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Some Considerations on Combination of Bullets which have the Axial Angle between the c-Axes of 90°

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Abstract

The arrangement of the a-axes and the c-axes of a combination of bullets in which the axial angle between the c-axes of the neighbouring two bullets was approximately 90° was examined. Samples examined were replicas of snow crystals collected by Kikuchi during over wintering from 1968 to 1969 at Syowa Station, Antarctica. In thirteen clear replicas of the examined samples, one of the a-axis of a bullet constituting a combination of bullets was in parallel to the c-axis of the another bullet when the axial angle between the neighbouring two bullets was about 90°. In two samples which were composed of more than four bullets, a pair of prism planes of two bullets whose axial angle was approximately 90° was almost in parallel with each other.

The axial angle between the c-axes of about 90° of a combination of bullets was explained by the combination of the c-axes constructed by the repetition of the formation of cubic structure on a basal plane. Further, the arrangement of the a-axes was determined with the same model. A possible three dimensional configuration of combination of bullets in snow crystals was considered.

1. Introduction

Recently, a number of considerations have been made on the formation mechanism of polycrystalline snow crystals. Several observations and experiments have been carried out in order to clarify the origin of polycrystalline snow crystals.

The observations of the axial angle between the c-axes of spatial branches of natural polycrystalline snow crystals were made carefully by Lee (1972). As a result, the peaks in the frequency distribution of the axial angle

around 70°, 55° and 40° were pointed out by Lee. The authors (1976 b) and Kobayashi et al. (1976 b) reconfirmed the results of the observations by Lee. Further, the authors (1979 a) ascertained that the axial angles between the c-axes of neighbouring two bullets of a combination of bullets were about 70°, whith minor excesses of about 55° and 40° by the examination of replicas of snow crystals collected at Syowa Station, Antarctica. And also, they pointed out that the axial angle of about 90° exsisted between the c-axes of neighbouring bullets. However, there have been few observations of the a-axes when the axial angle between the c-axes is 90°.

Polycrystalline snow crystals were expected to be grown from frozen cloud droplets. In order to clarify the origin of polycrystallization of supercooled water droplets, freezing experiments were carried out by many workers such as Hallett (1964), Higuchi and Yosida (1966), Aburakawa and Magono (1972), Murray and List (1972) and Uyeda and Kikuchi (1976 a). Further, the authors (1979 b) suggested a possible arrangement of the directions of the c-axes by the freezing experiment of hemispheric water drops.

Several workers have tried to explain the origin of polycrystalline snow crystals. Lee (1972) explained his observational results by a basal misfit theory. Iwai (1971) adopted the consideration of a penetration twin theory. On the other hand, Kobayashi et al. (1976 a) explained it by the "Generalized Coincidence Lattice Site (G.C.L.S.)" theory. Further, Kobayashi et al. (1976 b) proposed a cubic structure model as the origin of polycrystalline snow crystals.

The origin of polycrystalline snow crystals is becoming clear by observations of snow crystals, freezing experiments on water droplets and the consideration of models on the formation mechanism of polycrystalline snow crystals. The axial angles of about 70°, 55°, and 40° between the c-axes of neighbouring crystals of polycrystalline snow crystal have been examined carefully to the present. In addition to these, the examination of the directions of the a-axes, when the axial angle between the c-axes is about 90°, would help the comprehension of one of the formation mechanisms of polycrystalline snow crystals. With the knowledge obtained by this examination, a possible three dimensional arrangement of the c-axes of polycrystalline snow crystals which includes the axial angle between the c-axes of about 90° could be constructed.

In this paper, the directions of the a-axes were examined on the replicas of combination of bullets when an axial angle between the c-axes of the neighbouring bullets is about 90°. Then a possible three dimensional configuration of a combination of bullets was introduced.

