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Twinnings in phlogopite

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Abstract

The surface microtopographs of phlogopite crystals occurring in the druses of lavas, ranging from trachybasalt to andesite, at ten localities in Japan were studied by the phase contrast and interference contrast microscopes. Both mono-molecular growth spirals and plateaus are essentially in five-sided forms bounded by $(\bar{1}0)$, (01), (11), and (13), or $(\bar{1}0)$, (01), and (11). Apparent single crystals consist of many five-sided thin domains in twin orientations with rotation angles of 60° × *n*. It is concluded that the twinnings were formed by the agglutination in twin orientations of platy crystals of five-sided forms which had already reached certain sizes and were moving around in the space like magic carpets. Twin relations were also found between the two neighboring growth spirals occurring on one plateau surface, and these are considered to have been formed by a stacking fault introduced during the growth of a single plateau.

Introduction

Twinning in mica minerals has been studied by several workers. Takano and Takano (1958) discussed the possibility of mistaking X-ray diffraction patterns of twinned crystals of mica minerals as a new polytype. Sadanaga and Takéuchi (1961) theoretically derived possible types of polysynthetic twins of mica minerals, and observed spiral twins with rotation angles of $\omega = 60°$, 180° and alternating twins of $\omega = 180°$ on phlogopite crystals from Mutsure-jima (=zima), Japan. Bloss *et al.* (1963) studied synthetic fluorophlogopite and showed that 29 percent of the specimens are composed of two or more polymorphs twinned about the [310] axis or much more rarely about [110]. They attributed this to irregularities in the location of the Mg ions (60° or 180°) and of the fluoride or hydroxyl ions (120°). These are mainly X-ray studies. Sunagawa (1964) observed five-sided (hexagonal truncated one side) growth spirals of probably one unit cell height on the (001) face of phlogopite from Mutsure-jima and showed that apparent single crystals of the phlogopite actually consist of many domains in twin orientations by rotation around the c^* axis with angles $\omega = 60°$, 120°, and 180°, judged from the orientations of five-sided growth patterns.

We have made further surface microtopographic observations of phlogopite crystals from ten localities in Japan. All crystals exhibit on the (001) face many five-sided plateau-like patterns bounded by either $(\bar{1}0)$, (01), and (11) or by $(\bar{1}0)$, (01), and (13), whose sizes range from 20μ to 850μ, and heights of the order of one micron. On the surfaces of these plateaus, growth spirals of one unit cell height (10Å, measured by multiple-beam interferometry) are observed. They are also bounded by $(\bar{1}0)$, (01), and (11), or $(\bar{1}0)$, (01), (13), and (11), depending on the localities. From the orientations of the five-sided plateaus and spirals, twin relations are easily detected. In addition to the twin type previously reported by Sunagawa (1964), a new twin type was observed in this study. Judging from the modes of their appearance, it was considered that both types are different in their genesis. The present paper describes the observations and discusses how the two types of twins were formed.

Samples and experimental technique

Phlogopite crystals investigated in this study were collected from the druses of lavas, ranging from trachybasalt to andesite, at the following ten localities:

Mt. Akagi, Gumma Prefecture [in two-pyroxene andesite, $\gamma \approx \beta$ 1.581, $2V(-) = 0°$].

Takura-yama, Hyogo Prefecture [in augite-bearing olivine–trachybasalt, $\beta \approx \gamma$ 1.573–1.574, $2V(-) = 1°–2°$].

Kurayoshi, Tottori Prefecture [in olivine–trachyandesite lava, $\beta \approx \gamma$ 1.568 $2V(-) = 0°$].

Mutsure-jima, Yamaguchi Prefecture [in olivine–trachybasalt lava, α 1.559, γ 1.607, $2V(-)$ = 22°–32° (by Kôzu and Yoshiki, 1929); a 5.335Å, b 9.245Å, c 10.136Å, β 100.1° (Sadanaga and Takéuchi, 1961)].

