Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Synthesis of Large-Area Monolayer and Few-Layer $MoSe_2$ Continuous Film by Chemical Vapor Deposition without Hydrogen assisted and Formation Mechanism

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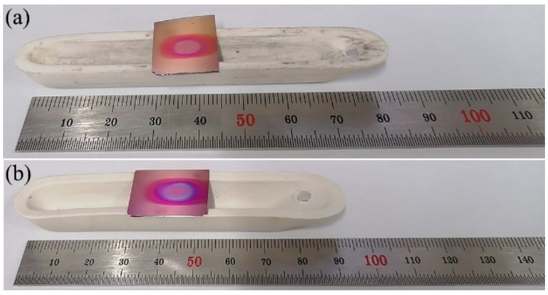

Fig. S1 Photographs of the typical $MoSe_2$ films grown on Si/SiO_2 substrates by using different sizes of alumina boat (a) 86×13×9.0 mm, (b) 95×17×11 mm.

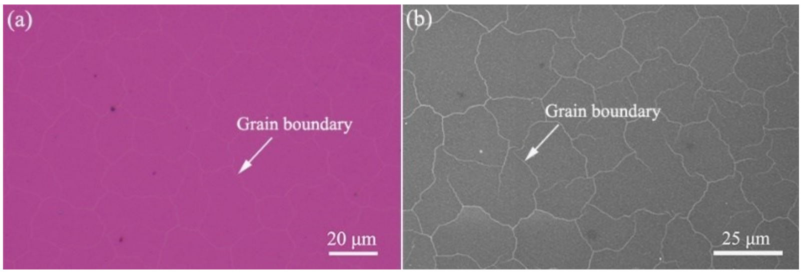

Fig. S2 (a) OM and (b) SEM image of the grain boundaries of monolayer $MoSe_2$ film.

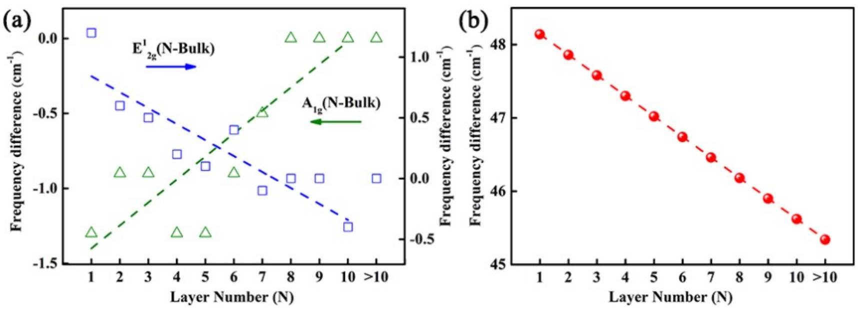

Fig. S3 (a) The frequency differences between the different layer and the bulk for the Raman mode A_{1g} and E^1_{2g} as a function of number of layers (*N*). The dashed lines are the linear fittings. (b) The frequency differences between the between E^1_{2g} and A_{1g} mode calculated by the simulated two formulas in (a) for different layers. The dashed lines are the fitting line in the main text.

The frequency differences between the different layer and the bulk for the Raman mode A_{1g} and E^1_{2g} as a function of number of layers (*N*) are illustrated in Fig. S3(a). For A_{1g} mode, the frequency di□erence $\Delta\omega(A_{1g})$ of *N*L $MoSe_2$ (*N*=1-10L) can be expressed as $\Delta\omega(A_{1g}) = 0.15N - 1.55$ and the value of Adj. R-Square is 0.68. For E^1_{2g} mode, the frequency di□erence $\Delta\omega(E^1_{2g})$ of NL $MoSe_2$ (N=1-10L) can be expressed as $\Delta\omega(E^1_{2g}) = -0.13N+0.97$ and the Adj. R-Square is 0.76. The two values of the Adj. R-Square are both smaller than 0.89 in the main text. As shown in Fig. S3(b), the frequency differences between E^1_{2g} and A_{1g} calculated by the above simulated two formulas are the same as the simulation of the main text.

