

## Supporting Information

### **Engineering of Annealing and Surface Passivation toward Efficient and Stable Quasi-2D Perovskite Light Emitting Diodes**

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## **Methods**

### **Materials**

PVK, PEABr, FABr, PbBr<sub>2</sub> and MACl were purchased from Xi'an Polymer Light Technology Corp. TPBi and LiF were purchased from Jilin Oled Material Technology Corp. Trimethylolpropane trimethacrylate was purchased from Energy-Chemical Corp. DMF, DMSO, and ethylacetate were purchased from J&K Scientific Corp. All the chemical materials were directly used without any further purifications.

### **Preparation of quasi-2D perovskite layers**

The quasi-two-dimensional perovskite precursor is configured by dissolving PEABr, FABr, PbBr<sub>2</sub> and MACl in DMSO at a stoichiometric ratio of 2:4:5:0.5, and the MACl acts as additive to improve the crystallization. The concentration of PbBr<sub>2</sub> is 0.2 M. After the solute is completely dissolved, 0.45  $\mu$ m organic filter is used to filter the solution. The precursor solution is spin-coated at 7000 rpm for 30 s, and then 100  $\mu$ L of ethyl acetate is poured onto the film at the 10th second, followed by annealing on a hot plate at 85 °C. For the TPTA modified film, TPTA is added to ethyl acetate at a concentration of 2 mM.

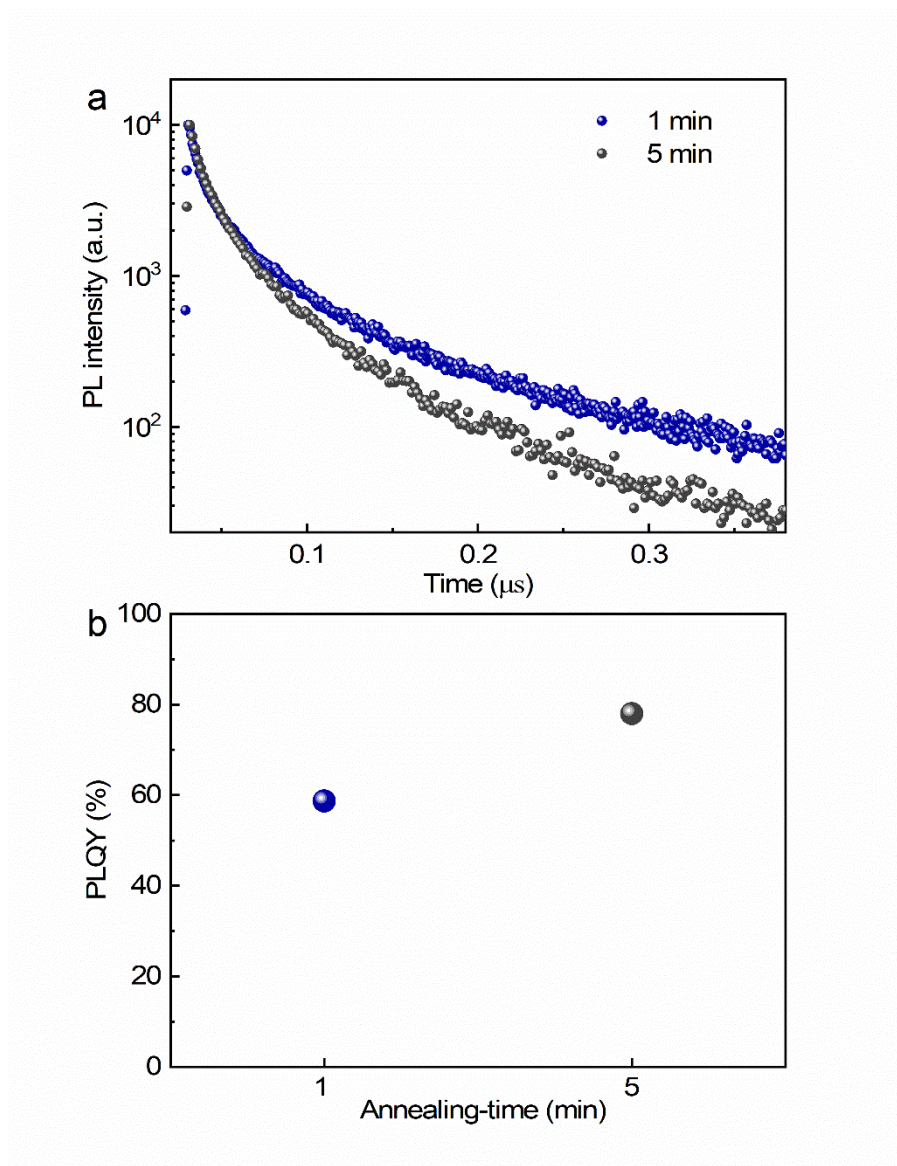
### **Light-Emitting Diodes Fabrication**

