Supporting Information

Highly Efficient Planar Perovskite Solar Cells via Acid-assisted Surface Passivation

Xin Zhang[‡†§], Zejiao Shi[†§], Haizhou Lu[†], Xiaoguo Li[†], Haoliang Wang[†], Sijian Yuan[†], Fengcai Liu[†], Yiyi Pan[†], Zhenhua Weng[†], Haijuan Zhang[†], Xiaolei Cui[†], Qi Liu[⊥], Chongyuan Li[⊥], Akrajas Ali Umar[#], Jiao Wang[†], Xiao-Chun Hang[⊥], Zhengyi Sun[⊥], Yiqiang Zhan[†]*

[‡] Academy for Engineering & Technology, Fudan University, Shanghai 200433, China.

[†] Center for Micro Nano Systems, School of Information Science and Technology (SIST), Fudan University, Shanghai 200433, China

[⊥] Key Laboratory of Flexible Electronics (KLOFE) & Institute of Advanced Materials (IAM), Jiangsu National Synergistic Innovation Centre for Advanced Materials (SICAM), Nanjing Tech University, 30 South Puzhu Road, Nanjing 211800, China.

[#] Institute of Microengineering and Nanoelectronics, (IMEN), Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia.

*corresponding author: yqzhan@fudan.edu.cn

[§] These two authors contribute equally to this work.

KEYWORDS: perovskite solar cells, tin oxide, interface, defect passivation

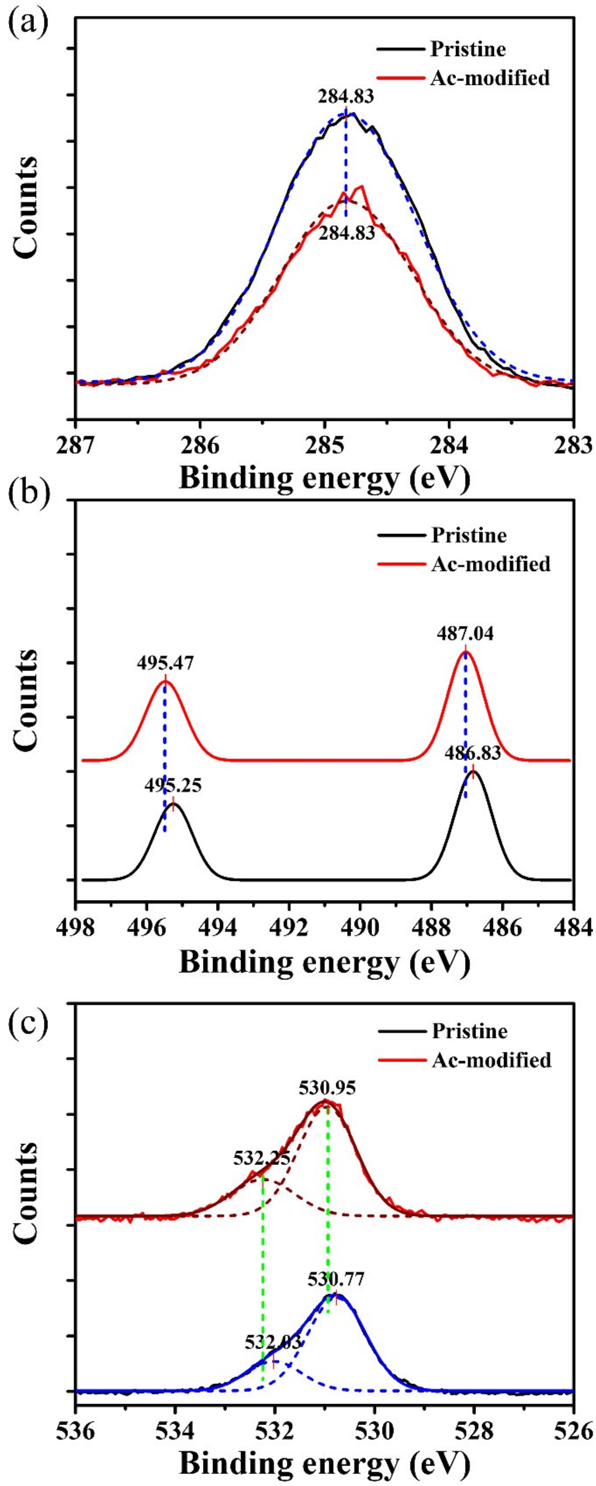
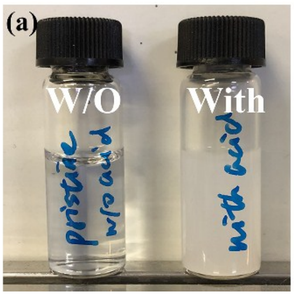


