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Imaging Coherent Structural Dynamics with Ultrafast Electron Microscopy

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Development of ultrafast electron and X-ray scattering methods has enabled direct routes to probing atomic-scale structural dynamics [1,2]. This in turn has led to new insight into lattice responses to a variety of dynamic processes associated with phase transformations, electron-lattice correlations, and nanoscale mechanical motion, to name a few. Currently, the most wide-spread approaches involve using table-top, laser-driven electron-scattering chambers or femtosecond (fs) X-ray beamlines to probe dynamics in reciprocal space and, in many instances, over specimen regions that are large relative to unit-cell dimensions and discrete (atomic and nanoscale) structural and morphological defects (*e.g.*, vacancies, line and screw dislocations, grain boundaries, interfaces, *etc*.). As a result, the transients monitored for relatively large-spot-size reciprocal-space measurements – typically some aspect of a Bragg reflection (intensity, position, width, shape) – may be comprised of a range of responses occurring within the probed region. To directly resolve the role of distinct morphologies and structural features on dynamics associated with the nucleation, emergence, evolution, and decay of coherent phenomena, one would ideally directly image the local, nanoscale behaviors and precisely correlate responses to the nature of the feature [3].

Here, I will show how such effects can be directly probed with ultrafast electron microscopy (UEM) [4]. After a brief overview of the operating principles of UEM, I will discuss how we are using real-space imaging to directly visualize coherent structural dynamics in a variety of nanostructured and nanoscale materials. Specifically, I will provide an overview of the results of our studies of photoinduced dynamics in: (i) transition metal dichalcogenides (TMDCs; MoS_2 and WSe_2) and (ii) thin Ge crystals [5-8]. In TMDCs, we have found that *in situ* fs photoexcitation leads to the generation of coherent phonon wavetrains at vacuum-crystal interfaces and extended crystal terraces. Correlative ultrafast parallel-beam diffraction studies, supplemented with finite-element modeling, indicate this occurs via an initial impulsive expansion along the *c*-axis stacking direction (*i.e.*, via an interlayer expansion) followed by launch of in-plane phonon wavefronts propagating at the speed of sound (*e.g.*, 5 nm/ps) and away from the specific interface. This indicates that such features serve as nucleation sites for the coherent, in-plane phonon wavetrains. In thin Ge crystals, we observe a variety of spatially-resolved effects; including direct imaging of coherent, hypersonic (*e.g.*, up to 35 nm/ps) acoustic-phonon wavefronts, delayed generation of the wavetrains (100 ps following photoexcitation) [9], and a time-varying phase-velocity relaxation to the bulk speed of sound occurring over one nanosecond [10].

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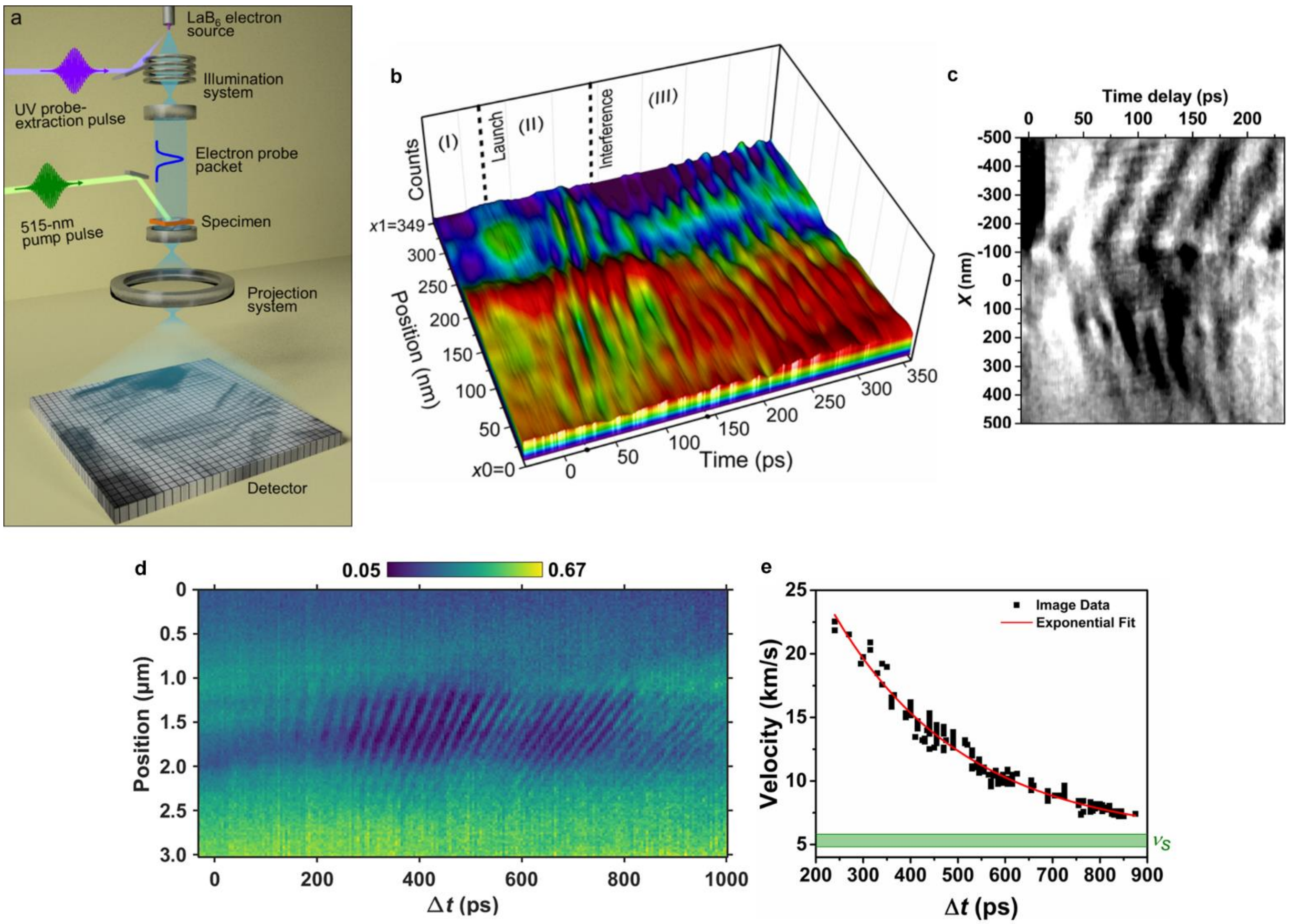

Figure 1. (a) Schematic of a typical UEM imaging experiment with all pertinent components labeled. Here, a 515-nm pump pulse is shown for illustrative purposes only (other wavelengths can be used). (b) Space-time surface plot showing coherent wavetrain propagation in an MoS_2 flake from approximately 0 to 140 ps, followed by interference of a counter-propagating wavetrain beyond 140 ps. (c) Space-time contour plot of two wavetrains consisting of individual acoustic-phonon wavefronts (dark, linear features) emanating from a WSe_2 crystal terrace (located at approximately X = 0 nm) and counter-propagating away from one another at the speed of sound (5 nm/ps). (d) Space-time contour plot of individual phonon wavefronts (dark, linear features) propagating through a Ge crystal. (e) Time-varying phase-velocity behavior of the phonon wavefronts propagating through a Ge crystal. The velocities decay from being hypersonic to approaching the bulk speed of sound (v_s).

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