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GROWTH FORM OF ICE CRYSTALS GROWN IN AIR AT LOW SUPERSATURATION AND THEIR GROWTH MECHANISM

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Abstract: The morphological instability and the growth mechanism of ice crystals grown in air at $-30°C$ and at supersaturation below 4% have been experimentally studied. Whether ice crystals grown under this condition would develop into long prismatic columns or into thin plates is dependent on the emergence of active screw dislocations on the $\{0001\}$ or $\{10\bar{1}0\}$ faces of the crystals. The morphological instability of ice crystals grown in air at low supersaturation is related to the emergence of active screw dislocations near the corners of the $\{0001\}$ or $\{10\bar{1}0\}$ faces. From the experimental results, the growth form and the growth mechanism of snow crystals at low supersaturation observed at Mizuho Station, Antarctica are discussed.

1. Introduction

Snow crystals precipitating at Mizuho Station, Antarctica are generally formed under the low absolute humidity. However, in Antarctica where the temperature is low, high or low supersaturation will be produced by a small variation of water vapor contents even under nearly constant temperature. Paying attention to single crystals, we have found by the observations at Mizuho Station (WADA and GONDA, 1985) that long hollow-prisms (long prismatic columns with large skeletal structures) precipitate with high frequency under about 10% supersaturation.

On the other hand, in the polar regions, long solid prisms (SHIMIZU, 1963), rectangular snow crystals (HIGUCHI, 1968), diamond-dust type ice crystals (KIKUCHI and HOGAN, 1979) and long prisms with skeletal structures which enclose air bubbles inside the crystals (KLINOV, 1960), etc. have been observed under the circumstance where the supersaturation is estimated to be small. Thereafter, the growth form and the growth mechanism of ice crystals have been experimentally studied in order to understand those of snow crystals observed in the polar regions (KOBAYASHI, 1965; GONDA and KOIKE, 1982; GONDA *et al.*, 1984; GONDA and GOMI, 1985; GONDA *et al.*, 1985). It has been found by these studies (*e.g.* GONDA *et al.*, 1985) that ice crystals growing at the supersaturation above about 10% grow by a two-dimensional nucleation mechanism, while those at the supersaturation below a few % grow by a screw dislocation mechanism.

The purposes of this article are to study experimentally the morphological instability and the growth mechanism of ice crystals growing in air at low temperature and low supersaturation, and from the obtained results, to discuss the growth form and the growth mechanism of snow crystals under low supersaturation observed at Mizuho Station, Antarctica.

2. Experimental

The structure of a growth chamber, the cooling and the temperature control of it, the supply of water vapor and its measurement, the supply of ice nuclei, etc. have been described in other papers (Kuroda and Gonda, 1984; Gonda *et al.*, 1984), so, further explanations related to the experimental methods are omitted in this paper.

To study the morphological instability of ice crystals growing at low temperature and low supersaturation, ice crystals were formed in air at 3.3×10^4 and 1.0×10^5 Pa at $-30°C$ and supersaturation below about 4%, and growing ice crystals were observed *in situ* using a differential interference microscope attached to a single-lens reflex camera. Next, the normal growth rates of a polyhedral ice crystal were measured in air at 4.0×10 Pa at $-30°C$ and supersaturation below 3%. To keep the physical properties of the crystal as constant as possible, the normal growth rates of the same crystal were measured as a function of supersaturation. At the same time, the surface microtopography of the crystals was observed using a differential interference microscope attached to a TV camera.

