

The origin of a tetrahedral diamond

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SUMMARY. The symmetry of diamond is still disputed, and is usually regarded as $4/m\bar{3}2/m$ or $\bar{4}3m$. The (rare) development of tetrahedral morphology has been cited in favour of the lower symmetry. A tetrahedral crystal of c. 4 mm edge-length was studied for evidence relevant to this controversy. Three similar quadrants have nearly plane tetrahedron faces, each surrounded by six curved surfaces belonging to forms $\{hkl\}$. The fourth (unique) quadrant, containing $(1\bar{1}\bar{1})$, differs topographically. In the three similar quadrants the tetrahedron faces exhibit coplanar banding of trigons parallel to $(1\bar{1}\bar{1})$, and the curved surfaces have striations parallel to it. The banding and striations are interpreted as stratigraphic etching of nitrogen-rich layers parallel to $(1\bar{1}\bar{1})$ in a type I diamond. Slip, and apparently polygonization, occur in the unique quadrant. The three similar $\{111\}$ faces were formed by cleavage, and the curved surfaces, banding, and striations by subsequent dissolution. It is probable that $(1\bar{1}\bar{1})$ was part of an original octahedral surface, since tetrahedra of diamond are so rare. The tetrahedral morphology does not necessarily indicate that this crystal belongs to class $\bar{4}3m$. The doubt cast upon tetrahedral morphology (and the inferred twinning on $\{100\}$ or about $\langle 100 \rangle$) as evidence in favour of lower symmetry strengthens the case for assigning diamond to class $4/m\bar{3}2/m$.

THE symmetry of diamond has been the subject of controversy for a very long time, most workers having assigned it variously to the point groups $4/m\bar{3}2/m$ or $\bar{4}3m$. The holosymmetric class appears to be indicated by structural studies, the lack of perceptible piezoelectric properties and the most characteristic morphologies of diamond. The occurrence of crystals that have been interpreted by some observers as twins on $\{100\}$, especially notched or grooved crystals of octahedral habit, and the occasional development of tetrahedral forms, is the evidence adduced for the lower symmetry. A concise statement of the position, citing the critical references, is given by Palache *et al.* (1944), who describe twinning on $\{100\}$ and reproduce a classical drawing of a grooved octahedron. More comprehensive accounts of views on the symmetry of diamond, and additional references, are given by Hintze (1904), Fersmann and Goldschmidt (1911), Williams (1932), and Orlov (1977).

When the opportunity of studying a tetrahedral diamond occurred it was gratefully accepted, in the

