A non-resonant dielectric metamaterial for the enhancement of thin-film solar cells

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Abstract
Recently, we have suggested a dielectric metamaterial composed of an array of submicron dielectric spheres located on top of an amorphous thin-film solar cell. We have theoretically shown that this metamaterial can decrease the reflection and simultaneously suppress the transmission through the photovoltaic layer because it transforms the incident plane wave into a set of focused light beams. This theoretical concept has been strongly developed and experimentally confirmed in the present paper. Here we consider the metamaterial for oblique angle illumination, redesign the solar cell and present a detailed experimental study of the whole structure. In contrast to our previous theoretical study we show that our omnidirectional light-trapping structure may operate better than the optimized flat coating obtained by plasma-enhanced chemical vapor deposition.

Keywords: thin-film solar cells, anti-reflecting coating, omnidirectional coating, light-trapping structure, focusing, metamaterial

1. Introduction

Currently, great attention is paid to thin-film solar cells (TFSCs), solar cells with a photovoltaic layer (doped semiconductor comprising p-nor p-i-n junctions) of sub-micron thickness. First, such TFSCs can be built on flexible substrates using the so-called roll-to-roll technology that decreases the market price per unit area of the solar cell by an order of magnitude compared to solar cells of the 2d generation [1]. Second, the decrease in the required amount of purified semiconductor (also by one or even two orders of magnitude) implies the corresponding reduction in toxic waste, and the production of TFSCs becomes practically harmless to the environment [2, 3]. Of course, in favor of these practical advantages one sacrifices the ultimate efficiency of TFSCs. However, if the overall photovoltaic (PV) efficiency in the operational band of a practical TFSC is equal to at least 5–7%, i.e. twofold smaller than that of commercially available solar cells, its industrial perspectives become favorable [1]. Notice that TFSCs are dedicated mainly to domestic needs and may be used throughout the world.

The increase in efficiency without dramatic enhancement of the fabrication costs has become a key issue for TFSCs. For this purpose one often utilizes an anti-reflecting coating (ARC). Optimized multilayer ARCs which are usable for wafer-type and multi-junction solar cells are not compatible with the concept of TFSCs since this is very expensive. A sufficient ARC for a TFSC is a simple blooming layer with optimized thickness and refractive index [1]. For example, for a TFSC with an amorphous silicon (a-Si) layer located on top (when the top electrode is a wire mesh) the silicon nitride covering is recommended. It may be fabricated using the plasma-enhanced vapour deposition (PECVD). This simple ARC with optimized thickness reduces the reflection losses from nearly 50% to 15% [4].

However, for TFSCs operating with visible light, their reflection losses are usually lower than their transmission ones. If the PV layer is as thin as 400 nm more than one half of the normally incident light energy (over the visible...
spectrum) transmits into its substrate (usually it is the rear electrode). This energy is lost, moreover its parasitic absorption results in the additional heating of the whole cell. The heating of the PV layer is very harmful since it produces the dark current which is subtracted from the photocurrent. Practically, one needs not only to prevent the reflection but also to concentrate the light inside the PV layer where the light at least partially converts into electricity. The problem of the local light concentration is very interesting and has recently resulted in a body of literature unified by a common topic: light-trapping structures (LTS) for TFSCs. The majority of work in this field corresponds to two approaches: so-called plasmon enhancement of TFSCs and so-called photonic crystal enhancement (see e.g. in [5–12]). We do not aim to review this literature. For our purposes it is enough to mention general drawbacks of all these structures. Photonic crystals embedded into TFSCs are expensive structures. Even in mass production the cost of such TFSCs will be much higher than that of TFSCs enhanced by a simple ARC. A twofold increase in the power output claimed in the best cases hardly justifies such costs. Plasmonic structures can be relatively cheap (e.g. nanosiland films of silver or gold), however, they possess inherent resonant losses in the metal elements. Therefore such LTSs do not usually stand comparison with the ARC. Several works claim more significant enhancement due to regular structures of gold or silver nanoelements. These regular arrays, when located beneath the PV layer or incorporated into it, can mimick an effective optical facet converting the incident plane wave into eigenmodes propagating in the PV layer as in the waveguide [10, 13]. Notice that usual textured optical facets cannot be applied to TFSCs due to their small thickness (this restriction is called Yablonovich’s limit). Alternatively, a regular array of specially designed metal nanoelements may convert the plane wave into collective oscillations of the array. These modes are located on the frequency axis rather far from the plasmon resonances of individual metal elements and are characterized by negligible parasitic losses in the metal [14, 15]. It makes this approach promising for the enhancement of TFSC. However, regular arrays of metal nanoelements obviously imply high fabrication costs, and their market prospects are also disputable.

