PHYSICAL REVIEW A **80**, 061402(R) (2009)

Complete real-time temporal waveform characterization of single-shot few-cycle laser pulses

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(Received 20 August 2009; published 7 December 2009)

A method for complete characterization of the waveform of individual few-cycle laser pulses is presented. By analyzing the "left" and "right" asymmetries of high-energy photoelectrons along the polarization axis using the recently developed quantitative rescattering theory, we show that the carrier-envelope phase (CEP), pulse duration, and peak intensity of each single-shot pulse can be readily retrieved. By CEP tagging each laser shot, the method permits the study of waveform-dependent processes be extended to relativistic beams and to wavelengths, where CEP stabilization is not yet possible.

DOI: 10.1103/PhysRevA.80.061402 PACS number(s): 32.80.Fb, 32.80.Rm, 42.30.Rx

Strong-field interactions are directly governed by the electromagnetic field; thus, knowledge of the waveform of intense laser pulses is a prerequisite for the interpretation of experiments. The waveform is characterized by its pulse envelope, which in turn can be determined from the peak intensity and pulse duration, as well as the carrier-envelope phase (CEP) that measures the offset between the peak of the electric field and the peak of the envelope. Specifically, a CEP-fixed waveform can be written as $E(t)=E_0(t)\cos(\omega t+\phi)$, where ω is the frequency of the carrier wave, ϕ is the CEP.

Different diagnostic tools have been developed for the determination of temporal parameters of a laser pulse. For example, pulse duration can be measured using autocorrelation or frequency-resolved optical grating [1], and intensity can be inferred from the pulse duration and the fluence in the interaction volume. These methods perform well with multicycle pulses, but not for few-cycle pulses. Lastly, the CEP is most commonly retrieved from the left or right yield of high-energy above-threshold-ionization (HATI) electrons along the polarization axis [2]. However, for this method to work well, the pulse duration and peak intensity have to be known accurately. The temporal structure of few-cycle pulses can also be characterized by attosecond streaking [3], but this technique requires a highly sophisticated installation and hours of integration time using phase-stabilized kHz repetition rate lasers. The stabilization of the CEP is rather complex and it has been demonstrated only up to 0.5 TW peak powers, while low repetition few-cycle pulses with multi-10-TW peak powers are already available [4]. These sub-three-cycle pulses hold promise for the generation of intense isolated attosecond pulses on solid surfaces [5]. To exploit the potential of these pulses to relativistic laser-plasma interactions, a robust single-shot complete characterization method for the temporal structure of the waveform of few-cycle pulses is needed.

Recently, Wittmann *et al.* [6] demonstrated that the left/right photoelectrons from individual single shots can be measured. In this Rapid Communication, we show that such data can be analyzed readily to retrieve the pulse duration, peak intensity, and the CEP using the recently developed quantitative rescattering (QRS) theory [7–9]. The method is robust and the laser pulses can be fully characterized in real time. By CEP tagging each single laser shot, this opens up the opportunity of studying waveform-dependent processes with non-phase-stabilized pulses at any laser intensities and wavelengths.

According to the rescattering model, HATI electrons are due to backscattering of returning electrons by the target ion. The QRS, as detailed by Chen *et al.* [7], puts this model in quantitative form. It shows that experimental HATI electron momentum distributions $D(p,\theta)$ can be expressed as

$$D(p,\theta) = W(p_r)\sigma(p_r,\theta_r), \tag{1}$$

where $W(p_r)$ is the returning electron wave packet and $\sigma(p_r,\theta_r)$ is the elastic differential cross section (DCS) between *free* electrons and the target ion. There are two wave packets: one from the "left" and another from the "right." For few-cycle pulses, the two wave packets differ. In this equation, the DCS is independent of lasers; thus, all the properties about lasers are contained in the wave packet. Since the DCSs for simple atomic target, such as xenon, are well known, one can extract $W(p_r)$ directly from the measured electron momentum spectra. We comment that experimental electron spectra are obtained by integrating over the interaction volume; thus, volume integration effect is included in $W(p_r)$. In the following, we illustrate how the laser parameters are retrieved from the measured electron momentum spectra.

Following Eqs. (50) and (51) of Chen *et al.* [7], the angles θ_r and θ are related by

$$\tan\theta = \frac{\sin\theta_r}{\pm(1/1.26-\cos\theta_r)}, \tag{2}$$

for the two wave packets and p and p_r are related by

$$p^2 = p_r^2(1.63-1.59\cos\theta_r). \tag{3}$$

For HATI electrons along the polarization axis, $\theta_r=\pi$ and $p=1.79p_r$.

