Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

InSe: a two-dimensional material with strong interlayer coupling

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Table S1 The calculated lattice parameters with optB86b-vdW and optB88-vdW functionals for bulk β-InSe and γ-InSe. The selected functional is emphasized with purple background.

	Bulk β-InSe			Bulk γ-InSe		
Functional	B86b-vdW	B88-vdW	Exp.[1]	B86b-vdW	B88-vdW	Exp.[2,3]
a	4.054	4.092	4.05	4.062	4.099	4.002
b	4.054	4.092	4.05	4.062	4.099	4.002
c	16.929	16.998	16.93	25.151	25.189	24.961

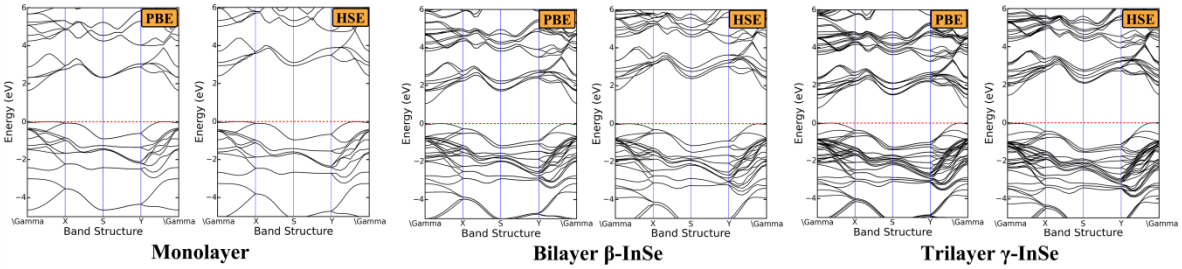

Fig. S1 The calculated PBE and HSE06 band structures for monolayer, bilayer β-InSe, and trilayer γ-InSe, respectively.

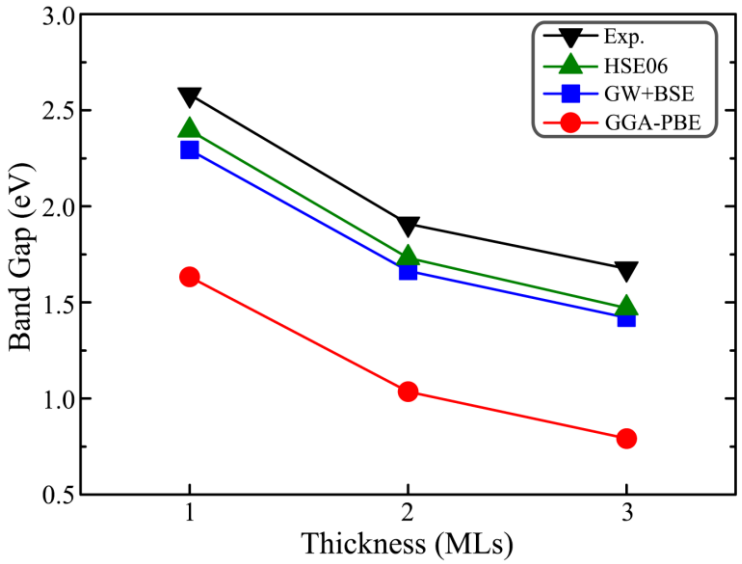

Fig. S2 Evolution of electronic band gaps with varying thickness from monolayer to trilayer γ-InSe as obtained from experiment and different theoretical approaches.

