Studies on Small Ice Crystals. I.
The Ice Crystals of Rectangular Plane Form.*

by

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Abstract

The author observed some ice crystals of rectangular plane form apparently belonging to the cubic system during the experiments of artificial precipitation of snow carried out at Asahigawa last winter. We were also able to verify the existence of such crystal form in our low temperature laboratory. The conditions under which they were formed will be described with some photomicrographic illustrations.

I. Introduction

As to the small ice crystals in the atmosphere which were observed on clear and very cold days in Manchuria, Sakhalin and other areas, a number of observational investigations have been reported. K. Ito[1] observed the small ice crystals everywhere in Manchuria and made full investigations on the form, size and number in relation to the atmospheric conditions. The forms of S. I. C. (small ice crystals) observed by him were mainly hexagonal twin prism, hexagonal prism, hexagonal plate and pyramid.

In laboratory experiments, artificial formation of S. I. C. can easily be brought about by seeding supersaturated water vapour with silver iodide or the like in a cold chamber. The influence of air temperature on the crystal form was critically examined by H. J. aufm Kampe, H. K. Weickmann and J. J. Kelly[2], and it was concluded that the crystal forms thus obtained were in accordance with the well-known Nakaya's diagram[3].

As far as we are aware, however, there have been made no thorough investigations on the form of ice crystals which are produced at low temperatures below ca. −30°C and with a scanty supply of water vapour.

In his work "Snow Crystals", U. Nakaya[4] mentions with reference to this problems as follows: "If we make a crystal by a very slow process at a low temperature and with a scanty supply of water vapour, the crystal ought to become a single crystal of nearly solid structure. X-ray investigations of ice crystals have been made by many physicists, but a definite conclusion is yet to be obtained.

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Barnes[5] caried out a very elaborate investigation by taking Laue photographs of single crystals of ice, and confirmed the view proposed by his predecessors Tamman, Bridgman, Bragg, and others that the ordinary ice crystal belongs to the hexagonal system. He went a step further and says that there seems to be every reason to believe that the ice crystal is D^4_{6h} (Dihexagonalbipyramidal). But Brandenburger[6] opposed Barnes's view, saying that the photographs taken by the latter provide little basis for such conclusion.*"

"Mügge[10] is opposed to the hexagonal-system theory and considers that ice is to be assigned to the rhombohedral system. Seljakov[11] conducted x-ray studies on ice crystals obtained by freezing water in various ways, and concluded that two modifications, α and β, exist in ordinary ice, α being hexagonal and β rhombohedral. Also Cohen and van der Horst[12] insist upon the existence of the ice crystal of the cubic system on the basis of their experiments. There are as yet various unclarified points with regard to the crystallization system of ice. It might be more complicated than is usually assumed."

U. Nakaya obtained some different forms of artificial snow crystals, one of them which apparently belonged to the rhombohedral system being produced in the case where the room temperature T_r was about -30°C and the supply of water vapour corresponded to the temperature of water T_w in the reservoir of about $+9$°C.

Another crystal form he obtained was rectangular and seemingly belonged to the cubic system. Such crystal was formed in 20 hr with T_r at about -28°C and with a scanty supply of water vapour, T_w being -6°C to $+7$°C. He also obtained rectangular skeletons among the frost crystals which were developed on the copper lid.

He concluded, however, that "although crystals of this type may be regarded as ice of cubic system, following Cohen, we cannot decide so by these photographs alone, because the hexagonal skeleton can grow to this rectangular shape if only one of its side is developed", and emphasized the necessity of further investigations along with x-ray studies.

Recently C. E. Hall[13] obtained the electron photomicrograph of ice crystals condensed at $-90° \sim -120$°C on the frozen distilled water surface by making use of replica and metal shadowing method. Judging from his photograph, they seem to belong to the cubic system. (In this connection, see also W. Rau's paper[14].)

The author observed by chance, in January 1954, some ice crystals of rectangular plane form apparently belonging to the cubic system, among many ice crystals which were produced by seeding the steam fog with silver iodide. After that we were able to confirm the existence of such crystal form in our low temperature

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* F. Rossmann[7] and F. Jona and P. Scherrer[8] inferred from their piezoelectrical examinations that, in contrast to Barnes's view, the crystal form of ice falls under the class C_{6v} (dihexagonalpyramid). It is to be pointed out that the existence of C_{6v} was confirmed already in 1935 by C. E. Morgan[9], through his electron-diffraction study.

laboratory. In the following, we shall describe the conditions under which these crystals were formed, giving some illustrations from the original photomicrographs obtained both in the field and in the laboratory.

