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Supplementary Information

Suppressing the Defects in Cesium-Based Perovskite via Polymeric Interlayer Assisted Crystallization Control

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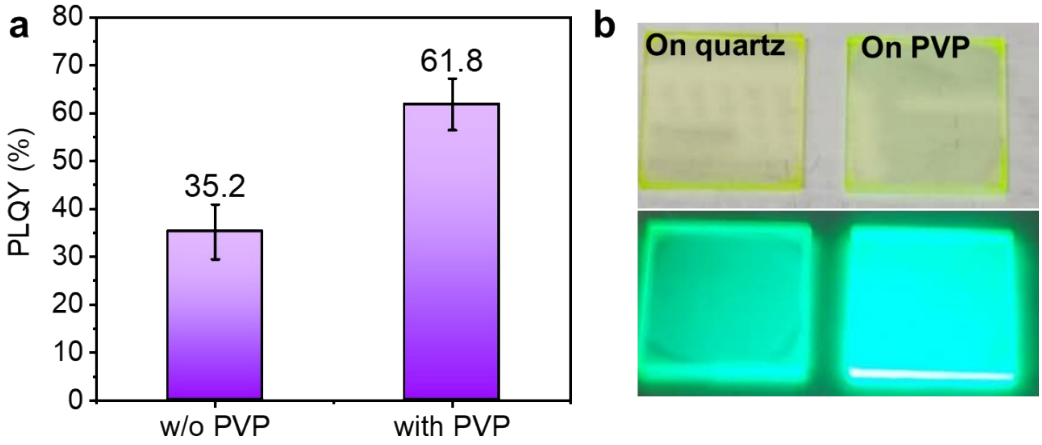

Figure S1. a PLQY of perovskite films without and with interfacial engineering. **b** photograph of perovskite films on quartz substrate and PVP substrate under daylight (top) and 365 nm UV-light (bottom).

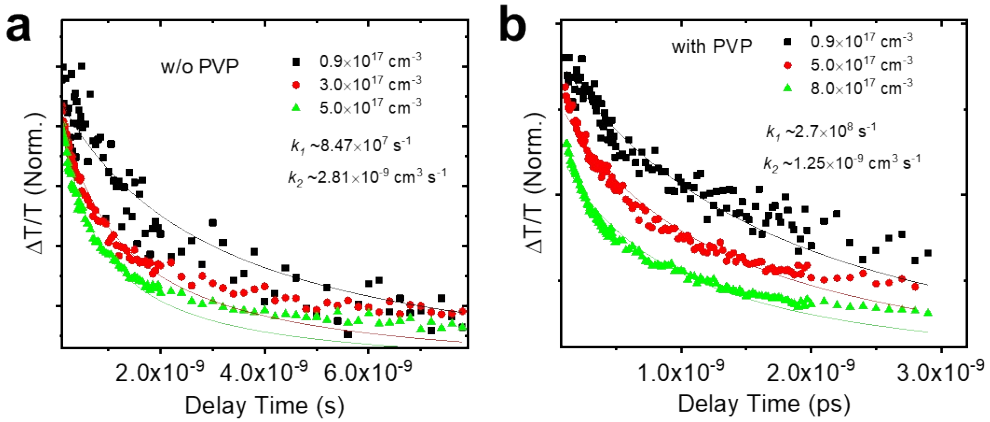

Figure S2. a-b TA kinetics under different pump fluence probed at the characterized bleaching peaks for the perovskite films without **a** and with **b** interfacial engineering with PVP.

