Broadband xuv supercontinuum generation via controlling quantum paths by a low-frequency field

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We propose a method to generate broadband extreme ultraviolet supercontinuum harmonics by using the combination of a phase-stabilized few-cycle laser pulse and a low-frequency field. It is shown that this method can significantly modulate the quantum paths of the harmonics. The harmonic spectrum is extended to $I_p + 8.2U_p$ and harmonics with a bandwidth of $4U_p$ become continuous. By adjusting the carrier-envelope phase of the few-cycle pulse, a close-to-Fourier-limit sub-50-as pulse is straightforwardly obtained.

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The generation of extreme ultraviolet (xuv) attosecond pulses has opened new fields of time-resolved studies with high precision. An isolated attosecond pulse is an important tool for detecting and controlling the electronic dynamics inside atoms, such as innershell electronic relaxation and ionization by optical tunneling [1]. High-order harmonic generation (HHG) enables the production of ultrashort radiation at the attosecond time scale. Both experimental and theoretical studies have shown the general characteristic of HHG: the harmonic spectrum decreases rapidly at low orders, then exhibits a broad plateau and ends up with an abrupt cutoff at the well-known frequency of $I_p + 3.17U_p$. The physical process of HHG can be well-explained by the three-step model [2]: the 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, it may return to the ground state by recombining with the parent ion. During the recombination, a photon with energy equal to the ionization potential plus the kinetic energy of the recombining electron is emitted. This process is repeated every half-cycle of the laser field, which results in an attosecond pulse train with a periodicity of half an optical cycle. However, an isolated attosecond pulse is needed to operate a pump-probe experiment. How to generate an isolated attosecond pulse has attracted a lot of attention [1,3–6]. If the driving laser pulse is sufficiently short, the highest order harmonics emits once and the cutoff region of the spectrum becomes a continuum, and then an isolated attosecond pulse can be filtered out. This scheme has been experimentally carried out by using a state-of-the-art 5-fs few-cycle laser pulse [1,3]. However, the bandwidth of the harmonics in the cutoff region is less than 20 eV, therefore the duration of the shortest attosecond pulse is about 250 as [1], which is greater than the natural time scale of the electronic process inside atoms (152 as, i.e., the period of electrons in the Bohr orbit of ground-state hydrogens), and then the application of the 250-as pulse is significantly limited. Many efforts are paid to broaden the bandwidth of the attosecond pulse and push the duration to even shorter time. It has been reported that broadband continuous harmonics can be observed using a few-cycle driving pulse with the polarization gating technique [7]. Recently, this scheme has been experimentally achieved by Sansone *et al.* [8]. In their experiment, a single cycle attosecond pulse with the duration of 130 as is generated with the chirp compensation technique. It has been suggested that the bandwidth of the attosecond pulse is more important than the duration in attosecond science [9]. Very recently, it has been theoretically demonstrated that the attosecond pulse bandwidth can be significantly broadened in a two-color laser field [10,11], which leads to isolated sub-100-as pulse generation.

In this paper, we propose a method for broadband xuv supercontinuum harmonic generation. The combination of a phase-stabilized few-cycle driving laser pulse and a low-frequency field is adopted. The harmonic spectrum is significantly extended to $I_p + 8.2U_p$ and the harmonics higher than $I_p + 4.2U_p$ are almost synchronically emitted for once, which results in an xuv supercontinuum with the bandwidth of $4U_p$. By adjusting the carrier-envelope phase of the few-cycle pulse, the supercontinuum spectrum bandwidth can be further broadened, and then a close-to-Fourier-limit sub-50-as pulse is straightforwardly obtained.

