Supporting Information (SI)

Tuning the Binding Energy of Excitons in Monolayer MoS_2 by Molecular Functionalization and Defective Engineering

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Fig. S1: The G_0W_0 band edges of pristine MoS_2 with 55 eV G_0W_0 self-energy as a function of vacuum thickness. Black dashed lines indicate the band edges of the MoS_2 with 30×30×1 k-point sampling and 15 Å vacuum thickness; CBM and VBM indicate the conduction band minimum and valence band maximum, respectively.

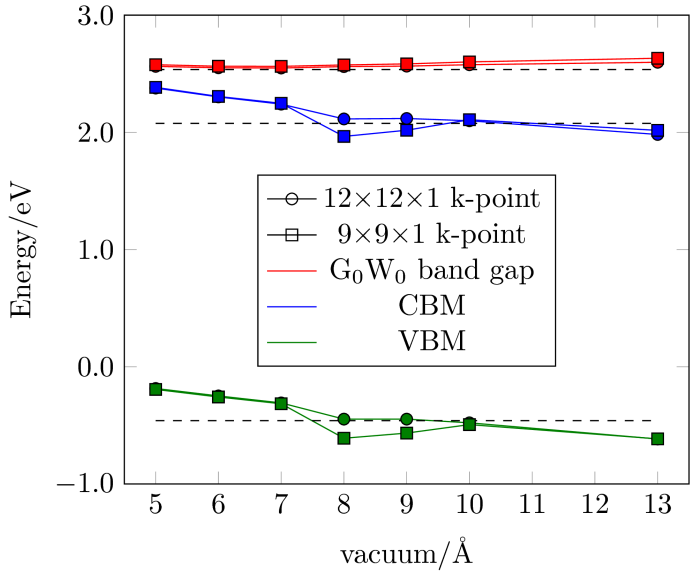
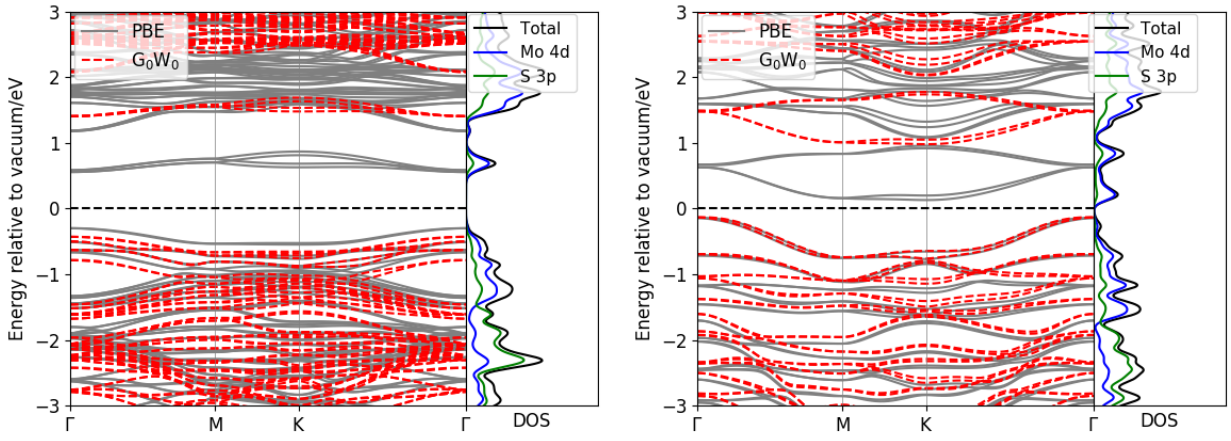


Table S1: The G_0W_0 band gaps of pristine MoS_2 with 55 eV G_0W_0 self-energy, 10 Å vacuum and 12×12×1 k-point sampling as a function of plane wave (PW) cutoff energy. CBM and VBM indicate the conduction band minimum and valence band maximum, respectively. Energies are in eV.

	PW=500	PW=600	PW=700	PW=800
CBM	3.323	3.322	3.321	3.321
VBM	0.746	0.745	0.745	0.744
G_0W_0 band gap	2.577	2.577	2.576	2.577

Fig. S2: Electronic band structures and density of states for defective MoS_2.



