Generation of an isolated sub-30 attosecond pulse in a two-color laser field and a static electric field*

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We theoretically investigate high-order harmonic generation (HHG) from a helium ion model in a two-color laser field, which is synthesized by a fundamental pulse and its second harmonic pulse. It is shown that a supercontinuum spectrum can be generated in the two-color field. However, the spectral intensity is very low, limiting the application of the generated attosecond (as) pulse. By adding a static electric field to the synthesized two-color field, not only is the ionization yield of electrons contributing to the harmonic emission remarkably increased, but also the quantum paths of the HHG can be significantly modulated. As a result, the extension and enhancement of the supercontinuum spectrum are achieved, producing an intense isolated 26-as pulse with a bandwidth of about 170.5 eV. In particular, we also analyse the influence of the laser parameters on the ultrabroad supercontinuum spectrum and isolated sub-30-as pulse generation.

Keywords: high-order harmonic generation, isolated attosecond pulse, supercontinuum spectrum, combined field

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1. Introduction

When an intense short laser pulse interacts with atoms or molecules, high-order harmonic radiation is generated.[1−3] Generally, a typical spectrum of high-order harmonic generation (HHG) shows a rapid decrease for the first few harmonics, followed by a broad plateau of almost constant conversion efficiency, ending up with a sharp cutoff. Because HHG covers a large spectral range from the infrared to soft x-ray region with equidistant frequency, it has become a candidate for breaking through the femtosecond (fs) limit. Additionally, HHG is a promising light source of isolated attosecond pulses. The physical origin of HHG can be well explained according to a semiclassical three-step model.[4] In detail, an electron first tunnels through the barrier formed by the Coulomb potential and the laser field. Then, it oscillates almost freely in the laser field. Finally, the ionized electron may recombine with the parent ion, emitting energetic photons. On the basis of this model, the maximum harmonic photon energy is equal to $I_{\rm p}+3.17U_{\rm p}$, where $I_{\rm p}$ and $U_{\rm p}$ correspond to the atomic ionization potential and the ponderomotive energy of the electron, respectively. For a long laser pulse, this process periodically occurs every half optical cycle, leading to the generation of an attosecond pulse train (APT). In practical application, an isolated atttosecond pulse is more important than an APT. Therefore, a great deal of effort has been made to produce isolated attosecond pulses. It has been demonstrated that an isolated attosecond pulse can be obtained by using a few-cycle laser pulse[5−7] or a polarization gating technique.[8] By decreasing the duration of the laser pulse to 3.3 fs, an isolated 80-as pulse, which is currently the shortest attosecond pulse, was realized in a recent experiment.[9] However, the duration of isolated attosecond pulses is still far greater than the natural time scale of electronic motion in an atom (24 as). Therefore, there is an urgent need to generate isolated attosecond pulses with broader bandwidth and shorter pulse duration.

Recently, it has been theoretically proposed that a two-color field scheme can broaden the attosecond pulse bandwidth and shorten the pulse duration. With this controlling scheme, many people have successfully synthesized broadband isolated attosecond pulses, such as a 65-as pulse using a two-color

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field,[10] a 67-as pulse using a multicycle two-color field,[11] a 96-as pulse using a two-color time-gating laser field,[12] a 26-as pulse using the combination of an intense few-cycle chirped laser and its high-order harmonic pulses,[13] a 59-as pulse using a multicycle chirped two-color pulse,[14] and a 39-as pulse using intense multicycle 800-nm laser and its 27th harmonic pulse.[15] An alternative method for generating a broadband attosecond pulse is through controlling quantum paths, which can significantly modulate the HHG process.[16,17] From a macroscopic point of view, the short path can be selected by carefully adjusting the phase-matching condition[18,19] or by spatial filtering.[20] For the single-atom response, selection of the short quantum path can be achieved by using a two-color field,[21,22] an APT,[23] or a static electric field.[24,25] Very recently, we also note that a variety of schemes using a static electric field or a low-frequency laser field have been used for HHG and attosecond pulse generation.[26−30] Wang *et al.*[26] studied the effect of a static electric field on HHG with the full quantum theory, and found that the presence of the static electric field can not only break the inversion and reflection symmetry, but also can effectively extend the HHG cutoff. Moreover, they also investigated the ellipticity dependence of the HHG in the presence of a static electric field and found two different mechanisms of HHG.[27] Hong *et al.*[28] succeeded in generating a broadband extreme ultraviolet (XUV) supercontinuum, and a close-to-Fourier-limit sub-50-as pulse by combining a phase-stabilized few-cycle laser pulse and a low-frequency field. Xiang *et al.*[29] theoretically demonstrated that both extension of the HHG cutoff and generation of an ultrabroad continuum spectrum covering about 700 order harmonics can be realized in the combined field of a chirped laser pulse and a static electric field, and they also obtained an isolated sub-100-as pulse without any phase compensation. Du *et al.*[30] produced an isolated sub-30-as pulse by using a two-color field and a high-order harmonic pulse. The low-frequency field strength is extremely high (0.1 a.u.), which can hardly be experimentally achieved at present.

