Electronic Supplementary Information for

2D SnSe-based vdW heterojunctions: Tuning Schottky barrier by reducing Fermi level pinning

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Table S1. Details on the Monkhorst-Pack (MP) sampling of k-points for each of the following heterostructures.

Heterostructure	Monkhorst–Pack k-points grid
SnSe/Graphene	7×1×1
SnSe/ *ZT-MoSe2*	3×1×1
SnSe/ *ZT-MoS2*	4×1×1
SnSe/ *H-NbS2*	3×1×1

Table S2. Lattice constants (in Å) and angles between them of the SnSe/G supercells. The last column gives the total number of atoms in the supercells of the heterostructure.

	2D SnSe a=4.30, b=4.36, α=90°			Graphene a=2.46, b=2.46, α=120°			Mismatch (%)			Number of atoms
	u	v	γ	u	v	γ	u	v	γ	
Case 1	4.26	39.36	90.00	4.30	39.21	90.00	0.88	0.39	0.00	100
Case 2	13.02	13.70	70.16	13.07	13.61	71.34	0.41	0.61	1.65	100
Case 3	4.26	44.28	90.00	4.30	43.57	90.00	0.88	1.64	0.00	112
Case 4	8.52	22.14	90.00	8.60	21.78	90.00	0.90	1.64	0.00	112

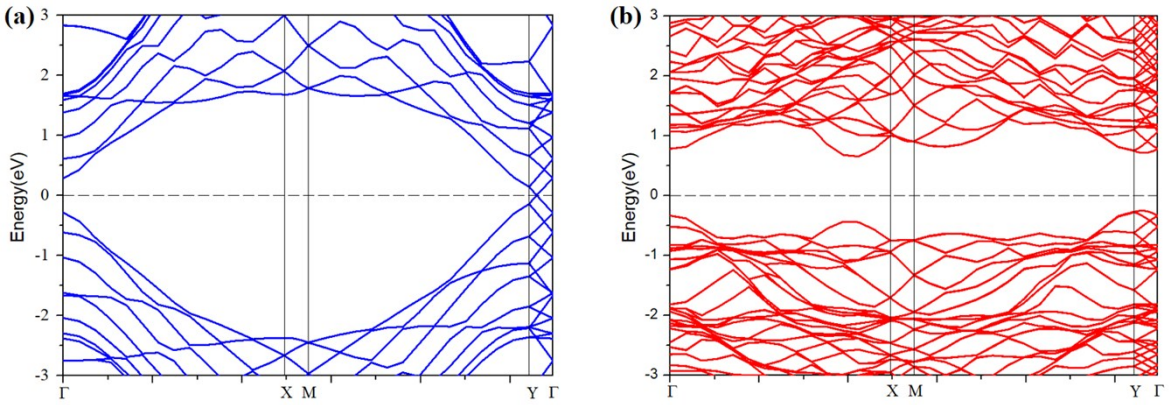


Fig. S1 Electronic band structure of (a) freestanding graphene, and (b) the freestanding SnSe sheet.

