Resolving the quantum dynamics of near cut-off high-order harmonic generation in atoms by Bohmian trajectories

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Abstract: We present an ab initio study of the quantum dynamics of high-order harmonic generation (HHG) near the cutoff in intense laser fields. To uncover the subtle dynamical origin of the HHG near the cutoff, we extend the Bohmian mechanics (BM) approach for the treatment of attosecond electronic dynamics of H and Ar atoms in strong laser fields. The time-dependent Schrödinger equation and the self-interaction-free time-dependent density functional theory are numerically solved accurately and efficiently by means of the time-dependent generalized pseudospectral method for nonuniform spatial discretization of the Hamiltonian. We find that the most devoting trajectories calculated by the BM to the plateau harmonics are shorter traveling trajectories, but the contributions of the short trajectories near the cutoff are suppressed in HHG. As a result, the yields of those harmonics in the region near the cutoff are relatively weak. However, for the last few harmonics just above the cutoff, the HHG intensity becomes a little higher. This is because the HHG just above the cutoff arises from those electrons ionized near the peak of the laser pulse, where the ionization rate is the highest. In addition, the longer Bohmian trajectories return to the core with lower energies, these trajectories contribute to the below-threshold harmonics. Our results provide a deeper understanding of the generation of supercontinuum harmonic spectra and attosecond pulses via near cutoff HHG.

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1. Introduction

High-order harmonic generation (HHG) has enabled the generation of coherent extreme ultraviolet (XUV) source in the attosecond time scale [1–3], leading to the real-time experimental observation of ultrafast electron dynamics in atomic and molecular systems [4–6]. HHG has attracted immense interest in the past decades, and it is now a forefront subject of much current significance in science and technology [7–9]. The harmonic spectrum exhibits a broad plateau structure where all harmonics have the similar amplitude and the plateau is followed by a sharp cutoff approximately at the energy \( I_p + 3.17U_p \), where \( I_p \) is the atomic ionization potential, and \( U_p \) is the ponderomotive potential.

Currently the investigations of HHG have focused on how to enhance the HHG plateau and extend the harmonic cutoff for the generation of stronger and shorter XUV source. Therefore, the analysis of the characteristics of the plateau and the underlying mechanism is vital in this respect. Several experimental and numerical studies have been done to discuss the dependence of the harmonic-generation cutoff on the laser intensity [10] and the laser wavelength scaling of
the HHG yield near cutoff region [11]. On the other hand, one of the most important features in HHG is the generation of supercontinuum harmonic spectra near cutoff in the intense laser field [12]. By superposing several supercontinuum harmonics near cutoff, an isolated ultrashort XUV attosecond pulses can be generated, leading to a number of exciting new advancements in ultrafast science, such as real-time observation and coherent control of atomic-scale electron and nuclear dynamics [13]. Therefore, it is not only necessary to generate intense harmonic spectra near cutoff, it is also very important to explore the dynamical origin. The interpretations from the three-step model [14,15] and strong-field approximation (SFA) [16] often used are valuable in providing qualitative insight regarding electron dynamics in laser-atom interaction. However, the quantum mechanical treatment appears to be inevitable to explore the underlying mechanism of the essential features of the harmonic generation. Fully *ab initio* quantum mechanical solution of time-dependent Schrödinger equation (TDSE) is currently feasible for one- and two-electron systems in intense laser field. Recently, by solving accurately the TDSE, we have demonstrated that the HHG near the cutoff are suppressed when the ionization process is pushed from the multiphoton regime into the tunneling regime [17], but the dynamical origin of the suppression near cutoff HHG still is less understood. More recently, it has been shown that the Bohmian mechanics (BM) [18] can provide an accurate trajectory-based scheme to causally interpret the electron wave packet dynamics and BM has been successfully applied as an alternative and complementary quantum approach to study the strong field processes such as HHG [19], laser-driven electron dynamics [20–24], etc.

In this paper, we present a fully *ab initio* three-dimensional and accurate treatment of the Bohmian trajectories beyond SFA, and we utilize it to explain the electron dynamical mechanism of the HHG near cutoff from H and Ar atoms. Numerical scheme to obtain Bohmian trajectories can be found in detail in our previous works [25,26]. The harmonic spectrum of H atom can be calculated accurately and efficiently by solving the TDSE [27] by means of the time-dependent generalized pseudospectral method (TDGPS) [28]. And many-electron quantum system of Ar atom can be treated accurately by means of the *self-interaction-free* time-dependent density-functional theoretical (TDDFT) approach [29–32] with proper long-range potential [33]. We find that the dynamical origin of harmonic emission near cutoff is associated with the competition between the short- and long-trajectories, leading to the harmonic suppression near the HHG cutoff. Our detailed electron-trajectory analysis calculated by the BM indicates that the short trajectories play an important role rather than those long trajectories in the harmonic suppression near cutoff.

