Broadband isolated attosecond pulse with high spatiotemporal quality in pre-excited medium by multi-cycle two-color fields

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Abstract: We propose a scheme that combines multi-cycle two-color pump pulses and pre-excited medium to simultaneously control the ionization and acceleration steps of electron wave packet. Intense two-plateau supercontinua are observed at microscopic level, which support isolated attosecond pulse with tunable central wavelength. We further investigate the propagation effects and find that the macroscopic response is mainly associated with the initial population of the excited state. Large initial population can distort the pump field and weaken the phase-matching of the supercontinua due to the high density of free electrons. On the contrary, small initial population of 3% can generate well phase-matched supercontinua, and an efficient isolated attosecond pulse with gaussian-like spatial distribution is directly achieved. The divergence angle of the generated pulse is less than 0.2 mrad, which indicates its beam quality is good. This pulse benefits for some potential applications of ultrafast measurements with high spatiotemporal resolutions.

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OCIS codes: (190.7110) Ultrafast nonlinear optics; (190.4160) Multiharmonic generation; (300.6560) Spectroscope, x-ray.

References and links
1. Introduction

High-order harmonic generation (HHG) has been recognized as one of the best ways of producing isolated attosecond pulses [1], which have successfully opened the new research areas of the ultrafast processes in much shorter time-scale, such as the inner-shell electron dynamics in atoms and molecules [2, 3]. For the practical applications of isolated attosecond pulses, two factors, i.e., the pulse efficiency and the pulse duration are very crucial. In the past few years, many efforts have been paid to broaden the bandwidth of supercontinuum and shorten the pulse duration for much higher time resolution [4–10]. The sub-100 as barrier has been first brought through by Goulielmakis and co-workers [4]. In their experiment, a near-single-cycle laser pulse has been employed as pump source to produce 40 eV supercontinuum and an isolated 80 as pulse with energy of 0.5 pJ has been obtained. On the other hand, attosecond radiation with high peak intensity has potential in opening new avenues for pump-probe studies. Ferrari et al. recently used above-saturation few-cycle (5 fs) pulses to generate an isolated attosecond pulse and the pulse energy reached nanojoule-level [11]. Nowadays, it is still extremely challenging to obtain intense broadband isolated attosecond pulse in multi-cycle regime.

The classical three-step model [12] indicates that the HHG process is mainly determined by the ionization, acceleration and recombination of the electrons in the laser field. This classical picture suggests that many approaches can be introduced to manipulate the electron trajectories for the broadband supercontinuum generation. One effective approach is to adopt wave-form-induced “time gating” by using two-color fields, i.e., the so-called “ionization gating” [9,10,13], “acceleration gating” [14, 15] and “recombination gating” [16, 17] schemes. Moreover, two-color methods offer increased control over attosecond pulse generation, allowing the use of multi-cycle pump pulses [15, 18]. According to the three-step model, the harmonic efficiency is mainly decided by the ionization rate when the intensity of the driving field is far below the saturation intensity of the target. The ionization gating scheme may be more meaningful to enhance harmonic efficiency. Recently, Mashiko et al. combined the ionization gating and the polarization gating techniques to simultaneously control the ionization and recombination steps, named double optical gating (DOG), and an efficient broadband supercontinuum was directly generated in the plateau [19]. The use of specific target may improve HHG properties, e.g., Rydberg atoms or medium with coherent superposition state. In the scheme with Rydberg atoms, due to stabilization of the Rydberg electron, intense laser may essentially interact with inner electron that have high ionization potential, thereby producing very high harmonics [20]. By contrast, using coherent superposition state of the target can provide an effective way to enhance the harmonic efficiency [21, 22]. In this system, part of electron wave packet are populated to excited state, they can be easily triggered to continuous state, while others at the ground state are hardly ionized owing to its high ionization potential. Very recently, Hong et al. used intense few-cycle laser pulses to drive a pre-excited medium, and found that the rapid depletion of the excited state takes place within a very short time and this rapid depletion also works as an ionization gating for isolated attosecond pulse generation [23]. Du et al. pushed this scheme to multi-cycle pump regime through employing two-color chirped and chirped-free pulses [24]. However, to the best of our knowledge, such chirped driving pulses are still difficult for current laser technology, which limits the application of their approach.

