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TCO and light trapping in silicon thin film solar cells

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Abstract

For thin film silicon solar cells and modules incorporating amorphous (a-Si:H) or microcrystalline (μc-Si:H) silicon as absorber materials, light trapping, i.e. increasing the path length of incoming light, plays a decisive role for device performance. This paper discusses ways to realize efficient light trapping schemes by using textured transparent conductive oxides (TCOs) as light scattering, highly conductive and transparent front contact in silicon p–i–n (superstrate) solar cells. Focus is on the concept of applying aluminum-doped zinc oxide (ZnO:Al) films, which are prepared by magnetron sputtering and subsequently textured by a wet-chemical etching step. The influence of electrical, optical and light scattering properties of the ZnO:Al front contact and the role of the back reflector are studied in experimentally prepared a-Si:H and μc-Si:H solar cells. Furthermore, a model is presented which allows to analyze optical losses in the individual layers of a solar cell structure. The model is applied to develop a roadmap for achieving a stable cell efficiency up to 15% in an amorphous/microcrystalline tandem cell. To realize this, necessary prerequisites are the incorporation of an efficient intermediate reflector between a-Si:H top and μc-Si:H bottom cell, the use of a front TCO with very low absorbance and ideal light scattering properties and a low-loss highly reflective back contact. Finally, the mid-frequency reactive sputtering technique is presented as a promising and potentially cost-effective way to up-scale the ZnO front contact preparation to industrial size substrate areas.

Keywords: Thin-film silicon; Solar cells; TCO; Zinc oxide (ZnO); Light trapping; Light scattering; Modeling; Absorption losses

1. Background and motivation

For the further development and world-wide market growth of photovoltaic (PV) power generation, a reduction of the investment costs of the PV system is one of the major issues. One approach which promises a significant cost reduction are thin film solar cells based on hydrogenated amorphous (a-Si:H) or microcrystalline (μc-Si:H) silicon and their alloys (see references in Rath, 2003 and Schropp and Zeman, 1998). An integral part of these devices are the transparent conductive oxide (TCO) layers used as a front electrode and as part of the back side reflector. When applied at the front side, TCO has to posses a high transparency in the spectral region where the solar cell is operating and a high electrical conductivity. These are necessary but not sufficient properties of good TCO. For the so-called "superstrate" or p–i–n configuration (Fig. 1) where the Si layers are deposited onto a transparent substrate (e.g. glass) covered by TCO, two further conditions have to be fulfilled: strong scattering of the incoming light into the silicon absorber layer and favorable physico-chemical properties for the growth of the silicon. For example, the TCO has to be inert to hydrogen-rich plasmas or act as a good nucleation layer for growth of nano- or microcrystalline material. For all thin-film silicon solar cells, scattering at interfaces between neighboring layers

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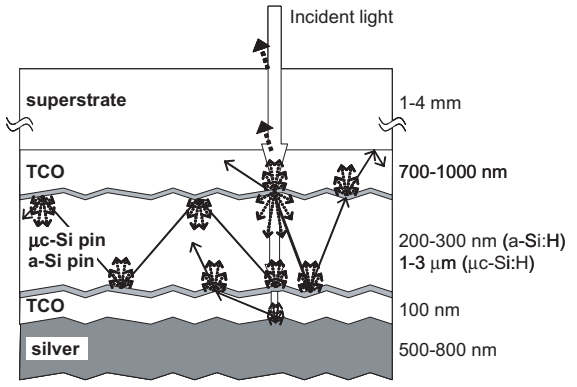

Fig. 1. Schematic sketch of the cross section of a silicon thin film p–i–n solar cell (a-Si:H and/or μc-Si:H) with rough interfaces. Thicknesses of the individual layers are typical values. The concept of light trapping is illustrated by the arrows representing incoming and scattered sun light. Different light paths and scattering events are sketched.

with different refractive indices and subsequent trapping of the incident light within the silicon absorber layers is crucial to gain a high efficiency (see light paths in Fig. 1). The reason is the absorption coefficient α of amorphous and microcrystalline silicon, which is shown in Fig. 2a for typical device grade material. Whereas α is high for short wavelength light, it strongly decreases towards longer wavelengths as the light energy approaches the optical band gap of the material (about 1.8 eV for standard a-Si:H and 1.1 eV for μc-Si:H). Due to the low band gap, light in μc-Si:H can be absorbed up to the near infrared spectral region allowing a better utilization of the solar spectrum. However, the band gap of single-, poly- or microcrystalline silicon is indirect thus limiting the absorption coefficient. As a result, in a thin film of not more than several microns thickness incoming light will not be completely absorbed during one single pass. On the other hand, to minimize process time and reduce light-induced degradation of amorphous Si, the absorber layer thickness should be as thin as possible. Hence, for all absorber materials optical absorption inside the silicon layers has to be enhanced by increasing the optical path of solar radiation.

For amorphous or microcrystalline thin film silicon solar cells light scattering is usually achieved by nano-texturing the front TCO electrodes (with a typical root-mean-square surface roughness of 40–150 nm) and/or nano-textured back reflectors. In the ideal case, these rough layers can introduce nearly completely diffuse transmission or reflection of light. Quantitatively, this is shown in Fig. 2b, where the total absorption $A = 1 - \exp(-\alpha \cdot d)$ (d = film thickness) was calculated for an a-Si:H and a μc-Si:H layer of typical device thickness (300 nm and 2 μm, respectively) for one single light pass using the absorption coefficients of Fig. 2a.

