

The Growth Mechanisms and Device Applications of Large-area MoS₂ Films Prepared by Sulfurization of Pre-deposited Molybdenum on Sapphire Substrates

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Abstract

Compared with the zero-bandgap 2-D crystal, graphene, transition metal dichalcogenides (TMDs) such as MoS₂ have revealed its potential for transistor applications due to the device's high ON/OFF ratios. In nowadays, one most commonly adopted approach for large-area TMD growth is by chemical vapor deposition (CVD) [1]. Although large-area and uniform MoS₂ films can be prepared by this method, the choice of suitable precursors and the in-capability of selective growth are the major disadvantages of this approach. In this report, we have demonstrated large-area MoS₂ growth by using sulfurization of Mo thin films pre-deposited on sapphire substrates in a hot furnace at 800 °C. By fixing the thickness of Mo films to 1 nm and decreasing the S powder weight, the Raman peak difference remains the similar, which suggest that the same average MoS₂ layer numbers are obtained for samples grown under different S powder amounts as shown in Fig. 1 (a). With the lowest FWHM value 2.48 cm⁻¹ corresponding to the in-plane vibration Raman peak E_{2g}¹ of the sample prepared with 1.5 g S power, it is demonstrated that large-area and uniform MoS₂ films with high crystalline quality can be obtained by using this approach. The XPS spectrums of the MoS₂ samples prepared with different amounts of S powder are shown in Fig. 1 (b). With S powder amount lower than 1.5 g, the peak corresponding to the Mo-O bonding located at 236 eV is observed. The atomic ratio of S : Mo is also reduced to 1.6 for this sample, which suggests that with insufficient S, the film is not completely sulfurized such that Mo oxides would be observed. The results also indicate that the deposited Mo films will be oxidized after exposing to air. The cross-sectional HRTEM image of the sample grown with < 1.5 g S is shown in Fig. 2 (a). As shown in the figure, there are small clusters spreading over the sample surfaces. The HRTEM image with higher magnification of the same sample is shown in Fig. 2 (b). It seems that the sample surface including the small clusters is covered by few-layer MoS₂. To verify the chemical compositions of the small clusters, EDX mapping of elements S and O are shown in Fig. 2 (c) and (d). As shown in the figure, it seems that the small clusters contain both elements S and O. Since the small clusters are fully covered by few-layer MoS₂ as shown in Fig. 2 (b), it is possible that the S signal comes from the covering MoS₂ films. In this case, it is reasonable to assume that the clusters contain mostly Mo oxides. Therefore, under S insufficient condition, the Mo oxides will undergo coalescence at first and form small clusters on the substrates. After that, sulfurization will still takes place and form few-layer MoS₂ covering the sample surface. The results indicate that Mo oxides will migrate on substrate surfaces at 800 °C. Under S sufficient condition, both Mo oxides and S adatoms will migrate on the surface. Epitaxially growth of MoS₂ will take place such that large-area and uniform MoS₂ films can be obtained. The bottom-gated MoS₂ transistors are fabricated by transferring the MoS₂ films prepared by sulfurizing 0.2, 0.5 and 1.0 nm Mo with 1.5 g S to 300 nm SiO₂/Si substrates with pre-deposited Au/Ti electrodes. The I_D-V_{GS} curves of the three devices are shown in Fig. 3. The corresponding MoS₂ layer numbers are 1, 3 and 5 as shown in the HRTEM images inserted in Fig. 3. The mobility values of these three devices are 0.02, 0.07 and 0.08 cm²/V-s, respectively. The On/Off ratios of devices increase from 3.0 × 10³, 1.2 × 10⁴ to 2.5 × 10⁴ with increasing MoS₂ layer numbers. The results have demonstrated that multi-layer MoS₂ transistors fabricated by using sulfurization of Mo films are promising for practical applications.

References

[1] C. R. Wu *et al.*, J. of Phys. D: Appl. Phys., 48 (2015) 435101.

Figures

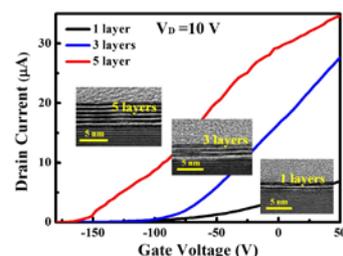
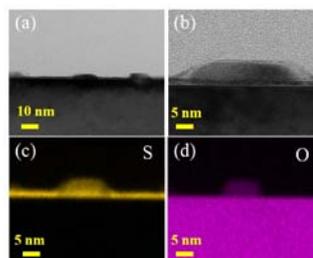
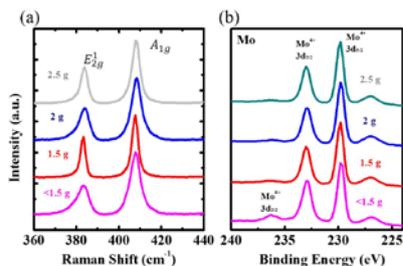


Fig. 1 (a) The Raman and (b) XPS spectra of the samples prepared with different S amounts.

Fig. 2 (a), (b) The cross-sectional HRTEM images and (c), (d) EDX mappings of the sample grown with S < 1.5 g.

Fig. 3 The I_D-V_{GS} curves of the three devices prepared by sulfurizing 0.2, 0.5 and 1.0 nm Mo with 1.5 g S.