Figure 1(a) shows the typical experimental left and right electron spectra from a single-shot measurement. Define $W_R(p_r)=D(p,\theta=0)/\sigma(p_r,\theta_r=\pi)$, where $\sigma(p_r,\theta_r=\pi)$ for atomic targets are easily calculated, for example, the right wave packet can be obtained. Similarly, a left wave packet $W_L(p_r)$ can also be obtained. By comparing these "experimental" wave packets with those obtained theoretically (in-

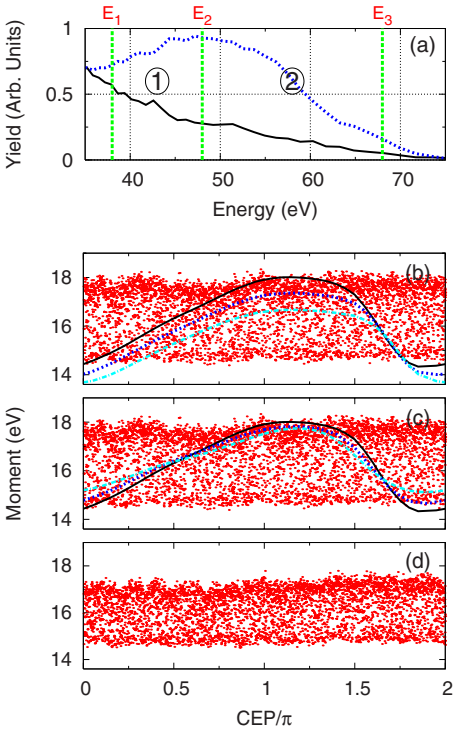

FIG. 1. (Color online) (a) Typical single-shot left-side (solid line) and right-side (dashed line) electron energy spectra along the polarization axis. (b) Energy moment of left-side spectra from experimental measurements (dots) compared with theoretical calculations at peak intensities of 1.2 (dash-dotted line), 1.3 (broken line), and 1.4×10^{14} W/cm^2 (solid line) with pulse duration of 4.5 fs. (c) Same as (b) but for theoretical calculations at peak intensity of 1.4×10^{14} W/cm^2 and pulse durations of 5.0 (dash-dotted line), 4.7 (broken line), and 4.5 fs (solid line), respectively. (d) Energy moment of right-side spectra from experimental measurements. The experimental data are from Wittmann *et al.* [6] (see text).

cluding volume integration), the laser parameters used in the experiment are retrieved. Since the wave packet in the QRS is obtained from the strong-field approximation, the calculation is a few thousands times faster than from solving the time-dependent Schrödinger equation. This speed up makes it possible to carry out real-time retrieval of laser parameters from experimental data.

In Ref. [6], 4500 single-shot data were collected. We first determine the peak intensity and pulse duration used in the experiment. For this purpose, we define a single quantity called energy moment M for the wave packet from each shot,

$$M = \frac{\int_{p_{r1}}^{p_{r3}} (p_r^2/2) W(p_r) dp_r}{\int_{p_{r1}}^{p_{r3}} W(p_r) dp_r}. \quad (4)$$

For example, from the experimental electron spectra, like Fig. 1(a), we estimate E_1 and E_3, where E_1 is close to about $5U_p$ and E_3 is about $10U_p$, where U_p is the ponderomotive energy. These selections are made since the QRS is valid only for HATI electrons. The precise values of E_1 and E_3 are not important. Using the left wave packet for all the 4500 shots, 4500 values of M's are calculated from Eq. (4). These calculated values are displayed in Fig. 1(b), where the horizontal axis is divided into 90 sections. The moments M calculated from the first 50 shots are placed in the first bin, at the vertical positions corresponding to the values of M. The M's from the next 50 shots are placed in the second bin. The process continues until the energy moments from all the shots are registered. Note that the M's are distributed nearly uniformly within a band. The experimental average value of M or $\bar{M}$ was calculated to be 16.46 eV. Using the QRS, the $\bar{M}$ values (averaged over the whole 2π range of the CEP) were found to be 15.64, 16.05, and 16.46 eV, respectively, for peak intensities of 1.2, 1.3, and 1.4×10^{14} W/cm^2. We thus assign the experimental peak intensity to be 1.4×10^{14} W/cm^2. From theoretical calculations, we found that $\bar{M}$ is independent of pulse duration for a given peak intensity. By choosing peak intensity at 1.4×10^{14} W/cm^2 and for pulse duration of 4.5, 4.7, and 5.0 fs, respectively, we found that the best fit to the (vertical) bandwidth is for pulse duration of about 4.6 fs [see Fig. 1(c)]. These two simple procedures allow accurate determination of the peak intensity and pulse duration directly. They are determined independently and are obtained with a much greater accuracy than it was possible with optical methods in Ref. [6].

