

## MECHANISM OF GROWTH OF HEMATITE

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### ABSTRACT

Surface structures on the basal planes and other faces of hematite crystals from many localities have been observed with both metallurgical and phase contrast microscopes. It is concluded that the growth takes place mainly by two-dimensional spreading of growth layers parallel to (0001), and that the other faces are formed mainly by the edges of these layers, although growth layers parallel to these other faces can develop when they become large.

It is also concluded that at an early stage, two-dimensional nucleation is responsible for the major growth, but later growth layers may originate from deformed portions of the surface, or from a group of dislocations, both of which are due to stresses applied to the crystal. Most of the thick growth layers are formed by the coalescence of thinner layers, but some originate independently. Only at the later stage, or under very low supersaturation, can typical spiral patterns be formed. From the characteristics of the growth patterns on crystals from different localities, a sequence is arranged, showing decrease in supersaturation conditions.

### INTRODUCTION

If the surface of a crystal is observed under a reflection microscope, a complex pattern which looks like contour lines of a topographic map is observed. These contour lines are sometimes polygonal according to the symmetry of a crystal, but may be circular or irregular. They are edges of layers, either of growth or etching, parallel to the face under observation. Therefore, by observing the surface structures of crystal faces, it is possible to determine the mechanism of crystal growth or etching. Although the surface structures which we are observing represent only the final stage of growth or etching, observations on many crystals of a wide range of sizes, as well as from many localities, give good information.

Both the layer growth and spiral growth theories have been established by studying the surface structures of crystals. There is no doubt that growth of crystals mainly takes place by two-dimensional spreading and piling up of growth layers and not by three-dimensional precipitation of molecules or atoms around a nucleus. It is also quite evident that screw dislocations play an important role in crystal growth. However, there are still many problems to be solved. Do spirals play an important role in growth throughout the process of crystallization, or are they important only at the latest stage of growth? Are there any other imperfections on the crystal surface by which growth can be greatly accelerated?

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The writer reported his observations on one hematite crystal on which the final history of growth can be observed (Sunagawa, 1960). Although this gave clear evidence that three-dimensional nucleation occurs before the spiral growth, it did not give a final answer to the above questions.

The writer has made observations on hematite crystals from the following localities: Ayumikotan, Hokkaido; Sasazawa, Nagano Pref.; Gihofuji, Okayama Pref.; and Saganoshima, Nagasaki Pref., in Japan; Vesuvius and Stromboli in Italy; the Azores Islands, Portugal; Ascension Island, and British West Indies. All hematite observed comes from volcanic lava, and is certainly crystallized by the reaction of iron chloride gas and water. This paper gives a brief description of the different structures observed on the basal and other faces and the mechanism of growth is discussed.

#### OBSERVATIONS UNDER LOW MAGNIFICATION

Under low magnification with an ordinary metallurgical microscope, the coarse surface structures on the basal plane look like contour lines on a topographic map. These lines are the edges of growth layers. One type is regularly triangular, and was observed only on crystals from Gihofuji and Ascension Island I. Circular or irregular patterns are much more common. Typical examples are shown in Figs. 1 and 2, respectively.

Commonly there are one or more growth centers. These are usually flat elevations, but in some cases there are smaller, fairly steep cones formed by rapid growth. Sometimes the ordinary metallographic microscope shows almost the entire surface as a very flat area, and thick growth layers are found only at the margin. The typical thin growth layers visible with a phase contrast microscope will be discussed later. On several crystals it was observed that the surface is broken into many small, slightly inclined areas (Fig. 3). In other cases, thick growth spirals start from small portions of the surface which are inclined to the main surface (Fig. 4). These two patterns are considered to be formed by deformation of the surface due to internal or external stresses. Since the misfit boundary between these portions consists of grids of screw dislocations, further growth preferentially takes place at these boundaries. Thus growth layers start either from growth centers, or from misfit or broken up portions, and spread two-dimensionally parallel to the basal plane.

Impurities or tiny crystals on the surface may have an effect on the spreading of growth layers. Figure 5 shows a tiny unoriented crystal of hematite, which has completely intercepted the spreading of thick growth layers, giving a pattern similar to the back current formed behind a rock in a stream. Figure 6 shows thick spiral growth layers

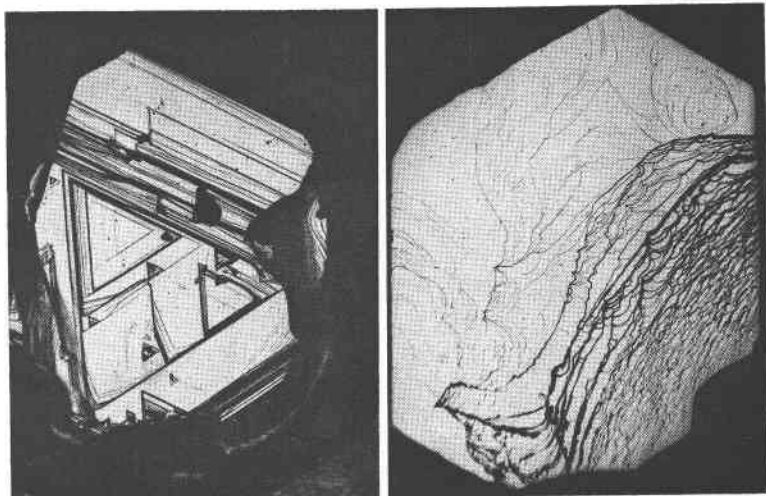


FIG. 1. (Left) Photomicrograph of thick triangular growth layers. Gihojuji, Japan.  $\times 5$ .  
 FIG. 2. (Right) Photomicrograph of thick irregular growth layers on the basal plane. Saganoshima, Japan.  $\times 5$ .

