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Performance improvement of organic solar cells with moth eye anti-reflection coating

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Abstract

The realization of highly efficient organic solar cells requires the understanding of all optical losses in the solar cell. In this paper, we demonstrate the use of a nano-replicated moth eye anti-reflection coating which functions as an effective medium at the air-substrate interface. We show that the reflection losses of the substrate are compensated, yielding an increase of the peak EQE values by 3.5%. This improvement is valid for a wide range of incidence angles and preserves the insensitivity of organic solar cells against oblique incident light.

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1. Introduction

The performance of bulk-heterojunctions organic solar cells has been steadily increasing since their first realization [1], now reaching efficiencies above 5% [2,3]. One of the possibilities to further increase the performance is to optimize the usage of the incident light. Since organic solar cells consist of a stack of several thin layers, a certain percentage of the light is always lost by reflection at the interfaces and absorption in layers other than the active layer. A possibility to reduce these losses is the usage of anti-reflection coatings.

Moth eyes are a two-dimensional sub-nanometer grating structure that can be found on the cornea of nocturnal insects. Their working principle was first explained by Bernhard [4]. The reflection from the cornea is reduced by employing an effective medium, that is, a gradual change in the index of refraction rather than an abrupt one.

The reflection and transmission properties of moth eye structures can be determined with effective medium theory (EMT), as long as the period of the moth eye is smaller than the wavelength of the incident light. The simplest version of EMT

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for one-dimensional gratings, which can be applied if the periodicity of the grating is negligibly small compared to the wavelength of the incident light, is called 0th order [5]. If this condition is no longer fulfilled, so-called 2nd order EMT developed by Rytov can be used [6].

The first artificial moth eyes fabricated by interference lithography were demonstrated by Clapham and Hutley [7]. Today, they can be produced on a large scale by interference lithography and subsequent replication and are therefore suitable for large-scale industrial production [8]. The fact that they can be produced on a large variety of flexible substrates makes them particularly interesting for organic solar cells. A SEM micrograph of a structure as it was used in our experiments is shown in Fig. 1. Unlike anti-reflection coatings that are based on multiple interference in a dielectric layer stack, their anti-reflecting properties are largely independent of the incidence angle and the wavelength, as long as the latter is larger than the dimension of the moth eye. The former property is of particular interest in the case of organic solar cells, as will be shown in this paper.

2. Experimental details

As the first step, 10 Ω /square indium tin oxide (ITO) coated glass substrates (ITO thickness: 325 nm) were cleaned with

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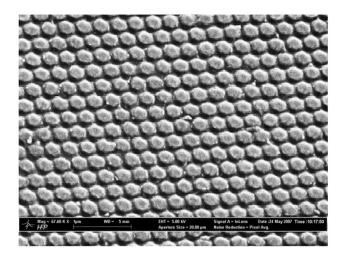


Fig. 1. SEM micrograph of a replicated moth eye. The scale bar represents one micron.

acetone and iso-propanol in an ultrasonic bath. The transparent substrates were then coated with a thin (\approx 20 nm) layer of poly (3,4-ethylenedioxythiophene) doped with poly(styrene sulfonate) (PEDOT:PSS Baytron®, Bayer AG) deposited by doctor blading. On top, a solution of poly(3-hexylthiophene): 1-(3-methoxycarbonyl) propyl-1-phenyl[6,6]C₆₁ (PCBM) was deposited by the same technique. The active layer was bladed from an ortho-xylene (o-Xy) solution yielding a 170 nm thick layer. Finally 100 nm of aluminum was thermally evaporated through a shadow mask, defining the 100 mm² active surface area of the device.

These solar cells were characterized under AM1.5G (100 mW cm⁻²) provided by an Oriel solar simulator. The cells showed a short circuit current density ($J_{\rm sc}$) of 8.6 mA cm⁻², a fill factor (FF) of 0.51, and an open circuit voltage ($V_{\rm oc}$) of 0.57 V, hence an overall efficiency (η) of 2.5%. The device performance of the investigated cells is slightly lower than for state of the art P3HT/PCBM cells. This does not restrain the general validity of our findings, since the application of an anti-reflection coating to a solar cell gives a relative increase of the short circuit current, independent of the starting values. For the spectrally resolved EQE measurements, the OSCs were illuminated by a chopped radiation of a Xe lamp dispersed by a grating monochromator. The current response of the OSCs was pre-amplified and detected by lock-in technique.

The cells were encapsulated and characterized outside the glove-box. The EQE measurements were performed for various angles of incidence, were 0° corresponds to normal incidence. The moth eyes that we received from Autotype Ltd. were replicated in an acrylic lacquer on a PMMA substrate. They were attached to the front of the solar cell with an optical adhesive. Reflection measurements were made with a Perkin-Elmer Lambda 35 spectrometer. All measurements were done for unpolarized light.

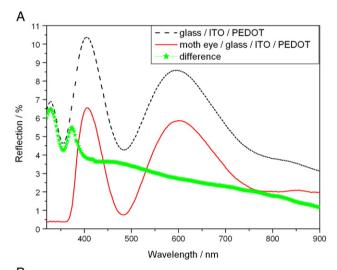
3. Results and discussion

The reflection of a glass/ITO/PEDOT stack and a complete organic solar cell with and without attached moth eye substrate

is shown in Fig. 2A) and B), respectively. The reflection spectra show oscillations caused by interferences in the dielectric layer stack. These oscillations are still present with an additional moth eye, but the overall reflection is clearly reduced due to the lower reflection at the air—glass-interface.

