nature physics

ARTICLES

PUBLISHED ONLINE: 19 APRIL 2009 | DOI: 10.1038/NPHYS1250

Single-shot carrier–envelope phase measurement of few-cycle laser pulses

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Full characterization of single ultrashort laser pulses, as needed for attosecond metrology and spectroscopy, requires precise measurement of the offset between the electric field and pulse envelope, or carrier–envelope phase (CEP). Until now, all CEP measurements have been made by averaging over a large number of phase-stabilized laser pulses. Here, we demonstrate the first single-shot CEP measurement of intense few-cycle laser pulses. We focus a laser pulse on a gas target and detect photoelectrons emitted in opposing directions ('left–right') parallel to the polarization of the laser. By comparing the left–right asymmetries of photoelectrons at different energies, we mapped the CEP of consecutive non-phase-stabilized pulses on a parametric plot. This new evaluation method enables us to determine the CEP without phase ambiguity at unprecedented measurement precision. Future investigation of phase-dependent phenomena with CEP tagging at a much lower phase jitter than accessible at present with phase-stabilized lasers is now possible.

A laser pulse can be considered as a sine-like electromagnetic wave modulated by an envelope function $\mathcal{E}_0(t)$. Accordingly, the field of the pulse can be written as $\mathcal{E}(t) = \mathcal{E}_0(t)\cos(\omega t + \phi)$, where ω is the frequency of the carrier wave and ϕ is the carrier–envelope phase[1] (CEP). The significance of the CEP ϕ—or rather the rate of its variation $\mathrm{d}\phi/\mathrm{d}t$—was first recognized and exploited in frequency metrology. $\mathrm{d}\phi/\mathrm{d}t$ is the offset of the frequency modes of a short-pulse laser from integer multiples of the pulse repetition rate and is therefore known as the offset frequency. Stabilization of this offset frequency[2,3] led to a revolution in frequency metrology and its pioneers, T. W. Hänsch and J. L. Hall, were awarded for their contributions with the Nobel Prize in 2005.

For few-cycle laser pulses, the pulse envelope changes appreciably within one optical cycle; therefore, the CEP effectively determines the temporal evolution of the underlying electric field. This is of great significance, as virtually all strong-field laser–matter interactions depend on the electric field rather than the intensity envelope of the pulse. For example, isolated attosecond extreme-ultraviolet pulses[4,5] are generated just after the most pronounced maximum of the electric field of a laser pulse[6–8]. Changes in the CEP shift the position of this maximum within the pulse envelope and subsequently the instant of attosecond pulse generation. Extension of waveform control to multi-terawatt few-cycle lasers[9,10] is expected to open the door to a plethora of new phenomena. One of the most intriguing ones is certainly the generation of isolated attosecond bursts with kiloelectronvolt photon energies[11]. Obviously, the present state-of-the-art in the ability to control the phase severely limits such applications.

Several techniques exploiting various phase-sensitive phenomena were shown to be able to retrieve the CEP in proof-of-principle experiments[12–16]. All of these methods need a large number of laser shots for a single measurement point, thus averaging over possible fluctuations of the CEP. It also implies that the CEP of the laser pulses is already stabilized, which, as a rule, is rather complex. Typically, the rate of change of the CEP is detected with a so-called f-to-2f interferometer[2,3,17], and the CEP drift is then readjusted with a control loop. This, in particular, was the way to stabilize the phase of intense few-cycle pulses[18] for the first time. Alternatively, quantum interference in photocurrents[12,19] or linear optical interferometry[20] can be used to derive the evolution of the CEP. All of these techniques cannot detect the actual value of the CEP but only its relative change. Therefore, measurement of the absolute phase requires a fundamentally different approach, ultimately one that can carry out this task using only a single laser shot.

The most recent step towards single-shot detection by improving the signal yield was made using half-cycle cutoffs in high-harmonic spectra[21]. This method, however, demands more than 0.1 mJ of pulse energy and CEP retrieval was demonstrated only by integrating over 2 s at a kilohertz repetition rate (2000 shots). In the pursuit of a single-shot measurement, a large variety of methods have been proposed[22–25], but no experimental demonstration has been reported yet. Here, we show for the first time single-shot CEP measurement of intense few-cycle laser pulses.