Fig. S1 XPS spectra of (a) C 1*s*, (b) Sn 3*d* and (c) O 1*s* with marked specific peaks position.

(a)



(b)

$$\mathbf{SnO_2 + CH_3COOH \longrightarrow Sn(OOCCH_3)_4 + H_2O}$$

Fig. S2 (a) Photographs of SnO_2 precursor solutions without (Left) and with (Right) adding acetic acid. (b) Chemical reaction of SnO_2 and acetic acid.

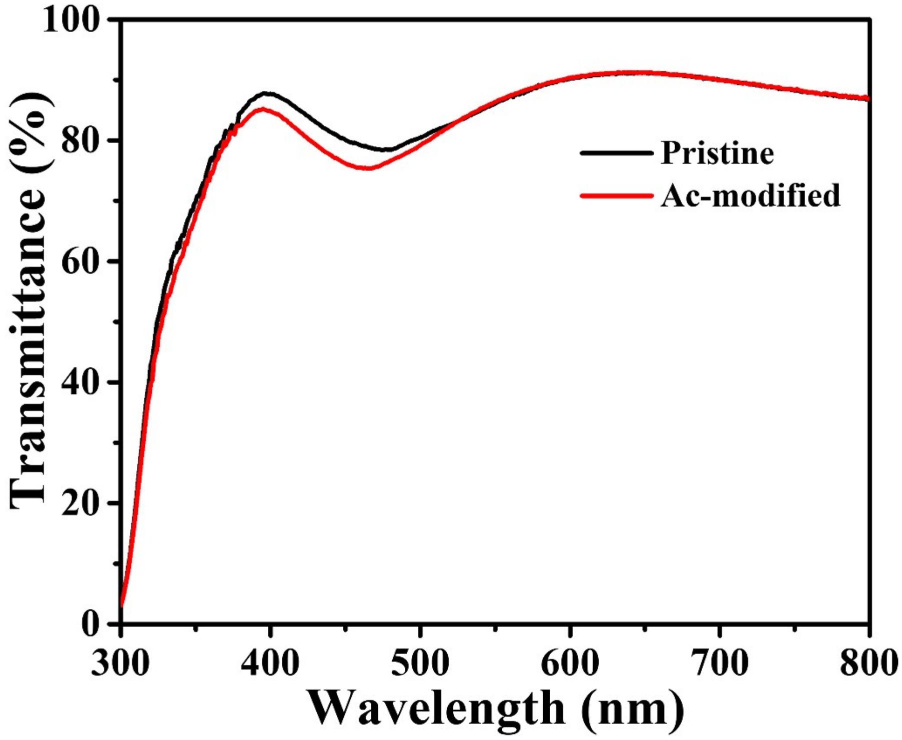


Fig. S3 Transmission spectra of pristine and Ac-modified SnO_2 films.

