Dramatic cutoff extension and broadband supercontinuum generation in multi-cycle two color pulses

Jianghua Luo,1,3 Weiyi Hong,1 Qingbin Zhang,1,4 Kunlong Liu,1 and Peixiang Lu1,2,*

1Wuhan National Laboratory for Optoelectronics and School of Physics, Huazhong University of Science and Technology, Wuhan 430074, China
2Key Laboratory of Fundamental Physical Quantities Measurement of Ministry of Education, Wuhan 430074, China
3College of Physical Science and Technology, Yangtze University, Jingzhou 434023, China
4zhangqingbin@mail.hust.edu.cn

Abstract: We propose a method to markedly increase the electron-recollision kinetic energy, using the combination of a 0.8 μm/13 fs driving pulse with a much weaker multi-cycle mid-infrared pulse at 10.4 μm. The results show that the synthesized field effectively lengthens the accelerated distance of electron wave packet and the harmonic cutoff is significantly extended to \( I_P + 26U_P \), which is covered with the water-window spectral region. In addition, only one single quantum path contributes to harmonics, and those higher than \( I_P + 15U_P \) become continuous. This supercontinuum can support the generation of an isolated sub-100 as pulse with tunable central wavelength and also the pulse with the duration below one atomic unit of time (24 as). Moreover, our scheme can further extend to more longer driving pulses, which provides a dramatic approach for cutoff extension and broadband supercontinuum generation with multi-cycle pump pulses.

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References and links
1. Introduction

High-order harmonic generation (HHG) known as high-nonlinear, non-perturbative laser-atom interaction has provided the most attracted source of sub-femtosecond pulses, opening new fields of time-resolved metrology on attosecond time scale [1,2]. An isolated attosecond-pulse is an important tool for probing and manipulating the electronic dynamics, and it has pushed fields of time-resolved metrology on attosecond time scale [1,2]. An isolated attosecond-pulse interaction has provided the most attracted source of sub-femtosecond pulses, opening new fields of time-resolved metrology on attosecond time scale [1,2].

So far, the production of attosecond-pulse is mainly dependent on HHG in gases. The process of HHG can be easily understood in the framework of electron-recollision model involved three-step [4]: ionization, acceleration and recombination. During the recombination, the maximum emission energy typically obeys cutoff law, $I_P \propto E_{\text{rec}}$, where $I_P$ and $U_P$ are the ionization potential and the ponderomotive energy, respectively. This process occurs every half-cycle, and leads to an attosecond-pulse train. Since practical application prefers an isolated pulse, many efforts have been paid to extract a single attosecond-pulse. Spectral filtering of several continuous harmonics near the cutoff generated by a few-cycle driving laser offers the simplest scheme. This scheme has been carried out in experiment directly using a two-cycle (5 fs) driving pulse [3]. However, owing to the limitation of the supercontinuum bandwidth, the duration of the shortest attosecond-pulse is only 250 as. To compress the pulse duration, another scheme
that uses a few-cycle (≈5 fs) driving pulse with the polarization gating techniques has been proposed [5]. And soon it was experimentally achieved by Sansone et al. In their experiment, a single 130 as pulse was filtered out after compensating the harmonic chirp [6]. Recently, by employing a near-single-cycle (≈2.5 fs) driving pulse, Goulielmakis et al broke through sub-100 as barrier and an isolated 80 as pulse was obtained in their work [7].

Currently, there is a great interest in the generation of isolated attosecond-pulse with a driving pulse substantially longer than two-cycle (5 fs). Because two-cycle pulses are notoriously hard to produce and control [8–10]. It has been proposed that two-color control scheme can effectively produce isolated attosecond-pulse based on multi-cycle regime [11,12]. Moreover, this approach also has the potential to select short or long path [13,14], extend the cutoff energy [15,16], enhance HHG conversion efficiency [17–19], and steer the electron motion in continuous state [20]. As we all known, the bandwidth of the supercontinuum plays a crucial role in performing ultra-fast measurements [21]. In order to obtain a broadband supercontinuum, the main challenge comes from the effective enlargement of HHG cutoff. All previous studies related to the cutoff extension, to our best knowledge, show the common case that the electron wave packet of first return only undergo once-oscillation, i.e., the ionized electrons initially depart then immediately return and recombine during the laser field first reverse. Usually the duration of the first return is limited within one optical cycle and the resulting distance of the accelerated electrons is very short, thus, we dub them "short-distance" acceleration mechanisms. In this case, it is hard to push the harmonic cutoff to over \( I_P + 10 U_P \), even if a DC component is imposed to energize the electron-recollision, such as the performances of quasi-static field in Ref. [15] and linear ramp offset in Ref. [16].

