Supporting Information for

The electric field modulated data storage in bilayer InSe

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Table S1 Lattice parameters of bulk and monolayer InSe via different calculation methods

Method	GGA-PBE	optB88-vdW	DFT-D3	Ref. calculation	Ref. experiment
a (Å) (bulk)	4.098	4.089	4.040	3.95[1]	4.005[2]
c (Å) (bulk)	18.783	17.061	16.904	16.92[1]	16.640[2]
a (Å) (ML)	4.095	4.077	4.038	4.041[3]	-

Table S2 Calculated parameters of bilayer InSe with different stacking configurations via GGA-PBE

Stacking	I	II	III	IV	V
a (Å)	4.097	4.095	4.096	4.094	4.097
d (Å)	3.847	4.518	3.897	4.415	3.790
ΔE(meV)	0.00	4.14	1.30	4.55	0.81
E_b (meV)	-10.87	-6.74	-9.57	-6.32	-10.07
E_g (eV)	1.10	1.21	1.11	1.19	1.09

Table S3 Calculated parameters of bilayer InSe with different stacking configurations via optB88-vdW

Stacking	I	II	III	IV	V
a (Å)	4.085	4.077	4.090	4.077	4.087
d (Å)	3.088	3.752	2.966	3.754	3.019
ΔE(meV)	10.45	102.14	0.00	102.51	4.81
E_b (meV)	-272.47	-180.78	-282.92	-180.41	-278.12
E_g (eV)	0.93	1.05	0.86	1.05	0.90

We have calculated the basic properties of trilayer InSe, the results are shown in Table S4. We found that trilayer InSe presents very similar trend to bilayer InSe with different stacking configurations. The trilayer configurations can be divided into two groups according to the interlayer distances as well. Among them, with the stacking configurations energy stabilities of trilayer InSe are predicted to be III>V>I>II>IV. It is notable that, compared with bilayer InSe, for each stacking configuration, the lattice constant *a* and interlayer distance *d* are steady. And the band gaps of trilayer InSe are smaller than those of bilayer InSe.

Table S4 Calculated parameters of trilayer InSe with different stacking configurations via DFT-D3

Stacking	I	II	III	IV	V
a (Å)	4.042	4.034	4.049	4.034	4.045
d (Å)	3.009	3.698	2.879	3.704	2.937
ΔE(meV)	20.62	206.62	0.00	206.68	8.87
Group	Group-S	Group-L	Group-S	Group-L	Group-S
E_b (meV)	-259.18	-166.18	-269.49	-166.15	-265.06
E_g (eV)	0.84	1.04	0.73	1.03	0.79

