Electronic Supplementary Material (ESI) for Nanoscale Advances.

Supporting Information

Structural and electronic properties of MoS_2 and $MoSe_2$ monolayers grown by chemical vapor deposition on Au(111)

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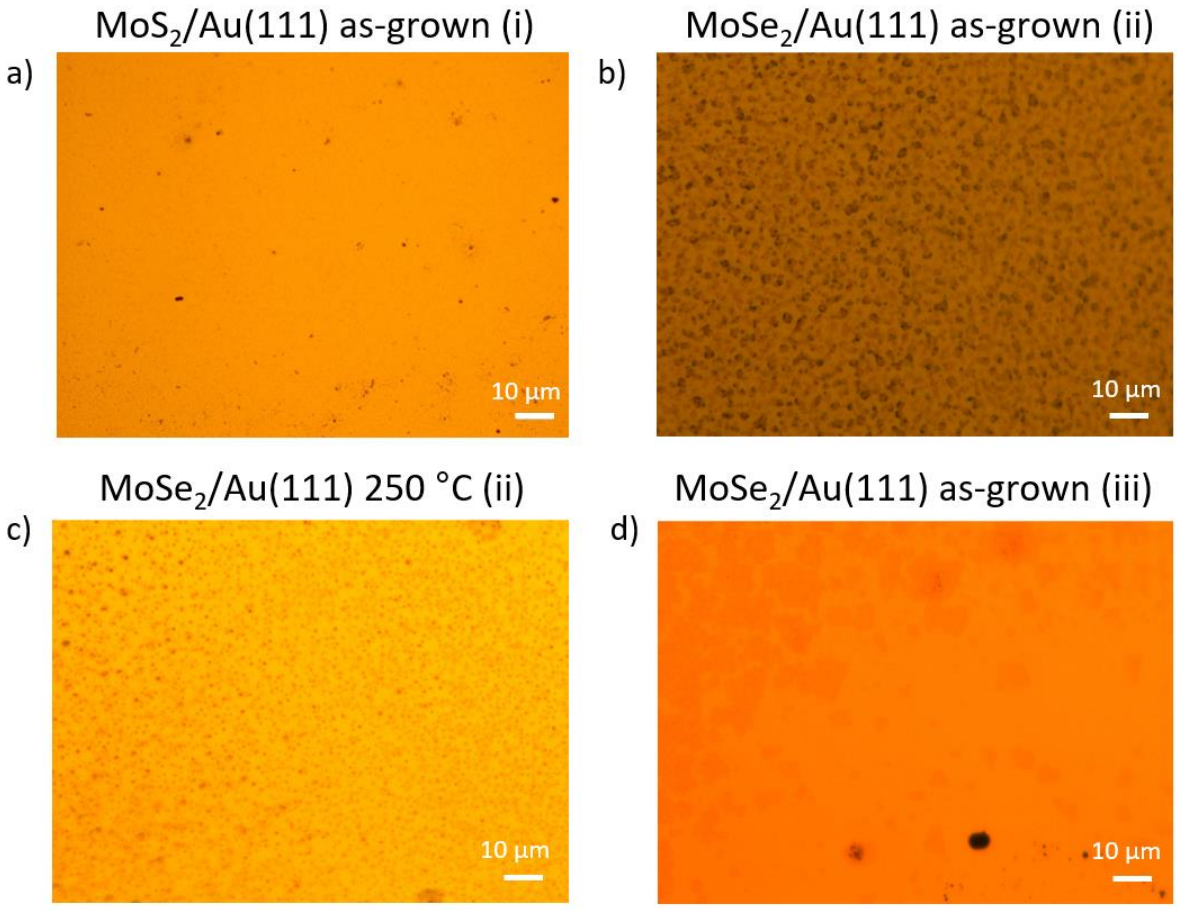

Figure S1: Optical microscope (OM) images of (a) as-grown MoS_2 (i), (b) as-grown and (c) annealed at 250 °C (in UHV) $MoSe_2$ (ii), as well as (d) as-grown $MoSe_2$ (iii) on Au(111). For MoS_2 a nearly continuous monolayer film is visible. For (b)-(d), the $MoSe_2$ crystals appear with a darker contrast compared to the gold substrate. The crystal size is from several to tens of micrometers. Sample (ii) and (iii) differ in the growth conditions. For sample (iii) a higher Se flow (higher Knudsen cell temperature) were used, which results in a higher Se concentration.