The indium tin oxide (ITO)-coated glass substrates are sequentially cleaned in detergent, distilled water, acetone and isopropanol by an ultrasonic cleaner. The pre-cleaned substrates are ultraviolet ozone treated for 30 min to make the surface hydrophilic, and then transferred into a nitrogen-filled glove box. PVK solution (10 mg mL<sup>-1</sup> in chlorobenzene) is spin-coated at 4000 rpm for 40 s and the films are baked at 120 °C for 30 min. After that, the perovskite film is deposited on PVK layer. Ultimately, TPBi (50 nm), LiF (1 nm), and Al (100 nm) are deposited by thermal evaporation, respectively. The active device area is 0.08 cm<sup>2</sup>

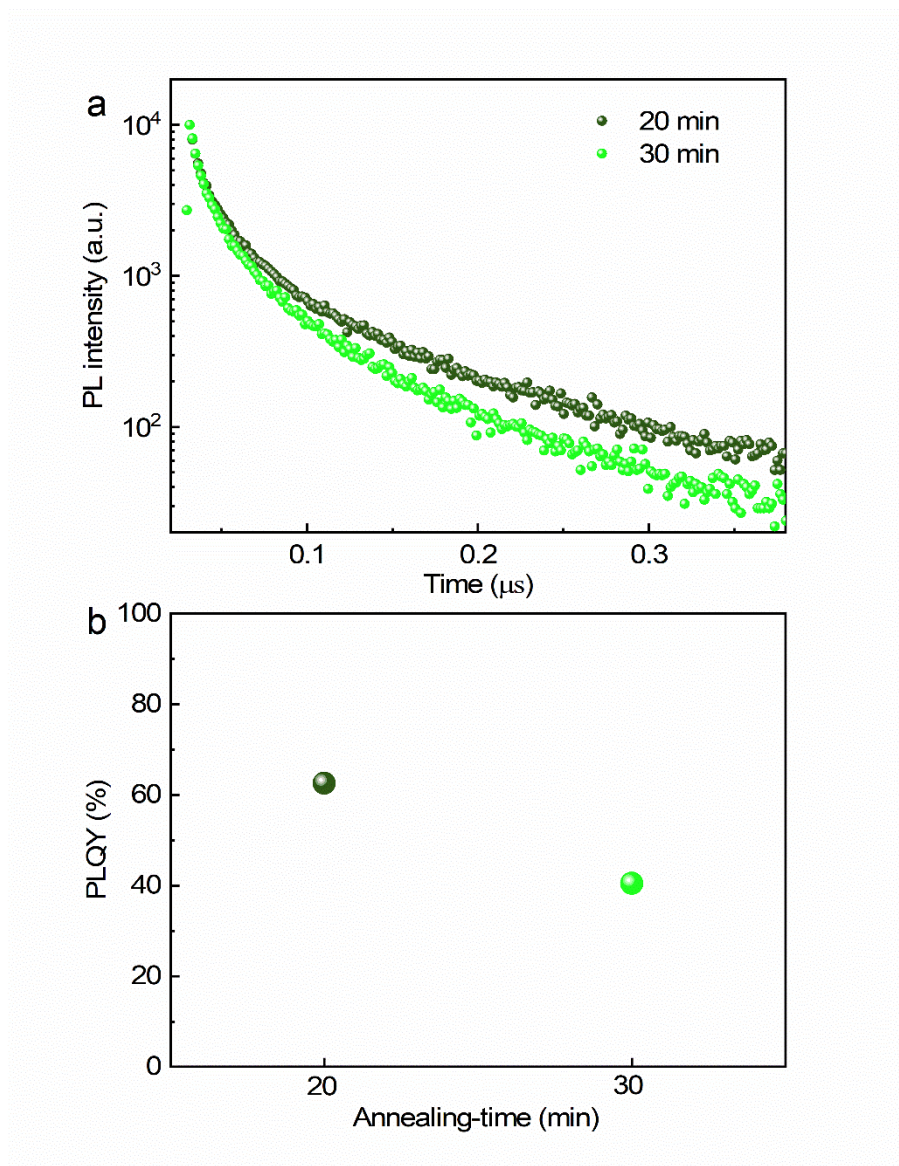
### **Characterization**

UV-Vis absorption spectra of quasi-2D perovskite layers were obtained by a Perkin-Elmer Lambda 35 UV-vis spectrometer. The GIXRD measurements were performed at Shanghai Synchrotron Radiation