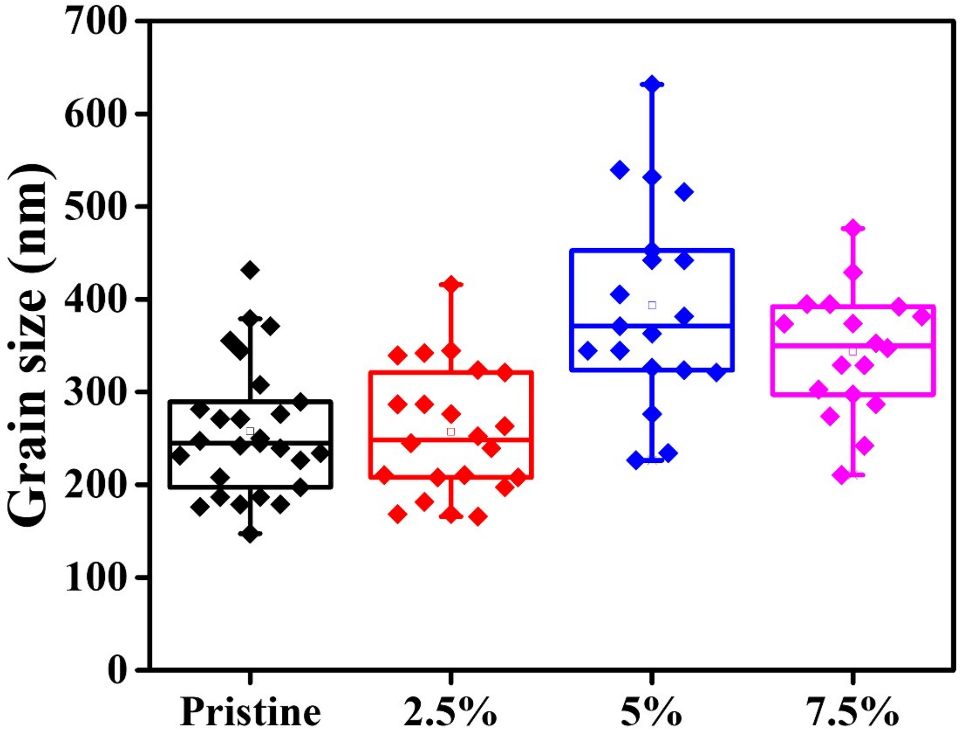


Fig. S4 The grain size distribution of perovskite films deposited on pristine and Ac-modified SnO_2 substrates.

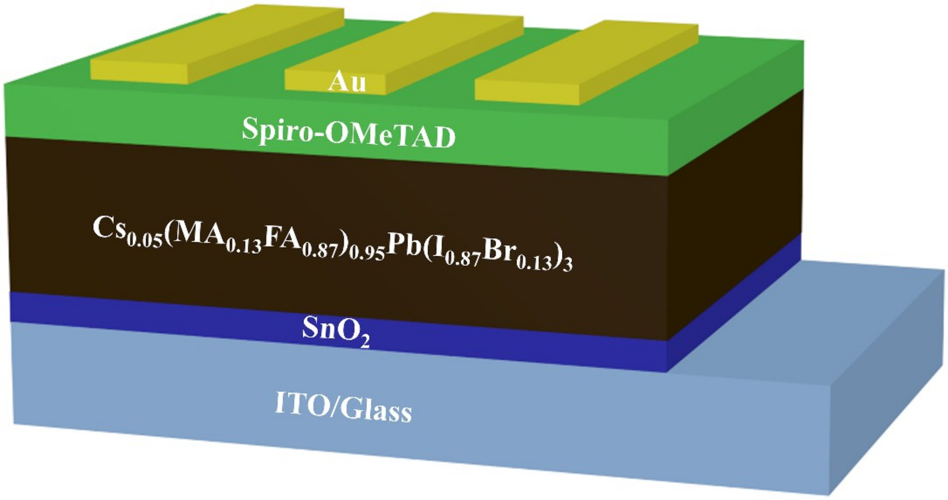


Fig. S5 Device structure of planar-type PSCs. For triple cation PSC, the active layer is $Cs_{0.05}(MA_{0.13}FA_{0.87})_{0.95}Pb(I_{0.87}Br_{0.13})_3$ and for dual cation PSC, the active layer is $(FAPbI_3)_{1-x}(MAPbBr_3)_x$).

