Classical effect for enhanced high harmonic yield in ultrashort laser pulses with a moderate laser intensity

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Abstract
We study the influence of pulse duration on high harmonic generation (HHG) by exploring a wide laser-parameter region theoretically. Previous studies have shown that for high laser intensities close to saturation ionization intensity, the HHG inversion efficiency is higher for shorter pulses since the ground-state depletion is weaker in short pulses. Our simulations show that this high efficiency also appears for a moderate laser intensity at which the ionization is not very strong. A classical effect relating to shorter travel distances of the rescattering electron in shorter pulses is shown to contribute importantly to this high efficiency. The effect can be amplified significantly if a two-color laser field is used, suggesting a potential approach to increasing the HHG yield and generating short and bright attosecond pulses.

Keywords: ultrashort laser pulse, harmonic spectrum, HHG yield

(Some figures may appear in colour only in the online journal)

1. Introduction

Because of its promising application as an attosecond light source [1–3], high harmonic generation (HHG) has attracted a great deal of interest in recent years [4–6]. According to the well-known three-step model [7], the maximal energy of the harmonic (the cutoff energy) in the HHG is \( I_p + 3.17 U_p \). Here, \( U_p = E_0^2/4\omega_0^2 \) is the ponderomotive energy with \( E_0 \) and \( \omega_0 \) being the laser amplitude and frequency. \( I_p \) is the ionization potential of the ground state. To obtain shorter attosecond pulses, a higher cutoff energy is expected. To increase the HHG cutoff, one can increase the laser intensity or the wavelength. However, there are limitations for both. First, high laser intensities can induce the important depletion of the ground state, which decreases the HHG yield [8, 9]. Secondly, due to the diffusion effect, the HHG yield also decreases very quickly as the laser wavelength increases [10–12]. This decrease of the HHG yield also results in a decrease of the HHG conversion efficiency [13], limiting wider application of the HHG.

A great deal of effort has been devoted to overcoming these difficulties [14–16]. It has been found that the target atom can survive higher laser intensities in ultrashort laser pulses, resulting in much higher HHG cutoff energy and yield at high laser intensities near the saturation ionization intensity [17–21]. In addition, the use of two-color laser fields has been shown to be a very effective approach for increasing the HHG cutoff and producing shorter and brighter attosecond pulses [22–26].

In this paper, we further theoretically explore a procedure which could increase the HHG yield in a wide laser-parameter region. We focus on the influence of the pulse duration on the HHG with varying laser intensity and wavelength and working in both one-color and two-color laser fields. Our simulations through numerical solution of the time-dependent Schrödinger equation (TDSE) show that for a moderate laser intensity with relatively low ionization probability, the HHG efficiency still increases remarkably in an ultrashort laser pulse. Our analyses reveal a classical effect, which affects this...
phenomenon significantly: during the fast falling part of the short pulse, the rescattering electron is capable of obtaining the same energy by traveling a shorter distance and therefore enjoys a more efficient recollision for the HHG than it does in the long pulse. This classical effect becomes more remarkable as a two-color ultrashort pulse is used with increasing the HHG conversion efficiency significantly at diverse laser wavelengths.

The paper is organized as follows. In section 2, we introduce our theoretical methods. In section 3, we show our two-dimensional numerical results for enhanced HHG efficiency in short laser pulses as the laser intensity is relatively low. The classical effect responsible for enhancing the HHG yield is discussed in section 4. Extended discussions for three-dimensional cases, for different forms of the laser envelope and higher laser intensities are presented in section 5. In section 6, we show the potential application of the classical effect. Extended discussions on macroscopic phase matching are presented in section 7. Section 8 is our conclusion.

2. Theoretical methods

To explore a wide range of laser wavelength, we first use a two-dimensional (2D) model He atom to simulate the HHG. The Hamiltonian of the model atom studied here is $H(t) = \frac{p^2}{2} + V(r) + r \cdot E(t)$ (in atomic units of $\hbar = e = m_e = 1$). The potential used here has the form of $V(r) = -Z/\sqrt{\xi + r^2}$ with $r^2 = x^2 + y^2$. $\xi = 0.5$ is the smoothing parameter which is used to avoid the Coulomb singularity. $Z$ is the effective charge, which is modulated in such a manner that the ground state of the model atom has the ionization potential of $I_p = 0.9$ a.u. The electric field used here has the form of $E(t) = \frac{\hat{e} f(t)}{E_0} (\sin \omega_0 t + \varepsilon \sin 2\omega_0 t)$ with $\varepsilon = 0$ for one-color cases and $\varepsilon = 0.5$ for two-color cases. $f(t)$ is the envelope function. $\hat{e}$ is the unit vector along the laser polarization which coincides with the $x$ axis. To check the influence of the pulse duration on the HHG and simplify our discussions, we use a $3n$-cycle laser pulse which is switched on and off linearly over $n$ optical cycles with $n = 1, 3, 8$. The whole pulse duration is $NT$ with $N = 3n$ and $T = 2\pi/\omega_0$.

