Supplementary Information

Effect of Sulfur Vacancy on Geometric and Electronic Structures of MoS_2 Induced by Molecular Hydrogen Treatment at Room Temperature

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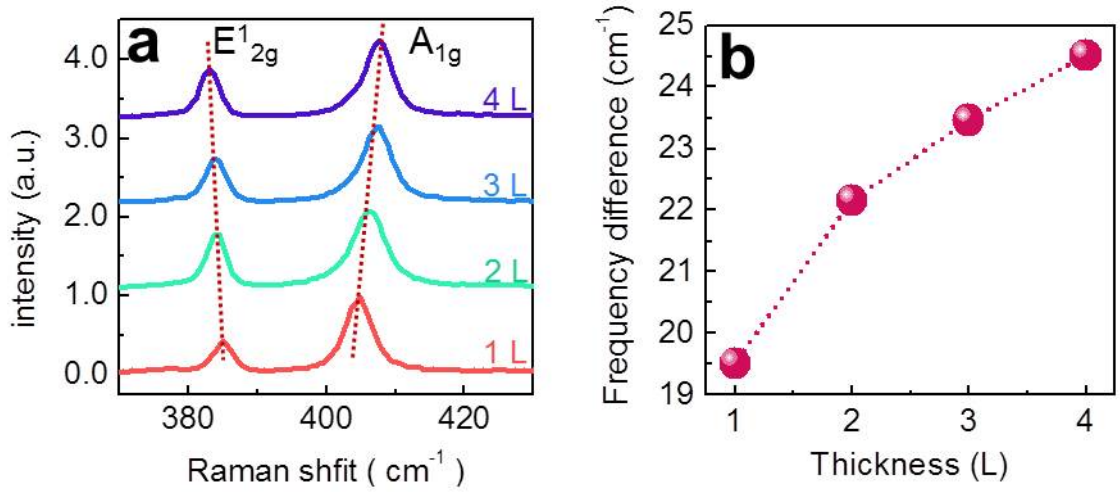

Figure S1. (a) The number of layer dependence of Raman spectra. (b) As the thickness increases, the frequency difference between E^1_{2g} and A_{1g} increases from 19.5 to 24.5 cm^{-1}. It is consistent with the previous literature.[S1-S2]