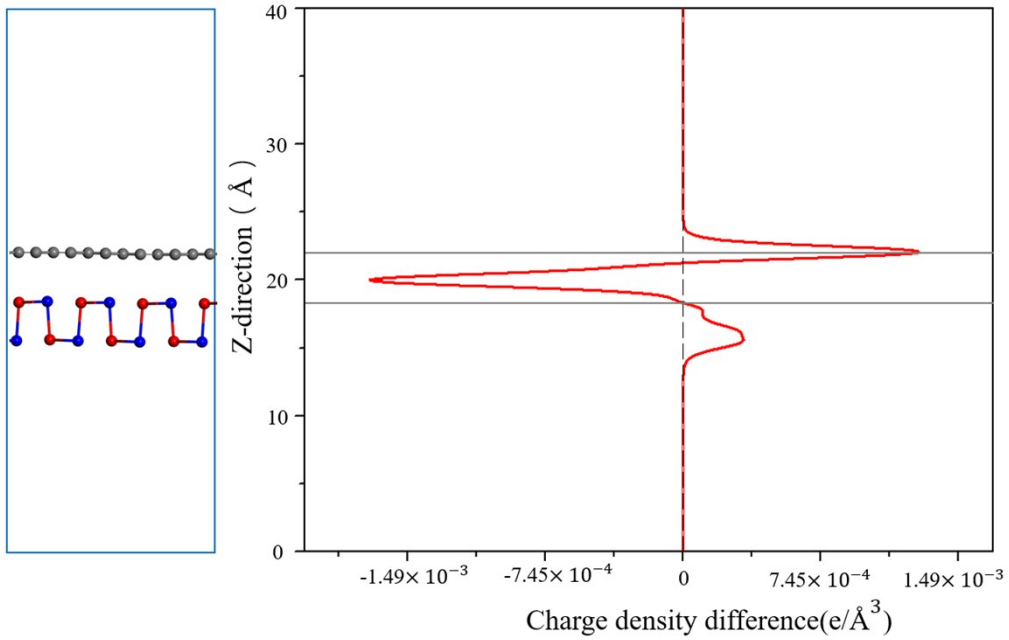


Fig. S2 Charge density difference of the SnSe/G heterostructure, where the averaged charge density is in the XY plane parallel to the interface. Gray, blue, and red spheres stand for C, Sn, and Se atoms, respectively.

