Supporting Information

Uniform vapor pressure based CVD growth of MoS_2 using MoO_3 thin film as a precursor for co-evaporation

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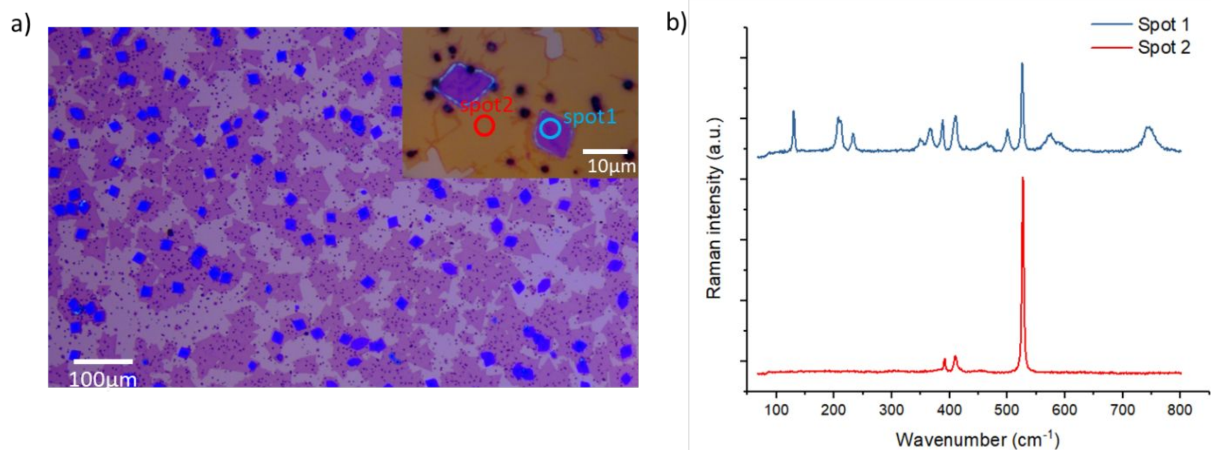

Figure S1. Growth results for co-evaporation of MoO_3 and sulfur powders. a) Optical micrographs for the crystals grown with ~1mg of MoO_3 powder and 600mg of S with the same setup. b) The Raman single spectra taken at spot 1 (blue) and spot 2 (red) shown in the inset of a). The Raman spectra is a combination at spot 1 is MoS_2/$MoOS_2$[1] and the reduced intensity of Si peak is an indication that these plates are very thick. Also, the high intensity of MoS_2 peaks implies that a thick layer of MoS_2 is grown on top of the oxysulfide plates. The spot 2 shows a Δ=18.92 cm^{-1} between E^1_{2g} and A_{1g} peaks which corresponds to monolayer MoS_2.

