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Lateral Crystallization of Amorphous Silicon films Induced by Flash Lamp Annealing

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Abstract. Flash lamp annealing (FLA) induces lateral crystallization of a few μm-thick precursor amorphous silicon (a-Si) films from film edges toward center. This lateral crystallization can be understood as explosive crystallization (EC), driven by the release of latent heat, and leaves behind periodic microstructures in poly-Si films with an interval of approximately 1 μm. Based on the results of crystallization induced by multi-pulse FLA, we estimate the velocity of lateral crystallization to be 2.5-5.0 m/s. This value is significantly smaller than that of liquid-phase EC of ~15 m/s, suggesting that observed EC includes solid-phase process.

Introduction

Thin-film silicon (Si) is an attractive photovoltaic material because of low Si consumption and resulting high adaptivity for future enormous increase in photovoltaic production. There are, however, some disadvantages to be overcome for chemical vapor deposited (CVD) Si thin films such as light-induced degradation for amorphous Si (a-Si) and difficulty of realizing high deposition rate for microcrystalline Si (μc-Si). Hence, other approaches to form polycrystalline Si (poly-Si) films on low-cost substrates have been proposed [1,2,3]. In particular, poly-Si films formed through solid-phase crystallization of a-Si films by conventional furnace annealing has demonstrated 10%-efficiency solar cells [1]. This crystallization process, however, requires hour-order, time-consuming annealing, resulting in low throughput, and we requires rapid crystallization technique. Another disadvantage of long-duration furnace annealing is thermal-equilibrium treatment, which prevents us to use substrates with poor thermal tolerance.

To overcome this issue, we have investigated flash lamp annealing (FLA), with millisecond-order duration, for a technique to crystallize micrometer-order-thick a-Si films. The millisecond-order duration of FLA leads to the sufficient heating of a few μm thick a-Si films with avoiding thermal damage to entire glass substrates. We have actually confirmed the formation of poly-Si films with a thickness up to 4.5 μm on quartz and conventional soda lime glass substrates [4,5]. We have also observed that hydrogen (H) atoms in Si films are hardly desorbed off during FLA, and formed poly-Si films contains H atoms on the order of 10^{21} /cm^3, which can be effectively utilized to terminate dangling bonds in flash-lamp-crystallized (FLC) poly-Si films by post-furnace annealing [6]. Diode and solar cell operations have been confirmed in devices fabricated using FLC poly-Si films [7]. Interestingly, the crystallization induced by FLA takes place laterally, and in this paper, we discuss the mechanism of this particular crystallization induced by FLA.

Experimental details

First, we deposited Cr films on 20×20×0.7 mm^3-sized quartz glass substrates by RF sputtering in order to prevent the peeling of Si films during FLA [4,5]. We then deposited 2.0-4.5 µm-thick a-Si films by catalytic CVD (Cat-CVD). The detailed deposition conditions have been summarized elsewhere [8]. We also attempted sputtered a-Si films as precursor films. We have confirmed the formation of FLC poly-Si films by using sputtered a-Si films and similar microstructure to those formed from Cat-CVD a-Si films.

Figure 1 shows the schematic diagram of an FLA system. Flash lamp light has a broad spectrum mainly in visible range, which can be seen elsewhere [9]. FLA was performed under a fixed duration of 5 ms, while its irradiance was systematically changed around 20 J/cm^2. Flash lamp light was irradiated onto an entire Si film surface with sufficient areal uniformity. Besides a single-pulse FLA system we have used so far, we also newly utilized a multi-pulse FLA system, whose pulse light is schematically illustrated in Fig. 2. The frequency of individual pulse emission can be systematically changed in the multi-pulse FLA system. The total irradiance of a set of a multi-pulse was also approximately 20 J/cm^2.

The microstructure of FLC poly-Si films was observed with scanning electron microscope (SEM), atomic force microscope (AFM), and transmission electron microscope (TEM). Crystallization and crystalline fraction of FLA Si films were evaluated by means of Raman spectroscopy.

