

Supporting Information:

Accelerated Discovery of Two-Dimensional Optoelectronic Octahedral Oxyhalides via High-Throughput *Ab Initio* Calculations and Machine Learning

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1. The computational methods

All geometric optimization of crystals was performed within the density functional theory as implemented within the Vienna ab initio simulation package (VASP)^{1,2} with projected augmented wave (PAW) method^{3,4} and the exchange-correlation interactions were treated within spin-polarized gradient approximation and Perdew-Burke-Ernzerhof generalized gradient approximation (PBE-GGA).⁵ The plane-wave cutoff energy is taken as 500 eV and $12 \times 9 \times 1$ k-points sampling are used for the ground state calculation. The lattice geometries and atomic positions were fully relaxed until the energy and force were converged to 10^{-6} eV and 0.01 eV/Å.

For high-throughput calculations, phonon dispersions were calculated by density functional perturbation theory (DFPT)⁶ using a $4 \times 4 \times 1$ supercell. The phonon calculations of six selected materials were performed using a $5 \times 5 \times 1$ supercell with the finite displacement method.^{7, 8} Ab initio molecular dynamics (AIMD) simulations were performed at 300 K using canonical ensemble (NVT) with Nosé-Hoover thermostat^{9, 10} lasting for 6 ps with a time step of 3 fs.

The carrier mobility (μ) was calculated based on the deformation potential theory.¹¹ The carrier mobility (μ) in 2D materials is given as $\mu_{2D} = \frac{e\hbar^3 C_{2D}}{k_B T m^* m_d (E_i)^2}$,

here, $\hbar$ is the reduced Planck constant, k_B is Boltzmann constant, and T is the temperature (here 300 K in this work). C_{2D} is the elastic modulus defined

as $C_{2D} = \frac{1}{S_0} \frac{\partial^2 E}{\partial (l/l_0)^2} \Big|_{l=l_0}$, where E is the total energy of the supercell and S_0 is the area

of the optimized supercell. $m^* = \hbar^2 [\partial^2 E(k) / \partial k^2]^{-1}$ is the carrier effective mass

along x and y directions, and $m_d = \sqrt{m_x^* m_y^*}$ is the carrier reduced effective mass. E_i

represents the deformation potential constant of the CBM (electron) and VBM (hole) along the transport direction, defined by $E_i = \Delta E / (\Delta l / l_0)$, where ΔE is the energy

change of the CBM or VBM under proper cell compression and dilatation, l_0 is the

lattice constant along the transport direction, and Δl is the length of deformation on l_0 ($\Delta l/l_0$ is set to be 0.3% in this work).

The GW approximation^{12, 13} was applied to obtain the quasiparticle (QP) energies via the perturbative solution to the Dyson equation

$$\left[-\frac{\hbar^2}{2m_e} \nabla^2 + V_{\text{ion}} + V_H + \sum (E_{\text{nk}}^{\text{QP}}) \right] \psi_{\text{nk}}^{\text{QP}} = E_{\text{nk}}^{\text{QP}} \psi_{\text{nk}}^{\text{QP}}$$

where m_e is the mass of electron, $\hbar$ is the reduced Planck constant, V_{ion} is the electrostatic potential contributed by ions, V_H is the Hartree potential, and $E_{\text{nk}}^{\text{QP}}$ is the quasi-particle energy.

The optical excitation energies and exciton wave functions are determined through Bethe-Salpeter equation (BSE)^{14, 15, 16}

$$(E_{\text{ck}}^{\text{QP}} - E_{\text{vk}}^{\text{QP}}) A_{\text{vck}}^S + \sum_{\text{v'c'k'}} \langle \text{vck} | K^{\text{eh}} | \text{v'c'k'} \rangle A_{\text{v'c'k'}}^S = \Omega^S A_{\text{vck}}^S$$

where $E_{\text{ck}}^{\text{QP}}$ and $E_{\text{vk}}^{\text{QP}}$ are the quasi-particle energies for conduction and valence bands, K^{eh} and Ω^S are the electron-hole interaction kernel and excitation energy, respectively.

The imaginary part is evaluated from the excitation energies and exciton wave functions with the following expression¹⁶

$$\varepsilon(\omega) = \frac{16\pi e^2}{\omega^2} \sum_{\text{S}} \left| \vec{\lambda} \langle 0 | \vec{v} | \text{S} \rangle \right|^2 \delta(\omega - \Omega^S)$$

where $\vec{\lambda}$ and $\vec{v}$ is the polarization vector of the incident light and velocity operator, respectively, and $\langle 0 | \vec{v} | \text{S} \rangle$ is the transition matrix element.

The absorption coefficient $\alpha(\omega)$ is calculated as follows¹⁷

$$\alpha(\omega) = \sqrt{2} \omega \left[\sqrt{\varepsilon_1^2(\omega) + \varepsilon_2^2(\omega)} - \varepsilon_1(\omega) \right]^{1/2}$$

where $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ are the real and imaginary parts of the dielectric function, respectively.

The kinetic energy cutoff for GW and BSE is 250 eV, and 10 occupied and 10 unoccupied orbitals are used to build the electron-hole interaction kernel.

We use the element-related properties data from the python mendeleev package 0.4.1¹⁸ and the valence electron orbit radius of atoms from Ref.[19].

2. Gradient boosting regression

The machine-learning was created using the Gradient boosted regression (GBR)^{20, 21} in the open-source scikit-learn package.²² The algorithm is based on the learning principle that finding many weakly accurate prediction algorithms can be much easier than finding a single highly accurate algorithm. After multiple iterations, the final regression algorithm is to combine several weak regression algorithms obtained by each of training into a single more accurate than any single weak algorithm, which reads

$$F_M(x) = \sum_m^M \gamma_m h_m(x)$$

where $h_m(x)$ are the weak regressions, x is the input data, γ_m is the weight of each

regression, and M is the number of the weak regressions. At each of iteration, the weight γ_m of next weak algorithms is obtained by minimizing the loss function for all

$$\text{data (count } N) \quad \gamma_m = \arg_{\gamma_m} \min \sum_{i=1}^N L[y_i, F_{m-1}(x_i) + \gamma_m h(x_i)]$$

where $F_{m-1}(x_i)$ is the current model.

Here, the five optimized hyper-parameters in the GBR model are obtained by grid searching method: loss function (least squares), learning rate (0.1), maximum depth of the individual regression estimators (7), maximum leaf nodes of the individual regression estimators (7) and the number of learning algorithms (200).

3. Model evaluation

The coefficient of determination (R^2), employed to evaluate the goodness of fit, is defined as

$$R^2 = 1 - \frac{\sum_i (y_i^{\text{True}} - y_i^{\text{Pred}})^2}{\sum_i (y_i^{\text{True}} - \overline{y_i^{\text{True}}})^2}$$

where y_i^{True} is the true value, y_i^{Pred} is the predicted value, N is the number of data and p is the number of fitting parameters. The closer to 1 the value of R^2 , the better the fitting degree on true values.

The mean squared error (MSE) represents the expected value of the square of the difference between the fitting values and true data, which is defined as

$$\text{MSE} = \frac{1}{N} \sum_i (y_i^{\text{True}} - y_i^{\text{Pred}})^2$$

The correlation between predictive value and real value can be reflected by Pearson coefficient (r):

$$r^2 = \frac{\sum_{i=1}^N (y_i^{\text{True}} - \overline{y_i^{\text{True}}})(y_i^{\text{Pred}} - \overline{y_i^{\text{Pred}}})}{\sqrt{\sum_{i=1}^N (y_i^{\text{True}} - \overline{y_i^{\text{True}}})^2 (y_i^{\text{Pred}} - \overline{y_i^{\text{Pred}}})^2}}$$

The value is between -1 and +1. If r is larger than zero, implying that the two variables are positively correlated. If r is less than zero, suggesting two variables are negatively correlated. Moreover, the greater the absolute value of r , the stronger the correlation.

