

## Supporting information

### **Modification of mixed-halide quasi-2D perovskites by aminophylline towards efficient and spectrally stable blue light-emitting diodes with low efficiency roll-off**

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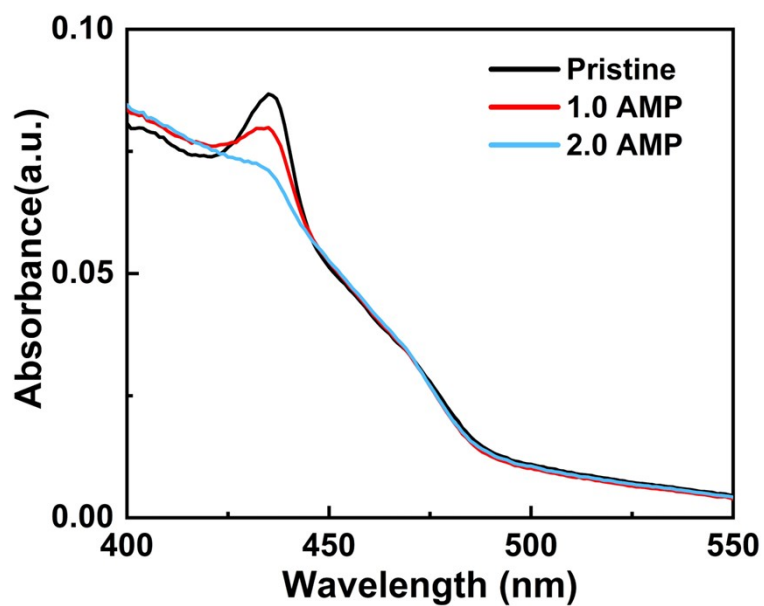


Figure S1. UV-vis absorption spectra of perovskite films with different AMP concentration (1.0 AMP and 2.0 AMP denoted as the modified perovskite films with  $1.0 \text{ mg mL}^{-1}$  and  $2.0 \text{ mg mL}^{-1}$  AMP in the precursor solutions respectively).

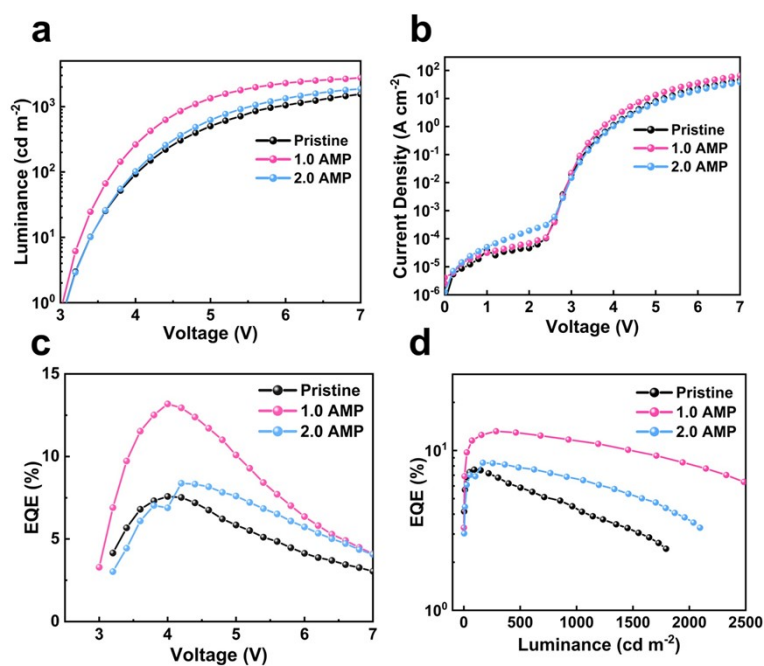


Figure S2. Device performance of PeLEDs using the perovskite films as the EMLs with different AMP concentration. (a) Current density-voltage characteristics. (b) Luminance-voltage characteristics. (c) EQE-voltage characteristics. (d) EQE-luminance characteristics.

