Supporting Information

First Principles Study on Thickness Dependent Structural and Electronic Properties Unveiling Growth and Stability of 2D Layered II-VI Semiconducting Compounds.

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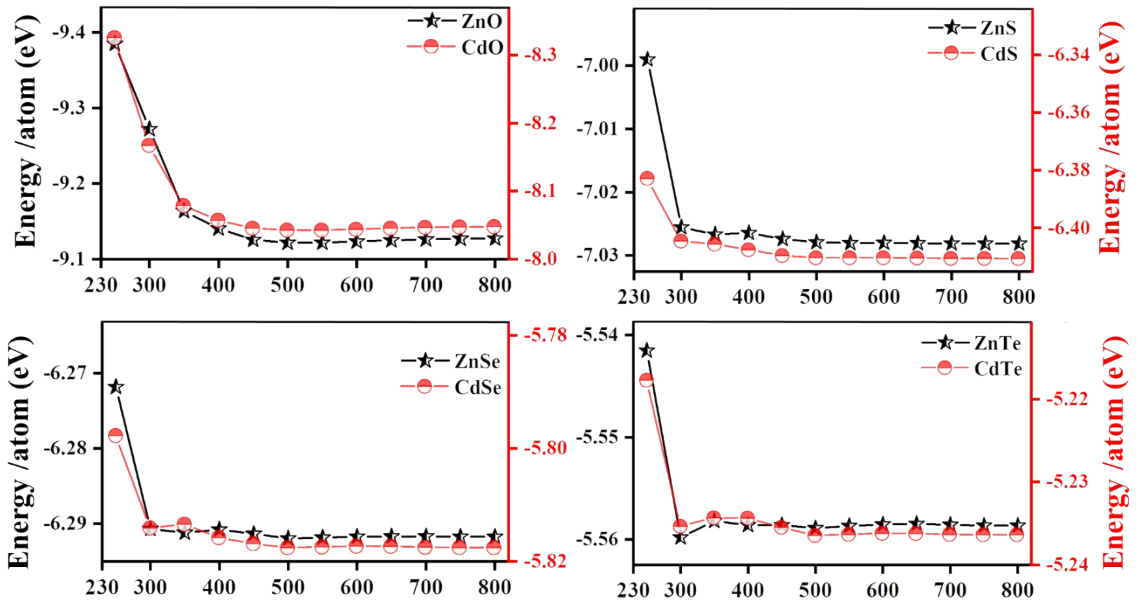


Fig. S1 : Cut-off energy optimization for WZ phase of MO and MX compounds.

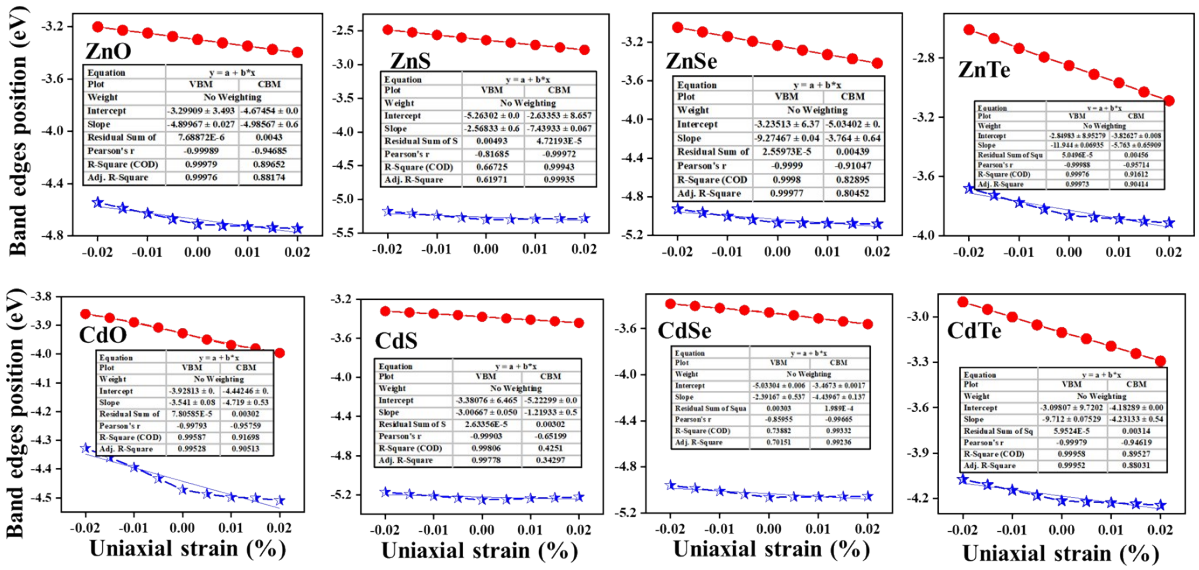


Fig. S2: CBM (red line) and VBM (blue line) as a function of uniaxial strain of MX 2L (high symmetry direction Γ→K)

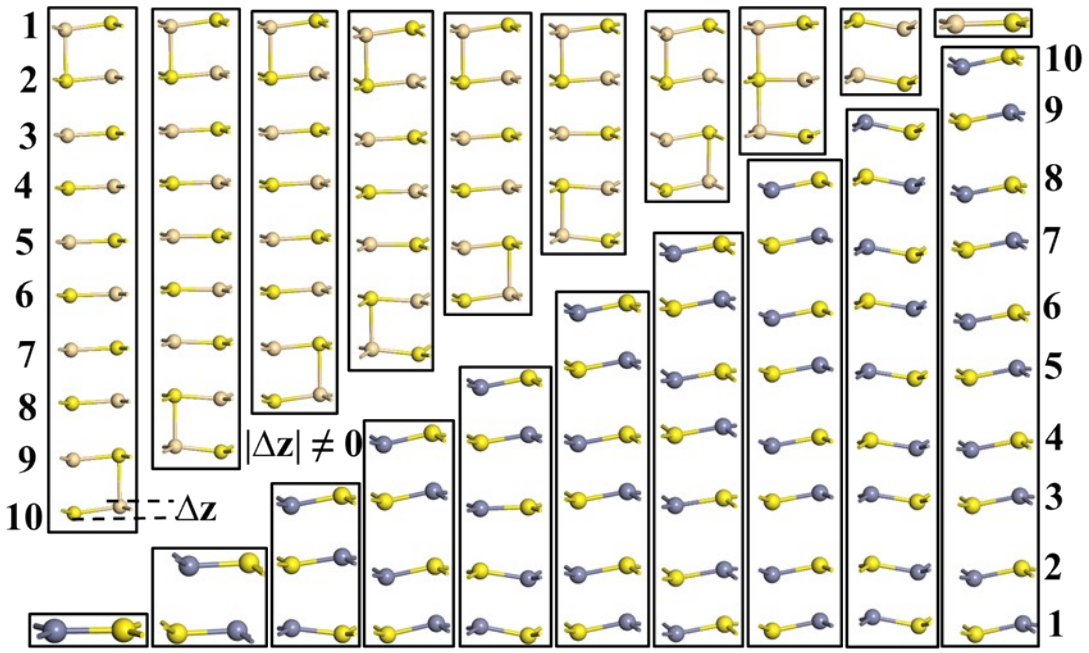


Fig. S3: Ball and stick model of optimized structures of various thick (1L to 10L) ZnS (lower diagonal) and CdS (upper diagonal) surface slabs. Grey, sandal and yellow coloured ball represent Zn, Cd and S atoms, respectively.

