MORPHOLOGY OF SINGLE SNOW CRYSTALS GROWING IN AIR AT LOW TEMPERATURES

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Abstract: In order to study the morphology and the growth mechanisms of single snow crystals observed in polar areas, ice crystals were formed in air at 1.0 atm at −30°C and various constant supersaturations. At low temperatures, minute plate-like and column-like ice crystals are formed at the same time under the same environmental conditions; the morphology of these crystals depends not only on temperature but also on supersaturation, crystal size and macroscopic surface structure. From our experimental results, it is inferred that on diamond dust particles observed in Antarctica, when columnar ice crystals are predominantly observed, the supersaturation is lower than about 2%, while when plate-like ice crystals are predominantly observed, the supersaturation is higher than about 2%. Moreover, it is inferred that long solid prisms and rectangular snow crystals observed in polar areas grow by the screw dislocation mechanisms at a supersaturation lower than about 2%.

1. Introduction

In accordance with KOBAYASHI's experiment (1961, 1965), in such low temperature regions as Antarctica and the Arctic areas, columnar snow crystals are expected to be formed. According to GONDA's experiment (1977) and the observations of diamond dust particles by KIKUCHI and HOGAN (1979), however, it has been found that even at low temperatures below −30°C, plate-like ice crystals are also formed.

On the other hand, KURODA and LACMANN (1982) have proposed theoretically that the habit of single snow crystals growing below −20°C depends not only on temperature but also on supersaturation. Thereafter, GONDA and KOIKE (1982) have studied experimentally the temperature and supersaturation dependences of the habit of single ice crystals growing in air at 1.0 atm at temperatures below −20°C; and it has been confirmed experimentally that except for the results at low supersaturation below about 2%, the change in habit of small ice crystals of 15 μm with temperature and supersaturation coincides with the theory of KURODA and LACMANN (1982).

The purposes of this study are to describe the characteristic of single ice crystals growing in air at 1.0 atm at −30°C, and to infer experimentally the growth mechanisms of such peculiar snow crystals as long solid prisms (SHIMIZU, 1963) and rectangular snow crystals (HIGUCHI, 1968) observed in polar areas.

2. Experimental Procedures

A schematic diagram of a growth chamber to form ice crystals at low temperatures is described in a previous paper (GONDA and KOIKE, 1982). The growth chamber is designed for controlling independently the temperature T_2 of a water vapor source and the temperature T_1 of a growth substrate. The chamber is cooled down to a specified temperature by applying an electric current of 3 to 5 amperes to the thermoelectric cooling panels. In order to keep the supersaturation in the chamber constant, the temperature difference T_2-T_1 between the water vapor source and the growth substrate is held constant. A small amount of sufficiently rarefied silver iodide smoke is inserted into the chamber in order to form ice crystals and to avoid the interaction among them when ice crystals grow on the substrate. By taking such precautions, we can form ice crystals at constant temperature and supersaturation. We observe *in situ* growing ice crystals by differential interference microscope.

The growth conditions of natural snow crystals are different from those of ice crystals described here, especially concerning the transport of the latent heat of sublimation. However, it is known that the transport effect of latent heat on the morphology of ice crystals is not so effective (GONDA and KOMABAYASI, 1971; GONDA, 1976). Furthermore, when we consider the growth of the first half of an ice crystal growing on a substrate, the effect of the crystal shape on the volume diffusion of water vapor may be ignored. Therefore, it will be possible to infer the growth forms and mechanisms of natural snow crystals on the basis of our experiments.

3. Habit of Ice Crystals Growing at Low Temperatures

In order to study the morphology and the growth mechanisms of single snow crystals observed in polar areas, ice crystals were formed in air at 1.0 atm at

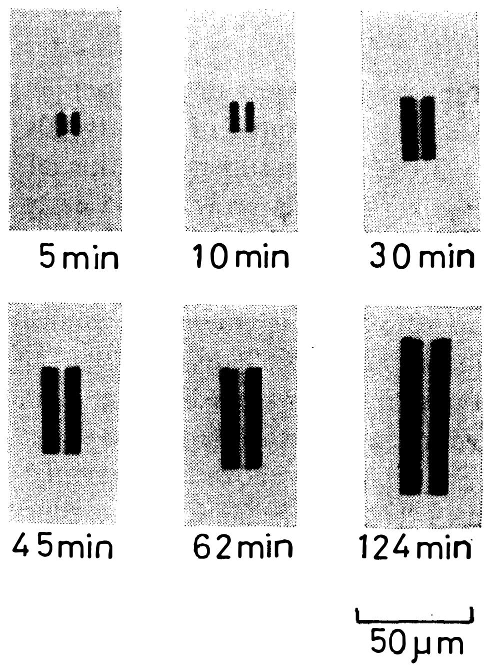

Fig. 1. Columnar ice crystal grown predominantly in air at 1.0 atm at −30°C and a supersaturation of 1.0%.

