Optimization of three-color laser field for the generation of single ultrashort attosecond pulse

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Abstract: We present an efficient and realizable scheme for the generation of an ultrashort single attosecond (as) pulse. The feasibility of such a scheme is demonstrated by solving accurately the time-dependent Schrödinger equation using the time-dependent generalized pseudospectral (TDGPS) method. This scheme involves the use of the optimization of the three-color laser fields. The optimized laser pulse is synthesized by three one-color laser pulses with proper relative phases. It can provide a longer acceleration time for the tunneling and oscillating electrons, and allows the electrons to gain more kinetic energy. We show that the plateau of high-order harmonic generation is extended dramatically and a broadband supercontinuum spectra is produced. As a result, an isolated 23 as pulse with a bandwidth of 163 eV can be obtained directly by superposing the supercontinuum harmonics near the cutoff region. We will show that such a metrology can be realized experimentally.

OCIS codes: (020.2649) Strong field laser physics;(020.4180) Multiphoton processes.

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1. Introduction

The availability of attosecond (as) extreme ultraviolet pulse is on the verge of opening up a new field of science [1-3]. Attosecond laser permitted observation of control of electron wave packets [4], probing of nuclear dynamics [5] and electronic dynamics [6], attosecond time-resolved spectroscopy [7], tomographic imaging of molecular orbital [8], etc. The generation of much shorter attosecond pulses has continued to attract much interest and has become one of the most active research directions in attosecond metrology today.

Among the approaches to produce attosecond pulses, the high-order harmonic generation

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(HHG) seems to be the most promising one[1, 9]. The HHG procedure can be well understood by the semiclassical three-step model [10]. According to the three-step model, the electron tunnels through the barrier formed by the Coulomb potential and the laser field in the beginning, then it oscillates quasifreely driven by the laser field and acquires additional kinetic energy, and finally it can recombine with the parent ion and emit radiation. The harmonic spectrum is characterized by a rapid drop at low orders followed by a broad plateau where all the harmonics have the same strength and a sharp cutoff around harmonic energy $I_p + 3.17U_p$, where I_p is the atomic ionization potential and U_p is the ponderomotive energy, i.e., the cycle-averaged kinetic energy of an electron gained in a monochromatic laser field.

At present, many schemes have been proposed to generate an ultrabroad supercontinuum spectrum and obtain extreme short attosecond pulse, such as using two-color laser fields [11, 12], a few-cycle laser pulse [13, 3], the chirped laser pulse [14, 15], quantum path control [16, 17], polarization gating [18, 19], and long-wavelength pumping [20, 21]. Among them, Sansone *et al*. have reported the generation of a 130 as isolated XUV pulse using the near-infrared driving pulse. Very recently, Goulielmakis *et al*. have experimentally demonstrated the generation of an isolated 80 attosecond pulses using only a single 3.3 fs pump pulse. Moreover, many theoretical works have been done to generate single sub-100 as pulse [22, 23].

In this paper, we present a realizable approach for efficiently ultrashort attosecond pulses generation by creating a longer acceleration time for the oscillating electrons allowing electrons to obtain more kinetic energy from laser fields. Our pulses are constructed by superposing a range of synchronized high harmonics driven by the optimized three-color laser field. We know that a one-color driving field interacts with an atom and generates coherent high-frequency radiation through ionizing, accelerating, and recombining processes. These three processes occur every half optical cycle [24, 25]. In our method, we choose a optimized three-color laser field that can be realized experimentally. This shape of laser pulse allows that the processes of ionization, acceleration, and recombination occur more than half optical cycle and enables the HHG plateau extended significantly. It is also important to note that our approach can obtain an efficient and very broadband supercontinuum than those of some other published works, such as the cases of two color laser pulse, the longer wavelength, and so on. Although our simulation of the single attosecond pulse generation is single atom response, the recent studies show that the single-atom spectrum is similar even considering propagation effects [26]. Our theoretical calculations show that this proposed scheme can obtain the efficient broadband supercontinuum harmonic spectra, which is used to produce a very short attosecond pulse less than one atomic unit of time (the atomic unit of time is about 24 as), which is the time scale of electron motion in atoms.

