

Effect of starting point formation on the crystallization of amorphous silicon films by flash lamp annealing

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Abstract

Flash lamp annealing (FLA) with millisecond-order pulse duration can crystallize μm -order-thick amorphous silicon (a-Si) films on glass substrates through explosive crystallization (EC). We investigate the effect of partially stacked a-Si parts on the crystallization of precursor a-Si films. Crystallization is initiated at the stacked a-Si part consisting of a-Si films formed by sputtering and catalytic chemical vapor deposition (Cat-CVD). Raman spectroscopy and optical microscopy reveal that the mechanism of the crystallization started from the stacked a-Si part is identical to the EC which we have reported previously. Partially stacked a-Si can thus be utilized for the starting point of EC induced by FLA.

1. Introduction

Thin-film polycrystalline silicon (poly-Si) solar cells formed on low-cost substrates, such as glass substrates, have been receiving attention as next-generation solar cells because of low material usage and high stability against light soaking. One of the methods to obtain poly-Si films is the crystallization of precursor amorphous Si (a-Si) films by annealing. Flash lamp annealing (FLA) is a millisecond-order annealing technique using pulse emission from a Xe lamp array, and can crystallize μm -order-thick a-Si films without serious thermal damage to glass substrates [1]. We have thus far confirmed the crystallization of a-Si films with a thickness of $>4\ \mu\text{m}$ [1]. The crystallization of a-Si films induced by FLA takes place laterally, and is driven by the release of latent heat corresponding to the enthalpy difference between crystalline Si (c-Si) and a-Si phases. This particular crystallization mechanism has been known as explosive crystallization (EC) [2,3]. EC is thought to start from high temperature parts [3]; however, we have not succeeded in the control of the initiation of EC thus far. The control of the initiation of EC will be essential particularly for the formation of large-area poly-Si films. Thicker a-Si films tend to have higher temperature than thinner ones [4], and the initiation of EC with high controllability is expected by using precursor a-Si with partially thick parts. In this work, we attempted to use a-Si films with partially stacked a-Si parts to form poly-Si films by FLA, aiming at the precise control of the initiation point of EC.

2. Experimental

We used alkali-free (Corning Eagle XG) flat glass substrates with a size of $19.8 \times 19.8 \times 0.4\ \text{mm}^3$. After the ultrasonic cleaning of the glass substrates in Semico Clean, ethanol, and pure water for ~ 10 minutes, respectively, Cr films with a thickness of 200 nm were deposited on them by sputtering. a-Si films with a thickness of $\sim 2\ \mu\text{m}$ was deposited along sides of the Cr films by sputtering by covering the rest of the Cr films with glass plates. a-Si sputtering was performed at a pressure of 1.1 Pa and at a Ar flow rate of 40 sccm. a-Si films were then deposited on the entire surface by catalytic chemical vapor deposition (Cat-CVD) at a catalyst temperature of $1800 \pm 50\ ^\circ\text{C}$, a substrate temperature of $200\ ^\circ\text{C}$, and at SiH_4 and H_2 flow rates of 50 and 10 sccm, respectively. The cross-sectional schematic of a precursor a-Si structure is shown in Fig. 1. A single shot of 7-ms-duration flash lamp pulse with a fluence of $\sim 12\ \text{J}/\text{cm}^2$ was supplied for each sample pre-heated at $500\ ^\circ\text{C}$ in argon (Ar) atmosphere. We evaluated the crystallization of Si films by Raman spectroscopy using the 632.8 nm line of a He-Ne laser. The full width at half maximum (FWHM) of a c-Si Raman peak obtained from a reference c-Si wafer was $\sim 3.5\ \text{cm}^{-1}$. The surfaces of Si films after FLA were observed by optical microscopy.

3. Results and discussion

Figures 2(a) and 2(b) show the surface images of Si films after FLA for the samples with and without partially stacked a-Si. Gray-colored parts are observed in Fig. 2(a), particularly in and near the stacked a-Si region, while no color change is seen in Fig. 2(b). Figure 3 shows the Raman spectra of the Si films, in which (1)–(3) correspond to the

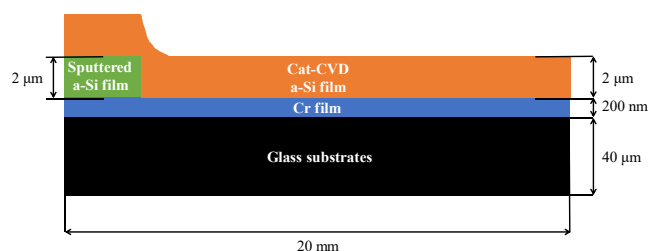


Fig. 1 Cross-sectional schematic of a precursor a-Si structure.

