Kinetic behavior of solid-liquid interface during Si solidification from the melt

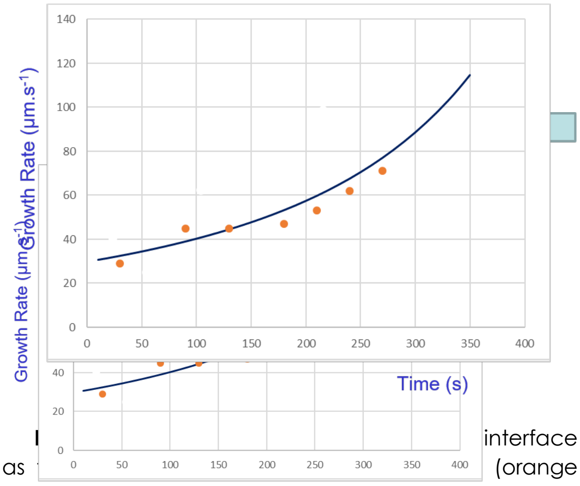
In situ observation of silicon solidification from the melt has shown that the facets, formerly expected to grow by 2D nucleation kinetic, are in fact surfaces vicinal to (111) facets. The variable terrace length is expected to depend on the facet-rough interface angle. The effect of the interaction of single dislocations with the vicinal facets is also described.

For two decades now, the IMR Tohoku is performing in situ measurements and visualization of the solid-liquid interface during solidification of metals and semiconductors, especially silicon. In parallel, more than 10 years ago, the group in Grenoble published a series of theoretical papers on the kinetics of grain boundaries in the growth of silicon from the melt. These papers predicted that a groove, constituted by (111) facets, should exist at the grain boundary-growth interface triple line.

The experimental results obtained in Sendai indeed showed that these facets and groove exist during silicon growth. The purpose of this two month visit, from 1st October to 30th November 2022, was to confront experiment and theory, for a better understanding of the grain boundary behavior during silicon growth.

During three one-hour seminars, I presented our understanding of Si kinetics, including dislocation behavior and twinning during growth. After discussions with all the researchers of Fujiwara laboratory, I more precisely focused my attention on the experimental results of two students working on their PhD, both with in-situ recording of Si growth. Shashank Mishra was using pre-oriented seeds in order to observe perfectly defined grain boundaries. Fan Wang on the contrary did not used seeds, so that she observed totally random grain boundaries.

A first step has been to determine the temperature gradient at the solid-liquid interface. As it was not possible to measure it precisely during the experiments, I developed a linear approximation of the heat flux in the sample, giving simultaneously the variation of growth rate and of temperature gradients (in liquid and solid) with time. As can be seen on the Figure 1, the agreement of the model with the measured growth rate was good and then the temperature gradients were reliable.



I interface as (orange dots), fit well with the theoretical calculation (blue line).

From these calculated temperature gradients, it was possible to measure the undercooling of the grain boundary grooves and of the growing facets.

In Shashank's experiments, thanks to the well oriented grains, it was possible to observe facets growing perpendicularly to the temperature gradient. It appeared immediately that the relationship between the growth rate and the undercooling of facets did not fit at all, neither with a 2D-nucleation growth law, neither with a dislocation-driven growth model. We finely concluded that the observed facets were indeed surfaces vicinal to (111) facets, growing through a terrace-step mechanism [1]. In order to better explain the results, we added the hypothesis that the terrace length depends on the angle between the vicinal surface and the rough solid-liquid interface.

Another result of these experiments was the angle measured between the grain boundary and the growth direction, in the case where no groove was observed. From this angle it has been possible to propose a ratio between the kinetic coefficients of the (100) and (110) oriented solid-liquid interfaces [2].

Concerning Fan's experiments, she observed large grooves at the grain boundary intersection with the solid-liquid interface. Here also, the measured undercooling did not fit to 2D-nuclation and vicinal surface growth kinetic was also concluded. From time to time a strange behavior occurred, leading to decrease and increase of the groove size and facet velocities. This phenomenon was explained by the periodic interaction with single dislocations impacting the facet kinetics [3]. Figure two shows an experimental picture of the groove variations and the results of a geometrical model that we developed on the basis of the dislocation interaction.

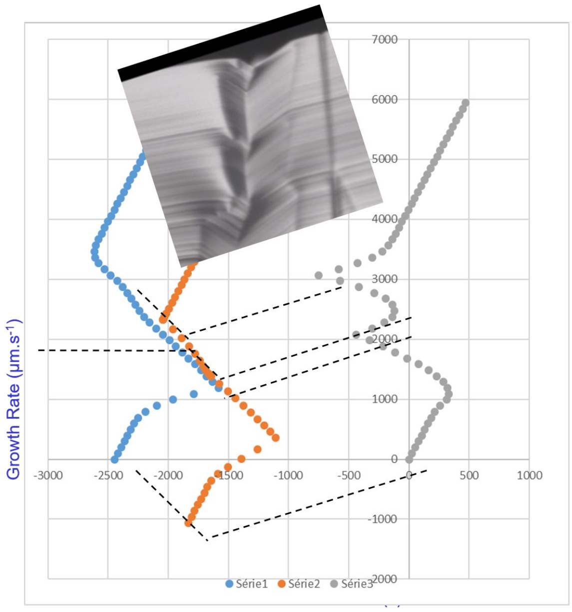


Fig. 2, shows the experimental observation of a grain boundary groove interacting with dislocations and the geometrical simulation of the kinetic mechanisms involved in these interactions.

In other experiments, it was observed that sometimes dislocations can pass through a twin during solidification, while in most experiments, they are blocked at the boundary. A model has been developed taking into account the possible continuity of gliding planes from one grain to the other through the twin boundary. It was shown that dislocations can glide along three common planes in the case of a $\Sigma 3$ twin and along one common plane only in the case of a $\Sigma 9$ twin.

In a final seminar, I presented to the laboratory a summary of our results and a number of recommendations for future work in the group, including the interest of numerical simulation for a better understanding of the experiments, and a series of experiments to perform in order to investigate more quantitatively the phenomena that we observed.

In conclusion, I would like to thank very much the institute for Materials Research of Tohoku University and Professor K. Fujiwara for this invitation to participate to this fascinating research program. Thanks to the excellent experimental work performed in this unit, we have been able to explain the kinetic behavior of the silicon facets and show that vicinal facet growth also exists in solidification and crystal growth from the melt. To the best of our knowledge, it is the first time that vicinal growth is demonstrated in the case of crystal growth from the melt.

References

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Poster presentation, ICCGE-19, July 30-August 4, 2023, Napoli, Italy
Paper for Scripta Materialia in progress

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