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### *The surface structure of crystals.*

(With Plates I-IX.)

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#### INTRODUCTION.

C. W. BUNN and H. Emmett<sup>1</sup> have observed the spreading of multi-molecular layers on growing crystals of artificial compounds, and have suggested that deposition takes place on the high-index surfaces at the edges of the layers. Their work led the author to examine crystals of several mineral species for evidence of the mechanism of growth, and this account is the result of a preliminary survey of the growth structures found.

So many growth structures could be seen with the naked eye or with a hand-lens that it was decided to use low magnifications in order that comparatively large areas of the crystal faces could be examined and photographed at one time. A metallurgical microscope with a prism reflector and 1-inch objective was used for the observations and most of the photography. This work is qualitative rather than quantitative, but it is intended to carry out a quantitative examination of the same specimens later.

The approach differs from that of Bunn and Emmett in two respects: the use of low magnifications and the observation of the crystals after rather than during growth. The structures to be described project above the general level of the crystal face, like etch hillocks. Growth by layer-spreading seems to offer the only satisfactory explanation of the origin of these structures and is further substantiated by their close similarity to the spreading layers which have been seen on growing crystals (e.g. Bunn and Emmett, loc. cit., 1949, figs. 4-7).

<sup>1</sup> C. W. Bunn and H. Emmett, Discuss. Faraday Soc., 1949, no. 5, p. 119. [M.A. 11-377.]

## STRUCTURES ON CRYSTAL FACES.

The origin of the structures can be understood more easily if the mode of layer-initiation and spreading is considered first. Several hypothetical cases are considered, in which layers are initiated on different parts of a face at either a constant or a variable rate and spread with either a constant or a variable velocity. Combinations of these variables give rise to several possible structures and types of habit. Since a number of these structures has been found it suggests that some, at least, of the hypothetical cases are valid.

## A GENERAL ACCOUNT OF CRYSTAL HABIT IN TERMS OF LAYER-SPREADING.

Two conditions are postulated:

1. The initiation of new layers, either monomolecular or multimolecular, is a comparatively infrequent event, the previous layer having spread completely, or nearly completely, across the crystal face before the new layer is formed.
2. The initiation of new layers takes place so rapidly that many of the earlier-formed layers are still spreading across the crystal face when the new layers are formed.

These two conditions must both be considered with each of the following cases:

- A. The layers are initiated within the edges of the crystal face.
- B. The layers are initiated at the edges of the face.
- C. The layers are initiated at the corners of the face.

The combinations of the conditions 1A, 1B, and 1C would all lead to the development of normal, plane-faced crystals. The combination 2A would lead to the growth of crystals with vicinal pyramids, the combination 2B would lead to the growth of hopper crystals, and 2C to skeletal or dendritic habits.

This concept of the development of hopper, skeletal, and dendritic crystals by very rapid layer formation agrees with the fact that such abnormal habits are usually developed only at very high supersaturations, when presumably layer-initiation is most rapid.

## A GENERAL CONSIDERATION OF LAYER-SPREADING.

If it is assumed that layers may be generated at a constant, increasing, or decreasing rate, and spread at a constant, increasing, or decreasing velocity, nine combinations will arise, the surface profiles of which

are given in table I. 'Increasing' and 'decreasing' in the column headed 'Velocity of layer-spreading' refer to the change in velocity of the edge of the layer as it spreads. 'Complex' is used for surfaces which have both concave and convex curvatures.

TABLE I. Possible combinations of rates of initiation and growth of layers and resultant surface profiles.

Case.	Rate of layer-initiation.	Velocity of layer-spreading.	Surface profiles.
1	Constant	Constant	Plane
2	Constant	Increasing	Concave
3	Constant	Decreasing	Convex
4	Increasing	Constant	Concave
5	Increasing	Increasing	Concave
6	Increasing	Decreasing	Concave or complex
7	Decreasing	Constant	Convex
8	Decreasing	Increasing	Convex or complex
9	Decreasing	Decreasing	Convex

Cases 1, 4, and 7 seem most likely to occur. Plane, concave, and convex surfaces have been found. Cases 6 and 8 seem least probable and no examples of them have been found.

The vicinal pyramid formed by rapid layer-initiation may be regarded as a large number of superimposed molecular layers growing outwards from a common origin and having a distance between adjacent layer-edges of a small number of atoms. Such a vicinal pyramid would have smooth surfaces with profiles as given in table I. Most vicinal pyramids have surfaces which are striated parallel to the layer-edges and appear to consist of multimolecular layers of approximately equal thickness. Alternating periods of rapid and slow layer-initiation, or periods of rapid initiation with pauses between them, would account for the striated vicinal pyramids. According to this interpretation each period of rapid initiation would produce one multimolecular layer, composed of a number of closely-spaced molecular layers. Each multimolecular layer would have plane, concave, convex, or complex profiles on its edges, depending upon its particular conditions of growth (see table I). The multimolecular layers would grow by deposition on the molecular steps.

#### THE HABIT OF GROWTH FORMS.

It has been observed that layers initiated within the edges of a crystal face may be polygonal, circular, or elliptical in plan, or bounded by intersecting arcuate lines, while occasionally they are ragged and irregular. Apart from the circular and irregular forms, the layers show the same degree of symmetry as the face on which they grew.

When layer-initiation is rapid a vicinal pyramid will be built up. In its usual crystallographic sense the word *pyramid* denotes a form in which all the faces are crystallographically equivalent. This is only true of *vicinal pyramids* in special cases, since, in the general case, they are bounded by a number of different forms, as in an etch pit. Steep vicinal pyramids closely resemble etch hillocks, so the term *vicinal hillock* will be used for all vicinal pyramids which are polygonal, or nearly polygonal in plan.

Since vicinal hillocks have been named by analogy with etch hillocks, it is desirable to use a different name for the less common circular and elliptical growth forms, for which the term *vicinal boss* is suggested. *Growth forms* will be used to embrace all vicinal hillocks and vicinal bosses.

#### TOPAZ (plates I and II).

##### *The prism* {110}.

One specimen, believed to come from Cornwall, has several topaz phenocrysts, some doubly-terminated, in a fine-grained rock groundmass. The (110) face of the largest crystal has five well-developed vicinal hillocks. One vicinal hillock has been photographed twice, with different lighting, to show the curvature on the sides towards (100) and (010) (figs. 1 and 2)<sup>1</sup>; figs. 3-6 show the other four vicinal hillocks.

All these figures emphasize the fact that the layers travel slowly from the point of origin towards the (010) edge, faster towards the (001) and (00 $\bar{1}$ ), and fastest towards the (100) edge. Thus strong striations are built up by the steep sides of the vicinal hillocks which face (010) (fig. 4). The latter are ( $hk0$ ) surfaces with  $h < k$ , and the opposite side of the vicinal hillock is bounded by ( $hk0$ ) surfaces with  $h > k$ . The other two sides of the vicinal hillock are bounded by pyramidal surfaces. The striations are not due to an oscillatory combination, if that is understood to mean an alternation of two planes. Figs. 2 and 4 show clearly that the surface profiles are concave.

The surfaces forming the striations all lie between the planes (110) and (120), except for the highest and steepest parts in figs. 3 and 6 which reflect light simultaneously with (120). Since the interfacial angle (110):(120) is  $18^{\circ} 44'$  it represents an unexpectedly steep slope for a vicinal form. On the side of the vicinal hillock facing (100) the slope is

<sup>1</sup> The use of face or form symbols in this paper to describe *directions* on a crystal face does not necessarily imply that those faces or forms are present on the crystal.



less steep, although somewhat variable (figs. 1, 4, and 6). Figs. 1, 2, and 4 show examples of concave surface profiles.

The salient edge between adjacent faces of the vicinal hillock may be straight or sinuous (figs. 4 and 3). The latter case indicates changes in the relative growth velocities of the layers on adjacent sides of the salient edge.

It is interesting that nearly all these vicinal hillocks have a minute particle of impurity at or immediately adjacent to the apex of the hillock. It is not possible to assert definitely that these impurities have caused initiation of growth as there are many other particles on the face which have no visible effect.

Some of these particles have held up growth layers, causing *growth shadows*. A 'cliff' due to this cause can be seen on the steep side of the vicinal hillock in figs. 1 and 2. A similar but longer cliff appears in fig. 6, while in fig. 3 the steep edge has been held back in two successive steps by impurities.

On the sides of the hillocks bounded by pyramidal surfaces the edges of the layers make an angle of approximately  $2^\circ$  with the edge (110) (001). Thus the pyramidal surfaces have an index of the type  $\{hkl\}$  and not  $\{hhl\}$ .

Fine curved lines can be seen on the flatter surfaces of some vicinal hillocks (figs. 1 and 2). Similar curved lines have also been observed macroscopically, bounding the layers on  $\{110\}$  of a topaz crystal from Nigeria. In the figures mentioned the central, steep vicinal hillock rests on an earlier-formed hillock, like a nearly square plinth. Layers have spread from another initiation point near the edge of the plinth. They remained quadrilateral until they reached the steep side of the larger hillock, when the layer-edges became curved. The re-entrant angles formed when the two sets of layers met offered preferential positions of attachment. The additional deposition at these positions, combined with the continued spreading of the layers from the second initiation point, would account for the curvature of the layer-edges.

Crystals of topaz from the Thomas Range, Utah, usually have many intergrown vicinal hillocks similar in habit to those described from Cornwall. One vicinal hillock is shown in fig. 7.

#### *The prism zone [001].*

The common forms on the brown crystals from Ouro Preto, Minas Geraes, Brazil, are  $\{110\}$ ,  $\{120\}$ , and  $\{111\}$ . The striated prism zone has a complex structure. Although alternating elements of  $\{110\}$  and  $\{120\}$  are present it is not only an oscillatory combination, as the adjacent

planes are often connected by concave surfaces, also many of the finer and some of the coarser striations are not parallel. Many of the narrower striations are composed of surfaces lying between  $\{110\}$  and  $\{120\}$ . Vicinal hillocks on  $(110)$  have the steep side facing  $(120)$ , while those on  $(120)$  have the steep side facing  $(110)$ . Fig. 8 shows a vicinal hillock on  $(120)$  elongated in the direction of the  $c$ -axis, parallel to which it forms striations. In fig. 8 the upper part of the steepest side is bounded by  $(110)$  (the white band), and concave pyramidal surfaces can be seen.

Fig. 9 shows a D-shaped vicinal hillock on  $(110)$  of a crystal believed to come from Nigeria. The steep side faces  $(120)$ .

The elongation of such a structure would account for the curved layers in fig. 10, seen on  $(110)$  of a magenta-coloured crystal from Ouro Preto. The gently arcuate layers spread across the face until they were stopped at a striation, which was thereby increased in height. The inclined surfaces forming the striations, which are dark in the figure, lie between  $(110)$  and  $(120)$ .

