

## Origin of optical variations in grossular-andradite garnet

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### Abstract

Optical textures in birefringent grossular-andradite garnets were studied from the standpoint of crystal growth. Grossular from Eden Hill, Vermont, shows one-to-one correlation between growth surface features consisting of well-defined pyramidal hillocks and the internal texture. Grossular from Fujikaramari, Japan, consists of growth sectors with fine lamellae which apparently correspond to growth steps. Andradite from Kamihogi, Japan, is composed of many triangular sectors which have the character of so-called lineage structure. The three specimens show sector twins whose planes are parallel, normal, or inclined to the [001] axis in a thin section parallel to the (110) growth plane. The texture can be interpreted by a general mechanism of formation of order-disorder growth sectors suggested by Akizuki (1981a). That is, various degrees of  $\text{Fe}^{3+}/\text{Al}$  ordering in the octahedral cation sites were produced on the side faces of surface steps during growth.

### Introduction

Optically non-cubic garnets, showing birefringence, have been reported by many mineralogists and petrologists since the last century. Brauns (1891) summarized optical studies of birefringent garnets with sectors, and Meagher (1980) reviewed recent studies. Birefringence is especially common in grossular-andradite garnets (grandite garnet), though garnets near both the end-member compositions are isotropic. Some grandite garnets show alternating isotropic and birefringent lamellae parallel to the {110}<sup>1</sup> growth planes (e.g., Murad, 1976). Also, grandite garnets show optical growth sectors which correspond to the {110} faces with or without {211} faces. Birefringent grandite garnet was transformed irreversibly into optically isotropic garnet by heating at 860°C by Stose and Glass (1938), though the birefringence was still observable in some garnets after quenching from a temperature of 1060°C (Ingerson and Barksdale, 1943).

The origin of birefringence in garnets has been variously attributed to residual strain in the structure (Chase and Lefever, 1960; Kitamura and Komatsu, 1978) and ordering of octahedral cations,  $\text{Fe}^{3+}$  and Al, thereby reducing the symmetry (Takéuchi et al., 1982). Takéuchi et al. (1982) refined the structures of some grandite garnets with X-ray data and found cation ordering. In grandite garnet from Munam, North Korea, the content of  $\text{Fe}^{3+}$  in the eight non-equivalent octahedral positions varied from 15(1)% to 49(1)%, and the space group was  $\bar{1}I$ .

Winchell and Winchell (1951) summarized the sectoral twinning in birefringent garnets, though fine twinning in each sector was not described. Little is known about the

origin of sectors and twinning, although Takéuchi et al. (1982) suggested that the twinning of garnets they used for X-ray refinement was produced during a transformation from disordered to ordered structure after crystal growth.

Through studies of a number of minerals showing so-called optical anomalies, such as adularia (Akizuki and Sunagawa, 1978), topaz (Akizuki et al., 1979), analcime (Akizuki, 1981a), and chabazite (Akizuki, 1981b), it was found that the internal sectors observable between crossed nicols correspond to growth planes and furthermore that fine textures correspond to the various vicinal faces on the growth planes, on which various degrees of cation or anion ordering may occur during growth. Also, a general mechanism of formation of order-disorder growth sectors was suggested by the present writer (Akizuki, 1981a).

Two problems of optically "abnormal" garnets are considered here: (1) why is the optical symmetry lower than that usually shown by X-ray analyses and morphology, and (2) what is the origin of the complicated textures that the "abnormal" garnets show without exception? The former question was explained in terms of  $\text{Fe}^{3+}/\text{Al}$  ordering (Takéuchi et al., 1982), while the latter has remained an open question since the last century.

The objective of the present paper is to describe the growth sectors and optical properties of some grandite garnets showing distinct birefringence and to discuss the origin of the optical sectors and twinning using the general mechanism suggested by Akizuki (1981a).

### Observations

Although birefringent grandite garnets from several localities were studied, only the garnets from three local-

<sup>1</sup> Throughout this paper indices refer to the ideal isometric axes.

ities are described in this paper. Most observations were made on thin sections (30  $\mu\text{m}$  thick) parallel and normal to the growth surface (110).

### Grossular from Eden Hill, Vermont

The garnet has cinnamon color and transparent dodecahedral form {110}, which is the so-called hessonite type, crystals are as large as 5 mm in diameter, and chemical composition is  $\text{Gr}_{85.7}\text{And}_{14.3}$ . The surface microtopography of crystal faces was studied with reflection-type interference contrast microscopy. The (110) growth surface is characterized by growth hillocks elongated parallel to the [001] direction (Fig. 1a). Each hillock consists of four vicinal faces in a pyramidal form. The faces intersect in edges parallel to [001] and at about  $50^\circ$  to [001] (in projection perpendicular to (110)). Some indistinct boundaries, across which optical contrast differs, are shown by white lines at the lower-left on Figure 1a. Summits of growth hillocks are observed at the intersections of the boundaries. There are six summits between two arrows marked A and A'. Also, the summit of a large hillock with some growth layers in the central area in Figure 1a is shown by arrow B.

A thin section parallel to the growth surface was prepared from the specimen, whose as-grown face was cemented on a glass slide, and was observed on the polarizing microscope with a universal stage in order to measure the optical orientation. Figure 1b shows a photomicrograph taken with crossed polars from the same area as that in Figure 1a. A clear correlation is observed between growth features (Fig. 1a) and the internal textures (Fig. 1b). Growth hillocks with four vicinal faces show internal fourling sectors between crossed polars, the fourling centers corresponding to the growth hillock summits. Six centers of fourling sectors between A and A' of Figure 1b correspond to the six summits of growth hillocks shown in Figure 1a. Likewise, the center shown by arrow B in Figure 1b corresponds to the summit of the growth hillock B in Figure 1a. Although the fourling vicinal faces cannot be observed clearly on the photograph (Fig. 1a), because of the small difference of inclination between two of the vicinal faces, the fourling sectors are clearly observed between crossed polars (Fig. 1b). The indistinct boundaries shown by the white lines at the lower-left in Figure 1a correspond to sector boundaries inclined about  $50^\circ$  to [001] in Figure 1b. The angle of these boundaries varies from  $90^\circ$  to about  $50^\circ$  with respect to the [001] axis from specimen to specimen. The growth hillock with summit B developed at a late stage of crystal growth, and one of the vicinal faces covered part of the growth hillock with summit indicated by a letter C (Fig. 1a). The sector boundary under the vicinal face is shown by a broken line. The growth hillock at C is much thicker as compared with the vicinal face covering the hillock C, and therefore the sector boundary corresponding to the broken line is distinct in Figure 1b. Frequently, a growth

feature varies during different stages of crystal growth, and therefore the correlation between optical properties and surface topography is not always good in some minerals (see Akizuki, 1981a). However, from the present observations, it is clear that the internal textures were produced during growth.

