Supporting Information

Charge transport layer dependent electronic band bending in perovskite solar cells and its correlation to light induced device degradation

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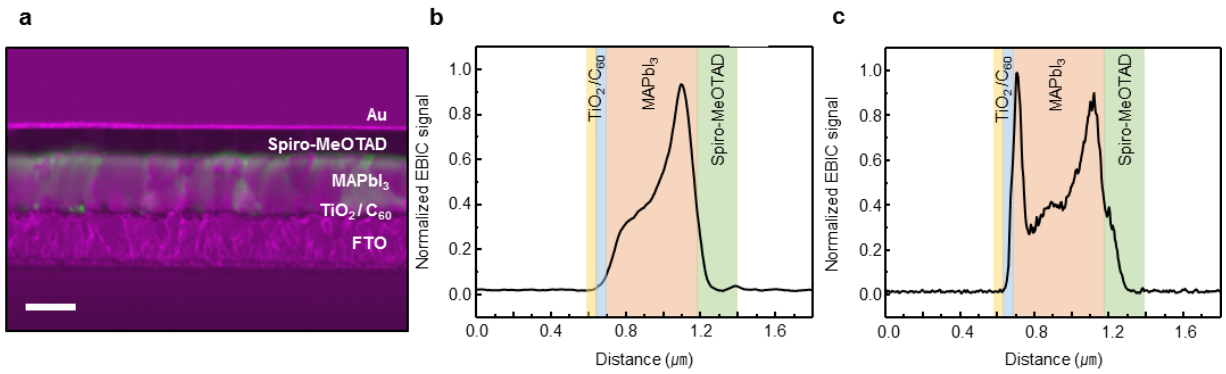


Figure S1. (a) Cross sectional EBIC image overlapped with SEM image of the TiO_2/C_{60}-based device. Line profiles of EBIC signal perpendicular to layers of the regions (b) without and (c) with a local pinhole in the C_{60} layer in the device. Scale bar: 500nm.

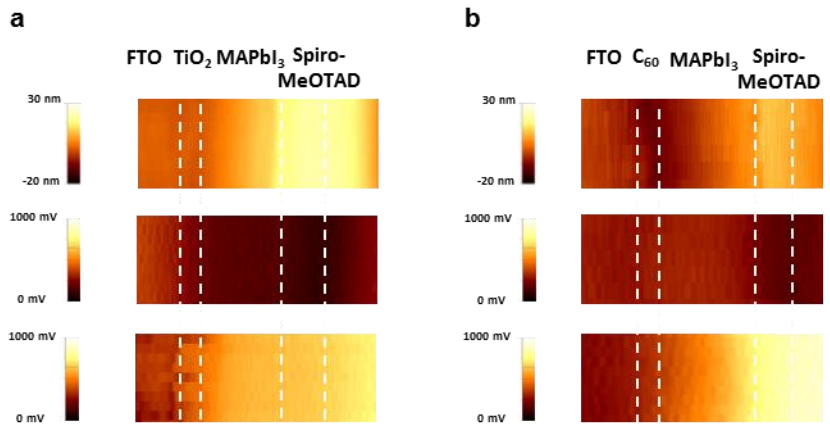
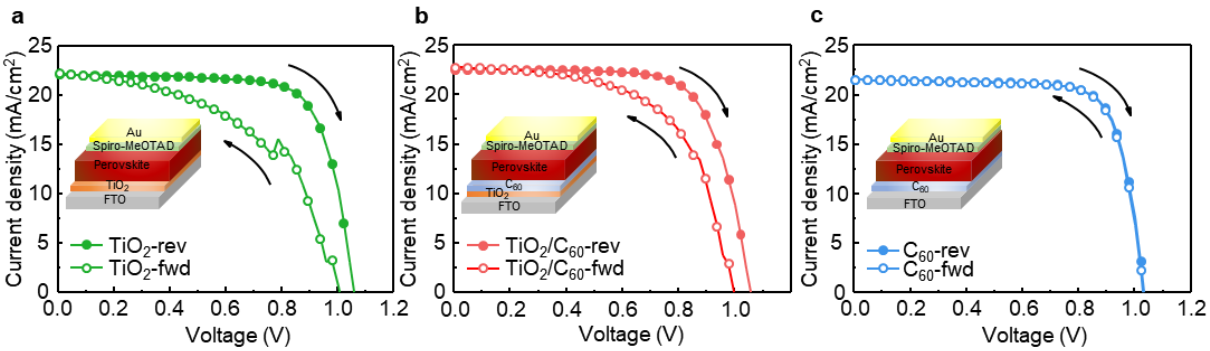