2. Theoretical methods

The HHG and attosecond pulse generation can be investigated by solving the time-dependent Schrödinger equation (TDSE). In the length gauge, the TDSE of hydrogen atom can be written as (in atomic units)

$$i\frac{\partial \psi(\mathbf{r},t)}{\partial t} = \hat{H}\psi(\mathbf{r},t) = [\hat{H_0} + \hat{V}(\mathbf{r},t)]\psi(\mathbf{r},t), \tag{1}$$

where $\hat{H_0}$ is the Hamiltonian of the Hydrogen atom.

$$\hat{H_0} = -\frac{1}{2}\frac{d^2}{dr^2} + \frac{\hat{L}^2}{2r^2} - \frac{1}{r}, \tag{2}$$

and $V(\mathbf{r},t)$ is the time-dependent atom-field interaction

$$\hat{V}(\mathbf{r},t) = -\mathbf{E(t)}\cdot\mathbf{r} = -zE(t). \tag{3}$$

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The TDSE is solved accurately and efficiently by means of the time-dependent generalized pseudospectral (TDGPS) method [13, 27]. Once the time evolution of the wave function $\psi(\mathbf{r},t)$ is determined, the HHG power spectrum can be obtained. The time-dependent induced dipole acceleration can be given by means of Ehrenfest's theorem [27]

$$d_A(t) =< \psi(\mathbf{r},t)|\frac{dz^2}{dt^2}|\psi(\mathbf{r},t) >=< \psi(\mathbf{r},t)|-\frac{z}{r^3}+E(t)|\psi(\mathbf{r},t) >, \tag{4}$$

and the HHG power spectrum can be determined, which is proportional to the modulus squared of the Fourier transform of $d_A(t)$. It is given by

$$P_A(\omega) = |\frac{1}{t_f - t_i}\frac{1}{\omega^2}\int_{t_i}^{t_f} d_A(t)e^{-i\omega t}dt|^2. \tag{5}$$

By superposing several harmonics, an ultrashort pulse can be generated,

$$I(t) = |\sum_q a_q e^{iq\omega t}|^2, \tag{6}$$

where $a_q = \int d_A(t)e^{-iq\omega t}dt$.

To study the detailed spectral and temporal structures of HHG, we perform the time-frequency analysis by means of the wavelet transform,

$$A_\omega(t_0,\omega) = \int d_A(t)\sqrt{\omega}W[\omega(t-t_0)]dt. \tag{7}$$

For the harmonic emission, a natural choice of the mother wavelet is given by the Morlet wavelet:

$$W(x) = \frac{1}{\sqrt{\tau}}e^{ix}e^{\frac{-x^2}{2\tau^2}}. \tag{8}$$

3. Results and discussion

Considering several experimental demonstrations obtained a few-cycle pulse (∼5fs) [3], the laser pulses we choose are near 5 femtosecond for the experimental realizations. In our calculations, the optimized three-color laser field has the following form:

$$E(t) = \sum_{i=1}^{3} E_i f(t)cos(\omega_i t + \phi_i), \tag{9}$$

where $f(t)$ is the Gaussian pulse with 5-fs pulse duration. E_i, ω_i, and ϕ_i are the amplitudes, frequencies, and relative phase for each laser pulse, respectively. We choose the intensity $I_1 = 1.0\times10^{14}W/cm^2$, $I_2 = 1.2\times10^{14}W/cm^2$ and $I_3 = 1.5\times10^{14}W/cm^2$. The frequencies ω_1=0.057 a.u. (800 nm), ω_2=0.023 a.u. (2000 nm) and ω_3=0.021 a.u. (2200 nm), and the relative phase $\phi_1 = \pi$, $\phi_2 = 0$ and $\phi_3 = -\frac{\pi}{2}$, respectively. This optimized pulse shape is presented in Fig. 1(d).

