Generating coherent broadband continuum soft-x-ray radiation by attosecond ionization gating

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Abstract: The current paradigm of isolated attosecond pulse production requires a few-cycle pulse as the driver for high-harmonic generation that has a cosine-like electric field stabilized with respect to the peak of the pulse envelope. Here, we present simulations and experimental evidence that the production of high-harmonic light can be restricted to one or a few cycles on the leading edge of a laser pulse by a gating mechanism that employs time-dependent ionization of the conversion medium. This scheme enables the generation of broadband and tunable attosecond pulses. Instead of fixing the carrier-envelope phase to produce a cosine driver pulse, the phase becomes a control parameter for the center frequency of the attosecond pulse. A method to assess the multiplicity of attosecond pulses in the pulse train is also presented. The results of our study suggest an avenue towards relaxing the requirement of few-cycle pulses for isolated attosecond pulse generation.

OCIS codes: (190.4160) Multiharmonic generation; (320.7150) Ultrafast spectroscopy; (190.2620) Frequency conversion; (300.6560) Spectroscopy, x-ray.

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Attosecond pulses are currently opening up a new field of time-resolved spectroscopy. The attosecond domain is the natural time-scale of atomic physics (1 atomic unit $\sim$24 as) and is therefore well suited for the temporal tracking of electronic wave-function dynamics and multi-electron correlation, in both valence and core electronic levels of matter. Attosecond pulse production typically utilizes high-harmonic generation [1–4] with multi-cycle [2] or few-cycle, carrier-envelope phase (CEP) stabilized laser pulses [3], possibly augmented by transient polarization control [4], leading to the generation of attosecond pulse trains (APTs) or isolated attosecond pulses (IAPs), respectively. The preferred attosecond mode (pulse train or isolated) depends on the spectroscopic application [5–7]. Many approaches rely on an isolated attosecond pulse [5, 6] or a well-known small number of pulses in the attosecond pulse train, such as a double pulse [7].

Here, we present simulations and experiments supporting an ionization gating mechanism similar to one described theoretically [8] that produces continuum emission of high-harmonics on the leading edge of a driving laser pulse, indicative of an isolated attosecond pulse. Previous studies indicated that laser-induced ionization can be used to shorten the harmonic pulse duration for a single harmonic order [9, 10]. In this study, we observe ionization gating to act on a sub-cycle time-scale, enabling the generation of coherent spectral continua spanning many harmonic orders. The bandwidth of the continuum soft-x-ray radiation produced at the leading edge can be enhanced beyond that of the traditional method [11] using equally short driver pulses, while the center photon energy can be tuned by varying the CEP; this means that broadband and tunable isolated attosecond pulses can be produced. Based on our findings, we also discuss the possibility to remove the absolute requirement of a few-cycle pulse for isolated attosecond pulse production. Finally, we present a method to experimentally measure the multiplicity of attosecond pulse production (number of pulses in the attosecond pulse train).

Our method is based upon the pioneering work of Haworth *et al.* [12]: if the driving laser pulse consists of several half cycles, one observes several local spectral maxima located at photon energies that correspond to the harmonic cutoff emission (attosecond pulses) generated from individual half cycles (termed half-cycle cutoffs) in the spectrum, enabling determination of the absolute CEP of the laser pulse [12]. We show here that this analysis can be extended to include ionization of the medium. This additional information is essential to observe ionization-gated high-harmonic generation on the single-optical-cycle level.

