Supporting Information

Inverted Planar Solar Cell with 13% Efficiency and Sensitive Visible Light Detector Based on Orientation Regulated 2D Perovskites

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Fig. S1 Absorption spectra of $(PEA)_2(MA)_4Pb_5I_{16}$ (n = 5) perovskite films with various amounts of NH₄SCN additive on ITO/PEDOT:PSS substrate.



Fig. S2 2D GIWAXS patterns of $(PEA)_2(CH_3NH_3)_4Pb_5I_{16}$ (n=5) films made by MA process (a) without and (b) with 1.5 SCN addition.



Fig. S3 (a) XRD patterns and (b) the corresponding diffraction peak intensity and FWHM of the MA-based $(PEA)_2(CH_3NH_3)_4Pb_5I_{16}$ (n=5) perovskite films with various addition ratio of NH₄SCN on ITO/PEDOT:PSS substrate.



Fig. S4. Raman spectra of pristine NH_4SCN (a) and $(PEA)_2(MA)_4Pb_5I_{16}$ films with various amounts of NH_4SCN addition after thermal annealing (b).



Fig. S5 Atomic force microscopy of the $(PEA)_2(CH_3NH_3)_4Pb_5I_{16}$ (n=5) perovskite films fabricated by (a) CA and (b) MA processes on ITO/PEDOT:PSS substrate.



Fig. S6 XRD patterns of perovskite films fabricated by CA and MA processes.



Fig. S7 (a) Time-resolved photoluminescence measurements, (b) Steady-state photoluminescence spectra of the perovskite films on the PEDOT:PSS substrates with $PC_{61}BM$ above.

Spectral densities of 1/f noise, shot noise and thermal noise of the MA-based device.

The 1/f noise $(S_l(1/f))$ can be calculated by equation of $S_l(1/f) = |l(f)|^2/(Fs \times N)$, in which l(f) is the discrete Fourier transform of the dark current waveform l(t), Fs is the sampling rate, N is the number of data points. The calculated spectral density of 1/f noise is shown in Fig. S8. At a modulation frequency of 10 Hz, the value of $S_l(1/f)$ is about 2.0×10⁻²¹A²/Hz.

The shot noise ($S_l(shot)$) can be expressed as $S_l(shot) = 2 \times q \times I_{dark}$, where q is the elemental charge, I_{dark} is the dark current of the device. The calculated $S_l(shot)$ is about 9.67×10⁻²⁷ A²/Hz.

The thermal noise (*S_i(thermal*)) is calculated by using Nyquist's equation, *S_i(thermal*) = $4 \times k_B \times T/r$, where k_B is the Boltzmann's constant, T is the temperature and *r* is the differential resistance of the device in the dark. The calculated *S_i(thermal*) at room temperature (300K) is about 5.00×10⁻²⁷ A²/Hz.



Fig. S8 Spectral density of 1/f noise for the MA-based device at -0.1V.



Fig. S9 Specific detectivity of the PVSC fabricated by MA process as the function of incident wavelength.



Fig. S10 Photocurrent of MA-based device under the illumination of white light with various intensities at -0.1V bias.



Fig. S11 Device stability of the corresponding unencapsulated 2D, 3D PVSC in air with different humidity.

Method	τ ₁ (ns)	frac. τ ₁ (%)	τ ₂ (ns)	frac. τ ₂ (%)	Lifetime (ns)
CA	0.97	21.99	12.67	78.01	10.10
MA	1.51	49.28	7.77	50.72	4.68

Table S1. Time-resolved PL data of (PEA)₂(CH₃NH₃)₄Pb₅I₁₆ perovskite films on ITO/PEDOT:PSS substrates.