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Highly doped layers as efficient electron–hole recombination contacts for tandem organic solar cells

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A key feature of stacked organic solar cells is an efficient recombination contact at the interface between the solar cells in the stack. Here, an electron current has to be converted into a hole current without loss of energy. Furthermore, the recombination contact has to be highly transparent. We present a new approach for small molecule organic solar cells using highly doped organic layers. Our approach adapts the use of tunnel diodes known from inorganic tandem solar cells. We compare a metal cluster based recombination contact reported in literature to the new approach using different organic tandem solar cell structures. For this purpose, current-voltage characteristics of adequate solar cells are measured. The experiments show that highly doped layers as recombination contacts in tandem organic solar cells are superior to the metal cluster based approach. The proposed concept allows an addition of the open circuit voltages of the subcells of a tandem solar cell, without absorption or reflection at the recombination contact. The results further show that our concept does not depend on the specific choice of materials as it is seen for metal cluster based recombination contacts. It therefore represents a general approach which is compatible to mass manufacturing.
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I. INTRODUCTION

Organic photovoltaics are potentially an important technology for future electricity generation because of low material consumption and the comparatively easy and cheap production process. The reason for the low material consumption is the high absorption coefficient of organic dyes, allowing high absorbances even for very thin layers of a few tens of nanometers. However, in most materials this absorbance is present only in narrow bands, limiting the overall power conversion efficiency of organic solar cells. One possibility to overcome this problem is to stack solar cells consisting of different materials absorbing in ideally nonoverlapping areas of the solar spectrum on top of each other. Theoretical calculations of the efficiency of such tandem solar cells result in values of about 16% to 23%,[1,2] in contrast to 10% to 15% for organic single solar cells.[1–3] The tandem solar cell concept has been realized for oligomer[4–6] as well as for polymer solar cells.[7–9]

A challenge of this concept is the need of an efficient recombination contact for electrons and holes in between the subcells of a tandem solar cell (see Fig. 1).

There are three main requirements for such a recombination contact: First, it has to ensure that the electrons from the first and the holes from the second subcell meet at the same energy level. Therefore a splitting of the electron and hole quasi-Fermi levels has to be avoided. If this condition is not fulfilled and a reverse voltage is produced at the recombination contact, the open circuit voltages (V_{oc}^{s}) of the subcells cannot sum up in the tandem solar cell (see Fig. 2). Second, the recombination contact has to be highly transparent to avoid absorption and reflection reducing the power conversion efficiency and disturbing the current matching in the tandem solar cell. Third, it should be compatible to future mass production processes.

A possible approach is the use of metal layers. If these are sufficiently thick, a closed metal layer is formed which acts as an Ohmic contact. This approach has been preferred in polymer tandem solar cells because the closed metal layer prevents the underlying layer from dissolving during spin coating of the second solar cell. Typically, the metal thicknesses used are in the range of 10–15 nm.[7,9] The disadvantage of this approach is the high absorbance and reflectance of the metal layer, which leads to losses and unbalanced absorption in the subcells. Recent work showed that efficient recombination contacts in polymer photovoltaics can be achieved with TiO_x (Ref. 10) or ZnO (Ref. 8) as transparent conducting interlayer. Nevertheless, these approaches still

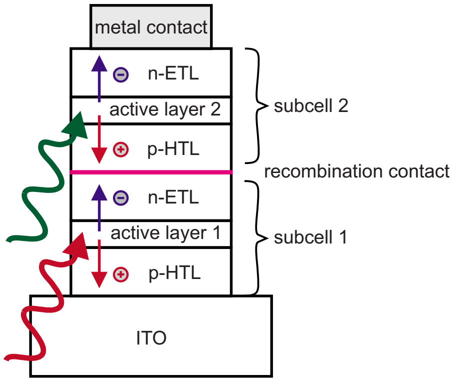

FIG. 1. (Color online) Basic principle of an organic tandem solar cell using the p-i-n-concept (Ref. 5). The arrows indicate the hole currents and the electron currents. ETL denotes the electron transport layers and HTL the hole transport layers. ITO is the conductive transparent indium tin oxide bottom contact.

