A robust all-non-optical method for the characterization of single-shot few-cycle laser pulses

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An all non-optical method for accurately determining the pulse parameters of individual few-cycle laser shots is presented. By analyzing the “left” and “right” asymmetry of high-energy photoelectrons along the polarization axis using the recently developed quantitative rescattering theory, we show that the carrier-envelope phase, the pulse duration and the peak intensity of each single-shot pulse can be readily retrieved. Unlike optical measurements, this method provides the laser intensity in the interaction region directly.

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Observation of the interaction of laser pulses with matter requires the knowledge of the waveform of the laser field. The waveform is characterized by its pulse envelope, including peak intensity and pulse duration, as well as the carrier-envelope phase (CEP) which measures the offset between the peak of the electric field and the peak of the envelope. Specifically, such a CEP-fixed waveform can be written as $E(t) = E_0(t) \cos(\omega t + \phi)$, where $\omega$ is the frequency of the carrier wave, $\phi$ is the CEP.

The stabilization of the CEP is of great importance, for example, in the generation of isolated attosecond XUV pulses [1], and in controlling the dissociation dynamics of molecules [2]. For atomic targets, the CEP affects the pulses [1], and in controlling the dissociation dynamics (especially noted.) Following the QRS [5, 9], the momentum $p_r$ of the returning wave packet (not directly measured) is related to the measured photoelectron momentum $p$ via

$$p \cos \theta = -A_r \mp p_r \cos \theta_r, \quad (2)$$
$$p \sin \theta = p_r \sin \theta_r \quad (3)$$

where $A_r$ is the vector potential of the laser field at the time of recollision. In Eq. (2), the upper sign refers to the right side while the lower sign refers to the left side. (All the equations are written in atomic units unless otherwise noted.) Following the QRS [3, 8], the magnitude of $A_r = p_r/1.26$. The angles $\theta$ and $\theta_r$ are defined with respect to the laser polarization axis, and $\theta_r$ is measured from the direction of the “incident beam”, i.e., of the returning electrons. For HATI electrons along the polarization axis, $\theta = 0$ (right) or $\pi$ (left), $\theta_r = \pi$, and $p = 2.26 p_r/1.26 = 1.79 p_r$.

According to the QRS [3, 4], the wave packet $W(p_r)$ depends mostly on the lasers only, while the DCS is solely the property of the target. Due to this separability the experimental HATI spectra can also be written as the product of a volume-integrated “wave packet” $W(p_r)$ with the DCS $\sigma$. For each pulse, there are two returning wave packets, one from the left, the other from the right, along the polarization axis. Fig. 1(a) shows the “left” and “right” electron spectra from a typical single-shot measurement. Using $W_R(p_r) = D(p, \theta = 0)/\sigma(p_r, \theta_r = \pi)$, 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 $W(p_r)$ obtained theoretically, the laser parameters used in the experiment are retrieved. Since the wave
packet in the QRS is obtained from 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 near real-time retrieval of laser parameters from experimental data. To perform volume integration, we assume that the spatial distribution of each laser shot is Gaussian and all the electrons from the interaction volume are collected. Thus only the peak intensity at the laser focus and its pulse length (defined as the full-width at half-maximum of the intensity), in addition to the CEP, will be retrieved. We also assume that the peak intensity and pulse duration of each individual shot do not change, i.e., only the CEP is changed randomly. The wavelength is assumed to be known.

