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Experimental Studies on Snow Crystals of Plane Type with Spatial Branches

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Abstract

Riming experiments of supercooled droplets on the surface of an ice plate of single crystal were made at various temperatures below freezing, and their crystal orientations were observed under a polarization microscope. It was found that the droplets were frozen into single crystal with c-axis parallel to that of the ice plate at temperature regions warmer than -15°C, while the droplets were frozen into poly crystal or single crystal with the c-axis different from that of the ice plate at temperature regions colder than -15°C.

Further, artificial snow-making experiments for snow crystals with spatial dendritic branches were made, utilizing such droplets rimed on the ice plate, and it was confirmed that spatial branches developed from the droplets with c-axes different from that of the ice plate. It seemed that larger ones among the rimed droplets acted as nuclei for spatial branches.

As a result of the experiments, it was concluded that some of rimed droplets which were frozen to ice surface at temperature regions colder than -15 or -20°C, became nuclei for the spatial branches. This result agrees well with the meteorological condition of natural snow crystals of plane type with spatial dendritic branches.

1. Introduction

The snow crystal of plane type with spatial branches is noted for its characteristic shape, as illustrated in Photo. 1, Plate I. Hereinafter, this snow crystal will be referred to simply as 'Spatial snow crystal' in this paper. Nakaya1) proposed that the formation mechanism of the snow crystal of this type was as follows. During the fall of a snow crystal of hexagonal type, many nuclei become attached to the lower surface of the hexagonal plane, when it falls keeping its plane horizontally, and moreover the supply of water vapor is more abundant on that surface.

Magono and his colleagues2), and later Lee and Magono3) made observations on natural snow crystals in Hokkaido, and it was confirmed that Nakaya's Ta-s diagram4) (air temperature *vs* supersaturation grade diagram) was applicable to the growth of natural snow crystals. During these observations, it was noted that spatial snow crystals were invariably observed when a temperature inversion layer around -20°C was included in a cloud layer. As a result of analysis of temperature condition of the cloud layer through which the spatial snow crystals passed, it was confirmed that the snow crystals were formed in the following manner. At first a snow crystal of plane type is formed in an upper layer of a cloud with a temperature around -17°C, then many nuclei become attached to the lower surface of the snow crystal when it passes through a middle layer of a colder temperature, for example -20°C, and then spatial branches develop from the nuclei when the snow crystal falls through a lower and warmer layer of the cloud with a temperature of about -15°C again. Thus the spatial snow crystal is recognized as an indicator of the existence of a temperature inversion layer around -20°C in a cloud.

As described above, the temperature conditions for the formation of spatial snow crystals was fairly well clarified, however the following questions arised concerning the basic mechanism of development of spatial branches.

1. What are the nuclei which act as the origin of the spatial branches?
2. Why do the nuclei attached within a limit of approximately -20°C act as the origin ?
3. It has been noted that the direction of spatial branches are not random but frequently have some particular angles with the basal plane of the snow crystal. Why ?

Concerning the nature of the nuclei, explanations were not given by Nakaya. However, it was believed that the nuclei may well be cloud droplets which rimed on the lower surface of a snow crystal of plane type. This may be easily understood by observing Photo. 2, Plate I which shows an early stage of spatial branches which seems to have originated from cloud droplets rimed on the basal plane of a snow crystal.

Hallet[5] made a laboratory experiment for the study of crystalline of supercooled water drops which were frozen on an ice plate of single crystal, and he showed that the water drops were frozen to ice with different crystal orientation from that of the basal ice place in a temperature region colder than -5°C, and demonstrated that branches originated from the frozen water drops and developed spatially. However, the size of the water drop used by him was too large compared with the size of natural cloud droplets.

Higuchi and Yosida[6] also carried out a similar laboratory experiment in which columnar branches developed spatially from supercooled water droplets rimed on a frost surface at -12°C, and they confirmed that the spatial columnar branches took some particular angles with the surface of the frost, including right angles. They explained these particular angles to be the results of misfitting phenomena in the lattice constants of the rimed droplets and the frost.

As a result of the studies described above, the formation mechanism of spatial snow crystals was fairly well clarified, however the second question concerning the temperature dependency of the mechanism still remains unanswered. Therefore a series of experiments were made in order to study the effect of temperature on the formation of the spatial branches.

