

## Supporting Information

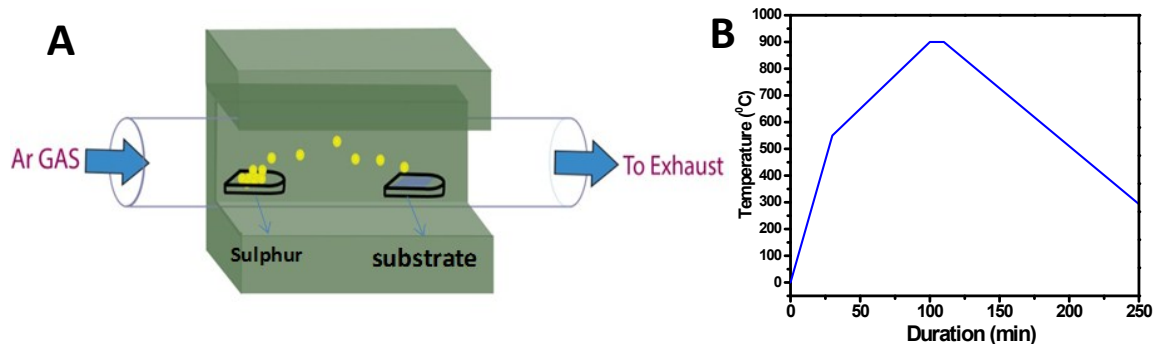
### Controllable Growth of Few-layer Spiral WS<sub>2</sub>

*Prasad V. Sarma, Prasanna Patil, Prahalad Kanti Barman, Rajeev N. Kini and Manikoth M. Shaijumon\**

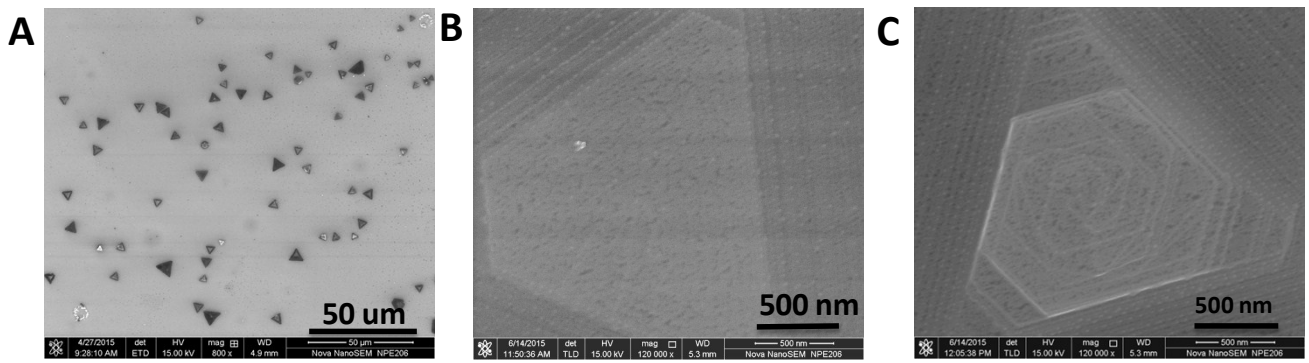
School of Physics, Indian Institute of Science Education and Research Thiruvananthapuram,  
Thiruvananthapuram, Kerala, 695016, India.

