Comparison and optimization of randomly textured surfaces in thin-film solar cells

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Abstract: Using rigorous diffraction theory we investigate the scattering properties of various random textures currently used for photon management in thin-film solar cells. We relate the haze and the angularly resolved scattering function of these cells to the enhancement of light absorption. A simple criterion is derived that provides an explanation why certain textures operate more beneficially than others. Using this criterion we propose a generic surface profile that outperforms the available substrates. This work facilitates the understanding of the effect of randomly textured surfaces and provides guidelines towards their optimization.

OCIS codes: (040.5350) Photovoltaic; (350.4238) Nanophotonics and photonic crystals; (350.6050) Solar energy; (290.0290) Scattering

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1. Introduction

Photon management in thin-film solar cells constitutes an indispensable tool to make them outperform the established wafer-based technology[1]. Thin-film solar cells made of hydrogenated amorphous silicon (a-Si:H) and microcrystalline silicon (μc-Si:H) have lower fabrication cost as their fabrication requires less energy and raw materials[2]. One major drawback of mass produced, stable a-Si:H cells is that the maximum thickness of their light absorbing layer is limited to about 300 nm, which is insufficient to absorb light with energies close to the electronic band gap. Therefore, an optical structure must be implemented to increase the absorption for wavelengths in the range of 600 to 800 nm, above which further efforts become pointless [3]. At smaller wavelengths a-Si:H is sufficiently absorptive such that all the incoming light is absorbed and no special care needs to be taken, whereas at larger wavelengths a-Si:H is such poorly absorptive that even an enhancement of the absorption by orders of magnitude would contribute marginally to a noticeable enhancement of the current.

Structures to increase the effective absorption include reflective back contacts [1], fluorescent dyes [4], dielectric gratings [5], photonic crystals [6], or plasmonic nanoparticles [7]. They aim at decreasing the cell reflectivity and at increasing the optical path length by deflecting normally incident photons obliquely into the active layer.

In this paper we consider a model system comprising randomly textured interfaces [8]. In most solar cells these interfaces are integrated by adapted growth conditions and/or by using an appropriate technique to etch a layer of a transparent conductive oxide, deposited on a glass substrate. On top of this interface the a-Si:H p-i-n structure is then deposited. The cells are either terminated by a back reflecting structure or by a transparent layer that may serve as, e.g., an intermediate reflector (IRL) in a tandem cell where the a-Si:H cell is equipped with an additional μc-Si:H p-i-n structure [9].

The intended optical effect of such a randomly textured surface is twofold [10]. On the one hand it should lower the reflection losses at the entrance facet and on the other hand it should scatter the light such that the optical path of each photon in the solar cell gets increased. The optimization of the surface profile is usually performed by heuristically modifying a selected

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fabrication parameter and observing the effect on the absorption enhancement [11]. This, however, is insufficient for obtaining a deeper understanding of the optimization of such textures. Moreover, this approach led to various substrates currently available which have quite distinctive morphologies with strongly deviating scattering properties.

Despite its importance regarding solar cell efficiency, the optical properties of such randomly textured surfaces were only sporadically theoretically/numerically investigated thus far and in most cases only in terms of their scattering properties [12]. The parameter in the focus of interest is usually the haze, which is defined as the share of non-specularly scattered light into the far-field. It is defined as:

$$haze = \lim_{r\to\infty}\left[\frac{\int_0^{\pi}\int_0^{2\pi} S_{\mathrm{r}}(r,\theta,\phi)d\theta d\phi - S_{\mathrm{r}}(r,0,0)}{\int_0^{\pi}\int_0^{2\pi} S_{\mathrm{r}}(r,\theta,\phi)d\theta d\phi}\right] \quad (1)$$

