Half-cycle cutoff in near-threshold harmonic generation

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Abstract

A half-cycle cutoff is identified in the harmonic generation spectra near the ionization potential for short driving laser pulses. Unlike the half-cycle cutoff in the high-energy region, the newly found low-energy cutoff is strongly affected by the ionic potential and multiple return trajectories. We show their contribution clearly in the observable harmonic spectrum based on the reference of this cutoff structure. Our results are calculated from a numerical solution to the 3D time-dependent Schrödinger equation and a classical trajectory Monte Carlo method. By comparing the results from both methods, we analyze the time-dependent sub-cycle electron dynamics and provide a transparent explanation to this half-cycle cutoff. We further investigate the low-energy harmonic yield as a function of CEP for different pulse durations. The modulation depth of this yield drops rapidly when the pulse duration is increased, which can explain the CEP-dependence recently observed in the experiment.

1. Introduction

The interaction of strong laser fields with atoms or molecules can lead to high-order harmonic generation (HHG), which can be applied as table-top coherent light sources in a wide frequency range [1–4]. Recently, much attention has been paid to the below or near-threshold harmonics because of its potential applications as coherent vacuum-ultraviolet light sources in different circumstances [5–9]. Chini et al [9] investigated low-energy harmonics around the ionization threshold within the framework of the double optical gating, in which the linear-polarization window of the driving laser is very short (about 1.5 fs). They observed a conspicuous dependence of the near-threshold harmonics on the carrier envelope phase (CEP), while the dependence becomes invisible for a 5 fs linear driving pulse.

The studies on the CEP effects in HHG have mainly focused on the high-energy regime, where high energy half-cycle cutoffs were observed [10, 11]. The CEP effects are related to the electron dynamics happening within a single laser cycle [12] and can be applied to detect the ultrafast electron dynamics or select a single attosecond pulse [13, 14]. Very recently, the high-energy cutoffs have also been observed at a 1600 nm driving laser in the mid-infrared range [15].

The CEP effects in the high-energy HHG can be well explained by the simple man’s model with the potential only and the SFA cannot correctly describe low-energy electrons as it ignores the potential [18]. Thus the theoretical study of low-energy harmonics mostly relies on the numerical solution to the time-dependent Schrödinger equation (TDSE) [19, 22, 23]. A rough understanding is mainly based on the simple man’s model with the potential only accommodated in the propagation step [24, 25].
In this paper, we investigate the low-energy HHG driven by a few-cycle mid-infrared pulse. The harmonic spectra are calculated by the exact numerical solution to the TDSE [19] and the classical trajectory Monte Carlo (CTMC) simulation. Through comparison studies, we confirm that the CTMC can provide correct descriptions for the near-threshold harmonics. We identify a low-energy half-cycle cutoff near the ionization potential and the electron dynamics corresponds to this cutoff is analyzed. By a careful study of the low-energy cutoff, we can identify the roles of the multiple-return trajectories and the important effects of the ionic potential. Although their impacts are usually difficult to directly observe in a harmonic spectrum [25–27], we explicitly demonstrate their contributions to the harmonic spectrum in the low-energy cutoff structure. In addition, we investigate the pulse-length dependence of the integrated yield to explain the sensitive dependence on the pulse duration, as recently observed in the experiment [9].

2. Theoretical methods

The most reliable theoretical description of near-threshold harmonics is the numerical solution to the TDSE [19], which is accurate within the single active electron approximation. Here we perform the TDSE calculation to get the time-dependent electron wave function. We calculate the harmonic spectrum through a Fourier transform to the electron dipole acceleration. Besides, the time information is also extracted through a Gabor transform [28].

To interpret the TDSE results, we apply a classical trajectory Monte Carlo method [29, 30] to calculated the harmonic spectrum. For the first time, we use the CTMC in the near-threshold regime to calculate the harmonic spectrum. We show that the CTMC gives not only an alternative way to describe these low-energy harmonics, but also provides a transparent understanding from electron trajectories. Specifically, an ensemble of electrons are initialized, each of whose probability is weighted by a quasi-static ionization rate at its ionization time [31]. Then these electrons are propagated according to the Newton’s equation in the combined field of the laser pulse and the ionic potential. When the electron travels close enough to the core, one assumes that this electron recombines and at the same time a photon is recorded in the harmonic spectra. The photon energy is the summation of the potential energy and the electron’s kinetic energy. After the end of the laser pulse, the final harmonic spectra is calculated by counting the number of photons in each energy bin.

