The Influence of Temperature and Supersaturation on the Habit of Ice Crystals Grown from the Vapour
Author(s): J. Hallett and B. J. Mason
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frequencies apparently disposes of three out of seven proposed models. However, the predicted dichroism for the second modification of the Bernal–Fowler model and for the Rundle model is so small in each case that it could have escaped detection. Only the Owston model can be excluded by the present analysis.

REFERENCES (Ockman & Sutherland)


The influence of temperature and supersaturation on the habit of ice crystals grown from the vapour

BY J. HALLETT AND B. J. MASON

Imperial College, London

[Plates 8 to 10]

Ice crystals, growing on a fibre in a water-vapour diffusion cloud chamber under conditions in which the temperature and supersaturation of the environment can be varied independently, undergo the following transitions of habit in the temperature range 0 to $-30\,^\circ\text{C}$: plates $\rightarrow$ needles $\rightarrow$ hollow prismatic columns $\rightarrow$ plates $\rightarrow$ dendritic crystals $\rightarrow$ plates $\rightarrow$ prismatic columns. These changes of habit are controlled mainly by the temperature of the environment, large variations of supersaturation influencing only secondary features such as the development of dendritic forms. When crystals growing at a particular temperature are suddenly transferred into a different environment, further growth follows the habit characteristic of the new conditions; this allows the production of crystals of composite habit. In contradiction to a recent report by Nakaya, the crystal habit does not appear to be influenced by the presence or absence of atmospheric aerosols, but small quantities of organic vapours, e.g. camphor and isobutyl alcohol, modify the habit to an extent depending upon their concentration.

INTRODUCTION

The remarkable variety of shape and pattern exhibited by natural snow crystals poses some very interesting problems in crystal physics. In particular, it seems important to determine to what extent the growth rates and habits of the crystals are governed by such obvious environmental factors as temperature and supersaturation and to discover what other factors, such as crystal structure, may play.

In recent years ice crystals have been collected in aircraft from different types of clouds having widely different conditions of temperature and water-vapour concentration, and a correlation between the crystal habit and the environmental conditions has been established as shown in table 8, which is based largely on the observations of Weickmann (1947).
The rather striking changes of crystal form encountered in natural clouds at different temperatures have been reproduced in the laboratory, for example, in supercooled clouds formed by introducing steam into room-size cold chambers (Mason 1953). In these experiments, in which ice crystals, having been initiated at the top of the cloud by seeding with a small pellet of dry ice, were collected near the floor after 2 to 3 min growth, it was possible to determine the various transitions

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<th>Table 8. The predominant crystal forms occurring in different cloud types</th>
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<th>Table 9. Changes of crystal habit with temperature in artificially produced water clouds (Mason 1953)</th>
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<td>temperature range (°C)</td>
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of crystal habit with respect to temperature rather precisely. The results, summarized in table 9, reveal a threefold transition in habit—plates→prisms→plates (and stars)→prisms—as the temperature falls from 0 to about -25 °C. Such marked changes in crystal form, involving transitions from preferred growth along the principal (c)-axis for prisms, to preferred growth along the secondary (a)-axes for plates, and occurring in such a small temperature range, are believed to be peculiar to ice.

In the cold-chamber experiments, and as is usually the case in the atmosphere, the ice crystals grew in the presence of a cloud of supercooled droplets when the environment, being saturated with respect to liquid water, was supersaturated with respect to ice to a degree determined solely by the temperature. Under those conditions, therefore, it was not possible to determine whether the observed changes in crystal habit were being produced by changes in temperature or by changes in supersaturation since the latter were governed by the former.
In an initial attempt to resolve this point, Shaw & Mason (1955) studied the growth of ice crystals formed on a chilled metal surface under conditions in which the temperature and supersaturation could be varied independently and the growth of individual crystals kept under continued observation. Crystals grown at temperatures between 0 and $-40^\circ C$ exhibited variations of habit with temperature very similar to those shown in tables 8 and 9. Furthermore, by varying the supersaturation over quite wide ranges at fixed temperatures, it was established that, for the ice crystals grown on the metal surface, changes of crystal habit were controlled very largely by the temperature of the plate, the supersaturation having no systematic effect.

