

Substrate Facet Effect on the Growth of Monolayer MoS₂ on Au Foils

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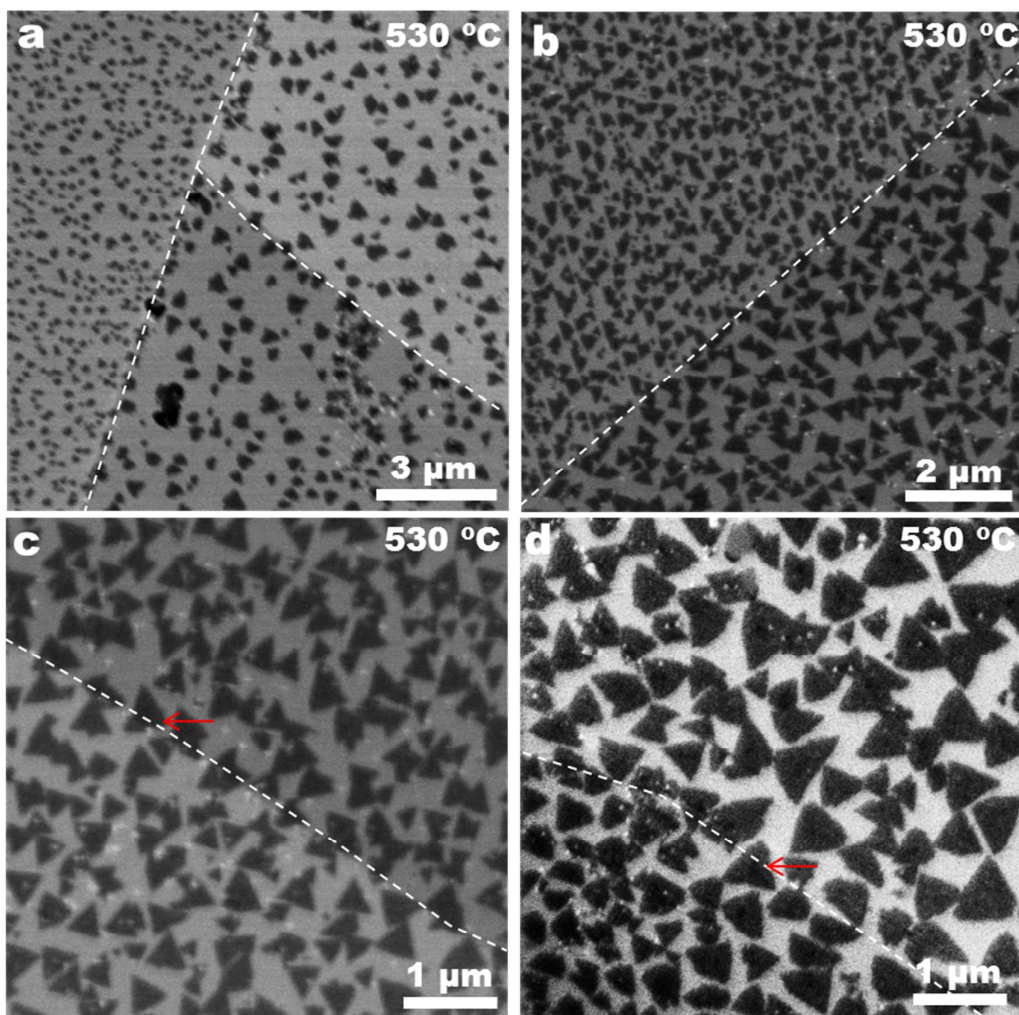


Figure S1. SEM images of monolayer MoS₂ triangles on Au foils (synthesized at 530 °C for 1 h under 50 sccm Ar carrier gas). (a,b) Low-magnification SEM images of triangular MoS₂ flakes synthesized on different Au facets, with the dotted lines indicating the Au grain boundaries. (c,d) High-magnification SEM images of MoS₂ triangles on two Au facets, with the red arrow indicating a MoS₂ flake riding over the Au grain boundary.