4. Initial features

Table S1. 62 Initial features with definition

Features	Definition
$r_A, r_{B1}, r_{B2}, r_{X1}, r_{X2}$	ion radii of A, B1, B2, X1 and X2-site atoms
$DSOf_1$	First distorted stacked octahedral factor defined as $\frac{r_A + r_{B2}}{r_{B1} + r_{B2}}$
$DSOf_2$	Second distorted stacked octahedral factor defined as $\frac{r_{B2}}{r_{B2} + r_{X1}}$
$DSOf_3$	Third distorted stacked octahedral factor defined as $\frac{r_A + r_{X1}}{r_{B1} + r_{X1}}$
$DSOf_4$	Fourth distorted stacked octahedral factor defined as $\frac{r_{X1}}{r_{B2} + r_{X1}}$
$DSOf_5$	Fifth distorted stacked octahedral factor defined as $\frac{r_A + r_{B1}}{r_{B1} + r_{B2}}$
$DSOf_6$	Sixth distorted stacked octahedral factor defined as $\frac{r_{B1}}{r_{B1} + r_{X2}}$
$DSOf_7$	Seventh distorted stacked octahedral factor defined as $\frac{r_A + r_{X2}}{r_{X2} + r_{B2}}$
$DSOf_8$	Eighth distorted stacked octahedral factor defined as $\frac{r_{X2}}{r_{B1} + r_{X2}}$
$IE_A, IE_{B1}, IE_{B2}, IE_{X1}, IE_{X2}$	Ionization energy of A, B1, B2, X1 and X2-site atoms
$EA_{B1}, EA_{B2}, EA_{X1}, EA_{X2}$	Electron affinity of B1, B2, X1 and X2-site atoms
$P_A, P_{B1}, P_{B2}, P_{X1}, P_{X2}$	Ionic polarizability of A, B1, B2, X1 and X2-site atoms
$X_A, X_{B1}, X_{B2}, X_{X1}, X_{X2}$	Martynov-Batsanov electronegativity of A, B1, B2, X1 and X2-site atoms
$IC_A, IC_{B1}, IC_{B2}, IC_{X1}, IC_{X2}$	Ionic charge of A, B1, B2, X1 and X2-site atoms

$n_A, n_{B1}, n_{B2}, n_{X1}, n_{X2}$	Number of valence electrons of A, B1, B2, X1 and X2-site atoms
$n_{A(in)}, n_{A(out)}$	Number of valence electrons in the inner/outer shell of A atom
$r_{A(in)}, r_{A(out)}$	Inner/outer valence electrons orbital radius of A atom
$n_{B1(s)}, n_{B1(p)}, n_{B2(s)}, n_{B2(p)}, n_{X1(s)}, n_{X1(p)}, n_{X2(s)}, n_{X2(p)}$	Number of s, p valence electrons of B1, B2, X1 and X2-site atoms
$r_{B1(s)}, r_{B1(p)}, r_{B2(s)}, r_{B2(p)}, r_{X1(s)}, r_{X1(p)}, r_{X2(s)}, r_{X2(p)}$	The s, p valence electrons orbital radius of B1, B2, X1 and X2-site atoms

5. Feature reduction

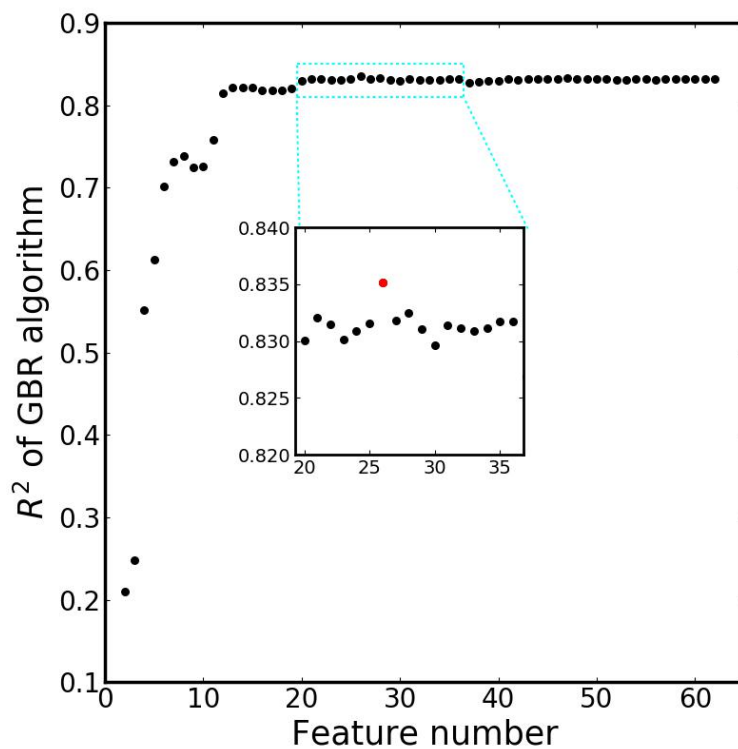


Figure S1. Determination coefficient (R^2) of GBR model with increasing the feature numbers. The position of the red pot is the maximum value of R^2 (0.835), corresponding to 26 feature numbers.

As shown in Figure S1, using the principal component analysis (PCA) method²³, we acquire the importance of all 62 initial features, and then we perform a feature reduction by excluding the features with less impact until R^2 achieves a maximum (0.835). In the end, we find that 26 features form an optimal vector of features.

6. Algorithm selection

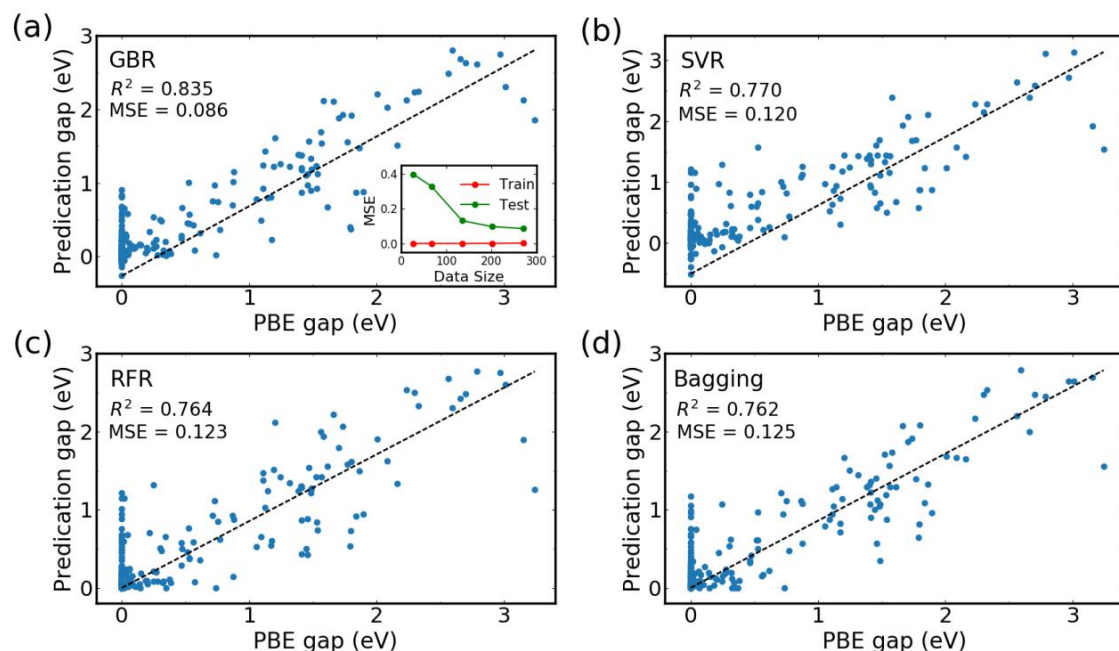


Figure S2. Ten-fold cross-validation plots for the four Machine-learning algorithms to predict the band gaps. Models are evaluated with coefficient of determination (R^2) and mean squared error (MSE) of (a) gradient boosted regression (GBR), (b) support vector regression (SVR), (c) random forest regression (RFR) and (d) bagging. The inset of (a) is the convergence of learning curve for ten-fold cross-validation split of the data.

We have tested various algorithms such as the support vector regression (SVR),²⁴ random forest regression (RFR),²⁵ bagging²⁶ and gradient boosting regression (GBR), which all have been successfully used for machine-learning prediction in materials.²⁷ In Figure S2, the results of 10-fold cross-validation regression analysis and grid searching best hyper-parameters technique show that the GBR model outperforms others and is the best model with the lowest mean square error ($MSE = 0.086$) and largest coefficient of determination value ($R^2 = 0.835$).

7. Comparison of various feature combinations

Table S2. Determination coefficient (R^2) and mean square errors (MSE) on test data for various feature combinations with the GBR algorithm

Feature combinations	R^2	MSE
<i>DSOf</i> + element-related properties	0.835	0.086
<i>DSOf</i> + <i>Of</i> + element-related properties	0.736	0.139
<i>Of</i> + element-related properties	0.741	0.152
Element-related properties	0.748	0.147

We consider *DSOf* as better features than *Of* since *DSOf* can describe the geometric relation of the inequitable atoms in octahedrons in an even better fashion. To verify, we compare four different feature vectors (features of element-related properties; features of element-related properties plus *Of*; features of element-related properties plus *DSOf*; features of element-related properties plus both *DSOf* and *Of*), and find that the feature vectors including element-related properties and *DSOf* give the best prediction accuracy among the others (see Table S2).

