ADVANCED ENERGY MATERIALS

Supporting Information

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A Simple Approach for Unraveling Optoelectronic Processes in Organic Solar Cells under Short-Circuit Conditions

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A Simple Approach for Unraveling Optoelectronic Processes in Organic Solar Cells Under Short-Circuit Conditions

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Table S1. Photoelectrical parameters of the solar cells used in this work under 1 sun solar simulator illumination.

	J_{SC} (mA/cm^2)	V_{OC} (V)	FF (%)	PCE (%)
PCE10:COTIC-4F	20.9 ± 0.1	0.57 ± 0.01	57.5 ± 0.6	6.8 ± 0.1
PCE10:IOTIC-2F$_a$	16.5 ± 0.4	0.80 ± 0.01	55.3 ± 0.1	7.3 ± 0.2
PM6:Y6	24.0 ± 0.9	0.78 ± 0.01	64.9 ± 2.7	12.1 ± 0.4

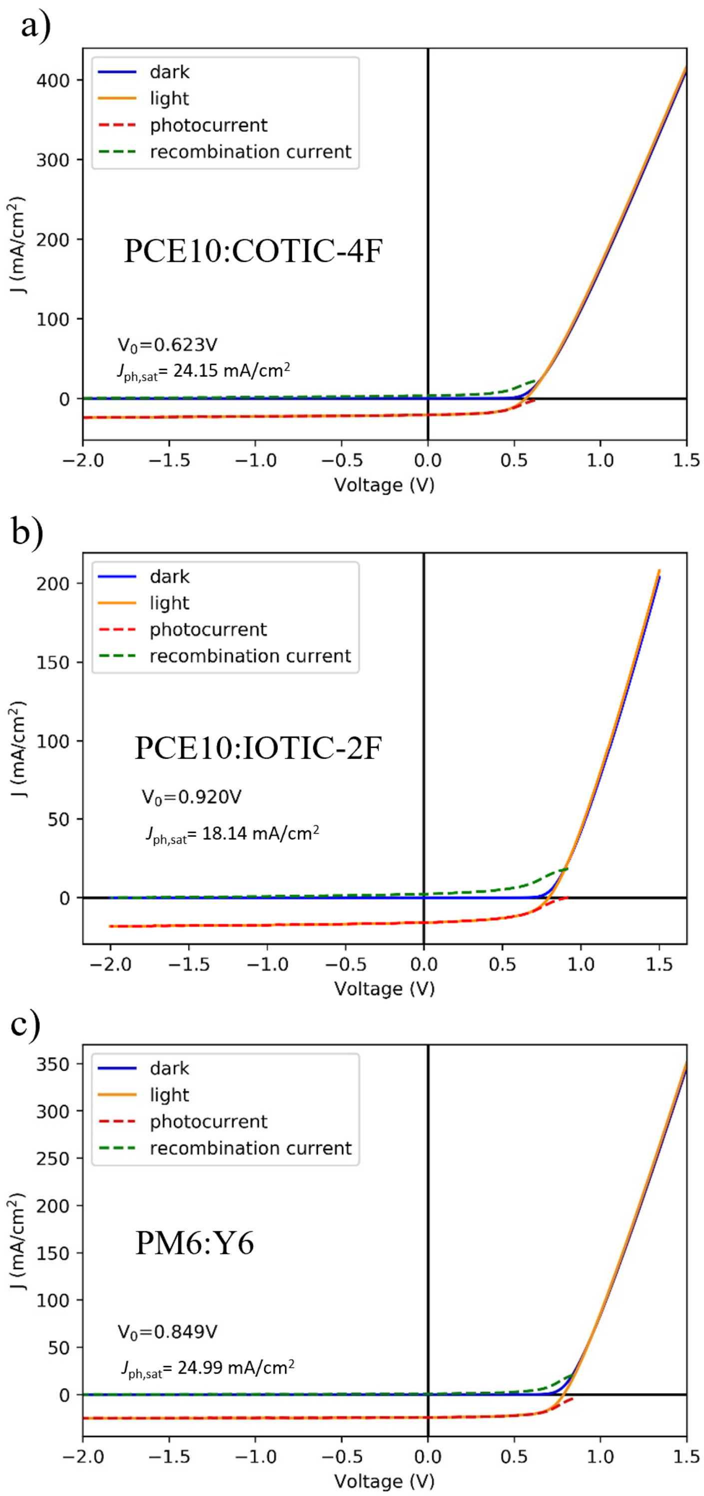

Figure S1. Current-voltage characteristics in the dark and under 1 sun illumination, photocurrent, saturated photocurrent and V_0 of a (a) PCE10:COTIC-4F, (b) PCE10:IOTIC-2F$_a$ and (c) PM6:Y6 solar cell.

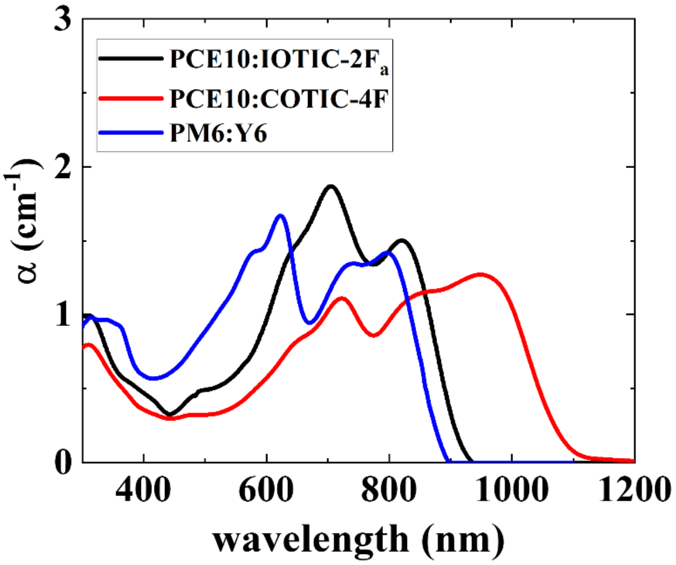

Figure S2. Absorption coefficient α of the three solar cell blends.

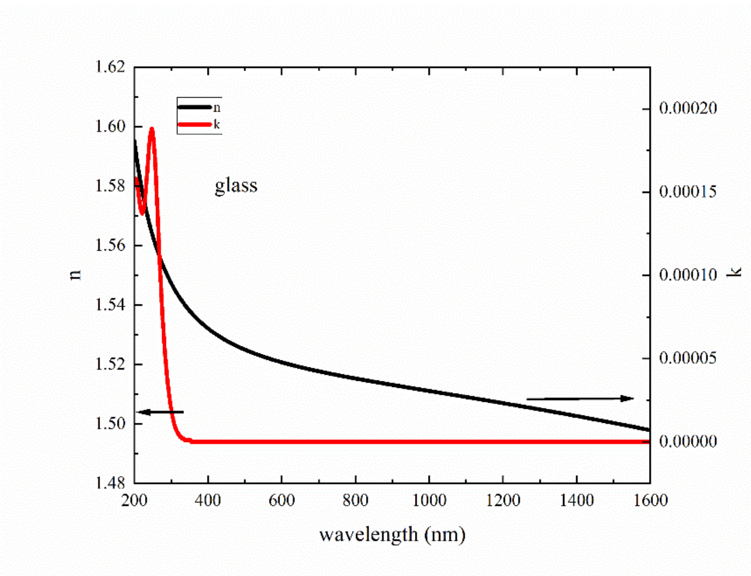

Figure S3. Optical constants of glass, determined with spectroscopic ellipsometry.