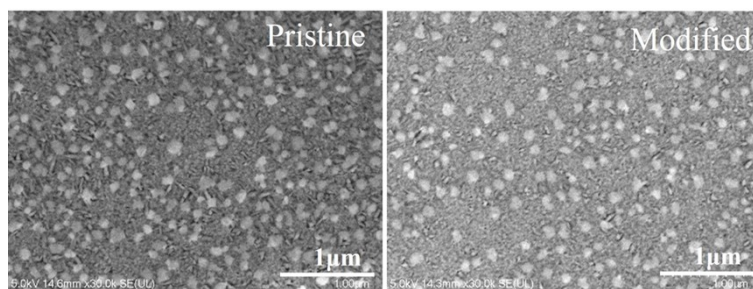


Figure S3. SEM images of pristine and modified perovskite films.

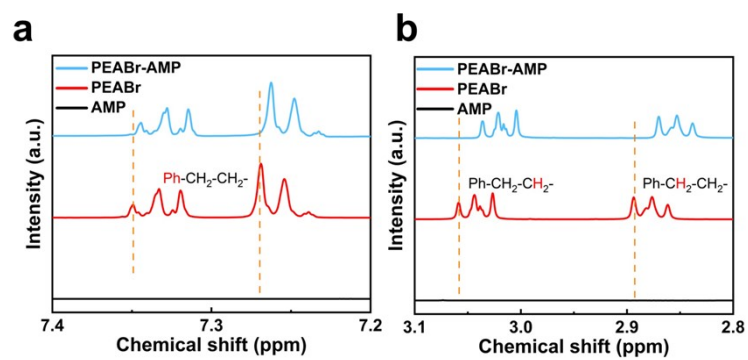


Figure S4.  $^1\text{H}$  NMR characterization of AMP, PEABr and PEABr-AMP.

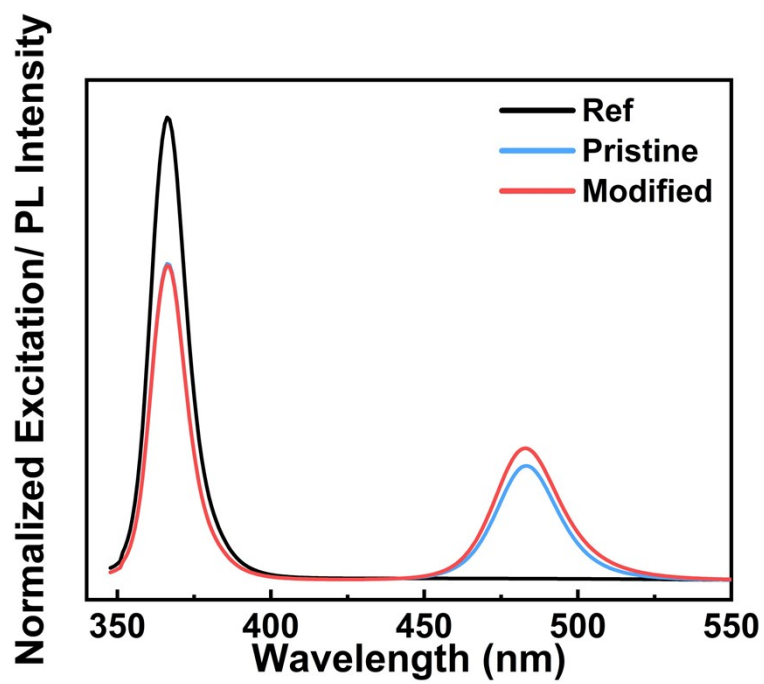


Figure S5. PLQY spectra of pristine and modified perovskite films.