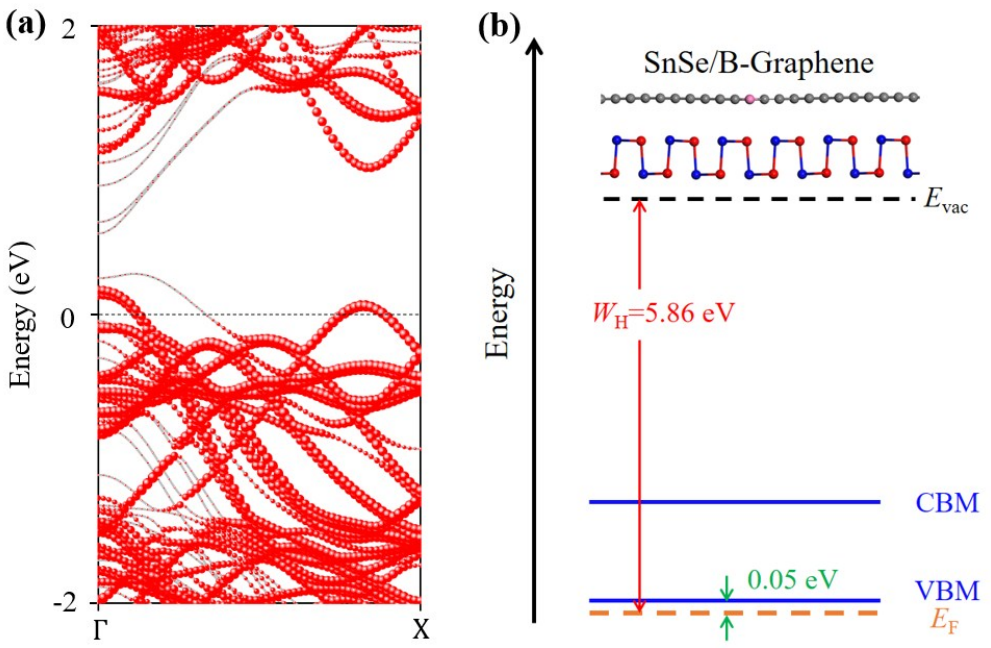
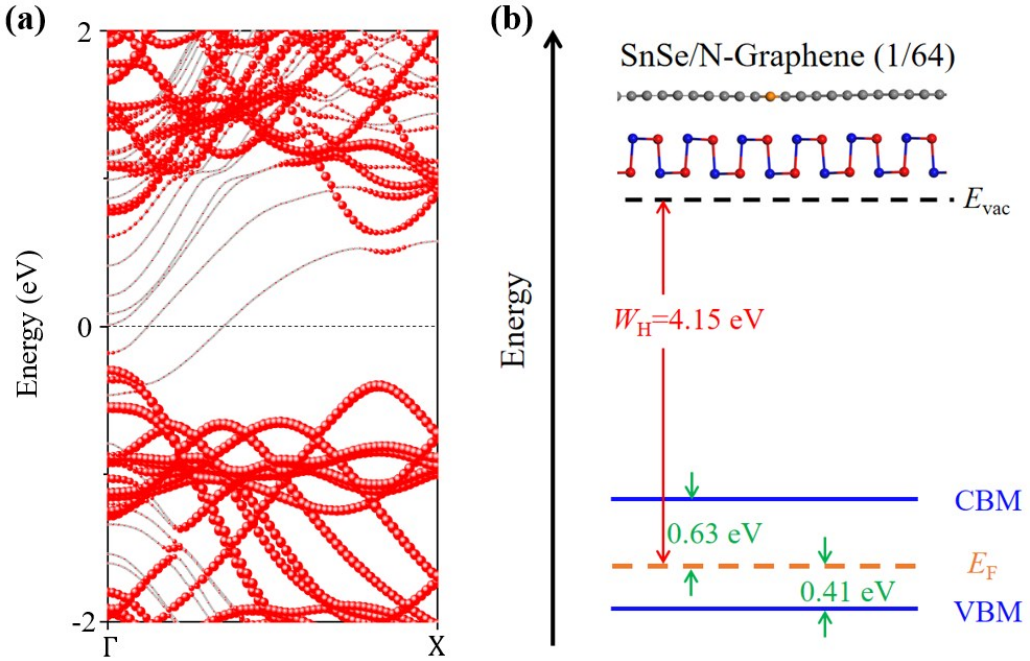
