Proc. Indian Acad. Sci. **A24** 1–24 (1946)

The crystal forms of diamond and their significance

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Received for publication, June 4, 1946

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1. Introductory statement

The crystallography of diamond presents problems of peculiar interest and difficulty. The material as found is usually in the form of complete crystals bounded on all sides by their natural faces, but strangely enough, these faces generally exhibit a marked curvature. The diamonds found in the State of Panna in Central India, for example, are invariably of this kind. Other diamonds – as for example a group of specimens recently acquired for our studies from Hyderabad (Deccan) – show both plane and curved faces in combination. Even those diamonds which at first sight seem to resemble the standard forms of geometric crystallography, such as the rhombic dodecahedron or the octahedron, are found on scrutiny to exhibit features which preclude such an identification. This is the case, for example, with the South African diamonds presented to us for the purpose of these studies by the De Beers Mining Corporation of Kimberley. From these facts it is evident that the crystallography of diamond stands in a class by itself apart from that of other substances and needs to be approached from a distinctive standpoint. It is essential, at the very outset, to emphasise the point—seemingly obvious but often overlooked—that a crystal which exhibits curved faces cannot properly be described in the usual terminology which is based on the existence of plane faces obeying the crystallographic law of rational indices.

One of the most firmly established results of physics is the dependence of the physical properties of a crystalline solid on the symmetry of its structure of which the external form is an indication. There can be little doubt, therefore, that a study of the crystal forms of diamond, pursued from an appropriate standpoint, would prove most helpful in understanding and interpreting the many remarkable properties of this substance. These considerations and the availability of the material referred to above – some 72 specimens in all – encouraged us to undertake a critical examination of the subject. The investigation had for its object the discovery of the factors determining the general shape and other distinctive features of the crystal forms, and of the connection between them and the internal architecture of the crystal. The studies have enabled us to establish some definite propositions concerning these matters which are stated below. The evidence on which our conclusions are based will be set out fully in the course of the paper.

I. Both the internal architecture of diamond and its external form are determined by the quadrivalence of the carbon atom and its intrinsic tetrahedral symmetry in the crystal.
II. The sharply defined edges appearing on the curved surface of the crystal are its intersections with the six symmetry planes of the fundamental tetrahedron, each containing two of the valence directions.
III. The general shape of a diamond crystal stands in the closest relation with the configuration of the edges on its surface.
IV. An edge is most pronounced when it coincides exactly with a valence direction and becomes less conspicuous as it deviates from the same.
V. The vertices of the crystal are the points where four or six edges meet.
VI. The crystal symmetry of diamond as revealed by the edges on its surface is in the majority of cases that of the tetrahedral class only.
VII. While many diamonds exhibit the features characteristic of tetrahedral symmetry, there is a manifest tendency towards the assumption of forms which are common to the tetrahedral and octahedral symmetry classes.
VIII. The crystal forms of diamond exhibit a recognizable sequence on which it is possible to base a theory of their formation.

2. General descriptive characters

The Panna diamonds: Fortythree of our specimens are from the State of Panna in Central India. They are of widely differing shape, size and quality and may be considered as fairly representative of the diamonds mined in that area. As stated in the Introduction, the Panna diamonds invariably exhibit curved faces. During the senior author's two visits to the Panna State Treasury, he had the opportunity of examining several hundreds of these diamonds, including several very large and exceptionally fine specimens and never once came across a crystal showing plane faces either alone or in combination with the usual curved forms. It is very remarkable also that though the Panna diamonds are found in conglomerate beds of obviously sedimentary origin, it is exceptional to find a specimen exhibiting signs of having undergone any wear and tear during the transit from the original site of formation to its final resting place in those beds. Indeed, amongst our 43 specimens, there are only two which give any indication of having suffered in this manner. Most of our specimens, in fact, exhibit a remarkable transparency and smooth lustrous faces on which the details are seen beautifully clear and sharp. There cannot therefore be any doubt that the Panna diamonds exhibit precisely the same form as that in which they originally crystallised.

In an earlier symposium, a paper appeared by one of us (Ramaseshan 1944) describing and depicting the forms of the Panna diamonds. At that time, our Panna collection was not so extensive as it is now, having since been enriched by the addition of fourteen specimens of great interest from the scientific point of view. Further, at that time, we did not recognise as fully as we do now, the futility of describing curved crystal forms in the usual language of geometric crystallography. At that time also we had not discovered the physical significance of the details seen on the surface of these diamonds. The shortcomings arising from these circumstances, however, do not effect the scientific value of the diagrams, photographs and descriptive detail set out in the earlier paper. It was, in fact, the attempt to explain the facts described in that paper which led us to the present investigation.

We may here briefly recapitulate the main facts which emerged from the earlier studies. The curved surface of a Panna diamond is not a single continuous sheet, but consists of distinct sections meeting sharply along well-defined edges. These edges appear elevated above the general level of the surface to an extent depending on the angle between the sections on either side of them. This angle and the prominence of the edge vary enormously. An edge may at one part of the surface be so pronounced as to form a visible ridge, while elsewhere it may be so little conspicuous as to be seen only on careful examination under suitable illumination. The points on the surface where four or six prominent edges meet appear as protuberances or vertices of the crystal form. On the other hand, regions where the edges are inconspicuous are areas of relatively small curvature of the surface, even at points where they intersect. In a general way and subject to certain variations determined by the general shape of the diamond, the pattern of edges may be described by the statement that it divides the superficies of the crystal into 24 triangular sectors. These sectors are approximately similar to each other if the diamond is of fairly symmetric shape, while on the other hand, the sectors may differ greatly in size and shape if the diamond is of unsymmetrical form.

The Hyderabad diamonds: Eleven of the diamonds having their natural form as crystals included in our collection are a recent addition. They were picket out and purchased from the stock of unset stones in the possession of a firm of jewellers at Hyderabad (Deccan). No information was available regarding the origin of these stones beyond the statement that they had been detached for sale from some ancient jewellery. Since the city of Hyderabad is the nearest market to various places in the Deccan where diamonds are found, it is not improbable that the stones are of South Indian origin. All the eleven specimens are small, but they are of particular interest, being, with one exception, quite different from the Panna diamonds in their general features. They represent a combination of plane and curved forms, but the proportion of plane to curved surface varies in the different specimens. Taken together, the ten stones illustrate the successive stages of the transition from the curved faces and edges of the Panna diamonds to the form having eight plane faces separated by grooves which is the nearest approach made by diamond to the standard forms of geometric crystallography.

The South African diamonds: The sixteen specimens presented to us by De Beers of Kimberley for the purpose of this investigation have proved very useful in enabling us to compare the South African forms with the Indian ones and determine the relationships between them. Two items of particular interest in the collection may be mentioned here. One is a remarkably perfect example of the form of diamond first described by Haidinger, illustrations of which are to be found in the standard texts on mineralogy. The other is a triangular twin of flat tabular form with beautifully sculptured edges, presenting an interesting comparison with the rounded contours of the triangular twins found at Panna. We shall have occasion to refer to both of these specimens later in the course of the paper.

3. Some theoretical considerations

Geometric crystallography is based on the fact that crystals exhibit plane faces bounded by straight edges, and the descriptions given of them specify the directions of the face-normals with reference to the crystallographic axes. The obvious advantages of this system are that the directions of the face-normals are readily determined by goniometry, and that no changes are necessitated in the description by reason of any unequal development in different direction—a very common feature in actual crystals. When, however, we seek to depict a crystal by means of a figure, what we actually do is to delineate its edges. It follows that a crystal can be described by specifying the directions of its edges instead of its face-normals, and that such a description should enable us to determine the symmetry class to which the crystal belongs quite as definitely as the orientation of its faces. In this connection, however, a minor complication which may arise has to be borne in mind, namely, that the unequal development of a crystal in different directions would not only alter the lengths of its edges, but may also bring into existence new edges along which faces which do not meet in a perfectly developed crystal intersect each other.

