Supporting information

Electronic-dimensionality Reduction of Bulk MoS_2 by Hydrogen Treatment

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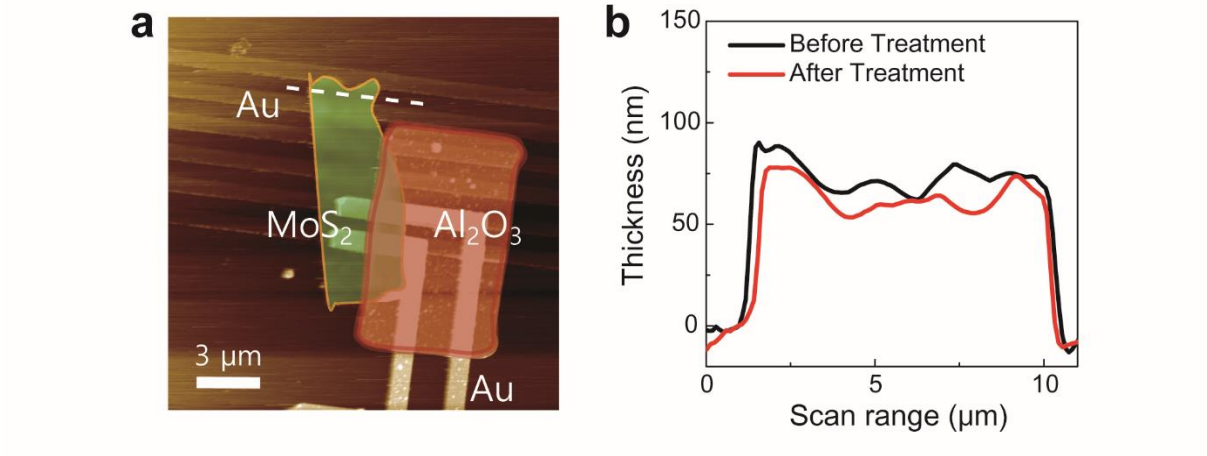

Figure S1: (a) The atomic force microscopy (AFM) image and (b) AFM height profile along the white line in the AFM image before (black line) and after (red line) hydrogen treatment. The average thickness of exfoliated few-layer MoS_2 after the treatment is reduced by 9.85 nm after the treatment.

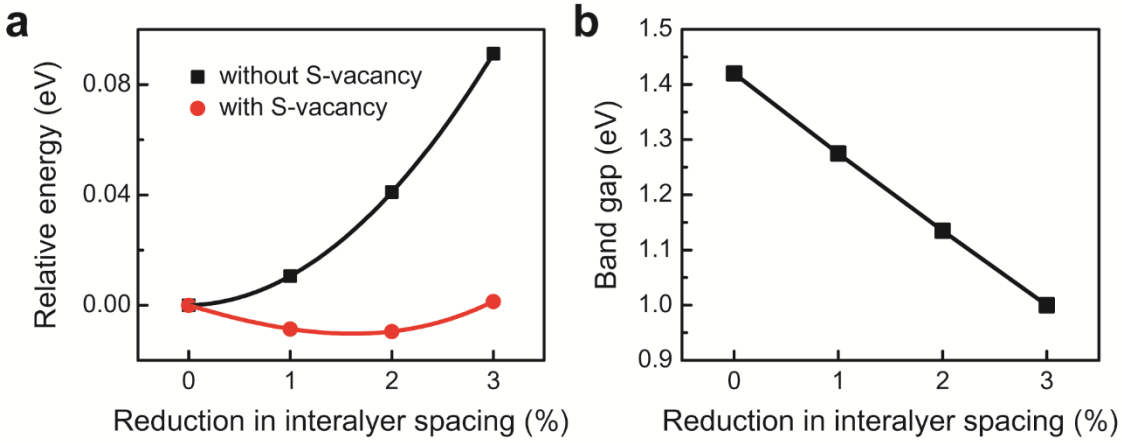

Figure S2: (a) Results of the relative energy of bilayer MoS_2 with (red) and without (black) a sulfur vacancy as a function of the reduction in interlayer spacing. For each, the total energies corresponding to the optimized interlayer distance (3.183 Å) of the vacancy-free pristine MoS_2 are taken as reference energy (zero). In contrast to the pristine MoS_2, the relative energy of the vacancy-included MoS_2 decreases as the interlayer distance reduces, reaching its minimum at around 2 % reduction of the interlayer distance. This indicates that the presence of a sulfur vacancy in hydrogen-treated MoS_2 results in reduced interlayer spacing between MoS_2 layers. (b) The calculated band gap energy of the vacancy-free pristine MoS_2 as function of the interlayer spacing. The bandgap energy is reduced by about 0.285 eV at the 2 % reduced interlayer spacing, which corresponds to the equilibrium interlayer distance for MoS_2 with a sulfur vacancy.

