Supporting information for

High-Performance Flexible and Self-Powered Perovskite Photodetector Enabled by Interfacial Strain Engineering

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Experimental section

Materials:

The etched PEN/ITO substrates for flexible photodetector were purchased from Advanced Election Technology CO., Ltd. Hole transport material-PEDOT:PSS (Clevios 4083) , electron transport material-$PC_{61}BM$, $CH_3NH_3PbI_{3-x}Cl_x$ precursors−methylammonium iodide (MAI), lead iodide (PbI_2) and lead chloride ($PbCl_2$) were purchased from Xi'an Polymer Light Technology Corp. All the organic solvents include γ-butyrolactone (GBL), Dimethyl sulfoxide (DMSO, 99.9%), 1,2-dichlorobenzene (ODCB, 99.9%), chlorobenzene (CB, 99.9%) were purchased from Sigma Aldrich. Hole transport material-PDCBT was purchased from 1-Material. All reagents and chemicals were used as received without further purification.

Perovskite precursor solutions:

$CH_3NH_3PbI_{3-x}Cl_x$ perovskite precursor solutions were prepared by mixing MAI, PbI_2 and $PbCl_2$ with a molar ratio of 1.4:1.25:0.15 in a mixed solvent of GBL: DMSO = 7:3 (v:v) with a slight amount of excessive PbI_2 and stirred at 55 °C for 12 hours in a N_2-filled glove box.

Sample preparation and device fabrication:

PEN/ITO substrates were sequentially cleaned by sonication with distilled water, isopropanol, ethanol and then treated with ultraviolet-ozone plasma before spin-coating of the PEDOT:PSS (Clevios 4083). The PEDOT:PSS (Clevios 4083) solution were spin-coated on the PEN/ITO substrates at 4000 rpm for 60 s, and then annealed on a hotplate at 120°C for 15 min. After depositing the PEDOT:PSS layer, an additional hole transport layer PDCBT was spin-coated at 5000 rpm for 50 s. In order to improve the wettability of the perovskite solution on the PDCBT layer, a modified layer was spin coated by a continuous two-step spin-coating method process at 4000 rpm for 20

and 10 s. During the first spin-coating step, 150 μL GBL was dripped on top of the PDCBT layer, and during the second spin-coating step, 50 μL CB was spin-coated. The perovskite layer was deposited by a continuous two-step spin-coating method process at 1000 rpm and 4000 rpm for 10 and 31 s, respectively. During the second spin-coating step, 400 μL CB was dripped on top of the perovskite film. Then, the flexible samples were annealed at 100°C for 10 min and cooled down to room temperature on a glass Petri dish. For the device fabrication, 60 μL $PC_{61}BM$ solution was deposited at 3000 rpm for 60 s on top of the perovskite film. Finally, it was sequentially deposited PEI (0.05 wt. % in isopropanol) layer at 5000 rpm for 30 s and 70 nm silver electrodes by vacuum thermal evaporation under a vacuum of 5×10^{-5} Pa. The working area of the device is 0.038 cm^2.

Device characterization:

Self-built quantum efficiency test system was used to test the responsivity and detectivity. The response speed was studied by an oscilloscope (MSO58, Tektronix) cooperated with a chopper. Surface morphologies of perovskite films were performed using SEM (JSM-6700F, JEOL). UV-vis spectra were conducted by a UV–vis spectrometer (UV-2600, Shimadzu). XRD measurements were obtained using an X-ray diffractometer (Empyrean, PANalytical). Self-built optical path for the linearly/circularly polarized photoexcitation-modulated photocurrent experiments is schematically shown in Figure S3. The excitation light source is 635 nm continuous-wave (CW) laser. The angle of quarter wave plate is adjusted to convert linearly polarized light and circularly polarized light, and the photocurrent are monitored by the source meter. Magneto-photocurrent measurements (Electrical transport measurement system, Model EM7, East Changing Technologies) were performed by recording photocurrent as a function of magnetic field. Steady and transient fluorescence

spectroscopic measurements were performed using a fluorescence spectrophotometer (Fluorolog-3, Horiba Scientific). Measurement of impedance spectrum was performed using impedance analyzer (E4990A, Keysight). The film thickness was measured by an ellipsometer (RC2-X, J.A. Woollam Co.).