When the faces of a crystal are curved, as in diamond, it becomes impossible to specify the directions of the face-normals by a finite set of numbers. But it remains possible to depict the form of the crystal exactly by delineating its edges. The edges would then naturally be curved, but it may very properly be assumed that if a crystal does exhibit a set of well-defined edges on its surface, the configuration of these edges must be related in some specifiable manner to its internal structure, and hence that a study of the same would enable us to determine the symmetry class of the crystal in an unequivocal manner.

X-ray studies have made it clear that the structure of diamond is essentially based on the quadrivalence of the carbon atom. The four axes of trigonal symmetry of the crystal are, in fact, also the directions of the valence bonds which link each atom of carbon in the structure with its four nearest neighbours. It stands to reason therefore that the visible signs of crystallinity exhibited by diamond in its natural forms should also be related in some simple manner to these valence directions. A specific indication as to the nature of such relationship is obtained by considering the form of the regular rhombic dodecahedron. It is readily proved that if a diamond had this form, every one of its 24 edges would coincide with one of the valence directions. Many actual diamonds do roughly resemble a rhombic dodecahedron, but they also exhibit features which cannot be reconciled with such a description. Nevertheless, a simple examination shows that the observable edges on such diamonds do approximately coincide with the valence directions. A more exact statement would be that the edges lie in the planes which contain the valence directions taken two at a time. This statement immediately makes intelligible the features observed on such diamonds which are irreconcilable with a description of them as rhombic dodecahedra.

We summarise the considerations set out above in the form of two propositions.

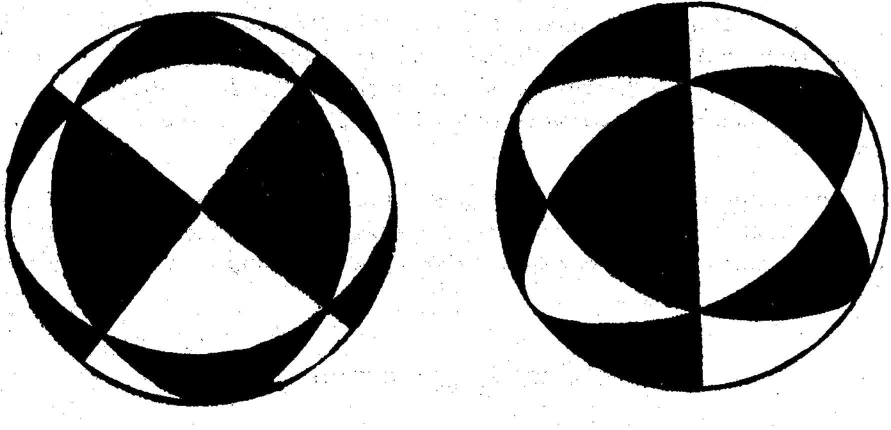
A. The configuration of the edges on the surface of a diamond is determined by the structure of the crystal and hence should exhibit its symmetry properties.
B. The configuration of the edges is also related in a simple way to the valence directions.

4. Geometric preliminaries

Before we proceed to discuss the observed forms of diamond in the light of the two foregoing propositions, it is useful to recall the symmetry properties of the crystal classes belonging to the cubic system. All five classes in that system have as a common feature the four axes of trigonal symmetry which are the cube body-diagonals. Taking these axes in pairs and drawing planes through them, we obtain the six diagonal planes of the cube. If these are symmetry planes, the crystal would belong to the tetrahedral class. All the elements of symmetry appearing in that class are represented by drawing through the centre of a sphere the six diagonal planes. The sphere then appears divided up into 24 equal spherical triangles.

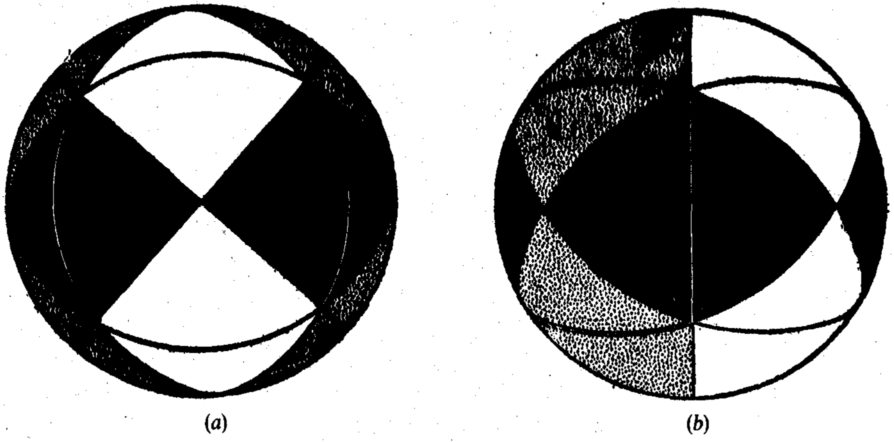
Figures 1(*a*) and (*b*) represent two views of a spherical surface divided up in this way. It will be seen that there are six points on the surface where four sectors meet and eight points where six sectors meet. There are respectively the intersections with the spherical surface of the three axes of diagonal symmetry and the four axes of trigonal symmetry. Any crystal of the tetrahedral class having a regular form, viz., a positive or a negative tetrahedron, a cube, a rhombic dodecahedron, a tetrakis-hexahedron or a hexakis-tetrahedron may be represented by its projection on the surface of a sphere. The edges of the crystal would appear as the sharp dividing lines between the areas on the surface of the sphere, the number of distinct areas being the same as the number of faces in the crystal, viz., 4, 6, 12 or 24 as the case may be. To illustrate this, the case of the rhombic dodecahedron is represented in figures 2(*a*) and (*b*). The shorter diagonals of the rhombic faces have been retained in the figures so as to enable the similarity between figures 1 and 2 to be perceived.

If, besides the six diagonal planes, the three axial planes of the cube are also planes of symmetry, the crystal would belong to the octahedral class. The elements of symmetry appearing in that class may be represented by drawing all the nine planes through the centre of a sphere. The surface of the latter would then appear divided up into 48 equal spherical triangles.



(*a*) (*b*)

Figures 1(*a*) and (*b*). Division of a spherical surface by the tetrahedral symmetry planes.


(*a*) (*b*)

Figures 2(*a*) and (*b*). Rhombic dodecahedron projected on a sphere.

Figure 3 illustrates the division of a spherical surface in this way by the symmetry planes of the octahedron. A regular crystal having the most general form of this class with 48 similar faces could be represented by its spherical projection, the edges of the crystal appearing as the dividing lines between the sectors of the sphere. Particular cases of the class with a smaller number of similar faces could also be represented in the same way by the simple device of leaving out some of the dividing lines on the surface and thereby reducing the number of distinct areas into which it appears divided. For instance, a regular octahedron would be represented by figure 3 with the six diagonal planes of symmetry omitted and only the three axial planes of symmetry retained, the surface of the sphere would then appear divided into 8 equal areas separated by sharp dividing lines. It would, of course, be impossible to exhibit the form of an octahedral

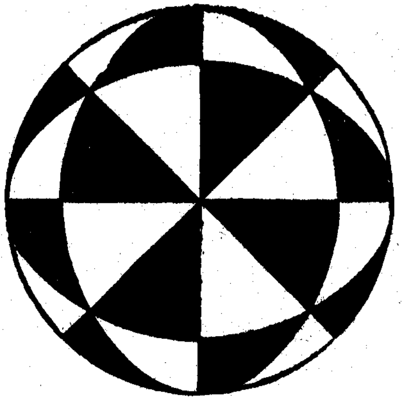


Figure 3. Division of spherical surface by the octahedral symmetry planes.

crystal with the aid of figure 1, since the three axial planes of symmetry are not present in that figure.

