

## Electronic Supplementary Information for

### Stepwise Growth of Crystalline MoS<sub>2</sub> in Atomic Layer Deposition

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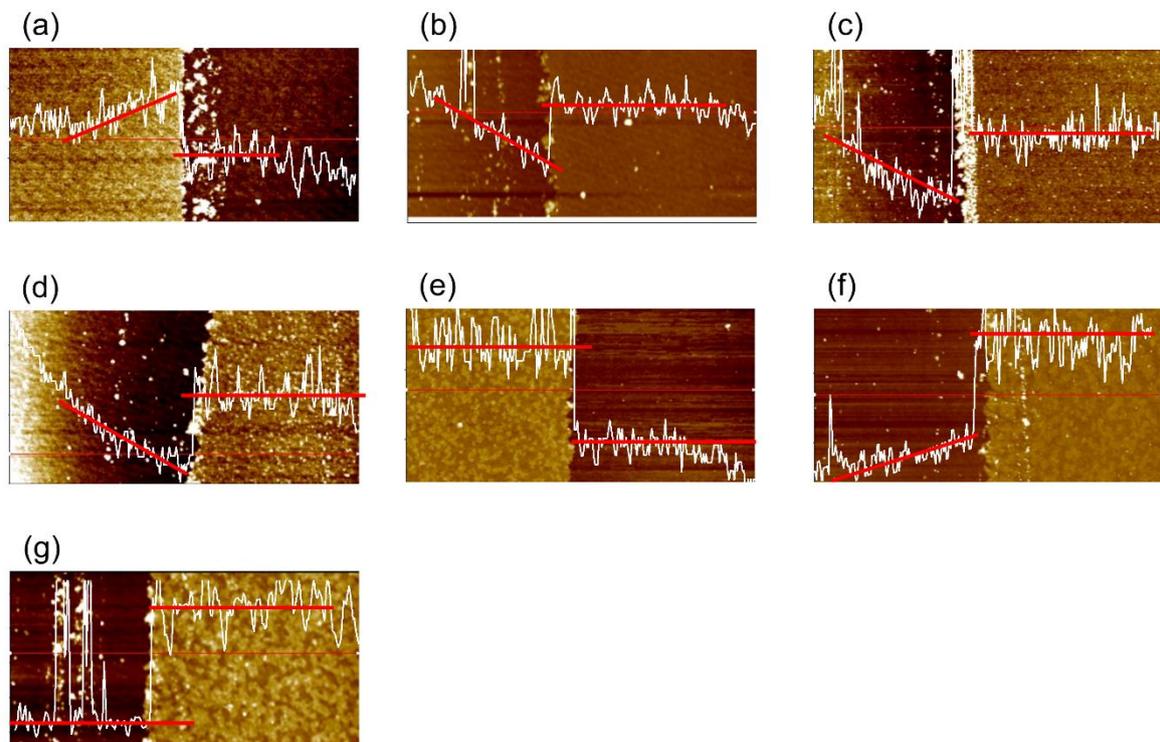
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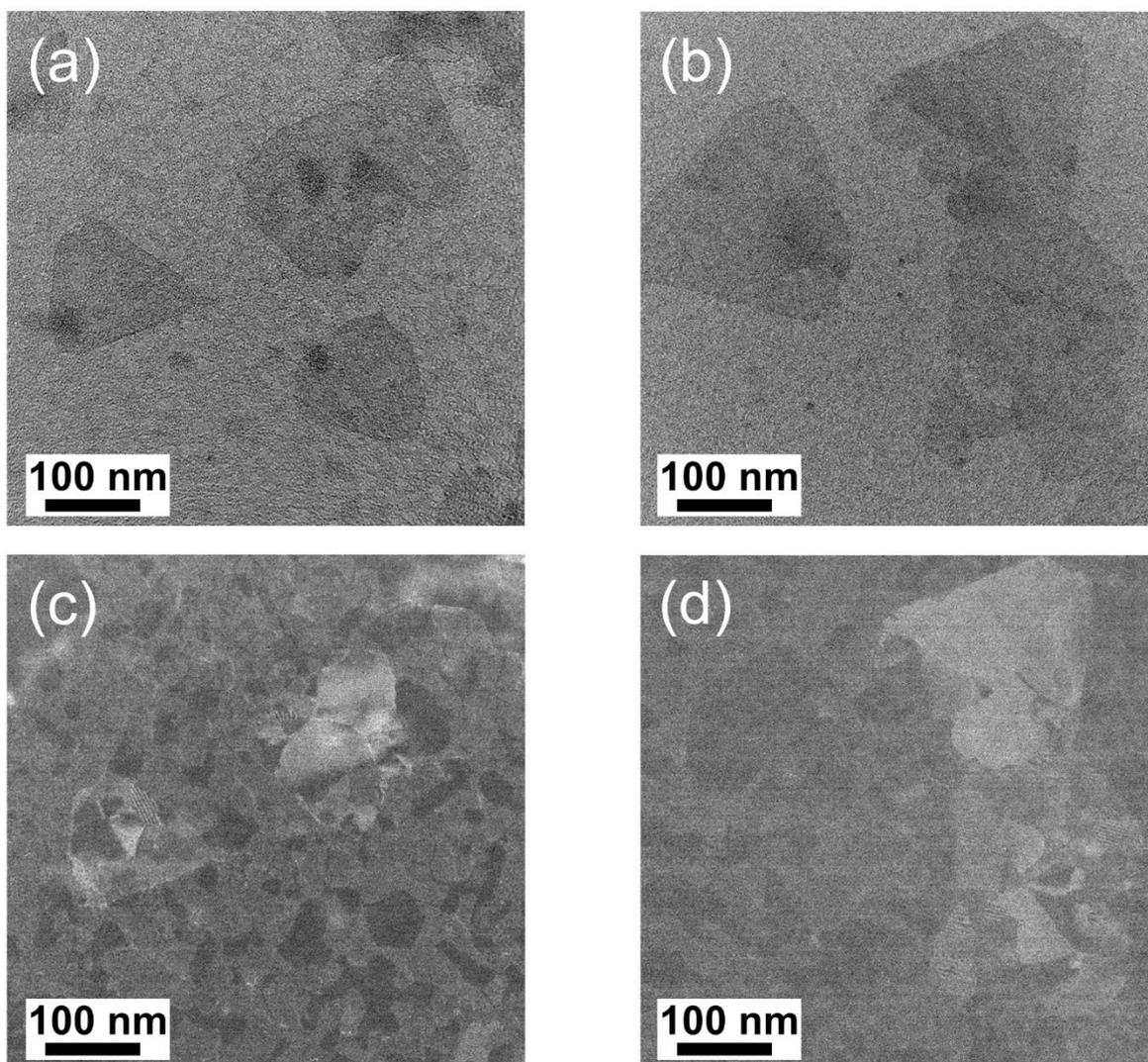
