Broadband water window supercontinuum generation with a $\omega + 3\omega/2$ multicycle two-colour pulse*

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We propose a method to generate a high-efficiency broadband water window supercontinuum with a $\omega + 3\omega/2$ multicycle two-colour pulse. Our results reveal that the $3\omega/2$ laser pulse can simultaneously modulate the acceleration step and the ionization step, which not only broadens the bandwidth but also enhances the yield of the generated supercontinuum. An ultra-broadband supercontinuum from 290 eV to 555 eV covering the whole water window is generated. Using this method, we expect that an isolated 62-as pulse with a minor pre-pulse can be directly obtained.

Keywords: high-order harmonic, attosecond pulse, supercontinuum

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1. Introduction

The generation of extreme ultraviolet (XUV) attosecond pulses makes it possible to study the dynamic of electrons in atoms and molecules.[1] Currently, the production of attosecond pulses is mainly based on high harmonic generation in atomic gases experimentally. In particular, the broadband supercontinuum covering the spectral range between the K-absorption edges of carbon (284 eV) and oxygen (543 eV), is more attractive for synthesizing the ultrafast water window X-ray pulse, which can capture images of live cells with high contrast. In the breakthrough work in Ref. [2], a train of attosecond pulses were generated with a multicycle laser pulse. However, for practical application, an isolated attosecond pulse is more useful.[3] So much effort has been made to extract a single attosecond pulse.[4,5] To generate an isolated attosecond pulse, the emission time of the harmonics should be confined in one-half driving pulse, which has been achieved by using a few-cycle driving pulse.[6] An isolated attosecond pulse has been first produced by filtering harmonics in the cutoff of the spectrum with a few-cycle laser pulse interacting with a jet of neon atoms.[7] Very recently, Goulielmakis *et al.*[8] brought through the 100-as-barrier. In their experiment, a sub-4 fs near-single cycle driving pulse has been employed to generate a 40-eV supercontinuum and an 80-as pulse has been obtained. Recently, Hong *et al.*[9] theoretically generated a broadband window supercontinuum in a neutral rare-gas medium by using a phase-stabilized few-cycle mid-IR pulse tailored by a much weaker 800-nm pulse. In our previous work, we proposed a method to generate a broadband supercontinuum using a few-cycle two-colour laser pulse and a high-order pulse,[10] from which an isolated 24-as pulse could be directly obtained. Wu *et al.*[11] proposed a method to extend high-order harmonics and generate an isolated attosecond pulse by optimizing the chirping parameters of the few-cycle laser pulses. Ye *et al.*[12] theoretically also generated isolated attosecond pulse by the two-colour pulse composed of IR field and UV field. However, such a few-cycle driving pulse can be achieved only with a state-of-the-art laser system, which assigns a limited applicability to the scheme.

The mechanism of high harmonic generation can be well understood using the three-step model:[13] ionization, acceleration and recombination. During the recombination, a photon is emitted with energy equal to the ionization potential plus the kinetic energy of a recombining electron. Since the ponderomotive energy is proportional to the square of the wavelength of the driving laser pulse, an effective approach to significantly extend the harmonic cutoff is to adopt a longer-wavelength driving laser pulse. Recently, it has been theoretically[14] and experimentally[15,16] shown

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that a mid-IR driving pulse not only improves harmonic conversion efficiency but also reduces harmonic chirps, which is beneficial for enhancing the generated attosecond pulse intensity and shortening the generated attosecond pulse duration.

To reduce extremely experimental requirements, many theoretical proposals[17−20] have been suggested to generate a single attosecond pulse. In particular, it has been proposed that the multicycle $\omega + 2\omega$ two-colour field can significantly enlarge the difference between the highest and the second highest half-cycle photon energies, thereby forming a broadband supercontinuum near the cutoff, from which a broadband isolated attosecond pulse is obtainable.[17,18] The polarization gating technique has also been used to produce broadband supercontinuum.[21] Zhang *et al.*[19] proposed a method to obtain an ultra-broadband supercontinuum using the modulated polarization gating. Very recently, we also proposed a method to obtain an ultra-broadband supercontinuum combining mid-infrared chirped-pulse modulation and generalized polarization gating,[22] which supports the isolated sub-100 attosecond pulses generation with tunable wavelength.

In this work, we propose a method to simultaneously control the acceleration step and the ionization step in the high harmonic generation (HHG) process for the efficient generation of a water window supercontinuum with a $\omega + 3\omega/2$ multicycle two-colour pulse. The $3\omega/2$ laser pulse not only broadens the bandwidth but also enhances the yield of the generated supercontinuum. The generated supercontinuum from 290 eV to 555 eV almost covers the whole water window, from which an isolated 62-as pulse with a minor pre-pulse can be directly obtained. The ionization probability of an He gas is also estimated by ADK model[23] to be below 0.1%.

