

## Electronic Supplementary Information

### ***In situ* nanocrystallization on the perovskite film surface for enhanced light emission**

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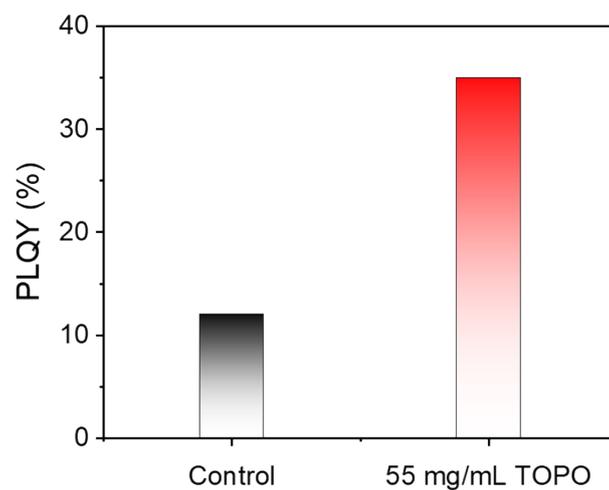
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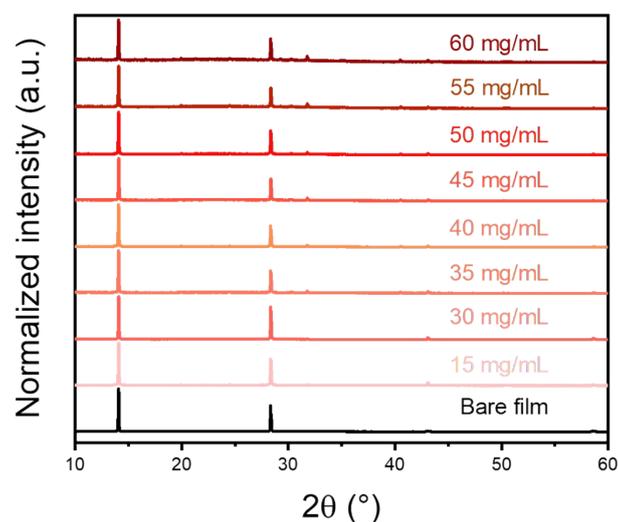
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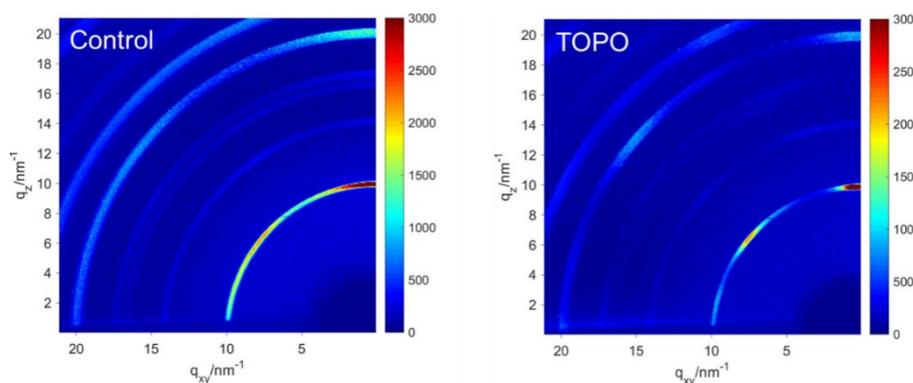
<sup>†</sup> H.T. and S.L. contributed equally to this work.



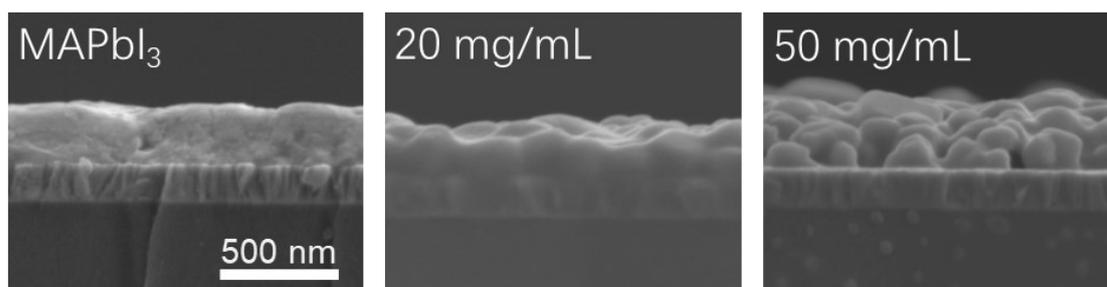
**Fig. S1.** PLQY results of control and 55 mg/mL TOPO solution-treated perovskite films.



