Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

SUPPLEMENTARY INFORMATION

Revealing the correlation between charge carrier recombination and extraction in an organic solar cell under varying illumination intensity

Abhishek Sharma[1,2,a], Mihirsinh Chauhan[3,a], Vishal Bharti[1], Manoj Kumar[4], Suresh Chand[1], Brijesh Tripathi[4,*], J. P. Tiwari[1*]

[1]Advanced Materials and Devices Division, CSIR-National Physical Laboratory New Delhi 110012 (India).

[2]Academy of Scientific and Innovative Research (AcSIR), CSIR-National Physical Laboratory, Dr. K. S. Krishnan Marg, New Delhi 110012, India

[3]Department of Solar Energy, School of Technology, Pandit Deendayal Petroleum University, Gandhinagar – 382007 (India).

[4]Department of Science, School of Technology, Pandit Deendayal Petroleum University, Gandhinagar – 382007 (India).

[a]Authors contributed equally to this work.

*Corresponding Authors: Ph. +91 79 2327 5455, +91-11-45608640, Fax +91 79 2327 5030, +91 45609310, Email: brijesh.tripathi@sse.pdpu.ac.in, jai_ti2002@yahoo.com, tiwarijp@mail.nplindia.org

Comparison between one diode and two diodes model

In this article two diode model has been used extensively to explain the experimental J-V curves of fabricated OSC as compared to conventional one diode model. Using both the models experimental J-V curves were fitted as shown in Fig. S1. The experimental data at low light intensity could not be accurately fitted with one diode model for experimentally determined reverse saturation current whereas two-diode fits well (curve in blue color in Fig. S1). This two diode model is more suitable due to the diffusion process of the excitons in the organic materials

[1, 4]. In OSCs exciton generation and consequently free electrons and holes creation by exciton dissociation is important under varying light intensity which affects the ideality factor.