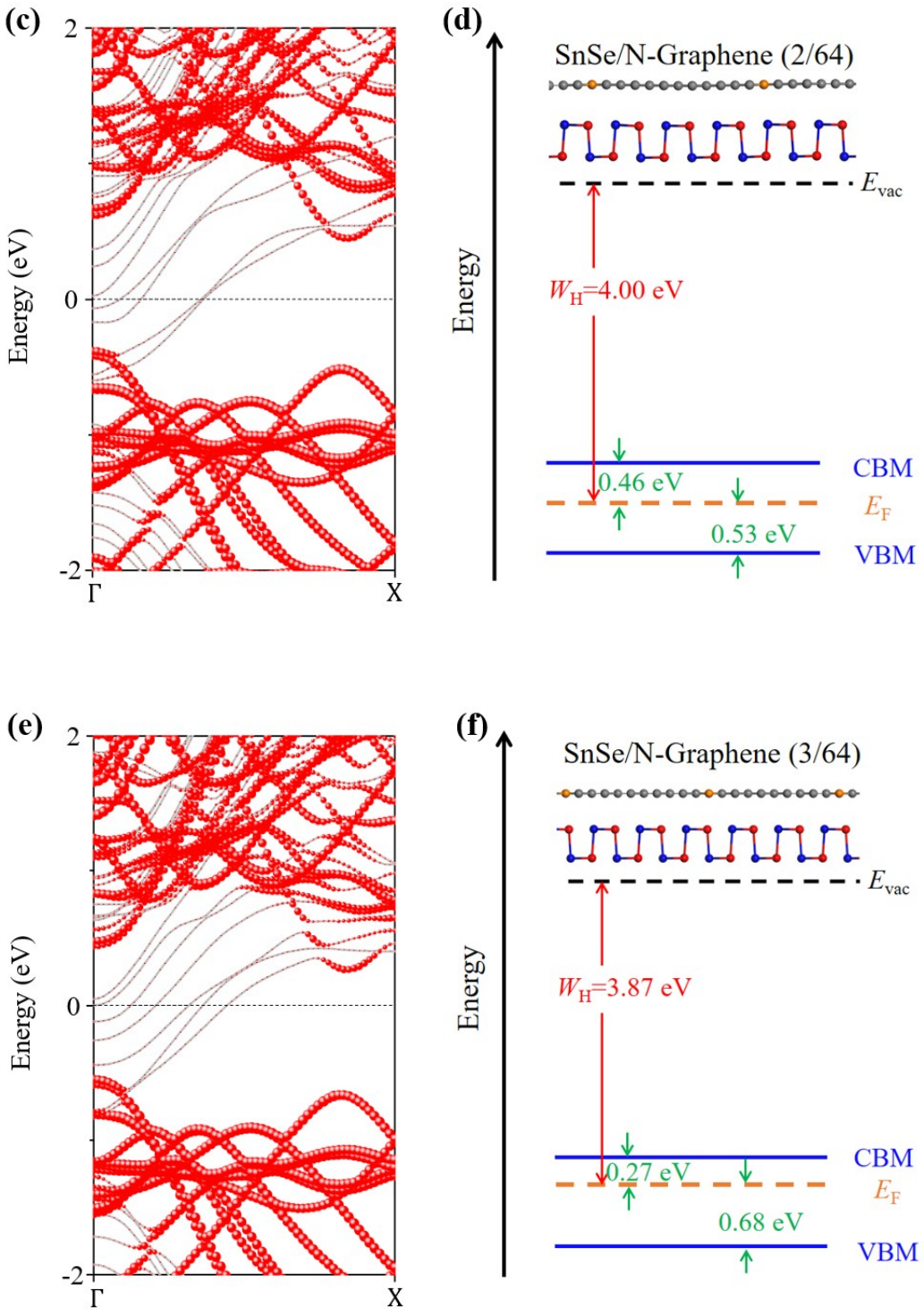


