Interpretation of Crystallographic Orientations in Accreted Ice

P. J. Rye and W. C. Macklin

Dept. of Physics, University of Western Australia, Nedlands

(Manuscript received 12 September 1972, in revised form 16 May 1973)

ABSTRACT

Measurements of c- and c'-axis orientation distributions in accreted ice deposits formed in the dry growth regime show peaks which may be related to the growth conditions. Theoretical studies of the factors governing the development of the equilibrium crystallographic orientation distributions in such deposits have been made. These show that surface roughness affects the sharpness of the peaks in both types of distributions. In addition, in the c'-axis distribution the position of the peak is shifted and hence the peak cannot be uniquely related to the growth conditions. If inferences of the growth environment of hailstones are to be made from crystal orientation measurements, the orientation distributions of the c-axes relative to the local normal to the surface of the hailstone should be used as these give the maximum possible resolution of the peaks.

1. Introduction

Aufdermaur *et al.* (1963), Knight and Knight (1968, 1970) and List *et al.* (1970) have determined the crystallographic orientation distributions of ice crystals in natural hailstones. Aufdermaur *et al.* (1963) and Levi and Aufdermaur (1970) have also investigated the crystallographic orientation distributions in ice deposits formed by accretion in an icing tunnel. Two types of distributions have been ascertained. In one the number of crystals is measured as a function of the angle between the c-axes of the crystals and the growth direction. In the other the number of crystals is measured as a function of the angle between the projections of the c-axes of the crystals onto the plane of the ice section being examined, termed the c'-axes, and the growth direction. However, the use of such distributions to interpret hailstone structure has not yet proved particularly fruitful because the factors controlling the development of the distributions have been inadequately understood. The purpose of the present paper is to provide a better theoretical basis for the interpretation of such data.

2. Preferred crystallographic orientation for droplets freezing during accretion

When a supercooled droplet impinges on an accreting surface whose temperature is below 0C (i.e., dry growth), freezing takes place in two stages. Initially, a dendritic sheet grows rapidly through the droplet warming it to a temperature near 0C. Subsequently, the remainder of the droplet is frozen by conduction of latent heat to the underlying surface (see Macklin and Payne, 1967). For supercoolings larger than 2–3C the direction of growth of the dendrites is inclined at an angle to the basal plane, the angle increasing with supercooling (Macklin and Ryan, 1965; Pruppacher, 1967). Levi and Aufdermaur (1970) suggest that those droplets which freeze by chance with the initial dendritic sheet parallel to the accreting surface give rise to preferred orientations in the bulk ice. The reason for this is that the ice crystals so formed present the largest cross-sectional area to later impinging droplets which then take up this particular orientation by epitaxial growth. This gives rise to a peak in the crystallographic orientation distribution corresponding to the growth angle of the dendrites.

Levi and Aufdermaur found that the crystallographic orientation distributions attained equilibrium in a very small distance (a fraction of a millimeter) from the cylinder on which the ice was grown. This occurred even with droplet temperatures as high as −3C, indicating a relatively large probability of crystallographic reorientation. At these warm temperatures they found also that the crystals in the bulk ice were relatively large, some millimeters in length. As they note, it seems contradictory that large crystals are formed under the same conditions where the probability of reorientation is supposedly high.

It is shown in Section 5 that reorientation alone cannot give rise to the crystallographic orientation distributions observed by Levi and Aufdermaur. We now propose an alternative mechanism by which preferred orientations can be established in accreted ice in the dry growth regime. Consider a supercooled droplet impinging at the boundary between two crystals. Dendritic sheets grow into the droplet from a nucleus on the surface. The most likely location of

