Electronic Supporting Information

Controlled Growth of Large-area Anisotropic ReS_2 Atomic Layer and its Photodetector Application

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1) Thermogravimetric analysis of ReO_3.

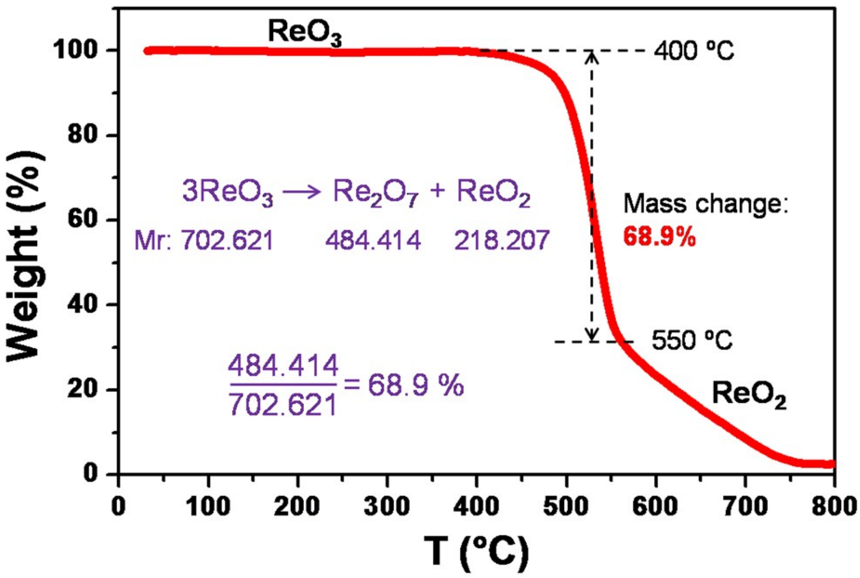

Fig. S1. Thermogravimetric analysis of ReO_3 measured under N_2 atmosphere. The temperature increment is 10 ºC/min.

For reasonable controlling the growth of ReS_2 film with ReO_3 as Re precursor, we studied the pyrolysis process of ReO_3 by using thermogravimetric (TG) analysis. As show in Fig. S1, the weight of ReO_3 decrease sharply in the temperature range from 400 to 550 ºC, attributing to the fast decomposition of ReO_3 into volatile Re_2O_7 and nonvolatile ReO_2 through a disproportionation reaction. For CVD growth of high quality 2D materials, the growth temperature usually need above 500 ºC with the temperature increment up to 25 ºC/min. In this case, the volatile Re_2O_7 (melting point: 220 ºC and boiling point: 360 ºC) would sublimes quickly into vapor at the growth temperature, leading to supersaturated Re source in the CVD system, and thus large amount of flower-like ReS_2 thick flakes preferably obtained when grow with conventional CVD approach.

2) Role of space-confinement in the CVD growth of ReS_2.

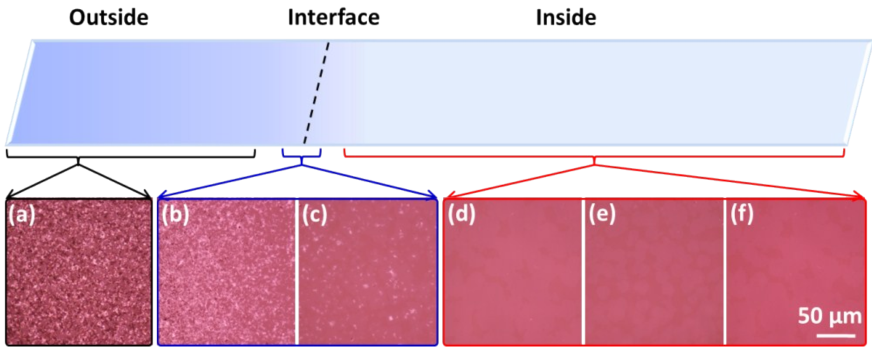

Fig. S2. Schematic diagram of various regions on the growth substrate which is covered by another mica substrate forms a micro-reactor. According to the cover region on the substrate, it was divided into three regions: outside, inside and interface. Corresponding optical images of the ReS_2 film grow at different regions of substrate. Scale bars for all these images are 50 μm.