3. Experimental Results

3.1. Morphological instability of ice crystals grown in air at −30°C and low supersaturation

Figure 1 shows an example of a columnar ice crystal grown in air at 1.0×10^5 Pa at $-30°C$ and 1.4% supersaturation. At 12.0 (b) and 39.3 (c) min after the growth stage (a), the crystal grew preferentially to the ⟨0001⟩ direction and a polyhedral columnar crystal was formed. From the observations of surface microtopography of many ice crystals, it is inferred that the growth of this crystal is related to the emergence of active screw dislocations near a center of the {0001} face. After 50.1 min (d), the {0001} face of the crystal became unstable and a long prismatic column with small skeletal structures was formed. At 84.3 min (e), after the initial skeletal structures on the {0001} face had been enclosed inside the crystal as the air bubbles, the secondary skeletal structures were formed on the {0001} face. At 97.3 min (f), the {0001} face became stable again and the polyhedral crystal was finally formed. The morphological instability under low supersaturation described above, in the later stage of the growth (d, e), is related to the emergence of active screw dislocations near the corners of the {0001} face (Gonda *et al.*, 1985). In addition, in the growth stages (e) and (f), the non-hexagonal columnar crystal was formed by the emergence of active screw dislocations on the {10$\bar{1}$0} face of the crystal.

Figure 2 shows an example of an ice crystal grown in air at 1.0×10^5 Pa at $-30°C$ and 0.8% supersaturation, whose size ratio c/a is close to unity. From the observations of the surface microtopography of many ice crystals at low supersaturation, it is inferred that both the {0001} and {10$\bar{1}$0} faces of the crystal grew by screw dislocations which emerged on the both faces. This crystal was photographed from the direction

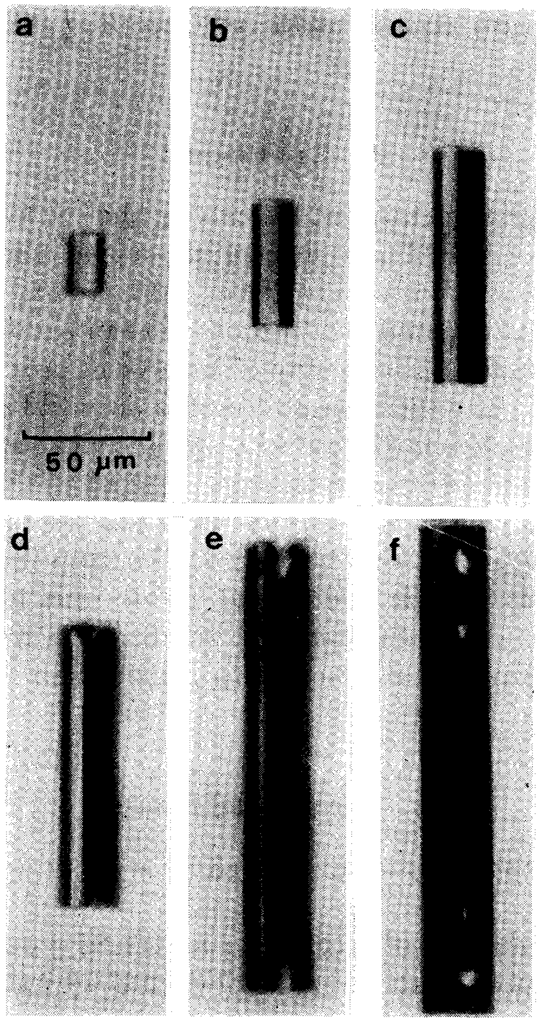

Fig. 1. Columnar ice crystal grown in air at 1.0×10^5 Pa at $-30°C$ and 1.4% supersaturation. (a) 0, (b) 12.0, (c) 39.3, (d) 50.1, (e) 84.3, (f) 97.3 min.

along *a*-axis. From the surface microtopography, it is inferred that active screw dislocations emerge near the corners of the $\{10\bar{1}0\}$ face of the right-hand side of the crystal. That is, the macroscopic steps advance from the outside to a center of the $\{10\bar{1}0\}$ face and the shallow concave was formed on the $\{10\bar{1}0\}$ face (c). After that, the steps further moved towards the center of the $\{10\bar{1}0\}$ face and a relatively deep concave was formed on the $\{10\bar{1}0\}$ face by the bunching of the growth steps (d, e). Moreover, the concave became gradually small and the $\{10\bar{1}0\}$ face became the flat surface (f) again. In the progressive process of the growth steps, the concave is captured into the crystal as the air bubbles when the concave on the $\{10\bar{1}0\}$ face became comparatively deep (GONDA and KOIKE, 1982).