We work with a space grid size of $L_x \times L_y = 1638.4 \times 102.4$ a.u. for the $x$ and the $y$ axes. The TDSE is solved using the operator-splitting method [27], where the quantum evolution operator is factored into a product of kinetic and potential propagators arranged in a symmetric way and a fast Fourier transform routine is used to transform the wave function between momentum space and coordinate space. The space steps used in our simulations are $\Delta x = \Delta y = 0.4$ a.u. and the time step is $\Delta t = 0.05$ a.u. The

![Figure 1. Wavelength dependence of the HHG yield $Y(N,\lambda)$ for N-cycle one-color (a, b) and two-color (c, d) laser pulses. Results are obtained through full TDSE simulations (a, c) and the short-trajectory simulations (b, d). The ratio of $Y(N)/Y(N_0)$ at $\lambda = 1400$ nm is also shown.](image.png)
accuracy of our TDSE simulations with the operator-splitting method is $\sim \Delta t^3$.

To avoid reflections of the wave packet from the boundaries, after each time step the TDSE wave function $\psi(r, t)$ of $H(r)$ for each dimension of $x$ and $y$ is multiplied by a mask function to absorb the continuum wave packet at the boundary. The mask function along the $x$ axis has the following form of $F(x) = \cos^2/8(|x| - x_0)/(L_x - 2x_0)$ for $|x| \geq x_0$ and $F(x) = 1$ for $|x| < x_0$. Here, $x_0$ is the absorbing boundary from where the mask function becomes to work along the $x$ axis. Similarly, the mask function $F(y)$ along the $y$ axis has the form of $F(y) = \cos^2/8(|y| - y_0)/(L_y - 2y_0)$. For the full TDSE simulations, we have used the parameters of $x_0 = L_x/8$ and $y_0 = L_y/8$. For the laser parameters explored in the paper, the maximal classical displacement $x_{\text{max}} = 2E_0/w_0^2$ [7] of the electron along the laser polarization can easily be represented in our numerical grids. The absorbing procedure with $x_0 = L_x/8$ and $y_0 = L_y/8$ is also sufficient to retain the contributions of the first return of the rescattering electron to the core (including long and short trajectories) and multiple returns [28] to HHG.

Alternatively, in the mask functions of $F(x)$ and $F(y)$, we can set the absorbing boundary $x_0$ at $x_0 = E_0/w_0^2$ with that at $y_0$ unchanged (i.e., $y_0 = L_y/8$) [29]. For our present cases, this treatment removes the contributions of the long trajectory and multiple returns to the HHG as the contribution of the short trajectory is not influenced basically. $E_0/w_0^2$ is the quiver amplitude of the classical electron in the laser field. It is also the maximal displacement the rescattering electron can travel along the short trajectory in the laser field. It should be stressed that multiple returns and collisions may lead to a quantum-path interference effect and can be important in both calculations [30] and experiments. Here, the absorbing procedure with $x_0 = E_0/w_0^2$ separates the contributions of short trajectories from other trajectories, allowing one to identify the properties of the different trajectories in short laser pulses. Below, for differentiation from the full TDSE simulation with $x_0 = L_x/8$, we denote the simulation with $x_0 = E_0/w_0^2$ as the short-trajectory simulation.

