

Field and Laboratory Observations of Ice Crystal Growth from the Vapor

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ABSTRACT

The temperature dependence of the habit of ice crystals growing in a water vapor diffusion chamber is related to the growth rate of ice crystals in a supercooled cloud. The rates of growth occurring between -4 and -6°C as needles or between -12 and -16°C as dendrites may be in excess of those at intermediate temperatures by as much as a factor of 100. The effect of seeding a supercooled cloud will therefore depend critically on its temperature.

Growth of crystals at large supersaturation between -4 and -6°C takes place as spikes² growing along a direction 25° to the c axis. These crystals have been observed in the diffusion chamber, and also as frost near hot springs in Yellowstone Park. The molecular processes responsible for these habit changes are discussed.

1. Introduction

In recent years there have been a number of laboratory and field studies of the influence of temperature and supersaturation on the habit³ of ice crystals growing from the vapor. (Summary in Mason, 1957; Hallett and Mason, 1958; Kobayashi, 1961.) These investigations showed that the crystal habit depended markedly on temperature, with transitions between plate type (maximum development in the a axis direction) to prism type (maximum development in the c axis direction) taking place at -3 , -8 and -25°C . On the other hand, the transition from plates to dendrites and from prisms to needles was dependent on supersaturation, crystal size and the fall velocity of the crystal. The purpose of this communication is to describe some new observations of the growth of ice crystals from the vapor, to examine the implication of these results for the growth of crystals in the atmosphere and finally to suggest a possible interpretation of the observations on the molecular scale.

2. Experimental investigations

A study of ice crystals growing along a fiber suspended down the center of a diffusion chamber (Hallett and Mason, 1958) revealed, in addition to the changes of habit with temperature, that there existed three temperature zones where the growth took place more rapidly than elsewhere, either as hollow prisms or

needles (-4 to -6°C) or as thin plates or dendrites (0 to -2°C and -14 to -16°C). With supersaturation in excess of 3 per cent at -4°C and of 8 per cent at -15°C , the growth rates in the direction of a and c axes could differ by as much as a factor of 100. The transition from plates to dendrites or from prisms to needles depended on supersaturation only, and took place at about water saturation in the diffusion chamber. Outside these temperature zones, growth invariably took place much more slowly as prisms, plates or hopper crystals, even with a supersaturation in excess of 300 per cent. An estimate of the rate of mass increase of crystals at different temperatures was made from the dimensions obtained from photographs. Although only giving an accuracy of about 30 per cent, this revealed a striking difference in the rate of increase of crystal mass at different temperatures. The excess vapor pressure was calculated at each temperature from the temperature of source and sink of water vapor. Fig. 1 shows the relative rate of growth obtained when the excess vapor pressure was equivalent to that of ice crystals growing in a supercooled water cloud. The most striking feature of this relation is that a small change in temperature can make a large difference in the growth rate. In particular, a minimum in the growth rate exists at about -8°C . The dependence of growth rate on temperature differs significantly from that expected when the controlling factor is the excess vapor pressure of water over ice alone.

When the supersaturation in the diffusion chamber was increased beyond about 200 per cent at -4°C , it was found that a distinct change of crystal habit took place. The maximum growth direction then occurred at an angle of $25^{\circ} \pm 3^{\circ}$ to the c axis direction, while hollow prisms grew behind the tip in shielded regions

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² Not to be confused with spikes formed when the supercooled liquid freezes (Hallett, 1960).

³ Defined by the relative growth rates parallel to and perpendicular to the c axis.

