Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information: Tuning the hydrogen evolution activity of MS_2 (M = Mo or Nb) monolayers by strain engineering

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1. Data for ΔG calculations

Table S1. Total energies for a isolated H atom E(H), a H atom in gas molecular state ($E(H_2)/2$), and their difference ΔE_{cp}. E_{ZPE}(gas) and E_{ZPE}(ads) are the zero point energies of a H atom in gas molecular state and adsorbed state, respectively. The calculated E_{ZPE}(gas) is 0.137 eV, in good agreement with the previous value of 0.135 eV.[1] $T\Delta S_H$ is obtained from the literature.[1]

	E(H)	$E(H_2)/2$	ΔE_{cp}	E_{ZPE}(gas)	E_{ZPE}(ads)	$T\Delta S_H$	$\Delta E_{ZPE} - T\Delta S_H$
1H-MoS_2					0.194		0.262
1T-MoS_2					0.228		0.296
	-0.000488	-3.380413	3.380	0.137		-0.205	
1H-NbS_2					0.229		0.297
1T-NbS_2					0.229		0.297

All the total energies calculated by VASP are with respect to the atomic energies in reference configuration (*i.e*. the configuration for which the pseudopotential is generated). Consequently, E(H) should be as small as possible. The small value of -0.000488 eV for E(H) indicates the high accuracy of the PAW potential. In addition, it is seen that E_{ZPE}(ads) is weakly dependent on different materials.

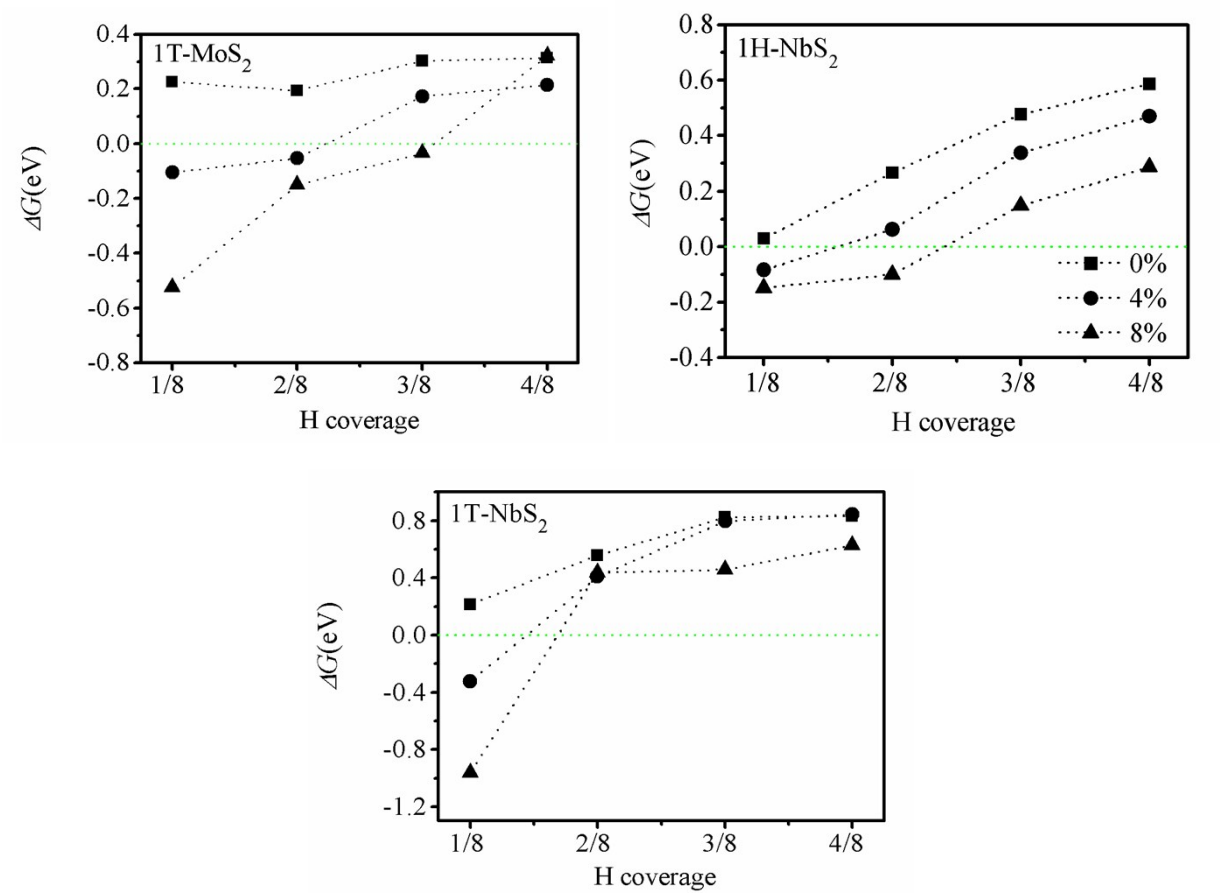

Fig. S1 (color online) H coverage dependent differential ΔG for monolayer 1T-MoS_2, 1H-NbS_2 and 1T-NbS_2 for biaxial tensile strain of 0% (squares), 4% (circles) and 8% (triangles). Only H coverage from 1/8 to 4/8 is considered.

Generally, ΔG increases with increasing H coverage for all the monolayer disulfides. For a given disulfide, the evolution of ΔG with H coverage, as indicated by dotted lines, may cross the green horizontal lines (ΔG = 0). The coverage corresponding to the crossover is selected to be the max coverage of H (MCH) defined in the text. This may lead to an average error of *ca*. ±1/16 for the MCH.

REFERENCES

1. J. K. Nørskov, T. Bligaard, A. Logadottir, J. R. Kitchin, J. G. Chen, S. Pandelov and U. Stimming, *J. Electrochem. Soc.*, 2005, **152**, J23-J26.