Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Layer-by-Layer Thinning of Two-Dimensional MoS_2 Film by Focused Ion Beam

Ding Wang, Yuqiu Wang, Xiaodong Chen, Yuankun Zhu*, Ke Zhan, Junhe Yang, Xianying Wang*

College of Materials Science and Engineering, University of Shanghai for Science & Technology, Shanghai, 200093, People's Republic of China.

* Corresponding author: xianyingwang@usst.edu.cn (Prof. X. Y. Wang), zhuyuankun@usst.edu.cn (Y. K. Zhu)

The precise control of the thinning process of MoS_2 thin films will be influenced mainly by the following 3 factors (but not only on accelerating voltage):

(1) Ga^+ ionic energy, which is mainly determined by the accelerating voltage. The ionic energy of imposed Ga^+ is attributed to the transformation from the electric potential energy to the kinetic energy due to the applied electric field (acceleration voltage). From this perspective, acceleration voltage is of great importance to control the surface damage of MoS_2 thin films. For the precise controlling the surface status, it is obvious that high accelerating voltage will cause damage to the samples. For our experiments, 10 kV is the largest voltage we used. Following the reviewer's suggestion, we tried 15 kV, 20 kV, and 30 kV. As expected, no Raman signals can be detected under 20 kV and 30 kV (Fig. S4), which means the structure of MoS_2 has been destroyed already under such conditions.

(2) Ga^+ dose per frame. The imposed Ga^+ dose (D_{frame}) per frame in FIB thinning is:

$$D = \frac{I * t}{x_{\text{p}} y_{\text{p}}}$$

where I is beam current, t (dwell time) is the time that the beam spends on a single pixel per pass, x_{p} and y_{p} (dwell spacing) is the pitch between two spots which depends on the spot size and overlap rate. A higher dose of Ga ions will lead to less control over the etching depth. During our experiments, we fixed the x_{p}, y_{p} and dwelling time to make I as the only parameter to be adjusted. To minimize the possible damage, the beam current was set to be as low as possible. As illustrated in the laser thinning experiments of MoS_2 (Nano Letters, vol. 12(6), pp 3187-3192 (2012)), there should exist a critical point of the maximum dose above which MoS_2 film will be damaged.

(3) The total amount of etched materials. If A is total thinning area, the total amount of etched material can be expressed as:

$$V = I * V_d * t_m$$

where V is the total etching volume, V_d is the sputtering rate describing the amount of material volume removed per charge. For a given material, V_d is a default value. V equals to $A*d$, where d is the tentative etching depths. During the FIB operating process, I and d can be set and t_m will be calculated by the system accordingly. Therefore, I and d are two key factors here.

Considering there are too many factors which will influence the FIB thinning process, according to the above analysis, we take the accelerating voltage V, ion beam current I and etching depth t as three main parameters to study the FIB thinning of few layer MoS_2 films, and the thinning results were characterized by Raman spectroscopy and optical microscope. Raman is a powerful method which can determine whether the structure of MoS_2 film is well-maintained and it is also thickness sensitive. We investigated many groups of etching parameters, and Fig. S5 is one of the representive results. Very low beam currents and small accelerating voltages are necessary for the precise thickness control.

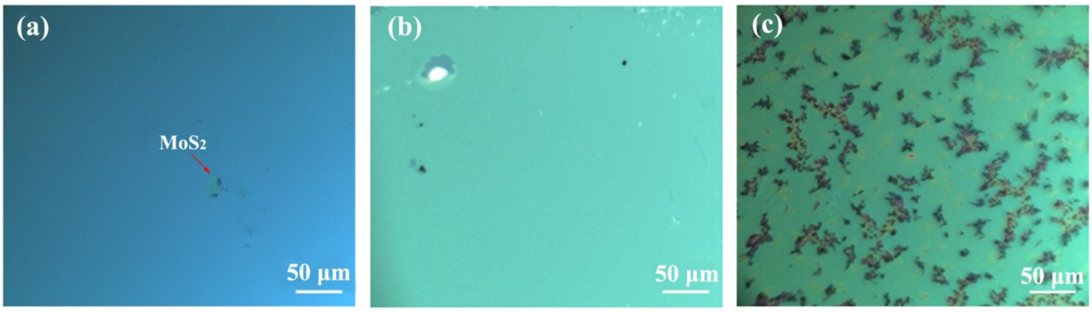

Fig. S1 OM images of MoS_2 samples prepared under different reaction temperatures of (a) 750 °C, (b) 800 °C and (c) 850 °C.

