonic generation in intense laser fields. The method is found to be computationally efficient and capable of providing high-quality time-dependent wavefunctions for accurate treatment of HHG processes. The method is applied to a detailed investigation of the feasibility of coherent control of HHG spectrum by tuning the pulse length, chirp, and intensity of the ultrashort intense pulsed laser fields. Several novel phenomena are predicted. These theoretical results are in good agreement with several recent experimental observations.

Extension of the method to the study of HHG and other multiphoton processes of many-electron system in intense laser fields by means of a generalized time-dependent density functional theory [36,37] is in progress.

Acknowledgements

This work was partially supported by the Division of Chemical Sciences, Office of Basic Energy Sciences of the US Department of Energy, and by National Science Foundation under contract No. PHY-95-12100.

References