Vanishing carrier-envelope-phase-sensitive response in optical-field photoemission from plasmonic nanoantennas

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At the surfaces of nanostructures, enhanced electric fields can drive optical-field photoemission and thereby generate and control electrical currents at frequencies exceeding 100 THz (refs. 1,2). A hallmark of such optical-field photoemission is the sensitivity of the total emitted current to the carrier-envelope phase (CEP)1,2,3,5,7. Here, we examine CEP-sensitive photoemission from plasmonic gold nanoantennas excited with few-cycle optical pulses. At a critical pulse energy, which we call a vanishing point, we observe a pronounced dip in the magnitude of the CEP-sensitive photocurrent accompanied by a sudden shift of π radians in the photocurrent phase. Analysis shows that this vanishing behaviour arises due to competition between sub-optical-cycle electron emission events from neighbouring optical half-cycles and that both the dip and phase shift are highly sensitive to the precise shape of the driving optical waveform at the surface of the emitter. As the mechanisms underlying the dip and phase shift are a general consequence of nonlinear, field-driven photoemission, they may be used to probe sub-optical-cycle emission processes from solid-state emitters, atoms and molecules. Improved understanding of these CEP-sensitive photocurrent features will be critical to the development of optical-field-driven photocathodes for time-domain metrology and microscopy applications demanding attosecond temporal and nanometre spatial resolution.

Our experimental set-up is shown in Fig. 1a. In our experiments, we illuminated an array of triangular, gold nanoantennas (nano-triangles) located on a conductive indium tin oxide (ITO) layer with pulses of near-infrared light having a central wavelength of 1,177 nm and a duration of 10 fs (that is, 2.5 optical cycles full-width at half-maximum, FWHM). The incident laser pulses are polarized along the altitudes of the triangular nanoantennas and drive electron emission. Field-enhancement factors of ≈30x are achieved near the apices of the triangles due to both plasmonic resonance and geometric enhancement effects1,2,5; when illuminated, the triangular nanoantennas predominantly emit electrons from their apices due to these localized, enhanced fields5. Following emission, a d.c. bias voltage sweeps the electrons from the antennas, through the interstitial air and across an insulating gap of ≈5 μm to a second ITO pad serving as a collector. Such a short gap size reduces the likelihood of electron capture and removes the need for the samples to be housed in vacuum. Optical and scanning-electron micrographs of the emitter electrode, gap, collector electrode and emitter structures are shown in Fig. 1b. (See Methods or ref. 1 for further experimental set-up details.)

With sufficiently high fields at the emitter apices, the tips are driven into the optical-field photoemission regime where the emission can be approximated by a quasi-static tunnelling rate1,2. In Fig. 1c,d, we show the emitted electron current, calculated using a quasi-static Fowler–Nordheim (FN) emission model, which has proven effective at predicting optical-field-driven photocurrents from gold nanotips and nanoantennas1–4 (see Methods for further details). For the calculation we use a 10 fs laser pulse with a central wavelength of λ = 1,177 nm and a peak field of E0 = 15 GV m−1. The work function of the nanoantennas was taken to be ϕ = 5.1 eV (we use this value of the work function for the remainder of this Letter). For FN tunnelling emission, current is emitted in sub-cycle bursts during half-cycles where the field is negative. Emission is suppressed during half-cycles where the field is positive as the surface electrons experience a force that drives them further into the emitter. As it will be convenient for later analysis, in Fig. 1c,d we have shaded and numbered half-cycle regions of the pulse (even half-cycles are shaded light red and odd ones are shaded light blue). From Fig. 1c,d, it is apparent that the CEP of the driving laser pulse, ϕcep, has a strong effect on the emission. As ϕcep is changed from 0 (Fig. 1c) to π (Fig. 1d), the total emitted charge per pulse switches from being dominated by even half-cycle contributions (Fig. 1c) to being dominated by odd half-cycle contributions (Fig. 1d). (Note that the half-cycle regions are defined relative to the centre of the intensity envelope, not the underlying field waveform, and thus do not move when the CEP is shifted.)