(a) 11% S defect coverage

(b) 25% S defect coverage

Fig. S3: The most favorable configurations for (a) NO and (b) $C_3H_3N_3$ molecules on pristine MoS_2 with 25% molecular coverage.

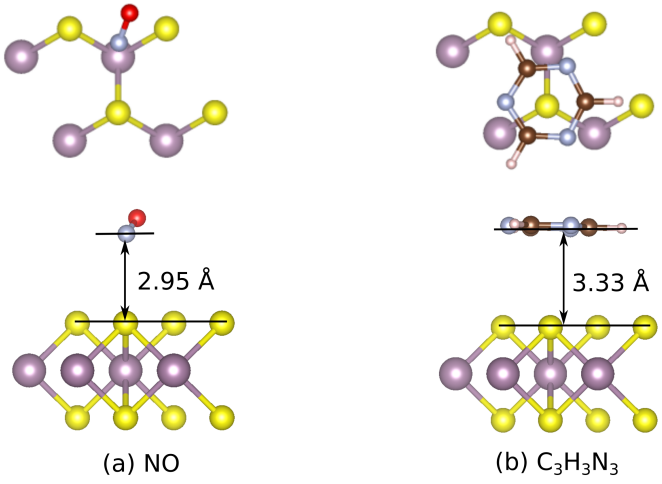


Fig. S4: The most favorable configurations for (a) NO and (b) $C_3H_3N_3$ molecules on defective MoS_2 with 25% molecular and defective coverage.