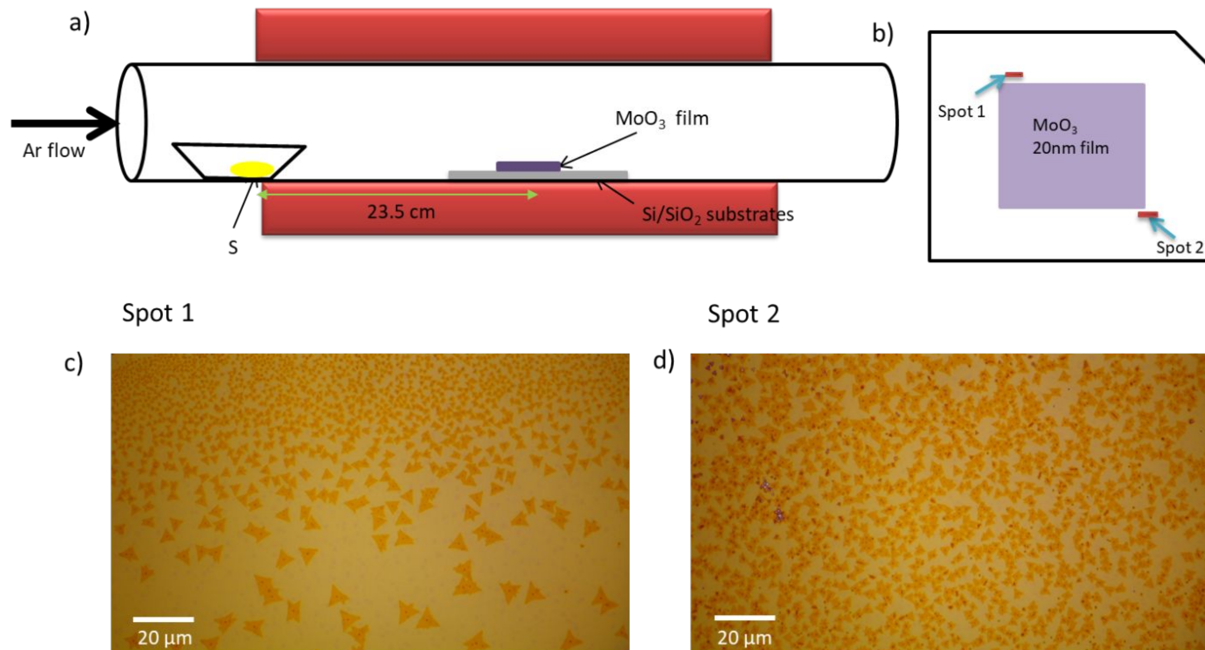

Figure S2. Lateral placement of source and substrate. a) Experimental setup showing relative positioning of substrate and precursors. b) Locations of MoS_2 growth on the substrate. c), d) Optical micrographs at spots 1 and 2 respectively. Using the same MoO_3 film thickness (20 nm) and same amount of S (600 mg), we observed that MoS_2 growth is limited to few spots near the source edges in lateral placement geometry.

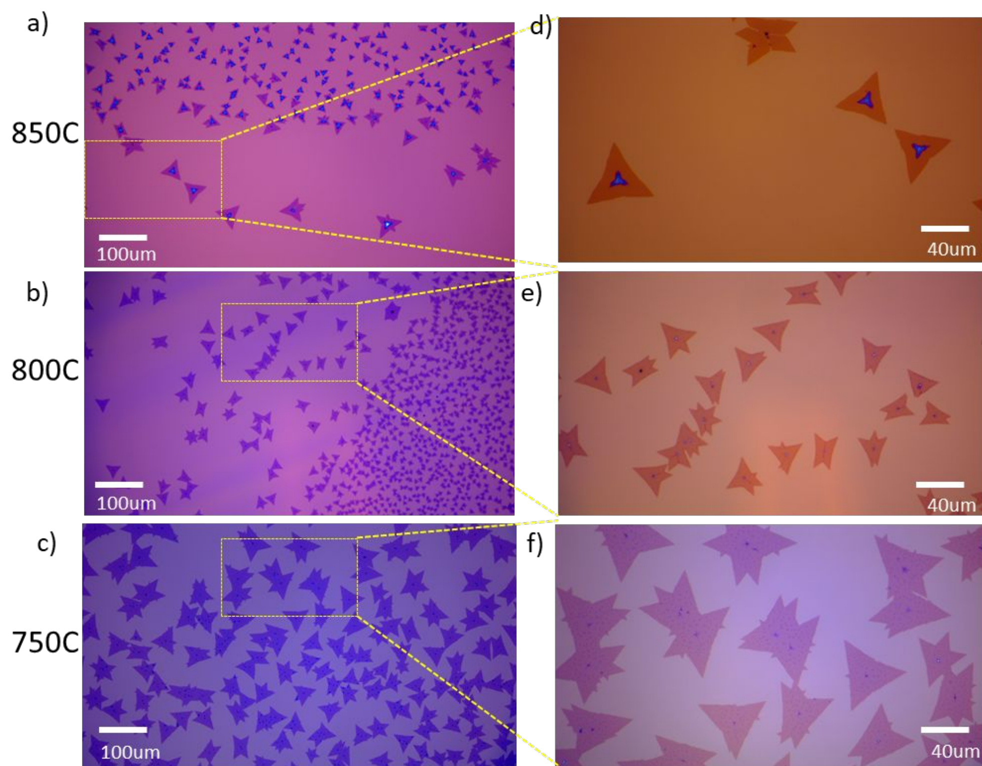

Figure S3. Growth at different temperatures. a)-c) Optical images of growth at 850C, 800C and 750C respectively taken at 20X magnification. d)- f) corresponding images at 50x magnification.

