

A Commercial Benchmark: Light-Soaking Free, Fully Scalable, Large-Area Organic Solar Cells for Low-Light Applications

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Low-light applications provide an exciting market opportunity for organic solar cells (OSCs). However, so far, studies have only considered OSCs of limited commercial viability. Herein, the applicability of a fully-scalable, flexible, inverted non-fullerene acceptor (NFA) containing OSC is demonstrated by showing its superior performance to silicon under low-light, achieving $40 \mu\text{W cm}^{-2}$ maximum power output at 1300 lx illumination. The effect of parasitic resistance and dark current on low-light performance are identified. Furthermore, an atmosphere sensitive light-soaking (LS) effect, critical for low-light performance and resulting in undesirable S-shaped current-voltage characteristics, is analyzed. By employing different interlayers and photo-active layers (PALs) the origin of this LS effect is identified as poor electron extraction at the electron transport layer (ETL)/PAL interface when the common ETL ZnO is used. Two strategies are implemented to overcome the LS effect: replacement of ZnO with SnO_2 nanoparticles to reduce ETL sub-gap electron trap states or tuning the NFA energy levels to optimize interfacial energetics. Finally, the commercial viability of these LS-free devices is demonstrated by fabricating fully printed large-area modules (21.6 cm^2) achieving a maximum power output of $17.2 \mu\text{W cm}^{-2}$, providing the most relevant example of the currently obtainable performance in commercial low-light OSCs.

of non-fullerene acceptors (NFAs), with efficiencies now reaching 16–18%.^[1,2] The potential for OPV commercialisation has never been higher. This is reflected in current research with more studies having an emphasis on using scalable deposition methods and non-chlorinated solvents.^[3–6] Despite these improvements, the efficiency and stability of OPVs still lag behind other technologies such as perovskite and silicon solar cells. It is therefore important to investigate potential niche markets for OPVs that take advantage of their low weight, flexibility, roll-to-roll (R2R) processing compatibility and many tuneable properties. One such market is indoor and low-light applications.

Low-light refers to irradiance well below 1000 W m^{-2} (1 Sun).^[7] For indoor applications, the intensity of light is orders of magnitude less ($\approx 1\text{--}10 \text{ W m}^{-2}$), and is more often measured as photometric illuminance with the units of lux (lx) which takes into account the perception of the human eye. Values of between 200–1300 lx

are common for indoor lighting with the recommended illuminance for an office being 200–500 lx, and 750–1000 lx in large retail environments.^[8,9] For the solar spectrum 1000 lx equates to a radiometric irradiance of approximately 7.9 W m^{-2} . It is also important to note that the emission spectra of indoor lights, usually fluorescent bulbs or LEDs, is much narrower than that of the solar spectrum and located in the visible range.^[10] In this respect, NFA based OPVs are a particularly exciting prospect for low-light applications due to their easily tuneable visible and near-IR absorption spectra that can be matched to the spectra of different light sources maximising photon harvesting, something previously inaccessible with the use of fullerene acceptors. Additionally, the higher open-circuit voltages (V_{OC}) afforded by tuneable NFA energy levels, and the lower trap-mediated recombination of some acceptors are beneficial for low-light performance.^[11] Combined with tuneable aesthetic properties, form-fitting ability, low-weight, and potentially low-cost R2R fabrication they provide a genuine option for low-light applications.

The use of OPVs at low-light excitation has been shown to be superior to amorphous silicon, and has already been used to replace Si to improve the performance of a wireless sensor

1. Introduction

Organic photovoltaics (OPVs) utilizing sunlight to generate electricity continue to break efficiency records due to the advent

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node.^[12] For a detailed review of recent progress in OPVs for low-light, the reader is directed to a recent review by Ryu et al.^[13] We mention a few significant works below. A recent study by Lee et al. is particularly promising; they fabricate a conventional, solvent annealed, solution processed, small molecule-based donor (BTR) and acceptor (PC₇₁BM) bulk heterojunction device to achieve an impressive power output of 78.2 $\mu\text{W cm}^{-2}$ at 1000 lx under fluorescent lamp illumination.^[14] The emission spectra used has sharp peaks that coincide with absorption peaks of the electron donor molecule (BTR),^[15] highlighting advantages of the tuneable band-gap of organic semiconductor materials. Although encouraging, the conventional device structure and solvent annealing processing step used in this reported study are not suitable for commercialisation due to stability and scale-up issues. Similar performances using a polymer:NFA blend have been reported by Cui et al. again employing a conventional device structure to achieve 72.1 $\mu\text{W cm}^{-2}$ at 1000 lx.^[16] This performance is achieved due to the high V_{OC} of the device, 1.03 V at 200 lx (1.24 V at 1 Sun), the importance of a high V_{OC} for low-light performance is also addressed elsewhere,^[17,18] although it should be noted that a high V_{OC} is not essential, with Lee et al. achieving their impressive performance with a V_{OC} of just 0.79 V (0.95 V at 1 Sun). Cui et al. also demonstrated that after 1000 h of indoor light illumination there is no reduction in solar cell performance, indicating that the stability issues of OPVs at 1 Sun may not be so important at low-light.^[16]

Fill factor (FF) at 1 Sun is a far less important indicator of low-light performance, with all studies showing an improved FF at low-light. This is due to the different role of parasitic series and shunt resistances in the low photocurrent regime encountered at low irradiance. Low-light performance has been shown to be essentially independent of series resistance, showing no significant drop in performance until more than 1000 Ω of external resistance are applied to the device.^[16,19,20] It has been demonstrated that shunt resistance at 1 Sun, and the concomitant dark current, are critical in determining low-light performance, with a low shunt resistance/high dark current leading to FF and V_{OC} losses at lower light intensities.^[20,21]

There has been some effort to upscale OPVs for low-light, with a few large area modules being fabricated. Lee et al. fabricate a spin-coated PCDTBT:PC₇₁BM conventional module with an area of 100 cm² and achieve a maximum power output of 938 μW at 300 lx.^[18] Another group have utilised spin-coated small molecule blends in an inverted architecture to give power outputs of 111 and 95.4 μW at 200 lx in 9.5 and 9.6 cm² modules, respectively.^[22,23] The applicability of NFAs to low-light modules is also demonstrated when the NFA IT-4F is used in a 20.4 cm² module to achieve an impressive maximum power output of 40.2 $\mu\text{W cm}^{-2}$ at 1000 lx.^[24]

The examples above highlight the progress and potential of OPVs for low-light applications. However, these modules and most lab-scale devices in the literature employ at least one of the following components: evaporated top interlayers and contacts, conventional device structure, spin-coated layers, thin photoactive layers (PALs), glass substrates, solvent vapour annealing, or expensive PAL components. All of these components make the devices unfeasible for large-scale R2R manufacture and therefore limit the commercial viability of OPVs.

