

Extraction of the *in situ* temporal information of few-cycle laser pulse from carrier-envelope phase-dependent high order harmonic spectrum

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We propose an *in situ* method to obtain information on a few-cycle pulse's temporal structure directly from the high order harmonic spectrum. When the carrier-envelope phase (CEP) changes, a half-cycle of a pulse scans over the pulse itself and directly maps the electric field to the trace of the half-cycle cut-off energy. This map can lead to a quick and simple way to extract the real temporal information, including the envelope and the chirp, of the pulse in the process of high-order harmonic generation. This method is verified theoretically by numerical simulations and experimentally by generating high-order harmonics using CEP-controlled few-cycle laser pulse. © 2014 Optical Society of America

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1. INTRODUCTION

In recent years, remarkable progress has been made on ultrashort laser pulses [1,2] with locked carrier-envelope phase (CEP) [3–5]. Because of the characteristics of high peak power, short temporal width, and controlled electric field, it has become an ideal tool to study ultrafast phenomena, such as high-order harmonic generation (HHG) [6,7], attosecond pulse generation, [8–10], and electron transfer [11,12]. With only a few optical cycles [1,2,13], the temporal structure of the electric field for an ultrashort laser pulse is much different from the sinusoidal curve. For example, its amplitude along the envelope varies dramatically. Many phenomena, such as molecular isomerization [14] and isolated attosecond pulse (IAP) generation based on amplitude gating [10], strictly depend on the temporal structure of the electric field. In IAP generation, when the envelope is steep enough and the CEP is close to zero, there is one half-cycle whose intensity is much stronger than all other half-cycles, so the harmonics from this half-cycle has the highest energy and can form an IAP.

Characterization of ultrashort laser pulses is a key to utilize them and to analyze the experimental results. A few-cycle pulse can be expressed as $E(t) = f(t) \cos(\omega t + \text{CEP} + g(t))$. The temporal structure of electric field $E(t)$ depends on its envelope $f(t)$, central frequency ω , CEP, and temporal dispersion $g(t)$. The temporal property of the ultrafast laser pulse can be measured by several methods such as autocorrelation [15], frequency-resolved optical gating (FROG) [16], and spectral phase interferometry for direct electric-field reconstruction (SPIDER) [17]. However, in many cases, nonlinear effects in an experiment, such as self-phase modulation and self-steepening [18], could severely change the pulse's

property, so the parameters of laser pulses from measurement may differ from that in the experiment. For example, in the process of HHG, nonlinear effects could dramatically modify the pulse's shape [19] temporally from being a Gaussian shape to having a flat shoulder at the falling edge. It is important to acquire the exact parameters of the laser pulse in the experiment even though the pulse shape modification is not always harmful. A temporally and spatially flattened pulse is beneficial for phase matching and can increase the yields of high order harmonics. Knowing the pulse's shape in the region of interaction can help to improve these experiments and to analyze the final results. An attosecond streaking camera developed recently provides a way to completely measure the temporal oscillation of electric fields [20], but this method requires an additional electron spectrograph and generating attosecond pulses, which increase the difficulties of measurement.

2. METHOD

In this paper, we propose an all-optical method to directly acquire the information of the temporal structure of a few-cycle laser pulse based on HHG. The temporal structure of a pulse can be directly obtained through the nonlinear effect itself. Using a CEP-dependent high order harmonic spectrum with distinguishable half-cycle cut-off (HCO), the CEP of the few-cycle driving laser pulse can be extracted [21]. We extend this method to acquire the important information of the envelope $f(t)$ and the temporal dispersion $g(t)$ of a few-cycle pulse at the interaction area from high-order harmonic spectrum. This gives people a simple and quick way to acquire the real temporal property of the driving pulse in the experiment

of HHG and IAP generation, which is very critical for optimizing these important laser sources.

Since the temporal width of a few-cycle pulse is too short to be directly recorded by any electronic device, in general the measurements are carried out in the spatial domain instead of in the temporal domain. In these measurements, a pulse is usually divided into two pulses with a controllable time delay; the two pulses are then focused into nonlinear media to generate a nonlinear signal. When the time delay is changed, the ultrashort pulse is scanned by its replica; then its temporal variation is mapped to the spatial domain through the time-delay-dependent signal, which is used to reconstruct the temporal structure of the pulse. In our method, which carries on this spirit and is based on the extreme nonlinear process of HHG, the electric field of a half-cycle scans the envelope of the pulse itself by simply changing the CEP, and finally important information about the envelope and the chirp can be extracted through the CEP-dependent high-harmonic spectrum.