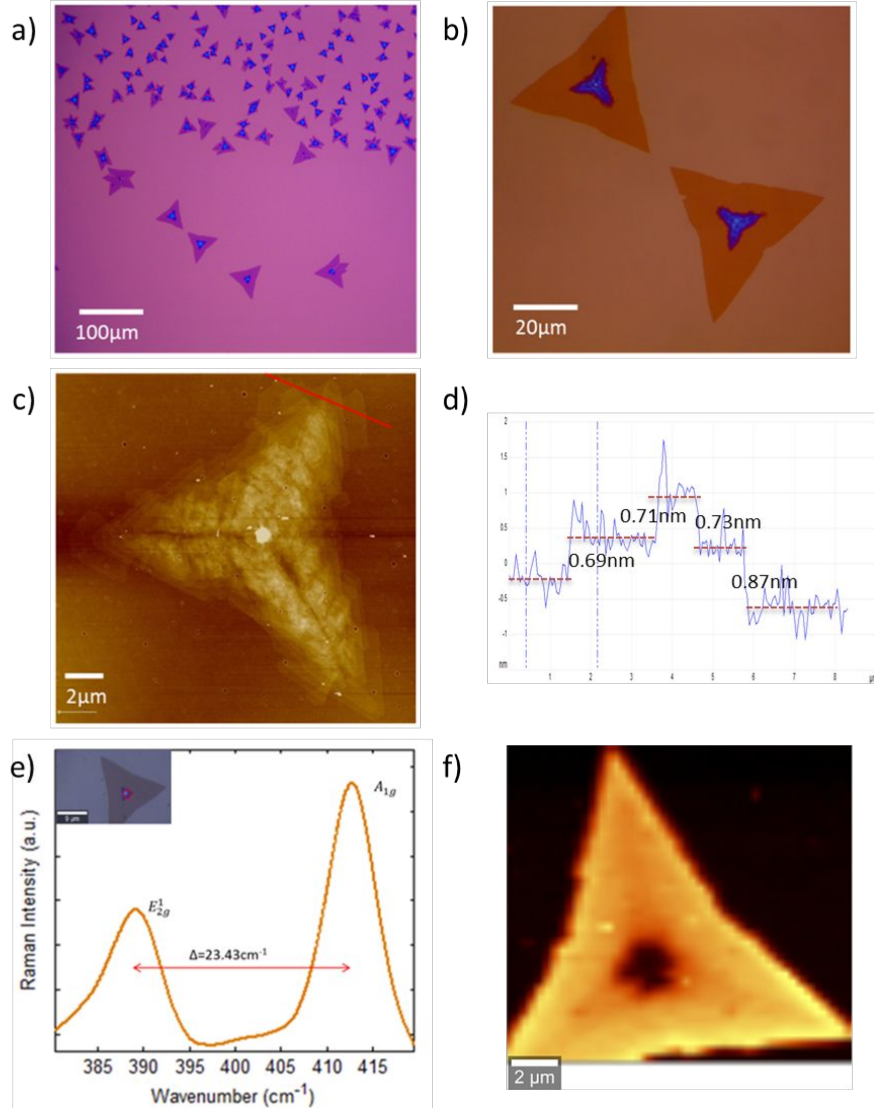

Figure S4. Multilayer growth around the nucleation center at 850°C. a), b) Optical images taken at 20x and 100x magnifications respectively. c) AFM topography and d) height profile showing step-like height variation which corresponds to individual MoS_2 layers. e) Raman spectra for the multilayers at the nucleation center. f) PL mapping centered at 1.8eV shows the PL intensity drop at the center due to the thick layers and uniform outer region consists of monolayer MoS_2