hope that it might yield new evidence concerning an old controversy.¹

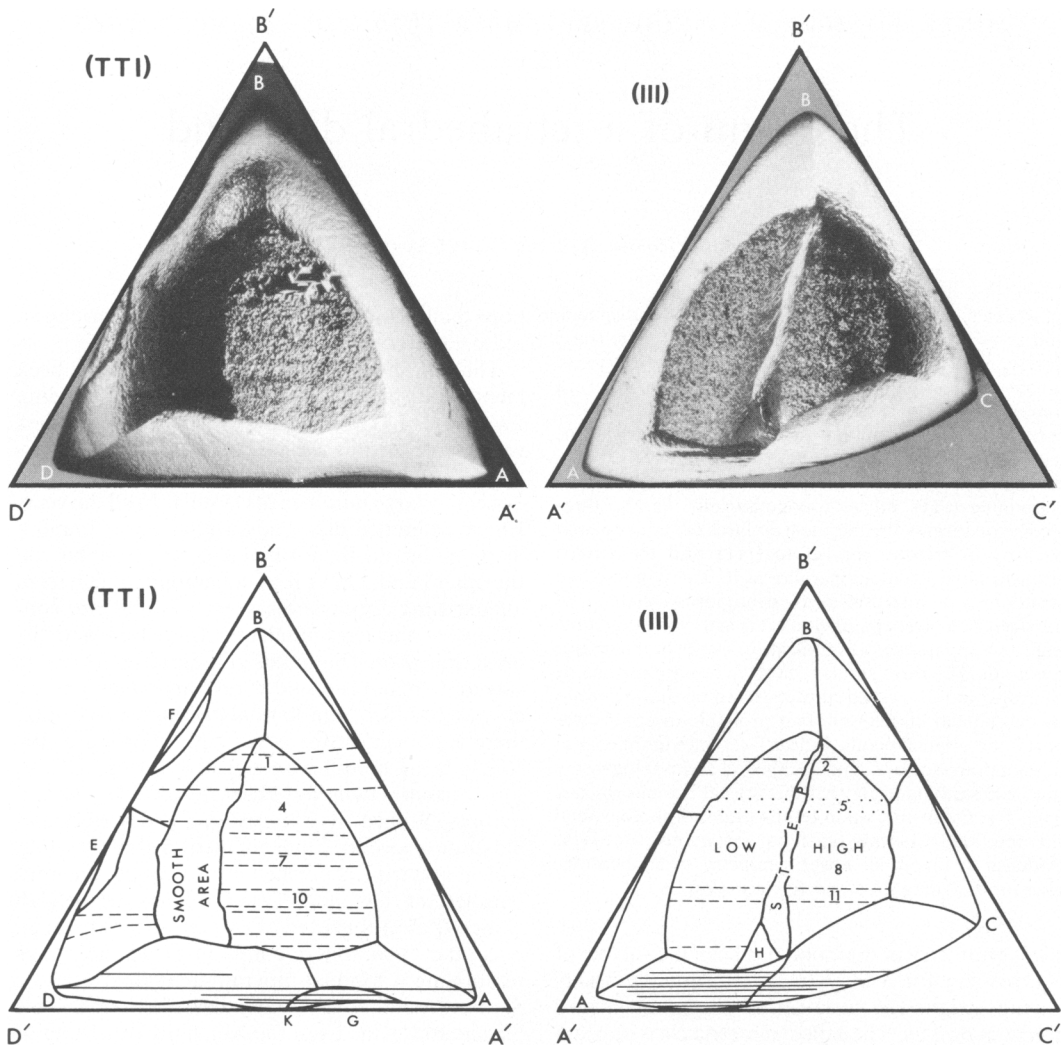
The tetrahedral habit of diamond has been mentioned by numerous authors, including Palache *et al.* (1944). Some significant observations were made on this subject by Williams (1932, pp. 448-9), who had the opportunity of studying extremely large numbers of crystals. 'In all the years I have examined diamonds, I have never found a diamond having the form of a tetrahedron; but, on the other hand, I have found diamonds which from all outward appearances seem to fulfil the conditions of the tetragonal tristetrahedron and the hextetrahedron. Diamonds of these latter forms are rare and are never found with sharp edges, but are always rounded.' 'In crystals of this description there is no sign of the edges and little sign of the coigns being modified by negative forms . . .'

Tetrahedral twins have been described by several authors. Williams (1932, pp. 497-9) describes and illustrates several kinds of tetrahedral crystals, which are triplets formed by twinning on non-parallel $\{111\}$ planes. A crystal from the Belgian Congo (Zaire) consisted of two tetrahedra forming a contact twin on a common (111) face, thus resembling a trigonal bipyramid (Polinard, 1950). Shafranovskii *et al.* (1966) describe remarkable tetrahedral twins from the Mir pipe, consisting of two octahedra twinned on (111) , but developing a tetrahedral habit by the suppression of certain faces.² Casanova *et al.* (1972) have given an account of a diamond from the Ivory Coast, West Africa, in which essentially tetrahedral units form a fivefold cyclic twin by repeated twinning planes of $\{111\}$, all components having a single $[\bar{1}10]$ direction in common.

Diamonds of tetrahedral morphology could arise in various ways. In holosymmetric crystals, tetrahedral form could be due to irregular growth of an octahedron or to twinning, whereas in crystals having the symmetry $\bar{4}3m$ a tetrahedron would be one of the possible forms. Crystals of

¹ The author is deeply indebted to Mr Eric Bruton for the loan of his tetrahedral diamond.

² The author is most grateful to Dr M. Moore for a translation of this Russian paper.



