Electronic Supplementary Material (ESI) for Materials Chemistry Frontiers.

Electronic Supplementary Information (ESI) for

Exploring correlation of pseudocapacitance with electronic structures in monolayer 1*T*-MoS_2 electrodes for supercapacitors

Zhenzhou Zhang,[a] Maokun Wu,[a] Lijing Wang,[a] Jin Wang,[a] Yahui Cheng,[a] Luyan Li,[b] Hong Dong,[a] Hui Liu,[a] Zhanglian Hong,[c] Kyeongjae Cho,[a,d] Feng Lu,[a] Weichao Wang,[*,a] Wei-Hua Wang[*,a]

[a] Department of Electronic Science and Engineering, and Tianjin Key Laboratory of Photo-Electronic Thin Film Device and Technology, Nankai University, Tianjin, 300071, P. R. China

[b] School of Science, Shandong Jianzhu University, Jinan 250101, P. R. China

[c] State Key Laboratory of Silicon Materials, School of Materials Science and Engineering, Zhejiang University, Hangzhou, 310027, P. R. China

[d] Department of Material Science and Engineering, the University of Texas at Dallas, Richardson, 75080, USA

*Emails: whwangnk@nankai.edu.cn; weichaowang@nankai.edu.cn

Table S1 Possible adsorption configurations and adsorption energies of H^+ ions on one side and two sides of monolayer 1*T*-MoS_2

Number of H^+ ions on one side	Coverage	Sites	E_{ad} (eV)	Number of H^+ ions on both sides	Coverage	$\boldsymbol{E}_{ad}$ (eV)
1	11.1%	1	-4.13	1	11.1%	-2.89
2	22.2%	1, 5	-2.74	2	22.2%	-2.14
		1, 2	-2.89			
3	33.3%	2, 4, 8	-2.25	3	33.3%	-1.83
		2, 4, 9	-2.32			
		1, 2, 3	-2.44			
4	44.4%	2, 4, 8, 9	-1.93	4	44.4%	-1.72
		2, 4, 7, 9	-2.01			
		1, 2, 4, 9	-2.05			
		1, 2, 3, 4	-2.08			
5	55.6%	1, 3, 5, 6, 7	-1.82	5	55.6%	-1.64
		1, 3, 5, 6, 8	-1.79			
		3, 5, 6, 7, 8	-1.88			
		5, 6, 7, 8, 9	-1.83			
6	66.7%	4, 5, 6, 7, 8, 9	-1.66	6	66.7%	-1.55
		1, 3, 5, 6, 7, 8	-1.76			
		1, 3, 5, 6, 7, 9	-1.70			
7	77.8%	1,2, 3, 5, 6, 7, 8	-1.49	7	77.8%	-1.48
		1, 3, 5, 6, 7, 8, 9	-1.63			
8	88.9%	1, 2, 3, 4, 5, 6, 7, 8	-1.53	8	88.9%	-1.37
9	100%	1, 2, 3, 4, 5, 6, 7, 8, 9	-1.19	9	100%	-1.25

