MASTER'S DEGREE PROGRAMME

Eco-Energy Systems

Modification of transparent nanostructures for light scattering in solar cells

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by

Peter Hirner

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Project supervisors

Dr.in Angelika Basch and Prof. Maan Alkaisi

ABSTRACT

The power conversion efficiency (PCE) of solar cells depends on many factors. In general, the fabrication process, material quality and the structural design are critical parameters to achieve high PCE. For the most frequently used polycrystalline silicon solar cells (Si-SC) it can be said that as the cell thickness increases up to 100 µm the PCE will rises until the intrinsic limit of ~29% is reached. [1] This is achieved as light penetrates an extended path through the solar cell with the probability of light absorption increasing.

In the same manner the effect of extending the path for absorption should be achieved by scattering of the light. Therefore, the top surface is fitted with a nanostructure. This scatters the incident light in a flat angle towards the photoactive layer. In that way the light absorption path is extended and has the same effect as the thickening of the photoactive layer like for Si-SC.

Such light scattering nanostructures at the top surface can be used for different solar cell types. Because thin film- or perovskite- solar cells are using a thinner photoactive layer, this technique is particularly suitable since the nanostructure is on the top layer, which is also the encapsulant, instead of the photoactive layer.

The light scattering should be enhanced by embedding 10 to 20 nm sized SiO_2-beads into a transparent UV-cured polymer (OrmoStamp). While the internal reflection should increase too due to the superior nanopyramid shape of the surface.

Shown in the AFM and SEM images, both types of pyramids are perfectly aligned and uniform in shape when comparing the morphology of the DCM/SiO_2- OrmoStamp mixture and the just OrmoStamp. While the direct transmittance measurements of the DCM/SiO_2- OrmoStamp pyramids show a higher transmittance than just OrmoStamp pyramids. This was contrary to the expected results. The reasons for this were explored and considered in more detail in accordance with the literature on light guiding.

In conclusion, the direct transmittance of nanostructures varies as SiO_2-beads are added into the transparent UV-curable polymer. Furthermore, the influence of these mixture on the transmission behaviour could be shown by changing the fabrication process and therefore the superior nanostructure from imprint to spin coating.

CONTENTS

1 INTRODUCTION

Modifying the scattering behaviour of the top layer for solar cells is the focus of this research investigation. The aim is to create nanostructures on the glass substrate which influence the direct transmittance. Details will be explained in the following subchapters.

1.1 RESEARCH OBJECTIVES

This thesis aims to observe the effect on the transmittance due to the modified nanostructures. By applying nanobeads the reflection should be reduced, and the light scattering might be increased. A valuable by-product of the top surface is the self-cleaning effect that is maintained by the nanostructures which should not be impaired by the added nanobeads.

It is the desired aim to reduce the reflection of the solar cell top surface, in order to increase transmitted light and hence increasing the probability of photonic conversion. The objective of increasing light scattering is to generate multiple reflections in the solar cell. In this way the probability that light is absorbed by the photoactive material is increased as is explained in the following paragraph.

1.2 PRESENT WORK

The possibility to increase the path length of incident light by texturing the surface with nanobeads embedded into a nanopyramid structure is part of this investigation. According to additional internal refraction between the nanopyramids made of UV-cured polymer and the nanobeads of silicon dioxide (SiO_2), see chapter 2.3. Furthermore, the small size of the nanobeads of 10 to 20 nm should lead to a scattering effect as described in chapter 2.4. By comparing the light path in the nanopyramids in Figure 1 with the textured nanopyramids in Figure 2 the desired scattering effect is shown.

The pyramids with pattern or pitch sizes (Λ) are demonstrated as v-grooves. The path of the light can be traced, indicated by the black arrows. In Figure 1 and in Figure 2 the incident wavelength (λ) is similar to pattern sizes (Λ). While Figure 2 additionally got SiO_2-beads inside the pattern structure, which should enhance the scattering effect.

As the SiO_2-beads are in range size below 1/10 of the applied wavelengths (λ) the Rayleigh scattering according to chapter 2.4 might affect the light trapping behaviour.

So, the idea is to combine the two scattering effects as described in chapter 2.2 for Case λ≈Λ and Case λ<<Λ. Additionally the reflection should be reduced and the refraction should be enhanced due to the implemented SiO_2-beads, as described in chapter 2.1.
Such new investigations to fabricate path length enhancement features are a potential application for solar cells or other light guiding devices.