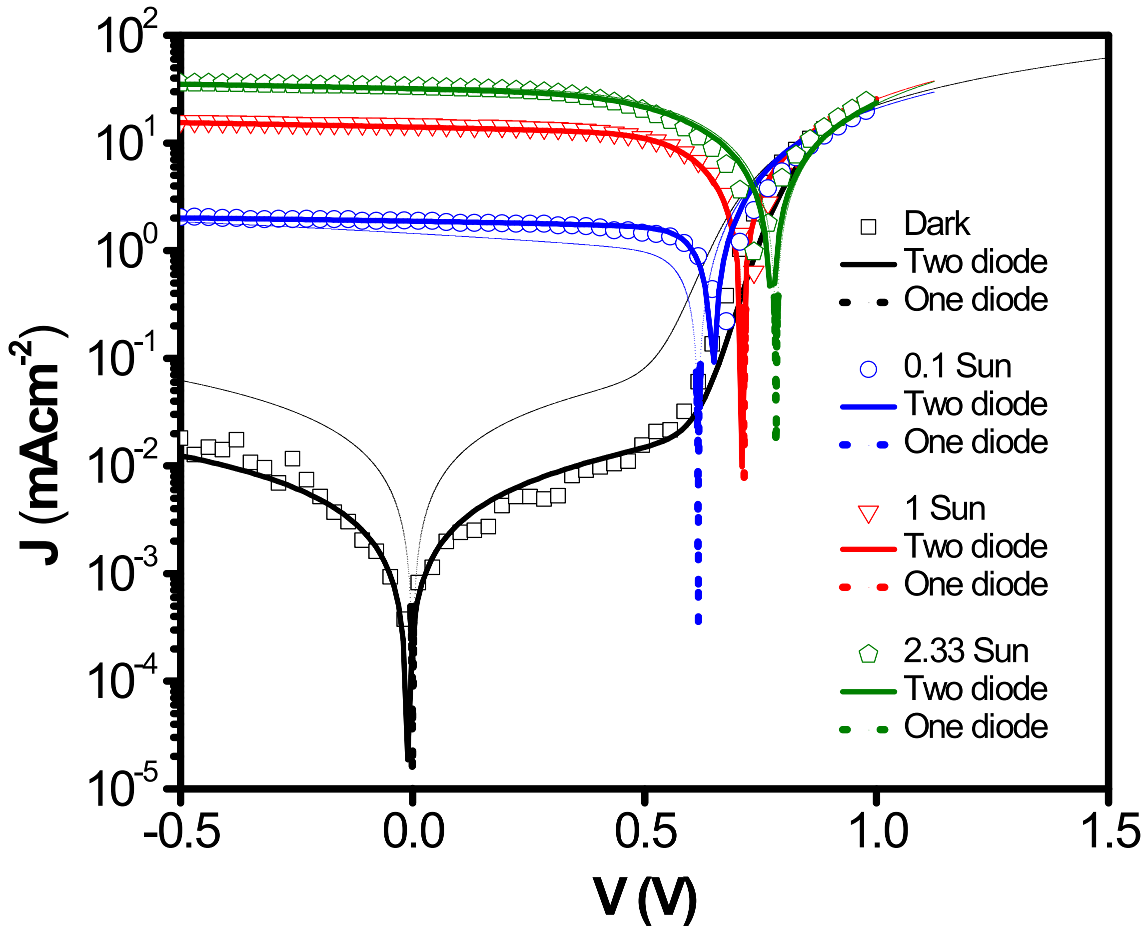

Figure S1: J-V curve of BHJ OSC. Symbols are for experimental data. Solid lines are fitting curves using the two-diode model. Dashed lines are fitting curves using the single-diode model.

Here, the effect of variation of parameters of both the models (one diode and two diode) on OSC performance as a function of irradiance intensity has been investigated. Under varying irradiance intensity only a single parameter has been varied at a time keeping all other parameters constant. The one diode and two diode model input parameters are listed in Table S1.

Table S1: Initial fitting parameter used for one diode and two diode model

Light Intensity ($mW \cdot cm^{-2}$)		**R_S ($\Omega \cdot cm^2$)**	**R_{SH} ($\Omega \cdot cm^2$)**	**J_{SC} ($mA \cdot cm^{-2}$)**	**V_{OC} (V)**	**n**	**m**
10	One diode	10.2	2700	1.84	0.630	2.12	-
	Two diode	10	2750	1.89	0.654	2.12	4.24
100	One diode	9.75	820	14.53	0.718	2.95	-
	Two diode	9.6	805	14.68	0.720	2.95	6.1
233	One diode	9.12	285	32.81	0.740	3.26	-
	Two diode	9	260	33.14	0.743	3.26	6.45

Initially, shunt resistance (R_{SH}) effects were investigated under varying light intensities using both the models and J-V curves were generated respectively (Fig. S2). The effect of R_{SH} on the open circuit voltage (V_{OC}) and efficiency (η) are shown in Table S2 and S4. It can be observed that at high light intensity (233 $mW \cdot cm^{-2}$ and 100 $mW \cdot cm^{-2}$) both the models show appropriate fitting with minimal deviation in V_{OC} and η. But at low irradiance intensity of 10 $mW \cdot cm^{-2}$, due to the small amount of generated currents, the resistive losses ($\sim J^2 R_S$) remain insignificant which results in larger deviation with the experimental values (deviation in V_{OC} = 6.06% and in η = 22.01%) by one diode model. Using two diode model we can fit this small amount of generated currents more appropriately with minimum change in V_{OC} (1.51% deviation) and η (0.62% deviation). Similarly, we investigated the influence of series resistance (R_S) on V_{OC} and η. The results of the plotted J-V curve (Fig. S3) are summarized in Table S3 and S5 and was concluded that two diode model is more appropriate compared to one diode model at varying illumination.

