Supplementary Information

Film thickness-induced optical and electrical modifications in large-area few-layer 2H-$MoSe_2$ grown by MBE

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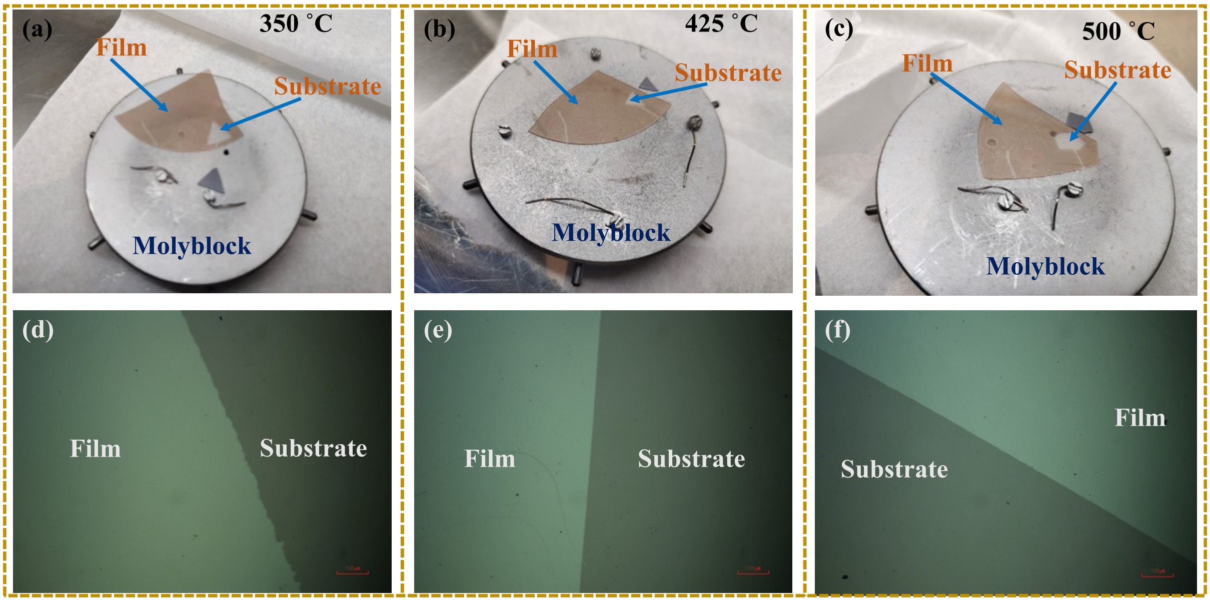

Fig. S1. Image of as-grown samples at (a) 350 ˚C, (b) 425 ˚C and (c) 500 ˚C, (d), (e) and (f) are the optical microscope image of the films, clearly showing step between substrate and film.

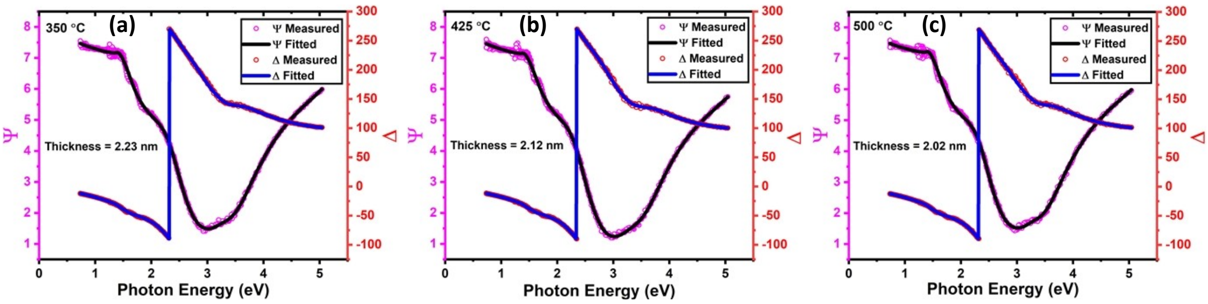

Fig. S2. Ψ and Δ fitting of (a) 350 ˚C, (b) 425 ˚C, (c) 500 ˚C grown samples by spectroscopic ellipsometry.