2. Model and method

The harmonics and the attosecond pulse generation can be studied by numerically solving the one-dimensional time-dependent Schrödinger equation. In our calculation, the soft Coulomb potential[24] $V(x) = -1/\sqrt{a + x^2}$ is chosen with $a = 0.484$ to match the ionization potential $E_0 = -0.903$ a.u. of a real He atom. The electric field can be expressed as

$$E(t) = E_0 f_0(t) \cos(\omega_0 t + \phi_{\rm CEP}) + E_1 f_1(t - \tau) \cos(\omega_0 (t - \tau)). \quad (1)$$

Here E_i and ω_i $(i = 0, 1)$ are the peak amplitudes and frequencies of the driving pulse and the control field, respectively; τ is the time delay between the driving pulse and the control field; $f_i(t) = \cos^2(\pi t/Tl_i)$ $(i = 0, 1)$ present the profiles of the driving pulse and the control field; $\phi_{\rm CEP}$ is the carrier-envelope phase of the driving pulse, and it is set to be 1.5π throughout the paper; Tl_0 and Tl_1 are set to be $10T_0$ and $14T_1$ respectively; T_0 and T_1 are the periods of the driving pulse and the control field, respectively.

The one-dimensional time-dependent Schrödinger equation is solved accurately and efficiently by means of the split-operator method.[25] Once the time evolution of the wave function $\psi(x, t)$ is determined, the time-dependent induced dipole acceleration can be given by means of Ehrenfest's theorem

$$a(t) = -\langle \psi(x, t) \left| \frac{\partial V(x)}{\partial x} - E(t) \right| \psi(x, t) \rangle. \quad (2)$$

The harmonic spectrum can be obtained, which is proportional to the modulus squared of the Fourier transform of $a(t)$

$$P_{\rm A}(\omega) = \left| \frac{1}{\sqrt{2\pi}} \int_{-5T_0}^{5T_0} a(t)\, {\rm e}^{-{\rm i}\omega t} {\rm d}t \right|^2. \quad (3)$$

By superposing several orders of the harmonics, an ultrashort pulse can be obtained with the temporal profile as

$$I(t) = \left| \sum_q a_q\, {\rm e}^{{\rm i}q\omega t} \right|^2, \quad (4)$$

where $a_q = \int_{-5T_0}^{5T_0} a(t)\, {\rm e}^{-{\rm i}q\omega t} {\rm d}t$.

3. Results and discussion

In order to demonstrate our scheme, we first investigate the HHG process using the semiclassical three-step model. In our calculation, we choose $\omega_0 = 0.0228$ a.u. and $\omega_1 = 3\omega_0/2$ corresponding to $\lambda_0 = 2000$ nm and $\lambda_1 = 1333$ nm, respectively. $E_0 = 0.1$ a.u. and $E_1 = 0.042$ a.u., which correspond to the intensities of 3.5×10^{14} W/cm^2 and 6×10^{13} W/cm^2, respectively. τ is set to be $-0.5T_0$.

The electron dynamics of the HHG process and the ionization rate by the ADK model[23] in the driving pulse and in the two-colour field are shown in Figs. 1 and 2, respectively. The ionization time and the recombination time are shown with crosses and unfilled circles, respectively. Figure 1(a) shows the

electric field of the multicycle driving pulse. Figures 1(b) and 1(c) indicate the classical electron trajectories and the ionization rate in the multicycle driving pulse alone, respectively. As shown in Fig. 1(b), there are many peaks of trajectories during the pulse and the difference between the highest and the second highest half-cycle photon energies is too small to produce supercontinuum. Figure 2(a) shows the electric waveforms of the fundamental (dotted line), control (dashed line) and two-colour field (solid line). From Fig. 2(c), one can clearly see that under the control of the $3\omega/2$ pulse, the ionizations take place mainly from $-0.4T_0$ to $-0.05T_0$ and from $0.05T_0$ to $0.4T_0$, and other ionization peaks are successfully suppressed. Consequently, it is clear from Fig. 2(b) that a large portion of the electron trajectory from $0.0T_0$ to $0.5T_0$ contributes to the supercontinuum generation. Taking into account the above results, we can conclude that a supercontinuum from 290 eV to 570 eV can be generated.

