



Carrier-envelope-phase measurement of few-cycle mid-infrared laser pulses using high harmonic generation in ZnO

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Abstract: High-harmonic generation (HHG) in crystals offers a simple, affordable and easily accessible route to carrier-envelope phase (CEP) measurements, which scales favorably towards longer wavelengths. We present measurements of HHG in ZnO using few-cycle pulses at 3.1 μm . Thanks to the broad bandwidth of the driving laser pulses, spectral overlap between adjacent harmonic orders is achieved. The resulting spectral interference pattern provides access to the relative harmonic phase, and hence, the CEP.

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1. Introduction

In recent years, strong-field laser physics has experienced a shift towards longer wavelengths, i.e., from the near-infrared to the mid-infrared (mid-IR) spectral range. On one hand this shift is motivated by the beneficial scaling law of the ponderomotive energy $U_p \sim \lambda^2$ [1,2]. As a result, electrons can be accelerated to very high kinetic energies, even at moderate field strength. Using mid-IR wavelength drivers, this has led to the demonstration of bright x-ray sources [3], soft x-ray attosecond pulses [4,5] high-resolution laser-induced electron diffraction [6,7] and a long-living, keV-temperature, high-charge-state plasma [8]. On the other hand, the low linear excitation rate of mid-IR light in wide band gap semiconductor and dielectric materials allows for applying electric fields nearly as strong as the the critical electric field strength [9,10]. This allows for transitioning from conventional non-linear optics to strong-field physics in solids without inflicting optical damage to the sample [11]. This transition is marked by non-perturbative high harmonic generation (HHG) in solids [12–17], including an HHG plateau, analogous to gases [18].

Accompanying the discovery of new physical phenomena tremendous progresses has been achieved improving the performance of mid-IR laser sources. Pulse energies up to tens of milli-joules [19], high repetition rates [20,21] and pulse durations down to a few optical cycles [19–23] are now available. Approaching the few cycle regime makes precise control over the carrier envelope phase (CEP) necessary [24]. CEP measurement is typically achieved using an f-2f-Interferometer, where an octave spanning fundamental spectrum is overlapped with its second harmonic signal. In the spectral overlap region, a fringe pattern reveals the *relative* CEP [25,26]. With mid-IR pulses, however, the overlap region between the fundamental and second harmonic may lie outside the detection range of well-developed and affordable Silicon based detectors. Some drawbacks of the f-2f interferometer can be overcome by the stereographic above-threshold ionization (Stereo-ATI) technique for single-shot measurements of the *absolute* CEP [27–29]. While the Stereo-ATI has been demonstrated to work up to the short-wave infrared [30], its application in the mid-IR has been hampered due to the unfavorable scaling of the recollision probability with increasing wavelength [31,32]. More recently, new techniques for determining the CEP of few-cycle pulses under ambient laboratory conditions have been demonstrated [33,34].

Here, we present measurements of HHG in ZnO using few-cycle laser pulses in the mid-IR (3.1 μm). We show that the interference between adjacent harmonic orders delivers a fringe pattern [35], from which the relative CEP can be readily extracted. Importantly, our scheme scales well towards longer wavelengths, where higher harmonic orders can be achieved. Our approach represents a simple and cost-effective method for measuring the relative CEP, and could be easily integrated into a CEP stabilization scheme.

2. Experimental approach

The experiments were carried out using the MIR laser at ELI-ALPS, which provides laser pulses centered around 3.1 μm at a repetition rate of 100 kHz. The output 140 μJ pulses from the optical parametric chirped pulse amplification (OPCPA) system [36] is compressed to ~ 25 fs (ca. 2.5 optical cycles) using non-linear spectral broadening in a 2 mm thick yttrium aluminum garnet (YAG) crystal [19,22,23] and a 1 mm thick Si plate, followed by a BaF₂ wedge pair. The CEP of the laser pulses is passively stable and additionally actively stabilized and controlled by an acousto-optical phase modulator (Fastlite, Dazzler) which receives feedback from an f-2f interferometer in combination with a CEP measurement processing unit (Fastlite, Fringezz). Besides stabilizing the CEP, the Dazzler can be used to vary the CEP continuously, while the f-2f interferometer monitors the CEP variations.