This result was at variance with the suggestion, made by Weickmann (1950) and by Marshall & Langleben (1954), that the crystal habit is principally determined by the excess of the ambient vapour density over that at equilibrium with the ice crystal at its own temperature, i.e. by the flux of vapour directed towards the crystal. It must be admitted, however, that conditions for crystal growth on the metal surface may not have fairly simulated those occurring in the free air, and it was to meet this point, and to study the whole question in more detail, that the present experiments were undertaken.

### 2. THE EXPERIMENTAL ARRANGEMENT

The crystals are grown on a thin nylon or glass fibre running vertically through the centre of a water-vapour diffusion chamber constructed on a principle similar to that described by Schaefer (1952). The chamber, 50 cm high, 30 cm in diameter, rests upon a solid aluminium block maintained at about $-60^\circ C$ by dry ice to serve as a heat sink of large thermal inertia. Cooled from below, with its top maintained either at or above room temperature, the chamber encloses a thermally stratified, convectively stable atmosphere in which steady-state conditions are readily achieved. Water vapour, evaporated from an extended surface of either water or ice (the source), diffuses downwards through the chamber towards an extended ice surface (the sink) held at lower temperature. The supersaturation regime is very largely determined by the temperatures of the water source and sink, the vertical separation of which can be varied.

The plane, rectangular walls of the chamber are made of Perspex. The outer plate-glass walls serve to reduce the lateral inflow of heat and to widen the vertical temperature gradient which is measured by a thermocouple attached to a micro-meter head. Lateral rotation of the thermocouple allows measurement of the horizontal distribution of temperature at any level. Once a steady-state is achieved, the horizontal temperature distribution in the chamber is very uniform except in the immediate neighbourhood of the walls; a typical vertical temperature profile is shown in figure 10, the separation of the 0 and $-40^\circ C$ levels being about 10 cm.

The chamber is illuminated by a parallel beam from a high-pressure mercury arc lamp, filtered to remove most of the radiant heat, and the crystals are viewed or photographed at right angles to the beam through a low-power microscope.
In one version of the chamber, the water source, sufficient for several days, is mounted on the underside of the top plate (see figure 10) and may be electrically heated. It takes the form of a large flat spiral of crimped metal foil which, after being immersed in water, allows the liquid to be retained by capillary forces in a network of small vertical cells. In such a chamber containing room air, condensation of water vapour upon the aerosol particles produces a dense cloud of tiny water droplets which freeze spontaneously on falling beneath the \(-40 \, ^\circ\text{C}\) level. In the presence of a persistent droplet cloud the air may be regarded as saturated relative to liquid water, the supersaturation relative to ice at any level being determined only by the temperature at that level. On the other hand, if the chamber is sealed and left for some hours, the aerosol particles, having become centres of condensation, are progressively removed by sedimentation leaving clean, highly supersaturated air in which condensation tracks produced by cosmic rays may be seen. Some variation of this very high supersaturation regime may be achieved by changing the temperature of the water source.

In order to simulate conditions in natural clouds, where ice crystals often grow at humidities below water saturation and therefore at supersaturations of only a few per cent relative to ice, the apparatus is modified as shown in figure 11. The crystals are now grown on a fibre suspended centrally between two parallel plane sheets of ice formed by the freezing of distilled water in shallow metal trays. The distribution of both temperature and supersaturation between the plates, the upper of which serves as a vapour source, is determined by their positions in the chamber which are adjustable. Further control is provided by electrical heating of the upper plate. A thermocouple passing through a hole in this plate allows the...
temperature distribution to be measured. Using both arrangements it is possible to grow crystals over the wide range of temperatures and supersaturations shown in figure 13.

![Diagram of cloud chamber](image)

