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High intensity supercontinuum in the plateau region of high-order harmonic generated by a phase delayed two-color laser field

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ABSTRACT

We theoretically demonstrate that an intense supercontinuum can be generated in the high-order harmonic plateau region by using a two-color laser field with appropriate electric intensity and phase delay. By using the semi-classic three-step model and the method of time–frequency analysis, it is proved that both the electric intensity and phase delay of the two-color field are important for the effective generation of smooth supercontinuum in the plateau region. As a case study, we show that an isolated 41 as pulse without phase compensation can be generated directly from the contribution of plateau region alone. These results are benefit for the final obtainment of single attosecond pulse with high-energy conversion efficiency.

1. Introduction

High-order harmonic (HH) generation has been studied extensively during the past years as tunable coherent light sources in the extreme ultraviolet region, which allows wide applications in the fields of ultrafast and ultraviolet spectroscopy, holography and photobiology [1,2]. As one of attosecond light sources, single attosecond pulse could be obtained by filtering out coherent ultraviolet supercontinuum around the cutoff region of the HH spectrum. However, the energy conversion efficiency is nevertheless quite low [3]. Since the spectrum in the plateau region are much stable and intense as compared with those in the cutoff region, and the trajectory choose, including the phase-locking has been realized by many different methods [4,5], it is possible to obtain an isolated intense single attosecond pulse in the time domain if less modulated or even supercontinuum can be generated in the frequency domain of plateau region [6]. Now, there are many well-known methods for the generation of continuous emission over the whole harmonic spectrum, such as polarization gating (PG) [7–10], ionization gating (IG) [11,12], and two-color gating (TCG) [13,14]. For the PG methods, the delay induced by the wave plates between the two circularly polarized pulses could reduce the conversion efficiency of the process; for the IG methods, the high intensity and CEP stabilization of few-cycle laser field are key points. As compared to them, TCG is easy to operate and realize. From the analysis of the semi-classical three-step model, we have known that the recombination of the freed electron with its parent ion steered by the driving fields that leads to the emission of HHs. Obviously, the ionization itself and evolution of the ionization-induced electrons, as well as the final recombination of the electrons with the parent ion, all can be controlled well by manipulating the driving fields. For example, it was demonstrated in experiments and calculations that a two-color laser field could significantly modulate the three-step ionization process [15], the long and short electronic trajectories [16], and eventually the energy conversion efficiency and coherence of the emitted HHs [17,18].

Therefore, in this work, we use the TCG methods and demonstrate that an intense supercontinuum can be generated in the plateau region by adding intense second harmonic (SH) pulses to the fundamental-wave (FW) pulses with appropriate relative phase delay. By choosing appropriate electric intensity and phase delay of these driving pulses, the electronic trajectory and recombination time can be controlled effectively and then limit some HHs are only or mainly emitted in a certain half-cycle (in units of FW optical period and used throughout). This finally induces the generation of high intensity supercontinuum in the plateau region (SCP) and then the single attosecond laser pulse with clean temporal profile.

2. Theory

In the following, we first consider the effect of external electric field on the electron movement, in order to obtain an intuitive picture of the control process. Fig. 1 presents the waveforms of the

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FW pulse, the SH pulse and the synthesized FW–SH pulse with a phase delay. We can see the differences between them, mainly are the oscillation frequency and symmetry. For the single color field (FW or SH), the oscillation of the waveform between the positive and negative half cycles is symmetric, and then ionized electron under the driving of laser field will accelerate deviate from the parent ion firstly and then decelerate when the electric field reverses its direction. When the electronic velocity decreases to zero, the direction of electronic movement begins to reverse, i.e., accelerates award the parent ion. During the whole process, electrons that can finally recombine with the parent ion experience the direction reverse only one time. While for the two-color laser field with a phase delay, the oscillation is irregular and quick, i.e., the direction of electric field changes many times, which may drive the electrons to be accelerated and decelerated many times at any possible time. Finally, the recombine time, kinetic energy, electronic trajectory and then emitted harmonic energy all will be changed. Based on this analysis, we think that if we can design a proper waveform of the two-color laser field, some of the ionized electrons may recombine with their parent ions at the same time. As a result, the harmonics can be emitted at the same time and then superposed coherently, which may promote the formation of SCP with high energy conversion efficiency.