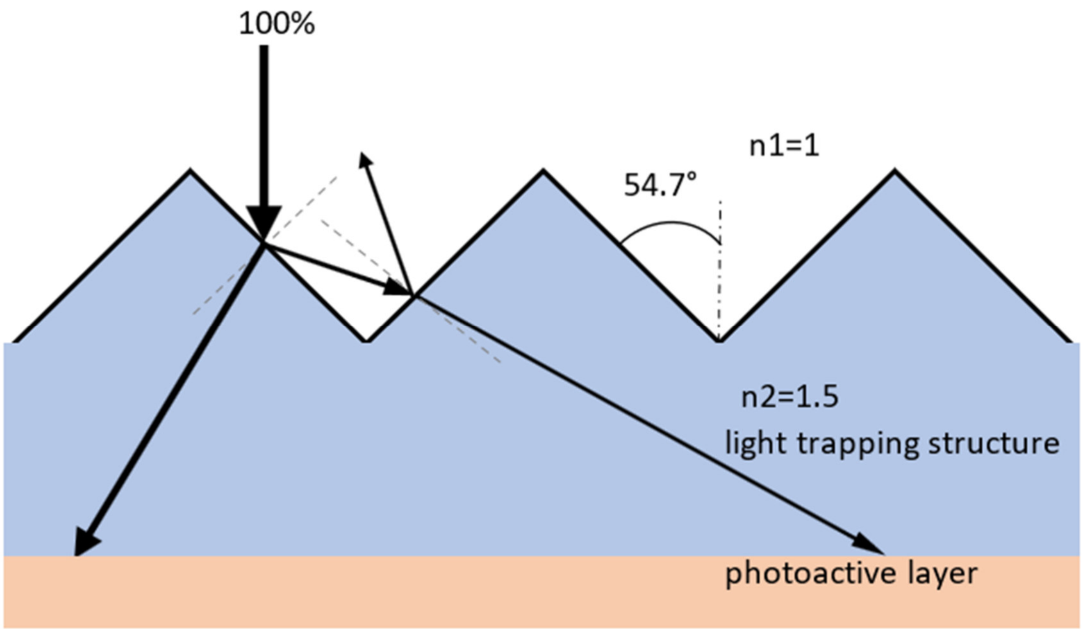


Figure 1: Schematic diagram of light paths in a V-groove pattern causes multiple refraction and reflection within the light trapping structure.

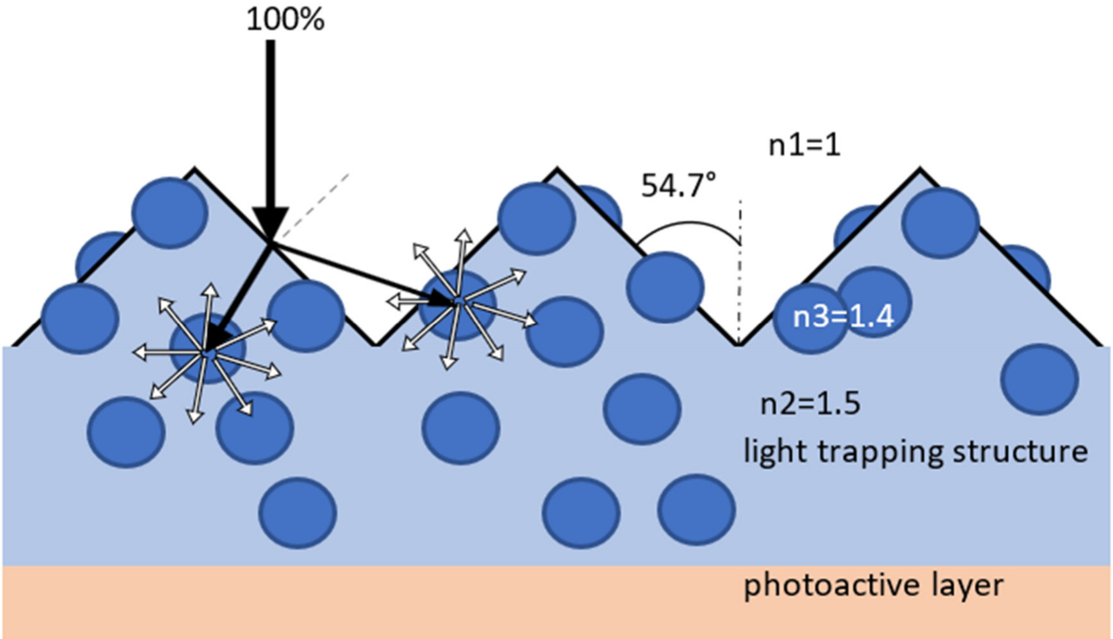


Figure 2: Schematic diagram of light paths in a V-groove pattern which lead to multiple reflection while the SiO_2-bead textures ensures the scattering within the light trapping structure.

1.3 Need of Solar Cell Research

As the energy demand worldwide is increasing and energy transition from fossil fuels to renewable energy sources is an ongoing process of the next decades it is also important to increase the efficiency of these more environmental-friendly power plants, Figure 3 and Figure 4.

“Note: In the Stated Policies Scenario (STEPS), the global economy returns to its pre-Covid-19 level in 2021,but remains around 7% smaller over the longer term than projected in the WEO-2019.