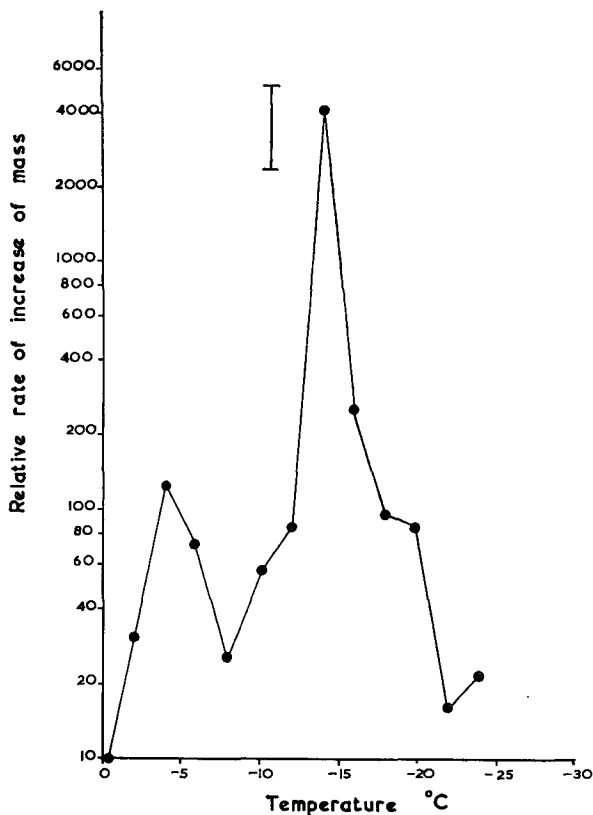


FIG. 1. Relative rate of mass increase of crystals growing at water saturation.

of lower supersaturation, (Fig. 2). Dendrites, (seen end on), have subsequently been grown on the ends of the prisms by lowering the crystal to -15°C to confirm the orientation of the spike. The spike did not lie in a simple crystallographic direction.

In nature, supersaturations as large as this occur only under very unusual conditions—for example, near hot springs when the air temperature is very low. Examples of these forms photographed in Yellowstone Park during 1962 are shown in Figs. 3–6. The air temperature was -30°C ; spring temperature $+10^{\circ}\text{C}$. These crystals grew overnight on thin grass stems projecting near the water surface. The formation of these crystals is shown schematically in Fig. 7. With increasing supersaturation, hollow prisms (Fig. 7a) grow on top of each other (Fig. 3) until the main line of growth is at an angle of 25° (Fig. 7b.) At even higher supersaturation, the only growth visible is in these directions, with the prisms occurring as a much smaller secondary growth, (Figs. 4, 5 and 7c). For each crystal, this gives 12 directions of high velocity growth (Fig. 7d) which means that the final crystal form may be very complicated, and will be three dimensional if shielding does not occur, (Fig. 6). It is also of interest that dendrites were observed growing at -1°C , a process which could never occur in a supercooled cloud in the atmosphere as the supersaturation at this temperature is only about 1 per cent (Fig. 8).

3. The influence of air flow

When growing crystals are moving with respect to their environment, the local concentration of water vapor near the crystal surface is enhanced. This causes the supersaturation dependent transitions—from plates to dendrites, and prisms to needles—to occur at a lower ambient supersaturation. If the crystal is in free fall, this transition also depends on its size and shape, in as far as this controls its terminal velocity, (Mason, 1953). In the diffusion chamber, where crystals are growing in an environment almost at rest, this transition takes place at about water saturation. Crystals which are falling in a supercooled cloud, on the other hand, readily develop as needles or dendrites at the appropriate temperature, indicating that this transition takes place at an ice supersaturation substantially less than that equivalent to water saturation.

4. Growth rate of crystals in a supercooled cloud

Formally, we may describe the growth rate of a crystal by an equation of the form