In this paper, we theoretically investigate the HHG from a helium ion model in a synthesized two-color laser in combination with a static electric field. It is shown that this method can not only remarkably increase the ionization yield of electrons, but also significantly modulate the quantum paths of HHG. As a result, the bandwidth of the supercontinuum spectrum is largely broadened and the efficiency of harmonics in the supercontinuum is greatly enhanced, compared with the case of a two-color field. By filtering some well-chosen continuous harmonics, an intense isolated 26-as pulse is directly obtained. Particularly, the influences of the laser parameters on the ultrabroad supercontinuum spectrum and isolated sub-30-as pulse generation are also investigated.

2. Model and methods

In our simulation, we use a soft-core Coulomb potential $V(x) = -z/\sqrt{x^2+a}$ and set $z = 2$ and $a = 0.5$ corresponding to the binding energy of the ground state of a helium ion, i.e., I_p =54.4 eV. The driving pulse is composed of a two-color field (fundamental: 5 fs, 800 nm; 2rd harmonic: 5 fs, 400 nm) and a static field. The intensity of the fundamental pulse is 1.3×10^{15} W/cm^2 and the intensities of the second harmonic (SH) pulse and static field are both 1.0×10^{14} W/cm^2. In fact, the duration of the SH pulse will not obviously affect our simulated results. The electric field of the driving pulse can be expressed as

$$E(t) = f(t)[E_0\cos(\omega_0 t) + E_1\cos(2\omega_0 t - \phi)] + E_2, \quad (1)$$

where $f(t) = \exp[-4\ln 2(t/\tau)^2]$ presents the profile of the laser field, and $\tau = 5$ fs is the pulse duration (full width at half maximum) of the laser field. E_0, E_1, and E_2 are the amplitudes of the fundamental, SH, and static fields, respectively. ω_0 is the frequency of the fundamental pulse. ϕ is the relative phase between two laser fields.

In order to verify our scheme, we perform a numerical investigation into the HHG spectrum and the attosecond pulse generation in the two-color field and the combined field of the two-color field with the static electric field, respectively. The HHG can be studied by numerically solving the one-dimensional time-dependent Schrödinger equation with the splitting-operator method,[31] which is based on the single-active-electron approximation and has been widely employed for HHG simulation. The harmonic spectrum is obtained by Fourier transforming the time-dependent dipole acceleration of a model atom. The temporal profile of attosecond pulses can be derived by simply performing inverse Fourier transformation of the XUV supercontinuum in different spectral regions.

3. Results and discussion

Figure 1(a) presents the harmonic spectra from He^+ with $\phi = 0$, where the dotted curve shows the harmonic spectrum in the two-color field. As shown in