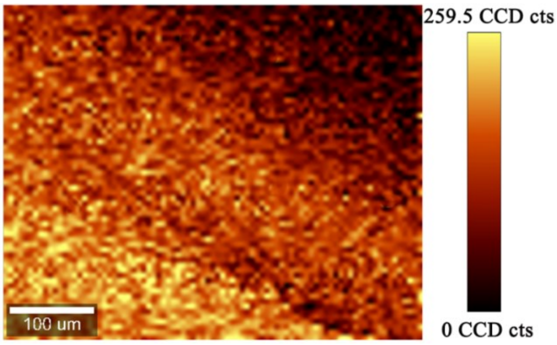

Fig. S4 Peak intensity mapping of Raman mode A_{1g} obtained from monolayer (bottom left) to multilayer (top right) $MoSe_2$ film.

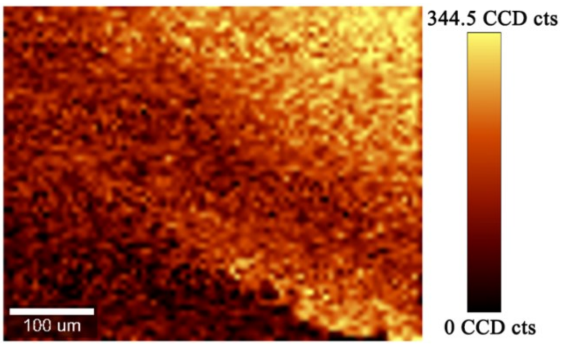

Fig. S5 Raman intensity map of D peak obtained from monolayer (bottom left) to multilayer (top right) $MoSe_2$ film.

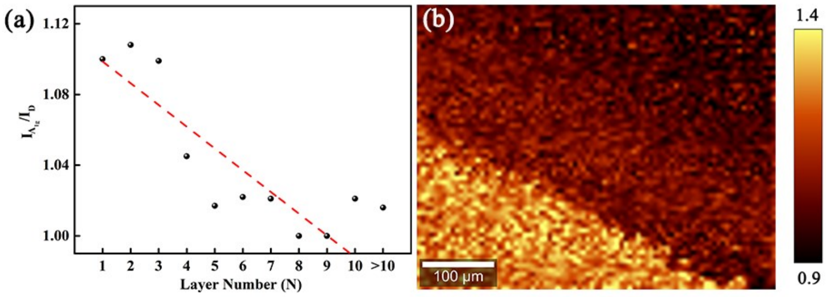

Fig. S6 (a) The peak intensity ratio of A_{1g}/D as function of the layer numbers. The dashed line is guide to the eyes. (b) The mapping of the peak intensity ratio of A_{1g}/D from monolayer (bottom left) to multilayer (top right) $MoSe_2$ film.

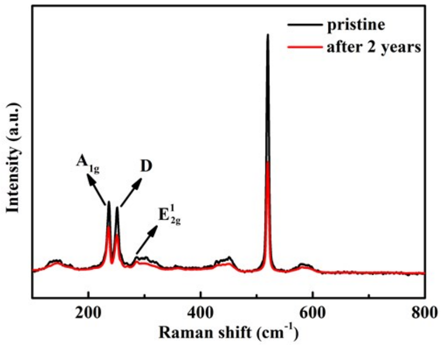

Fig. S7 The Raman spectra of the $MoSe_2$ monolayer freshly prepared and stored for two years.

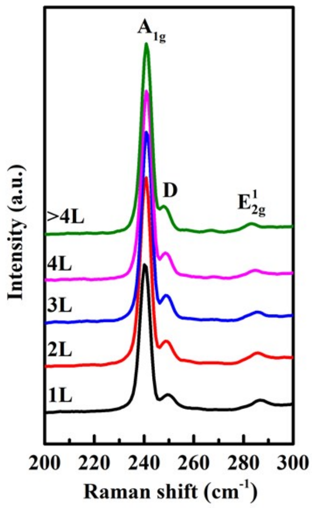

Fig. S8 The Raman spectra of the $MoSe_2$ film with different number of layers synthesized with 1mg MoO_3 and 50 mg Se power as precursors.

