EPJ Web of Conferences **205**, 08011 (2019) https://doi.org/10.1051/epjconf/201920508011
UP 2018

Antiresonant-like behavior in carrier-envelope-phase-sensitive sub-optical-cycle photoemission from plasmonic nanoantennas

Phillip D. Keathley[1,*], *William P.* Putnam[1,2,3] , *Richard G.* Hobbs[1,4] , *Karl K.* Berggren[1] , and *Franz X.* Kärtner[1,2,5]

[1]Massachusetts Institute of Technology, Department of Electrical Engineering and Computer Science and Research Laboratory of Electronics, 77 Massachusetts Ave., Cambridge, MA 02139, USA

[2]University of Hamburg, Department of Physics and Center for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany

[3]Northrop Grumman Corporation, NG Next, 1 Space Park Blvd., Redondo Beach, CA 90278, USA

[4]Centre for Research on Adaptive Nanostructures and Nanodevices (CRANN), Advanced Materials and Bio-Engineering Research Centre (AMBER), and School of Chemistry, Trinity College Dublin, Dublin 2, Ireland

[5]Center for Free-Electron Laser Science and Deutsches Elektronen-Synchrotron (DESY), Notkestraße 85, 22607 Hamburg, Germany

Abstract. We study carrier-envelope-phase-sensitive (CEP-sensitive) photoemission from plasmonic nanoantennas excited with laser pulses of varying incident optical intensities. The CEP-sensitive current exhibits antiresonant-like features that we attribute to competing sub-optical-cycle bursts of photoelectrons.

It has recently been shown that the electric field of ultrafast optical pulses can control electrical currents around nanoscale vacuum electronic structures at frequencies exceeding 100 THz [1-4]. Such extreme electrical control requires optical-field emission, *i.e.* the emission of sub-optical-cycle bursts of electrons via optical-field-driven tunneling. A hallmark of optical-field emission is sensitivity of the total emitted current to the carrier-envelope phase (CEP) of the driving optical pulse. Such CEP-sensitive photocurrent has been observed in the emitted electron spectra from isolated nano-tips [1,2], and more recently in total current yields from nanoscale structures [2-4]. Here, we present a study of optical-field-controlled, CEP-sensitive photoemission from nanoantennas performed under varying optical intensity. We observed antiresonant-like behavior in the photocurrent's CEP response where at a critical intensity the CEP-sensitive photocurrent exhibited a significant dip in magnitude as well as a dramatic 180° shift in phase. Using a quasi-static tunneling model, we attribute this behavior to competition between sub-optical-cycle bursts of charge.

Our experimental approach followed earlier work [4] and is illustrated in Fig. 1a. Arrays of gold nanotriangles were fabricated onto a transparent and conducting indium tin oxide (ITO) film on a sapphire substrate. An ITO collector electrode was located across a ≈ 5 μm gap from the nanotriangle emitter array; micrographs are provided in Fig. 1b. The nanotriangles were illuminated with tightly-focused (≈ 5 μm beam diameter) few-cycle laser pulses (≈ 10 fs duration at a center wavelength of 1177 nm corresponding to ≈ 2.5 cycles). When excited by few-cycle laser pulses, the nanotriangles' localized surface plasmon resonance (LSPR) and sharp features result in enhanced local optical fields. Our measurements indicate a field enhancement of ≈ 32, which agrees well with prior work under

* Corresponding author: pdkeat2@mit.edu

identical illumination conditions [4]. With incident pulse energies ranging between 50-160 pJ, we have enhanced peak fields of 18-32 V/nm (*i.e.* Keldysh parameters of 0.67-0.38). Such strong-fields lead to large photoemission currents (tens of nA). Additionally, with such high fields the photoemission occurs in the strong-field regime where the emission process resembles quasi-static optical tunneling, and the emitted current becomes CEP-sensitive.

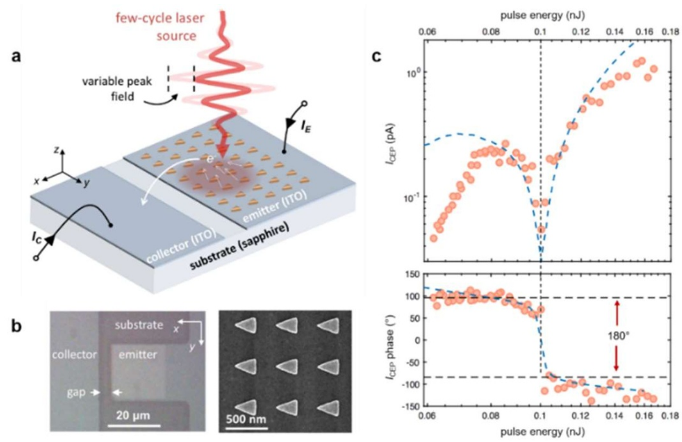

Fig. 1: a. Experimental setup. Few-cycle laser pulses of tunable pulse energy and peak field produce photoemission currents from gold nanotriangles, and these currents are pulled from an emitter electrode to a collector (emitter and collector currents are labeled I_E and I_C respectively). **b.** Optical and electron microscope images of a nanotriangle emitter array. **c.** Measured CEP-sensitive current (I_{CEP}) versus the incident pulse energy. In the bottom panel, the relative phase of I_{CEP} and the f_{CEO} reference is displayed. Simulated I_{CEP} values (blue, dashed line) calculated using a quasi-static tunneling model show remarkable agreement to the experimental data.

