

Plasmon-driven ultrafast point-projection electron microscopy

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Abstract. We implement a plasmon-driven ultrafast electron source in a point-projection electron microscope. A proof-of-principle experiment investigating the charge propagation in a single nanoresonator demonstrates an unprecedented spatiotemporal resolution of 20 nm and 25 fs.

1 Introduction

Directly watching localized plasmonic fields on their natural few-nm length and few-fs time scales is an open challenge for today's ultrafast microscopy techniques. Although significant progress has been made in, for example, ultrafast electron microscopy, the desired resolution has not been reached yet. So far, the chromatic dispersion of short photoemitted electron pulses prevents a temporal resolution much below 100 fs [1,2].

Recently, we developed a plasmon-driven electron source, capable of delivering single electron pulses originating from a few-nm, few-fs spatiotemporal volume without the requirement of a micrometer-sized laser focus at the source position [3]. Compared to direct electron emission with a short laser pulse, plasmon-driven electron emission turned out to be 50 times more efficient. This is achieved by efficiently focusing optical energy via a propagating surface plasmon on a tapered gold wire, leading to a concentration of the surface plasmon at the taper apex, followed by multiphoton electron emission.

Here, we implement this electron source in a point-projection electron microscope and perform a first pump-probe experiment. We demonstrate that the source permits source-sample distances of 1 μm and below to efficiently prevent temporal electron pulse broadening, resulting in a temporal resolution of less than 20 fs. The resolution is deduced from the ultrafast transmission change of the probing electrons when they pass a single nanoresonator after optical excitation so that a small charge cloud is emitted [4].

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2 Results and discussion

Ultrashort carrier-envelope phase stable laser pulses with a pulse duration of 16 fs at a central wavelength of 1700 nm are generated by two home-built optical parametric amplifier laser systems with subsequent difference frequency generation [5]. A schematic of the experimental setup is shown in Fig. 1a. A first laser pulse excites a surface plasmon polariton (SPP) on a sharply etched metallic taper. Starting at a distance of 80 μm from the taper apex, the SPP experiences a shrinking taper diameter and hence an increasing effective refractive index. This concentrates the SPP into a nanometric volume, solely defined by the sharpness of the taper. At the taper apex, the surface plasmon's electric field becomes strong enough to induce ultrafast localized multiphoton electron emission.

The emitted electron wavepackets, consisting of a single electron on average, diverge spatially and interact with a sample, positioned 2700 nm in front of the taper. In a classical picture, a fraction of the electrons is absorbed or scattered by the sample. A second laser pulse excites the sample with variable time delay, leading to additional forces acting on the probing electrons. The electrons transmitted through the sample plane propagate towards an electron detector in a distance of 75 mm. Here, a magnified shadow image of the sample is formed. Furthermore, the ultrafast charge dynamics in the sample are imprinted into additional modifications of the shadow image.

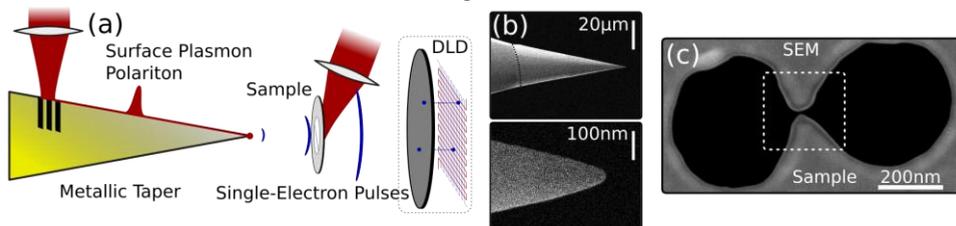


Fig. 1. a) Schematic of the experimental setup, showing the metallic taper, the sample and the delay line electron detector (DLD). b) Scanning electron microscope (SEM) images of a sharply etched gold taper, showing an apex radius of 15 nm and a grain boundary in a distance of 80 μm to the taper apex. c) SEM image of the 30 nm-thin gold film sample.

