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SUPPORTING INFORMATION

Toward an Alternative Approach for Preparation of Low-Temperature Titanium Dioxide Blocking Underlayer for Perovskite Solar Cells

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SI-1. Photoelectron spectroscopy of underlayers

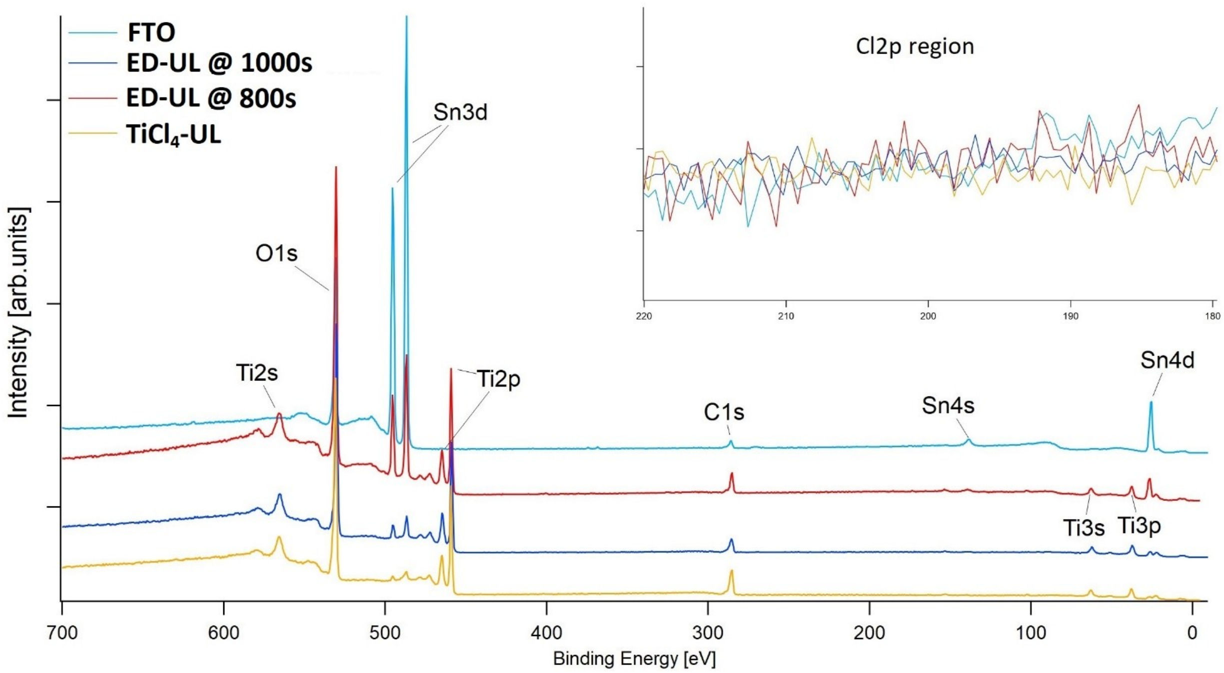

Fig. S1. Overview photoelectron spectra of TiO_2 underlayers compared to an FTO reference measured with a photon energy of 2100 eV. Inset: zoom on the spectral region where Cl 2p should be observed. No Cl is seen for UL samples confirming that no Cl from the precursors remains in the layers.