In our recent publication, an effective acceleration mechanism for cutoff extension and broadband supercontinuum generation has been presented in multi-cycle two-color pulses [15]. Compared with the chirped and chirped-free two-color configuration in [24], our scheme does not require a chirped driving field to obtain broadband isolated attosecond pulse. This offers better feasibility for experimental demonstration. Unfortunately, in helium atoms the efficiency of the generated continuous harmonics is unsatisfactory, because of the wave packet spreading during the long-distance acceleration. In this paper, we introduce pre-excited system to over-
come this defect. Microscopically, it is found that the combination of the multi-cycle two-color pump pulses and pre-excited medium can simultaneously control the ionization and acceleration steps. Then intense two-plateau supercontinua, i.e., one plateau with bandwidth of approximately 80 eV located in extreme ultraviolet (XUV) region and another plateau with bandwidth of approximately 100 eV located in water-window region, are observed in single-atom response. This two-plateau structure can support isolated attosecond pulse with tunable central wavelength. Macroscopically, we investigate the propagation effects [25, 26]. Our results reveal that the initial population of the excited state significantly determines the performance of the supercontinua. Large population of the excited state will lead to high density of free electrons in the medium. The presence of such high-density free electrons will distort the pump field-form and weaken the phase-matching of the supercontinua. Whereas, small population of the excited state at 3% can lead to good phase-matching, and an efficient isolated attosecond pulse with gaussian-like spatial distribution is directly achieved. The divergence angle of the generated pulse is less than 0.2 mrad, which implies the beam quality is good. This pulse benefits for some potential applications of ultrafast measurements with high spatiotemporal resolutions.

2. Theoretical methods

Our theoretical investigation of HHG in the pre-excited medium includes calculations of the dipole response at single-atom level induced by the laser field and the co-propagation effects of the pump laser and harmonic beams.

2.1. Single-atom response

In the single-atom response, the initial state of the pre-excited system can be written as

$$\Psi(r,t_0) = \alpha|g\rangle + \beta|e\rangle,$$

(1)

where the ground state $|g\rangle$ and excited state $|e\rangle$ are chosen to be the $1s$ and $2s$ states of the $He^+$ ions with the binding energies of 54.4 eV and 13.6 eV, respectively. $\alpha$ and $\beta$ are the amplitudes of the ground and excited states, which satisfies the condition: $\alpha^2 + \beta^2 = 1$. The initial population of the excited state can be expressed as $p = \beta^2$. The instantaneous dipole moment of the system in single-atom response is described by

$$d_{nl}(t) = \langle \Psi(r,t)|\hat{x}|\Psi(r,t)\rangle,$$

(2)

The instantaneous dipole moment is calculated by numerically solving the time-dependent Schrödinger equation. The calculation details can be found in [27]. The harmonic spectrum is then obtained by Fourier transforming the time-dependent dipole acceleration $\ddot{a}(t)$:

$$a_q = \frac{1}{T} \int_0^T \ddot{a}(t)\exp(-iq\omega t)|^2,$$

(3)

where $\ddot{a}(t) = \ddot{d}_{nl}(t)$, $T$ and $\omega$ are the duration and frequency of the driving pulse, respectively. $q$ corresponds to the harmonic order.

