Ultrafast optical-field controlled photoemission from plasmonic nanoparticle arrays

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Abstract: Exciting plasmonic nanoparticles with two-cycle optical pulses, we observe photoemission across few-micron gaps under ambient conditions. The photoemission is modulated by the carrier-envelope phase with a signal-to-noise ratio exceeding 20 dB at 1 Hz resolution-bandwidth.
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Attosecond science fundamentally relies on the manipulation and control of ultrafast, attosecond bursts of electrons with optical waveforms. Extending this control from gaseous media to solid-state nanostructures would provide avenues to exciting scientific and technological developments. In the past few years, signatures of strong-field emission and electron re-scattering from nano-tip emitters [1-4] and plasmonic nanoparticles [5] have been observed. Additionally, a hallmark of optical waveform control, carrier-envelope phase (CEP) sensitivity, has been observed in the photoelectron energy spectra [2] and total emission current from single nano-tip emitters [4].

In this contribution, we explore photoemission from plasmonic nanoparticle arrays under ambient conditions, i.e. out of vacuum, and on the surface of a chip. We observe stable, strong-field emission of electrons across few-micron gaps at a high, oscillator repetition rate. We also show that the photoemission current can be controlled by the CEP of the exciting laser pulse. The CEP sensitive current shows > 20 dB signal-to-noise ratio (SNR) at a resolution bandwidth of 1 Hz. Considering that the excitation pulse has less than half an octave of optical bandwidth at full width at half maximum, our devices might find applications as compact, solid-state CEP detectors.

Our experimental configuration is illustrated in Fig. 1a. Our laser source is an Er:fiber seeded supercontinuum (SC) femtosecond source [6,7]. The SC pulses are two-cycle (≈ 8.5 fs) in duration at a central wavelength of 1.2 μm (SC spectrum ranges from 1 - 1.4 μm). The SC source has a repetition rate of 78.4 MHz and a pulse energy of up to 0.3 nJ. The carrier-envelope offset frequency (f_{CEO}) is stabilized to a local oscillator at 2 kHz. The SC pulses are focused by a reflecting objective to a 5.5 μm diameter spot on our devices.

Our devices consist of arrays of plasmonic nanoparticles. The arrays lie on patterned, indium-doped tin oxide (ITO) electrodes on a sapphire substrate. In the image in Fig. 1a, the array of nanoparticles is not visible, however, a

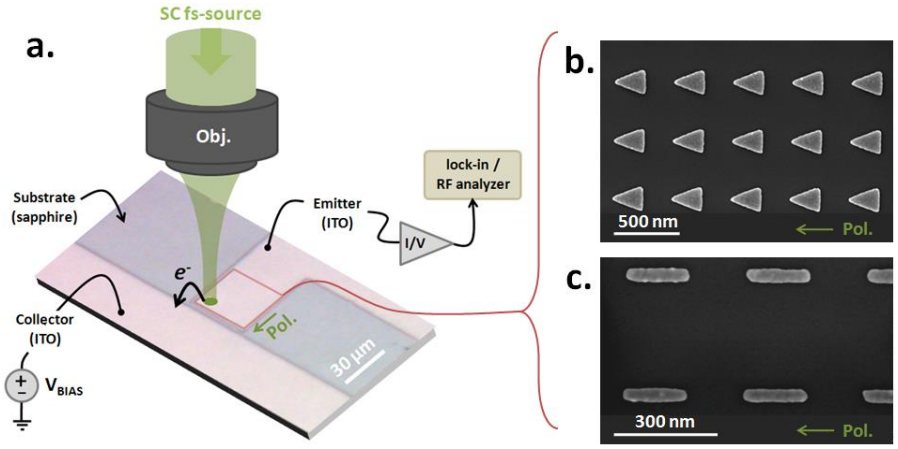

Figure 1: a. Experimental configuration. SC fs-pulses excite a plasmonic nanoparticle array resulting in electron emission. Electrons jump across a small gap from the emitter electrode to the positively biased collector electrode. The photoemission current is amplified in a transimpedance amplifier (I/V) and sent to a lock-in amplifier or a radio-frequency (RF) spectrum analyzer. **b.** SEM image of Au nano-triangle array. **c.** SEM image of Au nano-rod array (polarization direction labeled by Pol.).

red box denotes the array location for clarity. The gray regions of this image correspond to the insulating sapphire substrate and the pink to the ITO electrodes. The emitter electrode connects to the nanoparticle array and is separated from the collector electrode by a 2.5 μm gap; this small gap allows for large bias fields with only modest bias voltages. When the SC pulses excite the device array, photoemitted electrons jump from the emitter to the collector (as illustrated in Fig. 1a). We use two nanoparticle array-types in our experiments—Au nano-triangles (SEM image in Fig. 1b) and Au nano-rods (SEM image in Fig. 1c). Both device-types are designed to have their resonance within the SC bandwidth (near 1.1 μm).