Figure 1(a) shows a schematic of the experimental setup. The laser pulses are focused by a gold coated concave mirror ($f = 15$ cm) to a spot size of ~ 200 μm . The peak intensity and field strength were approximately 3 TW/cm^2 and 0.4 $\text{V}/\text{\AA}$, respectively and clearly below the reported damage threshold value [12]. A 100 μm thick c-cut ZnO crystal (Crystec GmbH) is placed in the focal plane. The generated high harmonic signal was focused to the entrance slit of a UV-VIS spectrometer (Avantes Avaspec ULS3648) by an $f = 5$ cm CaF₂ lens. The integration time was typically 1 ms, which corresponds to averaging over 100 laser shots. A fused silica window was placed after the sample and in front of the spectrometer to avoid focusing intense mid-IR light onto the entrance slit of the spectrometer. All experiments were carried out at room temperature and ambient atmosphere (21.5°C, 45% relative humidity). In addition, measurements with the 134 fs pulses were performed, using 500 μm thick non-centrosymmetric a-cut ZnO as a target. The laser spectra with and without post-compression are shown in Fig. 1(b).

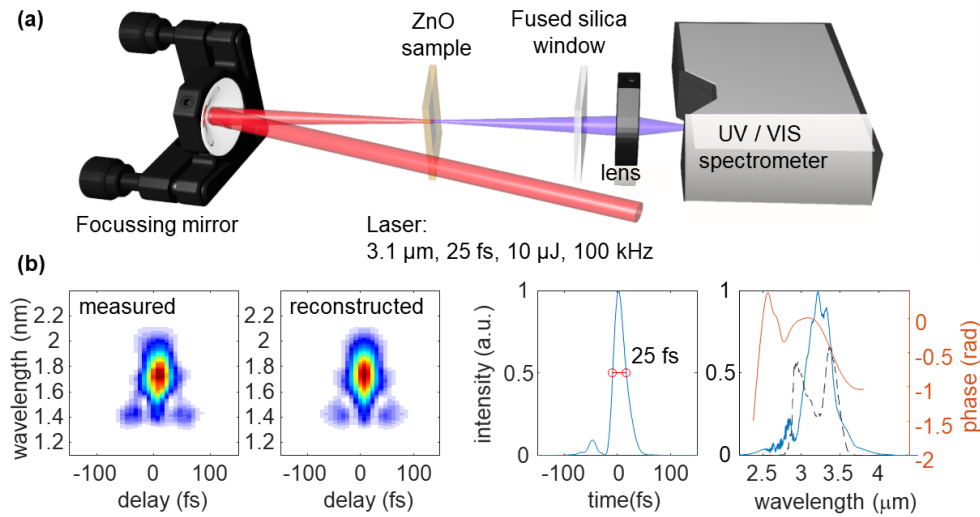


Fig. 1. (a) Experimental setup for CEP measurements in the mid-IR using HHG in ZnO. Mid-IR laser pulses with controllable CEP are focused onto a ZnO crystal. The fundamental laser light is removed using a fused silica window. The high harmonic signal is collected with a CaF₂ lens and detected with a UV-VIS spectrometer. (b) Pulse characterization results using frequency resolved optical gating (FROG). Shown are (from left to right) the measured and reconstructed FROG traces, the retrieved pulse shape in the time domain, and the measured spectrum (blue, solid line) and retrieved spectral phase. The dashed black line shows the spectrum without spectral broadening.

