

Analysis of the Scattering Properties of Textured TCO Structures for Thin Film Silicon Solar Cells

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ABSTRACT: The texture of transparent conducting oxides (TCO) plays an important role for the performance of amorphous (a-Si:H) and microcrystalline (μ c-Si:H) thin-film silicon solar cells. Different TCO morphologies and feature sizes lead to different light in-coupling, light trapping, and absorbance in the cell. Induced by TCO scattering, the light path is enhanced which results in a higher absorbance in the silicon layer. We have investigated the scattering properties of sputtered and post-etched ZnO substrates. Varying the etching time results in different feature sizes. To find relations between the surface topology and the scattering behaviour we developed a model, which allows for an analysis of the surface morphology and the simulation of the angle resolved scattering behaviour of randomly textured TCOs. The model uses atomic force microscope data and is based on geometric optics. In this article experimental and simulated results are analysed and discussed.

Keywords: thin film silicon solar cells, TCO structure, scattering

1 INTRODUCTION

For high performance thin-film silicon solar cells transparent conductive front contacts (TCO) are widely used. TCO texturing leads to a significant enhancement of the short circuit current compared to cells made on smooth TCO substrates [1-3]. The texturing can be realized either directly by APCVD (LPCVD) growth of SnO_2 (ZnO) or by a post-etching step of sputtered ZnO. For the light path enhancement which leads to a higher absorption in the silicon layer, (i) the amount of scattered light - the haze - and (ii) the angle distribution of scattered light are of great importance. These two key parameters are correlated with the TCO morphology and feature sizes. Although a lot of work has been done in this area [4-7] clear optimization criteria are missing.

We have investigated in detail the optical scattering properties of sputtered, Al-doped ZnO substrates on glass, which are textured by a post-etching step. Using different etching times the roughness and lateral feature sizes can be varied over a broad range [4, 7]. To gain more insight in how the surface structure is correlated with the scattering properties, we developed a model based on geometric optics. In section 2 we describe the structure of post-etched TCO substrates and their scattering properties, as a function of the etching time. In section 3 we introduce our geometric optics model and we use this model to determine the inclination angles and to analyse the ARS scattering behaviour of the post-etched TCO substrates. Although our simple geometric optics model can account for the observed general trends there remain also deviations between the simulated and measured scattering behaviour. In section 4 we will discuss possible explanations of the observed differences and in section 5 the location of inclination angles on the surface and its relation to the ARS is analysed. Finally, conclusions about the scattering behaviour of nano-textured TCO structures are given.

2 ZNO ETCHING SERIES

During the etching process crater-like structures are immediately formed. With increasing etching time, the initially small craters accrete by and by to larger ones, increasing both in depth and lateral size. Figure 1 (left) shows the AFM scan of a 3 seconds etched substrate. Only a few small craters are randomly distributed over an otherwise almost flat substrate. For etching times shorter than 20 seconds, individual craters with lateral structure sizes between 100 nm and 1.2 μm are formed. Substrates with longer etching times (≥ 20 s) show a more homogenous randomly textured surface. Large craters with lateral feature sizes between 1 and 3 μm are located directly next to each other and are not separated by flat regions (Fig. 1, right).

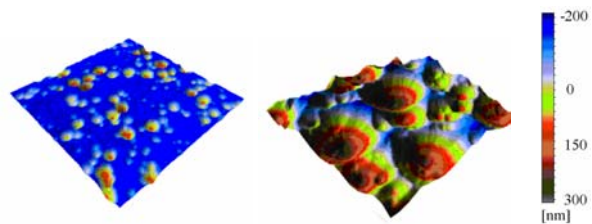


Figure 1: 7 x 7 μm AFM scans of 3 seconds etched (left) and 70 seconds etched (right) ZnO substrate.

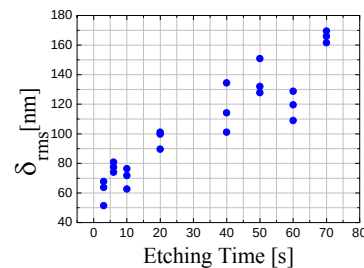


Figure 2: Roughness of ZnO substrates as a function of the etching time.

