

Supporting Information

Mechanistic Understanding of the Interfacial Properties of Metal-PtSe₂ Contacts

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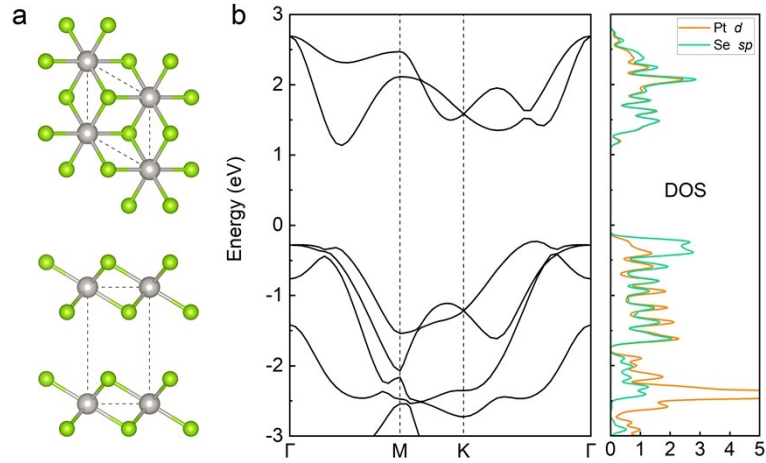


Figure S1. (a) Atomic structures of bulk PtSe₂ with 1T phase. (b) Band structures and density of states of monolayer PtSe₂.

Table S1. Total energy (eV) of possible stacking configurations for metal-PtSe₂ contacts.

Energy (eV)	Type I	Type II	Type III
Ag-PtSe ₂	-189.7475	-189.7505	-189.7764
Al-PtSe ₂	-220.0505	-220.0588	-220.0629
Au-PtSe ₂	-217.4000	-217.3996	-217.3914
Cu-PtSe ₂	-215.8513	-215.9233	-
Pd-PtSe ₂	-295.8074	-295.8145	-295.9051
Pt-PtSe ₂	-338.2266	-338.2190	-338.2775
Sc-PtSe ₂	-197.7589	-197.8269	-
Ti-PtSe ₂	-395.1130	-395.2772	-395.1684

Table S2. The structural parameters (Å) of the metal-PtSe₂ contacts. The values in parentheses represent the structural parameters of the metal-BL PtSe₂ contacts.

	Ag	Al	Au	Cu	Pd	Pt	Sc	Ti
lattice	7.59	7.56	7.58	6.65	7.41	7.44	6.57	7.67
constants	(7.59)	(7.56)	(7.58)	(6.65)	(7.41)	(7.44)	(6.57)	(7.67)
interlayer	2.49	2.42	2.58	2.19	2.17	2.26	2.13	2.07
distances	(2.46)	(2.37)	(2.55)	(2.17)	(2.15)	(2.34)	(2.11)	(2.03)

