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In-situ determination of the effective absorbance of thin μc-Si:H layers growing on rough ZnO:Al

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Abstract In this study optical transmission measurements were performed in-situ during the growth of microcrystalline silicon (μc-Si:H) layers by plasma enhanced chemical vapor deposition (PECVD). The stable plasma emission was used as light source. The effective absorption coefficient of the thin μc-Si:H layers which were deposited on rough transparent conductive oxide (TCO) surfaces was calculated from the transient transmission signal. It was observed that by increasing the surface roughness of the TCO, the effective absorption coefficient increases which can be correlated to the increased light scattering effect and thus the enhanced light paths inside the silicon. A correlation between the in-situ determined effective absorbance of the μc-Si:H absorber layer and the short-circuit current density of μc-Si:H thin-film silicon solar cells was found. Hence, an attractive technique is demonstrated to study, on the one hand, the absorbance and the light trapping in thin films depending on the roughness of the substrate and, on the other hand, to estimate the short-circuit current density of thin-film solar cells in-situ, which makes the method interesting as a process control tool.

1 Introduction

Light trapping in thin-film silicon solar cell devices plays a major role at today's research activities to increase the efficiency of solar modules. Rough surfaces are commonly used to scatter the light in the thin absorber layer of the solar cell. Therefore, random textures are used which can be realized by e.g. the transparent conductive oxide (TCO) window and front contact layer applied in thin-film silicon solar cells [1–5]. By light scattering and diffraction, the path of the light traversing through the absorber layer is enhanced and the light absorption is increased [6]. It was shown that different types of TCO with different roughnesses are able to scatter and diffract light in a different way such that the scattering property can be varied over a broad range [6–8]. This is important to adapt the interface texture to different types of solar cells, e.g. single junction or multi junction concepts, whereby always the optimal texture for different absorber layer thicknesses is used.

In the present study, we show a way to determine the effective absorbance of silicon thin-films, which depends on the light path enhancement and thus on the substrate texture. The transient signal of transmission measurements, which were performed in-situ during the deposition of microcrystalline silicon, was used. In our earlier work, we have demonstrated that these transmission measurements can be used to determine the thickness and the crystallinity of the growing layers [9]. Additionally, we showed that especially by in-situ controlling the thickness of the deposited absorber layers, it was possible to fabricate tandem solar cells in which the top and bottom cell generate the identical short-circuit current density which is referred to as "current matched" [10]. In these two studies, it was demonstrated that in-situ transmission measurements are very interesting to use them as process control in the industrial production line. In the present study, we focus on the interpretation of the transient transmission signal which can be correlated to the effective absorbance of the silicon absorber layers and can be used to estimate the short-circuit current density of solar cell devices.

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2 Experimental setup

Figure 1 shows the setup of the in-situ transmission measurement. Plasma enhanced chemical vapor deposition (PECVD) processes were used to fabricate silicon thin films integrated in solar cell devices [11]. A parallel plate

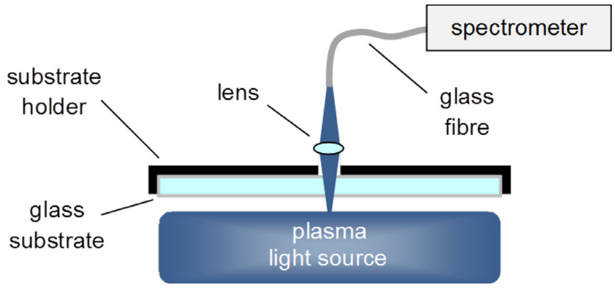

Fig. 1. Experimental setup. PECVD process including an optical port on the backside of the substrate carrier for transmission measurements. The plasma emission is used as light source.

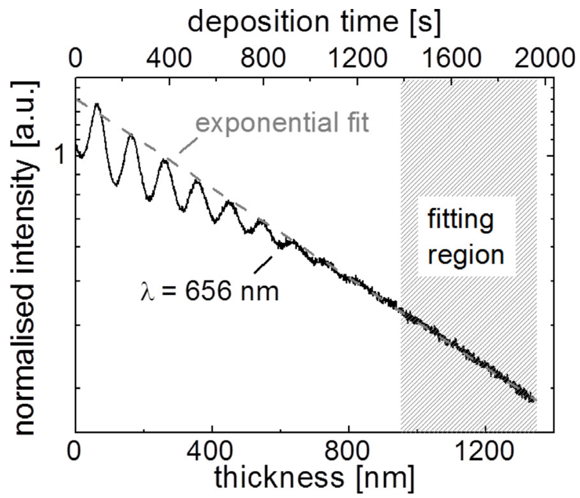

Fig. 2. Transient transmission of the plasma emission line at 656 nm through a growing μc-Si:H film. An exponential fit function (dashed curve) was applied at the last 400 nm of the μc-Si:H layer deposition (see gray fitting region).