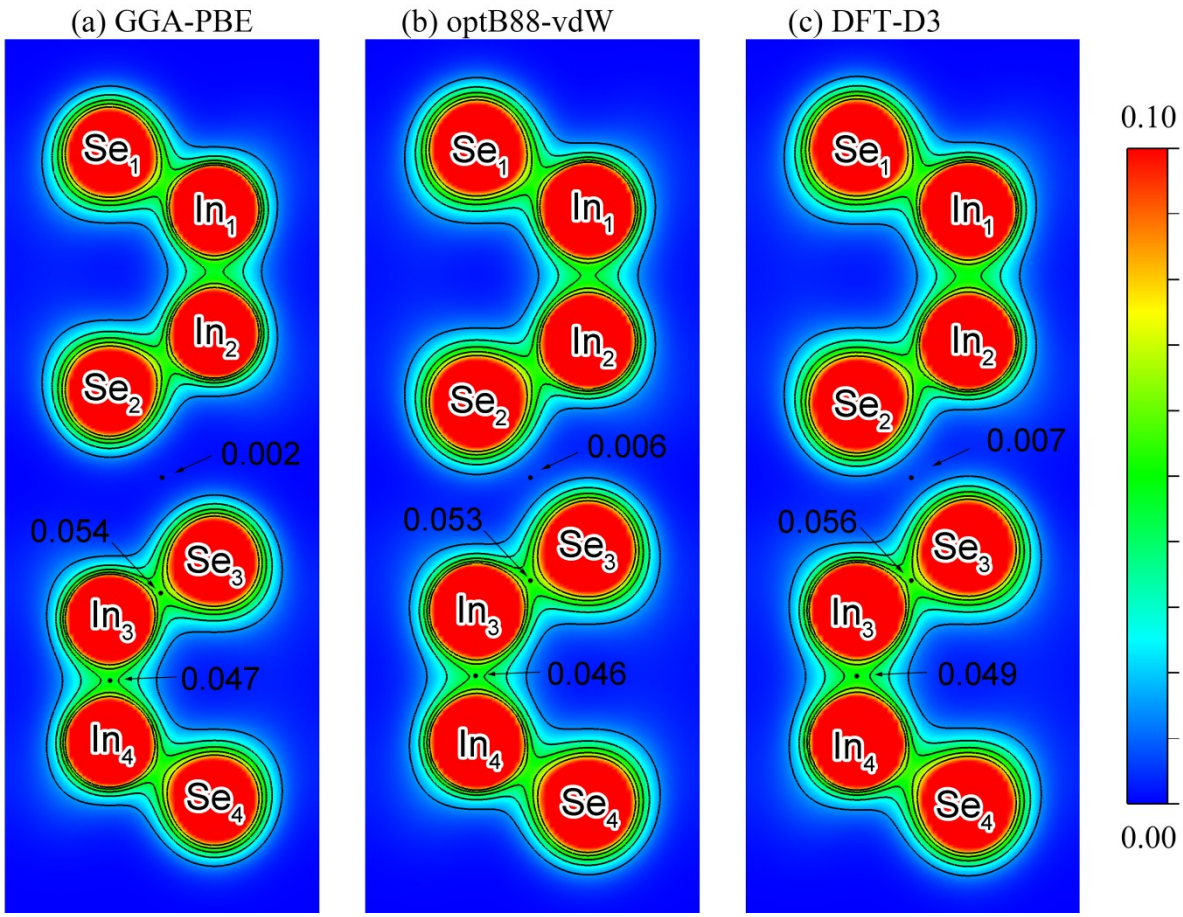

Figure S1 The charge density plots of bilayer InSe with stacking I using (a) GGA-PBE, (b) optB88-vdW and DFT-D3 method in the (10) plane. Contour lines are plotted at 0.015 e/au^3 intervals.

