Generation of an isolated sub-100 attosecond pulse in the water-window spectral region*

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We propose a scheme to generate isolated attosecond pulses in the water-window spectral region. Based on the numerical solutions of the single active electron model, we investigate high-order harmonic generation in helium atoms driven by a multi-cycle two-colour optical field synthesized by an intense 2000 nm, 20 fs pulse and its frequency-doubled pulse. When the latter is slightly detuned and properly phase shifted with respect to the fundamental laser pulse, an ultra-broad extreme ultraviolet supercontinuum with a spectral width of 130 eV can be generated in the 270–400 eV spectral regions. A supercontinuum from 280–340 eV in the water window can be selected to yield an isolated 67 attosecond pulse without employing any phase compensation. This water window coherent x-ray pulse with less than 100 attosecond duration is a potential tool for studying the ultrafast electronic dynamics of biological samples in water.

Keywords: x-ray, ultraviolet spectra, optical harmonic generation
PACC: 9580N, 3320L, 4265K

1. Introduction

Isolated extreme ultraviolet (XUV) pulses of attosecond durations are an important tool for exploring electron dynamics in atoms and molecules.[1−8] The most successful way to generate isolated attosecond pulses has been frequency synthesis of high-order harmonic supercontinuum at the cutoff region and the plateau region driven with laser pulses shorter than two optical cycles.[2] For instance, Peng and Zeng[9] numerically demonstrated that an XUV supercontinuum can be generated in the high harmonic plateau region by coherently synthesizing three different harmonic frequencies with fundamental-wave, second harmonic (SH) and third-harmonic pulses. This supercontinuum is significant for the efficient generation and further control of single attosecond pulses. This approach is limited by the shortest available pulse duration, and it has been difficult to generate pulses with duration less than $\sim$ 100 attosecond until a recent breakthrough.[8] Isolated attosecond pulses have also been generated by employing a polarization gating method,[3] which also requires few-cycle driving laser pulses. Besides, for efficient high-order harmonic generation (HHG) with ultrashort laser pulses, it is important to have a perfect phase matching between the driving laser pulse and the generated harmonic signal. Peng *et al.*[10] reported experimental results along with a theoretical analysis that the phase-matching condition and HHG efficiency can be readily controlled by using an SH laser field.

HHG driven by a two-colour laser field does not require a driving pulse duration of less than two cycles and has been utilized to generate attosecond pulses.[4,11] More recently, calculations have been presented for the generation of isolated attosecond pulses in the multi-cycle two-colour field.[12,13] They have shown that the detuning of the SH can be used to break down the symmetry of the electric field over many optical cycles and provide a coherent control for the formation of an isolated attosecond pulse. The use of multi-cycle driving laser pulses offers higher energy for the generated attosecond XUV pulses.

In the so-called water-window, from 4.4 nm to 2.3 nm (from 280 eV to 550 eV), carbon-containing bi-

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ological samples absorb radiation efficiently but water is comparatively transparent. High-contrast biological spectroscopy in this spectral window has long been one of the most important objectives of the development of coherent XUV sources.[14] High order harmonic emission in the water window with attosecond pulse duration is, therefore, strongly desired for studying the ultrafast electronic dynamics of biological samples in water. However, the wavelengths of currently available isolated attosecond pulses are much longer than 10 nm.[1−8]

The scaling law for the cutoff energy of HHG as a function of driving laser wavelength indicates that a longer wavelength infrared (IR) laser pulse will shorten the wavelength of attosecond pulses, but will do so with reduced conversion efficiency. Recently a scheme for the generation of 2-cycle (15.7-fs), 740-μJ mid-infrared (MIR) pulse at the carrier wavelength of 2.1 μm and the repetition rate of 1 kHz, from a broadband optical parametric chirped pulse amplification (OPCPA) system was proposed.[15] For the detuned frequency-doubled pulse, it can be obtained by using the spectral modulation of an MIR femtosecond laser pulse induced by molecular alignment revivals.[16] In this work, we explore how the two-colour scheme using MIR driving laser pulses can be used to generate an intense isolated attosecond pulse in the water window.

