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Attosecond ionization gating for isolated attosecond electron wave packet and broadband attosecond xuv pulses

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An attosecond ionization gating is achieved using a few-cycle laser pulse in combination with its second harmonic. With this gating, the generation of the electron wave packet (EWP) is coherently controlled, and an isolated EWP of about 270 as is generated. An isolated broadband attosecond extreme ultraviolet pulse with a bandwidth of about 75 eV can also be generated using this gating, which can be used for EWP measurements as efficiently as a 50-as pulse, allowing one to measure a wide range of ultrafast dynamics not normally accessible before.

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High harmonics are generated when atoms or molecules are irradiated by an intense laser field [1]. This process is well understood with the recollision model [2]. In detail, an electron wave packet (EWP) is first created through tunneling ionization, then is accelerated in the laser field, and finally recombines with the parent ion. This process can also be viewed as an electron interferometer [3], which opens the door to attosecond science. The interferogram has been used to map attosecond EWP motions [4], to image the molecular orbitals with tomographic metrology [5], and to measure *in situ* the return time of recollision EWPs [3]. Moreover, the recollision EWP has been used to probe the ultrafast dynamics in atoms and molecules [6] and light waves [7] with an attosecond resolution.

On the other hand, attosecond extreme ultraviolet (xuv) pulses are produced by filtering several harmonics [8–10]. The attosecond xuv pulse allows one to trace the ultrafast dynamics inside atoms and molecules in real time. The straightforward attosecond metrology prefers an isolated attosecond xuv pulse [10,11], which can be generated using a few-cycle laser pulse [12]. Note that the restriction on the few-cycle laser pulse can be relaxed [13,14]. However, the bandwidth of the supercontinuous harmonics is only $\sim$10 eV; thus the shortest attosecond xuv pulse is about 250 as [9]. The recent result suggests that the bandwidth of the attosecond pulse is a more important parameter than the duration for attosecond science [15]. Therefore, intense research is currently afoot to broaden the attosecond pulse bandwidth and push the pulse duration to even shorter time [16–18].

The gating technique is an efficient way to coherently control the ultrafast process and has been used in a wide range of measurements. For instance, the polarization gating technique makes the EWP miss the parent ion on its return [19,20], which has been used to measure the EWP motion with attosecond resolution [21] and to generate an isolated broadband attosecond xuv pulse [17,18]. In this Rapid Communication, we propose a different gating technique to coherently control the generation of EWPs, i.e., the ionization process, so we name it ionization gating. This ionization gating is achieved with a few-cycle laser pulse in combination with its second harmonic (SH), and the sketch is shown in Fig. 1. The dotted line in Fig. 1(a) shows the electric field of the few-cycle laser pulse. When atoms are exposed to this field, EWPs are dominantly generated at the times t_1, t_2, and t_3. By synthesizing the SH [the dashed line in Fig. 1(b)], the generation of EWPs can be coherently controlled, forming an ionization gate. By adjusting the SH, we can open the gate at the pulse peak (t_2), since the fundamental field is enhanced by the SH, and close the gate at t_1 and t_3, since the fundamental field is weakened. Then an isolated attosecond EWP will be generated, which may be used to probe the ultrafast dynamics. In addition, this gating technique also can be used to generate an isolated broadband attosecond xuv pulse, allowing one to produce a single-cycle attosecond pulse and to measure a wide range of ultrafast processes not normally accessible before.