Fig. S2 shows the spectroscopic ellipsometry Ψ and Δ variation and fitting of 350 ˚C, 425 ˚C and 500 ˚C grown samples. The thickness of the 350 ˚C, 425 ˚C and 500 ˚C grown films are 2.23 nm, 2.12 nm, and 2.02 nm, respectively. Slight variations of thickness due to the growth temperature effects.

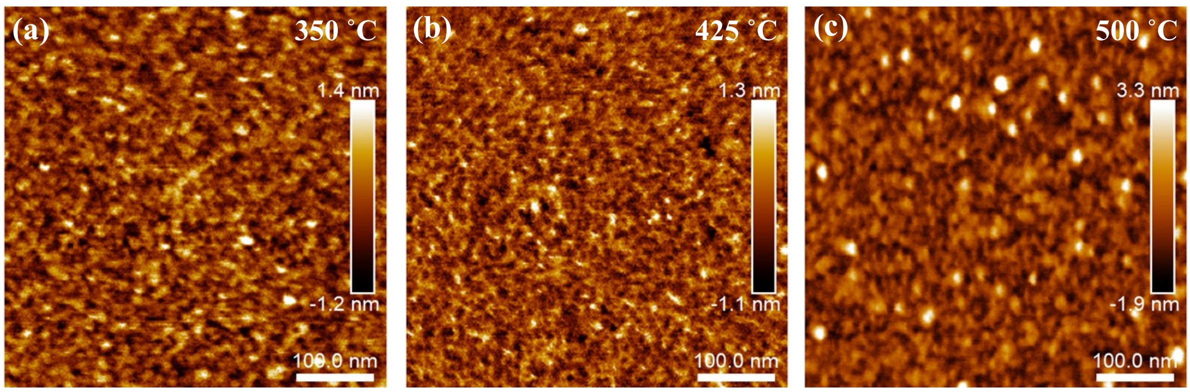

Fig. S3. AFM surface topography of (a) 350 ˚C, (b) 425 ˚C, and (c) 500 ˚C grown samples (500 × 500 nm^2 area).

Fig. S3 shows a 500 × 500 nm^2 area AFM image of $MoSe_2$, showing small grains of $MoSe_2$ mostly merged and making a uniform surface.

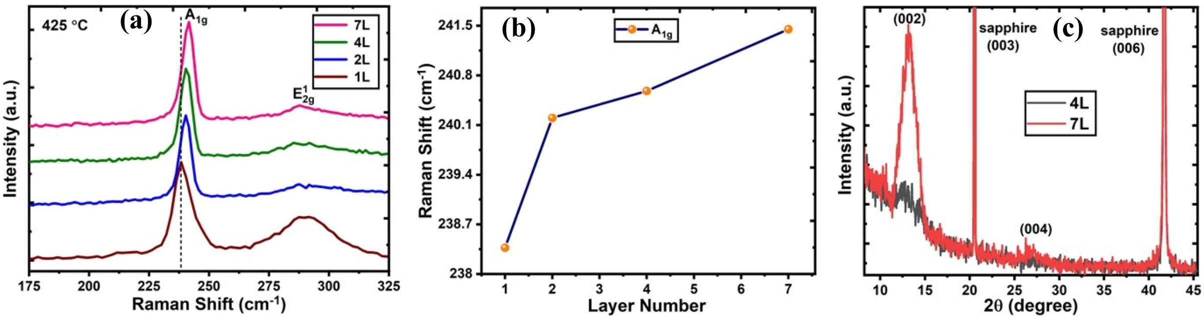

Fig. S4. (a) Raman spectroscopy of different thicknesses of $MoSe_2$ layers, (b) Shift of Raman A_{1g} peak position with layer number, (c) XRD of 4L and 7L $MoSe_2$ films.

Fig. S4 shows the Raman spectroscopy of 1L, 2L, 4L and 7L of $MoSe_2$ films, there is a redshift of the Raman A_{1g} peak position with increasing layer thickness. Raman A_{1g} is the out-of-plane vibration mode of $MoSe_2$, as the number of layers is added, the A_{1g} vibration mode is also affected by that and as a result, there is a redshift in the peak position. We only found XRD for 4L and 7L samples, as 1L and 2L samples are too thin to get any XRD intensity. The XRD peak was found from the (002) family of planes, which indicates out-of-plane van der Waals growth of $MoSe_2$ films.