We detected photoelectrons generated by above-threshold ionization (ATI) from single laser shots. In ATI, photoelectrons possibly gain much more energy from the strong laser field (up to tens of electronvolts) than they need for being liberated from an atom. ATI spectra from few-cycle laser pulses are highly phase sensitive, resulting in an asymmetry if two spectra recorded at opposing directions with respect to the laser polarization are compared[14]. We introduced a new method where the asymmetries are calculated from two different energy intervals of the ATI spectra, leading to a two-dimensional parametric plot mapping the CEP without any ambiguity. Facilitated by our single-shot detection, we measured the CEP of consecutive non-phase-stabilized laser pulses

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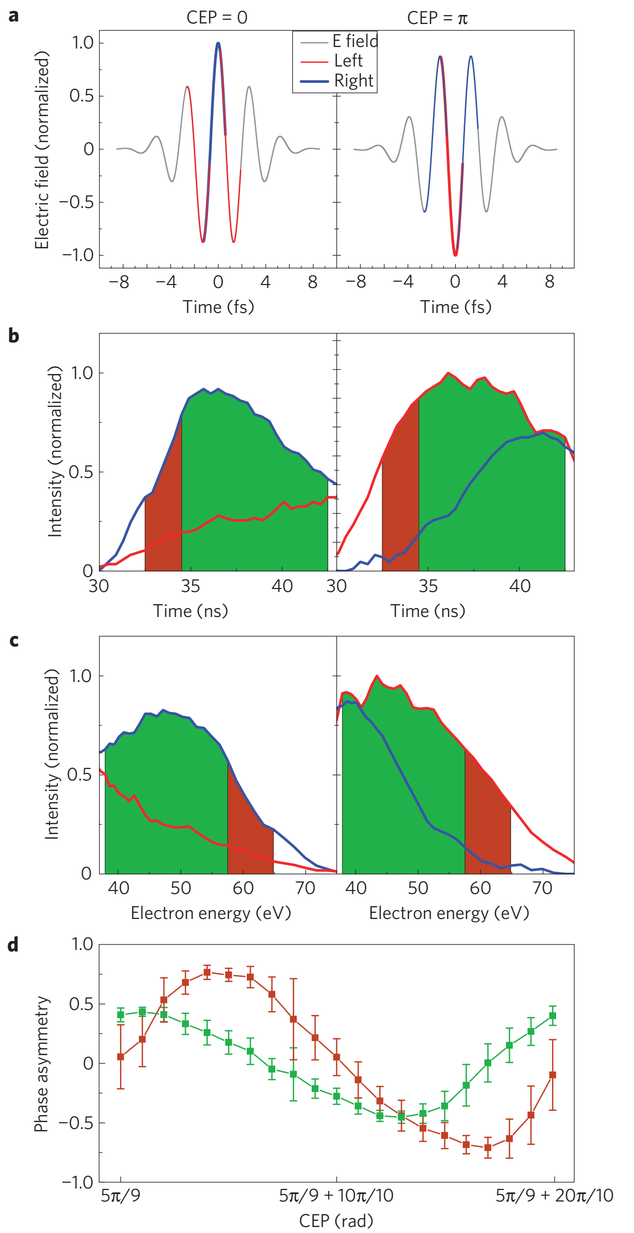