The magenta crystal has D-shaped vicinal hillocks on  $(120)$  (fig. 11). They are similar to the one in fig. 9, but face in the opposite direction, with the steep side towards  $(110)$ . The outer layers are D-shaped, although the inner ones are quadrilateral, and show a transition from quadrilateral to arcuate layers.

Some of the brown crystals from Ouro Preto have D-shaped vicinal hillocks in which the arcuate part has a slightly concave profile. Several of these structures have a small particle of impurity at the apex.

#### BERYL (plates II and III).

On a blue crystal from Brazil, showing hexagonal prism and basal pinakoid, the layers on  $\{10\bar{1}0\}$  appear to have spread rapidly in the direction of the  $c$ -axis and slowly normal to it, thereby forming elongated vicinal hillocks with steep prismatic and more gently inclined pyramidal surfaces. Each end of the vicinal hillock may have one or two pyramidal surfaces. Occasionally the pyramidal ends are irregular; the parallel layers with sinuous edges of fig. 12 are asymmetric extensions of a vicinal hillock, beside which is a normal hillock.

Most of the aquamarine crystals examined from Spitzkopje, South-West Africa, show the forms  $\{10\bar{1}0\}$ ,  $\{11\bar{2}0\}$ ,  $\{10\bar{1}1\}$ ,  $\{11\bar{2}1\}$ , and  $\{0001\}$ . The crystals are usually prismatic in habit, but two are tabular on  $\{0001\}$ . On the prism  $\{10\bar{1}0\}$ , the numerous vicinal hillocks present are similar to those in fig. 12, but both the prismatic and pyramidal surfaces have convex profiles.

On the prism  $\{11\bar{2}0\}$  the layers form vicinal bosses which are elliptical or nearly circular in plan, and each boss has an etch pit at its centre. Fig. 13 shows the initiation points of four vicinal bosses and part of two other bosses. In fig. 14 the prismatic surfaces are concave and the pyramidal surfaces conical and slightly concave.

The bipyramid  $\{11\bar{2}1\}$  has numerous vicinal hillocks, one of which is shown in fig. 15. The layers are nearly in the form of a parallelogram, although there is a slight curvature of the edges. The two upper faces are seen, and one of the lower, which is in deep shadow. The two upper surfaces are convex, and presumably have high indices. They lie in zones passing through  $\{11\bar{2}1\}$  and  $\{10\bar{1}1\}$ . The two lower surfaces lie in zones passing through  $\{11\bar{2}1\}$  and  $\{10\bar{1}0\}$ . The two upper faces thus belong to one form, or set of forms, and the two lower faces to another. There is a sudden change in the slope of the vicinal hillock.

On the bipyramid  $\{10\bar{1}1\}$  only one vicinal hillock has been found. It is a comparatively thin layer which is almost an equilateral triangle in plan. One side is parallel to the intersection with  $\{10\bar{1}0\}$ , and the other two to the intersection with  $\{11\bar{2}1\}$ , therefore at least two pyramidal forms are involved as growth surfaces.

The basal pinakoid (fig. 16) shows series of hexagonal layers which have spread from numerous centres. The layer-edges are straight and parallel to the intersections with  $\{10\bar{1}0\}$ , and are bounded by a form in zone with  $\{10\bar{1}0\}$  and  $\{0001\}$  which makes a small angle with the latter.

#### BARYTE (plates III and IV).

*The basal pinakoid.*—The vicinal hillocks commonly have the shape of a parallelogram in plan, the edges of which are parallel to the intersections of  $\{001\}$  and  $\{110\}$ . The edges of the layers are thus bounded by  $\{hhl\}$  surfaces. Vicinal hillocks are well developed on the yellow crystals from Frizington, Cumberland, which have been described by Miss J. M. Sweet.<sup>1</sup> They are elongated on the *b*-axis (figs. 17 and 18). The crystals often consist of several slightly mis-aligned individuals, the ledges or steps between which appear to have exerted a considerable influence on growth. In fig. 17 a series of layers can be seen, bounded on one side by a step. It appears that, as soon as the growing layers passed the end of the step, some of them 'doubled back' along the other side. The sinuous nature of the salient edge of the vicinal hillock shows that

<sup>1</sup> J. M. Sweet, *Min. Mag.*, 1930, vol. 22, p. 268.

the layers on adjacent sides have spread with changing relative velocities, the group of layers on one side then on the other travelling faster alternately. This suggests a structure-sensitive control of growth. In fig. 18 several series of layers can be seen to have started from the ends of steps. The central parallelogram seems to have had two initiation points, resulting in two obtuse corners on one side. The curvature of the layer-edges between the two corners is reminiscent of the more smoothly curved layers on the topaz from Cornwall and Nigeria described above, and may be due to the same cause.

On crystals from the Silverband mine, Westmorland, which are elongated on the  $a$ -axis,  $\{011\}$ ,  $\{001\}$ , and  $\{102\}$  are the best developed forms. The vicinal hillocks commonly have the same form as those described from Frizington, but are not so finely striated. Sometimes the vicinal hillocks are six-sided, with an additional pair of sides in the zone  $[010]$ . On some crystals the  $\{hhl\}$  and  $\{hOl\}$  surfaces are convex.

A crystal from Nutfield, Surrey, shows vicinal hillocks on  $\{001\}$ , most of which have a minute crystal of chalcopyrite and of calcite in the centre.

*The prism*  $\{110\}$ .—Crystals from the north of England often have fine striations on  $\{110\}$ , parallel or normal to the  $c$ -axis, or in both directions. In many cases rectangular vicinal hillocks can be seen, their direction of elongation determining that of the major striations. Fig. 19 shows a vicinal hillock elongated parallel to the  $c$ -axis, initiated at, or very near, the edge of the face. Fig. 20, from the same specimen, shows the major striations in the other direction. The layers are bounded by pyramidal surfaces  $\{hhl\}$ , and by prismatic surfaces  $\{hko\}$ , with  $h > k$  on one side and  $h < k$  on the other. As the layers spread outwards they sometimes break up into a large number of minute rectangular steps, as in fig. 20, giving rise to the curved lines sometimes seen on  $\{110\}$ .

*The brachydome*  $\{011\}$ .—Some crystals of baryte from the Silverband mine have numerous circular layers, as in fig. 21. They are similar to the structures on ammonium sulphate described by J. Ames, T. L. Cottrell, and Avis M. D. Sampson.<sup>1</sup> In fig. 21 the upper surfaces of the layers are plane, but in most cases the vicinal bosses consist of rounded hummocks. The latter are presumably formed when layer-initiation continues to the end of growth.

*The macrodome*  $\{102\}$ .—Triangular vicinal hillocks are present on some crystals from the Silverband mine.

<sup>1</sup> J. Ames, T. L. Cottrell, and A. M. D. Sampson, *Trans. Faraday Soc.*, 1950, vol. 46, p. 940, and fig. 5.

## FLUORITE (plates IV–VI).

Reference will be made to four specimens in describing the structures on fluorite. The first, from Weardale, County Durham, is a mauve 4-cm. cube, twinned on the spinel law, on which only  $\{100\}$  is present. The second is a pale mauve specimen consisting of twinned 4–8-mm. cubes, with a hexakisoctahedron, from the Sedling mine, Weardale. The third is a mauve specimen consisting of twinned and untwinned cubes up to 2 cm., in combination with an  $\{hk0\}$  and an  $\{khl\}$  form. The locality is uncertain, but the specimen has the appearance of fluorite from Weardale, and will be referred to as the 'mauve fluorite'. The fourth specimen consists of two green twinned 2-cm. cubes from County Durham, and will be called the 'green fluorite'.

Fig. 22 shows part of the (100) face of the 4-cm. cube, on which a vicinal hillock has been initiated by the twin crystal. The plan view is approximately square, but the edges are slightly curved, as is common in fluorite. The salient edge of the vicinal hillock is shown in figs. 23–26, in which the angle between adjacent layer-edges is approximately  $126^\circ$ . Thus the edge of each layer makes an angle of  $18^\circ$  with the nearest cube edge. Sir Henry Miers<sup>1</sup> records vicinal  $\{hk0\}$  forms on fluorite, but they would not account for the curved edges of the layers. The (001) face of the 4-cm. cube has a re-entrant corner of the twin crystal projecting through it. All three edges of the re-entrant corner initiate a vicinal hillock at the point where they touch the cube face. Two of these vicinal hillocks, which are octagonal, not square, are shown in figs. 27 and 28. The alternate interior angles of the octagons are approximately  $127^\circ$  and  $143^\circ$ . Farther from the point of origin the layers do not form a continuous smooth curve between adjacent salient edges of the vicinal hillock, but break up into a number of triangular layers which form projecting tongues (as in fig. 29, of the green fluorite) or form layers with zigzag edges composed of many such tongues (fig. 30, green fluorite). In the aggregate these tongues give a gentle curvature to the outline of the vicinal hillock in plan view. The edges of the tongues in fig. 29 make an angle of approximately  $18^\circ$  with their nearest cube edges. In the specimen from the Sedling mine the layer-edges on both sides of the salient edge of the vicinal hillock are parallel to the intersection of the cube and hexakisoctahedron, and the layer-edges make an angle of approximately  $18^\circ$  with their nearest cube edge. The surface on the edge of the cube layers on the three specimens described and on the mauve fluorite thus appears to be a hexakisoctahedron.

<sup>1</sup> H. A. Miers, *Mineralogy*. 2nd edit., 1929, p. 358.

The  $\{hko\}$  faces of the mauve fluorite are interrupted by numerous projections. Some are shown in fig. 31, which is a view normal to (100), so that  $(hk0)$  appears dark. As the microscope was focused on the projections, the layers on the cube face are slightly out of focus. The projections have surfaces parallel to (100) with a zigzag edge (angle  $143^\circ$ ). It is clearly an extension of the layer-system of the cube projecting from  $(hk0)$ . The two lower faces of the projection belong to an  $\{hkl\}$  form, for they reflect light simultaneously with such a form which is developed on the corners of the crystal. One of the  $\{hkl\}$  faces of another projection is shown in fig. 32, looking approximately normal to  $(hk0)$ . The latter, slightly rounded and mottled, appears on the right, and the edges of the layers seen above it are on a cube face, which is viewed obliquely. On the left is the projection, looking like a spiral staircase; the 'treads' being the cube planes (similar to those seen from above in fig. 31) and the 'rises' belonging to an  $\{hkl\}$  form.