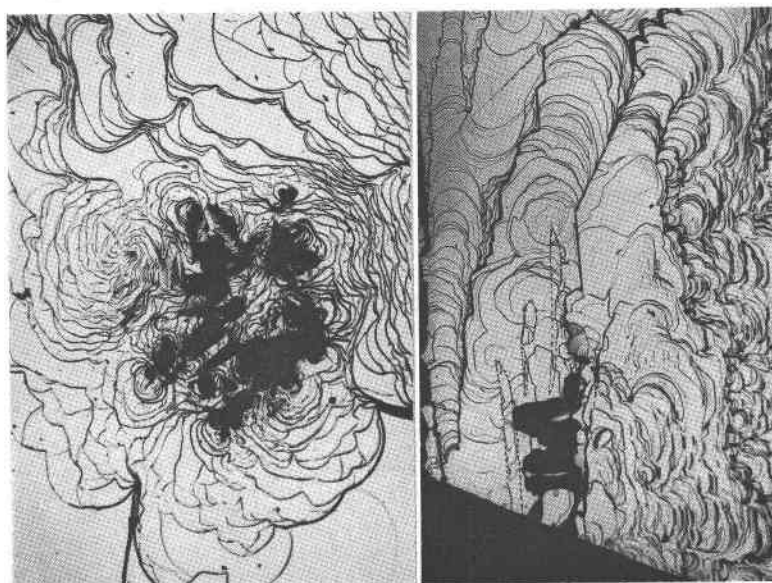


FIG. 3 (Left) Portion of surface broken up by internal stresses. Saganoshima, Japan.  $\times 25$ .  
 FIG. 4. (Right) Thick spiral layers starting from inclined portions (marked by dotted lines). The Azores Islands, Portugal.  $\times 25$ .

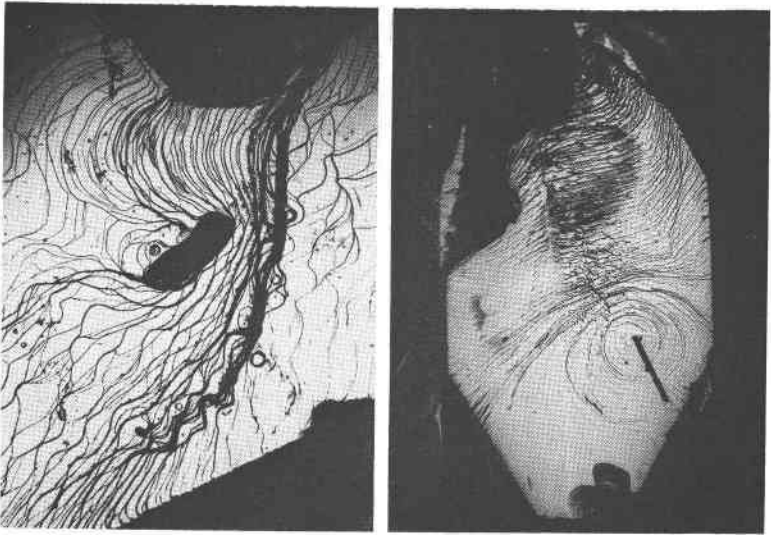


FIG. 5. (Left) Effect of an unoriented crystal on the spreading of thick growth layers, Ayumikotan, Japan.  $\times 25$ .

FIG. 6. (Right) Formation of a spiral pattern of thick growth layers around a twinned crystal (black rod). Saganoshima, Japan.  $\times 5$ .

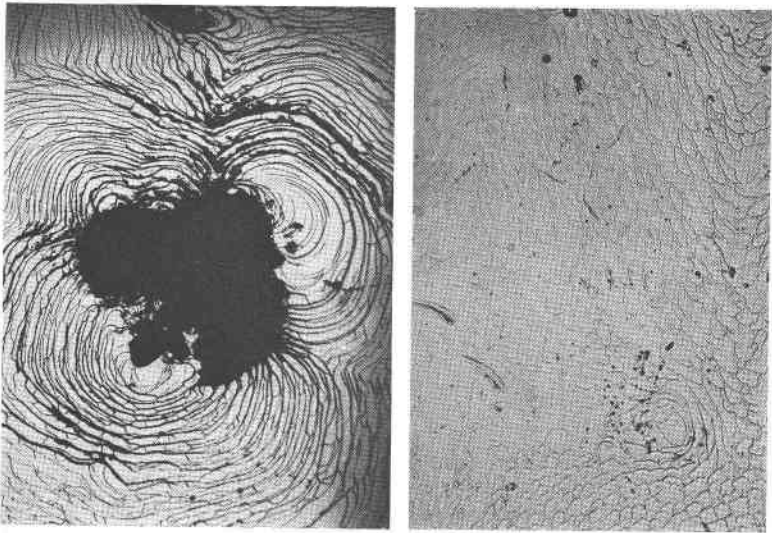


FIG. 7. (Left) Thick growth layers on the basal plane, starting from an attached crystal. Saganoshima, Japan.  $\times 25$ .

