Type-II Band Alignment and Enhanced Optical Properties in InP/Bi_2Se_3 van der Waals Heterojunctions: A First-Principles and FDTD Study

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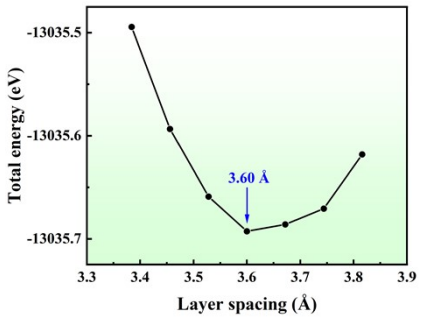

Figure S1 Binding energy curve analysis (E_{bind}(d)) for the H1 configuration, presented as the binding energy as a function of interlayer spacing.

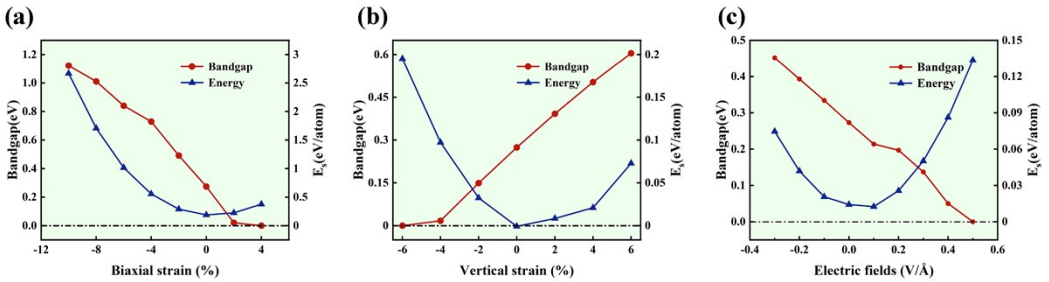

Figure S2 The influence of (a) biaxial strain, (b)vertical strain and (c)electric field on the bandgap and binding energy of the InP/Bi_2Se_3 vdWH.

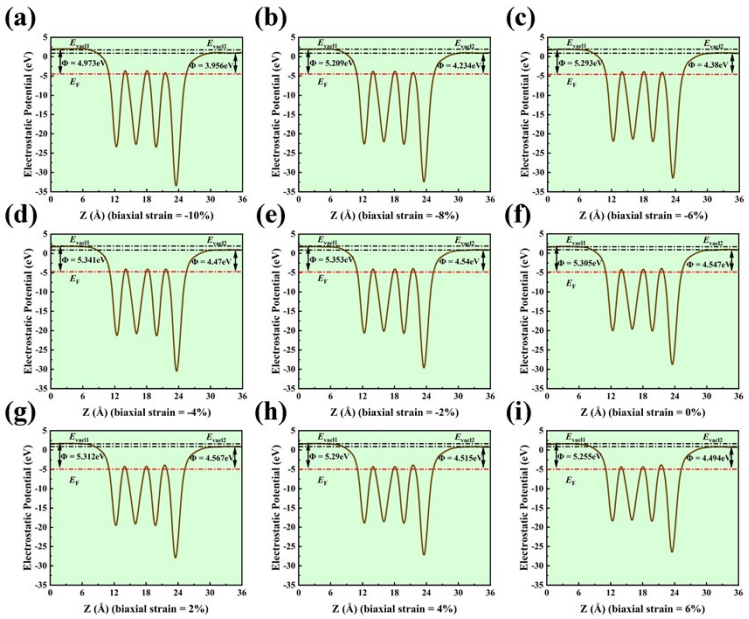

Figure S3 The variation of planar electrostatic potential and work function of the InP/Bi_2Se_3 vdWH under biaxial strain ranging from -10% to 6%. (The work function is marked on the corresponding graph.)