5. The configuration of the edges

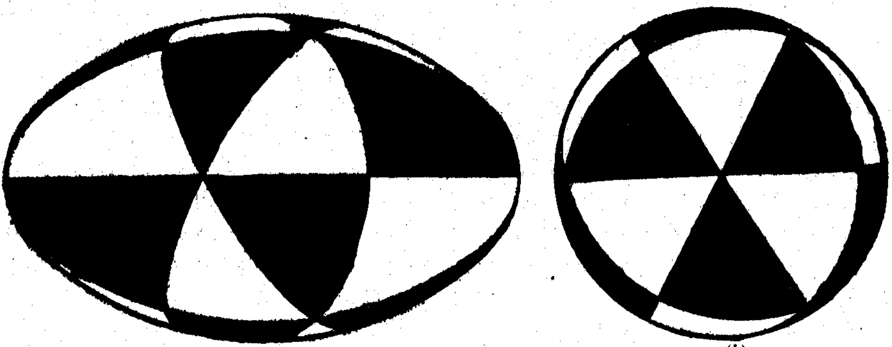
Even from the results of the earlier studies of the forms of the Panna diamonds referred to above (Ramaseshan, *loc. cit.*), it is evident that the edges seen on the surface of these diamonds represent the division of the superficies of the crystal into 24 sections by the symmetry planes of the fundamental tetrahedron. Many of the diamonds do indeed show marked deviations from the simplicity and regularity of the pattern depicted in figure 1. These deviations are however readily explained and do not represent any essential departure from the principles which determine the configuration of the pattern.

In the first place, the actual shape of the diamond has to be taken into consideration. The influence of this may be illustrated by considering the intersection of the symmetry planes of a tetrahedron with various surfaces other than a sphere, e.g., a prolate spheroid, an oblate spheroid or an ovaloid of revolution, which roughly represent the shape of the smaller Panna diamonds. In dealing with such cases, it is natural to suppose that the orientation of the tetrahedral axes with respect to the surface would not be arbitrary, but would be related to it in some specific fashion, viz., one of the trigonal axes of symmetry would coincide with the rotation axis of the surface. It is noteworthy that this view is borne out by the actual facts, viz., that the configuration of the edges on the surface of a diamond is very clearly related to the general shape of the crystal.

The division of the surface of a prolate spheroid into 24 sectors by the six diagonal planes of symmetry is shown in figure 4. Figure 4(*a*) is a side view and figure 4(*b*) is an end view. The sectors of the surface are still of roughly triangular form, but they are now of unequal area, those near the ends being considerably enlarged in relation to the others. A prolate spheroid has an axis of rotation and a plane of symmetry bisecting that axis. But the pattern on its surface evidently does not exhibit these features.

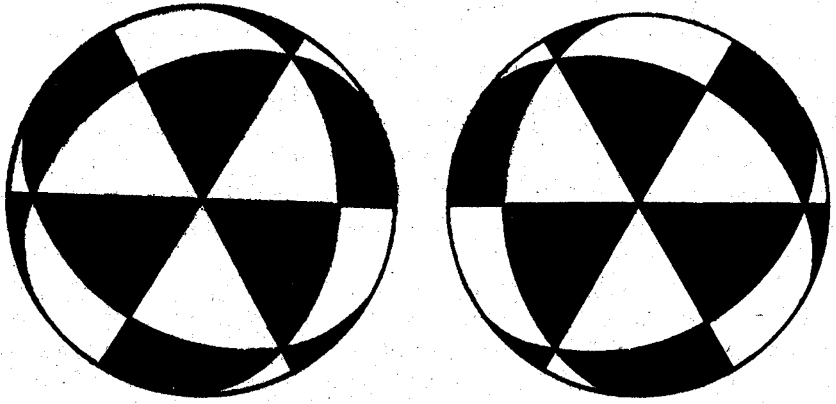
The division of the surface of an oblate spheroid into 24 segments by the six diagonal planes of symmetry is illustrated in figures 5(*a*) and (*b*), the two figures being the front and back views of the surface. It will be noticed that the front and back views are different and that one of them would require to be rotated through 180° to enable them to be brought into coincidence, thereby showing clearly that while the oblate spheroid has a plane of symmetry bisecting its axis of rotation, the pattern on its surface does not share that feature. Later in the paper, we shall have occasion to consider the features appearing in figures 5(*a*) and (*b*) in relation to the theory of formation of the flat triangular twins of diamond.

Figure 6 illustrates the division of the surface of an ovaloid of revolution into 24 segments by the symmetry planes of a tetrahedron. The figure of the ovaloid



(a) (b)

Figures 4(*a*) and (*b*). Division of a prolate spheroid by the tetrahedral symmetry planes.



(a) (b)

Figures 5(*a*) and (*b*). Division of an oblate spheroid by the symmetry planes of a tetrahedron.

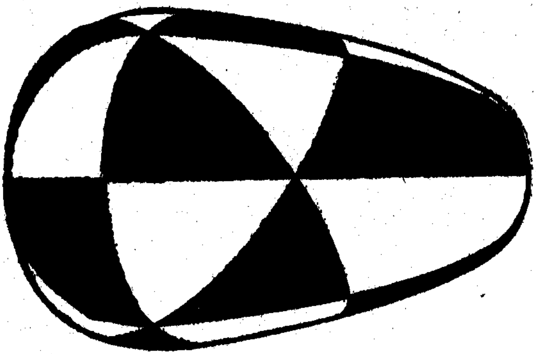


Figure 6. Division of an ovaloid of revolution by the symmetry planes of a tetrahedron.

does not possess a centre of symmetry and hence a pattern drawn on its surface exhibits the tetrahedral symmetry in a more obvious fashion than similar patterns on the surface of a sphere or spheroid of revolution.

In the models illustrated in figures 4, 5 and 6, the six diagonal planes of symmetry were drawn so as to pass through a single point within the surface, viz., the centre of the spheroid or the centre of mass of the ovaloid. As a consequence, the patterns are similar to that drawn on the surface of a sphere in their general features, viz., the division of the surface into 24 segments of triangular shape, four of which meet at six common points on the dyad axes and six at eight common points on the triad axes. The patterns are thus fundamentally related to the pattern of edges presented by a hexakis-tetrahedron or a tetrakis-hexahedron with 24 exactly similar faces. We know, however, that when the general shape of a crystal departs from regularity, the pattern of edges exhibited by it is substantially altered. While the *directions* of the edges which persist remain the same, their positions are altered, and new edges appear along the lines of intersection of the planes which did not previously meet. A similar situation would arise in our present problem of the configuration of the edges on a curved surface, and similar results would naturally follow. Hence, the configuration of the edges in the vicinity of the dyad and triad axes would be altered to an extent varying with the general shape of the diamond and to different extents at the various points. The nature of such variations may be readily deduced by shifting the edges laterally while retaining their general directions and drawing intermediate edges connecting the broken ends together.

This has been done in figure 7 for the case of four edges which fail to meet exactly on a dyad axis, with the result that a fresh edge connecting up the broken ends appears on the surface. It may be remarked that this type of irregularity is seldom noticed in diamond. The reason for this is that in the vicinity of the dyad axes, the valence directions lie in two perpendicular planes and the edges meeting on these axes are usually very pronounced.

On the other hand, the six edges which run towards a triad axis often fail to meet exactly on that axis when they traverse a part of the surface which is very nearly flat. The various types of deviation which may be expected to occur are

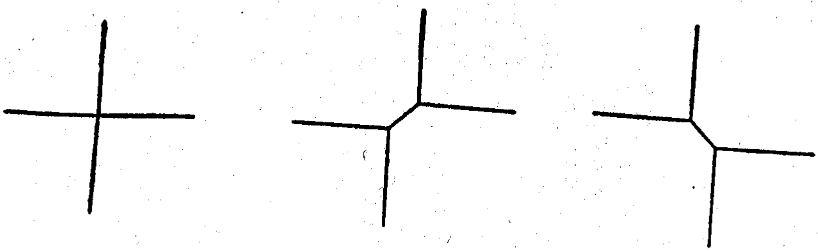


Figure 7. Pattern of edges in the vicinity of a dyad axis.

