Electronic Supplementary Information (ESI)

Hydrogenation of Monolayer Molybdenum Diselenide *via* Hydrogen Plasma Treatment

Kyung Yeol Ma,[ae] Seong In Yoon,[ae] A-Rang Jang,[af] Hu Young Jeong,[d] Yong-Jin Kim,[e] Pramoda K. Nayak,[a] and Hyeon Suk Shin*[bce]

[a] Department of Energy Engineering, [b] Department of Chemistry, [c] Low Dimensional Carbon Materials Center, [d] UNIST Central Research Facilities (UCRF), Ulsan National Institute of Science and Technology (UNIST), UNIST-gil 50, Ulsan 44919, Republic of Korea

[e] Center for Multidimensional Carbon Materials (CMCM), Institute of Basic Science (IBS), UNIST-gil 50, Ulsan 44919, Republic of Korea

[f] Department of Engineering Science, University of Oxford, Oxford OX1 3PJ, United Kingdom

Corresponding author

* Hyeon Suk Shin: shin@unist.ac.kr

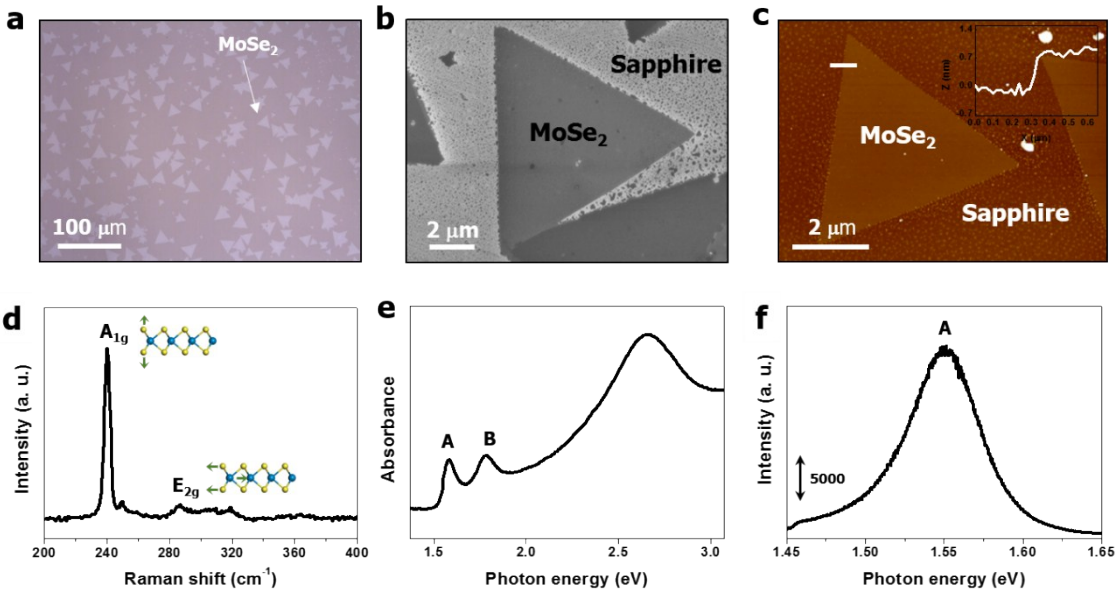


Fig. S1 Microscopic and spectroscopic characterization of as-prepared monolayer $MoSe_2$ by CVD method. (a) optical microscopy, (b) scanning electron microscopy image and (c) atomic force microscopy height topography. (Inset) Height profile of white line marked in (c). (d) Raman, (e) UV-vis-NIR and (f) photoluminescence spectrum measured on sapphire substrate.

