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 Nb_2CT_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}
Nb_2C/SeMoS	I	203.39	Nb_2C/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_2CF_2/SeMoS	I	68.229	Nb_2CF_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_2CO_2/SeMoS	I	111.65	Nb_2CO_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
$Nb_2C(OH)_2$/SeMoS	I	19.12	$Nb_2C(OH)_2$/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 $Nb_3C_2T_2$-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}
Nb_3C_2/SeMoS	I	491.68	Nb_3C_2/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
$Nb_3C_2F_2$/SeMoS	I	16.67	$Nb_3C_2F_2$/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
$Nb_3C_2O_2$/SeMoS	I	33.57	$Nb_3C_2O_2$/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
Nb_3C_2 $(OH)_2$/SeMoS	I	64.56	Nb_3C_2 $(OH)_2$/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 MX_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}
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
NbS_2/SeMoS	I	91.57	NbS_2/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
TaS_2/SeMoS	I	83.47	TaS_2/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
VSe_2/SeMoS	I	82.07	VSe_2/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_2C	1.46	0	0.67	0	0.66	0
Nb_2CF_2	3.58	3.57	1.42	1.34	7.17	6.45
Nb_2CO_2	4.60	4.86	1.55	1.57	11.01	11.92
Nb_2COH_2	1.20	1.88	0.87	1.08	0.78	2.18
Nb_3C_2	1.42	0	0.64	0	0.58	0
$Nb_3C_2F_2$	4.02	4.08	1.49	1.48	8.93	8.94
$Nb_3C_2O_2$	4.50	4.63	1.53	1.52	10.52	10.76
$Nb_3C_2(OH)_2$	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_{tint}	D_{tot}	ΔV	ΔW	W_M
ML-MoSSe	Nb_2CF_2	0.77	0.86	-0.25	-0.48	-0.70	0.69	4.37
	Nb_2CO_2	1.57	0.03	-0.08	-0.31	-0.22	0.11	5.81
	Nb_2COH_2	-0.10	1.52	-1.13	-1.37	-2.41	2.33	2.22
	$Nb_3C_2F_2$	1.03	0.50	-0.28	-0.51	-0.76	0.62	4.89
	$Nb_3C_2O_2$	1.59	0.05	-0.21	-0.44	-0.51	0.44	5.48
	$Nb_3C_2(OH)_2$	-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
BL-MoSSe	Nb_2CF_2	0.63	0.27	-0.57	-1.04	-1.47	1.44	4.37
	Nb_2CO_2	1.38	0.04	-0.15	-0.61	-0.30	0.13	5.81
	Nb_2COH_2	-0.09	0.78	-1.62	-2.08	-3.62	3.64	2.22
	$Nb_3C_2F_2$	0.72	0.26	-0.58	-1.04	-1.28	0.98	4.89
	$Nb_3C_2O_2$	1.12	0.02	-0.29	-0.75	-0.59	0.37	5.48
	$Nb_3C_2(OH)_2$	-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
TL-MoSSe	Nb_2CF_2	0.51	0.25	-0.77	-1.24	-2.06	1.07	4.37
	Nb_2CO_2	1.17	0.02	-0.14	-0.60	-0.27	0.08	5.81
	Nb_2COH_2	-0.08	0.06	-2.31	-2.77	-3.60	3.47	2.22
	$Nb_3C_2F_2$	0.51	0.04	-0.73	-1.20	-1.28	0.91	4.89
	$Nb_3C_2O_2$	0.95	0.03	-0.33	-0.79	-0.55	0.32	5.48
	$Nb_3C_2(OH)_2$	-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_2CF_2	0.18	1.55	0.30	0.53	0.78	0.01	4.37
	Nb_2CO_2	1.17	0.41	0.32	0.56	0.92	-0.10	5.81
	Nb_2COH_2	-0.17	1.66	-0.80	-0.57	-1.68	1.58	2.22
	$Nb_3C_2F_2$	0.33	1.21	0.31	0.54	0.67	-0.11	4.89
	$Nb_3C_2O_2$	1.00	0.58	0.34	0.57	0.81	-0.07	5.48
	$Nb_3C_2(OH)_2$	-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
BL-MoSSe	Nb_2CF_2	0.02	1.05	0.34	0.81	0.74	0.02	4.37
	Nb_2CO_2	0.67	0.19	0.53	0.99	1.36	-0.51	5.81
	Nb_2COH_2	-0.11	1.24	-0.97	-0.50	-1.74	1.61	2.22
	$Nb_3C_2F_2$	0.05	1.02	0.38	0.85	0.13	-0.25	4.89
	$Nb_3C_2O_2$	0.67	0.29	0.68	1.15	1.52	-0.19	5.48
	$Nb_3C_2(OH)_2$	-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
TL-MoSSe	Nb_2CF_2	-0.01	0.35	0.38	0.84	0.72	0.02	4.37
	Nb_2CO_2	0.13	0.47	0.94	1.41	1.98	0.08	5.81
	Nb_2COH_2	-0.04	0.21	-1.87	-1.41	-1.62	3.47	2.22
	$Nb_3C_2F_2$	0.01	0.84	0.54	1.00	3.51	-0.32	4.89
	$Nb_3C_2O_2$	0.24	0.39	0.63	1.09	1.65	-0.18	5.48
	$Nb_3C_2(OH)_2$	-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_{tint}	D_{tot}	ΔV	ΔW	W_M
-6%	Nb_2CF_2	0.58	1.00	-0.27	-0.49	-0.85	0.43	4.37
	Nb_2CO_2	1.46	0.04	-0.11	-0.32	-0.37	0.39	5.81
	Nb_2COH_2	-0.20	1.52	-0.48	-0.69	-2.09	2.32	2.22
	$Nb_3C_2F_2$	1.11	0.33	-0.27	-0.49	-0.87	0.66	4.89
	$Nb_3C_2O_2$	1.38	0.04	-0.18	-0.39	-0.48	0.32	5.48
	$Nb_3C_2(OH)_2$	-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_2CF_2	0.67	0.94	-0.27	-0.49	-0.80	0.52	4.37
	Nb_2CO_2	1.51	0.04	-0.11	-0.33	-0.32	0.47	5.81
	Nb_2COH_2	-0.15	1.52	-0.95	-1.16	-2.19	2.30	2.22
	$Nb_3C_2F_2$	1.08	0.40	-0.28	-0.50	-0.81	0.62	4.89
	$Nb_3C_2O_2$	1.45	0.04	-0.18	-0.40	-0.49	0.37	5.48
	$Nb_3C_2(OH)_2$	-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_2CF_2	0.73	0.89	-0.26	-0.49	-0.75	0.60	4.37
	Nb_2CO_2	1.55	0.03	-0.09	-0.32	-0.28	0.54	5.81
	Nb_2COH_2	-0.12	1.52	-1.03	-1.26	-2.29	2.30	2.22
	$Nb_3C_2F_2$	1.07	0.44	-0.28	-0.51	-0.76	0.59	4.89
	$Nb_3C_2O_2$	1.58	0.05	-0.20	-0.42	-0.50	0.40	5.48
	$Nb_3C_2(OH)_2$	-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

