Orientation Control of Large Grain Poly-Si on Glass by Interfacial Oxide Layer Controlled Al-Induced Crystallization

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1. Introduction

Oriented large-grain poly-Si on glass substrates is strongly required to realize high-efficiency thin-film solar cells and advanced system-in-displays. Recently, Al-induced crystallization (AIC) of Si, where a-Si/Al stacked structures having interfacial Al oxide layers change into Al/poly-Si structures by low-temperature annealing (<450°C), has been investigated by many researchers [1-3], because this process realizes oriented poly-Si with large grains ($\sim 20 \mu m$). However, there is a discrepancy in the preferential orientations among the researchers; i.e., (001) [2] or (111) [3]. The clarification of the reason and its control are very important.

We focus on effects of the interfacial oxide layer thickness on crystal orientation of poly-Si, because interfacial oxide layers acts as barriers of Si and Al, and plays an important role in AIC growth features [4]. In the present study, the control of the preferential orientation of poly-Si grains is demonstrated, and the mechanism is discussed.

2. Experimental Procedure

The sample fabrication is schematically shown in Fig.1. The Al films (100 nm thick) were deposited on quartz substrates by DC magnetron sputtering, where the XRD measurements revealed that the Al films were preferentially oriented to (111). Some samples were exposed to air for 5 min-24 h to form native Al oxide layers on the surfaces, and other samples were not exposed to air. Subsequently, a-Si films (100 nm thick) were deposited on the Al oxide layers by an MBE system (base pressure: ~10⁻¹⁰ Torr). Here a-Si was evaporated using an electron-beam gun. All depositions and oxidation were carried out at room temperature. Finally, they were annealed at 450°C for 20-40 h in a furnace under a dry nitrogen ambient. The annealing temperatures were selected to be below the eutectic temperature of the Al-Si (577°C).

After etching the top Al layers, crystal orientations of poly-Si layers were characterized by electron backscatter diffraction (EBSD) analysis.

3. Results and Discussion

Figs.2(a)-2(d) show Nomarski optical micrographs of the annealed samples with different air exposure times. The annealing times are shown in the figures. In the case of long-time air exposure (24 h), layer exchange of Si/Al layers did not complete in 20 h; therefore, additional annealing for 20 h was performed, which indicated that the interfacial Al oxide thickness became thicker with increasing air exposure time. It is found that the crystal grains become large from <1 μ m [Fig.2(a)] to ~100 μ m [Fig.2(d)] with increasing air exposure time, and large grains (~20 μ m) are obtained for all samples with air exposure. In order to evaluate the crystal orientation of the growth layers, EBSD measurements were performed. Figs.2(e)-2(l) show EBSD maps and inverse pole figures (IPF) as a function of air exposure time. These characterizations were performed along the normal direction to the sample surfaces. In the case of the sample without air exposure, crystal orientations are random. However, preferential (001) or (111) orientation is recognized for the sample exposed to air for 5 min or 24 h.

In order to quantitatively investigate the preferential orientation, the orientation distribution of the samples (air exposure: 5 min, 60 min, 24 h) are summarized in Figs.3(a), 3(b), and 3(c), respectively. From these figures, the fractions of the preferential (001) and (111) orientations are obtained as a function of the air exposure time, as shown in Fig.4, where the fractions of pixels in the EBSD maps with angles within 20° from (001) or (111) are counted. In the case of short-time air exposure (5 min), a high fraction $(\sim 80\%)$ of the growth surface is oriented to (001). On the other hand, the fraction of the (111)-oriented regions increases with increasing air exposure time, and a high fraction (~85%) of the film surface is oriented to (111) for the samples with very long-time air exposure (24 h). These results indicate that the interfacial oxide layers play an important role in not only control of the diffusion of atoms but also alignment of the crystal orientation.

To explain these preferential orientations in the interfacial oxide dependent AIC, we propose a model considering the phase transition of the native Al oxide, as shown in Fig.5. In the present study, the Al films are preferentially oriented to (111). Thus, we speculate that the Al oxide change into (111)-oriented γ -Al₂O₃ by sold-phase crystallization during long time annealing at 450°C [5]. Since the lattice constant of γ -Al₂O₃(111) is close to that of Si(111) (mismatch: $\sim 2.4\%$) [6], nucleation of Si(111) is expected at the γ -Al₂O₃/Al(111) interfaces. This well explains our results of the preferentially (111) orientation of the samples with long-time air exposure (B in Fig.5(a)), where inter-diffusion of Si and Al atoms takes long time due to the thick diffusion barrier, as shown in Fig.5(c). On the other hand, inter-diffusion of Si/Al atoms fast proceeds in the sample with short-time air exposure (A in Fig.5(a)), because of the very thin diffusion barrier. Thus, the Al oxide layers should remain amorphous in the stage of the Si nucleation. Recently, Schneider *et al.*, reported a preferentially (001) oriented nucleation of Si at amorphous-Al₂O₃/Al(111) interfaces based on the free energy calculation [7]. This should explain the preferential (001) orientation of the sample with short-time air exposure, as shown in Fig.5(b). This model qualitatively explains the present experimental results of preferential orientation.

4. Summary

Control of the preferential orientation of large grains $(>20 \ \mu\text{m})$ are realized by interfacial oxide controlled AIC. The oriented growth was well explained by the model con-

sidering the phase transition of the native Al oxide. This technique should be useful to achieve advanced thin-film solar cells and system-in-displays.

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Fig.1. Experimental procedure for Al-induced crystallization of a-Si on insulator.





Fig.2. Nomarski optical micrographs (a-d), EBSD mapping (e-h), and IPF (i-l) of AIC-Si layer for the samples (air exposure: without, 5 min, 60 min, and 24 h). These observations were performed along the normal direction to the sample surfaces.

Fig.3. Histograms of the orientation distribution for various air exposure times [5 min (a), 60 min (b), and 24 h (c)].



Fig.4. Fraction of (001) and (111) orientation as a function of the air exposure time.



Fig.5. Model for interfacial oxide layer-dependent AIC growth of Si (a), based on Si(001) and (111) nucleation for thin (b) and thick Al oxide (c).