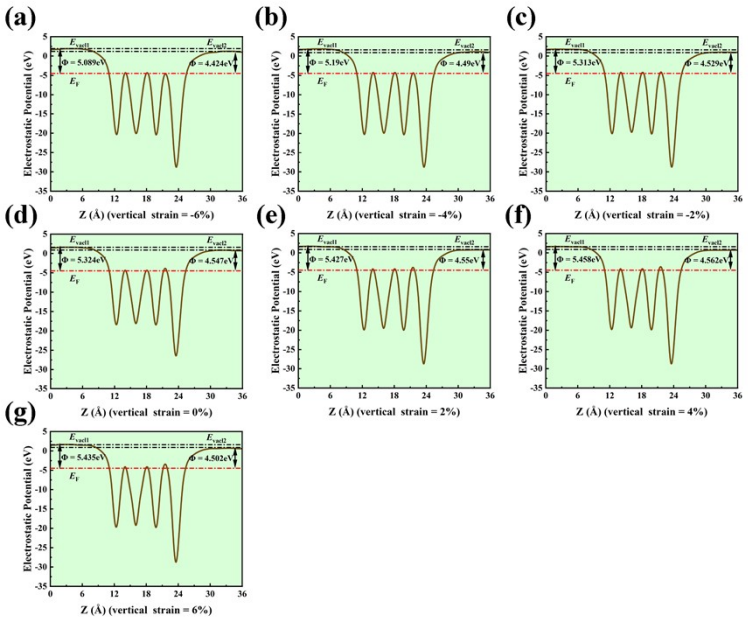

Figure S4 The variation of planar electrostatic potential and work function of the InP/Bi_2Se_3 vdWH under vertical strain ranging from -6% to 6%. (The work function is marked on the corresponding graph.)

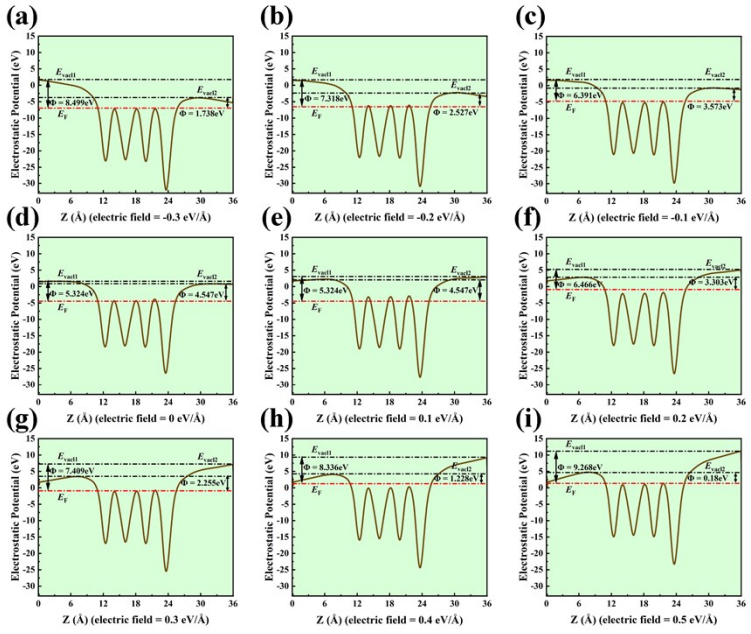

Figure S5 The variation of planar electrostatic potential and work function of the InP/Bi_2Se_3 vdWH under external electric fields ranging from -0.3 to 0.5 V/Å. (The work function is marked on the corresponding graph.)

