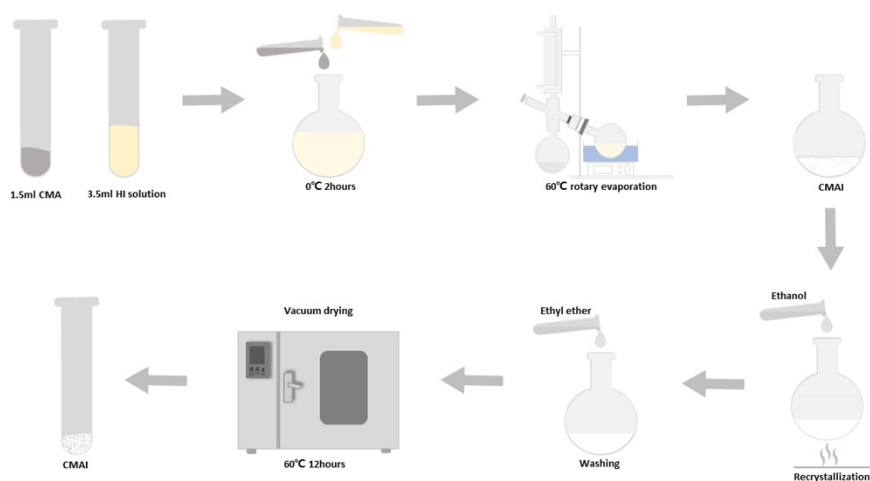


# Supporting Information for

## Enhanced Photovoltage and Stability of Perovskite Photovoltaics Enabled by Cyclohexylmethylammonium Iodide-Based 2D Perovskite Passivation Layer

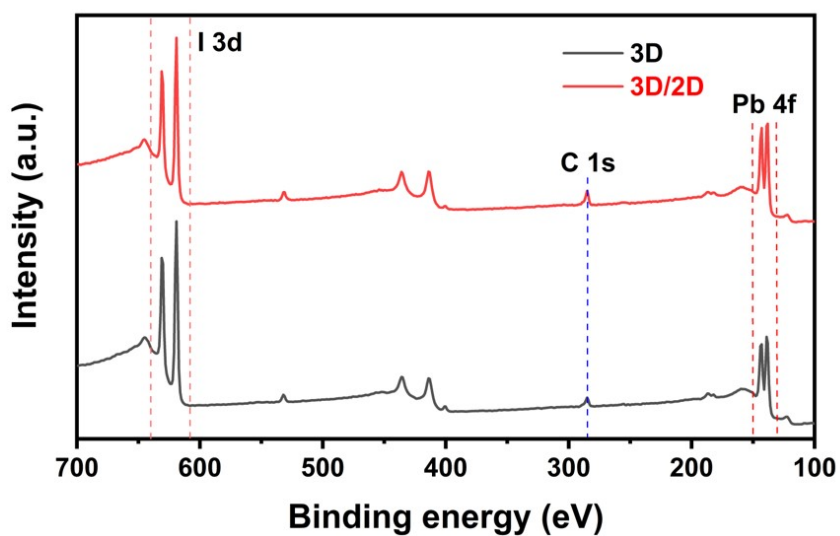
Weihai Sun<sup>a,\*</sup>, Jinjun Zou<sup>a,†</sup>, Xiaobing Wang<sup>a</sup>, Shibo Wang<sup>a</sup>, Yitian Du<sup>a</sup>, Fengxian Cao<sup>a</sup>, Zhang Lan<sup>a</sup>, Jihuai Wu<sup>a,\*</sup>, Peng Gao<sup>b</sup>

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<sup>b</sup> *Fujian Institute of Research on Structure Matter, CAS, Xiamen 361021, Fujian, China*

