Supporting Information

Inorganic Perovskite Engineering Through Incorporation of Carboxylic Acid Containing Ligand for Performance Enhancement in Perovskite Light-Emitting Diodes

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1 Experimental

1.1 Materials: Cesium bromide (CsBr), lead (II) bromide ($PbBr_2$), poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, 4083), 1,3,5-tris(N-phenylbenzimidazol-2-yl)benzene (TPBI) and phenmethylammonium bromide (PMABr) were used as purchased from Xi'an Polymer Light Technology Corp., 4-(Aminomethyl)benzoic acid (97%) was purchased from Aladdin. Hydrobromic acid (HBr) was purchased from Fuchen (Tianjin) Chemical Reagents Co. Ltd., dimethyl sulfoxide (DMSO) was purchased from J&K Scientific Ltd., tetrahydrofuran (THF), ethanol and toluene were purchased from Guangzhou Chemical Reagent Factory. All materials were used without further purification.

1.2 Device Fabrication and Characterization. Indium tin oxide (ITO)-coated glass substrates were ultrasonically cleaned prior to O_2 plasma treatment. Subsequently, PEDOT:PSS was spin-coated on the treated ITO at a speed of 3000 rpm for 60 s, following by thermal annealing for 30 min at 150 ℃ under atmospheric environment, then the substrates were treated with O_2 plasma again for 15 s to make the surface more hydrophilic. After that, the substrates were transferred into a glovebox and the

perovskite precursor solutions were spin-coated onto the PEDOT:PSS films at 5500 rpm for 50 s and annealed at 65 ℃ for 20 min. Then, the perovskite-coated substrates were transferred to a thermal evaporation chamber, wherein organic and metal cathode layers were sequentially deposited under vacuum pressure of 3×10^{-4} Pa, leading to an active area of 9 mm^2. The current density−voltage−luminance characteristics and electroluminescence (EL) spectra of PeLEDs were measured using a Keithley 2400 source combined with a Photo Research PR735 spectrometer under room temperature, while external quantum efficiency (EQE) was calculated based on a computer program.

2. Supplementary tables and figures

Table S1 TRPL characteristics of perovskite films.

Perovskite	λ (nm)	B_1 (%)	τ_1 (ns)	B_2 (%)	τ_2 (ns)	B_3 (%)	τ_3 (ns)	τ_{avg} (ns)
$CsPbBr_3$	520	4.74	6.50	6.12	201.64	89.14	0.33	12.94
x=1.0	517	38.57	0.82	40.52	8.99	20.91	49.08	14.22
x=1.5	516	37.18	6.97	12.27	153.28	50.55	1.54	22.17
x=2.0	514	27.30	17.36	34.55	147.23	38.15	0.66	55.86
x=2.5	507	31.03	11.53	15.10	186.41	53.87	1.88	32.74
$(PMA)_2CsPb_2Br_7$	466	14.14	7.67	10.97	94.53	74.89	0.81	12.06
	517	20.26	8.97	27.99	120.39	51.75	0.92	35.99

The TRPL profiles were fitted by a three-exponential decay model as $I(\tau)=B_1\cdot\exp(-t/\tau_1) + B_2\cdot\exp(-t/\tau_2) + B_3\cdot\exp(-t/\tau_3)$, wherein τ_1, τ_2, τ_3 are the lifetimes of the first, second and third components, respectively; B_1, B_2, B_3 are the proportions of the first, second and third lifetime components, respectively, and $B_1 + B_2 + B_3 = 1$. The average lifetime (τ_{avg}) was calculated by $\tau_{avg}=(B_1\cdot\tau_1 + B_2\cdot\tau_2 + B_3\cdot\tau_3)/(B_1 + B_2 + B_3)$.

Table S2 Electrical performance of the PeLEDs based on $CsPbBr_3$, $(HOOC\text{-}PMA)_xCsPb_2Br_{(5+x)}$ (x=1.0, 1.5, 2.0, 2.5), and $(PMA)_2CsPb_2Br_7$ perovskites.

Device	λ_{EL}[a] (nm)	L_{max}[b] (cd m^{-2})	CE_{max}[c] (cd A^{-1})	EQE_{max}[d] (%)	FWHM[e] (nm)	CIE[f] (x, y)
$CsPbBr_3$	520	205	0.08	0.02	22	0.136,0.730
x=1.0	515	949	0.61	0.12	25	0.090, 0.716
x=1.5	514	1641	0.93	0.29	25	0.101, 0.725
x=2.0	514	8692	5.27	1.61	25	0.096, 0.723
x=2.5	508	2985	1.29	0.40	26	0.074, 0.620
$(PMA)_2CsPb_2Br_7$	515	1765	0.93	0.33	23	0.107,0.732

[a]EL peak wavelength, [b]maximum luminance, [c]maximum current efficiency, [d]maximum external quantum efficiency, [e]full width at half maximum, [f]1931 CIE coordinates at a voltage of 6 V.