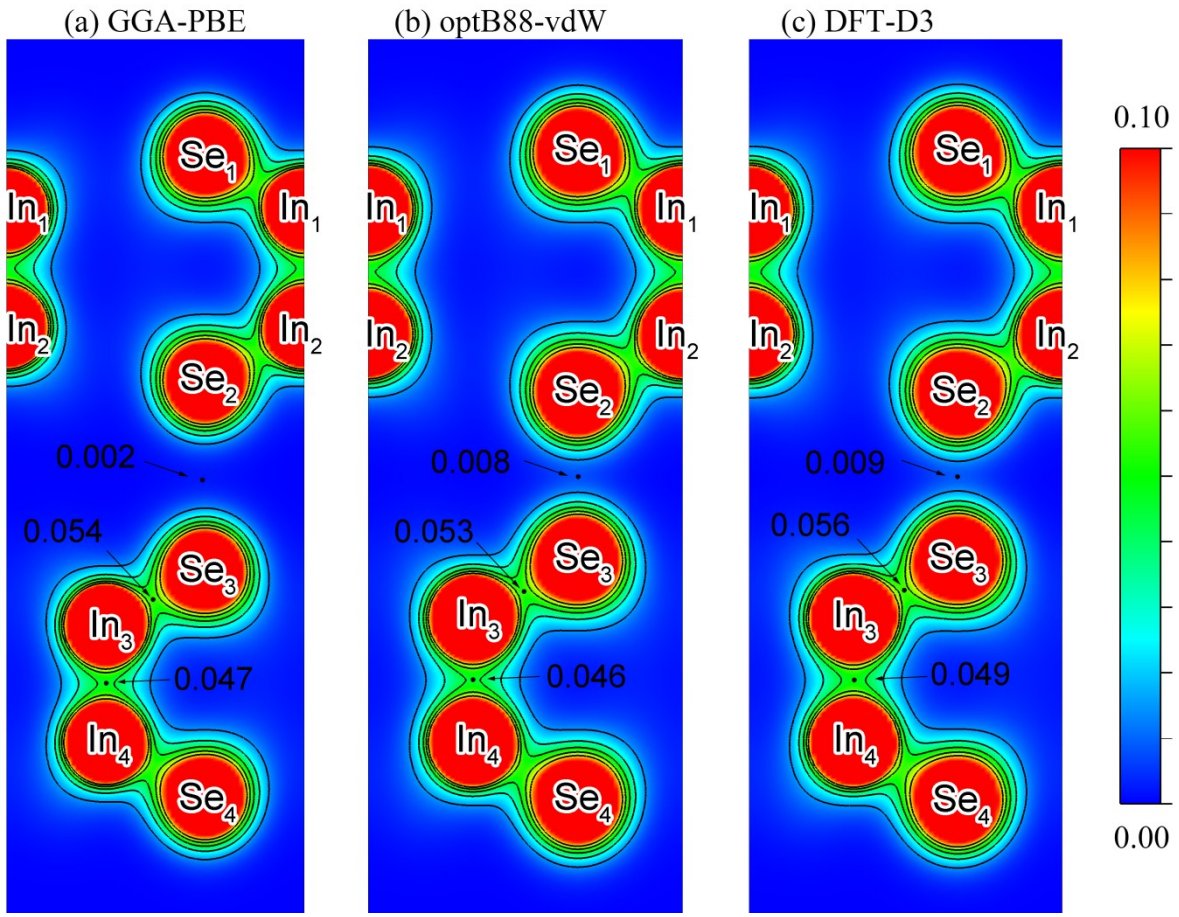

Figure S2 The charge density plots of bilayer InSe with stacking II using (a) GGA-PBE, (b) optB88-vdW and DFT-D3 method in the (10) plane. Contour lines are plotted at 0.015 e/au^3 intervals.