8. Comparison between Machine-learning-predicted and DFT-calculated band gaps

Table S3. Preliminary screened 73 OOHs, the comparison between the band gaps predicted by Machine-learning and DFT calculations and mean squared error (MSE)

No.	Structure	E_g^{ML}	E_g^{DFT}
1	As ₂ O ₂ BrI	1.364	1.597
2	Sb ₂ O ₂ BrF	1.083	1.597
3	Sb ₂ O ₂ BrI	0.926	1.589
4	In ₂ OSBrF	1.062	1.577
5	As ₂ O ₂ ClI	1.342	1.550
6	Bi ₂ S ₂ BrF	1.403	1.550
7	Al ₂ Se ₂ I ₂	1.250	1.518
8	Al ₂ Se ₂ BrI	1.229	1.514
9	P ₂ O ₂ Cl ₂	1.070	1.507
10	Bi ₂ S ₂ ClF	1.330	1.503
11	In ₂ OSeCl ₂	1.200	1.495
12	Al ₂ OSeClI	1.234	1.470
13	In ₂ OSClI	1.384	1.469
14	Al ₂ Se ₂ ClBr	1.443	1.436
15	Ga ₂ S ₂ ClBr	1.348	1.430
16	In ₂ SSeI ₂	1.252	1.429
17	Bi ₂ SSeBr ₂	1.295	1.424
18	Ga ₂ S ₂ Cl ₂	1.252	1.418
19	Al ₂ OSeBrI	1.176	1.411
20	Bi ₂ OSClF	1.409	1.406
21	Al ₂ Se ₂ Cl ₂	1.264	1.398
22	In ₂ OSeBr ₂	1.280	1.385
23	Bi ₂ OSClBr	1.568	1.381
24	Bi ₂ OSClI	1.524	1.374
25	In ₂ SSeClBr	1.083	1.373
26	Bi ₂ OSFI	1.459	1.367
27	P ₂ O ₂ F ₂	1.221	1.363

28	$\text{Al}_2\text{Se}_2\text{ClI}$	1.121	1.348
29	Bi_2OSCl_2	1.459	1.341
30	In_2SSeBrI	1.158	1.339
31	$\text{In}_2\text{SSeCl}_2$	0.958	1.338
32	$\text{P}_2\text{O}_2\text{ClBr}$	0.955	1.324
33	$\text{Al}_2\text{S}_2\text{FI}$	1.030	1.323
34	Ga_2OSBrF	1.131	1.280
35	In_2OSI_2	1.338	1.255
36	In_2SSeClI	0.995	1.255
37	Al_2SSeClF	1.290	1.243
38	Al_2SSeBrF	1.309	1.206
39	$\text{Sb}_2\text{O}_2\text{FI}$	0.964	1.198
40	Bi_2OSBr_2	1.557	1.183
41	$\text{Bi}_2\text{Se}_2\text{I}_2$	1.169	1.175
42	$\text{Bi}_2\text{Se}_2\text{Br}_2$	1.193	1.172
43	Bi_2SSeBrF	1.171	1.171
44	$\text{Bi}_2\text{Se}_2\text{BrI}$	1.223	1.165
45	Bi_2OSF_2	1.341	1.122
46	$\text{Sb}_2\text{S}_2\text{Cl}_2$	0.921	1.118
47	Al_2OSFI	1.387	1.109
48	In_2OSFI	1.111	1.100
49	Bi_2OSI_2	1.469	1.072
50	Al_2OSeF_2	1.438	1.067
51	Al_2OSI_2	1.419	1.062
52	$\text{Sb}_2\text{S}_2\text{BrF}$	1.013	1.051
53	$\text{As}_2\text{O}_2\text{FI}$	1.551	1.049
54	$\text{Sb}_2\text{S}_2\text{Br}_2$	1.147	1.047
55	In_2OSeClI	1.107	1.028
56	$\text{Ga}_2\text{S}_2\text{BrI}$	1.306	1.025
57	$\text{Ga}_2\text{SSeBr}_2$	0.937	1.016
58	$\text{Bi}_2\text{Se}_2\text{ClF}$	1.008	1.012
59	Al_2OSeBrF	1.599	1.002
60	$\text{P}_2\text{O}_2\text{ClI}$	0.957	0.960

61	In ₂ S ₂ FI	1.074	0.960
62	Sb ₂ S ₂ BrI	0.903	0.950
63	Al ₂ OSeFI	1.142	0.946
64	Ga ₂ S ₂ I ₂	1.398	0.943
65	In ₂ S ₂ BrF	1.094	0.941
66	Ga ₂ OSBrI	1.052	0.903
67	Al ₂ TeSeII	1.047	0.777
68	Bi ₂ OSeFI	0.917	0.763
69	Ga ₂ OSClI	1.063	0.754
70	Bi ₂ OSeBrF	1.103	0.739
71	Al ₂ S ₂ BrF	1.264	1.877
72	Bi ₂ S ₂ Cl ₂	1.505	1.815
73	Al ₂ OSClI	1.434	1.725
MSE		0.071	

9. Structural details

Table S4. Structural details for Bi₂Se₂Br₂, Bi₂Se₂BrI, Bi₂Se₂I₂, In₂S₂BrF, In₂SSeClBr and In₂SSeCl₂ monolayers

Structure	Spacegroup	Lattice Constants	Atom positions
Bi ₂ Se ₂ Br ₂	Pmmn: No.59	a=4.2052 Å b=5.8758 Å	Bi1: (0, 0, 0.18858)
			Bi2: (0.5, 0.5, 0.06713)
			Se1: (0, 0.5, 0.16846)
			Se2: (0.5, 0, 0.08725)
			Br1: (0, 0.5, 0.96540)
			Br2: (0.5, 0, 0.29031)
Bi ₂ Se ₂ BrI	Pmm2: No.25	a=4.2627 Å b=5.8645 Å	Bi1: (0, 0, 0.18637)
			Bi2: (0.5, 0.5, 0.0649)
			Se1: (0, 0.5, 0.16533)
			Se2: (0.5, 0, 0.08629)
			Br: (0, 0.5, 0.96412)
			I: (0.5, 0, 0.30012)
Bi ₂ Se ₂ I ₂	Pmmn: No.59	a=4.3204 Å b=5.8538 Å	Bi1: (0, 0, 0.18852)
			Bi2: (0.5, 0.5, 0.06719)
			Se1: (0, 0.5, 0.16630)
			Se2: (0.5, 0, 0.08941)
			I1: (0, 0.5, 0.95429)
			I2: (0.5, 0, 0.30142)
In ₂ S ₂ BrF	Pmm2: No.25	a=3.6994 Å b=5.3477 Å	In1: (0, 0, 0.18556)
			In2: (0.5, 0.5, 0.08249)
			S1: (0, 0.5, 0.17437)
			S2: (0.5, 0, 0.09386)
			Br: (0, 0.5, 0.98393)
			F: (0.5, 0, 0.24692)
In ₂ SSeClBr	Pmm2: No.25	a=3.8386 Å b=5.5574 Å	In1: (0, 0, 0.17912)
			In2: (0.5, 0.5, 0.0761)
			S: (0, 0.5, 0.16456)
			Se: (0.5, 0, 0.0844)

$\text{In}_2\text{SSeCl}_2$	Pmm2: No.25	$a=3.7990 \text{ \AA}$ $b=5.5521 \text{ \AA}$	Cl: (0, 0.5, 0.98713)
			Br: (0.5, 0, 0.27581)
			In1: (0, 0, 0.18028)
			In2: (0.5, 0.5, 0.07792)
			S: (0, 0.5, 0.16705)
			Se: (0.5, 0, 0.08479)
			Cl1: (0, 0.5, 0.98844)
			Cl2: (0.5, 0, 0.26864)

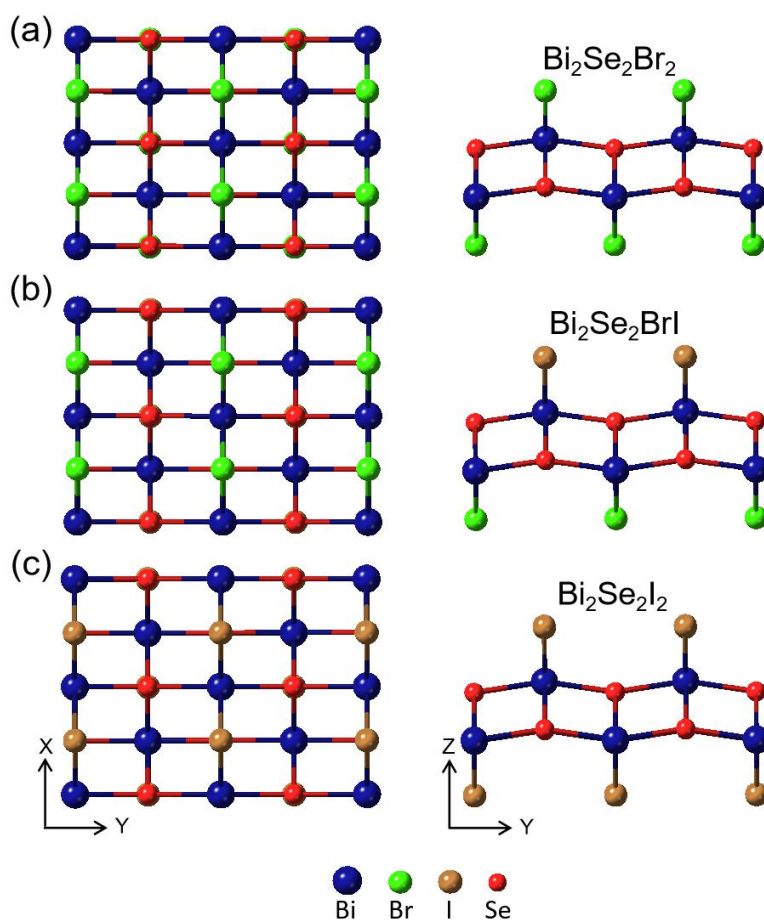


Figure S3. Crystal structures of three candidates of OOHs. Top view from z axis and side view from x axis of 2x2 unit cell of (a) $\text{Bi}_2\text{Se}_2\text{Br}_2$, (b) $\text{Bi}_2\text{Se}_2\text{BrI}$ and (c) $\text{Bi}_2\text{Se}_2\text{I}_2$.