	VSe_2	1.06	0.63	-0.12	-0.35	-0.27	0.21	5.46
2%	Nb_2CF_2	0.70	0.84	-0.24	-0.48	-0.64	0.78	4.37
	Nb_2CO_2	1.58	0.04	-0.07	-0.31	-0.18	0.16	5.81
	Nb_2COH_2	-0.07	1.52	-1.25	-1.49	-2.53	-2.22	2.22
	$Nb_3C_2F_2$	0.97	0.56	-0.30	-0.54	-0.69	0.62	4.89
	$Nb_3C_2O_2$	1.58	0.07	-0.22	-0.45	-0.51	0.47	5.48
	$Nb_3C_2(OH)_2$	-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
	Nb_2CF_2	0.53	0.74	-0.23	-0.48	-0.65	0.93	4.37
	Nb_2CO_2	1.53	0.04	-0.05	-0.30	-0.13	0.19	5.81
	Nb_2COH_2	-0.01	1.39	-1.39	-1.64	-2.71	-2.22	2.22
	$Nb_3C_2F_2$	0.93	0.60	-0.27	-0.51	-0.65	0.63	4.89
	$Nb_3C_2O_2$	1.43	0.07	-0.20	-0.44	-0.45	0.47	5.48
4%	$Nb_3C_2(OH)_2$	-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
	Nb_2CF_2	0.32	0.70	-0.22	-0.47	-0.54	0.92	4.37
	Nb_2CO_2	1.30	0.03	-0.01	-0.26	-0.02	0.15	5.81
	Nb_2COH_2	-0.05	1.18	-1.56	-1.81	-2.94	-2.22	2.22
	$Nb_3C_2F_2$	0.79	0.65	-0.29	-0.54	-0.59	0.62	4.89
	$Nb_3C_2O_2$	1.19	0.04	-0.14	-0.39	-0.31	0.44	5.48
6%	$Nb_3C_2(OH)_2$	-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_2CF_2	0.04	1.54	0.17	0.38	0.47	0.06	4.37
	Nb_2CO_2	1.00	0.50	0.27	0.48	0.83	-1.35	5.81
	Nb_2COH_2	-0.31	1.65	-0.56	-0.35	-1.33	1.54	2.22
	$Nb_3C_2F_2$	0.42	1.01	0.15	0.36	0.72	0.25	4.89
	$Nb_3C_2O_2$	0.85	0.62	0.28	0.49	0.74	-0.15	5.48
	$Nb_3C_2(OH)_2$	-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_2CF_2	0.05	1.56	0.19	0.41	0.59	0.16	4.37
	Nb_2CO_2	1.07	0.47	0.29	0.51	0.93	-0.24	5.81
	Nb_2COH_2	-0.24	1.54	-0.63	-0.41	-1.44	1.55	2.22
	$Nb_3C_2F_2$	0.38	1.10	0.14	0.36	0.72	0.24	4.89
	$Nb_3C_2O_2$	1.09	0.46	0.33	0.55	0.80	-0.11	5.48
	$Nb_3C_2(OH)_2$	-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_2CF_2	0.07	1.57	0.26	0.49	0.69	0.24	4.37
	Nb_2CO_2	1.13	0.43	0.31	0.53	0.88	-0.19	5.81
	Nb_2COH_2	-0.08	1.48	-0.71	-0.49	-1.56	1.56	2.22
	$Nb_3C_2F_2$	0.35	1.16	0.28	0.51	0.77	0.25	4.89
	$Nb_3C_2O_2$	1.04	0.52	0.32	0.54	0.82	-0.09	5.48
	$Nb_3C_2(OH)_2$	-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

	VSe_2	0.59	0.87	0.34	0.56	0.88	-0.07	5.46
2%	Nb_2CF_2	0.07	1.43	0.31	0.55	0.75	0.43	4.37
	Nb_2CO_2	1.22	0.38	0.34	0.58	0.89	-0.02	5.81
	Nb_2COH_2	-0.09	1.60	-0.90	-0.67	-1.79	1.60	2.22
	$Nb_3C_2F_2$	0.30	1.26	0.33	0.57	0.82	0.29	4.89
	$Nb_3C_2O_2$	0.96	0.64	0.36	0.60	0.88	-0.03	5.48
	$Nb_3C_2(OH)_2$	-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
4%	Nb_2CF_2	0.04	1.20	0.30	0.55	0.71	0.57	4.37
	Nb_2CO_2	1.15	0.32	0.36	0.61	0.96	0.06	5.81
	Nb_2COH_2	-0.05	1.53	-1.02	-0.77	-1.93	1.60	2.22
	$Nb_3C_2F_2$	0.30	1.27	0.34	0.58	0.84	0.31	4.89
	$Nb_3C_2O_2$	0.86	0.54	0.40	0.64	0.91	0.03	5.48
	$Nb_3C_2(OH)_2$	-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
6%	Nb_2CF_2	0.03	0.98	0.27	0.52	0.61	0.60	4.37
	Nb_2CO_2	1.02	0.18	0.39	0.64	1.15	0.12	5.81
	Nb_2COH_2	-0.08	1.42	-1.15	-0.90	-2.12	-2.22	2.22
	$Nb_3C_2F_2$	0.12	1.29	0.34	0.59	0.84	0.33	4.89
	$Nb_3C_2O_2$	0.73	0.44	0.43	0.67	0.95	0.14	5.48
	$Nb_3C_2(OH)_2$	-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

