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On the effect of the underlying ZnO:Al layer on the crystallization kinetics of hydrogenated amorphous silicon

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In this contribution, we analyze the thickness effect of the underlying aluminum doped-zinc oxide (ZnO:Al) layers on the structural properties and crystallization kinetics of hydrogenated amorphous silicon (a-Si:H) thin films. It is shown that the disorder in as-deposited a-Si:H films, as probed by Raman spectroscopy, decreased with increasing ZnO:Al roughness. This caused an earlier nucleation upon crystallization when compared to a-Si:H layers directly grown on SiNx-coated glass. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4809517]

Thin-film polycrystalline silicon (poly-Si) solar cells are an appealing alternative to wafer-based technology as they combine the low cost production typical of thin-film technologies with the superior opto-electronic properties of crystalline silicon. Poly-Si thin films for photovoltaic applications have already been achieved in the past two decades by solid phase crystallization (SPC) of hydrogenated amorphous silicon (a-Si:H).1–5 CSG Solar has developed a solar cell technology where poly-Si is fabricated on glass by SPC of a-Si:H with a conversion efficiency up to 10.4%.2 Poly-Si thin films on foreign supporting materials are particularly appealing. One of the most promising substrates for low cost photovoltaics is glass, as it is inexpensive, transparent (enabling the superstrate configuration and/or bifacial modules), long-term stable, and readily available on large scale. Light trapping can be enhanced by the implementation of a transparent conductive oxide layer such as ZnO:Al. An important aspect of poly-Si solar cell technology is the deposition of a-Si:H on the ZnO:Al followed by a thermal annealing step to induce the transition from amorphous to crystalline phase. The kinetics of the crystallization process (incubation-nucleation-grain development)6 in a-Si:H controls the grain size evolution and, thus, significantly influences the material quality. Sontheimer et al.7,8 have reported on the crystallization of amorphous silicon (a-Si) deposited on ZnO:Al and SiNx-coated glass. They have demonstrated that the crystallization of a-Si/ZnO:Al is faster than in a-Si/SiNx and the former contained smaller crystals than the latter. This difference in crystallization kinetics was attributed to the roughness of ZnO:Al, although no further studies have been specifically reported on the influence of the ZnO:Al roughness on the a-Si:H microstructure, in terms of SiHx bond distribution and short-to-medium range order.

Therefore, the present contribution aims to address the influence of the ZnO:Al texturing on the microstructure of a-Si:H by following its crystallization kinetics upon SPC treatment.

Polycrystalline ZnO:Al films were deposited using an expanding thermal plasma chemical vapor deposition (ETP-CVD) technique.9–12 An argon (Ar) plasma was generated in a high pressure (360 mbar) cascaded arc and expanded into a low pressure deposition chamber (0.3–1.5 mbar). The precursors for film deposition, i.e., diethylzinc (DEZ), oxygen, and trimethylaluminium (TMA), were injected downstream via injection rings. More details about this deposition tool have been reported elsewhere.9–12 Thin (130 nm) and thick (800 nm) ZnO:Al films were deposited on glass at 200 °C. As roughness scales with thickness for ETP deposited ZnO:Al films,9,11,12 thin and thick films were used to obtain two extreme values of surface roughness. NT-MDT Solver P47 was used to perform atomic force microscopy (AFM) measurements on ZnO:Al films. 200 nm and 1000 nm thick ETP-CVD a-Si:H films were deposited on ZnO:Al layers at 400 °C. The Ar-H2 thermal plasma was generated in a cascaded arc at typical pressures of 0.2–0.6 bar. Silane, which was injected downstream, was dissociated by atomic hydrogen mainly into SiH3 radicals for relatively high H2 flow rates through the arc (≥5 sccs).9–12 200 nm thick a-Si:H films were analyzed by Raman spectroscopy using a Renishaw Raman system. Thin (200 nm) a-Si:H films ensure that the Raman laser wavelength (514.5 nm) penetrates through the whole layer thickness. a-Si:H layers deposited on ZnO:Al and SiNx coated glass substrates were annealed up to 600 °C: the ramp started at room temperature and developed up to 400 °C at 10 °C/min and from 400 °C to 600 °C at 1 °C/min. The incubation time (time before the onset of crystallinity) was defined as the time needed to observe the c-Si (111) XRD peak rise to 1/10 of its final intensity.15–17 To study the crystallization kinetics, an Anton Paar DHA 1100 heating unit attached to a PANalytical XRD instrument was used to anneal the samples in an argon atmosphere. 1000 nm thick a-Si:H films were used for in situ XRD measurements as thick films led to high intensity signal (low signal-to-noise ratio). It is worth mentioning here that no microstructural differences were observed between thin (200 nm) and thick (1000 nm) a-Si:H films. Finally, crystallized film...
composition was measured by time of flight secondary ion mass spectrometry (TOF-SIMS).

The surface roughness of ZnO:Al films is found to increase with film thickness, as earlier reported. AFM measurements showed that thin (130 nm) and thick (800 nm) ZnO:Al films have rms roughness (δrms) values of 8.5 and 41 nm, respectively. In order to understand the effect of the underlying ZnO:Al layer, the structural analysis of a-Si:H layers was performed by Raman spectroscopy where the underlying ZnO:Al layer, the structural analysis of a-Si:H between the surface roughness of ZnO:Al films and the structural properties (specifically, short- and medium range order) of a-Si:H are also affected by the surface roughness of underlying ZnO:Al. However, the relationship between the surface roughness of ZnO:Al films and the enhanced surface diffusion of growth radicals during a-Si:H deposition requires further investigation.

Furthermore, to investigate the influence of the underlying ZnO:Al film on the crystallization kinetics of a-Si:H, annealing was performed at 600°C for 10 h. A resistivity increase in a-Si:H films, as a function of the ZnO:Al thickness.

FIG. 2. Variation of (a) peak position, ω (cm⁻¹) and FWHM (b) ITA/ITO and ILO/ITO in a-Si:H films, as a function of the ZnO:Al thickness.

FIG. 1. Variation of A_HSM and R* as a function of the ZnO:Al thickness. The inset shows the stretching mode of a-Si:H deconvoluted in the LSM and HSM contributions by two Gaussians. The glass/SiNx/a-Si:H data point is also reported in the graph. Top x-axis shows the rms roughness (δrms) values.
level) were found in the glass/ZnO:Al/poly-Si layers, as indicated by TOF-SIMS measurements. Therefore, we can conclude that the observed difference in crystallization between glass/ZnO:Al/a-Si:H and glass/SiNₓ/a-Si:H is due to the improvement in structural order caused by the underlying ZnO:Al layer.

In conclusion, the analysis of the influence of the underlying ZnO:Al layer on the microstructure (R*, short- and medium-range order) of the as-grown a-Si:H and on its crystallization kinetics has been presented. The results have shown that the decrease in nano-size void density with increasing thickness of the underlying ZnO:Al led to the improvement in the structural order of the as-grown a-Si:H, and resulted in a faster nucleation in the a-Si:H layer during annealing. The understanding of the crystallization kinetics of a-Si:H on ZnO:Al is expected to play an essential role not only in poly-Si solar cell technology but also in other technologies such as a-Si:H and tandem a-Si:H/µc-Si:H solar cells.

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