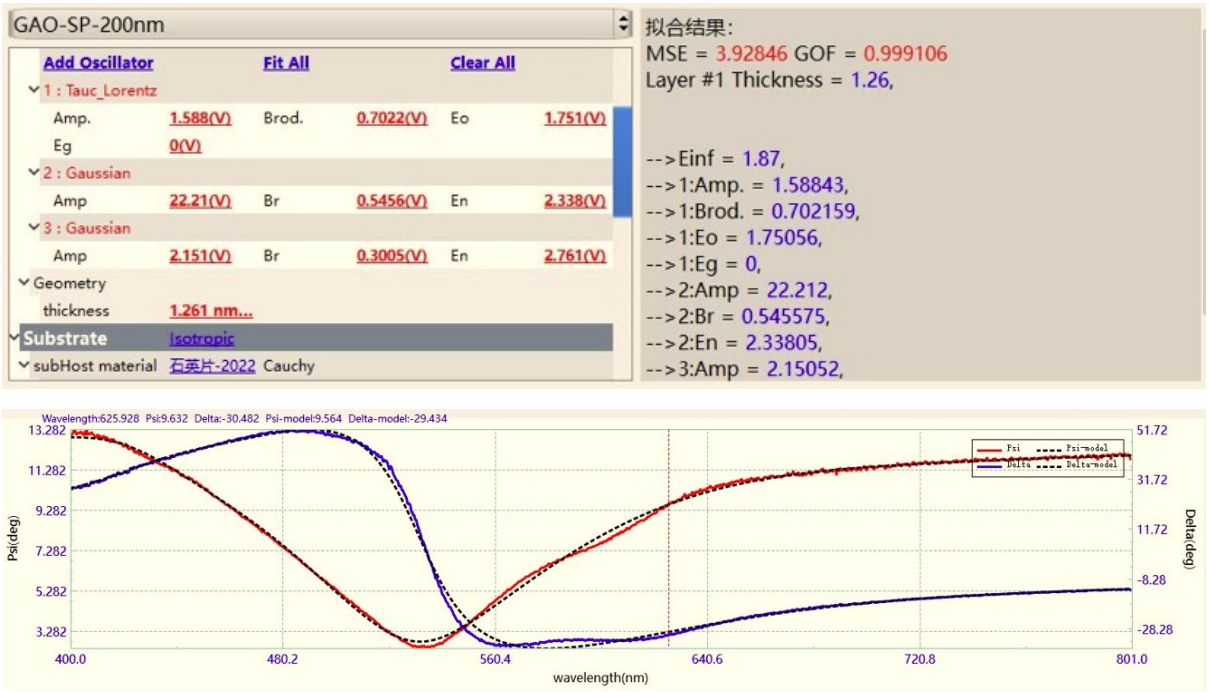

Figure S1. The thicknesses of PDCBT layer, which was measured by an ellipsometer.

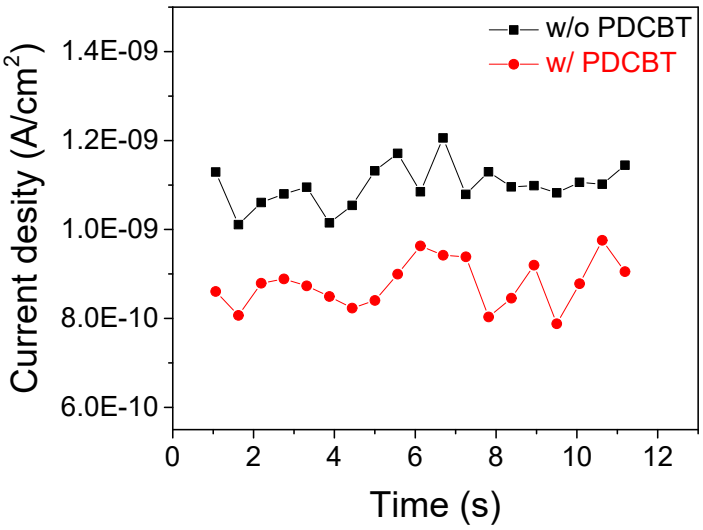

Figure S2. Dark current density under zero bias of $MAPbI_{3-x}Cl_x$-based FSPPD

with/without PDCBT layer.

