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FULL PAPER

Measuring the External Quantum Efficiency of Two-Terminal Polymer Tandem Solar Cells

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Tandem configurations, in which two cells are stacked and connected in series, offer a viable approach to further increase the power conversion efficiency (*PCE*) of organic solar cells. To enable the future rational design of new materials it is important to accurately assess the contributions of individual subcells. Such accurate measurement of the external quantum efficiency (*EQE*) of the subcells of two-terminal organic or polymer tandem solar cells poses specific challenges, caused by two characteristics of these cells, *i.e.* a sub-linear light intensity dependence of the current and a field-assisted charge collection. These properties necessitate that *EQE* experiments are carried out under representative illumination conditions and electrical bias to maintain short-circuit conditions for the addressed subcell. We describe a method to determine the magnitudes of the bias illumination and bias voltage during *EQE* measurements, based on the behavior of single junction cells and optical modeling. The short-circuit current densities of the subcells obtained by convolution of the *EQE* with the AM1.5G solar spectrum are consistent with those obtained from optical modeling and correctly predict the current density–voltage characteristics of the tandem cell under AM1.5G conditions.

1. Introduction

Polymer solar cells can possibly contribute to the future global demand for renewable and sustainable energy.[1] Although polymer solar cells based on a single active layer now reach power conversion efficiencies of over 7%,[2] ultimately a tandem cell configuration may be required to reach commercially viable efficiencies. In a tandem cell two photoactive layers, designed to convert different spectral regions of the sunlight are stacked on top of each other separated via a recombination layer that serves to electrically connect the two subcells. The stack is sandwiched between top and bottom electrodes (**Figure 1a**). Tandem solar cells can reduce the transmission and thermalization losses that are encountered in single junction cells, where photons with energies exceeding the optical band gap energy lose the surplus energy via non-radiative relaxation and photons with energies smaller than the band gap are not absorbed. Hence, by combining wide and small band gap subcells, tandem cells retain more of the absorbed photon energy and offer increased power conversion efficiency. In recent years, several successful strategies have been developed for solution processed polymer tandem solar cells and efficiencies up to 6.5% have been reported.[3–14]

For a detailed understanding of the operation and optimization of polymer tandem solar cells, the rational design of new and improved materials, and the accurate determination of the power conversion efficiency it is important to be able to assess the performance of the individual subcells in a tandem configuration.[11,15] This can be accomplished by determining the monochromatic external quantum efficiency (*EQE*) and internal quantum efficiency (*IQE*), which are defined as the number of electrons collected at short-circuit per incident and per absorbed photon at that wavelength, respectively. Using the *EQE* of the subcells, it is possible to assess which of the two is limiting the performance and assess how further rise in efficiency might be possible by improved materials design. The *EQE* is also necessary to establish accurate power conversion efficiencies,[16,17] because it determines the spectral mismatch factor that is used to correct for differences between white light from a solar simulator and the tabulated AM1.5G spectrum.[18]

EQE measurements of single junction solar cells are preferably carried out by measuring the effect of a small amount of monochromatic light under short-circuit conditions with appropriate bias light to bring the cell close to the standard global air mass 1.5 (AM1.5G) sunlight operation conditions. Measuring the *EQE* for a tandem cell is significantly more challenging, especially for the technologically attractive two-terminal configuration where the intermediate contact is inaccessible and subcells cannot be individually addressed. The tandem cell is only able to pass current when both the wide *and* the small band gap subcells are simultaneously excited with light and care must be taken that the response to the additional monochromatic light indeed represents the response of the subcell under study and not that of the other one. Hence, the subcell under study must be current-limiting over the entire spectral range.[19] To address this difficulty, detailed protocols for *EQE* measurement of two-terminal tandem and triple junction solar cells have been developed.[20–25] While these methods are generally applicable to inorganic solar cells, specific characteristics, such as a reduced fill factor and a sub-linear light intensity dependence, limit their