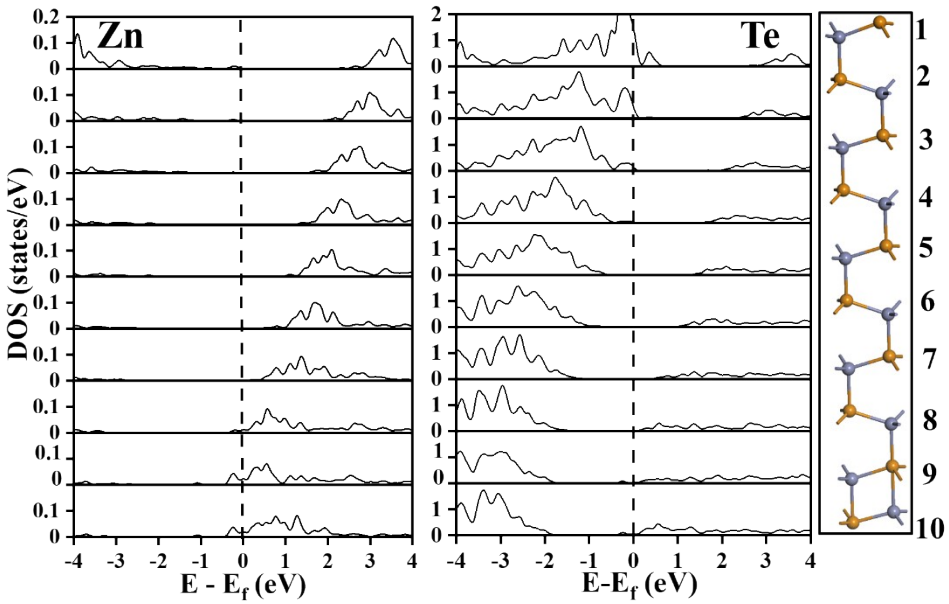


Fig. S4 : Layer resolved DOS of 10L thick WZ slab of ZnTe compound is given. For clarity, atomic structure is provided in right panel where layers are indexed. This figure clearly indicates the holes and electrons are spatially separated in the surface slab. This kind of feature can be seen in WZ slabs of these compounds.

Table S1 : Typical Zn-X bond distances (in Å) along xy-axis of various thick (1L to 10L) of 2D-MX surface slabs. For comparsion, bulk bond distances are given at last row.

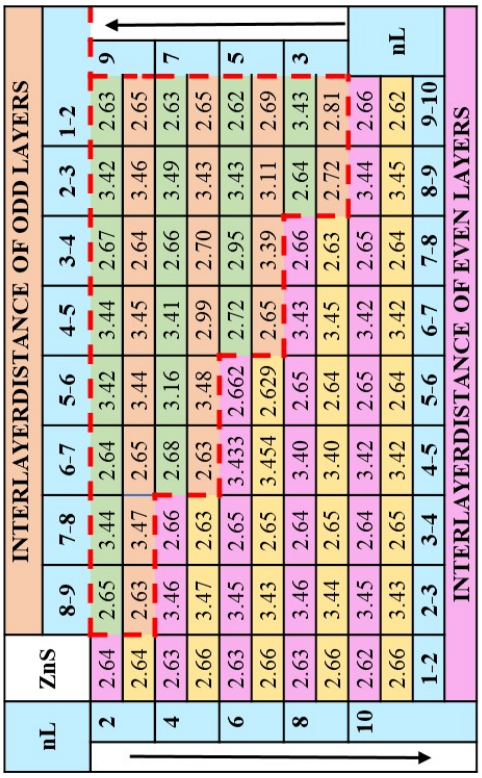
nL	Zn-O	Zn-S	Zn-Se	Zn-Te
1	1.896	2.236	2.364	2.563
2	1.925	2.303	2.448	2.649
3	1.935	2.269 - 2.293	2.383 - 2.439	2.567 - 2.647
4	1.942	2.297 - 2.303	2.444 - 2.447	2.647 - 2.648
5	1.946	2.258 - 2.303	2.383 - 2.446	2.566 - 2.654
6	1.949	2.295 - 2.305	2.443 -2.449	2.648 - 2.650
7	1.952	2.304 - 2.277	2.375 - 3.449	2.599 - 2.671
8	1.954	2.294 - 2.304	2.385 - 2.469	2.648 - 2.650
9	1.955	2.295 - 2.305	2.383 - 2.449	2.564 - 2.652
10	1.956	2.295 - 2.306	2.390 - 2.455	2.646 - 2.648
Bulk	1.99	2.32	2.45	2.63

Table S2 : Typical Cd-X bond distances (in Å) along xy-axis of various thick (1L to 10L) of 2D-MX surface slabs. For comparsion, bulk bond distances are given at last row.

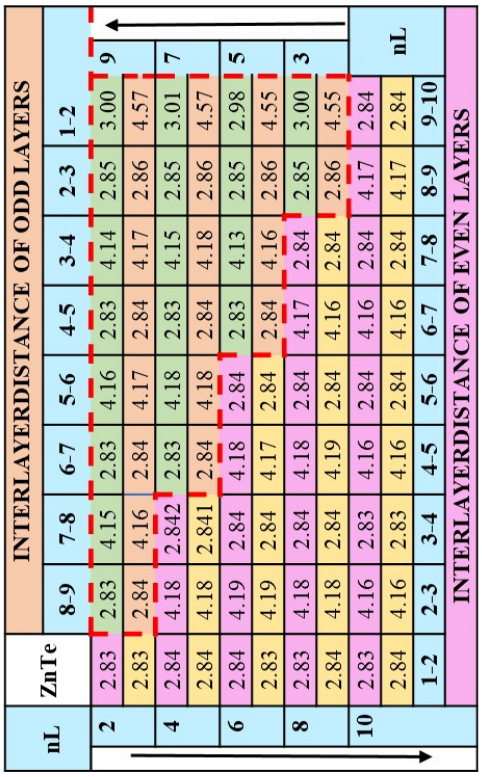
nL	Cd-O	Cd-S	Cd-Se	Cd-Te
1	2.115	2.446	2.572	2.760
2	2.153	2.517	2.649	2.840
3	2.170	2.497 - 2.510	2.594 - 2.646	2.758 - 2.840
4	2.179	2.513 - 2.525	2.642-6.50	2.840 - 2.842
5	2.185	2.508 - 2.528	2.589 - 2.658	2.756 - 2.844
6	2.190	2.512 - 2.531	2.639 - 2.652	2.838 - 2.842
7	2.193	2.516 - 2.537	2.587 - 2.666	2.756 - 2.844
8	2.196	2.518 - 2.538	2.640 - 2.655	2.801 - 2.868
9	2.198	2.519 - 2.540	2.597-2.662	2.743 - 2.848
10	2.199	2.521 - 2.540	2.643 - 2.656	2.800 - 2.873
Bulk	2.23	2.55	2.68	2.81

In these Tables (S1 & S2), M-X bond distances, along xy axis of layers, in odd nL are observed to varied significanly as compared to even nL, owing to instability of former case.

