

## TWINNING IN THE DIAMOND

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### ABSTRACT

The geometrical concept of twinning in the diamond is inadequate because a reflection across either (111) or  $(11\bar{2})$  will give identical relationships. The twinning plane can be identified by observing the contact or by demonstrating structurally that (111) lends itself to the growth phenomena involved in twinning. In either normal or twinned growth across (111) the first and second coordinations are identical and differences appear only in the third or higher coordinations. The statistical opportunity for twinned structures to be initiated is therefore high.

The probability of atoms falling into the normal or twinned position is not wholly a function of the structure. The surface energy distribution on a growing crystal face may be modified by the type of impurity that forms an adsorbed film on the surface. The resultant energy pattern may increase or decrease the statistical probability of the atoms attaching themselves in the normal or twinned position. By slowing the rate of migration of atoms to the surface the impurity film will promote the orderly accretion of atoms. Structural conditions indicate that diamonds do not grow by the accretion of single atoms.

Twinning is highly characteristic and what are apparently random intergrowths are more often multiple twins. The classical examples of contact, penetration, polysynthetic, and cyclic twins, by focussing attention on the geometrical symmetry of highly unusual and non-characteristic growth forms, obscure the true structural basis of twinning. Tables useful in the analysis of complex twins of non-symmetrical external form are given.

Crystallographic continuity exists across a twinning plane. All other boundaries between twinned individuals are suture contacts which are very often the loci of voids and inclusions.

### INTRODUCTION

Twinning is commonly defined in terms of a geometrical relationship which exists between the individuals comprising the twin. However, in the cubic system, a geometric concept alone is inadequate because more than one plane of reflection may describe the geometric relationship between two crystals which are twinned with respect to each other. This condition arises because of the equally spaced coordinate system.

If the diamond is assumed to have hexoctahedral symmetry, its twinning may be defined on a geometric basis as (1) a reflection across the rational plane (111) or, as (2) a reflection across the rational plane  $(11\bar{2})$ . These planes are mutually perpendicular, and both are also perpendicular to a secondary plane of symmetry. Hence, either plane will serve to define the relationship which exists between the twinned individuals (Fig. 1). There are three planes of the type  $(11\bar{2})$ , and in the idealized interpenetrating cubes (Fig. 2) after the spinel law (which are common in diamonds from the Belgian Congo) we have a total of four planes of symmetry, any one of which may be a twinning plane if such a plane is defined on a geometric basis only. Also, any of the four axes normal to these planes may be the twinning axis. This twinning may also be de-

fined geometrically as a rotation of  $70^{\circ} 32'$  about a binary axis of symmetry. This geometrical operation, however, does not meet the accepted concepts of a twinning law.

Twinning is not a geometric phenomenon but a deviation in the normal process of crystal growth. While this deviation gives rise to a discontinuity in the crystal structure, it is a controlled deviation—an ordered process—and therefore we have crystallographic continuity across the twinning plane from one individual to the other. The twinning plane is that plane along which the ordered deviation in the crystal structure

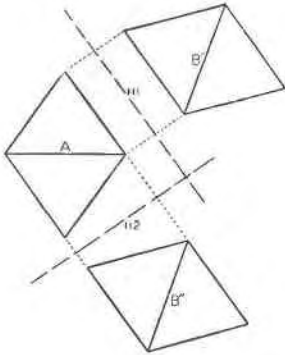


FIG. 1

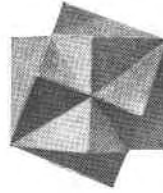


FIG. 2

takes place. In the diamond it is the (111) plane. In the microscopic sense twinning is typical and characteristic of the diamond. No single diamond that has been subjected to detailed examination has been found free from twinning of the mosaic type.

Much of the confusion in regard to twinning arises from the classical conception of twinning as a geometrical phenomenon. Given the complete goniometric measurements of a twinned diamond, it would be impossible to determine whether the twinning law was (111) or  $(11\bar{2})$ . The decision as to which plane is the twinning plane can be made by observing the twinning contact between the two individuals. This contact plane may generally be readily observed in contact twins, but in such substances as fluorite, which occurs only in "penetrations twins" (Fig. 2), no positive determination can be made except by observing the contact on an oriented polished surface. This condition arises because the crystal habit of fluorite is cubic while the twinning is octahedral. In doubly refracting crystals the two individuals can be easily differentiated, but in singly refractive substances the presence of twinning other than that revealed by gross macroscopic features often goes unobserved. Figure 3 shows fine suture-like twinning in the diamond which has been revealed

by its hardness variation. Twinning contacts in quartz are brought out by etching a polished surface, revealing the "electrical twinning," which is not differentiated by double refraction.