In order to give an explanation of why such three-color combination was chosen, the optimizing process is presented in Fig. 1(a)-(d). Figure 1(a) shows a 5-fs 800 nm laser pulse with peak intensity $I = 1.0\times10^{14}W/cm^2$ (black solid line) and a 5-fs 2000 nm laser pulse with peak intensity $I = 1.2\times10^{14}W/cm^2$ (red dotted line), respectively. If we choose this few-cycle 800nm laser pulse to generate attosecond pulse, it can only obtain a single 200 as-300 as pusle [13]. If we desire to extend the HHG plateau and obtain ever shorter attosecond pulse, the longer driving wavelength is a good choice [20, 21]. Such as this 2000 nm laser pulse is presented

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in Fig. 1(a), because the ponderomotive energy is proportional to $I\lambda^2$, which implies that the harmonic cutoff can be extended using longer wavelength of the driving laser field. However, only using one-color longer wavelength driving field has a serious disadvantage that the harmonic yield is greatly decreased than the shorter wavelength laser pulse [28]. To enhance the harmonic yield, some schemes have been proposed to use a longer wavelength laser pulse in combination with a higher frequency pulse [29]. Figure 1(b) presents the combination of the 800 nm and 2000 nm laser pulses as shown in Fig. 1(a). Here, the major local maximum of peaks are marked by *A*, *B*, *C*, and *D*. We can see that the effective wavelength is increased for

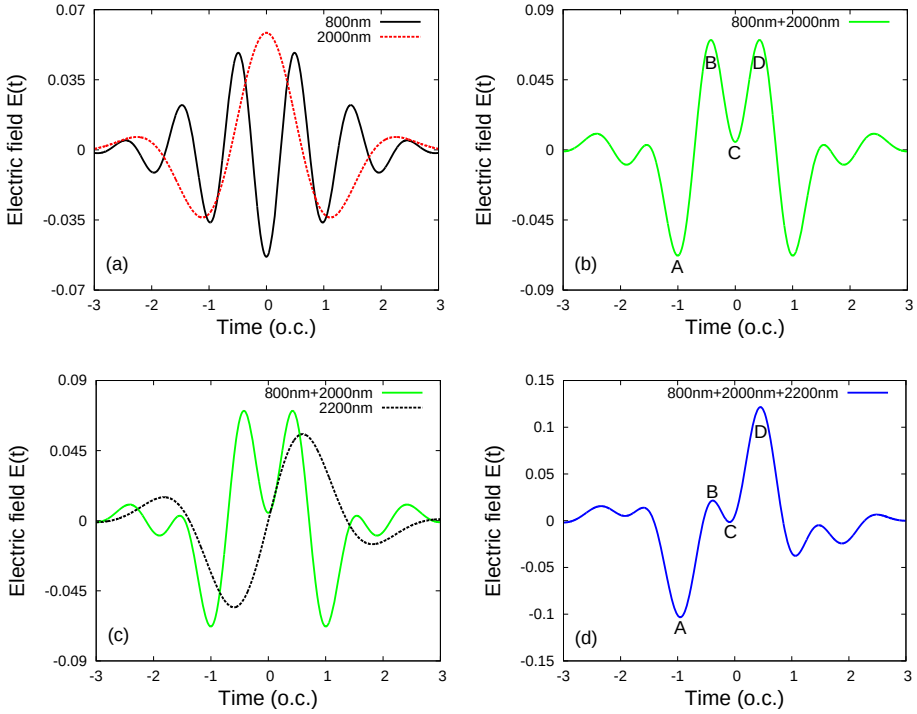

Fig. 1. (a) A 5-fs 800 nm laser pulse with peak intensity $I = 1.0 \times 10^{14} W/cm^2$ (black solid line) and a 5-fs 2000 nm laser pulse with peak intensity $I = 1.2 \times 10^{14} W/cm^2$ (red dotted line). (b) The combination of the 5-fs 800 nm and 2000 nm laser pulse shown in (a). (c) A 5-fs 2200 nm laser pulse with peak intensity $I = 1.5 \times 10^{14} W/cm^2$ (black dotted line). For reference, two-color combination case (green solid line) is also presented. (d) The optimized three-color laser field.

