

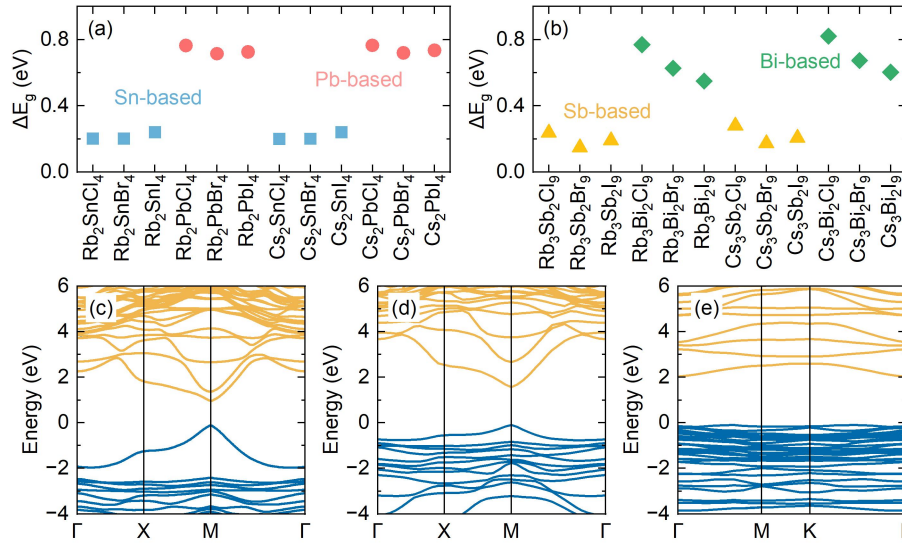
## **Supplementary Information**

### **Facet-Dependent Exfoliation Feasibility and Optoelectronic Properties of Two-Dimensional All-Inorganic Halide Perovskites**

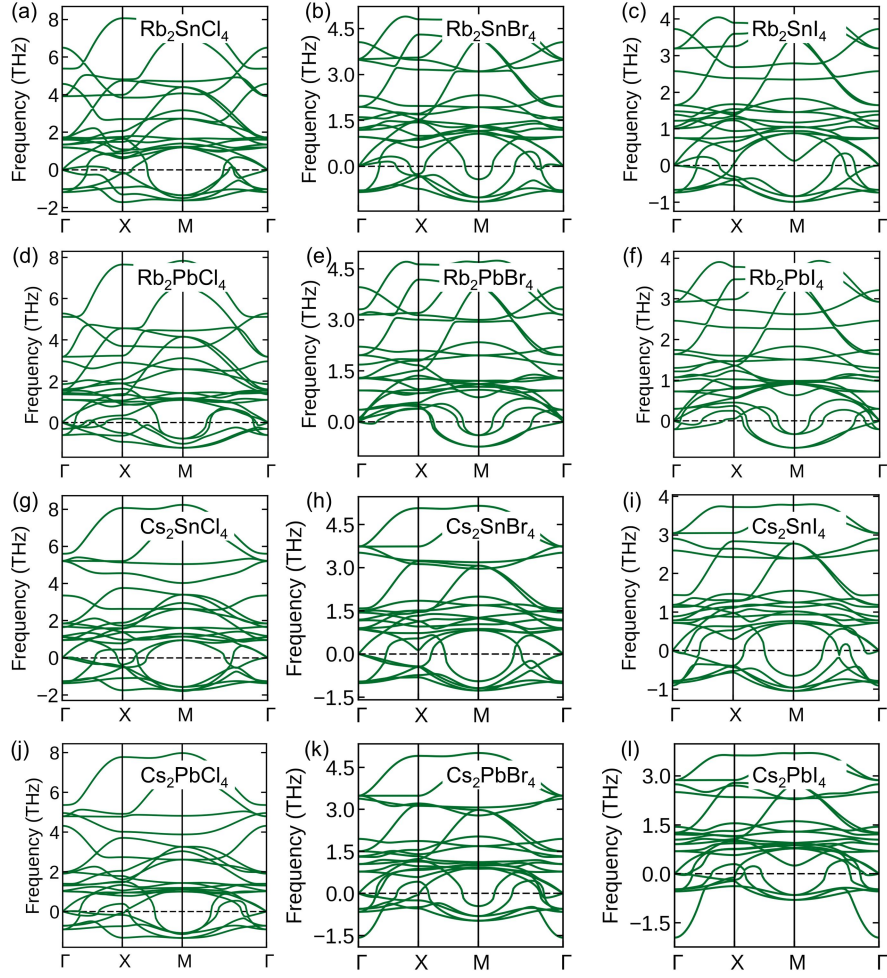
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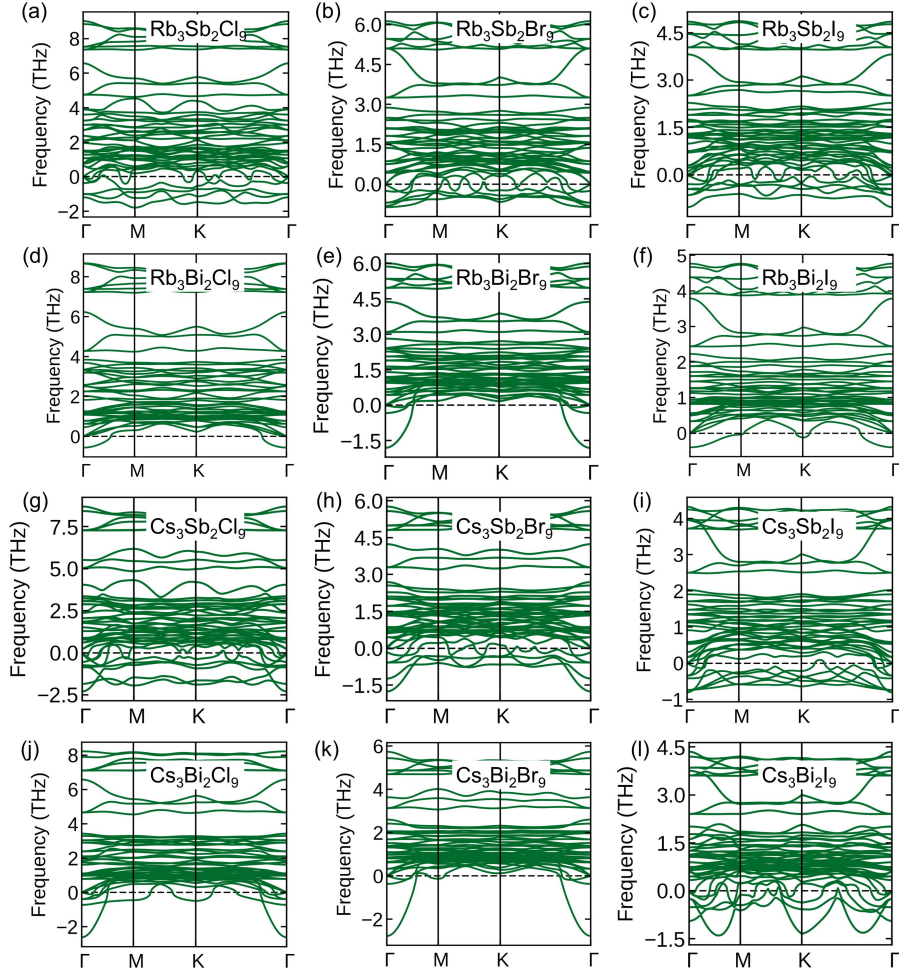
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**Figure S1.** Differences of band gaps ( $\Delta E_g$ ) for monolayer (a)  $A_2BX_4$  and (b)  $A_3B'_2X_9$  obtained by PBE and PBE+SOC methods. The SOC effect is substantial in Pb and Bi compounds but considerably weaker in Sn and Sb systems. Predicted band structures for (c)  $Rb_2SnBr_4$ , (d)  $Cs_2PbI_4$ , and (e)  $Cs_3Bi_2I_9$  by employing the HSE06+SOC approach. It is obvious that the triple degeneracy at the CBM of  $Rb_2SnBr_4$  is broken when SOC is considered.



