

Supplementary Information

Contact engineering for 2D Janus MoSSe/metal junctions

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Table S1 The calculated relative total energies ΔE_{tot} (10^{-3} eV) of the six possible configurations for Nb₂CT₂-MoSSe contacts. (Total energy of the most stable configuration is set as the reference of zero energy for each configuration)

System	Configuration	ΔE_{tot}	System	Configuration	ΔE_{tot}
Nb ₂ C/SeMoS	I	203.39	Nb ₂ C/SMoSe	I	373.96
	II	255.53		II	59.64
	III	10.10		III	196.26
	IV	0.00		IV	159.48
	V	274.83		V	0.00
	VI	166.30		VI	376.29
Nb ₂ CF ₂ /SeMoS	I	68.229	Nb ₂ CF ₂ /SMoSe	I	74.82
	II	6.59		II	1.21
	III	7.20		III	6.37
	IV	0.00		IV	0.00
	V	68.99		V	13.01
	VI	5.64		VI	75.28
Nb ₂ CO ₂ /SeMoS	I	111.65	Nb ₂ CO ₂ /SMoSe	I	127.48
	II	4.57		II	16.20
	III	6.11		III	0.00
	IV	6.80		IV	13.96
	V	70.45		V	85.44
	VI	0.00		VI	19.42
Nb ₂ C(OH) ₂ /SeMoS	I	19.12	Nb ₂ C(OH) ₂ /SMoSe	I	0.00
	II	11.64		II	77.859
	III	0.00		III	66.985
	IV	3.60		IV	67.971
	V	61.03		V	43.825
	VI	21.25		VI	73.215

Table S2 The calculated relative total energies ΔE_{tot} (10^{-3} eV) of the six possible configurations for $\text{Nb}_3\text{C}_2\text{T}_2$ -MoSSe contacts. (Total energy of the most stable configuration is set as the reference of zero energy for each configuration)

System	Configuration	ΔE_{tot} (	System	Configuration	ΔE_{tot}
$\text{Nb}_3\text{C}_2/\text{SeMoS}$	I	491.68	$\text{Nb}_3\text{C}_2/\text{SMoSe}$	I	160.33
	II	279.30		II	508.80
	III	401.64		III	0.00
	IV	358.71		IV	443.32
	V	0.00		V	122.76
	VI	419.92		VI	125.61
$\text{Nb}_3\text{C}_2\text{F}_2/\text{SeMoS}$	I	16.67	$\text{Nb}_3\text{C}_2\text{F}_2/\text{SMoSe}$	I	21.41
	II	59.86		II	65.53
	III	6.85		III	7.98
	IV	3.97		IV	10.20
	V	0.00		V	0.00
	VI	60.77		VI	66.18
$\text{Nb}_3\text{C}_2\text{O}_2/\text{SeMoS}$	I	33.57	$\text{Nb}_3\text{C}_2\text{O}_2/\text{SMoSe}$	I	39.51
	II	70.33		II	0.00
	III	5.46		III	74.74
	IV	0.00		IV	2.96
	V	6.12		V	74.97
	VI	71.37		VI	4.46
$\text{Nb}_3\text{C}_2(\text{OH})_2/\text{SeMoS}$	I	64.56	$\text{Nb}_3\text{C}_2(\text{OH})_2/\text{SMoSe}$	I	70.58
	II	22.27		II	0.00
	III	17.45		III	39.32
	IV	28.56		IV	37.41
	V	0.00		V	24.22
	VI	27.40		VI	5.97

Table S3 The calculated relative total energies ΔE_{tot} (10^{-3} eV) of the six possible configurations for $\text{MX}_2\text{-MoSSe}$ contacts. (Total energy of the most stable configuration is set as the reference of zero energy for each configuration)

System	Configuration	ΔE_{tot}	System	Configuration	ΔE_{tot}
VS_2/SeMoS	I	67.03	VS_2/SMoSe	I	71.95
	II	0.28		II	0.00
	III	1.54		III	2.71
	IV	4.28		IV	3.92
	V	0.00		V	7.71
	VI	59.47		VI	68.72
$\text{NbS}_2/\text{SeMoS}$	I	91.57	$\text{NbS}_2/\text{SMoSe}$	I	80.30
	II	0.00		II	0.34
	III	14.02		III	0.00
	IV	9.65		IV	13.306
	V	14.05		V	1.53
	VI	1.05		VI	76.586
$\text{TaS}_2/\text{SeMoS}$	I	83.47	$\text{TaS}_2/\text{SMoSe}$	I	75.06
	II	0.00		II	0.00
	III	8.96		III	3.97
	IV	13.95		IV	1.66
	V	7.39		V	10.04
	VI	79.75		VI	71.82
$\text{VSe}_2/\text{SeMoS}$	I	82.07	$\text{VSe}_2/\text{SMoSe}$	I	82.01
	II	0.00		II	2.78
	III	5.61		III	0.00
	IV	16.33		IV	7.46
	V	2.79		V	8.21
	VI	78.10		VI	78.09

Table S4 Tunneling barrier height Φ_{TB} , width w_{TB} and comprehensive factor C of 2D metal-MoSSe contacts

2D metal	Φ_{TB} (eV)		w_{TB} (Å)		C (eV·Å ²)	
	Se	S	Se	S	Se	S
Nb ₂ C	1.46	0	0.67	0	0.66	0
Nb ₂ CF ₂	3.58	3.57	1.42	1.34	7.17	6.45
Nb ₂ CO ₂	4.60	4.86	1.55	1.57	11.01	11.92
Nb ₂ COH ₂	1.20	1.88	0.87	1.08	0.78	2.18
Nb ₃ C ₂	1.42	0	0.64	0	0.58	0
Nb ₃ C ₂ F ₂	4.02	4.08	1.49	1.48	8.93	8.94
Nb ₃ C ₂ O ₂	4.50	4.63	1.53	1.52	10.52	10.76
Nb ₃ C ₂ (OH) ₂	1.10	1.81	0.82	1.10	0.74	2.19
VS ₂	4.80	4.82	1.60	1.50	12.32	10.91
NbS ₂	4.50	4.73	1.37	1.49	8.41	10.48
TaS ₂	4.53	4.84	1.52	1.58	10.44	12.03
VSe ₂	4.15	4.32	1.50	1.50	9.38	9.71

Table S5 Electronic SBH Φ_n (eV), hole SBH Φ_p (eV), interface dipole D_{int} (Debye), tot dipole D_{tot} (Debye), potential step ΔV (eV), the difference between the metal and MSJs work functions ΔW (eV), metal work functions W_M (eV) of 2D metal-SeMoS contacts (ML, BL, and TL represent monolayer, bilayer, and trilayer, respectively)

	Metal	Φ_n	Φ_p	D_{int}	D_{tot}	ΔV	ΔW	W_M
ML-MoSSe	Nb ₂ CF ₂	0.77	0.86	-0.25	-0.48	-0.70	0.69	4.37
	Nb ₂ CO ₂	1.57	0.03	-0.08	-0.31	-0.22	0.11	5.81
	Nb ₂ COH ₂	-0.10	1.52	-1.13	-1.37	-2.41	2.33	2.22
	Nb ₃ C ₂ F ₂	1.03	0.50	-0.28	-0.51	-0.76	0.62	4.89
	Nb ₃ C ₂ O ₂	1.59	0.05	-0.21	-0.44	-0.51	0.44	5.48
	Nb ₃ C ₂ (OH) ₂	-0.33	1.49	-1.20	-1.43	-2.42	2.45	2.10
	VS ₂	1.48	0.06	0.05	-0.18	0.12	0.09	6.02
	NbS ₂	1.25	0.08	0.08	-0.15	0.18	-0.01	6.08
	TaS ₂	1.21	0.11	0.02	-0.21	0.04	0.01	5.92
	VSe ₂	0.92	0.50	-0.15	-0.38	-0.25	0.26	5.46
	Nb ₂ CF ₂	0.63	0.27	-0.57	-1.04	-1.47	1.44	4.37
	Nb ₂ CO ₂	1.38	0.04	-0.15	-0.61	-0.30	0.13	5.81
BL-MoSSe	Nb ₂ COH ₂	-0.09	0.78	-1.62	-2.08	-3.62	3.64	2.22
	Nb ₃ C ₂ F ₂	0.72	0.26	-0.58	-1.04	-1.28	0.98	4.89
	Nb ₃ C ₂ O ₂	1.12	0.02	-0.29	-0.75	-0.59	0.37	5.48
	Nb ₃ C ₂ (OH) ₂	-0.23	0.83	-1.70	-2.16	-2.02	3.11	2.10
	VS ₂	1.28	0.02	0.08	-0.38	0.22	0.09	6.02
	NbS ₂	0.78	0.00	0.10	-0.36	0.25	-0.03	6.08
	TaS ₂	0.77	0.01	0.13	-0.34	0.17	-0.01	5.92
	VSe ₂	0.71	0.03	-0.14	-0.60	-0.29	0.28	5.46
	Nb ₂ CF ₂	0.51	0.25	-0.77	-1.24	-2.06	1.07	4.37
	Nb ₂ CO ₂	1.17	0.02	-0.14	-0.60	-0.27	0.08	5.81
	Nb ₂ COH ₂	-0.08	0.06	-2.31	-2.77	-3.60	3.47	2.22
	Nb ₃ C ₂ F ₂	0.51	0.04	-0.73	-1.20	-1.28	0.91	4.89
TL-MoSSe	Nb ₃ C ₂ O ₂	0.95	0.03	-0.33	-0.79	-0.55	0.32	5.48
	Nb ₃ C ₂ (OH) ₂	-0.14	0.20	-2.46	-2.93	-3.83	3.74	2.10
	VS ₂	1.19	0.02	0.19	-0.27	0.23	0.11	6.02
	NbS ₂	0.85	0.00	0.15	-0.31	0.27	-0.04	6.08
	TaS ₂	0.78	0.01	0.05	-0.41	0.15	-0.01	5.92
	VSe ₂	0.65	0.02	-0.17	-0.64	-0.33	0.30	5.46

