Attosecond manipulation of ionization and efficient broadband supercontinuum generation in stretched molecules*

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We theoretically investigate the high-order harmonic generation from stretched molecules in a linearly polarized intense field. By adopting an infrared pulse combined with an ultraviolet (UV) attosecond pulse, the ionization process can be controlled effectively. In this excitation scheme, the harmonic spectrum beyond $I_p + 3.17U_p$ is significantly enhanced by two orders, where I_p and $U_p = e^2E_0^2/(4m_e\omega^2)$ are the ionization and ponderomotive potential, then smooth broadband supercontinuum with the bandwidth of about 120 eV is obtained, which leads to an isolated sub-60-as attosecond pulse with a high signal-noise ratio. Moreover, the bandwidth of the supercontinuum is weakly dependent on the location and pulse duration of the UV pulse.

Keywords: attosecond manipulation, attosecond pulse, supercontinuum generation

PACC: 3280F, 3300, 3365, 3365C

1. Introduction

The generation of extreme ultraviolet (UV) attosecond pulses has opened new fields of time-resolved studies with high precision.[1] The attosecond pulses are an important tool for detecting and controlling the electronic dynamics inside atoms and molecules, such as inner-shell electronic relaxation and ionization by optical tunneling.[2] High harmonic generation (HHG) is a subject of great interest due to its potential applications in the generation of attosecond pulses.[3−5] It was demonstrated by Paul *et al.* that a train of attosecond pulses were produced with a multicycle laser pulse.[6] However, straightforward metrology prefers an isolated attosecond pulse[7] and then much effort has been paid to extract an isolated pulse.[8−10] The effort opens up new possibilities for investigating and controlling electron dynamics in atoms and molecules in attosecond time scale.[11] The HHG process can be well-explained by the three-step model:[12,13] the electron first tunnels through the barrier formed by the Coulomb potential and the laser field, then it oscillates almost freely in the laser field, and finally, it may return to the ground state by recombining with the parent ion in an atom.

By manipulating the different steps of this dynamical process, one can control HHG in principle. Thus most effort focuses on several schemes for HHG control.[3,14−16] One of the schemes has demonstrated that the return probability of the electron to the parent ion has been manipulated by using the polarization gating technique.[14,15] An alternative method to control HHG is using an additional field, then the trajectory of the electron in the continuum can be efficiently altered.[3,16] The effect of those methods on the ionization process is of secondary importance. A different scheme has been investigated to control HHG by manipulating the ionization of the electron.[5,11,17] It was demonstrated by Lan *et al.*[5] that one can trigger the ionization process in atoms and the harmonic spectrum is varied evidently by using an intense attosecond UV pulse, which leads to an isolated 130-as pulses. However, the critical problems are that the HHG control is very sensitive to the delay between the attosecond UV pulse and fundamental pulse, and the bandwidth for isolated attosecond pulse should be further broadened.

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In recent years, the theoretical and experimental researches on HHG control from molecules have also been of great interest.[18−21] It was found that HHG from molecules shows similar spectral pattern to that from atoms. However, since the molecules have more degrees of freedom than atoms, their physical phenomena in strong fields are much richer. Thus high order harmonics can be generated in molecules. The theoretical investigation shows that the harmonic spectrum can be extended to $I_{\rm p}+8U_{\rm p}$ from molecules with a specific internuclear distance,[21,24] which can be obtained by dissociating the molecule.[25,26] This HHG process can be well explained by the three-step model: the molecule has more than one nuclei, the electron may be detached from one nucleus, then be accelerated by the laser field towards another nucleus. Moreover, it has been investigated that the emission times of harmonics with energy beyond $I_{\rm p}+3.17U_{\rm p}$ are nearly the same, i.e., well phase-locked.[20] Taking into account those effects, the stretched molecules are attractive candidates for broadband XUV supercontinuum and shorter isolated attosecond pulses. However, due to the low ionization, the intensity of the harmonic spectrum with energy beyond $I_{\rm p}+3.17U_{\rm p}$ is very low.

