Optical-Field-Controlled Photoemission from Plasmonic Nanoparticles with a Sub-Two-Cycle, 6 nJ, Octave-spanning Ti:sapphire Oscillator

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Abstract. We developed an optimized, carrier-envelope-phase-stable Ti:sapphire oscillator that generates 6 nJ pulses compressible to under sub-two- optical-cycles in duration. We used the optimized oscillator to demonstrate carrier-envelope-phase-sensitive (CEP-sensitive) photoemission from metallic nanoparticles in the near-infrared.

1 Introduction

Strong-field light-matter interactions driven by few-cycle visible to near-infrared light pulses enable the study of matter's electronic response with sub-optical-cycle, i.e. attosecond, precision [1,2]. Strong-field interactions with solids typically require field strengths >1 GV/m; such fields are typically achieved by focusing high energy (μJ- to mJ-level) CEP-locked amplifier systems. In recent years, it has been shown that with plasmonic nanoantennas, one can locally enhance the optical field strength and reduce the required optical pulse energy for achieving strong-field light-matter interactions by two to three orders of magnitude [2]. Thereby, strong-field interactions can be driven by nJ-level laser pulses from unamplified, mode-locked oscillators at MHz-level repetition rates.

Using laser oscillators to directly generate few-optical-cycle nJ-level pulses is challenging. Octave-spanning spectra with <3-nJ pulse energy have been generated from Ti:sapphire oscillators [3,4]; however, the performance and applicability of such oscillators depend on the quality of the highly resonant cavities (typically with 1% output coupling) based on double chirped mirror pairs (DCMPs) [3,4]. Until now the octave-spanning spatiotemporal dynamics in Ti:sapphire oscillators have not been completely understood and optimized [3,4,5]. In this work, we firstly developed a numerically optimized

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Ti:sapphire oscillator capable of generating carrier-envelope-phase(CEP)-stabilized few-cycle pulses with ≈ 6 nJ pulse energy at an 85 MHz repetition rate. Secondly, after locking this source's carrier-envelope-offset frequency, denoted as f_{CEO}, to a stable 100 Hz reference, we used it to study CEP-sensitive, optical-field-controlled photoemission from an array of tailored metallic nanoantennas. Our results build on a previous work where we demonstrated optical-field-controlled photoemission from nanoantenna arrays using an amplified Er:fiber-based supercontinuum source centered around 1.2 μm [2]; this source was limited to sub-nJ pulse energy due to the use of highly nonlinear fibers [2,6]. Our results verify that the underlying emission physics scales to visible to near-IR optical frequencies, and demonstrate the potential for optimized Ti:sapphire oscillators to drive on-chip strong-field photoemission for both scientific research and technical applications.

2 Experimental demonstration

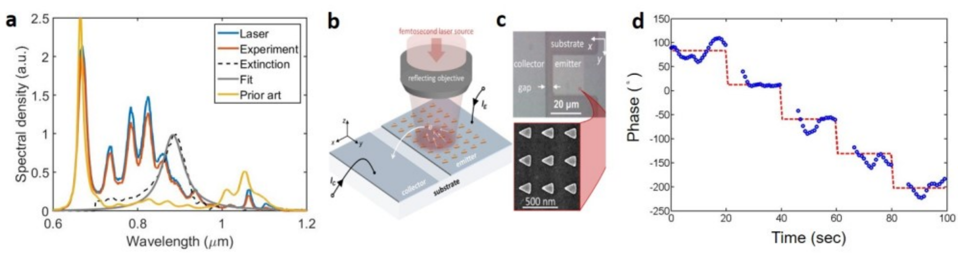

Fig. 1. (a) Output spectrum from the optimized laser cavity (blue) and experimental spectrum (red) illuminated on the nanotriangle arrays, compared with the previous state-of-the-art output (yellow) supporting the generation of 3.6fs-TL pulses, and measured extinction spectrum (gray) for the nanotriangle arrays (with damped harmonic oscillator fits (black, dashed line)). (b) Experimental setup and (c) microscope image of tip array emitter and collector contacts (top) and SEM of nanotriangles exhibiting laser surface plasma resonances near 895 nm (bottom). (d) Dependence of the phase of the CEP current signature measured at 100 Hz while inserting a BaF_2 wedge by 1.5 mm every 20 sec (blue, circle). An average CEP phase shift of 72° (red, dashed line), is consistent with our source spectrum. The ability to precisely control this phase via the wedge insertion is a true indication of CEP controlled current.

