Supplementary Information (SI) for Nanoscale.

Electronic supplementary information

Pure Red Emission with Spectra-Stable in Full Iodine-based Quasi-2D Perovskite Films by Controlling Phase Distribution

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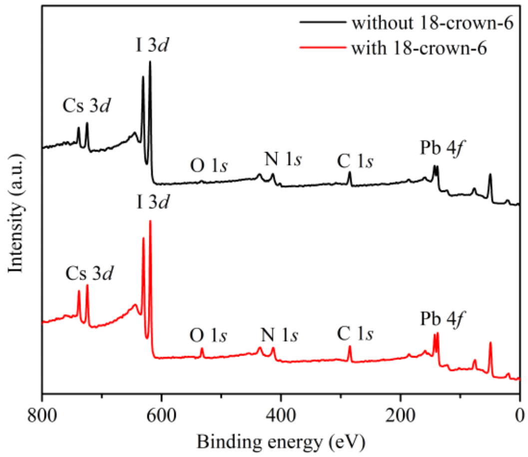

Fig. S1 XPS spectra of quasi-2D perovskite films without and with 18-crown-6.

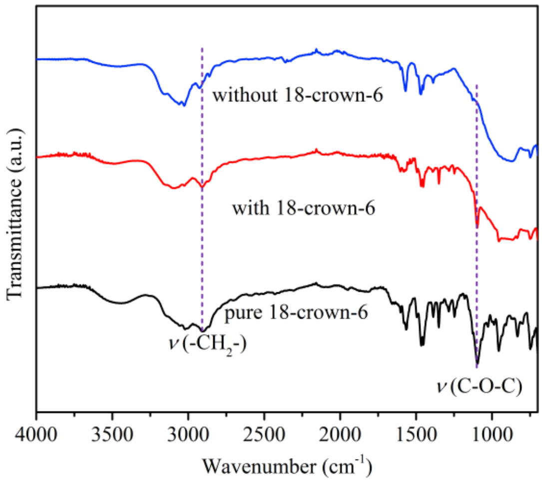

Fig. S2 FTIR spectra of quasi-2D perovskite films without and with 18-crown-6 in comparison with the pure 18-crown-6.

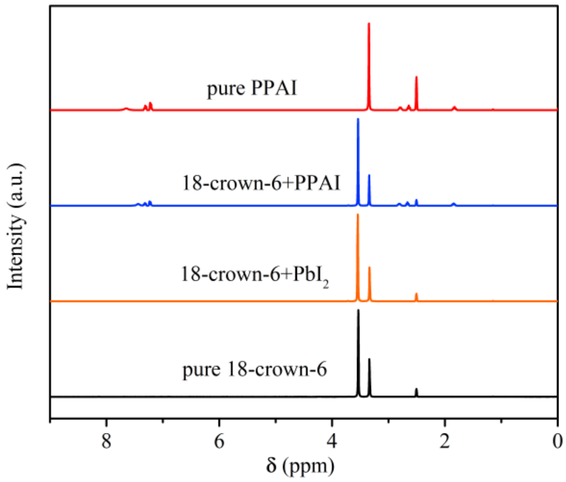

Fig. S3 ^{1}H NMR spectra of pure 18-crown-6, 18-crown-6 with PbI_2, 18-crown-6 with PPAI and pure PPAI in deuterated DMSO.