this combined pulse, and the intensity of the peaks is also. Therefore, the quasifreely electrons can obtain higher cutoff energy when the electrons are ionized from the major local ionization peak *A* and return to emit the HHG radiation at the major local emission peaks *B* and *D*. However, such simple combination of two-color laser field can not efficiently be used for generation of a broadband supercontinuum due to the interaction of HHG emission from peaks *B* and *D*. In order to generate a broadband supercontinuum, we must eliminate the impact of peak *B*. So we add a 5-fs 2200 nm laser pulse with peak intensity $I = 1.5 \times 10^{14} W/cm^2$ (black dotted line) to the combined two-color laser pulse, as shown in Fig. 1(c). It just can eliminate the peak *B*. This results are presented in Fig. 1(d). Namely, We obtain a optimized three-color laser pulse. We can see that the intensity of peak *B* is deceased, and the intensity of peak *A* and *D* is increased. The advantages of such changes are not only to significantly extend the plateau of HHG, but

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also obtain a broadband supercontinuum. This is very important for the successful generation of the single ultrashort attosecond pulse.

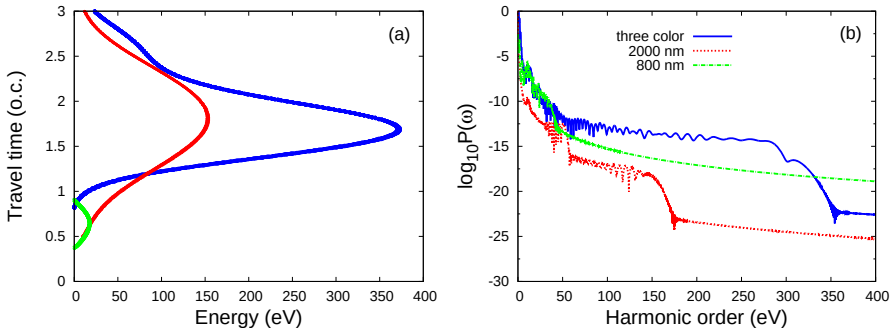

Fig. 2. (a) The travel time versus returning energy for the 800 nm laser pulse (green line), the 2000 nm aser pulse (red line), and the optimized three-color laser field (blue line), respectively. The same laser parameters are used in Fig. 1(a) and Fig. 1(d). (b) Corresponding to the high-order harmonic spectra.

To explain extension of the plateau of the HHG from the optimized combination of three-color laser pulse, we analyze the dependence of the returning kinetic energy on the travel time of the electrons for the 800 nm laser pulse, the 2000 nm laser pulse, and the optimized three-color laser pulse, respectively. The laser parameters are the same as Fig. 1(a) and Fig. 1(d). According to the semiclassical three-step model [10], the motion of the electron after tunneling ionization can be described by the classical mechanics. We can obtain the dependence of the returning kinetic energy on the travel time of electron by solving Newtonian equation. For the 800 nm laser pulse case, and travel time with the maximum return energy is around 0.6 optical cycle, the corresponding maximum return energy is about 30 eV. For the 2000 nm laser pulse case, and travel time with maximum return energy is about 1.65 optical cycles (using 800 nm laser period), and the corresponding maximum return energy is about 150 eV. For the optimized three-color laser case, the travel time with the maximum return energy is also about 1.65 optical cycles, and the corresponding maximum return energy is about 365 eV, which is much higher than the 2000 nm laser pulse case. Namely, the plateau of HHG can be effectively extended by the optimized three-color laser field. On the other hand, the shape of the optimized three-color laser pulse allows the processes of the ionization, acceleration, and recombination occur in 3.0 optical cycles. But for one-color 800 nm laser field case, these processes happen only in 1.0 optical cycle. Though the longer processes have happened for 2000 nm laser pulse case, it can not obtain a lager harmonic cutoff than the optimized three-color case [see Fig .2(b)]. In Fig. 2(b), we compare the HHG spectra from hydrogen atom driven by these three corresponding laser fields, respectively. It is seen that the HHG cutoff for these three cases agree well with the semiclassical simulations shown in Fig. 2(a). For the 2000 nm laser pulse case, there are two plateaus, the cutoff energy of the first plateau is about 55 eV, which is generated from the first returning electrons. The cutoff energy of the second plateau is about 150 eV, which comes from the contribution of the multi-rescattering electrons. In general, the conversion efficiency of the harmonic by multi-rescattering electrons is lower. Though the higher cutoff energy of the HHG is obtained for the 2000 nm laser case, the harmonic yield is greatly decreased than the optimized three-color laser field. Moreover, it does not generate an efficient broadband supercontinuum. In fact, our optimized three-color laser pulse not only can significantly enhance the harmonic spectrum, but also greatly extend the harmonic generation cut-off. It is also important to note that our approach can obtain an efficient and very broadband