In order to demonstrate our scheme, we first investigate the HHG process in terms of the semiclassical three-step model [2], which presents a clear physical picture. The driving pulse is expressed by $E(t) = f(t-\tau)E_0 \cos[\omega_0(t-\tau) + \phi_{CEP}] + E_1 \cos(\omega_1 t)$, where $f(t-\tau) = \sin^2[\pi(t-\tau)/T]$ presents the profile of the few-cycle laser field, here $T = 552$ a.u. corresponding to a duration of 5 fs full width at half maximum. E_0, ω_0, and ϕ_{CEP} are the amplitude, frequency, and the carrier-envelope phase of the few-cycle driving pulse, respectively. τ is the time delay between the few-cycle pulse and the low-frequency field and is always set to zero in our work. E_1 and ω_1 are the amplitude and the frequency of the low-frequency field, respectively. We choose $\omega_0 = 0.057$ a.u. and $\omega_1 = 0.0045$ a.u. corresponding to $\lambda_0 = 800$ nm and $\lambda_1 = 10$ μm, respectively. In our scheme, one half-cycle of the low-frequency field is longer than the duration of the whole few-cycle driving pulse, i.e., the harmonic generation is only affected by one half-cycle of the low-frequency field, therefore the low-frequency pulse can be treated as a planar wave in the simulation for convenience. $E_0 = 0.13$ a.u. and $E_1 = 0.042$ a.u., which correspond to the

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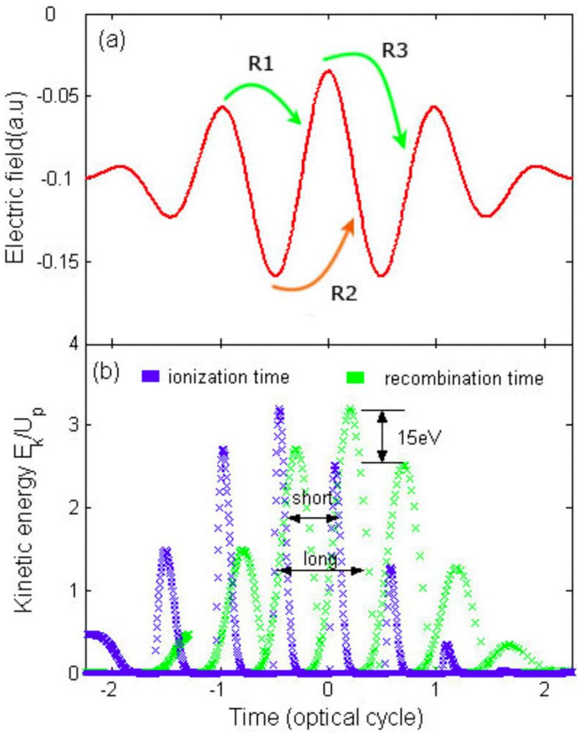

FIG. 1. (Color online) (a) The sketch of electron dynamics in a few-cycle laser pulse with $\phi_{CEP}=0$. (b) The dependence of the kinetic energy E_k on the ionization (blue crosses) and recombination times (green crosses). The laser intensity is 6×10^{14} W/cm^2 and the wavelength is 800 nm.

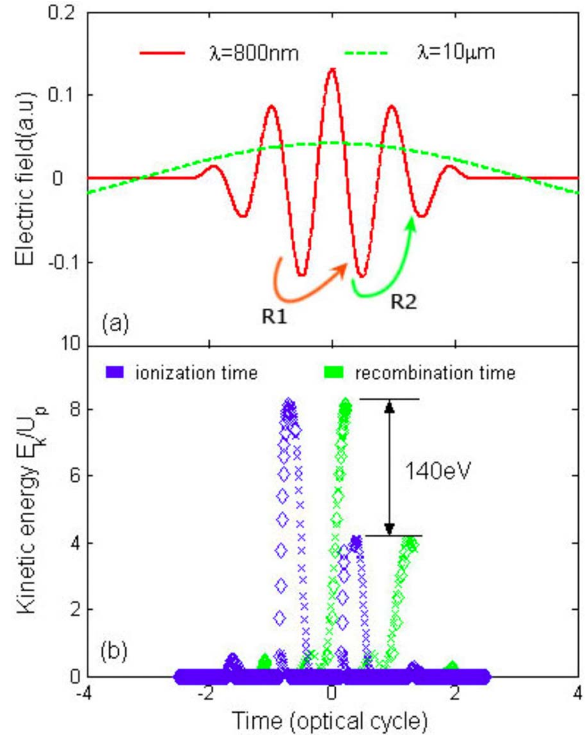

FIG. 2. (Color online) (a) The sketch of electron dynamics in a few-cycle driving pulse ($\lambda=800$ nm) with $\phi_{CEP}=0$ in combination with a low-frequency field ($\lambda=10$ μm). (b) The dependence of the kinetic energy E_k on the ionization (blue crosses for first return and blue diamonds for second return) and recombination times (green crosses for first return and green diamonds for second return). The intensities of the driving pulse and the low-frequency field are 6×10^{14} and 6×10^{13} W/cm^2, respectively.