SI-2. UV-VIS Spectra of underlayers

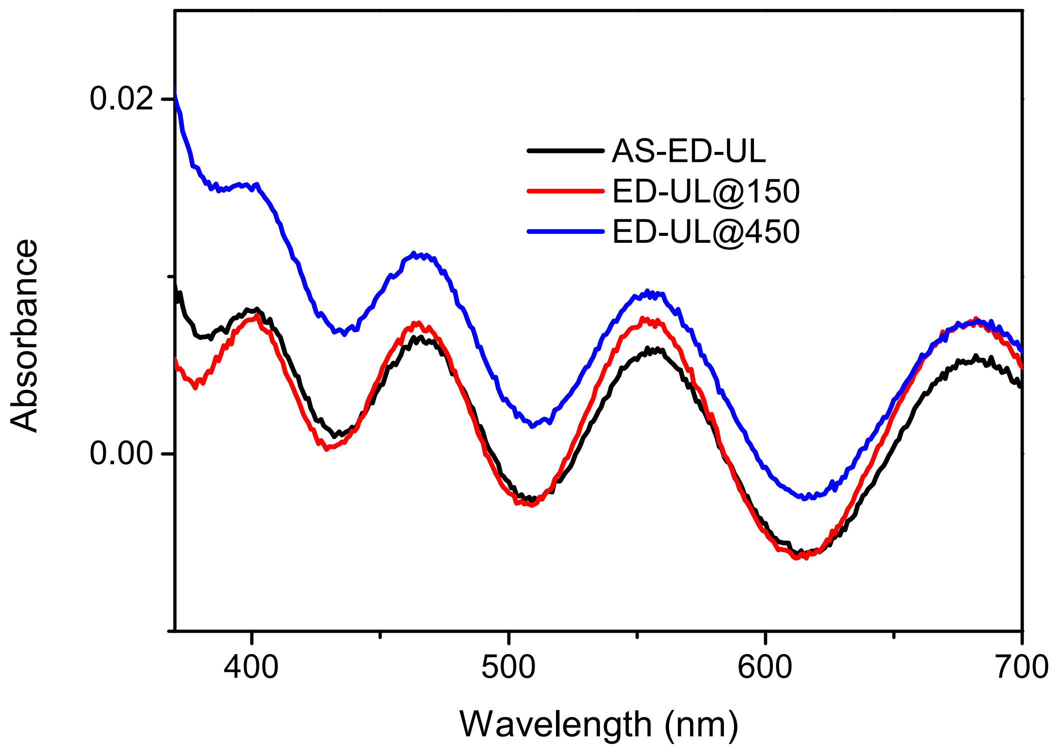

Fig. S2. UV-Vis absorption spectra of Ti (IV) oxo polymer immediately after electrodeposition (black), after sintering at 150 ^oC for 5 hours (red) and after sintering at 450 ^oC for half an hour (blue). The interference pattern is attributed to the FTO glass support.

SI-3. Statistical data of mesoscopic solar cell performance measurements.

Samples	T (˚C)	N	V_{OC} (V)	J_{SC} (mA cm^{-2})	*FF*	PCE (%)
SP-UL	450	30	1.069±0.037 (1.117)	23.5±0.5 (24.2)	0.75±0.02 (0.78)	19.0±1.0 (20.9)
ED-UL	450	30	1.067±0.0130 (1.121)	23.4±0.4 (24.1)	0.76±0.01 (0.78)	19.0±0.7 (20.5)

Electrodeposition time 1000s and electrodeposited underlayer sintering temperature 450 ˚C during 30min for all data. Best performance data in parenthesis.

V_{OC}	open-circuit photovoltage
J_{SC}	short-circuit current density
FF	fill factor
PCE (%)	solar to electricity efficiency
T:	sintering temperature
t_{dep}:	electrodeposition time
N:	number of samples
BEST	data for cells with best performance
AV	average
SD	standard deviation

SI-4. Incident photon-to-current conversion efficiency measurements: experimental conditions

Incident photon-to-current conversion efficiency (IPCE) measurements were performed with a setup including a Xenon light source (spectral products AB-XE-175), a monochromator (Spectral Products CM110), and a Keithly 2700 multimeter. The light intensity was calibrated with a certified reference solar cell (Fraunhofer ISE).

SI-5. Comparison of mesoporous solar cell short-circuit photocurrents and photocurrents obtained by integration of the IPCE spectrum.

Integrated current table and solar cell performance. These solar cells are different than the ones included in the main text.