The Sustainable Development Scenario (SDS) is based on the same economic and public health outlook as the STEPS but puts energy systems on a different trajectory, where CO_2 emissions fall to under 27 Gt in 2030.” [2] As Figure 3 shows the level of CO_2 emissions (orange line) will not reach the pre-crisis-level of 2019 before 2027, while the energy demand (blue line) will be recovered in 2023. This is mainly due to the huge growth of renewable energy source and the decreasing demand of coal needed for energy.

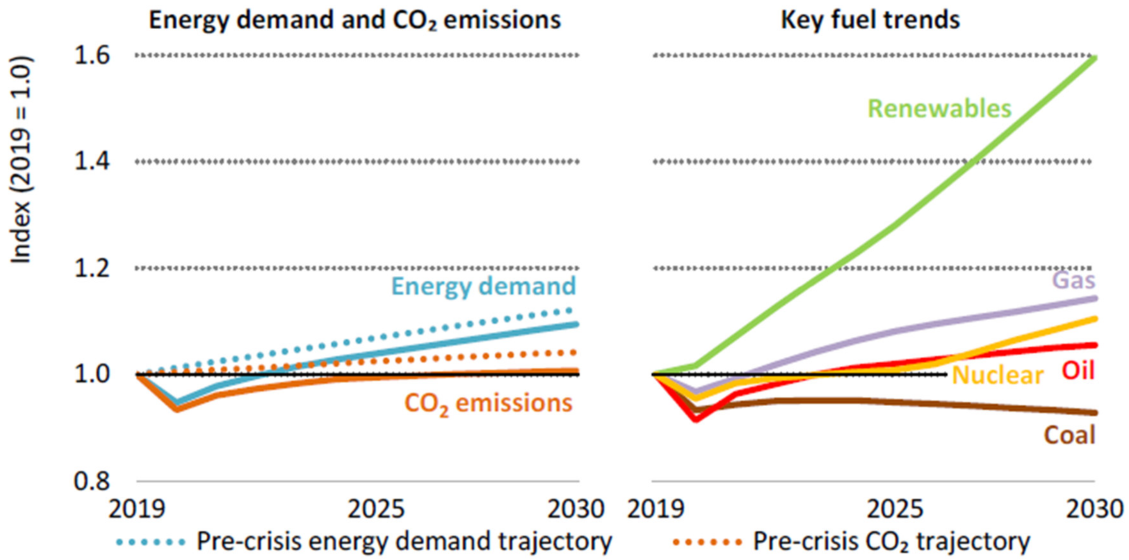


Figure 3: Energy demand returns to pre-pandemic levels in early 2023, but CO_2 emissions do not until 2027 due to resilient growth in renewables and reduced coal demand. Graphic taken from [2].

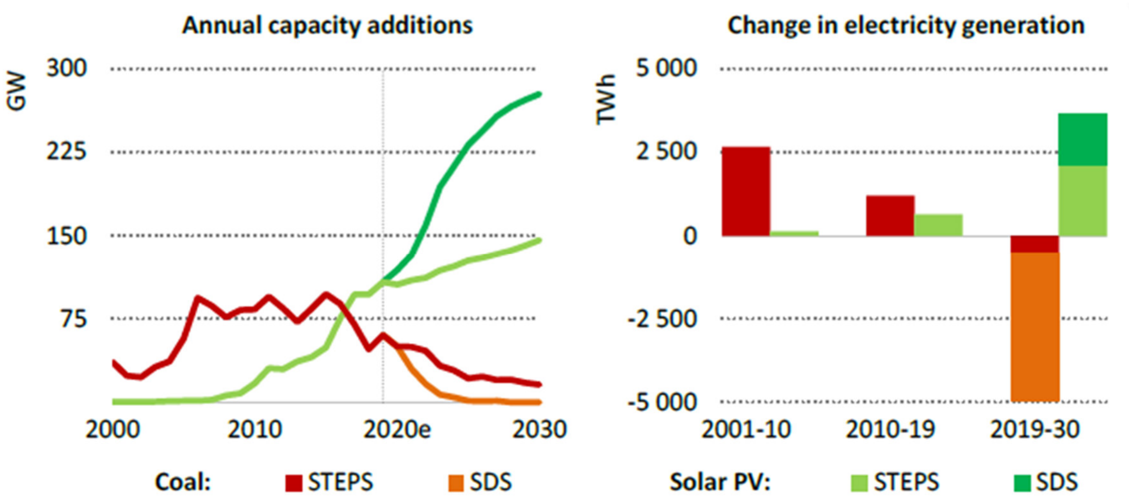


Figure 4: Solar PV becomes a preeminent force in electricity supply in the STEPS due to falling costs and policy support, but the potential is given for much more rapid growth. Graphic taken from [2].