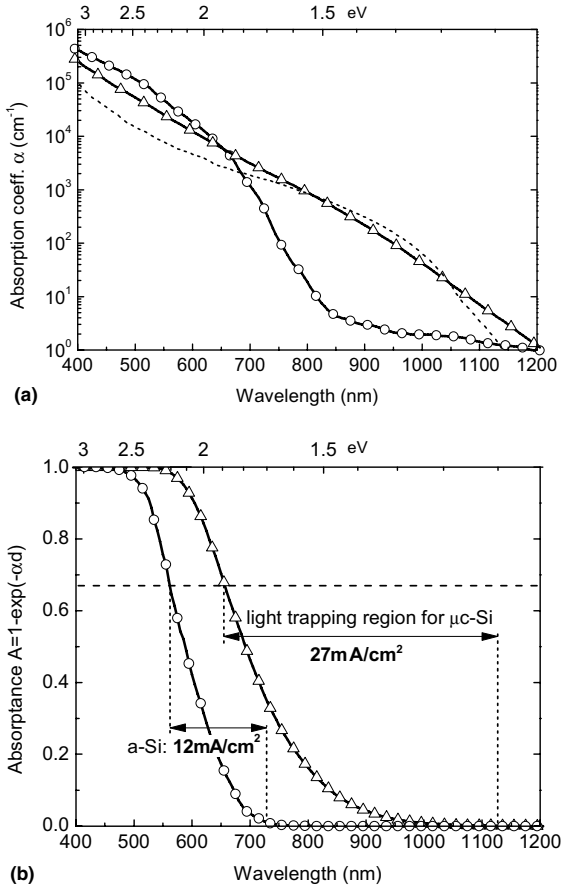

Fig. 2. (a) Absorption coefficient of amorphous (circles), microcrystalline (triangles) and crystalline (dashed line) silicon. (b) Light trapping spectral region for a typical amorphous (*i*-layer thickness 300 nm) and microcrystalline (2000 nm) silicon solar cell, with the maximum current value achievable in this region.

Fig. 2b illustrates the necessity of light trapping for long wavelengths and gives the maximum current values that can be gained in this region if ideal light trapping is realized.

TCO is also used between silicon and the metallic contact as a part of the back reflector to improve its optical properties and to act as a diffusion barrier (Morris et al., 1990; Beneking et al., 1994). Furthermore, applied in a-Si:H/μc-Si:H tandem solar cells ("micromorph" cell; Meier et al., 1994), TCO can be used as an intermediate reflector between top and bottom cell to increase the current in the thin amorphous silicon top cell (Fischer et al., 1996; Pellaton Vaucher et al., 1998; Yamamoto et al., 2002). Finally, nano-rough TCO front contacts act as an efficient antireflection coating due to the refractive index grading at the TCO/Si interface.

In conclusion, TCO plays an important role in the thin film silicon solar cell structure and has a decisive influence on the efficiencies presently achievable in state-of-the art amorphous, microcrystalline or "micromorph" solar cells. As the conditions for the best values of several material parameters have to be found in a multidimensional deposition space, optimizing TCO for solar cells constitutes a very complex problem and deserves further research, including ideally a simultaneous up-scaling of the research results to industrially relevant substrate sizes.

While in the past strong efforts have been taken to improve optical and electrical properties of the most widespread TCO materials tin oxide (SnO_2) and indium–tin oxide (ITO), we concentrate here more on the recent development of zinc oxide (ZnO) layers, which are more resistive to hydrogen plasmas, can be prepared by large area and potentially cheap techniques and consist of abundant materials. More specifically, focus will be on magnetron-sputtered aluminum-doped ZnO:Al, which is post-deposition textured in a wet-chemical etching step (Löffl et al., 1997; Kluth et al., 1999a). This technique offers the unique possibility to separate the electro-optical properties on the one hand and the light scattering behavior on the other hand and allows to produce light scattering TCO films for both p–i–n and n–i–p cells. More detailed information on preparation and use of sputtered and texture-etched ZnO in solar cells can be found e.g. in Löffl et al., 1997; Rech et al., 1997; Kluth et al., 1999b; Rech et al., 2002, Kluth et al., 2003. In the following, we study the light scattering properties and the light trapping mechanism in amorphous and microcrystalline silicon single and tandem junction solar cells. Focus will be put on p–i–n ("superstrate") devices, but all the modeling of light trapping in solar cells, with the help of the model *CELL* (Springer et al., 2002b), can be applied in the same way to n–i–p ("substrate") structures, deposited on opaque substrates like metal or metalized plastic foils. A general roadmap for using TCO to achieve highest solar cell efficiencies from an optical point of view will be outlined, and finally TCO up-scaling issues will be briefly discussed.

2. TCO materials and experimental results

2.1. TCO materials

As a rule of thumb, the sheet resistance R_{sq} of high-quality TCO should be not larger than about 10 Ω and the average absorption of TCO on highly transparent glass $A_{400-1100}$ between 400 and 1100 nm should be below about 6–7%. Such absorption values from optical transmission and reflection data are best measured with an index matching fluid to avoid errors due to the surface roughness of the TCO (Gotoh et al., 1987). However, the minimum requirements for the optical and electrical properties of the TCO depend on the structure and the absorber material of the solar cell. For amorphous silicon absorber layers a high transparency for visible light (wavelength $\lambda = 400$–750 nm) is sufficient, while for solar cells incorporating μc-Si:H the TCO has to be highly transparent up to the near infrared (NIR) region (400–1100 nm). This imposes certain restrictions on the carrier density n of the TCO material, as increased free carrier absorption leads to a reduction of IR transmission (see e.g. Hartnagel et al., 1995; Agashe et al., 2004). Fluorine-doped tin oxide films (SnO_2:F), which fulfill these requirements to a large extent, have been developed by Asahi Glass (Asahi Type U, Sato et al., 1992). SnO_2:F prepared by a CVD process is also the most commonly used TCO material for thin film Si solar module manufacturing (Beneking et al., 1999), but despite recent progress cost-effective SnO_2-coated glass substrates of the same quality as Asahi-U are still not being supplied as a standard substrate on large areas ($\sim$1 m^2).

Doped ZnO has been established as a successful alternative on laboratory scale. Examples are boron-doped ZnO prepared by low-pressure chemical vapor deposition (LPCVD) (van den Berg et al., 1993; Faÿ et al., 2000) or intrinsic and aluminum-doped films deposited by expanding thermal plasma CVD (Groenen et al., 2001). A very promising approach is the use of magnetron sputtering together with a post-deposition chemical etching step. The sputtering process leads to highly conductive and transparent but smooth ZnO:Al films. A simple chemical etching step in diluted acid yields a textured surface which can be adjusted to give optimal light scattering over a wide wavelength range. Typical R_{sq}-values of 5–8 Ω for films of 650 nm thickness and an absorption $A_{400-1100}$ of only 5–6% proof the high quality of these films.