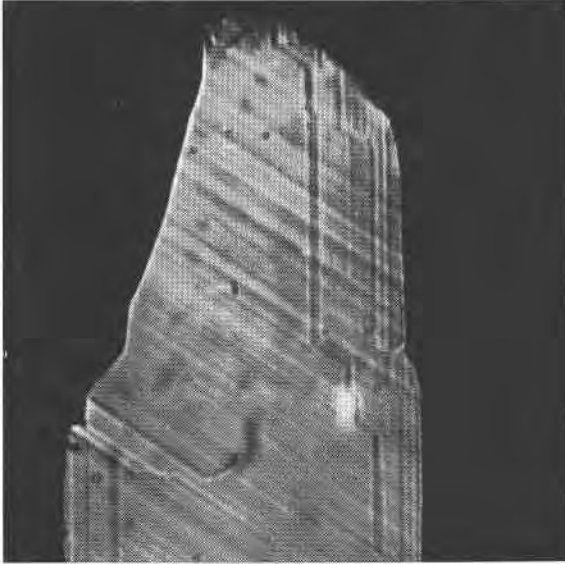


FIG. 3.

Polishing lines run lower right to upper left. Twinning lines vertical.

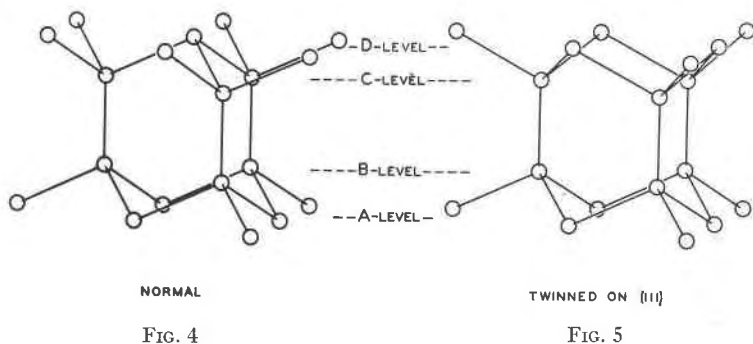
Where two geometrically possible twinning planes exist the choice can generally be made from structural considerations. One plane will have an energy distribution such that twinning across this face will be more probable than across the other.

#### STRUCTURAL TWINNING

In Figs. 4 and 5, four layers of atoms in the normal and the twinned position are represented with the octahedral face in the horizontal plane. In the twinned structure all primary bonds are completely satisfied and no free energy develops at the interface as a result of the relative positions of the *B* and *C* layers. The crystal is therefore continuous across the interface or twinning plane, although there is a discontinuity in the structure. The twinning plane is parallel to (111) but cannot be definitely localized, because the *B* and *C* levels of atoms may be considered as lying in either the upper or lower individual.

For purposes of the discussion, it is desirable to consider that the twinning plane is located between the *B* and *C* levels. Thus identified,

the adjacent octahedral surfaces of both individuals, normal and twinned, consist of carbon atoms arranged in equilateral triangular patterns held together by bonds normal to the surface. If any atom in the *C* level of the normal structure (Fig. 4) be taken as a center and the *C* and *D* levels be rotated sixty degrees, a structure identical to the twinned structure (Fig. 5) would be developed. Each atom of the third layer would still have its counterpart directly above it.



In the growth of a crystal by accretion on the octahedral surface a discrete aggregate of ordered atoms drifting toward the octahedral surface would have two positions, sixty degrees apart in which it could attach itself to the crystal. The distribution patterns of surface energy on the contacting surfaces determine which of the two positions, normal or twinned, would be chosen. The outermost layer of atoms on each surface are identical and would make identical contributions to the distribution patterns of surface energy. They would be dominant in forming this pattern. The layer of atoms next to the surface would produce a secondary trigonal distortion in the distribution pattern at the surface (Figs. 6 and 7).

Burger<sup>1</sup> has expressed a somewhat similar idea in stating that the higher coordination spheres control twinning. In the diamond structure it can be demonstrated that both the primary and secondary coordinations are identical in both the normal and the twinned positions, and that variation is introduced only in the tertiary coordinations. Its influence is so remote that it seems most unlikely that it could affect the structure. But in the case of surface energy distribution patterns the layer of atoms immediately below the surface would exercise a considerable influence upon the patterns.