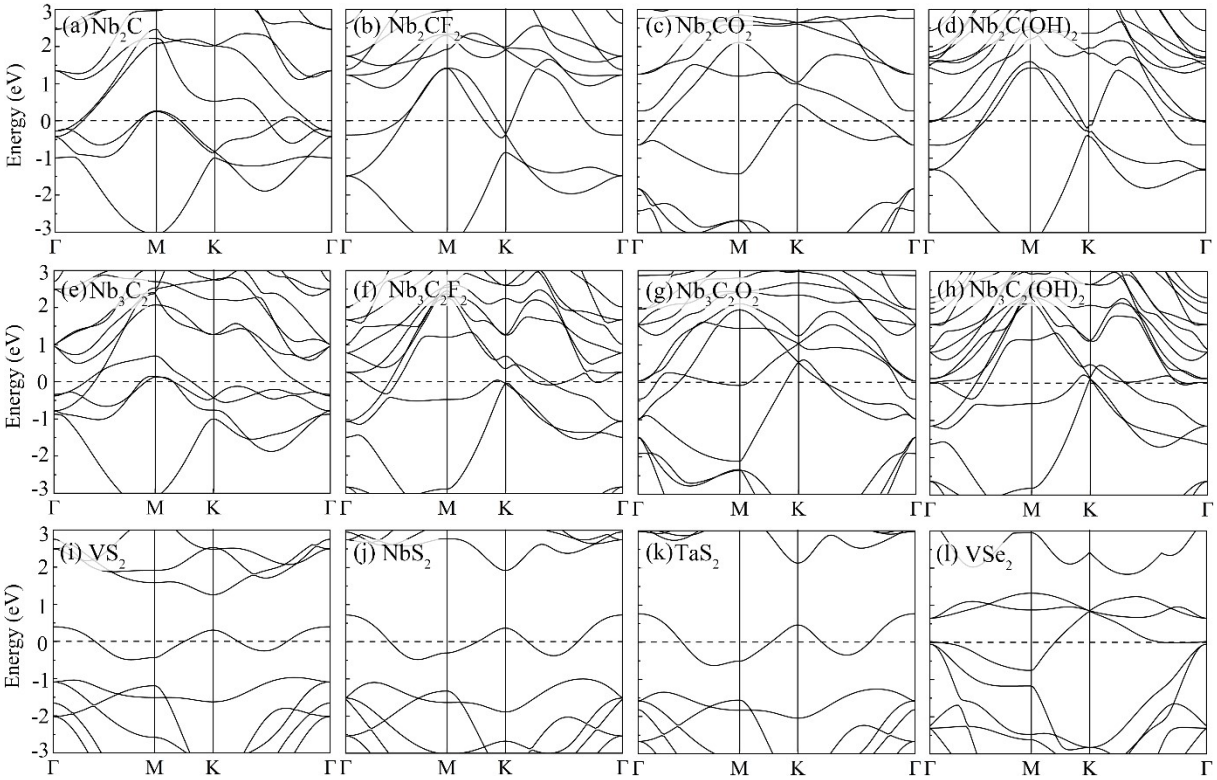

Fig. S1 The band structures of (a) Nb_2C, (b) Nb_2CF_2, (c) Nb_2CO_2, (d) $Nb_2C(OH)_2$, (e) Nb_3C_2, (f) $Nb_3C_2F_2$, (g) $Nb_3C_2O_2$, (h) $Nb_3C_2(OH)_2$, (i) VS_2, (j) NbS_2, (k) TaS_2, and (l) VSe_2 monolayers.

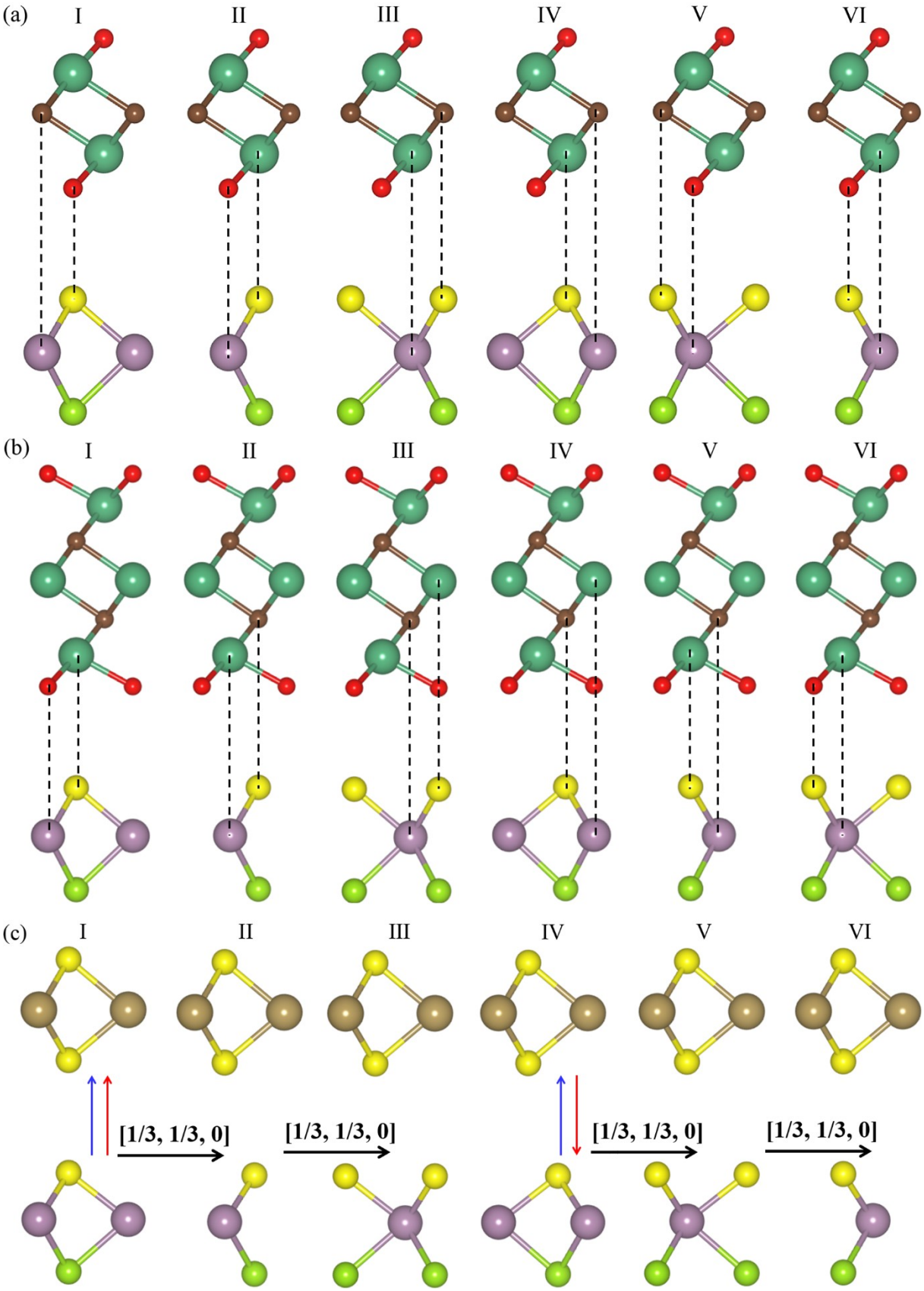

Fig. S2 Side view of (a) Nb_2CT_2-MoSSe, (b) $Nb_3C_2T_2$-MoSSe (c) MX_2-MoSSe contacts with six possible stacking configurations.