It is a well-observed phenomenon that OPVs require a few minutes of light-soaking (LS) to maximise FF and reach

a stable, optimal performance. The origin of LS induced performance improvement has been ascribed to different processes: ZnO UV activation,^[25] adsorbed oxygen on indium tin oxide (ITO),^[26] trap filling within the oxide layers,^[27] and ITO/electron transport layer (ETL) interface energetic barriers,^[28] and unfavourable phase-separation leading to energetic barriers.^[29] Often this LS effect is ignored at 1 Sun excitation, as at this light intensity the effect is already maximised after only a few minutes of solar illumination. Although it is common practice to light-soak a device before measuring the illuminated J - V curve, this LS effect on OPVs targeted for low-light applications, where it can be particularly detrimental, has not been investigated in detail, or by using commercially scalable device architectures, limiting an important market opportunity for OPVs.

Herein we investigate the low-light OPV performance of commercially viable inverted device structures, employing scalable fabrication methods and readily available relatively low-cost polymer (P3HT) and NFA (O-IDTBR) as active layer materials. In particular, the use of the homopolymer P3HT lowers the cost of the active layer considerably, whilst the improved stability and higher V_{OC} afforded by O-IDTBR compared to PCBM offsets the increased costs of the NFA synthesis.^[30–32] In these devices, we demonstrate superior low-light OPV performance compared to silicon and highlight the importance of dark current, and series-resistance-independent low-light performance. More importantly, we establish poor charge extraction at the interface between the ETL and the PAL as the origin of the LS effect which is particularly detrimental for OPV low-light performance. The presence of defect states in the metal oxide ETLs (e.g., ZnO) is attributed to be the key contributor for this by limiting ETL conductivity. This is further compounded by deep lowest unoccupied molecular orbital (LUMO) level of acceptors that provide a barrier to charge extraction. We demonstrate that by properly selecting ETL and NFA materials this undesirable interfacial barrier can be removed, and the LS effect can be avoided. We develop this further by fabricating a fully printed and R2R compatible large area module using our LS-free device structure, demonstrating the potential of OPVs for commercially scalable low-light applications.

2. Results and Discussion

2.1. Organic Versus Silicon Solar Cell Low-Light Performance

To highlight the applicability of NFA-based organic solar cells (OSCs) for low-light applications we first compare the performance of a standard silicon reference cell against a commercially viable, laminated OSC with the inverted device structure IM1/ZnO/P3HT:O-IDTBR/PEDOT:PSS/AgSP (see Experimental Section for more information) (**Figure 1**). We highlight the use of a thick PEDOT:PSS layer (≈ 290 nm in total) that is required to protect the PAL from the pressure applied during the screen-printing process used to deposit the Ag contact. The extracted device parameters of these devices are shown in **Figure 1**. Both devices show a fairly linear reduction in short-circuit current (J_{SC}) with decreasing light intensity as expected. However, the maximum power output decrease of the silicon cell deviates from this linearity at low-light excitation due to

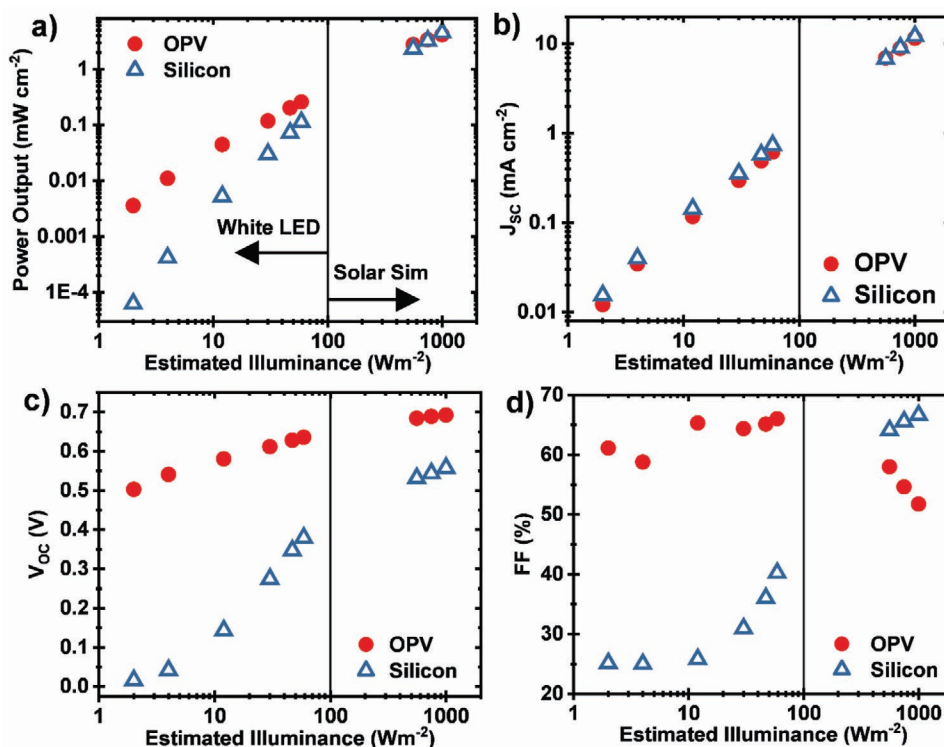


Figure 1. Maximum power output (a), J_{sc} (b), V_{oc} (c), and FF (d) as a function of illuminance for both a laminated, scalable OSC (IMI/ZnO/P3HT:O-IDTBR/PEDOT:PSS/AgSP) and a silicon reference cell.

significant losses in both FF and V_{oc} . The OSC, on the other hand, shows only a small decrease in V_{oc} with decreasing light intensity, and an increased FF due to improved charge collection arising from an improved mobility lifetime product of photogenerated charge carriers at low-light compared to 1 Sun illumination.^[14] This stable FF, and minimal V_{oc} loss of the OPV device is consistent with the characteristics previously reported on spin-coated devices and highlights the superior performance of OPVs for low-light applications.^[14] Our R2R compatible device architecture gives reproducible low-light performance with the best device achieving $44.4 \mu\text{W cm}^{-2}$ at 1300 lx under a typical white LED used for indoor lighting applications. We return to discuss the commercial viability these OPVs for large area low-light application in the final section of this work.