Figure 1 | ATI with few-cycle pulses. a, Travel times of rescattered electrons (red and blue lines) in the ATI process illustrated in the electric field of a 1.5 cycle (4.1 fs) cosine and anti-cosine laser pulse (grey lines). Red and blue indicates whether the electron rescattered after its trip is detected by the left or right detector, respectively, in the stereo-ATI phase meter. The trajectories were calculated classically. **b**,**c**, Single-shot left and right TOF spectra (**b**) and converted energy spectra (**c**) recorded in the experiment. A −25 V blocking potential allows only electrons with a kinetic energy >25 eV to reach the detectors, which is why only the high energy parts of the spectra are shown. The green and brown areas indicate the spectral ranges used to derive the phase asymmetry parameters *x* and *y*. Parameter *x* was calculated as $P_L - P_R/P_L + P_R$, where P_L and P_R are the integrated signals between 42.5 ns and 34.5 ns on the left (red) and right (blue) TOF spectra (corresponding to an energy range from 37.9 eV to 57.5 eV). *y* was calculated in an analogous manner with integration ranges from 34.5 ns to 32.5 ns on the TOF spectra (57.5–64.8 eV). **d**, Dependence of the phase asymmetry parameters *x* and *y* on the CEP (here measured for phase-stabilized pulses). Depending on the selection of the spectral ranges from which the parameters are derived the sinusoidal curves are shifted, in our case by 60°.

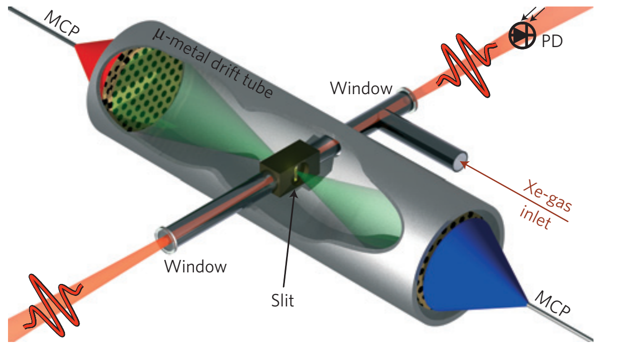

Figure 2 | Single-shot stereo-ATI phase meter. Two opposing TOF spectrometers are mounted in a compact high-vacuum apparatus, carefully shielded (symbolized by the μ-metal-shielding tubes) from electrical and magnetic fields. Xenon atoms at a pressure of 1.6×10^{-2} mbar fill the inner part of a differential pumping stage. The xenon atoms are ionized near the focus of the laser beam and enter the ultrahigh-vacuum drift tubes through the vertical slits. The electrons are detected with a pair of MCP detectors. The colour coding of the detectors corresponds to the colours used in the spectral plots in Fig. 1. Vacuum pumps are not shown. PD: photodiode

without being limited by the fluctuations of the phase stabilization and ended up with unprecedented precision.

Mapping the CEP with rescattered ATI electrons

Our approach for phase measurement relies on ATI in an isotropic medium (gaseous xenon) by a linearly polarized few-cycle pulse. Asymmetry of the field of the laser pulse results in asymmetry of photoelectron emission in opposite directions parallel to the laser polarization. There are two distinctively different mechanisms leading to typical ATI photoelectron spectra. Most photoelectrons leave the atoms directly with low energy (<25 eV under typical conditions of this experiment). A tiny fraction (<1%) revisits the ion core and rescatters at a time t_1 approximately three quarters of an optical cycle after initial ionization at time t_0. Rescattered ATI electrons can acquire substantial kinetic energy (up to 60 eV in our case) and form a characteristic plateau[26] at the high-energy side of the photoelectron spectra. Our phase measurement relies exclusively on the investigation of these plateau electrons because they exhibit a phase dependence stronger by about an order of magnitude as compared with direct photoelectrons[27,28]. The explanation is in fact quite comprehensive: for low-energy (direct) electrons, asymmetric ionization yields at instant t_0 are largely cancelled by the deflection of the photoelectrons in the laser field[29]. High-energy (rescattered) electrons revisit the ion core as already noted. After backscattering at time t_1, they can be accelerated to high energy only if the field strength is large during the optical cycle following the scattering event. In addition, the field strength should also be high at the instant t_0 of initial ionization to provide high ionization probability. As t_0 and t_1 differ by approximately three quarters of an optical cycle, both conditions are hard to meet for a few-cycle pulse. This, together with some other boundary conditions in the kinematics of recolliding electrons, gives rise to the strong CEP dependence of the plateau part of the photoelectron spectra.