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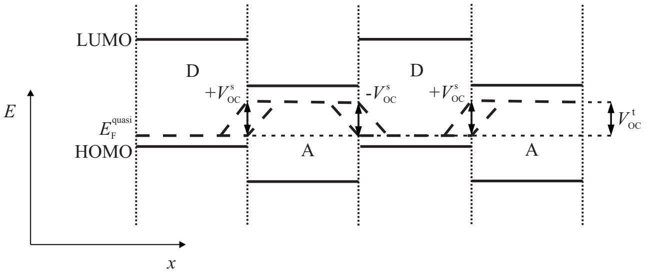

FIG. 2. Schematic energy level diagram of a double heterojunction solar cell without recombination contact under illumination at open circuit. D denotes the donor material and A the acceptor material. Each heterojunction produces a voltage V^{s}_{oc} which results from the quasi-Fermi level splitting at the heterojunction. The reverse heterojunction in the center generates a voltage $-V^{s}_{oc}$. The overall voltage of the tandem solar cell is V^{t}_{oc}.

suffer from a small loss in the open circuit voltage of the tandem solar cells (V^{t}_{oc}) of at least 3% compared to the sum of the single solar cell voltages.

In small molecule based organic solar cells, mostly very thin metal layers have been used as recombination contacts. Their thickness of 0.5 to 2 nm does not lead to the formation of closed metal layers but to metal clusters.[6,11] The reflectance is reduced compared to closed layers. However, due to Mie-resonances there are still absorption and reflectance peaks,[12,13] reducing the power conversion efficiency of these solar cells. The working principle of a recombination contact with metal clusters is associated with their ability to act as recombination centers. If the associated recombination rate is higher than the generation due to illumination, the quasi-Fermi level splitting is removed. As this splitting equals the V_{oc} of the according heterojunction, with metal clusters no reverse voltage at the recombination contact is expected. This situation is depicted in Fig. 3. However, a strong dependence of the used materials, especially the metal, on the efficiency of the recombination contact was shown in Refs. 14 and 15. Possible explanations are the influence of interface dipoles occurring between metal and organics resulting in a built-in voltage of the interface[16] or doping effects of the metal.[17]

As a result of that, up to now, only three material combinations in small molecule based organic tandem solar cells have been reported that show a full addition of the V_{oc}: Ag clusters as recombination contact in a double heterojunction of PTCBI/CuPc (Ref. 6) and Au clusters as recombination contact in a tandem solar cell consisting of P3HT:PCBM and ZnPc:C_{60} bulk heterojunctions.[18] Furthermore, by supporting the metal cluster based recombination contact with doped layers, a doubling of V_{oc} in a double bulk heterojunction of ZnPc:C_{60} was observed.[19] All other published cell configurations suffer from at least some loss of V_{oc} in the tandem solar cell.[4,20,14,15]

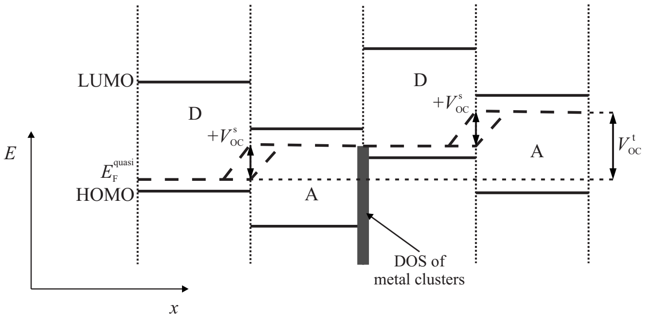

FIG. 3. Schematic energy level diagram of a double heterojunction solar cell with a metal cluster based recombination contact under illumination at open circuit. D denotes the donor and A the acceptor material.

Accordingly it would be advantageous to have a concept of recombination contacts that is independent of the used materials and a specific deposition procedure. This concept should exclude metal layers or metal cluster layers, considering a possible future roll-to-roll production process of organic solar cells where metal interlayers would lead to an interruption of the organic evaporation process.

In this paper, we present tandem organic solar cells with highly electrically doped layers as recombination contacts, which avoid the use of metal cluster interlayers. In this concept, we adapt the approach of tunnel diodes used in inorganic tandem solar cells.[21] We use different material combinations for the heterojunction to show the independence of this concept on the materials used.

