Supporting Information

Small-rotation-angle moiré structures of 2H $TaSe_2$ monolayers on Au(111)

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1. Experimental details

All experiments were carried out in an Omicron Surface Science cluster system equipped with two ultrahigh vacuum (UHV) chambers. Growth of $TaSe_2$ was conducted in the preparation chamber with a base pressure of ~2×10^{-9} mbar. Before growth, Au(111) was cleaned by repeating cleaning cycles of Ar^+ sputtering at 1 kV and Ar pressure of ~1.0×10^{-6} mbar at room temperature for 5 min and following annealing at 500 ^{o}C for 15 min. The cleanliness of Au(111) was checked by low electron energy diffraction (LEED) and STM. The appearance of clean and sharp hexagonal diffraction patterns in LEED, along with long-range herringbone reconstruction in STM was considered as the criterion for a clean Au(111) surface in this study. Se was deposited from a home-built thermal evaporator with the temperature of Se source (Alfa Aesar, 99.999%) at 210 ^{o}C for 10 min. Ta was deposited from a UHV compatible home-built sputterer[1] with a Ta target (Alfa Aesar, 99.997%). The high voltages for grid and target were 300 V and 3 kV, respectively. The emission current was 15 mA at the grid. Background Ar pressure was kept at ~6.0×10^{-5} mbar during Ta sputtering and the deposition process lasted for 10 min. The deposition sequence of Se and Ta and corresponding discussions are mentioned in the main text. Both Se and Ta deposition were carried out when the sample was at room temperature. After deposition, Au was annealed to 400 ^{o}C for 20 min in Se vapor (Se source was kept at ~210 ^{o}C) to prevent lack of Se during high temperature annealing.

2. Characterizations

Scanning tunneling microscopy (STM), X-ray photoelectron spectroscopy (XPS) and high-resolution electron energy loss spectroscopy (HREELS) were conducted in an analysis chamber (connected to the preparation chamber) with a base pressure of ~8×10^{-11} mbar. STM measurements were taken by an Omicron ambient temperature STM with a tungsten tip at a constant current mode (with the tip grounded and sample biased) and STM images were analyzed by WSXM software. XPS was acquired at room temperature with a Mg Kα X-ray radiation using the analyzer-Omicron EAC 125 and the analyzer controller-Omicron EAC 2000. CasaXPS software was used for XPS data analysis. Shirley backgrounds and finite Lorentzian (LF) line shape[2, 3] were employed in the curve fitting. Details of key fitting parameters are listed in Table S1. HREELS was acquired at room temperature with ELS5000 (LK technologies) with the primary electron beam of 5 eV and the incident angle of 65° at a specular scattering geometry.

Table S1. Key fitting parameters for XPS. Binding energy E_b. Spin-orbital splitting Δ_{so} values were referred to the reference[4]. LF stands for finite Lorentzian (LF) line shape.

Component	E_b (eV)	LF parameters	Δ_{so} (eV)
Ta $4f_{7/2}$	23.5	LF(1,1.2,8,30)	1.9
Se $3d_{5/2}$ ($TaSe_2$)	53.7	LF(1,1.2,8,30)	0.9
Se $3d_{5/2}$ (elemental Se)	55.4	LF(1,1.2,8,30)	0.9
Au $5p_{3/2}$	57.4	LF(1,2,3,50)	–

3. Growth results at different experimental conditions when Se was deposited first on the substrate

Since the growth was always conducted in a Se-rich environment, the evolution of surface structures was explored at different deposited Ta amount. Figure S1a-c demonstrate topographies of surface structures after Ta was deposited for 6 min, 10 min and 14 min, followed by annealing at 400 °C, respectively. At a low deposited Ta amount, the surface was covered mainly by particles and occasionally triangle-like islands were found (figure S1a). Depositing Ta for 10 min (the default growth time used for samples in the main text) resulted in the growth of triangular $TaSe_2$ flakes, as well as other irregular species on the surface (figure S1b). $TaSe_2$ flakes exhibit an approximate coverage of 8% of the entire surface while the irregular clusters cover ~69%. These irregular clusters could possibly be ascribed to $TaSe_x$ species which did not grow into good $TaSe_2$ crystalline lattices. Longer Ta deposition time of 14 min resulted in the growth of a bigger coverage of triangular $TaSe_2$ flakes of ~14% (figure S1c). Meanwhile, irregular particles were observed to be attached to the edges and corners of the as-grown flakes.

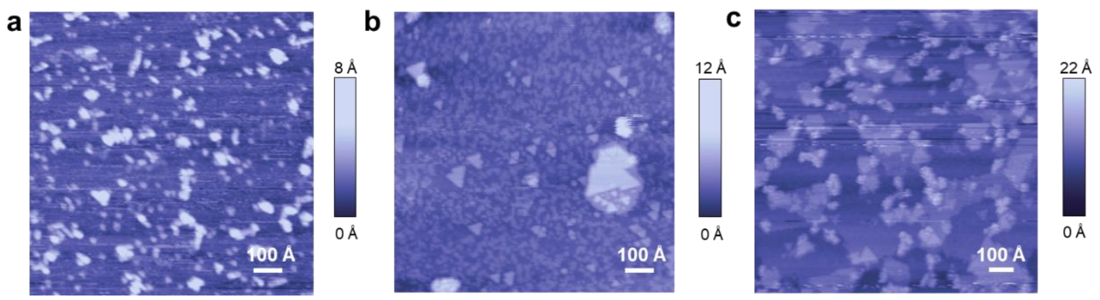

Figure S1. **a-c**, At the condition of Se deposited first on the substrate, STM topographic images of surface structures after growth under different Ta deposition time for 6 min, 10 min and 14 min, respectively.