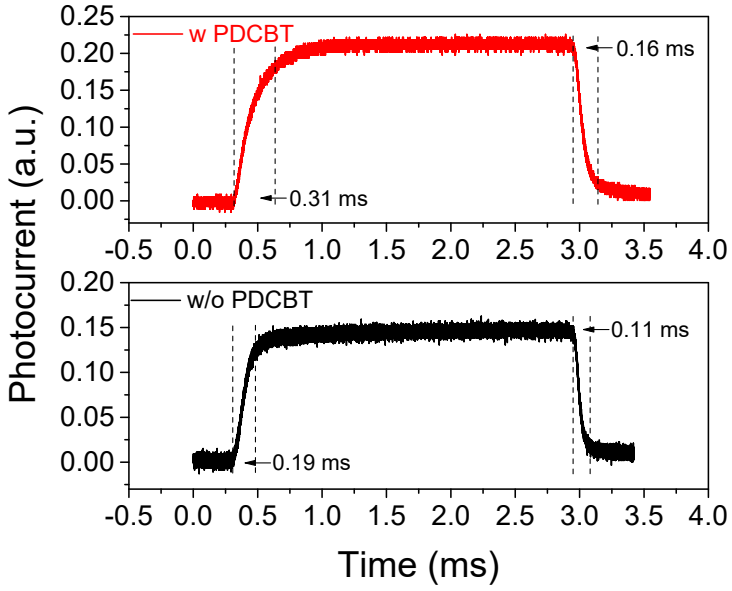

Figure S3. The rise time/decay time for FSPPD with/without PDCBT layer. The rise time of FSPPD was calculated by the time interval for the photocurrent to reach 90% of its highest value, while the decay time was calculated by the time interval for the photocurrent to fall 10% of its highest value.

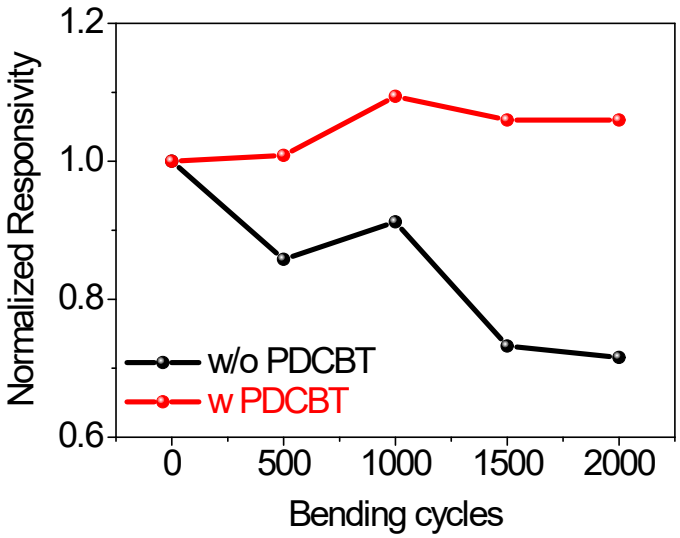

Figure S4. Normalized responsivity of $MAPbI_{3-x}Cl_x$-based FSPPD as a function of

bending cycles with bending radius of 5 mm.

