Isolated attosecond pulse generation from atom radiated by a three-color laser pulse*

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We theoretically investigate high-order harmonic and attosecond pulse generation from helium atom in a three-color laser field, which is synthesized by 10 fs/800 nm Ti-sapphire laser and a two-color field consisting of 30 fs/532 nm and 30 fs/1330 nm pulses. Compared with harmonic spectrum generated by a monochromatic field, the harmonics generated from the synthesized three-color field show a supercontinuum spectrum with a bandwidth of 235 eV, ranging from the 154th to the 306th order harmonic. This phenomenon can be attributed to the fact that the ionization of atoms as well as motion of ionized electron can be effectively controlled in the three-color field. Therefore, an isolated 46-as pulse can be generated by superposing supercontinuum from the 160th to the 210th order harmonics.

Keywords: three-color laser field, high-order harmonic, isolated attosecond pulse

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1. Introduction

The attosecond pulse allows one trace and detect electronic motions in atoms, molecules and condensed matters,[1–3] so many attempts have been made to obtain attosecond pulses. So far, the high-order harmonic generation (HHG) is the only way to achieve attosecond light pulses in experiment due to its particular plateau structure.

When atoms, molecules, or clusters are irradiated by an intense laser pulse, high-order harmonics of the laser frequency can be generated. The HHG process can be well explained by the semi-classical three-step model proposed by Corkum in 1993.[4] The electron is firstly ionized under the action of the external field, then oscillates almost freely with the field and is driven back to its ion when the laser field reverses, finally comes back to its initial state and simultaneously emits a harmonic photon whose energy is equal to the ionization energy of atom plus the energy obtained from the laser field. This model successfully explains the cut-off rule of the atomic high-order harmonic spectra: $E_{\rm cutoff} = I_{\rm p} + 3.17U_{\rm p}$, here $I_{\rm p}$ and $U_{\rm p} = E^2/4\omega^2$ are the ionization energy and the pondermotive energy of atom, respectively.

In obtaining attosecond pulses by using an HHG source, the generation of isolated attosecond pulses attracts more attention due to its wide application in the pump-probe experiments. So far, the isolated 80-as and 67-as short pulses have been successfully obtained in experiments by adopting the few-cycle scheme[5] and the double optical gating technique, respectively,[6] but the intensities of the pulse obtained are low. Further research will focus on the generation of the isolated attosecond pulse with higher intensity and shorter pulse duration. The two-color or three-color field schemes are proposed theoretically to obtain optimized isolated attosecond pulses by extending the width of supercontinuum harmonic spectra. Zeng *et al*. obtained an isolated 65-as pulse by imposing a second harmonic pulse on a fundamental pulse.[7] Lan *et al*. generated 135-as pulses with the "ionization gating" scheme, in which the electrons are ionized within an interval of 270 as.[8] Compared with the two-color field case, trajectory of electron motion can be further controlled in the three-color field scheme, which provides an effective way for the generation of the isolated attosecond pulse. Chen *et al*. obtained an isolated 38-as pulse by imposing a 27th-order harmonic with 1 fs pulse duration on the two-color laser field consisting of 5 fs/800 nm and 5 fs/1600 nm pulses.[9] Similarly, adding a 5 fs/2000 nm pulse and a 5 fs/2200 nm pulse to the 5 fs/800 nm pulse, Li et al. obtained a supercontinuum spectrum with a bandwidth of 163 eV and an isolated 23-as pulse.[10] However, the pulse durations used in the three-color field schemes above are very short (even less than one optical cycle), which makes them difficult to be realized in the experiment.

In this paper, we theoretically investigate high-order harmonic generation from helium atom in a three-color field with longer pulse duration, and obtain effective isolated attosecond pulse generation. Here, the three-color field is synthesized by

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a 10 fs/800 nm Ti-sapphire laser pulse and a two-color field consisting of 30 fs/532 nm and 30 fs/1330 nm pulses. Addition of a two-color field locally changes the electric field profile of the 800-nm Ti-sapphire laser pulse, further control electronic ionization at several particular moments, thus the bandwidth of supercontinuum spectrum is extended dramatically to 235 eV due to the reduction of harmonic interference effects (compared with multiple ionization-recombination processes in the case of monochromatic laser field). After a wavelet transform for the front part of supercontinuum spectrum ranging from the 160th to the 210th order harmonics, an intense isolated 46-as pulse is obtained.

