# Supporting Information

# Theoretical Design of an InSe/GaTe vdW Heterobilayer: A Potential Visible-Light Photocatalyst for Water Splitting

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**15 pages, 1 text, 1 table, 10 figures**

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**Text S1. Band alignment strategy for InSe/GeTe HBL**

We calculate the $E_{CBM}$ and $E_{VBM}$ combined with the macroscopic average electricstatic potential ($\bar{\bar{V}}$), which has proven to be successful for several semiconductor heterostructures. The static electric potential $V(\vec{r})$ is calculated by solving Poisson equation:

$$\nabla^2 V(\vec{r}) = -\frac{\rho(\vec{r})}{\varepsilon_0} \quad (1)$$

where the charge density $\rho(\vec{r}) = \rho_e(\vec{r}) + \rho_i(\vec{r})$. The valence electron density of the occupied states $\rho_e(\vec{r})$ and the charge density of atomic cores $\rho_i(\vec{r})$ can be calculated as follows: $\rho_e(\vec{r}) = \sum_{occ}|\varphi_i(\vec{r})|^2$ and $\rho_i(\vec{r}) = \sum_j q_j \delta(\vec{r} - \vec{r}_j)$, where $q_j$ and $\vec{r}_j$ are the charge and position of the *j*th atomic core. To visualize the static electric potential profiles extending throughout the layers, $V(\vec{r})$ is then averaged on the plane (*xy*-plane) perpendicular to the layers using the equation:

$$\bar{V}(z) = \frac{1}{S_{xy}} \int V(\vec{r}) dx dy \quad (2)$$

The integration on the *xy*-plane is carried using a 60 × 60 point mesh. $S_{xy}$ represents the area of the *xy*-plane of the layers. Furthermore, the macroscopic average $\bar{\bar{V}}(z)$ is accomplished by averaging $\bar{V}(z)$ over a distance ($L$) corresponding to InSe and GaTe layers, respectively :

$$\bar{\bar{V}}(z) = \frac{1}{L} \int_{-2/L}^{2/L} \bar{V}(z') dz' \quad (3)$$

The $\bar{V}$ and $\bar{\bar{V}}$ are the relative positions aligned to the vacuum level (set to zero).

The band edge positions of InSe or GaTe layer in the HBL can be determined as follow:

$$E_{CBM/VBM}^{InSe/GaTe} = \bar{\bar{V}}_{HLs}^{InSe/GaTe} + \left[E_{CBM/VBM} - \bar{\bar{V}}\right]_{InSe/GaTe} \quad (4)$$

where the first term represents the $\bar{\bar{V}}$ positions of InSe or GaTe layer in the InSe/GaTe HBL, the second term represents the $E_{CBM/VBM}$ positions relate to $\bar{\bar{V}}$ in the distorted InSe and GaTe monolayers, respectively.

The conduction band offset ($\Delta E_C$) and valence band offset ($\Delta E_V$) of the InSe/GaTe HBL can be evaluated using the equation:

$$\Delta E_{C/V} = \left|E_{CBM/VBM}^{InSe} - E_{CBM/VBM}^{GaTe}\right| \quad (5)$$

**Table S1.** Lattice constant *a* (Å) of bulk InSe using PBE and different VDW functionals.

| System | Method | *a* (Å) |
|---|---|---|
| Bulk InSe | PBE | 4.093 |
| | optB88 | 4.071 |
| | PBE-D2 | 3.964 |
| | PBE-D3-G | 4.086 |
| | PBE-D3-BJ | 4.057 |
| | optPBE | 4.106 |
| | revPBE | 4.132 |
| | Expt.[1] | 4.05 |