At first, a riming experiment of supercooled water droplets on an ice plate was made at various temperatures, then the crystal axes of the rimed droplets were observed. As a second step of the experiment, spatial branches of dendritic type were reproduced artificially on an ice plate, utilizing the rimed droplets as nuclei. This paper will describe the results of the experiments.

2. Experimental method

In order to simulate the plane of a snow crystal, an ice plate of single crystal was used. The crystal axis of the ice plate was perpendicular to the plane of the plate. The artificial riming of supercooled water droplets on the ice plate was made in such a way as shown schematically in Fig. 1. The ice plate was cemented on a slide glass. Water droplets were sprayed from an atomizer in a cold chamber at various temperatures, namely −5, −10,.... and −45°C. The size of the droplets ranged from 10 to 100 μ in diameter. The distance between the ice plate and the nozzle of the atomizer was about 3 meters which was sufficiently large to cool down the droplets to below freezing temperature.

The crystal axis of droplets rimed on the ice plate was observed under a polarization microscope, utilizing the difference in color due to the difference in the direction of the crystal axis, as stated in the previous paper by Magono and Suzuki(7).

3. Results

Color photographs of rimed droplets under the polarization microscope are shown in photographs in Plate II. The color of the background, namely the color for the ice plate was colored red or violet, owing to the effect of a color sensitizing plate which was used in order to increase the change in

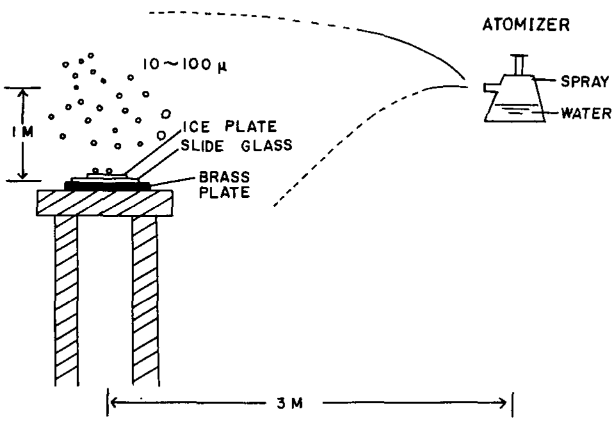

Fig. 1 Method for artificial riming of supercooled droplets on an ice plate of single crystal.

color due to the change in the crystal axis.

Photo. 3 shows an example of a microscopic photograph of droplets which were rimed on an ice plate at –15°C. It may be seen that all droplets were commonly colored red reflecting the color of the background. This means that all droplets were frozen into ice particles of single crystal whose axes were the same as that of the basal ice plate, namely their axes were all vertical to the plane of the paper.

Photo. 4 shows an example of droplets which were rimed at –10°C. It may be seen that the color of the droplets were roughly the same as that of the ice plate, but the center part of larger droplets had several colors which were different from that of the background. This means that the larger droplets were frozen partially into poly crystals whose axes were different from that of the basal ice plate. This tendency agrees with that of Hallet's result.

Photo. 5 shows a result in experiments at –15°C. It may be seen that the crystal axes of larger droplets were different from that of the basal ice plate. It may also be noted that several small ice crystals of hexagonal plate type were observed in the photograph. Presumably the ice crystals were formed already in air at –15°C. In other words they were not rimed on the ice plate, but only fell on it, as ice crystals. It is well known that ice crystals develop to the hexagonal plate type at air temperature around –15°C.

In the case of Photo. 6 which shows a result of experiments at –20°C, it may be clearly seen that almost all droplets were frozen into poly crystal.

