Robust generation of isolated attosecond pulse against the variation of carrier envelope phase of driving laser pulses

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We propose a scheme for generating isolated attosecond pulse (IAP) via high-order harmonic generation in gases using a chirped two-color laser field of multicycle duration. In contrast to previous techniques where the stable carrier-envelope phase (CEP) of the driving laser pulses is a prerequisite for IAP generation, the proposed scheme is robust against the large variation of CEP. We show the generation of IAP with an intensity fluctuation less than 50% and an intensity contrast ratio higher than 5:1 when the CEP shift is as large as 1.35π.

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I. INTRODUCTION

The attosecond pulses produced via high-order harmonic generation (HHG) in atoms and molecules is important for probing and controlling of ultrafast electronic dynamics. Light pulses with a pulse duration close to 100 as have been demonstrated either as an attosecond pulse train (APT) with a repetition period of half the driving laser cycle [1] or as an isolated attosecond pulse (IAP) [2,3]. The process of HHG is well understood in terms of the semiclassical three-step model [4], strong field approximation (SFA) [5], and Feynman's path-integral approach in a quantum-mechanical theory frame [6]. The IAP can be produced only if one is able to isolate the HHG emission within half a cycle of the driving laser field.

By using few-cycle laser pulses with stable carrier-envelope-phase (CEP), different techniques have been demonstrated for the generation of IAP in the extreme ultraviolet (xuv) spectral region [2,3,7–10]. The driving laser field can be shaped precisely in the subcycle scale by mixing a controllable second-color laser pulse to the fundamental driving pulse, and the electron motion as well as HHG can be controlled consequently in the two-color field in the light of the three-step model. Several methods were proposed to generate sub-100 as IAP's by mixing a second harmonic (SH) pulse to a few-cycle fundamental laser pulse [11–14]. More recently, the generation of IAP by using multicycle driving laser pulses was also reported [15–17]. However, all of these schemes [8–17] require state-of-art CEP-stabilized laser facilities.

In this article we propose a scheme to overcome the vital requirement of CEP stabilization in the generation of IAP's by using a chirped driving laser field with multioptical-cycle duration. We calculate the HHG in helium by the numerical solution of a single-active-electron (SAE) model with strong-field approximation (SFA). We demonstrate that the generation of IAP is insensitive to the CEP variation when a chirped two-color laser field is used. To generate the required chirp for the driving laser field, we consider the chirp due to the self-phase modulation (SPM) induced in a fused silica plate placed in the path of the laser beams, which can be generated and controlled properly and easily by adjusting the laser intensity and propagation length (silica plate thickness).

II. MODEL AND NUMERICAL METHOD

We calculated the response a helium atom exposed to a synthesized laser field consisting of an 1800 nm/50 fs laser pulse and a 900 nm/40 fs pulse. The method of HHG calculation was described in detail in Ref. [18], which is based on the SAE model and has been widely used. The atomic dipole moment is calculated using Eq. (34) in Ref. [18] and the harmonic spectrum is obtained by Fourier transforming the time-dependent dipole moment. Both of the two laser pulses' peak intensities at the focus where HHG occurs are assumed to be 4.0×10^{13} W/cm^2. The driving laser field has the form

$$E_s(t) = E_1 \exp\left[-2\ln(2)t^2/\tau_1^2\right]\cos[\omega_1 t + (2\pi/\lambda_1)n_2 I_1(t)L + \delta_{\mathrm{CEP}}] + E_2 \exp\left[-2\ln(2)(t+t_0)^2/\tau_2^2\right] \times \cos[\omega_2(t+t_0) + (2\pi/\lambda_2)n_2 I_2(t+t_0)L + 2\delta_{\mathrm{CEP}}], \quad (1)$$

where E_i, ω_i, λ_i, δ_{CEP}, and τ_i $(i = 1,2)$ are the amplitude, frequency, wavelength, CEP, and the full-width-at-half-maximum (FWHM) pulse duration for the two-color laser pulses, respectively. t_0 denotes the relative time delay between the two pulses.

