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Angular resolved scattering by a nano-textured ZnO/silicon interface

M. Schulte,[1] K. Bittkau,[1,a)] K. Jäger,[2] M. Ermes,[1] M. Zeman,[2] and B. E. Pieters[1]
[1]*IEK5–Photovoltaik, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany*
[2]*Photovoltaic Materials and Devices Laboratory, Delft University of Technology, P. O. Box 5031, 2600 GA Delft, The Netherlands*

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Textured interfaces in thin-film silicon solar cells improve the efficiency by light scattering. A technique to get experimental access to the angular intensity distribution (AID) at textured interfaces of the transparent conductive oxide (TCO) and silicon is introduced. Measurements are performed on a sample with polished microcrystalline silicon layer deposited onto a rough TCO layer. The AID determined from the experiment is used to validate the AID obtained by a rigorous solution of Maxwell's equations. Furthermore, the applicability of other theoretical approaches based on scalar scattering theory and ray tracing is discussed with respect to the solution of Maxwell's equations. © *2011 American Institute of Physics*. [doi:10.1063/1.3640238]

Despite the reduction of spurious reflections, optimized light scattering by textured transparent conductive oxides (TCO) is well-known to improve the efficiency of thin-film silicon (Si) solar cells.[1–9] Therefore, experimental and theoretical methods to study light scattering by TCO/Si interfaces are of great importance for further improving the solar cell performance. Commonly, these TCO properties are investigated using a textured TCO on a glass substrate without the solar cell on top of it. In this configuration, only the light scattering properties of TCO/air interfaces can be studied. But for the comprehension of light propagation in thin-film silicon solar cells, the TCO/Si interfaces need to be studied since light scattering properties at an interface strongly depend on the refractive indices of the surrounding media. To describe these scattering properties theoretically, different approaches based on the scalar scattering theory,[10] rigorous diffraction theory,[4] and ray tracing[11] are commonly applied. For all these theoretical approaches, the validation by experimental results concerning the relevant TCO/Si interface is missing.

In this letter, an experimental technique is developed to investigate the angular intensity distribution (AID_{Si}) at TCO/Si interfaces. The experiment is performed by using a sample with a polished hydrogenated microcrystalline silicon (μc-Si:H) layer on top of a rough TCO. The TCO/Si layer stack is situated on a flat Corning glass substrate and is illuminated from the glass side. In this configuration, the AID_{Si} can be measured up to the total reflection angle for the μc-Si:H/air interface. These experimental results agree well with simulations performed by means of rigorous diffraction theory.[12–14]

Simulations based on finite-difference time-domain (FDTD) take the refractive indices and extinction coefficients of the materials and an interface profile measured by atomic force microscopy (AFM) into account. Since FDTD is extremely expensive[14,15] with respect to computational time and use of memory capacity, different approximations have been developed to predict light scattering properties of nano-textured interfaces. In this letter, different approaches based on the scalar scattering theory[16–19] and ray tracing[11] are compared to rigorous diffraction theory. To compare different models, a rough interface between TCO and μc-Si:H half spaces is investigated. This allows the validation of the scattering theories for the application to thin-film solar cells.

The investigated sample is prepared by sputtering aluminum-doped zinc oxide (ZnO:Al) on a glass substrate. The texture is achieved by post-etching in 0.5 wt. % diluted hydrochloric acid for 30 s and shows a root mean square roughness of (78 ± 2) nm. After depositing a 3 μm thick μc-Si:H layer by plasma-enhanced chemical vapor deposition, the Si surface is polished resulting in a root mean square roughness of (9 ± 2) nm. The average thickness of the Si layer after polishing is estimated to be approximately 2.3 μm.

In Fig. 1(a) the sample geometry is shown. Two AFM images of the rough ZnO:Al surface before the deposition of the μc-Si:H layer are shown in Figs. 1(b) and 1(c). The surface has crater-like features with typical lateral sizes below 1 μm. For the polished sample, the AID in air is measured by using the automated reflectance/transmittance analyser (ARTA) equipment for PerkinElmer spectrophotometer, illuminating the sample from glass side.[20,21] The measured light intensity $I(\theta_{air})$ is multiplied by the solid angle element to get $AID_{air}(\theta_{air}) = I(\theta_{air})\sin\theta_{air}$. From the AID in air, the AID in the silicon layer can be determined up to the critical angle θ_c of the μc-Si:H/air interface: $\theta_c = \arcsin(n_{air}/n_{Si})$, where n_{air} and n_{Si} are the refractive indices of air and μc-Si:H, respectively. Therefore, different optical effects have to be taken into account. First, the transmittance τ at the μc-Si:H/air interface according to Fresnel equations depends on the incident angle at the interface. Second, the larger the scattering angle at the TCO/μc-Si:H interface is, the longer the light

