SURFACE MICROMORPHOLOGY OF COLUMNAR ICE CRYSTALS GROWING IN AIR AT HIGH AND LOW SUPERSATURATIONS

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Abstract: The growth mechanisms of long prisms with skeletal structures precipitating in the polar regions are studied by observing the surface micromorphology of columnar ice crystals growing in air at high and low supersaturations. It is concluded that long hollow prisms, that is, long prisms with large skeletal structures grow by a two-dimensional nucleation mechanism under supersaturation above about 10%, while long prisms with small skeletal structures grow by a screw dislocation mechanism under supersaturation below about 2%.

1. Introduction

Before the 1980's there had been only a few observations on snow single crystals precipitating in the polar regions, long solid prisms (SHIMIZU, 1963) and rectangular crystals (HIGUCHI, 1968), and on diamond dust type ice crystals (KIKUCHI and HOGAN, 1979) and so on. The morphology and the growth mechanisms of these polyhedral crystals have been experimentally studied by KOBAYASHI (1965), GONDA and KOIKE (1982) and GONDA (1983).

Recently, a long hollow prism has been observed at Mizuho Plateau, Antarctica (SATOW, 1983). Thereafter, WADA and GONDA (1985) have quantitatively discussed the crystallographic properties and the growth conditions of the long hollow prisms observed at Mizuho Station, Antarctica. Meanwhile, the habit and the morphological instability of columnar ice crystals grown at low temperatures have been experimentally studied by GONDA *et al.* (1984) and GONDA and GOMI (1985). Throughout a series of these studies, the growth mechanisms of long prisms with skeletal structures have become of major interest.

In this paper, the growth mechanisms of long prisms with skeletal structures precipitating in the polar regions are discussed on the basis of the surface observations of the columnar ice crystals grown in air at high and low supersaturations.

2. Experimental Procedures

A growth chamber of ice crystals is described in detail in a previous paper (GONDA and KOIKE, 1982). The growth chamber is cooled by flowing an electric current of 0–5 amperes to the thermoelectric cooling panels attached at the top and the bottom of the chamber, keeping a growth substrate at a slightly higher temperature than that of an ice plate. The temperature of the growth substrate and the ice plate is controlled by regulating the electric current to flow to the thermoelectric cooling panels.

When the growth substrate reaches a desired temperature, water vapor is supplied by keeping the ice plate at a slightly higher temperature than that of the growth substrate. Minute ice crystals are formed in air by inserting 2 cm^3 of sufficiently diluted silver iodide smoke into the growth chamber. Minute ice crystals formed in air fall quickly and grow on the growth substrate. Growing ice crystals are observed *in situ* using a differential interference microscope.

The surface micromorphology of columnar ice crystals was observed in a desired air pressure after a large polyhedral ice crystal was formed under low air pressure. The surface photographs were taken using a single-lens reflex camera or a video camera which is attached at the top of a cylindrical mirror.

3. Experimental Results

3.1. Columnar ice crystals with large skeletal structures grown under relatively high supersaturation

Figure 1 shows a columnar ice crystal with large skeletal structures grown in air of 1.0 atm at $-30°C$ and 8.8% supersaturation. Under this growth condition, the skeletal structures are formed on the (0001) face of the crystal when the crystal size reaches about 20 μm. The skeletal structures develop with increasing crystal size, that is, the crystal becomes more unstable. Here, this crystal is photographed from the $[11\bar{2}0]$ direction, and because the $(10\bar{1}0)$ face of the crystal is out of focus, a boundary layer along *a*-axis of the $(10\bar{1}0)$ faces is invisible. The surface micromorphology of columnar ice crystals was observed in order to study the growth mechanisms of these crystals.