In fig. 23 three sets of edges can be seen, which all belong to the same  $\{hkl\}$  form. Since growth would be likely to occur on any of them with equal facility, the layers may travel normal to the length of the step or laterally along it. The latter is well illustrated by some thin layers in fig. 25, which also shows that the forward movement of a layer may begin at the salient edge of the vicinal hillock. A projection appears at the corner of the layer, and gives rise to layers which have travelled along and not across the steps. In fig. 22 a 'transgression' can be seen. The vicinal hillock exhibits a sudden change of slope, and the boundary between the steep and shallow parts is transgressive, the layers of the steeper part cutting obliquely across those beneath. Thus the upper layers must have been travelling faster than those beneath. The steeper part has a convex surface. Convex surfaces are also seen on the green fluorite (fig. 33), in which the ellipses are due to layers parallel to the cube face growing out of the sloping side of the vicinal hillock. (Very smooth concave surfaces have been observed on a specimen from the Stotsfieldburn mine, Weardale.)

*The effect of fractures on growth.*—Within the 4-cm. cube are many veil-like fractures several centimetres in length, containing liquid and gaseous inclusions. It is evident that some of the fractures were present at the surface during growth, for they have affected the layer-pattern. Where two of them cross smooth areas of the faces (100) and (010) a line of triangular, tongue-like layers appears. In some places the fractures appear to have held up the layers temporarily, producing minute steps, the smallness of which testifies to the relative ease with which the layers

were able to bridge the gaps. Fig. 22 shows the only example of a marked obstruction which was found. A shallow triangular depression may be seen near the upper left-hand corner of (100). It is bounded by a fracture on one side of which many layers collected before the obstruction was overridden. By the time growth ceased the steep front of the arrested layers was two millimetres beyond this fracture.

*Growth shadows.*—Most of the re-entrant corners which appear within the boundary of a cube face on the mauve fluorite have formed growth shadows. Layers initiated at one end of the re-entrant corner have not spread all round the obstructing corner but continue almost undeviated after passing it, thus building up a 'cliff', as seen in fig. 34. The end of the re-entrant corner is on the left and the layers in the upper part of the figure were spreading from left to right, and formed a cliff after passing the corner. No layers were initiated by this edge of the re-entrant corner.

#### PYRITE (plates VI and VII).

*The cube.*—Variation in the shape of the layers is well illustrated by pyrite, in which four-, six-, and eight-sided layers have been found on {100}. The crystal shown in fig. 35 is bounded by {100}, {111}, and an {hkl} form. The layers approximate to squares, but are somewhat irregular. The layer-edges are parallel to the intersections of {100} and {111}. Another crystal with the same forms has octagonal layers (fig. 36). The layer-edges are bounded by a diploid and pentagonal dodecahedron. Neither of these crystals shows the striations characteristic of pyrite.

In some crystals of pyrite it can be seen that the oscillatory combination of adjacent pairs of faces of {210}, which may or may not be truncated by {100}, is produced by vicinal hillocks. Each ridge of the striations is one vicinal hillock, which is hexagonal in plan and very much elongated. The long sides have pentagonal dodecahedral surfaces and each end of the hillock is bounded by two surfaces which are probably diploidal. The latter are sometimes clearly defined, but more often have an irregular character. Fig. 37 shows the hexagonal vicinal hillocks on a slightly striated cubo-octahedron from Elba, and fig. 38 shows them on a very deeply striated cube the locality of which is unknown.

*The octahedron.*—Most of the octahedron faces examined have equilateral triangular vicinal hillocks. Figs. 39 and 40 show one face of a crystal from Elba. On (111) the edges of the vicinal hillock have a clockwise rotation of some  $15^\circ$  from the edges of the face, which is bounded by

{321}. On ( $1\bar{1}1$ ) the rotation is anti-clockwise. Supplementary twinning brings about the coincidence of (111) and ( $1\bar{1}1$ ), and produces a 're-entrant angle' in the vicinal hillock, as shown in fig. 40, in which the black line is the twin-boundary. The surfaces of the vicinal hillock appear to be diploidal, with a higher index than {321}.

Another habit is shown in fig. 41. The crystal is predominantly octahedral, with small {100}, {210}, and { $hkk$ } faces, the last forming the sharply defined edges of the vicinal hillock.

The orientation of the vicinal hillocks which are bounded by diploids on {111} is not constant, but shows different amounts of rotation on different specimens, even when allowance is made for the forms bounding {111}.

#### APATITE (plate VIII).

Interesting growth forms were found on the first-order prism and bipyramid of asparagus-stone from Durango, Mexico. The centre of a vicinal hillock on the prism is shown in fig. 42. Fig. 43 shows another part of the same hillock. The vicinal hillock is bounded laterally by prismatic surfaces, which form striations parallel to the  $c$ -axis, and at the ends by { $hk\bar{l}m$ } or { $h.h.2\bar{h}.m$ } surfaces. Several fractures, some of which have veil-like inclusions, cross the confused area at the centre of the vicinal hillock. The remarkable regularity of the layer-spacing in both the prismatic and pyramidal zones in fig. 43 suggests the operation of a rhythmic process controlling the initiation of layers. One of the vicinal bosses on the pyramid is shown in fig. 44.

A crystal from Cornwall shows a vicinal hillock on ( $10\bar{1}0$ ) with layer-edges bounded by prismatic and { $h0\bar{h}l$ } surfaces (fig. 45). Layers are initiated at, or very near, the edge of the face. Hillocks of this habit have also been found on vanadinite crystals from the Apache mine, Arizona.

#### QUARTZ (plates VIII and IX).

Figs. 46 and 47 (smoky quartz found near Andermatt, Switzerland) show that the very characteristic striations normal to the  $c$ -axis on the prism faces of quartz crystals are caused by elongated vicinal hillocks. The long edges of the layers are bounded by rhombohedral surfaces and the short edges are bounded by trigonal trapezohedron or trigonal pyramid surfaces, on which growth is much more rapid than on the rhombohedral surfaces.<sup>1</sup> In fig. 47 growth shadows can be seen, caused

<sup>1</sup> Since this paper was read G. van Praagh and B. T. M. Willis have corroborated this explanation of the origin of the horizontal striations on quartz. (*Nature*, 1952, vol. 169, p. 623).



by particles of impurity on the surface. Figs. 48 and 49, of a crystal from the same locality, show the usual triangular vicinal hillocks of the unit rhombohedron. The straight layer-edges have rhombohedral surfaces, and give rise to striations parallel to the edge of the prism. One of the most interesting features is the apparent ease with which the layers completely surround particles of impurity on the surface of the crystal. These particles may be seen in all stages of encirclement in fig. 49. In fig. 50, a rhombohedron face of a crystal from the iron mines of Cumberland, the remarkable regularity of spacing of the layer-edges seems to indicate a rhythmic process of layer-initiation.

#### MAGNETITE (plate IX).

The crystals have the combination  $\{111\}$  and  $\{110\}$ . The  $\{111\}$  faces are brilliant, while the  $\{110\}$  faces are dull and laminated, being formed by the edges of layers which have spread across the octahedron faces. The equilateral triangular layers on  $\{111\}$  are bounded by an  $\{hhl\}$  form near  $\{111\}$  in orientation (fig. 51).

#### CELESTINE (plate IX).

Fig. 52 shows a vicinal boss on (011). It occurs on a crystal from Yate, Gloucestershire, on the surface of a ghost crystal which has been re-exposed by fracture along a parting. Other crystals from this district have rectangular vicinal hillocks on  $\{001\}$  and  $\{102\}$ , both having one set of edges parallel to their common intersection and another set at right angles to it.

#### TOURMALINE (plate IX).

Vicinal hillocks have been found on the basal plane of two crystals from Arassuahy, Brazil. One example is shown in fig. 53. On the other crystal the layers have the shape so characteristic of basal sections of tourmaline, an 'equilateral triangle' in which the sides are rounded.

#### GROWTH FORMS ON OTHER MINERALS.

Vicinal bosses have been found on stilbite, and vicinal hillocks on galena, haematite, wulfenite, chessylite, diopside, chabazite, and harmotome.

#### DISCUSSION OF OBSERVATIONS.

One of the most interesting points is the very widespread occurrence of growth forms on crystals. The frequent appearance of curved surfaces on growth forms seems to support the assumptions underlying table I, that they are due to changes in the rate of initiation or spreading of the layers, or to both factors combined.

*Frequency and origin of multimolecular layers.*

Multimolecular layers have been seen during growth on many crystals of soluble substances, and the layers recorded here show such similarity to them that they probably also grew as multimolecular layers. If so, their development seems to be more common than is generally to be expected on theoretical grounds.

According to the dislocation theory of crystal growth multimolecular layers may be generated at low supersaturations by a single screw dislocation having a large Burgers vector, or by a closely spaced group of dislocations. At high supersaturations multimolecular layers may be initiated by the same mechanism, or they may arise from the aggregation or bunching of monomolecular layers formed by two-dimensional nucleation or by dislocations.

In most of the growth forms illustrated in this paper a large number of layers can be seen to have a common origin. (In some cases the origin has moved during growth, but the subsequent layers are again concentric with the new origin, as in fig. 1.) This seems to suggest that growth may have been caused by dislocations, unless there is any reason why successive layers formed by two-dimensional nucleation should be concentric. Several further observations seem to suggest the action of dislocations, although they do not prove it.

In a number of vicinal hillocks on topaz, particles of impurity were seen at the points of origin of the layers. In baryte, layers were seen to emanate from points on the surface at which structural deformations ended (fig. 18). Both phenomena suggest the presence of structural discontinuities, and hence the possibility of dislocations, in Frank's sense,<sup>1</sup> existing at the initiation points. Since etching is likely to start at points of structural discontinuity, the etch pits at the centres of the elliptical layers on beryl (fig. 13) may have the same significance, but it should be noted that there are many other etch pits from which no layers emanate.

*The velocity of layer-spreading.*

In the dislocation theory the rate of advance of monomolecular layers has been calculated. It has been shown that the rate is dependent on the degree of supersaturation, and increases with it. Although no detailed predictions have been made for multimolecular layers, it is suggested that their velocity of spreading will likewise be governed by supersaturation.

<sup>1</sup> F. C. Frank, Discuss. Faraday Soc., 1949, no. 5, p. 48.

Some of the observations presented in this paper suggest that the degree of supersaturation does not entirely determine the velocity of layer-spreading on a given face of a particular crystal. Consider the salient edge between two adjacent faces of a vicinal hillock. If the two faces are crystallographically equivalent the salient edge should be straight, because the layers on both sides of the edge should have the same growth velocity, and any change in the supersaturation should affect them identically. (Differences of supersaturation should be negligible over such small distances.) But, in fact, appreciable departures from rectilinearity occur, and the salient edge has a sinuous form, often with quite sharp curvatures. Such is the case for baryte in fig. 17. Highly sinuous salient edges between crystallographically equivalent forms have also been observed on  $\{100\}$  of fluorite and  $\{001\}$  of wulfenite. The layers on adjacent sides of a salient edge are clearly spreading with changing relative velocities. Measurement shows that in the lower part of fig. 17 the layers on the right of the salient edge were spreading just over five times as fast as the corresponding layers on the left. Immediately below this area the layers on the left were spreading with approximately eight times the velocity of those on the right. These are large factors; much larger than are likely to be caused by possible slight variations in supersaturation.

