Supplementary material

Two-dimensional layered Janus-\( \text{In}_2\text{SeTe}/\text{C}_2\text{N} \) van der Waals heterostructures for photocatalysis and photovoltaics: First-principles calculations

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1. The band structure and electrostatic potentials with/without dipole corrections
2. Two types of the heterostructures fabricated by \( \text{C}_2\text{N} \) and Janus \( \text{In}_2\text{SeTe} \) monolayers
3. The molecular dynamics simulations of Janus-\( \text{In}_2\text{SeTe}/\text{C}_2\text{N} \) vdWHs
4. The exciton binding energies of Janus-\( \text{In}_2\text{SeTe}/\text{C}_2\text{N} \) vdWHs and their components
5. The band structures and alignments of \( \text{InSe}/\text{C}_2\text{N} \) and \( \text{InTe}/\text{C}_2\text{N} \) vdWHs
6. Computational detail of the carrier mobility
7. Orthorhombic lattices of Janus-\( \text{In}_2\text{SeTe}/\text{C}_2\text{N} \) vdWHs and their parent monolayers
8. The elastic modulus and the deformation potential calculations of \( \text{C}_2\text{N} \) and Janus-\( \text{In}_2\text{SeTe} \) monolayers
9. The elastic modulus and the deformation potential calculations of Janus-\( \text{In}_2\text{SeTe}/\text{C}_2\text{N} \) vdWHs
10. Discussion S1
1. The band structure and electrostatic potentials with/without dipole corrections

![Graphs showing band structures and electrostatic potentials](image)

Fig. S1. (a) The band structure and electrostatic potentials of Janus In2SeTe monolayer with/without dipole corrections applied. (b) The electrostatic potentials of Janus In2SeTe monolayer with dipole corrections applied. Band structures (c) TeIn2Se/C2N, (d) SeIn2Te/C2N heterostructure obtained with/without dipole corrections. All results are calculated with PBE functional.

2. Two types of the heterostructures fabricated by Janus-In2SeTe and C2N monolayers

![Heterostructures](image)

Fig. S2. Top and side views of (a) TeIn2Se/C2N and (b) SeIn2Te/C2N vdWHs with γ-stacking configuration.
3. The molecular dynamics simulations of Janus-In$_2$SeTe/C$_2$N vdWHs

![Energy plots](image1)

**Fig. S3.** The results of molecular dynamics simulation of (a,c) TeIn$_2$Se/C$_2$N and (b,d) SeIn$_2$Te/C$_2$N heterostructures.

4. The exciton binding energies of Janus-In$_2$SeTe/C$_2$N vdWHs and their components

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5. The band structures and alignments of InSe/C2N and InTe/C2N vdWHs

![Graphs showing band structures and alignments](image)

**Fig. S4.** The band structures of (a) InSe/C2N and (b) InTe/C2N heterostructures and (c) their band alignments. The redox potentials of water splitting at pH 0 (red dotted line) is shown for comparison.

6. Computational detail of the carrier mobility

Using the acoustic phonon limited method, we quantitatively estimate the carrier mobility for Janus-In2SeTe/C2N vdWHs and their parent monolayers. In 2D materials, the carrier mobility can be calculated by the formula [1-2]

\[
\mu_{2D} = \frac{2e\hbar^2 C_{2D}}{3k_B T m^* (E_i^o)^2}
\]

where \(e\), \(\hbar\), \(k_B\) and \(T\) is the electron charge, reduced Planck constant, Boltzmann constant and temperature, respectively. The temperature is set to be 300 K in the calculations. \(C_{2D}\) is the in-plane stiffness of 2D material calculated by \(C_{2D} = (\partial^2 (E - E_0)/\partial \varepsilon^2) / S\), where \(E - E_0\) is the total energy shift activated by strain effect and \(S\) is the equilibrium area, \(m^*\) is the effective mass in the transport direction for the electron or hole, which can be obtained by \(m^* = \hbar^2 (\partial^2 E / \partial k^2)^{-1}\). \(E_i^o\) is the deformation potential constant of the valence band maximum (VBM) for holes and conduction band minimum (CBM) for electrons along the transport direction, defined by \(E_i^o = \Delta V_i / (\Delta l / l_0)\), in which \(\Delta V_i\) is the energy change of the \(i\) band under proper cell compression and dilatation, \(l_0\) is the lattice constant in the transport direction, and \(\Delta l\) is the deformation of \(l_0\).

For convenience, orthorhombic lattices instead of pristine hexagonal lattices are used to calculate the uniaxial strain responses along x and y directions, as shown in Fig. S5. The \(m^*\) of electrons and holes are calculated by fitting parabolic functions to CBM and VBM along the transport direction. Then, the \(C_{2D}\) and \(E_i^o\) are obtained by parabolic fitting of the total energy shift and linearly fitting of the band edge positions of the VBM and CBM as a function of the uniaxial strain along x and y directions, respectively (see Fig. S6 and S7).

7. Orthorhombic lattices of Janus-In$_2$SeTe/C$_2$N vdWHs and their parent monolayers

Fig. S5. The orthorhombic lattices used to calculate the uniaxial strain responses along $x$ and $y$ directions for (a) C$_2$N, (b) Janus-In$_2$SeTe monolayers, (c) TeIn$_2$Se/C$_2$N, (d) SeIn$_2$Te/C$_2$N vdWHs.
8. The elastic modulus and the deformation potential calculations of C$_2$N and Janus-In$_2$SeTe monolayers

Fig. S6. The relationship between energy and strain for (a,b) C$_2$N and (c,d) In$_2$SeTe monolayers. The CBM and VBM along the x and y directions as a function of deformation proportion for (e,f) C$_2$N and (g,h) In$_2$SeTe monolayers.
9. The elastic modulus and the deformation potential calculations of Janus-\text{In}_2\text{SeTe}/\text{C}_2\text{N} \text{vdWHs}

Fig. S7. The relationship between energy and strain for (a,b) \text{TeIn}_2\text{Se}/\text{C}_2\text{N} and (c,d) \text{SeIn}_2\text{Te}/\text{C}_2\text{N} \text{vdWHs}. The CBM and VBM along the x and y directions as a function of deformation proportion for (e,f) \text{TeIn}_2\text{Se}/\text{C}_2\text{N} and (g,h) \text{SeIn}_2\text{Te}/\text{C}_2\text{N} \text{vdWHs}. 
10. Discussion S1:

As we know, in contrast to the quasiparticle selfconsistent GW method, the hybrid functional calculations (HSE) could underestimate the band gap of semiconductors. Meanwhile, we noted that the electronic structures of 2D Te-based semiconductors and Janus structures can be affected by the spin-orbital coupling (SOC) effect [Phys. Chem. Chem. Phys. 2018, 20, 18571-18578]. To check the quality of the HSE06 functional adopted in this work, we performed test calculations for Janus-In2SeTe monolayer using HSE06 and G0W0 functionals with/without SOC correction. The bandgap obtained with G0W0 is 2.28 eV, while G0W0+SOC gives a result of 1.97 eV, which is slightly higher than the HSE06 value (1.82 eV) obtained in our work. Indeed, higher level functionals may result in more accurate result. Unfortunately, the G0W0+SOC method is computationally very expensive and thus out of reach when large data sets of periodic systems are studied, as done in this work, especially for the heterostructure systems. Thus, the HSE06 method is probably a good option in our work. Of course, for future related experimental studies, the band gaps obtained in this work should be taken with care since it is likely is slightly underestimated.