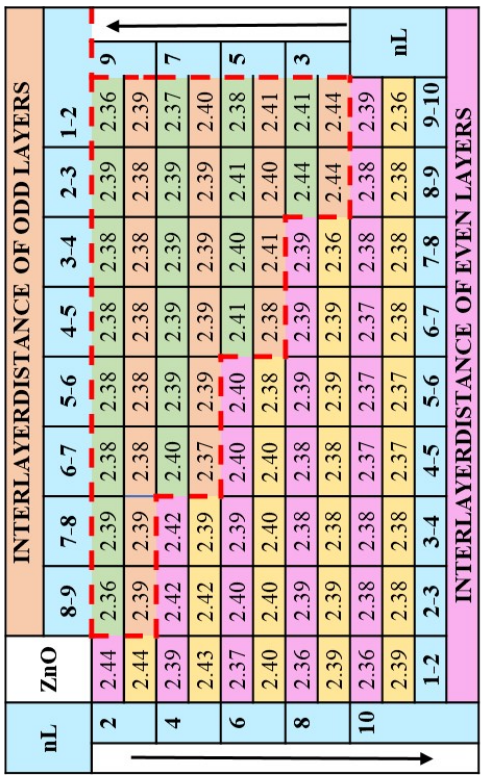
Table S3 : Interlayer distances (in Å) of various thick (nL) of 2D-ZnX surface slabs are reported. Left and right diagonal panels correspond to even and odd number layered surface slabs, repectively.



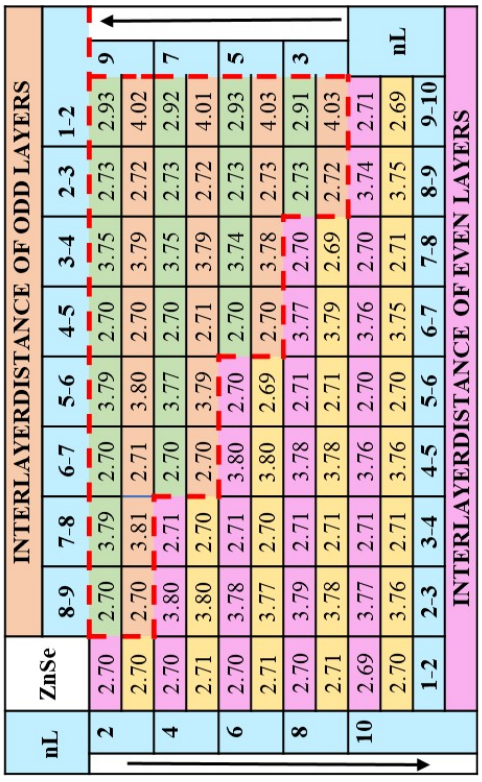
nL	ZnS	INTERLAYERDISTANCE OF ODD LAYERS									
		8-9	7-8	6-7	5-6	4-5	3-4	2-3	1-2		
2	2.64	2.65	3.44	2.64	3.42	3.44	2.67	3.42	2.63	9	
	2.64	2.63	3.47	2.65	3.44	3.45	2.64	3.46	2.65		
4	2.63	3.46	2.66	2.68	3.16	3.41	2.66	3.49	2.63	7	
	2.66	3.47	2.63	2.63	3.48	2.99	2.70	3.43	2.65		
6	2.63	3.45	2.65	3.433	2.662	2.72	2.95	3.43	2.62	5	
	2.66	3.43	2.65	3.454	2.629	2.65	3.39	3.11	2.69		
8	2.63	3.46	2.64	3.40	2.65	3.43	2.66	2.64	3.43	3	
	2.66	3.44	2.65	3.40	2.64	3.45	2.63	2.72	2.81		
10	2.62	3.45	2.64	3.42	2.65	3.42	2.65	3.44	2.66	nL	
	2.66	3.43	2.65	3.42	2.64	3.42	2.64	3.45	2.62		
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10	
		INTERLAYERDISTANCE OF EVEN LAYERS									



nL	ZnTe	INTERLAYERDISTANCE OF ODD LAYERS									
		8-9	7-8	6-7	5-6	4-5	3-4	2-3	1-2		
2	2.83	2.83	4.15	2.83	4.16	2.83	4.14	2.85	3.00	9	
	2.83	2.84	4.16	2.84	4.17	2.84	4.17	2.86	4.57		
4	2.84	4.18	2.842	2.83	4.18	2.83	4.15	2.85	3.01	7	
	2.84	4.18	2.841	2.84	4.18	2.84	4.18	2.86	4.57		
6	2.84	4.19	2.84	4.18	2.84	2.83	4.13	2.85	2.98	5	
	2.83	4.19	2.84	4.17	2.84	2.84	4.16	2.86	4.55		
8	2.83	4.18	2.84	4.18	2.84	4.17	2.84	2.85	3.00	3	
	2.84	4.18	2.84	4.19	2.84	4.16	2.84	2.86	4.55		
10	2.83	4.16	2.83	4.16	2.84	4.16	2.84	4.17	2.84	nL	
	2.84	4.16	2.83	4.16	2.84	4.16	2.84	4.17	2.84		
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10	
		INTERLAYERDISTANCE OF EVEN LAYERS									