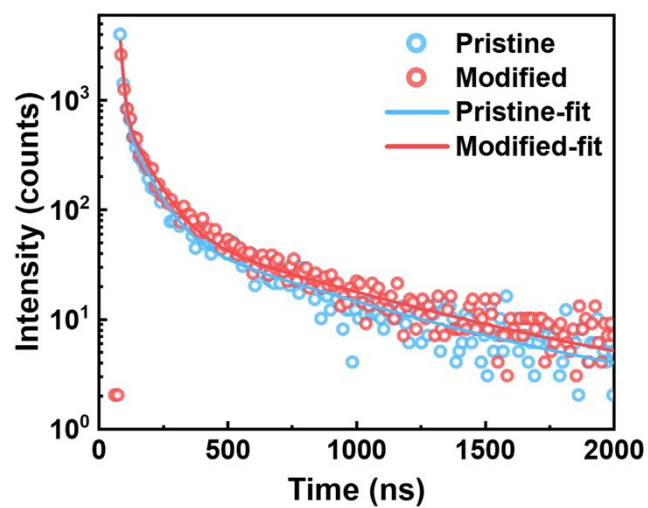


Figure S6. TRPL decay curves of pristine and modified perovskite films.

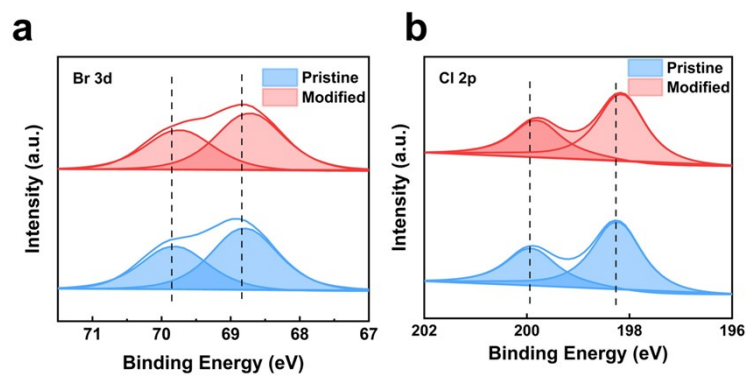


Figure S7. XPS spectra of Br 3d (a) and Cl 2p (b) of pristine and modified perovskite films.



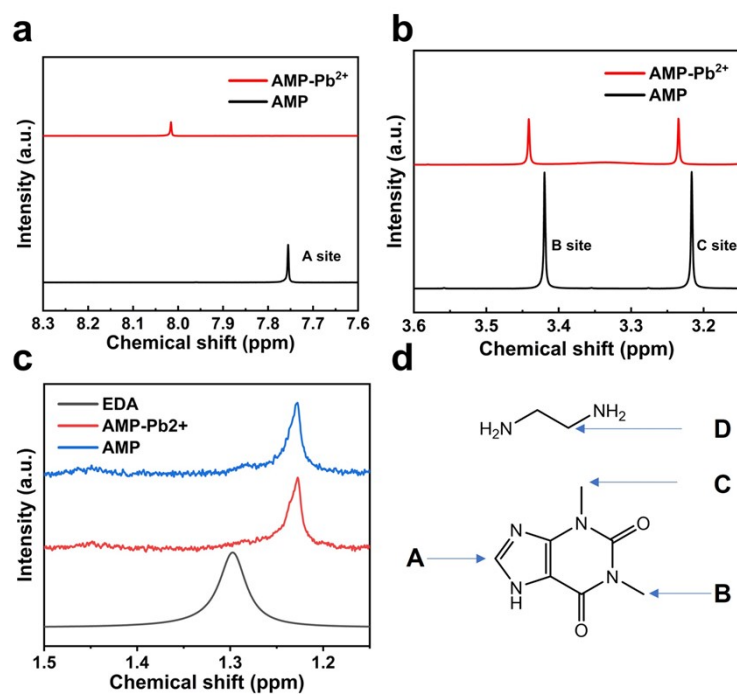


Figure S8. (a-c) <sup>1</sup>H NMR characterization of AMP and AMP-Pb<sup>2+</sup>. Here EDA is Ethylenediamine in AMP. (d) Chemical structure of AMP.