2.2. Macroscopic propagation effects

The collective response of the macroscopic medium is described by the co-propagation of the laser and the harmonic field, which is performed by separately solving Maxwell wave equations for laser and harmonic field in cylindrical coordinates [28].

$$\nabla^2 E_f(\rho,z,t) - \frac{1}{c^2} \frac{\partial^2 E_f(\rho,z,t)}{\partial t^2} = \frac{\omega_p^2(\rho,z,t)}{c^2} E_f(\rho,z,t)$$

(4)

$$\nabla^2 E_h(\rho,z,t) - \frac{1}{c^2} \frac{\partial^2 E_h(\rho,z,t)}{\partial t^2} = \frac{\omega_h^2(\rho,z,t)}{c^2} E_h(\rho,z,t) + \mu_0 \frac{\partial^2 P_{nl}(\rho,z,t)}{\partial t^2}$$

(5)
where $E_f$ and $E_h$ are laser and harmonic fields, $\omega_p = e\sqrt{4\pi n_e(\rho,z,t)/m_e}$ is the plasma frequency, and $P_{nl}(\rho,z,t) = [n_0 - n_e(\rho,z,t)]d_{nl}(\rho,z,t)$ is the nonlinear polarization generated by the medium. $n_0$ is the gas density and $n_e(t) = n_0 \left[1 - \exp \left(-\int_{-\infty}^{t} w(t') dt' \right)\right]$ is the free-electron density in the gas. This model takes into account both temporal plasma induced phase modulation and the spatial plasma lensing effects on the laser field. Here Eq. (4) and (5) can be numerically solved using the Crank-Nicholson method. The calculation details can be found in [29].

3. Result and discussion

In our simulation, the pump laser field is given by $E_p(t) = E_0 f(t) \cos(\omega_0 t)\hat{x} + E_1 \cos(\omega_1 t - 0.5\pi)\hat{x}$, $0 \leq t \leq T$. $\hat{x}$ is the polarization vector. $E_0$, $E_1$ and $\omega_0$, $\omega_1$ are the amplitudes and frequencies of the driving laser at 0.8 $\mu$m and control laser at 10.4 $\mu$m, respectively. $f(t) = \sin^2(\frac{\pi t}{T_0})$ is the pulse profile, and $T = 13T_0$ is the pulse duration corresponding to 13 fs full width at half maximum (FWHM), where $T_0$ denotes the optical cycle of the driving pulse. Here we choose $1 \times 10^{14}$ W/cm$^2$ and $1.2 \times 10^{13}$ W/cm$^2$ as the intensity of the driving pulse and the control pulse, respectively. In this condition, the pump field can not cause the ground state electrons to tunnel. But, for the electrons at excited state, the tunneling ionization is much easier to take place. Because the ionization potential at excited state (13.6 eV) is much lower than that at ground state (54.4 eV). The sketch of our scheme is shown in Fig. 1. The grey line in Fig. 1(a) shows the time evolution of the pump field, and the red dots and the green circles represent the classical ionization and recombination. According to Fig. 1(a), the acceleration gating is formed between 4.42$T_0$ and 4.61$T_0$. Details of this acceleration mechanism have been presented in our recent publication [15]. Figure 1(b) gives the corresponding normalized populations of the excited (yellow area) and the ground state (grey area) versus time during the pump field. As shown in Fig. 1(b), the population of the ground state is approximately constant. While, for the population of the excited state, there are three steep decreases within the time.
interval between $2.1T_0$ and $2.6T_0$, between $3.3T_0$ and $3.6T_0$, and between $4.4T_0$ and $4.7T_0$. This implies that the ionization rate in these three time regions is significantly higher than others. Notably, the acceleration gating exactly locates in the time interval of the third steep decrease (between $4.4T_0$ and $4.7T_0$). Thus, the combination of the multi-cycle two-color pulses and the pre-excited system can simultaneously gate the ionization and acceleration steps, forming a double control gating. It is worth mentioning that appropriate driving intensity and control intensity are extremely crucial to form such a double control gating. Too high pump intensity may result in the rapid depletion of the electrons at excited state to zero before the acceleration gating.

Fig. 2. (a) The microscopic harmonic spectrum and (b) its time-frequency structure from the pre-excited medium with different initial population of the excited state of $p = 0.1$ (blue curve), $p = 0.06$ (red curve) and $p = 0.03$ (green curve).