The origin of these sudden, large changes of relative velocity must be sought in the structure of the face rather than in the state of the solution. There is no evidence of lineage structure or rows of dislocations parallel to the edges of the layers which might have retarded them. If, however, the edges of these multimolecular layers are vicinal in character, as suggested by Bunn and Emmett, and the index of the surface changes during growth, a change in the growth velocity would also be expected. Vicinal forms are known to be very susceptible to changes of index during growth, through causes not yet understood. A difference of the index on adjacent sides of the salient edge would account for the phenomena observed.

Another phenomenon which does not seem to be explicable in terms of the dislocation theory is the transgression of higher layers over lower ones on fluorite in fig. 22. The upper layers of the vicinal hillock were evidently spreading faster than those beneath, and it is possible that the latter had even ceased to grow. This transgression could be accounted for if the rate of layer-spreading, at a given supersaturation, varied with the index of the form bounding the edges of the multimolecular layers. That growth may proceed on some parts of a crystal surface and not on

others is shown by the lack of growth at growth shadows, as seen in topaz, quartz, and fluorite (figs. 1, 3, 6, 47, and 34).

Another point of apparent disagreement with the predictions of dislocation theory is the steepness of many of the vicinal forms illustrated in this paper. Frank<sup>1</sup> contrasts the 'unusually steep vicinal faces' (e.g.  $3\cdot0^\circ$ ) caused by twinning in fluorite 'with  $20'$ ', which is the most which is usually observed for growth pyramids centred on some ordinary point in the face of a crystal<sup>2</sup>. He says that in the steep vicinal pyramids on fluorite the rate of emission of growth fronts, compared with their rate of spreading, is of the order of ten times as great as when the initiating centre is a simple dislocation or group of dislocations.

No exact measurements have yet been made on the growth forms described here, but in the case of topaz one side of a vicinal hillock seems to show an inclination of over  $18^\circ$ . There is evidence that some of the vicinal hillocks have inclinations of over  $5^\circ$ . All the text-figures are photomicrographs, except figs. 22, 33, and 50. In the photomicrographs any face appears dark when its normal is inclined at approximately  $5^\circ$  (or more) to the optic axis of the microscope. This suggests that the faces of several vicinal hillocks have an inclination of  $5^\circ$  (or more), while some of the moderately dark faces may have inclinations of, say,  $2^\circ$  to  $4^\circ$ . Twinning was not responsible for these steep growth forms, except in the case of fluorite and possibly also in figs. 46 and 47, both of quartz.

#### GENERALIZATIONS DRAWN FROM THE OBSERVATIONS.

*Regularity of layer-spacing.*—A phenomenon of interest is the remarkable regularity of the layer-spacing around some initiation points, as in figs. 43 and 50, which strongly suggests a rhythmic process of layer-initiation. W. E. Garner<sup>2</sup> has suggested that layer-initiation is associated with periodic changes of the supersaturation. These may also be the cause of the regular layer-spacing.

*Changes of slope on vicinal hillocks.*—Some vicinal hillocks show sudden, discontinuous changes in the slopes of their surfaces. One vicinal hillock on the 4-cm. cube of fluorite shows five such changes. Other examples are seen in fluorite (figs. 29 and 33), beryl (fig. 15), and topaz (fig. 2). Since the layers show no evidence of transgressions in these cases a probable explanation seems to be a sudden change in the rate of initiation. Sir Henry Miers noted sudden changes in vicinal forms during growth.<sup>3</sup>

<sup>1</sup> F. C. Frank, Discuss. Faraday Soc., 1949, no. 5, p. 187.

<sup>2</sup> W. E. Garner, Discuss. Faraday Soc., 1949, no. 5, p. 187.

<sup>3</sup> H. A. Miers, Phil. Trans. Roy. Soc. London, Ser. A, 1904, vol. 202, p. 459.

*Nature of growth surfaces.*—It has been shown that growth often takes place on a general form, not necessarily of lowest index, or on special forms of high index. In the case of fluorite, deposition is taking place on  $\{hkl\}$  surfaces when layers spread across  $\{100\}$ .

*Effect of flaws and particles of foreign substances on spreading layers.*—In fluorite most of the flaws which reach the surface seem to have hindered the spreading layers remarkably little. In quartz the layers appear to have spread round and engulfed solid particles of an impurity with no indication of hindrance (fig. 49). In topaz and quartz the layers passed the particles of impurity and left growth shadows (figs. 1, 3, 6, and 47).

*Habit of growth forms.*—The variable habit of growth forms is well illustrated by those on  $\{110\}$  of topaz and on  $\{100\}$  and  $\{111\}$  of pyrite (figs. 1, 9, 35–38, 39, and 41). Bunn and Emmett (loc. cit., 1949, p. 121) have observed a change in the shape of layers when specific soluble impurities were added. The variation described here is probably due to the same cause.

*Striations on crystals.*—It has been shown how the striations on crystals of several minerals are formed by the slow spreading of the layers on one side or two opposite sides of a vicinal hillock, while the layers travel faster on the other sides (e.g. topaz, figs. 1, 4, 8, and 9; beryl, fig. 12; quartz, figs. 46, 48, and 49; and pyrite, figs. 37 and 38). It is hard to visualize any other mechanism which would account for the striations and oscillatory combinations. The surface of the striations may be plane, concave, or convex.

#### THE VIEWS OF BUNN AND EMMETT.

The present work gives general support to their views on crystal growth by deposition on the high-index surfaces at the edges of layers. They usually found only one system of spreading layers on each face. That the author sometimes found quite large numbers of vicinal hillocks on a face may be due to the larger crystals observed, the majority of faces having areas between 0.25 and 2 square cm. In fig. 16 (beryl) the spacing of the vicinal hillocks varies from about 0.1 to 0.3 mm. Bunn and Emmett (loc. cit., 1949, p. 120) and Bunn<sup>1</sup> record that layers very often start from the centres of faces. The initiation points of the layers seem to have a random distribution on the crystal faces. No evidence has been found to suggest that they are preferentially situated at face-centres. Apart from two possible exceptions (figs. 19 and 45) none of the initiation points were situated at the corners or edges of crystal faces.

<sup>1</sup> C. W. Bunn, *Discuss. Faraday Soc.*, 1949, no. 5, p. 140.

The growth layers were all observed on substances of low solubility. Even in the presence of mineralizers at high temperatures and pressures, it seems unlikely that these substances would have solubilities as high as 6 %, which was the lowest solubility of any substance on which Bunn and Emmett saw layers during growth.<sup>1</sup>

The different habits of layers recorded here are very likely to have been produced by impurities, as noted by Bunn and Emmett (*loc. cit.*, 1949, p. 121).

These authors suggest that the absence of flat surfaces on faces and edges of layers during growth may be due to indiscriminate deposition (Bunn and Emmett, *loc. cit.*, 1949, p. 128). D. R. Hale<sup>2</sup> quotes the rounded surfaces on the basal pinakoid of artificially grown quartz in support of this view. A very high degree of order, in structure and growth, is the outstanding characteristic of crystals. It seems wrong to invoke indiscriminate processes if ordered processes can explain phenomena satisfactorily. Indiscriminate deposition suggests a random process. Random deposition would, in time, add uniform amounts of solute all over a face, and would not tend to make it rounded, either locally or as a whole. It seems more satisfactory to account for curved surfaces as closely-spaced series of molecular layers which have grown under one of the conditions which can give rise to curved surface profiles, as shown in table I.

The author finds it hard to accept Bunn's view (*loc. cit.*, 1949, p. 132) that diffusive flow, once started, can initiate layers. An inconceivably complex structure of diffusive flow would have to be evolved to deliver sufficient solute to initiate layers at points about 0.2 mm. apart but not between them. It seems more likely that the controlling factor is a structure-sensitive process of some kind.

It also seems improbable that once growth is started its velocity bears no relation to supersaturation.

#### GROWTH OF MULTIMOLECULAR LAYERS.

A qualitative hypothesis is suggested below to account for the observed growth properties of multimolecular layers, in particular the variable velocities of spreading and the stopping and restarting of growth. It takes into account deposition on the high-index edges of layers while also allowing a relation between supersaturation and rate of growth. Five postulates are made:

<sup>1</sup> C. W. Bunn and H. Emmett, *Discuss. Faraday Soc.*, 1949, no. 5, p. 124.

<sup>2</sup> D. R. Hale, *Discuss. Faraday Soc.*, 1949, no. 5, p. 189.

1. That growth velocity is a function of supersaturation, such that an increase of supersaturation increases growth velocity.
2. That in general, at a given supersaturation and temperature, forms of higher index grow more rapidly than those of lower index.
3. Each form has a critical supersaturation below which growth will not take place on a perfect face.
4. The value of the critical supersaturation bears an inverse relation to the index of the form, forms of low index having a high value of the critical supersaturation, and vice versa.
5. Deposition is entirely, or almost entirely, confined to the high-index surfaces forming the edges of the layers.

1 and 2 above are widely accepted; and 5 seems to be supported by observation. There is a small amount of evidence for 3, but it is very difficult either to prove or to disprove experimentally.

*Nature and functions of growth layers.*—The layers on crystals may be regarded as having two morphologically distinct parts with different functions, the high-index edge, or *growth surface*, and the comparatively large low-index plane upper surface, or *resting surface*. The growth surface has an index which is, in general, high, but may vary during growth. Sir Henry Miers has recorded changes of index of vicinal forms during growth (loc. cit., 1904, p. 459).

If the present hypothesis is correct the growth surface will have a low but variable value of the critical supersaturation required for growth, and the low-index resting surface will have a high value of the critical supersaturation, growth only taking place at high, perhaps very high, supersaturations. A crystal placed in a solution of moderate supersaturation will grow by deposition on the growth surfaces, thus extending the resting surfaces, on which no deposition is taking place.

This concept of growth surfaces and resting surfaces on crystals emphasizes the dual functions of the layers. The growth surface provides a surface on which growth can take place easily while the plane resting surface keeps the crystal polyhedral, and tends to minimize the surface energy of the whole crystal. In fluorite, for example, the growth surface is  $\{hkl\}$  and the resting surface is  $\{100\}$ . The cube appears to be the equilibrium form for fluorite grown from solution. Sir Henry Miers says, 'The habit of fluor is remarkably constant; no mineral crystallises more persistently in cubes'.<sup>1</sup> If the cube is indeed the equilibrium form it is the one best suited to be the resting surface, as it possesses the lowest surface energy.

<sup>1</sup> H. A. Miers, *Mineralogy*. 2nd edit., 1929, p. 357.