Here, in a proof-of-principle experiment, a double nanohole structure milled into a 30 nm thin freestanding gold film is used as the sample. The second laser pulse, polarized along the vertical direction in Fig. 1c, photoemits approximately 30 electrons per laser shot in the 30 nm-small gap region. Depending on the temporal delay between the generation of the electron cloud and the probing electron pulse, different expansion states of the cloud can be recorded. This is shown in Fig. 2a. A pronounced reduction of the electron transmission in the central gap region within the first 100 fs after optical excitation can be observed. For later times, the electron cloud expands further, becoming more and more transparent for the probing electrons. After 300 fs, the static image is almost completely recovered.

We use this series of images to deduce the spatiotemporal resolution of our microscope. First, we determine the spatial resolution by analyzing a micrograph recorded while the excitation laser was off. A cross cut along a horizontal line (Fig. 2b) shown in Fig. 2c reveals a resolution of 20 nm. By examining all recorded images at certain positions on the white arrow shown in Fig. 2b, we infer a temporal resolution of less than 25 fs, given by the decay of the electron transmission from 90% to 10% of its minimum. This is, to the best of our knowledge, a significant improvement by a factor of 4 compared to the temporal resolution reached so far in ultrafast electron microscopy experiments [1,2]. With this unique new technique, we trace the ballistic motion of electrons that are ejected from the plasmonic hot spot of the nanohole antenna with unprecedented resolution. We can directly

see the spreading of the electron cloud and extract quantitative information about the released electron wavepacket such as their momentum and kinetic energy distribution.

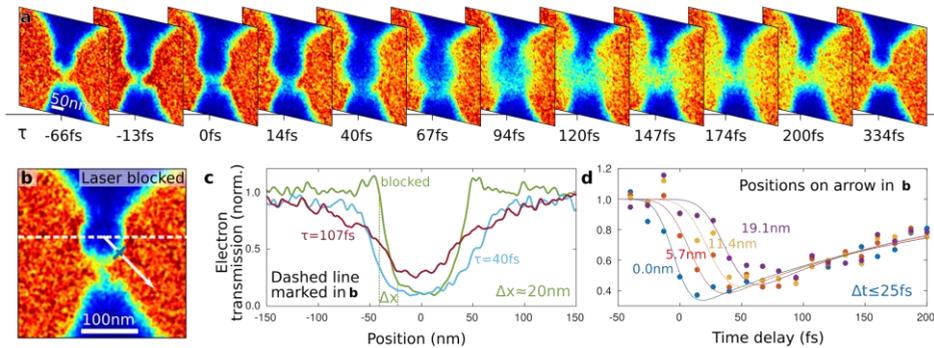


Fig. 2. a) A series of point-projection electron microscope images of the central gap region for different pump-probe time delays. A clear reduction of electron transmission in the middle of the image can be observed for time delays of up to 100 fs, which is fully restored within 300 fs. b) Reference image recorded with a blocked pump laser. c) Cross cuts along the dashed line in b) for different time delays, giving an upper limit for the spatial resolution of the microscope of 20 nm. d) Temporal evolution of the electron transmission for different positions on the white arrow shown in b), starting at the blue cross. This gives an upper limit for the temporal resolution of 25 fs.

3 Summary and Outlook

In summary, we demonstrate electron microscopy with so far unrivaled spatio-temporal resolution on a single nanostructure. We use this to record the ultrafast expansion of a charge cloud in a single nanostructure. This new plasmon-driven point-projection electron microscope can, in a next step, be used to perform electron holography [6] with ultrahigh temporal resolution. Furthermore, energy-resolved detection of shadow images or holograms, as already depicted in Fig. 1a and implemented in our laboratory, will permit the imaging of local plasmonic fields, similar to PINEM experiments [7], but with a temporal resolution in the 10 fs regime and below. With this, streaking experiments in nanolocalized near-fields can be performed, bringing the capture of coherent field oscillations in nanometric systems into direct reach.

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