$$\frac{dm}{dt} = 4\pi DFAC\Delta\rho, \tag{1}$$

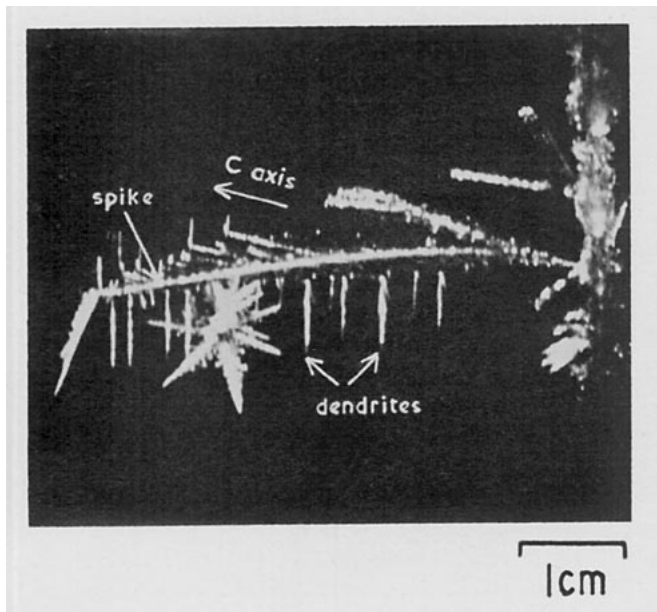


FIG. 2. A spike grown at -4°C in a water vapor diffusion chamber, supersaturation 250 per cent, showing a maximum growth rate in a direction at 25° to the *c* axis. The glass fiber suspension appears on the right.

FIGS. 3-8. Ice crystals growing near a thermal spring, Yellowstone Park, 28 January 1962.

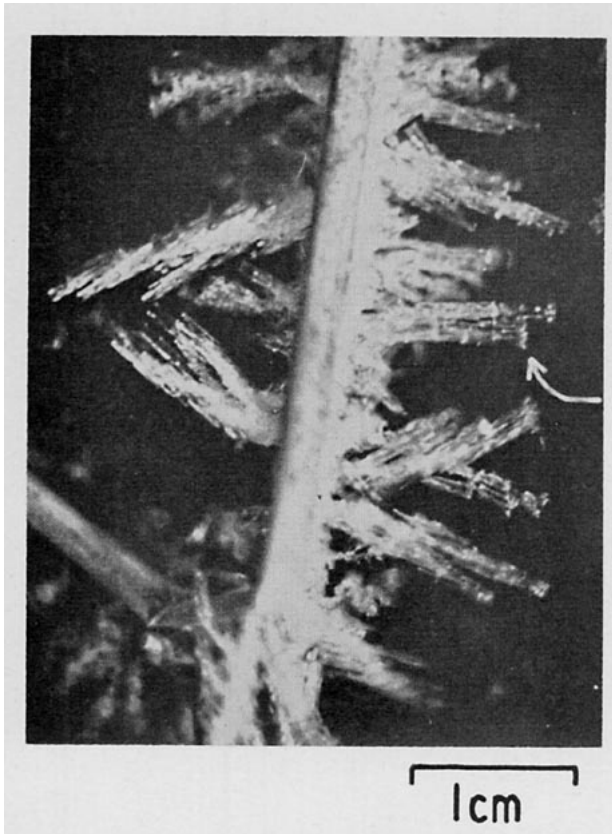


FIG. 3. Bundles of prisms, a prelude to spike growth, at intermediate saturation, with plates just beginning to develop on the ends as the local air temperature rose to about -2°C .