The HHG spectrum for one harmonic $\omega$ along the laser polarization $\hat{e}$ can be evaluated using [31]

$$F(\omega) = \int \langle \psi(r, t) | \hat{e} \cdot \nabla \psi(r, t) \rangle e^{i\omega t} dt. \quad (1)$$

The HHG power spectrum $S(\omega)$ is $S(\omega) = |F(\omega)|^2$. Once the power spectrum $S(\omega)$ for the harmonic $\omega$ is obtained, we calculate the average HHG yield $Y(N, \lambda)$ for an $N$-cycle laser pulse using [32]

$$Y(N, \lambda) \propto \frac{1}{N(\Omega_2 - \Omega_1)} \int_{\Omega_1}^{\Omega_2} S(\omega) d\omega. \quad (2)$$

Here, $\Omega_1 = I_\nu$ and $\Omega_2$ is the cutoff energy of the spectrum which depends on the laser parameters. $\lambda$ is the laser wavelength. For the pulse shape used here, $Y(N, \lambda)$ can be used to

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Figure 2. HHG spectra for $N$-cycle one-color (a, b) and two-color (c, d) laser pulses at $\lambda = 1400$ nm. Results are obtained through full TDSE simulations (a, c) and the short-trajectory simulations (b, d), and divided by the cycle number $N$. 

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[Diagram showing HHG spectra for one-color and two-color laser pulses at different cycles and for different $N$ values, with log-log scale for intensity and harmonic energy.]

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For the harmonic $n$, $\omega_n$ is the cutoff energy of the spectrum for the harmonic $n$.
compare the HHG conversion efficiency at different pulse durations directly [13].

In the paper, unless mentioned elsewhere, our discussions will be performed for the peak laser intensity of $I = 3 \times 10^{14}$ W cm$^{-2}$. With this moderate peak intensity, the ionization yield of the model He atom is low.

### 3. Enhanced HHG yield

In figure 1(a), we plot the wavelength dependence of the average HHG yield in a one-color field for different pulse durations. The contrast of the curves is remarkable. One can observe that the average HHG yield is the highest for the 3-cycle pulse at different wavelengths. This yield decreases as the cycle number $N$ increases. At long wavelengths such as $\lambda = 1400$ nm, the yield of the 3-cycle pulse is several times higher than the 9-cycle result, and one order of magnitude higher than that of the 24-cycle pulse. Note that, in this case, the whole HHG yield of $NY(N, \lambda)$ for $N = 3$ is also several times higher than that for $N = 24$. The contrast of the HHG yields at different pulse durations becomes more remarkable when the short-trajectory simulations are performed. As shown in figure 1(b), when the contributions of the long trajectory and multiple returns are excluded, the short-trajectory HHG yield for $N = 3$ is one order of magnitude higher than $N = 9$, and almost two orders of magnitude higher than $N = 24$. For a two-color laser field, however, this remarkable difference occurs even for the full TDSE simulations, as shown in figure 1(c). Here, the ratio of $Y(3, \lambda)$ versus $Y(24, \lambda)$ with $\lambda = 1400$ nm arrives at 82, implying a significant increase of the HHG efficiency. This significant increase of the efficiency is further amplified as the short-trajectory contributions are considered, as shown in figure 1(d). Since the short-trajectory simulations are closely associated with the classical motion of the electron, these results in figures 1(b) and (d) imply that the potential mechanism, which increases the HHG yields at short pulses, is related to the classical aspect of the electron. Next, we explore the mechanism in detail.

Figure 2 plots the HHG spectra of 3-cycle versus 9-cycle pulses for the typical wavelength of $\lambda = 1400$ nm at peak laser intensity $I = 3 \times 10^{14}$ W cm$^{-2}$. For comparison, these spectra are divided by the cycle number $N$. We mention that the wavelength of 800 nm is usually used in experiments. In this paper, the wavelength of 1400 nm is chosen as a typical case. First, with the long laser wavelength of 1400 nm, the cutoff of the HHG spectrum increases remarkably in comparison with the case of 800 nm at the same laser intensity. With the wide spectral regime, the structure of the HHG spectrum can be read more clearly. Secondly, it has been shown that the long laser wavelength shows some merits for the generation of isolated attosecond pulses in the spectral region both in theories [15] and experiments [33, 34]. Lastly,
many laboratories now have optical parametric amplifiers which can easily go to 1400 nm.

For the one-color case in the left column of figure 2, one can observe from figure 2(a) the full simulations: (i) the spectrum with $N = 3$ (the solid-black curve) is higher than that of $N = 9$ (the dashed-red curve), especially for the low-energy part. (ii) In both cases, the HHG yield decreases as the harmonic energy increases. (iii) The spectrum of $N = 3$ shows three plateaus with the cutoff positions of $\omega = 1.7$ a.u., 4.5 a.u. and 7.8 a.u. respectively. Around the second cutoff $\omega = 4.5$ a.u. (corresponding to the electron kinetic energy of $E_p = \omega - I_p = 1.7U_p$), a robust peak can be observed. A similar multiple-plateau structure of the HHG spectra in ultrashort laser pulses has been observed in [19], where the important role of the laser phase is addressed. As the short-trajectory simulation is executed, the robust peak of $N = 3$ survives our treatments and the spectrum of $N = 9$ becomes flat, resulting in a remarkable contrast of these two spectra with different $N$, as shown in figure 2(b).