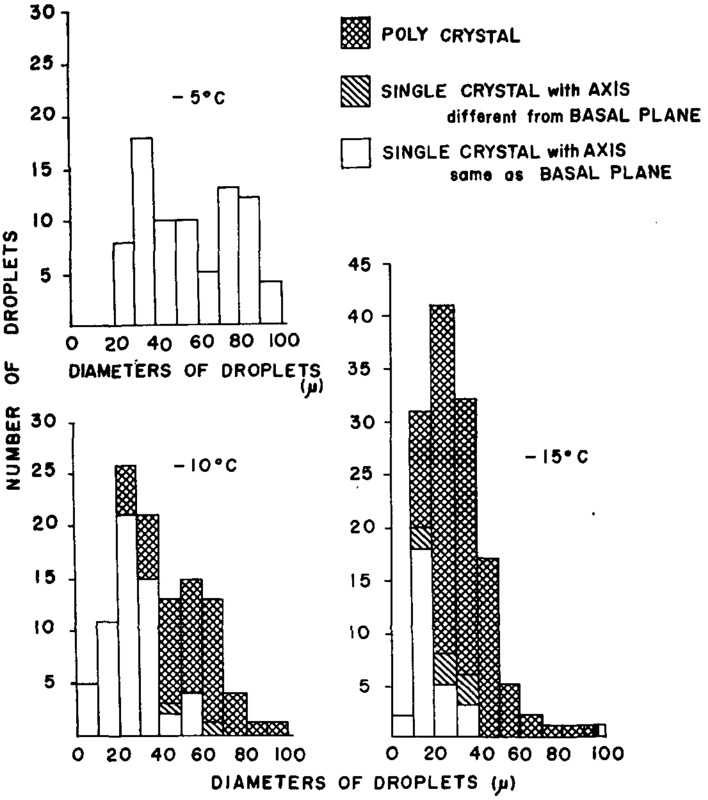

Fig. 2 Size distribution of droplets frozen on an ice plate at temperatures of −5, −10 and −15°C.

Photos. 7 and 8 show the result of experiments at −25 and −30°C respectively. They also show that larger droplets were frozen into poly crystal. The results of experiments at −35, −40 and −45°C were the same as those at warmer temperature regions between −20 and −30°C as described above, although the respective photographs are not given here.

The results of experiments in all temperature regions are shown in the form of a histogram in Figs. 2, 3 and 4. In the figures, white areas of the column mean that all rimed droplets were of single crystal and their crystal axes were common to that of the basal ice plate, and the dashed areas show that rimed droplets of single crystal but their axes were different from that of the ice plate, and cross dashed areas indicate that rimed droplets were

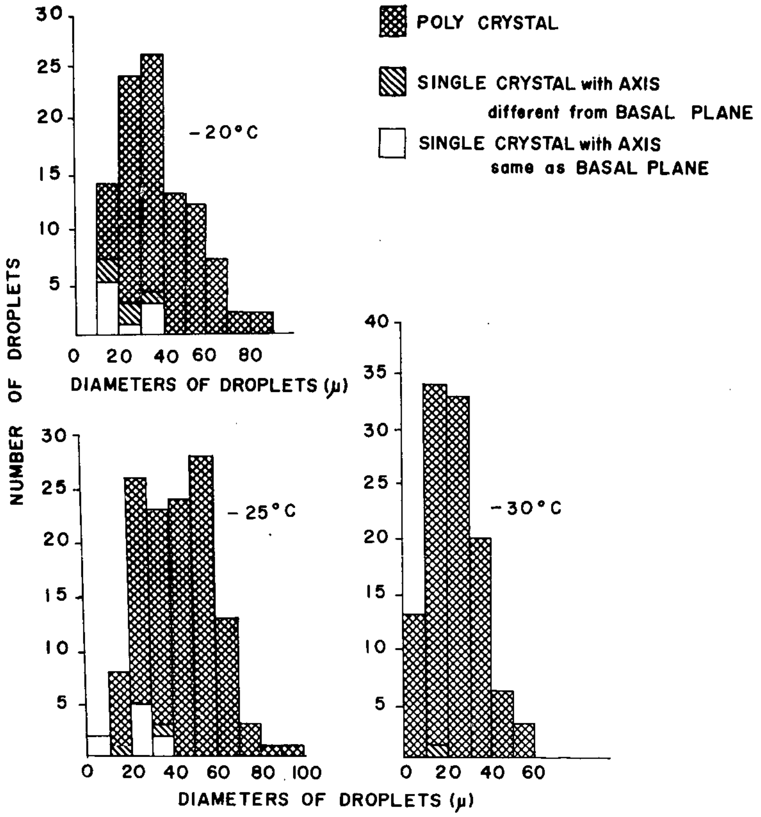

Fig. 3 Size distribution of droplets frozen on an ice plate at temperatures of −20, −25 and −30°C.

frozen into poly crystals whose axes were of course different from that of the ice plate. In the histogram, horizontal axis shows a size distribution with a width of 10 μ, and vertical axis shows the number of rimed droplets whose axes were observed.

