Enhanced high-order harmonics and an isolated short attosecond pulse generated by using a two-color laser and an extreme-ultraviolet attosecond pulse

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An efficient method to generate an isolated short attosecond pulse is presented theoretically. We show that, by adding a 0.5 fs, 62.3 nm extreme ultraviolet (xuv) attosecond pulse to a synthesized two-color field at a proper time, the harmonic intensity is effectively enhanced by 1–3 orders of magnitude compared with the two-color case and the harmonic spectrum higher than 260th order reveals an ultrabroad xuv supercontinuum. In addition, the short quantum path is selected to effectively contribute to the high order harmonics, and an isolated 40 as pulse is obtained. Our calculated results also show that, by adding a 0.5 fs, 29.6 nm xuv attosecond pulse to the synthesized two-color field at a proper time, the enhancement of the high-order harmonics and an ultrabroad xuv supercontinuum is observed, and the selection of the long quantum path can be achieved, then an isolated attosecond pulse as short as 39 as is generated.

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High-order harmonic generation (HHG) has been a topic of great interest due to its potential application as a coherent soft-x-ray source [1] and the generation of attosecond (as) pulses [2,3]. So far the HHG is the most promising approach to generate attosecond pulses. The HHG process can be well understood by the three-step model [4]: ionization, oscillation, and recombination. During the recombination, a photon with energy equal to the ionization potential plus the kinetic energy of the recombining electron is emitted. This process is repeated every half an optical cycle of the laser field and generates two attosecond pulses in each optical cycle. However, for practical application, the straightforward attosecond metrology prefers an isolated attosecond pulse [5], thus much effort has been paid out to obtain an isolated attosecond pulse. It has been shown that a few-cycle laser pulse can produce an isolated attosecond pulse by filtering several consecutive harmonics in the cutoff region [6]. Due to the limitation of the supercontinuum bandwidth, the minimum duration of the isolated pulse is 250 as [7]. Using a few-cycle laser pulse with the polarization gating techniques, Sansone *et al.* [8] generated a broad supercontinuum with a bandwidth of 36 eV, then a single 130 as pulse was obtained after compensating the harmonic chirp. Isolated 80-as pulse of extreme ultraviolet light was obtained by recent experiment [2]. Very recently, it has been proposed that two-color field scheme can broaden the bandwidth of the supercontinuum and obtain an isolated attosecond pulse with much shorter pulse duration. To realize the goal, a great deal of work has been done. Zeng *et al.* [9] theoretically showed that a 148-eV-broadwidth supercontinuum could be generated by a two-color field, then an isolated 65 as pulse was attained. Zhai *et al.* [10] proved a combined two-color field scheme can broaden the high order harmonics plateau, and obtained an isolated 63 as pulse. However, due to the inherent characteristic of the two-color scheme, the efficiency of the continuous harmonics is low, which limits the application of generating single attosecond pulse. Hence, how to enhance the efficiency of the continuous harmonics has attracted a lot of attention. Biegert *et al.* [11] theoretically predicted and experimentally confirmed that the enhancement of HHG is possible by combining an attosecond pulse train with an IR driving laser. It is shown that the addition of an xuv field to a strong IR field leads to the appearance of new harmonics in the harmonic generation spectra [12]. Zhang *et al.* [13] demonstrated that the HHG efficiency could be efficiently enhanced by combined the two-color field with the coherent superposition state and obtained an intense isolated 47 as pulse. Moreover, control of quantum paths is another efficient method to generate an isolated attosecond pulse. For the single-atom response, the quantum path selection can be achieved by two-color field [14] or an attosecond pulse train [15]. Macroscopically, the short path can be realized by the phase-matching condition [16] or spatial filtering [17].