We also explored the effect of growth temperature. As shown in the following figure S2, sample annealing at a low temperature of 200 °C demonstrates small triangle islands as well as nanobelts grown along three directions (figure S2a). These nanobelts are Se chains grown along the crystallographic orientations of Au(111). After annealing at 400 °C (the default growth temperature used for the samples in the main text), triangular $TaSe_2$ islands were grown (figure S2b). A higher annealing temperature of 600 °C led to decomposition of $TaSe_2$, leaving no triangles but nanoparticles on the surface (figure S2c).

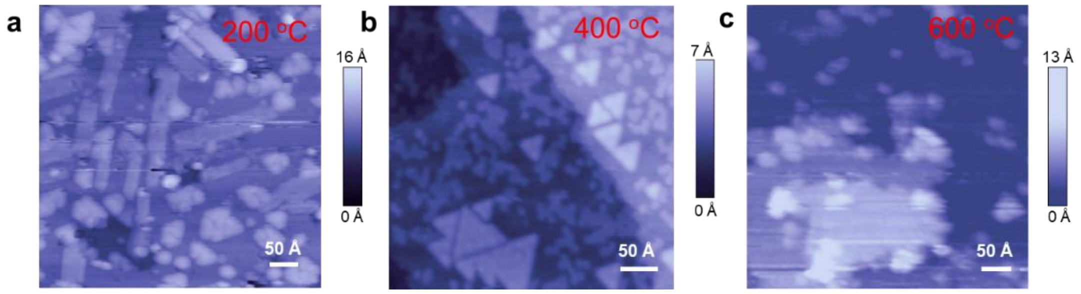

Figure S2. **a-c**, STM topographic images of surface structures after growth under different temperatures at 200 °C, 400 °C, 600 °C, respectively.

4. Additional experimental results

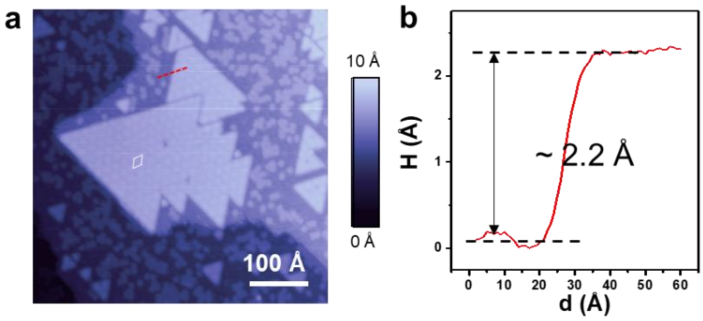

Figure S3. **a**, An STM image of the as-grown $TaSe_2$ monolayer flakes. The white rhombus indicates the moiré unit cell. **b**, A line profile along the red dashed line in (a).

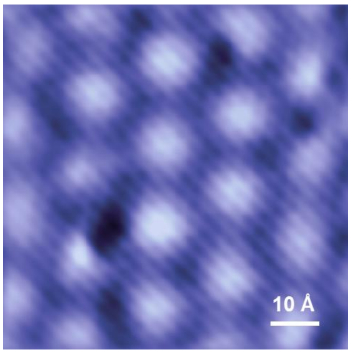

Figure S4. A bigger area STM image on the same $TaSe_2$ flake with that in Figure 1b.

5. Growth results at different experimental conditions when Ta was deposited first on the substrate

Figure S5 shows the results of the same series of experiments (as those in figure S1) at the condition of Ta deposited first on the substrate. The general evolution of the growth results is similar to that when Se was deposited first (figure S1): increased $TaSe_2$ coverage in accordance with the longer Ta deposition time. When Ta was deposited for a short time of 6 min, the as-grown $TaSe_2$ flakes are small on the surface (figure S5a). As Ta was deposited longer for 10 min, bigger triangular $TaSe_2$ flakes were grown (figure S5b). We also notice that the substrate is cleaner (without many irregular species) compared with that after the growth by depositing Se first (figure S1b). This indicates there are fewer $TaSe_x$ species resulting from the growth when Ta is deposited first on the substrate. A longer deposition time of 14 min resulted in the growth of big $TaSe_2$ flakes as well as small and thick $TaSe_2$ islands (figure S5c).

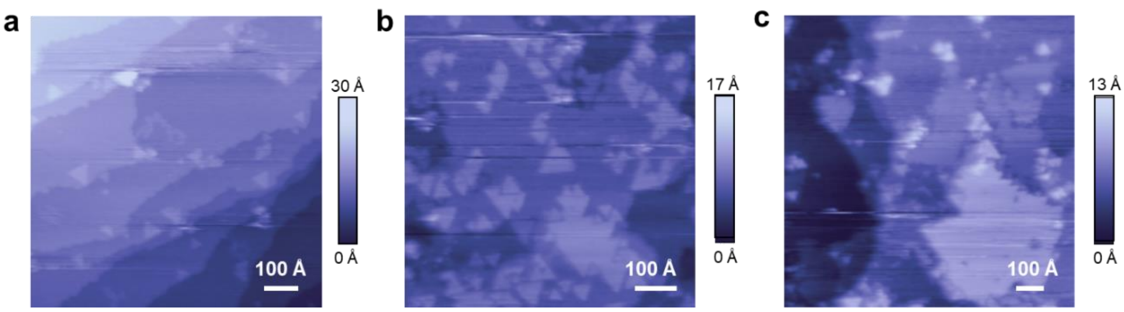

Figure S5. **a-c**, At the condition of Ta deposited first on the substrate, STM topographic images of the surface after growth under different Ta deposition time for 6 min, 10 min and 14 min, respectively.

References

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