The optical orientation and  $2V$  value in the light-colored sectors of Figure 1b are shown in Figure 2. The optical vibration direction  $X$  is normal to the growth plane (110), and the other two directions,  $Y$  and  $Z$ , rotate up to  $4^\circ$  from [001] and  $[\bar{1}10]$  around the  $X$ -axis. The  $2V_z$  value

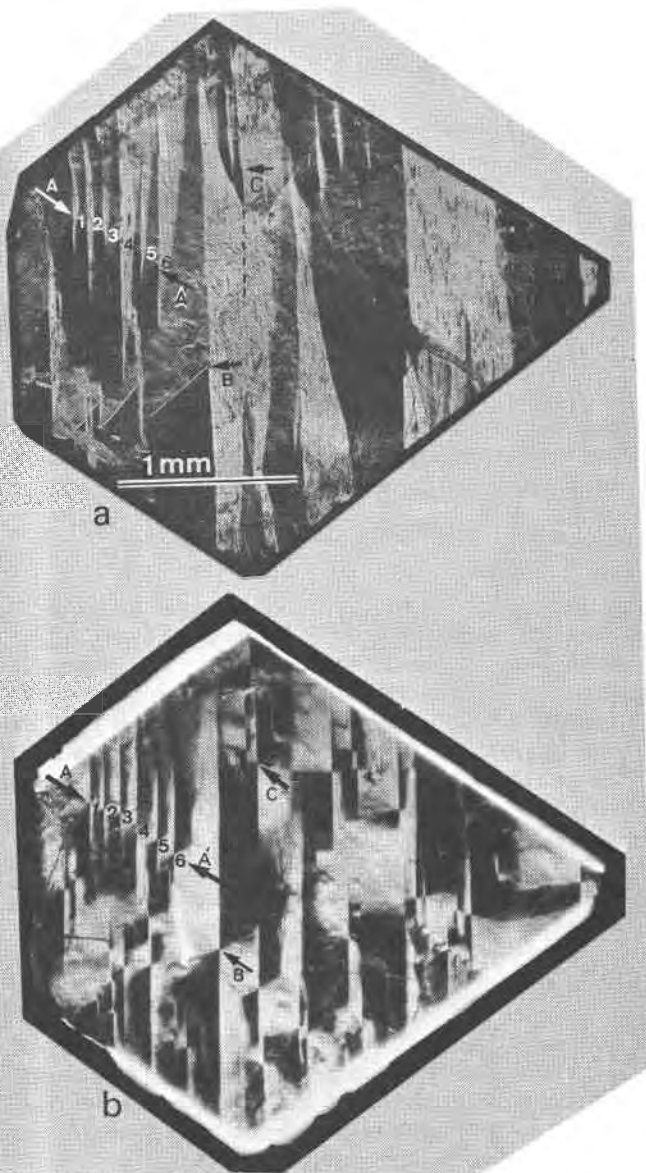


Fig. 1. Corresponding reflection interference-contrast (a) and cross polarized transmission (b) photomicrographs of a (110) section of garnet from Eden Hill. [001] is vertical, and the polarizing plane is rotated  $3^\circ$  clockwise from vertical. See text for details.

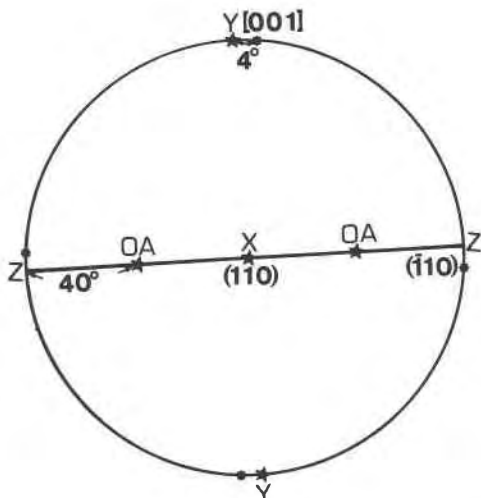


Fig. 2. Stereographic projection of the optical properties measured on a (110) thin section of garnet from Eden Hill. Principal vibration direction ( $X$ ) is normal to the (110) growth surface and optic axes are indicated by OA.

is (+)80°. The four sectors corresponding to the four vicinal faces on the growth hillocks are reflection twins to each other. Other transparent grandite garnets from Torino, Italy, showed a texture similar to that of Eden Hill garnet. However, detailed study was impossible, because of the small size of the crystals.

#### *Grossular from Fujikaramari, Fukushima Prefecture, Japan.*

The specimens, which are red-brownish and translucent, have well-developed {110} faces with small {211} faces and are about 10 mm in diameter. The {110} faces have some irregular growth steps and the {211} faces have fine striations. Fine cracks, some of which are filled by calcite, are abundant in the crystal. Growth bands parallel to the (110) face are due to chemical variations, which were observed with an electron microprobe. The average chemical composition is  $Gr_{79.7}And_{20.3}$ .

Figure 3a shows fine growth steps on the (110) surface, whose growth center seems to exist near the crystal edge. In general, the crystal faces do not show regular growth patterns such as those of Eden Hill garnet. Frequently, some straight lines, which continue through several crystal faces, are observed to modify the growth steps slightly. The straight lines may be produced during crystal growth by slip. Such straight lines were not observed in garnets from the other two localities. A cross-polarized microphotograph shows fine lamellae correlated with the growth steps on the surface (Fig. 3b). The internal textures of the fine lamellae and their optical properties are described below.

Figure 4 shows another thin section with growth sectors corresponding to the outline of the (110) face. The many fine black lines are due to irregular cracks. Triangu-

lar sectors, shown by arrows, correspond to one of the fourling sectors produced on the four vicinal faces of Eden Hill garnet (Fig. 1b). The sectors shown by stars have rims parallel to the outline of the (110) plane. A magnified schematic sketch of the texture in the rectangle of Figure 4 is shown in Figure 5. The extinction directions shown by arrows are inclined to [001] in the (110) thin section, though they are nearly parallel to [001] in some areas. Lamellae with different extinction angles repeat alternately parallel to one of the outlines of {110} faces in each sector. Thus, a clear correlation exists between orientations of the lamellae and extinction (Fig. 5). One of the growth sectors shown in Figure 5 has a rim with two extinction angles inclined 15° to the (110) outline. This suggests that a thick hillock with large faces existed on the surface during growth. The fourling sectoral texture as found in Figure 1b was not observed in this specimen.

