EPITAXIAL SI GROWTH ON POLYCRYSTALLINE SI SEED LAYERS AT LOW TEMPERATURE

B. Rau[1,*], J. Schneider[1], M. Muske[1], I. Sieber[1], S. Gall[1], M. Stöger-Pollach[2], P. Schattschneider[2] and W. Fuhs[1]

[1]Hahn-Meitner-Institut Berlin, Silizium-Photovoltaik, Kekuléstr. 5, D-12489 Berlin, Germany
[*]Tel.: +49 30 8062-1329, Fax: +49 30 8062-1333, E-mail: bjoern.rau@hmi.de
[2]Institute for Solid State Physics, Vienna University of Technology, Wiedner Hauptstr. 8-10, A-1040 Vienna, Austria

ABSTRACT: The epitaxial thickening of polycrystalline Si films on glass substrates is of great interest for the realisation of crystalline Si thin film solar cells and other large-area thin film devices. In this paper we report on the epitaxial growth of Si deposited below 600 °C on a polycrystalline Si seed layer on glass. The films were grown by electron-cyclotron chemical vapor deposition (ECRCVD) on seed layers made by aluminium-induced crystallisation. The quality of the grown films strongly depends on both the seed layer preparation in general and on the orientation of the underlying grain in particular. Due to a mainly favourable orientation of the seed layers more than 73 % of a seed layer are epitaxially thickened by a 400 nm thick film. The degree of epitaxy depends on the total film thickness, due to the local breakdown of epitaxy of some grains after a certain critical thickness depending on the specific grain orientation. The results show, that the epitaxial thickening of a polycrystalline Si seed layer on glass by ECRCVD is a promising route to crystalline Si thin-film solar cells on glass. But they also illustrate the challenges on the way to a low-temperature grown epitaxial absorber layer.
Keywords: Si-Film, Epitaxy, Low-temperature

1 INTRODUCTION

An attractive low-temperature route to a poly-crystalline Si (poly-Si) thin-film solar cell on glass bases on the seed layer concept [1]. This route is followed in the framework of the European Commission project METEOR [2]. In such a cell concept, we use a thin large-grained poly-Si seed layer on glass formed by aluminium-induced crystallisation (AIC) [3,4]. The crystalline absorber layer is grown on this seed layer in a subsequent epitaxial growth process. The substrate temperature of such a growth process is limited by the softening point of the glass to temperatures of about 600 °C. At these low temperatures it becomes necessary to apply deposition techniques which provide additional non-thermal energy to the surface of the growing film. Crystal orientation and surface cleaning of the substrates are much more critical for epitaxial growth than for conventional high-temperature deposition techniques. In previous works it was demonstrated that ion-assisted techniques like electron-cyclotron resonance chemical vapor deposition (ECRCVD) [5] or ion-assisted deposition (IAD) [6] are in principle suitable for low-temperature epitaxial growth of Si. First epitaxial growth of Si on AIC seed layers was achieved using IAD [7]. Here we report on the succesful epitaxial growth of Si on poly-Si seed layers on glass by low-temperature ECRCVD.

2 EXPERIMENTAL PROCEDURE

2.1 Specimen characterisation

To investigate the structural properties and the crystal orientations of seed layers and deposited films a Hitachi S4100 (25 keV) scanning electron microscope (SEM) equipped with a cold field emission cathode and the electron backscattered diffraction (EBSD) system ORKID of ThermoNORAN were used. For the EBSD measurements the samples were tilted by an angle of 70 ° to the electron beam. The crystal orientations were determined by the analysis of the Kikuchi pattern. For a detailed investigation of the sample cross section transmission electron microscopy (TEM) and electron diffraction experiments were carried out with a TENCAI F20 ST at the TU Vienna.

2.2 Seed layer preparation

To realise the epitaxial growth of Si on a foreign substrate like glass a seed layer is necessary which provides an ordered surface with a crystalline morphology. One way to create such a poly-Si seed layer is a so called aluminium-induced layer exchange (ALILE) process based on AIC [3]. Two major advantages of such seed layers are the realisation of large grains (above 10 µm) with a preferential (100) orientation. A 300 nm thick Al film was deposited by DC magnetron sputtering directly on a RCA cleaned glass substrate (Corning 1737F). After 1 hour exposure of the specimen to air a thin aluminium oxide layer was formed. On top of this a 375 nm thick amorphes Si film was deposited also by DC magnetron sputtering. In a subsequent annealing step the specimen was exposed for 6 hours to a temperature of 500 °C. This procedure leads to an exchange of the Al and Si layers combined with the concurrent transition from amorphes to crystalline Si. As a result of this process a poly-Si layer is formed directly on the glass substrate. The remaining Al layer on top contains Si inclusions. This layer and the oxide layer below were removed by wet-chemical etching and chemical-mechanical polishing (CMP) resulting in a smooth surface of the poly-Si seed layer.

