

## ***Supporting Information***

# **Simultaneously Enhancing Performance of Perovskite Solar Cells and Suppressing Lead Leakage Via Interface Modification Strategy**

Yushuang Gao, Zhengyan He, Quanming Geng, Xiangrui Jia, Shufang Zhang\*, Dehua Wang\*

*School of Physics and Photoelectronic Engineering, Ludong University, Yantai 264025, P. R. China.*

**\*Corresponding author**

*E-mail address:* zhang.shufang@ldu.edu.cn (S.F. Zhang)

lduwdh@163.com (D.H. Wang)

## Experimental Section

### Materials

Cesium iodide (CsI, 99.5%), Lead iodide (PbI<sub>2</sub>, 99.9%), and Formamidinium iodide (FAI, 99.9%) were purchased from Advanced Election Technology CO.,Ltd, PTASS was purchased from Shandong Taihe Water Treatment Technologies Co.,Ltd, N, N-dimethylformamide (DMF, 99.5%), and dimethyl sulfoxide (DMSO, 99.5%) were purchased from Aladdin. SnO<sub>2</sub> colloidal dispersion (diluted 15% in H<sub>2</sub>O) was purchased from Xi'an Polymer Light Technology Corp. Unless otherwise specified, all powders and solutions are used directly without further purification.

### Device fabrication

Firstly, ultrapure water, acetone, and ethanol are used to clean FTO substrate in an ultrasonic cleaner for 40 minutes in turn and then put into a UV cleaning box for the mixed treatment of UV and O<sub>3</sub> for about 45 min. For the SnO<sub>2</sub> electron transport layer, firstly, the used solution was prepared from SnO<sub>2</sub> nanocrystal solution (diluted to 4.5% with H<sub>2</sub>O), then spin-coated on the FTO substrate for 30 s at 5,000 rpm/s, and finally heated at 160 °C in an air environment annealed for 35 minutes. For the SnO<sub>2</sub> film modified by PTASS, firstly, different concentrations of PTASS powder were dissolved in IPA, then dripped on the fresh SnO<sub>2</sub> film cooled to room temperature, spin-coated at the speed of 4000 rpm/s for 30 s, and then heated on a hot plate for 10min. 0.23 g FAI, 0.65 g PbI<sub>2</sub>, and 0.04 g CsI were mixed in 1 mL DMSO and DMF mixed solvent (4:1, v/v), and the FA<sub>0.9</sub>Cs<sub>0.1</sub>PbI<sub>3</sub> perovskite precursor solution with a concentration of 1.35 M was prepared, which was then put into a stirrer and stirred at 50 °C for 8 hours until it was completely dissolved. The perovskite precursor solution was dropped onto the SnO<sub>2</sub> substrate and spin-coated at 5000 rpm/s for 40 s. The anti-solvent was added dropwise during spin coating by adding 250 μL of EA dropwise

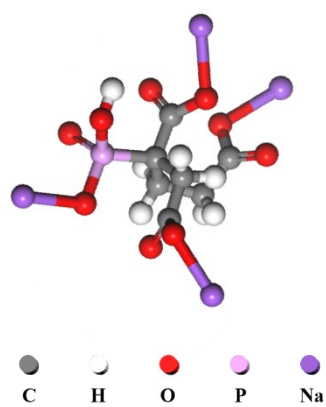
after spinning for 10 seconds. Then, the perovskite films were annealed in a nitrogen atmosphere for 10 minutes with a hot plate temperature of 140 °C. The hole transporter, dissolving the hole transport material spiro-OMeTAD in a mixed solution of tert-butylpyridine (TBP) and chlorobenzene (0.225 g spiro-OMeTAD in 0.05 g TBP and 1.10 mL chlorobenzene), and the additive bistrifluoromethanesulfonimide lithium salt (LiTFSI) was added, the solution was then spin-coated on the device surface for 35 s at 4500 rpm/s. Finally, the top electrode was covered with a mask, and 60 nm thick Au electrodes were deposited by thermal evaporation deposition coating equipment.