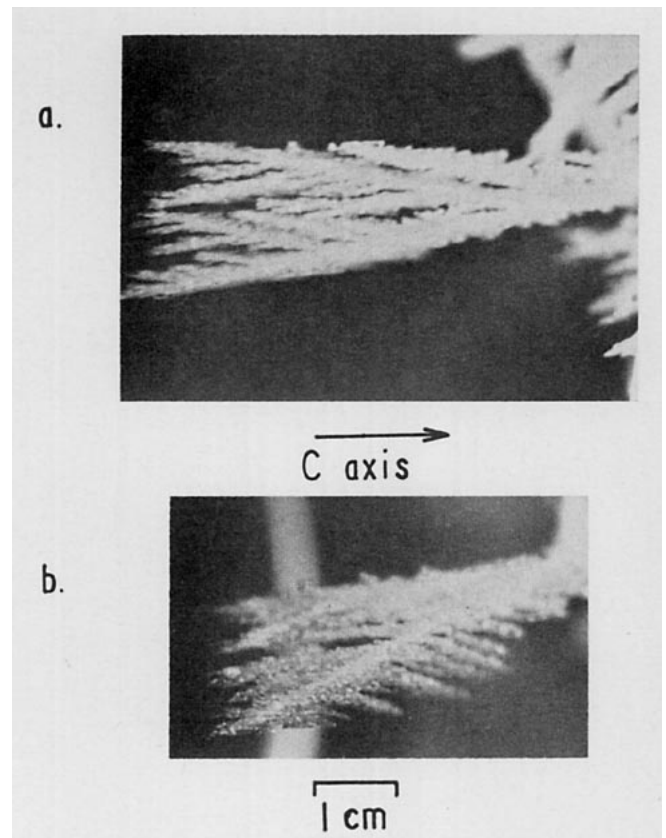


FIG. 4. a, b. Photographs of the same crystal at 90° , showing spike growth along two of the twelve possible directions of Fig. 7d. There is also subsequent prism development parallel to the c axis.

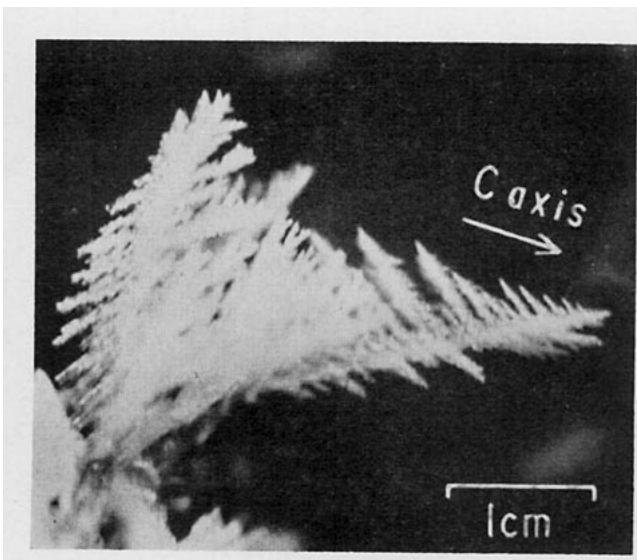


FIG. 5. At larger supersaturation, spike development occurs in more directions and dominates the growth form; here, on the right, growth is occurring with angles of 130° between the spikes. The arrow gives the direction of the c axis of this crystal.

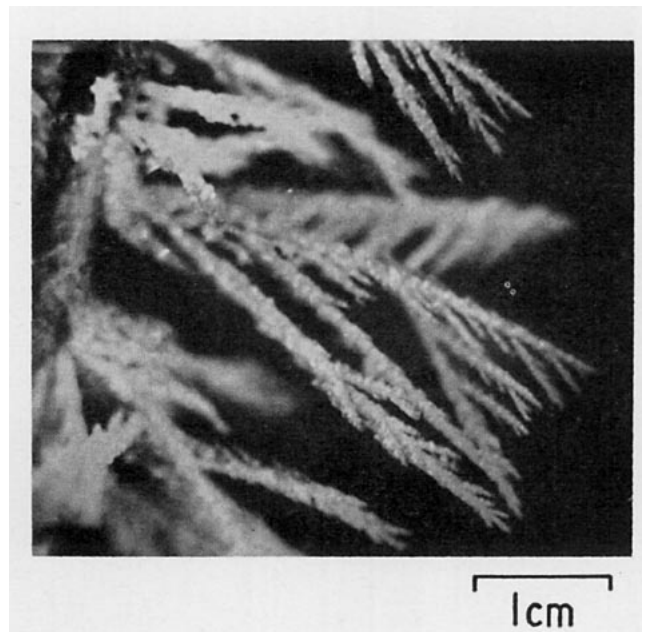


FIG. 6. An extensive spike array.

