Electronic Supplementary Material (ESI) for Nanoscale Advances.

Electronic Supplementary Information

Barrier-assisted vapor phase CVD of large-area homogeneous MoS_2 monolayers with high spatial homogenity

Santhosh Durairaj,[a] P. Krishnamoorthy,[a] Navanya Raveendran,[a] Beo Deul Ryu,[b] Chang-Hee Hong,[b] Tae Hoon Seo,[c] S. Chandramohan[a]*

[a]2D Materials and Devices Laboratory, Department of Physics and Nanotechnology, SRM Institute of Science and Technology, Kattankulathur 603 203, Tamil Nadu, India

[b]Department of Semiconductor Science and Technology, Semiconductor Physics Research Center, Chonbuk National University, Jeonju 54896, South Korea

[c]Nano-photonics Convergence Technology, Korea Institute of Industrial Technology, Gwangju61012, South Korea

E-mail: chandras3@srmist.edu.in

MoS_2 grown at different temperatures:

The effect of growth temperature is also studied by performing the growth experiments at different temperatures from 700 to 800 °C. Irrespective of the growth time, only triangular flakes were formed at a growth temperature of 700 °C. When the temperature is increased to 750 °C, the MoS_2 coverage on the substrate significantly increased and at an elevated temperature of 800°C despite continuous MoS_2 growth, nucleation of additional layers as well as secondary phases have been observed on the first layer.

