Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting information

Enhancing Two-dimensional Perovskite Photodetector Performance through Balancing Carrier Density and Directional Transport

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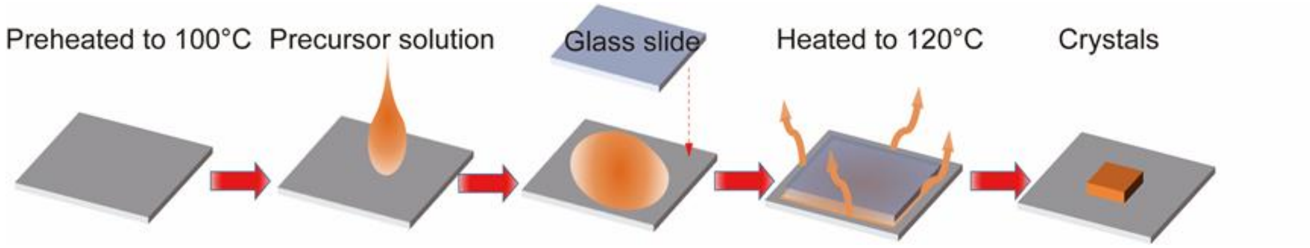

Fig. S1 Schematic illustration of the in situ inverse temperature crystallization method to grow 2D layered $PEA_2MA_{n-1}Pb_nI_{3n+1}$ single crystals.

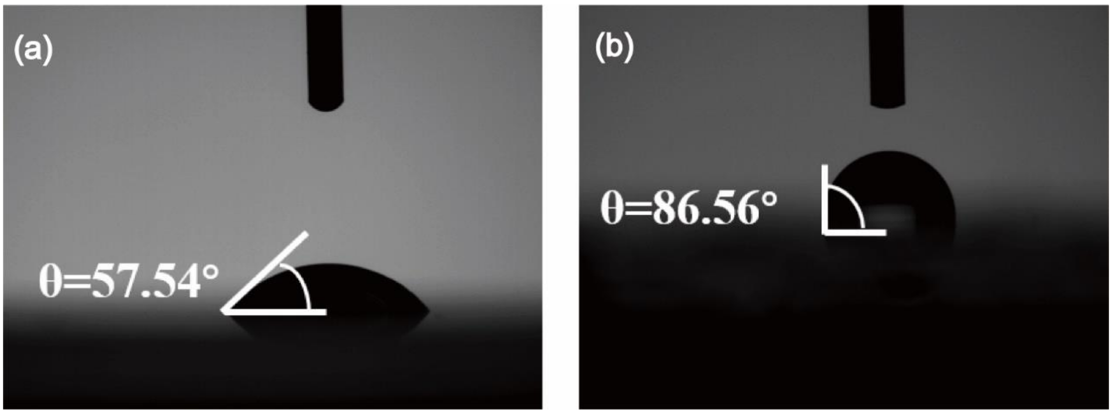

Fig. S2. Comparison of the hydrophobicity with glass substrates. (a) Substrate without hydrophobic treatment. (b) Substrate after hydrophobic treatment.

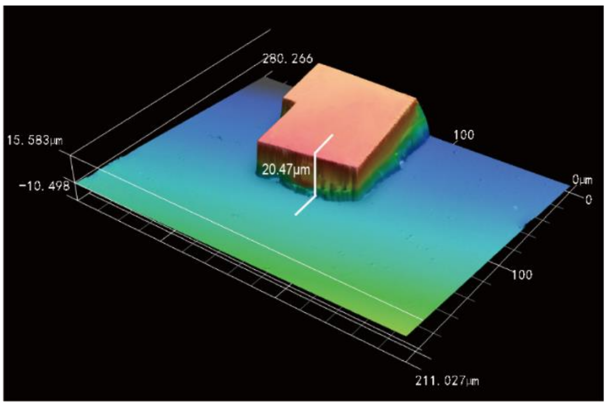

Fig. S3. 3D pseudo color plot of the $PEA_2MA_3Pb_4I_{13}$ single crystals.

