Spatio-temporal sampling of near-petahertz vortex fields

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Measuring the field of visible light with high spatial resolution has been challenging, as many established methods only detect a focus-averaged signal. Here, we introduce a near-field method for optical field sampling that overcomes that limitation by employing the localization of the enhanced near-field of a nanometric needle tip. A probe field perturbs the photoemission from the tip, which is induced by a pump pulse, generating a field-dependent current modulation that can easily be captured with our electronic detection scheme. The approach provides reliable characterization of near-petahertz fields. We show that not only the spiral wave-front of visible femtosecond light pulses carrying orbital angular momentum (OAM) can be resolved, but also the field evolution with time in the focal plane. Additionally, our method is polarization sensitive, which makes it applicable to vectorial field reconstruction.

I. INTRODUCTION

The precise knowledge of the electro-magnetic field oscillations of light is not only the backbone of ultrafast science [1, 2], but the indispensable prerequisite for many applications such as time-domain terahertz spectroscopy [3–5] and field-resolved mid-infrared spectroscopy [6, 7]. Common techniques for field sampling reaching from the near-infrared to the visible spectral region include attosecond streaking [8–14], electro-optic sampling [15], femtosecond streaking [16], non-linear photoconductive sampling [17], or the tunneling ionization with a perturbation for the time-domain observation of an electric field (Tiptoe) [18–25]. While the sampling of electric field waveforms in the time domain is well established, its simultaneous spatial characterization has remained challenging. Whereas for terahertz radiation, where the wavelength is much longer than for visible light, sub-focal size resolution can be achieved, for instance, by using cameras [26] or small sensors [27], spatially resolved field measurements in the focus of a visible light beam are challenging as the typical focal size is of the order of a few micrometers. Here, a sub-micrometer probe is necessary.

Furthermore, measuring the field of an optical vortex beam brings additional difficulties related to the spatial phase structure in the focus. Because of its helical phase, the field has a π phase-shift at opposite sides of the OAM mode [28, 29]. Thus, for measurement techniques that are only sensitive to the focal averaged field, a complete cancellation of the signal can be expected. Consequently, the field-resolved measurement of vortex beams requires sub-focal spatial sampling. In attosecond streaking experiments, an extreme ultraviolet (XUV) pulse is used to sample the electric field of a co-propagating (near-petahertz) field. Here, the XUV-focal size is much smaller than the sampling beam focal size, so the spatio-temporal characterization may be possible. Such measurements, however, require complex vacuum setups, electron spectroscopy, and would be experimentally demanding. It is thus not surprising that the use of attosecond streaking for the spatial reconstruction of near-petahertz vortex fields has not been reported yet. The measurement of vortex fields has mostly been limited to the measurement of the corresponding OAM carried by the light [29–31], not the field itself.

To date, approaches using near-field methods to achieve spatial resolution could either not resolve the electric field of light itself [33–36], were limited to much lower frequency ranges in the THz regime [4, 5], or detected a spatially averaged signal from several sensors [23, 25]. We note, that the work presented in Ref. [25] has demonstrated a measurement of spatio-temporally coupled laser pulses. Here, the frequencies were limited to the infrared range and spatial resolution only achieved in one dimension (limited by the pixel size to (5.2 µm)). Here, we overcome these limitations by employing a single nanometric needle tip as a localized probe for the near-field sampling of femtosecond light fields. The approach, termed nanoTiptoe and illustrated in Fig. 1a), inherits the method for sampling the
FIG. 1. The nanoTiptoe approach for spatio-temporal field sampling: a) Experimental setup: The pump pulse (red line) and signal pulse (black line) were focused onto a tungsten tip with an off-axis parabola (OAP). The photocurrent generated by the emitted electrons was trans-impedance amplified and lock-in detected. b) Finite-difference time-domain simulations of the field around an exemplary tungsten tip with 10.5° half-opening angle and a radius of r = 15 nm have shown that the field is enhanced by a factor of around 5, in agreement with literature values [32]. The arrows indicate the near-field polarization. c) Shadowgraphy image obtained by scanning the tip across the x, y-plane while detecting the transmitted intensity. The opening angle in the experiment was determined to be (21 ± 2)°. d) Detected ionization current as a function of position. The signal in region A corresponds to the ionization at the apex, whereas the signal in region B is due to a small contribution from ionization near the rear-end of the needle shank, which can be spatially discriminated.

The electric field from Tiptoe[18] and achieves high spatial resolution from field localization at the nanometric needle tip.

In nanoTiptoe, a few-cycle pump pulse drives electron emission in the tunneling regime that depends nonlinearly on the electric field. Due to this nonlinearity and a short pump pulse, the photoemission is limited to the strongest half-cycle of the laser pulse, and suppressed otherwise. We note that only electric field vectors pointing into the surface cause photoemission from the nanometric needle tip[37]. Similar to Tiptoe, the emission burst during the strongest half-cycle opens a sub-cycle temporal gate that is perturbed linearly with the signal pulse [18], enabling characterization of its field. We measured the resulting photoemission current from the needle tip after transimpedance amplification employing lock-in-detection. Importantly, the fields driving photoemission were the locally enhanced near-fields, which were strongest near the apex of the tip, cf. Fig. 1b). The current measurement approach makes complex ultra-high-vacuum-based time-of-flight spectroscopy [34, 38] obsolete, which is a major advance in simplifying such measurements.