### **Characterization**

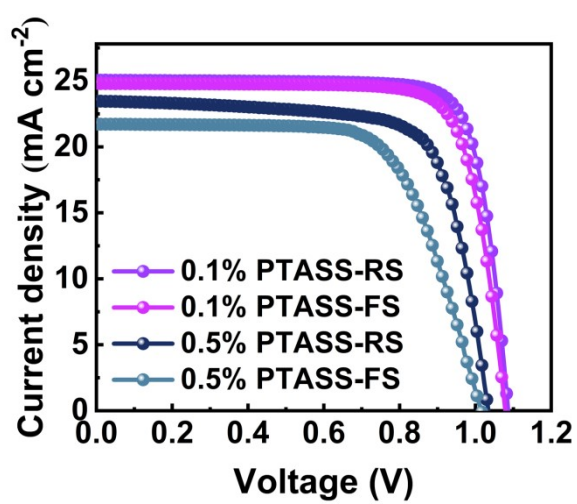
*Characterization of large instruments:* FTIR spectra were measured by IRTracer-100 (Shimadzu, Japan). The ultraviolet-visible spectrum of the sample was recorded by ultraviolet-visible-near infrared spectrophotometer (UV-3600i, Shimadzu). Surface morphologies of perovskite thin films were observed by atomic force microscopy (Bruker AFM Multimode 8) and scanning electron microscopy (FEI NOVA NanoSEM 450). The XRD of the samples was measured on an Empyrean X-ray diffractometer (PANalytical B.V. Co., Netherlands) with Cu K $\alpha$  radiation. The steady-state PL spectra of the samples were measured using Steady State & Transient State Fluorescence Spectrofluorometer FluroMax-4P with an excitation light source wavelength of 325 nm. TRPL measurements were performed by Time-Resolved Fluorescence Spectrofluorometer TemPro-01 with an excitation light source wavelength of 478 nm. X-ray photoelectron spectroscopy (XPS) Measurement of samples by K-alpha (Thermo Scientific, USA). The linear scanning current-voltage curve of PSC was measured with Keithley 2400 digital source meter under the standard sunlight simulator (100mw cm<sup>-2</sup>, Solar Simulator 94023A, Newport). Lead

ion concentration was measured by inductively coupled plasma optical emission spectrometer (Plasma 2000, SGN). For stability, unencapsulated perovskite films and devices were placed in an air environment at 25 °C and 40% relative humidity (RH) for long-term monitoring.