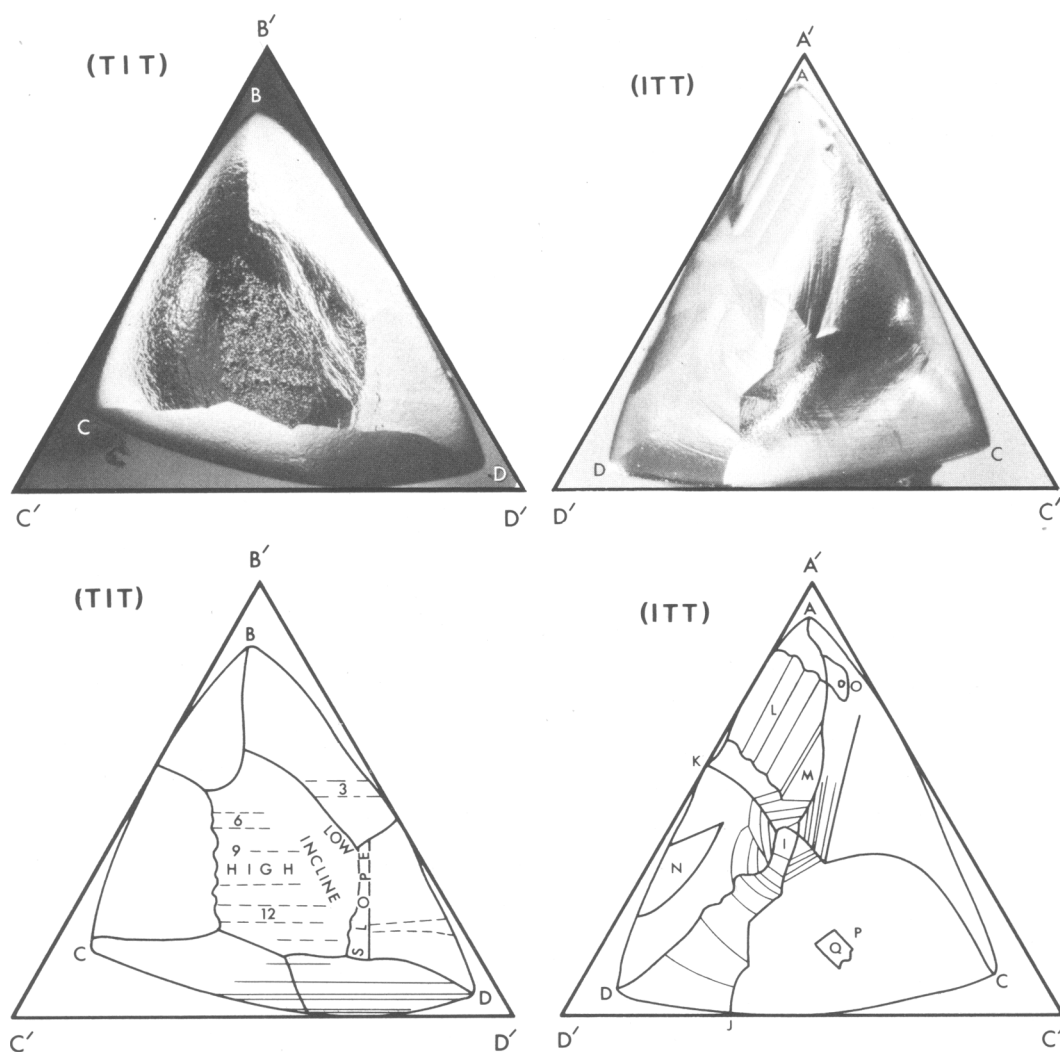
FIGS. 1-8. The photographs and drawings of the four quadrants of the tetrahedral diamond (all $\times 14$) are each enclosed in an equilateral triangle with sides in $\langle 110 \rangle$ directions. The more important features are lettered in each drawing: for description see text. FIGS. 1-4, top; 5-8, bottom. FIGS. 1 and 5. The quadrant ABD, including $(\bar{1}\bar{1}1)$. FIGS. 2 and 6. The quadrant ABC, including (111) . FIGS. 3 and 7. The quadrant BCD, including $(\bar{1}\bar{1}\bar{1})$. FIGS. 4 and 8. The quadrant ACD, nominally including $(1\bar{1}\bar{1})$.

either class of symmetry could give rise to tetrahedral cleavage fragments. Thus the development of tetrahedra does not necessarily imply that the symmetry of diamond is lower than that of the holosymmetric class.