As we know, if the phase change $\varphi(t)$ of laser pulses is induced only by the SPM, it is proportional to the product of nonlinear refractive index n_2 (measured in square centimeters per watt) and the laser intensity $I(t)$ (measured in watts per square centimeter) and has the form of $\varphi(t) = (2\pi/\lambda)n_2 I(t)L$, where L is the thickness of the fused silica plate and λ is laser wavelength [19]. $I_1(t)$ and $I_2(t+t_0)$ are the intensities of the two-color pulses during the propagation in the fused silica plate, which are assumed to be 5.66×10^{12} and 4.91×10^{12} W/cm^2, respectively. In our calculations, we assumed that both of the n_2 in the fused silica plate for 1800 and 900 nm laser pulses are $n_2 = 2.5 \times 10^{-16}$ cm^2/W [19,20]. We neglect here and in the following part a possible spatial variation of the induced SPM owing to radial nonuniformity of the laser intensity. Such an assumption relies on the possibility to enlarge the beam size and then select only its central part [16].

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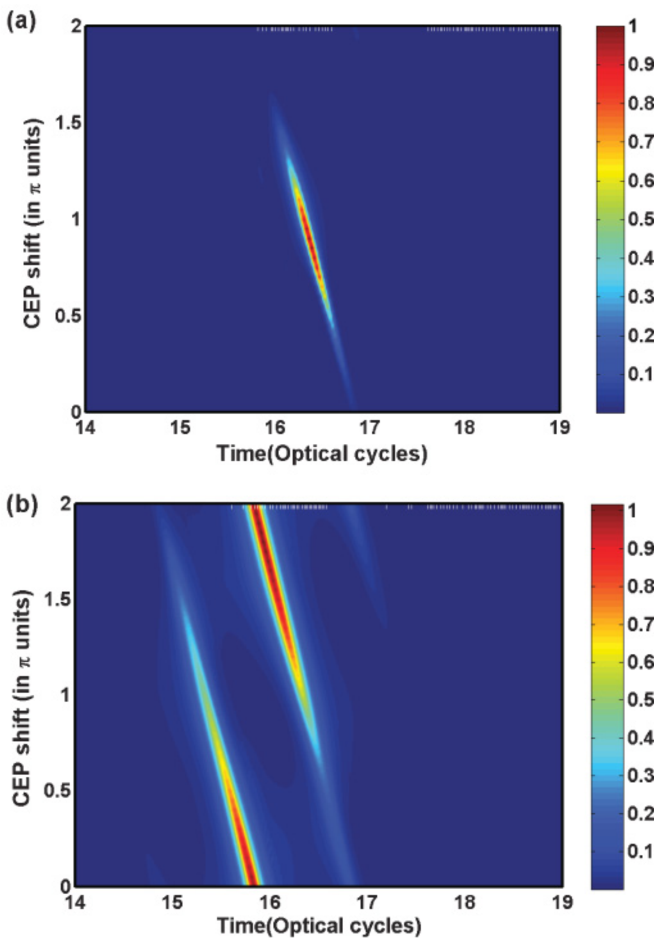

FIG. 1. (Color online) Temporal intensity profiles of the harmonic emission as functions of CEP, for the harmonic emission (a) in the range of 97–117 eV generated in the chirped laser field and (b) in the range of 77–97 eV generated in the chirp-free laser field. The time delay between the two-color pulses as 5.79 fs and the thickness of the fused silica plate is 1.0 mm.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the calculated temporal intensity profiles for the cutoff range of HHG in helium driven by the chirped and chirp-free [$\varphi(t) = 0$] two-color field with a time delay of 5.79 fs between the two pulses, as functions of CEP from 0 to 2π. Only the harmonic emission near the cutoff with a bandwidth of 20 eV is selected (i.e., from 97 to 117 eV in the chirped field case and from 77 to 97 eV in the chirp-free field case). The silica plate thickness is 1.0 mm. Note that in the chirped field case the cutoff of harmonic spectrum extends to a higher-energy region in comparison with the chirp-free field case.