#### Contents:

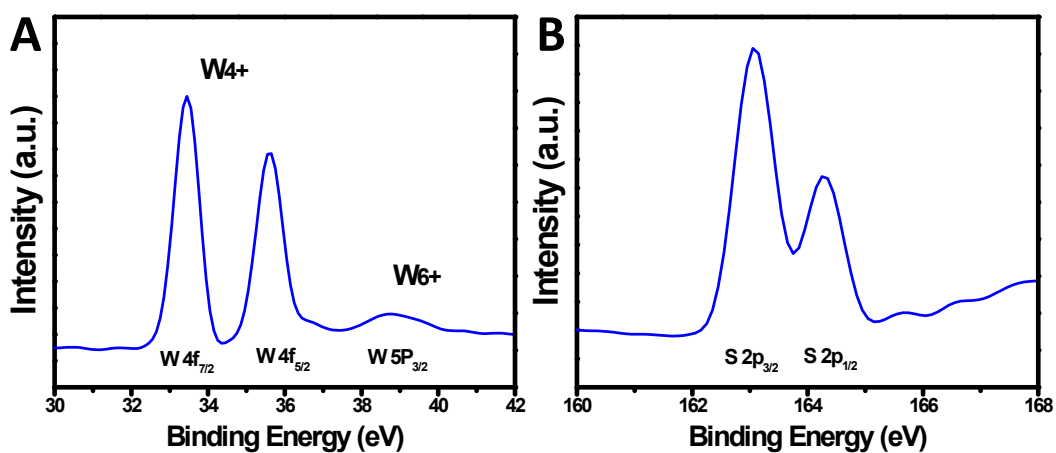
- (1) Schematic of Chemical Vapor Deposition Unit and temperature profile of the deposition
- (2) SEM images of triangular domain growth having Spiral and LBL stacking morphology
- (3) X-ray photoelectron Spectroscopy data from WS<sub>2</sub> domains
- (4) AFM, SEM and Raman mapping data from WS<sub>2</sub> domains grown on substrate where concentration of Tungsten precursor was 0.1mg/ml.
- (5) AFM image and height profile of Ribbon kind of growth observed on top of triangular domains
- (6) SEM images of WO<sub>3</sub> particles dropcasted on the sample before and after annealing at 900 °C without Sulphur
- (7) AFM and SEM images of WS<sub>2</sub> domains grown on substrate where concentration of Tungsten precursor was 0.5mg/ml
- (8) SEM images of WS<sub>2</sub> flakes grown *via* CVD process at different temperatures



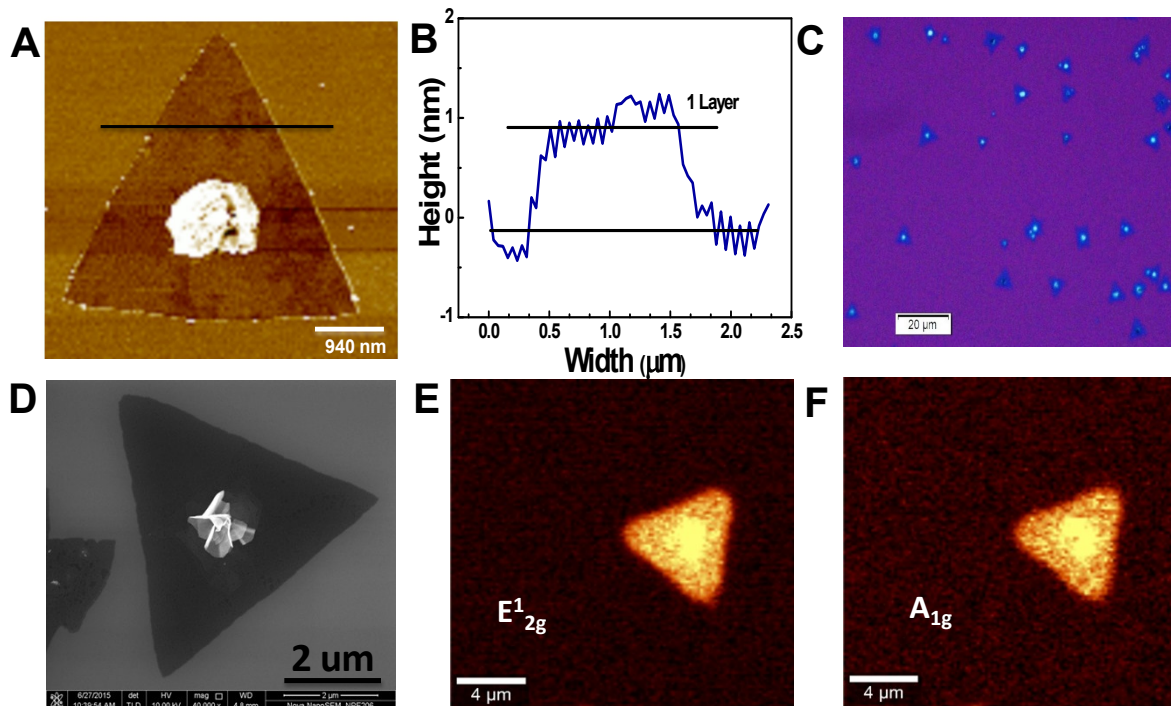
**Fig. S1:** (A) Schematic illustration of  $WS_2$  chemical vapor deposition set up.  $WO_3$  powder drop-casted on  $SiO_2/Si$  substrate is kept in alumina boat placed at the centre of heating zone, while Sulphur powder is kept at upstream end. (B) Temperature profile of chemical vapor deposition. After holding at  $900\text{ }^\circ\text{C}$  for 10 min, furnace is allowed to cool down to room temperature naturally.