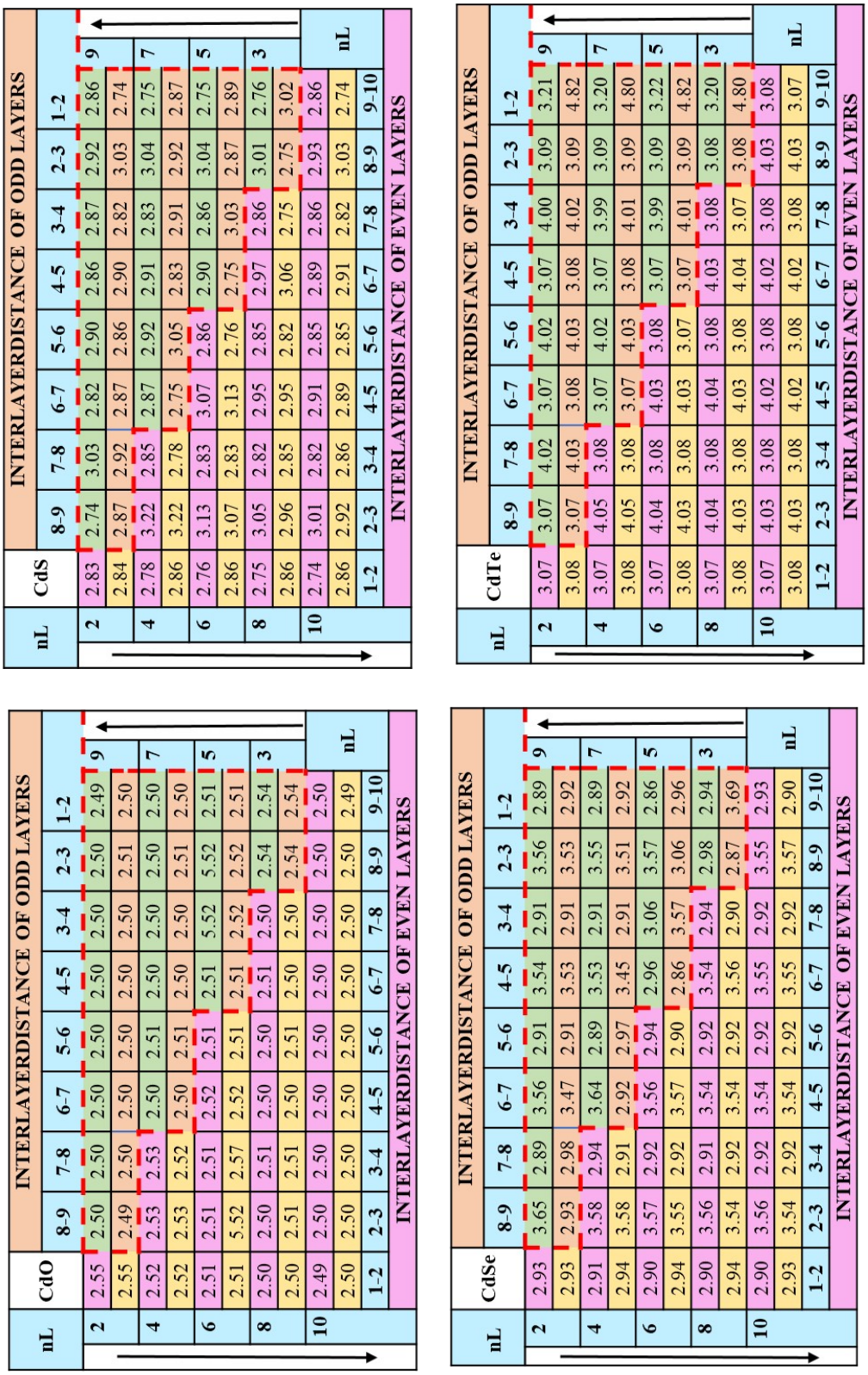


nL	ZnO	INTERLAYERDISTANCE OF ODD LAYERS									
		8-9	7-8	6-7	5-6	4-5	3-4	2-3	1-2		
2	2.44	2.36	2.39	2.38	2.38	2.38	2.38	2.39	2.36	9	
	2.44	2.39	2.39	2.38	2.38	2.38	2.38	2.38	2.39		
4	2.39	2.42	2.42	2.40	2.39	2.39	2.39	2.39	2.37	7	
	2.43	2.42	2.39	2.37	2.39	2.39	2.39	2.39	2.40		
6	2.37	2.40	2.39	2.40	2.40	2.41	2.40	2.41	2.38	5	
	2.40	2.40	2.40	2.40	2.38	2.38	2.41	2.40	2.41		
8	2.36	2.39	2.38	2.38	2.39	2.39	2.39	2.44	2.41	3	
	2.39	2.39	2.38	2.38	2.39	2.39	2.36	2.44	2.44		
10	2.36	2.38	2.38	2.37	2.37	2.37	2.38	2.38	2.39	nL	
	2.39	2.38	2.38	2.37	2.37	2.38	2.38	2.38	2.36		
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10	
		INTERLAYERDISTANCE OF EVEN LAYERS									



nL	ZnSe	INTERLAYERDISTANCE OF ODD LAYERS									
		8-9	7-8	6-7	5-6	4-5	3-4	2-3	1-2		
2	2.70	2.70	3.79	2.70	3.79	2.70	3.75	2.73	2.93	9	
	2.70	2.70	3.81	2.71	3.80	2.70	3.79	2.72	4.02		
4	2.70	3.80	2.71	2.70	3.77	2.70	3.75	2.73	2.92	7	
	2.71	3.80	2.70	2.70	3.79	2.71	3.79	2.72	4.01		
6	2.70	3.78	2.71	3.80	2.70	2.70	3.74	2.73	2.93	5	
	2.71	3.77	2.70	3.80	2.69	2.70	3.78	2.73	4.03		
8	2.70	3.79	2.71	3.78	2.71	3.77	2.70	2.73	2.91	3	
	2.71	3.78	2.71	3.78	2.71	3.79	2.69	2.72	4.03		
10	2.69	3.77	2.71	3.76	2.70	3.76	2.70	3.74	2.71	nL	
	2.70	3.76	2.71	3.76	2.70	3.75	2.71	3.75	2.69		
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10	
		INTERLAYERDISTANCE OF EVEN LAYERS									

Table S4 : Interlayer distances (in Å) of various thick (nL) of 2D-CdX surface slabs are reported. Left and right diagonal panels correspond to even and odd number layered surface sla bs, repectively.



nL	CdS	INTERLAYERDISTANCE OF ODD LAYERS										nL
		8-9	7-8	6-7	5-6	4-5	3-4	2-3	1-2			
2	2.83	2.74	3.03	2.82	2.90	2.86	2.87	2.92	2.86	9		
	2.84	2.87	2.92	2.87	2.86	2.90	2.82	3.03	2.74			
4	2.78	3.22	2.85	2.87	2.92	2.91	2.83	3.04	2.75	7		
	2.86	3.22	2.78	2.75	3.05	2.83	2.91	2.92	2.87			
6	2.76	3.13	2.83	3.07	2.86	2.90	2.86	3.04	2.75	5		
	2.86	3.07	2.83	3.13	2.76	2.75	3.03	2.87	2.89			
8	2.75	3.05	2.82	2.95	2.85	2.97	2.86	3.01	2.76	3		
	2.86	2.96	2.85	2.95	2.82	3.06	2.75	2.75	3.02			
10	2.74	3.01	2.82	2.91	2.85	2.89	2.86	2.93	2.86			
	2.86	2.92	2.86	2.89	2.85	2.91	2.82	3.03	2.74			
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10		
		INTERLAYERDISTANCE OF EVEN LAYERS										

nL	CdTe	INTERLAYERDISTANCE OF ODD LAYERS										nL
		8-9	7-8	6-7	5-6	4-5	3-4	2-3	1-2			
2	3.07	3.07	4.02	3.07	4.02	3.07	4.00	3.09	3.21	9		
	3.08	3.07	4.03	3.08	4.03	3.08	4.02	3.09	4.82			
4	3.07	4.05	3.08	3.07	4.02	3.07	3.99	3.09	3.20	7		
	3.08	4.05	3.08	3.07	4.03	3.08	4.01	3.09	4.80			
6	3.07	4.04	3.08	4.03	3.08	3.07	3.99	3.09	3.22	5		
	3.08	4.03	3.08	4.03	3.07	3.07	4.01	3.09	4.82			
8	3.07	4.04	3.08	4.04	3.08	4.03	3.08	3.08	3.20	3		
	3.08	4.03	3.08	4.03	3.08	4.04	3.07	3.08	4.80			
10	3.07	4.03	3.08	4.02	3.08	4.02	3.08	4.03	3.08			
	3.08	4.03	3.08	4.02	3.08	4.02	3.08	4.03	3.07			
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10		
		INTERLAYERDISTANCE OF EVEN LAYERS										