To demonstrate the ionization gating technique, we investigate the interaction of helium and a few-cycle laser pulse in combination with its SH. The fundamental laser pulse and its SH are both linearly polarized in the $\hat{\mathbf{x}}$ direction and the electric field is given by $\mathbf{E}(t) = E_0 f(t)\cos[\omega_0(t-T/2)]\hat{\mathbf{x}} + E_1 f(t)\cos[2\omega_0(t-T/2)+\phi]\hat{\mathbf{x}}$. $f(t) = \sin^2(\pi t/T)$ is the pulse envelope and T determines the pulse duration. E_0 and E_1 are the amplitudes, ω_0 is the frequency of the fundamental laser, and ϕ is the relative phase. The pulse duration is 5 fs full width at half maximum. The intensity and wavelength of the fundamental laser pulse are 6×10^{14} W/cm^2 and 800 nm; the intensity of the SH is 4% of the fundamental field. In this work, we adopt a laser pulse with an intensity lower than the saturation intensity, which is different from Refs. [14,16]. In this condition, only a few atoms are ionized, and the depletion of neutral atoms can be negligible. Thus the generation of EWPs and high harmonics is determined by the ionization rate [2], which can be calculated with the Ammosov-Delone-Krainov (ADK) theory [23]. The solid line in Fig. 1(a) shows the ionization rate of helium in the fundamental few-cycle laser pulse. One can see that three dominant EWPs are generated. The situation is distinct in the two-color field. The solid line in Fig. 1(b) shows the ionization rate of helium in the two-color field

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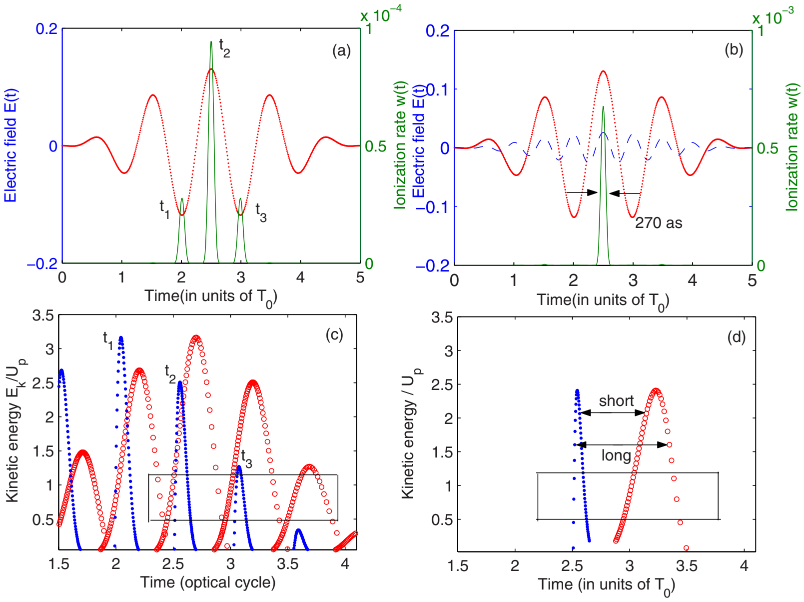

FIG. 1. (Color online) (a) Electric field of the fundamental few-cycle laser pulse (dotted line) and ionization rate (solid line) of helium in this laser field. (b) Electric fields of fundamental few-cycle laser pulse (dotted line) and its SH (dashed line). The solid line is the ionization rate of helium in this two-color laser pulse. (c) and (d) show the dependence of the electron kinetic energy on the ionization (dots) and recombination times (open circles) in the cases of (a) and (b), respectively. The square indicates the spectral window. The parameters are shown in the main text.

with the relative phase of $\phi=0$. One can see that the gate is open at the peak of the laser pulse and an EWP is set free in an interval of about 270 as, the gate is closed at other times and the EWPs are eliminated. In this way, the generation of EWPs is coherently controlled, forming an attosecond ionization gating.

Figure 2 shows the ionization rate in the two-color field at different relative phases. One can see that only one EWP is set free when the relative phase is within $\pm 0.2\pi$, and many EWPs are generated at other phases. In experiments, establishing a dedicated relative phase shift can be done by introducing small amounts of glass. Alternatively, our calculation shows that similar result can also be achieved by adjusting the delay between the two laser pulses. Note that the amplitude of the main EWP is always two orders of magnitude higher than the satellite EWPs when the intensity of the SH varies from 2.5% to 15% of the fundamental laser pulse. In addition, our calculation indicates that the ionization gating will be achieved using a laser pulse with a duration less than 6 fs and an intensity ranging from 3×10^{14} to 1×10^{15} W/cm^2. Note that the laser intensity of 1×10^{15} W/cm^2 is close to the saturated intensity, which indicates a satisfactory yield of electron ionization and high-harmonic generation. On the other hand, the ionization gating is formed with a two-color field and it has been demonstrated that harmonic yield is enhanced in such a field [22].