**FIGURE 11.** The cloud chamber modified to produce low supersaturations.

### 3. Evaluation of Supersaturation in the Chamber

Since there appears to be no satisfactory method of measuring directly the supersaturation in the chamber without disturbing the environment, the experimental arrangement was designed so that this quantity might be calculated. In the central part of the chamber as modified to produce low supersaturations, we may regard the plates as two infinite parallel plane sheets of ice held at fixed temperatures $T_1$, $T_2$. Assuming a steady state, with the vapour being transported downwards solely by molecular diffusion (turbulence is strongly damped by the temperature inversion), and the diffusion coefficient to be independent of temperature, the diffusion equation predicts a linear profile of vapour density between the plates. The actual value of the vapour density at any level will be fixed by the boundary conditions; here we assume that the air immediately in contact with the two plates is just saturated with respect to the particular ice surface. Having obtained the vertical temperature profile by measurement, it is now possible to
calculate the supersaturation prevailing at each level at distances remote from the growing crystals. Figure 12 shows the results of a specimen calculation in which the distribution of supersaturation, computed relative to both ice and water, is displayed alongside the measured temperature profile. A more detailed calculation, which takes into account the variation of diffusion coefficient with temperature, shows the simplified treatment underestimates the supersaturation by only a few per cent. The effects of thermal diffusion would also appear to be negligible.

Figure 12. Specimen calculations of the supersaturation profile in the modified diffusion chamber based on a measured temperature profile. ..., supersaturation relative to water; --, supersaturation relative to ice; ——, temperature profile.

If, as sometimes occurs when the plates are several centimetres apart, the onset of a slow convection leads to an appreciably non-linear temperature profile, then it is reasonable to suppose that the associated vapour density profile will be of the same shape, and the supersaturation at each level between the plates may still be calculated from the measured temperature profile. The validity of this method of calculating the supersaturation was checked experimentally as follows. Having established, in clean air, a certain supersaturation regime between the plates, a small quantity of sodium chloride aerosol was introduced into the chamber. On occasions when the humidity everywhere between the plates was calculated to be
below water saturation, the aerosol particles remained invisible, indicating that very little growth had occurred. When, however, the humidity was predicted to surpass water saturation at a particular level, a droplet cloud was observed, its sharp upper boundary being located very close to the level calculated.

In the original chamber the much higher supersaturations in clean air were calculated in a similar manner using, this time, the temperature of the water source as an upper boundary condition. With such large separations of source and sink, it is very difficult to suppress convection completely and therefore the higher values of supersaturation indicated in figure 13 should be regarded as only approximate. However, the location of cosmic-ray tracks indicated that the calculated values of the highest supersaturation ratios were not in error by more than 20%.

![Figure 13](image_url)

**Figure 13.** The growth habits of ice crystals in relation to the temperature and supersaturation (relative to ice) of the environment.

4. **The correlation of crystal habit with temperature**

In a preliminary investigation of the influence of environmental conditions on crystal habit, the crystals were allowed to grow by sublimation on a thin nylon fibre hanging vertically in the centre of the chamber. For the most part, the crystals grew under conditions of high supersaturation achieved by removing the condensation nuclei from the air by sedimentation (see §2).

Figure 14, plate 8, shows a general view of the variation of crystal habit along a part of the fibre over which the temperature varied from −2 to −16 °C and the saturation ratio* everywhere exceeded 2:0. The crystal habit changes sharply at

* Defined as the actual vapour pressure relative to the equilibrium vapour pressure over a plane surface of ice at the same temperature.
Habit of ice crystals grown from vapour

-3 °C from thin hexagonal plates (shown in more detail in figure 15, plate 8) to the needles and hollow prisms seen more clearly in figure 16, plate 8. The needles are an exaggerated form of hollow prism in which both the prism and basal faces are often incomplete. They are formed only if the supersaturation exceeds about 5% (see figure 13). In figure 16 the needles give way, at -6 °C, to the much more compact and perfect hollow prisms, the detailed structure of which can be seen in figure 17, plate 8.

A reversion from hollow prisms to plates occurs at about -8 °C (figures 14, 16), the transition sometimes being effected via hexagonal scrolls and cups which will be described further in §7. The plate regime is interrupted between about -12 and -16 °C by the pronounced dendritic growth shown in figures 14, 19, plate 8, this being very reminiscent of the richly branching star-shaped crystals which occur at similar temperatures in natural snow. The plate-like development, which is resumed at about -16 °C, continues until about -25 °C where there occurs a final changeover to hollow prismatic columns (figure 20, plate 9), this transition being much less sharp than those just described for higher temperatures. The prisms occur at all lower temperatures down to -50 °C, the lower limit of the present experiments.

5. THE INFLUENCE OF SUPERSATURATION ON CRYSTAL HABIT

In order to distinguish between the possible influences of temperature and supersaturation on the habit of ice crystals, a series of experiments were performed in which crystals were grown in a particular temperature range but under widely different degrees of supersaturation—varying from a few per cent to about 300% relative to a plane ice surface. The procedure was then repeated at a number of different temperatures, particular attention being given to the transition zones separating one crystal form from another.

