Single-shot measurement of few-cycle optical waveforms on a chip

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The measurement of transient optical fields has proven critical to understanding the dynamical mechanisms underlying ultrafast physical and chemical phenomena, and is key to realizing higher speeds in electronics and telecommunications. However, complete characterization of optical waveforms requires an ‘optical oscilloscope’ capable of resolving the electric-field oscillations with sub-femtosecond resolution and with single-shot operation. Here we show that strong-field nonlinear excitation of photocurrents in a silicon-based image sensor chip can provide the sub-cycle optical gate necessary to characterize carrier-envelope phase-stable optical waveforms in the mid-infrared. By mapping the temporal delay between an intense excitation and weak perturbing pulse onto a transverse spatial coordinate of the image sensor, we show that the technique allows single-shot measurement of few-cycle waveforms.

The development, characterization and control of intense, few-cycle optical fields have enabled the generation of isolated attosecond pulses1 and ushered in a new regime of field-resolved optical spectroscopy and microscopy (or ‘fieldoscopy’2) techniques3-5. These advances, in turn, have stimulated the development of pulse characterization tools capable of measuring the complex electric field—including the carrier-envelope (or ‘absolute’) phase (CEP)6—of a few-cycle laser field. So far, these techniques have been based either on field-driven ‘streaking’ of photoionized electrons7,8, electro-optic sampling (EOS)9,10 or the optical perturbation of a strong-field response11, and in all cases have required measurement of the spatial variation of the fundamental beam profile (Methods, Supplementary Note 1 and Supplementary Figs. 2 and 3), and a time delay onto the transverse position17, we further take advantage of the spatial resolution inherent to image sensors to realize single-shot detection of mid-IR laser waveforms.

The experimental measurement, which is an extension of the TIPTOE (tunnelling ionization with a perturbation for the time-domain observation of an electric field) technique6,14, is described schematically in Fig. 1a and in the Methods section. Briefly, an intense fundamental pulse with a central wavelength of 3.4 µm creates charge packets in the pixels of a silicon-based image sensor via multiphoton excitation, leading to detectable photocurrents. The probability of excitation is perturbed by the field of a weak perturbation pulse, leading to a modulation in the excitation probability and therefore in the magnitude of the detected photocurrent. We have previously shown that, for collinear fundamental and perturbation pulses, the dependence of the modulation in the excitation probability on the time delay between the two pulses encodes the time-varying electric-field waveform of the laser pulse11. Here, by using a crossed-beam geometry with cylindrical focusing, we map the time delay onto a transverse spatial coordinate of the image sensor chip to achieve single-shot detection.

The experimental image obtained from the sensor chip is shown in Fig. 1b, along with lineouts showing the magnitude of the photocurrent as a function of the spatial position on the detector with and without the perturbation pulse present. Here, the image and lineouts represent a true single-shot measurement, without integrating over multiple pulses or averaging multiple images. The signal is dominated by multiphoton excitation by the fundamental pulse, and the perturbation pulse produces no photocurrent on its own. Instead, the presence of the perturbation pulse is observed as a weak modulation on the signal produced by the fundamental pulse. Importantly, this modulation does not arise due to linear interference between the fundamental and perturbation pulses, but rather as a cross-correlation between the perturbation pulse and a sub-cycle electro-optic gate arising due to the multiphoton excitation of charge packets in the image sensor chip14. From the perturbed photocurrent signal, the modulation waveform of the perturbation pulse (Fig. 1c) can be obtained by subtracting the fundamental-only signal, followed by a normalization process to remove the effects of the spatial variation of the fundamental beam profile (Methods, Supplementary Note 1 and Supplementary Figs. 2 and 3), and a higher signal-to-noise ratio can be obtained by averaging over multiple identical laser shots. Finally, we take the Fourier transform of the modulation waveform to obtain the frequency spectrum, which can be compared to an independent measurement of the spectrum, and the spectral phase.

