Exploring Thermal Properties of MoS₂ Using In Situ Quantitative STEM

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We present an investigation on the thermal properties of MoS₂ using *in situ* quantitative STEM. Behind the thermal properties of condensed matter are phonons that can be manipulated by phonon engineering. Spatial confinement of phonons in nanostructures and thin films offer a new way of controlling thermal transport, which can lead to exciting advancements in the heat management of electronic, thermoelectric and optoelectronic devices (*e.g.* [1]). A major contribution to the thermal conductance of the system comes from defects, making the understanding of thermal motion at or near structural defects, such as dislocations or at epitaxial interfaces critical [2]. A study at these locations would require information about the atomic scale thermal motion. While Raman or EELS vibrational spectroscopy [*e.g.* 3] can reveal thermal motions, it is still difficult to resolve the thermal motions at the atomic scale because the techniques have limited spatial resolution.

In this work, we use quantitative STEM to determine thermal vibration information in MoS₂ at the atomic scale. Quantitative STEM measures the absolute scattering intensity at high angles that can be directly compared to simulated intensities [4]. Since the amplitude and width of the scattering intensity depend directly on thermal vibration [5], the Debye-Waller (DW) factors of each atomic column (or single atom) can be quantified from a single quantitative STEM image. This provides new possibility of characterizing atomic thermal motions at or near defects and interfaces, which can deliver new understanding in thermal properties of novel functional materials.

Prior to the characterization of the thermal motions near defects, the DW factors of "normal" MoS₂ structure must be known. The exact DW factors for the structure, however, remain elusive. Previous reports suggested that the DW factors of MoS₂ may be substantially different depending on the bonding type, substrate, or the number of layers in the sample [*e.g.* 6]. It is also possible that the measured DW factors can be affected by the dynamic structural response of the material upon illumination [7]. In the present work, we measure the DW factors of MoS₂ layers as a function of temperature and number of layers in an *in situ* quantitative STEM condition. Our data can serve as a reference for the characterization of potentially abnormal thermal motions near defects or interfaces, and also provide the information about the size of the electron source incoherence [4] that is difficult to estimate otherwise.

We use our layer-by-layer etching technique to precisely control the number of MoS₂ layers in the sample [8]. The sample will be directly transported to the MEMS chip of the FEI Nano-EX *in situ* holder (Fig. 1a and b) that ensures fast heating rate and excellent thermal stability with minimal sample drift [9]. Quantitative STEM images are taken from both plan-view and cross-sectional views (Fig. 1b and c). Figure 2a shows the HAADF STEM images of MoS₂ (2 layers) simulated using frozen phonon multislice algorithm at 0 K with estimated DW factors [6]. Same simulations as a function of temperature revealed that the amplitude of the Gaussian fit to the atomic column decreases as a function of temperature, while the width of the Gaussian function does not change significantly (Fig. 2b to d). Using a χ^2 fitting of the simulated Gaussian profiles to the experimental images, the DW factors for each

atomic species in MoS_2 can be measured as a function of the number of MoS_2 layers. Our *in situ* experiment also provides a more reliable way to estimate the incoherent source size [4], because the incoherent source function must be independent of the sample temperature. Effects of error sources, such as shot noise and possible scan noise, will be also discussed.

