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Supplementary Information

Origin of the Enhanced Edge Optical Transition in Transition Metal Dichalcogenide Flakes

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Keywords: edge state, optical transition, transition metal dichalcogenides, density functional theory

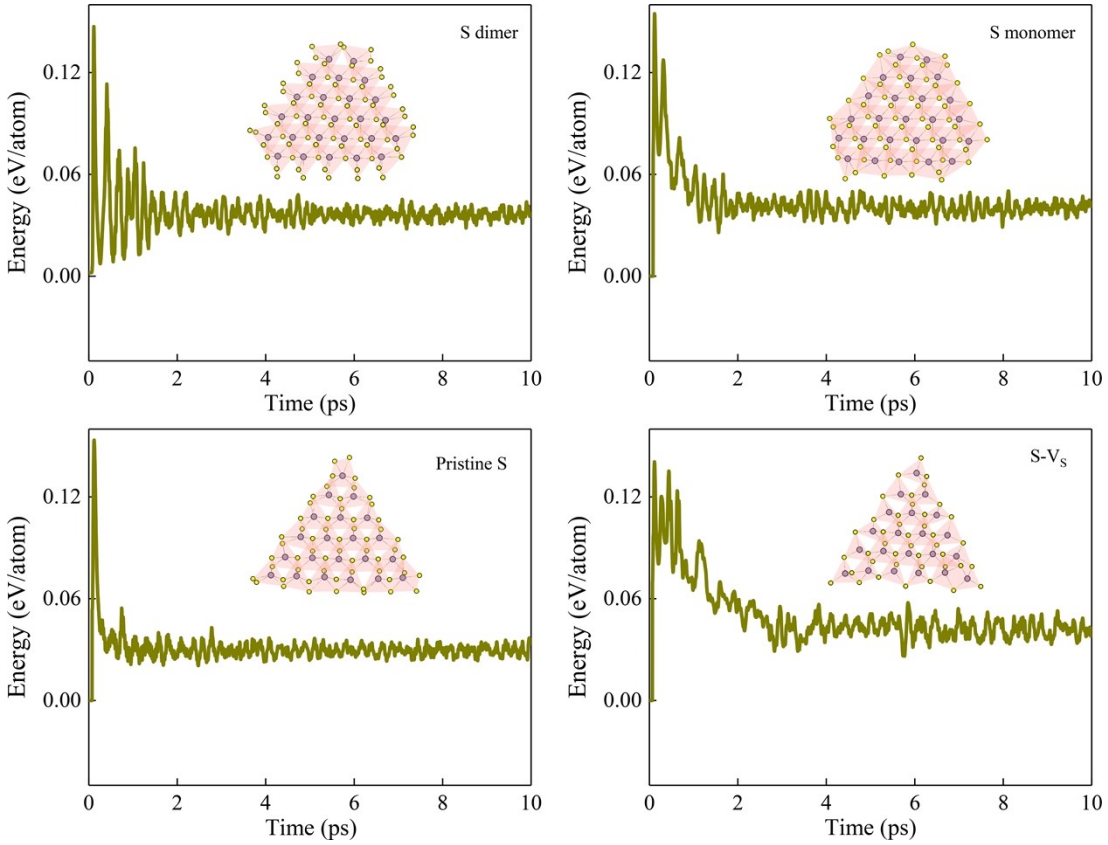

Fig. S1 AIMD global optimization (0-10 ps) for the triangular MoS_2 flakes with S dimer/monomer, pristine S or S-V_S edge.

