

Self-contained optical enhancement film for printed photovoltaics



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ARTICLE INFO

Keywords:

Light management
Printed photovoltaics
Absorption enhancement
Light trapping
Organic solar cells
Embedded nanostructure

ABSTRACT

Printed photovoltaics promise lightweight and flexible light harvesting devices for conformal integration into buildings, portable electronics or vehicles. This is enabled by employing thin photoactive layers, which can reduce the use of sometimes costly and scarce absorber materials. Since this in turn comes along with an incomplete light absorption and hence restrains the power conversion efficiency of printed photovoltaics, their development was since long accompanied by integrated light management. However, besides a mere efficiency enhancement, new strategies should consider both functionality and additional costs of the light management. Optical enhancement strategies should consequently avoid complications with the delicate printing of the photovoltaic layers and therefore structures on the light incident surface of the device have been proposed. However, these air-faced approaches are prone to the impacts and stress of operating conditions and hence a protected solution is desired. Here we introduce a carefully designed photonic nanostructure embedded in a self-contained transparent film, which offers a conformal device attachment. An efficiency enhancement of 11% is demonstrated on an organic photovoltaic device. Angle dependent measurements moreover suggest a yearly increase of 13%, exploiting the seasonal asymmetry of the incident solar power by a built-in asymmetric diffraction.

1. Introduction

Certified record efficiencies for printable photovoltaic technologies are ranging between 22–23% for CIGS, CdTe and perovskite solar cells [1–3] and between 10–13% for CZTS, dye-sensitized and organic solar cells [4–7]. The continuous improvements during the past years were enabled on the one hand through material and process development [8–10] and on the other hand by a wide variety of optical enhancement schemes [11–13]. Existing solutions have thereby often been introduced as scattering or diffractive structures in the vicinity of or even directly in the active layer, in order to obtain coupling into guided modes [14–16]. If metallic layers are involved, surface plasmon polaritons can additionally be excited which in turn can give rise to strong confined fields in the absorber layer [17–19]. However, light management strategies should ideally neither compromise the electronic properties of the solar cell [20,21] nor add costs or complications to the large-area fabrication of the photovoltaic devices [12,14,22]. Whereas the implementation of structures inside the functional photovoltaic layers could hinder appropriate morphology and phase formation, increase interface recombination or introduce additional parasitic absorption in the device, light management structures located on the surface provide an excellent alternative solution. Absorption enhancements through light redirection and antireflection strategies

have been reported [23–32], which successfully avoid manipulation of the printed functional layers. However, these structures have their functional optical interface exposed to air and hence cannot sustain typically outdoor conditions of printed photovoltaic operation without losses in their optical properties. Up to now, only a single approach provided a protected surface structure that is designed with an unstructured air interface, but with efficiency enhancement only below 7% [33].

Here we propose a self-standing and non-absorbent light management film with an unstructured sunward surface, which can be independently attached to the substrate of thin film photovoltaics. The film consists of a carefully designed high refractive index periodic nanostructure of large aspect-ratio, which is embedded in a polymer matrix to protect it from external influences and contamination. By introducing an optimized asymmetric shape, a large portion of the light is diffracted into the first order [34] and is further trapped in the device. The structure generates a 11% enhancement in the power conversion efficiency (PCE) of an organic photovoltaic device printed in ambient atmosphere.

2. Optical simulations

The commercial software SETFOS (Fluxim AG, Switzerland) was

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<http://dx.doi.org/10.1016/j.solmat.2017.01.015>

Received 31 October 2016; Received in revised form 23 December 2016; Accepted 10 January 2017

Available online 17 January 2017

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used to calculate the optical properties of the solar cell. The absorption in the active layer is determined dependent on the propagation angle in the substrate θ_S , which is the angle accessible through light redirection structures. Secondly, for each geometry of the ZnS nanostructure (see Fig. S1), the diffraction efficiencies η_m and the resulting propagation angles up to the fourth order $m = \pm 4$ were calculated for both polarizations using rigorous coupled wave analysis (RCWA) [35]. Through the initial diffraction event, a set of possible light propagation angles $\theta_S(m)$ is defined for every wavelength λ . The above calculation is hence repeated for incidence angles $\theta_S(m)$ for both directions of incidence (top and bottom) and the results are stored in bidirectional scattering distribution functions BSDF(λ) with dimension equal to the number of propagating diffraction orders. Once the coherent optics of the solar cell stack and the nanostructure layer are calculated, they can be treated as interfaces [36] with angle dependent reflection, transmission or absorption coefficients. As described in [37,38] the BSDF and the results from SETFOS are then used to calculate the optics of the incoherently coupled system, connected only through transparent thick layers (glass/polymer). From the resulting absorption in the active layer, the current density can be evaluated for various incident angles.