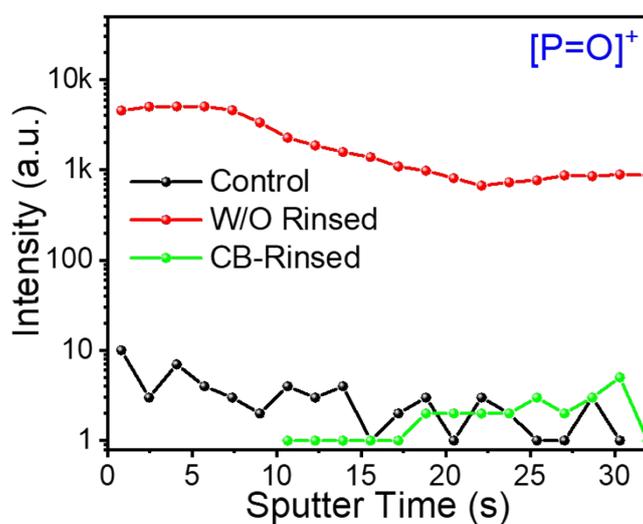
**Fig. S2.** XRD results of MAPbI<sub>3</sub> films with different concentrations of TOPO solutions.



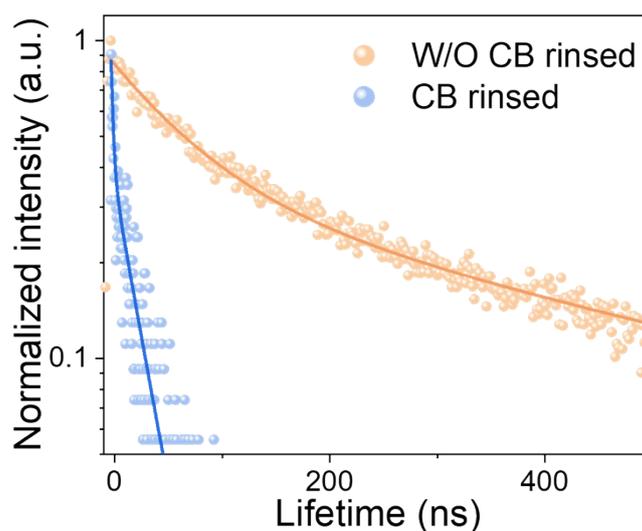
**Fig. S3.** GIWAXS results of MAPbI<sub>3</sub> films without (control) and with the TOPO treatment.



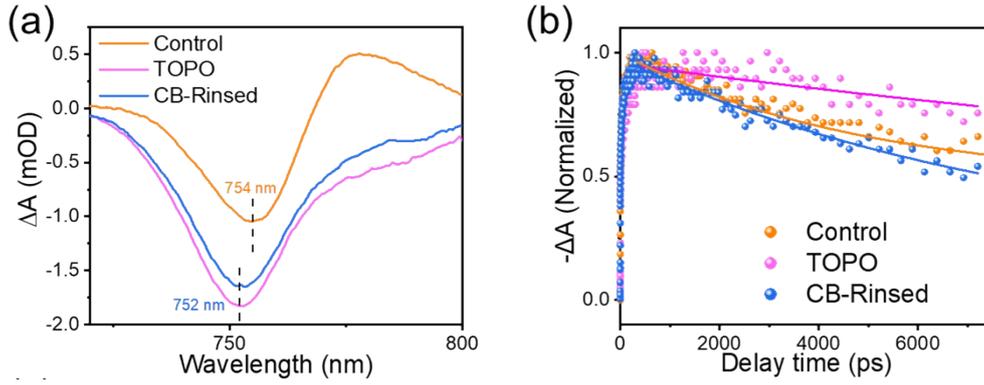
**Fig. S4.** Cross-sectional SEM images of the perovskite film with a thickness of 200 nm, treated with different concentrations of TOPO solutions.



