

**Supporting Information for  
Lateral and Vertical Heterostructures of Transition Metal Dichalcogenides**

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Here, we provide

1. The cohesive energies  $E_c$  of 2D and 3D  $\text{MX}_2$  structures, calculated according to Eq. (1).
2. The formation energies  $E_f$  of 2D and 3D  $\text{MX}_2$  structures, calculated according to Eq. (2).
3. The formation energies  $E'_f$  of 2D  $\text{MX}_2$  structures relative to the parent 3D bulk crystals, calculated according to Eq. (3).
4. The average cohesive energies  $\bar{E}_c$  of lateral and vertical  $(\text{MoTe}_2)_p/(\text{NiTe}_2)_q$  heterostructures, calculated according to Eq. (4).
5. The average formation energies  $\bar{E}_f$  of lateral and vertical  $(\text{MoTe}_2)_p/(\text{NiTe}_2)_q$  heterostructures, calculated according to Eq. (5).

in the following tables:

- Table S1 for SL 2D, 2L, 3L, and 3D parent-crystals of  $\text{MoTe}_2$  and  $\text{NiTe}_2$ .
- Table S2 for Armchair lateral heterostructures of  $\text{A}:(\text{MoTe}_2)_p/(\text{NiTe}_2)_q$ .
- Table S3 for Vertical heterostructures of  $\text{V}:(\text{MoTe}_2)_p/(\text{NiTe}_2)_q$ .

**The cohesive energy  $E_c(MX_2)$  per formula unit is given by**

$$E_c(MX_2) = E_{\text{atom}}(\text{M}) + 2E_{\text{atom}}(\text{X}) - E(MX_2), \quad (1)$$

where  $E_{\text{atom}}(\text{M})$  and  $E_{\text{atom}}(\text{X})$ , denote the total energies of M and X atoms, respectively, and  $E(MX_2)$  is the total energy of  $\text{MX}_2$ .

**The formation energy  $E_f(MX_2)$  per formula unit is given by**

$$E_f(MX_2) = E_c(MX_2) - E_{\text{bulk}}(\text{M}) - 2E_{\text{bulk}}(\text{X}) \quad (2)$$

where  $E_{\text{bulk}}(\text{M})$  and  $E_{\text{bulk}}(\text{X})$ , denote the cohesive energies of constituent elements M and X in their equilibrium bulk phases, respectively, and  $E_c(MX_2)$  is the cohesive energy of  $\text{MX}_2$ .

**The formation energy  $E'_f(MX_2)$  per formula unit relative to the parent 3D crystals given by**

$$E'_f(MX_2) = E_c(3\text{D } MX_2) - E_c(2\text{D } MX_2) \quad (3)$$

where  $E_c(3\text{D } MX_2)$  denotes cohesive energy of parent 3D crystal and  $E_c(2\text{D } MX_2)$  is cohesive energy of the 2D SL, 2L or 3L of  $\text{MX}_2$ .

**The average cohesive energies  $\bar{E}_c(p/q)$  per Mo(Ni)-Te pair of 2D lateral A:(p/q) and 3D vertical V:(p/q) heterostructures is given by**

$$\bar{E}_c(p/q) = [pE_{\text{atom}}(\text{Mo}) + qE_{\text{atom}}(\text{Ni}) + 2(p+q)E_{\text{atom}}(\text{Te}) - E(p/q)]/(p+q) \quad (4)$$

where  $E_{\text{atom}}(\text{Mo})$ ,  $E_{\text{atom}}(\text{Ni})$  and  $E_{\text{atom}}(\text{Te})$  denote the total energies of Mo, Ni and Te atoms, respectively, and  $E(p/q)$  is the total energy of  $(\text{MoTe}_2)_p/(\text{NiTe}_2)_q$  lateral or vertical heterostructure.