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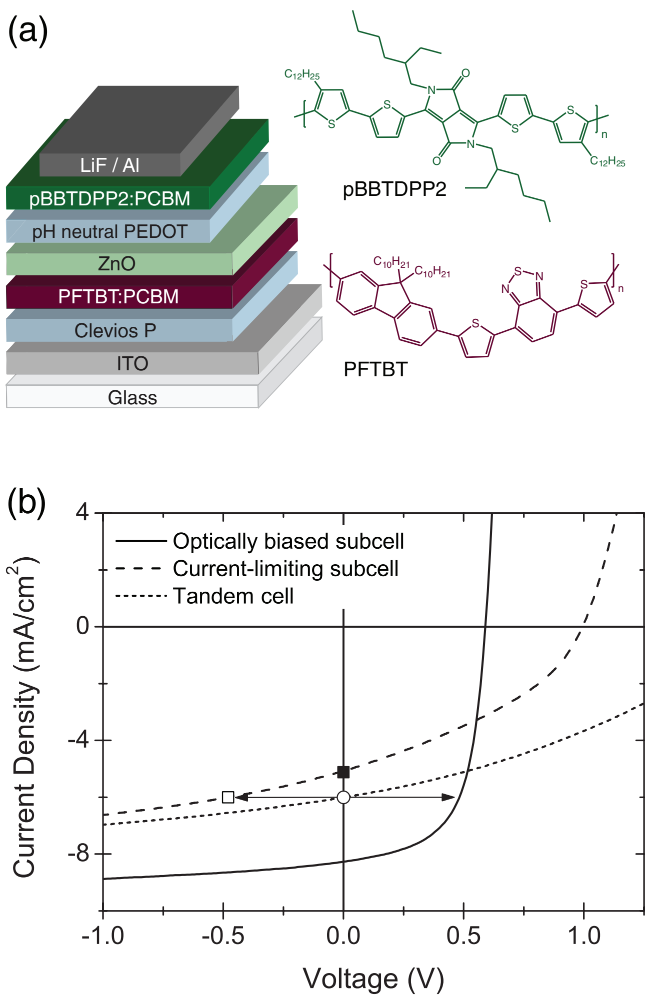

Figure 1. (a) Device layout of the studied polymer tandem solar cell and the molecular structure of the applied polymers. (b) Influence of the bias voltage of the optically biased subcell on the current-limiting subcell. ■ is the target value while □ is the obtained value when measuring the tandem cell under short-circuit conditions.

use for polymer tandem solar cells. With the increasing interest for polymer tandem solar cells,[3–14,26,27] there is a strong need for a customized protocol to measure the *EQE* of these cells correctly.

It is evident that to determine the *EQE* of individual subcells, bias illumination is required. The spectrum and intensity of bias illumination must be chosen such that the bias light is predominantly absorbed by one of the two subcells and results in charge generation in that subcell that largely exceeds the charge generation in the other subcell. As a consequence, the latter subcell will be current-limiting for the tandem cell over the entire wavelength range and the *EQE* of the tandem cell represents the *EQE* of the current-limiting subcell. Yet, by optically biasing the tandem cell, the much more strongly absorbing subcell (denoted as "optically biased subcell") will be operating close to its open-circuit voltage (V_{oc}) because it will produce many more charges than can be transported through the other, "current-limiting, subcell". Because the total cell is measured at short circuit ($V = 0$) and the optically biased cell is operating at a forward bias $V = +V'$, close to V_{oc} the current-limiting subcell will be operating at $V = -V'$, *i.e.* under reverse bias. For organic solar cells, where charge collection is often field dependent, the reverse bias creates additional photocurrent and, hence, an overestimation of the *EQE*. The effect is illustrated in Figure 1 for a 4.9% polymer tandem solar cell comprising wide and small band gap polymers and a ZnO|PEDOT recombination layer.[11] Figure 1b shows the current density – voltage (*J–V*) characteristics of the subcells in the tandem and reveals that bias illumination of one subcell leads to a significant increase in photocurrent for the non-biased, current limiting cell (*cf.* ■ and □ in Figure 1b). In the example shown, the error induced is about 16%, but it can be larger or smaller depending on the magnitude of the slope of *J-V* curve around $V = 0$. To correct for this effect and correctly measure the *EQE* of the subcell under short-circuit conditions, the reverse bias created has to be compensated by applying a forward electrical bias on the tandem cell. The crucial question is how much light bias and electrical bias are required.

Here, we present a method to accurately determine the magnitude of the required optical and electrical bias to determine the *EQE* of the subcells in a two-terminal polymer tandem solar cell. In this method, representative single junction dummy – or replica – cells, identical to the tandem subcell, are used in combination with optical modeling to mimic the behavior of the tandem subcell. Using these single junction dummy cells one can successfully correct for the complications caused by the significant difference in light absorption by the two subcells in the tandem during the measurements and obtain a reliable *EQE* that describes the operation of the cell under operation at solar light intensity. We verify the validity of the method in two ways. First, by comparing the measured *EQE* with the *EQE* obtained from the product of the *IQE* of single junction dummy cells and the calculated fraction of absorbed light in the tandem. Second, by calculating the short-circuit current density (J_{sc}) via convolution of the *EQE* with the AM1.5G solar spectrum and comparing the results to those obtained with a class-A solar simulator.

