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Influence of differently formed interfacial aluminium oxide on the structural properties of poly-Si films prepared by aluminium induced crystallisation

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The influence of differently formed interfacial aluminium oxide layers on the structural properties of poly-Si thin films prepared by Aluminium Induced Crystallization (AIC) of a-Si:H is reported. The Al layers were deposited by evaporation or by magnetron sputtering on an unheated substrate or at a 300ºC substrate temperature (T_S). Different methods for preparation of the aluminium oxide were applied: exposure of the Al film to air or sputtering of an Al_2O_3 target. The Al oxide layer topographies were studied by AFM. The a-Si:H films were deposited by magnetron sputtering at T_s = 250 ºC. After annealing in H_2, the poly-Si films were characterized by microprobe Raman spectroscopy and optical microscopy. The results indicate that poly-Si films prepared using evaporated Al precursors have larger crystallites than those prepared by sputtering. In the case of a sputtered Al precursor, T_s = 300 ºC is found to be more favorable for larger grain formation. The preparation of poly-Si films by AIC of glass/Al/Al_2O_3/a-Si:H stacks with a sputtered interfacial oxide is also demonstrated.

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1. Introduction

The method of Aluminium Induced Crystallization (AIC) has been widely investigated for the preparation of poly-Si thin films on glass substrates by the annealing of glass/Al/a-Si stacks [1-3]. One of the factors influencing this process is the interfacial aluminium oxide layer, which has an effect on the nucleation rate of the Si crystallites and on the Si or Al diffusion through the oxide membrane [1]. It was observed that a thicker oxide leads to longer annealing times [1,2]. In the present paper, a study of the influence of the interfacial oxide layer, formed by different methods, on the structural properties of poly-Si films prepared by AIC is presented. The aluminium oxide was formed either by exposure of the Al film to air for 24h or by sputtering from an Al_2O_3 target on top of the Al layer. The Al layers were deposited by sputtering at two substrate temperatures: room temperature (RT) and 300 °C, or by evaporation at RT.

The structure of the poly-Si films was investigated by microprobe Raman spectroscopy and optical microscopy. The surface topography of the Al films was studied by Atomic Force Microscopy (AFM).

2. Experimental details

Hydrogenated amorphous Si films (a-Si:H) were deposited on Al coated glass substrates by RF magnetron sputtering of a c-Si target at a substrate temperature of 250 ºC in a gas mixture of Ar (0.5 Pa) and H_2 (0.05 Pa). These conditions resulted in hydrogen concentrations (measured by Elastic Recoil Detection Analyses) in the a-Si:H of about 6 at.%. The base pressure in the chamber was 10^{-4} Pa.

In order to investigate the effect of the Al deposited at RT by different methods, two sets of samples with thermally evaporated (sample EVD) and RF magnetron sputtered Al precursor layers (sample SPA) were prepared. The Al layers were exposed to air for 24 h prior to the deposition of the a-Si:H films.

In order to investigate the influence of the interfacial oxide layer for the sputtered Al precursor, three sets of samples were prepared: where the Al layers were deposited at RT (sample SPA) or at 300ºC (sample SPB) and then exposed for 24h to air, and the third one, (sample SPC) where Al_2O_3 (about 4 nm) was sputtered upon Al layers deposited at 300 ºC.

All samples were annealed in hydrogen at 500 ºC for 7h. The Al layer remaining on top of the poly-Si, formed due to the characteristic aluminium induced layer exchange process of AIC, was removed using a standard Al etch solution. The technological parameters for all sets of samples are given in Table 1.

The crystallinity of the poly-Si films was investigated by microprobe Raman spectroscopy, using a Leica DMLP microscope coupled to a Kaiser Optical Systems Raman system. The laser spot diameter on the sample was 8 μm and the total laser power incident on the specimen was kept below 1.4 mW. The values of the Si-Si TO-like peak position (ω_{TO}) and its Full Width at Half Maximum (FWHM) were acquired by fitting to a Gaussian profile.

Surface roughness characterization of the Al layers was performed by obtaining topographical images in a non-contact mode of small areas of each sample, employing a Quesant Instrument Corporation AFM Q-Scope 250. The optical microscopy images were acquired using a Neophot 32 optical microscope (Jenoptic, Jena).

