Optical scattering properties of a nano-textured ZnO-silicon interface

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ABSTRACT

The scattering properties of transparent conductive oxide (TCO) layers are fundamentally related to the performance of thin film silicon solar cells. In this study we introduce an experimental technique to access light scattering properties at textured TCO-silicon interfaces. Therefore we prepared a sample with a polished microcrystalline silicon layer, which is deposited onto a rough TCO layer. We used the measured results to validate calculations obtained with rigorous diffraction theory, i.e. a numerical solution of Maxwell's equations. Furthermore we evaluated four approximate models based on the scalar scattering theory and ray tracing and compared them to the rigorous diffraction theory.

Keywords: scattering, surface-textured TCO, microcrystalline silicon, ZnO, FDTD, scalar scattering theory, ray tracing

1. INTRODUCTION

State of the art thin film silicon solar cells contain rough interfaces in order to increase their short circuit current.[1–4] The rough interfaces scatter the incoming light, which leads to an increased photon path length and partial total internal reflection in the absorber layer of the solar cell and results in increased absorption in the absorber layer. The increased absorption finally leads to a higher short circuit current. Usually the textured interfaces are introduced into the solar cells by depositing the silicon layers onto a surface-textured transparent conductive oxide (TCO) layer. The most important textured interface in the solar cell is the TCO-silicon interface.

In order to optimise the interface morphology of the textured interfaces, their light scattering properties have to be investigated experimentally and theoretically. Usually not TCO-silicon interfaces but TCO-air interfaces are investigated by using a textured TCO on a glass substrate without the solar cell on top of it. In this configuration, only light scattering by TCO-air interfaces can be studied. However, light scattering by an textured interface strongly depends on the refractive indices of the two media constituting the interface. Therefore the scattering properties of TCO-silicon interfaces need to be studied if one wants to investigate how light propagates in thin film silicon solar cells.

To theoretically describe scattering by textured interfaces, many different approaches such as the rigorous diffraction theory,[5,6] the scalar scattering theory[7–12] and ray tracing[13] can be found in literature. For all these theoretical approaches, the experimental validation of the predicted scattering behaviour of the relevant TCO-silicon interface is missing.

In this work we present an experimental technique to measure the angular intensity distribution (AID) at TCO-silicon interfaces. As TCO we used sputtered aluminium-doped zinc oxide (ZnO:Al). The investigated sample consists of a polished hydrogenated microcrystalline silicon (µc-Si:H) layer on top of a ZnO:Al layer deposited onto a flat Corning glass substrate. We further compare the measured AID to rigorous results obtained by the finite-difference time-domain (FDTD) method.[14,15] The FDTD results are then used to evaluate results obtained with three different models based on the scalar scattering theory and a ray tracing approach based on geometrical optics. This comparison allows us to estimate the predictive power of the different theoretical approaches.

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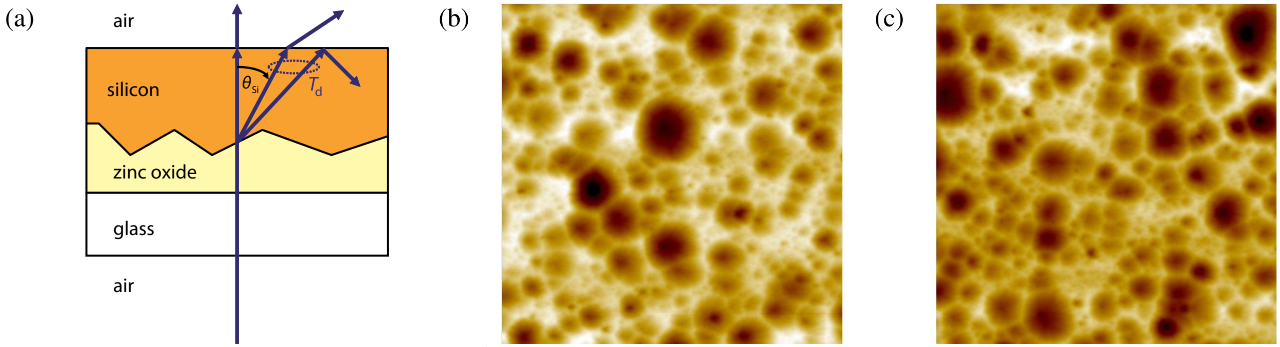

Figure 1. Sample geometry (a) and two 10×10 µm^2 AFM scans of the rough, crater-like ZnO interface at different positions (b,c) with a height range of 610 nm (b) and 545 nm (c).