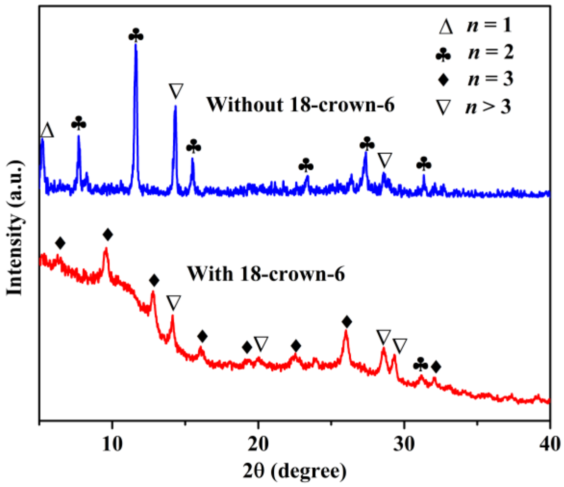

Fig. S4 GIXRD spectra of quasi-2D perovskite films without and with 18-crown-6. The film without 18-crown-6 exhibited typical diffraction peaks with 2θ periodicity of 3.9°, corresponding to (0k0) reflections. We calculated the d-spacing was about 2.26 nm, which was similar to $BA_2MAPb_2I_7$.[1] Thus, we attributed above diffraction peaks (0k0) to *n* = 2 perovskite. Similarly, the film with 18-crown-6 exhibited typical diffractions with 2θ periodicity of 3.2°, corresponding to (0k0) reflections. We calculated the d-spacing was about 2.76 nm, similar to $BA_2MA_2Pb_3I_{10}$.[2] Thus, these diffraction peaks could be ascribed to *n* = 3 perovskite crystal planes.

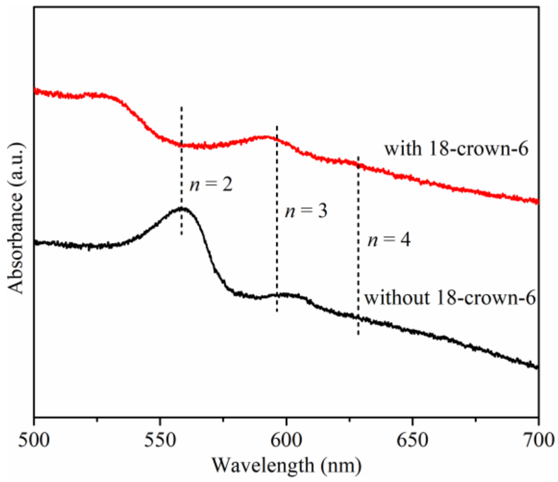

Fig. S5 UV-vis absorption spectra of perovskite films without and with adding 18-crown-6 additions.

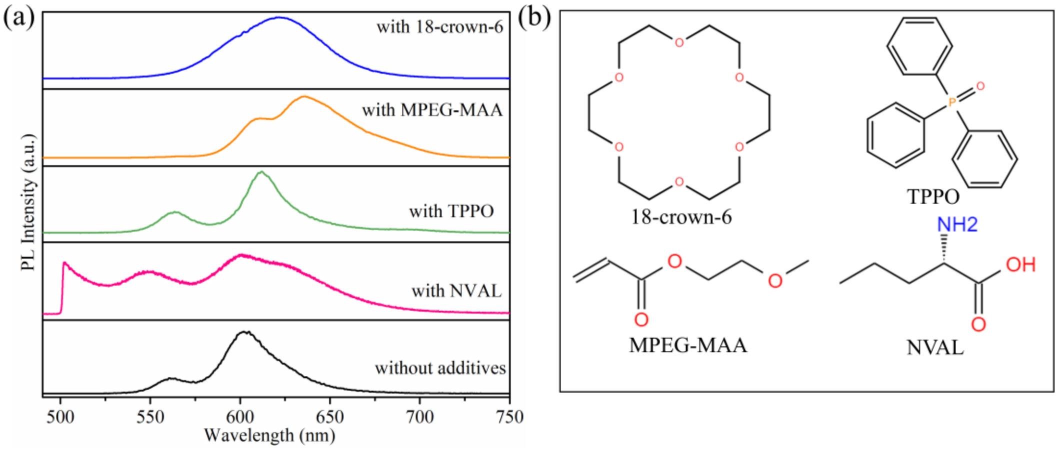

Fig. S6 (a) Log PL spectra of perovskite films without and with various types of additives under 488 nm excitation. (b) The molecular structure of various additives including 18-crown-6, poly(ethylene glycol)methyl ether acrylate (MPEG-MAA), triphenylphosphine oxide (TPPO) and L-Norvaline (NVAL).