Figure 2 shows the microscopic photographs of the (0001) face of a columnar ice crystal grown in air of 1.0 atm at $-30°C$ and 13% supersaturation, where the size ratio c/a of this crystal is about 1.5. When we observe the surface micromorphology of an ice crystal, as the air is introduced into the chamber after a large polyhedral ice crystal having the size ratio c/a nearly equal to unity was formed in air at low pressure, the size ratio c/a of the ice crystal is relatively small except for the experiments at very low supersaturation. In the case of this photograph, though the two-dimensional nuclei at the corners are not visible, it will be evident that the growth layers on the (0001) face originate in the two-dimensional nuclei at the corners of the crystal. At 21.2 min (b) after the growth stage (a), the center of the (0001) face became concave in order that the growth layers originated in two-dimensional nuclei did not reach the center of the (0001) face. At 36.1 min (c), the concave at the center of the (0001) face became considerably deep by the bunching of the growth layers running near the center of the crystal. At 46.5 min (d), it is seen that four giant steps were formed on the (0001) face by the bunching of relatively thin growth layers. At 50.7 min (e), this phenomenon became more clear. At 58.8 min (f), another new giant step is seen.

Figure 3 shows the video photographs of the (0001) face of a columnar ice crystal grown in air of 1.0 atm at $-15°C$ and 10% supersaturation, where the size ratio c/a of this crystal is about 2.6. Here, it is seen that the video scanning lines (dark bold

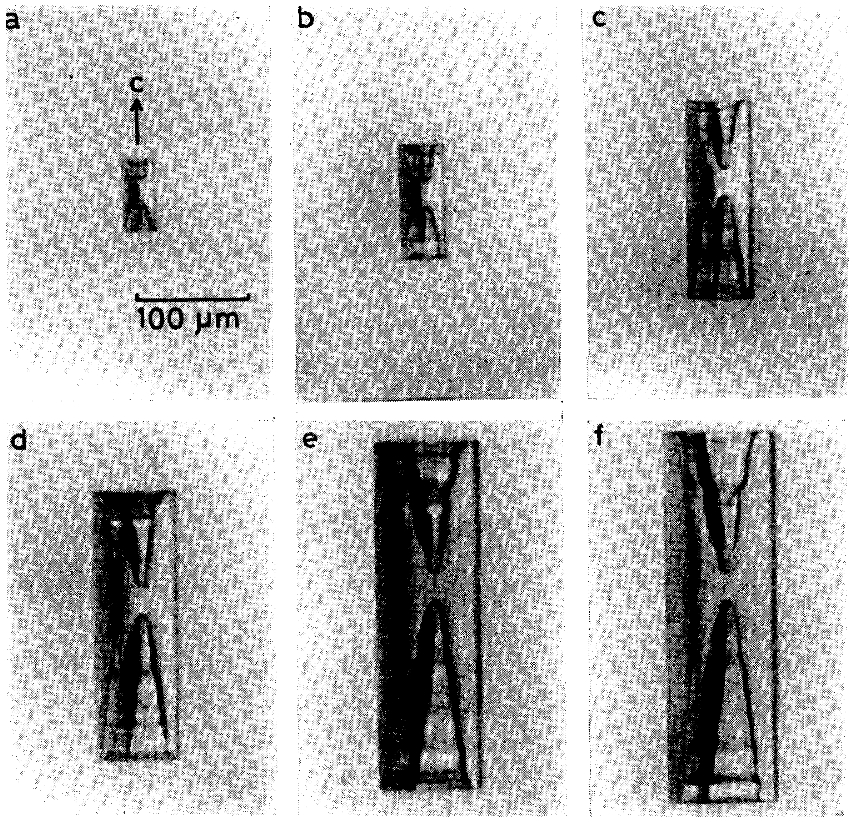

Fig. 1. *Columnar ice crystal with large skeletal structures grown in air of 1.0 atm at −30°C and 8.8% supersaturation. (a) 0, (b) 9.9, (c) 37.3, (d) 76.0, (e) 143.7, (f) 173.6 min.*

