



# Laser patterning of vacuum processed small molecular weight organic photovoltaics



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## ABSTRACT

In this paper, we report on laser patterned vacuum processed organic solar cells using an infrared ultra short pulse laser. Laser patterning is performed on all layers, electrodes as well as organic layers, to produce fast, reproducible, and precise scribes. The structuring enables monolithically interconnecting several cells to mini-modules. For patterning the silver top electrode, a plasma process is introduced which lowers the ablation threshold and minimizes protrusion-induced shunt formation. Finally, we demonstrate the up-scaling of organic mini-modules to an area of 64 cm<sup>2</sup> with a geometric fill factor of 94% and a power conversion efficiency on the aperture area of 4.3%.

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## 1. Introduction

In recent years, the commercial maturity of organic photovoltaics (OPV) has improved for both processing technologies: (i) solution processed polymers and solution processed small molecules have reached power conversion efficiencies (PCE) on individual cells above 10%, partly by using tandem structures [1–3]. (ii) For thermally evaporated vacuum processed small molecule multi-junction solar cells a PCE of 13.2% has been reported [4], and first commercial modules of building integrated OPV were recently installed [5]. Although these PCE values are not directly transferable to large area applications, fast roll-to-roll (R2R) processing on flexible and light weight films, semi-transparent solar cells, and promising long-term stability results indicate the potential for both technological approaches [6–20].

R2R processing is typically the method of choice for up-scaling organic devices. Contrary to crystalline silicon solar cells, the monolithic interconnection of single cell stripes to modules can be realized in situ during the coating process as known from conventional thin-film photovoltaics [21–25]. In general, layers can either be

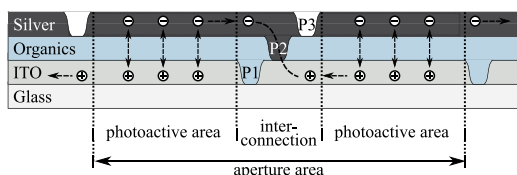
directly patterned during the coating process or be coated homogeneously over the whole area and then selectively be removed. For direct patterning of thermally evaporated OPV in vacuum processes, shadow masks are usually used [26]. Solution processing uses ink-jet printing, slot-die coating, doctor blading, screen printing, or gravure printing as a direct structuring method [11].

For the commercialization of OPV, the PCE related to the module area is of high importance. Like others [27], we define the aperture area  $A_{ap}$  as the area of the photo-active solar cell itself (cell area  $A_{ac}$ ) plus the area of the interconnection zone (see Fig. 1). The latter does not contribute to the power generation and is sometimes called the “dead” area [28]. The aperture area has to be further differentiated from the total, or module area, which specifies the outer limits of a module. We define the geometric fill factor (GFF) as the ratio between  $A_{ac}$  and  $A_{ap}$ . In previous studies about laser structuring on OPV, this factor could be raised above 98% [29].

Modules realized via screen printing or slot-die coating suffer in contrast to laser patterned modules from higher apertures losses. Thus, only GFFs between 40 and 80% have been published [20]. Both values obviously still highly depend on the module layout. One basic advantage of patterning with ultra short pulse lasers is the small and precisely reproducible scribe width (in the range of 20–100 μm – depending on the laser system), leading to a small area loss in the interconnection zone.

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**Fig. 1.** Scheme of the series interconnection of two solar cells to form a solar module. The necessary laser scribes are denominated P1–P3. The organic stack consists of a bulk heterojunction incorporating two absorber materials (donor either  $F_4$ -ZnPc or HDR14, acceptor  $C_{60}$ ) sandwiched between highly doped organic transport layers.

Ultra short pulse laser patterning has a further advantage besides being fast, reliable and scalable: it is already a successfully established patterning technique for various purposes [30–32] and has also been investigated for OPV related technologies such as the production of OLEDs, OTFTs, and single layers [23,24,33–35].

Up to now, the highest reported PCE for laser patterned OPV on PET films was published to be 6.0% for vacuum processed organic solar cells produced on an aperture area of 1171 cm<sup>2</sup> [6]. For solution processes, several contributions on laser structuring from the Brabec group have been published so far [28,29,36–38]. Recently, Lucera et al. demonstrated a PCE of 4.0% on PET on an area of 98 cm<sup>2</sup> [29]. On glass substrates, an efficiency of 5.3% was shown in the same publication on 35 cm<sup>2</sup>. A higher PCE of 5.7% was also already published for tandem modules on PET, but only on a small area of 0.1 cm<sup>2</sup> [36]. Semi-transparent, flexible OPV modules have been demonstrated with an efficiency of 1.2% on an area of 156 cm<sup>2</sup> [39].

This work focuses on laser patterning processes and parameters for a full R2R-compatible vacuum process of small molecule organic solar modules using an ultra short pulse laser. We demonstrate the structuring of the transparent bottom electrode ITO (laser scribe called P1) on glass, of the organic layers containing the photoactive bulk heterojunction and doped transport layers (P2), and of the top electrode silver (P3). The patterning is used to monolithically interconnect cells to modules, which are finally scaled up to a mini-module with six cell stripes, an aperture area of 64 cm<sup>2</sup>, and a PCE on the aperture area of 4.3%.

