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Microstructures of polycrystalline silicon films formed through explosive crystallization induced by flash lamp annealing

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1. Introduction

Thin-film polycrystalline silicon (poly-Si) films have been expected as a next-generation solar cell material. Solar cells with a high conversion efficiency (>10%) have been demonstrated using poly-Si films formed by solid-phase crystallization (SPC) of precursor a-Si films through hour-order furnace annealing [1]. The application of a rapid crystallization technique would lead to higher throughput, and also enables us to use low-cost substrates with poor thermal tolerance.

Flash lamp annealing (FLA) is an annealing technique using a millisecond-order pulse light, and can crystallize micrometer-order-thick a-Si films without heating of whole glass substrates due to its appropriate annealing duration. We have demonstrated that FLA can form 4.5-µm-thick poly-Si films even on soda lime glass substrates [2], and the flash-lamp-crystallized (FLC) poly-Si films can be processed to solar cells demonstrating rectifying and photovoltaic properties [3]. We have also clarified that the flash-lamp-induced crystallization progresses laterally through explosive crystallization (EC), autocatalytic crystallization associated with the liberation of latent heat [4]. The EC induced by FLA leaves behind periodic structures along the lateral crystallization directions [5]. The FLC poly-Si films have two regions with different features such as grain size and surface morphology, which form the periodic structures [5]. For the further understanding of these microstructures, we have precisely investigated the microstructures of FLC poly-Si films by means of cross-sectional transmission electron microscopy (TEM).

2. Experimental procedure

Cr adhesion films of 60 nm in thickness were first sputtered on quartz glass substrates of 20×20×0.7 mm^3 in size, followed by the deposition of 4.5-µm-thick a-Si films by catalytic chemical vapor deposition (Cat-CVD). FLA was performed under pulse duration of 5 ms and irradiance of approximately 20 J/cm^2. Only one shot of flash irradiation is performed for one sample. We selected a partially crystallized Si film sample for cross-sectional TEM observation. The surface appearance of the FLC poly-Si film is shown in Fig. 1, indicating that lateral crystallization is ignited at film edges and crystallized area expands towards center. A cross section for the TEM observation was formed at a a-Si/c-Si boundary. According to the results of Raman spectroscopy, the crystallized parts have a high crystalline fraction close to unity, which is consistent with those previously reported [2,5].

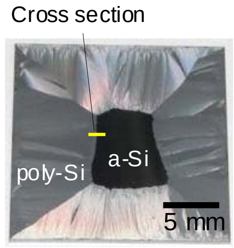

Fig. 1 Surface appearance of a FLC poly-Si film used for TEM observation. The position of cross section is also indicated.

3. Results and discussion

Figure 2 shows the low-magnification cross-sectional TEM image of the FLC poly-Si film. One can clearly see

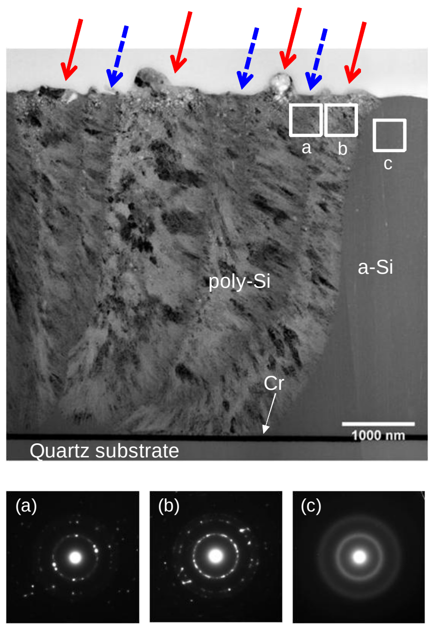

Fig. 2 Cross-sectional TEM image (bright field) of the FLC poly-Si film. Straight and dashed arrows indicate the parts of large-grain and fine-grain regions, respectively. EBD patterns recorded at fine-grain, large-grain, and amorphous regions are also shown.

two characteristic regions in the image: a region containing surface projections and relatively large, stretched grains with sizes of more than 100 nm, and a region with flat surface and containing no 100-nm-sized large grains. Electron beam diffraction (EBD) patterns of the two regions, also shown in Fig. 2, reveal that the former has higher degree of orientation than the latter. The EBD pattern of the position c, approximately 1 μm distant from the a-Si/c-Si boundary, indicates a complete halo ring, showing quite abrupt phase change at the boundary.