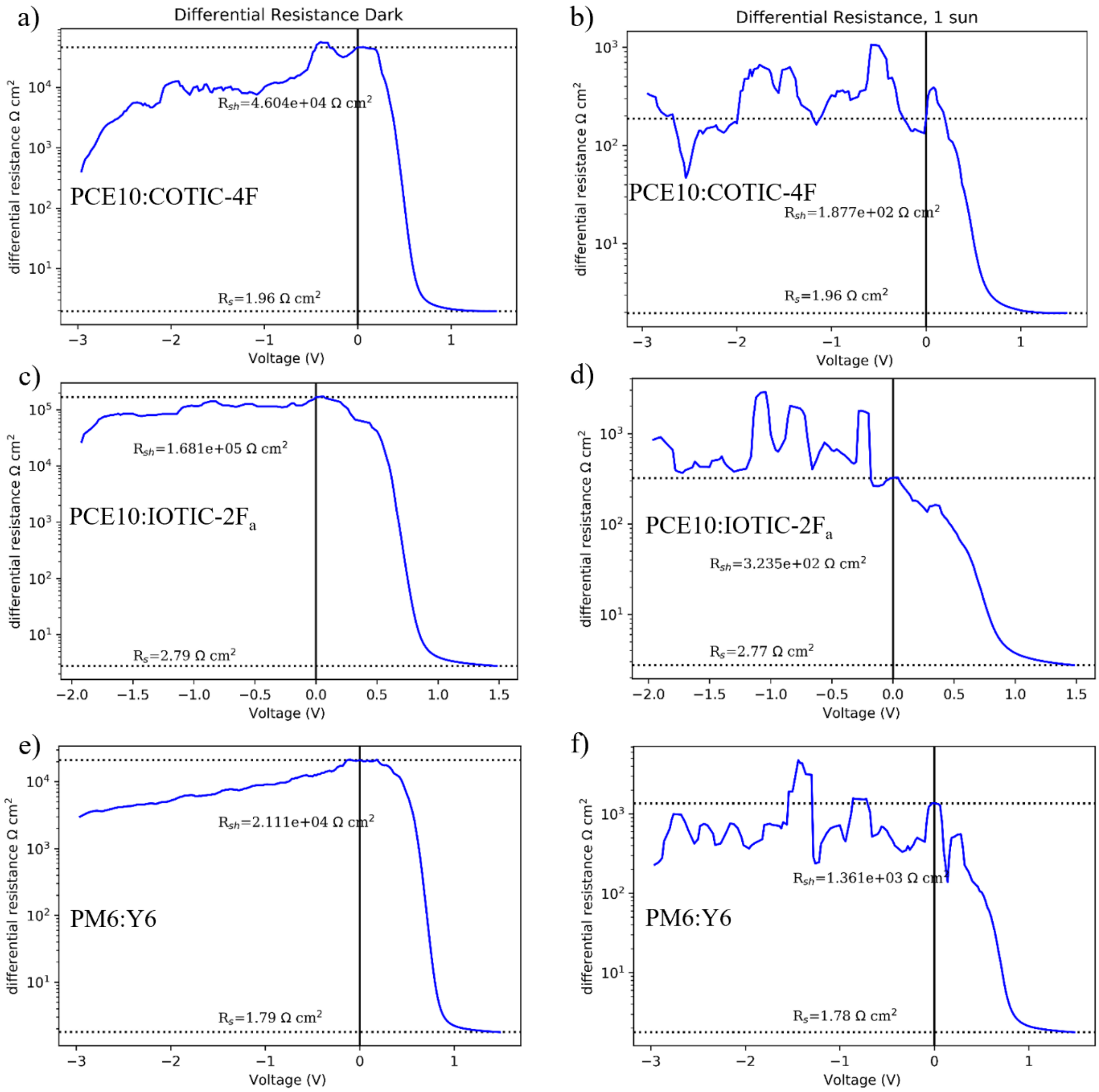

Figure S4. Differential Resistance of (a),(b) the PCE10:COTIC-4F solar cells in the dark and under 1 sun illumination, of (c),(d) the PCE10:IOTIC-2F$_a$ solar cells in the dark and under 1 sun illumination and (e),(f) the PM6:Y6 solar cells in the dark and under 1 sun illumination.