3. Results and discussion

In Fig. 2(a), a typical recorded spectrum with harmonics up to the 13th order is shown. Using the c-cut crystal, only odd harmonic orders are detected due to the inversion symmetry of the c-cut ZnO crystal [37]. The even orders appear when an a-cut crystal is used. For clarity of the peak structure, we present data for 134 fs pulses.

Figure 2(b) shows the recorded CEP-dependence of the harmonic spectrum. In this measurement, the CEP of the driving laser pulses was varied with a triangular wave using the Dazzler, i.e. a constant slope from 0 to 2π and back. Between all odd harmonics we observe a clear oscillating structure of diagonal lines as a function of the CEP. The variations in the signal strength (vertical features) are attributed to short-term fluctuations of the laser power or pointing. Importantly, with regard to CEP measurements, the CEP-dependent interference pattern is not notably affected by these fluctuations. We attribute this insensitivity to the very similar intensity scaling of adjacent order harmonics in the non-perturbative regime [12]. Moreover, pointing fluctuations do not significantly affect the fringe pattern since different harmonic orders propagate collinearly.

Another pronounced feature of the spectra presented in Fig. 2 consists in the strong modulations of the 5th harmonic, which are independent of CEP. These oscillations result from the interference between the 5th harmonic radiation generated at the front and rear surfaces of the ZnO sample. Thus, the spectral spacing in the 5th harmonic is given by:

$$\Delta\lambda = \frac{\lambda^2}{c\Delta t} = \frac{\lambda^2}{cd} \left(\frac{1}{v_g(\lambda)} - \frac{1}{v_g(\lambda/5)} \right)^{-1} \quad (1)$$

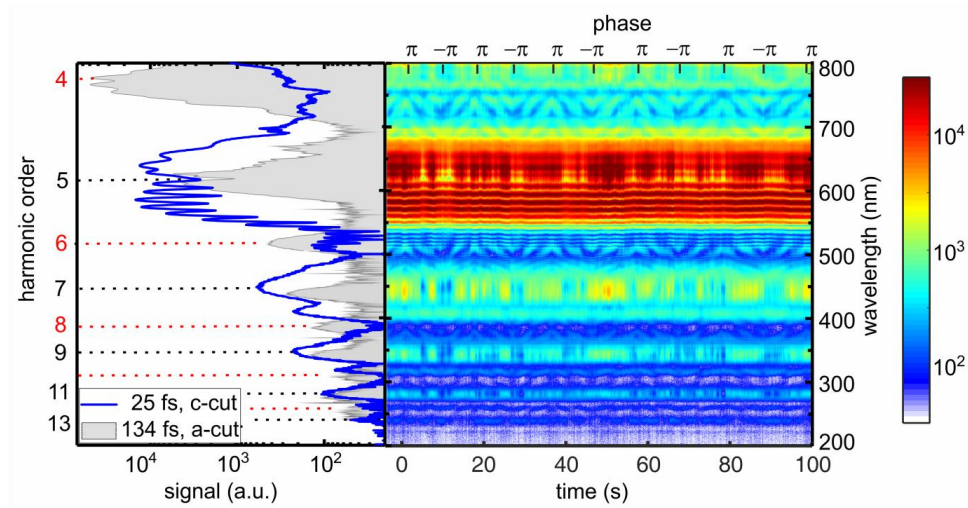


Fig. 2. a) Typical spectrum showing high harmonics of various orders, as indicated. Data is presented using two different crystal geometries and pulse durations, as specified in the legend. b) Measured series of high harmonic spectra using 25fs pulse and a c-cut ZnO crystal, while the CEP is varied using the Dazzler. The colorbar encodes the measured signal strength on a logarithmic scale.

Here d denotes the thickness of the ZnO sample (100 μm), $v_g = \frac{c}{n} \left(1 - \frac{\lambda}{n} \frac{dn}{d\lambda}\right)^{-1}$ the group velocity of the fundamental beam and the 5th harmonic, n the refractive index and c the speed of light.

