Epitaxial growth of 2D Se-Ta-S Janus membranes on Au(111)

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2D transition metal chalcogenides (TMC) Janus membranes are asymmetric materials characterized by a transition metal layer (M) sandwiched between two different chalcogen layers (X and X'). Due to the intrinsic electric field generated by the differing partial charges of the chalcogens, they exhibit unique properties. For instance, breaking out-of-plane symmetry can lead to strong piezoelectric effects and Rashba spin splitting [1, 2].

Here, we report the controlled synthesis and atomic-scale characterization of Se-Ta-S Janus membrane grown on Au(111) under ultra-high vacuum. Tantalum is first deposited onto clean Au(111) and annealed in an H_2S atmosphere, resulting in tantalum monosulfide (TaS) under sulfur-poor conditions (Fig. 1a). From our previous works, we know that this phase can be further converted to TaS_2 by additional sulfurization (Fig. 1b) [3, 4]. Exploiting this layer-wise growth mechanism, we instead introduce selenium in a second step, enabling the formation of Se-Ta-S structures (Fig. 1c). Scanning tunneling microscopy reveals large-area, well-defined islands with atomic precision, providing strong evidence for the formation of Se-Ta-S Janus membranes. A similar growth behavior is observed in the TaSe system (Fig. 1d), where TaSe can be transformed into $TaSe_2$ (Fig. 1e), suggesting that our protocol can also enable Janus membranes with inverted polarity. Compared with existing approaches that rely on H_2 plasma stripping to introduce the second chalcogen, our method avoids this defect-inducing step, yielding more uniform, well-oriented samples. These results establish a reliable route for fabricating Janus TMCs with controlled composition and polarity.

References:
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[2] L. Zhang et al., Nanoscale 11 (2019) 5666.
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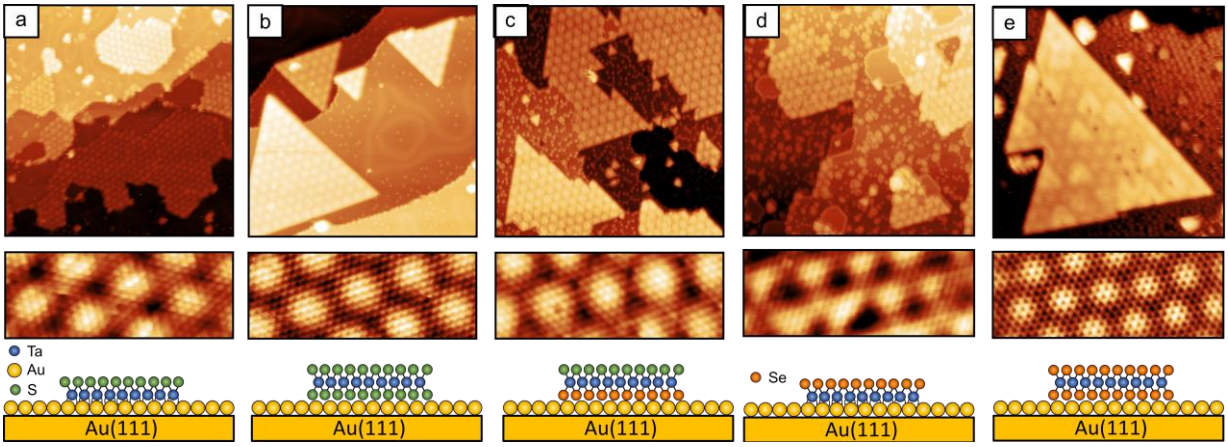

Fig. 1: STM images of (top) large-scale islands, (middle) their atomic and moiré resolution at a size 14.5 nm x 6.5 nm (bottom) corresponding model for (a) TaS, (b) TaS_2, (c) SeTaS, (d) TaSe and (e) $TaSe_2$. Image parameters: (a, top) V = -2.25 V, I = 0.01 nA, (a, middle) V = -0.91 V, I = 0.10 nA, (b, top) V = -0.83 V, I = 0.09 nA, (b, middle) V = -0.44 V, I = 3.80 nA,(c, top) V = -1.42 V, I = 0.17 nA, (c, middle) V = -0.30 V, I = 1.7 nA. (d, top) V = -1.66 V, I = 0.02 nA, (d, middle) V = -0.14 V, I = 0.14 nA, and (e, top) V = -0.35 V, I = 0.04 nA, (e, middle) V = -0.35 V, I = 0.58 nA, Image Sizes: (a, top) 21.5 nm × 21.5 nm, (b, top) 46.5 nm × 46.5 nm, (c, top) 49.0 nm × 49.0 nm, (d, top) 43.5 nm × 43.5 nm, and (e, top) 22.5 nm × 22.5 nm.