$-30°C$ and various constant supersaturations. At $-30°C$ and one constant supersaturation, the experiments of ice crystal growth have been done 23–25 times. About two ice crystals were formed in one experiment in the field of view of a microscope. Accordingly, the total number of ice crystals formed at one constant supersaturation is about fifty-five, the number of single crystals forty-two and that of polycrystals thirteen.

Figure 1 shows an example of ice crystals formed with high frequency in air at 1.0 atm at $-30°C$ and a supersaturation of 1.0%. At low supersaturation like this, columnar ice crystals grow predominantly in the early stage of growth. The columnar ice crystals are transformed into long solid prisms with increase of elapsed

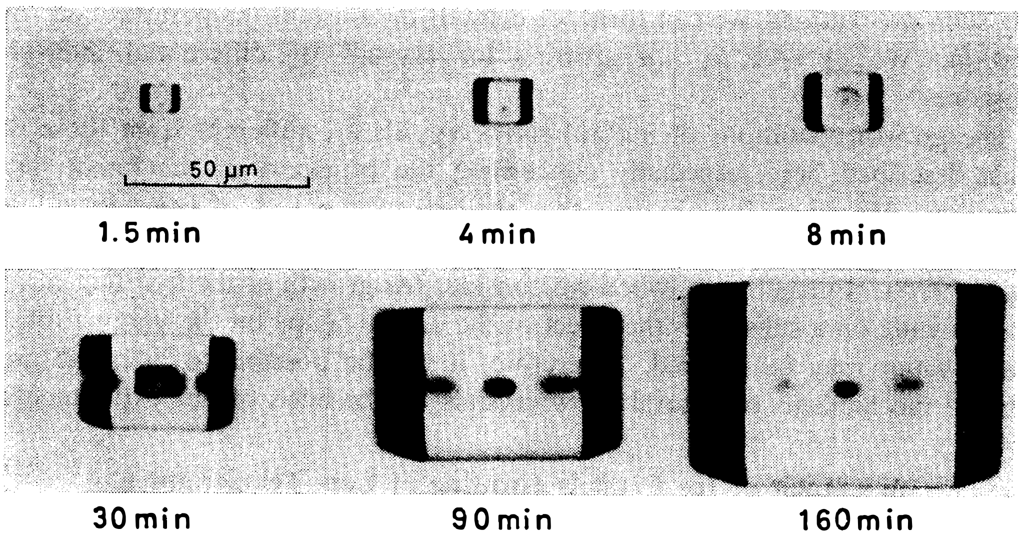

Fig. 2. Plate-like ice crystal grown predominantly in air at 1.0 atm at —30°C and a supersaturation of 8.7%.

time as the result of preferential growth along the $\langle 0001 \rangle$ directions. Figure 2 shows an example of ice crystals formed with high frequency in air at 1.0 atm at $-30°C$ and a supersaturation of 8.7%. At moderate supersaturation, solid plate-like ice crystals grow in the early stage of growth. However, when the crystal size increases, skeletal structure is generated only on the prismatic faces of the crystals. Although the skeletal structure develops with increase of the crystal size up to about 80 μm, it gradually diminishes with further increase of crystal size. In growth stages as the skeletal structure develops, the growth rate along the $\langle 10\bar{1}0 \rangle$ directions of the crystals is larger than that along the $\langle 0001 \rangle$ directions as compared with the growth rate in the stages in which the skeletal structure diminishes.