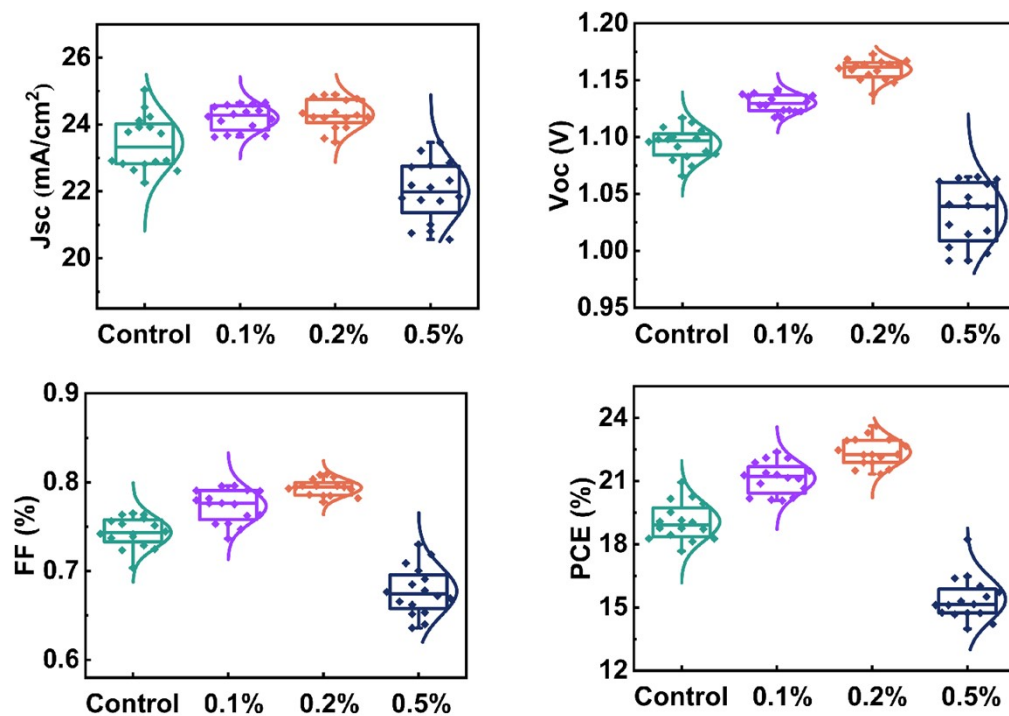
## Figures and Tables



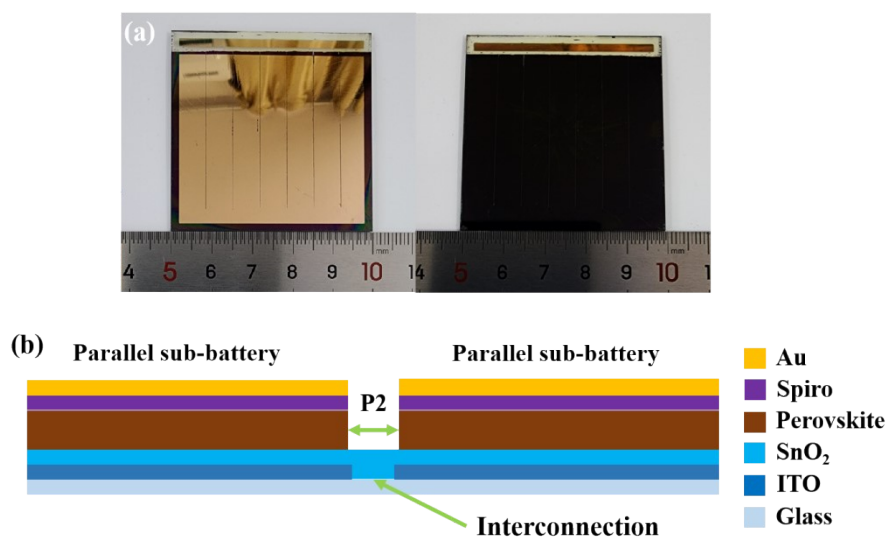
**Figure S1.** Molecular structure of PTASS.



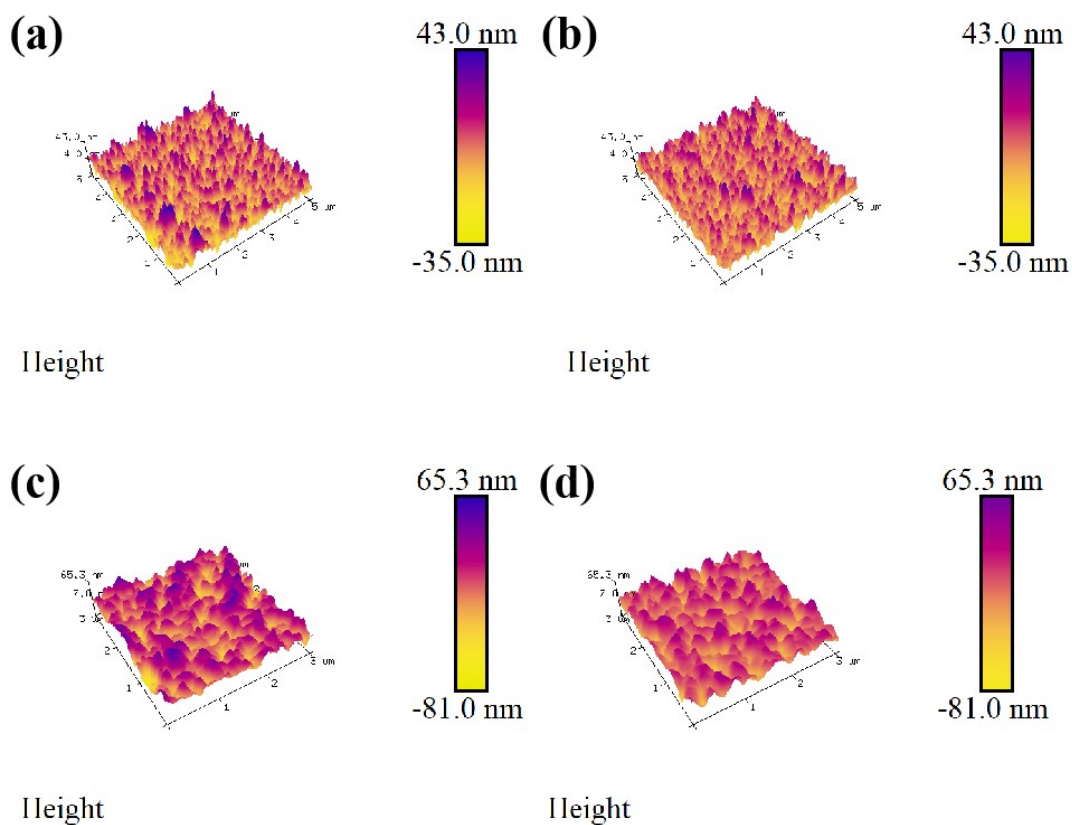
**Figure S2.**  $J$ - $V$  curves of the champion cells based on different perovskite films.