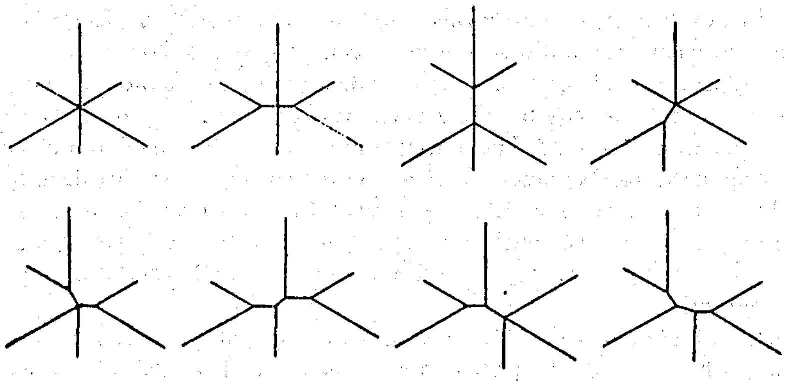


Figure 8. Pattern of edges in the vicinity of a triad axis.

indicated by the diagrams in figure 8. It may be remarked that they correspond closely to the features actually observed in our specimens.

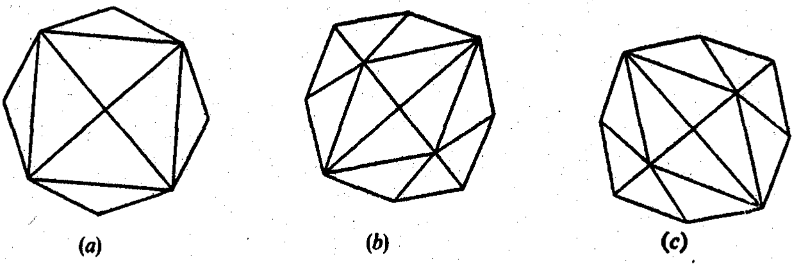
All the Panna diamonds without exception exhibit on their surface a pattern of edges which may be described as its intersections with the tetrahedral planes of symmetry of the structure—subject to the modifications described and illustrated above. This fact is all the more remarkable when it is recalled that some of the specimens in our collection bear no resemblance whatever to the conventional descriptions of a crystal. The actual configuration of the edges varies with the shape of the diamond and when this is irregular, the edges meander in their course. We shall refer to the points on the surface where four and six edges meet respectively as the dyad and triad vertices of the crystal. The actual shape of any particular specimen is closely connected with the configuration of the edges in the vicinity of the dyad and triad axes and the relative prominence of the two types of vertices. There are, of course, various possibilities, and many of them are illustrated by the specimens in our collection. We shall return to these matters later in the paper, but meanwhile we may turn to the fundamental question—What is the crystal symmetry of diamond?

6. The crystal symmetry of diamond

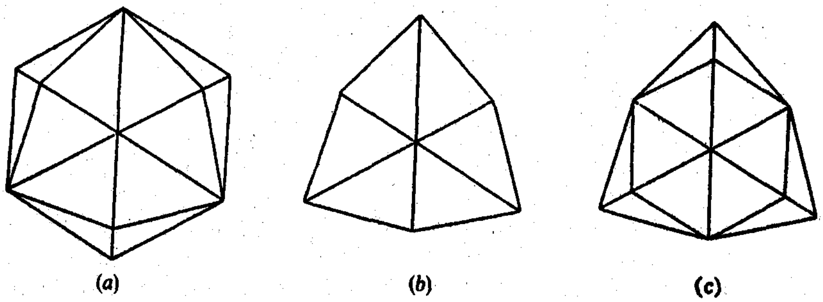
As illustrated in figure 3, the planes of symmetry in the octahedral class divide the sphere into 48 equal sectors. If diamond had octahedral symmetry of structure, we may expect it to exhibit such a subdivision, or at least the edges lying in the axial planes of symmetry. There is not the slightest hint or indication of any such edges in the Panna diamonds. Geometric crystallography tells us that the tetrahedral and octahedral symmetry classes have some forms in common, viz., the cube, the rhombic dodecahedron, and the tetrakis-hexahedron. If a substance crystallised exclusively in these forms, it would not be possible to decide between the two alternative possibilities. A decision can only be based on the appearance or non-appearance of forms definitely indicative of the higher or the lower symmetry as the case may be. We are accordingly justified in applying similar tests in the case of diamond. The non-appearance in the Panna diamonds of the edges lying in the axial symmetry planes is an indication that we are dealing with only the lower and not the higher symmetry. But evidence of a more positive character is desirable. We must however know what it is we have to look for. Here, again, we may usefully draw upon the ideas and results of geometric crystallography.

Figure 9(*a*) represents a tetrakis-hexahedron viewed along a dyad axis in either direction, while (*b*) and (*c*) represent a hexakis-tetrahedron also viewed along a dyad axis in the two opposite directions respectively.

Figure 10(*a*) represents a tetrakis-hexahedron viewed along a triad axis in either direction, while figures 10(*b*) and (*c*) represent a hexakis-tetrahedron


(*a*) (*b*) (*c*)

Figures 9(*a*), (*b*) and (*c*). (*a*) Tetrakis–hexahedron; (*b*) and (*c*) hexakis–tetrahedron viewed along a dyad axis.


(*a*) (*b*) (*c*)

Figures 10(*a*), (*b*) and (*c*). (*a*) Tetrakis–hexahedron; (*b*) and (*c*) hexakis–tetrahedron viewed along a triad axis.

viewed along a triad axis in the two opposite directions respectively. The forms illustrated in figures 9 and 10 both belong to the tetrahedral symmetry class, but the tetrakis-hexahedron can also be regarded as exhibiting octahedral symmetry, the dyad axis then becoming a tetrad axis passing through a centre of symmetry. As can be seen from a comparison of the figures, the characteristic feature of tetrahedral symmetry is that the dyad vertices appear as ridges instead of as peaks, while the triad vertices appear as peaks and as domes respectively at the opposite ends of each axis instead of as peaks at both ends.

Examination of our Panna collection discloses in numerous cases the specific features of tetrahedral symmetry indicated above. It is a very common occurrence to find the dyad vertices appearing as elongated ridges formed by the meeting of two edges nearly parallel to each other, while the two other edges which are transverse to them go up and down the slopes of the ridge. Then, again, one frequently finds the eight triad vertices falling clearly into two groups, one set of four forming fairly well-defined peaks, while the other set of four opposite to them appear as flattened domes. It is very significant also that such configurations of the dyad and triad vertices appear in association with each other. In other words, it is the same diamonds that either show or do not show the stated features in respect of the dyad and triad vertices. Further, these features are clearly related to the general shape of the diamond. The crystals that do not exhibit these features are of highly symmetrical form. On the other hand, the specimens that do exhibit these features possess a symmetry of general shape which is obviously of a lower order. Some of the finest diamonds in our Panna collection—beautiful water-white crystals with smooth lustrous faces—present an external form which does not possess a centre of symmetry. It is impossible in the face of these facts to doubt the truth of the proposition that *the internal symmetry of the diamond structure is that of the tetrahedral class only and not that of the octahedral*, at least in all the cases now under consideration.

It remains to be explained, however, why diamond seems to prefer the forms whose symmetry may be indifferently assigned either to the lower or the higher class, and why even in the forms in which the lower symmetry is observable, it does not appear as conspicuously as might have been expected. One possible explanation is that the more symmetric forms have a smaller surface per unit volume than the less symmetric ones. If, as the curvature of the surfaces suggests, diamond was formed by solidification from carbon liquefied under pressure, the form having the smaller surface would have less surface energy and is therefore more likely to be assumed at the instant of solidification. The second possible answer, which is supported by much relevant evidence, is that the positive and negative tetrahedral forms appear intermingled in the crystal. If this be the case, the crystal form would tend to imitate octahedral symmetry even if it does not exhibit the specific characters of that symmetry.

