## **Electronic Supplementary Information**

## 21.7% Efficiency Achieved in Planar *n-i-p* Perovskite Solar Cells via Interface Engineering with Water-Soluble 2D $TiS_2$

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Fig. S1 UV-vis spectra of  $SnO_2$  and  $SnO_2/2D$  TiS<sub>2</sub> on ITO substrates.



Fig. S2 AFM height images of (a) silicon wafer and (b) silicon wafer/2D TiS<sub>2</sub>. The scan area of both images is  $2 \times 2 \ \mu m^2$ .



Fig. S3 c-AFM images of (a) ITO/SnO<sub>2</sub> and (b) ITO/SnO<sub>2</sub>/2D TiS<sub>2</sub>. The scan area of both images is  $5 \times 5 \ \mu m^2$ .



Fig. S4 J-V characteristics of devices with configuration of  $ITO/(SnO_2 \text{ or } SnO_2/2D \text{ TiS}_2)/Al$ .



Fig. S5 J-V curves of Pero-SCs with different thicknesses of 2D TiS<sub>2</sub> as interlayer.

Thickness (nm)	<i>V</i> (V)	$J_{\rm sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
2	1.08	23.79	76.02	19.58
5	1.09	24.34	76.39	20.34
12	1.09	24.34	73.68	19.54

Table S1. Optimization of the thickness of 2D TiS<sub>2</sub> as interlayer in Pero-SCs.



Fig. S6 Surface potential images of (a) bare ITO, (b)  $ITO/SnO_2$  and (c)  $ITO/SnO_2/2D TiS_2$  films.



Fig. S7 J-V curves of Pero-SCs for the different delay time.

Table S2. Device	parameters of	of Pero-S	Cs for the	different	delay	time.
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Condition	$V_{\rm oc}$ (V)	$J_{\rm sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
1 ms, FS	1.08	24.44	73.2	19.29
1 ms, RS	1.09	24.45	76.9	20.40
10 ms, FS	1.05	24.46	71.9	18.39
10 ms, RS	1.06	24.42	76.3	19.72
100 ms, FS	1.04	24.41	71.3	18.12
100 ms, RS	1.05	24.40	76.2	19.57
1000 ms, FS	1.04	24.39	71.7	18.17
1000 ms, RS	1.05	24.41	76.3	19.56
10000 ms, FS	1.04	24.39	72.5	18.38
10000 ms, RS	1.05	24.41	76.9	19.63

**Table S3.** *n* and  $J_0$  extracted from the *J*-*V* curves in the dark.

ETL	п	$J_0 (\mathrm{mA} \mathrm{cm}^{-2})$
SnO <sub>2</sub>	2.47	6.5 × 10 <sup>-9</sup>
$SnO_2/2D TiS_2$	2.18	$6.6 \times 10^{-10}$
$SnO_2/2D$ $T1S_2$	2.18	$6.6 \times 10^{-10}$



Fig. S8 (a) UV-vis absorption spectra and (b) XRD patterns of Pero-TFs on different ETLs.

Table S4. The parameters of the equivalent circuit for Pero-SCs based on different ETLs.

ETLs	$R_{\rm s}(\Omega)$	$R_{ m tr}(\Omega)$	$C_1$ (nF)	$R_{\rm rec}(\Omega)$	$C_2$ (nF)
SnO <sub>2</sub>	24.3	45.9	3.3	52.9	7.1
SnO <sub>2</sub> /2D TiS <sub>2</sub>	27.6	17.4	4.0	111.1	3.5

Table S5. Lifetimes and weighted fractions fitted from the transient PL spectra.

ETL	$\tau_1$ (ns)	$f_1$ (%)	$\tau_2$ (ns)	$f_2$ (%)
SnO <sub>2</sub>	16.6	22.2	121.6	77.8
$SnO_2/2D TiS_2$	11.6	29.1	83.9	70.9



**Fig. S9** Time dependent PCE of Pero-SCs based on different ETLs. The test was conducted at dry air (10% relative humidity).