Table S6 Electronic SBH Φ_n (eV), hole SBH Φ_p (eV), interface dipole D_{int} (Debye), tot dipole D_{tot} (Debye), potential step ΔV (eV), the difference between the metal and MSJs work functions ΔW (eV), metal work functions W_M (eV) of 2D metal-SMoSe contacts (ML, BL, and TL represent monolayer, bilayer, and trilayer, respectively)

	Metal	Φ_n	Φ_p	D_{int}	D_{tot}	ΔV	ΔW	w_m
ML-MoSSe	Nb ₂ CF ₂	0.18	1.55	0.30	0.53	0.78	0.01	4.37
	Nb ₂ CO ₂	1.17	0.41	0.32	0.56	0.92	-0.10	5.81
	Nb ₂ COH ₂	-0.17	1.66	-0.80	-0.57	-1.68	1.58	2.22
	Nb ₃ C ₂ F ₂	0.33	1.21	0.31	0.54	0.67	-0.11	4.89
	Nb ₃ C ₂ O ₂	1.00	0.58	0.34	0.57	0.81	-0.07	5.48
	Nb ₃ C ₂ (OH) ₂	-0.28	1.58	-0.86	-0.63	-1.72	1.70	2.10
	VS ₂	1.12	0.47	0.47	0.70	1.22	0.12	6.02
	NbS ₂	0.66	0.46	0.49	0.72	1.20	0.03	6.08
	TaS ₂	0.59	0.57	0.42	0.65	1.03	0.02	5.92
	VSe ₂	0.43	0.63	0.33	0.57	0.84	-0.01	5.46
	Nb ₂ CF ₂	0.02	1.05	0.34	0.81	0.74	0.02	4.37
	Nb ₂ CO ₂	0.67	0.19	0.53	0.99	1.36	-0.51	5.81
BL-MoSSe	Nb ₂ COH ₂	-0.11	1.24	-0.97	-0.50	-1.74	1.61	2.22
	Nb ₃ C ₂ F ₂	0.05	1.02	0.38	0.85	0.13	-0.25	4.89
	Nb ₃ C ₂ O ₂	0.67	0.29	0.68	1.15	1.52	-0.19	5.48
	Nb ₃ C ₂ (OH) ₂	-0.20	1.45	-1.02	-0.55	-1.71	1.70	2.10
	VS ₂	0.42	0.32	0.76	1.22	2.07	0.14	6.02
	NbS ₂	0.13	0.37	0.77	1.23	1.74	0.03	6.08
	TaS ₂	0.04	0.49	0.80	1.26	1.67	0.02	5.92
	VSe ₂	0.04	0.61	0.60	1.06	1.53	0.01	5.46
	Nb ₂ CF ₂	-0.01	0.35	0.38	0.84	0.72	0.02	4.37
	Nb ₂ CO ₂	0.13	0.47	0.94	1.41	1.98	0.08	5.81
	Nb ₂ COH ₂	-0.04	0.21	-1.87	-1.41	-1.62	3.47	2.22
	Nb ₃ C ₂ F ₂	0.01	0.84	0.54	1.00	3.51	-0.32	4.89
TL-MoSSe	Nb ₃ C ₂ O ₂	0.24	0.39	0.63	1.09	1.65	-0.18	5.48
	Nb ₃ C ₂ (OH) ₂	-0.04	1.10	-1.08	-0.61	-1.63	1.66	2.10
	VS ₂	0.54	0.23	0.54	1.01	1.21	0.13	6.02
	NbS ₂	0.36	0.38	0.35	0.81	0.93	0.01	6.08
	TaS ₂	0.06	0.51	0.45	0.91	0.98	0.01	5.92
	VSe ₂	0.03	0.66	0.39	0.86	0.78	0.00	5.46

Table S7 Electronic SBH Φ_n (eV), hole SBH Φ_p (eV), interface dipole D_{int} (Debye), tot dipole D_{tot} (Debye), potential step ΔV (eV), the difference between the metal and MSJs work functions ΔW (eV), metal work functions W_M (eV) of 2D metal-SeMoS contacts under different strains

	Metal	Φ_n	Φ_p	D_{int}	D_{tot}	ΔV	ΔW	W_M
-6%	Nb ₂ CF ₂	0.58	1.00	-0.27	-0.49	-0.85	0.43	4.37
	Nb ₂ CO ₂	1.46	0.04	-0.11	-0.32	-0.37	0.39	5.81
	Nb ₂ COH ₂	-0.20	1.52	-0.48	-0.69	-2.09	2.32	2.22
	Nb ₃ C ₂ F ₂	1.11	0.33	-0.27	-0.49	-0.87	0.66	4.89
	Nb ₃ C ₂ O ₂	1.38	0.04	-0.18	-0.39	-0.48	0.32	5.48
	Nb ₃ C ₂ (OH) ₂	-0.22	1.51	-0.93	-1.15	-2.16	2.46	2.10
	VS ₂	1.40	0.13	0.01	-0.20	0.04	-0.09	6.02
	NbS ₂	1.36	0.37	0.02	-0.19	0.02	-0.12	6.08
	TaS ₂	1.34	0.40	-0.04	-0.25	-0.09	0.01	5.92
	VSe ₂	1.03	0.63	-0.12	-0.33	-0.41	0.18	5.46
-4%	Nb ₂ CF ₂	0.67	0.94	-0.27	-0.49	-0.80	0.52	4.37
	Nb ₂ CO ₂	1.51	0.04	-0.11	-0.33	-0.32	0.47	5.81
	Nb ₂ COH ₂	-0.15	1.52	-0.95	-1.16	-2.19	2.30	2.22
	Nb ₃ C ₂ F ₂	1.08	0.40	-0.28	-0.50	-0.81	0.62	4.89
	Nb ₃ C ₂ O ₂	1.45	0.04	-0.18	-0.40	-0.49	0.37	5.48
	Nb ₃ C ₂ (OH) ₂	-0.17	1.50	-1.01	-1.23	-2.20	3.79	2.10
	VS ₂	1.44	0.10	0.03	-0.19	0.07	-0.02	6.02
	NbS ₂	1.43	0.32	0.01	-0.21	0.01	-0.08	6.08
	TaS ₂	1.40	0.36	-0.04	-0.26	-0.12	0.06	5.92
	VSe ₂	1.03	0.65	-0.12	-0.33	-0.34	0.20	5.46
-2%	Nb ₂ CF ₂	0.73	0.89	-0.26	-0.49	-0.75	0.60	4.37
	Nb ₂ CO ₂	1.55	0.03	-0.09	-0.32	-0.28	0.54	5.81
	Nb ₂ COH ₂	-0.12	1.52	-1.03	-1.26	-2.29	2.30	2.22
	Nb ₃ C ₂ F ₂	1.07	0.44	-0.28	-0.51	-0.76	0.59	4.89
	Nb ₃ C ₂ O ₂	1.58	0.05	-0.20	-0.42	-0.50	0.40	5.48
	Nb ₃ C ₂ (OH) ₂	-0.12	1.49	-1.10	-1.33	-2.29	2.44	2.10
	VS ₂	1.47	0.07	0.04	-0.19	0.09	0.04	6.02
	NbS ₂	1.35	0.25	0.03	-0.20	0.07	-0.04	6.08
	TaS ₂	1.46	0.30	-0.02	-0.25	-0.06	0.04	5.92

2%	VSe ₂	1.06	0.63	-0.12	-0.35	-0.27	0.21	5.46
	Nb ₂ CF ₂	0.70	0.84	-0.24	-0.48	-0.64	0.78	4.37
	Nb ₂ CO ₂	1.58	0.04	-0.07	-0.31	-0.18	0.16	5.81
	Nb ₂ COH ₂	-0.07	1.52	-1.25	-1.49	-2.53	-2.22	2.22
	Nb ₃ C ₂ F ₂	0.97	0.56	-0.30	-0.54	-0.69	0.62	4.89
	Nb ₃ C ₂ O ₂	1.58	0.07	-0.22	-0.45	-0.51	0.47	5.48
	Nb ₃ C ₂ (OH) ₂	-0.28	1.49	-1.30	-1.54	-2.53	2.45	2.10
	VS ₂	1.01	0.04	0.09	-0.15	0.21	0.13	6.02
	NbS ₂	1.00	0.04	0.12	-0.12	0.27	0.03	6.08
	TaS ₂	0.99	0.05	0.05	-0.19	0.12	0.03	5.92
	VSe ₂	0.81	0.32	-0.13	-0.37	-0.28	0.34	5.46
4%	Nb ₂ CF ₂	0.53	0.74	-0.23	-0.48	-0.65	0.93	4.37
	Nb ₂ CO ₂	1.53	0.04	-0.05	-0.30	-0.13	0.19	5.81
	Nb ₂ COH ₂	-0.01	1.39	-1.39	-1.64	-2.71	-2.22	2.22
	Nb ₃ C ₂ F ₂	0.93	0.60	-0.27	-0.51	-0.65	0.63	4.89
	Nb ₃ C ₂ O ₂	1.43	0.07	-0.20	-0.44	-0.45	0.47	5.48
	Nb ₃ C ₂ (OH) ₂	-0.21	1.49	-1.40	-1.64	-2.64	2.48	2.10
	VS ₂	1.17	0.02	0.12	-0.12	0.29	0.16	6.02
	NbS ₂	0.77	-0.02	0.14	-0.10	0.32	0.06	6.08
	TaS ₂	0.76	0.03	0.08	-0.17	0.17	0.05	5.92
	VSe ₂	0.68	0.21	-0.11	-0.35	-0.24	0.34	5.46
6%	Nb ₂ CF ₂	0.32	0.70	-0.22	-0.47	-0.54	0.92	4.37
	Nb ₂ CO ₂	1.30	0.03	-0.01	-0.26	-0.02	0.15	5.81
	Nb ₂ COH ₂	-0.05	1.18	-1.56	-1.81	-2.94	-2.22	2.22
	Nb ₃ C ₂ F ₂	0.79	0.65	-0.29	-0.54	-0.59	0.62	4.89
	Nb ₃ C ₂ O ₂	1.19	0.04	-0.14	-0.39	-0.31	0.44	5.48
	Nb ₃ C ₂ (OH) ₂	-0.14	1.45	-1.50	-1.75	-2.78	2.53	2.10
	VS ₂	0.93	-0.01	0.14	-0.11	0.34	0.19	6.02
	NbS ₂	0.57	-0.01	0.16	-0.09	0.35	0.09	6.08
	TaS ₂	0.56	0.02	0.09	-0.15	0.20	0.07	5.92
	VSe ₂	0.58	-0.09	-0.08	-0.33	-0.17	0.32	5.46

