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Influence of deposition conditions and substrate morphology on the electrical properties of sputtered ZnO:Al grown on texture-etched glass



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ABSTRACT

The focus of this work is the growth of aluminum-doped zinc oxide (ZnO:Al) on texture-etched glass substrates. We investigated the influence of sputter parameters, pressure and temperature on the charge carrier mobility of ZnO:Al films grown on different substrate textures. An optimized sputtering process was developed which led to charge carrier mobilities on textured substrates that are close to those on flat substrates. Based on X-ray diffraction measurements, we qualitatively explain the effect of different sputtering conditions. Furthermore, the ZnO: Al charge carrier mobility was related to the substrate morphology. ZnO:Al films on U-shaped surface morphologies showed significantly higher charge carrier mobilities than those on V-shaped structures. ZnO:Al damp heat stability and etching behavior provided evidence that the number of ZnO:Al growth disturbances on textured substrates can be reduced by adequate substrate morphology and sputtering conditions.

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1. Introduction

Textured interfaces improve the light trapping in silicon thin film solar cells [1–6]. Thereby the short circuit current density increases and thus higher solar cell efficiencies are achieved. Commonly, the transparent conductive oxide (TCO) is textured [2–4]. Sputter deposition of aluminum-doped zinc oxide (ZnO:Al) layers onto flat substrates and subsequent etching in HCl, HF or via electrochemical methods leads to textured ZnO:Al surfaces [7–9]. Doped zinc oxide grown via low pressure chemical vapor deposition (LPCVD) can also produce self-textured surface morphologies which induce light trapping in solar cells [3,5].

The texture and thus the quality of light trapping, however, depend on the specific growth conditions, such as deposition pressure, temperature [10] or layer thickness [5,6]. Hence, the deposition parameters have to be carefully adjusted. Additionally, there is a tradeoff between optical, electrical and texture properties, e.g. a thicker layer may enhance the light trapping capability of the textured TCO, but it increases at the same time the parasitic absorption in the TCO layer [5,11].

Here, we present texture-etched glass substrates to overcome the abovementioned obstacles of textured zinc oxide. The etched glass provides the texture. The subsequently sputter-deposited ZnO:Al layer must be optimized regarding electrical and optical properties only. Hence, textured glass substrates allow the decoupling of texture on

the one hand and electrical as well as optical properties on the other hand. Specifically, the layer thickness can be adjusted with regard to the layer resistance, thereby reducing the parasitic absorption in the ZnO:Al layer. However, optical and electrical properties are not decoupled completely, because the light-incoupling on textured substrates remains a function of the TCO layer thickness [12].

Although various etching methods [8] or plasma treatments [13] of textured zinc oxide can modify the resulting surface morphology to a certain extent, textured glass substrates offer a larger variety of surface morphologies.

Besides wet-chemically etched glass presented in this paper, further methods to obtain rough glass have been presented in literature. Nanoimprint lithography is a sophisticated method to produce textured substrates [14–16]. Furthermore, reactive ion etching leads to suitable surface morphologies [17,18]. Additionally, rough glass is obtained by ion beam treatment of a sputter-etched ZnO:Al mask, thereby transferring the ZnO:Al texture into the glass [19]. Textured glass was also obtained by aluminum induced texturization [20].

Despite the high number of differently textured glass substrates, studies about the ZnO:Al growth on these substrates are very limited. The growth of sputtered ZnO:Al on rough substrates with regard to its damp heat stability has been investigated for the application in chalcopyrite-based solar modules [21–23]. The challenge of ZnO:Al growth on textured substrates is the decrease of charge carrier mobility in comparison to flat substrates. Moreover, damp heat treatment of ZnO:Al on rough substrates leads to a strong resistivity increase.

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ZnO:Al growth disturbances, also called extended grain boundaries, were given as a reason for this behavior.

Nevertheless, a comprehensive investigation regarding the influence of different sputtering conditions and substrate morphologies on the charge carrier mobility is not available.

