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Ultrafast Transmission Electron Microscopy with nanoscale Photoemitters

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Ultrafast transmission electron microscopy (UTEM) is a rapidly developing technique which aims to elucidate laser-triggered ultrafast processes at the nanometer scale [1]. In UTEM, ultrafast imaging and spectroscopy with sub-picosecond temporal resolution is achieved by utilizing electron pulses in a stroboscopic laser-pump/electron-probe scheme (Fig 1a). The previously employed flat-surface photocathodes put a severe limit on the achievable spatial resolution due to their intrinsically large electron source size, demanding novel pulsed electron source concepts.

Here, we report on the development and first applications of a novel UTEM instrument at the University of Göttingen which incorporates a tip-shaped photocathode for the generation of high-quality electron pulses [2-4]. Specifically, in a modified JEOL JEM-2100F microscope, we obtain localized single-photon photoelectron emission from the apex of a ZrO-coated tungsten tip using 3.1 eV optical pulses. Using the local depression of the work function due to the Schottky effect, photoelectron emission is largely confined to the front facet of the crystalline tip. At the sample position, the laser-triggered electron pulses can be focused down to 3 nm spot sizes (Fig.1b, inset), which results in a root-mean-square (rms) emittance of less than 4 nm mrad.

The temporal structure of the electron pulses is probed by electron-photon cross-correlation, utilizing inelastic electron scattering in laser-driven optical near-fields [5] around a gold nanotip. An electron traversing an optical near-field experiences an energy gain or loss in multiples of the photon energy (Fig. 1b,d), giving rise to multiple photon sidebands in the transmitted electron energy spectrum. Varying the temporal delay between the optical driving pulse and the arrival time of the electron bunch at the near-field, allows to map the temporal bunch shape. For, on average, less than one electron per pulse, electron-photon cross-correlation results in a temporal width of the electron pulses as small as 300 fs (Fig. 1c, upper panel). At larger bunch charges, space-charge induced spectral and temporal broadening sets in, leading to a strongly correlated chirped electron pulse structure (lower panel, Fig. 1c).

Low-emittance ultrashort electron pulses, as reported here, are particularly suited for time-resolved imaging applications which require, either, large transverse coherence lengths, as for example in Lorentz microscopy and electron holography, or small probe sizes. In a first application of nanoscale probing, we investigated quantum interference effects in the inelastic electron near-field scattering [6]. In particular, the population of certain photon sidebands is almost completely suppressed for specific driving field strengths. We quantitatively describe these scattering spectra taking into account quantum path interferences between different photon absorption/emission sequences which lead to the same final state.

In conclusion, we report on the development of a novel ultrafast transmission electron microscope employing a tip-shaped photo-emitter. Implications of coherent near-field scattering processes for the optical control of free electron states are discussed.

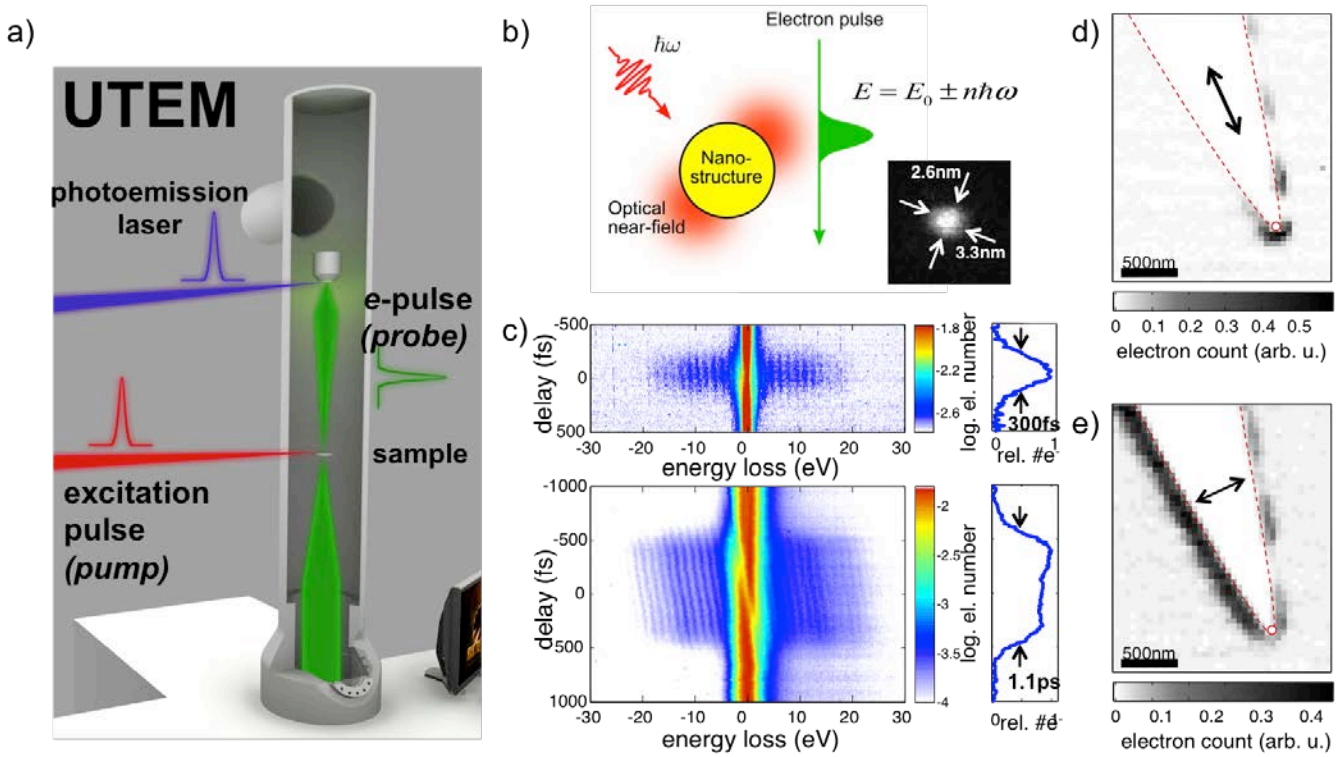

Figure 1. a) Laser-pump/electron-probe scheme. Ultrashort electron pulses are generated by photoemission and probe at well-defined temporal delays the state of a sample after laser excitation. b) Schematics of inelastic electron scattering in the laser-driven near-field of a nanostructure. Inset: achievable electron spot diameter employing laser-triggered Schottky field emission. c) Electron energy spectra after inelastic near-field scattering for single-electron (top panel) and multi-electron pulses (bottom panel). Photon-order sidebands are only observed for temporally overlapping photon and electron pulses at the optical near-field around a gold nanotip, giving rise to the electron-photon cross-correlation traces shown on the right. (d-e) Raster-scan of the intensity of gain-scattered electrons imaging the mode structure of the laser-driven near-field. The driving laser field is polarized either in the tip direction (d) and perpendicular to the tip direction (e). Broken red line: geometrical contour of the nanotip.

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