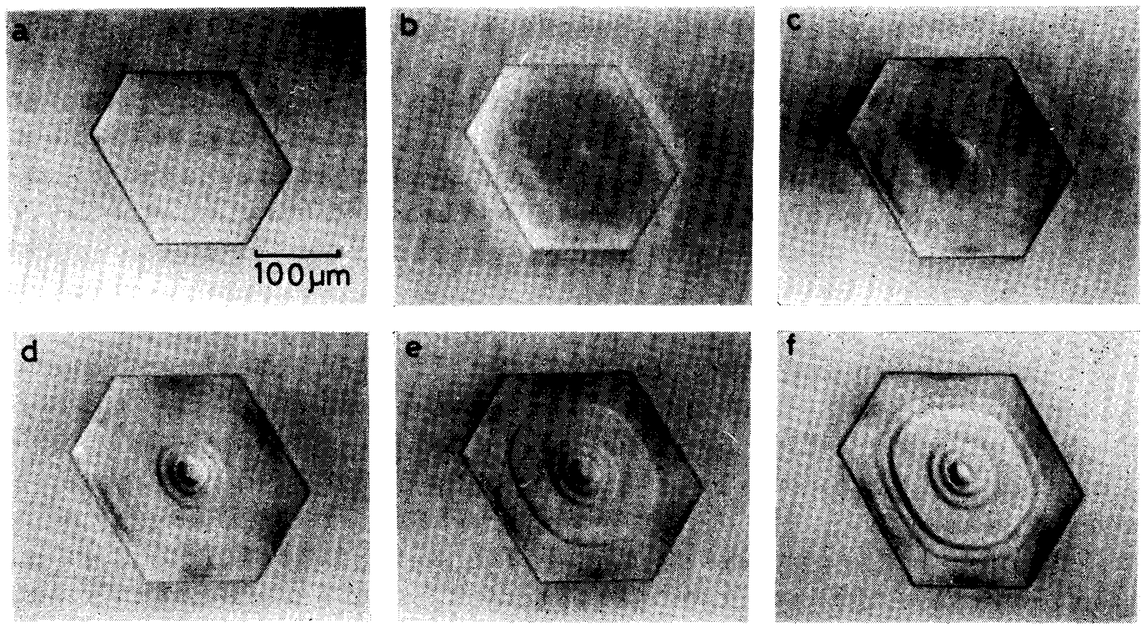

Fig. 2. *Microscopic photographs of the (0001) face of a columnar ice crystal grown in air of 1.0 atm at −30°C and 13% supersaturation. (a) 0, (b) 21.2, (c) 36.1, (d) 46.5, (e) 50.7, (f) 58.8 min.*

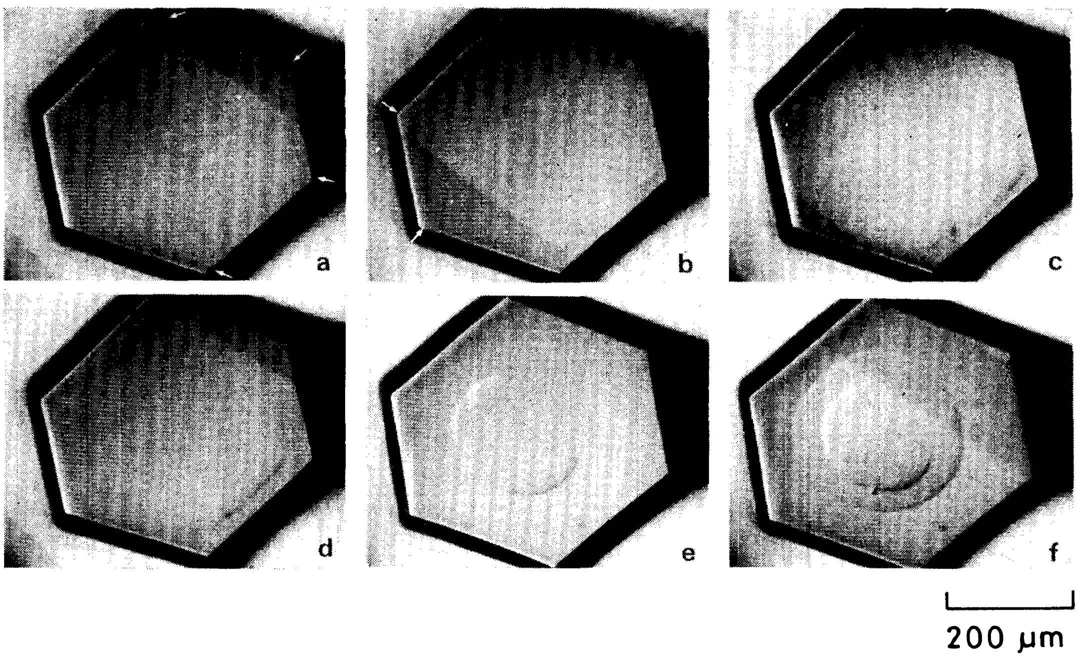

Fig. 3. Video photographs of the (0001) face of a columnar ice crystal grown in air of 1.0 atm at −15°C and 10% supersaturation. (a) 0, (b) 0.3, (c) 0.5, (d) 1.3, (e) 7, (f) 11.8 min.