Figures 6a and 6b show the sectoral textures of another

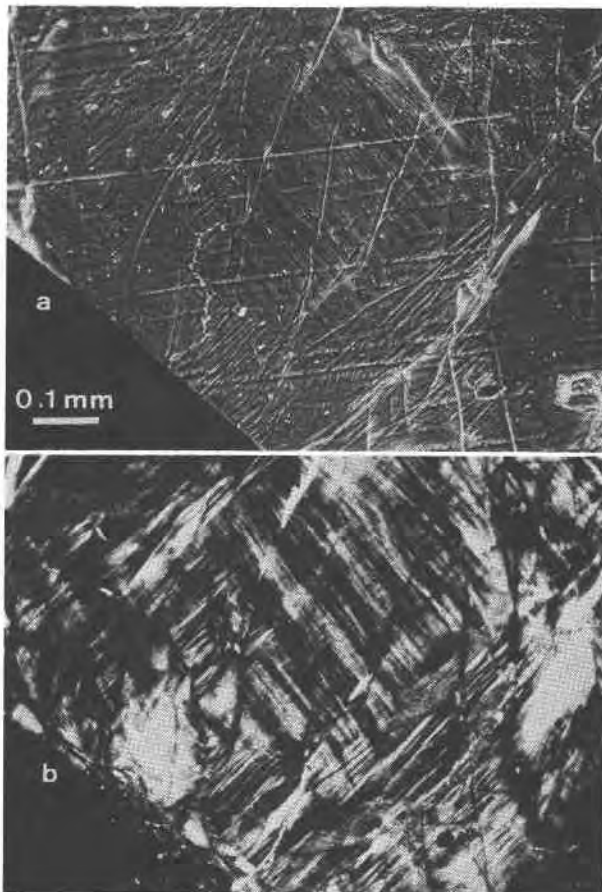


Fig. 3. Corresponding reflection interference-contrast (a) and cross polarized transmission (b) photomicrographs of a (110) section of garnet from Fujikaramari. [001] is vertical, and the polarizing plane is rotated 4° clockwise from vertical. Roughly horizontal lines may be produced by crystal slip. Roughly vertical lines are due to cracks. See text for details.

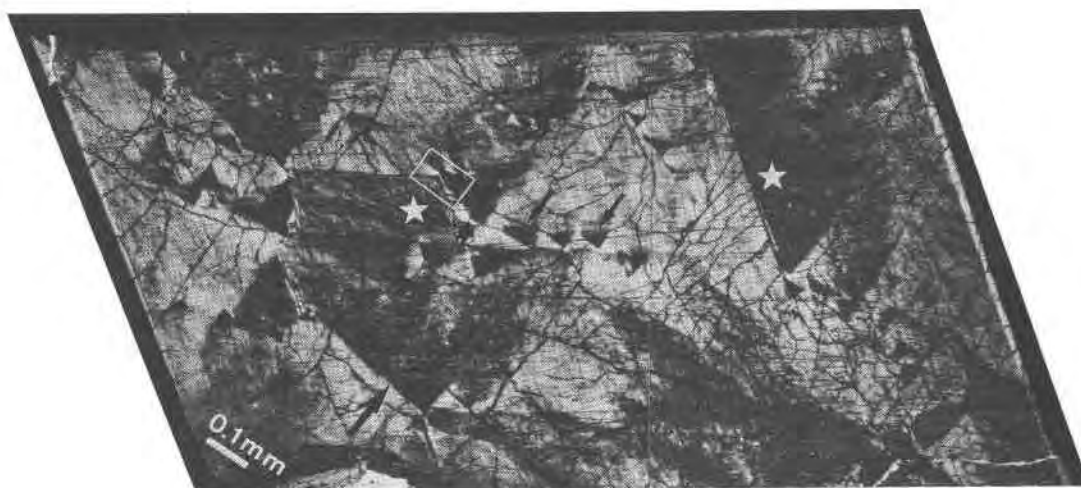


Fig. 4. Cross polarized photomicrograph of a thin section parallel to the growth surface (110) of a garnet from Fujikaramari. Black arrow is [001], and the polarizing plane is rotated 12° counter-clockwise from vertical. The three edges of the crystal with black bands are natural, while the fourth is cut. See text for details.

specimen observed between crossed polars. The crystal is divided into sectors by vertical boundaries, and extinction angles are roughly symmetrical with respect to [001] on the (110) face. Two kinds of lamellae alternate in the sectors (Fig. 6b). A magnified photograph of the rectangular area in Figure 6a is shown in Figure 7. Both elongation direction and optical extinction of the lamellae are roughly symmetrical with respect to the sector boundaries in some specimens. However, the lamellae are not always parallel to the outlines of (110) faces, but may be inclined, curved or kinked (Fig. 7). Extinction angles of the lamellae vary along the curves. The sectoral boundaries are sectoral twin planes roughly parallel to [001].

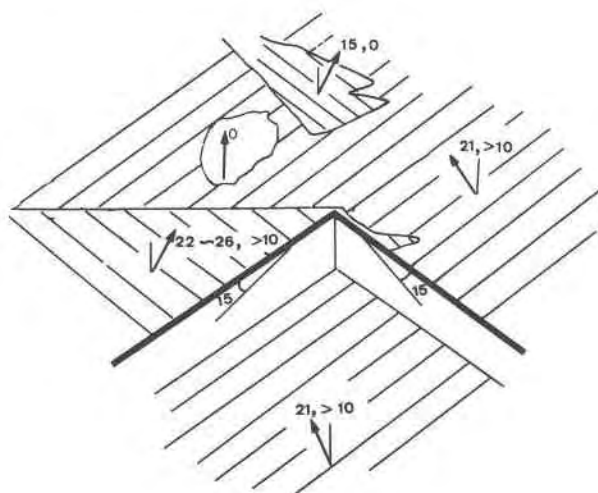


Fig. 5. Schematic diagram of growth sectors in the rectangle shown in Fig. 4. Growth lamellae, optical extinction directions (arrows), and extinction angles are shown. The sectors consist of alternate lamellae showing larger extinction angle (left number) and smaller angle (right). [001] is vertical.

Thickness of the lamellae varies parallel and normal to the crystal surfaces, and frequently the lamellae cross each other in thin sections parallel to the growth plane. Also, the lamellae are inclined to the growth surface (110) and cross growth bands in thin sections normal to the surface. A small difference of Al/Fe<sup>3+</sup> ratio was detected between the two kinds of lamellae by electron microprobe. Although an attempt was made to determine the optical orientation and 2V value, they were not clearly defined, because the optical extinction was not clear. However, the structure is assumed to be triclinic, because the crystal is optically biaxial and the indicatrix rotates variously with respect to the crystal axes. Internal texture similar to that of the Fujikaramari specimen commonly is observed in garnets from other localities in Japan.

#### *Andradite from Kamihogi, Yamaguchi Prefecture, Japan*

The specimens, which are greenish brown and translucent, consist of well-developed {110} faces and are about 10 mm in diameter. The average chemical composition is And<sub>79.8</sub>Gr<sub>20.2</sub>. Very fine non-isotropic chemical zoning, which causes a strong iridescence, was observed in thin sections normal to the growth surface (Akizuki, Nakai, and Suzuki, in prep.).