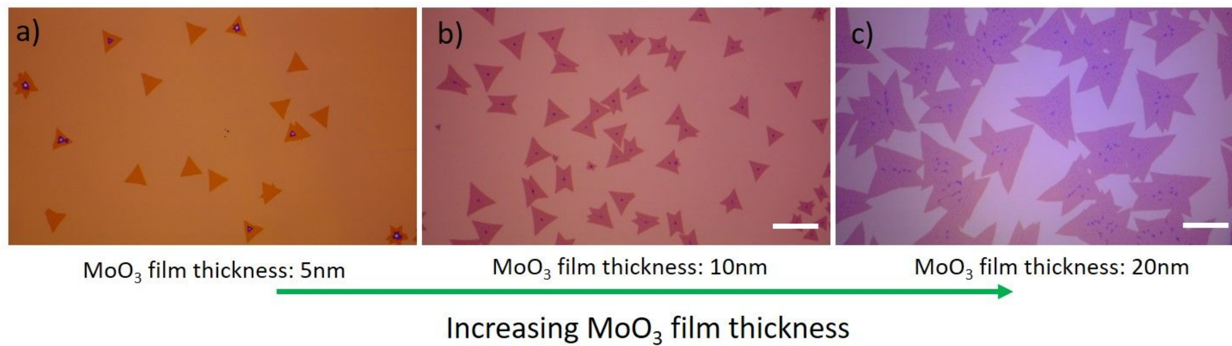

Figure S5. Effect of MoO3 film thickness. Optical micrographs of crystals grown with a)5nm, b) 10nm, c) 20nm MoO3 precursor thickness with 600mg of sulfur. Scale bar is 40μm in each image.

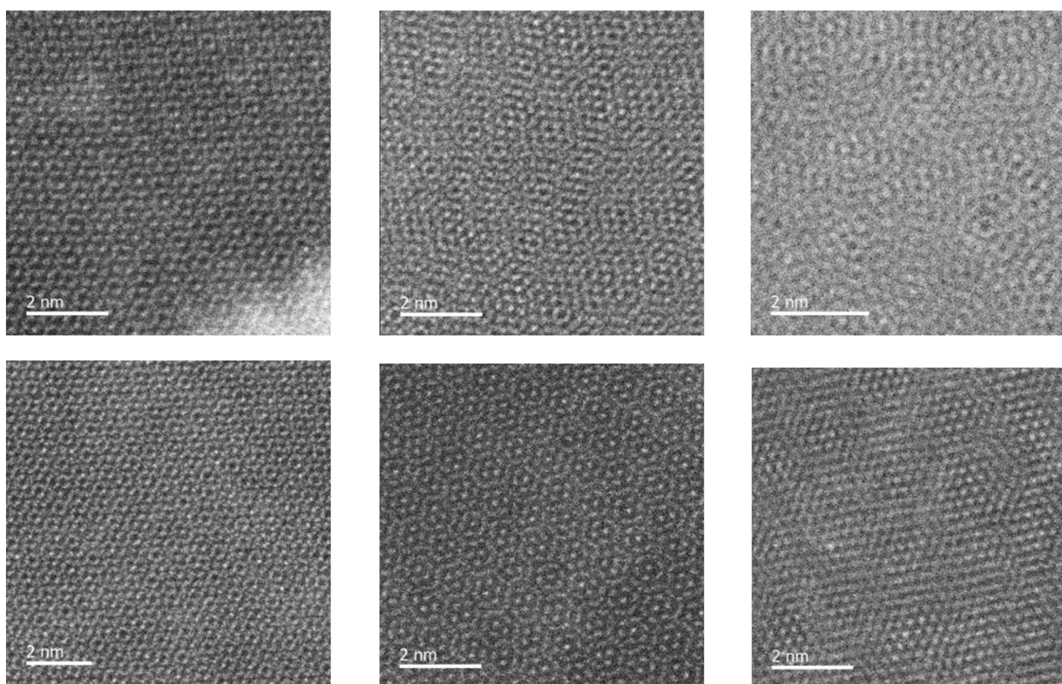

Figure S6. Supplementary TEM images. Different Moire patterns were seen at some locations of the transferred flakes depending on the angle of orientation of different layers at the polycrystalline regions and/or different folding orientations of the transferred films.