On the basis of these observations it was possible to allocate quite well-defined zones of a temperature-supersaturation diagram to particular crystal forms. Such a diagram, shown in figure 13, shows that large variations of supersaturation do not change the basic crystal habit, i.e. a change from a basically plate-like crystal to a prism, or vice versa although, of course, the growth rates are profoundly effected. In other words, the transitions between one crystal form and another occur at the same temperatures irrespective of the degree of supersaturation. This point is illustrated in figures 18, 16, 21, plates 8 and 9, which show the transition from prisms to plates to occur within a degree of -8 °C with supersaturations ranging from about 100% to only 5%. Similar results were obtained at -3 °C, where the changeover from plates to prisms usually occurs within one half of one degree Centigrade. Although the final transition from plates to prisms was less clear-cut, being spread over a few degrees around -25 °C, again large changes in supersaturation produced no systematic effect.

Although we may conclude from these observations that the degree of supersaturation does not determine the basic habit of ice crystals, we see from figure 13 that needles, and cups, sector plates and the dendritic forms are produced only if
the supersaturation surpasses certain limits which, in these experiments, corresponded roughly to water saturation. These secondary features of crystal development, which are supersaturation-dependent, will now be discussed in more detail.

6. Doritic and dendritic growth

There are three interesting features of crystal development which appear to be markedly influenced by supersaturation. They are the needle-like extensions of hollow prisms, the development of spikes and segments at the corners of hexagonal plates, and the fern-like development of the star-shaped crystals. We propose to call the first two of these doritic (spear-like) growth, reserving the term dendritic to describe the branching structure illustrated in figure 19, plate 8.

Crystals growing in our chamber at temperatures between \(-12\) and \(-16\) °C reacted in the following manner to changes of supersaturation. Once the latter exceeded a certain value, which depended upon the temperature, the corners of hexagonal plates developed segmental extensions (see figure 22, plate 9) to form what Nakaya has called sector plates. Higher supersaturations, causing preferential growth of the corners, produced, first of all, long spear-like growths which thicken only slowly. At higher supersaturations still, these developed lateral growths, usually at 60° to the \(a\)-axes, to form the branching structures typical of the larger snow crystals. Although this pronounced dendritic growth occurs only if the supersaturation exceeds about 20% relative to ice, it also appears to be restricted to the temperature range \(-12\) to \(-16\) °C, as shown in figure 23, plate 9. This progressive development from doritic to dendritic growth is undoubtedly exhibited by natural snow crystals, but there the changes occur at rather lower supersaturations than those indicated in figure 13. This we attribute to the fact that dendritic growth is encouraged by the temperature and vapour density gradients in the vicinity of a falling crystal being enhanced by its motion relative to the air, in the manner described by Mason (1953). Plates formed at temperatures between 0 and \(-3\) °C also exhibited dendritic growth if the supersaturation exceeded about 80%.

The development of doritic growth from the corners of plates, shown in figure 20, plate 9, was particularly marked at temperatures between \(-20\) and \(-25\) °C.

The needle-like crystals, which are a skeletal form of hollow prism, occurred predominantly at temperatures between \(-3\) and \(-5\) °C and at supersaturations in excess of 10%. Typical examples are shown in figure 16, plate 8. They arise as the result of rapid growth along the principal axis; not only are the basal faces incomplete, but growth may occur only on some of the edges of the basal plane or, in extreme cases, only from one or more corners. From the external forms alone, it is not difficult to confuse some of the doritic growths, which develop from the corners of a small hexagonal plate along the \((1\overline{1}20)\) direction, with needles growing along the principal axis. When in doubt, we identified the particular form by moving it into a different temperature region and observed the subsequent development as described in the next section.
7. Metamorphic and transitional crystal forms

The effect of suddenly changing the temperature and supersaturation on the growth form of a particular crystal could be observed after raising or lowering the fibre in the chamber. Whenever a crystal was thus transferred into a new environment, the continued growth assumed a new habit characteristic of the new conditions.