2. MEASURING THE AID OF TEXTURED TCO-SILICON INTERFACES

While the AID* of TCO-air interfaces has been measured for many years,[8, 16] measuring the AID of TCO-silicon interfaces is an involved task due to a number of reasons. First, the silicon layer absorbs the light traversing it. Therefore one has to use detectors that are able to detect low intensities or one has to use a light source that is strong enough to provide the detectors with a sufficient intensity. However, light trapping is most important in the infrared, where the silicon has a low absorptance. Due to the low absorptance, measuring in the near infrared is possible. Second, when a silicon layer is deposited onto a rough TCO layer, the silicon-air interface also will exhibit a certain roughness. If one deposits microcrystalline silicon, the material itself will be rough due to its microstructure. Therefore light traversing the layer stack will not only be scattered by the TCO-silicon interface, but also by the silicon-air interface. This makes it impossible to extract the AID of the TCO-silicon interface from the AID measured for the whole layer stack. To solve this problem, one can polish the rough silicon surface so that the light is only scattered by the TCO-silicon interface. Third, even if the silicon-air interface is polished, light interacts with this interface: It is partially reflected back into the silicon and it is refracted at the silicon-air interface. Considering these three effects, we then can relate the AID at the TCO-silicon interface (i.e. inside the silicon) to the AID in the air via

$$\mathrm{AID}_{\mathrm{Si}}(\theta_{\mathrm{Si}}) \cdot \Omega_{\mathrm{Si}} = \mathrm{AID}_{\mathrm{air}}(\theta_{\mathrm{air}}) \cdot \frac{1}{\tau} \cdot \exp\left(\alpha_{\mathrm{Si}} \frac{d}{\cos\theta_{\mathrm{Si}}}\right) \cdot \Omega_{\mathrm{air}}, \tag{1}$$

where τ is the transmittance of the silicon-air interface as obtained from the Fresnel equations, d is the average thickness of the silicon layer and α_{Si} is the absorption coefficient of the silicon. The scattering angles θ_{Si} and θ_{air} are related to each other via Snell's law. Due to the refraction at the silicon-air interface, the solid angle Ω_{Si} of the detector differs from the solid angle Ω_{air} into which the light that reaches the detector is emitted. These two solid angles are related to each other via

$$\frac{\Omega_{\mathrm{Si}}}{\Omega_{\mathrm{air}}} = \frac{n_{\mathrm{Si}}^2}{n_{\mathrm{air}}^2} \cdot \frac{\cos\theta_{\mathrm{Si}} \cdot \cos\psi_{\mathrm{Si}}}{\cos\theta_{\mathrm{air}} \cdot \cos\psi_{\mathrm{air}}} = n_{\mathrm{Si}}^2 \cdot \frac{\cos\theta_{\mathrm{Si}}}{\cos\theta_{\mathrm{air}}}. \tag{2}$$

In Eq. (2) we took into account that the detector moves around the $x-z$ plane and that the azimuth ψ therefore is zero. We further approximated the refractive index of the air n_{air} to be 1. We thus find that the AID inside the silicon can be retrieved from the measured AID in air with the equation

$$\mathrm{AID}_{\mathrm{Si}}(\theta_{\mathrm{Si}}) = \mathrm{AID}_{\mathrm{air}}(\theta_{\mathrm{air}}) \cdot \frac{1}{\tau} \cdot \exp\left(\alpha_{\mathrm{Si}} \frac{d}{\cos\theta_{\mathrm{Si}}}\right) \cdot n_{\mathrm{Si}}^2 \cdot \frac{\cos\theta_{\mathrm{Si}}}{\cos\theta_{\mathrm{air}}}. \tag{3}$$

In reality, light that leaves the layer stack via the silicon-air interface after multiple reflections also will contribute to the measured AID and therefore also will contribute to the $\mathrm{AID}_{\mathrm{Si}}$ that is obtained with Eq. (3). The $\mathrm{AID}_{\mathrm{Si}}$ therefore differs from the (hypothetical) $\mathrm{AID}_{\mathrm{Si}}$ of light that is scattered into a half space filled with silicon. We also note that the $\mathrm{AID}_{\mathrm{Si}}$ only can be obtained up to the critical angle $\theta_c = \arcsin\left(n_{\mathrm{Si}}^{-1}\right)$.

