

Supplementary Materials

A universal Ti_3C_2 assisted strategy facilitates hole extraction of PEDOT:PSS anode interface layer for over 19% efficiency organic photovoltaics

Yi-lin Liu^{1,#}, Xiaojian Zheng^{1,2,#}, Gaoyang Wang¹, Wenquan Wang¹, Jing Guo³, Luobin She¹, Zhixin Liu^{1,2}, Zhenye Li^{1,*}, Xingzhu Wang^{1,2,*}

¹College of Mechanical Engineering and School of Electrical Engineering, University of South China, Hengyang 421001, Hunan, China.

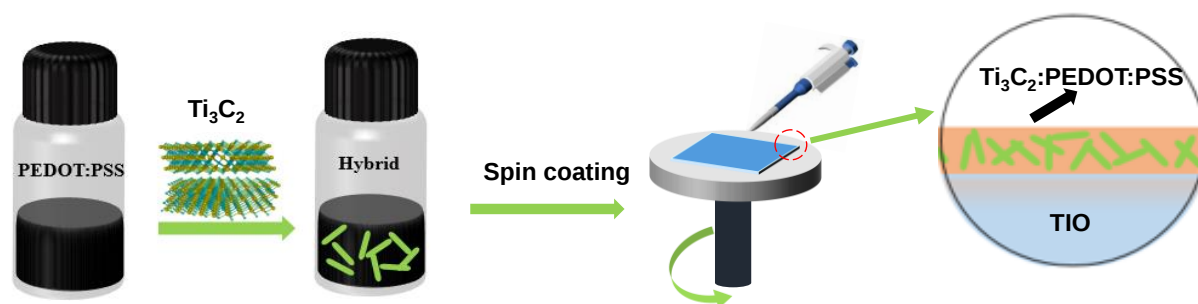
²Engineering and Research Center for Integrated New Energy Photovoltaics & Energy Storage Systems of Hunan Province, University of South China, Hengyang 421001, Hunan, China.

³School of Pharmaceutical Science, University of South China, Hengyang 421001, Hunan, China.

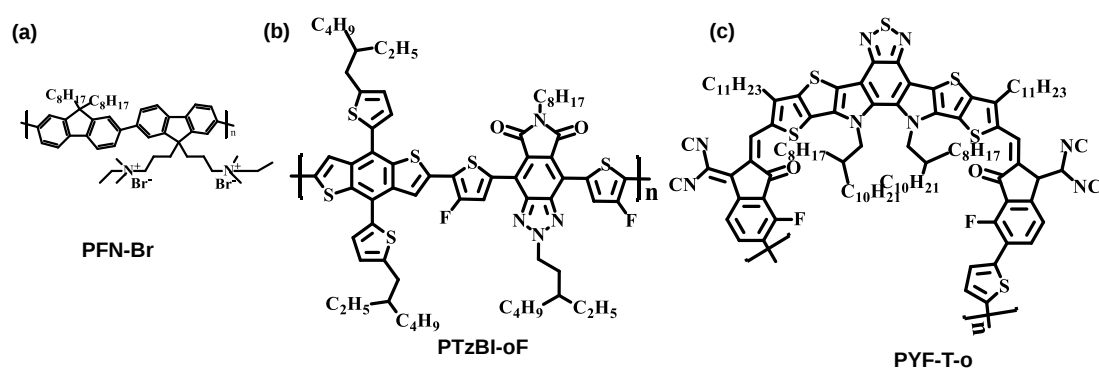
[#]Authors contributed equally.

***Correspondence to:** Prof. Zhenye Li, College of Mechanical Engineering, University of South China, Hengyang 421001, Hunan, China. E-mail: lizhenye@usc.edu.cn; Prof. Xingzhu Wang, Engineering and Research Center for Integrated New Energy Photovoltaics & Energy Storage Systems of Hunan Province and School of Electrical Engineering, University of South China, Hengyang 421001, Hunan, China. E-mail: 2022000041@usc.edu.cn

