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

Supporting Information

Ligand Induced Structure and Property Change of 1T-MoS_2

Yaoyao Linghu,[a] Na Li,[a] Yaping Du[b*] and Chao Wu[b*]

E-mails: chaowu@xjtu.edu.cn , ypdu@nankai.edu.cn

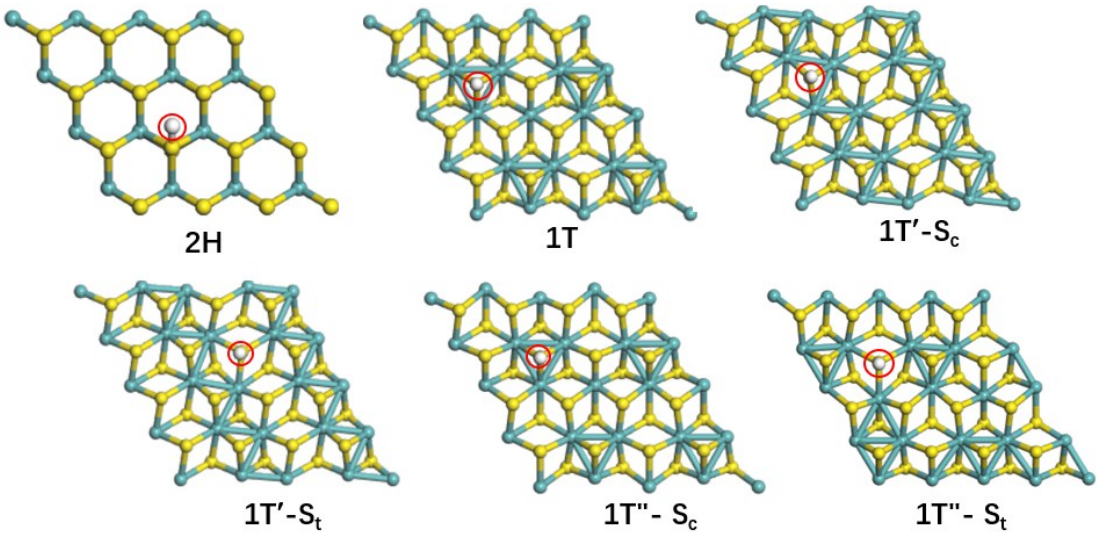

Figure S1. Optimized structures of single H atom adsorbed MoS_2 monolayers. The H atoms (white balls) are highlighted with red circles.