Table. S1 Photovoltaic parameters of triple cation PSCs based on different SnO_2/ITO substrates, the device architecture in this study is the regular planar structure of ITO/SnO_2/Perovskite/Spiro-OMeTAD/Au.

Samples		V_{oc} (V)	J_{sc} (mA·cm^{-2})	FF	PCE (%)
Pristine	Average	1.137 ± 0.004	21.83 ± 0.26	0.749 ± 0.006	18.57 ± 0.17
	Best	1.141	22.07	0.748	18.84
2.5%	Average	1.143 ±0.007	21.86 ± 0.11	0.781 ± 0.006	19.52 ± 0.15
	Best	1.141	21.92	0.787	19.68
5%	Average	1.153 ± 0.005	22.31 ± 0.09	0.791 ± 0.003	20.33 ± 0.17
	Best	1.158	22.39	0.793	20.56
7.5%	Average	1.145 ± 0.006	21.31 ± 0.25	0.779 ± 0.004	19.02 ± 0.18
	Best	1.145	21.36	0.784	19.17

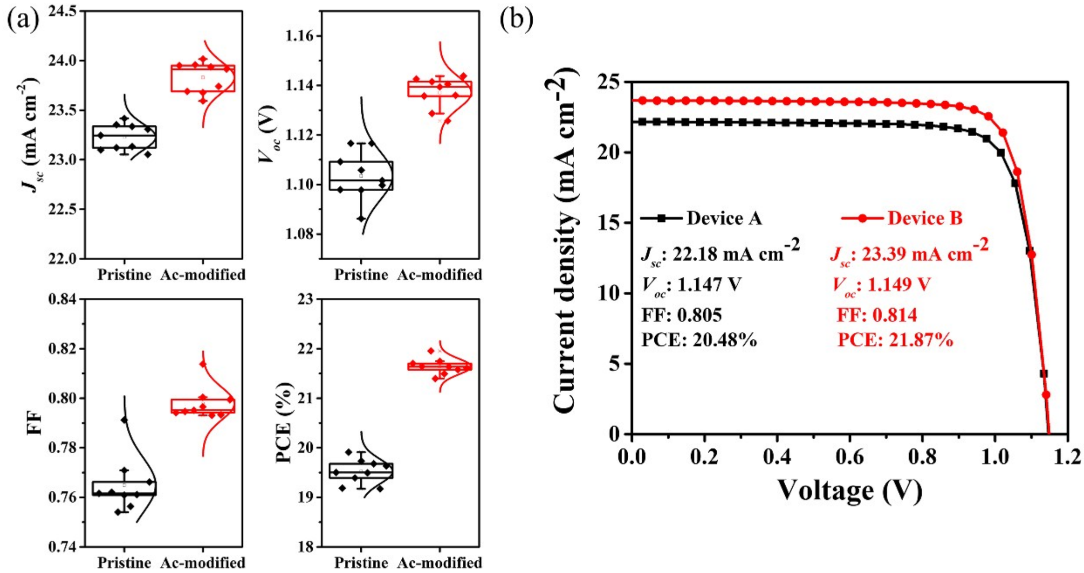
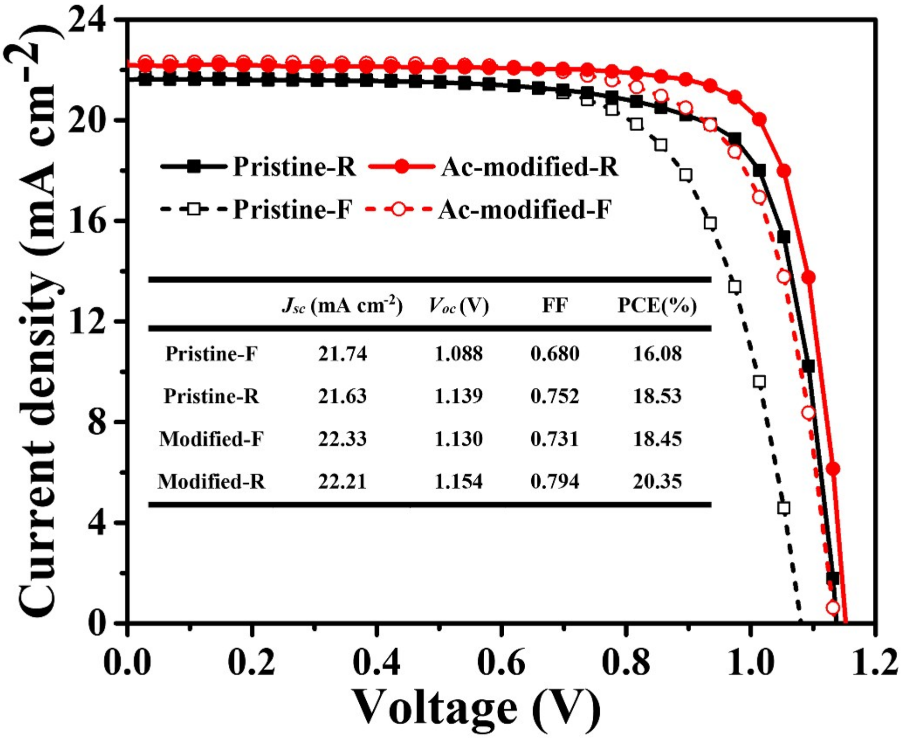