Fig. 3. The macroscopic on-axis harmonic spectrum in the pre-excited medium with different initial population of the excited state of $p = 0.1$ (blue curve), $p = 0.06$ (red curve) and $p = 0.03$ (green curve).

To confirm the classical sketch, we perform the quantum calculations on the harmonics generation at single-atom level. The microscopic harmonic spectrum is shown in Fig. 2(a). The harmonic spectrum from the pre-excited system with $p = 0.1$ is plotted by blue curve. One can see that two obvious continuous harmonic plateau with bandwidth of 80 eV and 100 eV.
are presented. The 80 eV supercontinuum (from 120th to 172th) locates in extreme ultraviolet (XUV) region and the 100 eV supercontinuum (from 195th to 260th) locates in water-window region. For deeper understanding of this spectral structure, the emission time of the harmonics in terms of time-frequency analysis is presented in Fig. 2(b). It shows that three dominated peaks with just left branch contribute to harmonics radiation, and the corresponding harmonic order is 170th, 195th and 260th. These characteristics agree well with the classical descriptions in Fig. 1(a), and simultaneously indicate that the harmonics from 120th to 172th and from 195th to 260th display supercontinuum, which is also consistent with the results in Fig. 2(a). In addition, the modulation on the harmonics between 172th and 195th comes from the interference of the electron wave packet in overlap area. The supercontinuum generation is also associated with the initial population of the excited state. Then, we present the harmonic spectrum with different initial populations of $p = 0.06$ and $p = 0.03$ in Fig. 2(a) by red curve and green curve. As shown in Fig. 2(a), the harmonic efficiency increases with increasing $p$. This can be interpreted that the initial populations determine the ionization probability and high initial populations make more electrons to be ionized and emit harmonics. At the same time, we should note that the variation of $p$ does not change cutoff and the spectral profile of the harmonics except the efficiency. On the other hand, the pump intensity in our scheme can not cause the ground state electrons to tunnel [see Fig. 1(b)]. In other words, the electrons at ground state can not be pumped to the continuum. Thus, in this intensity condition, we can not obtain the harmonic spectrum from ground state.

![Fig. 4. The electric fields of the pump pulses after propagation with the initial populations of $p = 0.1$ (blue line), $p = 0.06$ (red line) and $p = 0.03$ (green line), and the initial pump field (dash line).](image)

In fact, the HHG are generated by macroscopic number of ionization atoms or ions interacting with focused laser field, and the ionization is a very crucial factor in this nonlinear process. In pre-excited system, the excited state is responsible for the ionization during the laser-matter interaction. As mentioned above, the initial population of the excited state directly determines the ionization probability of the system and the resulting free-electron density in the medium. High density of free electrons strongly changes the spatiotemporal propagation dynamics of the laser pulses, such as the spatial defocusing of the beam, the temporal distortion of the pump field (dephasing), and thereby the phase-matching of HHG and the isolated attosecond generation. In the following, we investigate the propagation effects of the supercontinuum generation in such pre-excited system with moderate gas pressure and medium length, and the non-adiabatic three-dimensional (3D) propagation simulations [28, 29] for both pump and har-
monic fields are performed in the gas target. Here we consider 3D propagation through a 0.5 mm gas jet with a density of $0.6 \times 10^{18}$ cm$^{-3}$. In our calculations, the driving and the control pulses are assumed as Gaussian beams and are both focused 2 mm before the gas jet, and the beam waists at focus of the two pulses are 30 μm and 120 μm, respectively. For this configuration, the confocal parameter of the driving pulse is approximately equal to that of the control pulse. Since, in our scheme, single quantum trajectory has been microscopically selected and two-plateau supercontinua have been successfully obtained in the single-atom response. We should focus on choosing appropriate initial population of the excited state, of which the density of the free electrons is safely low and the influence of ionization-induced defocusing and dephasing are slight to the spatiotemporal coherence of the supercontinua. Figure 3 presents the macroscopic on-axis harmonic spectrum in the pre-excited medium with different initial populations of $p = 0.1$ (blue curve), $p = 0.06$ (red curve) and $p = 0.03$ (green curve). One can see that the supercontinua in the case of $p = 0.03$ are smooth, which indicates the harmonics after propagation are well phase-matched. In comparison with the case of $p = 0.03$, some regions in the supercontinua show slight modulation when $p = 0.06$, therefore the bandwidth of the continuous part is shortened. Instead, in the case of $p = 0.1$, the harmonic modulation becomes substantially deep and the supercontinua can not be macroscopically obtained. These phenomena can be attributed to two reasons. The first one is that high-density free electrons originating from large initial population of the excited state weaken the phase-matching of the supercontinua. The other reason is the distortion of the electric field during propagation. Figure 4 shows the on-axis electric field of the pump pulse at the end of the medium for above three cases. For comparison, the initial pump field at entrance of the gas jet is also presented by dash line, and the enlargement of a part field is shown in the inset for visual convenience. As shown in this figure, the electric-field distortion becomes noticeable as the initial population of the excited state increases.