In this paper, we propose a method to markedly increase the electron-recollision kinetic energy with multi-cycle two-color pulses. It is shown that a so-called "long-distance" acceleration can be achieved when using a 0.8 \( \mu m \)/13 fs driving pulse in combination with a mid-infrared control field at 10.4 \( \mu m \). While the intensity of the control field is 8% of the driving field, the harmonic cutoff is surprisingly extended to \( I_P + 26 U_P \), and the harmonics above \( I_P + 15 U_P \) are emitted almost in phase, leading to a smooth supercontinuum with the bandwidth of 11\( U_P \). At the same time, we can obtain supercontinuum with variable cutoff by slight adjustment of the two-color field intensity, which allows the production of isolated sub-100 as pulse with tunable central wavelength from ultraviolet to the whole water-window domain.

2. Result and discussion

In order to demonstrate our scheme, we first investigate the HHG process in terms of the classical three-step model [4], which provides a deep insight into the physics. In our simulation, the synthesized field is given by \( E(t) = E_0 f(t) \cos(\omega_0 t + \varphi) \hat{x} + E_1 \cos(\omega_1 (t - \tau) - 0.5 \pi) \hat{x}, 0 \leq t \leq T \).

\( \hat{x} \) is the polarization vector. \( E_0, E_1 \) and \( \omega_0, \omega_1 \) are the amplitudes and frequencies of the driving field at 0.8 \( \mu m \) and control field at 10.4 \( \mu m \), respectively. \( f(t) = \sin^2(\frac{t}{T}) \) is the pulse profile, and \( T = 13 T_0 \) is the pulse duration corresponding to 13 fs full width at half maximum (FWHM). Here, \( T_0 \) denotes the optical cycle of the driving pulse. \( \tau \) is the time-delay between the driving field and the control field, and the carrier-envelop phase \( \varphi \) is set to 0. We choose \( 3 \times 10^{14} \) W/cm\(^2\) and \( 2.4 \times 10^{13} \) W/cm\(^2\) as the intensity of the driving laser and the control pulse, respectively. In this condition, the intensity ratio between the control field and the driving field is 8%. Since the wavelength of the control field (10.4 \( \mu m \)) is much greater than that of the driving field (0.8 \( \mu m \)), and the driving pulse is entirely within one-cycle of the control field. The control field has no contribution to the ponderomotive energy [22], and the ponderomotive energy \( U_P = E_0^2 / 4 \omega_0^2 \) is calculated to be 18 eV.

The classical HHG process from helium driven by the 13 fs laser pulse alone is shown in Fig. 1. Figure 1(a) plots the time evolution of the driving field. Figure 1(b) shows the depen-
Fig. 1. (a) The time evolution field-form of a 0.8 μm/13 fs driving pulse and (b) the corresponding classical ionization (blue dots) and recombination (red circles). The intensity of the driving pulse is $3 \times 10^{14}$ W/cm².

Fig. 2. (a) The schematic description of time-domain field distribution by a 0.8 μm/13 fs driving field (red solid line) in combination with an 10.4 μm control field (blue dashed line) and (b) the corresponding classical ionization (blue dots) and recombination (red circles). The intensity of the driving field and the control field are $3 \times 10^{14}$ W/cm² and $2.4 \times 10^{13}$ W/cm², respectively.

dence of kinetic energy on the ionization (blue dots) and recombination (red circles) times. As shown in Fig. 1(b), within every emission cycle, each harmonic contains two ionization times and two recombination times, which correspond to two branches in each peak. The branch with earlier ionization but latter recombination is named long path, and the other one with later ionization and earlier emission is named short path. Since the maximum kinetic energy of the highest peak ($P_1$) is very close to that of the second-highest peak ($P_2$), it is difficult to be used to generate a supercontinuum.