Fig. S6 (a) Comparison of photovoltaic parameters of the dual cation PSCs based on pristine and Ac-modified SnO_2. (b) The *J-V* curves of different devices with the highest FF (Device A is triple cation PSC and Device B is dual cation perovskite).

Table. S2 Photovoltaic parameters of dual cation PSCs based on different SnO_2 with device architecture of ITO/SnO_2/Perovskite/Spiro-OMeTAD/Au.

Samples		V_{oc} (V)	J_{sc} (mA·cm^{-2})	FF	PCE (%)
Pristine	Average	1.103 ± 0.009	23.23 ± 0.13	0.765 ± 0.011	19.52 ± 0.25
	Best	1.117	23.13	0.771	19.92
Ac-modified	Average	1.137 ±0.006	23.83 ± 0.15	0.798 ± 0.007	21.64 ± 0.15
	Best	1.139	23.68	0.814	21.95



	J_{sc} (mA cm^{-2})	V_{oc} (V)	FF	PCE(%)
Pristine-F	21.74	1.088	0.680	16.08
Pristine-R	21.63	1.139	0.752	18.53
Modified-F	22.33	1.130	0.731	18.45
Modified-R	22.21	1.154	0.794	20.35

Fig. S7 The *J*–*V* curves of the devices with pristine and Ac-modified SnO_2 ETL measured under reverse and forward scanning directions.

Table. S3 Parameters of the time-resolved PL spectra of triple cation perovskite based on pristine SnO_2, Ac-modified SnO_2 and glass substrates.

Sample	τ_1 (ns)	τ_2 (ns)
Pristine	4.83	22.15
Ac-modified	4.31	21.17
Glass	23.5	167.7