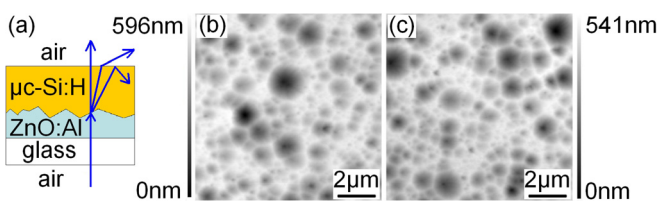

FIG. 1. (Color online) Sample geometry (a) and two 10×10 μm^2 AFM scans of the rough, crater-like ZnO interface at different positions (b), (c).

a)Electronic mail: k.bittkau@fz-juelich.de.

path through the μc-Si:H layer is, and the large the absorption. Thus, the light intensity striking the μc-Si:H/air interface depends on the scattering angle. Third, the detector in air collects light propagating from the μc-Si:H/air interface within a specific solid angle. Since the transformation of the solid angle due to the refraction at the μc-Si:H/air interface depends on the scattering angle, the measured intensities have to be corrected. The transformation of the AID measured in air to the AID in μc-Si:H is given by

$$\mathrm{AID}_{\mathrm{Si}}(\theta_{\mathrm{Si}}) = \mathrm{AID}_{\mathrm{air}}(\theta_{\mathrm{air}})\exp\left(\frac{\alpha d}{\cos\theta_{\mathrm{Si}}}\right)\frac{n_{\mathrm{Si}}\cos\theta_{\mathrm{Si}}}{\tau\cos\theta_{\mathrm{air}}}, \quad (1)$$

where α is the absorption coefficient, d the average thickness of the Si layer, and θ the scattering angle. The refractive indices n of μc-Si:H and ZnO:Al at a wavelength of $\lambda = 780$ nm were determined by ellipsometry to be 3.70 and 1.65, respectively. The absorption coefficients α of μc-Si:H and ZnO:Al were 860 and 369 cm^{-1}, respectively. Light which is multiply reflected inside the Si layer contributes to the measured intensity and is therefore also part of the AID inside the μc-Si:H layer. Thus, back-propagating light exists, which does not contribute to the AID in Eq. (1).

The resulting $\mathrm{AID}_{\mathrm{Si}}$ determined from the experiment is shown in Fig. 2 at a wavelength of 780 nm. A maximum at 4.3° is observed. Approaching the critical angle θ_c, the noise level increases. This is related to the low light intensity and the large correction factor in Eq. (1) since θ_{air} approaches π.

Rigorous diffraction theory is performed by using the open source software Meep[22] that implements the FDTD method.[12–14] The geometry shown in Fig. 1(a) is used for the FDTD simulation with a spatial resolution of 20 nm and metallic boundary conditions. By fast Fourier transformation of the complex electric field directly below the flat μc-Si:H/air interface, the $\mathrm{AID}_{\mathrm{Si}}$ in Si is obtained. The simulation results for both AFM topographies are plotted together with the measured $\mathrm{AID}_{\mathrm{Si}}$ in Fig. 2.

The simulated $\mathrm{AID}_{\mathrm{Si}}$ obtained by applying rigorous diffraction theory to the AFM scans 1 and 2 from Fig. 1 show zigzag like features due to the finite size of the calculation domain. The experimental behavior is qualitatively reproduced for angles θ between 0° and 15°. The maximum in the scattered light intensities is found for angles of 4.6° for both AFM scans. Above 15°, scan 2 shows a stronger pronounced fluctuation with a local minimum at 23°. The larger craters at the edges of scan 2 might be responsible for such artifacts. The simulated $\mathrm{AID}_{\mathrm{Si}}$ of both scans decays to 0 at approximately 70°.

The results in Fig. 2 considers a sample structure with a rough ZnO:Al/μc-Si:H and a flat μc-Si:H/air interface. This includes multiply reflected light and allows the comparison of rigorous diffraction theory and experiment. The evaluation of more simple scattering models is still a great challenge since models based on scalar scattering theories consider an interface between two half spaces. Thus, the rough interface between ZnO:Al and μc-Si:H half spaces is investigated in the following.

For comparison, different theoretical approaches that were published by different authors in recent years are taken into account: First, the grating model,[17] where the topography is given by a superposition of periodic gratings and the scattering angles are defined by the reciprocal lattice constants. Here, diffraction into the first order is assumed to be the dominant scattering process, and only the shape of the AID can be determined. Second, the phase model,[18] where the local phase shift $\exp[ikz(x,y)(n_{\mathrm{ZnO}} - n_{\mathrm{Si}})]$ of the electromagnetic wave while traversing the scattering volume defines the scattering. Third, the Born-Fraunhofer approximation[19] that assumes that the shape of the $\mathrm{AID}_{\mathrm{Si}}$ is determined by the phase changes $\exp[ikn_{\mathrm{Si}}z(x,y)]$ obtained in the second medium, i.e., Si, while its strengths are controlled by a scattering potential which is proportional to $(n_{\mathrm{Si}}^2 - n_{\mathrm{ZnO}}^2)$. Both, the phase model and the Born-Fraunhofer approximation assume that the local phase variation of light defines the scattering. In the three models above, $k = 2\pi/\lambda$ is the wavenumber *in vacuo* and z denotes the height profile as obtained by AFM. Fourth, a ray tracing approach,[11] where geometrical optics is applied considering Snell's law. Here, light refraction defines the scattering behavior. These four models are compared to rigorous diffraction theory, whose reliability was already shown in Fig. 2 for the polished μc-Si:H layer by experimental results. The extinction coefficient of μc-Si:H was set to zero to allow a propagation into a half space.