Table S8 Electronic SBH Φ_n (eV), hole SBH Φ_p (eV), interface dipole D_{int} (Debye), tot dipole D_{tot} (Debye), potential step ΔV (eV), the difference between the metal and MSJs work functions ΔW (eV), metal work functions W_M (eV) of 2D metal-SMoSe contacts under different strains

	Metal	Φ_n	Φ_p	D_{int}	D_{tot}	ΔV	ΔW	w_m
-6%	Nb ₂ CF ₂	0.04	1.54	0.17	0.38	0.47	0.06	4.37
	Nb ₂ CO ₂	1.00	0.50	0.27	0.48	0.83	-1.35	5.81
	Nb ₂ COH ₂	-0.31	1.65	-0.56	-0.35	-1.33	1.54	2.22
	Nb ₃ C ₂ F ₂	0.42	1.01	0.15	0.36	0.72	0.25	4.89
	Nb ₃ C ₂ O ₂	0.85	0.62	0.28	0.49	0.74	-0.15	5.48
	Nb ₃ C ₂ (OH) ₂	-0.30	1.54	-0.92	-0.71	-2.11	2.40	2.10
	VS ₂	0.70	0.87	0.46	0.68	1.35	-0.08	6.02
	NbS ₂	0.71	1.03	0.52	0.73	0.56	-0.50	6.08
	TaS ₂	0.67	1.06	0.45	0.66	1.25	-0.09	5.92
	VSe ₂	0.55	1.15	0.32	0.53	0.91	-0.22	5.46
-4%	Nb ₂ CF ₂	0.05	1.56	0.19	0.41	0.59	0.16	4.37
	Nb ₂ CO ₂	1.07	0.47	0.29	0.51	0.93	-0.24	5.81
	Nb ₂ COH ₂	-0.24	1.54	-0.63	-0.41	-1.44	1.55	2.22
	Nb ₃ C ₂ F ₂	0.38	1.10	0.14	0.36	0.72	0.24	4.89
	Nb ₃ C ₂ O ₂	1.09	0.46	0.33	0.55	0.80	-0.11	5.48
	Nb ₃ C ₂ (OH) ₂	-0.16	1.50	-1.01	-0.79	-2.21	2.43	2.10
	VS ₂	0.80	0.74	0.47	0.69	1.30	0.00	6.02
	NbS ₂	0.79	0.82	0.51	0.73	0.46	-0.47	6.08
	TaS ₂	0.75	0.88	0.44	0.66	1.17	-0.07	5.92
	VSe ₂	0.57	1.04	0.33	0.55	0.90	-0.14	5.46
-2%	Nb ₂ CF ₂	0.07	1.57	0.26	0.49	0.69	0.24	4.37
	Nb ₂ CO ₂	1.13	0.43	0.31	0.53	0.88	-0.19	5.81
	Nb ₂ COH ₂	-0.08	1.48	-0.71	-0.49	-1.56	1.56	2.22
	Nb ₃ C ₂ F ₂	0.35	1.16	0.28	0.51	0.77	0.25	4.89
	Nb ₃ C ₂ O ₂	1.04	0.52	0.32	0.54	0.82	-0.09	5.48
	Nb ₃ C ₂ (OH) ₂	-0.12	1.50	-1.11	-0.88	-2.31	2.41	2.10
	VS ₂	0.89	0.63	0.47	0.69	1.25	0.06	6.02
	NbS ₂	0.77	0.63	0.50	0.73	0.36	-0.45	6.08
	TaS ₂	0.73	0.73	0.43	0.66	1.09	-0.02	5.92

2%	VSe ₂	0.59	0.87	0.34	0.56	0.88	-0.07	5.46
	Nb ₂ CF ₂	0.07	1.43	0.31	0.55	0.75	0.43	4.37
	Nb ₂ CO ₂	1.22	0.38	0.34	0.58	0.89	-0.02	5.81
	Nb ₂ COH ₂	-0.09	1.60	-0.90	-0.67	-1.79	1.60	2.22
	Nb ₃ C ₂ F ₂	0.30	1.26	0.33	0.57	0.82	0.29	4.89
	Nb ₃ C ₂ O ₂	0.96	0.64	0.36	0.60	0.88	-0.03	5.48
	Nb ₃ C ₂ (OH) ₂	-0.22	1.54	-1.32	-1.08	-2.55	2.46	2.10
	VS ₂	0.98	0.31	0.49	0.73	1.22	0.16	6.02
	NbS ₂	0.56	0.30	0.51	0.75	0.20	-0.42	6.08
	TaS ₂	0.50	0.40	0.43	0.67	1.00	0.04	5.92
	VSe ₂	0.31	0.56	0.33	0.57	0.81	0.05	5.46
	Nb ₂ CF ₂	0.04	1.20	0.30	0.55	0.71	0.57	4.37
4%	Nb ₂ CO ₂	1.15	0.32	0.36	0.61	0.96	0.06	5.81
	Nb ₂ COH ₂	-0.05	1.53	-1.02	-0.77	-1.93	1.60	2.22
	Nb ₃ C ₂ F ₂	0.30	1.27	0.34	0.58	0.84	0.31	4.89
	Nb ₃ C ₂ O ₂	0.86	0.54	0.40	0.64	0.91	0.03	5.48
	Nb ₃ C ₂ (OH) ₂	-0.16	1.53	-1.42	-1.18	-2.68	2.50	2.10
	VS ₂	0.87	0.17	0.54	0.78	1.22	0.18	6.02
	NbS ₂	0.48	0.16	0.54	0.79	0.19	-0.43	6.08
	TaS ₂	0.41	0.27	0.46	0.70	1.02	0.06	5.92
	VSe ₂	0.22	0.42	0.32	0.57	0.77	0.11	5.46
	Nb ₂ CF ₂	0.03	0.98	0.27	0.52	0.61	0.60	4.37
	Nb ₂ CO ₂	1.02	0.18	0.39	0.64	1.15	0.12	5.81
	Nb ₂ COH ₂	-0.08	1.42	-1.15	-0.90	-2.12	-2.22	2.22
6%	Nb ₃ C ₂ F ₂	0.12	1.29	0.34	0.59	0.84	0.33	4.89
	Nb ₃ C ₂ O ₂	0.73	0.44	0.43	0.67	0.95	0.14	5.48
	Nb ₃ C ₂ (OH) ₂	-0.08	1.48	-1.52	-1.27	-2.80	2.54	2.10
	VS ₂	0.74	0.06	0.57	0.82	1.31	0.20	6.02
	NbS ₂	0.39	0.06	0.57	0.82	0.17	-0.42	6.08
	TaS ₂	0.34	0.14	0.48	0.73	1.04	0.08	5.92
	VSe ₂	0.15	0.29	0.33	0.58	0.74	0.15	5.46
	VSe ₂	0.15	0.29	0.33	0.58	0.74	0.15	5.46

