Supporting information for

Direct Z-scheme WTe_2/InSe van der Waals heterostructure for overall water splitting

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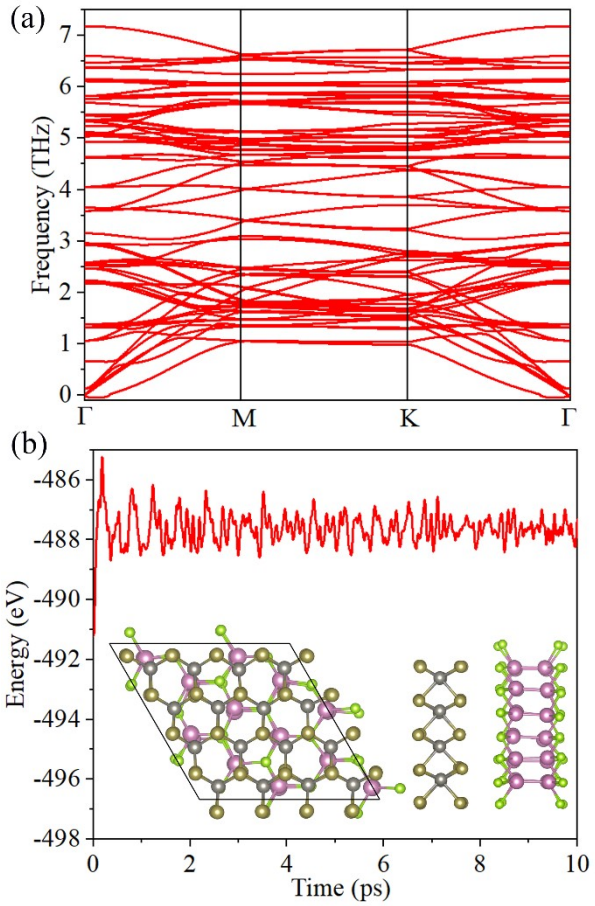

Figure S1. (a)The phonon dispersion curves and (b) evolution of total energy and snapshot structure from AIMD simulations of WTe_2/InSe heterostructure.

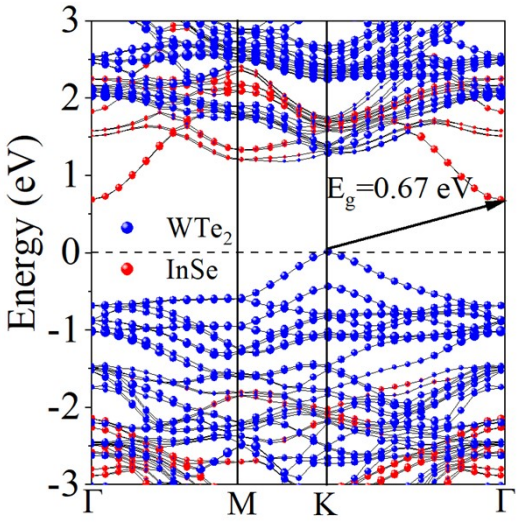

Figure S2. The projected band structure of WTe_2/InSe vdW heterostructure by using HSE06+SOC functional.

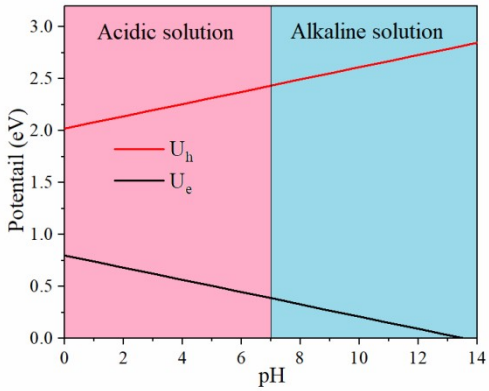

Figure S3. The external potentials U_e and U_h as a function of pH value.