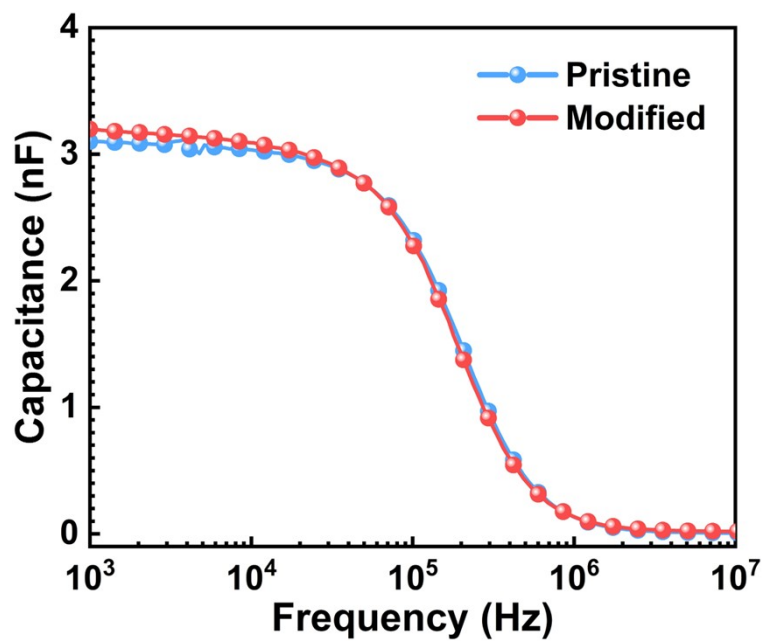


Figure S9. Capacitance-Frequency curves of the devices based on pristine and modified perovskite films.

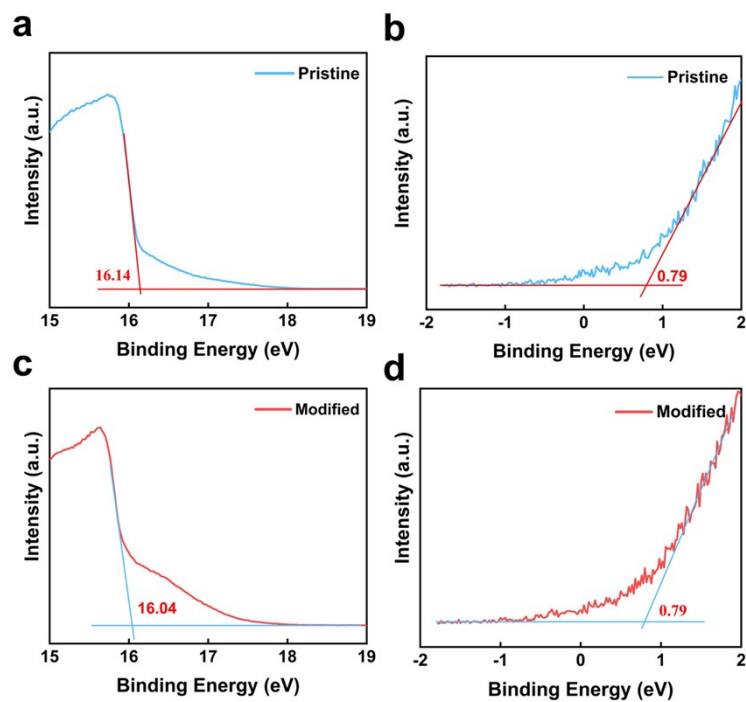


Figure S10. UPS spectra of pristine and modified perovskite films.

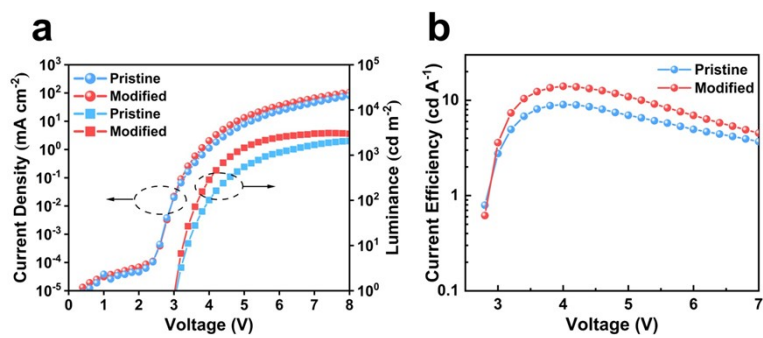


Figure S11. Voltage-dependent current density, luminance and current efficiency of pristine and modified PeLEDs.

