**Electronic Supplementary Information**

# Role of hydrogen in chemical vapor deposition growth of $MoS_2$ atomic layers

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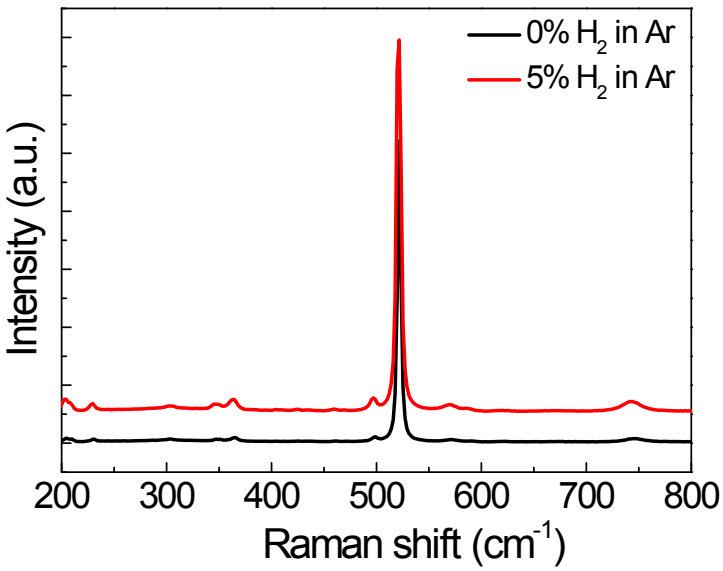


**Figure S1.** Raman spectra of the samples obtained with 0% and 5% $H_2$ after first-step annealing. No typical peaks ($E^1_{2g}$ and $A_{1g}$) are observed when $H_2$ content is below 5%.

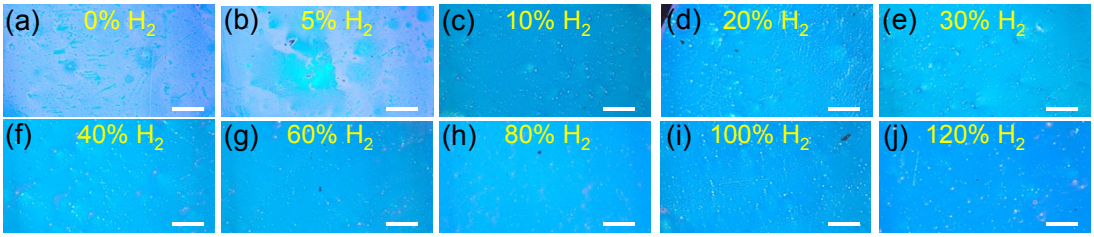


**Figure S2.** Low magnification optical microscopy images of samples obtained with different $H_2$ concentrations (in Ar) after first-step annealing. (a) 0%. (b) 5%. (c) 10%. (d) 20%. (e) 30%. (f) 40%. (g) 60%. (h) 80%. (i) 100%. (j) 120%. Scale bars: 100 μm.

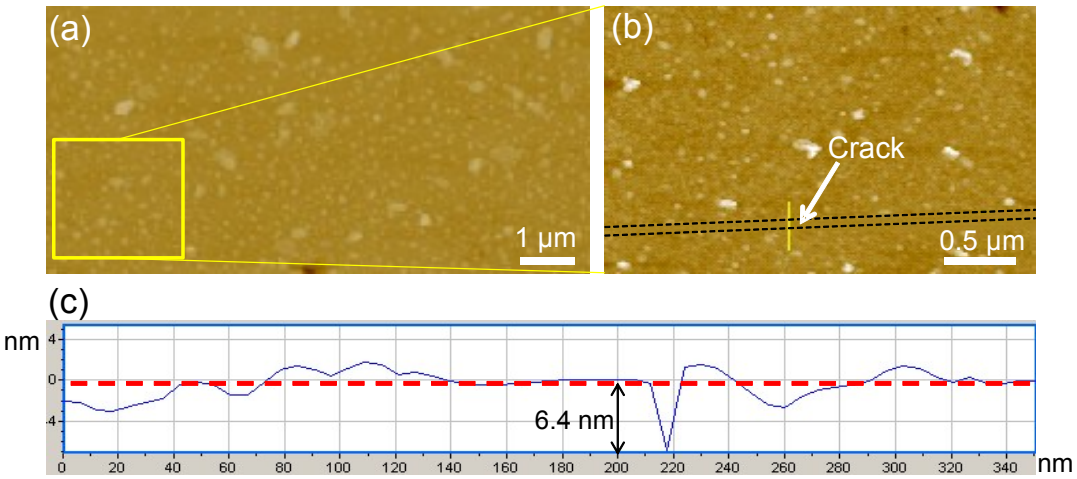


**Figure S3.** (a) AFM image of a typical sample obtained with 20% $H_2$ after sulfurization annealing. (b) Enlarged view of the marked area in (a). (c) Corresponding height profile of the yellow line marked in (b).