The etching time was limited to 70 seconds as then the crater bottom already reaches the glass substrate. The etching time determines the feature sizes of the substrate. Short

etching times result in small feature sizes, while long etching times result in large feature sizes. The root mean square roughness δ_{rms} increases almost linearly with etching time (Fig. 2). The value of δ_{rms} varies between 61 nm (3 seconds) and 166 nm (70 seconds).

The change of δ_{rms} is clearly reflected in the scattering behaviour of the ZnO sample (Fig. 3). The haze is defined as the ratio between diffuse transmitted and total transmitted light. A decrease of the haze with the wavelength and an increase with the etching time is observed. This behaviour can be described by the scalar scattering theory [8]: $H(\delta_{\text{rms}}, \lambda) = 1 - \exp(-4 \pi \cdot f(\lambda) \cdot \delta_{\text{rms}} / \lambda)^2$ with $f(\lambda) = c \cdot (n_{\text{TCO}}(\lambda) - n_{\text{Air}})$ and the wavelength dependent TCO refractive index n_{TCO} . The fit parameter c for the investigated etching series was between 0.45 and 0.52 for etching times longer and equal 10 seconds, which correlates to substrates with a more homogeneous surface texture. Only for the samples with an etching time of 3 s and 6 s a good fit was achieved for values of $c=0.35$ and 0.37 , respectively. These values strongly deviate from the expected value of 0.5 [9].

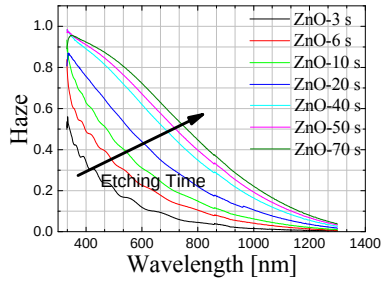


Figure 3: Measured haze of ZnO substrates with different etching times.

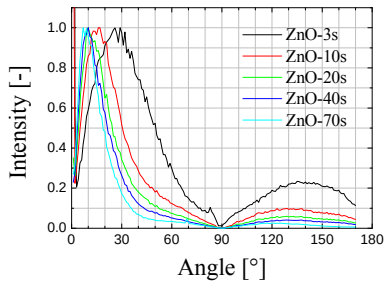


Figure 4: Measured ARS distributions of ZnO substrates with different etching times and determined by using a 550 nm laser.

Besides the haze also the angle resolved scattering (ARS) behaviour depends on the etching time. The ARS experiment was performed by using a 550 nm laser with normal incidence on the sample. A mobile photodetector records the scattering intensity for different angles. Considering an isotropic scattering behaviour to the axis of the laser beam the entire intensity for each angle is calculated by integration over the corresponding angle segment. The resulting intensity profiles are normalized to the maximum intensity of the scattered light. The measured ARS characteristics for selected samples of the etching

series are plotted in Figure 4. For an etching time of 3 seconds we observe in transmission (0° - 90°) a maximum located at high angles (29°). With increasing etching time the position of the maximum shifts from 29° (3 seconds) to 7° (70 seconds). Furthermore, the large angle contributions in the range between 40° and 90° and the ARS distribution in reflection decreases with etching time.

The observed deviations of the experimentally determined haze and the haze as predicted by scalar scattering theory show that the surface roughness is not sufficient to describe the scattering behaviour of the TCO and the surface topology has to be taken into account. Also the observed evolution of the ARS distribution with the etching time can only be explained by considering the surface topology.

3 ANALYSIS OF STRUCTURE AND ARS DISTRIBUTION

To analyse the relation between surface morphology and ARS behaviour we developed a model based on the surface topology of randomly textured surfaces measured by AFM data. To calculate the local inclination angles we determined the normal vectors from the plane equations for three neighbouring points. The inclination angle is defined as the angle of the normal vector with respect to the z-axis. In Figure 5 the principle of plane formation is illustrated. For our investigations AFM scans of an area of $7 \times 7 \mu\text{m}^2$ with a lateral resolution of 13 nm were carried out.