The typical spectrum of high-order harmonics consists of the plateau region in which harmonic intensity keeps almost constant for all orders and the cut-off region in which the highest order is arrived at and the harmonic intensity drops dramatically. In each half-cycle of the laser pulse, the medium emits high-order harmonics once, in which the cut-off energy is proportional to the intensity of the half-cycle, i.e., the height of the envelope at the position of the half-cycle. With the variation of the CEP, the relative phase between the peak of electric field and that of the envelope, the electric field of a specific half-cycle scans over the pulse envelope and generates HCO photons with different energy. This gives the basic idea of our method, shown in Fig. 1, of extracting the information of the driving pulse's temporal structure from this CEP-dependent trace of HCO photon energy. In this method, the electric field of a certain half-cycle is a probe and scans the pulse itself when the delay changes. It is also a map from

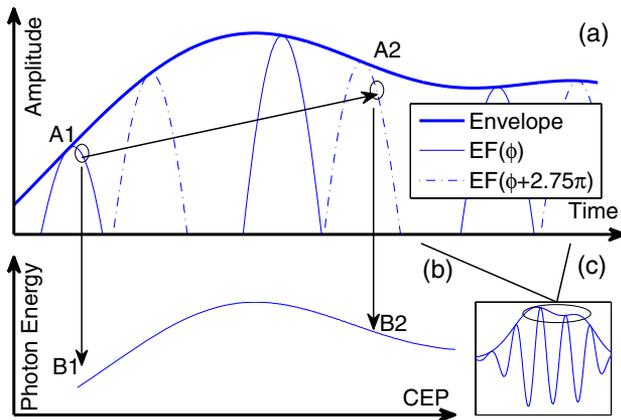


Fig. 1. Carrier envelope phase maps the electric field under the envelope to the cut-off photon energy of high order harmonics. EF stands for electric field. Cut-off energy is calculated from the strong field approximation (SFA) model [22]. Electric field in (a) is the elliptical part of a pulse in (c). (a) The envelope (full thick line), electric field ($\text{CEP} = \phi$) (full thin-line), and electric field ($\text{CEP} = \phi + 2.75\pi$) (dashed-dotted line) of a few-cycle pulse. (b) Cut-off photon energy from a certain half-cycle (HC) at different CEP. When CEP changes from ϕ to $\phi + 2.75\pi$, the H.C. moves from A1 to A2 in (a); accordingly, in (b) the trace of cut-off photon energy shows a similar shape, from B1 to B2.

space to time through the relation $t = \text{CEP}/\omega = (1/v_g - 1/v_p)z$. z is the direction of laser propagation and v_g and v_p are the group velocity and the phase velocity of the pulse, respectively.

The capability of extracting the pulse's temporal structure relies on the light-field-dependent, not the light-intensity-dependent, cut-off energy. The cut-off law says that the maximal photon energy of high order harmonics is proportional linearly to the light intensity. This original law neglects the time-dependent variation of the light field and strictly speaking is only valid for a single color laser. For a pulsed laser field, the cut-off energy of a certain half-cycle is a function of the electric field and varies when the half-cycle is at different position under the envelope. This variation can be used to extract the envelope and the chirp of the laser pulse.

3. SIMULATION

In order to reveal the relations between the CEP-dependent high-order harmonic spectrum and the pulse's temporal structure, we calculated the harmonic spectrum numerically by solving a one-dimensional soft-core time-dependent Schrödinger equation (TDSE) [23] and the HCO photon energy from one half-cycle by using strong field approximation (SFA) [22] model for comparison. In the following, we first set temporal dispersion $g(t) = 0$, only considering the influences of different envelope $f(t)$ to the harmonic spectra, then taking the $g(t)$ into account. The results show that the CEP-dependent high-order harmonic spectra directly give the information about the pulse's envelope $f(t)$ and temporal dispersion $g(t)$. The HCO spectrum can also give the information about the process of HHG, such as the harmonic yields at different temporal position of the pulse.