Scanning electron microscopy (SEM) images (**Figure S1a,b**) present a relative uniform distribution of monolayer MoS₂ triangles on different facets of Au foils, although the flake sizes vary a little on different Au facets. Intriguingly, the triangular MoS₂ flakes can ride over the Au grain boundary with their shapes and orientations nearly unchanged, as indicated by the red arrow in **Figure S1c,d**. This should facilitate the growth of large domain MoS₂ flakes on polycrystalline Au foils.

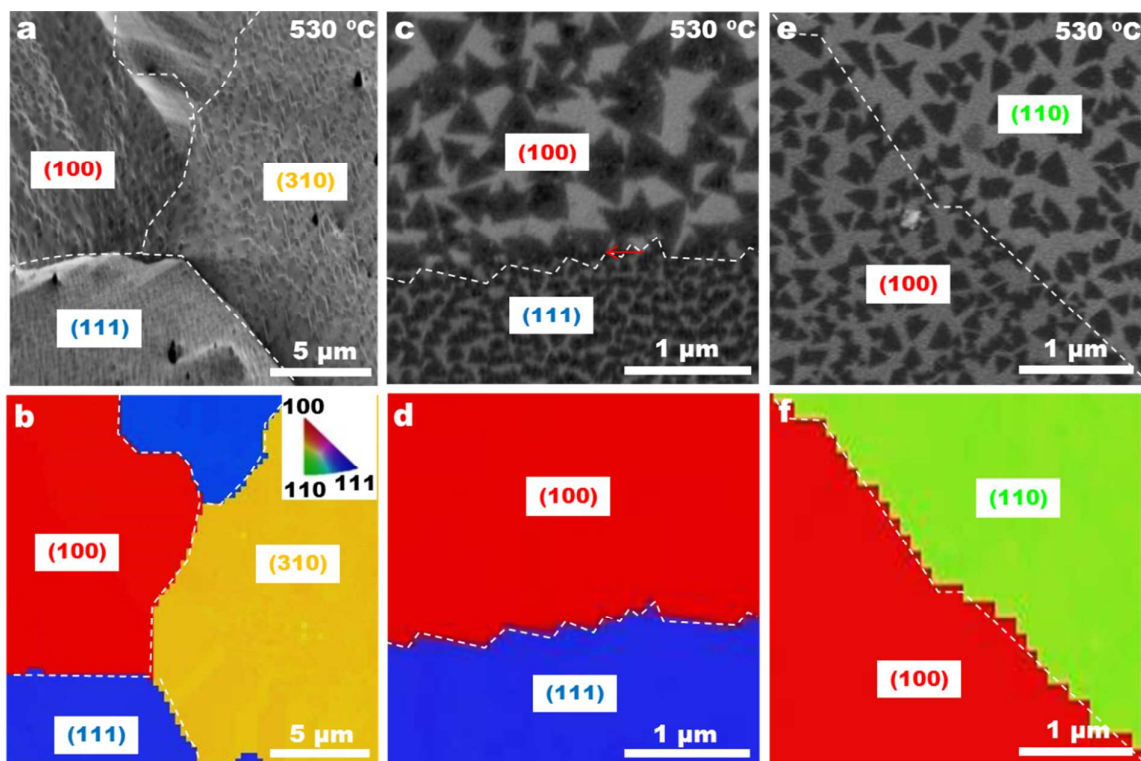


Figure S2. SEM and corresponding EBSD images of monolayer MoS₂ flakes on different Au facets grown at low growth temperature of 530 °C. (a) Low-magnification SEM image of MoS₂ flakes on three facets of Au foils, with the dotted lines indicating the Au grain boundaries. (b) Corresponding EBSD image of the same region in (a) using the standard EBSD color key, showing three Au facets (Au(100), Au(111) and Au(310)). (c,e) High-magnification SEM images of MoS₂ triangles on two neighboring Au facets of Au(100) and Au(111), and Au(110) and Au(111), respectively. (d,f) Corresponding EBSD images of the same regions of (c) and (e), respectively.

In order to identify the crystal structure of the underlying Au foil visually, SEM images (Figure S2a,c,e) and corresponding electron backscatter diffraction (EBSD) maps (Figure S2b,d,f) are obtained to correlate the domain size of MoS₂ with the Au crystallographic orientation. Notably, larger domain MoS₂ triangles preferably evolve on the Au(100) and Au(110) facts than that on Au(111) at 530 °C growth.