At present the orientation of the grains cannot be reliably controlled. There are different mechanisms under discussion for the formation of preferential orientation. The impact of the annealing temperature on the crystal orientation has been demonstrated by Kim et. al [8]. Very recently investigations performed by our group have shown that also the properties of the thin oxide layer between the aluminum and the a-Si layer have influence on the crystal orientation.

In our case we used seed layers which grains are preferentially (100) orientated. Fig. 1 shows the pole figure gained from an EBSD orientation mapping of such

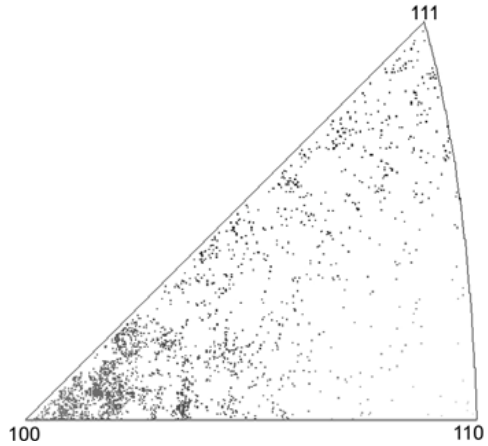

Figure 1: EBSD pole figure of a poly-Si seed layer on glass made by ALILE showing the statistics of grain orientation.

a seed layer. The mapping was about 64 × 50 µm² including a representative number and distribution of grains (average grain size: 10 µm). The pole figure shows, that there is a preferential orientation of the grains in the region of (100) indicating a grain surface orientation tilted by a specific angle with respect to (100). From the analysis of the orientation map we obtained a statistic information about the share of the seed layer area consisting of grains having a crystal orientation of (100) but are tilted by an angle which is smaller than a certain value. This is shown in Table I where the share of area is given in percent of the total seed layer area in dependence on the maximum tilt angle to (100).

Table I: Statistic analysis of the seed layer area consisting of grains which crystal orientations are tilted (100) surfaces with a maximum tilt angle less than a specific value.

max. tilt angle from (100)	10 °	15 °	20 °
share of area	41 %	64 %	77 %

Note, these statistics did not include any information about the atomic surface structure of a particular grain. This structure (i.e. the shape of the formed terraces on the grain surface) strongly depends on the direction of the tilt.

2.3 Epitaxial growth

Si films with high crystallographic quality can be grown on Si(100)-wafers by ECRCVD up to film thicknesses above 2 µm [9]. However substrate orientations different from (100) lead to epitaxial films with a high degree of disorder or complete finecrystalline films, respectively [10]. From this investigations it follows, that a preferential (100) orientation of the seed layer is favourable for its epitaxial thickening. The crystalline Si films discussed here were grown in an ECRCVD system with a RR 250 PQ (Roth & Rau, Germany) plasma source decomposing SiH_4 by an H_2 plasma. The seed layers were treated by a standard RCA cleaning process and a final HF dip (30 s, 2 % HF:H_2O solution). In the process chamber, prior to the deposition, the substrates were held in hydrogen atmosphere (mTorr-range) for 30 min to obtain stable thermal process conditions. The substrate temperature was 585 °C. A microwave power of 1000 W was used and the total pressure amounted to 6.5 mTorr. The resulting growth rate was 20 nm/min. The process gas consisted of 10 sccm SiH_4 and 10 sccm H_2. The base pressure of the system was about 4×10^{-7} Torr at the process temperature.

3 RESULTS

In Fig. 2 a SEM image of a 400 nm thick Si film is given grown on the seed layer described in Fig. 1. In front, the cross section of the sample with film, seed layer and glass substrate is shown. The upper part of the image shows the surface of the film. Different grains are well distinguishable resulting from different degrees of crystallographic order. The grain in the center and front has a smooth fraction edge and also a smooth grain surface indicating the succesful epitaxial thickening of the underling seed layer grain. The neigbouring grain on the top left is finecrystalline grown. No epitaxy took place on the corresponding grain there. As a reference, Si was deposited on a crystalline Si(100) wafer in the same experiment. The resulting film was homo-epitaxially grown without any disordered regions or inclusions.