### 2. Theoretical methods

The harmonic spectrum of H and many-electron Ar atoms can be calculated accurately and efficiently by solving the TDSE and the *self-interaction-free* TDDFT approach, respectively. The TDSE can be written in the dipole approximation in atomic units (a.u.) as

\[
\frac{i}{\hbar} \frac{\partial \psi(r, t)}{\partial t} = [H_0 + E(t) \cdot r] \psi(r, t),
\]

where

\[
H_0 = \frac{1}{2} \nabla^2 - \frac{Z}{r},
\]

and \(E(t)\) is the electric field. For the H atom, \(Z=1\). For many-electron systems, we extend the *self-interaction-free* TDDFT approach for nonperturbative treatment of multiphoton processes. In the TDDFT frame, the time-dependent Kohn-Sham equations of \(N\)-electron systems in intense
laser fields can be written as,

\[
\frac{i}{\hbar} \frac{\partial}{\partial t} \psi_{i\sigma}(\mathbf{r}, t) = H(\mathbf{r}, t)\psi_{i\sigma}(\mathbf{r}, t)
= \left[-\frac{1}{2} \nabla^2 + V_{\text{OEP}}^{\sigma}(\mathbf{r}, t)\right]\psi_{i\sigma}(\mathbf{r}, t),
\]

where $V_{\text{OEP}}^{\sigma}(\mathbf{r}, t)$ is the time-dependent optimized effective potential depending upon the total electron density $\rho(\mathbf{r}, t)$, $N_\sigma (=N_\uparrow$ or $N_\downarrow)$ is the total number of electrons for a given spin $\sigma$, and the total number of electrons in the system is $N = \sum_\sigma N_\sigma$. The total electron density $\rho(\mathbf{r}, t)$ is determined by the single-electron orbital wave functions $\psi_{i\sigma}(\mathbf{r}, t)$ as

\[
\rho(\mathbf{r}, t) = \sum_\sigma \sum_{i=1}^{N_\sigma} \rho_{i\sigma}(\mathbf{r}, t) = \rho_\uparrow(\mathbf{r}, t) + \rho_\downarrow(\mathbf{r}, t).
\]

The TDSE and TDDFT are solved by means of the TDGPS method [28]. It has been shown that the TDGPS is considerably more accurate and computationally more efficient than the conventional time-dependent propagation techniques using equal-spacing grid discretization. The TDGPS method has been applied successfully to the study of a broad range of multiphoton ionization and HHG of atomic and molecular systems in strong fields. The numerical scheme of the TDGPS method consists of two essential steps: (i) The spatial coordinates are optimally discretized in a nonuniform spatial grid by the generalized pseudospectral technique [28]. This discretization uses only a modest number of grid points, and it is characterized by denser grids near the nuclear origin and sparser grids for larger distances. (ii) A second-order split-operator technique in the energy representation is used for the time propagation of the wave function, which can be expressed as

\[
\psi(\mathbf{r}, t + \Delta t) \approx \exp(-iH_0 \frac{\Delta t}{2}) \times \exp[-iV(\mathbf{r}, \theta, t + \frac{\Delta t}{2})\Delta t] \times \exp(-iH_0 \frac{\Delta t}{2})\psi(\mathbf{r}, t) + O(\Delta t^3).
\]

When the time-dependent wave function $\psi(\mathbf{r}, t)$ is available, we can calculate the expectation value of the induced dipole moment in the length and acceleration forms, which can be written as

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

\[
a(t) = \langle \psi(\mathbf{r}, t)|\frac{d^2}{dt^2}|\psi(\mathbf{r}, t)\rangle,
\]

respectively. The spectral density of radiation energy can be calculated, within the semiclassical approach, by the following expression [34,35]:

\[
S(\omega) = \frac{2\omega^4}{3\pi c^3} |\tilde{d}(\omega)|^2 = \frac{2}{3\pi c^3} |\tilde{a}(\omega)|^2,
\]

where $c$ is the speed of light, $\tilde{d}(\omega)$ and $\tilde{a}(\omega)$ are the Fourier transforms of the time-dependent dipole moment in the length and acceleration forms, given by

\[
\tilde{d}(\omega) = \int_{-\infty}^{\infty} d(t)e^{i\omega t} dt,
\]

\[
\tilde{a}(\omega) = \int_{-\infty}^{\infty} a(t)e^{i\omega t} dt.
\]
\[ \tilde{a}(\omega) = \int_{-\infty}^{\infty} a(t)e^{i\omega t} dt, \]  

(10)

respectively. In our calculation, the time-dependent dipole moment in acceleration form is used for efficient and accurate treatment of HHG, and it allows us to achieve full the convergence of the time-dependent wave function by using smaller number of grid points.