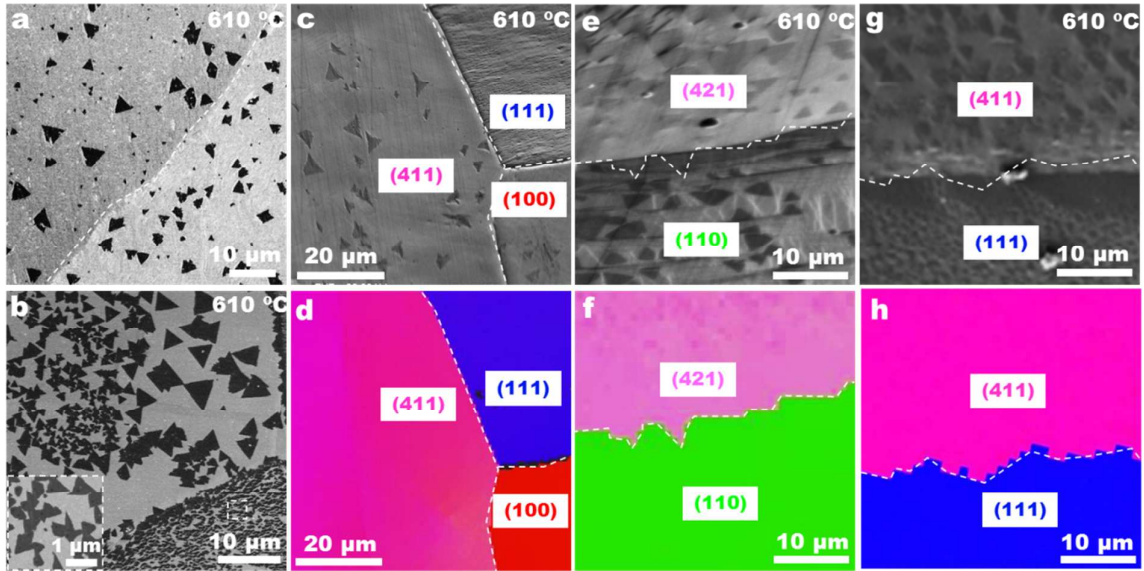


Figure S3. Edge-length and shape dependence of MoS₂ flakes on different Au facets grown at a medium temperature of ~ 610 °C (CVD growth for 1 h under 50 sccm Ar carrier gas). (a,b) Low-magnification SEM image of MoS₂ flakes on different Au facets. **(c,e,g)** SEM images of MoS₂ flakes on different Au facets. **(d,f,h)** Corresponding EBSD images of the same regions of **(c)**, **(e)** and **(g)**, respectively.

Upon increasing the growth temperature to 610 °C, the edge length and shape of MoS₂ flakes present a clear difference on disparate Au facets. Large domain, triangular MoS₂ flakes with sharp edges were usually obtained on the low-index facet of (100) and (111). In contrast, only small and irregular MoS₂ flakes were achieved on high-index facets, such as (411) and (421).