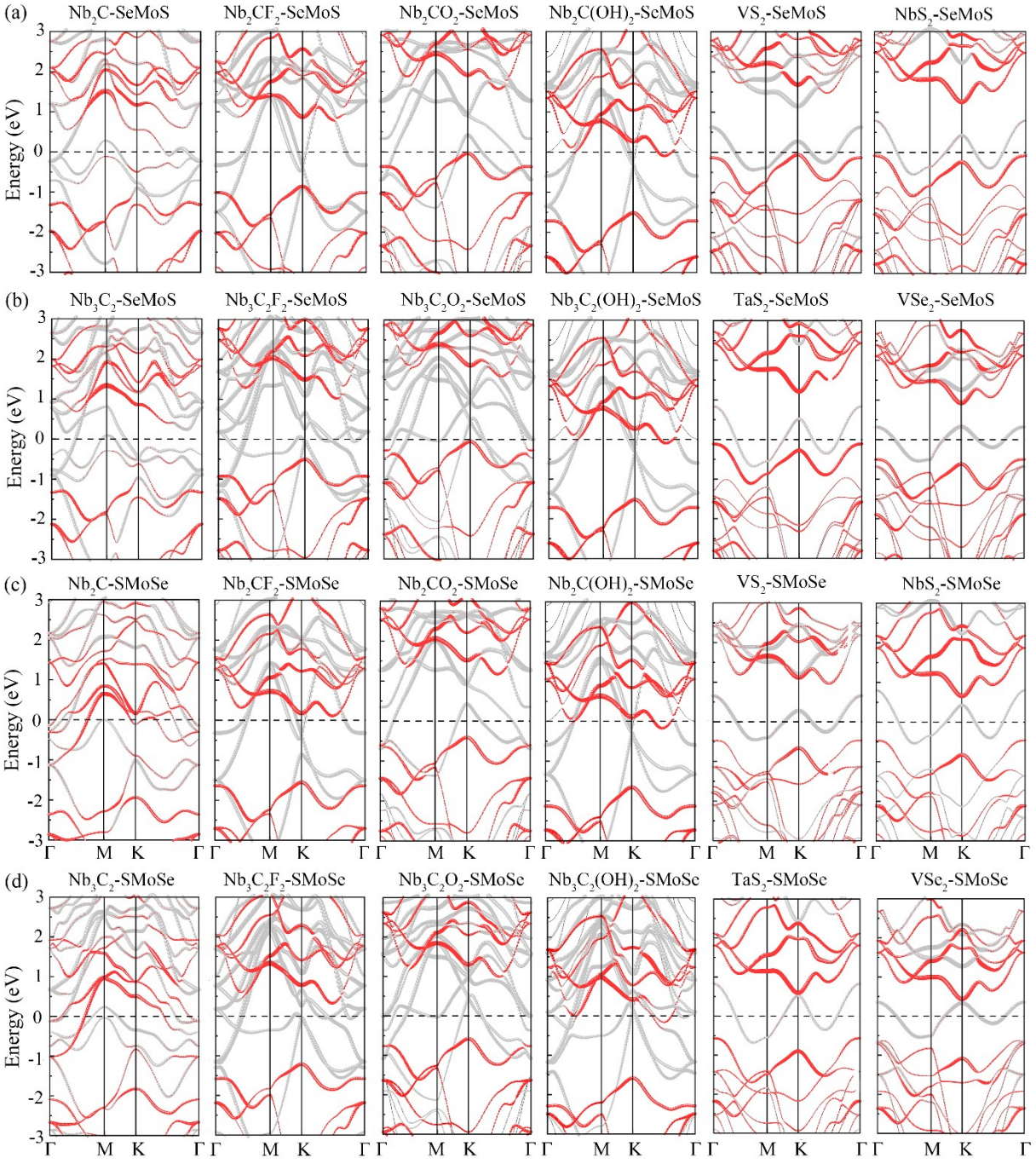

Fig. S3 The projected band structures of (a, b) metal-SeMoS and (c, d) metal-SMoSe contacts. The red and gray spheres represent the contributions from MoSSe and metal layers, respectively.

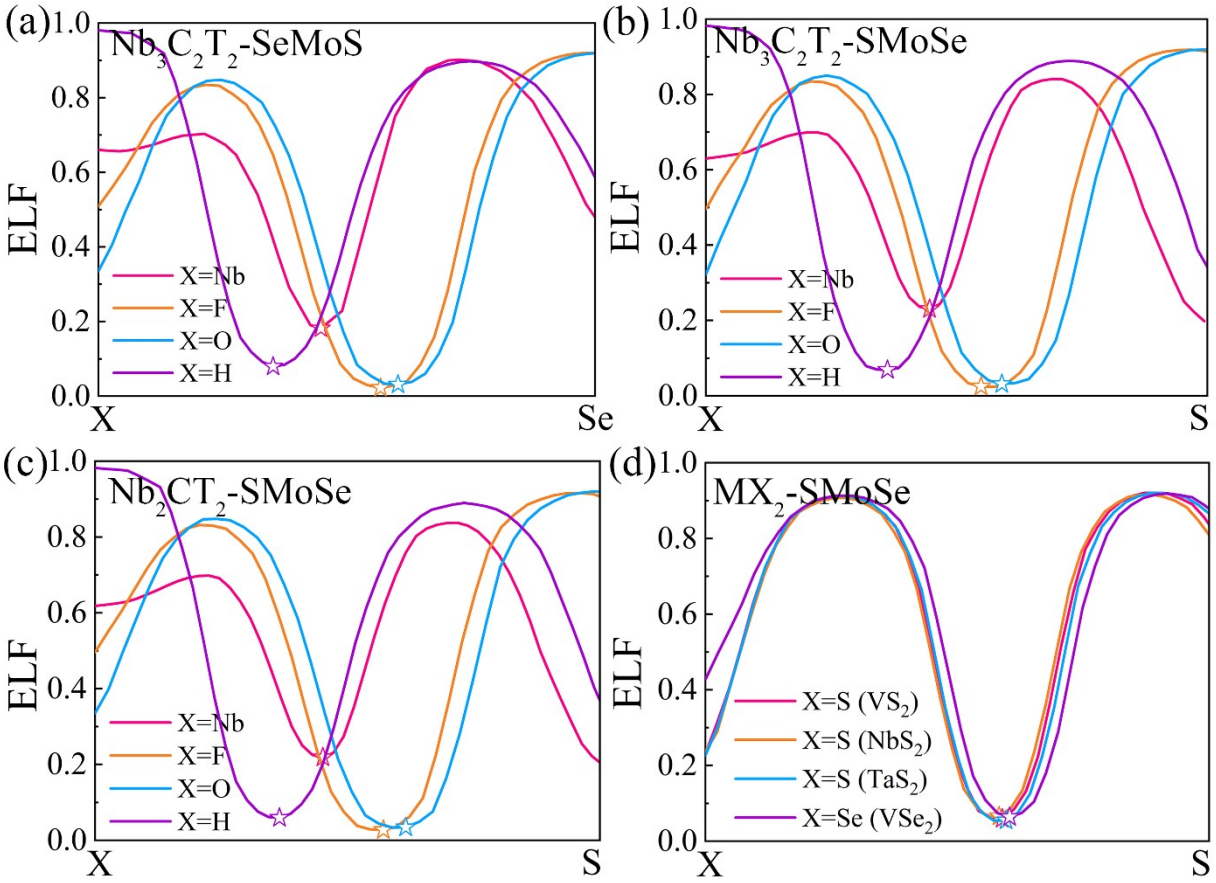

Fig. S4 The ELF 2D line profiles and bond points of the (a) X–Se and (b-d) X–S bonds in 2D metal-MoSSe contacts. The pentagrams represent the bond points of the X–Se and X–S bonds in 2D metal-MoSSe contacts.