As the global solar PV capacity has increased almost 20 - fold over the last decade, the STEPS-scenario indicates it will set to triple over the coming decade. “Targeted policies in over 130 countries and technology cost gains have been key drivers of this expansion, and they have helped in turn to bring down the cost of financing, which accounts for 20-50% of the overall levelized cost of generation for new utility-scale solar PV projects.” [2]

One of the prominent solar cell technologies are perovskite solar cells (SC) or a combination of perovskite and crystalline Silicon SC. While Si-SC are indirect semiconductors with a band gap of about 1.1 eV, the perovskite has a direct band gap, which is engineerable within the range of 1 to 3 eV as Hu *et al.* reported. [3] A combination of Si and perovskite leads to a wide-bandgap so called tandem SC. As for Si-SC the biggest share of costs is the Si-ingot material, while for perovskite SC the necessary material demand is lower, and the fabrication process can be realised in a simple wet chemistry process. Next to the dye-SC the perovskite-SC are still under investigation in research and new fabrication methods of printing or spraying these SC have laboratory status. The use of lead components in SC manufacturing is currently possible in the European Union because PV modules are currently not covered by the Regulation on the Avoidance of Lead in the Manufacture of Electronic Equipment (RoHS).

Solar energy is the initial source of all renewable energy on earth with exception of geothermal and tide energy. The direct conversion from solar irradiation to electrical energy without intermediate conversion into other energy states is the desired aim of many scientists. This is feasible, however with non-negligible energy losses, like recombination and absorption or the losses prior the radiation reaches the photoactive layer. Or the optical losses like reflection caused by the top surface of a solar module. This top surface is also often the encapsulation and protection layer of the solar cells beneath.

1.4 MOTIVATION AND VISON

The power conversion efficiency (PCE) as a benchmark for the solar cell performance is dependent on many factors in the fabrication process, structure type and material quality.

A large number of structure types exists, for the most frequently used polycrystalline silicon solar cells (Si-SC). Whereas in general, for the crystalline (c-Si) with an indirect bandgap, it can be said that as the cell thickness and thereby the photoactive layer increases the PCE rises. This is mainly caused by the higher probability of light absorption, which increases the generation of electron-hole pairs too. In that case, light penetrates an extended path through the solar cell. Very simplified it means that as deeper the light can penetrate through photoactive layer the higher the electron-hole generation, followed by a higher electrical energy harvesting, is.

Note, this generalization does not consider recombination and other effects, which are stronger with thickening photoactive layer and therefore have a performance-reducing effect at the same time.

In the same manner the effect of extending the path for absorption should be achieved by scattering of light. Therefore, the top surface scatters the incident light in a flat angle through the photoactive layer. In that way the light absorption path gets extended and has the same effect as thickening the photoactive layer. In this case the mentioned negative performance-reducing effects do not have so much influence.

Such light scattering can be achieved by the top surface and can be used for various of different solar cell types. This is also true for the so-called third generation of solar cells. The most prominent representatives are currently the perovskite solar cells, which are most commonly a hybrid of organic and inorganic material. The development of this technology is subject to high expectations and a large amount of research is ongoing.

Further research studies concerning this solar cell type could bring this product to market in the upcoming years. Although here, too, costs and resources can be saved by using a thinner photoactive layer. In addition, light scattering could increase the power conversion efficiency of the perovskite solar cells or other solar cell structures.

As described from J. H. Noh *et al.*, the absorption of a certain wavelength range can be adjusted for perovskite solar cell (PSC), also known as wavelength selectivity. [4] This feature could bring benefits for fruit growing in greenhouses. Thereby semi-transparent solar cells are applied as roofing and walls instead of ordinary glass of such fruit-growing farms.

Here the necessary wavelengths for growing vegetables and fruits are transmitted throw the solar cell. For plant growing unusable wavelengths are absorbed by the semi-transparent solar cells and converted into electrical energy. This double use of the greenhouse roofs shows the benefit of reducing area and material consumption. Moreover, it cuts the cost for the construction of such a combination of greenhouses and solar cell technology.

A crucial point to enable the combined electrical power generation and plant growth is that the very top layer of the solar cell will trap as much light as possible. Therefore, the direct reflection of the incident light needs to be reduced and the light scattering has to be increased.

The light scattering nanostructures on solar cells have the potential of improving the amount of incident light reaching the photoactive absorption layer.

2 Light Guiding

In 1960 Dale *et al.* demonstrated that front surface texturing can increase the efficiency of solar cells. [5] During that time the focus in research laid mainly on silicon solar cells (Si-SC) than on other solar cell types, driven by the semiconductor industry as well as the space program. In the 21st century a change of research projects can be observed, since 60% of the filed patents from the top inventor companies for SC contain thin film and emerging SC like organic-SC or perovskite-SC technologies. The increase of scientific publications and patents demonstrates the advancement of SC-technology into a significant research sector. Measured in terms of the number of patents filed in 2014. [6].