We determined the $\mu\tau$ at two illumination conditions (OD1 and 1 sun) since the illumination intensity of the used EQE setup is in between OD1 and 1 sun illumination. The average of the obtained values is displayed in Figure 5. The differential resistance at OD1 was assumed to be identical with the data for 1 sun for the analysis, which is justified by the minor changes observed comparing the differential resistance in the dark and under 1 sun illumination.

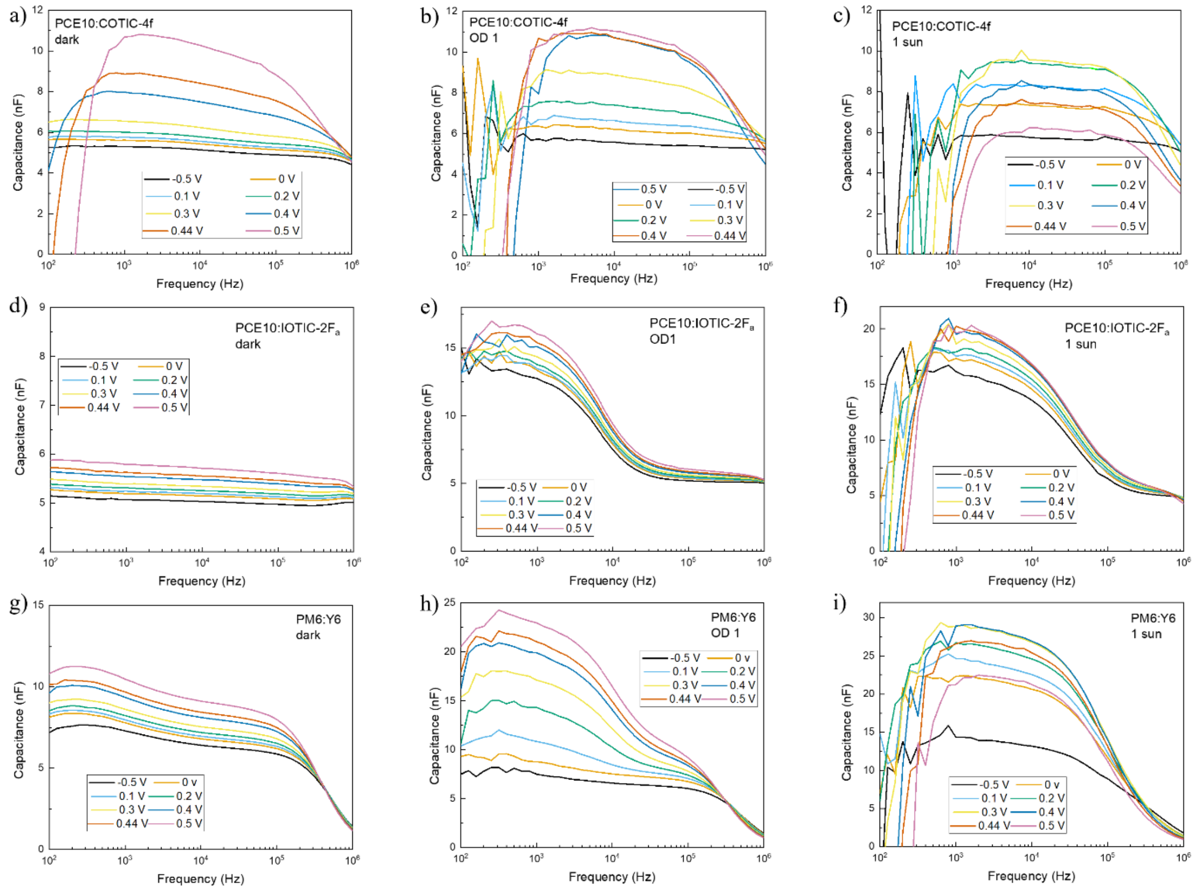

Figure S5. Frequency-dependent capacitance-voltage spectra of (a) a PCE10:COTIC-4F solar in the dark, (b) at an illumination of 0.1 sun and (c) at an illumination of 1 sun, (d) capacitance-voltage spectra of a PCE10:IOTIC-2F$_a$ solar in the dark, (e) at an illumination of 0.1 sun and (f) at an illumination of 1 sun,(g) capacitance-voltage spectra of a PM6:Y6 solar in the dark, (h) at an illumination of 0.1 sun and (i) at an illumination of 1 sun.