lines) run obliquely across the crystal corners. As shown in the figure, in the growth stage (a) the growth layers (arrows ↑) originated in the two-dimensional nuclei at the corners of the crystal can be seen. At 0.3 min (b) after the growth stage (a), the growth layers (arrows ↑) starting from the other corners can be seen, too. After 0.5 min (c), it is seen that the growth layers which started from each corner spread along each edge of the crystal and a circular thick growth layer begins to be formed. Here, a screw dislocation had emerged near the corner at the top of the right on the (0001) face of this crystal. Accordingly, the growth layer of the top of the right spreads toward the center of the (0001) face. After 1.3 min (d), this phenomenon can be clearly seen. After 7.0 min (e), a circular concave is seen in the center of the (0001) face. After 11.8 min (f), another thick growth layer advances from the outside to the center of the (0001) face, and as a result the concave of the (0001) face became a double bottom.

From these surface observations, the growth layers originated in two-dimensional nuclei at each corner of the (0001) face at relatively high supersaturation advance toward the center of the (0001) face, and as a result, large skeletal structures are formed on the (0001) face.

3.2. *Columnar ice crystals with small skeletal structures grown under low supersaturation*

Figure 4 shows a long prism with small skeletal structures grown in air of 1.0 atm at −30°C and 1.4% supersaturation. Under low supersaturation like this, it is known that long prisms grow by screw dislocations emerged on the (0001) face of the crystals (GONDA and KOIKE, 1982). In the growth stage (a), the crystal grows in the state of polyhedral form. However, at 35.4 min (b) after the growth stage (a), small skeletal structures are formed on the (0001) face and the crystal grows unstablly. After

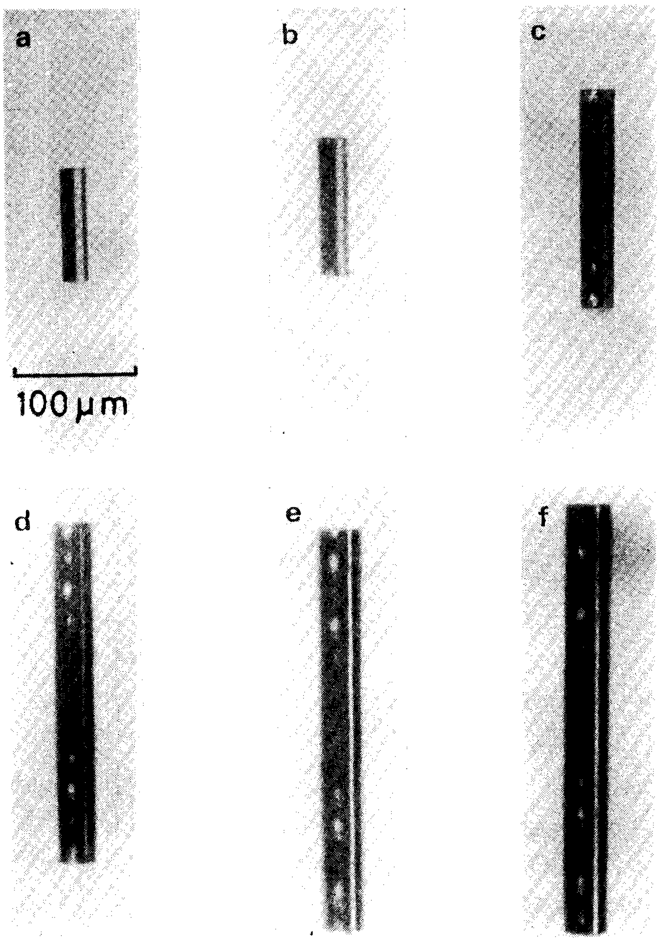

Fig. 4. Long prism with small skeletal structures grown in air of 1.0 atm at −30°C and 1.4% supersaturation. (a) 0, (b) 35.4, (c) 72.5, (d) 141.9, (e) 187.7, (f) 212 min.