Figure 1 (a) illustrates the geometry of the investigated sample. To prepare the sample we deposited sputtered ZnO:Al onto a Corning glass substrate. After deposition the ZnO:Al was etched in 0.5 wt% diluted hydrochloric acid for 30 s. Due

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*By "AID" we mean the "AID in transmission" throughout this manuscript.

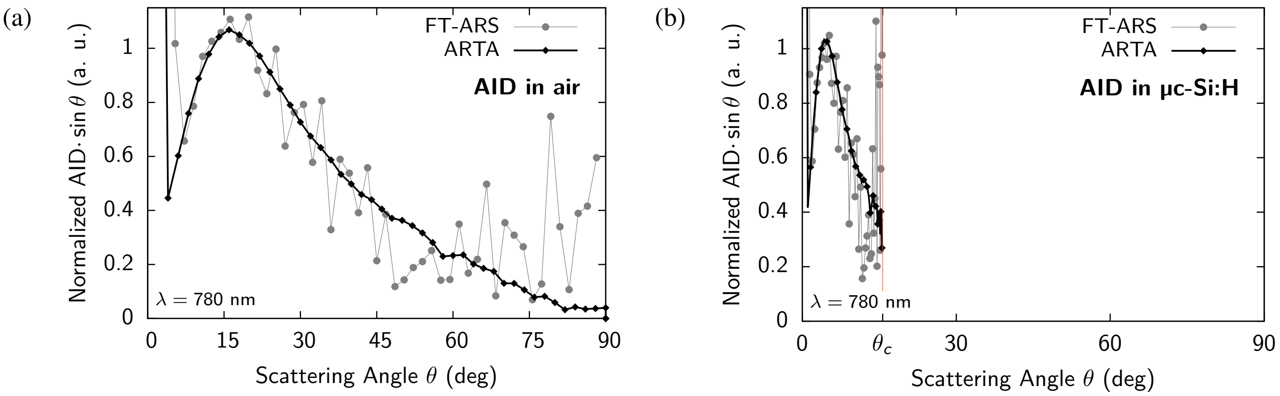

Figure 2. (a) The AID$\cdot \sin\theta$ in air measured with the Fourier transform angular resolved scattering setup (FT-ARS) and the Automated Reflectance / Transmittance Analyser (ARTA). (b) The AID$\cdot \sin\theta$ inside the silicon layer obtained from the results in (a) with Eq. (3). The critical angle θ_c of the silicon-air interface is indicated.

to the etching a crater-like texture with a root mean square roughness of 78 ± 2 nm was created, as can be seen in Fig. 1 (b) and (c) that show atomic force microscopy (AFM) pictures of the ZnO:Al surfaces. Onto the ZnO:Al we deposited a 3 μm thick μc-Si:H layer by plasma-enhanced chemical vapour deposition and polished the silicon surface, resulting in a root mean square roughness of 9 ± 2 nm. The average thickness of the silicon layer after polishing is estimated to be approximately 2.3 μm. We extracted the refractive indices and absorption coefficients with ellipsometry: The refractive indices at 780 nm are $n_{\mathrm{ZnO}} = 1.65$ and $n_{\mathrm{Si}} = 3.70$. The absorption coefficients at 780 nm are $\alpha_{\mathrm{ZnO}} = 369$ cm^{-1} and $\alpha_{\mathrm{Si}} = 860$ cm^{-1}. The critical angle at the silicon-air interface is then $\theta_c = 15.68°$.