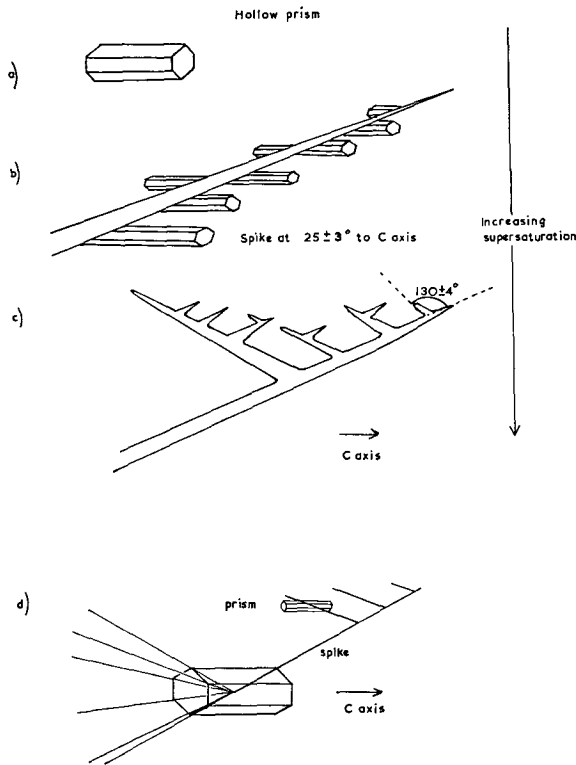


FIG. 7. Change of ice crystal habit at -4°C with supersaturation.

where

- m = crystal mass,
- t = time,
- D = molecular diffusion coefficient of water vapor in air,
- F = ventilation coefficient: a function of the Reynolds number of the flow, crystal shape and its orientation to the air flow,
- A = accommodation coefficient: a function of temperature and crystal face,
- C = the electrostatic capacity of the crystal
- $\Delta\rho$ = excess vapor pressure

together with a similar equation for heat flow.

If the crystal is falling freely

$$dt = Udz$$

U = terminal velocity, depending on the same parameters as F

Z = height.

Solutions of this equation have only been obtained under simplified conditions, assuming the crystal shape to be that of a disc or ellipsoid and an accommodation coefficient of unity (Houghton, 1950; Mason, 1953). A detailed solution for a real crystal is obviously a difficult task, as we have inadequate knowledge of most of the terms involved. Qualitatively, however, it can be seen that, since the limits of integration of (1) will

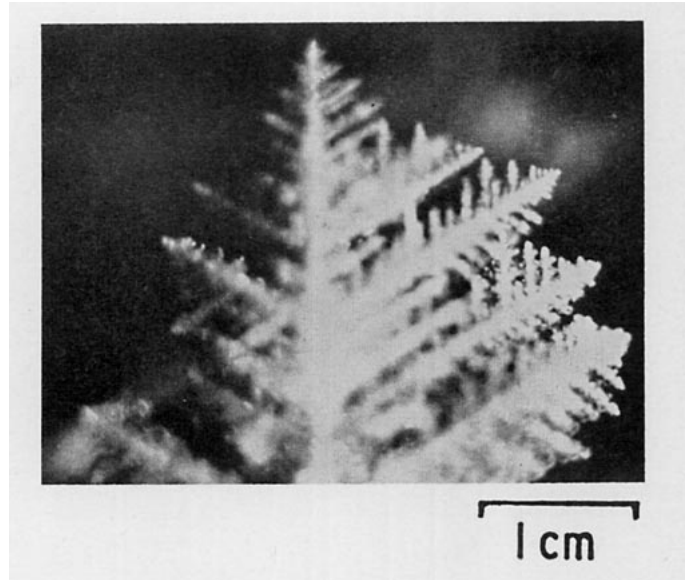


FIG. 8. Dendrites growing at about -1°C .