A promising technique for SC, which lead to higher power conversion efficiency (PCE) is the reduction of reflectivity and light trapping, as outlined in the following.
The purpose of reflectivity reduction is obvious, as the radiation, which is reflected back towards its source is lost. The second optical improvement is the light trapping, which is maintained by increasing the internal path length of incident light, in order to increase the possibility of absorption by the photoactive area. For Si-SC, where silicon is a semiconductor with indirect band gap the absorption of radiation with photon energies close to band gap energy (E_{Gap}) is weak. While the extending of the internal path length promotes the absorption. This can lead to an advantage of lesser material use since SC's can be produced thinner, which reduces material costs. For thin film- or perovskite SC this extended path length is mandatory too for an increase of absorption in the photoactive layer.

2.1 Reflection reduction

One way to reduce the front surface reflection is the destructive interference shown in Figure 5 demonstrates the two reflections are 180° angel (π) out of phase and cancel each other.

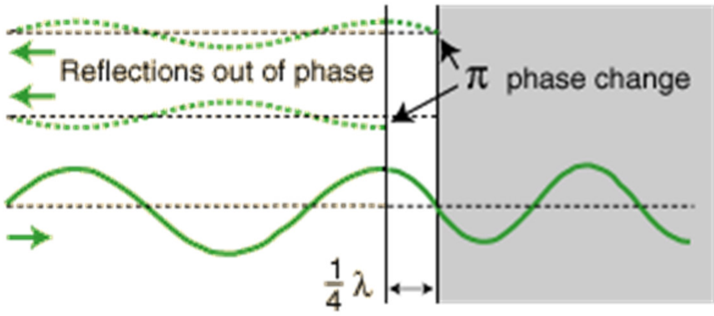


Figure 5: Anti-reflective coatings work by producing two reflections which interfere destructively with each other. Graphic taken from [7].

As shown in Figure 6, the two red arrows labelled as I and II demonstrate the reflection of the waves which are out of phase and therefore causing a destructive interference. Note that the infinite subsequent reflection between the interfaces is neglected.

Some boundary condition must be fulfilled to ensure this destructive interference, since this holds true for just a particular wavelength. The layer thickness must be λ/4 or a multiple of uneven numbers. The refractive index must be higher than the refractive index of the previous layer in which the radiation transmits. In that way the light penetrates through the layers to ensure a phase shift on the top surface and on the interface between the antireflection coating (ARC) and the substrate, meaning that $n_1 < n_{ARC} < n_S$. The intensity of the reflected waves has to be equal, this can be ensured by Fresnel`s equations. The ideal refractive index n_{ARC} of the ARC can be calculated according to Equation 1 and the thickness (d) of the ARC-layer can be determined by Equation 2. This additional layer reduces the surface reflection but does not contribute to an increase of the internal path length.

Equation 1

$$n_{ARC} = \sqrt{n_1 \cdot n_s}$$

Equation 2

$$d = \frac{\lambda}{4 \cdot n_{ARC}}$$

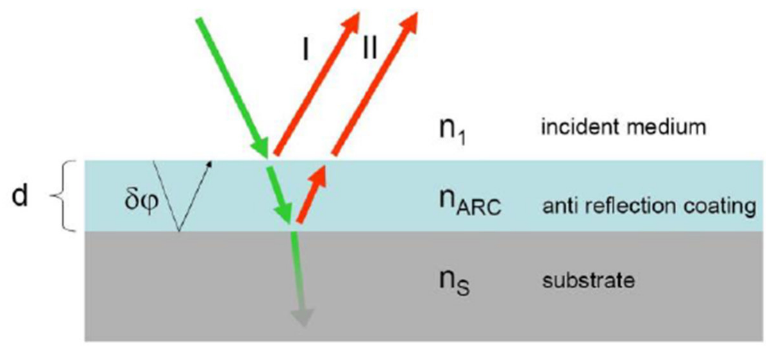


Figure 6: Sketch of a single layer antireflection coating. Graphic taken from [8].

2.2 PATH LENGTH ENHANCEMENT

An increase of the internal path length can promote the increase of absorption in the photoactive layer of a SC. For example to accomplish this, the perpendicular incident light is guided through the structure of the top surface layer in a low tilted angel towards the photoactive layer. This guiding effect of the structures is also called scattering.
To guide the incident light with structures such as pyramids, the ratio of feature size to wavelength plays a mandatory role. The effect of three cases of varying ratios is shown in Figure 7 and can be described by geometric optics in the following paragraph showing a V-groove pattern of different pattern period Λ and pattern size.