intensity of 6×10^{14} and 6×10^{13} W/cm^2, respectively. Then the intensity of the controlling field is 10% of the driving pulse. The ponderomotive energy $U_p=E_0^2/4\omega_0^2$ is calculated to be 36 eV. Note that $\omega_0 \gg \omega_1$, and the few-cycle pulse envelope is entirely within one half-cycle of the low-frequency field, then the low-frequency field does not change its direction during the few-cycle pulse, thus the low-frequency field has no contribution to the ponderomotive energy [12].

Figure 1(a) illustrates the sketch of the HHG process from atoms driven by a few-cycle laser pulse with $\phi_{CEP}=0$. As shown in the figure, the driving pulse contains only two optical cycles, thus the electron is mainly ionized near the peaks of the electric field and forms three dominant returns (marked as R1, R2, and R3). Figure 1(b) presents the dependence of the kinetic energy E_k on the ionization (blue crosses) and recombination times (green crosses). It is shown that there are two classes of trajectories corresponding to the same energies of the returning electrons in each half optical cycle. The first trajectory with earlier ionization but later recombination times is called the long trajectory, and the other one with later ionization but earlier emission times is called the short trajectory. The maximum kinetic energies of R1, R2, and R3 are $2.71U_p$, $3.17U_p$, and $2.52U_p$, respectively. Therefore only R2 contributes to the harmonics higher than $I_p+2.71U_p$, which can be filtered out to generate an isolated attosecond pulse. However, the bandwidth of this pulse is only 15 eV, and the minimum pulse duration is about 270 as.

The electron trajectories for HHG driving by a few-cycle laser pulse can be significantly modulated by adding a low-frequency field. The sketch of the electron dynamics is shown in Fig. 2. The red solid line and the green dashed line in Fig. 2(a) present the few-cycle laser pulse and the low frequency field, respectively. The electron is no longer ionized near the peak of the driving field, instead it is ionized from −0.8 to −0.4 T and from 0.2 to 0.6 T forming only two returns (marked as R1 and R2). In every optical cycle of the few-cycle pulse (T_0), the low-frequency field does not change its direction. In the half cycle of $t=0$, the total field is greatly enhanced and the electron accelerated here gains much more energy (R1), therefore the harmonic spectrum can be significantly extended, i.e., the cutoff region is much higher than the well-known value of $I_p+3.17U_p$. From our calculation the maximum return time of R1 for the second return is about $1T_0$, therefore the second return should be considered in this case. Figure 2(b) shows the dependence of the kinetic energy on the ionization and recombination times. The ionization and recombination times for the first return are presented by blue and green crosses, respectively, and that for the second return are presented by blue and green diamonds, respectively. As shown in this figure, for R1 there is only one main trajectory both for the first and the second return. More surprisingly, the recombination times for the

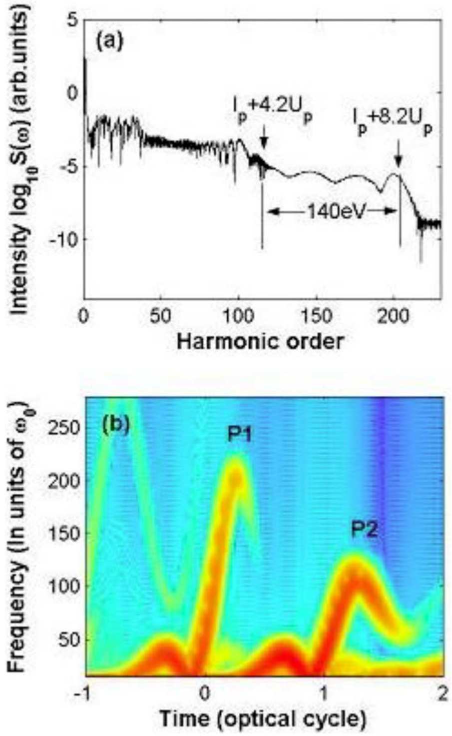

FIG. 3. (Color online) (a) The harmonic spectrum and (b) the time-frequency distribution of HHG in the combination of a few-cycle laser pulse and a low-frequency field. The parameters are the same as in Fig. 2(b).

