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Growth kinetics of elementary spiral steps on ice prism faces grown in vapor and their temperature dependence

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References

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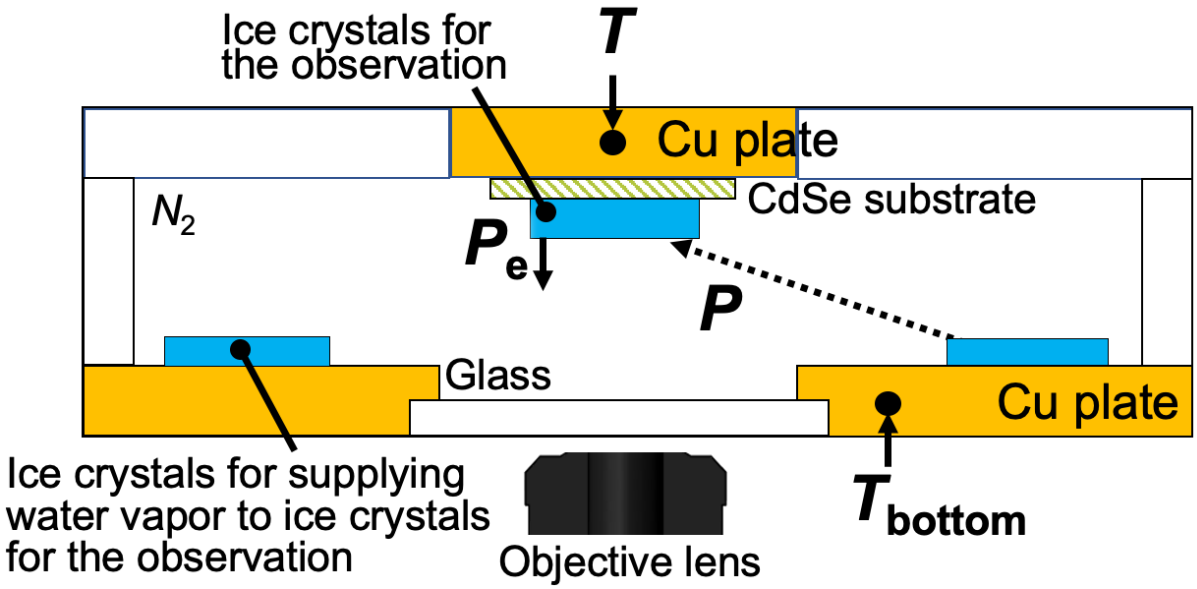

Fig. S1. A schematic drawing of an observation chamber. The observation chamber was composed of upper and lower Cu plates, whose respective temperatures T and T_{bottom} were separately controlled using Peltier elements. At the center of the upper Cu plate, a $\{10\bar{1}0\}$ CdSe substrate crystal was attached using heat grease. Ice crystals for the LCM-DIM observation were grown heteroepitaxially on the CdSe crystal. Ice crystals for supplying water vapor to the ice crystals for the observation were grown on the lower Cu plates. The total pressure in the observation chamber was kept at 0.1 MPa (atmospheric pressure) by filling the chamber with pure nitrogen gas. The volume of the ice crystals for supplying water vapor was significantly larger than that of the ice crystals for the observation. Hence, partial pressure of water vapor P in the observation chamber was determined by T_{bottom}. In addition, equilibrium partial vapor pressure P_{e} of the ice crystals for the observation was determined by T. By separately controlling T and T_{bottom}, we can control the growth temperature T of the ice crystals for the observation and the supersaturation $\sigma = (P - P_{\mathrm{e}})/P_{\mathrm{e}}$ in the observation chamber separately.