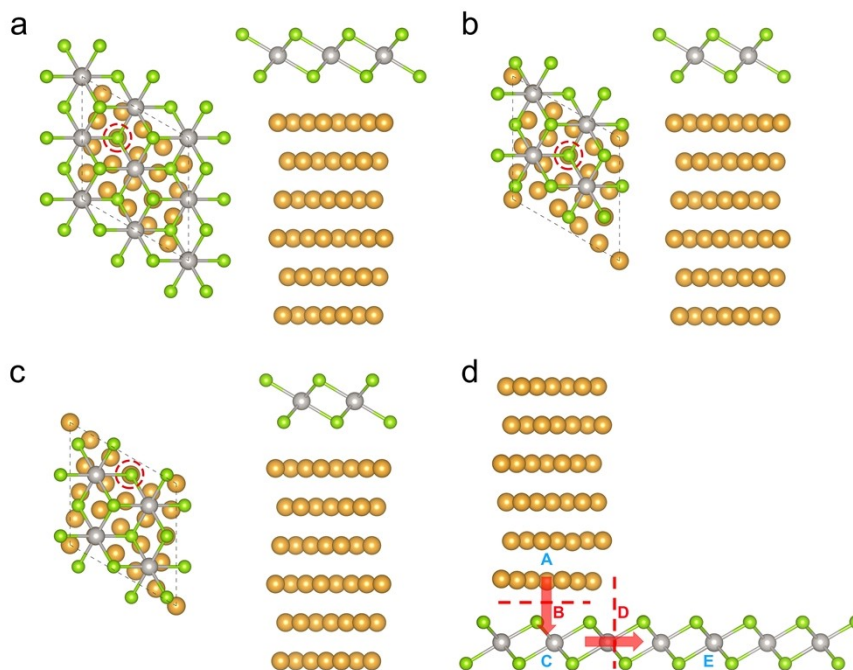


Figure S2. (a)-(c) Three different high symmetric stacking configurations of Au-PtSe₂ contacts. The red dotted circles in (a)-(c) represent the FCC hollow site, HCP hollow site and top site for the Se atom in the circle, respectively. (d) Schematic view of metal-PtSe₂ contacts. A, C, and E denote the metal, PtSe₂ at the region of contact and channel, respectively. B and D are the vertical and lateral interface, respectively. Red arrows represent the pathway of electron injection from metal to PtSe₂ channel.

Table S3. Binding energy E_b (J/m²) of metal-PtSe₂ contacts.

	Ag	Al	Au	Cu	Pd	Pt	Sc	Ti
E_b (ML)	1.27	0.97	1.23	1.82	2.08	2.00	2.25	2.61
E_b (BL)	1.36	1.07	1.31	1.94	2.22	2.15	2.43	2.83

Table S4. Binding energy E_b (eV) of Ag, Al, Au, Cu, Pd, Pt, Sc and Ti based metal-monolayer PtSe₂ contacts. Here, the binding energy are calculated as: $E_b = (E_{\text{metal}} + E_{\text{PtSe}_2} - E_{\text{heterostructure}})/N$ for comparison with previous studies.

E_b	Ag	Al	Au	Cu	Pd	Pt	Sc	Ti	Ref
PtSe₂	-0.99	-0.75	-0.96	-1.45	-1.55	-1.50	-1.75	-2.08	Our results
MoS ₂	-0.27	-0.24	-0.20	-	-0.69	-0.48	-	-	[1]
MoS ₂	-0.50	-	-0.31	-	-	-0.57	-1.18	-1.81	[2]
MoSe ₂	-0.13	-0.04	-	-	-	-0.29	-	-0.71	[3]
WS ₂	-0.62	-	-0.28	-0.76	-	-0.82	-1.27	-1.08	[4]
WSe ₂	-0.30	-0.29	-1.18	-	-0.60	-0.53	-0.92	-	[5]