As seen in the left upper histogram in Fig. 2, at -5°C all droplets were frozen into single crystal with the same axis as that of the basal ice plate. However as seen in the left lower histogram, at -10°C about half of the droplets were frozen into poly crystals. In the case of -15°C, the majority of rimed droplets were poly crystal, as seen in the right hand histogram in Fig. 2. In the temperature regions of -20 and -25°C, almost all droplets were poly crystal, as seen in the left hand histogram in Fig. 3. The fraction of droplets of a

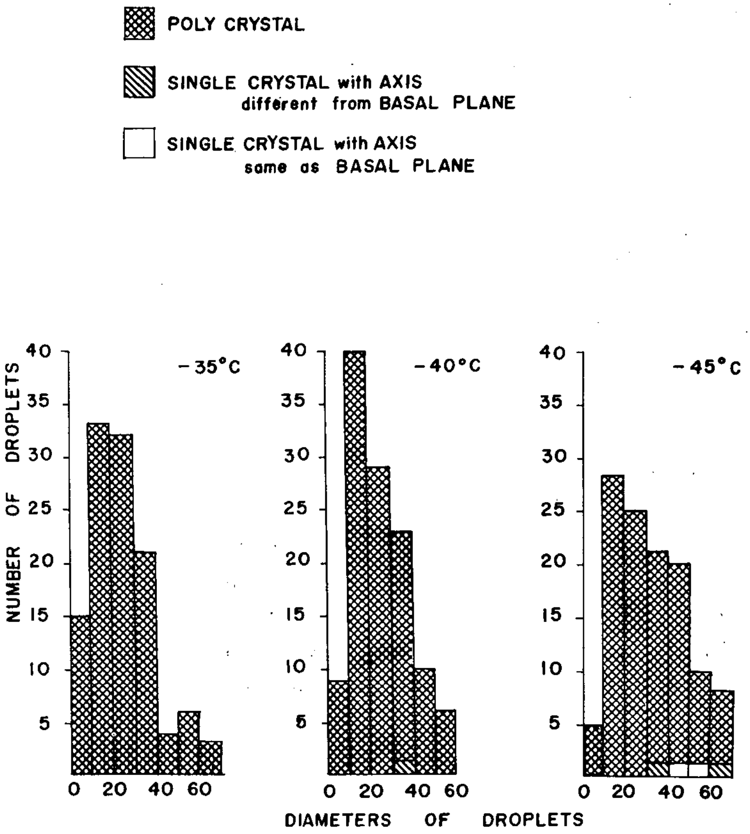

Fig. 4 Size distribution of droplets frozen on an ice plate at temperatures of −35, −40 and −45°C.

single crystal with axes different from that of the ice plate was very small in a temperature region colder than -10°C. In the coldest temperature regions lower than -30°C, all droplets were frozen into poly crystal as seen in Fig. 4.

4. Considerations

The results described above may be summarized as follows.

1. In a temperature region warmer than -5°C, all droplets were frozen to single crystal with the same axes as that of the basal ice plate, as generally expected from the epitaxy theory.
2. In temperature regions colder than -10°C, as the temperature decreased,

the fraction of droplets with axes different from the basal plate increased rapidly.

3. The threshold temperature for freezing of droplets to different crystal axes was between –5 and –10°C.

The former two results agree with that obtained by Hallet⁵⁾, and the results may be qualitatively understood if we consider the epitaxy theory. Namely when the grade of supercooling is small, droplets have sufficient time to freeze to ice with the same crystal orientation as that of the ice plate under the effect of crystal habit of the surface of the ice plate, while when the grade of supercooling is great, droplets are frozen to ice with random crystal orientations, having no time to receive the effect of the surface of the ice plate. However it seems that the crystal orientation of the rimed droplets is not perfectly random, but has some particular angles with each other. The some particular angles may be related to the misfitting phenomenon, as pointed out by Higuchi and Yosida⁶⁾.

