

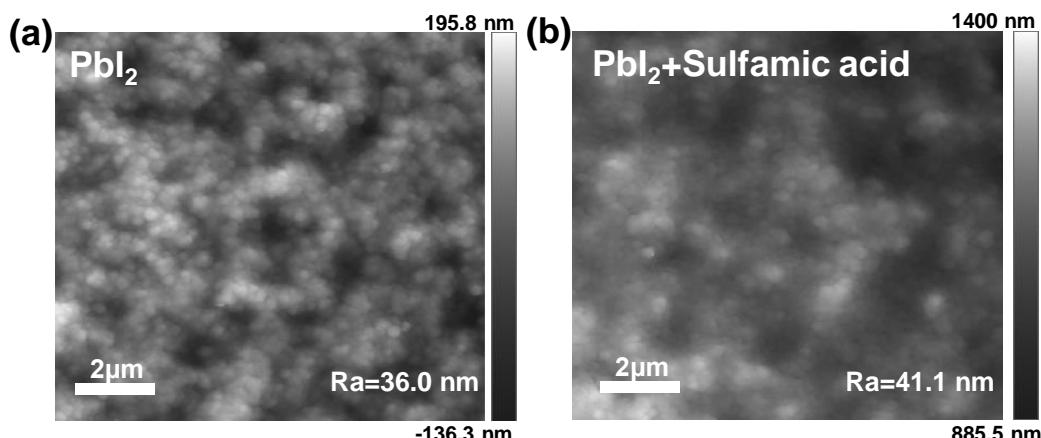
Supporting Information

Interdiffusion-controlled nucleation strategy for efficient sequential deposited perovskite photovoltaics

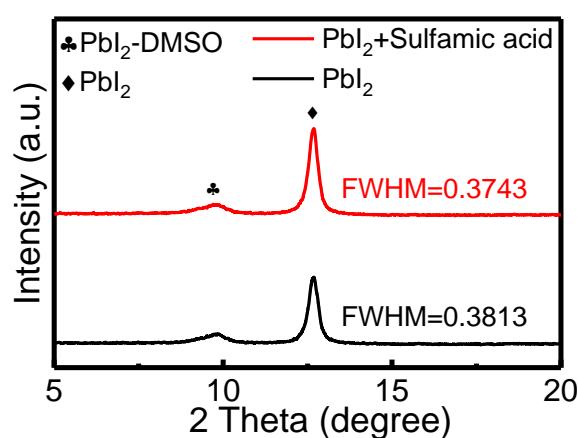
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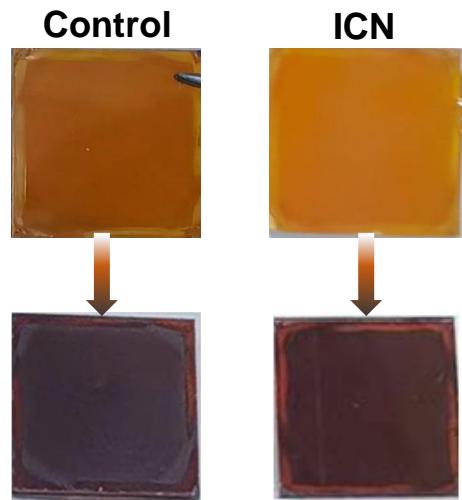
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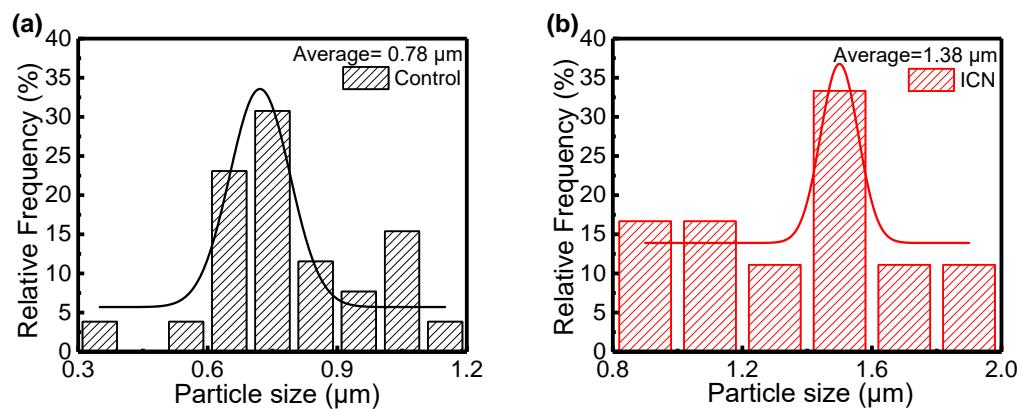
**Fig.S1.** Surface roughness of (a) the PbI<sub>2</sub> film and (b) the PbI<sub>2</sub> film with ICN modification, which was measured by AFM.