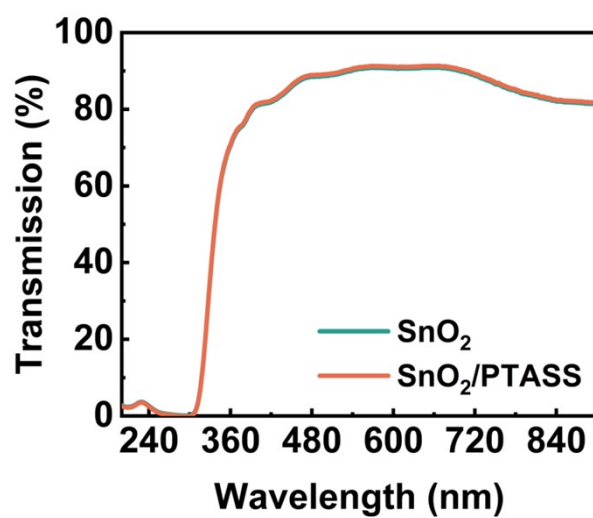
**Figure S3.** The photovoltaic parameters statistics for 20 PTASS-modified PSCs with different concentrations of PTASS.



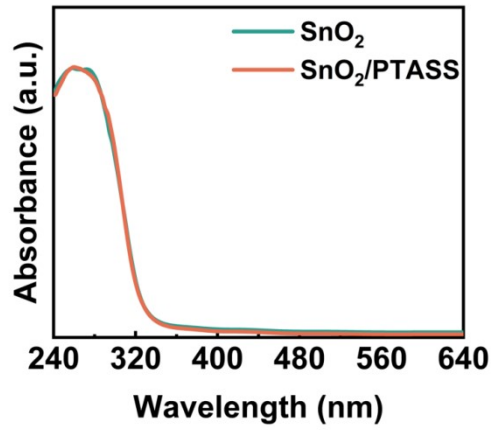
**Figure S4.** (a) Optical image of large-area perovskite modules. (b) Cross-sectional schematic illustration of the fabricated module coupled with the deposition methods of the functional layers.



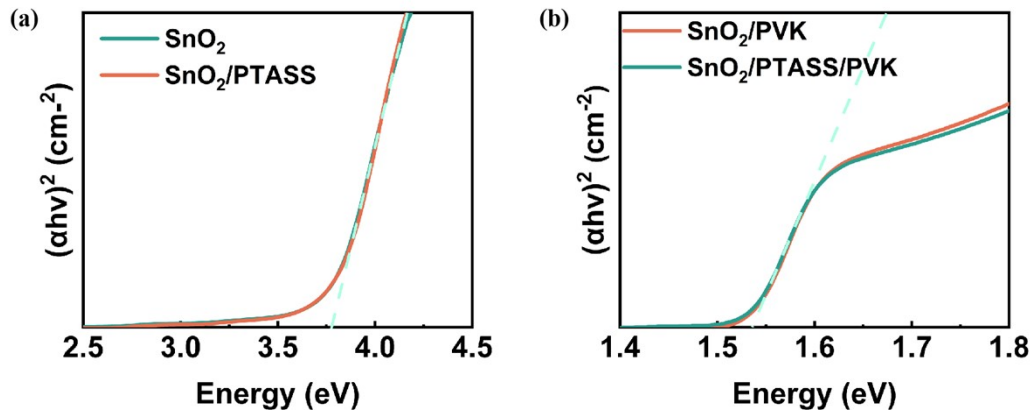
**Figure S5.** The 3D AFM images of SnO<sub>2</sub> films (a) with or (b) without PTASS modification. The 3D AFM images of perovskite films deposited on (c) unmodified SnO<sub>2</sub> and (d) PTASS-modified SnO<sub>2</sub>.



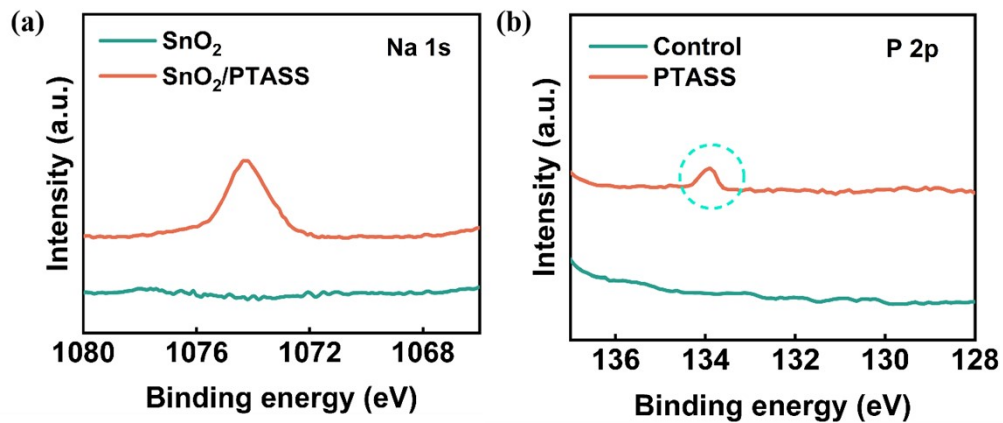
**Figure S6.** Transmission spectrum of SnO<sub>2</sub> and SnO<sub>2</sub>/PTASS films.



