Controlled Synthesis of High-quality Monolayered α-In_2Se_3 via Physical Vapor Deposition

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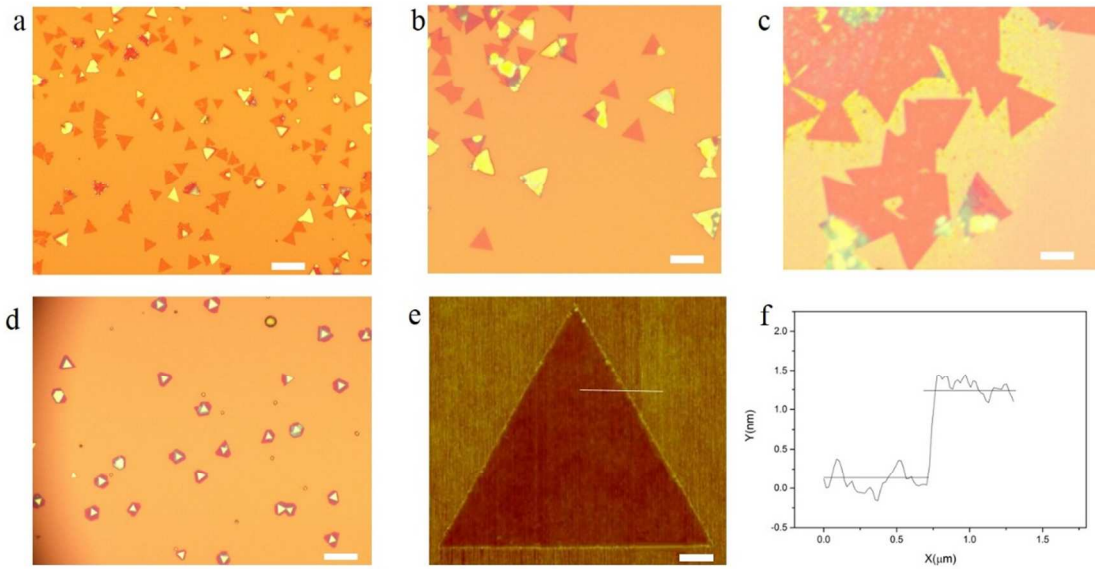

Figure S1. Size dependence on growth time. Growth time: (a) 5 min, (b) 10 min, (c) 12 min, and (d) 15 min. (a-c) Scale bar is 10μm, 10μm, 10μm, and 20 μm, respectively. (e-f) The AFM image and thickness of In_2Se_3 monolayer. Scale bar is 0.2 μm.