In Figs. 6 and 7 an attempt has been made to represent schematically

<sup>1</sup> *Am. Mineral.*, **30**, 172 (1945)

the trigonal pattern of energy distribution produced by the surface and second layers of atoms on a crystal face. If a crystal grows by the addition of groups of atoms rather than single atoms the primary bonds of the surface layers will be equally as well satisfied by the normal or the twinned position. They will also exercise the dominant control in pulling the two surfaces together in one or the other of the two positions, which are sixty degrees apart. An aggregate of atoms drifting toward a crystal face in the halfway or thirty degree orientation must rotate to one or the other of the bonding positions. The polarity of the surface energy dis-

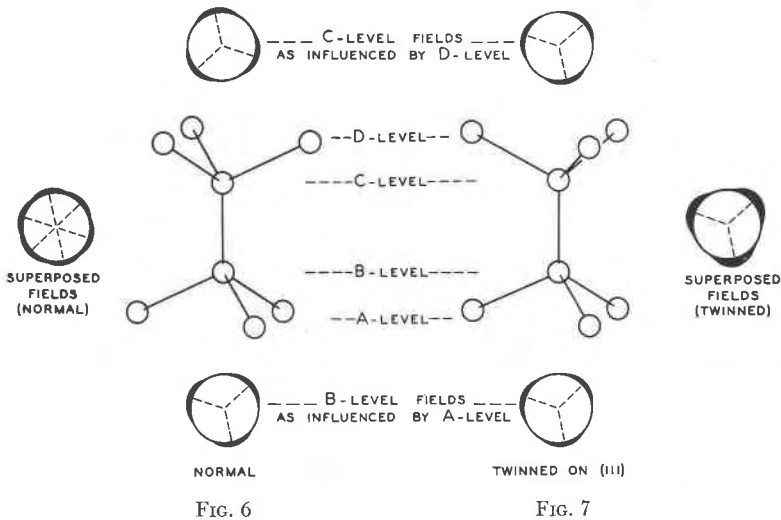


FIG. 6

FIG. 7

tributions as far as it is determined by the surface layers of atoms will be identical and would exercise no control over the attachment of drifting atoms in either the normal or twinned position. The control must come from the contribution to the surface pattern made by the second layer of atoms, and it might be assumed that if the approach was midway between the bonding positions, the aggregate would rotate to the normal position and attach itself to the crystal. There might be some intermediate position of approach where the drifting aggregate could take the lesser angular rotation toward the twinned position rather than the greater toward the normal bonding. On a structural basis alone one might assume that whether this neutral position is at  $15^{\circ}$ - $45^{\circ}$ ,  $10^{\circ}$ - $50^{\circ}$ , or some other point, would determine the statistical prevalence for the normal or the twinned position.

Such an argument as that just given assumes that the surface energy distribution of a crystal is a function of its structure only and does not

take into consideration the nature of the environment in which the crystal is growing. The unsatisfied bond energy at the surface of the crystal attracts and holds selectively molecules or atoms from the environment in which it exists. This film not only modifies the energy distribution of the surface but serves as a barrier through which the attaching atoms must migrate. A corresponding film on the surface of the arriving atoms exercises a similar control. The character of the film and consequently its influence will vary with the character of the environment. The choice then of whether the arriving atoms will join the nucleus in a normal or twinned position becomes dependent also upon the "reaction" that develops between the crystal face and the environment.

It appears to be necessary to have crystal growth on the octahedral surface of the diamond proceed not by the addition of a single layer of atoms, but at least by a double layer. From Figs. 4 and 5 it can be seen that if the *C* layer were added to the *B* layer, the resulting surface would have three times the free surface energy it had previously (three unsatisfied bonds per surface atom instead of one). By the addition of a double layer of *C* and *D* atoms the free surface energy remains the same. It appears therefore necessary to have crystal growth proceed by the accretion of two or multiples of two layers of atoms. This naturally holds true for both the normal and twinned positions.

