Probing time-resolved emission in laser-driven electron-multirescattering in a high-order harmonic generation

Peng-Cheng Li, Chun-Xiao Li, Xiong-Yuan Lei, Yae-Lin Sheu, Xiao-Xin Zhou and Shih-I Chu

College of Physics and Electronic Engineering, Northwest Normal University, Lanzhou, People’s Republic of China; Department of Physics, National Taiwan University, Taipei, Taiwan; Department of Chemistry, University of Kansas, Lawrence, USA

ABSTRACT

We present an ab initio study of quantitative extraction of the time–frequency spectra of multiple rescattering processes of the electrons driven by the mid-infrared laser field in a high-order harmonic generation (HHG). The HHG is calculated by solving the three-dimensional time-dependent Schrödinger equation by means of the time-dependent generalized pseudospectral method. We extend a synchrosqueezed transform (SST) technique to extract the individual contributions of multiple rescattering processes in HHG. Combining with an extended semiclassical analysis and the SST time–frequency spectrum, the role of quantum trajectories in multiple rescattering processes in HHG is clarified. We find that the SST allows us to distinguish the individual contribution of multiple rescattering processes in HHG and show the details of the spectral and temporal fine structures of the HHG, which provides an important tool for a deep understanding to the dynamics of multiple rescattering processes in HHG.

ARTICLE HISTORY

Received 30 July 2018
Accepted 9 November 2018

KEYWORDS

High-order harmonic generation; intense laser field; time–frequency analysis; multiple rescattering processes; time-dependent Schrödinger equation

1. Introduction

High-order-harmonic generation (HHG) is produced by the interaction of atoms or molecules with strong laser fields, leading to the emission of extreme ultraviolet (XUV) photons. Its potential application (1–6) as an attosecond light source has attracted much interest in ultrafast science and technology. The attosecond pulse generation provides an important tool to probe and control the dynamical behaviours of atoms, molecules, and condensed matter (7–9). The essential features in HHG, such as a rapid drop at low-order harmonics, above-threshold harmonic plateau, and finally a sharp cutoff beyond which no further harmonic emission, have been observed experimentally. The HHG cutoff is estimated approximately at the energy $I_p+3.17U_p$, where $I_p$ is the atomic ionization potential, and $U_p$ is the ponderomotive energy. The HHG process can be well understood by a semiclassical three-step model (10,11). Firstly, the bound electron tunnels through the barrier formed by the Coulomb potential and the laser field, then it is subsequently accelerated and obtain the kinetic energy from the laser field. Finally, the electrons can be driven back toward the core and recombine with the ground state to convert the binding energy and kinetic energy into an emitted harmonic photon. For most of the study of the HHG, the major attention has been focused on the first rescattering in HHG. However, the multiple rescattering processes also play an important role in HHG and the dynamical origin of the HHG associated with multiple rescattering is less understood and largely unexplored.

In the last decade, considerable effort has been devoted to the HHG related multiple rescattering processes (12–17). More recently, Hickstein et al. (18) have studied the laser-driven electron multiple rescattering in strong-field ionization. The results show that the photoelectron angular distributions generated by mid-IR lasers are created by electron trajectories that scatter from the parent ion after passing by the ion several times. Haessler et al. (19) have studied that the energy transferred from the field to the electron may be released as attosecond-duration extreme ultraviolet emission in HHG on the rescattering with the parent ion. He et al. (20) have studied the quantum trajectories in HHG from multiple rescattering events in the long-wavelength regime. The result shows that the HHG yield ratio associated with multiple rescattering raises gradually with the increasing laser wavelength. Hernández-García et al. (21) have explored the conditions for resolving high-order electronic recollisions in HHG spectroscopy. Zhang et al. (22) have studied the multiple rescattering processes in HHG from the molecules. Francois Risoud et al. (23) have presented a method to quantitatively extract the emission...
times of the HHG via the determination of instantaneous frequencies. Recently, we have explored the laser-driven electron-multirescattering dynamics in HHG (24), we found that the multiphoton- and tunneling-ionization regimes are competitive in the multiple rescattering processes.

In this paper, we extend a synchronous squeezed transform (SST) technique (25,26) to extract the individual contributions of multiple rescattering in HHG. The harmonic spectrum of the hydrogen atom can be calculated accurately and efficiently by solving the time-dependent Schrödinger equation (TDSE) by means of the time-dependent generalized pseudospectral method (TDGPS) (27). By comparing the results with the extended semi-classical analysis, the Hilbert time–frequency spectrum for the emission time of the HHG, and the individual multiple rescattering contributions from the quantitative extraction of the HHG time–frequency spectrum based on the SST method, we find that the extended SST method provides an important tool for quantitatively probing the dynamics of the multiple rescattering processes in HHG.