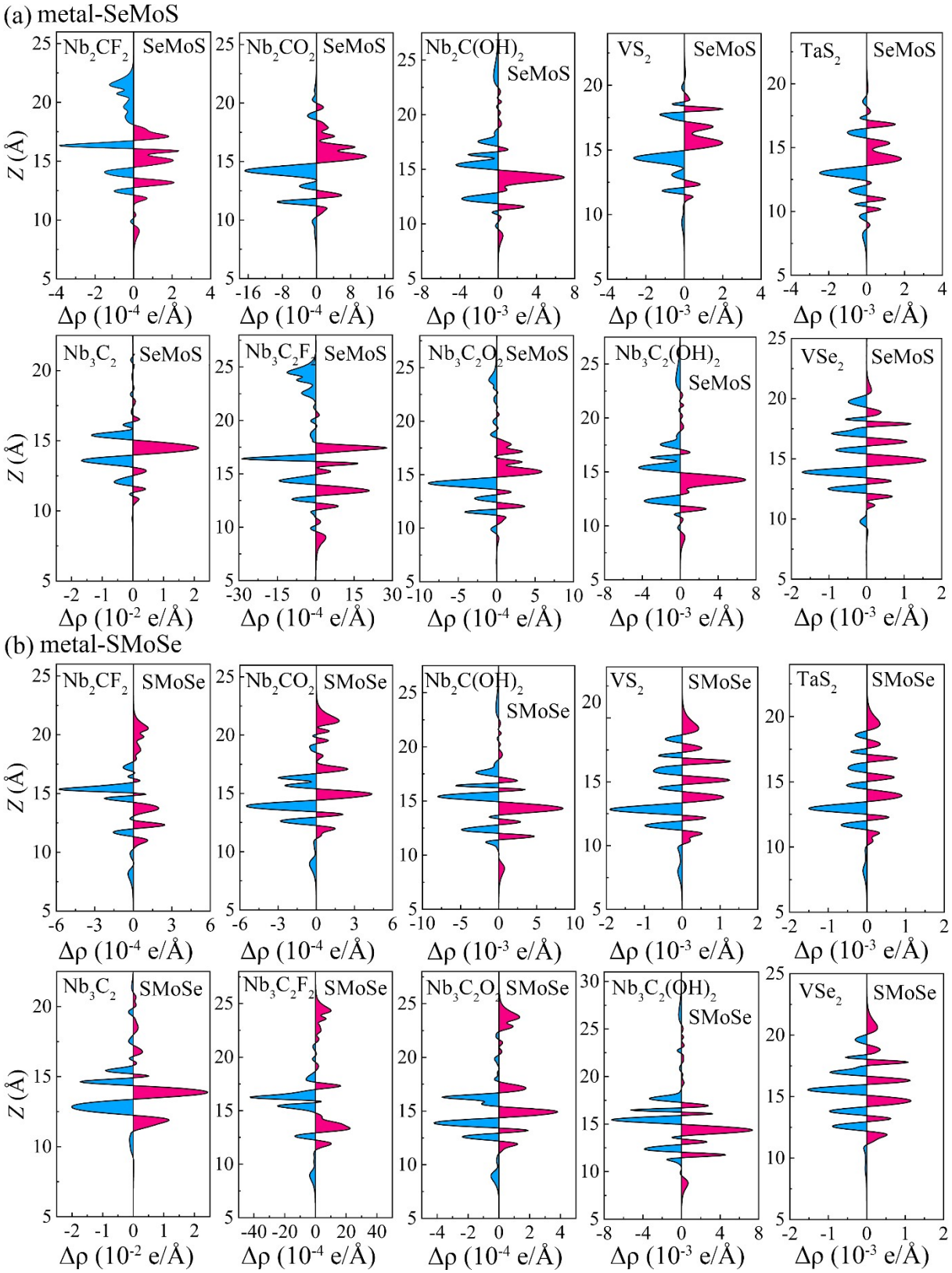

Fig. S5 The plane averaged charge density difference for (a) metal-SeMoS and (b) metal-SMoSe contacts. Pink indicates charge accumulation, azure indicates charge depletion.

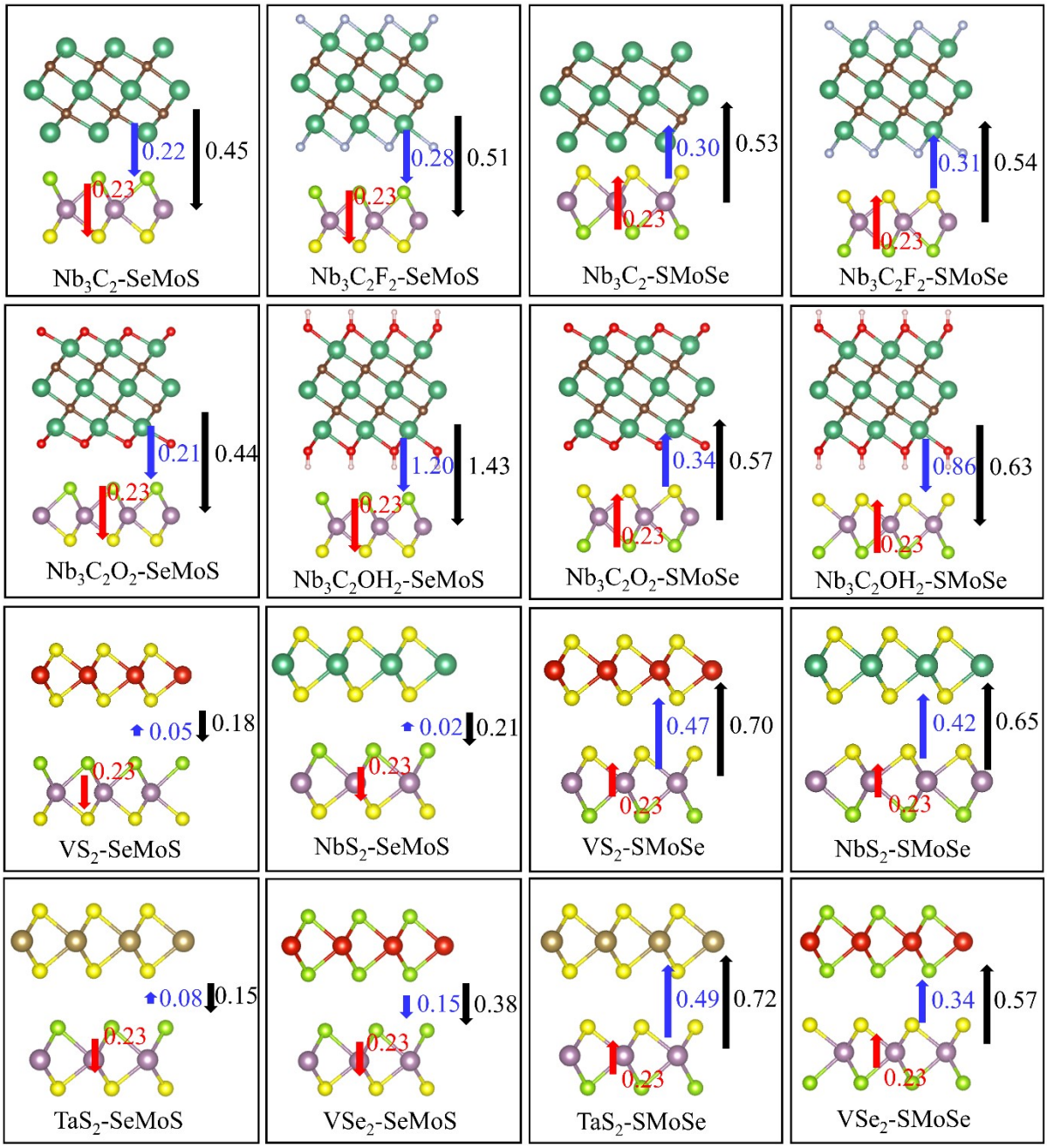

Fig. S6 The dipole moment distributions of 2D metal-MoSSe contacts. The red, blue and black arrows represent the intrinsic dipole, interface dipole and tot dipole, respectively.

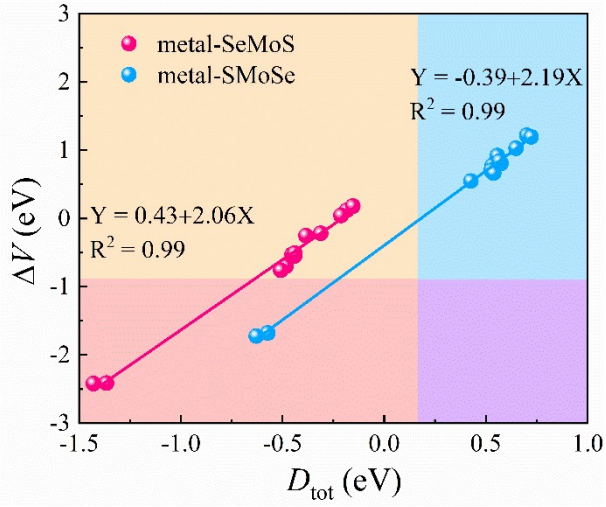

Fig. S7 The change of ΔV with tot dipole moment (D_{tot}) for 2D metal-MoSSe contacts.