Figure 3 shows an example of ice crystals formed with high frequency in air at 1.0 atm at $-30°C$ and a supersaturation of 41%. At high supersaturation like this, columnar ice crystals with skeletal structure on both basal and prismatic faces grow predominantly in the early stage of growth. As shown in the figure, when the crystal size increases up to about 100 μm in size, the skeletal structure on the basal faces diminishes although that on the prismatic faces still develops. That is,

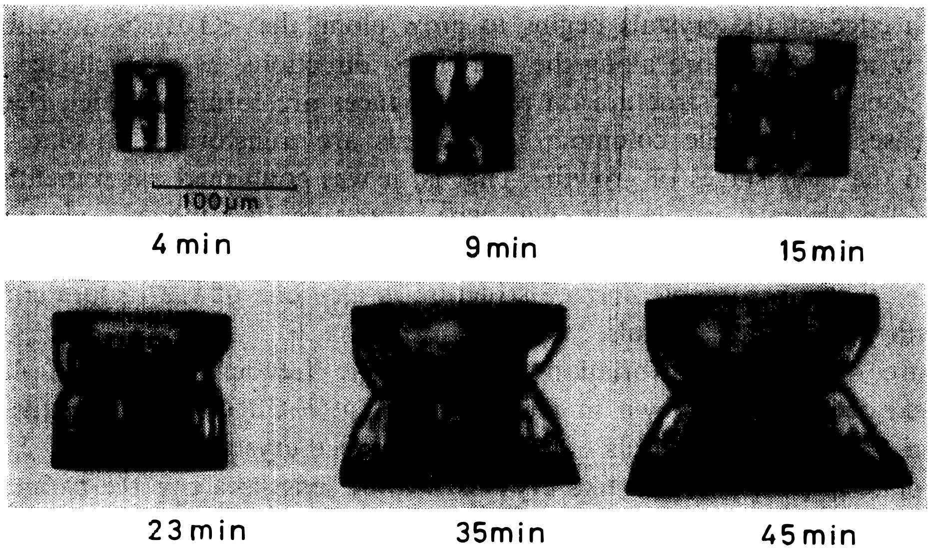

Fig. 3. *Ice crystal grown predominantly in air at 1.0 atm at —30°C and a supersaturation of 41%.*

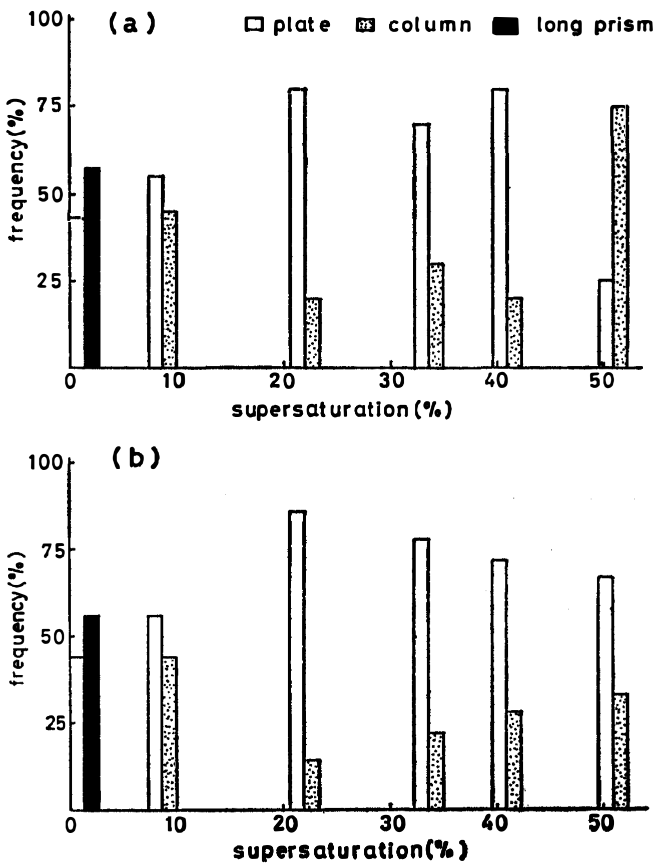

Fig. 4. *Percentage frequencies of plate-like and column-like ice crystals grown in air at 1.0 atm at —30°C versus supersaturation. (a): Ice crystals of 15 μm. (b): Ice crystals of 50 μm.*

once each edge of the crystals begins to grow along the $<10\bar{1}0>$ directions, the edges grow more and more along the $<10\bar{1}0>$ directions, as a result, ice crystals with developed skeletal structure on prismatic faces are formed. When the crystal size increases further, the columnar ice crystals are transformed to plate-like ice crystals in the later stages of growth. That is, it was confirmed experimentally that columnar ice crystals growing at high supersaturation like this are always transformed to plate-like ice crystals in the later stages of growth. This experimental fact means that macroscopic surface structure is also one of the factors controlling the growth form of ice crystals.