#### TWINNING MORPHOLOGY

Because classical crystallography was founded on the external morphology of crystals, the concepts of contact and penetration twins developed. While the penetration twin may still be useful in describing some of the external features of twinning, and therefore assist in the recognition of minerals which are found characteristically twinned, it has no structural or theoretical significance. The spatial patterns of atoms cannot interpenetrate, and twinned crystals can only be in contact with each other. Interpenetrating twins of diamond are common only in the Belgian Congo production. These crystals are of the type shown in Fig. 2 and are usually yellow to brown in color and opaque. Occasional twins of this type are glassy, colorless to white, and may be as large as one hundred carats or more.

Gross manifestations of twinning represent exceptional cases where the two individuals have grown to approximately the same size. The De Beers Consolidated Mines, Ltd., give the percentages of macles (twins) of all stones recovered from the Dutoitspan, Bultfontein, and Wesselton Mines as 7.39, 5.25, and 3.71 per cent, respectively, or an average of 4.37 per cent. The percentages of "flats" (which are commonly twinned) from these mines are 9.05, 5.63, and 11.21, or an average of

8.63 per cent. Approximately 40% of the production consists of industrial stones which are not sorted into twinned and untwinned classifications. Macle is a classification of gem quality stones from which a fair recovery of polished stones can be made. They may be parted along the twinning plane by a process practically identical with cleaving. It is the impression of cleavers that they "cleave" along the twinning plane slightly easier than along a true cleavage, but that the resulting surfaces are apt to be more irregular. With such a high percentage of diamonds showing gross evidence of twinning, it is not surprising that microscopically all diamonds have internal twinned zones or areas.

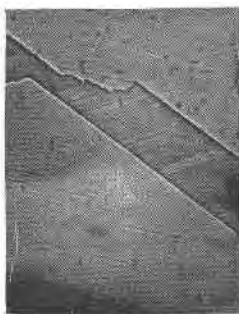


FIG. 8

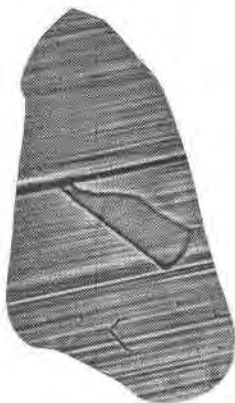


FIG. 9



FIG. 10

Polishing lines are nearly horizontal.

Figure 8 shows the polished surface of a diamond which has been lapped in such a direction that the directional variation in hardness brings into relief the twinned zone. This zone itself is further twinned. The orientation of the twin differs from that of the surrounding diamond, and when it is being lapped on a softer direction, the twinned zone presents a hard direction to the lap and resists abrasion. When viewed in the reflecting microscope, the beveled edge along the twinning contact stands out in relief, because it does not reflect the vertically incident light uniformly with the rest of the flat surface. This relief is accentuated when the microscope is slightly out of focus and appears to move parallel to the contact when the tube of the microscope is raised and lowered in a manner similar to the Becke line. When the tube is raised, the line moves towards the area in lower relief, in this case, away from the twinned zone.

The diamond cutting profession has developed a terminology to dis-

nate the various types of cross-grained structure (twinning) that causes difficulties in polishing a diamond. When the diamond cutter is lapping on the softer or more rapid cutting direction of the main part of the stone, the twinned area is presenting a hard direction, and vice versa. By choosing an intermediate direction, he finds one in which the two areas both cut at the same rate. If a third area is twinned in a direction not parallel to either of the other areas, it is impossible to remove all evidence of the twinning on a polished surface.

The penetration and contact twin of the mineralogist is called a *macle* by the diamond cutter. He designates as a *block* an enclosed twin that appears as a regularly shaped area on a polished surface (Fig. 9). In the early stages of the growth of the twin one individual has become dominant and completely encloses the other. A *knot* is an enclosed twin of irregular outline and often two or more individuals comprise the knotted area. A *ping* is a small speck-like area that stands out on the polished surface and from which a comet-like tail usually streams in the direction of lapping. It can be demonstrated that all these types are twinned on (111) by the trace of the twinning contact on an oriented polished surface or by measuring the angle between the trace and the lapping direction at which host and the twin abrade at equal rates. Contact twins rarely consist of only two individuals but commonly are polysynthetic, at least in the region where it is macroscopically evident that the twinning plane lies. Microscopically, the commonest types of twinning observed are the speck-like inclusions (*pings*) and paper-thin layers which on a polished surface appear as continuous (Fig. 10) or discontinuous (Fig. 3) lines parallel to traces of the octahedron. A single



