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Morphological Modulation of Acoustic Phonons Imaged with Ultrafast Electron Microscopy

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Atomic-scale manipulation and control of phonon modes has been proposed and vigorously pursued for enabling and enhancing myriad technological developments [1]. Indeed, the formulation of a comprehensive microscopic description of the real-time interaction of propagating modes with individual lattice discontinuities having primary features best visualized at the atomic level (*e.g.*, grain boundaries, step-edges, strain fields, *etc.*) would constitute a significant advance toward ultraprecise coherent energy manipulation and control. Fundamentally, structural dynamics of this nature are amenable to study with ultrafast methods that make use of the dependence of scattering wavevectors on lattice orientation and symmetry [2]; movement or spacing/symmetry changes of the reciprocal lattice on a fixed Ewald sphere produces a commensurate modulation or re-configuration of the resulting coherent-scattering pattern. Importantly, phase information is retained when probing dynamics in real space, thus enabling spatiotemporal localization of discrete phonon-nucleation events and resolution of propagation dynamics and frequency dispersion at morphological discontinuities.

Here we report direct, real-space imaging of the emergence and evolution of single-phonon wavefronts at individual atomic-scale defects in layered WSe_2 and crystalline Ge (Fig. 1). Via stroboscopic, bright-field imaging with an ultrafast electron microscope [3], transient elastic deformations induced by propagating lattice waves manifest as spatially-localized contrast modulations due to femtosecond perturbation of the Bragg condition. By isolating and quantifying the transient contrast modulation, we extract the spatially-dependent vibrational-mode shapes and properties – namely, symmetries, frequencies (GHz), phase velocities (*e.g.*, 6 nm/ps), and decay times (hundreds of picoseconds) – over fields-of-view on the order of the phonon wavelengths. In addition, we are able to track the velocity and frequency dispersion of speed-of-sound acoustic waves as they traverse morphological features endemic on the nanoscale. In this way, the roles played by individual atomic-scale defects, spatially-varying lattice orientations, and associated nanoscale strain fields on phonon nucleation and dynamics can be determined, thus providing insight for ultraprecise generation and control of coherent energy propagation in real materials [4].

References:

[1] Y. J. Hu *et al*, Nat. Nanotechnol. **10** (2015), p. 701.
[2] R. J. D. Miller, Science **343** (2014), p. 1108.
[3] D. A. Plemmons *et al*, Chem. Mater. **27** (2015), p. 3178.
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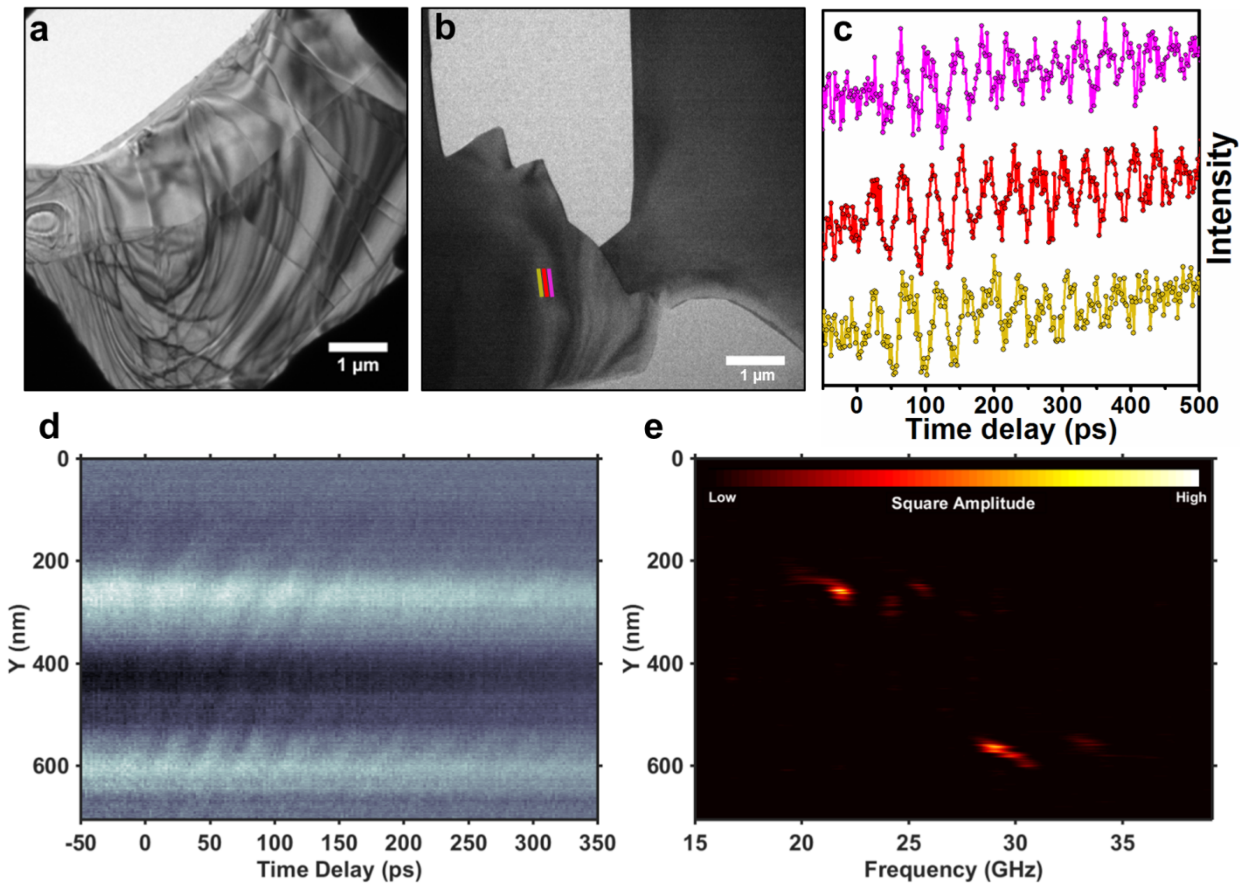

Figure 1. Imaging and quantification of acoustic-phonon dynamics in WSe_2 and Ge. (a) Thermionic bright-field image of a WSe_2 flake. (b) Photoelectron bright-field image of crystalline Ge. (c) Time-intensity traces generated from adjacent regions marked by the colored lines in (b). (d) Space-time plot taken along the direction of contrast-wave propagation showing dispersion as well as change in wave velocity with time. (e) Space-frequency plot corresponding to (d), wherein dispersion of traveling contrast waves is observed.

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