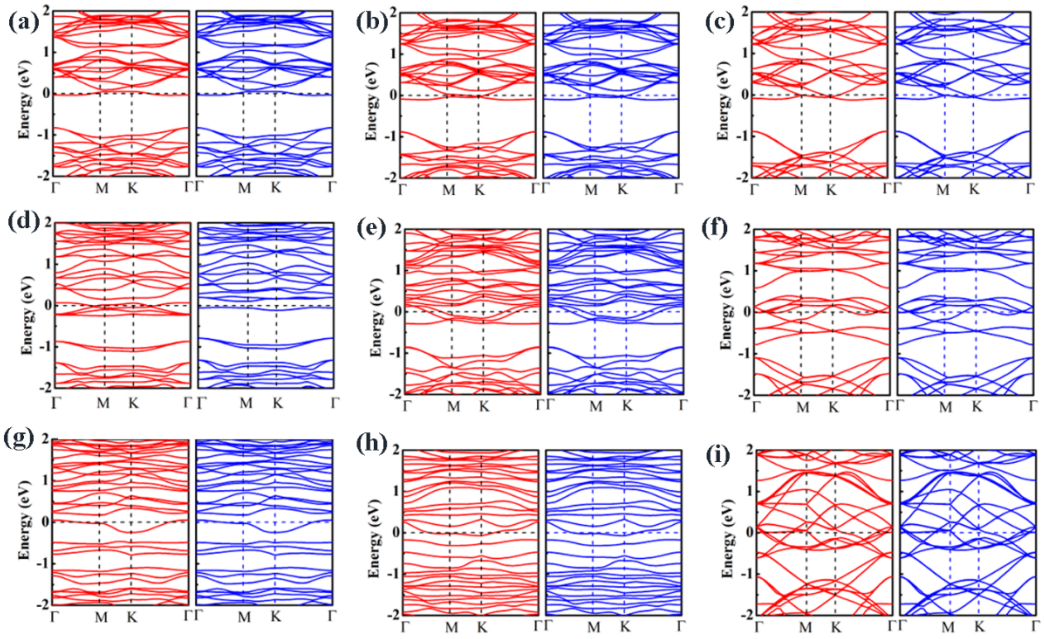

Fig. S1 Band structures of 1*T*-MoS_2 under different adsorption coverage of H^+ ions on one side of 1*T*-MoS_2. The coverage is 11.1% in (a), 22.2% in (b), 33.3% in (c), 44.4% in (d) 55.6 % in (e) 66.7 % in (f) 77.8% in (g), 88.9% in (h), 100 % in (i). The Fermi level is set to be 0.0 eV.

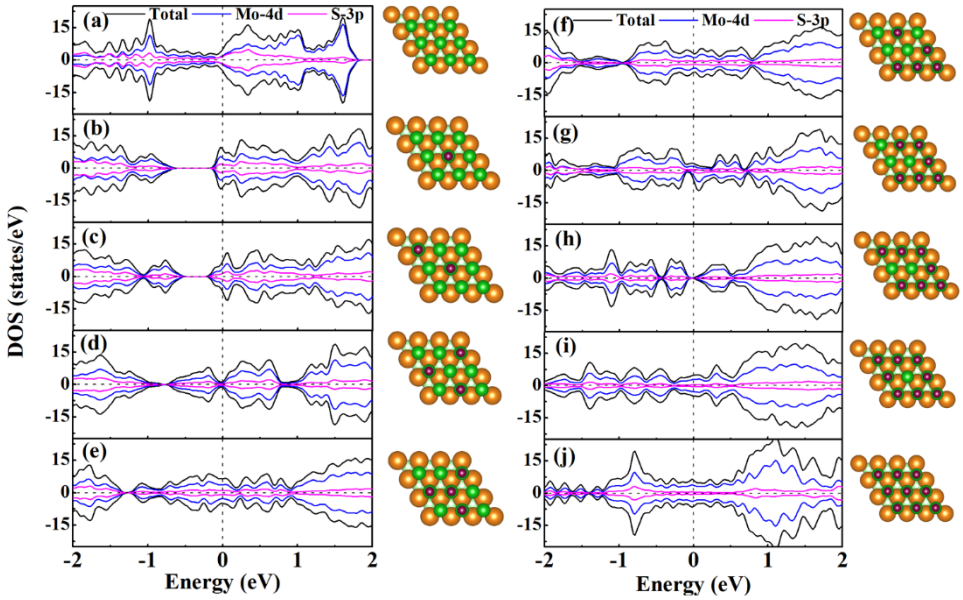

Fig. S2 Total DOS, PDOS and geometric structures of monolayer 1*T*-MoS_2 under different adsorption coverage of H^+ ions on both sides of 1*T*-MoS_2. Pristine 1*T*-MoS_2 in (a), and the coverage is 11.1% in (b), 22.2% in (c), 33.3% in (d), 44.4% in (e) 55.6 % in (f) 66.7 % in (g) 77.8% in (h), 88.9% in (i), 100 % in (j). The Fermi level is set to be 0.0 eV.