*Velocity of spreading of growth layers.*—Sinuous edges between 'crystallographically equivalent' faces of vicinal hillocks, caused by differential growth rates, could be accounted for by a difference in the index of the growth surfaces on the two sides. The 'equivalence' would apply to the zone in which the faces lie, but not to their indices. Thus in fig. 17 all the growth surfaces would have an index  $\{hhl\}$ , but the value of  $l$  would vary.

*Stopping and restarting of growth.*—The stoppage of growth of a face has been observed by Bunn (loc. cit., 1949, p. 132) and S. P. F. Humphreys-Owen.<sup>1</sup> The latter has also seen the restarting of growth. According to this hypothesis growth will cease when the growth surface changes from one of higher index to one of lower index, if the critical supersaturation for growth of the new face is higher than the supersaturation prevailing in contact with the crystal. The restarting of growth may be due to an increase in the supersaturation above the critical value for the new face, or to a dynamic interchange of a few ions or molecules between the crystal and its environment, producing a minute element of face with a low enough critical supersaturation to allow growth to proceed. The latter suggestion would agree with Humphreys-Owen's observation that the restarting of growth began at a single point on the face.

*Justification of high-index surfaces.*—A crystal which is in equilibrium with its exactly saturated solution will be bounded by the form or combination of forms, the equilibrium form(s), giving the lowest total surface energy for the crystal. Equilibrium forms can only be expected to appear at negligibly small supersaturations and are of theoretical rather than practical importance. They should not be expected, nor should their non-appearance be regarded as anomalous, on crystals grown at appreciable supersaturations. In the thermodynamic sense crystallization is a spontaneous process and therefore not a reversible reaction. Thus the maximum energy cannot be obtained from the change. Of the total energy potentially available in the reaction the major part will be liberated as heat, but the rest will be retained as surface energy, so that the total surface energy of the crystal will be above the minimum value. This implies that forms will be developed which have a higher surface energy than the equilibrium form(s).

#### STIMULATION OF GROWTH BY TWINNING.

In fluorite growth may be stimulated by twinning. Sir Henry Miers (loc. cit., 1929, p. 358) states, 'In these twins, at the exact point where an

<sup>1</sup> S. P. F. Humphreys-Owen, Discuss. Faraday Soc., 1949, no. 5, p. 144.



edge emerges, the cube-face is often raised into a very low pyramid consisting of four vicinal faces  $\{hk0\}$ . In the same connexion Frank (loc. cit., 1949, p. 187) states, 'Each face of these twin crystals shows a pronounced growth pyramid of unusually steep vicinal faces centred on the common line of the twin faces which meet on the composition plane'. The latter is taken to mean that the centre of the growth pyramid may occupy any position on any of the three lines common to the cube face and the re-entrant corner. The author's observations suggest that *steep* vicinal pyramids only form at the exact point of contact of the *edge* of a re-entrant corner with a cube face. A few layers may sometimes be seen to have spread from the common lines of the cube face and re-entrant corner, but no steep vicinal pyramids have been observed in such positions. The edges of the re-entrant corner seem to be particularly active in initiating layers, even when bevelled by  $\{hk0\}$ . The layers then start from the edges common to adjacent pairs of  $\{hk0\}$  faces, and not from the edges between  $\{hk0\}$  and  $\{100\}$ . All three edges of one re-entrant corner may initiate layers, as on the 4-cm. cube, two edges of its re-entrant corner being seen in figs. 27 and 28. The layers may be square, 'squares' with gently curved sides, or octagonal in plan. When only one edge seems to have initiated layers the interesting question arises as to why the other two edges were not similarly active.

The phenomenon of the stimulation of growth by the re-entrant angles of twins has been raised by I. N. Stranski<sup>1</sup> and F. C. Frank (loc. cit., 1949, p. 186). While it certainly applies in many cases to fluorite, it seems doubtful if it applies to all twins with re-entrant angles. The very thin crystals of cerussite, *tabular on*  $\{010\}$ , which form stellate trillings do not support this contention, and many other mineral examples could be cited in this connexion. The re-entrant angles of some twins may be self-perpetuating, while others may not, as in the cube and octahedron, respectively, twinned on the spinel law. In the latter case, if stimulation of growth occurred in the re-entrant angle, it would soon cease to exist, and so would the stimulus. The very presence of the re-entrant angle in these twins argues against its effectiveness in promoting growth.

#### EFFECT OF MODE OF GROWTH ON THE DONNAY-HARKER HYPOTHESIS.

This hypothesis<sup>2</sup> postulates that the morphological importance of a form is proportional to its reticular density. If growth takes place by two-dimensional nucleation and subsequent spread of the layers, the

<sup>1</sup> I. N. Stranski, Discuss. Faraday Soc., 1949, no. 5, p. 69.

<sup>2</sup> J. D. H. Donnay and D. Harker, Amer. Min., 1937, vol. 22, p. 446. [M.A. 7-241.]

growth-velocity of a face is entirely determined by the rate of initiation of layers and is unaffected by their rate of spreading. If the Donnay-Harker hypothesis is true this implies an inverse relationship between reticular density and rate of layer-initiation.

On the other hand, growth may take place by the spreading of multi-molecular layers originating at screw dislocations. If, as suggested above, the growth-velocity of a face, at a given supersaturation, depends on the nature of the growth surface at the edge of the layer, the change of index which occurs on vicinal surfaces during growth would make the calculation of reticular density, and hence of morphological importance, impossible. In fluorite it would be useless to calculate the reticular density of  $\{100\}$  in order to determine the morphological importance of the cube, if growth is taking place on  $\{hkl\}$  surfaces; while in topaz a layer which has spread across  $\{110\}$  has grown on three different kinds of surface,  $\{hk0\}$ ,  $\{k0h\}$ , and  $\{hkl\}$ , and each of these surfaces might consist of several forms in the zone concerned.

#### EXPLANATION OF PLATES I-IX.

##### PLATE I.

- FIG. 1. Topaz, Cornwall? (110), *c*-axis parallel to length of page. Steep, dark side of vicinal hillock faces (120). Concave surface on left faces (100).  $\times 55$ .
- FIG. 2. Topaz, same as fig. 1. Shows fine curved lines. Concave surface on right faces (120).  $\times 55$ .
- FIG. 3. Topaz, Cornwall? (110), *c*-axis parallel to length of page. Steep side of vicinal hillock faces (120). Two growth shadows near apex of hillock formed by particles of impurity.  $\times 55$ .
- FIG. 4. Topaz, Cornwall? (110). Steep side of vicinal hillock forms striations parallel to *c*-axis. Striations have concave surfaces.  $\times 55$ .
- FIG. 5. Topaz, Cornwall? (110), *c*-axis parallel to length of page. Angle of  $4^\circ$  between edges of adjacent vicinal hillocks indicates high-index pyramidal surfaces.  $\times 55$ .
- FIG. 6. Topaz, Cornwall? (110). Striation, in shadow, parallel to *c*-axis. Particle of impurity at apex of vicinal hillock causes long growth shadow.  $\times 55$ .

##### PLATE II.

- FIG. 7. Topaz, Thomas Range, Utah. (110), *c*-axis parallel to length of page. Steep side of vicinal hillock faces (120).  $\times 43$ .
- FIG. 8. Topaz, Ouro Preto, Brazil. (120). Longer striations parallel to *c*-axis.  $\times 43$ .
- FIG. 9. Topaz, Nigeria? (110), *c*-axis parallel to length of page. D-shaped vicinal hillock; steep side faces (120).  $\times 33$ .

- FIG. 10. Topaz, Ouro Preto, Brazil. (110). Arcuate layers form striations parallel to  $c$ -axis.  $\times 43$ .
- FIG. 11. Topaz, Ouro Preto, Brazil. (120). D-shaped vicinal hillock showing transition from quadrilateral to arcuate layers. Steep side faces (110).  $\times 50$ .
- FIG. 12. Beryl, Brazil. (10 $\bar{1}$ 0). Longer sides of vicinal hillock form striations parallel to  $c$ -axis. Sinuous layers are asymmetric extension of another vicinal hillock.  $\times 36$ .

## PLATE III.

- FIG. 13. Aquamarine, Spitzkopje, South-West Africa. (11 $\bar{2}$ 0),  $c$ -axis parallel to length of page. Vicinal bosses, each with an etch pit at the centre.  $\times 33$ .
- FIG. 14. Aquamarine, Spitzkopje, South-West Africa. (11 $\bar{2}$ 0),  $c$ -axis parallel to length of face. Vicinal boss with concave surfaces.  $\times 33$ .
- FIG. 15. Aquamarine, Spitzkopje, South-West Africa. (11 $\bar{2}$ 1). Vicinal hillock showing convex curvature and abrupt change of slope. Trace of  $c$ -axis parallel to length of page.  $\times 33$ .
- FIG. 16. Aquamarine, Spitzkopje, South-West Africa. (0001). Edges of hexagonal layers parallel to intersections of  $\{10\bar{1}0\}$  and  $\{0001\}$ .  $\times 33$ .
- FIG. 17. Baryte, Frizington, Cumberland. (001). In upper part of the figure layers spread approximately from left to right, prevented from spreading downwards by the ledge or step. When end of step was reached layers spread round it and returned along lower side, from right to left.  $\times 33$ .
- FIG. 18. Baryte, Frizington, Cumberland. (001),  $b$ -axis coincides with the major axis of the parallelogram-shaped layers. Layers are initiated at the ends of steps.  $\times 33$ .

## PLATE IV.

- FIG. 19. Baryte, Frizington, Cumberland. (110). Long edges of layers parallel to  $c$ -axis. Layers initiated at, or very near, edge of face.  $\times 33$ .
- FIG. 20. Baryte, same specimen as fig. 19. (110). Long edges of layers normal to  $c$ -axis. Edges of layers stepped.  $\times 33$ .
- FIG. 21. Baryte, Silverband mine, Westmorland. (011). Circular vicinal bosses.  $\times 36$ .
- FIG. 22. Fluorite, Weardale, County Durham. Part of (100) face of 4-cm. cube, showing transgression of steeper, convex part of vicinal hillock over layers beneath, projection of layers along the salient edge, and depression in top left-hand corner where layers were hindered by a fracture.  $\times 2.1$ .
- FIG. 23. Fluorite, Weardale, County Durham. (100) face of 4-cm. cube, showing part of salient edge of vicinal hillock. Layers have travelled from right to left along the steps from the salient edge.  $\times 55$ .

## PLATE V.

- FIG. 24. Fluorite, Weardale, County Durham. (100) face of 4-cm. cube, showing part of salient edge of vicinal hillock. The layer-edges on adjacent sides are not at right angles, as they would be if growth had taken place on an  $\{hk0\}$  form.  $\times 36$ .