The recombination process does not have the classical correspondence, thus we are careful about the description of the recombination process in the CTMC model. The electrons travel around the core after ionization, when the distance of the electron to the core is smaller than a certain value, we assume the recombination process happens. In our model, we choose the recombination distance as the tunneling point, which is different for different trajectories depending on their ionization time. As a more crude approximation, we also calculate the harmonic spectrum at a fixed recombination distance (from 1 to 6 a.u.) for all the trajectories. We find that this crude choice does not change the resultant harmonic spectrum qualitatively. However, for a fixed distance, the short trajectories ionized when the electric field is small cannot be correctly described. As for these trajectories, they are ionized quite far away from the core when the electric field is quite small, and the electric field reverses immediately after the ionization. Those electrons have a long distance to be accelerated to the core and get a high energy when recombining. Although it does not affect the harmonic spectrum due to the low ionization rate, this is incorrect for the description of short trajectories. To avoid this problem, we choose the tunneling exit as the recombination distance.

3. Results and discussion

3.1. Comparison results and sub-cycle electron dynamics

In figure 1, we show the harmonic spectra for a model Helium [32] with the potential

\[ V(r) = -1 + (1 + 27r/16) \exp(-27r/8)/r. \]

The driving field is at 1800 nm with a peak intensity of \( 7 \times 10^{14} \text{ W cm}^{-2} \) and a pulse duration of 2.3-cycle full width in a sin-square pulse shape. The driving pulse is in the deep tunneling region, which ensures the correctness of the ionization probability. And the selection of He atom with a large ionization potential avoided the bound–bound transition from the ground state [19]. Both the driving field and the model atom are selected to make the CTMC method grab the main physics of this process. The results by the CTMC method agree very well with those from the TDSE. The well understood high-energy half-cycle cutoffs can be clearly observed. If one looks closely at the energy range around the ionization potential (about 0.9 a.u.), some CEP-dependent structures can also be identified in both figures 1(a) and (b). We show the amplified spectra for the low-energy part in a linear scale in figures 3(a) and (b). A low-energy cutoff structure can be identified, and it moves toward a lower energy when the CEP \( \phi \) increases from 0 to about 1.6 rad. When \( \phi \) is further increased, the spectrum becomes complicated due to the multiple return trajectories. In the rest part of this paper, we will only focus on these near-threshold harmonics.
At first, we look at the sub-cycle electron dynamics from the point view of the classical trajectories. By performing a Gabor transform in the TDSE calculations, one can get the emitting time (recombination time) at different harmonic energies [28]. In the CTMC simulations, the ionization time can be extracted along with the recombination time. The electrons’ trajectories are determined for certain ionization and recombination times. We present the time information at two different CEPs of the driving pulse, respectively calculated by TDSE shown in figure 2 (a) (b) and CTMC in figure (c) (d). It achieves good agreement for the recombination time calculated from the two different methods, including those multiple return trajectories. In the CTMC results in figures 2(c) and (d), the white dots represent all possible ionization times. The ionization time spreads around since there are lots of trajectories with a low ionization rate, which can not be observed in the color plot at the corresponding recombination time.

Now we can focus on the trajectories of low-energy electrons in figure 2. For the case of \( \phi = 0 \) shown in figures 2 (a) and (c), the harmonic signal mainly comes from those electrons ionized around time \( t_1 \) and \( t_2 \) with its recombination time respectively at \( t_{r1} \) and \( t_{r2} \) (the denoted time here represents a time interval instead of a single time point). The electrons ionized around time \( t_1 \) have a probability of a second return and can generate photons below the ionization threshold. However, because of the small electric field at this ionization time, one cannot observe too much low-energy signals in the spectra at this CEP. The signal near the ionization potential is mainly the contribution of the electrons ionized around \( t_{o1} \) for its first return. These electrons do not have time to reach a zero energy before the end of the pulse and their recombination at the end of the pulse forms the low

Figure 1. The harmonic spectra in the log scale, calculated respectively by TDSE in (a) and by CTMC in (b), for different CEP \( \phi, A, B, C \) marks the cutoffs for \( \phi = \pi/2 \), whose recombination time is respectively indicated in figure 2.

Figure 2. The recombination time calculated from TDSE for (a) \( \phi = 0 \), (b) \( \phi = \pi/2 \), in which the black line indicates the driving pulse. In (c) and (d), the recombination time calculated by CTMC is shown together with all the possible ionization time (white dots). The other laser parameters are the same with those in figure 1.
energy cutoff around 1.4 a.u. For the case of $\phi = \pi/2$, as shown in figures 2(b) and (d), the ionization times shifts leftwards to $t_i'$ and $t_i''$, respectively. The signals from electrons ionized at $t_i'$ almost disappear. And the electrons ionized at time $t_i''$ have enough time to propagate and acquire lower energies. This is the reason why the low energy half-cycle cutoff moves towards a lower energy when the CEP increased.