72.5 min (c), the (0001) face which became flat came to be unstable again. After 141.9 min (d), it is seen that the skeletal growth on the (0001) face is repeated four times. After 187.7 min (e), the skeletal growth is repeated five times. After 212 min (f), the polyhedral crystal grows again. As shown in this figure, the stable and unstable growth is repeated many times at low supersaturation with increasing crystal size. As a result, many air bubbles are enclosed into the crystal and a long prism with small skeletal structures grows. Here, there is no skeletal structure at the center of the $(10\bar{1}0)$ face of this crystal because the growth rate of the $(10\bar{1}0)$ face is very small as compared with that of the (0001) face, so the $(10\bar{1}0)$ face grows stable.

Figure 5 shows the microscopic photographs of the (0001) face of a columnar ice crystal grown in air of 1/3 atm at −30°C and 2.8% supersaturation, where the size ratio c/a of this crystal is about 1.5. In the case of this crystal, though a screw dislocation at the corner is invisible, it is evident that a screw dislocation emerges at a corner on the (0001) face of the crystal. The reason is that an evaporation pit and evaporation layers due to a screw dislocation were formed because the chemical potential in the position where a screw dislocation emerges is high when the crystal was kept in an evaporation state. The second is that two-dimensional nuclei are not formed on the ice crystal surface under such low supersaturation as 2.8% from the theoretical consideration. In the growth stages (a) and (b), it is understood that relatively thick growth layers are formed by the bunching of thin growth layers and advance from

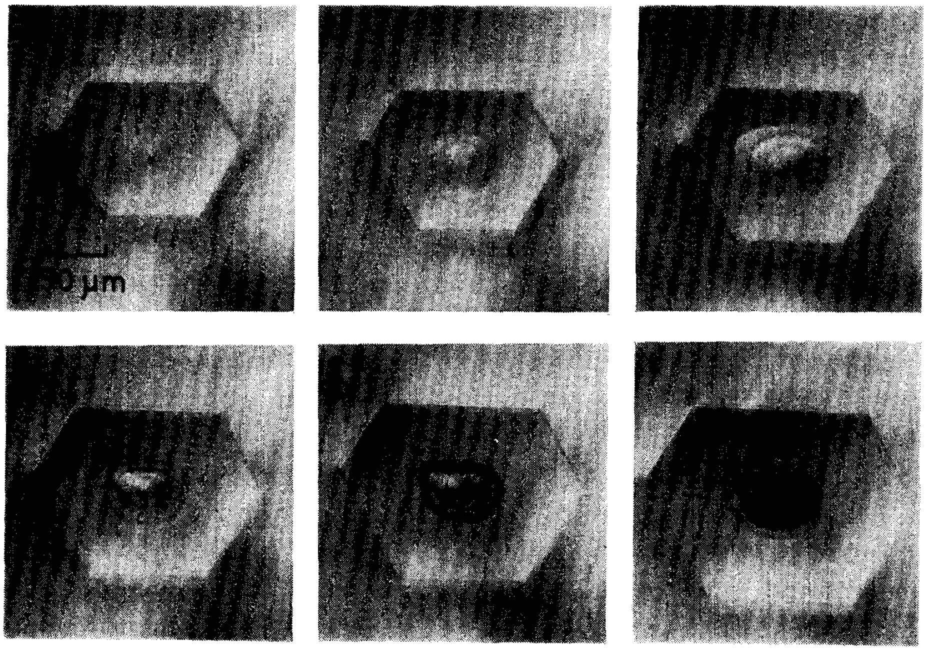

Fig. 5. *Microscopic photographs of the (0001) face of a columnar ice crystal grown in air of 1/3 atm at −30°C and 2.8% supersaturation. (a) 0, (b) 0.6, (c) 2.0, (d) 3.6, (e) 5.1, (f) 8.1 min.*

