Generation of an Extreme Ultraviolet Supercontinuum in a Two-Color Laser Field

Zhinan Zeng, Ya Cheng,* Xiaohong Song, Ruxin Li, and Zhizhan Xu†

State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics, CAS, P. O. Box 800-211, Shanghai 201800, China

(Received 13 December 2006; published 16 May 2007)

We theoretically investigate the high-order harmonic generation in a helium atom with a two-color optical field synthesized by an intense 6 fs pulse at 800 nm and a relatively weak 21.3 fs pulse at 400 nm. When the frequency-doubled pulse is properly time shifted with respect to the fundamental pulse, an ultrabroad extreme ultraviolet supercontinuum spectrum with a 148 eV spectral width can be generated which directly creates an isolated 65 as pulse even without phase compensation. We explain this extraordinary phenomenon by analyzing maximum electron kinetic energies at different return times.

DOI: 10.1103/PhysRevLett.98.203901 PACS numbers: 42.65.Ky

The creation of attosecond (as) extreme ultraviolet (XUV) pulses opens the route to time-resolved atomic physics since the electron motion around the atom is at the subfemtosecond (fs) time scale [1]. Although there exist several approaches to as pulse generation such as Fourier synthesis of Raman sidebands or high-order harmonics generated in noble gases [2], single as XUV pulses demonstrated to date are basically created by two techniques: namely, high-order harmonic generation (HHG) from a few-cycle driving pulse [3] and temporal confinement of the HHG by polarization-gating [4]. Limited by the currently available shortest pulse duration, the former approach is difficult to offer as pulses with durations less than ~100 as. With the latter approach, ~130 as XUV pulses have been demonstrated [5]. However, the polarization-gating technique also requires state-of-the-art few-cycle (~5 fs) pulses, and the as pulses generated with this technique are usually weak due to the fact that only a fraction of the driving pulse energy is used in the generation of as pulses. In this Letter, we will address these issues in the generation of sub-100 as XUV pulses using two-color laser field with a controlled phase delay.

In our simulation, a 6 fs/800 nm and a frequency-doubled 21.3 fs/400 nm (16 optical cycle) pulses are combined to serve as the driving pulse for generating as pulses. In fact, the pulse duration of the 400 nm pulse will not strongly affect the simulation results. The model atom in the simulation is helium (He). The method of the simulation for HHG has detailed description in Ref. [3], which is based on single-active atom approximation and has been widely used. The atomic dipole moment is calculated using Eq. [34] in Ref. [3], and the high harmonic spectrum is obtained by Fourier transforming the time-dependent dipole moment. The intensities of the fundamental and the frequency-doubled pulses are 1.4×10^{15} W/cm^2 and 5.5×10^{13} W/cm^2, respectively. The expression of the synthesized field can be written as

$$E_s = E_1 \exp[-2\ln(2)t^2/\tau_1^2]\cos(\omega t) + E_2 \exp[-2\ln(2)(t+t_0)^2/\tau_2^2]\cos[2\omega(t+t_0)]. \quad (1)$$

Here, E_1 and E_2 are the amplitudes of the electric fields of the fundamental and the frequency-doubled pulses, respectively; ω is the frequency of the fundamental pulse; and τ_1 and τ_2 are the corresponding pulse durations (FWHM). The t_0 in Eq. (1) defines the initial time delay between the fundamental and the frequency-doubled pulses, creating a relative phase of $4\pi t_0/T$ where T is the optical period of the fundamental wave. First, we carried out our simulation only using the 6 fs fundamental pulse. Next, we included the frequency-doubled pulse in our simulation and scanned the relative phase between the two pulses in order to obtain an optimum XUV supercontinuum. It is noteworthy to mention that we searched for the *optimum* supercontinuum spectrum which had not only an ultrabroad spectral width, but also a flat profile with lest spectral modulation. Both these two conditions are important for generating as pulses with clean temporal profiles. Shown in Fig. 1 are the three electric fields of the 6 fs fundamental pulse (dashed line), the two-color pulses without a time delay (dashed-dotted line), and with a $t_0 = -166.7$ as time delay (solid line).

