Supporting Information

A Nanomesh Electrode for Self-Driven Perovskite Photodetectors with Tunable Asymmetric Schottky Junctions

Meng Zhang,[a,b,c] Youdi Hu,[a] Shuaiqi Wang,[a] Yaru Li,[a] Chunwu Wang,[d] Ke Meng,[a] and Gang Chen*[a,b,c]

[a] M. Zhang, Y. Hu, S. Wang, Y. Li, K. Meng, Prof. G. Chen
School of Physical Science and Technology
ShanghaiTech University
Shanghai 201210, China
E-mail: gchen@shanghaitech.edu.cn

[b] M. Zhang, Prof. G. Chen
Shanghai Advanced Research Institute
Chinese Academy of Sciences
Shanghai 201210, China

[c] M. Zhang, Prof. G. Chen
University of Chinese Academy of Sciences
Beijing 100049, China

[d] C. Wang
School of Environment and Architecture
University of Shanghai for Science and Technology
Shanghai 200093, China

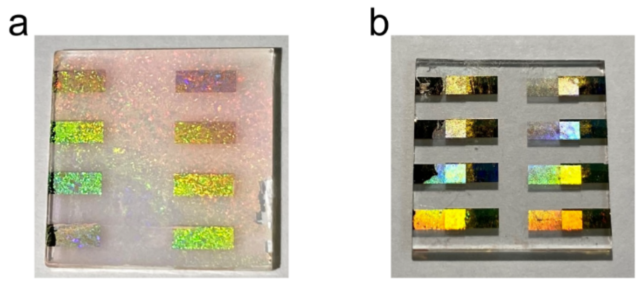

Figure S1. The photographs of (a) the bottom electrode fully covered by the PS-MS monolayer and (b) the final nanomesh electrode.

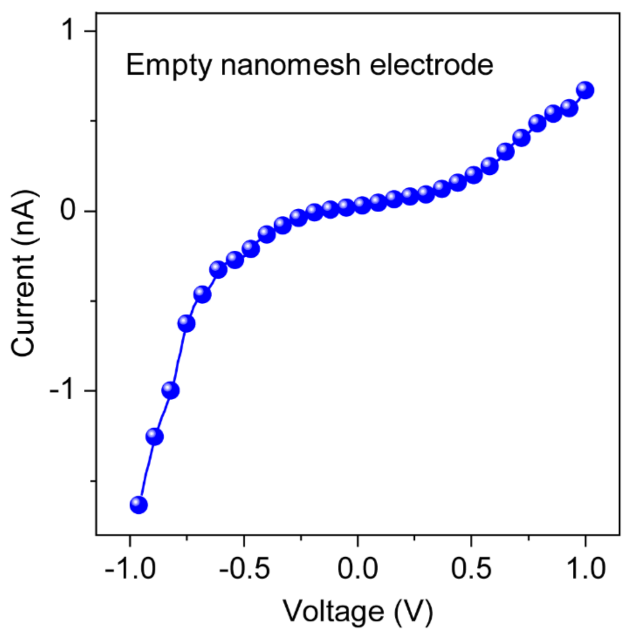

Figure S2. The *I*-*V* curve of the empty nanomesh electrode measured by scanning the voltage from +1 V to -1 V.