**The average formation energy  $\bar{E}_f(p/q)$  per Mo(Ni)-Te pair of 2D lateral A:(p/q) and 3D vertical V:(p/q) heterostructures is given by**

$$\bar{E}_f(p/q) = \bar{E}_c(p/q) - [pE_c(3\text{D } \text{MoTe}_2) + qE_c(3\text{D } \text{NiTe}_2)]/(p+q) \quad (5)$$

where  $E_c(3\text{D } \text{MoTe}_2)$  and  $E_c(3\text{D } \text{NiTe}_2)$  denote the cohesive energies of 3D bulk  $\text{MoTe}_2$  and  $\text{NiTe}_2$  crystals, respectively, and  $\bar{E}_c(p/q)$  is the average cohesive energy of the composite structure.

TABLE S1. The values of  $E_c$  (eV/formula unit),  $E_f$  (eV/formula unit),  $E'_f$  (eV/formula unit), the direct ( $d$ ) or indirect ( $i$ ) band gap  $E_g$  (eV), the lattice constants  $a = b$  and  $c$  (Å), the Mo(Ni)-Te bond lengths  $d_{M-X}$  (Å), the layer thickness  $d_t$  (Å), the interlayer spacing  $d_{int}$  (Å) for 2D [single-layer (SL), bilayer (2L), trilayer (3L)] and 3D crystals. The k-meshes used in structural optimizations (str. opt.) and DOS calculations are given in columns 11 and 12, respectively.

	$E_c$	$E_f$	$E'_f$	$E_g$	$a = b$	$c$	$d_{M-X}$	$d_t$	$d_{int}$	k-mesh (str. opt.)	k-mesh (DOS)
3D MoTe <sub>2</sub> (LDA)	15.822	0.917		0.884 <sup><i>i</i></sup>	3.469	13.738	2.692	3.597	3.272	37×37×15	47×47×11
3D MoTe <sub>2</sub> (PBE)	13.117	1.770		0.751 <sup><i>i</i></sup>	3.515	14.076	2.718	3.617	3.421	37×37×15	47×47×11
3L MoTe <sub>2</sub> (PBE)	13.003	1.656	0.114	0.834 <sup><i>i</i></sup>	3.516		2.720	3.620	3.367	37×37×1	47×47×1
2L MoTe <sub>2</sub> (PBE)	12.947	1.599	0.171	0.943 <sup><i>i</i></sup>	3.519		2.720	3.617	3.367	37×37×1	47×47×1
2D MoTe <sub>2</sub> (LDA)	15.661	0.756	0.160	1.253 <sup><i>d</i></sup>	3.466		2.688	3.589		37×37×1	47×47×1
2D MoTe <sub>2</sub> (PBE)	12.789	1.442	0.328	1.145 <sup><i>d</i></sup>	3.519		2.722	3.623		37×37×1	47×47×1
3D NiTe <sub>2</sub> (LDA)	13.152	0.483			3.619	11.608	2.551	2.928	2.876	37×37×15	47×47×11
3D NiTe <sub>2</sub> (PBE)	11.009	1.243			3.686	11.703	2.592	2.960	2.892	37×37×15	47×47×11
2D NiTe <sub>2</sub> (LDA)	12.795	0.125	0.357		3.589		2.535	2.918		37×37×1	47×47×1
2D NiTe <sub>2</sub> (PBE)	10.510	0.744	0.499		3.672		2.585	2.959		37×37×1	47×47×1

TABLE S2. The values of lateral heterostructures, A:(MoTe<sub>2</sub>)<sub>*p*</sub>/(NiTe<sub>2</sub>)<sub>*q*</sub> for  $1 \leq p = q \leq 10$ . The optimized values of semiconducting MoTe<sub>2</sub> (*S*) and metallic NiTe<sub>2</sub> (*M*) stripes' layer thicknesses  $d_t^S$  and  $d_t^M$  (Å), Mo–Te and Ni–Te bond lengths  $d_{Mo-Te}$  and  $d_{Ni-Te}$  (Å), in interface (*i*) and bulk region (*b*). The k-meshes used in structural optimizations (str. opt.) and DOS calculations are given in columns 6 and 7, respectively.