this figure, the harmonic spectrum exhibits a double-plateau structure with two cutoffs: one is at the 131st-order harmonic corresponding to an energy of $I_p + 148$ eV, the other is at the 218th-order harmonic corresponding to an energy of $I_p + 284$ eV. The harmonics above $149\omega_0$ ($\sim$ 232 eV) form a supercontinuum with a bandwidth of about 107 eV. However, the intensity of harmonics in the supercontinuum is about 2–6 orders of magnitude lower than that of harmonics in the first plateau. In addition, the supercontinuum shows a strongly modulated structure. This can be understood as follows: each harmonic in the supercontinuum region originates from two dominant quantum paths (the long and short paths) in the two-color case, and the contributions from the long and short paths to the harmonic generation are almost comparative. Therefore, the interference between these two quantum paths brings about an obvious modulated structure in the supercontinuum. The above results show that, in our two-color case, single quantum path selection and isolated attosecond pulse generation can not be achieved. This aim can be realized by adding a static field to the synthesized two-color field. The harmonic spectrum in the presence of the static field is shown by the solid curve in Fig. 1(a). It can be seen that the spectral cutoff is significantly extended to $423\omega_0$, which is far larger than that in the two-color field. Moreover, the harmonics higher than $172\omega_0$ (267 eV) are regular and continuous, forming a more favorable ultrabroad XUV supercontinuum with a bandwidth of about 389 eV. Note that the photon energy of the generated supercontinuum covers all over the water window spectral region (284–543 eV), which paves the way to some significant applications in water. In addition, the modulation in the supercontinuum nearly disappears, implying a single quantum path is picked. We also note the harmonic intensity of the generated broadband XUV supercontinuum is effectively enhanced by 2–5 orders of magnitude compared with the two-color case. With such a broad and strong XUV supercontinuum, an intense ultrashort isolated attosecond pulse can be produced directly.

In Fig. 1(b), the time-frequency distribution of the HHG spectrum corresponding to the solid curve in Fig. 1(a) is presented in order to well understand the spectral structure. As shown in this figure, there are two main peaks contributing to the harmonics, which are marked as A and B, respectively. The maximal harmonic orders of peaks A and B are about 172 and 423, respectively. The intensities of peaks A and B are comparable, and the harmonics above $172\omega_0$ are only contributed by the peak B. Thus an ultrabroad XUV supercontinuum with a bandwidth of 389 eV is formed, which is consistent with the solid curve in Fig. 1(a). Additionally, one can clearly see that for the supercontinuum there is only a dominant quantum path contributing to the harmonic generation, which implies that the harmonics beyond the 172nd order are almost emitted once, i.e., these harmonics are well phase-locked. For harmonics lower than 172nd order, the interference between peaks A and B leads to an irregular spectral structure. Considering all the results above, we come to a conclusion that, by using a static field, a broadband XUV supercontinuum with a single quantum path and high conversion efficiency is produced, which can support the generation of an intense ultrashort isolated attosecond pulse.

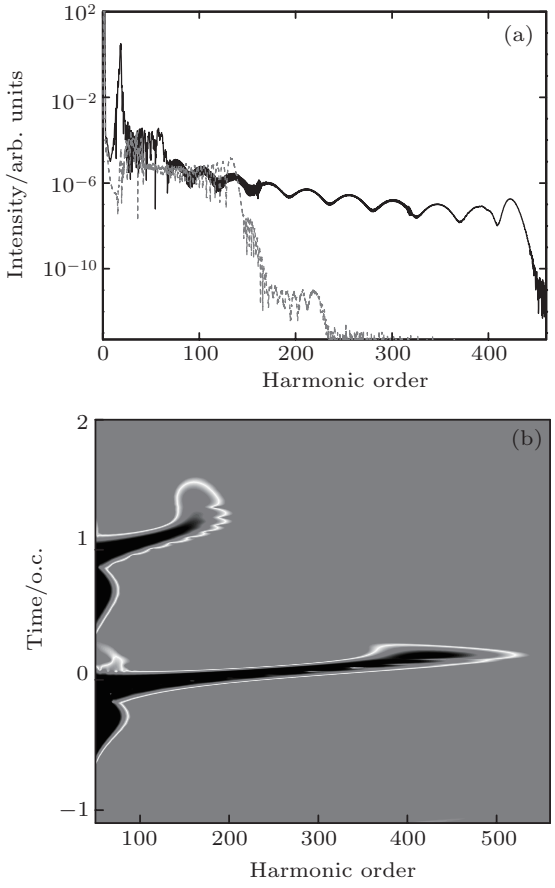


Fig. 1. (a) Harmonic spectra from He^+ in the two-color field synthesized by a 5 fs/800 nm fundamental field and its second harmonic field (dotted curve), and in the combined field of the synthesized two-color pulse and a static electric field (solid curve). (b) Time-frequency distribution of the HHG spectrum corresponding to the solid curve in Fig. 1(a). The intensities of the fundamental, SH, and static fields are 1.3×10^{15}, 1.0×10^{14}, and 1.0×10^{14} W/cm^2, respectively. The relative phase ϕ is chosen to be 0.