## 2. Materials and methods

The deposition of the organic layers and the laser patterning was performed in a custom-built in-line vacuum tool with a base pressure of  $10^{-6}$  to  $10^{-7}$  mbar. In this setup, different chambers are used for the deposition of organic layers, metal, for laser patterning, and plasma treatment, respectively. Only one sample is processed at a time. Deposition of the organic layers is done as the sample is moved successively over various linear evaporation sources with typical feed motion speeds between 0.2 m min<sup>-1</sup> and 1.0 m min<sup>-1</sup>, and typical dynamic rates that correspond to stationary evaporation rates between 0.2 nm s<sup>-1</sup> and 1.0 nm s<sup>-1</sup>. The precise value of the feed motion speed and evaporation rate depend mainly on the desired layer thickness and the cost of the material, as the material evaporated while heating up, while controlling mixing ratios before deposition, and when cooling down is not exploited in the used setup. The variation of evaporation rate, however, has not shown a dominant effect on the processed solar cell. Laser patterning is realized stationary with a fast moving beam.

For laser patterning, the commercially available mode-locked solid state laser TruMicro 5050 from Trumpf GmbH + Co. KG was used, that emits laser pulses of 6 ps duration at a wavelength of 1030 nm with a maximum pulse energy of 125  $\mu$ J and a beam quality factor  $M^2$  of 1.2. A ps-laser was chosen instead of a fs-laser

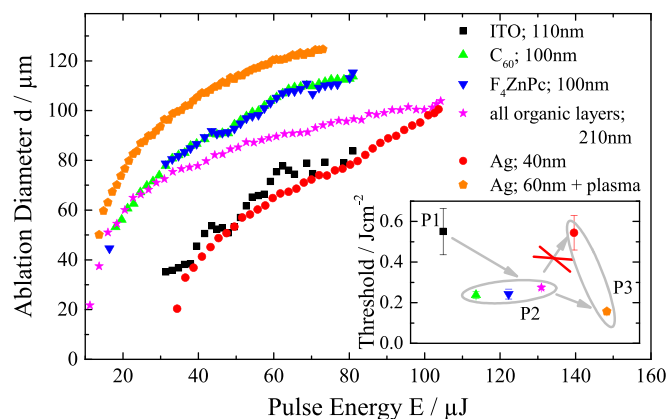
for reasons of stability, technical aspects and economy. The same laser was used to pattern the ITO layer, the organic layers, and the metal electrode. Repetition rates of 50 kHz for ITO and 400 kHz for both metal and organic layers were used. The laser beam is focused via an optic with movable mirrors onto the sample with a beam waist of 97  $\mu$ m (gaussian intensity distribution radius with  $1/e^2$  intensity), allowing for scribe velocities between 0.1 and 8.0 m s<sup>-1</sup>.

All solar cells are built on glass substrates purchased fully covered with a 110 nm ITO layer from LfB, University of Stuttgart. During the processing, the ITO is structured at first (P1), followed by the deposition and patterning of the organic layers (P2), then the silver electrode is deposited and patterned (P3). Fig. 1 shows the schematic cross-section of the layer sequence and the nomenclature of the laser scribes from P1 to P3.

All solar cells contain a bulk heterojunction (BHJ) made of two absorber materials (donor either  $F_4$ -ZnPc [40] or HDR14, acceptor  $C_{60}$ , provided by BASF SE, Heliatek GmbH, MTR Ltd, respectively) sandwiched between doped organic transport layers. P-doped layers used NHT18 as a wide-bandgap transport material doped with NDP9, n-doped layers were realized with  $C_{60}$  doped with NDN45. NHT18, NDP9 and NDN45 were purchased from Novaled GmbH. The precise layer sequences for the presented solar cells and modules are given in the following. Layer thicknesses can be found in parentheses, doping ratios are given in weight-%, and all donor: acceptor blends are produced with D:A volume fractions of 1:1. The layer sequence for cells and modules containing  $F_4$ -ZnPc is: ITO/p-NHT18 (50 nm, 10 wt%)/n- $C_{60}$  (10 nm, 20 wt%)/ $C_{60}$  (10 nm)/ $F_4$ -ZnPc:  $C_{60}$  (50 nm)/p-NHT18 (40 nm, 10 wt%)/Ag (50 nm). Single junctions cells and modules containing HDR14 have the same layer sequence as above, only replacing  $F_4$ -ZnPc:  $C_{60}$  with HDR14:  $C_{60}$  (30 nm). Tandem cells and modules containing HDR14 were built using the following recipe: ITO/p-NHT18 (50 nm, 8 wt%)/n- $C_{60}$  (10 nm, 20 wt%)/ $C_{60}$  (10 nm)/HDR14:  $C_{60}$  (30 nm)/p-NHT18 (120 nm, 8 wt%)/n- $C_{60}$  (10 nm, 20 wt%)/ $C_{60}$  (10 nm)/HDR14:  $C_{60}$  (30 nm)/p-NHT18 (40 nm, 8 wt%)/Ag (50 nm).

The absorber materials are deposited on a pre-heated substrate ( $T \approx 90^\circ \text{C}$ ) to facilitate the formation of a favorable morphology in the blend layer. No post-annealing procedure is performed.

The photoactive area is defined by the overlap of the ITO and the silver electrode. Between ITO and silver, the series interconnection is created by removing the organic layers with the P2 scribe and by preventing the lateral conduction paths in the electrodes using the P1 and P3 scribes (compare Fig. 1). In order to optimize the aperture efficiency of the interconnected cells, it is



**Fig. 2.** Ablation diameters for circles with equivalent area for various layers of an organic solar cell. In the inset, the ablation thresholds for all used layers and material combinations are shown. It can be seen that the organic layers have a lower ablation threshold in comparison to the electrode layer of ITO. The ablation threshold for Ag in comparison to the organic layers is only lower, and thus damage free, when an oxygen plasma treatment is performed.

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