II. EXPERIMENTAL

Before use, all organic materials were purified at least twice by vacuum gradient sublimation, except for F4-TCNQ which was sublimated once. N,N,N,N-tetrakis4-methoxyphenyl-benzidine (MeO-TPD) (Sensient, Wolfen, Germany) [highest occupied molecular orbital (HOMO): −5.1 eV] and N,N′-di(naphthalen-l-yl)-N,N′-diphenyl-benzidine (α-NPD) (Sensient, Wolfen, Germany) (HOMO: −5.4 eV) serve as matrix materials for the hole transport, C_{60} (Kurchatov Institute, Moscow, Russia) [lowest unoccupied molecular orbital (LUMO): −4.0 eV] is used as active material and matrix for electron transport. As p-dopant, 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethan (F_4-TCNQ) (TCI Europe, Zwijndrecht, Belgium) (LUMO: −5.2 eV) and the comparable NDP2 (Novaled AG, Dresden, Germany) are used while for the n-side either acridine orange base (AOB) (Sigma-Aldrich, St. Louis, USA) or the comparable NDN1 (Novaled AG, Dresden, Germany) is chosen. The use of NDP2 and NDN1 lead to comparable device performances as the use of the commonly used dopants,[22,23] but alleviates materials handling and device stability. All devices are prepared on indium tin oxide-coated glass substrates (Thin Film Devices Inc., Anaheim, USA) with a sheet resistance of 30 Ω/sq which were precleaned with organic solvents in an ultrasonic bath. Thermal evaporation is performed at $p < 10^{-6}$ mbar and deposition rates of ~0.5 Å/s in one of two different UHV systems: A multichamber UHV system is used in which the intrinsic layers, all doped layers, as well as the metal layers are deposited in separate chambers without breaking the vacuum. Alternatively, the layers are deposited successively in a single large chamber (Kurt J. LESKER Co. Ltd., Hastings, U.K.) where large wafers containig several solar cell substrates can be coated by vacuum deposition simultaniously. That way, it is possible to fabricate several devices with different structures on the same substrate, ensuring the

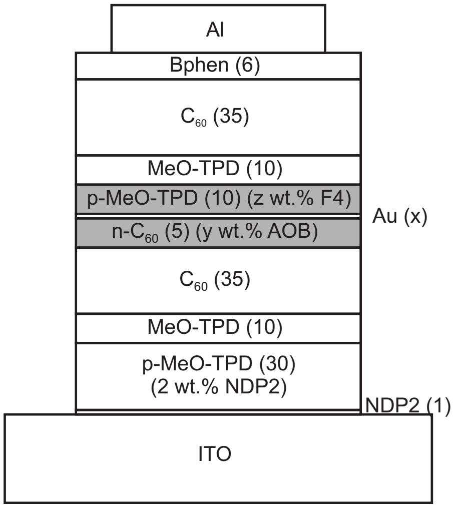

FIG. 4. Structure of the MeO-TPD/C_{60} tandem solar cell. Solar cells with the following variations in this structure are prepared: without the doped layers displayed in gray and x=0 (device Al); without the doped layers displayed in gray and x=1 (device A2); x=0, y=5, z=20 (device A3) and x=0, y=8, z=35 (device A4). All values are given in nanometer except for the doping concentrations.

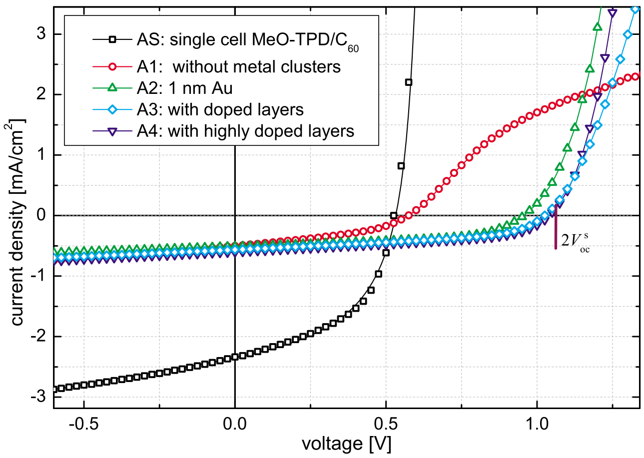

FIG. 5. (Color online) Current density-voltage characteristics of tandem solar cells with two MeO-TPD/C_{60} heterojunctions connected by different recombination contacts.

