Design of a New Ultrafast Transmission Electron Microscope

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In the past decades, spatial resolution of Transmission Electron Microscopes (TEM) has been improved down to the sub-Angstrom range. More recently, studies have begun to focus on *in-situ* experiments for imaging a variety of dynamic processes at the atomic scale. However, most of the nanoscale processes occur at very short time scales, in the micro- to femtosecond domain. Studies on such processes are limited by the acquisition frequency of the TEM detectors (~few milliseconds), thus missing the salient details of the sample dynamics, e.g. defect processes, phase transformations, or nucleation phenomena. For such studies, a much higher temporal resolution is required. This can be obtained using short electron pulses in a pump-probe approach. After excitation (pump) the sample is probed with an electron packet at a variable time delay with respect to the excitation. Repeating this process at different delays allows time-resolved studies.

These pulsed electron imaging studies can be carried out in two different operating modes:

- **Single shot mode**, required for studying irreversible processes, using high intensity pulses with a sufficient number of electrons ($>10^8$) to capture single-shot images of transient states in the material on nanosecond to microsecond timescales.

- **Stroboscopic mode** allows imaging and spectroscopy of reversible processes with high spatial and energy resolution through the accumulation of millions of low-intensity electrons pulses (1-100 electrons) at MHz repetition rates.

In this contribution, we present the setup and the first results of the new Ultrafast Transmission Electron Microscope (UTEM) developed at the IPCMS (Strasbourg, France). The microscope is the first instrument designed to operate in both stroboscopic and single-shot modes. At the present time, the microscope operates in stroboscopic mode, while the single-shot mode is under development.

The equipment is based on a JEOL 2100 transmission electron microscope combined with a femtosecond fiber laser. The initial infrared laser beam (1030 nm, pulse length 370 fs) is split into two beams. The pump beam is focused onto the specimen and excites the material. The probe beam is frequency-quadrupled to ultra-violet (257 nm) and then focused onto the filament of the TEM gun to generate electron pulses via photoemission. The delay between both pulses is controlled by tuning the path length of one laser beam with respect to the other, using a high-precision optical delay line. A new condenser lens (C_0 lens) was integrated for focusing the electron beam above the conventional condenser lens system, thus increasing the beam current.

In the present configuration (stroboscopic mode) the high repetition rate of the laser (1 kHz - 40 MHz) generates a continuous train of pulses that illuminates the TEM sample, allowing picosecond events to be investigated in imaging, diffraction and spectroscopy modes. The repetition rate of the laser is set such that the sample relaxes to its ground state

between the pulses. Images and spectra are generated through the integration of millions of pump-probe events.

The first results show comparable image resolution (0.23 nm) for thermionic and photoelectron modes, while the energy resolution for EELS (< 0.7 eV) is much improved in the photoelectron mode (Figs. 1a and b). Detailed characterization of the electron pulses (duration and intensity distribution) as a function of UTEM parameters is done through the Photon-Induced Near-Field Effect (PINEM). This effect is based on the photon-electron interaction in the near-field of nanoscale objects, i.e. electrons coinciding with the photon pulse at the specimen loose or gain entire quanta of photon energy which is visible in the electron energy-loss spectrum (Fig. 1c).

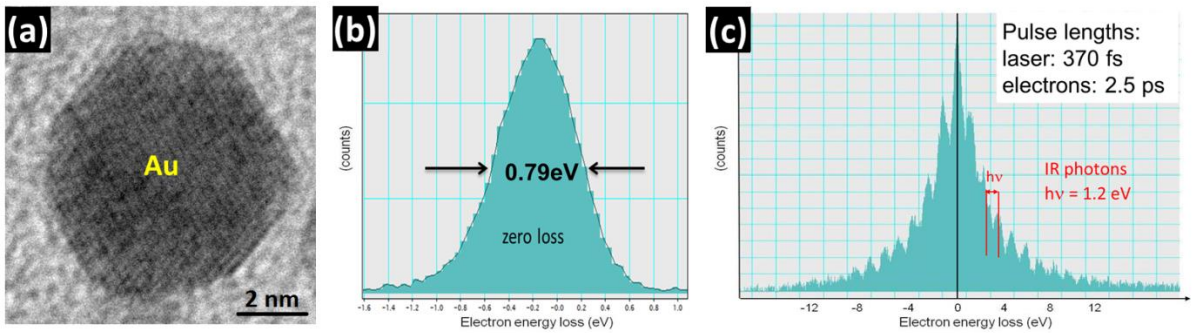

Figure 1: Spatial resolution in the photoelectron imaging mode, without laser on the specimen (a), zero-loss peak showing the EELS resolution in the photoelectron mode (b), broadening of the zero-loss peak in the PINEM spectrum (c).