The travel times of a few rescattered electrons in the laser field are shown in Fig. 1a for cosine ($\phi = 0$) and anti-cosine ($\phi = \pi$) pulses. Red and blue lines indicate electrons propagating to the left and right detectors respectively. The asymmetry of the photoelectron spectra (Fig. 1b,c) is quantified by the normalized phase asymmetry parameter: $(P_L - P_R)/(P_L + P_R)$, where P_L and P_R are the numbers of electrons integrated over a certain energy interval emitted in the left and right directions. It has been demonstrated experimentally and numerically[14,27,28] that the phase asymmetry parameter thus

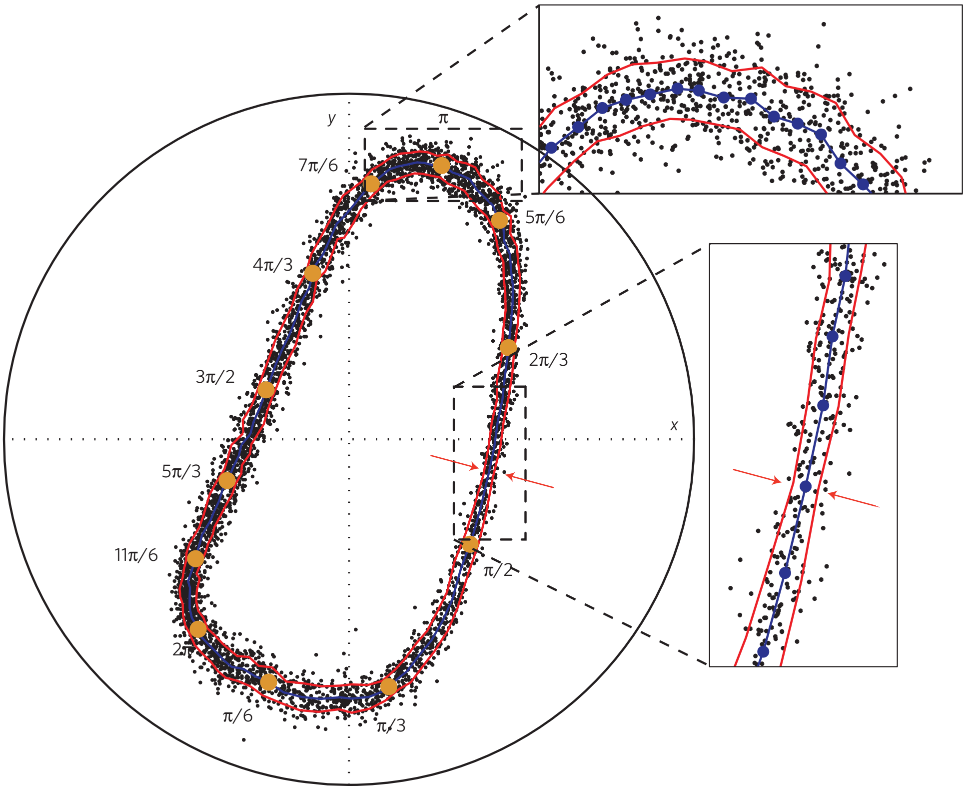

Figure 3 | Mapping the CEP of phase-stabilized and non-phase-stabilized consecutive laser pulses. Consecutive single laser shots (black dots) from a non-phase-stabilized laser at 3 kHz repetition rate depicted on a parametric plot in which the axes x and y are the phase asymmetry parameters derived for different energy ranges in the ATI spectra. The CEP of the 4,500 measured laser shots varies randomly in a 2π range leading to a distribution along a nearly elliptical curve (blue line). Using the random CEP distribution of non-phase-stabilized laser shots, the phase asymmetry was linearized: 90 blue dots (zoomed in view) indicate $\pi/45$ phase intervals similar to the minute ticks on a watch. The values of the CEP indicated around the quasi-ellipse (orange dots) were retrieved by one-dimensional TDSE simulations. The red lines show the standard deviation of the measurement. At the operation point of $\phi = 5/9\pi$, highlighted by red arrows and close to the zero value of phase asymmetry parameter y (highest energy range of the ATI spectra) the measurement precision was evaluated to be $\pi/300$ (s.d.).