7. Classification of the crystal forms

A survey of all the specimens in our collection indicates that in spite of the great variety of forms exhibited by diamond, it is possible to arrange them in a regular sequence which is evidently connected with the physical circumstances of their formation. The ordering of the forms in such a sequence is obviously an advantage. It avoids the necessity for a minute description of all the individual specimens, and it also enables us, at least tentatively, to put forward a physical explanation of the features appearing in them. Indeed, we may advantageously reverse the order and derive from physical considerations an idea of what the forms of diamond should be and then proceed to fit the observed forms into a sequence based on such considerations.

It appears highly probable that diamond results from the solidification of carbon which has assumed the liquid state under conditions of high pressure and temperature. The state of the atoms in the liquid state is an important point needing consideration. The thermal agitation would certainly prevent a perfect ordering of the valence bonds within the liquid. Hence, it follows that the molten carbon would assume a rounded shape and this would be the more likely, the smaller the volume under consideration. Solidification is accompanied by a fixation of the valence bonds but not necessarily by any radical change of shape. On this basis, it is easy to understand why the crystals formed have curved faces. It may be remarked that the smaller diamonds in our Panna collection exhibit a highly marked curvature of the surface on which a pattern of edges appears as described earlier in the paper. The formation of these patterns is readily explained. At the surface of the molten carbon there would be some free valences which may attach themselves to the surrounding material. The valences not thus disposed of would link each carbon atom to its three nearest neighbours on the surface and hence would tend to align them with respect to its position in the valence directions. Accordingly, the first indication of the regular internal structure manifesting itself on the surface of the solidified material would be the formation of edges along the directions of the valence bonds, or in the planes containing them which are also the tetrahedral planes of symmetry of the structure.

If the edges of the crystal could align themselves completely along the valence directions, the surfaces between them would be plane, and the form of the crystal would be that of the rhombic dodecahedron. However, the curvature of the surfaces would prevent such a complete ordering of the edges. In consequence, the form would only approximate to a rhombic dodecahedron; the edges would not stop at the triad vertices but would be continued along the shorter diagonals, thereby dividing the superficies into 24 parts and not 12. Since these "continuation edges" deviate considerably from the valence directions, they would be relatively inconspicuous and would also meander on the surface to fit its varying curvature. These features are exhibited by several of our South African specimens. One of them (N.C. 26) is a beautiful crystal which might easily be mistaken for the regular rhombic dodecahedron of geometric crystallograhy, but is seen on a more careful examination to exhibit the features indicated above. The other dodecahedroid diamonds are less symmetrical in shape and exhibit corresponding variations in the configuration of their edges. These variations, however, are fully explicable on the same basis as in the case of the Panna diamonds already discussed in the foregoing pages.

If the valence bonds within the liquefied carbon have at least a semblance of the regular ordering which exists in the crystal, it would follow that the form assumed by the mass would deviate notably from a spherical shape. In a separate paper by Ramaseshan appearing in the present symposium, it is shown that the surface energy per unit area varies with the orientation of the surface in respect of the valence directions, being a minimum in the directions normal to the triad axes and a maximum in directions normal to the dyad axes. Accordingly, the liquid mass would tend to assume the shape of an octahedron with rounded edges, the largest proportion of the area appearing in the vicinity of the triad axes and the smallest near the dyad axes. On solidification, this general shape would be maintained but modified by the formation of the usual pattern of edges in the planes containing the valence bonds. Many of the larger Panna diamonds, including three examples in our collection (N.C. 2, N.C. 4, N.C. 8), have the shape indicated here. They may be referred to as "octahedroid" diamonds, but are not true octahedra, since they do not exhibit any edges in the axial planes. It may be remarked that the edges in the vicinity of the triad axes are much less conspicuous in the octahedroid diamonds than in the dodecahedroids, since they necessarily deviate more from the valence directions.

A further stage in the sequence of the crystal forms of diamond is reached when the influence of the thermal agitation is diminished sufficiently to enable the surface of the molten carbon to adjust itself exactly to the condition of minimum surface energy. This would exhibit itself by the surface in the immediate vicinity of the triad axes appearing as perfect planes in the solid crystal. The subsequent stages in the sequence would correspond to increasing areas of such plane areas in the crystal and a corresponding contraction of the curved surfaces, until finally a crystal form is attained in which nearly the whole surface consists of optical planes normal to the triad axes of symmetry.

The appearance of optically plane or "splendent" faces in combination with curved surfaces is represented in all its stages in our collection. The first indication of it to appear is a peculiar waviness or rippling of the surface in the vicinity of the triad vertices, often of a regular character and forming a hexagonal network of lines surrounding these vertices. The next stage in the sequence is the appearance of plane areas at and around these vertices. If these are continuous and of sufficient extent, they appear as a truncation or slicing off of the curved surface of the crystal and indeed have the form which would result from such a process, viz., a hexagon with three acute and three obtuse angles, the vertices appearing exactly at the points where the edges running across the curved surfaces meet the plane. Not infrequently also, the planes appear at the top of a succession of terraces. These terraces run parallel to the perimeter of the plane area, and the edges which have traversed the curved surfaces can be traced through the whole series of terraces up to the plane surface before they finally disappear. As the plane areas enlarge further in extent and the curved surfaces diminish correspondingly, the terraces or slopes—sometimes both terraces and slopes—fringing the plane areas persist, with the result that the crystal presents finally the appearance of an octahedron with deep grooves along its edges. The successive stages by which this result is reached can be followed in the Hyderabad specimens in our collection, thereby making it evident that it is not an accidental circumstance but a specific feature of the crystal forms of diamond.

8. The Haidinger diamond

As mentioned earlier, our South African collection includes a magnificent example (N.C. 25) of the particular form of diamond originally observed and figured by Haidinger and of which illustrations are to be found in the standard treatises on mineralogy. The general form is that of an octahedron, but the octahedral edges are absent. In their place, we have the special feature of the form, namely, conspicuous V-shaped grooves with smooth surfaces which widen from the middle outwards. Four of these grooves meet at each vertex of the octahedron and terminate in four sharp straight edges converging to a point on the symmetry axis. *These edges lie in the planes containing the valence bonds, and our measurements indicate that they are parallel to the valence directions.* They form part of the system of edges lying in the valence planes characteristic of all diamonds. Continuations of them, though much less conspicuous, may be traced running up the terraces on the faces of the octahedron and disappearing at the vertices with acute angles on each face. Another set of edges can be seen cutting across each groove and dividing it at its narrowest part into two parts sharply inclined to each other. These same edges can also be traced climbing the terraces and meeting the plane octahedral faces at the vertices with obtuse angles.

Apart from the fact that the particular specimen is a remarkably perfect one, the features which it exhibits can also be seen in several of our Hyderabad diamonds. It is therefore clear that the Haidinger diamond is not a rare or accidental occurrence but is a typical form of deep significance in the crystallography of this substance. The appearance of grooves or re-entrant edges is a characteristic feature of twinning, and in view of the independent evidence showing that the crystal symmetry of diamond is ordinarily that of the tetrahedral class, the only reasonable description of the Haidinger form is that it is an interpenetration twin of positive and negative hexakis-tetrahedra truncated by planes normal to the trigonal axes. Further, since the form mimics octahedral symmetry, it furnishes an excellent example of the result of such interpenetration in suppressing the external manifestation of the inherent tetrahedral symmetry of diamond.