II. EXPERIMENTAL DETAILS

The output of a commercial 10 kHz Ti:Sa chirped pulse amplifier is broadened in a hollow-core fiber to an octave spanning spectrum ranging from 500 nm to 1000 nm, with a central wavelength of 750 nm. The pulses are then compressed to a duration of around 4.2 fs using chirped mirrors (UFI PC70). The laser beam was actively stabilized in angle and position. The pulses were split into a strong pump pulse and a weak signal pulse in a Mach-Zehnder interferometer (not shown in Fig. 1a)), where the signal pulse is chopped at half the repetition rate. To facilitate the precise control of the delay between signal and pump pulses, the pump arm is provided with a retro-reflector mounted on a closed-loop piezo-stage (MCL OPM100) with 100 µm travel range. The beams were focused with variable temporal delay onto a nanometric tungsten needle tip inside a vacuum chamber (2 × 10⁻³ mbar) using an off-axis parabolic mirror (OAP, f = 101.6 mm). For the shadow image in Fig. 1c), we used f = 25.4 mm to get a sharper contour. The needle was directly soldered to a BNC pin, which was mounted onto a 3D closed-loop piezo stick-slip stage. The photocurrent is amplified by 10⁹ V/A using a low-noise high-gain transimpedance amplifier (FEMTO
DLPCA-200) and detected using a lock-in amplifier (Zürich Instruments HF2LI). The upper cut-off frequency ($f_{\text{−3dB}}$) of the transimpedance amplifier is 1.1 kHz, which is below the repetition rate of our laser of 10 kHz and 5 kHz for pump and signal beam, respectively. The expected damping of the signal can be estimated from the measured amplifier response curve provided by the manufacturer to -20 dBV and -14 dBV for 10 and 5 kHz, respectively. These values correspond to a damping factor of the voltage signal of 10 and 5, in that order. Whenever we estimated a number of emitted electrons, we took these factors into account. For the lock-in detection, we used a demodulation bandwidth of 1.459 Hz ($f_{\text{−3dB}}$). From a noise reference measurement with blocked laser beams, we calculated the normalized noise density to $4.8 \mu V/\sqrt{\text{Hz}}$ and $4.2 \mu V/\sqrt{\text{Hz}}$ for the 5- and 10 kHz components, respectively. This is close to the specified value of $4.3 \text{fA/V}\sqrt{\text{Hz}}$ noise current at our amplification of $10^9 \text{V/A}$. In order to be detectable, the minimum current modulation thus has to be larger than the noise current times the damping, that is: $e \cdot n_e / s > 4.8 \text{fA} \times 5$. Here, $e$ is the elementary charge, and $n_e$ the number of electrons. This corresponds to a current modulation of at least 30 electrons per shot, assuming a 1 Hz demodulation bandwidth for illustration purposes. In the high-gain mode of the amplifier, the amplification bandwidth would be even larger, such that damping becomes negligible, and a modulation of only 6 electrons per shot would become detectable in theory. We found best signal-to-noise performance, however, in the low-noise mode, where the cut-off frequency was below the repetition rate, as discussed earlier. The lock-in detection separates the contributions from pump pulse and signal pulse as they have different repetition rates. We were therefore able to directly measure a modulation current without the current caused by the pump beam. For a reference measurement using conventional TiPTOE, a pair of copper electrodes with a distance of $(120 \pm 15) \mu \text{m}$ was employed to detect the total ionization yield in gas (i.e., air at 50 mbar). A bias voltage of 10 V between the electrodes was applied directly by the transimpedance amplifier. The sampling speed was around 10-14 data points per second, corresponding to a time interval longer than the time-constant of the lock-in amplifier of 47 ms. In addition, the data acquisition was paused for 100 ms after each step in space, in order to wait for the decay of currents induced by the movement of the tip.

### III. RESULTS AND DISCUSSION

Before performing the actual field measurements, a large raster scan of the nanometric needle tip in the $x,y$-plane in the laser focus was performed to confirm that the current is generated at the apex of the tip with a laser beam polarized along the tip axis (Fig. 1d)). Simultaneously, we recorded the transmitted light in an imaging geometry resulting in the shadow image of the tip shown in Fig. 1c). The comparison of Figs. 1c) and d) demonstrates that photoemission occurred predominantly at the apex of the needle tip (region A), whereas currents from sharp features at the needle shank (region B) were prevented by suitable positioning of the tip. As there is no emission between region A and B, we conclude that there is no emission from the side of the nanometric needle tip. Emission away from the tip apex occurs in regions characterized by a large surface roughness. Therefore, the distance between A and B corresponds to the upper limit for the size of the scanning region. Based on a broad parameter study [32] and the experimentally determined enhancement of around $5.1^{+1.2}_{-0.9}$ (see SI), as well as the opening angle $((21 \pm 2)\degree)$, we estimated an apex radius of the tip of $r = 14^{+11}_{-7}$ nm.

The nanoTiPTOE measurement obtained for linearly polarized sample and pump pulses (polarized along the needle direction) with the needle tip placed in the center of the focus is presented in Fig. 2a). The obtained waveform is compared to a reference obtained via conventional TiPTOE. We note that the enhanced field on the needle tip exhibits a phase shift of typically $0.2\pi$ to $0.5\pi$ compared to the incident field [32]. However, since both beams in nanoTiPTOE experience the same shift due to the enhancement, the overall phase difference is zero and therefore does not affect the measurement. The excellent agreement between the nanoTiPTOE measurement and the reference indicates a rather flat spectral response and demonstrates the capability of nanoTiPTOE to sample near-petahertz laser fields. This conclusion is further supported by the similarity of the measured spectral phases obtained using both methods (see Fig. 2b)). The nanoTiPTOE measurements are only slightly red-shifted, as evident from the spectral amplitudes of the measured pulses in Fig. 2b) and the calculated response, see SI Fig. S5. This difference relates to the response function of the nanometric needle tip. The good agreement of the time-domain waveforms indicates a secondary importance of this small red-shift to most applications.