II. Observations

In January, 1954, we performed some basic experiments on artificial precipitation of snow at Asahigawa in Hokkaido(15), in which the supercooled steam fog over the river was seeded with silver iodide particles. The air temperature during the period of experiments was about $-16°C$ to $-31°C$, and the air near the ground was estimated in most cases to be saturated with respect to ice but not to water, from the measurement of water content absorbed by phosphorus pentoxide.

In each experiment, many tiny ice crystals were observed in the neighbourhood of AgI generator, and the form of the crystals was mainly hexagonal prism and hexagonal plate, as identified by the aid of Schaefer's replica method(16).

Jan. 24th was the coldest day during the period. The air temperature was about $-30°C$ at 7 o'clock, the relative humidity being about 110% with respect to ice. Immediately after the AgI smoke was generated, and that during only ten minutes, we came across several crystals apparently having the form of thin rectangular plates. Fig. 1 and Fig. 2 show typical examples of such crystals.

When the silver iodide are smoked out, rapid appearance of a number of crystal embryos causes a considerable decrease of supersaturation of air in close proximity to the AgI generator. Therefore, the degree of supersaturation around the generator might have been much smaller than the above-mentioned value, that is, only a few per cent, when the rectangular plane crystals were observed. It may be assumed that the condition under which the rectangular plane crystals appeared was very similar to that of Nakaya's observation, although the form of crystals obtained by him was rectangular solid.

Being encouraged by this interesting fact, we attempted to reproduce the ice crystals of the same form in our low temperature laboratory,** and after a few trials succeeded in observing many ice crystals which seemingly belonged to the cubic system.

We used the low temperature room, kept at the temperature of about $-30°C$. The humidity in the room was always estimated to be just saturated with respect to ice, since the surface of the cooling tubes and other objects was constantly covered with hoar frost. When the door of the room was opened for an instant and the warm out-door air crept into the room, tiny ice crystals, some-

** We have five rooms (12 ft. long × 12 ft. wide × 9 ft. high) that can be kept, each independently, at any low temperature down to $-57°C$ by ammonia-refrigerating machines. Access to each room is through an "air lock" (6 ft. long × 6 ft. wide × 9 ft. high) maintained at a temperature intermediate between that of the room and the outside air. It was one of the air locks that was used for our experiments.

thing like "Diamond Dust", were observed to fall glittering in the electric light.

For the observation of crystal forms we used Schaefer's replica method like before. The several slide glasses covered with 1% replica solution were placed one above another at proper intervals so as to avoid catching broken pieces of the hoar frost directly falling from the cooling tubes on the slide glasses.

As the temperature in the room falls, the time required for the evaporation of ethylene dichloride increases. In our experiment at about −30°C, it took 3 to 5 hr to fix complete replicas.

After a few trials, we obtained on a slide glass many rectangular plane crystals considered to belong to the cubic system, besides hexagonal plane crystals (Fig. 3—Fig. 18). In this case, the room temperature was −27°C, the out-door air temperature being −3° to −5°C, so that the supply of water vapour when the door was opened must have been rather scanty. On this slide, none of the column-like crystals were observed, but only the plate-like crystals of both rectangular and hexagonal forms. The rectangular plates were greater in number than the hexagonal, while the former was about one-tenth in size of the latter, which presumably means that the rate of growth of rectangular plane crystals is comparatively much slower.

It is worthy to note that, while the condition of such crystal formation was very similar to that of Nakaya's observation, no rectangular solid forms made their appearance. For the critical examination of the conditions, under which the rectangular plates are developed, further investigations will be necessary. For the present, we intend to examine the crystals obtainable under various conditions by the use of polarization microscope, although the very existence of cubic ice crystals seems to have been established by our own observation as well as by Cohen.

Explanation of Plates

Plate I

Fig. 1, Fig. 2 (× 130) :

Ice crystals of rectangular plane form observed during the experiments at Asahigawa. Ordinary hexagonal prism and plates are seen in the left upper corner of Fig. 2.

Fig. 3—Fig. 18 (× 500) :

Ice crystals obtained in the low temperature laboratory.

Fig. 3, Fig. 4: Rectangular planes.

Fig. 5, Fig. 6: Rectangular planes with skeleton structure.

Plate II

Fig. 7, Fig. 8: Square planes.

Fig. 9: Rectangular plane and needles.

Fig. 10, Fig. 11, Fig. 12: Crystals of irregular form which are likely composed of several rectangular plates.

Plate III

Fig. 13: Simple hexagonal plane.

Fig. 14, Fig. 15: Hexagonal sectors.

Fig. 16: Deformed hexagonal plane.