Figure 2(a) shows the HHG spectra generated in the three optical fields in Fig. 1. The XUV supercontinuum

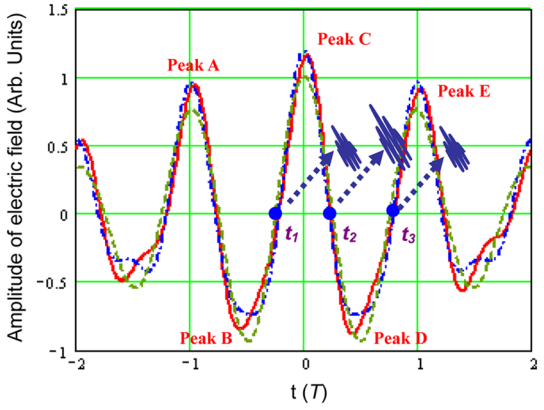

FIG. 1 (color online). Electric fields of the single 6 fs pulse (dashed line), the two-color pulses without a time delay (dashed-dotted line), and with a −166.7 as time delay (solid line).

spectrum in the cutoff region produced by the single 6 fs pulse alone shows a bandwidth of approximate 30 eV, as indicated by the curve I in Fig. 2(a). As has been reported in Ref. [6], the XUV supercontinuum in the cutoff region generated by a few-cycle optical pulse can be used for producing a single 250 as pulse. For generating as pulses with sub-100 as durations, XUV supercontinua with broader spectral widths are required. A significantly broadened XUV supercontinuum with a spectral width of ~75 eV can be achieved by superposing the weak frequency-doubled pulse onto the fundamental pulse with a zero time delay, as can be seen in Fig. 2(a) (curve II). In this case, the contribution to the broadening of the XUV supercontinuum spectrum is mainly from the extension of the cutoff region. It has been pointed out recently in Ref. [7] that the significant extension of the cutoff photon-energy after superposing a weak heterodyne pulse onto an intense fundamental pulse is a result of the heterodyne mixing of laser fields. Thus, our result for the two-color field without a time delay between the two pulses is consistent with this physical picture [7].

Surprisingly enough, a dramatically increased XUV supercontinuum spectral width was obtained when we introduced a relative phase shift between the two-color pulses. The curve III in Fig. 2(a) shows that an XUV supercontinuum of ~148 eV spectral width was produced with a time delay of $t_0 = -166.7$ as. In addition, Fig. 2(b) shows the phase profiles of the XUV supercontinua generated in the three laser fields. In particular, the nearly parabolic phase profile across the 148 eV XUV supercontinuum spectrum (curve III) implies that the phase is mostly linearly chirped, although some high-order dispersions still exist. By simply making an inverse Fourier transformation of the XUV supercontinuum in the 148 eV spectral width (we blocked all the low-order harmonics), it is shown in Fig. 3 (curve II) that such a broad XUV spectrum can support an isolated 65 as pulse even without any phase compensation. Furthermore, if all the phase dispersion over the 148 eV spectral width can be properly compensated for, an isolated 23 as pulse with a clean temporal profile could be theoretically predicted, as shown by the dashed curve I in Fig. 3. This stunning value of the as pulse duration indicates that extremely short XUV pulses with durations of a few tens of attoseconds could be attainable from the HHG in noble gases. For comparison, we also present the as pulse generated in the single-color 6 fs field by inversely Fourier transforming the 30 eV XUV supercontinuum in the cutoff region without using a phase compensation (curve III in Fig. 3). In this case, although a single as pulse is generated, its temporal profile shows two subpulses of comparable peak intensities, and each subpulse is longer than 100 as.

In order to understand the physics behind the dramatically broadened XUV supercontinuum spectrum in the two-color light field with an optimum phase delay, we

