GROWTH FORMS AND GROWTH MECHANISMS OF SINGLE SNOW CRYSTALS GROWING AT A LOW TEMPERATURE

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Abstract: Single ice crystals have been grown in air at various constant pressures at $-30°C$ and various constant supersaturations, and measurements of normal growth rate and *in situ* observations of the surface micromorphology of ice crystals have been made. As a result, it has been found that the habit and the morphological instability of ice crystals grown at $-30°C$ vary markedly not only with supersaturation and crystal size but also with air pressure. On the basis of this study, it is considered that many snow crystals formed in polar regions at a supersaturation below about 2% grow by a screw mechanism, while at a supersaturation about above 10% they grow by a nucleation mechanism.

1. Introduction

It has become clear from theoretical studies (KURODA and LACMANN, 1982; KURODA, 1982) and experimental studies (GONDA, 1977, 1980; GONDA and KOIKE, 1982a, b) that the habit of small ice crystals grown in air at 1.0×10^5 Pa at a temperature below $-22°C$ depends not only on temperature but also on supersaturation. Moreover, it has been pointed out that the habit of ice crystals depends also on crystal size and the external form and internal structure of minute ice crystals below 1–2 μm (GONDA and KOIKE, 1982b).

In order to study the mechanism of the habit change of ice crystals, we must form polyhedral ice crystals in air at low pressure where the surface kinetic process for incorporation of water molecules into the crystal lattice is the rate determining process, and it is necessary to measure the supersaturation dependence of the normal growth rate of the {0001} and {10$\bar{1}$0} faces of ice crystals (GONDA and KOIKE, 1983); in addition, we must observe the surface micromorphology of growing ice crystals. Moreover, we should form ice crystals in air at various constant pressures and should study the effects of air pressure on the normal growth rate of each face of ice crystals.

The purpose of this study is to infer the growth forms and growth mechanisms of single snow crystals forming in the polar regions, on the basis of studies of the air pressure and supersaturation dependences of the growth forms of ice crystals growing at $-30°C$ and observations of the surface micromorphology of growing ice crystals at $-30°C$.

2. Experimental Procedures

A cold chamber for *in situ* observation of ice crystals growing at $-30°C$ has been described in a previous paper (KURODA and GONDA, 1984). The cold chamber is designed to independently cool an ice plate for supplying water vapor and a substrate for ice crystal growth. The chamber is cooled by flowing an electric current to the thermoelectric cooling panels for the water vapor supply and for the growth substrate, keeping the ice plate at a temperature lower than that of the growth substrate. The temperature on the ice plate and on the growth substrate is accurate to $\pm 0.05°C$. Temperature control is carried out by automatically turning on and off the electric current flowing to the thermoelectric cooling panel for ice plate (2A) and that for growth substrate (5A), using the temperature regulators.

Thereafter, the air in the chamber is evacuated for about 10 min using a vaccum pump until the air pressure reaches about 4.0×10 Pa. When about 3 cm^3 of sufficiently diluted silver iodide smoke is inserted into the chamber in order to nucleate ice crystals, keeping the ice plate at a temperature slightly higher than that on the growth substrate, minute ice crystals are nucleated in air at low pressure at a constant supersaturation and fall on the growth substrate. Then, the residual air in the chamber is evacuated for about 1 min for the growth of ice crystals under as lower pressure as possible, and photomicrographs of ice crystals are taken. Ice crystals were grown in air at 4.0×10, 3.3×10^4 and 1.0×10^5 Pa at $-30°C$ and under various constant supersaturations.

3. Experimental Results

3.1. Habit and the morphological instability of ice crystals growing at −30°C

Figure 1 shows a long solid prism grown predominantly in air at 4.0×10 Pa at $-30°C$ and a supersaturation of 0.7%. In air at 4.0×10 Pa, the (0001) and $(10\bar{1}0)$ faces of ice crystals are stable. At a supersaturation of 0.7%, long solid prisms which grow preferentially in the $\langle 0001 \rangle$ direction are formed even in air at 1.0×10^5 Pa (GONDA and KOIKE, 1982a).