Mutsumi-mura, Yamaguchi Prefecture [in two-pyroxene olivine–basalt, $\beta \approx \gamma$ 1.616–1.617, $2V(-)$ = 14–25°].

Sekiyama, Shimonoseki-shi [in amphibole-bearing olivine–trachybasalt, $\beta \approx \gamma$ 1.569–1.570, $2V(-)$ = 0°].

Nokono-shima, Fukuoka-shi [in augite-bearing amphibole–olivine–trachybasalt, $\beta \approx \gamma$ 1.563–1.565, $2V(-)$ = 0°].

Iwagami-yama, Kumamoto-shi [in two-pyroxene amphibole–andesite, $\beta \approx \gamma$ 1.570–1.572, $2V(-)$ = 1°–2°].

Miyakono-mura, Oita Prefecture [in two-pyroxene andesite, $\beta \approx \gamma$ 1.565–1.566, $2V(-)$ = 4°–6°].

Kakara-jima, Saga Prefecture [in hortonolite–anorthoclase trachyte, $\beta \approx \gamma$ 1.616–1.617, $2V(-)$ = 5°–26° (Ishibashi, 1971)].

The phlogopites from Kurayoshi, Mutsure-jima, Mutsumi-mura, Sekiyama, Nokono-shima, and Kakara-jima, occur in lavas belonging to the Sanin-Kita-Kyushu alkali basalt petrographic province, others from different provinces. It is associated with pargasite, ilmenite, augite, hypersthene, and rarely magnetite, hematite, tridymite, or plagioclase. Most of the mineralogical data on these phlogopites, together with the data on coexisting druse minerals, were reported by Oota (1958, 1959). The phlogopite from Mutsure-jima has been studied by several workers (Kôzu and Yoshiki, 1929; Kozu and Tsurumi, 1931; Tomisaka, 1958, 1962; Sadanaga and Takéuchi, 1961). This phlogopite is a biotitic phlogopite with $1M$ structure, and is twinned by rotation, in the forms of spiral or alternation twins (Sadanaga and Takéuchi, 1961). The phlogopites from the other localities are also $1M$ polytype, judging from both X-ray data and morphology of growth spirals.

Crystals were silvered in a vacuum evaporation apparatus to secure high reflectivity suitable for observation under the reflecting-phase or interference-contrast microscopes. When necessary, crystals were etched with 10 percent HF solution, after which silvering was applied.

Observations and discussion

Figure 1a is a transmission polarized photomicrograph and 1b a reflection plus transmission photomicrograph of a phlogopite crystal. This crystal has a hexagonal outline and gives the appearance of a single crystal, if its surface microtopograph or internal heterogeneity is neglected. As clearly seen in these photomicrographs, the crystal consists of many superimposed five-sided domains.

The five-sided domains are rotated with respect to each other by angles of 60° × n. The straight lines bounding five-sided domains correspond, without a doubt, to the edges of the five-sided plateaus occurring on the surface, and this can be easily identified in Figure 1b. There are also some domains which are inside the crystal and do not appear on the surface. In Figure 1a, five-sided plateaus on both basal faces, upper and lower, are seen superimposed, together with those inside the crystal. Figure 1a also shows clearly that the extinction angles vary from place to place in the crystal, depending on the rates of super-

