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Effect of leakage current and shunt resistance on the light intensity dependence of organic solar cells

Christopher M. Proctor[1] and Thuc-Quyen Nguyen[2,3,a)]
[1]*Materials Department, University of California, Santa Barbara, California 93106, USA*
[2]*Department of Chemistry, Faculty of Science, King Abdulaziz University, Jeddah, Saudi Arabia*
[3]*Department of Chemistry and Biochemistry, University of California, Santa Barbara, California 93106, USA*

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In this report, we demonstrate that parasitic leakage currents dominate the current voltage characteristics of organic solar cells measured under illumination intensities less than one sun when the device shunt resistance is too low ($<10^6\,\Omega\,\mathrm{cm}^2$). The implications of such effects on common interpretations of the light intensity dependence of the solar cell open circuit voltage, fill factor, short circuit current, and power conversion efficiency are discussed in detail. © *2015 AIP Publishing LLC*.
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Measuring the light intensity dependence of the current-density voltage (*JV*) characteristics has proven to be a powerful tool for identifying the primary recombination loss mechanisms in organic photovoltaic (OPV) devices.[1–3] Unlike other opto-electronic techniques for probing recombination mechanisms, light dependent *JV* studies do not require extensive experimental equipment or expertise. A solar testing setup (light source and *JV* measuring unit) and a series of neutral density filters or other means to attenuate the light intensity are all that is needed.

Nonetheless, as we demonstrate here, one must be very careful to use high quality devices when studying the light intensity dependence of solar cells as the light dependence can be strongly influenced by parasitic leakage currents. Such losses are well known to effect all types of solar cells;[4–7] however, many reports in the OPV literature have seemingly overlooked the influence of leakage currents when interpreting light dependent behavior.

Leakage current in a solar cell can be considered as undesirable current that is injected from the electrodes prior to the turn on voltage. Within the operating regime (0 V to open circuit voltage), leakage current flows opposite to the photocurrent and thereby reduces the light current. This phenomenon is typically described using a simple circuit model (see, Figure 1(a)) in which leakage (shunt) current can travel through the shunt resistor (R_{sh}) that is in parallel to the photocurrent source and diode.[6] The magnitude of the leakage current is then determined by the magnitude of R_{sh} – the higher R_{sh}, the less current that runs through it.

From this model, it follows that

$$J(I,V) = J_d(V) + J_{sh}(V) - J_{ph}(I,V), \tag{1}$$

where J is the net output current density, J_d is the diode current density, J_{ph} is the photogenerated current density, and J_{sh} is the leakage current density that flows through R_{sh}. It is worth noting that J_D and J_{sh} depend only on voltage (V) while J_{ph} scales with the incident light intensity (I) as well and thus both J and J_{ph} are functions of V and I. Consequently, the relative influence of J_{sh} on J will increase at lower light intensities. In the event that J_{sh} is non-negligible, this can lead to significant decreases in both open circuit voltage (V_{oc}) and fill factor (FF) at low light intensities.

Following Ohm's law, the leakage current through R_{sh} can be expressed as

$$J_{sh} = \frac{V - JR_s}{R_{sh}}, \tag{2}$$

where R_s is the series resistance. For an organic solar cell, R_s is typically taken to be the inverse slope of the dark current