reactor with a showerhead electrode was used to deposit the intrinsic microcrystalline silicon (μc-Si:H) absorber layers at which the transmission measurements were performed. The plasma emission was used as light source for this purpose. The plasma emission spectrum contains dominant peaks at wavelengths of 656 nm, 760 nm and 890 nm which were used in this study. An example of such emission spectrum can be found in reference [10]. A hole in the backing plate of the substrate holder allows the light collection from the plasma via a lens through the substrate and the growing silicon thin films. The lens focusses the light into a glass fiber which is connected to a spectrometer.

As substrate for the solar cell deposition, 10×10 cm^2 glasses (Corning Eagle X) were used on which ZnO:Al layers were sputtered. The ZnO:Al layers were etched in liquid HCl solution for texturing [4]. Thus, the textured ZnO:Al films serve as front electrode of the solar cell and also as scattering layer to increase the light absorbance in the solar cell. By varying the etching time between 5 s and 40 s, various random surfaces textures were fabricated to study the influence of the surface texture on the effective absorbance of the silicon thin films.

Microcrystalline thin-film silicon solar cells in p-i-n configuration were fabricated to investigate the influence of the in-situ determined effective absorbance of the intrinsic layers on the short-circuit current density of the device. In the PECVD processes, a gas mixture of silane and hydrogen was used. The excitation frequency was 13.56 MHz at a power of 1 W/cm^2. The deposition pressure was 10 Torr at a silane concentration (SC_i = $[SiH_4]/([SiH_4]+[H_2])$) of 0.58% resulting in a Raman-crystallinity of about 60% for the intrinsic μc-Si:H layers. The p-type and n-type layers of the solar cells were deposited in separate chambers with cross flow configuration using trimethyl-boron and phosphine as doping gas, respectively. A ZnO:Al/Ag layer stack was deposited through a shadow mask as back reflector and electrical contact resulting in active solar cell areas of 1×1 cm^2.

The ZnO:Al surface textures, on which the solar cells were deposited, were characterized by atomic force microscopy (AFM). The Raman-crystallinity of the solar cell material was measured after the deposition of the whole layer stack through the n-layer using a laser wavelength of 647 nm [12]. The microcrystalline silicon solar cells were electrically characterized under AM1.5 illumination.

3 Results

Figure 2 shows an example of the transient optical transmission signal at a wavelength of 656 nm which was recorded during the deposition of μc-Si:H on an etched ZnO:Al substrate with a root mean square roughness of 110 nm. The shape of the transient transmission curve is known from our earlier studies [9, 10]. We show it here again to briefly summarize the signal shape which helps to follow our further evaluation and the discussion. The lower axis of abscissae shows the film thickness of the μc-Si:H layer and at the upper axis of abscissae the actual deposition time of the μc-Si:H layer is provided. Using the maxima and minima of the curve, the deposition time can be transferred to film thickness as described in reference [9]. The axis of ordinates shows the (normalized) intensity of the plasma emission at a wavelength of 656 nm. The signal is normalized to the first minimum of the curve. The fringes of the transient transmission signal result from interference effects due to multiple reflections inside the growing silicon layer. The exponential decrease of the transmission signal intensity is due to the absorbance of the silicon which increases with increasing film thickness.

By using an exponential function, the transient transmission signal, which is plotted as function of film thickness, was fitted for the last 400 nm of the silicon deposition (see fit in Fig. 2 – a detailed discussion of the fit will follow in the discussion section). With the fit function the effective absorption coefficient (α_{eff}) of the growing layer was determined using the Lambert-Beers law:

$$I = I_0 e^{-\alpha_{\text{eff}} d} = I_0 e^{-w\alpha d}. \quad (1)$$

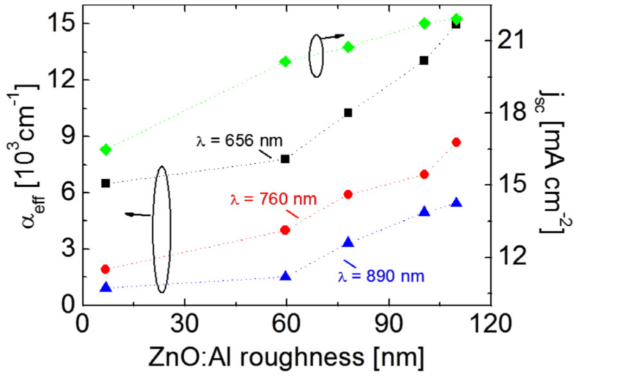

Fig. 3. Effective absorption coefficient α_{eff} and short-circuit solar cell current density j_{sc} as a function of RMS roughness of the different ZnO:Al layers. The thickness of the cells is ~1.4 μm.