FIG. 11

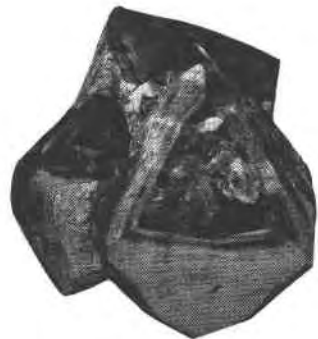


FIG. 12

crystal commonly reveals twinning on more than one of the octahedral faces. Complex twins such as that in Fig. 11, a seventy-five carat diamond consisting of four cubes, are classified in the *bort* category and



crushed into grit for grinding wheels. Much of the bort that at first glance appears to be a random intergrowth can be demonstrated to be complexly twinned.

Most of the literature on twinning deals with crystals of the lower symmetry classes. In the case of the plagioclase feldspars, repeated twinning on (010) gives rise to polysynthetic twins with a distinctive geometric pattern, because there is only one direction of growth which the twinned crystal may take. However, in rutile, which twins on (101), there are four structurally equivalent faces. In knee-shaped twins there is one plane of symmetry to the combination, and if subsequent twinning bends in the same direction and maintains the plane of symmetry, a cyclic twin develops. This is a very specialized case, but because of its unusual geometry, it has been considered characteristic. A zig-zag type of cyclic twinning which calls for a repetition of a distinctive pattern of alternation is also represented in museum specimens as a characteristic twinning. A more complex type of twinning gives rise to what has been called alphabetical rutile, because the bizarre arrangements often resemble letters of the alphabet. The finest museum specimens of this type represent repeated twinning in which all individuals lie in one plane. The more complex forms of twinned rutile in which twinning occurs randomly in three directions on any of the structurally equivalent (101) faces are usually dismissed as random intergrowths. The exceptional case where three or four successive twinings have followed the same pattern and the individuals have approximately the same size give rise to distinctive geometric forms which make excellent museum specimens but obscure the structural implications of twinning.

The complexity of twinning in the diamond with its still higher symmetry multiplies so rapidly that a twinned aggregate of more than three individuals nearly defies analysis. Figure 12 is a 14.60 carat gem quality diamond consisting of four octahedrons, three of which are apparent in the photograph. Because the three are not exactly equal in size and because they also partially interpenetrate, it is not at first evident that we are dealing with two octahedrons twinned on a central octahedron. The fourth octahedron, on the underside, is also part of the twinned complex.

Figure 13 illustrated a cyclic twin on (111) of three individuals. *A* and *C* answer all the geometrical conditions of twinning on (114), because it is a common rational plane to which both are symmetrical. If upon continued growth of the aggregate, *A* and *C* became dominant and enclose *B*, an intergrowth of the type shown in Fig. 14 will develop, which is a form observed among diamond intergrowths.

The possibilities of multiple twinning give rise to such diverse aggregates in the diamond that it is practically impossible to analyze them.

If the individuals of an aggregate are all of the same size, the geometrical symmetry may serve as a basis for analysis. Because the probability of equal size is so remote, the very existence of such highly specialized types leads to the conclusion that irregular twinned aggregates must be common. After examining hundreds of aggregates it does not seem unreasonable to state that most of the intergrowths of diamond commonly classed at random are multiple twins.

#### ANALYSIS OF TWINNED INTERGROWTHS

When dealing with a twinned intergrowth of three or more individuals, the methods of analytical geometry are much simpler to handle than

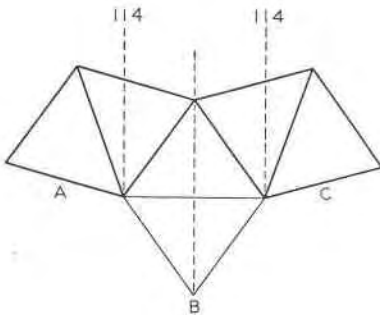


FIG. 13

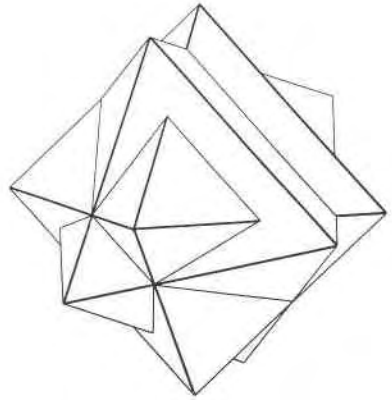


FIG. 14

trigonometric calculations involving interfacial angles. One works with linear equations in three unknowns with coefficients which are small whole numbers, and the algebraic processes are simpler than the complex computations using trigonometric functions which are not rational.