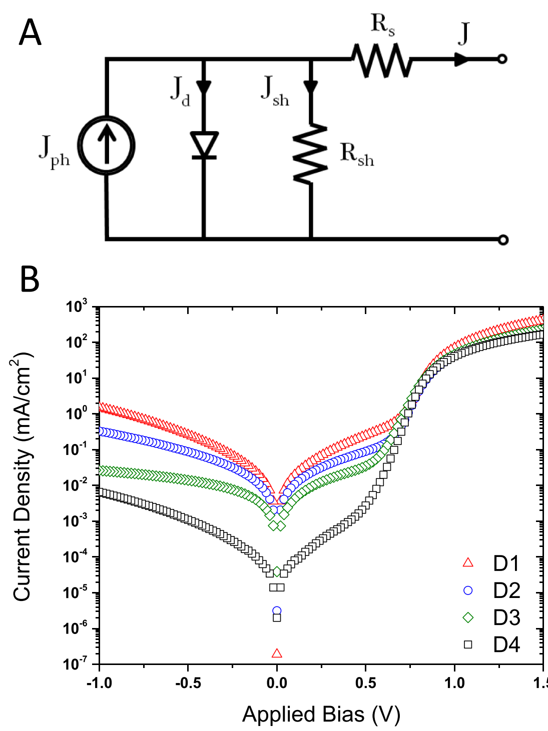

FIG. 1. (a) Simple circuit model showing current pathways and resistances in a typical solar cell. (b) The current-voltage response measured in the dark of *p*-DTS(FBTTh$_2$)$_2$:PC$_{71}$BM solar cell devices with leakage currents ranging from low to very high.

a)Email: quyen@chem.ucsb.edu

around $J(0\,\mathrm{mW/cm^2},\ 1.5\,\mathrm{V})$ and R_{sh} is the inverse slope around $J(0\,\mathrm{mW/cm^2}, 0\,\mathrm{V})$.

Figure 1(b) shows the dark current of four different organic solar cells with varying magnitudes of leakage current. The R_s and R_{sh} for each device are shown in Table I. The solar cell devices were prepared using the high performing solution processed small molecule system 7,7′-(4,4-bis(2-ethylhexyl)-4H-silolo[3,2-b:4,5-b′]dithiophene-2,6-diyl)bis(6-fluoro-4-(5′-hexyl-[2,2′-bithiophen]-5-yl)benzo[*c*][1,2,5]thiadiazole), (*p*-DTS $(FBTTh_2)_2$) as the donor material and phenyl-C71-butyric acid methyl ester ($PC_{71}BM$) as the acceptor following the optimal procedures described by Van der Poll *et al.*[8] Though devices were prepared from identical solutions and procedures, a large variation can be seen in the dark current with device D1 exhibiting orders of magnitude higher current from 0 to 0.75 V as compared to device D4. From Figure 1(b), it is evident that D1 has the highest leakage current followed by D2, D3, and D4. This is also reflected in the R_{sh}, where D1 has the lowest R_{sh} followed by D2, D3, and D4. Such large variation in the leakage current of organic solar cells is not uncommon and is known to be affected by substrate cleaning procedures, film thickness, electrode interlayers, and film deposition techniques.[5,9,10] In the case of the devices here, the range in R_{sh} is most likely a consequence of spin coating from a hot solution which led to variations in film thickness and film density which in turn affects the degree of cathode diffusion into the active layer.

The effect of leakage current (i.e., low R_{sh}) at open circuit conditions is of particular interest as the light intensity dependence of the open circuit voltage is often used to understand the nature of charge carrier recombination. It has previously been shown by Koster *et al.* that for an ideal system with only bimolecular recombination and negligible leakage current, the open circuit voltage can be expressed as

$$V_{oc} = \frac{E_{gap}}{q} - \frac{kT}{q}\ln\left(\frac{(1-P)\gamma N_c^2}{PG}\right), \tag{3}$$

where E_{gap} is the band gap, q is the elementary charge, k is Boltzmann's constant, T is temperature, P is the dissociation probability of a bound electron-hole pair, γ is the bimolecular recombination rate coefficient, N_c is the effective density of states, and G is the photogeneration rate.[2] As G is the only term in Eq. (3) that depends on light intensity Koster *et al.* predicted and confirmed that for a system with only bimolecular recombination the V_{oc} should have a logarithmic dependence on light intensity with a slope of kT/q.[2] Subsequently, it was shown that a system with trap-assisted recombination will have a slope greater than kT/q.[11] The light dependence of the V_{oc} has hence been used to distinguish bimolecular and trap assisted recombination in a variety of organic solar cell systems.[1,12,13]

TABLE I. Series resistance and shunt resistance extracted from the dark current of solar cell devices D1, D2, D3, and D4 as well as the slope of the V_{oc} light intensity dependence of each device.