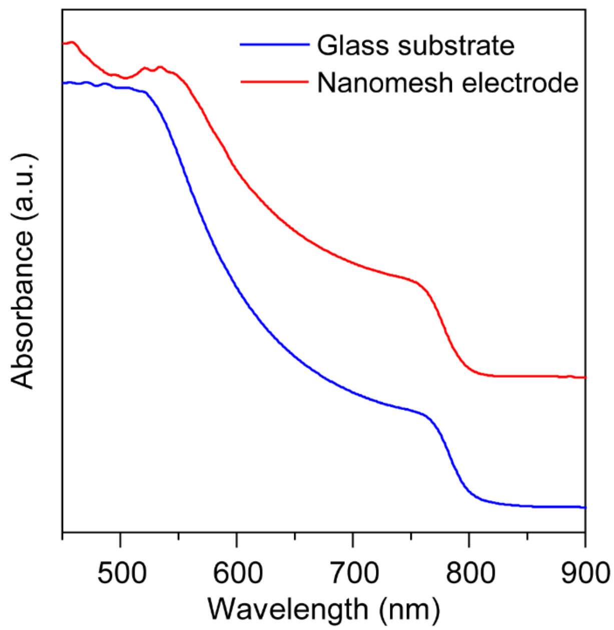

Figure S3. The absorption spectra of the perovskite films on the nanomesh electrode and the glass substrate.

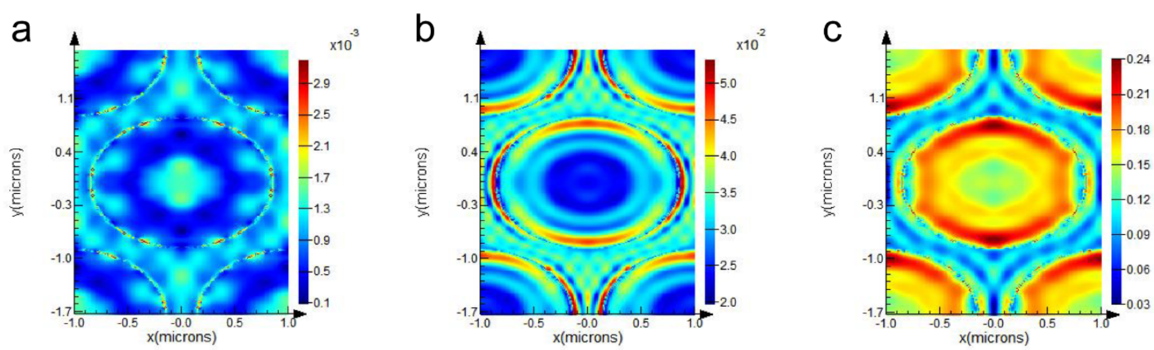

Figure S4. The electric field intensity profiles $|\vec{E}|^2$ at the nanomesh electrode simulated for the light wavelengths of (a) 530 nm, (b) 640 nm and (c) 800 nm.

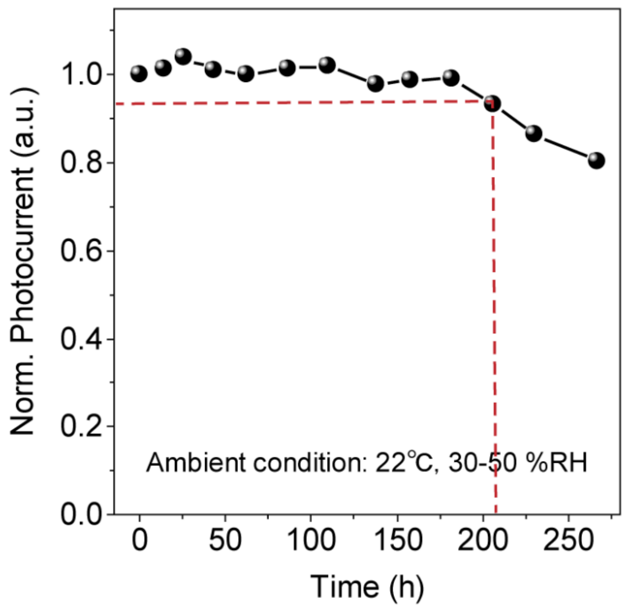

Figure S5. Photocurrent changes of the photodetector being placed in an environment of 30-50% relative humidity.