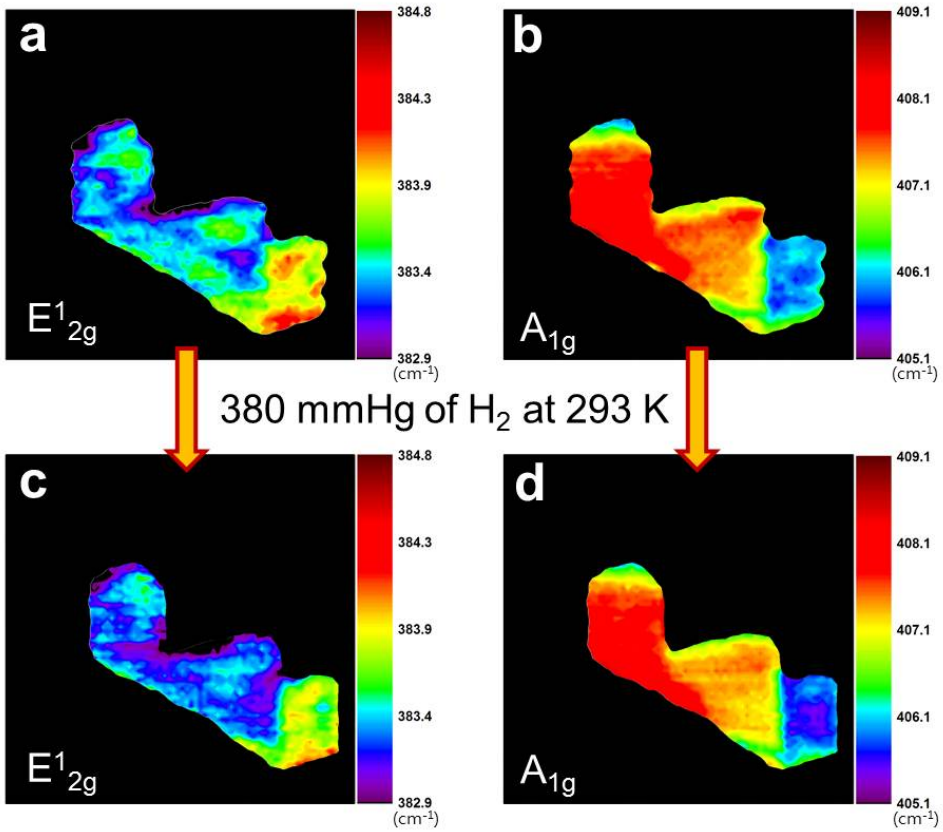

Figure S2. The red-shift of Raman spectra after exposure to 380 mmHg of H_2 at room temperature. First, we prepared the chamber for Raman study in vacuum. Second, we obtained (a)**,** E^1_{2g} and (b)**,** A_{1g} peaks in vacuum and finally the both peaks were extracted after H_2 exposure. (c) and (d)**,** The red-shift occurred in both peaks.

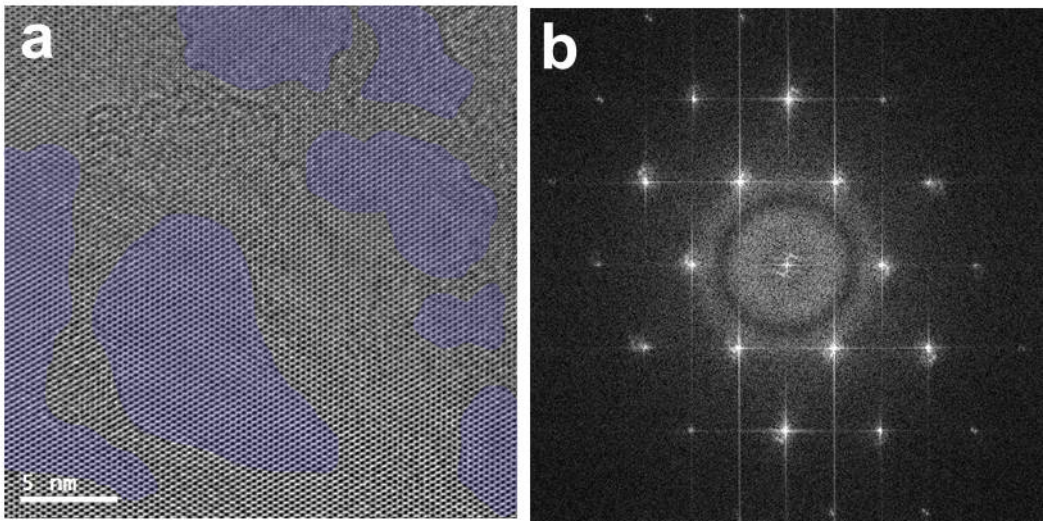

Figure S3. TEM study of multilayer MoS_2. (a) TEM image of the surface shows that several pieces of additional layers were attached on the main layer. The coloured parts indicate the even surface of the main layer. (b) ED pattern of the TEM image.

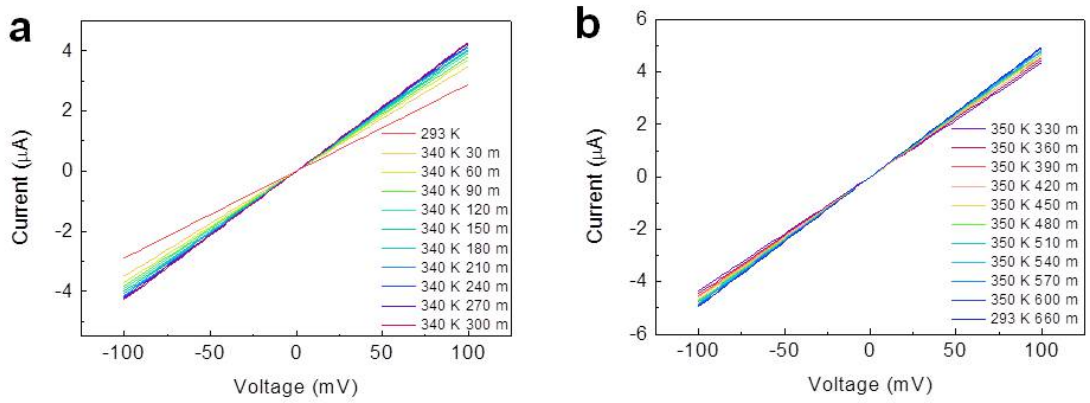

Figure S4. Temperature-dependent electrical transport properties of MoS_2 in high H_2 pressure from 293 K to 350 K. (a) and (b) *I-V* characteristics.