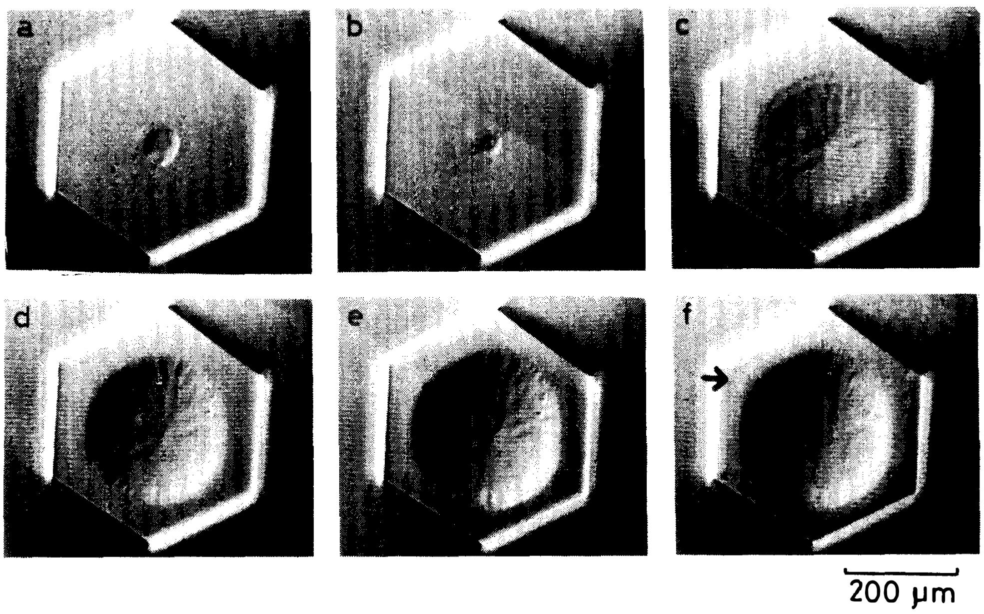

Fig. 6. *Video photographs of the (0001) face of a columnar crystal grown in air of 1.0 atm at −7°C and 1.6% supersaturation. (a) 0, (b) 1.0, (c) 3.0, (d) 5.3, (e) 6.2, (f) 6.7 min.*

the outside to the center of the (0001) face. At 2.0 min (c) after the growth stage (a), a new growth layer advances toward the center of the crystal. After 3.6 min (d), the giant steps are formed by further bunching of relatively thick growth layers, and as a result the skeletal structures are formed on the (0001) face. After 5.1 min (e) and 8.1 min (f), the skeletal structures on the (0001) face tend to diminish and the (0001) face tends to become stable. However, when the crystal grows further it is found that new thin growth layers advance from the outside to the center of the (0001) face. That is, the growth process of (a)–(f) is repeated many times.

Figure 6 shows the video photographs of the (0001) face of a columnar ice crystal grown in air of 1.0 atm −7°C and 1.6% supersaturation, where the size ratio c/a of this crystal is about 1.2. In the figure, it is seen that the video scanning lines (dark bold lines) run oblique to the crystal edges. In order to increase the supersaturation in the growth chamber when the air was introduced into the chamber, two-dimensional nuclei were formed at the corners of the crystal and the deep concave was formed near the center of the (0001) face (growth stage (a)). After that, the supersaturation in the growth stage (a) was kept at 1.6%. Therefore, further nucleation at the corners of the crystal is not formed. In the case of this crystal, a screw dislocation emerges at the position indicated by an arrow (photograph (f) is the evaporation stage). At 1.0 min (b) after the growth stage (a), it is seen that the growth layers with thin steps run from the outside to the center of the (0001) face. After 3.0 min (c), the deep concave produced when the air was introduced in the chamber diminishes and the growth layers with relatively thin steps advance in succession toward the center of the (0001) face. After 5.3 min (d) and 6.2 min (e), the skeletal structure with shallow concave which is characteristic at low supersaturation is formed by the bunching of the thin growth layers. After 6.7 min (f), as soon as the crystal is kept in the evaporation state, the evaporation began to arise preferentially at the dislocation site indicated by the arrow. It is seen that a circular (0001) facet is also formed by the evaporation at the center of the concave. Moreover, it will be understood that there are evaporation steps at the edges of the crystal if we carefully observe the surface of this crystal in evaporation state (f).

Based on the surface observations described above, it is understood that when screw dislocations emerge near the corners of the (0001) face under low supersaturation, the skeletal structures are formed on the (0001) face.