obtained depends on the CEP close-to-sinusoidally if P_L and P_R are derived from the entire high-energy spectra. The differential asymmetry, that is, the asymmetry calculated for a given range of electron energies, changes in a nearly sinusoidal way with a shift of the CEP; however, there is a gradual phase shift in this dependence with increasing electron energy. We demonstrate a novel method for measuring and retrieving the CEP, which exploits this observed shift (Fig. 1d) and further relies on the unique features of our single-shot measurement technique. We defined two phase asymmetry parameters with integration ranges of 37.9–57.5 eV for the first, and 57.5–64.8 eV for the second parameter. These ranges are indicated with green and brown areas on the time-of-flight (TOF) and energy spectra for the cosine and anti-cosine waveforms of the laser pulse in Fig. 1b and c, respectively. We used the TOF spectra for CEP retrieval because those are the raw unprocessed signals from our measurement apparatus, which is shown in Fig. 2. The choice of the two ranges was optimized for high phase asymmetry and low measurement noise. The two (close-to-sine) phase asymmetry curves obtained in this way are shifted with respect to each other by approximately 60°. Instead of the conventional linear representation (phase asymmetry versus phase), new insight can be gained by plotting these sine-like phase asymmetries on a Lissajous-like parametric plot with each axis corresponding to one of the two phase asymmetries. In Fig. 3, laser shots with a random CEP are shown in that representation. With the two phase asymmetry parameters, the CEP of each shot (in the entire 2π range) is mapped to one point on the ellipse-like curve, which implies that there is no phase ambiguity. The CEP in the figure was retrieved by carrying out one-dimensional time-dependent Schrödinger equation (TDSE) simulations on rescattered electrons with the parameters used in the experiment. The reason for a slightly deformed ellipse is due to the fact that the phase asymmetry curves are per se not perfectly sinusoidal and that the responses of the two electron detectors are not exactly identical.

Single-shot detection

The CEP measurements were carried out with a single-shot stereo-ATI phase meter (Fig. 2). In this twin TOF detector, ATI electrons from xenon atoms are detected by two microchannel-plate (MCP) detectors in opposing directions parallel to the laser field's polarization. To detect only the electrons that are most phase sensitive at high energies (see above), low-energy direct electrons ($E < 25$ eV) are discriminated by an electrostatic repeller. For single-shot performance, a large number of electrons have to be detected per shot of the laser. This is realized by the use of a differentially pumped vacuum apparatus that is carefully shielded from external electromagnetic fields. The single-shot electron signal is detected with a digital data acquisition system making consecutive detection at high repetition rates (>100 kHz) possible. Our method requires only ~40 μJ of pulse energy and it is non-invasive as the electron density in the target is too low to affect the laser beam. It can even be realized at an intermediate focus in a laser beam, without any impact on the spatial and temporal characteristics of few-cycle pulses. For a detailed technical description of the apparatus and of the CEP-stabilized laser system used in the measurements, see the Methods section.

