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Time-resolved photoemission electron microscopy of a plasmonic slit resonator using 1 MHz, 25 fs, UV-to-NIR-tunable pulses

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Abstract. We discriminate different field dynamics across distances as small as 33 nm within a plasmonic slit resonator using aberration-corrected photoemission electron microscopy and a tunable broadband optical parametric amplifier at 1 MHz repetition rate.

1 Introduction

Tailored optical antennas made from single crystalline gold substrates are known to support localized surface plasmon modes (LSP) [1] which give rise to electromagnetic field dynamics on nanometer length scales and femtosecond time scales [2]. Exposing single or few quantum systems (e.g., molecules or quantum dots) to this local field distribution, the detection of the altered LSP properties is of high interest for sensing and spectroscopy applications. Conventional optical methods offer the required few femtosecond temporal resolution, but the spatial resolution is limited by optical diffraction. Employing ultrafast optical excitation and high-resolution photoelectron imaging capabilities, time-resolved photoemission electron microscopy (TR-PEEM) is applicable for the investigation of dynamical phenomena on the nano–femto scale [3,4]. However, conventional TR-PEEM experiments using oscillator lasers offer only little wavelength flexibility in the visible, while 1 kHz laser systems suffer from poor counting statistics or space-charge image distortion.

To resolve these drawbacks, we present the combination of aberration-corrected PEEM and fully tunable broadband excitation pulses rendered by noncollinear optical parametric amplification (NOPA) at 1 MHz repetition rate. Different delay dependencies of the multiphoton photoemission yield across a mean distance of 33 nm are resolved in a phase-resolved autocorrelation experiment with a spatial resolution of 8.8 nm ± 1.0 nm upon laser excitation.

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2 Experimental Setup

A two-branch NOPA is pumped with the second and third harmonics of an Yb-doped fiber laser (Amplitude Systèmes, Tangerine HP) at 1030 nm with a pulse energy of 20 µJ and pulse duration of ~320 fs at a repetition rate of 1 MHz. Tunable pulses in the range of 400 nm – 650 nm and 630 nm – 970 nm can be generated by pumping with the third and second harmonics, respectively, while simultaneous emission of the two branches is possible [5]. By second harmonic generation of the NOPA output, pulses in the range of 215 nm – 400 nm (5.8 eV – 3.1 eV) are available (Fig. 1, inset, for exemplary spectra throughout the available total regime). Output pulses are compressed typically to sub-25 fs with a prism compressor.

For the specific application here, a pair of phase-coherent and time-delayed pulses (700 nm, 22 fs, 12 nJ each) is generated with an actively stabilized Mach-Zehnder-type interferometer and focused into the main PEEM ultrahigh vacuum chamber (4×10^{-11} mbar base pressure) in normal incidence to the sample surface. Electrons emitted through a multiphoton absorption process are spatially imaged with aberration-corrected PEEM (Elmitec) as a function of the inter-pulse delay (Fig. 1).

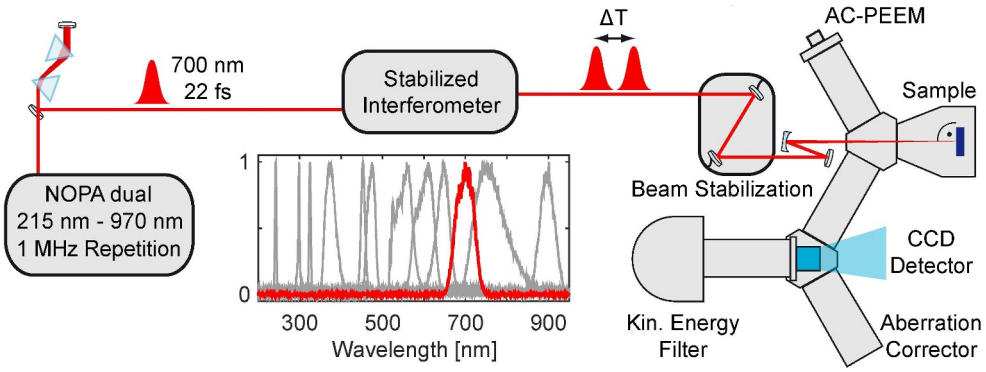

Fig. 1. Experimental setup: Phase-coherent pulse pairs are generated from a two-branch noncollinear optical parametric amplifier (NOPA) and an actively stabilized Mach-Zehnder-type interferometer. Representative NOPA spectra (gray) are shown as well as the 700 nm spectrum used in the experiment (red). The spatial distribution of emitted electrons is detected with aberration-corrected photoemission electron microscopy following active beam stabilization.