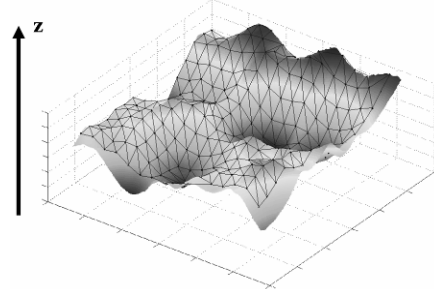


Figure 5: AFM data is used to calculate the local inclination angles with respect to the z-axis.

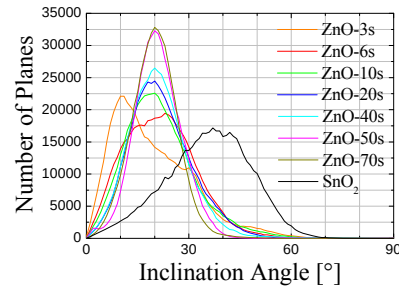


Figure 6: Calculated inclination angles of sputtered ZnO with different etching times and an APCVD SnO_2 substrate.

First the inclination angles of the post-etched substrates with different etching times were determined. The distributions of inclination angles for a variety of samples are plotted in Figure 6. Surprisingly, the maximum of the distribution located at approximately 20° is found to be nearly independent of the etching time. Only the distribution of the 3 seconds etched substrate deviates from the others

with its maximum at 10° . To compare the results for the ZnO etching series, we also plotted the inclination angle distribution of a commercial APCVD SnO_2 substrate with a pyramid-like structure.

The ARS spectra are calculated using the refractive indices $n_{\text{TCO}}=1.8$ and $n_{\text{glass}}=1.5$ and the surface topography measured by AFM. The light beam strikes on the glass/TCO stack under normal incidence with respect to the mean surface level. Light scattering in reflection and transmission at the individual interfaces is determined by the Snellius law and the Fresnel functions and the optical path within the layer stack is calculated using a ray tracing approach. In Figure 7 (top) we compare the simulated ARS of a 40 seconds etched ZnO substrate and an APCVD SnO_2 sample with experimental data. Angles between 0° and 90° represent the transmitted light and the reflected light is represented by angles between 90° and 180° . The general trends observed in the experiments are well reproduced within the simulations (Fig. 7, top). However, clear deviations can also be observed. For the ZnO substrate the angle of the calculated maximum in the ARS data (10°) is slightly shifted to smaller values in comparison to the experimentally determined value (18°). For the LPCVD SnO_2 substrate a lower position of the maximum is simulated than is experimentally observed. Furthermore, for both substrates the large angle distributions (40° - 90°) in transmittance and the contribution in reflection are slightly underestimated in the calculation.

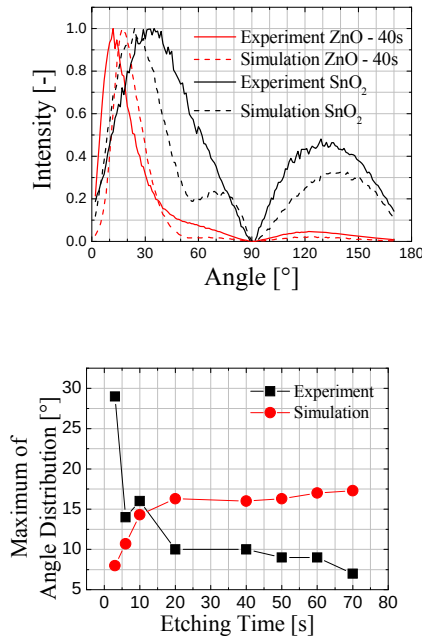


Figure 7: Simulated and experimental ARS data of a 40 seconds etched ZnO and an APCVD SnO_2 sample. A simple geometric optics model was used for the simulations (left). The different position of the maximum obtained by experiment and simulation (right).