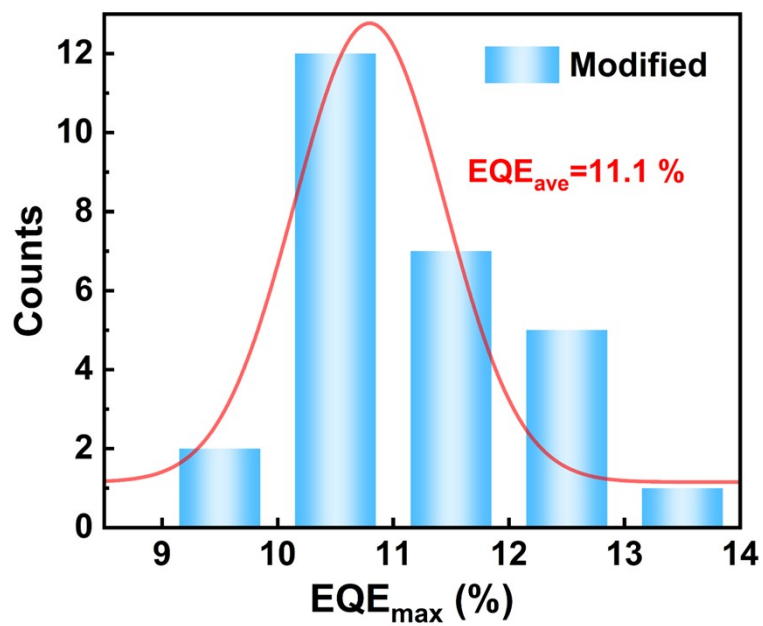


Figure S12. Histogram of maximum EQEs of the devices based on the AMP-modified EMLs.

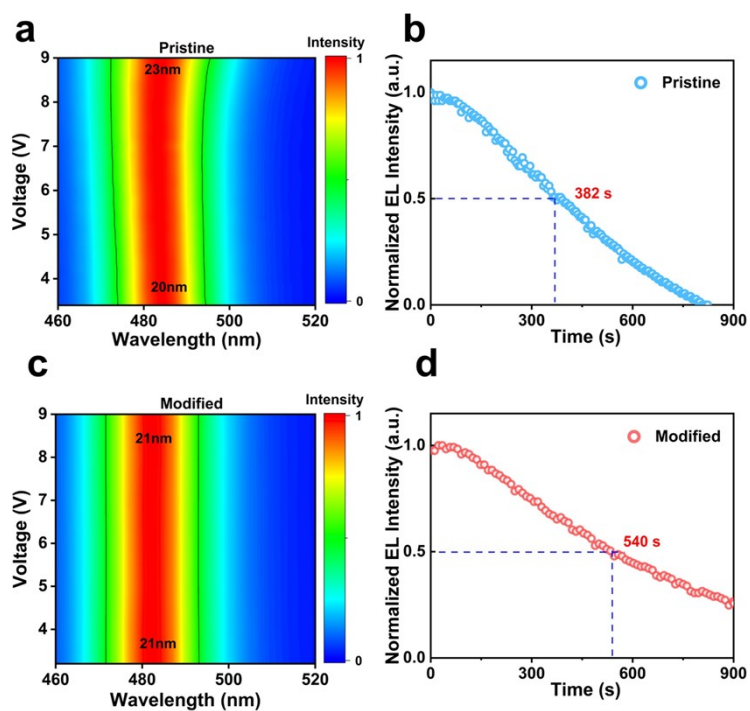


Figure S13. Voltage-dependent EL spectra of pristine (a) and modified (c) PeLEDs. Lifetime of pristine (b) and modified (e) PeLEDs under the initial luminance of  $100 \text{ cd m}^{-2}$ .