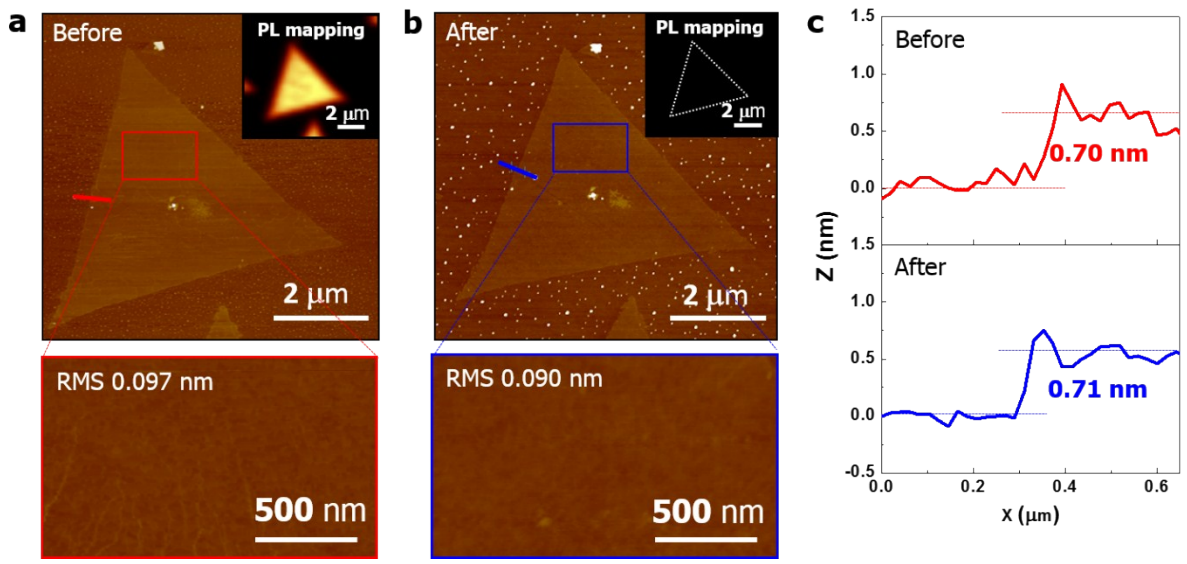


Fig. S2 AFM height topography of monolayer $MoSe_2$ on sapphire (a) before and (b) after 20 W for 40 sec hydrogen plasma treatment. Inset: homogeneous PL (1.56 eV) peak of monolayer $MoSe_2$ (a) before and (b) after hydrogenation. (c) Height profile of (a) and (b). $MoSe_2$ flakes remained intact after H_2 plasma treatment.

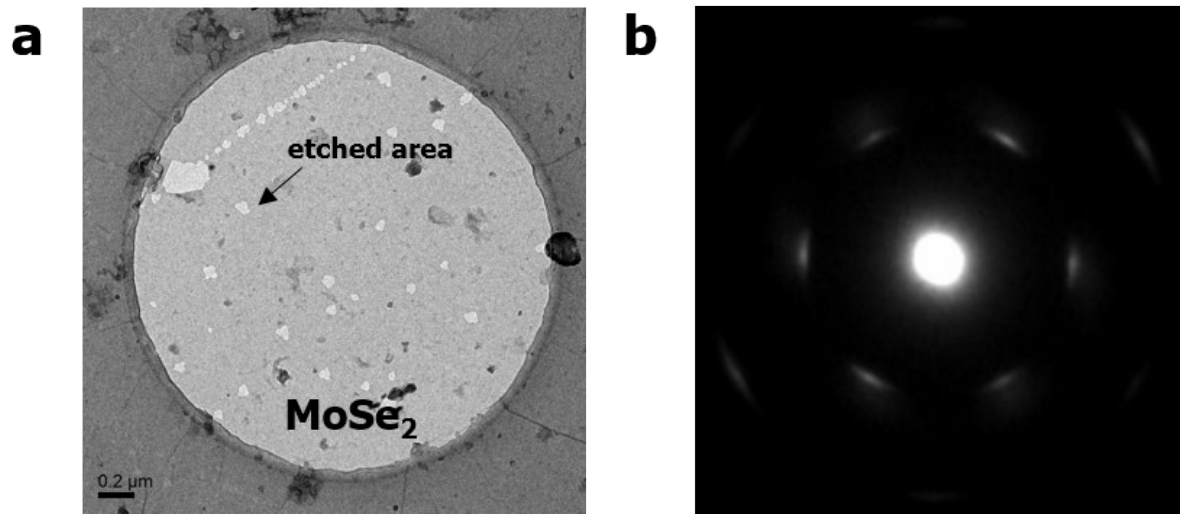


Fig. S3 (a) Bright field image and (b) SAED pattern of $MoSe_2$ treated for 60 seconds, etching of $MoSe_2$ was observed in the $MoSe_2$ treated for 60 seconds.