10. Electronic structures

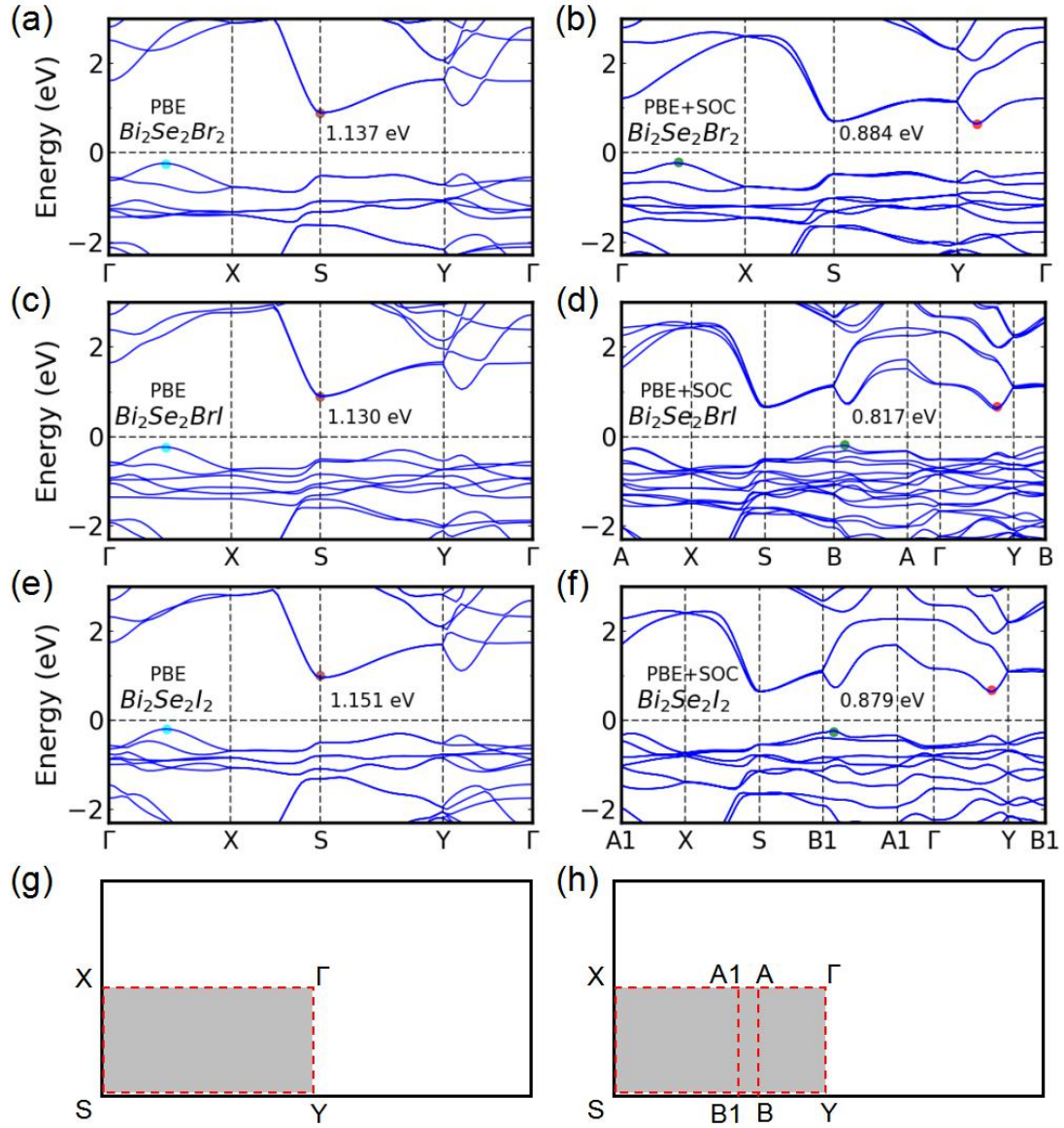


Figure S4. Electronic band structures of $\text{Bi}_2\text{Se}_2\text{Br}_2$ (a) and (b), $\text{Bi}_2\text{Se}_2\text{BrI}$ (c) and (d), and $\text{Bi}_2\text{Se}_2\text{I}_2$ (e) and (f) calculated at the PBE, and PBE+SOC level, as well as the high symmetry points labeled in the first Brillouin zone (g) and (h).

Detailed analysis of the influence of SOC effect on conduction band minima (CBM) and the valence band maxima (VBM) reveals that the position of CBM moves from S point to the point between Γ and Y. For $\text{Bi}_2\text{Se}_2\text{BrI}$ and $\text{Bi}_2\text{Se}_2\text{I}_2$ monolayers, the position of VBM shifts from the point in the high symmetry path to the point inner Brillouin zone (Figure S4(g) and (h)).

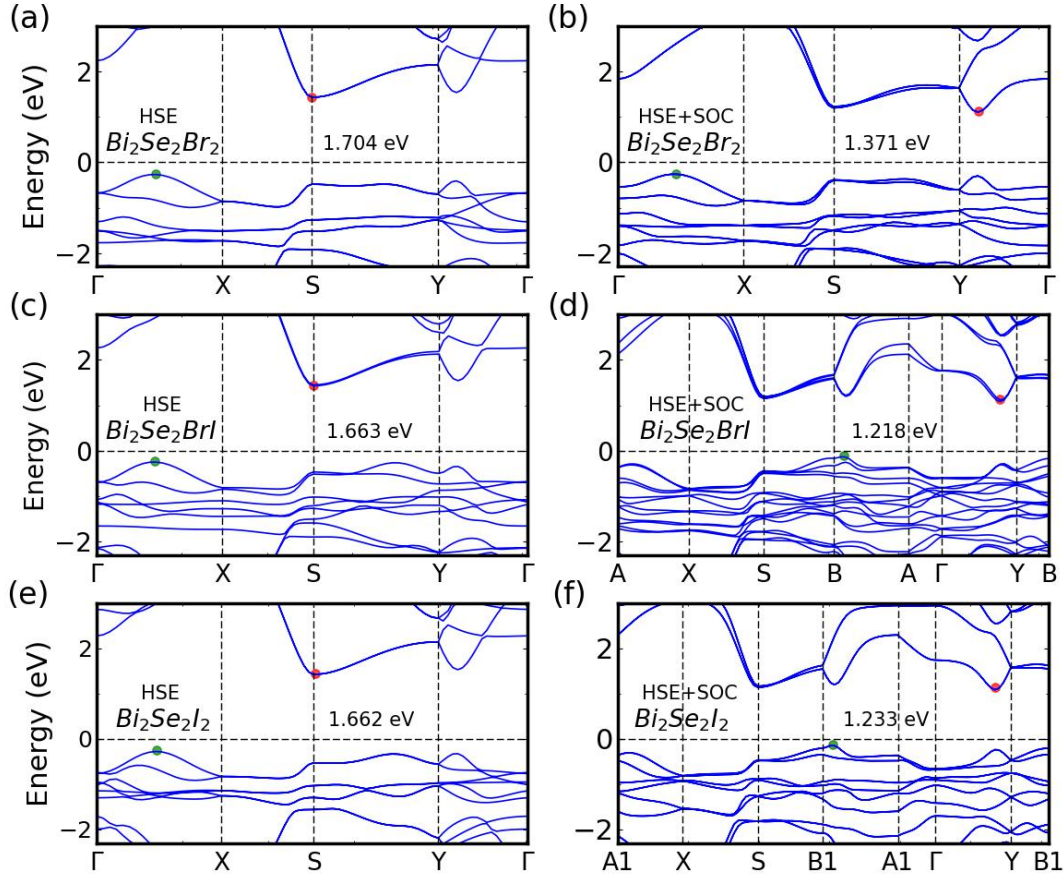


Figure S5. Electronic band structures of $\text{Bi}_2\text{Se}_2\text{Br}_2$ (a) and (b), $\text{Bi}_2\text{Se}_2\text{BrI}$ (c) and (d) and $\text{Bi}_2\text{Se}_2\text{I}_2$ (e) and (f) calculated at the HSE, and HSE+SOC level.

For $\text{Bi}_2\text{Se}_2\text{Br}_2$, $\text{Bi}_2\text{Se}_2\text{BrI}$ and $\text{Bi}_2\text{Se}_2\text{I}_2$ monolayers, the more elaborate hybrid HSE06 method²⁸ was further employed to obtain the accurate electronic band structures. Spin-orbit coupling (SOC) was considered in calculations due to the existence of heavy elements. But, at HSE06+SOC level, the band gaps become 1.371, 1.218 and 1.233 eV, respectively, indicating that these band gaps are suitable for photovoltaic devices.