Figure 3(a) shows an enlarged view of the 5th harmonic spectrum, where the change of the variation of the fringe spacing is apparent. Using Eq. (1), the fringe spacing $\delta\lambda(\lambda)$ is calculated and compared to the measurements in Fig. 3(b). The measured and calculated values coincide nearly perfectly, indicating that due to the wide spectrum of the 5th harmonic the modulation frequency changes over the spectrum. The spectral interference disappears below ~ 500 nm, due to absorption of the light produced at the front surface of the ZnO crystal. Thus, below 500 nm, only the harmonic light generated at the rear surface reaches the detector.

We now turn towards extracting the CEP from the phase-dependent interference pattern observed in Fig. 2(b). To this end, measurements were performed, in which the CEP was varied in a triangular pattern, Fig. 4(a), and in a sinusoidal pattern, Fig. 4(b). The spectra were recorded using a window filter with transmission in the shown wavelength range. They show the 9th and 11th harmonic of the 3.1 μm laser, and clear CEP-dependent interference patterns in the vicinity of the harmonics. The triangular and sinusoidal phase variations are clearly imprinted in the interference patterns.

Thanks to the high fringe contrast (up to 3:1) of the interference pattern, the relative phase of adjacent harmonics can be retrieved from individual spectra. This is achieved using a fast Fourier transform of the spectrum in the range (260, 400) nm. The relevant frequency of the interference fringes is identified in the Fourier power spectrum and its phase is unwrapped and plotted in Figs. 4(c) and 4(d) for the triangular and sinusoidal patterns, respectively. The reconstructed phase from the HHG signal (blue line), is compared to twice the CEP measured using the f-2f interferometer at the laser output (red dots). It is important to note that the relative phase of the harmonics with order n and $(n+2)$ varies by $2\Delta\varphi$ when the CEP is changed by $\Delta\varphi$. Therefore, the reconstructed phases in Figs. 4(c) and 4(d) oscillate between 0 and 4π while the CEP was varied between 0 and 2π .

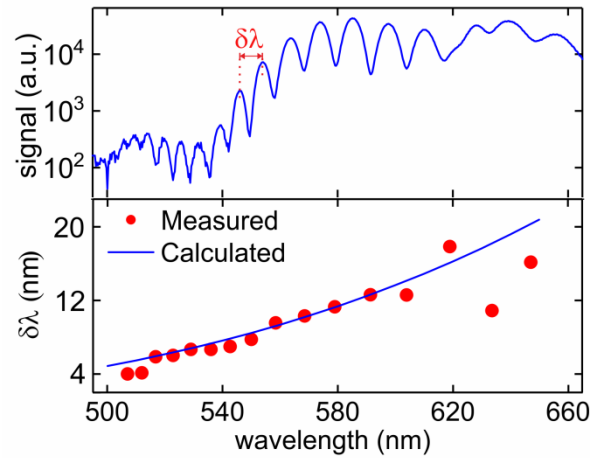


Fig. 3. Spectral interference in the 5th harmonic. (a) Measured spectrum in the vicinity of the 5th harmonic, exhibiting a narrow fringes pattern due to interference between harmonics generated on the front and back surface of the ZnO sample. (b) Measured and calculated (using Eq. (1)) spectral fringe spacing in the 5th harmonic.

