Direct in-situ single-shot measurements of the absolute carrier-envelope phases of ultrashort pulses

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Received XX Month XXXX; revised XX Month, XXXX; accepted XX Month XXXX; posted XX Month XXXX (Doc. ID XXXXX); published XX Month XXXX

Many important physical processes such as non-linear optics and coherent control are highly sensitive to the absolute carrier-envelope phase (CEP) of driving ultrashort laser pulses. This makes the measurement of CEP immensely important in relevant fields. Even though relative CEPs can be measured with a few existing technologies, the estimate of the absolute CEP is not straightforward and always requires theoretical inputs. Here we demonstrate a novel in-situ technique based on angular streaking that can achieve such a goal without complicated calibration procedures. Single-shot measurements of the absolute CEP have been achieved with an estimated precision of 0.19 radians.

http://dx.doi.org/10.1364/OL.99.099999

Carrier-envelope phase, defined as the phase shift between the carrier wave and the intensity envelope of an ultrashort pulse $E(t) = E_0 e^{\left(-\frac{t}{\tau}\right)^2} \cos(\omega t + \phi)$, is required to fully characterize the electric fields of such pulses. In nonlinear optics and strong field science, because the response of systems to intense laser pulses closely depends on the instantaneous electric field as well as the intensity, it is thus important to obtain CEP information besides the duration, spectra phase, amplitude etc. Determining CEP is even more critical for few-cycle pulses because it dramatically shapes the temporal evolution of the electric field and can produce different results in light-matter interactions. For example, in high harmonic generation, absolute CEPs can determine whether a single isolated attosecond pulse or two pulses will be produced, assuming all other pulse parameters are equal[1].

While there have been no direct measurements of the absolute CEPs, considerable effort have been made in measuring the relative CEP of ultrashort pulses, both in the fields of frequency metrology and high field science. The f-to-2f interferometric method was developed to measure and stabilize the CEP of a frequency comb[2-4] and was later adapted to single-shot measurements on pulses produced from Ti:Sapphire amplifier systems at multiple kHz[5-7]. Paulus et al. developed the powerful stereo ATI-phasemeter method, which exploits the phase-dependent above-threshold-ionization process (ATI)[8]. By measuring the photoemission asymmetry along the polarization direction at different energy ranges, the phase can be retrieved in real-time for pulses with repetition rates up to 100 kHz[9-11].

Even though the stereo-ATI phasemeter is able to estimate the absolute CEP, it employs the assumption that the highest energy photoelectrons are produced at an absolute phase of 0.3π[10]. This assumption was based on classical calculations and the experimental accuracy was estimated to be 0.1π. It should be noted this result was achieved with a phase-stabilized laser and therefore not a single-shot measurement. A few other studies have shown the correlation between CEP and certain experimental observables such as recoil momentum of produced cations[12-14]. However, in order to estimate the absolute CEP, comparisons between experimental and theoretical modeling are always required[15, 16].

All of the aforementioned methods used linearly polarized ultrashort pulses. A proposal was put forward almost 20 years ago to measure the absolute CEP using circularly polarized light[17]. The concept is quite simple: due to the high nonlinearity of strong field ionization, the direction of the peak electric field in the plane of the polarization, which is uniquely associated with the absolute CEP, has the highest ionization rate. If one can measure the angle dependent ionization rates, the absolute CEP can be directly obtained. However, as recently shown[18], the final measured lab-frame angle is subject to uncertainty due to population depletion and Coulomb field deflection (see also Fig. 1a and b). Therefore, it can only be applied to a limited laser intensity range and to electrons within a certain energy range. Furthermore, the implemented measurement cannot be carried out in single-shot fashion. As such, even though the original proposal has inspired considerable research in revealing detailed dynamics of strong field ionization [19-21], an experiment to fulfill its main purpose of determining the absolute CEP of short pulses has yet to appear.

In this work, we show the absolute CEP of each individual pulse at 1 kHz can be measured with an angular steaking technique using elliptically polarized strong fields instead of circularly polarized light. Employing elliptically polarized light is critical: it completely mitigates the complicating factors (Coulomb field deflection and population depletion) and thus allows a direct correlation between the angle of electron ejection and the absolute CEP. We achieved this with single-shot photoelectron imaging using a novel apparatus that can access the full 2D momentum of electrons in the plane of polarization. We further suggest that this method can also be used for characterizing the absolute CEP of linearly polarized few-cycle pulses.