For the two-color case, the contrast of the two spectra at different pulse durations is more remarkable, even for the full TDSE results, as shown in the right column of figure 2. In this case, the spectrum of $N = 3$ in figure 2(c) of full simulations shows four plateaus with the cutoff positions of $\omega = 1.4$ a.u., 4.8 a.u., 7.1 a.u. and 10.3 a.u., respectively. Around the second plateau, which also has a robust peak at $\omega = 4.8$ a.u. (corresponding to $E_p = 1.85U_p$), the spectrum of $N = 3$ is one order of magnitude higher than that of $N = 9$. By comparison, the spectrum of $N = 9$ shows two striking plateaus corresponding to the third and the fourth plateaus of $N = 3$. As we perform the short-trajectory simulations, the intensity of the second plateau does not decrease basically, as shown in figure 2(d). The origin of the plateaus can be well understood through wavelet analysis of the TDSE dipole acceleration combined with the quantum–orbit (QO) theory [28, 35], as discussed below. These comparisons in figure 2 explain the remarkable differences of the HHG yields at different pulse durations discussed in figure 1. From the comparisons, one can also conclude that the high HHG efficiency of short pulses is closely related to the robust HHG peaks of $1.7U_p$ and $1.85U_p$ indicated in figure 2, which are less influenced by our different absorbing procedures in TDSE simulations. These results are further explored through the time-frequency analysis.

4. Classical effects

In figure 3, we show wavelet-analysis [36] results (the color coding) for the corresponding spectra in figures 2(a) and (b).
Figure 5. Same as figure 4, but for two-color laser pulses.

For comparison, we also show the electron trajectory of the first return with the excursion time of the electron shorter than a laser cycle (the black-square curve), predicted from the QO model. For $N = 3$, from figure 3(a) of the full TDSE results, one can observe that (i) the distributions match the theory predictions basically; (ii) the distributions imply three HHG cutoffs. The first one with $E_p = 0.5U_p$ around the return time $t_r = 2.4T$ has the largest amplitude. The second one with $E_p = 1.7U_p$ also appears around $t_r = 2.4T$ and has a comparable amplitude with the first one, as indicated by the dashed arrow. The third one with $E_p = 3.2U_p$ near to $t_r = 2T$ has the smallest amplitude. The first one can be identified as arising from multiple returns, and the second and the third ones come from the first return. For short-trajectory simulations in figure 3(b), the first one disappears, as the second and the third ones keep their amplitudes with the prevailing role of the second one. Note, the contributions of the long trajectory around the minus-chirp part of the black-square curve also disappear basically due to the absorbing procedure used here. For $N = 9$, the situation is different. The full TDSE results in figure 3(c) show large amplitudes around the electron trajectories of multiple returns (these trajectories are not shown here), in agreement with the results in [10]. In figure 3(d) of short-orbital simulations, the contributions of multiple returns disappear. The survived distributions, however, show smaller amplitudes than that around $E_p = 1.7U_p$ in figure 3(b). To understand the large amplitude located at $E_p = 1.7U_p$ in figure 3(b), in figure 4, we further compare the maximal displacement $x_m$ [29], which the electron can travel as it ionizes at the time $t_i$ and returns at $t_f$ for $N = 3$ versus $N = 9$.

