A General Guideline for Vertically Resolved Imaging of Manufacturing Defects in Organic Tandem Solar Cells

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Organic tandem solar cells recently made great improvements with power conversion efficiencies (PCEs) over 15%, making them attractive for further large-scale production and industrial applications. However, compared to their single-junction counterparts, the complicated device architectures of organic tandem solar cells strongly restrict their processing and upscaling to larger scales. Therefore, fast and reliable quality control measures are crucial for developing organic tandem photovoltaic technologies towards commercialization. Some of the most widely used means for quality control are luminescence imaging and lock-in thermography respectively. While effective techniques, they are limited in some respects. For example, determining the lateral position of a defect is easily possible, while the exact resolution in which layer of a thin film stack a defect is located, is challenging. This is particularly the case for tandem cells with complicated multi-layer cell architectures. This approach to overcome this challenge is the introduction of well-defined artificial defects into certain layers of an organic tandem cell stack and subsequently performing imaging analysis of the defected cells with several complementary methods. The unique response from cells with artificial defects using different imaging techniques and excitation sources can then be transferred to the imaging of devices with naturally occurring manufacturing defects.

1. Introduction

Recent developments show a dramatic increase in the power conversion efficiency (PCE) of organic photovoltaics (OPV) over the past years, mainly due to the emergence of novel non-fullerene acceptor materials,[1] with organic tandem devices routinely achieving values over 15%[2–4] and a record efficiency of 17.4% reported.[5] For this reason, OPV in general, and organic tandem solar cells in particular are becoming increasingly interesting for commercialization. Although the improvement in performance is crucial for organic solar cells (OSCs) becoming marketable, another just as important step that needs to be considered is the upscaling of the technology from small area laboratory cells to the fabrication of larger area modules using roll-to-roll processing.[6–9] One of the most critical challenges for upscaling is to avoid manufacturing defects such as pinholes, scratches or small dust particles on the surface since such defects typically cause an unwanted connection of two layers, which can lead to power dissipation for the whole device. To identify such problems as early as possible, reliable quality control measures that can provide in-depth information on possible manufacturing defects are essential. Besides standard electrical characterization with current-voltage curves (J–V curves), imaging analysis of solar cells can provide further valuable information on defects, most importantly the position on the samples, which can be vital for adjusting the production process accordingly.

To this end, a multitude of different imaging methods can be used to find defects and their position on a sample. Typically used imaging methods either probe the heat dissipation in a sample (lock-in thermography) or the radiative recombination in a sample (luminescence imaging). Both methods can be coupled with different means of excitation, typically electrical excitation—dark lock-in thermography[10,11] (DLIT) and electro-luminescence imaging[12,13]—or by optical excitation—illuminated lock-in thermography[14,15] (ILIT) and photoluminescence imaging[16]—with each providing different information on the physical processes in a solar cell.

While these imaging methods have been successfully used both for defect detection in many different single junction solar cell architectures, classic silicon solar cells,[17–22] but also on novel thin film cells like CIGS,[23–25] perovskite,[26–29] or organic solar cells,[30–37] tandem cells have only rarely been investigated.[38–43] Other imaging methods such as Ptychographic X-Ray Imaging[44] or light beam induced current (LBIC)[45,46] have also been successfully used for defect detection in organic tandem devices. For tandem solar cells, in-depth information on manufacturing defects is of special importance due to the increased number of deposited layers. The introduction of a second
absorber layer and a recombination layer for serially connecting the two photoactive layers can easily double the number of layers in the tandem configuration, thus increasing the overall probability for defects. This is especially the case for solution processed cells in ambient conditions due to the possibility for, for example, pinholes or dust particles in any of the layers.

Although using only one of the aforementioned imaging methods can be helpful for simply finding a defect and its position, using a combination of all different imaging methods can provide a deeper understanding on which type of defect is present in a sample. Depending on which layer is defective, a combination of imaging techniques often shows a unique response which allows for the exact pinpointing of a defect by layer.