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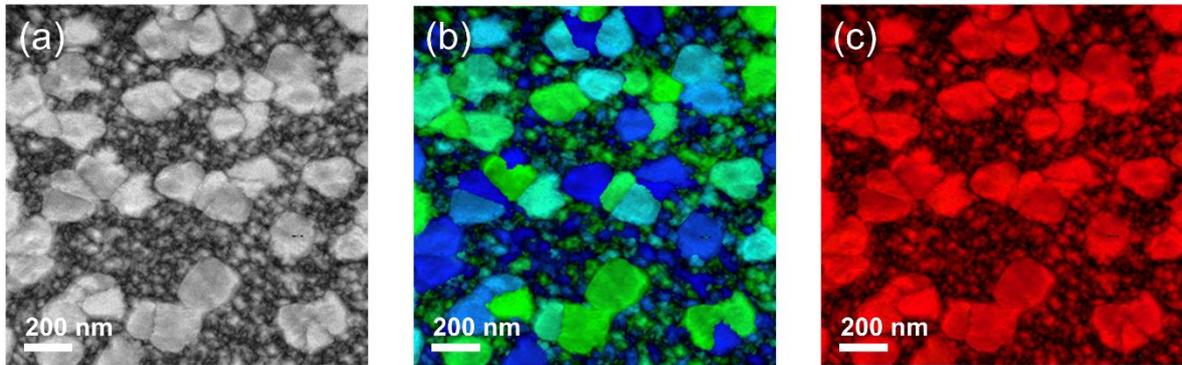
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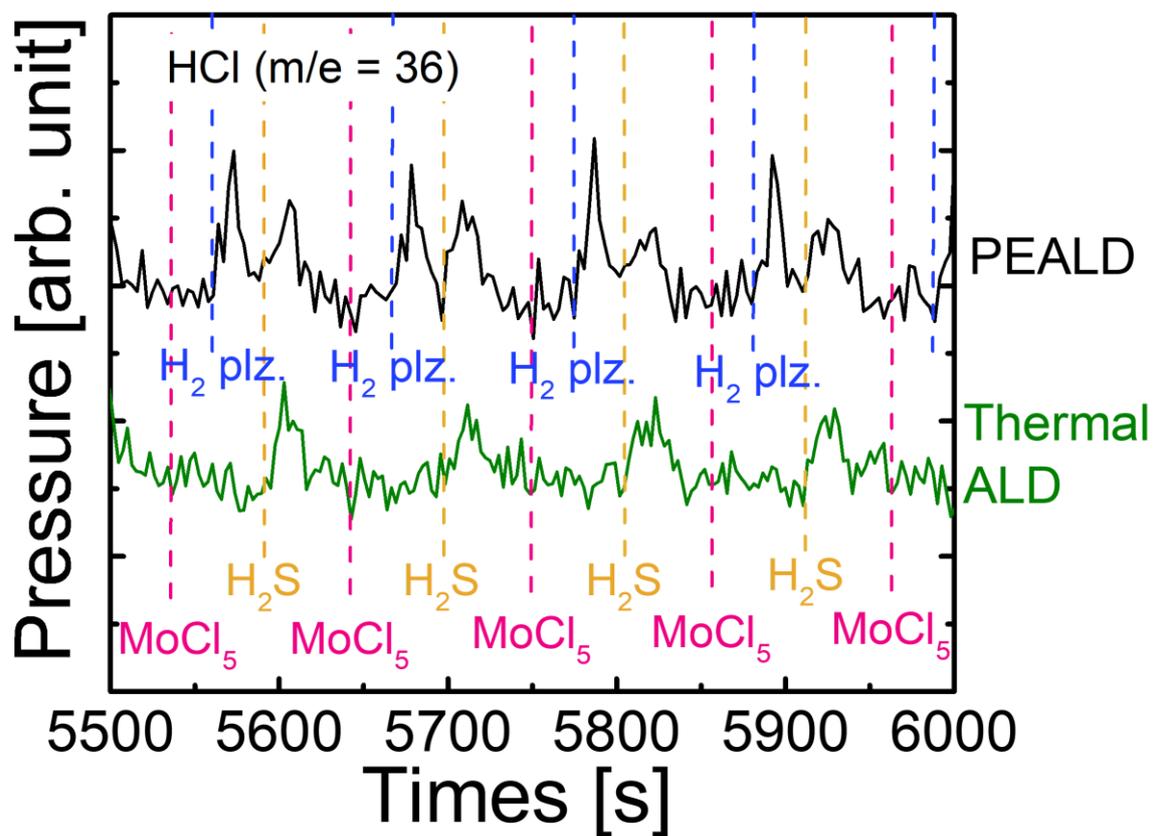
**Figure S1.** AFM images and height profiles of the PEALD-grown MoS<sub>2</sub> on SiO<sub>2</sub>/Si for (a) 50, (b) 100, (c) 200, (d) 300, (e) 400, (f) 500, and (g) 700 cycles.