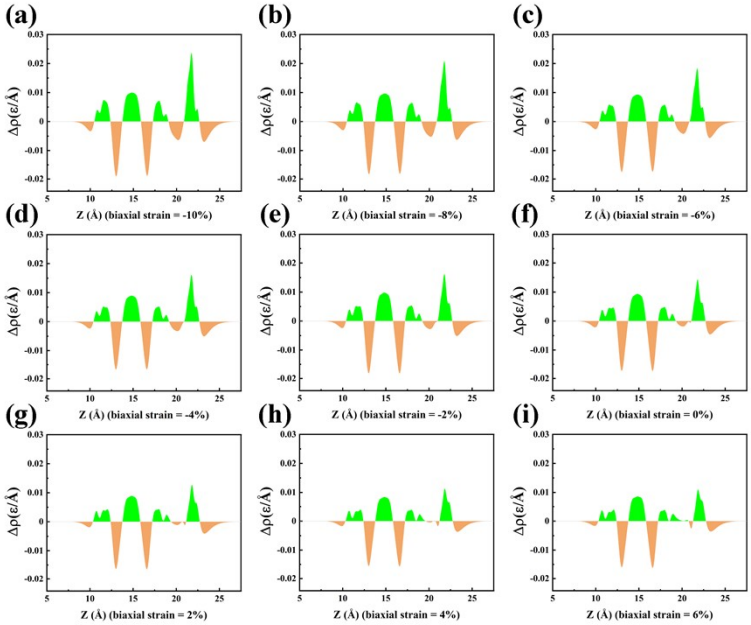

Figure S6 Differential charge density variation of the InP/Bi_2Se_3 vdWH under biaxial strain ranging from -10% to 6%.

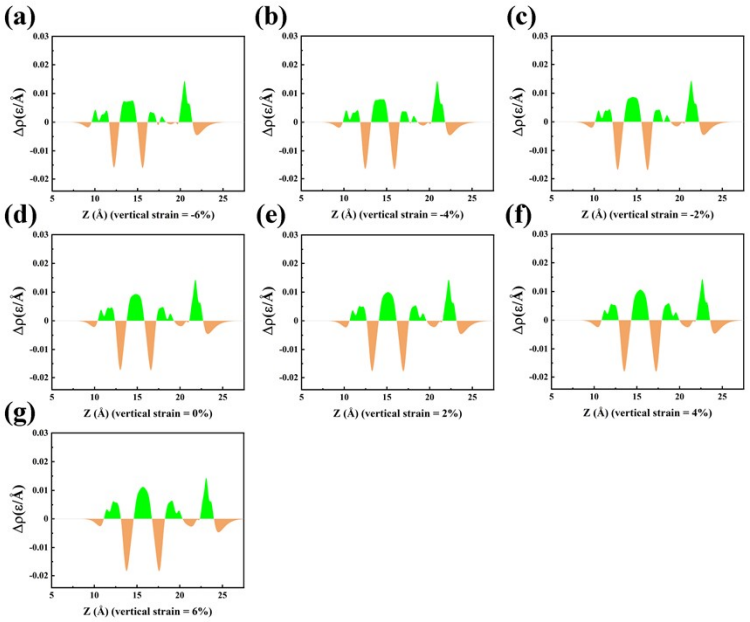

Figure S7 Differential charge density variation of the InP/Bi_2Se_3 vdWH under vertical strain ranging from 6% to 6%.