The most direct way to determine this unique response for each defective layer is the introduction of artificial defects into each layer of the cell stack and subsequently performing imaging with different methods and excitations. In our previous work, we showed the introduction of artificial defects into single-junction OSCs by using fs-laser ablation and inkjet-printing layers intentionally containing pinholes.[47] However, the introduction of controlled defects into solar cells is also possible by using a variety of other means, for example, transfer lamination[48] or removing part of the layer with solvent.[49] In this work, besides using the established method of introducing defects by fs-laser, we resorted to masking part of our samples with tape during blade coating in order to introduce defects into certain layers of the tandem devices.

The photoactive layers of the investigated system consisted of two organic bulk heterojunctions using P3HT, Poly(3-hexylthiophene-2,5-diyl), and pDPPST-2, a diketopyrrolopyrrolequinque thiophene alternating copolymer, as electron donors in conjunction with PC60BM, Phenyl-C61-butyric acid methyl ester, as the electron acceptor. The subcells were connected in series with a recombination layer consisting of ZnO Nanoparticles and PEDOT:PSS (Figure 1a). Tandem systems consisting of one or both of these two donor polymer materials have been extensively studied and successfully used for both serially[38,50,51] and parallel[52] connected tandem cells. Both polymers can be processed by doctor blading under ambient conditions with good stability and reproducibility and an acceptable efficiency of ≈5% for tandem solar cells using these material systems. While tandem OPV with state of the art materials can reach much higher efficiencies,[53] the systems at hand were deemed suitable for this study. This is mainly due to their desirable properties such as, their complementary absorption spectra (Figure 1c), which allow for excitation of each individual subcell using monochromatic optical excitation of suitable wavelengths, which is ideal when probing the photoluminescence of each individual subcell.

**Figure 1.** a) Schematic of the investigated tandem device stack b) cell layout of the samples with artificial defects and reference cells on the same sample c) normalized absorption spectrum of single junction cells for both utilized active layer materials d) normalized photoluminescence spectrum of single junction cells for both utilized active layer materials.
2. Results and Discussion

2.1. Interface Layer Defects

The first investigated defects are concerning the interface layers between electrode and photoactive layer, namely the electron transport layer (ETL) on top of the bottom electrode (ITO) and the hole transport layer (HTL) below the top electrode (Ag). Defects into the bottom ETL were introduced using fs-laser ablation as discussed previously,[47] as an ablated circular area with a diameter of 400 µm. Between 1 and 3 defects have been introduced into the samples, leading to a defective area between roughly 1% and 5% of the complete cell area. The defected samples were investigated by standard J–V analysis and furthermore by electroluminescence imaging (EL), dark lock-in thermography (DLIT) and photoluminescence imaging (PL). For analysis of the images in the following, the signal intensity on the defected area was compared to not defected parts of the sample, when referencing an increase or a decrease in intensity. The PL images were recorded using two different excitation wavelengths, 532 and 808 nm, through the ITO-coated glass substrate. The 532 nm light is almost entirely absorbed by the bottom P3HT:PC60BM active layer, while the bottom active layer is almost transparent to the 808 nm excitation light, which then yields the PL signal from the top pDPP5T-2:PC60BM active layer. The EL signal, however, is the integral of the EL intensity over the whole spectrum originating from the combination of both subcells. Similarly, the DLIT signal is not sensitive to one of the subcells but shows heat dissipation for the whole tandem cell. The comparison of the J–V curves of a defected sample and a reference sample, as well as the imaging results, is summarized in Figure 2.

The J–V curves show practically no difference between the defective cell and the reference cell, which clearly indicates that these type of interface defects do not lead to obvious power dissipation for small amounts of defective area (up to ≈5% tested). This is similar to the effects observed for laser defects in the ETL of single-junction OSCs.[54] Also the effect on the images is practically the same, with a general reduction of the DLIT and the EL signal at the defect positions due to an increased barrier for charge injection at the ITO/AL interface in comparison to the ITO/ETL/AL interface. The PL signal, on the other hand, is only barely affected for either excitation wavelength used, with the signal being slightly lower at both wavelengths.