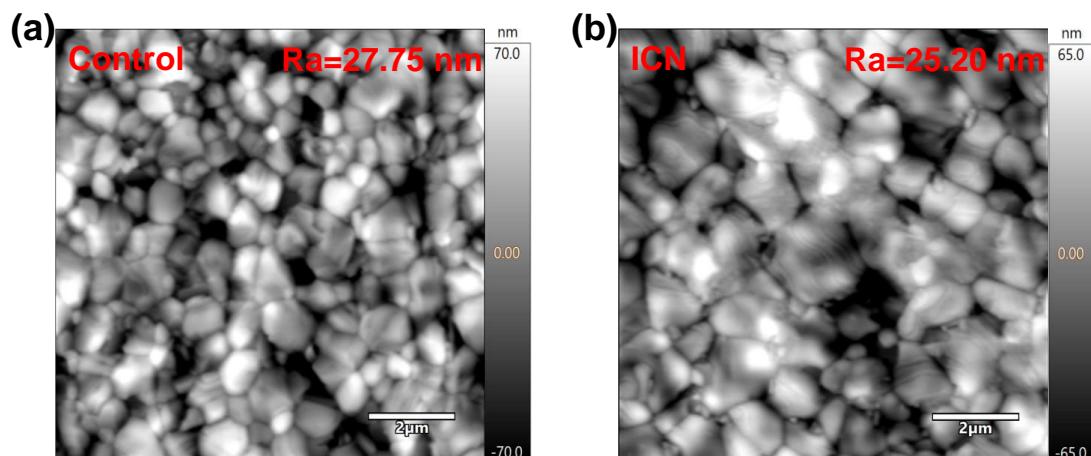
**Fig. S2.** XRD patterns spectra of the PbI<sub>2</sub> films without and with Sulfamic acid modification.



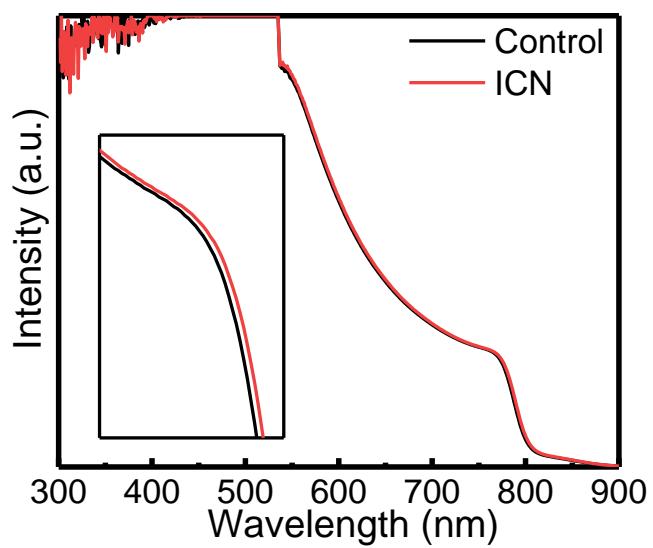
**Fig.S3.** Photograph of unannealed perovskite films over time.