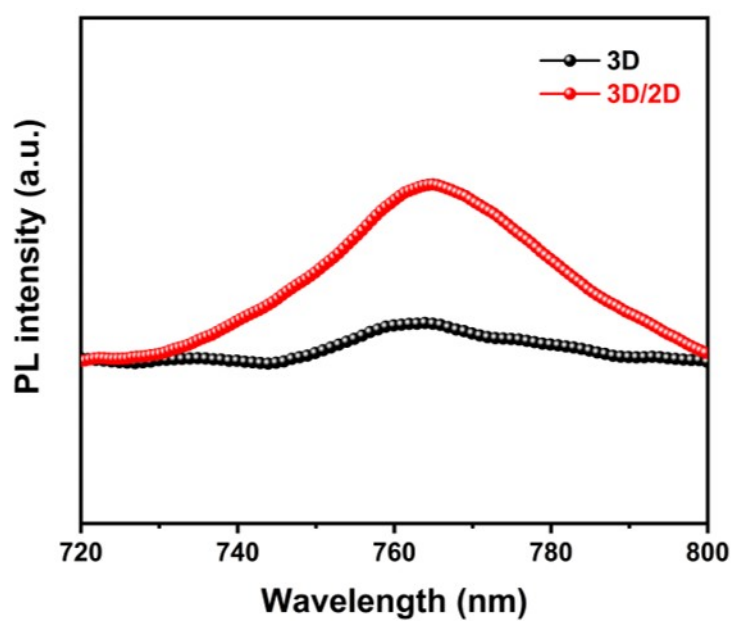


**Fig. S1** Schematic illustration of the synthesis of CMAI.

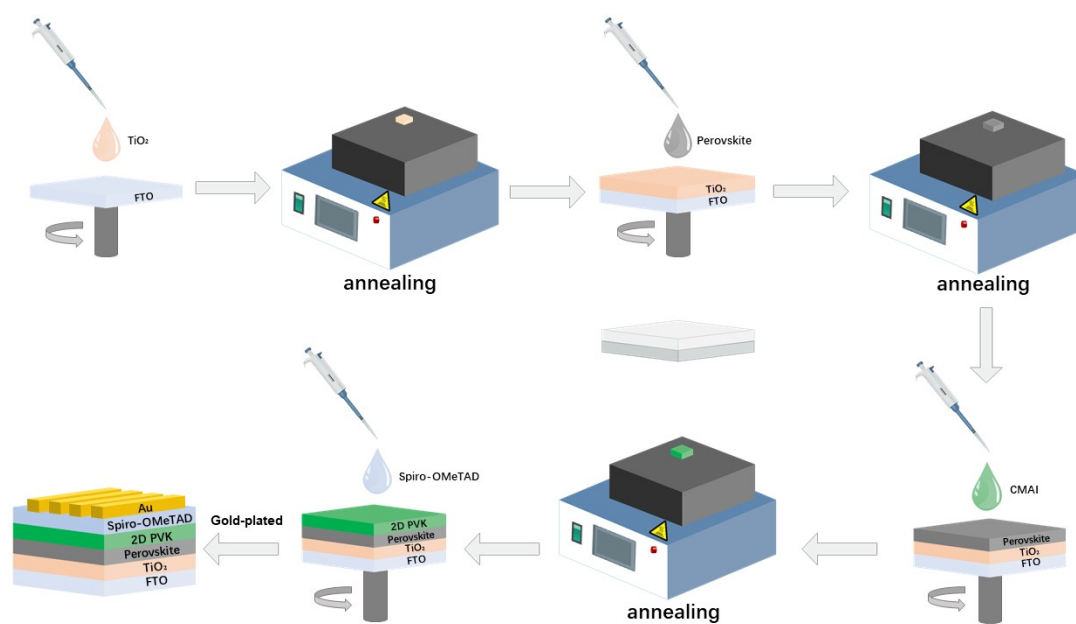
\* E-mail: [sunweihai@hqu.edu.cn](mailto:sunweihai@hqu.edu.cn); [jhwu@hqu.edu.cn](mailto:jhwu@hqu.edu.cn).



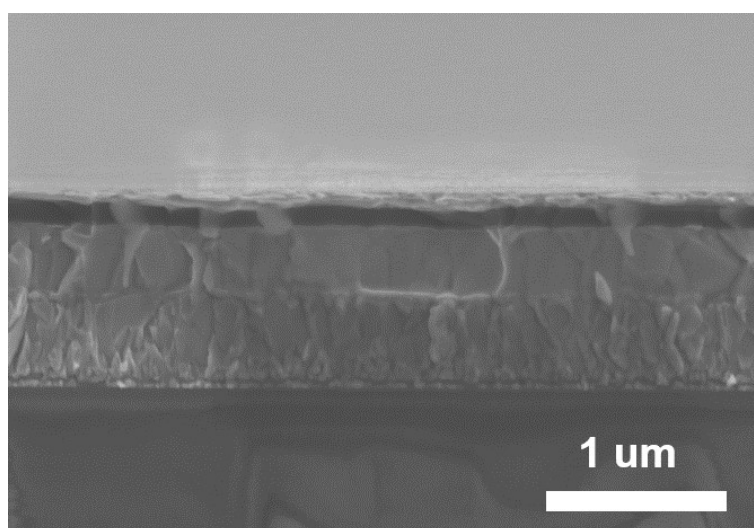
**Fig. S2** XPS survey spectra of the perovskite films with and without CMAI treatment.



