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Catching them in Action: Ultrafast Transmission Electron Microscopy

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Recent developments in instrumentation have made it a very exciting time to perform both fundamental and applied research in the electron microscope. *In situ* microscopy is moving forward at a rapid pace with the development of gas/liquid stages that permit reaction processes to be imaged and analyzed at atomic resolution. Moreover, the development of nanosecond and faster photoemission electron sources offers the chance to move the high spatial resolution world of electron microscopy into the ultrafast world of materials dynamics. Conventional *in situ* TEM coupled with ultrafast TEM can be utilized to gain a fundamental understanding of dynamic processes occurring in materials and biological structures. The combination of these capabilities allow for vast improvements of *in situ* TEM studies limited by video rate in that many processes span multiple time and length scales. Ultrafast dynamic *in situ* electron microscopy promises to answer challenging questions in the fields ranging from materials science and chemistry to nanoscience and biology.

One of the long-standing challenges in developing fundamental understanding in synthesis and processing of materials is observing the evolution of the structures, *in situ*. One such example is the synthesis/processing of materials using lasers, which has emerged as a technique that can be used to fabricate complex structures with a high degree of control.[1] The interaction of the materials subjected to either a pulsed or a continuous laser beam generates extremes of temperature and pressure in the material.[2] Because of this reason, incorporation of high temporal probing capabilities and lasers to the TEMs has been a critical development required to discover new knowledge about the dynamic evolution of nanostructures under processing conditions on varying time and length scales.

Figure 1 illustrates a schematic of the UTEM in our laboratory at the University of Connecticut with multi-mode operation (stroboscopic and single-shot) capabilities. High time resolution of the UTEM is achieved by creating short electron pulses that are used to illuminate the specimen. To initiate the laser induced structural and chemical evolution in nanostructures, a sample excitation laser pulse is used. This method is called "pump-probe scheme" and the delay between the two laser pulses, sets the timing of the observation. From the two approaches that enable a time resolution in the range of nanoseconds to femtoseconds, namely stroboscopic[3-5] and single-shot,[6,7] the laser-matter interactions in the field of processing of materials requires single-shot technique due to the irreversible nature of the investigated dynamics. Figure 2 represents a set of single-shot experiments performed on Ag nanoparticle (200 nm) system at two different time delays, namely, 10 ns and 1400 ns. As can be seen in Fig. 2a, very early dynamics of nanoparticle motions due to the sample excitation laser pulse is observed at 10 ns. At 1400ns, the initial stages of nanoparticle sintering and melting is captured in Fig. 2b.

The incorporation of accurate physics for the mechanisms during laser-matter interaction also requires a computational approach that incorporates the atomic scale models to include laser energy absorption, electron-phonon coupling, heat generation/transfer, and structural dynamics. Such links can be investigated using a coupled atomistic-continuum approach by combining classical molecular dynamics simulations with a continuum two-temperature model to model laser energy deposition, heat conduction and phase transformation at the atomic scales.[8,9] Additionally, multiphysics simulations within the finite element framework to determine the laser-matter interactions (electromagnetics simulations), heat transfer processes, fluid flow and various hydrodynamic effects,[10] are often critical to obtain a fundamental understanding into the mechanisms associated with the dynamic processes involved during these very complex processes. In this presentation, examples of single-shot electron microscopy studies and relevant computational approaches will be presented for selected systems. Additionally, the potential of novel *in situ* stages for various data acquisition scheme to push the envelope of ultrafast electron microscopy for the investigation of materials will be discussed.