2. Results and Discussion

2.1. Experimental Set-Up for the *EQE* Measurement

A schematic diagram of the *EQE* set-up is given in **Figure 2a**. It consists of a tungsten-halogen white light that is modulated with a mechanical chopper and then dispersed by monochromator that includes a set of ordering filters to obtain modulated monochromatic probe light. Further two cw semiconductor lasers are used for bias illumination with wavelengths tuned to excite the photoactive layers in the different subcells. Their intensity can be varied with circular variable neutral density filters. The three light beams are combined at tandem cell under study using lenses, mirrors, and a beam splitter. A lock-in-amplifier referenced to the mechanical chopper is used as measuring unit. Optical components like lenses and mirrors to focus the incident light on the sample are omitted from

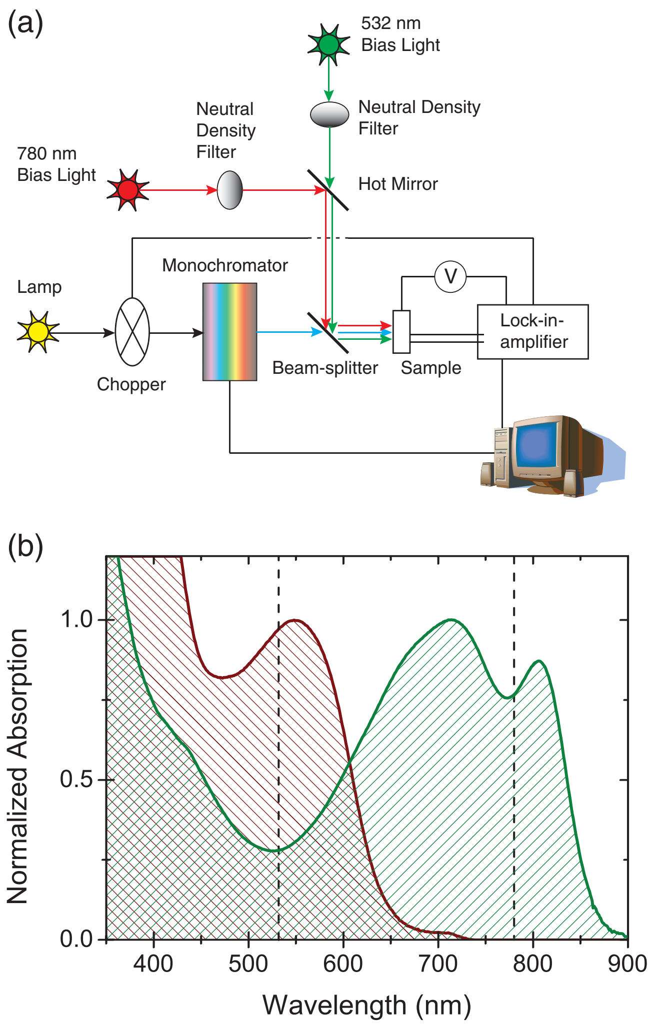

Figure 2. (a) Set-up for the *EQE* measurement of tandem solar cells. (b) Normalized absorption spectra of both individual active layers (PFTBT:PCBM in wine and pBBTDPP2:PCBM in green). The dotted lines represent the wavelengths of the bias illumination.

Figure 2a for clarity. Further details are included in the Experimental section.

The device whose *EQE* measurement is presented here, has been described previously and is based on poly[2,7-(9,9-didecylfluorene)-alt-5,5-(4′,7′-di-2-thienyl-2′,1′,3′-benzothiadiazole)] (PFTBT) as wide band gap polymer and poly[3,6-bis(4′-dodecyl-[2,2′]bithiophenyl-5-yl)-2,5-bis(2-ethylhexyl)-2,5-dihydropyrrolo[3,4-]pyrrole-1,4-dione] (pBBTDPP2) as small band gap polymer, both in combination with [6,6]-phenyl-C_{61}-butyric acid methyl ester ([60]PCBM or PCBM in short) (Figure 1a).[11] The tandem structure with a 180 nm thick PFTBT:PCBM front cell and a 125 nm thick pBBTDPP2:PCBM back cell resulted in an overall efficiency of 4.9%.

The normalized absorption spectra of individual films of the active layers of the subcells are displayed in Figure 2b and show the wavelengths used for the bias illumination. The PFTBT:PCBM layer is insensitive to light above 750 nm while pBBTDPP2:PCBM still absorbs up to 900 nm. As a result, bias illumination with a 780 nm laser is well suited to generate excess charges in the small band gap back cell and measure the current-limiting wide band gap front cell in the tandem configuration. On the other hand, the low absorption of pBBTDPP2:PCBM in the 500 to 550 nm region enables selective addressing of the small band gap back cell in the *EQE* measurement of the tandem cell with bias illumination from a 532 nm laser.

2.2. Challenges in *EQE* Measurements of Polymer Tandem Solar Cells

In contrast to inorganic solar cells, the *EQE* of polymer solar cells under reduced light intensity is often overestimated compared to the response under standard operating conditions due to the sub-linear light intensity dependence of polymer solar cells.[28] By using monochromatized white light, the light intensity on the (sub)cells is orders of magnitude lower than the AM1.5G light intensity (1000 W m^{-2}) (**Figure 3a**). Representative illumination conditions can be obtained by sufficient additional continuous flood light. Ideally this flood light would be simulated AM1.5G solar light to simulate operating conditions in both subcells, but this is in our set-up not practically feasible. We note that apart from flood light, determining the *EQE* of tandem cells also requires specific bias illumination to optically bias one subcell such that the other subcell, which is