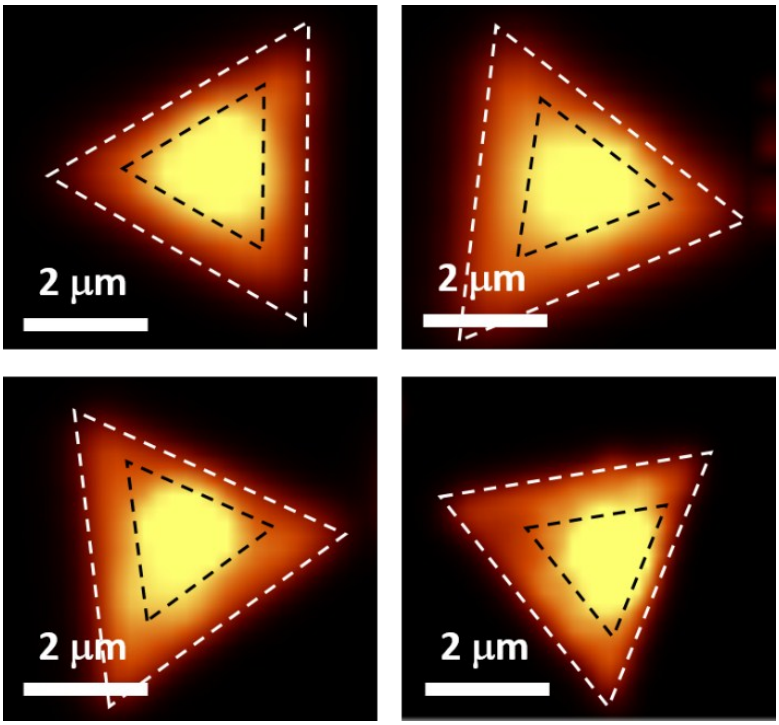


Fig. S4 PL intensity (the A exciton at 1.56 eV) mapping of 4 hydrogenated $MoSe_2$ flakes at early stage (2 seconds). The initial site of hydrogenation is the edge of the flake because the reduction of PL intensity by the hydrogenation was observed at the edge. In the meantime, the interior parts of flakes show strong PL intensity, indicating no hydrogenation. For reference, look at the PL mapping image of a $MoSe_2$ flake before the plasma treatment in the inset of Fig. S2(a).