In Ref. [4], 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 each shot,

$$M = \frac{\int_{p_{r\min}}^{p_{r\max}} \left(\frac{p_r^2}{2}\right) W(p_r) dp_r}{\int_{p_{r\min}}^{p_{r\max}} W(p_r) dp_r}$$  \hspace{1cm} (4)

where the momentum $p_r$ of the returning electron is related to the momentum $p$ along the polarization axis (and energy $E_r = p_r^2/2$) by $p = 1.79 p_r$. For example, from the experimental electron spectra like Fig. 1(a) (or better the spectra summed over all the shots, on the left or on the right) 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), using fixed $E_1$ and $E_3$. 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 till the energy moments from all the shots are registered. Note that the $M$’s are distributed nearly uniformly within a band. The average value of $M$, or $\overline{M}$, was calculated to be 16.46 eV and was found to be independent of the pulse duration. Using the QRS, the $\overline{M}$ values (averaged over the whole $2\pi$ 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 $1.4 \times 10^{14}$ W/cm$^2$. By choosing peak intensity at $1.4 \times 10^{14}$ W/cm$^2$, and for pulse durations of 4.5, 4.7 and 5.0 fs, respectively, we found that the best fit to the (vertical) band width is for pulse duration of about 4.6 fs, see Fig. 1(c). Thus the energy moments calculated over the single shots allow the determination of the peak intensity and pulse duration easily.

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,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 $1.3 \times 10^{14}$ 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. [4]. 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 $E_3$.
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. [4], the $(A_1, A_2)$ for each laser shot is plotted as a point in 2D, 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 $1.4 \times 10^{14}$ 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 fs 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.

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 CEP’s are insensitive to such choices. Take laser shots, #829, #1138 and #4000 as examples. We retrieve the CEP by drawing a straightline 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 $129^\circ$, $279^\circ$ and $16^\circ$, respectively, with an error of about $3^\circ$. No effort was made to optimize the choices of the three energy points.

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 the “phase-stabilized” laser pulses. We have electron momentum spectra from Kling et al. [10]. From their HATI electron momentum spectra, we deduced that the peak intensity is $3.0 \times 10^{13}$ W/cm$^2$ and the pulse duration is 5.0 fs. The mean wavelength of the laser used was 738 nm. In Fig. 4(a), we show the parametric plots using $(E_1, E_2, E_3)=(10.0, 13.0, 20.0)$ eV 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 the 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” laser pulses. This large scattering reflects the lack of good phase stabilization. As shown in Wittmann et al., performing single-shot measurements on these “phase-stabilized” pulses shows shot-to-shot CEP variations up to about $20^\circ$. 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), where two sets of energy values were used to obtain the CEP for each measurement. The straightline in Fig. 4(b) was drawn so that the line best fits the deduced CEP’s from
The determination of CEP is easier for shorter pulses. Previously, Kling et al.\textsuperscript{11} reported HATI electron momentum spectra for longer pulses. Their data were analyzed by Micheau et al.\textsuperscript{12} earlier, also using the QRS, but the method assumed that CEP differences between successive measurements are constant. For this same set of data, Micheau et al. found a pulse length of 6.7 fs and peak intensity of $1.05 \times 10^{14}$ W/cm$^2$. Using the present method we analyzed the 40 “shots” from Kling et al.\textsuperscript{11}, and found that the pulse length is 7.0 fs and the peak intensity is $1.05 \times 10^{14}$ W/cm$^2$, close to the values from Micheau et al.\textsuperscript{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 phase stabilized pulses from shot to shot, the scattering of the experimental asymmetries around the theory ellipse are much larger. For the ten measurements over the $2\pi$ range, we notice again that the retrieved CEP’s depend more sensitively on the $(E_1, E_2, E_3)$ used for the retrieval, 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 to efficiently retrieve accurate peak laser intensity, pulse duration and the CEP of each single laser shot from the measured high-energy ATI electron spectra. The method is very robust. The computational effort is very small and requires no iterations. Unlike Wittmann et al.\textsuperscript{4}, we show that there is no need to retrieve the peak intensity and pulse duration using the optical method. In the future, CEP dependence measurements can be carried out using non-phase-stabilized lasers. By simple CEP tagging, the strong-field effects on the waveform of ultrashort pulses can be accurately carried out to achieve temporal resolution of a few attoseconds. The present all-non-optical method also characterizes the laser pulses in the interaction region directly and avoid errors introduced by the propagation of the laser beam.

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