The TCO surface also influences the electrical properties of the solar cell, as it determines the interface area and chemistry of the TCO/Si-p-layer interface. One major challenge in producing high-quality SnO_2 is the fact that usually surface roughness (and hence light scattering) and thickness are correlated. Often a certain minimum thickness is required to develop a surface of sufficient feature size and roughness, leading to an increased absorption of the TCO. Here, sputtered and texture-etched zinc oxide possesses the significant advantage that the surface roughness is *not* introduced during growth, but is achieved by a post-deposition etching step, leaving absorption and surface morphology largely independent from each other.

The light scattering ability of a TCO is determined by the feature sizes and shapes of the TCO surface. However, there is still no general coherent description linking the structural properties of the statistically rough ZnO surface with its light trapping behavior in the finished

solar cell, although a number of experimental studies have been performed and empirical relations have been found (Kluth et al., 1999b; Frammelsberger et al., 2000; Müller et al., 2002; Rech et al., 2003; Hüpkes et al., 2003, 2004). As an example for two typical TCO structures, Fig. 3 shows SEM images of APCVD-grown SnO_2:F (Asahi type U) and magnetron-sputtered and texture-etched ZnO:Al. Surface structures of ZnO materials prepared by other deposition techniques can be found in the respective publications (Faÿ et al., 2000; Groenen et al., 2001). Particularly striking is the fact that the average feature sizes of SnO_2 (around 200 nm in lateral direction, root mean square roughness $\delta_{rms} = 40$–50 nm) are significantly smaller than for the rf magnetron-sputtered and etched ZnO (500 nm–1 μm, $\delta_{rms} = 80$–120 nm), whereas the structures perform quite similar when applied in amorphous silicon solar cells (Rech and Wagner, 1999). Larger feature sizes become, however, important when light of longer wavelengths is to be scattered as necessary for microcrystalline silicon (μc-Si:H) absorber layers, which absorb sunlight up to about 1100 nm. In fact, it has been shown that magnetron-sputtered and texture-etched ZnO performs particularly well in μc-Si:H single junction or a-Si:H/μc-Si:H tandem solar cells (Vetterl et al., 2000; Müller et al., 2001a). Applying these ZnO films, stable efficiencies of 11.2% for an a-Si:H/μc-Si:H tandem solar cell and 10.1% for an a-Si:H/μc-Si module (aperture area 64 cm^2) have been achieved (Repmann et al., 2003). Recent efforts of TCO manufacturing companies point in the direction of increasing average feature sizes (Kambe et al., 2003; Toyama et al., 2003).

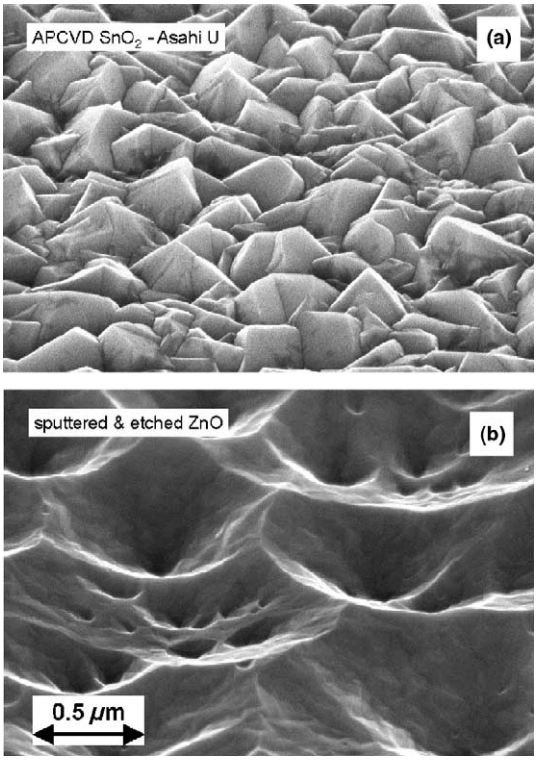

Fig. 3. SEM images of two typical surfaces of CVD-grown SnO_2:F (Asahi Glass Co., type U) and rf magnetron-sputtered and texture-etched ZnO.

In addition, one has to make sure that the front side encapsulation of the solar module, i.e. the glass sheet for devices in superstrate structure, is also highly transparent for NIR light, which can be realized by using glass with low-iron content (Hirata et al., 2001; Uchino et al., 2000). For example, typical absorption values of 3 mm thick glass at a wavelength of 1000 nm are 2–3% for low-iron glass as compared to 10–11% for standard float glass.

2.2. Film preparation and characterization

For the following experiments the ZnO films were prepared either by rf (Kluth et al., 1999a) or by dc (Müller et al., 2001b) magnetron sputtering from ceramic ZnO:Al_2O_3 targets. The ZnO-films, which are smooth in the as-deposited state, undergo a wet-chemical etching step in diluted acid (usually 0.5% HCl) yielding a rough and light scattering surface (root mean square roughness $\delta_{rms} = 80$–120 nm typically).

The p–i–n solar cells were deposited by plasma enhanced chemical vapor deposition (PECVD) at 13.56 MHz excitation frequency in a system for 30×30 cm^2 substrate size and with diode-type electrode configuration (Repmann et al., 2000, 2002).

TCO films were characterized by scanning electron microscopy (SEM), optical transmission and reflection measurements and by four-point probe sheet resistance (R_{sq}) and Hall effect measurements. For the solar cells we additionally measured quantum efficiency (QE) and illuminated I–V-curves using a class A double light source solar simulator (Wacom-WXS-140S-Super). For some of the cells, QE was measured without bias and under reverse bias conditions (−0.5 or −1.0 V), which yielded a relative difference in short circuit current of only 1% indicating excellent current collection. In the light trapping spectral region, the light beam undergoes multiple passes through silicon and ZnO layers and multiple reflections at the back reflector. Therefore, the limitations for a residual absorption in supporting layers (front TCO, back reflector, glass) are very strict and the absorption loss in each layer (or at the back reflector) has to be measured with a high precision. Photothermal deflection spectroscopy (PDS) (Amer and Jackson, 1984) was used for such measurements (Deng and Narasimhan, 1994; Springer et al., 2004), because it yields a high precision in the case of low absorption.