In Fig. 1, the calculated half-cycle harmonic cutoff (HCO) energies are plotted as a function of CEP for a 3-cycle full-width-at-half-maximum (FWHM) laser pulse using a classical calculation [1], taking into account the full transient of the electric field on the leading edge

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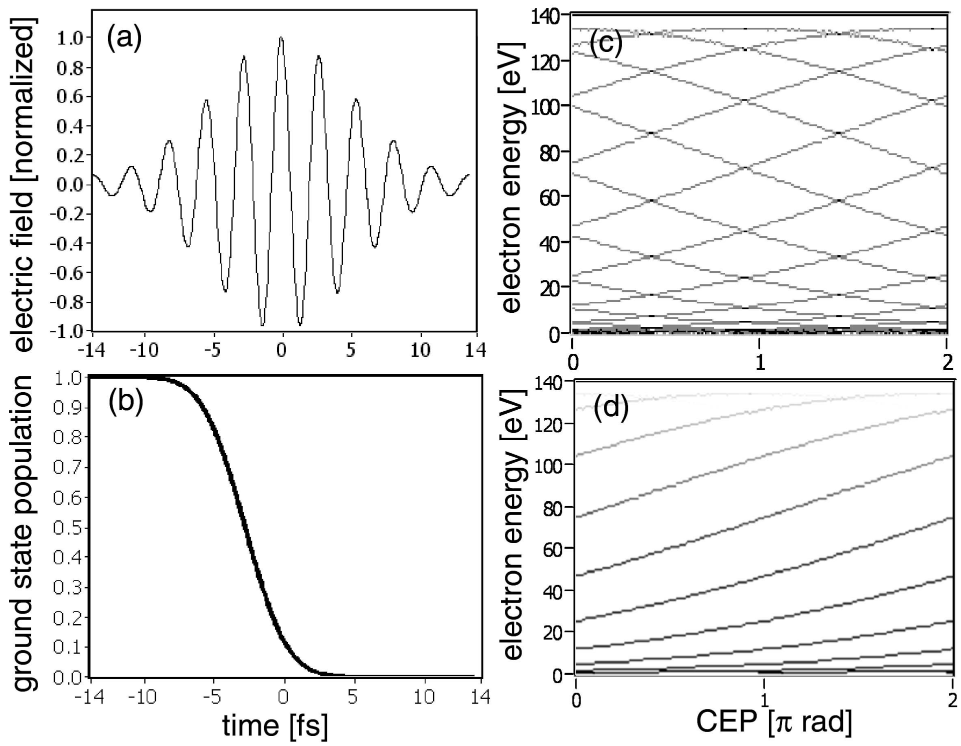

Fig. 1. Half-cycle resolved cutoff energy calculation (kinetic energies of the recombining electron) for a 3-cycle FWHM pulse shown in (a) by including depletion of the harmonic medium (b). For illustration purposes, an error-function shape is assumed in the model as an approximation of a step-like function that would be expected for tunnel ionization. HCO positions were calculated in (c) by neglecting and in (d) by including depletion. The intensity of the lines indicates the spectral intensity at the HCO position.

of the pulse. As the CEP is modulated, these cutoffs shift in energy due to the change of the accelerating electric field underneath the pulse envelope. The highest half-cycle cutoffs occur close to CEPs of 0 and π, corresponding to the well-known cases of the cosine and minus-cosine pulses, respectively, as discussed in [11]. Using the HCO plots (Fig. 1(c)) the emergence and bandwidth of the continuous harmonic spectrum in [11] can be explained by the difference between the highest-energy HCO curve to the next lower ones. This difference is largest at the CEP of π (cosine pulse) and smallest (zero) at 0.5π (sine pulse), where the two highest HCO curves cross. Spectral modulation (odd-order harmonic peaks) in the case of the sine pulse then arises from two attosecond pulses produced at successive half-cycles having the same HCO energy. We return to this interference when we discuss the multiplicity of attosecond pulses in the pulse train. Also, note that the bandwidth available for an isolated attosecond pulse can be extracted from the HCO plot by considering the energy difference between the highest and the second-highest HCO values for a fixed CEP.