Needles were grown at temperatures between $-3.5$ and $-5.5 \, ^\circ C$ and at high supersaturation; on being moved up in the chamber, to about $-1.5 \, ^\circ C$, plates developed on the very ends of the needles (figure 24, plate 9), which thereafter showed little further thickening. When similar needles were lowered to about $-14 \, ^\circ C$, they gave way to star-shaped crystals as shown in figure 25, plate 9.

Figure 26, plate 10 shows a quadruple transition; needles, grown at $-4 \, ^\circ C$, were transferred to $-13 \, ^\circ C$ where plates developed on their ends, then to $-8 \, ^\circ C$ where the plates developed into cup-shaped crystals and finally, back to $-4 \, ^\circ C$, where needles grew from the corners of the cups. Figure 27, plate 10, illustrates a similar sequence of changes but based, this time, upon a long protuberance from the corner of a hexagonal plate grown at $-13 \, ^\circ C$.

The effect of transferring a well-developed dendritic crystal grown at $-15 \, ^\circ C$ into the plate-like region at $-2 \, ^\circ C$ is shown in figures 28a, b, plate 10; the ends of the crystal branches now develop as plates, this transition being a common feature of natural snow crystals. It may be induced either by changing the temperature or by reducing the supersaturation (see §5).

In order to determine whether there are any essential structural differences between the prismatic crystals growing at about $-6 \, ^\circ C$ and those appearing below $-25 \, ^\circ C$, prisms formed initially at $-30 \, ^\circ C$ were transferred into the higher temperature zone. On most occasions the new growth grafted perfectly on the original form as shown clearly in figure 29, plate 10, but, in some cases, new prisms grew only from the corners of the old.

These are only some examples of metamorphosis which were observed when crystals were suddenly transferred to a new environment; in fact, combination forms of all the basic crystal types shown in figure 13 are readily produced in this way.

A very interesting crystal form sometimes develops at temperatures between about $-6$ and $-10 \, ^\circ C$. It takes the form of either a hexagonal cup or a scroll and appears to represent an intermediate form between the prism and plate-like habits. Both scrolls and cups are essentially skeletal prisms with ‘hopper’ development of both prism faces (rectangular) and basal faces (hexagonal), as shown in figure 30, plate 10. It seems that they grow in both diameter and depth by the addition of thin rod-like crystals to the edges of a small central hexagonal prism. Sometimes the growth is symmetrical, the hexagonal edges are complete, and the crystal takes the form of a cup. On other occasions, they show a spiral (or scroll) formation and closely resemble the very large crystals of this type described by Mason & Owston (1952) as growing at $-10 \, ^\circ C$ on the cooling pipes of a cold store. The edges of the spirals are accurately parallel and the angles between neighbouring
faces are accurately 120°. Such a development might be initiated by some prism faces growing more slowly than others, perhaps through being partially shielded from the vapour by neighbouring crystals, so that the hexagonal edges of the basal planes do not join up. Scrolls are therefore more likely to grow in stagnant than in moving air; they are not observed among natural snow crystals but are found in ice caves and among frost crystals growing in crevasses.

Occurring at temperatures in the transition zone between plates and hollow prisms, the cups and scrolls exhibit development along both the c- and a-axes, but they become progressively more shallow at the lower temperatures.

8. THE INFLUENCE OF AEROSOLS AND FOREIGN VAPOURS ON CRYSTAL HABIT

Nakaya (1955) has reported that the presence of aerosols in the air may profoundly affect the habit of ice crystals growing from the vapour. Thus, when he replaced room air in his chamber by air from which all suspended particles had been removed by a cotton-wool filter followed by a thermal precipitator, crystals growing in the temperature ranges normally associated with plates and dendritic crystals now appeared as prisms or needles.

We have been unable to confirm this observation. In the present experiments, the aerosol was removed by sealing the diffusion chamber and allowing it to ‘rain out’. Crystals grown in these conditions, in droplet-free air, exhibited a very similar variation of habit with temperature as was obtained in the presence of a droplet-cloud formed by condensation on atmospheric aerosol. However, the appearance of needle-like crystals, at all temperatures, along the whole length of the fibre, was observed when a small trace of camphor vapour was accidentally introduced into the chamber. The result is shown in figure 31, plate 10. Rather similar effects have been obtained with traces of other organic vapours, for example, isobutyl alcohol, thus supporting an observation by Vonnegut (1948) that a cloud of thin hexagonal ice plates, produced by the seeding of a supercooled cloud, became transformed into prisms by the addition of butyl alcohol at about 10⁻² mb. partial pressure.