2.2. The Effect of Parasitic Resistances on Device Performance

We now investigate the PAL thickness dependence on low-light performance of fully scalable OPV devices to investigate the effect of parasitic resistances. At 1 Sun (Figure 2a) there is clear thickness-dependent performance with thinner and thicker devices performing worse than the optimal 320 nm PAL devices; this trend closely follows the linear trend in J_{sc} , with thin devices limited by absorption and thick devices limited by charge extraction and trap states in the PAL (Figure 2c).^[33] The poor extraction in the thicker devices (590, 757, and 1125 nm) is characterised by very low FFs, with the 1125 nm devices showing an average FF of 31%. This low FF is caused by an excessive series resistance (average of 70Ω for 320 nm devices

and 119Ω for 1125 nm devices) as expected for much thicker devices. The V_{oc} is also seen to decrease with increasing active layer thickness mainly due to the reduced built in field in thicker devices. We notice that some of the 1125 nm devices performed well with improved J_{sc} and FF, but with very large standard deviation.

Importantly, at 400 lx excitation (post-LS, this is defined in the following section), all thicker devices (>320 nm) show good performance with $15 \mu\text{W cm}^{-2}$ of max power output (Figure 2b). This highlights the ruinous series resistance at 1 Sun being less important at low-light excitation, consistent with reports on conventional architecture devices as mentioned above,^[16,19] and confirms series resistance independent low-light performance on scalable inverted OPV devices. Conversely, the thinner devices, including the devices optimised for 1 Sun excitation condition, perform poorly. To understand possible causes, we first look at the relationship between low-light performance and reverse dark current (Figure 2d). At dark currents greater than $\approx 0.03 \text{ mA cm}^{-2}$ (black dividing line) there is a log-log linear relationship between dark current and low-light performance at 1300 lx illumination, with increasing dark current leading to reduced maximum power output, consistent with conventional architecture devices reported.^[19–21] Below a dark current threshold of 0.03 mA cm^{-2} low-light performance is essentially independent of dark current. The threshold for independent low-light performance shifts to a lower level of dark current at lower light excitation (e.g., $\approx 0.015 \text{ mA cm}^{-2}$ at 200 lx), indicating that lowering dark current is more critical at lower light excitation conditions to achieve maximum power output. A relationship between shunt resistance (measured at

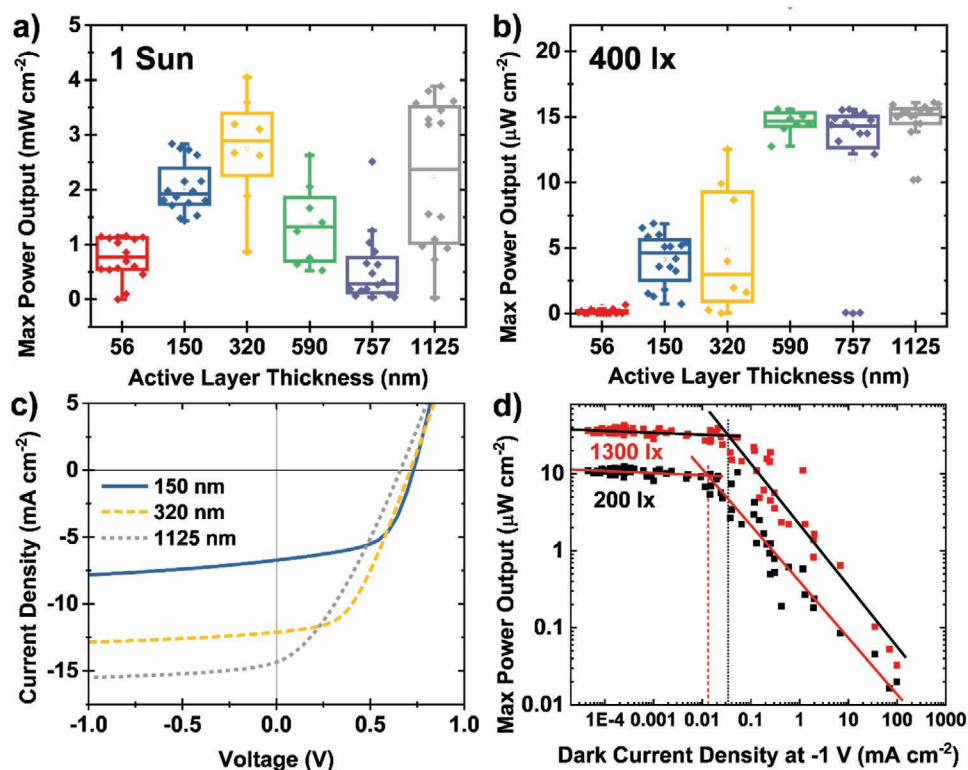


Figure 2. PAL thickness-dependent device max power output for IMI/ZnO/P3HT:O-IDTBR/PEDOT:PSS/Ag devices at a) 1 Sun and b) 400 lx illumination; c) representative $J-V$ curves of thin (150 nm), optimized (320 nm) and thick (1125 nm) PAL device at 1 Sun, highlighting the loss of J_{SC} in thin devices and the detrimental effect of series resistance for thick devices at this illumination. d) relationship between dark current density at -1 V and device performance at 1300 and 200 lx, linear fits are shown to highlight the regions in which maximum power output is dependent and independent of dark current density.

1 Sun) and low-light performance is also observed but with more data scatter than when dark current is compared (Figure S3, Supporting Information) indicating that dark current is a better criterion of low-light performance than shunt resistance. Thinner devices are poor at low-light excitation due to the generally higher dark currents they exhibit, consistent with the relationship detailed above. These findings highlight the importance of optimising and designing devices specifically for low-light applications; devices that do not work at 1 Sun can perform admirably at low-light. This is particularly promising for large scale roll-to-roll printing where a thicker PAL is easier to process with higher film uniformity.