In order to well understand the underlying physics of both the extension and enhancement of the HHG, as

well as the formation of the ultrabroad XUV supercontinuum, we perform a classical trajectory simulation using the three-step model and calculate the ionization probability in the two optical fields with $\phi = 0$, respectively. The results are shown in Fig. 2. Figure 2(a) presents the electric field of the two-color laser pulse. Figure 2(b) shows the dependence of the kinetic energy $E_{\rm k}$ on the ionization and emission times. As shown in this figure, in the two-color case, the electrons are mainly ionized near the peaks of the laser field, which are marked as A, B, and C, respectively. The highest kinetic energies of electrons ionized near the peaks A, B, and C are respectively 177, 280, and 148 eV, corresponding to harmonic energies $I_{\rm p} + 177$, $I_{\rm p}+280$, and $I_{\rm p}+148$ eV. $I_{\rm p}+148$ eV is well consistent with the cutoff energy of the first plateau ($131\omega_0$), and $I_{\rm p} + 280$ eV roughly accords with the cutoff energy of the second plateau ($218\omega_0$). Moreover, one can see that the harmonic photons with energy higher than $I_{\rm p} + 177$ eV are attributed to two electron paths. The path with earlier ionization time but later emission time is called the long path, and the path with later ionization time but earlier emission time is called the short path. As the kinetic energy increases, the emission time of the short path increases, and that of the long path decreases. Finally the emission times of two paths are equal for the maximum kinetic energy. This result indicates that the harmonics in the supercontinuum for the two-color case are not synchronized. Hence, the superposition of several harmonics will result in two radiation pulses in every half cycle of the fundamental pulse. Although we obtain an XUV supercontinuum with a bandwidth of about 103 eV, the efficiency of the supercontinuum is low, leading to attosecond pulses with low intensity, which limits their application. This is because the ionization of electrons ionized near peak B, from which the supercontinuum originates, takes place slowly (see the dotted curve in Fig. 2(a)). Thus, the harmonic yield of the supercontinuum is comparatively inefficient owing to the fact that the harmonic efficiency is mainly determined by the ionization rate according to the three-step model.

We expect that, under the influence of a static electric field, the quantum paths of HHG in the synthesized two-color field can be significantly modulated and the ionization yield of electrons contributing to the harmonic emission can be obviously improved. The solid curve in Fig. 2(c) shows the electric field of the combined field. Figure 2(d) shows the dependence of the kinetic energy $E_{\rm k}$ on the ionization (gray diamonds) and emission times (black circles) in the combined field. From Figs. 2(c) and 2(d), one can see that there are two major ionization processes (marked as A and B), which indicates that the HHG mainly comes from the contributions of the processes A and B. The highest kinetic energy that on electron gains from the driving field is raised to 593 eV, corresponding to a cutoff energy of $I_{\rm p} + 593$ eV. Since the highest kinetic energy of an electron in the presence of the static field is much more higher than that in the synthesized two-color field, the spectral cutoff for the case of the combined field is greatly extended compared with the two-color case. However, the second highest kinetic energy of electrons only reaches 213 eV, corresponding to the harmonic photon with energy of $I_{\rm p}+213$ eV. In comparison with the case of two-color field, the energy difference between peaks B and A becomes much larger. The larger energy difference is the reason why an ultrabroad XUV supercontinuum spectrum can be produced in the presence of a static electric field. Furthermore, when the static electric field is added to the two-color field, the time variation of the ionization yield of electrons is different. The corresponding result is given by a dotted curve in Fig. 2(c), which shows that after about -1.0 optical cycle (o.c.) the ionization probability is much larger than that in the two-color field, implying that the ionization yield of electrons is significantly enhanced. In our scheme, the HHG, especially the supercontinuum part, is mainly attributed to the ionized electrons in the time range from -0.72 to -0.17 o.c., while the ionization probability in this time range is higher than that in the time range from -0.53 to -0.22 o.c. for the two-color case, where the supercontinuum appears. Therefore, the harmonic efficiency of the supercontinuum in the presence of a static field is greatly improved compared with the two-color case. It should be noted that the long path of the peak B in Fig. 2(d) is removed and only the short path is selected after the addition of the static field. Once a single quantum path is selected, an isolated short attosecond pulse can be generated directly. The above results are well consistent with that derived from the time-frequency analysis as shown in Fig. 1(b).