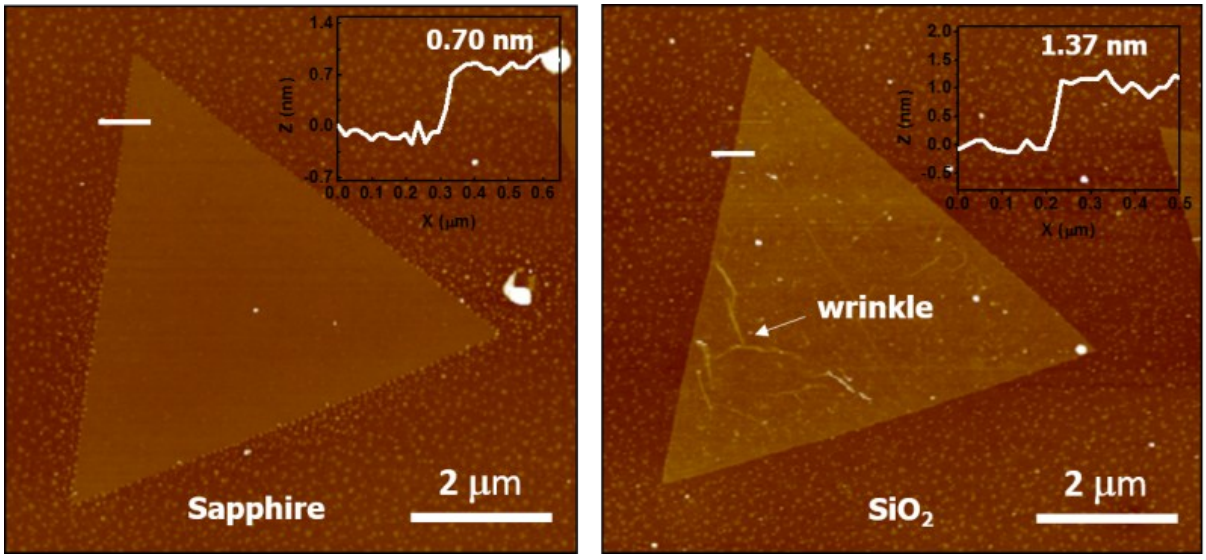
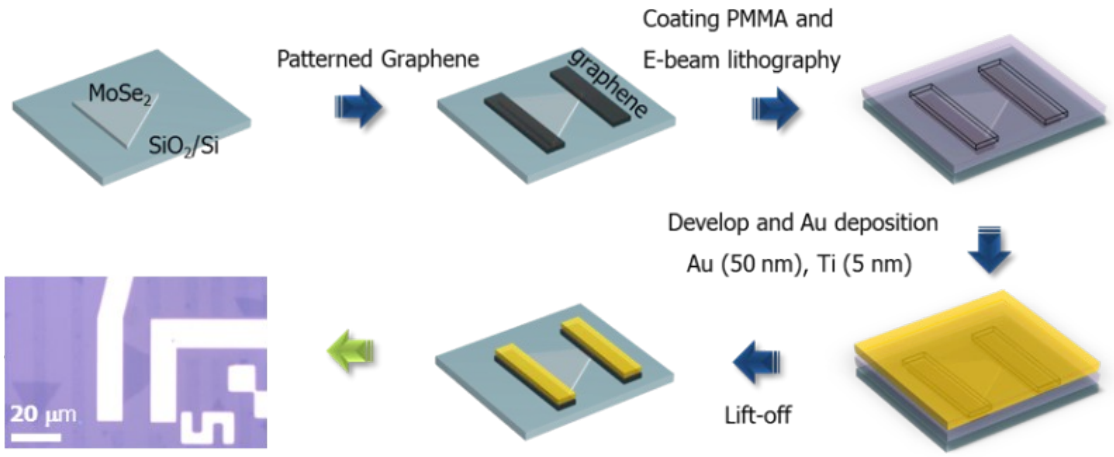


Fig. S5 AFM images of as-prepared $MoSe_2$ on sapphire (growth substrate) and SiO_2/Si substrate (after transfer).



Schematic description of $MoSe_2$-channel back-gate FET device fabrication process after the transfer of $MoSe_2$ to Si/SiO_2 wafer and optical microscopic image of device.

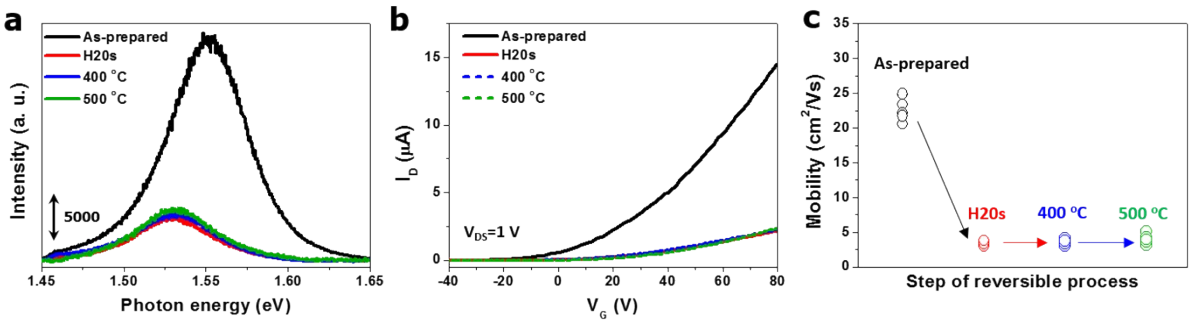


Fig. S6 Non-reversible hydrogen desorption of hydrogenated $MoSe_2$ by heat treatment. (a) photoluminescence spectra of the as-prepared, hydrogenated $MoSe_2$ and heat treated hydrogenated $MoSe_2$. (b) Electron transport characteristics (I_D-V_G) also exhibits that reversible hydrogen adsorption and desorption were not observed in hydrogenated $MoSe_2$ for 20 seconds. (c) Calculated electron mobility values of 6 $MoSe_2$ FETs.