Description of the tetrahedral diamond

Morphologically the crystal is a tetrahedron combined with a 'hextetrahedron'. The latter is not a plane-faced form, but has the curvature so characteristic of these surfaces on diamond, in

which the curvature increases greatly towards the edges and even more near the coigns. Each curved surface may be deemed to consist of a large number of plane elements or faces, all of which belong to forms of the type $\{hkl\}$. This connotation will be implied in subsequent usage of the term ' $\{hkl\}$ surfaces'. This crystal closely resembles one illustrated by Williams (1932, pl. 155, fig. 2b). The tetrahedron has been arbitrarily designated $\{111\}$ and the four coigns lettered A-D. Individual quadrants are defined by their three coigns, and will include the $\{hkl\}$ surfaces, $\{111\}$ faces, and any



other features within the area. Figs. 1-4 are photographs of the four quadrants. Each is inscribed in an equilateral triangle, having edge directions $\langle 110 \rangle$, the corners of which bear the same letter as the adjacent coign, but are distinguished by a superscript dash. Each quadrant is also illustrated by a sketch of the more important features (figs. 5-8), since it is impossible to make them all visible on a single photograph with one form of lighting, or at a low magnification. Interpretation is facilitated by describing the crystal quadrant by quadrant, correlating the structures on the tetrahedral faces with those on the adjacent $\{hkl\}$ surfaces.

Three of the quadrants are similar, having a nearly plane $\{111\}$ central area, which is surrounded by $\{hkl\}$ surfaces. The fourth quadrant is markedly

different, with a more rugged topography, lacking the plane central area, but having smooth curved surfaces with lamellar structure near corner C. All tetrahedral surfaces on this specimen are covered with trigons (Sutton, 1928), the triangular pits so common on $\{111\}$ faces of diamond. The trigons have the same orientation as the tetrahedron face, and would be designated negative on the classification of Frank *et al.* (1958). Although some pits on this crystal have been truncated on one or more corners, producing 4, 5, and 6-sided pits, they will be referred to collectively as trigons. The $\{hkl\}$ surfaces typically have small-scale structures somewhat resembling those characteristic of $\{110\}$ faces of diamond (Williams, 1932, pl. 174), but lacking all symmetry, in accordance with their orientation

(fig. 9, top left and bottom right). There is usually a narrow transition zone between the $\{111\}$ and $\{hkl\}$ surfaces, observable at higher magnifications. These zones are smoother and brighter than the adjacent areas: they bear a few trigons (presumably on elements of $\{111\}$), which are much smaller and more widely separated than on the adjacent $\{111\}$ face, and other small features are also present. No additional faces occur on the coigns or edges of the crystal, but slight abrasion and a small conchoidal fracture are present.

The crystal is brown, with a pinkish tinge, and the strongly curved edges vary in length from 3.9 to 4.9 mm. Determination of the absorption characteristics in ultraviolet light was prevented by the tetrahedral shape of the diamond, but luminescence was observed. The colour was determined visually and the duration of phosphorescence estimated. Under long-wave UV (with most output about 366 nm) fluorescence is fairly strong and pale yellow. Phosphorescence is moderately strong for *c.* $\frac{1}{3}$ s and very pale yellow, followed by a longer period (30 s?) with a very dim glow of indeterminate colour. Under short-wave radiation (maximum intensity about 254 nm and smaller peak at 366 nm) fluorescence is weak and pale brownish yellow. Phosphorescence is very weak, of indeterminate colour, with a duration of possibly 30 s.

Quadrant ABD, including $(\bar{1}\bar{1}1)$. The face $(\bar{1}\bar{1}1)$, the six surrounding $\{hkl\}$ surfaces, and the more significant features are shown in figs. 1 and 5. This is the most symmetrical quadrant of the crystal. The large dark area to the left of $(\bar{1}\bar{1}1)$ is an exceptionally wide transition zone, inclined at a small angle to $(\bar{1}\bar{1}1)$. When rotated slightly this zone becomes the brightest area of ABD in reflected light, and will be called the 'smooth area'.