Figure S2. (a) A topographic image (Top), CPD distribution images under short-circuit (middle), and open circuit (bottom) conditions of the TiO_2-based device. (b) A topographic image (Top), CPD distribution images under short-circuit (middle), and open-circuit (bottom) conditions of the C_{60}-based device. RMS of roughness in the topographic images of the TiO_2-based and C_{60}-based devices are 9.32 nm, 5.55 nm, respectively.



	V_{oc} (V)	J_{sc} (mA/cm^2)	FF(%)	Efficiency(%)
TiO_2-rev	1.06	22.02	73.84	17.20
TiO_2-fwd	1.00	22.08	55.54	12.31
TiO_2/C_{60}-rev	1.02	22.25	76.72	17.38
TiO_2/C_{60}-fwd	1.00	22.30	65.93	14.64
C_{60}-rev	1.04	20.52	76.57	17.08
C_{60}-fwd	1.03	21.52	76.58	17.04

Figure S3. J-V characteristic of the (a) TiO_2-based, (b) TiO_2/C_{60}-based, (c) C_{60}-based devices measured in reverse (full circle) and forward (hollow circle) scan.

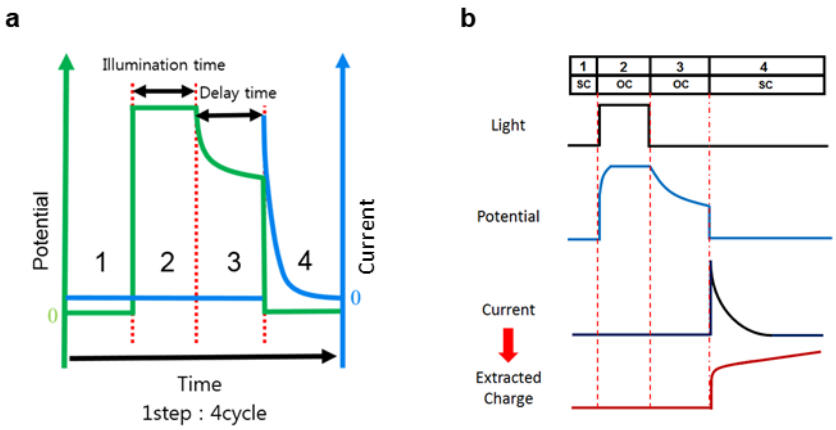
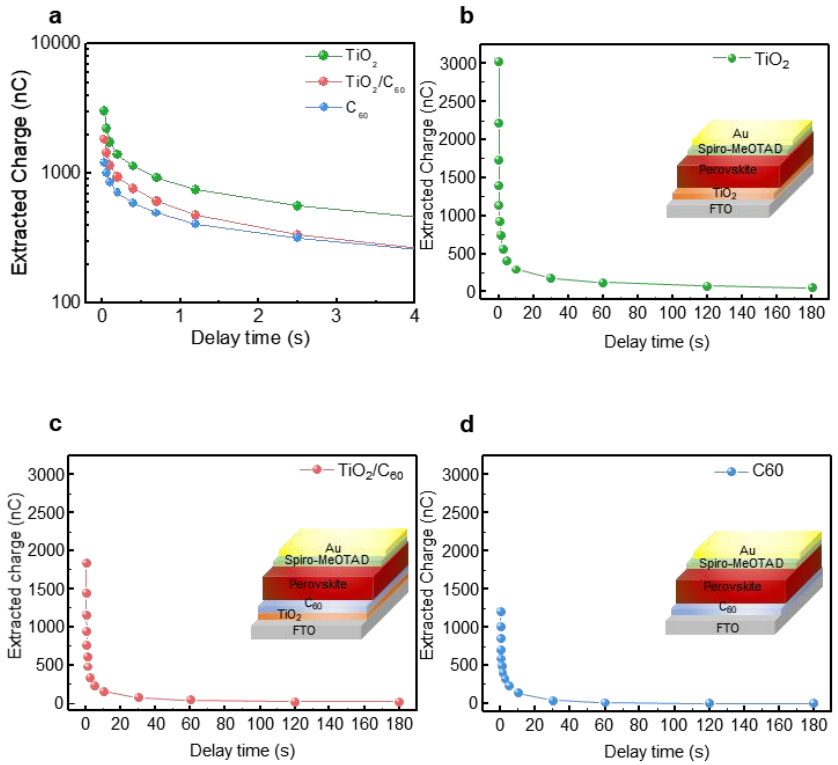