To obtain the maximal displacement $x_m$, we first calculate the complex ionization time $t_i^{\text{fl}}$ and the return time $t_f^{\text{fl}}$ (the real parts of the complex times are considered the physical ionization time $t_i$ and the return time $t_f$) by the QO model. Then we evaluate the maximal displacement using [37]

$$x_m \equiv x_m(t) = (E_0/\omega_0^2) \text{Re} \left[ \sin \omega_0 t - \sin \omega_0 t_i^{\text{fl}} \right]$$

$$+ p_0 (\omega_0^2/E_0) (t - t_i^{\text{fl}})$$

with $\nu(t) = \text{Re} [P \omega_0/\omega_0^2 \cos(\omega_0 t)] = 0$ and $\text{Re}(t_i^{\text{fl}}) < t < \text{Re}(t_f^{\text{fl}})$. $\nu(t)$ is the electron velocity, and $P = (E_0/\omega_0) [\sin \omega_0 t_i^{\text{fl}} - \sin \omega_0 t_f^{\text{fl}}]/[\omega_0 (t_f^{\text{fl}} - t_i^{\text{fl}})]$ is the saddle-point momentum. The above expression of $x_m(t)$ is related to the following process. The rescattering electron is first accelerated and is driven far away from the core by the laser field. As the laser field changes its direction, the electron is decelerated. At a certain time $t$, when the velocity of the electron $\nu(t)$ is decelerated to zero, the electron arrives at its maximal displacement $x_m(t)$. Then it is driven back into the core.

We mention that the maximal displacement $x_m$ obtained here agrees well with that obtained using the classical procedure introduced in [29]. For the first return of the electron to the core including short and long electron trajectories, the value of $x_m$ is larger for a larger excursion time $\tau = t_f - t_i$ and therefore it reflects the effects of quantum diffusion (the spreading of the continuum wave packet) [28]. For multiple...
returns, the situation is somewhat more complex. The values of $x_m$ for multiple returns is similar to the long-trajectory one for trajectories with similar returning energy. However, Coulomb focusing can play an important role in the case of multiple returns, which decreases the diffusion effects [10].

Our comparisons are performed for two typical long trajectories (denoted using $L_1$ and $L_2$ in figure 4) with the same return energy $E_p = 1.5U_{pp}$, near $E_p = 1.7U_{pp}$ of the large amplitude in figure 3(b). Both trajectories have the ionization times $t_i$ located in the flat-top part of the pulse and near to the peak of the field, as shown in figures 4(c) and (f), and are expected to contribute importantly to the HHG. The return times of the two trajectories are different. As the $L_1$ trajectory returns to near the flat-top part of the pulse, the $L_2$ trajectory returns in the falling part of the pulse (the laser pulse starts to fall at $2T$ for $N = 3$ and $6T$ for $N = 9$). For the 3-cycle case in figure 4(b), the displacement $x_m$ for the $L_2$ trajectory is 81 a.u. It is 160 a.u. for $L_1$, two times larger than the $L_2$ one. Considering that the spreading of the wave packet is proportional to the electron displacement, these results imply that the $L_2$ trajectory has an amplitude several times larger than the $L_1$ one, in agreement with the wavelet-analysis result in figure 3(b). For the 9-cycle case in figure 4(e), the situation is different. The $L_2$ trajectory has a maximal displacement of $x_m = 142$ a.u. which is near the 164 a.u. of the $L_1$ one. As a result, the wave packet spreading is comparable for the two trajectories in the 9-cycle case, resulting in similar amplitudes for them, as seen in figure 3(d). We mention that short trajectories have smaller displacements $x_m$ than the corresponding long ones. However, they usually ionize at a time farther away from the peak of the field and therefore have smaller amplitudes than the long ones (this can also be seen from the wavelet-analysis results in figure 3. For this reason, we do not discuss them here).

In combination with the distributions in figure 3(b) versus figure 3(d), the contrast of the maximal displacements $x_m$ for the $L_2$ trajectory in figure 4(b) versus figure 4(e) suggests that the higher HHG efficiency for the short pulse of $N = 3$, observed in figure 1(b), is closely related to the shorter excursion distance of the rescattering electron in the fast-falling part of the short pulse. In the following, we call the effect associated with this shorter travel distance of the electron in short pulses and which increases the HHG efficiency at moderate laser intensities, the classical effect. It is different from the effect which gives rise to the high HHG inversion efficiency in short laser pulses with high laser intensities [17–21]. The latter effect is associated with weaker depletion of the ground state of the system in shorter pulses and is a quantum effect. It should be noted that the classical effect plays an important role only in the lower HHG energy region of the spectra, when the quantum effect contributes to the whole HHG energy region.