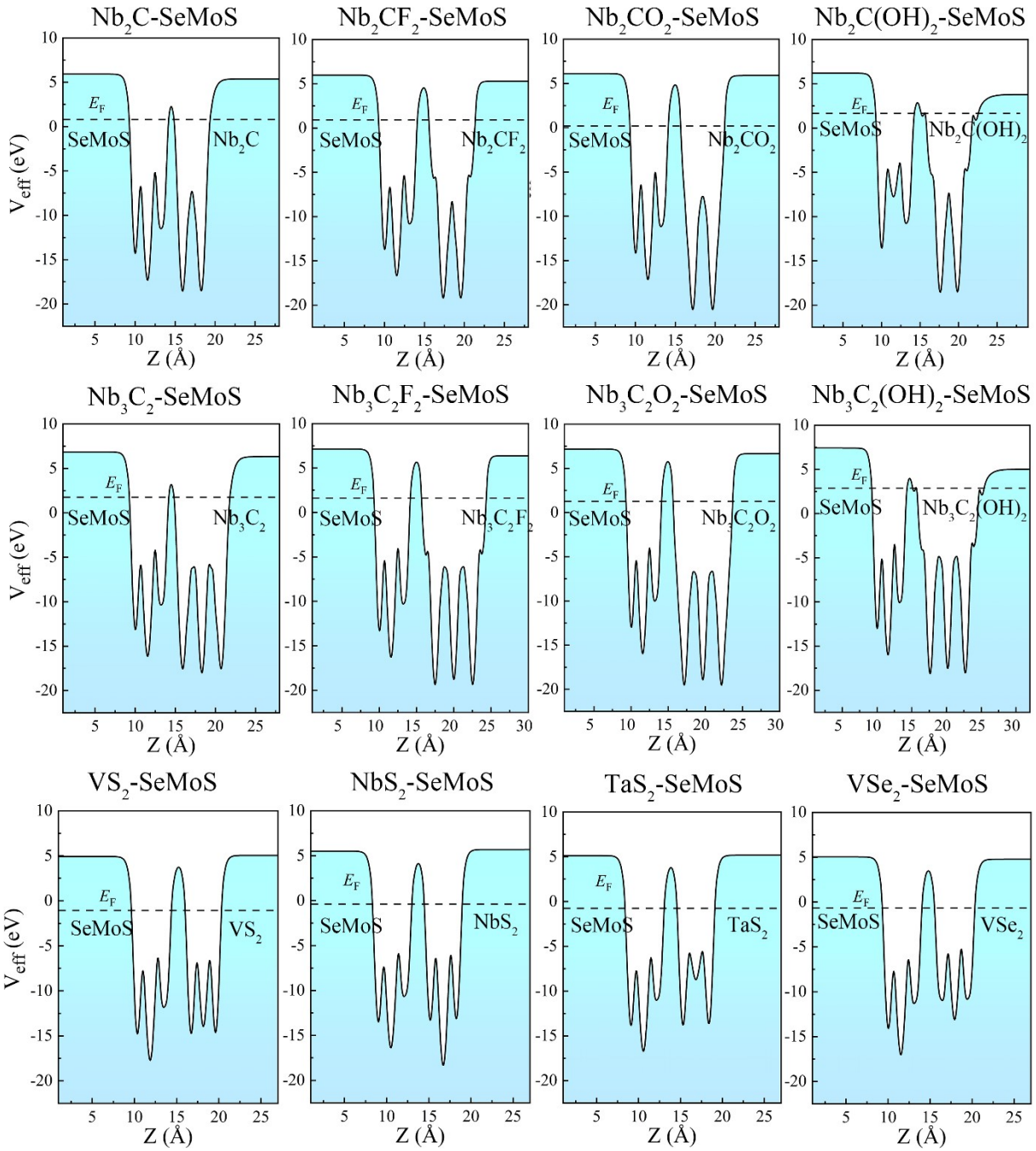

Fig. S8 Effective electrostatic potential of 2D metal-SeMoS contacts.

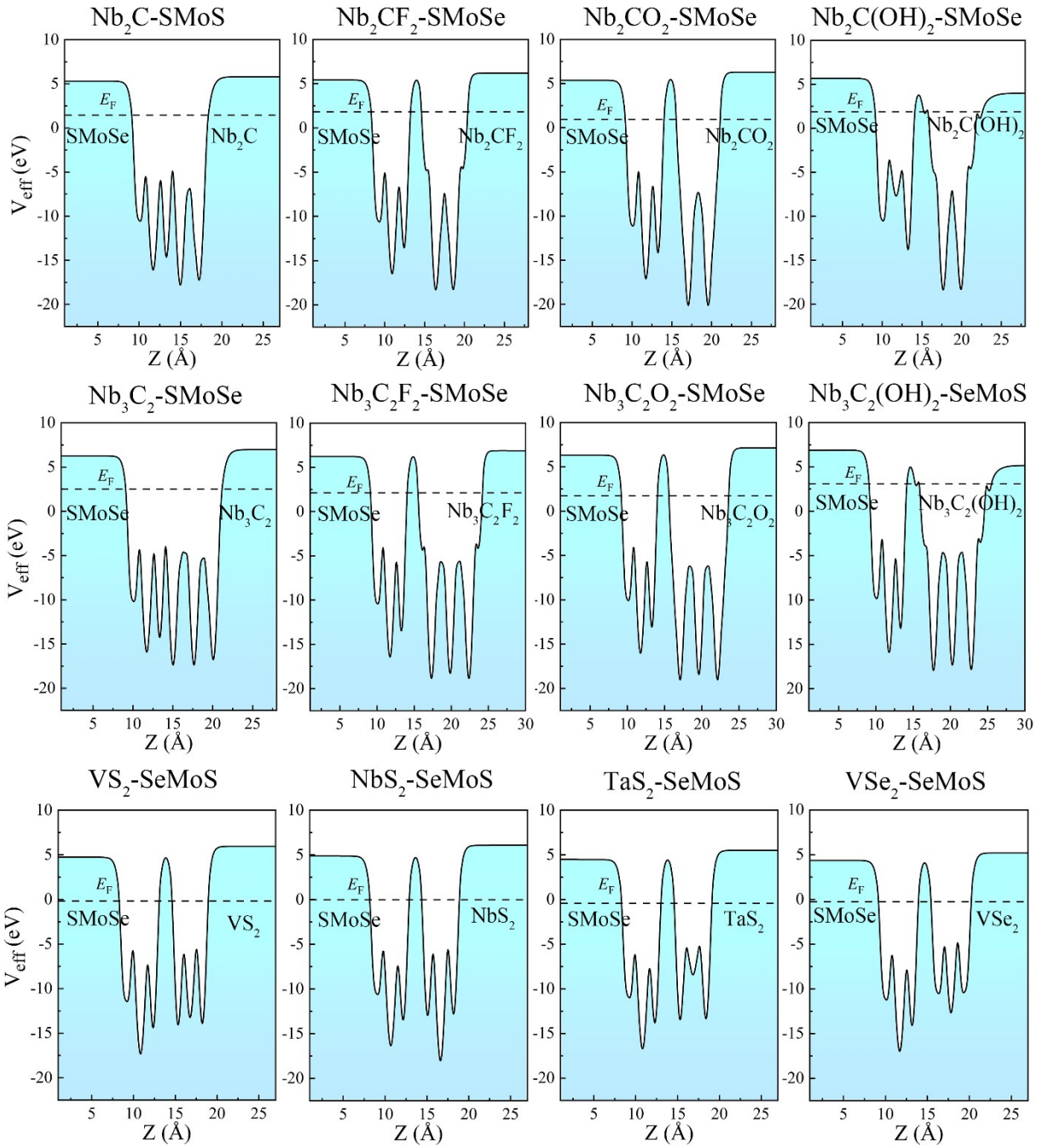

Fig. S9 Effective electrostatic potential of 2D metal-SMoSe contacts.