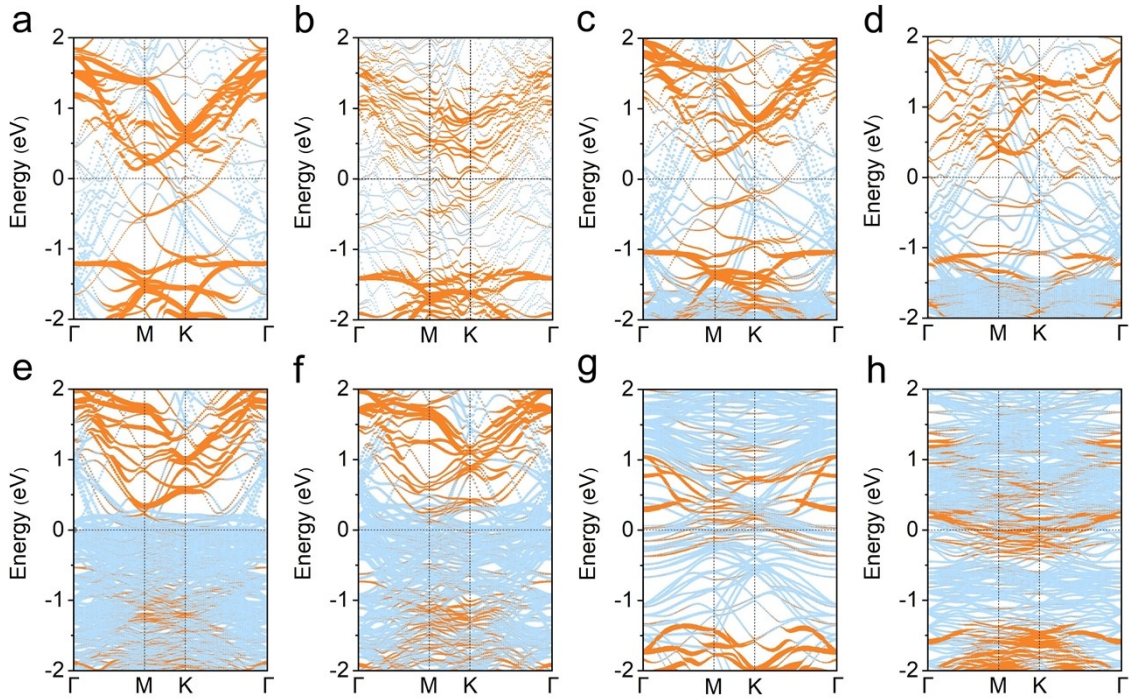


Figure S3. The projected band structures of Ag, Al, Au, Cu, Pd, Pt, Sc and Ti based metal-monolayer PtSe₂ contacts in (a)-(h). Orange and blue colors represent the contributions from PtSe₂ and metal, respectively.

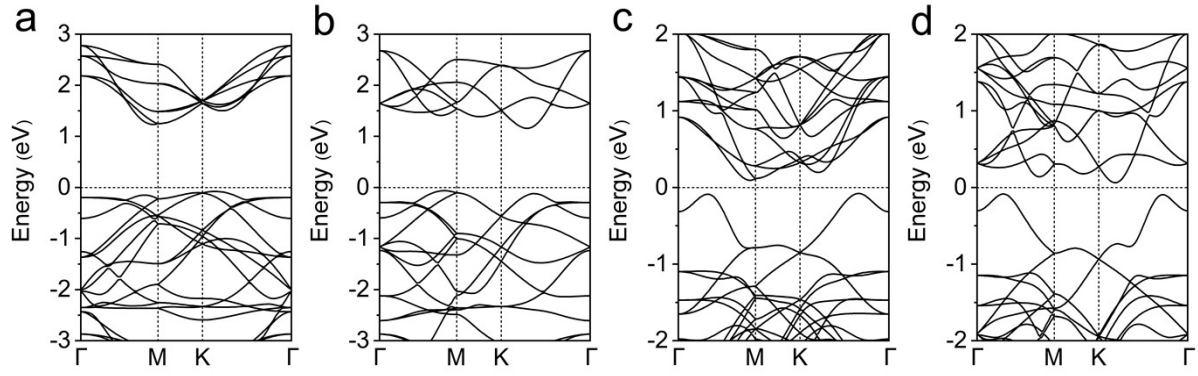


Figure S4. The band structures of original supercell PtSe₂ in metal-PtSe₂ contacts. (a) and (c) represent the cases of ML and BL PtSe₂ with Ag, Al, Au, Pd, Pt and Ti, respectively. (b) and (d) represent the cases of ML and BL PtSe₂ with Cu and Ti, respectively.

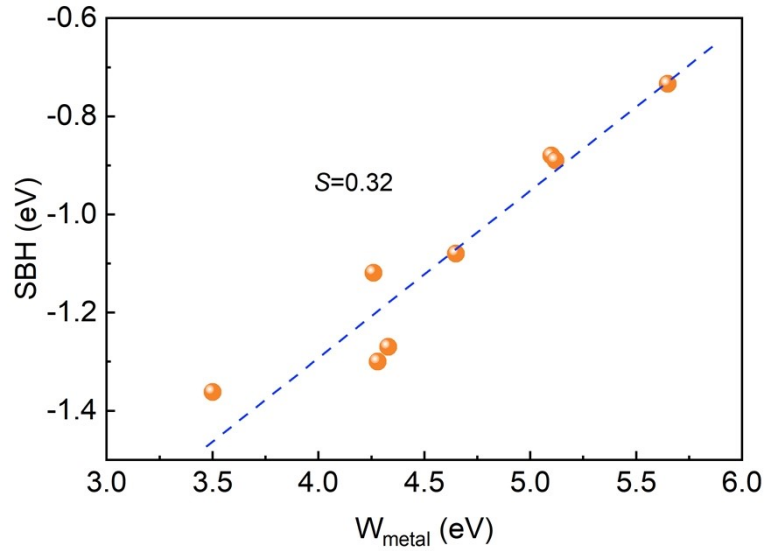


Figure S5. The fitting of FLP factor for Ag, Al, Au, Cu, Pd, Pt, Sc and Ti based metal-monolayer PtSe₂ contacts.