Unauthenticated | Downloaded 02/24/24 07:34 AM UTC

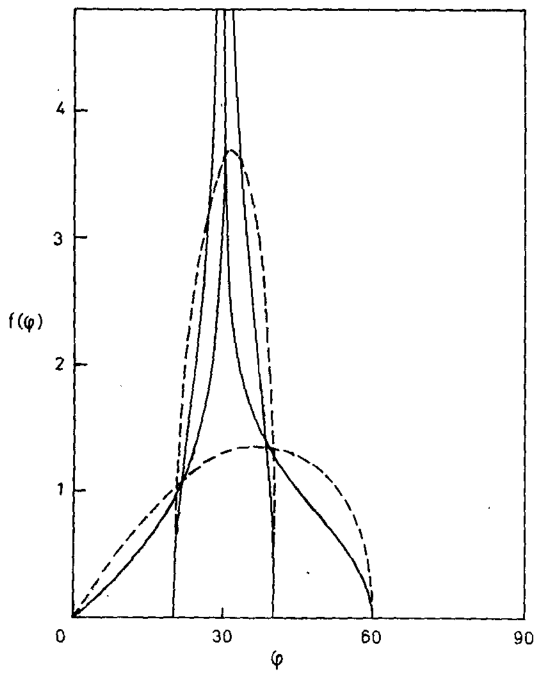

FIG. 1. Typical curves for $f(\phi)$ for $\psi=30°$ and $\alpha=10°$ and 30°. The continuous lines are for rough cylinders and the dashed lines for rough spheres.

the nucleus is at the crystal boundary, where the dendrites are equally likely to take the crystallographic orientation of either of the two crystals involved. During the second stage of freezing, the ice growing above the dendrites takes their crystallographic orientation. Consequently, the larger part of the frozen droplet is, in general, taken by the crystal from which the dendrite grows, and the fraction it takes increases as the angle between the dendrite and the surface decreases. The result is a migration of the crystal boundary. Consequently, the crystal whose growth is favored is the one whose initial dendrites lie parallel to the surface. After the accretion of a large number of droplets, an equilibrium situation is produced in which all c-axes are inclined to the normal to the surface at the angle between the growth direction of dendrites and the basal plane. This angle will subsequently be denoted ψ.

The selection mechanism described here requires an initial supply of random crystal orientations. In the case of accreting cylinders, this may come from one or both of two sources. First, the droplets which are initially accreted on a cylinder core freeze to some extent with random crystallographic orientations. Second, there is a probability that crystallographic reorientation takes place when a supercooled droplet impinges on an ice surface. Although measurements of this probability are not currently available, it can be easily shown that the present mechanism requires a much lower value than that of Levi and Aufdermaur. Calculations based on the crystal sizes near the cylinder core of Fig. 3a of their paper show that, in the absence of a selection process, attainment of crystallographic equilibrium in the distance measurable on the figure (0.5 mm) requires a reorientation probability of about 0.02. When the proposed selection process is operating, this probability is reduced to 0.002. Once crystallographic equilibrium is established, the crystals produced by further reorientation do not grow because their orientation is not favored (Knight, 1971).

This mechanism for producing preferred orientations in accreted ice gives the same result as that proposed by Levi and Aufdermaur. However, since it does not require a large probability of crystallographic reorientation at warm droplet temperatures, it is operative at all temperatures. Again a peak in the c-axis orientation distribution occurs at the growth angle ψ.

3. The effect of surface roughness on the crystallographic orientation distribution

To obtain theoretical crystallographic orientation distributions which may be compared with those obtained experimentally, we define two functions:

(i) The exact analogue of the measured distributions, termed $f(\phi)$. This is defined so that $f(\phi_1)d\phi$ is the probability that the angle ϕ between the c-axis and the overall growth direction, at any point in the accreted ice, lies in the range $\phi_1<\phi<\phi_1+d\phi$.

(ii)
$$h(\phi)=\frac{f(\phi)}{2\pi\sin\phi}. \quad (1)$$

If the c-axis orientation is axially symmetric about the overall growth direction, $h(\phi)$ is the c-axis orientation probability per unit solid angle.

In general, accreted ice surfaces are macroscopically rough. Levi and Aufdermaur (1970) note that such roughness has an effect on the crystallographic orientation distribution. To allow for this it is necessary to include an area distribution function which provides the weighting factor to allow for the relative contributions from small areas of the ice surface with normals inclined at various angles to the overall growth direction, and to transform the c-axis orientations from a coordinate system with the Z-axis normal to the surface to one with the Z-axis parallel to the overall growth direction. This may be readily done geometrically.