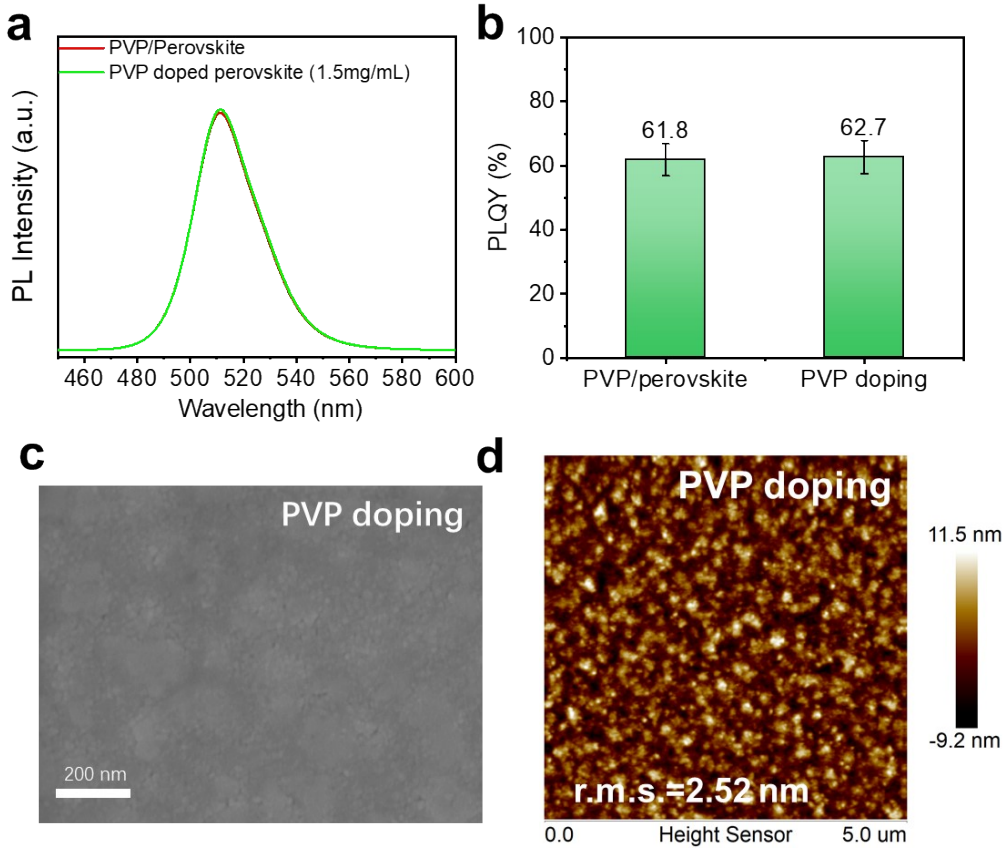

Figure S3. Perovskite films with PVP doping. a PL spectrum of perovskite films with PVP layer and with PVP doping (1.5 mg/mL). **b** PLQY statistics of two kinds of perovskite films. **c** SEM image of perovskite film with PVP doping. **d** AFM image of perovskite film with PVP doping.

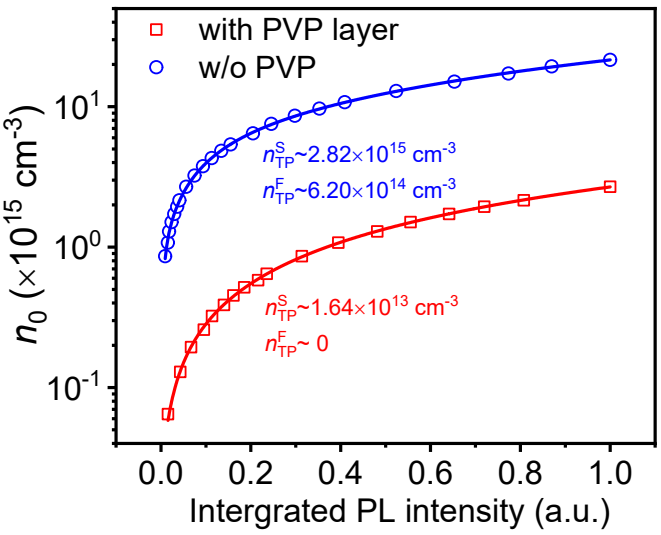

Figure S4. PL intensity as a function of pump fluence, measured to estimate the trap density.

Notes: The detailed trap state densities were estimated with injected carrier density dependent PL measurement. The photo-generated carrier density can be calculated as:

$$\rho_{photocarrier} = \frac{light\ fluence\ density\ of\ single\ puulse\ \times light\ absorbance}{photo\ energy\ \times film\ thickness}$$

Where the light absorbance is obtained from UV-visible absorbance spectrum. Film thickness is 80 nm, which is tested by step profiler.

The trap states density can be estimated by [16]:

$$n_c(0) = \sum_i n_{TP}^{i}(0)(1 - e^{-\frac{a_i \tau_0 I_{PL}}{k}}) + I_{PL}/k$$

Where the $n_c(0)$ is the initial photogenerated charge carrier density, n_{TP}^{i} is the trap density, a_i is the product of the trapping cross section and the carrier velocity. Fitting the dependence of injected charge carrier density and PL intensity with the above equation yields two types of traps in the perovskite films, with the intrinsic traps

exhibiting fast trapping times, while the surface/interfacial traps exhibiting slow trapping times. [1-3] The intrinsic bulk trap density is calculated as n_{TP}^{F}~6.20×10^{14} cm^{-3}, while the surface/interfacial trap density is n_{TP}^{S}~2.82×10^{15} cm^{-3}. By comparison, for the PVP modified perovskite films, the bulk trap density is beyond measurement, the surface defect density is n_{TP}^{S}~1.64×10^{13} cm^{-3}.

Table S1. Trap state density of perovskite films with and without PVP layer.

	n_{TP}^{S} **[cm^{-3}]**	n_{TP}^{F} **[cm^{-3}]**	n_{TP}^{total} **[cm^{-3}]**
w/o PVP	2.82±0.14 ×10^{15}	6.20±1.15 ×10^{14}	3.44±0.255 ×10^{15}
With PVP	1.64±0.34 ×10^{13}	0±0	1.64±0.34 ×10^{13}