nL	CdO	INTERLAYERDISTANCE OF ODD LAYERS										nL
		8-9	7-8	6-7	5-6	4-5	3-4	2-3	1-2			
2	2.55	2.50	2.50	2.50	2.50	2.50	2.50	2.50	2.49	9		
	2.55	2.49	2.50	2.50	2.50	2.50	2.50	2.51	2.50			
4	2.52	2.53	2.53	2.50	2.51	2.50	2.50	2.50	2.50	7		
	2.52	2.53	2.52	2.50	2.51	2.50	2.50	2.51	2.50			
6	2.51	2.51	2.51	2.52	2.51	2.51	5.52	5.52	2.51	5		
	2.51	5.52	2.57	2.52	2.51	2.51	2.52	2.52	2.51			
8	2.50	2.50	2.51	2.50	2.50	2.51	2.50	2.54	2.54	3		
	2.50	2.51	2.51	2.50	2.51	2.50	2.50	2.54	2.54			
10	2.49	2.50	2.50	2.50	2.50	2.50	2.50	2.50	2.50			
	2.50	2.50	2.50	2.50	2.50	2.50	2.50	2.50	2.49			
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10		
		INTERLAYERDISTANCE OF EVEN LAYERS										

nL	CdSe	INTERLAYERDISTANCE OF ODD LAYERS										nL
		8-9	7-8	6-7	5-6	4-5	3-4	2-3	1-2			
2	2.93	3.65	2.89	3.56	2.91	3.54	2.91	3.56	2.89	9		
	2.93	2.93	2.98	3.47	2.91	3.53	2.91	3.53	2.92			
4	2.91	3.58	2.94	3.64	2.89	3.53	2.91	3.55	2.89	7		
	2.94	3.58	2.91	2.92	2.97	3.45	2.91	3.51	2.92			
6	2.90	3.57	2.92	3.56	2.94	2.96	3.06	3.57	2.86	5		
	2.94	3.55	2.92	3.57	2.90	2.86	3.57	3.06	2.96			
8	2.90	3.56	2.91	3.54	2.92	3.54	2.94	2.98	2.94	3		
	2.94	3.54	2.92	3.54	2.92	3.56	2.90	2.87	3.69			
10	2.90	3.56	2.92	3.54	2.92	3.55	2.92	3.55	2.93			
	2.93	3.54	2.92	3.54	2.92	3.55	2.92	3.57	2.90			
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10		
		INTERLAYERDISTANCE OF EVEN LAYERS										

Table S5: Interlayer distances (in Å) of MX (expect CdO) various thick surface slabs (nL) are provided. Pink and yellow color marked boxes correspond to M-X bonding side and non-bonding side of surface slabs (refer Fig. 2), respectively.

	nL	INTERLAYERDISTANCE OF WURZITE SLABS								
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10
ZnO	7	2.16	2.13	2.12	2.13	2.14	2.22			
		2.88	3.00	3.02	3.02	3.01	2.94			
	8	2.14	2.09	2.08	2.08	2.09	2.10	2.18		
		2.91	3.04	3.06	3.07	3.07	3.07	3.01		
	9	2.13	2.07	2.07	2.07	2.07	2.07	2.08	2.16	
		2.93	3.08	3.11	3.11	3.11	3.11	3.10	3.04	
	10	2.12	2.06	2.06	2.05	2.05	2.05	2.05	2.07	2.14
		2.94	3.10	3.14	3.14	3.14	3.14	3.13	3.13	3.07
ZnS	6	2.56	2.52	2.50	2.51	2.49				
		2.96	3.60	3.55	3.57	3.64				
	7	2.51	2.46	2.45	2.45	2.45	2.45			
		3.19	3.69	3.67	3.67	3.67	3.74			
	8	2.43	2.40	2.39	2.39	2.39	2.39	2.40		
		3.71	3.80	3.79	3.79	3.79	3.79	3.87		
	9	2.42	2.38	2.38	2.38	2.38	2.38	2.38	2.39	
		3.75	3.82	3.82	3.82	3.82	3.81	3.81	3.89	
	10	2.41	2.37	2.37	2.37	2.37	2.37	2.37	2.38	2.39
		3.77	3.84	3.83	3.82	3.82	3.83	3.83	3.83	3.90
ZnSe	5	2.62	2.64	2.68	2.67					
		3.89	3.77	3.92	2.82					
	6	2.59	2.60	2.60	2.63	2.66				
		3.96	3.85	3.83	3.93	2.84				
	7	2.56	2.56	2.56	2.57	2.59	2.65			
		4.00	3.91	3.92	3.90	3.96	2.88			
	8	2.53	2.53	2.53	2.54	2.54	2.57	2.66		
		4.02	3.94	3.96	3.96	3.93	3.97	2.91		
	9	2.52	2.52	2.51	2.52	2.52	2.53	2.56	2.67	
		4.04	3.97	3.98	3.98	3.98	3.95	3.98	2.91	
	10	2.52	2.51	2.51	2.51	2.51	2.51	2.52	2.56	2.67
		4.05	3.99	4.00	4.00	4.00	4.00	3.97	3.99	2.92
ZnTe	4	2.86	2.92	2.82						
		4.24	4.44	2.88						
	5	2.77	2.78	2.81	2.80					
		4.39	4.24	4.40	2.88					
	6	2.73	2.74	2.74	2.76	2.79				
		4.44	4.31	4.28	4.37	2.90				
	7	2.70	2.71	2.71	2.71	2.73	2.79			
		4.48	4.33	4.33	4.29	4.37	2.90			
	8	2.69	2.70	2.70	2.70	2.70	2.72	2.78		
		4.51	4.35	4.35	4..5	4.30	4.36	2.91		
	9	2.68	2.69	2.71	2.69	2.69	2.69	2.71	2.78	
		4.52	4.37	4.38	4.37	4.37	4.33	4.38	2.91	
	10	2.68	2.68	2.68	2.68	2.68	2.68	2.68	2.70	2.78
		4.53	4.38	4.37	4.37	4.37	4.36	4.31	4.35	2.91

	nL	INTERLAYERDISTANCE OF WURZITE SLABS								
		1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10
CdS	8	2.66	2.71	2.69	2.70	2.70	2.73	2.75		
		3.46	3.42	3.47	3.47	3.41	3.54	3.02		
	9	2.63	3.64	2.64	2.64	2.64	2.64	2.67	2.77	
		3.76	3.70	3.72	3.72	3.72	3.69	3.72	3.10	
	10	2.62	2.62	2.61	2.61	2.61	2.62	2.62	2.65	2.76
		3.85	3.81	3.83	3.83	3.83	3.84	3.80	3.81	3.15
CdSe	7	2.75	2.76	2.76	2.76	2.80	2.86			
		4.06	3.94	3.96	3.90	4.06	3.08			
	8	2.73	2.73	2.73	2.73	2.74	2.78	2.88		
		4.15	4.06	4.08	4.07	4.03	4.43	3.10		
	9	2.72	2.71	2.71	2.71	2.72	2.72	2.77	2.88	
		4.20	4.13	4.14	4.14	4.13	4.08	4.16	3.11	
	10	2.72	2.71	2.70	2.70	2.71	2.71	2.73	2.76	2.89
		4.22	4.16	4.17	4.17	4.17	4.15	4.11	4.16	3.11
CdTe	6	2.94	2.95	2.95	2.98	3.00				
		4.63	4.47	4.43	4.61	3.13				
	7	2.91	2.92	2.92	2.93	2.96	3.01			
		4.69	4.53	4.53	4.48	4.64	3.15			
	8	2.89	2.90	2.90	2.90	2.91	2.95	3.01		
		4.73	4.56	4.57	4.57	4.53	4.68	3.16		
	9	2.89	2.88	2.89	2.89	2.89	2.90	2.95	3.01	
		4.76	4.59	4.59	4.59	4.60	4.54	4.69	3.17	
	10	2.88	2.88	2.88	2.88	2.88	2.88	2.89	2.94	3.01
		4.78	4.61	4.61	4.61	4.60	4.60	4.54	4.68	3.17

Note that in last two layers (particularly for MSe, MTe cases), the M-X interlayer distances in both bonding and non-bonding sides are almost equal due to formation of bilayer in the WZ slabs.