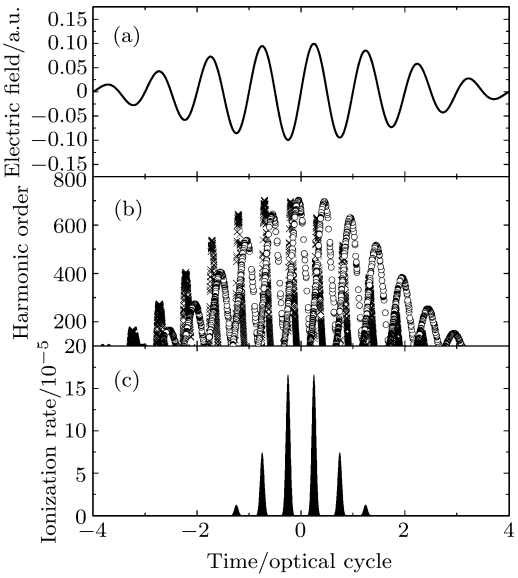


Fig. 1. (a) Electric waveform of multicycle driving field, (b) dependence of kinetic energy E_k on ionization time (crosses) and recombination time (unfilled circles), and (c) Ammosov–Delone–Krainov (ADK) ionization rate (solid gray line).

In the following, we investigate the harmonic generation to confirm the above results. The harmonic spectrum (thick solid line) is shown in Fig. 3(a). As shown in this figure, the spectrum cutoff is approximately at the 895th harmonic (555 eV). The spectrum above the 470th (290 eV) becomes continuous. The modulations on the supercontinuum are due to the interference between the short and the long quantum paths. For comparison, the spectrum (thin dotted line) of the high harmonics with the fundamental field alone is also shown in the figure. It reveals that the $3\omega/2$ pulse significantly broadens the bandwidth of the supercontinuum and enhances the harmonic yields by one order. In order to further understand the spectrum structure shown in Fig. 3(a), we also investigate the emission time of the harmonics for the case in terms of the time-frequency analysis method.[26]

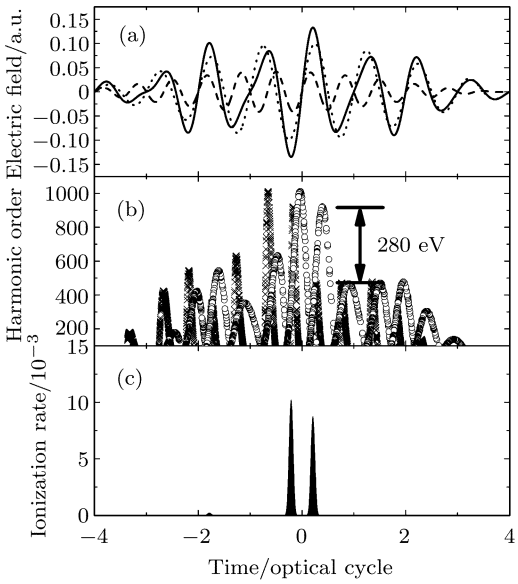


Fig. 2. (a) Electric waveforms of the fundamental (dotted line), control (dashed line) and two-colour field (solid line); (b) dependence of the kinetic energy E_k on the ionization (crosses) and recombination times (unfilled circles); and (c) ADK ionization rate (solid gray line).

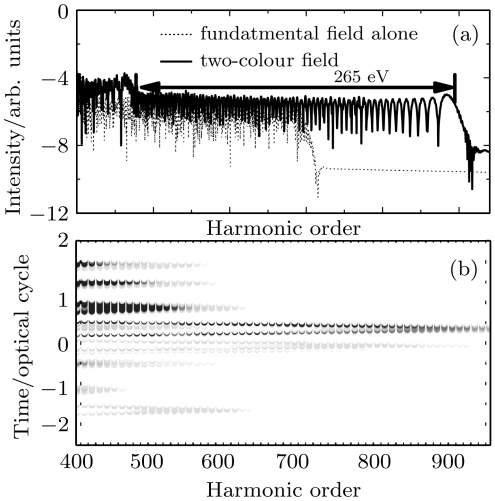


Fig. 3. (a) HHG spectra and (b) their time-frequency distributions in two-colour field.

The result is shown in Fig. 3(b). One can see that the harmonics above the 470th (290 eV) are mainly contributed by the quantum path from $0.0T_0$ to $0.5T_0$, forming a supercontinuum from 290 eV to 555 eV almost covering the whole water window. These results

are in good agreement with those obatined by the classical approaches shown in Fig. 2. In our scheme, the result is sensitive to the delay τ between the two laser pulses. Figure 4 shows the sensitivity of the harmonics spectrum to the variation of the pulse delay. The harmonic spectra with the delay $\tau = -0.6T_0$ and $\tau = -0.4T_0$ are shifted down 6 and 3 units for clarity, respectively. The bandwidth of the supercontinuum decreases for $\tau = -0.6T_0$ and $\tau = -0.4T_0$. Therefore, it is important to control precisely the delay of the two-colour field in order to obtain broad supercontinuum.