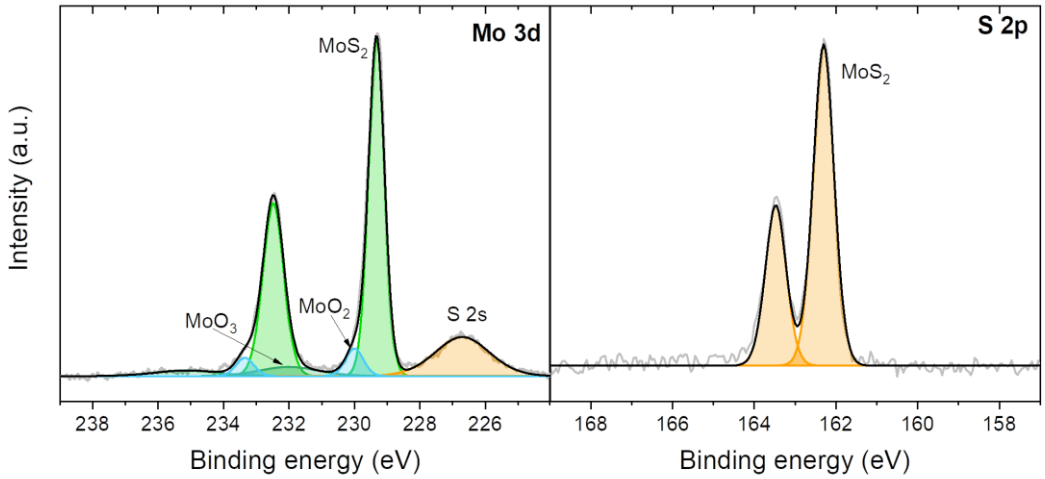

Figure S2: High-resolution XP spectra of MoS_2 monolayers on Au(111). For the growth of this sample the temperature of the Knudsen cell with sulfur was slightly reduced (~ 180 °C) compared to the MoS_2 sample discussed in Fig. 2a of the main paper. The Mo 3d and S 2p spectra show the peaks with similar binding energies such as in Fig. 2a. Note that the Mo to S ratio of this sample is (2.1 ± 0.2):1 which is lower than that of the sample presented in Fig. 2a of the main manuscript.