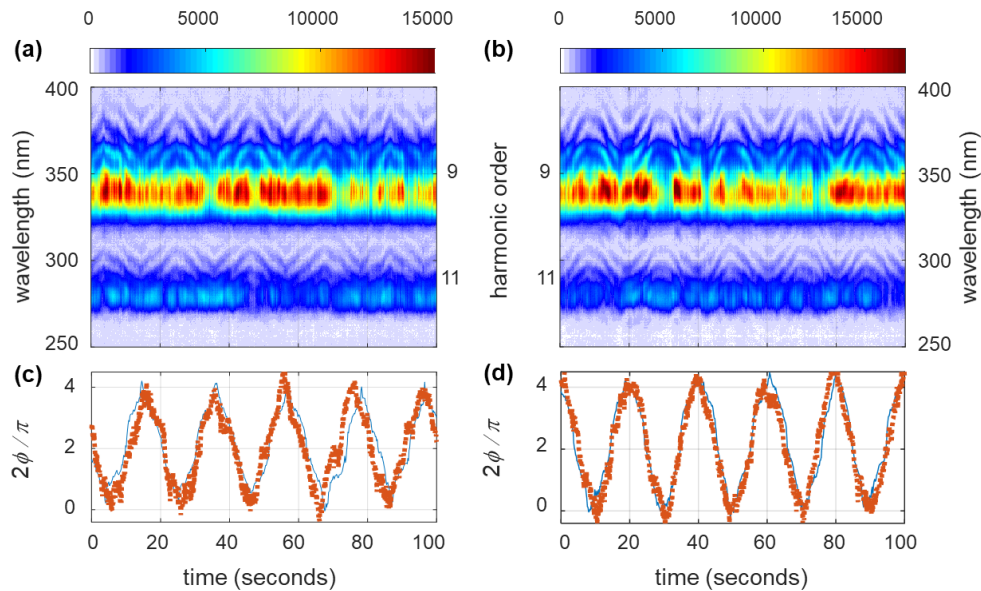


Fig. 4. CEP measurement using HHG in ZnO. Shown are series of spectra in the region around the 9th and 11th harmonic. The CEP of the fundamental pulses is varied in a triangular pattern (a) or a sinusoidal pattern (b) with a frequency of 1/20 Hz. The fringe patterns in between the harmonic signals is used to obtain the relative phase between adjacent odd-order harmonics, which corresponds to twice the CEP ϕ . Panels (c) and (d) show the reconstructed values of 2ϕ (blue solid lines) along with the corresponding values recorded by the f-2f interferometer (red dotted lines).

The increased sensitivity to CEP variations leads to an ambiguity of π when measuring the CEP via spectral interference of odd order harmonics. This ambiguity could be removed by using non-isotropic media such as a-cut ZnO, where even harmonics can be generated. In that case, the same procedure could be applied to neighboring even and odd order harmonics, whose relative phase varies by $\Delta\varphi$ when the CEP is changed by $\Delta\varphi$. A typical spectrum using a-cut ZnO is shown in Fig. 2(a). CEP-dependent oscillations are observed for a sufficiently broad bandwidth, in between odd and even order harmonics.

We have shown above that the *relative* CEP can be reliably measured using the spectral interferences in the HHG signal. In the following, we utilize this capability to obtain a phase-resolved high harmonic spectrum from the measured data. In Fig. 5, we present the normalized difference, $\frac{S(\phi)-R}{S(\phi)+R}$, between the CEP-dependent harmonic spectra, $S(\phi)$, and the CEP-averaged spectra $R = \frac{1}{2\pi} \int S(\phi) d\phi$. This representation of the data shows red and blue areas with pronounced tilts, corresponding the interference fringes between the different harmonic orders. As has been shown in the context of multiphoton ionization [38], the tilts are related to the uncompensated spectral phase in the fundamental spectrum, see Fig. 1. In addition, the yield of the high harmonics exhibits a clear CEP dependence with a π periodicity, as well. Contrary to the interference fringes between the harmonics, the modulations on the maxima of the 9th and 11th harmonics do not exhibit a tilt along the wavelength axis. This suggests that they do not originate in spectral interference but rather in the HHG process itself. Therefore, the modulations in the harmonic yield can be used to determine the *absolute* CEP on target, except for a π ambiguity. For this determination, knowledge over what value of the CEP leads to the maximum harmonic yield is required from theory.