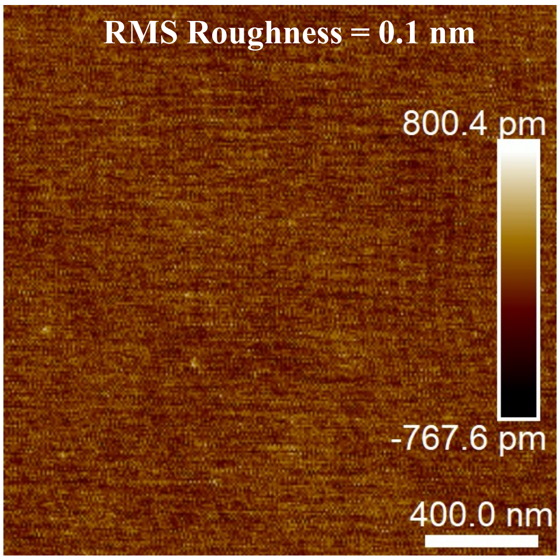

Fig. S5. AFM image of the clean sapphire substrate.

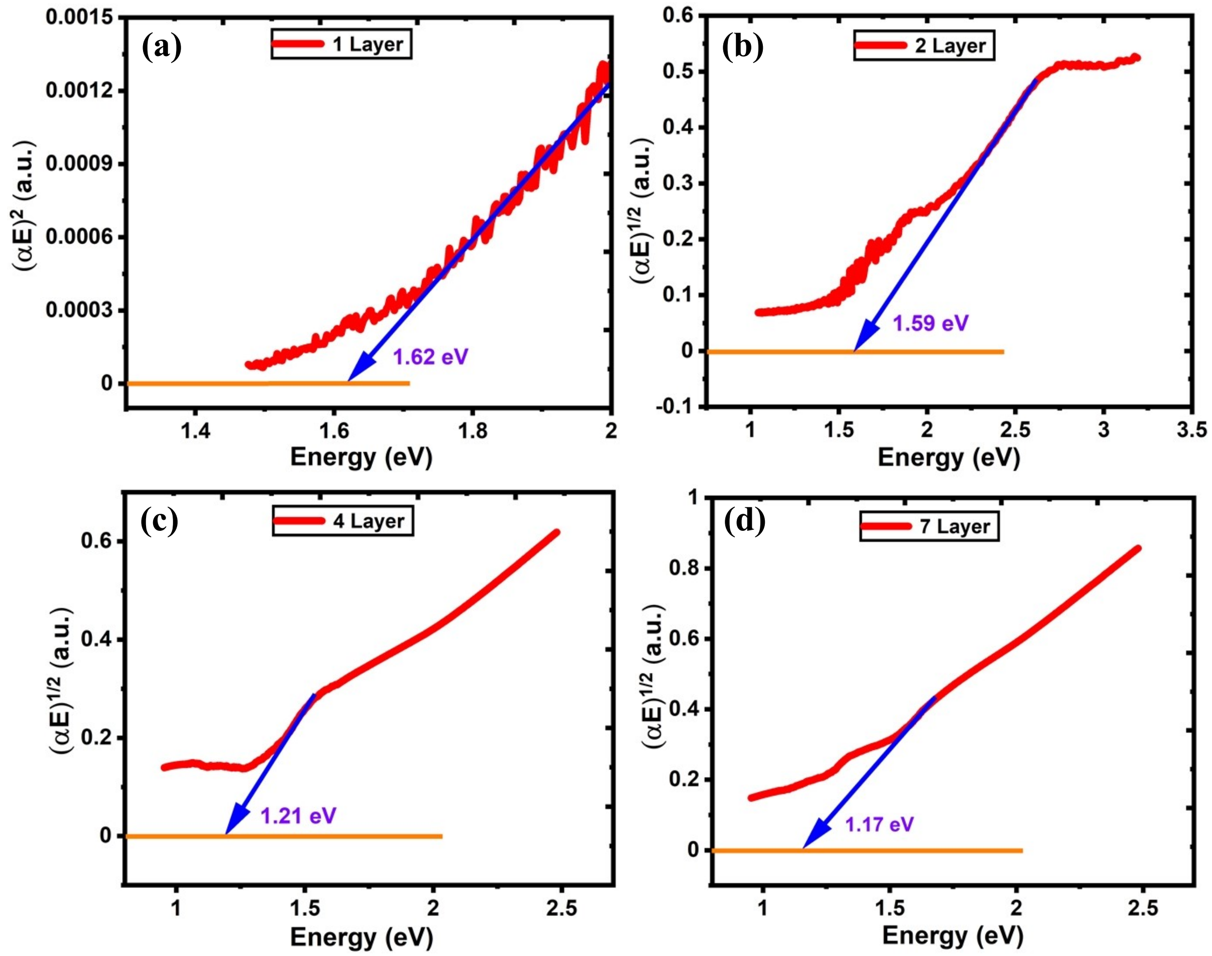

Fig. S6. UV-Vis band gap of 1L, 2L, 4L and 7L $MoSe_2$ films.

Calculation of lattice constant from XRD:

2θ value corresponding to the (002) peak is 13.22 degrees.

According to Bragg's law, $2d \sin\theta = n\lambda$ and $d = C/\sqrt{(h^2+k^2+l^2)}$, where C is out of the plane (c-axis) lattice constant.

Now, putting the value of h=0, k=0, l=2 we get $d = C/2$.

By putting $d = C/2$, we get

$C \sin\theta = n\lambda$.

Now, for (002) peak n = 1. The value of λ is 1.54 Å for the Cu k-alpha x-ray source.

Putting all these values in the equation, we get

$C \sin(13.22/2) = 1.54$

or C = 13.3 Å.

Therefore, the out-of-plane (c-axis) lattice constant (C) of the $MoSe_2$ film is 13.3 Å based on the (002) peak.

Ellipsometer parameters:

All the Tauc-Lorentz parameters for 2L, 4L and 7L have been given below.

Oscillators No.	Tauc-Lorentz Parameters	2 Layer	4 Layer	7 Layer
1.	E_g (eV)	1.579	0.982	0.991
	E_0 (eV)	2.585	2.386	2.305
	A	15.1905	79.0042	57.2263
	B	0.710	0.943	0.575
2.	E_g (eV)	1.119	1.232	1.109
	E_0 (eV)	3.381	4.346	4.296
	A	127.8341	242.0918	352.6824
	B	0.863	0.861	0.860
3.	E_g (eV)	1.361	1.091	1.140
	E_0 (eV)	3.413	4.381	4.395