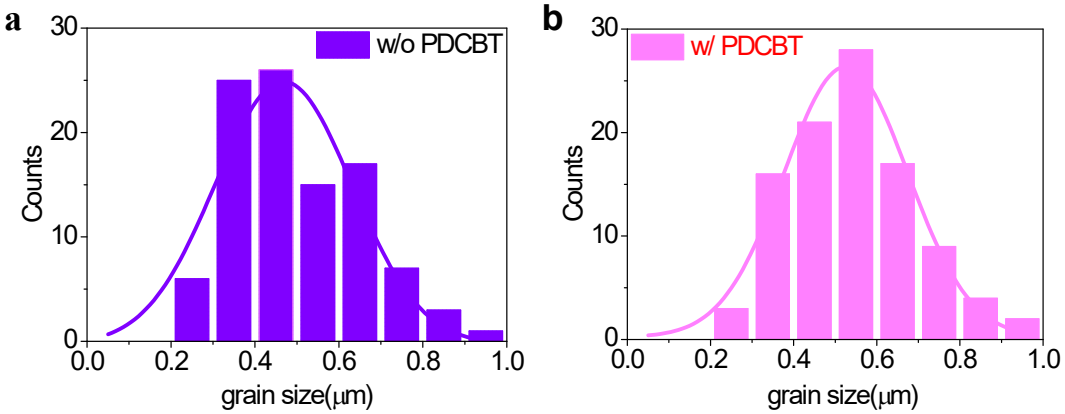

Figure S5. Grain size distribution histogram of $MAPbI_{3-x}Cl_x$ film prepared (a) without PDCBT and (b) with PDCBT, obtained by the top view of SEM images.

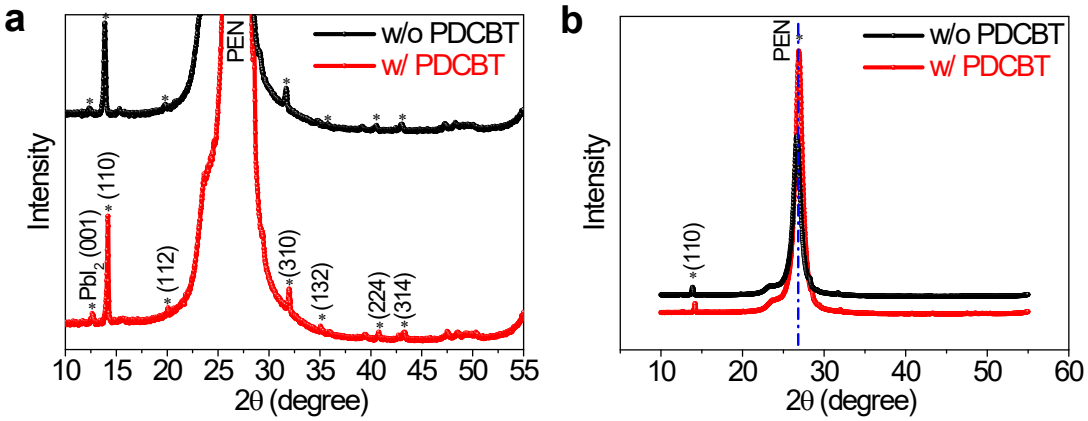

Figure S6. XRD patterns of $MAPbI_{3-x}Cl_x$ film prepared with/without PDCBT (a) excluding and (b) including PEN peak.

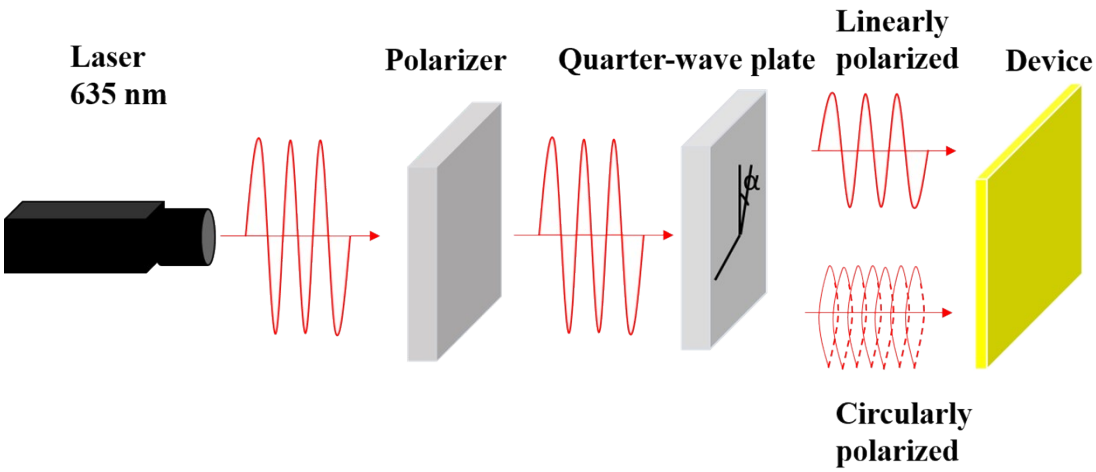

Figure S7. Experimental setup for the linear/circular photoexcitation-modulated photocurrent measurements.