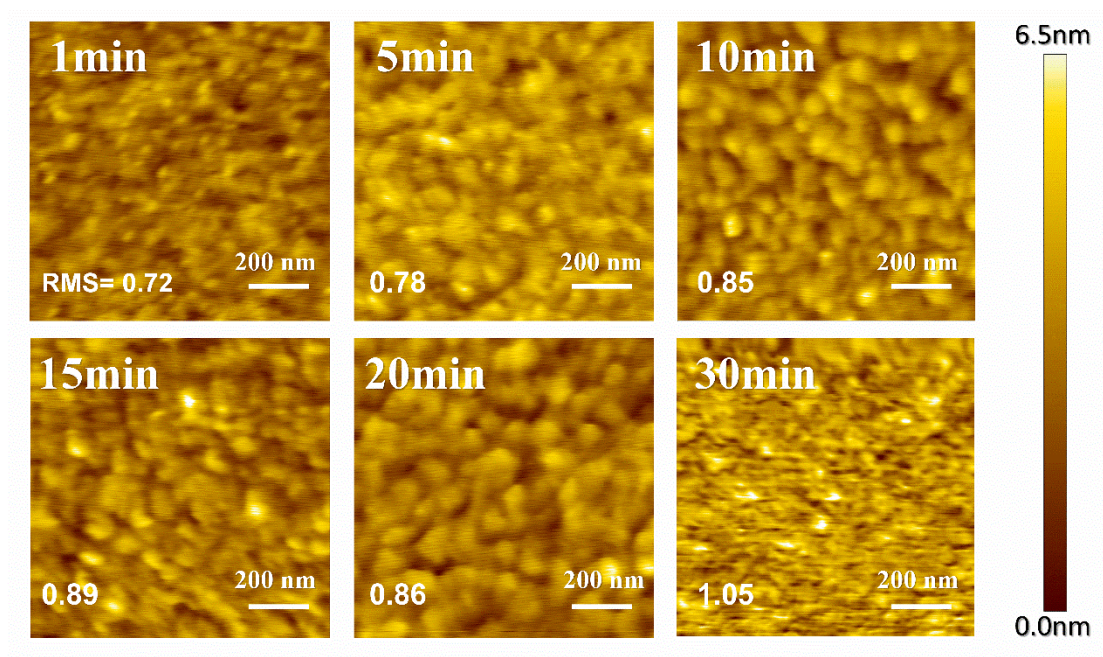
Facility using X-ray with a wavelength of 1.24 Å and an incidence angle of 0.13 °. PL spectra and PLQY were measured by a HORIBA FL3C-111 spectrofluorometer equipped with an integrating sphere, and the excitation wavelength is 385 nm from a xenon lamp light source. The transient PL-decay curves were measured with an Edinburgh FLS-980 fluorescence spectrophotometer with an excitation source of a 390 nm laser. Topography images of the corresponding films were collected by SPI3800N AFM. XRD curve was obtained using a Bruker D8 diffractometer with a wavelength of 1.54 Å and an incidence angle of 0.2 °. All the PeLEDs performance tests were performed with an Ocean Optics LED integrating sphere test system in glove box.