Figure 2 shows a polyhedral plate-like ice crystal grown predominantly in air at 4.0×10 Pa at $-30°C$ and a supersaturation of 8.8%. As seen in Figs. 1 and 2, and the statistical data (GONDA and KOIKE, 1983), when the supersaturation increases under constant temperature and air pressure, columnar ice crystals are transformed to plate-like ice crystals with increasing supersaturation. That is to say, at this growth temperature, the habit of ice crystals depends on the supersaturation.

Figure 3 shows a columnar ice crystal with skeletal structure grown predominantly in air at 1.0×10^5 Pa at $-30°C$ and a supersaturation of 8.8%. Under this growth condition, polyhedral ice crystals grow only when the crystal size is below about 20 μm or above about 330 μm.

Figure 4 shows the air pressure dependence of the instability of the (0001) face of 80 μm columnar ice crystals grown at $-30°C$ and a supersaturation of 8.8%. As seen from Figs. 2–4 when air pressure varies from 4.0×10 to 1.0×10^5 Pa under constant temperature and supersaturation, the habit of ice crystals varies from

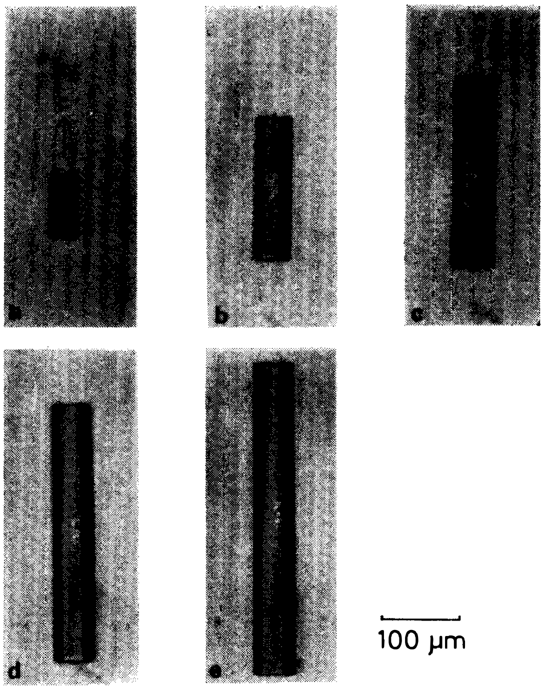

Fig. 1. Long solid prism grown in air at 4.0×10 Pa at −30°C and a supersaturation of 0.7%: (a) 1.4, (b) 3.5, (c) 5.0, (d) 7.0, (e) 10.2 min.

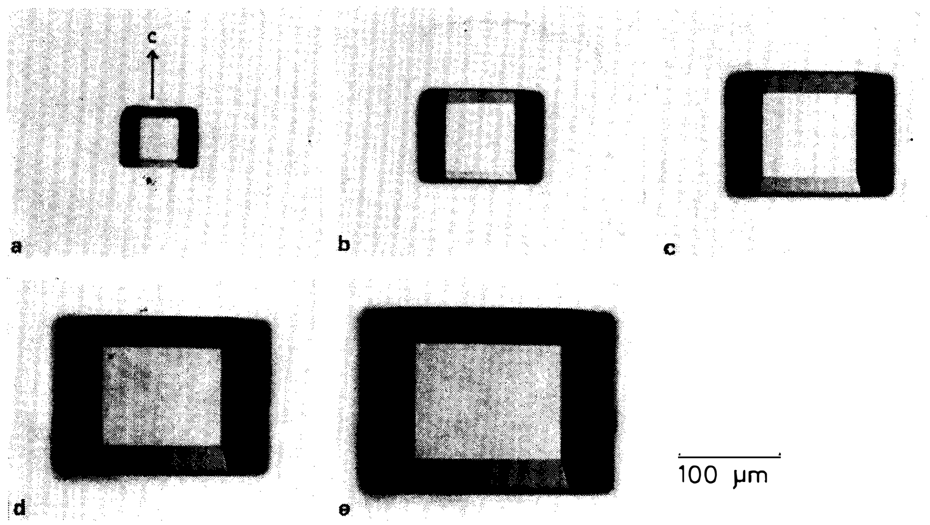

Fig. 2. Plate-like ice crystal grown in air at 4.0×10 Pa at −30°C and a supersaturation of 8.8%: (a) 0.4, (b) 0.8, (c) 1.2, (d) 1.7, (e) 2.3 min.

plate-like to column-like (Fig. 5); at the same time polyhedral ice crystals are transformed to skeletal crystals. This means that the habit and morphological instability of ice crystals depend markedly on air pressure.

