Abstract of Doctoral Dissertation

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Surface Melting of Polycrystalline Ice Thin Films
（氷多結晶薄膜の表面融解に関する研究）

A vast amount of ice exists on the earth, and surface melting of ice forms thin liquid water layers, so-called quasi-liquid layers (QLLs), on ice surfaces even below the melting point (0 °C). Because QLLs govern properties of ice surfaces, surface melting of ice plays crucially important roles in a wide variety of natural phenomena on the earth. To understand such phenomena, unraveling the surface melting of ice is indispensable. In nature, a large proportion of ice is present in a polycrystalline state. Hence, in this work the behavior of QLLs on polycrystalline ice was investigated.

Polycrystalline ice samples were prepared by quenching pure water using liquid N_2. Then the surfaces of the polycrystalline ice thin films thus formed were observed using a laser confocal microscope combined with a differential interference contrast microscope (LCM-DIM). In addition, the growth and sublimation of the polycrystalline ice thin films were monitored using a Linnik interferometer. The presence of QLLs on the polycrystalline ice thin films was proved, for the first time, by observing macroscopic fluidity of QLLs. During observations, temperature of ice samples and water vapor pressure were controlled separately.

Firstly, the behavior of grains in polycrystalline ice thin films was investigated. It was found that some grains exhibited relatively fast grain growth on a time scale of approximately 30 min. To eliminate the effects of the grain growth, the grains that exhibited the relatively fast grain growth was excluded from further investigations of the surface melting of the polycrystalline ice thin films.

Next, the formation of QLLs in grooves of grain boundaries and on grain surfaces was investigated. Polystyrene particles (653 nm in diameter) were used as a probe for monitoring the presence of QLLs in grooves of grain boundaries. Then the author found that with increasing temperature, QLLs, whose volume was sufficient to move the polystyrene particles, preferentially appeared in grooves of grain boundaries at −1.9 ± 0.4 °C. After the appearance of the QLLs in the grooves, the QLLs continued to exist. Water vapor pressure dependency of the appearance of the QLLs in the grooves demonstrates that QLLs in grooves are formed by melting of grain boundaries to eliminate lattice mismatches between adjacent grains. In addition, the author further increased temperature, and found that droplet-type QLLs appeared on grain surfaces at −0.7 ± 0.2 °C but they disappeared within 5 ± 3 min even though the temperature and water vapor pressure were kept constant. These results suggest that droplet-type QLLs on grain surfaces are a metastable phase, as those on ice single crystals. The author proposed two plausible causes for the disappearance of the QLLs, based on faster growth of grain surfaces and changes in distribution of water vapor pressure in the vicinity of the grain surfaces. Moreover, the author found that thin-layer-type QLLs also appeared on the grain surfaces at temperatures higher than −0.7 ± 0.3 °C.

Finally, the behavior of QLLs was investigated at temperatures very close to the melting point. Then the author found that a large amount of QLLs spontaneously emerged from grain boundaries, and then the QLLs thus formed fully covered the surfaces of the polycrystalline ice thin films at −0.3± 0.1 °C, although ice grains still remained solid (unmelted) beneath the QLLs. Hence, the author concluded that it was not bulk melting. Because basal faces of ice single crystals were not fully covered with QLLs at the same temperature, it was expected that wettability of surfaces of polycrystalline ice thin films is significantly higher than that of basal faces. Interferometry observations suggest that at temperatures just below the melting point, QLLs on polycrystalline ice thin films are formed mainly by the melting of grain boundaries.

The experimental results found in this study clearly demonstrate that the behavior of QLLs on a polycrystalline ice thin film is significantly different from that on an ice single crystal. In particular, the author found that grain boundaries play a more important role in the surface melting of polycrystalline ice than grain surfaces. The author expects that the insights obtained in this study will provide a key to unlocking a wide variety of natural phenomena related to surface melting of ice.