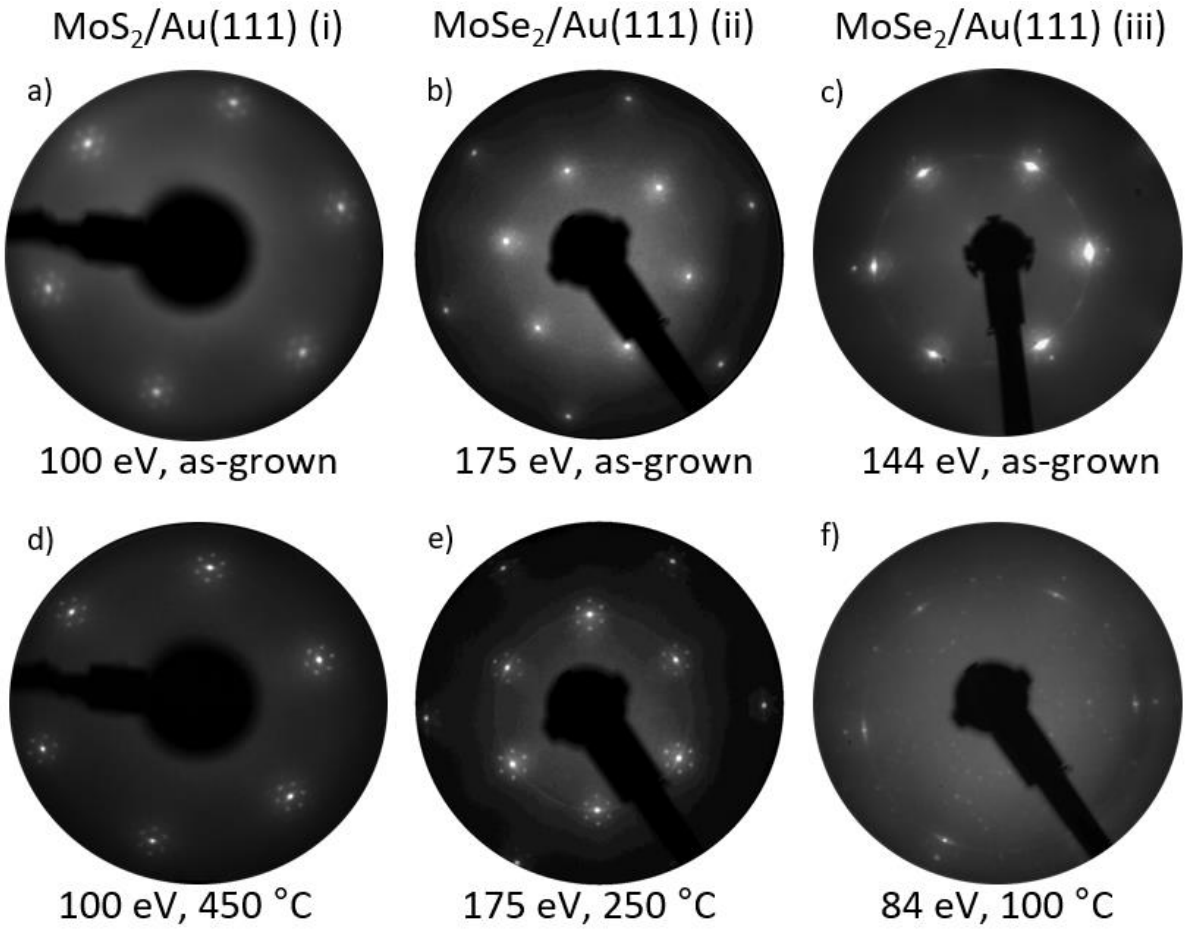

Figure S3: LEED images of (a) as-grown and (d) annealed at 450 °C MoS_2 (i), (b) as-grown and (e) annealed at 250 °C $MoSe_2$ (ii), (c) as-grown and (f) annealed at 100 °C $MoSe_2$ (iii) on Au(111). After annealing the contrast of the moiré pattern of MoS_2 sample (i) and $MoSe_2$ sample (ii) increases significantly. For as-grown $MoSe_2$ sample (iii) we observe the moiré pattern similar to sample (ii), but with a weaker contrast. After annealing at 100 °C, the moiré structure vanishes and the structure of the Se interlayer appears. The $MoSe_2$ lattice is still visible. Used LEED devices: (a,d) SMCP (Scienta Omicron); (b,c,e) BDL800IR MCP2 (OCI); (f) MCP2-SpectaLEED (Omicron).