Figure 4 shows the variation of percentage frequencies of plate-like and column-like ice crystals grown in air at 1.0 atm at $-30°C$ versus relative supersaturation σ. (a) is ice crystals of 15 μm, (b) is ice crystals of 50 μm. Here, the sizes of plate-like and column-like crystals are the lengths along the a- and c-axes of the crystals, respectively. As shown in the figure, in the case of ice crystals of 15 μm, when $\sigma <$ about 2%, long solid prisms are formed with high frequency. However, when 40% $> \sigma >$ about 2%, solid plate-like ice crystals are formed with high frequency. At a supersaturation above about 50%, short prisms are formed with high frequency. Next, in the case of ice crystals of 50 μm, when $\sigma <$ about 2%, long solid prisms are also formed with high frequency. When $\sigma >$ about 2%, plate-like ice crystals predominantly grow: especially, at high supersaturation above about 50%, short prisms are transformed to skeletal plates with increasing crystal size (see Figs. 3 and 4). As described here, at low temperatures, plate-like and column-like ice crystals are formed simultaneously under the same environmental conditions. Moreover, growth frequencies of these ice crystals growing at low temperatures depend on supersaturation and crystal size, too. (The figure of growth

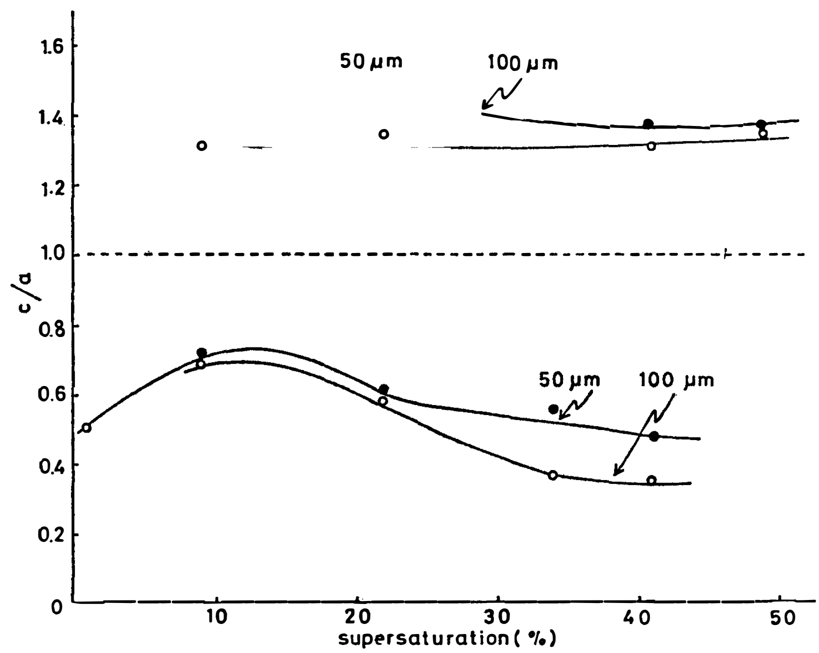

Fig. 5. Supersaturation dependence of the ratios of the axial lengths c/a of plate-like and column-like ice crystals grown in air at 1.0 atm at —30°C.

frequency of these crystals versus crystal size is not shown.)

Figure 5 shows the supersaturation dependence of the ratios of the axial lengths c/a of plate-like and column-like ice crystals grown in air at 1.0 atm at $-30°C$. In the figure, the sizes of plate-like and column-like crystals are the lengths along the *a*- and *c*-axes of the crystals, respectively. In the case of columnar ice crystals, long solid prisms grow when the supersaturation is low; but short prisms grow with increasing supersaturation. In the case of plate-like ice crystals, relatively thin plates grow when the supersaturation is low; but thick plates grow at moderate supersaturation; and very thin plates grow at high supersaturation. Moreover, as seen in the figure, the ratios of the axial lengths c/a of ice crystals vary with crystal size. Therefore, it is understood that the habit of ice crystals depends not only on temperature but also on supersaturation and crystal size.

