Supporting Information to

Rapid, stable and self-powered perovskite detectors via a fast chemical vapor deposition process

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Experimental Section

Methylamine iodide (MAI): Methylamine (33 wt % ethanol) and hydroiodic acid (HI, 45 wt % in water) were reacted in 250 mL three-neck flask at 0 °C for 3 h with continuous stirring by a magnetic stirrer. HI was added dropwise during the stirring process. After evaporated in a rotary evaporator at 60 °C for 3 h, and the white precipitation was then dissolved in ethanol and precipitated with the addition of 400 mL of diethyl ether. Finally, MAI (CH_3NH_3I) white powder was vacuum-dried for overnight.

Perovskite photovoltaic Devices: FTO glasses (15 Ω/sq) were washed with soap, acetone, ethanol and deionized water using ultrasonication. TiO_2 compacted layer was then deposited on the FTO substrates by spin-coating of titanium isopropoxide (750 μL in 5 mL ethanol) diluted in the acid ethanol solution and sintered at 500 °C in air and subsequently treated in 40 mM aqueous solution of $TiCl_4$ for 30 min at 70 °C. The compacted layer was formed after annealing at 500 °C for 30 min again. Lead iodide (PbI_2, 99.99%) poweder was dissolved in N,N-dimethylformamide (DMF, Aladdin) at a concentration of 460 mg mL^{-1} and stirred at 70 °C for 10 hours. 80 μL dissolved solution was spin-coated on FTO/ c-TiO_2 substrates at 4000 rpm for 30 s. After annealing at 60 ^{o}C for 30min, the PbI_2 substrates and MAI powder were placed in the furnace, as shown in Fig. 1a. The system was pumped to 10^{-3} Pa by turbo molecular pump, and using Ar as carrier gas, followed by subtracted heated to 100 ^{o}C, 120 ^{o}C, respectively. After reacted, the $CH_3NH_3PbI_3$ films were in-suit annealing for 30min in the Ar atmosphere in the furnace zone II. The hole-transport layer was spin-coated at 3500 rpm for 30 s as previous report.[1, 2] Finally, 120nm thick silver as cathode was deposited by thermal evaporation (~0.5 Å s^{-1}). The active area is 0.09 cm^2. All the processes were in relative humidity below 40%.

Characterization: The phase of products were identified by X-ray powder diffraction (XRD) utilizing a Cu Kα radiation with the scanning rang from 10° to 60°. The morphology of the samples was investigated by the field emission scanning electron microscopy (FE-SEM, Sigma Zeiss), atomic force microscopy (AFM, Dimension), respectively. The electrical characterization of the perovskite devices were conducted with a semiconductor parameter analyzer system (Keithley 4200-SCS). Noise current was mesured using a lock-in amplifier (SR830) in a dark room shielded with metal from 1Hz to 700Hz. To investigated the response speed, a home-built measurement system combining a laser diode (650 nm), oscilloscope (Tektronix, TDS2012B) and pulse generator was applied by modulated pulsed light illumination (100 Hz ~ 4 MHz). Photocurrent density–voltage (*J*–*V*) measurements of perovskite solar cells were carrid out on a Keithley 2636 system sourcemeter with a Xenon Lamp Solar Simulator equipped with a light intensity of ∼100 mW cm^{-2}. All the measurements were performed at zero bias and the room temperature with the relative humidity below 40%.

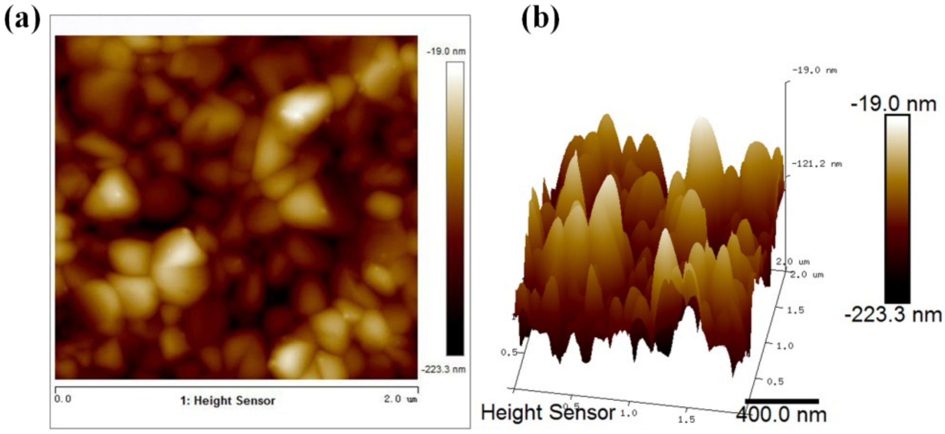

Fig. S1 The atomic force microscopy (AFM) graph of perovksite film (a) 2D and (b) 3D height image (2 × 2 μm).