Recently, all-dielectric LTSs exploiting different kinds of resonances in an array of densely packed dielectric spheres (whispering gallery resonance, magnetic Mie resonance, a combination of these resonances, and spatial resonance of a photonic crystal of dielectric spheres) have been suggested for the enhancement of TFSCs [16–19]. In view of commercially available micron and submicron dielectric spheres, including quartz and silicon ones, these structures seem to be promising. However, their resonant behavior implies a more or less narrow frequency range of the light trapping effect which is not completely compatible with the idea of so broadband object as a solar panel (e.g. solar cells based on a-Si operate in the whole visible range). Therefore, in [20], we suggested and theoretically studied a non-resonant, i.e. fundamentally broadband LTS for the PV layer of total thicknesses 300–500 nm. Our structure is a metamaterial of densely packed dielectric spheres as in [16–19], however, our design parameters allow different (unfortunately, competing) physical mechanisms for the suppression of both reflection and parasitic transmission. The reflection is suppressed due to the quadrupole polarization of spherical inclusions forming the metamaterial. The quadrupole radiation of polarized metamaterial dominates in the shortwave part of the spectrum. In the reflected wave this radiation destructively interferes with the dipole re-radiation from the bulk of the PV layer. In the long-wave part of the spectrum the roughness of the metamaterial layer resulting from the curvature of nanospheres becomes not very significant for the reflected wave. For the range 400–600 nm our LTS refers to the class of quadrupole metamaterials, and, in what concerns the reflective properties, our metamaterial operates almost as an ARC. This property of an array of densely packed spheres has been studied earlier (see e.g. in [21, 22]). However, in what concerns the parasitic transmission through the optically thin photovoltaic layer, in [20] we revealed the novel mechanism of light-trapping. The parasitic transmission is suppressed over the whole visible range due to a simple macroscopic effect: light focusing by individual spheres, which holds in spite of their modest optical size. Over the axis of wavelengths the focusing mechanism appears when the wavelength (in free space) becomes smaller than the sphere diameter. So, the device enabling a usual optical focusing would allow the field concentration inside the PV layer, which in turn increases the absorption and suppresses the transmission. The elements of our LTS in [20] were slightly submicron spheres formed of a transparent dielectric like polystyrene or silica [20]. The best diameter of spheres for maximal suppression of the reflectance over the visible range is nearly equal to $d=500$ nm, whereas the focusing functionality requires larger spheres. The optimal diameter of silica spheres, $d=900$ nm, was found from the compromise between the minimal reflection and minimal transmission [20].

Though metamaterials of dielectric nanospheres for the enhancement of semiconductor TFSCs had been suggested before [20] they exploited different design parameters of the structure and therefore implemented only resonant, rather narrow-band, mechanisms of the local field enhancement (see e.g. [16–19]). In these works the overall impact of the metamaterial was more modest than that theoretically demonstrated in our work [20]. In [20] we claimed a 44% gain in the PV absorption granted by an array of optimized densely packed spheres located on top of a silicon PV layer. The last one was backed by alumina zinc oxide (AZO) half-space operating as a rear electrode. A noticeable part of this gain resulted from light-trapping. However, the solar cell in [20] had an impractical design, and was difficult to realize experimentally. It was only an initial step of our theoretical studies in this direction. Further investigations have shown that the whole approach needs to be revised.