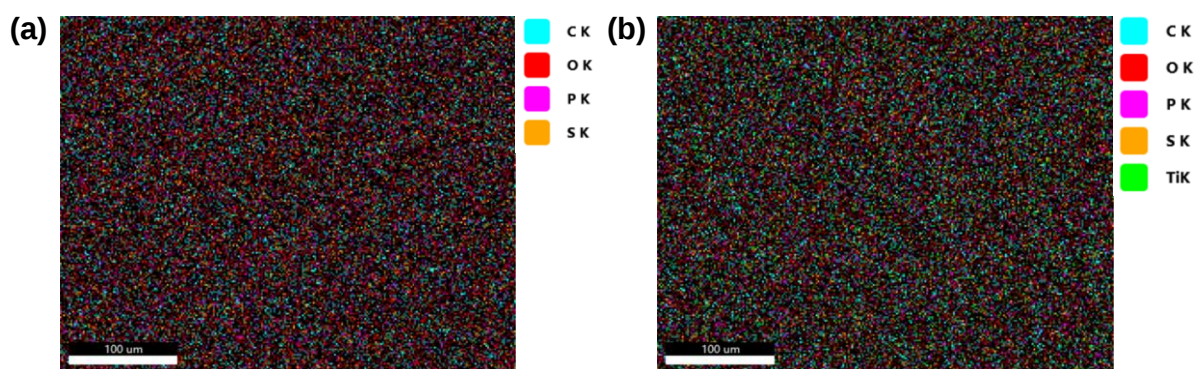
Supplementary Figures and Tables



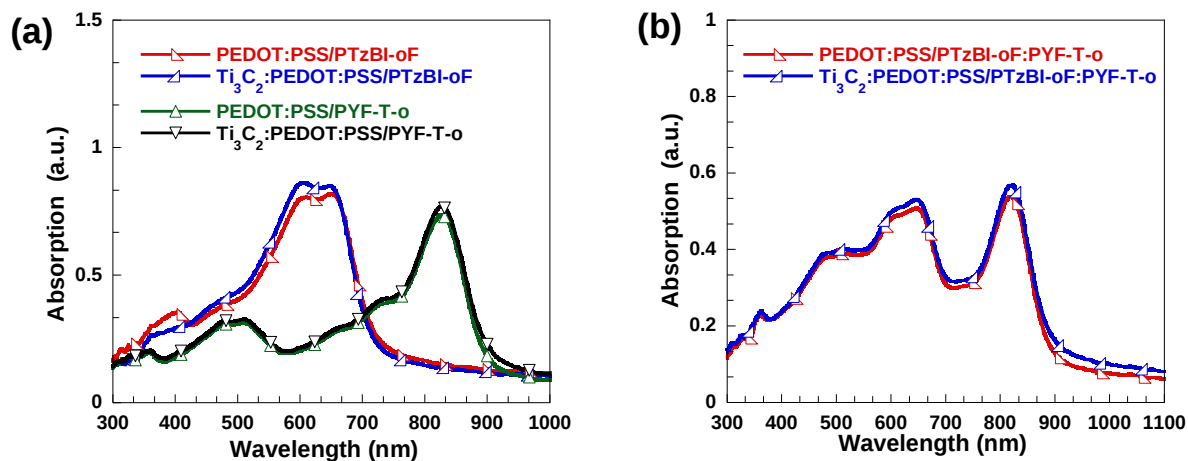
Supplementary Figure 1. Preparation process of Ti_3C_2 :PEDOT:PSS hybrid AIL.



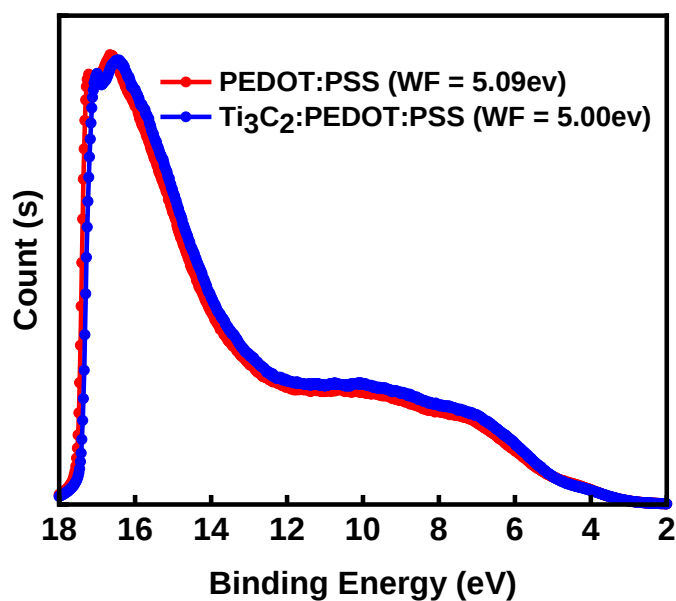
Supplementary Figure 2. Molecular structures of (a) PFN-Br, (b) PTzBI-oF and (c) PYF-T-o.



Supplementary Figure 3. EDS images of PEDOT:PSS (a) and Ti_3C_2 :PEDOT:PSS (b) interlayers.