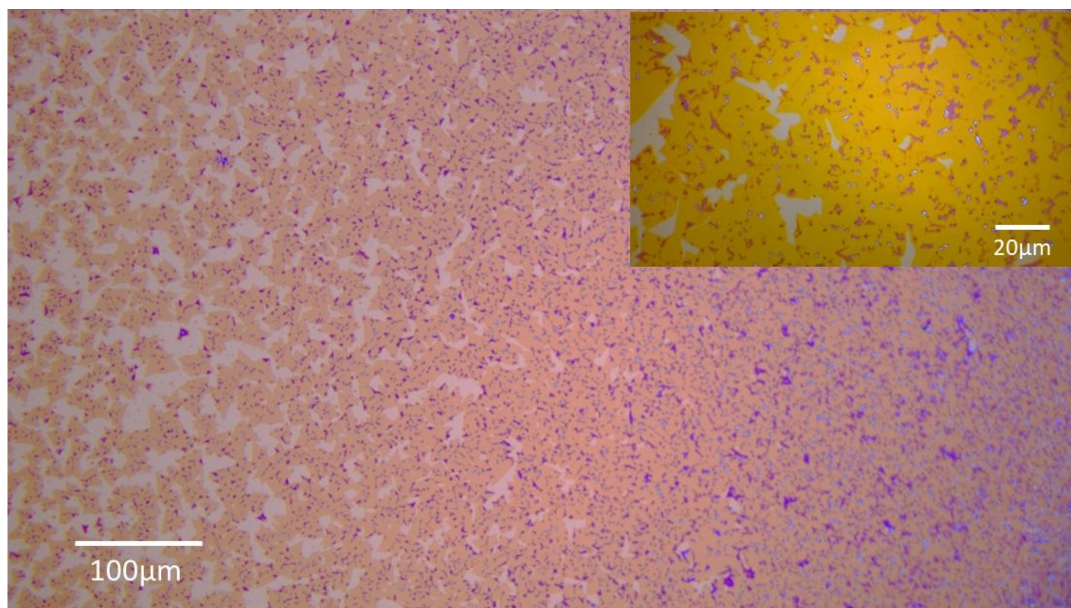

Figure S7. MoS_2 growth on source substrate. An optical micrograph of the 20nm MoO3 source substrate after the growth. An enlarged image is shown (100X) in the inset. We observed this growth at few regions of the source substrate and most of these regions are polycrystalline. Grain boundaries and overlapping is also visible in the optical images.

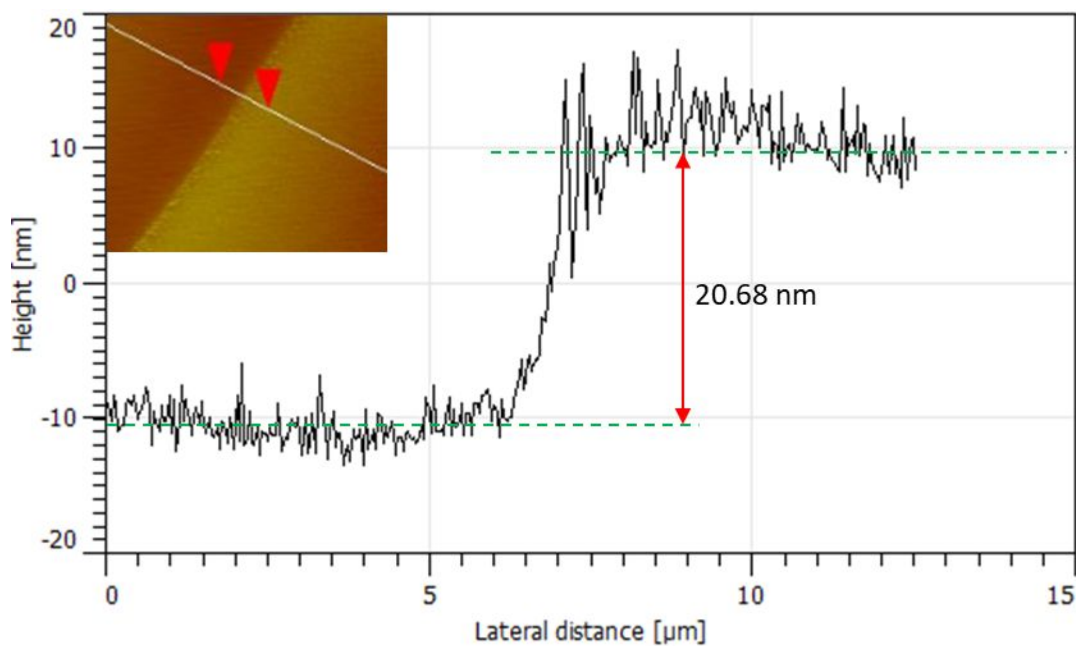

Figure S8. AFM characterization of thermally deposited thin film with target thickness of 20nm. AFM topography image shown in the inset express high uniformity of the deposited thin film. Height profile shows a clear step height of 20.68 nm at the substrate-thin film boundary

References

(1) Pondick, J. V.; Woods, J. M.; Xing, J.; Zhou, Y.; Cha, J. J. Stepwise Sulfurization from MoO3 to MoS2 via Chemical Vapor Deposition. *ACS Applied Nano Materials* **2018,** *1* (10), 5655-5661.