Figure 2 shows the CEP-dependent high order harmonic spectra from Fourier transform limited pulses with different envelopes. Figure 2(a) shows the high-order harmonic spectra as a function of CEP from a Gaussian shape-driving pulse. The spectra are smoothed to get the position of HCO [21]. The black line shows the cutoff energy from one half-cycle and is consistent with the results of numerical simulation. The sign of the CEP is defined such that when the CEP increases, each half-cycle moves to the direction of positive time. It clearly shows a similar shape between the envelope of the pulse and the trace of cutoff energy. Figure 2(a) also shows asymmetric yields of harmonics between the rising edge and falling edge of the pulse; most of the harmonic signal comes from the rising edge. This is because most of the medium is quickly ionized in the rising edge, so it remains less in ground state at the falling edge of the pulse, which leads to the small yields.

Figure 2(b) shows the spectrum from a temporal asymmetric laser pulse. This temporal shape is similar to that in Gaarde's simulation [19] on the first experiment of IAP generation [10]. The simulation shows that the temporal intensity profile of the driven laser pulse changes dramatically in the process of HHG. The on-axis part of the pulse temporally is from the Gaussian shape to that with a peak in the rising edge and a depressed portion in falling edge. In our simulation in Fig. 2(b), a similar pulse with a Gaussian-like rising edge followed by a flat region at the falling edge is used; the generating trace of HCO energy is similar to the envelope of the pulse. Harmonics above 140 eV, generated from the falling edge, are much more intense than those from the rising edge

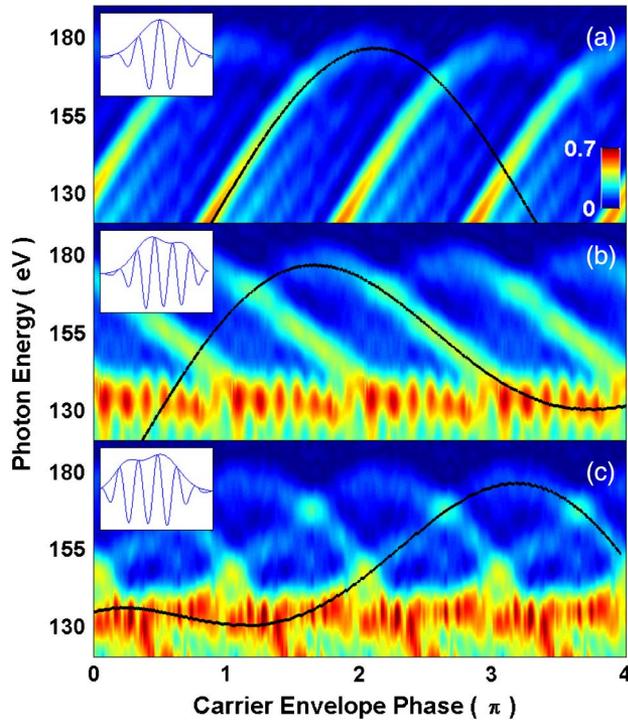


Fig. 2. CEP-dependent high-order harmonic spectra driven by few-cycle pulses with different envelopes. The insert in each spectrum is the corresponding electric field of the driving pulse, and the coordinates are the same as that in Fig. 1(a). All pulses are Fourier transform limited. A color map is calculated from TDSE. The black line is the trace of cut-off energy from one half-cycle and is calculated from SFA. (a) Gaussian shape with 4.5 fs temporal width. Electric fields in (b) and (c) are temporally opposite. All color bars are the same in (a).

because the rising edge is steeper than that in Fig. 2(a) and have fewer half-cycles. It can also be seen that the intensity of harmonics around 130 eV is stronger than other energy ranges and shows some interference pattern. This is because under the relatively long flat region at the falling edge, several half-cycles simultaneously contribute to the same energy range.

In Fig. 2(c) the temporal shape of the driven pulse is opposite temporally to that in Fig. 2(b). The CEP dependent harmonics spectrum also shows the opposite envelope shape. However the distribution of harmonics' intensity in Fig. 2(c) is much different from that in Fig. 2(b). Harmonics above 140 eV are contributed equally from both the rising edge and falling edge of the driving laser. The spectrum around 130 eV is stronger than that in Fig. 2(b). This is because the flat region locates at the rising edge; thus a more natural medium is available for the generation. All the three cases show that scanning the high-order harmonic spectrum from one half-cycle at different CEP, we can obtain the information about the envelope $f(t)$ of the driven laser and the temporal yields of harmonics through the pulse.