We measured the AID of the glass-ZnO-silicon[†] layer stack in the near infrared at 780 nm. We used two setups: A Bruker IFS 66v Fourier transform infrared spectroscopy that was equipped with a self-assembled angular resolved scattering accessory (FT-ARS) and a PerkinElmer Lambda 950 spectrophotometer equipped with an Automated Reflectance / Transmittance Analyser (ARTA).[17] Figure 2 (a) shows the AID in air that was measured with the two setups. Although the trends of the FT-ARS and the ARTA measurements resemble each other, the FT-ARS measurement is much noisier. While the ARTA results keep decreasing with increasing angles, for the FT-ARS a saturation is detected for angles larger than 60°. The specular peak is broader for the FT-ARS. This is due different detector opening angles and light beam sizes at the two setups. Figure 2 (b) shows the AID in silicon as it was obtained from the AID in air with Eq. (3). Due to the large refractive index of silicon, only the AID for angles smaller than 15.68° can be obtained. For the further analysis we will proceed with the data obtained by the ARTA since it is much smoother. All the AIDs discussed in this paper are normalised to allow a better comparison. The normalisation procedure is discussed in the appendix.

3. RIGOROUS CALCULATIONS WITH FDTD

We performed the rigorous diffraction theory calculations with the open source software Meep[18] that implements the FDTD method.[14,15] The calculations take the refractive indices and extinction coefficients of the materials into account. We implemented the textured interfaces by the AFM data. The calculations were done on the geometry shown in Fig. 1 (a) with a spatial resolution of 20 nm and metallic boundary conditions. From the calculated light intensities slightly beneath the flat silicon-air interface, the AID in silicon is obtained by a fast Fourier transform. The FDTD results for both AFM topographies together with the measured AID are plotted in Fig. 3 (a). Even though the FDTD results show zigzag-like features due to the finite size of the calculation domain, a good agreement between calculations and measurements can be found.

In the next section we compare different approximate models for calculating the AID to each other and to the FDTD values. With the approximate models the AID in a half space filled with a non-absorbing material with the (real part of the) refractive index of silicon can be calculated. As we mentioned already above, multiple reflections inside the silicon layer influence the AID. The AID shown in Fig. 3 (a) was obtained from FDTD calculations of the structure depicted in Fig. 1 (a). To compare the AID from FDTD with the AIDs from the approximate models one should, however, calculate it for

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[†] For the sake of simplicity we denote "ZnO:Al-(μc-Si:H)" by "ZnO-silicon" for the remainder of this manuscript.

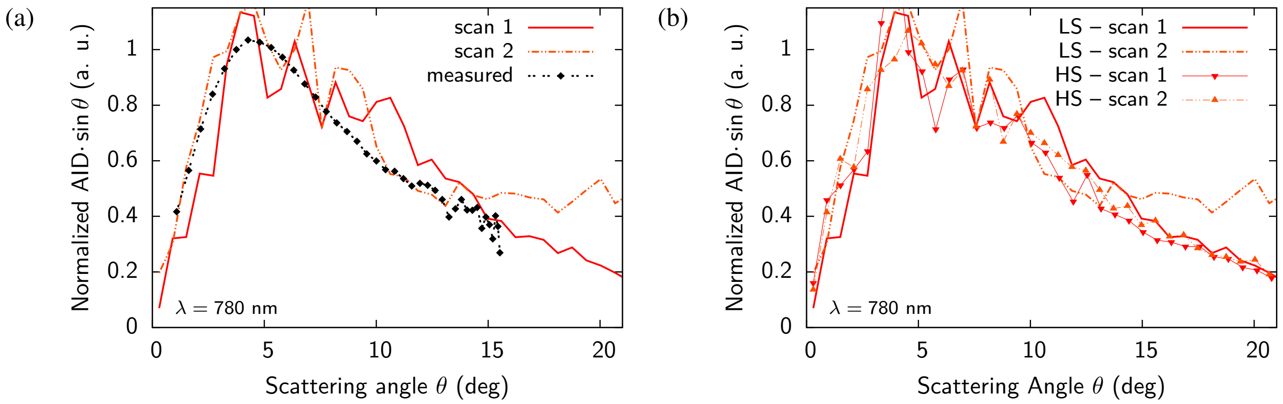

Figure 3. The AID$\cdot \sin\theta$ inside the silicon layer obtained from FDTD calculations and measurements. (b) The AID$\cdot \sin\theta$ inside the silicon layer stack (SL, thick lines) and in a half space filled with a silicon-like non-absorbing material (HS, thin dotted lines) as obtained from FDTD.

the half space. Figure 3 (b) shows FDTD calculations for the silicon layer and the half space filled with a non-absorbing material with the (real part of the) refractive index of silicon.

