Supplementary Information

Enhanced Electronic and Optical Properties of Three TMD Heterobilayers

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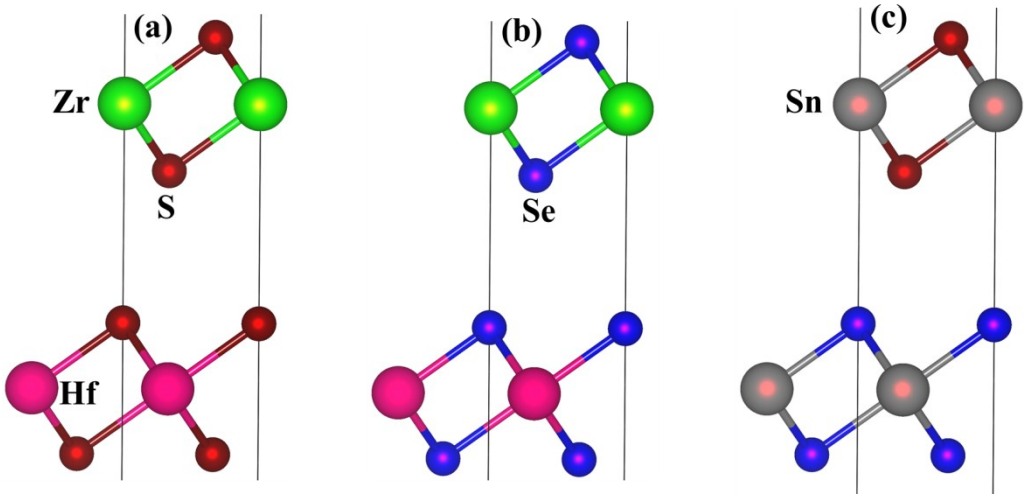
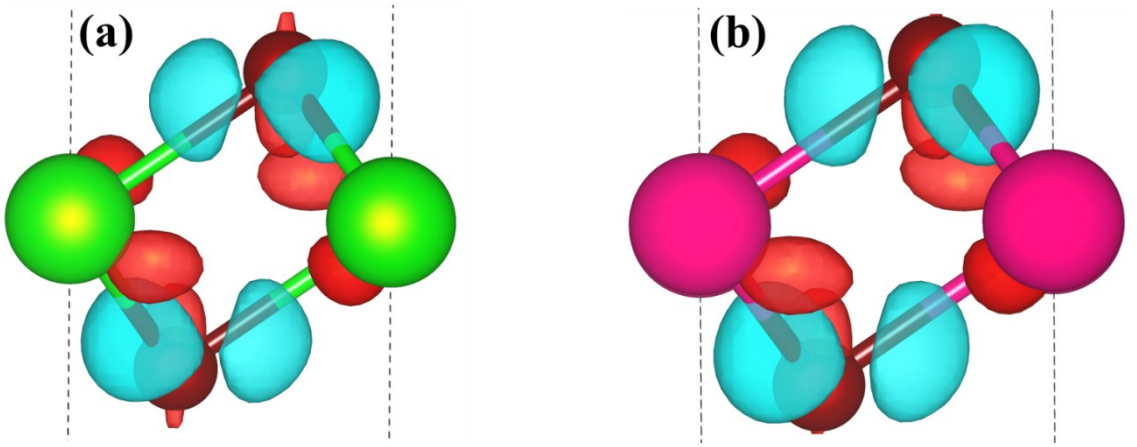


S1: The side view of the optimized structures with AB-stacking: (a) ZrS_2/HfS_2; (b) $ZrSe_2/HfSe_2$ and (c) $SnS_2/SnSe_2$ heterobilayers.

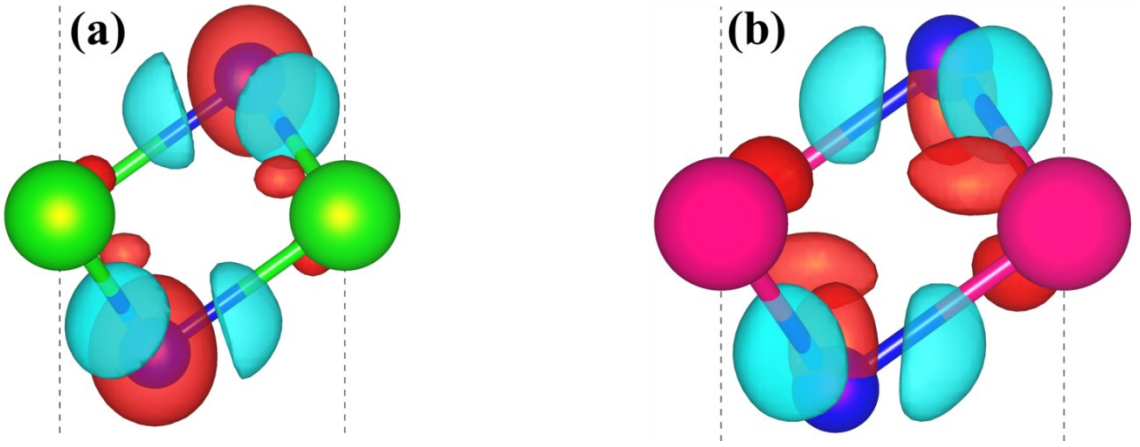
Table. S1: Geometric parameters (lattice constant a, spacing d, bond length and binding energy E_b for the predicted heterobilayers (L= Hf, Zr and Sn) with AB stacking.