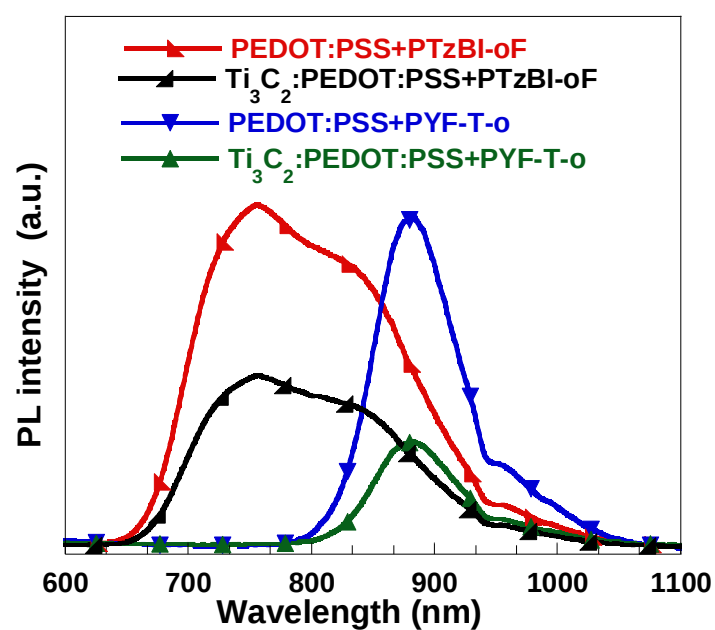
Supplementary Figure 4. (a) Absorption spectra of neat PTzBI-oF and PYF-T-o films spin-coated on the PEDOT:PSS with or without Ti_3C_2 ; (b) Absorption spectra of PTzBI-oF:PYF-T-o blend films spin-coated on the AIL with or without Ti_3C_2 .



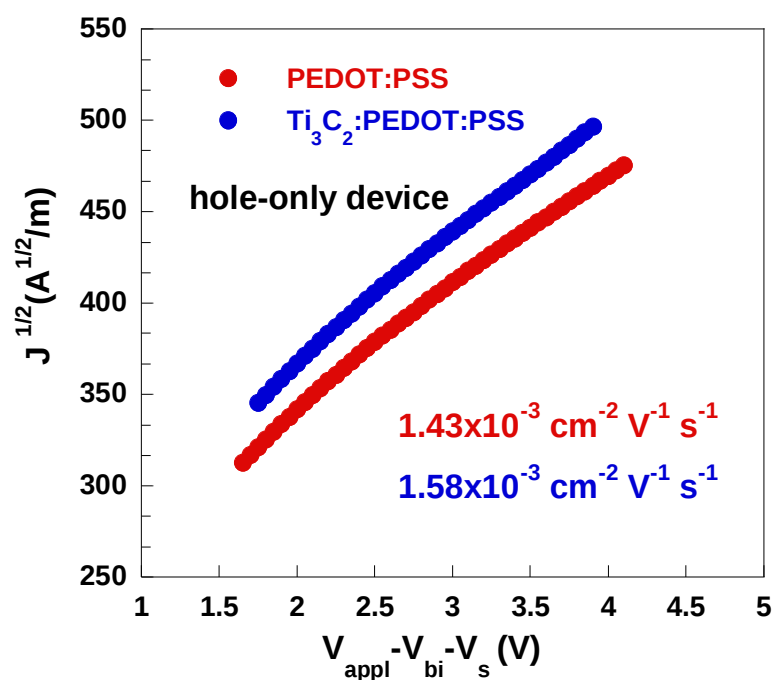
Supplementary Figure 5. Ultraviolet photoelectron spectrometer of PEDOT:PSS and Ti_3C_2 :PEDOT:PSS interlayers.

Supplementary Table 1. Work function of PEDOT:PSS and Ti_3C_2 :PEDOT:PSS

AIL	Work function (eV)
PEDOT:PSS	-5.00
Ti_3C_2 :PEDOT:PSS	-5.09

Supplementary Figure 6. PL spectra of neat PTzBI-oF and PYF-T-o films that spin-coated on the PEDOT:PSS films with or without Ti_3C_2 dopants.Supplementary Table 2. Conductivity parameters of PEDOT:PSS and Ti_3C_2 :PEDOT:PSS films