2. Numerical methods

The HHG is produced by the interaction of an intense laser field with atomic or molecular systems and can be calculated by solving the time-dependent Schrödinger equation (TDSE) (in atomic units)

\[ i \frac{\partial \psi(r, t)}{\partial t} = \hat{H} \psi(r, t) = [\hat{H}_0 + \hat{V}(r, t)] \psi(r, t), \quad (1) \]

where \( \hat{V}(r, t) \) is the atom-field interaction in the electric dipole approximation. For a linearly polarized laser field, which is given as

\[ \hat{V}(r, t) = -E(t) \cdot \mathbf{r} = -zeE(t), \quad (2) \]

And \( \hat{H}_0 \) is the unperturbed Hamiltonian for the hydrogen atom, which is written as

\[ \hat{H}_0 = -\frac{1}{2} \nabla^2 - \frac{1}{r}. \quad (3) \]

The TDSE is solved by means of the time-dependent generalized pseudospectral method (TDGPS) (27) in the spherical coordinates. The TDGPS numerical technique has been successfully applied to the study of a broad range of multiphoton ionization and HHG of atomic and molecular systems (28,29). This numerical scheme comprises two significant components: (i) The GPS technique allows the radial coordinates to be optimally discretized in a nonuniform radial grid. With only a modest number of grid points, the discretization is characterized by denser grids near the nuclear origin and sparser grids for larger distances. (ii) The second-order split-operator technique in the energy representation permits elimination of undesirable fast-oscillating high-energy components and thus leads to efficient and accurate time propagation of the wave function according to the relation expressed as

\[ \psi(r, t + \Delta t) \simeq \exp \left( -i \hat{H}_0 \frac{\Delta t}{2} \right) \times \exp \left[ -i \hat{V}(r, \theta, t + \frac{\Delta t}{2}) \Delta t \right] \times \exp \left( -i \hat{H}_0 \frac{\Delta t}{2} \right) \psi(r, t) + O(\Delta t^3). \quad (4) \]

The unitarity of the wave function is automatically preserved by Equation (4). Once the time-dependent wave function \( \psi(r, t) \) is obtained, the induced dipole moment in the length and acceleration forms can be calculated as follows:

\[ d_L(t) = \langle \psi(\mathbf{r}, t) | \mathbf{z} | \psi(\mathbf{r}, t) \rangle, \quad (5) \]

\[ d_A(t) = \langle \psi(\mathbf{r}, t) | -\frac{z}{\tau^2} + E(t) | \psi(\mathbf{r}, t) \rangle. \quad (6) \]

The HHG power spectra can be obtained by the Fourier transformation of time-dependent dipole moment, it is expressed as

\[ P_L(\omega) = \left| \frac{1}{t_f - t_i} \int_{t_i}^{t_f} d_L(t)e^{-i\omega t}dt \right|^2, \quad (7) \]

\[ P_A(\omega) = \left| \frac{1}{(t_f - t_i)\alpha^2} \int_{t_i}^{t_f} d_A(t)e^{-i\omega t}dt \right|^2. \quad (8) \]

To extract the individual multiple-rescattering contributions in HHG, we extended a SST technique (30) based on Gabor transform (31) to explore the frequency characteristics of the laser-driven atom associated with the multiple rescattering events. The SST is defined by

\[ S(t, \xi) = \int V(t, \omega) \frac{1}{\alpha} h(t - \omega) \left( \frac{\xi - \omega f(t, \omega)}{\alpha} \right)d\omega, \quad (9) \]

where \( \alpha > 0, h(t) = e^{-t^2}/\sqrt{\pi}, \) and \( V(t, \omega) \) is the Gabor transform of the harmonic spectra, which can be written as

\[ V(t, \omega) = \int d(t')g(t' - t)e^{-i\omega(t' - t)}dt', \quad (10) \]

where \( d(t') \) is the time-dependent dipole moment and the sweeping function \( g(t' - t) \) is given by

\[ g(x) = \frac{1}{\sqrt{2\pi}\tau} e^{-x^2/\tau}. \quad (11) \]
Here, we choose the parameter $\tau = (3\omega_0)^{-1}$ to perform the Gabor transform. The instantaneous frequency information function $\omega_f(t, \omega)$ is defined by

$$\omega_f(t, \omega) = \begin{cases} -i\partial_t V(t, \omega) \frac{V(t, \omega)}{V(t, \omega)}, & \text{for } V(t, \omega) \neq 0 \\ \infty, & \text{for } V(t, \omega) = 0 \end{cases}$$ (12)

The causality property of the dipole moment is preserved, so it allows to an inverse transform.