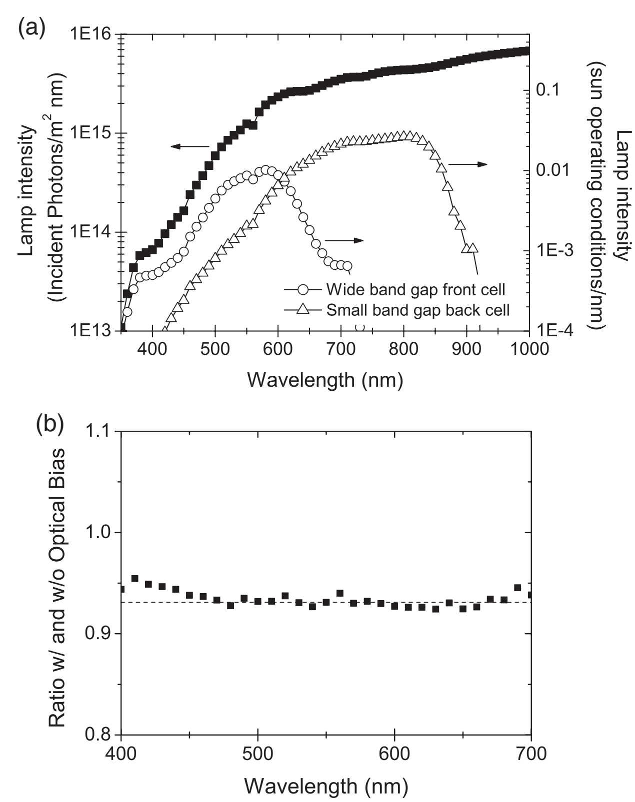

Figure 3. (a) Intensity of the modulated monochromatic light expressed in incident photons m^{-2} (■) and relative to one-sun operating conditions for the wide band gap front cell and small band gap back cell (open symbols). (b) Ratio between the *EQE* determined with and without optical bias on a wide band gap single junction dummy cell.

under investigation, is current-limiting over the entire wavelength range.

The approach to solve this problem differs for the two subcells. For the small band gap back cell, sufficient flood light can be provided by the 532 nm bias illumination. This can explained by considering that while the 532 nm light is primarily absorbed by the wide band gap front cell (creating the optical bias), also the small band gap back cell absorbs a substantial amount at this wavelength and the intensity of the 532 nm can be tuned to generate one-sun operating conditions in the small band gap back cell. As a consequence, the response of the monochromatic light is measured under the appropriate conditions. On the other hand, the wide band gap front cell is insensitive to the 780 nm light, used to bias the small band gap back cell. To correct for this, a mathematical correction of the measured *EQE* is applied to correct for the sub-linear light intensity dependence of the current. This correction factor is determined as the average ratio between the *EQE* measurements with and without one-sun intensity flood light of a wide band gap single junction dummy cell (Figure 3b).

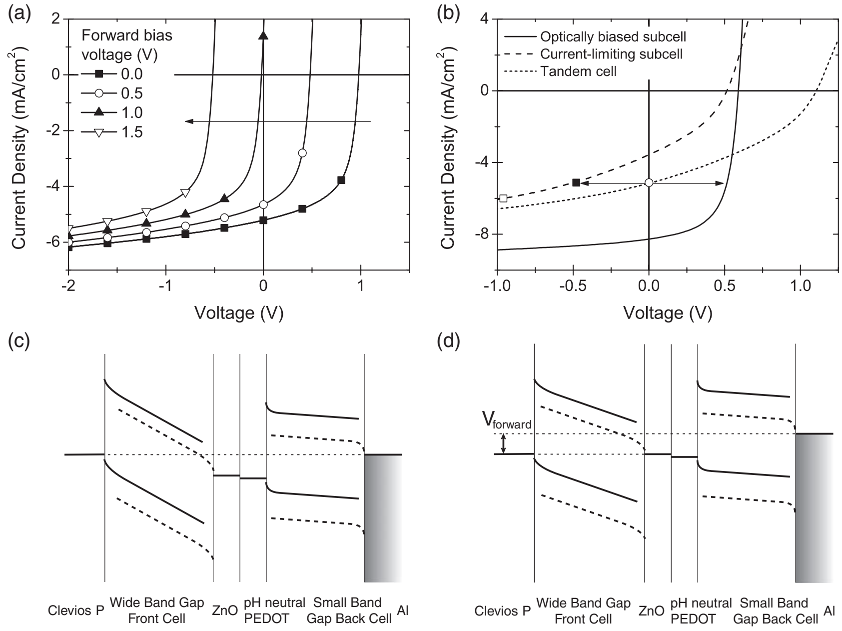

Figure 4. (a) The effect of a forward bias voltage on a single junction solar cell and (b) on the subcells of a tandem solar cell. This enables the measurement of the current-limiting subcell at short-circuit conditions in contrast to Figure 1b. (c) Energy band diagram of the tandem cell under illumination of 780 nm light at short-circuit condition of the tandem cell and (d) with an applied forward bias voltage to keep the current-limiting wide band gap front cell at short-circuit.

