Supporting Information

Stable, Color-Tunable 2D SCN-Based Perovskites: Revealing the Critical Influence of Asymmetric Pseudo-Halide on Constituent Ions

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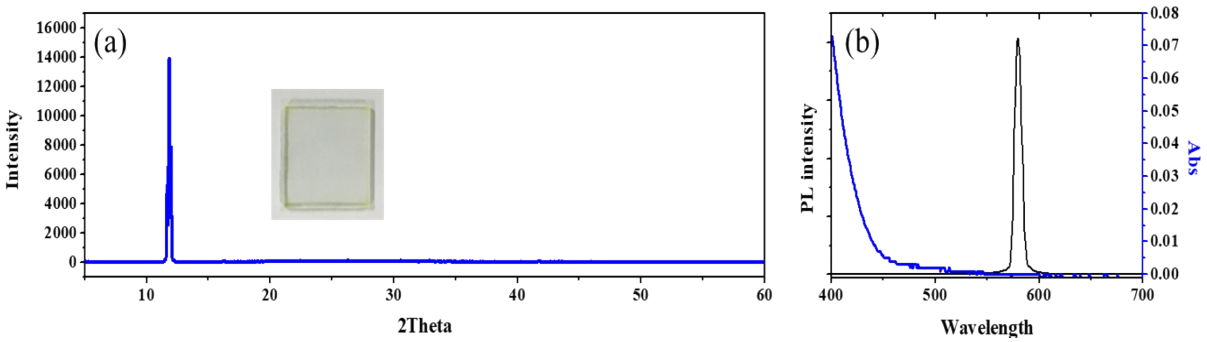

Figure S1. (a) The XRD pattern and (b) UV-vis absorption and PL spectrum of the as-cast film made from the precursor solution containing FAI and $Pb(SCN)_2$ with the aim to form 2D $FA_2PbI_2(SCN)_2$. Insert is the real picture of the as-cast film, displaying matt white color. The result indicates the failure in forming 2D $FA_2Pb(SCN)_2I_2$, as compared to the results of $MA_2Pb(SCN)_2I_2$ and $Cs_2Pb(SCN)_2I_2$ (Figure 1) prepared by same deposition technique.

Table S1. The optimal unit cell parameters and energy of the studied SCN-based Cs-perovskites.

Systems	a	b	c	volume	Total energy
	Å	Å	Å	Å^3	eV
Cs-I	18.64	6.39	6.38	760.61	-114.85
Cs-bb	18.64	6.14	6.38	729.90	-116.30
Cs-bc	18.69	6.32	6.20	733.33	-116.27
Cs-cb	18.67	6.33	6.20	732.73	-116.27
Cs-cc	18.91	6.41	6.04	732.04	-116.34
Cs-Br	18.62	6.15	6.06	693.86	-117.55

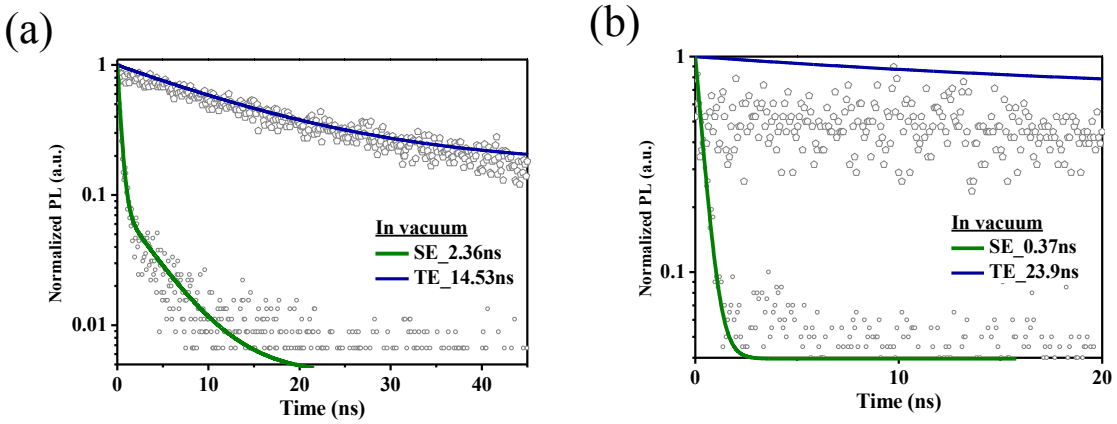

Figure S2. TRPL spectra of the (a) $Cs_2Pb(SCN)_2I_2$ and (b) $(MA)_2Pb(SCN)_2I_2$ films measured in vacuum.

Table S2. TRPL carrier lifetime for the studied 2D perovskite films.