2. Simulation results and discussion

In our calculation, a 20 fs/2000 nm pulse and a detuned frequency-doubled 20 fs/909 nm pulse, both linearly polarized, are synchronized to form the driving field for HHG in helium atoms. The HHG calculation method, which is based on the single-active-electron approximation, has been widely used.[2] The Lewenstein model is used to calculate the dipole moment of an atom in the nonadiabatic formula[17] and the high harmonic spectrum can be obtained by Fourier transformation of the time-dependent dipole moment. The intensities of the fundamental and the frequency-doubled pulses are assumed to be 2.0×10^{14} W/cm^2 and 1.0×10^{14} W/cm^2, respectively. The synthesized laser field is described by

$$E_{\rm s}(t) = E_1 \exp\left[-2\ln(2)t^2/\tau_1^2\right]\cos(\omega_1 t+\phi_1) + E_2 \exp\left[-2\ln(2)t^2/\tau_2^2\right]\cos(\omega_2 t+\phi_2), \quad (1)$$

where E_1 and E_2 are the electric field amplitudes of the fundamental and the frequency-doubled pulses, respectively; ω_1 and ω_2 are the angular frequencies of the two-colour pulses; τ_1 and τ_2 are the corresponding pulse durations (full width half maximum, FWHM); ϕ_1 and ϕ_2 are the corresponding absolute phases.

Figure 1(a) shows the electric field of the fundamental laser pulse. The temporal profile of the harmonic emission driven only by the fundamental field is shown in Fig. 1(b). It is clear that the attosecond pulses appear at almost every 0.5 optical cycle. In Fig. 1(c) we show the synthesized electric field when an intense detuned frequency-doubled field ($\omega_2 = 2.2\omega_1$) is added to the fundamental field. The temporal profile of the harmonic emission driven by two-colour field is presented in Fig. 1(d). The attosecond pulses with the highest intensity (labelled by I and II) are separated by about 3 optical cycles as shown in Fig. 1(d). If such a two-colour field is employed, a laser pulse 6 times longer than the 2 cycle limit could be used to generate isolated attosecond pulses.

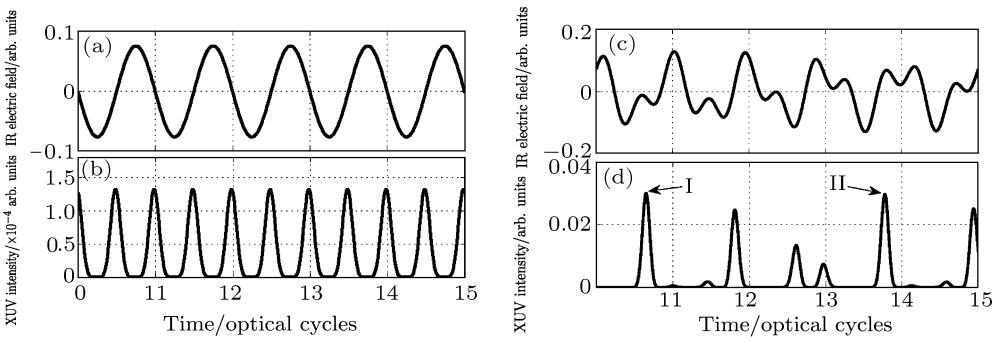


Fig. 1. (a) Electric field of the fundamental laser pulse (2000 nm, ω_1). (b) Temporal profile of the harmonic emission driven only by the fundamental field. (c) Synthesized electric field for the fundamental (2000 nm, ω_1) and its detuned frequency-doubled harmonic (909 nm, $\omega_2 = 2.2\omega_1$). (d) Temporal profile of the harmonic emission driven by the synthesized laser field. The relative phase between the two pulses is -0.3π.

To understand HHG better in the detuned two-colour field, we may observe the harmonic emission in the spectral domain. Significant differences from the single colour case are revealed. The calculated harmonic spectra generated by single colour (ω_1) and the detuned two-colour ($\omega_1 + \omega_2$) fields are plotted in Fig. 2. The most pronounced difference between the two spectra is the significant extension of cutoff energies from 270 eV in the single colour field case to 400 eV in the two-colour case. In addition, the harmonic intensity is increased by about one order of magnitude in the two-colour case. This can be easily understood in the light of the semi-classical three-step model of HHG.[5]