2.3. Effects of LS on Low-Light Device Performance

Whilst characterising low-light performance we commonly observed a distinctive LS effect, which we investigate further below. Unless otherwise stated, we define the LS effect as initial device characteristics changing after about 1 min of 1 Sun illumination (solar simulator, 1000 W m^{-2}). Hereafter, the term low-light indicates 1300 lx illumination.

Figure 3a shows the 1 Sun LS effect on low-light performance of OPVs. Prior to LS, the $J-V$ curve under low-light excitation shows a distinct S-shape, limiting FF. After LS, the curve changes to a conventional J-shape with an increased FF

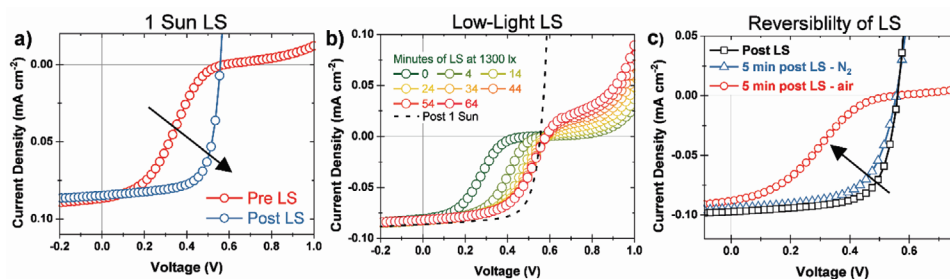


Figure 3. a) The impact of 1 Sun LS on low-light (1300 lx) excitation performance of an IMI/ZnO/P3HT:O-IDTBR/MoO_x/Ag device; b) low-light LS effect on low-light performance of a similar device; c) reversibility of LS effect in different atmospheric conditions of an IMI/ZnO/P3HT:O-IDTBR/PEDOT:PSS/Ag device.

and a 1.5× increase in maximum power output (Figure S4, Supporting Information).

A similar but much slower LS effect is achieved when devices are illuminated by low-light instead of 1 Sun. Upon continuous low-light exposure (60 min), the initial S-shaped $J-V$ curve changes to J-shape (Figure 3b) with a concomitant increase in the maximum power output of the device from 12 to 27 $\mu\text{W cm}^{-2}$ (Figure S4, Supporting Information). The low-light excitation intensity (1300 lx) used here is still relatively high for certain indoor applications and we presume that at a much lower light intensity this LS effect would take even longer to saturate.

Importantly, the LS effect is found to be non-permanent, with light-soaked devices returning to the initial S-shaped $J-V$ curve after stopping LS, the rate of which depends on the environment in which the device is kept and device architecture. If kept under nitrogen the return to the initial S-shape is much slower compared to in air and even slower if the device is continuously operated under low-light excitation, this is highlighted in Figure S5, Supporting Information. This return to the s-shape is also slowed by replacing the printed PEDOT:PSS/AgSP hole transport layer (HTL) with evaporated MoO_x/Ag . These results indicate that the LS effect is sensitive to air, with the more permeable top contacts the more rapid diminishing of its effect. They also highlight undesirable unstable power outputs and demonstrate that an initial 1 Sun LS of OPV devices is insufficient to ensure constant high device performance for low-light applications when devices are not encapsulated or not optimised.

2.4. Effects of Charge Transport and Photoactive Materials on LS

Now, we investigate the effects of ETL, HTL, and PAL materials on the LS effect. Figure 4 shows representative $J-V$ curves at 1300 lx for a variety of materials in devices. For the statistical

device data at 1300 lx before and after LS, as well as at 1 Sun the reader is directed to Figures S6 and S7, Supporting Information. The effects of different ETL and HTL materials on LS are tested with the cost-effective and scalable P3HT:O-IDTBR PAL.^[34]

For different ETL materials, a clear LS effect is observed when the common sol-gel ZnO ETL is employed regardless of HTL materials. ZnO is known to be sensitive to UV light. If we conduct strong UV treatment of the ETL prior to PAL deposition, the resulting devices show a reduced LS effect, but device performance is significantly reduced limiting the usefulness of this as a method to remove the LS effect (for more information see Figure S8, Supporting Information). Instead, we replace ZnO with SnO_2 nanoparticles and find that this effectively removes this LS effect, with SnO_2 containing devices showing reproducible J-shaped $J-V$ curves before and after LS. Although SnO_x has been shown to be beneficial to remove LS effects at 1 Sun,^[35] its effect on low-light applications in particular when using solution-processable SnO_2 nanoparticles as an ETL hasn't been reported.

For different HTL materials, if MoO_x is replaced by the more industrially scalable organic PEDOT:PSS (with ZnO ETL), a more pronounced initial S-shape is observed, despite post-LS performance being similar for both HTL devices. In contrast, devices with SnO_2 ETL show no such HTL dependence in initial and post-LS device performances, with only a small reduction in J_{SC} for PEDOT:PSS device. This suggests that the choice of ETL is more critical than the HTL in determining the LS effect.

To investigate this further, we also tested different PAL materials with the same ZnO ETL and MoO_x HTL materials. The performance of these devices at 1 Sun, and a brief discussion concerning these are summarised in Figure S7, Supporting Information. Similarly to the P3HT:O-IDTBR devices, both the archetypal P3HT:PC₆₀BM and high-performance PBDB-T:ITIC

