Examination of Ice-Crystals by X-Rays

By

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Abstract

The orientation of ice-crystals obtained under various conditions was examined by means of X-rays, and it was found that all directions parallel to the basal plane of the hexagonal crystal of ice are equally suited for its growth.

For the determination of the orientation of the crystallographic axes of ice, the optical method, in which a polarized light is used, is only effective for the direction of the principal axis of the hexagonal ice-crystal, and has no bearing on the other crystallographic directions. With X-rays, however, not only is this defect of the optical method remedied, but some-times even the arrangement of the microcrystals oriented in some regular manner can be determined. Thus the writers have examined roentgenographically some specimens of ice obtained under various conditions. The method and the results obtained are described below.

In order to take a Laue-photograph with ice, we must keep the specimen frozen throughout the whole exposure of the photographic plate. For this purpose a simple apparatus as shown in Fig. 1 was employed. In this figure C is a copper rod, and the specimen to be examined is inserted in a small vertical hole A in the upper part of the rod C. The small horizontal hole B passing through the upper part of the rod C is a slit for the X-rays. The X-rays starting from the molybdenum target of a Coolidge tube illuminate the specimen of ice in the hole A, after passing through the slits S and B; and the pattern of the diffracted X-rays is impressed on the photographic plate P standing perpendicularly to the incident X-ray beam. The specimen of ice is kept frozen, in winter, by dipping the lower part of the copper rod C in a suitable freezing mixture.

The orientation of the crystallographic axes of the ice-crystal was determined by treating the Laue-spots, taken with the specimen to be tested, with the globe and the spherical scale first devised by one[1] of the writers.

First the writers examined the orientation of the crystallographic axes of ice, which had been frozen on the surface of calm water by being exposed to the cold air during a fine night in winter. The Laue-photograph taken with a piece of such ice consists usually of one set of Laue-spots belonging to a single crystal of ice, as seen in Fig. 1, Plate I. This fact indicates that the ice obtained under such circumstances is composed of single crystals of considerable size. The orientation of the crystallographic axes of seven different pieces of ice, obtained on different days in different localities, is shown in Fig. 2 in a stereographic projection. In this figure the direction of the normal to the flat surface of ice, that is the direction of the surface normal of water is represented by a dot in reference to the directions of the crystallographic axes 0001, 10$\bar{1}$0 and 11$\bar{2}$0 of the hexagonal crystal of ice. As is seen in this figure, all the dots are crowded near the direction of the principal axis 0001. This fact shows that under the circumstances mentioned above, the water freezes in such a manner, that the basal plane of the hexagonal crystal of ice

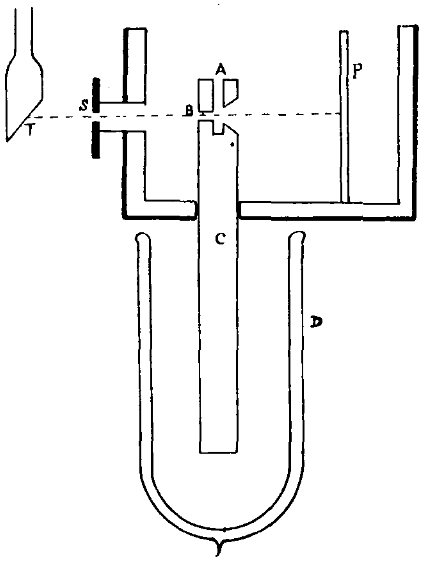

Fig. 1

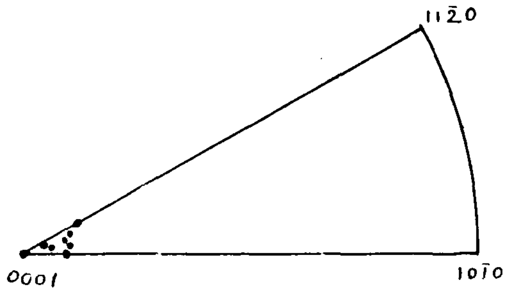

Fig. 2

1 U. Yoshida; Japanese J. Phys., **4**, 133 (1927)

produced is nearly parallel to the surface of the calm water.