2. Theoretical methods

We use Lewenstein's strong-field approximation[12] model to calculate the harmonic radiation, the electronic time-dependent dipole moment in the external field can be described as (unless otherwise stated, atomic units are used throughout this paper)

$$\begin{aligned} d_{nl}(t) = & 2\,\mathrm{Re}\left\{ \mathrm{i}\int_{-\infty}^{t} \mathrm{d}t' \left(\frac{\pi}{\varepsilon + \mathrm{i}(t-t')/2}\right)^{3/2} \right. \\ & \times d^{*}\left[p_{\mathrm{st}}(t',t) - A(t)\right] d\left[p_{\mathrm{st}}(t',t) - A(t')\right] \\ & \left. \times \exp\left[-\mathrm{i}S_{\mathrm{st}}(t',t)\right] E(t') \right\} \\ & \times \exp\left[-\int_{-\infty}^{t} w(t')\,\mathrm{d}t'\right], \end{aligned} \tag{1}$$

where $A(t)$ is the vector potential of laser pulses, and ε is a small positive number, the electric field of the linearly polarized laser pulses $E(t)$ is given by

$$\begin{aligned} E(t) = & E_1 f_1(t)\cos(\omega_1 t) + E_2 f_2(t)\cos(\omega_2 t) \\ & + E_3 f_3(t)\cos(\omega_3 t). \end{aligned} \tag{2}$$

Here, ω_i $(i = 1,2,3)$ are the angular frequencies of three pulses whose wavelengths are 800 nm, 1330 nm, and 532 nm, respectively. The $f_i(t) = \exp\left[-2\ln 2\left(t^2/\tau_0^2\right)\right]$ are the corresponding pulse envelop functions, in which the pulse durations are in turn 10 fs , 30 fs, and 30 fs, and E_i are the peak amplitudes of the electric fields.

In Eq. (1), the stationary momentum P_{st} and the quasiclassical action S_{st} are given by

$$P_{\mathrm{st}}(t',t) = \frac{1}{t-t'}\int_{t'}^{t} A(t'')\,\mathrm{d}t'', \tag{3}$$

$$S_{\mathrm{st}}(t',t) = (t-t')I_{\mathrm{p}} - \frac{1}{2}p_{\mathrm{st}}^2(t',t) + \frac{1}{2}\int_{t'}^{t} A^2(t'')\,\mathrm{d}t'', \tag{4}$$

where the electric dipole transitions $d(p)$ for hydrogen-like atoms can be approximated as

$$d(p) = \mathrm{i}\frac{2^{7/2}\left(2I_{\mathrm{p}}\right)^{5/4}}{\pi}\frac{p}{\left(p^2+2I_{\mathrm{p}}\right)^3}. \tag{5}$$

According to the Ammosov, Delone, Krainov (ADK) model,[13] the ionization rate can be calculated as (the depletion of ground state has been taken into consideration in Eq. (1))

$$w(t) = \omega_{\mathrm{p}}\left|C_{n^*}\right|^2\left(\frac{4\omega_{\mathrm{p}}}{\omega_{\mathrm{t}}}\right)^{2n^*-1}\exp\left(-\frac{4\omega_{\mathrm{p}}}{3\omega_{\mathrm{l}}}\right), \tag{6}$$

where $\omega_{\mathrm{p}} = I_{\mathrm{p}}/\hbar$, $\omega_{\mathrm{t}} = |E_1(t)|/\sqrt{2I_{\mathrm{p}}}$, $\left|C_{n^*}\right|^2 = \frac{2^{2n^*}}{n^*\Gamma(n^*+1)\Gamma(n^*)}$, and $n^* = Z\left(I_{\mathrm{ph}}/I_{\mathrm{p}}\right)^{1/2}$, in which Z is the net resulting charge of the helium atom, and I_{ph} is the ionization potential of the hydrogen atom.

