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Ultrafast transmission electron microscopy using a laser-driven field emitter: Femtosecond resolution with a high coherence electron beam

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ABSTRACT

We present the development of the first ultrafast transmission electron microscope (UTEM) driven by localized photoemission from a field emitter cathode. We describe the implementation of the instrument, the photoemitter concept and the quantitative electron beam parameters achieved. Establishing a new source for ultrafast TEM, the Göttingen UTEM employs nano-localized linear photoemission from a Schottky emitter, which enables operation with freely tunable temporal structure, from continuous wave to femtosecond pulsed mode. Using this emission mechanism, we achieve record pulse properties in ultrafast electron microscopy of 9 Å focused beam diameter, 200 fs pulse duration and 0.6 eV energy width. We illustrate the possibility to conduct ultrafast imaging, diffraction, holography and spectroscopy with this instrument and also discuss opportunities to harness quantum coherent interactions between intense laser fields and free-electron beams.

1. Introduction

The continuing advancement of electron microscopy within physics and chemistry, materials science, and structural biology [1–3] provides us with ever-increasing precision in viewing structure and composition on the nanoscale. A detailed microscopic understanding of the structural, electronic and magnetic properties of natural and synthetic materials demands - besides atomic-scale spatial characterization - the investigation of the response of these systems to external perturbation. The growing importance of in-situ approaches in transmission electron microscopy [4], scanning electron microscopy [5], X-ray diffraction [6], scanning tunneling and atomic force microscopy [7], and other areas testify to this development.

Time-resolved experiments, following the dynamical response of a system to a pulsed excitation, represent an especially powerful form of in-situ probing, which yields direct time-domain access to the character and strengths of the couplings between structural, electronic and spin degrees of freedom. Ultrafast electron [8–13] and X-ray [14–17] diffraction are well-established techniques to track structural relaxation with femtosecond temporal resolution, widely applied to homogeneous and thin film systems. The observation of spatiotemporal relaxation processes in heterogeneous systems [18–23], however, such as excitation and energy transfer across functional interfaces, is particularly challenging, requiring simultaneous nanoscale spatial and ultrafast temporal resolutions. To this end, various experimental approaches are pursued very actively at present, including time-resolved variants of scanning tunneling microscopy (STM) [24–26] and scanning near-field optical microscopy (SNOM) [27–29]. Furthermore, imaging techniques using ultrashort electron pulses such as compact point-projection electron imaging [30–33] and ultrafast scanning electron microscopy [34–38] are being developed.

Beyond these approaches, ultrafast transmission electron microscopy (UTEM) promises to become one of the most powerful experimental tools for the investigation of ultrafast dynamics on the nanoscale, joining femtosecond temporal resolution with the vast opportunities in imaging, diffraction and spectroscopy provided by state-of-the-art electron optics. Early pioneering works at the Technical University Berlin [39], Caltech [40] and Lawrence Livermore National Labs [41] demonstrated the feasibility of pump-probe studies in electron microscopy, either in a stroboscopic fashion [42] or using single-shot imaging [43]. Motivated by various notable individual results highlighting its broad potential, time-resolved electron microscopy is currently explored in a growing number of laboratories worldwide [44–50].

Being considered one of the most exciting frontiers in electron microscopy, the area of ultrafast transmission electron microscopy is

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presently at a pivotal moment of its development. Facing great challenges in obtaining intense high-quality electron pulses, time-resolved electron microscopy is in particular need of benchmarking the currently achievable spatio-temporal resolution limits and electron beam figures-of-merit. Quantitative characterizations will be required to facilitate systematic progress and to connect this emerging field to the well-established, powerful experimental and theoretical framework of electron microscopy [51,52]. In order to harness the full imaging and spectroscopy capabilities of today's electron microscopes, it is highly desirable to integrate higher-brightness pulsed electron guns into the electron optics environment of a transmission electron microscope, in particular based on laser-triggered field emitter concepts. Not unlike the scientific leaps that are associated with technological breakthroughs in bright continuous electron sources in the past [53,54], significant advances in pulsed electron source quality promise a path to uncharted territory in ultrafast nanoscale dynamics.

In this contribution, we describe the first implementation of an ultrafast transmission electron microscope based on laser-triggered electron emission from a nanoscale photocathode (Ch. 2). We provide a quantitative characterization of the spatial and temporal electron beam properties for a variety of electron-optic illumination conditions (Ch. 3), demonstrating electron pulse durations down to 200 fs, energy widths of 0.6 eV and a focusability of the photoelectron beam to sub-nm dimensions. We illustrate a range of possible applications for this instrument, which include bright- and dark-field imaging, convergent beam electron diffraction (CBED) from nanoscale areas, phase-contrast imaging and Lorentz microscopy, holography and spatially-resolved electron spectroscopy (Ch. 4). Beyond adding femtosecond temporal resolution to this set of conventional electron microscopy techniques, the advanced electron beam properties of the field-emitter UTEM render it ideally suited to be applied in contrast mechanisms and phenomena that are exclusive to ultrafast electron microscopy, such as photon-induced near-field electron microscopy (PINEM) or the quantum coherent manipulation of free-electron beams (Ch. 5).