Fig. S3 (a) Band structure projected on the SnSe sheet, and (b) band alignment of SnSe/B-Graphene with B doping concentration of 1/64. W_H is the work function of the heterostructure. E_{vac} represents the vacuum level. Pink, gray, blue, and red spheres stand for B, C, Sn, and Se atoms, respectively.




(c)
2
0
-2
Energy (eV)
Γ
X
(d)
SnSe/N-Graphene (2/64)
E_{vac}
W_H=4.00 eV
Energy
CBM
0.46 eV
E_F
0.53 eV
VBM
(e)
2
0
-2
Energy (eV)
Γ
X
(f)
SnSe/N-Graphene (3/64)
E_{vac}
W_H=3.87 eV
Energy
CBM
0.27 eV
E_F
0.68 eV
VBM

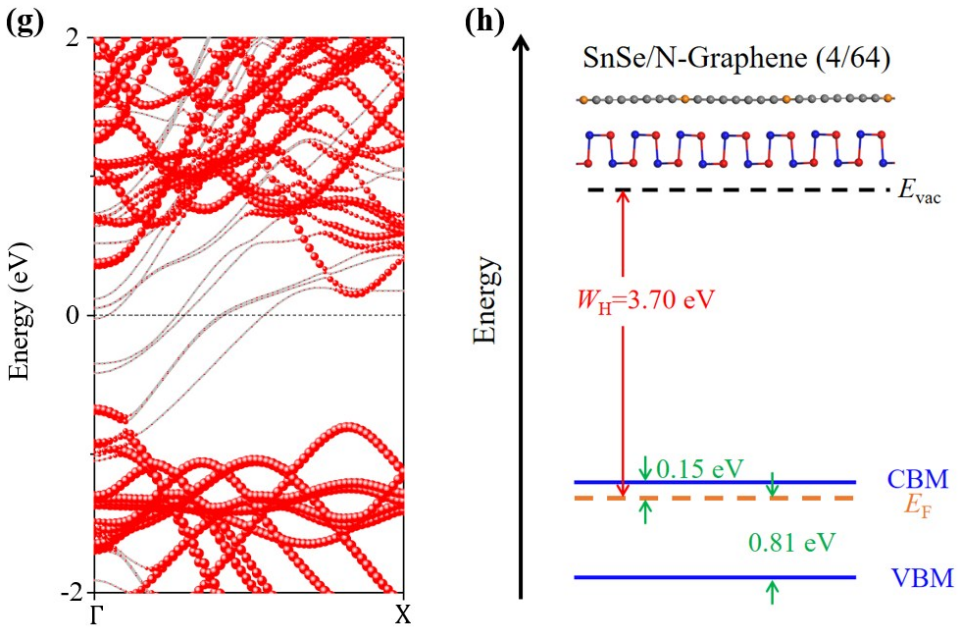


Fig. S4 (a), (c), (e), and (g) Projected Band structures on the SnSe sheet, and (b) (d), (f), and (h) band alignments of the SnSe/N-Graphene heterostructures with N doping concentration of 1/64, 2/64, 3/64 and 4/64, respectively. W_H is the work function of the corresponding heterostructure. E_{vac} represents the vacuum level. Orange, gray, blue, and red spheres stand for N, C, Sn, and Se atoms, respectively.