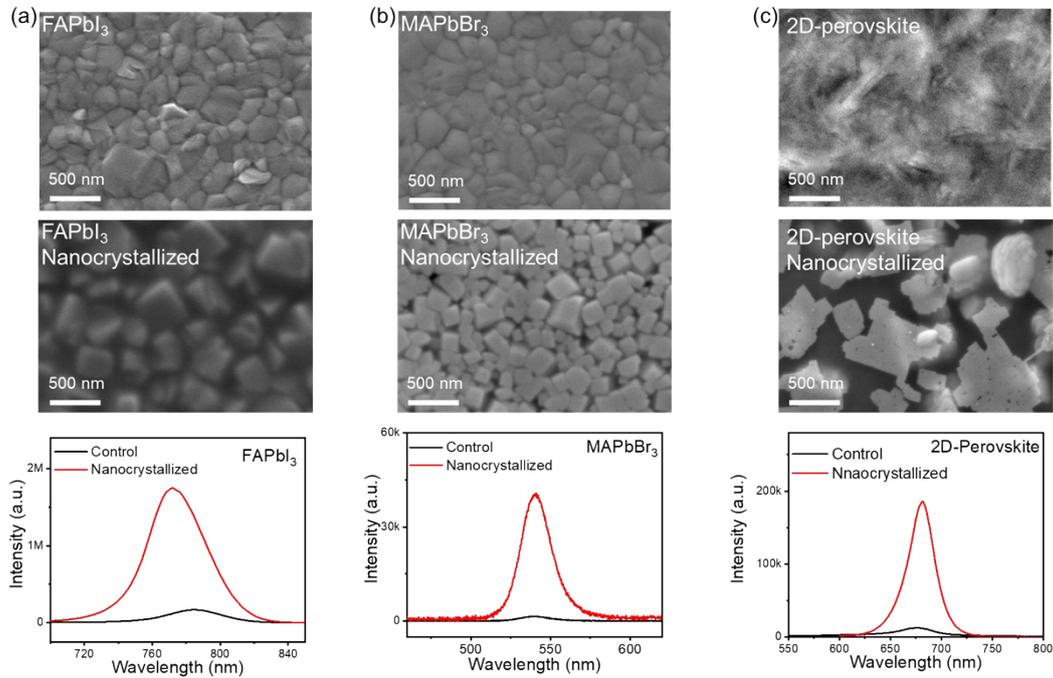
**Fig. S5.** ToF-SIMS results of perovskite films of control, without CB rinsing (W/O Rinsed) and with CB rinsing (CB-Rinsed).



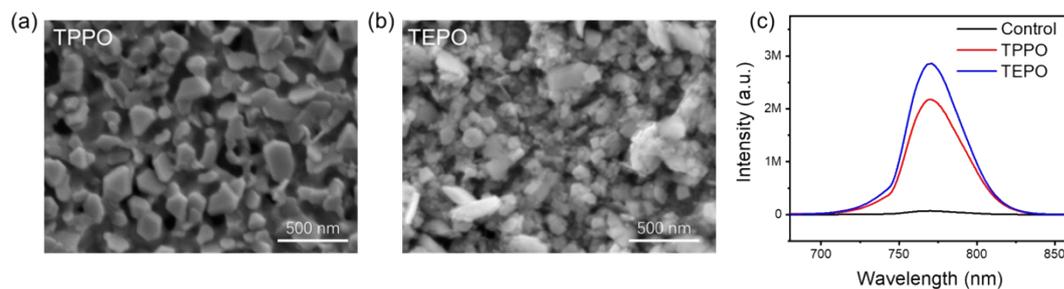
**Fig. S6.** Double-exponential fitting carrier lifetime of without CB-rinsed and CB-rinsed perovskite films.