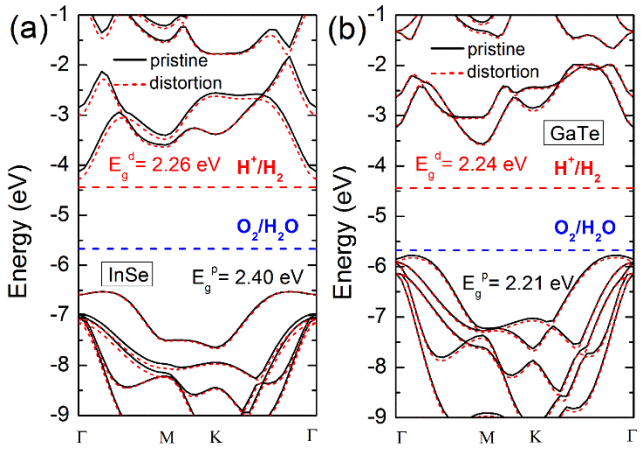


**Figure S1.** Band structures of (a) InSe monolayer and (b) GaTe monolayer calculated by HSE06 functional, respectively. The black solid lines represent the pristine isolated InSe and GaTe layers, the red dotted lines represent the distorted InSe or GaTe layer in the InSe/GaTe HBL.

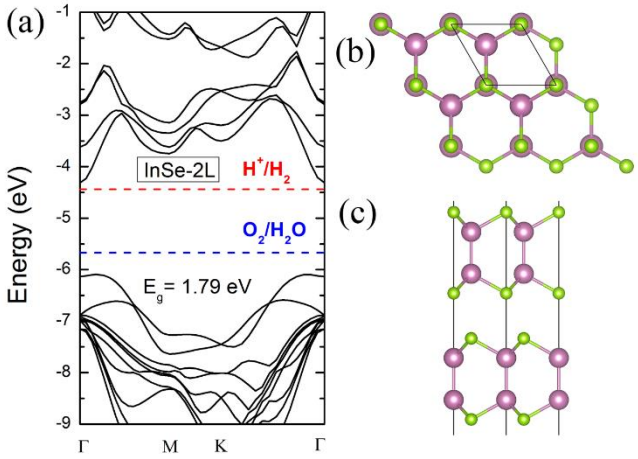


**Figure S2.** (a) Band structure of InSe bilayer calculated by HSE06 functional. (b) Top and (c) side views of the bilayer structure.