One can see a dramatic difference in the temporal intensity profiles as functions of CEP for the chirped field and chirp-free field cases. As shown in Fig. 1(a) a clean IAP is generated for a wide range of CEP shift in the chirped field case. However, in the chirp-free field, as shown in Fig. 1(b), IAP can be generated with some CEP values around $0(2\pi)$, while two attosecond pulses with different intensities can be found for most of the CEP values, which is consistent with the previously reported sensitive effect of CEP on IAP generation [21].

Previous works have shown that CEP stabilization is a strict prerequisite of stable IAP generation [21]. According to the three-step model of HHG, if the selected harmonic emission is confined to a single revisit of the mother core by a freed electron, a single burst of xuv emission in time domain and supercontinuum emission in spectral domain can be observed. For a less than two cycle driving laser field, the xuv emission occurs only during a small fraction of the optical cycle of the driving laser field with the largest amplitude. For a multicycle driving laser field, IAP can also be generated by the synthesized field with a dedicated temporal shape with which the electronic trajectories are properly controlled [13,14]. Without CEP stabilization, the synthesized field cannot have a stable temporal shape from shot to shot, and therefore does not warrant a stable IAP generation.

We find that, however, the CEP variation does not affect the effective generation of the IAP when the multicycle driving filed is properly chirped, although there are some fluctuations in the intensity and pulse width of IAP and the absolute timing of IAP with respect to the driving laser field, as shown in Fig. 1(a). The underlying physics for the robust generation of IAP against the CEP variation in the chirped laser field is as follows. Due to different chirps for the leading edge and tailing edge of the driving laser pulses, which are introduced via the laser-intensity-related SPM in the fused silica plate, and by adjusting the time delay between the two pulses, the periodicity of the driving laser field in most of the optical cycles is destroyed and only one suitable trajectory for IAP generation of the freed electron can be selected by photon energy filtering. When the CEP is changed, the synthesized electric field slips under the multicycle envelope, the timing of produced IAP also slips due to the shift in the electric field, but it is always an isolated emission for most of the CEP values. This point can be illustrated in detail by performing a time-frequency analysis.

We performed the time-frequency analysis by means of a wavelet transform of the induced dipole response of the He atom to the laser fields [22,23], as shown in Fig. 2. The parameters are the same as those in Fig. 1. In Figs. 2(a) and 2(b) for the chirp-free laser field case, two attosecond pulses with comparable intensity appear at $\delta_{\mathrm{CEP}} = 0.4\pi$, while one main pulse and two weak pulses appear at $\delta_{\mathrm{CEP}} = 1.3\pi$, respectively. When the driving laser pulses is chirped properly, the emission of satellite attosecond pulses are suppressed successfully with only one attosecond pulse dominating (the harmonic emission from 97 to 117 eV) as shown in Figs. 2(c) and 2(d) for either $\delta_{\mathrm{CEP}} = 0.4\pi$ or $\delta_{\mathrm{CEP}} = 1.3\pi$, respectively, in agreement with the results shown in Fig. 1(a).

Although the robust generation of IAP against CEP shift has been demonstrated with a chirped laser field, the intensity and pulse width of the IAP changes as CEP varies, as shown in Fig. 1(a). Figure 3 shows the peak intensity and pulse duration (FWHM) of the IAP as functions of CEP. We define the CEP range for the IAP intensity variation from the peak value to its 50% as CEP width. The wider the CEP width the more stable the IAP intensity against CEP variation is. For the same parameters as those for Fig. 1(a) where the silica plate thickness is 1.0 mm, the CEP width is 0.65π (from 0.55π to 1.2π). Because