Fig. 17, Fig. 18: Crystals of irregular form.

Acknowledgments

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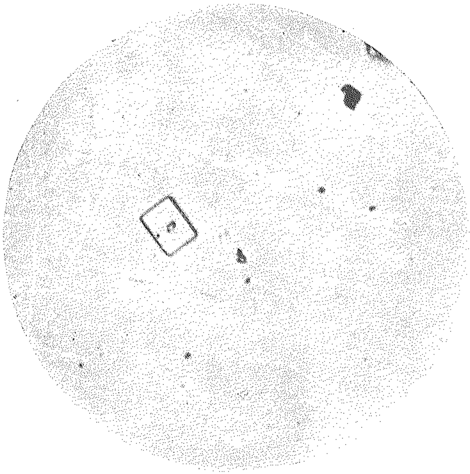


Fig. 1

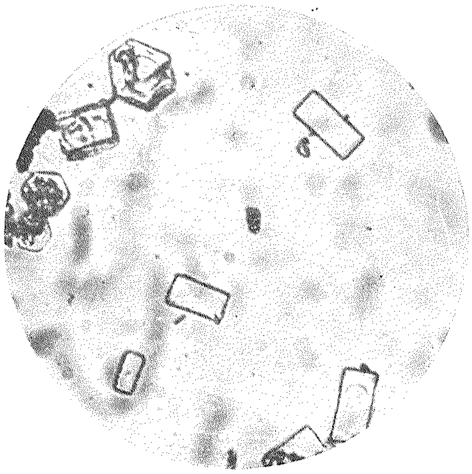


Fig. 2

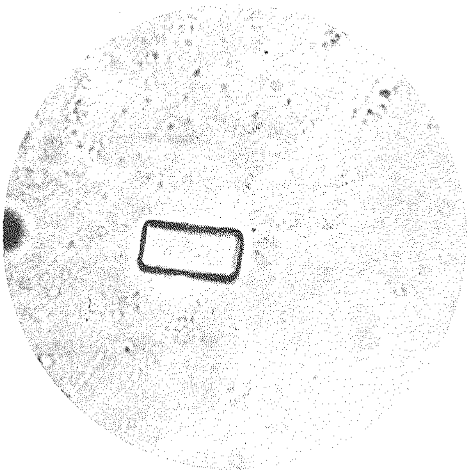


Fig. 3

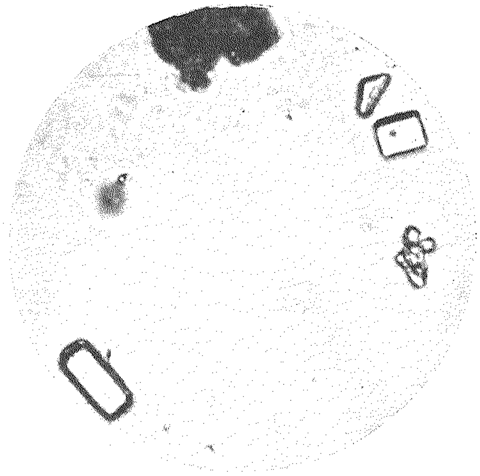


Fig. 4

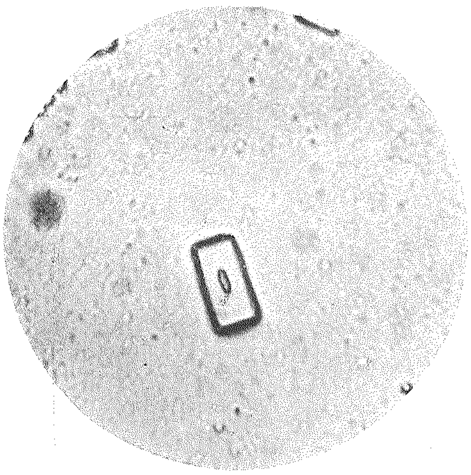


Fig. 5

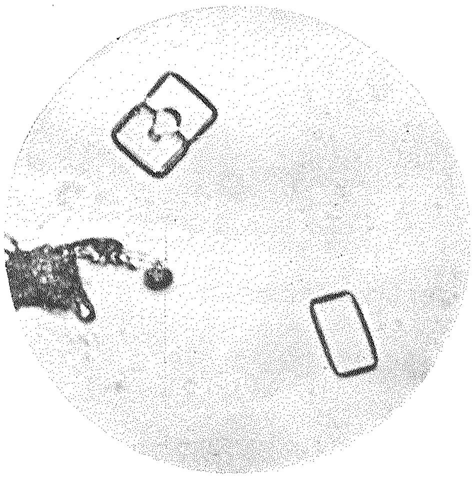