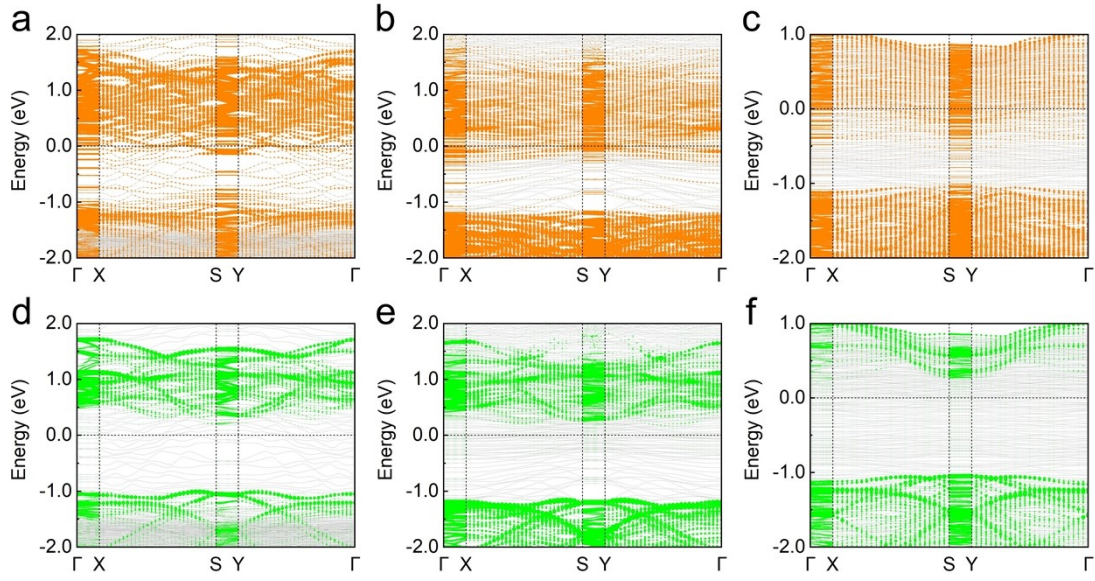


Figure S6. Band structure of PtSe₂ in the large supercell model at the vertical metal contact region (indicated by orange) and the lateral free-standing PtSe₂ region (indicated by green) for Cu ((a) and (d)), Sc ((b) and (e)) and Ti ((c) and (f)) based systems. (noting that the Fermi level is same in the corresponding systems, respectively)

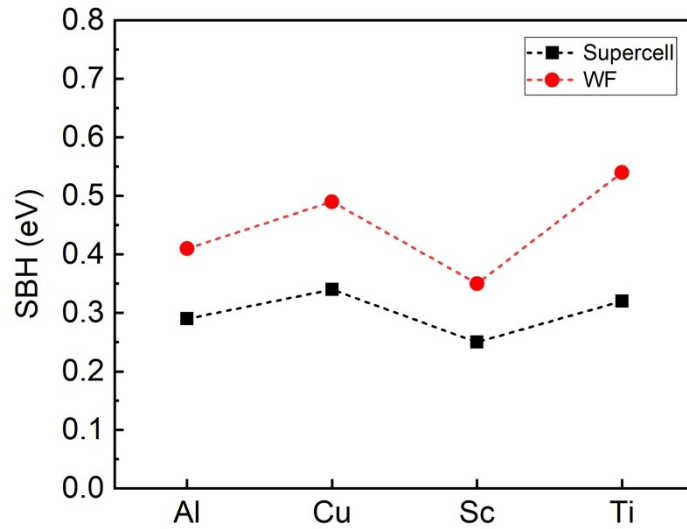


Figure S7. Schottky barrier height of the metal-PtSe₂ systems based on the work function approximation and large supercell model.

Table S5. The actual distance d (Å) between metal atoms and interfacial Se atoms as well as the covalent distance (sum of the covalent radii of metal atom and Se atom).

	Ag	Al	Au	Cu	Pd	Pt	Sc	Ti
d	2.85	2.78	3.06	2.58	2.53	2.59	2.75	2.64
	2.84	2.77	2.66	2.56	2.53	2.58	2.70	2.47
	2.82	2.75	2.66	2.33	2.52	2.58	2.56	2.47
	2.58	2.51	2.66	-	2.35	2.37	-	2.47
shortest d	2.58	2.51	2.66	2.33	2.35	2.37	2.56	2.47
average d	2.77	2.70	2.76	2.49	2.48	2.53	2.67	2.51
covalent distance	2.50	2.34	2.50	2.33	2.44	2.46	2.60	2.48

Table S6. The tunneling barrier and tunneling probability for the case of van der Waals interlayer distance (3.5Å) between metal and PtSe₂ in metal-PtSe₂ contacts.

vdW distance	ΔV	Δd	TB
Au	4.37	1.78	2.21%
Cu	4.13	1.88	2.00%
Sc	3.14	1.58	5.69%
Ti	3.60	1.66	3.97%

Table S7. Tunneling barrier height Φ_{TB} (eV), width d_{TB} (Å), tunneling probability (%) and tunneling-specific resistivity ($\Omega \text{ cm}^2$) of metal-monolayer PtSe₂ contacts.