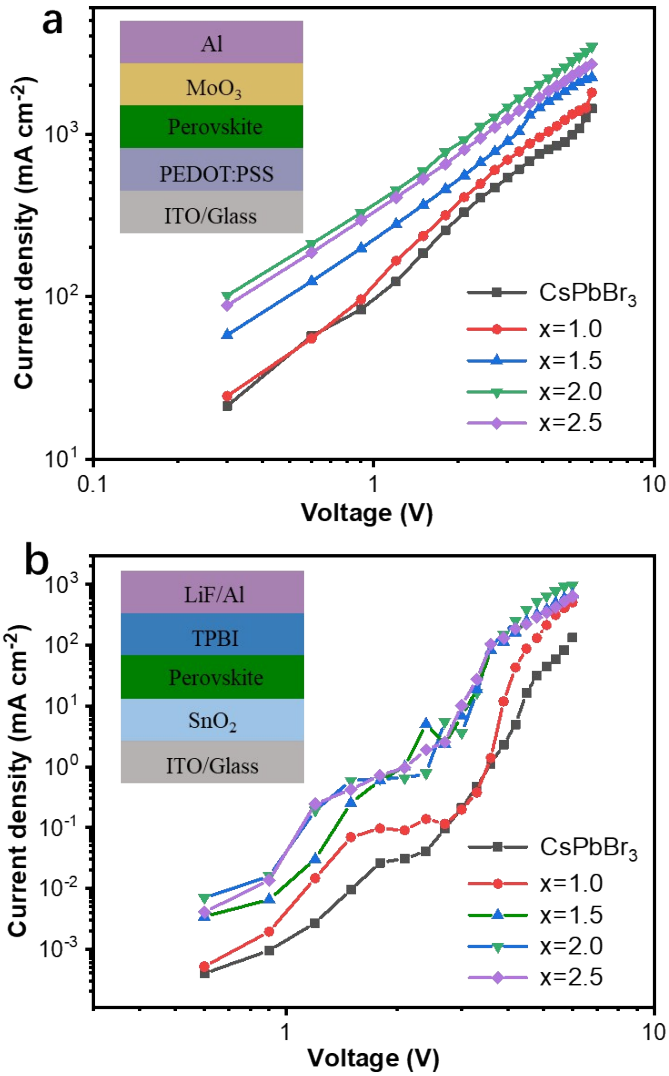

Figure S1. Current density–voltage curves for (a) hole-only and (b) electron-only devices. Inset: device configurations of the hole-only and electron-only devices.

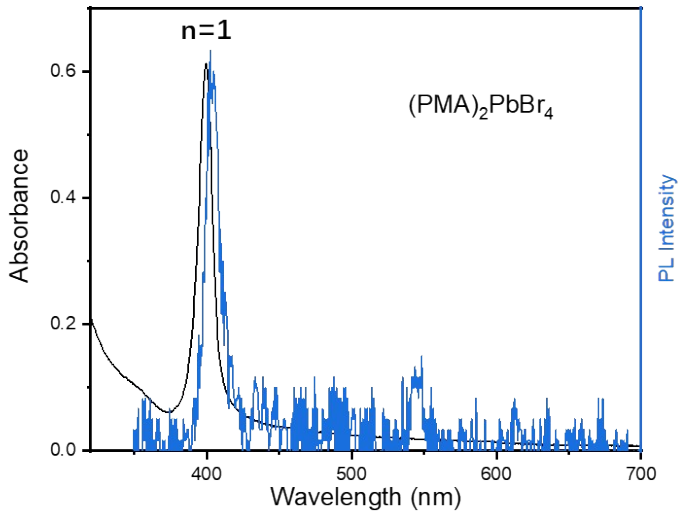

Figure S2. UV-vis absorption and PL spectra of $(PMA)_2PbBr_4$ film.

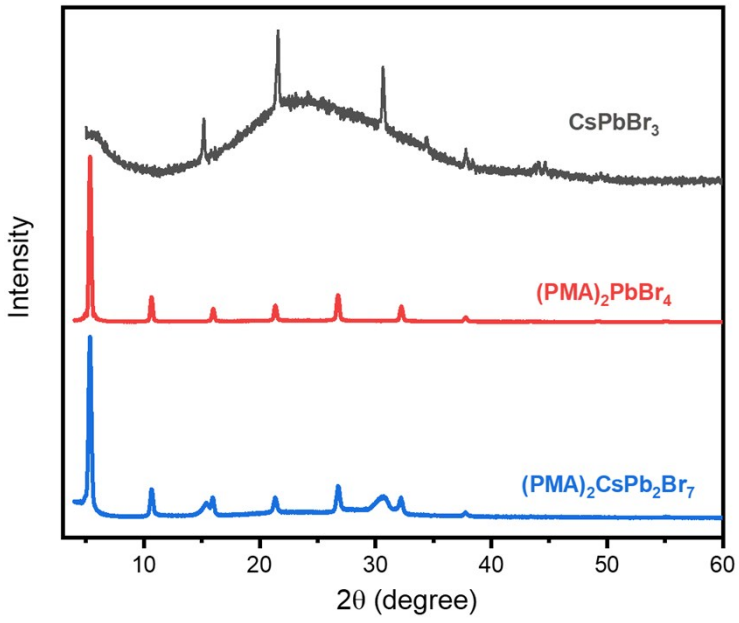

Figure S3. XRD profiles of $CsPbBr_3$, $(PMA)_2PbBr_4$ and $(PMA)_2CsPb_2Br_7$ perovskites.