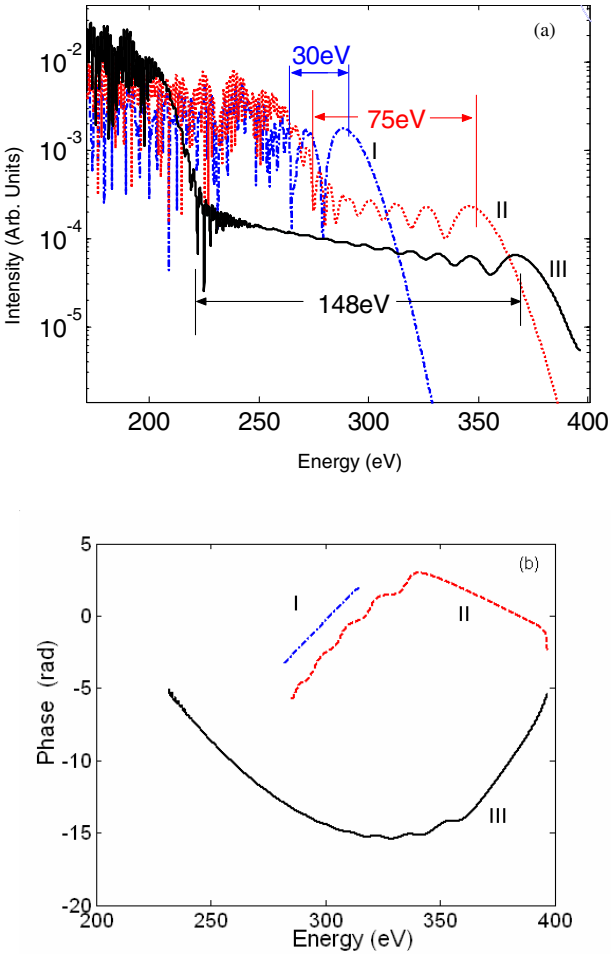

FIG. 2 (color online). (a) The spectra of the XUV supercontinua generated with the single 6 fs pulse (curve I), the two-color pulses without a time delay (curve II), and with a −166.7 as time delay (curve III). (b) The phase profiles of the XUV supercontinua in (a).

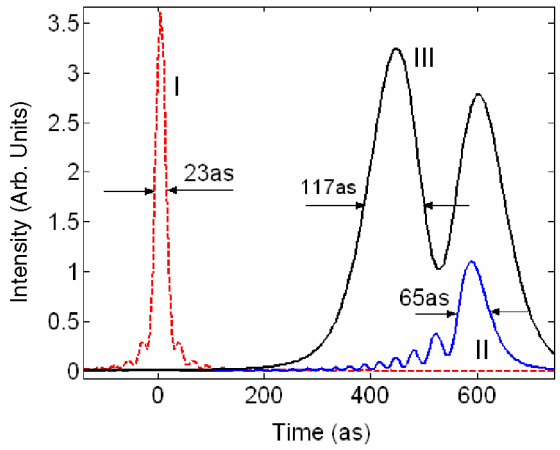

FIG. 3 (color online). The temporal profiles of the as pulses generated from the XUV supercontinuum of 148 eV spectral width with (curve I) and without (curve II) phase compensation. For comparison, the as pulse generated by the single-color 6 fs pulse without phase compensation is shown as the curve III.

performed time-frequency analyses of the dipole responses of the He atom to the three different optical fields [8,9], as presented in Fig. 4. From the time-frequency analyses, we can obtain the information on the emission times of the photons with different photon energies. In particular, we focus our attention on three photon-energy peaks which are indicated as A, B, and C in each diagram. The three peaks indicate the recombination times as well as the maximum kinetic energies of the electrons returning to their parent ions. It can be seen in Fig. 4(a) that for the XUV photons produced by the single 6 fs pulse, either the cutoff-energy difference between the peaks B and A or the cutoff-energy difference between the peaks B and C is relatively small, which limits the supercontinuum spectral width. Here, the cutoff energies of different peaks are defined as the maximum electron return energies in different half optical cycles plus the ionization potential of He atom, because for a few-cycle pulse, the electric field amplitude varies significantly from one half-cycle to the next [10]. When the frequency-doubled pulse is superposed upon the fundamental pulse with a zero time delay, the height of the peak B in Fig. 4(b) is greatly increased, though there is no significant change in either the peak A or peak C. Furthermore, after a temporal shift of the frequency-doubled pulse by -166.7 as, it can be found in Fig. 4(c) that both the peaks A and C are now greatly suppressed, whereas the peak B in Fig. 4(c) is further enhanced in comparison with that in Fig. 4(b). The enhanced cutoff-energy difference between the peaks B and A or between the peaks B and C explains why the XUV supercontinuum spectrum generated in the phase-shifted two-color field can extend towards both the short- and long-wavelength ends.