FIG. 8. (Right) Growth layers on the  $r(10\bar{1}1)$  face. Ayumikotan, Japan.  $\times 5$ .

formed around a small hematite crystal in twinned position. Thick growth layers may start from an unoriented crystal as a growth center, as shown in Fig. 7.

The surface structure of  $r\{10\bar{1}1\}$ ,  $m\{10\bar{1}0\}$  or  $a\{11\bar{2}0\}$  is quite different from that of the basal plane. When they are small, only striations parallel to the edges of the basal plane are observed. Growth layers may be formed parallel to larger faces, but their spreading is strongly affected by the edges of growth layers parallel to the base. Figure 8 shows a scale-like growth pattern on a  $r\{10\bar{1}1\}$  face. The growth layers are elongated parallel to the basal plane. The growth layers often start from the edges between the face and the basal plane and spread inward. This gives a hint as to the mechanism of the formation of skeletal faces commonly observed on hematite.

In the case of  $\{h0\bar{h}l\}$  or  $\{hk\bar{h}l\}$  faces, only striations parallel to the edges between these faces and the basal plane can be observed on the surface. This suggests that these faces are entirely formed by the edges of growth layers parallel to the basal plane.

From these observations, we can conclude that the major growth of hematite takes place by two-dimensional spreading and piling up of growth layers parallel to the basal plane, and the other faces are usually formed by the edges of these growth layers. Only when rhombohedral and prism faces develop, can new growth layers parallel to these faces develop, starting from the edges and spreading inwards.

#### OBSERVATIONS WITH A PHASE CONTRAST MICROSCOPE

If the surface is examined with a phase contrast microscope, very thin growth layers or spirals can be observed on the surface of thick growth layers, growth centers or flat areas. The following is a brief summary of observations of such surface patterns. All photomicrographs shown in this section are taken with a positive phase contrast setting, thus the white fringes always appear on the higher side of the step.

##### *Surface structures of growth centers*

There are two types of growth centers; one is flat and the other steep conical. The former are the growth centers of main growth layers, while the latter are formed by rapid growth and are the centers of local growth.

On the top of a steep conical center, many spiral growth fronts are usually observed. A typical example is shown in Fig. 9. It can be seen that spiral growth layers originated along a line, and not from a point. They are not typical spirals originating from a single screw dislocation, but originate from a misfit boundary consisting of a grid of dislocations.

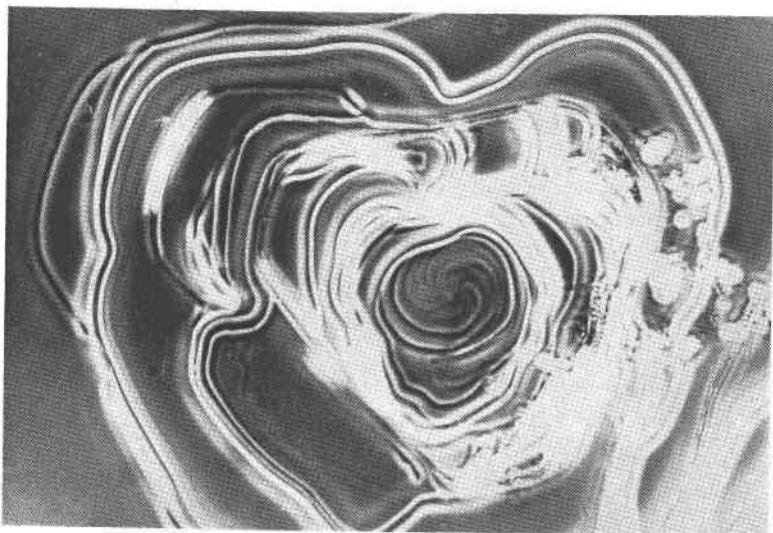


FIG. 9. Positive phase contrast photomicrograph of the surface of a steep conical growth center. Ayumikotan, Japan.  $\times 175$ .