Figure 5 shows the air pressure and supersaturation dependences of the ratio of axial lengths c/a of 80 μm ice crystals grown in air at 4.0×10, 3.3×10^4 and 1.0×10^5 Pa at -30°C. Each point is a mean value of about 10 ice crystals. As seen in the

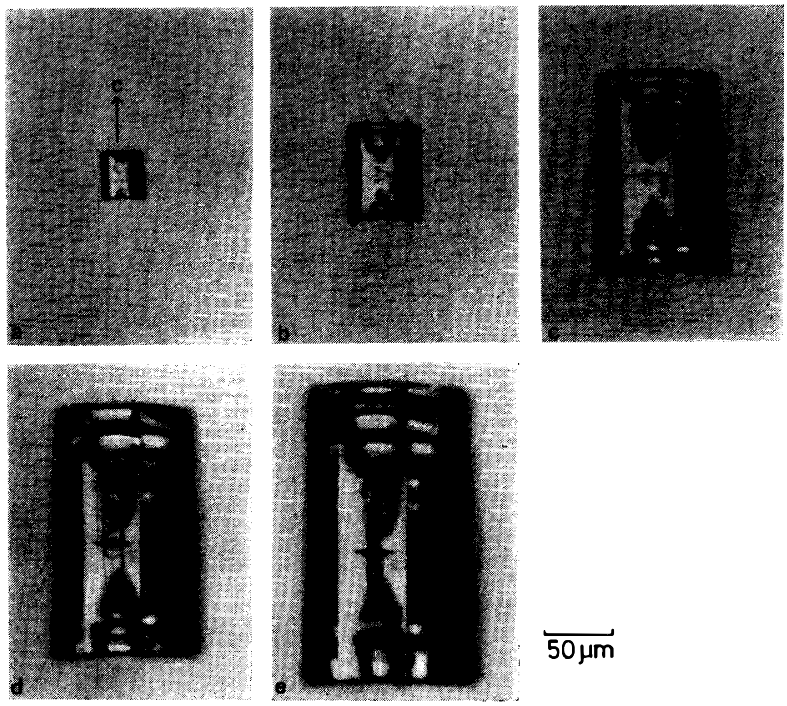

Fig. 3. *Columnar ice crystal with skeletal structures grown in air at 1.0×10^5 Pa at $-30°C$ and a supersaturation of 8.8%: (a) 1.8, (b) 14.3, (c) 60.5, (d) 92.0, (e) 142.9 min.*