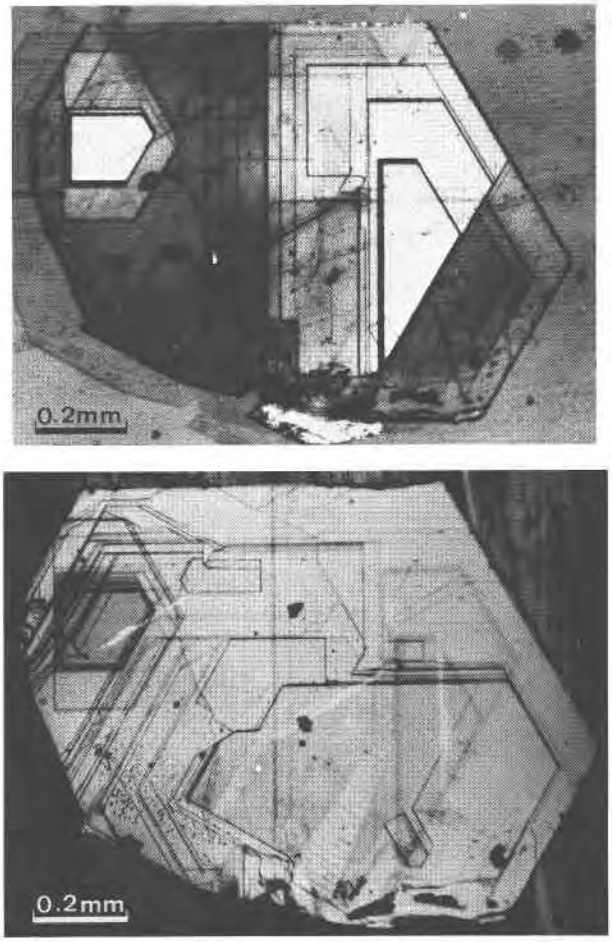

FIG. 1. (a) Polarized transmission photomicrograph (top) and (b) reflection plus transmission photomicrograph of an apparent single crystal of phlogopite (bottom)

imposition of differently oriented domains. This is perhaps the cause for the values of $2V$ showing some variation, ranging from 0° to 32°, in the previous reports.

X-ray diffraction spots of such a crystal give trigonal symmetry. Detailed investigations of intensity distribution reveal that the crystal has $1M$ structure and is twinned by the rotation of $60° \times n$ perpendicular to c^*, with composition plane (001). Only when a thin foil is cleaved off and examined by X-ray, are diffraction spots of $1M$ symmetry obtainable.

These results clearly show that the phlogopite crystals, although appearing as single crystals, are in fact twinned crystals consisting of many five-sided domains superimposed and rotated at angles of $60° \times n$ with respect to each other.

Figure 2 is a reflection photomicrograph of a typical plateau. Its edges are straight and sharp, and have no connection with the step patterns developing on the surface of the base on which this plateau occurs. This is a general relation observed between the basal surface and the edges of plateaus. Only exceptionally may one find steps consisting of the straight edge of a plateau continuing to the steps appearing on the basal surface.

When thinner growth layers bunch together to form a thicker layer, the bunched thick layer usually takes on a more irregular form than the thinner layers. When they reach the edges of the crystal, they become straight and join with the side faces. This is observed universally. Therefore the straight edges of the plateau must represent the edges of a platy crystal that has already reached the present size, and are not the thick layers formed by bunching of thinner layers developing on the substrate surface. Five-sided forms and straight edges of the plateaus must have been formed before they agglutinated on the surface of the already well-developed platy crystal.

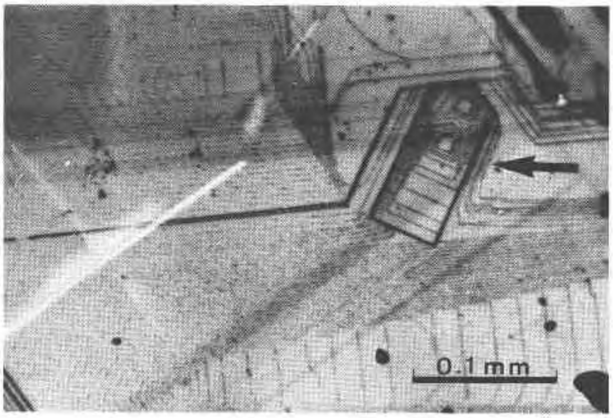

FIG. 2. Showing the relation of a plateau indicated by an arrow and mono-molecular growth spirals. Twin relation is also seen.