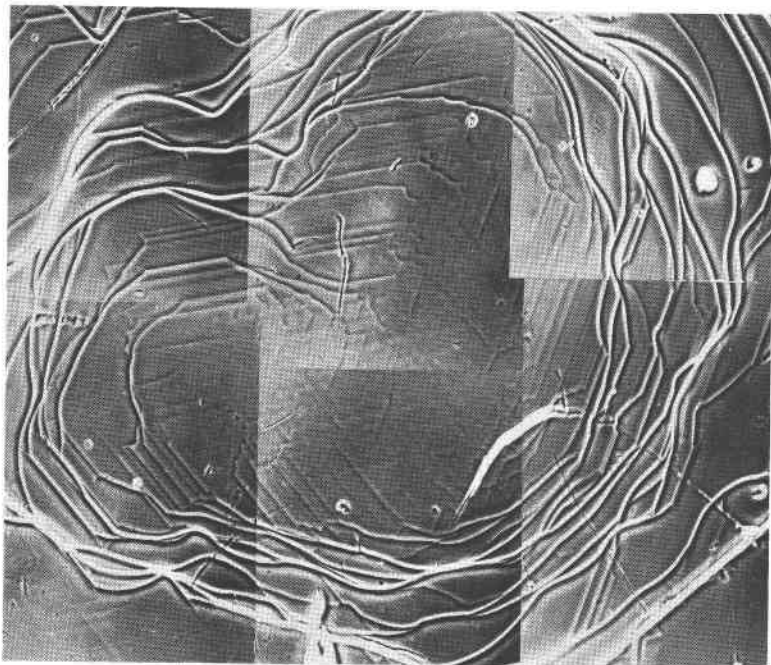


FIG. 10. Positive phase contrast photomicrograph of a flat growth center. Saganoshima, Japan.  $\times 75$ .

On a flat growth center, one can usually see large numbers of very thin growth layers; some irregular or circular, and others triangular. They usually originate from many spirals concentrated in a small area or along a line. Figure 10 shows many spiral-like triangular growth layers which originate along a curved line. However, if the surface is very flat over a wide area, on which no growth pattern is observed under an ordinary metallurgical microscope, this surface is usually covered with a spiral pattern starting from a single screw dislocation point or a few dislocation points, not from a group of dislocations. Figure 11 shows the

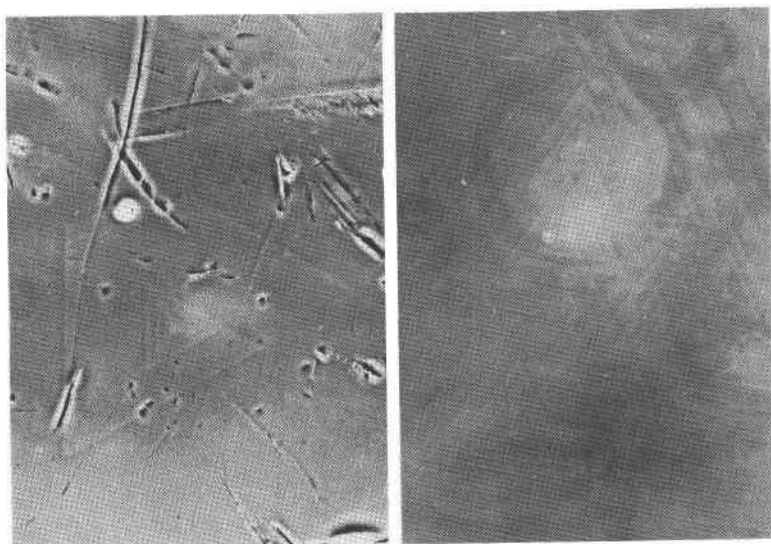


FIG. 11. (Left) Positive phase contrast photomicrograph of a triangular spiral which covers nearly half the surface of a basal face. Saganoshima, Japan.  $\times 150$ .

FIG. 12. (Right) Positive phase contrast photomicrograph of the central area of a very flat basal face. Several spirals are visible. Ascension Island.  $\times 150$ .

central part of the flat area of a crystal, on which triangular spiral layers starting from a single dislocation point can be seen. Although several weak spirals join the main one as it spreads, they do not interfere with the spreading of the main spiral. The height of this spiral is measured with multiple-beam interferometry (Tolansky, 1948) and found to be  $20 \text{ \AA}$ . Figure 12 is another example of the surface pattern of a flat area. In this case, as clearly seen in the photomicrograph, there are several typical growth spirals of  $4.6 \text{ \AA}$  step height situated close together. A wide flat area of this crystal is covered with the growth layers starting from these spirals. In the case of these spirals which cover the flat area, the step height of spiral layers is invariably very small. As a result of

measurement with multiple-beam interferometry, the step heights of 4.6, 7, 10, 20 Å, etc. are confirmed (Sunagawa, 1961).

These flat surfaces covered with a few very thin spirals are not a common feature. While in usual growth centers, growth layers are normally continuous from the center to the marginal part, in the case of very flat surfaces, there is a discontinuity between the flat area and the area consisting of thick layers. This suggests that the area covered with typical spirals is formed at the latest stage of crystallization.

In the case of crystals from the Ascension Island I, surface patterns are quite different from those of crystals from other localities. Thick growth layers have regular triangular form. On the surface of growth centers of these thick triangular layers, many thin triangular layers are observed. No spiral pattern can be seen on these surfaces.

#### *Structures on the surface of thick growth layers*

It has been observed that there are many thin growth layers as well as growth spirals on the surface of thick growth layers. Three different patterns are observed.

In the first type, only thin triangular growth fronts are found on the surface, and no growth spiral is observed.