Figure 8 shows growth features on a (110) surface consisting of many blocks whose forms are the same as the outline of a rhombic (110) growth plane. The blocks, which have fine striations parallel to the [001] direction, are sometimes slightly misoriented, that is, this garnet has the so-called lineage structure as found in galena (Buerger, 1932).

One-to-one correspondence between the surface features and the internal texture is not observable in the specimen, because the area just below the growth surface

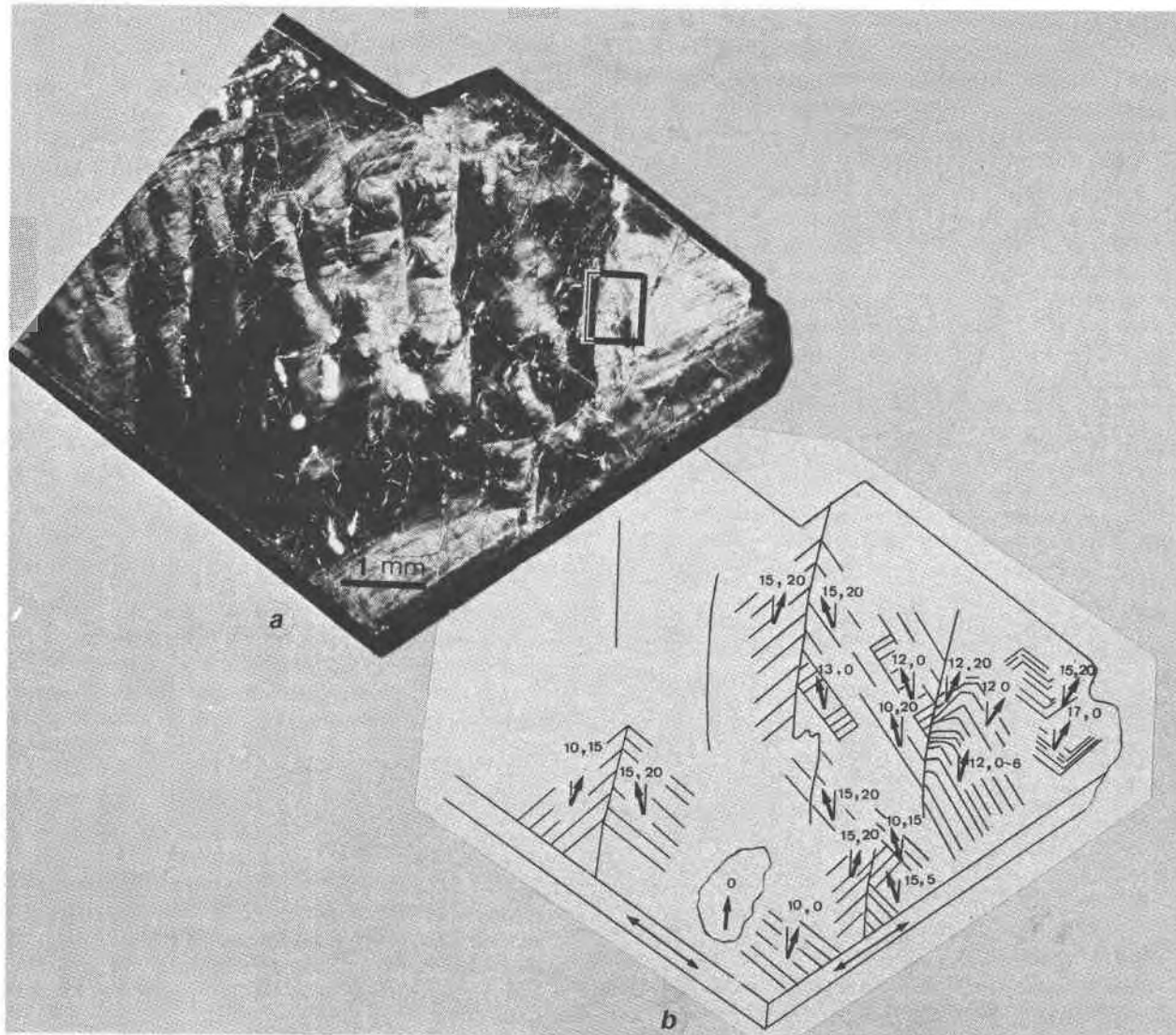


Fig. 6. Cross polarized photomicrograph (a) and schematic diagram (b) of a thin section parallel to the growth surface (110) of garnet from Fujikaramari. [001] is vertical and the polarizing plane is rotated about  $12^\circ$  counter-clockwise from vertical. See text and caption of Fig. 5 for details.

shows a fine granular texture whose development is presumably due to strain. However, many triangular sectors correlating to the outline of the (110) face are observed in the area deeper than about 0.5 mm below the surface (Figs. 9a, 9b and 10), and schematic figures of the sectors are shown in Figure 11. The optical orientations of sectors with solid and dashed lines (Fig. 11) are roughly symmetrical with respect to the [001] direction. The sectors are more or less optically homogeneous and have no lamellae corresponding to the striations on the surface (Fig. 8). Figures 9a and 9b show some triangular sectors indicated by letters B and E, corresponding to those of Figure 11. Since the triangular sectors are in contact or overlap with each other, their shapes are modified from the ideal shapes of Figure 11.

Figure 10 shows growth sectors which correspond to a hillock with four vicinal faces. This arrangement of growth sectors is extremely rare. The different optical contrasts shown by the four starts in Figure 10 are the result of chemical zoning. Figure 12 shows a cross-polarized photomicrograph of a thin section normal to the (110) growth surface. The texture crossing the horizontal growth lamellae is a profile of growth sectors like those shown in Figure 9. The textures are optically modified where they intersect. One example of the triclinic optical orientation is shown in Figure 13. Although the rotation of the optic axial plane is  $33^\circ$  from the  $[\bar{1}10]$  axis in the (110) plane in this example, the rotation angles vary from about  $25^\circ$  to  $35^\circ$  in the specimen, and frequently the angle and the  $2V$  value are not clearly defined. This type of

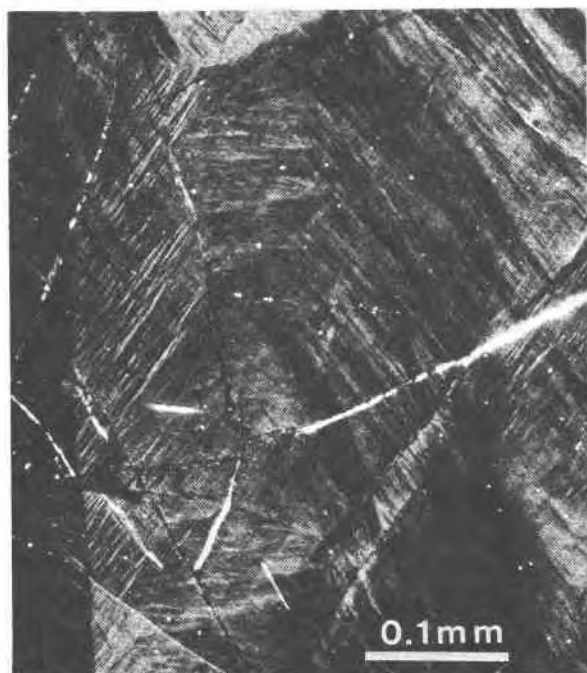


Fig. 7. High-magnification cross polarized photomicrograph of fine lamellae in the rectangle shown in Fig. 6a. [001] is vertical, and the polarizing plane is rotated 3° clockwise from vertical.

texture was not observed in garnet from other localities in Japan. Iridescent grandite garnet from Nevada, however, shows a texture similar to that of the present garnet (Ingerson and Barksdale, 1943, their Fig. 8).