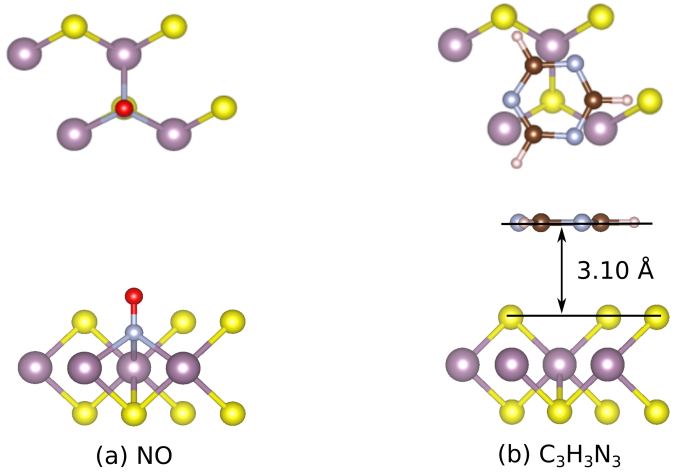
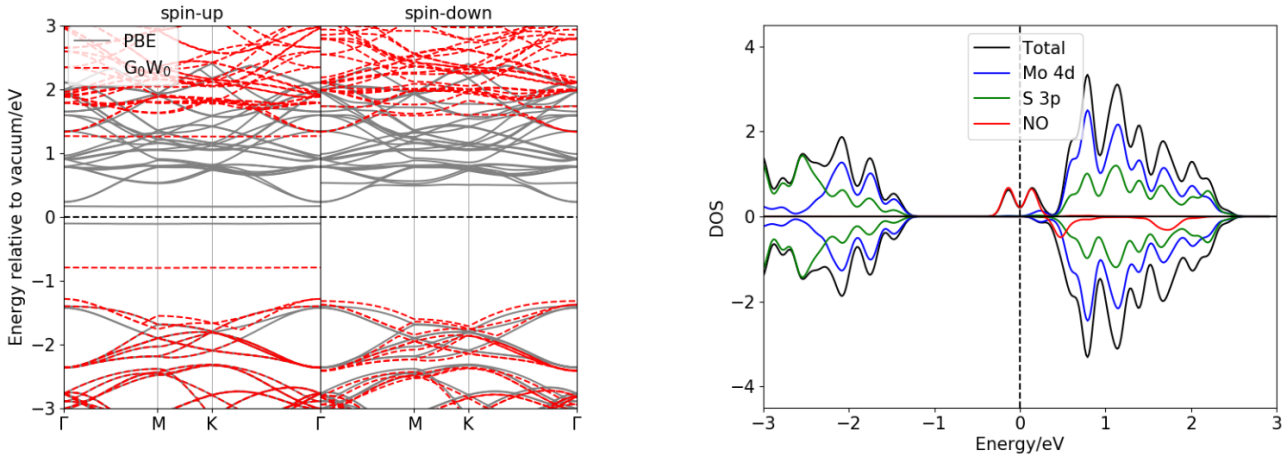
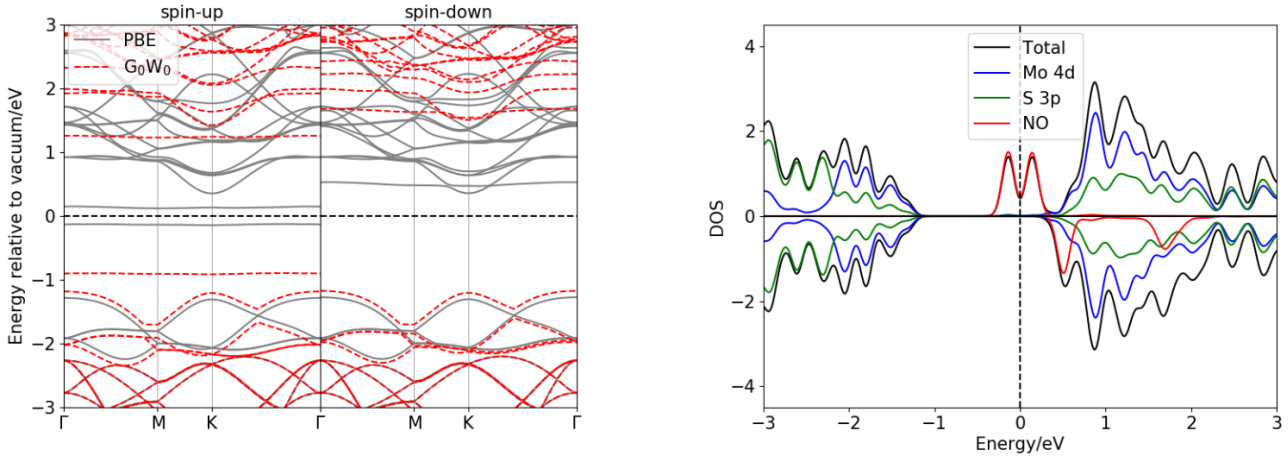


Fig. S5: Electronic band structures and density of states for the NO molecule adsorbed on pristine MoS_2 with (a) 11% and (b) 25% molecular coverage.