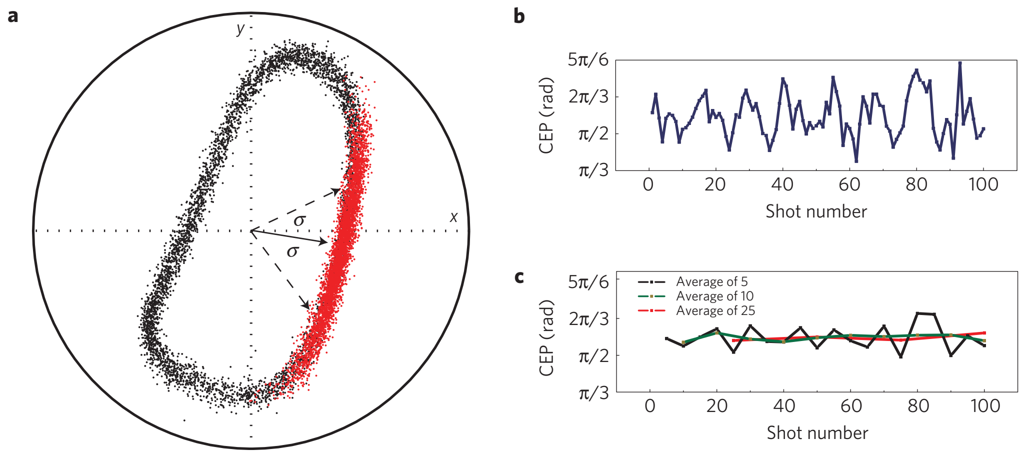

Figure 4 | Evolution of the CEP of consecutive laser shots. a, 4,500 consecutive phase-stabilized (red dots) and non-phase-stabilized (black dots; same as in Fig. 3) laser shots. The stabilized shots are distributed around the preset CEP with a standard deviation of $\sigma = 278$ mrad, which is indicated by arrows on both sides of the median CEP point. As both our measurement fluctuation and the distribution of the stabilized shots are nearly Gaussian, with a crude approximation the ratio of measurement precision over laser stability equals the ratio of width over length of the 'stripe' made by the red dots. **b**, Shot-to-shot evolution of the CEP of 100 consecutive phase-stabilized laser shots. **c**, Same evolution with the CEP values of 5/10/25 shots being averaged. The corresponding standard deviations (calculated from 4,500 shots) are 195 mrad, 140 mrad and 110 mrad, respectively.

High-precision single-shot CEP measurement

As CEP measurement techniques have so far relied on phase scans carried out with a sophisticated, but highly specialized, phase-stabilized laser, the precision of the measurement was inherently limited by the CEP fluctuations of the stabilized laser itself [18,30,31]. For state-of-the-art terawatt-class few-cycle lasers, phase stability with sufficiently low fluctuations[10] has so far been demonstrated only for a few minutes, and for even larger systems[9], stabilization has not yet been reported. Therefore, attempting to apply phase-scan measurements to these systems makes little sense. By exploiting single-shot detection without phase ambiguity, which is the unique feature of our measurement, we were able to determine the precision of our measurement without the need for phase stabilization.

The concept of such a first-principle calibration is the following: single-shot ATI spectra are measured with a non-phase-stabilized few-cycle pulse laser. From these spectra, the phase asymmetry parameters as defined above are evaluated and plotted in a parametric representation, see Fig. 3. Each point on the map (4,500 in total) corresponds to a single laser shot. All measurement points are distributed evenly around a loop, which represents a 2π phase range. This is in accordance with the fact that the phase distribution of a non-phase-stabilized laser is random (see Supplementary Movie S1 about consecutive non-phase-stabilized laser shots). Therefore, selecting any subset of, for example, 50 of the 4,500 points that are neighbouring each other by their polar angle on the plot means that these 50 points correspond to an interval of CEP phases of $\pi/45 (50 * 2\pi/4{,}500 = \pi/45)$. The mean points of the consecutive subsets of 50 points are indicated by 90 (4,500/50) blue dots in Fig. 3. In this way, we linearized the phase asymmetry parameters so that the blue dots around the elliptical curve indicate $\pi/45$ CEP shifts similarly to the minute ticks on a watch. A comparison with one-dimensional TDSE simulations leads to the CEP values on the loop. The points with the highest asymmetry at the cutoff energy (that is, the points with the maximum absolute value along the y axis) define the distinguished CEP settings of $\phi = 0$ and $\phi = \pi$. Higher resolution can be obtained by selecting smaller subsets of points. It is evident that the sensitivity of the phase measurement is highest at the CEP where the separation between two adjacent blue dots is largest. It is reasonable to assume that the width of the distribution in the radial direction is equivalent to the width of the distribution in the tangential direction for a fixed CEP. Figure 3 confirms that the distribution perpendicular to the loop is narrower the longer the distance between the blue dots indicating the $\pi/45$ intervals is. In this way, the precision of our method for different CEP settings can be evaluated, again solely relying on the random distribution of non-phase-stabilized few-cycle pulses. At the CEP of $5\pi/9$ (close to $y = 0$), indicated by red arrows in the figure, our measurement is most precise with a standard deviation of $\pi/300$ for its value, which corresponds to a temporal jitter of 4.5 as. This high precision is mainly due to the extreme phase sensitivity of highest energy rescattered electrons around the point where one phase asymmetry parameter is zero. The measurements are robust concerning pulse energy fluctuations and within the pulse duration fluctuations of state-of-the-art ultrafast lasers.