2. Examinations

2.1 Samples and method

The samples examined here were replicas of snow crystals of a combination of bullets collected by one of the authors (K.K.) during over wintering from 1968 to 1969 at Syowa Station, Antarctica. Uyeda and Kikuchi (1979 a) examined the axial angles of about 70°, 55° and 40° between the c-axes of neighbouring two components of a combination of bullets with the same samples. They also noted the axial angles of about 90° between the c-axes of neighbouring two components of a combination of bullets.

In this paper, one of the possible arrangement of the a-axes and the c-axes was examined in the combination of bullets which have an axial angle of about 90° between the c-axes of neighbouring two components. The method of the examination was the same as that adopted by Uyeda and Kikuchi (1979 a). Namely, a three dimensional configuration of a combination of bullets was sketched first under a stereographic microscope and next microphotographs of them were taken.

The number of the combination of bullets examined which have the axial angle between the c-axes of about 90° was 46 crystals on the whole. Among them, 32 crystals were composed of three bullets and 14 crystals were composed of more than four bullets. Eleven crystals in the 32 crystals and two crystals in the 14 crystals were replicas clear enough to determine the directions of the a-axes and the c-axes. For these 13 crystals, an errangement of the a-axes and the c-axes was examined.

2.2 Results of examinations

Fig. 1 shows the examples of replicas of the combination of bullets with three bullets which have the axial angle of about 90° between the c-axes of neighbouring components. The directions of the a-axes and the c-axes of each component are illustrated on the right of each photograph of the crystals. Here, the arrows with symbols of C and C′ indicate the directions of the c-axes of the main and supplementary columns (bullets). The line segments $\overline{A_1A_2}$, $\cdots$, $\overline{A_6A_1}$ and $\overline{A_1'A_2'}\cdots$, $\overline{A_6'A_1'}$ indicate the directions of the a-axes of bullets. For all of the clear replicas with three components of bullets which have the axial angle between the c-axes of about 90°, one of the a-axes of the supplementary crystal was in parallel with the c-axes of the main crystal. For example, in Fig. 1(a) and (b), the angle between C and C′ was about 90° and the line segment $\overline{A_2'A_3'}$ or $\overline{A_5'A_6'}$ was parallel to the direction of C. In Fig. 1