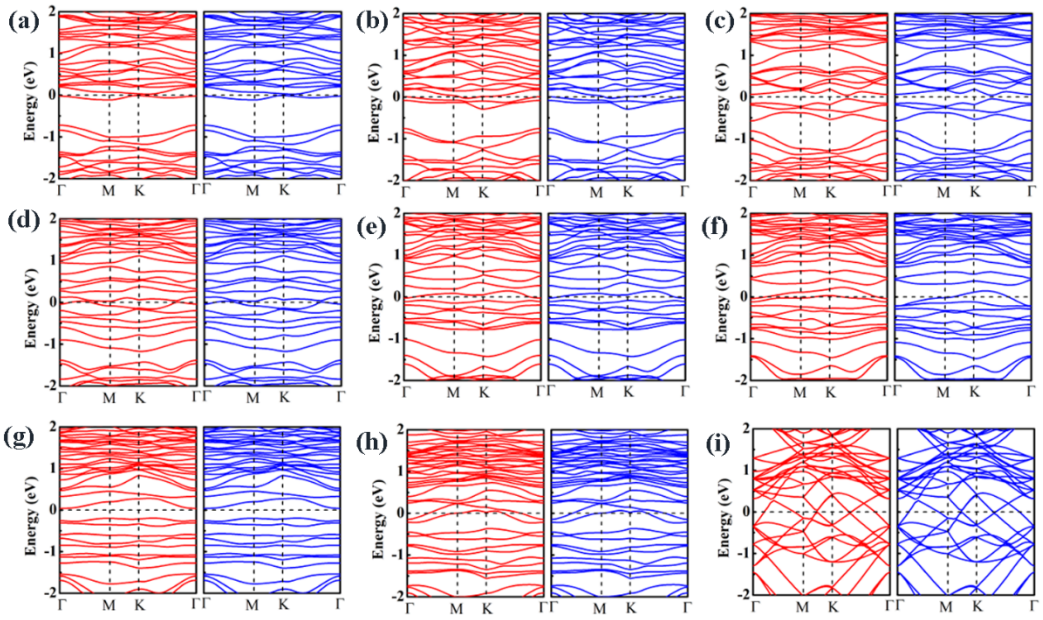

Fig. S3 Band structures of 1*T*-MoS_2 under different adsorption coverage of H^+ ions on both sides of 1*T*-MoS_2. The coverage is 11.1% in (a), 22.2% in (b), 33.3% in (c), 44.4% in (d) 55.6 % in (e) 66.7 % in (f) 77.8% in (g), 88.9% in (h), 100 % in (i). The Fermi level is set to be 0.0 eV.

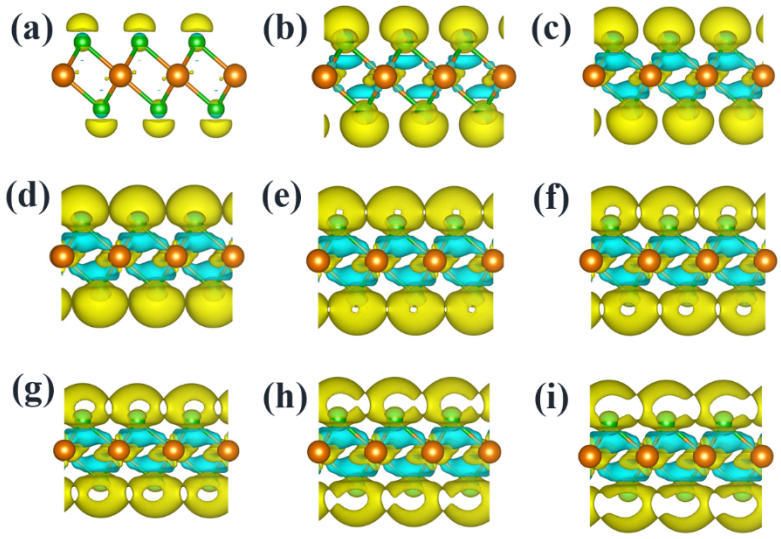

Fig. S4 Charge density difference distribution of monolayer 1*T*-MoS_2 in charging state with the additional electrons of 0.92 e in (a), 1.88 e in (b), 2.91 e in (c), 3.93 e in (d), 4.96 e in (e), 5.91 e in (f), 7.08 e in (g), 7.92 e in (h), 9.45 e in (i). The yellow and cyan isosurface with an isovalue of 0.005 e/$Å^3$ correspond to charge accumulation and depletion respectively.