Due to the similar inclination angles of substrates with different etching times (Fig. 6) the simulation

provides nearly identical ARS distributions for most etching times (not shown). Hence, the simulated ARS distributions from samples with different etching times ($\neq 3$ seconds) have nearly the same ARS distributions as the 40 seconds etched sample with a maximum at around 18° (Fig. 7, top). Figure 7 (bottom) summarizes the simulated and experimentally observed positions of the ARS maximum as a function of the etching time. The simulated ARS has a maximum at 9° for an etching time of 3 seconds. For larger etching times the position of the maxima shifts to an angle of approximately 18° . The experimental data (Fig. 4) shows for increasing etching time a shift of the position of the maximum to smaller angles. The 3 seconds etched sample has its maximum located at 30° which decreases with increasing etching time to an angle of 7° for the 70 seconds etched substrate. Besides the position of the maximum, for all samples, particularly for those with small lateral feature sizes (etching times ≤ 20 seconds), large angle contributions are underestimated.

For the ARS scattering behaviour of nano-textured TCO substrates two effects can be observed which cannot be solely described by geometric optics: (1) in particular, substrates with a low roughness show large angle ARS distributions between 40° and 90° in transmission. This contribution decreases with increasing roughness. (2) The experimentally observed maximum in ARS for long etching times ($t \geq 20$ s) is located at smaller angles than found in the simulations.

The local feature sizes we used in the model are smaller than the wavelength we used to measure the ARS. This could provide an explanation of the observed differences between the simulated and experimental data. In the next section we will investigate two different approaches to improve our simulations.

4 EFFECTIVE MEDIUM AND LARGER PLANE APPROACH

Since the wavelength of the incident light is larger than the local feature sizes, an effective medium approach and a larger grid approach is tested. In Figure 9 the two approaches are illustrated. The impact of both approaches on the scattering behaviour of the substrates is discussed for the 40 seconds etched ZnO substrate.

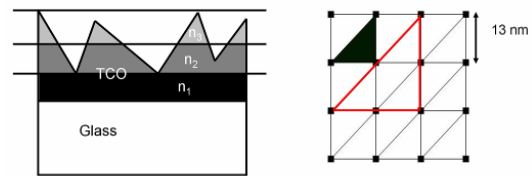


Figure 8: Effective medium approach (left) and larger grid approach (right).

The *effective medium approach* assumes for thin films a local variation of the refractive index at different surface heights [10]. We applied a approach, where n_{TCO} changes in 3 discrete zones. In our calculation we used $n_1=1.8$, $n_2=1.65$ and $n_3=1.5$ as indicated in Figure 8 (left).

For the *larger plane approach* we mapped the AFM scan to a grid with a larger grid length (the original grid has a grid length of 13 nm). Figure 8 (right) shows the smallest grid size (black triangle) originating from the AFM scan and as an example a larger plane using a triangle with 26 nm edge

length (red triangle).

Figure 9 shows the simulations in comparison to the experiment of a ZnO-40s sample for both approaches. A good match of the maximum in ARS is achieved, when the effective medium approach or a larger edge length is considered. However, the contribution for larger angles between 40° and 90° remains underestimated within the simulations, which can be likely explained by Mie scattering [11]. Also the additional amount of reflected light scattered into large angles compared to the ARS in transmission gives a hint for Mie scattering [11] where a high amount of light can be scattered backwards.

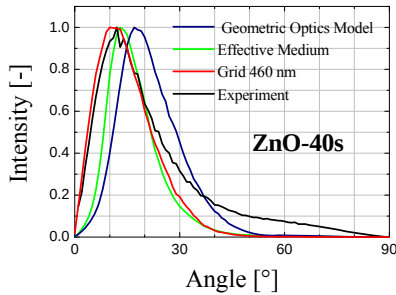


Figure 9: Simulation ARS distribution considering the effective medium approach and the larger grid approach in comparison to the experiment.