In this paper, based on the single-active electron approximation, the HHG and attosecond pulses generation can be investigated by solving the one-dimensional time-dependent Schrödinger equation with the splitting operator method. In our simulation, we choose a soft-core Coulomb potential model $V(x)=-z/\sqrt{x^2+a}$ and set $z=2$ and $a=0.5$ corresponding to the binding energy of 54.4 eV for the ground state of a He^+ ion. The electric field of the driving laser can be expressed as $E(t)=E_0f_0(t)\cos(\omega_0 t)+E_1f_1(t)\cos(\omega_1 t)+E_{\mathrm{xuv}}f_{\mathrm{xuv}}(t-\tau_{\mathrm{delay}})\cos[\omega_{\mathrm{xuv}}(t-\tau_{\mathrm{delay}})]$, here E_i and ω_i $(i=0,1,\mathrm{xuv})$ are the peak amplitudes and frequencies of the fundamental, subharmonic, and xuv pulses, respectively. $f_i(t)=\exp[-4\ln 2(\frac{t}{\tau_i})^2]$ denotes the Gauss envelopes $(i=0,1,\mathrm{xuv})$. τ_i is the pulse duration at full width of half-maximum $(i=0,1,\mathrm{xuv})$. τ_{delay} is the time delay of the xuv pulse with respect to the fundamental pulse.

We first investigate the HHG of He^+ ion in a two-color field (5 fs, 800 nm and 10 fs, 1200 nm) and the combination of the synthesized two-color field and an xuv pulse (5 fs, 800 nm, 10 fs, 1200 nm, and 0.5 fs, 62.3 nm, also called the combined field), respectively. In our calculation, the intensities of the fundamental, subharmonic and xuv pulses are cho-

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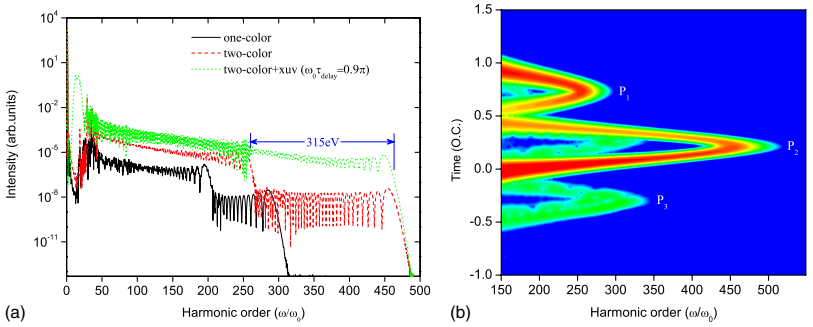

FIG. 1. (Color online) (a) Harmonic spectra of He^+ in a 5 fs, 800 nm laser field (solid black curve), in the synthesized field of a 5 fs, 800 nm and a 10 fs, 1200 nm laser pulses (dashed red curve) and in the combination of the synthesized two-color field and an xuv attosecond pulse (0.5 fs, 62.3 nm) at $\omega_0\tau_{delay}=0.9\pi$ (dotted green curve). (b) The time-frequency distribution of the HHG spectrum corresponding to the dotted green curve in Fig. 1(a).

sen to be 2×10^{15}, 1×10^{14}, and 3×10^{13} W/cm^2, respectively. For comparison, the harmonic spectrum of He^+ ion in the fundamental field alone is also presented. As shown by solid black curve in Fig. 1(a), the harmonic spectrum shows a double-plateau structure, and the spectrum cutoff is only at the 287th order harmonic, and the spectral structure above $220\omega_0$ is smooth. For the two-color case, the harmonic spectrum also reveals a double-plateau structure similar to that of one-color field as shown by dashed red curve in Fig. 1(a), whereas the spectrum cutoff is significantly expanded to $453\omega_0$. In addition, the harmonic intensity in the first plateau is enhanced effectively, and the harmonic spectrum above $310\omega_0$ is smooth. Thus a broad supercontinuum spectrum is formed in the second plateau. However, the low efficiency and large modulation of the supercontinuum is not beneficial for us to produce an intense isolated attosecond pulse. By adding a 0.5 fs, 62.3 nm xuv pulse to the synthesized two-color field at $\omega_0\tau_{delay}=0.9\pi$, the overall harmonic spectrum is uplifted, and almost does a plateau exist, as shown by dotted green curve in Fig. 1(a). The harmonic intensity is effectively enhanced by 1–3 orders of magnitude compared with the two-color case. The spectrum cutoff is at about $453\omega_0$ and is no apparent change, this is because the contribution of the xuv attosecond pulse to the ponderomotive energy is negligible. Furthermore, the harmonic spectrum from the $260\omega_0$ becomes smooth but much less modulated, and the bandwidth of the continuous harmonics is much broader than the two-color case. Particularly, we note that the intensity of the broad supercontinuum is greatly enhanced by 2 orders of magnitude and is comparable with that of the low-order irregular harmonics. Such a strong and broad supercontinuum is highly likely to support the generation of an intense isolated short attosecond pulse.