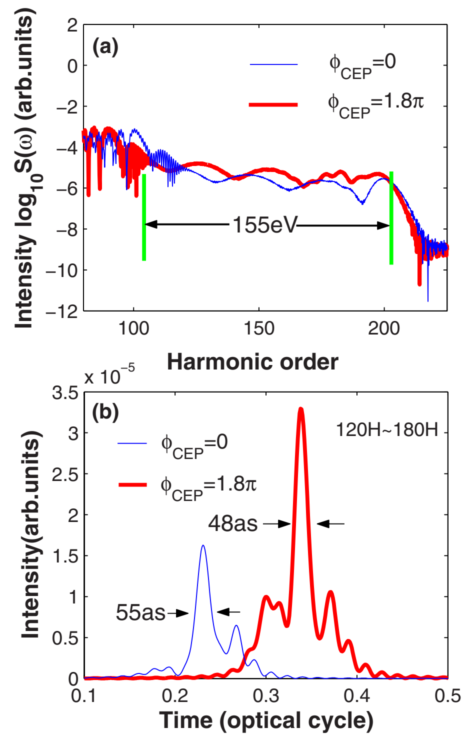

FIG. 4. (Color online) (a) The harmonic spectrum of HHG in the combination of a few-cycle laser pulse and a low-frequency field for $\phi_{CEP}=0$ (blue line) and $\phi_{CEP}=1.8\pi$ (red line). (b) The temporal profile of the isolated attosecond pulse by superposing the harmonics from 120 to 180. Other parameters are the same as in Fig. 2(b).

second return are almost the same with that for the first return. The maximum kinetic energy of R1 reaches $8.2U_p$ while that of R2 only reaches $4.2U_p$. Taking into account all the results above, we can conclude that under modulation of the low-frequency field, the HHG spectrum can be extended to $I_p+8.2U_p$, and the harmonics higher than $I_p+4.2U_p$ are almost locked in phase and emit once, i.e., become continuous, which results in a broad supercontinuum spectrum with the bandwidth of 140 eV, and then an isolated 30-as pulse can be obtained in the Fourier transform limit.

Following, we investigate the harmonics and the attosecond pulse generation by numerically solving the time-dependent Schrödinger equation by means of the split-operator method [13]. In our simulation, we use a soft-core potential model $V(x)=-1/(\alpha+x^2)^{1/2}$ and choose the softening parameter α to be 0.484 corresponding to the ionization energy of 24.6 eV for the ground state of a helium atom. Thus it can be treated as a planar wave here. Figure 3(a) shows the harmonic spectrum in the combination of the 5-fs driving pulse and the low-frequency field. One can clearly see that the spectrum shows the structure as expected in our semiclassical approach: it is irregular for the low harmonics and fascinatingly becomes regular and continuous from the 115 harmonic to the 205 harmonic, corresponding to $I_p+4.2U_p$ and $I_p+8.2U_p$, respectively. In order to further understand the spectrum structure shown in Fig. 3(a), we also investigate the emission time of the harmonics for the case in terms of the time-frequency analysis method [14]. The result is shown in Fig. 3(b). As shown in this figure, there are two main peaks contributing to the harmonics (marked as P1 and P2), and the maximum harmonic orders for P1 and P2 are about 115 and 205, respectively. One can clearly see that the harmonics for P1 are almost locked in phase, which is in great agreement with the semiclassical approach shown in Fig. 2. For the harmonics below 115, the interference of P1 and P2 leads to the irregular spectrum structure which is shown in Fig. 3(a).