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Details of the nonlinear compression set-up and the CEP stabil-

mirror. The incoming laser pulse is separated into an intense fundamental pulse $E_1$ (Supplementary Fig. 12). However, this is not an inherent limitation to increase the signal-to-noise ratio (Supplementary Note 2 and ~100 laser shots to use the full dynamic range of the camera and the compressed pulses, each experimental image is integrated over optical cycles, as shown in Fig. 2a. Owing to the low intensity of the mid-IR pulses are described in the Methods section and bulk yttrium aluminium garnet (YAG) and silicon windows 20–22.

To achieve this, we temporally compress CEP-stable mid-IR pulses to a duration of ~2.5 optical cycles or below and with stable CEP. To do this, the CEP of the fundamental pulse is set to zero by replacing the CEP of the perturbation pulse, but rather the relative CEP between the fundamental and perturbation. Measuring the full electric-field waveform—including the CEP—of the perturbation pulse provides a pulse measurement but not necessarily a waveform measurement. In those measurements, as was the case in earlier work using nonlinear optical amplitude of the second-harmonic modula-
tion provides a pulse measurement. As a result, the modulation at the second-harmonic frequency is strongly suppressed, leaving instead only a weak oscillation at the fourth harmonic ($4\omega$). However, for sine-like pulses, or for multi-cycle pulses, the excitation events in adjacent half-cycles will be enhanced and suppressed, respectively, by the addition of the second-harmonic pulse. As a result, the modulation at the second-harmonic frequency is strongly suppressed, leaving instead only a weak oscillation at the fourth harmonic ($4\omega$) of the fundamental field. Figure 3 shows the modulated signals associated with a second-harmonic perturbation pulse using cosine- and sine-like fundamental pulses, as well as the CEP-dependent amplitude of the second-harmonic modulation signal. The CEP was controlled by varying the thickness of a CaF$_2$ wedge pair placed in the incident beam. As shown, the CEP of the fundamental pulse can be set to zero simply by maximizing the amplitude of the second-harmonic modulation.

Fig. 1 | Experimental set-up and principle of single-shot measurement. a, Schematic of the experimental set-up. BS, beamsplitter; CCM, concave cylindrical mirror. The incoming laser pulse is separated into an intense fundamental pulse $E_1$ and weak perturbation pulse $E_2$, which are focused with a cylindrical mirror onto a silicon-based complementary metal–oxide–semiconductor image sensor. For CEP determination, the perturbation pulse can be replaced with a second-harmonic pulse. b, Experimentally measured single-shot image (top) and lineouts (bottom) showing the modulated nonlinear photocurrent induced by the perturbation pulse. F, fundamental; P, perturbation. c, Measured modulation waveform after subtraction of the fundamental-only signal and normalization. The inset shows a zoomed-in region near the center of the temporal profile. d, Spectra and spectral phases of the measured mid-IR waveforms. The spectrum measured using a grating spectrometer with a PbSe detector is shown by a shaded area for comparison. The grey lines in c and d correspond to ten consecutive single-shot images obtained under identical conditions and without averaging, and the red line shows an averaged waveform, obtained by averaging over the ten images. Details of the experimental set-up and normalization procedure are provided in the Methods section.

Our previous work using band fluorescence to detect the modulation of the multiphoton excitation probability showed that the modulated signals in Fig. 1 reflect the electric-field envelope and the time-dependent phase (that is, the carrier frequency and the frequency chirp) of the perturbation pulse, even for multi-cycle pulses. That is to say, the modulation provides a pulse measurement but not necessarily a waveform measurement. In those measurements, as was the case in earlier work using nonlinear optical detection19, the CEP of the modulation waveform does not represent the CEP of the perturbation pulse, but rather the relative CEP of the fundamental and perturbation. Measuring the full electric-field waveform—including the CEP—of the perturbation pulse therefore requires that the CEP of the fundamental pulse be set to zero. This, in turn, requires the use of few-cycle pulses with a duration of ~2.5 optical cycles or below and with stable CEP. To achieve this, we temporally compress CEP-stable mid-IR pulses to a duration of 2.1 optical cycles using nonlinear propagation in bulk yttrium aluminium garnet (YAG) and silicon windows20–22. Details of the nonlinear compression set-up and the CEP stability of the mid-IR pulses are described in the Methods section and Supplementary Figs. 1 and 7, respectively. In this case, the modulated signal corresponds to a pulse with a duration of 24.0 fs, ~2.1 optical cycles, as shown in Fig. 2a. Owing to the low intensity of the compressed pulses, each experimental image is integrated over ~100 laser shots to use the full dynamic range of the camera and thus increase the signal-to-noise ratio (Supplementary Note 2 and Supplementary Fig. 12). However, this is not an inherent limitation of the technique, and single-shot measurements of slightly longer few-cycle pulses are presented in Supplementary Figs. 11 and 12. We confirm the validity of these measurements both by comparing to an independent measurement of the spectrum (Fig. 2b) and by measuring the spectral phase associated with the dispersion of a 2-mm-thick CaF$_2$ window placed in the perturbation pulse beam path (Fig. 2c), which agrees well with the phase calculated from the Sellmeier equation23.