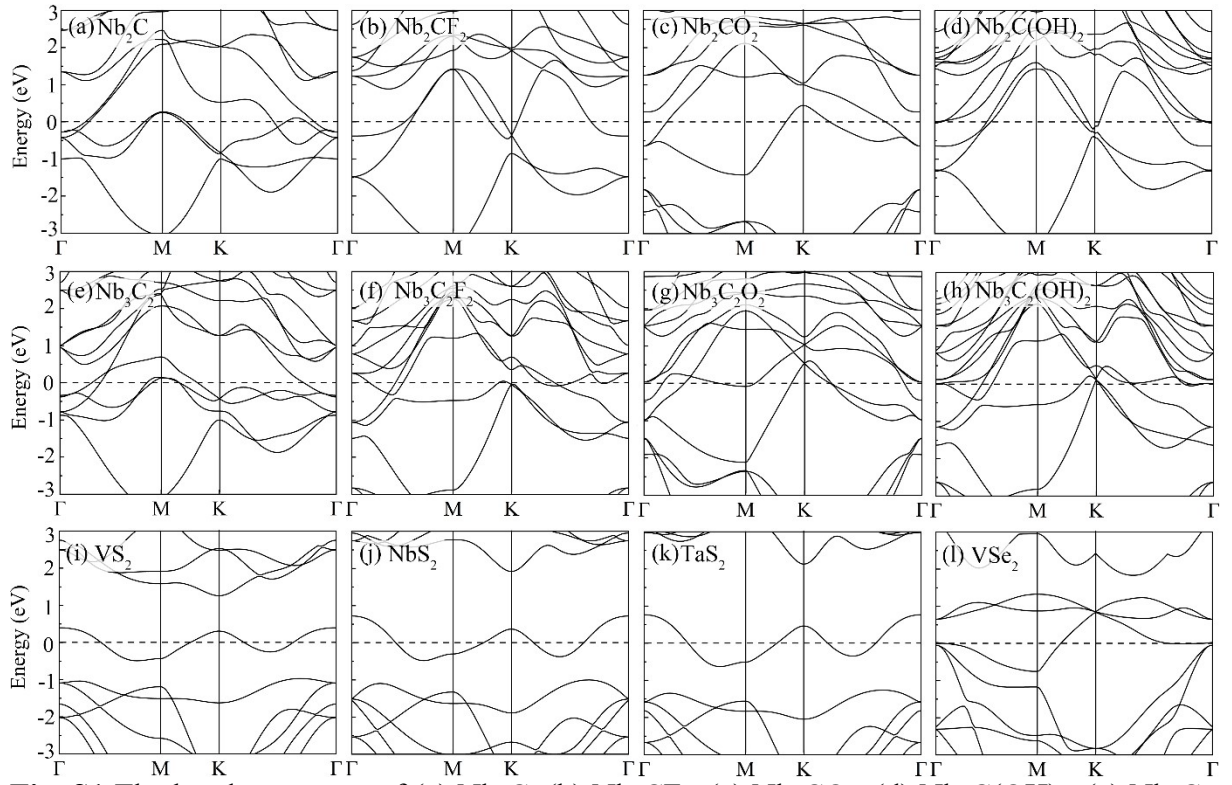


Fig. S1 The band structures of (a) Nb_2C , (b) Nb_2CF_2 , (c) Nb_2CO_2 , (d) $\text{Nb}_2\text{C}(\text{OH})_2$, (e) Nb_3C_2 , (f) $\text{Nb}_3\text{C}_2\text{F}_2$, (g) $\text{Nb}_3\text{C}_2\text{O}_2$, (h) $\text{Nb}_3\text{C}_2(\text{OH})_2$, (i) VS_2 , (j) NbS_2 , (k) TaS_2 , and (l) VSe_2 monolayers.

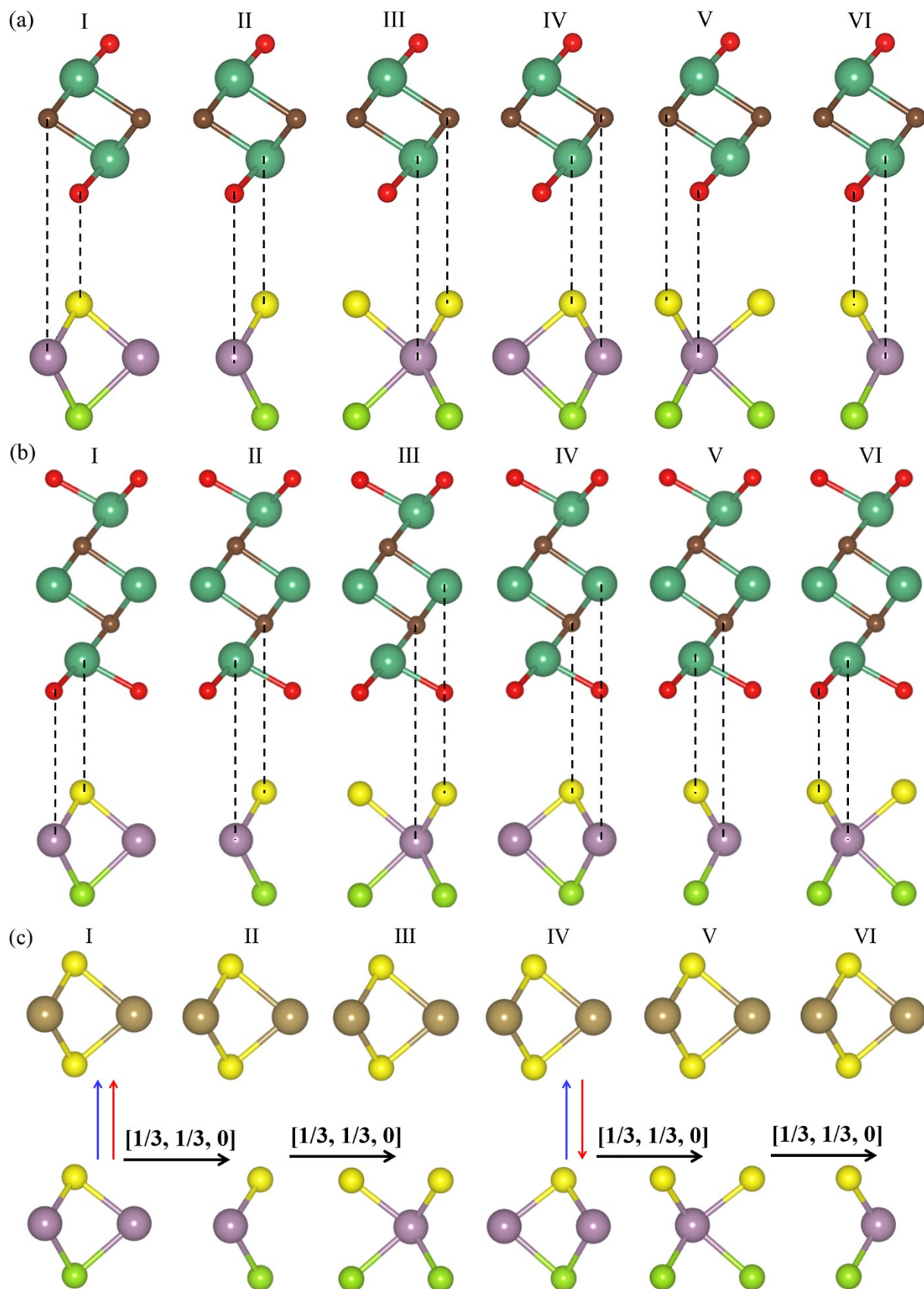


Fig. S2 Side view of (a) $\text{Nb}_2\text{CT}_2\text{-MoS}_2$, (b) $\text{Nb}_3\text{C}_2\text{T}_2\text{-MoS}_2$ (c) $\text{MX}_2\text{-MoS}_2$ contacts with six possible stacking configurations.

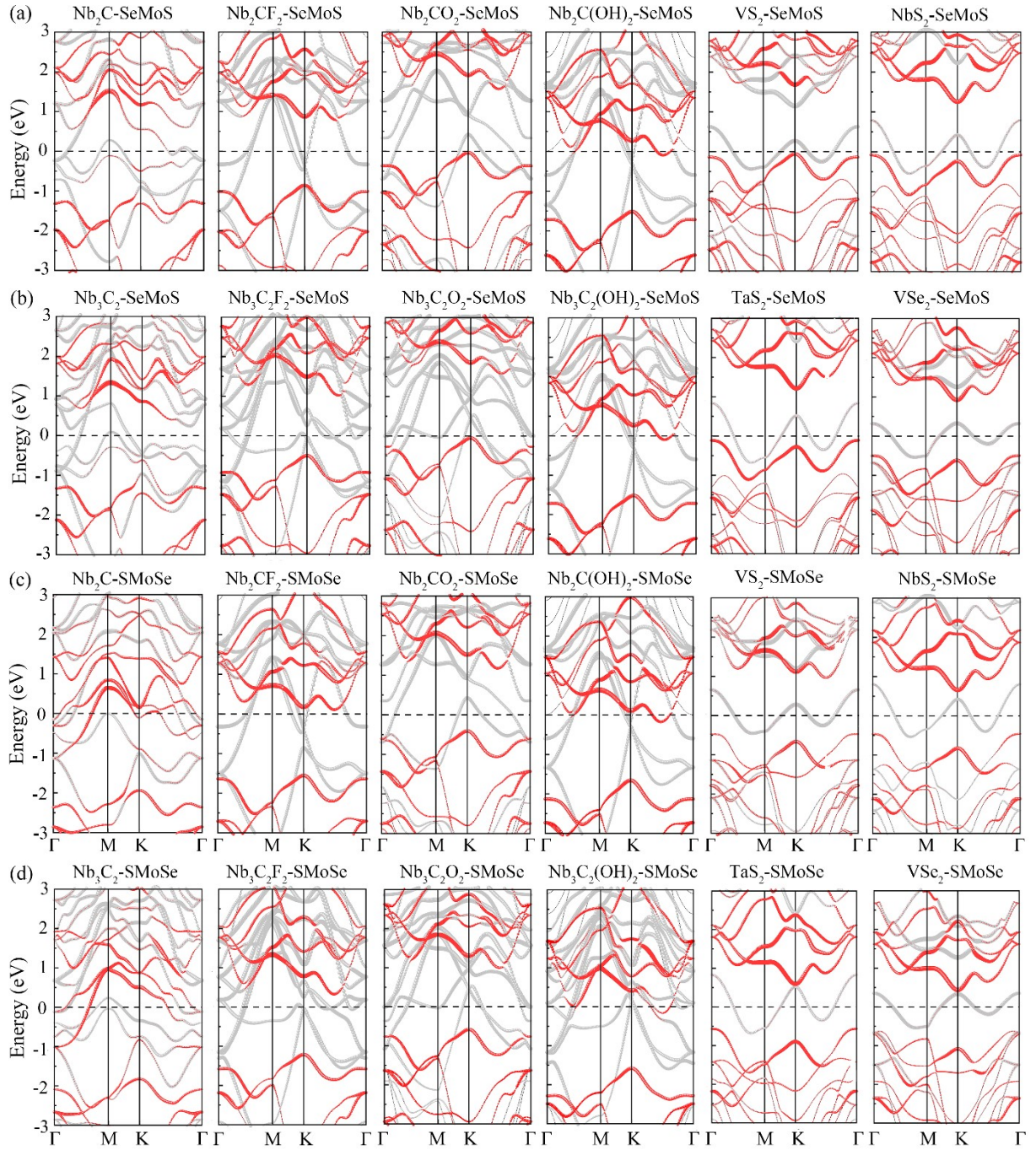


Fig. S3 The projected band structures of (a, b) metal-SeMoS and (c, d) metal-SMoSe contacts. The red and gray spheres represent the contributions from MoS₂ and metal layers, respectively.