Composite	$d_t^S$	$d_t^M$	$d_{Mo-Te}$	$d_{Ni-Te}$	k-mesh (str. opt.)	k-mesh (DOS)
A:(MoTe <sub>2</sub> ) <sub>1</sub> /(NiTe <sub>2</sub> ) <sub>1</sub>	3.751	3.187	2.653	2.595	41×21×1	41×41×1
A:(MoTe <sub>2</sub> ) <sub>2</sub> /(NiTe <sub>2</sub> ) <sub>2</sub>	3.644	2.906	2.701	2.548	19×21×1	39×41×1
A:(MoTe <sub>2</sub> ) <sub>3</sub> /(NiTe <sub>2</sub> ) <sub>3</sub>	3.329 <sup><i>i</i></sup>	3.098 <sup><i>i</i></sup>	2.713	2.534	13×21×1	27×41×1
	4.140 <sup><i>b</i></sup>	2.807 <sup><i>b</i></sup>				
A:(MoTe <sub>2</sub> ) <sub>4</sub> /(NiTe <sub>2</sub> ) <sub>4</sub>	3.039 <sup><i>i</i></sup>	3.134 <sup><i>i</i></sup>	2.724 <sup><i>i</i></sup>	2.536 <sup><i>i</i></sup>	9×21×1	19×41×1
	3.906 <sup><i>b</i></sup>	2.864 <sup><i>b</i></sup>	2.707 <sup><i>b</i></sup>	2.542 <sup><i>b</i></sup>		
A:(MoTe <sub>2</sub> ) <sub>5</sub> /(NiTe <sub>2</sub> ) <sub>5</sub>	3.427 <sup><i>i</i></sup>	3.119 <sup><i>i</i></sup>	2.704 <sup><i>i</i></sup>	2.522 <sup><i>i</i></sup>	7×21×1	15×41×1
	3.624 <sup><i>b</i></sup>	2.948 <sup><i>b</i></sup>	2.714 <sup><i>b</i></sup>	2.555 <sup><i>b</i></sup>		
A:(MoTe <sub>2</sub> ) <sub>6</sub> /(NiTe <sub>2</sub> ) <sub>6</sub>	3.310 <sup><i>i</i></sup>	3.071 <sup><i>i</i></sup>	2.707 <sup><i>i</i></sup>	2.533 <sup><i>i</i></sup>	6×21×1	13×41×1
	3.668 <sup><i>b</i></sup>	2.923 <sup><i>b</i></sup>	2.705 <sup><i>b</i></sup>	2.541 <sup><i>b</i></sup>		
A:(MoTe <sub>2</sub> ) <sub>7</sub> /(NiTe <sub>2</sub> ) <sub>7</sub>	3.113 <sup><i>i</i></sup>	3.176 <sup><i>i</i></sup>	2.715 <sup><i>i</i></sup>	2.508 <sup><i>i</i></sup>	6×21×1	13×41×1
	3.620 <sup><i>b</i></sup>	2.976 <sup><i>b</i></sup>	2.683 <sup><i>b</i></sup>	2.534 <sup><i>b</i></sup>		
A:(MoTe <sub>2</sub> ) <sub>8</sub> /(NiTe <sub>2</sub> ) <sub>8</sub>	3.396 <sup><i>i</i></sup>	3.135 <sup><i>i</i></sup>	2.701 <sup><i>i</i></sup>	2.496 <sup><i>i</i></sup>	6×21×1	13×41×1
	3.631 <sup><i>b</i></sup>	3.025 <sup><i>b</i></sup>	2.692 <sup><i>b</i></sup>	2.515 <sup><i>b</i></sup>		
A:(MoTe <sub>2</sub> ) <sub>9</sub> /(NiTe <sub>2</sub> ) <sub>9</sub>	3.323 <sup><i>i</i></sup>	3.132 <sup><i>i</i></sup>	2.705 <sup><i>i</i></sup>	2.511 <sup><i>i</i></sup>	4×20×1	9×40×1
	3.620 <sup><i>b</i></sup>	3.022 <sup><i>b</i></sup>	2.687 <sup><i>b</i></sup>	2.527 <sup><i>b</i></sup>		
A:(MoTe <sub>2</sub> ) <sub>10</sub> /(NiTe <sub>2</sub> ) <sub>10</sub>	3.344 <sup><i>i</i></sup>	3.153 <sup><i>i</i></sup>	2.704 <sup><i>i</i></sup>	2.492 <sup><i>i</i></sup>	4×21×1	9×41×1
	3.607 <sup><i>b</i></sup>	3.024 <sup><i>b</i></sup>	2.693 <sup><i>b</i></sup>	2.518 <sup><i>b</i></sup>		