Figure 3 shows the instability limit of the $\{10\bar{1}0\}$ face of columnar ice crystals grown in air at 3.3×10^4 Pa at $-30°C$ and relatively low supersaturation. The morphological instability of the ice crystals under low supersaturation was produced by active screw dislocations which emerged near the corners of the $\{0001\}$ face (GONDA *et al.*, 1985). As shown in Fig. 1, the morphological instability at low supersaturation produced by screw dislocations can be repeated such as the stable growth to the un-

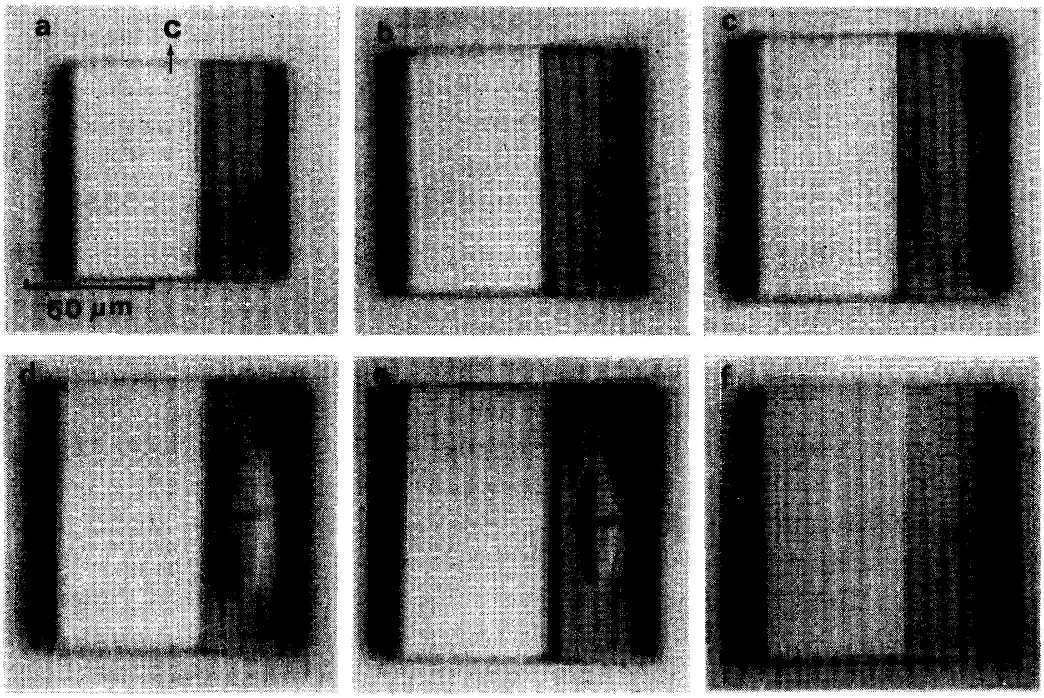

Fig. 2. Ice crystal grown in air at 1.0×10^5 Pa at $-30°C$ and 0.8% supersaturation, whose size ratio c/a is close to unity. (a) 0, (b) 44.7, (c) 74.6, (d) 89.6, (e) 96.6, (f) 100 min.