It is considered that branches develop spatially from rimed droplets with crystal orientation different from the basal ice plate, being parallel to their a-axes. However the value of the threshold temperature for the different crystal orientation of rimed droplets (–5 to –10°C) was much warmer than –20°C which would be expected from the natural case. This difference seems to originate from the size of the droplets.

The diameter of natural cloud droplets rimed on snow crystals are usually smaller than 40μ with an average value of 10∼15μ (Nakaya⁸⁾), (Kikuchi and Magono⁹⁾). Therefore water droplets smaller than 20μ should be considered in the application of the results of experiments to natural riming phenomena in clouds. Therefore if we only consider droplets smaller than 20μ, it may be seen in Fig. 2 that the threshold temperature for different crystal orientation was about –15°C, however a value difference of 5°C still remains between the experiment and the natural spatial snow crystals. Concerning this disagreement, a possible explanation may be as follows. Rimed droplets with different crystal orientation do not always act as nuclei for spatial branches, but only certain droplets may be effective as the nuclei. This explanation is only an idea, therefore it requires some experimental proof.

On the other hand, the simulation in the experiment described above was considered to be imperfect in the following two points. One is that the droplets in the experiment fell from above and were rimed on the upper surface of the ice plate, but in a natural case cloud droplets are rimed on the lower surface of falling snow crystals. This means that droplets on the surface of the ice plate were not always rimed droplets but a portion of them might be ice particles which were already frozen in the air before colliding with the ice plate. The portion might be particularly great in the coldest temperature region lower than –30°C. The other point is that considerably large droplets as compared with natural cloud droplets were included in the experiment.

5. Improved experiment

Because the experiment was not perfect for the simulation of the growth of spatial snow crystals, three improvements were made as follows.

When artificial riming of supercooled droplets was made on an ice plate the plate was inclined as shown schematically in the right picture of Fig. 5 where supercooled droplets were attached to the lower surface of the ice plate, in order to ensure the collection of rimed droplets only. If supercooled droplets were frozen in the air prior to falling on the ice plate, they would not be attached to the lower surface of the ice plate.

The atomizer was improved to produce droplets with diameters smaller than 30 μ.

In order to ensure that spatial branches were surely developed from rimed droplets with different crystal orientations from the ice plate, the plate was suspended in an artifiical snow-making apparatus of the Nakaya type[10]), as shown in the left picture of Fig. 5. The air temperature of the space was kept at about –15°C and moist air was supplied from the bottom where a water reservoir was set. It was considered that when branches originated from suitable nuclei, they would develop rapidly to dendritic form.

The practical treatment in the experiment was as follows.

1. A cross mark × was curved on the surface of an ice plate of single crystal with its axis vertical to the surface, in order to record the exact site on the surface.
2. Supercooled water droplets with sizes smaller than about 30 μ were made to collide with the lower surface of the ice plate, as shown in Fig. 5 at air temperatures of –10 or –25°C.
3. The site near the cross mark was photographed with a polarization microscope.
4. The ice plate was suspended in the space of –15°C in the snow-making apparatus and was exposed to moist air, as shown in Fig. 5.

5. After about 10 minutes, the site previously photographed was photographed again, focussing both at the surface of the ice plate and at the level of spatial branches.

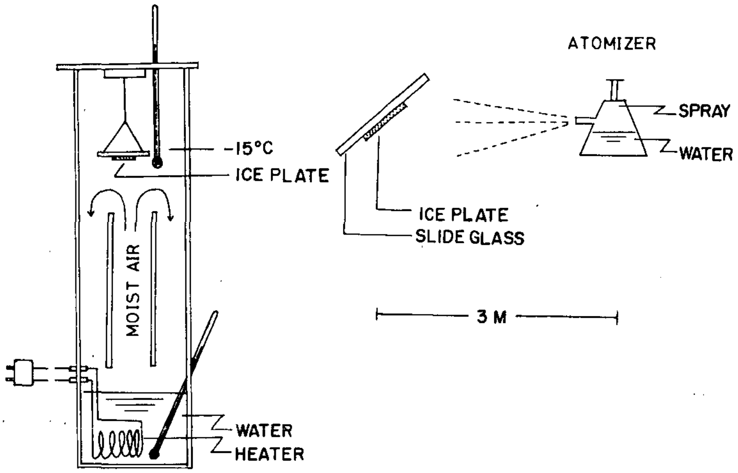

Fig. 5 Method for making spatial dendritic branches on an ice plate.