If laser-induced ionization of the harmonic medium occurs on the leading edge of the driver pulse and is taken into account in the classical model (Fig. 1(b)), the CEP plot is modified so that the quasi-axial symmetry with respect to π (Fig. 1(c)) is broken and only the increasing slopes of the CEP curves are retained (Fig. 1(d)). This result can be understood intuitively; the symmetry is created by the fact that a given half-cycle of the laser pulse grows in amplitude as

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it approaches the peak of the pulse envelope (with increasing CEP) and thus produces progressively higher electron energies, whereas after passing the peak of the pulse at π, the half-cycle will decline in amplitude and therefore produce progressively less energetic recombining electrons. However, this time-symmetric process can be observed only if the medium is not ionized and perfect phase-matching is achieved at all times during the driver pulse. If ionization is present, more efficient production of harmonics occurs on the leading edge of the driving pulse when the degree of ionization is minimal, and harmonic production is inhibited at later times when the ionization level is too high. In addition to the effect presented by Cao *et al.* [8], where they only simulate the single-atom response, we note that other mechanisms such as ionization-induced phase mismatch and defocusing [13] can also inhibit the harmonic generation process at a certain time during its leading edge, even for much less than full depletion (5-10%) of the ground state. We now turn to the experiments that test the predictions of leading-edge harmonic production via ionization gating.

Our experimental system consists of a Ti:sapphire multipass amplified laser system, delivering carrier-envelope phase stabilized (root-mean-squared fluctuation rms $\sim$0.25 rad) 0.8 mJ pulses of 25 fs FWHM duration with a repetition rate of 3 kHz. These pulses are focused into a neon-filled hollow-core fiber (inner diameter 250 μm) and sent through a chirped mirror setup for pulse compression to 5-7 fs at a center wavelength $\sim$720 nm. High harmonics are produced in a vacuum system, using a 2 mm long gas cell ($\sim$100 μm diameter holes on laser input and output side) filled with 180 mbar of Ne gas . As an ionization gate is expected to be significant at higher driver intensities, the driver pulse was focused very close to the gas jet. A home-built spectrometer consisting of two zirconium filters of 200 nm thickness each, a Si_3N_4 based transmission grating with a period of 100 nm, and a soft-x-ray sensitive backside-illuminated (thinned) charge-coupled device (CCD) detector was used to detect the high-harmonic radiation. The spectrometer is arranged in such a way that only the center portion of the harmonic beam is spectrally analyzed.

In Fig. 2(a), we show the high-harmonic spectra observed for several different values of the CEP of our laser pulse. A group of intense harmonics is located in the region of 80-100 eV, and a less intense group at around 100-120 eV, both of which shift to shorter wavelengths with increasing phase. The overall spectral minimum at 100 eV and the reduced spectral sensitivity beyond 100 eV are due to the absorption edge of Si in the Si_3N_4 grating that is used for spectral analysis. We obtain the spectra shown in Fig. 2(b) after filtering out (by means of Fourier transform) the odd-harmonic modulating component of the spectrum at twice the laser photon energy ($2\hbar\omega_L \cong 3$ eV), in order to extract the more slowly varying local intensity maxima caused by the half-cycle cutoffs. We observe another faster modulation with a period of $\sim$1.5 eV, which will be discussed below for determining the attosecond pulse multiplicity. This modulation can be filtered out analogously to retain only the HCO maxima structure (as was done in [12]). The resulting spectra are divided by the CEP-averaged harmonic spectrum (dotted line in Fig. 2(b)) in order to remove fast modulations in the spectral sensitivity of the spectrometer that could otherwise obscure the HCO positions. The spectra are then plotted as a function of relative CEP (Fig. 3(a)). Here, we take the HCO periodicity of π into account and only show the spectrum from 0 to π relative CEP. The black dots mark the positions of the resulting HCO maxima.

If we require the measured half-cycle cutoffs to fit the global cutoff of the laser pulse (intensity maximum at the peak of the pulse), a fit to a 4 fs gaussian pulse (Fig. 3(b)) returns the closest result. The free parameters in the fit are peak intensity, pulse duration, and CEP. This fit does not describe the observed behavior of the HCO energies. While the fit shows a saturation at the highest photon energies, the measured photon energies increase linearly up to the global cutoff at 120 eV. However, by comparing the data with a calculation of the HCO positions *at*