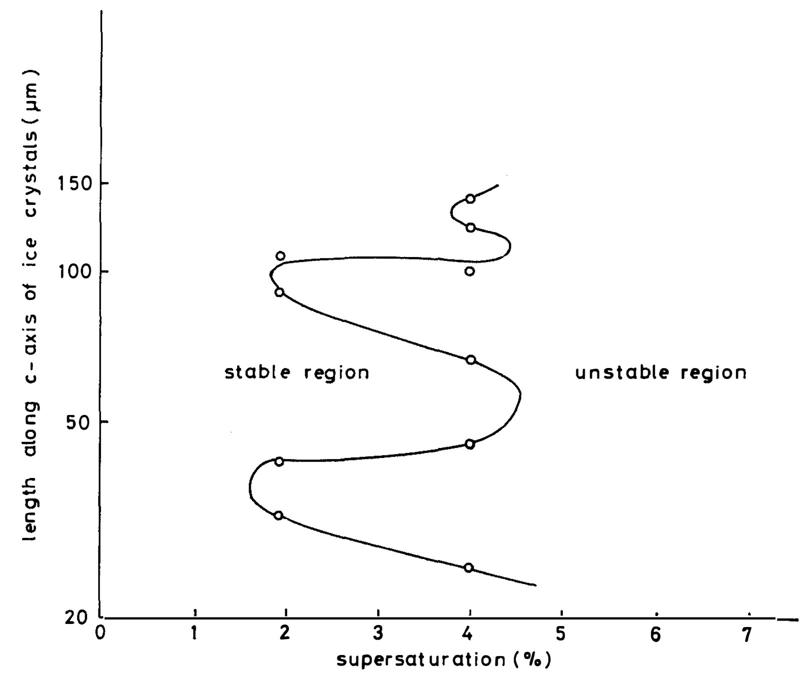

Fig. 3. Instability limit of the $\{10\bar{1}0\}$ face of columnar ice crystals grown in air at 3.3×10^4 Pa at $-30°C$.

stable growth or *vice versa*. For example, as shown in Fig. 3, the morphological instability of the $\{10\bar{1}0\}$ face under about 2% supersaturation is repeated twice with increasing crystal size. Furthermore, when ice crystals grow over a long period of time, the morphological instability of the crystals is repeated many times. That is, the morphological instability produced by screw dislocations is drastically different from that produced by two-dimensional nuclei, which occurs only one time. On the other hand, the morphological instability of ice crystals depends also on the ratio of growth rates R_b/R_p and the size ratio c/a of ice crystals (GONDA and GOMI, 1985). Here, R_b and R_p show the growth rates of the {0001} and {10$\bar{1}$0} faces of ice crystals, respectively.

3.2. The growth rate and the surface microtopography of ice crystals grown in low air pressure at −30°C and low supersaturation

Figure 4 shows the supersaturation dependence of the normal growth rates of the {0001} and {10$\bar{1}$0} faces of a columnar ice crystal grown in air pressure of 4.0×10 Pa at −30°C and low supersaturation. In this growth condition, the resistance of the volume diffusion of water vapor can be ignored. As shown in the figure, the normal growth rates of the {0001} and {10$\bar{1}$0} faces of the crystal quadratically increase with increasing supersaturation in the region of supersaturation below about 1.5% and then linearly increase with increasing supersaturation in the region of supersaturation between 1.5 and 3%. These experimental results coincide with the theoretical consideration by BURTON *et al.* (1951). Therefore, it is considered that both the {0001} and {10$\bar{1}$0} faces of the crystal grow by a screw dislocation mechanism.

Figure 5 shows the video photographs of the {0001} and {10$\bar{1}$0} faces of ice crystals grown in air pressure of 4.0×10 Pa at −30°C and low supersaturation. Here, the upper crystal is that grown at 1.8% supersaturation while the lower crystal is that grown at 1.4% supersaturation. In the figure, circular white parts are the cavities introduced between ice crystals and a growth substrate. As shown in the figure, an