4. APPROXIMATE MODELS FOR CALCULATING THE AID

For the comparison, we considered four approximate model that were recently developed. While the phase model,[9] the Born-Fraunhofer model[10] and the grating model[11] are based on the scalar scattering theory,[‡] the ray tracing approach[13] uses geometrical optics. In the scalar scattering theory the electromagnetic vector fields are replaced by a complex scalar field. The scalar scattering theory requires the dielectric function across the sample varying so slowly with position that it is effectively constant over distances of the order of the wavelength. Even though this requirement is clearly not fulfilled for TCO-air or TCO-silicon interfaces, the models based on the scalar scattering theory can predict the AID of TCO-air interfaces very well.

The three models based on the scalar scattering theory use the insight that in a first order approximation the AID is related to the Fourier transform of the scattering object. The three models, however, differ from each other in detail. In the phase model the AID is proportional to

$$\mathrm{AID}_{\mathrm{phase}}(\theta) \propto \lambda_0^2 \cos\theta \,|\mathscr{F}\{\exp[ik_0 z(x,y)(n_{\mathrm{TCO}}-n_{\mathrm{Si}})]\}|^2, \tag{4}$$

where $z(x,y)$ is the height of the texture at the point (x,y) as obtained with AFM images and n_{TCO} and n_{Si} denote the refractive indices of the materials surrounding the interface. The Fourier transform $\mathscr{F}$ is taken of the phase $kz(x,y)(n_{\mathrm{TCO}} - n_{\mathrm{Si}})$ that is accumulated when light traverses the textured interface at the position (x,y).

While the phase model naturally contains the refractive indices of the materials surrounding the scattering interface, this is not the case for the Born-Fraunhofer model that in principle deals with a scattering object *in vacuo*. The AID is in this case proportional to

$$\mathrm{AID}_{\mathrm{BF}} \propto \cos\theta \left| \frac{k_0^2}{4\pi} \left(n_{\mathrm{TCO}}^2 - 1\right) \mathscr{F}\langle (ik_0)^{-1}\{1-\exp[ik_0 z(x,y)]\}\rangle \right|^2 . \tag{5}$$

To be able to calculate the AID of TCO-silicon interfaces one has to place the scattering structure in a surrounding filled with silicon. In the equation this can be done by replacing the wave vector *in vacuo* k_0 by the effective wave vector in silicon, $k_{\mathrm{eff}} = k_0 \cdot n_{\mathrm{Si}}$, and by replacing the refractive index n_{TCO} by the relative refractive index, $n_{\mathrm{eff}} = n_{\mathrm{TCO}}/n_{\mathrm{Si}}$.

The biggest difference between Eqs. (4) and (5) is the presence of the refractive indices in the exponents of the exponential functions that undergo the Fourier transform. While the exponent in the phase model contains the difference of the refractive indices $n_{\mathrm{TCO}} - n_{\mathrm{Si}}$, the exponent in the Born Fraunhofer model only contains the refractive index of the surrounding silicon that is incorporated in the effective wave vector k_{eff}.

[‡] See for example Ref. [19, Sec. 8.4 and 13.1].