To further validate the nanoTiPTOE technique for field sampling, we also performed scans of the dispersion of the signal pulse, its carrier-envelope-phase, the field-strength ratio (see corresponding SI sections) and its polarization. For the investigation of the polarization dependence, we kept the pump beam polarized along the tip axis and rotated the polarization of the signal beam, see Fig. 2c). As the superposition of both beams drives the ionization process, we would expect a scaling of the signal in free-space as $\sim |\cos(\beta)|$, where $\beta$ is the angle between the polarizations of the two laser pulses (solid blue line in Fig. 2c)). However, at a nanostructure, the pump beam generates surface normal near-fields that the signal beam can interfere with (cf. Fig. 1c) and Ref. [32]). Our polarization scan (Fig. 2c)) suggests that the interference of the signal beam with surface normal near-fields has only a minor influence on the
FIG. 2. Field measurement using nanoTiptoe: a) Average and standard deviation of five nanoTiptoe field measurements in the center of the laser focus together with a reference obtained using Tiptoe in gas. We corrected the data for a small offset by removing frequency components below 0.01 PHz. The enhanced intensities were $(7.2 \pm 0.9) \times 10^{13}$ W cm$^{-2}$ and $(4.6 \pm 0.6) \times 10^{12}$ W cm$^{-2}$ for pump and signal beam in the nanoTiptoe regime, respectively. The second axis indicates the current modulation in the nanoTiptoe regime. The excellent agreement of nanoTiptoe and standard Tiptoe can be seen in the inset. b) Spectral amplitudes with error bar and spectral phase, obtained via a Fourier-transform of the data in a). c) Detected amplitudes depending on polarization angle of the linearly-polarized signal beam (red dots). As expected, the signal scales proportional to $|\cos(\beta)|$. The pump polarization remained the same.

signal taken with nanoTiptoe, as the signal amplitude for perpendicular polarization nearly reaches the electronic noise amplitude (dashed blue line, Fig. 2c)). Therefore, nanoTiptoe exhibits a polarization sensitivity that allows to map the two-dimensional polarization state.

Having established that nanoTiptoe provides the electric field waveform of the sampling field, we can now investigate how scanning the needle tip across the focal plane provides spatially-resolved data. Some care has to be taken in such scanning measurements. As the measured waveform samples only the relative phase between signal and pump pulse [23, 24], the mode size of the pump beam has been made smaller by a factor of roughly 2.4, causing a larger focal spot size. This increase in size leads to a rather flat pump beam intensity and phase profile over the area that is scanned for the sampling pulse. In order to demonstrate the capability of nanoTiptoe in spatio-temporal field sampling of near-petahertz fields, we sampled a light beam carrying orbital angular momentum (OAM) [28, 29]. The signal beam was shaped by a vortex plate (Vortex Photonics V-780-20-1), that preserves linear polarization, into an OAM beam. As the wave plate had a limited bandwidth, a suitable bandpass filter was added, which increased the pulse duration to 33 fs. The pump pulse, however, was not modified such that the temporal gate remained short. The vortex beam resulting from the beam shaping of the signal pulse is expected to exhibit a field distribution with a singularity on the propagation axis as well as a helical phase shape (cf. Fig. 1a) and Fig. 3c)).

In order to map the evolution of the vortex field in the focal plane, we scanned the tip through the beam while varying the delay over a few oscillations of the most intense part of the signal pulse. The result is depicted in Fig. 3a). A clear rotational motion of the field amplitudes around the center of the focal spot is observable. The extracted amplitude and phase as a function of the needle tip position are shown in Fig. 3b) and d), together with a theoretical expectation for a Laguerre-Gaussian mode (c) and e)), see SI for details. An animated version of Fig. 3a) can be found in the supplementary material. The field distribution exhibits a typical doughnut-shape, the minimum of which is visible in the middle despite the pump beam being maximal there. In order to validate that nanoTiptoe provides full spectral resolution at every point in space, we also performed scans over the full pulse length, but only at selected points marked as A and B in Fig. 3b). The corresponding data was Fourier-transform filtered and is shown in Fig. 3f).
FIG. 3. Spatially-resolved measurements of OAM beams: 

a) The field amplitudes as a function of space and time, normalized to unity. 

b) The amplitude of the measured current modulation induced by the OAM beam together with the expected shape for the theoretical Laguerre-Gaussian beam shown in c). Deviations between experimental data and the simple OAM profile can be attributed to astigmatism in the focusing optics as well as to a non-Gaussian mode shape that has been used for the vortex generation. 

d) The extracted phase at each point in the sampled plane exhibits a helical profile as theoretically shown in e). 

f) Field sampling with nanoTiptoe at the points A and B at opposite sides of the focus, indicated in b). 

g) The phase for each point within the dashed circle in d) (red dots) shows a linear trend with $\alpha$ (dashed blue line). The raw data used for the plots in a), b), d) and g) can be seen in supplementary figure S1. To generate the plot in f), a Fourier filter from 0.1 to 1 PHz has been applied in order to remove nonlinear distortions.

The points A and B were chosen at opposite sides of the mode, since this is where we expect the spatial phase difference to be maximum. Indeed, the corresponding waveforms exhibit a clear $\pi$-phase difference, which is due to the OAM of the signal beam. As can be seen in Fig. 3d), one of the main features of light carrying orbital angular momentum, the helical phase front, agrees well with the theoretical prediction, Fig. 3e). 