These works[18−21] only considered the single-molecule response. For the real scenario, a full description of HHG requires the propagation of the driving field and high harmonics through the gas medium at the macroscopic level. Propagation affects the time-frequency structure of the harmonics due to different phase-matching conditions for different quantum paths.[22] However, for the stretched molecules, the harmonics from two different paths are synchronously emitted.[20] Then the time-frequency structures of the harmonics in the second plateau will hardly change under different phase-matching conditions.[23] In this paper, we investigate manipulation of the ionization process from stretched molecules by adding a UV attosecond pulse to an infrared few-cycle pulse, which effectively controls HHG and generates an efficient broadband XUV supercontinuum. It is shown that the harmonic spectrum beyond $I_{\rm p}+3.17U_{\rm p}$ is significantly enhanced by two orders and a sub-60-as isolated attosecond pulse with high signal-noise ratio is efficiently obtained. Moreover, the bandwidth of the supercontinuum is weakly dependent on the duration of the UV pulse and the delay between the attosecond UV pulse and fundamental pulse.

2. Formulation

In this paper, we investigate the interaction of a diatomic molecule with a linearly polarized intense laser pulse by solving the one-dimensional time-dependent Schrödinger equation (TDSE) numerically. This equation describing the motion of the electron in the presence of the combined field is expressed as (atomic units are used throughout this paper)

$$\mathrm{i}\frac{\partial\psi(x,t)}{\partial t}=\left[-\frac{1}{2}\frac{\partial^2}{\partial^2 x}+V(x,R)-E(t)x\right]\psi(x,t), \quad (1)$$

$$V(x,t)=-\frac{1}{\sqrt{0.5+(x-R/2)^2}}-\frac{1}{\sqrt{0.5+(x+R/2)^2}}, \quad (2)$$

in Eq. (1), $V(x,R)$ is the model potential of the molecule, and we will consider the soft Coulomb potential as Eq. (2), $E(t)$ is the electric field. R in Eq. (2) is the internuclear distance, and the two nuclei are fixed at $\pm R/2$. The electric field $E(t)=E_{\rm ir}+E_{\rm UV}$ is a combination of an infrared ($E_{\rm ir}$) and UV ($E_{\rm UV}$) light. The two components in $\hat{X}$ direction are given by

$$E_i(t)=E_i f_i(t-\tau_i)\cos[\omega_i(t-\tau_i)], \quad (3)$$

where i=ir and UV. E_i and ω_i are the amplitudes and the frequencies, and $f_i(t)$ and τ_i are envelopes and peak positions of the pulses respectively. A Gaussian envelope shape is adopted and the delay between the UV and infrared pulses is $\tau_{\rm delay}=\tau_{\rm UV}-\tau_{\rm ir}$.

The time-dependent Schrödinger equation can be solved with the Crank–Nicolson method,[27] starting from the field-free ground state which is obtained by propagation in imaginary time, and finally the wave function $\psi(x,t)$ can be obtained. The space range is from –500 to 500 a.u. To prevent the reflection of the wave function from boundary, the wave function is multiplied by a $\cos^{1/8}$ mask function that varies from 1 to 0 over a width of 250 a.u. at the outer boundary. The spatial and temporal steps are 0.1 and 0.08 a.u., respectively. According to the Ehrenfest theorem,[28] the time-dependent dipole acceleration $a(t)$ is

$$a(t)=-\langle\psi(x,t)|\frac{\partial V(x,R)}{\partial x}-E(t)\,|\psi(x,t)\rangle\,. \quad (4)$$

The harmonic spectrum can be obtained from the Fourier transform of $a(t)$

$$S_q(\omega)\sim|a_q|^2\,, \quad (5)$$

$$a_q=\int a(t)\mathrm{e}^{-\mathrm{i}\,q\omega_0 t}\mathrm{d}t, \quad (6)$$

where q is the harmonic order. By superposing several harmonics, an ultrashort pulse is generated and the temporal shape of the pulse is

$$I(t) = \left| \sum_q a_q \mathrm{e}^{\mathrm{i}\, q\omega_0 t} \right|^2 . \tag{7}$$

3. Result and discussion

We first investigate the HHG of the stretched molecule exposed to the 5-fs infrared pulse alone. The intensity is 5×10^{14} W·cm^{-2} and the central wavelength is 800 nm. It has been demonstrated that the photon energy at the cutoff reaches $8U_\mathrm{p} + I_\mathrm{p}$ when internuclear distance $R = \pi E_0/\omega^2$,[21,24,25] then R is set to 115.3 a.u. The curve in Fig. 1 shows the harmonic spectrum of the stretched molecule in the infrared laser pulse. The harmonic spectrum has two cutoff marked by the arrows. The first cutoff is at the 77-th order harmonic, corresponding to the energy $I_\mathrm{p} + 3.17U_\mathrm{p}$, and the second one is at the 170-th, corresponding to the energy $I_\mathrm{p} + 8U_\mathrm{p}$. According to the classical discussion in Ref. [20], the first plateau is mainly due to the electrons which ionize from one nuclei and recombine with their parent nuclei, here we call it the first approach. The second plateau is mainly contributed by the electrons ionized from one nuclei and recombine with the neighbouring nuclei, here we call it the second approach. As shown in Fig. 1, the intensity of the second plateau is very weak, which limits the application of the harmonic spectrum.