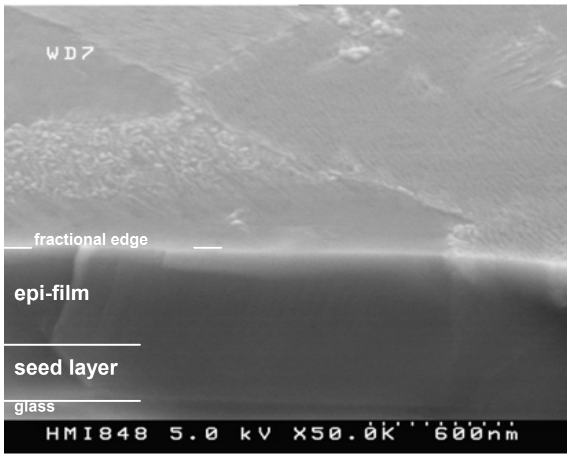

Figure 2: Fractional edge and surface of a 400 nm thick crystalline Si film grown on a poly-Si seed layer on glass (SEM image).

The reason for the different kind of Si growth on the seed layer is the difference in crystal orientation of the different grains. As mentioned above, the substrate orientation is very important for the epitaxial growth of Si at low temperatures. To analyse this effect in more detail a crystal orientation mapping of the film on the seed layer was carried out [11]. Fig. 3 presents the orientation map of the grown film shown in Fig. 2. Grey-shadowed regions display epitaxially thickened grains, where a surface orientation could be observed. Black areas mark regions, where no orientation could be determined. No Kikuchi pattern could be detected due to the finecrystalline structure of the grown film here. In

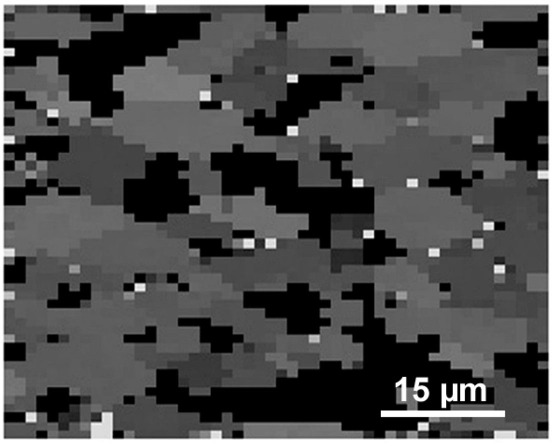

Figure 3: EBSD orientation map of the Si film shown in Fig. 2 (gray regions: epitaxial thickened grains, black regions: finecrystalline thickened grains).

these cases the grains of the seed layer have crystallographic orientations which are not suitable for the epitaxial thickening at the present process conditions.

In order to investigate the correlation between seed layer orientation and morphology of the deposited film in more detail TEM experiments were carried out and electron diffraction patterns were analysed. In Fig. 4 a TEM cross section (left) of the Si film on the seed layer is given. The image shows the glass susbtrate (bottom), the seed layer (center), and the Si film (top). The circles mark the positions of the selected area aperture for electron diffraction. Two regions of the film are well distinguishable (B and D) caused by two different oriented grains of the seed layer (A and C). The left region of the film is finecrystalline grown starting right at the interface to the seed layer. Whereas the right region is epitaxially grown. The corresponding diffraction patterns are presented on the right of Fig. 4.

The diffraction patterns A and B are identical. This indicates the successful epitaxial thickening (B) of the seed layer (A) in this region. The analysis of these patterns led to a clear (100) orientation of the grain surface. Although pattern C is nearly the same as A, no epitaxial growth took place at the left grain of the seed layer. The deposited film here has a finecrystalline structure. The reason for this is a 7.8 degree tilt of the grain surface with respect to the strictely (100) oriented neighbouring grain. By analysing the pattern D in more detail, the spots of pattern C can be found again, but also many additional spots are visible. This is due to the fact that the needle-like grains in this film region are preferentially oriented corresponding to the seed layer grain.

This pronounced difference in the growth mode for both grains with strict (100) orientation and a slightely tilted (100) orientation is remarkable. It emphasizes that the (100) orientation is not the only requirement for Si epitaxy at low temperatures. Because from the EBSD analysis of the grown film in Fig. 3 we know that also grains with a larger tilt angle than 7.8 degree could be epitaxially thickened. Therefore we assume, that also the atomic structure of the grain surfaces (i.e. the form of the terraces on the surface) seems to influence the epitaxial growth process.