Figure 1(b) presents the time-frequency distribution of the HHG [18] shown by the dotted green curve in Fig. 1(a). From this figure, one can clearly see that there are three main peaks marked as P_1, P_2, and P_3 with the maximal harmonic orders of 250, 453, and 300, respectively. P_2 and P_1 have comparable intensity, and P_3 is weaker than P_1 and P_2 and its contribution to the HHG can be ignored. Considering the above results, the bandwidth of the supercontinuum is greatly broadened to about 315 eV and is larger than the value of classical calculation, this is because the effect of the ionization probability on harmonic efficiency is considered in quantum calculation. Particularly, for the peak P_2, the intensity of the short path is much higher than that of the long path. This implies that the short path is selected effectively to contribute to the HHG. Furthermore, the efficient enhancement of the short path also implies that the yields of short-path harmonics are rather efficient, which leads to that the harmonic spectrum almost presents a plateau. Note, for the peak P_2, the leading edge of the xuv pulse also enhances the yields of long-path harmonics to a certain extent, which is similar to the result of Ref. [19]. An efficient method for removing the long path is to reduce the duration of the xuv pulse or to adjust the time delay of the xuv pulse with respect to the fundamental pulse. Hereinafter, one will see the expected results.

Next, we investigate the attosecond pulse generation in the combined field. The temporal profiles of the attosecond pulses can be obtained by superposing several harmonics. From Fig. 1(b), one can note the HHG is mainly emitted near 0.065 optical cycle (o.c.); this implies that a short isolated as pulse can be generated by superposing several harmonics which are emitted in phase. We select the harmonics from 265th to 325th order, an isolated 40 as pulse is obtained, as shown in Fig. 2(a). From our calculation, the intensity of the attosecond pulse is 4 orders of magnitude higher than that in the one-color field and is 3 orders of magnitude higher than that in the two-color field. Note, after the main pulse there is a satellite pulse originating from the long path, but the intensity ratio of the satellite pulse to the main pulse is so small (only 0.05) that it can be ignored. In addition, we investigate the dependence of the isolated attosecond pulse on the intensity, duration and time delay of the xuv pulse. Figure 2(b) shows that the intensity of the generated attosecond pulse is significantly enhanced with increasing the intensity of the xuv pulse, whereas the duration of each attosecond pulse is 40 as. Note that the rapid advancement of HHG has made it available for an xuv pulse with a intensity of about 1×10^{14} W/cm^2 [20,21], thus a more intense isolated attosecond pulse can be generated by using a stronger xuv pulse. Figure 2(c) shows the temporal profiles of the attosecond pulses for three different durations. One can see that, for these three cases, clean isolated attosecond pulses are obtained, and the intensities of the satellite pulses are about 0.02, 0.05, and 0.11 of the main pulses, respectively. The durations of the main pulses for these three cases are, respectively, 43, 40, and 40 as. According to our calculation, under the condition of $\tau_{xuv}\leq1$ fs, we can keep the intensity ratio of the satellite pulse to the main pulse below 20%. Figure 2(d) shows the temporal profiles of the generated attosecond pulses for three different delays. For the cases of $\omega_0\tau_{delay}=0.7\pi$ and $\omega_0\tau_{delay}=0.9\pi$, isolated attosecond pulses are observed, but the former is much lower than the latter in pulse energy. It should be noted that the long quantum path is

