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In situ observation of grain-boundary development from a facet-facet groove during solidification of silicon

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ABSTRACT

The evolution of the facet-facet groove at grain boundaries in multi-crystalline Si during solidification was investigated by *in situ* observation of the melt/crystal interface. The grain boundaries changed their propagation direction without any new grain formation or grain boundaries interaction during crystal growth. We monitored the melt/crystal interface over time and carefully estimated the growth velocities on two facets. We found that the facet velocities are different in some of our experimental observations and the development of random grain boundary is strongly dependent on the facet velocities. On the basis of our experimental observations, we discussed the direction of random grain boundary during solidification by considering the growth velocities on two facets.

1. Introduction

Multi-crystalline silicon (mc-Si) grown by directional solidification is widely used in photovoltaic applications because it is very cost-effective. Mc-Si is used in more than 60% of solar cells today [1]. The photovoltaic properties of mc-Si are strongly dependent on its grain structure, e.g., its grain size, crystallographic orientation, and the presence of defects [2,3]. Grain boundaries in mc-Si are very important because lattice defects, which interact with minority carriers, are typically located at grain boundaries [4–7]. Moreover, grain boundaries act as getters, attracting impurities, which results in impurity segregation at grain boundaries [8–10]. Therefore, the development and control of grain boundaries in mc-Si ingots is very important [11].

Several groups have studied the growth of grain boundaries through experiment [12–14] and simulation [15,16]. Duffar et al. discussed the morphology of the grain-grain-liquid triple phase line in mc-Si from the theoretical point of view and presented three possible grain boundary groove configurations at the tri-junction: rough-rough, rough-facet, and facet-facet [17]. These authors also argued that the grain boundary groove configuration determines the grain boundary direction. One possibility is in a facetted/facetted grain boundary groove, the model states that the grain boundary propagates along the bisector plane of the two facets in a facet-facet groove. Duffar et al. suggested that the growth velocities of the two facets are the same because they experience the same undercooling. At this stage, in a facet-facet groove, grain boundary development should follow the bisector between the two facets. Experimentally, Tandjaoui et al. directly observed the evolution of grain boundary grooves during solidification by using *in situ* X-ray imaging [18]. They showed that the grain boundary direction of a non $\Sigma3$ grain boundary followed the bisector angle of the two facets if the growth rates of the two facets were the same in a facet-facet groove. For a $\Sigma3$ {111} twin boundary, it followed the common {111} plane between two grains [13]. Also, recently, Lin and Lan examined various types of grain boundaries in mc-Si moving at different drift speeds in order to determine if they satisfied the bisector rule [19]. They obtained the eight {111} plane vectors by EBSD measurement and analyzed selected grain boundaries for all possible bisector planes. They concluded that only random grain boundaries at high growth rates and some CSL grain boundaries obeyed the bisector rule for facet-facet grooves. In those models, the difference in growth velocities of two facets was not considered because the undercooling in the melt at the grain boundary groove was assumed to be uniform. On the other hand, the experimental study for grain boundary development from a facet-facet grooves during crystal growth is limited. Therefore, direct experimental evidence is needed to clarify the mechanism of grain boundary development during solidification.

In this study, we monitored the directional growth of mc-Si

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using *in situ* observations, and focused on the evolution of the grain boundary groove at the melt/crystal interface. The experimental results suggested that random grain boundary development is strongly dependent on the growth velocities of the two facets of the groove. We discuss the development of random grain boundary for facet-facet grooves by considering the growth velocities of the two facets. The development model of grain boundary showed good agreement with our experimental results.

2. Experimental

An *in situ* observation system consisting of a furnace and microscope was used to observe the grain boundary grooves at the melt/crystal interface during solidification [20,21]. We prepared orientation-controlled Si single crystals, Si (100) and Si (110), in 1 mm thickness sheet form. The Si (100) wafer was rotated to set the growth direction to ⟨110⟩ + 20º-25° in order to obtain grain boundary groove that could be clearly examined by our observation system. The grain boundaries would be determined by two orientation-controlled Si sheet crystals. In doing so, the grain boundary between two seed grains was expected to be a random grain boundary. Two Si sheet crystals were set between two quartz plates to fix the position of the Si sheet crystals and keep the Si melt surface flat during solidification, as shown in Fig. 1(a). The furnace chamber was evacuated to low vacuum and filled with ultrapure argon gas. Two graphite heaters were used to control the furnace temperature during melting/crystallization of the Si sheet crystals. The two graphite heaters were set to different temperatures to generate a temperature gradient during solidification and initiate melting from the same end of the Si sheet crystals. The temperatures of the two heaters were reduced before the Si sheet crystals were fully melted to initiate crystal growth from the unmelted seed crystals, as shown in Fig. 1(b). Thus, the growth of the grain boundary between two orientation-controlled grains could be observed during solidification. A video recording of the process was made through a 50x magnification lens for analysis. After crystallization, the quartz plate on the observed side was removed for crystallography orientation analysis. Grain orientation and grain boundary characterization was performed through electron backscattering diffraction (EBSD) using a JSM-6610 A (JEOL) high-performance scanning electron microscope equipped with OIM Data Collection software.