In the ionization gating, an isolated attosecond electron pulse of about 270 as is obtained [see Fig. 1(b)]. It seems likely that attosecond science will use the attosecond electron pulse and the attosecond photon pulse equally [6]. For instance, with an attosecond electron pulse, the technique of classical electron diffraction can be extended to the attosecond range, allowing one to investigate instantaneous structural changes as well as ultrafast dynamics in atoms and molecules [24]. In contrast to attosecond photon pulses, the attosecond electron pulse is produced with a higher efficiency, and has a higher probability of interaction [6]. In the previous investigations, attosecond EWPs have been used to probe the molecular dynamics [6] and light waves [7] with attosecond resolution.

Moreover, the attosecond ionization gating can be used to coherently control the recollision process and to produce an isolated broadband attosecond xuv pulse. Following, we investigate high-harmonic and attosecond pulse generation by solving the time-dependent Schrödinger equation [4]. The harmonics and attosecond xuv pulses generated in the few-cycle laser pulse in combination with its SH are shown in

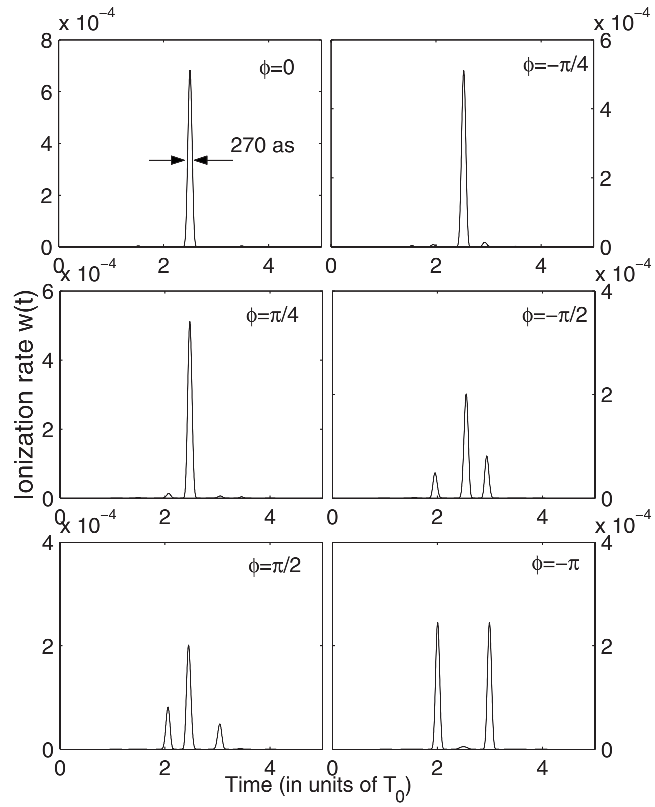

FIG. 2. Ionization rate of helium in a few-cycle laser field in combination with its SH at different relativistic phases.