2. Concept of the metamaterial

The mechanism of the additional enhancement was found by inspecting the operation regime for the oblique incidence and averaging the results for the PV absorption over the incidence
focusing is mainly collimation [20]. Such collimated beams are individually. Since it has quite a small numerical aperture, this for the normal incidence every nanosphere focuses the light improves the focusing properties of our metamaterial. Really, a tilt to the southern side, a typical Finnish roof.

As to the suppression of the overall reflectance, we propose to use the multilayer TFSC based on a-Si which in the plain case (without ARC or LTS) would possess sufficiently small reflectance over the visible range. It can be obtained if we increase the number of layers involving the top electrode of transparent conducting oxide, such as AZO. Optical losses in AZO are substantial, however the figure of merit is higher than in a-Si, and the refractive index of AZO is inbetween that of a-Si and that of free space. Therefore the top electrode of AZO can partially implement the functionality of ARC. Additionally one may introduce an optical contrast into our a-Si layer performing it as a p-i-n structure: an i-type layer has lower optical losses than p- and n-type layers. If the rear electrode, also formed of AZO, is not very thick a certain contribution to the interference damping of the reflectance can be made by a transparent substrate (wafer) of our TFSC. This six-layer structure may reflect much less than the three-layer one from [20].

3. Modelling

3.1. Plain solar cell

A piece of a TFSC designed in accordance with our last speculation is depicted in figure 2(a). This sketch is copied from our simulation project (Ansys HFSS). In this figure it is denoted: 1. optical glass substrate of thickness 500 μm; 2. 270 nm thick AZO; 3. p-type 70 nm thick a-Si; 4. i-type 400 nm thick a-Si; 5. n-type 100 nm thick a-Si; 6. 220 nm thick AZO. Some of these design parameters were dictated (with possible deviations of a few nm) by the available fabrication process, the others resulted from optimization. We numerically (HFSS) simulated the absorption coefficient \( A(\lambda) \) defined as the power absorbed in the PV layer per unit area normalized to the incident power flux. This coefficient was calculated over the visible range \( \lambda = 400–800 \) nm for three incidence angles. Then for every incidence angle we calculated the absorption efficiency \( \eta \) defined as the integral of \( A(\lambda) \), with the weight function representing the solar spectrum. Assuming the collection efficiency close to unity at all wavelengths (it is an adequate assumption for a sandwiched
TFSC) the photocurrent is proportional to the product of $\eta$ and PV spectral response of a-Si averaged over the visible range (see e.g. in [2, 7–9]). Since the spectral response of silicon does not depend on the cell design the gain in the absorption efficiency of the cell due to the presence of either ARC or LTS is practically equal to the gain in the photocurrent [7–9, 14, 15, 20].

Optical parameters of the media involved were taken from [24] (we assume the same doping level of p-type and n-type sub-layers as in this work). Besides HFSS simulations, we calculated $A(\lambda)$ analytically using the Matlab code based on the transfer matrix method. This code was presented in [23]. The results for $A(\lambda)$, in the case of normal incidence presented in figure 2(b) show that the plain TFSC with multilayer structure absorbs the visible light quite well. The agreement between the Matlab code and the HFSS simulator is rather good. Similar calculations were done for angles $\theta=30^\circ$ and $60^\circ$, and the design parameters were optimized for the mean value of the absorption efficiency.