Table S6- Buckling distances (Δz in Å) of various thick 2D – ZnX (X=S, Se, Te) surface slabs.

Layer	1L	2L	3L	4L	5L	6L	7L	8L	9L	10L
1L	0.02									
2L	0.473	-0.475								
3L	0.254	0.006	0.26							
4L	0.402	-0.33	0.331	-0.4						
5L	0.318	-0.171	0.005	0.162	-0.309					
6L	0.371	-0.275	0.213	-0.212	0.279	-0.372				
7L	0.007	-0.005	0.002	0	0.002	0.005	-0.007			
8L	0.335	-0.222	0.134	-0.108	0.109	-0.135	0.228	-0.336		
9L	0.324	-0.203	0.096	-0.045	0.002	-0.035	0.089	-0.194	0.318	
10L	0.318	-0.197	0.099	-0.06	0.044	-0.046	0.064	-0.103	0.202	-0.321
1L	0.409									
2L	0.724	-0.723								
3L	0.271	0.477	-0.584							
4L	0.688	-0.656	0.653	-0.658						
5L	0.022	-0.018	0	0.018	-0.022					
6L	0.684	-0.65	0.632	-0.632	0.649	-0.682				
7L	0.658	-0.629	0.595	-0.595	0.51	-0.426	-0.297			
8L	0.679	-0.642	0.626	-0.622	0.622	-0.623	0.642	-0.677		
9L	0.658	-0.63	0.603	-0.609	0.597	-0.599	0.511	-0.426	-0.296	
10L	0.673	-0.637	0.619	-0.619	0.618	-0.619	0.621	-0.623	0.641	-0.675
1L	0.565									
2L	0.985	-0.984								
3L	0.66	0.942	-0.948							
4L	0.98	-0.974	0.974	-0.98						
5L	0.662	0.94	-0.938	0.966	-0.965					
6L	0.981	-0.974	0.971	-0.972	0.973	-0.981				
7L	0.663	0.939	-0.936	0.962	-0.954	0.969	-0.97			
8L	0.98	-0.973	0.969	-0.97	0.97	-0.969	0.973	-0.98		
9L	0.668	0.942	-0.939	0.962	-0.955	0.964	-0.957	0.969	-0.968	
10L	0.978	-0.97	0.97	-0.969	0.969	-0.968	0.969	-0.969	0.969	-0.978

ZnS

ZnSe

ZnTe

This buckling distances is higher value of MTe case, owing to having high covalency.

Table S7- Buckling distances (in Å) of various thick 2D– CdX (X=S, Se, Te) surface slabs.

Layer	1L	2L	3L	4L	5L	6L	7L	8L	9L	10L
1L	0.04									
2L	0.5	-0.5								
3L	0.429	0.348	-0.272							
4L	0.477	-0.444	0.446	-0.476						
5L	0.458	-0.392	0.074	0.364	-0.441					
6L	0.472	-0.44	0.421	-0.42	0.441	-0.473				
7L	0.461	-0.436	0.382	-0.343	0.082	0.401	-0.458			
8L	0.476	-0.443	0.427	-0.419	0.418	-0.423	0.439	-0.469		
9L	0.448	-0.392	0.029	0.359	-0.391	0.42	-0.42	0.444	-0.469	
10L	0.469	-0.438	0.422	-0.416	0.416	-0.417	0.416	-0.421	0.438	-0.469
1L	0.293									
2L	0.728	-0.727								
3L	-0.445	0.669	-0.682							
4L	0.711	-0.701	0.695	-0.712						
5L	-0.44	0.663	-0.663	0.705	-0.707					
6L	0.712	-0.702	0.695	-0.699	0.707	-0.717				
7L	0.438	0.655	-0.658	0.695	-0.685	0.703	-0.703			
8L	0.711	-0.702	0.695	-0.693	0.696	-0.694	0.706	-0.713		
9L	-0.437	0.657	-0.66	0.695	-0.688	0.698	-0.696	0.705	0.704	
10L	0.719	-0.706	0.693	-0.694	0.698	-0.699	0.696	-0.692	0.7	-0.713
1L	0.46									
2L	0.95	-0.95								
3L	0.642	0.905	-0.898							
4L	0.945	-0.943	0.943	-0.944						
5L	0.689	0.887	-0.878	0.908	-0.903					
6L	0.945	-0.945	0.945	-0.944	0.94	-0.941				
7L	0.649	0.918	-0.911	0.941	-0.935	0.941	-0.942			
8L	0.947	-0.944	0.942	-0.943	0.945	-0.944	0.941	-0.943		
9L	0.65	0.922	-0.913	0.936	-0.943	0.943	-0.935	0.943	-0.939	
10L	0.946	-0.944	0.945	-0.945	0.947	-0.945	0.945	-0.946	0.947	0.948

CdS

CdSe

CdTe

This buckling distances is higher value of MTe case, owing to having high covalency,

We calculated the exfoliation energy (E_{exf}) to form n number of monolayers from respective surface slabs, with their thicknesses varying from 2L to 10L, from following equation.

$$E_{exf} = \frac{n * E_{1L} - E_{nL}}{n}$$

where E_{nL} is total energy of surface slab with thickness of nL, E_{1L} is the energy of the monolayer and n is the number of layer. Deduced E_{exf} values for 2D-MO and 2D-MX surface slabs are given in Fig. S5. It shows that thinner slabs are easier to exfoliate, as compared to thicker slabs.