4. Interface Stability of Ice Crystals Growing in Air

Figure 6 shows the stability limit of {0001} and $\{10\bar{1}0\}$ faces of plate-like ice crystals growing in air at 1.0 atm at $-30°C$. Each value in the figure is for an average of 10–20 ice crystals. As shown in the figure, the stability limits of the {0001} and $\{10\bar{1}0\}$ faces of the crystals depends on both supersaturation and crystal size; here, the crystal sizes which are related to the stability limit of the {0001} and $\{10\bar{1}0\}$ faces are the lengths along the *c*-axis, L_c and that along the *a*-axis, L_a respectively.

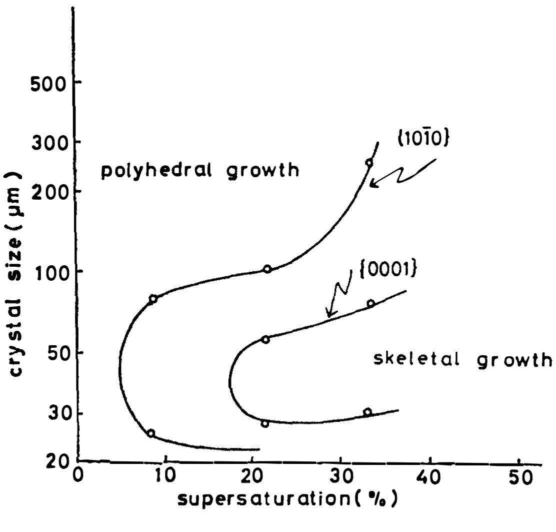

Fig. 6. Stability limit of the {0001} and $\{10\bar{1}0\}$ faces of plate-like ice crystals grown in air at 1.0 atm at $-30°C$.

For example, the stability of the {0001} and $\{10\bar{1}0\}$ faces of the ice crystals growing in air at 1.0 atm at $-30°C$ and a supersaturation of 20% is as follows. When $L_a < 22\ \mu m$, both the {0001} and $\{10\bar{1}0\}$ faces of the crystals are optically smooth, that is, polyhedral (solid) ice crystals grow. When $100\ \mu m > L_a > 22\ \mu m$ and $L_c < 30\ \mu m$, ice crystals with skeletal structure on the $\{10\bar{1}0\}$ faces and optically smooth {0001} faces grow. When $100\ \mu m > L_a > 22\ \mu m$, and $50\ \mu m > L_c > 30\ \mu m$, ice crystals with skeletal structure on both the $\{10\bar{1}0\}$ and {0001} faces grow.

However, when $100\,\mu m > L_a > 22\,\mu m$ and $L_c > 50\,\mu m$, ice crystals with skeletal structure on the $\{10\bar{1}0\}$ faces and optically smooth $\{0001\}$ faces grow. When $L_a > 100\,\mu m$ and $L_c > 50\,\mu m$, ice crystals with optically smooth $\{10\bar{1}0\}$ and $\{0001\}$ faces grow.

Furthermore, the instability region of the $\{10\bar{1}0\}$ faces of the ice crystals is larger than that of the $\{0001\}$ faces of the crystals.

5. Discussion of Results

It is characteristic of single ice crystals growing at low temperatures that plate-like and column-like ice crystals grow simultaneously under the same environmental conditions. The growth frequencies of plates and columns depend on both supersaturation and crystal size; moreover, as shown in Fig. 5, the ratios of the axial lengths c/a of these ice crystals depend on both supersaturation and crystal size, too.

Moreover, the c/a of the ice crystals growing in air at 1.0 atm at high supersaturation deviates from unity. This experimental result means that the morphology of ice crystals growing in air is affected by the volume diffusion of water vapor.

In the next place, as shown in Fig. 6, the interface stability of plate-like ice crystals growing in air at 1.0 atm at $-30°C$ depends on both supersaturation and crystal size. The qualitative reason is described in a previous paper (GONDA and KOIKE, 1982). Moreover, the instability region of the $\{10\bar{1}0\}$ faces of the ice crystals growing in this growth condition is larger than that of the $\{0001\}$ faces. The reason is that because of the effect of the crystal shape on the volume diffusion of water vapor, water vapor flux concentrates along the $\langle 10\bar{1}0 \rangle$ directions of the crystals rather than along the $\langle 0001 \rangle$ directions.