In this analysis it is found that the fitting of the ideality factor using one diode model does not match with experimentally extracted value from J-V curve below 1 sun illumination and lead to a wrong estimate of carrier recombination as well as reverse saturation current. Two-diode model is suitable to explain the experimental results as it matches with experimental data with a minimum error. This can be well described by two diode model which includes the dominance of recombination saturation current density (J_{02}) at lower biased region as well as dominance of diffusion current density (J_{01}) at higher biased region.

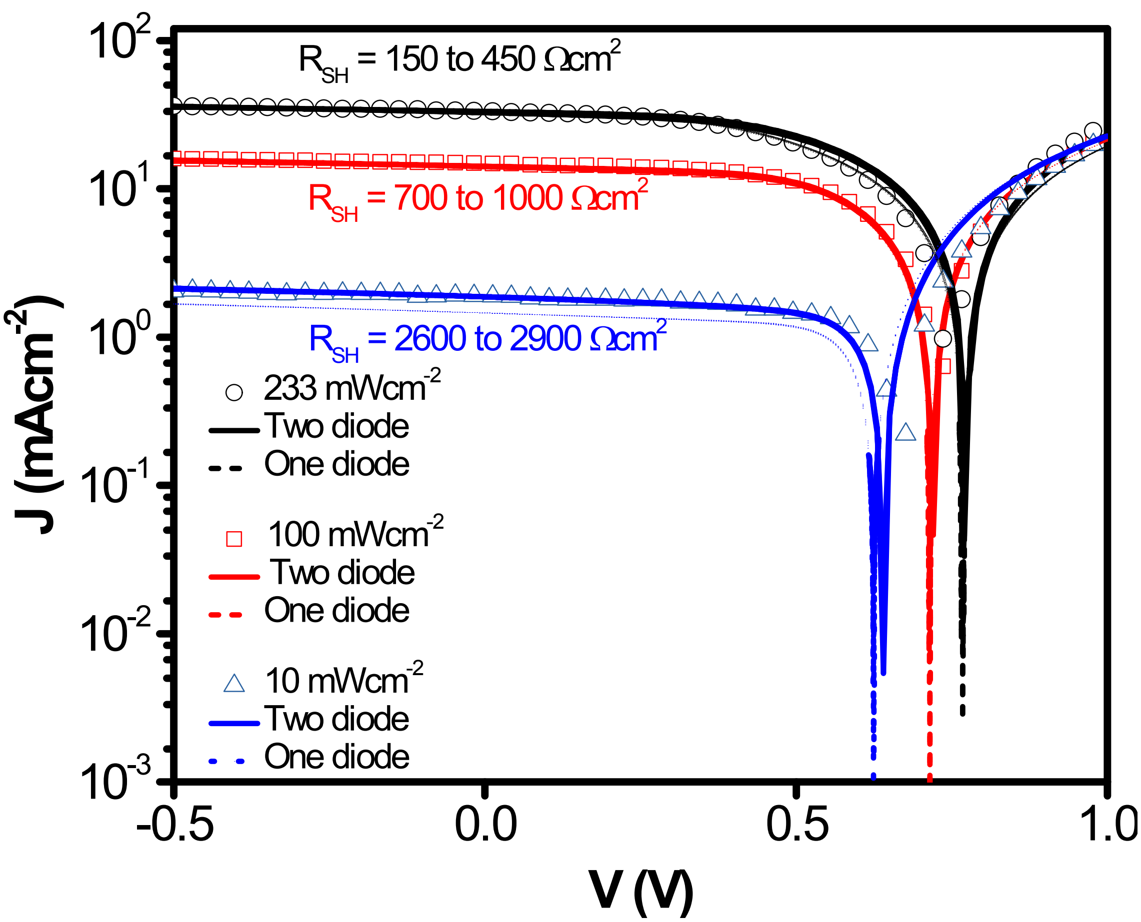

Figure S2: J-V curve of BHJ OSC for varying shunt resistance. Symbols are for experimental data. Solid lines are fitting curves using the two-diode model. Dashed lines are fitting curves using the single-diode model.