### 3. Results and discussions

To understand the physical picture of multiple rescattering processes in HHG, in Figure 1(a), we show several classical trajectories in HHG calculated by using the three-step model in the intense 800-nm, 1200-nm, and 1800-nm laser pulses with the peak intensity $I = 1.0 \times 10^{14}$ W/cm$^2$, respectively. In our calculation, the laser field has the following form

$$E(t) = E_0 f(t) \cos(\omega t),$$ (13)

where $f(t)$ is the cosine-squared pulse with 10 optical cycles. $E_0$ and $\omega$ are the amplitude and frequency, respectively. The classical trajectories show the multiple rescattering behaviours become more obviously in HHG with the increasing laser wavelength, it is in good agreement with the previous works (20). On the other hand, we find that the first rescattering marked by 1st occurs in one optical cycle, but the multiple rescattering marked by 2nd, 3rd, 4th and 5th occurs less than the half optical cycle. Figure 1(b) shows the HHG power spectrum of the hydrogen atom calculated by solving the three-dimensional TDSE, the laser parameters used are the same as those in Figure 1(a). It is clearly seen that the energies of the cutoffs are located at 30, 55, and 100 eV for the laser wavelength 800-nm, 1200-nm, and 1800-nm laser pulses, respectively. The results show that the HHG plateau is extended when the laser wavelength is increased.

As the discussion above, the multiple-rescattering contribution in HHG is raising with the increasing laser wavelength. Therefore, we adopt an 1800-nm laser pulse as the driven field to explore the individual multiple rescattering events for time-resolved high-order harmonic emission. In Figure 2(a), a comparison of the HHG time–frequency spectra based on the Gabor transform and classical simulation calculated by the three-step model is shown. The other laser parameters used are the same as those in Figure 1. The semiclassical return energies are obtained by solving the Newton’s law of motion along with the Coulomb effect $-\nabla(-1/r)$. The green lines (the 1st rescattering) and black lines (the 2nd rescattering) indicate the return energies as a function of the emission time, respectively. Although the multiple rescattering trajectories beyond twice returning are observed in the classical simulation as shown in Figure 1(a), the trajectories in the Gabor time–frequency spectra based on the TDSE are not obviously as shown in Figure 2(a), the reason is that the contribution of the multiple rescattering trajectories beyond twice returning is weak in HHG. It is well known that there are two types of dominant quantum trajectories (short trajectory and long trajectory) in HHG to contribute to the harmonic generation. The results show that the harmonic generation is due to the short trajectory belongs to the 1st rescattering, and the 2nd rescattering mainly contributes to the plateau region less than the 100th order (about $2U_p$). The short trajectories have the main contributions to the harmonic generation in the intense laser field due to the higher returning probability of the electrons. In fact, the previous study suggests that the short trajectory is the dominant contribution to the HHG and cutoff extension. It has been shown theoretically (32) and experimentally (33) that three-dimensional propagation effect favour contributions from the short trajectories.

![Figure 1.](image-url)
In Figure 2(b), we show a comparison of the HHG time–frequency spectra and the quantitative extraction of emission time associated with the 2nd rescattering process from Hilbert transform (only shows the contribution of the 2nd rescattering). Hilbert transform is given by

$$H_d(t) = \frac{1}{\pi} P \int \frac{d(t'')}{t - t'} dt'.$$

Here, $d(t'')$ is a filtered dipole moment and $P$ is the Cauchy principal value. In the Hilbert transform method, $d(t'')$ is obtained by filtering the dipole moment $d(t')$ below the ionization energy $I_p$ where the contribution of multiple rescattering is weak. Combining with the results of the Hilbert transform and the Gabor time–frequency spectra of the HHG, we obtain a similar result as shown in Figure 2(a), the 2nd rescattering mainly contribute to the plateau region less than the 100th order (about $2U_p$). The emission time associated with the 2nd rescattering process can be extracted by using the Hilbert transform.

To extract the components of the HHG associated with the individual multiple-rescattering trajectories from the TDSE calculation, we extend an SST time–frequency technique. The SST has been applied mainly to the analysis of medical signal (34) in recent years. The first application to the quantum phenomenon was presented in Ref. (30). One novel feature of SST is that it can preserve the causality property of the signal, leading to an inverse transform. Therefore, we choose the SST time–frequency analysis as a tool to explore the details of the spectral and temporal fine structures of the HHG. As the discussion above, the main contribution to the above threshold harmonics is due to the short trajectories. In Figure 3(a), we present the SST time–frequency spectra of the HHG, and the same laser parameters used are the same as those in Figure 2. The green lines and the black lines indicate the semiclassical trajectories of the 1st rescattering and the 2nd rescattering, respectively. As shown in Figure 3(a), the SST time–frequency transform allows us to explore the subtle details of the multiple-rescattering trajectories in HHG. It is clear that not only the semiclassical trajectories of the first returning are matched very well but also the multiple-rescattering trajectories are in good agreement with the SST time–frequency spectra. Combining with the results of the semiclassical simulations, the SST may identify the maximum energy of the second returning, it is located at

![Figure 2](image1.png)

Figure 2. (a) Comparison of the HHG time–frequency spectra and classical trajectories simulation in an intense 1800-nm laser field. The classical energy map is calculated by the three-step model, the green lines and the black lines indicate the 1st rescattering energies and the 2nd rescattering energies as a function of the emission time, respectively. The colour bar (colour online) is in a logarithmic scale. (b) Same as (a), a comparison of the HHG time–frequency spectra and the quantitative extraction of emission time related to the 2nd rescattering process from Hilbert transform (black lines). The other laser parameters used are the same as those in Figure 1.