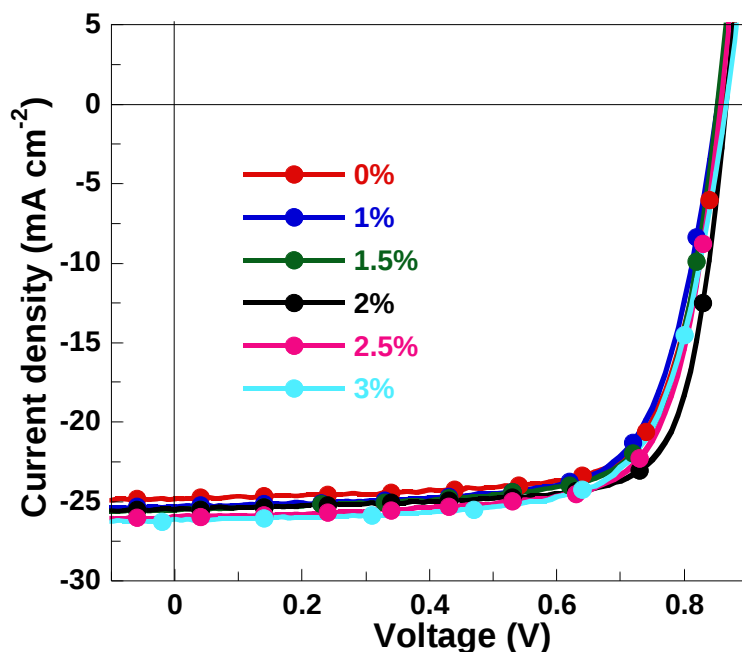
Film	Conductivity (S m^{-1})
PEDOT:PSS	3.20×10^{-3}
Ti_3C_2 :PEDOT:PSS	5.47×10^{-3}



Supplementary Figure 7. SCLC hole mobility for PTzBI-oF:PYF-T-o devices with (or without) Ti_3C_2 in the PEDOT:PSS interlayer.

Supplementary Table 3. SCLC hole mobility parameters for PTzBI-oF:PYF-T-o control films and PTzBI-oF:PYF-T-o optimized films with different AIL

AIL	μ_h ($\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)	Thickness (nm)
PEDOT:PSS	1.43×10^{-3}	~120
Ti_3C_2 :PEDOT:PSS	1.58×10^{-3}	~120



Supplementary Figure 8. Current density versus voltage (J – V) characteristic curve of PTzBI-oF:PYF-T-o device with different contents of Ti_3C_2 in the PEDOT:PSS.

Supplementary Table 4. Photovoltaic parameters of PTzBI-oF:PYF-T-o devices with different contents of Ti_3C_2 in the PEDOT:PSS measured under AM1.5 illumination at 100 mW/cm^2

Device	Ti_3C_2 contents	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
PTzBI-oF:PYF-T-o	0%	0.86	24.94	72.80	15.61
	1%	0.86	25.31	72.14	15.70
	1.5%	0.86	25.53	73.84	16.21
	2%	0.86	25.60	76.43	16.83
	2.5%	0.86	25.92	73.71	16.43
	3%	0.86	26.11	71.02	16.06

Supplementary Table 5. Relevant parameters obtained from $J_{\text{ph}}-V_{\text{eff}}$ curves for PTzBI-oF:PYF-T-o devices with different AIL

Active layer ^a	AIL	J_{ph}^b (mA cm ⁻²)	J_{sat} (mA cm ⁻²)	G_{max} (m ⁻³ s ⁻¹)	η_{diss}
PTzBI-oF:PYF-T-o	PEDOT:PSS	23.07	23.54	1.47×10 ²⁸	0.980
	Ti ₃ C ₂ :PEDOT:PSS	24.09	24.85	1.55×10 ²⁸	0.985

Supplementary Table 6. Detailed GIWAXS (010) peak information in OOP of PEDOT:PSS with or without Ti₃C₂:PEDOT:PSS

Film	Peak	Peak location (Å ⁻¹)	π - π stacking distance (Å)	FWHM (Å ⁻¹)	Lamellar stacking distance (nm)
PEDOT:PSS	(010) in OOP	1.80	3.49	2.80	0.20
Ti ₃ C ₂ :PEDOT:PSS	(010) in OOP	1.86	3.38	1.95	0.29

Supplementary Table 7. Area covered by PEDOT, PSS and PEDOT/PSS ratio of PEDOT:PSS and Ti₃C₂:PEDOT:PSS hybrid AIL

AIL	PEDOT area (eV)	PSS area (eV)	PEDOT/PSS ratio
PEDOT:PSS	0.164	1.675	0.10
Ti ₃ C ₂ :PEDOT:PSS	0.204	1.507	0.14

Supplementary Table 8. Photovoltaic parameters of D18:L8-BO (1:1.2) and PM6:L8-BO (1:1.2) devices with and without Ti₃C₂ doping in the PEDOT:PSS interlayer measured under AM1.5 illumination at 100 mW cm⁻²

Device	Ti ₃ C ₂ content	<i>V</i> _{oc} (V)	<i>J</i> _{sc} (mA cm ⁻²)	FF (%)	PCE (%)
D18:L8-BO	0%	0.902	26.32	75.51	17.92
	2%	0.905	27.37	76.94	19.05
PM6:L8-BO	0%	0.872	26.71	66.48	15.49
	2%	0.882	28.31	69.23	17.33