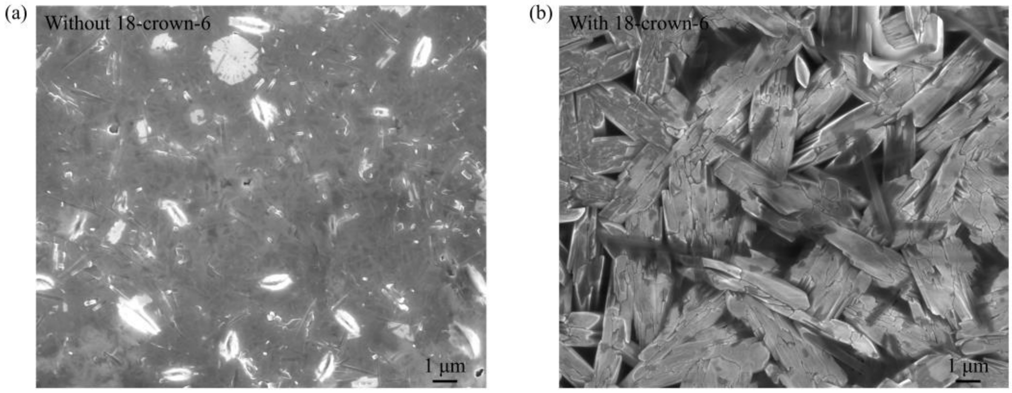

Fig. S7 Top-view SEM images of perovskite films without (a) and with (b) 18-crown-6 additives.

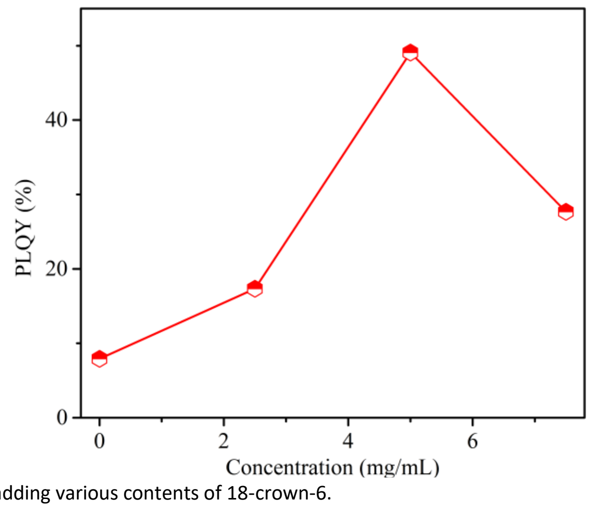

Fig. S8 PLQY of perovskite films with adding various contents of 18-crown-6.