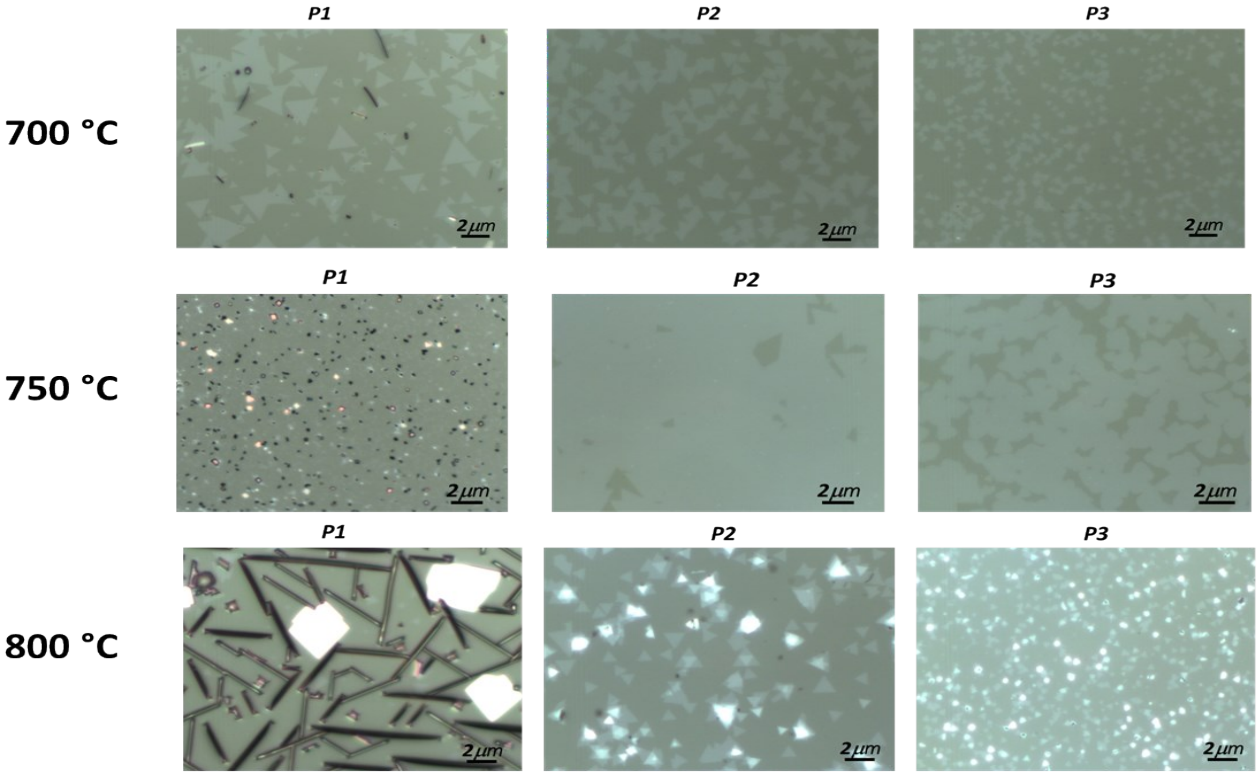

Fig. S1. Optical microscope images of MoS_2 grown at different temperatures (700, 750 and 800 °C) at different substrate positions P_1, P_2 and P_3.The second and few layer nucleation sites (brighter regions) are clearly visible for the sample grown at 800°C.

Characterization of MoS_2grown at 800 °C:

At 800 °C, the growth was very rapid and hence no control over the nucleation of additional layers is facilitated. The optical microscope image clearly shows several triangular flakes on the first layer. Furthermore, the bright spots are identified as few- and/or multi-layered flakes, according to Raman spectra (Fig. 3) which is further corroborated by TEM analysis. According to the TEM image shown in figure b, the bulk-like MoS_2 is identified to have many triangular flakes stacked one over another.

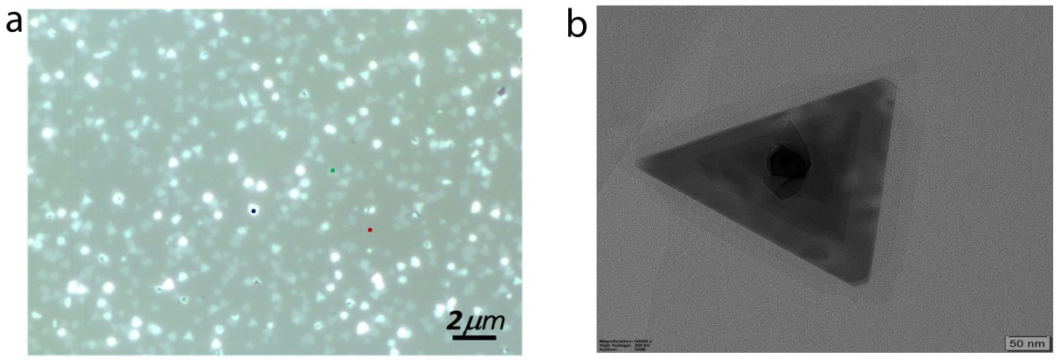

Fig. S2. (a) Optical microscope image of MoS_2 grown at800 °C in P_3 position. **(b)** TEM image of bulk layer nucleation site.