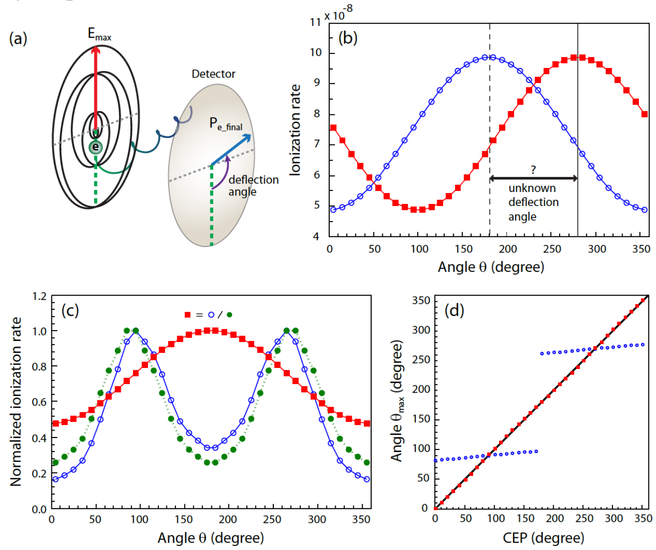

Fig. 1 (a) Schematic of angular steaking of strong field ionized electrons by few-cycle circularly polarized pulses. Electrons tunnel out at the direction opposite to the electric field and gain a final momentum in the laser field. (b) The calculated angle dependent ionization rates of a 5 fs circularly polarized pulse. The calculated rates (blue) show a single peak at 180 degree, corresponding an absolute CEP of π. However, due to an unknow deflection angle, from the supposedly measured yield (red), this CEP cannot be determined. (c) The calculated angle dependent ionization rates of the same pulse but with an ellipticity of 0.92 and a CEP of π (blue circle), the CEP-averaged angle dependent ionization rates with the same elliptically polarized pulse (green dot) and the normalized ratios between the two cases, showing the CEP angle at 180 degrees was retrieved. Note the zero angles are different between (b) and (c) by the deflection angle. (d) The angles with maximum yields obtained from angle dependent ionization rates with (red dot) and without (blue dot) scaling by the CEP-averaged angle dependent ionization rates vs. the CEP. The red dots show an almost perfect retrieval of all CEPs while the blue ones only loosely depend on the CEPs. The modeling of strong field ionization rates followed ref.[22] The simulations did not model electron propagation after ionization.

We first demonstrate the principle of using elliptical polarized light to retrieve the absolute CEP through simulations (Fig. 1). As mentioned earlier, experiments employing circularly polarized light have difficulties in determining the absolute CEP due to the unknown deflection angle between the electric field direction at the moment of ionization and the final lab-frame electron momentum (this will be π/2 without Coulomb field interaction and population depletion). With elliptical polarized light, such a deflection angle can be directly measured by integrating many single-shot electron images to average out the phase dependence while preserving the ellipticity dependence. The angle between the measured minimum (maximum) yield angle and the minor (major) axis of the polarization ellipse is the deflection angle, which is the result of all effecting factors including the vector potential, Coulomb field deflection, population depletion and ionization delay. The latter three are difficult to assess directly and the topic of ionization delay is even controversial. By measuring the angle directly, we can remove such an uncertainty for the purpose of determining the absolute CEP. However, due to the ellipticity, each electron image does not have a single maximum yield angle anymore (Fig. 1c) [20]. Also, the angles with maximum yields only loosely depend on the absolute CEP (Fig. 1d). However, it turns out that if we scale the angle dependent yield of each image with the averaged angle dependent yield, the phase dependent yield can be fully recovered and thus the CEP angle can be extracted as shown in Fig. 1c and d. Furthermore, if we set the angle of the lowest (highest) ionization yield in the averaged image to zero, the peak yield angle of each individual single-shot image will automatically become the absolute phase of the minor (major) axis of the electric field ellipse. In this case, the deflection angle is completely removed from the measurements regardless of its absolute value.