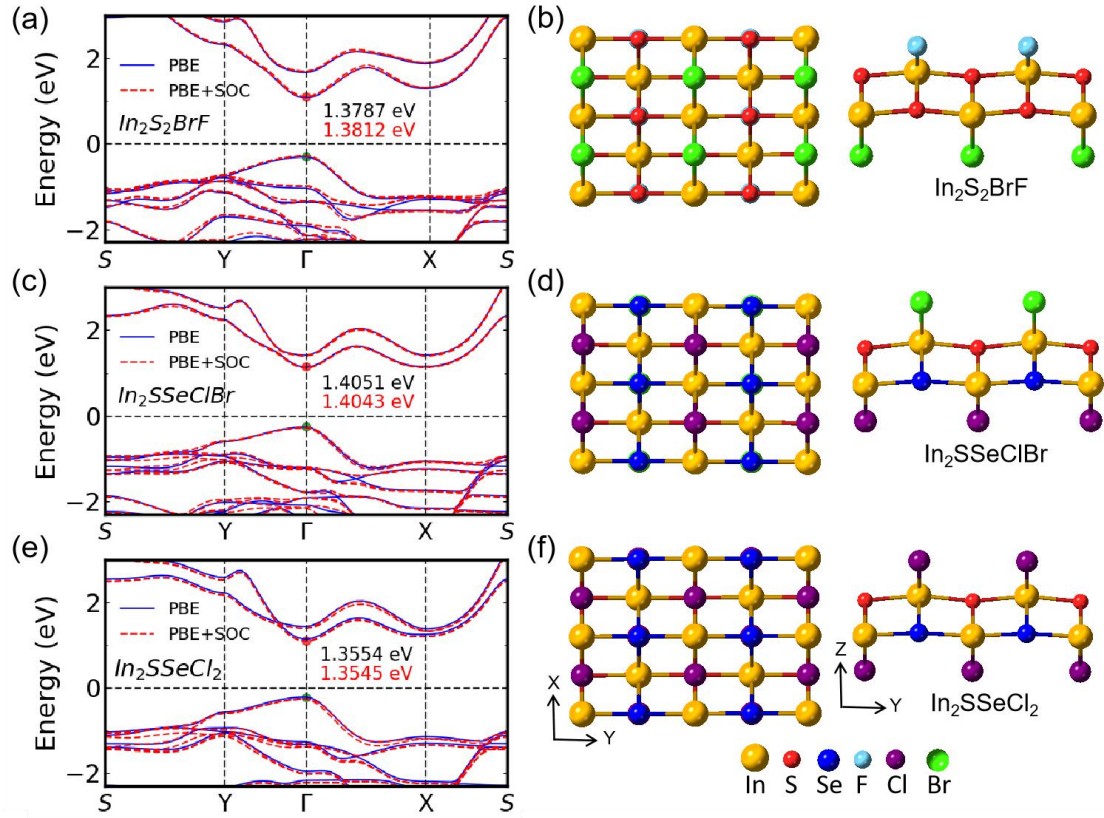


Figure S6. Electronic band structures and crystal structures. Band structures of $\text{In}_2\text{S}_2\text{BrF}$ (a), $\text{In}_2\text{SSeClBr}$ (c) and $\text{In}_2\text{SSeCl}_2$ (e) calculated at the PBE (blue line), and PBE+SOC level (red dash line). Top view from z axis and side view from x axis of 2×2 unit cell of $\text{In}_2\text{S}_2\text{BrF}$ (b), $\text{In}_2\text{SSeClBr}$ (d) and $\text{In}_2\text{SSeCl}_2$ (f).

As is shown in Figure S6, the SOC effect is not pronounced and the band structures obtained by PBE and PBE+SOC are almost same. Therefore, we neglected the SOC effect on $\text{In}_2\text{S}_2\text{BrF}$, $\text{In}_2\text{SSeClBr}$ and $\text{In}_2\text{SSeCl}_2$.

11. Calculated phonon dispersions and AIMD evolutions for the monolayers

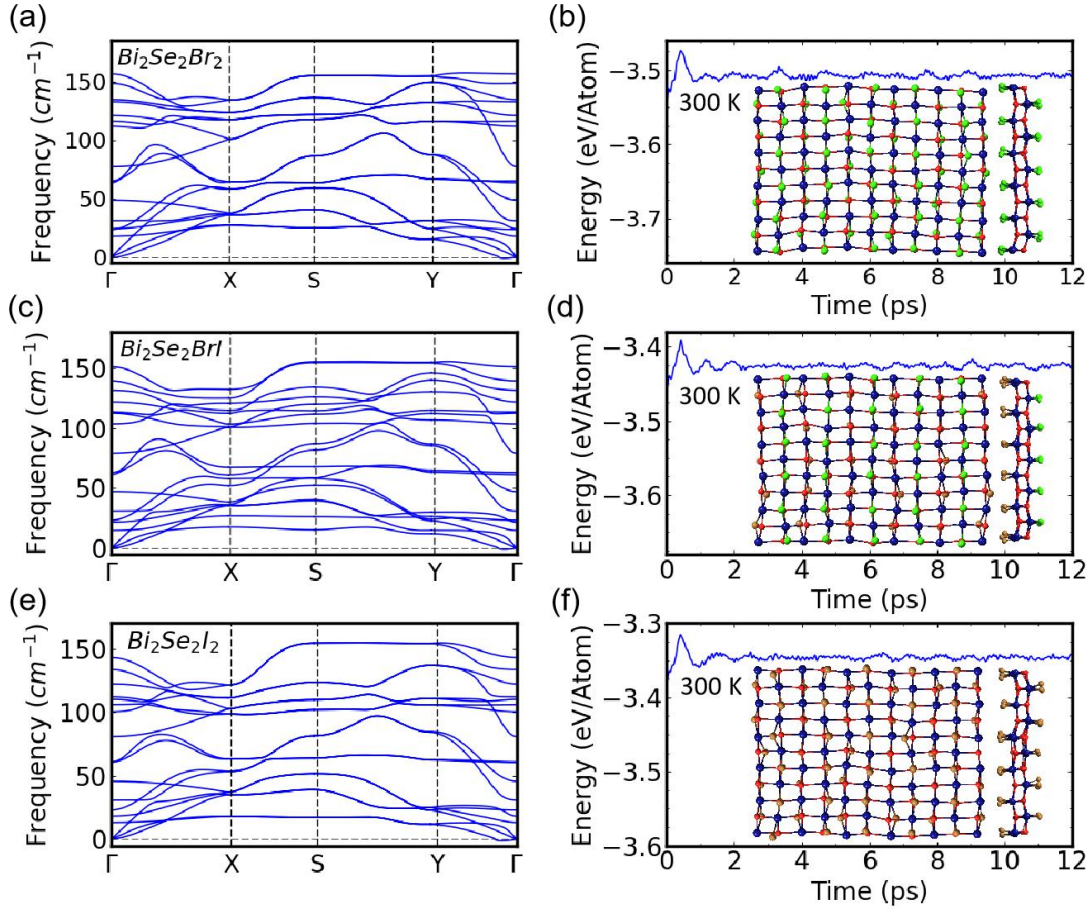


Figure S7. Calculated phonon dispersions and AIMD evolutions of average energy per atom for $\text{Bi}_2\text{Se}_2\text{Br}_2$ (a) and (b), $\text{Bi}_2\text{Se}_2\text{BrI}$ (c) and (d), and $\text{Bi}_2\text{Se}_2\text{I}_2$ (e) and (f) monolayers. The insets show the snapshots of the three structures at 300 K after 12 ps AIMD simulations.