To quantify the spiral phase, we introduced a polar angle $\alpha$, and evaluated all phase points within the dashed circle in Fig. 3d). We fixed the central point and calculated the corresponding $\alpha$ for every data point in the region of interest. The result (red dots in Fig. 3g)), is in qualitative agreement with the expected linear increase of the phase with $\alpha$. We attribute the small deviations from the linear scaling for $\alpha \in [120^\circ : 220^\circ]$ to a curved pulse front of the pump beam, which is even visible without vortex plate, see SI for details.
IV. CONCLUSIONS

In conclusion, we have demonstrated that nanoTiptoe enables the spatially resolved measurement of near-petahertz optical field oscillations with sub-cycle resolution. The localized probe enabled the spatio-temporal characterization of optical fields by employing a nanometric needle tip instead of the conventional electrodes. The field enhancement allowed the characterization of laser fields with moderate intensity - a major advance compared to techniques requiring high-power laser sources. As compared to previous approaches for electronic field detection with nanotips, which were often limited to the low terahertz region [4, 5], nanoTiptoe increases the temporal resolution by nearly three orders of magnitude. While the detected bandwidth is comparable to that in Ref. [38] using attosecond streaking spectroscopy, the nanoTiptoe approach is much simpler and avoids a complex vacuum beamline. A combination of nanoTiptoe with latest approaches in time-resolved scanning tunneling microscopy [35, 36] seems promising in characterizing the light-induced near-fields of a nanometric sample with attosecond precision. Theoretically, the resolution here is limited by the interaction of the nanometric needle with the field close to the apex, as extensively studied in Ref. [32]. For our tip geometry, we expect a maximum resolution in the order of the tip diameter, and even down to 1 nm [39] using smaller tips. Finally, orienting the needle along the propagation direction of the laser beam [40] may pave the way towards the measurement of the longitudinal component of strongly focused light with nanoTiptoe. Such measurements would offer a more detailed understanding of the properties of focused light in superresolution microscopy.

AUTHOR CONTRIBUTIONS

J.B. and J.S. contributed equally to this work. J.S. and M.F.K. conceived the nanoTiptoe concept. P.H. contributed expertise on nanometric needle tips. A.S., M.F.K., and P.C. conceived the experiment with OAM beams. J.B., J.S., A.M., and N.S. performed the measurements. Z.W. and P.R. supported laser operations. The data was analyzed by J.B. and J.S. The manuscript was written by J.B., J.S., B.B. and M.F.K. and reviewed by all authors.

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DISCLOSURES

The authors declare no competing interests.
DATA AVAILABILITY

Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

SUPPLEMENTAL DOCUMENT

See Supplement 1 for supporting content as well as the supplementary animation.

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Spatio-temporal sampling of near-petahertz vortex fields: supplemental document

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This document provides supporting information on the main article. We present not only additional data, but also investigate different aspects of our work theoretically. Among them, the response function as well as the intensity scaling. The additional data includes the data for spatio-temporal scans, an intensity calibration, a dispersion scan, a carrier-envelope-phase scan as well as for a non-linearity scan.

I. SPATIO-TEMPORAL SCAN OF ORBITAL ANGULAR MOMENTUM BEAMS

NanoTiptoe is a reliable tool in sampling optical waveforms in space. Supplementary Figure 1 shows the raw data used to generate Figs. 3a, b, d and g in the main text. For this purpose, the tip was scanned over the focal plane and the delay was sampled over 2.5 optical cycles of the orbital angular momentum beam at each point. Please note that the detected signal was a voltage signal despite the fact that a current has been generated, as the transimpedance amplifier was converting the current to a voltage. To each measured point, we fit a sine wave of the form \( u(t) = u_0 \cdot \sin(2\pi f \cdot t + \phi) + u_c \) to the data. There, \( u_0 \) is the amplitude, \( f \) the frequency, \( \phi \) the phase and \( u_c \) a constant offset that models the non-linear distortions (see also SI Sec. VI). The resulting image for \( \phi(x, y) \) is shown in Fig. 3d in the main text. The offset \( u_c \) has been subtracted in main text Fig. 3a). Besides the phase information, we also got the amplitude information as we always scanned over the main pulse, as shown in main text Fig. 3b) and SI Fig. 2a). The current generated by the pump pulse is shown in part b) of SI Fig. 2. Despite the different mode sizes by a factor of 2.4, the comparison of SI Fig. 2a) and b) suggests that probe and pump beam have an equal size. However, this is misleading as firstly, when changing a Gaussian beam of fixed waist to an OAM beam, the apparent focal size becomes larger, as the Gaussian function is multiplied by a factor \( \sim r^{1/4} \) for \( l = 1, p = 0 \), c.f. (1). And secondly, the detected signal is proportional to the field (part a)), whereas the injection current (part b)) is proportional to the ionization rate, that is, a non-linear function of the field which makes the spot size occurring smaller.

As mentioned in the main text, we can of course measure the complete electric field of the laser pulse. To prove that we indeed resolve the complete spectrum, we compared the calculated spectrum of the nanoTiptoe measurement to the spectrum taken with a commercial spectrometer (Thorlabs CCS200/M) and an integrating sphere, as can be seen in SI Fig. 2. The spectrum of the nanoTiptoe data is exactly as broad as the bandpass filter supports. The spectrometer data shows the same peaks in the spectrum with similar height, but is slightly blue-shifted. This can be explained by the wavelength precision that the device has, as there are up to 2 nm tolerance according to the producer. This is around the observed difference of 2-3 nm. For the OAM data, we averaged the two full-time scans performed at points A and B (c.f. Fig. 2 in the main part). The average was performed, as the integrating sphere is also averaging over the mode. However, we found that the difference in the spectra between point A and B is rather small. A complete scan over the mode, especially also without bandpass filter would be very interesting regarding the wavelength dependent properties of the OAM generation. However, such a measurement would be practically hard to implement under long-term controlled conditions, as with the scan times in our experiments the duration would be of the order of a day.

For the expected field distributions of the Gaussian Beams and OAM beams, we used an expression for the electric
FIG. 1. Spatio-temporal scan of orbital angular momentum beams - At each point in the $x,y$-plane, we scanned over 2.5 optical cycles of the OAM beam. This measurement not only yields the phase information, but also the amplitude information, as we scanned over the maximum of the laser pulse. This is the raw data used to generate parts a), b), d) and g) of Fig. 3 in the main text.

