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Development of a high brightness ultrafast Transmission Electron Microscope based on a laser-driven cold field emission source

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ABSTRACT

We report on the development of an ultrafast Transmission Electron Microscope based on a cold field emission source which can operate in either DC or ultrafast mode. Electron emission from a tungsten nanotip is triggered by femtosecond laser pulses which are tightly focused by optical components integrated inside a cold field emission source close to the cathode. The properties of the electron probe (brightness, angular current density, stability) are quantitatively determined. The measured brightness is the largest reported so far for UTEMs. Examples of imaging, diffraction and spectroscopy using ultrashort electron pulses are given. Finally, the potential of this instrument is illustrated by performing electron holography in the off-axis configuration using ultrashort electron pulses.

1. Introduction

Since its invention in 1931, Transmission Electron Microscopy (TEM) has allowed giant steps in our fundamental understanding of many fields of science including chemistry, biology or physics. Many quantitative techniques have been developed to get structural and chemical information either from imaging (direct space), diffraction (reciprocal space) or spectral analysis (energy space). Three types of electron sources can be found in TEMs: thermionic, Schottky and cold-field emission sources. In a thermionic electron source, the electrons are pulled out of a cathode by thermal excitation produced by a strong electric current. In Cold-Field Emission (CFE) electron sources, no heating of the cathode is required. An extraction voltage is instead applied on a sharp conical metallic tip. This voltage, enhanced by the lightning rod effect at the tip apex lowers the potential in vacuum enough to allow for efficient tunneling of electrons out of the tip from the Fermi level. The very small size of the emitting zone is at the origin of the high brightness of field-emission electron sources. The development of field emission guns has allowed TEM to enter in a new era [1]. Following the initial suggestion of Gabor in 1948, new techniques such as off-axis electron holography have started to exploit the high spatial coherence of the electron beam to produce interferograms from which modifications of the phase of the electronic wavepacket can be retrieved [2]. These studies have shown that the phase of the electron wave is a very sensitive probe of the electrostatic field, strain field or magnetic field which allows the quantitative mapping of these observables with nanometer resolution [3–6]. The brightness of the source, defined as the current per unit area and solid angle, is the figure of merit that must be optimized in order to get the highest spatial coherence or the highest spatial resolution for applications like holography or spatially resolved spectroscopies (in this latter case, the improved energy resolution of the cold FE sources is also very important). Schottky guns are halfway and use a field assisted thermionic emission process [7]. They are equipped with a so-called suppressor anode to confine the region from which electrons are emitted to the apex of the tip. For the most demanding applications, CFEGs are the first choice as they exhibit the highest brightness and the lowest energy spread thanks to their unique combination of small virtual source size and low angular current density.

Pioneering work at the Berlin Technical University [8,9] and Lawrence Livermore National Laboratory [10] have clearly established time-resolved Transmission Electron Microscopy as one of the most active line of instrumental developments in TEM. High speed Transmission Electron Microscopes or Dynamical Transmission Electron Microscopes have provided a unique insight into irreversible processes such as phase transitions, melting or ablation for instance [11,12]. Their spatio-temporal resolution was however limited by the large number of electrons in each pulse [13]. By using pulses containing only a few electrons, the group of A. Zewail at Caltech has overcome this limitation and performed time-resolved studies with both nanometer spatial resolution and sub-

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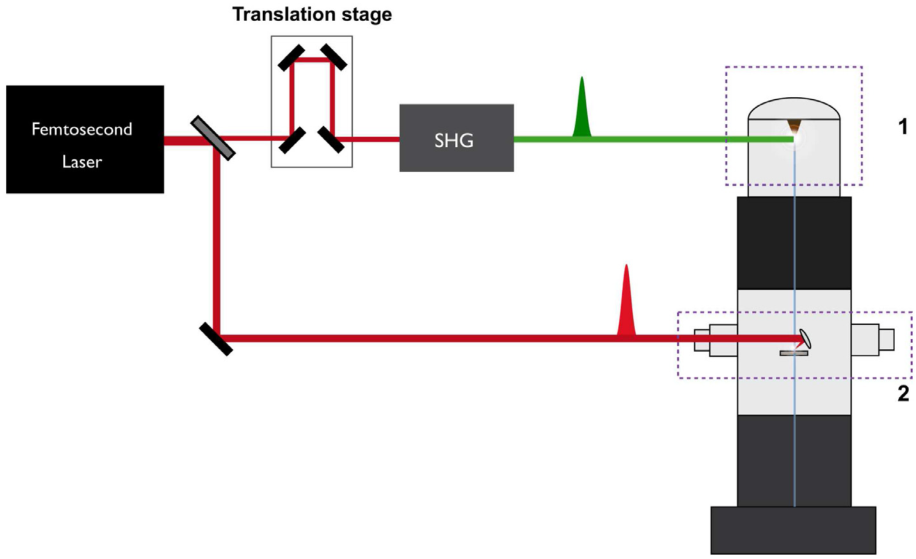