In the case of the complete solar cell, this reduction is strongest for wavelengths between 350 nm and 600 nm, where the P3HT:PCBM is strongly absorbing. For longer wavelengths, most of the reflection is due to light that is not absorbed but reflected at the metal back electrode, so that a reduction of the reflection at the air-substrate interface has a smaller relative effect.

The effect of the moth eye on the performance of the solar cell is shown in Fig. 3, which presents the angle-dependent EQE of the solar cell with a moth eye coating, as well as the EQE of a cell without moth eye at normal incidence as a reference (dashed line). In excellent agreement with the expectations for a highly efficient anti-reflection coating, the peak values of the EQE at 0 ° increase by a relative 3.5%. This



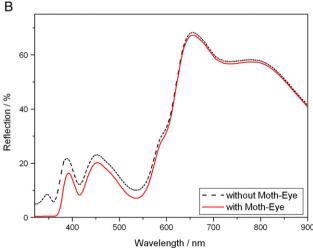


Fig. 2. Measured reflection with (full red line) and without (dashed black line) from a layer stack of glass substrate/ITO/PEDOT (A) and a complete P3HT: PCBM solar cell (B). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

value is close to the total reflection of a PET substrate under normal incidence. The $J_{\rm sc}$ obtained from convolution over the AM 1.5 spectrum from 400 nm to 750 nm is shown in an inset. For wavelengths above 400 nm, an increase of the EQE is visible. At normal incidence, this results in an increase of the calculated $J_{\rm sc}$ of 2.7% from 7.43 mA/cm² to 7.63 mA/cm². The low EQE below 400 nm is not an intrinsic property of the moth eye, but due to absorption in the PMMA substrate that was used in this case.

One of the big advantages of organic solar cells is their low sensitivity if light is incident under an oblique angle [9]. This behavior is explained by an increased light path and thus increased absorption in the active layer. Only at very large angles, the current drops as a consequence of high reflection at the air-substrate interface. In our measurement, this behavior is conserved in the case of an attached moth eye: $J_{\rm sc}$ increases slightly by 2.4% with the incidence angle, reaching its peak value at about 50°, dropping down sharply after that. This is the typical behavior for organic solar cells.

However, the sharp drop of $J_{\rm sc}$ at 60° was not expected, since the moth eye should significantly reduce the reflection at large angles. To clarify the reason for this decrease for large incidence angles, we have repeated the measurement with large-area solar cells so that we were sure that all the light of the incident beam hits the active area of the solar cell.

In addition, we have performed optical simulations of the absorption with the transfer matrix method [10,11]. The effect of the moth eye was accounted for by treating the air-substrate interface separately. For the reflection of the moth eye, we used values that were obtained by an EMT calculation using the Ryatov 2nd order method for one-dimensional gratings. These values can also be used to describe the reflection of two-dimensional structures as used in our experiment with sufficient accuracy.

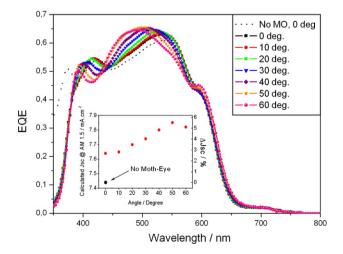


Fig. 3. EQE measurement of a P3HT:PCBM solar cell with a moth eye antireflective coating for different angles of incidence, compared with the EQE without the moth eye at normal incidence (dotted line). The inset shows the short circuit current that was obtained from the spectra by convolution with the AM1.5 solar spectrum.

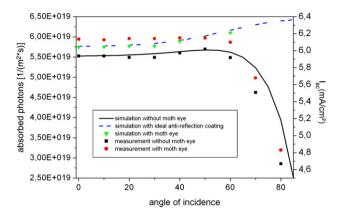


Fig. 4. Optical simulation of the number of photons absorbed in the active layer of a P3HT:PCBM depending on the angle of incidence, compared with the $J_{\rm sc}$ obtained from EQE measurements. Simulations: no anti-reflective coating (continuous line), hypothetical 'ideal' anti-reflection coating (dashed line), typical moth eye coating (triangles). Measurements: $J_{\rm sc}$ without moth eyes (squares) and with moth eyes (circles).

The result of the simulations (number of absorbed photons) as well as the measurement (J_{sc}) is shown in Fig. 4. The solid line represents the number of absorbed photons in a 160 nm thick P3HT:PCBM solar cell without moth eye coating, the dashed line the number in a solar cell for which the reflectivity at the air-substrate interface has been artificially set to zero. In this case, the number of photons is higher, and instead of a reduced absorption at high incidence angles, we observe an increase. If we use reflectivity values for the moth eyes that were obtained with an EMT calculation as described above, the absorption is still very similar to that of the ideal anti-reflective coating (triangles). However, if one compares these values with the $J_{\rm sc}$ that was obtained from the convolution of the measured EQE with the solar spectrum for the large-area solar cell with (circles) and without (squares) moth eyes, we find that the relative improvement due to the moth eye is always about 3%, independent of the angle.

We believe that this discrepancy between the simulation and the measurement is due to the imperfect matching of the moth eye substrate and the solar cell substrate, and that the even more favorable angle-dependence predicted by the simulations could be achieved if solar cell and moth eyes were integrated on a single substrate.

In conclusion, we have shown that the addition of a moth eye anti-reflection coating can improve the performance of an organic solar cell by approximately 2.5–3%, contributing to the improvement of their efficiency in general. In particular, this improvement is present for all angles of incidence.

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