To better quantify the impact of ϕcep on the measured photocurrent, we need to define the key observables for both our measurements and subsequent analysis. In Fig. 1e we plot the integrated total photoemission current Itot(ϕcep) (see Methods for calculation details). Note that Itot has been normalized in the plot. Using harmonic analysis, the integrated total photoemission current can be written as Itot(ϕcep) = ∑n |In|cos(nϕcep + θn) where In is the complex amplitude of the nth harmonic of the CEP-dependent total photoemission current. In Fig. 1f we plot |In| for n = 0–4 on a
Fig. 1 | Experimental set-up and illustration of CEP sensitivity. a, Illustration showing optical excitation and charge extraction from the nanoantenna array in the experimental set-up. The photocurrent is pulled by a bias voltage, $V_C = 30\, \text{V}$, from the nanoantenna array into an isolated collector electrode, where it is then detected using a transimpedance amplifier. b, Optical microscope image of the emitter array and collector region (left) and scanning electron micrograph of the triangular gold nanoantennas used in the experiment (right). ROC, radius of curvature. c, Simulated waveform (red curve) and the calculated electron emission rate (filled blue curves) as a function of time with $\psi_{\text{cep}}=0$. Half-cycle numbers are labelled across the bottom. Note that the half-cycle regions are determined relative to the centre of the pulse intensity envelope, not the field waveform (that is, they do not depend on $\phi_{\text{cep}}$). d, Same as in c, but with $\psi_{\text{cep}}=\pi$. e, Plot of the total emitted current as a function of $\psi_{\text{cep}}$. The average total current $I_n$ is labelled. The magnitude of the CEP sensitivity was found to be $|I_n|/I_0 = 0.004$. f, Magnitudes of the harmonic components of the CEP-sensitive current $I_n$ for $n=0$–4.

logarithmic scale. We note that $I_n(\psi_{\text{cep}})$ is dominated by the CEP-independent average total photocurrent $I_0$ and the first harmonic $I_1$, that is, the fundamental CEP-dependent sinusoidal component of the photocurrent. We also define the complex ratio $I_n/I_0$ as the CEP sensitivity.

For experimental characterization, we phase-stabilized our optical source with a fixed carrier-envelope offset frequency $f_{\text{coo}} = 100\, \text{Hz}$ such that $\psi_{\text{cep}} = 2\pi f_{\text{coo}} t$, and used lock-in detection to characterize $I_n$. (see Methods and refs. 1,22 for further details). We then monitored $I_n$ while scanning through a range of incident pulse energies; the experimental results are plotted in Fig. 2 (orange circles). Intuitively, it would seem that at higher intensities, when the overall photoemission current is larger and when we are deeper into the optical-field photoemission regime, that we would observe a larger CEP-sensitive photocurrent. However, we find a striking behaviour in the experimental data where at a critical pulse energy of $\approx 0.1\, \text{nJ}$ we observe a dramatic dip in $|I_n|$ and a corresponding phase-shift of $\pi$ radians in $\angle I_n$. We refer to this critical pulse energy as a vanishing point; the behaviour at this vanishing point was measured multiple times at various locations on the sample surface over several days of measurement (Supplementary Fig. 2 and Supplementary Section III). We note that the observed dip in amplitude and the phase shift are reminiscent of the characteristics of an antiresonance. (The antiresonance analogy holds in some respects beyond just the appearance of the data; however, the observed dip and phase shift appear with changes in driving pulse energy, not frequency, on the horizontal axis and are not representative of an antiresonance in a strict sense.)