In the following, we characterize four differently etched glass substrates. We present an extensive growth study and reveal optimized growth conditions for ZnO:Al on textured glass leading to high charge carrier mobilities. On the basis of X-ray diffraction pole figures, we explain in a simple, qualitative model the impact of different deposition conditions on the ZnO:Al growth on textured substrates. Furthermore, we show that the extent of mobility decrease depends on the specific glass surface morphology. Damp heat and etching experiments suggest different amounts of ZnO:Al growth disturbances to occur on the various texture-etched glass substrates.

2. Experimental details

Sputtering with a circular magnetron was used to deposit aluminum-doped zinc oxide films in a high-vacuum system (Lesker Inc., USA) in radio-frequency mode from a ceramic target with 0.5 wt.% Al $_2$ O $_3$ at a base pressure of ~2 × 10 $^{-5}$ Pa. The target diameter was 15.2 cm, the target-to-substrate distance was 7.8 cm and pure argon was used as sputtering gas.

All ZnO:Al thin films were deposited on low-iron solar glass (EuroWhite, Euroglas, Germany). After initial basic etching, the morphology of the glass surface was modified in a second etching step. The substrates were treated in an acid mixture consisting of 45 wt.% of sulfuric acid and 0.5 wt.% of hydrofluoric acid. For glass substrates I to IV, the etching time of the second step was increased from 0 s to 120 s in 40 s steps (Fig. 1). Subsequently, the glass substrates were coated with a 140 nm SiO_xN_y barrier layer. For details about this process, see Ref. [24]. This layer slightly smoothed the glass texture, but preserved

the general morphology. The substrate size was $3 \times 10~\text{cm}^2$. We always coated two substrates within one deposition: a textured glass and a flat reference. The resulting $6 \times 10~\text{cm}^2$ glass area was positioned over the center of the target. Therefore, the outer parts of the substrates were above the race-track which had a diameter of roughly 10 cm. Measurements characterizing the layers were performed in the center of each substrate.

The ZnO:Al layer thickness was measured on the flat reference substrates to be between 600 nm and 700 nm using a surface profiler (Dektak 3030, Veeco, USA). We assumed the layer thickness on the textured substrates to be the same as that on the flat reference substrate. Hall measurements were conducted using the van der Pauw method (RH2030, PhysTech, Germany). The surface morphology was investigated by atomic force microscopy (AFM) in a Nanostation 300 (SIS, Germany) using non-contact mode and by scanning electron microscopy (SEM) in a LEO 1550 VP GEMINI (Zeiss, Germany). Damp heat degradation was carried out in a climatic chamber (Nema NCC4020) at 85 °C and 85% humidity. X-ray diffraction pole figures of the ZnO:Al (002)-reflex were recorded by a Philips X'Pert Pro MRD with a Eulerian cradle using CuK $_{\alpha}$ radiation in order to investigate the texture.

3. Results

3.1. Substrate textures

AFM images of four investigated etched glass substrates are shown in Fig. 1. In a first etching step, we obtained substrate morphology I that is characterized by pyramids with sharp valleys and high plateaus (Fig. 1a). In a second etching step, the sharp features are predominantly attacked by the acid, therefore widening and rounding the valleys. Applying this second etching step for different duration, we modified the etched glass type I towards round, smooth, crater-like surface

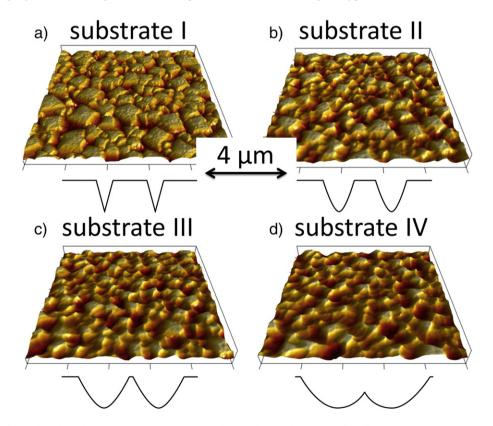


Fig. 1. AFM measurements of the etched glass substrates I–IV. The glass substrates underwent the second etching step for different times: (a) 0 s, (b) 40 s, (c) 80 s, and (d) 120 s. The corresponding rms values are: (a) 99 nm, (b) 114 nm, (c) 98 nm, and (d) 107 nm. Schematic diagrams show the valley shaping effect of the second etching step.

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