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Process monitoring of texture-etched high-rate ZnO:Al front contacts for silicon thin-film solar cells

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ABSTRACT

In this study angular resolved scattering measurements are used to monitor the surface texture properties of sputter-etched ZnO:Al and to identify the influences of process parameter changes on the surface texture. Variations in pressure and temperature are possible drifts in the industrial fabrication that can influence the ZnO:Al properties. Therefore – with respect to industrial relevance – this study focuses on the influence of these parameters. Deposition parameter dependent trends in the angular resolved scattering are studied. Correlations between changes in the results of the angular resolved scattering measurements and the performance of a-Si:H/μc-Si:H tandem solar cells are shown. Additionally, it is demonstrated that the measurements can be used to optimize the surface texture of ZnO:Al for single-junction μc-Si:H solar cells by adjusting the deposition conditions according to the previously identified trends.

1. Introduction

Sputtered and texture-etched ZnO:Al is a promising front contact in silicon thin-film solar cells [1–3]. For thin-film devices a well-defined surface texture determines the performance significantly [4,5]. The consistency of the texture is one of the predominant drawbacks in industrial production of sputter-etched ZnO:Al front contacts because of its strong dependence on the deposition conditions. Hence, even small changes or process instabilities can lead to changes in the surface texture [6].

Since changes in process temperature and pressure do not always occur deliberately, but are due to inhomogeneity especially in industrial large area fabrication, means of process control need to be established to facilitate a stable front contact quality. A common process control approach is the optical haze measurement which has been found to be insufficient to describe texture changes adequately with respect to the solar cell performance [7]. Atomic force microscope imaging allows a more detailed analysis of the surface texture [8] but this method does not fulfill the requirements for fast and cost effective process control. Angular resolved scattering (ARS) is an inexpensive, fast optical measurement that gives detailed information regarding the surface texture. Using two optical models the ARS results – called angular intensity distribution (AID) – can be interpreted as an image of inclination angles or feature sizes on the surface [9,10]. Besides the ability to describe the textured surface the AID has also proven to be a predictive tool for solar cell current. It has been shown already that the AID data can be used to correlate the large angle scattering intensity with the short-circuit current densities of thin-film silicon solar cells [7,11,12].

In the presented study ARS measurements are applied as a monitoring tool for high-rate sputtered, texture-etched ZnO:Al. Process parameter dependent trends in the AID are identified. a-Si:H/μc-Si:H tandem cell parameters are presented to confirm that the detectable differences in the ARS monitoring are significantly influencing the solar cell performance. Furthermore an example is presented showing that the known correlation between AID and single-junction μc-Si:H cell performance and the identified trends can be combined to optimize the surface texture of ZnO:Al for single-junction μc-Si:H solar cells.

2. Experimental details

All samples were deposited on commercially available float glass *ipawhite* (from *Interpane Glas Industrie AG*). In a first production step all float glass substrates were coated with an approximately 150 nm thick SiO_xN_y-interlayer. This interlayer has three functions: first of all, it is a barrier-layer to prevent Na^+-ions from diffusing out of the glass substrate. Secondly, the interlayer works as an anti-reflective coating between the glass substrate and the subsequently deposited transparent conductive oxide (TCO) layer. Finally, the interlayer also serves as a seed-layer for the growth of the TCO layer.

The SiO_xN_y-film is sputter-deposited in a reactive process using two planar *Sispa*® targets (by *Heraeus Materials Technology GmbH & Co. KG*). Oxygen and nitrogen were supplied to the chamber during the sputtering process. The excitation mode was magnetron-assisted mid-frequency at

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40 kHz with a power output of 4 kW. To simulate the use of multiple cathodes in an industrial in line system the samples were deposited in the so called dynamic mode with a moving carrier passing the cathodes multiple times.

In a second production step ZnO:Al was sputtered onto the interlayer using two 0.5 wt.% aluminum-doped ceramic tube targets. These rotatable tube targets were operated in magnetron configuration at mid-frequency mode with 40 kHz. The power output was kept constant at 10 kW to facilitate high-deposition rates of up to 62.6 nm∗m/min. Similar to the SiO_xN_y-coating the ZnO:Al deposition was executed in dynamic mode with the substrate carrier passing the targets multiple times during the deposition. While output power and gas flow stayed constant during the deposition processes, the temperature and pressure values were varied from one deposition to the next to simulate drifts and inhomogeneity in an industrial large area fabrication. These parameter changes were repeated for two series of different thickness of which the first, thicker series achieved film-thicknesses around 1280 nm (10 passes), whereas the second, thinner series yielded thicknesses around 900 nm (7 passes). All ZnO:Al layers and interlayers were deposited in a vertical inline sputter deposition system *VISS 300* by *von Ardenne Anlagentechnik Dresden GmbH* [6]. For further deposition parameter details see Table 1.