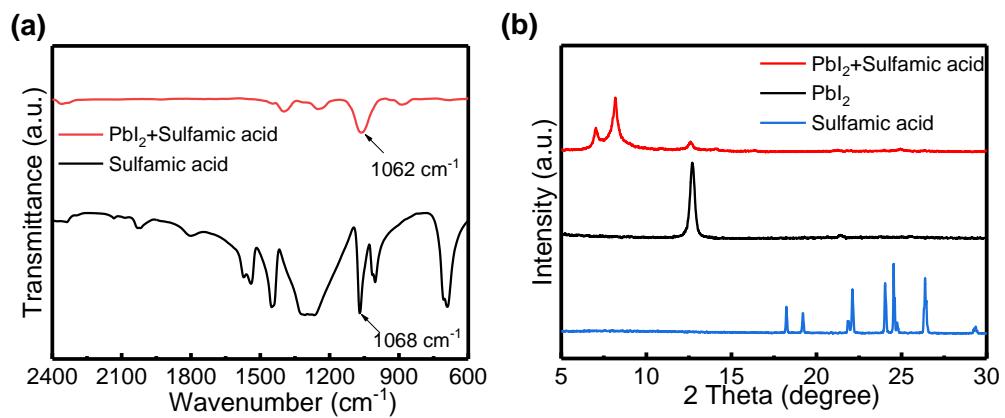
**Fig.S4.** Statistical distribution of grain size of the perovskite films (a) without and (b) with ICN modification.



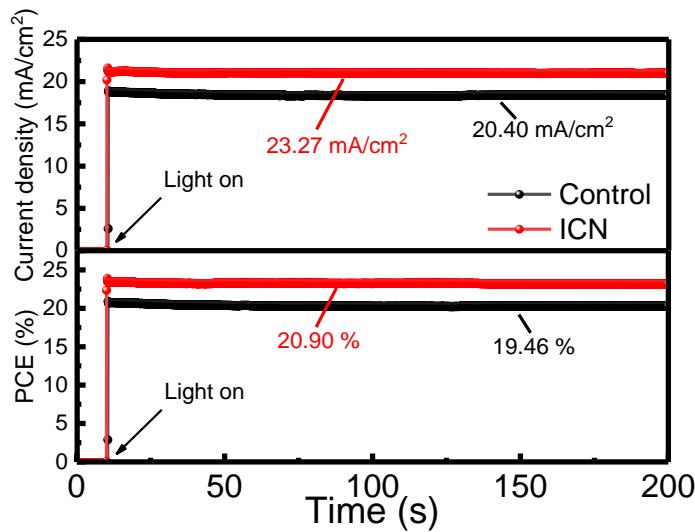
**Fig.S5.** Surface roughness of the perovskite films (a) without and (b) with ICN modification, which were measured by AFM.



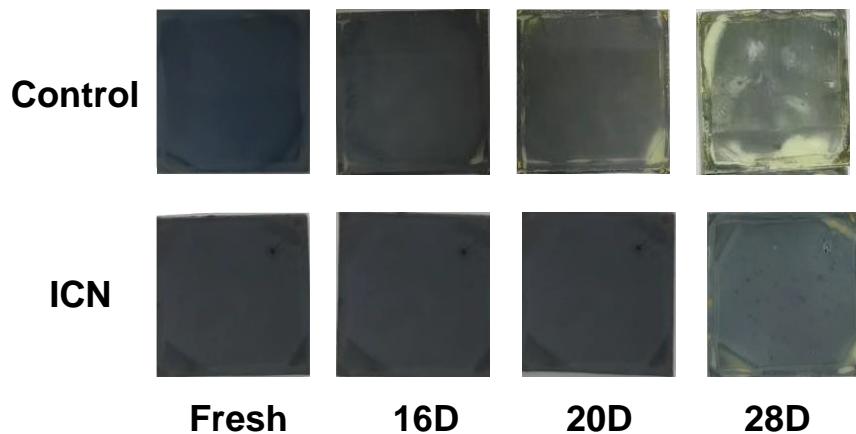
**Fig.S6.** UV-vis absorption spectra of the perovskite films with and without ICN modification.