It has been reported that HHG and attosecond pulse generation driven by a few-cycle pulse can be significantly affected by their carrier-envelope phase [15]. In this work, we also investigate the HHG by adjusting the carrier-envelope phase of the few-cycle driving pulse and find that the bandwidth of the continuous harmonics is even broadened when $\phi_{CEP}=1.8\pi$. The result is shown in Fig. 4(a) by a red line. For comparison, we also present the HHG spectrum for $\phi_{CEP}=0$ (blue line). As shown in this figure, the continuous harmonic range is from 105 to 205 harmonic, corresponding to a bandwidth of 155 eV, which is 15 eV broader than that for $\phi_{CEP}=0$. Further, we consider the isolated attosecond pulse generation for $\phi_{CEP}=0$ and $\phi_{CEP}=1.8\pi$. Figure 4(b) shows the temporal profile of the isolated attosecond pulse by selecting the harmonics with a bandwidth of 95 eV (from 120 to 180). For $\phi_{CEP}=0$ (blue line), a close-to-Fourier-limit pulse with the duration of 55 as is directly obtained; and for $\phi_{CEP}=1.8\pi$ (red line), the pulse duration is even shorter (48 as). Moreover, the intensity is two times higher than that for $\phi_{CEP}=0$.

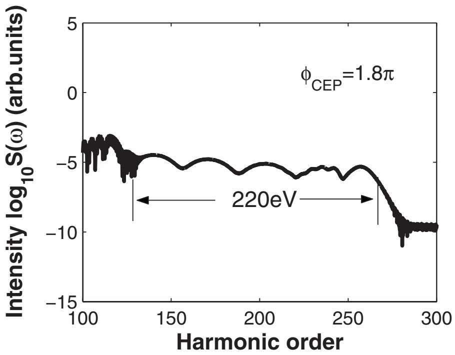

FIG. 5. The harmonic spectrum of HHG in the combination of a few-cycle laser pulse and a low-frequency field with the intensity of 8×10^{14} and 8×10^{13} W/cm^2.

Note that the supercontinuum can be obtained with the intensity ratio ranging from 8% to 15%, and 10% is the optimal value to obtain the most broad continuous spectrum. If we keep this ratio and increase the intensities of these two fields, the bandwidth of the supercontinuum can be broadened to more than 200 eV. Figure 5 shows the harmonic spectrum in the combination of the few-cycle laser pulse and the low-frequency field with the intensities of 8×10^{14} and 8×10^{13} W/cm^2. In this case, the bandwidth of the continuous harmonics reaches 220 eV. By compensating the chirp with a proper media, a pulse with the duration of about 20 as will be obtained in the Fourier transform limit, which is a powerful tool to study and control the ultrafast electronic processes. Experimentally, this scheme can be carried out with a 10 mJ, 40 fs at 1 kHz Ti:sapphire laser system. The laser beam can be split into two beams: one is compressed to 5 fs pulses through a cascade filamentation compression technique, and another is used to produce such low-frequency laser pulses via an optical parametric amplifier (OPA). In our method, it is worthwhile to consider how long the helium atom can survive in the low-frequency field alone, which determines how long a low-frequency pulse can be used. From our calculation the atom can hardly be affected by the low-frequency field alone, even with a pulse duration longer than 1 ps.

In conclusion, we present a method to generate broadband extreme ultraviolet supercontinuum harmonics by using the combination of a phase-stabilized few-cycle laser pulse and a low-frequency field. It is achieved via significantly modulating the quantum paths of HHG. The harmonic spectrum can be extended to $I_p+8.2U_p$ and the harmonics higher than $I_p+4.2U_p$ are almost synchronically emitted for once, which results in an xuv supercontinuum with the bandwidth of $4U_p$. By adjusting the carrier-envelope phase of the few-cycle pulse, the supercontinuum spectrum bandwidth can be further broadened, and then a close-to-Fourier-limit sub-50-as pulse is straightforwardly obtained. By simultaneously increasing the intensity of the few-cycle laser pulse and the low-frequency field, the continuous harmonic bandwidth can be broadened up to 220 eV, and by compensating the chirp the pulse duration can be less than 1 a.u. (24 as).

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