Overlaid on our experimental results in Fig. 2 are the predicted CEP-dependent behaviours from our quasi-static tunnelling model (shown as a blue dashed line). Note that for the model results in Fig. 2, we have accounted for the effects of the plasmonic resonance of the nanoantennas using a damped harmonic oscillator model and spatial averaging over the beam spot (see Methods for calculation details). From Fig. 2, we see that the quasi-static model shows excellent agreement with the experimental data in both magnitude and phase, especially at high intensities where the photoemission is deep in the optical-field-emission regime. When modelling the CEP-sensitive photocurrent response, we used a plasmonic resonance wavelength of $\lambda_{\text{res}} \approx 1.105\, \text{nm}$ and a field enhancement of 32.6; these values were extracted from extinction and photocurrent scaling measurements on the nanoantenna arrays (see Methods and Supplementary Sections IV–VI). Extinction measurements indicated a plasmonic damping time of $\tau = 6.4–6.9\, \text{fs}$ in our model, we used $\tau = 6.8\, \text{fs}$. We should note that changes in the modelled damping time of just 100 as lead to visually apparent changes in the shape of $|I_n|$ and $\angle I_n$ as a function of intensity near the vanishing point (Supplementary Fig. 5). The discrepancy between our model results and the experimental data is most pronounced at low intensities; this is not unexpected because, at these intensities, the emission approaches the multiphoton regime, and thus the quasi-static FN model provides a poorer estimate of the total current response.

The simulation results shown in Fig. 2 accurately account for the experimental behaviours and give us confidence that our modelling of optical-field photoemission as a quasi-static tunnelling process is reasonable. However, the simulation results provide little insight into the physical origin of the dip and phase shift in the CEP-dependent photocurrent. To get a sense for these origins, we now turn to a simplified version of the model presented in Fig. 2. In this simplified model, we ignored the filtering of the incident pulse by the plasmonic response of the nanoantennas as well as the focal spot averaging. We simply used a transform-limited pulse with similar duration (10 fs FWHM) and central wavelength (1.177 nm) to that used in the experiment and considered emission from a single nanoantenna. The results are presented in Fig. 3.

In Fig. 3 we examine the properties of the electron emission as a function of intensity assuming emission only from the light red and light blue shaded regions of the optical pulse shown in the top left insets of Fig. 3a–c. The top panels of Fig. 3a–c show $|I_n|$
and the bottom panels show $\angle I_\phi$ as a function of optical intensity. Note that the shaded regions encompass an increasing number of half-cycles moving from the centre of the pulse out to the wings: in Fig. 3a only one half-cycle is shaded (we consider emission from only the centre half-cycle, that is, half-cycle 0), in Fig. 3b three are shaded (we consider emission from half-cycles 0 and ±1) and in Fig. 3c nine are shaded (we consider emission from half-cycles −4 to +4). As with Fig. 1cd, we shade even half-cycles in light red and odd half-cycles in light blue. Additionally, in each plot of $\angle I_\phi$, there is a light red dashed line ‘even’ showing the phase response of the sum of the even half-cycle contributions to $I_\phi$ (that is, contributions from half-cycles 0, ±2, ±4, …), and a light blue dashed line called ‘odd’ showing the phase response of the sum of the odd half-cycle contributions to $I_\phi$ (that is, contributions from half-cycles ±1, ±3, ±5, …). Note that the sum of the even half-cycle contributions always peaks at $\varphi_{cep} = 0$, while the sum of the odd half-cycle contributions always peaks at $\varphi_{cep} = ±\pi$.

In Fig. 3d–f we analyse the emission from half-cycle 0 at the intensity labelled by the red dot in Fig. 3a. Figure 3d shows the normalized emitted photocurrent (red) and half-cycle 0 of the driving electric field waveform (black curve) for $\varphi_{cep} = 0$. Figure 3e similarly shows the normalized emitted photocurrent and driving electric field for $\varphi_{cep} = \pi$. Note that because tunneling requires a negative field, a large photocurrent pulse appears for $\varphi_{cep} = 0$, while no photocurrent appears for $\varphi_{cep} = \pi$. In Fig. 3f, we plot $I_{cep}(\varphi_{cep})$ with a red dashed line, and we plot $I_0 + |I_\phi|$ cos($\varphi_{cep} + \angle I_\phi$) with a red solid line. From Fig. 3f, we see that the emitted current is maximized at $\varphi_{cep} = 0$ and minimized at $\varphi_{cep} = \pi$ (where there is no emission).