Device	R_s (Ω cm^2)	R_{sh} (Ω cm^2)	V_{oc} slope (kT/q)
D1	1.2	2.2×10^3	2.0
D2	2.4	5.7×10^3	1.4
D3	2.2	2.7×10^4	1.2
D4	3.6	1.2×10^6	1.0

Equation (3) was derived by considering that for an ideal device with negligible leakage current, generation is cancelled about by recombination at open circuit. However, in the case of a nonideal device with low shunt resistance, the shunt current also contributes to cancelling out the photogenerated current such that at open circuit

$$J_{ph} = J_{rec} + J_{sh}, \tag{4}$$

where J_{rec} is the recombination current. Following a previously described model for a metal-insulator-metal diode with only bimolecular recombination, the recombination current can be expressed as

$$J_{rec} = qL(1-P)\gamma np, \tag{5}$$

where L is the active layer thickness, n is the density of free electrons, and p is the density of free holes.[2] Likewise, when the photocurrent is measured under a strong reverse bias such that the photocurrent is saturated, the photocurrent can be expressed in terms of the generation rate G, as

$$J_{ph} = qLPG. \tag{6}$$

It should be noted that P may be voltage dependent as some organic solar cell systems have been shown to exhibit voltage dependent photogeneration.[14,15] From Eq. (2), it is evident that at open circuit $J_{sh} = \frac{V_{oc}}{R_{sh}}$ and thus it follows from Eqs. (4)–(6) that

$$PG = (1-P)\gamma np + \frac{V_{oc}}{qLR_{sh}}. \tag{7}$$

At open circuit, the quasi-Fermi levels across the device are approximately constant and their energy difference equal to the applied voltage, therefore

$$np = N_c^2 \exp\left(\frac{qV_{oc} - E_{gap}}{kT}\right). \tag{8}$$

Combining Eqs. (7) and (8), one can obtain an expression for V_{oc} similar to that of Eq. (3) but now also considering the effect of shunt current, such that

$$V_{oc} = \frac{E_{gap}}{q} - \frac{kT}{q}\ln\left(\frac{(1-P)\gamma N_c^2}{PG - V_{oc}/qLR_{sh}}\right). \tag{9}$$

In the ideal case, R_{sh} is large such that even at low light intensities J_{sh} is negligible and Eq. (9) simplifies to the expression in Eq. (3). In the nonideal case, R_{sh} may be small such that the shunt current is non-negligible and thus the R_{sh} term in Eq. (9) cannot be neglected. The effect of low R_{sh} will be to increase the V_{oc} light intensity slope such that even a system with purely bimolecular recombination may appear to have a slope $>kT/q$. This is demonstrated in Figure 2(a), where the V_{oc} is plotted versus the incident light intensity for the four devices presented in Figure 1. The symbols represent data points and the lines are fits for a natural logarithmic dependence of V_{oc} on light intensity. While the low leakage