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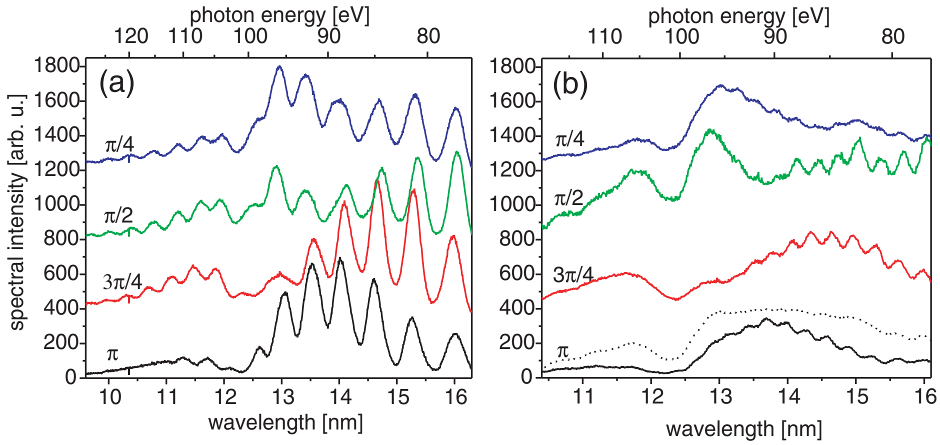

Fig. 2. (a) High-Harmonic spectra measured for different CEP values (solid lines). Filtering out the harmonic modulation at $2\hbar\omega_L \sim 3$ eV reveals several HCO maxima that shift to higher photon energy with increasing CEP (b). The remaining modulation of $1\hbar\omega_L$ is indicative of three attosecond pulses in this spectral range. A global minimum around 100 eV is an artifact caused by partial absorption in our Si_3N_4 spectrometer grating, the dotted line shows the harmonic spectrum averaged over 4 different CEP values after removing both $2\hbar\omega_L$ and $1\hbar\omega_L$ modulations.

the leading edge for a laser pulse of 7 fs as shown in Fig. 3(c), we obtain very good agreement and can extract a value for the absolute CEP to within an integer multiple of π. The closure of the ionization gate prevents the generation of harmonic photon energies higher than 120 eV even though the laser pulse intensity at the peak would allow for the production of > 145 eV harmonic photons. From the expected HCO energy at the peak of the laser pulse we extract a peak laser pulse intensity of $6.2{\cdot}10^{14}$W/cm^2 in the generation volume, which is lower than the focal intensity at the beam waist estimated using gaussian beam propagation of $4{\cdot}10^{15}$W/cm^2. This discrepancy could occur if the harmonic generation is not directly in the focus or if defocusing occurs due to ionization prior to the focus. We also used an 8 fs pulse in the model and compared it to the data, resulting in another good fit (Fig. 3(d)). Note, however, that the extracted CEP varies by almost 0.5π (the data points are shifted along the CEP axis) if a 7 fs pulse is assumed compared to the 8 fs pulse, which means that a sine pulse could be mistaken for a cosine pulse if the pulse duration is not precisely known. The strong dependence of the extracted CEP on the pulse duration arises because the absolute value of the CEP is conventionally defined as the local phase of the electric field at the maximum of the envelope. In the case presented here, high-harmonics are only produced on the leading edge of the laser pulse, and the HCO analysis cannot yield information about the phase of the electric field at the intensity maximum of the laser pulse, which occurs later in time. Only if the pulse shape is precisely known can we infer the CEP at the peak of the pulse. However, as will become clear below, the CEP commonly defined with respect to the peak of the laser pulse is irrelevant for isolated attosecond pulse generation on the leading edge.