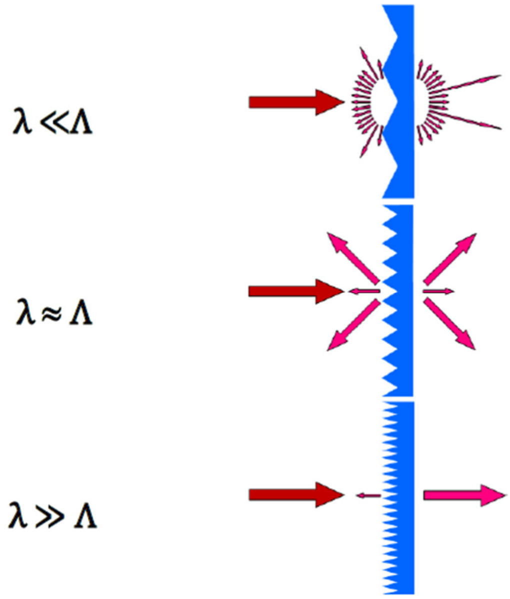


Figure 7: Schematic view of optical effects caused by periodically textured surfaces of varying special frequency for a given wavelength. Graphic taken from [9].

- Case λ<<Λ:

As the pattern period Λ and size of the v-groove structure is much larger than the wavelength λ, a variety of diffractions occurs. Depending on the angels of the groove structure a certain refraction angle can be achieved, which causes an internal path length enhancement. In addition, multiple reflections occur due to the surface structure which reduces the reflection again, as depicted in Figure 8. [10]

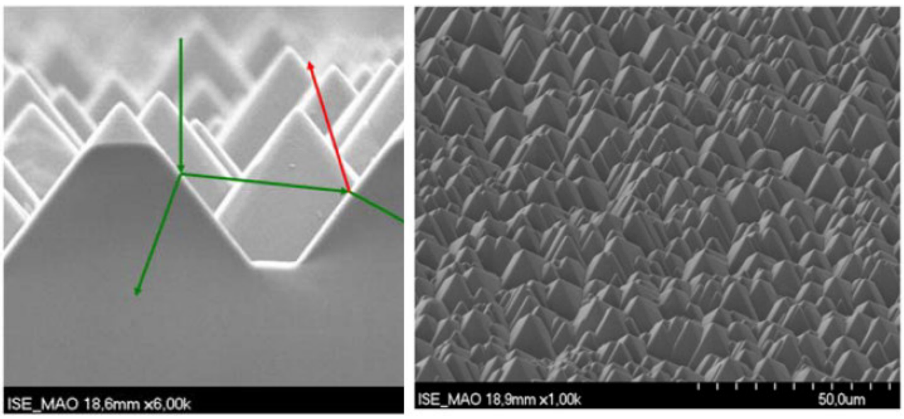


Figure 8: Scanning electron microscope (SEM) images of Random Pyramids on monocrystalline silicon. In the cross-sectional view light-rays are sketched to demonstrate multiple reflections. In the tilted view the different sizes of pyramids are shown. Graphic taken from [8].

- Case $\lambda \approx \Lambda$:

The reflection and transmission properties of the surface structure depends on the structure size, if the incident wavelength λ is similar to the pattern sizes Λ. The structure leads to interference effects and therefore to a reflection reduction. Furthermore, it determines the path length enhancement. For the periodic pattern the interaction is usually just valid for a narrow spectral range due to the structure dimensions. [10]

- Case $\lambda >> \Lambda$:

If the periodic feature size is much smaller than the incident light the effective medium theories can be used to describe an effective reflective index. The behaviour of the incident light with the wavelength λ can be understood as the structure with dimensions Λ in subwavelength range, which cannot be resolved by the wavelength direct the incident wavelength. Therefore, the incident light transmits through the structure and is also partially reflected similar to a flat surface. Such small features determine the optical effect on transmission and reflection, but do not contribute to light guiding. [10]

2.3 Refraction and Reflection of Light

In general, incident light will be partially reflected and partially transmitted as a refracted ray by the top surface. [7] If the surface got a pattern the reflective part could be directed again towards the surface, which would lead to an additional refraction and lowers the whole share of reflectivity.

In the example of v-grooves, as shown in Figure 9, where the incident wavelength λ is similar to the pattern sizes Λ or pitch, the path of the light can be traced, indicated by the blue arrows.