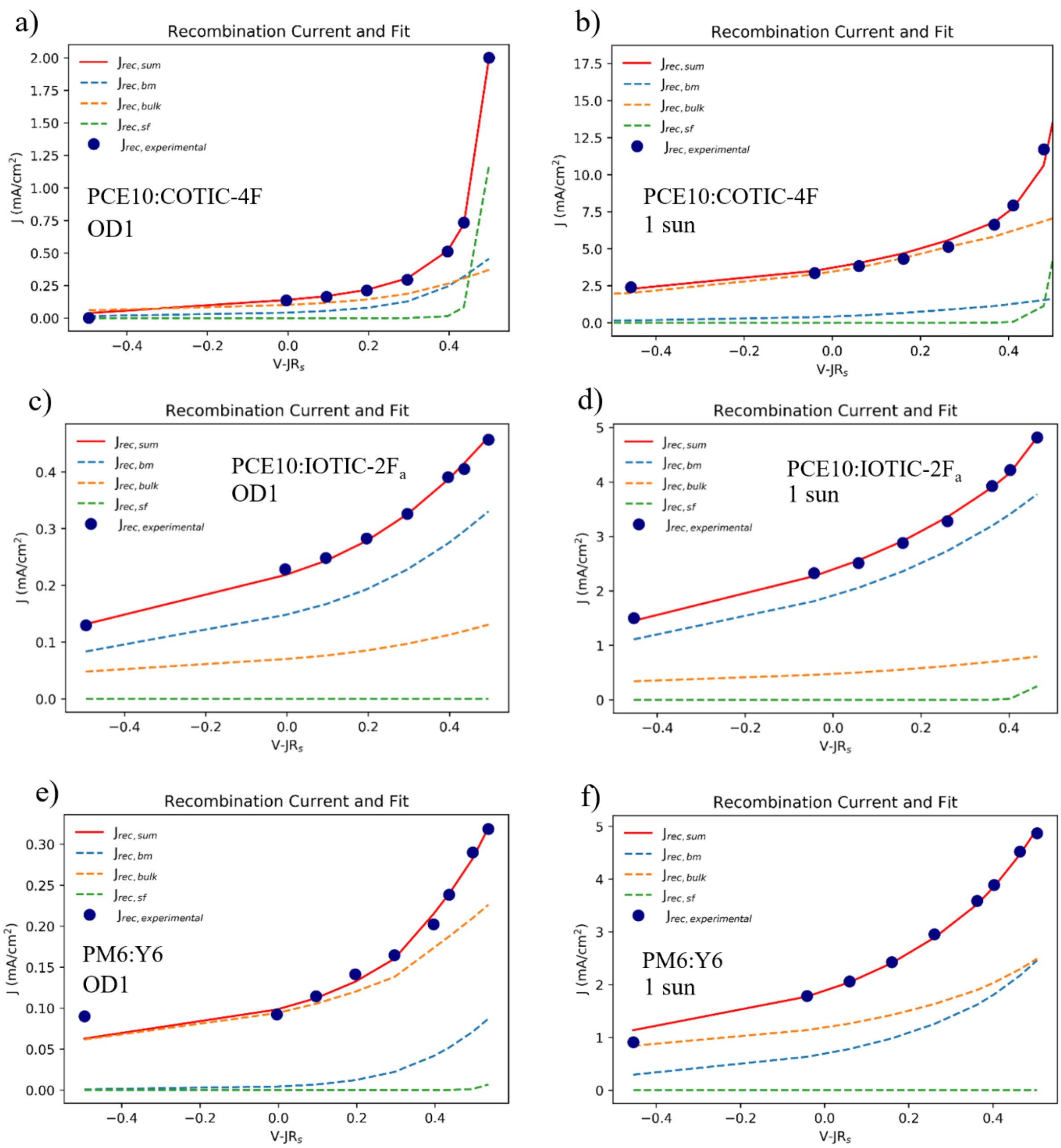

Figure S6. Experimental recombination current and fit of the recombination current based on the parameters determined by the analysis of the impedance spectroscopy data of (a) a PCE10:COTIC-4F solar cell under 0.1 sun and (b) 1 sun illumination, of (c) a PCE10:IOTIC-$2F_a$ solar cell