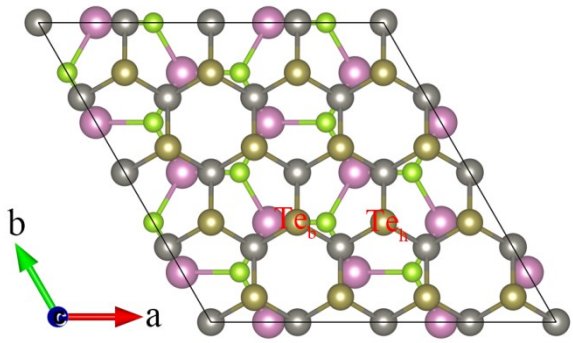

Figure S4. The different sites of Te atom in WTe_2/InSe heterostructure

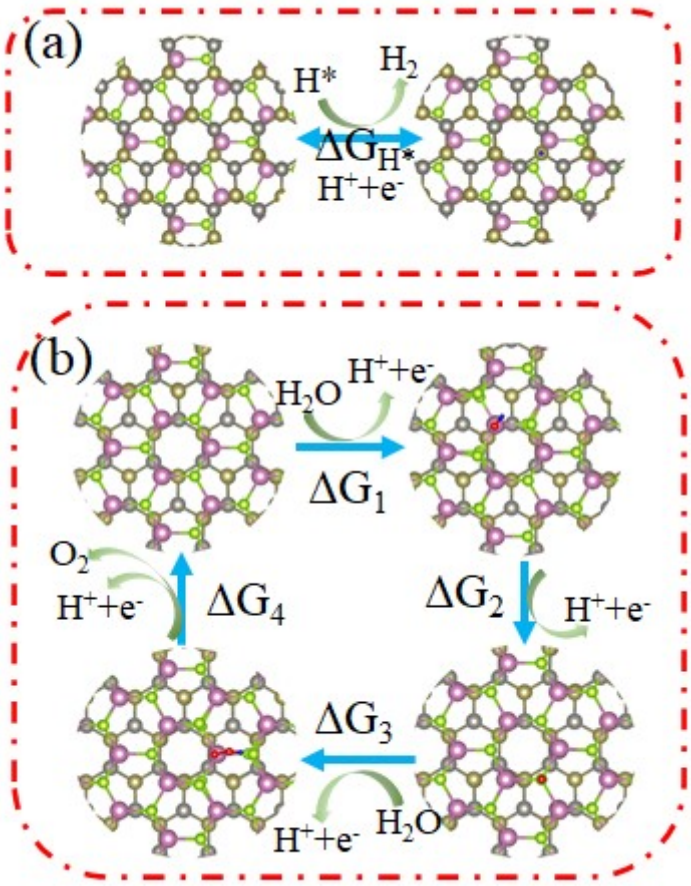

Figure S5. Proposed photocatalytic pathways and the most stable absorbed site of intermediates on the WTe_2/InSe heterostructure (a) HER and (b) OER.

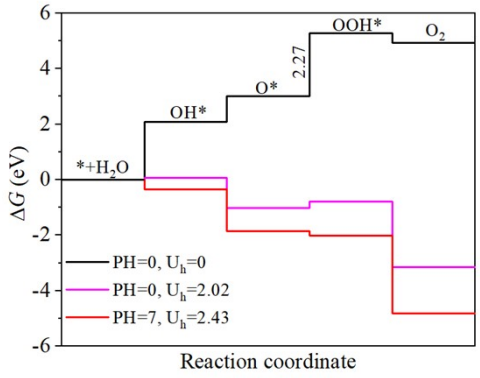

Figure S6. Free energy diagrams for OER on perfect WTe_2/InSe vdW heterostructure.

Table S1. The calculated lattice a, interlayer distant d, formation energy E_f and binding energy E_b of WTe_2/InSe heterostructure for three different stacking configurations

Configuration	a (Å)	d (Å)	E_f (eV)	E_b (meV/Å)
I	7.032	3.53	-0.8098	16.465
II	7.033	3.55	-0.8101	16.470
III	7.033	3.55	-0.8096	16.455

Table S2. Energy conversion efficiency of light absorption (η_{abs}), carrier utilization (η_{cu}) and STH (η_{STH}) for WTe_2 and InSe monolayers and WTe_2/ InSe vdW heterostructure

System	η_{abs}(%)	η_{cu}(%)	η_{STH}(%)
WTe_2	63.07	5.01	3.1
InSe	31.01	35.85	11.1
WTe_2/InSe	89.07	63.55	56.6