**Table S1** Fitting results of TRPL decay curves from the pristine and modified perovskite films. And the calculated average PL lifetimes, radiative and non-radiative recombination rates are also listed for comparison.

Comment	$\tau_1$ (ns)	$A_1$	$\tau_2$ (ns)	$A_2$	$\tau_3$ (ns)	$A_3$	$\tau_{avg}$ (ns)	$K_r$ (ns <sup>-1</sup> )	$K_{nr}$ (ns <sup>-1</sup> )
Pristine	8.58	0.28	72.10	0.40	576.83	0.32	213.8	$2.8 \times 10^{-3}$	$1.9 \times 10^{-3}$
Modified	11.17	0.24	84.91	0.44	623.77	0.31	236.2	$3.1 \times 10^{-3}$	$1.1 \times 10^{-3}$

$$I = \sum A_i e^{-\frac{t}{\tau_i}}, i = 1, 2, 3 \quad (S1)$$

$$\tau_{avg} = \frac{\sum (A_i \tau_i)}{\sum A_i}, i = 1, 2, 3 \quad (S2)$$

$I$  is normalized PL intensity,  $t$  is time,  $\tau_1$ ,  $\tau_2$  and  $\tau_3$  represent decay lifetimes of carrier,  $A_1$ ,  $A_2$  and  $A_3$  are corresponding to the amplitude of decay components and  $A_1 + A_2 + A_3 = 1$ .

The average PL lifetimes of  $\tau_{avg}$  are calculated from equation S2.

$$\frac{1}{\tau_{avg}} = K_r + K_{nr} \quad (S3)$$

$$\frac{1}{\eta_{PLQY}} = \frac{K_r}{K_r + K_{nr}} \quad (S4)$$

The  $K_r$  and  $K_{nr}$  are corresponding to radiative and non-radiative recombination rates, respectively.

**Table S2** Fitting parameters of the decay kinetics at the selected wavelength according to equation S5.

Sample	phase	$\tau_{et}$ (ps)	$\tau_1$ (ps)	$\tau_2$ (ps)	$\tau_3$ (ps)	$A_0$	$A_1$	$A_2$	$A_3$
pristine	n=3	0.07	0.25	1.93	76.28	13.54	-9.20	-0.58	-0.22
	bulk	0.49	0.57	34.10	1001.84	15.33	-12.00	-1.70	-1.04
Modified	n=3	0.10	0.21	1.71	54.48	8.82	-8.14	-0.83	-0.04
	bulk	0.22	0.21	48.07	1196.68	25.67	-0.90	-1.88	-21.56

$$M(t; A, \tau) = \frac{\sqrt{4 \ln(2)}}{\sqrt{\pi} FWHM} e^{-\frac{4 \ln(2) \cdot (t - D_0)^2}{FWHM^2}} \otimes H(t - D_0) \cdot \left( A_0 e^{-\frac{t - D_0}{\tau_{et}}} + A_1 e^{-\frac{t - D_0}{\tau_1}} + A_2 e^{-\frac{t - D_0}{\tau_2}} + A_3 e^{-\frac{t - D_0}{\tau_3}} \right) \quad (S5)$$



**Table S3** Performance of reported blue PeLEDs with EQE<sub>max</sub> exceeding 10%

Number	$\lambda_{\text{EL}}$ (nm)	EQE <sub>max</sub> (%)	EQE <sub>1000</sub> (%)	EQE <sub>1000</sub> /EQE <sub>max</sub> (%)	Ref.
1	485	14.4	2.1	14.7	1
2	484	11.5	3.2	28.1	2
3	475	14.2	4.1	28.6	3
4	487	20.4	6.0	29.6	4
5	484	21.4	7.4	34.5	5
6	486	16.2	5.8	36.0	6
7	483	11.3	4.6	41.0	7
8	488	12.1	6.1	50.6	8
9	477	11.0	5.7	52.1	9
10	493	16.5	9.8	59.4	10
11	489	12.5	8.7	69.4	11
12	486	11.2	7.8	69.6	12
13	484	13.2	11.5	87.1	this work

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