Fig. 5. Normalized temporal profiles of the XUV attosecond pulses with the initial populations of (a) $p = 0.03$, (b) $p = 0.06$ and (c) $p = 0.1$. Spatiotemporal profiles of the XUV attosecond pulses at the end of the medium with the initial populations of (d) $p = 0.03$, (e) $p = 0.06$ and (f) $p = 0.1$.

Next, we investigate the temporal characteristics of the macroscopic on-axis harmonic spectrum by means of the application of a square window with a width of 20 harmonics (31 eV). For convenience, the harmonics from 130th to 150th in the XUV region and the harmonics from 220th to 250th in the water-window region are chosen. Figure 5(a)-5(c) show the normalized temporal profiles of the on-axis XUV attosecond pulses for the cases of $p = 0.03$, $p = 0.06$
and $p = 0.1$, and Fig. 6(a)-6(c) show the corresponding water-window attosecond pulses. For $p = 0.03$, isolated XUV and water-window attosecond pulses can be directly filtered out from the supercontinua. For $p = 0.06$, a weak satellite pulse appears in the water-window region. When $p$ is further increased to 0.1, the water-window satellite pulse becomes noticeable and multi-peaks attosecond burst emerges in the XUV region. Moreover, compared with the cases of $p = 0.03$ and $p = 0.06$, the emitted time of the main pulse in the XUV region is changed. Here not only the poor phase-matching of the harmonics caused by the high-density free electrons but also the significant distortion of the laser field should be responsible for this change. In addition, the spatial distribution is also an important characteristic of the generated attosecond pulse. Figure 5(d)-5(f) show the the spatiotemporal profiles of the XUV attosecond pulses for the cases of $p = 0.03$, $p = 0.06$ and $p = 0.1$, and Fig. 6(d)-6(f) show the corresponding spatiotemporal profiles of the water-window attosecond pulses. Similarly, the spatial distributions of the attosecond pulses are related to the density of free electrons. As the population of the excited state increases, free electrons induced refractive index may cause the laser phase front to advance faster on axis than off axis, resulting in the spatial defocusing of the beam. As shown in Fig. 5(d)-5(f) and Fig. 6(d)-6(f), the higher density of free electrons for $p = 0.1$ makes its spatiotemporal distribution much more complicated than those for $p = 0.03$ and $p = 0.06$.