While the bottom ZnO layer can easily be removed via laser ablation, defects in the top HTL are more difficult to introduce without damaging the underlying layers. In the case of PEDOT:PSS as the top HTL, a possible method to introduce a defect is to apply a stripe of scotch tape to the layer after coating and drying and then remove it. The adhesion between the PEDOT:PSS layer and the top AL is relatively weak in comparison to the interaction between the scotch tape and the PEDOT:PSS layer, which allows for controlled removal of parts

Figure 2. a) Representative examples EL, DLIT, and PL Images of a full cell with artificial defects in the interface ETL with normalized intensity b) illuminated and dark (inset) J–V curves of the same serially connected tandem cell including artificial defects in the interface ETL and a reference cell on the same sample c) schematic representation of an ETL/HTL defect. Due to the increased injection barrier at the electrode-semiconductor interface compared to the electrode-interface layer-semiconductor interface, less charge carriers can be injected, thus lowering the EL/DLIT signal at the defect.
of the layer. The observations for imaging the top HTL defects are generally similar to the observations on the bottom ETL defects (Figure S1, Supporting Information). A clear difference, however, can be seen in the effect on the PL images. While the PL signal is slightly lowered for the bottom AL, similar as for the ETL defects, the PL signal is slightly increased for the top AL. While the reason for this increase in radiative recombination for one of the layers at the defect position is not completely clear, it could be used to clearly distinguish between top and bottom interface defects, at least for this system.

2.2. Active Layer Defects

Other layers in which manufacturing defects can occur are the two photoactive layers. For single-junction solar cells, such defects typically lead to a shunt in the sample due to the connection of the two interface layers (electrodes) forming an ohmic contact. These types of defects can usually easily be characterized by using DLIT since the current flowing through the ohmic contact leads to an increased heat dissipation. The defective cells typically exhibit most prominently a decrease in shunt resistance $R_{sh}$ as well as a decrease in $V_{OC}$, Fill Factor and PCE. For tandem cells, however, a defect in the active layer should not lead to an ohmic shunt due to the second active layer separating the interface layers.

The defects in the bottom AL were again introduced with fs-laser ablation at first using the same diameter ($d = 400 \mu m$) as for the ETL defects. The $J$–$V$ curves for a cell with one defect in the bottom AL compared to a reference cell and EL, DLIT, and PL images are shown in Figure 3. The $J$–$V$ curves show a significant decrease for the $V_{OC}$ while the other parameters are only barely affected, leading to a general decrease of the PCE as a result of the introduced defect. This loss in $V_{OC}$ when one of the subcells is defected or shorted was expected and already shown in simulations for different tandem systems.[58] Depending on how significantly the shunt resistance is decreased for the defected subcell, the $V_{OC}$ of the tandem cell decreases up to the value of the non-defected single subcell. To get a better idea of the reason for this $V_{OC}$ loss caused by the defect, we need to additionally consider the imaging results of the defective cell, starting with DLIT. In the DLIT image the defect appears as a bright spot in comparison to the rest of the cell area, meaning an increased heat dissipation. Usually, such a hot spot is associated with a short circuit in the cell. However, when an additional DLIT measurement at reverse bias was conducted, it did not show any heat signal at all from the sample, which means that the defect in the bottom active layer does not lead to a shunt pathway in the sample.

The EL image also shows a strong signal at the defect position. This means that despite the defect in the bottom active layer, radiative recombination still occurs at the defect position which can only originate from the top active layer. This could elucidate the reason for $V_{OC}$ loss induced by the defect in the bottom active layer. At the defect position, the tandem cell is effectively only operated as a single junction cell causing the $V_{OC}$ to decrease.

![Figure 3](image_url)

**Figure 3.** a) Representative examples of EL, DLIT, and PL Images of a full cell with an artificial defect in the bottom AL. For EL and DLIT absolute intensities are shown due to the very strong increase in signal at the defect position, while for the PL images the intensities were normalized. b) Illuminated and dark (inset) $J$–$V$ curves of the same cell including artificial defects in the bottom AL and a reference cell on the same sample c) schematic representation of an AL defect. The missing active layer at the defect position leads to lower resistance path for charge injection at the HTL–ETL–HTL interface compared to the rest of the sample.
This is further corroborated by comparison of $J-V$ measurements for different AL defect sizes. Introducing only small holes, with a length of ca. 50 µm and a width of ca. 30 µm, into one of the active layers results in the same appearance of the defects in both EL and PL Imaging as the larger defects, but no significant drop in the $V_{OC}$ (Figure S2, Supporting Information). However, the heat dissipation can no longer be resolved using DLIT for these defects, probably due to their small areas.