A second problem to address is the reverse electrical bias $V = -V'$ on the current-limiting subcell that is created by the bias light absorbed by the optically biased subcell which operated at $V = +V'$ (close to V_{oc}) under conditions where the total tandem cell is at short-circuit $V = V' - V' = 0$. This phenomenon is more relevant in polymer tandem solar cells than in inorganic tandem devices, where the influence is stated to be marginal.[22,23] Polymer solar cells often exhibit a sloped *J–V* curve under reverse bias (Figure 1b) and can display field-assisted charge collection. In a phenomenological device model this characteristic is represented by a low shunt resistance. As shown in Figure 1b this results in a higher current density in the current-limiting subcell and to an overestimation of the *EQE* in measurements, when the *tandem* cell is operating at short-circuit conditions.[11]

To compensate for this overestimation a forward electrical bias is required. Applying a forward bias voltage on a solar cell moves the *J–V* curve of the entire cell towards more negative voltage (**Figure 4a**). In a tandem cell, the optically biased subcell is continuously operating close to V_{oc} regardless the voltage over the tandem cell. Therefore, applying a forward bias voltage on the tandem cell moves the *J–V* curve of the tandem and the current-limiting subcell to more negative voltages enabling the measurement of the current-limiting subcell operating at short-circuit conditions (Figure 4b). This is illustrated by an energy band diagram of the tandem cell under 780 nm bias illumination in Figure 4c and 4d.

2.3. Determination of the Magnitude of the Optical Bias

For *EQE* measurements on the small band gap back cell, the appropriate, AM1.5G equivalent intensity for 532 nm laser flood light can be determined from the absorption profile of the tandem subcells obtained by calculating the number of absorbed photons using optical modeling as described before.[11] The optical modeling requires knowing the refractive index and extinction coefficient of all layers as function of wavelength as well as the thickness of each individual layer. For the example described here we find that at this intensity and wavelength (532 nm), the wide band gap front cell absorbs ca. three times more light, which ensures that the small band gap back cell remains current-limiting.

On the other hand, for the *EQE* measurement of the wide band gap front cell, the 780 nm laser light can only be used as bias light for the small band gap back cell, but cannot act as flood light to have the wide band gap front cell operating under one-sun conditions. Hence, the intensity of this 780 nm bias illumination is arbitrary as long as the charge generation in the small band gap back cell exceeds the charge generation in the wide band gap front cell at any wavelength of the modulated probe light to make it not current-limiting. The intensity of the 780 nm bias light was tuned to AM1.5G equivalent intensity for the small band gap dummy cell, comparable to the 532 nm bias light.

2.4. Determination of the Magnitude of the Electrical Bias

In section 2.2 we explained that under bias illumination and short-circuit conditions, the current-limiting subcell is actually operating under reverse bias because the optically biased subcell operates close to V_{oc}. A forward bias voltage on the tandem cell is required to compensate for this effect and to avoid the overestimation of the *EQE*, which can be as large as 16% for the materials studied here (Figure 1b), In a first-order approximation, the optically biased subcell is operating close to V_{oc} (**Figure 5**). However, as shown in Figure 5b this voltage can deviate substantially from V_{oc}. The precise magnitude of the electrical bias is hard to define in a two-terminal solar cell where the intermediate contact is inaccessible. Nevertheless, the correct electrical bias can be determined by making use of *J–V* characteristics of single junction dummy cells under illumination conditions that are representative for the subcells in the tandem as obtained by calculating the number of absorbed photons of the tandem cell via optical modeling.

During the *EQE* measurement, the tandem cell also experiences the modulated probe light on top of the bias illumination generating additional current in the subcells. The intensity of the probe light, however, varies for different wavelengths and causes a variable electrical bias for different wavelengths. For the ease of the experiment we opted for an average value of the electrical bias at wavelengths of minimal and maximal additional current generation of the modulated probe light. These wavelengths are 350 nm and 550 nm, respectively for the wide band gap front cell and 350 nm and 800 nm for the small band gap back cell.