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supercontinuum.

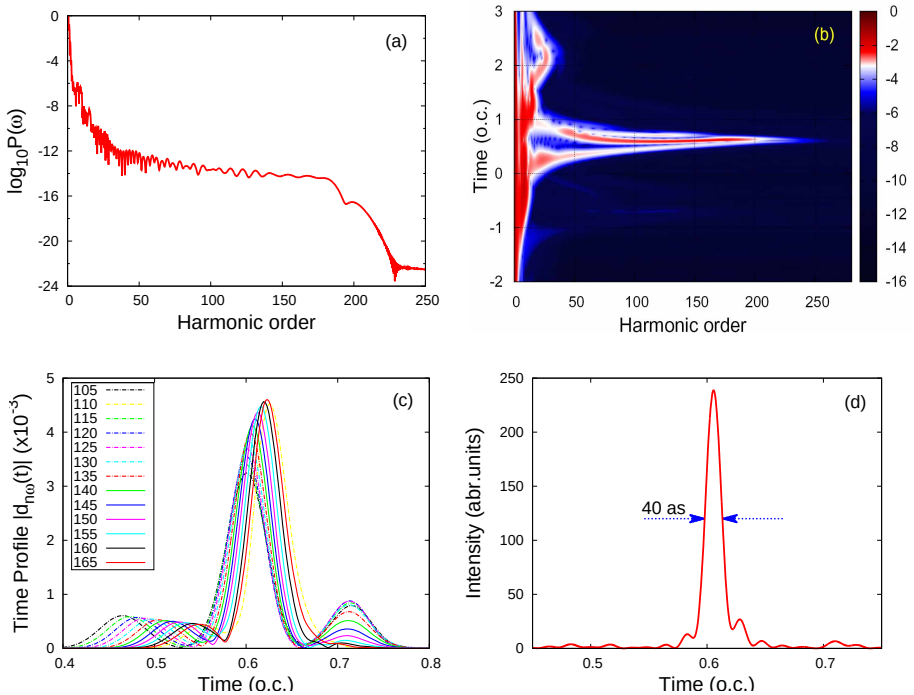

Fig. 3. (a) The HHG power spectra from hydrogen atom driven by the optimized three-color laser field shown as in Fig. 1(d). (b) Wavelet time-frequency profile of the HHG power spectra. (c) Dipole time profiles of consecutive harmonics near the cutoff. (d) The temporal profiles of the attosecond pulse by superposing the harmonic from 105^{th} to 165^{th} order.

Now we analyze the single ultrashort attosecond pulse generation by the optimized three-color laser field. In Fig. 3(a), we observe the broadband supercontinuum spectra near cutoff region driven by the optimized three-color laser field, the laser field parameters are same as the previous case. To figure out the detailed spectral and temporal time dependent structures of HHG, we perform a time-frequency analysis by means of the wavelet transform of the induced dipole acceleration, which is shown in Fig. 3(b). There is one major emission burst, which the contribution of the long quantum trajectory is dominant, and the short quantum trajectory is weak. On the other hand, the long trajectory electrons emit in a shorter time range and contribute to each order of harmonics near cutoff region. The corresponding time profiles of the consecutive harmonics of 105^{th} to 165^{th} harmonic order near the cutoff region are shown in Fig. 3(c). The emission time of the short trajectory electrons is dispersed from 0.4 optical cycle to 0.57 optical cycle. But the long trajectory electrons exhibit higher intensity peaks, which are generated in the relatively concentrated emission time. So it is possible to obtain a single attosecond pulse by superposing the harmonics near cutoff region. In Fig. 3(d), we show the generation of the attosecond pulse with the duration of 40 as, which is observed by superposing 60 order harmonics from the 105^{th} to the 165^{th} order. It is significant that this obtained attosecond pulse is strong and regular.