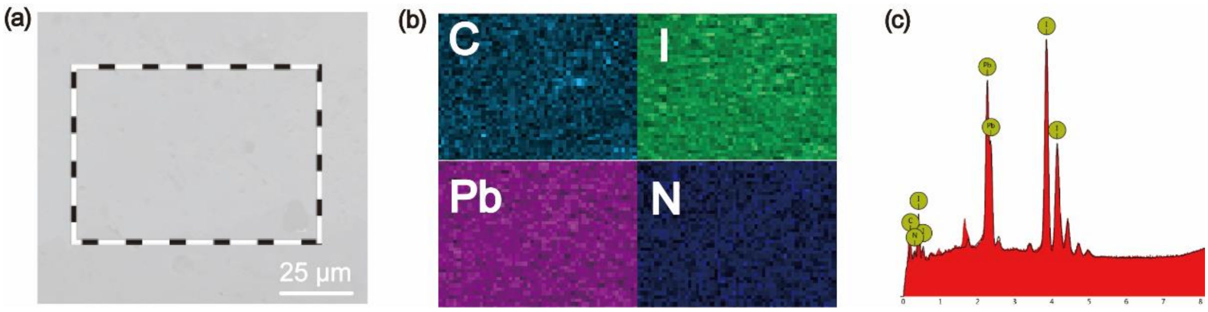

Fig. S4 (a) SEM image of $(PEA)_2(MA)_3Pb_4I_{13}$ single crystal surface. (b) EDS mapping of $(PEA)_2(MA)_3Pb_4I_{13}$ single crystal, which shows uniform elemental distribution of carbon, lead, iodine and, nitrogen respective. (c) The EDS spectra of selected area.

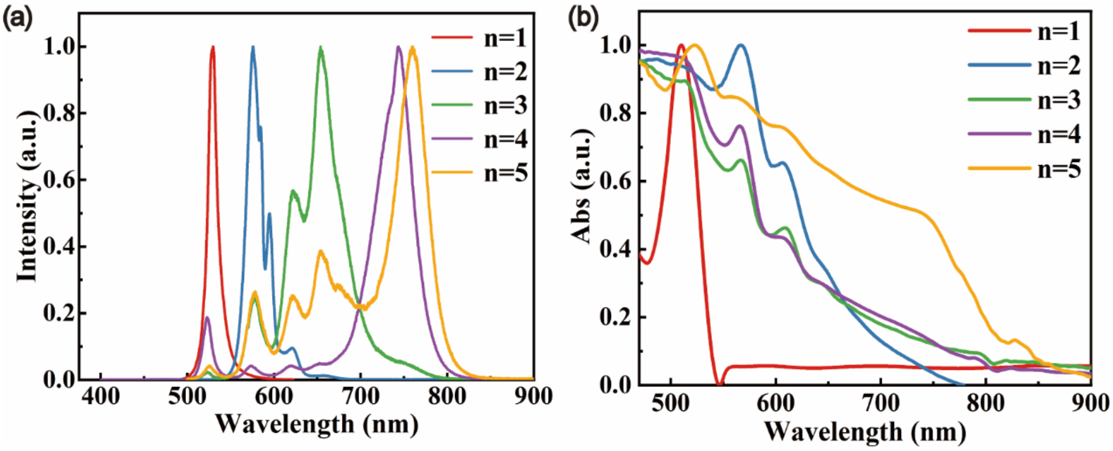

Fig. S5 (a) Photoluminescence spectra and (b) absorption spectra of 2D perovskite polycrystalline films with different layers.

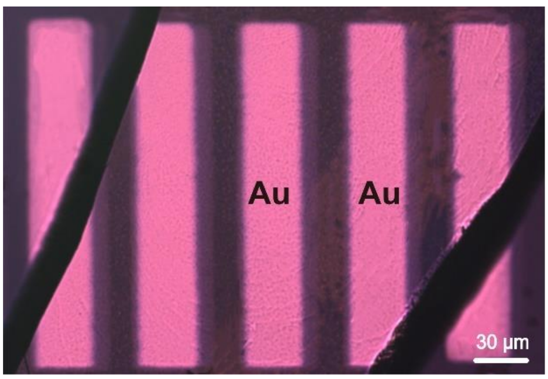

Fig. S6 The optical image of Au electrodes evaporated on the surface of crystal.