**Figure S1.** a) Transient fluorescence decay and b) PLQY of quasi-2D perovskite layer obtained by annealing for 1 min and 5 min.

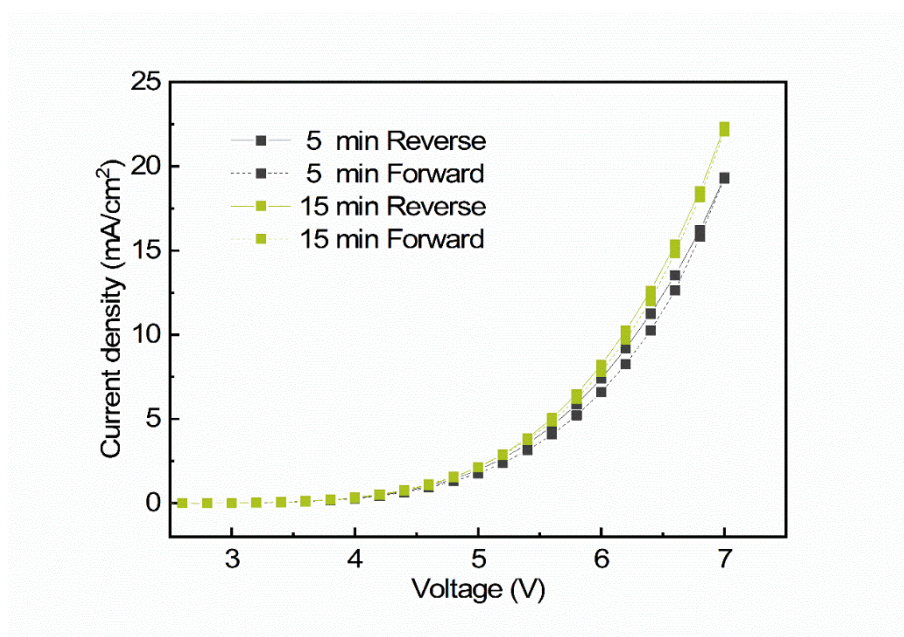


**Figure S2.** a) Transient fluorescence decay and b) PLQY of quasi-2D perovskite layer obtained by annealing for 20 min and 30 min.