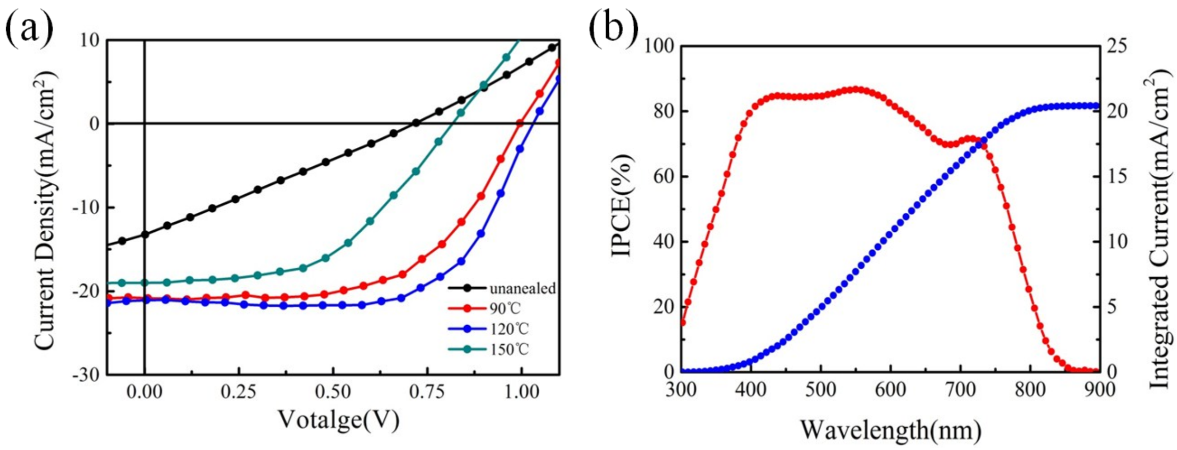

Fig. S2 (a) The current density–voltage (*J*–*V*) characteristic curves of the photovoltaic devices annealed at different temperatures under AM 1.5 irradiation. (b) the IPCE spectrum and the accumulative photocurrent density integral of the perovskite photovoltaic devices.

Table S1 Device performance parameters of perovskite photovoltaic devices.

Temperature [^{o}C]	V_{oc} [V]	J_{sc} [mA cm^{-2}]	FF	PCE (%)
unannealed	0.73	13.45	0.25	2.46
90	0.98	20.85	0.60	12.26
120	1.03	21.25	0.69	15.10
150	0.81	19.12	0.51	7.90

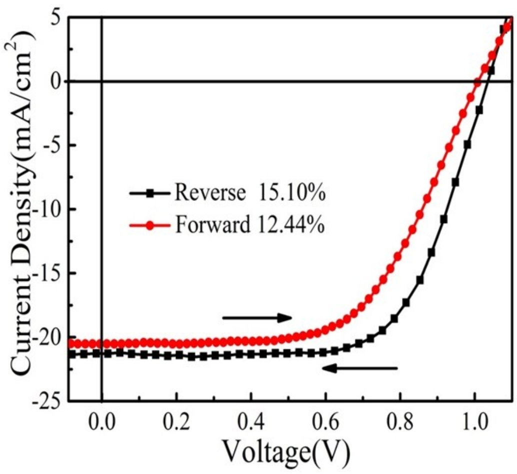

Fig. S3 *J*–*V* curves of perovskite photovoltaic device measured under forward and reverse scans.