2.3. Influence of front side TCO

Fig. 4 demonstrates the influence of TCO surface roughness for two μc-Si cells deposited on glass/ZnO superstrates. One cell was deposited onto a smooth 490 nm thick ZnO superstrate without etching. The second one was prepared on another piece of this ZnO/glass substrate which was etched for 30 s to obtain a textured surface. Here the resulting ZnO thickness was 330 nm. Fig. 4 shows the QE of these two cells as well as the $(1-R)$ curves (R = total cell reflectance). The difference between $(1-R)$ and QE curves is a measure for the optical absorption losses in ZnO, back reflector and photovoltaically non-active Si layers (doped p, n) supposing that all photoexcited charge carriers in the undoped μc-Si layer are collected. As expected, the cell on rough ZnO is superior to the smooth cell. The main gain in generated short circuit current density (j_{sc}) comes from the red/IR region due to introduced light trapping. Additionally, QE is increased due to an antireflection effect (Frammelsberger et al., 2000). Note, however, that the difference between $(1-R)$ and QE is also increased by the rough TCO substrate pointing to higher absorption losses due to multiple light passes (see Section 3). Note also that, for the textured cell, interference fringes are weak but still visible. This shows that part of the light is not scattered and hence there is still room for further improvement.

Optical transparency and electrical conductivity of front TCO are usually closely related. Both thinner TCO layers and lower free carrier concentration n reduce absorption in the visible (VIS)/IR region, but at the same time the sheet resistance R_{sq} often increases. This is illustrated in Fig. 5 where the absorption coefficients of doped ZnO:Al ($n = 4$–6×10^{20} cm^{-3}) and undoped ZnO ($n \sim 5\times10^{18}$ cm^{-3}) films are plotted. To study the effect of thickness experimentally, μc-Si cells were deposited on ZnO of three different initial thicknesses yielding films with 330, 870 and 1370 nm final thickness and similar surface roughness after 30 s of etching in HCl (Fig. 6). The thick substrate has a lower R_{sq}, which increases FF due to reduced series resistance losses (Table 1). However, lower optical transparency leads to lower

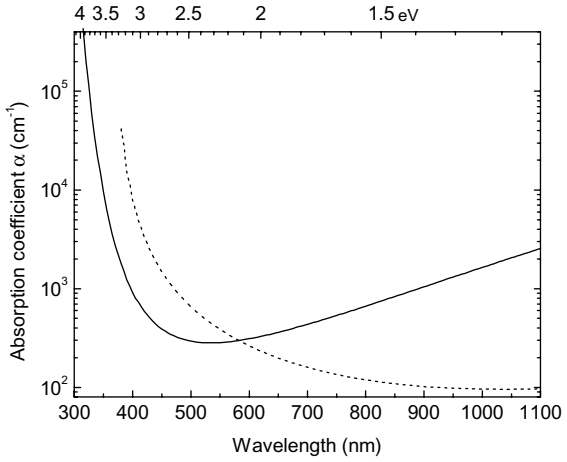

Fig. 5. Absorption coefficient of highly conductive Al-doped front contact ZnO, rf-sputtered from a ZnO:Al_2O_3 target ($\rho = 3$–5×10^{-4} Ω cm, $n = 4$–6×10^{20} cm^{-3}, $\mu = 30$–40 cm^2/V s, solid line) and of less conductive undoped ZnO as used for the interlayer ($\rho = 0.2$ Ω cm, $n = 3$–7×10^{18} cm^{-3}, $\mu = 5$–10 cm^2/V s, dashed line). In the UV region doping shifts the absorption edge to a higher energy due to Burstein–Moss shift, in the red/IR region the absorption is increased in the doped sample by free carrier absorption. The optical data shown in Fig. 5 were used in the calculations.

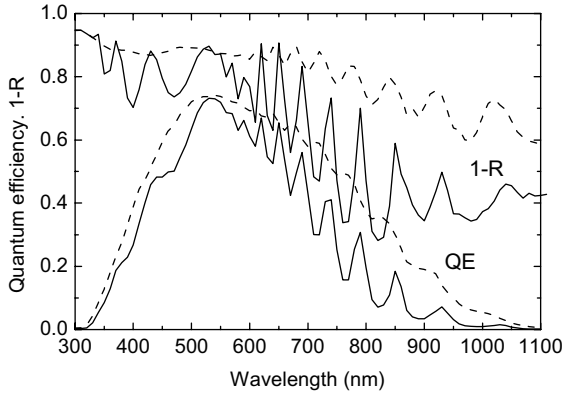

Fig. 4. Quantum efficiency QE and $(1-R)$ curves of μc-Si pin solar cells (i-layer thickness 1.2 μm, ZnO/Ag back reflector) on thin texture-etched (dashed lines) and smooth ZnO (solid lines). Resulting short-circuit densities are 20.8 and 15.3 mA/cm^2 respectively. R = total cell reflectance.

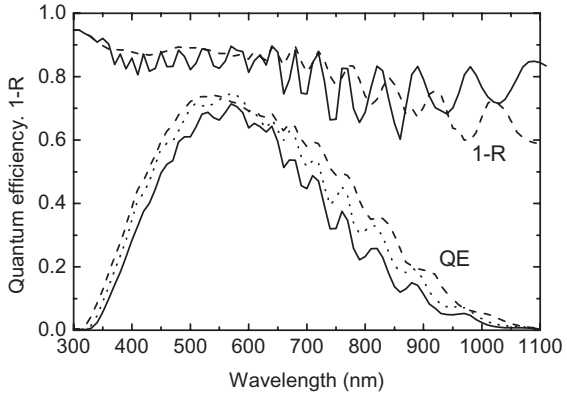

Fig. 6. Quantum efficiency QE and $(1-R)$ curves of μc-Si pin solar cells (i-layer thickness 1.2 μm, ZnO/Ag back reflector) for front ZnO with different thickness: 330 nm (dashed lines), 870 nm (dotted) and 1370 nm (solid). R = total cell reflectance.