under 0.1 sun and (d) 1 sun illumination and (e) a PM6:Y6 solar cell under 0.1 sun and (f) 1 sun illumination.

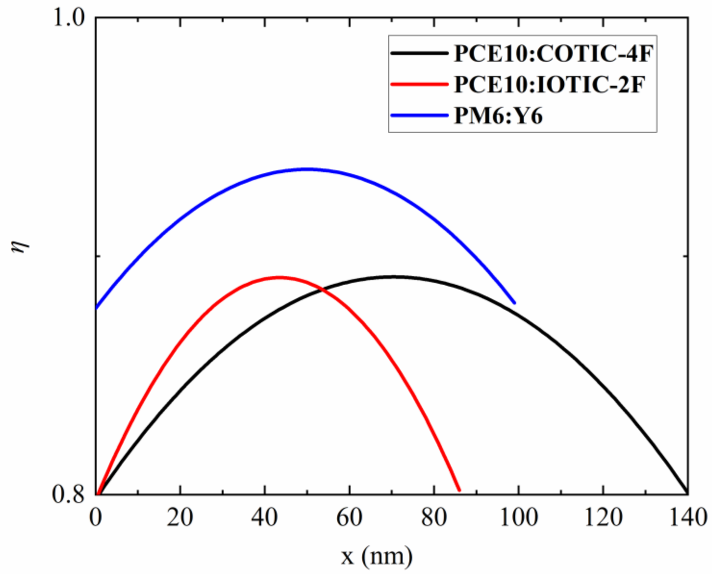

Figure S7. Spatially resolved and extraction efficiencies for the three investigated solar cells at 0 V and spatially averaged value of the extraction efficiencies $\bar{\eta}$.