4. Discussion

In order to study the growth mechanisms of long hollow prisms precipitating in the polar regions, the surface micromorphology of columnar ice crystals growing in air at relatively high and low supersaturations were studied using a differential interference microscope. As a result, it is understood that the large skeletal structures formed at high supersaturation are formed by the bunching of thick growth layers originated in two-dimensional nuclei at the corners of the (0001) face, which advance from the corners to the center of the (0001) face where the surface supersaturation is the lowest. Columnar ice crystals with large skeletal structures growing under high supersaturation remain in the state of unstable growth with increasing crystal size, and as a result, sheath-like crystals are formed.

On the contrary, small skeletal structures of columnar ice crystals growing under low supersaturation are formed by the bunching of thin growth layers originated in screw dislocations which emerge near the corners of the (0001) face. However, when screw dislocations emerge near the center of the (0001) face of the crystal under supersaturation below about 2%. The growth layers originated in screw dislocations run from the center of the (0001) face to the outward direction, and as a result, polyhedral ice crystal is formed (figure is not shown). That is, it is understood that the instability of the (0001) face under low supersaturation depends on screw dislocations emerging either at the corners or at the center of the (0001) face.

On the other hand, the step height of the growth layers due to two-dimensional nuclei forming at high supersaturation is high and the step velocity is small, while the step height of the growth layers due to screw dislocations forming at low supersaturation is low and the step velocity is large. This fact means that large skeletal structures and small skeletal structures are formed at high and low supersaturations, respectively. Moreover, step patterns and step motion on ice crystal surfaces which was described above are reproducible many times in our experiments. Therefore, it is concluded that long hollow prisms, that is, long prisms with large skeletal structures observed at Mizuho Station, Antarctica (Fig. 7), were formed by the bunching of relatively thick growth layers originated in two-dimensional nuclei at the corners of the (0001) face.

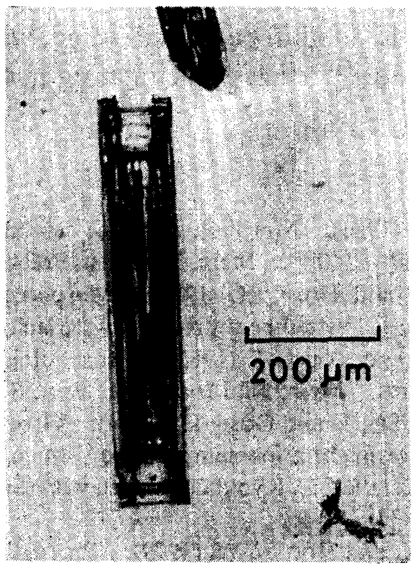

Fig. 7. Long hollow prism observed at Mizuho Station, Antarctica (WADA and GONDA, 1985).

On the other hand, it is considered that long prisms with small skeletal structures which have symmetrically many air bubbles inside the crystal (KLINOV, 1960) are formed by the bunching of thin growth layers originated in the emergence of screw dislocations near the corners of the (0001) face. When the observations of natural snow crystals and diamond dust particles under supersaturation below about 2% are carefully made in the polar regions, the same long prisms as shown in Fig. 4 should be found.

5. Conclusion

The surface micromorphology of columnar ice crystals growing in air at high and low supersaturations were observed in order to study the growth mechanisms of long prisms with skeletal structures precipitating in the polar regions. The results obtained in this study are as follows.

1) Large skeletal structures on the (0001) face of columnar ice crystals growing in air at supersaturation above about 10% are formed by the bunching of relatively thick growth layers originated in two-dimensional nuclei at the corners of the (0001) face.

2) Small skeletal structures on the (0001) face of columnar ice crystals growing in air at supersaturation below about 2% are formed by the bunching of relatively thin growth layers originated in screw dislocations which emerged near the corners of the (0001) face.

3) When screw dislocations emerge near the center of the (0001) face of a columnar ice crystal growing in air at supersaturation below about 2%, the columnar ice crystal grows in the state of polyhedral form.

4) It is concluded from the experimental results that long prisms with large skeletal structures precipitating in the polar regions are formed by a two-dimensional nucleation mechanism under supersaturation above about 10%.

5) Long prisms with small skeletal structures having many air bubbles inside the crystals should be found when we carefully observe natural snow crystals and diamond dust particles precipitating in the polar regions.

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