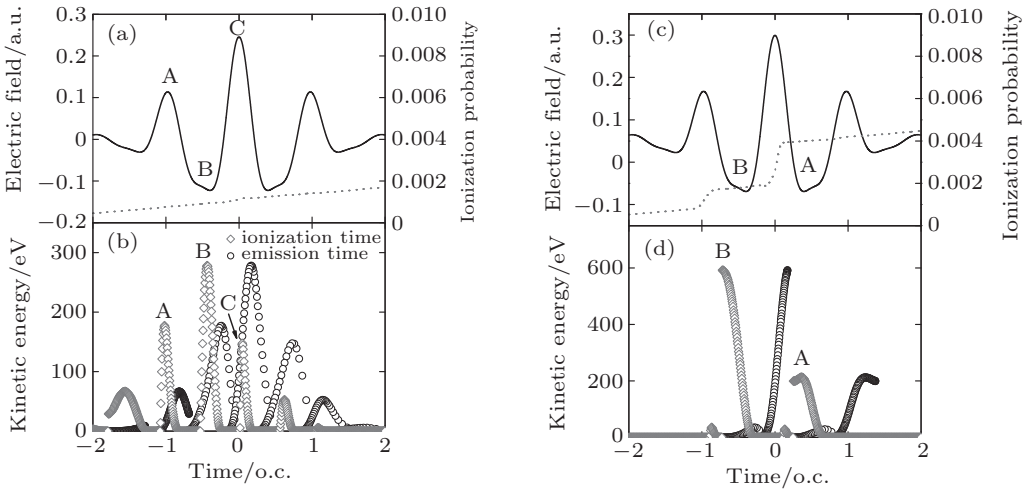


Fig. 2. (a) Electric field (solid curve) and time dependence of the ionization probability (dotted curve) in the synthesized two-color pulse with $\phi = 0$. (b) Dependence of the kinetic energy E_k on the ionization (gray diamonds) and emission times (black circles) in the field shown in Fig. 2(a). (c) Electric field (solid curve) and time dependence of the ionization probability (dotted curve) in the combined field ($\phi = 0$). (d) Dependence of the kinetic energy E_k on the ionization (gray diamonds) and emission times (black circles) in the field shown in Fig. 2(c). The laser parameters are the same as in Fig. 1.

According to the above analysis, under the influence of a static electric field, we realize the selection of the short path and produce an ultrabroad XUV supercontinuum with high conversion efficiency, which powerfully supports us in generating an intense short isolated attosecond pulse. In order to check this viewpoint, we investigate the attosecond pulse generation in the combined field. For comparison, the temporal profile of the attosecond pulse generated by the two-color field is also presented in Fig. 3(a). As shown in this figure, two attosecond bursts are produced by superposing the harmonics from 180th to 220th order. This is because that each harmonic in the supercontinuum (see the dotted curve in Fig. 1(a)) has two different emission times, and the harmonics are not locked in phase. As shown in Fig. 3(a), the weak 68-as pulse originates from the short path, the intense 59-as pulse originates from the long one. Figure 3(b) shows the temporal profile of the attosecond pulse in the combined field. By superposing the harmonics from the 190th to 300th order, an ultrashort isolated 26-as pulse with a bandwidth of 170.5 eV is directly obtained without phase compensation, which is also very close to the value predicted by the formula $\Delta T = T_0/N$.[32] Compared with the case of two-color field, the intensity of the generated isolated attosecond pulse is at least 5 orders of magnitude higher. The isolated 26-as pulse contains only 2.4 optical cycles of the central wavelength of 2.8 nm. Such a few-cycle isolated attosecond pulse allows one to study and manipulate the electronic dynamics in atoms and molecules when the processes are triggered by the electric field of the attosecond pulse rather than by the intensity profile.