Figure S4. The sequence of the charge extraction measurement: 1) light off & short-circuit (SC) step in which charge remained in the device is extracted from the previous cycle; 2) light on & open-circuit (OC) step in which photocurrent is generated and carriers are recombined; 3) light off & OC step in which charge generation is stopped and accumulated charge inside the device is relaxed and recombined under certain delay time; 4) light off & SC step in which remnant accumulated charge is extracted. (a) An illustration of the potential and current values as a function of the steps, and (b) values of the extracted charge by integration of the current profile.



Q (nC)	0.03 s	0.06 s	0.1 s	0.4 s	1.2 s	5 s	10 s	30 s	60 s	180 s
TiO_2	3025.52	2213.63	1738.15	1134.74	747.73	410.59	296.04	183.12	124.87	56.33
TiO_2/C_{60}	1833.67	1448.08	1154.97	762.99	476.79	229.69	152.36	73.60	41.13	13.80
C_{60}	1207.13	1012.23	849.92	591.36	406.43	229.33	143.29	40.44	12.18	2.04

Figure S5. a) Quantity of extracted charges as a function of switching delay time up to 4s for the three different devices. Quantity of extracted charges delay time up to 180 s of b) the TIO_2-based device, c) TiO_2/C_{60}-based and d) C_{60}-based device, respectively.

