Supplemental Materials for
Simple linear response model for predicting energy band alignment of two-dimensional vertical heterostructures

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Supplementary Note

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SUPPLEMENTARY NOTE

In order to calculate vacuum dipole step in the DFT, we extract local electrostatic potential energy across the supercell. Fig. S1 shows the potential energy of the SiSe-SnS heterostructure as an example, where its supercell is shown on the top of the plot. The vacuum energy for each layer is reached within a few Å away from the corresponding layer. Here vacuum energies are 2.24 eV for the SiSe and 2.02 eV for the SnS layer, which results in a vacuum dipole step of $eV_h = 0.22\,\text{eV}$.

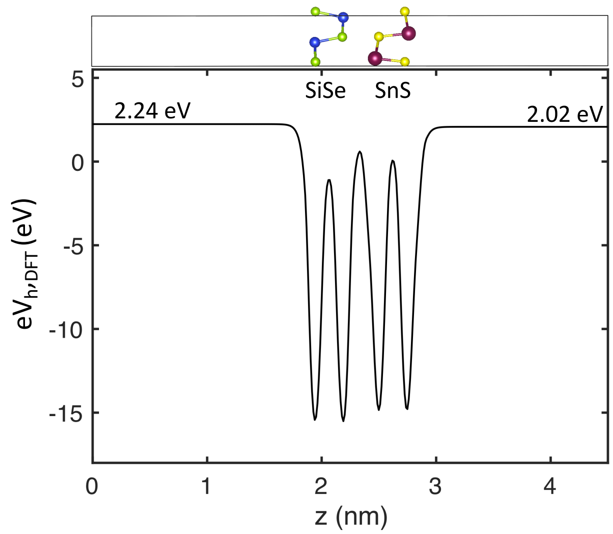

Figure S1. Electrostatic potential energy of SiSe-SnS heterostructure. The vacuum dipole step, $eV_{h,\mathrm{DFT}}$, is obtained by the difference between the vacuum energy on the SiSe side (2.24 eV) and on the SnS side (2.02 eV) within the supercell, that is 0.22 eV for this case.

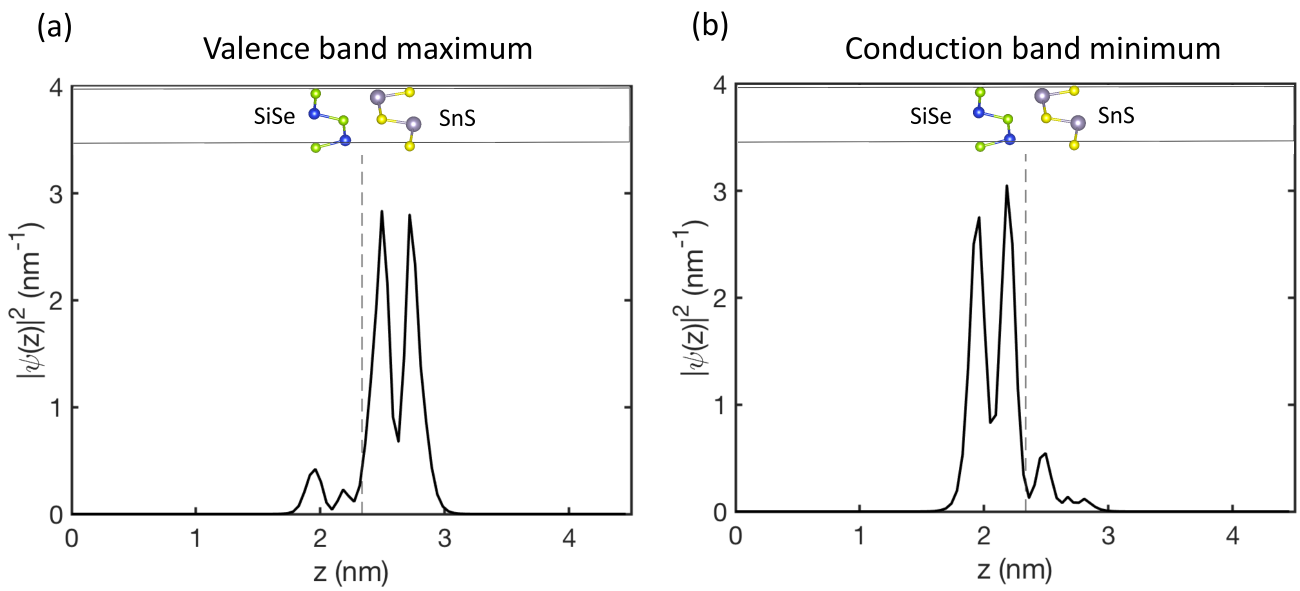

Figure S2. (a) Wave function projection at valence band maximum, showing weak band hybridization with 10% contribution from SiSe, and 90% from SnS layer. (b) Wave function projection at conduction band minimum, showing strong band hybridization with 87% contribution from SiSe, and 13% contribution from SnS layer. Vertical dashed lines shows the interlayer region. Unit cell is shown in the inset.