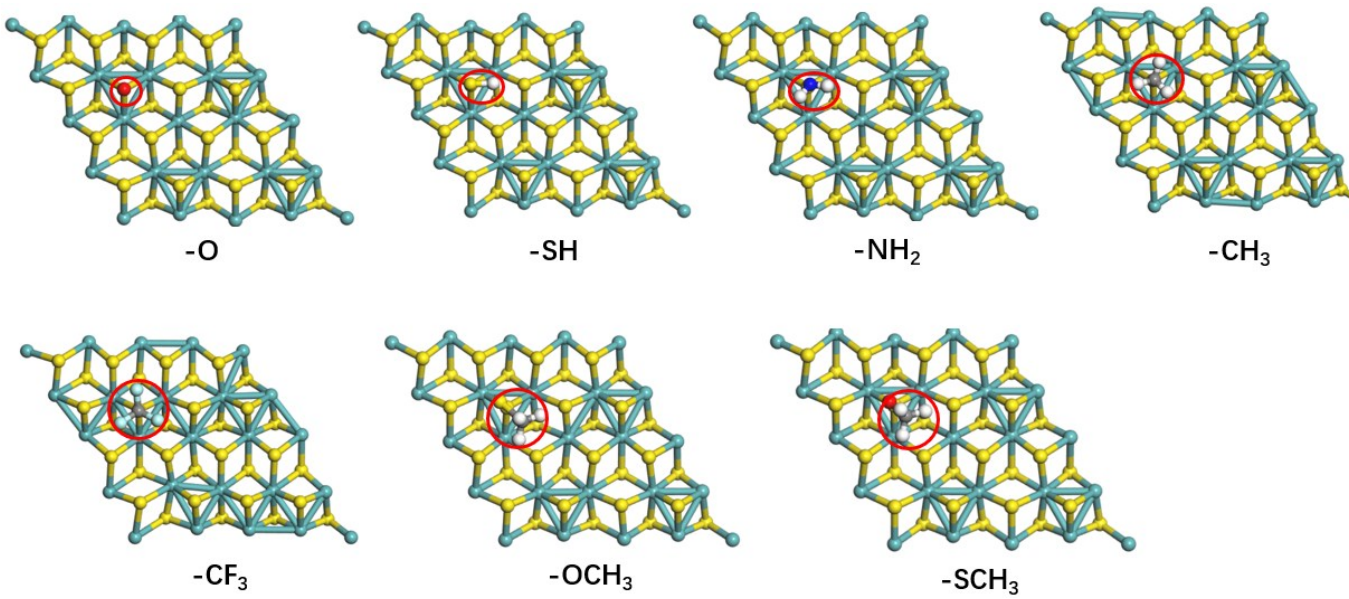

Figure S2. Optimized structures of single functional group (-O, -SH, -NH_2, -CH_3, -CF_3, OCH_3 and SCH_3) adsorbed 1T-MoS_2 monolayers. The functional groups are

highlighted with red circles.

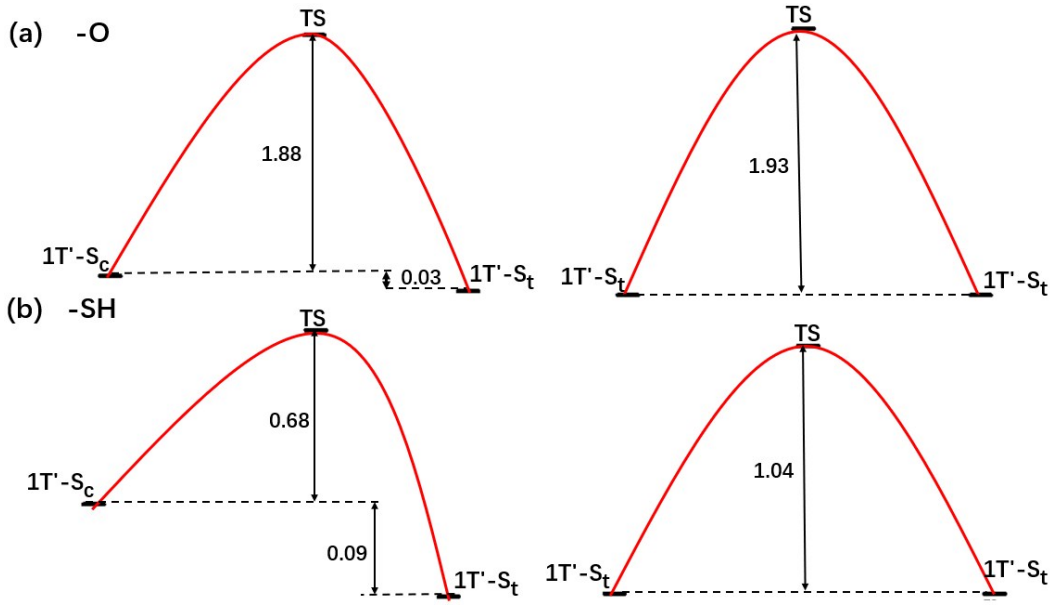

Figure S3. The functional groups (a) –O and (b) -SH diffusion energy (eV) profiles from S_c to S_t and from S_t to S_t.

