Comprehensive study of sub-grain boundaries in Si multicrystals toward defect engineering for high-efficiency solar cells

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Control of microstructures and defects in Si multicrystals, to supply high-quality wafers, is of crucial importance toward further improvement of conversion efficiency of solar cells. Among various defects, we have recently revealed that “sub-grain boundaries”, which consist of dislocation clusters in a crystal grain, are serious ones based on estimation of recombination velocity of carriers by combining spatially resolved X-ray rocking curve analysis and surface photovoltage method [1]. Therefore, it is necessary to somehow suppress generation of sub-grain boundaries to obtain high-quality Si multicrystals for solar cells.

In this contribution, we present comprehensive study on sub-grain boundaries in Si multicrystals to include fundamental mechanisms of generation, spatial distribution, electrical properties, and impact on solar cells.

To disclose the generation mechanisms of sub-grain boundaries, we performed model crystal growth experiments using a seed crystal with artificially controlled grain boundary configuration, namely <110> tilt grain boundary (Fig.1 left). X-ray rocking curve analysis of the grown crystal revealed that sub-grain boundaries are generated from a grain boundary as evidenced by the peak shift in the rocking curve profile (Fig.1 right). Interestingly, the amount of the peak shift was found to be strongly dependent on the initial grain boundary configuration. This phenomenon was well correlated with the amount of the calculated local shear stress in the slip plane at the vicinity of the grain boundary by taking anisotropic elastic constants into account as shown in Fig.2. Therefore, it can be concluded that the introduction of the local shear stress around grain boundaries is the origin of the generation of sub-grain boundaries during directional growth.

The generation of sub-grain boundaries from grain boundaries was confirmed not only in the model crystal but also in Si bulk multicrystals grown by the practical casting method. The sample was cut from an ingot parallel to the growth direction to include the bottom part. At the initial stage of the crystal growth, random grain boundaries are necessarily formed by collision of randomly nucleated crystal grains. The generation of sub-grain boundaries was confirmed as multiple peaks in the rocking curve profile especially when we measured a region around random grain boundaries.
Resultantly, sub-grain boundaries are not homogeneously distributed but tend to densely localize in the narrow area around random grain boundaries and lengthen in the growth direction.

Furthermore, electrical properties of sub-grain boundaries and their impact on solar cell performance were investigated by electroluminescence (EL) imaging with external forward and reverse bias to monitor active defects and shunts, respectively. Decrease in the resistivity of the base crystal resulted in the appearance of bright spots around sub-grain boundaries in the EL image with reverse bias, which could be associated with avalanche breakdown sites (Fig.4). Reduction of shunt resistance by sub-grain boundaries was confirmed by EL imaging with energy filtering (Fig.5). These results further confirm that sub-grain boundaries are detrimental defects for solar cells.

In summary, we revealed that the sub-grain boundaries, which seriously affect solar cell performance, are generated from grain boundaries owing to the introduction of the local shear stress. Especially, random grain boundaries are shown to be the source of sub-grain boundaries to give large impact on inhomogeneous spatial distribution of sub-grain boundaries. This fundamental knowledge will be implemented to crystal growth method to obtain high-quality Si multicrystals for high-efficiency solar cells.

REFERENCES
Fig. 2 Calculated shear stresses in adjacent crystal grains to construct a <110> tilt grain boundary. $\theta_1$ and $\theta_2$ represent in-plane orientation to specify the grain boundary configuration. Gray circles show the configuration of the crystal grains of the grown crystals. It is seen that the amount of the shear stresses is in good agreement with the amount of the peak shift in the rocking curve profiles to be associated with generation of sub-grain boundaries.

Fig. 3 (a) Two-dimensional mapping of integrated intensity of X-ray rocking curve profiles of Si multicrystals grown by the casting method: Boundaries between black and bright regions correspond to random grain boundaries. (b) An example of modification of X-ray rocking curve profiles along a grain boundary as growth proceeds: Appearance of sub-grain boundary is evidenced by appearance of an additional peak.
Fig. 4 Electroluminescence images of a solar cell based on Si multicrysals with (left) forward and (right) reverse bias. Dotted red circle indicates sub-grain boundaries. These sub-grain boundaries were detected as bright spots in the image obtained by applying reverse bias especially when the base resistivity is decreased. This indicates that sub-grain boundaries are decorated with impurities, leading to reduction of shunt resistance.

Fig. 5 (a) Ratio of Electroluminescence intensities measured with 960nm and 800nm long pass filters. (b) Electroluminescence intensities measured with a 860nm long pass filter. The sub-grain boundaries were detected as darker area than random grain boundary in the image of (a). This indicates that the electrical currents are shunted more strongly in sub-grain boundaries region than that in random grain boundary region.