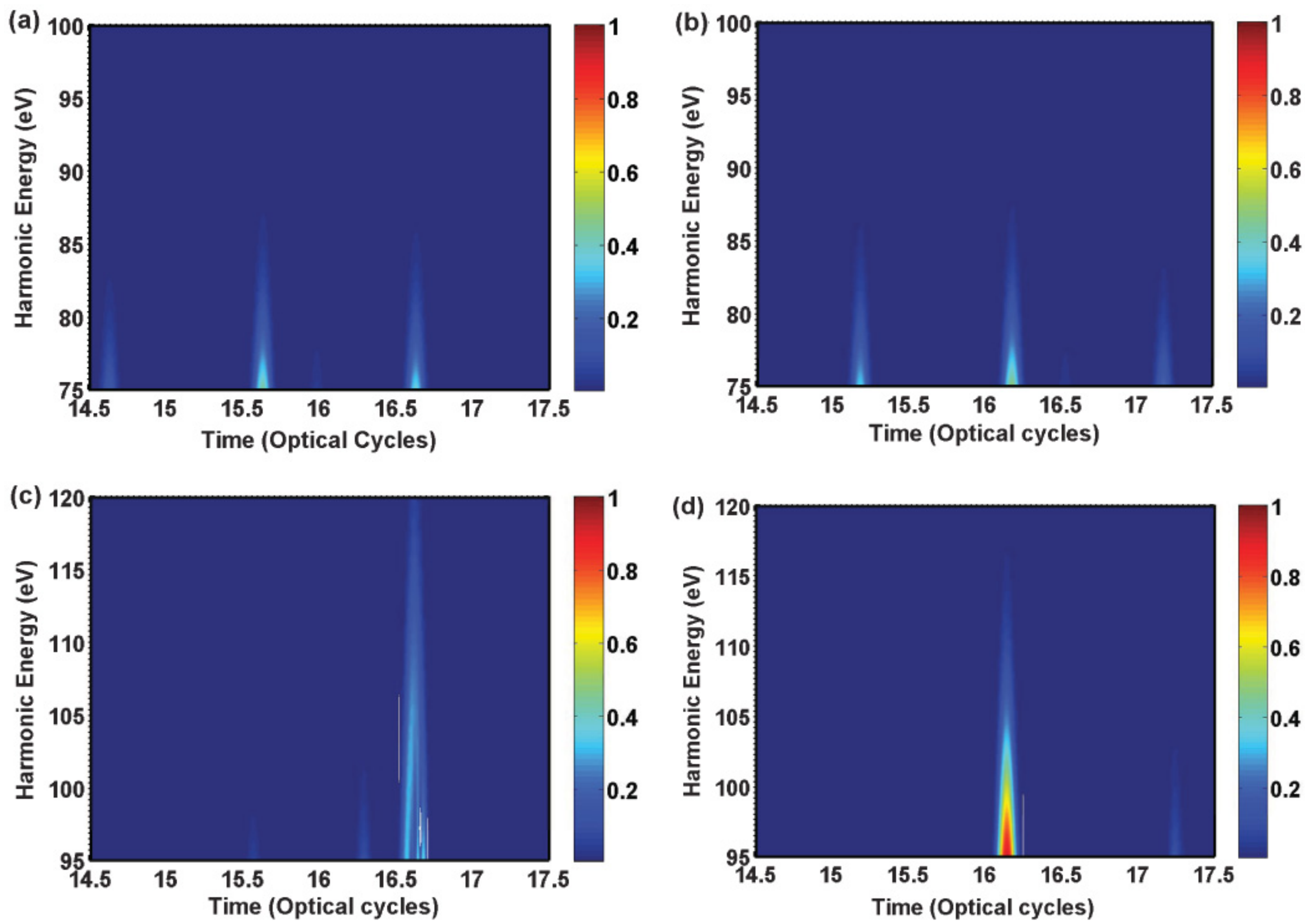

FIG. 2. (Color online) Diagrams of the time-frequency distribution for harmonic emission produced by the (a,b) chirp-free and (c,d) chirped driving laser field. The CEP values are (a,c) $\delta_{\mathrm{CEP}} = 0.4\pi$ and (b,d) $\delta_{\mathrm{CEP}} = 1.3\pi$, respectively. The laser parameters and the silica plate thickness are the same as those in Fig. 1.