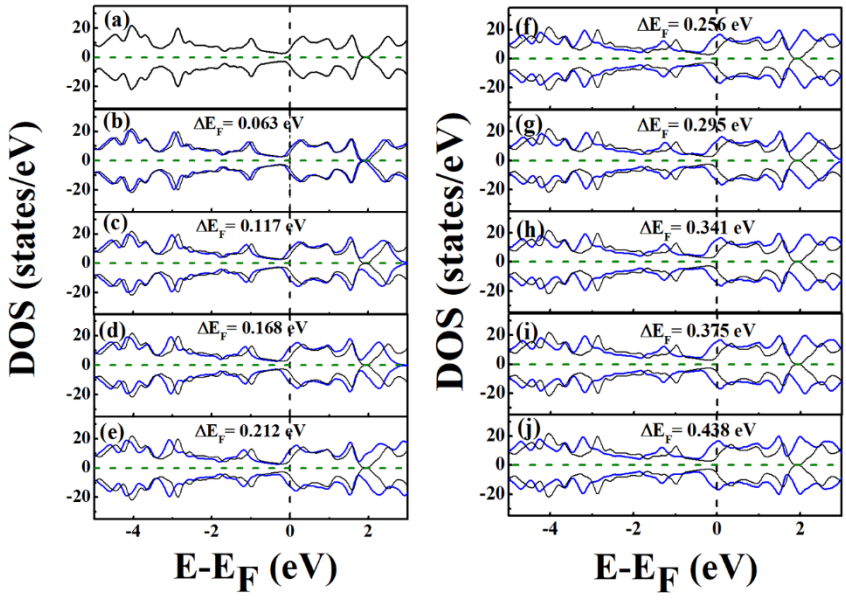

Fig. S5 Total DOS of monolayer 1*T*-MoS_2 in charging states. ΔE_F is Fermi level shift. Pristine monolayer 1*T*-MoS_2 without additional electrons in (a), and with 0.92 e in (b), 1.88 e in (c), 2.91 e in (d), 3.93 e in (e), 4.96 e in (f), 5.91 e in (g), 7.08 e in (h), 7.92 e in (i), 9.45 e in (j). The Fermi level is set to be 0.0 eV.

Table S2 Number of H^+ ions on one side (N_{H^+}), Fermi level shift (ΔE_F), work function of monolayer 1*T*-MoS_2 adsorbed H^+ ions at neutral state (WF_{MoS_2-nH}) and monolayer 1*T*-MoS_2 ($WF_{MoS_2{}^{\Delta Q-}}$) at negatively charged state, voltage change ($|\Delta V|$), charge transfer (ΔQ) and pseudocapacitance (C_{redox})

N_{H^+}	ΔE_F (eV)	$WF_{MoS_2{}^{\Delta Q-}}$ (eV)	WF_{MoS_2-nH} (eV)	$\|\Delta V\|$ (V)	ΔQ (e)	C_{redox} (μF/cm^2)	C_{redox} (F/g)
1	0.063	5.037	4.58	0.457	0.92	40.87	134.74
2	0.117	4.983	4.06	0.923	1.88	41.34	136.33
3	0.168	4.932	3.62	1.312	2.91	45.03	148.45
4	0.212	4.888	3.29	1.598	3.93	49.92	164.60
5	0.256	4.844	3.12	1.724	4.96	58.40	192.56
6	0.295	4.805	2.86	1.945	5.91	61.68	203.37
7	0.341	4.759	2.64	2.119	7.08	67.83	223.63
8	0.375	4.725	2.39	2.335	7.92	68.85	227.02
9	0.438	4.662	2.16	2.502	9.45	76.67	252.81

Table S3 Number of H^+ ions on both sides (N_{H^+}), Fermi level shift (ΔE_F), work function of monolayer 1*T*-MoS_2 adsorbed H^+ ions at neutral state (WF_{MoS_2-nH}) and monolayer 1*T*-MoS_2 ($WF_{MoS_2{}^{\Delta Q-}}$) at negatively charged state, voltage change ($|\Delta V|$), charge transfer (ΔQ) and pseudocapacitance (C_{redox})