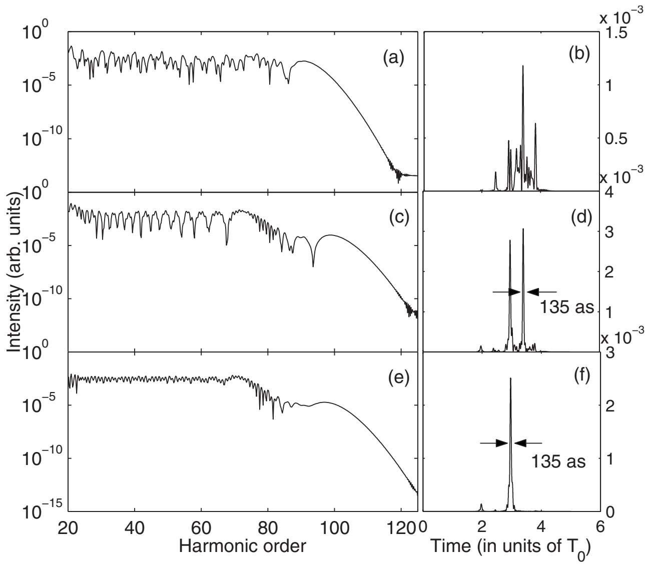

FIG. 3. Harmonics and attosecond pulses generated using the fundamental laser pulse (a), (b) and in combination with its SH (c), (d). The laser parameters are the same as in Fig. 1. In (e) and (f), we show the harmonics and attosecond pulses generated in the fundamental field in combination with its SH by taking account of an intensity variation of 5%.

Figs. 3(c) and 3(d), respectively. The parameters are the same as those in Fig. 1. For comparison, the harmonics and attosecond xuv pulses generated in the fundamental laser pulse alone are shown in Figs. 3(a) and 3(b). To obtain the temporal profile of the attosecond xuv pulse, a square window is applied to the harmonic spectrum; then it is Fourier transformed to the time domain [17]. The width of the spectral window is 33 eV, and the central frequency is $35\omega_0$. As shown in Fig. 3(a), the spectral structure is irregular for the lower harmonics in the plateau and becomes continuous for the harmonics in the cutoff. As shown in Fig. 3(b), the irregular harmonics in the plateau lead to a train of attosecond xuv pulses. Note that the continuous harmonics in the cutoff will lead to an isolated attosecond xuv pulse. However, the bandwidth is only about 10 eV. While in the ionization gating formed by the two-color field, the harmonic spectrum shows a different structure. As shown in Fig. 3(c), the overall spectrum is smooth and regularly modulated for the harmonics through the plateau to cutoff. The temporal profile of the attosecond xuv pulse is shown in Fig. 3(d), which shows a two-burst structure. The above results can be interpreted qualitatively within the semiclassical recollision model [2]. Figure 1(c) shows the dependence of electron kinetic energy on the ionization and recombination times in the fundamental pulse. One can see that EWPs are dominantly generated at the times $t=2.0T_0$, $2.5T_0$, and $3T_0$. After ionization, the EWP oscillates quasifreely in the laser field, accumulating kinetic energy, and finally returns to the parent ion through two quantum trajectories [2]. By filtering the harmonics in the plateau, a train of attosecond xuv pulses is generated. While in the two-color field, the EWP is set free only in a short interval at $t=2.5T_0$ [see Fig. 1(d)], and returns to the parent ion through two quantum trajectories. It takes about a half cycle of the laser field for the short trajectory, and about one cycle for the long trajectory. Therefore, by filtering these harmonics, two attosecond xuv pulses are generated at $t=3.0T_0$ and $3.4T_0$. The pulse duration is about 135 as, close to the Fourier-transform limit [25]. Note that the attosecond pulses show the same temporal profile if we use another spectral window, only the pulse duration is slightly increased to 155 as when a Gaussian window with a width of about 23 eV is used. We also calculated the temporal profile of the attosecond pulse by varying the central frequency of the spectral window. The results clearly show that the emission time of the attosecond pulse originated from the short trajectory increases linearly with increasing harmonic frequency, indicating a positive chirp, and the long trajectory indicates a negative chirp. These time-frequency characteristics are consistent with the results shown in Figs. 1(c) and 1(d), which has also been observed experimentally [3,25]. Due to this chirp, the attosecond pulse duration is limited. In addition, one can see from Fig. 3(c) that there is a regular modulation in the spectrum. This is owing to the interference of the short and long quantum trajectories, analogous to Young's two-slit experiment in the spatial domain.