6. Results

In the improved experiment, it was noted that when supercooled droplets were rimed on an ice plate, they did not become poly crystals but became single crystals even when their crystal axes were different from that of the basal ice plate. This may be because of their small size.

The process of the growth for spatial branches from rimed droplets are illustrated in Photos. 9, 10 and 11 on the left side of Plate III. Photo. 9 shows six droplets rimed on an ice plate at -25°C. It may be seen that their colors are different from that of the background, namely their crystal orientations were different from that of the background. The cross mark to indicate the site is partially seen at the left end of the photograph.

Photo. 10 shows the spatial branches which developed from the respective rimed droplets after the ice plate was suspended in moist air of -15°C. It may be seen in the photograph that each droplet was deformed, to some extent, due to exposure to the moist air, however spatial branches developed from four droplets, namely the other two droplets were not active as nuclei. Photo. 11 shows the same photograph as Photo. 10 but with a little higher focussing level. It should be noted that remarkable spatial dendritic branches developed from only one nucleus at the center of the field of view. It is obvious that the dendritic branches developed in the direction of the a-axis. As series of these photographs indicate that rimed droplets with different crystal orientation do not always act as nuclei for spatial branches, but particular droplets, perhaps the larger droplets among them are effective as active nuclei.

Photo. 12 shows that droplets rimed at temperatures as warm as -10°C. The droplets had the same crystal orientation as that of the ice plate, as expected. After the ice plate was exposed to moist air of -15°C temperature, each droplet was changed to minute hexagonal columns or plates with axes common to the basal ice plate whose c-axis was vertical to the plate, as shown in Photo. 13. Several ice particles with different colors are seen in the photograph, however they were not droplets previously rimed, but some ice particles which were attached during the exposure to the moist air of -15°C.

As well known, dendritic branches develop in the basal plane along the a-axis of an ice crystal in mosit air at -15°C, however their growth rate in parallel with c-axis is negligible, if the vapor supply is scanty.

In the case of Photos. 9, 10 and 11, their a-axes were inclined to the basal ice plane, therefore dendritic branches developed spatially along the a-axes. In the case of Photos. 12 and 13, their a-axes coincided with the ice surface of the basal plate, therefore no branches could develop spatially. The rimed droplets were deformed only to minute hexagonal column or plate.

Photo. 14 shows a typical example of artificial spatial dendritic branches developed from droplets which were rimed on an ice plate at -25°C. It is also seen in the photograph that spatial branches developed from only particular droplets. The other nonfocussed spots show nonactive droplets on the ice plate.

7. Concluding remarks

As a result of the experiments, it was confirmed that in temperature regions warmer than -15°C, supercooled droplets with cloud droplet size were frozen into ice of single crystal with a crystal orientation common to that of the basal ice plate and none of them acted as nuclei for spatial branches even when they were exposed to moist air of -15°C. In contrast to this, in temperature regions colder than -15°C, supercooled droplets were frozen to ice with a crystal orientation different from that of the basal ice plate, and dendritic spatial branches developed from some of these frozen droplets on the plate when they were exposed to moist air of -15°C.

According to the results of the present experiments, it was concluded that the threshold temperature for rimed droplets with crystal orientation different from that of basal ice plate was about -15°C, however it was not always true that these droplets act as nuclei for spatial branches, but some portion of them, perhaps larger droplets only acted as the nuclei. Considering this fact, it may be said that the threshold temperature of rimed droplets for the growth of spatial branches is slightly colder than -15°C, namely it is about -20°C which agrees with that for natural spatial snow crystals.

Thus the reason why snow crystals of spatial branch type are observed in a temperature inversion around -20°C was explained qualitatively. However there remains still another problem. Why are snow crystals of the spatial branch type not observed in temperature regions colder than -30°C? The authors think that this problem is closely related to the question why riming phenomena on snow crystals do not occur in such a cold temperature region. Although we have no particular experimental evidence to answer this, the following explanation may be possible. Because the concentration of freezing nuclei in the atmosphere is very high in such cold temperatures, the chance of the existence of a supercooled cloud is very rare. Accordingly the riming phenomena would have difficulty to occur in temperatures lower than -30°C.