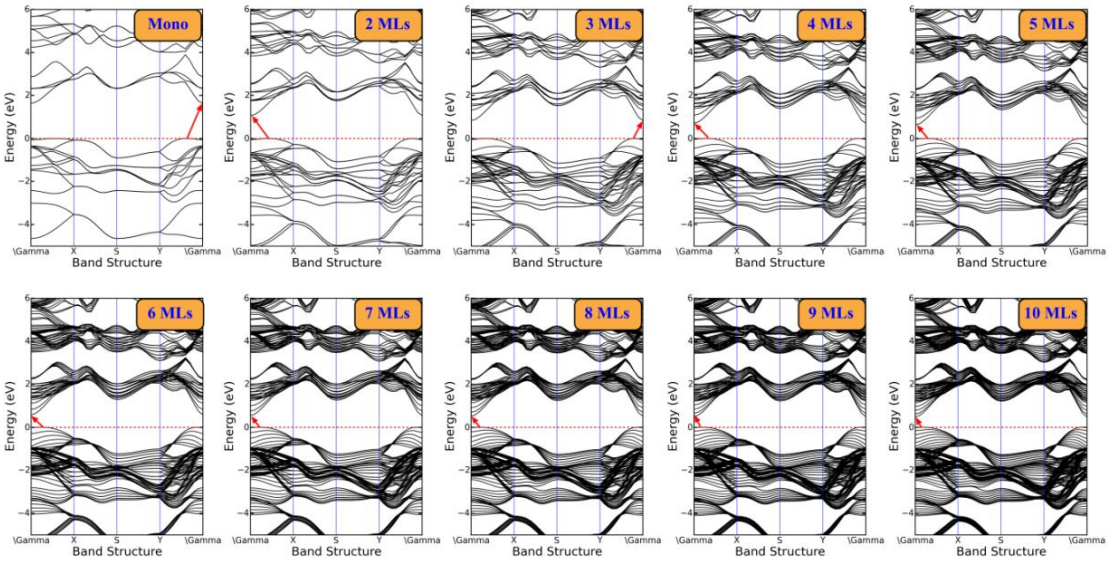

Fig. S3 The calculated PBE band structures of β-InSe from *n* = 1 to *n* = 10-MLs. The arrows point the indirect gap positions.

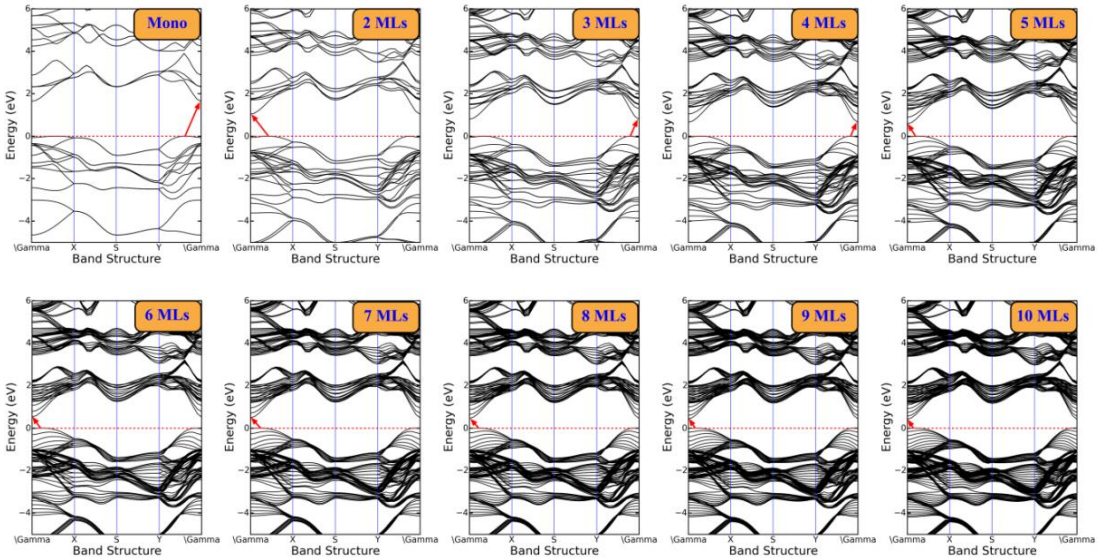

Fig. S4 The calculated PBE band structures of γ-InSe from *n* = 1 to *n* = 10-MLs. The arrows point the indirect gap positions.

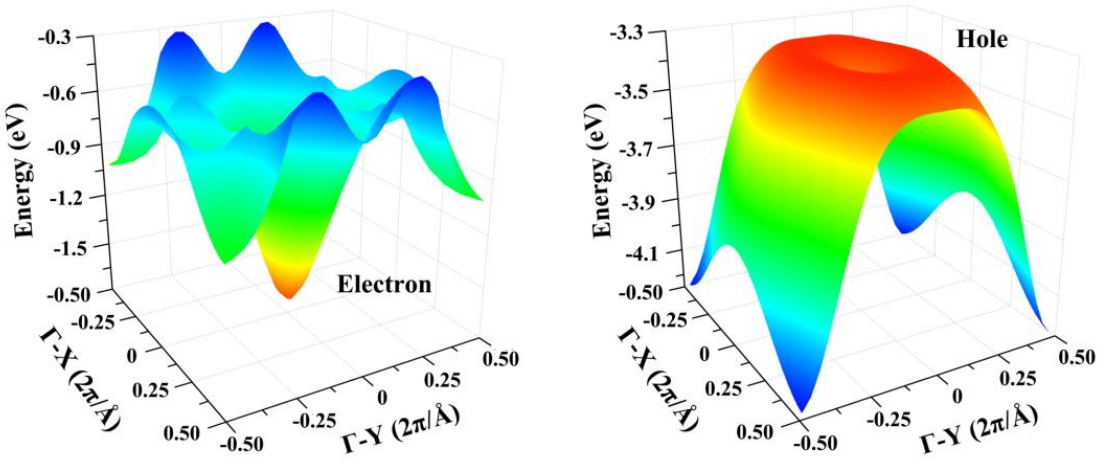

Fig. S5 Contours of the lowest CB and the highest VB of monolayer InSe at the 2D plane of Brillouin zone.