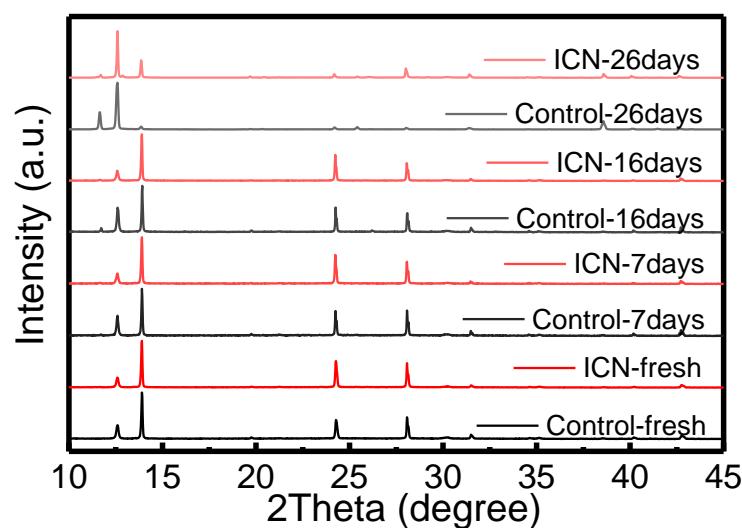
**Fig.S7.** (a)Fourier transform infrared spectroscopy of Sulfamic acid powder and  $\text{PbI}_2$ +Sulfamic acid. (b)XRD patterns spectra of  $\text{PbI}_2$  with ICN modification,  $\text{PbI}_2$ , and Sulfamic acid.



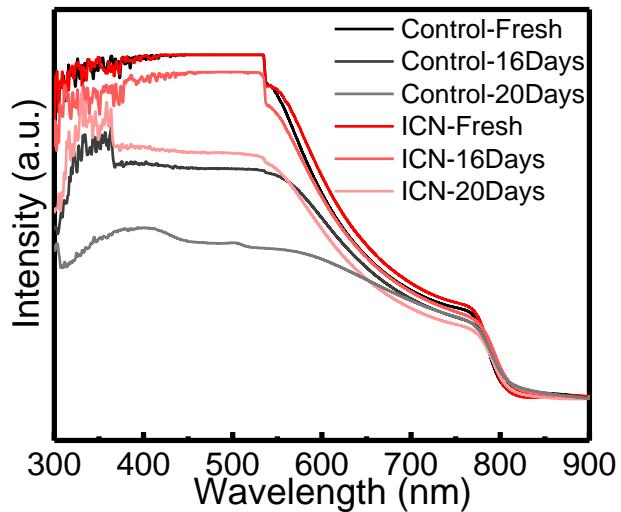
**Fig.S8.** Current density and PCE were measured at the maximum power point for 200 s.