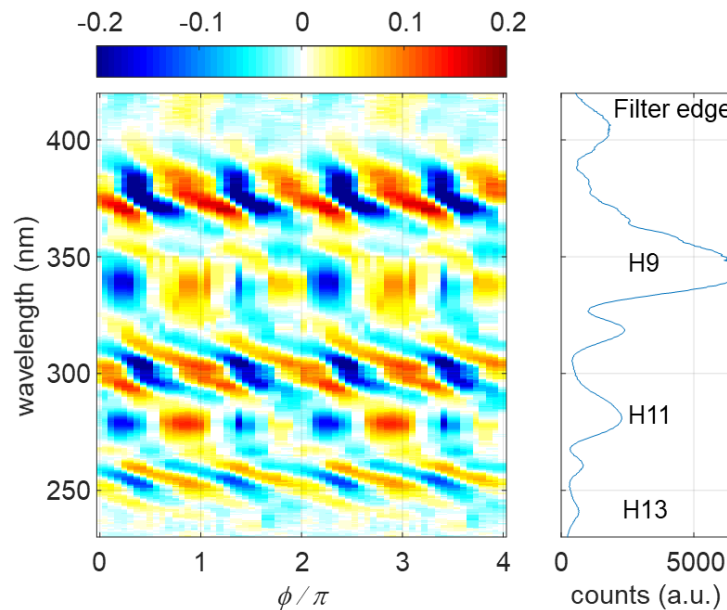


Fig. 5. CEP dependence of the high harmonic spectrum generated by a few-cycle laser pulses at 3.1 μm . The color code denotes the normalized difference between the signal obtained at a given CEP and the CEP-averaged signal. The relative CEP ϕ is obtained from the interference fringes between the harmonics using the method described in the text. Harmonic orders and the filter edge near 400 nm are indicated in the spectrum

We note that the CEP dependence of the HHG process in crystals is not yet well understood, even though it has been investigated in various materials and for different driving laser wavelengths [39–43]. In particular, a shift of the photon energy of high harmonics as a function of the CEP, i.e. a tilt along the wavelength axis, have been reported in GaSe [39], fused silica [41], MgO [40] and Si [43] using THz, NIR and mid-IR few-cycle laser pulses, respectively. A detailed explanation of the origin of the tilt-free modulations observed in our experiments using ZnO is beyond the scope of the present work and will be the topic of further experiments.

4. Conclusion and outlook

In conclusion, we have shown that HHG in solids such as ZnO allows for a simple detection scheme for the CEP of mid-IR laser pulses. Because it is based on the spectral interference of high harmonics of orders $(n, n + 2)$ ($n = 3, 5, 7, \dots$), it does not require an octave spanning spectrum; rather, a spectrum with a relative bandwidth $\frac{\Delta\lambda}{\lambda} \geq \frac{2}{n}$ is sufficient to obtain overlap between adjacent harmonics of order n or higher. Using non-centrosymmetric crystals further relaxes this condition due to the appearance of even order harmonics. This would also remove the π ambiguity in the CEP measurement but may require usage of a polarizer, because the polarization states of even and odd-order harmonics may be different [44,45]. Similar to its use in an f-2f interferometer, the polarizer would allow for tuning the fringe contrast between even and odd-order harmonics.

The presented approach scales favorably towards longer wavelength, as the amount of overlap between adjacent harmonics increases with the harmonic orders, and longer wavelengths produce higher-order harmonic orders for suitable targets [16]. The possibility to detect the CEP using light in the visible spectral range allows for the use of affordable silicon-based photon detectors, and makes the presented method very economical. In addition, modulations in the harmonic yield would in perspective allow for the measurement of the absolute CEP. With improved signal-to-noise ratio, it is in principle possible to perform single shot measurements of the high harmonic spectra, and thus, measuring the CEP even for lasers, which are not CEP stable.

Finally, the interference patterns between different pairs of harmonics carries information on the “attochirp” [46], i.e. the relative phase of the various harmonics as they are generated. This information could directly reveal the time structure of the harmonic pulses, an important milestone towards attosecond pulse generation from solids [42].

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