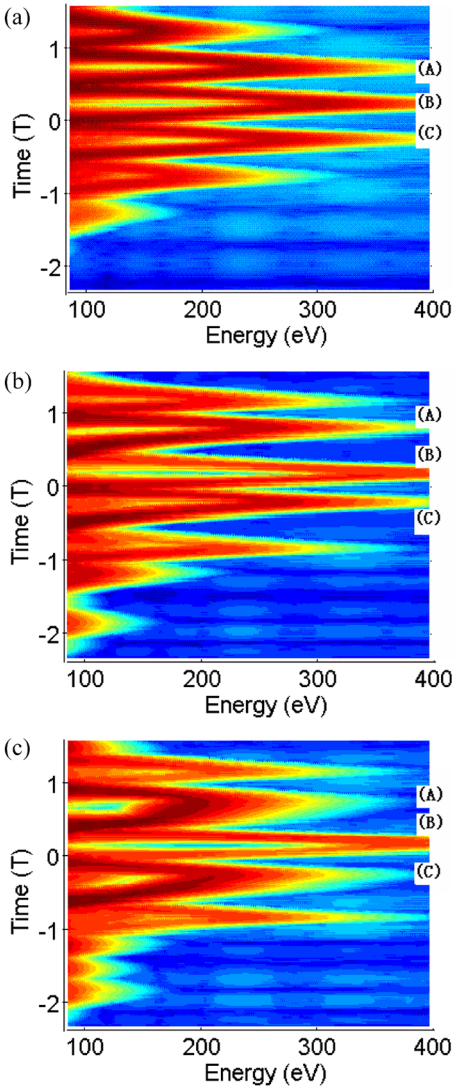

FIG. 4 (color online). Diagrams of the time-frequency analyses of the XUV supercontinuum spectra generated by (a) the single 6 fs pulse, the two-color pulses (b) without a time delay and (c) with a -166.7 as time delay. Note that the color scale in all diagrams is in logarithmic unit.

At first sight, the dramatic change of the peak heights in Figs. 4(b) and 4(c) seems a little bit hard to understand because if we now reexamine the curves in Fig. 1, we can see that the difference between the two electric fields formed by the different two-color pulses is actually not so significant. Thus, in order to get an insight into the augmented difference between the two XUV supercontinuum spectra, we analytically calculated the maximum electron kinetic energies at three return times: namely, t_1, t_2, and t_3 as indicated in Fig. 1. Note that the three return times are slightly different for the ionized electrons in the single 6 fs field and the two two-color fields. The expressions of the synthesized electric fields (E_s) have three different forms with $E_2/E_1 = 0$ for the single 6 fs pulse, $E_2/E_1 = 0.198$ and $t_0 = 0$ for the two-color pulse without a time delay, and $E_2/E_1 = 0.198$ and $t_0 = -166.7$ as for the two-color pulse with the relative time delay. Thus, both the times of the birth of the electrons (t_i^b) and of their returning to the parent ions can be obtained by solving the following equation

$$\int_{t_i^b}^{t_i}\left(\int_{t_i^b}^{t} E_s(t)dt\right)dt = 0, \tag{2}$$

where $i = 1$, 2, and 3. And the maximum kinetic energies of the electrons returning to their parent ions at t_i can be calculated as

$$E_{ki} = \kappa\left(\int_{t_i^b}^{t_i} E_s(t)dt\right)^2, \tag{3}$$

where the coefficient κ is a constant related to the mass of the electron. Combining Eqs. (1)–(3) gives out the maximum electron kinetic energies at the return times t_i in all the three optical fields. The calculated results are listed in Table I. Clearly, one can see that in the case of using a single 6 fs driving pulse, the maximum kinetic energy at the return time t_2 is the lowest one in the t_2 row of Table I, whereas the corresponding maximum electron kinetic energies at the return times t_1 and t_3 are the highest in the t_1 and t_3 rows of Table I, respectively. On the contrary, when the two-color field with the -166.7 fs time delay is used,

TABLE I. Maximum electron kinetic energies at different return times in the three optical fields.

Optical field Return time	Single 6 fs pulse	Two-color pulse $t_0 = 0$	Two-color pulse $t_0 = -166.7$ as
t_1	$\sim 0.033\kappa E_1^2$	$\sim 0.033\kappa E_1^2$	$\sim 0.029\kappa E_1^2$
t_2	$\sim 0.037\kappa E_1^2$	$\sim 0.041\kappa E_1^2$	$\sim 0.043\kappa E_1^2$
t_3	$\sim 0.032\kappa E_1^2$	$\sim 0.031\kappa E_1^2$	$\sim 0.027\kappa E_1^2$

the situation becomes totally inverted. In this case, the maximum kinetic energy at the return time t_2 is increased by $\sim$16.2% compared to that obtained in the single 6 fs driving pulse; whereas the maximum kinetic energies at the return times t_1 and t_3 are, respectively, decreased by $\sim$12.2% and $\sim$15.6% compared to those obtained in the single 6 fs pulse. This feature is qualitatively consistent with the result shown in the time-frequency diagrams in Fig. 4. Since the Coulomb potential of the He atom is not considered in this classical model calculation, quantitative differences between the results in Table I and Fig. 4 still remain.