with θ and ϕ being the usual spherical coordinates and $S_{\mathrm{r}}(r,\theta,\phi)$ being the radial component of the Poynting vector $\mathbf{S}(r,\theta,\phi)$ in the far-field. This can only be regarded as a first step since the relation to absorption enhancement is usually not established [13]. Moreover, due to experimental constraints the scattering properties are usually only investigated against air [14]. These scattering properties are usually only accessible after fabricating the patterned surface into the transparent conductive oxide (TCO). If the surfaces are integrated into the final cell the scattering process cannot be accessed anymore. Moreover, if a high permittivity material is attached at the backside, a large portion of the scattered light would be trapped by total internal reflection and it is difficult to extract it. But the characterization of the scattering process against air is insufficient to a certain extent, since in the solar cell the light is scattered at the randomly textured interface into a medium of high permittivity such as silicon.

In this paper we shall lift these limitations whereas our contribution will be fourfold. At first, we argue that the scattering properties against a high permittivity medium have to be considered when evaluating the performance of a certain texture. At second, we show that the haze is an insufficient criterion since it neglects the angular dependency of the scattered light which is important for the path length enhancement. At third, we introduce a criterion that correlates the scattering properties of a substrate to the absorption enhancement. The criterion shall not be understood in a rigid manner but merely as a guideline. Its limitations are discussed. We apply this criterion to three referential randomly textured surfaces, called the substrates. We consider a commercially available Asahi-U substrate, a substrate from the IEF-5 in Jülich, Germany, (Jülich substrate) and a substrate from the IMT in Neuchâtel, Switzerland (Neuchâtel substrate). From an optical point of view it is found that the Neuchâtel substrate outperforms the others. On the base of the introduced criterion it can be reasonably explained why. At fourth, we use the criterion and scalar optical theory to optimize the surface texture of a generic profile and show that this surface prevails against the substrates as considered here, hence constituting at least a proposal for a further improved substrate. Prior to any further consideration it has to be stressed that we only aim at characterizing the optical properties of the substrates. Other aspects such as, e.g., their electrical properties or their integration into the final solar cell, will certainly affect the performance and must be accounted for when ultimately selecting the optimum substrate.

2. Comparing and optimizing substrates

The considered substrates are shown in Fig. 1. The surface profiles of fabricated samples were measured with an atomic force microscope (AFM). The substrates are either commercially available [Fig 1(c) - Asahi-U] or were fabricated with procedures as described in literature. Details for the Jülich [Fig. 1(a)] and the Neuchâtel substrate [Fig. 1(b)] can be found in Refs.

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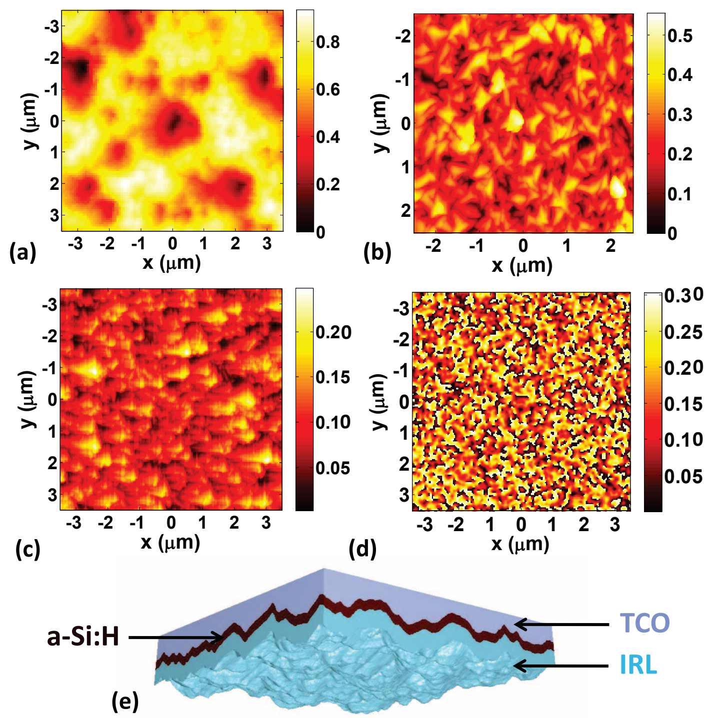

Fig. 1. Topology of the various considered randomly textured surfaces. (a) the Jülich substrate, (b) the Neuchâtel substrate, (c) the commercial substrate Asahi-U, and (d) a generic topography as a result of optimization in this work. The sequence of layers of the cell is shown in (e).

