Thin Solid Films 616 (2016) 213–219

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Effects of the Si/Al layer thickness on the continuity, crystalline orientation and the growth kinetics of the poly-Si thin films formed by aluminum-induced crystallization

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ARTICLE INFO

Article history:
Received 22 March 2016
Received in revised form 5 August 2016
Accepted 5 August 2016
Available online 12 August 2016

Keywords:
Aluminum-induced crystallization
Polycrystalline silicon
Thin films
Surface coverage
Grain orientation
Nucleation

ABSTRACT

The joint impact of the Si/Al layer thickness on the growth kinetics, the crystalline orientation and the size of the poly-Si grains resulting from aluminum-induced crystallization process is analyzed. It is shown that the surface coverage of resulting poly-Si layers rapidly decreases together with annealing temperature and the Si/Al ratio. The surplus of a-Si over the Al needed to ensure continuity of the poly-Si thin film is in the range of 35%–50% for Al layers thicker than 225 nm, but rapidly goes up to 200% as the thickness of the Al layer decreases below 50 nm. It is demonstrated that the angular distribution of grain orientations is discrete and shifts towards the {111} direction as the Si/Al increases. It is reported that during an isothermal annealing, the nucleation of Si grains occurs in two steps. Finally, a simple model of the aluminum-induced crystallization process explaining the two-step nucleation is proposed.

1. Introduction

The aluminum-induced layer exchange (ALILE) process based on the aluminum-induced crystallization (AIC) has gained significant attention over the last two decades for its ability to form continuous polycrystalline Si thin films with relatively big grain size (films with the grains larger than 50 μm were reported) and controlled crystalline orientation [1–7] on the dielectric substrates. The conventional approach to perform the AIC consists in depositing a micro-crystalline Al layer on a silica-based dielectric substrate followed by an amorphous Si layer with a buffer oxide layer in-between and consecutive annealing at a temperature below the eutectic temperature of Si/Al alloy of 577 °C. Another popular approach called "inverted Aluminum-Induced Layer Exchange (iALILE)" consists in depositing the a-Si layer first. After iALILE, the poly-Si layer appears on top of the Al layer on glass, which can be advantageous for photovoltaic applications, since the Al layer can be used as a back electric contact [8]. Although the crystallization process is faster when a-Si is deposited prior to Al, the overall layer exchange phenomenon occurs independent of the layer sequence [9].

Back in 1998, Nast et al. reported [10] that the resulting poly-Si layer had exactly the same thickness as the initial Al layer. It was concluded that the initial a-Si and Al layer should be roughly of the same thickness. However, a fraction of the Si material is consumed during the AIC. Therefore, in order to create a continuous poly-Si thin film, the initial a-Si layer should be slightly thicker than the Al layer [11]. The exact amount of the Si surplus is open to question. The values of the Si and of the Al layer thickness as reported in the key articles on the Si ALILE process are traced in Fig. 1. Fox a fixed Al layer thickness, the value of the Si layer thickness significantly differs from article to article. In addition, the dispersion of data points widens as the Al layer thickness is decreased. The latter fact is particularly important in the frame of the recent focus on the crystalline orientation control by modulating the thickness of the Al layer below 200 nm [3,4]. However, the role of the Si layer in orientation control has not been discussed so far.

In this paper, the impact of the Si/Al layer thickness ratio on the Aluminum-Induced Crystallization process is analyzed. It will be shown that a small change of the Si layer thickness alone leads to significant changes in the kinetics of the AIC process, grain size, orientation and the total surface coverage.