Fig. 6

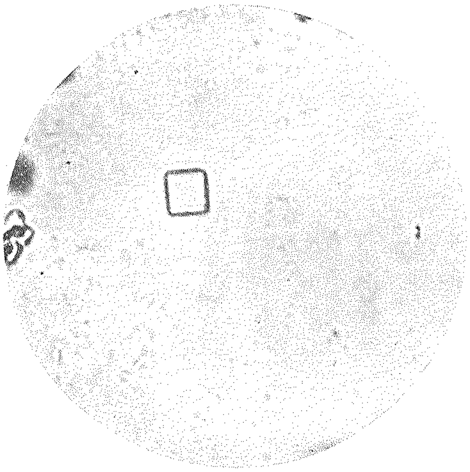


Fig. 7

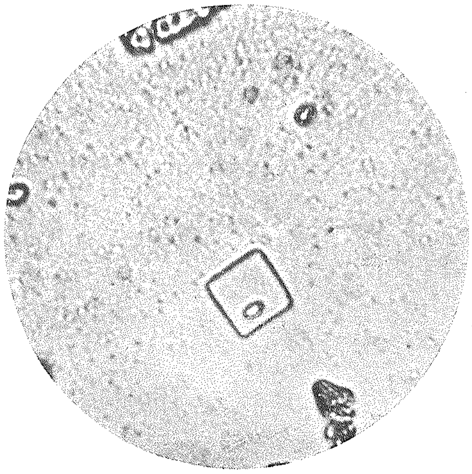


Fig. 8

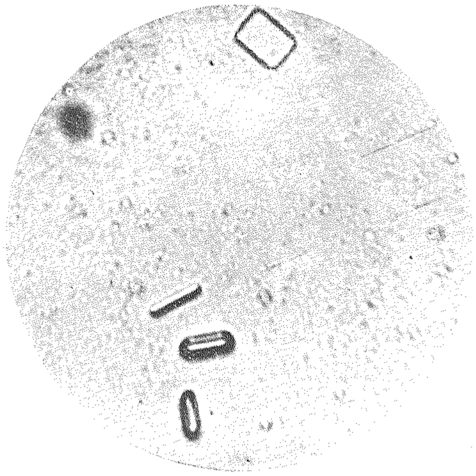
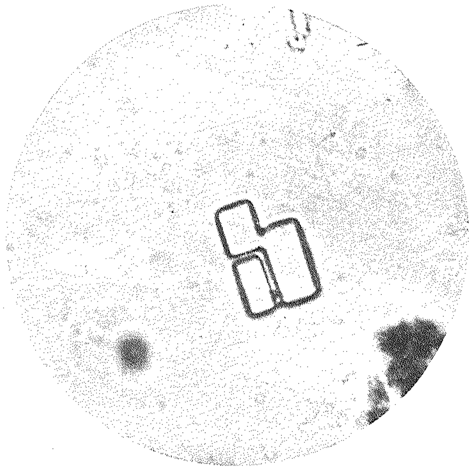


Fig. 9



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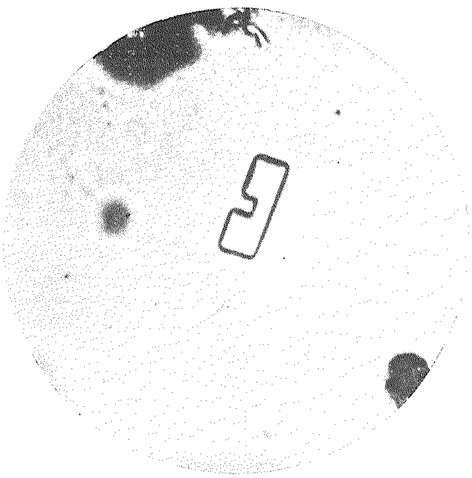

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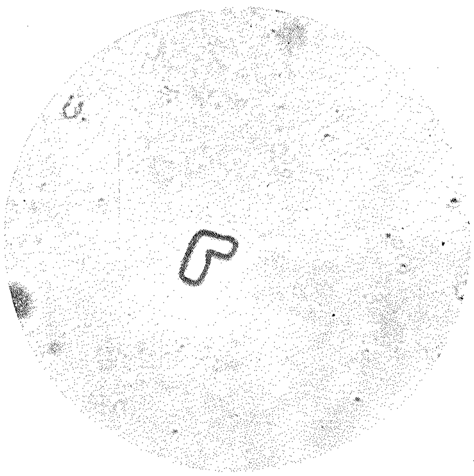