**Figure S7.** UV-vis absorption spectra of SnO<sub>2</sub> and SnO<sub>2</sub>/DAD films.

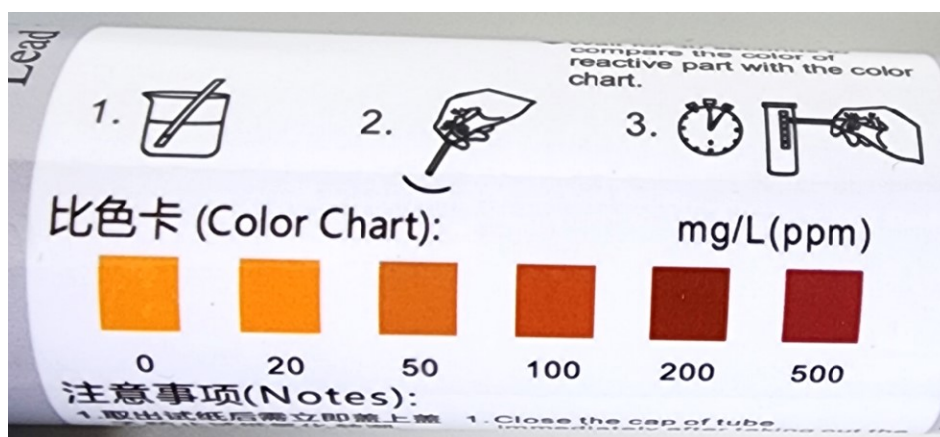


**Figure S8.** Tauc plots of (a) SnO<sub>2</sub> films and (b) perovskite films without or with PTASS modification.



**Figure S9.** (a) Na 1s and (b) P 2p XPS spectra of SnO<sub>2</sub> films without or with PTASS modification.





**Figure S10.** The corresponding lead ion concentration color card.

**Table S1.** The photovoltaic performance parameters of the optimal devices based on different perovskite films.

Name	Scan direction	$J_{sc}$ [ $\text{mA cm}^{-2}$ ]	$V_{oc}$ [V]	FF [%]	PCE [%]	HI [%] <sup>b)</sup>
Control	Reverse	24.516	1.117	76.525	20.956	8.239
	Forward	24.113	1.089	73.229	19.229	
0.1% PTASS	Reverse	24.621	1.142	79.58	22.376	4.067
	Forward	24.309	1.121	77.931	21.237	
0.2% PTASS	Reverse	24.892	1.173	80.889	23.618	1.316
	Forward	24.662	1.169	80.845	23.308	
0.5% PTASS	Reverse	23.463	1.065	72.994	18.240	15.778
	Forward	21.719	1.021	69.276	15.362	

b)  $HI = (PCE_{Rev} - PCE_{For}) \times 100 / PCE_{Rev}$

**Table S2.** The photovoltaic performance parameters of the optimal modules based on large area.

Name	Scan direction	$J_{sc}$ [mA]	$V_{oc}$ [V]	FF [%]	PCE [%]	HI [%]
Module device (Area: 19.32 $\text{cm}^2$ )	Reverse	0.454	1.076	76.81	19.421	4.33
	Forward	0.447	1.058	75.902	18.58	

**Table S3.** Summary of the Relevant Parameters of PL Lifetimes.

Name	A <sub>1</sub>	τ <sub>1</sub> [ns]	A <sub>2</sub>	τ <sub>2</sub> [ns]	τ <sub>ave</sub> [ns] <sup>a)</sup>
SnO <sub>2</sub> /PVK	0.251	8.88	0.694	49.766	38.903
SnO <sub>2</sub> /PTASS/PVK	0.402	3.805	0.617	26.272	17.409
Glass/PVK	0.252	29.081	0.694	162.685	127.068
Glass/PTASS/PVK	0.341	33.802	0.57	325.005	216.04

a)  $\tau_{ave} = \frac{A_1\tau_1 + A_2\tau_2}{A_1 + A_2}$

**Table S4.** Summary of the results of fitting EIS.

Name	R <sub>s</sub>	R <sub>tr</sub>	R <sub>rec</sub>
Control	86.237	94761	1675700
0.2% PTASS	31.218	56847	3790700