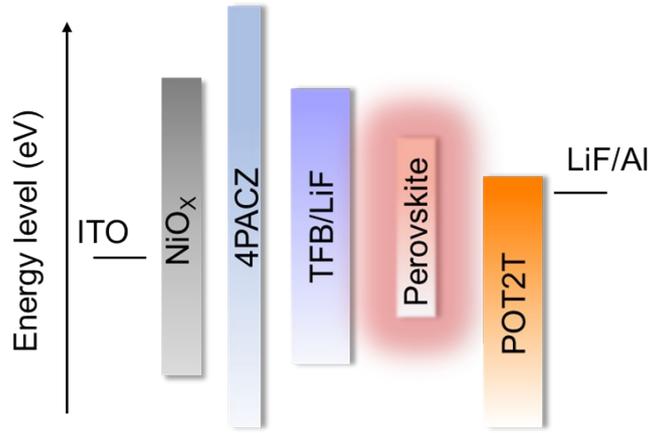
**Fig. S7.** (a) Transient absorption spectroscopy results and (b) normalized transient decays plots of Control, TOPO-modified and CB-rinsed samples.



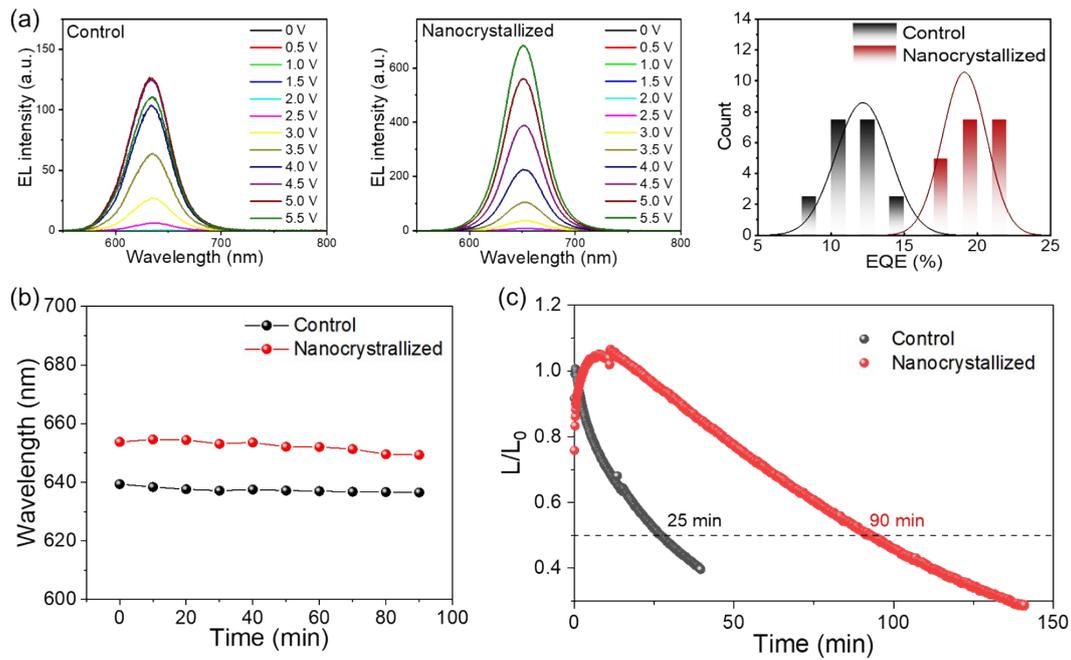
**Fig. S8.** SEM and PL results of with and without nanocrystallization of FAPbI<sub>3</sub>, MAPbBr<sub>3</sub> and quasi-2D perovskite films modified by TOPO.



**Fig. S9.** SEM and PL results of MAPbI<sub>3</sub> films modified by triethylphosphine oxide (TEPO) and triphenylphosphine oxide (TPPO).



**Fig. S10.** Energy level diagram of perovskite light-emitting diode in this work.



**Fig. S11.** (a) EL spectra of PeLED devices at different voltages, and statistical EQEs of control and nanocrystallized PeLEDs. (b) EL peak position evolution and (c) operating lifetime for the PeLEDs.