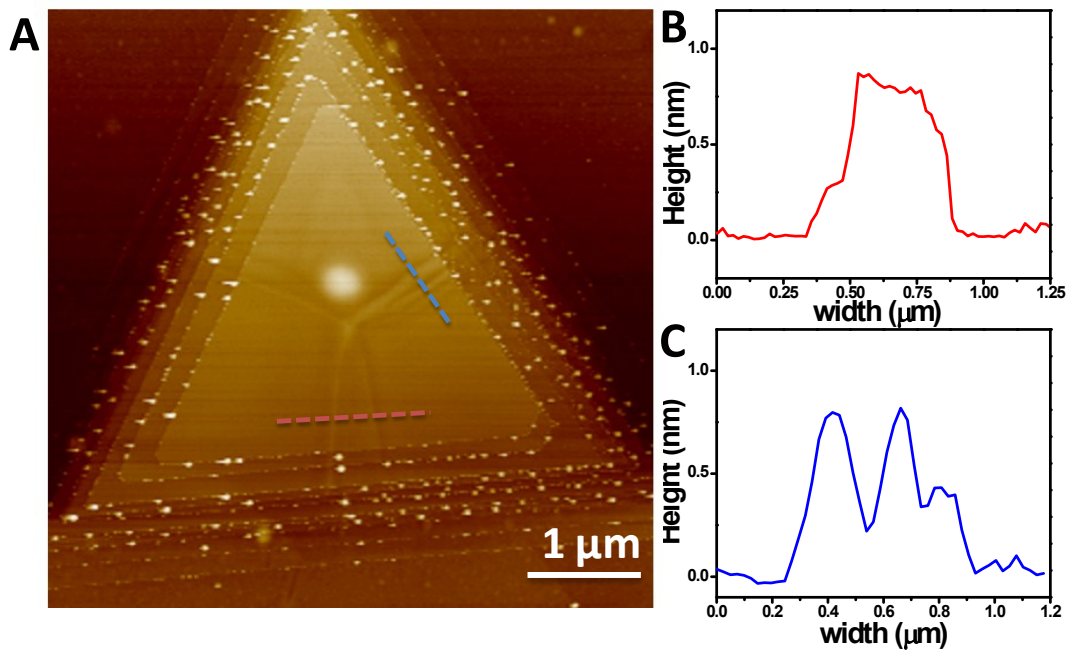
**Fig. S2:** (A) SEM image of CVD grown WS<sub>2</sub>. High resolution SEM images of (B) LBL and (C) SDD grown WS<sub>2</sub> pyramid. (B) shows the flat surface on top of the domain and a clear image of spirals on top of another domain is illustrated in (C).



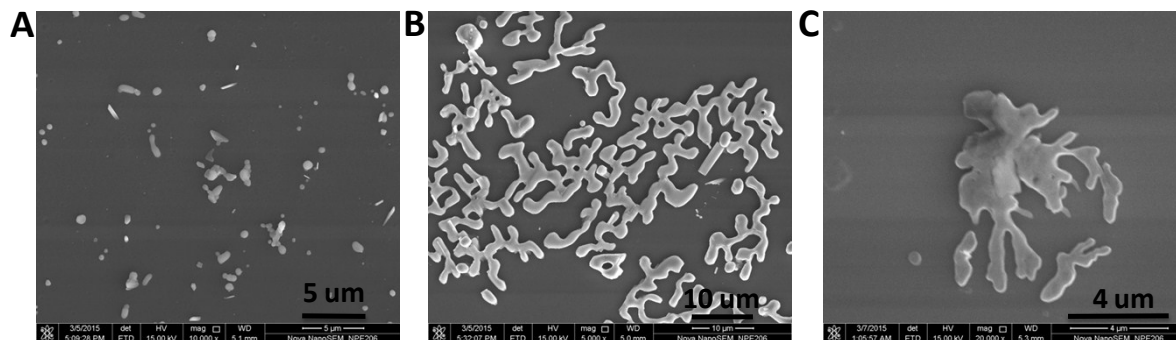
**Fig. S3:** XPS spectra of CVD grown  $WS_2$ . (A) shows characteristic XPS data from Tungsten that clearly illustrate peaks corresponding to  $W^{4+}$  oxidation state. The small intensity peak  $\sim 38$  eV, in (A), attributed to 6+ oxidation state of Tungsten which could be due to the presence of  $WO_3$ . XPS spectrum from Sulphur is shown in (B) where characteristic peaks at 162.3 eV ( $S 2P_{3/2}$ ) and 163.57 eV ( $S 2P_{1/2}$ ) are present.