- FIG. 25. Fluorite. (100) face of 4-cm. cube, showing a part of the salient edge at which projecting layers appear. They give rise to layers which travel laterally along the steps on the faces of the vicinal hillock.  $\times 36$ .
- FIG. 26. Fluorite. (100) face of the 4-cm. cube, showing a part of the salient edge at which a thick layer appears to transgress across two thinner layers.  $\times 36$ .
- FIG. 27. Fluorite. (001) face of the 4-cm. cube. Vicinal hillock initiated by re-entrant corner of twin crystal. The layers are octagonal, and represent an  $\{hkl\}$ , not an  $\{hk0\}$ , vicinal form.  $\times 36$ .
- FIG. 28. Fluorite. As fig. 27; octagonal layers initiated by another edge of the same re-entrant corner.  $\times 36$ .
- FIG. 29. Fluorite, County Durham. A cube face. Surface of vicinal hillock on the green fluorite, showing two abrupt changes of slope and three projecting tongue-like layers.  $\times 33$ .

## PLATE VI.

- FIG. 30. Fluorite, County Durham. Face of vicinal hillock on cube face of the green fluorite. Layers with zigzag edges; an aggregate of many triangular tongues.  $\times 50$ .
- FIG. 31. Fluorite, Weardale? View of the mauve fluorite normal to (100), which reflects the light. Triangular layers project from the (dark)  $(hk0)$  face.  $\times 36$ .
- FIG. 32. Fluorite. Another projection on same specimen as fig. 31, but viewed approximately normal to the  $(hk0)$  face (mottled). Upper surfaces of projections bounded by  $\{100\}$ , lower surfaces by  $\{hkl\}$ .  $\times 36$ .
- FIG. 33. Fluorite, County Durham. Cube face of the green fluorite showing steep vicinal hillock with convex surfaces, curved edges of the layer-aggregates forming the faces of the hillock and ellipses due to layers parallel to the cube face projecting from the sloping surface of the vicinal hillock.  $\times 2.2$ .
- FIG. 34. Fluorite, Weardale? (100). Growth shadow (broad dark band running left to right) on the mauve fluorite caused by re-entrant corner of twin crystal (black triangle on left).  $\times 36$ .
- FIG. 35. Pyrite. (100). Irregular square layers with edges parallel to intersections of  $\{100\}$  and  $\{111\}$ .  $\times 110$ .

## PLATE VII.

- FIG. 36. Pyrite. (100). Octagonal layers with edges parallel to intersections of  $\{100\}$  with  $\{hk0\}$  and  $\{hkl\}$ .  $\times 33$ .
- FIG. 37. Pyrite, Elba, Italy. (100). Hexagonal layers with length parallel to striations on (100). Bounding surfaces on long edges are  $\{hk0\}$ , on short edges probably  $\{hkl\}$ .  $\times 36$ .
- FIG. 38. Pyrite. (100). Very deeply striated cube. Shows one striation, formed of elongated vicinal hillock, similar in habit to fig. 37.  $\times 50$ .
- FIG. 39. Pyrite, Elba. (111). Triangular vicinal hillock probably bounded by diploidal surfaces.  $\times 36$ .

- FIG. 40. Same crystal as fig. 39. (111). Re-entrant angle formed in vicinal hillock by supplementary twinning. Black line is the twin boundary.  $\times 33$ .
- FIG. 41. Pyrite. (111). Very regular layers bounded on their edges by an  $\{hkk\}$  form.  $\times 36$ .

## PLATE VIII.

- FIG. 42. Apatite, Durango, Mexico.  $(10\bar{1}0)$ ,  $c$ -axis parallel to longer layer-edges. Centre of vicinal hillock. It is intersected by several fractures.  $\times 33$ .
- FIG. 43. Apatite, Durango, Mexico. Another part of vicinal hillock seen in fig. 42, showing remarkably regular spacing of the layer-edges.  $\times 33$ .
- FIG. 44. Apatite, Durango, Mexico. Circular vicinal boss on pyramid face.  $\times 33$ .
- FIG. 45. Apatite, Cornwall.  $(10\bar{1}0)$ .<sup>‡</sup> Longer striations parallel to  $c$ -axis. Vicinal hillock with sinuous salient edge, initiated at, or very near, edge of face.  $\times 33$ .
- FIG. 46. Smoky quartz, near Andermatt, Switzerland.  $(10\bar{1}0)$ . The longer edges of the vicinal hillocks form the characteristic striations normal to the  $c$ -axis.  $\times 33$ .
- FIG. 47. Smoky quartz, near Andermatt, Switzerland. Same crystal as fig. 46. A similar vicinal hillock on  $(10\bar{1}0)$  with growth shadows caused by particles of impurity.  $\times 33$ .

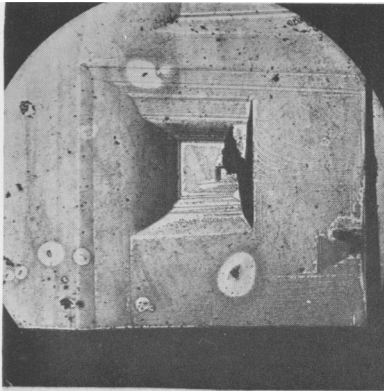
## PLATE IX.

- FIG. 48. Smoky quartz, near Andermatt, Switzerland. Vicinal hillock on rhombohedron face forms striations parallel to prism edge.  $\times 33$ .
- FIG. 49. Smoky quartz, near Andermatt, Switzerland. Same crystal as fig. 48, and showing particles of impurity in all stages of encirclement by the layers.  $\times 33$ .
- FIG. 50. Quartz, Cumberland. Remarkable regularity of layer spacing on a rhombohedron face.  $\times 4.3$ .
- FIG. 51. Magnetite. (111).  $\times 33$ .
- FIG. 52. Celestine, Yate, Gloucestershire. (011). Vicinal boss on surface of a ghost crystal re-exposed by fracture along a parting.  $\times 33$ .
- FIG. 53. Tourmaline, Arassuahy, Minas Geraes, Brazil. Triangular growth layers on basal plane.  $\times 33$ .

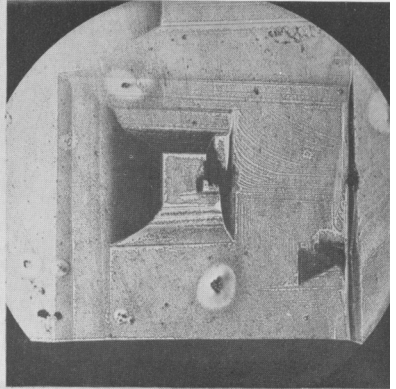
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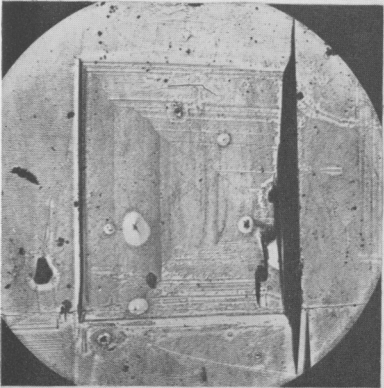
This paper forms part of a thesis approved for the degree of Ph.D. by the University of London.



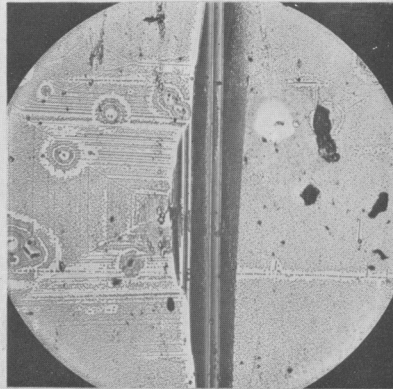
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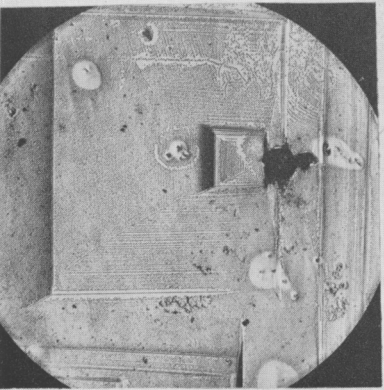
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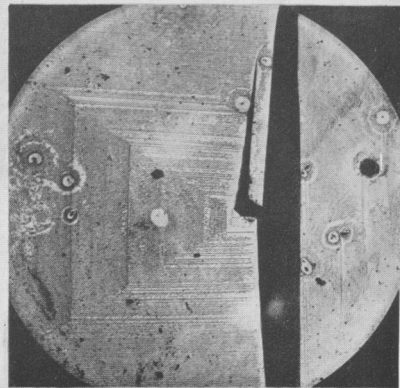
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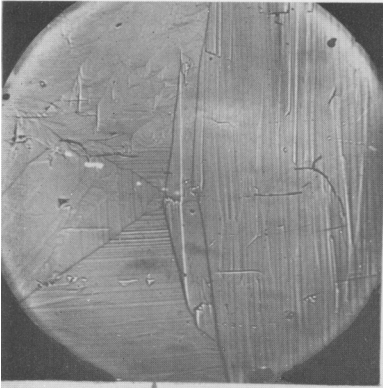
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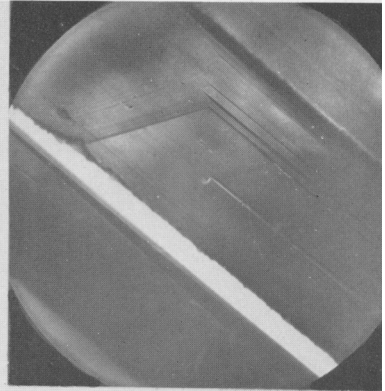
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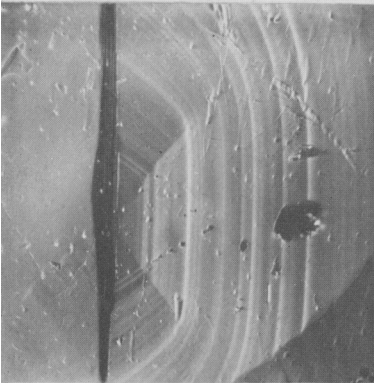
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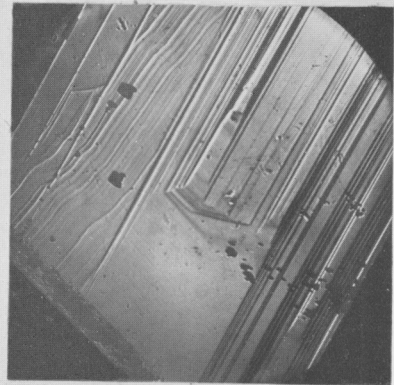
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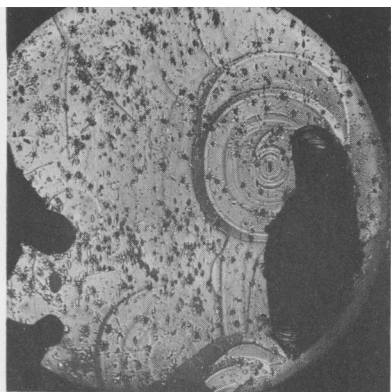