Table 1
Short circuit current density j_{sc} and fill factor FF of μc-Si pin solar cells (*i*-layer thickness 1.2 μm, ZnO/Ag back reflector) for front ZnO with different thickness (values after 30 s of etching)

Initial R_{sq} (Ω)	Etched ZnO thickness (nm)	j_{sc} (mA/cm^2)	Fill factor (%)
10.1	330	20.8	68
4.4	870	18.5	72
2.7	1370	16.6	75

QE and j_{sc}. Comparing the thin and the thick substrate, optical losses are mostly in the red/IR region ($\lambda > 600$ nm) due to free carrier absorption in the front ZnO (Fig. 5). In this wavelength region the current density loss Δj_{sc} as compared to the thin ZnO is ≈3 mA/cm^2. This is enhanced by multiple passes of scattered light within the solar cell (compare cells with and without light trapping in Fig. 4). Smaller losses ($\Delta j_{sc} = 1$ mA/cm^2) are observed in the short wavelength region ($\lambda < 600$ nm). This experiment demonstrates the conflicting requirements of good electrical and optical TCO properties and the necessity of careful optimization with respect to each other.

2.4. Influence of back reflector

For effective light trapping, a highly reflective back contact (BR) is necessary when the average photon is reflected many times at the back side of the solar cell. To satisfy this optical condition together with sufficient electrical conductivity of the back contact, aluminum and silver are commonly used materials as metal back reflectors. Often aluminum is preferred in an industrial module production due to its lower price and better adhesion properties. Introducing a TCO layer between silicon layers and metal increases the reflectivity of the back reflector (Morris et al., 1990; Beneking et al., 1994), which will be discussed in the following.

Four metal back reflectors (ZnO/Ag, Ag, ZnO/Al and Al) were compared from an optical point of view in Fig. 7 for a-Si:H and μc-Si:H pin solar cells deposited on texture-etched ZnO. The ZnO/Ag and the Al are the best and the worst BRs, respectively, having different reflectance over the whole light trapping region. Interestingly, in a-Si p–i–n solar cells (Fig. 7a) both Ag and ZnO/Al show a similar effect on cell performance, while in μc-Si cells (Fig. 7b) the Ag BR gives higher QE and consequently higher j_{sc} than ZnO/Al. Table 2 helps with the explanation. For a-Si cells, good reflectivity is important in the region between 550 and 750 nm (see Fig. 2). The reflectance at 600 nm (which we take as a measure) is very similar for Ag and ZnO/Al. On the other hand, for microcrystalline silicon cells good reflectivity of the BR is essential in the whole region 600–1100 nm. Within this wavelength range an aluminum interband absorption

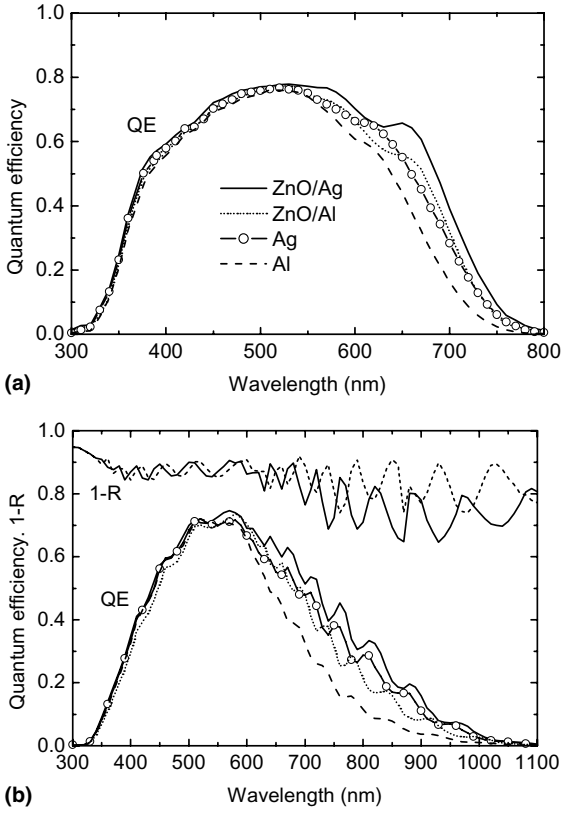

Fig. 7. Quantum efficiency QE and $(1 - R)$ curves of a-Si (a) and μc-Si (b) pin solar cells (*i*-layer thickness 350 nm and 1.2 μm, respectively) for different back reflectors: ZnO/Ag (solid lines), Ag (open circles), ZnO/Al (dotted line) and Al (dashed line). R = total cell reflectance.

occurs at $\lambda \approx 800$ nm (Palik, 1985), which causes enhanced absorption in the Al and hence low reflectivity for the ZnO/Al back reflector.

Table 2 shows the experimentally measured difference in j_{sc} for the different cells and BRs together with calculated internal reflectance values for the a-Si/BR structure at 600 nm and the μc-Si/BR structure at 800 nm. For the reflectance calculation in Table 2 literature data and Fresnel's equations were used. Two problems arise here: First, the optical properties of thin films and particularly their interfaces may significantly differ from bulk material data. It has been demonstrated via comparison of experiment and numerical analysis that the growth and the interface properties of Al or Ag are improved by deposition on ZnO, which also leads to a higher reflectivity (Stiebig et al., 1994). Secondly, the calculations are valid only for smooth interfaces, while a rough metal interface has a lower total reflectance than a smooth one (Beaglehole and Hunderi, 1970) due to a surface plasmon absorption (Raether, 1988; Harbeke, 1985; Fontana and Pantell, 1988). This situation arises

Table 2
Internal reflectance for the a-Si/BR structure at 600 nm (R_{600}) and the μc-Si/BR structure at 800 nm (R_{800}, BR = back reflector) calculated by using Fresnel's equations

BR	$R_{600\ nm}$	$R_{800\ nm}$	Δj_{SC} (mA/cm^2) (μc-Si:H)	Δj_{SC} (mA/cm^2) (a-Si:H)
ZnO/Ag	0.94	0.96	0.0	0.0
Ag	0.87	0.94	−1.0	−1.3
ZnO/Al	0.85	0.77	−2.6	−1.1
Al	0.70	0.65	−4.7	−2.0

Last two columns: Experimentally determined current losses Δj_{sc} in a-Si:H and μc-Si:H cells, respectively, with different types of back reflectors as compared to the ZnO/Ag back reflector.