To explore the quantum dynamics resolved by the electron trajectories in HHG, we perform the BM calculations based on the time-dependent wave function \( \psi(r, t) \). For the atoms in linearly polarized laser fields, the angular momentum projection onto the polarization \( z \)-axis direction of the field is conserved. That means the dependence of the wave function on the rotation angle \( \phi \) about the \( z \)-axis is reduced to the factor \( e^{im\phi} \), where \( m \) is the angular momentum projection quantum number. For \( m=0 \), the wave function does not depend on \( \phi \) at all, thus the gradient of the wave function \( \psi \) can be calculated with respect to the radial coordinates \( r \) and angle \( \theta \) between the radius-vector and \( z \)-axis:

\[ \nabla \psi = e_r \frac{\partial \psi}{\partial r} + e_\theta \frac{1}{r} \frac{\partial \psi}{\partial \theta}, \]  

(11)

\[ = e_r \frac{\partial \psi}{\partial r} - e_\theta \frac{1}{r} \frac{\partial \psi}{\partial \cos \theta}, \]  

(12)

\( e_r \) and \( e_\theta \) are the unit vectors of spherical coordinate system. The equation for the Bohmian trajectories [25] reads as

\[ \frac{d r}{d t} = \text{Im} \frac{\nabla \psi}{\psi}, \]  

(13)

and the Bohmian trajectory lies in the plane defined by the initial radius-vector and the \( z \)-axis. The vector equation (13) is equivalent to a set of three 1D equations:

\[ \frac{d r}{d t} = \text{Im} \left( \frac{1}{\psi} \frac{\partial \psi}{\partial r} \right), \]  

(14)

\[ \frac{d \theta}{d t} = -\frac{\sin \theta}{r} \text{Im} \left( \frac{1}{\psi} \frac{\partial \psi}{\partial \cos \theta} \right), \]  

(15)

\[ \frac{d \phi}{d t} = 0. \]  

(16)

The equations (14) and (15) are solved numerically with the help of the fourth-order Runge-Kutta (RK4) method, yielding the electron quantum trajectories. Since the quadrature points for RK4 differ from the original GPS grid points, we need to perform an additional interpolation using the GPS interpolation formula.

3. Results and discussions

Figure 1 shows the HHG spectral densities of radiation energy of H atom driven by an intense 800-nm infrared laser pulse with the peak intensity (a) \( I=0.8I_0 \) \( (I_0=10^{14}\text{W/cm}^2) \), (b) \( I=1.5I_0 \), and (c) \( I=2.5I_0 \), respectively. In this calculation, the laser field has the following form:

\[ E(t) = E_0 f(t) \cos(\omega t), \]  

(17)

where \( f(t) \) is the cosine-squared pulse with 20 optical cycles (o.c.). \( E_0 \) and \( \omega \) are the amplitude and frequency, respectively. To achieve full convergence of the time-dependent wave functions and HHG spectra, the parameters of the radial range \( r_{\text{max}} \) used are from 130 a.u. to 150 a.u., 800 radial grid points, 80 partial waves, and an absorber is placed at 80 a.u. to filter out the ionizing wave packet. In Fig. 1(a), the laser peak intensity is 0.8\( I_0 \), and the corresponding Keldysh parameter \( \gamma \) \( (\gamma=\sqrt{I_p/2U_p}) \) is equal to 1.2. Note that the multiphoton ionization is typically
classified by $\gamma > > 1$, while tunneling ionization is classified by $\gamma << 1$. The cutoff of the HHG is located at the 19th harmonic order indicated by the red dashed lines. An interesting feature of the HHG is that the yield of the harmonics near the cutoff between the 13th order and the 19th order is suppressed as shown in inset, and the lowest yield in HHG is located at the 17th harmonic order. In Fig. 1(b) and 1(c), the spectral suppression near the cutoff still exists when the laser intensity is increased to $1.5I_0$ ($\gamma \approx 0.9$) and $2.5I_0$ ($\gamma \approx 0.7$) corresponding to the tunneling regime, and the lowest yields in HHG near cutoff are located at the 25th harmonic order and the 37th harmonic order, respectively.