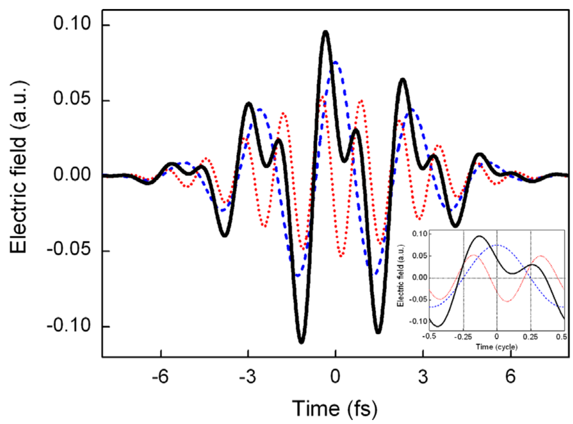

Fig. 1. The FW field (blue dashed line), SH field (red dotted line) and the synthesized FW–SH two-color field with the relative phase delay of 0.7π (black solid line). All these pulses are all 6 fs in pulse duration. The inset is the magnified view of these three waveforms in one cycle. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Furthermore, consider from the equation of the semi-classical three-step model of the HH generation, we may find the exact parameters that can be used to control the electronic motion effectively. The ionized electron is assumed to move along the classical trajectory $x(t)$ driven by the laser field with zero initial velocity $v(t_0)=0$ at time t_0 and position $x(t_0)$. In a single-color laser field $E(t)=E_\omega \cos(\omega t)$, the ionized electron returns to the parent ion at a recombination time t determined by $x(t)-x(t_0)=-(E_\omega/\omega^2)[\cos(\omega t)-\cos(\omega t_0)+\omega(t-t_0)\sin(\omega t_0)]=0$, with the corresponding kinetic energy $E_k=U_p[\sin(\omega t)-\sin(\omega t_0)]^2$, where $U_p=(E_\omega{}^2/4\omega^2)$ is the ponderomotive potential of the single-color laser filed. While in a two-color laser field $E(t)=E_\omega \cos(\omega t)+E_{2\omega}\cos(2\omega t+\Delta\varphi)$, the free-evolution time $(t-t_0)$ and the corresponding kinetic energy at the recombination time of the ionized electron can be written as

$$\begin{aligned} x(t)-x(t_0)=&-\frac{E_\omega}{\omega^2}[\cos(\omega t)-\cos(\omega t_0)+\omega(t-t_0)\sin(\omega t_0)]\\ &-\frac{E_{2\omega}}{4\omega^2}[\cos(2\omega t+\Delta\phi)-\cos(2\omega t_0+\Delta\phi)\\ &+2\omega(t-t_0)\sin(2\omega t_0+\Delta\phi)]=0, \end{aligned} \tag{1}$$

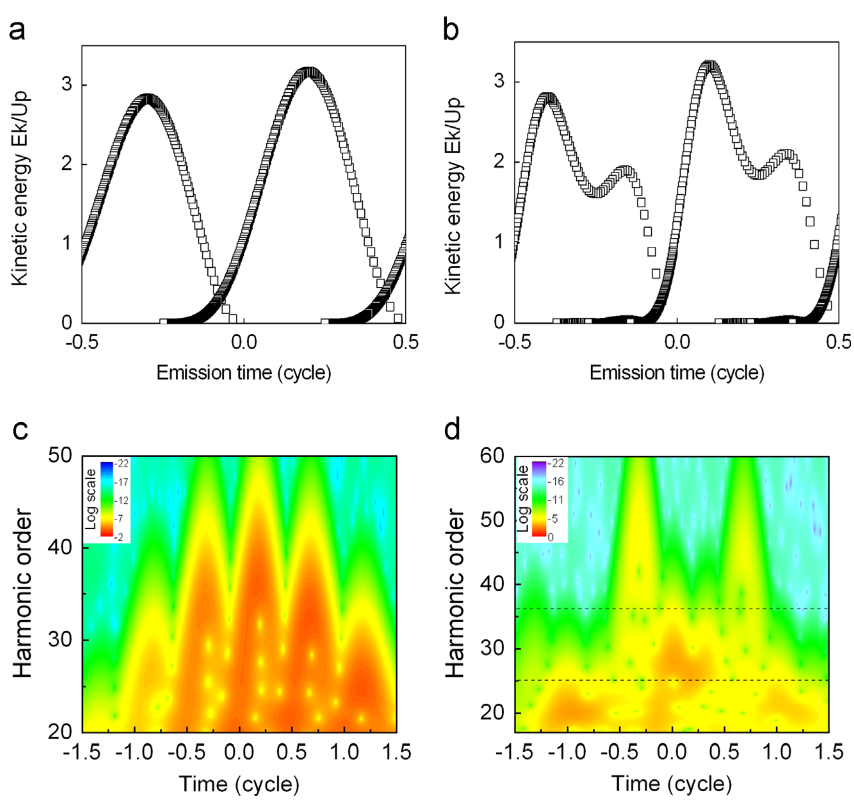

Fig. 2. (a) and (b) Electronic return kinetic energy dependent on the emission time, and (c) and (d) time–frequency characteristics driven by FW [(a), (c)], or synthesized FW–SH two-color field [(b), (d)]. The intensity of the FW pulse in the single-color field is 2.06×10^{14} w/cm^2, and the intensities of the FW and SH pulses in the two-color field are 2.0×10^{14} w/cm^2 and 1.0×10^{14} w/cm^2, respectively. The other parameters are the same as those in Fig. 1. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