Today, several groups have characterized the phase stability of CEP-stabilized lasers by measuring the relative CEP change with out-of-loop f-to-2f interferometers[31,32]. These measurements were made at acquisition rates far below the laser repetition rates and by averaging over tens to hundreds of shots. Here, we present the first consecutive single-shot characterization of a stabilized laser using this same method as described for non-stabilized lasers, and further demonstrate superior precision in comparison with established methods. This was enabled by our new evaluation method that could decouple the phase fluctuations of the laser from the precision of the apparatus. The red points in Fig. 4a indicate 4,500 consecutive laser shots recorded with the same laser as above but with the CEP stabilized[33] to $5\pi/9$. (Supplementary Movie S2 shows a phase scan carried out in $\pi/10$ steps over a 2π range.) Even with our state-of-the-art stabilization scheme (see the Methods section), the standard deviation of the CEP of the stabilized shots was 278 mrad. That is more than 25 times worse than our precision at that CEP setting and it is also worse than reported previously[5]. The reason is that the standard deviation was derived from the CEP values of all individual shots, whereas in other measurements the CEP values are averaged over several shots. Figure 4b shows the shot-to-shot evolution of the CEP over 100 consecutive stabilized shots. Figure 4c shows that this evolution is more and more flattened out when the number of shots used for averaging the CEP increases, indicating that the single-shot measurement provides new insight into the CEP evolution of a laser system. Such measurements with our non-invasive diagnostics can be conducted along with running experiments on any type of intense few-cycle lasers and give rise to a deeper understanding of the CEP evolution for low-repetition-rate high-power lasers as well as for high-repetition (greater than kilohertz) lasers.

Summary and outlook

We present a robust single-shot CEP measurement technique without the need for previous phase stabilization. The CEP is measured without any ambiguity owing to a novel evaluation method of ATI spectra at a precision that can be as high as $\pi/300$ at an optimum measurement point.

The maximum acquisition rate of the apparatus is several times higher than the repetition rate of amplified few-cycle sources available at present, and only a small fraction of typically delivered pulse energies is necessary for a non-invasive CEP measurement. These unique features of the apparatus make it extremely versatile. As a direct application, the CEP of each laser pulse can be measured with a precision more than 25 times higher than with state-of-the-art stabilization techniques. High-order harmonic generation, ATI or terahertz emission are just a few of those strong-field phenomena that are directly governed by the electromagnetic field and actively studied with few-cycle pulses. With our single-shot measurement technique, experiments with non-phase-stabilized lasers can now be carried out and by simple CEP tagging, the dependence of strong-field effects on the electromagnetic waveform can be reconstructed with very low phase jitter. Gaining access to the CEP of a single laser shot will enable us for the first time to characterize the entire waveform of single ultrashort laser pulses using our apparatus along with well-established single-shot all-optical pulse characterization techniques[34,35].

The capability of our apparatus is not limited to CEP measurements. One of the foremost challenges in ultrafast optics is the measurement of steadily decreasing pulse durations now approaching the single-cycle limit[5]. Our measurement technique can also be used for that purpose in a very simple way. As expected, our numerical simulations show that the amplitudes of the sine-like phase asymmetries are increasing with decreasing pulse durations. Therefore, the polar angle of a measurement point on the parametric plot gives the CEP and its radius provides a measure of the pulse duration. Unlike conventional pulse duration measurement techniques, this method is dispersion-free and unlimited in bandwidth, and its accuracy is increasing with shorter durations. As the ATI cutoff is a direct measure of the pulse intensity, we will be able to measure the three most important parameters of few-cycle laser pulses with our apparatus in a single shot: CEP, pulse duration and intensity.