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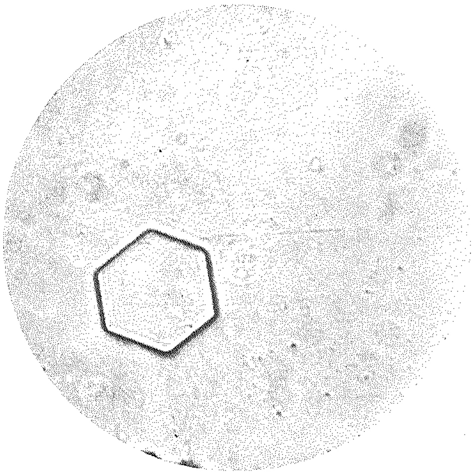
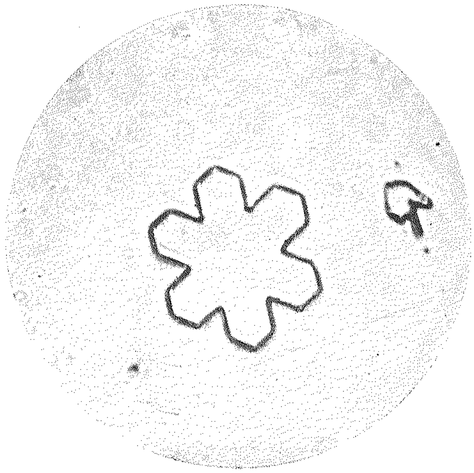


Fig. 13



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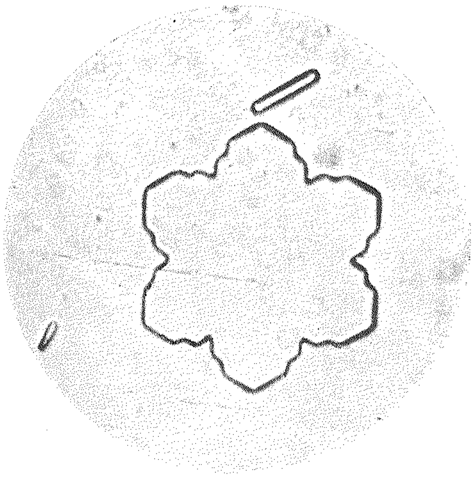

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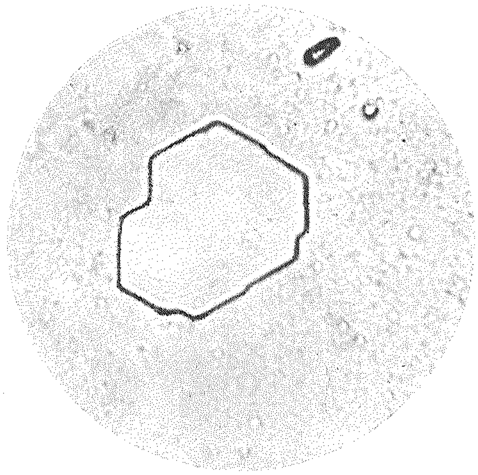

Fig. 16

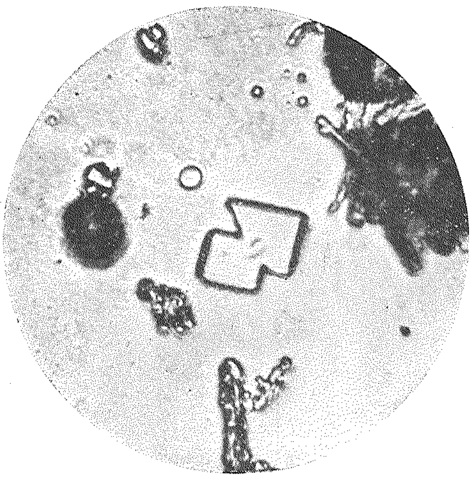

Fig. 17

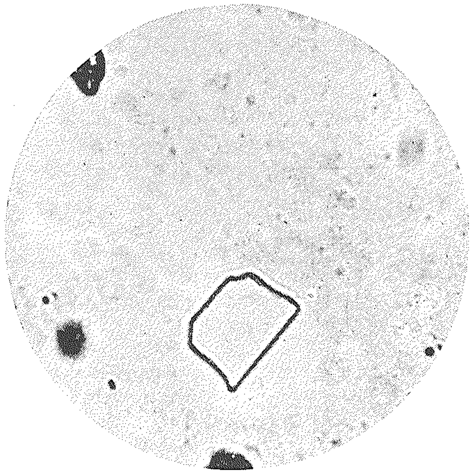

Fig. 18