	Φ_{TB}	d_{TB}	Probability	Resistivity
Ag	1.69	0.71	38.63	3.00E-11
Al	0.93	0.50	61.04	1.80E-11
Au	2.22	0.80	29.38	3.86E-11
Cu	0.69	0.38	72.65	1.33E-11
Pd	0.79	0.33	74.08	1.06E-11
Pt	0.98	0.39	67.47	1.23E-11
Sc	0.00	0.00	100.00	0
Ti	0.00	0.00	100.00	0

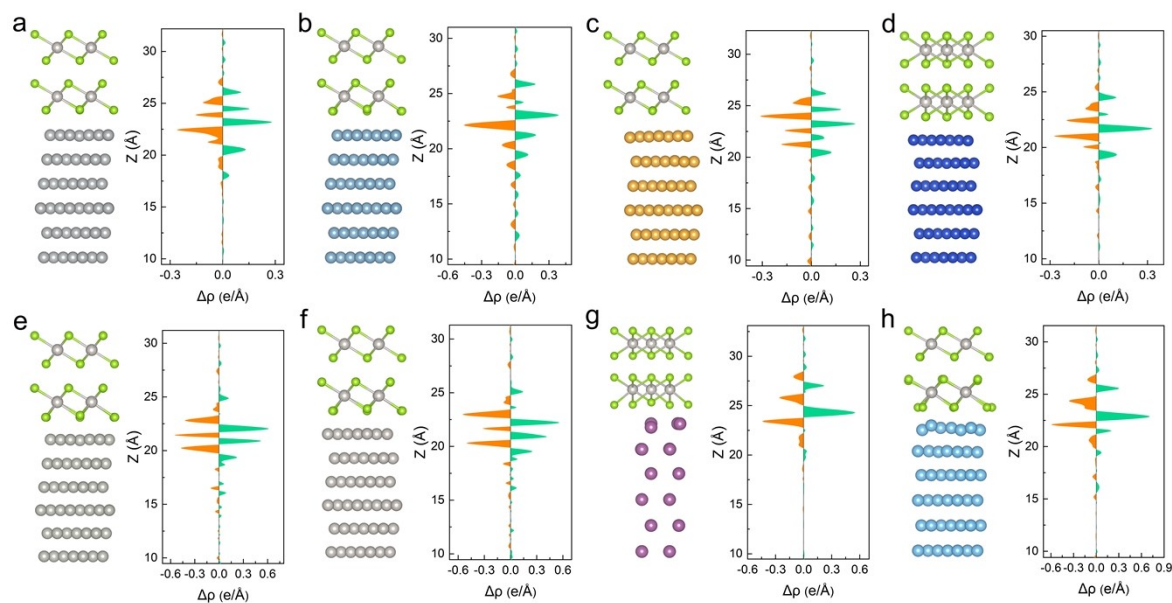


Figure S8. Side view of the optimized atomic structures and plane charge density differences for Ag, Al, Au, Cu, Pd, Pt, Sc and Ti based metal-bilayer PtSe₂ contacts in (a)-(h), respectively.