Table S2: Error analysis for R_{SH} (varied in the range of ± 150 Ω·cm² in order to check the validity of both models) input parameters in term of deviation in V_{OC} of organic solar cell

Light Intensity (mW·cm⁻²)	**R_{SH} (Ω·cm²)**	**V_{OC} (Exp.) (V)**	**V_{OC} (One-diode) (V)**	**V_{OC} (Two-diode) (V)**	**% deviation in V_{OC} using One-diode**	**% deviation in V_{OC} using Two-diode**
10	2750	0.660	0.62 ± 0.01	0.65 ± 0.01	± 6.06	± 1.51
100	850	0.721	0.72 ± 0.01	0.72 ± 0.01	± 0.13	± 0.13
233	300	0.744	0.74 ± 0.01	0.742 ± 0.01	± 0.53	± 0.26

Table S3: Error analysis for R_S input parameters in terms of deviation in V_{OC} of organic solar cell

Light Intensity ($mW\cdot cm^{-2}$)	R_S ($\Omega\cdot cm^2$)	V_{OC} (Exp.) (V)	V_{OC} (One-diode) (V)	V_{OC} (Two-diode) (V)	% deviation in V_{OC} using One-diode	% deviation in V_{OC} using Two-diode
10	10 ± 1	0.660	0.640 ± 0.01	0.655 ± 0.01	± 3.03	± 0.75
100	10 ± 1	0.721	0.725 ± 0.01	0.723 ± 0.01	± 0.55	± 0.13
233	10 ± 1	0.744	0.747 ± 0.01	0.743 ± 0.01	± 0.40	± 0.13