**Figure S2.** Phonon structures of monolayer  $A_2BX_4$  based on the finite-displacement method with the  $3 \times 3 \times 1$  supercells.



**Figure S3.** Phonon structures of monolayer  $A_3B'_2X_9$  based on the finite-displacement method with the  $3 \times 3 \times 1$  supercells.

**Table S1.** SCAPS simulation parameters for the TCO, various ETLs and HTLs, and the  $\text{Rb}_2\text{SnBr}_2$  absorber, including thickness (L), band gap ( $E_g$ ), electron affinity ( $\chi$ ), dielectric permittivity ( $\epsilon$ ), conduction band effective density of states ( $N_C$ ), valence band effective density of states ( $N_V$ ), electron mobility ( $\mu_n$ ), hole mobility ( $\mu_p$ ), and defect density ( $n_t$ ).

parameters	TCO		ETL			absorber		HTL		
	FTO	PCBM	TiO <sub>2</sub>	SnO <sub>2</sub>	ZnO	Rb <sub>2</sub> SnBr <sub>4</sub>	PATT	PEDOT:PSS	Spiro-MeOTAD	Cu <sub>2</sub> O
L (nm)	500	300	30	100	50	500	100	50	200	50
$E_g$ (eV)	3.6	1.9	3.2	3.6	3.3	1.34	3.3	1.6	3	2.2
$\chi$ (eV)	4	4.2	4	4	4	3.99	2.3	3.4	2.2	3.4
$\epsilon$	9	4	9	9	9	9.95	3	3	3	7.5
$N_C$ (cm <sup>-3</sup> )	$2.2 \times 10^{18}$	$2.5 \times 10^{21}$	$2 \times 10^{18}$	$2.2 \times 10^{18}$	$3.7 \times 10^{18}$	$1.48 \times 10^{19}$	$2.2 \times 10^{19}$	$2.2 \times 10^8$	$2.2 \times 10^{18}$	$2.8 \times 10^{19}$
$N_V$ (cm <sup>-3</sup> )	$1.8 \times 10^{19}$	$2.5 \times 10^{21}$	$1.8 \times 10^{19}$	$1.8 \times 10^{19}$	$1.8 \times 10^{19}$	$1.39 \times 10^{19}$	$1.8 \times 10^{19}$	$1.8 \times 10^{19}$	$1.8 \times 10^{19}$	$1 \times 10^{19}$
$\mu_n$ (cm <sup>2</sup> ·V <sup>-1</sup> ·s <sup>-1</sup> )	20	0.2	20	100	100	4.5	$2 \times 10^{-3}$	$4.5 \times 10^{-2}$	$2.1 \times 10^{-3}$	12
$\mu_p$ (cm <sup>2</sup> ·V <sup>-1</sup> ·s <sup>-1</sup> )	10	0.2	10	25	25	1.5	$4 \times 10^{-3}$	$4.5 \times 10^{-2}$	$2.16 \times 10^{-3}$	2.8
$n_t$ (cm <sup>-3</sup> )	$1 \times 10^{15}$	$1 \times 10^{15}$	$1 \times 10^{15}$	$1 \times 10^{15}$	$1 \times 10^{15}$	$1 \times 10^{15}$	$1 \times 10^{15}$	$1 \times 10^{15}$	$1 \times 10^{15}$	$1 \times 10^{15}$