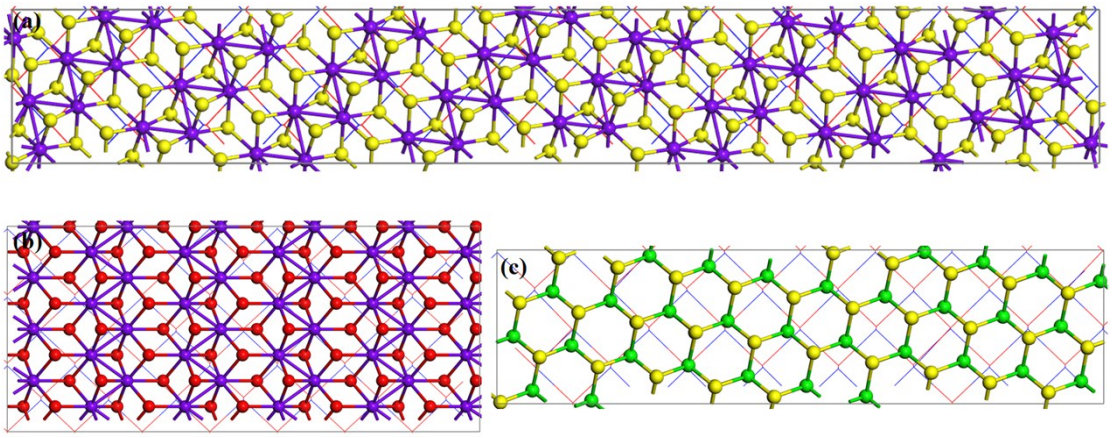
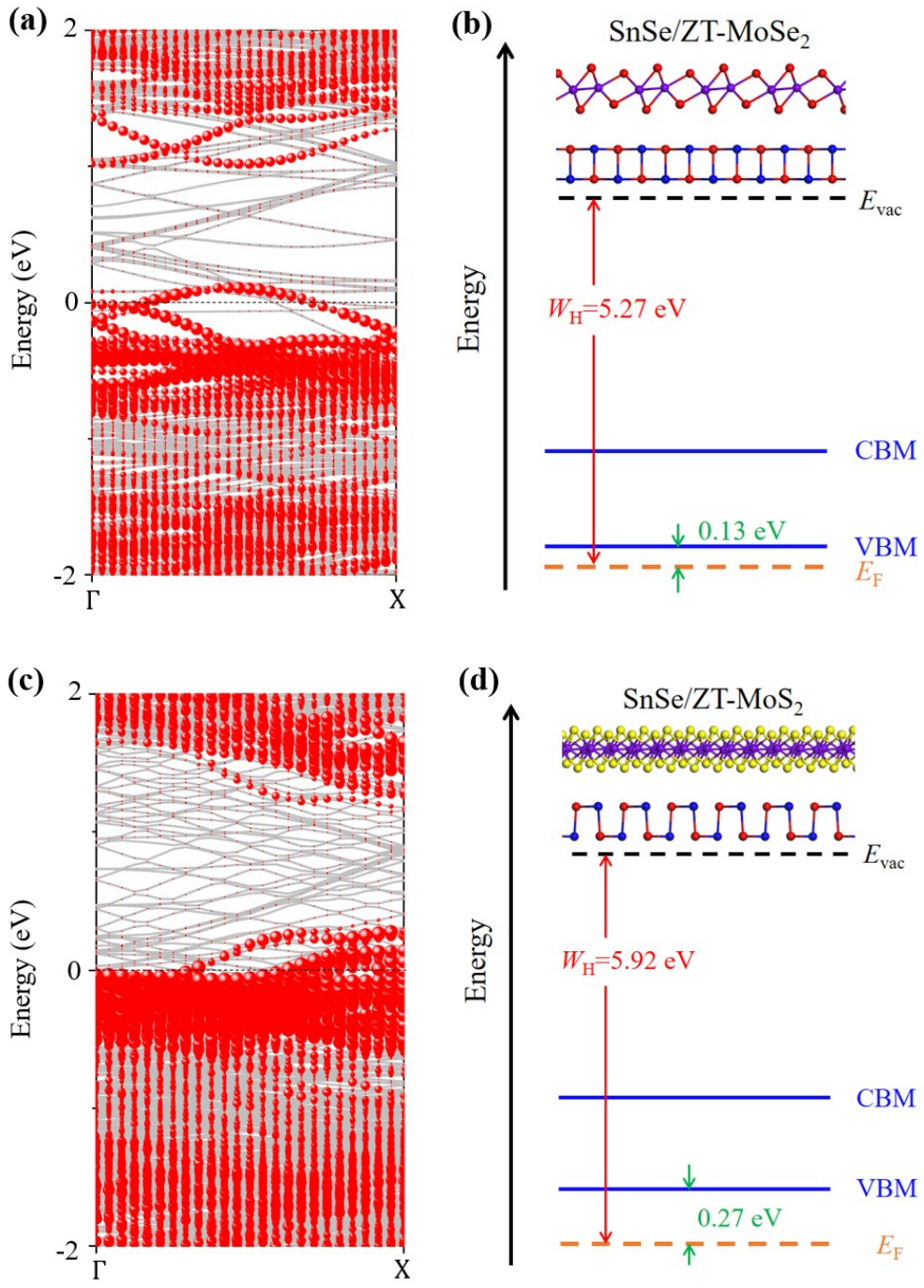