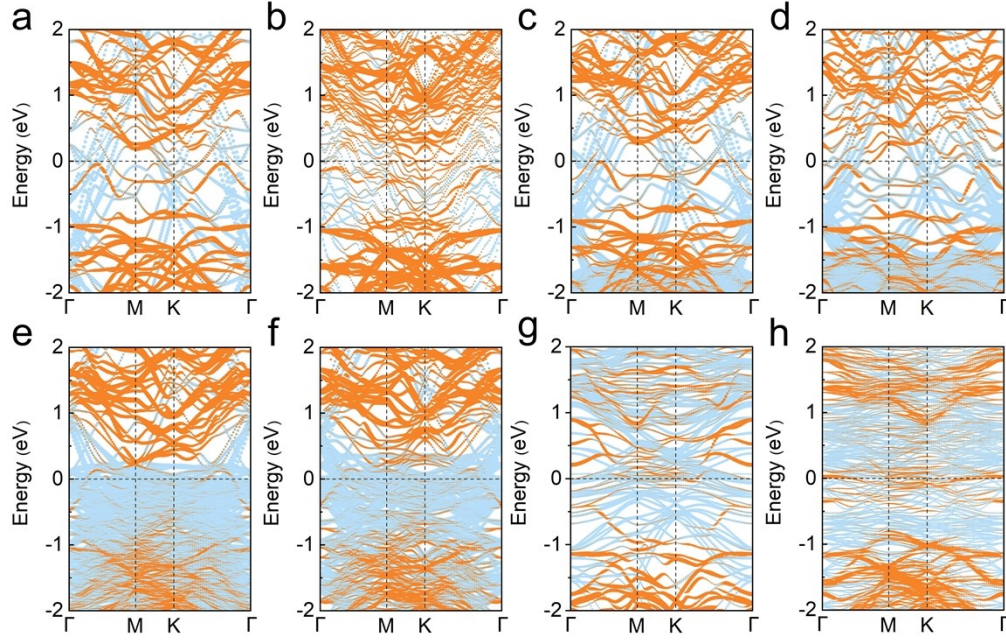


Figure S9. Projected band structures of Ag, Al, Au, Cu, Pd, Pt, Sc and Ti based metal-bilayer PtSe₂ contacts in (a)-(h).

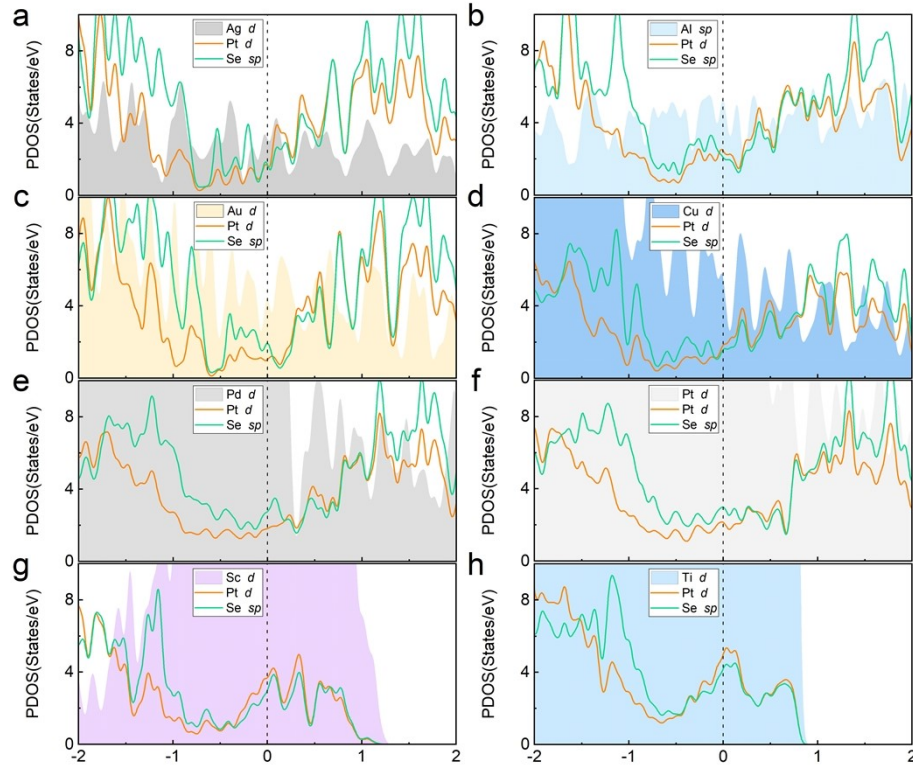


Figure S10. Partial density of states for PtSe₂ and metal in Ag, Al, Au, Cu, Pd, Pt, Sc and Ti based metal-bilayer PtSe₂ contacts in (a)-(h).