**Fig. 1.** HHG spectral densities of radiation energy of H atom driven by an intense 800-nm infrared laser pulse with the peak intensity (a) $I=0.8I_0$ ($I_0=10^{14}$ W/cm$^2$), (b) $I=1.5I_0$, and (c) $I=2.5I_0$, respectively. The inset shows an enlarged view of the spectral densities of the radiation energy near cutoff HHG. The red vertical dashed lines indicate the HHG cutoff.

A better understanding of the electron dynamics during the ionization process can be achieved by analysis of the time evolution of the electron density. Figure 2(a) is a snapshot of H atom electron density at $-0.1$ optical cycles in an intense laser field. The laser parameters used are the same as those in Fig. 1(a). As illustrated in this figure, several distinct density portions are shaped and detached from the H atom within a half optical cycle of the laser field. The detailed mechanism of generation of these distinct wave packets is presented in our previous work [25]. Besides some of the wave packets that ionize directly and never return to core (shaded in gray), when the laser field changes sign, some of the density portions change direction and return to parent ion (shaded in green and red, and labeled by 1 and 2, respectively). Next, we will discuss the contribution of these returning wave packets in harmonic generation separately. Figure 2(b) shows BM trajectories which represent the time evolution of electron density of the system within the central optical cycle of the laser field. Bohmian trajectories provide a detailed perspective on the evolution of quantum electron density. In general, there is a close resemblance of the trajectory patterns and the electron density. To better illustrate this fact, we used the same colors and labels.
to indicate the corresponding groups of trajectories with respect to the density portions shown in Fig. 2(a). For each case, a few trajectories (bold solid lines) are selected as representatives for each group of trajectories. These bold trajectories are some representative trajectories in each group for further analysis on harmonic generation. Figure 2(c) shows the evolution of the electron energy in some of the individual trajectories. The energy is calculated as $E_k + E_p$, where $E_k$ and $E_p$ are the kinetic and Coulomb potential energies, respectively. Several prominent features are observed. (i) Although the first and second traveling wave packets (shaded in green and red) are not completely separated under this laser field, the short trajectories belonging to the first group (green, labeled 1) mainly contribute to lower order harmonics. This contribution, however, is subordinate, compared to the dominant role of multiphoton bound-bound resonance for this range of harmonics [31]. This effect is well pronounced in the structureless time profiles of these harmonics, presented in Fig. 2(d). (ii) Bound states resonances continue to effect the harmonic structures in the plateau region as well. The diminution of this aspect can be clearly seen in the time profiles of the harmonics 11 through 17. The contribution of quantum trajectories in these harmonics, on the other hand, increments throughout the plateau. The most devoting trajectories to this energy interval are shorter traveling trajectories which represent the dynamics of the outset of the second electron density portion (red). The most salient feature is the dramatic phase change, right at the 17th harmonics, which is shown in Fig. 2(d). This is a clear evidence that the energy contributions to the harmonics after this point would be from the quantum trajectories and the multiphoton bound-bound resonances play no role in generating these harmonics anymore. (iii) A few phase locked harmonics around cutoff (namely harmonics 19, 21, and 23) are generated by the small group of trajectories with a much synchronized emission time (between 0.1 to 0.15 o.c.). This can be seen in the energy profiles of the contributing trajectories in Fig. 2(c) and the corresponding time profiles in Fig. 2(d). (iv) As illustrated in Fig. 2(b) and 2(c), the longer trajectories in the group 2 (red) return to the core with very low energies. These trajectories can contribute below-threshold harmonics.

To explore the role of quantum trajectories in the spectral suppression of the HHG near the cutoff, Fig. 3(a) presents two group of Bohmian trajectories arise from only those short- and long-trajectories of the electrons released in the positive z-direction at $-0.6$ optical cycles of the laser field. In Fig. 3(b), the returning energies of two groups as a function of emission time are presented by using the BM calculations based on the time-dependent wave function. It is seen clearly that the distribution of the returning energies near HHG cutoff related to the short trajectory group is small, but the long Bohmian trajectories with lower energies is dominant, these trajectories contribute to the below-threshold harmonics. This result is in good agreement with the discussions above as shown in Fig. 2(b) and 2(c). For comparison, in Fig. 3(c), we present the wavelet time-frequency spectra of the HHG [12] and the semiclassical simulation. In our calculations, the classical results of H atom are obtained by solving the Newton’s equation, given by

