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## Temperature dependence of the growth kinetics of elementary spiral steps on ice basal faces grown from water vapor

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Ice is one of the most abundant materials on the earth. Hence, crystal growth of ice governs a wide variety of phenomena in nature. For example, most rains fallen outside tropical regions are formed by the melting of ice crystals (snowflakes) that were grown in the sky and then descended to the ground [1]. Therefore, to understand the growth kinetics of ice crystals is extremely important. Recently, we improved laser confocal microscopy combined with differential interference microscopy (LCM-DIM) [2] further, and then succeeded in visualizing individual elementary steps (0.37 nm in thickness) on ice crystal surfaces growing in supersaturated water vapor by LCM-DIM [3]. Hence, in this study, we tried to reveal the temperature dependence of the growth kinetics of elementary steps on ice basal faces.

We measured velocity  $V_{\text{step}}$  of isolated elementary spiral steps and distance  $L_{\text{eq}}$  between adjacent equivalent spiral steps on ice basal faces by LCM-DIM. We determined the step kinetic coefficient  $\beta$  from  $V_{\text{step}}$  measured under various supersaturations. We performed similar experiments under various temperatures  $T$ , and determined the temperature dependence of  $\beta$  of ice basal faces, for the first time, in the temperature range of  $-26.0$  to  $-2.7^\circ\text{C}$ . When  $-6.2 \leq T \leq -2.7^\circ\text{C}$ , the value of  $\beta$  decreased significantly with decreasing  $T$ . In contrast, when  $-15.0 \leq T \leq -6.2^\circ\text{C}$ , the value of  $\beta$  increased with decreasing  $T$ , and had the maximum at  $T \approx -15^\circ\text{C}$ . When  $-26.0 \leq T \leq -15.0^\circ\text{C}$ , the value of  $\beta$  decreased monotonically with decreasing  $T$ . Such complicated temperature dependence of  $\beta$  strongly implies the existence of unknown phenomena in the temperature range examined.

To obtain a clue to the complicated behavior of  $\beta$ , we also measured dependence of  $L_{\text{eq}}$  on surface supersaturation  $\Delta\mu^{\text{surf}}$ . When  $-9.2 \leq T \leq -3.2^\circ\text{C}$ , plots of  $L_{\text{eq}}$  vs.  $1/\Delta\mu^{\text{surf}}$  satisfactorily follow the spiral growth model. However, when  $-26.0 \leq T \leq -13.0^\circ\text{C}$ , the  $L_{\text{eq}}$  vs.  $1/\Delta\mu^{\text{surf}}$  plots do not follow any model: this temperature range agrees with the temperature range in which  $\beta$  decreased monotonically with decreasing  $T$ .

### References

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- 3) Sazaki, G.; Zepeda, S.; Nakatsubo, S.; Yokoyama, E.; Furukawa, Y., Elementary steps at the surface of ice crystals visualized by advanced optical microscopy. Proceedings of the National Academy of Sciences of the United States of America 2010, 107, (46), 19702-19707.

### Significance statement

We measured the growth kinetics of “elementary steps” on ice basal faces under various temperatures. Then we found that the temperature dependence of the growth kinetics shows a significantly complicated behavior, demonstrating that there still exist unknown phenomena in the growth of elementary steps on ice basal faces.

**Primary author:** Mr INOMATA, Masahiro (Institute of Low Temperature Science, Hokkaido University)

**Co-authors:** Prof. SAZAKI, Gen (Institute of Low Temperature Science, Hokkaido University); Dr ASAKAWA, Harutoshi (Graduate School of Sciences and Technology for Innovation, Yamaguchi University); Dr NAGASHIMA, Ken (Institute of Low Temperature Science, Hokkaido University, Japan); Dr MURATA, Ken-ichiro (Institute of Low Temperature Science, Hokkaido University); Mr NAKATSUBO, Shunichi (Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency); Prof. FURUKAWA, Yoshinori (Institute of Low Temperature Science, Hokkaido University)

**Presenter:** Mr INOMATA, Masahiro (Institute of Low Temperature Science, Hokkaido University)

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