Simple models for the surface roughness have been used to determine $f(\phi)$. For rotating cylinders the surface undulations have been taken to be cylindrical in form with axes parallel to the axis of rotation, while for hailstones the roughness has been assumed to take the form of spherical caps. The surface roughness may then be characterized by a single parameter,

Unauthenticated | Downloaded 02/24/24 07:34 AM UTC

namely, the maximum angle between the local surface normal and the overall growth direction, termed α. Calculations of $f(\phi)$ have been made for a range of values of ψ and α and typical distributions are shown in Fig. 1. For both types of roughness the distribution is broadened and, in the case of hailstones, the sharpness of the peak is reduced. Due to the $\sin\phi$ term in the conversion from $h(\phi)$ to $f(\phi)$, the curves tend to zero as ϕ goes to zero. However, in the case of rough rotating cylinders, $f(\phi)\neq 0$ when $\psi=0$.

4. Distributions expressed as a function of c'-axis orientation[1]

Here the angle between the c'-axis and the overall growth direction is taken to be ζ, and the corresponding probability per unit change of ζ for a crystal with c'-axis aligned at ζ, is defined to be $F(\zeta)$. For hailstones, it may be shown that $F(\zeta)$ is a simple mathematical transform of $f(\phi)$, i.e.,

$$F(\zeta)=\frac{2}{\pi\cos\zeta}\int_{\zeta}^{\pi/2}\frac{f(\phi)\cos\phi d\phi}{(\cos^2\zeta-\cos^2\phi)^{\frac{1}{2}}}. \qquad (2)$$

For cylinders, $F(\zeta)$ can be derived geometrically:

$$F(\zeta)=\frac{1}{\pi}\left\{\sin^{-1}\left[\min\left(1,\frac{\tan(\alpha-\zeta)}{\tan\psi}\right)\right]+\sin^{-1}\left[\min\left(1,\frac{\tan(\alpha+\zeta)}{\tan\psi}\right)\right]\right\}. \qquad (3)$$

Fig. 2 shows the effect on the distribution of using c'-axes rather than c-axes in the measurement of crystal orientations. For both types of roughness the angle at which the mode of the distribution occurs and the sharpness of the peak in the distribution are reduced. For hailstones the reduction is about $\frac{2}{3}\alpha$ and for cylinders it is exactly α. Where α is greater than ψ, the distribution $f(\phi)$ transforms to an $F(\zeta)$ distribution with a broad peak at $\zeta=0$.

It is evident that measurements of crystallographic orientation distributions in terms of the c'-axes cause loss of much of the information available from the crystal structure. In particular, it is not possible to obtain quantitative estimates of the temperature of the droplets from which the accreted ice was formed.

5. The effect of crystallographic reorientation on the c-axis distribution[1]

It is possible that at relatively cold droplet and ice deposit temperatures the probability of crystallographic reorientation is sufficiently high to dominate the selection process described in Section 2. Reorientation must also be an important factor in the determination of crystal orientations when the dry growth selection mechanism is weakened as the wet growth limit is approached. A theoretical model has been developed on the basis of a set of discrete reorientation angles, such as postulated by Higuchi and Yosida (1966). The probability of reorientation of the c-axis of an accreted droplet to an angle of η_i from the substrate orientation is dependent on the angle ϕ between the substrate c-axis and the growth direction (Hallett, 1964). This probability is termed $p_i(\phi)$. It may be shown that the equilibrium distribution for $h(\phi)$ is then given by

$$\sum_{\eta_i\neq 0} p_i(\phi)h(\phi) =\frac{1}{\pi}\int_{-\pi/2}^{\pi/2}\sum_{\eta_i\neq 0} p_i[\cos^{-1}(\cos\eta_i\cos\phi+\sin\eta_i\sin\phi\sin y)] \times h[\cos^{-1}(\cos\eta_i\cos\phi+\sin\eta_i\sin\phi\sin y)]dy. \qquad (4)$$

Several numerical solutions of Eq. (4) have been determined using appropriate values of $p_i(\phi)$ and η_i. These indicate that the basic form of $f(\phi)$ may be adequately represented by considering only the most common reorientation angle, $\eta_1=90°$. In this case the solution of Eq. (4) reduces to

$$F(\phi)=\frac{K\sin\theta}{p_1(\phi)}, \qquad (5)$$

[1] Derivations of Eqs. (2)–(4) will be made available on request.

