Spatially-Resolved Strain-Wave Dynamics in MoS\textsubscript{2}

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Band structures and, therefore, electronic and optical properties of transition metal dichalcogenides are sensitively dependent on crystal thickness (\textit{i.e.}, number of layers) and applied elastic strain in the few to single-layer limit \cite{mak2010,conley2013}. The most widely studied example is MoS\textsubscript{2}, in which the band gap was discovered to increase from 1.3 eV in the bulk to 1.9 eV for a single layer, with a commensurate transition from an indirect to direct gap. Consequently, MoS\textsubscript{2} and other TMDs and heterostructures are being explored as active elements in thin, flexible electronic and photonic devices \cite{jariwala2014}. The interesting structure-function properties of these materials has spurred interest in structural and mechanical effects on charge-carrier and transport dynamics, as well as the possibility of strain-engineering phase transformations \cite{fujihara2018}. In addition, the importance of grain boundaries, vacancies, terraces, and interfaces with respect to electronic, optical, and mechanical properties has been demonstrated \cite{fujihara2018}. While the transient response of charge-carrier dynamics has been extensively studied with ultrafast spectroscopic approaches \cite{miller2014}, the structural response to coherent (rapid) perturbation (via photoexcitation, for example) is not well understood, especially on the scale of individual crystal defects.

Here, we describe our observations of photoinduced, nanoscale coherent elastic strain-wave dynamics in individual freestanding flakes of MoS\textsubscript{2} \cite{zhang2018}. Via femtosecond bright-field imaging with an ultrafast electron microscope \cite{miller2014}, we are able to observe and quantify the evolution of acoustic-phonon wavetrains – from spatially-resolved nucleation and subsequent propagation to initial interference and scattering of individual wavefronts (Figure 1). Dynamics are triggered by \textit{in situ} photoexcitation of the specimen with a femtosecond laser pulse, and the structural response is quantified by analyzing the motion of bend contours spanning picoseconds to microseconds (Figure 1 illustrates dynamics spanning approximately the first 2 ns following photoexcitation). In this way, both the frequencies (tens of GHz) and propagation directions and velocities (speed of sound) can be determined. Because dynamics are studied with real-space imaging, additional information pertaining to nucleation points/regions and local interference and scattering processes can be determined \cite{zhang2018}. Importantly, we find these discrete behaviors in the overall dynamics to be highly specimen dependent; flake-specific morphologies and boundary conditions significantly impact the precise dynamics observed. For example, terraces formed by oriented few-layer sections having smaller lateral dimensions than the entire flake serve as specific nucleation regions for phonon wavetrains. Further, the orientation of the terraces dictates the wavetrain propagation direction (normal to the terrace). This subsequently dictates the precise nature of scattering and interference effects. It remains to be seen if such specimen-to-specimen variation also affects timescales for coupling mechanisms or phase-transformation nucleation and growth kinetics, for example \cite{miller2018}.

References:

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Figure 1. Elastic strain-wave dynamics in MoS$_2$. (a) Photoelectron bright-field image of a freestanding MoS$_2$ flake prior to in situ femtosecond photoexcitation. The yellow and red rectangles ($x_1$ and $x_2$, respectively) denote regions of interest (ROIs) from which dynamics were quantified. The dark feature in the lower-left corner of the image is the Cu grid substrate. Scale bar = 1 µm. (b) and (c) Space-time plots of the ROIs in (a). Each time point in panels (b) and (c) is comprised of image counts averaged along the width of the ROIs, thus producing a snapshot in time of the one-dimensional contrast profile. Strain waves passing through these ROIs cause time-dependent contrast variations, from which the spatially-resolved oscillation frequencies (b) and directionality (c) can be determined. In (c), the dashed green and orange lines are meant to guide the eye to specific linear contrast features that arise from individual wavefronts propagating through the ROI. (d) through (f) Space-frequency plots of the region highlighted in the yellow ROI for three temporal ranges: 0 to 0.49 ns (d), 0.5 to 0.99 ns (e), and 1 to 1.49 ns (f). The warmer colors represent larger amplitudes.