FIG. 2. Amplitude and spectrum of the OAM beam: a) The amplitude of the current modulation in space has a ring shape. b) The corresponding injection current does not carry orbital angular momentum and is a round spot rather than a ring. c) The blue filled area is the calculated spectrum of the nanoTiPTOE measurement. It is located exactly in the region that the bandpass filter supports (dashed grey line). The spectrometer data (red) reproduces the peaks in the spectrum very well, but is slightly shifted in wavelength. This can be explained by the wavelength uncertainty of the device that is specified to be 2 nm.
field in space adapted from Ref. [1] which reads

\[ E_p(r, \varphi, z, t) = E(t) \frac{C(l, p)}{\sqrt{1 + z^2/z_R^2}} \left( \frac{r \sqrt{2}}{w^2(z)} \right)^{|l|} L_p^l \left( \frac{2r^2}{w^2(z)} \right) \times \exp \left[ -\frac{r^2}{w^2(z)} + \frac{-ikr^2z}{2(z^2 + z_R^2)} \right] \times \exp \left[ -il\varphi + i(2p + l + 1) \arctan \left( \frac{z}{z_R} \right) \right] \times \exp \left[ -ikz + i\omega t \right]. \]

Here, \( r, \varphi \) and \( z \) are cylindrical coordinates, \( E(t) \) is the temporal envelope, \( z_R \) is the Rayleigh length, \( w(z) \) is the beam waist, \( L_p^l(.) \) are associated Laguerre polynomials and \( C(l, p) = \sqrt{\frac{2^p}{\pi \Gamma(p+|l|)!}} \) is a constant defined by the azimuthal and radial indices \( l \) and \( p \), respectively. The central frequency is \( \omega \) and the \( k \) is the wavenumber. In most cases, we assumed to be directly in the focal plane, i.e. we set \( z = 0 \) and \( w(z = 0) = w_0 \). For a Gaussian beam, we set \( l = p = 0 \), whereas for our OAM beam, we set \( l = 1, p = 0 \).

Please note, we often found that the measured phase front exhibits a linear behavior in space that can be quite large. It turned out to be that this was caused by imperfectly centered modes of pump and probe beam. In an illustrating picture, this can be explained by the \( k \)-vectors of the pump and signal beam propagating under an angle through the focus which ends up in a linear phase relation in space between the two beams. This mode mismatch could be caused by parallel but slightly shifted beams. In the measurements presented here, we prevented this additional phase term by carefully aligning not only parallelism of the beams, but also on centering one beam into the other. The small additional curvature of the phase front we detected in the vortex beam measurements (c.f. main text Fig. 3d)) was caused by a slightly quenched optic in the pump beam path and therefore even present without vortex plate.

II. DISPERSION SCAN

To verify that nanoTiptoe probes the light field, we systematically scanned different light properties as the dispersion. Therefore, we moved the fused silica wedges in the signal arm while keeping the pump beam untouched, i.e. compressed. The temporal overlap was realigned using a manual stage that the piezo stage was attached to. Please note, that neither bandpass filter nor vortex plate was used here. Additionally, a fused-silica window (thickness \( 1.05 \pm 0.05 \) mm) could be added to increase the scan range. As can be seen in SI Fig. 3a), the calculated spectrum from the data is similar for all glass insertions, whereas the time-domain pulse shapes are either chirped or compressed, see SI Fig. 3c). The spectral phase, however, clearly shows a quadratic behavior when the pulse is chirped. Additionally, there are some modulations on the phase that are most likely caused by the 8 pairs of double angle chirped mirrors (PC70, Ultrafast Innovations) used for the pulse compression. To extract the group-delay-dispersion (GDD), we fit a third-order polynomial to the spectral phase. We weighted the fit with the spectral amplitudes in order to avoid overfitting of phase terms with low amplitude. The resulting second order dispersion is shown in SI Fig. 3b). It clearly follows a linear scaling and a fit yields a dispersion coefficient of \( 40.59 \text{fs}^2/\text{mm} \), which is only \( 0.63\% \) deviation from the literature value of \( 40.335 \text{fs}^2/\text{mm} \) for fused silica at 750 nm central wavelength (0.4 PHz).

III. RESPONSE FUNCTION OF NANO TIPTOE

The response function of nanoTiptoe consists of two major contributions: Firstly, the incident fields are enhanced due to the geometric shape of the tip and secondly, the ionization process at a surface which is the field sampling process. The response function of the latter has already been discussed in many details in Ref. [3, supplementary], where the Tiptoe technique has been applied to nano triangle arrays. We will therefore give a brief overview only. Tiptoe-like methods rely on a few-cycle pump pulse that ideally only ionizes some medium during the strongest half-cycle creating a field-dependent current. The total generated charge, i.e. the time-integral over the ultrafast current is then detected. This charge is now modulated by a weak perturbing field, the signal field. As this modulation happens only during one sub-cycle ionization burst, the modulation will follow the signal field, depending on the temporal delay between pump and signal field [4], see SI Fig. 4a). We can write the emitted charge \( Q \) in terms of the ionization rate \( w(t) \) and the delay \( \tau \) [3, 4]:

\[ Q(\tau) \propto \int w(E_p(t - \tau) + E_s(t)) dt, \tag{2} \]
FIG. 3. Dispersion scan with nanoTIPTOE: a) The calculated spectra and phases for two measured pulses with different dispersion show the same spectral amplitudes but different quadratic phases. A fit of a third-order polynomial, weighted with the spectral amplitudes, delivers the value for the group-delay-dispersion. The result of a systematic scan of the dispersion in the signal arm is shown in b), clearly following a linear trend. A linear fit yields a GVD of 40.59 fs$^2$/mm, which is only 0.63% deviation from the literature value of 40.335 fs$^2$/mm [2]. c) Time-domain signals corresponding to the spectra shown in a).