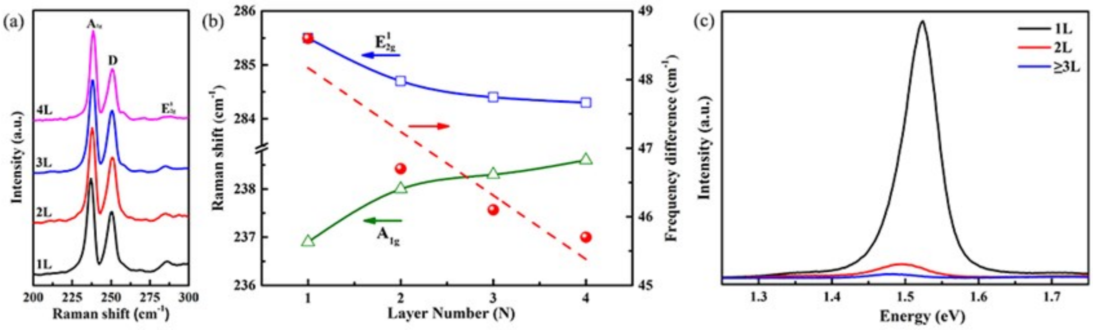

Fig. S9 Spectral analysis of another $MoSe_2$ film synthesized by APCVD method. (a) Raman spectra of $MoSe_2$ film with different number of layers. Labels indicate the corresponding number of layers. (b) Frequency (ω) of E^1_{2g} and A_{1g} and the frequency difference ($\Delta\omega$) between E^1_{2g} and A_{1g} as a function of number of layers (N). (c) PL spectra of monolayer, bilayer and multilayer $MoSe_2$.

Spectral analysis of another $MoSe_2$ film synthesized by APCVD method is shown in Fig. S9. Fig. S9(a) shows the Raman spectra of $MoSe_2$ film with different number of layers (N=1-4L). The frequency (ω) of E^1_{2g} and A_{1g} and the frequency difference ($\Delta\omega$) between E^1_{2g} and A_{1g} as a function of number of layers (N) are shown in Fig. S9(b). The position of E^1_{2g} and A_{1g} show opposite trends with increasing thickness from 1L to 4L. The A_{1g} Raman mode shows a blue shift while the E^1_{2g} Raman mode exhibits a redshift. The frequency difference between A_{1g} and E^1_{2g} decreases gradually with the increase of $MoSe_2$ film layers as shown in Fig. 9S(b), and the frequency difference $\Delta\omega(E^1_{2g} - A_{1g})$ of NL $MoSe_2$ (N=1-4L) can be expressed as $\Delta\omega(E^1_{2g} - A_{1g}) = 49.0 - 0.9N$. Fig. S9(c) shows the PL spectra of monolayer, bilayer and multilayer $MoSe_2$. As the thickness of $MoSe_2$ film increased the PL peak shows redshifted. And the PL intensities of bilayer and multilayer regions are significantly reduced compared to that of the monolayer. These spectral analysis results are similar to those in the main text.

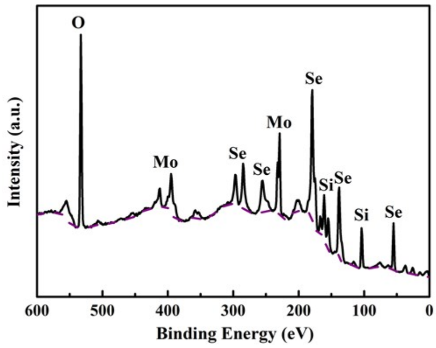

Fig. S10 XPS spectrum of the sample. Four elements are present: Mo and Se (from the $MoSe_2$ film), Si and O (from the substrate).