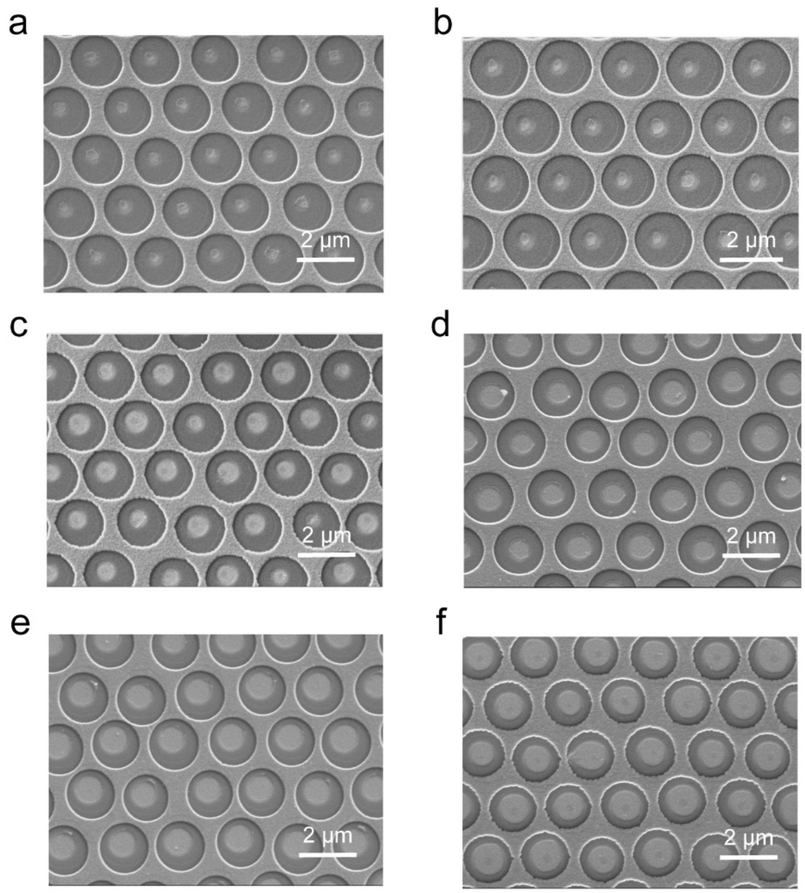

Figure S6. The SEM images of the nanomesh electrodes prepared at T_a of (a) 90 ℃, (b) 95 ℃, (c) 100 ℃, (d) 105 ℃, (e) 110 ℃, (f) 115 ℃.

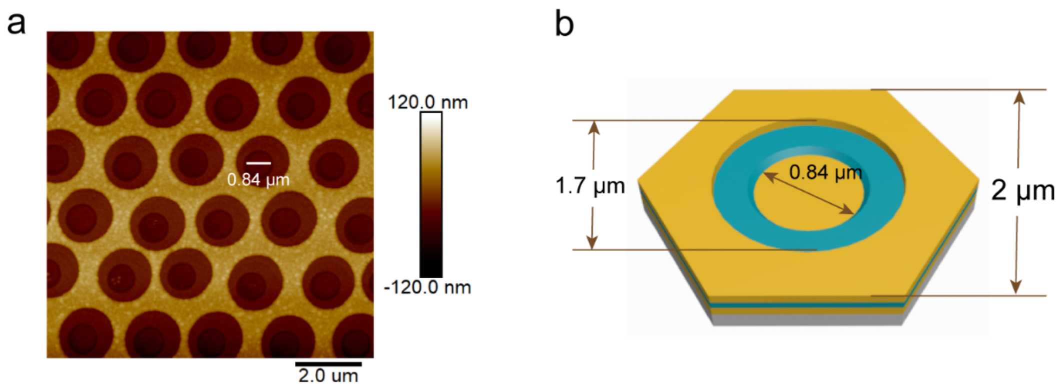

Figure S7. (a) AFM topography of the nanomesh electrode at T_a of 105 ℃. (b) The structural parameters of a single nanoelectrode.