One of the most noticeable features of $(\bar{1}\bar{1}1)$ is a group of large trigons. At higher magnification it is apparent that the larger trigons are aligned in bands, the more obvious of which have been numbered 1 to 12 in figs. 5-7. The largest trigons on $(\bar{1}\bar{1}1)$ are in band 4. Where the prolongation of this band crosses the smooth area, the latter has an even lower density of pits than elsewhere, and a higher reflectivity. A continuation of band 4 can also be traced across the $\{hkl\}$ surfaces to AB and almost to BD by a change in the microtopography, although no trigons are present. On $(\bar{1}\bar{1}1)$ the band is parallel to A'D', but it has a slight deflection on the curved surfaces. Subsequently the direction of bands will be stated for the tetrahedron faces only, without repeated mention of the deflection. Band 1, parallel to A'D', is relatively featureless on the smooth area, has fairly large trigons on $(\bar{1}\bar{1}1)$ and continues as a smooth band across an $\{hkl\}$ surface to AB. Below

band 4 several less distinct bands of trigons are grouped as band 7, below which is band 10, the most prominent on this face. Immediately below it is another band, which extends across the smooth area and the adjacent $\{hkl\}$ surface to BD, widening as it approaches the edge. It appears less rough than the rest of the $\{hkl\}$ surface. Additional bands are present to the lower edge of $(\bar{1}\bar{1}1)$.

At least six very narrow striations are present on the $\{hkl\}$ surfaces adjacent to AD. They are virtually rectilinear and parallel to A'D', but show a very small change of direction on crossing the salient edge between the two $\{hkl\}$ surfaces. One striation runs from A to D; on the very strongly curved surface near coign D the striation is deflected towards B.

The feature E has a surface parallel to $(\bar{1}\bar{1}1)$ on which many PD (pyramidal, deep) and some FD (flat-bottomed, deep) trigons are present: the steep arcuate bounding surface appears to be composed of cleavage steps. The various types of trigon have been classified by Frank and Lang (1965).

Feature F superficially resembles E, but its base is not parallel to $(\bar{1}\bar{1}1)$ and no trigons were seen.

A ridge on $(1\bar{1}\bar{1})$ meets AD at K, forming a projection when viewed on $(\bar{1}\bar{1}1)$. The recess on the right is feature G, through which the striations continue uninterrupted, and virtually parallel to A'D'.

Quadrant ABC, including (111) . The face (111) , the six surrounding $\{hkl\}$ surfaces, and more important topographic features are shown in figs. 2 and 6. This quadrant resembles ABD in many respects: there is a plane central tetrahedron face, bearing trigons of the same orientation, surrounded by six curved, hummocky $\{hkl\}$ surfaces (fig. 9, bottom right), and in some areas a transition zone is present between the latter and (111) . Banding of trigons also occurs, parallel to A'C', and there are strong striations near the edge AC. One of the most notable differences is the step, atypical of diamond, that nearly bisects (111) , and widens into a feature like a delta at H. The high side of the step is adjacent to BC.

The banding of trigons is less obvious than on $(\bar{1}\bar{1}1)$. Band 2 contains some fairly large trigons on (111) and can be traced across the face and the adjacent surfaces to the edges AB and BC. On the former it meets band 1. Bands 5 and 11 extend the width of (111) , and between them are some ill-defined bands, which will be grouped together as band 8. Additional bands, apparently six, are present below band 11. Nearly all the trigons are PD, but one large and several small FD trigons are present.

A series of striations occurs on the two $\{hkl\}$ faces adjacent to the edge AC (figs. 2, and 9 bottom right).