References

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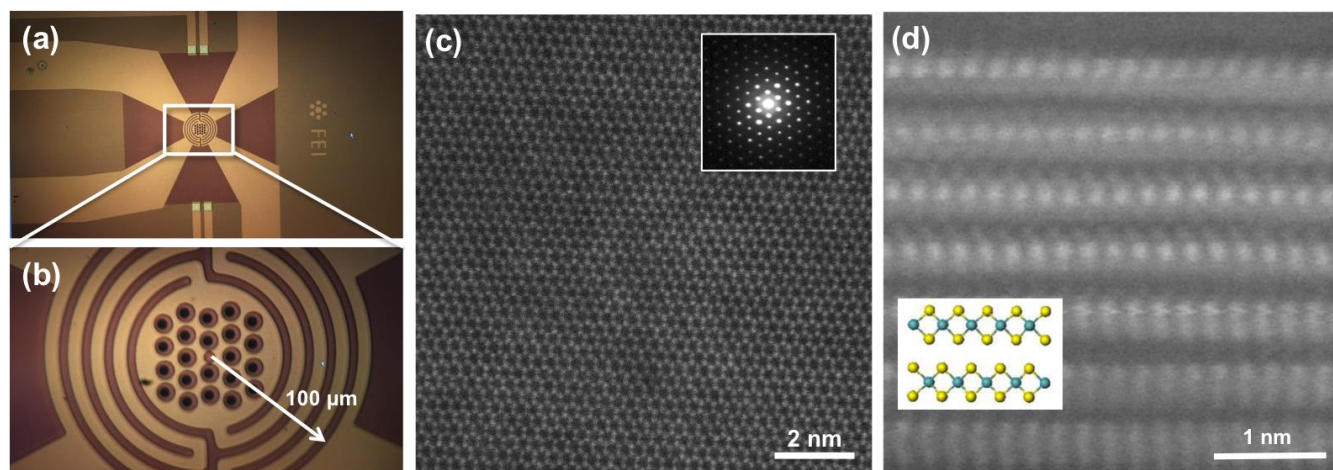
Figure 1. (a and b) The in situ heating element in FEI Nano-EX TEM holder [9]. Experimental STEM ADF image of (c) plan-view multilayered MoS_2. The diffraction pattern (inset) confirms that the sample is single crystalline. (d) Cross-sectional view of the multilayered MoS_2 sample [8].
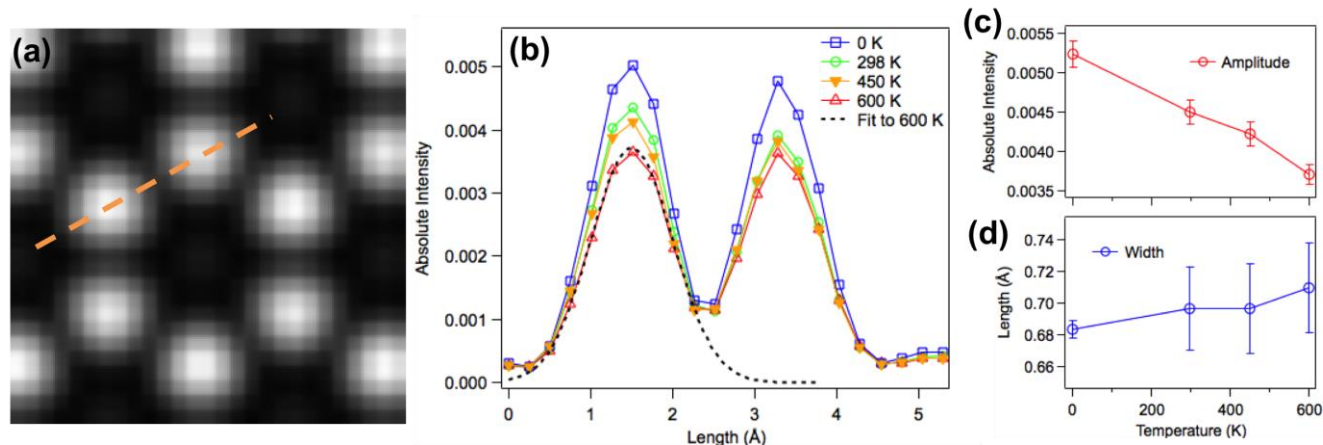
Figure 2. (a) Simulated HAADF image of 2 layers of MoS_2. (b) Line profiles of the dashed line shown in (a), and the (c) amplitude and (d) width of the Gaussian fit used in (b) as a function of temperature.