To generate an isolated attosecond pulse, one of the short or long trajectories must be eliminated. It has been demonstrated that the short and long trajectories depend distinctly on the laser intensity [26]. The long-trajectory component depends sensitively on the laser intensity, and is more likely to experience destructive interference. Hence, the long trajectory will not survive if we sum the harmonic spectra obtained at different laser intensities [26]. Here we calculated harmonic spectra for N pulses with an intensity variation of 5%, and then add the spectra as [26] $d(\omega)=\Sigma_i^N d_i(\omega)/N$ where d_i is the oscillating dipole for the ith pulse. Figures 3(e) and 3(f) show the final harmonic spectrum and attosecond pulse, respectively. As shown in Fig. 3(e), the harmonics are supercontinuous through the plateau to cutoff. It is because the long trajectory is eliminated and the interference is weakened as well. By filtering the supercontinuous harmonics, an isolated attosecond pulse is generated at $t=3T_0$ [see Fig. 3(f)] which corresponds to the short trajectory. In real experiments, the laser intensity will be varied along the propagation axis by focusing the laser beam before the gas jet; then the long trajectory will be eliminated as in our simulation. This method has been implemented and verified experimentally [8,18,25]. One can see from Fig. 3(f) that the attosecond xuv pulse is enhanced by a factor of about 2 in the two-color field compared to Fig. 3(b). In addition, the duration of this attosecond xuv pulse is 135 as, containing only 1.8 optical cycles of the central frequency ($35\omega_0$). Analogously to the few-cycle infrared pulses [12], such few-cycle isolated attosecond xuv pulses may pave the way to investigate and manipulate ultrafast dynamics by varying the attosecond pulse phase in the xuv regime [18]. Further, one can see from Fig. 3(e) that the bandwidth of the supercontinuous harmonics is broadened to 75 eV, an isolated attosecond pulse of about 50 as will be generated in the Fourier-transform limit. However, the harmonic chirp prevents production of the Fourier-transform-limited pulse. It should be emphasized that such a broadband attosecond xuv pulse, even though it is chirped, still allows one to trace a wide range of ultrafast processes not normally accessible before. Since the recent results show that the important parameter for attosecond pulse is not the pulse duration but the bandwidth, long chirped pulses can be used for EWP measurements (and perhaps many other types of attosecond measurement) as effectively as Fourier-transform-limited pulses of the same bandwidth [15]. On the other hand, it has been shown that the positive chirp can be compensated by a material with a negative group delay dispersion [18,25]. Al and Si filters have almost constant transmittance (like the square frequency window used in the above simulation) and a negative group delay dispersion from 20 to 60 eV and 25 to 90 eV [18,27], respectively, which can be used to compensate the harmonic chirp in experiments. With this technique, it is anticipated that an isolated xuv pulse of 50 as can be generated.

We also analyzed the influence of the fluctuation of laser intensity on the broadband attosecond xuv pulse. Supercontinuous harmonics and broadband attosecond xuv pulses are retrieved using a few-cycle laser pulse with an intensity ranging from 2×10^{14} to 8×10^{14} W/cm^2 and a duration less than 6 fs. Moreover, our result still survives when the intensity of the SH varies from 2% to 15% of the fundamental field; and small changes of the absolute phase ($\leq\pm0.1\pi$) of the fundamental field and relative phase ($\leq\pm0.2\pi$) between the SHs do not influence the generation of the broadband attosecond xuv pulse. Note also that the few-cycle laser pulse usually is not clean in experiment, but is surrounded by a weak shoulder pulse. We have also analyzed this effect, and the result shows that the shoulder pulse does not influence our results when the intensity of the shoulder pulse is less than 30% of the main pulse. This laser system is currently available, indicating the demonstration of our scheme.

In conclusion, we propose a gating technique using a fundamental few-cycle laser pulse in combination with its SH to coherently control the generation of an EWP. The EWP is set free only in an interval of 270 as, forming an attosecond ionization gating, and then an isolated attosecond EWP is obtained. In addition, an isolated broadband attosecond xuv pulse with a bandwidth of about 75 eV is generated, and an isolated single-cycle attosecond xuv pulse of 135 as is obtained, which may pave the way for investigation and manipulation of ultrafast dynamics by changing the phase of the attosecond pulse. The pulse duration is anticipated to be reduced to about 50 as after compensating the chirp, allowing one to measure a wide range of ultrafast dynamics not normally accessible before. This attosecond ionization gating is achieved with a two-color field, which is accessible in experiment, and is beneficial for enhancing the harmonic and attosecond xuv pulse yields.

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