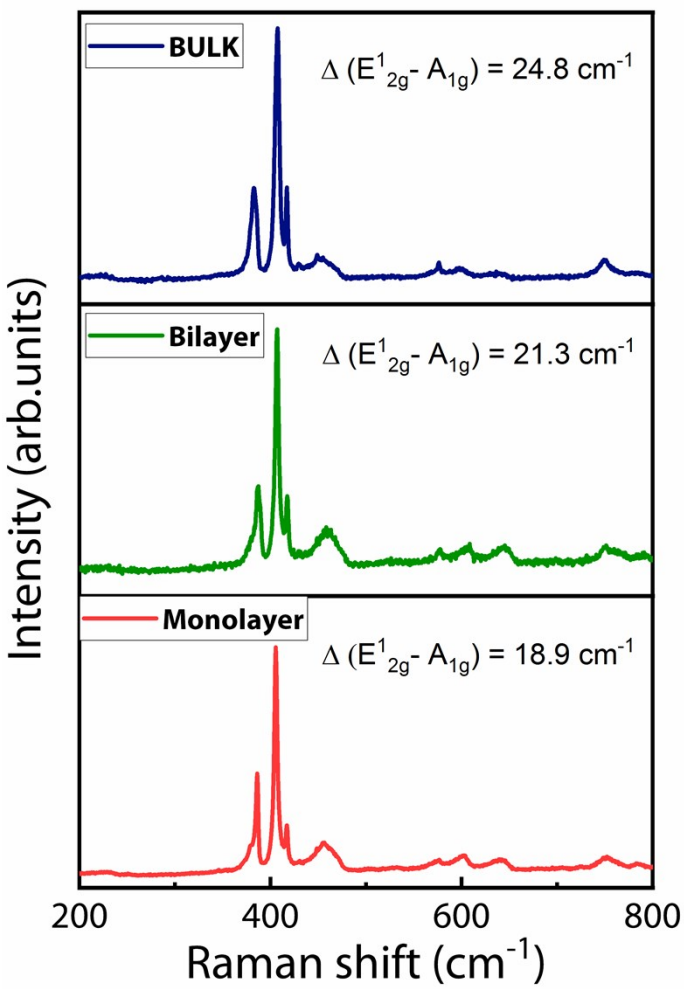

Fig. S3. Raman spectra collected at different points of optical contrast seen in the optical microscope image in figure 2a.By acquiring the spectrum at different points, we ensured that the different optical contrast seen in the optical image correspond to different layer thickness of MoS_2.

Effect of growth time: Case study for 750 °condition

Based on the results, growth temperature of 750 **°C** can be considered optimum to grow continuous MoS_2 without second layer nucleation. Since a growth time of 10 minutes did not result continuous MoS_2, we varied the growth time and studied the growth behavior at this particular temperature. The optical images of the sample grown at 10, 20, and 30 minutes are shown in figure 4. It is obvious that the coverage of the MoS_2 increased with increasing growth time. Furthermore, there is no obvious formation of secondary phases or additional layers in the form of triangular flakes observed. However, complete coalescence did not happen even at a growth time of 30 minutes. Hence, a growth temperature slightly above 750 **°C** is attempted in this study to get fully coalesced MoS_2monolayers.

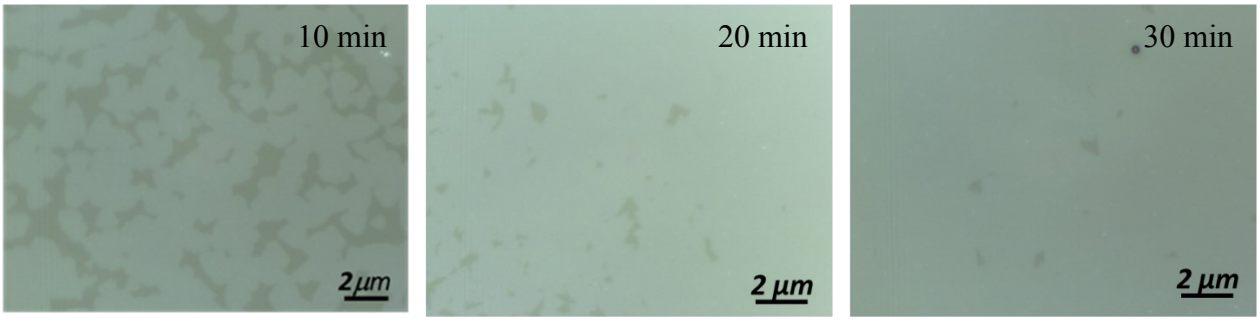

Fig. S4. Optical microscope images depicting morphology evolution with increasing growth time from 10 minutes to 30 minutes for MoS_2 grown at 750 °C.

Large-scale uniformity of monolayer MoS_2 grown at 775°C:

To check the quality and thickness uniformity of MoS_2 over the entire substrate, we acquired the Raman and PL spectrum at different sampling points as illustrated in figure 5a. The frequency difference ($\Delta\omega$) between the E^1_{2g} and the A_{1g} modes is considered as a figure of merit to evaluate the thickness uniformity of the grown MoS_2. The $\Delta\omega$ values were found to be within the range (from 18.9 to 19.45 cm^{-1}) reported for monolayer MoS_2. Furthermore, the PL spectrum taken at several points showed similar results with a strong emission peak around 1.87-1.89 eV.