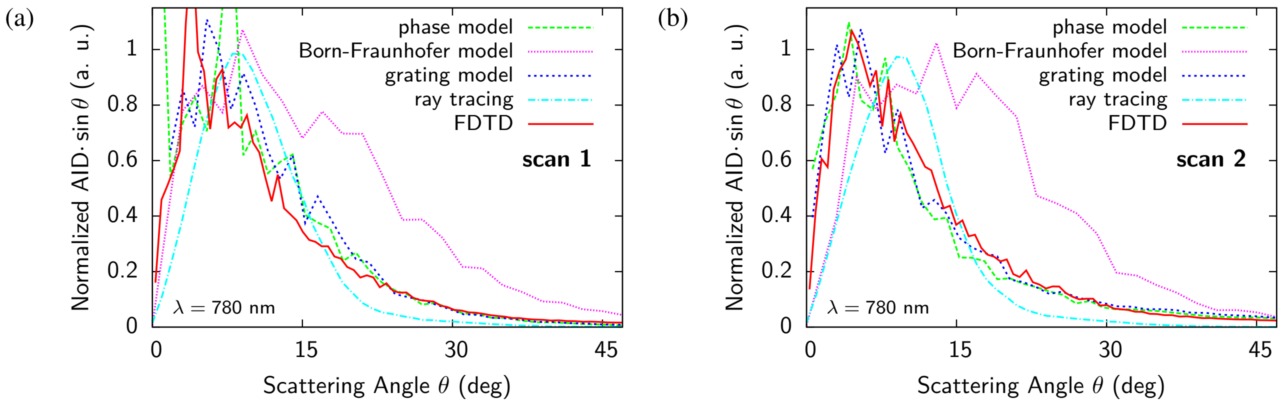

Figure 4. The AID· $\sin\theta$ in a half space filled with a non-absorbing silicon-like material obtained with four approximate model and FDTD calculations. The results shown in (a) and (b) were obtained with the AFM scans shown in Fig. 1 (b) and (c), respectively.

In the grating model, the scattering interface is decomposed into a superposition of periodic gratings by fast Fourier transform. To obtain the AID, the first diffraction orders of the gratings are superposed. If necessary, also the higher diffraction orders of the gratings can be taken into account. In difference to the phase model and the Born-Fraunhofer model, which use the height distribution of the texture as input, the grating model only considers the relative height modulation of the texture. It therefore is not sensitive to stretching the profile vertically.

In the ray tracing approach, the scattering interface is decomposed into a collection of small facets. Each of these facets refracts the incident light according to Snell's law. A ray is sent through each facet. The (unnormalised) AID at a scattering angle θ is then given by the number of rays that are scattered into a small interval around θ .

Figures 4 (a) and (b) show the AID of the ZnO-silicon interface as obtained with the four approximate models. In (a) AFM scan 1 [Fig. 1 (b)] was used, while the results in (b) were obtained with scan 2 [Fig. 1 (c)]. As mentioned before, the AID shown in the figure is obtained for a non-absorbing material with the refractive index of silicon. While the AID obtained from ray tracing is smooth, all the other approaches lead to zigzag-like results. One can observe some trends anyway. While the phase model, the grating model and the FDTD calculation resemble each other, the ray tracing approach and the Born-Fraunhofer model clearly deviate from the rest. The deviations of the ray tracing approach are not very surprising, since the features of the scattering interface are in the range of the wavelength and wave-optical effects are not considered in this approach. The reason for the large deviation of the Born-Fraunhofer model is the absence of the term $n_{\mathrm{TCO}} - n_{\mathrm{Si}}$ as it is present in the phase model. For TCO-air interfaces results obtained with both the phase model and the Born-Fraunhofer model are very similar. Since n_{TCO} typically lies between 1.5 and 2 and n_{air} can be assumed as 1, $n_{\mathrm{TCO}} - n_{\mathrm{air}}$ will not deviate too much from 1, i.e. the Fourier transforms in both the phase model and the Born-Fraunhofer model lead to similar results. Due to the high refractive index of silicon, $n_{\mathrm{TCO}} - n_{\mathrm{Si}}$, however, deviates a lot from 1, leading to very different results for the phase model and the Born-Fraunhofer model, as can be seen in Fig. 4.

We also can analyse the validity of the different approaches by comparing the angles $\theta_{\max}$ at which AID· $\sin\theta$ takes its maximum. Table 1 shows the average $\theta_{\max}$ from scans 1 and 2 as they were obtained from the fitting procedure described in the appendix. Similar to what we discussed above, $\theta_{\max}$ from the FDTD, the phase model and the grating model are close to each other while the maximum angles from the Born-Fraunhofer model and the ray tracing approach are much higher.