$$\frac{E_k}{U_p}=\frac{2}{1+(E_{2\omega}/2E_\omega)^2}\left\{\sin(\omega t)-\sin(\omega t_0)+\frac{E_{2\omega}}{2E_\omega}[\sin(2\omega t+\Delta\phi)-\sin(2\omega t_0+\Delta\phi)]\right\}^2, \quad (2)$$

where the ponderomotive potential of the two-color laser field is defined as $U_p=(E_\omega{}^2/4\omega^2)+(E_{2\omega}{}^2/16\omega^2)$. This simplified calculation predicts that the electronic trajectory, recombine time and the recombine kinetic energy all can be controlled by varying the electric intensity, phase delay and field frequency of the two-color laser field.

3. Results and discussion

Based on the theoretical analysis above, we firstly use the method of time–frequency transform [19,20] to find the proper waveform oscillation, which can generate the SCP effectively [21]. The model atom considered here is argon (Ar), and the so-called single-electron approximation is used for the response calculation of atom in the intense field, which recurs most of the basic aspects of experiments well. The simulation is performed by the numerical integration of the time-dependent Schrödinger equation (in atomic units) [22]. The electric field of the two-color driving laser pulse $E(t)$ is

$$E(t)=E_\omega \exp(-2\ln 2t^2/\tau_\omega^2)\cos(\omega t)+E_{2\omega}\exp(-2\ln 2t^2/\tau_{2\omega}^2)\cos(2\omega t+\Delta\phi), \quad (3)$$

where E_ω and $E_{2\omega}$ are the electric intensities of the FW and SH pulses with the pulse duration of τ_ω and $\tau_{2\omega}$, respectively. The phase difference $\Delta\phi$ defines the initial phase delay between them, and both the carrier–envelope phases of the two driving laser pulses are considered as stable. By testing different intensity ratios and phase delay of the two-color laser field, we finally find some conditions that the SCP can be formed effectively. Fig. 2 compares the results driven by the FW field, and synthesized two-color field. To make their difference clearly, we use the same value of ponderomotive potential under these two conditions by choosing different intensities of FW and SH pulses. In the single-color driving field, the FW intensity is selected as $I_\omega=2.06\times10^{14}$ W/cm^2. While in the case of two-color field, the peak intensities of FW and SH pulses are chosen as $I_\omega=2.0\times10^{14}$, and $I_{2\omega}=1.0\times10^{14}$ W/cm^2, respectively. The pulse durations are all 6 fs. For the phase delay, the SCP all can be formed when $\Delta\phi$ is between 0.6π and 0.8π, and 0.7π is chosen here for example.

Firstly, by analyzing the inset in Figs. 1 and 2 comparatively, we can see that, in the single-color laser field, the waveform of electric field is symmetric between the positive and negative half cycles, then the freed electron experiences the acceleration, deceleration and acceleration again in the opposite direction only once for every electron recombination, then each electron recombination time profile has only one peak (see Fig. 2(a)). When the waveform of FW is dramatically changed rather than a small perturbation by the intense SH addition, the waveform of the synthesized electric field oscillates quickly and irregularly. The electron is accelerated and decelerated many times and as a result, the route of electronic movement is lengthened or shortened, which induces the electron to return to the parent ion earlier or later than the corresponding return times under the FW field only. As a result, the electron recombination time profile divides into two peaks in each half-cycle (see Fig. 2(b)). Additionally, according to the rigorous quantum mechanical treatment of the whole processes, we know that the HH emission intensity depends on the ionization and recombination probabilities in the driving electric field, which can also be controlled by varying the corresponding optical waveform. As shown in Fig. 2(d), when the phase delay of the two-color laser field is set at 0.7π, the Fourier components ω and 2ω are synthesized properly to allow HHs emit in one dominant half-cycle with a certain spectral span in the plateau, and its emission intensity is about five orders of magnitude stronger than those in other half cycles. HHs from other half cycles can be therefore neglected, leading to the SCP generation. On the other hand, we should notice that the intensity of SCP in the case of two-color field is comparable with the intensity of the cut-off region of the single color field, i.e., our method offers another advantage in terms of efficiency compared to the conventional spectral filtering and double optical gating (DOG) techniques [3,10].

