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## Development of 3-D interferometer for crystal growth

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To understand the mechanism of crystal growth in solution, it is very important to know the concentration field around the growing crystal, especially, around the crystal-solution interface. Growth mechanism changes as the supersaturation increases. The solute concentration near the crystal-liquid interface, however, is smaller than that of the bulk solution because the crystal grows by incorporating solute in the solution. Local concentration distribution in the crystal interface can cause instability of the form on the crystal surface, it is important to determine the concrete concentration distribution in the crystal interface to discuss the mechanism of the crystal growth. Therefore, to discuss the relation between the crystal growth mechanism and growth rate of the crystal, it is necessary to measure the concentration around the crystal-liquid interface, not of the bulk.

There were many previous researches of measurement of the concentration field, but many of them were two-dimensional (2-D) observations, namely, only from one direction. However, the information obtained by the 2-D observations is integrated along the direction of the observation, so the local information, e.g., concentration around the crystal-liquid interface, is not obtained.

To solve the problem on the 2-D observation, a method of computer tomography (CT) has been adopted by some authors. By using the CT method, one can reconstruct the information of the three-dimensional concentration field around the growing crystal based on 2-D observations obtained from several directions (3-D observation). Previous works of 3-D observations revealed the three-dimensional structure of solute convection around the growing crystal. However, there is quite a few observations of the concentration field around the crystal-solution interface.

In the present study, we carry out 3-D observation to measure the three-dimensional concentration field very close to the crystal-liquid interface growing in solution quantitatively. We newly develop two types of optics; microscopic Mach-Zehnder interferometer.

For quantitative 3-D measurement of concentration field, we developed 3-D microscopic Mach-Zehnder interferometer. 3-D observations by using our newly developed microscopic Mach-Zehnder interferometer can provide us the detailed information of concentration field around a growing crystal with high magnification and high sensitivity.

For the application of the 3-D observation by using our Mach-Zehnder interferometer, we measured the concentration field around protein crystal growing in solution. The growth rate of the protein crystal is quite low, so the decrease of concentration in solution should be much smaller. The interference fringes obtained are straight, but curved slightly only in a very narrow area close to crystal surface. In spite of the very small movements of the interference fringes, we successfully reconstructed the 3-D concentration field around the growing protein crystal. From the reconstructed image, we identified two low concentration regions on the vicinity of the crystal surface. These positions corresponded to the positions of spiral growth centers, which were observed by the confocal laser scanning microscope just after the growth experiment. The 3-D reconstruction of the concentration field around a protein crystal growing in solution was performed for the first time in this work.

This is the first time to confirm the correlation between the position of spiral growth center on the growing crystal surface and the low concentration region obtained by the 3-D reconstruction. The 3-D observation system we developed in this study is a powerful tool to obtain the experimental data concerning the interaction between the concentration field and the layer-by-layer growth mechanism on faceted crystal surface. It is expected that we can deepen the understanding of the layer-by-layer growth theory based on the result of the quantitative 3-D observation.