in Si thin film solar cells on textured TCO, where the front surface roughness is either adapted by the silicon layers and "transferred" to the back contact or, as is sometimes the case for μc-Si, the absorber layer itself introduces additional roughness during growth. In fact, a reduced reflectance of a rough silver back reflector has been observed by Stiebig et al. (2000) in n–i–p and by Springer et al. (2000) and Springer et al. (2002a) in p–i–n devices. Furthermore, the absorption coefficients of textured Ag films similar to the ones incorporated in a real solar cell were measured by Springer et al. (2004) using photothermal deflection spectroscopy (Amer and Jackson, 1984). In the IR region, a rough ZnO/Ag interface has two to three times higher absorption A ($A = 1 - R$) than the smooth one. This is shown in Fig. 8, where the reflectivity of the layer system glass/ZnO(smooth or texture etched)/Ag/ZnO(30 nm) was measured from the ZnO(30 nm) side. Note that the 30 nm thick ZnO-layer was only used to protect the Ag from contact with ambient atmosphere, which we observed to cause changes in reflectance properties. The exact value will also depend on the surface topography and the quality of the ZnO/Ag BR, but the general trend of Fig. 8 should be always visible in the device.

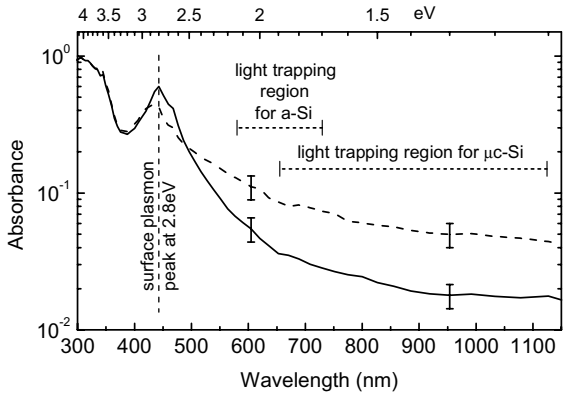

Fig. 8. Absorbance A of a smooth (solid line) and rough (dashed line) ZnO(30 nm)/Ag/ZnO back reflector measured by photothermal deflection spectroscopy (PDS) as a function of wavelength (lower scale) or light energy (upper scale). The 30 nm thick ZnO layer is only used to protect the underlying Ag from oxidation and samples were measured from the ZnO(30 nm) side. Error bars for the PDS measurement are also included.

3. Modeling of light trapping

A number of approaches attempting to predict the performance of thin film Si solar cells by theoretical modeling have been reported in literature (e.g. Stiebig et al., 1994; Leblanc et al., 1994; Hishikawa et al., 1997, 1999; Zeman et al., 2000; Krč et al., 2002; Springer et al., 2002b). In the optical model "CELL" (version 7), for silicon thin film solar cells developed at the Institute of Physics, Prague (Springer et al., 2002b), the coherent contribution of the multilayer structure is evaluated using wave theory taking into account scattering losses at each surface and interface. Then ray tracing of scattered photons is calculated up to the final absorption in any layer of the solar cell or photon loss due to reflection into air. This is schematically sketched in Fig. 1 for a thin film silicon p–i–n cell.

This approach allows to analyze all optical losses and the Monte Carlo method is a very suitable tool for this task. Experimentally determined optical constants of all materials and experimentally deduced absorption losses in the rough ZnO/Ag back reflector are used.

Only the photons absorbed in the intrinsic layer are considered as contributing to the photo generation of free electrons and holes. Assuming 100% carrier collection, absorbance in the intrinsic silicon layer equals the external quantum efficiency (QE). Finally, the short circuit current is obtained by multiplying QE with the AM 1.5 spectrum at 100 mW/cm^2 and the elementary charge.

In the optical model the following approximations are applied in order to describe the influence of the rough surfaces/interfaces: the effective media approximation (EMA) (Brass, 1995) for the description of antireflection properties of rough interfaces (graded index

of refraction) and the scalar scattering theory (Beckmann and Spizzichino, 1963; Poruba et al., 2000).

For high cell efficiency it is crucial to scatter as much of the light as possible into angles larger than the critical angle for total reflection in silicon (outside of the so-called "escape cone") to get the light trapped within the silicon absorber layer (Springer et al., 2002b). According to the scalar scattering theory the intensity of diffusely transmitted light depends on differences in the refractive indices of ZnO and Si and on the ratio of wavelength λ to root mean square surface/interface roughness δ_{rms} for the case of $L < \lambda$, where L is the surface correlation length (Beckmann and Spizzichino, 1963). Ideally, the angular distribution of scattered light inside the silicon layers should be estimated experimentally, which turns out to be very complicated and requires silicon surface polishing before measurement (Springer et al., 2002b). Therefore, a theoretical Lambertian distribution is used as an approximation.

3.1. Analysis of optical losses

Optical losses in an amorphous/microcrystalline silicon tandem cell (micromorph) prepared by PECVD with a sputtered ZnO intermediate reflector were analyzed by calculating external quantum efficiency QE, total reflectance R and absorbance in each layer (Fig. 9). Cell structure was glass (1 mm)/texture-etched ZnO:Al (500 nm)/a-Si:H pin (250 nm)/ZnO (100 nm)/μc-Si pin (2.8 μm)/ZnO (90 nm)/Ag. The glass/ZnO:Al superstrate was texture-etched in diluted (0.5%) HCl resulting in a residual film thickness of 500 nm after etching. The top cell is a standard amorphous silicon cell with a μc-Si/a-Si p-layer (total thickness about 15 nm) and a μc-Si n-layer (thickness about 20 nm). As there are no strong requirements on conductivity of the 100 nm thick ZnO interlayer, ZnO without Al-doping was used. The measured optical absorption coefficients of the a-Si:H and μc-Si:H absorber layers are the same as in Fig. 2a. The cell deposition was finished by a 90 nm thick undoped ZnO-layer and a silver back reflector which define the cell area of 1×1 cm^2.