### 3.2. Solar cell with ARC

This structure is depicted in figure 3(a). It differs from figure 2(a) only by the presence of a blooming layer with thickness $h$ and refractive index $n$, which optimized. The PV absorbance (integral over the visible range) versus $h$ for different values of the refractive index is shown in figure 3(b) as a polar plot for the case $\theta=0$. Similar plots were calculated for $\theta=30^\circ$ and $60^\circ$. The optimal parameters corresponding to the maximal mean value of $\eta$ (averaged over three incidence angles) were found $n=1.45$, $h=80$ nm. The medium with practically dispersionless and lossless refractive index $n=1.45$ is silica. Our ARC demonstrates the best operation at $\theta=30^\circ$. For $\theta=0$ and $60^\circ$ its presence slightly worsens the absorption efficiency. This is not surprising since the blooming effect is based on the wave interference which cannot be robust to the incidence angle. We have checked that an ARC optimized for the normal incidence strongly decreases the absorption efficiency for $\theta=60^\circ$ and is, therefore, far from being optimal.

### 3.3. Solar cell with LTS

A piece of the structure with $3 \times 3$ spheres of silicon dioxide replacing the flat ARC is shown in figure 4(a). This picture is just to show that spheres should stay side by side. We completed the simulations only for one cell, with one sphere on top limited by master and slave boundaries. We have simulated $A(\lambda)$ for this structure and compared with $A(\lambda)$ calculated for the plain TFSC and for the TFSC with ARC. For the normal incidence this comparative plot is shown in figure 4(b). Figure 5(a) shows the plot for $\theta=30^\circ$ and figure 5(b) shows the plot for $\theta=60^\circ$. The results for LTS corresponds to $d=1 \mu$m. The optimized diameter $d$ of spheres is slightly smaller, however, within the interval $d=1\pm0.1\mu$m the result for the absorption efficiency depends on $d$ rather weakly. Since the silica spheres with $d=1 \mu$m are commerically available in a liquid host we show the results for such spheres.

To confirm our idea on the improved (cascaded) focusing for the oblique incidence we show two color maps of the electric field amplitude $E$ at $\lambda=500$ nm. Both color maps correspond to the optimized structure. First, in figure 6(a) we depict the distribution of $E$ in the vertical cross section of the structure for the normal incidence. In this regime the spheres focus the light individually. The parasitic transmittance though the PV layer in this case is close to 20%. Second, in
Figure 6(b) we present the similar color map for the oblique incidence. The picture clearly corresponds to the cascade focusing, for which the result is better utilization of the volume of semiconductor. In this regime we observe two collimated beams per one sphere and the absorption holds in the whole volume. The parasitic transmittance in this regime is much smaller than 1% (note that the color map in this figure is logarithmic). It is worth noticing that the PV absorption in all cases strongly dominates over the absorption in the top AZO electrode though the focus area is partially located in it. This is so because optical losses of AZO are much lower than those of a-Si.
To conclude this section we present the table for the gain in absorption efficiency granted by our ARC and by our LTS to our TFSC for three incidence angles. The mean gain due to our ARC is equal to 9%, whereas that offered by our LTS is 34%.

4. Experiment

To demonstrate the efficiency of the proposed dielectric metamaterial an a-Si based TFSC with LTS was fabricated. The present approaches to the fabrication of a p-i-n junction include in situ doping during PECVD deposition of a-Si. Since our facilities are not suitable for the preparation of doped a-Si layers, we have developed an unusual method for the fabrication of p-i-n structure.

4.1. Process flow

Firstly, the glass wafer was covered with 272 nm thick AZO film using the ALD process [25]. On the top of the AZO layer was fabricated a p-i-n junction from a-Si with a thickness of 570 nm. For this purpose a 70 nm thick intrinsic amorphous silicon was deposited by PECVD and p-type doped by ion implantation. In a typical TFCS the front side of the p-i-n junction is a p-doped layer. In our case the inverse order of layers was used to avoid the creation of a parasitic p-n junction. This junction can appear due to contact of intrinsic p-AZO with n-type doped a-Si.
Chips of TFSC with dimensions 1 × 2 cm² were fabricated on double side polished glass wafers of thickness 0.5 mm and diameter 100 mm, from Siegert Wafer GmbH.