Sample	*P* (mWcm^{-2})	J_{SC} (mAcm^{-2}) (device)	$J_{IPCE(1)}$ (mAcm^{-2}) integrated from IPCE spectrum for AM 1.5G irradiance 100 mWcm^{-2}	$J_{IPCE(2)}$ (mAcm^{-2}) integrated from IPCE spectrum for *P*	$J_{IPCE(2)}/J_{SC}$	V_{OC} (V)	FF	PCE (%)
SP-UL	100.6	24.0	23.2	23.4	0.97	1.113	0.74	19.7
ED-UL	100.4	23.7	23.4	23.5	0.99	1.124	0.74	19.7

Electrodeposited underlayer, sintering temperature 450 ˚C during 30min

SP-UL underlayer generated by spray pyrolysis
ED-UL underlayer generated by electrodeposition
P irradiance
V_{OC} open-circuit photovoltage
J_{SC} short-circuit current density
J_{IPCE} integrated photocurrent density
FF fill factor
PCE (%) solar to electricity efficiency

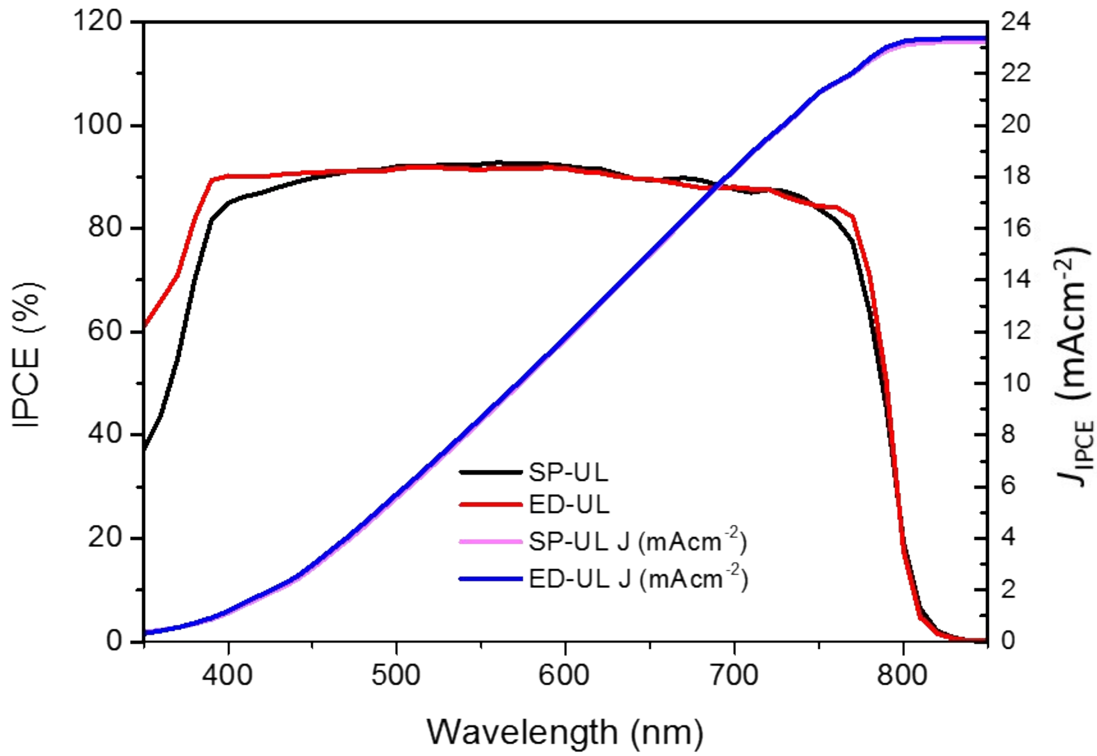

Fig. S5. IPCE spectrum and integrated photocurrent vs. wavelength

SI-6. Hysteresis data

Measurements were performed on cells different than these described in the main text.

Hysteresis behaviors of devices with SP-ULs, ED-ULs @450°C under 100% sun. Note. ED-UL(A) refers prepared at room temperature and heated @450°C (30 mins).