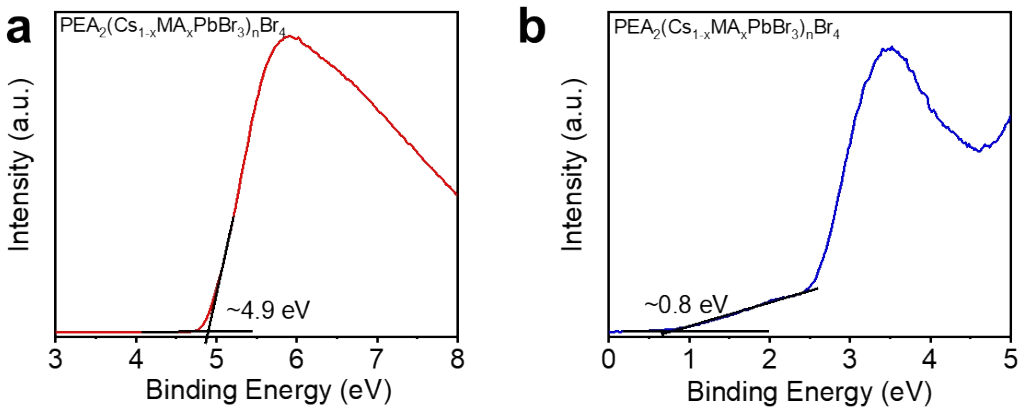

Figure S5. UPS measurement of perovskite films. The work function (W_F) is 4.9 eV, valence band maximum (VBM) is 0.8 eV. The bandgap is 2.7 eV, calculated from absorption spectrum in Figure 1b.

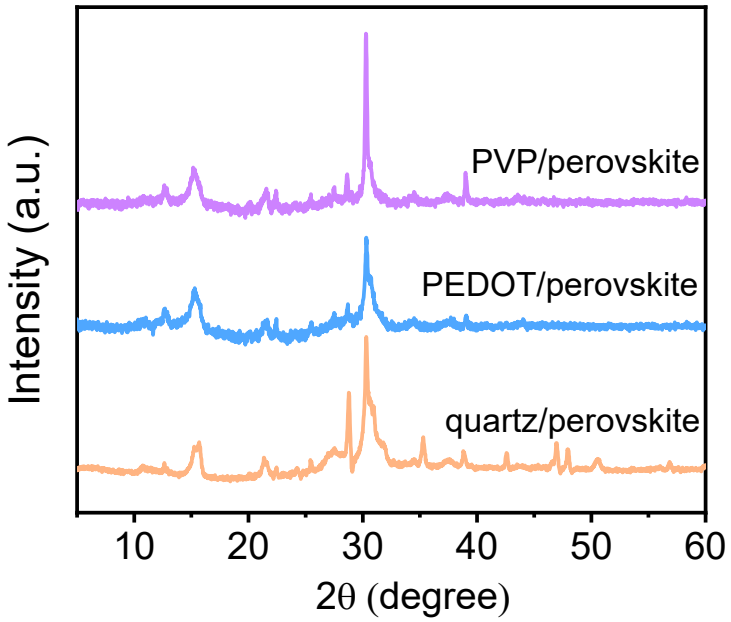

Figure S6. XRD pattern of perovskite film on quartz, PEDOT: PSS and PVP substrate.

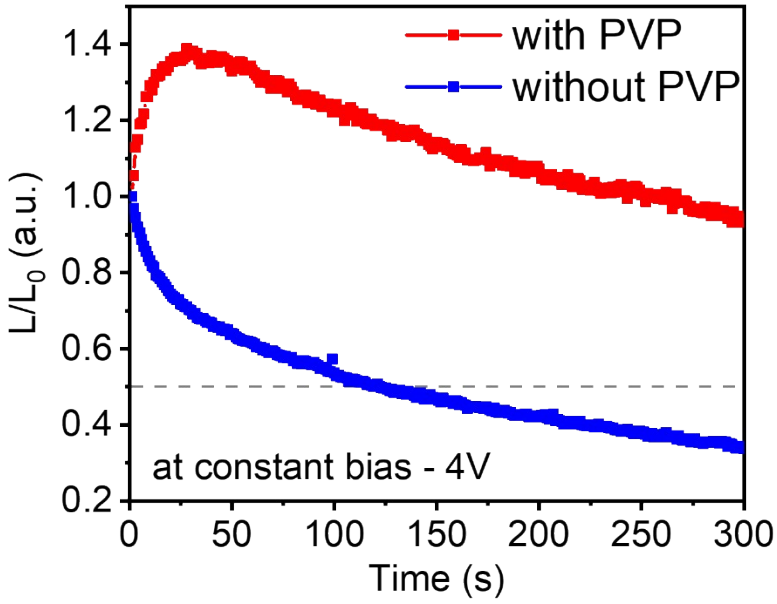

Figure S7. Operational stability of devices at constant bias (4V) with and without PVP interlayer.

Reference

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[2] G. Xing, G. Xing, M. Li, E. J. Sie, D. Wang, A. Sulistio, Q. Ye, C. Huan, T. Wu, and T. C. Sum, *Appl. Phys. Lett*. 2011, **98**, 102105;

[3] G. Xing, N. Mathews, S. Sun, S. S. Lim, Y. Mimg, M. Grätzel, S. Mhaisalkar and T. C. Sum, *Science*, 2013, **342**, 344.