		$Cs_2Pb_2(SCN)_2I_2$	$(MA)_2Pb_2(SCN)_2I_2$
In vacuum	**SE**	2.36 ns	0.368 ns
	TE	14.53 ns	23.9 ns
In ambient air	**SE**	3.97 ns	0.359 ns
	TE	12.37 ns	17.09 ns

Table S3. Fitting parameters for the TRPL dynamics for the studied 2D perovskite films.

	$Cs_2Pb_2(SCN)_2I_2$				$(MA)_2Pb_2(SCN)_2I_2$			
	In vacuum		**In air**		**In vacuum**		**In air**	
	SE	**TE**	**SE**	**TE**	**SE**	**TE**	**SE**	**TE**
A1	31.75	163	41.55	152.55	187	7.29	177	28.5
τ_1 (ns)	4.19	14.53	6.34	12.38	0.369	23.9	0.359	5.45
A2	358	-	417	-	-	-	-	37.1
τ_2 (ns)	0.34	-	0.42	-	-	-	-	20.75

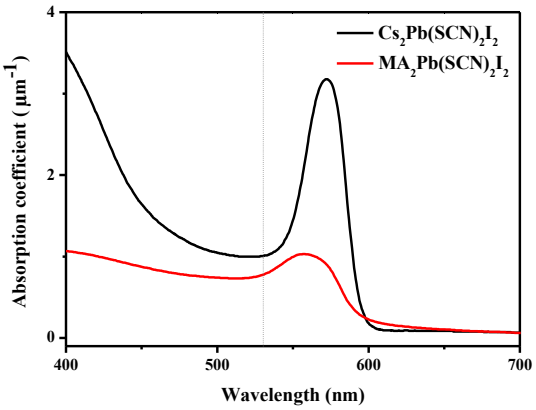

Figure S3. Absorption coefficients of $Cs_2Pb(SCN)_2I_2$ and $MA_2Pb(SCN)_2I_2$.

Table S4. Enthalpies of decomposition of each system calculated from Eqn (1), (2), and (3).

	Compound	CS-I	CS-bb	CS-bc	CS-cb	CS-cc	CS-Br
Enthalpy of decomposition (eV)	**Eqn 1**	12.69	13.32	13.37	13.34	13.75	13.52
	Eqn 2	3.52	3.84	3.81	3.79	3.76	3.94
	Eqn 3	12.98	13.66	13.63	13.57	13.76	14.00

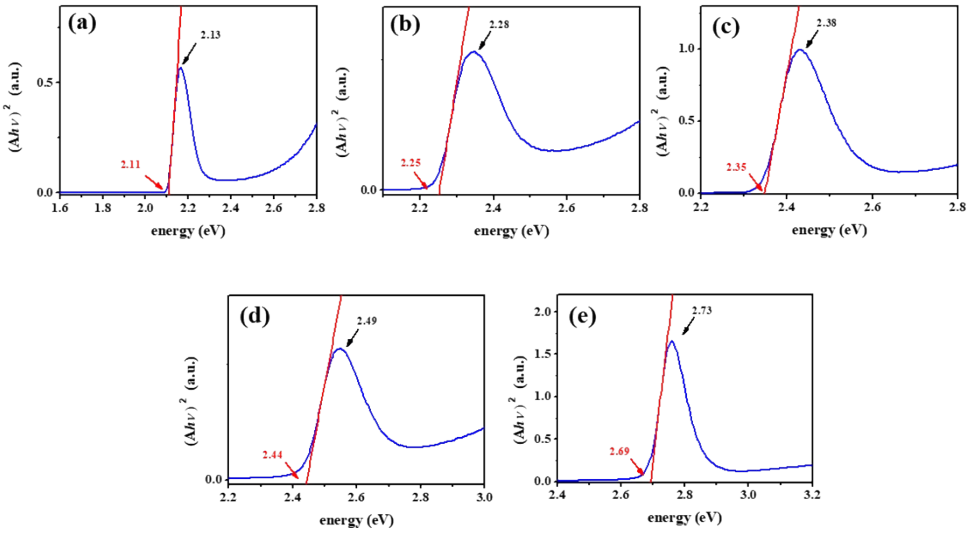

Figure S4. The Tauc plots of the studied $Cs_2Pb(SCN)_2(I_{1-x}Br_x)_2$ films: (a) x = 0, (b) x = 1/3, (c) x = 1/2, (d) x = 2/3, and (e) x = 1.