The CEP of the fundamental pulse is set to zero by replacing the perturbation pulse with a weak second-harmonic field, as previously described in ref. 11. For a sufficiently short pulse with a cosine-like waveform, multiphoton excitation is temporally confined to a single half-cycle at the centre of the pulse. In this case, the detector signal will exhibit modulations at the second-harmonic frequency $2\omega$. However, for sine-like pulses, or for multi-cycle pulses, the excitation events in adjacent half-cycles will be enhanced and suppressed, respectively, by the addition of the second-harmonic pulse. As a result, the modulation at the second-harmonic frequency is strongly suppressed, leaving instead only a weak oscillation at the fourth harmonic ($4\omega$) of the fundamental field. Figure 3 shows the modulated signals associated with a second-harmonic perturbation pulse using cosine- and sine-like fundamental pulses, as well as the CEP-dependent amplitude of the second-harmonic modulation signal. The CEP was controlled by varying the thickness of a CaF$_2$ wedge pair placed in the incident beam. As shown, the CEP of the fundamental pulse can be set to zero simply by maximizing the amplitude of the second-harmonic modulation.
We next demonstrate the single-shot measurement of laser electric-field waveforms with different CEP values. We first set the CEP of the fundamental to zero, as described above, and then scan the CEP of the perturbation pulse by varying the thickness of a wedge pair placed in the perturbation pulse beam path. The results are shown in Fig. 4. By varying the CEP of the perturbation pulse by $\sim \pi$ rad, we observe that the signal modulations transform from a positive to a negative sine-like waveform. The observed CEP change agrees well with that expected from the insertion of the glass wedge.

The ability to resolve the electric-field waveform of a few-cycle light pulse in a single shot presents numerous opportunities to resolve attosecond dynamics in light–matter interactions$^{24}$, as well as the impulsive responses of molecules to intense ultrashort fields and their time-domain signatures$^{25}$. Moreover, the inherent spatial resolution associated with the use of a two-dimensional detector...
(Supplementary Note 5 and Supplementary Figs. 13 and 14) is likely to enable new perspectives into the rich spatiotemporal behaviour found throughout nonlinear optics. Broader applications, however, will require extension of this technique to both shorter and longer wavelengths. Although the technique described here remains valid in both the multiphoton and tunnelling excitation regimes, and therefore the experimental set-up based on the use of a silicon image sensor is probably suitable for the detection of longer-wavelength pulses, a different detector technology will be required for extension to the near-IR and potentially the visible spectrum. We have previously shown that multiphoton excitation in ZnO is suitable for wavelengths down to ~900 nm (ref. 15) and therefore it may be possible to spatially resolve the band fluorescence from ZnO or other dielectric materials with larger bandgaps. Alternatively, AlGaN image sensors, developed for solar-blind detection in the ultraviolet–NIR range, may provide a purely opto-electronic solution.