2. Experimental

The poly-Al and the a-Si thin films were deposited onto quartz (qtz) substrates by DC magnetron sputtering at the base chamber pressure of 3×10^{-5}Pa. The poly-Al layer was deposited at 100 W and the Ar pressure of 1.0 Pa, whereas the a-Si layer was deposited at 500 W and the

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http://dx.doi.org/10.1016/j.tsf.2016.08.016
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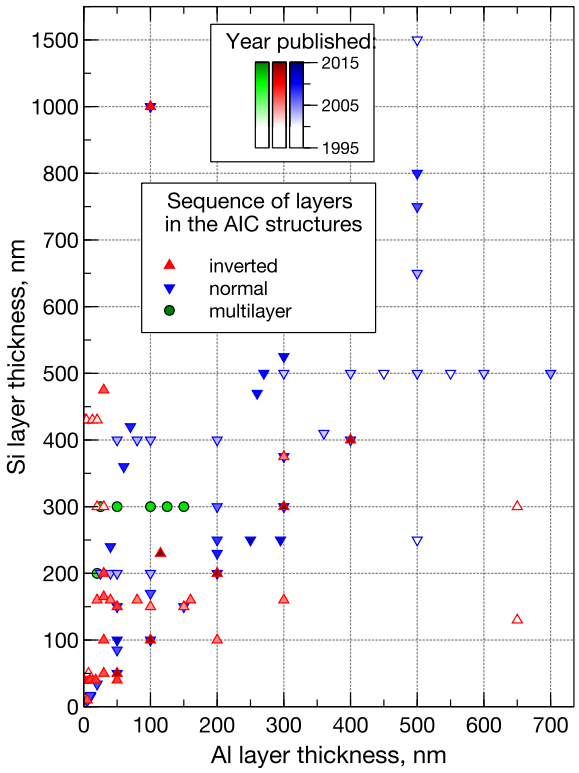

Fig. 1. Thickness of the Al and of the Si layers used in the a-Si/poly-Al/qtz structures reported in the literature. The blue points stand for the normal bilayer a-Si/poly-Al/qtz structures (SiO2/poly-Al/AlO_x/a-Si), the red points stand for the inverted bilayer poly-Al/a-Si/qtz structures (SiO2/a-Si/SiO_x/poly-Al). The thickness of the unit Si/Al layers in multilayer structures are given in green. The publication year of the papers is coded with the brightness of the data points. If the same a-Si/poly-Al/qtz or poly-Al/a-Si/qtz structure has been reported in multiple articles, the brightness indicate the age of the most recent article. The high-resolution version of this figure with referring papers indicated in labels to the corresponding data points can be found in supplementary materials to this article. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Ar pressure of 0.2 Pa, which corresponds to the deposition rates of 0.26 Å/s and 2.8 Å/s, respectively. The thickness of the Al layer was varied from 22 nm to 275 nm and the ratio of Si/Al layer thickness was within the range of 0.8 to 2.75. Prior to depositions, the substrates were cleaned in the piranha solution (H_2SO_4:H_2O_2 = 3:1 for 10 min) followed by the two-step RCA cleaning (HNO_3:H_2O_2:H_2O = 4:1:1 and HCl:H_2O_2:H_2O = 4:1:1 at 70 °C for 10 min each), sonification in deionized water for 10 min and finally dried under a N_2 stream. The buffer AlO_x layer was formed by exposing the poly-Al layer to atmosphere for 5 min. The particular samples were exposed to atmosphere for 24 h. The samples were annealed at the temperatures within the range of 400 °C to 550 °C in the N_2 atmosphere. Annealing was performed in a window-equipped furnace to enable in-situ observations of the AIC process with the Keyence VHX-2000 optical microscope. After annealing, the Al, AlO_x and the residual a-Si layers were etched off by immersing the sample to the 5% HF solution for 5 min and then to H_2O:HF:$K_2Cr_2O_7$ = 320 ml:40 ml:0.8 g for 2.5 min in order to bare the surface of the poly-Si layer for the further analysis. The Scanning Electron Microscope (SEM) pictures were taken at accelerating voltage of 15 kV using a JEOL JSM70001FA microscope equipped with an EDAX detector for recording Electron Back-Scatter Diffraction (EBSD) maps. The microscope photos were taken at fixed settings of the brightness, contrast and of the white balance and then processed with Adobe Photoshop CC 2015 software. The percentile of surface coverage was calculated as a fraction of non-white pixels on a photo after subtracting the background in lab colour mode by normalizing the histogram using the same shadow and highlights levels for all the photos analyzed. The nucleation density was calculated by hand-counting the crystallites on the fixed surface area of the sample of 333 × 250 μm.