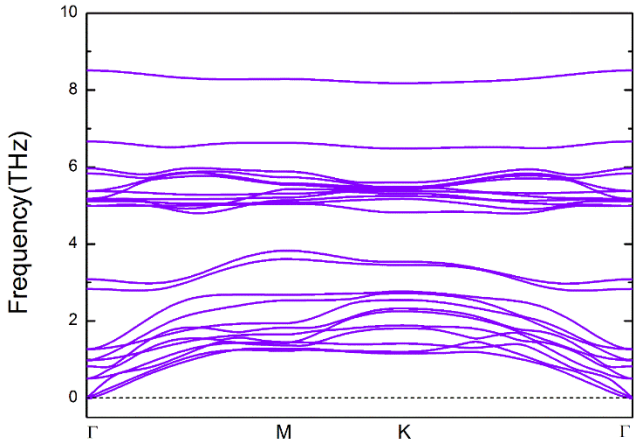


**Figure S3.** Calculated phonon dispersion of the InSe/GaTe HBL with stacking pattern II.

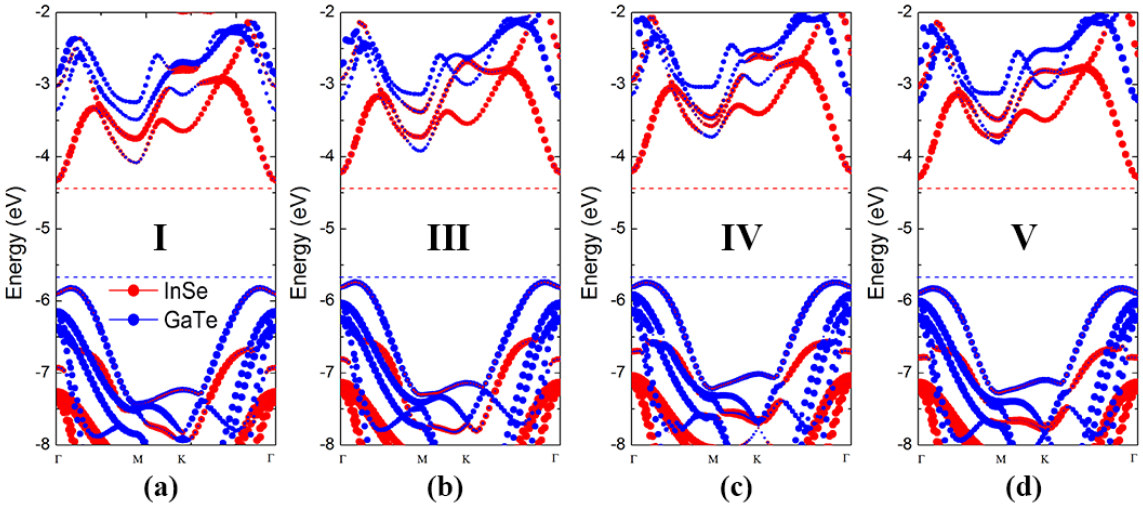


**Figure S4.** Orbital-resolved band structures of InSe/GaTe HBL with stacking pattern (a) I, (b) III, (c) IV and (d) V. The sizes of the red and blue dots represent the weights of InSe and GaTe, respectively. The dotted red and blue lines represent the reduction potential of $H^+/H_2$ and the oxidation potential of $O_2/H_2O$, respectively.