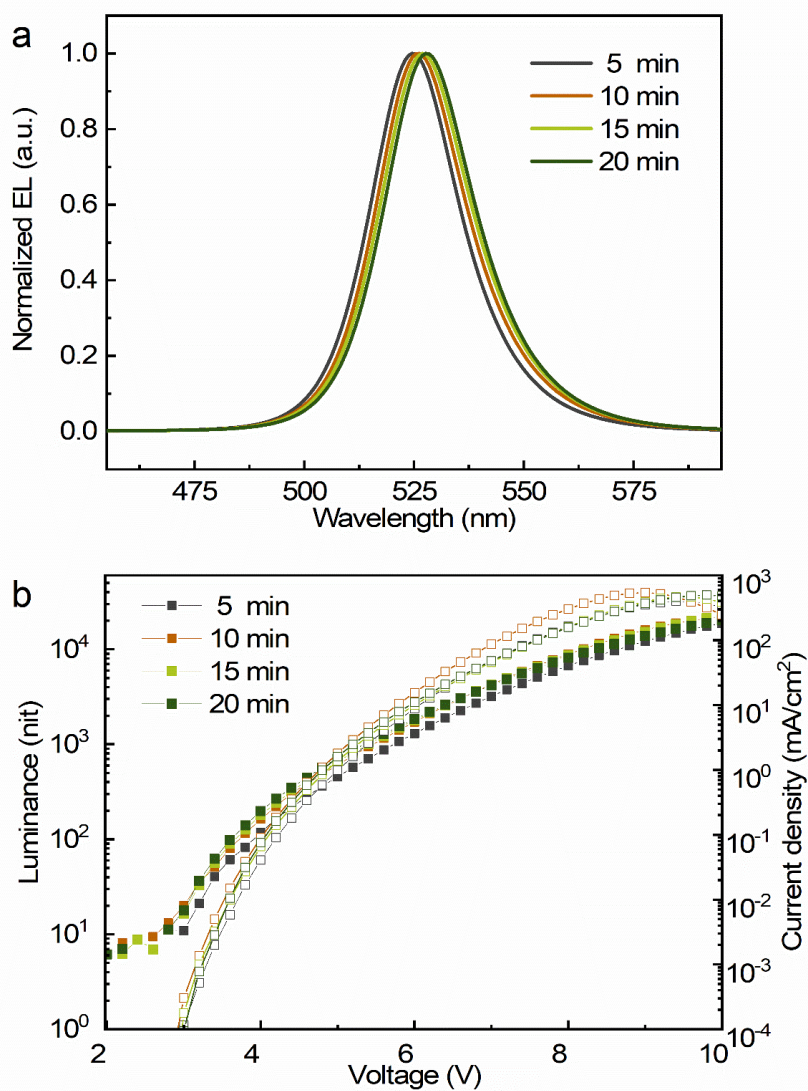


**Figure S3.** AFM images of quasi-2D perovskite films by annealing for different time.



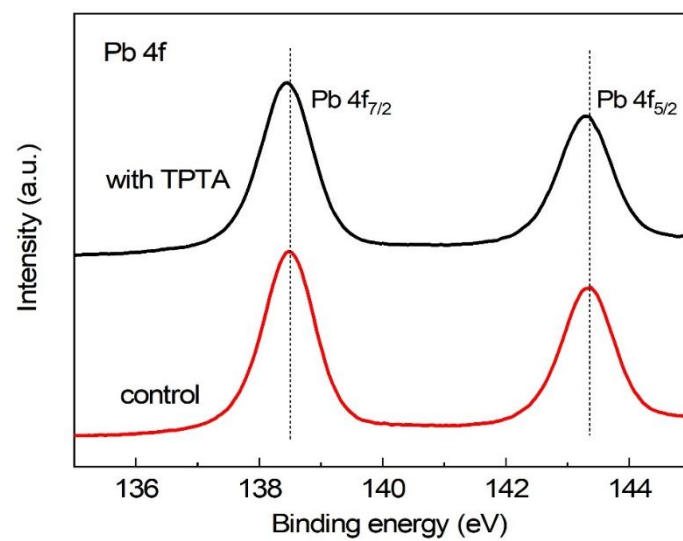


**Figure S4.** Current density-voltage hysteresis of PeLEDs prepared