In Fig. 3g–i we similarly analyse emission from half-cycles 0 and ±1. However, in Fig. 3g,h we show the photocurrent emission at two intensities: in red, we show the photocurrent from the intensity labelled (i), and indicated with a red dot in Fig. 3b, and in blue, we display the photocurrent at the intensity labelled (ii) and shown with a blue dot in Fig. 3b. Figure 3g shows the normalized emitted photocurrents (red and blue) and half-cycles 0 and ±1 of the driving electric field waveform (black) for $\varphi_{cep} = 0$. In Fig. 3g the photocurrents plotted in red and blue are multiplied by normalization constants $C_{red}$ and $C_{blue}$, respectively, such that their maximum values are 1. Figure 3h shows the emitted photocurrents (red and blue) and half-cycles 0 and ±1 of the driving electric field waveform (black) for $\varphi_{cep} = \pi$. In Fig. 3h, the same normalization constants as used in Fig. 3g ($C_{cep}$ and $C_{cep}^{\pi}$) are used; that is, the photocurrents are normalized relative to those plotted in Fig. 3g.

Consider the peak values of the photocurrents in Fig. 3h. The dashed line in Fig. 3h denotes 0.5. The photocurrents plotted in red lie just below this line, and those in blue lie just above it. Therefore, if we sum the photocurrents in red from the ±1 half-cycles, we expect that this total current will be smaller than that from half-cycle 0. Similarly, if we sum the photocurrents in blue from the ±1 half-cycles, we expect that this current will be larger than that from half-cycle 0. In other words, we expect that at intensity (i), $I_{cep}(\varphi_{cep})$ will be maximized at $\varphi_{cep} = 0$, while at intensity (ii), $I_{cep}(\varphi_{cep})$ will be maximized at $\varphi_{cep} = \pi$. Moreover, between intensities (i) and (iii), we anticipate an intensity where the sum of the photocurrents from the ±1 half-cycles and the photocurrents from half-cycle 0 are about equal. Here, we expect $I_{cep}(\varphi_{cep} = 0) \approx I_{cep}(\varphi_{cep} = \pi)$, and we therefore expect that the first harmonic component of $I_{cep}(\varphi_{cep})$ will approximately vanish (that is, $I_\phi = 0$). This intensity is the vanishing point, and we label it as intensity (ii) and mark it with a purple circle in Fig. 3b.

In Fig. 3i, we plot $I_{cep}(\varphi_{cep})$ for intensities (i), (ii) and (iii) with red, purple and blue dashed lines, respectively, and we overlay plots of $I_0 + |I_\phi|$ cos($\varphi_{cep} + \angle I_\phi$) for these three intensities with solid lines.
of the same colour. Note that in Fig. 3i we observe the exact behaviours described in the preceding paragraph: moving from intensity (i) to (ii) to (iii), $I_0(\varphi_{op})$ transitions from being peaked at $\varphi_{op}=0$, to having a minimal first harmonic, to being peaked at $\varphi_{op}=\pi$. We should also note that these behaviours are consequences of the nonlinear character of the field-driven photoemission. In the inset of Fig. 3b, we display the emitted charge, that is, the integrated photocurrent, from an individual half-cycle of varying intensity. The intensities of the 0 and ±1 half-cycles at intensity (i) are labelled with red dots, and the intensities of the 0 and ±1 half-cycles at intensity (iii) are shown with blue dots. Additionally, the slope of the emission curve near the red dots is shown with a red dashed line and near the blue dots with a blue dashed line. Note that, due to the nonlinearity of the emission, the slope decreases at higher intensities. This decrease in the emission rate growth means that the blue dots are closer together than the red dots. In other words, at higher intensities, the emission from the ±1 half-cycles catches up to the emission from half-cycle 0. Indeed, in the inset we show the ratio of the charge emitted from one of the ±1 half-cycles to that emitted from half-cycle 0 as $Q_1/Q_0$, and we see that this ratio increases from intensity (i) to (iii). This nonlinear, ‘catching up’ behaviour underlies the shift in $I_0(\varphi_{op})$ from intensity (i) to (iii) and the vanishing point behaviour.