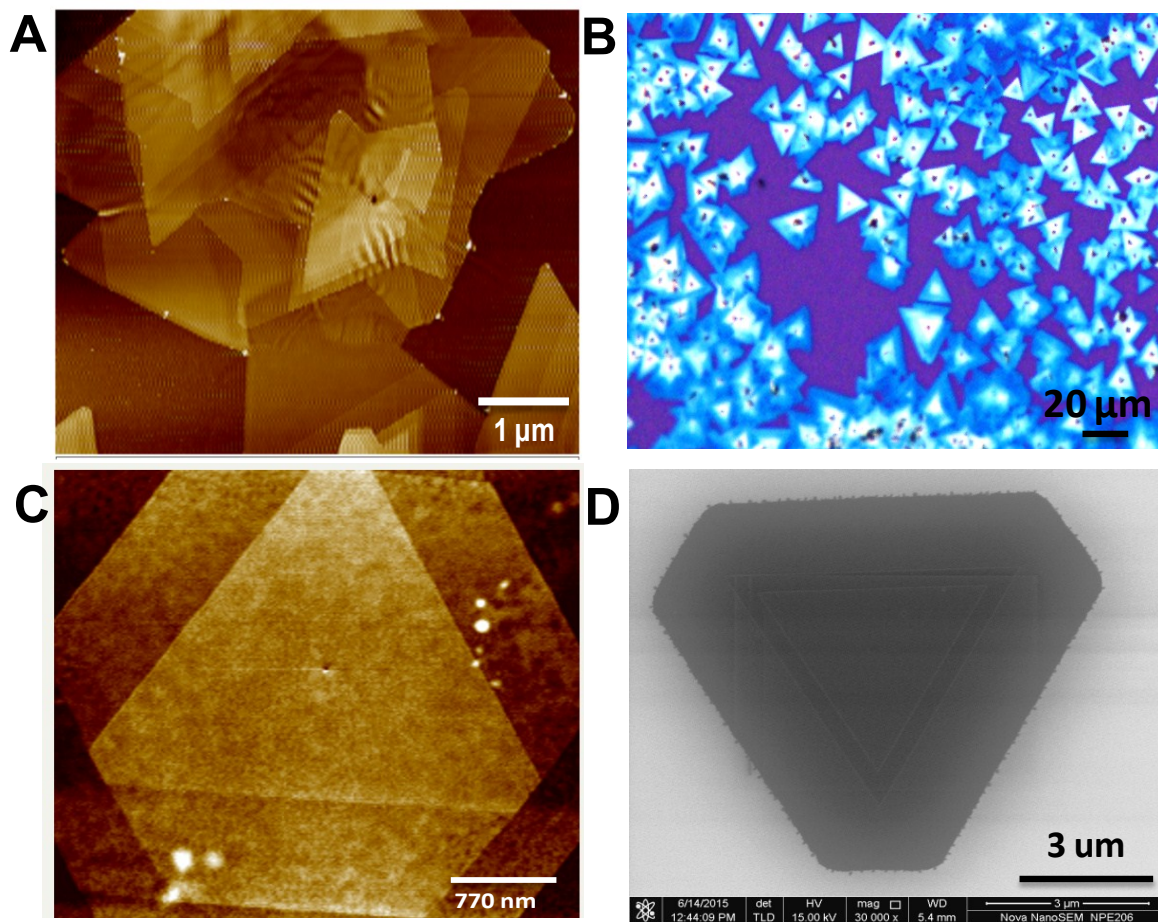
**Fig. S4:** Morphological and Raman characterization of monolayer WS<sub>2</sub> grown by using 0.1 mg/ml concentration of WO<sub>3</sub> precursor. (A) AFM and (B) corresponding height profile of monolayer WS<sub>2</sub>. (C) Optical microscopy image and (D) SEM image of monolayer WS<sub>2</sub> flake (E), (F) Raman mapping of monolayer WS<sub>2</sub> by using 488 nm excitation. Here the domain size is around 4-6  $\mu\text{m}$ . Optical image clearly demonstrate the uniform growth of monolayer WS<sub>2</sub> throughout the substrate. Raman mapping images show uniform Raman intensity throughout the domains except for the particle or cluster at the centre.



**Fig. S5:** (A) Ribbon-like morphology observed on top of WS<sub>2</sub> domains. (B), (C) shows the corresponding height profile along the marked lines in red and blue colors respectively. Heights of these ribbons matches with the thickness of monolayer WS<sub>2</sub>.