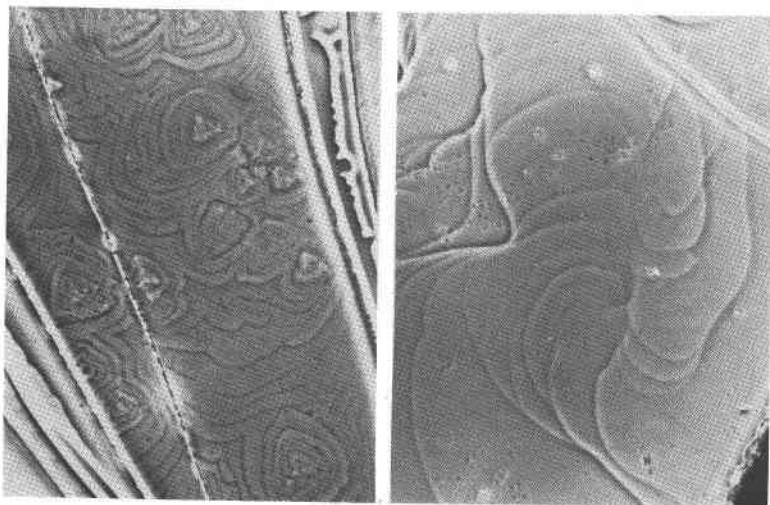


FIG. 13. (Left) Positive phase contrast photomicrograph showing the surface of thick growth layers covered with many spirals. The Azores Islands.  $\times 165$ .

FIG. 14. (Right) Positive phase contrast photomicrograph showing spirals starting from a row of dislocations, observed on the surface of thick growth layers. Saganoshima, Japan.  $\times 155$ .



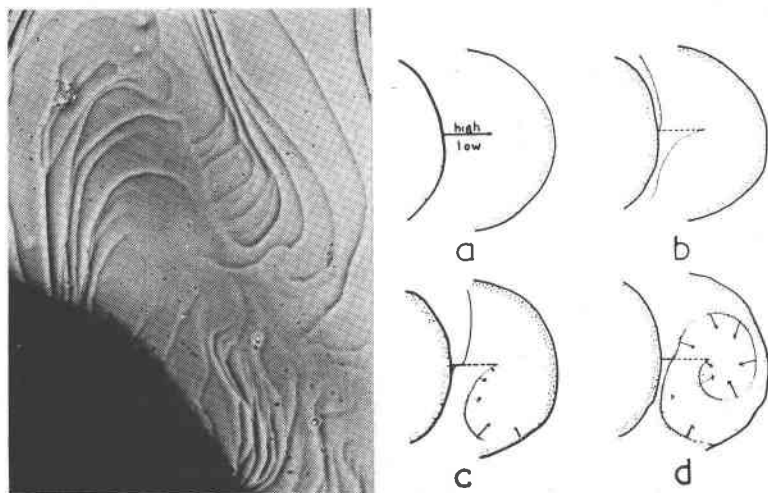


FIG. 15. (Left) Positive phase contrast photomicrograph showing negative spirals originating from a row of dislocations. Ayumikotan, Japan.  $\times 130$ .

FIG. 16. (Right) Drawing showing the process of formation of a depression spiral on the surface of thick growth layers. Dots show the higher side.

In the second type there are several growth spirals on the surface of a thick growth layer (Fig. 13). Screw dislocation points are scattered evenly over the surface, and there are many triangular spiral cones with step heights of  $2.3 \text{ \AA}$ .

A third type consists of a group of irregular spirals originating from a group of dislocations (Fig. 14). Irregular thin growth layers start from a line. They are not typical spirals, for the growth fronts have only a half turn, and do not complete themselves. The most likely reason for this line is a misfit boundary formed by mechanical deformation during growth, as in the case of similar patterns observed on the surface of steep conical growth centers. A negative form of this pattern is also often observed (Fig. 15). In this case, mechanism of formation is different from the usual spiral growth. Figure 16 is a figure showing the process of formation of negative spiral patterns. This mechanism can also be applied to the formation of depression spirals, which are sometimes observed on the surface of thick growth layers. It is often observed that there is a misorientated or inclined portion at the end of or near these irregular spirals. A typical example is shown in Fig. 17, in which the white portion is inclined to the basal plane. This suggests that the misorientated portion has close genetic relationship with the origin of spirals of this type. In other words, a row of dislocations from which

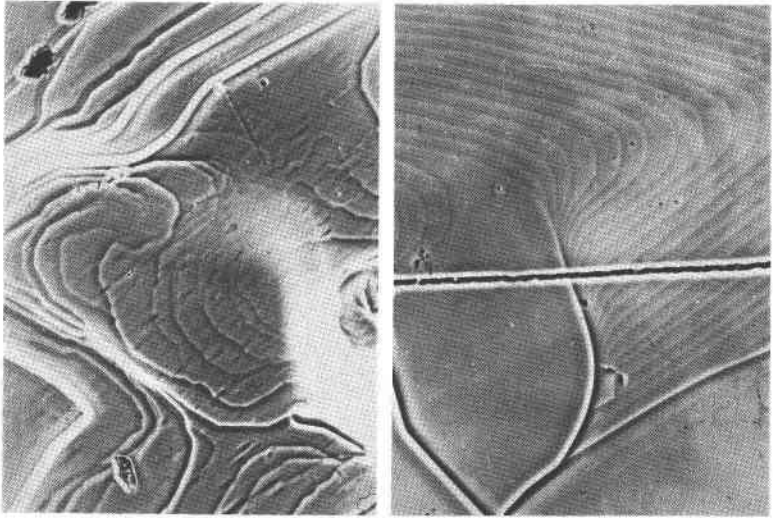


FIG. 17. (Left) Positive phase contrast photomicrograph showing relation between misoriented portion and groups of dislocations. Small white area is inclined to the main surface. Ayumikotan, Japan.  $\times 115$ .