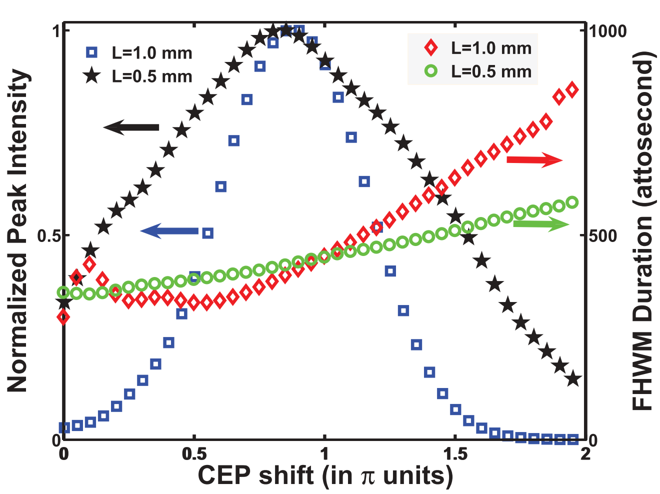

FIG. 3. (Color online) The intensity and pulse width of IAP as functions of CEP when the thickness of the fused silica plate is 0.5 mm (stars and open circles) and 1.0 mm (squares and open diamonds), respectively. The IAP corresponds to the harmonic emission from 97 to 117 eV produced in the chirped laser field. Note that the time delay between the two-color pulses is 4.98 fs for $L = 0.5$ mm and 5.79 fs for $L = 1.0$ mm, respectively.

we use the propagation in a fused silica plate to introduce the controllable chirp for the driving laser pulses, we can adjust the CEP width by changing the thickness of fused silica plate. We show also in Fig. 3 the results for a 0.5-mm thick silica plate and in this case the optimized time delay between the two-color pulses is 4.98 fs. Now we obtain a doubled CEP width as 1.4π (0.15π–1.55π)and therefore the IAP intensity is less changed with the variation of CEP. The variation of IAP pulse width at different CEP values is also significantly reduced when the thickness of the silica plate is 0.5 mm.

Another issue for a clean IAP generation is that the intensity contrast ratio between the main attosecond pulse and the weak satellite attosecond pulses should be as high as possible. As shown in the left panel of Fig. 4, the temporal intensity profile of IAP generated in the chirped two-color field as a function of CEP follows a 2π periodic structure. We find that there are some CEP values at which double attosecond pulses of comparable intensity appear, for example, when CEP is in the range from 1.65π to 1.825π as shown in the right panel of Fig. 4, which cannot be distinguished in Fig. 1(a) due to the limited dynamic range of the color image.

The intensity contrast ratio of the two attosecond pulses (i.e., IAP1 and IAP2) changes with the CEP. A high-contrast