Twinnings of this type are not formed during the nucleation stage (Buerger, 1945), because of the wide range in sizes of twin components and a large number of components constituting the twinned crystal. The only possible explanation for the observations is that these twinnings are formed by the agglutination in twin orientations of thin platy crystals which have already attained certain sizes and are bounded by well-developed straight crystallographic side faces. Such thin platy crystals must have been moving around in the space like a flying magic carpet while they were growing, and when they agglutinated with other platy crystals or on the surface of a larger crystal they settled in the twin positions. Hartman (1956) discussed the possibility of such agglutination as a cause of growth twins.

As to whether thin platy crystals of certain sizes can move around in the space like flying magic carpets, we may point out two observations. Sunagawa *et al.* (1968) and Sunagawa and Endo (1971) proposed, on the basis of observations on auto-epitaxial growth of fluor-phlogopite from the vapor phase onto the cleavage surface of melt-grown fluor-phlogopite, that thin platy crystallites which are formed by the coalescence of molecules in the vapor phase are transported certain distances like flying magic carpets, and settled on the cleavage surface at the cooler end. When they settled, they took, with equal probability, orientations rotated by $60° \times n$. Kitazawa *et al.* (1971) also observed through in situ observations of the growth process of CdI_2 crystals in aqueous solution that hexagonal platy crystals moving around in the solution often agglutinate, and that just before they agglutinate they adjust their orientations so as to meet with crystallographic orientations.

We have observed many crystals and found that practically all five-sided plateaus are rotated at angles of $60° \times n$ with respect to each other, *i.e.* in the relation of twinning. However, only exceptionally we find cases that five-sided plateaus are rotated approximately 28°, 13°, and 7°.

If we assume that the (001) face of phlogopite consists of a regular hexagonal network, and that the bonds directed outwards are perpendicular to the surface, the settlement of a platy crystal onto the basal plane of the other crystal by the rotation of angles of $60° \times n$ requires exactly the same energy as required for the settlement in the correct orientation. Probability of agglutination in twinned and single orientation is, therefore, equal. The probable orientations which come next to these may be calculated on the basis of Kronberg-Wilson theory of Coincidence-Site Lattice (Kronberg and Wilson, 1949). For a hexagonal network with bonds perpendicular to the plane (Kobayashi and Furukawa, 1975), $\Sigma = 7$ comes next to the twin orientations of rotation angles $60° \times n$ ($\Sigma = 1$), followed by $\Sigma = 13, 19, 31$, *etc.* Σ is designated by the ratio of size of the Coincidence-Site Lattice (multiple cell) to the original unit cell. These correspond to rotation angles of 21.8°, 27.8°, 13.2°, 17.9°, *etc.*, respectively. The observed angle of misoriented five-sided plateaus agree well with $\Sigma = 13$, 19, and 61. This again supports the proposed genesis of this twinning.

Mono-molecular growth spirals occurring on one plateau surface have, in most cases, the same orientation as that of the plateau (*e.g.* Figure 1b and 2). However, there are cases, though only occasionally and only on the crystals of certain localities, in which two neighboring growth spirals, occurring on one plateau surface, are rotated with respect to each other at angles of $60° \times n$. Figure 3 shows rotations of 60°. Although the angles of rotation are the same, these are quite different from the twinning described above, in that the previous cases are twin relations among platy crystals which have already reached certain sizes and have agglutinated on the surface of a pre-existing crystal, whereas in this case twinnings are between the two neighboring mono-molecular growth spirals occurring on the same plateau surface. Their origins should be quite different.

Giving attention to the modes of growth of spiral layers in this photomicrograph, especially to the portions where two spiral layers meet, we note special features which are not commonly observed in the case of two spiral layers with the same orientation meeting.