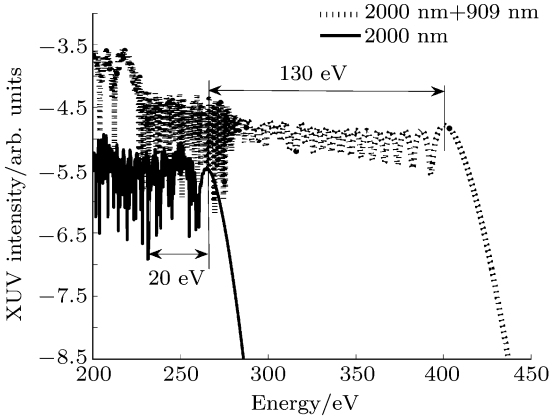


Fig. 2. The harmonic spectra for helium atoms. The solid curve shows the spectrum generated only by the fundamental laser field. The dashed curve shows the spectrum generated by the two-colour field. The relative phase between the two pulses is -0.3π.

To clarify the role of the shaped laser field played in the two-colour scheme, the ionization and the classical dynamics of electrons are calculated. The results are shown in Fig. 3, in which Fig. 3(a) illustrates the dependence of the kinetic energy on the recombination time in the single colour fundamental field and in the two-colour field. It is shown that there are two classes of trajectories corresponding to the same energies of the returning electrons in each half optical cycle for the fundamental field. The first trajectory, with positive chirp, is called short trajectory, characterized by later ionization but earlier emission time; the second trajectory, with negative chirp, is called long trajectory which displays earlier ionization but later recombination time.

In the single colour field case, the recombination of the electron with the parent core will generate harmonic emission with slightly different maximum photon energy between the two adjacent half fundamental laser cycles. Consequently, the harmonic supercontinuum with a bandwidth of only 20 eV at the cutoff can be spectrally selected for an isolated attosecond pulse. After the detuned frequency-doubled pulse was added to the fundamental laser pulse, the returning electron energy at $t = 16.2T_0$ (T_0 is the optical cycle of the fundamental field) is enhanced greatly from 250 to 380 eV, which coincides well with the significant broadening of the XUV supercontinuum as shown in Fig. 2 (dashed curve).

It can be seen from Figs. 2 and 3 that the extension of the cutoff is consistent with the additional kinetic energy of the returning electron wave packets provided by the detuned frequency-doubled field. The XUV supercontinuum of 130 eV bandwidth is generated when the relative phase between the two pulses is -0.3π. In Fig. 2 we have noticed that in the dramatically extended plateau region, the harmonic spectrum shows less intensity modulation than that in the single colour case where a very large modulation exists inherently.[18] This is the region where a spectral filter can be applied to isolate the attosecond pulse.

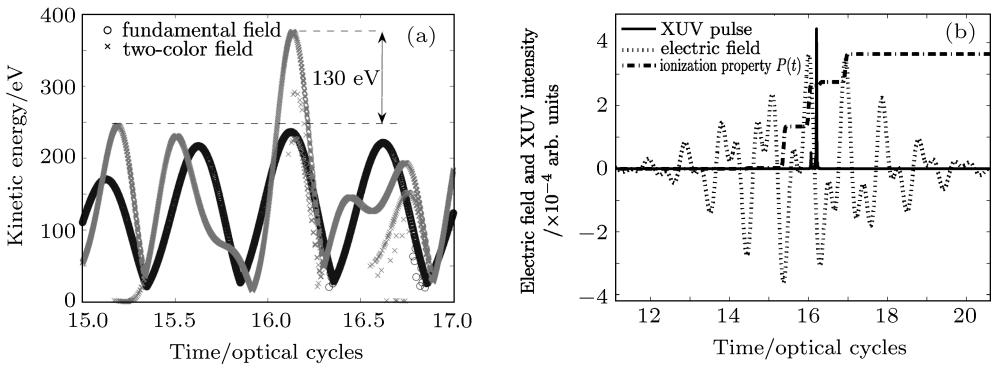


Fig. 3. (a) The kinetic energies of a returning electron in the fundamental field (◯) and in the two-colour field (×) as functions of the recombination time. (b) The electric field of the two-colour field (dotted curve), the ionization probability (dot-dashed curve) and the harmonic emission (solid curve) selected with a numerical spectral filter ranging from 280 to 340 eV, as functions of time in the two-colour field.