From the point of view of basic ice physics, one more question arises. Why is the threshold temperatute for different crystal orientation about -15°C in freezing of supercooled droplets ? It is considered that some knowledge from the theory of property of matter will be required to answer this question.

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Plate I Natural snow crystals with spatial dendritic branches.

Photo. 1 Spatial dendritic branches grown from a basal stellar crystal. ×32

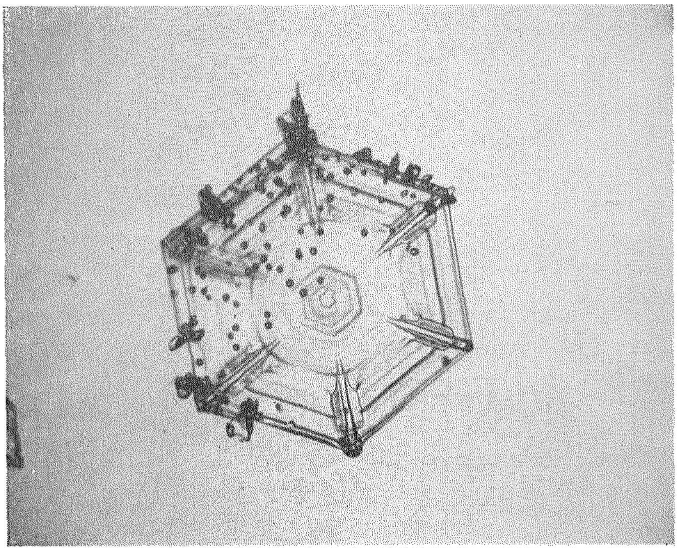

Photo. 2 Early stage of spatial dendritic branches from a hexagonal plate crystal. ×42

Plate II Crystal orientation of droplets frozen on an ice plate of single crystal.

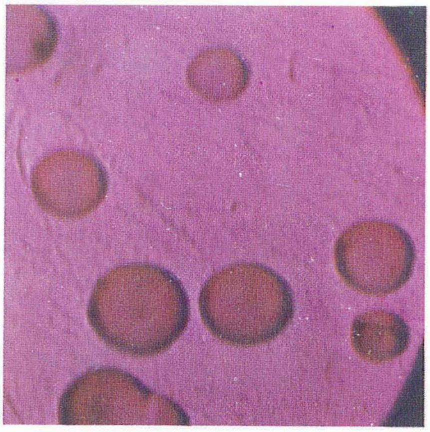

Photo. 3 At −5°C, all droplets had axes the same as that of basal ice plate. ×237

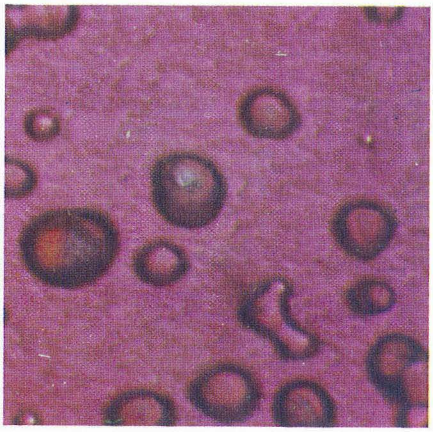

Photo. 4 At −10°C, axes of larger droplets were frozen to poly crystal. ×237

Photo. 5 At −15°C, hexagonal ice crystals were seen besides droplets. ×237

Photo. 6 At −20°C, almost all droplets were frozen to poly crystal. ×237

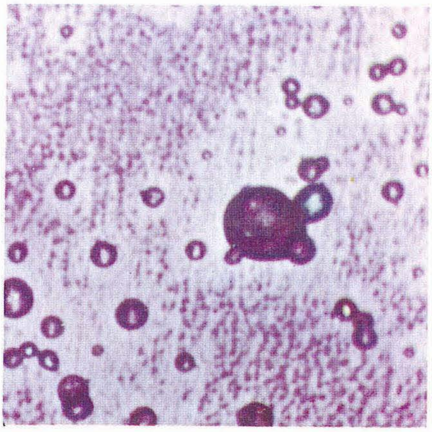

Photo. 7 At −25°C, about a half of droplets were frozen to ice with axes different from basal plate. ×237