The dipole acceleration can be obtained from the time-dependent dipole moment, $a(t) = \frac{\partial}{\partial t^2}d_{nl}(t)$. The harmonic spectrum is calculated by Fourier transforming the dipole acceleration

$$P_{\mathrm{A}}(\omega) = \left|\frac{1}{t_{\mathrm{f}}-t_{\mathrm{i}}}\frac{1}{\omega^2}\int_{t_{\mathrm{i}}}^{t_{\mathrm{f}}} a(t)\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}t\right|^2. \tag{7}$$

By properly superposing several harmonics on the harmonic spectrum, an ultrashort attosecond pulse can be generated with the temporal profile

$$I(t) = \left|\sum_q a_q \mathrm{e}^{\mathrm{i}q\omega t}\right|^2, \tag{8}$$

where $a_q = \int a(t)\,\mathrm{e}^{-\mathrm{i}q\omega t}\,\mathrm{d}t$.

3. Results and discussion

To understand the physics behind isolated attosecond pulse generation by using a three-color pulse with longer pulse duration, we firstly simulate the HHG spectra. The HHG spectrum from helium atom subjected to the synthesized three-color pulse is presented as solid blue curve in Fig. 1. The electric fields of three pulses are set as E_1=0.17, E_2=0.05, and E_3=0.04 in turn, so the peak amplitude of a synthesized pulse is 0.26 at the appropriate phase condition. For comparison, we also performed simulation of HHG from the same atom subjected to a single Ti:sapphire fundamental pulse with peak amplitudes $E_1 = 0.17$ (dotted black curve) and $E_1 = 0.26$ (dashed red curve), respectively. It can be seen from the figure that the cut-off positions of the harmonic spectra in the three cases are obviously different. When a low-peak-amplitude fundamental pulse ($E_1 = 0.17$) is adopted as the driving pulse, the cut-off position of harmonic plateau is the 140th order. When a high-peak-amplitude fundamental pulse ($E_1 = 0.26$) is adopted, the cut-off position extends to the 305th order. The two cut-offs are in agreement with the cut-off position formula. In contrast to the HHG in the fundamental pulse, the cut-off position of the harmonic spectrum in the three-color pulse is the 306th order, which is nearly consistent with that of harmonic spectrum of the fundamental pulse with a peak amplitude of $E_1 = 0.26$.

More importantly, in the three-color case there exists a broadband supercontinuum ranging from 154th to 306th order harmonics which can be used to synthesize an isolated attosecond pulse with a short duration. While in the monochromatic field case the harmonic spectrum experiences a great modulation, leading to the absence of supercontinuum structure.

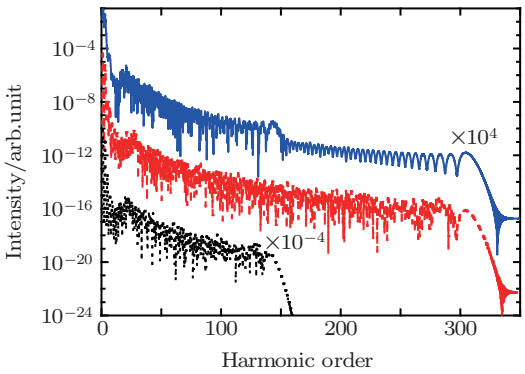


Fig. 1. (color online) HHG spectra from helium atom exposed to an 800-nm fundamental pulse with a peak amplitude $E_1 = 0.17$ (dotted black) and $E_1 = 0.26$ (dashed red), respectively. Solid blue curve: the harmonic spectrum from helium atom exposed to the three-color field. For comparison, the harmonic intensities are multiplied by a factor of 10^{-4} and 10^4 in the case of an 800-nm fundamental field with $E_1 = 0.17$ and the three-color field, respectively.

In the following, we analyze the reason for the generation of supercontinuum spectrum in the three-color laser pulse.