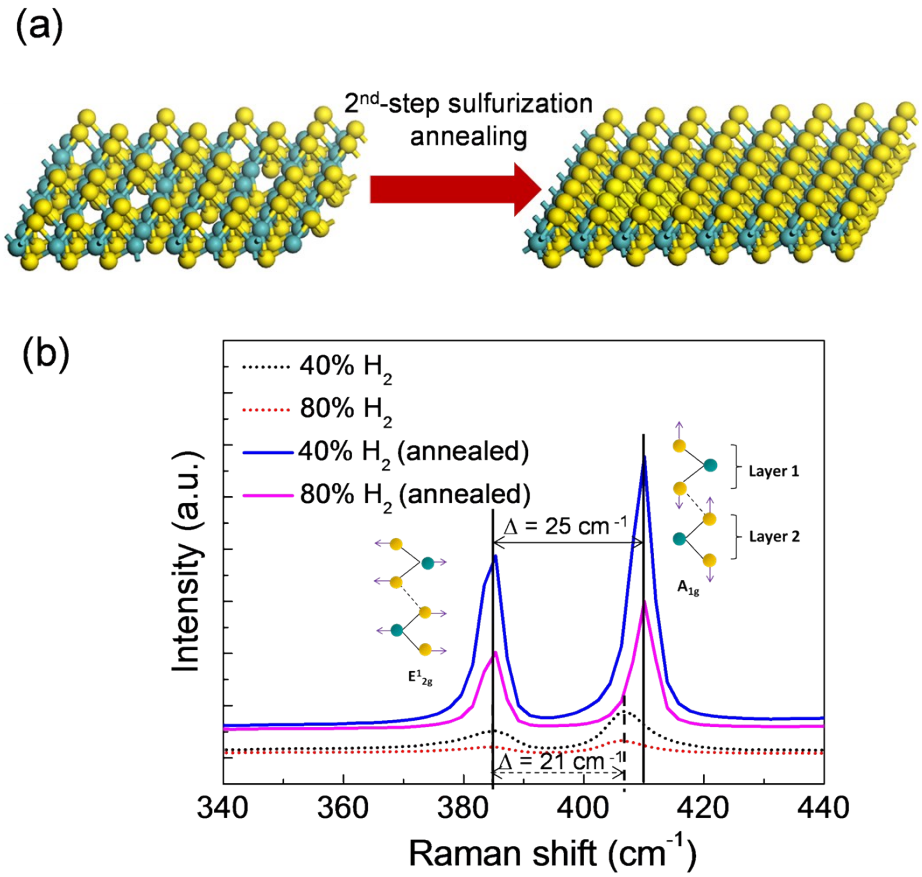


**Figure S4.** (a) Schematics of the film before and after sulfurization annealing. (b) Raman spectra of the samples obtained with 40% and 80% $H_2$ before and after sulfurization annealing. Insets show schematic illustrations of two typical Raman-active phonon modes ($E^1_{2g}$, $A_{1g}$).