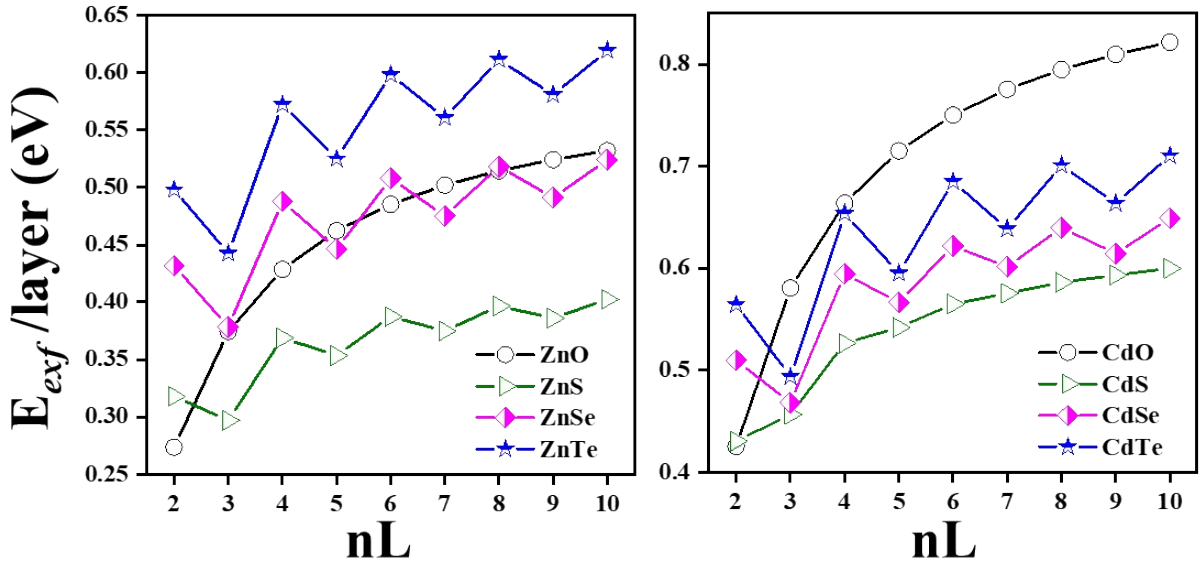


Fig. S5: Calculated E_{exf} values for various thick (2L to 10L) 2D-MO and 2D-MX surface slabs are shown.

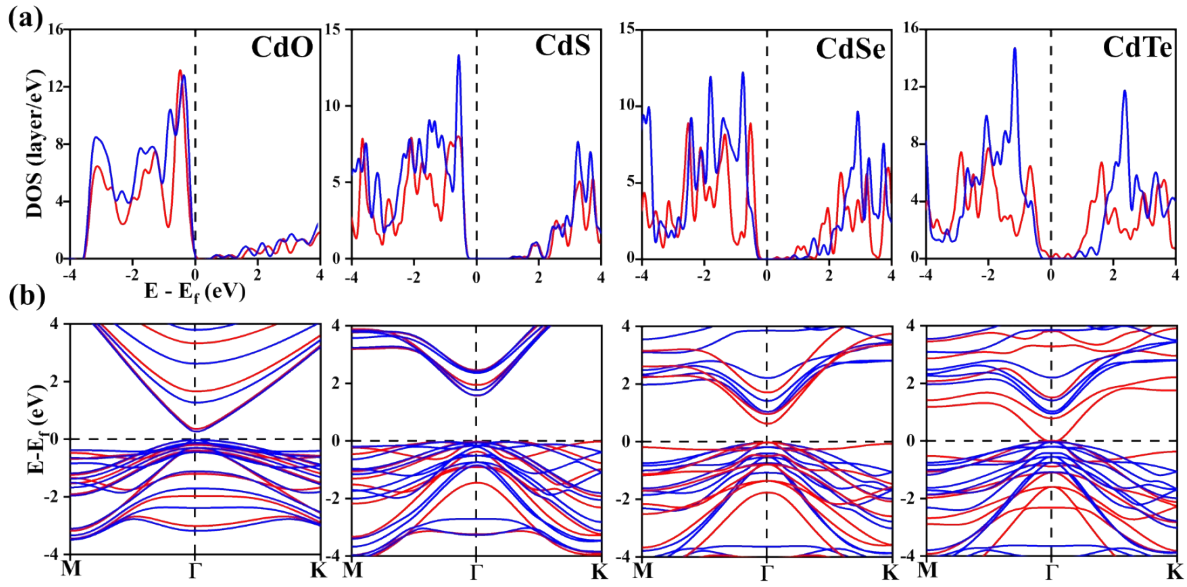


Fig. S6: DOS (a) and BS (b) of 3L (red line) and 4L (blue line) layered 2D- CdX surface slabs.

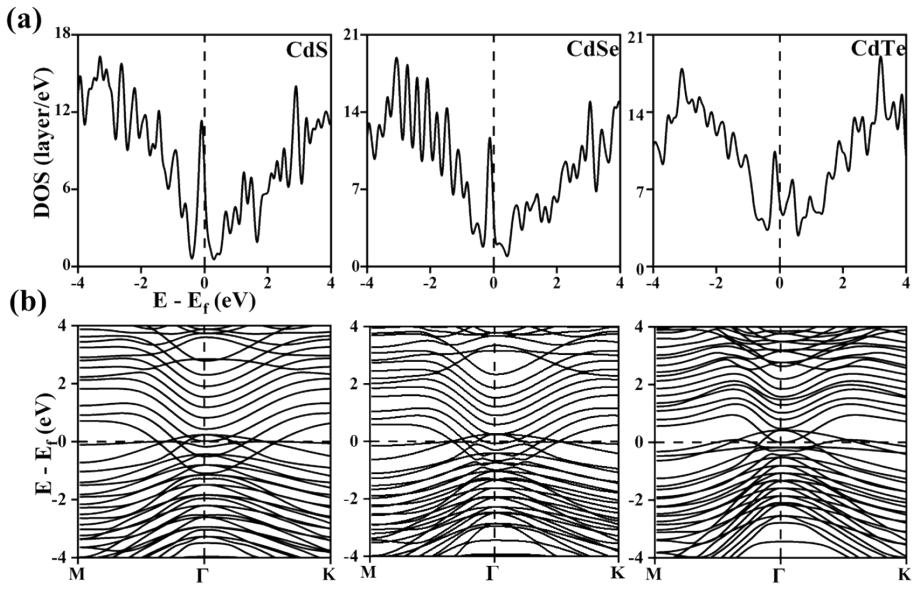


Fig. S7: DOS (a) and BS (b) of 10L thick WZ surface slab of MX compounds.

Table S7 : Elastic constants (C_{11}, C_{12}, C_{22}, C_{66} in unit of N/m), layer modulus (γ, N/m), Young's modules (Y, N/m), Poisson's ratio (υ) of 2L 2D-MX compounds are given.

MX	Elastic Constants (N/m)			Layer modulus (γ)	Young modulus (N/m)	Poisson's ratio (υ)
	C_{11}	C_{12}	C_{66}			
ZnO	124.4	79.09	22.66	90.43	74.139	0.636
ZnS	72.98	28.36	22.31	39.51	61.967	0.389
ZnSe	61.45	21.61	19.92	31.57	53.85	0.352
ZnTe	54.08	16.74	18.67	26.07	48.895	0.31
CdO	95.40	76.74	9.33	81.40	33.673	0.804
CdS	53.30	27.98	12.66	34.31	38.608	0.525
CdSe	42.93	19.38	11.77	25.27	34.178	0.452
CdTe	39.41	15.85	11.78	21.74	33.027	0.402