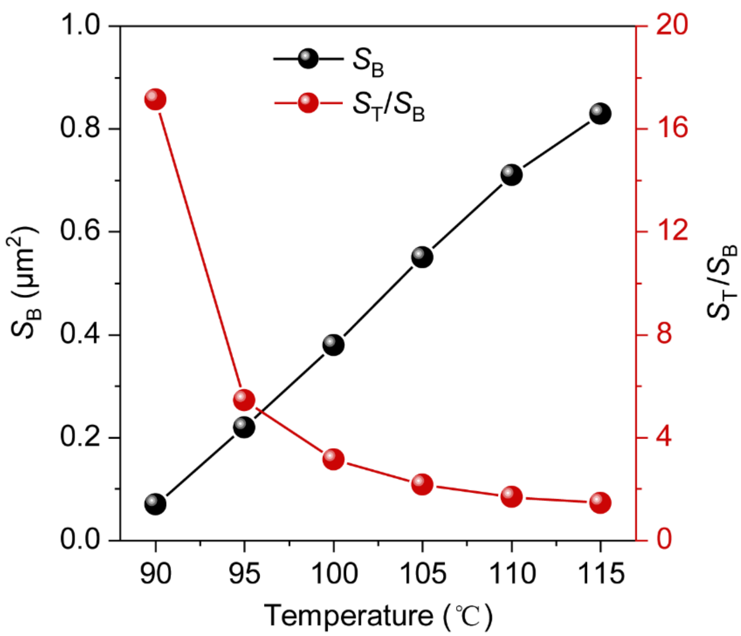

Figure S8. The area of the bottom electrode (S_B) and the areal ratio of the top and bottom electrodes (S_T/S_B) plotted as a function of T_a.

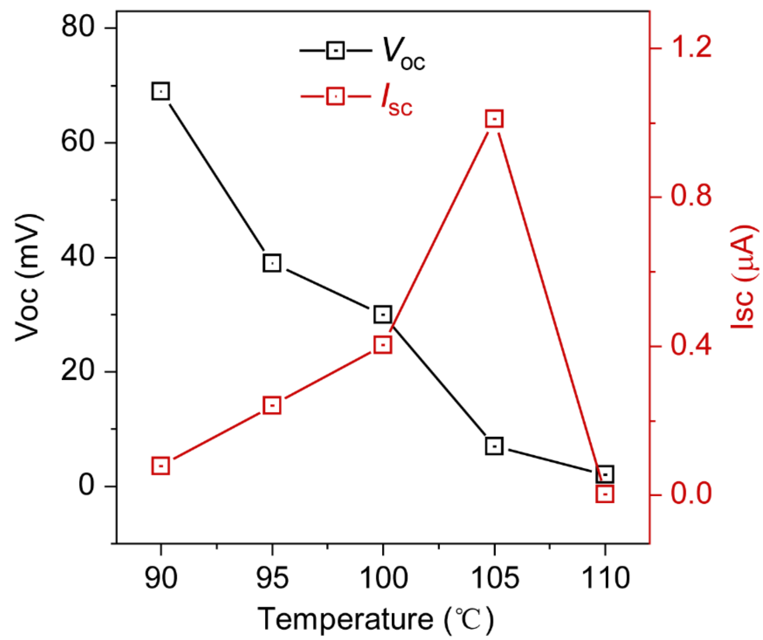

Figure S9. The open-circuit voltage (V_{oc}) and short-circuit current (I_{sc}) of the nanomesh photodetectors plotted as a function of T_a.