Fig. S5 Optimized geometrical structure of the supercell for (a) SnSe/ ZT-MoS_2, (b) SnSe/ ZT-$MoSe_2$, and (c) SnSe/ H-NbS_2 heterostructure. Blue, red, violet, yellow and green spheres stand for Sn, Se, Mo, S, and Nb atoms, respectively


(a)
2
0
-2
Energy (eV)
Γ
X
(b)
SnSe/ZT-MoSe2
Energy
Evac
WH=5.27 eV
CBM
0.13 eV
VBM
EF
(c)
2
0
-2
Energy (eV)
Γ
X
(d)
SnSe/ZT-MoS2
Energy
Evac
WH=5.92 eV
CBM
VBM
0.27 eV
EF

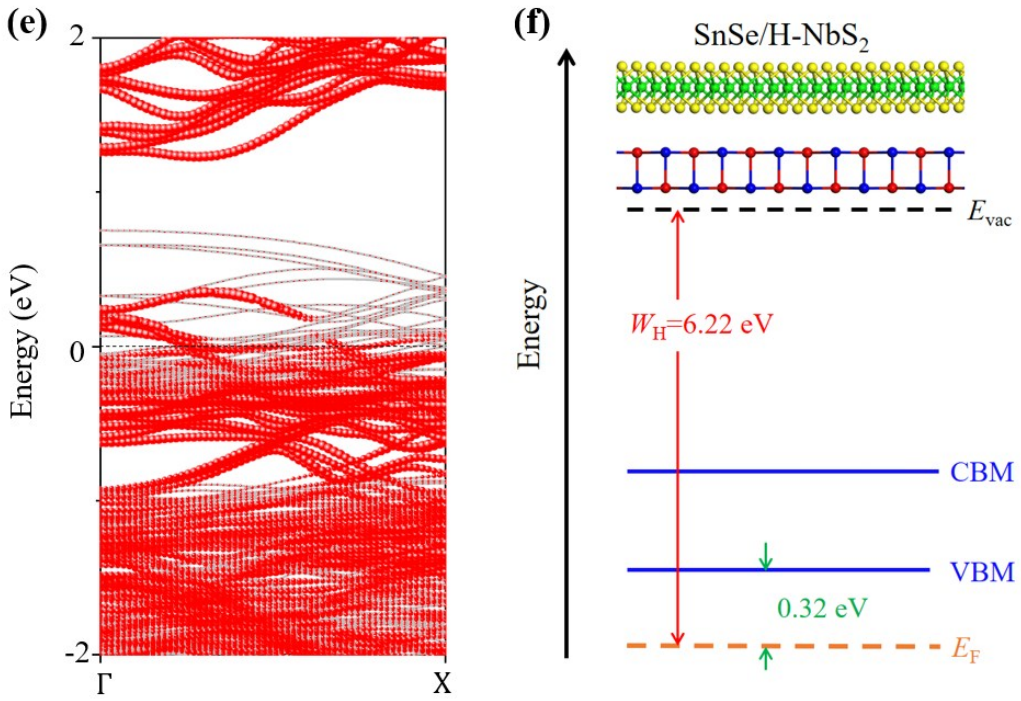


Fig. S6 (a), (c), and (e) Projected band structures on the SnSe sheet, and (b), (e), and (f) band alignments of the SnSe/ ZT-$MoSe_2$, SnSe/ ZT-MoS_2, and SnSe/ H-NbS_2 heterostructures, respectively. W_H is the work function of the corresponding heterostructure. E_{vac} represents the vacuum level. Blue, red, violet, yellow and green spheres stand for Sn, Se, Mo, S, and Nb atoms, respectively.

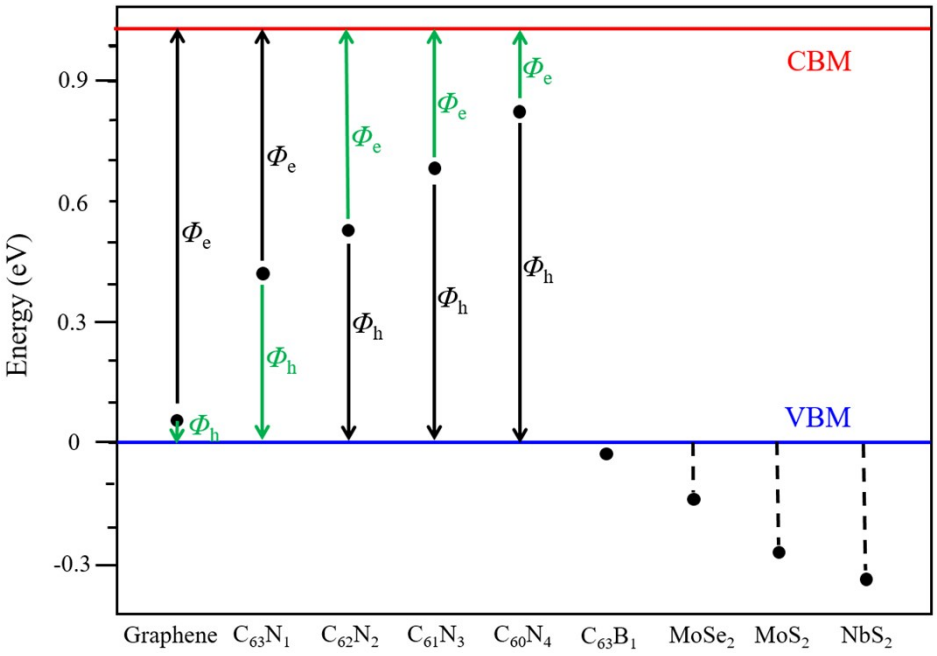


Fig. S7 Schottky barrier heights for the heterostructures of the single-layer SnSe sheet contacting with the 2D monolayers as the electrode, respectively. The red and blue lines represent the CBM and VBM of SnSe, which are the band edges of the heterojunctions. Black dots represent the Fermi levels of the corresponding heterojunctions. Φ_e and Φ_h stand for the Schottky barriers of electrons and holes, respectively. VBM is set to zero. Ohmic contacts are achieved in the heterostructures with the Fermi levels (black dots) below the CBM (the dash lines denote the Ohmic contacts).

Table S3 Lattice parameters (u and v, in Å) and the angle (γ, in degree) between them for SnSe/ ZT-$MoSe_2$, SnSe/ ZT-MoS_2, and SnSe/ H-NbS_2 supercells. The last column shows the total number of atoms in these heterostructures.

Heterostructure	u	*v*	γ	Number of atoms
SnSe/ *ZT-$MoSe_2$*	13.10	30.10	90.00	204
SnSe/ *ZT-MoS_2*	8.60	61.00	90.00	286
SnSe/ *H-NbS_2*	8.70	34.40	90.00	157