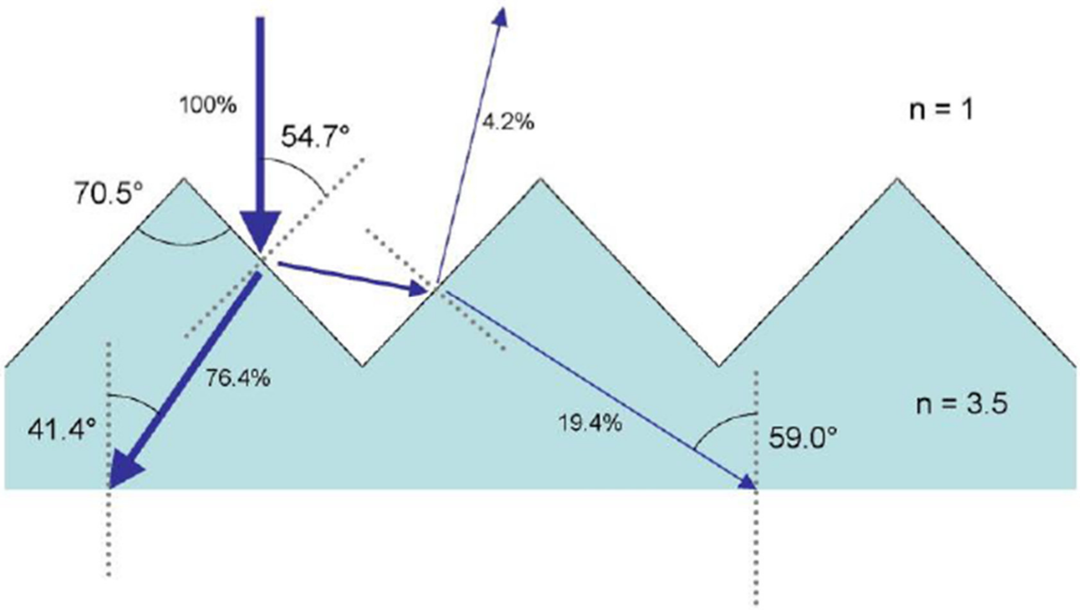


Figure 9: Schematic diagram of light paths in a V-groove pattern for the case of a double reflection on opposite surfaces. Angles correspond to <111> planes in a <100> silicon wafer. The figures given represent the case for light with a wavelength of 1040 nm and a solar cell with an antireflection coating of silicon oxide with a thickness of 105 nm. Graphic taken from [8].

The angle of reflection and refraction of the light can be derived from Fermat's principle. Called the law of reflection the angle of incidence light on a plane surface is equal to the angle of reflection. [7] Figure 9 also shows the refraction of light when it passes from a fast medium to a slow medium and bends the light ray toward the normal to the interface between the two media. Depending on the indices of refraction (n) the two media to the direction of the incident light in the slow medium can be determined by Snell's Law, see Equation 3. [7]

Equation 3

$$\frac{n_1}{n_2} = \frac{\sin\theta_2}{\sin\theta_1}$$

2.4 Rayleigh Scattering

Rayleigh refers primarily to the scattering of light by a spherical particle, whose diameter is less than about 1/10th the wavelength (λ) of the incident light.
Figure 10 demonstrates the dependency of the scattering intensity on the direction of the incident light, which is one-half the forward intensity at a right angle direction. [11]

Rayleigh scattering can be considered to be elastic scattering, since the photon energies of the scattered photons are not changed. Whereas scattering in which the scattered photons have either a higher or lower photon energy is called Raman scattering, whose share of intensity is lower and, therefore, outside the scope of this thesis. [7]

2.5 Mie Scattering

Mie scattering is predominant for particle sizes similar to the wavelength (λ). This scattering produces an intensity pattern like an antenna lobe, whereas for larger particles a sharper and more intense forward lobe can be seen in Figure 10. [11]

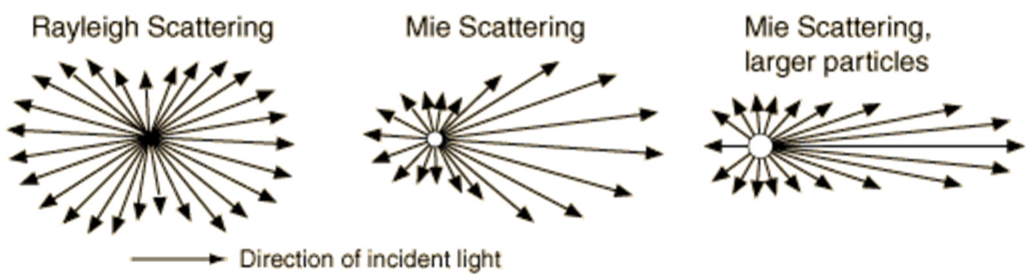
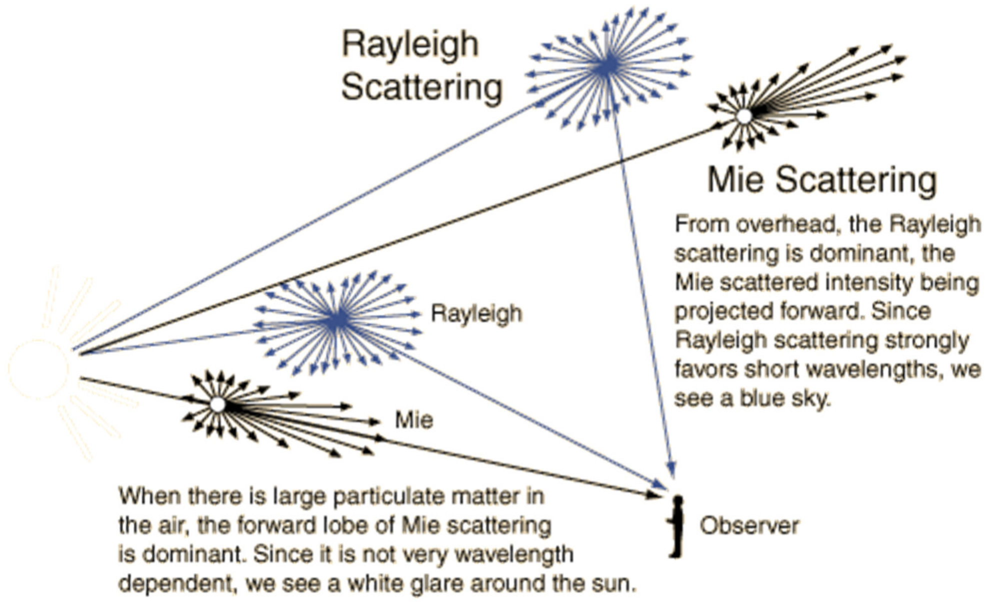