11 and 15, respectively. The optimized profile, as shown in Fig. 1(d), corresponds to a computer-generated hologram whose generic profile will be motivated below. In our rigorous analysis the substrates as shown in the figures are fully considered [16].

For the sake of comparability, the considered thin-film solar cell is identical for all substrates and depicted in Fig. 1 (e). The medium in the illuminating region is a semi-infinite TCO (n = 1.915). The modulation of the TCO surface corresponds to the height of the AFM measured substrate. A glass substrate was not explicitly considered since the height of the TCO layer on top of it may be subject to optimization and is not known to us for all substrates. Moreover, it is anticipated that the absolute thickness is of minor importance since the TCO is optically thick (over 500 nm), the index contrast between a glass substrate and the TCO is rather low and the optical actions are dominated by the texture. Below this randomly textured surface we assume a conformally deposited a-Si:H layer with a thickness subject to variations (125 nm, 250 nm, 375 nm). The optical properties of a-Si:H were taken from literature [17]. The material at the back is assumed to be an intermediate reflector (n = 2). We have to stress that the analysis of an entire tandem cell is too challenging for current computational resources. Therefore, our results can strictly be applied only to a single layer solar cell and we choose to concentrate on the a-Si:H top cell which usually limits the current in a stabilized tandem solar cell. Extrapolation towards tandem solar cells is only possible if the thickness of the intermediate layer is so large that it is no more important [10]. We evaluate the absorption enhancement in the a-Si:H layer at two different wavelengths (633 nm and 720 nm) corresponding roughly to the upper and lower edge of the spectral domain essential for absorption enhancement. There, the

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absorption tends to be small and the thickness of the active layer is insufficient to absorb all the incoming light with just one single passage. The structure is illuminated by a linearly polarized plane wave at normal incidence (0°). We use the finite-difference time-domain (FDTD) method to compute the field in the entire space [18]. The method is rigorous in a sense that it directly solves Maxwell's equations on a spatial grid. We assumed a spatial resolution of 12.5 nm. The computational domain in the growth direction is terminated by perfectly matched layers, whereas periodic boundary conditions are applied along the lateral directions. Prior to these calculations we verified that the considered substrates are sufficiently large such that this periodicity does not affect the conclusions. From the calculated field distribution the absorption can be obtained as described in literature [19]. Basically it only requires the integration of the divergence of the Poynting vector over the spatial domain occupied by the a-Si:H. The absorption enhancement is finally calculated by normalizing it to the absorption in a solar cell with the same thickness of the a-Si:H layer and flat interfaces. To quantify the scattering properties

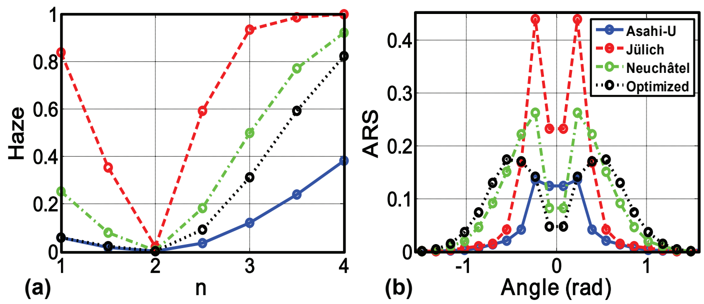

Fig. 2. (a) Haze for different substrates where the surface separates TCO and a medium whose refractive index n is subject to variation. (b) The ARS function for the different substrates if the adjacent medium has a refractive index of n = 4. Data is rigorously calculated at a wavelength of 633 nm. Lines are only guide to the eyes.