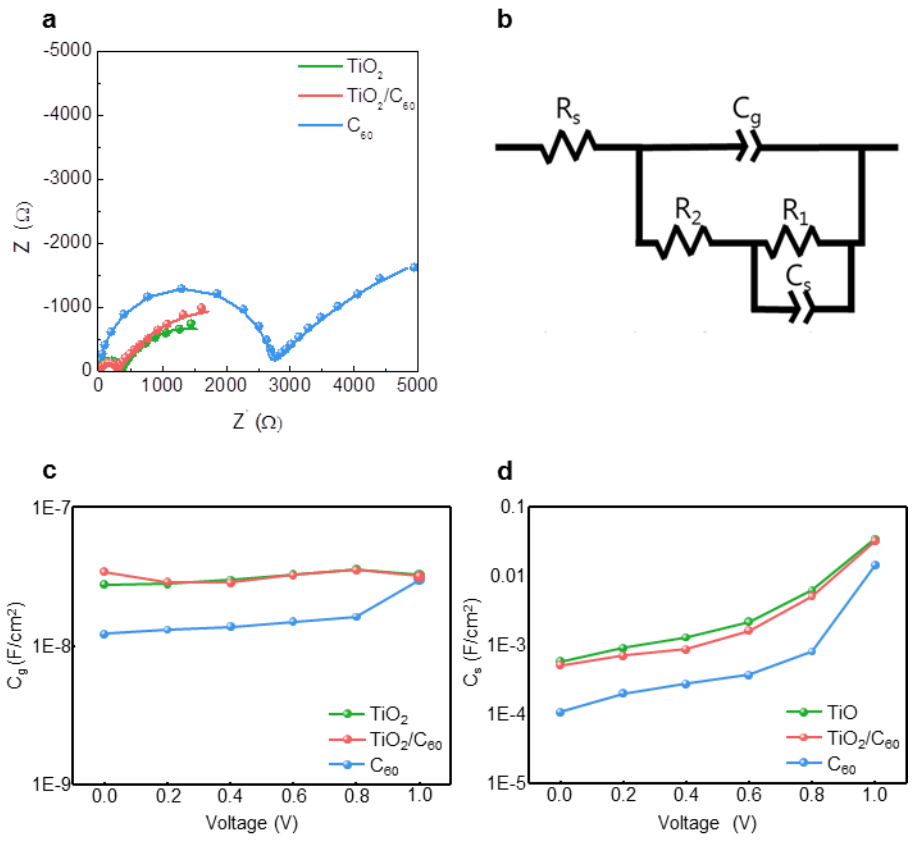


Figure S6. (a) Nyquist plots (Z”-Z’) of the PSCs with different ETLs. (b) An equivalent circuit employed in this study and (c) the values of the geometrical capacitance and (d) surface accumulation capacitance depending on the applied voltage. EIS measurements are performed under a LED light source and ambient conditions. Here, R_s is ohmic contribution of contacts and wires; C_g is geometrical capacitance, which is dielectric response of the perovskite layer; C_s is capacitance of surface charge accumulation at the interfaces; and R_1 and R_2 are the resistance of recombination current flux.