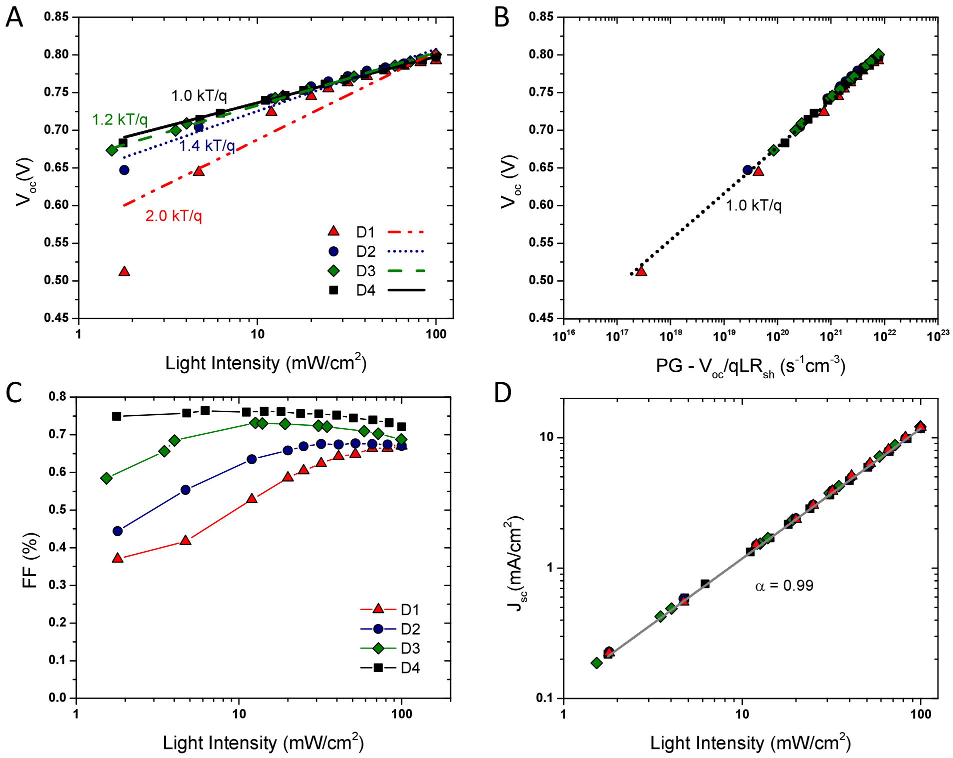

FIG. 2. The light intensity dependence of the V_{oc} (a), FF (c), and J_{sc} (d) of devices D1, D2, D3, and D4. The solid and dashed lines in (a) are fits to the data using Eq. (3). (b) shows the V_{oc} of devices D1, D2, D3, and D4 plotted versus $PG - V_{oc}/qLR_{sh}$ where the dashed line has a slope of kT/q.

device, D4, exhibits a slope of 1.0 kT/q the slope steadily increases with increasing leakage current with D3, D2, and D1 exhibiting slopes of 1.2, 1.4, and 2.0 kT/q, respectively. In contrast, when the effect of leakage current is accounted for by plotting V_{oc} versus $PG - V_{oc}/qLR_{sh}$, each device exhibits approximately the same slope of 1.0 kT/q as predicted by Eq. (9) (Figure 2(b)) for a system dominated by bimolecular recombination.

The effect of leakage current is also evident in the dependence of the device FF on incident light intensity as shown in Figure 2(c). In the case of device D4, the FF appears steady at ~0.75 from ca. 2 mW/cm^2 to 20 mW/cm^2 after which it decreases slightly with increasing light intensity to ~.0.70 at 100 mW/cm^2. The decrease of FF with increasing light intensity has been reported to have two origins: series resistance and bimolecular recombination.[1,5,12,16] The effects of bimolecular recombination have gained considerable attention in particular, as bimolecular recombination is known to be a significant loss mechanism in most organic solar cells.[1,17,18] Therefore, the FF dependence on light intensity has been used sporadically to infer differences in recombination dynamics across various organic solar cell systems which are often in turn then attributed to observed morphological changes. However, leakage currents can also influence the FF dependence on light intensity as illustrated by devices D1, D2, and D3 wherein the FF is observed to decrease at lower light intensities. The light intensity below which the FF decreases is determined by the shunt resistance. For instance, the FF of the lowest shunt resistance devices, D1, continuously decreases starting at intensities below 100 mW/cm^2 where as the FF of D3 increases from 100 mW/cm^2 to ~10 mW/cm^2 and then decreases at lower light intensities. As with the V_{oc}, the effect of leakage current on FF becomes increasingly prominent at lower intensities because the leakage current is independent of light intensity where as the magnitude of the photocurrent steadily decreases with decreasing light intensity.

It is worth noting that unlike the V_{oc} and FF, the dependence of the short circuit current (J_{sc}) is *not* strongly influenced by leakage currents. This can be understood from Eq. (1), which for the case of V = 0 and $JR_s \approx 0$, reduces to $J(I, 0) = J_{ph}(I, 0)$. As shown in Figure 2(d), the J_{sc} for all four devices here exhibits an identical dependence on light intensity which can be described with a power law fit to $J_{sc} \propto I^{\alpha}$ with $\boldsymbol{\alpha} = 0.99$ which is a typical value for organic solar cells such as *p*-DTS(FBTTh$_2$)$_2$:PC$_{71}$BM that have balanced carrier mobilities and only modest bimolecular recombination losses at short circuit.[1,15,19]