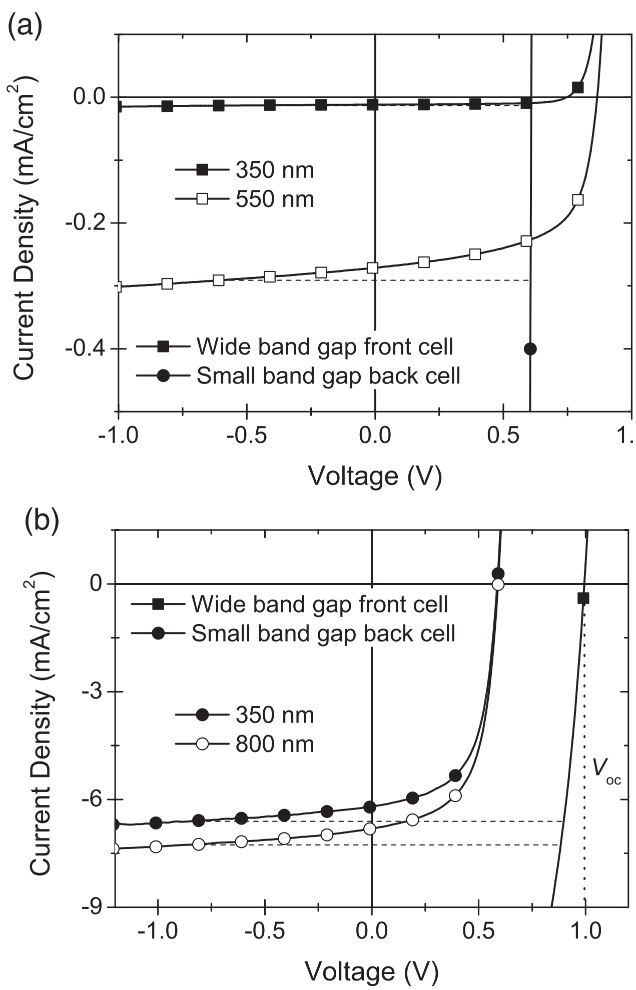

Figure 5. *J–V* curves of wide and small band gap dummy cells under illumination conditions representative for the subcells during the *EQE* measurements: (a) represents the situation during the *EQE* measurement of the wide band gap front cell, (b) reflects the situation during the *EQE* measurement of the small band gap back cell. The required electrical bias is indicated by the dotted lines.

To show how determining the magnitude of the electrical bias is accomplished, we have plotted in Figure 5 the *J–V* curves of the dummy cells under bias illumination plus probe light (350 and 550 nm for Figure 5a; 350 and 800 nm for Figure 5b). The *J–V* curve of the optically biased subcell hardly changes with variation of the probe light wavelength and therefore only one curve is shown. The forward bias voltage, to be applied on the tandem cell to ensure short-circuit conditions for the measured subcell, is the absolute value of the voltage of the data points of the *J–V* curves with equal current densities and voltages opposite in sign indicated by the horizontal dashed lines in Figure 5. For example, in Figure 5a for 550 nm probe light the horizontal dashed line connects the point (J, $-V'$) on the curve of the wide band gap cell (–□–) with the point (J, $+V'$) on the curve of small band gap cell (–•–). When the tandem cell operates at (J,0) the current limiting subcell will be at (J, $-V'$) and the optically biased subcell at (J, $+V'$). Hence a bias of $V = V'$ is required to bring the current limiting subcell to zero bias. From Figure 5b we infer that the average electrical bias needed in the *EQE* measurements was 0.60 and 0.90 V for the wide band gap front cell and the small band gap back cell, respectively. Note that the electrical bias to be applied on the small band gap back cell deviates substantially from the V_{oc} of the PFTBT:PCBM dummy cell (0.98 V) (see Figure 5b).

2.5. Combination of Light and Electrical Bias

The *EQE* of the subcells of the polymer tandem solar cell changes significantly under influence of applied optical and electrical bias (**Figure 6**). Consequently, also the J_{sc}, estimated by convoluting the *EQE* with the AM1.5G solar spectrum,[16] changes considerably (**Table 1**). Without optical and electrical bias, the J_{sc} of the wide band gap front cell is overestimated by 16%. For the small band gap back cell the deviation is only related to the electrical bias, but this alone accounts for an error of 16%. This clearly demonstrates that carefully determining the magnitude of the optical and electrical bias is essential for accurate *EQE* measurements. The convoluted J_{sc} of the wide band gap front cell is 5.8 mA cm^{-2} with a maximum *EQE* of 0.44 at 540 nm and of the small band gap back cell 7.0 mA cm^{-2} with a maximum *EQE* of 0.37 at 700 nm. The fact that the *EQE* of *both* cells would be overestimated by ~16% without an electrical bias, is a coincidence. In other cases the error made can be larger or smaller depending on the slope of *J–V* around $V = 0$.

2.6. Verification of the Method

If we assume that the internal quantum efficiency (*IQE*) of the subcells is identical to the (known) *IQE* of the representative single junction dummy cells,[11] the *EQE* of the tandem subcells can also be calculated by calculating the number of absorbed photons in each subcell of the tandem by optical modeling and using the *IQE* data of the single junction dummy cells. The calculated