Table S1 TRPL fitting data of 2D perovskite with different layers.

n values	**p_1 (%)**	**t_1 (ns)**	**p_2 (%)**	**t_2 (ns)**	**t_{ave} (ns)**
n=1	22.77	0.118	77.23	0.516	0.425
n=2	57.37	0.918	42.63	5.348	2.807
n=3	58.2	1.781	41.8	6.584	3.789
n=4	56.67	1.204	43.33	12.398	6.054
n=5	81.77	2.497	18.23	59.944	12.967

Note: $t_{ave}= p_1 \times t_1 + p_2 \times t_2$, among which t_{ave} is the average carrier lifetime; t_1, t_2 are the carrier lifetimes; and f_1,f_2 are the fractional contributions.

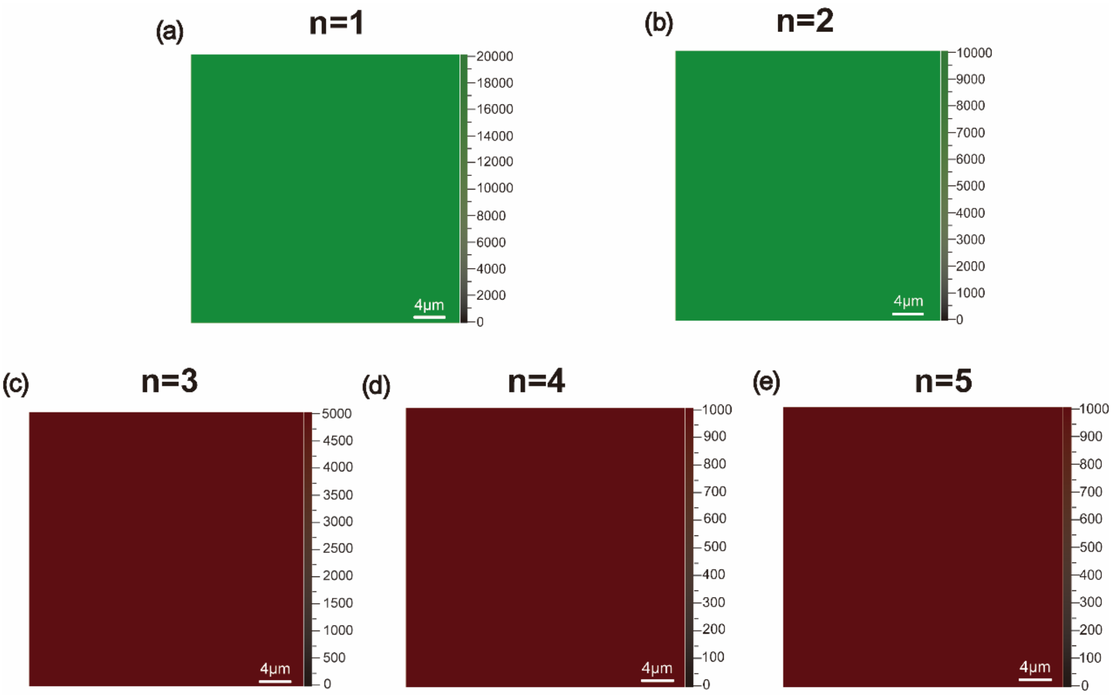

Fig. S7 (a)-(e) PL mapping intensity of 2D single crystals (n=1-5) with the excitation of 473 nm laser.

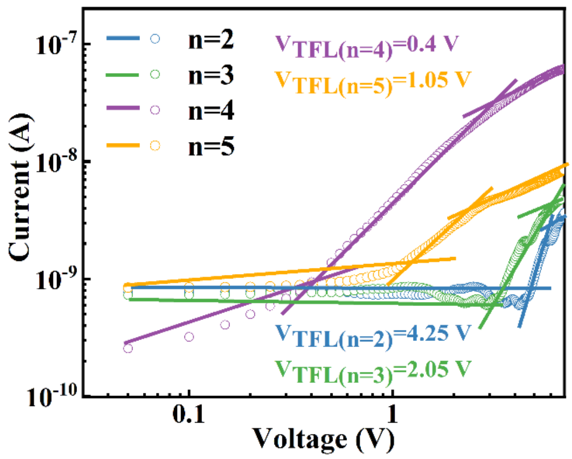

Fig. S8 SCLC curves of 2D single crystals.