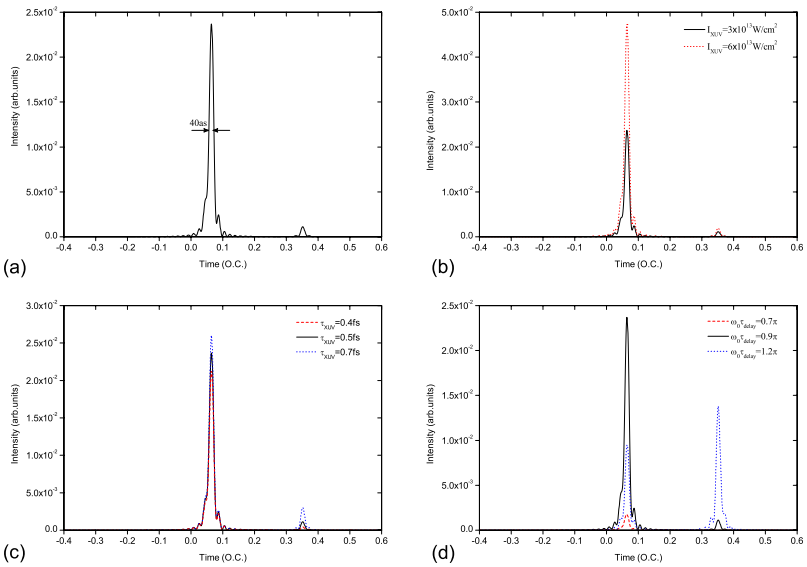

FIG. 2. (Color online) The temporal profiles of the attosecond pulses in the combined field at a proper time. (a) $I_{xuv}=3\times10^{13}$ W/cm^2, $\omega_0\tau_{delay}=0.9\pi$. (b) Solid black curve: $I_{xuv}=3\times10^{13}$ W/cm^2; dotted red curve: $I_{xuv}=6\times10^{13}$ W/cm^2. (c) Dashed red curve: $\tau_{xuv}=0.4$ fs; solid black curve: $\tau_{xuv}=0.5$ fs; dotted blue curve: $\tau_{xuv}=0.7$ fs. (d) Dashed red curve: $\omega_0\tau_{delay}=0.7\pi$; solid black curve: $\omega_0\tau_{delay}=0.9\pi$; dotted blue curve: $\omega_0\tau_{delay}=1.2\pi$. Others parameters are the same as in Fig. 1.

removed at $\omega_0\tau_{delay}=0.7\pi$. For the case of $\omega_0\tau_{delay}=1.2\pi$, two 40 as bursts are emitted, the intensity of the prepulse is lower than that of the postpulse, but the intensities of the two attosecond pulses are lower than the case of $\omega_0\tau_{delay}=0.9\pi$. This indicates that both the intensity and the profile of the attosecond pulses have a certain degree of dependence on the time delay.

In order to study the underlying physics of enhancement of the HHG, we calculate the ionization probability in the combined field. In our simulation, the intensity of the laser pulse is much lower than the saturation intensity of the He^+ ion. According to three-step model, the harmonic efficiency is mainly laid on the ionization rate. In our two-color scheme, the continuous harmonics mainly originate from the electrons ionized in the time range from −0.57 to −0.3 o.c., since the ionization rate of the electrons ionized in the time range is slower (see dotted blue curve in Fig. 3), the efficiency of the continuous harmonics is lower. However, by adding a 0.5 fs, 62.3 nm xuv pulse to the synthesized two-color field at $\omega_0\tau_{delay}=0.9\pi$, the situation is completely different. As shown by dashed red curve in Fig. 3, the ionization probability increases rapidly near $t=-0.45$ o.c., which indicates that the ionization rate of the electrons contributing to the continuous harmonics is greatly increased, resulting in the efficient enhancement of the continuous harmonics. It should be emphasized that, after the ionization time later than −0.47 o.c., the short path dominates over the long path. In fact, the 62.3 nm xuv pulse can promote electronic transition from 1 *s* to 2 *s* or 2 *p* state for a He^+ system. The electrons populated in the states are easily ionized by the driving field owing to the low binding energy. Thus the ionization yields are increased significantly. As a result, the conversion efficiency of the HHG is improved dramatically compared with the two-color case.