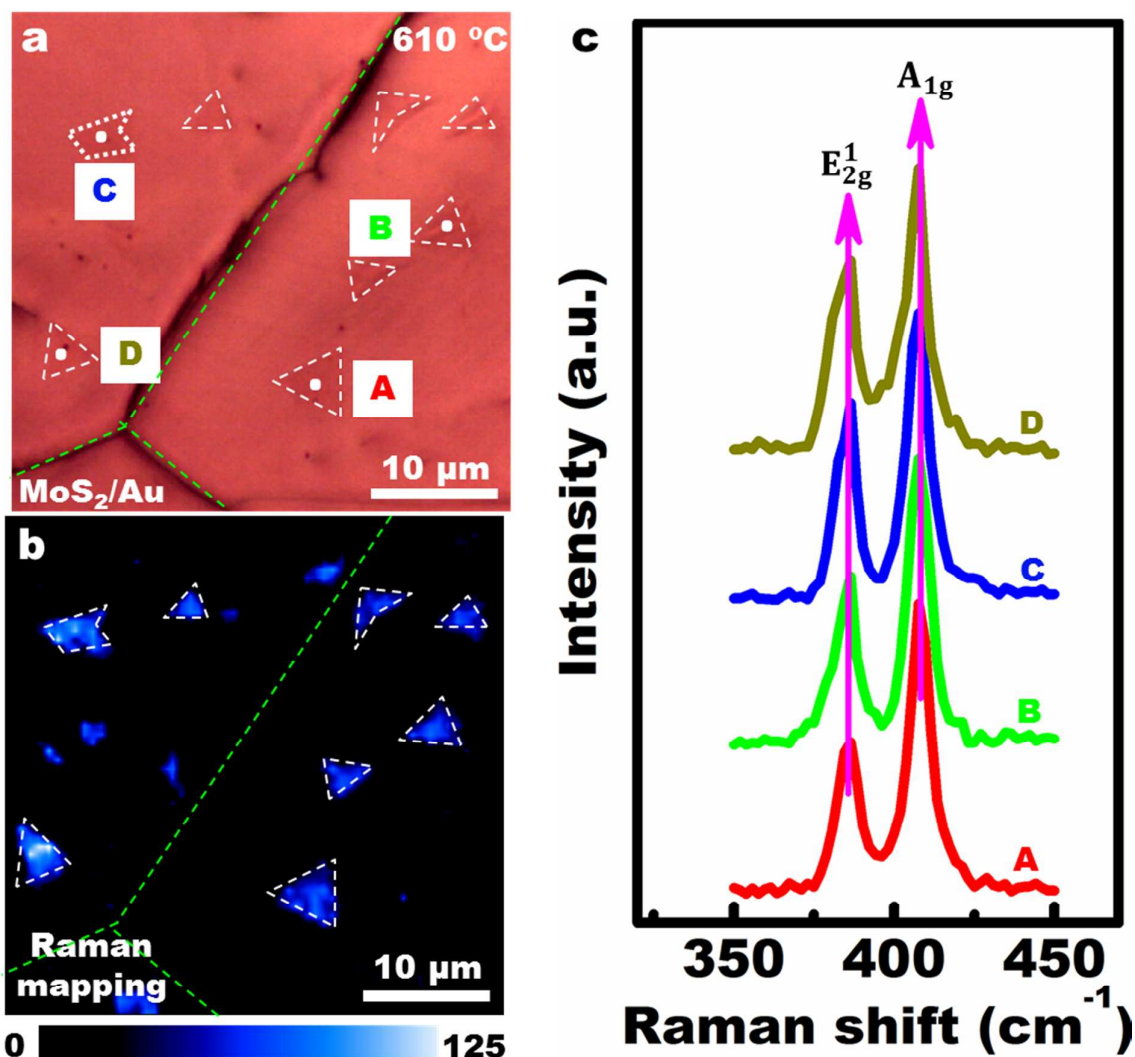


Figure S4. Raman characterization of MoS₂ flakes on polycrystalline Au foils at a medium growth temperature of 610 °C. (a,b) Optical microscope and Raman mapping images of MoS₂ flakes on different Au facets. (c) Raman spectra captured from MoS₂ flakes as indicated in (a).

Optical microscope (OM) and Raman mapping were employed to reveal the facet-dependent growth feature of MoS₂ on Au foils (**Figure S4a,b**). The relative homogeneous Raman mapping signal confirms the rather high thickness uniformity of as-grown MoS₂ on Au foils. Besides, single point Raman spectra captured from MoS₂ flakes (**Figure S4a**) reconfirms the high thickness uniformity, in view of the fixed location of the typical vibrations modes (A_{1g} and E_{2g}¹) as shown in **Figure S4c**.