Figure 2(a) shows the evolution of the electric field amplitude as a function of time in the fundamental pulse with the peak amplitude $E_1 = 0.17$ and $E_1 = 0.26$, respectively. It is known from the three-step model that the cut-off of the harmonic spectrum is determined by the maximum kinetic energy the electron possesses as it returns to the nucleus. Along with the increase in the electric field amplitude of laser pulse, the ionized electrons obtain larger kinetic energy as it returns to the nucleus, leading to the extension of harmonic plateau. However, in a single fundamental field with larger amplitude, the electrons experience the ionization-recombination process at nearly every peak of electric field, so harmonics emitted at different moments interfer with each other, thus leading to the absence of supercontinuum structure. As a result, it cannot be used to synthesize the isolated attosecond pulse. To solve this problem, we impose a two-color field consisting of a 30 fs/532 nm and a 30 fs/1330 nm pulses on a 10 fs/800 nm laser pulse, thus the synthesized laser field can be used to control the electrons from three aspects: ionization moments, motion after ionization, and their kinetic energies at recombination moments.

The amplitude evolution of the synthesized three-color field as a function of time is shown in Fig. 2(b) by the dotted red curve, we also give the amplitude evolutions of a single 800 nm pulse with the peak amplitude $E_1 = 0.17$ and two-color pulse in Fig. 2(b) by the dashed green and dotted red curves, respectively. To clearly illustrate the influence of the three-color field on the HHG spectra, we label the shadow region as A_1, A_2, and A_3. In region A_1, we lower the ionization rate. The peak amplitude of the two-color field is opposite to the 800-nm fundamental field in region A_1, thus reducing the electric field strength of the fundamental field. As a result, the ionization probability in region A_1 is so low that there is nearly no harmonics generated in region A_1. The ionization in region A_2 makes a major contribution to the harmonic spectrum. Now we can control the motion of the ionized electrons by introducing the two-color field to shift the secondary peak of the fundamental field towards the right, so that the synthesized field benefits the harmonic generation of short trajectory and realizes the trajectory selection. In region A_3, we extend the cut-off of the harmonic spectrum. The two-color field and fundamental field are in the same direction, and the increased field strength accelerates the electrons ionized at the secondary peak efficiently. As a result, the electrons obtain more energy and emit high-energy photons as they recombine with the nucleus. This prolongs the cut-off of the harmonic plateau and generates a supercontinuum spectrum with a large bandwidth of about 235 eV. Therefore, the optimized three-color field scheme plays an important role at three key points for HHG process: reducing the ionization, which is beneficial to macroscopic propagation effect, controlling the motion of ionize electrons, which is beneficial to trajectory selection, and controlling the peak amplitude, which is beneficial to a larger extension for the cut-off. Thus, the three-color scheme overcomes the problems caused by a monochromatic field, for instance, over ionization under the higher electric strength, interference among the harmonics generated at different moments, etc.

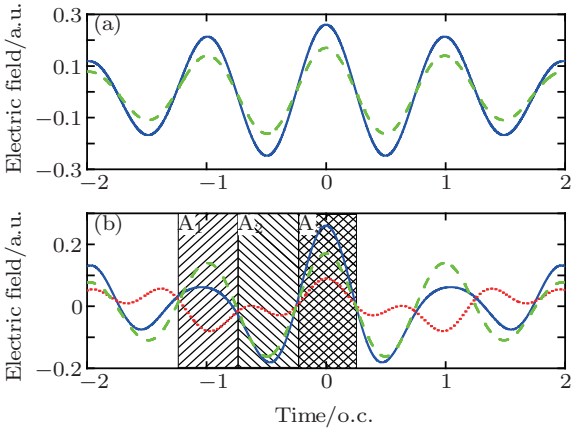


Fig. 2. (color online) (a) Variation of the electric field amplitude of a single 800-nm pulse with a peak amplitude of $E_1 = 0.17$ (dashed green) and $E_1 = 0.26$ (solid blue), respectively. (b) Variation of the electric field amplitude of a single 800-nm pulse with a peak amplitude of $E_1 = 0.17$ (dashed green), two-color pulse (dotted red), and three-color pulse (solid blue).

In the following, a semi-classical three-step model will be used to illustrate the dynamical behavior of electrons dur-

ing the above processes and analyze the mechanism of isolated attosecond pulse generation in the three-color field in details.