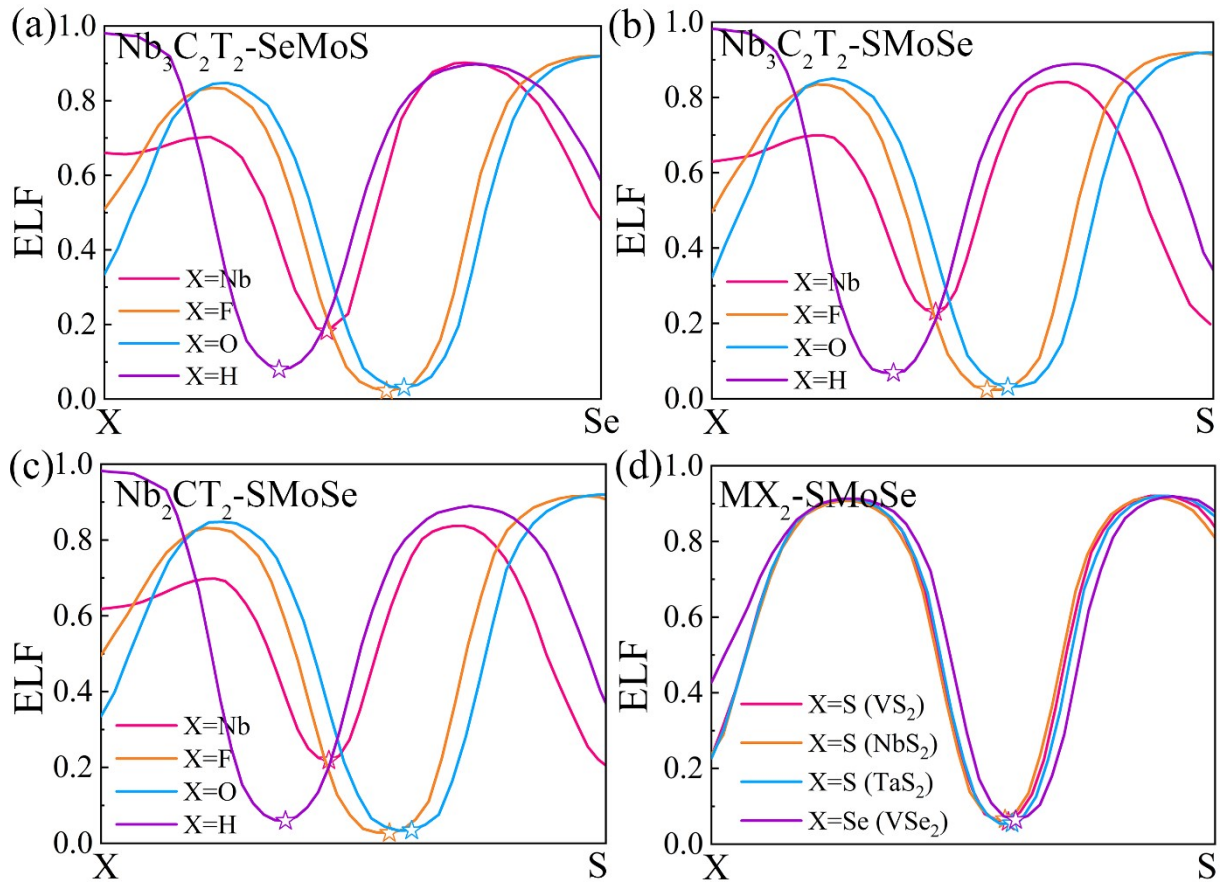
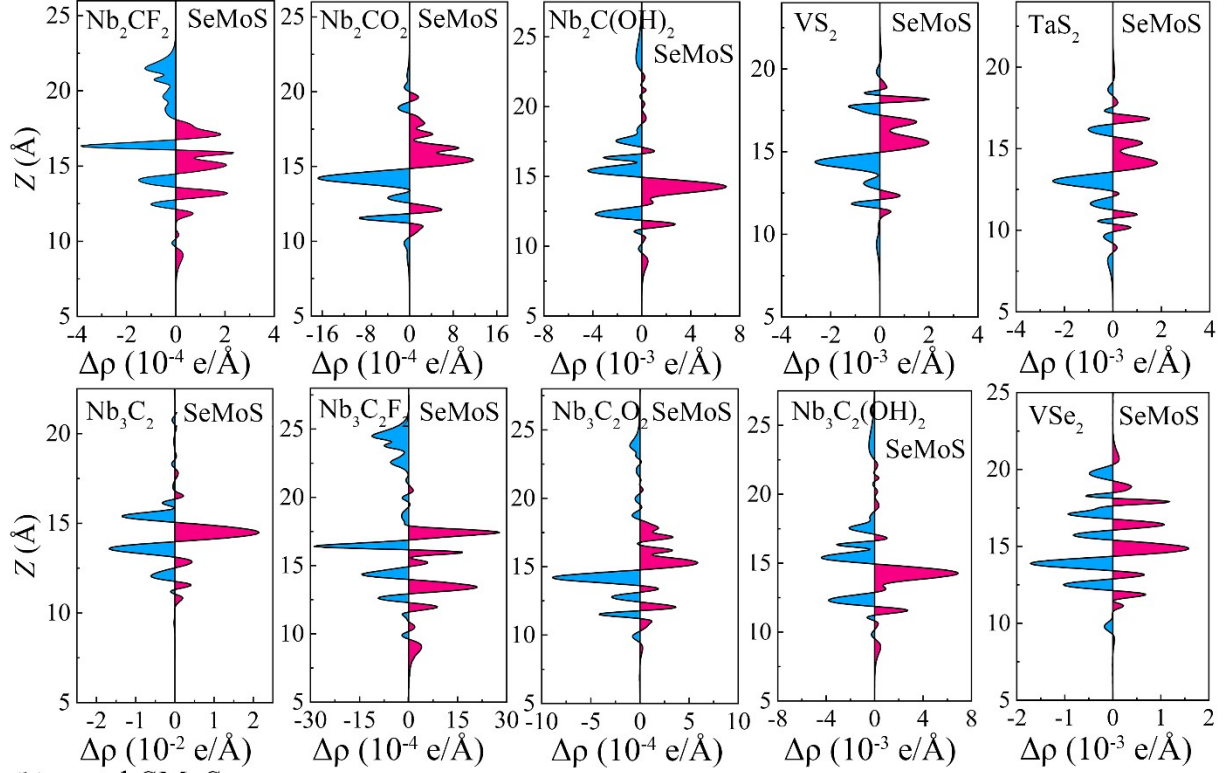


Fig. S4 The ELF 2D line profiles and bond points of the (a) X–Se and (b-d) X–S bonds in 2D metal-MoSSe contacts. The pentagrams represent the bond points of the X–Se and X–S bonds in 2D metal-MoSSe contacts.