by different perovskite annealing time.

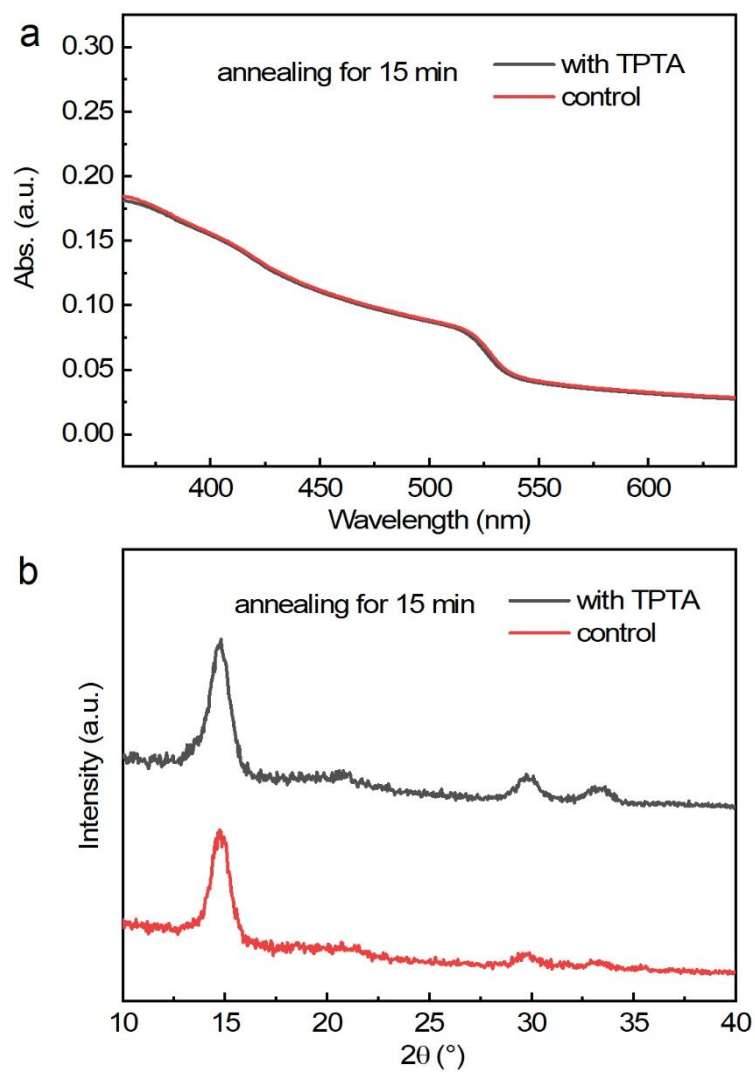


**Figure S5.** a) EL spectra of quasi-2D PeLEDs under the identical bias. b) Luminance-voltage-current density characteristics of fabricated LED devices.