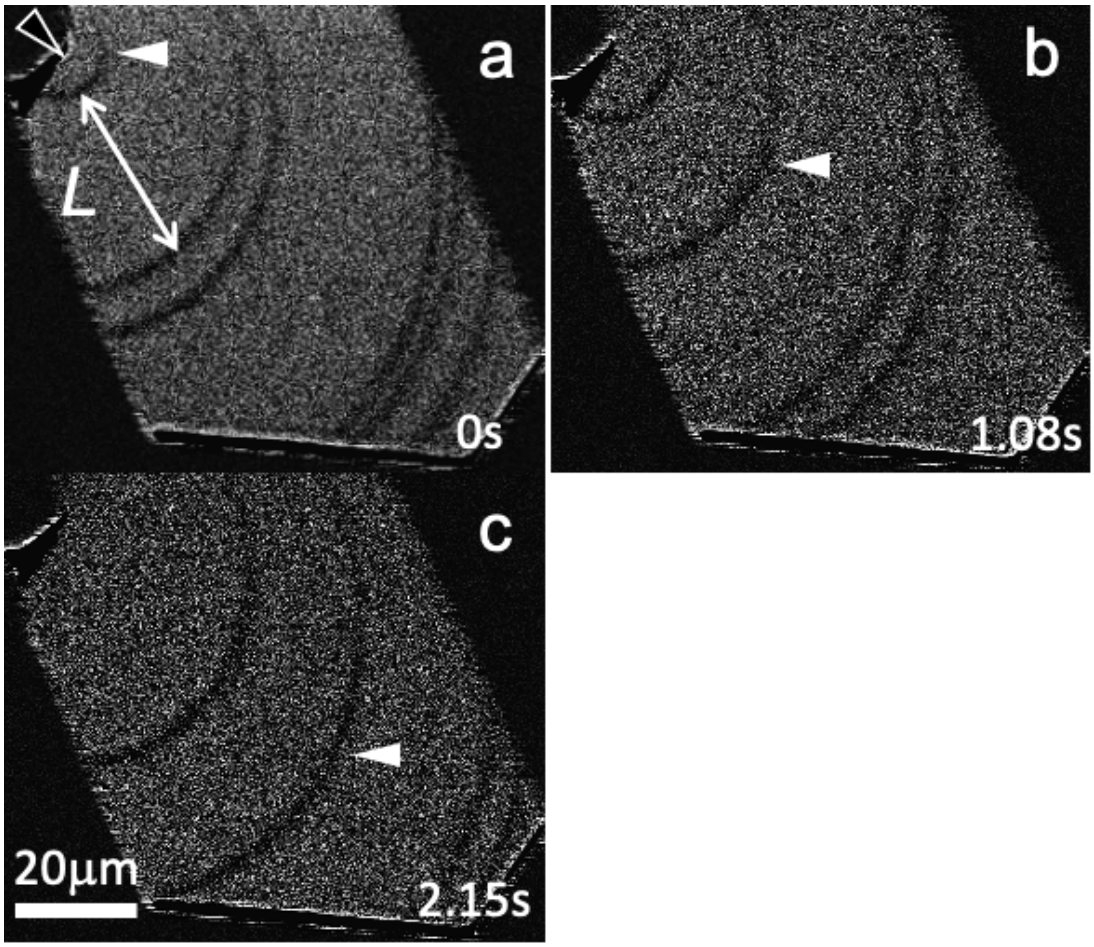

Fig. S2. LCM-DIM images of elementary spiral steps grown on an ice basal face at T=-15.0 °C and σ= 0.18: (a) 0 s, (b) 1.08 s, and (c) 2.15 s. A black arrowhead presents the location of a screw dislocation. On basal faces, screw dislocations are always located at the edges of ice crystals.[1, 2] In addition, distance L between adjacent spiral steps on basal faces exhibits a significantly large variation (also see Fig. S5b).[1, 2] The velocity V_{step} of elementary spiral steps was determined from the time evolution of the position of an identical step (marked by white arrowheads). Reprinted with permission from a reference.[2] Copyright 2018 American Chemical Society.