TABLE S3. The values of vertical heterostructures,  $V:(\text{MoTe}_2)_p/(\text{NiTe}_2)_q$  for  $1 \leq p = q \leq 5$ . The optimized values of semiconducting  $\text{MoTe}_2$  ( $S$ ) and metallic  $\text{NiTe}_2$  ( $M$ ) layer thicknesses  $d_t^S$  and  $d_t^M$  (Å), Mo–Te and Ni–Te bond lengths  $d_{\text{Mo}-\text{Te}}$  and  $d_{\text{Ni}-\text{Te}}$  (Å),  $\text{MoTe}_2-\text{NiTe}_2$ ,  $\text{MoTe}_2-\text{MoTe}_2$  and  $\text{NiTe}_2-\text{NiTe}_2$  interlayer spacings  $d_{int}^{S-M}$ ,  $d_{int}^{S-S}$  and  $d_{int}^{M-M}$  (Å), respectively, in interface ( $i$ ) and bulk region ( $b$ ). The k-meshes used in structural optimizations (str. opt.) and DOS calculations are given in columns 9 and 10, respectively.

Composite	$d_t^S$	$d_t^M$	$d_{\text{Mo}-\text{Te}}$	$d_{\text{Ni}-\text{Te}}$	$d_{int}^{S-M}$	$d_{int}^{S-S}$	$d_{int}^{M-M}$	k-mesh (str. opt.)	k-mesh (DOS)
$V:(\text{MoTe}_2)_1/(\text{NiTe}_2)_1$	3.561	3.019	2.732	2.563	3.108			$37 \times 37 \times 17$	$37 \times 37 \times 17$
$V:(\text{MoTe}_2)_2/(\text{NiTe}_2)_2$	3.568	3.052	2.731	2.565	3.227	3.337	2.880	$37 \times 37 \times 8$	$37 \times 37 \times 8$
$V:(\text{MoTe}_2)_3/(\text{NiTe}_2)_3$	3.568 <sup>i</sup>	3.051 <sup>i</sup>	2.730 <sup>i</sup>	2.575 <sup>i</sup>	3.271	3.292	2.940	$37 \times 37 \times 6$	$37 \times 37 \times 6$
	3.564 <sup>b</sup>	3.077 <sup>b</sup>	2.729 <sup>b</sup>	2.576 <sup>b</sup>					
$V:(\text{MoTe}_2)_4/(\text{NiTe}_2)_4$	3.567 <sup>i</sup>	3.037 <sup>i</sup>	2.732 <sup>i</sup>	2.570 <sup>i</sup>	3.203	3.347 <sup>i</sup>	2.909 <sup>i</sup>	$37 \times 37 \times 4$	$37 \times 37 \times 4$
	3.565 <sup>b</sup>	3.059 <sup>b</sup>	2.731 <sup>b</sup>	2.574 <sup>b</sup>		3.339 <sup>b</sup>	2.949 <sup>b</sup>		
$V:(\text{MoTe}_2)_5/(\text{NiTe}_2)_1$	3.611 <sup>i</sup>	3.112	2.722 <sup>i</sup>	2.563	3.306	3.380 <sup>i</sup>		$37 \times 37 \times 6$	$37 \times 37 \times 6$
	3.608 <sup>b</sup>		2.721 <sup>b</sup>			3.330 <sup>b</sup>			
$V:(\text{MoTe}_2)_1/(\text{NiTe}_2)_5$	3.516	2.969 <sup>i</sup>	2.745	2.578 <sup>i</sup>	3.105		2.883 <sup>i</sup>	$37 \times 37 \times 6$	$37 \times 37 \times 6$
		2.993 <sup>b</sup>		2.587 <sup>b</sup>			2.915 <sup>b</sup>		