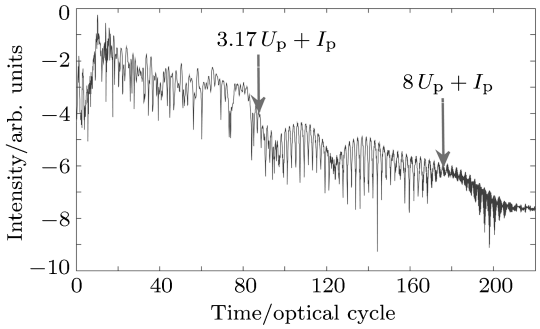


Fig. 1. The harmonic spectrum from diatomic molecules with large internuclear distance exposed to a 5-fs 800-nm infrared pulse alone.

In order to understand the underlying physics of the HHG with two-plateau structure in Fig. 1, we discuss the HHG process in stretched molecules using the semiclassical three-step model and the Ammosov–Delone–Krainov (ADK) model.[29] In this scheme, we can assume that the effect of the neighbouring potential can be neglected because internuclear distance of diatomic molecules is large enough.[29] Figure 2 illustrates the electron dynamics of HHG process in the infrared laser field. In our simulation, the parameters are the same as those in Fig. 1. Figure 2(a) shows the dependence of the kinetic energy E_k on the ionization and recombination time. The ionization times for the first and second approaches are shown by thin solid curve and thin dots, and the return times for the first and second approaches are presented by thick solid gray curve and thick black dots, respectively. As shown in Fig. 2(a), it is seen that there are two classes of trajectories corresponding to the same energy for the second approach in each half optical cycle. The one with earlier ionization time is called long trajectory, and the another one with later ionisation time is called short trajectory. The interference of the long and the short trajectories will lead to a coherent structure of the supercontinuum. From Fig. 2(a), the highest return energy can reach $8U_\mathrm{p}$, while the second and third highest return energies are about $7U_\mathrm{p}$. This result indicates that the bandwidth of the supercontinuum is only U_p. According to the three-step model, when the intensity of the laser pulse is much lower than the saturation intensity of the model atom, the harmonic efficiency is mainly determined by the first step, i.e., the ionization rate. Figure 2(b) shows the tunnel ionization rate calculated in terms of the ADK model (gray filled curve). As shown in Fig. 2(b), the ionization rates of the electrons corresponding to the highest peak are very low, as the rectangle marks.

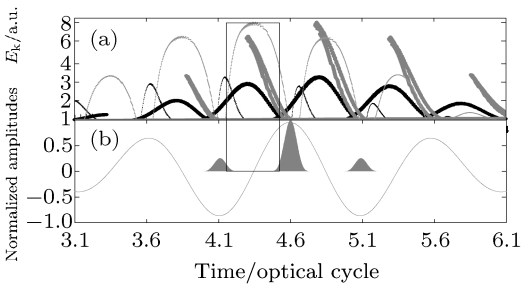


Fig. 2. (a) The dependence of the kinetic energy E_k on the ionization (thin solid curve and thin dashed curve) and recombination (thick solid curve and thick dashed curve) times, and (b) the tunnel ionization rate calculated by the ADK model (gray filled curve). The parameters are the same as those in Fig. 1.

Thus the intensity of the second plateau is two orders lower than that of the first one. In order to enhance the harmonic efficiency, we can manipulate ionization

process by adding a UV attosecond pulse to an infrared few-cycle pulse at the time marked by the black rectangle.