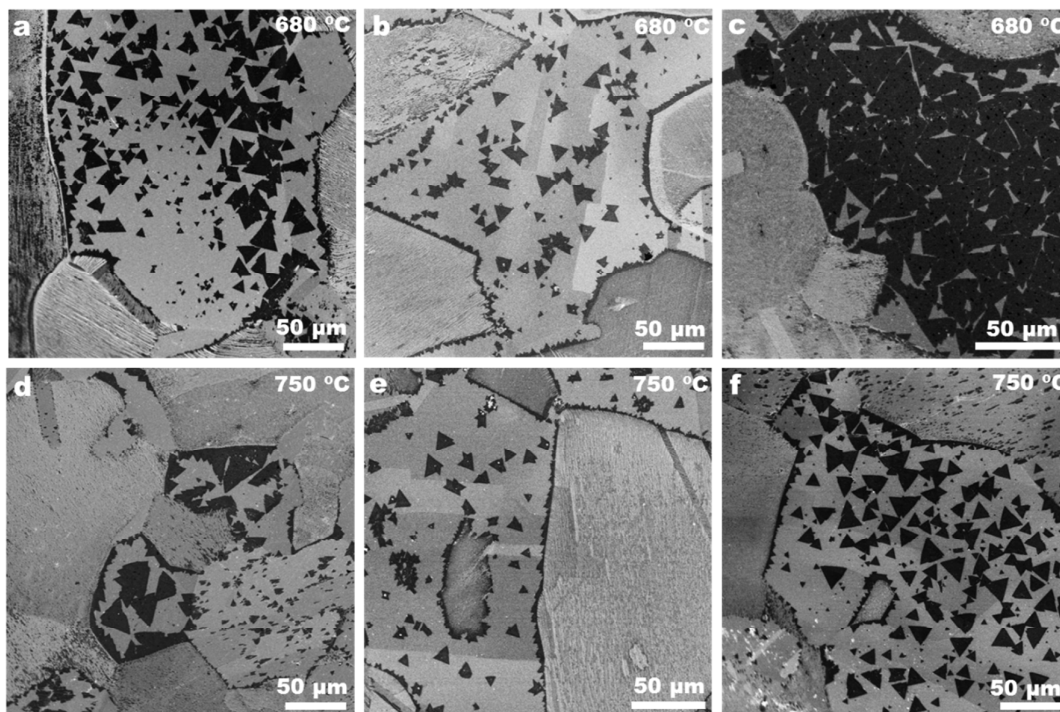


Figure S5. SEM images of monolayer MoS₂ triangles on Au foils (synthesized at 680 °C and 750 °C for 1 h under 50 sccm Ar carrier gas, respectively). (a-c) SEM images of monolayer MoS₂ triangles synthesized at 680 °C. (d-f) SEM images of MoS₂ flakes synthesized at 750 °C.

At high growth temperature of 680 °C and 750 °C, uniform triangular and polygonal MoS₂ flakes were obtained on specific Au facets, as evidenced by SEM images. And no MoS₂ flakes were observed on the neighboring facets. Meanwhile, the edge length of the triangular MoS₂ flake is greatly enhanced (~ 50 μm at large) along with a dramatic decrease of the nucleation density.

