## Electronic Supplementary Information (ESI)

## Controlled Growth of MoS<sub>2</sub> Nanopetals and Their Hydrogen Evolution Performance

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Fig. S1 SEM images show features of spiral growth. (a and b) Top-view SEM images of sample A4. (c) Cross-section SEM image of sample C2.



Fig. S2 XRD of Sample A4, B2, C2, D, E and 500 nm SiO<sub>2</sub>/Si substrate.

XRD measurements confirm the complete chemical conversion from MoO<sub>3</sub> to MoS<sub>2</sub>. High resolution X-ray scans of the five investigated samples A4, B2, C2, D, E and 500 nm SiO<sub>2</sub>/Si substrates are compared in Fig. S2. A strong sharp (002) diffraction peak is observed at  $2\theta = 14.5^{\circ}$ , corresponding to the MoS<sub>2</sub> films along the basel plane. Higher order peaks (004), (006) and (008) are observed, confirming that MoS<sub>2</sub> thin films are highly basal-oriented. All peaks are indexed to the MoS<sub>2</sub> according to JCPDS card No. 37-1942.



Fig. S3 (a) TEM image of sample B2. (b) Corresponding SAED pattern.

TEM samples are prepared via a following procedure: some products are stripped off and dispersed in alcohol with the aid of ultrasonic vibration for 1 h, and then a drop of the solution is transferred onto a standard holey carbon covered copper TEM microgrid.



Fig. S4 (a) SEM image of sample B2. (b and c) SEM images of sample C2.

Some particles and sheets which arised from the incomplete sulfuration were sometimes observed either at the roots or among the  $MoS_2$  nanopetals (Fig. S4a). Influenced by dense sulphur atmosphere, large layers sometimes formed on the top of  $MoS_2$  nanopetals. The edges were hided (Fig. S4b and S4c ).



Fig. S5 (a and b) Cross-section SEM images of sample D. (c and d) Cross-section SEM images of sample E. Compared regions are shown with blue (vertical  $MoS_2$  nanopetals) and red (horizontal  $MoS_2$  films) in this two samples (b and d).

According to cross-section SEM images of sample D and E, we can clearly see the boundary between vertical  $MoS_2$  nanopetals, horizontal  $MoS_2$  films and 500 nm SiO<sub>2</sub> layer. Different areas can be derived by image software to calculate the ratio of vertically aligned  $MoS_2$  nanopetals / total  $MoS_2$  (F).<sup>1</sup> We calculated it as the ratio between the area of vertically aligned  $MoS_2$  nanopetals (S<sub>v</sub>, blue) and horizontal  $MoS_2$  films (S<sub>h</sub>, red) according to:

$$F = \frac{S_v}{S_v + S_h} \times 100\%$$

This type of calculation requires the sheets to be aligned and close-packed in 3D space. With the increase of the growth temperature from 900 to 950  $^{\circ}$ C, vertically aligned MoS<sub>2</sub> nanopetals and horizontal MoS<sub>2</sub> films both become thicker. The value of F decreased from 82.1% to 79.5%.



Fig. S6 XRD data of products synthesised with  $S/MoO_3$  mass ratio less than 10:1.



Fig. S7. (a and b) SEM images of  $MoS_2$  nanopetals grown on the CNT films at 800 °C for 1 h. (b) Raman spectra of the CNT films and  $MoS_2/CNT$  films.



Fig. S8 (a and b) Cross-section SEM images of  $MoS_2$  nanopetals on the CNT films. (c) TEM image of  $MoS_2$  nanopetals on the CNT films (Inset: Line-scan EDS spectra). (d) HRTEM image (Inset: corresponding FFT patterns).

Fig. S8a and S8b show the interfacial structure of  $MoS_2$  on the CNT films, assuring that there is a robust atomic connection between the interface through which efficient electron transfer may occur. Fig. S8c suggests that strong adhesion between  $MoS_2$  nanopetals and the CNT film is still maintained after ultrasonic vibration for 1 h. Fig. S8d clearly shows different regions of  $MoS_2$  and CNTs with crystal lattice. In addition, the FFT pattern clearly identifies diffraction rings corresponding to the planes of hexagonal-phase  $MoS_2$  (Inset of Fig. S3d)

## References

S. Esconjauregui, R. Xie, M. Fouquet, R. Cartwright, D. Hardeman, J. Yang and J. Robertson, J. Appl. Phys., 2013, 113, 144309.