Figure 3c,j–l parallels Fig. 3b,g–i while considering emission from half-cycles −4 to +4. Interestingly, in Fig. 3c, when more half-cycles are included, it is possible to observe multiple vanishing points. Although the origins of the vanishing points in Fig. 3c are not as readily apparent as in Fig. 3b, close inspection reveals that the vanishing point labelled (ii) in Fig. 3c results from the emitted current from the ±4 half-cycles catching up to that from the ±3 half-cycles. Labelling the charge emitted from half-cycle ±n as $Q_n$, we find that at intensity (i) in Fig. 3c, $Q_0+Q_2<Q_2+Q_0$ and $Q_0+2Q_2+Q_2<2Q_2+Q_0$, while at intensity (iii) in Fig. 3c, $Q_0<2Q_2+Q_0$, and $Q_0+2Q_2+3Q_2>2Q_2+Q_0$. In other words, due to the reduction in emission rate growth from the ±3 half-cycles, there is a relative increase in the emission contribution from the ±4 half-cycles that enables the cumulative emission from the even half-cycles to overtake that of the odd ones. (The factor of two in front of $Q_n$ for $n \neq 0$ derives from the symmetric pairs of half-cycles about half-cycle 0.) The vanishing point at the lower intensity similarly arises from the emitted current from the ±3 half-cycles catching up to that from the ±2 half-cycles. In line with the preceding discussion, as successive half-cycles are included, there remains to be competition between even and odd half-cycle contributions to the CEP-sensitive photocurrent, until the optical field strength, and hence the emitted current, of the higher-numbered half-cycle regions weakens significantly, at which point $I_0$ converges (that is, $I_0$ stops changing significantly as more half-cycles are included in the calculation). For the intensity range shown and the pulse duration used in the model, $I_0$ converges after summing over half-cycles −4 to +4 (Fig. 3c,j–l).

At each vanishing point in Fig. 3a–c, the phase jumps instantaneously between 0 and ±$\pi$. However, in the experimental and simulation results in Fig. 2, which use realistic pulses with residual chirp and plasmonic damping, the phase response near the vanishing point is smoother with a slight downward slope. Initial modelling has indicated that the slope of $\angle I_0$ near the vanishing point is a function of the chirp and asymmetry in the pulse shape. By examining the CEP-sensitive response in detail, including the slope of $\angle I_0$ and the precise location and depth of the dip in $I_0$, one can likely obtain a great deal of information about the time-domain profile of the optical waveform at the surface of the emitter. (See a description of the sensitivity of vanishing points to changes in pulse dispersion in Supplementary Fig. 9 and Supplementary Section VIII.) Indeed, the sensitivity of the dips and phase shifts to subtle changes in the optical electric field allowed us to confirm the field enhancement at the apices of the nanoantennas and to refine our characterization of the damping time of the nanoantennas’ plasmonic response.

The appearance of vanishing points in the CEP-sensitive current response is a general phenomenon that should be observable for any system where the current is field-driven with similar nonlinear characteristics to that of FN tunnelling (Supplementary Section VII). Further experimental and theoretical analyses of vanishing point behaviours have the potential to probe the physics of strong-field electron emission dynamics in solids as well as gases, to better characterize the precise waveforms driving optical-field photoemission, and to sense subtle changes in the electronic structure of surfaces.

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Author contributions
P.D.K. and W.P.P. conceived the experiment. W.P.P., R.G.H. and Y.Y. fabricated the devices. P.D.K., W.P.P. and P.V. performed the measurements, collected the experimental data and performed the numerical analysis. P.D.K., W.P.P. and P.V. composed the manuscript. P.D.K., W.P.P., P.V., R.G.H., Y.Y., K.K.B. and F.X.K. interpreted the results and contributed to the final manuscript.