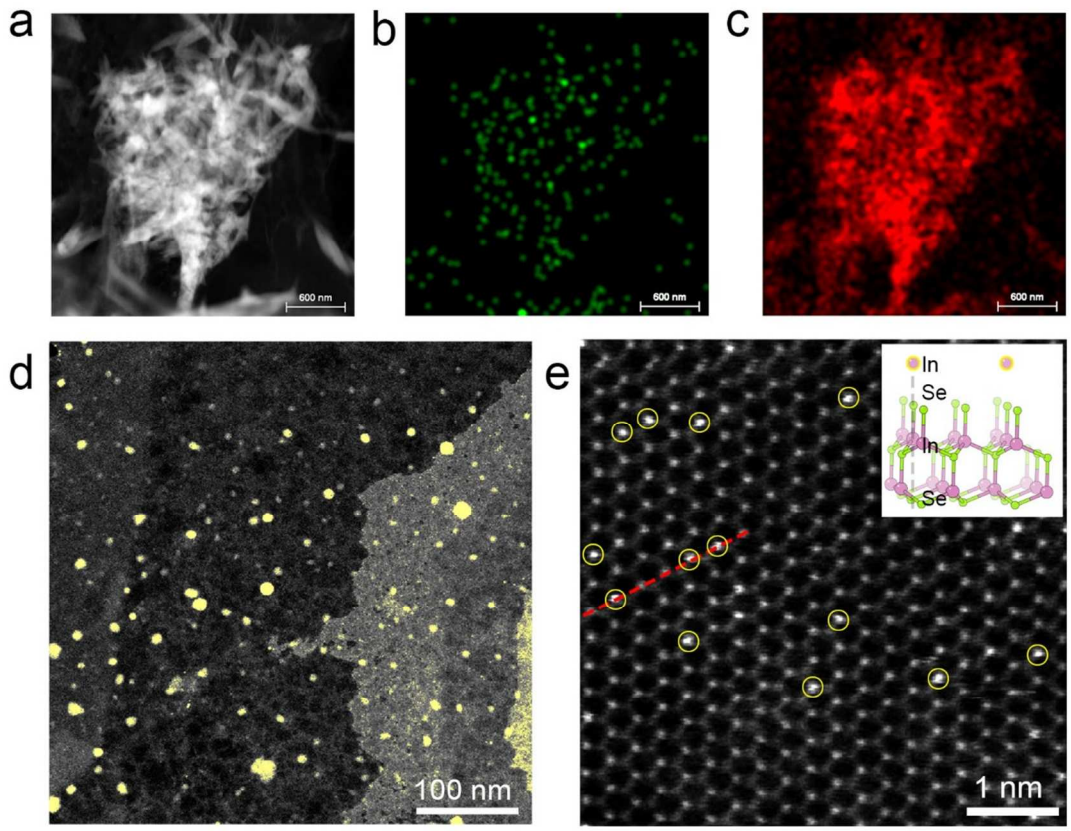

Figure S2 TEM characterization of indium particles. (a) TEM image of an large indium particle ~ 1μm. (b, c) Elemental mapping of Se and In, respectively. Very strong indium signal is found and the Se signal is weak, suggesting this is an indium riches particles. (d) Relative small indium particles (yellow) can be found from the atomic layered In_2Se_3 ranging from a few nm to 20 nm. (e) Some dopant are also found in the monolayer In_2Se_3 as indicated by the yellow circles. More interesting, these sites are identical to the column of Se-In-Se-In-Se. These dopant are more likely to be a single indium atom with a possible stacking shown in the inset.