It is convenient to assume that all planes pass through the origin (Fig. 15) in a manner analogous to the procedures used with the stereographic projection where points are determined by normals to the faces drawn from the center of the reference sphere. If it is desired to find the indices of the new plane ( $tuv$ ) with respect to the old axes when a given plane ( $pqr$ ) is reflected through the twinning plane ( $hkl$ ), the equations of these three planes are written as:

$$\begin{aligned} px + qy + rz &= 0 \\ hx + ky + lz &= 0 \\ tx + uy + vz &= 0 \end{aligned}$$

By a procedure similar to that by which the reciprocal lattice is derived, the plane ( $pqr$ ) can be defined by the point  $p, q, r$  which will lie on the

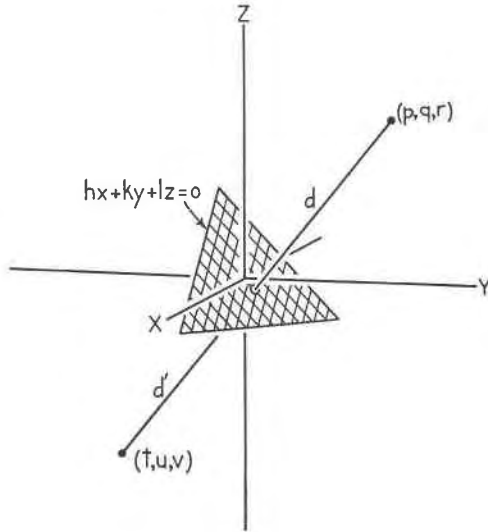


FIG. 15

normal to the plane from the origin. The reflection of that point in  $(hkl)$  gives rise to the point  $t, u, v$ , which lies on the normal to the plane  $(tuv)$  that passes through the origin. The distances of the points  $p, q, r$  and  $t, u, v$  from the plane  $hx + ky + lz = 0$  are:

$$d = \frac{hp + kq + lr}{\sqrt{h^2 + k^2 + l^2}} \quad d' = \frac{ht + ku + lv}{\sqrt{h^2 + k^2 + l^2}}$$

These distances are equal and opposite in sign, and therefore

$$(1) \quad hp + kq + lr = -ht - ku - lv.$$

If the point  $t, u, v$  is the reflection of  $p, q, r$ , then  $t, u, v$  lies on the line that passes through  $p, q, r$  and is normal to  $hx + ky + lz = 0$ . The equations of this line in symmetric form are:

$$\frac{x - p}{h} = \frac{y - q}{k} = \frac{z - r}{l}$$

from which we derive

$$kx - kp = hy - hq \quad \text{and} \quad lx - lp = hz - hr.$$

Because the point  $t, u, v$  lies on this line we may substitute  $t, u,$  and  $v$  for  $x, y,$  and  $z$  giving:

$$(2) \quad kt - kp = hu - hq \quad \text{and} \quad (3) \quad lt - lp = hv - hr.$$

Rearranging equations (1), (2), and (3) we have

$$\begin{aligned} ht + ku + lv &= -hp - kq - lr \\ kt - hu &= kp - hq \\ lt - hv &= lp - hr. \end{aligned}$$

By substituting the indices of a plane ( $pqr$ ) and the indices of the twinning plane ( $hkl$ ), we have three simultaneous equations which may be solved for  $t$ ,  $u$ , and  $v$ . These are actually coordinates of a point, but they also represent the plane ( $tuv$ ). Because ( $pqr$ ) and ( $hkl$ ) are rational ( $tuv$ ) is obviously also rational. In dealing with a twinned crystal, it is therefore possible to use a single set of crystallographic axes and all planes of both individuals will have rational indices on these axes. If a third individual is twinned with the second (Fig. 12), this twinning plane may be referred to the axes of the first and will have rational indices. Then following the procedure above all planes of the third individual can be given rational indices with respect to the axes of the first.