**Fig.S9.** Photos of morphological changes of the control and the perovskite films with ICN modification over time.

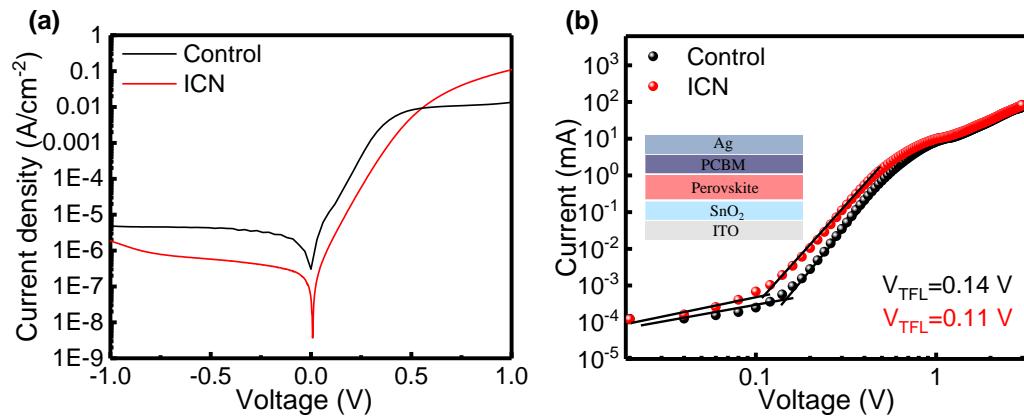


**Fig.S10.** XRD patterns of the perovskite films over time.

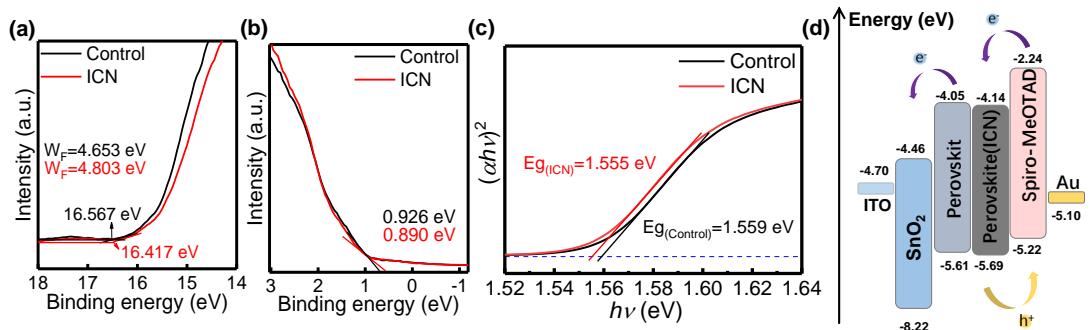


**Fig.S11.** UV-vis absorption spectrum of the perovskite films change over time.

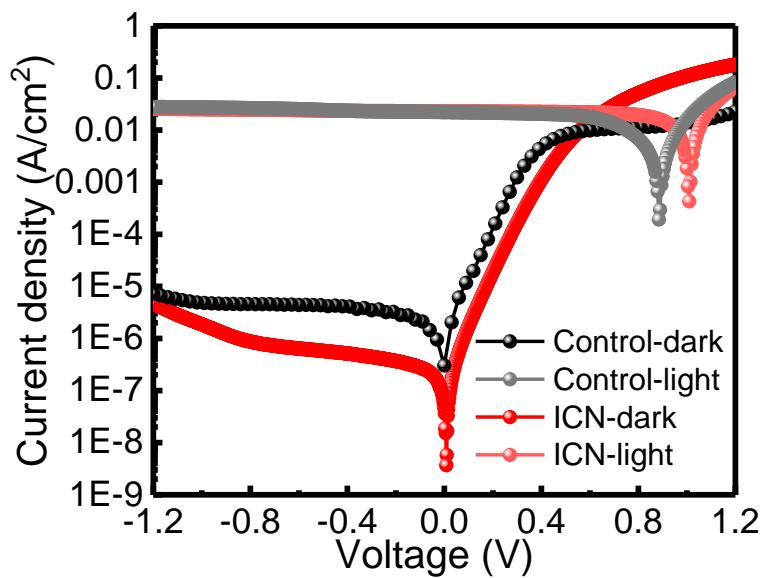
In addition, we also performed SCLC on the electron transport layer-only devices (Fig. S12b). Calculation analysis confirmed that the  $V_{TFL}$  of control and ICN-modified perovskite films is 0.14 V and 0.11 V, respectively. Consequently, the trap density of the control and ICN-modified perovskite films were calculated to be  $2.41 \times 10^{15} \text{ cm}^{-3}$  and  $1.89 \times 10^{15} \text{ cm}^{-3}$ , respectively.