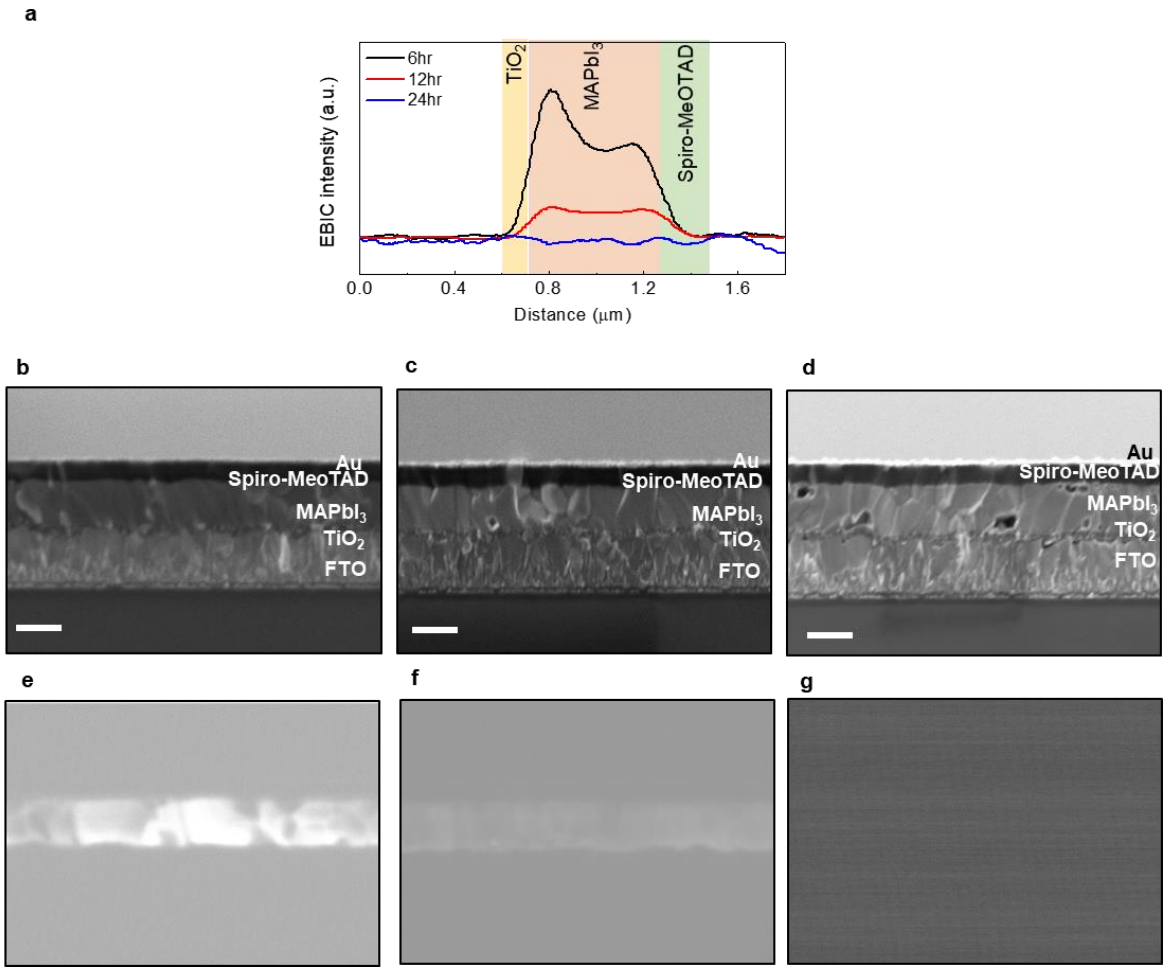


Figure S7. Time evolution of cross-sectional SEM and EBIC images of TiO_2-based device. a) Line profiles of EBIC signal perpendicular to the layers of the TiO_2-based device with different light soaking time. Cross-sectional SEM image of the TiO_2-based device after b) 6 hr under 1 sun light illumination, c)12 hr under 1sun light illumination and, d) 24 hr under 1 sun illumination. Cross-sectional EBIC image of the TiO_2-based device after e) 6 hr 1sun light illumination, f)12 hr 1sun light illumination and g) 24hr 1sun light illumination. All samples were stored under 1sun in air. Scale bars, 500nm