Table 1. The angle $\theta_{\max}$ at which AID· $\sin\theta$ takes its maximum for the different approaches.

Approach	$\theta_{\max}$
(ARTA)	(4.2°)
FDTD	4.3°
phase model	5.4°
Born-Fraunhofer model	8.5°
grating model	3.9°
ray tracing approach	8.8°

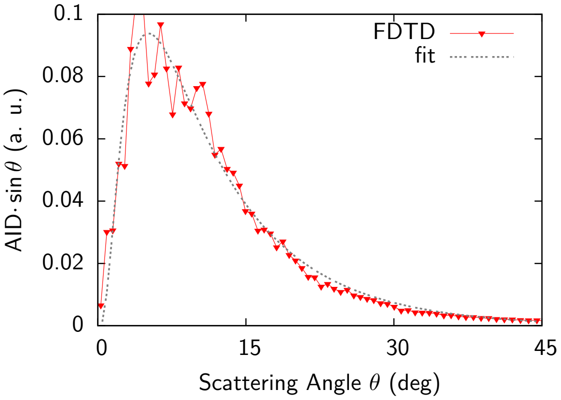

Figure 5. The function $f(\theta)$ [Eq. (6)] fitted to AID· sin θ inside the silicon layer as obtained by FDTD.

5. CONCLUSIONS

We presented a novel approach to measure the angular intensity distribution (AID) of light that was scattered at a textured ZnO-silicon interface. We further found good agreement between the measured AID and calculations based on the rigorous diffraction theory. Based on these results, we tested different established scattering models. In difference to the measurements, where the AID inside a silicon layer was investigated, the scattering models were applied to a textured interface between two halfspaces.

As a reference, we took the calculated AID in the silicon halfspace as it was obtained from the rigorous diffraction theory. We tested three different models based on the scalar scattering theory: The phase model, the Born-Fraunhofer model and the grating model. Further we tested a ray tracing approach based on geometrical optics. Both the results obtained from the phase model and the grating model resemble the calculations performed with the rigorous diffraction theory. The ray tracing approach predicts the maximum of the AID at larger angles and the Born-Fraunhofer model overestimates the AID at larger scattering angles. Since the lateral sizes and heights of the typical texture features define which physical mechanism dominates the scattering behaviour, textures that significantly differ from those studied in this work have to be investigated in the same manner to further test the validity of the scattering models.

The novel experimental technique allows to obtain essential information on scattering into the absorber material of thin film solar cells. This information allows investigating the scattering mechanisms inside the solar cells and is therefore much more valuable than scattering properties obtained at TCO-air interfaces.

APPENDIX A. THE FITTING FUNCTION

In this work we compare AIDs obtained from various approaches. To be able to perform this comparison, two measures have to be taken. First, we do not plot the AID but the AID· sin θ, which corresponds to the intensity that is scattered into the *ring* corresponding to the scattering angle θ. Contrary to the AID, which has its maximum at $\theta = 0°$, AID· sin θ peaks at larger angles. The position of this peak indicates how strongly the light is scattered away from the specular direction. Second, we normalise this peak to 1. The AID from several approaches is zigzag-like, as can be seen in Figs. 3 and 4. To compare the different AIDs it therefore is more convenient to normalise to a fitted function. As fitting function we can use

$$f(\theta) = \frac{a}{\tan\theta} \exp\left[-\frac{1}{2}\left(\frac{\ln(\tan\theta) - b}{c}\right)^2\right] \tag{6}$$

with the fitting parameters a, b and c. This function resembles the log-normal distribution[20] with the difference that θ was substituted by $\tan\theta$. This substitution is performed in order to ensure that $f(\theta) \to 0$ at 0° and 90° as this is the case for AID· sin θ. The function $f(\theta)$ takes its maximum $f_{\max}(\theta) = a \cdot \exp(0.5\,c^2 - b)$ at $\tan\theta_{\max} = \exp(b - c^2)$. Figure 5 shows, as an example, $f(\theta)$ fitted to AID· sin θ inside the silicon layer as obtained by FDTD.

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