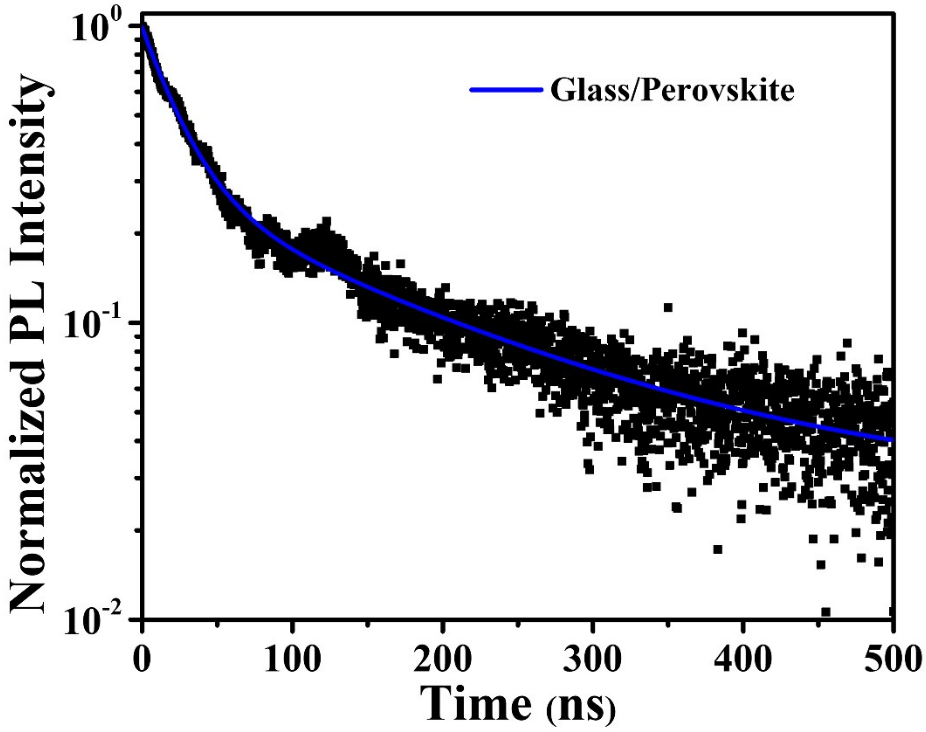


Fig. S8 Time-resolved spectrum of glass/perovskite device with larger timescale.

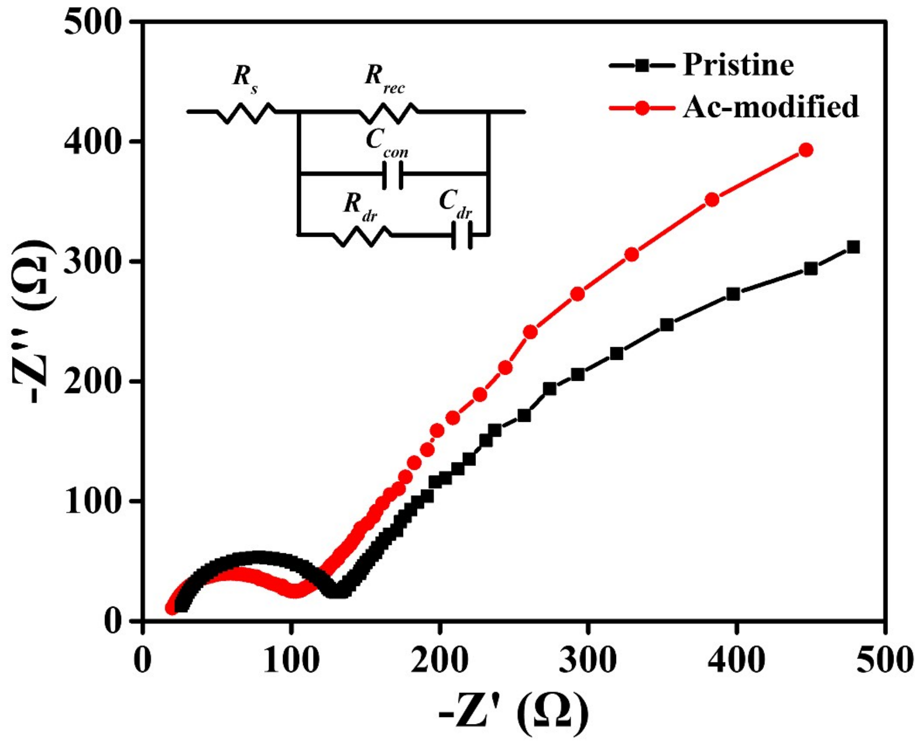


Fig. S9 Nyquist plots of PSCs based on pristine and Ac-modified SnO_2 measured under constant light illumination of 100 mW cm^2 from 1 Hz to 1 MHz. The inset gives the equivalent circuit model, R_s is series resistance, R_{rec} is recombination resistance, R_{dr} is dielectric relaxation resistance, C_{con} is selective contact capacitance and C_{dr} is dielectric relaxation capacitance of the perovskite.