This classical effect becomes more remarkable in the two-color case, as shown in figure 5. Here, our analyses are also performed for two typical long trajectories of $L_1$ and $L_2$. The ionization times of the both long trajectories are located...
at the flat-top part of the pulse and are near the peaks of the two-color field, as shown in figures 5(c) and (f). In addition, the electric-field amplitudes at the two ionization times of L1 and L2 in the two-color case are nearer each other than in the one-color one. For the short-pulse case of \( N = 3 \), one can observe from figure 5(b) that the maximal displacement of the L2 trajectory is \( x_m = 65 \) a.u., as the L1 trajectory shows a maximal displacement of \( x_m = 166 \) a.u., almost three times larger than the L2 one. For the long-pulse case of \( N = 9 \), they are 121 a.u. and 169 a.u., respectively, as shown in figure 5(e). The shorter excursion distance of the L2 trajectory in figure 5(b) of \( N = 3 \) suggests that this trajectory contributes significantly to the HHG in the short-pulse case. It corresponds to the cutoff of the second plateau with high intensity in figures 2(c) and (d). We mention that the TDSE cutoff position of the second plateau in figure 2(c) is \( U_{1.85} \) for the 3-cycle case, somewhat higher than the model prediction of \( U_{1.7} \) in figure 5(a). This is also the one-color case of \( 1.7U_p \) in figure 2(a) versus \( 1.5U_p \) in figure 4(a). This difference can partly arise from the nonadiabatic effect in ultrashort pulses. In comparison with \( x_m = 81 \) a.u. with \( E_p = 1.5U_p \) in figure 4(b), this shorter excursion distance \( x_m = 65 \) a.u. of the L2 trajectory with the higher return energy \( E_p = 1.7U_p \) in figure 5(b) also suggests that the HHG efficiency is higher in the two-color case than in the one-color case, in agreement with our analyses in figure 1.

5. Extended considerations

5.1. Three-dimensional simulations

To check our results, we have also performed three-dimensional (3D) simulations for the model He atom with the soft-core potential of \( V(r) = -\frac{Z}{\sqrt{\xi + r^2}} \) and \( r^2 = x^2 + y^2 + z^2 \). The definitions of the parameters \( Z \) and \( \xi \) are the same as in our 2D cases. The 3D calculations are very time-memory consuming. Here, we work with a grid size of \( L_x \times L_y \times L_z = 819.2 \times 51.2 \times 51.2 \) a.u. for the \( x \), \( y \), and \( z \) axes, respectively. Our calculations are performed for \( I = 3 \times 10^{14} \) W cm\(^{-2} \) and \( \lambda = 1400 \) nm corresponding to the laser parameters used in figure 2. Similar absorbing procedures as in 2D cases are also used in performing full TDSE simulations and short-trajectory simulations. The results are presented in figure 6.

One can observe from figure 6 that the 3D results are similar to the 2D ones in figure 2. First, the spectra of \( N = 3 \) show a robust peak which appears around \( E_p = \omega - I_p = 1.7U_p \) for one-color cases in the left column of figure 6 and around \( E_p = 1.85U_p \) for two-color cases in the right column. Secondly, this robust peak is more remarkable in short-trajectory simulations than in full simulations and in two-color cases than in one-color cases. As discussed in figure 2, this robust peak arises from the classical effect, which increases the HHG efficiency importantly in short pulses. The 3D

![Figure 7](image-url)
results in figure 6 also show that this HHG efficiency is strikingly higher in the short pulse than in the long one. All of the characteristics are in agreement with our 2D results in figure 2.

5.2. Sin-square-envelope pulses

To check our results, a sin-square-envelope pulse with the total duration of $N$ optical cycles (not FWHM) is also used in our calculations and relevant results are presented in figure 7. The results in figure 7 are also similar to those in figure 2, with showing a robust peak in the spectra of $N = 3$. Here, the position of the peak is around $E_p = 0.75U_p$ for one-color cases and $E_p = 0.9U_p$ for two-color cases, somewhat lower than those in figure 2. In addition, the maximal cutoff position of the spectrum of $N = 3$ is also somewhat lower than that of $N = 9$. However, in comparison with the results in figure 2, the results for $N = 3$ presented here show higher inversion efficiency. For example, in figure 7(b), the spectrum of $N = 3$ is higher than that of $N = 9$ in the whole energy region, different from the results in figure 2(b). In addition, the curve of $N = 3$ in figure 7(d) also shows a cutoff located at $\omega = 8.3$ a.u. Around the cutoff, the spectrum of $N = 3$ is one order of magnitude higher than that of $N = 9$. All of the characteristics can be understood by virtue of the analyses of the quantum–orbit and the maximal displacement of the rescattering electron, and can be attributed to shorter excursion distances of the rescattering electron in shorter pulses for obtaining the same kinetic energy.