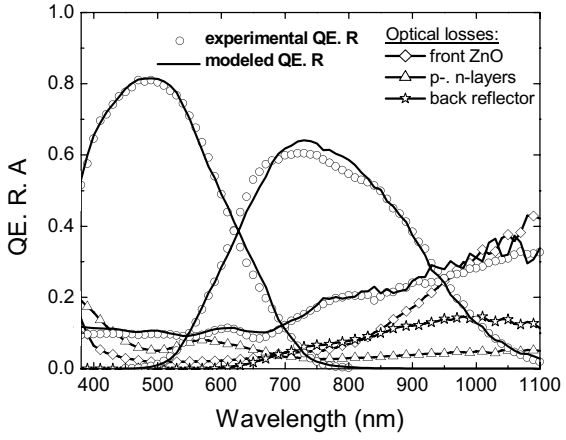

Fig. 9. Experimental and model data for the external quantum efficiency QE and total reflectance R of an amorphous/microcrystalline silicon p–i–n tandem solar cell with internal reflector. QE of the top a-Si:H and bottom μc-Si:H cell was measured separately using red and blue bias light illumination, respectively. Optical absorption losses A in each layer were computed with the help of the model.

In the light trapping region, above 850 nm, optical losses due to free carrier absorption in the front ZnO layer dominate. Residual interference effects can be seen because of incomplete light scattering, which is best visible in the reflectivity curves. Even if no antireflection coating is used, reflection is reduced due to the antireflective properties of rough interfaces (graded index of refraction). In the region of light trapping the absorption losses resulting from the rough Ag back reflector are also seen. The blue response is reduced by absorption in the non-ideal p-layer, but losses in the n-layer are low. Note that the importance of losses in non-photoactive layers of amorphous Si solar cells has also been highlighted recently in an analysis based on an experimental study (Lechner et al., 2000).

3.2. Road map for high-efficiency thin film silicon solar cells

Present data on the initial efficiency of micromorph tandem cells range between 12% and 14.5% (Meier et al., 2000; Rech et al., 2002; Yamamoto et al., 2002). Using the results of this analysis and the same cell structure as described above as a starting point, we applied our model to predict an efficiency limit for the micromorph cell structure and point out ways to further increase cell performance (Vanecek et al., 2003). For this analysis, we limited the top cell thickness to a value of less than 200 nm, which should guarantee a good stability of the tandem structure. This assumption follows recent results by Meier et al. (2002), who reported a stable behavior of the whole micromorph cell upon thickness reduction of the a-Si:H top cell below 200 nm. Keeping top cell thickness small is critical for device stability upon light exposure, as amorphous silicon undergoes a light-induced degradation process (Staebler and Wronski, 1977), while the microcrystalline bottom cell is considered reasonably stable under illumination.

Although the assumptions for the electrical quality of the absorber layers were quite optimistic using a fill factor of 73% of the 1.8 μm thick μc-Si:H bottom cell, further improvements in μc-Si:H film preparation are likely to reach a quality of both amorphous and microcrystalline Si absorber materials which could be sufficient to achieve the modeled stable solar cell efficiencies of 15% (Fig. 10). So one central result of the

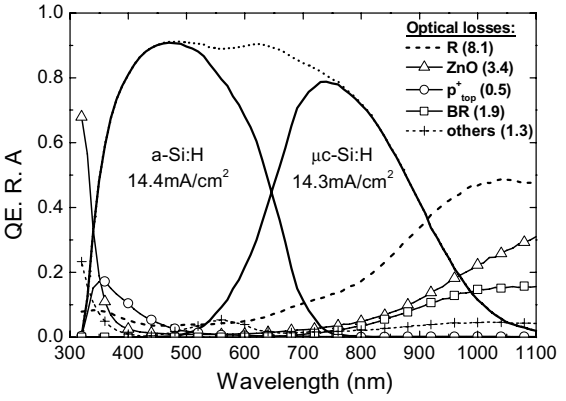

Fig. 10. Simulated quantum efficiency of a micromorph tandem cell with an intermediate reflector using optimized layers as described in the text. Assuming realistic values for fill factor (73%) and open circuit voltage (1.43 V) of the tandem structure yields an efficiency of 15%. The thickness of a-Si top and μc-Si bottom cell is 190 nm and 1.8 μm, respectively. Main losses (in mA/cm^2) are listed in the upper right corner of the figure (numbers in brackets). R: total reflection; ZnO: front TCO; p$^+$: front doped layer; BR: back reflector.

modeling is that the main bottleneck lies in the supporting layers (rough front TCO, doped p- and n-layers, back reflector). The results presented in Fig. 10 were obtained under the following assumptions:

(1) The front ZnO acts as an ideal Lambertian scatterer, scattering more than 90% of the incoming light intensity.
(2) Absorption losses in ZnO are reduced by a factor of three (compared to the Al-doped ZnO of Fig. 5, solid line) in the light trapping region by increasing the electron mobility and keeping carrier density low.
(3) A dielectric back reflector with an average reflectance of 96–98% is used.
(4) Reduced absorption losses in doped layers with an optical absorption coefficient in the defect region about 200 cm^{-1}.
(5) The superstrate glass has antireflection coatings (double layer at front side, single layer between glass and ZnO).
(6) An intermediate ZnO reflector of optimized thickness between the amorphous and microcrystalline cell compensates for the extremely thin (190 nm) a-Si:H top cell.

These assumptions are discussed in the following.

3.3. Front TCO

At the present state of development, the most important optical component for achieving 15% stable efficiency in a micromorph p–i–n cell is the quality of the front ZnO, which additionally has to exhibit a good homogeneity of these properties over large areas (~1 m^2).

Approaching 15% efficiency might not require any new discovery here, but definitely calls for a very intensive development and optimization. This is the area where the research on thin-film silicon solar cells (<2 μm of silicon absorber) should concentrate. For our modeling, the angular distribution of an ideal Lambertian scatterer was applied. In reality, this distribution will be different, but it is assumed that a distribution function favoring large (oblique) scattering angles over small ones would lead to more effective light trapping. This might be taken as one criterion for further optimization, but we underline here the necessity of a quantitative understanding of the correlation between surface texture and light scattering. Furthermore, it has been shown recently that it is possible to achieve electron mobilities of 45 cm^2/V s for sputtered ZnO on glass and thus reduce infrared absorption while keeping high-enough electrical conductivity (Agashe et al., 2004). Significantly higher mobilities have been reported for other TCO materials like Cd_2SnO_4 ($\mu \sim 80$ cm^2/V s; $E_g = 3.1$ eV, Coutts et al., 2000). However, a textured TCO material with these properties which is also feasible for the use in thin-film silicon solar cells still has to be developed.