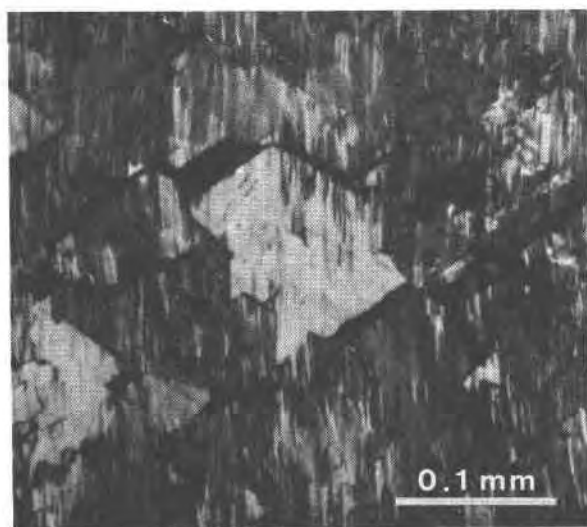


Fig. 8. Reflection interference contrast photomicrograph of the growth pattern on a (110) surface of garnet from Kamihogi. [001] is vertical.

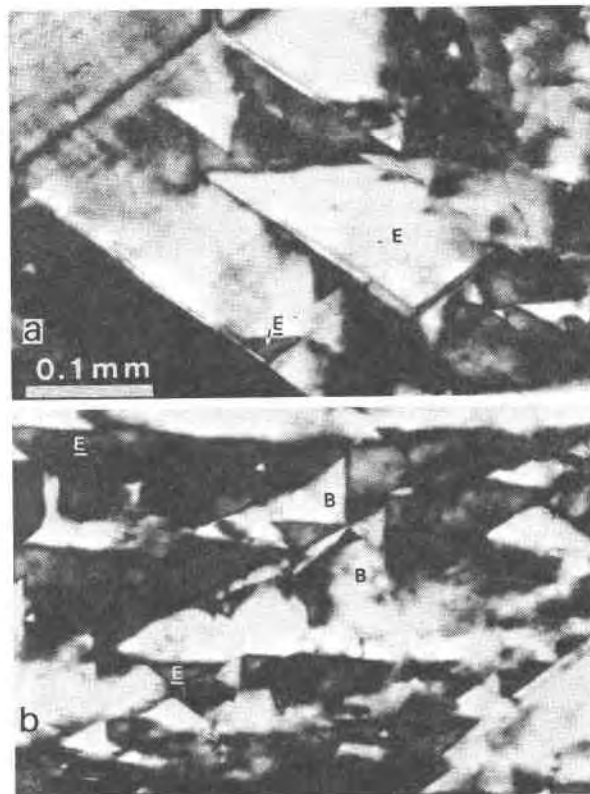


Fig. 9. Cross polarized photomicrograph showing sectors in a thin section parallel to the (110) growth surface of garnet from Kamihogi. The letters B and E correspond to those in Fig. 11. [001] is vertical and the polarizing plane is rotated 20° counter-clockwise from vertical.

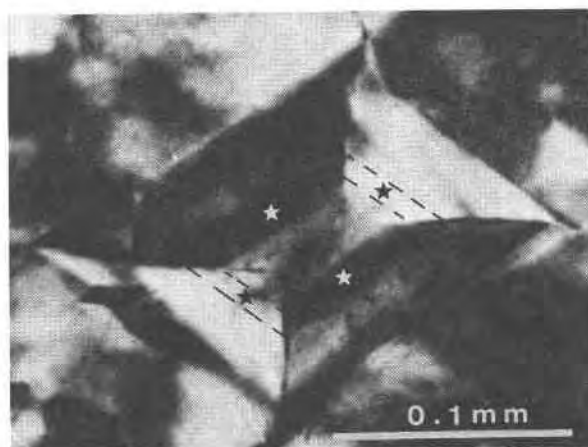


Fig. 10. Cross polarized photomicrograph of sectoral fourling twinning in thin section parallel to the (110) growth surface of garnet from Kamihogi. [001] is vertical and the polarizing plane is rotated 28° counter-clockwise from vertical.

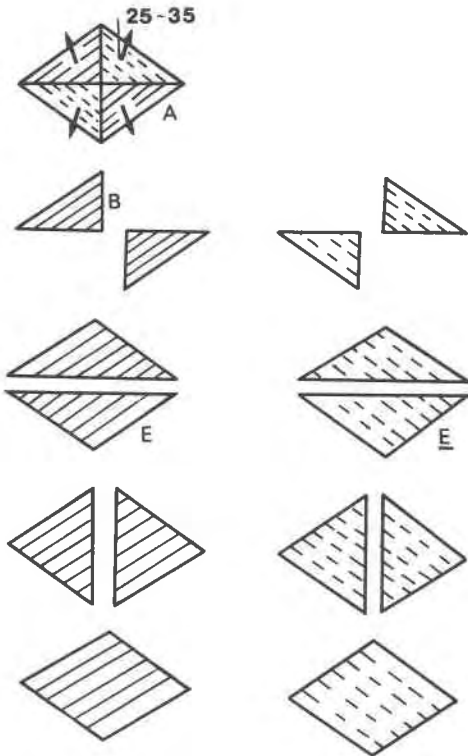


Fig. 11. Schematic figures of sectors observed in thin sections parallel to the (110) growth surfaces of garnet from Kamihogi. Extinction directions of the sectors with solid and dashed lines are symmetrical with respect to [001], shown by arrows in Fig. A. [001] is vertical.