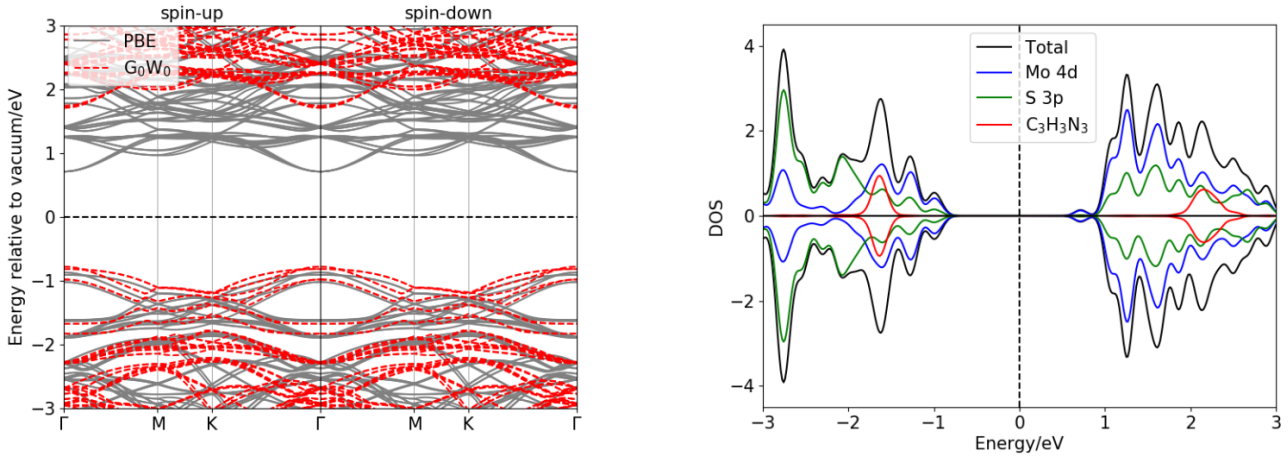


(a) 11% NO coverage

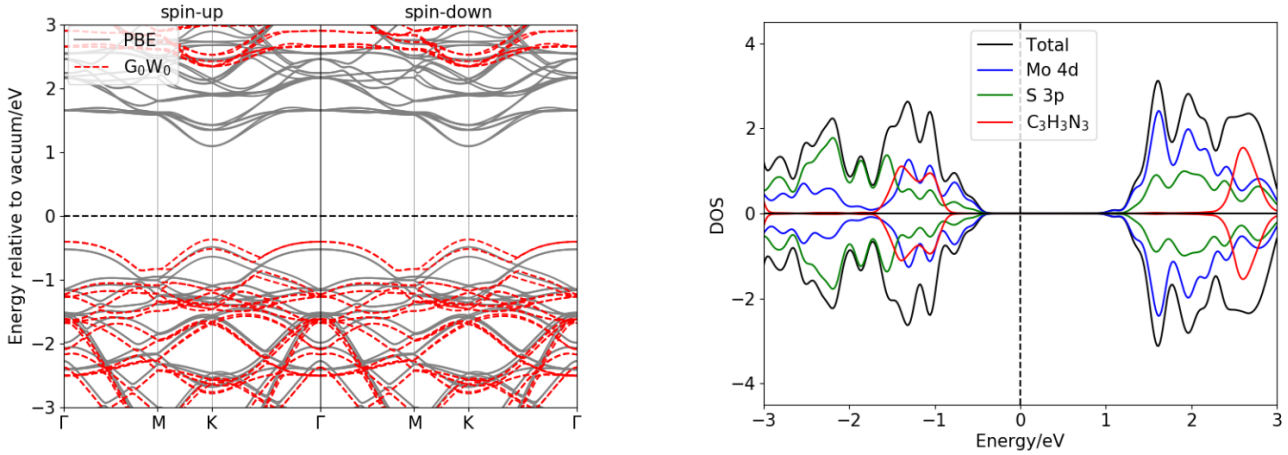


(b) 25% NO coverage

Fig. S6: Electronic band structures and density of states for the $C_3H_3N_3$ molecule on pristine MoS_2 with (a) 11% and (b) 25% molecular coverage.