Our experimental results are summarized in Fig. 2. Fig. 2a shows the total photoemission current from the nano-triangle devices for several bias voltages. At pulse energies below ~ 40 pJ, the photoemission is in the multiphoton regime, and the current scales as the fifth power of the intensity. Above ~ 40 pJ, the emitted current falls off from this multiphoton scaling and enters the strong-field emission regime. Additionally, in the inset of Fig. 2a, we compare the current measured from the emitter and that measured at the collector. Under ambient conditions and over a three-minute period, these currents match very well and show good stability (root mean square deviation from the mean current < 4% of the mean current).

The results from the CEP-control measurements are shown in Fig. 2b and 2c. Fig. 2b shows the radio-frequency spectra of the emitter currents from the nano-triangle and nano-rod devices. A large peak at f_{CEO} = 2 kHz is visible in the emitter current (I_E) from the nano-triangle devices, while no peak is visible for the nano-rods. The highly non-linear strong-field photoemission process occurs only when the optical electric field points away from the Au surface. Therefore, the asymmetric nano-triangles only emit electrons during positive half-cycles of the excitation pulse and, thereby, show a strong CEP sensitivity. The symmetric nano-rods, on the other hand, emit electrons in both positive and negative half-cycles, and accordingly, their CEP sensitivity is greatly reduced.

To confirm the CEP sensitivity, the phase of the emitter current at f_{CEO} = 2 kHz (measured on a lock-in amplifier) is measured as a BaF_2 wedge is moved across the SC pulse train (Fig. 2c). The wedge is moved in 2.5 mm increments every 10 s. With a carrier wavelength of 1.2 μm, each increment should result in a ~ 56° phase-shift to the CEP of the SC pulses. This shift should then appear as a phase-shift on the lock-in amplifier. The experimental results for the nano-triangle devices very closely resemble this prediction, as denoted by the orange line in Fig. 2c.

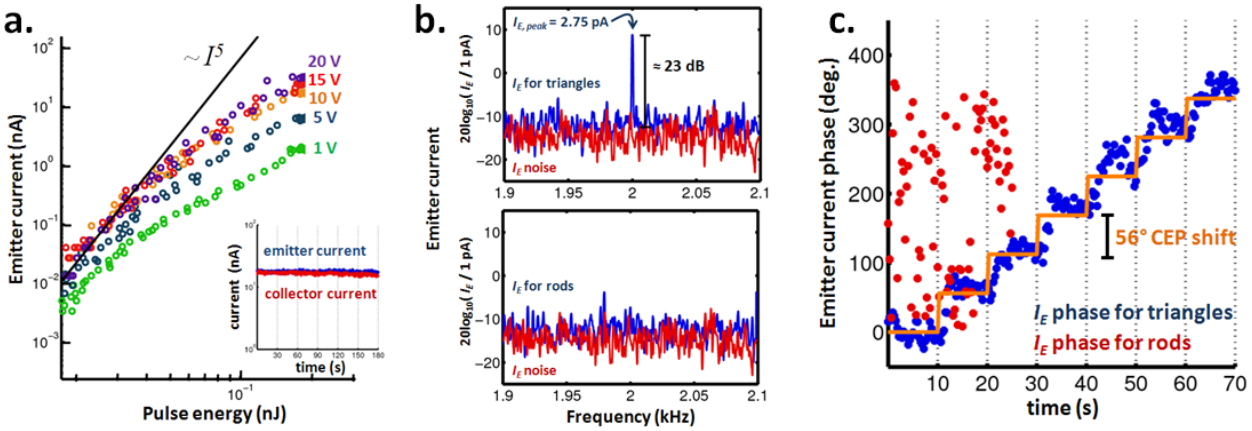

Figure 2: a. Strong-field emission and emitter / collector current stability. **b.** RF spectra (1 Hz resolution-bandwidth) of emitter current (I_E) from nano-triangle devices (top) and nano-rods (bottom). The red traces show the emitter current (I_E) without laser excitation. **c.** Emitter current (I_E) phase shifts for the nano-triangle devices (blue data points) and the nano-rod devices (red data points, only shown for the first 25 s).

In summary, we have shown strong-field photoemission from plasmonic nanoparticles across few-micron gaps under ambient conditions and at a high repetition rate. We have also demonstrated that the photoemission current consists of a strong carrier-envelope phase-sensitive component. These results are encouraging for the prospects of steering and manipulating attosecond electrons on a chip as well as for future compact, solid-state CEP detectors.

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