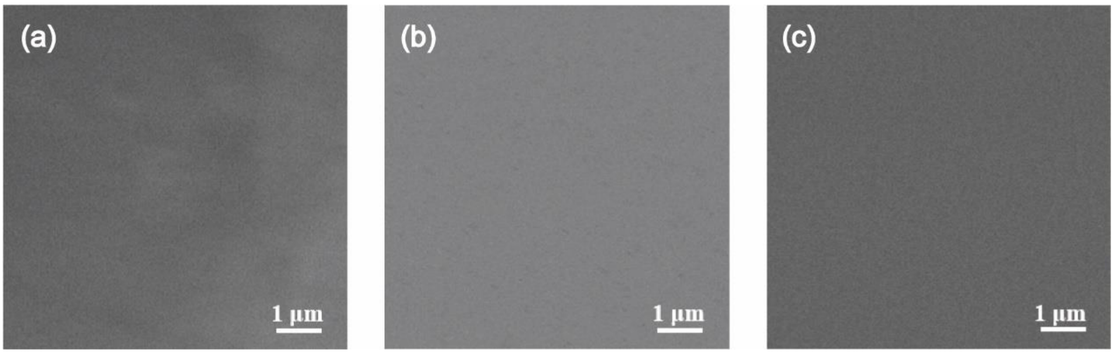

Fig. S9 SEM images of 2D perovskite single crystals with different layers. (a) n=2. (b) n=3. (c) n=5.

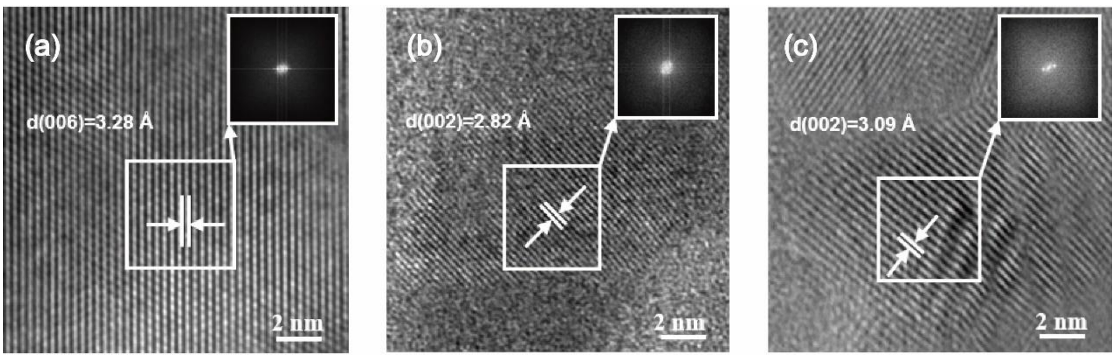

Fig. S10 HRTEM images of 2D perovskite single crystals with different layers. The embedded images are the FFT transform of the selected area. (a) n=2. (b) n=3. (c) n=5.