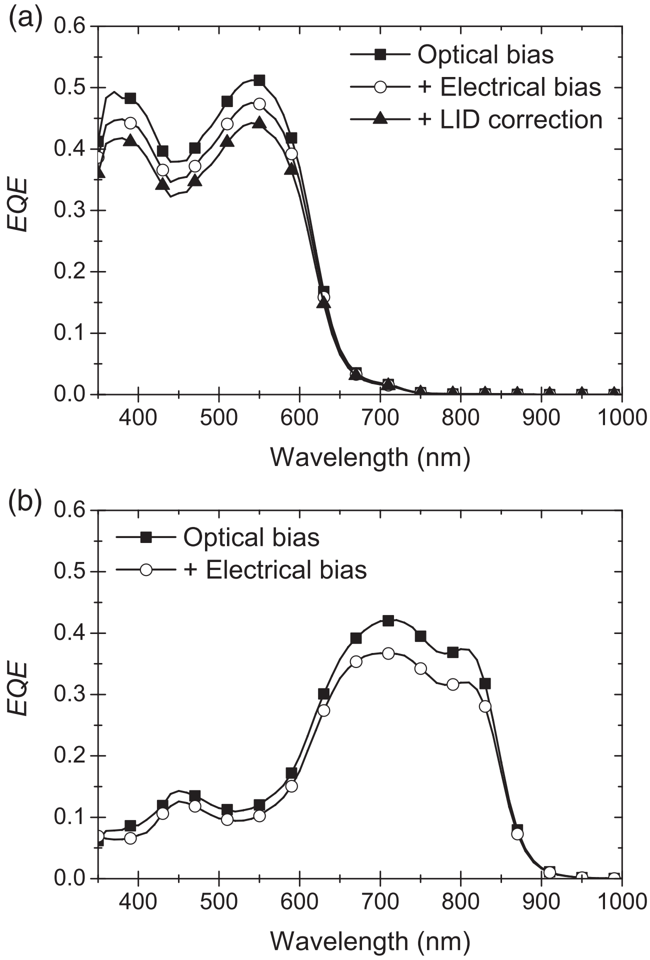

Figure 6. The EQE of the (a) wide band gap front cell and (b) small band gap back cell after applying relevant optical and electrical biases. For the wide band gap front cell an extra light intensity dependence (LID) correction is required.

spectral light absorption of both subcells in the tandem cell multiplied by the IQE of the subcells for these active layer thicknesses (0.69 and 0.60 for the wide band gap front cell and small band gap back cell respectively) provide the EQE values:

$$EQE(\lambda) = \text{absorption}(\lambda, \%) \cdot IQE \quad (1)$$

Figure 7 shows that the magnitude and spectral shape of the measured EQE is similar to the calculated EQE for both

Table 1. The short-circuit current densities of the subcells in the tandem cell obtained from EQE measurements after convolution with the AM1.5G solar spectrum.

Subcell	Corrections	J_{sc} (mA cm^{-2})
Wide band gap front cell	Optical bias	6.7
	+ electrical bias	6.2
	+ LID correction	5.8
Small band gap back cell	Optical bias	8.1
	+ electrical bias	7.0

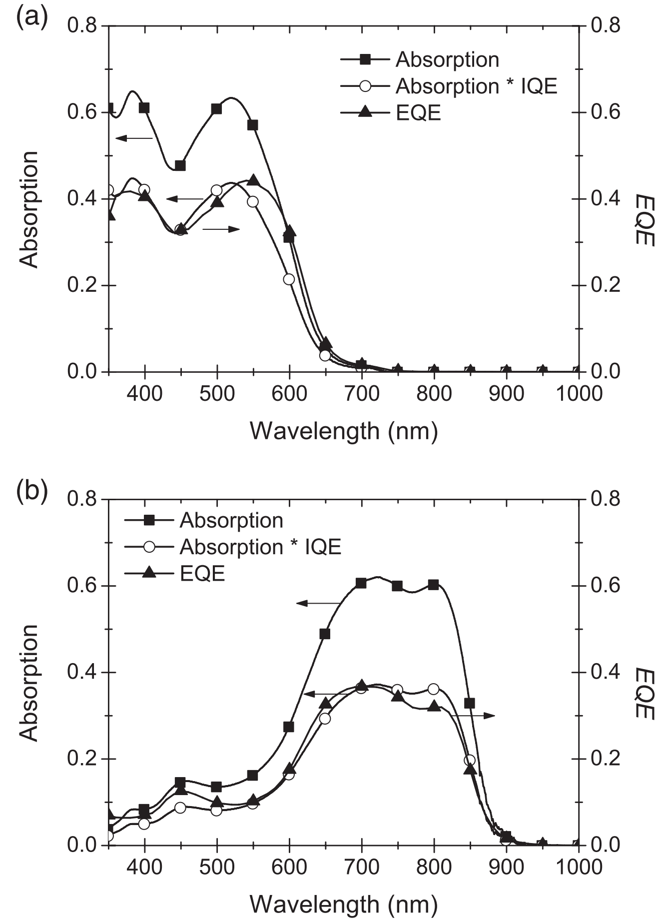

Figure 7. Comparison between the calculated and measured EQE for the (a) wide band gap front cell and (b) small band gap back cell of the tandem cell.

subcells. This demonstrates the validity of the assumption that the IQE of the subcells and single junction dummy cells are comparable. Moreover, the good match between measured and calculated EQE is a strong indication that the EQE measuring protocol provides accurate results. The shape of the two curves is comparable demonstrating that the optical modeling closely approximates the optical processes in the tandem stack.