### Discussion

Correlations between growth features on the crystal surface and the internal texture as found in Eden Hill and Fujikaramari garnets have been observed in adularia (Akizuki and Sunagawa, 1978), analcime (Akizuki,

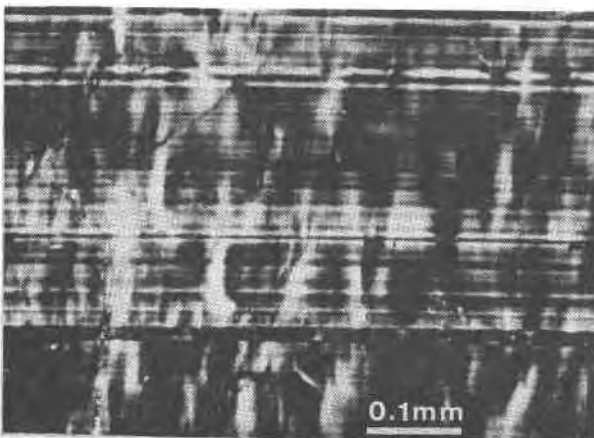


Fig. 12. Cross polarized photomicrograph of a thin section normal to the (110) growth surface of garnet from Kamihogi.

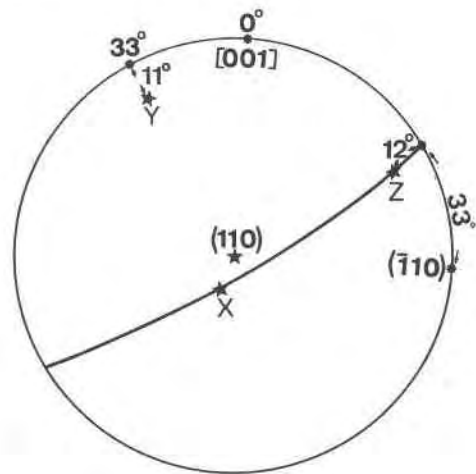


Fig. 13. Stereographic projection of optical properties measured in a (110) thin section of garnet from Kamihogi. Principal vibration directions (X,Y,Z) and optic axial plane are shown. X is not normal to the growth surface (110).

1981a), and chabazite (Akizuki, 1981b). Therefore, the origin of optical variations in the present specimens will be interpreted by the general mechanism of formation for order-disorder growth sectors suggested by Akizuki (1981a).

### Crystal growth mechanism

The pyramidal shape of the growth hillocks on garnet from Eden Hill suggests spiral growth. The garnets from both Fujikaramari and Kamihogi, however, consist of many sectors without a growth center. In Fujikaramari garnet, fine lamellae may correspond to growth steps that nucleate at the sector boundary or the crystal edge, suggesting that Fujikaramari garnet grew by the two-dimensional nucleation mechanism. Kamihogi garnet, with lineage structure, was presumably produced by two-dimensional crystal growth as well, because sectors with a growth center are extremely rare. The clear correlation between surface features and internal textures strongly suggests that the internal textures of garnet from Eden Hill and Fujikaramari were produced during growth, not by phase transition after growth. In minerals whose growth conditions changed abruptly at the latest stage of crystal growth, surface features are not always correlated to internal textures (Akizuki, 1981a). The internal textures in Kamihogi garnets (Figs. 9a, 9b and 10) are similar to the growth sectors of garnets from the other two localities (Figs. 1a, 1b and 4); therefore they were presumably produced during growth as well, though the one-to-one correlation between the surface features and internal texture is not observed in the crystal. Thin sections normal to the (110) growth surfaces of some garnets show various textures crossing zonal growth lamellae parallel to the (110) surface (as in Fig. 12), and therefore it has been suggested that the textures were produced after

growth by a phase transition (Takéuchi et al., 1982) or by strain. Such a texture, however, is frequently produced during growth. The textures crossing the growth lamellae shown in Figure 12 correspond to growth sectors which were observed in thin sections cut parallel to the growth surface (110) (Figs. 9a and 9b).

### $Fe^{3+}/Al$ ordering

Takéuchi et al. (1982) represented the structure of garnet using an array of octahedra on a pseudo-trigonal axis, which is similar to the rod-packing model of the garnet structure suggested by O'Keeffe and Andersson (1977). The cation ordering can be explained by differences in the immediate surroundings of the  $Fe^{3+}/Al$  sites of the octahedral array exposed on the growth steps. Figure 14 indicates the relation between the pseudo-trigonal axes shown by arrows and the crystal form of garnet. Four of the eight octahedral arrays in the unit cell lie on the four axes. Assuming growth of the crystal is in the direction shown by the arrow on the (110) plane, the immediate surroundings of octahedra in these four arrays differ from each other with respect to the growth direction on the surface. Thus, the four octahedral sites are not equivalent during growth. If this non-equivalence results in different relative preferences for iron and aluminum on the sites, then ordering will occur and the symmetry of the crystal will be reduced.

Although the formal charges of the two cations are the same, partitioning might occur on the growth surface due to the different ionic radii (0.57 Å for  $Al^{3+}$ , 0.67 Å for  $Fe^{3+}$ ) and electronegativities. Akizuki *et al.* (1979) predicted from optical observations that ordering of F and OH could occur on growth surfaces of topaz, because of different ionic radii. This hypothesis was confirmed by the neutron diffraction studies of Parise (1980).

If a crystal grows in the directions of the arrows in Figure 15, which are symmetrical with respect to  $(\bar{1}10)$

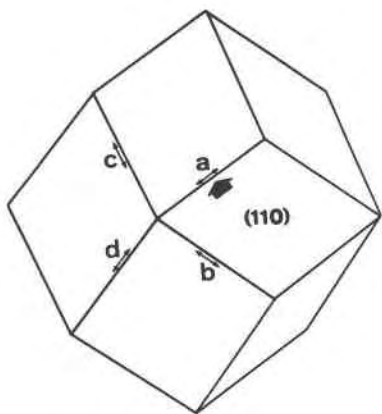


Fig. 14. Relation between the crystal form of garnet and directions of the octahedral arrays shown by arrows labelled a, b, c, and d. Growth steps discussed in text move in the direction shown by the thick arrow on the (110) face.

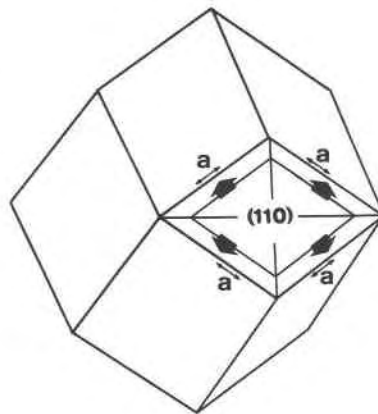


Fig. 15. Relation between formation of fourling sectoral twins and growth directions (heavy arrows) of vicinal faces on the (110) surface of garnet. The letters "a" correspond to the "a" of Fig. 14.

and (001), the same situation will occur on each vicinal face, and fourling twins will be formed during growth (Figs. 10 and 15). If the growth steps kink or curve during growth, the  $Fe^{3+}/Al$  partitioning at each site may differ from growth step to growth step, and complicated optical textures will be produced in the crystal during growth (Figs. 6a, 6b, and 7). Ordered structures with various  $Fe^{3+}/Al$  ratios can be produced in the garnets except the end-members. Birefringence is observed even near the end-members, because optics are very sensitive to ordering in the structure.