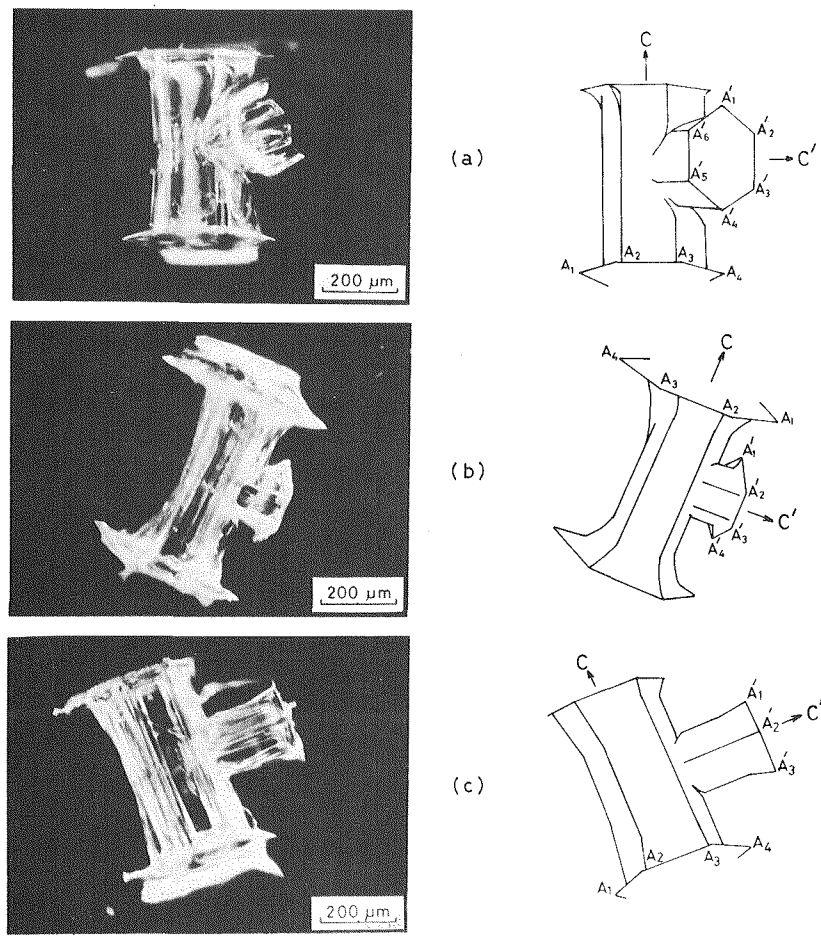

Fig. 1 Examples of replicas of a combination of bullets with three bullets which have the axial angle of about 90° between the c-axes of neighbouring components. The directions of the a-axes and the c-axes of the components are illustrated on the right hand side of each photograph.

(c), the angle between C and C′ was about 90° and the line segment $\overline{A_2A_3}$ of the main crystal was parallel to the direction of C′ of the supplementary crystals.

Fig. 2 shows the examples of replicas of the combination of bullets with more than four bullets which have the axial angle of about 90° between the c-axes of neighbouring components. The arrows with symbols of C, C′ and C″ in sketches on the right hand side indicate the directions of the c-axes

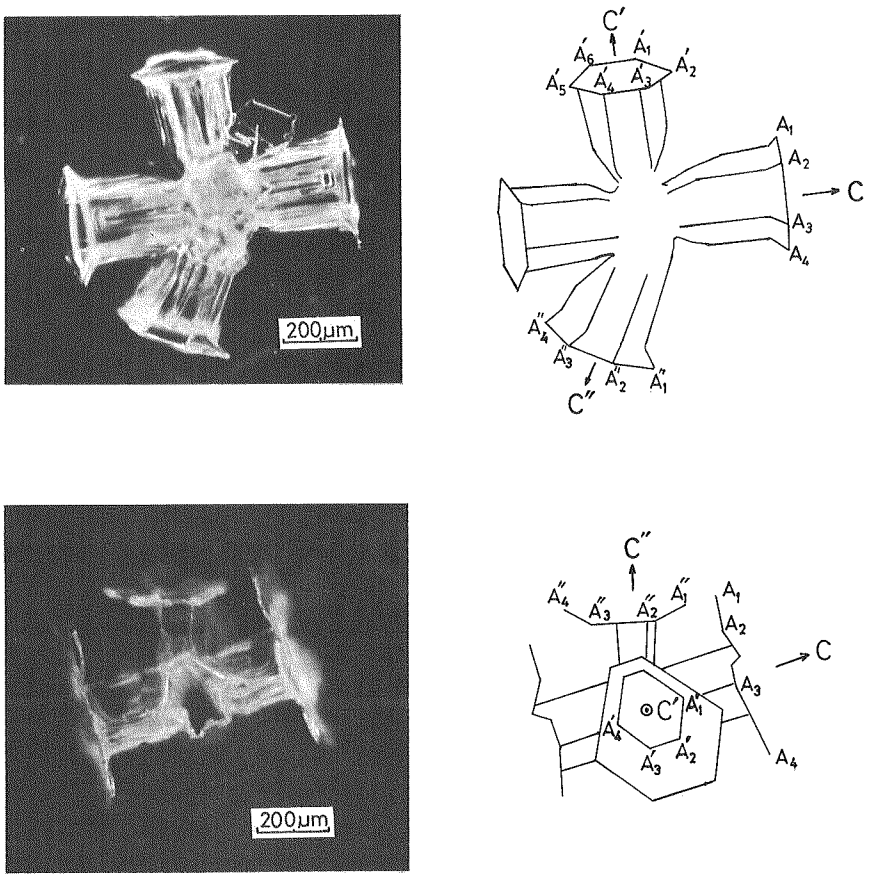

Fig. 2 Examples of replicas of a combination of bullets with more than four bullets which have the axial angle of about 90° between the c-axes of neighbouring components. The directions of the a-axes and the c-axes of the components are illustrated on the right hand side of each photograph.