Heterobilayers	ZrS_2/HfS_2	$ZrSe_2/HfSe_2$	$SnS_2/SnSe_2$
a (Å)	3.63	3.63	3.63
d (Å)	5.49	5.49	5.49

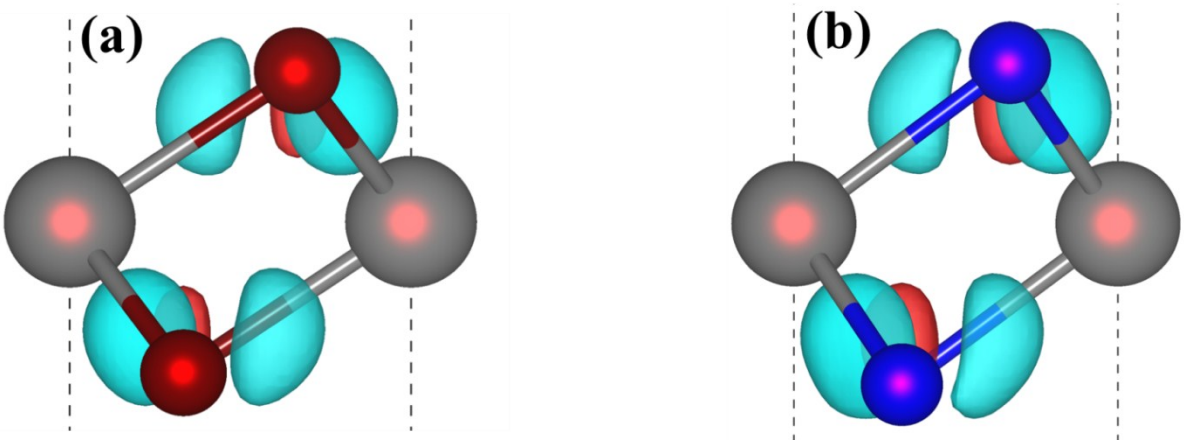
L-S (Å)	2.55	-	2.57
L-Se (Å)	-	2.65	2.68
E_b (eV/atom)	-0.021	-0.011	-0.030



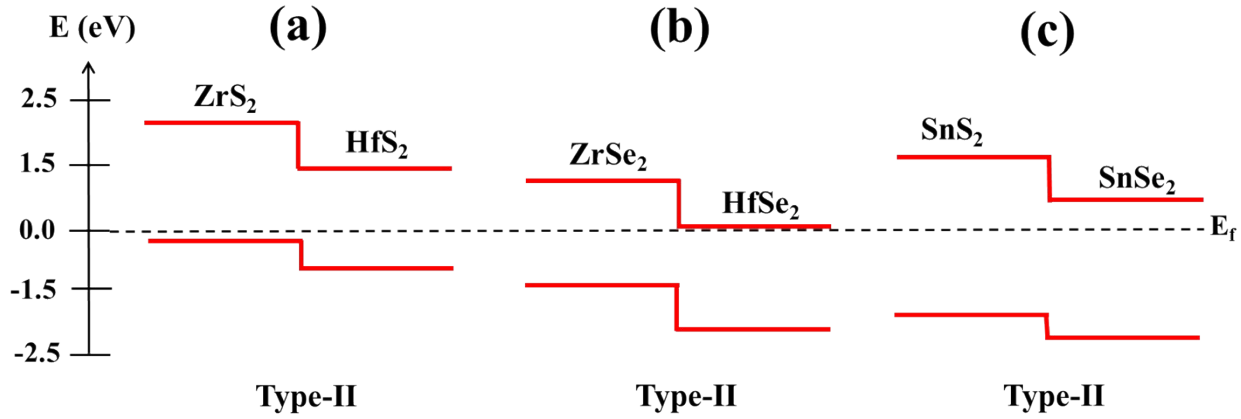
S2: Electronic charge density difference for (a) ZrS_2 monolayer and (b) HfS_2 monolayer at the isosurface of 5.3×10^{-4} $e/\text{Å}$. Where red color indicates charge accumulation (electrons) and cyan color charge depletion (holes).



S3: Electronic charge density difference for (a) $ZrSe_2$ monolayer and (b) $HfSe_2$ monolayer at the isosurface of 5.3×10^{-4} $e/\text{Å}$. Where red color indicates charge accumulation (electrons) and cyan color charge depletion (holes).



S4: Electronic charge density difference for (a) SnS_2 monolayer and (b) $SnSe_2$ monolayer at the isosurface of 5.3×10^{-4} $e/\text{Å}$. Where red color indicates charge accumulation (electrons) and cyan color charge depletion (holes).



S5: Schematic diagram of the Type-II band alignment between two isolated monolayers: (a) ZrS_2 and HfS_2; (b) $ZrSe_2$ and $HfSe_2$; (c) SnS_2 and $SnSe_2$.