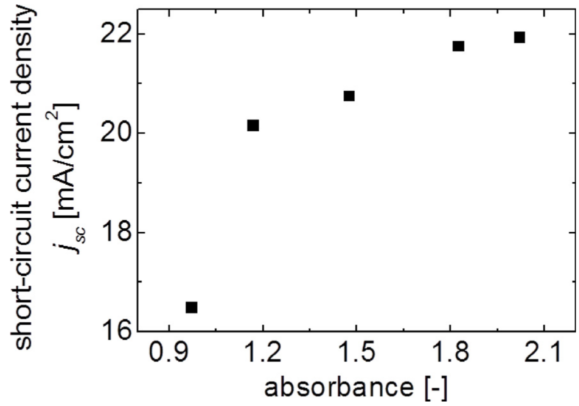

Fig. 4. Short-circuit current density j_{sc} as a function of effective absorbance $= \alpha_{\text{eff}} d$. The short-circuit current density of the cells was measured under standard AM1.5 illumination (ex-situ). The effective absorption coefficient was determined using exponential fits of the transient transmission signal at a wavelength of 656 nm (in-situ). The final thickness d of the solar cells was measured after deposition using a surface profiler (Dektak).

In equation (1) I is the signal intensity, I_0 is the initial signal intensity, d is the film thickness and α_{eff} is the effective absorption coefficient which is the product of the absorption coefficient α of the growing silicon (at a distinct wavelength of 656 nm) and the light path enhancement factor w which derives from the scattering of the light by the textured ZnO:Al layer. The light path enhancement factor w is known from reference [6] and will be further discussed later.

In classical reflection-transmission (RT) measurements, equation (1) is transformed into:

$$\frac{T}{1-R} = e^{-w\alpha d}. \tag{2}$$

For layer thicknesses d large enough to prevent effects of light coherence, a constant reflection R can be assumed. Therefore, the light path enhancement factor w can be determined with the transmission signal T only.

In the experiment five different ZnO:Al layers on glass substrates with different root mean square roughnesses (6.7 nm, 69.5 nm, 78.0 nm, 100.4 nm and 109.8 nm) were investigated. On the different ZnO:Al layers, μc-Si:H solar cells were fabricated. During the deposition of the intrinsic absorber layer, in-situ transmission measurements were performed. After fabrication, the solar cells were characterized using a class A sun simulator.

Figure 3 shows the effective absorption coefficient α_{eff} of the intrinsic absorber layer as a function of ZnO:Al roughness for three different wavelengths. Additionally, the short-circuit current density j_{sc} of the solar cells as a function of ZnO:Al roughness is plotted. (Please note, that the roughness is used in this study to distinguish the different ZnO:Al films by a simple parameter, though the roughness is known to be insufficient to describe the surface texture completely [4].)

By increasing the ZnO:Al roughness, the value of the effective absorption coefficient α_{eff} increases for all wavelengths. This is caused by the enhanced light scattering effect of rougher interfaces and, thus, an enhanced light path represented by w in equation (1). Also, the short-circuit current density j_{sc} of the solar cells is increased by increasing the roughness of the ZnO:Al which is in agreement with earlier studies [4,6].

With this experiment, it is demonstrated that an increasing absorbance of the i-layer of the solar cell with increasing substrate roughness can be detected in-situ and that it is possible to correlate the increasing absorbance to an increasing short-circuit current density of the solar cell device. Figure 4 shows the correlation plot between these two parameters. The absorbance (here) is the product of the effective absorption coefficient determined at a wavelength of 656 nm and the thickness of the solar cell which was measured using a surface profiler (dektak). The correlation shown in Figure 4 points out the attractiveness of the in-situ method for a j_{sc} prediction during the deposition of solar cells. It is interesting for an active process control in which the thickness of the solar cell absorber layer can be controlled to generate a distinct current which is important, e.g. for the fabrication of multi-junction solar cells [10].

