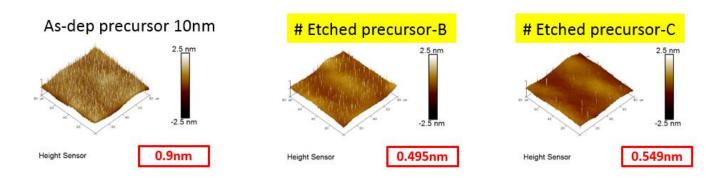


**Figure 1.** Scheme of large-area and continuous TMDs film synthesized by general CVD nucleation and growth process with local non-uniformity.

**Figure 2.** Schematic process flow of specific layer TMDs film synthesis with large area uniformity through ultrathin and flat precursor film preparation. An ion-assisted atomic-scale plasma etch is applied for precursor etching. Following by sulphurization process, uniform and layered TMDs can be precisely controlled.



**Figure 3**. Images of AFM roughness analysis in (a) as-deposited (b) partially etched and (c) heavily etched precursor film (WO<sub>3</sub>) treated by ion-assisted plasma etching approach. The rms data for etched WO<sub>3</sub> film are around 0.5nm  $\pm 0.05$ nm

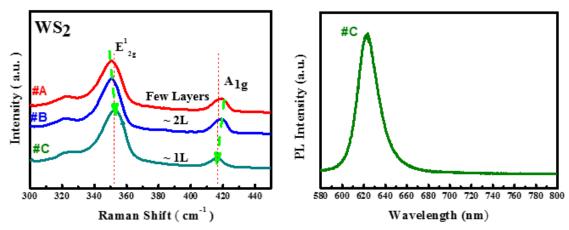


Figure 4. Raman and photoluminescence spectrum of  $WS_2$  film synthesized from the sulphurization of etch precursor films.