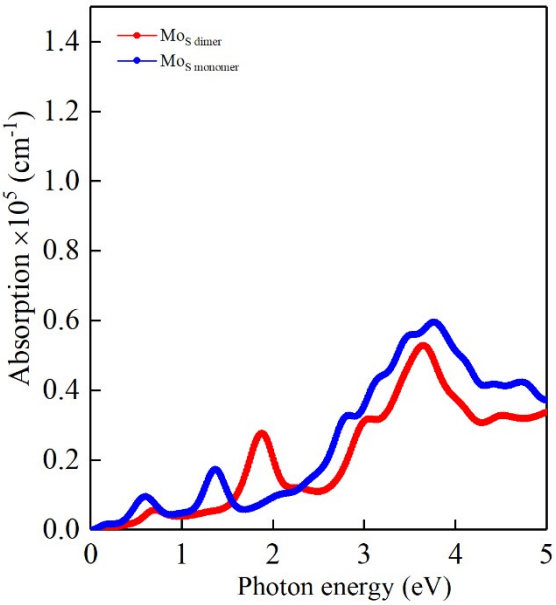

Fig. S2 Absorption spectrum for truncated triangular (hexagonal) MoS_2 flake with S dimer/monomer at Mo edge ($Mo_{S\ dimer}$/$Mo_{S\ monomer}$ in **Figure** 1c).

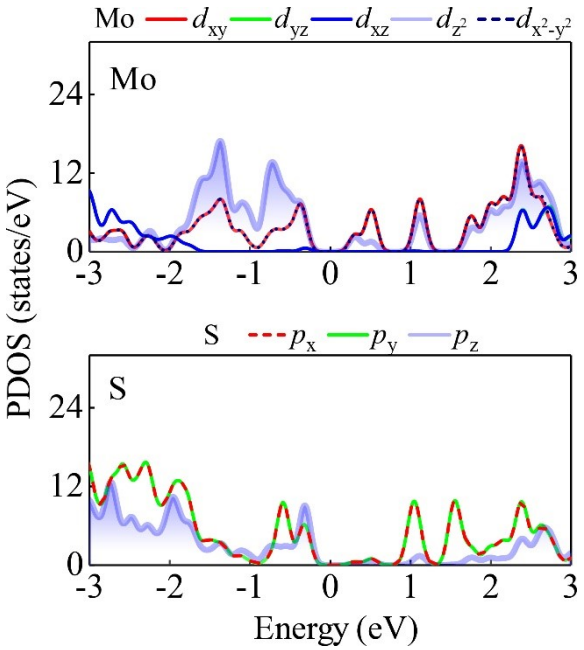

Fig. S3 PDOS for the *l*=5 triangular MoS_2 flake with S dimer.

Table S1. The TDM intensity for the strong transition $\psi_{VB}^{281,282,283} \rightarrow \psi_{CB}^{295,296,297}$ in triangular MoS_2 flake with S dimer edge.

TDM (debye^2)	ψ_{VB}^{281}	ψ_{VB}^{282}	ψ_{VB}^{283}
ψ_{CB}^{295}	102.503	59.551	0.001
ψ_{CB}^{296}	38.465	27.876	98.194
ψ_{CB}^{297}	27.827	38.218	76.227

Table S2. The TDM intensity for the weak transition $\psi_{VB}^{286,287,288} \rightarrow \psi_{CB}^{292,293,294}$ in triangular MoS_2 flake with S dimer edge.

TDM (debye^2)	ψ_{VB}^{286}	ψ_{VB}^{287}	ψ_{VB}^{288}
ψ_{CB}^{292}	40.121	32.498	0
ψ_{CB}^{293}	6.145	7.516	28.943
ψ_{CB}^{294}	7.523	6.13	29.776