4 Discussion

The first part of the discussion covers the light path enhancement factor w which is derived from the fitting equation of the transient transmission signal (see Fig. 2 and Eq. (1)). The wavelength dependent absorption coefficient α is an intrinsic material property. In transmission and reflection measurements for example, α is derived from well defined, flat sample geometries. The present experiment on textured surfaces has shown a dependence of the effective absorption coefficient α_{eff} on the roughness of the substrate surface. Since rough surfaces scatter the light into the silicon, the optical path length of incident light in the silicon is enhanced. In the past, it was shown that the light path enhancement factor w can be derived

Table 1. Light path enhancement factors as function of ZnO:Al roughness. The light path enhancement factors of three different wavelengths (656 nm, 760 nm, 890 nm) were calculated from in-situ transmission measurements ($w_{\text{in-situ}}$) and quantum efficiency measurements of the solar cells ($w_{\text{ex-situ}}$).

	656 nm		760 nm		890 nm	
ZnO:Al roughness [nm]	$w_{\text{in-situ}}$	$w_{\text{ex-situ}}$	$w_{\text{in-situ}}$	$w_{\text{ex-situ}}$	$w_{\text{in-situ}}$	$w_{\text{ex-situ}}$
6.7	1.3	1.36	1.6	2.31	3.1	3.09
59.5	1.56	1.61	3.33	3.96	5.09	5.81
78.0	2.05	1.77	4.92	4.49	11.08	6.61
100.4	2.61	1.92	5.80	5.45	16.51	8.49
109.8	2.99	1.97	7.22	5.97	18.13	9.16

from e.g. quantum efficiency (QE) measurements [6] according to:

$$w(\lambda) = -\frac{\ln(1 - QE(\lambda))}{\alpha(\lambda)d}. \qquad (3)$$

It was demonstrated that for certain textures, the light path enhancement factor increases with increasing surface roughness of the ZnO:Al, which is in agreement with the observations in this study. But here, the light path enhancement factor is derived from in-situ measurements during the deposition of the intrinsic absorber layer of μc-Si:H single junction solar cells (see Eq. (1)). Thus, the absorbance of only the absorber layer is measured using the in-situ method in contrast to QE measurements which are performed after the accomplishment of the complete solar cell device. Hence, the influence of the front contact layer, the back reflector and the doped layers of the solar cell device is not included using the in-situ transmission measurements and thus, e.g. parasitic absorption has no contribution. Furthermore, because only the change in the in-situ transmission signal during the deposition of the intrinsic absorber layer is evaluated, the initial reflectance of the solar cell device has no influence on the signal in contrast to the QE measurements were the initial reflectance, which is in the range of 10%, has an significant impact on the measurement signal. Another difference in the two experiments is given by the layer stack and the direction of light propagation. In the QE measurements, a complete solar cell with back reflector is specular illuminated from the glass side. This means that, for the rough samples, light scattering at the front contact occurs at an interface between TCO and silicon which morphology is only defined by the texture-etched TCO. In contrast to the QE measurement setup, at the in-situ transmission measurement setup the sample is illuminated from the silicon layer side by a diffuse (plasma) light source (see Fig. 1). Furthermore a layer stack without back reflector is investigated in-situ. Here, light scattering at the in-coupling to silicon occurs at an interface between air and silicon which morphology is not only defined by the texture-etched TCO, but also by the growth of the silicon layer [12, 13]. Light scattering properties are mainly defined by the interface morphology and the difference in refractive indices. Both are different in the two experiments, in particular for samples with large roughness. Furthermore, the in-situ measurement detects light which is scattered within the acceptance angle of the lens (see Fig. 1). Assuming a change in the scattering properties with increasing silicon layer thickness (e.g. by changing surface morphology), the portion of transmitted light, which is detected by the setup, modifies. This issue is also addressed below in the second part of the discussion.

Therefore, the light path enhancement factor $w_{\text{in-situ}}$ which is derived from the in-situ transmission measurements at single absorber layers is not the same as the light path enhancement factor $w_{\text{ex-situ}}$ which is derived from QE measurements at solar cell devices. Table 1 shows examples of values for $w_{\text{in-situ}}$ calculated from α_{eff} in comparison to values for $w_{\text{ex-situ}}$ calculated from QE measurements of the considered cells according to equation (3). The material specific absorption coefficient α was taken from photo thermal deflection spectroscopy measurements at μc-Si:H reference samples. The thickness d of the silicon was measured using a dektak surface profiler after the solar cell fabrication.

