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In situ observation of interaction between grain boundaries during directional solidification of Si

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ABSTRACT

The interaction between grain boundaries (GBs) at a silicon crystal/melt interface was studied using in situ observations during directional solidification. Two small-angle GBs (SAGBs) interact with each other to produce a new SAGB with increased misorientaion. However, the interaction between Σ3 GBs and SAGBs reveals a different phenomenon. The extending directions and misorientations of the SAGBs and Σ3 GBs show no change before and after the convergence of these two planar defects at the crystal/melt interface.

Multicrystalline silicon (mc-Si) grown through directional solidification is the dominant material in photovoltaic applications. Grain boundary (GB) in mc-Si is one of the most significant factors deteriorating conversion efficiency of solar cells. However, not all GBs have a negative impact on solar cell efficiency. The coherent coincidence site lattice (CSL) {111}Σ3 GBs are electrically inactive [1,2]. CSL GBs with low CSL indices, such as Σ3 and Σ9, generally contain lower concentrations of precipitated metals than GBs with high CSL indices or non-CSL GBs [3]. Therefore, either a reduced number of GBs or dominant {111}Σ3 GBs was once considered to be a promising way to achieve high-quality mc-Si material [4–6]. In recent years, a new growth method has been used to produce high-performance multicrystalline silicon (HP mc-Si), which exhibits even higher conversion efficiency when used as a substrate material for solar cells [7,8]. HP mc-Si has lower dislocation density than traditional mc-Si and possesses a high fraction of random GBs. The distribution of GBs has a critical influence on the defect distribution and materials property. GB distribution is actually the result of interaction between GBs. Therefore, in this study, we have attempted to elucidate mechanisms behind GB interactions through in situ observation during crystal growth.

Experiments were performed using an in situ observation system consisting of a microscope and a crystallization furnace [9–11]. High purity silicon raw materials were placed in a quartz crucible with inner dimension of 22 mm × 13 mm × 8 mm and then melted completely under an argon atmosphere using a pair of resistive graphite heaters in the furnace. The two heaters were controlled to create a temperature gradient between both sides of the quartz crucible, and directional solidification of the silicon began from the side with the lower temperature. The experiments were conducted without seed crystals, so that the GBs were generated directly from the inner walls and extended directionally during solidification. This created an environment in which observation of the GB growth without intervention was possible. A high-speed microscopic camera was used to record the crystal growth process, including the formation and annihilation of GBs. The solidified silicon crystal samples were analyzed using electron backscattering diffraction (EBSD) to determine the grain orientations and the structure of the GBs.

Fig. 1(a) shows a series of snapshots captured during in situ observation of silicon crystal growth for sample A. The growth direction is leftward. Several small-angle GBs (SAGBs) are clearly observed growing almost perpendicular to the solidification interface toward the left. A pair of straight {111}Σ3 GBs, confirmed by EBSD measurement (see Fig. 2(d), crystal orientations are along the growth direction), grow slanting toward the lower left of the figure. {111}Σ3 GBs are well known to exhibit straight features in silicon crystals. This pair of {111}Σ3 GBs eventually met another pair growing from the lower part, again confirmed through EBSD (see Fig. 2(d)). The two pairs of {111}Σ3 GBs converged and only one pair finally remained. In contrast to this situation, Σ3 GBs did not interact with SAGBs during solidification. From the in situ observation snapshots shown in Fig. 1(a), the SAGBs grew through {111}Σ3 GBs without changing the direction of growth. Both types of GBs kept moving along the initial directions without perceptible changes. Fig. 1(b) shows a series of snapshots for sample B during solidification with a leftward growth direction. A SAGB approaches a pair of Σ3 GBs, also confirmed by EBSD measurement (see Fig. 3(b)), in the beginning. When the SAGB met the first Σ3 GB, the