we carried out an extra simulation where it was assumed that the considered surfaces separate a half space with a refractive index of TCO from a half space with varying refractive index. It constitutes a reduction of the thin-film solar cell to only the relevant interface. Using the FDTD method we computed the field upon illuminating it from the TCO side with a linearly polarized plane wave. By Fourier-transforming the field in the second half space the angular distribution of the scattered light in transmission can be obtained. From this angular distribution the haze as well as the angularly resolved scattering (ARS) function can be calculated. For both functions we rely on definitions as documented in literature [20]. We concentrate here only on the wavelength of 633 nm since it will be the design wavelength of the optimized substrate. Moreover, supplementary simulations yielded that the deviations tend to be insignificant among different wavelengths around the absorption edge.

Considering at first the haze as shown in Fig. 2(a), it can be seen that the Jülich substrate has the largest haze, whereas the Asahi-U substrate has the smallest. Results are independent from the refractive index contrast at the interface. The haze close to zero at a refractive index of $n = 2$ is attributed to the disappearance of the optical interface if the index is sufficiently matched to that of TCO. Most notably, the haze of the Jülich substrate is close to unity for $n > 3$, indicating that there is no specularly diffracted light. However, from the ARS, shown in Fig. 2(b) for a medium with $n = 4$ in the second half space, it can be seen that the light is only scattered into a small angular domain around the optical axis. This is detrimental since paraxially propagating light does not exhibit an essentially increased optical path length. The Neuchâtel substrate performs quite better in this respect by scattering light into a much larger

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angular domain.

Both properties can be also deduced from the substrate morphology. The modulation depth of the Jülich substrate is large but the craters are too big to induce a sufficiently large transverse momentum. Hence, the scattered light remains restricted to an angular domain close to the optical axis. It is also reflected in a large *rms* value (160 nm) and a large autocorrelation length (1.4 μm). The autocorrelation length is a measure for a predominant structure size occurring in the sample which dictates the direction into which the light is scattered. Obviously, on the one hand the autocorrelation length should not be too large, since then the scattering angles would be too small. On the other hand, it should also not be too small, because in the limiting case of a vanishing predominant structure size when compared to the effective wavelength the structure is sub-wavelength and the illuminating light will rather experience an effective medium. A similar argumentation can be enforced for the *rms* value. If it is too small the scattering strength is negligible because the induced phase delay is marginal. In contrast, a large rms value is beneficial in general, though beyond a certain value it is pointless to increase it further since the induced phase delay will be randomly distributed between 0 and 2π. By contrast, the required fine details that lead to high spatial frequencies can be found in the Neuchâtel substrate which explains the large angular domain into which light is scattered. Nonetheless, the modulation amplitude of the surface morphology is too low to induce a sufficient phase delay and a portion of light is diffracted into the forward direction, i.e., the light is not diffracted at all. This explains the lower haze in air. The *rms* value and the autocorrelation length of the Neuchâtel substrate amount to 81 nm and 140 nm, respectively. The Asahi-U substrate has an *rms* value which is quite low (35 nm), although the autocorrelation length (160 nm) compares to the Neuchâtel substrate.