On the other hand, as seen in Fig. 4, long solid prisms predominantly grow at a supersaturation below about 2%. From the comparison of theoretical and experimental curves of normal growth rates versus supersaturation (KOIKE *et al.*, 1982), it is concluded that the growth mechanism of the $\{0001\}$ faces of long solid prisms which were experimentally formed is the screw dislocation mechanism. Therefore it is inferred that the $\{0001\}$ faces of long solid prisms observed in

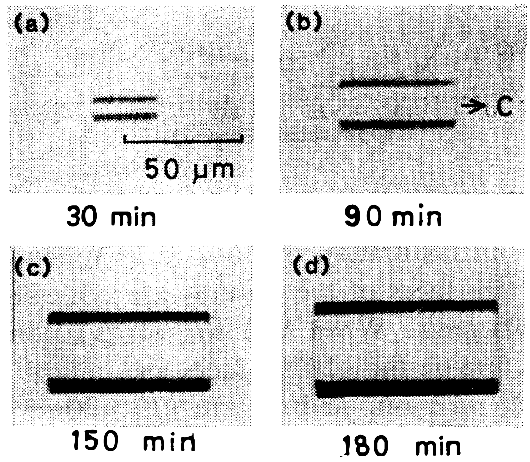

Fig. 7. Rectangular ice crystal grown in air at 1.0 atm at −30°C and a supersaturation of 1.0%.

Antarctica (SHIMIZU, 1963) grow by the screw dislocation mechanism at low supersaturation below about 2%.

Figure 7 shows an example of rectangular ice crystals (crystal with two remarkably developed prismatic faces) formed in air at 1.0 atm at $-30°C$ and a supersaturation of about 1.0%. When screw dislocations emerge only on the $\{0001\}$ faces of the crystal, the crystal grows preferentially along the $\langle 0001 \rangle$ directions as seen in Fig. 7(a). Here, in a certain stage of the growth process, when screw dislocations are introduced on the $(0,\bar{1},1,0)$, $(1,\bar{1},0,0)$, $(0,1,\bar{1},0)$ and $(\bar{1},1,0,0)$ faces, the normal growth rates of these faces increase. As a result, the areas of the two prismatic faces perpendicular to the paper remarkably increase, that is, the columnar ice crystal (a) is transformed to the rectangular ice crystal (d). Accordingly, it is inferred that the growth mechanism of the rectangular snow crystals observed in polar areas (HIGUCHI, 1968) is the same mechanism as that described above.

The diamond dust particles observed in Antarctica (KIKUCHI and HOGAN, 1979) are made up of plate-like and column-like ice crystals. This is characteristic of single snow crystals forming at low temperatures. From our experimental results (see Fig. 4), it is inferred that, when columnar ice crystals are predominantly observed, the supersaturation relative to ice is lower than about 2%, while when plate-like ice crystals are predominantly observed, the supersaturation is higher than about 2%.

6. Conclusions

In order to study the morphology of single snow crystals forming in polar areas and the growth mechanisms of such peculiar snow crystals as long solid prisms and rectangular snow crystals forming in these areas, ice crystals were formed in air at 1.0 atm at $-30°C$ and various constant supersaturations. The results which were obtained are as follows.

(1) The morphology of single ice crystals forming at low temperatures is determined in the stage of minute ice crystals, that is, at low temperatures, plate-like and column-like ice crystals are formed simultaneously in the same environmental conditions.

(2) The morphology of single ice crystals growing at low temperatures depends not only on temperature and supersaturation but also on crystal size and macroscopic surface structure.

(3) The interface instability of single ice crystals growing from the vapor is produced when the ice crystals are formed in air at high supersaturation; and the stability limit of the crystals is a function of both supersaturation and crystal size. The instability region of the $\{10\bar{1}0\}$ faces of plate-like ice crystals is larger than that of the $\{0001\}$ faces of the crystals.

(4) Long solid prisms (SHIMIZU, 1963) and rectangular snow crystals (HIGUCHI, 1968) observed in polar areas are explained by the introduction of screw dislocations on the $\{0001\}$ and $\{10\bar{1}0\}$ faces of the crystals growing at a supersaturation below about 2%.

(5) On diamond dust particles observed in Antarctica (KIKUCHI and HOGAN, 1979), it is inferred that when column-like ice crystals are predominantly observed, the supersaturation is lower than about 2%, while when plate-like ice crystals are predominantly observed, the supersaturation is higher than about 2%.

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