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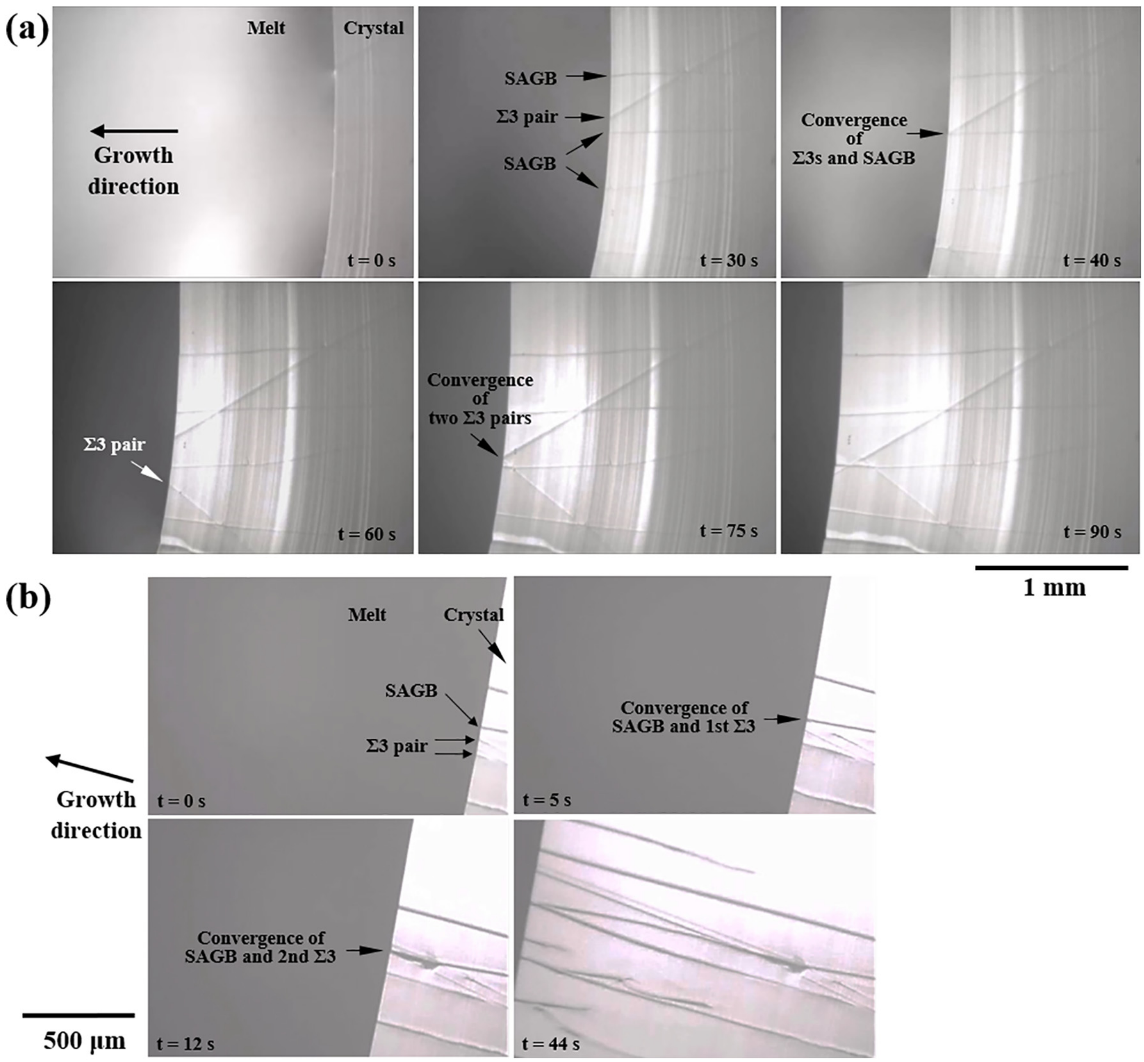

Fig. 1. Crystal/melt interface during directional solidification of mc-Si for (a) sample A and (b) sample B taken by microscopic camera at certain time points. The crystal growth direction is leftward.

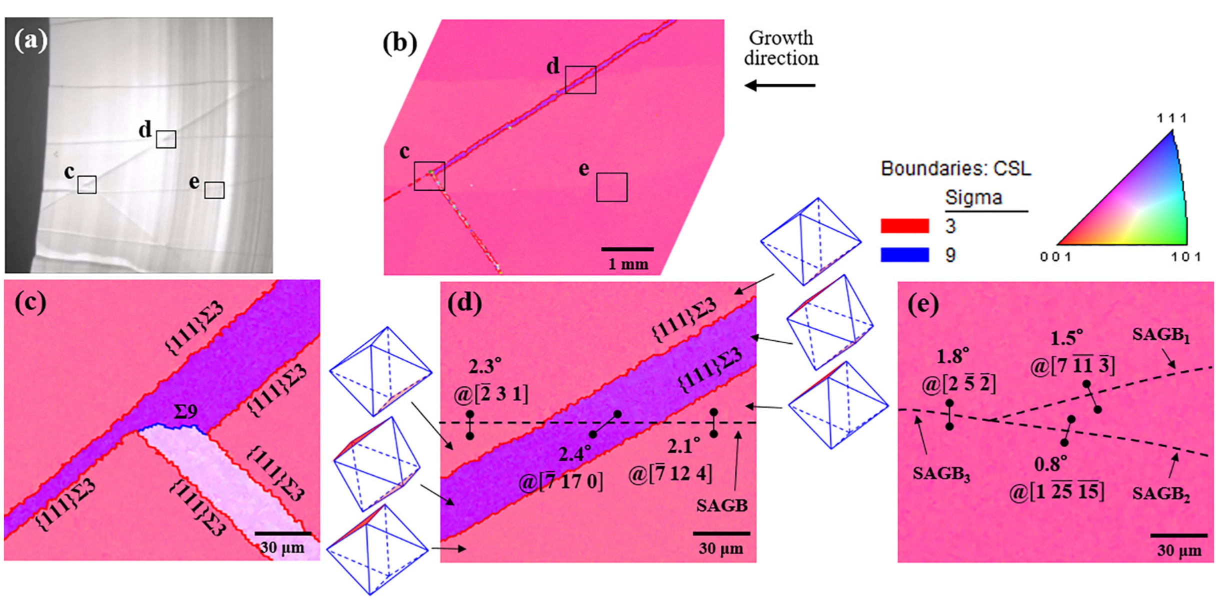

Fig. 2. (a) Image recorded by optical microscope for sample A. Orientation images by EBSD along the growth direction for (b) whole area of sample A, (c) interactions between Σ3 and Σ9 GBs, (d) interactions between SAGBs, and (e) interactions between Σ3 GBs and a SAGB. The growth direction is leftward.