We stabilized the CEP of our few-cycle laser source by locking the carrier-envelope offset frequency, f_{CEO}, to a stable 100 Hz reference. We measured the CEP-sensitive component of the emission current, *i.e.* the component of the current at f_{CEO}, via lock-in detection in a 125 mHz bandwidth. In the top panel of Fig. 1c, the CEP-sensitive current, denoted I_{CEP}, is displayed versus driving pulse energy; in the bottom panel the phase of I_{CEP} relative to the f_{CEO} reference is also plotted. We see that I_{CEP} displays antiresonant-like behavior: near 100 pJ pulse energies the magnitude of I_{CEP} abruptly dips, and the phase of I_{CEP} swings by $\approx 180°$. To understand the origins of this behavior, we used a quasi-static tunneling model. We started with $\cos^2$-shaped model pulses (10 fs duration and 1177 nm center wavelength) and then filtered these pulses with the experimentally-characterized transfer function of the plasmonic nanoantennas (the particles were modeled as resonators). Next, we calculated the quasi-static Fowler-Nordheim (FN) tunneling currents driven by these filtered pulses with varying CEP's. After integrating the current over each pulse in both space and time, we then calculated total emitted charge versus CEP and estimated I_{CEP}. We repeated this process for pulses of varying energy to build a simple model of our experiment. The model results are plotted in Fig. 1c (blue, dashed curve) and show excellent agreement with the experimental data.

To provide further insight into the antiresonant-like behavior, we removed the plasmonic filtering from our quasi-static model and repeated I_{CEP} calculations using a 10 fs duration

$\cos^2$ pulse centered at 1177 nm (see waveforms in Fig. 2a). In Fig. 2a, the current profiles from our quasi-static model are shown for two possible CEP phases, $\varphi_{\text{CEP}} = 0$ and $\varphi_{\text{CEP}} = \pi$ (red and blue curves respectively). For analysis purposes, half-cycle regions are highlighted (red and blue regions). Note that the even half-cycles (red regions) exhibit peak yield at or near $\varphi_{\text{CEP}} = 0$, and the odd half-cycles (blue regions) at or near $\varphi_{\text{CEP}} = \pi$. The magnitude and phase of the calculated I_{CEP} is plotted in Fig. 2b as a function of peak optical intensity at the emitter surface. We found that the cause of the antiresonant-like feature in I_{CEP} shown in Fig. 2b is a transition from the photoemission being dominated by odd half-cycle contributions to being dominated by even half-cycle contributions. This is evidenced in the response of total charge yield as a function of φ_{CEP} before and after the dip in I_{CEP}. For example, at a peak intensity of 50 TW/cm^2 charge emission, Q, peaks at $\varphi_{\text{CEP}} = \pm\pi$ (blue dot and top inset in Fig. 2b), and at 125 TW/cm^2 it peaks at $\varphi_{\text{CEP}} = 0$ (red dot and bottom inset in Fig. 2b). At a critical intensity near 80 TW/cm^2, the even half-cycle contributions match those of the odd half-cycles, resulting in a sharp decrease in I_{CEP} as the two are separated in phase by 180°. As the critical intensity point represents the switch between even and odd half-cycle dominance, the phase of I_{CEP} also experiences a sudden 180° swing at this point (right panel of Fig. 2b).

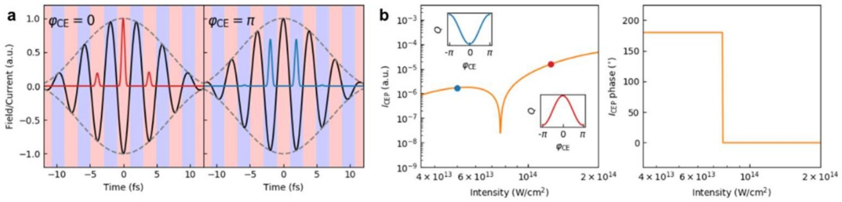

Fig. 2: a. Fowler-Nordheim tunneling emission (red and blue curves) from a $\cos^2$-shaped model pulse (black curve) with $\varphi_{\text{CEP}} = 0$ (left panel) and $\varphi_{\text{CEP}} = \pi$ (right panel). Even half-cycles are shaded red, and odd blue. Note that here we define $\varphi_{\text{CEP}} = 0$ to correspond to a negative peak field. **b.** The CEP-sensitive current (left) and relative phase (right) for a $\cos^2$-shaped model pulse with 10 fs duration exciting a gold surface (with work function 5.1 eV). The insets show the normalized charge (Q) emitted for varying CEPs at two different intensities: 50 TW/cm^2 and 125 TW/cm^2.

In summary, we measured CEP-sensitive photoemission currents from plasmonic nanoantennas driven by few-cycle laser pulses with optical pulse energies ranging between 50-160 pJ. We observed antiresonant-like behavior in the CEP-sensitive current and attribute this behavior to competing emission from different optical half-cycles of the driving laser pulses. While we observed this behavior in total current yields from nanostructured devices, we emphasize that this antiresonant-like behavior in strong-field photoemission is general in nature and should be observable using other systems, such as atoms or molecules.

References

1. M. Krüger, M. Schenk, and P. Hommelhoff, Nature **475**, 78 (2011).
2. B. Piglosiewicz, et al., Nat. Phot. **8**, 37 (2014).
3. T. Rybka et al., Nat. Phot. **10**, 667 (2016).
4. W. Putnam et al., Nat. Phys. **13**, 335 (2017).