3. Experimental

3.1. Device fabrication

In order to be compatible with large-area production, interference lithography [39] is used to create the designed nanostructure with period Λ from which a nickel master is created. This template further enables cost-effective and large-area continuous nanoimprint lithography [40,41] through either hot embossing into flexible substrates or UV-lithography of a sol-gel [42,14]. Fig. 1(a)–(d) show how the latter is used here to imprint an UV-curable polymer (micro resist technology, OrmoStamp), which is dispensed over the full light incident side of the solar cell substrate. Once it is hardened on the area of 25 cm² by UV light and successively baked it exhibits a high mechanical (hardness of 36 MPa, Young's modulus of 0.65 GPa) and thermal (up to 270 °C) stability and is further highly transparent for UV and visible light [43,44]. Tilted evaporation (45°) of the transparent and high index-of-refraction material zinc sulfide (ZnS) is used to obtain a periodically modulated asymmetric optical interface through self-shadowing of the previously molded pattern [34]. Once the structure is embedded with another layer of the UV-polymer it forms a self-contained diffractive film, which is attached to the photovoltaic substrate. A SEM image of the final nanostructure is shown in Fig. 1(f) (Fig. S1) and reveals the ZnS coated side faces. Polymer solutions with standard architecture of ITO(150 nm)/ Clevis P-VP AI4083(30 nm)/ Merck Lisicon PV-D4610:PC₆₀BM [1:2](130 nm)/ Ca(30 nm)/ Al(100 nm) were doctor-bladed in ambient atmosphere (Fig. 1) as described previously [45]. The standard device architecture and small device area is used to test

the light management on an increased number of reference devices. Since the light management film is decoupled from the device and independent on its fabrication, no tests on durability or up-scaling of the organic solar cells are performed and can be found elsewhere [46–49].

3.2. Sample characterization

J-V curves were recorded under illumination of 1 sun, provided by the xenon arc lamp of a solar simulator (sciencetech SS2.5 kW) equipped with an AM1.5 filter and a collimation of $\pm 3^\circ$. A power density of 100 mW cm⁻² was ensured using a calibrated Si-photodiode (VLSI Standards). The samples were masked with an aperture of $A=0.04$ cm², mounted in a N₂-filled housing and illuminated through a quartz window. A Keithley 2400 source meter was used to ramp the voltage from -1.3 to 1.3 V in steps of 0.01 V with a dwell time of 60 s to avoid heating of the device. The obtained current densities were multiplied with a correction factor accounting for the spectral mismatch and the Fresnel reflection at the quartz window ($n \approx 1.5$). Additional data on statistical variations and error analysis can be found in Fig. S2. For angle dependent measurements a constant distance to the light source was ensured for all angles of incidence. External quantum efficiency (EQE) was measured in a N₂-filled glove-box using the light of a xenon lamp passing through a monochromator and guided to the masked ($A=0.04$ cm²) sample through an optical fiber. Currents obtained from integration of the EQE-curves were compared to the values obtained from J-V measurements (see Fig. S3) yielding deviations below 6%.

The diffraction efficiency of the nanostructure was measured with a semi-cylindrical glass lens attached on the backside of the sample to preserve the diffraction angle. Collimated white light was incident on the structure and intensity spectra were recorded behind the sample in steps of 2 degrees.

4. Results and discussion

4.1. Design of the nanostructure

Compared to previously proposed air-faced light management, an embedded diffractive nanostructure is not only protected against environmental influences but provides beneficial additional optical properties. As known from previous reports, an oblique light propagation can enable a more preferred electromagnetic field distribution throughout the photoactive layer and in consequence an increased absorption [50–52]. For a periodic nanostructure this can be achieved by transmitted light of non-zero diffraction orders $T_{\pm 1, \pm 2, \dots}$ [45] which propagate at angles $\theta_S(\lambda)$ in the substrate (see Fig. 2a). Whereas $\theta_S(\lambda)$ is dependent on the period Λ , the amount of light coupled to a diffraction order at a certain wavelength can be controlled by the material and

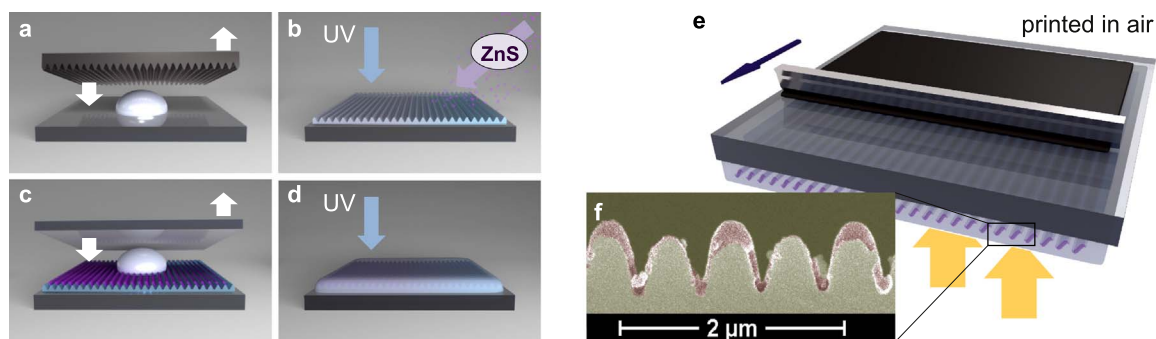


Fig. 1. Fabrication of the light-management layer onto a solar cell substrate. (a) An UV-curable polymer is dispensed onto a large area and molded by a structured master. (b) After UV curing, a high-index zinc sulfide (ZnS) layer is obliquely evaporated onto the hardened structure. Through self-shadowing of the grating pillars an asymmetric layer is formed. (c)–(d) A second layer of the polymer is then applied and cured to embed the ZnS nanostructure. (e) The resulting diffractive film is attached to the substrate for printing of the photovoltaic layers. (f) SEM picture taken of the cross section of the high-index nanostructure layer.

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