Measurement and control of high-power (much greater than terawatt) few-cycle lasers is another intriguing future application of our measurement technique. Extending the waveform control to few-cycle sources that deliver $>10^{18}$ W cm^{-2} of focused intensity, holds promise for the synthesis of intense isolated attosecond extreme-ultraviolet pulses in the kiloelectronvolt photon energy range. These unique sources are expected to open the door to a wide range of applications in physics, biology and chemistry.

Methods

Single-shot stereo-ATI phase meter. The single-shot stereo-ATI phase meter is composed of two ideally identical TOF spectrometers in opposing orientation to each other. They measure ATI electron spectra from xenon atoms at the two sides of the polarization axis of few-cycle laser pulses. Xenon gas is located in a small cell in the centre of the apparatus held at a constant pressure of 1.6×10^{-2} mbar by a continuous gas flow and simultaneous pumping. In all other parts of the apparatus, differential pumping by a turbo pump maintains a high vacuum of 4×10^{-6} mbar. This is necessary for safe operation of the MCP detectors and ensures free flight of the electrons.

A 10% split-off of the 400 μJ total laser output is focused into the interaction cell by a spherical mirror with a focal length of 250 mm. Alternatively, the apparatus can be operated as a non-invasive CEP diagnostics tool by using the entire beam and focusing it slightly. Electrons are ionized by the linearly polarized laser field close to the focus at an intensity of 8×10^{13} W cm^{-2} and leave the cell through 0.7-mm-thin vertical slits, which hold back electrons generated outside the laser focal region. After a 15.5 cm flight, the electrons are detected by MCP detectors (Del Mar Photonics). A −25 V blocking potential allows only electrons with a kinetic energy >25 eV to reach the detectors. As explained earlier, these electrons have a much higher phase sensitivity than low-energy electrons. The drift tubes and the interaction zone are protected from electric and magnetic fields by a μ-metal-shielding. The signals of the MCP detectors are amplified by two wide-band amplifiers and recorded by a digital oscilloscope (Tektronix). With the digital data acquisition we are able to consecutively record pulses up to 100 kHz repetition rates. At present, the maximum number of pulses is limited to ∼4,500 shots by the oscilloscope, which corresponds to 1.5 s at 3 kHz repetition rate of the laser. With suitable digitizer cards, which are now available commercially, this limit vanishes and recording of all laser shots is technically possible.

CEP-stabilized laser system. Our CEP-stabilized laser system[32] is composed of a Ti:Sa oscillator (5 fs, 5 nJ, 70 MHz) and a multipass amplifier (FEMTOPOWER compact PRO CEP, 3 kHz) delivering 20 fs pulses. These pulses are spectrally broadened in a hollow-core fibre (HCF) filled with neon[36] and temporally compressed on multilayer mirrors ('chirped' mirrors). The CEP measurements were carried out for pulses with a duration of 4.1 fs and an energy of 40 μJ, which is 10% of the total pulse energy from the system. To ensure that the pulses have identical CEP, we use an 'f-to-0' phase-stabilization scheme[37] ('fast-loop') for the oscillator and an 'f-to-2f' set-up[18] ('slow-loop') after the amplifier. To minimize the phase jitter, we installed the slow loop behind the HCF rather than directly after the amplifier. This resulted in a CEP-stabilization quality superior to previous schemes. Here, the extra noise from spectral broadening inside the f-to-2f measurement set-up has been eliminated by directly using the octave-spanning spectrum after the HCF. Moreover, this way the phase noise added by the HCF is also included in the determination of the error signal, which is fed back to modulate the pumping power of the oscillator for phase stabilization.

Received 8 October 2008; accepted 20 March 2009; published online 19 April 2009

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Acknowledgements

We thank L. Veisz for advice during the measurements and F. Krausz for his support. G.G.P. acknowledges support by the Welch foundation (A-1562) and the Airforce Office of Scientific Research AFOSR. R.K. acknowledges financial support from the Sofja Kovalevskaja Award of the Alexander von Humboldt Foundation. The work was supported by the DFG Cluster of Excellence: Munich Centre for Advanced Photonics (www.munich-photonics.de).

Additional information

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