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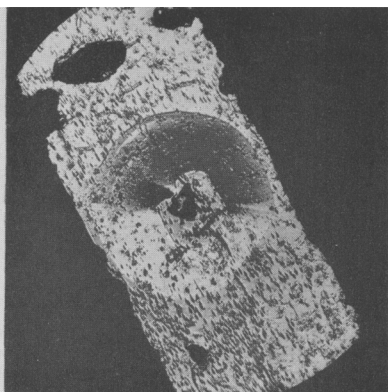


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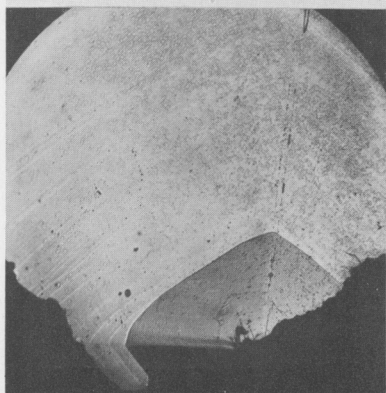
A. F. SEAGER: SURFACE STRUCTURE OF TOPAZ AND BERYL



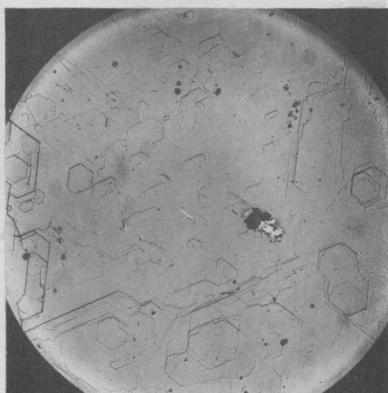
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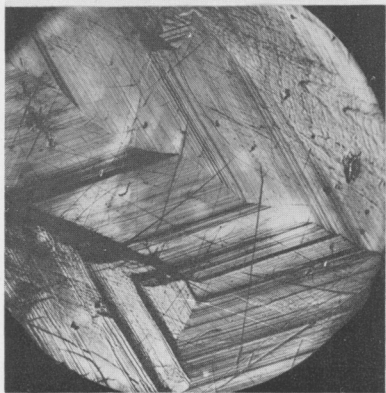
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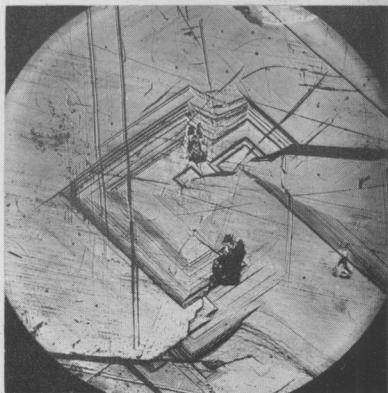
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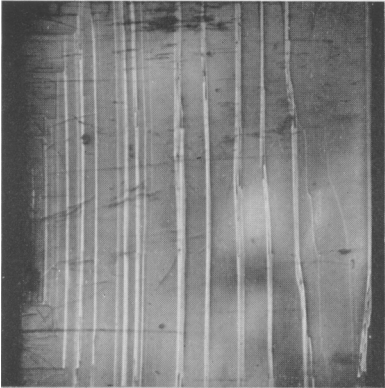
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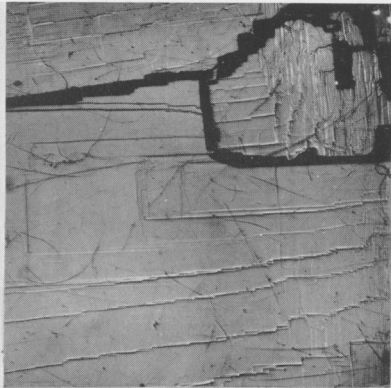
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A. F. SEAGER: SURFACE STRUCTURE OF BERYL AND BARYTE