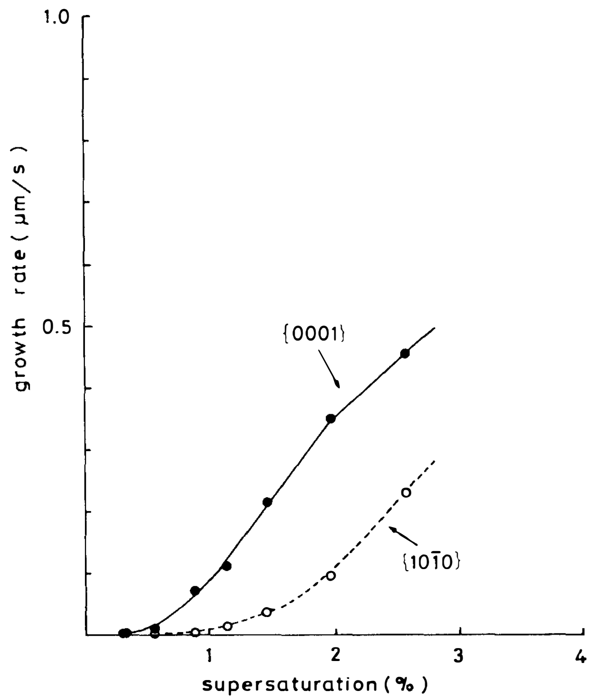

Fig. 4. Supersaturation dependence of the normal growth rates of the {0001} and {10$\bar{1}$0} faces of a columnar ice crystal grown in air at 4.0×10 Pa at −30°C.

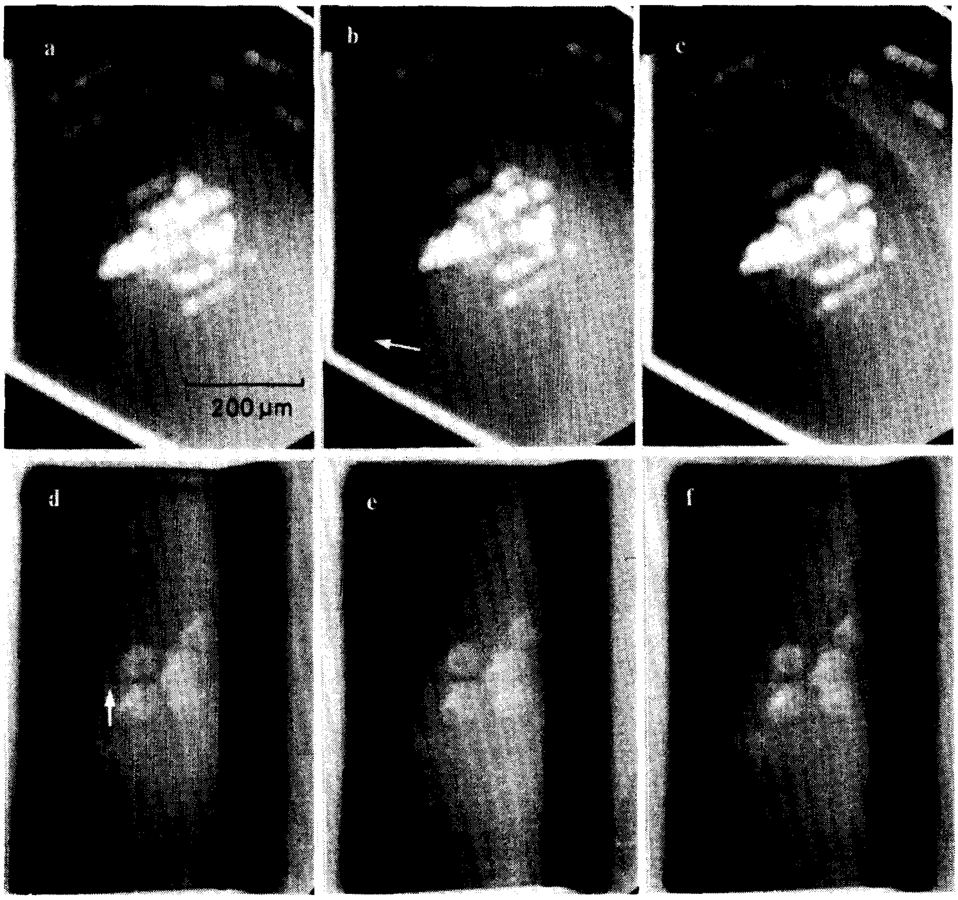

Fig. 5. Surface microtopography of the {0001} and {10$\bar{1}$0} faces of ice crystals grown in air at 4.0× 10 Pa at −30°C. The upper and lower crystals show the {0001} and {10$\bar{1}$0} faces, respectively. (a) 0, (b) 5, (c) 10 s; (d) 0, (e) 2, (f) 4 s.

active screw dislocation emerges near a corner of the {0001} face (arrow ↑), and the growth layers advance from a center of the screw dislocation. On the other hand, active screw dislocations emerge on a defect running perpendicularly to the *c*-axis on the {10$\bar{1}$0} face (arrow ↑), which may be stacking faults proposed by KOBAYASHI and OHTAKE (1974), and the growth layers move from the centers of screw dislocations to the outside of the crystal. It is concluded from the experimental results that ice crystals grown at −30°C and low supersaturation grow by a screw dislocation mechanism.