Online content

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References

1. Chini, M., Zhao, K. & Chang, Z. The generation, characterization and applications of broadband isolated attosecond pulses. Nat. Photon. 8, 178–186 (2014).
2. Alismail, A. et al. Near-infrared molecular fieldscopy of water. Proc. SPIE 10882, 1088231 (2019).
3. Sederberg, S. et al. Attosecond optoelectronic field measurement in solids. Nat. Commun. 11, 430 (2020).
4. Pupeza, I. et al. Field-resolved infrared spectroscopy of biological systems. Nature 577, 52–59 (2020).
5. Fattahi, H., Fattahi, Z. & Ghorbani, A. Prospects of third-generation femtosecond laser technology in biological spectromicroscopy. J. Opt. 20, 054005 (2018).
6. Paulus, G. G. et al. Measurement of the phase of few-cycle laser pulses. Phys. Rev. Lett. 91, 253004 (2003).
7. Goulielmakis, E. et al. Direct measurement of light waves. Science 305, 1267–1269 (2004).

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Methods
Experimental set-up. A schematic of the experimental set-up for single-shot measurement of optical waveforms is shown in Supplementary Fig. 1. Mid-IR pulses are produced by a two-stages optical parametric amplifier (OPA, Light Conversion ORPHUS-ONE, LCO), which is pumped by a commercial Yb:KGW amplifier (KGW, potassium gadolinium tungstate; Light Conversion Carbide). The OPA idler is tunable from 2.2 to 4.5 μm, and provides ~10 μJ, 90-fs pulses at a repetition rate of 10 kHz for the wavelengths used in the experiments. First, the single-shot on-chip detection technique was demonstrated by characterising multi-cycle mid-IR pulses, as in ref. 11. To do so, OPA idler pulses with central wavelengths of 3.1, 3.4 and 3.8 μm were selected. The mid-IR pulse characterization and dispersion measurements are displayed in Supplementary Figs. 9 and 10, respectively. Although the multi-cycle pulse measurement is not sensitive to the CEP of the perturbation pulse, the results confirm the good performance of the technique for pulse characterization. Measuring the full optical waveform requires that the CEP of the fundamental pulse be set to zero. To do so requires the use of CEP-stable fundamental pulses with a pulse duration below 2.5 optical cycles, and a second-harmonic perturbation pulse to detect the fundamental CEP11. Using a 2J-to-j interferometer17, we find that the CEP of the idler pulses is passively stable, as shown in Supplementary Fig. 7, which was expected based on the OPA geometry. For measurements with few-cycle pulses, the OPA idler waveform was set to 3.5 μm, and the second-harmonic pulses centred at 1.75 μm were generated with ~2% efficiency using an AlGaS2 (AGS) crystal (thickness of 1 mm) cut for type I phase matching. After the AGS crystal, a pair of CaF2 wedges were used to control the CEP of the fundamental pulse, which was used to split the fundamental and second-harmonic pulses. For pulse compression, the mid-IR pulses were focused by a silicon lens (f = 100 mm) through three windows (2-mm-thick silicon, 5-mm-thick YAG and 2-mm-thick silicon)18. The first window was placed 50 mm before the focal spot, while the front surfaces of the YAG and the second silicon plates were placed 2 and 10 mm after the focal spot, respectively. After the three plates, the mid-IR pulses with energy of ~4μJ were collimated by a CaF2 lens. After collimation, the few-cycle mid-IR pulses were split into an intense fundamental and weak perturbation pulse using two pairs of uncoated CaF2 wedges as beamsplitters. The transmitted and reflected pulses, containing 85% and 0.1% of the incident pulse energy, served as the fundamental and perturbation arms, respectively. The second wedge pair was also used to collinearly combine the mid-IR perturbation pulse with the second-harmonic pulse with a fixed time delay, as discussed in the following. In the mid-IR perturbation arm, an additional wedge pair was used to balance the dispersion between the fundamental and signal arms and to vary the CEP of the perturbation pulse. The fundamental and perturbation beams were finally focused by a cylindrical concave mirror (f = 50 mm for single-shot measurements and f = 100 mm for integrated measurements) onto an 8-bit complementary metal–oxide–semiconductor image sensor (Thorlabs DCC1545M), which serves as both the nonlinear medium and the detector. The intensity of the fundamental pulse was ~10 GW cm−2. The two beams overlapped on the detector with a crossing angle of ~3.5°, allowing the time delay τ to be mapped onto the transverse spatial coordinate of the sensor. By varying the position of the translational stage installed in the fundamental arm, the transverse position of the maximum of the modulation waveform can be varied and, in this way, the time delay can be calibrated as shown in Supplementary Fig. 3. A 1-mm-thick anti-reflection coated silicon window was placed in front of the detector, both to block the visible light and to compensate for the dispersion of the CaF2 wedges in the set-up. After passing through the silicon plate, a pulse duration of 24.0 fs was obtained, as shown in Fig. 2. After transmitting through the dichroic mirror, the second-harmonic pulses were reflected by a periscope to set their polarization parallel to that of the mid-IR and second-harmonic pulses overlapped on the detector with a crossing angle of ~3.5°, allowing the normalization procedure to obtain the electric-field waveform. The modulated detector signal, emerging from the combined influence of the fundamental pulse E0 and perturbation pulse EP, can be approximated (to first order in the perturbation) as S ∝ w(E0 + EP) ≈ w(E0) + (w(E1) − w(E0)) EP (ref. 17), where w(E) is the excitation rate. It is then possible to extract the field of the perturbation pulse from the difference between the detector signals with and without the perturbation field.