Table. S4 EIS parameters of the pristine and Ac-modified PSCs obtained by Z-view software.

Sample	R_s (Ω)	R_{rec} (Ω)	C_{con} (F)	R_{dr} (Ω)	C_{dr} (F)
Pristine	24.61	1208	9.23×10^{-9}	112.80	1.22×10^{-5}
Ac-modified	16.39	2068	3.51×10^{-9}	88.39	8.40×10^{-6}

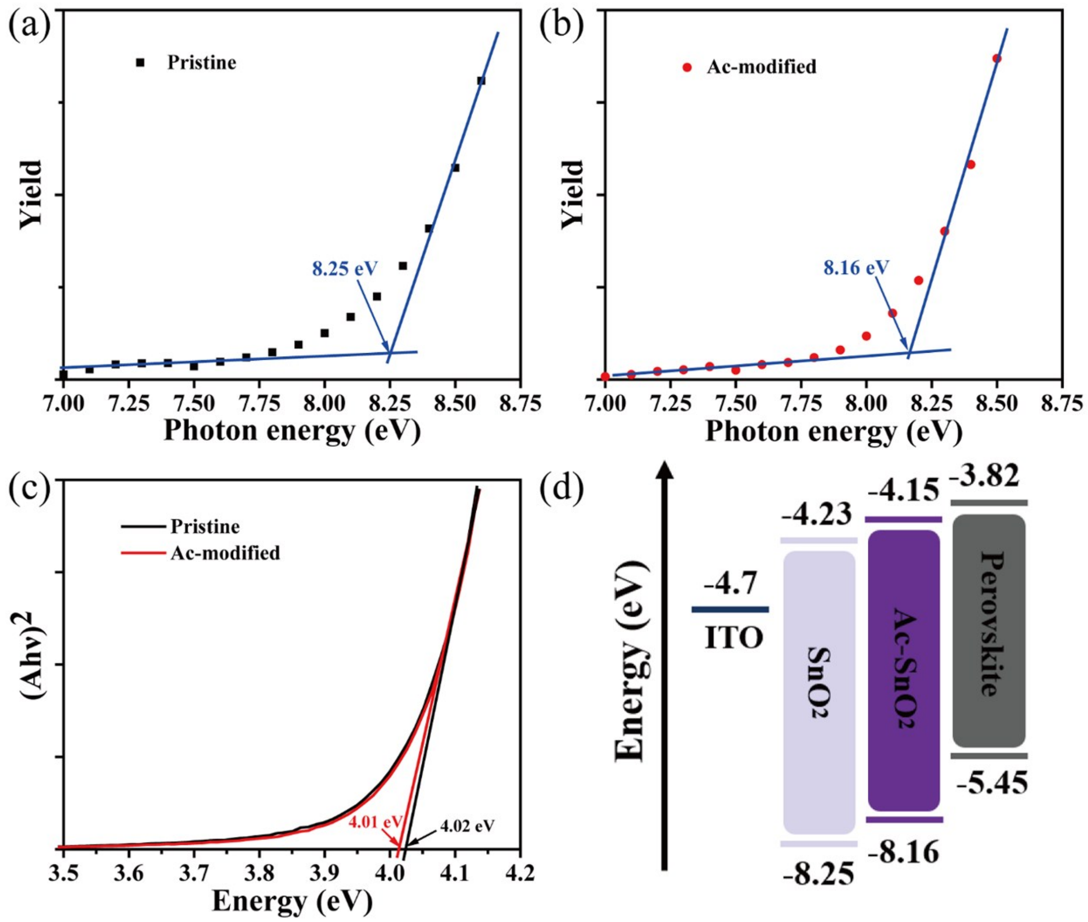


Fig. S10 (a) IPS spectrum of pristine SnO_2. (b) IPS spectrum of Ac-modified SnO_2. (c) Tauc plots for pristine and Ac-modified SnO_2 films. (d) Energy level diagram of the PSCs.