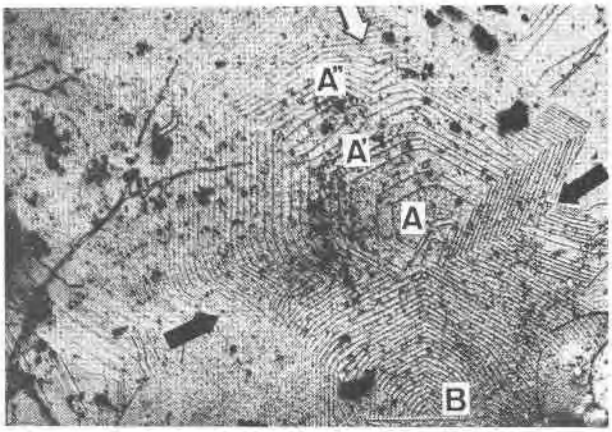

FIG. 3. Twin relations of the second type, *i.e.* between the two neighboring mono-molecular growth spirals. 60° rotation. See text.

Two-dimensional re-entrant corner effect is noted universally at the intersections of two spirals having the same orientation. An example may be seen at the intersections indicated by the white arrow in Figure 3. The intersection indicated by the white arrow in Figure 3 which exhibits typical two-dimensional re-entrant corner effect is the intersection between the dominant spiral *A* and dominated spirals indicated by *A'*, *A''*, *etc.*, all having the same orientation. In contrast, at the intersections indicated by black arrows in Figure 3 between the two spirals, *A* and *B*, which are rotated 60° with respect to each other, the re-entrant corner effect is either not seen at all or is very weak compared to the above case. This is remarkable, and has never been observed at any intersections between the two spirals of the same orientation.

Another point in this photomicrograph is the fact that the spacings between the successive arms of $(\bar{1}0)$ steps are much wider and regular at the intersection than those of the $(\bar{1}0)$ steps appearing at places other than the intersection, or of isolated spirals. This is clearly seen if the spacings of $(\bar{1}0)$ steps are compared between those at the intersection (indicated by the right black arrow) and those of the spiral *B*.

Although the modes of interaction between the two nearby spirals in twinned orientation are different case by case, *i.e.* according to the different rotation angles, it is clearly and commonly observed that whenever two spirals occur rotated with respect to each other at the angles of rotation producing twin relations, peculiar morphological features which are quite different from the case where two spirals have the same orientation are invariably produced along the intersections.

As to the genesis of twinning of this type, the previous agglutination mechanism is not applicable, since the two components are mono-molecular in height and occur on the same surface of a plateau.

Since two spirals on the same plateau surface are in twin orientation, the relation should have been created on the surface which was originally single and in the course of growth (thickening) of the plateau which was growing by spiral mechanism. Such twin relations may be created by the introduction of a stacking fault on the growing surface due to some reasons, such as by the settlement of a mono-molecular slice in twin orientation on a growing surface or by slip (Sunagawa, 1960). If such a stacking fault is introduced, a structural gap is introduced along the intersection between the domain which follows the original stacking and the domain newly appeared due to the stacking fault, even when both surfaces reach the same level. The modes of the gap are different depending on the rotation angles. For 60° and 180° rotations, both tetrahedral and octahedral layers have difficulties in matching along the intersections. For 120° rotation, the octahedral layer may continue with slight modification, but the tetrahedral layer leaves a misfit along the intersection. Morphological characteristics observed at the intersections are considered to have resulted from such structural gaps.

Conclusion

Twinnings of two different types are found through the surface micro-topographic observations of the (001) faces of phlogopite crystals from ten localities in Japan. Commonly observed twinnings, which consist of five-sided thin domains in twin orientations with rotation angles of 60° × *n*, are formed by the agglutination in twin orientations of platy crystals of five-sided form which attained their present size and were moving around in space like magic carpets. Rarely-observed twin relations which hold between the neighboring growth spirals are formed by a stacking fault introduced during the spiral growth on the surface.

Acknowledgments

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