From computing the absorption it turned out that the Neuchâtel substrate performs best (see Fig. 3). This conclusion holds for both wavelengths as well as for all considered thicknesses of the a-Si:H layer. Since the haze of the Neuchâtel substrate is smaller than that of the Jülich substrate, the haze as a criterion to predict the absorption enhancement is obviously insufficient. From rational understanding it is hypothesized that it is of primary importance that the substrate strongly scatters the light. The quantity which was traditionally taken into account for this aspect is the normalized integrated ARS spectrum excluding the directly transmitted light, i.e. the haze, denoted here as A_{Int}. However, the substrate shall also scatter into a wide angular domain. To access this property we performed a least square fit of the ARS with a Gaussian function (full width at half maximum σ_{A}) for large scattering angles. It is documented in literature and confirmed by our work (not shown) that such a functional dependency describes the experimental observations [20]. With the assumption that the absorption has to be proportional to both parameters A_{Int} and σ_{A}, the only open issue was to adjust their exponents to relate this figure of merit to the absorption enhancement. The ultimate goal was to achieve largely a linear functional dependency. This was possible by assuming that the absorption depends linearly on the product $A_{\mathrm{Int}}\sigma_{\mathrm{A}}^2$. To illustrate this linear behaviour, Figs. 3(a) and (b) show the absorption enhancement as a function of this parameter, whereas each parameter signifies a certain substrate as indicated in Fig. 3(b). This criterion, however, is only a qualitative one, since it neglects certain aspects. First of all, the absorption enhancement cannot grow infinitely, because the overall absorption cannot exceed 100%. Therefore, the absorption enhancement has to saturate which is not reflected by the above criterion. Moreover, especially for small values of $A_{\mathrm{Int}}\sigma_{\mathrm{A}}^2$ it was not always possible to relate it to the absorption enhancement. Despite these shortcomings the major tendency can be predicted, simply, because the formulation of the criterion relies on basic considerations which naturally hold. Roughly the same functional dependency holds if the second half space is assumed to be air instead of the intermediate reflector, as can be seen in Fig. 3 (c).

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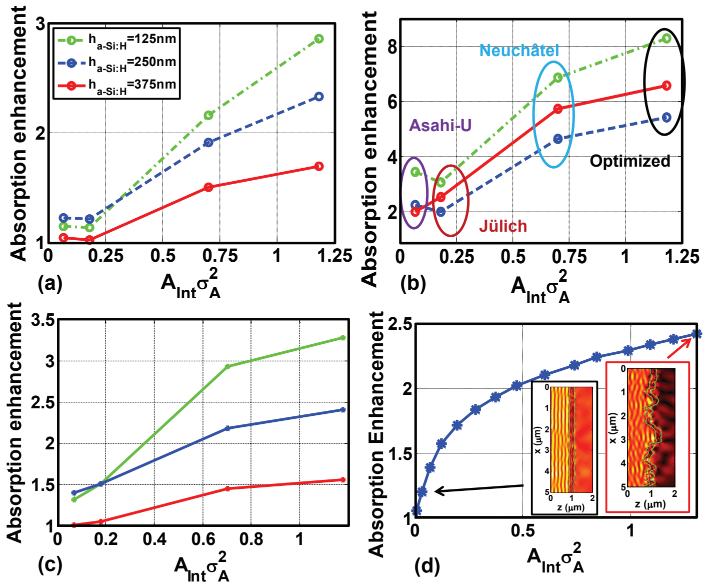

Fig. 3. Absorption enhancement of a thin-film solar cell with different textured surfaces. Results for a wavelength of 633 nm are shown in (a) and for a wavelength of 720 nm in (b). The geometry of the solar cell is described in the main body of the text. Three different thicknesses for the a-Si:H layers were considered and the different quality factors $A_{Int}\sigma_A^2$ signify different substrates. The link between the quality factor and the substrate is shown in (b). The absorption enhancement at a wavelength of 633 nm is furthermore shown in (c) where it was assumed that the medium of the infinite half space into which the light is scattered is air. The lines in (a)-(c) are only guide to the eyes which connect the different data points. In (d) we finally show the results of the absorption enhancement depending on the merit criterion. The data points were generated by scaling the height of the Neuchâtel profile and evaluating the scattering response against air as the medium of the second infinite half space and its ability to enhance the absorption at 633 nm for a 250 nm a-Si:H thickness for each texture individually .

