

# Sputtered indium zinc oxide rear electrodes for inverted semitransparent perovskite solar cells without using a protective buffer layer

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## ABSTRACT

In this work, we demonstrate inverted semitransparent perovskite solar cells (PSC) comprising an indium zinc oxide (IZO) layer sputtered at room temperature without a protective buffer layer between the organic electron transport layers (ETL) and the transparent sputtered electrode. The damage done to the ETL layer during sputtering is cured with a short annealing step at a moderate temperature and semitransparent PSCs with fill factors of almost 70% and with power conversion efficiencies (PCE) over 13% are achieved with negligible hysteresis. We investigate the influence of the layer thicknesses of the ETL double layer consisting of PCBM and BCP on the solar cell characteristics as well as the influence of the illumination direction of the bifacial device.

## 1. Introduction

Perovskite solar cells have reached high power conversion efficiencies (PCE) of over 22% [1] in only a few years of research and can be fabricated by low-cost solution-based, large-area deposition techniques. Additional benefits of perovskite solar cells are their inexpensive starting materials, an easily tunable bandgap and their potential usage as topcell in tandem devices in combination with copper indium gallium diselenide (CIGS) or silicon (Si) solar cells to improve the overall PCE while lowering the costs per watt peak. For the application in tandem cells the top cell has to be semitransparent and needs a transparent contact on top of the perovskite instead of an opaque contact made of materials like silver, aluminium or gold. The best semitransparent perovskite solar cells have already reached PCEs of over 16% in standard [2,3] as well as in inverted architecture [4,5] very recently. Materials used as transparent conductive electrodes are mainly indium tin oxide (ITO) [3,6–9], aluminium doped zinc oxide (ZAO) [4,10], hydrogen doped indium oxide (In<sub>2</sub>O<sub>3</sub>:H) [11] (in combination with ITO [2,12]), silver nanowires (Ag-NWs) [13,14] and thin metal layers [5,15,16]. Several research groups have already shown highly efficient tandem cells in two-terminal [2,7,12] as well as in four-terminal configuration [3–5,8–11,14,17] and with standard or inverse stacked perovskite solar cells.

In this work, we use sputtered indium zinc oxide In<sub>2</sub>O<sub>3</sub>:ZnO (IZO) as a transparent conductive oxide (TCO) with a wide range of high transparency up to long wavelengths for the first time in semitransparent inverted perovskite solar cells. It has already been proven

to be an interesting alternative in perovskite cells in standard configuration [17] and in CIGS technology to replace Al-doped zinc oxide [18,19], as IZO shows higher carrier mobility up to 50 cm<sup>2</sup>/Vs and improved moisture stability. Furthermore, compared to other sputtered TCOs, IZO layers do not need a heating treatment during or after the deposition to achieve good transmittance and sheet resistance values, allowing low-impact deposition on sensitive substrates. In addition, the transmittance at wavelengths larger than 1100 nm is much better than for ITO or ZAO, which might be an important factor, if the semitransparent perovskite cells are applied in a tandem cell in combination with low gap copper indium diselenide (CIS), CIGS or Si solar cells. We present highly efficient semitransparent inverted perovskite solar cells by sputtering the TCO directly on an organic double layer made from bathocuproine (BCP) and [6,6]-Phenyl-C61-butyric acid methyl ester (PCBM) without using a protective inorganic buffer layer like zinc oxide nanoparticles (ZnO-NP) or aluminium doped zinc oxide nanoparticles (AZO-NP) [4,6,7]. A PCBM/BCP double layer was used as electron transport layer (ETL) combination as it allows easily processable stacks with very good electrical properties to guarantee very low hysteresis due to well adjusted interface energy levels. It has been shown for BCP/metal interfaces, that the BCP layer shifts the vacuum level at the interface [20], which is beneficial for the application in perovskite solar cells.