For the detection in which of the active layers the defect is located, PL imaging is most important. When a defect is located in the bottom AL, the PL signal at the defect position is completely absent when using 532 nm excitation (sensitive to the bottom P3HT:PC$_{60}$BM AL) while the signal is only slightly lowered when using 808 nm excitation (sensitive to the top pDPPST-2:PC$_{60}$BM AL). For a defect in the top AL, the case is the exact opposite, with the signal being quenched at the defect position when using 808 nm excitation, while the signal is only slightly lowered for 532 nm excitation (Figure S3, Supporting Information). Comparing defects in top and bottom AL in general, the increased EL and DLIT signal is characteristic for AL defects regardless of in which of the active layers the defect occurs (Figure S4, Supporting Information). Also, the effect on the $J-V$ curves is the same with a clear decrease of the $V_{OC}$ for larger defect areas. For this reason, conducting PL Imaging for each subcell is especially important for unique assignment of the defects to the correct layer.

### 2.3. Recombination Layer Defects

In order to connect the two subcells in series in a two-terminal tandem configuration, a recombination layer (RL) is necessary to enable the recombination of holes from one subcell and electrons from the other subcell. Thus, typically a recombination layer consists of at least two layers on top of each other, an HTL and an ETL, between the two photoactive layers. Besides ensuring an efficient series connection, some features an efficient recombination layer should include are optical transparency and robustness towards certain solvents in order to protect the bottom active layer from dissolution when applying the top active layer. In terms of manufacturing defects in the recombination layer three possibilities are conceivable: first, a defect in the HTL part of the RL second, a defect in the ETL part of the RL and third, a defect through the whole RL connecting both active layers.

Here, we only focused on the first and second scenarios. First, we investigated defects in the HTL part of the recombination layer (PEDOT:PSS in our case). The defects were introduced in the same way as the top HTL defects by applying a thin stripe of scotch tape to the sample after applying the PEDOT:PSS layer and then removing it, thus removing a thin stripe of PEDOT:PSS.

Figure 4 shows $J-V$ curve of a cell with a defect in the PEDOT:PSS part of the recombination layer compared to the curve of a reference cell and the EL, PL, and DLIT images.

![Figure 4](image)

**Figure 4.** a) Representative examples of EL, DLIT, and PL Images of a full cell with an artificial defect in the recombination layer with normalized intensity b) illuminated and dark (inset) $J-V$ curves of the same cell including artificial defects in the PEDOT:PSS part of the recombination layer and a reference cell on the same sample c) schematic representation of an RL (HTL) defect. The missing PEDOT:PSS layer leads to a dissolution of the bottom AL upon deposition of the top AL, rendering the defect area electrically inactive.
The $J-V$ curves show a clear loss of Fill factor and $J_{SC}$ for the defected cells, while the $V_{OC}$ of the cell is only slightly affected. The EL and DLIT images show no signal at all at the defect position, suggesting that the area is completely inactive, meaning that no power dissipation is directly caused by the defect. Furthermore, the PL signal for the bottom active layer is strongly quenched at the defect position, while the PL signal for the top active layer is much larger at the defect compared to the rest of the sample. As shown in Figure S5, Supporting Information, it is clear that the bottom active layer was simply dissolved by the application of the top active layer at the position where PEDOT:PSS was removed. This dissolution leads to a mixture of all the deposited layers, which exhibits a strong PL signal with 808 nm excitation, but is otherwise inactive. As mentioned before, the recombination layer serves a variety of purposes, including protection of the bottom active layer from dissolution. As a polymer which is mainly soluble in polar solvents, PEDOT:PSS, provides excellent protection of the bottom active layer from non-polar solvents which are typically used for the deposition of the photoactive layers. Thus, the deposition of a closed PEDOT:PSS layer is not only critical to prevent possible power dissipation from defects, but also generally to prevent the dissolution of the bottom active layer.