extend over the a definite depth of the atmosphere, the final crystal mass will be strongly influenced by the inclusion of a rapid growth zone. This will have two effects: first, the absolute growth rate of the crystal will increase and second, since the mass will be distributed over a much larger area, its fall speed will increase much more slowly, a greater time will be available for growth and the final size of the crystal will be correspondingly greater. The crystal form which finally falls from the cloud base will, of course, reflect growth over the whole range of temperature of the cloud; most of the crystal, however, will have grown in those regions where the growth rate is large, so that its form will be typical of these temperatures. For example, a crystal which grew in a supercooled cloud extending from -20 to -10°C would begin growing as a small plate between -20 and -16°C , then grow rapidly as a dendrite between -16 and -12°C , and finally grow small plates on the end of the dendrite branches between -12 and -10°C . The effect of its motion will be to enhance the growth rate differential still further, by causing dendritic or needle growth to be more pronounced.

Similarly, the effect of seeding a supercooled fog would depend very much on temperature; a fog with temperature -4 to -6°C would be expected to produce a much greater effect than if the temperature lay between -6 and -8°C . A striking example of this process was observed near Salt Lake City, Utah, on Saturday, 3 February 1962. The area had been covered by a supercooled fog for several days. At about 1030 MST (local time) the surface temperature was -4°C , visibility 30 m and surface wind ~ 2 kts., northerly. It was estimated that the fog was ~ 1000 ft thick with top temperature of -6°C . A car with a basket containing 50 lb of dry ice was driven around a circuit in the

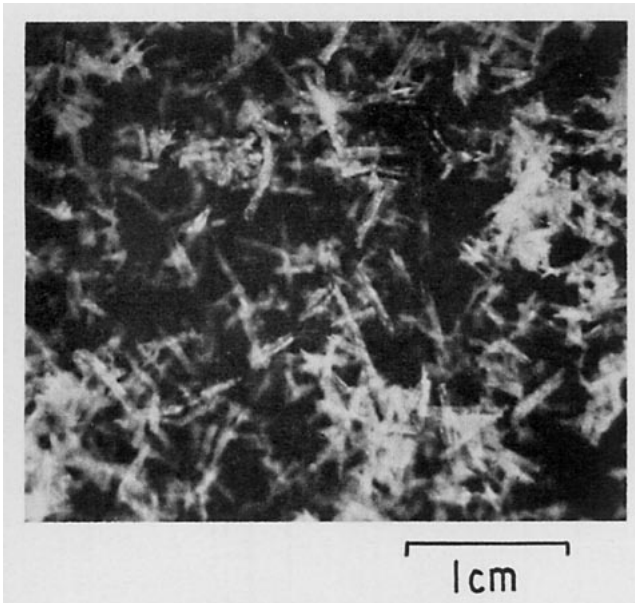


FIG. 9. Snow crystals fallen from a supercooled fog at -4°C , 15 minutes after seeding with dry ice. 1145 MST, Airport terminal, Salt Lake City Utah, 3 February 1962.

neighborhood of the airport terminal for 15 minutes, when a very slight precipitation was noticed. (No precipitation had occurred in the previous two hours.) This was 1120 local time. The precipitation increased, the particles increasing in size, until after a further 15 minutes, needles of length 3 mm and diameter 0.2 mm were falling, and continued for a further five minutes. There followed an increase of visibility to 200 yds, and momentary breakthrough of the sun. The needles are shown in Fig. 9, and a general view of the snowfall in Fig. 10. Needles are characteristic of growth in the region -4 to -6°C . The extent of the snowfall was about 1 mile downwind of the airport terminal, with no sign of snow upwind of the seeding site. A very similar result was obtained by Isono *et al.* (1956) after seeding a supercooled fog at -4.4°C . Seeding a fog with a temperature range of -6 to -8°C would be expected to produce much less spectacular results.