5.3. High laser intensities

As the laser intensity increases and becomes close to the saturation intensity, the situation is somewhat different. As shown in figure 8(a), for the case of $I = 1 \times 10^{15}$ W cm$^{-2}$ and $\lambda = 1400$ nm, the HHG spectrum of $N = 3$ is higher than that of $N = 9$ in the whole energy region, even the average over the cycle number $N$ is not performed here. This can be understood from the ground-state depletion. For the long pulse of $N = 9$, the ionization is stronger than that of $N = 3$, resulting in a significant depletion of the ground state and accordingly a remarkable decrease of the HHG yield. However, in this case, the classical effect can still be read from the spectrum, which manifests itself as a harmonic peak around $E_p = 1.67U_p$, as indicated by the dashed-dotted arrow. This peak is more remarkable in short-trajectory simulations, as shown in figure 8(b). In addition, the cutoff position in the spectrum of $N = 3$ in figure 8(a) seems somewhat larger than that of $N = 9$. This phenomenon is clearer for the case of $I = 9 \times 10^{14}$ W cm$^{-2}$ and $\lambda = 1600$ nm, as shown in figure 8(c). We mention that for the case in figure 8(c), the ionization is somewhat weaker than that in figure 8(a). Accordingly, the spectra of $N = 3$ and $N = 9$ are nearer to each other here. In addition, a harmonic peak around

Figure 8. HHG spectra for $N$-cycle one-color laser pulses at $I = 1 \times 10^{15}$ W cm$^{-2}$ and $\lambda = 1400$ nm (a, b) and $I = 9 \times 10^{14}$W cm$^{-2}$ and $\lambda = 1600$ nm (c, d). Results are obtained through full TDSE simulations (a, c) and the short-trajectory simulations (b, d) in 2D cases.
$w = 1.68U_\phi$ can also be observed in the spectrum of $N = 3$ in figure 8(c). This peak is more striking in figure 8(d) of short-trajectory simulations.

6. Potential application of this effect

In figure 9, we show the HHG spectra and the ellipticity of relevant harmonics obtained for 2D $\text{H}_2^+$ with the internuclear distance $R = 2$ a.u. for $N$-cycle one-color (a, b) and two-color (c, d) laser pulses at $I = 5 \times 10^{14}$ W cm$^{-2}$ and $\lambda = 1400$ nm. Results are obtained through short-trajectory simulations, and the HHG spectra in (a) and (c) are divided by the cycle number $N$. The orientation angle (the angle between the molecular axis and the laser polarization) is $\theta = 40^\circ$.

It should be noted that in figures 9(b) and (d), the ellipticity for $N = 9$ is higher than $N = 3$ near the 100th harmonic. However, the HHG intensity for $N = 9$ is remarkably lower than that for $N = 3$ in figures 9(a) and (c). One therefore can expect that the intensity of the elliptically-polarized EUV pulse generated with $N = 9$ is weaker than that with $N = 3$.

In figures 10(a) and (b), we plot the train of pulses $I_x(t)$ and $I_y(t)$, the expressions of which are as follows [1]:

$$I_{\ell x,y}(t) = \int_{\omega_0}^{\omega_{\text{cut}}} |d_{\ell x,y}(\omega)|e^{-i\omega t}d\omega|^2. \quad (4)$$

Here, $d_{\ell x,y}(\omega)$ denotes the component of harmonic $\omega$ of the dipole moment, which is parallel to the laser polarization ($d_x$) or perpendicular to the laser polarization ($d_y$). Our calculations are performed for the spectral region from the 67th to the 90th (i.e., $\omega_0 = 67\omega_0$ and $\omega_{\text{cut}} = 90\omega_0$), which is located in the interference-effect-dominating energy region of the spectra shown in figures 9(a) and (c). Note, parallel harmonics (i.e., harmonics which are emitted parallel to the laser polarization) are used for the calculation of $I_x(t)$ and perpendicular harmonics (i.e., harmonics emitted perpendicular to the laser polarization) are used for $I_y(t)$. In addition, the integration on $\omega$ is used in the expressions of $I_x(t)$ and $I_y(t)$.
in our simulations, instead of the sum on odd harmonics as used in [1]. Since, in short laser pulses, the single odd harmonic cannot be resolved from the spectra.