After deposition the ZnO:Al films were texture-etched in 0.5% HCl to roughen the surface. The etch step was conducted at room temperature for 120 s for all thick and 40 s for all thin ZnO:Al films. The textured ZnO:Al substrates were investigated using a scanning electron microscope (SEM) to gain insight into the surface features. Furthermore, the samples were inspected optically using an angular resolved light scattering set-up. The ARS set-up uses a green laser with a wavelength of 550 nm. The laser beam is chopped at a frequency of 230 Hz and then split into a reference beam and a measurement beam. The reference beam is led directly onto a silicon photodiode whereas the measurement beam passes through an aperture, striking the smooth glass side of the sample perpendicularly. A planar silicon photodiode is used to detect the transmitted, scattered light on the other side of the sample. The detector is free to move on a circular path at a fix distance around the sample. The step size of the moving detector can be set to 1° leading to an equivalent angular resolution of the measured signal. The measured photodiode current was used to calculate the normalized angular intensity distribution (AID) taking the solid angle into account.

The investigated ZnO:Al films were applied as front contact layer in a-Si:H/μc-Si:H tandem solar cells to study their influence on the solar cell performance. The silicon absorber layers were deposited in a single-chamber process. Details regarding the silicon deposition system and process are described elsewhere [13,14]. Room-temperature sputtered ZnO:Al and silver were employed as back contact and the area of the tandem solar cell was defined by laser scribing. The IV-characteristic of the solar cells were measured in a class A sun simulator using an AM 1.5 spectrum. Additionally, external quantum efficiency (EQE) measurements were conducted to identify the current distribution in the tandem device.

Table 1
Process parameters or parameter intervals of SiO_xN_y interlayer deposition and ZnO:Al deposition.

Process parameter	Unit	SiO_xN_y	ZnO:Al
Heater temperature	[°C]	25	430–500
Pressure	[Pa]	0.3	0.3–1
Power	[kW]	4	10
Φ_{Ar}	[sccm]	200	200
Φ_{O2}	[sccm]	18	–
Φ_{N2}	[sccm]	100	–
$V_{substrate\ carrier}$	[mm/s]	3	8
Passes		2	7 or 10

3. Results and discussion

3.1. Temperature series

Fig. 1 shows the SEM images of the deposited ZnO:Al surfaces after the texture-etch. The ZnO:Al samples in the left column show a heater temperature series from 430 °C to 500 °C with all deposited layers having a thickness around 1280 nm-thick series. The right column of SEM-images shows the same temperature series from 430 °C–500 °C for thin deposited films with a thickness of around 900 nm. The thick layer series shows a development in surface topography leading from rather smooth surfaces with small crater-like features at 430 °C to a very rough surface with large craters at 450 °C. Raising the process temperature even further the features seem to flatten again, leading to an almost smooth surface at 500 °C. For the thin layer series the behavior is similar. Small craters at 430 °C are increasing in diameter while the process temperature is increased leading to very distinct craters at 450 °C. Further increase in process temperature facilitates even larger craters, however, this is accompanied by a general flattening of the surface features.

Fig. 2 shows the AID results from 0° to 90° scattering angle of the two temperature series. Fig. 2A) displays the thick series whereas Fig. 2B) shows the thin series. The gray arrows within the graphs indicate the order of the measured samples from low temperature to high deposition temperature. The thick samples in Fig. 2A) have a decreasing ARS signal intensity at large angles (≥35°) with increasing deposition temperature. At the same time the scattered light intensity at angles below 30° increases and the maximum or peak value of scattered light intensity shifts to smaller angles with increasing temperature. The thin samples in Fig. 2B) show a similar behavior. With increasing deposition temperature the scattered light intensities at angles below 30° increases and the peak value shift towards smaller angles. The scattered intensity at large angles, however, shows a different trend. Beginning with a low temperature of 430 °C the scattered light intensity at large angles at first increases with temperature, reaching a maximum for the sample deposited at 450 °C. Increasing the temperature further leads to a decrease in scattered light intensity at large angles alike the thick series.

The SEM finding may be explained using the modified Thornton Model by Kluth et al. [15] or the further advanced study by Berginski et al. [16]. Both studies state that the deposition temperature has a significant impact on the growth of sputter deposited ZnO:Al. With increasing temperature the films become denser. The difference in density becomes visible after etching when the surfaces reveal different textures. These textures are grouped into three types: type A or I (low temperature, low density) reveals very small features and reveals almost no roughness after etching, type B or II (medium temperature, medium density) shows a homogeneous distribution of large feature on the surface, type C or III (high temperature, high density) shows even larger features than B but with heterogeneous distribution. The SEM images in Fig. 1 show for both temperature series – thin and thick – the evolution from type A surface at 430 °C to a type B surface at 450 °C and further to a type C surface at 500 °C.

The angular resolved scattering results can be interpreted using several optical models [9,10,17]. This paper will focus on the following two models for interpretation. The first model is based on the ray-tracing theory [9]. Hence, every single ray that strikes the textured ZnO:Al surface will undergo scalar scattering in accordance with Snell's law. The measured angular resolved scattering intensities are therefore the superposition of all single scattering events in the illuminated surface area. Large or “flat” surface angles will be visible in the small angle range of the scattered intensity whereas small or “steep” angles appear in the large angle intensities. The second model treats the ZnO:Al surface as a superposition of diffraction gratings with different lattice constants [10]. In this model large grating constants or “large surface features” will appear in the small angle range of scattered intensities