Using the effective medium approach a better agreement between simulations and the experimental data is achieved, because a lower refractive index value results in smaller scattering angles. Next, we have analysed the scattering behaviour of the whole etching series using the effective medium model. We found that the simulated position of the maximum does not significantly shift with etching time. For all samples a shift of around 5° in comparison to the standard approach was determined. Only for the sample with a strong inhomogeneous texture (3 s) a deviation in respect to all other samples is observed. Since the etching step leads to a crater-like texture, the nearly flat regions which make up the largest part of the surface are all located in zone 1 with a refractive index n_1 . For SnO_2 samples no significant shift of the maximum is observed because of the pyramid-like structure. Due to the pyramid-like structure, the total area of regions with a lower refractive index is significantly smaller in comparison to textures with a crater-like texture. Thus, this simple approach dividing the peak to peak value in three regions can not explain the shift in ARS with etching time. However, device simulations based on the rigorous solution of the Maxwell equation point out that the shape of the texture and the lateral dimension have a significant influence on the light in-coupling properties of thin-film silicon cells [12]. Therefore a better adjustment of the individual height levels has to be considered.

The general observations using the larger plane approach are the following: with increasing edge length the maximum of the ARS shifts to smaller angles since an average slope of the area is determined over a broader area. However, this effect is more pronounced for smaller lateral feature sizes

since an averaging of the peak to peak values occurs at smaller distance. For the 40 s-etched sample a larger plane approach with a 460 nm edge length accurately describes the position of the maximum. For different etching times we could not obtain a better agreement between our geometric optics model and the experiments using the large plane model. On the contrary, the trend of the position of the maximum as a function of etching time is in the opposite direction of the experimental observations since the shift in position of the maximum to smaller angles is more pronounced for decreasing lateral feature sizes (etching time).

As the geometric optics model is only valid for relatively large feature sizes, the model should be more accurate for the samples with a large feature size than for samples with small feature sizes. Consequently, a better description of the scattering behaviour for substrates with long etching times than for substrates with short etching times is expected.

5 LOCAL ANGLE ANALYSIS

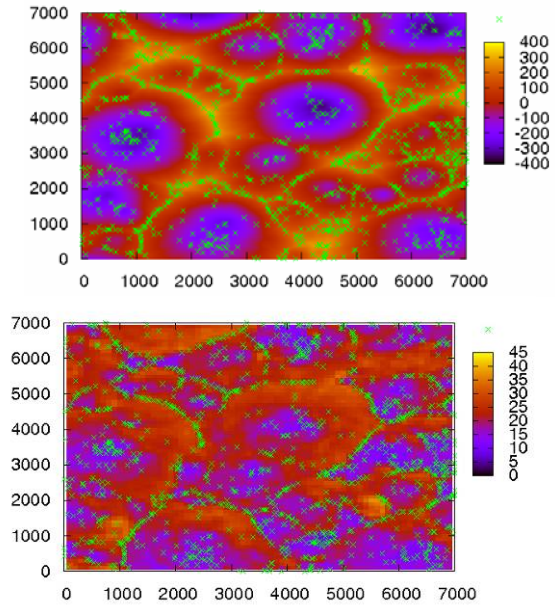


Figure 10: Top: 3D topography of a 60 seconds etched ZnO substrate (units in nm). Bottom: Local inclination angles (in nm and degrees) on the surface. The green crosses are located along the crater edges.

The nano-textured substrates consist of craters with different lateral feature sizes and nearly flat areas depending on the etching time. Since only a part of the light is diffusely scattered the question arises if scattering in small angles is determined by the craters itself. Therefore the surface topology of the textured surfaces is analysed. The surfaces are analysed by plotting the topography (Fig. 10/11, top) and the local angle distributions (Fig. 10/11, bottom) of the $7 \times 7 \mu\text{m}^2$ AFM scans of a long time (60 s) and a short time (3 s) etched sample. To calculate the local angle distributions the surface is divided by a mesh into $100 \times 100 \text{ nm}^2$ small areas. For this area the most frequent inclination angle is determined and plotted. To allow for a better comparison of topography and inclination angles crater edges are marked as green crosses. In the vicinity of the crater bottoms angles around 15° were determined. Larger angles ($\sim 25^\circ$) are located mainly in the intermediate regions of the crater walls.