In general: *In any twinning complex of two or more individuals, the crystallographic axes of one individual may be taken as reference axes for all individuals and all planes of the twinned complex will have rational indices with respect to these axes.* In Table I, the forms resulting from twinning of the octahedron, the cube, and the dodecahedron planes on  $(111)$ ,  $(\bar{1}\bar{1}\bar{1})$ , and  $(1\bar{1}\bar{1})$  are given in the third column. Structurally these represent all the twinning possibilities, because  $(\bar{1}\bar{1}\bar{1})$ ,  $(1\bar{1}\bar{1})$ , and  $(1\bar{1}\bar{1})$  are equivalent planes. However, geometrically a completely twinned octahedron could be surrounded by eight octahedrons which structurally represent only four equivalent pairs. In this sense an octahedron twinned on  $(111)$  and  $(\bar{1}\bar{1}\bar{1})$  is identical structurally with one twinned on  $(111)$  and  $(1\bar{1}\bar{1})$ , but the physical appearance and geometry of the two groupings are different. In the first instance the two outer octahedrons would be symmetrical to  $(100)$  of the central octahedron and in the second symmetrical to  $(011)$ . This apparent anomaly arises only when one attempts to interpret twinning in terms of the geometry of the aggregates. It again emphasizes the fact that twinning is not a geometric process but a deviation in the process of crystal growth related to the structure of the crystal.

The results of a second twinning of the octahedron only are given in the fifth column of Table II. The rapid increases in the number of possible twinings demonstrates the degree of complexity which is introduced by multiple twinning. The continued appearance of certain integers in interchanged positions due to the high symmetry is readily apparent.

#### INCLUSIONS AND TWINNING

If a twinned crystal of diamond is included within another crystal (Figs. 8 and 9), the contacting surfaces will consist of parallel planes (the twinning planes) and vacillating juncture surfaces which, because of their irregular character, may be called suture contacts to differentiate them from twinning contacts. The twinning junctures may be offset as

TABLE I

ORIGINAL PLANE	TWINNING PLANE	FIRST TWINNING
100	111	122
	111	122
	111	122
	111	122
010	111	212
	111	212
	111	212
	111	212
001	111	221
	111	221
	111	221
	111	221
110	111	114
	111	110
	111	110
	111	114
101	111	141
	111	101
	111	141
	111	101
011	111	411
	111	411
	111	011
	111	011
110	111	110
	111	114
	111	114
	111	110
101	111	101
	111	141
	111	101
	111	141
011	111	011
	111	411
	111	011
	111	411

TABLE II

ORIGINAL PLANE	TWINNING PLANE	FIRST TWINNING	TWINNING PLANE	SECOND TWINNING
111	111	111	111	111
	111	511	511	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
111	111	511	511	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
111	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
111	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111
	111	111	111	111

in Fig. 8, but they still retain their parallelism, and their traces on a polished surface can be readily distinguished from the suture traces by their regularity. From the foregoing tables it can be seen that the non-twinning boundaries between the two individuals might consist of rational planes with unlike indices in contact with each other, but usually in the diamond the juncture is between step-like faces of either the octahedron or dodecahedron along which each individual is trying to maintain its dominant habit. They resemble closely the type of boundary that exists between two crystals of a random intergrowth. It is commonly the locus of inclusions and open spaces of microscopic size.

A corollary is that the presence of inclusions is indicative of twinning in the adjacent area. Diamond cutters have observed and commented on the fact that inclusions are apt to indicate an area of hardness. It has been postulated that strain in the vicinity of any inclusion has given the diamond a greater hardness at that point, but the logical answer is that the presence of twinning gives an area that presents a harder direction

of the diamond to the lap. The high correlation between inclusions and twinning indicates that the diamond cutters term *macle* for those diamonds which the mineralogist calls twins probably is a corruption of the French word *macle*, meaning spot or blemish.

In many crystals, for example quartz, internal phantom crystals are outlined by inclusions which lie on crystallographic faces representing an earlier growth level of the crystal. It is characteristic of the diamond that when it contains clouds of minute inclusions they do not lie along crystallographic directions but follow the irregular vacillating planes that characterize suture contacts between two twinned individuals.

#### CONCLUSIONS

The examination of large numbers of diamonds indicates that twinning is a dominant characteristic of the diamond. The variation in hardness with direction can be utilized to reveal the twinning which is not observed in polarized light, the common method by which microscopic twinning is observed. The structure of the diamond is such that twinning on (111) should be expected as a normal deviation in the growth process. Because of the high symmetry, twinning usually occurs on more than one of the octahedral faces at the same time. This gives rise to multiple twins of complex nature which have commonly been considered random intergrowths. Inclusions within the diamond are correlated with twinning.

#### ACKNOWLEDGMENTS

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