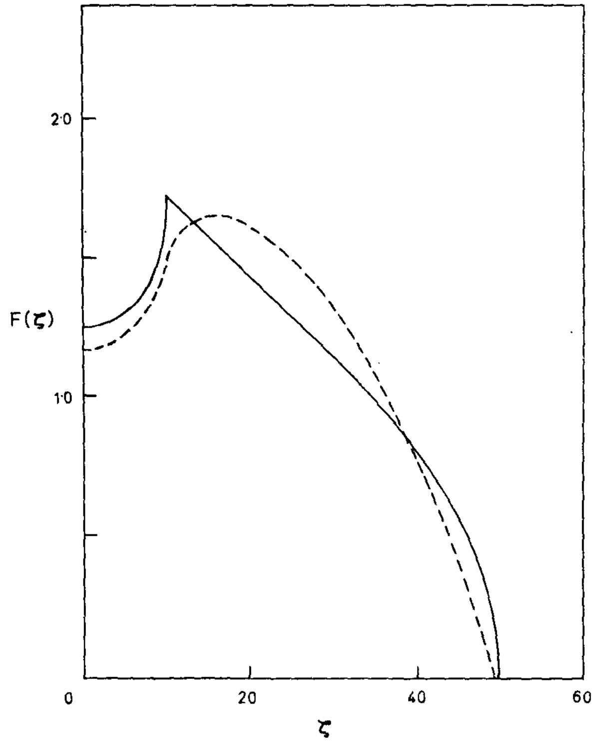

Fig. 2. The effect of using the c'-axis to determine crystallographic orientation distributions for $\psi=30°$ and $\alpha=20°$. The continuous lines are for rough cylinders and the dashed lines for rough spheres.

Unauthenticated | Downloaded 02/24/24 07:34 AM UTC

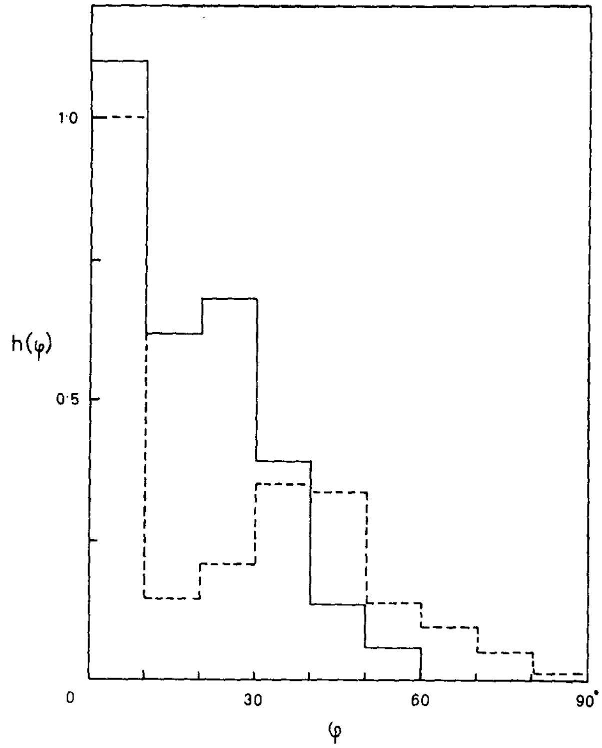

FIG. 3. Distributions $h(\phi)$ corresponding to $f(\phi)$ distributions shown in Fig. 1b (continuous line) and 1c (dashed line) of Levi and Aufdermaur (1970).

where K is a constant. An estimate of $p_1(\phi)$ can be obtained in the following way. When $\phi=0$, the value of $p_1(\phi)$ is the probability of crystallographic reorientation on the basal face of ice; $\phi=\pi/2$ corresponds to reorientation on a surface perpendicular to the basal plane, such as a prism face. Hallett (1964) has shown that, at small supercoolings, $p_1(\pi/2)$ is much less than $p_1(0)$. Under these conditions $f(\phi)$ has a sharp peak at $\phi=\pi/2$. However, crystal faces may not be well defined on the microscopically rough surface of a hailstone. This results in a reduction in the variation of $p_1(\phi)$ with ϕ. The extreme case, where $p_1(\phi)$ is constant, gives

$$f(\phi)=\sin\phi. \tag{6}$$

The implications of Eq. (6) are discussed in Section 6.

It should be noted that the calculations based on this model show that there is no physically reasonable mechanism by which crystallographic reorientation alone can give rise to the distributions observed by Levi and Aufdermaur (1970).