The influence of temporal dispersion, i.e., the chirp, is also investigated in simulation. Figure 3 shows CEP-dependent high-order harmonic spectrum driven by a Gaussian shape pulse with different chirp. A positive chirped pulse gives a spectrum in (a). Because fewer cycles of electric field are in the rising edge than in the falling edge, more harmonics are generated from the falling edge. With the comparison

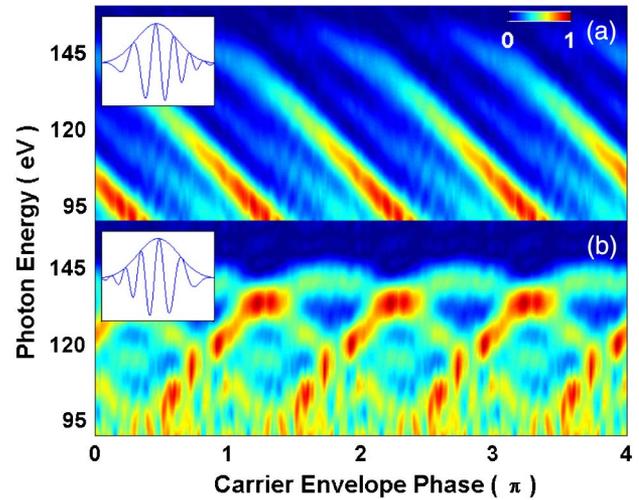


Fig. 3. CEP-dependent high-order harmonic spectra driven by a Gaussian shape pulse with positive chirp (a) and negative chirp (b), respectively. The coordinates of the pulse are the same as that in Fig. 1(a). All color bars are the same in (a).

of the spectrum generated by the zero chirped pulse as shown in Fig. 2(a), the positive chirp leads to slight modulation around 95 eV in Fig. 3(a). Figure 3(b) gives the spectrum from a negative chirped pulse, the rising edge of which generates more harmonics than that of the positive pulse and from which the whole spectral structure is more complex than that from the positive chirped pulse. The negative chirp leads to strong modulation below 120 eV, although with the same envelope the two chirped pulses in Fig. 3 give very different high-order harmonic spectra compared with the spectrum generated from the Fourier transform-limited pulse in Fig. 2(a). Any change of a pulse-temporal envelope, dispersion, and CEP leads to corresponding change in CEP-dependent high-order harmonic spectrum, so it is possible to characterize the pulse *in situ* directly from the spectrum.

4. EXPERIMENT

To verify the method, a few-cycle CEP stabilized laser pulse with pulse energy of 0.4 mJ was used to generate high-order

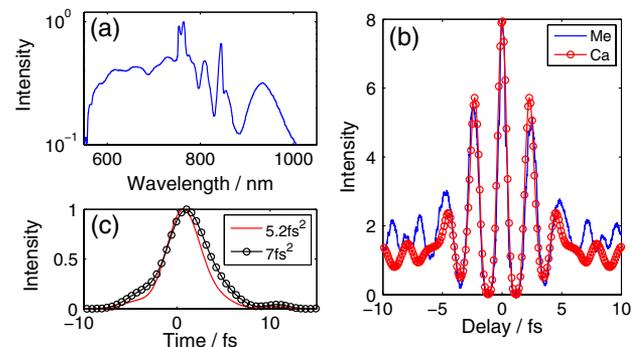


Fig. 4. Laser parameters. (a) Experimental spectrum of the driven laser. (b) Interferometric auto-correlation traces. Blue line is the measured (Me) trace. Red circle line is the fitted trace, which is calculated (Ca) from the experimental spectrum and has the group velocity dispersion (GVD) of 5.2 fs². The two traces show the same duration 4.5 fs. (c) Calculated envelopes. The red line has 5.2 fs² GVD and show the duration of 4.5 fs. The black circle line has 7 fs² GVD and show the duration of 5.2 fs.