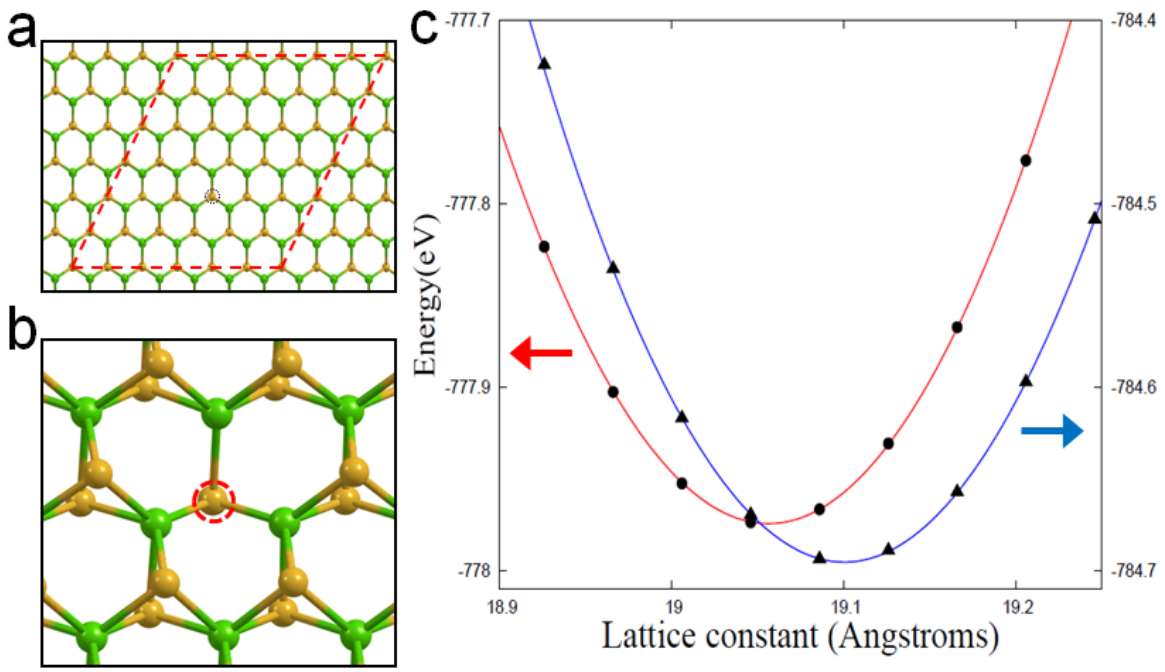

Figure S5. Structure and lattice length of pure single layer MoS_2 and that with sulfur vacancy per 6x6 supercell. (a) The geometry of the MoS_2 6x6 supercell with one sulfur vacancy. (b) A zoomed-in view of the region of the sulfur vacancy. (c) Lattice constant of the triangular lattice of the 6x6 supercell of perfect MoS_2 (solid triangles) and that with one sulfur vacancy (sold circles).

References

S1. Lee, C.; Yan, H.; Brus, L. E.; Heinz, T. F.; Hone, J.; Ryu, S., Anomalous Lattice Vibrations of Single- and Few-Layer MoS_2. *ACS Nano* **2010**, *4*, 2695-2700.

S2. Li, H.; Zhang, Q.; Yap, C. C. R.; Tay, B. K.; Edwin, T. H. T.; Olivier, A.; Baillargeat, D., From Bulk to Monolayer MoS_2: Evolution of Raman Scattering. *Adv. Funct. Mater.* **2012**, *22*, 1385-1390.