Next, we investigate the HHG of the stretched molecule exposed to the laser field synthesized by the infrared few-cycle pulse and the UV attosecond pulse. For the UV attosecond pulse, the intensity is 3×10^{13} W·cm^{-2}, the central wavelength is 100 nm, and the pulse duration is 0.6 fs full width at half maximum (FWHM). The parameters of the infrared pulse are the same as those of Fig. 1. Figure 3 shows the harmonic spectrum of the stretched molecule in the synthesized laser pulse (solid curve). For comparison, the harmonic spectrum in the infrared pulse alone is also presented (dashed curve). It is seen that the position of the second cutoff in the synthesized field is the same as that in the one-colour field. But the intensity of the second plateau is about two orders higher than that of the one-colour case, then a broadband XUV supercontinuum of the second plateau is obtained. Moreover, in Fig. 3, the second plateau of the harmonic spectrum in the one-colour case is modulated evidently. This is easily understood in Fig. 2: the interference between the trajectories in the adjacent half optical cycles leads to the modulation of the harmonic spectrum. By adding the UV attosecond pulse to the infrared few-cycle pulse at the time marked in Fig. 2(a), the ionization process can be manipulated and the ionization rates of the electrons corresponding to the highest energy are much higher than those of other electrons for the second approach. Then only the trajectories corresponding to the highest energy are selected to effectively contribute to the harmonics. Thus the interference will be weak and the harmonic spectrum becomes more regular and smooth.

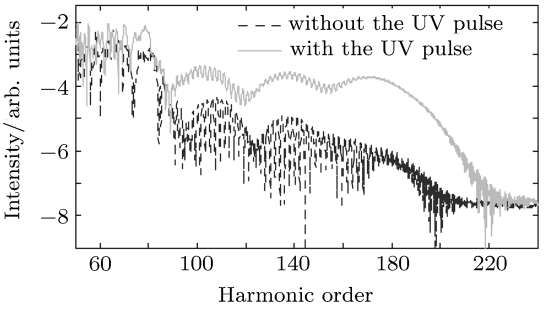


Fig. 3. The harmonic spectrum of the stretched molecule in the synthesized laser pulse (solid curve) and in the infrared pulse alone (dashed curve).

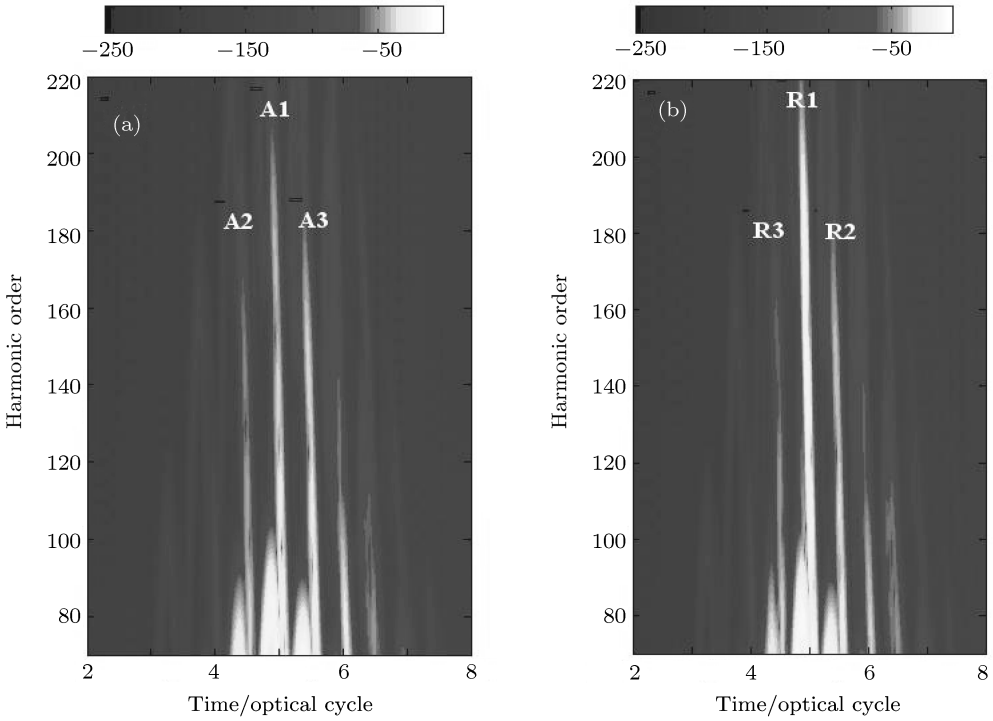


Fig. 4. The time-frequency analysis of the stretched molecule in (a) in the infrared pulse alone and (b) the synthesized laser pulse. The parameters are the same as those in Fig. 3.

The time-frequency analysis in the synthesized field and the infrared field alone are shown in Fig. 4, to gain a deeper insight into the HHG process. The parameters are the same as those of Fig. 3. As shown in Fig. 4(b), the intensity of the right arm marked as R1 is much higher than that of R2 and R3. Contrarily, the intensity of the arm A1 is a little higher than that of the arm A3 in Fig. 4(a), leading to the modulation of the harmonic spectrum in the infrared field alone. The results are consistent with those of Fig. 3.