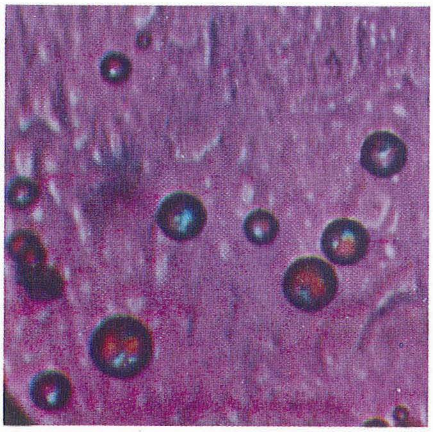

Photo. 8 At −30°C, all droplets were frozen to poly crystal. ×237

Plate III Artificial spatial dendrites grown from an ice plate of single crystal.

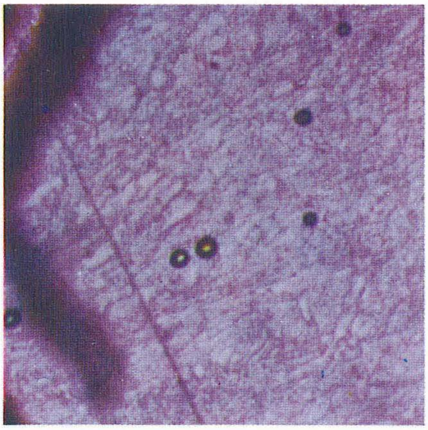

Photo. 9 Rimed droplets on an ice surface at −25°C. ×104

Photo. 12 Rimed droplets on an ice surface at −10°C. ×104

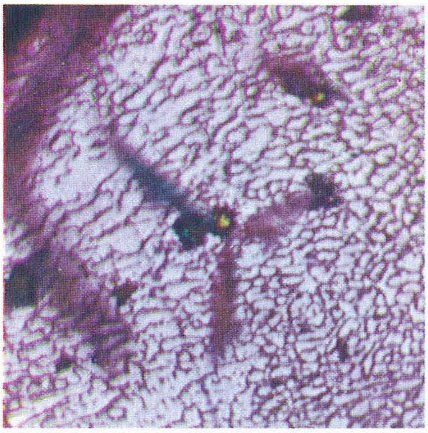

Photo. 10 Spatial branches grown at −15°C from droplets the same as above. Focussing on bottom. ×104

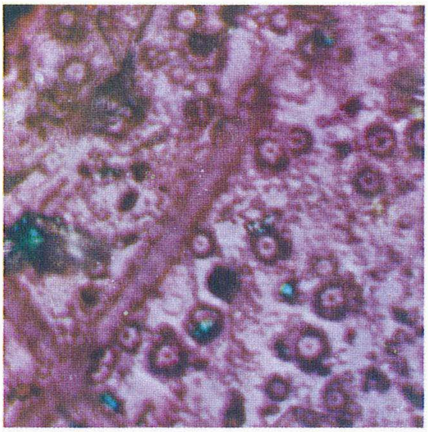

Photo. 13 Almost all droplets grew to hexagonal plates at −15°C. ×104

Photo. 11 The same as Photo. 10. Focussing on branch's level. ×104

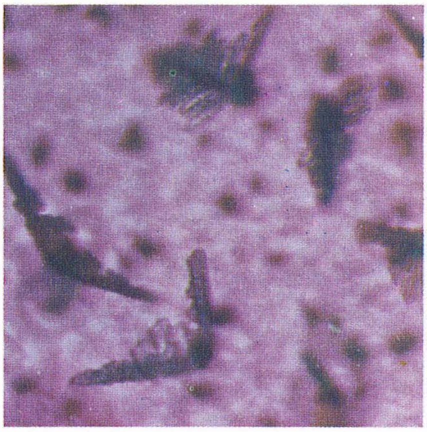

Photo. 14 Typical spatial dendrites originated from rimed droplets. ×104