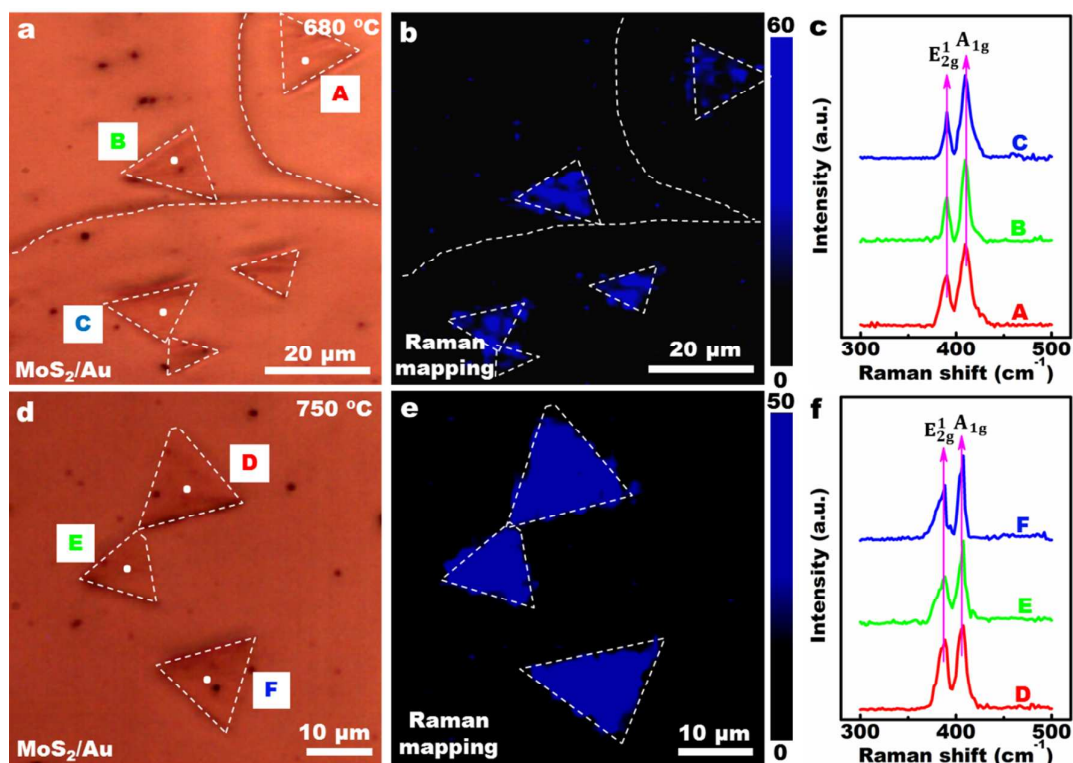


Figure S6. OM and Raman characterizations of monolayer MoS₂ flakes on Au foils (synthesized at 680 °C and 750 °C for 1 h under 50 sccm Ar carrier gas, respectively). (a,b) OM and Raman mapping images of monolayer MoS₂ flakes synthesized at 680 °C. (c) Single point Raman spectra captured from MoS₂ flakes as indicated in (a). (d,e) OM and Raman mapping images of monolayer MoS₂ flakes on Au foils synthesized at 750 °C. (f) Single point Raman spectra captured from MoS₂ flakes as indicated in (d).

OM and Raman mapping were employed to confirm the thickness uniformity and the crystal quality of MoS₂ flakes on Au foils grown at relative high temperature of 680 °C and 750 °C (**Figure S6a,b** and **Figure S6d,e**), respectively. The typical vibrations modes (A₁' and E₂') for MoS₂ flakes synthesized on different Au grains exhibit almost the same locations, as well as the same frequency difference of $\Delta \sim 19.9 \text{ cm}^{-1}$. These data suggest substantially the monolayer feature of MoS₂ triangles on Au foils (**Figure S6c,f**).

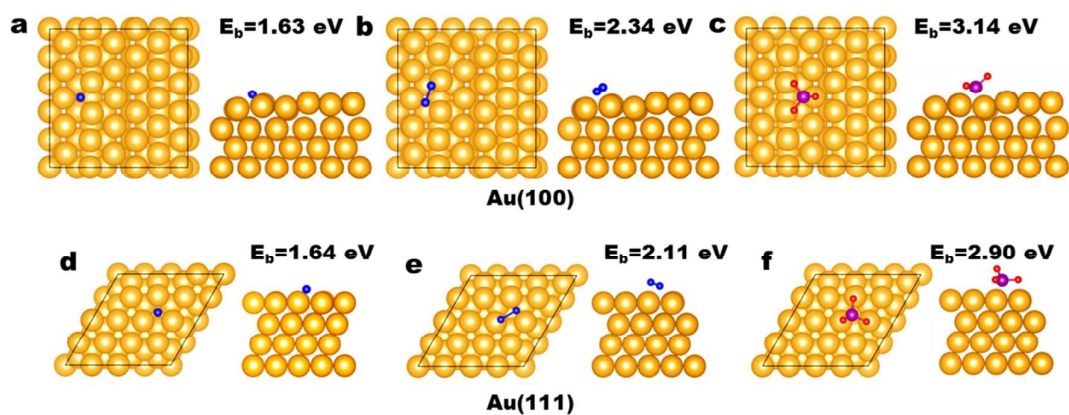


Figure S7. Top and side views of the most stable adsorption geometries of different adsorbates on (5×1) reconstructed Au(100) and Au(111) facets. (a,d) a single S atom; (b,e) a S₂ molecule; (c,f) a MoO₃ cluster, with the marked values showing the binding energies of S, S₂ and MoO₃ on Au(100) and Au(111) facets.