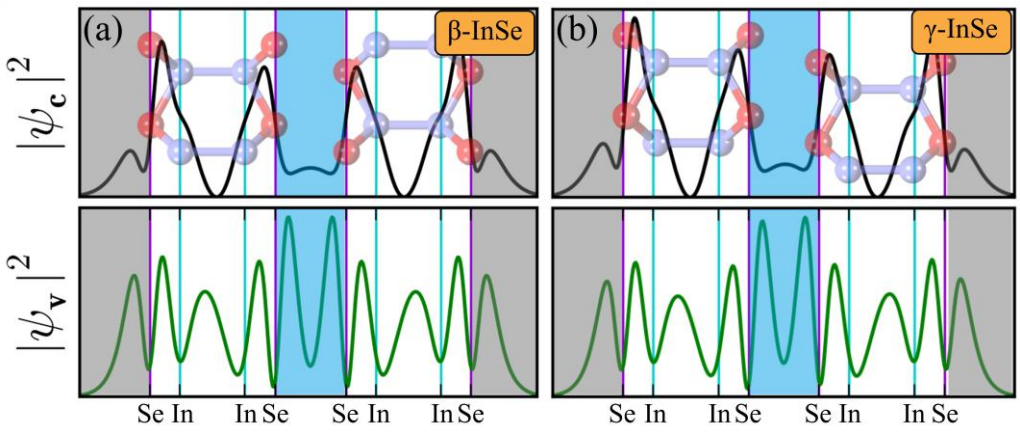

Fig. S6 Planar-averaged squared magnitude of wave functions of the CBM and VBM states of bilayer (a) β-InSe and (b) γ-InSe, respectively. The dodgerblue and grey parts represent interlayer and vacuum regions, respectively.

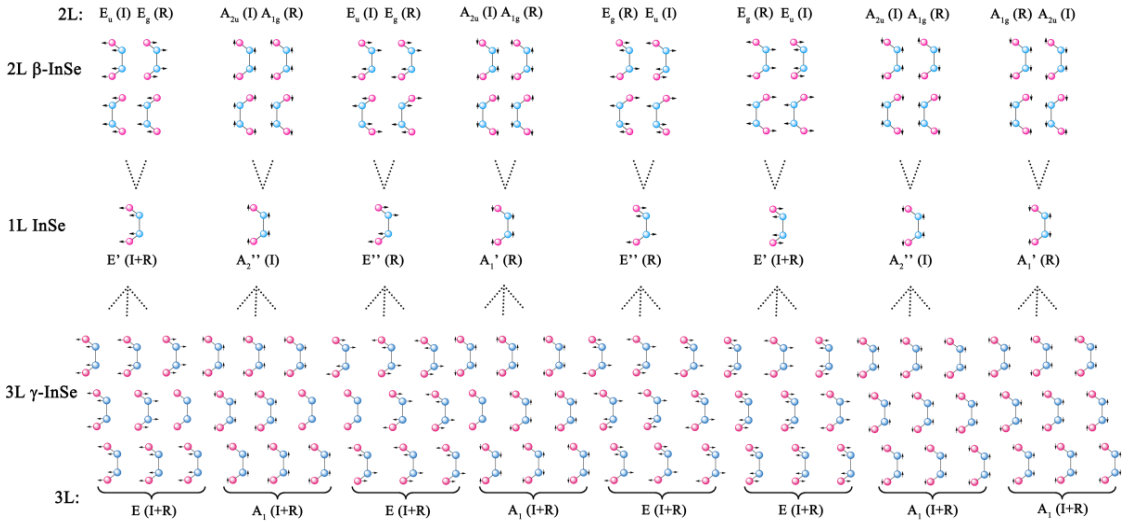

Fig. S7 Vibration modes, symmetry representation, and optical activities (Raman: R; Infrared: I) of the phonon modes for monolayer, bilayer β-InSe, and trilayer γ-InSe, respectively.

Table S2 The effective mass, deformational potential, 2D elastic modulus and the carrier mobility along x and y direction for β-InSe from *n* = 1 to *n* = to 10-MLs.

