doi:10.1017/S1431927616009156
Microsc. Microanal. 22 (Suppl 3), 2016

Imaging Photoinduced Structural and Morphological Dynamics of a Single MoS_2 Flake With Ultrafast Electron Microscopy

Alyssa J. McKenna[1], Jeffrey K. Eliason[1], and David J. Flannigan[1]

[1.] Department of Chemical Engineering and Material Science, University of Minnesota, 421 Washington Ave SE, Minneapolis, Minnesota, United States of America.

Real-space imaging of ultrafast photoinduced dynamics in molybdenum disulfide (MoS_2) offers a comprehensive understanding of the material's optical response, the transition from charge-carrier excitation to mechanical motion, and the decay of that motion. In particular, this knowledge will inform and improve the design and performance of photodectors and flexible electronics, of which MoS_2 is a material of interest [1,2]. Recent work on MoS_2 photoinduced electronic and structural dynamics focused mainly on the initial moments following excitation and used reciprocal-space studies to directly probe the lattice response [3,4]. Here, we directly image the photoinduced structural dynamics of an individual MoS_2 flake using ultrafast electron microscopy (UEM) [5]. The individual flake (Fig. 1 and 2) was excited at 515 nm with a 270-fs pulse, and the subsequent structural response was followed via real-space bright-field UEM imaging at time steps of 1 picosecond (ps), 10 ps, and 10 ns. The dynamic response was determined from the real-space images by mapping the trajectories of low-index diffraction-contrast features (*e.g.*, bend contours and moiré fringes).

From the UEM images series, we are able to directly follow the structural response of the individual MoS_2 flake to photoexcitation, and we find that initial high-frequency modes (~50 GHz) attributable to acoustic-phonon modes quickly scatter and interfere with one another during the first few hundred ps. These high-frequency modes soon give way to lower frequency mechanical oscillations comprised of multiple modes that subsequently relax into a single mode. The initial fast oscillations are summarized in Figure 1. The displacement of a single diffraction contrast feature is followed as a function of time, the temporal response of which is plotted in Figure 1(b). The FFT of this displacement [Figure 1(b) inset] reveals a single oscillation frequency of 50 GHz, in accord with the excitation of an acoustic-phonon mode. This high-frequency oscillation quickly damps over the first 200 ps and relaxes into lower frequency (~1 GHz) oscillations, as illustrated by the FFT spatial map in Figure 1(c). Soon after, these oscillations are converted into still lower-frequency mechanical oscillations, as quantified from the UEM nanosecond image series (Figure 2). Three distinct modes are observed in this nanosecond image series, all being a few to several MHz and with (1/*e*) lifetimes of tens of microseconds [Figure 2(c) and inset]. From the timescales and associated temporally-dependent behavior, we find that initial photoexcitation of charge carriers quickly couples to acoustic-phonon excitation within the first few ps. These acoustic-phonon modes quickly couple to lower frequency modes, as evident in the 10-ps image series, the precise timescales of which appear to strongly depend upon the geometrical boundary conditions of the flake. Within one nanosecond, these single-GHz modes couple to mechanical-oscillatory modes, which then dissipate the remaining energy via ring down over tens of microseconds. This work provides insight into the photoinduced structural response of few-layer MoS_2 with implications for future applications [6].

References:

[1] Q. H. Wang, *et al.*, *Nature Nanotechnol.* **7** (2012), pp. 699-712.

[2] O. Lopez-Sanchez, *et al.*, *Nature Nanotechnol.* **8** (2013), pp. 497-501.
[3] E. M. Mannebach, *et al.*, *ACS Nano* **8** (2014), pp. 10734-10742.
[4] E. M. Mannebach, *et al.*, *Nano Lett.* **15** (2015), pp. 6889-6895.
[5] D. A. Plemmons, *et al.*, *Chem. Mater.* **27** (2015), pp. 3178-3192.
[6] We acknowledge support by 3M in the form of a Nontenured Faculty Award under Award Number 13673369, by the Donors of the American Chemical Society Petroleum Research Fund under Award Number 53116-DNI7, and by the Arnold and Mabel Beckman Foundation in the form of a Young Investigator Award. This material is based upon work supported by the National Science Foundation Graduate Research Fellowship Program under Grant No. DGE-1348264.

