Direct observation of elementary growth processes of ice crystals by advanced optical microscopy

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Growth and melting processes of ice crystals govern wide variety of phenomena on earth. Hence, this issue has been a subject of interest for centuries. To understand crystal growth of ice at the molecular level, one has to observe in-situ “elementary steps”, which play a key role during growth and melting processes on ice crystal surfaces. However, since observation of ice crystal surfaces by scanning probe microscopy is very difficult, so far only one group has succeeded in such observation by atomic force microscopy \cite{1}. In this study, we adopted laser confocal microscopy combined with differential interference contrast microscopy (LCM-DIM), by which elementary steps of protein crystals (3-6 nm in height) could be visualized with sufficient contrast levels \cite{2}, and tried to visualize molecular-level surface morphologies on ice crystal surfaces. We first attempted to observe the air-ice interface, since this interface has a much larger reflectivity than water-ice interfaces. By further improving LCM-DIM and growing ice crystals of higher quality, we finally succeeded in observing ice crystal surfaces grown by the two-dimensional (2D) nucleation growth mechanism. When steps of neighboring 2D islands coalesced with each other, the contrast of steps disappeared completely. Such disappearances of the step contrasts were commonly observed all over the crystal surface confirming that we succeeded in observing elementary steps (0.37 nm in height), for the first time, by optical microscopy. We also succeeded in observing surface melting processes at air-ice interfaces. We could visualize the appearances of two types of quasi-liquid layers (bulk-liquid like drops (BLD) and thin-liquid like layers (TLL)) and growing elementary steps simultaneously on the same crystal surface.