3. Results and discussion

The AFM topographical images of the different sputtered and evaporated Al precursors used for the preparation of the poly-Si films are shown in Fig. 1. The surface average heights of the sputtered precursor Al films used in samples SPA (Fig. 1a), SPB (Fig. 1b) and SPC (Fig. 1c) are: 21.3, 120.6 and 23.6 nm, respectively. The respective root mean square (RMS) values of these heights are 5.2, 23.5, and 9.6 nm. Al films sputtered at 300ºC and exposed to air for 24 h (Fig. 1b) exhibit larger grains than the other Al films, as seen in Fig. 1. Due to the observation that a small-grained polycrystalline Al precursor results in a faster nucleation rate and smaller grains of the poly-Si film [1], it can be suggested that Al sputtered at 300 ºC should be more suitable as a precursor for the preparation of poly-Si films with larger grains by the AIC method. The surface average height and RMS values obtained from AFM of the evaporated Al (Fig. 1d) are 30.8 and 14.2 nm, respectively. The AFM images of both Al precursor layers, sputtered and evaporated at RT (Fig. 1a and Fig. 1d, respectively), reveal that the evaporated Al has a greater roughness and larger grains.

Fig. 2 shows optical images of the poly-Si films prepared using a sputtered Al precursor. The films deposited at RT and with native oxide formation (exposed to air for 24 h) – (Fig.1a), or from Al capped by sputtered aluminium oxide (Fig. 1c), exhibit dendritic grains. In contrast, poly-Si prepared using Al deposited at 300ºC and with native oxide (Fig. 1 b) have more dense structures and smoother surfaces. In this case, the grains are closely packed and the inter-grain material is hardly distinguished. These results can be correlated with the larger grains of the respective Al films observed by AFM.

The crystalline structure of the poly-Si films was verified by microprobe Raman spectroscopy. The FWHM and the ω_{TO} of the Si-Si TO-like peak taken from the microprobe Raman spectra of poly-Si prepared by AIC of all investigated samples with different interfacial aluminium oxide layers are presented in Table I. The TO-like peak of a c-Si wafer, used as a reference, is observed at 520.7

Table 1. Peak position (ω_{TO}) and FWHM of the Si-Si TO-like band of AIC poly-Si films (annealed in H_2) prepared using different sputtered (SPA, SPB, SPC) and evaporated (EVD) precursors. The microprobe Raman spectra were measured at grain or inter-grain positions.

Samples	Precursor structure with Al and Al_2O_3	T_s^{Al} ºC	Al exposure to air, h.	ω_{TO}, cm^{-1}		FWHM, cm^{-1}	
				grain	inter-grain	grain	inter-grain
SPA	Glass/Al/a-Si:H	RT	24	519.8±0.5	517.2±0.5	9.0±0.5	9.5±0.5
SPB	Glass/Al/a-Si:H	300	24	521.1±0.5	521.4±0.5	7.2±0.5	7.4±0.5
SPC	Glass/Al/sputtered Al_2O_3/a-Si:H	300	-	521.2±0.5	521.0±0.5	8.1±0.5	8.2±0.5
EVD	Glass/Al/a-Si:H	RT	24	522.9±0.5	520.5±0.5	8±0.5	8.4±0.5
c-Si reference		-	-	520.7±0.5		5.9±0.5	

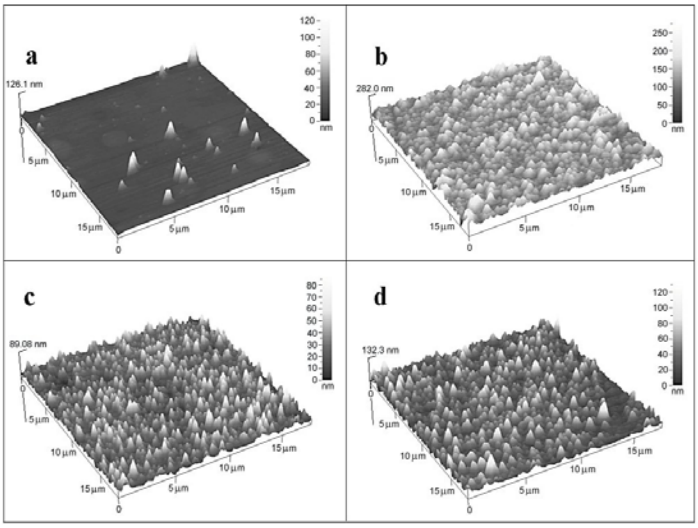

Fig.1. AFM images of different sputtered (a, b, c) and evaporated (d) Al precursors: (a) room temperature deposition and exposure for 24h to air; (b) 300 ºC deposition and 24 h exposure to air; (c) 300 ºC deposition and capping by sputtered Al_2O_3, (d) room temperature deposition and 24 h exposure to air.

cm^{-1}, and its FWHM is 5.9 cm^{-1}. It can be seen that the poly-Si films prepared from Al sputtered at 300ºC and stored in air for 24 h (sample SPB) or capped by sputtered Al_2O_3 (sample SPC) exhibit ω_{TO} values very close to the c-Si reference value, for both conditions under which the Raman signal has been obtained: from the grain and intergrain material. The results indicate a good homogeneous crystalline structure of these poly-Si films. The spectra of the poly-Si films prepared from Al deposited at RT and exposed for 24h to air (sample SPA), measured on the grains and on the inter-grain regions have ω_{TO} values shifted to lower wave numbers - 517.2 and 519.8 cm^{-1}, respectively.