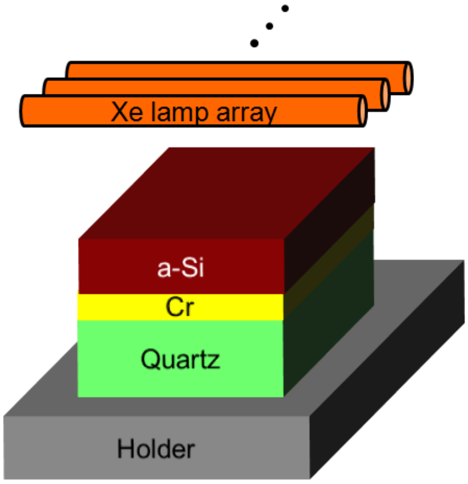

Fig. 1 Schematic diagram of an FLA system used in this study.

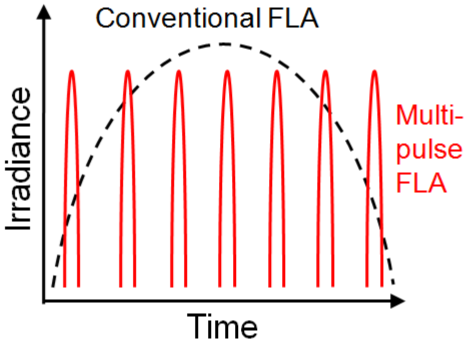

Fig. 2 Schematics of conventional (single-pulse) and multi-pulse flash-lamp light.

Results and discussion

Figure 3 shows surface images of Si films after FLA with various lamp irradiances. One can see partial crystallization in the vicinity of Si film edges after low-irradiance FLA, and crystallized area enlarges towards center with increasing lamp irradiance. These facts indicate that crystallization starts from edges and propagates laterally. We can also observe rainbow-colored surfaces on FLC poly-Si films.

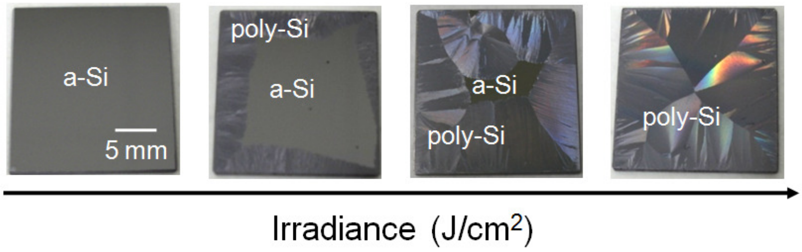

Fig. 3 Surface images of Si films after FLA with various lamp irradiances.

Figure 4 shows SEM images of an FLC poly-Si film. One can see a periodic microstructure along a crystallization direction. This structure is formed spontaneously during crystallization induced by FLA. Surface root-mean-square roughness (R_{rms}) of the FLC poly-Si surface, evaluated by AFM measurement, is typically 120 nm, which is significantly larger than R_{rms} of precursor a-Si films of less than 10 nm. These periodic structures act as an optical grating, resulting in the emergence of rainbow-colored surface on FLC poly-Si films. We have confirmed that this surface roughness effectively acts to reduce optical reflection on the FLA poly-Si surface [10].

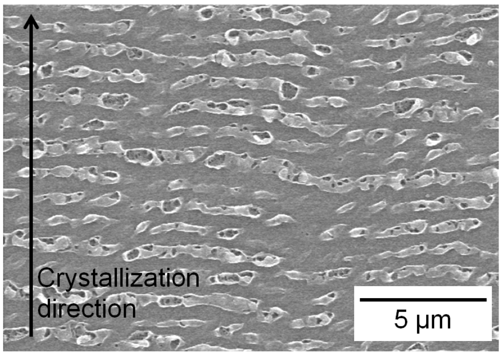

Fig. 4 Surface SEM image of an FLA poly-Si film.

Figure 5 shows the cross-sectional TEM image of an FLC poly-Si film. The cross section was formed along a crystallization direction. Periodic surface roughness, shown in the SEM image, is also shown in the TEM image. We can also see periodic structures inside poly-Si, which consist of two kinds of regions with different grain features. One region, large-grain region, contains surface protrusions on the surface and relatively large, a few hundred nm-sized grains inside Si film, some of which are stretched along the lateral crystallization direction. The other region, fine-grain region, connects to flat surface and includes no hundred-nm-sized large grains.