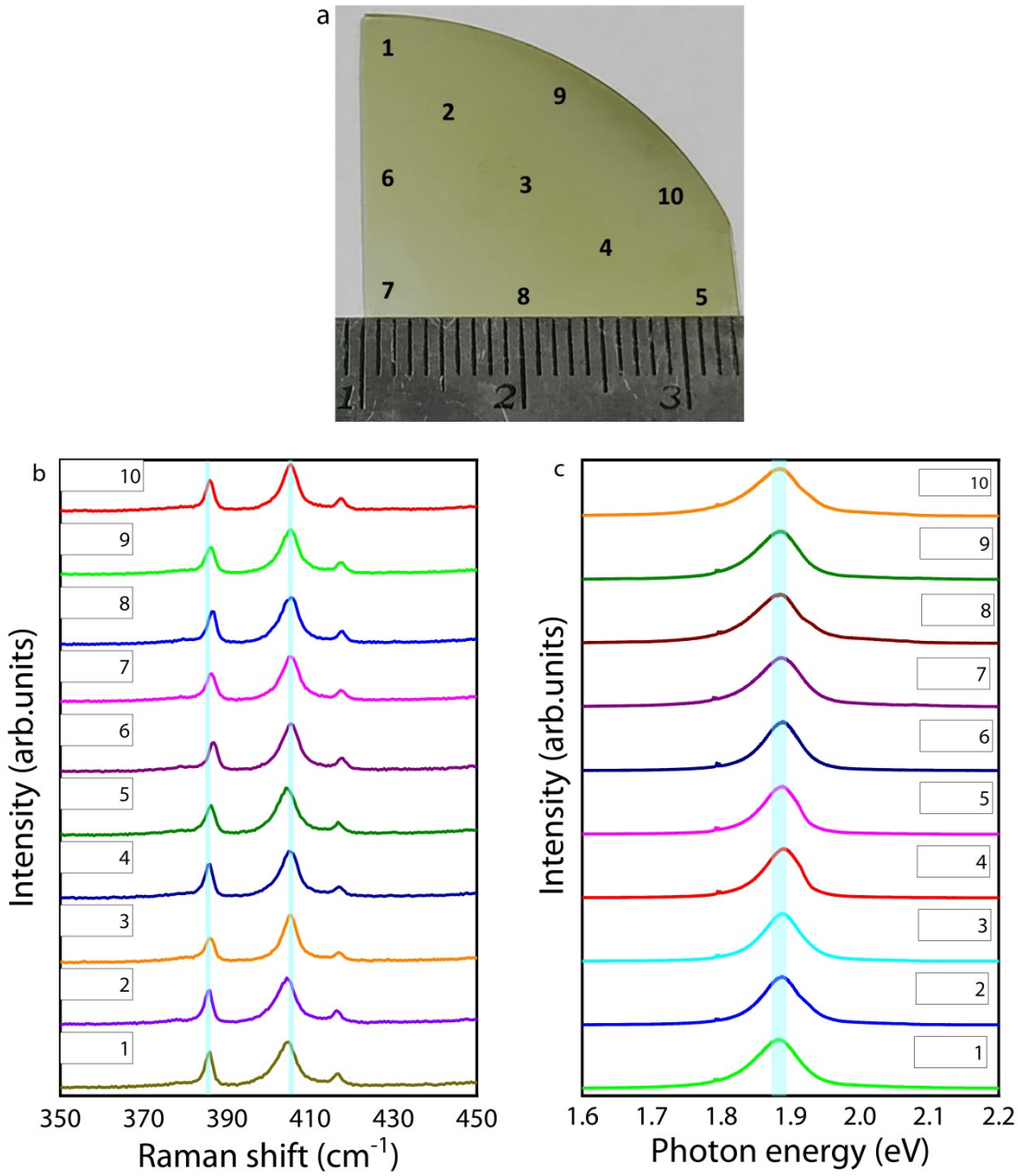

Fig. S5. (a) Photograph of a typical sample grown at position P_3. The numbers on the image are representative points at which micro-Raman and PL measurements were performed. **(b, c)** Raman and PL spectra of MoS_2 film confirming the thickness and optical homogeneity.