(a) metal-SeMoS



(b) metal-SMoSe

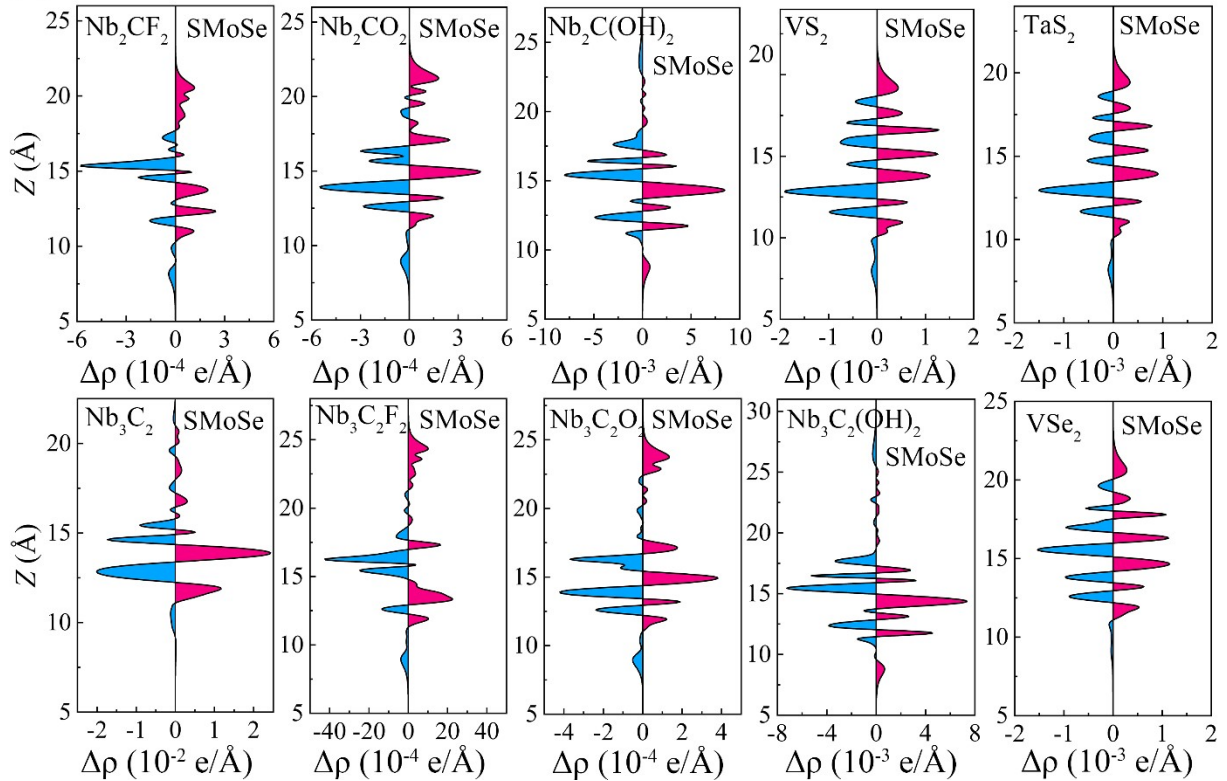


Fig. S5 The plane averaged charge density difference for (a) metal-SeMoS and (b) metal-SMoSe contacts. Pink indicates charge accumulation, azure indicates charge depletion.

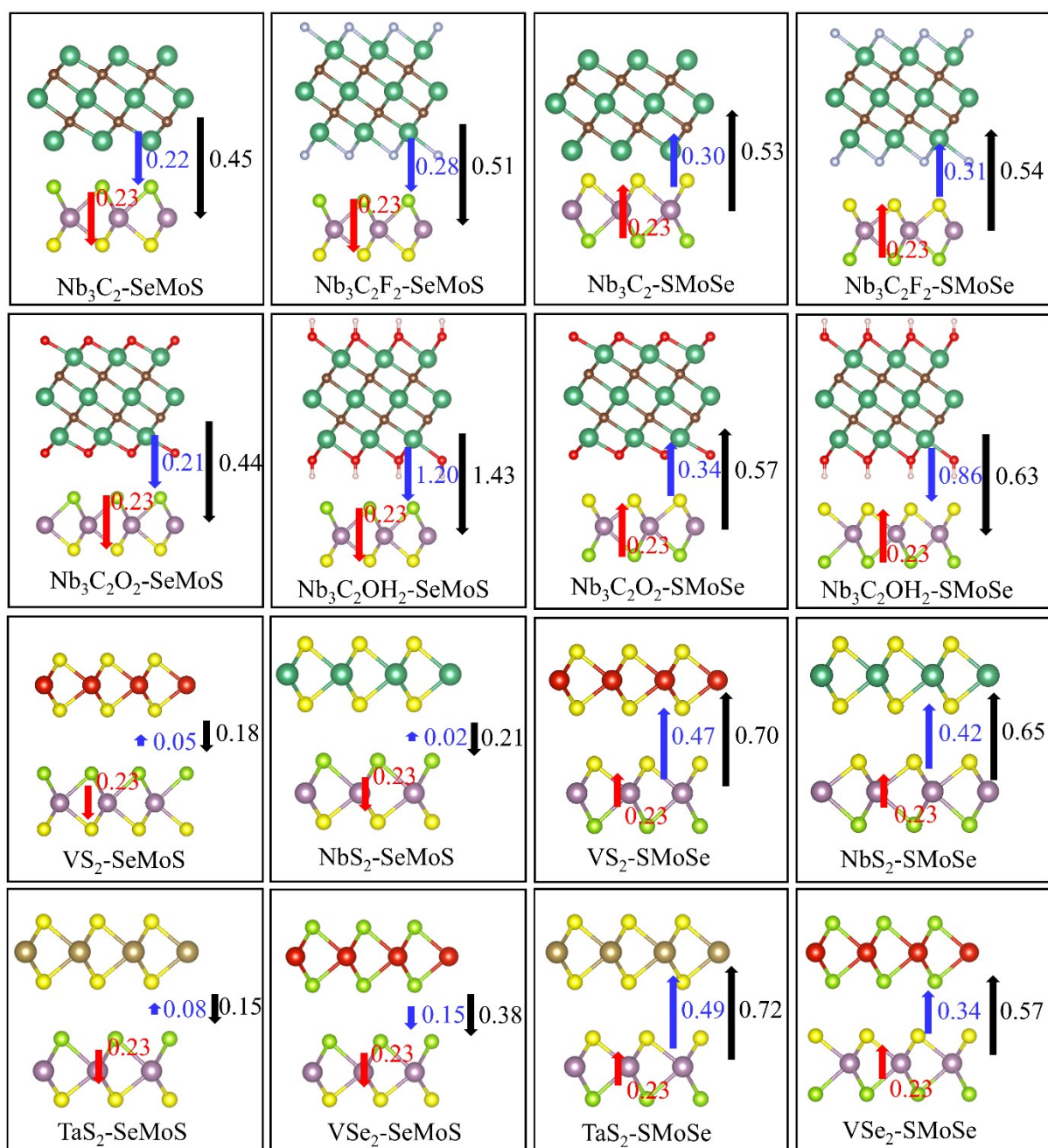


Fig. S6 The dipole moment distributions of 2D metal-MoSSe contacts. The red, blue and black arrows represent the intrinsic dipole, interface dipole and tot dipole, respectively.

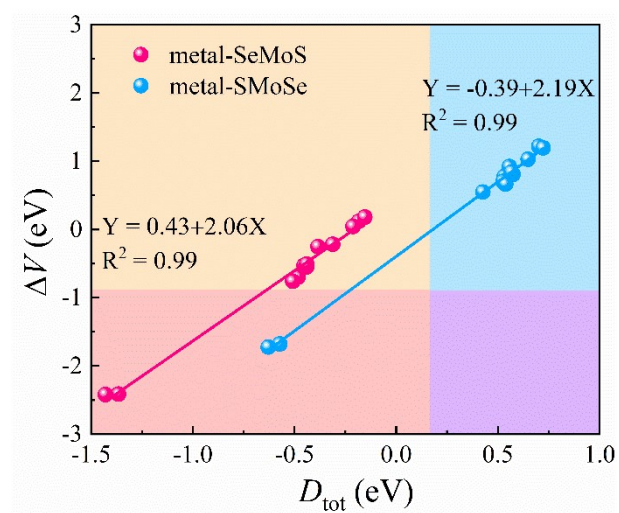


Fig. S7 The change of ΔV with tot dipole moment (D_{tot}) for 2D metal-MoSSe contacts.

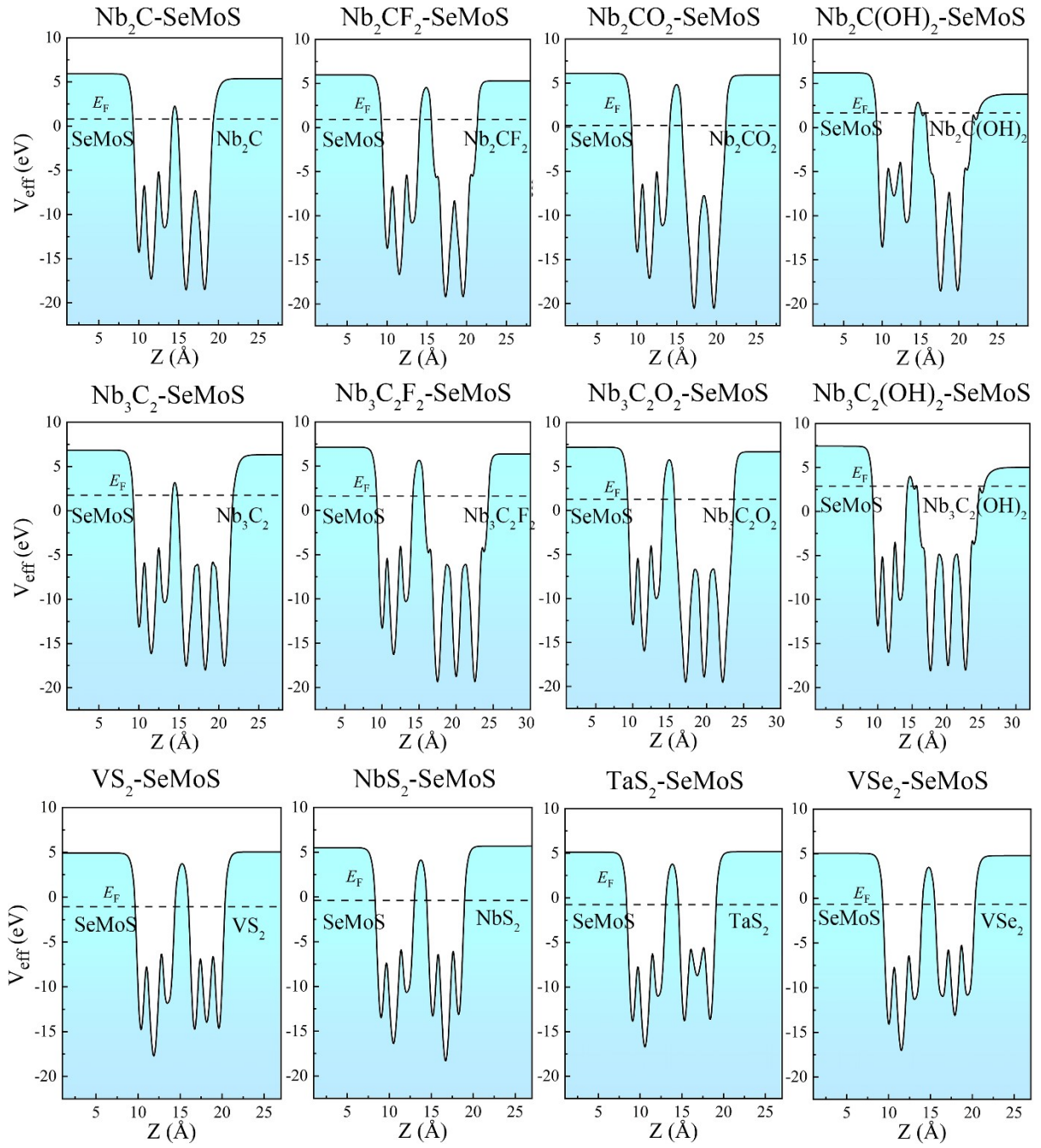


Fig. S8 Effective electrostatic potential of 2D metal-SeMoS contacts.