Fig. 1. Schematic of a time-resolved pump-probe TEM experiment. The emission of electron pulses is triggered by a laser pulse inside the electron gun of the microscope (1). The objective lens of the latter must be adapted to allow for light injection inside the column to excite the sample (2). SHG: Second Harmonic Generation.

picosecond temporal resolution [14,15]. Until recently, UTEMs were all based on flat photocathodes implemented in thermionic electron guns. The large size of the illuminated area on the photocathode was limiting the brightness of these electron sources and prevented their use for the most demanding TEM applications such as electron holography.

A decade ago, it has been shown that laser-driven nanoemitters could provide an exciting alternative to conventional photocathodes [16–18]. Due to the enhancement of the laser electric field at the apex, it is possible to trigger electron emission from a small region and investigate light-matter interaction in strong optical fields [19–22]. Processes such as electron rescattering which are at the heart of attosecond physics can be investigated with low power high repetition rate laser systems whereas similar studies on dilute systems demand amplified lasers [23]. The confinement of the emission region to strong field regions at the tip apex further yields a brightness in the laser-driven mode which is similar to the conventional DC mode and makes laser-driven nanoemitters very promising for ultrafast coherent electron microscopies [24]. The first implementation of a laser-driven field-emission tip in a TEM has been achieved in Göttingen on a Schottky-type electron source [25,26]. The spectacular improvement in brightness achieved has allowed to perform unique experiments which would not have been possible on the previous generation of UTEMs [27–29]. The development of an ultrafast electron source based on a cold-field emission gun has been recently demonstrated but its potential for ultrafast TEM has not been explored yet [30].

It is the purpose of this paper to describe the potential of such an ultrafast Transmission Electron Microscope based on a modified cold field emission source. We here follow a different line from the development achieved in Göttingen. The tight arrangement of Schottky sources makes the integration of short focal distance optics inside the electron source difficult. In this case, the optics used to focus the laser beam has therefore been placed outside the TEM column and the region from which electrons are emitted is restricted by the use of the additional suppressor electrode available on Schottky electron guns and/or by chemical selectivity using a zirconia wetting layer on the [100] oriented front facet of tungsten tips [7,29]. In the present work, we have modified the cold field emission source to integrate optics in the immediate vicinity of the field emission cathode to minimize the size of the laser focal spot on the tip apex, minimize the size of the emission region and therefore optimize the brightness of this new kind of ultrafast TEM electron source. Furthermore, CFE sources do not need to be heated to operate in continuous (DC) emission and therefore the proposed architecture can operate either in conventional DC or ultrafast mode and switching between the two modes is easily done by changing the extraction voltage. In the following, we present the design of the instrument, its performances and illustrate the potential of the new ultrafast source on a few TEM applications.

2. Development of an ultrafast Transmission Electron Microscope based on a Cold Field emission source

2.1. Accessing temporal resolution in TEMs

Most of ultrafast time-resolved TEM experiments are pump-probe experiments which involve an optical pulse and a delayed electron pulse [15]. As shown in Fig. 1, the optical *pump* pulse first brings the sample, located in the TEM objective lens, out of equilibrium and the electron *probe* pulse, delayed with respect to the excitation, is used to probe the sample during its relaxation. By systematically changing the delay between pump and probe, it is possible to record the dynamical evolution of the sample as it goes back to equilibrium. The delay between pump and probe can be controlled by moving the mechanical delay stage placed on one of the optical paths. In time-resolved TEM experiments, the temporal resolution depends on the laser pulse duration, electron emission characteristics (initial energy spread, number of electrons per pulse), and propagation inside the TEM (acceleration length, voltage...) [13]. As shown in Fig. 1, the generation of the electron probe pulse is triggered by a second optical pulse originating from the same laser source as the pump pulse and is therefore synchronized with the latter.

Time-resolved TEM experiments can be performed in two different modes. In the *single-shot mode*, only one electron pulse containing a sufficient number of electrons to yield an exploitable signal (image, diffraction pattern, spectrum) is used to probe the sample with a delay with respect to the excitation of the sample. This mode is used to investigate irreversible processes such as phase transitions for instance. The large number of electrons inside each electron bunch can have a detrimental effect on the temporal and spatial resolution [13]. Whereas flat photocathodes can yield electron bunches having more than 10^9 electrons per