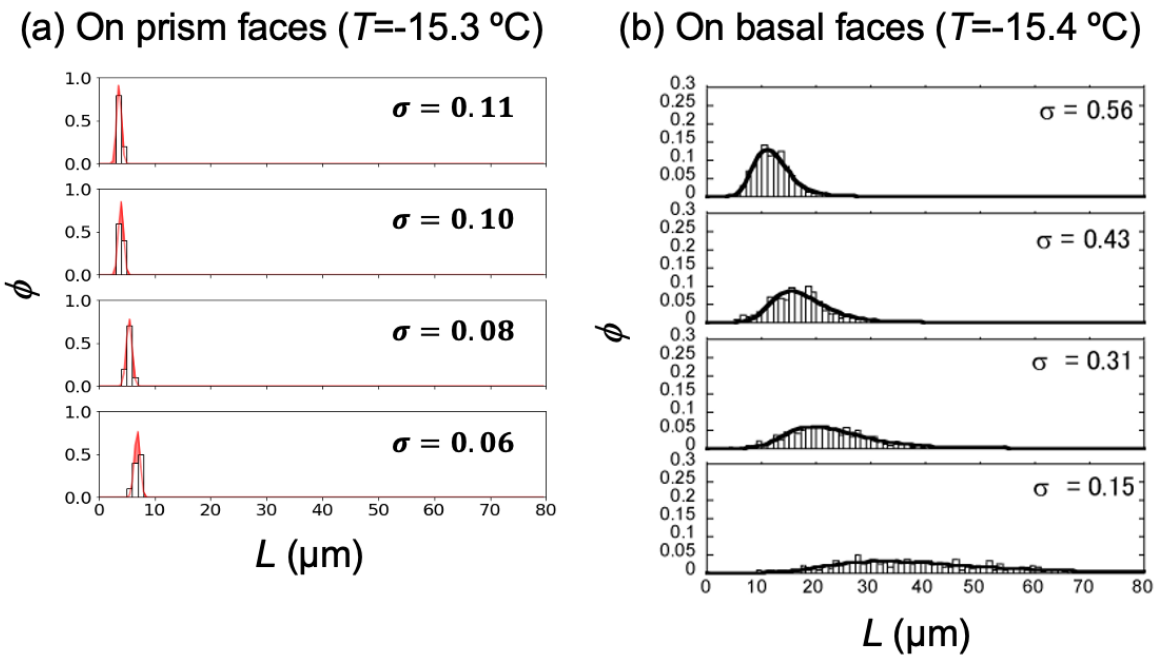

Fig. S3. Distribution of distance *L* between adjacent elementary spiral steps. (a) On prism faces measured in this study, and (b) on basal faces reported in our previous study. The panel (b) is reprinted with permission from a reference.[2] Copyright 2018 American Chemical Society.