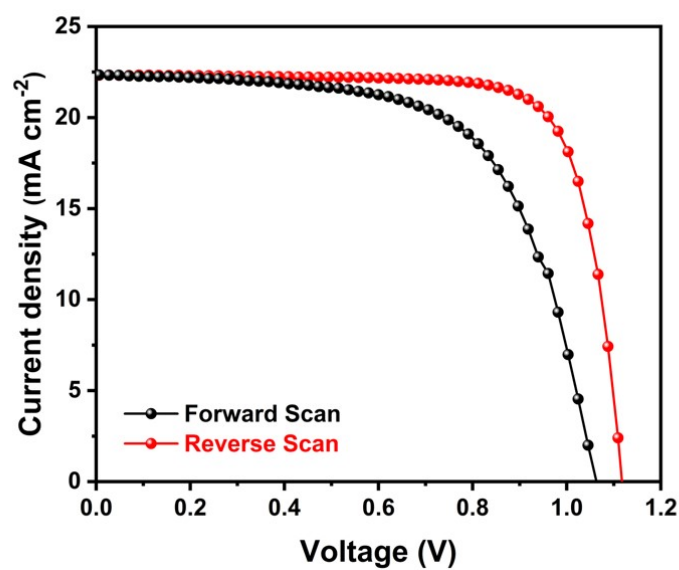
**Fig. S3** PL spectra of different perovskite films covered with HTL.



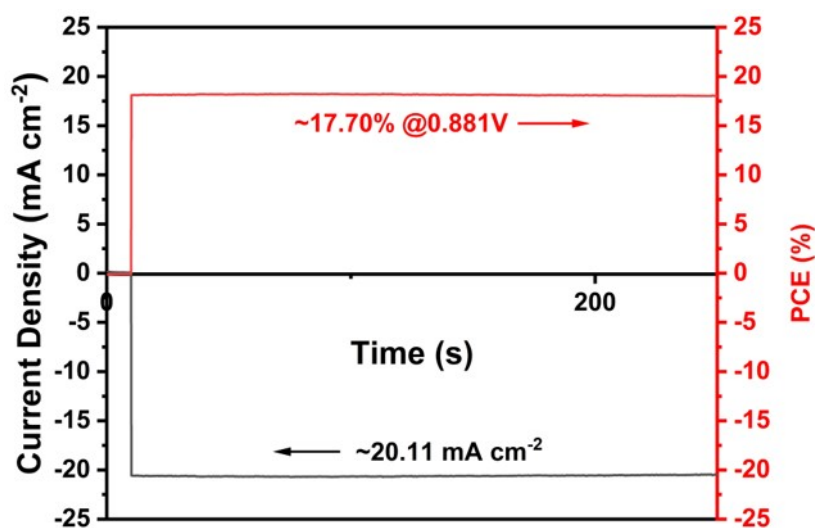
**Fig. S4** Schematic illustration of the fabrication of CMAI-treated PSCs.



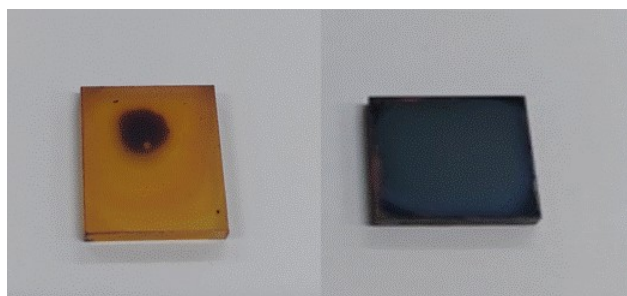
**Fig. S5** Cross-sectional SEM image of the 3D device.



**Fig. S6**  $J$ - $V$  curves of the 3D device under both forward and reverse scans.



**Fig. S7** The output of current densities and corresponding efficiencies of the pristine device by holding the voltage at 0.88 V.



**Fig. S8** Photographs of 3D perovskite film (left) and 3D/2D perovskite film (right) after 35 days storage.

**Table S1.** TRPL data of 3D and 3D/2D perovskite films on FTO substrates.

samples	$\tau_{\text{ave}}$ (ns)	$A_1$ (%)	$\tau_1$ (ns)	$A_2$ (%)	$\tau_2$ (ns)
3D	68.88	20.67	9.84	79.33	84.27
3D/2D	88.10	11.22	13.16	88.78	97.57

**Table S2.** Photovoltaic data of 3D and 3D/2D PSCs with different scan direction.

Device	Scan direction	$V_{\text{OC}}$ (V)	$J_{\text{SC}}$ (mA cm <sup>-2</sup> )	FF	PCE (%)	HI (%)
3D	Reverse	1.12	22.33	0.77	19.26	21.60
	Forward	1.06	22.26	0.64	15.10	
3D/2D	Reverse	1.19	23.18	0.80	22.06	4.99
	Forward	1.16	23.17	0.78	20.96	