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Experimental Evaluation of the Light Trapping Potential of Optical Nanostructures for Thin-Film Silicon Solar Cells

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Abstract

We describe a method based on nanoimprinting and non-absorbing insulating silicon nitride electrodes for evaluating the light trapping potential of photonic nanostructures for thin-film silicon solar cells. We validate our method by relating the optical reflectance of the full solar cell stack to the external quantum efficiency of functional cells. Our method provides a useful experimental tool to compare different nanostructures circumventing complications arising from parasitic absorption and electrical cell performance.

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1. Introduction

Excellent light trapping is essential to achieve high conversion efficiencies with thin-film silicon solar cells. Approaches that have already been successfully employed to increase light absorption in thin-film silicon modules on millions of square meters include the growth of transparent nanotextured zinc oxide (ZnO) electrodes with randomly-oriented pyramids via chemical vapour deposition (Fig. 1(a)) [1] and wet etching of crater-like structures into sputtered ZnO films (Fig. 1(b)) [2]. In particular, the pyramidal morphology has demonstrated outstanding light trapping capabilities and has led to several certified world record conversion efficiencies [3, 4]. Despite intensive experimental and theoretical efforts, neither the ideal surface morphology nor the ideal scattering characteristics have been identified to date.

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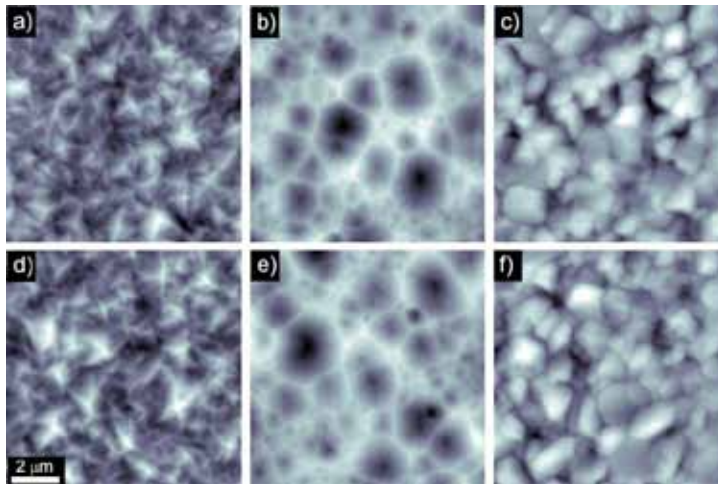


Fig. 1. Atomic force microscopy images of (a) ZnO pyramids, (b) ZnO craters, (c) rough silver. The corresponding replicas fabricated by nanoimprinting are shown in (d), (e) and (f), respectively.

Direct comparison of different nanophotonic structures via the external quantum efficiency (EQE) of the cell, from which the short-circuit current density can be derived, is often difficult as the surface morphology usually strongly affects the electrical cell performance metrics such as open-circuit voltage and fill factor. Although mild carrier collection problems may be eliminated by measuring the external quantum efficiency under an external bias voltage, the integration of more aggressive photonic structures often leads to Ohmic shunts in the cell, which render such a measurement impracticable. Furthermore differences in material properties of the electrodes, such as the free carrier density, have a direct impact on the parasitic light absorption in the cell. Consequently it is often not straightforward to identify if an enhanced spectral response is due to increased light trapping or reduced parasitic absorption [5].

Here we describe a method, based on nanoimprinting and non-absorbing insulating silicon nitride electrodes recently presented in Ref. [6], which allows us to compare different photonic nanostructure in the absence of parasitic absorption. We use our method to compare three classical photonic structures used to improve light absorption in thin-film silicon solar cells: pyramidally-textured ZnO grown by chemical vapour deposition (Fig. 1(a)) and sputtered-etched ZnO films with craters (Fig. 1(b)), both commonly used in the superstrate configuration. We also include rough silver (Ag) films (Fig. 1(c)), which are often used to enhance light trapping in the substrate configuration [7].

2. Experiment

Pyramidally-textured, slightly boron-doped ZnO layers were grown via low-pressure chemical vapour deposition at 180 °C and subjected to a 20 min argon plasma treatment to optimise the morphology for the growth of the cells [8]. Aluminium-doped ZnO layers were sputtered at 350 °C and etched in 0.5 % hydrochloric acid for 35 s. Sputtered silver films were deposited at 300 °C. At such high temperatures, silver undergoes a roughening transition. In all three cases, a 0.5 mm thick borosilicate glass substrate was used. All three test structures were then replicated into a transparent UV-curable lacquer using our recently developed high fidelity nanoimprinting technique [9, 10]. The replicated silver film serves as a good example, how nanoimprinting allows one to transpose an opaque master structure into a transparent

replica for use in the superstrate configuration [11]. The lacquer used for the fabrication of the replica on the borosilicate glass behaves optically like glass with no parasitic absorption and refractive index of 1.5.