$$\ddot{r} = -\nabla V(r) - E(t)z,$$

where $E(t)$ is the laser field and $V(r)$ is the Coulomb potential. We define two kinds of the initial conditions, one corresponds to initially the electrons with an initial velocity are released at the core, here the minimum of initial velocity in the laser polarized direction $z$ is zero and the maximum is the height of the barrier at the release time. Another one corresponds to initially the electrons with an instantaneous position are released at the core. The results indicate that the contribution of short trajectories near cutoff is small in HHG. That is to say, the short trajectories play a more important role for the spectral suppression near the HHG cutoff, which is associated with a competition between the quantum trajectories as the discussion above in Fig. 2(c) and 2(d). Therefore, the yield of harmonics near cutoff is suppressed due to the weak contributions of the short trajectories, but the contribution of the short- and long-trajectories merged is dominant in the last few harmonics just above the cutoff as shown in Fig. 3(c).
Fig. 2. (a) Multiple portions of the electron density detached from the H atom as seen at \(-0.1\) optical cycles in an intense laser field. Different colors and labels indicate different returning portions of the electron density. The laser parameters used are the same as those in Fig. 1(a). (b) Thousand of the Bohmian trajectories initiated at \(-0.6\) optical cycles, representing the time evolution of H atom in the same laser field. Shaded in green and red colors (labeled by 1 and 2, respectively) are distinct group of trajectories which represent the evolution of the corresponding portions of electron density in (a). (c) The evolution of the electron energy with time for some of the selected trajectories. (d) The dipole time profiles of the consecutive harmonics within half optical cycle.
Fig. 3. (a) Bohmian trajectories of H atom arise from only those short- and long-trajectories at −0.6 optical cycles in the positive z-direction. The short- and long-trajectories are shaded in green (green group) and red (red group), respectively. (b) Returning energy $E_R$ of as a function of emission time of the green group and red group. (c) Wavelet time-frequency spectra of the HHG. For comparison, the green curves and the red curves indicate the semiclassical trajectories. The laser parameters used are the same as those in Fig. 1(a).
To check the sensitivity of the spectral suppression in HHG near cutoff associated with the laser intensity, we calculate the spectral densities of radiation energy of H and Ar atoms as a function of the laser peak intensity as shown in Fig. 4(a) and 4(b). It is seen clearly that there is a spectral suppression near cutoff in HHG. This phenomenon is especially significant when the laser peak intensity is increased beyond $1.4I_0 \sim 1.8I_0$ ($\gamma \approx 1$). Note that the HHG of Ar atom exhibits a deep suppression between the 31st and the 35th harmonic orders generated by an 800-nm laser pulse when the laser intensity is larger than $2 \times 10^{14}$W/cm$^2$. This is the well-known Cooper minimum phenomenon [36] produced due to a zero dipole moment between the $p$ ground-state wave function and the $d$ wave function of the photon ionized electron. On the other hand, in our previous work [14], we have confirmed that the competition between the multiphoton and tunneling ionization regimes leads to a weak contribution associated with the quantum trajectories in HHG near cutoff. The spectral suppression in HHG near cutoff is not sensitive to the laser wavelength.

Fig. 4. Spectral densities (Log scale) of radiation energy as a function of the laser peak intensity. (a) H atom. (b) Ar atom. The other laser parameters used are the same as those in Fig. 1.
4. Conclusions

In conclusion, we extend the BM approach to explore the laser-driven electron dynamics, which allows us to obtain an accurate and clear physical picture in HHG. We present the spectral suppression feature in HHG near cutoff predicted from H and Ar atoms in the intense laser field for the first time by accurately solving the TDSE or TDDFT, respectively. To explore the mechanism and electron dynamics of the spectral suppression near cutoff in HHG, we have performed the Bohmian trajectories calculations. We find that the spectral suppression of those harmonics in the region near the cutoff happened because the contributions of the short trajectories are small in HHG. However, the HHG intensity becomes a little higher for the last few harmonics just above the cutoff. This is because the HHG just above the cutoff arises from those electrons ionized near the peak of the laser pulse, where the ionization rate is the highest. The longer Bohmian trajectories dominated return to the core with lower energies, these trajectories contribute to the below-threshold harmonics. In addition, we find that the spectral suppression in HHG near cutoff is associated with the electronic dynamical origin in the laser field rather than the electronic structure of the atoms. Our results enable us to obtain a deeper understanding of the mechanism and electron dynamics of the HHG, and provide fresh new insight to this important field of ultrafast science and technology.

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