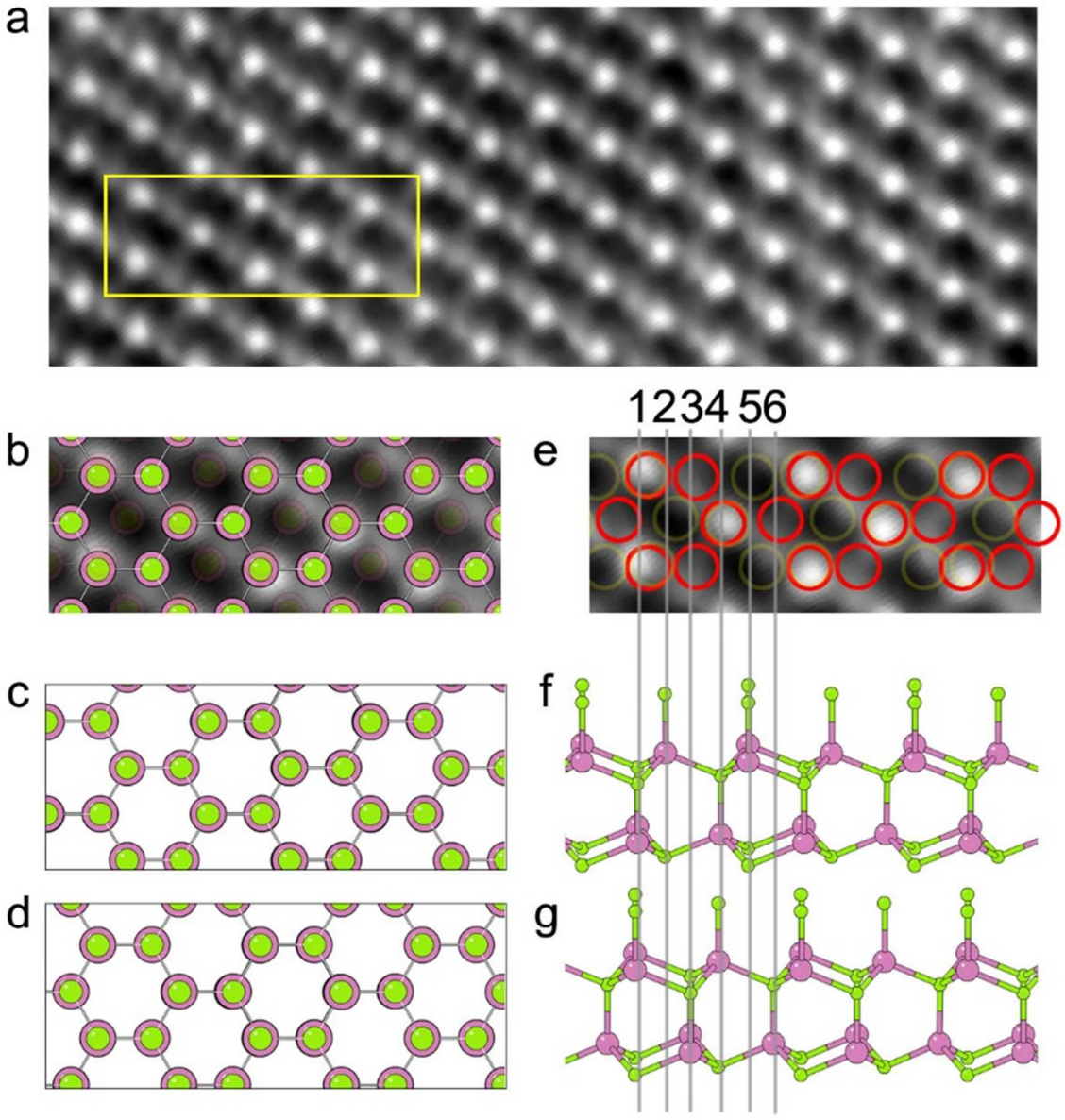

Figure S3. Bi-layered In_2Se_3. (a) Atomically resolved STEM imaging of bi-layer In_2Se_3 with AB stacking. (b) A close-up of bi-layer In_2Se_3 in (a) highlighted by a yellow rectangle, overplayed by a top view model. (c-d) The two different layers of In_2Se_3. (e-f) Side view of bi-layered In_2Se_3 and possible stacking of the atoms for each sites. Yellow and red sites correspond to different layers. A possible stacking of each columns: 1) Se-In-Se-In-Se; 2)Se-In-Se; 3) Se-In; 4) Se-In-Se-In-Se (same as 1 but has only one sites); 5) Se-In-Se (same as 2 but has two sites); 6) Se-In (same as 3 but has only one site).

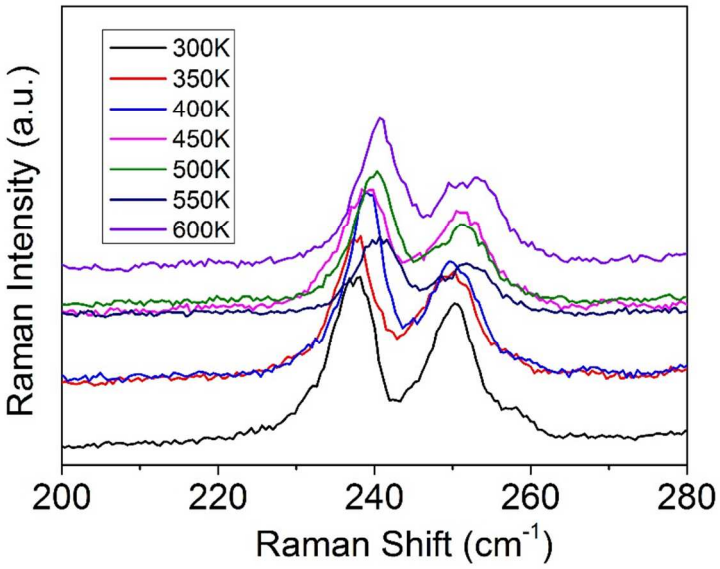

Figure S4. Raman spectra of monolayer In_2Se_3 at various temperatures.