The three specimens are optically biaxial, and of them, the two specimens from Kamihogi and Fujikaramari may be triclinic, judging from the relations between optical and crystallographic orientations. However, the symmetry of Eden Hill garnet may be monoclinic. Growth steps on the growth hillocks of garnet from Eden Hill are inclined to the [001] axis on the (110) surface, and therefore the optic vibration direction does not coincide with the [001] axis, because one-dimensionally ordered arrangements repeat parallel to the growth step on the surface, resulting in a two-dimensionally ordered structure. If the two-dimensionally ordered structures stack regularly in the direction normal to the growth plane (110) during slow growth, a diad axis may occur in the direction parallel to the [110] axis, which coincides with one of the principal optical vibration axes. If so, the garnet from Eden Hill will be monoclinic (Fig. 2).

An orthorhombic structure cannot be produced on the vicinal surface with growth steps inclined to the [001] axis on the (110) face, but may be formed on the vicinal surface with growth steps parallel or normal to the [001] axis. Whether such a structure exists on the (110) surfaces of some garnets is not known. According to X-ray diffraction analysis of garnets from Kamaishi, Japan, by Takéuchi et al. (1982), the cell parameters  $a$  and  $c$  for the cubic cell are equal but the angle  $\beta$  is not  $90^\circ$ ; the cubic  $\alpha$



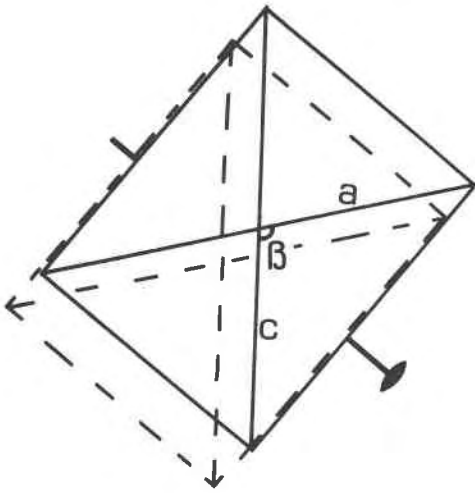


Fig. 16. Monoclinic unit cell of garnet. Pseudo-cubic cell dimensions  $a$  and  $c$  are equal, as are  $\alpha$  and  $\gamma$ , which are not equal to  $90^\circ$ . Solid and dashed lines show two-dimensional unit cells parallel to each other. Diad axis is normal to the  $(110)$  growth surface. The unit cell data are those of garnet from Kamaishi, Japan, which was analyzed by Takéuchi *et al.* (1982).

and  $\gamma$  are equal but are not  $90^\circ$ . The garnet is monoclinic, with the diad axis parallel to  $[110]$  of the cubic cell as shown in Figure 16, though Takéuchi *et al.* (1982) reduced their data in the orthorhombic system, because the lattice angles  $\alpha$  and  $\gamma$  are very close to  $90^\circ$ . Monoclinic symmetry is consistent with the optical observations for Eden Hill garnet.

### Twinning

According to crystallographic definition, optical properties such as  $2V$  and optical orientation must be symmetrical with respect to a twin plane. Because the sector twin components of the garnets discussed above, especially those from Kamihogi and Fujikaramari, are not optically homogeneous, the  $\text{Fe}^{3+}/\text{Al}$  ordered structure is not always *rigorously* symmetrical with respect to the "twin" planes, although the optical orientations of the sectors are *roughly* symmetrical with respect to the  $[001]$  axis. Therefore, these sector twins are not twins in a strict crystallographic sense, and the term sector twin here is used to denote a relation between two sectors whose growth directions and optical orientations are roughly symmetrical with each other.

The traces of sector twin planes of garnets from Kamihogi and Fujikaramari are parallel and normal to  $[001]$  on the  $(110)$  face, while in garnet from Eden Hill, one sector twin plane is parallel to the  $[001]$ , whereas the other varies from  $90^\circ$  to about  $50^\circ$  from the  $[001]$  axis. These differences are a result of differences in the growth features.

Takéuchi *et al.* (1982) distinguished two kinds of octa-

hedral sites, positive and negative octahedral, by differences in the ratio of  $\text{Fe}^{3+}/\text{Al}$ . For instance, in Munam garnet ( $\text{Gr}_{68}\text{And}_{32}$ ) analyzed by Takéuchi *et al.* (1982), the Fe content varies from 41 atom percent to 49 atom percent in the four sets of positive octahedra and from 15 atom percent to 29 atom percent in negative octahedra. Figure 17 shows schematically the twinned structure on the symmetrical vicinal faces of the  $(110)$  growth plane. This figure was modified from the twinned structure shown by Takéuchi *et al.* (1982) to interpret the growth twinning observed in the present garnets. In grandite garnet, the twelve  $\{110\}$  growth planes produce twelve growth sectors, and each surface has various vicinal faces which make many fine sectors. These complexities make it essentially impossible to ascertain the type of twin using a random section of garnet.

### Fine lamellae

The optical orientations of the fine lamellae (Figs. 5 and 6b) are roughly symmetrical with respect to the sector twin boundaries. The lamellae never cross a sector twin boundary, though they cross growth bands parallel to the  $(110)$  growth surface. The internal lamellae in garnets from Fujikaramari and Eden Hill correlate with the growth steps on the surface (Figs. 1 and 3). Garnet with fine striations on a small  $(211)$  face from Fujikaramari shows fine lamellae normal to the growth surface in the

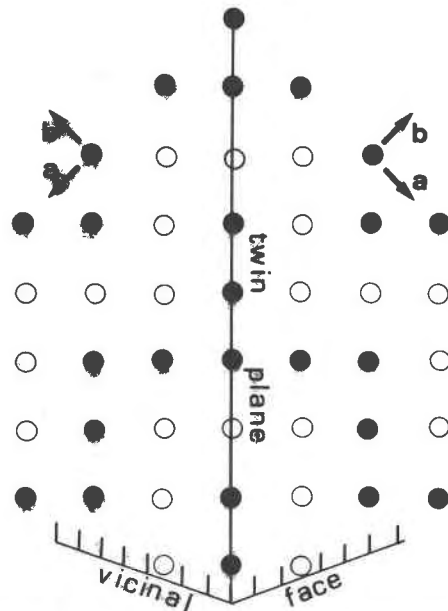


Fig. 17. Schematic diagram of twinned structure produced on vicinal faces of the  $(110)$  growth surface of garnet. The figure was modified from the twinned structure shown by Takéuchi *et al.* (1982). Solid and open circles show octahedral cation sites,  $M_1$  and  $M_2$ , at which  $\text{Fe}^{3+}/\text{Al}$  occupancy is different. The twin plane is normal to the  $(110)$  growth surface. The crystal axes  $a$  and  $b$  of the pseudo-cubic cell are shown.