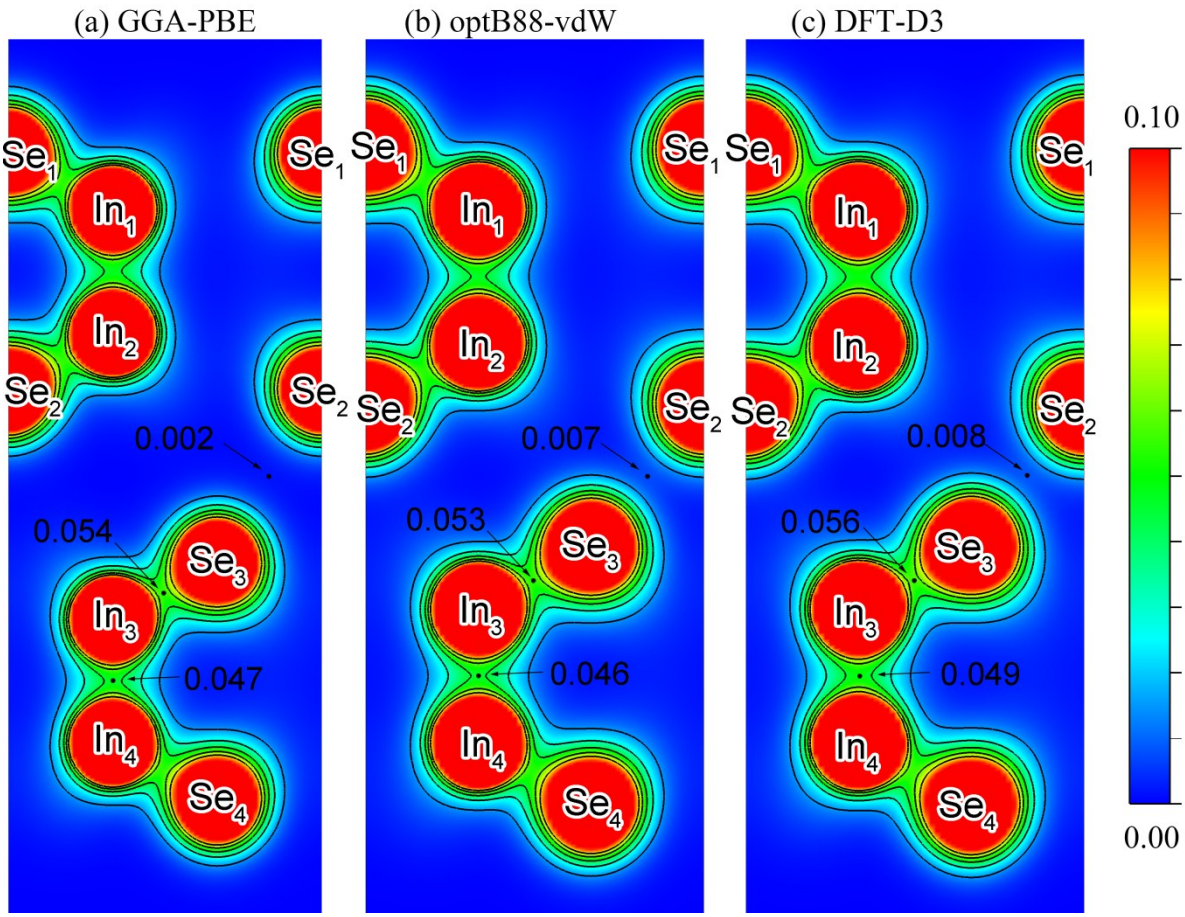

Figure S3 The charge density plots of bilayer InSe with stacking III using (a) GGA-PBE, (b) optB88-vdW and DFT-D3 method in the (10) plane. Contour lines are plotted at 0.015 e/au^3 intervals.

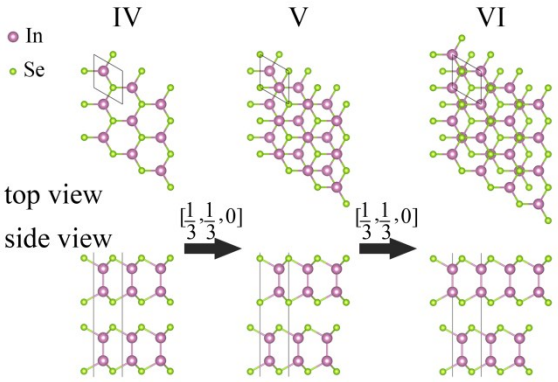

Figure S4 Top and side views of bilayer InSe with stacking IV, V and VI.