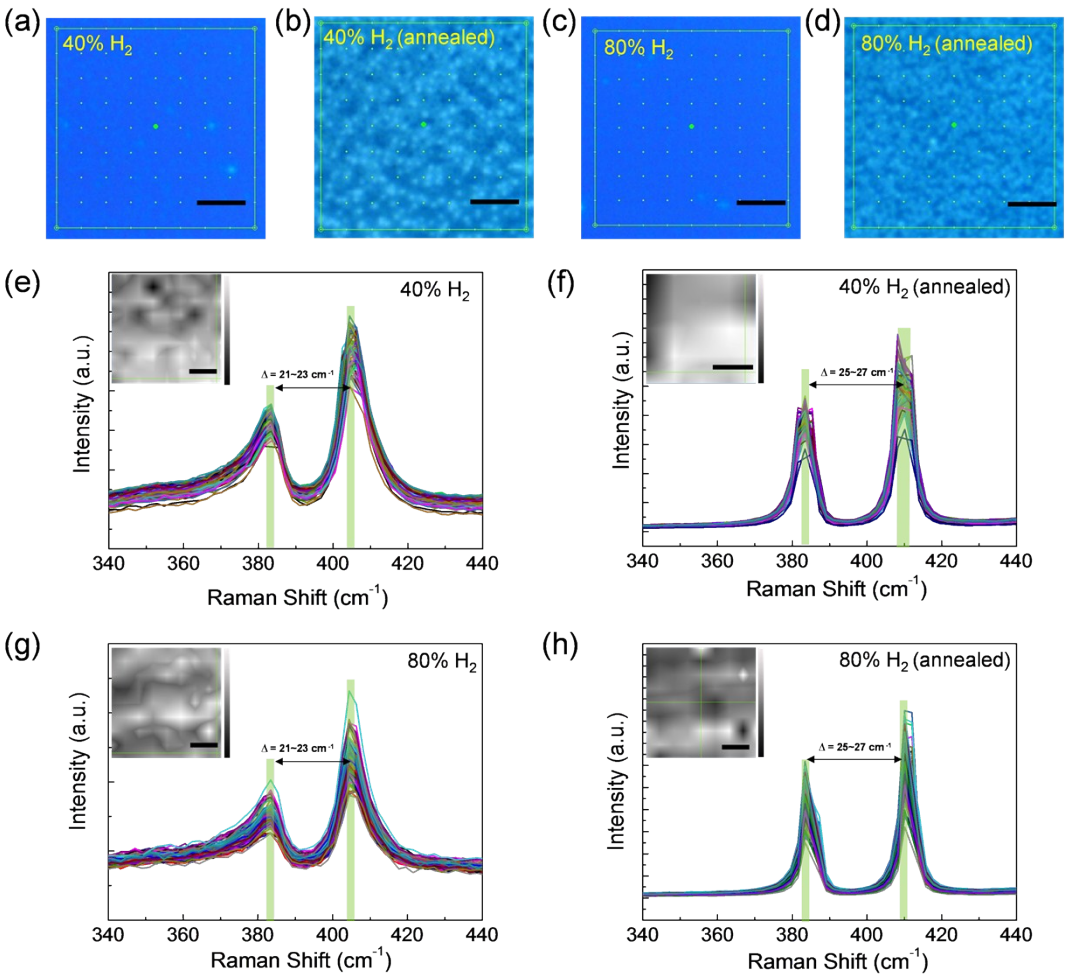


**Figure S5.** Raman mapping of 40% and 80% $H_2$ samples before and after sulfurization annealing. (a-d) Testing areas (40×40 μm$^2$). The step was set as 5 μm for both horizontal and vertical directions. Scale bars: 10 μm. (e,f) Raman mapping data of 40% $H_2$ sample. (g,h) Raman mapping data of 80% $H_2$ sample. Insets in (e-h) show corresponding Raman mapping images. Scale bars: 10 μm.

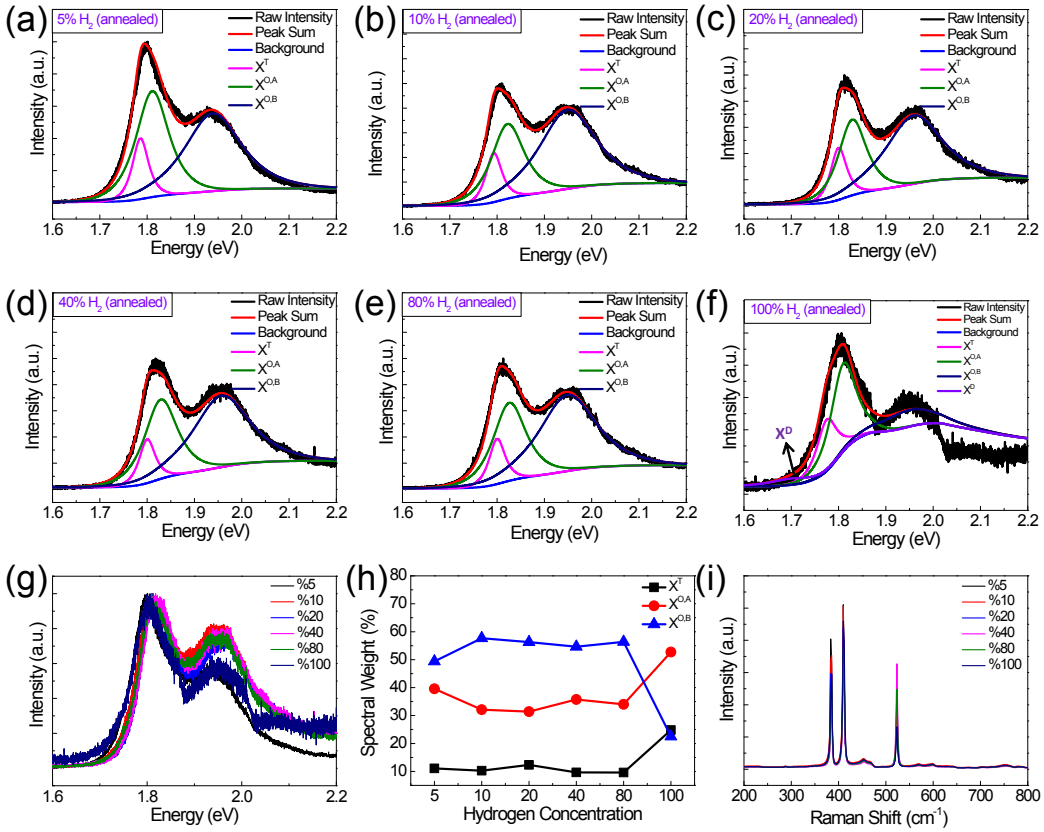


**Figure S6.** PL and Raman characterizations of 5%, 10%, 20%, 40%, 80%, 100% $H_2$ samples after sulfurization annealing. (a-f) PL curve fittings. (g) Normalized PL spectra for all samples. (h) Spectral weight evolution of charged exciton ($X^T$), A-exciton ($X^{O,A}$) and B-exciton ($X^{O,B}$) peaks at different $H_2$ contents. (i) Raman spectra of all samples.