same preparation conditions, i.e., comparability of the solar cells under consideration. Doping is realized by coevaporation of matrix and dopant material from different crucibles while the deposition rates are controlled individually by quartz oscillators. The resulting solar cell devices have an average active area of 6.44 mm^2. After processing, all devices are transfered from vacuum directly into an inert gas atmosphere and are encapsulated there with epoxy sealed glass covers. The j-V characteristics of the encapsulated samples are measured in air shortly after sample preparation and are recorded under a simulated air mass (AM) 1.5 illumination provided by either a sun simulator SOL 1200 (Dr. Hönle AG, Gräfelfing, Germany) or a sun simulator SC 1200 (K.H. Steuernagel Lichttechnik GmbH, Möhrfelden-Walldorf, Germany). While the current is detected with a Keithley Source Measure Unit, the intensity at the sample position is registered with the help of a silicon reference cell (Fraunhofer Institut für Solare Energiesysteme, Freiburg, Germany).

A possible mismatch between the simulated and the AM 1.5 spectrum or between the spectral responses of the reference cell and the test device is not accounted for.

III. RESULTS AND DISCUSSION

A. MeO-TPD/C_{60}-double heterojunction solar cells

For the proof of principle of the proposed recombination contact concept, a simple tandem solar cell structure consisting of two identical planar heterojunctions is chosen (see Fig. 4). In this structure, the layers in between the two subcells forming the recombination contact are varied. Different doping concentrations and furthermore a 1 nm Au interlayer are compared. A single solar cell with only one MeO-TPD/C_{60} heterojunction (device AS) is prepared under exactly the same experimental conditions as the tandem solar cells to compare the characteristic values, in particular the V_{oc}, with the tandem solar cell structure. The V_{oc}, the j_{sc}, and the fill factor (FF) of the single solar cell are measured to be 0.53 V, 2.3 mA/cm^2, and 50%, respectively. The tandem solar cells in this study are not optimized in terms of the optical field distribution so that a lower short circuit current j_{sc} for the tandem solar cells is expected.

The j-V characteristics (Fig. 5) show that in case of an undoped recombination contact without metal interlayer (device Al), the forward current is limited by the backward current of the reverse heterojunction formed at the recombination contact which results in a S-shaped j-V curve. The voltage of the tandem solar cell is therefore reduced by the V_{oc} of this heterojunction. The S-shape of the current-voltage (IV)-curve only results from this backward solar cell. By inserting a 1 nm Au layer as recombination contact (device A2) V^t_{oc} increases to 0.96 V which is a 9% loss compared to $2V^s_{oc}$. The FF slightly increases to 53%. By using a 5 wt % AOB-doped C_{60} layer and a 20 wt % F_4-TCNQ-doped MeO-TPD layer without metal interlayer (device A3) V^t_{oc} increases to 1.02 V which corresponds to a loss of V^t_{oc} of 4% compared to $2V^s_{oc}$. The FF of 48% is slightly decreased compared to the single solar cell.

With higher doping concentrations of 8 wt% AOB in C_{60} and 35 wt% F_4-TCNQ in MeO-TPD (device A4), a further increase in V^t_{oc} is observed which results in a V^t_{oc} only decrased by 1.9% compared to $2V^s_{oc}$. The FF of 49% is nearly the same as in the single solar cell. From the higher forward current of the corresponding j-V characteristics, one can conclude that the further increase in V^t_{oc} is mainly caused by a lower series resistance of the whole device for increasing doping concentration. In contrast, the lower V^t_{oc} of device A2 cannot be explained by a lower forward current because it is as high as in device A4. This implies different mechanisms for the reduction in V_{oc} in the device with Au as recombination contact (A2) and the devices with highly doped layers as recombination contact (A3 and A4). The characteristic values of the prepared solar cells are displayed in Table I.

B. α-NPD/C_{60}-double heterojunction solar cells

By using acceptor materials with a lower lying HOMO, it is possible to increase the open circuit voltage of organic

TABLE I. Characteristic values of the prepared solar cells. The value "loss of V_{oc}" is defined as $1-(V^{t}_{oc}/2V^{s}_{oc})$.

