STEM Characterization of Epitaxial Monolayer MoS_2

Xiaoxu Zhao[1, 2], Deyi Fu[1], Zhongxin Chen[1, 2], Wu Zhou[3], Kian Ping Loh[1]

[1.] Department of Chemistry and Center for Advanced 2D Materials (CA2DM), National University of Singapore, 3 Science Drive 3, Singapore 117543, Singapore
[2.] NUS Graduate School for Integrative Sciences and Engineering, National University of Singapore, Centre for Life Sciences, #05-01, 28 Medical Drive, Singapore 117456, Singapore
[3.] Materials Science & Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, United States.

Growing single crystal monolayer MoS_2 at large scale is the basis for high performance electronics and optoelectronics. Currently, large-area MoS_2 films are almost inevitable polycrystalline, with deteriorated electrical properties ascribed to the existence of various grain boundaries. [1] Epitaxial growth is a promising approach for achieving single crystal film with solely limited mirror grain boundaries (m-GBs) at merging areas, which are even predicted beneficial for electrical properties. [2] Such merging areas are critical; however, it has rarely been studied and reported, especially at atomic scale.

Here we apply aberration corrected scanning transmission electron microscopy annular dark field (STEM-ADF) imaging to disclose the atomic structure of monolayer MoS_2 and utilize electron energy loss spectroscopy (EELS) to identify the elemental distribution. Monolayer MoS_2 film was grown by molecular beam epitaxy (MBE) on an exfoliated BN substrate, and was transferred onto TEM grids through a polymer free method. The STEM experiments were performed on an aberration-corrected Nion-UltraSTEM-100 at 60 kV.

Figure 1(a) shows a large scale STEM-ADF image containing dozens of triangular MoS_2 flakes on BN substrate. All triangles point either in the same direction, or position at a relative 60° rotation, which indicates an epitaxial growth of MoS_2 on BN. Figure 1(b) reveals a typical MoS_2 triangle flake without extended defects inside the crystal. From the fast Fourier transform (FFT) pattern, the same orientation of MoS_2 (red circle) and BN (yellow circle) further confirms an epitaxial growth nature. EELS mapping (Figure 1c) demonstrates a uniform elemental distribution of Mo and S in triangles, and a homogeneous distribution of B and N across the whole substrate.

It is known that MoS_2 has a 3-fold symmetry in (001) plane. Suppose all MoS_2 flakes share the same or a relative 60° rotation. When two random flakes merge, the merging area should have only two scenarios – perfect merging (no GBs) or consisting of merely m-GBs. These two cases are both frequently observed. A typical m-GB is shown in Figure 2(a), with a S-4|4E type (mirror grain boundary consists of strings of 4- fold rings with Sulphur edge sharing) structure (Figure 2c) [3]. We have analyzed dozens of grain merging cases, and surprisingly, only S-4|4P (composed of 4-fold rings with Sulphur point sharing) and S-44|E types were found, while Mo-oriented GBs were never observed. This is most likely due to the high formation energy of Mo-oriented m-GBs. [4] Figure 2(b) and 2(d) indicate a potential perfect grain boundary merging area with no misalignment. Herein, our STEM results demonstrate the possibility in fabrication of large scale single crystal MoS_2 via an epitaxial growth approach. Only m-GBs were observed and the material holds potential for high performance device applications. [5]

References:

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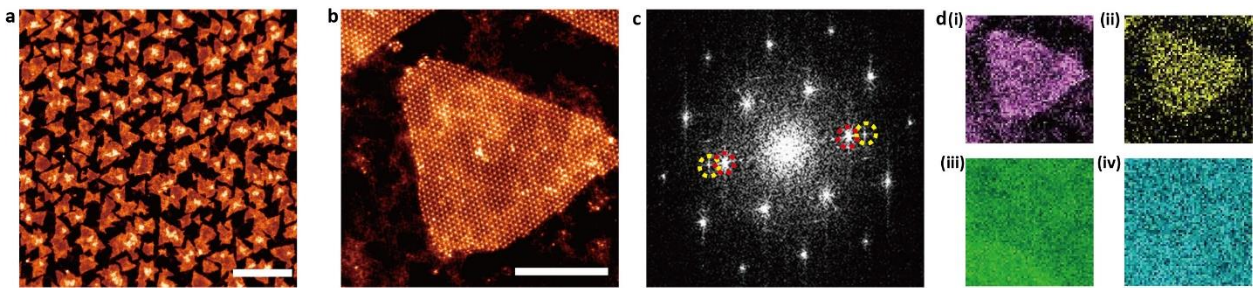

Figure 1. **(a)** STEM-ADF images of a monolayer MoS_2 film at a large field of view and **(b)** a typical triangular shape monolayer MoS_2 flakes and its corresponding **(c)** FFT pattern. **(d)** EELS elemental mapping of (**i**) Mo, (**ii**) S, (**iii**) B, and (**iv**) N in the same area of b. Scale bar: 100nm in (a), and 5nm in (b).

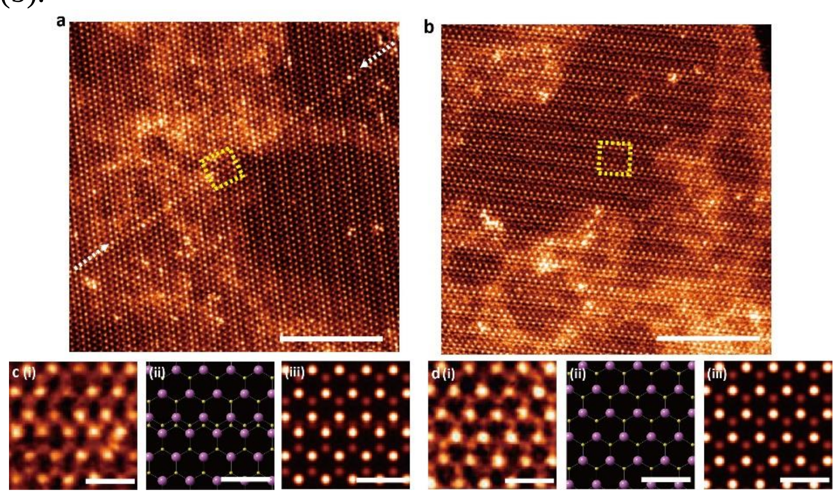

Figure 2. (a) STEM-ADF image of a monolayer MoS_2 film containing a m-GB at merging area indicated by the white arrows. (b) STEM-ADF image of a potential merging area in monolayer MoS_2 film with perfect grain merging. **(C) (i)** Zoom in image of the yellow box area in (a) showing the m-GB structure. **(ii)** Atomic structure of the m-GB and **(iii)** simulated ADF images based on the structure model in (ii). **(d) (i)** Zoom in image of the yellow box area in (b) showing perfect merging. **(ii)** Atomic structure of the merging area and **(iii)** simulated ADF images based on the structure model in (ii). Scale bar: 5nm in (a), and 0.5 nm in (b) and (c).