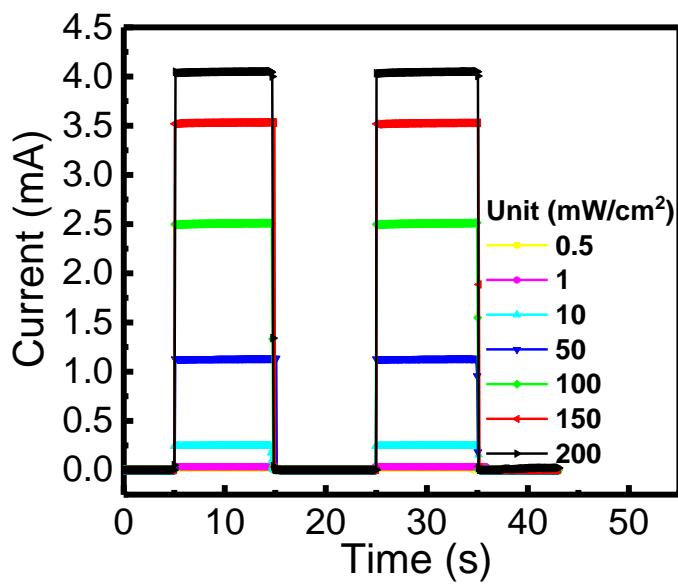
**Fig.S12.** (a)The dark current of the PSCs with and without ICN modification. (b) Dark J–V curves of the electron transport layer-only devices.



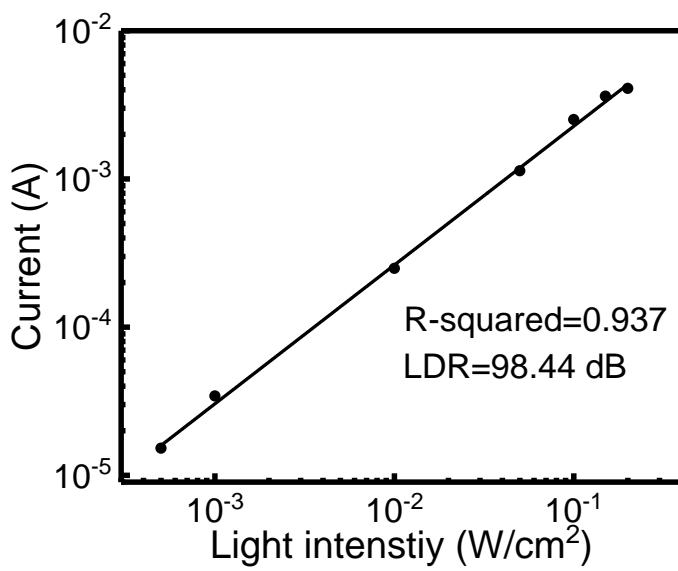
**Fig.S13.** (a, b) Ultraviolet photoelectron spectra of control and perovskite film with ICN modification, respectively. (c) UV–vis absorption spectra of control and the perovskite film with ICN modification. (d) Energy level diagram of device components.