Next, we focus on the case when the mid-infrared control with time-delay of 0 is added. Figure 2(a) illustrates the schematic of time-domain field distribution in our method. The red
Fig. 3. (a) The classical motion of the sample electron ionized at 5.36\(T_0\) as \(\tau=0\) (red circles), \(\tau=-0.5T_0\) (gray circles), \(\tau=-1T_0\) (blue circles), \(\tau=-1.5T_0\) (purple circles) and \(\tau=-2T_0\) (green circles), respectively. (b) The maximum kinetic energy dependent on the time-delay of \(\tau=0\), \(\tau=-0.5T_0\), \(\tau=-1T_0\), \(\tau=-1.5T_0\) and \(\tau=-2T_0\). Other parameters are the same as in Fig. 2.

solid line and the blue dashed line represent the driving field and the control field, respectively. Figure 2(b) presents the curves of the kinetic energy versus the ionization (blue dots) and recombination (red circles) times. As can be seen from Fig. 2(b), two major emission events take place near the peaks labeled \(P'_1\) and \(P'_2\). The maximum kinetic energy of \(P'_1\) reaches \(26U_P\), while that of \(P'_2\) reaches \(15U_P\). Additionally, it is the left branch that contributes to the emission between \(P'_2\) and \(P'_1\). From these features we can conclude that, under the mid-infrared control, the harmonic cutoff is extended to \(1P+26U_P\), and the harmonics over \(1P+15U_P\) are almost locked in phase and emit once, i.e., become supercontinuous. Another interesting phenomenon presented in Fig. 2(b) is that the recombining electrons near \(P'_1\) experience much longer time in continuous state. This phenomenon can be addressed that the added mid-infrared field assists these electrons to surmount once-oscillation. Thus, the electronic oscillating period is pushed to a few optical cycles, forming a long-distance acceleration channel.

To clearly understand how this long-distance mechanism affects the electron-recollision, we further investigate the classical electron motion corresponding to the harmonic process. Here, only the motion of the electrons of the first return is considered. In the range between \(P'_2\) and \(P'_1\), the recombining electrons can be traced back to the ionization from 5.33\(T_0\) to 5.38\(T_0\) [see the inset in Fig. 2(b) for details]. For convenience, the sample electrons ionized at 5.36\(T_0\) are chosen to be computed. We plot the results in Fig. 3(a) by red circles. Because the control field changes its direction at \(t=6.5T_0\), we define the direction from 0 to 6.5\(T_0\) (from 6.5\(T_0\) to 13\(T_0\)) as \(-x (+x)\) hereafter. For the sampled point of 5.36\(T_0\), the ionized electrons will be accelerated apart from nucleus along \(-x\). After the synthesized field changing its direction along \(+x\) in the next half cycle, the electronic acceleration reverses its direction. However, the electrons can not return to the nucleus in the neighboring half cycle. It can be explained that the control field direction has been changed at 6.5\(T_0\) (from \(-x\) to \(+x)\), thus the symmetry of the driving field relative to the point of 6.5\(T_0\) is broken. This situation continues until the time of about 9\(T_0\), close to the peak region of the mid-infrared field in \(+x\) direction. At this time, the control field has enough energy to resist the fast alternation of the driving field, then the electrons will be accelerated to return back to the nucleus. From our calculations the recombination time of the sample electron is about 10.2\(T_0\). Note that the control field does not change its direction (always along \(+x\)) during the whole return time, and the duration of the return is much longer than one half cycle.
and simultaneously covers the peak region of the control field. That is to say, the acceleration and total acceleration time of those electrons are greatly increased. Therefore, the recombining electrons gain much more kinetic energy, and the so-called long-distance acceleration channel is erected successfully. In addition, we trace the electrons ionized before $5.33T_0$ (from $5.15T_0$ to $5.33T_0$), and find these electrons can not be driven back to the nucleus due to the lack of driving energy of the synthesized field.

Following, the impact of the time-delay is taken into account for quantitatively analyzing the classical electron-recollision. With respect to the same ionized time at $5.36T_0$, the electron motions in the presence of time-delay at $\tau=-0.5T_0$, $\tau=-1T_0$, $\tau=-1.5T_0$ and $\tau=-2T_0$ are shown in Fig. 3(a) by gray, blue, purple and green circles, respectively. Here, other parameters remain unchanged. According to Fig. 3(a), when the time-delay decreases, both the electronic acceleration time and the acceleration distance reduce. As decreasing the time-delay to $-2T_0$ or less, the accelerated electrons work on the once-oscillation scheme, being similar to the one-color case, which implies that the long-distance channel is broken. In view of the influence of time-delay on HHG cutoff, we turn to computing maximum kinetic energy dependent on aforementioned parameters. In the case of $\tau=0$, $\tau=-0.5T_0$, $\tau=-1T_0$, $\tau=-1.5T_0$ and $\tau=-2T_0$, the corresponding maximum kinetic energy is calculated to be $26U_P$, $20.6U_P$, $14.8U_P$, $9.4U_P$ and $6.7U_P$, respectively. These values are plotted in Fig. 3(b). One can see that the maximum kinetic energy almost linearly increase with increasing the time-delay.