In the above analysis, only the energy moments from the left wave packets were considered. If the right and left detectors are exactly identical, then the same peak intensity and pulse duration should be obtained from the right wave packets. From Figs. 1(c) and 1(d), it is clear that the two detectors are not exactly the same. If we were to use the data from Fig. 1(d), we would obtain a peak intensity of 1.33×10^{14} W/cm^2 and pulse duration of 4.8 fs. We checked that these conclusions are not changed much when the values of E_1 and E_3 are varied.

Once the peak intensity and pulse duration are known, we retrieve the CEP for each shot following the procedure of Wittmann *et al.* [6]. In their method, between E_1 and E_3, another intermediate energy E_2 was chosen [see Fig. 1(a)]. The total electron yield Y_L between E_1 and E_2 from the left detector is evaluated, and a Y_R from the right detector in the same energy range is calculated. Define the asymmetry $A_1=(Y_L-Y_R)/(Y_L+Y_R)$. A similar asymmetry parameter A_2 is defined for the electron yields between E_2 and E_3. Using $(E_1,E_2,E_3)=(37.9,57.5,64.8)$ eV as in Wittmann *et al.* [6], the (A_1,A_2) for each laser shot is plotted as a point in two dimension, and the results for all the shots are shown in Fig. 2(a). On top of the plot, three theoretical curves are shown, for peak intensity of 1.4×10^{14} W/cm^2 and pulse durations of 4.5, 4.7, and 5.0 fs, respectively. From the three curves, a duration of 4.7 fs gives the best overall fit to the experimental data. This number happens to be the average of 4.6 fs and 4.8 derived from Fig. 1. Note that the theory curve is simply a Lissajous parameter plot of A_1 and A_2 versus the implicit variable the CEP. The theory expects a perfect ellipse. Due to the intrinsic errors in experimental electron spectra, the experimental "ellipse" acquires a width, and the ellipse is distorted due to the difference in the left and right detectors. Comparing Fig. 2(a) to Fig. 3 of Wittmann *et al.* [6], we note

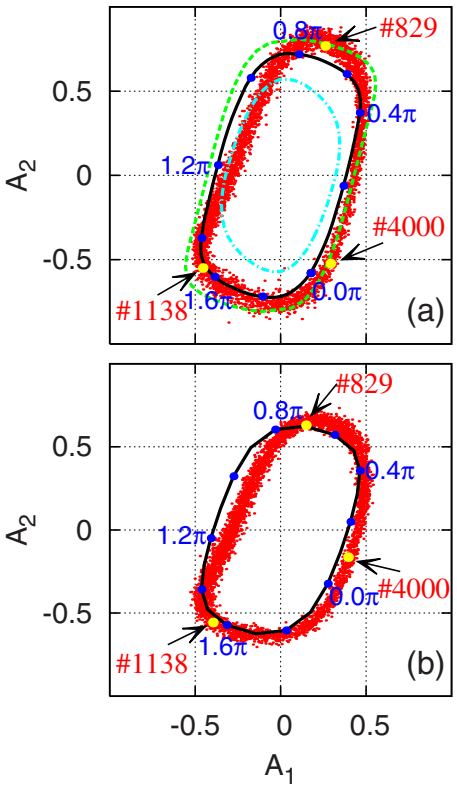

FIG. 2. (Color online) Comparison of the asymmetry ellipse from experiment with theory. The experimental points are calculated from the data of Wittmann *et al.* [6]. The peak intensity used in the theoretical simulations is 1.4×10^{14} W/cm^2. (a) The energy range used is $(E_1,E_2,E_3)=(37.9,57.5,64.8)$ eV, and pulse durations used in theoretical simulations are 4.5 (broken line), 4.7 (solid line), and 5.0 fs (dash-doted line), respectively. (b) The energy range is $(E_1,E_2,E_3)=(38.6,48.7,82.0)$ eV and the pulse duration for theory is 4.7 fs (solid line).

that the absolute values of the CEP assigned in Wittmann *et al.* are off by about 23°. This is because the theoretical model used in the latter is not adequate for accurate absolute CEP determination. We also comment that in Wittmann *et al.*, no method was offered to determine the peak intensity and the pulse duration.