If we consider that the ice on the surface of such calm water grows horizontally, from a certain nucleus, along the surface of the water, the horizontal orientation of the basal plane of the hexagonal crystal of ice seems to be caused by the fact that the growth of the ice crystal is easier in the directions parallel to the basal plane of the hexagonal ice crystal.

The same phenomenon was also observed in ice columns in the ground. Usually an ice column in the ground takes a long prismatic form, each prism consisting of several smaller prisms of single crystals of ice of about 0.5 mm. in diameter. The orientation of the crystallographic axes of these single crystals of ice were determined by taking their Laue-photographs, and the direction of the axis of the prisms of such single crystals is represented, in a stereographic projection, by the position of the small circles in Fig. 3, refering to the direction of the crystallographic axes. Here again we can see that the direction of the growth of the ice crystal is perpendicular to that of the principal axis, namely parallel to some direction in the basal plane of the hexagonal crystal.

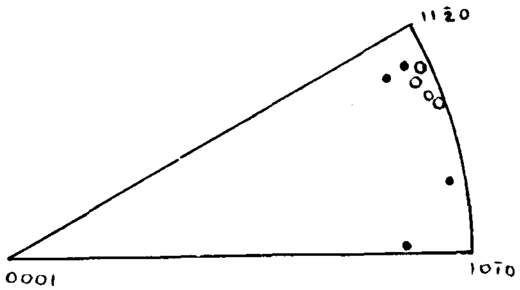

Fig. 3

Sometimes it was very difficult to split a prism of an ice column in the ground into component prisms of single crystals. In such a case a Laue-photograph was taken with such a prism as a whole. The resulting Laue-spots, consisting of two or more sets of the Laue- spots belonging to two or more single crystals of ice in different orientations of their crystallographic axes, were classified into two or more sets by trial with the spherical scale and the globe previously described; and the orientations of the crystallographic axes of the separate crystals corresponding to the separate sets of the Laue-spots were determined at the same time. Fig. 2 in Plate I is shown as an example of this. It was found, in the manner stated above, that the Laue-spots in this figure were due to two single crystals of different orientations. The result of classification of these Laue-spots into two sets due to two separate single crystals is shown in Fig. 4, where all the small circles are due to a single crystal and all the dots are due to another single crystal of different orientation. The big dot in that figure indicates the position of the impression of the direct X-ray beam, and the arrow represents the direction parallel to that of the axis of the prism of the ice column in the ground.

The growth of ice is also slow in the course of the development of an icicle. In the portion near its upper root it is formed of many small crystals of ice, and at its lower end it consists usually of a rather slender single crystal of ice. The orientation of the crystallographic axes of the ice crystal was determined in four such slender single crystals forming the lower ends of four different icicles. The dot in Fig. 3 represents the vertical direction, that is the direction of growth of the ice crystal in this case, in reference to the crystallographic axes. The direction of growth of the ice crystal is also seen to be nearly parallel to some direction in the basal plane of the hexagonal crystal, as was seen in the case of the ice on the calm water surface and in that of the ice column in the ground. The distribution of dots and small circles in Figs. 2 and 3 shows us that all directions parallel to the basal plane of the hexagonal crystal of ice are equally suited for the growth of ice. The Laue-photograph taken with a piece of ice at the lower slender end of an icicle is shown in Fig. 3, Plate I.

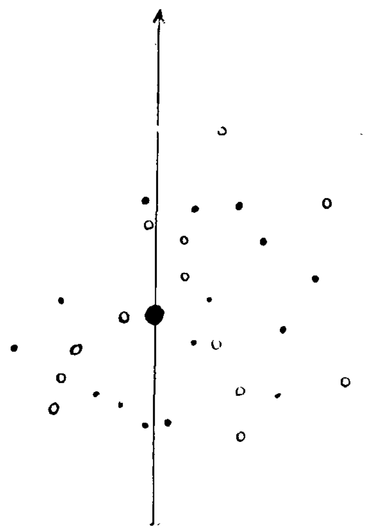

Fig. 4

When ice is formed by rapid cooling, for example as in the case of artificial ice, no such regular orientation of the ice crystal as that stated before could be detected. Artificial ice is composed of many long cylindrical ice-crystals; and the direction of the axes of such cylinders may be looked for as the direction of growth of the ice crystal. The results of experiment on several such cylindrical pieces of ice show that there is no regularity in the orientation of the crystallographic axes, as is seen in Fig. 5. In this figure the dot represents the direction of the axis of the cylinder of ice in reference to the crystallographic axes. The ice formed by the condensation of water vapour in the air on the surface of a super-cooled metal consists of an assemblage of small ice crystals. The X-ray examination shows that the orientation of such small crystals is entirely at random.