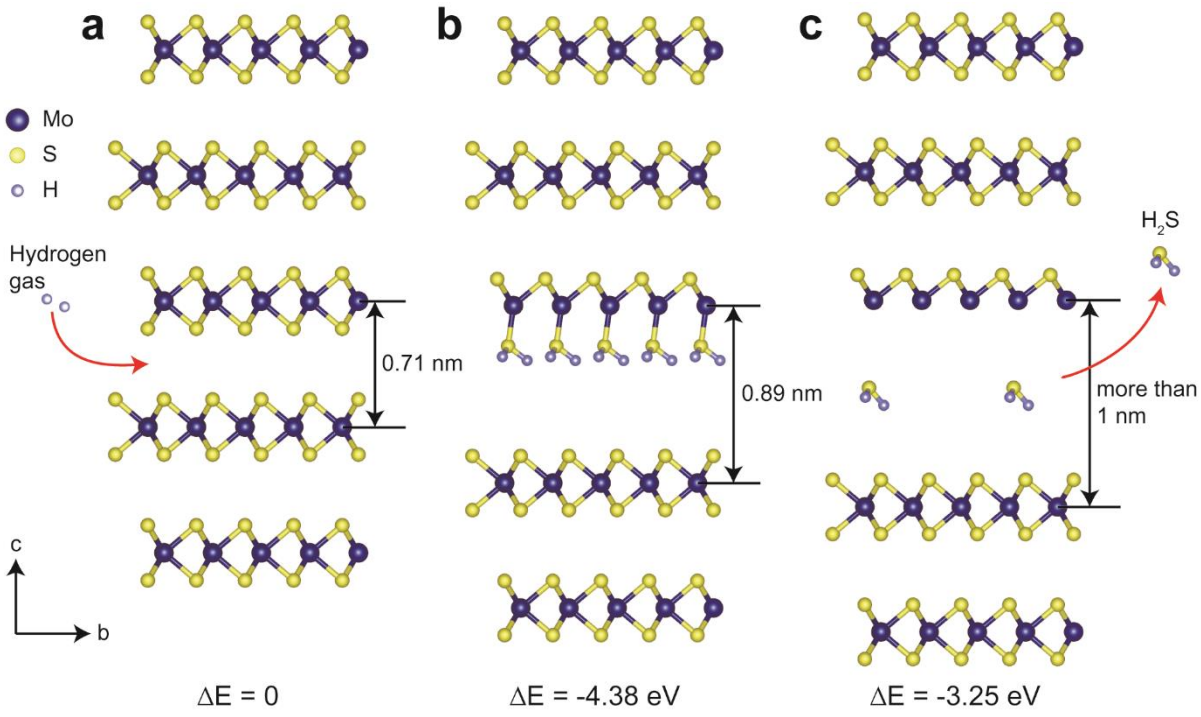

Figure S3: The simulation of the process of the hydrogen treatment. (a) Hydrogen molecule easily penetrates into the particular layer of MoS_2. The interlayer distance between the Mo planes is found to be about 0.71 nm. The total energy of MoS_2 before hydrogen treatment is taken as reference energy (zero). (b) The system energy with the chemical reaction between the hydrogen and sulfur atoms, where both H atoms of H_2 chemisorb to one of S atoms in MoS_2, is lower by ~4.38 eV than that without the chemical reaction. The Mo-Mo interlayer distance increases by about 0.18 nm compared to the distance without the reaction. (c) Finally, sulfur escapes from the crystal through the formation of H_2S. In this case, the distance is more than 1 nm, and the system energy is still lower than that without the hydrogen treatment. The red arrows in (a) and (c) indicate the in- and out-movement of the hydrogen gas away from the MoS_2 layers, respectively.