In order to obtain the emitted power of the attosecond pulses, We further calculate the radially integrated attosecond pulses in terms of the relationship

$$P_r = \int_0^R \left( \frac{1}{2} E_0 c E_a^2 \right) 2\pi rdr,$$

where $E_a$ is the electric field of the attosecond pulses at the end of gas jet. The radially integrated temporal profiles of the XUV attosecond pulses in the cases of $p = 0.03$ (green line), $p = 0.06$ (red line) and $p = 0.1$ (blue line) are shown in Fig. 7(a), and the corresponding power profiles of the water-window attosecond pulses are shown in Fig. 7(b). It is clear that, the radially integrated attosecond pulse of $p = 0.1$ exhibits several peaks with irregular temporal structure. Besides, the emitted power of the attosecond pulse is lower than those of $p = 0.03$ and $p = 0.1$, although its on-axis harmonic efficiency is higher (see Fig. 3). This may be due to its
more complicated spatiotemporal distribution, and thereby the destructive integration of the harmonics in both temporal and spatial domains. Following, we take $p = 0.03$ as example to evaluate the emitted power of the attosecond pulses in the XUV and water-window region. According to Fig. 7(a), in the XUV region, a pure isolated attosecond pulse with the pulse duration of approximately 155 as and the emitted power of $5.6 \times 10^5 \, \text{W}$ can be directly filtered out. The energy of this pulse is estimated to be approximately 0.1 nJ. In the same way, a pure isolated pulse with the pulse duration of approximately 160 as and the emitted power of $6 \times 10^3 \, \text{W}$ can also be obtained in the water-window region, see Fig. 7(b). The corresponding pulse energy is estimated to be approximately 1.4 pJ.

Finally, we take $p = 0.03$ as example to further explore the beam qualities of the isolated attosecond pulses. We calculate the far-field distributions of the generated isolated attosecond pulses by the Hankel transformation from the near-field harmonic emissions at the end of the gas jet. The calculation details can be found in [30]. The divergences of the isolated attosecond pulses in the XUV region and in the water-window region are presented in Fig. 8(a) and 8(b), respectively. The insets show the spots with the distance of 0.5 m from the exit of the gas jet for above two cases. One can see that the spatial distributions of the XUV and water-window pulses are both gaussian-like. The divergence angles of the two beams are 0.1 and 0.15 mrad, which indicates good beam quality for both cases.

4. Conclusion

In conclusion, we present a scheme to simultaneously control the ionization and acceleration steps of electron wave packet corresponding to the harmonic process. Intense two-plateau supercontinua that consist of 80 eV XUV plateau and 100 eV water-window plateau are observed in single-atom response, which support broadband isolated attosecond pulse with tunable central wavelength. Macroscopically, it is found that the temporal and spatial characteristics of the isolated attosecond pulses are governed by the initial population of the excited state. Large initial population induced high-density free electrons not only distort the pump field but also weaken the phase-matching of the supercontinua. Instead, small initial population of 3% can generate well phase-matched supercontinua, and sub-200 as isolated attosecond pulses with good spatial quality of gaussian-like distributions and small divergence angles are achieved.
Furthermore, we would like to discuss the generality of this scheme. On the one hand, our calculations show that 2%-5% initial population of the excited state can produce broadband isolated attosecond pulse. When it increases to 6%, some regions in the supercontinua are modulated and therefore satellite pulses are emerged. When the initial population of the excited state becomes more, for instance, 10%, the supercontinua becomes deeply modulated and multi-peaks attosecond pulses with equal weighted intensities are appeared. On the other hand, it is worth mentioning that the laser intensity choice of driving and control pulses should be appropriate to form the double control gating. Too high pump intensity may result in the rapid depletion of the electrons at excited state to zero before the acceleration gating.

Experimentally, the driving pulse can be realized by Ti:sapphire laser and the control pulse can be carried out with tuned CO$_2$ laser. Since they are both located in the multi-cycle regime, the two-color fields are easy to produce and control for current laser technology. Moreover, the obtaining of such pre-excited medium can be realized by multiphoton resonant excitation [21, 31, 32].

Acknowledgment

This work was supported by the National Natural Science Foundation of China under Grant Nos. 10904045, 11104092 and 60925021, the 973 Program of China under Grant No. 2011CB808103, and the Technology Creative Project of Excellent Middle & Young Team of Hubei Province under Grant No. T201204.