(a) 11% $C_3H_3N_3$ coverage

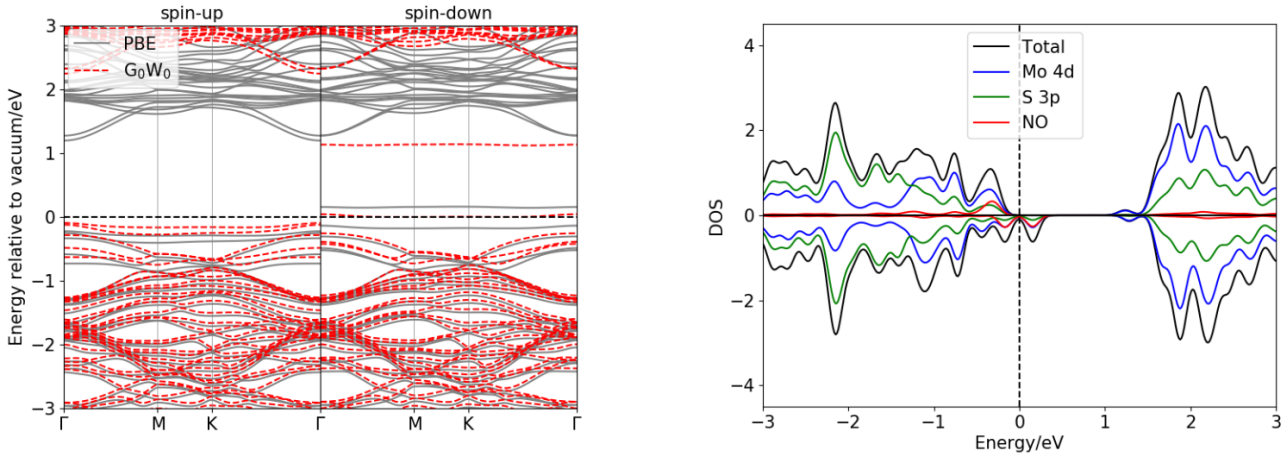


(b) 25% $C_3H_3N_3$ coverage

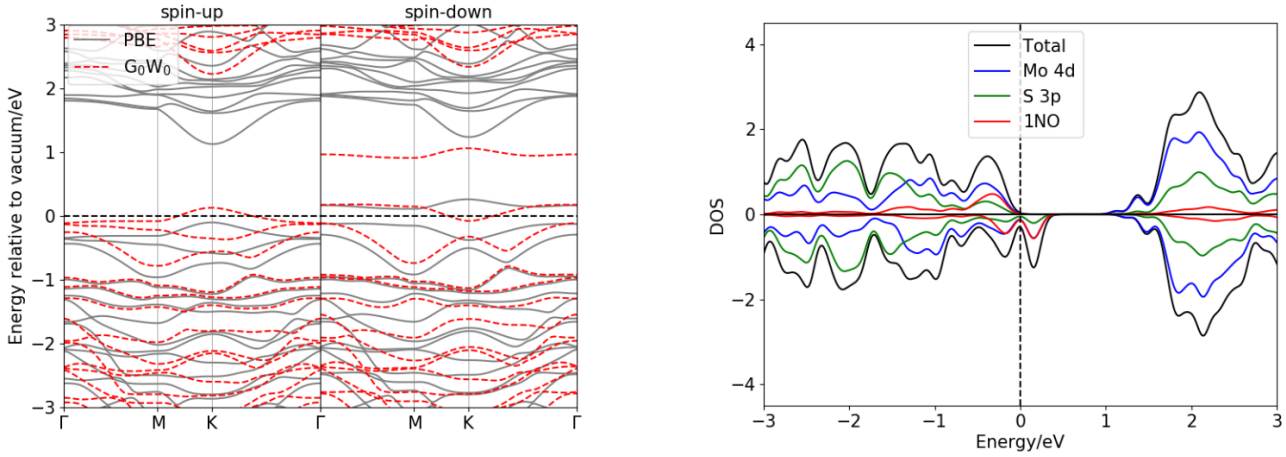
Table S2: The DFT band gaps with PBE functional for NO and $C_3H_3N_3$ molecules adsorbed on pristine MoS_2 with 11% and 25% molecular coverage. Energies are in eV.

	free	NO		$C_3H_3N_3$	
		11%	25%	11%	25%
DFT band gap	1.58	0.27(↑)/1.63(↓)	0.28(↑)/1.66(↓)	1.58	1.58

Fig. S7: Electronic band structures and density of states for the NO molecule on defective MoS_2 with (a) 11% and (b) 25% molecular and defective coverage.