Second, defects in the ETL part of the recombination layer were investigated (a layer of ZnO nanoparticles in our case). The defects were introduced by masking part of the sample, coating the ZnO layer and then removing the mask, resulting in a defect in form of a thin stripe. Figure 5 shows the $J-V$ curves of a cell with a defect in the ZnO part of the recombination layer compared to a reference cell and the DLIT, EL, and PL images for the same sample.

The effect on the $J-V$ curves, in this case, is entirely different compared to the defects in the PEDOT:PSS part of the RL, with the $J_{SC}$ of the cell not affected, but a clear decrease in the $V_{OC}$ and FF of the cell. This leads us to the conclusion that defects in the ZnO part of the RL, in stark contrast to defects in the PEDOT:PSS part of the RL, do not lead to a dissolution of the bottom active layer upon further processing of the tandem cell. Also, a decrease in the shunt resistance compared to the reference sample can be observed. Thus, for the system at hand, the cause and effect of different recombination layer defects can be clearly distinguished. For the first case the part of active layers without PEDOT:PSS were mixed due to solvent penetration, which leads to reduced FF and $J_{SC}$ and the defected part being inactive. For the second case, the part of active layers without ZnO could still be separated by PEDOT:PSS, but due to no charge carrier selectivity for the top cell, it acts as the combination of the bottom subcell with a series resistant. This is confirmed by the imaging analysis, with a strong increase in signal at the defect position in EL imaging and DLIT. However, it is important to notice, that in PL imaging no strong change in signal can be observed.

![Figure 5](image_url)

Figure 5. a) Representative Examples of EL, DLIT, and PL Images of a full cell with an artificial defect in the recombination layer with normalized intensity b) illuminated and dark (inset) $J-V$ curves of the same cell including artificial defects in the ZnO part of the recombination layer and a reference cell on the same sample c) schematic representation of an RL (ETL) defect. The damage in the ZnO part of the RL leads to the top AL acting as unipolar hole conducting layer, lowering the resistance, and thus increasing the EL and DLIT signal.
regardless of the excitation wavelength. Thus, we can clearly discriminate between active layer defects and recombination layer defects, despite their very similar effect on the electrical parameters, the EL signal and the DLIT signal, by additionally recording PL images of the device.

For this particular system, it would be sufficient to apply EL imaging, DLIT, and PL imaging with two different excitation wavelengths to distinguish between defects in each layer of the tandem cell, since both possible types of recombination layer defects appear very differently. For a more generalized approach, disregarding the dissolution occurring for defects in the HTL part of the RL, a method to distinguish between which part of the recombination is affected needs to be established. This could be realized by not only measuring the PL intensity, but also the EL intensity of each subcell individually, since, depending on which part of the RL is damaged, the EL signal of each subcell should be affected differently. However, it is impossible to selectively excite only one subcell electrically in a two-terminal, serially-connected tandem cell. For this reason, optical filters are the only possibility to record the EL signal of only one subcell selectively. In order to properly select suitable filters that block the EL signal for one of the subcells, EL spectra of single-junction devices using both active layer materials and EL spectra of the tandem device were recorded (Figure 6).

The EL spectra show that the pDPP5T-2:PC60BM active layer emits mainly from roughly 800 to 1200 nm, similar to the PL emission, but also shows a weak emission deeper in the IR part of the spectrum. The P3HT:PC60BM active layer emits mainly from 1100 to 1400 nm, thus being strongly red-shifted compared to the PL emission from roughly 600 to 900 nm (Figure 1), since the EL emission from P3HT:PC60BM almost exclusively originates from charge-transfer states. The spectrum of the tandem cell generally overlaps with the P3HT:PC60BM mainly. Since the pDPP5T-2:PC60BM emission extends deep into the IR, overlapping with the P3HT:PC60BM spectrum, we decided to employ a short pass filter blocking all emission above 950 nm in order to observe the pDPP5T-2:PC60BM EL signal exclusively, while a long pass filter with a cutoff wavelength 1350 nm was used in order to observe P3HT:PC60BM mainly. Since the pDPP5T-2:PC60BM emission extends deep into the IR, overlapping with the P3HT:PC60BM emission, it was not possible to completely block the EL emission of the top subcell, but employing a long pass filter with a cutoff wavelength of 1350 nm allowed us to mainly observe bottom active layer. Figure 7 shows the EL images for both employed filters.