These following formulae are used to calculate mechanical properties mentioned in table as follows:

$$\gamma = \frac{1}{4}(c_{11} + c_{12} + 2c_{12})$$

$$Y = \frac{(C_{11}C_{22} - C_{12}C_{21})}{C_{22}}$$

$$\upsilon = \frac{c_{21}}{C_{22}}$$

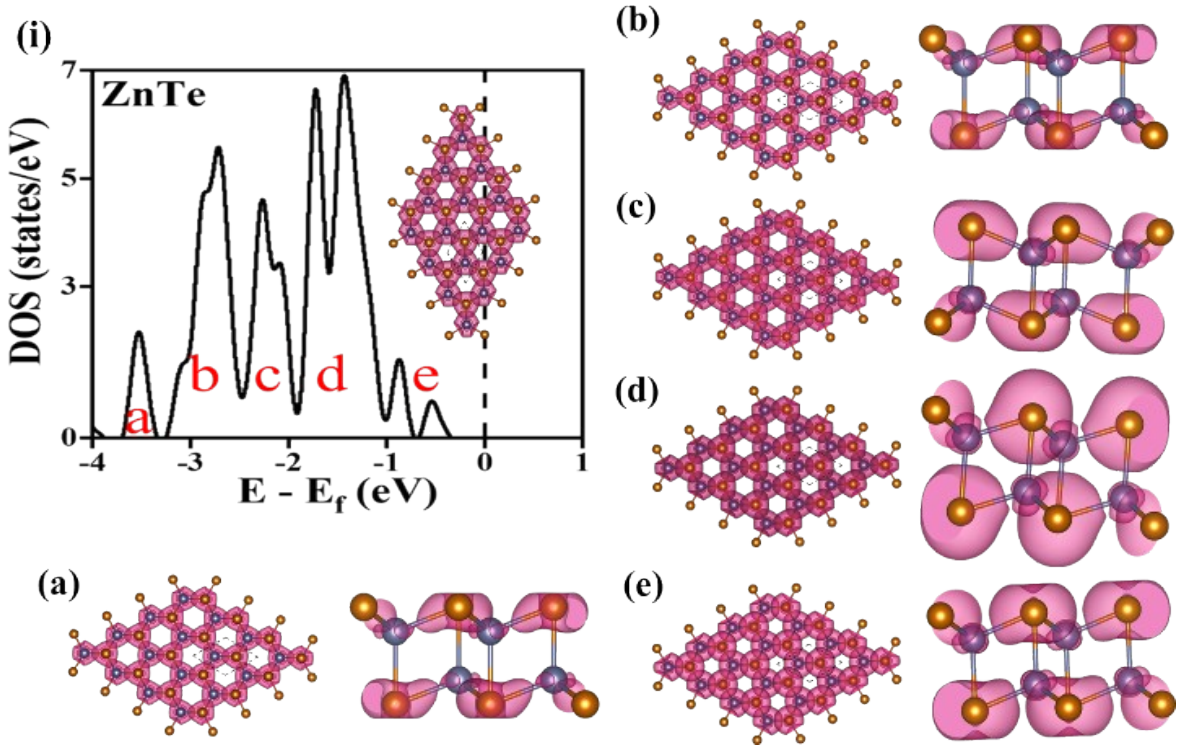


Fig. S8: Occupied DOS of 2L ZnTe is shown, where different discrete peaks (a to e) are marked. The partial change density isosurface plots are plotted for those peaks and their side and top views are presented.

Despite of vdW driven interlayer interaction in 2D-MX surface slabs, the bonding nature within layer is explored by performing partial charge density diagrams for 2L ZnTe cases and they are shown in Fig. S5. Structurally, sp^3 hybridized cations (M) are located in the centre of tetrahedra that is formed by X atoms. Each X atom interacts with three M atoms and holding one lone pair, forming trigonal pyramid shape with bond angle of 107°. It is also seen in the charge density diagrams, donut like shapes (mixing of p_x and p_y orbitals) that are presented in the both sides of bonding states (sp^3 orbitals).

Table S9: Band gap (in eV) of various thicknesses (nL) of 2D-MX surface slabs are shown, along with WZ bulk.

nL	ZnO	ZnS	ZnSe	ZnTe	CdO	CdS	CdSe	CdTe
1	1.66	2.61	1.91	1.694	0.83	1.68	1.33	1.31
2	1.41	2.66	1.85	1.01	0.55	1.87	1.61	1.11
3	1.28	1.54	0.62	0.06	0.39	1.60	0.66	0.00
4	1.20	2.34*	1.67	0.91	0.30	1.61*	1.28*	0.99
5	1.15	1.98	0.55	0.047	0.24	1.44*	0.85*	0.00
6	1.11	2.12*	1.55*	0.885	0.20	1.52*	1.09*	0.96
7	1.08	1.86	0.52	0.073	0.17	1.47	0.51	0.00
8	1.07	2.00*	1.46*	0.883	0.14	1.47*	0.98*	0.90
9	1.05	1.91	0.54	0.07	0.12	1.43*	0.47*	0.00
10	1.04	1.96*	1.40*	0.87	0.10	1.42*	0.90	0.88
∞L	0.94	1.81	1.29	0.93	0.00	1.25	0.74	1.06
WZ Bulk	0.76	2.28	1.40	1.36	0.00	1.23	0.67	0.78

The band gap is calculated by deducing BS calculations. We observed direct band gap in all the cases. It is also noticed that indirect band gap value, in a few cases (* marked), is also lied closer to direct band gap.

Table S10 (a): Effective mass (e^*) of various thicknesses of 2D-ZnX various surface slabs

nL	electron (e^*_e)				hole (e^*_h)			
	ZnO	ZnS	ZnSe	ZnTe	ZnO	ZnS	ZnSe	ZnTe
1	0.28	0.19	0.14	0.12	1.07	0.72	0.71	0.97
2	0.24	0.21	0.15	0.12	1.19	1.35	1.03	1.38
3	0.22	0.2	0.15	0.11	1.26	0.68	0.74	1
4	0.21	0.2	0.15	0.12	1.3	1.03	1.12	1.12
5	0.2	0.19	0.15	0.12	1.33	0.8	0.82	0.66
6	0.2	0.19	0.15	0.13	1.36	1.26	0.4	1.01
7	0.2	0.19	0.15	0.12	1.37	0.82	0.82	0.99
8	0.19	0.19	0.15	0.13	1.39	1.72	3.31	1.1
9	0.19	0.19	0.15	0.12	1.43	2.92	0.82	0.98
10	0.19	0.19	0.15	0.13	1.44	2.56	3.07	1.1

Table S10 (b): Effective mass (e^*) of various thicknesses of 2D-CdX various surface slabs.

nL	electron (e^*_e)				hole (e^*_h)			
	CdO	CdS	CdSe	CdTe	CdO	CdS	CdSe	CdTe
1	0.2	0.16	0.14	0.13	0.53	0.65	0.69	0.67
2	0.17	0.12	0.19	0.13	0.76	1.13	0.89	0.93
3	0.16	0.16	0.14	0.13	1.31	0.86	0.89	0.99
4	0.16	0.18	0.16	0.14	1.41	0.89	2.66	1.22
5	0.16	0.16	0.13	0.14	1.87	0.91	0.75	1
6	0.17	0.17	0.15	0.16	1.4	1.49	0.28	1.26
7	0.18	0.17	0.15	0.13	1.4	1.06	0.89	1.01
8	0.19	0.2	0.14	0.13	1.92	1.21	0.8	1.69
9	0.21	0.18	0.14	0.13	1.72	1.28	0.9	1
10	0.22	0.19	0.13	0.13	1.64	1.41	1.58	1.54