Figure 10: Scattering behaviour depending on particle size. Rayleigh scattering for molecules and very tiny particles smaller than 1 /10 λ, scatters backward and towards the incident light direction. Mie Scattering predominated for larger particles with similar size as the incident light with more scattering toward the direction of the incident light. Graphic taken from [7].

An interesting side note on the distinction between these two scattering effects is briefly explained below. Unlike Rayleigh scattering, Mie scattering is not strongly wavelength dependent. Furthermore, Figure 11 shows the overruling effect of Mie scattering if a lot



of particulate material is present in the air, which causes the almost white glare around the sun.

Figure 11: Dominating scattering effects of Rayleigh or Mie Scattering define what the Observer detects. [7]

2.6 NANOSTRUCTURES FOR SC

A continuous development in the light trapping technology can be observed in the recent years indicated by the increase of publications. Industrialized common techniques for silicon solar cells (Si-SC) are the pyramid structures in upright or inverted shape [12], [13] aligned or randomly arranged or similar textures. [14], [15], [16] These structures for light trapping usually have sizes between 3 to 10 µm. [17] As for third generation solar cells the usual thickness of the photoactive material is in the same range of thickness. [17] These surface structures are unpracticable to apply on the photoactive material itself, e.g. perovskite- or thin film-SC.

These third generation SC's are a promising renewable energy source which should reduce the material consumption and cut costs as well. But to lead to similar effect of light trapping as it arises with microstructures for Si-SC, it also shows beneficial impact on perovskite solar cells while using nanostructures, as reported in [18] and [19].
Whereas these nanopyramids and some promising others, such as periodic grating structures as reported in [20] and [21] also show light trapping behaviour.
Similar structures like photonic crystal structures [22], nanowires [23], random scattering surfaces [24], [25] and plasmonic structures [26], [27] are the most prominent methods for light trapping in solar cells.

Mainly two effects are crucial to improve the nanostructures for Si-SC. Firstly, the reduction of reflectance over a broad wavelength range and secondly, the improvement in the substrate quality, with fewer imperfections due to lower etching depth in nano-size region. While features in micron scales require deep etching and will generate defects in the material. [28]
For crystalline Si-SC such light trapping structures are implemented directly on the photoactive layer, while for thin film-, dye- and perovskite- SC this is not achievable. On one hand, the layer thicknesses are too thin and on the other hand, these materials are not resistant enough for dry- or wet- etching processes, which are common in structure fabrication for Si-SC's. [19]
One option to generate similar effects is to place a top layer with such a light trapping structure on top of the photoactive material. The fabrication of this structured top layer is described in chapter 1.2.
As the analytical results from Yu *et al.* show that light trapping can be significantly enhanced while embedding low-index absorptive inclusions in a high-index base material. [29] OrmoStamp in cured state as the base material has a refractive index of 1.505 at 1000 nm and of 1.535 at 400 nm as the material with the higher refractive index. While the embedded nanoparticles of SiO_2-beads with a size of 10 to 20 nm have the lower refractive between 1.450 at 1000 nm and of 1.470 at 400 nm.

As Yu *et al.* write “By roughening the semiconductor–air interface […], one randomizes the light propagation direction inside the material. The effect of total internal reflection results in a much longer propagation distance inside the material and hence a substantial absorption enhancement.” [29] This would lead to the upper absorption enhancement factor limit. While using a shallow grating on the surface of a relative thick medium, which is significantly thicker than half of the wavelength. In these size ranges, of the so called nanophotonic regime, this limit of the absorption enhancement factor can be exceeded, beyond the Yablonovitch limit. [29]

To increase the share of the absorbed wavelengths in the thin photoactive layer it is necessary to guide the wavelengths. This guidance is achieved through increasing the optical path length by scattering of the light and simultaneously reducing the reflection caused by the top surface.

Nanostructures, such as pyramids, can be used to fulfil this. Furthermore, recent studies showed that if the pyramids got doped with gold nanoclusters the light absorbance at the shorter wavelength region increases. [19]

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