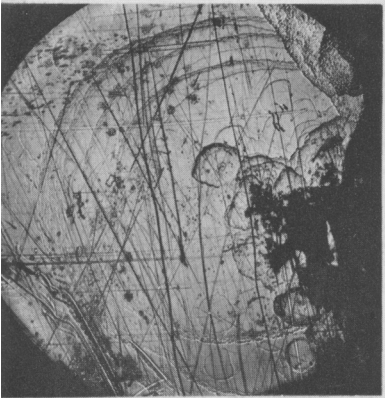




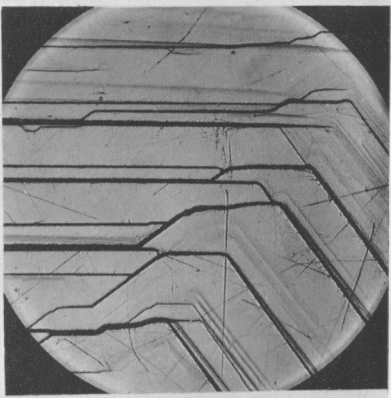
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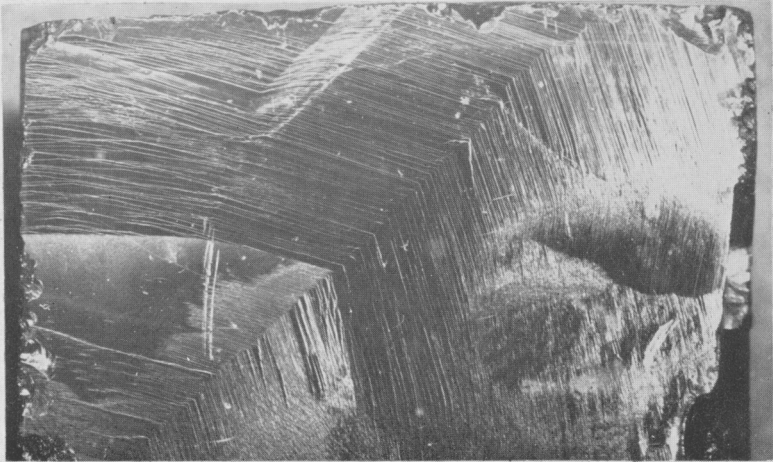
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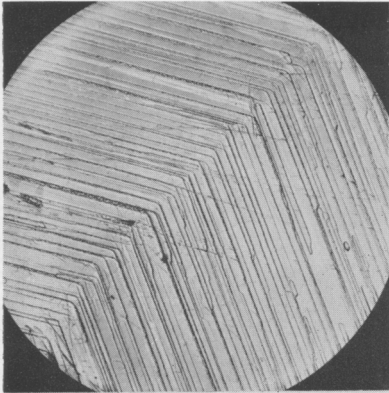
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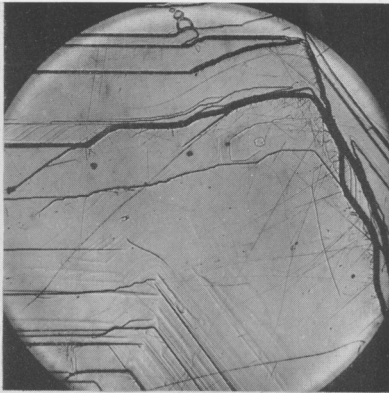
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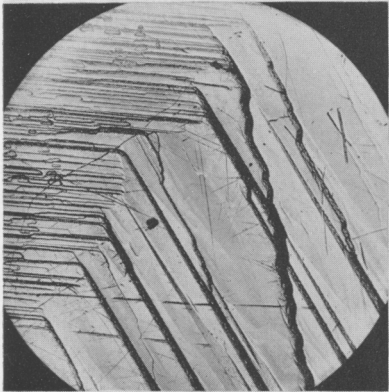
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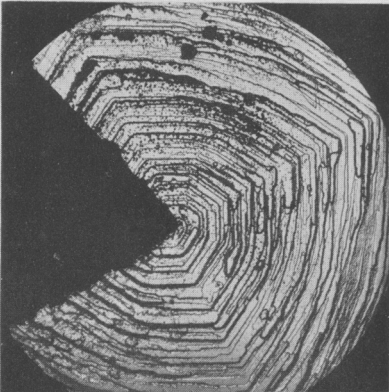
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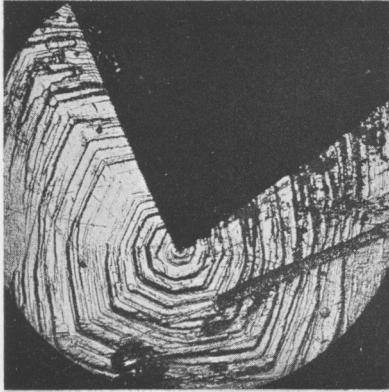
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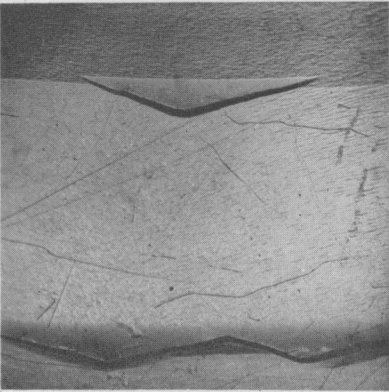
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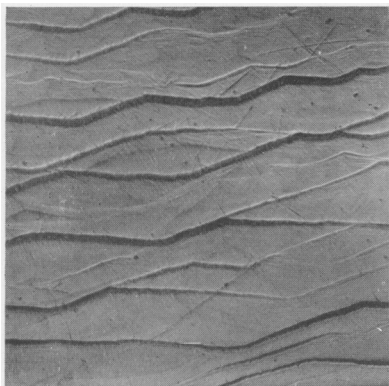
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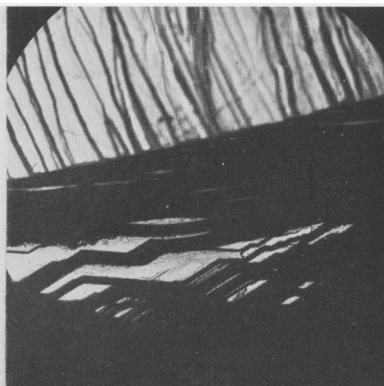
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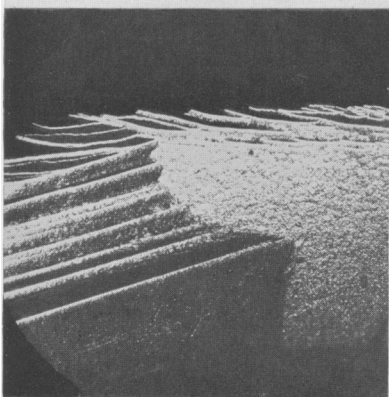
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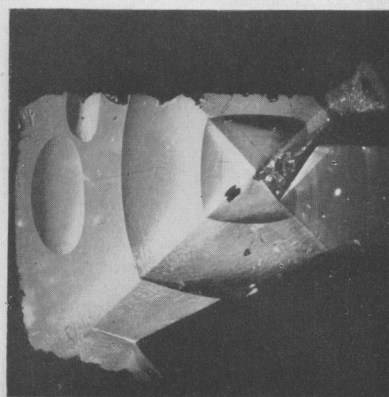
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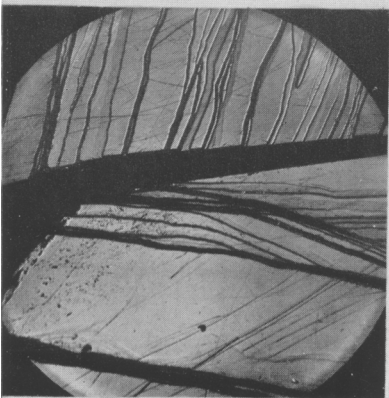
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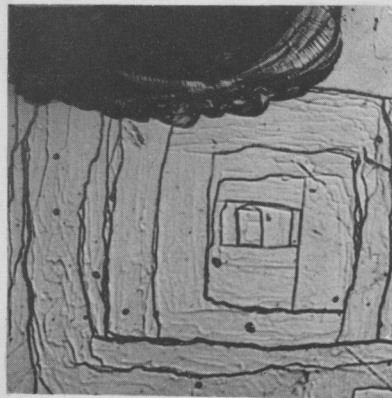
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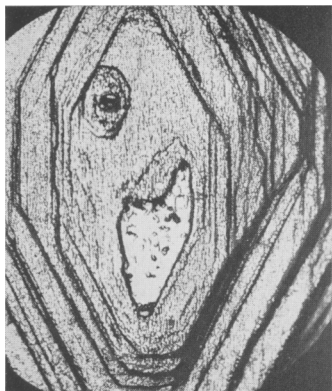
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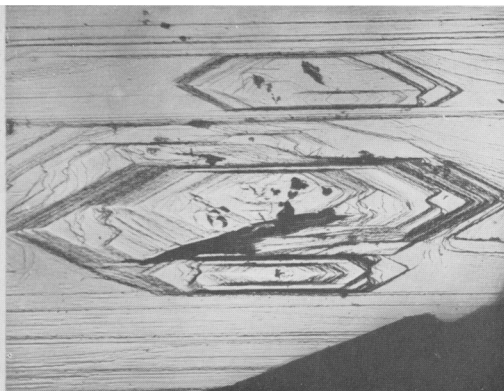
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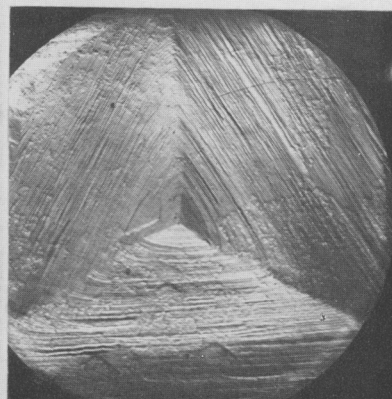
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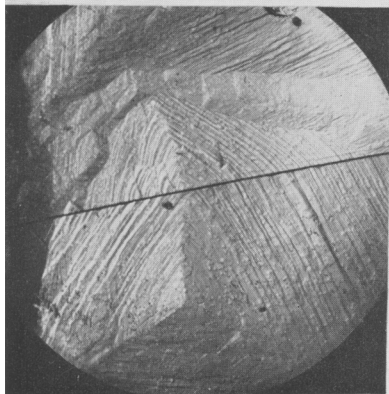
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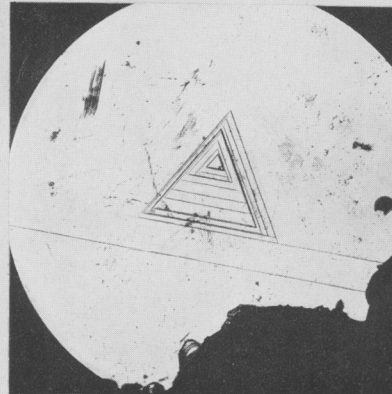
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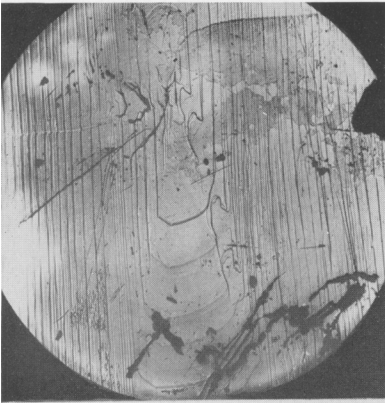
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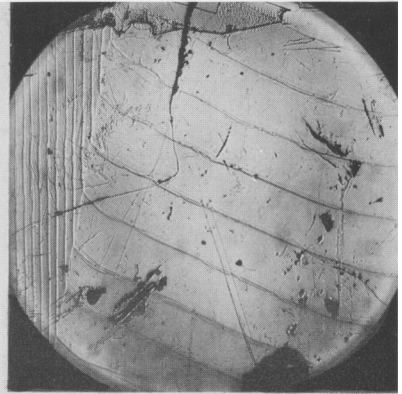
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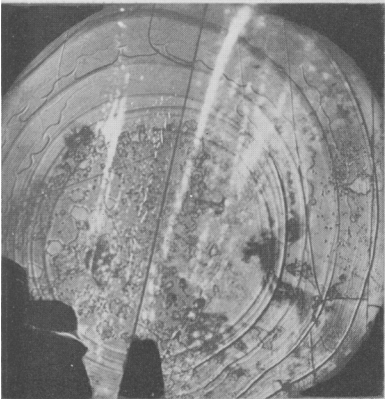
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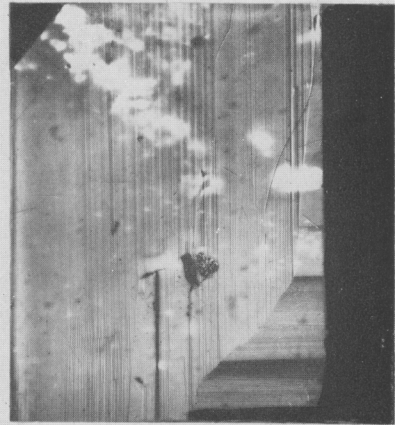
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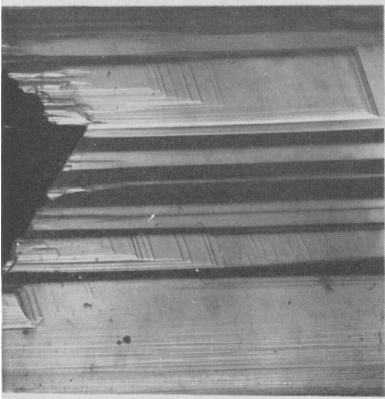
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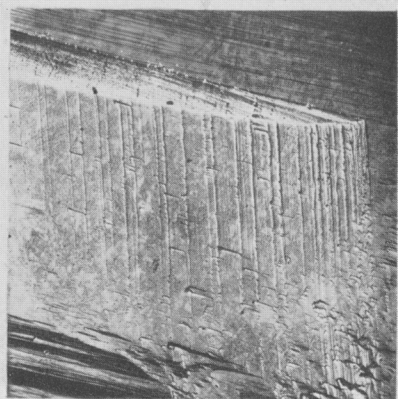
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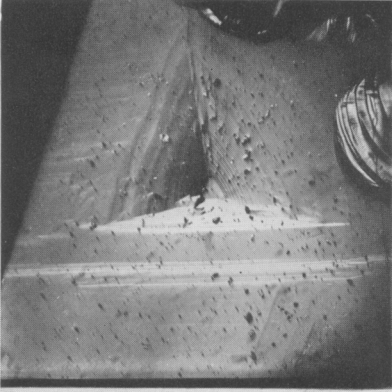


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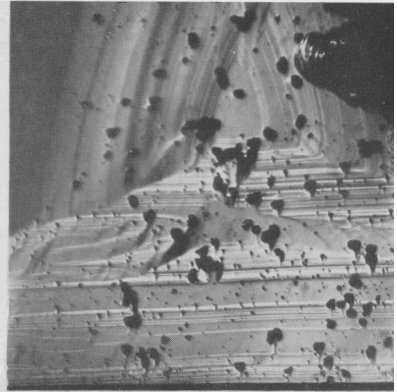


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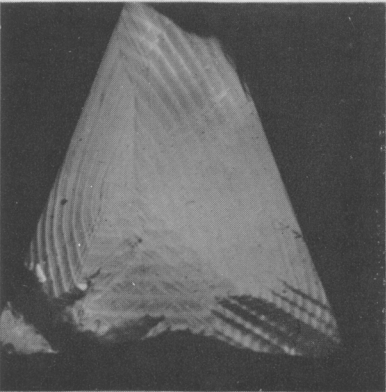




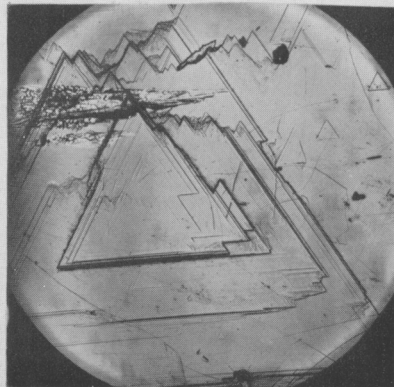
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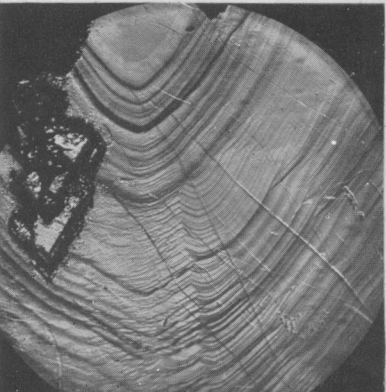
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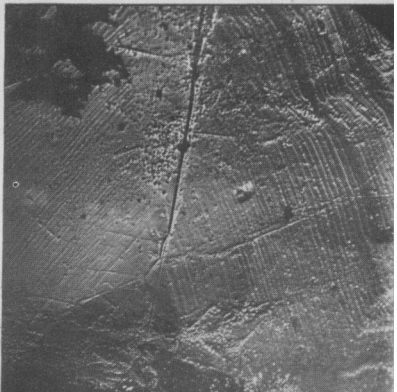
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51



52



53

A. F. SEAGER: SURFACE STRUCTURE OF QUARTZ, MAGNETITE, CELESTINE, AND TOURMALINE