For more clearly revealing the role of space-confinement in the CVD growth of ReS_2, we performed an in-situ contrast experiment by covering half of the mica surface with another mica layer. The covered part forms a micro-reactor for ReS_2 growth. In this case, the growth substrate can be divided into three regions (Fig. S2), one region at the outside of micro-reactor, another region at the inside of micro-reactor, and the third region at their interface. Clearly, the results of ReS_2 grow at these three regions show obvious difference. Flower-like ReS_2 thick flake with irregular morphology are densely grown on the outside region, while uniform monolayer film can be obtained on the inside region with a transition from thick layer to monolayer at the interface region. The stark contrast of the grown results between outside and inside regions highlights the importance of the constructed micro-reactor, which plays a space-confinement role in the controlled growth of ReS_2 film.

3) Layer number dependent Raman and fluorescence spectrum of grown ReS_2.

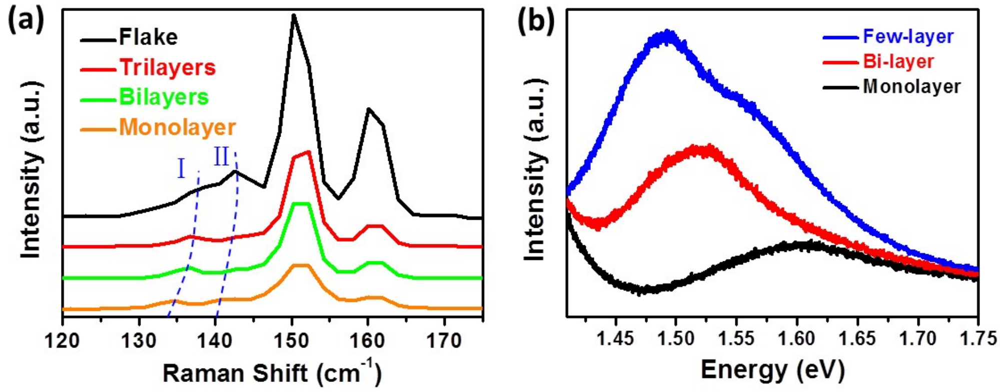

Fig. S3. (a) Raman and (b) fluorescence spectra of CVD grown ReS_2 film with thickness vary from monolayer to few-layer. The energy position of PL increases with decreasing layer numbers, ranging from 1.48 eV of few-layer to 1.61 eV of monolayer, while the PL intensity decreases with layer number decreasing.

4) Substrate dependent growth behavior of ReS_2 atomic layer.

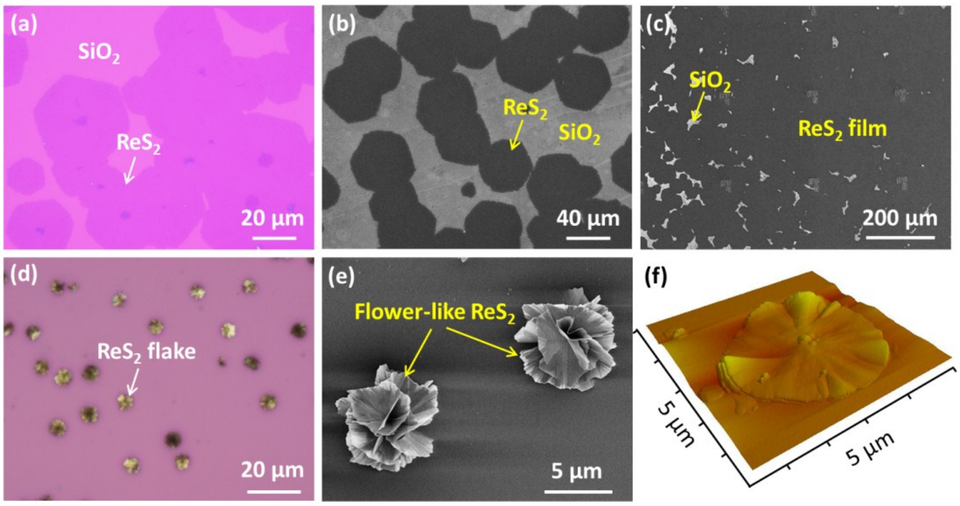

Fig. S4. Morphology characterizations of ReS_2 film grow on mica and SiO_2 substrates. (a) OM and (b,c) SEM images of ReS_2 transferred from mica onto SiO_2 substrate. (d) OM, (e) SEM and (f) 3D AFM images of ReS_2 grown on SiO_2 substrate.