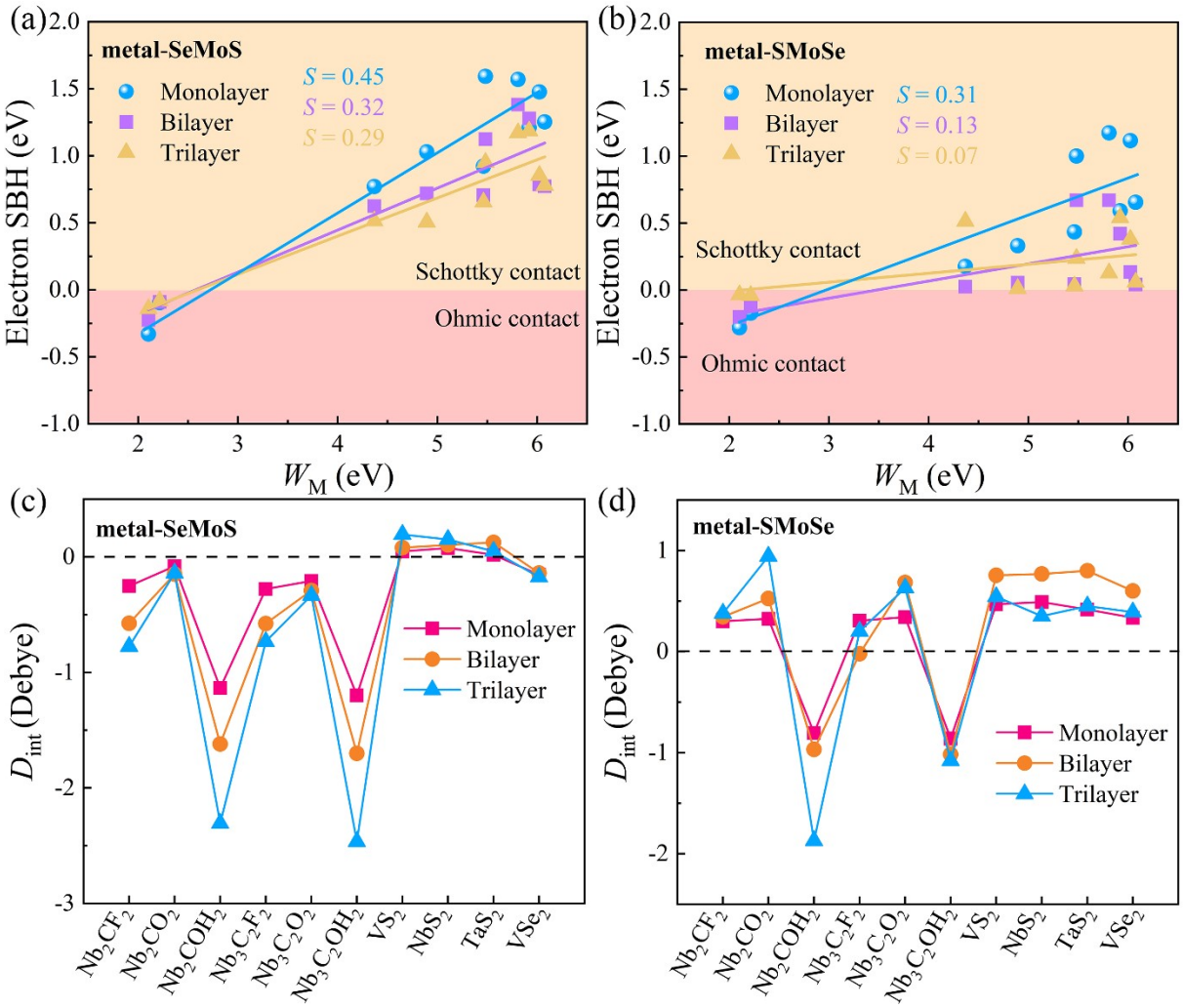

Fig. S10 The variation of electron SBH as a function of metal work functions (W_M) for (a) metal-SeMoS (b) metal- SMoSe contacts. The Fermi level pinning factor S is defined as the value of the fitted slope. Variation of the interface dipole (D_{int}) in the (c) metal-SeMoS and (d) metal-SMoSe contacts.