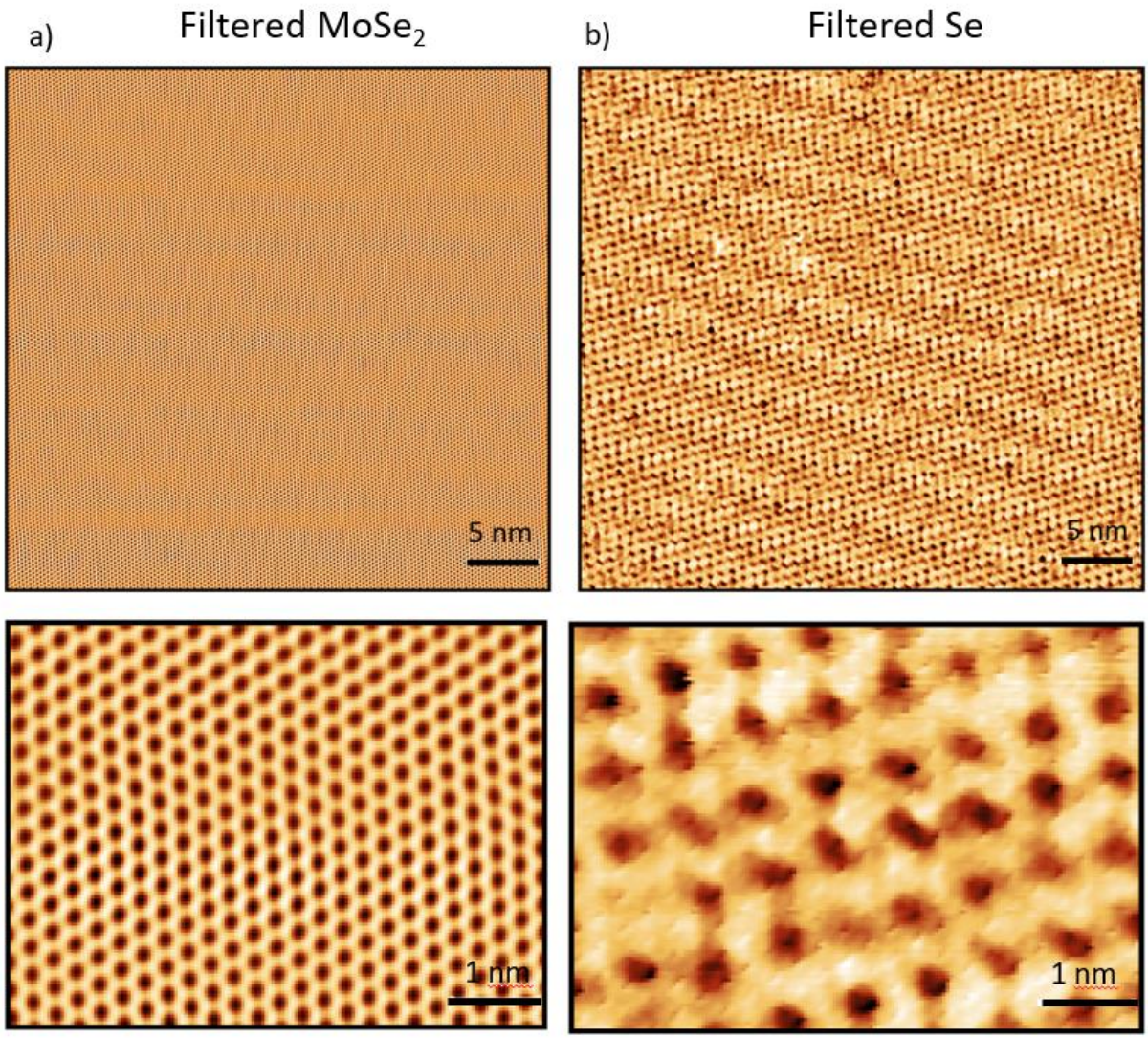

Figure S4: Fourier filtered images of the LT-STM image in Fig. 3c of $MoSe_2$ sample (iii) on Au(111). On the one hand, in (a) the Se structure is filtered out. We see a hexagonal structure corresponding to the $MoSe_2$ monolayer. On the other hand, in (b) the $MoSe_2$ structure is filtered out. So, the Se_8 superstructure[1] discussed in Fig. S5 below is visible. Combined with our Raman spectroscopy results, this confirms that the $MoSe_2$ monolayer is on top of the Se interlayer on Au(111). Measuring conditions: 4.2 K, 0.1 V, 80 pA.