provided that the scaling factor w is known. While in the scanning geometry, the excitation occurs only near the peak of the fundamental pulse, the scaling factor (w(E1) − w(E0)) is constant, the single-shot measurements here are sampled over a region of the spatial profile of the fundamental pulse, and therefore the scaling factor varies across the beam profile due to the nonlinearity of the excitation rate with field strength. However, the variations induced due to non-uniformity of the fundamental pulse beam profile can be easily removed using a simple normalization procedure.

For excitation in both the multiphoton and tunnelling regimes, the excitation rate can be approximated as w ∝ I, where I is the laser intensity and q is the effective multiphoton scaling parameter. Then, w = w0Iq, which is proportional to I1−q/2. As described above, the perturbing field can be obtained from EP = (w(E1) − w(E0)) EP, which simplifies to EP = w0 Iz, where n = 2Nlq−1 and Sz is the detector signal arising from the fundamental pulse only. Because q can be easily measured from the dependence of the detector signal on the fundamental laser intensity (Supplementary Fig. 4), the normalization procedure can be straightforwardly applied to obtain the perturbation field, even when the fundamental beam profile varies. The normalization procedure is demonstrated in Supplementary Fig. 2, and further details of the derivation of the normalization factor are described in Supplementary Note 1 and Supplementary Figs. 2 and 3. The normalization procedure described above requires successive measurements to be made with and without the perturbation pulse, and therefore imposes limits on the shot-to-shot stability of the laser pulse. However, this requirement can be relaxed, because, in general, the experimental geometry can be chosen such that the fundamental beam profile varies on spatial scales that are larger than those associated with the perturbation-field oscillations. In this case, the measurement of Sz can be replaced by a Fourier spectral filter of the perturbed signal S to isolate the fast oscillations associated with the perturbation pulse from the slowly varying background associated with the fundamental pulse.

Data availability
The data that supports the plots within this paper and other findings of this study are available at https://stars.library.ucf.edu/cgi/preview.cgi?article=1001&context=datasets.

Code availability
The codes that produced the modelled data within this paper and other findings of this study are available at https://stars.library.ucf.edu/cgi/preview.cgi?article=1001&context=datasets.

References
27. Thür, N. et al. 4-W, 100-kHz, few-cycle mid-infrared source with sub-100-nrad carrier-envelope phase noise. Opt. Express 25, 1505–1514 (2017).

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Author contributions
M.C. had the idea for the single-shot waveform measurement scheme and oversaw the research team. Y.L. led the experimental effort and performed most of the measurements and simulations. J.E.B. assisted with the measurements of the carrier-envelope phase dependence. J.N. and S.G.-M. assisted with the construction of the experimental set-up and with the data collection. All authors contributed to data analysis and the creation of the manuscript.

Competing interests
The authors declare no competing interests.

Additional information

Selfies from the lab: https://www.youtube.com/watch?v=VQ4ZdDFJ0fQ

Supplementary information
This material is available at www.nature.com/reprints.

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