2. Instrumentation

Ultrafast transmission electron microscopy is a stroboscopic imaging technique, in which dynamics in an investigated sample are triggered by short (typically optical) excitation pulses. At well-defined delay times after excitation, the evolving state of the sample is probed by an ultrashort electron pulse (Fig. 1a). Accumulating, for a given delay time, the signal derived from many electron pulses yields a stroboscopic snapshot of the transient state of the system [8,40]. Importantly, the temporal resolution of such a pump-probe approach is given by the electron pulse duration and is not limited by the speed of the electron detector. In the past, employing photoemission driven by ultrashort laser pulses has enabled the generation of electron bunches with femtosecond duration, which are now finding increasing use in time-resolved electron imaging, diffraction and spectroscopy techniques [8,10–12,20,40,44,55–57].

The Göttingen UTEM instrument is based on a JEOL JEM-2100F transmission electron microscope, which was modified to allow for both optical sample excitation and pulsed electron emission. In contrast to previous implementations of time-resolved TEM, we employ a laser-driven Schottky emitter, which confines the photoemission to the nanoscale front facet of a ZrO/W emitter tip [37,58]. The emitter is side-illuminated with 400 nm laser radiation, focused to a spot diameter of about 20 μm full-width-at-half-maximum (FWHM). Optical access to the emitter tip is given through a side window on the TEM gun and by a further optical steering assembly inside the ultrahigh vacuum chamber of the electron source. For time-resolved experiments, frequency-doubled femtosecond optical pulses from a regenerative Ti:Sapphire amplifier (Coherent RegA) are used at pulse energies of about 10 nJ and at a tunable repetition rate of up to 800 kHz. For alignment of the photoelectron beam into the TEM electron optics, and for characterization of electron beam properties in the space-charge-free regime, a continuous diode laser is employed at an average optical power of typically 20 mW. Utilizing the usual field geometry of a continuous Schottky source, the laser-triggered emitter is placed into an electrostatic suppressor-extractor electrode assembly, characterized by a dimensionless parameter Γ [59], which allows for tuning the extraction field at the emitter apex and the divergence of the photoelectron beam. (For further details on tailoring photoelectron beams in a Schottky emitter assembly, see Ref. [59]). Electrons far from the optical axis are cut by a hard aperture, which is placed in the electrostatic gun lens. By changing the voltages applied to these three electrodes, the electron gun can be operated in different modes, e.g., optimized for a high electron yield or a high beam coherence (cf. Ch. 3.2). Finally, after acceleration up to 200 keV, the probing electron beam is formed by the condenser system of the TEM column.

For optical sample excitation, we devised two optical beam paths by inserting mirror assemblies into the TEM column. First, access through a port conventionally used for adding an energy-dispersive X-ray spectrometer allows for optical excitation at an angle of incidence of 55° relative to the electron beam. Second, excitation close-to-parallel to the electron beam is provided by an illumination through the objective lens pole piece. For both cases, typical optical focal spot sizes on the sample at a central wavelength of 800 nm are about 50 μm FWHM. The delay time of optical excitation, optical fluence, polarization state and

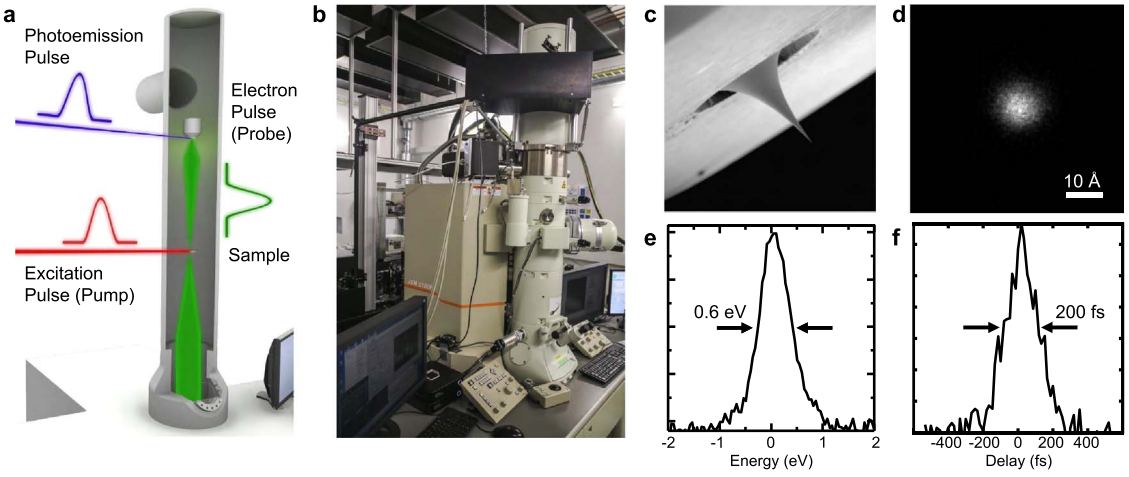

Fig. 1. Schematic setup and electron pulse properties of the Göttingen UTEM instrument. A laser-driven Schottky field emission electron gun (a) is combined with the column of a JEOL JEM-2100F (b). Side illumination of a nanoscopic ZrO/W(100) tip emitter (c) enables the generation of ultrashort electron bunches, which can be focused down to 0.89 nm (d), with an energy width of 0.6 eV (e) and a duration of 200 fs (f) (apertured beam, at 200 kV acceleration voltage).