Uniform and Repeatable Cold-Wall CVD Synthesis of Single-Layer MoS$_2$
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Supplementary Information
MoS$_2$ was grown in a cold-wall CVD reactor using MoO$_3$ and S deposited from alumina coated W baskets in a flowing Ar ambient. This reactor allows independent control over all deposition parameters. Several techniques were used to analyze the uniformity and composition of our samples. Auger electron spectroscopy (AES) was used to map elemental composition across the samples. The method for acquiring spectra is presented in the Experimental Methods section of the main text. Fig. S1(a) displays two differentiated, dN/dE, Auger spectra acquired over the energy range 100 eV < E < 300 eV to focus on the Auger transitions relevant to growth of MoS$_2$. N is the number of electrons detected with kinetic energy E. Several prominent peaks are evident in these spectra. For analysis of our MoS$_2$ films, the S$_{\text{LMM}}$ and Mo$_{\text{MNN}}$ Auger transitions at 151 eV and 186 eV, respectively, are particularly useful. These transitions are identified by green arrows in fig. S1(a). Also evident are the Si$_{\text{LVV}}$ (91 eV), Mo$_{\text{MNN}}$(223 eV) and C$_{\text{KLL}}$(274 eV) Auger transitions. Due to our focus on Mo and S Auger transitions, the O$_{\text{KLL}}$ transition at 511 eV is not shown.

In fig. S1(a), the red curve plots the dN/dE Auger spectrum acquired from a bulk MoS$_2$ standard formed by compressing 99% MoS$_2$ powder to 100 MPa. In this manner, two separate bulk standards were fabricated and a total of 5 Auger spectra were acquired from these standards using identical acquisition parameters. While the absolute intensities of
the S and Mo transitions varied, the ratio, \( R = \frac{I_{S_{LMM}}}{I_{Mo_{MNN}}} \) (\( I \) is the peak-to-peak intensity of a particular Auger transition) was nearly constant, yielding \( R = 11.1 \pm 0.14 \).

We use this ratio as a convenient fingerprint for stoichiometric MoS\(_2\) and to derive the equivalent coverage of stoichiometric MoS\(_2\) nanocrystals in our films from the intensity of the \( S_{LMM} \) Auger transition. The equivalent coverage of stoichiometric MoS\(_2\) nanocrystals in our films is simply \( R \frac{I_{S_{LMM}}}{\theta_{Mo}} \) where \( I_{S_{LMM}} \) is the intensity of the \( S_{LMM} \) Auger transition (labeled in fig. S1(a)) and \( \theta_{Mo} \) is the areal density of Mo atoms in a single layer of MoS\(_2\). \( I_{Mo_{MNN}} \) can be directly calibrated to the absolute coverage of Mo atoms in our films by acquiring an Auger spectrum and RBS data from the same sample region.

Figure S1: (a) The blue curve plots the differentiated dN/dE AES spectrum measured at the center of a sample grown following the temperature vs. time profile shown in Fig. 2 of the main text. The sample was grown at 100 Torr with an Ar flow rate of 150 sccm. SEM images of this sample are shown in Fig. 6(a). The red curve in (a) is the dN/dE Auger electron spectrum measured from a bulk standard. The peak-to-peak heights of the \( S_{LMM} \) and \( Mo_{MNN} \) Auger transitions are identified by the green arrows. Oxygen peaks lie outside the range of the spectrum shown. (b) \( S_{LMM}/Mo_{MNN} \) peak height ratio vs. position relative to sample center for the same sample from which the blue curve displayed in (a) was obtained. The green line is the average of the ratios obtained over the points acquired at positions within 4 mm of the sample center. Within this central region, the S/Mo ratio measured using AES varies by about \( \pm 0.5 \) from its average of 6.7. The S/Mo ratio measured from the bulk standard, is 11.1, which is characteristic of the ratio for stoichiometric MoS\(_2\).
Figure S2(a) shows a height mode AFM image of a MoS$_2$ nanoparticle grown using the temperature vs. time profile presented in fig. 2 of the manuscript. Ar pressure during growth was 100 Torr at a flow of 150 sccm. The average height of this nanocrystal is ~0.7 nm above the SiO$_2$ substrate as shown in fig. S2(b), which is indicative of single layer MoS$_2$. Fig. S2(c) and (d) are similar data obtained from a nanocrystal grown using growth parameters identical to the sample shown in fig. S2(a) but at an Ar pressure of 30 Torr. The average height of the nanocrystal shown in figs. S2(c) and S2(d) is about 1.4 nm above the substrate, which corresponds to bilayer MoS$_2$.

**Figure S2:** (a) 250 nm x 250 nm AFM height image of typical MoS$_2$ nanocrystals produced by our synthesis method. (b) corresponding linescan along the line indicated in (a) shows a nanocrystal height of ~0.7 nm, the expected height for single layer MoS$_2$. (c) 500 nm x 500 nm AFM height image of sample grown at an Ar pressure of 10 Torr. (d) shows the linescan height profile along the line indicated in (c) indicating a 2 ML high nanocrystal as discussed in the text.
Figs. S3(a) and S3(b) show SEM images of the same region of the sample from which the AFM image of fig. S2(c) was acquired. These SEM images demonstrate that larger features grow at lower densities when Ar pressures of less than 30 Torr are used during growth. For comparison, see figs. 5, which display SEM images of samples grown using Ar pressures of 100 Torr. The total Mo coverage measured using RBS is still not accounted for even if all of the MoS$_2$ islands evident in the SEM images are a bilayer high.

**Figure S3:** SEM images showing the relatively large features grown with a low nucleation density across the substrate at Ar pressures \( \leq 30 \) Torr. Scale bars are 10 µm in (a) and 500 nm for (b).

The SEM image shown in fig. S4(a) shows the same sample presented in fig. 4(d). The higher MoO$_3$ source temperature resulted formation of two distinct MoS$_2$ nanocrystal morphologies, as shown. The first are the planar, 2D nanocrystals similar to those shown in figs. 5. The second are 3D features evident with lighter contrast. Fig. S4(b) shows a cross-sectional SEM image of the same sample, which clearly demonstrates the 3D morphology. The height of these features is ~30 nm above the SiO$_2$ substrate.
Figure S4: SEM images of sample exhibiting both 2D and 3D features. (a) plan view and (b) cross-section SEM image. In (b), the SiO$_2$/Si substrate is labeled. 3D refers to the three-dimensional nature of the white features evident in (a), which have an average height of ~30 nm above the SiO$_2$ surface. The white scale bar is 300 nm for both images.

To gain further insight into film morphology variations shown in fig. 6(b), we measured the temperature variation along the substrate. The IR pyrometer employed during this investigation samples a 3mm diameter circular region of the substrate. To improve the spatial resolution of this measurement, we placed a 1mm wide slit between the pyrometer and the sample. To determine the temperature variation along the sample, we translated the slit and pyrometer relative to the sample to acquire the temperature at 2 mm increments. This measurement was repeated 4 times and the average temperatures are plotted vs. sample position, relative to center, in fig. S5. The error bars are 2 standard deviations long. The standard deviation was never greater than 1.5% of the average temperature.
Figure S5: Variation of temperature along substrate. The IR pyrometer samples a ~ 3 mm diameter circular area. To improve the spatial resolution of the temperature measurement, we inserted a 1 mm wide slit in between the pyrometer and the substrate and translated the slit and pyrometer relative to the sample in 2 mm increments.