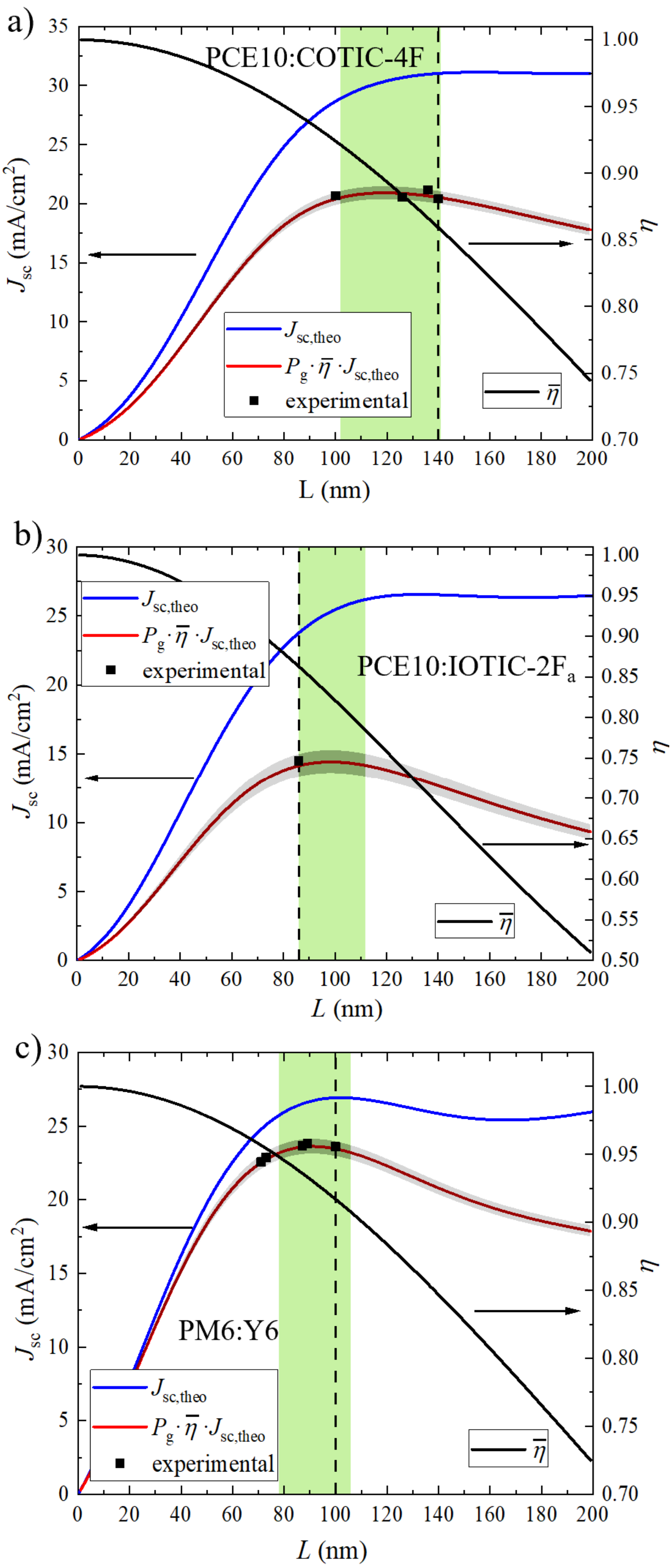

Figure S8. $J_{sc,theo}$, calculated based on optical transfer matrix method simulations, extraction efficiency $\bar{\eta}$ and the predicted J_{sc} as the product of P_g, $J_{sc,theo}$ and $\bar{\eta}$, in dependence on the active layer thickness L for the three studied systems, as well as experimental data points. The green range indicates the thickness range in which the highest J_{sc} is expected (98% of maximum). The dashed line indicates the active layer thickness that was used in this study.

Applicability to Fullerene Systems:
To demonstrate the applicability of the approach for fullerene systems, a P3HT:PC_{60}BM solar cell with the device architecture glass/ITO(130 nm)/ PEDOT:PSS (20 nm)/P3HT:PC_{60}BM(123 nm)/Al (63 nm) was fabricated and analyzed. The approach was carried out as described in the main text but the μτ was calculated from the EQE at 550 nm. We find good agreement between the impedance spectroscopy results, which were obtained in the same manner as described for the other systems, and those delivered by our approach for $\mu\tau$. The optical properties were used as reported in literature for films processed under the same conditions.[1]