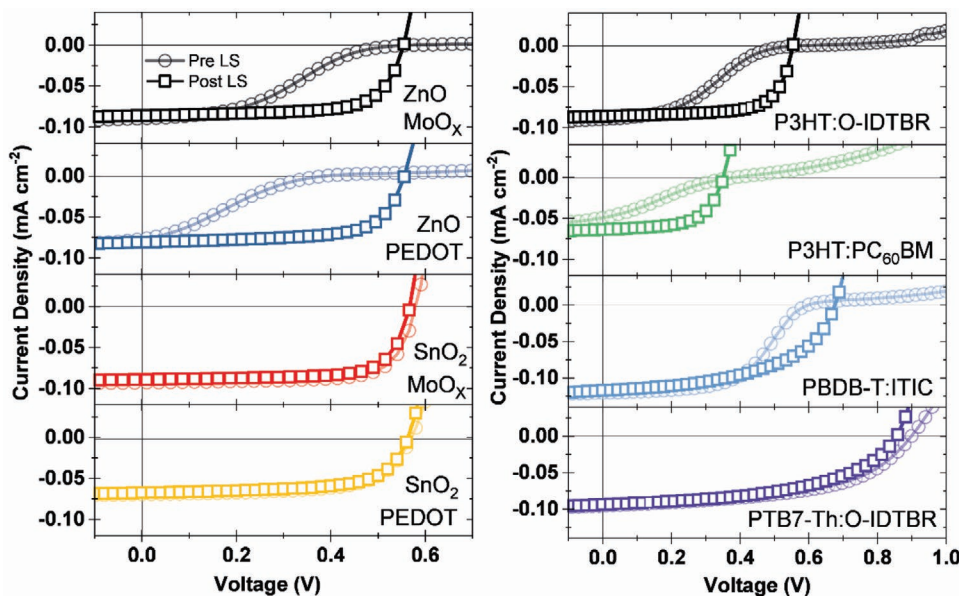


Figure 4. Representative $J-V$ curves, taken at 1300 lx, before and after LS at 1 Sun. In the left column, the active layer is P3HT:O-IDTBR with different ETLs and HTLs; the right shows different PAL with ZnO and MoO_x .

blend devices show a clear LS effect. Surprisingly, the S- and J-shaped curves of PBDB-T:ITIC have a similar maximum power (Figure S9, Supporting Information). This stems from the high dark current (0.1 mA cm^{-2}) observed in the PBDB-T:ITIC devices after LS, which as highlighted above is detrimental to low-light performance. Promisingly devices utilising the high-stability and high-performance PTB7-Th:O-IDTBR PAL display no LS effect, demonstrating a near stable performance, albeit with a slight decrease in V_{OC} after LS. This signifies that the LS effect is not solely dependent on ZnO ETL but is instead sensitive to the ETL/PAL interface.

2.5. The Origin of The LS Effect

To investigate the ETL/PAL interface and the origin of the LS effect we compare P3HT:O-IDTBR devices with ZnO and SnO_2 ETLs in more detail. We first probe the physical interface by water contact angle measurements of the ETLs. The contact angle of ZnO (44.3°) is smaller than SnO_2 (54.4°) indicating a more hydrophobic nature of the SnO_2 surface (Figure S10, Supporting Information). This would result in improved wetting of the hydrophobic PAL solution, possibly leading to a more homogenous ETL/PAL interface in SnO_2 devices. We next investigate the effect of LS on the dark currents of full devices as shown in Figure 5a,b. Prior to LS, the ZnO device exhibits poor diode characteristics with a small rectification ratio in current (e.g., 10^2 at $\pm 1\text{V}$). After LS the current in forward bias is increased significantly by >3 orders of magnitude, indicating improved electron injection from ZnO to the PAL, leading to a large rectification ratio (10^5 at $\pm 1\text{V}$). SnO_2 devices on the other hand show a large rectification ratio in current before LS (10^4), with only a small improvement after LS. This LS effect on the forward bias dark current, where ZnO ETL shows a large LS effect, whilst SnO_2 ETL does not, is similar to what is observed for low-light device performance (Figure 4).

The ETL/PAL interface was further investigated by surface photovoltage (SPV) measurements (Figure 5c). SPV which measures the change in surface potential upon illumination induced by the redistribution of photogenerated charge carriers within the PAL has been used as a powerful tool to probe charge accumulation and extraction at ETL/PAL and HTL/PAL interfaces.^[36–38] A quartz tungsten halogen lamp with excitation ranging from 1.2–3.1 eV is used. When illuminated at a nominal

intensity of 14 mW cm^{-2} the ZnO/PAL samples show a high, negative SPV signal (-428 mV) with slow, non-square turn-on and turn-off characteristics. The high, negative SPV signal represents efficient electron extraction from PAL to ETL leading to high hole accumulation on the PAL surface upon illumination. The slow SPV transient behaviors, however, suggest that this charge extraction process is initially slow at the ZnO/PAL interface and significant charge trapping occurs after the light is turned off. Compared to ZnO/PAL, the SnO_2 /PAL samples show a similarly high SPV value (-354 mV) but a much faster turn-on/off transient without any long-lived charge trapping, indicating highly efficient electron extraction at the SnO_2 /PAL interface.

A similar SPV measurement is also conducted on ZnO and SnO_2 ETLs only, without the PAL on top. It is important to notice that ZnO ETL itself generates a much larger, negative SPV signal with very slow turn-on/off transients and a continuous increase under illumination (180 mV after the first 100 s of illumination, Figure 6a). The slow SPV transient behaviors are similar to those observed in the ZnO/PAL samples, indicating that the ZnO is responsible for such slow photoresponse of the ZnO/PAL samples and potentially causing the LS effect in OPV devices. For SnO_2 samples, the magnitude of SPV (36 mV after 100 s of illumination) and slow photoresponses are significantly reduced.

We consider the possible causes of such a large negative SPV response and slow turn-on/off characteristics of the ZnO samples. ZnO has an optical bandgap of 3.38 eV ^[26] larger than the excitation energy ($<3.1 \text{ eV}$) used for SPV measurements, so we exclude direct optical band gap excitation as a cause for generating SPV signals. Instead, we attribute it to the excitation of the sub-gap electronic states (e.g., trap states such as oxygen vacancies, ionic defects, and adsorbed species).^[41–44] This sub-gap electronic state excitation was further confirmed by the presence of a clear negative SPV response when excited with a smaller energy light source (2.7 eV , Figure S11, Supporting Information). The negative sign of the SPV signal indicates a reduced density of electrons at the surface of the ZnO layer under illumination. This negative SPV signal is expected for metal oxide n-type semiconductors such as ZnO where electrons are initially trapped in their sub-gap states causing an upward band-bending, which is removed under illumination as the trapped electrons are photo-excited to the conduction band (CB) and transported away (Figure 6b). Such de-trapping