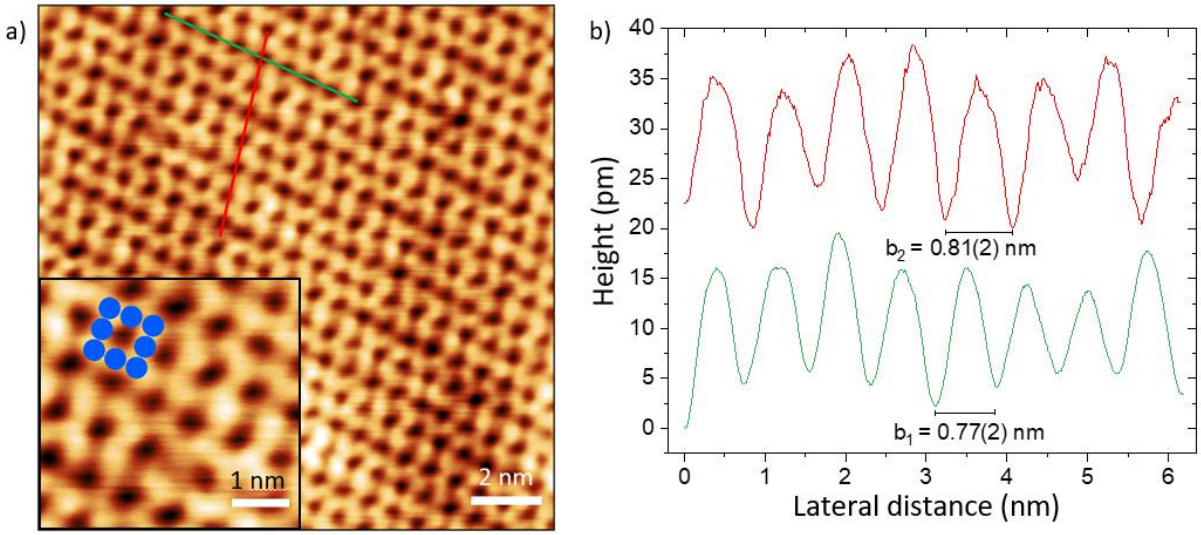

Figure S5: (a) LT-STM image of the Se_8 superstructure[1] on Au(111). This structure was taken at positions without $MoSe_2$ and consists of 8 selenium atoms arranged in a square (blue circles in the inset). The interior of the square (dark area) is empty resulting in a lower STM height. The lengths of the lattice vectors obtained from (b) line scans are around 0.8 nm with an enclosing angle of ~ 80 °. Measuring conditions: 4.2 K, 0.1 V, 30 pA).