	A	134.0846	79.3382	128.9343
	B	0.460	0.669	0.684
4.	E_g (eV)	1.581	1.201	1.118
	E_0 (eV)	3.845	4.789	4.978
	A	37.5584	18.1531	52.4017
	B	0.556	0.534	0.408
5.	E_g (eV)	1.539	1.221	1.221
	E_0 (eV)	3.102	3.531	3.650
	A	334.5042	215.6816	218.5192
	B	0.468	0.459	0.464
6.	E_g (eV)	1.585	1.229	1.171
	E_0 (eV)	3.396	3.607	3.898
	A	12.4537	56.8666	77.4226
	B	0.437	0.676	0.633

Here, $\mathbf{E_g}$**:** Band gap, $\mathbf{E_0}$**:** Peak transition energy, **A:** Amplitude parameter, **B:** Broadening term [1, 2].

All the Lorentz oscillator model parameters for 1L $MoSe_2$ film have been given below.

Oscillators No.	Lorentz Oscillator Parameters	1 Layer
1.	E_n (eV)	4.353
	A	8.011463
	B	5.3143
2.	E_n (eV)	3.171
	A	37.339463
	B	1.2389
3.	E_n (eV)	3.205
	A	34.346659
	B	1.1510
4.	E_n (eV)	2.969
	A	1.744886
	B	0.3943
5.	E_n (eV)	2.798
	A	0.783280
	B	0.2307

Here, $\mathbf{E_n}$**:** Center energy, **A:** Amplitude parameter, **B:** Broadening term [3].

References:

1 G. E. Jellison and F. A. Modine, *Appl. Phys. Lett.*, 1996, **69**, 371–373.

2 J. Xie, D. Zhang, X. Q. Yan, M. Ren, X. Zhao, F. Liu, R. Sun, X. Li, Z. Li, S. Chen, Z. B. Liu and J. G. Tian, *2D Mater.*, 1998, 313–314, 394–397.

3 R. A. Synowicki, *Thin Solid Films*, 1998, **313–314**, 394–397.