## **Supplementary Figures**



Supplementary Figure 1. The W core level XPS spectrum obtained on a SCSA deposited WSe<sub>2</sub>

film.



**Supplementary Figure 2**. A single flake layer  $WSe_2$  film on a glass substrate prepared by the SCSA method. The area of the substrate is 22.5 cm<sup>2</sup>. A few scratches are noted on the film due to rough handling.



**Supplementary Figure 3**. Additional cross-sectional SEM images. Panel (a) shows a tilt-shift view (stage at 3°) of the SCSA film to highlight the single-flake nature of the film and the slight roughness induced by the FTO surface. Panel (b) shows a representation of the large aggregates present in the AC film. The SEM images are coloured to highlight the F:SnO<sub>2</sub> substrate (blue) and the WSe<sub>2</sub> (brown).



**Supplementary Figure 4**. The J-V data for the conductivity measurement on the 60  $\mu$ m channel device as described in the main text. We note the shift of the minimum current by 1 V in conductivity measurement of AC film, which can be attributed to capacitance created during the scan.



**Supplementary Figure 5**. Cross-sectional SEM image of a 2 layer SCSA WSe<sub>2</sub> film. A similar morphology is observed as in the 1 layer case (Figure 3c, main text)



Supplementary Figure 6. Linear scanning voltammetry curves for the WSe<sub>2</sub> under constant dark or illumination conditions. Scan rate 10 mV s<sup>-1</sup> in 1M H<sub>2</sub>SO<sub>4</sub> electrolyte.



**Supplementary Figure 7**. Linear scanning voltammetry under intermittent illumination of the best performing 2-layer SCSA WSe<sub>2</sub> film before (red curve) and after (blue curve) Pt deposition.



**Supplementary Figure 8.** H<sub>2</sub> measurements and Faradaic efficiency calculation. (a) Shows the calibration curve constructed using Pt electrodes. (b) Shows the raw photocurrent data from the SCSA WSe<sub>2</sub> photocathode measured.



**Supplementary Figure 9**. Comparison of the linear scanning voltammetry under intermittent 1 sun illumination in 1M H<sub>2</sub>SO<sub>4</sub> electrolyte of the Pt-functionalized AC WSe<sub>2</sub> film.

## Supplementary Tables

Device geometry			Measured conductivity (S m <sup>-1</sup> )	
Channel	Length	Channel Width (mm)	SCSA film	AC film
(µm)				
60		1	6.5×10 <sup>-4</sup>	1.8×10 <sup>-4</sup>
20		3	7.2×10 <sup>-4</sup>	2.4×10 <sup>-4</sup>

## Supplementary Table 1. Summary of conductivity measurements.