6. Comparison with experiment

a. c-axis distributions

The distributions shown in Figs. 1b and 1c of Levi and Aufdermaur (1970) are similar in form to the theoretical curves given in Fig. 2 except that $f(\phi)$ does not fall to zero at $\phi=0$. This effect is best shown by the function $h(\phi)$ as indicated in Fig. 3. Both distributions possess pronounced peaks at $\phi=0$ as well as at $\phi=\psi$. The peaks at $\phi=0$ are considered to be due to the warming of the water layer near the ice-water interface which occurs during the initial stage of freezing of the individual droplets. Assuming that the warming raises the temperature to about 0C, then the crystals whose c-axes are normal to the ice surface produce dendritic sheets which are parallel to the surface. Such crystals are always in a preferred orientation. The peak at $\phi=\psi$ remains because dendrites from other crystals can grow (at a constant rate) through the warmed layer (the thickness of which increases as time$^{\frac{1}{2}}$) into the unwarmed region of the droplet.

In Fig. 8 of their paper, Knight and Knight (1970) have eliminated the effect of surface roughness (pronounced lobes in this case) by measuring the angles between the c-axes and the local normal to the hailstone surface. They observe that their distribution is a close approximation to a sine curve. Eq. (6) shows that such a distribution may result from crystallographic reorientation. Statistical calculations based on Knight and Knight's data indicate that the deviations from a sinusoidal distribution are not random but are due to well-defined selection effects. Table 1 shows the probabilities that the counts in each of the 10° intervals equal or exceed the experimental values, assuming that the overall distribution is sinusoidal. The probability that a crystal has an orientation in any one interval was calculated on the basis of a binomial distribution. There are two peaks which are very unlikely to be random, namely, those occurring at $20°\leqslant\phi\leqslant 29°$ and $70°\leqslant\phi\leqslant 79°$.

Knight and Knight (private communication) have given a complete fabric diagram for the hailstone section whose c-axis distribution was measured, including the c'-axis orientations of each crystal. Fig. 4 shows the locations in the hailstone section of the crystals with c'-axes in the ranges where peaks occurred. The crystals with orientations in the 20°–30° interval occur mostly in the outer zone of the section, while those with orientations in the 70°–80° interval lie mainly inside the zone where bubble layering

TABLE 1. Probabilities for the distribution quoted by Knight and Knight (1970).

Interval (deg)	Number n of crystals	Expected value assuming sine distribution	Binomial probability that number $\geqslant n$
0–9	6	4.04	0.220
10–19	14	12.00	0.316
20–29	29	19.59	0.023
30–39	20	26.60	0.931
40–49	26	32.78	0.917
50–59	29	37.98	0.956
60–69	38	42.02	0.774
70–79	58	44.78	0.021
80–90	46	49.19	0.538

Unauthenticated | Downloaded 02/24/24 07:34 AM UTC

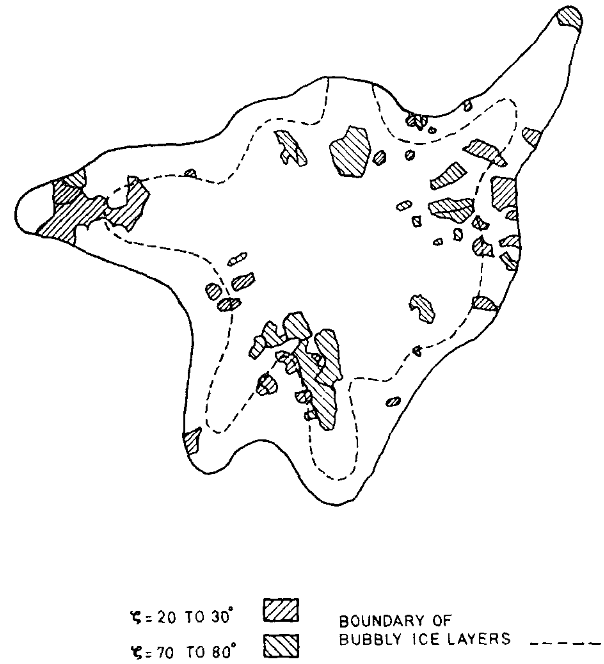

Fig. 4. Location of crystals with c'-axis orientations in the ranges 20°–30° and 70°–80° for the thin section of hailstone OR-Y12 analyzed by Knight and Knight (1970).