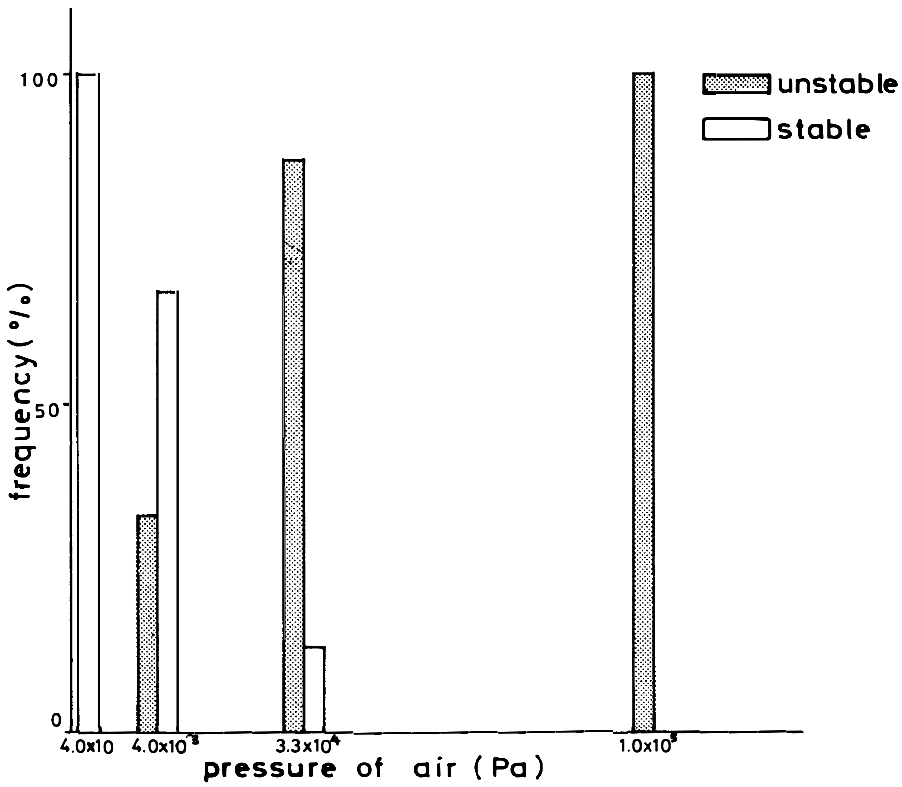

Fig. 4. *Air pressure dependence of the instability of the (0001) face of 80 μm columnar ice crystals grown at $-30°C$ and a supersaturation of 8.8%.*

figure, at a supersaturation below about 2%, long solid prisms grow and become still longer in the ⟨0001⟩ direction when the air pressure increases. On the contrary, at a supersaturation above about 2%, in air at 4.0×10 Pa plate-like ice crystals grow, at 3.3×10^4 Pa column-like ice crystals grow, and at 1.0×10^5 Pa relatively long prisms with skeletal structures grow. That is, the habit of ice crystals growing at -30°C varies markedly not only with supersaturation but also with air pressure.

Figure 6 shows the instability limit of the (0001) face of column-like ice crystals grown in air at 3.3×10^4 and 1.0×10^5 Pa at -30°C. Each point is a mean value of about 10 ice crystals. As shown in this figure, the instability limit of the (0001) face depends not only on supersaturation and crystal size but also on air pressure, that is,

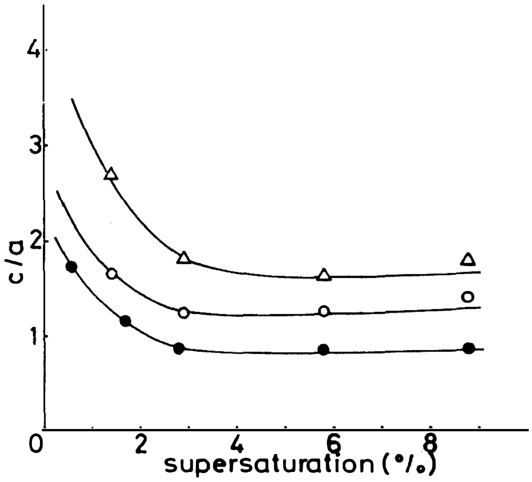

Fig. 5. Air pressure and supersaturation dependences of the ratio of axial lengths c/a of 80 μm ice crystals grown in air at 4.0×10, 3.3×10^4 and 1.0×10^5 Pa at -30° C. ●, ○ and △ show mean values of ice crystals grown in air at 4.0×10, 3.3×10^4 and 1.0×10^5 Pa, respectively.