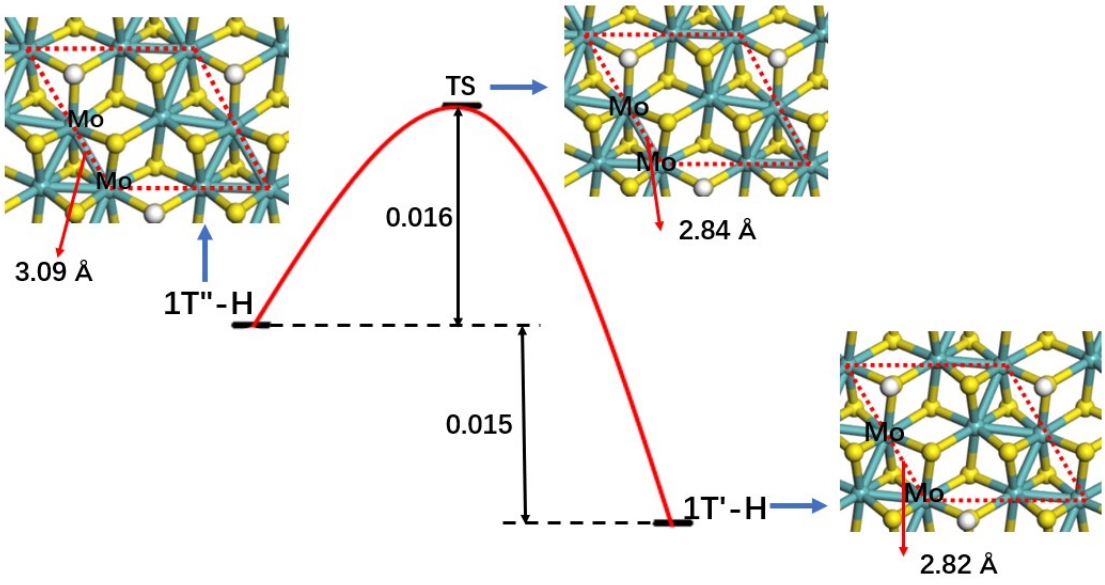

Figure S4. Minimum energy pathways of the phase transition 1T"->1T' with an adsorbed H atom. The energy barrier (eV f.u.$^{-1}$) is given. TS denotes the transition states. The computed supercell is marked by red dashed parallelograms. The Mo-Mo distance that distinguishes the 2H and 1T' phases is indicated by the red arrow.

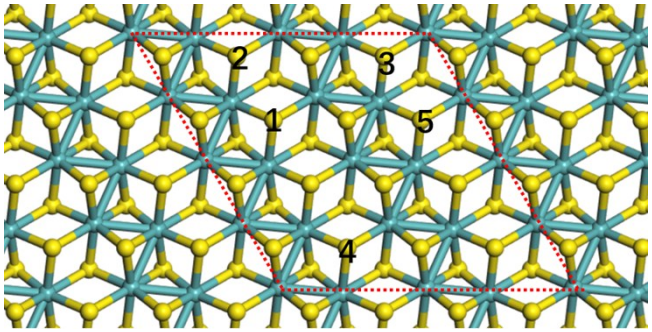

Figure S5. Configurations of two functional groups adsorbed on 1T'-MoS_2 monolayers. The number represents the adsorption site. (1,2), (1,3), (1,4) and (1,5) correspond to 1NN, 2NN, 3NN and 4NN with two adsorbed H atoms (one at site 1 and the other at sites 2, 3, 4 and 5, respectively). The computed supercell (4×4) is marked by red dashed parallelograms.