where the time integral extends over the pulse length. Since the signal field is weak, the kernel of the integral can be Taylor-expanded up to first order:

$$Q(\tau) \propto \int w(E_p(t - \tau)) \cdot E_s(t) dt,$$

(3)

where the last term corresponds to the cross-correlation of the gating function with the signal field [3]. The first term corresponds to the rate without signal field. Moreover, the gating function for a tunneling emission burst at $t_0$ can be approximated as a delta-function $\delta(t - \tau - t_0)$, yielding an expression for $\Delta Q$:

$$\Delta Q(\tau) \propto \int \frac{dw}{dE} \bigg|_{E_p(t - \tau)} \cdot E_s(t) dt \propto \frac{dw}{dE} \bigg|_{E_p(t_0)} \cdot E_s(t_0 + \tau).$$

(4)

For a single emission burst, the charge modulation is thus approximately proportional to the electric field. For several tunneling emission bursts, the last expression on the left hand side becomes a sum over all burst times $t_k$. This is illustrated in Fig. 4b) for a pump pulse (not shown) with $\varphi_{\text{CEP}} = \pi$, where two dominating equal emission bursts occur. As can be seen, in this case, signal frequencies at $0.5 f_0$ (red line) and $1.5 f_0$ (black line), where $f_0$ is the pump pulse frequency, do not lead to a modulation of the emitted charge. The reason is that the emission modulation of the first burst is counteracted by the second burst since the signal fields point into opposite directions. This is not the case for $\varphi_{\text{CEP}} = 0$. Please note, that in our coordinate system, the tip is pointing in negative $x$-direction. Therefore, a pulse with $\varphi_{\text{CEP}} = 0$ will cause a single ionization burst, as its strongest half-cycle points into the tip surface. The resulting CEP-dependence of the spectral response function can be seen in Fig. 4c), and has been extensively discussed in Ref. [3]. The spectral response function can be calculated either by Fourier transforming the gating function in Eq. 3, or by evaluating the full expression for the emitted charge for a given pump and signal field in Eq. 2 subsequently forming the ratio of $\Delta Q$ and $E_s$ in the Fourier-domain. We chose the latter approach, since it does not rely on the first-order Taylor-expansion. The results shown here use a Fowler-Nordheim emission rate[5, 6] assuming a work function of 4.5 eV, a pump pulse at an intensity-FWHM of 4.2 fs, a central wavelength of 750 nm and intensity of approximately $3 \cdot 10^{13}$ W/cm$^2$. In order to obtain a large bandwidth, we chose a signal pulse duration of 2 fs at 750 nm wavelength and a relative field strength of $10^{-5}$.

Figure 4c) shows the amplitude (solid lines) and phase (dashed lines) of the spectral response function $H(f)$ for a CEP of 0 (red) and $\pi$ (black), respectively. As discussed above, for $\varphi_{\text{CEP}} = \pi$, the amplitude response exhibits zeros at $f = 0.5 f_0 + n \cdot f_0$, where $n$ is an integer. The response function for $\varphi_{\text{CEP}} = 0$ only shows a slight modulation caused by the small satellite emission bursts one cycle earlier and later as shown in Fig. 4a). Around $f_0$, the phase
of the response function changes with the CEP of the pump pulse. As pointed out in Refs. [3, 7, 8], if both pump and signal pulse originate from the same laser source, this allows the auto-characterization of the pulse without CEP-stabilization, since the relative phase of both pulses does not change. However, this statement only holds true if the bandwidth is relatively narrow. For very broadband laser pulses, the CEP dependence of the response amplitude is expected to play a role. However, in our experiments we found only minor influence of the CEP of the driving pulse on the detected spectrum.

FIG. 4. The nanoTiptoe response function: a) The nanoTiptoe-principle: The ionization rate (blue line area) of a strong pump pulse (red line) is perturbed by a weak signal pulse (red dashed line). The change in ionization is essentially proportional to the perturbing electric field at the time of the emission peak. At a surface, electron emission only occurs if the electric field points into the surface, that is, with \( \varphi_{\text{CEP}} = 0 \) in our coordinate system. b) Illustration of the reason for the minima in the amplitude response for \( \varphi_{\text{CEP}} = \pi \) at 0.5 \( f_0 \) (red line) and 1.5 \( f_0 \) (black line). c) Amplitude (solid lines) and phase (dashed lines) of the response function for \( \varphi_{\text{CEP}} = 0 \) (red) and \( \varphi_{\text{CEP}} = \pi \) (black) for a pulse width of 4.5 fs (FWHM) calculated using the Fowler-Nordheim rate. For a 10 fs pump pulse (blue line), practically no CEP-dependence is observed (intensity \( 3 \times 10^{13} \text{W/cm}^2 \), work function=4.5 eV).

Additional to these properties inherited by the ionization process, the near-field enhancement plays in. The reference measurement presented in main text Fig. 2 allows to calculate the spectral response function as shown in SI Fig. 5. The function shown there is the difference in response between nanoTiptoe and the reference, as the reference might still have a (potentially negligibly flat) response. Note, that we can only calculate the response function up to a scaling factor, as the amplitudes of the data are not comparable and were normalized first (main text Fig. 2). For comparison, we simulated the response function of the enhancement (red line in SI Fig. 5) using a commercial finite-difference time-domain (FDTD) solver (Lumerical FDTD). We simulated a nanotip modeled by a truncated cone with 10.5° half-opening angle and a half-sphere with radius 15 nm as termination and a Gaussian pulse with 4.5 fs duration. The simulated response function is rather flat over the spectral region of interest, but agrees with the nanoTiptoe data within the uncertainty range.