In our simulation, we find that not only the short path but also the long path can also be selected to effectively contribute to the HHG. For the case, the calculated results are presented in Fig. 4. Where we add a 0.5 fs, 29.6 nm xuv pulse to the synthesized two-color field and choose $\omega_0\tau_{delay}=1.22\pi$. Others laser parameters are the same as in Fig. 1. Figure 4(a) shows the harmonic spectrum of He^+ ion in the combination of the synthesized two-color field and the 0.5 fs, 29.6 nm xuv pulse. As shown in this figure, the harmonic spectrum exhibits a two-plateau structure with two cutoffs, one is at the 250th harmonic, the other is at the 453rd harmonic. The harmonics in the second plateau not only are smooth and less modulated but also are 1–2 orders of magnitude higher than the two-color case in harmonic intensity. Figure 4(b) shows the corresponding time-frequency distribution. For the harmonics above $265\omega_0$, although there are two dominating quantum paths contributing to the same harmonic, the contribution of the long path is much higher than that of the short one, i.e., the long-path harmonics is effectively selected. Thus the harmonics in the second plateau are almost emitted in phase, and then the superposition of these synchronous harmonics leads to an isolated attosecond pulse. Figures 4(c) and 4(d) check the above analyses. Figure 4(c) illustrates the single attosecond pulse generated by superposing the harmonics from 265th to 470th order. As shown in this figure, a single 55 as pulse with several weaker subpeaks is obtained. When the harmonics superposed are from 265th

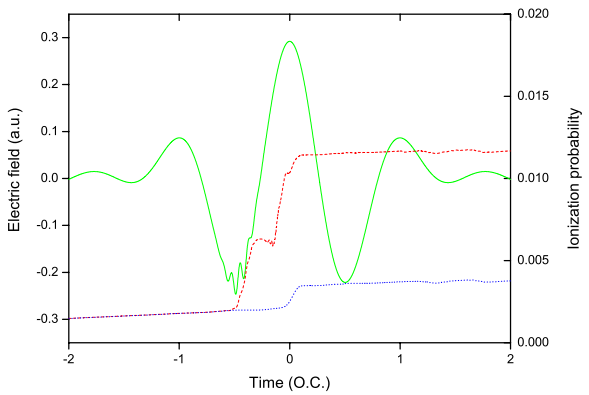

FIG. 3. (Color online) The electric field (solid green curve) and the dependence of the ionization probability on time (dashed red curve) in the combined field at $\omega_0\tau_{delay}=0.9\pi$ and (dotted blue curve) in the two-color field. The laser parameters are the same as in Fig. 1.

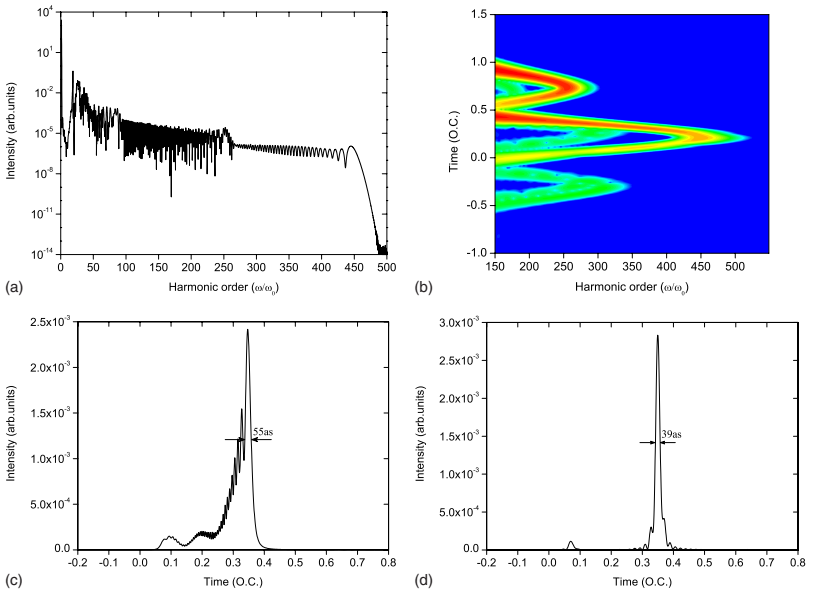

FIG. 4. (Color online) (a) Harmonic spectrum. (b) The time-frequency distribution. The temporal profiles of the attosecond pulses by superposing the harmonics (c) from 265th to 470th order and (d) from 265th to 325th order. A 0.5 fs, 29.6 nm xuv pulse is chosen and the time delay is at $\omega_0\tau_{\mathrm{delay}}=1.22\pi$, other laser parameters are the same as in Fig. 1.

to 325th order, an isolated 39 as pulse with a bandwidth of about 93 eV pulse is obtained, as shown in Fig. 4(d). The intensity of the attosecond pulse is 2 orders of magnitude higher than the two-color case. Further, the satellite pulse before the main pulse originates from the short path. However, the intensity of the satellite pulse only occupies 0.04 of the main pulse, so it can be ignored.