As an LSP model system, we fabricated plasmonic slit resonators of different lengths and widths at the edges of a chemically grown single-crystalline gold flake using ion-beam milling techniques. A scanning electron micrograph of the sample topology is shown in Fig. 2 (left).

3 Results

Pulses tuned to 700 nm and a polarization perpendicular to the long axis of the slit excite an LSP mode as predicted by finite difference time domain (FDTD) simulations, showing in Fig. 2 (second column) the simulated local electric field intensity distribution inside the slit for a single cross-section of the geometry of interest. The local plasmonic field strongly enhances non-linear photoemission processes, i.e., three photons are necessary to overcome the substrate work function. The experimentally obtained one-pulse photoelectron image (Fig. 2, third column) resembles the plasmonic mode distribution and interference with the

external field. Taking the complex near-field distribution at all facets into account, two major electron emission spots are expected in correspondence to the experimental data.

The dynamics of the most intensive photoemission signatures with a spatial distance of ~200 nm reveals out-of-phase oscillatory behavior in the range of 28 fs – 39 fs inter-pulse delay (faint blue and faint red in Fig. 2, right). Moreover, we show similar dephasing at adjacent cross-line locations with a mean spatial distance of 33 nm in the range of 28 fs – 34 fs (dark blue and dark red). The possibility to disentangle LSP dynamics on such small length scales is an important prerequisite for the exploration of interactions with close-by quantum systems. For the system investigated here, time-resolved FDTD simulations reveal a complex local phase behavior with strong excitation wavelength dependency which is, in general, accessible with our tunable laser source.

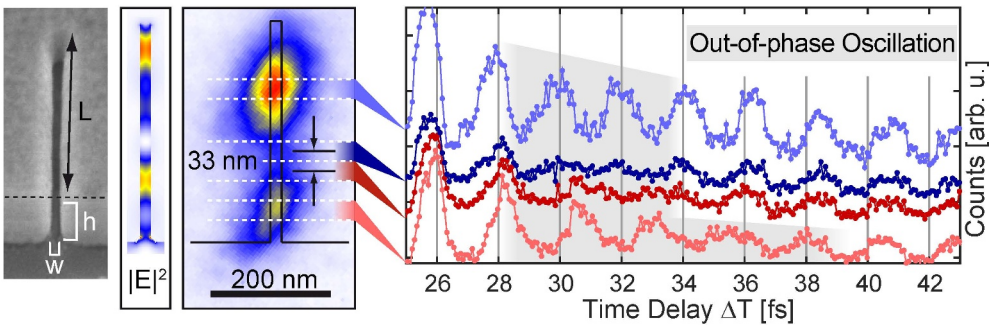

Fig. 2. From left: Scanning electron micrograph of a sample slit with L = 360 nm, h = 31 nm, w = 19 nm; simulated field intensity distribution from finite difference time domain simulations with the same geometrical parameters; photoemission electron microscopy image. Horizontal dashed lines indicate cross-line ranges for delay-dependent PEEM data (right).

4 Conclusion

We introduce a time-resolved aberration-corrected photoemission electron microscopy setup using broadband tunable excitation pulses at 1 MHz repetition rate. Our approach allows us to disentangle local field dynamics across so far unprecedented small spatial distances when using all-parallel image acquisition techniques. Future applications will target the investigation of hybrid nanostructures consisting of plasmonic nanoresonators and single quantum systems with nonlinear multi-dimensional spectroscopy techniques.

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