FIG. 5. Response function - The extracted response \( |H| = \frac{|E_{\text{nanoTiptoe}}|}{|E_{\text{ref}}|} \) compared to FDTD simulations (red line) with a 4.5 fs Gaussian beam.
IV. CEP DEPENDENCE OF NANO TIPTOE

Recent literature suggests that TIPTOE-type measurements only detect the relative phase between the pump and signal pulse\cite{3, 7, 8}. This corresponds to a phase term of the response function that is proportional to the carrier-envelope-phase of the pump pulse\cite{3}. For a more detailed discussion, see also section III. We experimentally confirmed that indeed the relative phase is detected. However, phase changes in one arm of the interferometer were indeed measurable which not only establish the applicability of nanoTIPTOE even to CEP-unstable lasers, but allow to detect modifications in one interferometer arm like the vortex generation.

For the investigations on the CEP dependence of both pulses, we changed their CEP simultaneously using the Dazzler in the laser amplifier. SI Figure 6a) illustrates that the measured CEP with nanoTIPTOE is nearly constant even though the carrier-envelope-phases of pump and signal pulse changes. If the response phase was independent of the CEP of the pump pulse, a linear increase would be expected (dashed line in SI Fig. 6a)).

To verify the phase sensitivity, we changed the CEP of the signal pulse only using fused silica wedges. Knowing the group and phase refractive index of \( n_{ph} = 1.4542 \) and \( n_{gr} = 1.4689 \)\cite{2}, we calculated the needed thickness change of the glass for a \( \pi \) phase flip to be \( x = \frac{\pi}{\lambda}(n_{gr} - n_{ph}) = 25 \mu m \). This change corresponds to a displacement of 730 \( \mu m \) at 2° opening angle of the wedge. From the variable \( x \), we calculated the group-delay and shifted the signal by the corresponding time-shift of 38.5 fs. We employed the full scanning range of the piezo delay stage which allows us to directly shift the signal by the time shift. Fig. 6b) shows that the unfiltered data after consecutive CEP shifts indeed shows a phase flip of \( \pi \) each time.

FIG. 6. CEP dependence: a) The measured CEP of the nanoTIPTOE signal is nearly constant (red dots), whereas the set CEP increases from 0 to 2\( \pi \). b) When only changing the CEP of the signal beam, the phase change can be measured well. The plot is showing raw data.

V. INTENSITY CALIBRATION

For the incident intensity, we found a power to intensity conversion given by

\[
I(P[W]) = 4.2 \times 10^{15} \cdot P[W] \frac{W}{cm^2} \\
\pm \left[0.9 \times 10^{15} \cdot P[W] + 4.2 \times 10^{15} \cdot \Delta P[W]\right] \frac{W}{cm^2}
\]  

(5)

for the pump beam with 4” focal length. The incident power \( P \) has the measurement tolerance \( \Delta P \). The above formula follows from an estimation via Gaussian optics. However, we corrected deviations by an image of the focal spot on a camera in a separate imaging setup. Accordingly, the values given above are only (30 \( \pm \) 5)% of the expected intensity by pure Gaussian optics. The given errors follow from a Gaussian error propagation of the uncertainties of all the initial quantities. The signal beam intensity is larger by a factor of around 5.9 as the mode size is different.

Taking also the lower repetition rate into account, the intensity should be around 11.8 times higher for the same incident power.

To estimate the local intensity at the tip, we investigated the intensity dependence of the emission current. For this purpose, the pump beam power is scanned while detecting the current, see SI Fig. 7. We estimated the number of emitted electrons per shot taking 10 kHz repetition rate, an amplification of \( 10^9 \frac{V}{A} \) and the damping rate, as discussed
in the main text, into account. The ionization current exhibits two different scaling laws: For low incident intensity, the current scales as $I^4$, whereas for higher intensities, the scaling is proportional to $I$. Similar curves have been observed in previous work [6, 9–13]. We follow a similar approach as in those references, especially Ref. [9] suggests the identification of the kink in the nonlinearity with the transition from multiphoton to tunneling ionization. This allows to set the Keldysh parameter $\gamma = \frac{\omega \Delta m \Phi}{e E}$ to unity at that point [14], which sets the enhanced intensity knowing the work function of tungsten of $\Phi = 4.5$ eV [15]. Even within different measurements with the same nanometric needle tip in different beam times, the kink in nonlinearity was always at an incident power of $P = (3.9 \pm 0.4) \times 10^2 \mu W$ for the 4° focal length optics. Regarding the nonlinearity in the multiphoton regime, the scaling of $\propto I^4$ is of higher order than expected from the number of photons needed to ionize the tip, that is 3 at a photon energy of 1.65 eV, see Ref. [16]. Similar observations have been made for instance in Ref. [9], as well as in other work on tungsten [17–22]. In those references, a higher density of states further below the Fermi level causes the higher nonlinearity. Having fixed the kink position, we can write the enhanced intensity as

$$I(P[W]) = 1.1 \times 10^{17} \cdot \frac{P[W]}{\text{cm}^2} \pm [1.2 \times 10^{16} \cdot P[W] + 1.1 \times 10^{17} \cdot \Delta P[W]] \frac{W}{\text{cm}^2}$$  \hspace{1cm} (6)$$

for the pump beam. For 1 mW incident power, the enhanced intensity at the tip is $I = (1.1 \pm 0.2) \times 10^{14} \frac{W}{\text{cm}^2}$. Comparing this to the incident intensity calibration above, we can estimate the intensity enhancement to $26^{+12}_{-9}$, or equivalently, a field enhancement of $5.1^{+12}_{-9}$. We note that Ref. [23] has demonstrated that the kink in the photoemission scaling is also connected to the onset of charge interaction. However, it is also stated there that, despite there being charge interaction, the measurement of field-dependent currents is still possible. Accordingly, we conclude that space-charge interaction might be present in our experiments, but does not affect the sampled waveform.