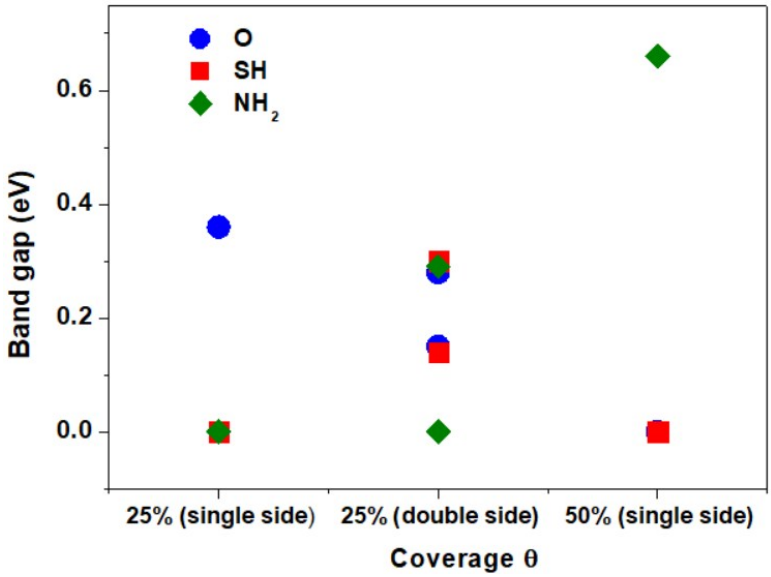

Figure S6. Calculated band gaps of the 1T'-MoS_2 monolayer adsorbed with different functional groups.

Table S1. The adsorption energy of one functional group on 1T'-MoS_2 with the PBE potential and the vdW correction (DFT-D3).

Structure	MoS_2-H	MoS_2-O	MoS_2-SH	MoS_2-NH_2	MoS_2-CH_3
E_{ad} (eV)	-0.15	-0.83	-1.86	-2.67	-2.58
E_{ad}-Vdw (eV)	-0.13	-0.98	-2.21	-3.19	-2.93

Structure	MoS_2-CF_3	MoS_2-OCH_3	MoS_2-SCH_3
E_{ad} (eV)	-1.79	-1.66	-1.50
E_{ad}-Vdw (eV)	-2.15	-2.04	-2.05

Table S2. The adsorption energy of two H atoms adsorbed on 1T'-MoS_2 with the PBE potential and the vdW correction (DFT-D3).

Structure	1NN	2NN	3NN	4NN
E_{ad} (eV)	-0.11	-0.27	-0.22	-0.13
E_{ad}-Vdw (eV)	-0.08	-0.24	-0.19	-0.11

Table S3. The adsorption energy E_{ad} and distance $d_{\text{functional- group}}$ between two functional groups adsorbed on 1T'-MoS_2.

Structure-2O	E_{ad} (eV)	$d_{O\text{-}O}$ (Å)
1NN	-1.97	3.14
2NN	-1.96	5.74
3NN	-1.96	6.35
4NN	-1.95	6.53

Structure-2SH	E_{ad} (eV)	$d_{SH\text{-}SH}$ (Å)
1NN	-4.26	3.70
2NN	-4.43	5.72
3NN	-4.30	6.23
4NN	-4.31	6.42

Structure-2NH_2	E_{ad} (eV)	$d_{NH_2-NH_2}$ (Å)
1NN	-5.66	3.19
2NN	-5.66	4.83
3NN	-5.62	6.38
4NN	-5.61	6.55

Structure-CH_3	E_{ad} (eV)	$d_{CH_3-CH_3}$ (Å)
1NN	-5.64	3.57
2NN	-5.82	5.74
3NN[1]	-5.75	6.31
3NN[2]	-5.79	6.54

Structure-$2CF_3$	E_{ad} (eV)	$d_{CF_3-CF_3}$ (Å)
1NN	-3.97	3.82
2NN	-4.29	5.73
3NN	-4.17	6.33
4NN	-4.23	6.55

Structure-$2OCH_3$	E_{ad} (eV)	$d_{OCH_3-OCH_3}$ (Å)
1NN	-3.94	3.30
2NN	-4.06	5.72
3NN	-4.01	6.39
4NN	-4.03	6.54

Structure-$2SCH_3$	E_{ad} (eV)	$d_{SCH_3-SCH_3}$ (Å)
1NN	-3.80	3.58
2NN	-4.08	5.74
3NN	-3.95	6.38
4NN	-4.04	6.55