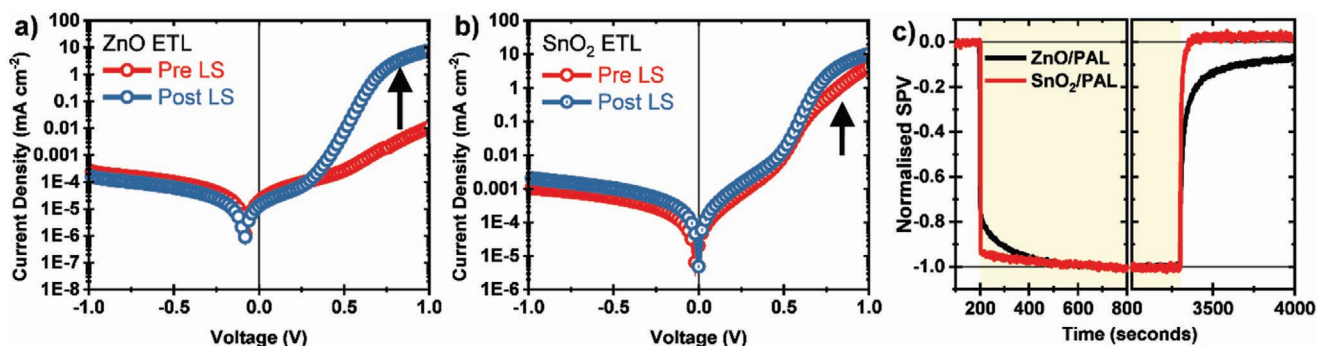


Figure 5. Representative dark J - V curves of a) IMI/ZnO/P3HT:O-IDTBR/ MoO_x /Ag and b) IMI/ SnO_2 /P3HT:O-IDTBR/ MoO_x /Ag devices before and after 1Sun LS; c) Normalised SPV measurements of samples with the structure IMI/ETL/P3HT:O-IDTBR.

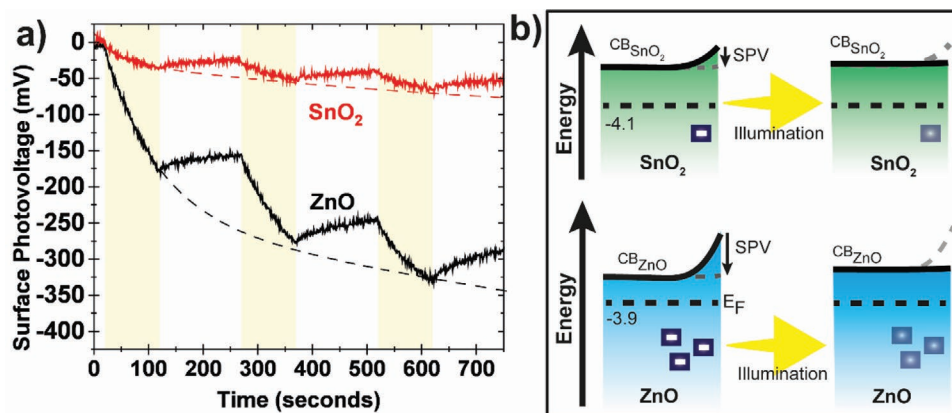


Figure 6. a) SPV measurements (initial value set to 0) of different ETLs without the PAL, highlighted regions indicate white light illumination, the dashed lines are exponential fits of the first illuminated period; b) Schematic showing the effect of illumination on the ETL surface, negative squares indicate filled intergap trap states that generate band-bending at the interface, after LS these charges are freed to the CB reducing the band-bending and generating a surface photovoltage.

of electrons from their trap states will increase the conductivity of ZnO.^[45]

In the device, these sub-gap trap states would initially inhibit extraction of photogenerated electrons from the PAL. With sufficient illumination, the de-trapping of ZnO sub-gap states may occur enabling efficient electron extraction at the ZnO/PAL interface and efficient electron transport within ZnO ETL. It can also account for the large increase in forward dark current of the ZnO devices after LS, as observed. This correlates well with the slow turn-on seen in the ZnO/PAL interface, during which the electron extraction and transport processes are improving as de-trapping of the ZnO sub-gap states is slowly initiated. Conversely, the fast turn on in the SnO₂/PAL samples correlates well with the low density of sub-gap trap states observed.

In addition, our observation on the oxygen sensitivity of the LS effect of ZnO devices (Figure 3c) indicates that electron trapping and de-trapping processes in ZnO sub-gap states can be mediated by oxygen adsorption and desorption. It has been reported that oxygen can adsorb to ZnO; the O₂ is ionized by free electrons in the ZnO which effectively traps the electron.^[45–47] Upon illumination photogenerated holes can quench these sites leading to desorption of molecular oxygen, freeing an electron to the CB, and improving conductivity. Compared to ZnO, we find a much smaller increase in forward current after LS in devices utilizing SnO₂, as well as the presence of much smaller SPV signals of the SnO₂ layer itself. These observations suggest that a similar light-driven de-trapping process occurs in the SnO₂ layer too, but these effects are much smaller than ZnO, possibly due to fewer defect/trap states present in SnO₂. The effect on charge extraction can also be dampened by the intrinsically higher carrier mobility of SnO₂.^[48]

Finally, we comment on why the LS affects device performance for certain PALs more than others. As discussed above, most devices with a ZnO ETL show a strong LS effect except the PTB7-Th:O-IDTBR blend (Figure 4). We consider the interfacial energetics at the ETL/PAL interface to understand these findings. The energy levels of the ETL/PAL materials are shown in Figure S12, Supporting Information. ZnO has a work function of -3.9 eV whilst SnO₂ has a slightly deeper work function

at -4.1 eV. As both are strong n-type materials their CB edge is expected to be close to the E_F (e.g., only ≈ 0.1 – 0.2 eV shallower than the E_F).^[40,49] Although it is difficult to measure the exact LUMO energy levels of NFAs in blends, the NFA LUMO levels are estimated to be -4.0 and -4.2 eV for IDTBR and ITIC respectively (see details in Experimental Section). Both deep LUMO levels would result in an electron extraction barrier with ZnO, which would result in an initial S-shaped J – V curve prior to LS, consistent with previously reported S-shaped J – V curves.^[50] As well as this barrier to electron extraction, the poor conductivity of ZnO prior to LS as discussed above is likely to inhibit efficient charge extraction and transport. We note that with the reported LUMO level of PCBM (-3.9 eV)^[51] there exists a smaller energetic barrier at the ZnO/PCBM interface. We postulate that the higher electron mobility of PCBM compared to O-IDTBR ($\approx 10^{-3}$ cm² V⁻¹ s⁻¹ and $\approx 10^{-6}$ cm² V⁻¹ s⁻¹ respectively)^[31,52] can lead to an accumulation of electrons at the ZnO/PCBM interface leading to resultant recombination losses, which will produce the non-optimized S-shape J – V characteristics as observed. SnO₂ has a higher electron mobility and with its deeper WF and CB, provides no energetic barrier to electron extraction, so no LS effect is observed when utilized as an ETL.