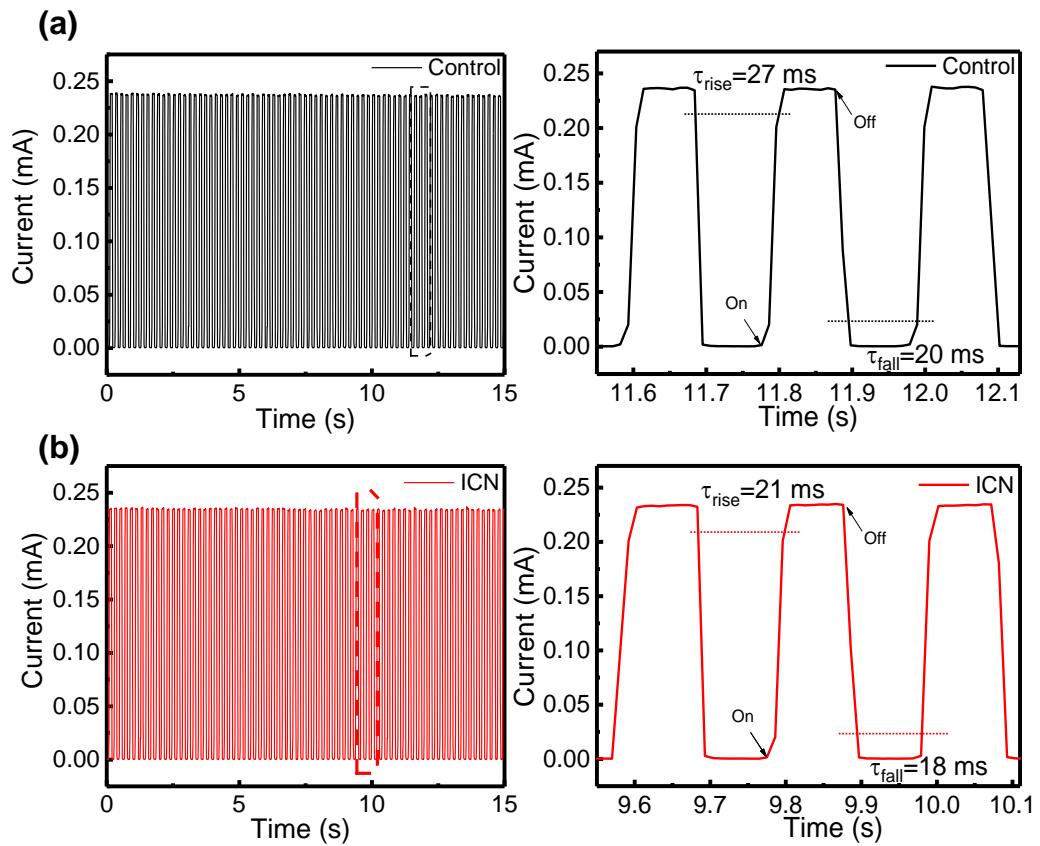
**Fig.S14.**  $J$ - $V$  curves of the devices in the dark and under white light illumination (100  $\text{mW}/\text{cm}^2$ ).



**Fig.S15.**  $I-T$  curves of the pristine photodetector under various light intensities under 0 V bias.



**Fig.S16.** Power-dependent photocurrent under 0 V bias for the pristine photodetector.



**Fig.S17.** The photocurrent response curves of (a) the pristine photodetector and (b) the pristine photodetector with ICN modification under 810 nm light source, respectively. Corresponding rise and decay time from the enlarged photocurrent response curve of photodetectors are shown in the right part.

**Table S1.** Photovoltaic parameters of the PSCs ( $\text{FA}_x\text{MA}_{1-x}\text{Pb}(\text{I}_y\text{Br}_{1-y})_3$ ) based on the control and the ICN-treated PSCs measured in reverse scan (RS).

Devices	$V_{\text{oc}}$ (V)	$J_{\text{sc}}$ (mA/cm)	FF (%)	PCE (%)
Control-Avg	1.119	24.84	74.54	20.67
Control-Max	1.127	24.95	76.87	21.09
ICN-Avg	1.135	25.05	77.49	22.04
ICN-Max	1.141	25.21	77.84	22.41

**Table S2.** Photovoltaic parameters of the PSCs ( $\text{FAPbI}_3$ ) based on the control and the ICN-treated PSCs measured in reverse scan (RS).

Devices	$V_{\text{oc}}$ (V)	$J_{\text{sc}}$ (mA/cm)	FF (%)	PCE (%)
Control-Avg	1.124	25.38	76.78	21.90
Control-Max	1.129	25.18	77.63	22.06
ICN-Avg	1.144	25.72	77.60	22.85
ICN-Max	1.154	25.47	78.51	23.08

**Table S3.** Fitting data of TRPL curves in Figure 4f based on double exponential function.

Samples	$\tau_1$ (μs)	$A_1$	$\tau_2$ (μs)	$A_2$	$\tau_{\text{ave}}$ (μs)
Glass/PVSK	0.85	0.42	4.30	0.58	3.87
Glass/PVSK(ICN)	1.31	0.27	7.62	0.73	7.23