In conclusion, we theoretically present an efficient method to generate an isolated attosecond pulse in the combination of a synthesized two-color field and an xuv attosecond pulse. We also show that the HHG enhancement and the selection of a specific quantum path can be realized by using an xuv pulse with different central energy at a proper time. Specifically, in our two schemes, the enhancement of the harmonics in the supercontinuum region is much more efficient. Since the xuv pulse holds the high photon energy and the extremely short pulse duration, the production time and property of the electron wave packet (EWP) can be operated. As a result the ionization yields of the electrons with a specific quantum path can be increased, i.e., one of two quantum paths is selected to effectively contribute to the HHG. It should be emphasized that, in our schemes, increasing the duration of the 1200 nm laser pulse up to 64 fs or keeping the wavelength of the subharmonic laser pulse in the region of 1160–1240 nm, we find that the results have little change. Currently, a 5 fs, 800 nm laser pulse is available in a good few laboratories, and a 10 fs, 1200 nm laser pulse can be generated by optical parametric amplifier (OPA) from an 800 nm laser pulse [22], besides, a 0.5 fs, 62.3 nm and a 0.5 fs, 29.6 nm xuv pulses can be produced by synthesizing the high-order harmonics such as Refs. [3,6]. Therefore, the schemes presented here appear feasible for an experimental demonstration in the near future. The importance we think is our schemes can overcome the limitation of the low efficiency in the HHG emission and produce an ultrabroad xuv supercontinuum, which benefits the generation of an intense short attosecond pulse.

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[1] M. Drescher *et al.*, Science **291**, 1923 (2001).
[2] E. Goulielmakis *et al.*, Science **320**, 1614 (2008).
[3] R. Kienberger *et al.*, Nature (London) **427**, 817 (2004).
[4] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[5] I. P. Christov, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. Lett. **78**, 1251 (1997).
[6] M. Hentschel *et al.*, Nature (London) **414**, 509 (2001).
[7] A. Baltuška *et al.*, Nature (London) **421**, 611 (2003).
[8] G. Sansone *et al.*, Science **314**, 443 (2006).
[9] Z. Zeng, Y. Cheng, X. Song, R. Li, and Z. Xu, Phys. Rev. Lett. **98**, 203901 (2007).
[10] Z. Zhai and X. S. Liu, J. Phys. B **41**, 125602 (2008).
[11] J. Biegert *et al.*, Laser Phys. **15**, 899 (2005).
[12] A. Fleischer, Phys. Rev. A **78**, 053413 (2008).
[13] G. T. Zhang and X. S. Liu, J. Phys. B **42**, 125603 (2009).
[14] C. M. Kim and C. H. Nam, J. Phys. B **39**, 3199 (2006).
[15] K. J. Schafer, M. B. Gaarde, A. Heinrich, J. Biegert, and U. Keller, Phys. Rev. Lett. **92**, 023003 (2004).
[16] P. Antoine *et al.*, Phys. Rev. A **56**, 4960 (1997).
[17] R. López-Martens *et al.*, Phys. Rev. Lett. **94**, 033001 (2005).
[18] P. Antoine, B. Piraux, and A. Maquet, Phys. Rev. A **51**, R1750 (1995).
[19] W. Hong *et al.*, J. Opt. Soc. Am. B **25**, 1684 (2008).
[20] A. D. Bandrauk, S. Chelkowski, and N. H. Shon, Phys. Rev. Lett. **89**, 283903 (2002).
[21] H. Mashiko *et al.*, Opt. Lett. **29**, 1927 (2004).
[22] C. Vozzi *et al.*, Opt. Lett. **32**, 2957 (2007).