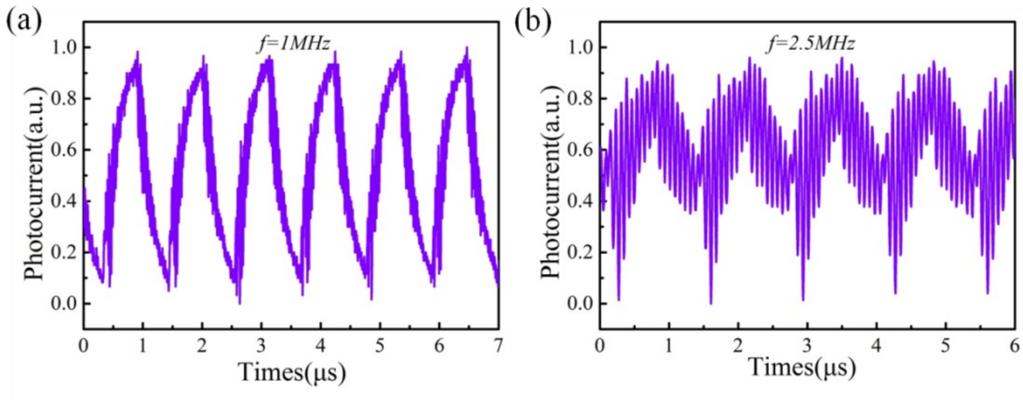

Fig. S4 Time response characteristics of the perovskite device under (a) 1 MHz and (b) 2.5 MHz pulsed light illumination.

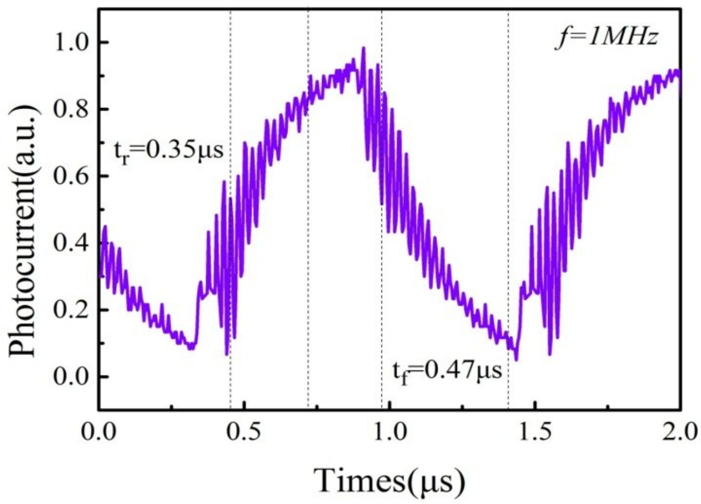

Fig. S5 The magnified and normalized plots of one response cycle 1 MHz.

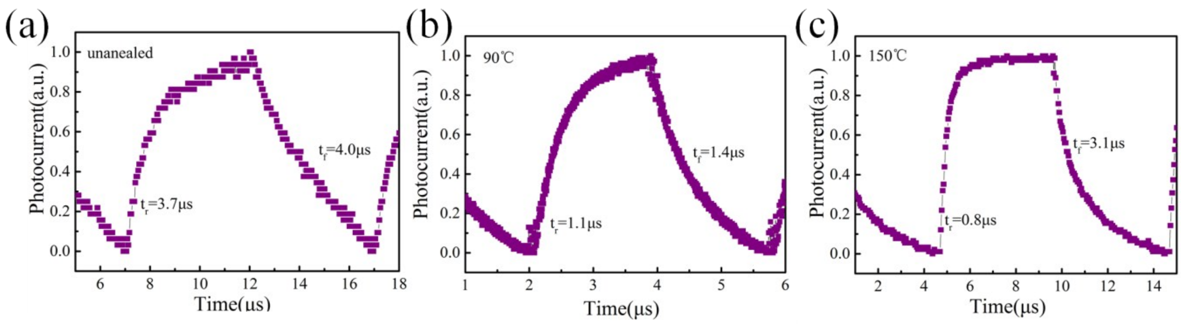

Fig. S6 The magnified and normalized plots of one response cycle 300 kHz with the annealing temperature at (a) unannealed; (b) 90^{o}C; (c)150 ^{o}C.

REFERENCE

1. N. J. Jeon, J. H. Noh, Y. C. Kim, W. S. Yang, S. Ryu and S. I. Seok, *Nat. Mater.*, 2014, **13**, 897-903.
2. C. Liu, J. Fan, X. Zhang, Y. Shen, L. Yang and Y. Mai, *ACS Appl Mater Interfaces*, 2015, **7**, 9066-9071.