The images show that EL emission of P3HT:PC60BM is present over the whole sample, but strongly increased at the defect position, similar to the EL image without any filter employed. The EL signal of pDPP5T-2:PC60BM, on the other hand, while relatively homogenous over the non-defective parts of the sample, is completely quenched at the defect position. Because of the recombination layer defect in the ZnO layer, no electrons can be injected into the top active layer, making radiative recombination via electroluminescence impossible. For the bottom active layer on the other hand, the EL emission is increased with respect to the rest of the sample, since at the defect position the top active with two HTLs together act like a “thick” HTL, causing the cell to be operated as a single junction device at the defect position, leading to the observed drop in VOC and Fill Factor. As a summary of all investigated types of defects, in Table 1 all layers which can be affected by a manufacturing defect in the investigated system and their according effect on different imaging methods and electrical parameters are summarized.

![Figure 6. Normalized EL spectra of a P3HT:PC60BM and a pDPP5T-2:PC60BM single junction devices (applied bias = 1.2 V) and a tandem device using the same active layer materials (applied bias = 2 V).](image)

![Figure 7. EL images of the same cell as in Figure 5, recorded with a long pass filter with a cutoff wavelength of 1350 nm (left) and recorded with a short pass filter with a cutoff wavelength of 950 nm (right).](image)
2.4. Application on a Naturally Occurring Manufacturing Defect

It is shown in Figure 8 how the previously acquired knowledge from artificial calibration defects can be used for finding a naturally occurring defect on a sample with the same layer stack that was used before.

The first step (1) shows the recorded $J–V$ curve of two tandem cells on the same substrate, with dark and illuminated $J–V$ analysis being the standard method for testing the quality of any kind of photovoltaic device. Here, a drop in the $V_{OC}$ (from 1.047 to 0.983 V) and the Fill Factor (from 57.4% to 52.7%) can be observed, but at this point a clear determination of the loss pathway for the weaker performing cell is difficult.

As a second step (2), a DLIT amplitude image is recorded of the worse performing cell. In DLIT, a defect with increased heat dissipation is observed including exact information on the lateral defect position. This is already valuable information, but the exact nature of the defect remains unclear. The next step (3) is the recording of an EL image. The EL image shows an increased emission at the defect position, meaning that an ohmic shunt can be excluded as a reason for the drop in $V_{OC}$ and the increased DLIT signal. Also, a defect in one of the top/bottom interface layers can be excluded at this point, leaving only the two active layers and the recombination layer as a possibility for the location of the defect. To further narrow the position down, spectrally filtered EL imaging can be employed (4). Here, the defect shows an increased signal for one of the subcells (AL 1), while the signal is decreased for the other subcell (AL 2). From this it can be concluded that either the ZnO part of the recombination layer, or the top active layer (AL 2) is defected. Now PL imaging can be used to clearly determine which of the remaining possibilities applies (5). Here we can see a clear quenching of the PL signal at the defect position for the top active layer, which leads to the final conclusion that the top active layer is defected or in some way shorted, leading to the initially observed efficiency loss of the tested device.