occurs. The hyperfine bubble structure is indicative of growth just beyond the wet growth limit where the droplets form a liquid film on the surface before freezing, so that the bubble structure is then similar to that obtained in the freezing of the bulk liquid (Carte, 1961). The clear ice zone is formed near the wet growth limit where the droplets freeze individually but sufficiently slowly for the dissolved air to escape by diffusion (Macklin, 1962; Brownscombe and Hallett, 1967). It is inferred that the peak between 20° and 29° is an enhancement due to dry growth while that between 70° and 79° is due to wet growth, as discussed by Levi and Aufdermaur (1970).

b. c'-axis distributions

Unique interpretation of the peaks in the c'-axis distributions is not possible because surface roughness markedly affects their position. However, in view of the considerable number of such distributions determined by various authors (Aufdermaur *et al.*, 1963; Knight and Knight, 1968; List *et al.*, 1970), attempts were made to convert these to equivalent c-axis distributions so that any peaks in the latter distributions could be related to the droplet temperature through the growth angle ψ. This was done by inverting the transform for $F(\zeta)$ in Eq. (2) using a series expansion. The main difficulty in interpreting the distribution $f(\phi)$ computed from $F(\zeta)$ is to decide whether any peaks obtained are now random. As shown in Section 4 there is a smoothing effect on peaks in $f(\phi)$ by the transform to $F(\zeta)$. Consequently, in the reverse process, random fluctuations in $F(\zeta)$ are accentuated.

Of the three works mentioned, that of Knight and Knight (1968) contains the most useful results, since the samples used were large (up to 281 crystals) and the counting intervals small (5°). Even so, it was necessary to smooth the data, by summing the counts in adjacent intervals, to obtain curves with sufficiently small random fluctuations. The theoretical analysis of each of the distributions shown in Figs. 2, 3b, 4 and 5 of Knight and Knight showed two significant peaks in the distribution $h(\phi)$, one at $\phi=0°$ and the other in the range $\phi=27°$ to $\phi=40°$, while Fig. 1 showed a significant peak only at $\phi=0°$. The curve for Fig. 3a was qualitatively similar to that for the hailstone whose crystallographic orientation distribution was shown in Fig. 8 of Knight and Knight (1970). As an example, Fig. 5 shows the curve $F(\zeta)$ of best fit to the distribution shown in their Fig. 2, together with the corresponding $h(\phi)$ distribution. The distributions obtained by Knight and Knight (1968) are therefore in good agreement with those of Levi and Aufdermaur (1970). The differences between the two sets of distributions are probably due to differing surface roughness.

7. Conclusions

The above studies indicate that the crystallographic orientation distributions of ice deposits formed by

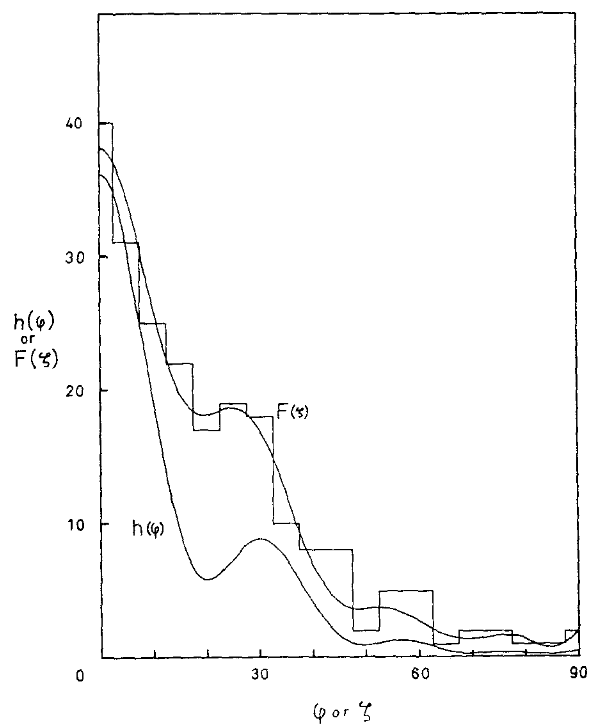

Fig. 5. Distributions $F(\zeta)$ and $h(\phi)$ calculated from the c'-axis distribution shown in Fig. 2 of Knight and Knight (1968).

