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Use of a Direct Electron-Detection Camera in Ultrafast Electron Microscopy for Low Dose Rate Time-Resolved Imaging

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Ultrafast electron microscopy (UEM), using pulsed photoelectron packets for imaging, provides intuitive means of studying the fundamental processes accompanying structural dynamics [1]. In UEM, a femtosecond (fs)-pulsed laser is used to excite a specimen, the moment of which defines the time zero and to induce the photoelectric effect on a cathode for generating pulsed electron beam as a "pump-probe" scheme (Fig. 1) [2]. In this scheme the optical-pump electron-probe pulses are repetitively cycled for integration until building a time-resolved image, a diffraction pattern, or an electron-energy-loss spectrum at each time delay between the pump and probe.

Yet, the use of fs photoelectron packets suffers from a significant problem, Coulomb repulsion between charged particles confined in space and time. This space-charge effect limits the formation and propagation of fs-long electron pulses considerably because ultrashort electron pulses broaden in space and lengthen in time [3]. Typically, the spatial resolution of pulsed electron packets generated by optical pulses of a few hundreds of fs at lower repetition rates than MHz seems to reside around several nanometers at best [2]. To circumvent the limitation, the approach has been the optimization of a gun region [4-6]. In this space-charge quasi-free regime, the number of electrons in each fs pulse has to be in the order of thousands or less depending on their density. To build up an image of affordably good quality integrated in seconds, a stroboscopic measurement at the repetition rate of MHz is typical for detecting at least tens of electrons per pixel on a conventional charge-coupled device (CCD) camera. This high repetition, however, narrows the choice of a specimen because the specimen upon photoexcitation must fully relax to its original configuration in less than microsecond (μs), the case of which is practically rare for typical TEM specimens, *e.g.*, because of slower heat dissipation therein. To expand the scope of the stroboscopic imaging, a clocking period has to be set long enough to ensure the full relaxation of a specimen in each cycle of excitation. Moreover, the number of electrons per probe pulse is desired to reside in the space-charge free regime, and the pulse duration needs to be brief enough to take snapshots of structural changes.

Here, we push the limit of the spatial resolution of UEM by an-order-of-magnitude improvement with demonstrating the direct visualization of the sub-nm plasmonic acoustic vibration of a single gold nanorod with the period of 30 picoseconds (Fig. 2). This was made possible by uniquely integrating a direct electron-detection camera (Fig. 1), which poses much higher sensitivity to incident electrons, to our UEM [2]. This allows for a less number of electrons to record an image, which means the lower repetition rate of stroboscopic imaging and the minimal space-charge effect for a high spatial resolution are possible.

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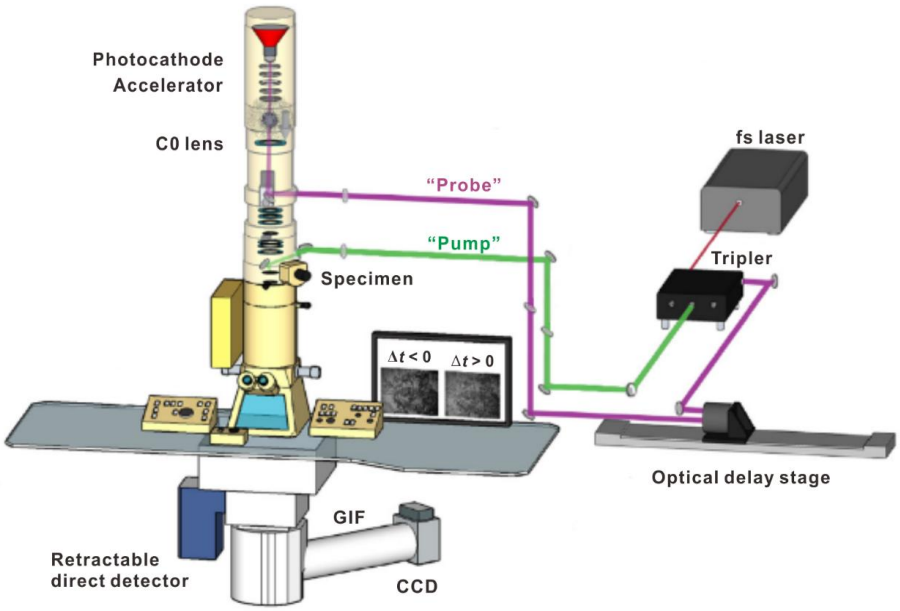

Figure 1. Schematic configuration of UEM at UNIST. A femtosecond laser system is interfaced to a modified TEM with two optical access ports to host laser pulses into a photocathode and a specimen in the TEM. An extra condenser lens and a direct electron-detection camera are integrated to the TEM.

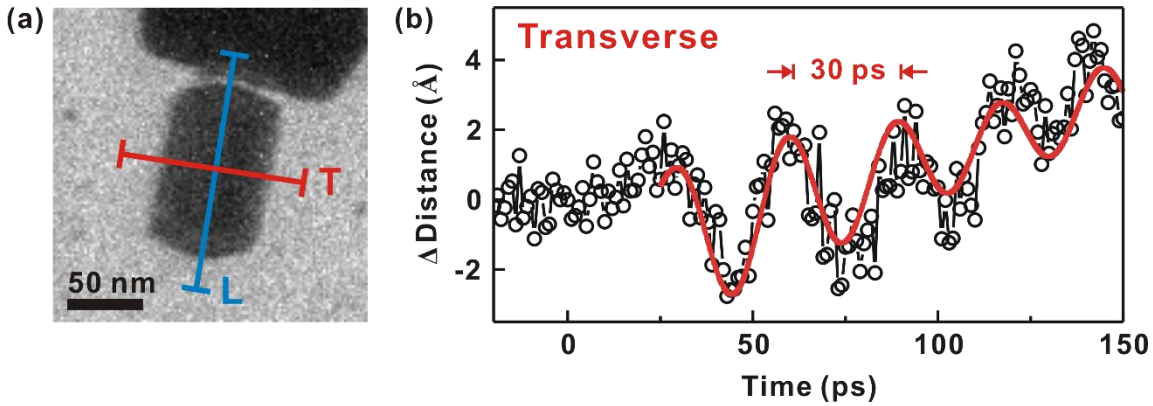

Figure 2. (a) The bright-field image of a vibrating gold nanorod. The quantification of the vibration was performed by extracting time-dependent change of the lengths along the longitudinal (L) and transverse (T) directions. (b) Acoustic vibration dynamics of the nanorod. The vibration dynamics of the nanorod was quantified in the transverse direction. All measured distances are relative to the at-rest position averaged before the time zero ($\Delta t < 0$). A Fit to a sum of damped sinusoidal functions is also depicted.

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