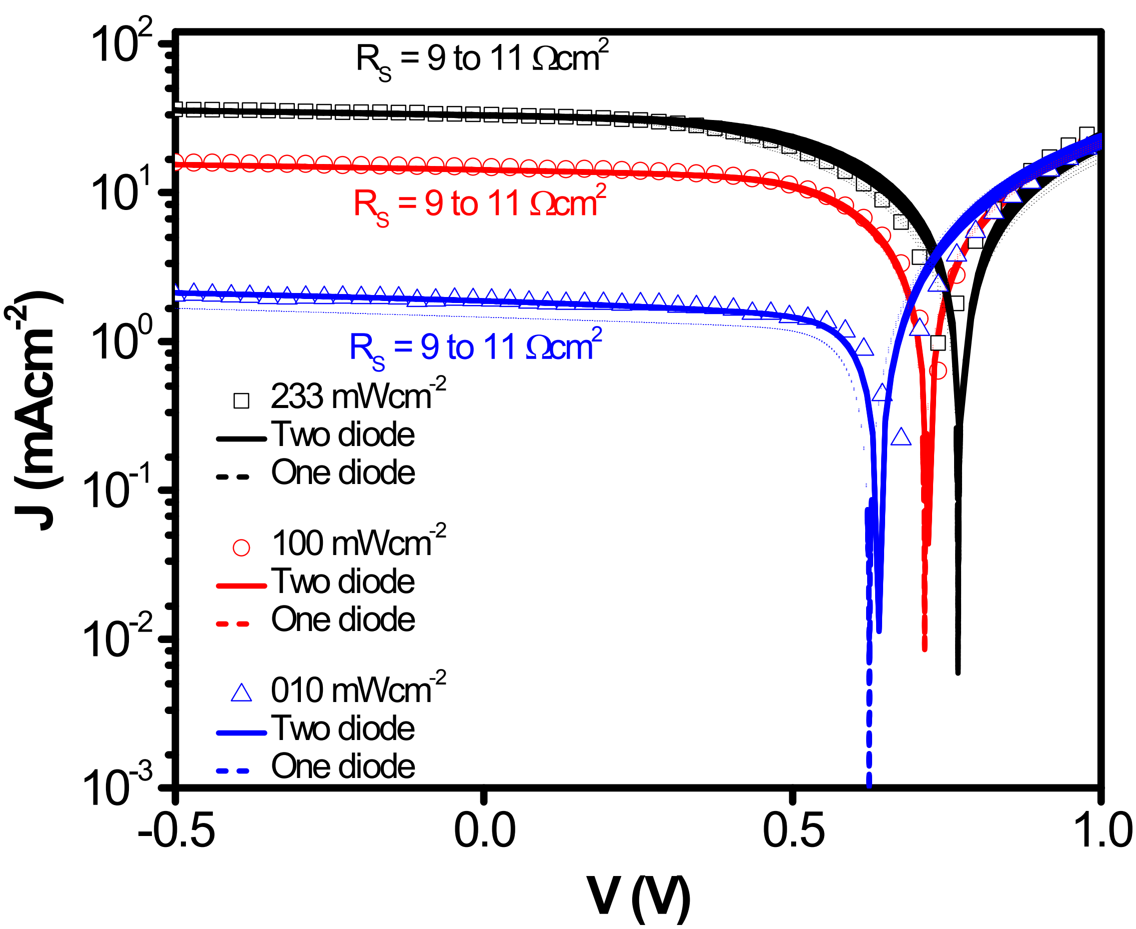

Figure S3: J-V curve of BHJ OSC for varying series resistance. Symbols are for experimental data. Solid lines are fitting curves using the two-diode model. Dashed lines are fitting curves using the single-diode model.

Table S4: Error analysis for R_{SH} (varied in the range of $\pm$ 150 $\Omega\cdot cm^2$ in order to check the validity of both models) input parameters in terms of deviation in η of organic solar cell

Light Intensity ($mW\cdot cm^{-2}$)	R_{SH} ($\Omega\cdot cm^2$)	η (Exp.) (%)	η (One-diode) (%)	η (Two-diode) (%)	% deviation in η using One-diode	% deviation in η using Two-diode
10	2750	7.95	6.2 $\pm$ 0.5	7.90 $\pm$ 0.5	$\pm$ 22.01	$\pm$ 0.62
100	850	5.52	5.4 $\pm$ 0.15	5.50 $\pm$ 0.07	$\pm$ 2.17	$\pm$ 0.36
233	300	4.40	4.5 $\pm$ 0.05	4.45 $\pm$ 0.08	$\pm$ 2.27	$\pm$ 1.13

Table S5: Error analysis for R_S input parameters in terms of deviation in η of organic solar cell

Light Intensity ($mW\cdot cm^{-2}$)	R_S ($\Omega\cdot cm^2$)	η (Exp.) (%)	η (One-diode) (%)	η (Two-diode) (%)	% deviation in η using One-diode	% deviation in η using Two-diode
10	10 $\pm$ 1	7.95	6.40 $\pm$ 0.5	7.92 $\pm$ 0.5	$\pm$ 19.49	$\pm$ 0.37
100	10 $\pm$ 1	5.52	5.45 $\pm$ 0.15	5.53 $\pm$ 0.05	$\pm$ 1.26	$\pm$ 0.18
233	10 $\pm$ 1	4.40	4.56 $\pm$ 0.05	4.42 $\pm$ 0.04	$\pm$ 3.63	$\pm$ 0.45

REFERENCES

1. N. v. Reich, W. Van Sark, E. Alsema, R. Lof, R. Schropp, W. Sinke and W. Turkenburg, *Solar Energy Materials and Solar Cells*, 2009, 93, 1471-1481.
2. K. Miyano, N. Tripathi, M. Yanagida and Y. Shirai, *Accounts of chemical research*, 2016, 49, 303-310.
3. O. Breitenstein, *Opto-Electronics Review*, 2013, 21, 259-282.
4. N. Craciun, J. Brondijk and P. Blom, *Physical Review B*, 2008, 77, 035206.