9. The triangular twins

The ideas developed in the preceding pages enable us to offer a simple explanation of the formation and peculiar shape of the well known "macles" or twin diamonds of triangular form and of small thickness, examples of which are often forthcoming. Referring again to figures 5(*a*) and (*b*) on an earlier page in which the front and back views of an oblate spheroidal model were represented, the remark may be recalled that for these two views to become identical, it would be necessary to rotate one through 180° with respect to the other. In the figures, the pattern of edges is represented as lying on the surface of the model. Actually, however, the dyad vertices would appear elevated above the surface, while the triad vertices would tend to be suppressed and become relatively inconspicuous, as is indeed the case in the majority of diamonds for reasons already explained. Hence, an oblate spheroidal diamond would tend to assume a triangular shape having its vertices on the dyad axes of the pattern, but the triangles on the two faces would be set oppositely, viz., vertex to base and base to vertex. Indeed, some of the specimens in our Panna collection (N.C. 6, N.C. 12) show such a form. The thinner the diamond, however, the greater would be the probability that this incompatibility between the front and the back of the same crystal would be redressed during its formation by one half of the form swinging round through 180° with respect to the other, thereby resulting in the formation of a triangular twin in which the two halves fit each other perfectly, vertex to vertex and base to base.

The pattern of edges formed by the intersections of the tetrahedral symmetry planes with the two surfaces of a model twin of triangular form is represented in figure 11. Only one side of the model is shown, since the other would be identical in the twin. Examination of the triangular twins in our Panna collection (N.C. 9 and N.C. 23) reveals a pattern of edges on their faces and their edges which corresponds closely with that represented in figure 11.

One of the South African specimens in our collection (N.C. 30) is also a triangular twin, but of a different type. It is much thicker, and also much more "like a crystal", that is to say the faces are much flatter and the edges much steeper than in the Panna examples. Examination of this specimen reveals the remarkable fact that its two faces and also the edges up to a third of the way down from each face exhibit a close similarity to the Haidinger form of diamond. We have the same triangular faces with terraces, the same steep grooves below them and the same set of four sharp edges in the valence planes meeting near the vertices of the form. These features are incompatible with the usual description of

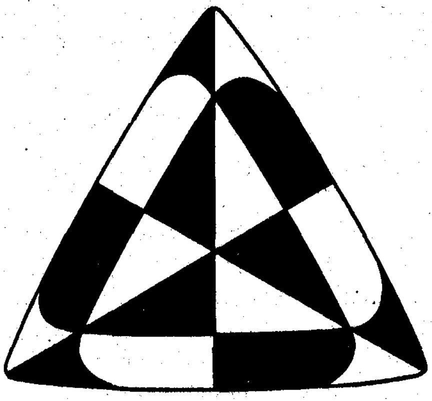


Figure 11. Intersection of the tetrahedral symmetry planes with the surfaces of a triangular twin.

the triangular diamonds as "spinel twins" obtained by cutting an octahedron in two and rotating one half through 180°. On the other hand, the specimen does exhibit in the vicinity of its median plane, the characteristic features of the form obtained in that way. It may accordingly be described as a remarkable but only partially successful attempt by a crystal of the tetrahedral class to mimic one of the characteristic form of octahedral symmetry.

10. Some descriptive notes

In the paper by Ramaseshan in the 1944 symposium, descriptions, drawings and photographs have already been given of several of our Panna diamonds. There is little need for reproducing the same of similar material here, especially as the numerous figures in the text of the present paper represent the theoretical counterparts of the features described in the earlier one. Since, however, several additions have been made to our collection, some of which are of special interest, it appears desirable to include descriptive notes and illustrations of a few of the new acquisitions.

N.C. 14—This diamond from Maharajpur (Panna) weighs 143 milligrams, and is water-white in colour, but exhibits a faint greenish surface tinge of the kind which is fairly common in the Panna diamonds. This is the specimen in our collection which makes the nearest approach in its shape to the regular hexakis-tetrahedron of geometric crystallography and hence most clearly demonstrates that diamond is a crystal of the tetrahedral symmetry class. The form is obviously lacking in a centre of symmetry. The eight triad vertices of the form fall into two groups of four each; in one group of four vertices, the edges meet accurately, and on the ridges connecting these vertices appear the six dyad vertices of the form. The other triad

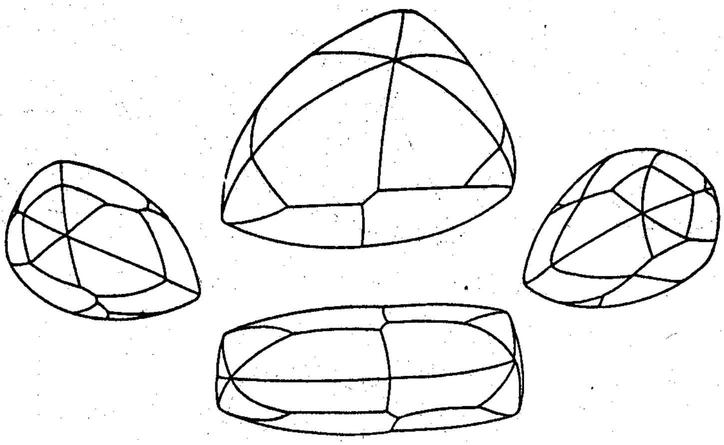


Figure 12. Front and side views of N.C. 14.

vertices appear on the four nearly flat faces of the form. In these vertices, the edges fail to meet and give rise to a pattern of broken lines as illustrated in figure 8 on an earlier page. Two of the triad vertices of the former group appear as pointed triangular tips at which three prominent ridges and three fainter ones midway between them meet. These tips are connected by an elongated ridge. The two other triad vertices of the same group are less prominent and are connected by a much shorter ridge, and the dyad vertex appearing midway between them is a prominent feature of the form. As a consequence of these features, both the general shape of the diamond and the details observed on it show only two of the planes of symmetry of the six which a perfect tetrahedron has. The two planes of symmetry are mutually perpendicular and contain between them all the eight triad vertices—four on each. Their intersection is a dyad axis of symmetry for the crystal.

N.C. 18—This diamond from Udasna (Panna) weighs 57 milligrams and is perfectly water-white in colour. Being a relatively small diamond, the curvature of the faces is very marked. Nevertheless, the specimen shows clearly enough the characters of tetrahedral symmetry. The form lacks a centre of symmetry, and exhibits a trigonal axis at the two ends of which very different features are noticed. One end of the axis is a sharp triangular tip where six edges meet. The other end is a nearly flat dome with a hexagonal perimeter bounded by three dyad and three triad vertices, all of which are fairly prominent. The four other triad vertices—appearing respectively on the triangular faces of the tip and on the dome at its end—are of a different description, being zig-zag patterns of broken lines. Three of the dyad vertices appear on the ridges which converge towards the triangular tip of the form, and the three others around the base of the dome.

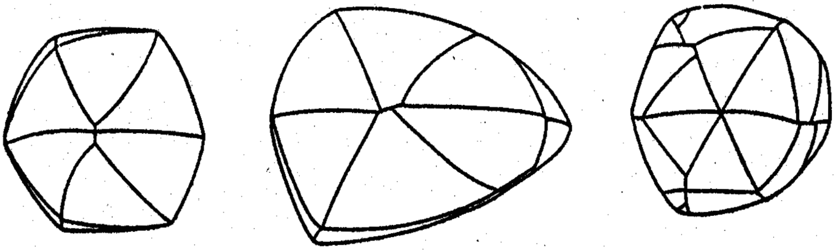


Figure 13. Front and end views of N.C. 18.