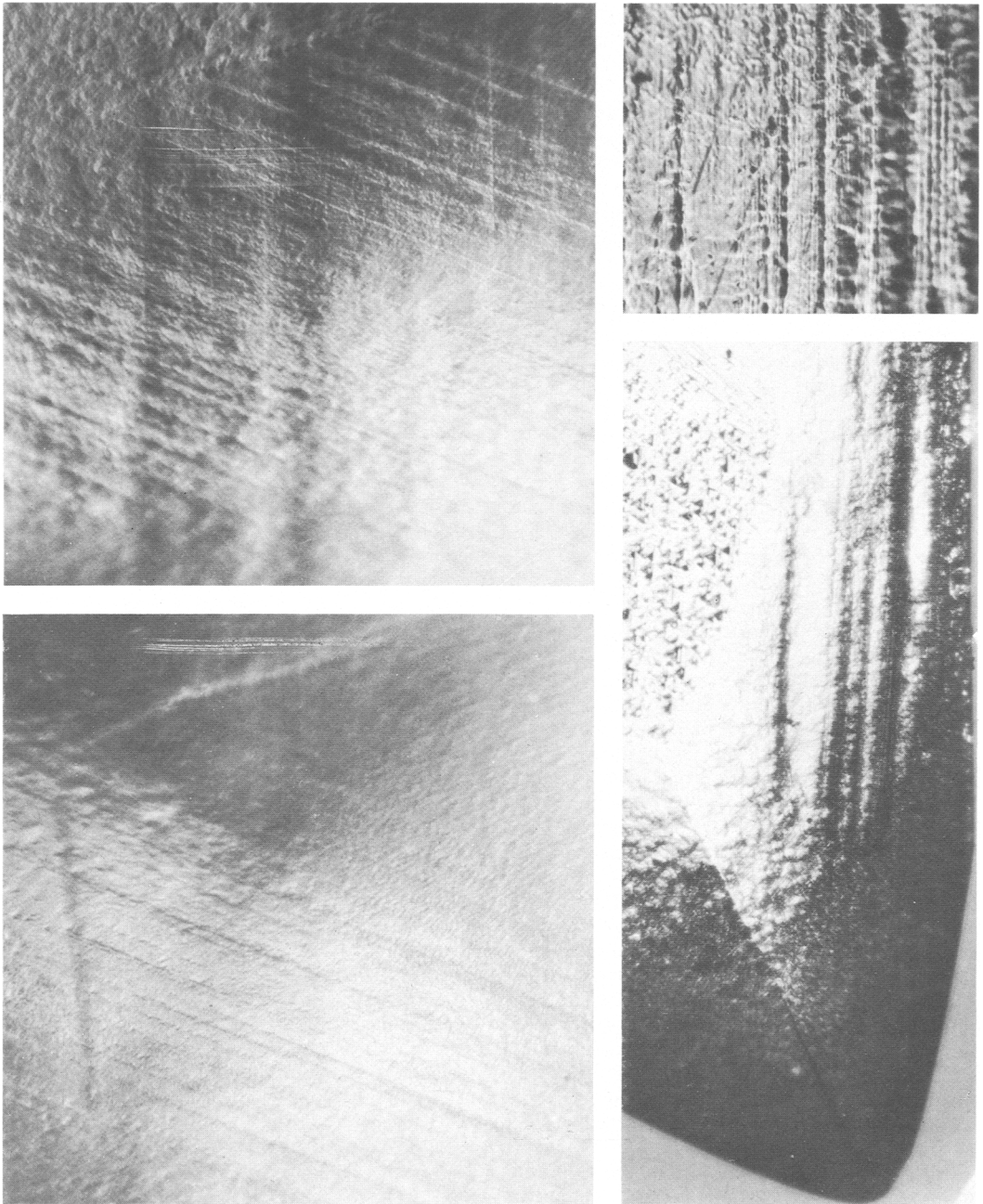


FIG. 9: (*Top left*). Part of the surface P on $(1\bar{1}\bar{1})$ with broad striations (and some narrow lamellae) parallel to $A'C'$ (vertical), and narrow lamellae parallel to $C'D'$. Typical $\{hkl\}$ surface features in top left-hand corner. $\times 102$. (*Bottom left*). The same area, slightly defocused, showing misorientated region. $\times 106$. (*Bottom right*). Corner A of ABC, showing trigons on (111) , dissolution-striations parallel to $A'C'$ on curved $\{hkl\}$ surfaces, the latter's typical hummocky topography, and a corner of the 'delta'. $\times 69$. (*Top right*). Detail of dissolution-striations on ABC, a little nearer C than those in the adjacent fig. $\times 280$.

Many more striations can be seen at higher magnification (fig. 9, top right). They almost reach A and extend over three-quarters of the length of AC, where the innermost striation cuts the highly curved edge nearer C. The striations are straight, except near the strongly rounded edge AC, from which they curve gently towards B, and there is a change of direction of $c. 2-4^\circ$ on crossing the salient edge between the two $\{hkl\}$ faces. The striations are almost parallel to A'C', and their mean direction is assumed to belong to the set $\langle 110 \rangle$.

At the foot of the steep step that nearly bisects (111), on the lower side towards AB, there is a relatively smooth narrow strip, which is comparatively free of trigons, but has numerous small surfaces of the type $\{hll\}$ parallel to A'B'. This strip appears to be a transitional area between the base of the steep step and the lower part of (111). In the delta-shaped area H the steps form a divergent pattern, and with the change of orientation there appear to be less well-defined steps $\{hll\}$ parallel to A'C'.

