Supporting information

Power conversion efficiency enhancement of low-bandgap mixed Pb-Sn perovskite solar cells by improved interfacial charge transfer

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Experimental sections

(FASnI$_3$)$_{0.6}$(MAPbI$_3$)$_{0.4}$ perovskite film preparation. The FASnI$_3$ solution was prepared by dissolving 0.372 g of SnI$_2$ (TCI) and 0.172 g of formanidinium iodide (FAI) (Xi’an Polymer Light Technology Corp.) with 10 mol% of SnF$_2$ (0.0156 g) in 480 μL N,N-dimethylmethanamide (DMF) (anhydrous, Sigma-Aldrich) and 120 μL dimethyl-sulfoxide (DMSO) (anhydrous, Sigma-Aldrich). The MAPbI$_3$ precursor solution was prepared by dissolving 0.461 g PbI$_2$ (TCI) and 0.159 g CH$_3$NH$_3$I (MAI) (TCI) with addition of 3.5 mol% (0.0113 g) of lead thiocyanate (Pb(SCN)$_2$) (Sigma-Aldrich). The (FASnI$_3$)$_{0.6}$(MAPbI$_3$)$_{0.4}$ precursor solution was formed by mixing stoichiometric amounts of FASnI$_3$ and MAPbI$_3$ perovskite precursor. The mixed solutions are kept for 30 min before spin coating. The (FASnI$_3$)$_{0.6}$(MAPbI$_3$)$_{0.4}$ precursors were spin-coated onto ITO/PEDOT:PSS or bare glass at 5,000 rpm for 50 s. 250 μL toluene was dropped onto the spinning substrate during the spin-coating. The films annealed immediately after spin-coating process were used as reference (without DA). For delayed annealing (DA) treatment, the perovskite films were firstly stored for about 15 min and then were annealed. The condition of annealing is under 100 °C for 10 min. Several films grown on bare glass were evaporated with 25 nm C$_6$H$_{60}$ for subsequent measurement. The samples for SEM, AFM, XRD measurement were perovskite films spin-coated onto ITO/PEDOT:PSS. The samples for UV-Vis absorption were films grown on bare glass. The sample for TRPL were films grown on ITO/PEDOT:PSS, films grown on bare glasses, films grown on bare glass and then evaporated with 25 nm C$_6$H$_{60}$ for comparisons.

Device fabrication. The pre-patterned ITO substrates were cleaned by ultrasonication
in diluted Micro-90 detergent, deionized water, acetone, and isopropanol for 15 min, respectively. PEDOT:PSS films were coated on the cleaned ITO substrate at 4000 rpm for 50 s and then dried at 150 °C for 30 min. The (FASnI$_3$)$_{0.6}$(MAPbI$_3$)$_{0.4}$ perovskite films were deposited by spin-coating the precursor solution onto PEDOT:PSS film at 5,000 rpm for 50 s. Toluene was dropped onto the spinning substrate during the spin-coating. After spin-coating, some films were annealed immediately under 100 °C for 10 min, while some were firstly stored for 15 min before annealing. Finally, C$_{60}$ (25 nm)/BCP(5nm)/Ag(80 nm) were sequentially deposited by thermal evaporation to complete the device fabrication.

**Film and device characterization.** Scanning electron microscopy (SEM) images were taken on a Hitachi SU8030 electron microscope. Atomic force microscopy (AFM) images of sample surfaces were obtained on a Cypher S Atomic Force Microscope. The absorption spectrum was measured by Agilent Carry 7000 UV-Vis spectrometer. X-ray diffraction measurement was carried out by X-pert Powder. TOPAS-Academic V6 software$^1$ was used for Rietveld refinements to obtain the lattice parameters. The background and peak shapes were fit using a shifted Chebyshev function with eight parameters and a Pseudo-Voigt function (TCHZ type), respectively. Current density−voltage ($J−V$) characteristics were measured under AM1.5G light (1000W/m$^2$) using the xenon arc lamp of a Class A solar simulator. Light intensity was calibrated using an Newport-calibrated mono crystalline Si diode. The Keithley 2400 source meter was used for I-V measurement. External quantum efficiency (EQE) was characterized with an integrated system (QER3018) from EnliTech. Steady-state PL and TRPL were
detected using a home-setup microfluorescence system. The excitation light (515nm) was generated by femtosecond laser, (Light Conversion Pharos, 1030 nm, <300 fs, 1 MHz). PL spectra were measured using a spectrograph (Princeton Instruments) with a liquid-N₂-cooled CCD (PrLoN-IR). TRPL decay kinetics were collected using a TCSPC module (PicoHarp 300) and a SPAD detector (IDQ, id100). Samples were kept in a N₂ filled cell at room temperature for all optical measurements. Transient photocurrent/photovoltage decays were measured on a home-built system. A 515 nm diode laser was used to modulate the \( V_{oc} \) with a constant light bias. The pulse duration is set to 1 \( \mu \)s and the repetition rate to 20 Hz. For the constant light bias, a continuous light source was coupled through a fiber to collimate on the active area of the solar cell under study. The intensity of the pulsed laser was set in a way that the modulated \( V_{oc} \) was \( \sim 20 \) mV to ensure a perturbation regime. For the transient photocurrent decay measurements, induced by the light perturbation was measured with a digital oscilloscope set to an input impedance of 1 MΩ. The charge recombination lifetime was analyzed by single-exponential decay fitting.

![Figure S1](image)

**Figure S1.** Top-view SEM images of \((\text{FASnI}_3)_{0.6}(\text{MAPbI}_3)_{0.4}\) perovskite films without and with DA treatment with higher magnification.
Figure S2. Photoluminescence emission of perovskite films without and with DA treatment grown on glass, PEDOT:PSS and films capped with 25 nm C$_{60}$.

<table>
<thead>
<tr>
<th>Conditions</th>
<th>$\tau_1$ (ns)</th>
<th>$A_1$ (%)</th>
<th>$\tau_2$ (ns)</th>
<th>$A_2$ (%)</th>
<th>Mean lifetime (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PEDOT:PSS /Perovskite with DA</td>
<td>4.58</td>
<td>45.9</td>
<td>117.25</td>
<td>54.1</td>
<td>65</td>
</tr>
<tr>
<td>PEDOT:PSS /Perovskite W/O DA</td>
<td>4.74</td>
<td>32.9</td>
<td>164.02</td>
<td>67.1</td>
<td>106</td>
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<tr>
<td>Perovskite/C$_{60}$ with DA</td>
<td>0.44</td>
<td>76</td>
<td>14.86</td>
<td>24</td>
<td>3.9</td>
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<tr>
<td>Perovskite/C$_{60}$ W/O DA</td>
<td>0.48</td>
<td>73.1</td>
<td>17.11</td>
<td>26.9</td>
<td>5.0</td>
</tr>
</tbody>
</table>

Table S1. Fitting parameters of TRPL for perovskite films with and without DA treatment interfaced with PETDOT:PSS, C$_{60}$.

Figure S3. Maximal steady-state photocurrent output and efficiencies of champion cell with DA at maximum power point.
Figure S4. $J$-$V$ curves of the PCE champion device measured freshly and 30 days later.

Reference
1. A. Coelho, TOPAS-Academic V6 software (Coelho Software, Brisbane, Australia, 2007).