To optimize our Ti:sapphire oscillator, we employed an extended three-dimensional model to capture its complete spatiotemporal features. Using DCMPs, one can design the cavity to generate pulses with transform-limited (TL) durations down to 3.6 fs (represented by the yellow spectrum in Fig. 1a). However, higher residual dispersion oscillations generate satellite pulses that hamper the onset of stable mode locking. Using complimentary designs of cavity end mirrors and output coupling windows, a phase-optimized cavity was designed and fabricated with <0.1 rad of intracavity residual phase oscillation over the whole operating bandwidth of 0.65 – 1.14 μm. By further shaping the cavity resonance and removing residual phase oscillations from DCMPs, the laser output can be tailored for different applications, e.g. simultaneous spectral enhancement around 570 and 1140 nm for better CEP stabilization via intracavity phase matching [5]. After optimization, we achieved 500 mW average output power at a repetition rate of 85 MHz. We obtained >10 dB enhancements in power both (1) around the center gain region, improving the capability as a driving source for the strong-field experiments, and (2) at 1140 nm, increasing the frequency-doubled yield required for precise, self-referenced CEP locking. For the generation of even shorter pulses, we also operated the oscillator with a broader spectrum supporting 3.9 fs TL pulses, and pulse compression down to 4.3 fs duration was characterized by two-dimensional shearing interferometry (2DSI). Fig. 1a shows an experimental spectrum (blue) from the optimized cavities with 5% output coupling compared with the prior-art spectrum (yellow). Aside from the oscillator development, we implemented a newly designed self-referencing *f-2f* scheme that (1) is capable of freely controlling the locked CEP frequency without laser repetition rate dependence and (2) allows for easy spatial and temporal alignment between the fundamental and frequency-doubled light.

Next, we demonstrated optical-field-controlled photoemission using our optimized source. We illuminated an array of triangular nanoantennas (see Fig. 1b) having a plasmonic resonance centered around 895 nm (see experimental spectrum, as well as extinction spectra in Fig. 1a, and device image in Fig. 1c). Using a reflective objective, the laser pulses were focused to a spot of roughly 7.8 μm by 4.6 μm FWHM, allowing us to estimate a peak intensity before enhancement on the order of 10^{11} W/cm^2 at 88 mW incident power. After locking f_{CEO} to a stable oscillator at 100 Hz, we were able to measure the CEP-sensitive current directly after a low-noise transimpedance amplifier. The spectral response of the output current shows a clear current signature, corresponding to a peak current of roughly 1.7 pA with a signal to noise ratio (SNR) of around 30 using a resolution bandwidth (RBW) of 300 mHz. Finally, to verify optical-field control of the CEP-dependent current, the phase of the 100 Hz photocurrent signal was measured via lock-in detection while inserting a BaF_2 wedge into the exciting optical beam. The wedge was inserted by 1.5 mm every 20 seconds, resulting in a phase shift of $\approx 72^{\circ}$ per step consistent with a central wavelength near 860 nm (see Fig. 1d). This result verifies that the current is indeed controlled via the optical field.

We have demonstrated octave-spanning pulses from a Ti:sapphire oscillator with high spectral power densities, allowing for close to single-cycle pulses by external shaping the output spectra. We also investigated CEP-sensitive photoemission from an array of plasmonic nanoantennas. Current studies are underway to determine optimal conditions for high SNR CEP detection using the nanoantennas by tuning pulse compression, beam spot size, and the central wavelength of the nanoantennas' plasmonic resonance. With the higher pulse energies available using our Ti:sapphire system, we are no-longer limited to few-micron spot sizes to achieve strong-field photoemission, and we anticipate that at least an order of magnitude improvement in CEP-sensitive current is within reach. Our high-energy CEP-locked Ti:sapphire oscillator design enables the development of novel lightwave/PHz electronic technologies and fundamental investigations of strong-field light-matter interactions throughout the visible to near-IR spectrum.

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