doi:10.1017/S1431927616004384
Microsc. Microanal. 22 (Suppl 3), 2016

Beyond Movie Mode: Bridging the Gap of Time Resolution

Bryan W. Reed[1], Sang Tae Park[1], and Daniel J. Masiel[1]

[1.] Integrated Dynamic Electron Solutions, Inc., Pleasanton, CA, USA

Movie Mode Dynamic Transmission Electron Microscopy (MM-DTEM) is a method of achieving nanosecond- to microsecond-scale multi-frame imaging of irreversible processes in an *in situ* TEM. MM-DTEM has revealed otherwise invisible details of fast, nonequilibrium chemical reactions, phase transformations, pulsed-laser metal nanoparticle sintering, and other processes of great fundamental and practical interest [1]. It works by initiating the process of interest with a nanosecond laser pulse directed at the sample and then repeatedly probing this process using a series of electron pulses. These pulses are brief (typically 10-50 ns) and intense (very often with 10^9 or more electrons each, enough to capture a high-quality real-space image in a single pulse). To achieve this, the thermionic cathode is replaced with a photocathode driven by an arbitrary-waveform pulsed ultraviolet laser, and the TEM's condenser lens system is modified (with the addition of a "C0" lens [2]) to accommodate the multi-milliampere peak current.

Between pulses, a fast two-dimensional electrostatic deflector switches states so that the next pulse will be directed to an as-yet-unexposed subregion on a large camera. This can be done in a 3x3 or 4x4 array, yielding 9 or 16 images in a span as short as one microsecond. The camera is read out at the end of the experiment, and the image is digitally segmented into 9 or 16 frames, producing a movie of the sequence of events in the sample. Each frame of the movie is precisely time-stamped relative to the time of arrival of the laser pulse that initiated the process, and the control system is able to allocate the 9 or 16 frames almost arbitrarily in time, extending experiments as far as 100 μs (by which time even the slowest-cooling sample will have essentially reverted to its initial temperature).

While MM-DTEM performs well at the tasks for which it was intended, no single tool is right for all experiments. Not all processes can be triggered to proceed on command with nanosecond precision. Some processes are too complex to have their essential nature revealed in only 9 or 16 frames. And, for many applications, nanosecond resolution is overkill; for many *in situ* and analytical applications (including STEM-EELS and STEM-diffraction), just being able to acquire continuously at multi-kHz frame rates would be more than enough. Such a solution would represent an as-yet-unattained middle ground, bridging the gap between nanosecond-scale burst-mode MM-DTEM and millisecond-scale continuous acquisition using modern TEM cameras.

The combination of the MM-DTEM deflector and (optionally) C0 systems enables this capability, allowing multiple frames to be acquired per captured frame without extensive modification of the TEM and without the need to add a powerful arbitrary-waveform laser to the system. We will discuss this approach, showing how the new operating modes enabled by this method can greatly enhance the data throughput of *in situ* and analytical (S)TEM. Ultimately, the combination of this kind of deflector-based system with one of the new generation of high-frame-rate cameras should enable true continuous-acquisition *in situ* experimentation on the microsecond scale. The main limitation becomes the beam current and not the acquisition system, and we will discuss approaches for expanding this bottleneck.