3. Results and discussions

3.1. Nucleation and grain growth

The impact of the Si/Al ratio on the flow of crystallization process was investigated by analyzing the in-situ photos of the quartz/Al interface taken during annealing. Once nucleated, the Si crystallites appear on photos as dark areas due to a lower reflectivity of Si as compared to Al (Fig. 2 (a)). The two parameters which can be directly measured on a such photo are the surface coverage (i.e. a percentile of the surface covered with recrystallized Si grains later referred as "X") and the nucleation density (i. e. a number of Si grains per unit surface of the sample). An example of a such graph is shown in Fig. 2(b). The surface coverage-time is a smooth S-shaped graph typical for phase-transformation processes. In contrast, the nucleation density-time curve has an inflection point. That means, the nucleation occurs in two steps. Such an inflection point is not seen on the nucleation density-time curves previously

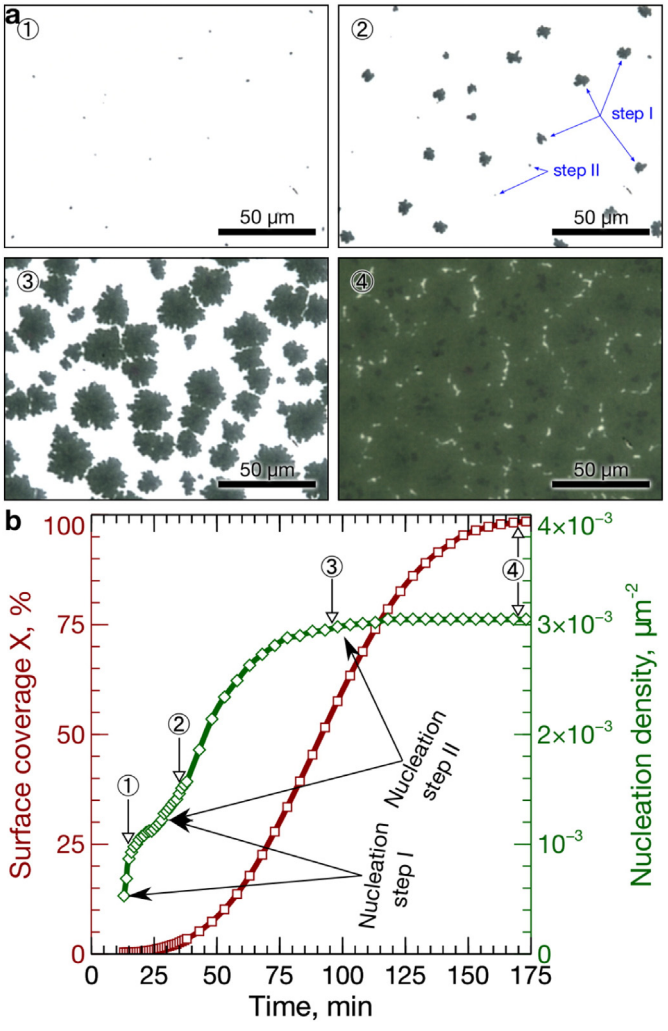

Fig. 2. (a): In-situ microscope photos of the interface between quartz substrate and the a-Si/poly-Al/qtz structures (Al = 225 nm, Si/Al = 1.3, T = 437 °C) taken at the beginning of nucleation step I (1), at the beginning of nucleation step II (2), when max. nucleation density reached (3) and when max. surface coverage reached (4). Dark areas designate Si grains, bright areas - the Al. The grains nucleated at step I and step II are indicated with blue arrows in photo (2). (b): Surface coverage and nucleation density as a function of time. The points on graphs corresponding to photos (1–4) are denoted by arrows with empty arrowheads. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)