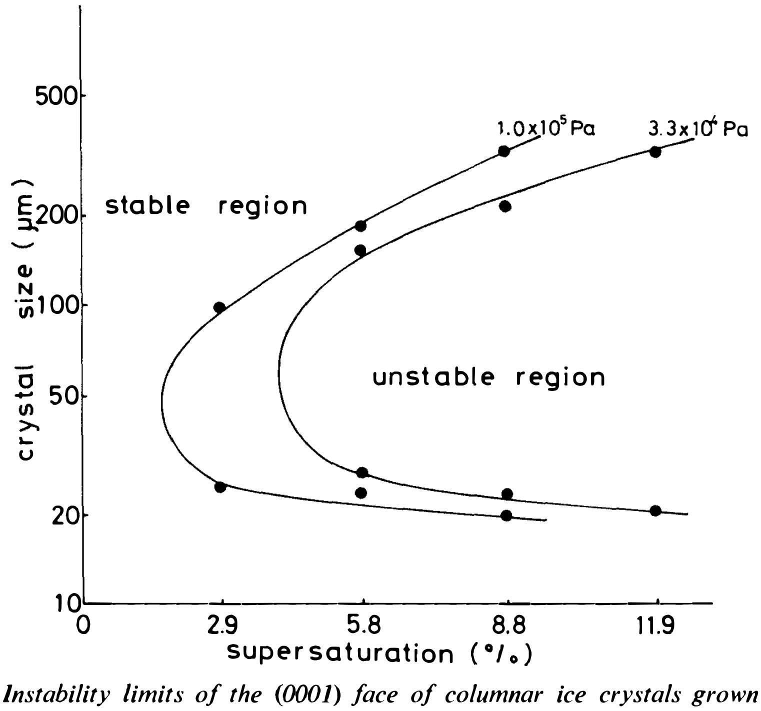

Fig. 6. Instability limits of the (0001) face of columnar ice crystals grown in air at 3.3×10^4 and 1.0×10^5 Pa at -30° C.

in air at 3.3×10^4 Pa polyhedral ice crystals are transformed to skeletal ice crystals at a supersaturation of about 4.1%, while at 1.0×10^5 Pa the same change occurs at a supersaturation of about 1.6%. This means that a morphological instability as well as habit change of ice crystals occurs with increasing air pressure.

3.2. Surface micromorphology of ice crystals growing at −30°C

Figures 7a and 7b show an example of the surface micromorphology on the (0001)

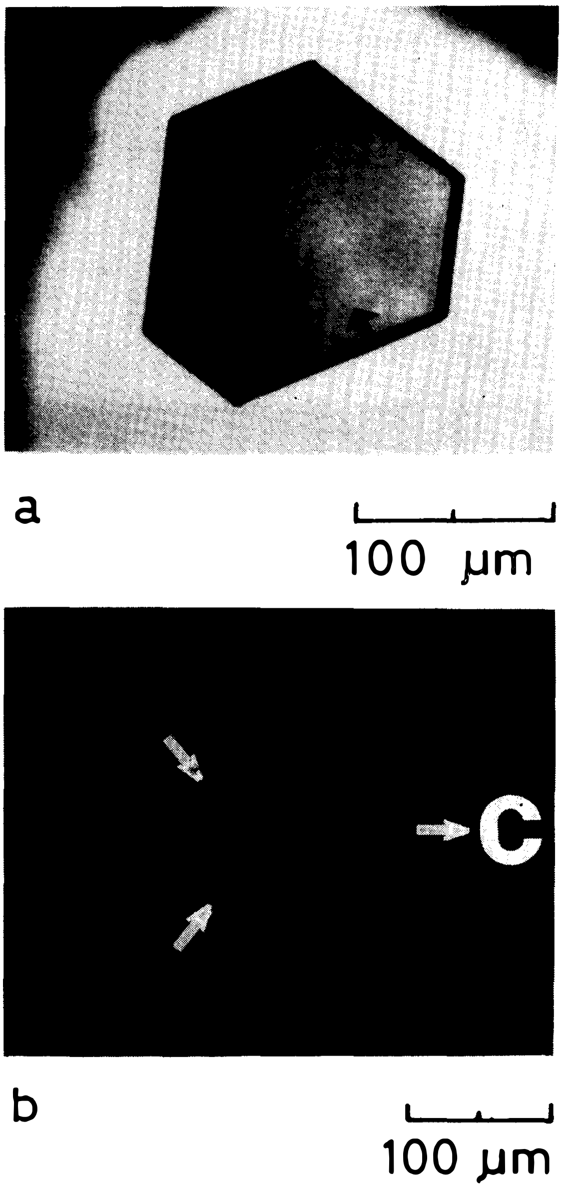

Fig. 7. Surface micromorphology of the (0001) and (10$\bar{1}$0) faces of ice crystals grown in air at 4.0×10 Pa at −30°C, (a) and (b) show the (0001) and (10$\bar{1}$0) faces, respectively; the arrow ↑ is a spiral center.