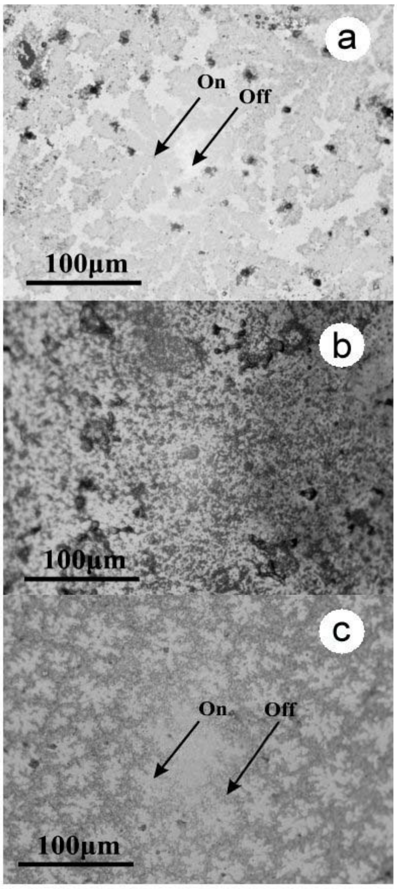

Fig. 2. Optical images of AIC poly-Si films prepared using different sputtered Al precursors: (a) RT deposition and 24h exposure to air; (b) 300ºC deposition and 24h exposure to air; (c) 300ºC deposition and capping by sputtered Al_2O_3. Arrows (on) point to grains and arrows (off) point to inter-grain material.

An estimate of the grain size and the stress in the films can be made from the downshift and FWHM of the Si-Si TO-like Raman peak [4-6]. The grain size is inversely proportional to the FWHM of the Si-Si TO-like peak [5]. The values of the FWHM of the SPB sample are the smallest. It can be deduced that deposition of Al at 300 ^{o}C results in poly-Si with larger grains compared to the case of Al deposited at RT (sample SPA), as also observed from AFM and optical images. Theslightly higher FWHM of the Si-Si TO-like peak the sample prepared from a structure consisting of an approximately 4 nm thick sputtered interfacial Al_2O_3 layer (sample SPC) indicates the formation of slightly smaller grains compared to those obtained from an Al precursor kept for 24h in air (SPB), as confirmed by optical microscopy observations. It is known that the diffusion of the Si through the aluminium oxide membrane during the process of AIC strongly depends on the oxide thickness – a thicker Al_2O_3 layer suppresses the crystallization velocity in the poly-Si films [2]. The thickness of the oxide in sample SPB could be estimated to be about 2 nm, about half that of sample SPC. The smaller grain size in the samples with sputtered Al_2O_3 could thus be a consequence of this. Thus, the thickness of the sputtered Al_2O_3 layer should be further optimized in order to obtain larger grained poly-Si films with homogeneous surfaces.

Microprobe Raman spectra of poly-Si prepared using an evaporated Al precursor exhibit closer values to the c-Si reference peak position. Thissuggests the presence of larger crystalline grains in poly-Si in the case of using of evaporated Al precursor compared to the case of sputtered Al, due to the larger grains in the precursor layer, as suggested from AFM.

The shift of the Si-Si TO-like peak towards higher or lower wave numbers can be related to the amount of compressive or tensile stress in the polycrystalline films, respectively [3,6]. It can be suggested that poly-Si films prepared by AIC using a sputtered Al precursor deposited at room temperature consist of grains with tensile stress, while those prepared from Al deposited at 300 ºC exhibit a compressive one. Although this result needs further investigation, it is possible to suppose that the presence of Al and more disordered Si in the inter-grain region could be a reason for the presence of the tensile stress [3, 6]. It can be noted that there is a correlation between the presence of the compressive stress and the larger grain size of the precursor Al films.

4. Conclusions

We have shown that the structural quality of poly-Si films prepared by the AIC of glass/Al/ a-Si:H stacks depends on the method and the temperature of deposition of the Al precursor film, and on the thickness and the formation method of the interfacial oxide. Microprobe Raman spectra of poly-Si films prepared by AIC using different Al precursors show that the grain size of the Al film is an important parameter in respect of the structure of the poly-Si. Poly-Si films prepared using an evaporated Al precursor exhibit larger crystallites than those prepared using sputtered Al (for the same annealing conditions). In the case of sputtered Al, a deposition temperature of 300 ºC is found to be more favourable for large grain formation. The preparation of poly-Si films with good structural quality by AIC of glass/Al/Al_2O_3/ a-Si:H stacks with sputtered interfacial oxide has been demonstrated.

Acknowledgments

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