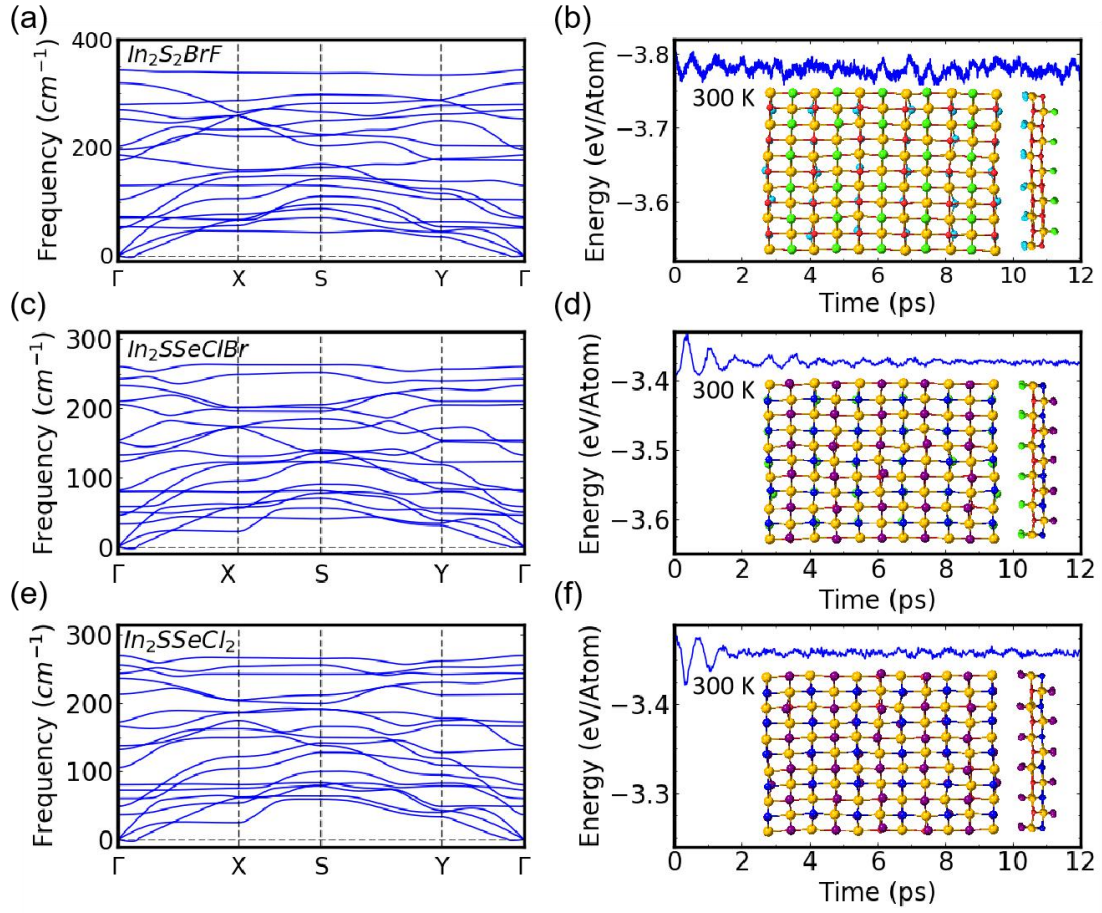


Figure S8. Calculated phonon dispersions and AIMD evolutions of average energy per atom for $\text{In}_2\text{Se}_2\text{BrF}$ (a) and (b), $\text{In}_2\text{SSeClBr}$ (c) and (d) and $\text{In}_2\text{SSeCl}_2$ (e) and (f) monolayers. The insets show the snapshots of the three structures at 300 K after 12 ps AIMD simulations.

12. Carrier mobility

Table S5. PBE-calculated elastic modulus (C_{2D}), effective mass (m/m_0), deformation potential constant (E_i), and carrier mobility (μ) in x and y directions for $\text{Bi}_2\text{Se}_2\text{Br}_2$, $\text{Bi}_2\text{Se}_2\text{BrI}$ and $\text{Bi}_2\text{Se}_2\text{I}_2$ monolayers at 300 K.

PBE	Carrier	direction	C_{2D} (N/m)	m/m_0	E_i (eV)	μ ($10^3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$)
$\text{Bi}_2\text{Se}_2\text{Br}_2$	electron	x	37.13	0.057	0.93	76.54
		y	59.73	0.762	-11.04	0.065
	hole	x		-1.375	-1.27	0.394
		y		-0.580	-3.61	0.187
$\text{Bi}_2\text{Se}_2\text{BrI}$	electron	x	36.37	0.059	1.04	59.56
		y	58.29	0.707	-11.06	0.069
	hole	x		-1.693	-1.61	0.183
		y		-0.539	-3.16	0.240
$\text{Bi}_2\text{Se}_2\text{I}_2$	electron	x	35.04	0.061	0.92	67.66
		y	57.06	0.727	-10.90	0.066
	hole	x		-1.899	-1.99	0.106
		y		-0.449	-2.84	0.360

Table S6. PBE+SOC-calculated elastic modulus (C_{2D}), effective mass (m/m_0), deformation potential constant (E_i), and carrier mobility (μ) in x and y directions for $\text{Bi}_2\text{Se}_2\text{Br}_2$, $\text{Bi}_2\text{Se}_2\text{BrI}$ and $\text{Bi}_2\text{Se}_2\text{I}_2$ monolayers at 300 K.

PBE+SOC	Carrier	direction	C_{2D} (N/m)	m/m_0	E_i (eV)	μ ($10^3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$)
$\text{Bi}_2\text{Se}_2\text{Br}_2$	electron	x	35.14	0.09	-0.56	89.8
		y	56.61	0.953	-4.31	0.23
	hole	x		-1.722	-1.42	0.22
		y		-0.57	-3.75	0.15
$\text{Bi}_2\text{Se}_2\text{BrI}$	electron	x	33.77	0.096	-0.64	56.16
		y	52.30	1.089	-3.26	0.30
	hole	x		-0.229	-2.23	1.66
		y		-0.62	-1.98	1.21
$\text{Bi}_2\text{Se}_2\text{I}_2$	electron	x	32.84	0.095	-0.72	47.7
		y	53.05	0.947	-3.94	0.26
	hole	x		-0.207	-1.87	2.37
		y		-0.801	-1.99	0.87

Table S7. PBE-calculated elastic modulus (C_{2D}), effective mass (m/m_0), deformation potential constant (E_i), and carrier mobility (μ) in x and y directions for $\text{In}_2\text{S}_2\text{BrF}$, $\text{In}_2\text{SSeClBr}$ and $\text{In}_2\text{SSe}_2\text{Cl}_2$ monolayers at 300 K.

PBE	Carrier	direction	$C_{2D}(\text{N/m})$	m/m_0	$E_i(\text{eV})$	$\mu (10^3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1})$
$\text{In}_2\text{S}_2\text{BrF}$	electron	x	77.42	0.362	-5.43	0.42
		y	57.92	0.363	-0.52	34.2
	hole	x		-2.1	-5.11	0.037
		y		-0.313	-1.96	1.25
$\text{In}_2\text{SSeClBr}$	electron	x	65.30	0.367	-9.10	0.13
		y	47.49	0.338	1.70	2.93
	hole	x		-1.895	-7.44	0.018
		y		-0.258	-0.90	7.04
$\text{In}_2\text{SSe}_2\text{Cl}_2$	electron	x	66.47	0.345	-8.66	0.16
		y	50.10	0.350	-1.80	2.70
	hole	x		-1.956	-7.82	0.017
		y		-0.244	-0.54	21.5

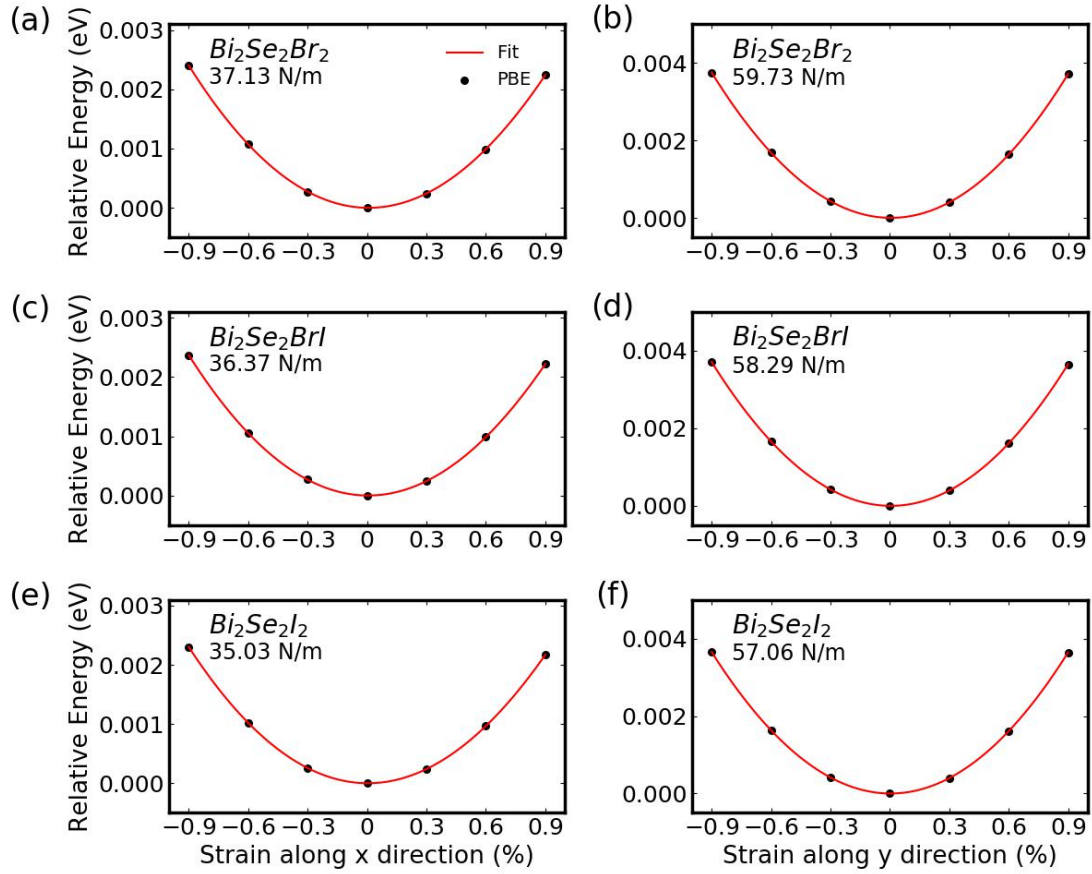


Figure S9. Relative energy between the total energy of unstrained and strained $Bi_2Se_2Br_2$ (a) and (b), Bi_2Se_2BrI (c) and (d), and $Bi_2Se_2I_2$ (e) and (f) monolayers along x and y directions, respectively, at PBE level. Corresponding elastic modulus are marked in each figure.