Figures 3(a) and (b) show bright- and dark-field images of the fine-grain region of the FLC poly-Si film. These images clearly show the existence of 10-nm-sized fine grains. Figure 3(c) shows the lattice image of the fine-grain region. One can confirm individual grains in the image, and amorphous phase is hardly seen between them. These results indicate that the fine-grain regions consist of randomly-oriented, densely-packed fine grains with little amorphous phase.

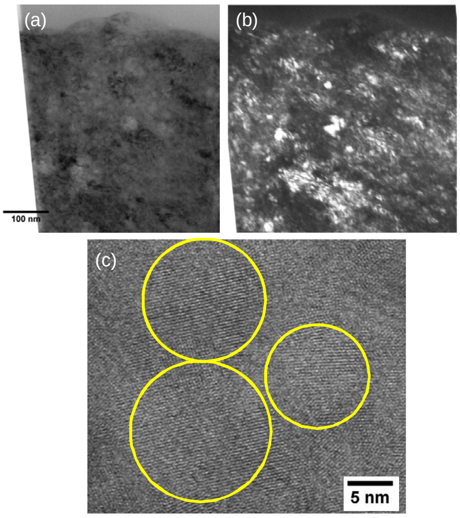

Fig. 3 TEM images of the fine-grain region of the FLC poly-Si film: (a) bright-field image, (b) dark-field image, (c) lattice image.

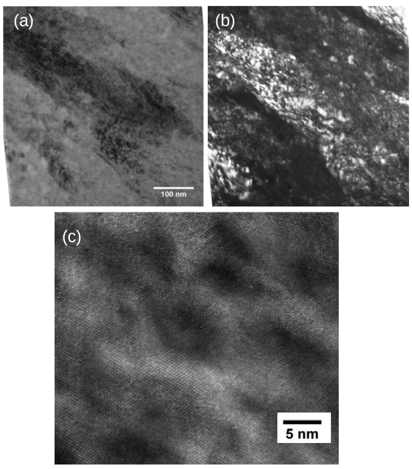

Fig. 4 TEM images of the large-grain region of the FLC poly-Si film: (a) bright-field image, (b) dark-field image, (c) lattice image.

Figures 4(a) and (b) show bright- and dark-field images of the large-grain region of the FLC poly-Si film. In contrast to the fine-grain region, large-sized grains are clearly seen. The direction of the grain stretching probably corresponds to the direction of thermal gradient during EC, and liquid-phase epitaxy (LPE) is most likely as their formation mechanism. Figure 4(c) shows the lattice image of the large-grain region of the FLC poly-Si film. No clear grain boundaries are seen in the image, which is also a clear indication of the formation of large grains.

We have mentioned the formation mechanism of the two regions in the previous paper: the fine-grain region is formed only through solid-phase nucleation (SPN), whereas the large grain region is governed by both SPN and LPE [5]. The results of TEM images are completely consistent with the proposed crystallization mechanisms. In the point of view of solar cell application, the FLC poly-Si with high crystalline fraction would lead to the effective transport of photogenerated carriers, while the passivation of grain boundaries would be one of the most important issues to realize high-efficiency solar cells using FLC poly-Si films because of the absence of a-Si tissues passivating grains, unlike CVD microcrystalline Si films.

4. Conclusions

TEM observations have clarified the difference of the microstructures of fine-grain and large-grain regions of FLC poly-Si films. Fine-grain region consists of randomly oriented, densely-packed fine grains of approximately 10 nm in size, whereas large-grain regions have stretched grains, probably formed through liquid-phase epitaxy.

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