The deposition of AZO was done by using ALD system Beneq TSF-500 at a temperature of 200 °C. The thickness deviation of ALD layers was less than 1 nm. A-Si was deposited by using a PECVD system Plasmalab 80 Plus from Oxford Instrument Plasma Technology at a temperature of 200 °C. P- and n-type doping was done by using an ion implanter Eaton NV 3206. Implantation of boron or phosphorous was done at energy 20 keV and surface dose 200 °C. P- and n-type doping was done by using an ion implanter Eaton NV 3206. Implantation of boron or phosphorous was done at energy 20 keV and surface dose

4.2. Experimental details

Chips of TFSC with dimensions 1 × 2 cm² were fabricated on double side polished glass wafers of thickness 0.5 mm and diameter 100 mm, from Siegert Wafer GmbH.

Immediately after ion implantation a 500 nm thick i-layer was deposited by PECVD. To create an n-doped layer at the front side of the TFCS an implantation of phosphorous was used. The estimated thickness of the n-layer is close to 100 nm. In the case of the p-doped front layer its thickness would be around 50% higher. It is one more reason to use inverse order of p-i-n layers in our TFCS. Above the n-Si the front layer of AZO, of thickness 221 nm, was deposited by ALD and patterned to serve as the top electrode. After that vias were opened to back the AZO layer, providing access to the bottom electrode. Finally, the wafer was cut into TFSC chips. The resulting structure of the TFSC without LTS is presented in figure 7. At this point some TFSCs were covered by ARC or a metamaterial layer.

4.3. Results

We could not measure the PV absorbance in the spectrum since it cannot be separated from the parasitic absorption in AZO. That is why we measured the external quantum efficiency (QE) of our TFSC. The result for the plain cell illuminated under different angles is shown in figure 8. Since QE is proportional to \( A(\lambda) \) we may state the good correspondence of the measured data with the theoretical predictions. The low absolute values of QE are related to the low doping level of a-Si (in our implanting process we cannot reach the minor carrier density \( 3 \cdot 10^{18} \) per cubic cm as in [26]).

Then some of the samples were covered with an ARC of silica using the PECVD. The measurements using the profilometer (Dektak/XT) have shown that the practical thickness is equal to 87 nm (averaged value for all samples). In accordance to our simulations the ARC of silica with thicknesses in the interval \( h = 80–90 \) nm have nearly the same operation characteristics. Therefore we have not remade these samples. The result for QE is shown in figure 10. In agreement with the theory the gain granted by our ARC is maximal for \( \theta = 30° \) and the spectral curves of QE for all three angles qualitatively fit our predictions for \( A \).

Some samples were covered by silica spheres with \( d = 1 \) µm. Available spheres have rather noticeable deviations of \( d \), namely \( d \) is within the interval 1–1.2 µm. We utilized the method of natural deposition (self-adhesion) of nanospheres suggested in [16] (see also [27]). This method allows a very good quality of LTS on the millimeter sized areas. However, the areas of touched spheres forming the desired metamaterial alternate on our samples with (also millimeter sized) areas of untouched spheres and with clean areas. The result of the deposition is illustrated by figure 9. Since only the areas of touched spheres operate properly, in our measurements we illuminated all solar cells by a collimated light beam tilted under the necessary angles \( \theta = 0 \), \( \theta = 30° \) and \( \theta = 60° \). The result for QE is shown in figure 11. In agreement with the theory the largest gain is achieved for \( \theta = 60° \) and the spectral curves for QE qualitatively repeat.
those for $A$ as it was obtained above for the plain TFSC and for that enhanced by the ARC. Notice that a sufficient number of measurements were reproduced with different samples and the achieved coincidence makes the claimed results fully reliable. This comment concerns all kinds of our solar cells; plain ones, those enhanced by the ARC and those with the LTS.

We have measured the photocurrent $J$ induced in all our samples using the same collimated beam. The measurements were done for all three incidence angles. The gain in $J$ is practically equal to the integral gain in $QE$. The results for the gain granted to our TFSC by both ARC and LTS are presented in table 2.