Table 1. List of all possible manufacturing defects in the tested organic tandem system and the effect of each defect on all employed imaging methods and photovoltaic parameters.

| Bottom Electrode (ITO) | EL | EL (filtered—only AL 1) | EL (filtered—only AL 2) | PL (only AL 1) | PL (only AL 2) | DLIT | $J_{SC}$ | $V_{OC}$ | FF |
|------------------------|----|------------------------|------------------------|----------------|----------------|------|----------|--------|----|
| ETL (ZnO N10) → Figure 2 | – – | – – | – – | 0/– | 0/– | – 0/– | 0/– | – 0/– | 0/– | – |
| AL 1 (P3HT:PC60BM) → Figure 3 | + – | + – | + – | 0/– | 0/– | + 0 – | 0/– | 0/– | 0/– | – |
| RL/HTL (PEDOT:PSS) → Figure 4 | – – | – – | – – | – – | + – | – 0 – | 0/– | 0/– | 0/– | – |
| RL/ETL (ZnO N10) → Figure 5 | + + | + – | + – | 0/– | 0/– | + 0 – | 0/– | 0/– | 0/– | – |
| AL 2 (pDPPST-2:PC60BM) → Figure 3/S3 | + – | + – | + – | – 0/– | 0/– | – 0 – | 0/– | 0/– | 0/– | – |
| HTL (PEDOT:PSS) → Figure 2/S1 | – – | – – | – – | (no Reflection) | (no Reflection) | – (no Reflection) | 0/– | 0/– | 0/– | – |

“– –” = no signal; “–” = decrease; “0” = no change; “+” = increase.

Figure 8. Schematic demonstrating the general procedure for determining the vertical position in a cell stack for a randomly occurring manufacturing defect.
3. Conclusion

By introducing artificial defects into different layers of a tandem cell stack, a calibration routine for the detection of manufacturing defects in a typical organic tandem solar cell system was demonstrated. By analyzing the different effects of specific defects using several complimentary imaging methods including DLIT, EL imaging, and PL imaging, not only a determination of the lateral defect position, but also a determination of the vertical defect position in the tandem cell stack was possible. Using two different excitation wavelengths for PL imaging and spectral filters for EL imaging, allowed us to selectively probe only specific subcells or layers of the tandem stack. This way, an exact pinpointing of the layer affected by a defect was possible. While particularly relevant for tandem devices, the application of such a calibration routine using artificial defects, has the potential to greatly improve the quality control procedures for any solar cell manufacturing line, since the underlying principle could be applied to any thin film solar cell technology.

4. Experimental Section

Solar Cell Preparation and Characterization: The solar cells were prepared on ITO covered glass slides (25 mm × 25 mm × 1.25 mm). As a first step, the glass slides were cleaned for 10 min each in an ultrasonic bath with acetone and 2-propanol. After drying the substrates with pressurized air, all the solar cell layers were deposited using doctor blading in ambient atmosphere, except of MoO3 and Ag which were evaporated onto the samples. First, a ZnO nanoparticle solution (N10, Avantama) was coated on top of ITO at 30 °C (30 nm). After annealing at 80 °C for 5 min, the bottom active layer solution was applied at 60 °C (400 nm). The bottom active layer solution consists of a 1:1 blend of P3HT (Merck) and PC60BM (Solenne BV) at a concentration of 3.5 wt % in chlorobenzene with 5 % p-bromoanisole as an additive.[57] After applying the bottom active layer, the HTL part of the recombination layer, consisting of PEDOT:PSS (HTL, Solar, Heraeus) was coated onto the samples at 60 °C (40 nm). As the next step, the samples were annealed in a nitrogen atmosphere for 10 min at 140 °C. After the annealing step, a second layer of ZnO nanoparticle solution was applied at 30 °C (30 nm), serving as the ETL part of the recombination layer. This was followed by application of the top active layer at 60 °C (100 nm). The top active layer solution consists of a 1:1 blend of pDPPST-2 and PC60BM at a concentration of 2.4 wt % dissolved in a mixture of 90% chloroform and 10% o-dichlorobenzene. After that, the top hole transport layer was applied, using either again PEDOT:PSS (40 nm) coated by doctor blading at 60 °C, or using a 10 nm evaporated layer of MoO3. As the last step, a 100 nm layer of silver was evaporated as top electrode. The active cell area of the cells was about 10.4 mm². Finished devices were glass–glass encapsulated with an UV-curable adhesive (DELO Katiobond LP 655, Delo, Windach, Germany).