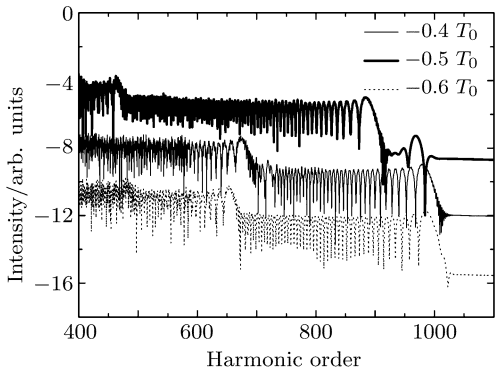


Fig. 4. HHG spectra with different pulse delays.

The sensitivity of the harmonics spectrum to the variation of the electric field E_1 of the control pulse is also investigated. The result is shown in Fig. 5. The harmonic spectra with $E_1 = 0.03$ a.u. (thin dotted line), $E_1 = 0.035$ a.u. (thin dashed line) and $E_1 = 0.04$ a.u. (thin solid line) have been shifted down 9, 6, and 3 units for clarity, respectively. It reveals that the bandwidth of the supercontinuum increases with the increase of electric field E_1 of the control pulse.

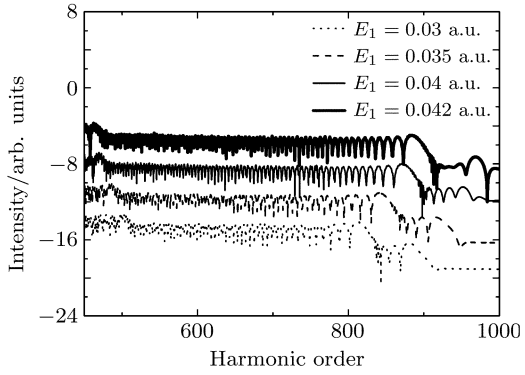


Fig. 5. HHG spectra with different control laser pulse intensities.

We further investigate the attosecond pulse generation by a square window in the multicycle two-colour pulse. Figures 6 and 7 show the temporal profiles of the attosecond pulses generated by selecting the harmonics with different central frequencies. As shown in Fig. 6, by adding a frequency window with a bandwidth of 115 order harmonics to the supercontinuum and making an inverse Fourier transformation, an isolated attosecond pulse with a minor pre-pulse is generated without any phase compensation. The shortest pulse duration shown in Fig. 6 is 62 as by selecting harmonics from 470th to 585th. By adding a frequency window with a bandwidth of 80 order harmonics to the supercontinuum, an isolated attosecond pulse with a minor pre-pulse is also generated without any phase compensation, which is shown in Fig. 7. The duration of the attosecond pulse changes a little with the increase of central frequency, though the intensity decreases. It reveals that the pulse duration is insensitive to the position of the frequency window.

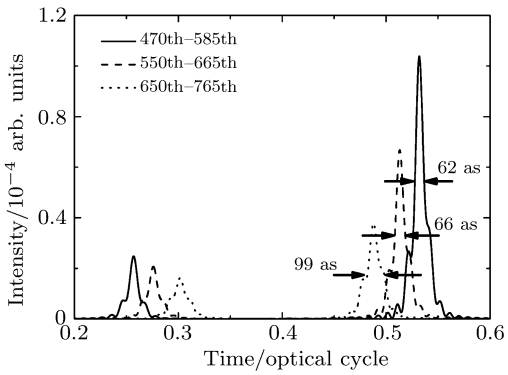


Fig. 6. Temporal profile of the isolated attosecond pulses by superposing different harmonics.

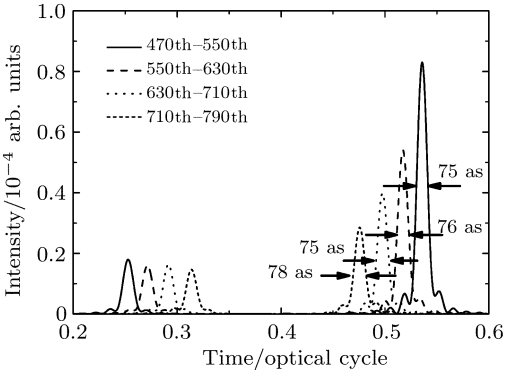


Fig. 7. Temporal profiles of the isolated attosecond pulses by superposing different harmonics.

4. Conclusion

We proposed a method to generate a high-efficiency broadband water window supercontinuum with a $\omega + 3\omega/2$ multicycle two-colour pulse. Our

results reveals that the $3\omega/2$ laser pulse can simultaneously modulate the acceleration step and the ionization step, which not only broadens the bandwidth but also enhances the yield of the generated supercontinuum from 290 eV to 555 eV. The generated supercontinuum almost contains the whole water window, from which an isolated 62-as pulse with a minor pre-pulse can be directly obtained.

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