Unauthenticated | Downloaded 02/24/24 07:34 AM UTC

accretion of supercooled droplets in the dry growth regime are determined primarily by a crystal selection process which is operative at all temperatures. Crystallographic reorientation is important only insofar as it supplies an initial distribution of orientations from which certain orientations are selected.

The effect of surface roughness, which develops during the accretion process, is to broaden any peaks in both the c- and c'-axis distributions. In the case of the c'-axis distributions there is also a shift of the location of the peaks to smaller angles so that they can no longer be related directly to the growth conditions. Clearly, future crystallographic orientation measurements should involve c-axes only and, if practicable, should be made relative to the local surface normal as this ensures maximum resolution of the peaks.

Distributions expressed in terms of the c-axis orientation probability per unit solid angle avoid the geometrical effect which results in the probability per unit angle being zero parallel to the growth direction. When the distributions measured by Levi and Aufdermaur (1970) and Knight and Knight (1968) are expressed in this way, two peaks are observed. One peak is located parallel to the growth direction and is probably due to droplet warming by latent heat released at the ice-water interface. The other occurs at an angle ψ, which is presumed to be the angle of growth of the dendritic sheet formed in the initial freezing of the droplets. Because of this preferred orientation it is in principle possible to infer from the crystallographic orientation distributions of some hailstone layers the ambient temperature at which they were formed. The ratio of the sizes of the peaks in the growth direction and at the angle ψ may also give empirical information on the growth conditions of the hailstone. Further investigations are required to determine the usefulness of this latter parameter.

Acknowledgments. This work was supported by the Australian Research Grants Committee. P. J. Rye is indebted to the Commonwealth Scientific and Industrial Research Organisation for the provision of a Postgraduate Studentship. We are indebted to Dr. C. A. Knight and Mrs. N. C. Knight for supplying detailed information on hailstone OR-Y12.

REFERENCES

Aufdermaur, A. N., R. List, W. C. Mayes and M. R. de Quervain, 1963: Kristallachsenlagen in Hagelkörnen. *Z. Angew. Math. Phys.*, **14**, 574–589.

Brownscombe, J. L., and J. Hallett, 1967: Experimental and field studies of precipitation particles formed by the freezing of supercooled water. *Quart. J. Roy. Meteor. Soc.*, **93**, 455–473.

Carte, A. E., 1961: Air bubbles in ice. *Proc. Phys. Soc.*, **77**, 757–769.

Hallett, J., 1964: Experimental studies of the crystallization of supercooled water. *J. Atmos. Sci.*, **21**, 671–682.

Higuchi, K., and T. Yosida, 1966: Crystallographic orientation of frozen droplets on ice surfaces. *Proc. Intern. Conf. Low Temperature Science*, Sapporo, Japan, 79–93.

Knight, C. A., 1971: Breeding of crystal nuclei by classical nucleation: Theory and some observations and experiments. *J. Cryst. Growth*, **11**, 201–217.

——, and N. C. Knight, 1968: Spongy hailstone growth criteria I. Orientation fabrics. *J. Atmos. Sci.*, **25**, 445–452.

—— and ——, 1970: Lobe structures of hailstones. *J. Atmos. Sci.*, **27**, 667–671.

Levi, L., and A. N. Aufdermaur, 1970: Crystallographic orientation and crystal size in cylindrical accretions of ice. *J. Atmos. Sci.*, **27**, 443–452.

List, R., J. G. Cantin and M. G. Ferland, 1970: Structural properties of two hailstone samples. *J. Atmos. Sci.*, **27**, 1080–1090.

Macklin, W. C., 1962: The density and structure of ice formed by accretion. *Quart. J. Roy. Meteor. Soc.*, **88**, 30–50.

——, and B. F. Ryan, 1965: The structure of ice grown in bulk supercooled water. *J. Atmos. Sci.*, **22**, 452–459.

——, and G. S. Payne, 1967: A theoretical study of the ice accretion process. *Quart. J. Roy. Meteor. Soc.*, **93**, 195–213.

Pruppacher, H. R., 1967: Growth modes of ice crystals in supercooled water and aqueous solutions. *J. Glaciology*, **6**, 651–662.

Unauthenticated | Downloaded 02/24/24 07:34 AM UTC