If we apply these ideas to the growth of crystals in a supercooled cloud in the laboratory (Aufm Kampe *et al.*, 1951; Mason, 1953), results will depend on whether or not any part of the cloud has a temperature at which rapid growth can take place. The coldest temperature will be in the neighborhood of the cooling pipes and the warmest at the top of the chamber, easily giving a difference of 5°C . The crystal type which first appears will be characteristic of the temperature at which growth rate is highest; crystals observed subsequently may have a different form.

5. Mechanism of crystal growth

Two recent papers have discussed possible molecular processes responsible for the changes of habit (Hallett,



FIG. 10. The extent of the snowfall can be seen as a light covering on this ice surface.

1961; Mason *et al.*, 1963). We will now examine the preceding results in the light of these ideas.

Growth processes must be consistent with the following observations:

1. Crystals growing in the direction of the basal plane have been observed to grow to a diameter of 100μ without thickening, providing that the supersaturation does not exceed a critical value, ~ 100 per cent.
2. The crystal grows in a particular direction by the propagation of thin layers at 90° to this direction; these may 'bunch' to produce thick ($\sim 1.0\mu$) visible layers.
3. Growth of these layers takes place by collection of molecules by a process of surface diffusion from a distance x on each side of the layer. Values of x have only been measured on the basal plane.
4. Molecules which impinge on flat regions of the crystal at a distance significantly greater than x from the growing edge will fail to be built into the crystal in spite of the excess saturation; these regions of the crystal will have accommodation coefficient of zero whilst others near the edge will have a value of unity.
5. Measurements of x show that on the basal plane it is strongly temperature dependent and is $\sim 6\mu$ at -10°C . The growth rate of a particular crystal face will therefore depend on the migration distance and the spacing of steps. (If the mean spacing of the steps is less than x , the growth rate is obviously independent of both.)
6. Hopper crystals develop when the supersaturation is greater at crystal corners than at the center of the face.

It is suggested that those crystal forms which occur where the habit is not excessive—prisms or plates—propagate only by growth at imperfections introduced into both crystals faces during growth, and that the distance between layers is usually sufficiently great to make growth by surface diffusion dominant. For slow

growth the corners are not greatly favored and solid forms result; for more rapid growth, the corners receive vapor preferentially, and hopper crystals develop with a shape characteristic of the temperature and the migration length on each surface. As the local supersaturation increases beyond a critical value, so the growth of a particular crystal face occurs by two-dimensional nucleation—this occurring first at the corners. The growth is now no longer dependent on the presence of steps on the crystal. Unfortunately the concentration of surface nuclei on prism or basal face is not easy to calculate. It will be influenced by surface energy and latent heat for the transition between the surface 'layer' and the solid, by temperature and also by the value of x for the particular face. When growth has begun at a corner this site is favored for further growth because of the diffusion field; a thin needle or dendrite arm results, with subsequent growth behind the tip being characteristic of the layer growth at lower supersaturation.

In the rapid growth zones, the growth direction was either parallel to a or c axes (low index faces) so that surface nucleation was occurring on only one plane. If, however, we increase the supersaturation sufficiently, surface nucleation would be expected to take place on *both* faces and this may be the cause of spike growth along a direction 25° to the c axis discussed in section 3. Layers are nucleated at the tip, grow a short distance, and then new layers appear at the tip as the supersaturation again builds up—the process suggested in Mason *et al.* (1963).

7. Conclusion

The ideas presented concerning the habit variation mechanism are necessarily speculative. It would be an interesting verification of these ideas to observe growth from the vapor on an ice substrate with known imperfection densities, and, in particular, to discover what changes in growth occur when the migration distance becomes comparable with the distance between steps resulting from the imperfections. If the suggested mechanism for rapid growth is correct, the calculation made from Eq. (1) of the growth rate of crystals in a

supercooled cloud would be in better agreement with observation in rapid growth zones than elsewhere. Investigations so far have not produced measurements of growth rate where temperatures have been outside rapid growth zones for the entire period of growth, and this comparison has not been made. It is hoped that this discussion will stimulate more intensive investigations of the result of seeding supercooled clouds when nature provides well defined temperature conditions.

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