similar to that described above examples. The line segments $\overline{A_1A_2}, \cdots, \overline{A_6A_1}$, $\overline{A_1'A_2'}, \cdots, \overline{A_6'A_1'}$ and $\overline{A_1''A_2''}, \cdots, \overline{A_6''A_1''}$ in these examples indicate the directions of the a-axes of bullets. For two clear replicas with more than four components of bullets which have the axial angle between the c-axes of about 90°, a pair of prism planes of two bullets was parallel to each other. For example, in Fig. 2(a), the angle between C and C′ directions is about 90°. The prism plane which is composed of C and $\overline{A_2A_3}$ and the prism plane which is composed of C′ and $\overline{A_3'A_4'}$ are almost parallel to each other. In Fig. 2(b),

the angle between C and C′ directions is about 90° and prism planes of two bullets, which are perpendicular to the plane of photograph, are almost parallel to each other.

Schematic figures of the combination of bullets which have the axial angle of 90° are shown in Fig. 3. One c-axis is perpendicular to the plane of Fig. 3 and another is on the plane of Fig. 3. In Fig. 3(a), one of the a-axes of one crystal is illustrated to be parallel to the c-axis of another crystal. In Fig. 3(b), on the other hand, two prism planes which are perpendicular to the surface of Fig. 3 are illustrated to be parallel to each other. The axial angle between a pair of the a-axes of neighbouring two crystals of Fig. 3 is listed in Table 1. The combination of bullets as shown in Fig. 1 corresponds to the type as illustrated in Fig. 3(a) and that as shown in Fig. 2 corresponds to the type as illustrated in Fig. 3(b).

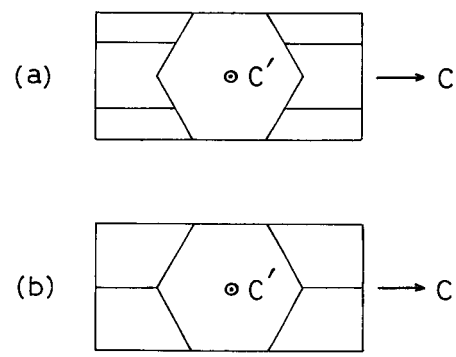

Fig. 3 Schematic figures of a combination of bullets with the axial angle of 90° between the neighbouring c-axes.

Table 1 The axial angle between a pair of the a-axes of neighbouring crystal illustrated in Fig. 3. The sets of symbols of (a_1, a_2, and a_3) and (a_1', a_2', and a_3') indicate the a-axes of the bullets shown in Fig. 3. The angle of (a) and (b) corresponds to Fig. 3 (a) and Fig. 3(b), respectively.

(a)

	a_1	a_2	a_3
a_1'	30°	64°	64°
a_2'	90°	90°	90°
a_3'	30°	64°	64°

(b)

	a_1	a_2	a_3
a_1'	90°	41°	41°
a_2'	90°	90°	90°
a_3'	90°	41°	41°

3. Explanation of the arrangement

The arrangement of the a-axes of the combination of bullets which has the axial angle between the c-axes of about 90° was made clear by careful scrutinization. The arrangement has to satisfy not to contradict a proposed model which explains the axial angles of about 70°, 55° and 40° between the c-axes. In the experiment by Uyeda and Kikuchi (1979 a), frozen water droplets became polycrystal when they were seeded with a basal plane of an ice needle. And a possible arrangement of the c-axes of a frozen water droplet which includes the axial angles of about 70°, 55°, and 40° between the c-axes was suggested. On the other hand, the formation mechanism of cubic structure on a basal plane was proposed by Kobayashi and Takahashi (1980).

In the present paper, the polycrystallization in a frozen water droplet was assumed to be formed according to the cubic structure model on a basal plane of ice. The processes of formation of several kinds of axial angles between the c-axes were considered by the following four steps.