It is found that for the samples with a small roughness, the values of $w_{\text{in-situ}}$ agree well with the light path enhancement factors $w_{\text{ex-situ}}$, showing that for samples with low light scattering properties, any differences in the two experimental techniques are well compensated by the transient approach of the in-situ measurement. With increasing roughness, and therefore increasing light scattering, the light path enhancement factors determined by in-situ transmission measurement are higher than those determined by QE measurement. The higher difference in refractive index at the front interface in the in-situ experiment leads to an improved light scattering efficiency compared to the configuration in the QE measurement [14]. This effect is the strongest pronounced for the largest wavelength (890 nm), since light scattering has the strongest impact on absorption. Beside this effect, discrepancies in the light path enhancement factor at a wavelength of 890 nm for larger roughness can also be explained by an increased parasitic absorption in the front TCO. Although the effective absorbance of the intrinsic silicon layer is strongly increased, parts of this improvement are compensated in the QE measurement by the reduced light intensity that reaches the silicon due to the absorbance in the TCO which is increased as well with the roughness. Since the in-situ measurement is directly sensitive to the effective absorbance in the intrinsic layer, higher values are measured. In summary, different optical effects occur which are hard to fully take into account but which are reasonable to explain the discrepancies in the light path enhancement factor determined by the two experiments. Since a quantitative comparison of the light path enhancement factors is difficult, the main focus will be on the correlation of the effective absorbance, determined by in-situ measurements, and the short-circuit current density of the solar cells.

The second point of the discussion covers the fitting procedure of the transient transmission signal which leads to the determination of the effective absorption coefficient. The estimation of α_{eff} was found to be reasonable for the deposition of the final 400 nm of the silicon where equation (1) was applied and α_{eff} can be correlated to j_{sc} of the solar cells. In Figure 2, the region wherein the exponential fit was performed is highlighted. In addition, it can be observed in Figure 2 that the slope of the transient transmission curve does not follow a single exponential function over the whole thickness range and a single exponential term is not sufficient to describe the whole curve. We have to mention that only for the example shown in Figure 2 the exponential fitting curve strikes the maxima of the fringes in the beginning. For other roughnesses this trend is different. The different slopes of the transmission signal can be explained by the modification of the growing surface of the μc-Si:H layer on the rough ZnO:Al.

First, in the beginning of the deposition, only a very thin silicon film is on top of the ZnO:Al. In that case the two interfaces, ZnO/Si and Si/air, are coplanar to a large extend and most light coherently can interfere upon reflection at both interfaces, whereas after deposition of a certain silicon thickness, these two interfaces are more and more separated and the light rays do not necessarily hit a parallel interface after passing through the thick absorber layer. Thus, after a certain silicon thickness, the coherence is destroyed, which is represented in the transient transmission signal by vanished interference fringes and a changed signal slope (see Fig. 2).

Second, the texture of the silicon surface which is deposited on the textured ZnO:Al gradually changes with increasing the layer thickness of the silicon [12, 13]. At the beginning, when only a very thin silicon layer is deposited, the ZnO:Al texture is present also on the silicon surface due to the conformity of the thin silicon on the rough ZnO:Al. At the end of the deposition, the texture on the silicon surface changed in comparison to the original ZnO:Al texture due to the non-conformal growth of thick silicon layers. Thus, the light scattering texture on the silicon surface is different at the beginning and at the end of the absorber layer deposition.

Both effects change the scattering behavior events at the silicon surface and, consequently, the light path enhancement during the silicon deposition. At the same time, the spatial coherence of the light is destroyed and interference fringes vanish with thickness of the silicon layer, leading to a reflection at the air-silicon interface which is independent on the layer thickness. For the usually quite thick cells, the diminished interference fringes and the steeper decay of transmission is already present, thus, it is reasonable, in accordance to equation (2), to fit only the last part of the transient transmission signal for the calculation of the effective absorption coefficient and the light path enhancement factor and this actual optical system can be correlated to the photovoltaic parameters of the accomplished device.

In summary, it is demonstrated in this study that the transient transmission signal of the in-situ measurements can be used for light scattering and light trapping studies in silicon thin-film solar cells. The effective light absorbance of only the absorber layer is investigated what makes this method unique and useful. Additionally, the correlation between the in-situ measured effective absorbance of the solar cell absorber layer and the resulting short-circuit current density of the solar cell device makes this method attractive as an active process control in which for example the deposition time of the absorber layer can be adapted to produce solar cells generating a restricted current.

5 Conclusion

It is shown that in-situ transmission measurements are feasible for the determination of the effective absorbance of the deposited intrinsic μc-Si:H absorber layers. The measured effective absorption coefficient α_{eff} can be correlated to the short-circuit current density of solar cell devices. This demonstrates, on the one hand, a tool to study optical scattering and light trapping effects at growing thin-film silicon absorber layers and, on the other hand, the possibility of an active process control.

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