FIG. 18 (Right) Positive phase contrast photomicrograph showing the formation of thick layers by bunching of thin layers. Horizontal straight line is a scratch on the surface. British West Indies.  $\times 140$ .

these irregular spiral layers originated is formed by breaking up or deformation of the surface during growth, external or internal stresses being responsible.

#### *Observations of very small crystals*

Surface structures described above are observed on large crystals. They are often too complicated, resulting from interference between growth layers from different centers, to clarify the mechanism of growth. For this, some observations have been specially made on very small crystals.

Crystals observed are those from Saganoshima, Japan, which occur in druses of decomposed basaltic lava. They are less than 1 mm in diameter. Of course, there is no fundamental difference of the surface patterns between these small crystals and other large crystals. However, two points come out clearly from these observations. These small crystals usually consist of a few portions which are slightly inclined towards each other. The boundary between two portions is sharp at one end, but disappears at the other and the two portions form a common surface. Growth layers are usually continuous from one portion to the other.

Usually one or several growth centers are situated near the edges of a crystal, but sometimes they start from the end of boundary lines. It is observed that thin growth layers or spirals usually start from a group or row of dislocations and have a pattern similar to spirals starting from a misfit boundary, and that there is no case in which the surface is covered with one or a few typical spirals as in the case of a very flat surface. This shows that at the early stage of growth, growth layers mainly originate from a structure such as a misfit boundary, which is considered to be formed by mechanical deformation of the surface during growth.

#### *Relation between thick and thin growth layers*

As described above, both very thick and very thin growth layers are observed on the surface of hematite crystals. Thick growth layers have step heights ranging from a few hundred to a few thousand Å, while thin growth layers have step height of 2.3 Å, 4.6 Å, 7 Å, 10 Å, 14 Å, 20 Å, etc. The following are observations concerning the relation between thick and thin growth layers.

Main growth centers have flat surfaces consisting of very thin growth layers, and in the marginal part of the surface, many thick growth layers are usually present. This suggests that as layers spread outward the rate of spreading decreases, and that there is close relation between thin and thick growth layers. Two reasons could account for the decrease of the rate of spreading. Since the total length of the growth edge of a layer increases as the layer spreads outwards, the rate of spreading decreases provided that atoms or molecules are supplied evenly all over the surface. Moreover, if the thickness of layers increases, the rate of advance will decrease, because there is a wider area on the cliff of thicker layers. This leads to the conclusion that thicker layers may be formed by the coalescing of thinner layers. This is observed on many crystals. Figure 18 is one of the most typical examples. Very thin growth layers (of step height of only 4.6 Å) coalesce to form a thick layer, the step height of which ranges from 4.6 Å to a few hundred Å. While thin growth layers have smooth and regular fronts, thick growth layers thus formed have irregular fronts. Retardation of spreading of thick growth layers is also observed in this photomicrograph. Therefore, it is quite clear that many thick growth layers are formed from very thin growth layers.

However, there is also another feature which clearly shows that thick growth layers can be formed from the beginning. For instance, as seen in Fig. 4, very thick growth spirals originate from large screw dislocations. No thin growth layers can be seen on the surface of these thick growth spirals. The writer has also reported that three-dimensional

growth islands having an average step height of  $40 \text{ \AA}$  are formed without the help of screw dislocations (Sunagawa 1960). These observations show that in some cases thick growth layers can be formed from the beginning without the help of bunching of thinner layers.

*Frequency of appearance of each different type of growth spirals*

Spiral patterns observed on the surface of the basal plane are roughly classified into the following types:

1. Triangular cones ( $2.3 \text{ \AA}$  triangular spirals)
2. Typical spirals having regular spacing between successive layers and small step heights ( $4.6 \text{ \AA}$ ,  $7 \text{ \AA}$ ,  $10 \text{ \AA}$ ,  $20 \text{ \AA}$ , etc.)
3. Spirals having very high step height.
4. Spirals originating from a group of dislocations.
5. Spirals originating from a row of dislocations.
6. Depression spirals, originating both from a single dislocation point and a row of dislocations.

Spirals of type 1 are observed on crystals from many localities. They occur not only on the surface of growth centers but also on the surface of thick growth layers. The surface of some crystals is completely covered with these cones, especially in the case of crystals from the Azores Islands. Frequency of appearance of this spiral varies in different localities. Spirals of type 2 are very rare. They have been observed only on a few crystals from Saganoshima, the Azores Islands, Ascension Island II, Stromboli, and British West Indies. They usually occur on the surface of a wide flat area but not on steep conical growth centers or on growth centers. They do not usually occur on the surface of thick growth layers, except on a few crystals from the Azores Islands. Spirals of type 3 are also not very common, but when they occur, the growth layers starting from these high step screw dislocations cover all the surface. Type 4 spirals are most common on nearly all crystals observed. They usually occupy the surface of growth centers of thick layers, but sometimes they occur on the surface of thick growth layers. Therefore, this type of spiral is considered to be the main origin of growth layers. Type 5 spirals are usually observed on the surface of thick layers and occur fairly commonly. Type 6 spirals are not very common and are observed mainly on the surface of thick growth layers.