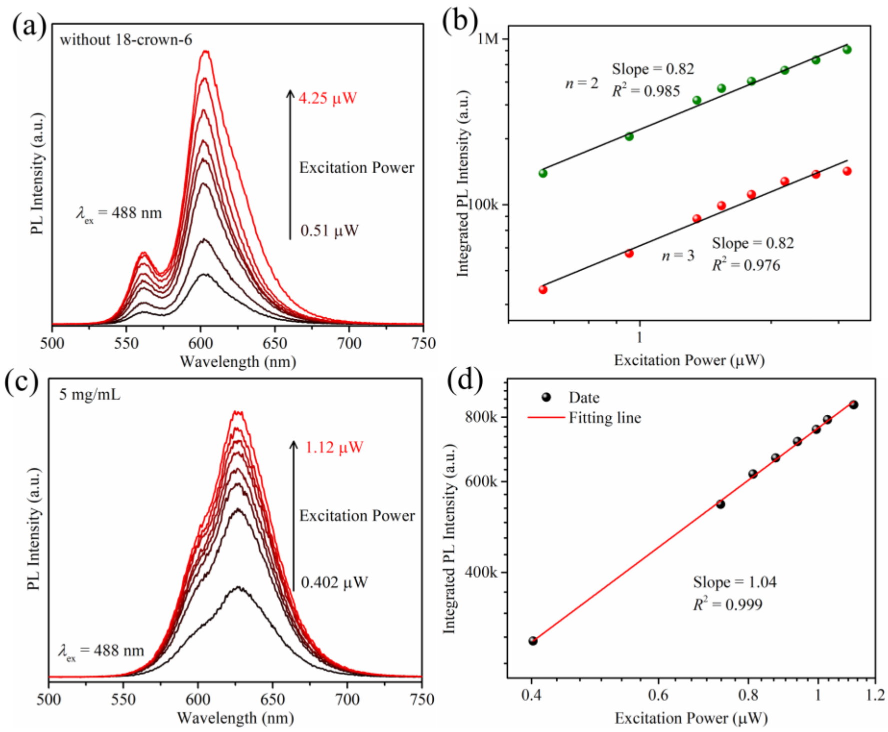

Fig. S9 Perovskite films with adding various contents of 18-crown-6: (a) (d) power dependent PL spectra, and (b) (c) PL intensity of emission peak under varying excitation power.

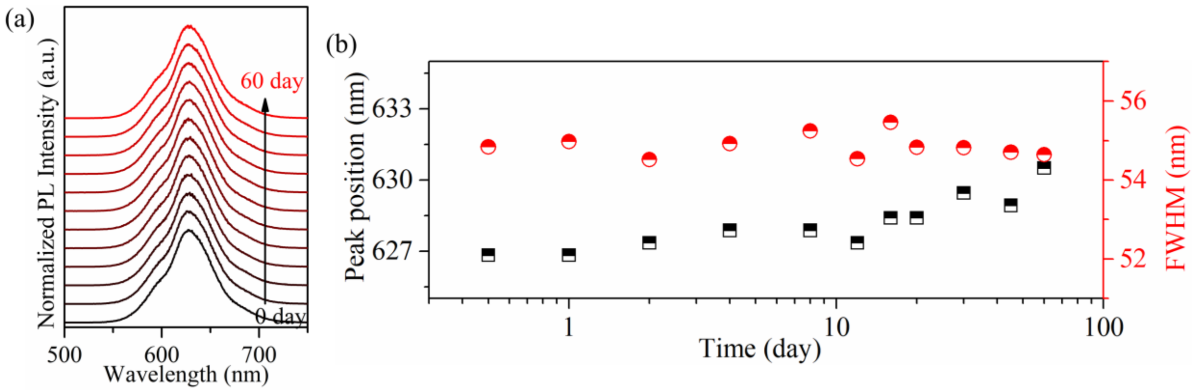

Fig. S10 (a) PL spectra and (b) corresponding the FWHM and peak position difference of quasi-2D perovskite films with 18-crown-6 after exposure to air for 0-60 days.

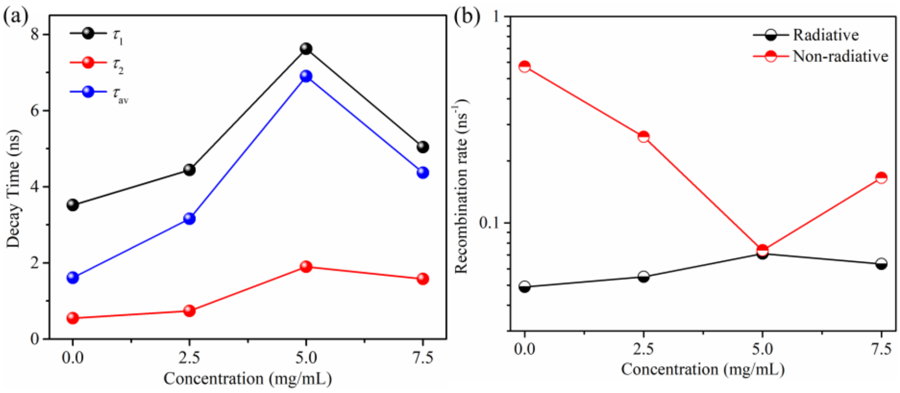

Fig. S11 (a) Slow decay lifetime τ_1, fast decay lifetime τ_2 and average decay lifetime τ_{av} (b) radiative and non-radiative recombination rate of perovskite films without and with adding various concentration of 18-crown-6 additions.