The estimated enhanced near-field intensities in the nanoTIPTOE regime were $(7.2 \pm 0.9) \times 10^{13} \frac{W}{\text{cm}^2}$ and $(4.6 \pm 0.6) \times 10^{12} \frac{W}{\text{cm}^2}$ for pump and signal beam, respectively, for the dataset presented in Fig. 2 in the main text. The incident intensities were lower according to the enhancement factor. In the TIPTOE reference measurements, the incident intensities were around $(4 \pm 1) \times 10^{13} \frac{W}{\text{cm}^2}$ for the pump beam and $(2.0 \pm 0.4) \times 10^{11} \frac{W}{\text{cm}^2}$ for the signal beam. The TIPTOE mechanism relies on the signal field being only a weak perturbation. Despite the signal beam intensity in our experiments being more than 6% of the pump intensity, we did not find significant non-linear distortion, see also the experimental investigation in section VI.

![Intensity scan: The number of emitted electrons (solid blue line and blue area) scales as $I^4$ (dashed black line) for low (local) intensities, and as $I^1$ (dashed red line) for higher (local) intensities. The dashed lines are only a guide for the eye.](image)

**FIG. 7.** Intensity scan: The number of emitted electrons (solid blue line and blue area) scales as $I^4$ (dashed black line) for low (local) intensities, and as $I^1$ (dashed red line) for higher (local) intensities. The dashed lines are only a guide for the eye.

### VI. DIFFERENT REGIMES OF NANOTIPTOE

Originally, TIPTOE has been derived for conditions where the signal field only constitutes a small perturbation of the pump field [4], but has also been extended beyond this condition at the expense of a more complicated retrieval [24, 25]. We experimentally verified that the linear approximation (see eq. (3)) in the nanoTIPTOE sampling process breaks
down if the field ratio between signal and pump beam is too large, see SI Fig. 8a). The scaling of the detected amplitudes leaves the linear regime as soon as the electric field ratio is larger than 0.3. To deduce the actual field-ratio, we employed similar mode sizes of signal and pump beam in order to avoid the uncertainties that come with the intensity calibration above. By doing so, we directly extract the field-strength ratio by the square-root of the power ratio, where we varied the signal field while keeping pump constant. Here, we made sure that the dispersion of both beams is balanced best. The results reproduce simulations using a Fowler-Nordheim tunneling [5] rate with comparable field-strengths as in our experiments, but with every second optical half-cycle suppressed [10].

At the highest signal powers, we observe a prominent asymmetry of the nanoTiptoe signal that is characterized by a low-frequency offset, reminiscent of an autocorrelation signal, see SI Fig. 8b) upper graph. In contrast, for lower signal field strength, no such offset is visible. We refer to that offset as non-linear offset or non-linear distortion, as it is only present when nanoTiptoe has left the linear regime. This low frequency offset can be seen in terms of higher orders of the Taylor expansion in (3). There, the second order term is proportional to $E_S^2(t)$ which is the time-domain representation of sum and differency frequency generation [4].

We also demonstrate that using Fourier transform filtering of a dataset with too large perturbation, we can reconstruct the sampled waveform taken with appropriate field strength, see SI Fig. 8c). There, the waveform shown in SI Fig. 8b) (red), has been Fourier transformed and filtered. Frequency components below 0.3 and above 0.6 PHz have been cut. The normalized back-transformation agrees well with another waveform sampled with a smaller perturbation of the ionization. As we employed a few-cycle pump pulse, a more sophisticated reconstruction algorithm [25] is not necessary.

FIG. 8. Nonlinearity scan: a) The amplitudes in positive and negative direction of the sampled waveform (red and blue dots) leave the linear scaling regime (dashed) at a field-ratio of around 0.3. The behavior follows the same nonlinearity as expected by simulations modeling the process with Fowler-Nordheim tunneling. b) Two waveforms with taken with a different signal field strength show a low-frequency background as well as harmonic distortions if the field strength is higher (red line). c) The blue line represents the raw data sampled with low signal field strength. It is well reproduced when applying a Fourier filter between 0.3 and 0.6 PHz to the waveform sampled with higher field strength (red).

VII. DAMAGE THRESHOLD FOR NANOTIPTOE

Similar intensities have been reached in other needle tip experiments[6, 9, 26] without observing damage to the tip. Specifically Ref.[9] states that under conditions similar to our experiment, even at intensities up to a factor 6 above the kink, no damage seems to occur to the tip. Several studies indicate that one of the major damaging mechanisms for nanostructures is heating by the electric field inside the material in combination with inefficient heat conduction[27–29]. Accumulated heating is rather a problem at MHz-repetition rates[27] which has also been shown to potentially alter the emission characteristics[30]. In contrast, at kHz rates there is enough time between consecutive pulses for the heat to be dissipated. Moreover, since the nanometric needle tip quickly takes on a micron length scales away from the apex due to the conical structure of the shank, a better heat dissipation can be expected compared to e.g. nanowires. Nevertheless, those nanowires exhibit a damage threshold of around $10^{13}$ W/cm$^2$ of incident intensity at kHz repetition rates[29]. Finally, as a general rule the damage threshold increases with decreasing pulse length. In this regard, our experimental conditions where a tungsten tip is irradiated by sub-two-cycle laser pulses at 10 kHz repetition rate and below, we do not expect damage as our incident intensities are even below $10^{13}$ W/cm$^2$, as can be calculated from (5). Indeed, we do not observe significant changes of the photocurrent over a several days long period of measurements. We also conclude that there is no significant damage by the repeatability of the measurement with
the same nanometric needle tip.

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