The J–V characteristics of the cells were measured with an Oriel Sol 1 A solar simulator from Newport (Irvine, California, U. S. A) under AM 1.5G irradiation. The light source was calibrated with a reference silicon solar cell. All cells were measured under ambient condition.

Imaging: For DLIT measurements an SC 8300 HD camera from FLIR System (Wilsonville, Oregon, U. S. A) was used. The camera is equipped with a Stirling cooled Indium–Antimonide (InSb) focal plane array detector (1344 × 784) which is sensitive between 3 and 5 µm. The lock-in measurements were carried out using a home-made LabVIEW (National Instruments—Austin, Texas, U. S. A) software. For EL and PL imaging a Ninox VIS-SWIR 640 camera from Raptor Photonics (Milbrook, Northern Ireland) was used. The camera is equipped with an air-cooled extended InSb–Galium–Arsenide detector (InGaAs) which is sensitive between 0.4 and 1.7 µm. The camera was controlled using the open source software “Micro-Manager”.[58]

Excitation for DLIT and EL Imaging was carried out with a Keysight B2901A Precision Source/Measure Unit from Keysight Technologies, Inc. (Santa Rosa, California, United States). For lock-in thermography, additionally a Berkeley Nucleonics Corporation (BNC) 555 pulse generator was used for synchronizing the camera and the excitation source. In case of DLIT a lock-in frequency of 4 Hz with an integration time of 5 ms was used. EL Images were measured with an integration time of 5000 ms and corrected for the background.

Excitation for PL Imaging was carried out with an RLMLG-532-2.5W 532 nm and an RLMLD-808-5W 808 nm fiber coupled laser diode (ROITHNER LASERTECHNIK GmbH) with a maximum output power of 2.5 and 5 W respectively. Both beams were expanded to an area of about 20 cm². An integration time of 100 ms was used for recording the PL images with either excitation wavelength. In addition to a background correction (subtraction of the image without illumination), a flat-field-correction was performed to account for typical deviations in illumination intensity (±10%).

For PL imaging, a long pass filter with a cutoff wavelength of 561 nm was used in front of the camera lens, in order to block reflections from the excitation beam. For spectrally filtered EL imaging measurements a short pass filter with a cutoff wavelength of 950 nm and a long pass filter with a cutoff wavelength of 1350 nm were used. All images were recorded from the glass substrate side. For PL imaging the samples were illuminated through the glass substrate and the transparent bottom electrode.

Introduction of Defects: The ETL and Bottom Active Layer defects were introduced using an “LS 7xF™” Setup (LS Laser Systems GmbH) with a “femtoREGEN™ UC-1040–8000 fs Yb SHC” laser source (High Q Laser GmbH) with a fundamental wavelength of 1040 nm. The laser was operated at 520 nm using SHG. The pulse duration is <350 fs at a repetition rate of 500 kHz. The laser has a beam quality of M² < 1.5 (@ 520 nm) and a pulse-to-pulse stability of <2%. The beam is scanned over the sample surface using a galvanometer-mounted mirror, which allows for patterning speeds of up to 4000 mm s⁻¹.

Defects in the PEDOT:PSS (HTL/RL) layers were introduced by placing a thin strip of scotch tape (d ≈ 1 mm) on the deposited PEDOT:PSS and then subsequently removing it, thus removing a thin strip of PEDOT:PSS. Defects in the RL ZnO layer and the top active were produced by masking part of the sample before deposition of the target layer with a thin stripe of Parafilm (d ≈ 1 mm), then coating the layer and subsequently remove the mask after drying.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements
This work was financially supported by the DFG research grant: BR 4031/6-1, “Entwicklung von bildgebenden Verfahren zur Defekterkennung in Tandem Solarzellen”. N.L. gratefully acknowledges the financial support from the DFG research grant: BR 4031/13-1. C.J.B. gratefully acknowledges the financial support through the “Aufbruch Bayern” initiative of the state of Bavaria (EnCN and “Solar Factory of the Future”), the Bavarian Initiative “Solar Technologies go Hybrid” (SolTech), and the SFB 953 (DFG, project no. 182849149).

Conflict of Interest
The authors declare no conflict of interest.
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