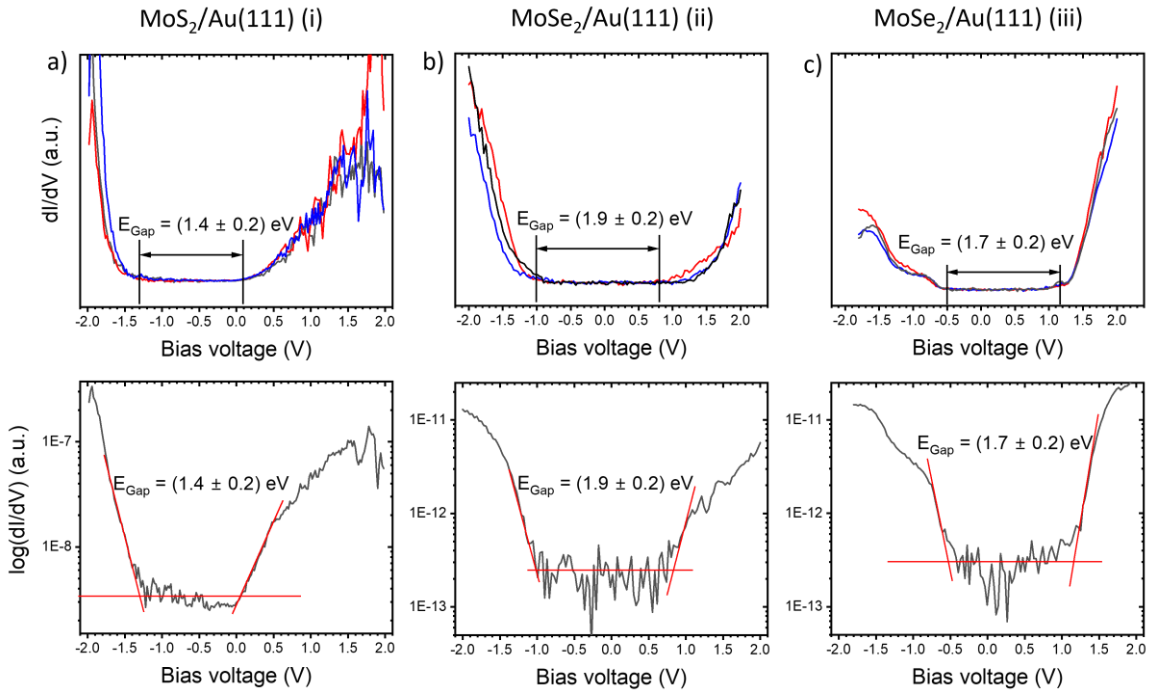

Figure S6: STS data of (a) MoS_2 sample (i) annealed at 100 °C, (b) $MoSe_2$ sample (ii) annealed at 250 °C and (c) $MoSe_2$ sample (iii) annealed at 100 °C. These data were acquired from the same samples represented in the STM images of Fig. 3 in the main text. In the top part three representative ST spectra for each sample are depicted. The onsets of the valence and conduction bands vary depending on the measured sample position, which might be due to point defects visible in the STM images. Yet, a detailed study of defects by STS is beyond the scope of this work. In the bottom part the ST spectra are displayed on a logarithmically scaled dI/dV axis for estimation of the onsets of the valence and conduction bands.[2] The onsets were determined based on the linear extrapolation of the slopes of valence and conduction bands intersecting with the horizontal region within the band gap.

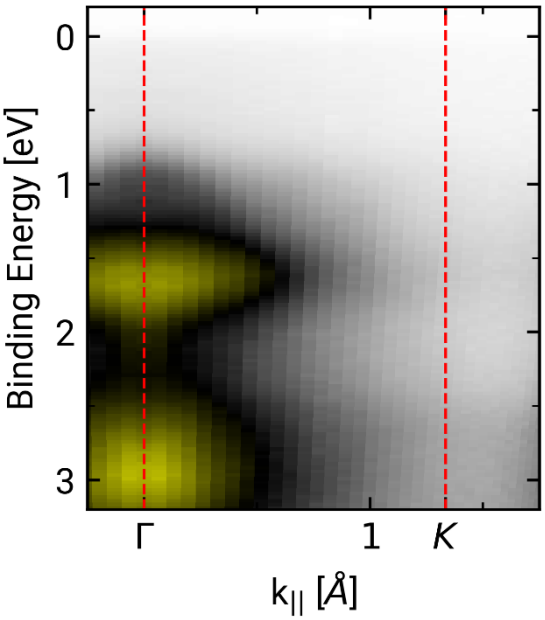

Figure S7: Angle-resolved UP spectra of as-grown $MoSe_2$ sample (ii) on Au(111). The band structure along Γ-K direction is shown. Smeared features at the Γ point are visible whereas no significant features are seen at the K point. That means that the $MoSe_2$ bands appearing after annealing at 100 °C or 250 °C (see Figs. 5b, c) cannot be measured because of contaminations on top of the samples.

References

1. Liu, B.; Zhuang, Y.; Que, Y.; Xu, C.; Xiao, X., STM study of selenium adsorption on Au (111) surface. *Chin. Phys. B* **2020,** *29* (5), 056801.
2. Ugeda, M. M.; Bradley, A. J.; Shi, S.-F.; Da Jornada, F. H.; Zhang, Y.; Qiu, D. Y.; Ruan, W.; Mo, S.-K.; Hussain, Z.; Shen, Z.-X., Giant bandgap renormalization and excitonic effects in a monolayer transition metal dichalcogenide semiconductor. *Nat. Mat.* **2014,** *13* (12), 1091-1095.