Figure 3(b) shows the electric field of the two-colour field (dotted curve), the ionization probability (dashed-dotted curve) and the high-order harmonic emission (solid curve) selected with a numerical spectral filter between 280 eV and 340 eV[19] as functions of time. One can clearly see that the ionization probability increases rapidly near the recombination time $t = 15.3T_0$ but keeps constant after $t = 17T_0$. The ionization rate at $t = 16.2T_0$ is relatively high, leading to the efficient broadband harmonic emission as shown in Fig. 2. This is consistent with the observation that the maximum energy of the returning electron appears at $t = 16.2T_0$, as shown in Fig. 3(a).

It should be noticed in Fig. 3(a) that the kinetic energy profile consists of several regular broad peaks separated by about half an optical period (T_0) in the single colour case. But the kinetic energy profile becomes irregular with a predominant peak at $t = 16.2T_0$ in the two-colour case. This change indicates that the electron trajectory has been changed after the detuned frequency-doubled field is added. Depending upon the instantaneous phase relationship between the fundamental and detuned frequency-doubled fields, the electrons may be accelerated and decelerated many times. The electron path is correspondingly stretched or shortened, which induces the returning time of electrons is earlier or later than that in the fundamental field case. Under certain conditions, only one quantum path gains the highest increase in kinetic energy while the others do not have any apparent change. In addition, the FWHM of the kinetic energy profile around $t = 16.2T_0$ is narrower in the two-colour case than that of the fundamental field case, indicating that the average time interval between emissions around $t = 16.2T_0$ decreases and, thus supports a single attosecond pulse with shorter duration.

In the time domain, an XUV supercontinuum from 280 to 340 eV (60 eV bandwidth), without employing phase compensation, can generate an isolated 67 as pulse shown in Fig. 4 (the solid curve). If the dispersion is well compensated for over the 60 eV spectral widths, a single 60 as pulse with a clean time profile can be obtained (the dashed curve in Fig. 4). For comparison, we also calculated the attosecond pulses generated in the single-colour fundamental laser field by taking the inverse Fourier transform of the 20 eV wide XUV supercontinuum in the cutoff region without employing phase compensation (Fig. 4, dot-dashed curve). Although a main attosecond pulse (peak I) is produced, two strong satellite pulses, peak II and peak III, also appeared.

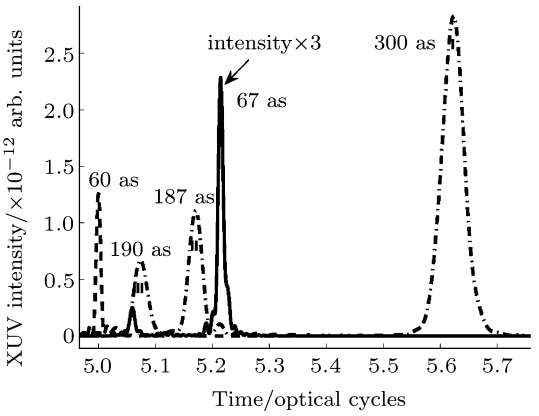


Fig. 4. The temporal profiles of the attosecond pulses with 60 eV bandwidth, obtained with (dashed) and without (solid) phase compensation. For comparison, the attosecond pulses generated by the single-colour 20 fs pulse without phase compensation is also shown (dot-dashed curve).

To show the importance of frequency detuning for the SH pulse and the relative phase shift between the two-colour pulses, we performed a time–frequency analysis of the dipole response of the He atom to the four different driving laser fields.[20,21] If the time window around a specific moment is applied to the dipole acceleration, the spectrum can represent the strength of the dominant frequencies at that moment. By calculating the spectra with a translation of the time window, we obtain the time–frequency distribution of the XUV radiation.[22] It can be seen in Fig. 5(a) that for the HHG produced by the single 20 fs pulse, there is only a minor difference in the XUV emission peaks marked by B, C, and D. The bandwidth of the supercontinuum at the cutoff region is very narrow. When the frequency-doubled pulse is added to the fundamental pulse with relative phase $\phi = -0.3\pi$, we can see from Fig. 5(b) that the photon energy of the two peaks labelled with A and C (separated by one optical cycle in time domain) are enhanced significantly. This illustrates that the symmetry of the electric field is already broken in the two-colour field. When the detuned frequency-doubled pulse is added upon the fundamental pulse with zero phase shift, only the maximum photon energy and intensity of peak D in Fig. 5(c) are greatly increased, while those of peak A, B and C are decreased. When a relative phase shift of the detuned frequency-doubled pulse of $\phi = -0.3\pi$ is employed, the intensities of peaks A, B and D are greatly suppressed, while the intensity of peak C is

further enhanced as shown in Fig. 5(d), indicating the electric field symmetry has been broken down further in a larger time window. From Figs. 5(a) to 5(d), we have shown the progress of isolating one attosecond emission by controlling the quantum path of a returning electron.