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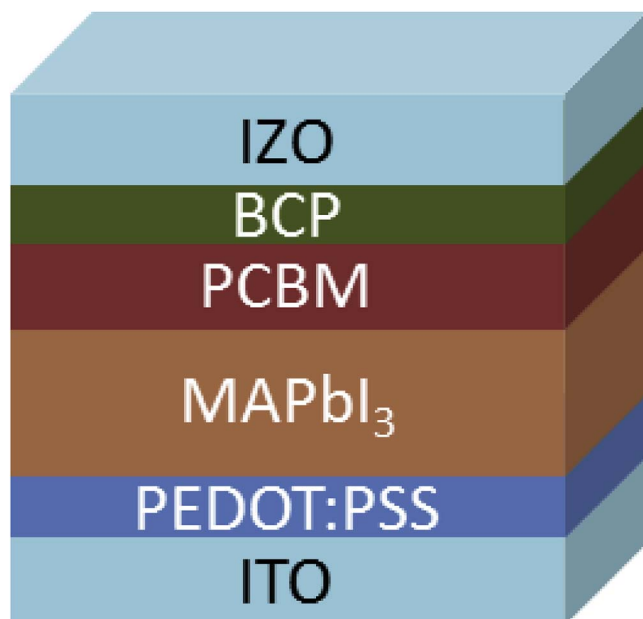


Fig. 1. Structure of semitransparent inverted perovskite solar cells.

## 2. Experimental

### 2.1. Materials

ITO coated soda lime float glass substrates were purchased from Visiontek (Rsq: 12–15  $\Omega$ /sq., thickness of ITO: 150 nm), poly-3,4-ethylenedioxythiophen (PEDOT:PSS, Clevis P VP AI 4083) was purchased from Heraeus, methylammonium iodide (MAI) was purchased from Dyesol, lead acetate trihydrate ( $\text{PbAc}_2 \cdot 3 \text{H}_2\text{O}$ , 99.999%), bathocuproine (BCP, sublimed grade, 99.99% trace metals basis), N,N-Dimethylformamide (DMF, anhydrous, 99.8%) and 1,2-dichlorobenzene (DCB, anhydrous, 99%) were purchased from Sigma-Aldrich. [6,6]-Phenyl-C61-butyric acid methyl ester (PCBM, 99.0%) was purchased from Solenne BV. All materials were used as received.

### 2.2. Device preparation

The structure of the devices is ITO/PEDOT:PSS/perovskite/PCBM/BCP/IZO as shown in Fig. 1.

Laser patterned ITO coated glass substrates ( $15 \times 15 \text{ mm}^2$ , thickness 1.1 mm) were plasma etched with argon before the deposition of PEDOT:PSS, which is prepared in a flowbox by spin-coating. Afterwards, the coated substrates were annealed at 120  $^\circ\text{C}$  for 10 min on a hot plate. For the following spin-coating steps the substrates were transferred to a nitrogen filled glovebox. All materials used for the preparation of the absorber were stored in a nitrogen filled glovebox and all the solutions were prepared there as well. A 40 wt% solution was made by dissolving methylammonium iodide and lead acetate (3:1 molar ratio) in anhydrous N,N-dimethylformamide (DMF). It was stirred on a hot plate (60  $^\circ\text{C}$ ) for about two hours before spin coating. The perovskite solution is spun at 6000 rpm for 22 s (with an additional ramp time of 8 s to reach 6000 rpm) and the substrates were left on the spin coater for 1 min before the solvent annealing (SVA) step was done (5 min at 100  $^\circ\text{C}$  with 10  $\mu\text{l}$  DMF under a petri dish). For the reference samples a 20 mg/ml PCBM solution in 1,2-dichlorobenzene (DCB) was spun at 1000 rpm for 30 s, which is followed by the spin coating of the BCP layer [21], unless otherwise noted: 1 mg/ml in ethanol, 3000 rpm 30 s). The samples were then taken out of the glovebox for sputtering the IZO layer, which was deposited in a Leybold Z600D inline system by DC magnetron sputtering from a high purity (99.95%)  $\text{In}_2\text{O}_3$ :ZnO target with a composition of 90 wt%  $\text{In}_2\text{O}_3$  (84 at% In to Zn). All the