β-InSe	**Carrier type**	**m_x (m_0) Γ-X**	**m_y (m_0) Γ-Y**	**E_{1x} (eV)**	**E_{1y} (eV)**	**C_{x_2D} (N/m)**	**C_{y_2D} (N/m)**	**μ_{x_2D} ($cm^2V^{-1}s^{-1}$)**	**μ_{y_2D} ($cm^2V^{-1}s^{-1}$)**
1 layer	Electron	0.20	0.23	5.70	5.76	52.35	52.89	801.09	689.20
	Hole	12.28	2.72	2.66	2.28	52.35	52.89	2.22	13.80
2 layers	Electron	0.17	0.20	5.37	2.78	100.57	100.18	2372.77	7496.29
3 layers	Electron	0.16	0.19	5.22	5.36	153.76	153.72	4313.90	3444.57
4 layers	Electron	0.15	0.18	5.67	5.95	201.33	204.14	5418.69	4157.81
5 layers	Electron	0.15	0.18	5.34	5.43	254.96	255.43	7736.44	6246.58
6 layers	Electron	0.15	0.17	5.45	5.60	308.76	308.43	9255.32	7726.56
7 layers	Electron	0.14	0.17	5.28	5.53	356.39	357.65	12623.09	9510.32
8 layers	Electron	0.14	0.17	5.04	4.98	410.67	411.65	15963.94	13497.60
9 layers	Electron	0.14	0.17	4.64	5.13	458.97	463.15	21050.21	14311.14
10 layers	Electron	0.14	0.17	4.32	4.64	511.17	514.72	27046.19	19441.17

Table S3 The effective mass, deformational potential, 2D elastic modulus and the carrier mobility along x and y direction for γ-InSe from *n* = 1 to *n* = to 10-MLs.

γ-InSe	**Carrier type**	**m_x (m_0) Γ-X**	**m_y (m_0) Γ-Y**	**E_{1x} (eV)**	**E_{1y} (eV)**	**C_{x_2D} (N/m)**	**C_{y_2D} (N/m)**	**μ_{x_2D} ($cm^2V^{-1}s^{-1}$)**	**μ_{y_2D} ($cm^2V^{-1}s^{-1}$)**
1 layer	Electron	0.20	0.23	5.70	5.76	52.35	52.89	801.09	689.20
	Hole	12.28	2.72	2.66	2.28	52.35	52.89	2.22	13.80
2 layers	Electron	0.17	0.19	5.34	5.37	100.56	100.08	2461.60	2167.55
3 layers	Electron	0.16	0.18	3.93	4.94	151.29	155.50	7693.67	4448.69
4 layers	Electron	0.15	0.18	5.10	5.15	205.81	202.75	6846.64	5512.10
5 layers	Electron	0.15	0.17	5.30	5.44	255.64	255.36	8102.90	6778.92
6 layers	Electron	0.14	0.17	5.44	5.48	311.08	309.89	10379.64	8391.39
7 layers	Electron	0.14	0.17	5.59	5.77	360.24	359.21	11383.52	8773.73
8 layers	Electron	0.14	0.17	5.50	5.19	415.22	413.09	13553.80	12470.88
9 layers	Electron	0.14	0.16	5.93	5.83	467.41	466.37	13528.86	12220.08
10 layers	Electron	0.14	0.16	5.54	5.79	518.14	516.62	17183.05	13724.44

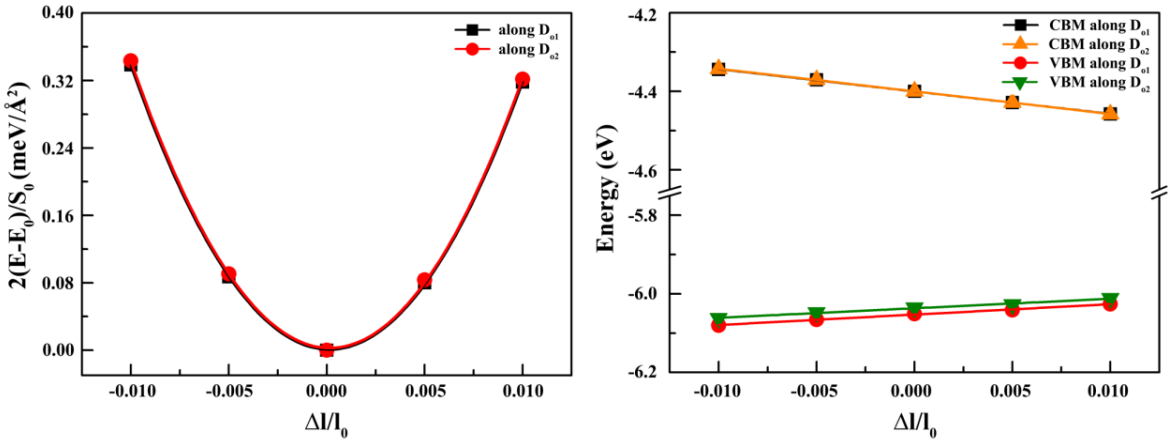

Fig. S8 The 2D elastic modulus is obtained by fitting the strain energy density curves $2(E-E_0)/S_0$ versus $\Delta l/l_0$ for shared monolayer InSe. The strain is applied along the D_{o1} and D_{o2} directions.

References

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