Highly transparent indium oxide (In_2O_3) electrodes were sputtered on top of the replicas dosing a small amount of water vapour to the argon/oxygen process gas mixture [12]. The addition of water vapour leads, after an annealing step at 200 °C, to extraordinarily high electron mobilities in the hydrogenated indium oxide films ($\text{In}_2\text{O}_3\text{:H}$) above 100 cm^2/Vs resulting in an excellent near-infrared transparency, as the carrier density can be kept low. $\text{In}_2\text{O}_3\text{:H}$ layers were covered by a thin sputtered aluminium-doped ZnO layer which serves as a protective barrier layer against reduction in the hydrogen-rich plasma during the subsequent cell deposition. Silicon nitride (SiN_x) layers were deposited by plasma-enhanced chemical vapour deposition using silane and ammonia as precursor gases. The refractive index of the SiN_x layers as measured by spectroscopic ellipsometry, was adjusted by modifying the ammonia/silane ratio, so as to obtain a refractive index comparable to the $\text{In}_2\text{O}_3\text{:H}$ films.

Micromorph p-i-n thin-film silicon tandem solar cells consisting of a 250 nm thick hydrogenated amorphous silicon (a-Si:H) top cell and 1100 nm thick hydrogenated microcrystalline silicon ($\mu\text{c-Si:H}$) bottom cell were deposited by plasma-enhanced chemical vapour deposition in an industrial reactor with parallel plate electrode configuration [13]. Silane and hydrogen were used as a source gas for intrinsic layers. For doped layers, we also used methane and carbon dioxide. P-type and n-type doping was achieved by adding borontrimethyl and phosphine.

The EQE of the top and bottom cell of the micromorph tandem was measured as described in Ref. [14], using red and blue bias light, respectively. Reflectance (R) measurements were carried out on a photospectrometer equipped with an integrating sphere. A white diffuse dielectric back reflector was mounted behind the back electrode for characterisation.

3. Results and discussion

Figure 2(a) shows external quantum efficiency measurements of a micromorph solar cell with $\text{In}_2\text{O}_3\text{:H}$ electrodes deposited on replicated ZnO pyramids shown in Fig. 1(b). Top as well as bottom cell EQEs are shown along with the sum of the two, which represents a useful quantity to characterise the total light conversion, that is, the light trapping, in the cell. Details on the performance of this cell are given in Ref. [13]. In order to be able to investigate light trapping in the absence of parasitic absorption in the cell, we rescale the EQE as described in Ref. [15] as well as in Refs. [5, 6]. In the case of ideal carrier collection, we have $\text{EQE} + R + A = 1$, where A represents the parasitic absorption in the cell. Assuming now zero parasitic absorption, we can define normalised quantities EQE' and R' satisfying $\text{EQE}' + R' = 1$. This normalisation is achieved by splitting A equitably between EQE' and R' : $\text{EQE}' = \text{EQE}/(1 - A)$ and $R' = R/(1 - A)$. The normalised $\text{EQE}' = 1 - R'$ is also shown in Fig. 2(a).

Figure 2(b) presents the reflectance measurement (plotted as $1 - R$) of the full micromorph cell stack with $\text{In}_2\text{O}_3\text{:H}$ electrodes. As the reflectance inside the band gap of $\mu\text{c-Si:H}$ (above 1100 nm), where $\mu\text{c-Si:H}$ absorbs no light, does not reach 100 %, we conclude that there is significant parasitic absorption in the near-infrared present. To determine where the different contributions to this parasitic absorption are coming from, we deposited an equivalent micromorph cell stack but replacing the $\text{In}_2\text{O}_3\text{:H}$ electrodes with insulating SiN_x electrodes. As the refractive index of SiN_x is similar to $\text{In}_2\text{O}_3\text{:H}$, we obtain an optically equivalent device, but free of parasitic absorption in the electrodes. This leads to a significant increase in the reflectance at 1200 nm due to the absence of free carrier absorption in SiN_x . However, as can be seen

from Fig. 2(b), still more than 20 % of light is absorbed parasitically at this wavelength. In a third step, we eliminated also the p- and n-doped silicon layers in both top and bottom cell. This increases the reflectance at 1200 nm further to almost 95 %.

In Fig. 2(c) we compare the normalised EQE' from Fig. 2(a) with the reflectance measurement of the cell stack without the doped silicon layers and with SiN_x electrodes from Fig. 2(b). Normalised EQE' and 1 - R almost coincide as we eliminated parasitic absorption in both cases, demonstrating the internal consistency of the two methods. Our approach therefore allows one to link the optical measurement to the electrical measurement and represents a useful tool for comparing the light trapping capacities of different photonic nanostructures. This can be done by a simple reflectance measurement on the cell stacks avoiding troubles with the electrical performance of the cells, which may eventually be solved by adapting the silicon layer deposition regimes to the new morphology.