It should be noted that the frequency-doubled pulse must be significantly weaker than the fundamental pulse in our scheme, although this seems somewhat counter-intuitive. The role of the frequency-doubled pulse is to enhance the amplitude difference between the central peak (C) and the remaining peaks (e.g., A, B, D, E) in the two-color electric field as shown in Fig. 1, which is equivalent to shortening the few-cycle driving pulse duration. One can see that after the frequency-doubled pulse is added, the amplitudes of peaks B and D are significantly reduced, whereas the amplitudes of the peaks A, C, and E are enhanced. Thus, if the frequency-doubled pulse is too intense, the peaks A and E will eventually become strong enough for generating energetic XUV photons, leading to a narrowing in the bandwidth of the XUV supercontinuum.

To conclude, we theoretically investigated the generation of ultrabroad XUV supercontinuum in two-color laser field with an optimized phase delay. Both numerical simulations and analytical calculations were carried out, and qualitatively consistent results were obtained. Our simulations are based on a single-active atom model; thus, the generated spectrum is robust and should not be very sensitive to propagation effects. Besides, the ionizations of He atoms are not strong under the peak intensities used in our modeling. The calculated ionization ratios are $\sim$6%, $\sim$27%, and $\sim$23% in the three simulations of the one-color field, the two-color fields without a time delay, and with the time delay, respectively. The efficiency of the generation of the Fourier-transform-limited, 23 as XUV pulse is estimated to be 1.5×10^{-11} by dividing the total energy of the single 23 as pulse by the total energy of the 6 fs driving pulse. Our results show again that the XUV supercontinuum generation is a highly nonlinear optical process sensitive to the slight change of the laser field. Before, this effect has been demonstrated by adding weak frequency-doubled pulses into strong fundamental pulses with relatively long pulse durations [11–13], and a recent experiment showed that superposing a weak second harmonic pulse upon a 9 fs driving pulse can result in an XUV supercontinuum which could support a single 200 as pulse [14]. In addition, significant enhancement of high-order harmonics in a two-color laser field synthesized by a strong frequency-doubled laser pulse and a weak fundamental pulse has also been reported recently [15]. Unlike these research, our work emphasizes the necessity of using fundamental pulses as short as possible and the importance of controlling the intensity ratio as well as the phase shift between the two-color pulses for the purpose of producing ultrabroad and spectrally flat XUV supercontinua. The obtained 148 eV spectral width is clear evidence of the potential of generating sub-50 as XUV pulses through HHG in noble gases, opening a new door for observing the electron motion with unprecedented time resolution. Lastly, we point out that the laser conditions in our simulations are presently available in quite a few laboratories around the world, facilitating an easy demonstration of this scheme.

The work was supported by the Major Basic Research Project of Shanghai Commission of Science and Technology, and Grants No. 2006CB806000 from National Basic Research Program of China and No. 60578049 from Natural Science Foundation of China.

*Electronic address: ycheng-45277@hotmail.com
†Electronic address: zzxu@mail.shcnc.ac.cn

[1] M. Drescher *et al.*, Nature (London) **419**, 803 (2002).
[2] P. Agostini and L. F. DiMauro, Rep. Prog. Phys. **67**, 813 (2004).
[3] T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545 (2000).
[4] I. J. Sola *et al.*, Nature Phys. **2**, 319 (2006).
[5] G. Sansone *et al.*, Science **314**, 443 (2006).
[6] A. Baltuška *et al.*, Nature (London) **421**, 611 (2003).
[7] T. Pfeifer *et al.*, Phys. Rev. Lett. **97**, 163901 (2006).
[8] X. M. Tong and Shih-I Chu, Phys. Rev. A **61**, 021802 (2000).
[9] J. J. Carrera, X. M. Tong, and Shih-I Chu, Phys. Rev. A **74**, 023404 (2006).
[10] C. A. Haworth *et al.*, Nature Phys. **3**, 52 (2007).
[11] T. Pfeifer *et al.*, Opt. Lett. **31**, 975 (2006).
[12] N. Dudovich *et al.*, Nature Phys. **2**, 781 (2006).
[13] J. Mauritsson *et al.*, Phys. Rev. Lett. **97**, 013001 (2006).
[14] Y. Oishi *et al.*, Opt. Express **14**, 7230 (2006).
[15] T. T. Liu *et al.*, Phys. Rev. A **73**, 063823 (2006).