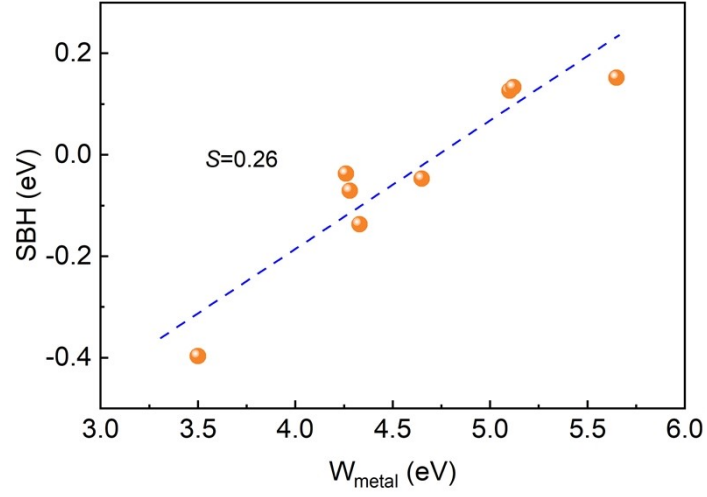


Figure S11. The fitting of FLP factor for Ag, Al, Au, Cu, Pd, Pt, Sc and Ti based metal-bilayer PtSe₂ contacts.

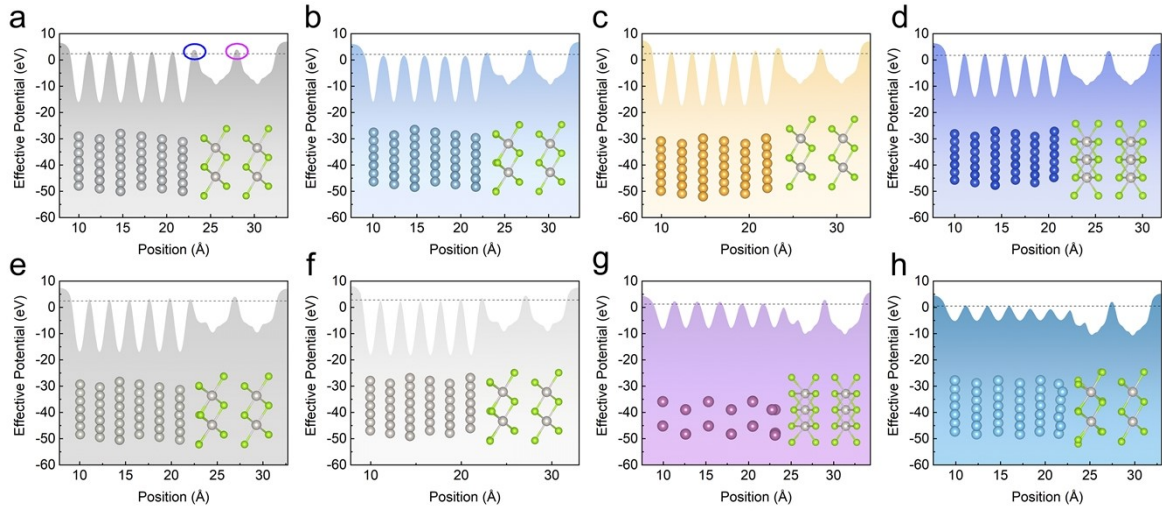


Figure S12. Effective potential along the vertical position for Ag, Al, Au, Cu, Pd, Pt, Sc and Ti based metal-bilayer PtSe₂ contacts in (a)-(h).

Table S8. Tunneling barrier height Φ_{TB} (eV), width d_{TB} (Å), tunneling probability (%) and tunneling-specific resistivity ($\Omega \text{ cm}^2$) of the interface between metal and first-layer PtSe₂ and the interface between the first-layer and second-layer in metal-monolayer PtSe₂ contacts.

	Metal-PtSe ₂				First layer PtSe ₂ -Second layer PtSe ₂			
	Φ_{TB}	d_{TB}	Probability	Resistivity	Φ_{TB}	d_{TB}	Probability	Resistivity
Ag	1.571	0.667	42.48	2.64E-11	1.647	0.649	42.62	2.50E-11
Al	0.697	0.416	70.07	1.50E-11	1.744	0.674	40.19	2.68E-11
Au	2.166	0.785	30.63	3.67E-11	1.763	0.682	39.56	2.74E-11
Cu	0.629	0.353	75.08	1.25E-11	1.685	0.628	43.40	2.35E-11
Pd	0.706	0.299	77.32	0.97E-11	1.650	0.659	42.03	2.58E-11
Pt	0.852	0.348	71.97	1.11E-11	1.678	0.666	41.34	2.62E-11
Sc	0.000	0.000	100.00	0	1.646	0.63	43.71	2.37E-11
Ti	0.000	0.000	100.00	0	1.571	0.601	46.24	2.19E-11
PtSe ₂	-	-	-	-	0.976	0.389	40.33	2.67E-11