To confirm the difference of the growth behavior of ReS_2 on mica and SiO_2 substrate, we compared the results of ReS_2 grown on these two substrates. Clearly, only thick flake or flower-like structure ReS_2 with small domain size (～5 µm) were grown on SiO_2 substrate. The flower-like structure grown on SiO_2 substrate, indicates an out-of-plane growth, arises much easier in most of our experiment and recent reported work. In contrast, large area, hexagon ReS_2 films with uniform monolayer thickness and large domain (～60 µm) size were grown on the mica substrate, indicating a well controlled epitaxial growth.

5) Temperature dependent crystal quality of CVD-grown ReS_2 atomic layer.

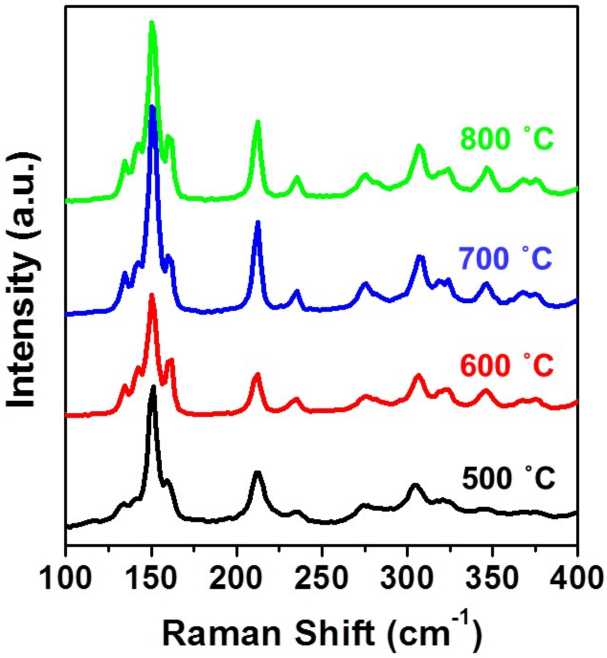

Fig. S5. Raman spectra of monolayer ReS_2 film grow at different temperatures.

6) Photoelectric property of the CVD-grown monolayer ReS_2 phototransistor.

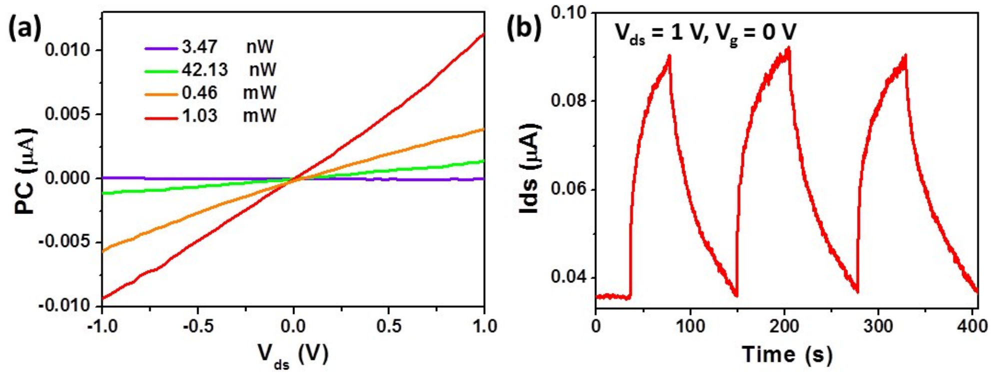

Fig. S6. (a) Photocurrent (PC) as a function of drain–source voltage (V_{ds}) under various light irradiation power at V_g = 0 V. (b) Time-dependence Ids of the device with and without the laser illumination measured at V_{ds} = 1 V and V_g = 30 V.

The dynamic response of our device during the on-and-off switching of an incident light exhibits a fast rising (60-80 ms) while a slow falling (30-50 s), which is compared with the results obtained from exfoliated thick layers ReS_2 and InSe photodetectors,[1, 2] indicates the existence of trap states. Such trap states exacerbated further due to the high surface-to-volume ratio of monolayer ReS_2 film.

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