To confirm the classical sketch above, we perform the quantum calculations by numerically solving time-dependent Schrödinger equation through split-operator method [23]. In our simulation, a soft potential model $V(x) = Z/\sqrt{\alpha + x^2}$ with $Z = 1$ and $\alpha = 0.484$ is used to match the ground ionization potential of 24.6 eV ($I_P = 0.903$ a.u.) for the real Helium atom. The harmonic spectrum of single-atom response is shown in Fig. 4(a) by green curve, and the parameters are the same as those in Fig. 2. In our calculations, the log scale is adopted. According to Fig. 4(a), the harmonic spectrum shows reasonable structure as expected in our classical prediction. In detail, the harmonics are irregular for the low orders and fascinatingly become regular and smooth from 192-order (295 eV) to 320-order (493 eV), i.e., from $I_P+15U_P$ to $I_P+26U_P$. Since the full description of harmonic process require solving the laser-atom interaction at macroscopic level. We further investigate the harmonic spectrum after three-dimensional (3D) propagation through a 0.5 mm helium gas jet with a density of $0.3 \times 10^{18}$ atoms/cm$^3$. The calculation details can be found in Ref [24]. In this simulation, the driving and the control pulses are assumed as Gaussian.
Fig. 5. (a) The supercontinuum generation with different time-delay of 0 (green solid), -100 as (pink dot) and +100 as (black dot). (b) The supercontinuum generation with different intensity ratio of 7% (red dashed), 8% (green solid) and 9% (gray solid). The supercontinuum generation with different intensity of (c) $2.5 \times 10^{14}$ W/cm$^2$, (d) $3.5 \times 10^{14}$ W/cm$^2$ and (e) $4 \times 10^{14}$ W/cm$^2$, and the corresponding intensity ratio is 9%, 7% and 6%, respectively.

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. The pink curve in Fig. 4(a) shows the continuous parts of macroscopic harmonic spectrum (on-axis) after 3D propagation. By comparison, it is found that the yield of the harmonics over the whole continuous range is enhanced by three to four orders of magnitude after propagation.

For deep understanding of the spectral structures shown in Fig. 4(a), the emission time of the harmonics in terms of time-frequency analysis are present in Fig. 4(b). As shown in Fig. 4(b), two main emission peaks contribute to the harmonics, and the maximum values of the highest peak and the second-highest peak are 320-order and 192-order, respectively. In particular, for the highest peak only the left branch survives. These characteristics agree well with the classical description in Fig. 2(b), and simultaneously indicate that the harmonics between 192-order and 320-order display supercontinuum, which is consistent with the depiction in Fig. 4(a).

It has been suggested that the spectral characteristics of HHG can be significantly affected by the relative time-delay and intensity of two-color field [15]. For this reason, we aim to discuss the influences of the time-delay and the intensity fluctuation. Our calculations unpleasantly imply that, when the time-delay exceeds ±200 as, the continuous harmonics in the plateau can not be obtained. Fortunately, in the current laser technology, the time-delay can be precisely controlled in dozens of attosecond by a piezoelectric translator [25]. Thus, we only consider the time-delay of ±100 as. The HHG spectrum with time-delay of 0, -100 as and +100 as are shown in Fig. 5(a) by green solid, pink dot and black dot, respectively. Figure 5(a) shows that harmonic modulations in the supercontinuum region are negligible with slightly changing the time-delay within 100 as. Then, the time-delay is set as 0 and the intensity ratio is scanned. The results indicate that if the ratio exceeds the range from 7% to 9%, there are some modulations in the supercontinuum, which lower the Signal to Noise Ratio (SNR) of the generated attosecond-pulse. In this case, for some chosen region in the supercontinuum, the intensity of the synthesized satellite pulse reaches 2% of the main pulse. Thus, the regular supercontinuum generation prefers the ratio range from 7% to 9%. In fact, if the allowed SNR set at 10, i.e., the
Fig. 6. The temporal envelopes of the water-window attosecond-pulses from the harmonics of (a) 190-220, 220-250, 250-280 and 280-310, and (b) the pulse from 260-300-order and its phase. The laser parameters are the same as the pink spectrum in Fig. 4(a).