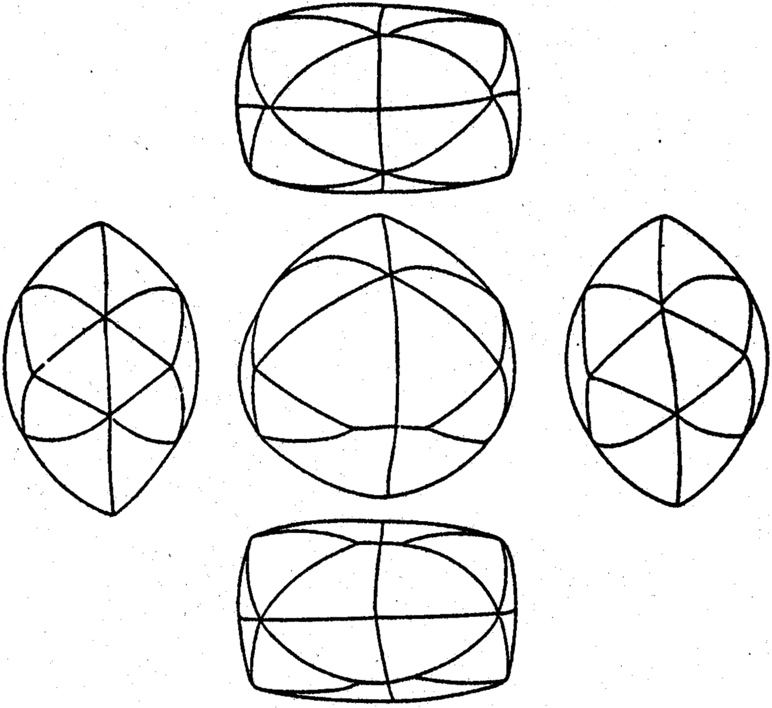


Figure 14. Front and side views of N.C. 10.

N.C. 10—This beautiful diamond from Maharajpur (Panna) weighing 182 milligrams, water-white in colour, with smooth lustrous faces, is well worthy of a detailed description as it illustrates a stage in the transition from the "tetrahedroid" to the "octahedroid" forms of diamond. The crystal exhibits a perfect symmetry of shape and of detail about two planes which are perpendicular to each other. These planes are two of the tetrahedral symmetry planes and the line along which they intersect is a dyad axis also for the actual shape of the crystal. The two ends of this axis are prominent dyad vertices of the form and exhibit the tetrahedroid character, appearing as ridges rather than as peaks. Since one of them is much more pronounced than the other, the crystal form has no centre of symmetry. The four other dyad vertices come next in order of prominence and are all exactly alike each other. They are peaks rather than ridges and thus exhibit an octahedroid character. The triad vertices form pairs, two of which appear in each of the two symmetry planes of the crystal. The four pairs of vertices are of progressively diminishing prominence. They illustrate the successive stages of the transformation of a triad vertex from a perfect meeting of six ridges to an elongated zig-zag of broken lines.

N.C. 25—This diamond from Bultfontein, South Africa, water-white in colour and weighing 211 milligrams, is the Haidinger form already described in some detail in section 8 above. A photograh with an accompanying sketch appear in figures 1 and 2 in plate I of this paper. Figure 10 in plate III is a photograph of one of the faces of the form showing its characteristic hexagonal shape, the terraces surrounding it and the triangular depressions or "trigons" appearing on the same. Another photograph of the same diamond reproduced as a negative appears as figure 11 in plate III.

N.C. 164—This is one of the Hyderabad diamonds whose forms were described and explained in sections 2 and 7 above. It is a small diamond weighing 62 milligrams and is nearly water-white in colour. It exhibits a combination of optically plane or "splendent" faces with curved surfaces separating them, the latter being smooth and lustrous and exhibiting edges analogous to those observed on the Panna diamonds. In this particular diamond, six of the plane faces are much larger than the other two. The photograph reproduced as figure 3 in plate I shows the smallest of all the plane faces as a dark area. Figure 9 in plate III is an enlarged picture of the same face, showing clearly the curved surfaces surrounding it as well as the edges crossing them and meeting the vertices of the plane face. The sketch reproduced in figure 4 in plate I shows in addition two of the larger plane faces of the diamond and the curved surface lying between them and separated into sections by a system of curvilinear edges. See also figure 12 in plate III where the photograph is reproduced as a negative.

N.C. 26—This diamond from South-West Africa, weighing 191 milligrams and water-white in colour, is the "rhombic dodecahedron" the features observed on which have been described and explained in section 7 above. Photographs and sketches of it from two different points of view are reproduced as figures 5, 6, 7 and 8 in plate II and in figures 13 and 14 in plate III.

11. The allotropic modifications of diamond

It is useful here to review our findings in so far as they have a bearing on the question of the symmetry of the internal structure of diamond. What the evidence indicates is that in the majority of the cases studied and possibly in all, the crystal symmetry is that of the tetrahedral class only and that none of the crystals appearing in our collection presents conclusive evidence of its possessing a true octahedral symmetry. There is, however, a pronounced tendency towards the assumption of crystal forms which may be .indifferently regarded as either tetrahedral or octahedral. This tendency would result from an interpenetration of positive and negative tetrahedral forms, and there is crystallographic evidence that such interpenetration does occur. But it should not be forgotten that the same situation would arise from the existence of diamond having a truly octahedral symmetry of structure but formed under conditions unfavourable for the intrinsic symmetry expressing itself to the fullest extent in the external form. The possibility has also to be borne in mind that the modifications of diamond having tetrahedral and octahedral symmetry of structure appear intertwinned with each other in the same crystal. In all such cases, we could scarcely expect the crystal forms to exhibit either tetrahedral or octahedral symmetry exclusively. It would then be necessary to rely on physical evidence, as for instance the infra-red absorption spectrum, to discriminate between the various possibilities and to establish the nature of any particular specimen.

Summing up the situation, we may say that while the study of the crystal forms in our collection shows clearly enough that the majority of diamonds have only a tetrahedral symmetry of structure, the results do not exclude the possibility that diamond has in some cases a truly octahedral symmetry of structure which for one reason or another fails to manifest itself fully in the external form of the crystal.

12. Summary

The paper describes the conclusions reached from a critical examination of some 72 diamonds in their natural form obtained from various sources. The curvature of the faces and other special features exhibited by diamond invalidate a description of its forms in the standard terminology of geometric crystallography. The proper basis for description and classification is furnished by the configuration of the sharply-defined edges which appear dividing the superficies of the crystal into 24 distinct sections. Subject to minor modifications, these edges lie along the intersections of the surface with the symmetry planes of a fundamental tetrahedron. These planes also contain the directions of the valence bonds between the carbon atoms in the crystal and the sharpness of the edges is determined by the angle which they make with the valence directions. The

configuration of the edges and the specific features exhibited by them in numerous specimens prove that in the majority of diamonds the crystal symmetry is that of the tetrahedral class only. The crystallographic evidence also shows that the positive and negative tetrahedral forms freely interpenetrate each other, and this explains the frequent appearance of forms common to the tetrahedral and octahedral symmetry classes. That diamond may in some cases possess a true octahedral symmetry is however entirely consistent with the observed facts. A physical theory of the formation of diamond is outlined which explains the observed features of the crystal forms and enables them to be classified in a regular sequence.

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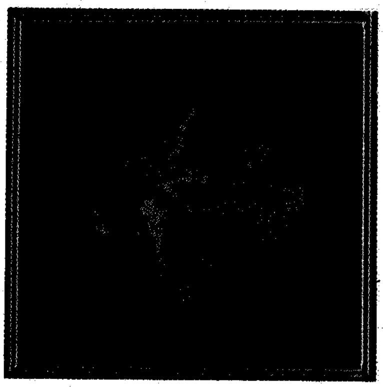


Figure 1

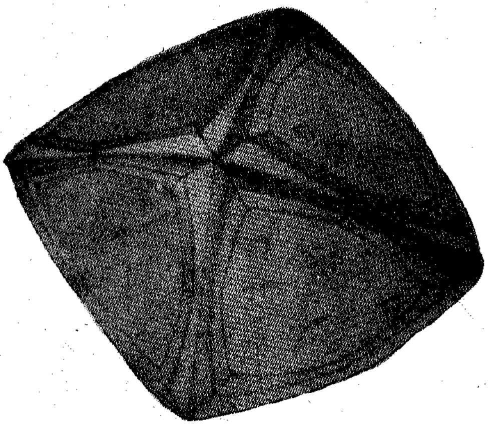


Figure 2

Haidinger diamond (N.C. 25)