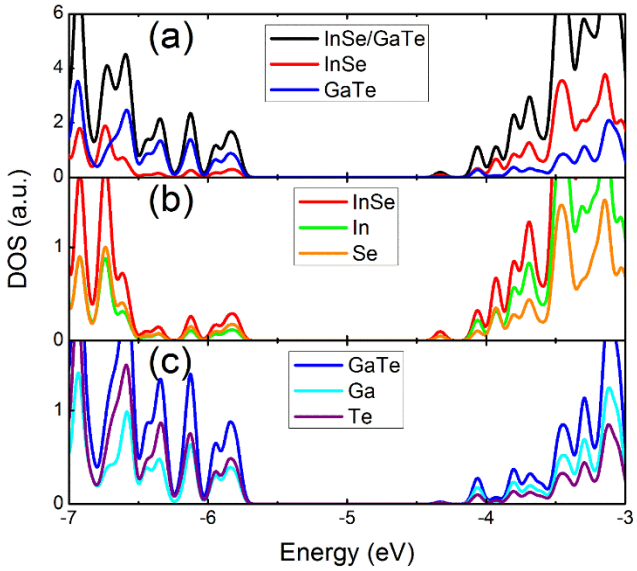


**Figure S5.** TDOS and PDOS of the (a) InSe/GaTe HBL, (b) InSe monolayer and (c) GaTe monolayer.

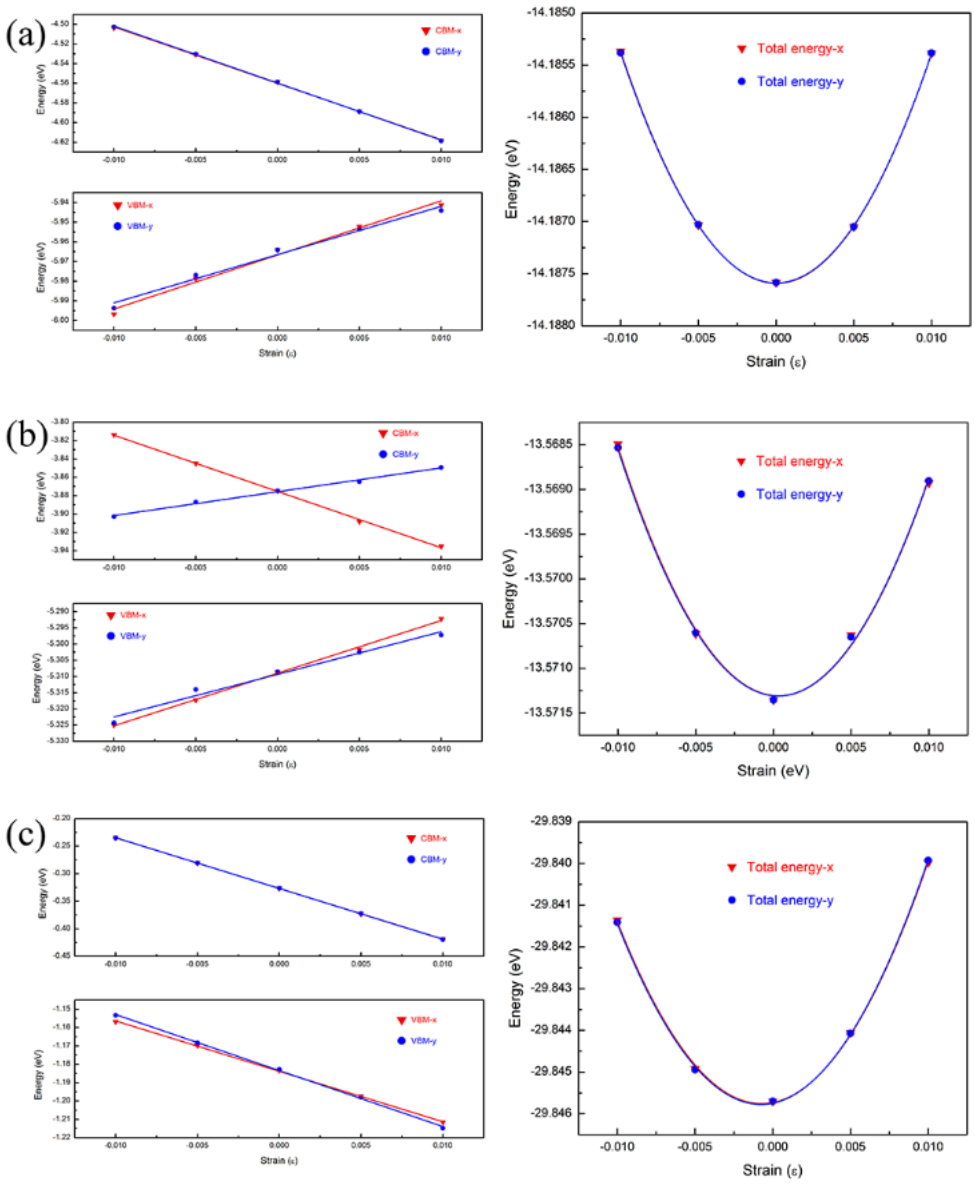


**Figure S6**. Linear fitting of band edges and quadratic fitting of total energy as a function of uniaxial strain for (a) InSe monolayer (b) GaTe monolayer and (c) InSe/GaTe HBL.