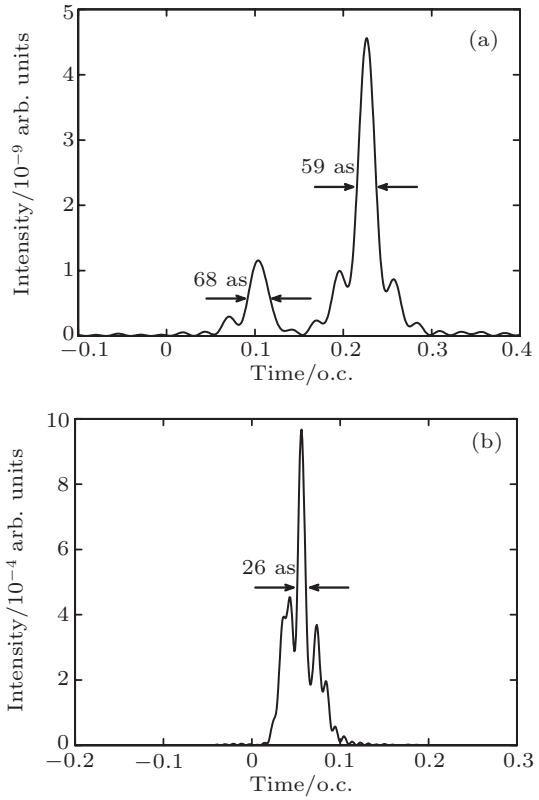


Fig. 3. (a) Temporal profile of the attosecond pulse by superposing the harmonics from 180th to 220th order in the synthesized two-color pulse. (b) Temporal profile of the attosecond pulse by superposing the harmonics from 190th to 300th order in the combined field. The laser parameters are the same as in Fig. 1.

It should be noted that, in our simulation, we find

that both the supercontinuum and the harmonic cutoff can be extended when the relative phase ϕ between two laser pulses changes from 0 to 0.79π, and isolated sub-30-as pulses are still obtained by superposing well-selected continuous harmonics. The intensity of the strongest subpeak is less than half of the main peak intensity. To verify this fact, we investigate the HHG and attosecond pulse generation in the combined field with $\phi = 0.3\pi$ and $\phi = 0.6\pi$, respectively, and the calculated results are shown in Fig. 4. Because the difference of the harmonic intensity in the plateau region is actually slight, we shift two curves for the cases of $\phi = 0.3\pi$ and $\phi = 0.6\pi$ so that the harmonic signals for $\phi = 0.3\pi$ and $\phi = 0.6\pi$ are scaled. The dashed gray and dotted gray curves in Fig. 4(a) show the harmonic spectra for the cases of $\phi = 0.3\pi$ and $\phi = 0.6\pi$, respectively. For comparison, the harmonic spectrum for the case of $\phi = 0$ is also presented by a solid curve in Fig. 4(a). For the case of $\phi = 0.3\pi$, the harmonics from the 182nd to the 458th are regular and continuous, corresponding to the supercontinuum with a bandwidth of about 428 eV, which is 39 eV larger than that in the case of $\phi = 0$. For the case of $\phi = 0.6\pi$, the supercontinuum ranges from the 185th to the 460th order, cover a bandwidth of 427 eV, which is superior to that in the case of $\phi = 0$. It is worth noting that the spectrum cutoff for $\phi = 0.46\pi$ is the highest. Furthermore, from Fig. 4(a), one can see that the modulation in the supercontinuum becomes stronger and stronger with the relative phase increasing, which has little effect on the generation of sub-30-as pulses. Next, we study the isolated attosecond pulse generation in cases of $\phi = 0.3\pi$ and $\phi = 0.6\pi$. Figure 4(b) presents the temporal profile of the attosecond pulse via superposing 112 order harmonics from 200th to 312nd in the supercontinuum at $\phi = 0.3\pi$. One can clearly see that a clean isolated 26-as pulse with a bandwidth of 173.7 eV is produced directly. Figure 4(c) presents the temporal profile of the attosecond pulse by superposing the harmonics from 215th to 315th order at $\phi = 0.6\pi$, an isolated 26-as pulse with a bandwidth of 155.1 eV is obtained.