Interestingly the LS effect is not significant in the PTB7-Th:O-IDTBR devices despite utilizing O-IDTBR. It is well known that the O-IDTBR acceptor can adopt different crystallinities in blends depending on the counterpart donor polymer used and fabrication processes applied.^[53,54] More crystalline domains (aggregates) of O-IDTBR acceptors are known to be formed when they are blended with semi-crystalline polymers such as P3HT. This crystalline, aggregate form of O-IDTBR shows a deeper LUMO energy level than less crystalline, non-aggregated O-IDTBR (-4.0 eV and -3.9 eV for aggregated and non-aggregated respectively). When blended with PTB7-Th polymer, the crystallinity of O-IDTBR is strongly suppressed, as demonstrated by the blue-shifted acceptor absorption (Figure S13, Supporting Information), leading to a shallower LUMO level. This shallower LUMO of O-IDTBR in PTB7-Th:O-IDTBR blend may reduce the energetic barrier at the ZnO ETL/PAL interface, removing the S-shape and thus the LS effect in J – V characteristics. We note that despite the LS effect not affecting

photovoltaic performance of PTB7-Th:O-IDTBR devices, there is still an increase in forward diode current after LS (Figure S14, Supporting Information) that results from the improved ZnO conductivity after LS.

2.6. Large-Scale Module Fabrication and Performance

To demonstrate the commercial viability of our LS free device structure (IMI/SnO₂/P3HT:O-IDTBR/PEDOT/AgSP) we fabricated fully scalable, R2R compatible modules optimized for low-light, comprised of six cells in series with a total area of 21.6 cm². To reduce dark current and increase photon collection we use a thick PAL of 590 nm. The best 1 Sun module *J*-*V* characteristics utilizing either SnO₂ or ZnO as an ETL are shown in Figure 7a. The best ZnO module achieved *V*_{OC} 4.19 V, *J*_{SC} 10.45 mA cm⁻², FF 48%, and PCE 3.44%, whilst the SnO₂ module gave a *V*_{OC} 4.18 V, *J*_{SC} 10.21 mA cm⁻², FF 38%, and PCE 2.68%. The FF and performance of the SnO₂ module are particularly limited by a high series resistance (74 Ω) and lower shunt resistance (673 Ω) than the ZnO modules (48 and 805 Ω respectively).

Figure 7b shows the *J*-*V* characteristics of the best performing modules at 1300 lx before and after LS. Similar to the lab-scale, ZnO modules show an S-shaped *J*-*V* curve prior to LS whilst those utilizing SnO₂ do not. At 1300 lx the maximum power output of the SnO₂ module is 21.7 μW cm⁻² which is approximately two thirds of that achieved at the lab-scale (29.5 μW cm⁻²) with a similar structure albeit with a thinner PAL and HTL and evaporated silver contacts. *J*_{SC} of the SnO₂ module (0.86 mA cm⁻²) is slightly improved compared to the lab-scale (0.68 mA cm⁻²) due to the thicker PAL in the module enabling more photon absorption. The *V*_{OC} of the module is 3.03 V, which equates to 0.50 V per cell, slightly lower than the lab-scale at 0.57 V. Whilst the FF is reduced from 65% to 50% upon scaling to the module. The performance of the SnO₂ module is maintained after encapsulation albeit with a slight decrease in *V*_{OC} and slight increase in *J*_{SC} (Figure S15, Supporting Information). We ascribe the losses upon scaling to increased parasitic resistance losses (increased series resistance and dark current) and non-optimal film uniformity. With further optimization of the PAL deposition and isolation of the cells, these losses can be reduced, and device performance can be further improved.

Figure 7c shows the SnO₂ module performance as a function of illuminance and a photograph of the module before

encapsulation, the other device parameters as a function of illuminance are shown in Figure S16, Supporting Information. The dependence is comparable to that observed in the lab-scale smaller devices, with the max power output and *J*_{SC} decreasing close to linearly, *V*_{OC} decreasing and FF improving. In the module, compared to the lab-scale, the *V*_{OC} decreases more with decreasing illuminance whilst the FF increases more. This results in the module showing 0.67% of the 1 Sun power output at 1000 lx, whilst the lab-scale is slightly lower at 0.56%.

After encapsulation, our LS-free module achieves a maximum output power of 17.22 μW cm⁻² at 1000 lx (17.95 μW cm⁻² before encapsulation). We believe that our module is the most demonstrative example of commercially scalable OPVs for low-light applications. This is because we have ensured the absence of LS; utilized a thick active layer (~590 nm); used a flexible PET substrate; the ETL, PAL, and HTL are all blade-coated in air from non-chlorinated solvents; the silver contacts are screen-printed, requiring a very thick HTL; and the PAL itself is comprised of synthetically simple and cost-effective materials that have already demonstrated scale-up potential. Modules measured at low-light in the literature fail to address all of these components, in particular, all inverted devices measured at low-light in the literature have MoO_x/Ag evaporated top contacts, something which is not viable for industrial scale-up.^[22–24] With all of these key factors considered, the power output density we achieve is promising for an R2R processing compatible device and provides an exciting starting point for further optimization and development. A few square centimeters of these cells would be able to power small electronic devices such as remote controls, smart labeling, or RFID Tags.^[55]