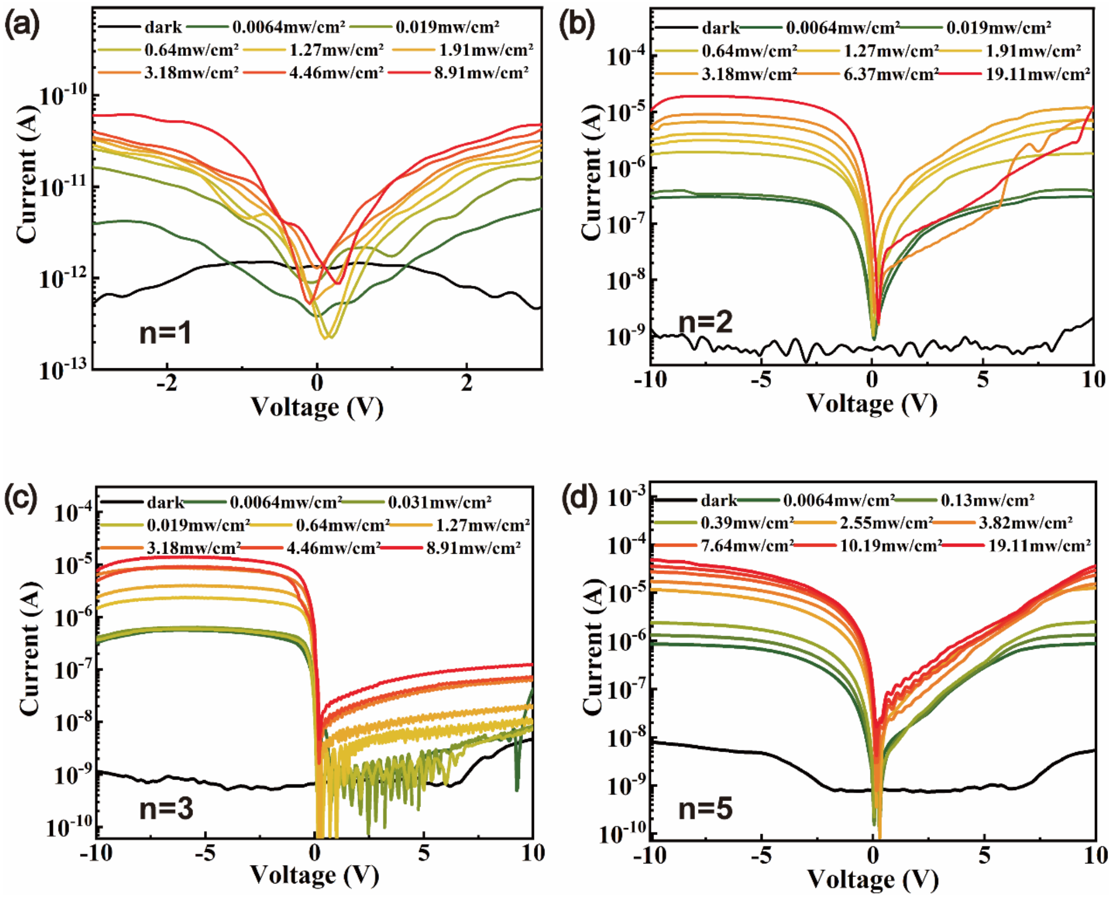

Fig. S11 The current–voltage (I–V) curves in dark and under 532 nm light illumination with varying light intensities for (a) n=1, (b) n=2, (c) n=3 and (d) n=5.

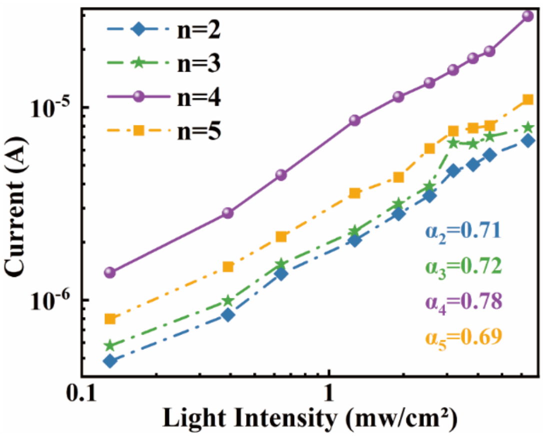

Fig. S12 Photocurrent values of the corresponding devices versus light intensity on a double-logarithmic scale for n=2,3,4 and 5. The bias is under 3V.