The experimental implementation requires a detection system capable of measuring the 2D momentum of electrons in the plane of the polarization. For single-shot measurements, many electrons (>500) needs to be detected from a single laser shot in order to achieve reasonable statistics. This multi-hit requirement effectively eliminates all 3D momentum detector such as delay-line[23] and camera-based 3D detectors[24, 25]. A conventional 2D imaging detector, which combines microchannel plates (MCPs) and a phosphor screen is the only option due to its massive multi-hit and 2D imaging capabilities. However, in a typical velocity map imaging (VMI) setup, in which the laser beam is propagated parallel to the plane of the detector, only one dimension of the electron momentum in the plane of the polarization can be accessed even though both momenta are required. Therefore, a different detector-laser beam configuration is needed. In this work, we designed and constructed a new VMI setup, in which the laser beam is pointed at the detector and thus enables direct imaging of 2D momenta in the plane of polarization. Fig. 2 schematically describes the experimental setup. Briefly, the ultrashort pulses utilized in this experiment were generated by first broadening the spectrum of 30 fs pulses from a Ti:Sapphire amplifier laser system (KMLabs, Red Dragon, 1 mJ/pulse at 1 kHz), using an argon filled 1-m long hollow-core-fiber (ICON, Imperial College London) and being further compressed with 7 pairs of chirped mirrors (Ultrafast Innovations GmbH, PC70). The compressed pulses were fully characterized using a dispersion scan (d-scan) technique[26]. The measured pulse duration was ~4.3 fs. The CEP of the laser was not stabilized. Using an ultrabroadband quarter-wave plate (United Crystals, AWP650-1100), we obtained elliptically polarized light with an ellipticity of 0.9. This beam was then loosely focused onto a continuous krypton gas jet, using a 35 cm focal-length concave mirror mounted on a translational stage. The focal spot was adjusted to be located after the atomic beam to minimize phase averaging arising from Gouy phase shift[27]. In principle, any gas can be used in this setup because the angular streaking technique is universal. Krypton was used here because it has a relatively low ionization potential and provided a high count-rate for a single laser shot (>600 counts). The laser beam was stopped by a beam block located in front of the MCP detector. The block has minimum effect on electrons because it was situated in the center of the donut-shaped momentum distributions and thus did not block any signal. We note that similar detector-laser beam configurations have been employed previously for measuring photoemission from surface[28] and photoelectrons produced by x-rays[29].

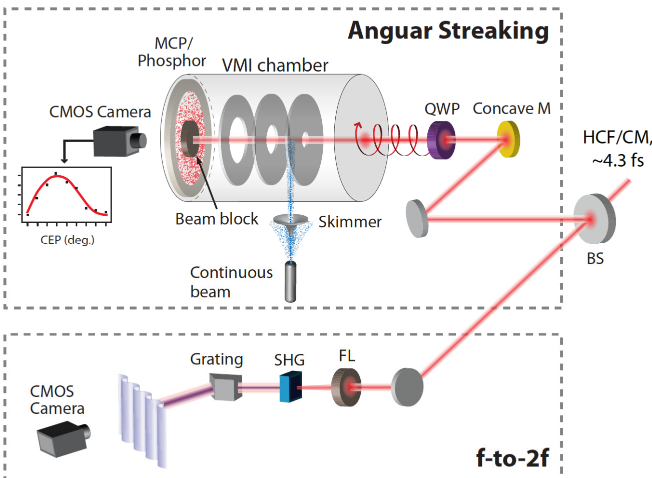

Fig. 2 Schematic of the experimental setups including both the single-shot angular streaking apparatus and a f-to-2f measurement setup. Both cameras were running at 1 kHz, the same as the laser repetition rate. Each camera image of the angular streaking setup was centroided to identify individual electron hits and their positions were recorded. Each camera image of the f-to-2f setup was reduced to 1-d interference pattern and was then filtered and Fourier-transformed. The phase of each pulse was recorded. The abbreviation of each component in the figure is as follows: HCF/CM: hollow core fiber/chirped mirrors; BS: beam splitter; CM: concave silver mirror; FM: flat silver mirror; FL: focus lens; QWP: quarter waveplate; VMI: velocity map imaging; SHG: second harmonic generation crystal.

To validate the phase measurement by the angular streaking technique, we also constructed an f-to-2f interferometric setup employing a fast CMOS camera, which read out the f-to-2f fringes and performed real-time fast Fourier transform at 1 kHz to retrieve the relative CEP of each individual pulse. Even though the f-to-2f method does not provide absolute CEPs, it will be used as a standard for estimating the precision of the angular streaking measurements[30].

The experimental results are shown in Fig. 3. A single-shot electron image and an averaged image of 5000 laser shots are shown in Fig. 3a and b, respectively. Fig. 3c shows the integrated angle dependent yields of single-shot and averaged images. It is clearly seen that main features are due to the ellipticity (two-peak structure). If the single-shot yield is scaled by that of the averaged image, the single peak structure is recovered, and the peak position can be located by peak-finding algorithm. Fig. 3d is the correlation plot between the