The EQE, convoluted with the AM1.5G solar spectrum is a measure for the short-circuit current density generating capacity of the two subcells under solar irradiation. Using the method that was introduced previously,[11] these J_{sc} generating capacities can be combined with the V_{oc}, FF and the shape of an equally thick single junction cell, to obtain the J–V curves of both subcells (**Figure 8**). Kirchhoff's law (*i.e.* adding the voltages of the subcells for the same current density) then allows construction of the J–V curve of the complete tandem cell. This constructed tandem J–V curve coincides with the actual J–V curve (Figure 8), measured under a class-A solar simulator and corrected for the spectral mismatch. This demonstrates that the magnitude of the J_{sc} generation capacity and thus the EQE has accurately been determined with the present measurement protocol.

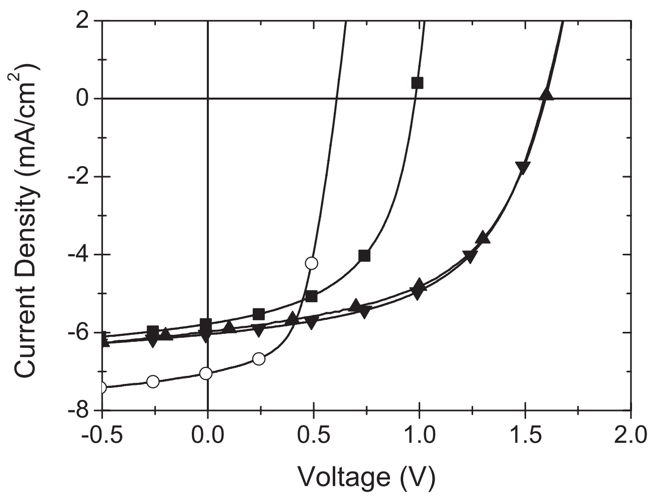

Figure 8. Constructed *J–V* curves of the wide band gap front cell (■) and small band gap back cell (◦) in the tandem cell with J_{sc} based on the *EQE* measurements. The constructed *J–V* curve of the tandem cell (▼) by addition of the *J–V* curves of the two subcells is compared to the *J–V* curve measured under simulated solar light (▲).

3. Conclusions

We described a method to measure the external quantum efficiency of two-terminal polymer tandem solar cells using an optical and electrical bias. The sub-linear light intensity dependence and field-assisted current generation complicate the measurements in organic tandem solar cells compared to inorganic tandem solar cells and require flood light to operate the subcell at one-sun equivalent light intensity conditions and a forward bias voltage to create short-circuit conditions for the addressed subcell. The method presented here relies on the behavior of single junction dummy cells, identical to the subcells. One-sun equivalent operating conditions in the subcell are generated for the *EQE* measurement of the small band gap back cell by tuning the intensity of the bias light, while the *EQE* measurement of the wide band gap front cell requires a mathematical correction. The magnitude of the electrical bias was determined by comparing *J–V* curves of the dummy cells under representative illumination conditions of the subcells in the tandem cell. For the specific materials used, the *EQE* changed by 16% under influence of applied optical and electrical bias demonstrating the importance of accurate determination of the magnitude of the optical and electrical bias. This resulted in short-circuit current density generating capacities of 5.8 mA cm^{-2} and 7.0 mA cm^{-2} for the wide band gap front cell and small band gap back cell respectively from convolution of the *EQE* with the AM1.5G solar spectrum. To verify the method, the measured and calculated *EQE* and the measured and constructed *J–V* curves of the tandem cell were compared. The good agreement found shows that the *EQE* measurement protocol results in a reliable estimate of the *EQE* and the method can be used for future characterization of polymer tandem solar cells.

4. Experimental Section

Devices Fabrication and Characterization: The fabrication of the tandem devices has been described in detail before.[17] The *EQE* measurements were done in a home-built set-up. The modulated monochromatic (Oriel Cornerstone 130 1/8 m, Newport) probe light (Halotone halogen lamp, Philips) was mechanically chopped at a frequency of 165 Hz with an optical chopper (SR 540, Stanford Research). The bias illumination was provided by a 532 nm (B&W, Tek Inc., 30 mW) and 780 nm (B&W, Tek Inc., 21 mW) solid state laser. The sample was stored in a nitrogen filled container during the measurements and illuminated through an aperture of 2 mm which is slightly smaller than the device area to avoid errors. The measurements were executed with a lock-in-amplifier (SR 830, Stanford Research) over a load of 50 Ω. All data were recorded by a Labview program on a computer. The electrical bias over the tandem cell was provided by the lock-in-amplifier.

Optical Modeling: Calculations of the optical electric field were performed with the Essential Macleod software package (Thin Film Center, Inc., Tucson, USA). The optical constants of glass and ITO were provided by Thin Film Center. The other optical constants were obtained from variable angle ellipsometry measurements or literature.[29]

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