Cell No.	Recombination contact	V_{oc} (V)	Loss of V_{oc} (%)	J_{sc} (mA/cm^2)	FF (%)
AS	⋯	0.53	⋯	2.34	50
Al	None	0.57	46.2	0.50	39
A2	Au clusters	0.96	9.4	0.51	53
A3	Doped layers	1.02	3.8	0.56	48
A4	Highly doped layers	1.04	1.9	0.60	49
BS	⋯	0.85	⋯	2.39	48
Bl	None	1.07	37	1.32	51
B2	Au clusters	1.35	20	1.13	41
B3	Ag clusters	1.07	37	0.89	41
B4	Highly doped layers	1.7	0	1.36	48

solar cells which also allows higher power conversion efficiencies.[24] Therefore, our approach of highly doped layers as recombination contact has also been tested with a double heterojunction of C_{60} and α-NPD, which has its HOMO at −5.4 eV compared to −5.1 eV of MeO-TPD.

Again, tandem solar cells with metal clusters and with highly doped layers as recombination contact are prepared. Because α-NPD can be doped more efficiently and stable with the proprietary acceptor NDP2 instead of F_4-TCNQ, we use this alternative p-dopant. We also use the proprietary donor dopant NDN1 for C_{60}.

Again, a single solar cell with only one α-NPD/C_{60} heterojunction (device BS) is prepared under exactly the same experimental conditions as the tandem cells to compare the characteristic values, especially the V_{oc}. Because of the low lying HOMO of α-NPD, a V^{s}_{oc} of 0.85 V is observed for such a single heterojunction. The j_{sc} is measured to be 2.39 mA/cm^2.

The structure of the tandem cells with metal clusters as recombination contacts are shown in Fig. 6(a) and that with highly doped layers in Fig. 6(b). The corresponding j-V characteristics are shown in Fig. 7. Again, the solar cell without metal clusters (device Bl) show that the recombination contact works as a solar cell in backward direction, as it was also observed with MeO-TPD/C_{60}-heterojunctions. Because the forward current is limited by the backward current of this reverse solar cell the j-V curve is S-shaped. With Au as well as Ag clusters, an addition of the V_{oc} is not achieved. With Au clusters, a strongly S-shaped IV characteristic is observed, resulting in a V_{oc} of only 1.35 V and a FF of 42% (device B2). With Ag clusters, no S-shape in the j-V characteristic occurs but the V_{oc} of 1.07 V is even lower and the FF is 41% (device B3). The S-Shape of device B2 is a hint for a strong interface dipole between gold and organics resulting in a barrier for the charge carriers. Using highly doped layers, a complete addition of the V^{s}_{oc} to 1.7 V is demonstrated (device B4). The FF was measured to be 48% and is thus as high as in the single cell structure. The forward current is also as high as in the single cell which implies that there is no additional series resistance due the insertion of highly doped layers. The characteristic values of the prepared solar cells are displayed in Table I.

C. Discussion

A strong material dependence of the recombination contact efficiency when different metals are combined with various organic materials is observed. With α-NPD, it is not

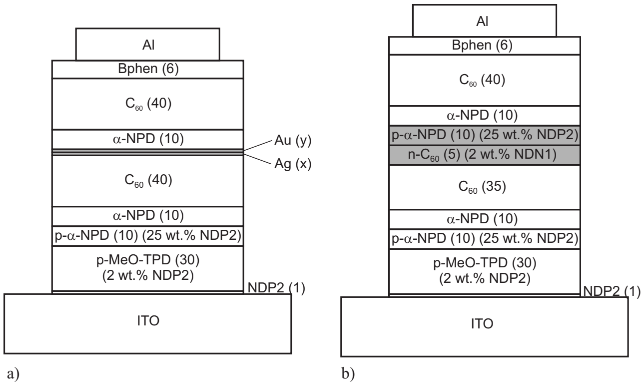

FIG. 6. Structure of the α-NPD/C_{60} tandem solar cells with (a) metal clusters and (b) highly doped layers as recombination contact. Solar cells with the following variations in structure (a) were prepared: x=0, y=0 (device Bl); x=0, y=0.5 (device B2) and x=0.5, y=0 (device B3). Structure (b) is denoted device B4 below. The layers forming the recombination contact are displayed in gray. All values are given in nanometer except for the doping concentrations.