Next, we investigate the isolated attosecond pulse generation in the synthesized field of the infrared field and the UV field. The temporal profile of the attosecond pulse by superposing 140–190-th harmonics is shown in Fig. 5. The bandwidth of the harmonics is 78 eV, which corresponds to an isolated 52-as pulse in the Fourier-transform limit. It is seen that the isolated pulse of 55-as with high signal-noise ratio is obtained.

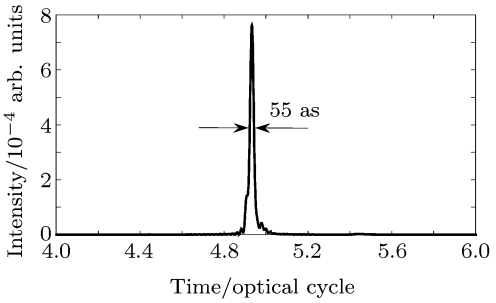


Fig. 5. Temporal profile of the isolated pulse in the synthesized laser pulse. The harmonics from 140-th to 190-th are selected. Other parameters are the same as those in Fig. 4(b).

As shown above, when the UV attosecond pulse is added to the infrared pulse, the HHG can be manipulated effectively and a sub-60-as attosecond pulse is obtained. Figure 6 shows the harmonic spectrum and the isolated attosecond pulses in the synthesized field with different durations of the UV attosecond pulse. The durations of UV pulses are set to 0.5 fs, 0.8 fs and 1 fs (FWHM), respectively, and here the harmonics of 140–190-th are selected. The other parameters are the same as those in Fig. 3. In Figs. 6(a)–6(c), it is seen that the modulation of the second plateau in the harmonic spectrum becomes strong when the duration of the UV pulse increases from 0.5 fs to 1 fs. However, the durations of the attosecond pulses in Figs. 6(d)–(f) are almost 55 as, and the maximum energies of the satellite pulses are only about 0.29%, 0.76% and 2.3% of the main pulses for the three cases, respectively. The results show that the bandwidth of the supercontinuum is weakly dependent on the pulse duration of the UV pulse. This is easily explained in Fig. 2, for the half optical cycle marked by the rectangle, the emission times of HHG corresponding to the short and long trajectories for the second approach are nearly equal.

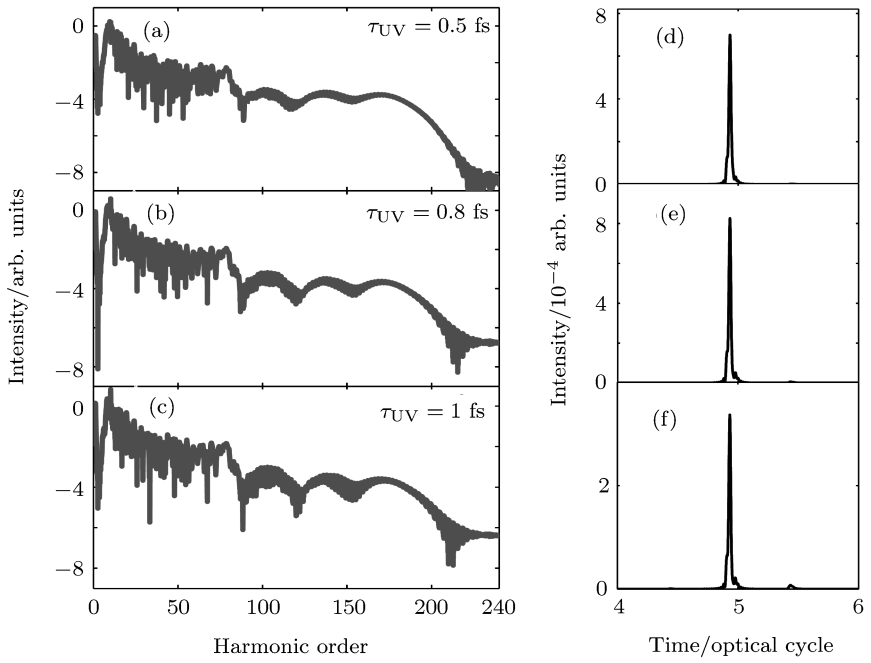


Fig. 6. The harmonic spectra in the synthesized laser pulse with different durations of the UV attosecond pulse [(a)–(c)]. The durations of the UV pulses in (a), (b) and (c) are set to 0.5 fs, 0.8 fs and 1 fs, respectively. The (d)–(f) show the temporal profile of the isolated pulse corresponding to the harmonic spectra in (a)–(c) respectively.