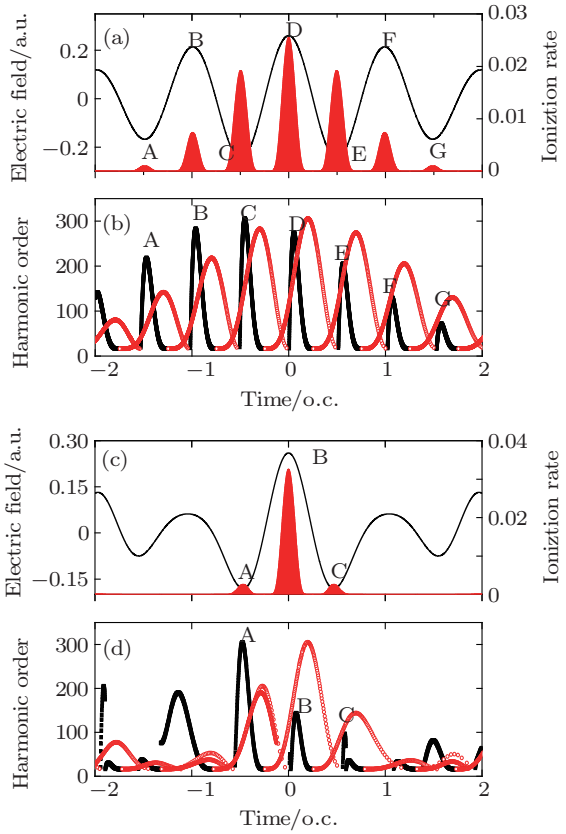


Fig. 3. (color online) (a) Variation of the amplitude of the 800-nm monochromatic field with a peak amplitude of 0.26 a.u. (solid black curve) and the ionization rate of the helium atom radiated by the fundamental field which is calculated from the ADK (filled red region). (b) Evolution of the harmonics with ionization (black dot) and recombination (red circle) time in the monochromatic field case, where the peak amplitude is 0.26 a.u.. (c) Variation of the amplitude of the three-color field (solid black curve) and the ionization rate of the helium atom radiated by three-color field which is calculated from the ADK (filled red region). (d) Evolution of the harmonics with ionization (black dotted curve) and recombination (red circle) time in the three-color field case.

In Figs. 3(a) and 3(b), the black curves show the amplitude evolution of the fundamental and three-color fields, respectively, and the filled red region denotes the corresponding ionization rate calculated from ADK model. The harmonic evolutions with ionization (black dot) and recombination (red circle), obtained from the semi-classical three-step model, are also given in these figures. It can be seen from Fig. 3(a) that in the monochromatic field case, there exists ionization at moments A, B, C, D, E, F, and G corresponding to the generation of the 222nd, 284th, 305th, 267th, 207th, 127th, and 73rd order harmonics, and these harmonics generated at different moments interfere with each other, resulting in the fierce modulation of harmonic spectrum, leading to the absence of supercontinuum. In contrast, in the three-color field case, the two-color field changes the electric field profile, making the electron ionized significantly at three moments: A, B, and C, and the ionizations at the three moments in Fig. 3(d) correspond to the generation of the 306th, 154th, and 107th order harmonics. The harmonic spectrum from the three-color field shown in Fig. 1 verifies the above analyses: the harmonics less than the 107th order experience fierce modulation, that ranging from the 107th to the 154th order experience much weaker modulation, and that ranging from the 154th to the 306th order form a broad supercontinuum. The supercontinuum ranging from the 154th to the 306th order harmonics originates mainly from two quantum trajectories, i.e., the long one which is ionized earlier and back to the nucleus later, and the short one which is ionized later but back to the nucleus earlier. However, the electrons are mainly ionized at three moments in the case of the three-color field scheme that overcomes obviously the over ionization and interference problem brought by a monochromatic field. Moreover, in the case in which the cut-off of the harmonic plateau dose not increase, a supercontinuum with much broader bandwidth than that in monochromatic field can be achieved, which is priority to the generation of shorter attosecond pulses.