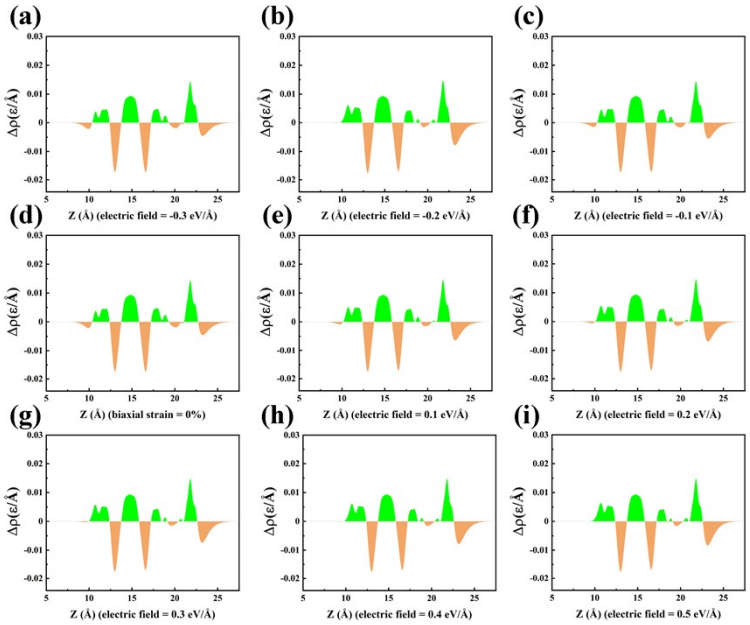

Figure S8 Differential charge density variation of the InP/Bi_2Se_3 vdWH under external electric fields ranging from -0.3 to 0.5 V/Å.

Table S1 Bader charge analysis of the InP/Bi_2Se_3 vdWH under different biaxial strains. Positive and negative values indicate electron accumulation and depletion, respectively.

Species	**0.90**	**0.92**	**0.94**	**0.96**	**0.98**	**1**	**1.02**	**1.04**	**1.06**
Bi	-1.01	-1.018	-1.03	-1.033	-1.03	-1.027	-1.021	-1.015	-1.008
Bi	-1.021	-1.031	-1.038	-1.036	-1.035	-1.033	-1.023	-1.02	-1.01
Se	0.621	0.625	0.627	0.628	0.627	0.625	0.626	0.623	0.62
Se	0.806	0.817	0.838	0.836	0.835	0.837	0.829	0.682	0.829
Se	0.619	0.622	0.623	0.623	0.621	0.618	0.614	0.61	0.603
In	-0.712	-0.749	-0.798	-0.793	-0.805	-0.811	-0.811	-0.806	-0.794
P	0.697	0.734	0.778	0.776	0.787	0.79	0.786	0.776	0.761

Table S2 Bader charge analysis of the InP/Bi_2Se_3 vdWH under different vertical strains. Positive and negative values indicate electron accumulation and depletion, respectively.

Species	**0.94**	**0.96**	**0.98**	**1**	**1.02**	**1.04**	**1.06**
Bi	-1.04	-1.038	-1.032	-1.028	-1.021	-1.016	-1.011
Bi	-1.046	-1.042	-1.038	-1.033	-1.027	-1.021	-1.011

Se	0.642	0.638	0.631	0.625	0.619	0.614	0.608
Se	0.837	0.837	0.837	0.838	0.837	0.836	0.833
Se	0.633	0.628	0.624	0.618	0.611	0.606	0.6
In	-0.816	-0.816	-0.815	-0.811	-0.807	-0.805	-0.801
P	0.789	0.791	0.792	0.79	0.787	0.786	0.782

Table S3 Bader charge analysis of the InP/Bi_2Se_3 vdWH under different electric fields. Positive and negative values indicate electron accumulation and depletion, respectively.

Species	**-0.3**	**-0.2**	**-0.1**	**0**	**0.1**	**0.2**	**0.3**	**0.4**	**0.5**
Bi	-1.03	-1.03	-1.031	-1.03	-1.031	-1.031	-1.03	-1.03	-1.03
Bi	-1.038	-1.038	-1.038	-1.038	-1.039	-1.038	-1.038	-1.038	-1.038
Se	0.629	0.63	0.63	0.63	0.629	0.63	0.63	0.63	0.629
Se	0.839	0.839	0.839	0.839	0.839	0.839	0.839	0.839	0.838
Se	0.622	0.622	0.622	0.622	0.622	0.622	0.622	0.622	0.622
In	-0.808	-0.808	-0.808	-0.808	-0.808	-0.808	-0.808	-0.808	-0.808
P	0.785	0.785	0.785	0.785	0.785	0.785	0.785	0.785	0.785