		V_{oc}(V)	J_{sc}(mA/cm2)	FF	PCE	Δ (PCE)
SP-UL-01	BW	1.075	23.4	0.74	18.9	1.8
	FW	1.065	23.2	0.68	17.1	
SP-UL-02	BW	1.074	23.3	0.74	18.9	2.0
	FW	1.061	23.4	0.68	16.9	
SP-UL-03	BW	1.073	23.3	0.75	19.0	2.3
	FW	1.062	23.5	0.66	16.7	
ED-UL-01	BW	1.069	23.4	0.74	18.8	2.5
	FW	1.062	23.4	0.65	16.3	
ED-UL-02	BW	1.070	23.4	0.74	18.8	2.5
	FW	1.061	23.4	0.65	16.3	
ED-UL-03	BW	1.058	23.6	0.73	18.5	3.2
	FW FS	1.058	23.5	0.61	15.3	

BW: Reverse direction scan, from open-circuit to short-circuit
FW: Forward scan, from short-circuit to open-circuit

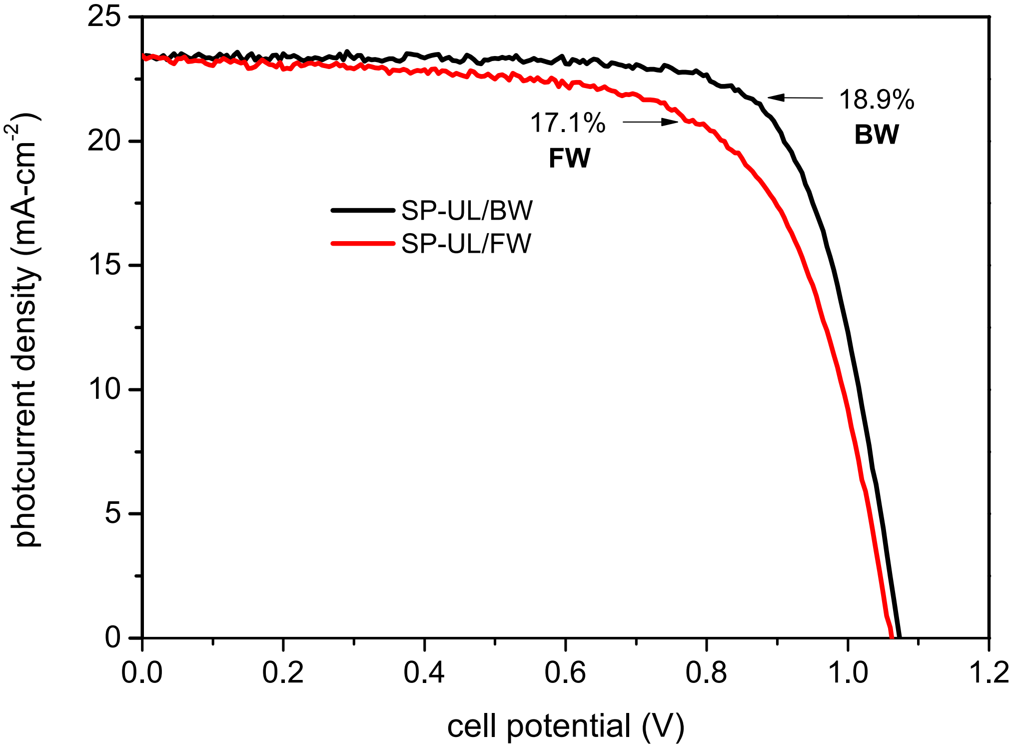

Fig. S6. Hysteresis behavior of SP-UL based mesoscopic -perovskite solar cells

Cell No SP-UL-03. BW: backward scan, from open-circuit to 0V. FW: forward scan, from 0V to open circuit.