3. Results

Fig. 2 shows the melt/crystal interface motion for the sample consisting of Si (110) and Si (100) in the ⟨110⟩ and ⟨110⟩+20° growth directions, respectively. Traces of the deepest of grain boundary groove at the melt/crystal interface revealed the development of grain boundary between two grains from remnant seed crystals. The grain boundary groove was formed in the early stages of crystallization and deepened with time because of the lower growth rate on the groove side compared to the global interface. The two facets forming the groove were clearly identified on the growth surface at t = 0 s. The groove deepened to its maximum depth, with the grain boundary direction close to the lower facet until around t = 5 s. Following this the grain boundary groove transferred from facet-facet to facet-rough and reduced its depth. Meanwhile, the grain boundary changed direction from left to right. At t = 16.25 s, the grain boundary groove underwent a groove transformation and started to deepen again. Also, the direction of grain boundary reverted to left. The grain boundary groove transformation could be due to rapid growth or new grains formation at the grain boundary groove [18]. It has been shown that grain boundary grooves are among the sources of {111} Σ3 twin boundaries due to the {111} facets [13,22]. Also, we recently found that grain competed at the encounter of grain boundaries and developed other grain boundaries with new neighbor grain [23]. It is therefore important to investigate the grain characteristics to clarify the reason of change in grain boundary direction in experimental observation. For the above purpose, the EBSD measurements employed in the area observed by *in situ* experiment.

The grain boundaries were characterized and the grain orientations were determined by EBSD measurements. Fig. 3(a) and (b) and (c) show a scanning electron micrograph, an image quality (IQ) map, and an orientation map for the growth direction (TD),

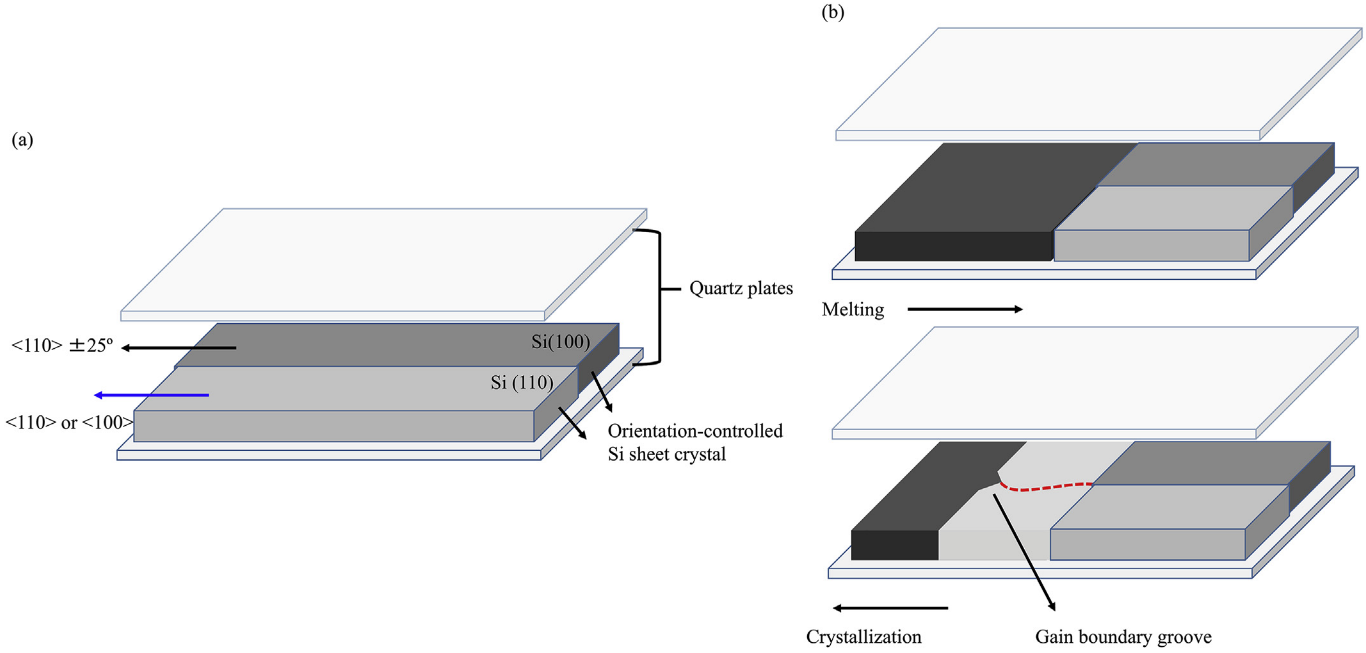

Fig. 1. (a) Schematic of Si sheet crystal preparation for observation of grain boundary development. (b) Schematics illustrating melting and crystal growth.