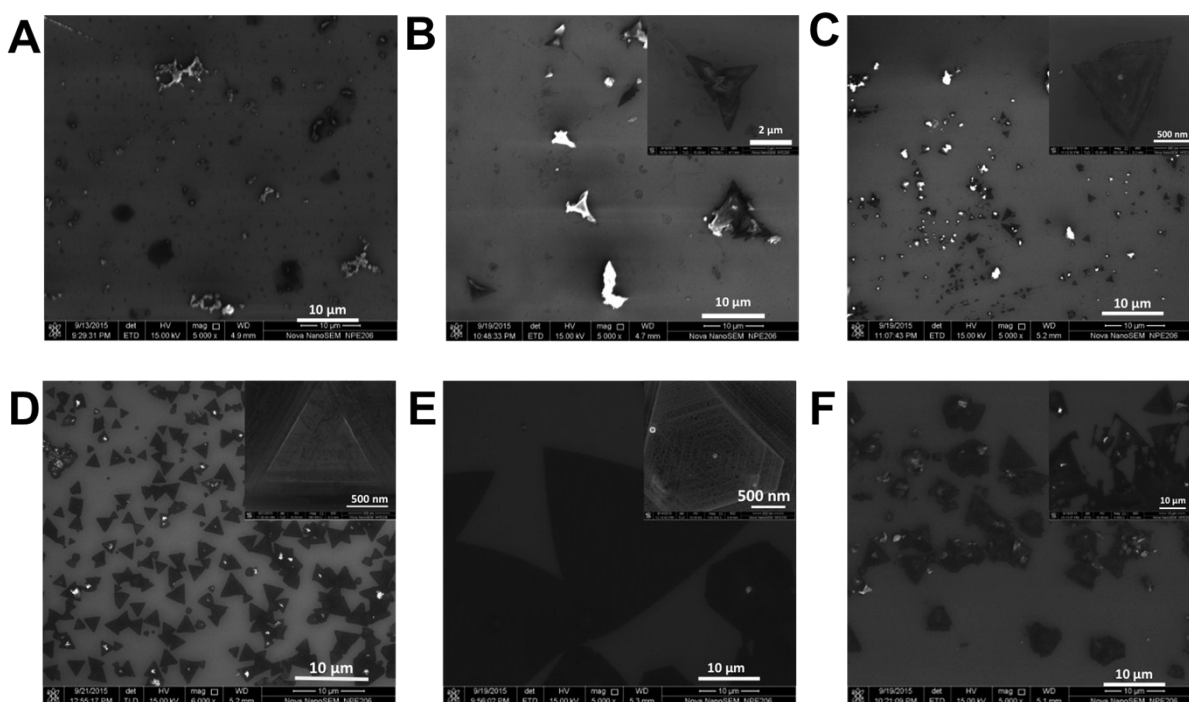


**Fig. S6:** (A) SEM image of  $\text{WO}_3$  powder drop-casted (0.2 mg/ml) on Si/SiO<sub>2</sub> substrate before annealing (B), (C) SEM images of  $\text{WO}_3$  drop-casted sample after annealing at 900 °C under controlled atmosphere of 200 sccm Argon flow. At this temperature  $\text{WO}_3$  powder completely melts and island kind of morphology is formed.



**Fig. S7:** AFM, SEM and optical microscopy characterization of CVD grown WS<sub>2</sub> flakes by using 0.5mg/ml concentration of WO<sub>3</sub> precursor. (A) AFM and (B) optical microscopy images clearly revealing cluttered growth. (C) shows the AFM image of pyramidal centre that follows LBL growth. (D) SEM image of a LBL grown WS<sub>2</sub> domain. Higher concentration of tungsten precursors triggers LBL growth and ends up in pyramids having flat faces on top.





**Fig. S8:** SEM images of WS<sub>2</sub> flakes grown *via* CVD process at (A) 700 °C, (B) 750 °C, (C) 800 °C, (D) 850 °C, (E) 900 °C and (F) 950 °C. Inset shows respective magnified images. All other parameters in the CVD process, including Argon gas flow rate (200 sccm), sulphur amount (500 mg) and WO<sub>3</sub> concentration (0.2 mg/ml in ethanol), remain unchanged. Uniform growth of triangular domains is observed at 850 °C and 900 °C, with respective average domains sizes of 3-5 μm and 15-20 μm. Further increase in CVD temperature to 950 °C yields smaller and broken triangles, while the growth was seen incomplete at temperatures less than 850 °C. 900 °C appears to be the optimum growth temperature for few-layer spiral WS<sub>2</sub> domains, under the given conditions. At 850 °C, growth is mostly thick triangular domains.