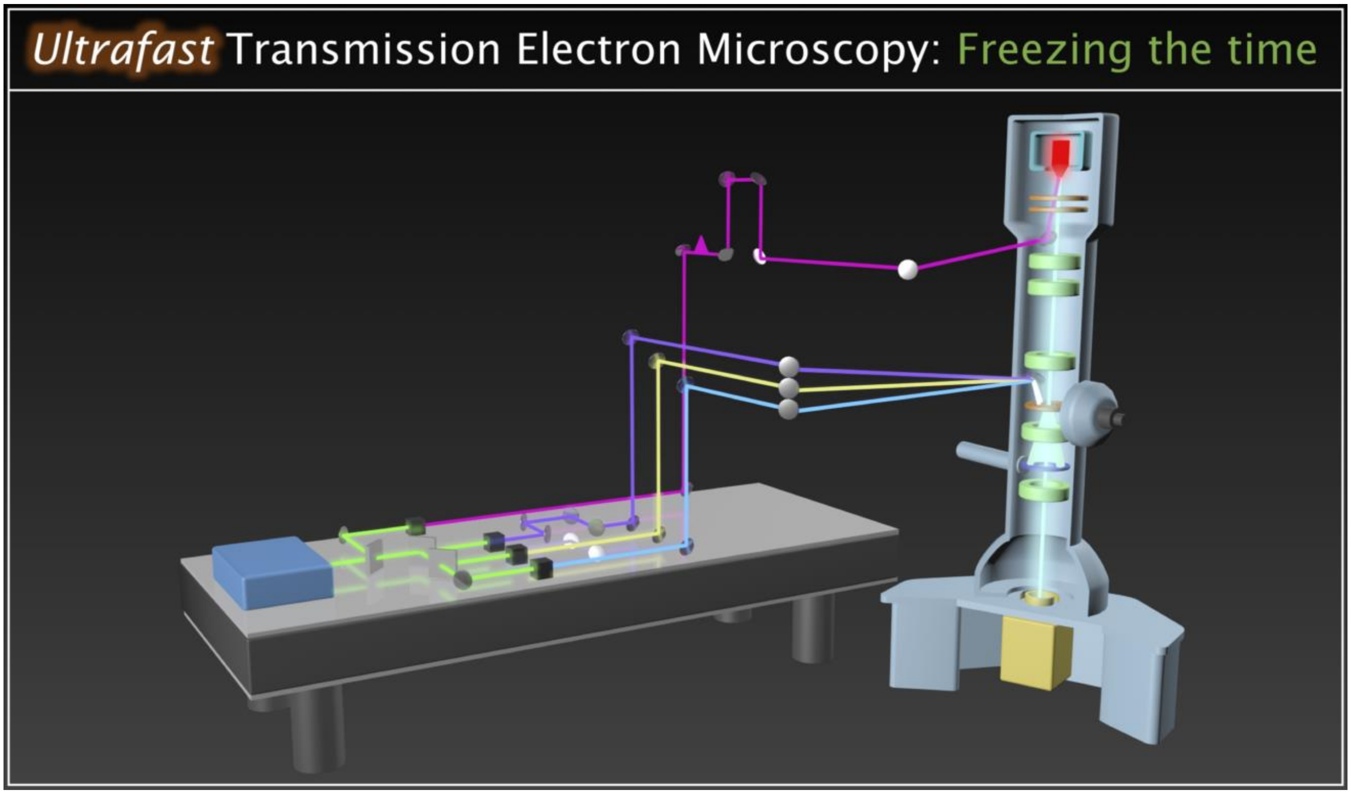

Figure 1. Figure1: Schematic illustration of the Ultrafast TEM in our laboratory at UConn.

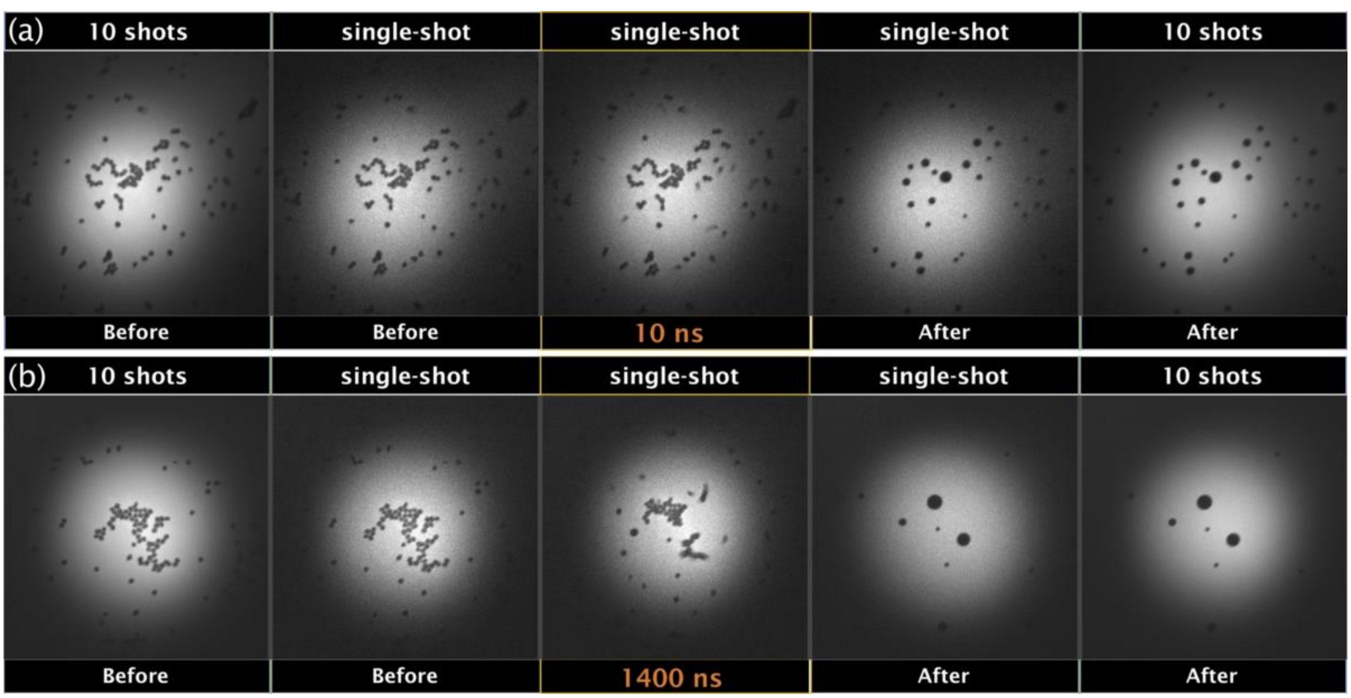

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Figure 2. Figure2: Laser induced dynamic evolution of 200 nm Ag nanoparticle system upon sample excitation laser. (a) 10 ns time delay and (b) 1400 ns time delay. Third images in each row are the single shot images showing the transient state of the dynamic process.

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