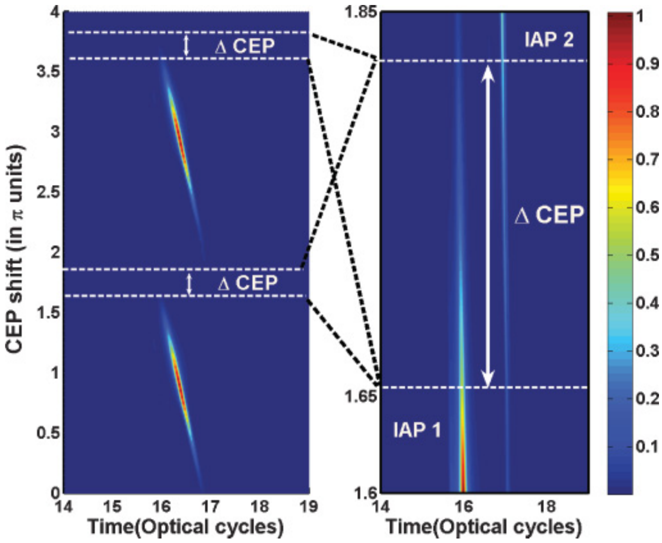

FIG. 4. (Color online) Left: Temporal intensity profile of the IAP as a function of CEP (from 0 to 4π), showing a 2π periodic structure. Right: The enlargement of the part obtained with CEP from 1.6π to 1.85π. The laser parameters and the silica plate thickness are the same as those in Fig. 1.

ratio is desired for the generation of clean IAP. We define the CEP range where the intensity ratio of IAP1 and IAP2 changes from 1:5 to 5:1 as ΔCEP. Note that $\Delta\text{CEP} = 0.175\pi$ for the parameters in the right panel of Fig. 4.

To make the CEP shift suitable for generating a clean IAP as large as possible, we need to minimize ΔCEP by adjusting the thickness of the fused silica plate. Figure 5 shows ΔCEP as a function of the thickness of the silica plate. The minimum value of ΔCEP is 0.09π when the thickness of the silica plate is 1.3 mm, after the time delay between the two pulses is optimized for each thickness in the range from 0.4 to 2.0 mm. However, at this thickness, the CEP width is reduced to 0.5π. For comparison, we show in Fig. 5 also the CEP width as a function of the thickness of the silica plate. A smaller chirp due to a thinner silica plate is good for broadening the CEP width, but the chirp becomes not enough to suppress the emission of satellite attosecond pulses. Therefore, a compromise between CEP width and ΔCEP should be made. At the best thickness of 0.5 mm for the silica plate under the current laser conditions, we can generate the IAP with an intensity fluctuation less than 50% and an intensity contrast ratio higher than 5:1 when the CEP shift is as large as 1.35π.

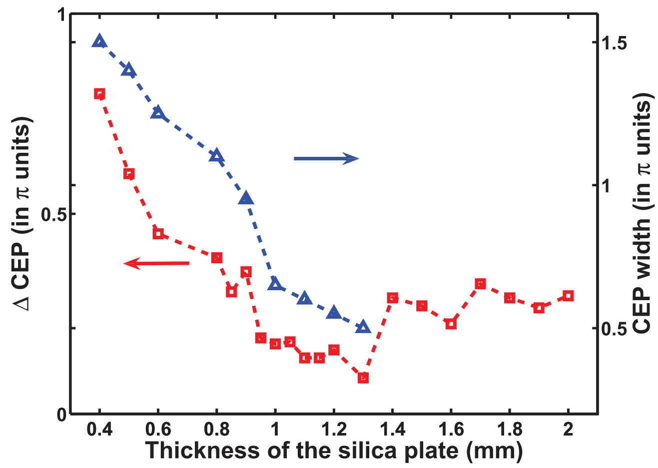

FIG. 5. (Color online) ΔCEP and CEP width as functions of the thickness of the silica plate, calculated for the IAP corresponding to the harmonic emission from 97 to 117 eV produced in the chirped laser field. Note that the time delay between the two pulses is optimized for each value of the silica plate thickness.

IV. CONCLUSION

In summary, we investigated the IAP generation in helium atoms subjected to a chirped two-color laser field of multicycle duration. We have found the specific conditions under which the generation of the IAP is robust against the CEP variation of the driving laser pulses. Therefore the CEP stabilization of the driving laser pulses is no longer a strict prerequisite of IAP generation. Although the intensity and pulse width of IAP and the absolute timing of IAP do change when CEP is shifting considerably, the requirement of IAP generation on CEP stabilization is greatly relieved by using the proposed scheme. This is very important for the generation and application of xuv IAP. The proposed scheme is easy to implement experimentally by using a typical ultrafast laser facility in most laboratories and therefore represents a crucial step to enable the access to IAP for a wide scientific community.

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