To further broaden the supercontinuum and compress the attosecond pulse duration, we now

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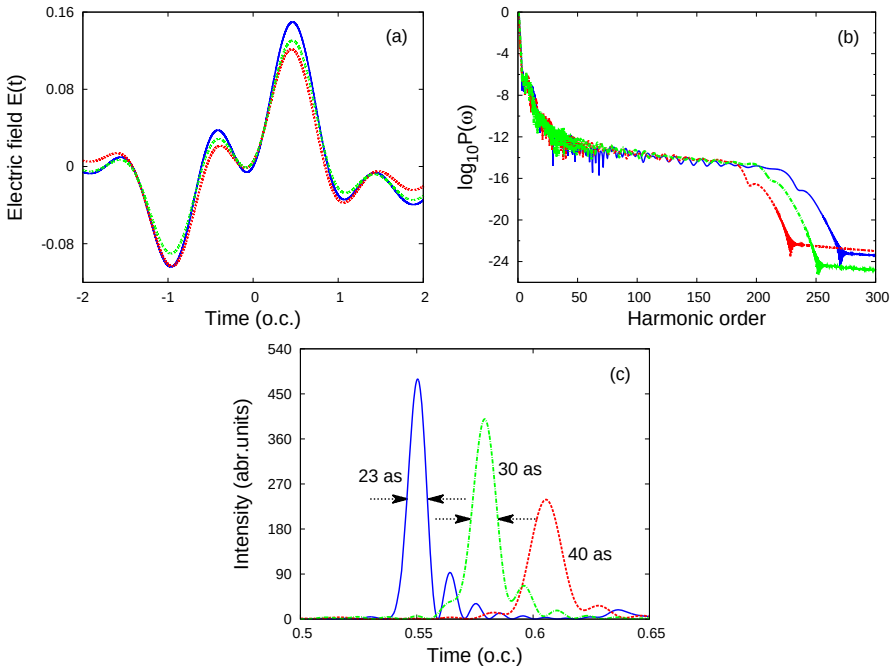

Fig. 4. (a) Optimized three-color laser field. The green dashed line represents laser frequency ω_2=0.019 a.u. (2400 nm). The blue solid line represents laser frequency ω_2=0.019 a.u. (2400 nm) and intensity $I_1 = I_2 = 1.5 \times 10^{14} W/cm^2$ and $I_3 = 1.7 \times 10^{14} W/cm^2$. The other laser parameters are the same as those in Fig. 1(d). (b) The corresponding HHG power spectrum and (c) Temporal profiles of the attosecond pulses by superposing the HHG near cutoff region. For comparison, the previous case (red dotted line) is also included in (a)-(c).