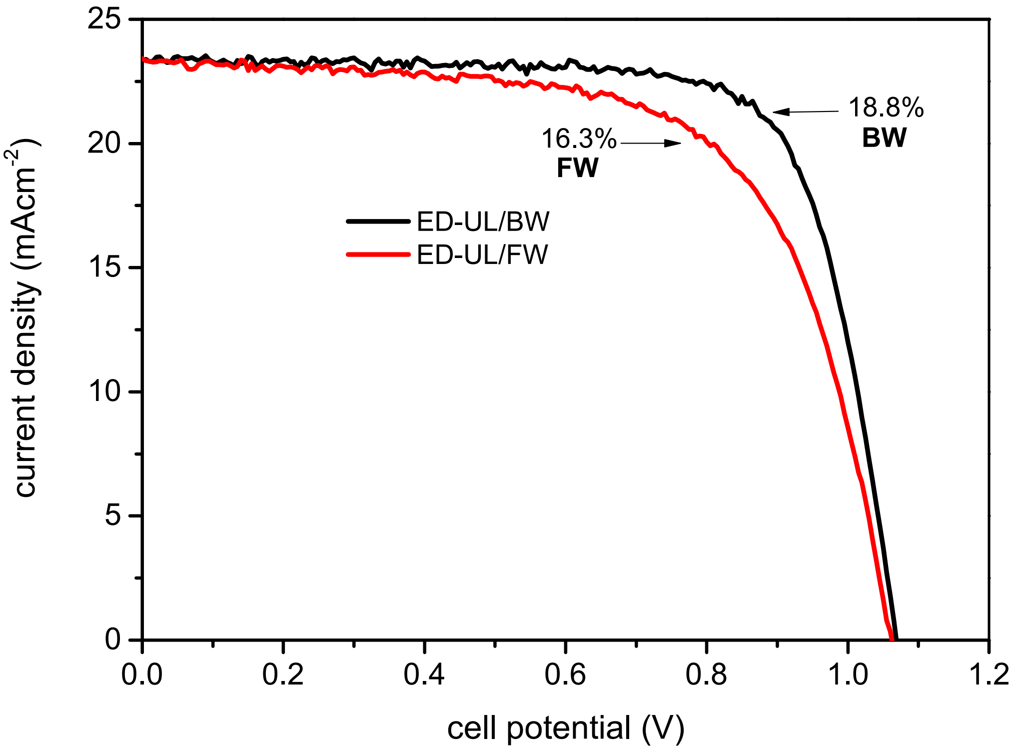

Fig. S7. Hysteresis behavior of ED-UL based mesoscopic-perovskite solar cell

Cell No ED-UL-01. BW: backward scan, from open-circuit to 0V. FW: forward scan, from oV to open circuit.

SI-7. Effect of electrochemical underlayer sintering time in the range 500s-1500s on the mesoscopic cell performance.

Sample	t_{dep}(s)	N	V_{oc} (V)	J_{sc}($mAcm^{-2}$)	FF	PCE(%)
SP-UL		3	1.109±0.008 (1.115)	23.8±0.1 (24.0)	0.75±0.00 (0.75)	19.6±0.2 (19.8)
ED-UL	500	3	1.068±0.003 (1.070)	23.7±0.1 (23.8)	0.70±0.02 (0.72)	17.6±0.4 (18.0)
ED-UL	800	3	1.111±0.003 (1.116)	23.6± 0.0 (23.6)	0.72± 0.00 (0.73)	18.7± 0.1 (18.7)
ED-UL	1000	5	1.116±0.006 (1.124)	23.6±0.1 (23.7)	0.75±0.01 (0.76)	19.7±0.078 (19.7)
ED-UL	1200	3	1.131±0.003 (1.135)	23.6± 0.1 (23.7)	0.73± 0.003 (0.74)	19.4± 0.1 (19.5)
ED-UL	1500	3	1.128±0.003 (1.133)	23.8± 0.1 (23.9)	0.73± 0.00 (0.74)	19.4± 0.1 (19.6)

Best performance data in parenthesis.

SI-8 Statistical data on planar perovskite cells.

Sample	Sintering Temp. (°C)	N	V_{oc} (V)	J_{sc}($mAcm^{-2}$)	FF	PCE(%)
SP-UL		10	1.059±0.036 (1.034)	19.2±2.7 (23.0)	0.68±0.04 (0.69)	15.9±1.2 (17.4)
ED-UL	150	10	1.040±0.060 (1.034)	19.2±2.7 (23.0)	0.56±0.11 (0.59)	11.5±2.8 (14.1)
ED-UL	450	10	1.059±0.036 (1.079)	21.9± 1.9 (23.5)	0.68± 0.04 (0.69)	15.9± 1.2 (17.4)

Best performance data in parenthesis.