If we progressively reconnect ('unwrap') HCO positions at CEP values $\varphi_{\mathrm{CEP}} = \pi$ with those at positions $\varphi_{\mathrm{CEP}} = 0$ due to their periodicity of π, we can follow one particular half-cycle-cutoff in time as it moves up to higher intensities on the leading edge of the driver pulse (Fig. 4(a)). The result of this procedure exhibits the leading-edge intensity of the laser pulse used for the fit in Fig. 3(d) reconstructed with attosecond precision (300 as corresponds to $\pi/4$ CEP) and the time of closure of the ionization gate (corresponding to the highest-energy

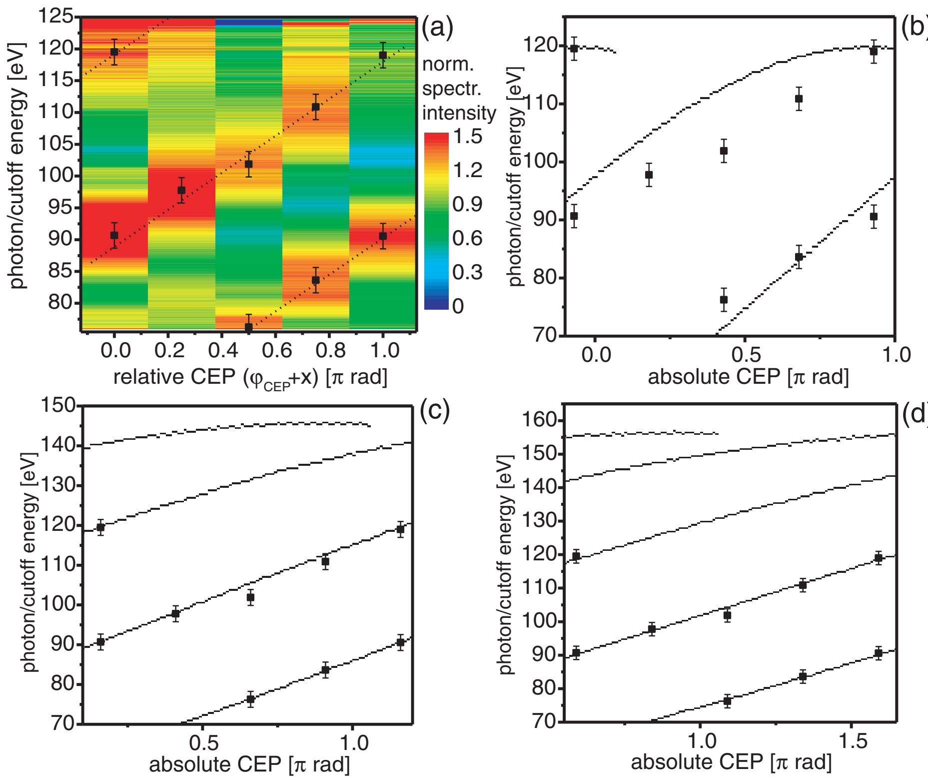

Fig. 3. Harmonic spectra from Fig. 2(b) after removal of harmonic modulations (Fourier filtering) and dividing by the CEP-average spectrum. Above 120 eV only experimental noise is observed in the spectrum. The positions of the half-cycle cutoffs are indicated by black squares (a) while the approximately constant slope lines indicate the linear increase of the HCO photon energy as a function of CEP. A fit to an HCO trace at a pulse duration of ∼4 fs shows poor agreement due to the predicted saturation of HCO energies at the peak of the laser pulse (b) while a fit to the leading edge of a 7 fs (c) or 8 fs (d) pulse results in very good agreement to the measured data.

half-cycle cutoff that was experimentally observed).

As can be seen in Fig. 3(a), the CEP can be used to tune the spectral maximum of harmonic emission. Figure 4(b) shows a harmonic spectrum obtained in 240 mbar of Ne with a more compressed driver pulse (∼6 fs FWHM) to achieve a faster-rising slope on its leading edge, which spectrally separates the harmonic contributions of adjacent half cycles. The photon energy maximum of this half-cycle can be significantly changed (∼10 eV) by varying the CEP. As only one maximum is obtained and only weak spectral modulation is present, we attribute the broad (15 eV FWHM) quasi-continuous harmonic emission to one half-cycle of the driver pulse. This implies a nearly isolated attosecond pulse (bandwidth limit 120 as) is produced by that particular half cycle. The slight residual modulation of the spectrum most likely stems from weaker attosecond pulses generated in the subsequent more intense half-cycle and/or the high-energy spectral tail of a weaker attosecond pulse generated in the previous (less intense) half cycle. For the case of polarization gating [14], harmonic continua of similar bandwidth have been observed. However, the additional complications of controlling the polarization state of an even shorter (5 fs) laser pulse are circumvented in the herein proposed scheme.