4. Discussion

The morphological instability and the growth mechanism of ice crystals grown in air at low temperature and low supersaturation have been experimentally studied. From the experimental results, the growth form and the growth mechanism of snow crystals observed at low supersaturation in Antarctica are discussed. The morphological instability of ice crystals grown at high supersaturation has been produced only once in the whole stage of growth (GONDA and KOIKE, 1982, 1983; GONDA and YAMAZAKI, 1982; GONDA and GOMI, 1985; GONDA *et al.*, 1984). However, the morphological instability of ice crystals grown at low supersaturation is repeated with increasing crystal size such as the stable growth to the unstable growth or *vice versa* (Figs. 1 and 3). In the case of high supersaturation, large skeletal structures have been formed by two-dimensional nuclei at the corners of the crystals (GONDA *et al.*, 1985; WADA and GONDA, 1985). In the case of low supersaturation, small skeletal structures have been formed by the screw dislocations which emerged near the corners of the {0001} face; this has been confirmed also by the surface observations of ice crystals grown in air at 1.0×10^5 Pa (GONDA *et al.*, 1985).

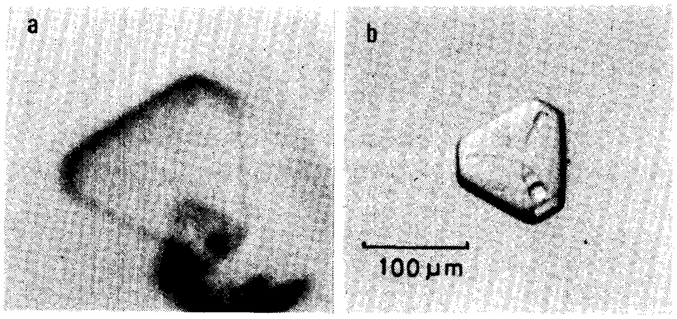

Fig. 6. Snow crystals observed at Mizuho Station, Antarctica. Air temperature at a height of 1.5 m above the snow surface is (a) −19.1 and (b) −22.6°C.

Figure 6 shows an example of snow crystals observed at Mizuho Station, Antarctica on (a) January 5, 1980 and (b) March 19, 1979. Let us discuss the growth mechanism of snow crystals observed at Mizuho Station from the experimental results described above and the other experiment (GONDA and SEI, 1986). GONDA and SEI (1986) found that the {0001} and $\{10\bar{1}0\}$ faces of an ice crystal growing at −7, −15 and −30°C and at a supersaturation below 5% grew by a screw dislocation mechanism. Therefore, it is considered that both the {0001} and $\{10\bar{1}0\}$ faces of ice crystals growing at a temperature between −7 and −30°C and at supersaturation below 5% grow by a screw dislocation mechanism. Accordingly, it is inferred that the non-hexagonal polyhedral snow crystal (a) was formed by the emergence of active screw dislocations near the centers of alternate $\{10\bar{1}0\}$ faces of the crystal under low supersaturation. On the other hand, it is inferred that the non-hexagonal snow crystal with small skeletal structure (b) was formed by the emergence of active screw dislocations near the corners of alternate $\{10\bar{1}0\}$ faces of the crystal under low supersaturation. Furthermore, it is understood by careful observations of the snow crystal that the morphological instability of the $\{10\bar{1}0\}$ face was produced twice or more. Figure 6 shows the characteristics of single snow crystals formed in Antarctica under low supersaturation.