coherently control the HHG in more intense optimized three-color laser field. This intense optimized three-color laser field can be easily realized by changing the wavelength or the intensity of one of the three one-color lasers. First, we only increase the wavelength for one of three one-color laser [ω_2=0.019 a.u. (2400 nm)], other parameters are the same as those in Fig. 1(d), this case is shown in Fig. 4(a) (green dashed line). We can see that the intensity of the maximum peak is higher than the previous case [For comparison, the previous case is also presented in Fig. 4(a)-4(c) (red dotted line)]. The corresponding HHG power spectrum are shown in Fig. 4(b). It is seen that the HHG cutoff is remarkably extended than the previous case. The supercontinuum spectra is more broadened, which means that a much shorter single attosecond pulse can be significantly obtained. In Fig. 4(c), we show the generation of the 30 as pulse (green dashed line) is observed by superposing the HHG from the 105^{th} to the 185^{th} order harmonics. Second, we intense the laser intensity ($I_1 = 1.5 \times 10^{14} W/cm^2$, $I_2 = 1.5 \times 10^{14} W/cm^2$ and $I_3 = 1.7 \times 10^{14} W/cm^2$), and the frequency ω_2 is kept to 0.019 a.u. (2400 nm), other parameters keep the same as the last one, this case is also shown in Fig. 4(a) (blue solid line). The intensity of the maximum peak is much higher than the previous two cases. The corresponding HHG power spectra and attosecond pulse (blue solid line) are also shown in Fig. 4(b) and 4(c), respectively. It is seen that the much broader supercontinuum spectra is produced. As a result, an isolated 23 as pulse with a bandwidth of 163 eV is obtained directly by superposing the HHG from the 90^{th} to the 195^{th} order harmonics. The duration of this attosecond pulse is less than one atomic unit of time, and reaches the time scale to the real time observation of the motion

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of electrons in atoms and molecules.

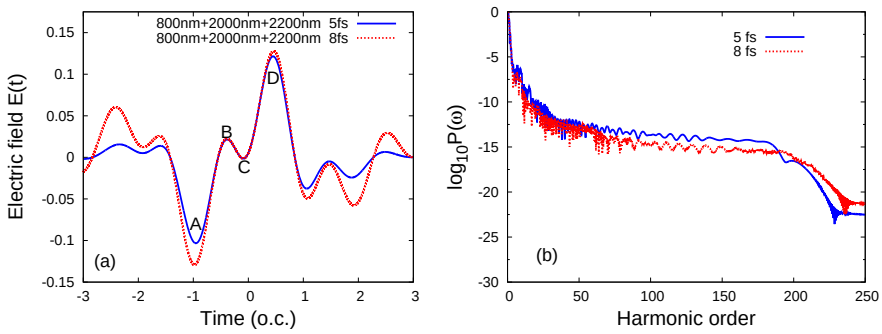

Fig. 5. (a) The optimized three-color laser pulse with 5-fs and 8-fs duration, respectively. The other laser parameters are the same as in Fig. 1(d). (b) Corresponding to the high-order harmonic spectra.

Indeed, the generation of a few-cycle laser pulse is a experimental challenge. However, we can obtain the similar shape of the optimized three-color laser pulse by short pulse with experimental realizable duration (6-10 fs). In Fig. 5(a), we present the optimized three-color laser pulse (red dotted line), which is combined by three one-color laser pulses with 8-fs full width at half maximum (FWHM), the other parameters are the same as Fig. 1(d). For reference, the previous case is also presented (blue solid line). The pulse shapes of these optimized three-color lasers are similar from peak *A* to peak *D* (the most decisive part of the generation of a single as pulse), only peak intensity of the 8 fs laser case is increased. These changes do not affect the generation of the supercontinuum harmonics spectra, as shown in Fig. 5(b). In Fig. 5(b), we present the HHG power spectrum from hydrogen atom driven by the optimized three-color laser pulses with 5-fs and 8-fs duration, respectively. We can see that a broadband supercontinuum is obtained from the Hydrogen atom driven by the 8 fs optimized three-color laser field. An isolated 35 as pulse can be generated by this broadband supercontinuum. Therefore, we present this method to produce a short as pulse by the pulse shape of the optimized three-color laser with longer duration, which is realizable experimentally.

4. Conclusion

In conclusion, we present an efficient scheme to produce a single ultrashort attosecond pulse by using the optimized three-color laser field. This scheme is demonstrated by solving accurately the TDSE using the TDGPS method. Our study confirms that the optimized three-color laser pulse can provide a longer acceleration time for the oscillating electrons, and allows electrons to gain more kinetic energy from the laser field. We show that the plateau of high-order harmonic generation is extended dramatically and the broadband supercontinuous spectrum is produced. As a result, an isolated 23 as pulse with a bandwidth of 163 eV can be obtained directly by superposing the supercontinuum harmonics near the cutoff region. We hope to this proposed scheme can be be realized experimentally.

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