Competing interests
The authors declare no competing interests.

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Methods

Few-cycle optical source. A supercontinuum-based fibre laser source with a repetition rate of 78.4 MHz, central wavelength of 1,177 nm and a duration of 10 fs (that is, ≈25 optical cycles FWHM) was used. For the experimental characterization of $I_c$, the carrier-envelope-offset frequency $f_{cep}$ of our incident pulse train was locked to a stable reference at 100 Hz. In this locked state, the 9th pulse in the train had a CEP phase of $\phi_{cep}(n) = 2\pi f_{cep} n + \phi_0$, where $f_{cep}$ is the repetition rate of the incident pulse train and $\phi_0$ is an absolute phase offset. We have characterized the CEP noise of our system to be $\pm 157$ mrad root-mean-square (r.m.s.) when integrated from 5 mHz to 5 MHz (ref. 1). For further details regarding the optical source and phase stabilization methods used in this work, see ref. 21.

Nanooptics layout and fabrication. The nano-triangles were arranged in a square grid (20 nm × 20 μm) with a nominal spacing of 480 nm. Each nano-triangle in the array was nominally isosceles in shape, with a target base width of 180 nm, altitude of 240 nm and thickness of 20 nm. The triangular shape of the antennas was chosen to break the system's inversion symmetry, thereby increasing the CEP sensitivity of the emitted photocurrent when driven into the regime of optical-field photoemission. Each nano-triangle had a radius of curvature at its apex of ≈10 nm.

The nano-triangle array and its supporting electrodes were fabricated in two main steps. First, the nano-triangle array was patterned onto an ITO-coated sapphire substrate via electron-beam lithography, then the ITO-coated substrate was patterned via optical lithography, and the ITO in designated areas was etched away to create the emitter and collector electrode structures. The details of each fabrication step are thoroughly described in the supplementary information of ref. 1.

Quasi-static tunnelling models. In our FN quasi-static tunnelling model, we express the field at the tip surface in the form $F(t) = \Re (F_0 \exp(\i\omega t + \phi_{cep}))$, where $F_0$ is the peak electric field strength, $\mathcal{F}(t)$ is the normalized complex amplitude envelope of the incident pulse and $g(\omega)$ is the transfer function due to the plasmonic nanostructure. We approximate the plasmonic nanostructure as a damped harmonic oscillator $e^{-\omega_\text{c nap}/(t)}$ and use

$$g(\omega) = \frac{1}{\omega_\text{c nap}^2 - \omega^2 - i \omega/\tau}$$

where $\omega_\text{c nap} = 2\pi c / \lambda_\text{nap}$ is the resonance frequency of the plasmonic nanoantenna and $c$ is the speed of light in vacuum. (The damped harmonic oscillator model is further described in the supplementary information of ref. 1.)

The field $F(t)$ modulates the surface potential barrier and, if sufficiently strong, it can pull the barrier down so far as to enable significant electron tunnelling from the nanoantenna and into the surrounding air or vacuum before reversing polarity. We used the FN expression

$$I(t) \propto F(t) \Theta(-F(t)) \exp\left[ \frac{2eV}{3kT}\right]$$

to account for the quasi-static tunnelling rate, where $\Theta(x)$ is the Heaviside step function (this function ensures we only consider emission when $F(t) < 0$), $m$ is the electron mass, $\phi$ is the material work function, $\hbar$ is the normalized Planck’s constant and $e$ is the electron charge. We integrated $I(t)$ over the entire pulse duration to determine the emitted charge per pulse $Q$, which is related to the integrated total photoemission current by the repetition rate of the laser: $I_\text{c} = Q f_{cep}$. To arrive at the model results presented in Fig. 2, we started with a cos2-shaped optical pulse. This model pulse was derived from a fit to the time-domain measurement of the experimental pulse that was characterized via two-dimensional spectral shearing interferometry (2DSI)13. The expected field profile at the nanoantennas' apices was then approximated by filtering this model pulse with the harmonic oscillator resonator response $g(\omega)$ and setting $F_0 = F_{cep} \times F_E$, where $F_E$ is the peak incident field strength of the laser pulse and FE is the field-enhancement factor of the nanoantenna.