The shape of the ellipse depends on the choice of (E_1,E_2,E_3) used in the calculation of A_1 and A_2. In Fig. 2(b), we show another choice of these parameters. The size and the orientation of the ellipse are changed. However, the actual retrieved CEPs are insensitive to such choices. Take laser shots #829, #1138, and #4000 as examples. We retrieve the CEP by drawing a straight line from $(A_1,A_2)=(0,0)$ to the experimental point (marked by large yellow dots). From the intercept of this line with the theoretical curve, we read out the CEP. For these three shots we found, their CEP values are 129°, 279°, and 16°, respectively, with an error of about 3°. We have tested with many other sets of (E_1,E_2,E_3) and found that the retrieved CEP fall within about 4° to 5° in general.

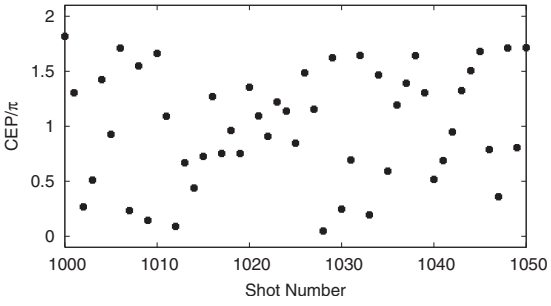

FIG. 3. Absolute CEP extracted for experimental measurements of Wittmann *et al.* [6] from shot number 1000 to 1050.

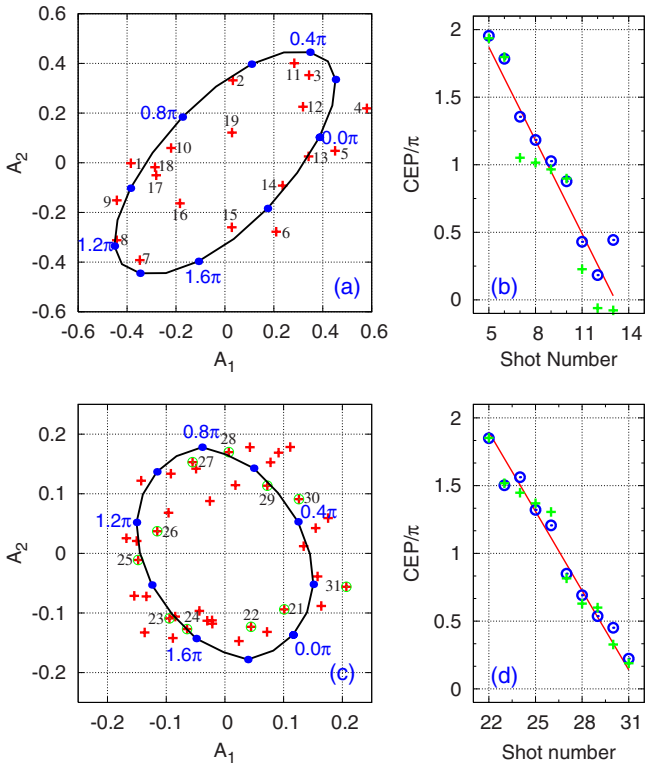

FIG. 4. (Color online) Retrieval of CEP for "sequential" "phase-stabilized" measurements. (a) Comparison of asymmetries from experiment (crosses) with asymmetry ellipse from theory (solid line). The asymmetries are calculated using $(E_1,E_2,E_3)=(10.0,13.0,20.0)$ eV. Experimental data for 19 shots are from Znakovskaya *et al.* [10]. (b) The absolute CEPs for some shots extracted from (a) (circles), compared to those extracted using $(E_1,E_2,E_3)=(10.0,15.0,20.0)$ eV (crosses). [(c) and (d)] Similar comparison for phase-stabilized long pulse measurements from Kling *et al.* [11]. The data consist of 40 sequential shots with wavelength of 760 nm. In (c), shot numbers from 21 to 31 are marked. The peak intensity used in the theory simulation is 1.05×10^{14} W/cm^2 and the pulse duration is 7.0 fs. The energies used for the asymmetry calculations are $(E_1,E_2,E_3)=(40.0,50.0,60.0)$ eV and the retrieved phases are shown as circles in (d). For the crosses in (d), the energies used are (40.0, 45.0, 55.0) eV. The straight lines in (b) and (d) are best fits to the deduced CEPs.