intensity of the satellite pulse is not more than 10% of the main pulse, the preferred intensity ratio can be extended to the range from 5.5% to 11.5%. Figure 5(b) presents the supercontinuum with ratio of 7% (red dashed), 8% (green solid) and 9% (gray solid), respectively. A comparison of the supercontinuum reveals some meaningful features. In detail, as increasing the intensity ratio the harmonic cutoff will extend, whereas the harmonic efficiency near the cutoff will decrease. Subsequently, we choose the laser intensity at $2.5 \times 10^{14}$ W/cm$^2$, $3.5 \times 10^{14}$ W/cm$^2$ and $4 \times 10^{14}$ W/cm$^2$, respectively, and scan their ratio in the same way. The corresponding range for regular supercontinuum is from 8% to 10%, from 6% to 8% and from 5% to 7%, respectively. The supercontinuum of the three intensities are plotted in Fig. 5(c), 5(d) and 5(e) in turn, where the intensity ratio corresponds to 9%, 7% and 6%, respectively. It is clear that synchronous adjustment of the two-color field intensity can achieve different spectrum region that involves the area of ultraviolet and soft x-ray. Particularly, by this approach, the famous water-window (185-352-order for 0.8 μm) is obtained successfully.

The generated supercontinuum (the pink curve) in Fig. 4(a) is investigated qualitatively, by means of the application of a square window with a width of 30 harmonics (47 eV). The temporal envelopes of the attosecond-pulses synthesized from the harmonics of 190-220, 220-250, 250-280 and 280-310-order are shown in Fig. 6(a). The pure isolated 107 as pulses with tunable central wavelength are directly obtained. To further shorten the duration of the attosecond-pulse, one can superpose more harmonics. Figure 6(b) shows an isolated 80 as pulse from 260-300-order without any chirp compensation. Its phase, which implies a certain number of chirp in the temporal envelope, is also presented in Fig. 6(b). It is worth mentioning that the present 198 eV bandwidth of the supercontinuum can support the pulse duration below 24 as (one atomic unit of time) by proper chirp compensation. Such a short isolated pulse is most valuable in probing and controlling the electronic dynamics inside atoms and molecules.

Finally, we further extend our scheme by using more longer driving pulse. The 0.8 μm driving pulse with the duration of 20 fs, 25 fs and 30 fs is discussed, respectively. It is found that when the driving pulse is entirely within one-cycle of the control pulse, the long-distance acceleration mechanism can work as our discussions above. The wavelength of the control pulse corresponds to 16 μm, 20 μm and 24 μm, respectively. We plot the HHG spectrum in Fig. 7(a), 7(b) and 7(c), whose intensity ratio is 7%, 5% and 4%, respectively. Where the other parameters are the same as those in Fig. 2. As shown in Fig. 7, the HHG presents broadband supercontinuum and its cutoff is also extended to water-window domain. Thus, our scheme is flexible, and can provide a dramatic approach for the cutoff extension and the broadband supercontinuum generation with multi-cycle pump pulses.
3. Conclusion

In conclusion, we propose an efficient method to produce broadband supercontinuum via a target helium atom in a combined field of a 0.8 µm/13 fs driving pulse and a mid-infrared control field. The results show that the mid-infrared field plays a robust tool for controlling the quantum acceleration path in long-distance mode. The harmonic spectrum is significantly extended to $I_P + 26U_P$ and the harmonics over $I_P + 15U_P$ are almost synchronically emitted for once, leading to a supercontinuum with the bandwidth of $11U_P$. On the other hand, we have obtained different spectrum region by synchronously adjusting the two-color field intensity, which enables the production of isolated attosecond-pulse with spectral bandwidth covering the whole water-window domain. Experimentally, the high mid-infrared field can be carried out with an 10.4 µm multi-cycle laser pulse. One can use tuned CO$_2$ laser to achieve such a pulse. Moreover, our scheme can extend to more longer driving pulse regime, which provides a dramatic approach for the cutoff extension and the broadband supercontinuum generation with multi-cycle pump pulses.

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