Comparing Figure 2(c) with the light dependence of the solar cell power conversion efficiency (PCE) shown in Figure 3(a), it is evident that the PCE light dependence is largely set by the trend in FF with the V_{oc} dependence also playing a role. At intensities close to one sun, the PCE of each device is similar as are the FFs however the difference is pronounced at lower light intensities where the leakage current competes favorably with the photocurrent. The origin of this is illustrated in Figure 3 which features the current voltage characteristics of D1 (Figure 3(b)) and D4 (Figure 3(c)) measured in the dark (black line) and at various illumination intensities ranging from 2–100 mW/cm^2 (color lines). As mentioned previously from 0 V to ~0.7 V, in both devices the dark current is dominated by the leakage current while the light current is dominated by the difference between photocurrent and leakage current. For device D1, the light current measured at 100 mw/cm^2 is over one order of magnitude higher than the leakage current across the operating regime and thus the FF and V_{oc} are not significantly decreased by the leakage current. However at lower light intensities, where the photo current is within one order of magnitude of the leakage current, the effect on the light current is

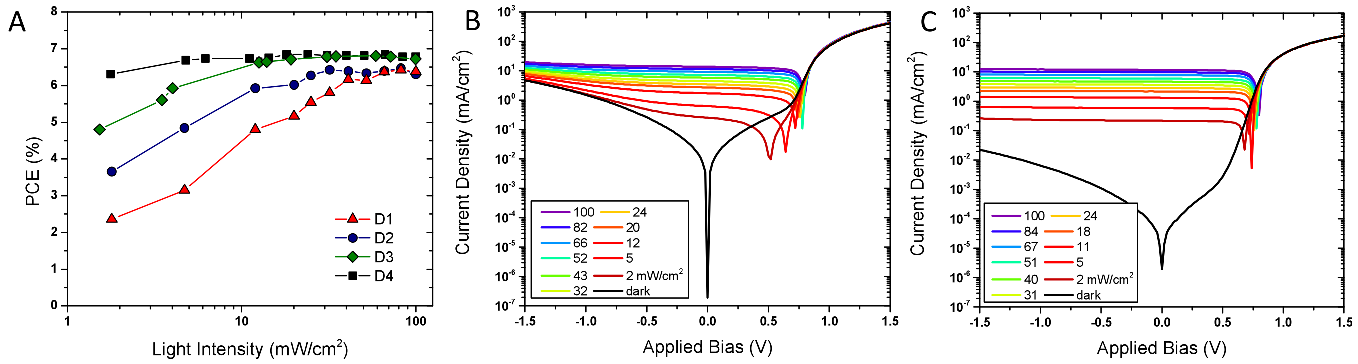

FIG. 3. (a) The light intensity dependence of the PCE of devices D1, D2, D3, and D4. Current density as a function of applied bias for devices D1 (b) and D4 (c) measured in the dark and under illumination up to 100 mW/cm^2.

pronounced resulting in steep drops in FF and V_{oc}. In contrast, as shown in Figure 3(c), for device D4 with low leakage current, even at relatively low light intensities, the light current is orders of magnitude greater than the dark (leakage) current and thus the FF and V_{oc} can be measured independent of the leakage current across a wide range of light intensities.

In conclusion, the effect of leakage current and shunt resistance on the light intensity dependence of a model organic solar cell system has been demonstrated. In the case of solar cell devices with low shunt resistance ($<10^6\,\Omega\,\mathrm{cm}^2$), current-voltage measurements conducted at incident light intensities less than one sun may be significantly skewed by parasitic leakage current. In such devices, the effect of the leakage current will be to decrease the device FF and the V_{oc} increasingly more as the light intensity is decreased. The slope of the logarithmic dependence of the V_{oc} on light intensity is particularly sensitive to the effect of leakage currents with slopes ranging from 1 kT/q to 2 kT/q for the same material system depending on the shunt resistance. This effect may explain some discrepancies about the light intensity dependence of various OPV systems reported in the literature. Thus, we assert that care should be taken to ensure that parasitic leakage currents are minimized when measuring the light intensity dependence of organic solar cells.

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