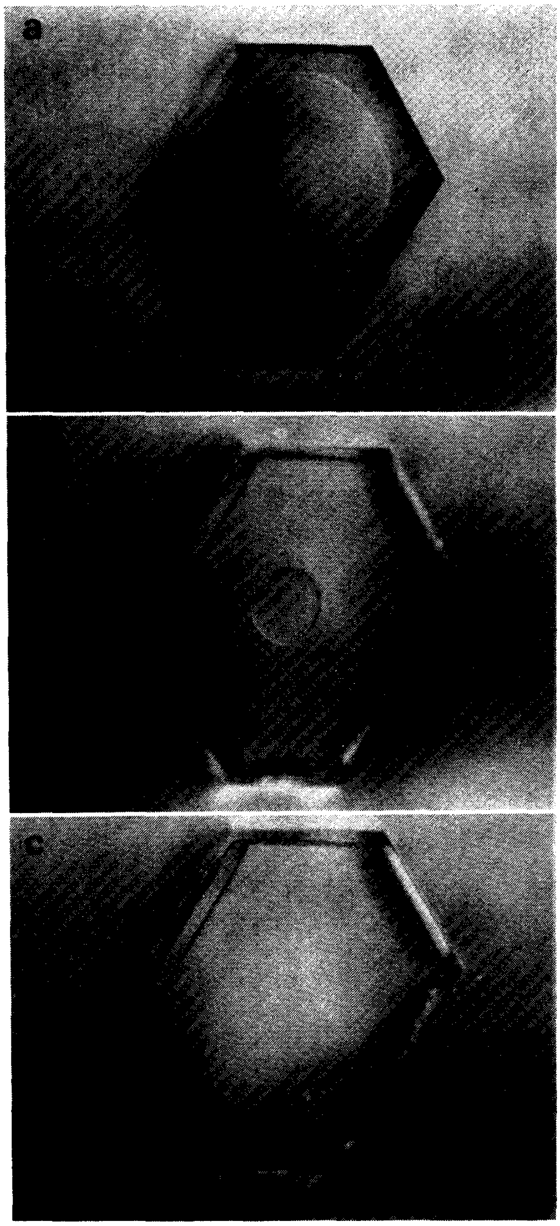

Fig. 8. Surface micromorphology of the (0001) face of ice crystals grown in air at 3.3×10^4 Pa at −30°C and a supersaturation of 11.9%: (a) 0, (b) 45.5, (c) 58.2 min.

and $(10\bar{1}0)$ faces of ice crystals grown in air 4.0×10 Pa at $-30°C$ and supersaturations of 0.4 and 2.7%, respectively. In Fig. 7a, it seems that there is a spiral center from which thin circular steps with equal separation are generated and advance successively toward each edge of the crystal. In Fig. 7b, part of the $(10\bar{1}0)$ face of an ice crystal is photographed to show the spiral center in detail. In Fig. 7b, a curved line which is seen at the center of the $(10\bar{1}0)$ face is the line dividing giant colliding steps originating from the two spiral centers shown by arrows; it runs in the direction along the *c*-axis. Such surface micromorphology as shown in Fig. 7 is always observed at the surface of ice crystals growing under the growth condition described above, so it is recognized that ice crystals growing at a supersaturation below about 2% grow by a screw mechanism when screw dislocations emerge on ice surfaces.