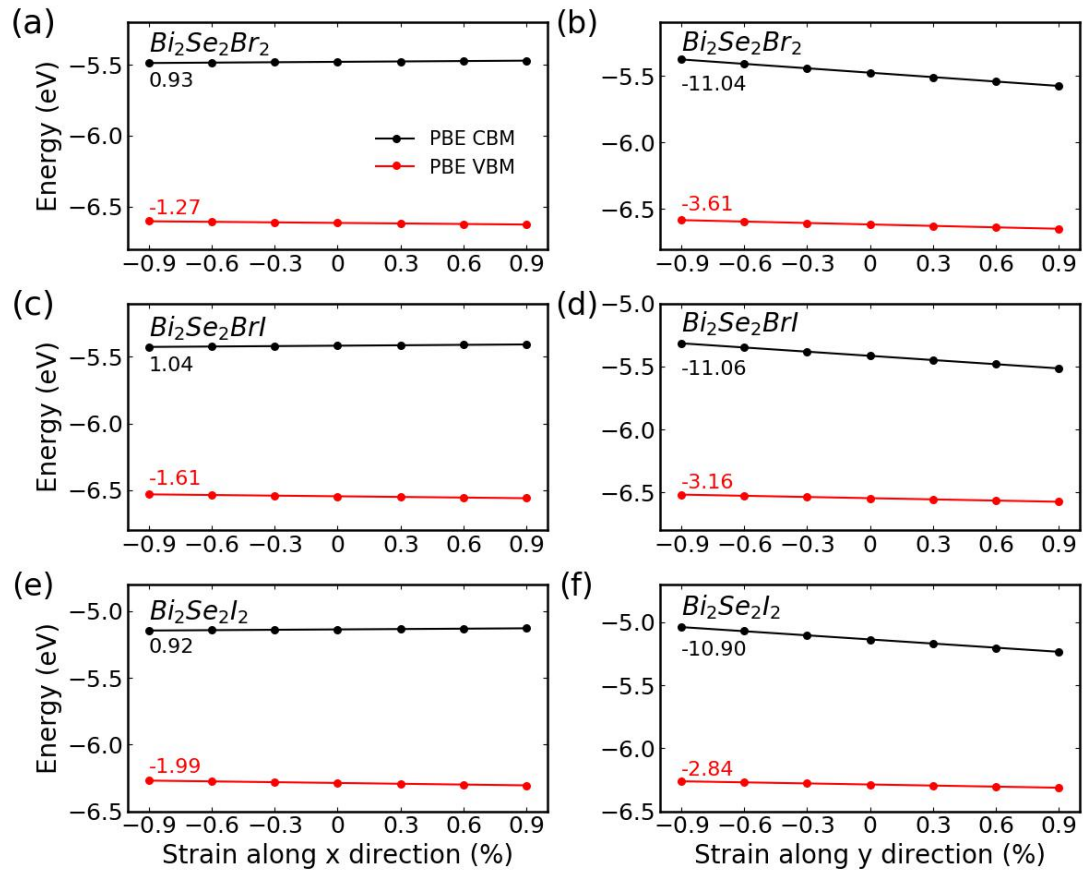


Figure S10. Energy shift of CBM and VBM for $\text{Bi}_2\text{Se}_2\text{Br}_2$ (a, b), $\text{Bi}_2\text{Se}_2\text{BrI}$ (c, d) and $\text{Bi}_2\text{Se}_2\text{I}_2$ (e, f) monolayers with respect to the lattice stretch and compression along x and y directions, respectively, at PBE level. Corresponding Deformation Potential constants are marked in each figure.

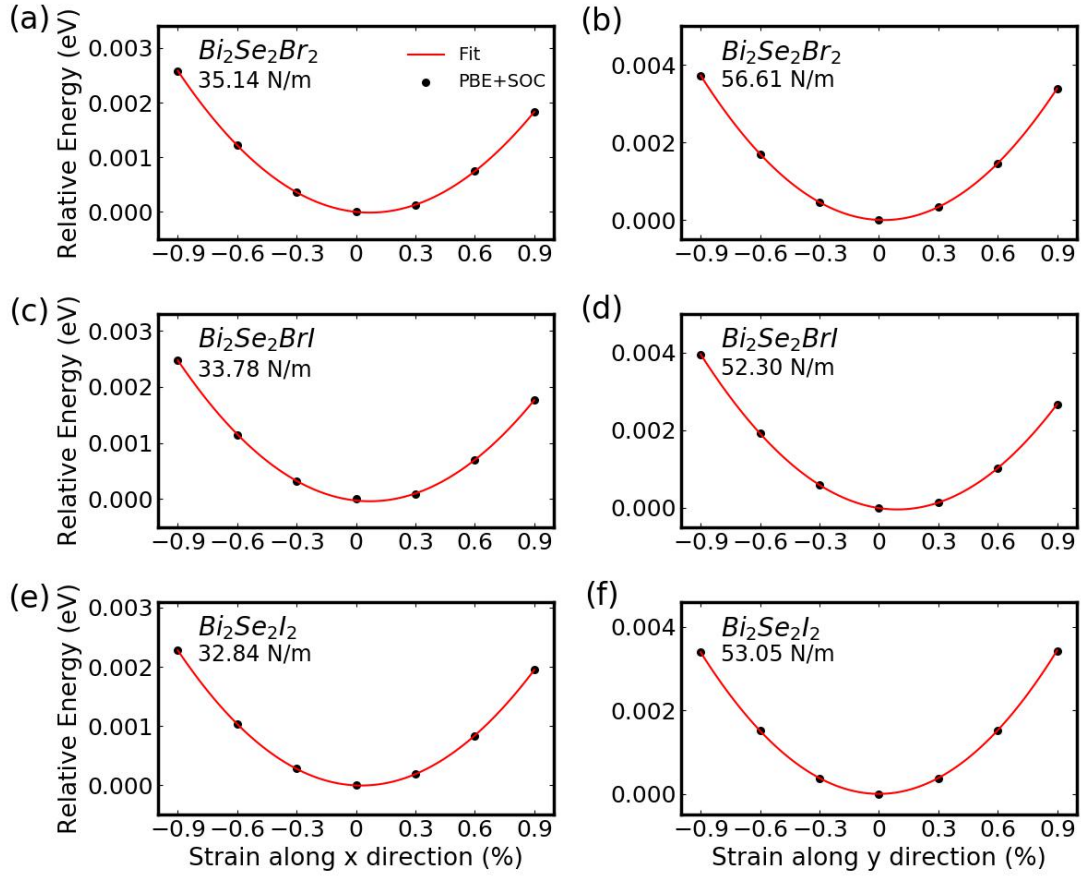


Figure S11. Relative energy between the total energy of unstrained and strained $Bi_2Se_2Br_2$ (a, b), Bi_2Se_2BrI (c, d) and $Bi_2Se_2I_2$ (e, f) monolayers along x and y directions, respectively, at PBE+SOC level. Corresponding elastic modulus are marked in each figure.