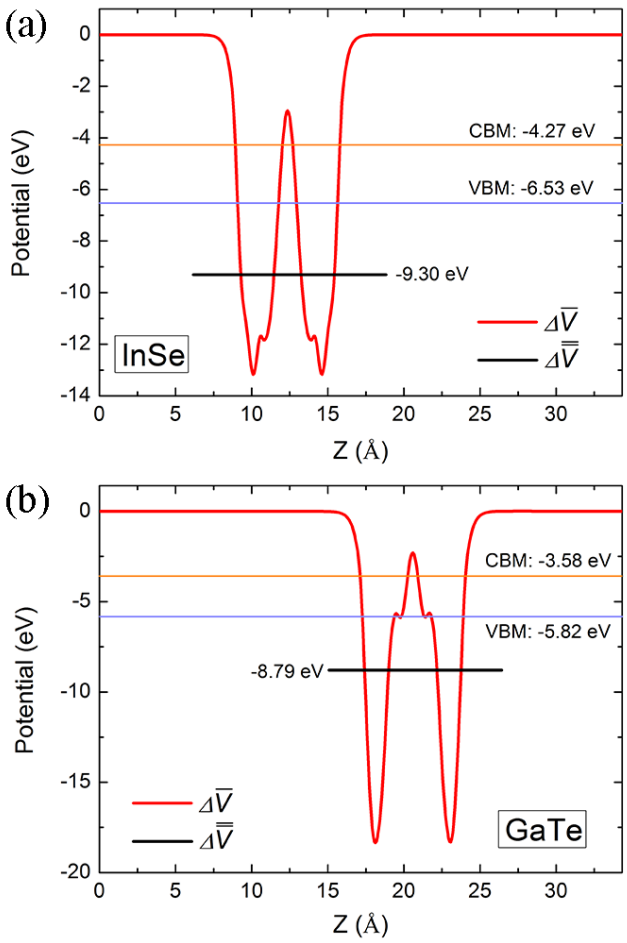


**Figure S7.** Plane-averaged potential $\bar{V}$ (red line) and macroscopic average potential $\bar{\bar{V}}$ (black line) for the distorted (a) InSe and (b) GaTe layers in the InSe/GaTe HBL calculated by the HSE06 functional. The brown and blue represent the CBM and VBM respectively.

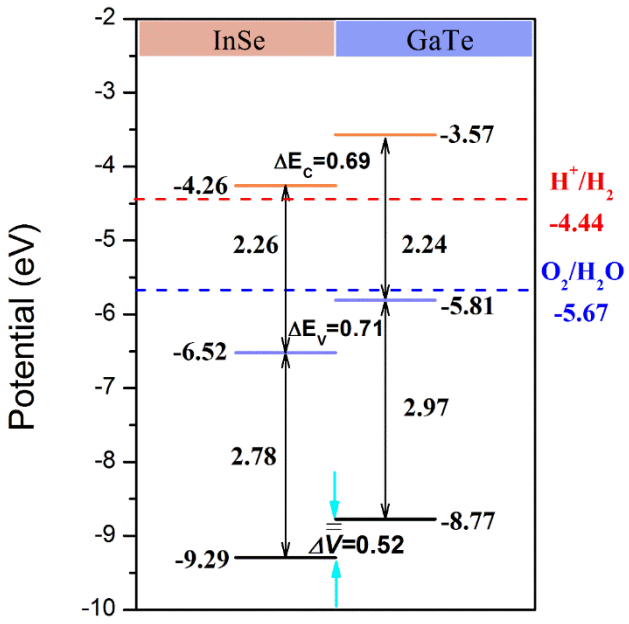


**Figure S8.** Band alignments of the InSe/GaTe HBL with stacking pattern II calculated by HSE06 functional methods. The brown, blue and black lines represent the CBM, VBM and macroscopic average potential, respectively. The energy differences are marked by the black arrows.