Both of the two trajectories can be selected to enhance the harmonic spectrum and broaden the XUV supercontinuum. Thus the duration of the UV attosecond pulse can reach approximate 1 fs, which equals the width of the rectangle. Moreover, an isolated sub-60-as attosecond pulse with high signal-noise ratio can be obtained.

Finally, we investigate the supercontinuum in the synthesized field with different delays between the attosecond UV pulse and the fundamental pulse. Figure 7(a) shows the dependence of the electron kinetic energy on the ionization time (circles) and return times (crosses) in the 4-th half optical cycle as well as the profile of UV pulse with different delays (solid curve for -0.7π, dots for -0.5π and dashed curve for -0.3π) on the ionization times. The other parameters are the same as those in Fig. 2. Figure 7(b) shows the harmonic spectra in the synthesized field with three different delays. As shown in Fig. 7, the ionization process can be triggered at different time by manipulating delays between the attosecond UV pulse and the fundamental pulse. However, the smooth supercontinuum with the bandwidth of about 120 eV is obtained in the three cases. The results show that the bandwidth of the supercontinuum is weakly dependent on the variation of the delay between the attosecond UV pulse and the fundamental pulse, which is very beneficial for the experiments.

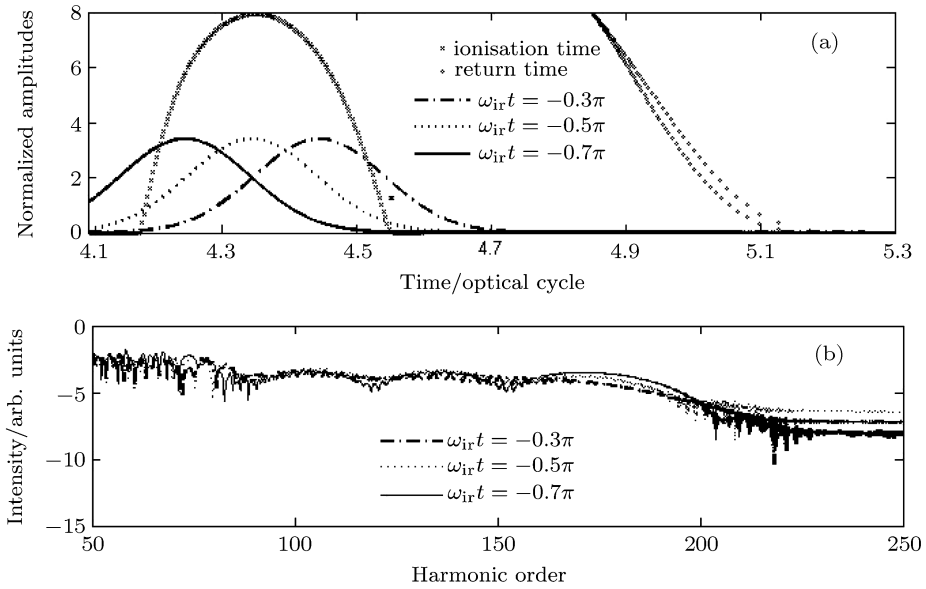


Fig. 7. (a) The dependence of the electron kinetic energy on the ionization time (circles) and return times (crosses) in the 4-th half optical cycle as well as the profile of UV pulse with different delays (solid curve for -0.7π, dotted for -0.5π and dashed curve for -0.3π) on the ionization times. The other parameters are the same as those in Fig. 2; (b) the harmonic spectra in the synthesized field with the three different delays.

4. Conclusion

In conclusion, a method to generate efficient broadband XUV supercontinuum from stretched molecules in a linearly polarized intense field is investigated.[30] By adopting an infrared pulse combined with the UV attosecond pulse, the ionization process can be effectively controlled. In this excitation scheme, the harmonic spectrum beyond $I_p + 3.17U_p$ is significantly enhanced by two orders, where I_p and U_p are the ionization and ponderomotive potential, and smooth supercontinuum with the bandwidth of about 120 eV is obtained, which leads to an isolated sub-60-as attosecond pulse with high signal-noise ratio. Moreover, the bandwidth of the supercontinuum is weakly dependent on the location and pulse duration of the UV pulse.

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