3. Conclusion

We have demonstrated the promising potential of commercially viable flexible inverted NFA containing OSCs for low-light applications, particularly their superior performance compared to silicon at indoor light intensities. PAL thickness-dependent measurements highlight the critical relationship between dark current and low-light performance and demonstrate that this performance is independent of series resistance. These conclusions provide a clear approach for the optimization of devices for low-light usage by increasing the thickness of the PAL to reduce dark current. This is particularly important as thick layers are easier to manufacture by R2R processes. On top of

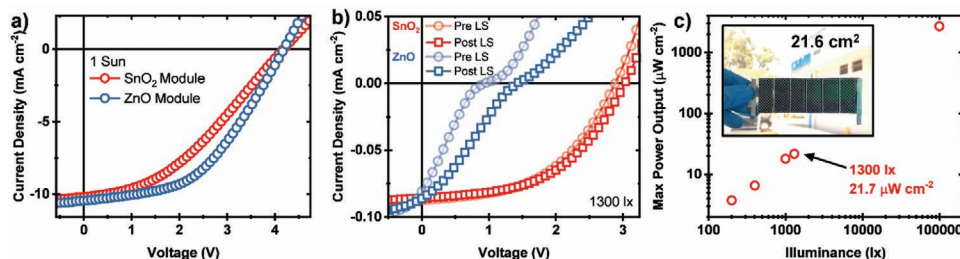


Figure 7. *J*-*V* curves of fully printed 21.6 cm² modules with the device structure IMI/ETL/P3HT:O-IDTBR/PEDOT:PSS/AgSP the ETL is either SnO₂ or ZnO, at 1 Sun (a), and at 1300 lx before and after 1 min of 1 Sun LS (b), light intensity-dependent performance of the best SnO₂ containing module, and photograph of said module (c).

this, our results highlight that screening potential low-light devices using 1 Sun illumination is not appropriate, as some devices that perform well at low-light appear to not work at all at 1 Sun.

More importantly, we have observed and characterized an atmosphere dependent LS effect that is critical for low-light performance of inverted OSCs. Prior to LS, the devices are limited by poor charge extraction at the ETL/PAL interface when ZnO ETL is employed due to its high density of sub-gap trap states and is compounded by the non-optimized energetics at the ZnO/PAL interface. Two clear strategies for avoiding this LS are demonstrated that overcome this poor charge extraction: replacing the ZnO ETL with SnO₂ nanoparticles or by employing an acceptor with a sufficiently shallow LUMO level to avoid any electron extraction barrier.

We take this further and fabricate a fully printed, large area, commercially viable module with the LS free SnO₂ ETL. Rather than optimize materials specifically for low-light applications we have used readily available materials that have shown scale-up potential. Doing so we achieve a performance that sets a promising precedent for the use of OPVs in low-light applications. For this exciting market opportunity to be realized, research must focus on optimizing and investigating devices specifically for low-light and transferring the state-of-the-art PAL materials in the literature to fully scalable devices.

4. Experimental Section

Device Preparation: Materials were purchased from a variety of sources for full details of these and the equipment used please see the ESI. All devices were fabricated on mechanically scribed, flexible PET/IMI (polyethylene terephthalate/ITO/Ag/ITO) substrates. Devices had an active area of 0.55 cm² whilst the large area module was comprised of six cells of 3.6 cm² in series giving a total area of 21.6 cm². All ETLs, active layers, and PEDOT:PSS layers were blade-coated from non-chlorinated solvents in air using optimized deposition parameters and annealing temperatures giving a variety of thicknesses, these are summarised in the supporting information (Table S1 and S2, Supporting Information). All ZnO layers were prepared using the sol-gel method, whilst SnO₂ layers were prepared from a nanoparticle dispersion. MoO_x layers, where used, were thermally evaporated to a thickness of 9 nm Silver electrodes were either screen-printed (AgSP, ≈12 μm) or evaporated (AgEvap, 200 nm). Cells were isolated using a laser where necessary.

J–V Measurements: J–V curves were obtained in dark, and under different illumination intensities. High irradiances (1000–500 W m^{−2}) were achieved using a calibrated solar simulator, low-light measurements were conducted under variable intensity white-light LED sources, the spectrum of which is shown in Figure S17, Supporting Information. A lux meter was used to measure illuminance whilst conversion to radiometric units was estimated by using the current generated by a c-Si reference cell (certified and sold by PV Measurements Inc, PVM 874) at different light intensities. It was appreciated that this was a flawed method of calculation and so power conversion efficiencies ($P_{\text{MAX}}/P_{\text{IN}}$) at low-light are an unreliable guide to device performance, instead, we present the maximum power output at particular illuminance levels.

Energetic Measurements: Kelvin probe (KP), ambient photoemission spectroscopy (APS), and SPV measurements were taken using an APS04 Air Photoemission system (APS04, by KP Technology) using a 2 mm gold tip under atmospheric conditions. All samples were measured on PET/IMI substrates to ensure proper grounding of the

thin films. Measurements were taken at multiple positions on the films to ensure reproducibility. Kelvin probe measurements are taken until a constant value was achieved. The result was a relative contact potential difference between the sample and tip which was converted to an absolute value by addition of the tip WF which was calculated using a silver reference sample. The APS data was processed using the protocol described by Baikie et al.^[56] This involves taking the cube root of the measured photoemission, fitting the resultant linear region, and extrapolating to zero photoemission to find the highest occupied molecular orbital (HOMO) level of the semiconductor. LUMO levels are estimated from these APS derived HOMO energies by addition of the optical bandgap derived from the absorption onset. SPV measurements were taken after a stable contact potential difference was recorded and, unless otherwise stated, with a white light 150 W DC regulated quartz tungsten halogen bulb (spectrum in Figure S17, Supporting Information) with an approximate intensity of 0.2 Suns (200 W m^{−2}).

Absorption: A Shimadzu UV-2550 UV-visible spectrophotometer was used to measure steady-state transmittance of substrates and samples. Absorbance was then calculated using the following equation to remove substrate contributions: $\text{Abs} = \log(T_{\text{Substrate}}/T_{\text{Sample}})$, with reflection and scattering effects being ignored.

Contact Angle Measurements: Contact angle measurements were carried using a Krüss DSA100 Goniometer. Water was used, with four drops on each sample being measured and fitted with the same protocol to ensure reproducibility.

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

interfacial energetics, light-soaking, low-light, organic solar cells, scalable

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