3.4. Back reflector

In the infrared region of low-absorption coefficient (Fig. 2a) most of the light travels through the 200 nm thick layer of amorphous silicon (top cell) and is only partly absorbed even in the typically 1–3 μm thick microcrystalline silicon film (bottom cell). As the frequently applied thin ZnO/silver double layer back reflector adopts the roughness of the underlying layers, it becomes rough itself. This is good for light scattering and trapping (if the front ZnO is not scattering enough), but leads to an additional light absorption in the rough metal as has been shown in Section 2.4.

The effect of surface roughness on absorption losses in metal is substantial, especially in the case of rough silver and cannot be avoided. A dielectric back reflector could be a solution of this problem and could possibly be realized even in a very cheap way, e.g. using white mat paint. Other, more sophisticated, approaches apply for example one-dimensional photonic crystals, which can be designed for high omnidirectional reflectivity in a wide spectral range (Lee and Yao, 2003 and references therein).

3.5. Other components

Antireflection coating of glass is a standard industrial process and is therefore not discussed here. It should be

mentioned that the nano-rough interfaces between silicon and ZnO act as a very efficient antireflection coating due to the refractive index grading. All this is included in the model CELL.

The light, which is trapped in the silicon absorber passes through doped p- and n-layers. Even though the absorption losses here are lower than the typical loss in front ZnO or back reflector, there is still room for improvement. Hence, the optimization and the reduction of absorption losses in the p-layer is still an ongoing process, which is usually performed also with the goal of increasing the open circuit voltage V_{oc} by engineering the doped layers and their interfaces with the silicon absorber.

The intermediate ZnO reflector between the amorphous and the microcrystalline cell is a crucial component for cell stability in our study. It reflects light back into the amorphous silicon top cell raising its current level. This allows to match the bottom cell current with a reduced a-Si:H thickness, which increases the stability of the amorphous cell (Fischer et al., 1996; Meier et al., 2002; Yamamoto et al., 2002).

4. Up-scaling and transfer to production

Despite the highly successful application of sputtered and texture-etched ZnO-films for cells and modules on laboratory scale, up-scaling to a cost-effective and industrially feasible large area process still remains a challenging task. Up to now, ZnO sputtering for large area p–i–n (superstrate) solar module manufacturing is only applied at room temperature as interlayer between the silicon and metal layers at the back side of the cell structure to enhance the reflectivity of the back contact (see Section 2.4). Note, however, that for industrially produced modules in n–i–p (or "substrate") configuration, TCO in combination with metal or dielectric layers is also used to manufacture highly reflective and light scattering back contacts on the carrier substrate (Yang et al., 1997).

For the application of ZnO as front contact in an industrial module production, the ZnO layers have to be prepared with cost-effective techniques at high-deposition rates and with excellent homogeneity on large areas ($\geqslant 1$ m^2). The rf sputtering method from ceramic ZnO:Al_2O_3 targets is not feasible here mainly due to the high target costs and low deposition rates. Going from rf to dc sputtering, which is possible by using effective arc suppression electronics and highly dense ZnO:Al_2O_3 targets, allows for significantly higher sputter rates (Müller et al., 2001b), but this still leaves the problem of the relatively high-priced targets. Although approaches to reduce target production costs exist (Weigert, 2002), the most promising concept to face the cost problem is the use of reactive sputtering from cheaper metallic Zn:Al alloy targets, where oxygen is added to the argon sputter gas. As the process stabilization of a reactive dc sputter process is difficult, one uses a dual magnetron cathode, where an ac voltage in the kHz-range ("mid frequency", e.g. 40 kHz) is applied between the two targets, which then alternatingly serve as cathode and anode. This mid-frequency sputtering technique avoids several problems of a standard reactive dc process (Malkomes et al., 2001). ZnO films of high electrical and optical quality prepared at static sputter rates up to 9 nm/s have been demonstrated on small areas (Szyszka, 1999) and recently on larger areas of 0.6 m^2 (Müller et al., 2002). In the latter case, dynamic sputter rates were around 700–1000 nm mm/s, i.e. a 700–1000 nm thick film is deposited while the substrate is passing the dual targets at a carrier speed of 1 mm/s. Upon etching in diluted acid, these mf-sputtered films also develop a rough surface structure and have been applied in efficient solar cells (Müller et al., 2002) and in first solar modules (Müller et al., 2003; Hüpkes et al., 2004). Although both the homogeneity of the films and their structural properties, reflected in the surface texture after etching, have to be further improved, the mid-frequency sputtering technique is one of the most promising approaches to manufacture high quality but cost-effective large area ZnO-films for a next generation of thin film Si solar modules.

5. Summary

For highly efficient and cost-effective thin film silicon solar cells and modules, light trapping is essential, as it allows to reduce absorber layer thickness and increase device stability upon light soaking. For p–i–n (superstrate) devices, transparent conductive oxide (TCO) films with rough surfaces are applied in combination with highly reflective back contacts. These films provide efficient coupling-in of incident sun light by refractive index grading and light scattering at the TCO/Si interface to increase the path length of the light.

Especially for devices incorporating μc-Si:H absorber layers, magnetron-sputtered and texture-etched Al-doped ZnO films turn out as a highly promising TCO material. Attempts to up-scale these films to large areas at low-costs have shown first successes by using the high-rate mid-frequency reactive sputtering technique. Furthermore, sputtered and etched ZnO films are ideally suited for systematic experiments to study the influence of electrical, optical and structural TCO properties on device performance. Both experimental studies and theoretical simulations using the model *CELL* show that absorption losses in front TCO and back reflector as well as the light scattering properties of the front TCO have the strongest influence on device performance. ZnO/Ag double layers make up a highly reflective back

contact, but the unavoidable roughness of the back reflector also leads to additional absorption losses, which can possibly be avoided by using alternative reflector concepts. Highest efficiencies are achieved for a-Si:H/μc-Si:H tandem solar cells additionally incorporating an intermediate ZnO-reflector between top and bottom cell. By minimizing the losses, stable efficiencies of about 15% seem to be achievable for such a tandem device as predicted by the model. Strong R&D efforts should be put into a better understanding and an improvement of current TCO materials and light trapping schemes to push the experimentally realized thin film silicon solar cell efficiencies towards this value.

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