N_{H^+}	ΔE_F (eV)	$WF_{MoS_2{}^{\Delta Q-}}$ (eV)	WF_{MoS_2-nH} (eV)	$\|\Delta V\|$ (V)	ΔQ (e)	C_{redox} (μF/cm^2)	C_{redox} (F/g)
2	0.118	4.982	4.535	0.447	1.90	86.29	284.51
4	0.209	4.891	4.195	0.696	3.82	111.42	367.37
6	0.285	4.851	4.070	0.781	5.68	147.64	486.79
8	0.370	4.730	3.755	0.975	7.81	162.61	536.16
10	0.452	4.648	3.485	1.163	9.79	170.88	563.44
12	0.543	4.557	3.210	1.347	11.90	179.34	591.32
14	0.639	4.461	2.915	1.546	13.92	182.53	602.67
16	0.741	4.359	2.665	1.694	16.15	193.53	638.13
18	0.896	4.204	2.435	1.769	18.62	213.67	704.53

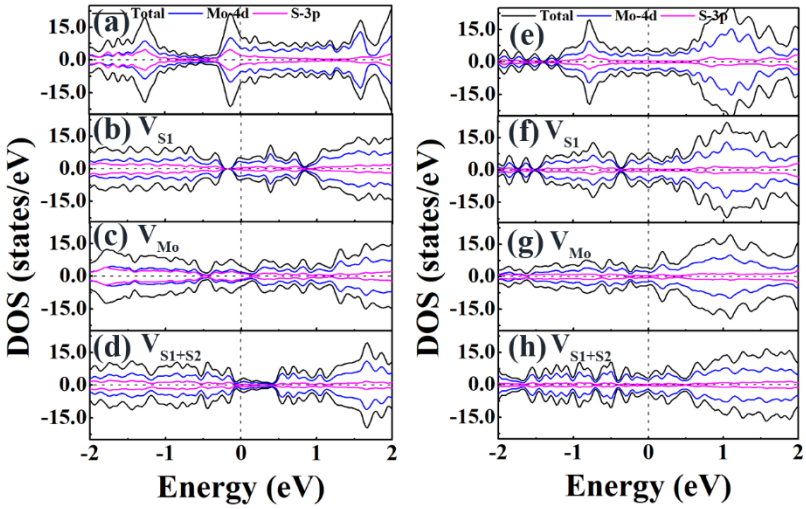

Fig. S6 Total DOS and PDOS of 1*T*-MoS_2 with one side adsorption of H^+ ions in (a)-(d) and two sides adsorption of H^+ ions in (e)-(h). Pristine 1*T*-MoS_2-9H and 1*T*-MoS_2-18H in (a) and (e), V_{S1} in (b) and (f), V_{Mo} in (c) and (g), V_{S1+S2} in (d) and (h). The Fermi level is set to be 0.0 eV.

Table S4. Configurations of fully coverage with H^+ ions under different intrinsic defects ($NH^+ - \text{defect}$), Fermi level shift (ΔE_F), work function of defect monolayer 1T-MoS_2 adsorbed H^+ ions at neutral state (WF_{MoS_2-nH}), work function of defect monolayer 1T-MoS_2 at negatively charged state ($WF_{MoS_2{}^{\Delta Q-}}$), voltage change ($|\Delta V|$), charge transfer (ΔQ) and pseudocapacitance (C_{redox}).

$NH^+ - \text{defect}$	ΔE_F (eV)	$WF_{MoS_2{}^{\Delta Q-}}$ (eV)	WF_{MoS_2-nH} (eV)	$\lvert\Delta V\rvert$ (V)	ΔQ (e)	C_{redox} (μF/cm^2)	C_{redox} (F/g)
$9H^+$-V_{Mo}	0.72	4.83	2.12	2.71	9.27	69.44	228.96
$9H^+$-V_{S1}	0.91	4.35	2.47	1.88	9.46	101.93	336.09
$9H^+$ V_{S1+S2}	0.78	4.30	2.64	1.66	9.42	115.20	379.83
$18H^+$-V_{Mo}	1.25	4.3	2.17	2.13	18.40	83.34	252.81
$18H^+$-V_{S1}	1.36	3.90	2.86/3.12	1.04/0.78	9.54/9.16	241.49/186.21	1410.24
$18H^+$-V_{S1+S2}	1.36	3.72	2.84	0.88	18.92	436.45	1439.08