1) When polycrystallization commenced at one point on a basal plane of a hexagonal ice, as expected by the cubic structure model, three directions of the c-axes would be constructed in the water droplet. The axial angle between the c-axes of the substrate and the c-axes of each crystal constituting the frozen water drop is about 70°. This explains the axial angle of about 70° (complementary angle of 110°) between the neighbouring two crystals of the frozen water droplet.

Fig. 4 shows a schematic figure of this arrangement of the directions of the c-axes. The double arrows are projections of directions of the c-axes (C_1, C_3, and C_5) onto the basal plane. The axial angle between the c-axis of the substrate and the c-axis of each of C_1, C_3, and C_5 is about 70°. The axial angles between any combination of two c-axes of C_1, C_3, and C_5 are about 70° (complementary angle of 110°) again. The relation explained above is equivalent to the c-axes of C_2, C_4, and C_6. This angle corresponds to the maximum peak of the frequency distribution of the axial angle between the c-axes of spatial branches of natural polycrystalline snow crystal.

2) When polycrystallization commenced at two points on two basal planes with an odd layer difference, the axial angles of about 55° and 40° would be constructed between a pair of the c-axes which are constituted at each nucleation point. In Fig. 4, two pairs of the c-axes stand on two basal planes with an odd layer difference. The arrangement of the c-axes of C_2, C_4, and C_6 coincides exactly a 60° rotation of the c-axes of C_1, C_3, and C_5 around the c-axis

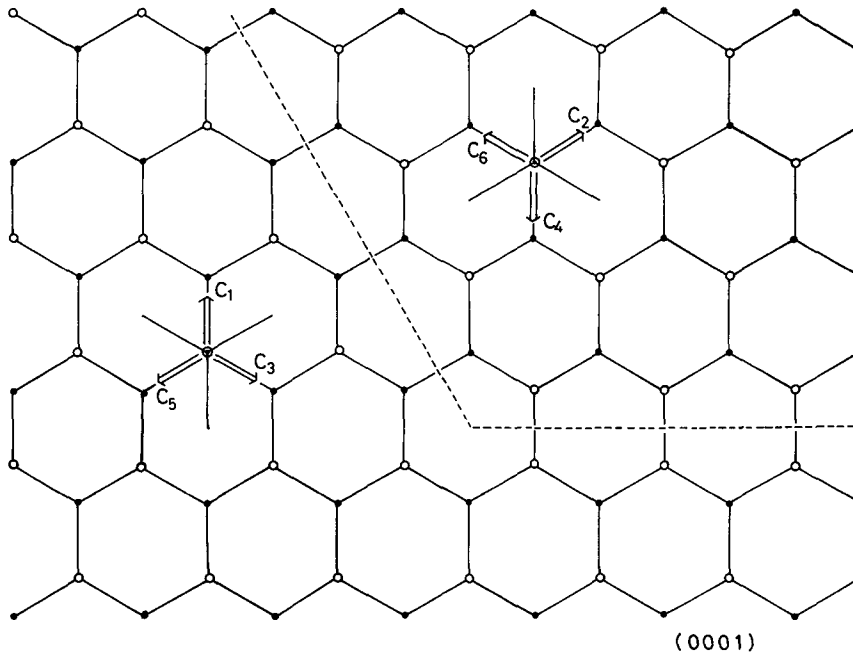

Fig. 4 Projection of directions of the c-axes ($C_1, \cdots, C_6$) formed on basal planes of a substrate. The one layer different basal planes are divided by the broken line. The angles between the c-axis of a substrate and each of $C_1, \cdots, C_6$ are about 70°. The axial angles between C_1 and C_3, C_2 and $C_4, \cdots$, are about 70°, the angle between C_1 and C_2, C_2 and $C_3, \cdots$, are about 55° and the angle between C_1 and C_4, C_2 and $C_5, \cdots$, are about 40°.

of substrate. The actual axial angles not projected on (0001) plane between the directions of C_1 and C_2, C_2 and $C_3, \cdots$, and C_6 and C_1 are about 55°, respectively and that of C_1 and C_4, C_2 and C_5, and C_3 and C_6 are about 40°, respectively. These angles correspond to the peaks of the frequency distribution of the axial angle between the c-axes of about 55° and 40° of natural polycrystalline snow crystals.