*Summary of the observations and consideration*

The following are summaries of the observations on thin growth layers:

1. Thin growth layers are observed on both growth centers and the surface of thick growth layers,
2. On the surface of growth centers, spiral-like growth layers originating from a small

- area or a line are usually observed. On the flat area, typical spirals originating from one or several dislocations cover the whole surface, but this is not a common feature.
3. On the surface of thick growth layers, sometimes only growth fronts of thin layers are observed, and in other cases all types of spirals are observed.
  4. In the case of small crystals, spirals originating from a group of dislocations are observed but typical spirals have never been seen. The surface of these small crystals is usually broken up into several slightly inclined portions.
  5. It is observed that many thick layers are formed by bunching of thin growth layers, but at the same time it is also observed that some thick layers originate from big screw dislocations and some are from three-dimensional islands.
  6. On some crystals from Ascension Island, no spiral pattern of any kind is observed. The surface is covered with only triangular growth sheets of both thick and thin layers. This shows that the crystals have grown without the help of screw dislocations. On one crystal from Saganoshima, three-dimensional islands are observed, on which no spiral is seen.

From these observations, it can be concluded:

- a) Growth layers mainly originated from a group of dislocations spread outwards, and bunch together forming thicker layers.
- b) A typical single screw dislocation has an important role in growth only in some special cases, and only at the latest stage of growth.
- c) Many crystals have misfit portions, mechanically deformed portions, etc. from which many growth layers originate.
- d) Most of the isolated typical spirals, especially  $2.3 \text{ \AA}$  spirals, are formed at the latest stage of growth.
- e) Three-dimensional growth islands are formed without the help of screw dislocations at the earliest stage of growth.

It is necessary to consider in more detail the origin of groups of dislocations from which main growth layers arise. These are certainly neither typical single screw dislocations, nor a typical row of screw dislocations. They often have a close relation to misfit, twisted, or misoriented areas on the surface. Therefore, their most probable origin is deformation of the surface due to internal or external mechanical stresses during growth. Two cases are considered; one is that many dislocation points already present migrate into some small area by stresses applied, and form a group of dislocations; the other is that the surface is actually broken up or twisted by external or internal stresses. Because of these processes, imperfections of the surface are concentrated in a small area, to which atoms or molecules which precipitate on the surface will migrate and preferentially be absorbed. Thus growth layers originate mainly from these small areas. That the mechanical deformations of the surface such as percussion or impact of other material can generate new growth centers on the surface is experimentally demon-

strated on cadmium iodide (Sultan, 1953). This is perhaps one of the origins of growth centers of hematite crystals.

From these observations and considerations, the mechanism of growth of hematite can be described in the following way. At the early stage of growth, when the supersaturation is very high, growth mainly takes place by two-dimensional nucleation or attachment of crystallites which are formed without the help of screw dislocations. When the crystals reach some critical size, the surface is broken up by internal stresses or impact from other crystals, forming imperfections on the surface. Internal or external stresses will also result in migration of dislocations already present into a small area. From the imperfection areas thus formed, growth layers preferentially start and spread two-dimensionally. Thin growth layers bunch together and form thicker layers while they are spreading. When crystals are growing under high supersaturation conditions, a typical single spiral pattern may not be formed on the surface, even if screw dislocations terminate on the surface. This is because of rapid spreading of main growth layers from the above described centers. The main growth layers will encompass or engulf the growth layers from a typical screw dislocation. However, as the supersaturation rate decreases, the rate of spreading of the main growth layers will decrease. In fact under very low supersaturation conditions, thick growth layers will not be able to advance. Under these conditions, thin growth layers can start freely from screw dislocation points already present on the surface. Different spiral patterns observed both on the surface of thick growth layers and flat growth centers are considered to be formed in this way. The typical spiral patterns of very small step heights usually observed on the very flat areas are also considered to be formed at the latest stage of growth, when the supersaturation rate is very low. These typical spirals constitute only the very surface of a crystal, and the main part of a crystal is formed by growth layers originating from the above described imperfection areas or from three-dimensional growth islands.

#### DIFFERENCE OF GROWTH PATTERNS ACCORDING TO THE DIFFERENT LOCALITIES AND THE CONDITION OF GROWTH

It has been noticed that the crystals from each locality have their own characteristic surface structures. Since the growth condition is responsible for the growth pattern, the above difference is considered to show the difference of growth conditions in each locality.

A summary of different growth patterns in different localities is as follows:

1. Crystals from Ascension Island I consist of only triangular growth layers, and no spiral of any kind is observed.
2. Thick growth layers on the crystals from Gihofuji are regular triangles, but thin

growth layers are irregular or deformed triangles. Typical spiral pattern is not observed.