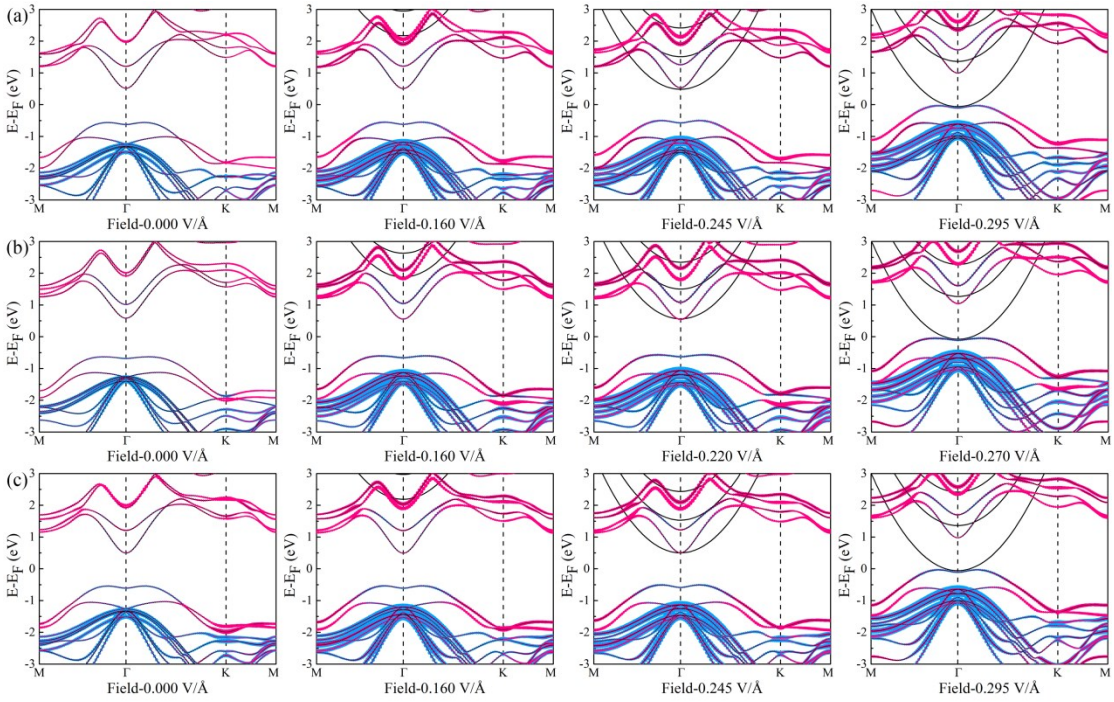

Figure S5 Band structure of bilayer InSe with stacking (a) I (group-S), (b) IV (group-L) and (c) V (group-S) under the different external vertical electric field. The size of the pink, azure circles illustrates the projected weight of the electrons from In and Se atoms, respectively.