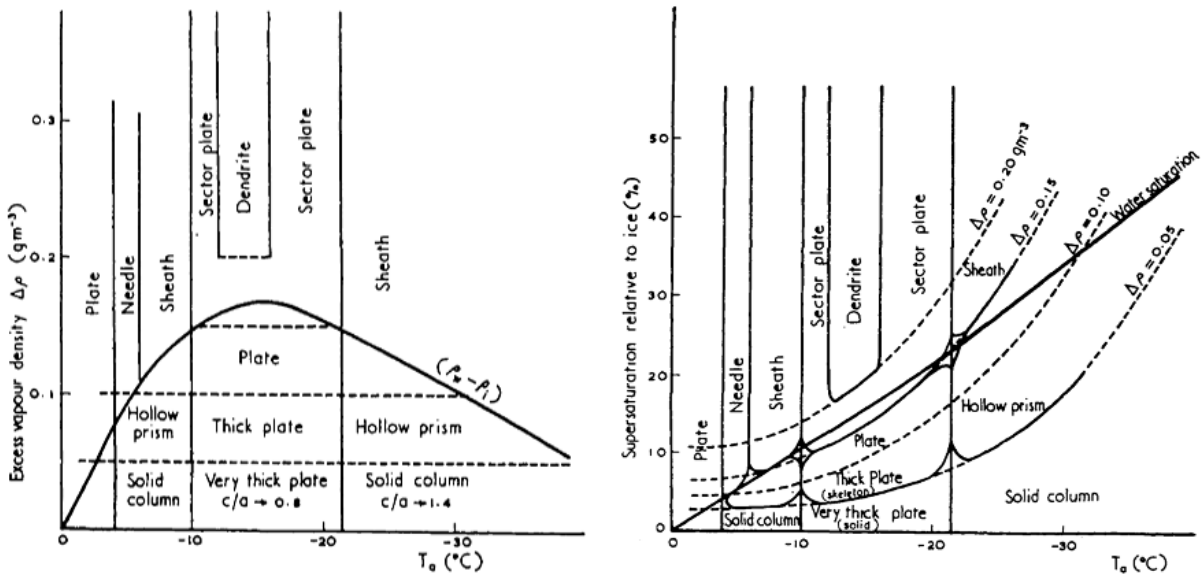

Fig. S4. Changes in the morphology of snow crystals as functions of water vapor density and temperature.[3, 4] With increasing water vapor density, a hexagonal crystal (hexagonal plate/column) is changed into a dendritic crystal with a hexagonal symmetry. This morphological change could be successfully explained by the diffusion-limited growth that occurs when the supply of water vapor by volume diffusion is much slower than the surface incorporation of water admolecules.[5, 6] In addition, with decreasing temperature, a snow crystal repeatedly shows a hexagonal-plate shape (0 to -4 °C), a hexagonal-column shape (-4 to -10 °C), a hexagonal-plate shape (-10 to -22 °C), and a hexagonal-column shape (< -22 °C). Kuroda and Lacmann tried to explain such morphological changes with temperature utilizing the differences in temperatures of the thermal roughening transition and surface melting on basal and prism faces.[7] However, after the study by Kuroda and Lacmann, Elbaum and coworkers found that basal faces never show the roughening transition even just below 0 °C, although prism faces exhibit the roughening transition at temperatures higher than -2 °C.[8] In addition, our recent studies revealed that quasi-liquid layers with droplet and thin-layer shapes appear on basal and prism faces at temperatures only higher than -2 °C.[9-12] Therefore, the study on the mechanism of the morphological changes with temperature had returned to the starting point. Reprinted with permission from a reference.[4] Copyright 1961 Taylor & Francis Group.