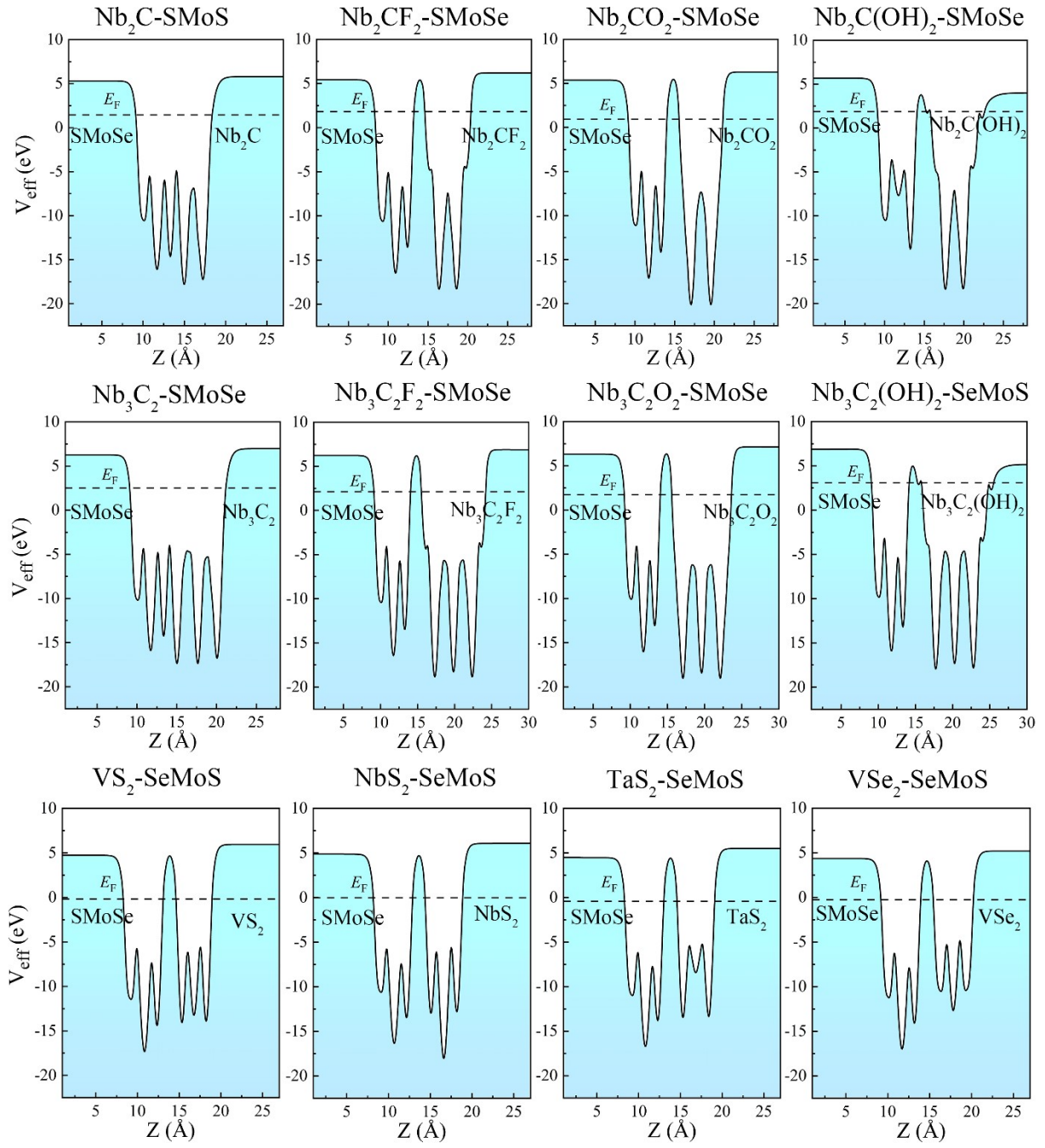


Fig. S9 Effective electrostatic potential of 2D metal-SMoSe contacts.

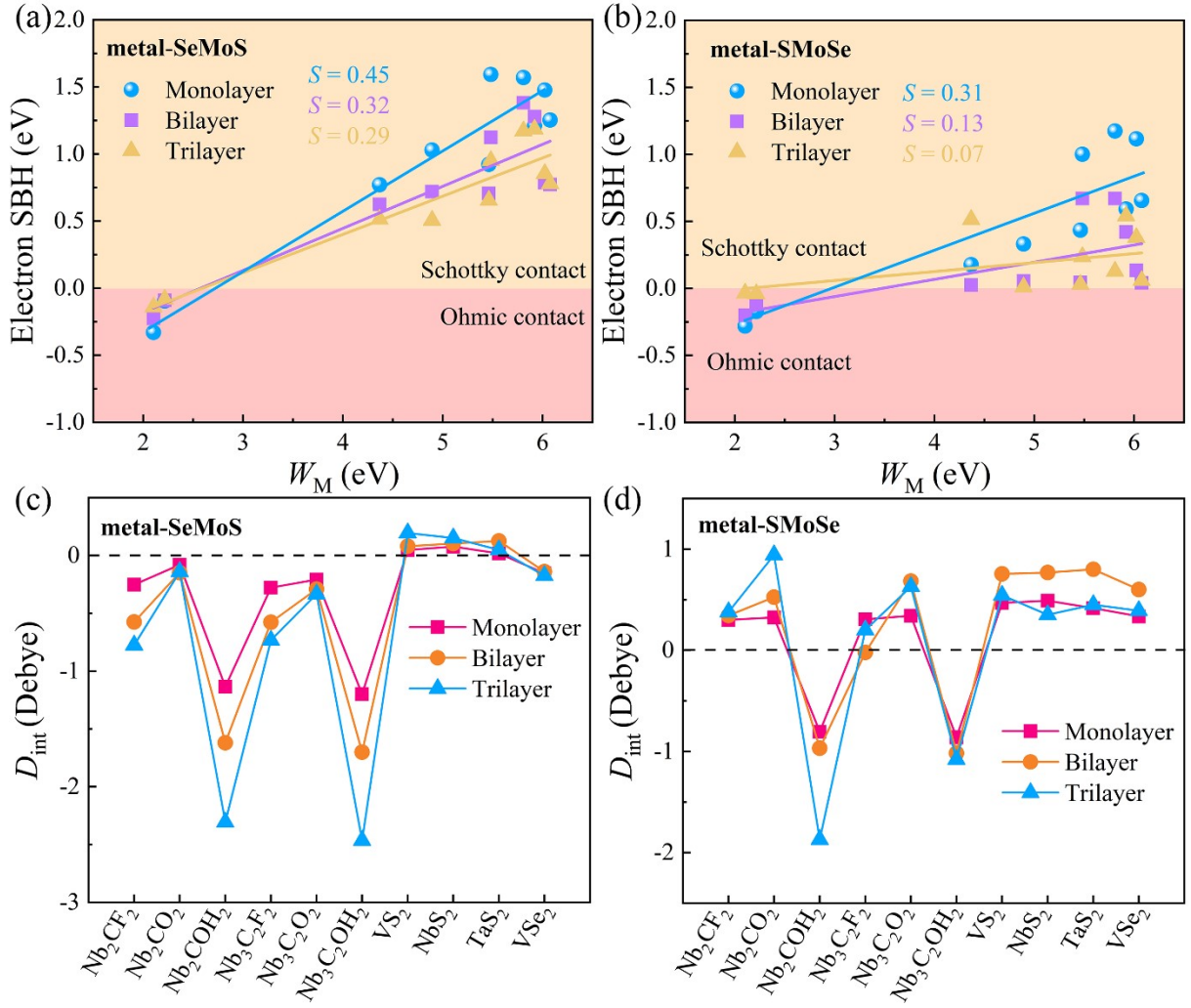


Fig. S10 The variation of electron SBH as a function of metal work functions (W_M) for (a) metal-SeMoS (b) metal-SMoSe contacts. The Fermi level pinning factor S is defined as the value of the fitted slope. Variation of the interface dipole (D_{int}) in the (c) metal-SeMoS and (d) metal-SMoSe contacts.