DESCRIPTION OF PLATE 8

FIGURE 14. The variation of crystal habit along a part of the fibre over which the temperature varied from −2 to −16 °C and the saturation ratio everywhere exceeded 2·0. The sequence is plates → needles → hollow prisms → plates → dendrites. Each stage is shown in more detail in the following photographs.

FIGURE 15. Thin hexagonal plates growing between −0·8 and −2·0 °C.

FIGURE 16. The sharp transitions between needles and hollow prisms at −6 °C and between prisms and plates at about −8 °C. These crystals were grown at medium supersaturations (ca. 15%).

FIGURE 17. A large hollow prism growing at −6·5 °C.

FIGURE 18. The transition from hollow prisms to plates at about −8 °C under conditions of high supersaturation (ca. 100%).

FIGURE 19. Well-developed dendritic growth at −16 °C under very high supersaturations.
DESCRIPTION OF PLATE 9

FIGURE 20. The transition from plates to hollow prisms at about −25 °C. The spear-like growth from a corner of a hexagonal plate is a common feature at these temperatures.

FIGURE 21. The transition from hollow prisms to plates at −8 °C under conditions of low supersaturation (ca. 5%).

FIGURE 22. Sector plates growing between −10 and −14 °C at medium supersaturations.

FIGURE 23. Dendritic growth is restricted to the temperature range −12 to −16 °C even under very high prevailing supersaturations.

FIGURE 24. Needles grown at about −4.5 °C developed plates on their ends when transferred into a new environment at about −1.5 °C.

FIGURE 25. Needles developed stars on their ends when transferred to the −14 °C region of the chamber.
Figures 26 to 32.
Habit of ice crystals grown from vapour

We have observed that, in the temperature range $-12$ to $-16$ °C, the normal dendritic habit is replaced respectively by plates, hollow prisms (see figure 32, plate 10), plates again, and finally by a malformed type of dendritic growth as the concentration of alcohol is increased from about $10^{-4}$ to 1 mb. partial pressure.

9. Discussion of Experimental Results

The pattern of crystal habit in relation to the temperature and supersaturation of the environment, shown in figure 13, is similar in many respects to that found by Nakaya (1951, 1954). He performed a large number of experiments in each of which one single crystal, supported on a short fibre, was allowed to grow in a rising stream of moist air which cooled as it ascended. Unfortunately, with this experimental arrangement, the conditions were not steady nor well defined because the strong convection in the apparatus gave rise to large fluctuations in both temperature and supersaturation. The average temperature of the air in the neighbourhood of the crystal was measured with an alcohol-in-glass thermometer, but only crude estimates of the supersaturation were possible.

The present observations, which cover a much wider range of supersaturation and extend to considerably lower temperatures, show the following differences from those of Nakaya: (i) the hexagonal plates which we observe between 0 and $-3$ °C are not reported by him; (ii) we observe hollow prisms rather than needles between $-5$ and $-8$ °C; (iii) we do not find prismatic columns occurring at temperatures between $-10$ and $-15$ °C when the supersaturation is low; (iv) we do not find that the presence of aerosol or water droplets in the air is an important factor in determining the crystal habit.

Although Nakaya was inclined to the opinion that the temperature rather than the supersaturation was the main factor controlling the crystal shape, Marshall &

Description of Plate 10

Figure 26. Crystals showing multiple changes of habit viz: needles→plates→hexagonal cups→needles as the result of being moved successively from $-4$ °C→$-13$ °C→$-8$ °C→$-4$ °C.

Figure 27. A crystal growing originally as a protuberance from the corner of a hexagonal plate at $-13$ °C developed a hexagonal cup when moved to $-8$ °C and, when moved later to $-4$ °C, needles grew from each corner of the hexagonal cup.

Figure 28(a), (b). Dendritic crystals grown at $-15$ °C develop plates at the ends of their branches when moved to $-2$ °C.

Figure 29. The grafting, at $-7$ °C, of hollow prisms on ‘low temperature’ prisms grown at $-30$ °C (see arrows).

Figure 30. Hexagonal scrolls growing at $-12$ °C.