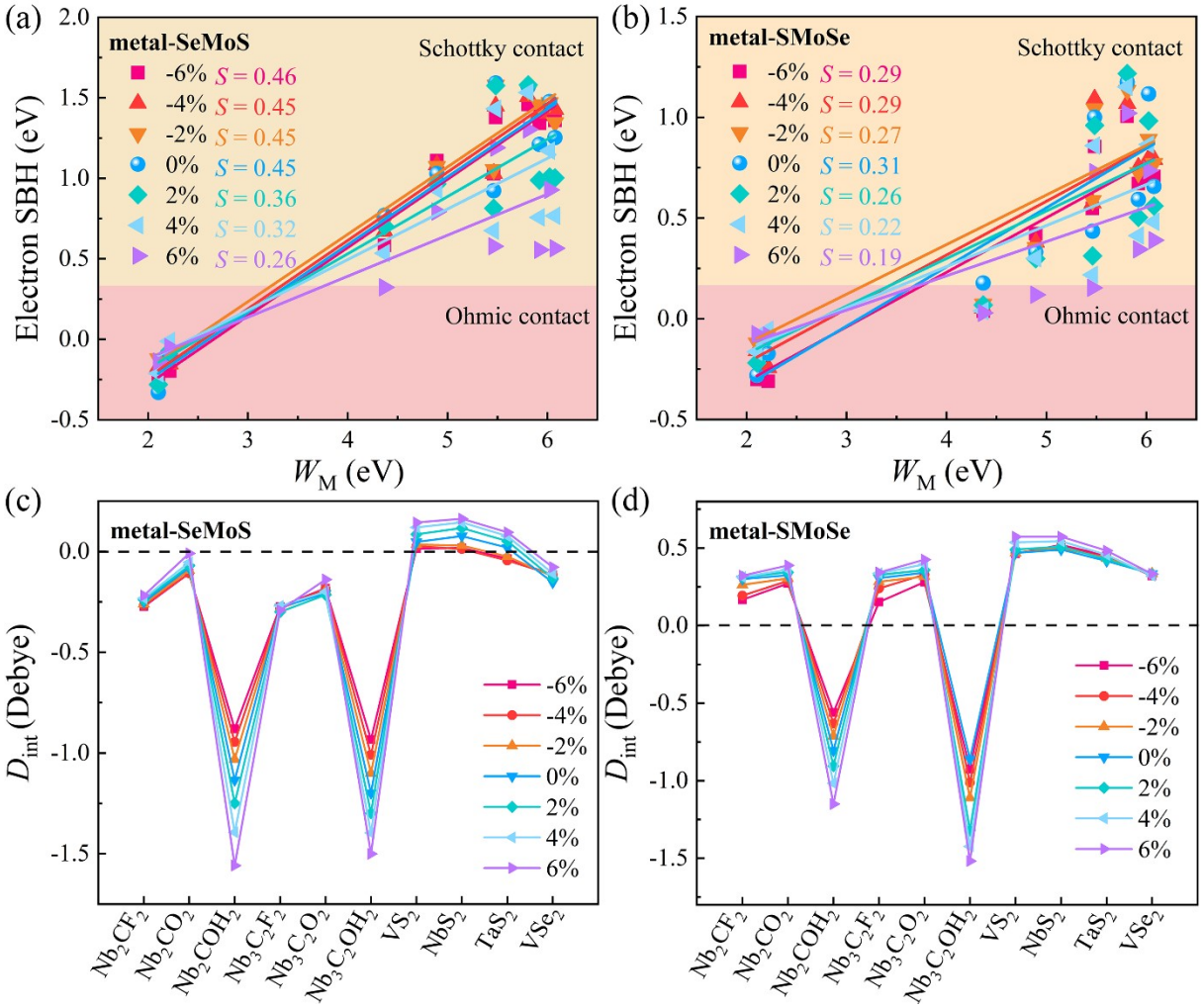

Fig. S11 The variation of electron SBH as a function of metal work functions (W_M) for (a) metal-SeMoS (b) metal-SMoSe contacts. The Fermi level pinning factor S is defined as the value of the fitted slope. Variation of the interface dipole (D_{int}) in the (c) metal-SeMoS and (d) metal-SMoSe contacts.

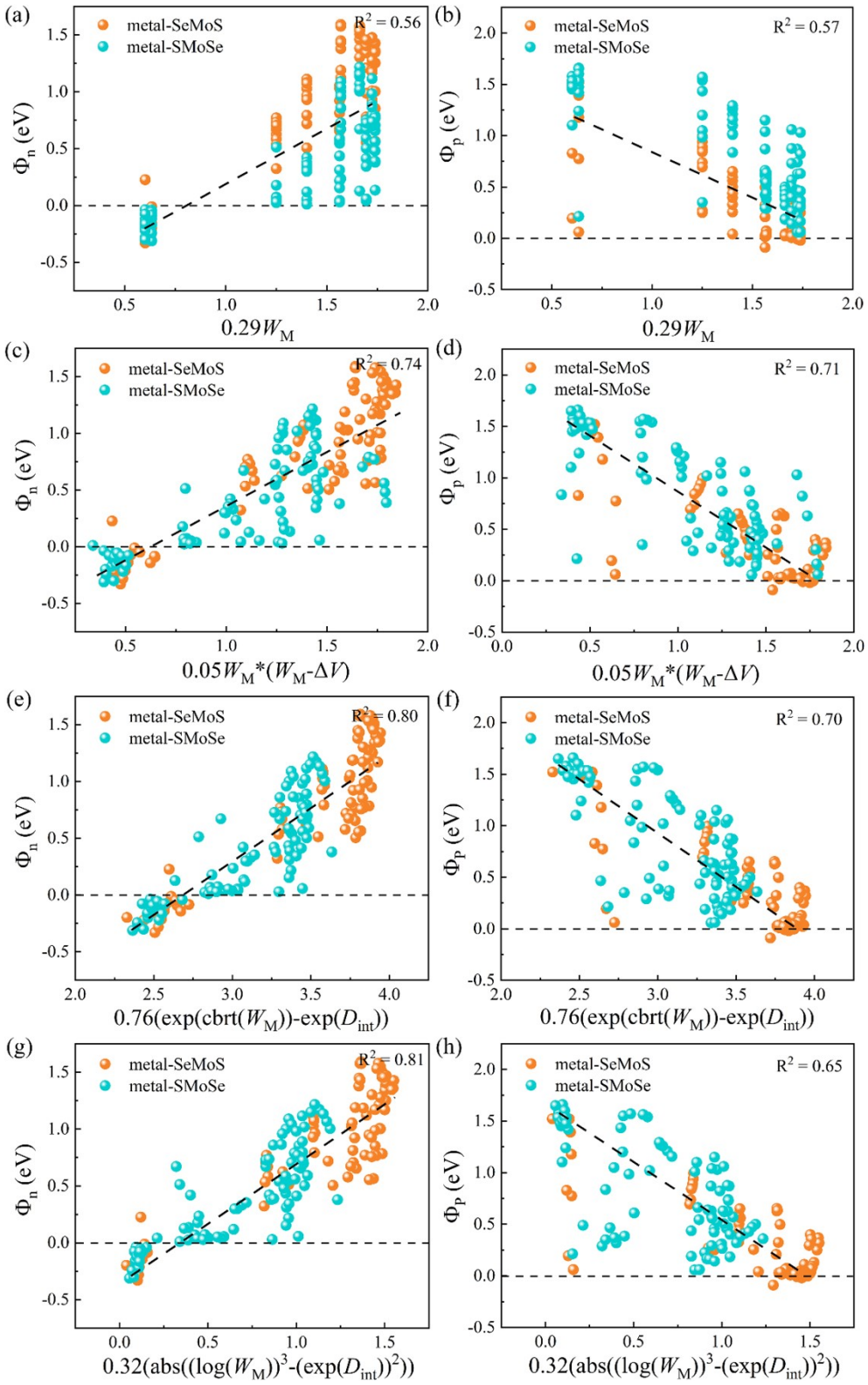

Fig. S12 The correlations of the calculated (a) Φ_n and (b) Φ_p values of metal-MoSSe contacts vs. predicted 0.29W_M formula from SISSO. The correlations of the calculated (c) Φ_n and (d) Φ_p values of metal-MoSSe contacts vs. predicted 0.05W_M *(W_M-ΔV) formula from SISSO. The correlations of the calculated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

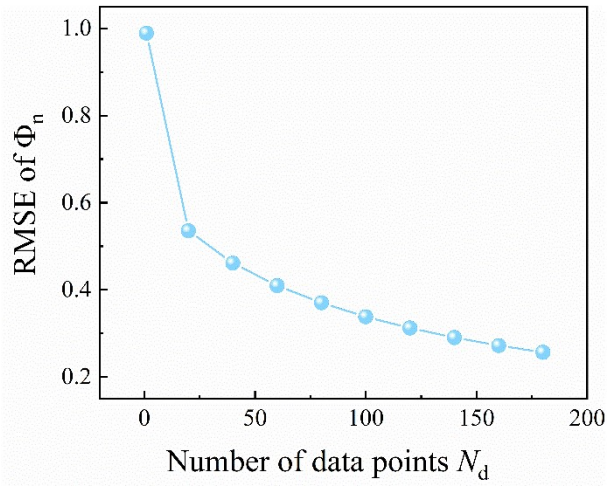

Fig. S13 The root-mean-square error (RMSE) of Φ_n is plotted as a function of the training data set size, N_d using the supervised data sampling method.