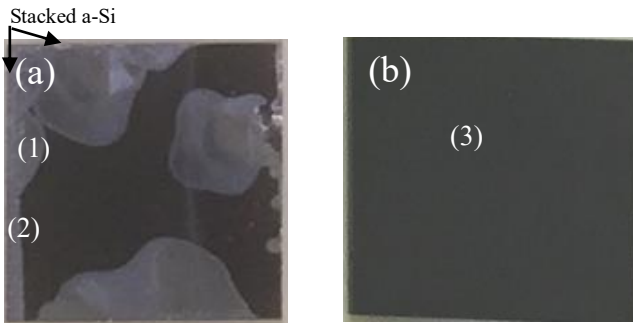


Fig. 2 Surface images of Si films after FLA for the samples (a) with and (b) without stacked a-Si.

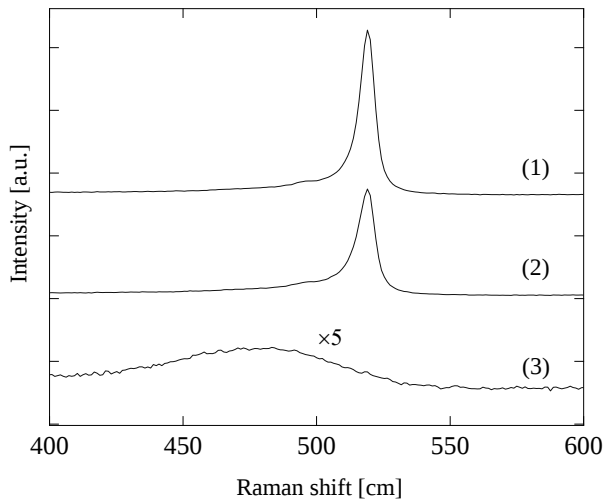


Fig. 3 Raman spectra of Si films after FLA obtained from the parts shown in Figs. 2(a) and 2(b).

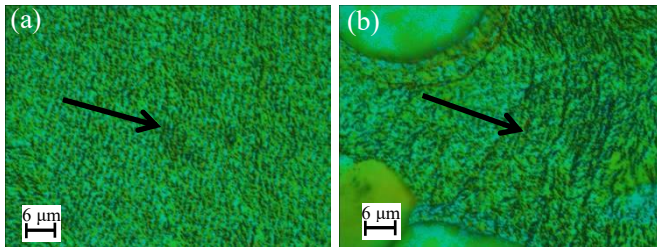


Fig. 4 Surface optical microscopic images of FLC poly-Si films observed on (a) region (1) and (b) region (2). Arrows on the images indicate the direction of EC.

regions shown in Figs. 2(a) and 2(b). The Raman spectra of regions (1) and (2) have a peak at around 520.5 cm^{-1} , corresponding to c-Si phase, while the spectrum of region (3) consists of a weak, broad peak at $\sim 480 \text{ cm}^{-1}$ originating from a-Si phase. These facts indicate that only the Si film with the partially stacked a-Si is crystallized by FLA. The Raman spectra of the flash-lamp-crystallized (FLC) poly-Si of regions (1) and (2) show high crystalline fraction close to unity, and a full width half maximum (FWHM) of $\sim 7 \text{ cm}^{-1}$ in these regions is larger than that of a c-Si wafer ($\sim 3.5 \text{ cm}^{-1}$). This fact indicates the existence of a number of nanometer-sized grains in the FLC poly-Si. One can see a slightly larger ($\sim 1 \text{ cm}^{-1}$) Raman shift value for the stacked Si region (2) compared to unstacked Si region (1). This is because of

the compressive stress of sputtered a-Si [5].

Figure 4 shows the surface optical microscopic images of an FLC poly-Si film observed on regions (1) and (2). One can clearly see periodic surface microstructures with an interval of $\sim 1 \mu\text{m}$ along a lateral crystallization direction. This characteristic microstructures are considered to be formed through EC with the alternative emergence of liquid-phase and solid-phase crystallization [4]. The existence of these microstructures means that the partially stacked precursor a-Si films are crystallized in the same EC mechanism as that in a-Si without partially stacked region which we have previously reported [3]. This result indicates that the stacked part can work as the starting point for the EC of a Cat-CVD a-Si film. As mentioned previously, the ease of crystallization depends on the reached temperature of a-Si during FLA, and thicker a-Si tends to have higher temperature. Once EC is initiated at the partially stacked part, heat generated by the crystallization propagates to a surrounding unstacked part, and EC continues there.

It should be noted that crystallization starts not only from the partially stacked Si part but also from other edges with no stacked a-Si, as shown in Fig. 2(a). One possible reason for this phenomenon is that a-Si films formed on the side surfaces of glass substrates during Si sputtering also acts as starting points for the EC of Cat-CVD a-Si films. This could be another way of controlling the initiation of EC.

4. Conclusions

We have investigated the effect of partially stacked a-Si part on the initiation of the EC of Cat-CVD a-Si films during FLA. Crystallization starts from the stacked a-Si region and expands to unstacked region. According to the experimental results of Raman spectroscopy and optical microscopy, the crystallized parts have particular characteristics of poly-Si films formed through EC with alternative emergence of liquid-phase and solid-phase crystallization. These facts indicate that the stacked a-Si can work as the starting point of the EC of a-Si films during FLA.

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