Considering the three-color laser field scheme, we further investigate high-order harmonic generation from helium atom by means of the wavelet transform.[13,14] The time-frequency analysis of the harmonics verifies the conclusion above. In Fig. 4, we can see that, for the harmonics ranging from the 154th to the 306th order, only the peak P_1 mainly contributes to their generation, and there are two branches at the left and right sides of the peak P_1, where the left branch corresponds to the short trajectory and the right one corresponds to the long trajectory. It can be seen that the left branch is stronger than the right one, indicating that the short trajectory plays the main role in harmonic generation and the long trajectory is depressed. It means that we can realize the trajectory selection through the optimization of three-color field scheme. It can also be seen from the figure that, along with the increase in the harmonics order, the intensities of both long and short trajectories become weak gradually. Therefore, in order to obtain an intense isolated attosecond pulse, one should superpose the front part of the supercontinuum for synthesizing attosecond pulse. Under this guide, the inverse Fourier transform of part of the harmonic spectrum is performed. It is found that an isolated pulse with a duration of about 50 as can be obtained by superposing harmonics covering arbitrary 50 orders, and the envelope of the isolated attosecond pulse obtained is shown in Fig. 5, which is identical to the time-frequency analysis. The attosecond pulse generated by superposing the harmonics from the 160th to the 210th order is one order of magnitude higher than that generated by superposing the harmonics from the 260th to the 305th order, in agreement with the above discussion. The final pulse duration of the former is as short as 46 as.

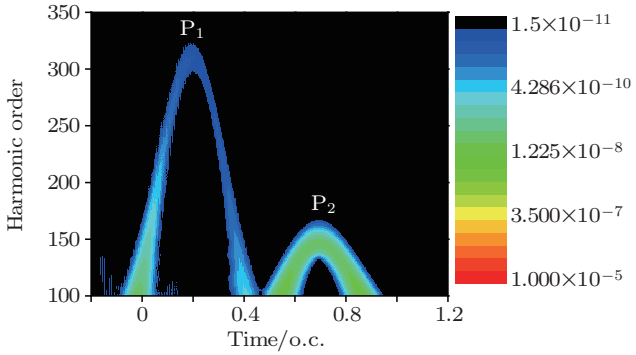


Fig. 4. (color online) Time-frequency analysis of the harmonics generated from helium atom irradiated by the three-color field.

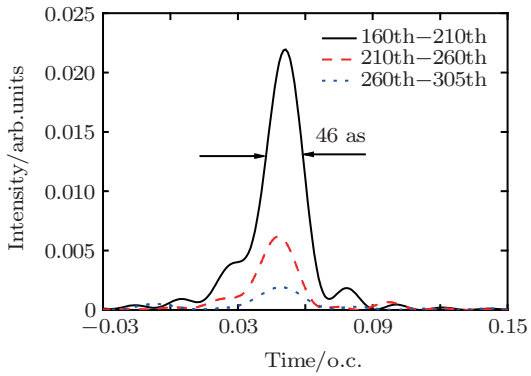


Fig. 5. (color online) Isolated attosecond pulses obtained by superposing the harmonics from the 160th order to the 210th order (solid black line), from the 210th order to the 260th order (dashed red line), and from 260th order to 306th order (dotted blue line).

4. Conclusion

In this paper, we numerically investigate the HHG spectra generated from the helium atom irradiated by a three-color field consisting of a 10 fs/800 nm Ti-sapphire laser and two attached pulses whose pulse duration/wavelength are 30 fs/532 nm and 30 fs/1330 nm, respectively. Compared with that generated by a monochromatic field, the harmonics generated from the three-color field show a supercontinuum with a bandwidth of 235 eV, ranging from the 154th to the 306th order harmonics. Then we explained the cause of the supercontinuum generation according to the three-step model in combination with the ADK ionization. The electrons are mainly ionized at three specific moments by imposing a two-color pulse, thus overcoming the problem that in the monochromatic field case, the harmonics generated at different moments interfere with each other, which limits the bandwidth of supercontinuum. Although the cut-off of harmonics is not increased, a supercontinuum with a 235 eV bandwidth can be obtained. Finally the theoretical simulation gives an intense isolated 46-as pulse by superposing the supercontinuum from the 160th to the 210th order harmonics.

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