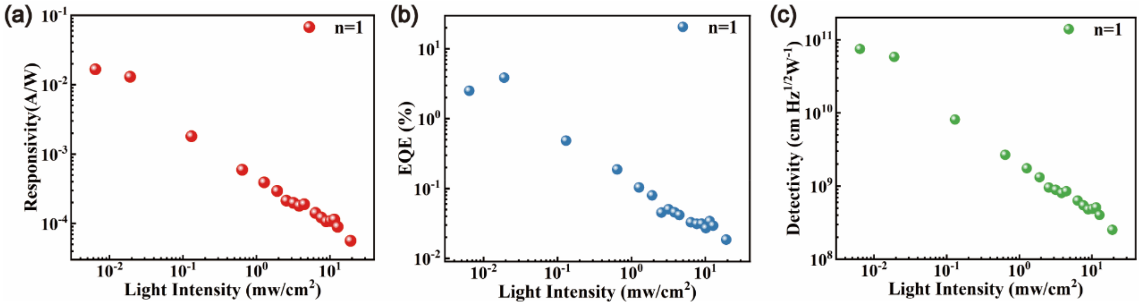

Fig. S13 (a) Responsivity, (b) EQE and (c) detectivity as a function of incident light intensity for n=1.

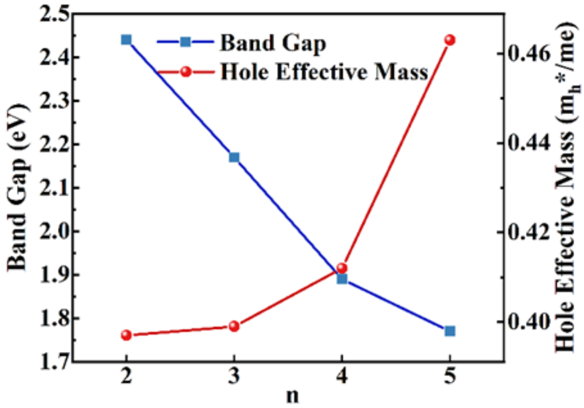

Fig. S14 The band gap and hole effective mass curves related to the number of layers.