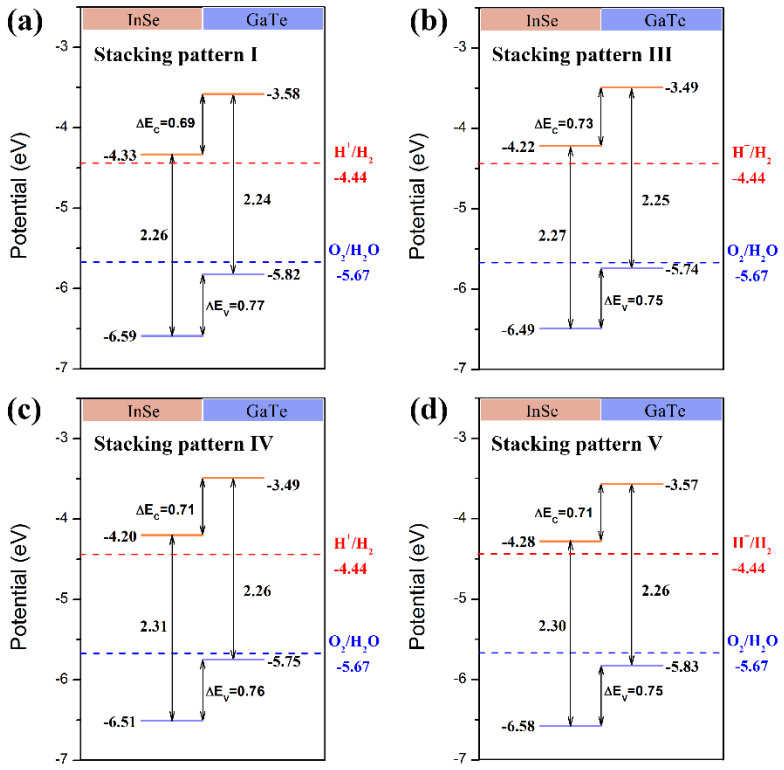


**Figure S9.** Band alignments of the InSe/GaTe HBL with stacking pattern (a) I, (b) III, (c) IV and (d) V. The brown and blue lines represent the CBM and VBM, respectively. The energy differences are marked by the black arrows.

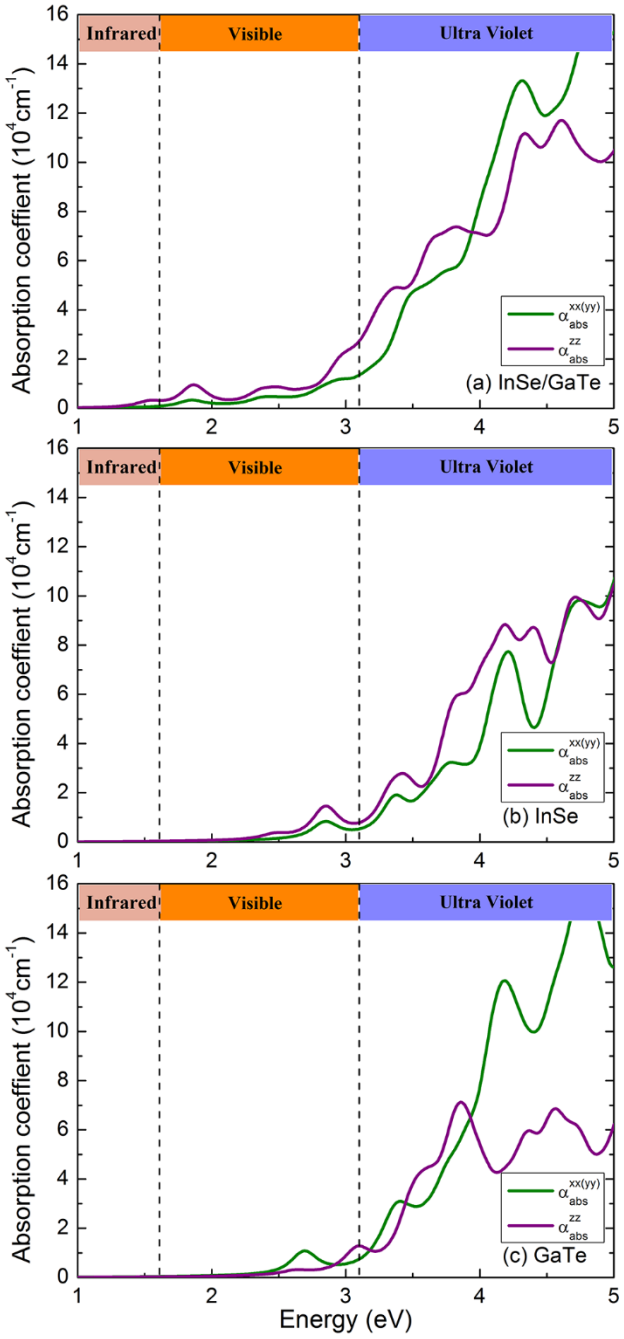


**Figure S10.** Calculated absorption coefficients of the (a) InSe/GaTe HBL, (b) InSe monolayer and GaTe monolayer, respectively. The green and purple lines represent the absorption coefficients parallel and normal to the planes of these three structures, respectively.

## REFERENCES

1. Chen, Z.; Biscaras, J.; Shukla, A., A high performance graphene/few-layer InSe photo-detector. *Nanoscale* **2015,** *7*, 5981-5986.