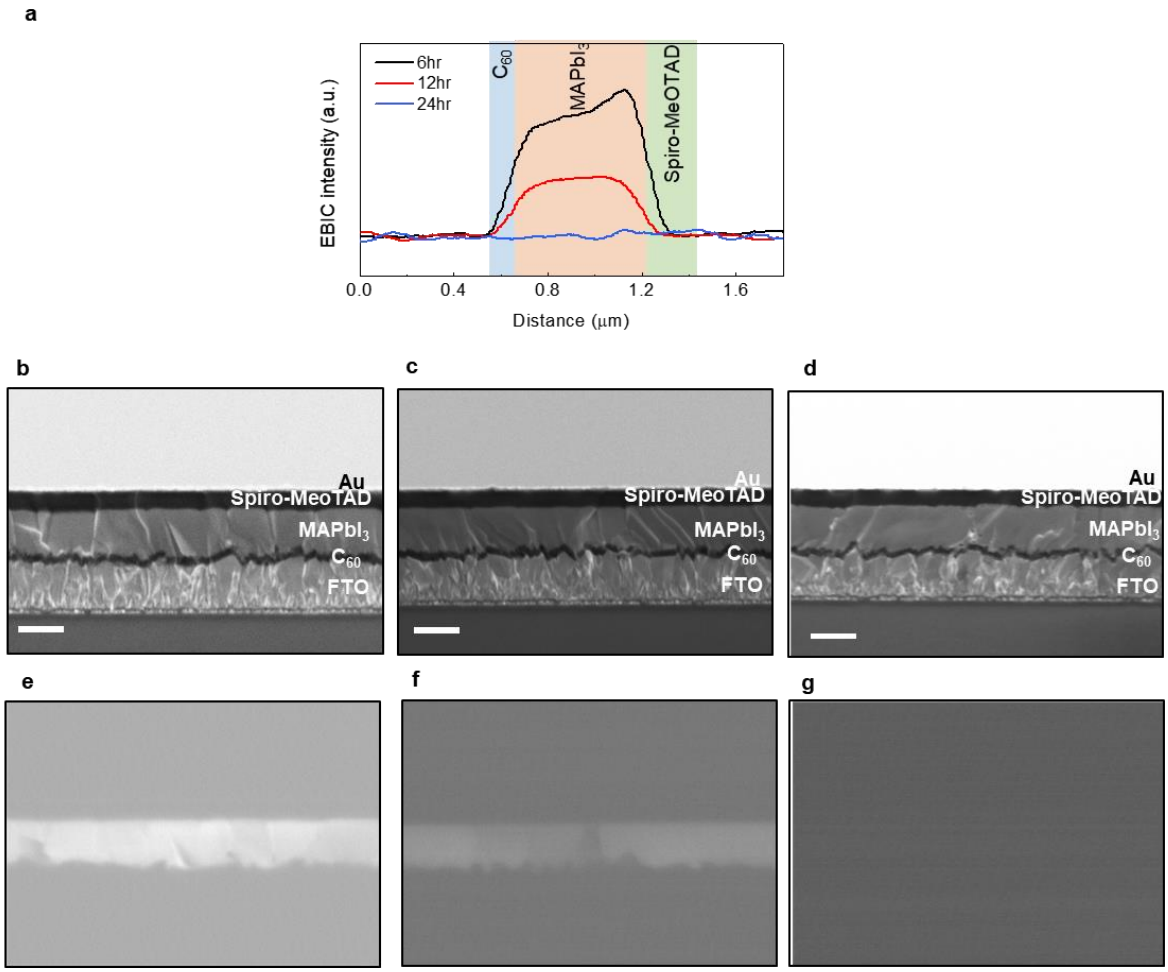


Figure S8. Time evolution of cross-sectional SEM and EBIC images of C_{60}-based device. a) Line profiles of EBIC signal perpendicular to the layers of the C_{60}-based device with different light soaking time. Cross-sectional SEM image of the C_{60} -based device after b) 6 hr under 1 sun light illumination, c)12 hr under 1 sun light illumination and, d) 24 hr under 1 sun illumination. Cross-sectional EBIC image of the C_{60}-based device after e) 6hr under 1sun light illumination, f)12 hr under 1sun light illumination and g) 24 hr under 1sun light illumination. All samples were stored under 1sun in air. Scale bars, 500 nm