3. Crystals from Ayumikotan usually have irregular thick and thin growth layers, but no typical spiral is observed on them. Triangular cones are sometimes observed.
4. Thick growth layers on the crystals from Sasazawa, Saganoshima, Vesuvius, the Azores Islands, Ascension Island II and British West Indies have irregular or circular form. Both triangular cones and typical spiral patterns are observed on these crystals.
5. Frequency of appearance of triangular cones is highest on the crystals from the Azores Islands, next on the crystals from Saganoshima. On the crystals from the other localities, it is very low compared with the above two localities.
6. Typical spiral patterns are observed on the flat growth centers of the crystals from Sasazawa, Saganoshima, Vesuvius, Ascension Island II, British West Indies, while on the crystals from the Azores Islands, they are observed both on the flat centers and the surface of thick growth layers.

The Frank's screw dislocation theory (Frank, 1949) is based on the concept that two-dimensional nucleation can occur only under very high supersaturation such as more than 25% (Burton *et al.*, 1951). This leads to a conclusion that the crystal having no spiral pattern must be crystallized under very high supersaturation conditions. Crystals from Ascension Island I are considered to belong to this category. Frank also gave a formula for the relation between the supersaturation rate and the spacing between successive arms of a spiral. According to this formula, it can be qualitatively said that the wider the spacing between successive arms of a spiral the lower the rate of supersaturation. Therefore, the crystals having typical spirals of wide spacing are considered to have grown under lower supersaturation than those having no typical spirals. This shows that the crystals from Gihofuji and Ayumikotan have crystallized under a higher supersaturation condition than the crystals from the other localities.

As discussed earlier, spirals observed on the surface of thick growth layers are formed at the latest stage of crystallization, when the supersaturation rate must be very low. Therefore, the crystals from the Azores Islands are considered to have grown under lower supersaturation condition than those from the other localities, which have typical spirals only on the flat centers. Triangular cones which consist of 2.3 Å spirals are also considered to be formed at the latest stage of crystallization. The mode of occurrences of triangular cones is in accordance with the above consideration based on the mode of occurrences of typical spirals. They occur more frequently on the crystals from the Azores Islands and Saganoshima than on the crystals from the other localities.

From these considerations, the following sequence can be arranged according to the decrease of supersaturation rate.

Ascension Island I—Gihofuji—Ayumikotan—Sasazawa—Vesuvius—(Ascension Island II, British West Indies, Stromboli)—Saganoshima—The Azores Islands.

In the case of crystals from Gihofuji, Ayumikotan, Saganoshima and the Azores Islands, a large number of crystals have been observed. It is noticed that there are several different surface patterns on the crystals from one locality. Although most crystals from Ayumikotan show irregular growth layers, a few crystals from the same locality show triangular growth layers. In the case of the Azores Islands, a few crystals show typical spiral patterns on the surface of thick layers, and other crystals have either irregular spirals or triangular cones on the surface of thick growth layers. This variation is considered to show different stages of crystallization in one locality.

#### CONCLUSIONS

Surface structures of the basal plane of hematite crystals from different localities have been observed with a metallurgical and a phase contrast microscope.

As a result of these observations, the following points are concluded.

1. Growth of hematite mainly takes place by two-dimensional spreading of growth layers parallel to the basal plane.
2. Faces other than the basal plane are mainly formed by the edges of growth layers parallel to the basal plane. In the cases of  $\{10\bar{1}0\}$ ,  $\{11\bar{2}0\}$  or  $\{10\bar{1}1\}$  faces, they are formed at first by the above mechanism, but after they develop, growth layers parallel to them can be formed.
3. In the early stage of crystallization, growth mainly takes place by two-dimensional nucleation or attachment of crystallites which are formed without the help of screw dislocations.
4. Main growth layers originate from misfit, twisted, misoriented portions or a group of dislocations, which are formed on the surface by internal, external or mechanical stresses during growth.
5. Thin growth layers bunch together and form thicker growth layers. However, some thick growth layers are formed from the beginning.
6. When the supersaturation rate decreases at the later stages, typical spiral patterns are formed on the surface.
7. Characteristics of the growth patterns vary in the different localities. Difference in supersaturation condition is responsible for this difference. The sequence in nine localities is arranged according to decrease of the supersaturation condition.

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## REFERENCES

- BURTON, W. K., N. CABRERA AND F. C. FRANK (1951), The growth of crystals and the equilibrium structure of their surface. *Phil. Trans.* **A243**, 299.
- FRANK, F. C. (1949), The influence of dislocations on crystal growth. *Disc. Faraday Soc.* No. 5, 48.
- SULTAN, F. S. A. (1953), Optical and interferometric studies of crystal growth from solution. Ph.D. Thesis (London University).
- SUNAGAWA, I. (1958), Crystal growth of hematite from Ayumikotan, Japan. *Jour. Mineral. Soc. Japan*, **3**, 543.
- (1960), Growth history of hematite. *Am. Mineral.* (in press).
- (1961), Step height of spirals on natural hematite crystals. *Am. Mineral.* **46**, 1216–1226.
- TOLANSKY, S. (1948), *Multiple-beam Interferometry*. Clarendon Press, Oxford.

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