SAED pattern for MoS_2 film at different points:

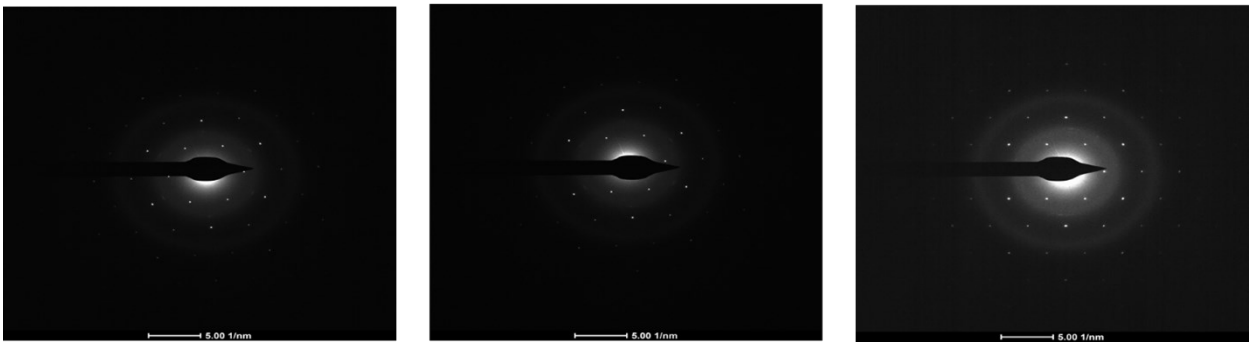

Fig. S6. SAED pattern for MoS_2 film at different points confirming the single crystal structure with (2H) hexagonal phase symmetry.

Thickness of the MoS_2 film on sapphire:

In order to measure the thickness of the as-grown MoS_2 on sapphire, a scratch was made on the sample and the film was investigated using AFM. As shown in the AFM line profile image, the approximate thickness of the MoS_2 is estimated to be 0.8 nm, which is close to monolayer film thickness reported in the literature.

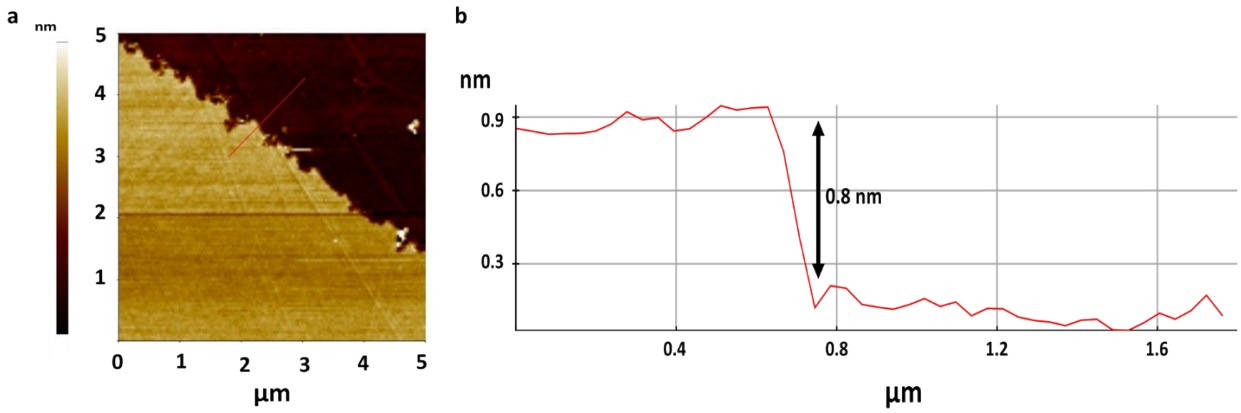

Fig. S7. (a) AFM surface topography of the MoS_2layer on sapphire substrate. A scratch is intentionally made to measure the film thickness. **(b)**AFM line-profile made across the film-substrate interface (the red line in figure a), representing to estimation of film thickness.