sputtering processes were performed at room temperature (no additional heating) with the following parameters: a power density of 2.8  $\text{W}/\text{cm}^2$ , transport speed of 12.7  $\text{cm}/\text{min}$ , total pressure of 10  $\mu\text{bar}$  and 1.8 vol%  $\text{O}_2$  content in the Ar sputter gas, which corresponds to an 170 nm thick layer with a sheet resistance of 22  $\Omega/\text{sq.}$  and an average transmittance (300–800 nm) of 71%. The active cell area was typically 0.135  $\text{cm}^2$ .

### 2.3. Instrumentation

Current voltage analysis was carried out in ambient air at 25  $^\circ\text{C}$  with a WACOM solar simulator (class AAA, AM 1.5, 1000  $\text{W}/\text{cm}^2$ ) and a Keithley 2400 source measure unit at 270  $\text{mV}/\text{s}$  and 20 ms delay time. There was no light-soaking or biasing done before the IV characteristics were recorded. The scanning electron microscope (SEM) images were taken with a FEI Sirion XL30 SFEG at a voltage of 5 kV. Transmittance measurements were done with a Perkin Elmer Lambda 900 spectrophotometer. External quantum efficiency (EQE) measurements were performed without light- or voltage-bias using an OptoSolar SR 300 setup. The X-ray diffraction (XRD) data were collected with a PANalytical Empyrean in Bragg-Brentano setup.

## 3. Results and discussion

As a consequence of inevitable damage caused by sputtering the IZO layer directly onto the organic layers (BCP/PCBM), all ‘as grown’ measured perovskite cells show an s-shaped IV curve as a result of an electron extraction barrier and charge accumulation at the sputter damaged electrode’s interface with the ETL, which leads to an unbalanced carrier extraction [17,22]. Therefore we applied a short annealing step at a moderate temperature (20 s at 90  $^\circ\text{C}$  on a hotplate) to cure the damage done by the sputtering process and to achieve IV characteristics without an s-shape (Fig. 2). Due to soft sputter conditions optimised for deposition on organic layers, such short annealing durations are already sufficient. Hence, the main damage appears to be related to a disordered interface manifested e.g. by a deranged order of the molecules on the organic layer’s surface. A similar behaviour is observed, when Al is sputtered on BCP/PCBM [23] or when an electrode is sputtered onto organic absorber layers [24–26]. In addition, the annealing step might help to passivate the shallow trap states deeper in the perovskite film (for example grain boundaries) by stimulating the diffusion of PCBM into the perovskite layer [27].

All results shown in this work are based on the characteristics after this annealing step, unless otherwise noted. In all our cells the

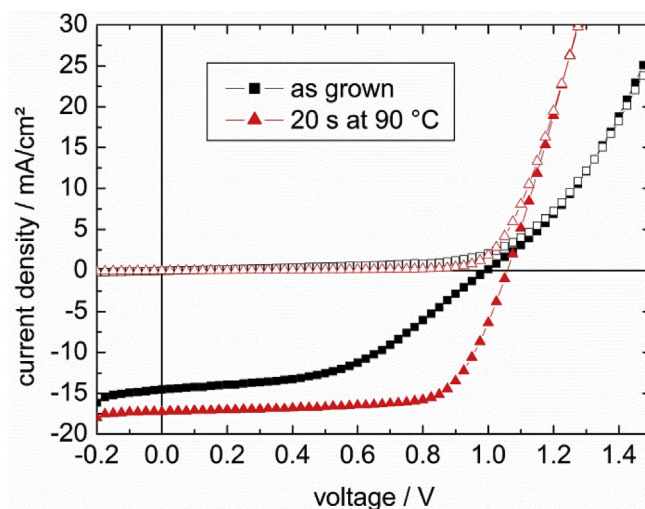


Fig. 2. IV characteristics before (“as grown”) and after a short annealing treatment (20 s at 90  $^\circ\text{C}$ ) of the complete cell stack measured in ‘up’-direction.

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