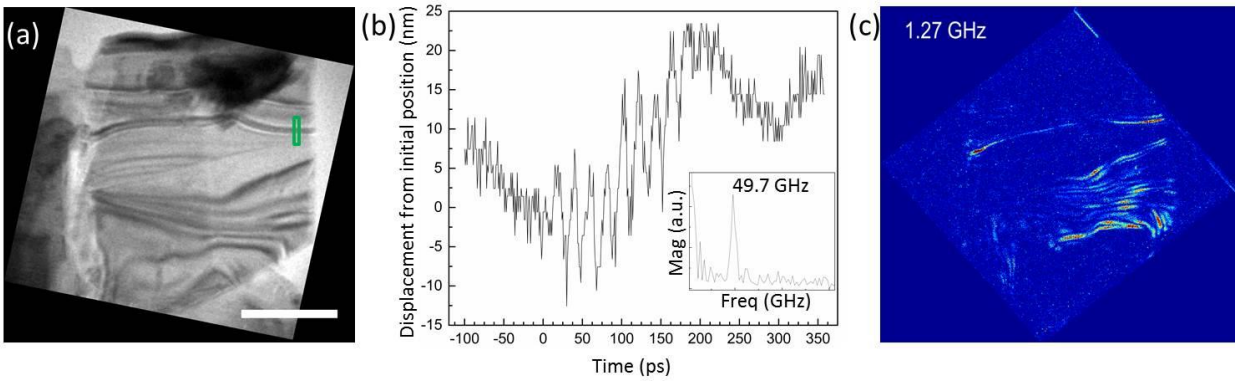

Figure 1. (a) Femtosecond UEM image of a single MoS_2 flake obtained at time zero. Scale bar = 1 μm. (b) Displacement from the initial position of the region of the bend contour in (a) contained within the green rectangle. The inset shows the FFT of the time trace, with a prominent peak at 49.7 GHz. (c) Magnitude of the FFT at each pixel at 1.27 GHz, where red corresponds to the highest magnitude value. Dominant frequencies are on the order of 1 GHz.

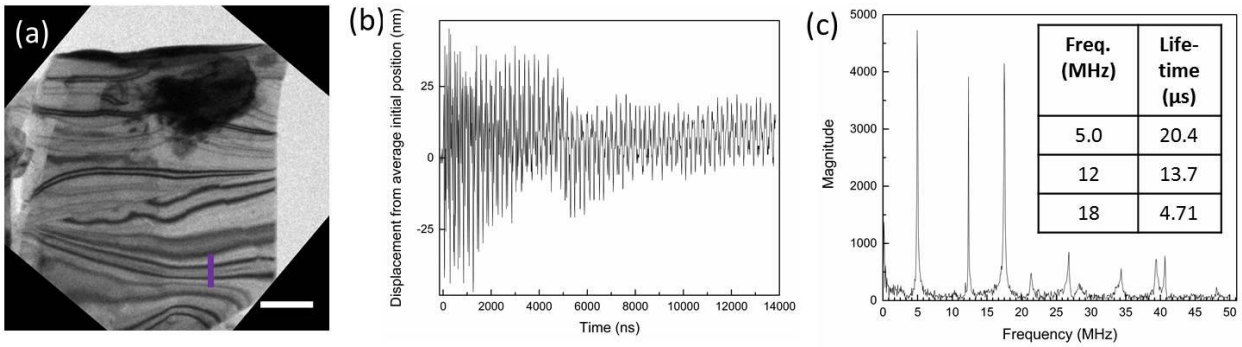

Figure 2. (a) Nanosecond UEM image of a single MoS_2 flake at negative time (*i.e.*, before photoexcitation). Scale bar = 500 nm. (b) Displacement from the initial position of the region of the bend contour in (a) contained within the purple rectangle. (c) FFT of the time trace in (b). The table inset lists the frequencies of three main peaks and the associated (1/*e*) lifetimes.

https://doi.org/10.1017/S1431927616009156 Published online by Cambridge University Press