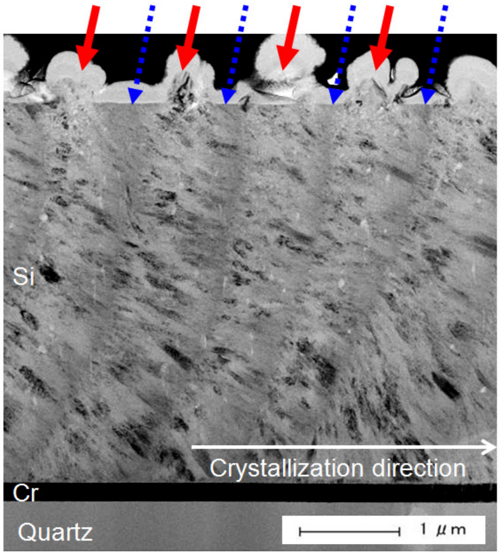

Fig. 5 Cross-sectional TEM image of an FLC poly-Si film. Solid and dashed arrows indicate large-grain and fine-grain regions, respectively.

The onset of crystallization at Si film edges can be understood as additional heating by angled flash lamp light into film edges. One may worry about thermal loss at Si film edges through convection or radiation. These effects are, however, negligible during ms-order short-duration process. More quantitative consideration about heat loss at Si film edges has been summarized elsewhere [11].

Since the lateral crystallization propagates ~1 cm during 5-ms flash lamp pulse, the crystallization velocity must be more than 2 m/s. Such a rapid lateral crystallization can be explained only by explosive crystallization (EC), autocatalytic crystallization driven by the release of latent heat [12,13,14,15], and both liquid-phase [16,17,18] and solid-phase EC [18,19] have been investigated experimentally and theoretically. FLC poly-Si films contain two regions with different grain features existing periodically, as shown above, and we can consider that these two regions are formed through different crystallization mechanisms. As we have discussed in a previous paper [11], the large-grain region is formed thorough a mixed phase of solid-phase nucleation and liquid-phase epitaxy, while the fine-grain region is formed only through solid-phase nucleation. This means that two types of EC occurs alternatively during one flash irradiation. The reason of this has not been fully clarified, and we will perform an analysis of lateral thermal diffusion including the effect of heat generation by solid-phase and liquid-phase crystallization.

Figure 6 shows the surfaces of FLC poly-Si films formed from sputtered a-Si films using the multi-pulse FLA system. We have confirmed that poly-Si films crystallized by using multi-pulse flash lamp light have microstructures and crystalline fraction similar to FLC poly-Si films formed using a single-pulse FLA system. One can see macroscopic stripes on the FLC poly-Si films. Since we have observed such stripes only on FLC poly-Si films formed using multi-pulse FLA and the width of stripes systematically varies depending on pulse emission frequency, we can understand that these stripes are formed by discrete pulses. By using known pulse emission frequency and the width of stripes, we can easily estimate the velocity of EC induced by FLA to be 2.5-5.0 m/s. In the case of simple liquid-phase EC, its velocity is governed by the speed of liquid-phase epitaxy that depends on the temperature of supercooled Si melt, and an EC speed of ~15 m/s has been experimentally confirmed [16]. On the other hand, an EC velocity of 0.5-3.0 m/s has been theoretically predicted in the case of complete solid-phase EC [19]. The EC velocity estimated for FLC poly-Si films is between the two typical values, indicating the validity of the proposed mechanism of mixed-phase EC. The estimated EC velocity is also consistent with the fact that 2×2 cm^2-sized a-Si films are not wholly crystallized when we use 1-ms pulse light for crystallization [20].

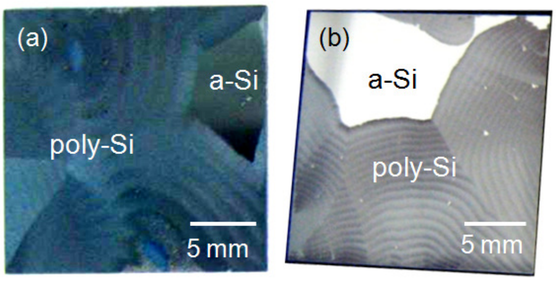

Fig. 6 Surface images of FLC poly-Si films formed using a multi-pulse FLA system with pulse frequency of (a) 5 kHz and (b) 7 kHz, respectively.

Summary

FLA induces EC of a-Si films, leaving behind periodic microstructures on the surface of and inside FLC poly-Si films. This periodic structure consists of two kinds of characteristic regions formed through different mechanisms: one is partially related to liquid-phase growth and the other is governed only by solid-phase process. This fact is consistent with an EC velocity of 2.5-5.0 m/s, estimated from the results of multi-pulse FLA, larger than the velocity of solid-phase EC and smaller than that of liquid-phase EC.

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