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ML 1	ML 2	a (Å)	b (Å)	d (Å)	E_{v1}	E_{v2}	E_{c1}	E_{c2}	eV_h	$E_{g,A}$	$E_{g,m}$	E_g	$E_{g,\mathrm{DFT}}$	$E_{g,\mathrm{HSE}}$
GeS	GeTe	4.47	3.98	2.90	-5.18	-3.74	-3.51	-3.55	0.23	0.19	0.19	0.19	0.21	1.16
SiSe	GeS	4.28	3.74	2.71	-4.42	-4.89	-3.63	-3.16	0.02	0.79	0.79	0.79	0.79	0.96
SiTe	GeTe	4.35	4.22	3.05	-4.22	-4.23	-3.59	-3.34	0.05	0.63	0.63	0.63	0.66	0.60
SnS	GeTe	4.36	4.22	3.01	-4.89	-4.29	-3.26	-3.36	0.11	0.93	0.93	0.93	0.85	1.16
SnS	SiTe	4.36	4.10	2.91	-4.73	-3.93	-3.07	-3.41	0.06	0.52	0.52	0.52	0.47	0.60
SiSe	SiTe	4.30	4.10	2.87	-5.04	-3.88	-3.95	-3.37	0.33	-0.07	0.51	0.29	0.12	0.55
SiSe	SnTe	4.65	4.55	2.52	-5.47	-4.21	-4.24	-3.27	0.23	-0.03	0.94	0.43	0.57	0.96
GeS	GeSe	4.24	3.97	2.79	-5.10	-4.61	-3.44	-3.37	0.10	1.16	1.23	1.23	1.04	1.53
GeS	SiTe	4.30	4.10	2.88	-5.25	-3.86	-3.64	-3.36	0.26	0.22	0.50	0.50	0.27	0.60
GeSe	GeTe	4.35	4.22	3.02	-5.14	-4.27	-3.72	-3.37	0.19	0.55	0.91	0.80	0.58	1.16
SiSe	GeSe	4.28	3.97	2.84	-4.84	-4.57	-3.81	-3.32	0.04	0.76	1.03	0.91	0.90	0.96
GeSe	SiTe	4.30	4.10	2.99	-4.87	-3.83	-3.55	-3.36	0.17	0.28	0.47	0.47	0.26	0.60
SiSe	GeTe	4.35	4.22	2.86	-5.17	-4.26	-4.09	-3.33	0.31	0.16	0.92	0.51	0.66	0.96
GeS	SnS	4.36	4.05	2.69	-5.25	-4.66	-3.63	-3.09	0.17	1.03	1.56	1.26	1.23	2.09
GeSe	SnS	4.36	4.05	2.75	-4.88	-4.66	-3.53	-3.08	0.07	1.13	1.35	1.26	1.15	1.53
SiSe	SnS	4.36	4.05	2.63	-5.05	-4.64	-3.97	-3.08	0.22	0.67	1.08	0.94	0.93	0.96
SnS	SnSe	4.49	4.27	2.61	-4.96	-4.53	-3.31	-3.27	0.10	1.22	1.26	1.26	1.20	1.44
SnS	SnTe	4.65	4.55	2.96	-5.21	-4.32	-3.59	-3.31	0.20	0.72	1.01	0.97	0.84	1.13
GeS	SnSe	4.49	4.27	2.67	-5.41	-4.47	-3.82	-3.26	0.26	0.65	1.21	0.96	1.00	1.44
GeSe	SnSe	4.49	4.27	2.77	-5.16	-4.47	-3.80	-3.26	0.14	0.67	1.20	0.92	0.90	1.44
GeTe	SnSe	4.49	4.27	2.99	-4.47	-4.45	-3.44	-3.29	0.06	1.01	1.03	1.03	0.81	1.14
SiSe	SnSe	4.49	4.27	2.60	-5.23	-4.48	-4.18	-3.23	0.31	0.30	1.05	0.65	0.79	0.96
SiTe	SnSe	4.49	4.27	2.88	-4.53	-4.46	-3.75	-3.27	0.03	0.71	0.78	0.78	0.65	0.60
SnSe	SnTe	4.65	4.55	3.07	-5.01	-4.30	-3.61	-3.34	0.14	0.69	0.96	0.89	0.75	1.13
GeS	SnTe	4.65	4.55	2.62	-5.62	-4.20	-3.87	-3.27	0.41	0.33	0.93	0.74	0.73	1.13
GeSe	SnTe	4.65	4.55	2.81	-5.38	-4.26	-3.97	-3.24	0.27	0.28	1.01	0.66	0.61	1.13
GeTe	SnTe	4.65	4.55	3.14	-4.96	-4.23	-3.81	-3.33	0.11	0.42	0.90	0.67	0.64	1.06
SiTe	SnTe	4.65	4.55	2.88	-4.91	-4.26	-4.11	-3.28	0.23	0.15	0.80	0.45	0.57	0.60

Table I. Relevant lattice constants (a and b), heterostructure interlayer distance (d), conduction band minimum (E_c), valence band maximum (E_v), DFT calculated vacuum dipole step of heterostructures (V_h), predicted bandgaps of heterostructures based on Anderson model ($E_{g,A}$), midgap model ($E_{g,m}$), linear response model (E_g), DFT calculated bandgaps ($E_{g,\mathrm{DFT}}$) for strained heterostrcutures, and HSE calculated bandgaps for unstrained heterostructures ($E_{g,\mathrm{HSE}}$) for group IV-VI heterostructures. All energies are in eV. Band edges of monolayers are computed according to the Anderson model.

Material	a (Å)	b (Å)	$E_{g,\mathrm{DFT}}$	$E_{g,\mathrm{HSE}}$
SiS	4.58	3.34	1.31	2.07
SiSe	4.28	3.74	0.50	0.96
SiTe	4.30	4.10	0.34	0.60
GeS	4.24	3.71	1.50	2.18
GeSe	4.22	3.97	1.06	1.53
GeTe	4.35	4.22	0.77	1.16
SnS	4.36	4.05	1.55	2.09
SnSe	4.49	4.27	1.04	1.44
SnTe	4.65	4.55	0.69	1.13

Table II. Relaxed lattice constants of group IV-VI monolayers and their bandgap values (in eV) based on DFT and HSE calculations.