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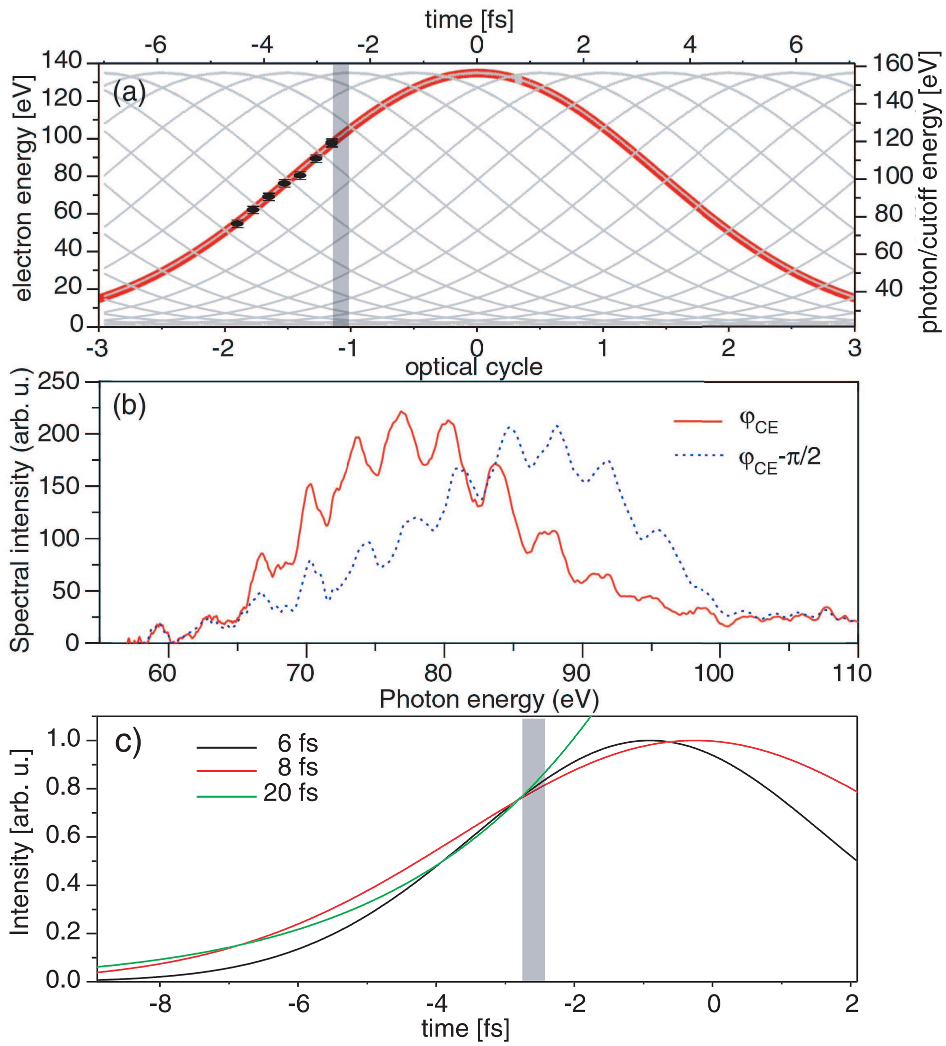

Fig. 4. (a) Reconstruction of the leading edge intensity envelope of a laser pulse by unfolding the HCO data shown in Fig. 3. A Gaussian laser pulse of 8 fs duration (solid red line) fits to both the HCO positions (thin grey lines, calculated as in Fig. 1(c) and unwrapped (see text)) and the measured data on the leading edge (black dots). The grey shaded area represents the time of closure of the ionization gate. (b) By improving the compression of the laser pulse to 6 fs the slope steepness is increased, leading to the experimental observation of a broad quasi-continuum that can be tuned in energy by varying the CEP. Most of this continuum stems from a single half-cycle cutoff while the slight modulation is caused by small contributions of the neighboring half-cycles. (c) A multi-cycle, 20 fs pulse (green line) produces the same steepness (slope) of the rising edge as a 6 fs pulse (black line) after increasing its peak electric field by a factor of 10. The red line shows an 8 fs pulse.