FE was approximated by fitting the quasi-static FN model of the total average photocurrent $I_c$ to measurements of $I_c$ over a range of incident intensities in the optical-field emission regime12. Using the measured spot size and pulse energy, $F_{cep}$ was calculated. The following iterative process was then used to fit FE: (1) The FN model was used for the entire dataset to obtain a first estimate for the field enhancement; (2) only those enhanced intensity values where $\gamma < 1$ were used to obtain the next field enhancement factor; (3) step 2 was repeated until the process converged to the final field enhancement factor value1. In Supplementary Fig. 4, we show the fit results for a single scan. Supplementary Table 1 compiles FE factors from nine separate scans for reference. The resultant FE fits ranged from 28.9 to 35.5 with an average FE of (32.6 ± 2.4)× (Supplementary Table 1, Supplementary Section 4 and Supplementary Section V). For the model results shown in Fig. 2, the FE was taken to be 32.6× to match the average measured field enhancement.

The damping time $\tau$ and resonant wavelength $\lambda_\text{nap}$ followed from fits to measurements of the nanoantennas' extinction spectra (Supplementary Fig. 3 and Supplementary Section IV). Fits to the nanoantennas' extinction spectra between 700 and 1,400 nm yielded $\lambda_\text{nap} = 1,105$ nm and $\tau$ values ranging from 6.4 to 6.9 fs (Supplementary Fig. 3; for further discussion see Supplementary Section IV). The final value of $\tau = 6.8$ fs was used to create the model results in Fig. 2 as was chosen as it provided the best fit to the experimental data.

To account for the fact that each nano-triangle was exposed to a different peak intensity due to tight focusing, we integrated the calculated electron emission from an array of nano-triangle apices over the focal spot of the incident beam. Using this integrated emission, we calculated the expected CEP sensitivity, that is, $I_c / \lambda_\text{nap}$, Multiplying this sensitivity by our measurement of $I_c$, we found the CEP-dependent current shown by the dashed blue line in Fig. 2. We note that, although recent results show that tightly focused, broadband optical pulses exhibit strong phase shifts in excess of the predicted Guoy phase near the region of the focus16, this effect can be safely ignored here as the emitters are only 20 nm tall and were located near the minimum beam waist where the CEP does not exhibit transverse dependence.

Measurements of CEP-sensitive photocurrent. The way in which the CEP-sensitive current was detected and analysed in this work using a slowly oscillating $\phi_{cep}$ and lock-in detection of $I_c$ is of critical importance. Photocurrents were first amplified and converted to a voltage signal using a transimpedance amplifier. Using lock-in detection, we measured the magnitude and phase of this amplified voltage signal oscillating at $f_{cep}$; this magnitude and phase corresponds to $|I_c|$ and $\angle I_c$, respectively. We verified that the measured $I_c$ signal was indeed controlled by the CEP of the incident pulse by translating a barium-fluoride wedge through the beam and verifying the expected phase shift imparted on $\angle I_c$ as a function of the wedge insertion (see results in Supplementary Fig. 1 and Supplementary Section I).

A variable neutral density filter was then used to alter the incident pulse energy while monitoring $I_c$. We note that, during the intensity scans, we observed a fluctuation in measurements of $|I_c|$ of less than 20% r.m.s., a fluctuation of $\angle I_c$ of less than 180 mrad r.m.s. and a noise floor for the current measurements of ~20–40 fA. Additionally, the magnitude of the CEP sensitivity (away from the vanishing point) was on the order of 5 × 10−10.

Data availability

The data that support the plots within this paper and other findings of this study are included in this article and its Supplementary Information and are available from the corresponding authors upon reasonable request.

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