Supporting Information

Electric-Field Assisted Perovskite Crystallization for High-Performance Solar Cells

Cong-Cong Zhang^a, Zhao-Kui Wang^{*b}, Meng Li^b, Zhi-Yong Liu^a, Ji-En Yang^a, Ying-Guo Yang^c, Xing-Yu Gao^c, and Heng Ma^{*a}

^{*a*} Henan Province Key Laboratory of Photovoltaic Materials & College of Physics & Materials Science, Henan Normal University, Xinxiang 453007, China. *E-mail: <u>hengma@henannu.edu.cn</u> (H. Ma);*

^b Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices, Institute of Functional Nano & Soft Materials (FUNSOM), Collaborative Innovation Center of Suzhou Nano Science and Technology, Soochow University, Suzhou, Jiangsu 215123, China. E-mail: zkwang@suda.edu.cn (Z.K.Wang)

^c Shanghai Synchrotron Radiation Facility, Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201204, China.

<u>Corresponding Authors:</u> hengma@henannu.edu.cn (H. Ma); zkwang@suda.edu.cn (Z.K.Wang)

Keywords: Perovskite solar cells; Electric-field assisted annealing; Ion migration; Ferroelectricity

Table of Contents

Figure S1. The construction of EEF system during the annealing treatment.

Figure S2. The perovskite films photograph taken during annealing process.

Figure S3. The schematic of the GIXRD and the integrated intensity plots around (110) plane along different direction.

Figure S4. *J-V* curves of PSCs with and without EEF treatment with a regular structure device.

Table S1. Detailed photostatic parameters of PSCs in Fig. S3.

Figure S5. The statistical average values and standard deviations of parameters.

Figure S6. The statistical histograms of PCE based on different treatments.

Table S2. The parameters of *J-V* hysteresis on PSCs with different annealing process.

Figure S7. The statistical IPCE histogram of PSCs with and without EEF treatment.

Figure S8. IPCE of PSCs with EEF-assist and thermal-annealing-only process.

Figure S9. Surface CPD patterns obtained by KPFM of different PSCs.

.



Figure S1. (a) The construction of measuring the spacing. (b) The construction of EEF system during the annealing treatment. (c) The energy diagram in perovskite solar cells.

Measurement the spacing



Figure S2. The perovskite films photograph taken during annealing process per 20 min. (a) is the reference perovskite film, and (b) is the 2.5 V/ μ m (up or down)-EEF based perovskite film.



Figure S3. (a) The schematic of the GIXRD experimental setup for detecting the crystalline information of the perovskite films, where IOP, and IIP are the scattering beam in the direction of out-plane and in-plane, respectively. The integrated intensity plots around (110) plane along the (a) out-plane direction and (b) in-plane direction.



Figure S4. *J-V* curves of PSCs with and without EEF treatment with a regular structure of ITO/TiO₂/CH₃NH₃PbI₃/Spiro-MeOTAD/Ag.

Table S1. Detailed photovoltaic parameters of PSCs in Fig. S3.

	$V_{\rm OC}$ (V)	$J_{\rm SC}$ (mA/cm ²) FF		PCE (%)
2.5 V/µm (up)	1.09	22.80	0.77	19.18
Ref.	1.03	22.33	0.73	16.77
2.5 V/µm (down)	1.01	22.13	0.74	16.57



Figure S5. The statistical curves of average values and standard deviations of (a) V_{OC} , (b) J_{SC} , (c) FF and (d) PCE.



Figure S6. The statistical histograms of PCE based on (a) the reference devices, (b) 1.0 V/ μ m (up), (c) 2.5 V/ μ m (up), (d) 1.0 V/ μ m (down), and (e) 2.5 V/ μ m (down).

Device	Scan direction	$V_{\rm OC}$ (V)	J _{SC} (mA/cm2)	FF (%)	PCE (%)
2.5 V/µm (up)	Forward	0.98	22.64	0.78	17.26
	Reverse	0.98	22.60	0.77	17.02
	Average	0.98	22.62	0.78	17.14
Ref.	Forward	0.93	21.80	0.75	15.33
	Reverse	0.93	21.62	0.73	14.68
	Average	0.93	21.71	0.74	15.01
2.5 V/µm (down)	Forward	0.92	21.90	0.75	15.18
	Reverse	0.94	21.98	0.75	15.49
	Average	0.93	21.94	0.75	15.34

 Table S2. Parameters of *J-V* hysteresis on PSCs with different annealing process by fitting linear part in Fig. 4 (d).



Figure S7. The statistical IPCE histogram of PSCs with and without EEF treatment.



Figure S8. IPCE of PSCs with EEF-assist and thermal-annealing-only process.



Figure S9. Surface CPD patterns obtained by KPFM based on (a-c) $CH_3NH_3PbI_{3-x}Cl_x$ and (d-f) $CH_3NH_3PbI_3$ films. (a, d) are after up-EEF treatment of 2.5 V/µm. (b, d) are the reference films. (c, d) are after down-EEF treatment of 2.5 V/µm. The inserts represent the statistic distributions CPD of different films. (g) The schematic diagram of KPFM to measure the surface CPD of the perovskite films.