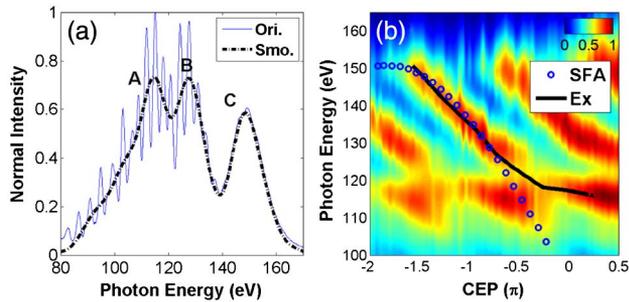


Fig. 5. Experimental trace of half-cycle cutoff in high-order harmonic spectra. (a) Harmonics spectrum when CEP equals -0.7π . The three peaks A, B, and C show the cutoff positions of three half-cycles. The solid blue thin line represents the original experimental data. The dashed line comes from smoothing the experimental data. (b) Harmonics spectrum as a function of CEP. The blue circles are the cutoff photon energy calculated from a 5.2 fs FT-limit Gaussian pulse based on SFA model. The black solid line is the trace of half-cycle cutoff energy in experiment.

harmonics. The spectrum of the laser pulse can be seen in Fig. 4(a). The trace of interferometric auto-correlation can be seen in Fig. 4(b) with the blue line and shows a duration of 4.5 fs. The red circle line is the fitted trace, which is calculated from the measured spectrum by adding 5.2 fs^2 group velocity dispersion (GVD). The continuous CEP locking time is up to 7.2 hours with 85 mrad (root mean square) fluctuation. A pair of wedges is used in front of the generation chamber to change the CEP of driving laser by adjusting the insertion. Neon gas is chosen as the medium for HHG.

The experimental spectrum of high order harmonics is shown in Fig. 5. The half-cycle cut-off can be roughly calculated by smoothing the mixed spectrum shown in Fig. 5(a), in which three peaks—A, B, and C—of the smoothed spectrum (black dashed line) are the cut-off positions of three half-cycles. With the change of the CEP, the three peaks move and form the CEP-dependent spectra shown in Fig. 5(b). The experimental trace is consistent with calculation (blue circle) only above 130 eV and has a flat structure between 110 and 120 eV. It indicates that the pulse in experiment has a similar shape in Fig. 2(b), a peak followed by a shoulder.

The temporal information of the pulse obtained from the HCO spectra contradict the results of the pre-experiment measurement. According to the trace of auto-correlation, the calculated envelope of the pulse before the HHG experiment shows the duration of 4.5 fs, as in Fig. 4(c) with the red line. However, the HCO spectrum shows that the pulse is similar to a 5.2 fs Gaussian pulse in rising edge and has a flat shoulder in falling edge. In our experiment, the driving laser pulses go into the vacuum chamber for HHG through a 1 mm fused silica window, which is considered in the pre-experiment measurement by putting a fused silica glass with the same thickness in the light-path for auto-correlation measurement. The difference between the results from the two methods could be introduced only by the HHG gas cell. Supposing a linear dispersion, by adding 7 fs^2 GVD, one could get a 5.2 fs pulse, whose envelope is shown in Fig. 4(c) with a black circle line. The two envelopes in Fig. 4(c) deduced from linear dispersion clear differ with the results obtained from the HCO spectrum, which shows a shoulder-like structure. We can safely conclude that the nonlinear effect in HHG distorted the temporal structure of the pulse.

The difference between the results from the pre-experiment measurement and the CEP-dependent HCO method is clear. It shows that a laser parameter from a pre-experiment measurement is not always valid to analyze experimental results in an extreme nonlinear experiment like HHG. Obviously it is very important to do *in situ* measurement even though it is qualitative. It is useful for people to analyze the experimental results and to optimize the experimental parameters. With the development of our *in situ* method, it might be probable to obtain quantitative information in the future.

5. CONCLUSION

In conclusion, we propose an *in situ* method to characterize the temporal structure of a few-cycle pulse using a CEP-dependent high order harmonic spectrum. When the CEP of the driving laser pulse is changed, the electric field of a half-cycle scans over the few-cycle pulse and map the light field to the half-cycle cut-off energy. This gives a direct way to know the envelope and the chirp of the driving pulse in the experiments of high-order harmonic generation and attosecond pulses generation. Using this information, one could further optimize the harmonic and attosecond pulse signal. This method also gives important parameters for experimental results analysis.

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