To quantify the success of epitaxy on the seed layer we analysed the orientation map shown in Fig. 2 in more detail. We determined the degree of epitaxial thickening by the ratio of the non-black area in comparison to the complete area of the orientation map. As a result, about 73 % of the surface of the film is poly-Si indicating the epitaxial thickening of the appropiate grains of the seed layer. Comparing with the seed layer orientation statistics of Table I this shows, that also grains with strong tilt to (100) can be epitaxial thickened up to a film thickness of 400 nm. The same result was obtained if a SEM top view image of the sample was investigated by determining the share of smooth surface regions in comparison to the total image area.

In order to use the ECRCVD-grown films as absorber layers in thin-film solar cells the film thickness has to be increased. Therefore we grew Si films of different thicknesses on seed layers at identical process conditions. For that we used specimen of the same seed layer sample as described in Fig. 1. As can be seen in Table II, the degree of completely epitaxially thickened grains of a

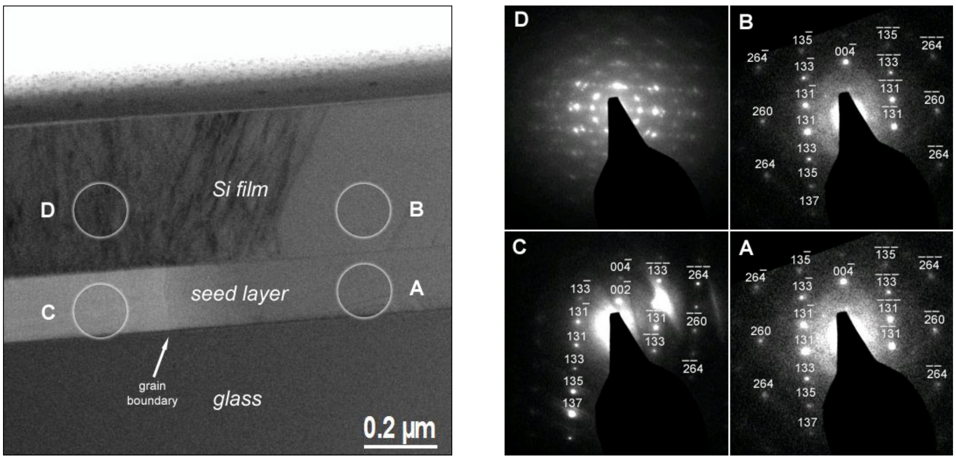

Figure 4: TEM cross section (left) and diffraction pattern (right) a the poly-Si film deposited on a seed layer on glass. The circles mark the positions of the selected area aperture for electron diffraction. The diffraction images are aligned with respect to the neighbouring TEM image.

sample decreases strongly with increasing film thickness. This occurs by a local breakdown of epitaxy of particular grains with crystal orientations which are not perfectly suitable for epitaxial growth. Such a breakdown is known as a typical effect for low temperature epitaxial growth at different growth techniques and strongly dependent on the crystal orientation [12,13]. As shown in Fig. 1 the seed layer we used had a large variation of crystal orientations inside a specific tilt angle to (100). Therefore the atomic structure of the surface varies on different grains with the same value of the tilt angle leading to different kinds of crystalline Si growth.

Table II: Estimated degree of completely epitaxially grown grains in dependence on the film thickness.

film thickness	400 nm	500 nm	1100 nm	1630 nm
degree of epitaxy	73 %	65 %	51 %	35 %

4 SUMMARY AND CONCLUSIONS

In summary, we obtained epitaxial growth of Si on poly-Si seed layers on glass at temperatures below 600 °C by ECRCVD. Approximately three quarters of a seed layer are epitaxially thickened at a film thickness of 400 nm. This degree of epitaxy confirms, that a preferential (100) orientation of a seed layer is favourable for epitaxial thickening by low-temperature ECRCVD. Clearly also additional phenomena, such as the shape of the formed terraces on the surface, may have an influence on the growth mode. This is also emphasized by the local breakdown of epitaxy on certain grains with increasing film thickness.

The results present a decisive step on the way to a poly-Si thin-film solar cell on glass. But they also show, that especially the seed layer orientation has to be improved further to realize an epitaxially grown absorber layer of a suitable thickness.

5 ACKNOWLEDGEMENTS

The authors would like to thank E. Conrad, G. Keiler, K. Jacob, and S. Kreutzmann for technical assistance and substrate preparation. The work has been financially supported by the European Commission (METEOR project, ENK5-CT-2001-00543).

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