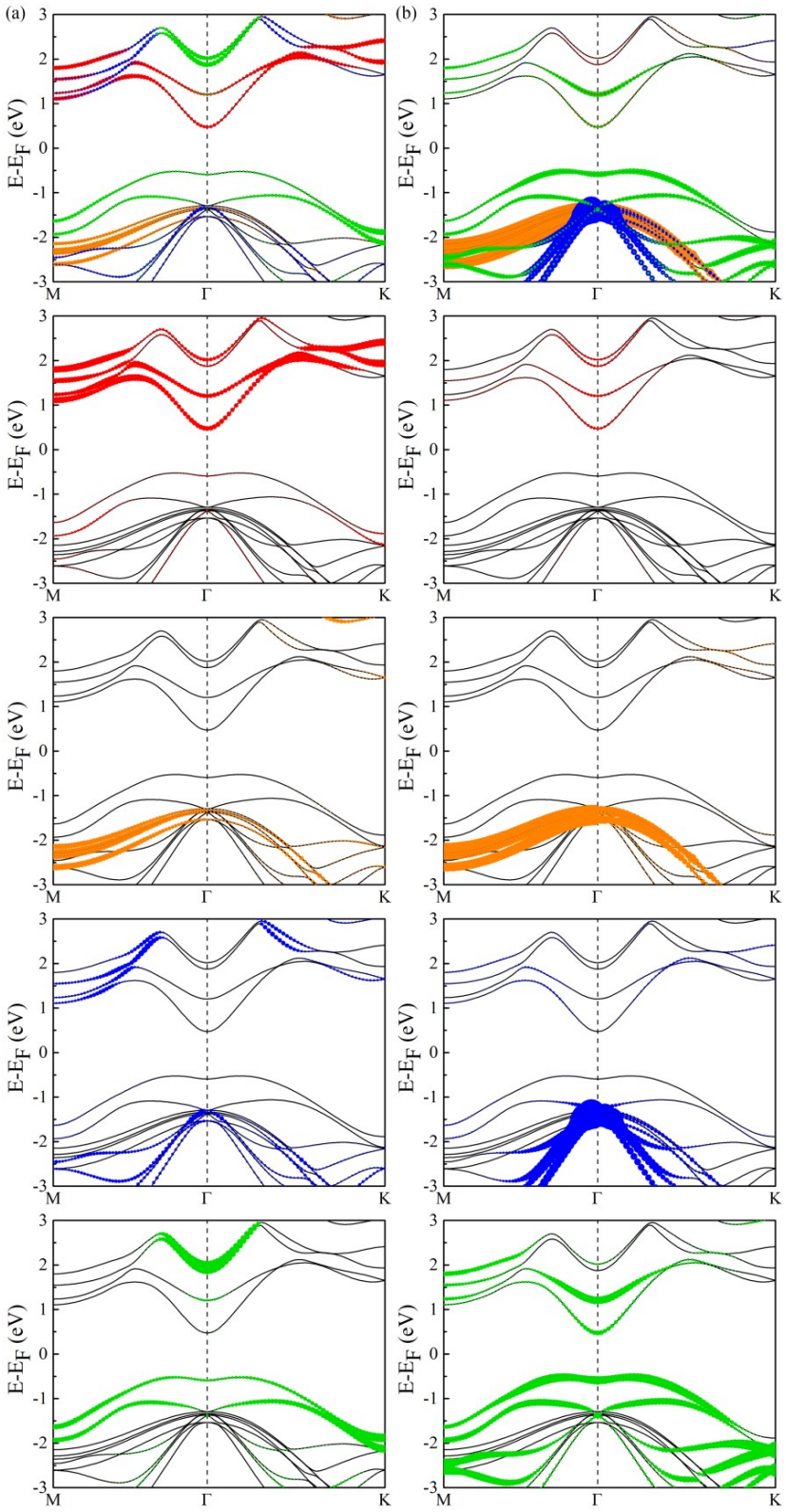

Figure S6 (a) The In electron orbital projected band structure of InSe with stacking III, where the size of the red, orange, blue and green circles illustrates the projected weight of the s, p_x, p_y and p_z orbitals from In atoms, respectively. (b) The Se electron orbital projected band structure of InSe with stacking III, where the size of the red, orange, blue and green circles illustrates the projected weight of the s, p_x, p_y and p_z orbitals from Se atoms, respectively.

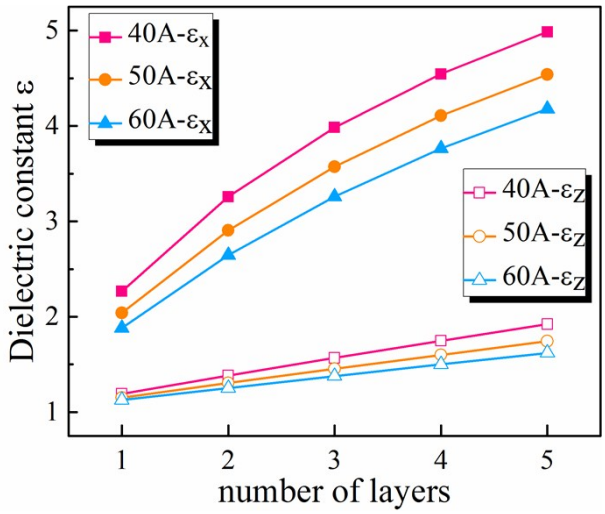

Figure S7 Dielectric constants of few layered InSe as a function of number of layers with different vacuum layer thickness.

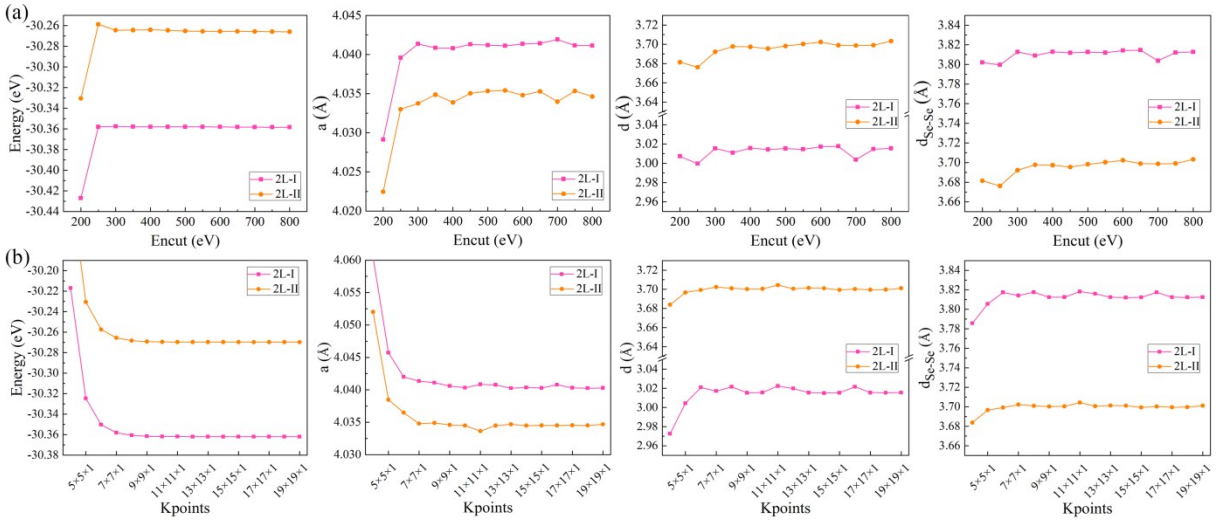

Figure S8 The convergence tests for (a) cutoff energy and (b) K-point grids of bilayer InSe with stacking configuration I (Group-S) and II (Group-L) by DFT-D3.

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