3) When the cubic structure is formed again on the basal plane of the crystal which were grown on the basal plane of substrate, the axial angle of about 90° between the neighbouring c-axes would be constructed. The arrangement of the c-axes is illustrated in Fig. 5. The directions of the c-axes of C_1, $C_2, \cdots$, and C_6 indicate the directions of the c-axes of C_1, $C_2, \cdots$, and C_6 in Fig. 4. If a cubic structure is formed again on a basal plane of the crystal, which c-axis is one of the c-axes of C_1, $C_2, \cdots$, and C_6, six new directions

of the c-axes would be made as shown on the right hand side in Fig. 5. Here, six new directions C_1', C_2',$\cdots$, and C_6' are taken toward C_1. Also, the angle between C_6 and any of C_1', C_2',$\cdots$, and C_6' are shown.

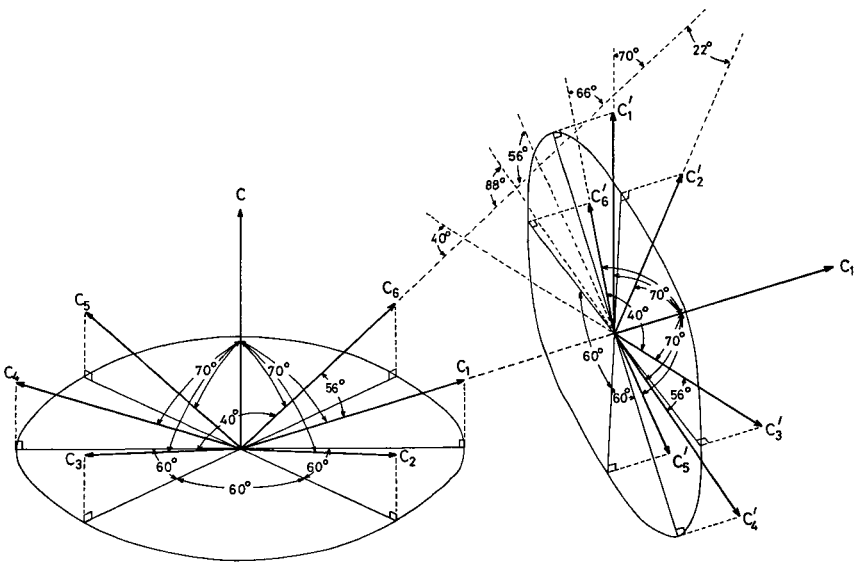

Fig. 5 An arrangement of the c-axes. The arrow symbolized as C indicates the direction of the c-axis of the substrate. The arrows symbolized as $C_1,\cdots,C_6$ indicate the c-axes formed on the basal plane of the substrate. The arrows symbolized $C_1',\cdots,C_6'$ indicate the c-axes formed on the basal plane of the crystal in which the c-axis is C_1.

Table 2 The list of the axial angles between the c-axes of one of $C_1,\cdots,C_6$ and one of $C_1',\cdots,C_6'$ in Fig. 5.

	c_1'	c_2'	c_3'	c_4'	c_5'	c_6'
c_1	70°	70°	70°	70°	70°	70°
c_2	70°	66°	56°	88°	40°	22°
c_3	70°	56°	0°	56°	70°	40°
c_4	70°	88°	56°	32°	56°	88°
c_5	70°	40°	70°	56°	0°	56°
c_6	70°	22°	40°	88°	56°	66°

The respective axial angles between one of C_1, C_2,$\cdots$, and C_6 and one of C_1', C_2',$\cdots$, and C_6' are listed in Table 2. According to this, the axial angles of 88°, 66°, 32°, and 22° are constructed beside 70°, 56° and 40° as expected by the cubic structure model. From this table, the frequency of 88° is more

twice as many as that of about 32° and 22°.