### Figure 9. Microscopic images of silica spheres on top of our TFSC for different areas. The quality of our LTS within the area of mutually touching spheres is illustrated by the SEM image (inset).

### Figure 10. Measured external quantum efficiency of the TFSC enhanced by spheres for different angles of incidence $\theta$. The domain of mutually touching spheres was illuminated by a collimated light beam.

### Figure 11. Measured external quantum efficiency of the TFSC enhanced by ARC for different angles of incidence $\theta$.

| Table 1. Gain in $\eta$ (theory). |
|----------------------------------|
| Gain (%) $\theta$(°) | 0 | 30 | 60 |
| ARC                | −8 | +36 | −1 |
| LTS                | +15 | +16 | +70 |
The mean gain due to our ARC is equal to 16%, whereas that offered by our LTS is 32%. The metamaterial LTS also in practice turns out to be noticeably more efficient than the ARC. We want to stress that this was achieved in spite of the better operation of the practical ARC. In practice the ARC offers a gain twice as big as was predicted. The reason for this strange result is, in our opinion, lower optical losses in the doped a-Si compared to our theoretical mode. Lower losses allow better matching of the whole structure as it usually holds for flat multilayers. So, better operation of the blooming layer is an indirect symptom of the insufficient doping (the last one we could not, unfortunately, control in our fabrication process). As to our LTS, its practical gain is almost equal to the prediction by our theory!

To better understand the role of the quality of our LTS we have experimentally studied the operation of our TFSC illuminating normally four different regions of the same sample (see figure 9). In figure 12, showing the power reflectance in the visible spectrum, one can see how strongly the anti-reflecting properties of the metamaterial depend on the package of spheres. In fact, the presence of spheres (even isolated) reduces the reflectance compared to the clean surface of the cell. However, this reduction becomes really significant (twofold) only for densely packed spheres. The lowest reduction of the reflectance corresponds to the case when the light beam illuminates the boundary between the regions of touched spheres and untouched spheres. The quantum efficiency of the sample turned out to be worse (and the photocurrent is smaller) than those of the plain TFSC if we illuminated the region with this boundary. At first glance, it seems strange because on the plots presented in figure 12 the maximal reflectance still corresponds to the clean surface. However, it is only the normal (mirror-type) reflectance. The illumination of poor regions of the sample results in the noticeable omnidirectional scattering which is maximal when the beam impinges on the boundary between two regions. This scattering is absent when we illuminate the high-quality regions of the sample. Our last study fits the results of [27], though in this work a different (resonant) mechanism of the enhancement was implemented.

5. Conclusions

In this work we have developed the concept of focusing all-dielectric metamaterial for the enhancement of thin-film solar cells, and found that its operation becomes advantageous compared to the anti-reflecting coating for the oblique incidence of sunlight. This advantage results from the cascade focusing effect. In order to prevent the strong reflection the solar cell should comprise several layers with optical contrast. To implement this concept we have theoretically studied the operation characteristics of the TFSC in the range of incidence angles $\theta = 0^\circ - 60^\circ$. For this range we theoretically found that our LTS (of silica spheres with micron radius) operates better than the optimal flat ARC (also of silica), whereas the gain due to the replacement of the ARC with the array of spheres equals 25%. We have practically implemented several samples of our TFSC with either LTS or ARC on top of the samples. Practical measurements have shown that our LTS offers nearly the same enhancement compared to the plain cell as we have theoretically predicted. The total advantage of our LTS compared to the ARC is 16% in the photocurrent.

So, our LTS is advantageous enough to open a new door for omnidirectional coatings; non-resonant, all-dielectric (lossless) omnidirectional coatings for broadband light-trapping. Our spheres are available commercially and the fabrication of the LTS promises to be cheap when in mass production. We consider our result quite inspiring and believe that our structures are helpful in the industrial adaptation of silicon TFSCs.

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