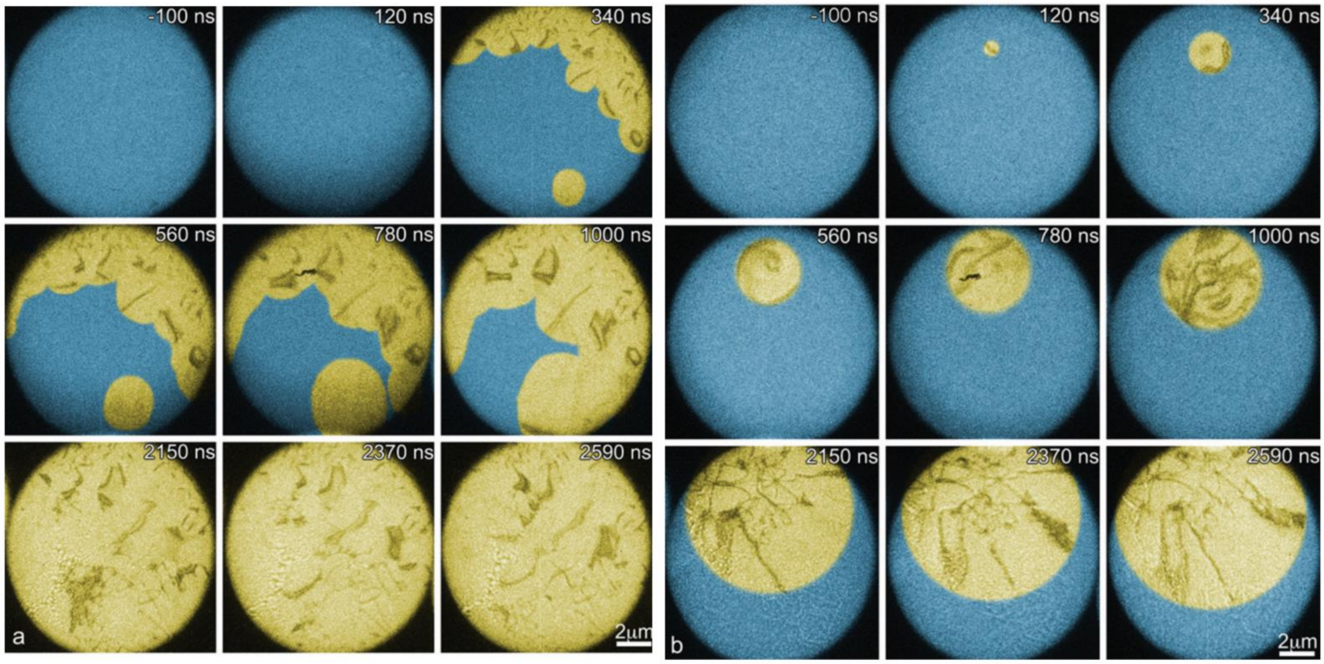

Figure 1. Two examples of nine-frame MM-DTEM sequences (with false color for clarity), showing nucleation and growth of GeTe crystals in an initially amorphous film [3].

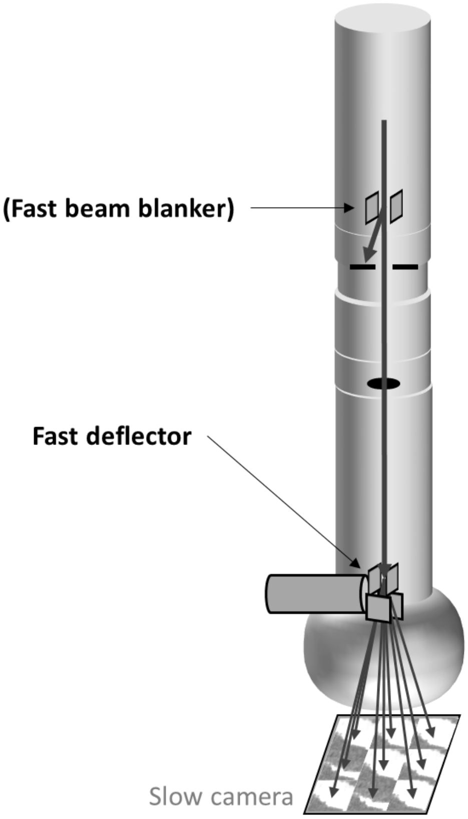

Figure 2. An electrostatic deflector system can be inserted into a standard 35 mm camera port. The system is capable of switching among up to 256 discrete deflection states in an arbitrary temporal pattern, with only 20 ns per switching operation, allowing a camera to act as an array of much faster cameras. An optional beam blanker eliminates the residual blur for extremely high-frequency operation.

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[4] This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences Small Business Innovation Research program under Contract DE-SC0013104.