According to the above electron trajectory analysis, the cutoff energy can be estimated in an intuitive way. For an electron ionized at time $t_i$ and recombines at $t_f$, its kinetic energy at $t_f$ is given by $E = [A(t_f) - A(t_i)]^2/2$. For the high-energy cutoff, we know the maximum electron energy can be approximated by $3.17 U_p$, where $U_p = E_p^2/4\omega^2$ is the ponderomotive energy. In that case, one searches all the possible electron energies when it returns and find the maximum energy. Taking the electrons ionized around time $t_i''$ as an example, the high-energy cutoff is denoted by ‘A’ in figure 2(d) and its corresponding spectrum is also marked in figure 1(b). For the low-energy cutoff structure, a similar searching process can be performed. Instead of looking for the maximum returning energy, one should find the minimum energy for all possible return electrons. For the electrons ionized around the time $t_i'$, the low-energy cutoff structure is calculated to be 1.6 a.u. (denoted as ‘B’ in figures 2 and 1). While for the electrons ionized around the time $t_i''$, the low-energy cutoff extends to the energy region below the threshold (denoted as ‘C’). The low-energy cutoff structure corresponds to the lowest photon energy that can be observed.

For the case of a long driving pulse, the minimum recombination kinetic energy can always get a zero value since there always exist $t_i$ and $t_f$, which satisfy $A(t_i) = A(t_f)$. Then the low-energy cutoff energy is determined by the potential term and its position is below the ionization threshold. But for a short driving pulse, the ionization rate significantly differs from one half-cycle to another. The electrons ionized at the beginning of the pulse can contribute to the low-energy structures (e.g., the signals ionized at $t_i'$ in figure 2(c)), but this trajectories usually have a low probability and their contribution can not be clearly identified in the spectra. The influences of the pulse duration will be discussed in the last part of this paper.

### 3.2. The role of multiple return trajectories

If the CEP $\phi$ is further increased, the multiple return trajectories become important. To illustrate this multiple return effect, we show the CTMC spectra with only the first return in figure 3(d), by restricting the propagation time of the electrons within about one optical cycle. Comparing the spectra in figures 3(b) and (d), one can clearly observe the contribution of multiple return trajectories. There are two main differences in these two spectra. Firstly, when the CEP is small, the multiple return trajectories enhance the signals at energies slightly higher than the low-energy cutoffs (the bright line at higher energies is caused by the overestimate of some trajectories). Secondly, when the CEP becomes larger than 1.6 rad, the signal near the ionization threshold is mainly contributed by the multiple return trajectories.

In the harmonic spectrum from TDSE shown in figure 3(a), these two impacts of multiple return trajectories can also be clearly identified. For the small CEPs, the multiple return signals from TDSE spread at energies...
higher than the low energy cutoff instead of localization at one bright line as in the CTMC. This is because of the inexact weight of multiple return trajectories in the CTMC simulations. When the multiple return trajectories dominate for $\phi$ larger than 1.6 rad, the strongest signal moves to a higher energy when $\phi$ is increased. This is a combined effect of the electron trajectory and the ionization rate, which cannot be simply explained by searching the minimum energy. However, as we can see from figure 3(b), the CTMC method is still able to provide a qualitatively correct description.

The main difference between TDSE and CTMC is that we can observe only two bright lines in the results of CTMC while for the TDSE calculations, many bright lines can be observed. The reason of the discrepancy is that the CTMC model does not include all the multiple return trajectories. Here in our model, the electrons recombine to the core immediately when they are close enough to the core. The multiple return trajectories mainly come from the electrons which have relative large initial vertical velocities. It already travels away in the vertical direction when it returns to the core. After its return, it flies over the core and feels the force of the potential and goes nearer in the vertical direction. Thus it is possible to recombine at its next return. While in the picture of SFA, the electrons move across the core without seeing anything for its first return. The electrons only interact with the core at its final return. Thus in our description, the first and second return are overestimated compared with SFA.

### 3.3. The effects of the ionic potential

Now we turn to investigate the role of ionic potential. For this purpose, we carry out a TDSE simulation for the harmonic spectra by a short range potential given by $V(r) = -2.37 \exp(-r)/r$. This potential supports only one bound state with the same ionization potential as that of the model He. As shown in figure 3(c), there are some distinctive differences from the results for the long-range potential in figure 3(a). The most obvious effect is that the signal below the ionization threshold disappears for the case of the short range potential. For the below-threshold harmonics (BTHs), the corresponding electron’s energy is small and can be captured by the core before its recombination [19]. As the short-range potential does not have any excited states, it is impossible for the electrons to be captured and generate BTHs. For the long-range potential case, these BTHs can be reproduced by CTMC as in figure 3(b), and the integrated yield is shown in figure 4(a). The position when the BTHs appear around $\phi = 1.6$ rad is well reproduced. However, the harmonic yield calculated by the TDSE is resonance-enhanced by the bound states, which is beyond the classical description.