To further quantify the validity of the merit criterion we took advantage of the computational approach and modified a selected profile in a deterministic manner, i.e., we changed the height of the Neuchâtel profile by a factor between 0 and 1.5. By this we generated a large number of different substrates with different scattering properties. We evaluated for each substrate the scattering properties and its ability to enhance the absorption in a 250 nm a-Si:H film at 633 nm. The material of the infinite half space into which the light is transmitted is here assumed to be air. Results are shown in Fig. 3 (d). The functional dependency as previously discussed can be fully disclosed, i.e., for small values of the merit criterion the absorption enhancement scales largely linear whereas it tends to saturate for higher values of the merit criterion.

The comparative aspect of this work ends here. It was found that the Neuchâtel substrate outperforms the other substrates since it suitably combines both properties: a strong scattering and scattering into a wide angular domain into a medium with a high refractive index. It has to be stressed that to evaluate the scattering properties one not necessarily needs to use resource consuming rigorous simulations. In the past various methods were developed that can be used for this purpose and they provide largely comparable results [22]. However, the ability to use such methods to compute the local absorption is less developed, but to decide which surface seems to be appropriate they may be exploited.

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Beyond such a descriptive contribution, the question naturally remains whether substrates can be designed that operate even more efficiently besides a mere scaling as shown in Fig. 3 (d). To find the pertinent surface profile, we used the thin-element and the scalar approximation as well as the iterative Fourier transform algorithm to optimize a phase-only transmission profile of a surface that shall scatter the light into a large angular domain with a Gaussian dependency.

We selected a surface profile which is characterized by a larger product $A_{Int}\sigma_A^2$ than all above substrates. The entire optimization procedure is identical to the one described in Ref. 21 for a 1D surface profile. The optimized phase profile was translated into a geometrical profile assuming 633 nm as the design wavelength and a refractive index of n = 4 for the medium of the second half space. Although scalar theory is inadequate to describe the scattering properties of such a surface, this optimization served as an initial guess whereby the final optical properties were evaluated by the FDTD method. From Fig. 2(a) it can be seen that the haze of the optimized surface is lower than that of the Neuchâtel substrate, but Fig. 2(b) indicates that a considerable amount of light is scattered into a domain with larger spatial frequencies. This is anticipated to be advantageous since the absorption enhancement depends on the square of this angular spectral width. This can be seen from Fig. 3. The optimized surface profile outperforms the existing substrates and shows the largest absorption enhancement with an absolute absorption of 83.1% at 633 nm and 10.5% at 720 nm within the 375-nm-thick a-Si:H layer. The *rms* value and the autocorrelation length of this artificial substrate are 88 and 200 nm, respectively, only a little above the values of the Neuchâtel substrate.

3. Conclusions

In conclusion, we have comparatively investigated various available textured substrates with regard to absorption enhancement in the top cell of thin-film tandem solar cells made of a-Si:H. It was shown that the structure should exhibit two features. The ideal substrate should scatter both strongly and into a wide angular domain. Based on this understanding we have related the scattering properties of various substrates to their respective absorption enhancement. An almost linear dependence of this enhancement on the product $A_{Int}\sigma_A^2$ has been identified. Both parameters were evaluated for a scattering response of the substrate against a high refractive index medium. A comparable correlation between scattering properties against air and absorption enhancement could not be verified, e.g., structures with a larger merit criterion that was evaluated from the scattering response of the interface separating TCO and air had a lower absorption enhancement. This suggests that the scattering response of the ZnO-interface against the active layer of the solar cell is of primary importance. We even went one step further and proposed an optimized surface profile for the randomly textured interface which outperforms the available substrates. Although its shape is quite complex, surfaces with such morphologies may be realized by advanced nano-fabrication techniques such as nano-imprint lithography. If the benefit of such surfaces is sufficiently large, they will certainly appear in commercially available solar cells in the near future. We finally wish to mention that the introduced merit criterion is not necessarily the only one which may be useful. Other criteria that are deduced from the scattering response can be applied as well and it remains open to the future to evaluate their predictive power.

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