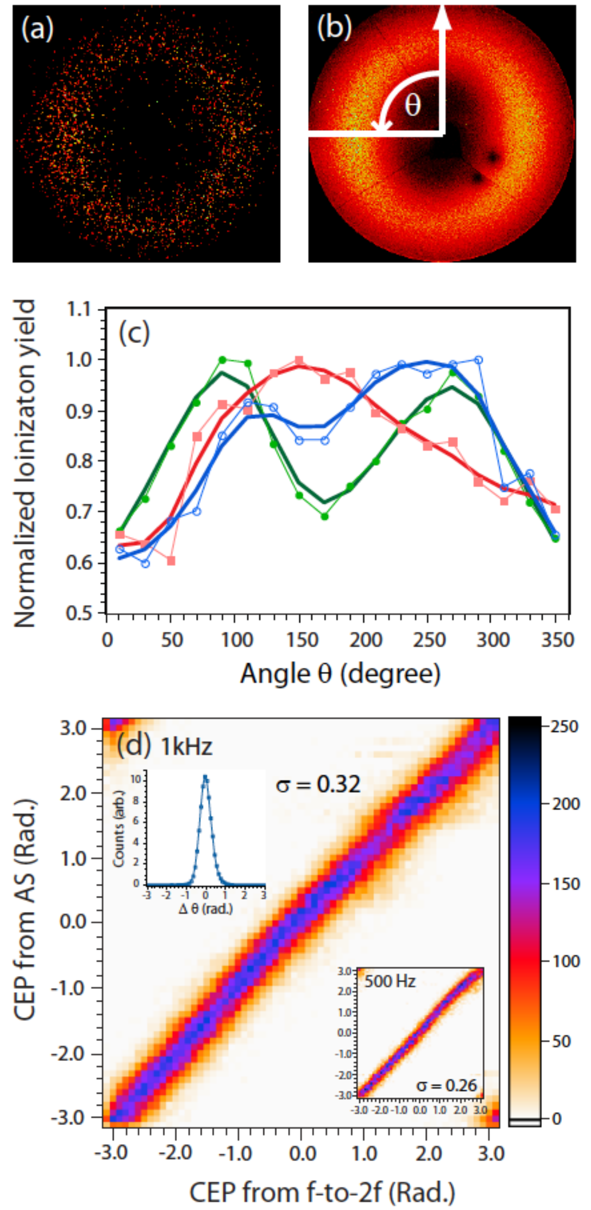

Fig. 3 (a) A single-shot photoelectron image. (b) An accumulated photoelectron image of 5000 laser shots. (c) Similar to Fig. 1c but with experimental data. The thick lines are the results of qubic-spline smoothing of the data. (b) The measured CEP correlation plot between angular streaking and f-to-2f methods at a repetition rate of 1 kHz. The left inset shows the distribution of phase difference while the right inset shows the correlation plot at 500 Hz.

CEPs measured by angular streaking and f-to-2f methods. The positive one slope validates the angular streaking measurements. The f-to-2f result was shifted to match the measured absolute CEPs from angular steaking. The thickness of the line in the correlation plot represents the uncertainty of both methods. For 1×10^5 laser shots, the standard deviation of the measured phase difference was 0.32 radians. When we used the previously obtained standard deviation of a similar f-to-2f setup (0.184 radians)[30], we obtained a standard deviation for the angular streaking method to be about 0.26 radians. The precision was mainly determined by the number of electrons detected in a single image, which averaged to about 600 electrons per laser shot. In order to improve this, we increased the count rate to 1700 electrons/shot while running the repetition rate at 500 Hz. The reduced repetition rate allowed the imaging detector to fully recover from the previous laser shot. The resulted 1 million counts/s is close to the limit of an MCP imaging detector before severe dead-time issue arises. The combined standard deviation was improved to 0.26 radians, which suggested the precision of the angular streaking measurement was better than 0.19 radians. This value is better than the best calibrated CEP measurements using a stereo ATI phasemeter (0.21 radians)[15]. It is possible to increase the count rate further by lowering the repetition rate. However, this will not allow tagging every shot of the laser. It is favorable, on the other hand, to use the current technique to calibrate existing relative CEP measurements such as f-to-2f or a stereo ATI phasemeter. Such a calibration only needs a single shot at a high count-rate and does not require any theory input. Once the calibration is done, the apparatus can be used to study phase-dependent strong field interactions. The new VMI apparatus offers great versatility in making momentum measurements of both ions and electrons. It can also be readily converted to a coincidence 3D momentum imaging apparatus[24, 25, 31]. Finally, even though the method was demonstrated with elliptically polarized light, because the in-situ absolute phases of both axes of the electric field ellipse are known from the measurement, by rotating the quarter waveplate to align either the fast or slow axis with the input polarization of the laser beam, the absolute CEP of the resulted linearly polarized light is also known.

Funding. U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES)(DE-SC0012628).

Acknowledgement. We would like to thank Adam Wyatt at the CIF, Rutherford Appleton Laboratory for assistance with the d-scan analysis.

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