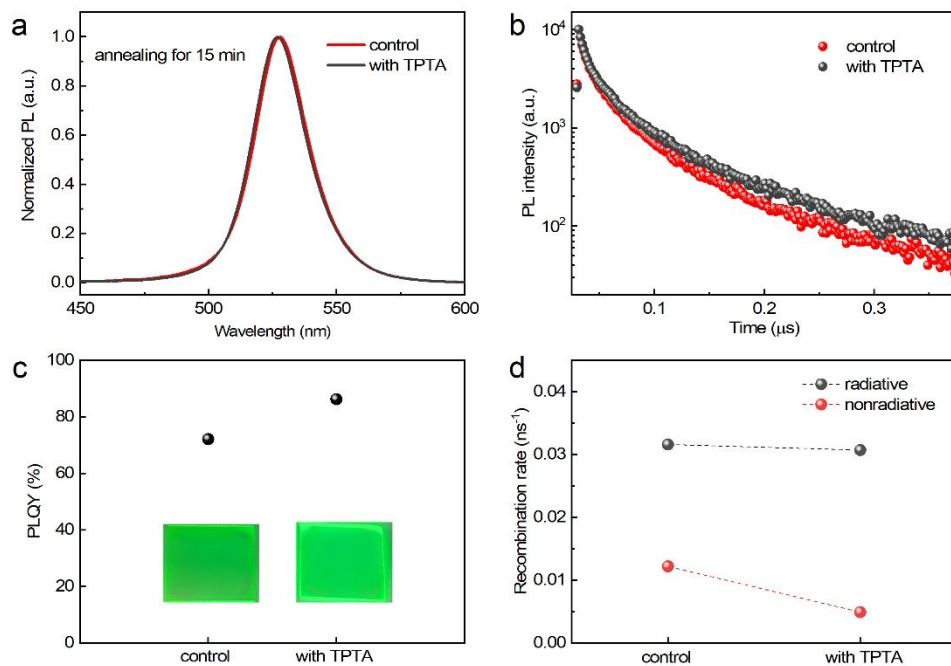




**Figure S6.** XPS spectra of the control and TPTA-treated films for Pb 4f orbit.



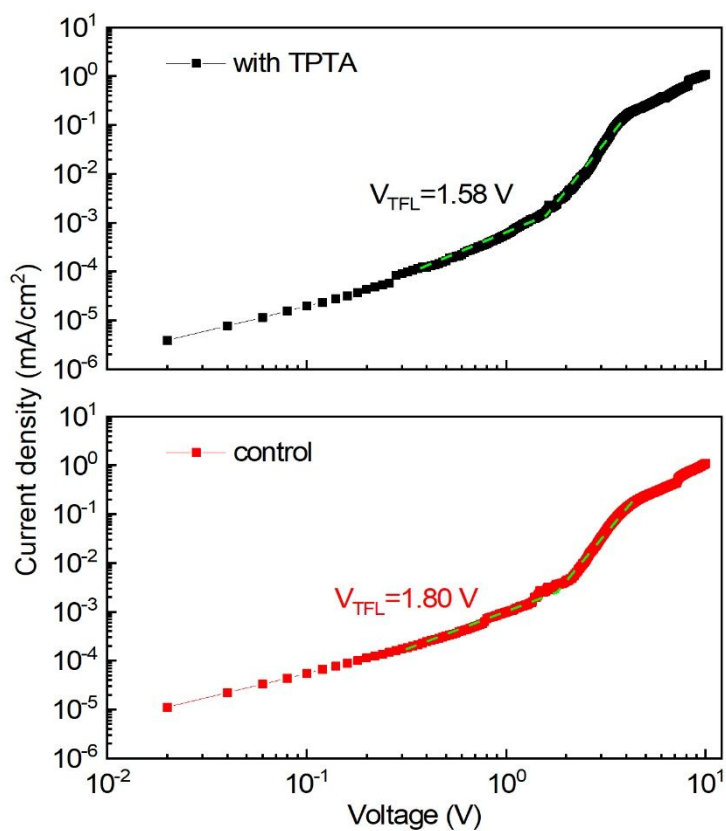
**Figure S7.** a) UV-Vis absorption spectra and b) XRD patterns of quasi-2D perovskite layer with or without TPTA.



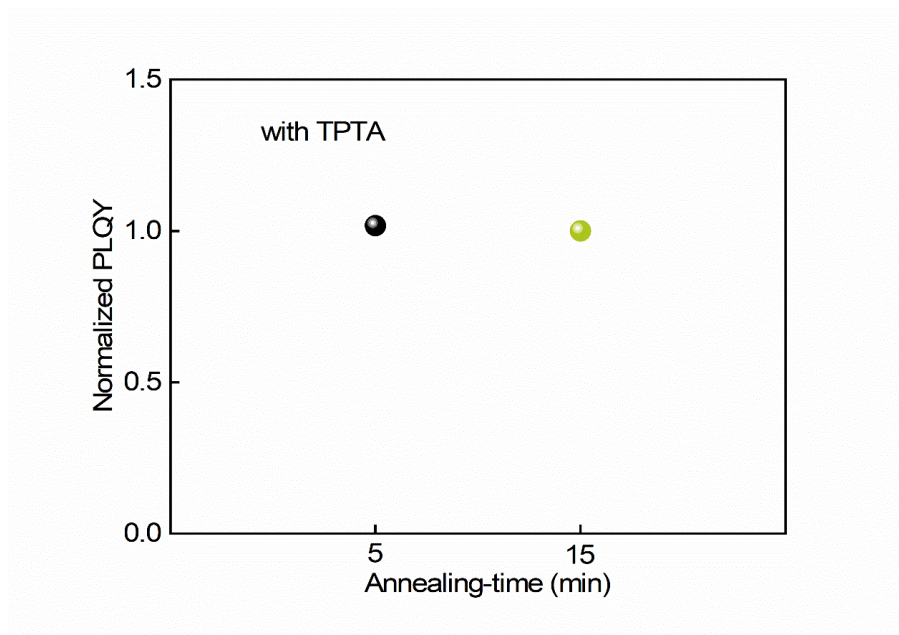
**Figure S8.** a) Steady-state PL, b) transient PL, c) PLQY, and d) radiative and nonradiative recombination rate of quasi-2D perovskite films with or without TPTA after annealing for 15 min.