Figure 8 shows an example of surface micromorphology of the (0001) face of an ice crystal growing in air at 3.3×10^4 Pa at $-30°C$ and a supersaturation of 11.9%. In Fig. 8a, duplicate circular macro-steps can be seen. The circular step advances towards a center of the crystal (Fig. 8b); and when the step reaches the center of the crystal, the smooth (0001) face is formed (Fig. 8c).

4. Discussion

Ice crystals have been grown in air at 4.0×10, 3.3×10^4 and 1.0×10^5 Pa at $-30°C$ and various constant supersaturations, and normal growth rates and surface micromorphology of ice crystals have been studied. As a result, the habit of ice crystals grown at $-30°C$ varies markedly not only with supersaturation and crystal size but also with air pressure. The air pressure dependence of the habit of ice crystals means that the three-dimensional diffusion field of water molecules and the absorption of air molecules on ice crystal surfaces are important factors in the habit change of ice crystals. It is seen from Fig. 5 that the ratio of axial lengths c/a of ice crystals increases with increasing air pressure. This means that air molecules restrain the growth of the $(10\bar{1}0)$ face more than that of the (0001) face of ice crystals. This experimental result supports the results obtained by Kuroda and Gonda (1984).

The instability limits of the (0001) face of ice crystals growing at $-30°C$ depend not only on supersaturation and crystal size but also on air pressure. This result means that the three-dimensional diffusion field of water molecules depends not only on supersaturation and the external form of ice crystals but also on air pressure. Because the morphological instability is concerned with two-dimensional nucleation at the corners of ice crystals, Fig. 6 means that the two-dimensional nucleation at the corners of ice crystals is easily formed when the air pressure becomes high. That is, when the air pressure becomes high, the gradient of water vapor pressure at the corners of the crystal becomes steep and lines of constant vapor pressure around the crystal become dense. In this growth condition, water vapor is concentrated onto the corners of the crystal and two-dimensional nucleation occurs at the crystal corners.

Because such surface micromorphology as shown in Fig. 7 is always observed at the surface of ice crystals grown at $-30°C$ and a supersaturation below about 2%, it is considered that ice crystals growing in air at 1.0×10^5 Pa at $-30°C$ grow by a screw mechanism at a supersaturation below about 2% when screw dislocations emerge on the crystal surfaces. On the other hand, from studies of the morphological instability (Figs. 4 and 6) and many observations of surface micromorphology of ice crystals growing in air using a video tape recorder (not shown in this paper), it is considered that ice crystals grow by a nucleation mechanism at a supersaturation above about 10%.

On the basis of experimental results described above, it is inferred that many solid snow crystals forming in the polar regions at a supersaturation below about 2% (SHIMIZU, 1963; HIGUCHI, 1968; KIKUCHI and HOGAN, 1979) grow by a screw mechanism, while many snow crystals with skeletal structures (SATOW, 1983) forming in the polar regions at a supersaturation above about 10% grow by a nucleation mechanism.

5. Conclusions

Single ice crystals have been grown in air at 4.0×10, 3.3×10^4 and 1.0×10^5 Pa at $-30°C$ and various constant supersaturations, and measurements of normal growth rates of the ice crystals and *in situ* observations of their surface micromorphology have been made. The results are as follows.

(1) The habit of ice crystals growing at $-30°C$ varies markedly not only with supersaturation and crystal size but also with air pressure. This fact means that the three-dimensional diffusion field of water molecules and the adsorption of air molecules on crystal surfaces play an important role in habit change of ice crystals.

(2) The morphological instability of ice crystals growing at $-30°C$ depends markedly not only on supersaturation and crystal size but also on air pressure.

(3) It is considered that ice crystals growing in air at 1.0×10^5 Pa at $-30°C$ grow by a screw mechanism at a supersaturation below about 2%, and by a nucleation mechanism at a supersaturation above about 10%.

(4) Therefore, it is inferred that many solid snow crystals formed in polar regions (SHIMIZU, 1963; HIGUCHI, 1968; KIKUCHI and HOGAN, 1979) grow by a screw mechanism at a supersaturation below about 2%, while many snow crystals with skeletal structures (SATOW, 1983) formed in the polar regions grow by a nucleation mechanism at a supersaturation above about 10%.

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