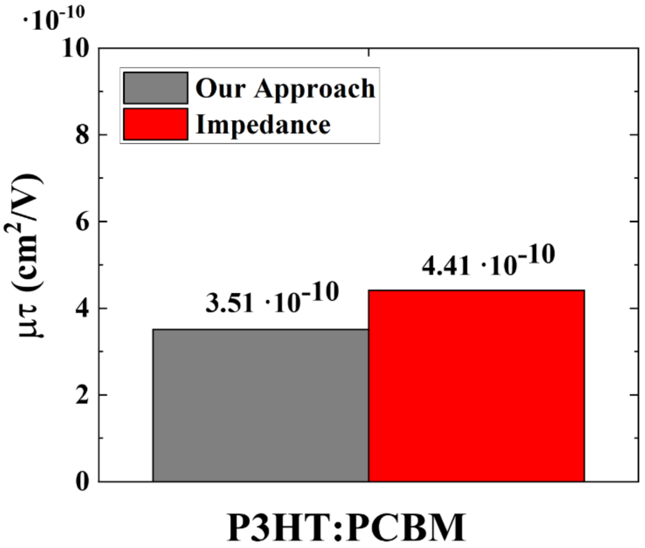

Figure S9. Comparison of the results obtained by our approach and by impedance spectroscopy for a P3HT:PCBM solar cell.

<u>Step-by Step Guide for the Application of our Approach to the Device Optimization:</u>

After obtaining the optical constants of the active layer and of other layers that are not provided in this work or other literature, the following steps lead to the prediction of the thickness-dependent $J_{sc.}$

<u>Experimental steps:</u>

1. Fabrication of at least one solar cell (thickness measurement should be done after completing the other experimental steps)
2. Measurement of the *JV* characteristics at 1 sun and in the dark (to obtain V_{bi})
3. Measurement of the EQE
4. Measuring *JV*-characteristics under monochromatic illumination

Simulation and calculation steps:

5. Simulating the $J_{sc,theo}$ under the monochromatic illumination used in step 4. (It is important to set other wavelengths to zero in the spectral distribution file.)
6. Calculation of P_g
7. Using the provided software (see below) to obtain $\mu\tau$ and the extraction efficiency η by fitting to measured *EQE*.
8. Calculation of the extraction probability $\eta(x)$ for each thickness L of interest, then averaging $\eta(x)$ to get extraction efficiency $\bar{\eta}$ for each L.
9. Simulation of the $J_{sc,ideal}$ for each L (no losses) with TMM under 1 sun AM 1.5 or the spectrum of your interest. (blue curve)

Product of $P_{g,}$ $\eta(L)$ and simulated $J_{sc,ideal}$ give predicted $J_{sc.}$ (red curve)

Software

The python-based software created for this work operates in conjunction with the transfer matrix method software that was developed by George F. Burkhard, Eric T. Hoke at Stanford University.[2]

Our part of the code includes:

- the visualization of the 3D Generation rate
- the calculation of the *EQE* in dependence on the position x in the device, the wavelengths and the $\mu\tau$
- fitting of the calculated *EQEs* to the experimental *EQE* and the output of the $\mu\tau$
- the calculation of the J_{sc} from measured *EQE* for AM1.5 illumination
- a visualization of the extraction efficiency $\eta(x)$ of electrons and holes at 0 V in dependence of the position in the device based on the determined $\mu\tau$
- a visualization of the extraction efficiency $\eta(x)$ of electrons and holes at -3 V in dependence of the position in the device based on the determined $\mu\tau$
- the average extraction efficiency $\bar{\eta}$ for different active layer thicknesses (required for device optimization)

The combined software can be accessed publicly and used under the GNU license agreement after the date of publication of this work. It can be found together with the optical constants provided in the manuscript under https://github.com/nschopp/Optoelectronic-Processes-in-Organic-Solar-Cells-Under-Short-Circuit-Conditions.

Supplemental References

[1] W. H. Lee, S. Y. Chuang, H. L. Chen, W. F. Su, C. H. Lin, *Thin Solid Films* **2010**, *518*, 7450.

[2] G. F. Burkhard, E. T. Hoke, M. D. McGehee, *Adv. Mater.* **2010**, *22*, 3293.