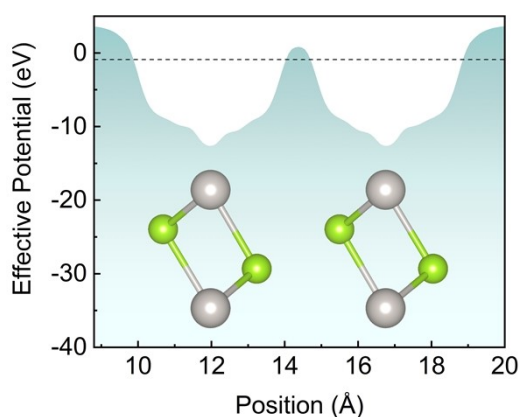


Figure S13. Effective potential along the vertical direction for bilayer PtSe₂. The dotted line represents the Fermi energy level.

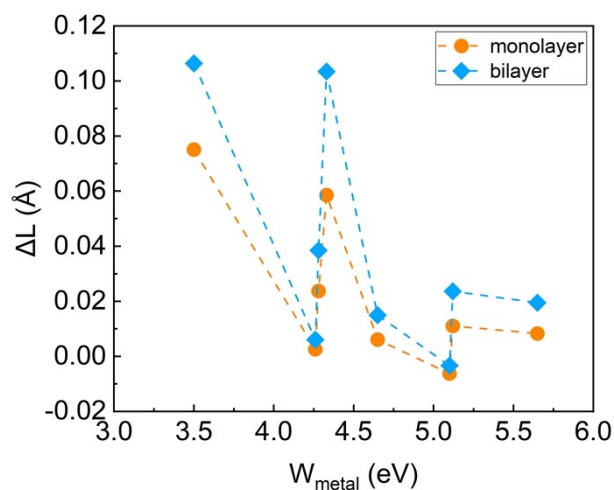


Figure S14. The change of the average bond length of the interfacial Se atom and Pt atom in PtSe₂ versus the work function of metals.

Table S9. The lattice parameters a (Å) and interfacial distance (Å) of metal-BN-PtSe₂ contacts.

	Ag	Al	Au	Pd	Pt	Ti
a	7.59	7.56	7.58	7.41	7.44	7.67
$d_{\text{Metal-BN}}$	3.16	3.57	3.23	2.97	3.16	2.18
$d_{\text{BN-PtSe}_2}$	3.36	3.38	3.34	3.35	3.34	3.28
$d_{\text{Metal-PtSe}_2}$	6.52	6.95	6.57	6.32	6.50	5.46

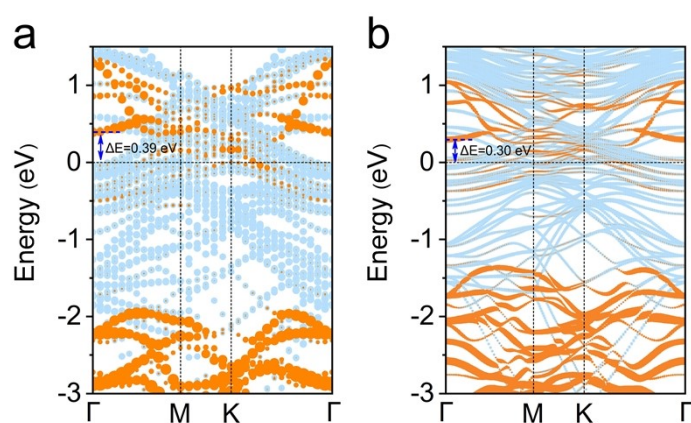


Figure S15. Comparison of the HSE06-based band structure (a) and the PBE-based band structure (b) of Sc-PtSe₂ contact. ΔE represents the energy difference between the Fermi level energy and one major conduction band of PtSe₂.

Table S10. The lattice mismatch and introduced strain for Metal-PtSe₂ contacts.

	Ag	Al	Au	Cu	Pd	Pt	Sc	Ti
Mismatch (%)	1.41	0.48	1.06	3.51	3.56	2.67	1.31	3.46
Strain (%)	0.71	0.24	0.53	1.79	-1.75	-1.32	0.66	1.76

References

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- (5) Wang, Y.; Yang, R. X.; Quhe, R.; Zhong, H.; Cong, L.; Ye, M.; Ni, Z.; Song, Z.; Yang, J.; Shi, J. et al. Does p-type ohmic contact exist in WSe₂-metal interfaces? *Nanoscale* **2016**, *8* (2), 1179.