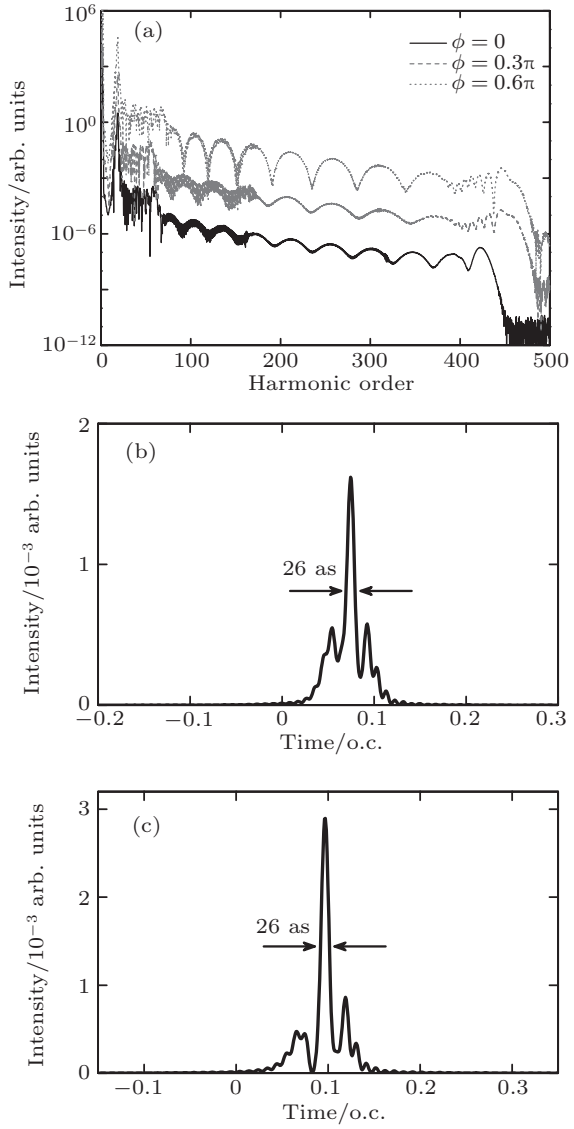


Fig. 4. (a) Harmonic spectra from He^+ in the combined field with the relative phase $\phi = 0$ (solid curve), $\phi = 0.3\pi$ (dashed gray curve), and $\phi = 0.6\pi$ (dotted gray curve), respectively. For the purpose of clarity, the harmonic intensities for $\phi = 0.3\pi$ and $\phi = 0.6\pi$ are multiplied by factors of 10^2 and 10^4, respectively. (b) Temporal profile of the attosecond pulse by superposing the harmonics from the 200th to the 312nd order for $\phi = 0.3\pi$. (c) Temporal profile of the attosecond pulse by superposing the harmonics from the 215th to the 315th order for $\phi = 0.6\pi$. Other laser parameters are the same as in Fig. 1.

Finally, we also investigate the influence of fluctuation of the laser intensity on the generation of the supercontinuum and broadband isolated sub-30-as pulse. According to our calculation, the supercontinuum and broadband isolated sub-30-as pulse can be produced by superposing some well-chosen harmonics for the fundamental pulse with an intensity ranging from 9.2×10^{14} to 1.8×10^{15} W/cm^2. For the static field, when its intensity changes from 8.2×10^{13} to 1.36×10^{14} W/cm^2, an isolated sub-30-as pulse and a supercontinuum with a bandwidth of larger than 300 eV can be generated, and the harmonic cutoff is higher than the 400th order. In our method, we also consider how long He^+ can survive in the static field alone. From our calculation, the ionization probability of He^+ is lower than 0.1 when the duration of the static field is not longer than 1 ps. In addition, our results are still obtained when the intensity of the SH pulse varies from 0 to 2.4×10^{14} W/cm^2. Though the