The results for AFM scans 1 and 2 are shown in Figs. 3(a) and 3(b), respectively. It is found that the Born-Fraunhofer approximation shows the highest scattering intensities at larger angles. The grating model and the phase model agree well with the rigorous diffraction theory for the whole angle range. The ray tracing model predicts larger angles for the maximum intensity. For this surface texture, light scattering properties of the rough ZnO:Al/μc-Si:H

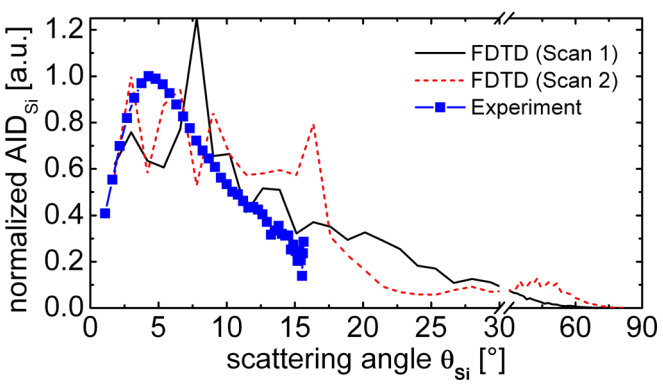

FIG. 2. (Color online) $\mathrm{AID}_{\mathrm{Si}}$ determined from experiment and rigorous diffraction theory based on the two AFM topographies shown in Figs. 1(b) and 1(c) at $\lambda = 780$ nm.

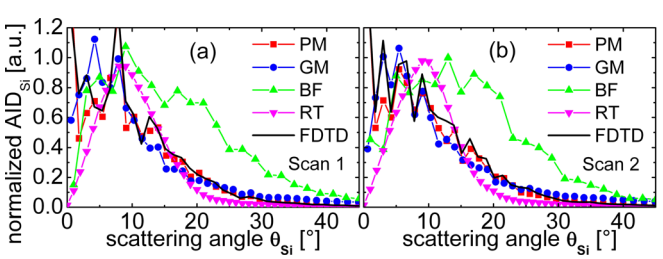

FIG. 3. (Color online) $\mathrm{AID}_{\mathrm{Si}}$ determined from rigorous diffraction theory (FDTD), phase model (PM), grating model (GM), Born-Fraunhofer approximation (BF), and ray tracing approach (RT) at $\lambda = 780$ nm for AFM scan 1 (a) and scan 2 (b).

interface can be well described by both the phase model and the grating model, while the ray tracing approach as well as the Born-Fraunhofer approximation overestimate the dominant scattering angles.

The agreement of the grating model to the rigorous diffraction theory means that the studied surface texture acts as an optical grating that diffract light into the first order. This assumption might not be reasonable for other surface textures. For the sample studied here, the phase model also shows a good agreement. Therefore, a deeper investigation for textures, where the different models predict different scattering, will help to better evaluate the dominant physical mechanism and the applicability of the models. All scalar scattering models, considered in this letter, assume a coherent incident plane wave. For structures with larger surface features, this condition might not be fulfilled. Then, the system is expected to be dominated by geometrical optics with a better agreement to the ray tracing approach. A crater-like interface with lateral feature sizes mostly below 1 μm is within the coherent limit also inside the Si, where the effective wavelength is lowered by a factor of $n_{Si} = 3.70$ (at $\lambda = 780$ nm). Hence, the surface profile and the surrounding media define the usability of the different simplified scattering models. Only by rigorous approaches, a general agreement to experimental results can be achieved. The comparison to simplified models identifies the dominant physical mechanism of light scattering for a given surface texture.

In conclusion, an experimental approach to investigate the angular resolved light scattering into the absorber material of a thin-film solar cell was introduced. These results were compared to simulations based on rigorous diffraction theory showing good agreements. Based on these results, different established scattering models were proven for a TCO/Si interface, where two half spaces are assumed and the results from rigorous diffraction theory taken as reference. The grating model and the phase model show good agreement to rigorous diffraction theory. The ray tracing approach predicts the maximum in the AID_{Si} at larger angles, and the Born-Fraunhofer approximation overestimates the AID_{Si} at larger scattering angles. Since the sizes of the typical texture features define which physical mechanism dominates the scattering behavior, textures that significantly differ from those studied in this letter have to be investigated in the same manner to prove the validity of the scattering models. The experimental technique will give essential information about the scattering into the absorber material of a thin-film solar cell. This information is much more important than light scattering properties into air.

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