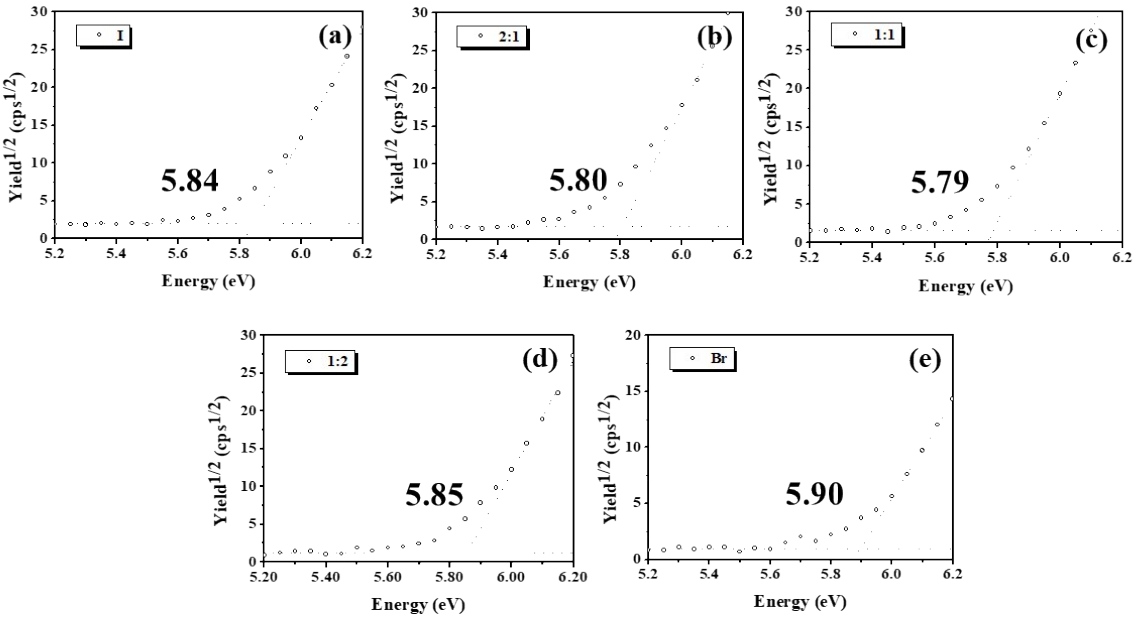

Figure S5. The HOMO level of the studied $Cs_2Pb(SCN)_2(I_{1-x}Br_x)_2$ films measured by photoelectron spectroscopy: (a) x = 0, (b) x = 1/3, (c) x = 1/2, (d) x = 2/3, and (e) x = 1.

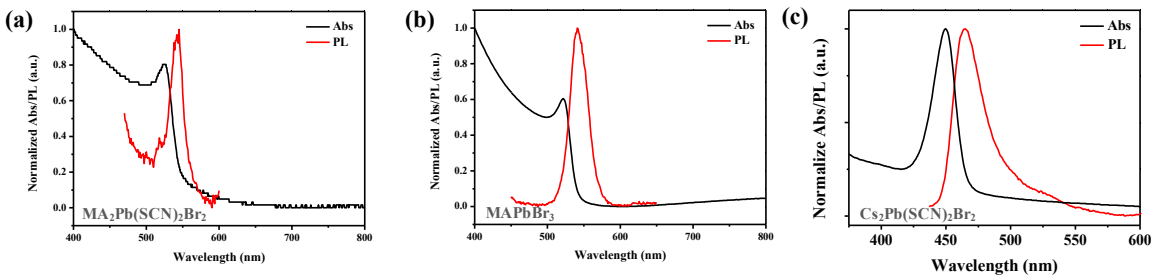

Figure S6. The UV-vis absorption and PL spectra of (a) the prepared $(MA)_2Pb(SCN)_2Br_2$, (b) $MAPbBr_3$, and (c) $Cs_2Pb(SCN)_2Br_2$ films.

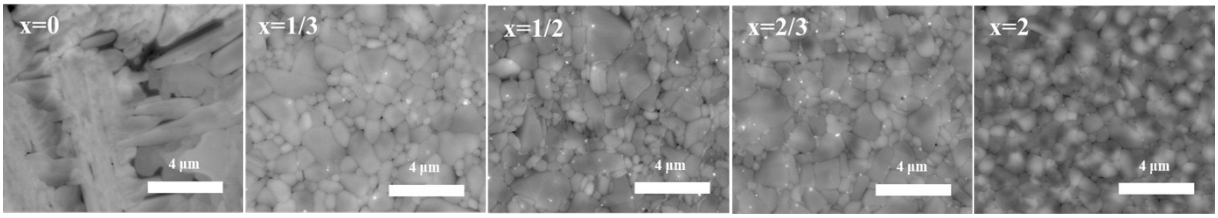

Figure S7. The SEM images (10000x) of the studied $Cs_2Pb(SCN)_2(I_{1-x}Br_x)_2$ films.