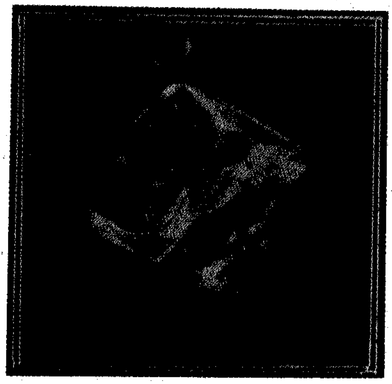


Figure 3

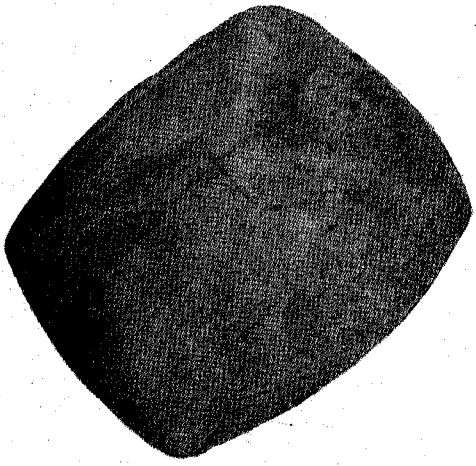


Figure 4

Hyderabad diamond (N.C. 164)

Plate I

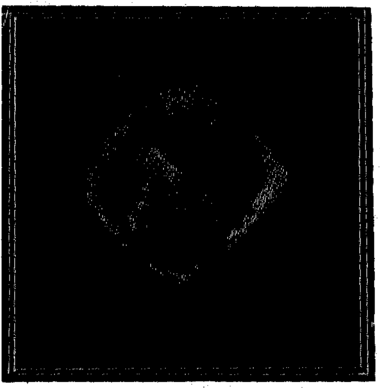


Figure 5

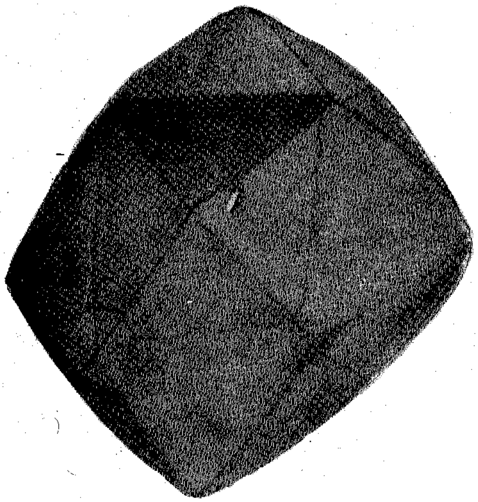


Figure 6

N.C. 26

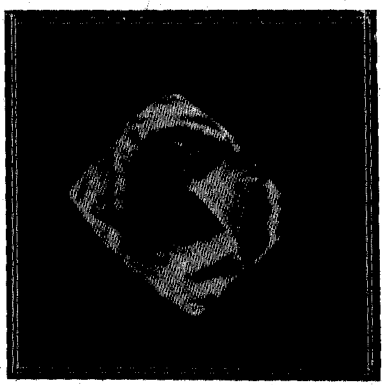


Figure 7

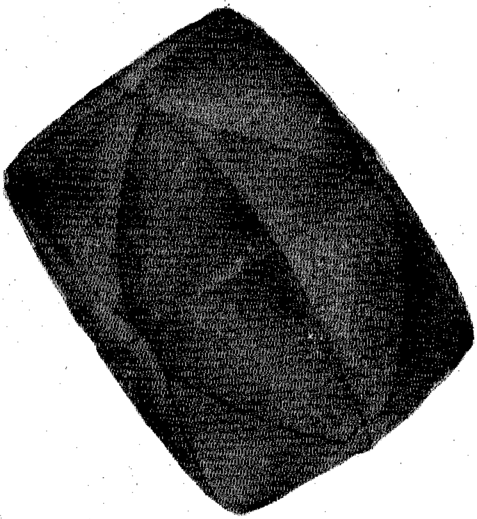


Figure 8

N.C. 26

Plate II

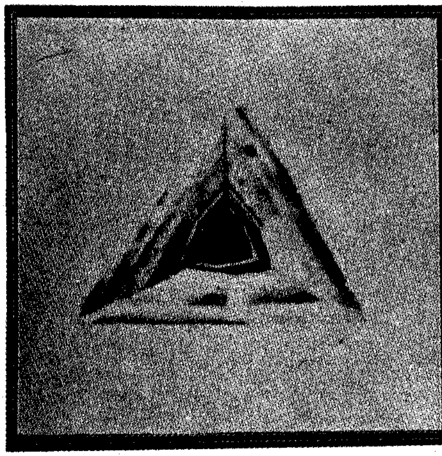


Figure 9

N.C. 164

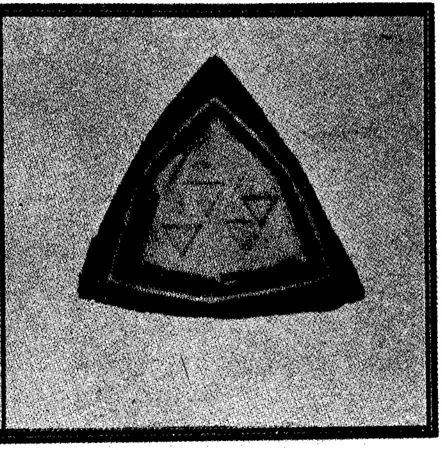


Figure 10

N.C. 25

Figure 11

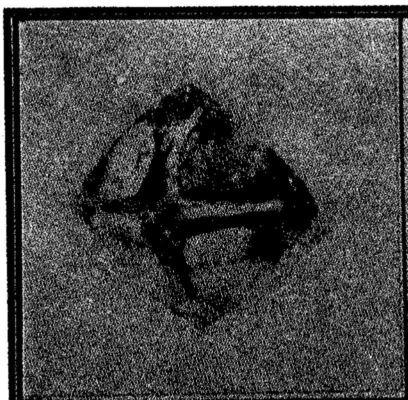
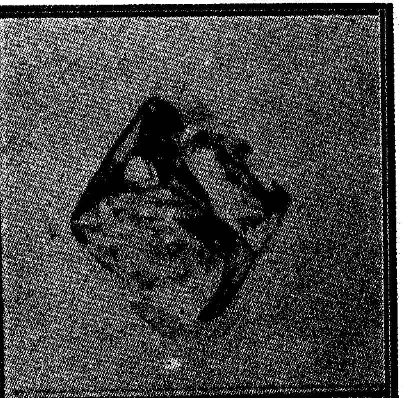


Figure 12



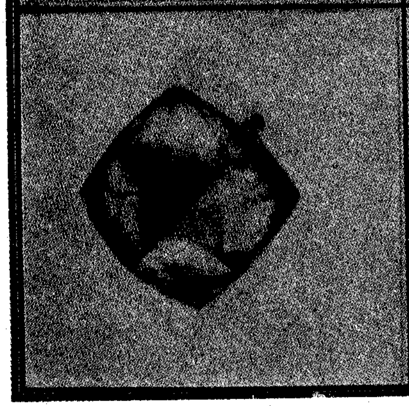


Figure 13

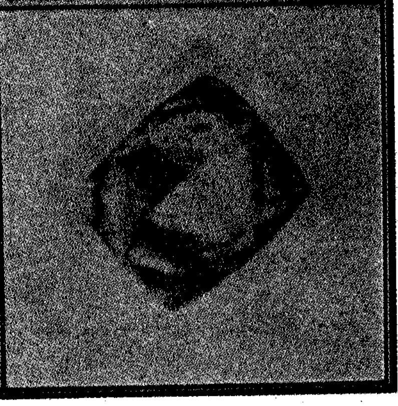


Figure 14

Plate III