Fig. 3(a) shows the corresponding three harmonic spectra generated by the FW field, synthesized FW–SH fields with relative phase delay $\Delta\phi=0.0\pi$ and 0.7π, respectively. For the single color field, a discrete spectrum containing only odd harmonics of the laser frequency is obtained due to the inversion symmetry between the positive and negative half cycles (the red dashed line in Fig. 3(a)) [23]. As is well known, by adding the SH pulses, the inversion symmetry is broken, and then a spectrum containing both even and odd harmonics will appear (the blue shorted line in Fig. 3(a)). For both the two-color field with $\Delta\phi=0.0\pi$ and 0.7π, the results are all quite in agreement with the classical results. Interestingly, under the case of $\Delta\phi=0.7\pi$, we can also see a smooth supercontinuum appeared in the plateau region of the spectrum (the black solid line in Fig. 3(a)), i.e. the SCP, whose frequency range is consistent with the strong emission intensity area in Fig. 2(d). This consistency of the spectral span in the plateau region confirms our analysis above well.

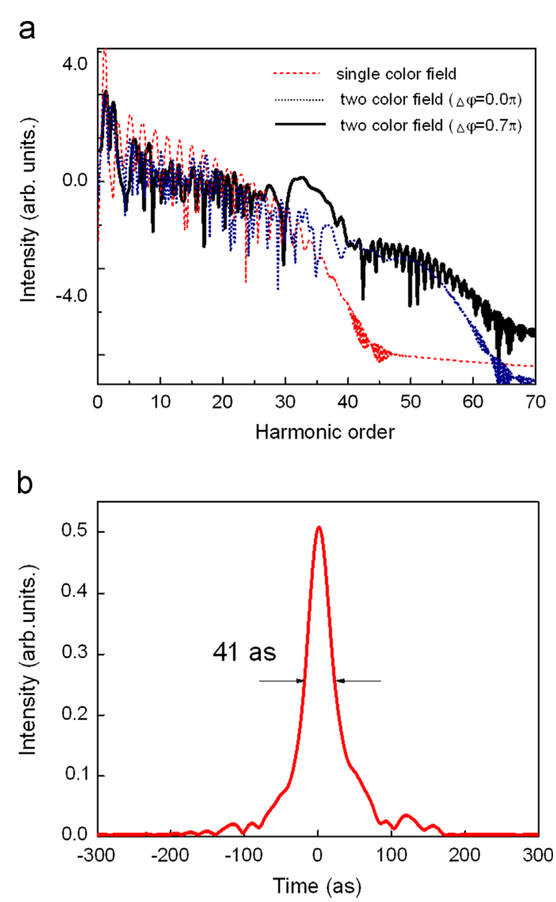

Fig. 3. (a) Harmonic spectra driven by FW field (red dashed line), FW–SH two-color field with the relative phase delay of 0.0π (blue dotted line), and 0.7π (black solid line), respectively. (b) Attosecond pulse produced by superposing the harmonics of plateau region from 25th to 36th order. The other parameters are the same as those in Fig.2. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Specially, we must emphasize that the intensity of SH pulses used here is much higher than that in other investigations of two-color field [17,18]. However, just because of the intense intensity of SH pulses added to the FW pulses, the waveform of synthesized electric field and the electronic route can be greatly changed, and then harmonics can be emitted in the same half-cycle with much stronger intensity than other in the adjacent half cycles. Therefore, under the driving of two-color electric field, both the phase delay and the intensity of SH pulses are important for the effective formation of the SCP.

In particular, by simply making an inverse Fourier transformation without any phase compensation of the supercontinuum in the plateau region (from the 25th to the 36th order), it is shown in Fig. 3(b) that an isolated 41 as pulse with a clean temporal profile can be theoretically obtained. We believe that this proposed two-color phase delayed field can provide an efficient method for the production of isolated ultrashort attosecond pulse for the future and may be extended to other systems.

4. Conclusion

We theoretically demonstrate the generation and control of high intensity SCP driving by a FW–SH two-color laser field with appropriate phase delay and electric intensity. It is proved that when the intense SH pulses are added to the FW pulses, the waveform of synthesized electric field can oscillate quickly and irregularly, and the route of electron movement can be greatly changed, and then harmonics can be emitted in the same half-cycle with much stronger intensity than other in the adjacent half cycles. Finally, smooth SCP can be formed with the corresponding spectral range consistent with the high intensity region in the time–frequency transform spectrum. Correspondingly, isolated single attosecond pulses with clean temporal profiles can be generated directly from the contribution of SCP alone even without the phase compensation.

It has been known that the SCP generation is especially desired for efficient single-attosecond-pulse generation in various spectral regions. Therefore, by utilizing SCP generation, many interesting phenomena, such as molecular clocks and molecular orbital imaging, may become practical, and further researches may promote the development of time-resolved spectroscopy of atoms and molecules and many other interdisciplinary fields.

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