**Table S1.** Summary of optical parameters of quasi-2D perovskite films with or without TPTA annealing for 15 min.

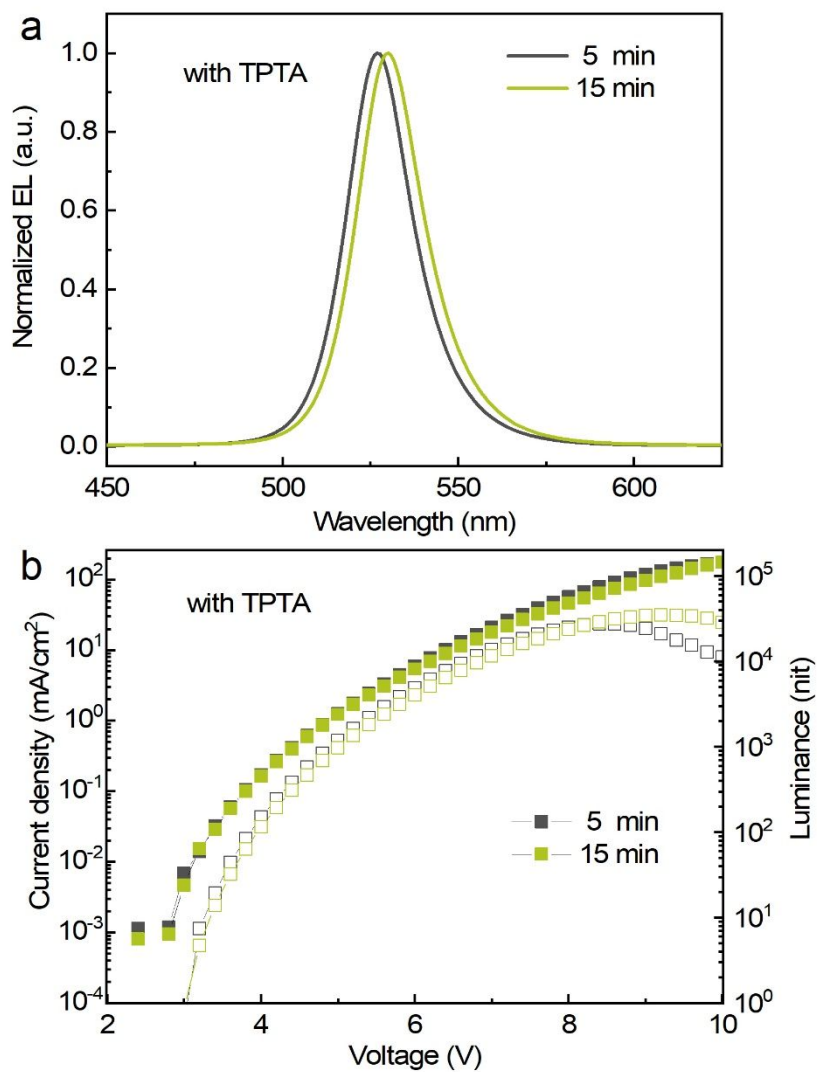
Sample	Wavelength (nm)	$\tau$ (ns)	PLQY (%)	$k_r$ (ns <sup>-1</sup> )	$k_{nr}$ (ns <sup>-1</sup> )
Control	527	22.8	72.1	0.0316	0.0122
With TPTA	527	28.0	86.2	0.0307	0.0049



**Figure S9.** Space-charge-limited current (SCLC) measurements of hole-only devices with structure of ITO/PVK/perovskite/MoO<sub>3</sub>/Ag.



**Figure S10.** PLQY of quasi-2D perovskite layer obtained by annealing for 5 min and 15 min with TPTA.



**Figure S11.** a) EL spectrum of TPTA-treated quasi-2D PeLEDs under the identical bias. b) Luminance-voltage-current density characteristics of TPTA-treated LED devices.



**Table S2.** Summary of device performance with or without TPTA.

Device	$V_{\text{on}}$ (V)	$L_{\text{max}}$ (cd m <sup>-2</sup> )	$\text{EQE}_{\text{ave}}$ (%)	Wavelength (nm)
Control, 5min	3.0	31931	17.7	526-523
Control, 10min	3.0	39400	15.7	530-526
Control, 15min	3.0	35713	14.0	530-526
Control, 20min	3.0	37244	12.6	530-526
With TPTA, 5min	3.0	27687	18.1	527-525
With TPTA, 15min	3.0	35025	18.2	530-527