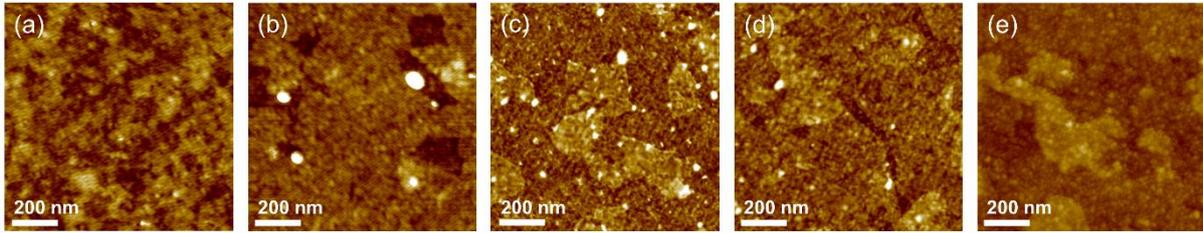
**Figure S2.** Bright-field TEM images of (a) 400-cycle-ALD-grown and (b) 700-cycle-ALD-grown MoS<sub>2</sub>. Dark-field TEM images of (c) 400-cycle-ALD-grown and (d) 700-cycle-ALD-grown MoS<sub>2</sub>.



**Figure S3.** (a) Index map and (b) In-plane and out-of-plane orientation maps combined with index map of PEALD-grown MoS<sub>2</sub> via ASTAR analysis.



**Figure S4.** Quadrupole mass spectrometry (QMS) signal of  $m/e = 36$  during the ALD process with  $MoCl_5$ - $H_2$  plasma- $H_2S$  and  $MoCl_5$ - $H_2S$  sequences.



**Figure S5.** AFM images of the thermal ALD-grown MoS<sub>2</sub> on SiO<sub>2</sub>/Si for (a) 100, (b) 300, (c) 400, (d) 500, and (e) 700 cycles.