5. Conclusions

The morphological instability and the growth mechanism of ice crystals grown in air at low temperature and low supersaturation have been experimentally studied. The growth form and growth mechanism of snow crystals at low supersaturation observed at Mizuho Station, Antarctica have been discussed from the experimental results. The obtained results and the conclusions are as follows.

(1) Whether ice crystals grown under the condition described above grow as long

prismatic columns or as thin plates depends on the emergence of active screw dislocations on the {0001} or {10$\bar{1}$0} faces of the crystals.

(2) Non-hexagonal ice crystals grown in air at low supersaturation are formed when active screw dislocations emerge on some {10$\bar{1}$0} faces of the crystals.

(3) Skeletal ice crystals (morphological instability) grown in air at low supersaturation are formed when active screw dislocations emerge near the corners of the {0001} or {10$\bar{1}$0} faces.

(4) The growth form and the growth mechanism of snow crystals observed in Antarctica (Fig. 6) can be interpreted by the experimental results described above and the other experiment (GONDA and SEI, 1986).

References

BURTON, W. K., CABRERA, N. and FRANK, F. C. (1951): The growth of crystals and structure of their surfaces. Philos. Trans. R. Soc. London, Ser. A, **243**, 299–358.

GONDA, T. and GOMI, H. (1985): Morphological instability of polyhedral ice crystals growing in air at a low temperature. Ann. Glaciol., **6**, 222–224.

GONDA, T. and KOIKE, T. (1982): Morphology of single snow crystals growing in air at low temperatures. Mem. Natl Inst. Polar Res., Spec. Issue, **24**, 148–156.

GONDA, T. and KOIKE, T. (1983): Growth mechanisms of single ice crystals growing at a low temperature and their morphological stability. J. Cryst. Growth, **65**, 36–42.

GONDA, T. and SEI, T. (1986): The growth mechanism of ice crystals grown in air at a low pressure and their habit change with temperature. 7th Symp. Phys. Chem. Ice in Grenoble, Sept. (in press).

GONDA, T. and YAMAZAKI, T. (1982): Morphological stability of polyhedral ice crystals grown from the vapor phase. J. Cryst. Growth, **60**, 259–263.

GONDA, T., SEI, T. and GOMI, H. (1984): Growth forms and growth mechanisms of single snow crystals growing at a low temperature. Mem. Natl Inst. Polar Res., Spec. Issue, **34**, 87–95.

GONDA, T., SEI, T. and GOMI, H. (1985): Surface micromorphology of columnar ice crystals growing in air at high and low supersaturations. Mem. Natl Inst. Polar Res., Spec. Issue, **39**, 108–116.

HIGUCHI, K. (1968): Kyokuchi ni furu yuki (Snow crystal forming in polar regions). Shizen (Nature), **23**(8), 38–46.

KIKUCHI, K. and HOGAN, A. W. (1979): Properties of diamond dust type ice crystals observed in summer season at Amundsen-Scott South Pole Station, Antarctica. J. Meteorol. Soc. Jpn., **57**, 180–190.

KLINOV, F. JA. (1960): Voda v Atmosfere pri Nizkikh Temperaturakh. Moskva, Izd. Akademii Nauk SSSR, 170 p.

KOBAYASHI, T. (1965): Vapor growth of ice crystal between −40 and −90°C. J. Meteorol. Soc. Jpn., **43**, 359–367.

KOBAYASHI, T. and OHTAKE, T. (1974): Hexagonal twin prisms of ice. J. Atmos. Sci., **31**, 1377–1383.

KURODA, T. and GONDA, T. (1984): Rate determining processes of growth of ice crystals from the vapour phase. Part II: Investigation of surface kinetic process. J. Meteorol. Soc. Jpn., **62**, 563–572.

SHIMIZU, H. (1963): "Long prism" crystals observed in precipitation in Antarctica. J. Meteorol. Soc. Jpn., **41**, 305–307.

WADA, M. and GONDA, T. (1985): Snow crystals of hollow-prism type observed at Mizuho Station, Antarctica. Nankyoku Shiryô (Antarct. Rec.), **86**, 1–8.

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