The axial angles between the a-axes are easily calculated with the cubic structure model for each axial angle between the c-axes. The axial angles between the a-axes when the axial angle between the neighbouring c-axes is 88° are shown in Table 3. These relations correspond to the arrangement of the c-axes and the a-axes of combination of bullets as shown in Fig. 1, and to the illustration of Fig. 3 (a).

Table 3 The axial angles between the a-axes, when the axial angle between the neighbouring c-axes is 88°.

	a_1	a_2	a_3
a_1'	34°	87°	27°
a_2'	60°	90°	60°
a_3'	71°	87°	67°

4) If it is possible for six new c-axes to be formed on C_1 in Fig. 5, there is the same possibility for six new c-axes to be formed on each of C_2, C_3,$\cdots$, and C_6. For the calculation of directions of these c-axes, C_1', C_2',$\cdots$, and C_6' are rotated 60°, 120°, and 180° around the c-axes of substrate in order to make six new directions of the c-axes on the basal planes of C_2, C_3, and, C_4.

Let's symbolize the six new c-axes on C_2, C_3, and C_4 as (C'_{21},$\cdots$, and C'_{26}), (C'_{31},$\cdots$, and C'_{36}) and ($C'_{41}\cdots$, and C'_{46}) respectively. By the combination of two of new c-axes, the axial angle between the c-axes is calculated. The axial angle between the following pairs is 88° (about 90°); C'_{12}-C'_{25}, C'_{13}-C'_{23}, C'_{13}-C'_{26}, C'_{14}-C'_{23}, C'_{15}-C'_{24}, C'_{16}-C'_{25}, C'_{12}-C'_{32}, C'_{12}-C'_{34}, C'_{14}-C'_{36}, C'_{16}-C'_{36}, C'_{13}-C'_{44}, C'_{14}-C'_{43}, C'_{14}-C'_{45} and C'_{15}-C'_{44}. The axial angle between the a-axes for all of these are equivalent to that listed in Table 3. This is the type as shown in Fig. 1 and illustrated in Fig. 3(a). The axial angle of C'_{12} and C'_{14} and of C'_{14} and C'_{26} is 86.5° (about 90°). These axial angles between the a-axes are shown in Table 4. This is the type shown in Fig. 2 and illustrated in Fig. 3(b). The same can be said for C_5 and C_6 as C_2 and C_3 by symmetry.

Table 4 The axial angle between the a-axes, when the axial angle between the neighbouring c-axes is 86.5°.

	a_1	a_2	a_3
a_1'	79°	87°	76°
a_2'	50°	87°	54°
a_3'	34°	90°	34°

The axial angles between the c-axes of a combination of bullets are explained by the combination of the c-axes produced by the process of the step 4).

4. Concluding remarks

An arrangement of the a-axes and the c-axes of a combination of bullets in natural snow crystals which have the axial angle of about 90° between the c-axes of neighbouring two bullets was examined using replicated snow crystals. On thirteen samples of clear replica, one of the a-axis of a bullet constituting a combination of bullets was parallel to the c-axis of another bullet when the axial angle between the neighbouring two bullets was about 90°. In two samples which were composed of more than four bullets, a pair of prism planes of two bullets whose axial angle was about 90° was almost parallel to each other.

Several kinds of axial angles between the c-axes of combination of bullets were explained by the combination of the c-axes which were constructed by the repetition of the formation of the cubic structure on a basal plane. At the same time, the arrangement of the a-axes was determined with this model. A possible three dimensional configuration of combination of bullets was constructed by a selection of one of the c-axes in Fig. 5 and one of the c-axes, which was formed by rotation of $\pm 60°$, $\pm 120°$ and 180° of C_1',$\cdots$, and C_6' in Fig. 5 around the principal c-axis of the substrate. The model explained in the section 3 is available for the understanding of three dimensional configuration of combination of bullets and the formation process of polycrystallization in water droplets. As a result the axial angles between the c-axes constructed by this model are the same as the axial angles between the c-axes formed by the repetition of m-c-c-m type of junction suggested by Kobayashi et al. (1976 b).