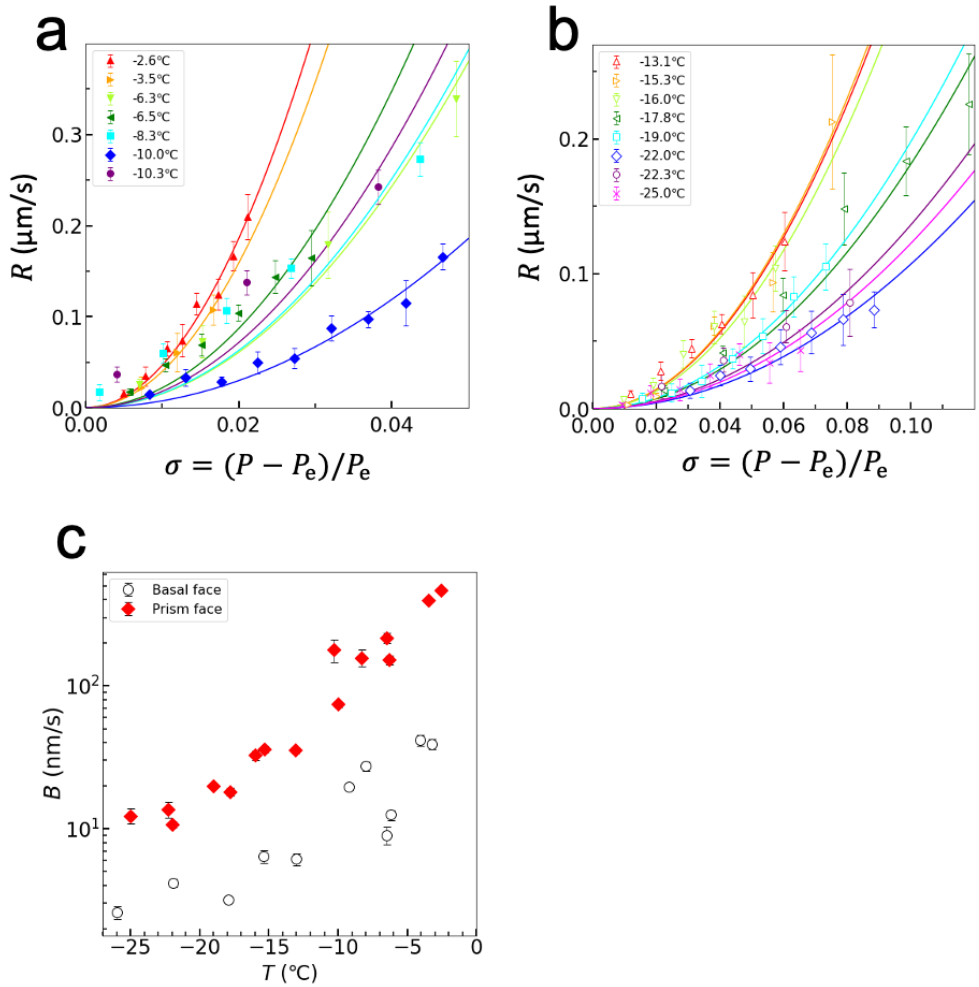

Fig. S5. Temperature dependence of the growth kinetics of normal growth rate R. (a) and (b) Changes in R as a function of σ under various T from -25.0 to -2.6 °C. (c) Changes in a proportional constant B as a function of T. Filled and open plots show the constant B of prism faces (this study) and basal faces (previous study[2]), respectively.

In addition to the determination of the temperature dependence of β, we also tried to evaluate the normal growth rate R at various σ and T, using the following relation:[5,6]

$$R = \frac{h}{L} V_{\text{step}}. \tag{S1}$$

Here, h is the height of elementary steps ($h = 0.39$ nm on prism faces, and 0.37nm on basal faces).[13] Because L is a distance between adjacent spiral steps, h/L shows the slope of a spiral growth hillock. The panels (a) and (b) shows the R vs. σ plots at various T. When spiral growth follows eq. (3) and σ is sufficiently small, it is well known that the R vs σ plot shows the following relation:[5,6]

$$R = B\sigma^2. \tag{S2}$$

Here, B is a proportional constant and hence shows the growth kinetics at a certain T (a kinetic coefficient of R). The panel (c) shows the changes in B determined from the panels (a) and (b) as a function of T. In this T range, the values of B of prism faces are significantly larger than those of basal faces, irrespective of T. Therefore, we could not explain the morphological changes of ice crystals with T (Fig. S3) even considering R. For the plausible reasons, see the main text.

Video S1. Time evolution of the lateral growth of an elementary spiral step on an ice prism face at T = -6.3 °C and σ = 0.02. A part of this video is shown in Fig. 2. The spiral step on the prism face was observed by LCM-DIM. Photomicrographs were acquired over a 5.5-s time interval.

Table S1 Summary of previous experimental studies on the difference between the growth of basal and prism faces

Author	Year	Pressure	Excess water vapor	Maximum temperature °C (σ [a])	Minimum temperature °C (σ [a])	Parameters measured
Hallett[14]	1961	1 atm	0.25 g/m^3	0 (0.05)	-40 (2.10)	Macrostep V [b]
Mason, *et al.*[15]	1963	1 atm	Adjusted	0 (0.5)	-40 (0.5)	Surface diffusion distance
Kobayashi[16]	1967	1 atm	0.21 g/m^3	0 (0.04)	-50 (5.5)	Macrostep V [b]
Lamb and Hobbs[17]	1971	R.P.[c]	1.3 Pa	0 (0.002)	-17 (0.009)	Face R [d]
Lamb and Scott[18]	1972	R.P.[c]	1.3 Pa	0 (0.002)	-17 (0.009)	Face R [d]
This study	2022	1 atm	Adjusted	-2.6 (0 ~ 0.02)	-25 (0 ~ 0.09)	Elementary step V [e]

[a] Supersaturation; [b] lateral velocity of macrosteps; [c] a vacuum formed by a rotary pump; [d] normal growth rate of a face; [e] lateral velocity of elementary steps

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