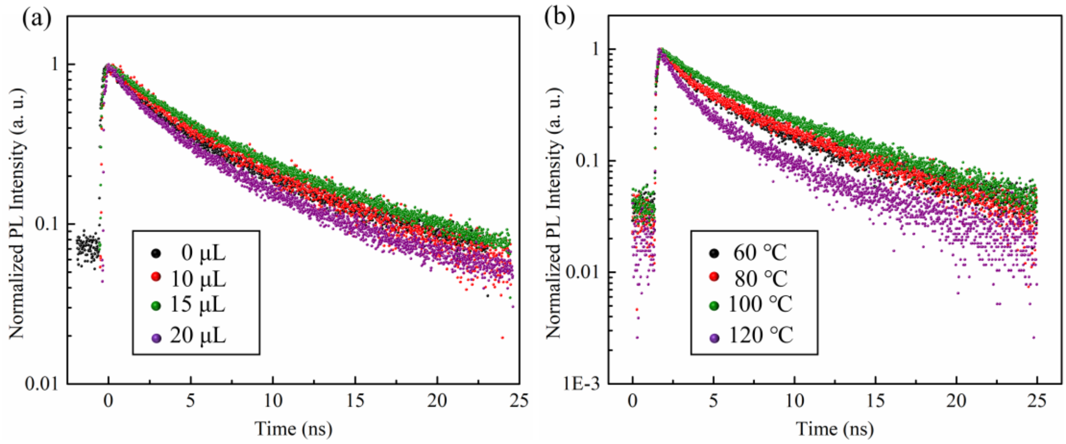

Fig. S12 TRPL decay curves of quasi-2D perovskite films with (a) various contents of CB and (b) different annealing temperature under 405 nm irradiation.