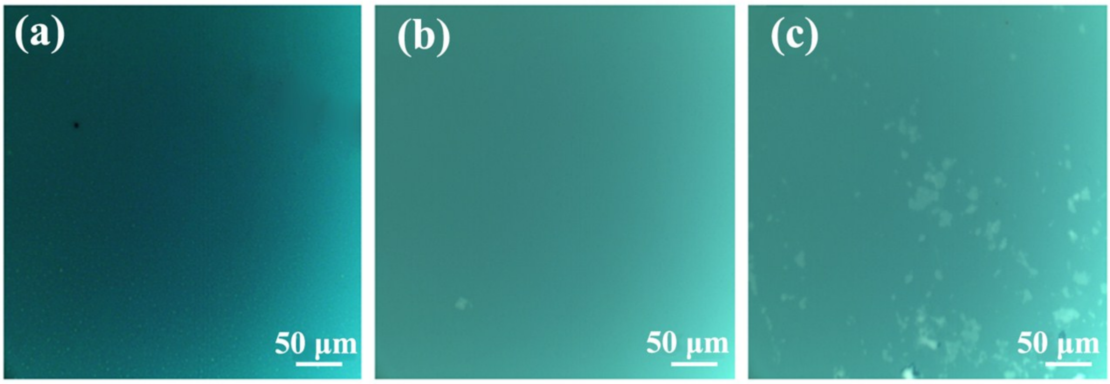

Fig. S2 OM images of MoS_2 samples prepared under different preservation times of (a) 5 minutes, (b) 10 minutes and (c) 15 minutes.

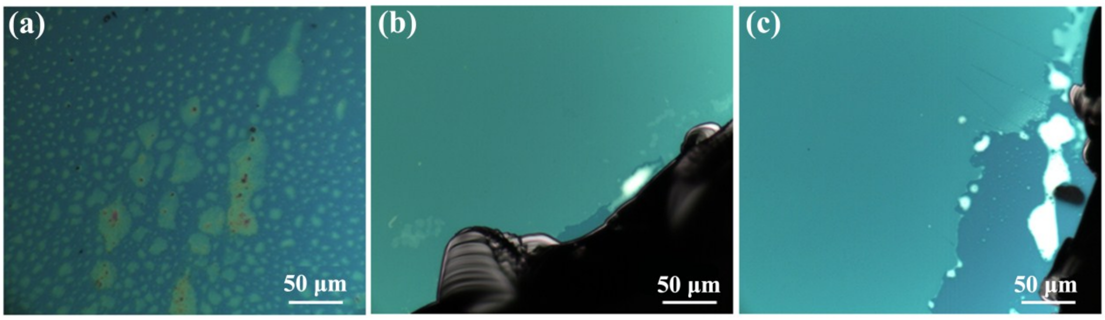

Fig. S3 OM images taken at the edge area of MoS_2 samples prepared under different thicknesses of Mo layer of (a) 0.5 nm, (b) 1 nm and (c) 1.5 nm.