By the combination of the c-axes explained above, the axial angles of 66°, 32° and 22° between the c-axes were constructed. These angles, however, were not clear by the examination of replicas used in this paper. These angles may be difficult to be found by the examination of replicas because precise observations of a sample from different directions other than the direction of perpendicular to the slide glass are difficult. On the other hand, the axial angle between the c-axes of about 90° is clear, because the percentage of the combination of bullets which have the axial angle between the c-axes of about 90° was 17% of that of replicas collected in the Antarctica (Uyeda and Kikuchi (1979 a)). One of the reasons for this may be that

the percentage of the axial angle between the c-axes of about 90° is more than twice of that of 66°, 32°, and 22°.

By repetition of the formation of the cubic structure on basal planes, many kinds of the axial angle between the c-axes are considered to be constructed. By decreasing of freezing temperature, the repetition of formation of the cubic structure is considered to occur many times. Steps (1) and (2) explain the axial angle between the c-axes of about 70°, 55°, and 40°. And Steps (3) and (4) explain the axial angle between the c-axes of about 90°. However, the consideration to the Step (4) of the section 3 would be enough to explain the three dimensional configuration of combination of bullets. In order to ascertain whether there are the axial angle of 66°, 32°, and 22° and other angles between the c-axes of neighbouring bullets, further observations of combination of bullets would be necessary.

Although replicas of combination of bullets were examined, the three dimensional arrangement of the c-axes proposed would be applicable to the explanation of other polycrystalline snow crystals. Polycrystalline snow crystals are considered to be made from the frozen cloud droplets, in which polycrystallization was formed by the processes as explained above. If the arrangement of the directions of the c-axes is determined in a cloud droplet at a freezing temperature, the type of snow crystals, such as combination of bullets, radiating assemblage of dendrite and so on would be determined according to the growth condition.

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References

Aburakawa, H. and C. Magono, 1972. Temperature dependency of crystallographic orientation of spatial branches of snow crystals. J. Meteor. Soc. Japan, **50**, 166–170.

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. Physics of Snow and Ice, Vol. 1 (Inst. Low Temp. Sci., Hokkaido Univ., Sapporo), 79–93.

Iwai, K., 1971. Note on snow crystals of spatial type. J. Meteor. Soc. Japan, **49**, 516–520.

Kobayashi, T., Y. Furukawa, K. Kikuchi and H. Uyeda, 1976a. On twinned structures in snow crystals. J. Crystal Growth, **32**, 233–249.

Kobayashi, T., Y. Furukawa, T. Takahashi and H. Uyeda, 1976b. Cubic structure models at the junctions in polycrystalline snow crystals. J. Crystal Growth, **35**, 262–267.

Kobayashi, T. and T. Takahashi, 1980. On the boundary structure of polycrystalline snow crystals and its formation mechanism. J. Jap. Assoc, Crystal Growth, **6**, 65–74.

Lee, C.W., 1972. On the crystallographic orientation of spatial branches in natural polycrystalline snow crystals. J. Meteor. Soc. Japan, **50**, 171–180.

Murray, W.A. and R. List, 1972. Freezing of water drops. J. Glaciology, **11**, 115–129.

Uyeda, H. and K. Kikuchi, 1976 a. On the orientation of the principal axis of frozen water droplets. J. Meteor. Soc. Japan, **54**, 267–275.

Uyeda, H. and K. Kikuchi, 1976 b. Remeasurement of axial angle between spatial branches of natural polycrystalline snow crystal. J. Fac. Sci., Hokkaido Univ., Ser. VII, **5**, 21–28.

Uyeda, H. and K. Kikuchi, 1979 a. Observations of the three-dimensional configuration of snow crystals of combination of bullet type. J. Meteor. Soc. Japan, **57**, 488–492.

Uyeda, H. and K. Kikuchi, 1979 b. Measurements of the principal axis of frozen hemispheric water droplets. J. Meteor. Soc. Japan, **58**, 52–58.