SH laser pulse has a wide range of intensity to adjust, this does not mean that the SH laser pulse is not important. On the one hand, the SH pulse can enlarge the intense difference between the strongest electric field peak and the electric field peak prior to it, and thus the electron ionized near the peak prior to the strongest one will obtain the highest kinetic energy from the laser field because the ionized electron experiences the longest accelerating time, which results in higher cutoff of the HHG. On the other hand, the harmonic spectrum in the presence of the SH pulse is more regular and smoother than that without it, which is beneficial to the generation of a shorter isolated attosecond pulse with more clean temporal profile. To confirm our judgement, we also present the harmonic spectra for different intensities of the SH pulse, i.e., 0, 1.0×10^{14}, and 2.4×10^{14} W/cm^2. The results are shown in Fig. 5. As we expect, the harmonic spectrum becomes smoother and the spectrum cutoff becomes higher with the increase of the intensity of the SH pulse, which best support our point of view.

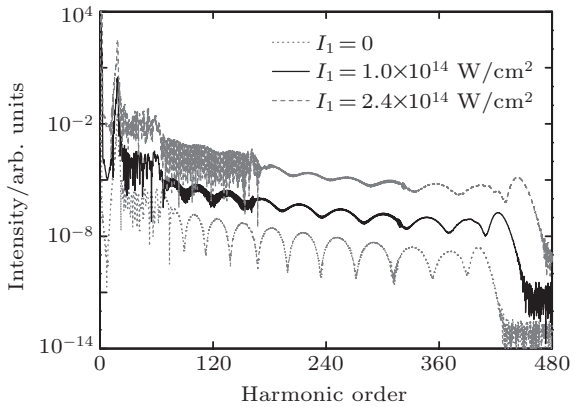


Fig. 5. Harmonic spectra for three different intensities of the SH laser pulse: 0, 1.0×10^{14} W/cm^2, and 2.4×10^{14} W/cm^2. For the purpose of clarity, the harmonic intensities in the cases of 0 and 2.4×10^{14} W/cm^2 are multiplied by factors of 10^{-2} and 10^2, respectively. Other laser parameters are the same as in Fig. 1.

4. Conclusion

In conclusion, we theoretically present an efficient method to generate an ultrashort isolated attosecond pulse by combining a synthesized two-color pulse and a static electric field. It is shown that, in the presence of the static electric field, the enhancement of the ionization yield and the control of quantum paths are both achieved. As a consequence, the bandwidth of the supercontinuum is significantly extended to 389 eV, and the efficiency of the supercontinuum is effectively enhanced by 2–5 orders of magnitude compared with that in the absence of the static field. The low harmonic efficiency in the absence of the static field is due to the intrinsic dynamic property of the corresponding electronic wave packet and the narrow bandwidth. Moreover, since the continuous harmonics cover an extremely broad bandwidth and are almost synchronically emitted at once, an ultrashort isolated 26-as pulse is directly obtained. Finally, we also analyse the influence of the laser parameters including the relative phase and laser intensity on the ultrabroad supercontinuum and isolated sub-30-as pulse generation. We also give the value range of the corresponding laser parameter. Presently, a 5-fs, 800-nm laser pulse is available in a good few laboratories, and a 5-fs, 400-nm laser pulse can be obtained from a BBO crystal. The static field strength in this paper is so high that it can hardly be experimentally realized nowadays. However, it has been shown that a CO_2 laser field or a strong terahertz field can work instead of a static field.[28,29,33,34] Our calculations also indicate that, by adding a 100-μm THz field to the synthesized two-color field, similar results are obtained. Overall, the scheme presented here appears to be feasible for experimental demonstration in the near future. The advantages of our scheme lie in producing an ultrabroad XUV supercontinuum spectrum and maintaining a high conversion efficiency, which can be used for the production of an intense ultrashort isolated attosecond pulse.

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