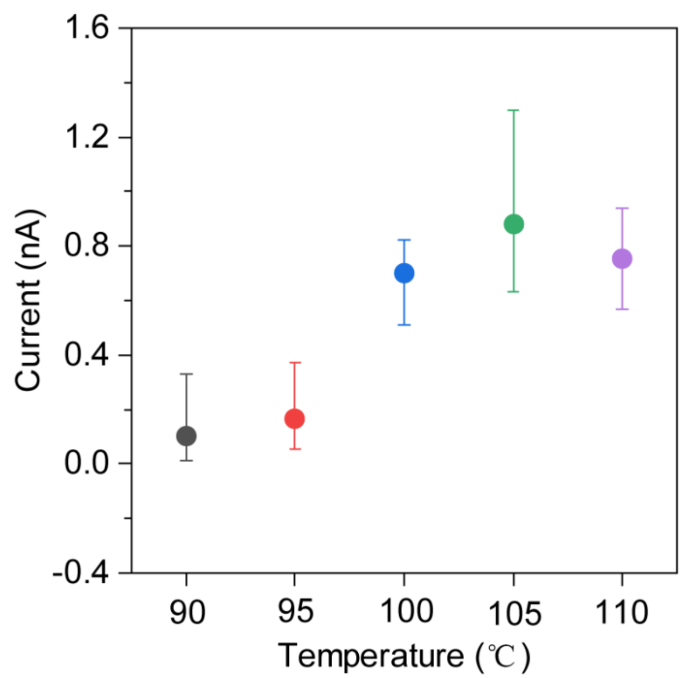

Figure S10. The statistical dark current distributions for photodetectors based on the nanomesh electrodes prepared at different PS-MS annealing temperatures.

Supplementary Note 1

Photoresponsivity (*R*) and specific detectivity (*D):** *R* is defined as the photocurrent generated per unit power of the incident light on the effective area which is obtained by the equation[1,2]

$$R = \frac{I_{ph} - I_d}{P_{opt}\, S} \tag{1}$$

where I_{ph}, I_d, P_{opt}, and S are the photocurrent, dark current, the incident-light intensity and the effective illuminated area, respectively. The spot size of the light source is large enough to cover the active area of the device of 1 mm^2. D^* is used to characterize the sensitivity of a photodetector and represents the ability of a detector to detect weak optical signals, which is defined as

$$D^* = \frac{\sqrt{S}\, R}{\sqrt{2qI_d}} \tag{2}$$

where q is the absolute value of the elementary charge, and R is the responsivity.

Supplementary Note 2

The Schottky barrier height variation at the junction in the nanomesh photodetector: Under light illumination, the carrier concentration and the energy levels in the semiconductor can be expressed as[3]

$$E_c - E_F = kT \ln \frac{N_c}{n} \tag{3}$$

$$N_c = 2\frac{(2\pi m_n^* kT)^{3/2}}{h^3} \tag{4}$$

where E_c is the conduction band, E_F is the electronic quasi-Fermi level, k is the Boltzmann constant, T is the absolute temperature, N_c is the effective state density of the conduction band, n is the electron concentration, m_n^* is the effective mass, and h is the Planck constant. Under the light illumination, the electron concentration increases at the Schottky junction interface and the Fermi level becomes closer to the conduction band, so the Schottky junction barrier at the semiconductor side with high electron concentration decreases, the asymmetric Schottky barriers are created in the nanomesh perovskite photodetector.

Table S1. The opening diameters of the top and bottom nanoelectrodes (D_T and D_B), the areas of the top and bottom nanoelectrodes (S_T and S_B), and their areal ratio (S_T/S_B) of the nanomesh electrodes prepared at different T_a.

T_a (°C)	90	95	100	105	110	115
D_T (μm)	0.7	0.7	0.7	0.7	0.7	0.7
S_T (μm^2)	1.2	1.2	1.2	1.2	1.2	1.2
D_B (μm)	0.29	0.53	0.70	0.84	0.95	1.03
S_B (μm^2)	0.07	0.22	0.38	0.55	0.71	0.83
S_T/S_B	17.14	5.45	3.16	2.18	1.69	1.45

References

[1] M. Yang, J. Wang, Y. Zhao, L. He, C. Ji, X. Liu, H. Zhou, Z. Wu, X. Wang and Y. Jiang, *ACS Nano*, 2019, **13**, 755-763.

[2] M. Long, Y. Wang, P. Wang, X. Zhou, H. Xia, C. Luo, S. Huang, G. Zhang, H. Yan, Z. Fan, X. Wu, X. Chen, W. Lu and W. Hu, *ACS Nano*, 2019, **13**, 2511-2519.

[3] D. A. Neamen, *Semiconductor Physics and Devices: Basic Principles*, McGraw-Hill, New York 2012.