Particularly for long etching times very large inclination angles (around 35°) were observed close to the crater edges. Figure 11 shows that for substrates with short etching times the smallest angles can be found between individual craters and at the crater centres.

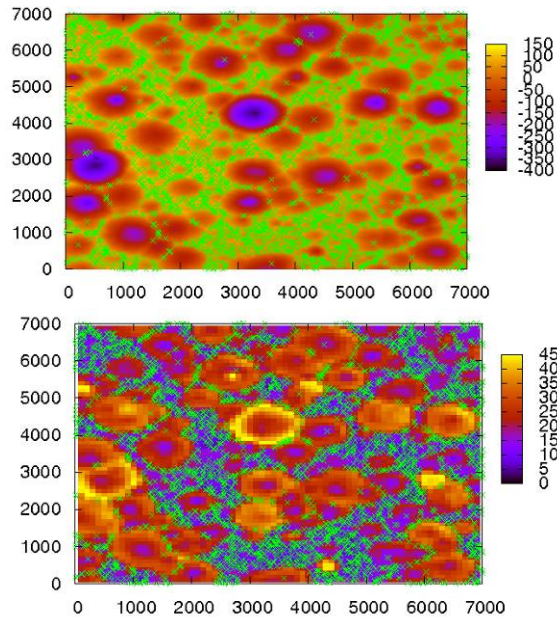


Figure 11: Top: 3D topography of a 6 seconds etched ZnO substrate (units in nm). Bottom: Local inclination angles (in nm and degrees) on the surface. The crosses are located along the crater edges.

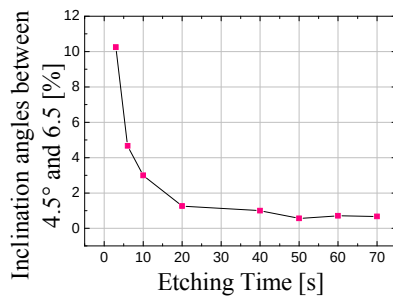


Figure 12: Amount of area with inclination angles of 5.5° in percent.

The ARS measurements show a maximum at an angle of 10° (Fig. 4). To reach a scattering angle of 10° by geometric optics and $n_{\text{TCO}} = 1.8$ an inclination angle of $\arcsin(n_{\text{Air}} / n_{\text{TCO}} \cdot \sin(10^\circ)) \approx 5.5^\circ$ is necessary. In Figure 12 the fraction of area with angles between 4.5° and 6.5° degrees are plotted as a function of etching time. Surprisingly, samples with long etching times do not show the highest amount of areas with an angle of around 5.5°. For etching times longer than 20 seconds less than 5% of the surface is covered by inclination angle of 5.5°. This means that the scattering angle of 10 degrees cannot be sufficiently explained by considering only the inclination angles in combination with geometric optics.

A possible explanation for the shift in the position of the maximum can be that geometric optical wave

fronts are formed at the crater walls and are disturbed at the edges (tips and dips).

6 CONCLUSIONS

We have investigated in detail the optical scattering properties of sputtered, Al-doped ZnO substrates on glass, which are textured by a post-etching step. To gain more insight in how the surface structure is correlated with the scattering properties, we developed a model based on geometric optics. The model uses AFM measurements of the surface topology as input data to simulate the ARS. The general trends observed in the experiments are well reproduced within the simulations. However, clear deviations can also be observed. A partial explanation was found in the local feature sizes we used, as these feature sizes were smaller than the wavelength we used to measure the ARS. The remaining differences between the geometric optics model simulations and the experimental results suggest a significant influence of Mie scattering and likely disturbed wave fronts formed at the crater walls and are disturbed at the edges (tips and dips).

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