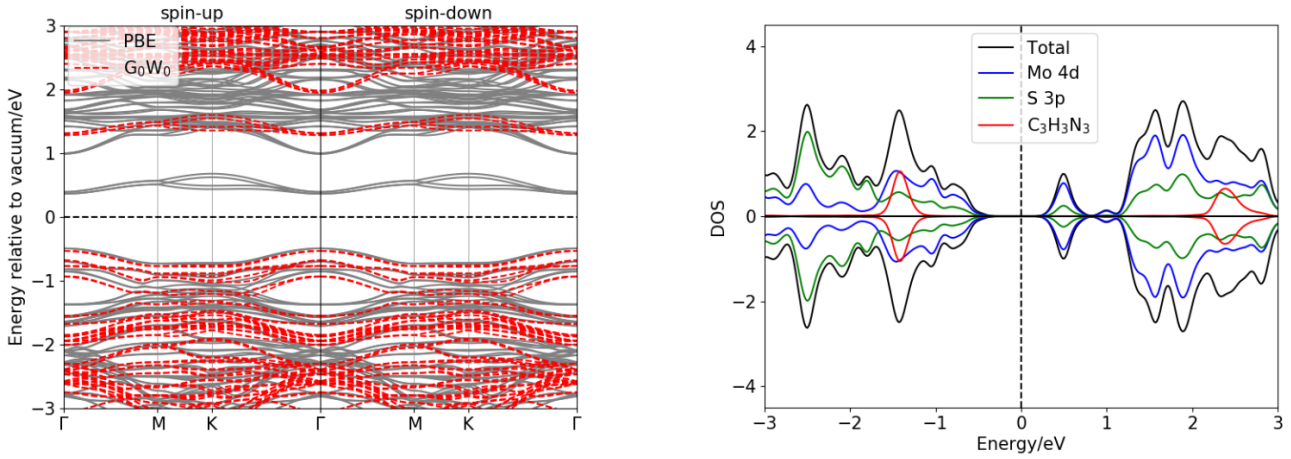


(a) 11% NO and S defect coverage

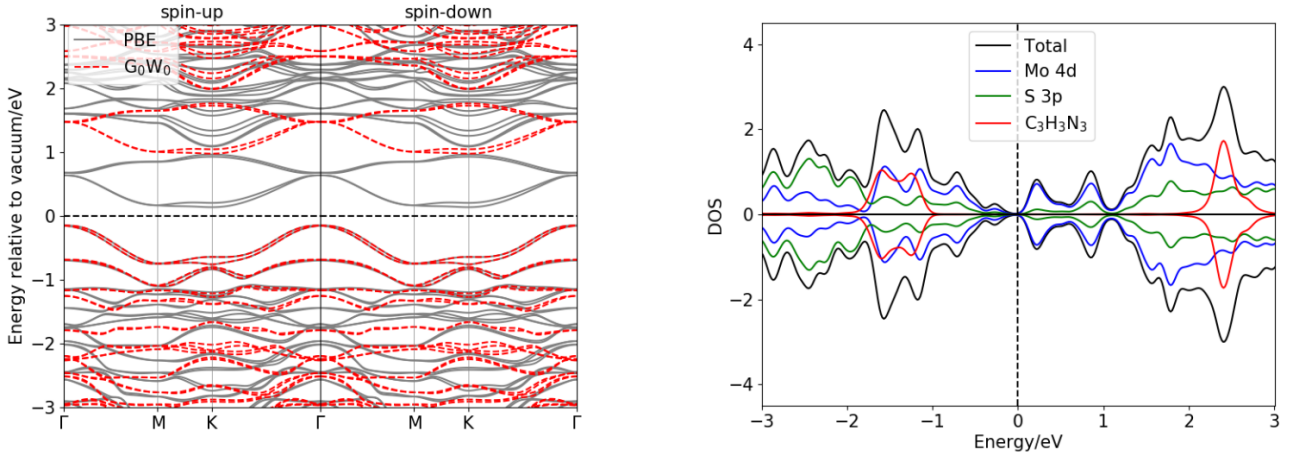


(b) 25% NO and S defect coverage

Fig. S8: Electronic band structures and density of states for the $C_3H_3N_3$ molecule on defective MoS_2 with (a) 11% and (b) 25% molecular and defective coverage.



(a) 11% $C_3H_3N_3$ and S defect coverage



(b) 25% $C_3H_3N_3$ and S defect coverage

Table S3: The DFT band gaps with PBE functional for NO and $C_3H_3N_3$ molecules adsorbed on defective MoS_2 monolayer with 11% and 25% molecular and defective coverage. Energies are in eV.

	S-defect (no molecule)		NO		$C_3H_3N_3$	
	11%	25%	11%	25%	11%	25%
DFT band gap	0.86	0.28	1.42(↑)/0.29(↓)	1.23(↑)/0.29(↓)	0.86	0.29