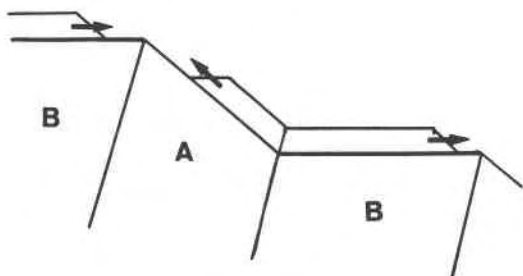


Fig. 18. Directions of motion of fine steps on thick growth steps. Degree of ordering and/or ratio of  $\text{Fe}^{3+}/\text{Al}$  are different in the two kinds of sectors, A and B.

{211} sector between crossed polars. From these observations it is thought that the lamellae were produced during growth.

Figure 18 is a schematic diagram showing a possible mechanism for formation of lamellae. If a growth step consists of monatomic layer, a step will move in only one direction along the surface, whereas if the growth step is thick, thin growth layers may move in two different directions (Fig. 18). If so, differences of chemical composition and/or degree of  $\text{Fe}^{3+}/\text{Al}$  ordering might occur on the two kinds of surface (A and B in Fig. 18), and therefore lamellae may be observed in the crystal. Striations on the (211) surface consist of alternating (110) and (101) faces, and polysynthetic twinning develops in the {211} sector.

The distance between steps varies during growth, and the thickness of steps can change by bunching or dispersing of steps. This explains the variation of thickness of the internal lamellae. If a surface feature changes by interruption of growth, it might result in two kinds of lamellae crossing each other in a thin section parallel to (110).

### Stability

It is likely that only the cubic structure is stable at the growth temperature in grandite garnet, because non-cubic grandite garnets are not homogeneous but have a heterogeneous texture characteristic of metastable minerals. No unequivocal evidence of phase transitions from the high form to the low form have been observed in these garnets. The crystal symmetry of the meta-stable garnet is not merely the result of the chemical composition, but results from  $\text{Fe}^{3+}/\text{Al}$  ordering in the octahedral sites. The degree of ordering in minerals can be affected by growth temperature and growth rate (Akizuki, 1981a,b,c). If the growth temperature of garnet is very high, a cubic disordered structure may be produced during growth. If the growth rate is very high or extremely low, a cubic disordered garnet may occur as well, because the disordered structures are favored by high growth rates, or are obtained by equilibrium growth at low growth rates. At intermediate growth rates and temperatures typical of hydrothermal or skarn deposits, the metastable ordered

structure is produced by the two-dimensional atomic arrangement exposed on the side faces of growth steps and is frozen during and after growth (Akizuki, 1981c).

Hariya and Kimura (1978) synthesized non-cubic grandite garnet at hydrothermal conditions and determined the "stability" field of pressure and temperature for the formation on non-cubic garnet. They compared the "stability" field with the phase transition temperature of natural non-cubic garnet as determined by heating. The non-cubic garnet formed at temperatures lower than about  $700^\circ\text{C}$  at low pressure, while the transition temperature of the natural garnet was slightly higher than  $800^\circ\text{C}$ . In the present interpretation, the metastable  $\text{Fe}^{3+}/\text{Al}$  ordered structure of synthetic garnet is produced on the free surface during growth, whereas the order-disorder transition occurs in the three-dimensional structure.

Some garnets show complicated internal textures which may be attributed to residual strain after crystal growth, and these textures may overlap with growth sectors in some thin sections. It is difficult to distinguish between the two textures in thin sections normal to the growth plane in some cases. The optical properties produced during growth may be modified by strain after growth in some specimens. It appears that Eden Hill garnet maintains the original structure and texture and that the modification of the garnets from Fujikaramari and Kamihogi is not large.

### General conclusions

Optical textures of natural grossular-andradite garnet can be interpreted in terms of  $\text{Fe}^{3+}/\text{Al}$  ordering which is produced on the surface during growth. Growth twins, whose mirror planes are in orientations parallel, normal, or inclined to [001] in (110) thin sections, correspond to sector boundaries at which growth steps are roughly symmetrical. Various types of twins and optical properties of garnet which were summarized by Winchell and Winchell (1951) may be interpreted by the same principles described in the present paper.

Even if a growth face is normal to the mirror planes of the ideal crystal structure and a growth hillock possesses the mirror planes, vicinal faces on the face are not always normal to the mirror plane. If the crystal has a growth hillock on the surface as shown in Figure 19a, the vicinal faces will be normal to the mirror planes (m), and therefore the structure produced on the hillock will have one mirror plane at least and will be disordered with respect to that plane. However, since the vicinal faces shown in Figure 19b are not normal to the mirror plane, the crystal structure does not possess the mirror plane and may be ordered, thereby reducing the symmetry. The crystal symmetry is not controlled by the macroscopic form of the crystal, but by the microscopic form of growth hillocks, that is, the order-disorder structure is determined by the directions of growth steps. In the case of Figure 19b, even if the crystal form has the mirror

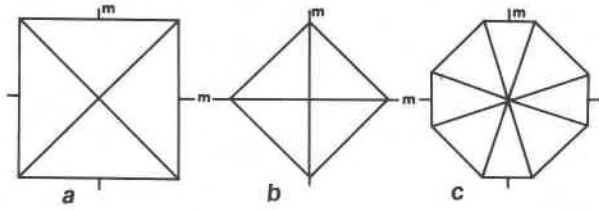


Fig. 19. Growth hillocks (summits in center) with mirror planes (m). Growth steps on hillock (a) are normal to the mirror planes, while the growth steps on hillock (b) are inclined to the mirrors. Hillock (c) consists of a combination of two kinds of vicinal faces (a and b). See text for details.

planes, the disordered structure with the mirror planes is not necessarily produced during growth. An example is chabazite crystals with rhombohedral form consisting of triclinic ordered structure (Akizuki, 1981b). The vicinal faces of growth hillocks on dodecahedral garnets are not normal to a mirror plane. Therefore, the ordered structure produced on the hillock is not orthorhombic, but monoclinic or triclinic. If a growth hillock is composed of a combination of the two cases (a and b of Fig. 19), as shown in Figure 19c, the sectoral texture may become more complicated. Surface features must be observed in order to understand minerals with anomalous optical properties. X-ray analyses should be performed on individual sectors after determining the optical properties as well as the chemical compositions. Optically homogeneous crystal flakes for X-ray analysis may be obtained from thin sections parallel to the growth surface.

### Acknowledgments

I thank Dr. D. R. Veblen, Johns Hopkins University, and Dr. E. Dowty, American Museum of Natural History, for valuable suggestions and English correction of the manuscript. Also, I thank Dr. R. Aines, California Institute of Technology, for critical reading of the manuscript. I am grateful to Drs. K. Yamaoka and H. Konno, Tohoku University for providing the samples.

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*Manuscript received, December 27, 1982;  
 accepted for publication, September 14, 1983.*