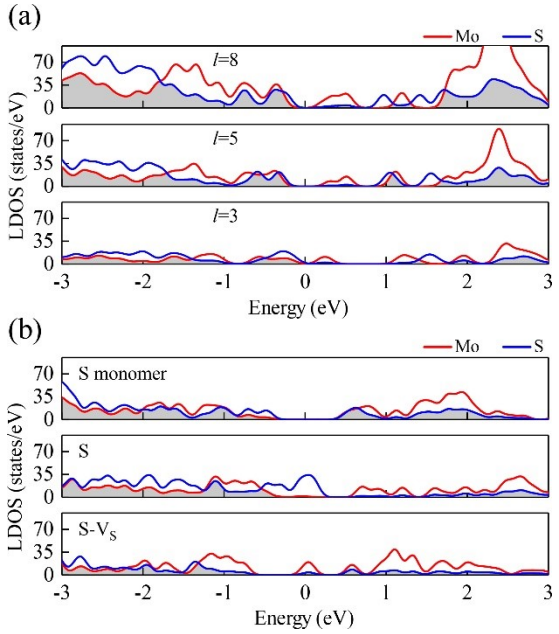

Fig. S4 (a) Overlapped *p*-*d* orbital of LDOS for the triangular MoS_2 flake with S dimer as a function of the size *l*. (b) LDOS for *l*=5 triangular MoS_2 flake with other edges (S monomer, pristine S and reconstructed S-V_S configurations).

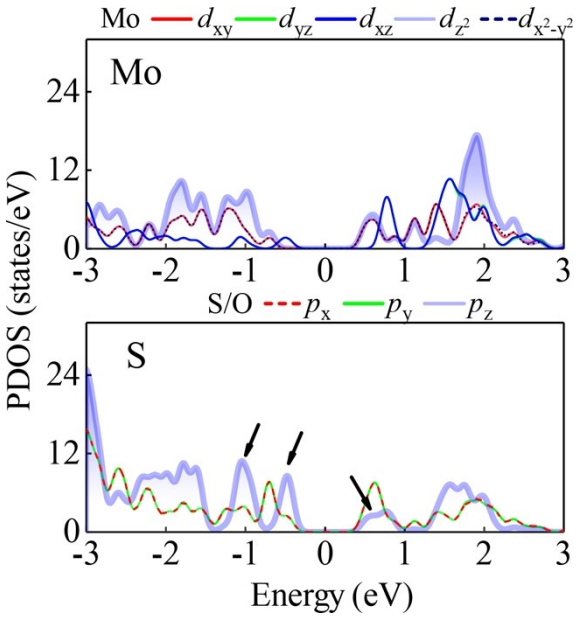

Fig. S5 PDOS for the *l*=5 triangular MoS_2 flake with S monomer.

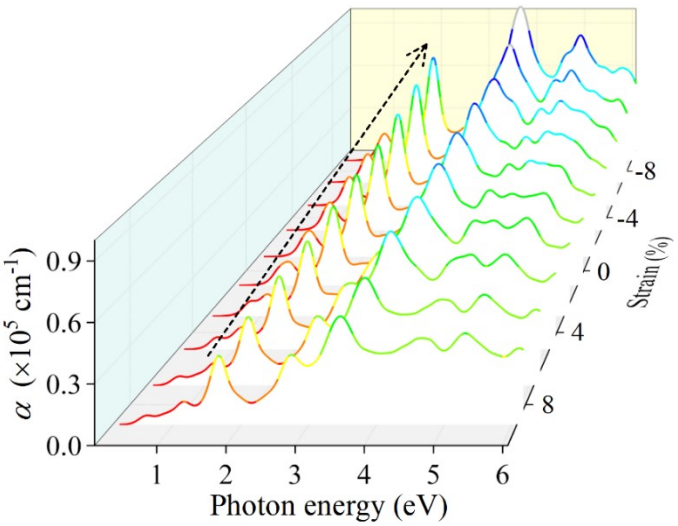

Fig. S6 In-plane strain engineering of the enhanced transition state for l=5 triangular MoS_2 flake.