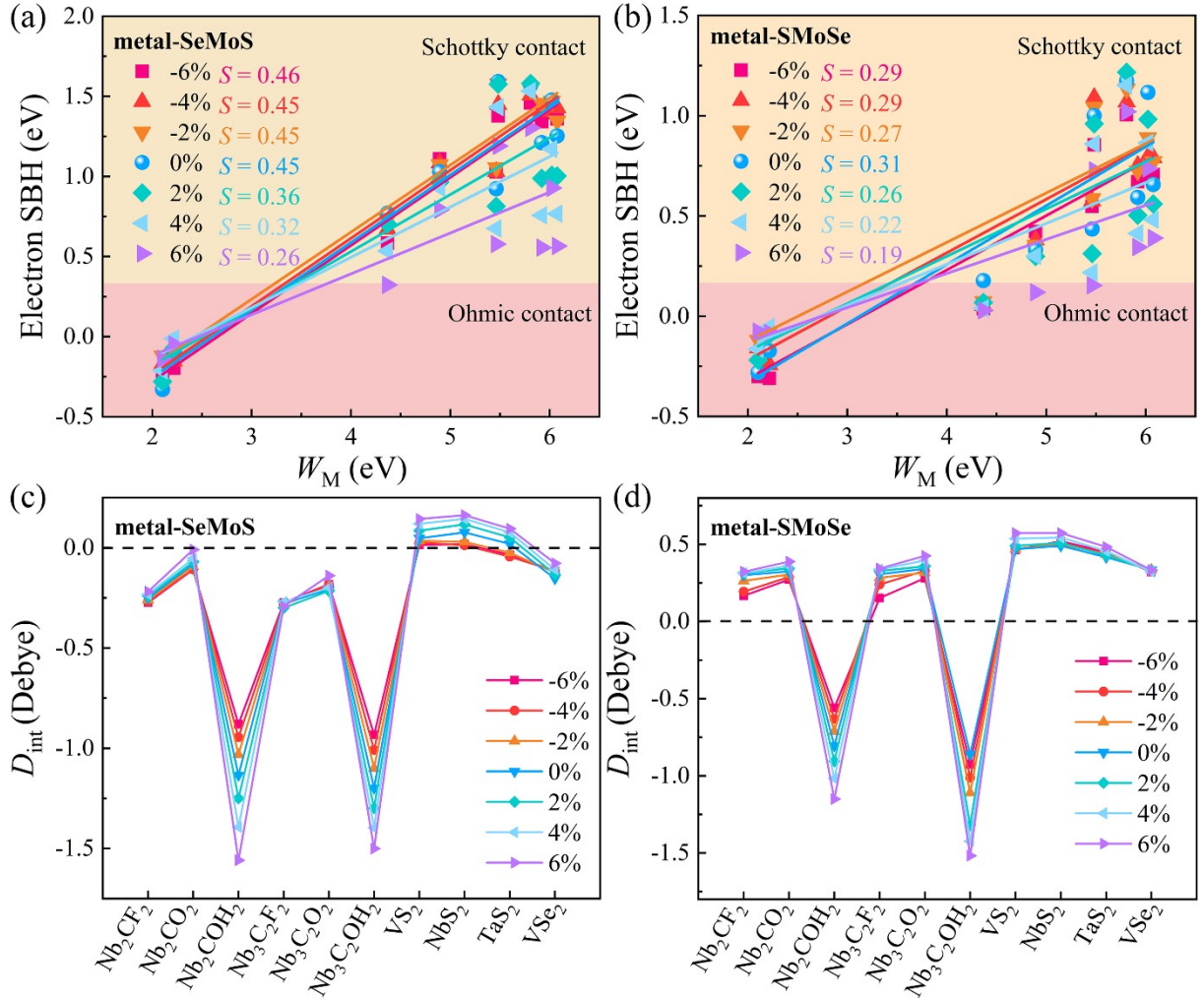


Fig. S11 The variation of electron SBH as a function of metal work functions (W_M) for (a) metal-SeMoS (b) metal-SMoSe contacts. The Fermi level pinning factor S is defined as the value of the fitted slope. Variation of the interface dipole (D_{int}) in the (c) metal-SeMoS and (d) metal-SMoSe contacts.

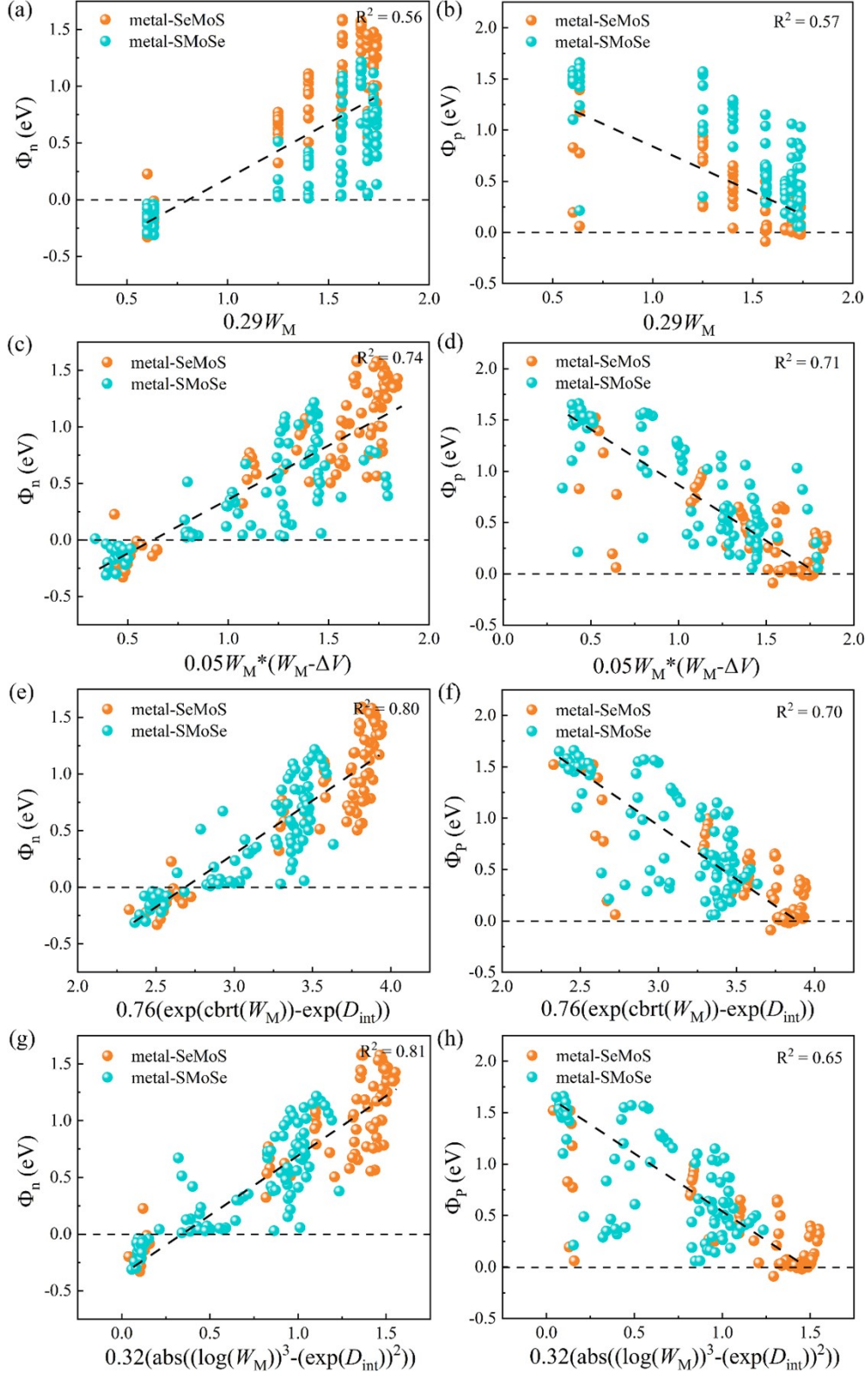


Fig. S12 The correlations of the calculated (a) Φ_n and (b) Φ_p values of metal-MoSSe contacts vs. predicted $0.29W_M$ formula from SISSO. The correlations of the calculated (c) Φ_n and (d) Φ_p values of metal-MoSSe contacts vs. predicted $0.05W_M*(W_M-\Delta V)$ formula from SISSO. The correlations of the calculated (e) Φ_n and (f) Φ_p values of metal-MoSSe contacts vs. predicted $0.76(\exp(\text{cbrr}(W_M))-\exp(D_{\text{int}}))$ formula from SISSO. The correlations of the calculated (g) Φ_n and (h) Φ_p values of metal-MoSSe contacts vs. predicted $0.32(\text{abs}((\log(W_M))^3-(\exp(D_{\text{int}}))^2))$ formula from SISSO.

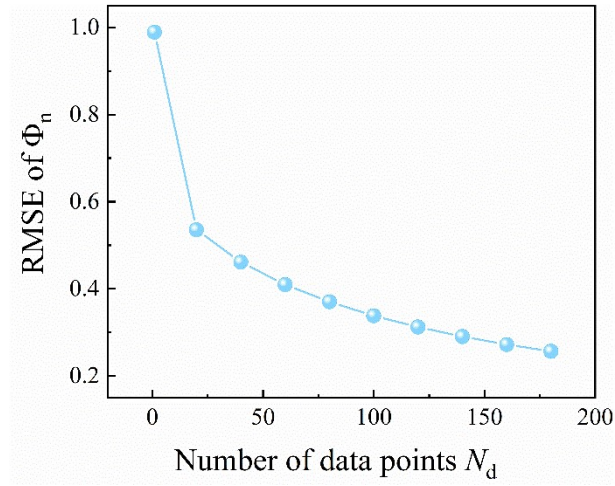


Fig. S13 The root-mean-square error (RMSE) of Φ_n is plotted as a function of the training data set size, N_d using the supervised data sampling method.