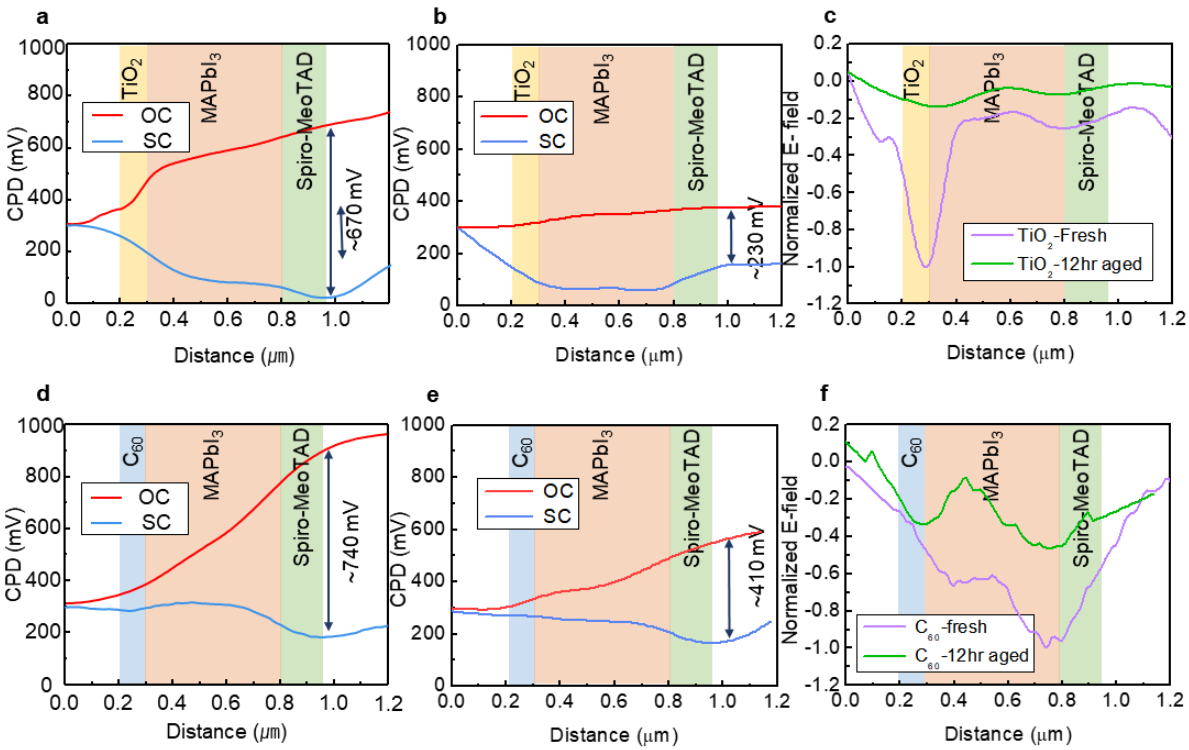


Figure S9. CPD distribution under illumination at open circuit condition (red line) and at short circuit condition (blue line) of fresh a) TiO_2-based and d) C_{60}-based device, respectively. CPD distribution under illumination at open circuit condition (red line) and at short circuit condition (blue line) of 12 hr aged b) TiO_2-based and e) C_{60}-based device, respectively. The arrow marks built up Voc. c) Normalized E-field distribution under illumination at open circuit condition of the fresh TiO_2-based (purple line) and 12 hr aged TiO_2–based device (green line). f) Normalized E-field distribution under illumination at open circuit condition of the fresh C_{60}-based (purple line) and 12 hr aged C_{60}–based device (green line).

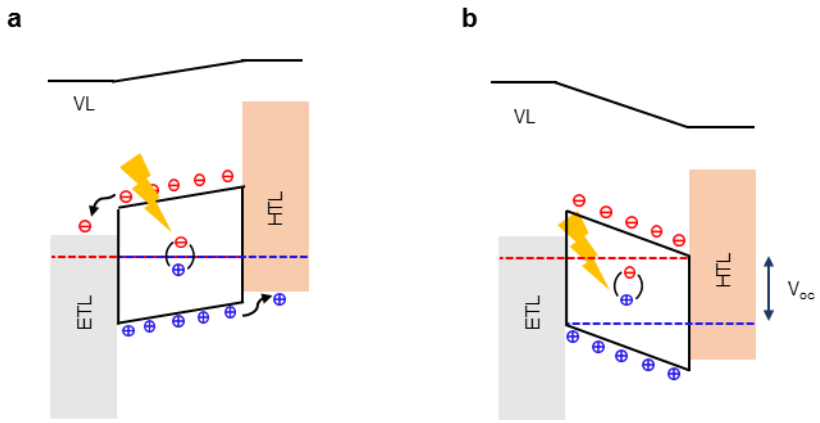


Figure S10. Schematic illustrations of band diagrams of device with ideal p–i–n junction type under light illumination in (a) short-circuit state and (b) open-circuit state. VL: vacuum level; ETL, electron-transporting layer; HTL, hole-transporting layer; V_{OC}, open-circuit voltage