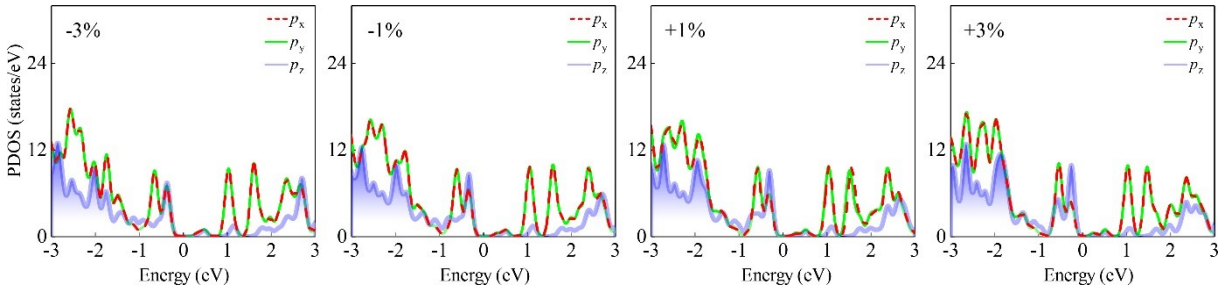

Fig. S7 PDOS for the compressive and tensile in-plane strained triangular MoS_2 flake, indicating the gradually enhanced band-edge p_z component for the tensile strain case.

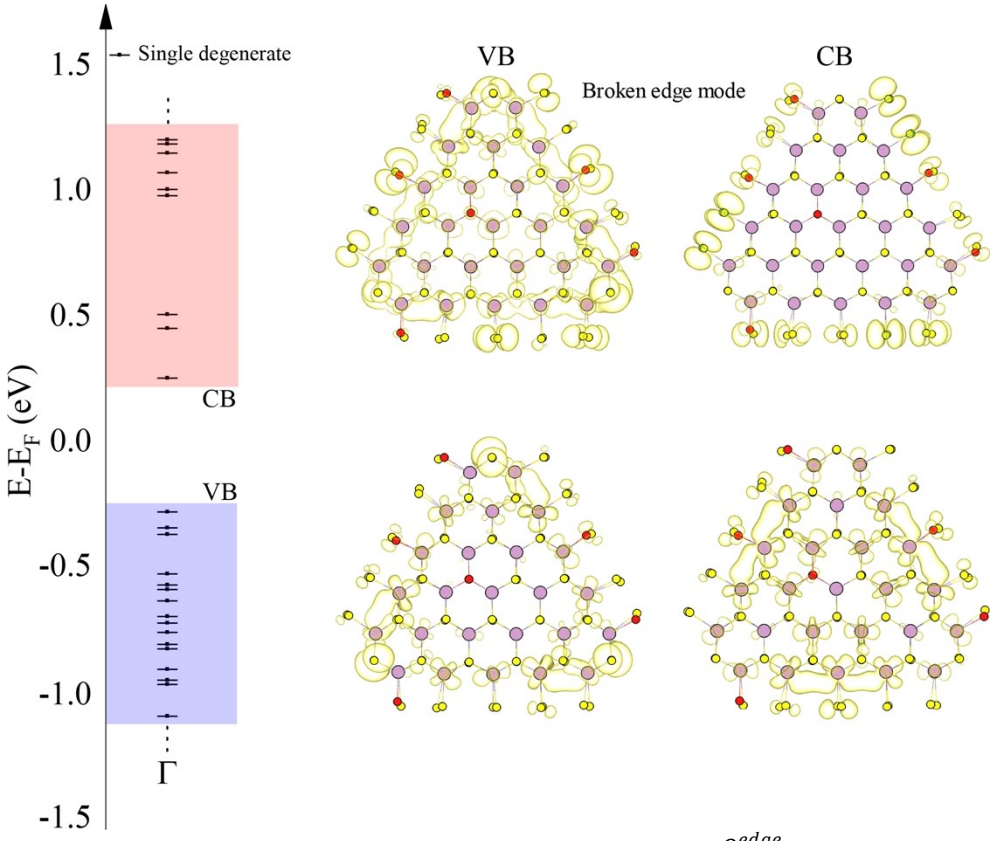

Fig. S8 Discrete level diagram for l=5 triangular MoS_2 flake with O_S^{edge} and the wavefunctions for edge states ($\rho = 2\times10^{-3}$ eÅ^{-3}).