To illustrate that the CEP of each laser shot indeed varies randomly, we show the retrieved CEP for shot numbers from 1000 to 1050 in Fig. 3. Clearly, the CEP varies randomly from shot to shot.

The present method can also be used to determine the CEP of "phase-stabilized" laser pulses. We have obtained the

laser parameters of the few-cycle pulses used in Znakovskaya *et al.* [10], where by changing the CEP the authors were able to control the attosecond dissociation dynamics of CO molecules. From their HATI data, we deduced that their pulses have the peak intensity of 3.0×10^{13} W/cm^2 and pulse duration of 5.0 fs. The mean wavelength of the laser used was 738 nm. In Fig. 4(a), we show the parametric plots for the 19 measured points. The "size" of the ellipse is about the same as in Fig. 2 since the pulse duration of 5.0 fs is close to 4.7 fs in Fig. 2. However, the scattering of the experimental data points from the theoretical ellipse is much larger for these phase-stabilized pulses. The large scattering reflects the remaining shot-to-shot phase variation of up to about 20° (see Wittmann *et al.*). Using the present method to retrieve the CEP, we found that the retrieved CEP depends more sensitively on the values of (E_1,E_2,E_3) used in the analysis [see Fig. 4(b)]. The straight line in Fig. 4(b) was drawn so that the line best fits the deduced CEPs from successive measurements. On the whole, the phase decreases as the "shot number" increases; thus, the phase has been stabilized at least partially.

The determination of CEP is easier for short pulses. Previously, Kling *et al.* [11] reported HATI electron momentum spectra for longer pulses. Their data were analyzed by Micheau *et al.* [12] earlier, also using the QRS. That method is more limited since it assumed that the CEP differences between successive measurements are constant. For this set of data, Micheau *et al.* found a pulse length of 6.7 fs and a peak intensity of 1.05×10^{14} W/cm^2. Using the present method, we analyzed the same data set and found that the pulse length is 7.0 fs and the peak intensity is 1.05×10^{14} W/cm^2, close to the values from Micheau *et al.* [12]. For these longer pulses, the asymmetries A_1 and A_2 are much smaller [see Fig. 4(c)]. Due to the remaining jittering of the CEP from shot to shot of such phase-stabilized pulses, the scattering of the experimental asymmetry parameters around the theory ellipse is much larger. For the ten measurements over the 2π range, we notice again that the retrieved CEPs depend more sensitively on the (E_1,E_2,E_3) used [see Fig. 4(d)] similarly to what was observed in Fig. 4(b).

In summary, using the recently developed quantitative rescattering theory, we propose an all-non-optical method that can accurately retrieve in real time the peak laser intensity, pulse duration, and the CEP of each single laser shot from the measured HATI electron spectra. The computational effort is very small and requires no iterations, and unlike Wittmann *et al.* [6], no optical method is needed for measuring the peak intensity and pulse duration separately. The accuracy of the present method in retrieving the pulse envelope also improves the accuracy of the CEP retrieval. Using this method, by simple CEP tagging, the dependence of strong-field effects on the waveform of ultrashort pulses can be carried out to achieve temporal resolution of a few attoseconds. CEP tagging with randomly changing nonphase-stabilized pulses can work as an "ultrafast phase scan" to perform experiments with any few-cycle pulses of arbitrarily high pulse energy and repetition rate. As today's few-cycle high-power optical parametric chirped lasers are not yet phase stabilized and might not be in the future either, CEP tagging is the only method that will allow phase-dependent laser-solid experiments. Our method is universal and it can also be used for the characterization of multishot phase-stabilized pulses. The present all-non-optical method moreover characterizes laser pulses directly in the interaction region and thus avoids errors introduced by the propagation of the laser beam.

We thank Matthias Kling and Marc Vrakking for providing high-energy electron momentum spectra in [10] for analysis, as shown in Figs. 4(a) and 4(b). C.D.L. also acknowledges earlier discussions on the single-shot measurements with Gerhard Paulus. This work was supported in part by Chemical Sciences, Geosciences and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy.

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