![Figure 3](image2.png)

Figure 3. (a) SST time–frequency spectra of the HHG of hydrogen atom driven by an 1800-nm laser pulse and the corresponding semiclassical simulation. (b) The quantitative extraction of the SST time–frequency spectra of the 2nd returning. The colour bar (colour online) is in a logarithmic scale. The green lines (the 1st rescattering) and black lines (the 2nd rescattering) indicate the return energies as a function of the emission time, respectively. The laser parameters used are the same as those in Figure 2.
Figure 4. (a) The quantitative extraction of time-dependent dipole moment based on the SST time-frequency spectra. (b) The individual HHG power spectrum (total, 1st rescattering, and 2nd rescattering) from the quantitative extraction of time-dependent dipole moment. The same laser parameters used are the same as those in Figure 2.

around the 100th harmonics. On the other hand, the SST allows us to extract the individual contributions of the multiple-rescattering trajectories. The dipole moment of multiple rescattering has its own frequency characteristics, we can separate these frequency-components as the first rescattering and second rescattering events by reconstructing the SST time–frequency spectra \(^{(35)}\). Once we obtain the time–frequency spectra of the first rescattering and second rescattering, it is easy to calculate the individual time-dependent dipole moment of the multiple rescattering by the inverse transform of the SST. In Figure 3(b), we present the quantitative extraction of the SST time–frequency spectra of the 2nd rescattering contributions. The black lines are the corresponding classical simulation, and it is in good agreement with the individual 2nd rescattering contributions from the TDSE calculations.

To obtain the individual harmonic spectra of the multiple-rescattering processes, we perform the Fourier transformation of the time-dependent dipole moment of the 1st rescattering and the 2nd rescattering to obtain the individual HHG power spectra, respectively. In Figure 4(a), we show the quantitative extraction of the time-dependent dipole moment of the 1st rescattering and 2nd rescattering by using the SST method. In Figure 4(b), the corresponding individual HHG power spectra are presented. It is clear that the contributions in HHG almost come from the 1st rescattering event, the 2nd rescattering mainly contributes to the harmonic generation from the 40th harmonic to the 80th harmonic. Note that, in our case, we only extract the individual contribution of the 1st rescattering and 2nd rescattering, because the contribution of the multiple rescattering beyond the 2nd returning is weak. It is clear that the HHG intensity of the 2nd rescattering is lower as three orders than the 1st rescattering case. In fact, the main contribution of HHG comes from the first rescattering, so the HHG plateau of the 1st rescattering is similar to the total HHG. But the strength of the total HHG has a little raising by comparing to the first rescattering one, because the total HHG includes all contributions from the first rescattering and other multiple rescattering, although the contributions of other multiple rescattering are weak.

4. Conclusions

In conclusion, we have presented an \textit{ab initio} study on the HHG of hydrogen atom in intense 1800-nm infrared laser fields by solving the TDSE by means of the TDGPS method accurately in space and time. To understand multiple rescattering effects of the HHG from an \textit{ab initio} simulation, we extended a new SST transform to extract the individual contribution of multiple rescattering from the time-dependent dipole moment of the hydrogen atom in an intense laser field. By comparing the decomposition of the SST time–frequency spectra, Hilbert time–frequency spectra as well as the extended semiclassical calculations, the role of the quantum trajectories in the multiple-rescattering process in HHG is clarified. We find that the extended SST transform is a powerful tool to probe the novel features of the spectral and temporal structures of the HHG. It allows us to obtain the quantitative extraction of the individual harmonic spectra of the multiple-rescattering processes in HHG.

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This work was partially supported by the Chemical Sciences, Geosciences and Biosciences Division of the Office of Basic Energy Sciences, Office of Sciences, and U.S. Department of Energy under grant number DE-FG02-04ER15504. P. C. Li. and X. X. Zhou are partially supported by the National Natural Science Foundation of China (Grants No.11364039, No. 11465016, No. 11674268, and No. 11764038). We also would like to acknowledge the partial support of the Ministry of
Science and Technology and the National Taiwan University (Grants No. 107L892901 and 107L104048).

References