Figure 31. The effect of introducing a small quantity of camphor vapour into the chamber; the crystals now assume a needle-like form at all temperatures between $-5$ and $-22$ °C.

Figure 32. The dendritic growth, which is normally characteristic of the temperature range $-12$ to $-16$ °C (see figure 23), is replaced by hollow prisms in the presence of isobutyl alcohol at about 1/100 mb partial pressure.
Langleben (1954) interpreted his results as showing that this is principally determined by the excess of the ambient vapour density over that which would be in equilibrium with the surface of the ice crystal, that is, by a quantity closely related to the supersaturation, and proportional to the flux of vapour directed towards the crystal. Marshall & Langleben hypothesize that resistance to growth will be greatest at the crystal corners and greater on the prism faces than on the basal faces, so that growth of the corners and the prism faces will occur only when the excess vapour density \( \Delta \rho \) becomes sufficiently large to overcome these inhibitions. Thus, prismatic columns would be expected to develop at relatively low values of \( \Delta \rho \), plates only when \( \Delta \rho \) is relatively large, and the corners to develop (doritic and dendritic growth) only when \( \Delta \rho \) achieves very high values. That a minimum critical value of the supersaturation is required to start growth on any crystal face was established by Shaw & Mason (1955), but this varied in an apparently random manner from face to face and from crystal to crystal; there was no systematic difference between the critical values for the basal and prism faces as required by Marshall & Langleben.

The results of the present experiments, plotted in figure 13, indicate quite clearly that the temperature is of prime importance in determining the crystal habit, which is temperature sensitive at supersaturations far in excess of those required to overcome the initial resistance to growth on any crystal face. Very strong support is also provided by the fact that, although the basic shape of a crystal is not changed by varying the supersaturation at constant temperature, a degree or two change in temperature at constant supersaturation can produce a radical change.

At the present time it is not possible to offer a convincing explanation for the observed variations of crystal habit with temperature. Present theories of crystal growth contain few parameters which are likely to be strongly face-dependent and, at the same time, sensitive to temperature changes of only a few degrees. The detailed mechanism of ice-crystal growth is clearly a matter for further experiment.

10. Experiments with heavy water

Experiments very similar to those described in §5 have recently been carried out using heavy water (99% pure) as the source. The variation of habit with temperature is almost identical to that observed with ordinary water, but the transition temperatures are all shifted upwards by 3 ± 0.5 °C in conformity with the melting point of pure D\(_2\)O being 3.8 °C.

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For some comments by J. D. Bernal on this contribution see p. 534.
Lattice disorder and physical properties connected with the hydrogen arrangement in ice crystals

By H. GRÄNICHER

Physikalisches Institut der Eidg. Techn. Hochschule, Zürich, Switzerland

The present knowledge of the hydrogen positions and of the physical properties sensitive to the hydrogen arrangement in ice crystals is reviewed. All possibilities of configurational changes in the ideal and the real crystal are considered. It is shown that the observed time-independence of the ionic conductivity leads to the conclusion that ions (H$_3$O$^+$ and OH$^-$) and Bjerrum defects (doubly-occupied and vacant bonds) must be present simultaneously. The concept that the hydrogen configurations are changed only by the diffusion of such defects proved to be the basis for a consistent theoretical explanation of the electrical, mechanical and nuclear relaxation phenomena and of the thermal properties.

STRUCTURAL EVIDENCE

Ice crystals have attracted a great deal of scientific interest for a long time. Their unusual structural and physical properties present many fundamental problems whose successful investigation has only become possible by new experimental methods and by the modern concepts of crystal physics. An essential feature of ice crystals is the fact that they are held together by hydrogen bonds exclusively. The various efforts to determine the structure and especially the hydrogen positions in ice crystals have been summarized in this discussion (Lonsdale 1958; Ockman & Sutherland 1958). Only the most important steps in the historical development of our knowledge of the hydrogen positions need therefore be recapitulated.

Bernal & Fowler (1933) in their famous paper made the following three suggestions:

(a) the hydrogen atoms lie on the lines connecting neighbouring oxygen atoms;
(b) there is only one hydrogen atom on each such linkage thus forming hydrogen bonds;
(c) each oxygen atom has two hydrogen atoms at a short distance (0.99Å) and hence water molecules are preserved.

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