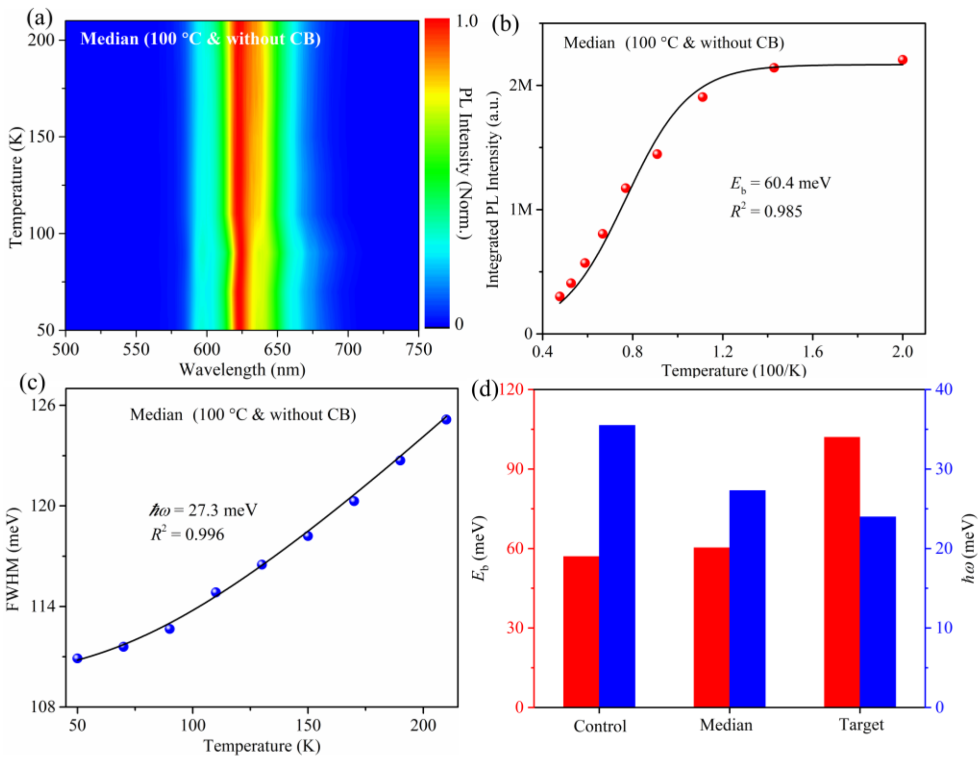

Fig. S13 Normalized temperature-dependent PL spectra of (a) median perovskite film (with annealing at 100 ℃ and without CB). (b) Relevant integration of PL intensity of median film and fitting curve for E_b. (c) FWHM of pure red emission peak as a function of temperature of median film. (d) Comparison of exciton binding energy (E_b) and longitudinal optical phonon energy ($\hbar\omega$) in different samples.

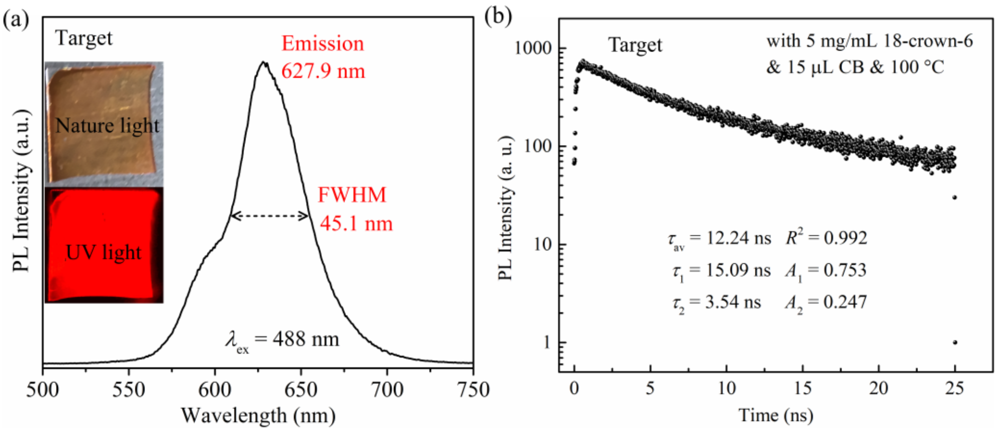

Fig. S14 (a) PL and (b) TRPL decay curve of target quasi-2D perovskite films under 405 nm irradiation.

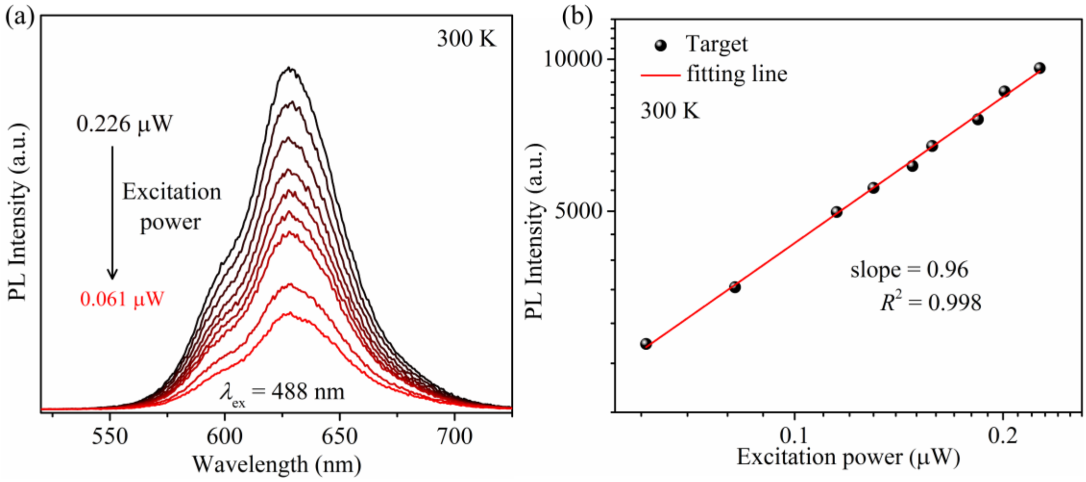

Fig. S15 (a) Power dependent PL spectra of optimized quasi-2D perovskite films under varying excitation power. (b) PL intensity of emission peak under varying excitation power.