Size distribution of MoS_2 flakes:

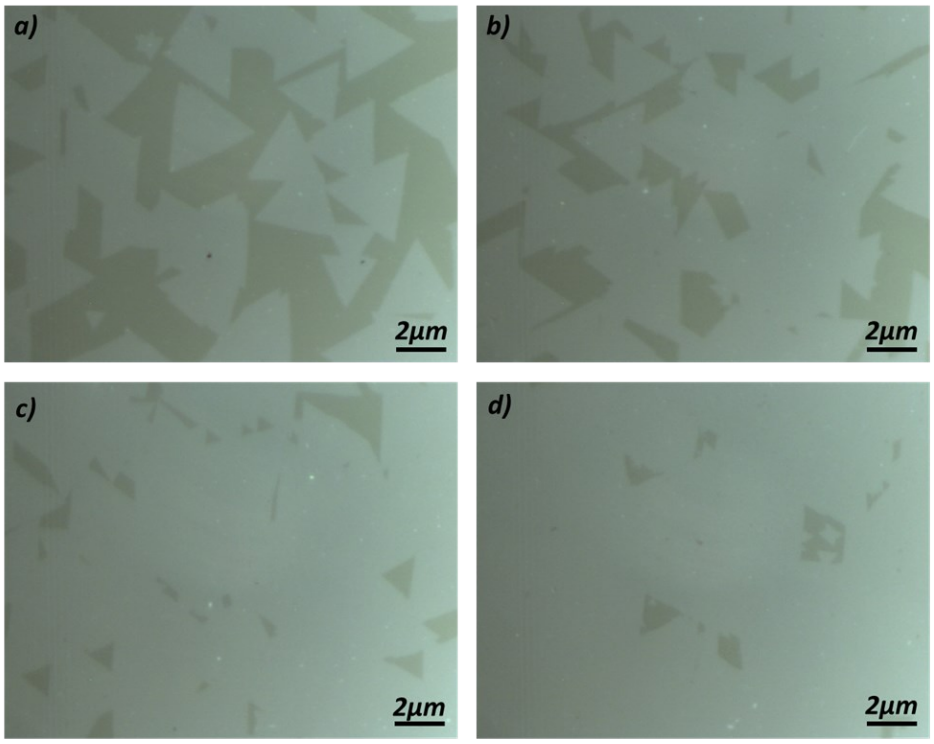

Fig. S8. Optical images of MoS_2 showing the size distribution of individual flakes. The average size of the flakes range from 3-10 μm.

MoS_2 field-effect transistor:

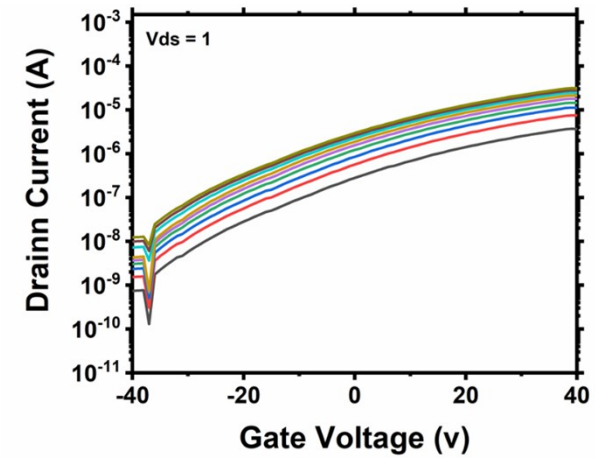

Fig. S9. Transfer characteristics measured for 10 devices and the estimated mobility values.