Another noticeable difference is that the low-energy cutoff position is different for the short-range and long-range potential. For better illustration of this effect, we calculate the low-energy cutoff by searching the minimum energy from the simple man’s model. We only focus on the electrons ionized around the time $t_{12}$, which varies with $\phi$. We show the minimum return energy in figures 3(a) and (c) as a red dashed line. The simple man’s model gives a better agreement with the actual cutoff for the short-range potential (black line in (c)) while the long-range potential has a lower actual cutoff energy (blue line in (a)). This observation means that the long range potential tends to shift the low-energy cutoff towards a lower energy. To quantify the shift as a function of the cut-off energy, we calculate the shift energy by subtracting the blue line in (a) and the black line in (c), and the difference is shown in figure 3(e). One notices that the shift increases when the cutoff energy decreases. To explain this phenomenon, we also calculate the shift from the CTMC method, shown in figure 3(c) as the red dashed line. The fact that the shift can be reproduced by the CTMC means that the shift is a classical effect. Actually, the ionic potential plays several roles here. On the one hand, the force of the potential changes the electron trajectory and thus the minimum kinetic energy increases due to this force. On the other hand, the potential energy must be accumulated to the electron’s total energy before its recombination, which reduces the total energy. This shift is caused by both the force of ionic potential in the propagation and the additional potential term in the recombination. This shift can only be observed in the low-energy cutoff. For the usually observed high-energy cutoff at $3.17 U_b + I_p$, the spectrum calculated from the short-range and the long-range potential is essentially identical. There is a third effect if we compare the harmonic yield for these two different potentials. We can clearly observe in both CTMC and TDSE calculations that the harmonic yield from the long-range potential is much larger than that of the short-range potential (results not shown graphically here). For the TDSE calculations, the ionization probability is higher for the long range potential and we cannot distinguish the effect of ionization probability from other effects. To quantify this problem, we calculate the total harmonic yield from the CTMC model. In the CTMC calculations, the ionization probability is the same for the same $I_p$ while the total harmonic yield is about 2.4 times higher from a long-range potential. We show the integrated harmonic yield calculated from CTMC in figure 5. As we can see, the yield from short range potential equals to the yield of a long regime potential when multiplied by a constant 2.4. All the other conditions are the same (except for the potential), the long range potential just drags more electrons back. The modulation of the yield by CEP is caused by different ionization probabilities in different fields. From a semi-classical point of view, the potential force can focus the electron in the propagation and enhance the rate of recombination.
Finally, we turn to investigate the pulse duration effect. Two effects have been observed recently in [9] when the pulse duration increases: the modulation depth drops and the harmonic yield fluctuates rapidly. To understand these observations, we integrate the harmonic yield near the ionization threshold for pulse duration of 2.3 cycles and 4 cycles, as shown in figure 4. For the shorter pulse case in figure 4(a), only one half-cycle dominates the tunnelling ionization so that the yield exhibits a single peak. The position of this peak will vary with the pulse duration, and this is a semi-classical effect which can be well reproduced by the CTMC method as shown in figure 4(a). When the pulse duration is further increased (shown in figure 4(b)), several half-cycles can contribute to the near-threshold harmonics. The two main effects observed in experiment [9] can be reproduced by the TDSE results. The first effect of this superposition is the decrease of the modulation depth, which can be well reproduced by the CTMC method. The second effect is the fluctuations observed in the TDSE, which comes from the interference between different trajectories. In the CTMC simulation, we do not include the phase information of different trajectories and the interference structures cannot be described. It should be noted that in [9], their driving laser is at 800 nm while the results here are from 1800 nm driving field. In our numerical calculations, we find that the main low-energy half-cycle cutoff structure does not change when the wavelength varies from 1800 to 800 nm. The results are shown in figure 6. As we can see, for the 1300 nm driving pulse, the cutoff structure is nearly the same for 1800 nm, except that the cutoff energy changes less with the CEPs and the extension of the cutoff is smaller. When it comes to the 800 nm driving pulse, the multi-photon effect gets involved and the extension of the cutoff is even smaller. However, we can still roughly observe the low energy cutoff.
In conclusion, a low-energy half-cycle cutoff in the near-threshold regime is observed and transparently interpreted. Based on the observation of this cutoff structure, multiple return trajectories and the role of ionic potential are studied as they are important in the low-order harmonics. Usually, their effects cannot be observed in a HHG spectrum since their impacts are not obvious for the high-energy harmonics. We directly identify their contributions to the observable spectra in the low-energy harmonics. This low-energy cutoff structure has been applied to explain the recent experimental observations in the near-threshold regime when the driving laser is short.

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Figure 6. The harmonic spectrum for 800 and 1300 nm, other parameters are the same as those in figure 1. The peak intensity is $7 \times 10^{14}$ W cm$^{-2}$ and the pulse duration of 2.3-cycle full width in a sin-square pulse shape.
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