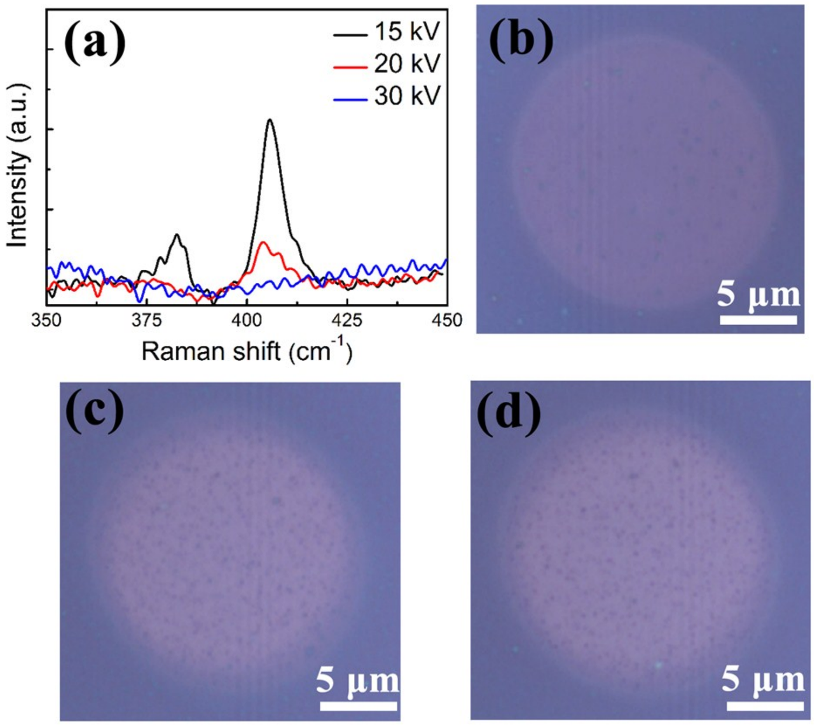

Fig. S4 (a) Raman spectra of MoS_2 films thinned under different accelerating voltages. (b)-(d) OM images of MoS_2 films thinned under 15 kV, 20 kV, and 30 kV, respectively.

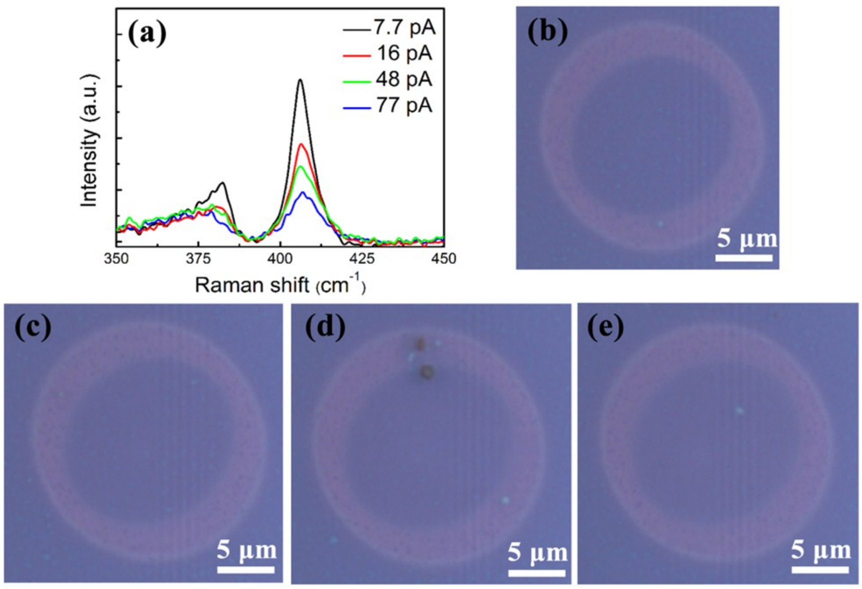

Fig. S5 (a) Raman spectra of MoS_2 films thinned under different beam currents. (b)-(e) OM images of MoS_2 films thinned under beam currents of 7.7 pA, 16 pA, 48 pA, and 77 pA, respectively.

Table S1 Raman shift of the CVD grown MoS_2 film before and after FIB thinning for different times.

Thinning time (s)	E1 2g peak frequency (cm^{-1})	A_{1g} peak frequency (cm^{-1})	Difference of E1 2g and A_{1g} (cm^{-1})
0	383.2	408.4	25.2
2	383.6	407.4	23.8
4	383.6	406.2	22.6
6	383.5	404.3	20.8
8	384.6	403.1	18.5