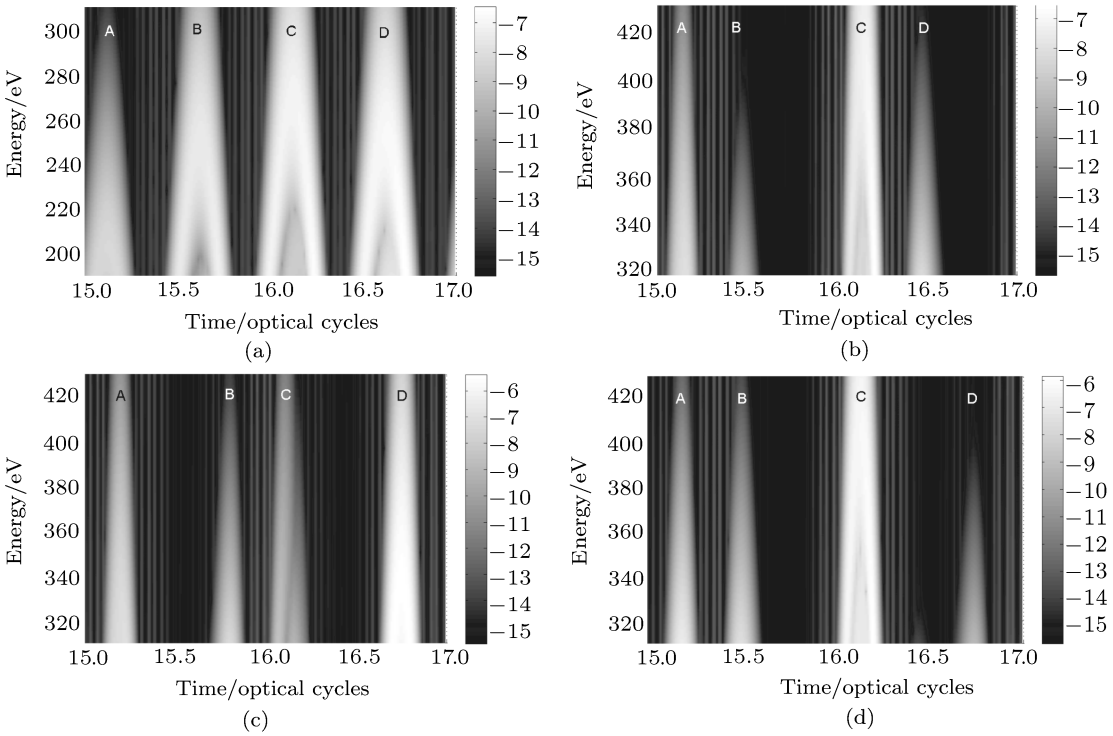


Fig. 5. Patterns of the time–frequency distributions of high harmonic emission generated by (a) the single 20 fs pulse, (b) the combination of a 20 fs, 2000 nm fundamental pulse and a frequency-doubled pulse with a relative phase of -0.3π, (c) the combination of the fundamental pulse and the detuned frequency-doubled pulse with zero phase shift, and (d) with a relative phase shift of -0.3π. Note that the colour scale is in logarithmic units.

Finally, it should be mentioned that this two-colour scheme requires the IR driving pulses to have carrier-envelope phase (CEP) stabilization in order to produce isolated attosecond pulses.

3. Summary

In summary, HHG in helium atoms driven by two-colour IR field has been investigated using the Lewenstein model. We have demonstrated the generation of isolated 67 attosecond pulses in the water window, when an appropriate phase shift between two driving pulses is employed. In addition, this two-colour scheme can enhance the XUV emission intensity by one order of magnitude, which is important for HHG in an IR field. This proposed scheme is promising for the development of intense isolated attosecond pulse sources useful in the analysis of biological samples in water.

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