In Fig. 2(d) we compare such reflectance measurements for cell stacks without doped silicon layers and SiN_x electrodes on replicated ZnO pyramids (Fig. 1(d)), replicated ZnO craters (Fig. 1(e)) and the replicated rough silver morphology (Fig. 1(f)). The reflectance measurements for a flat reference cell stack with no light trapping are also shown. Interestingly, for wavelengths above 950 nm, where light trapping is crucial, as the absorption coefficient of μc-Si:H becomes small, all three structures perform optically identically within the experimental errors.

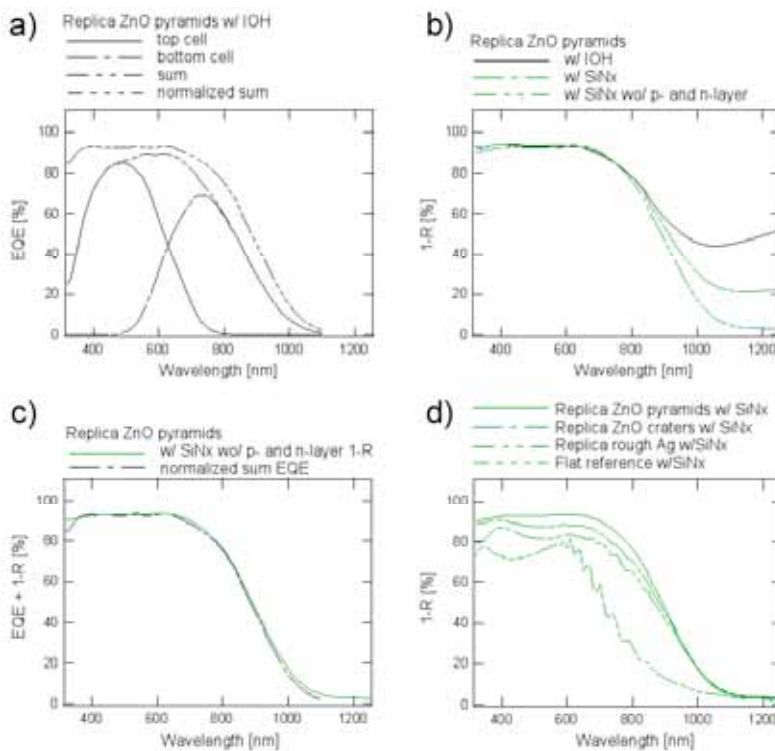


Fig. 2. (a) External quantum efficiencies, (b) reflectance measurements, (c) direct comparison between normalised external quantum efficiency and reflectance measurement, (d) reflectance measurements of the cell stack on the three classical nanostructures of Fig. 1 and a flat reference cell stack.

At wavelength below 950 nm, the pyramids clearly exhibit the best optical performance. In this spectral range, more light is reflected from the cell stack with craters. For the rough silver morphology the reflection further increases and interferences fringes become clearly apparent.

For the ZnO crater morphology, this is a known problem. To minimise optical reflection at the ZnO-silicon interface, Fujibayashi *et al.* [16] and later Das *et al.* [17] developed a TiO₂ antireflection layer with intermediate refractive index and were able to demonstrate a clear improvement in EQE. Buehlmann (now Cuony) *et al.* [18] applied similar TiO₂ antireflection layers at the interface between ZnO pyramids and silicon. They observed that the suppression of reflection upon introduction of a TiO₂ layer is strongly dependent on the roughness of the ZnO pyramids. Rougher pyramids also exhibit an antireflective effect as the effective refractive index between the two optical media changes gradually across a sufficiently rough interface.

While increasing roughness typically improves light incoupling into the silicon absorber layers, electrical cell performance usually suffers due to the formation of porous areas between microcrystalline grain agglomerates, known as cracks, caused by the growth dynamics on rough morphologies. These cracks are detrimental for both the open-circuit voltage and the fill factor [19]. Promising recent developments towards more morphology-tolerant silicon layers provide a clear roadmap for the integration of even more aggressive optical nanostructures into micromorph solar cells [20-22].

4. Conclusions

We described and validated a new method based on nanoimprinting and insulating SiN_x electrodes to compare the light trapping performance of different photonic nanostructures for thin-film silicon solar cells in the absence of parasitic absorption and electrical complications. We applied our method to three different classical light trapping structures and discussed ways to further improve light absorption in thin-film silicon cells.

Acknowledgements

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