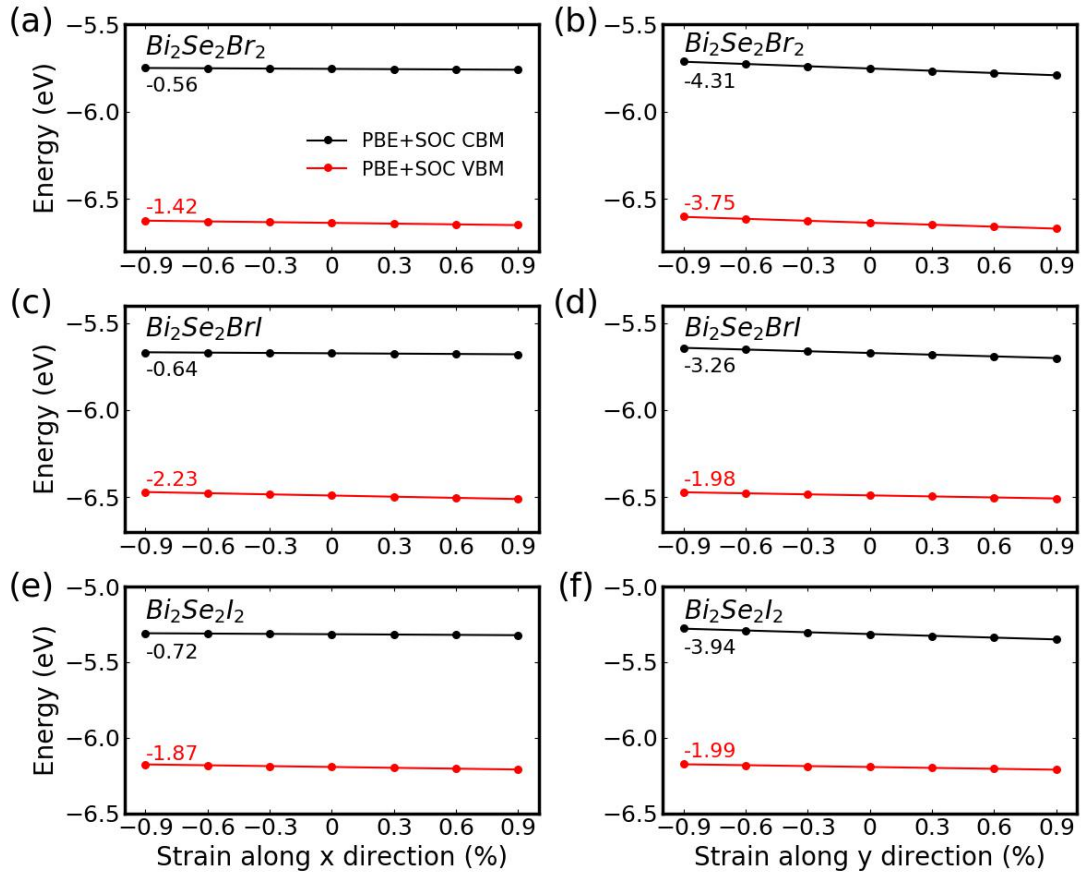


Figure S12. Energy shift of CBM and VBM for $\text{Bi}_2\text{Se}_2\text{Br}_2$ (a, b), $\text{Bi}_2\text{Se}_2\text{BrI}$ (c, d) and $\text{Bi}_2\text{Se}_2\text{I}_2$ (e, f) monolayers with respect to the lattice stretch and compression along x and y directions, respectively, at PBE+SOC level. Corresponding deformation potential constants are marked in each figure.

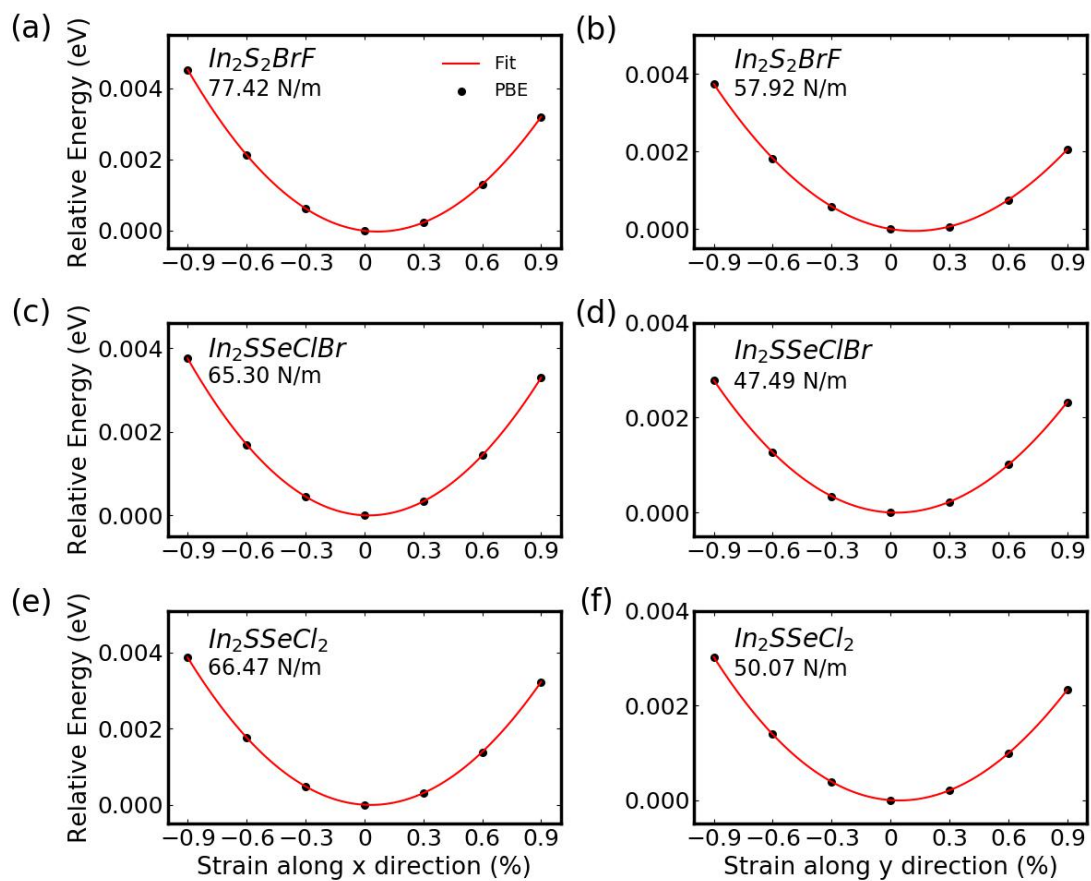


Figure S13. Relative energy between the total energy of unstrained and strained $\text{In}_2\text{S}_2\text{BrF}$ (a, b), $\text{In}_2\text{SSeClBr}$ (c, d) and $\text{In}_2\text{SSeCl}_2$ (e, f) monolayers along x and y directions, respectively, at PBE level. Corresponding elastic modulus are marked in each figure.

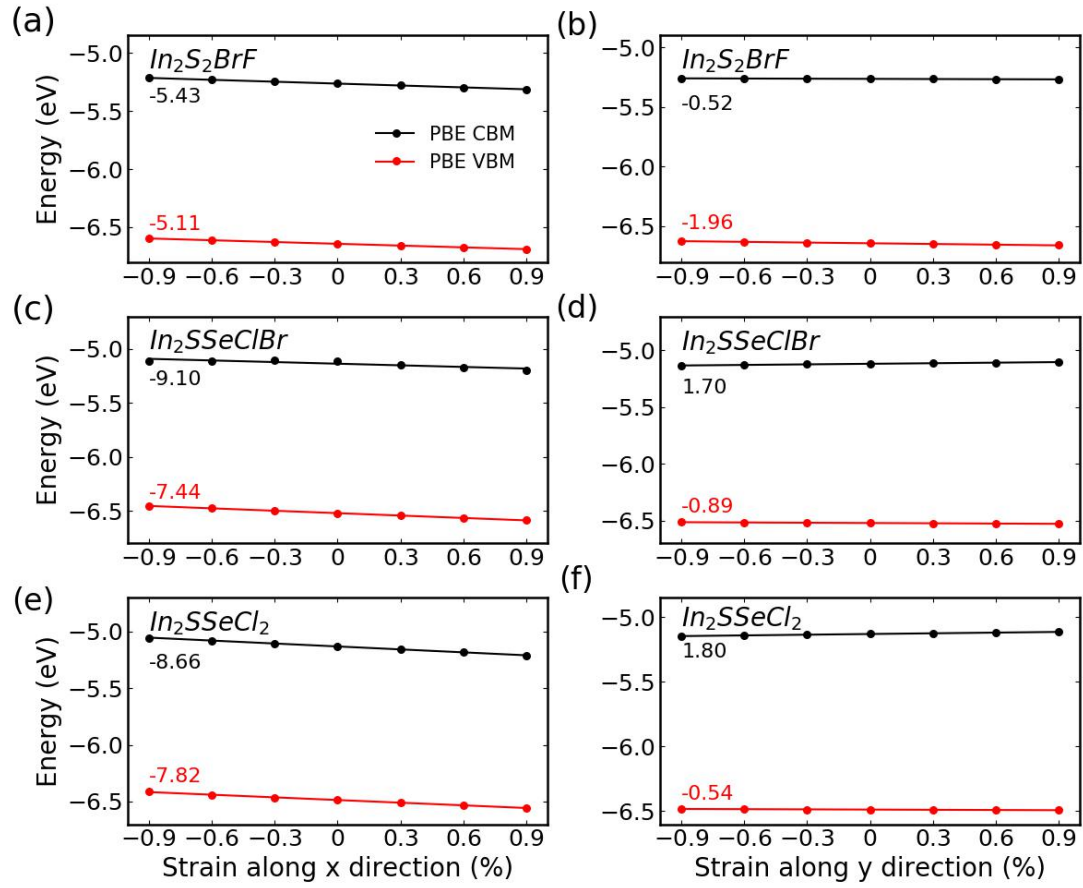


Figure S14. Energy shift of CBM and VBM for the $\text{In}_2\text{S}_2\text{BrF}$ (a, b), $\text{In}_2\text{SSeClBr}$ (c, d) and $\text{In}_2\text{SSeCl}_2$ (e, f) with respect to the lattice stretch and compression along x and y directions, respectively, at PBE level. Corresponding deformation Potential constants are marked in each figure.

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