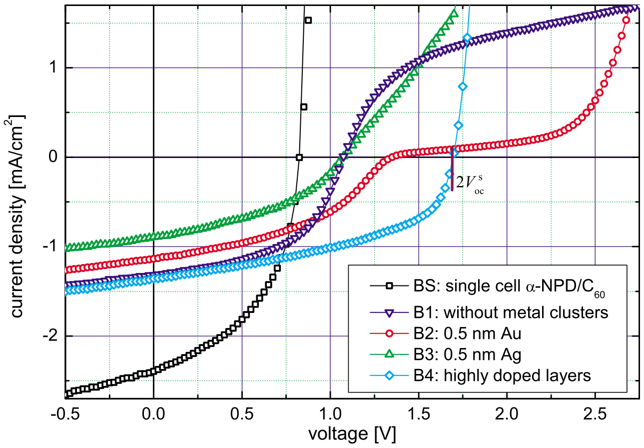

FIG. 7. (Color online) Current density-voltage characteristics of tandem solar cells with two α-NPD/C_{60} heterojunctions connected by different recombination contacts.

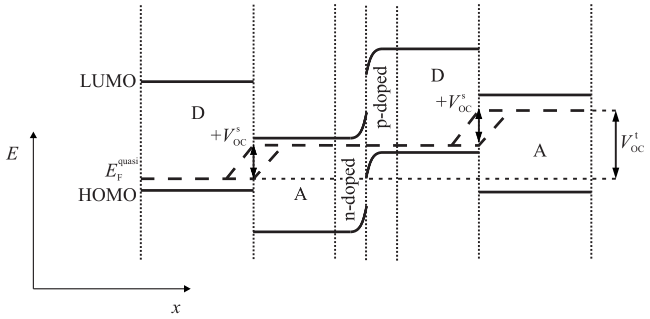

FIG. 8. Schematic energy level diagram at open circuit of a double heterojunction solar cell with highly doped layers as recombination contact. D denotes the donor material and A the acceptor material.

possible to form a working recombination contact neither with Ag nor with Au clusters. Besides, the very different behavior of the recombination contacts with Au and with Ag in α-NPD/C$_{60}$ as well as the difference of Au clusters in Meo-TPD/C$_{60}$ and α-NPD/C$_{60}$ indicate different interaction mechanisms between metal and organics. This is another hint for interface dipoles as reason for the different behavior of different metals in recombination contacts. In contrast, the highly doped layers approach does not show a material dependence.

The working principle of this pn-junction approach observed in this study is caused by the high carrier density introduced by the doping.[25] The Fermi level in the doped layers shifts toward the transport level,[26–28] resulting in a very small HOMO-LUMO offset and narrow space charge layers of only a few nanometers.[29] Second, the high doping concentrations imply high charge carrier concentrations on both sides of the recombination contact. Both are leading to a high recombination rate at the contact which removes the quasi-Fermi level splitting at this heterojunction. As stated above, this is the requirement for an addition of the V_{oc} in tandem solar cells. The schematic energy level diagram of this situation is shown in Fig. 8. A detailed ultraviolet photoelectron spectroscopy/x-ray photoelectron spectroscopy (UPS/XPS) study on p/n-heterojunctions confirms this model and will be published elsewhere.[30]

Besides the materials used in this study, the proposed concept was also applied to other organic materials which proves the versatility of our approach. The results show that the interconnection of the subcells in tandem organic solar cells is no more a problem for small organic molecules.[31] After realizing an efficient recombination contact allowing an addition of the voltages of the subcells, an optimimization of efficiencies should be addressed. Therefore, in a next step absorber materials with complementary absorption spectrum should be used to achieve a gain in efficiency of the tandem solar cell compared to the single cells. Using a highly doped recombination contact and complementary absorbers, recently a small molecule organic solar cell with a certified efficiency of 6.07% was prepared.[32]

Another important aspect that need to be considered, especially for future mass production, is the life time of such recombination contacts. We have not performed aging tests of the prepared solar cells but a systematic aging study of comparable pin-type small molecule organic solar cells showed that solar cells with doped layers are very stable.[33] Structures similar to the recombination contact shown in this work are used as charge generation layers in stacked white organic light emitting diode (OLEDs) which have resulted in lifetimes of 100.000 h at high current densities.[34]

IV. CONCLUSION

We show that highly doped layers as recombination contacts in tandem organic solar cells are superior to the metal cluster based approach. The proposed concept allows an addition of the open circuit voltages of the subcells of a tandem solar cell without absorption or reflection at the recombination contact. The results further show that our concept does not depend on the specific choice of materials. It is thus a general approach which should be compatible to mass manufacturing.

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