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An important advantage of isolated attosecond pulse generation on the leading edge of the driver pulse is that the available soft-x-ray bandwidth can be much larger than at the peak of the driver. This is a consequence of the larger difference of intensities of successive half-cycles at the leading edge compared to the slower variation of intensity at the peak of the laser pulse. Without an ionization gate, one must work at the peak of the (cosine) pulse for isolated attosecond pulse production, since both earlier and later half-cycles need to produce less photon energy to enable selection of the spectral contribution produced only from the most intense cycle. With an ionization gate, however, the later half-cycles are turned off and only the earlier half-cycles of the pulse need to be less intense. The CEP in this case becomes a free parameter to tune the photon energy of the isolated attosecond pulse.

The existence of an ionization gate makes it clear that the pulse duration of the driver pulse is no longer the important parameter for spectral isolation of harmonics from one half-cycle since harmonic emission after part of the leading edge is turned off and the further evolution of the pulse intensity is irrelevant. The only requirement to obtain a high-bandwidth isolated attosecond pulse is for the leading edge of the driver to be sufficiently steep. Note that for a clean Gaussian laser pulse of any duration, the rising edge can be made as steep as desired only by increasing its intensity e.g. by focusing more tightly; the gate will then move towards earlier times, but the relative change of intensity per unit time just before the gate closure can be as large as for a few cycle pulse (Fig. 4(c)), allowing the same spectral separation of individual half-cycle cutoffs and therefore the production of an isolated attosecond pulse after high-pass filtering. However, as is also well known in the literature on harmonic generation, the efficiency of harmonic production decreases for longer drivers since all the driver's pulse energy after the ionization gate closure is 'wasted'.

Finally, we discuss the possibility of determining the multiplicity of attosecond pulses in experimentally produced attosecond pulse trains. It can be seen that the $1\hbar\omega_L$ modulation in Fig. 2 is only present at low photon energies, while it completely vanishes above 100 eV. The $2\hbar\omega_L$ modulation in the raw data persists up to much higher photon energies (up to 120 eV for some values of the CEP). A $2\hbar\omega_L$ modulation is caused by the interference of harmonics produced in neighboring half-cycles, whereas the spacing of $1\hbar\omega_L$ is caused by interference of the harmonic emission of one half-cycle with that produced a full cycle T later. Since lower photon energies can be produced in a larger number of laser cycles on the leading edge than high photon energies, the $1\hbar\omega_L$ modulation ceases at lower photon energies than the $2\hbar\omega_L$ modulation. The visibility of the $1\hbar\omega_L$ modulation thus indicates the presence of at least three attosecond pulses in the attosecond pulse train, while only two are present at photon energies where only the $2\hbar\omega_L$ modulation is observed. This analysis therefore complements the previously established knowledge [11] that the appearance of a continuum in the harmonic spectrum (which also occurs for a CEP of π in Fig. 2(a)) indicates the generation of an isolated attosecond pulse.

In conclusion, we experimentally observed the role of ionization-gating for high-harmonic generation on the leading edge of the laser pulse. The ionization gate can be beneficial for the generation of broadband, isolated attosecond pulses with a center photon energy that can be tuned by varying the carrier-envelope phase. It also provides a route to isolated attosecond pulse generation with multi-cycle driver pulses. Using the half-cycle cutoff analysis, we observed the signature of individual attosecond soft-x-ray bursts. A characteristic interference between successive half-cycles can be experimentally used to provide information on the multiplicity of attosecond pulses.

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