When a drop of water is brought into contact with a piece of a single crystal of ice, whose temperature is a little lower than 0°C, then the drop of water solidifies slowly. The orientation of the crystallographic axes of ice thus newly formed was confirmed, by the X-rays, to be entirely the same as that of the mother crystal of ice. However, when the temperature of the mother crystal of ice, on which the drop of water added solidifies, is much lower than 0°C, then the drop of water freezes rapidly. The ice thus formed suddenly from the drop of water was ascertained, by means of X-rays, to be composed of many small crystals in a random orientation. Fig. 4 in Plate I is a long slit Laue-photograph[1] taken with a thin and long specimen of ice, which is composed mostly of a single crystal of ice and of a small piece obtained by sudden cooling from a drop of water added to the upper end of the single ice-crystal. The presence of many spots distributed rather irregularly in the upper part of the figure indicates that the ice thus formed by sudden cooling from a drop of water is an aggregate of many small crystals in an irregular orientation.

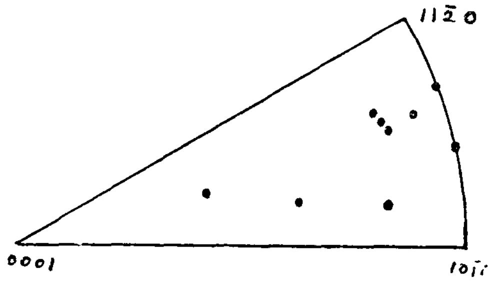

Fig. 5

1 U. Yoshida and K. Tanaka: Nature, **118**, 912 (1926)

Plate I

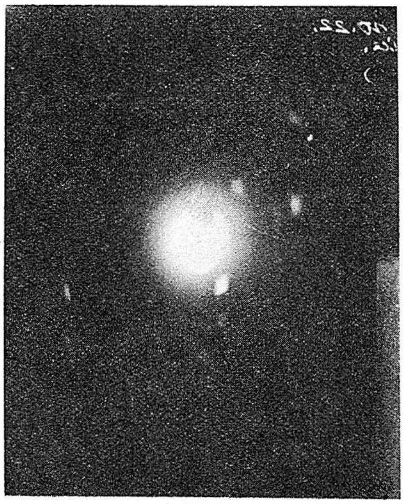

Fig. 1

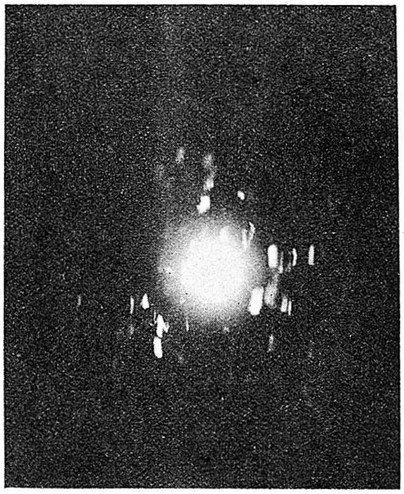

Fig. 2

Fig. 3

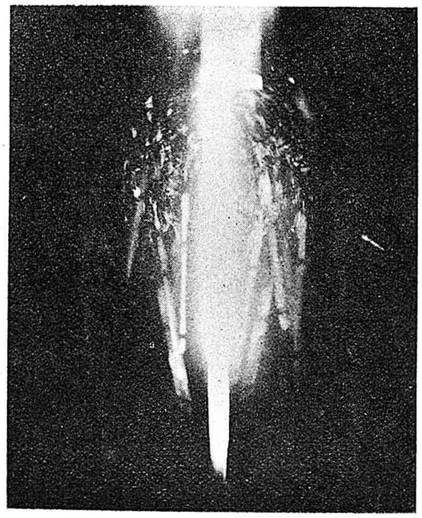

Fig. 4