Note S1

All PL decay curves could be well fitted by a bi-exponential decay function: $I(t) = A_1\exp(-t/\tau_1)+A_2\exp(-t/\tau_2)$, where A_1 and A_2 are constant, τ_1 and τ_2 are the slow decay lifetime of corresponding radiation recombination and the fast decay lifetime of corresponding non-radiation recombination. [3] The average lifetime is calculated using the equation: $\tau_{av} = (A_1\tau_1^2+ A_2\tau_2^2)/(A_1\tau_1+ A_2\tau_2)$. Based on results of PLQY and decay lifetime, we calculated the radiative and non-radiative recombination rate (k_r, k_{nr}) of films using the equation of PLQY = $k_r /(k_r + k_{nr})$, $\tau_{av} = 1/(k_r + k_{nr})$.

Note S2

All temperature-dependent integration of PL intensity of target and control films curves could be well fitted by Arrhenius equation: $I(T) = I_0/(1+A\exp(-E_b/kT))$, in which $I(T)$ and I_0 are the integrated PL intensity under the temperature of T K and 0 K, k is the Boltzmann constant, and E_b is the exciton binding energy.[4]

Note S3

The broadening emission band with increasing temperature could be fitted based on the independent Boson model: $\Gamma(T) = \Gamma_0+\sigma T+\Gamma_{op}/(\exp(\hbar\omega/kT)-1)$, where Γ_0 represents the temperature-independent inhomogeneous broadening, the other terms represent the homogeneous broadening, attributing to the exciton-acoustic phonon interaction (σ) and the exciton-longitudinal optical phonon interaction (Γ_{op}).[5]

Table S1. Progress of pure red full iodine-based quasi-2D perovskite films.

Composition	Difference of pure red peak position (nm)	FWHM (nm)	Reference
$(PBA_xMBZA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$ (x = 1)	4	49	[6]
$(PBA_xMBZA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$ (x = 0.75)	2	49	[6]
$(PBA_xMBZA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$ (x = 0.5)	9	47	[6]
$(PBA_xMBZA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$ (x = 0.25)	20	42	[6]
$(PBA_xPOEA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$($x$ = 0.75)	1	46	[6]
$(PBA_xPOEA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$($x$ = 0.5)	6	45	[6]
$(PBA_xPOEA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$($x$ = 0.25)	10	42	[6]
$(PBA_xPOEA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$($x$ = 0)	12	41	[6]
$(POEA_xMBZA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$($x$ = 0.75)	15	38	[6]
$(POEA_xMBZA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$($x$ = 0.5)	20	38	[6]
$(PEA_{0.5}NMA_{0.5})_2CsPb_2I_7$	5	46	[7]
$PEA_2(Cs_{0.3}MA_{0.7})2(Pb_{1-x}Zn_x)_3I_{10}$ (x = 0.45)	7	61	[8]
$BA_2Cs_{n-1}Pb_nI_{3n+1}$(No.2)	4	55	[9]
$PBA_2Cs_{n-1}Pb_nI_{3n+1}$ (PBA_1PI)	13	48	[10]
$PPA_2Cs_{n-1}Pb_nI_{3n+1}$	20	46	[11]
$POEA_2CsPb_2I_7$	15	45	[12]
$(PEA/NMA)_2Cs_{n-1}Pb_nI_{3n+1}$	18	47	[13]
$(PEA/PBA/NMA)_2Cs_{n-1}Pb_nI_{3n+1}$	3	48	[14]
$PPA_2Cs_{n-1}Pb_nI_{3n+1}$ (This work)	**2.1**	**45.1**	-

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