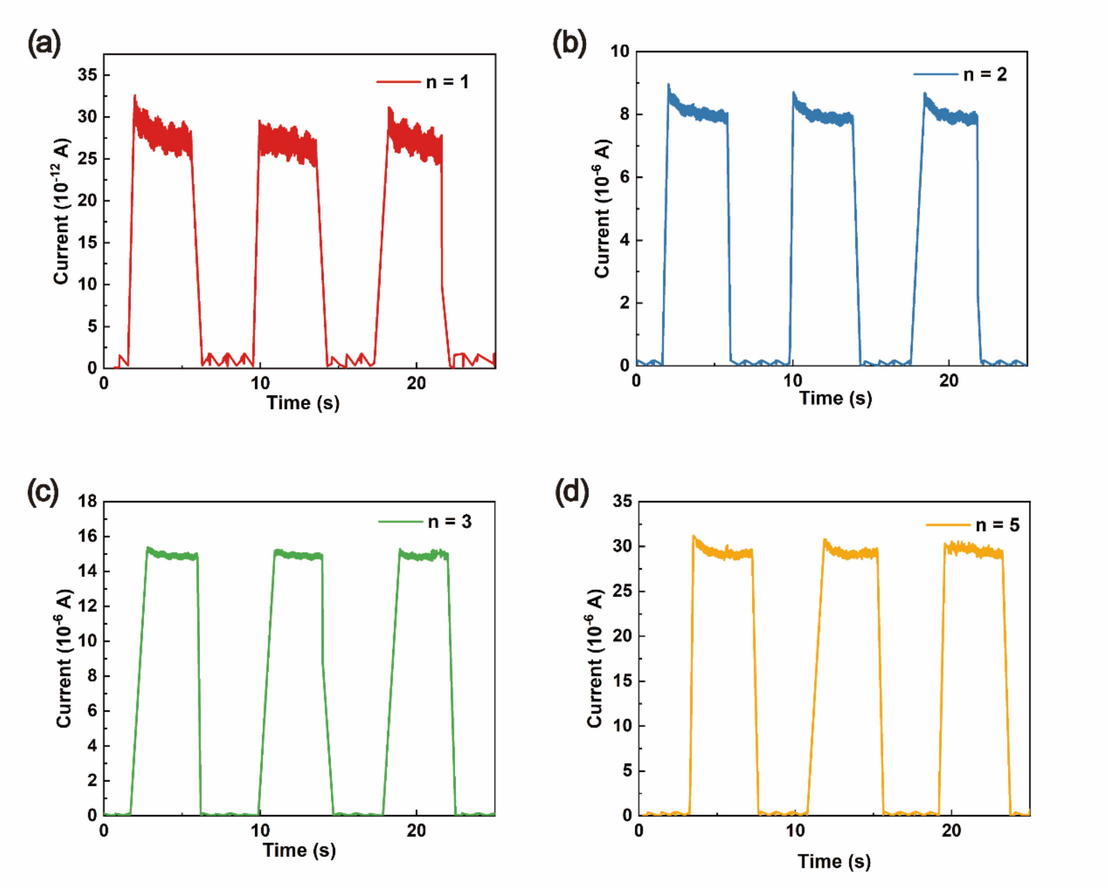

Fig. S15 temporal photocurrent response of 2D perovskite photodetectors with varying layers (a)-(d) n = 1, 2, 3, 5.

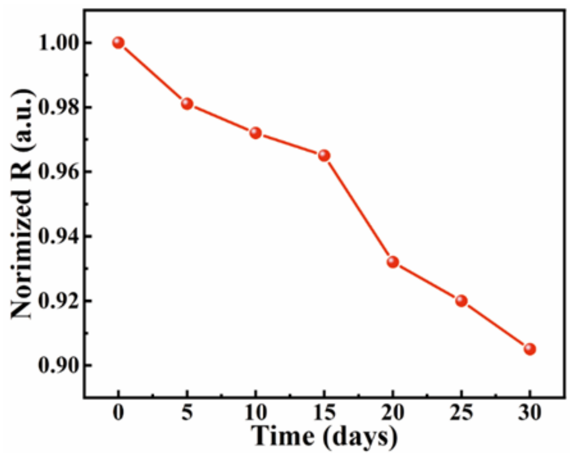

Fig. S16 Stability of $(PEA)_2(MA)_3Pb_4I_{13}$ single crystals photodetector.

Table S2. Performance comparison of 2D perovskite-based photodetectors.

Material	Modify	Light Power /Wavelength	Bias (V)	R (A/W)	EQE (%)	D (Jones)	Ref.
$(PEA)_2PbI_4$	No	0.08 μW cm^{-2}/460 nm	4	98.17	2.65×10^5	1.62×10^{15}	***Nat. Com.*** 2018,9,5302
$(PEA)_2PbI_4$	No	10 μW cm^{-2}/460 nm	5	139.6	3.77×10^5	1.89×10^{15}	***Matter*** 2019,2,465
$(PA)_2(FA)Pb_2I_7$	No	46 μW cm^{-2}/637 nm	10	----	----	1.73×10^{14}	***Small*** 2019,15, 1901194
$BDAPbI_4$	No	0.0088 μW cm^{-2}/637 nm	10	0.927	----	1.23×10^{11}	***Small*** 2020,16,2003145
$(PEA)_2(MA)_2Pb_3I_{10}$	No	0.038 μW cm^{-2}/598 nm	9	149	----	2×10^{12}	***Adv.Sci.***2020,7, 2000776
$(PEA)_2(MA)_3Pb_4I_{13}$	PCBM	2.5 μW cm^{-2}/570 nm	----	0.44	----	3.38×10^{12}	***Adv. Mater.***2021,33, 2101714
$(BA)_2FAPb_2I_7$	No	3.5 μW cm^{-2}/488 nm	10	5	----	3.5×10^{11}	***Adv. Funt. Mater.***2021, 2112277
$(CH)_2(MA)_3Pb_4I_{13}$	FTO	1.5 mW cm^{-2}/470 nm	3	0.046	----	----	***Sci.Rep.***2022,12, 2176
$(PEA)_2(MA)_3Pb_4I_{13}$	**No**	**6.4 μW cm^{-2}/532 nm**	**3**	**3077**	**7.19×10^6**	**2.23×10^{12}**	**This work**