Figures 10(a) and (b) provide information for the relative intensity of the pulses, generated with harmonics parallel versus perpendicular to the polarization of the driving laser field. In figures 10(c) and (d), we also plot the curve of \( I_y(t) \) as a function of \( I_x(t) \). The results are obtained through short-trajectory simulations with \( I = 5 \times 10^{14} \) W cm\(^{-2} \) and \( \lambda = 1400 \) nm for 3-cycle one-color (a, c) and two-color (b, d) laser pulses.

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7. Extended discussion on macroscopic phase matching

It should be stressed that in the above discussions based on single-atom responses to strong laser fields, the macroscopic propagation of both fundamental and harmonic fields is not considered. In real experiments, to obtain a bright HHG output beam which may be applied to attosecond science, the emission of a large number of target atoms over an extended region of the nonlinear medium must be added in phase. This phase matching may alter the conclusions obtained for a single atom. This phase matching condition is met when the driving laser and the generated HHG radiation travel at the same phase velocity in the nonlinear medium [34]. Factors which influence the phase velocity of the laser field or the HHG radiation (such as the neutral atom dispersion and the dispersion of the free-electron plasma that is created as the medium is ionized) can result in phase mismatch and therefore reduce the HHG yield.

A great deal of effort has been devoted to the phase matching effect of HHG in recent years [42–46]. It has been well recognized that a long driving laser wavelength (\( \sim 2 \) \( \mu \)m) leads to a shorter phase matching window and the resulting HHG emission arises from one or sub-laser cycles. A short driving wavelength results in a longer phase matching window and causes HHG emission from multiple laser cycles. Consequently, with the use of a long laser wavelength, a single attosecond pulse can be obtained from the HHG [33]. In addition, the HHG conversion efficiency is higher for longer driving laser wavelengths due to phase matching [15], resulting in a scaling of \( \lambda^{4.1} \) [34] for the highest energy harmonic that can be phase matched in the soft x-ray region. In particular, when intense few-cycle driving laser pulses with long laser wavelengths are used, phase mismatch of HHG can be remarkably reduced and isolated attosecond pulses can be extracted from plateau harmonics [15].

3D calculations with long propagation distance are very time-memory consuming. Here, with the above discussions on macroscopic phase matching of HHG, we discuss the
potential influence of phase matching on the classical effect explored in the paper. Firstly, the classical effect stands for short laser pulses where phase mismatch can be reduced in comparison with long laser pulses, as shown in [15, 42]. Secondly, the classical effect occurs even with moderate laser intensities at which the ionization is not strong and the influence of the ionization-induced plasma on the phase velocity of the driving laser is small. In this case, the phase matching of HHG is easier to achieve [33]. Thirdly, with the use of quasi-phase-matching (QPM) methods [47], one can shift the phase-matching window in time. This could create a short temporal window closer to the temporal regime where the classical effect dominates in the HHG [48]. Lastly, the classical effect works for long laser wavelengths at which conditions for ideal phase-matching can be fulfilled and the HHG yield can be increased remarkably [15, 33, 34]. Therefore we expect that with short laser pulses at long laser wavelengths and moderate laser intensities and with QPM methods, it is possible to observe the classical effect in experiments.

8. Conclusion

In summary, we have shown that the HHG efficiency in an ultrashort laser pulse is influenced significantly by a classical effect. The effect is closely associated with the shorter excursion distance of the rescattering electron as it ionizes near the peak of the short laser pulse and returns in the fast falling part of the pulse. This shorter excursion distance suppresses the spread of the wave packet and increases the efficiency of the HHG. Harmonics with high intensity relating to the classical effect are generally emitted in a half laser cycle and therefore provide a potential way to generate short and strong EUV pulses, which have important applications in ultrafast science such as tracking the nonlinear response of bound electrons [49]. By choosing the molecule as the target, harmonics associated with this classical effect can also be used to generate short and bright elliptically-polarized EUV pulses. We expect that this effect also has an important influence on rescattering induced other strong-field processes such as high-order above threshold ionization (HATI) and nonsequential double ionization (NSDI). This effect increases the amplitudes of the electrons which return in the fast falling part of the short pulse. If these returning electrons with large amplitudes are rescattered by the core, they can manifest themselves in the energy spectra of HATI. Similarly, as these electrons with large amplitudes recollide with the core, they can also excite or ionize other bound electrons and leave their footprints in the momentum distributions of NSDI.

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