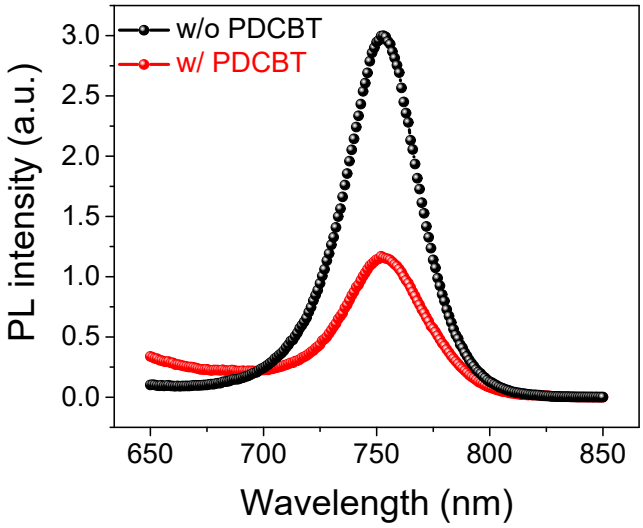

Figure S8. PL spectra of $MAPbI_{3-x}Cl_x$ films prepared with/without PDCBT.

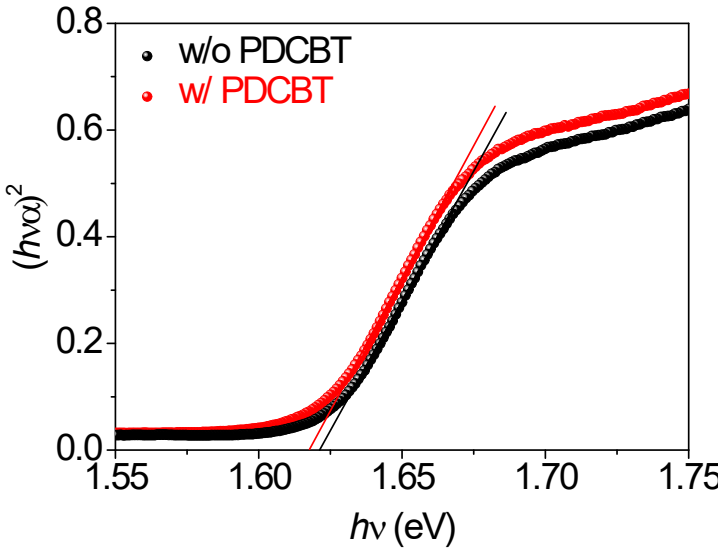

Figure S9. The band edges of $MAPbI_{3-x}Cl_x$ film prepared with/without PDCBT determined from Tauc plots.

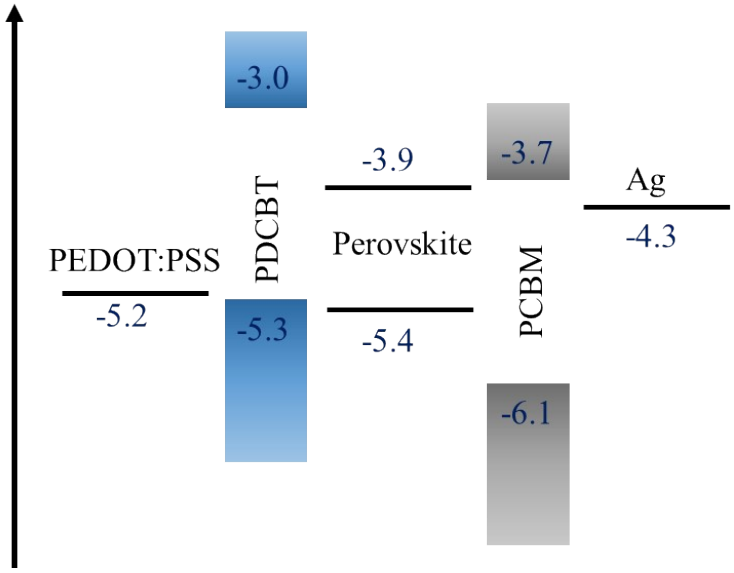

Figure S10. Energy level diagram of the FSPPD with PDCBT.