Quadrant BCD, including $(\bar{1}\bar{1}\bar{1})$. The face $(\bar{1}\bar{1}\bar{1})$, the six surrounding $\{hkl\}$ surfaces, and more important topographic features are shown in figs. 3 and 7. The nearly plane face $(\bar{1}\bar{1}\bar{1})$ is interrupted by an inclined surface (the 'incline') sloping down towards the edge BD and nearly parallel to it. The face $(\bar{1}\bar{1}\bar{1})$ is abruptly terminated towards corner D by a feature which will be called the 'slope'. The latter, which is prominent under different conditions of illumination, is normal to the bands of trigons and C'D'.

Most of the banding of trigons is not visible at low magnifications. Band 3 contains relatively large, well-spaced trigons, and band 6 has somewhat larger trigons, but the largest are in band 12. Additional banding is present between bands 6 and 12 (grouped together as band 9), and also below band 12. The most striking of the latter is the one immediately below 12, which appears dark on $(\bar{1}\bar{1}\bar{1})$ and continues as a pale smooth band on the adjacent $\{hkl\}$ face, widening as it extends to the edge BD, where it meets the similar band 10, which also widened up to the edge BD. Band 12 lies in a $\langle 110 \rangle$ direction, but the others cannot be measured accurately.

It may be noted that the incline has reflecting facets $\{hll\}$, which are parallel to B'D'. The incline cuts across the banding and is virtually confined to $(\bar{1}\bar{1}\bar{1})$. At the end near B, there is an area of the transitional surface on each side of the incline.

There are striations on the $\{hkl\}$ surfaces adjacent to CD, which deviate very slightly as they cross the salient edge, but are virtually parallel to C'D'.

Quadrant ACD, nominally including $(\bar{1}\bar{1}\bar{1})$. This

quadrant (figs. 4 and 8) is completely different in character from the three described previously. There are large or small $\{hkl\}$ surfaces adjacent to the coigns, but between them is a more rugged topography. The small triangle I forms the highest area on the face, from which five ridges radiate to A, C, and D, and also to the points J and K, on the edges of CD and AD respectively. Some of these ridges have the shield-shape characteristic of salient edges on diamond. Only very small elements of $(\bar{1}\bar{1}\bar{1})$ faces are present on some of the curved surfaces, and these bear trigons. Between A and the ridge IK is a series of prominent ridges L, which are parallel to A'D' (within the limits of experimental error), the topmost ridge M having the largest area of trigons on ACD. On the opposite side of IK is a depressed area N, adjacent to the edge AD. Near coign A is a large trigon O, which contains other trigons, and is surrounded by an extensive irregular sloping surface. A pattern of intersecting lamellae is present on the very smooth curved $\{hkl\}$ surface P (fig. 9, top left) and the adjacent $\{hkl\}$ surface, from C to about the mid point of CA. Owing to the very strong curvature of P adjacent to the edge and coign, the outcrop of many of the lamellae is slightly curved, but the longer ones appear to be parallel to C'D' and the shorter ones to A'C'. At a magnification of $c. \times 100$ the lamellae appear as very narrow lines, many of which are not resolved at lower magnifications. At $\times 880$ there are linear arrays of small asymmetric features, which are assumed to be etch pits. There are also some broader striations on P, up to $c. 0.1$ mm wide, parallel to A'C', a few of which have one of the lamellae within their width (fig. 9, top left). The lamellae parallel to C'D' appear to cross the striations without deviation. Within the area P is a discrete region Q, 0.42×0.33 mm, having a few parallel dark bands, resembling the broader striations, but inclined at $c. 9^\circ$ to them. These misorientated structures, which appear to be just below the crystal surface, are only visible under particular lighting conditions, due, perhaps, to total internal reflections from internal surfaces (fig. 9, bottom left).

Review of work relevant to interpretation of the observations

The origin of trigons. The common trigons on octahedron faces of diamond are oppositely oriented relative to the face and are said to have negative orientation (Frank *et al.*, 1958). The origin of trigons has been the subject of prolonged controversy: whether they are formed by processes of growth or etching. References to authors holding opposing views are given by Varma (1967). The

controversy was largely due to the fact that trigons are oppositely orientated to the etch pits formed in the majority of experiments, which have positive orientation, and to neglect, or inadequate consideration of the possible effect of different solvents on the orientation of etch pits. Frank *et al.* (1958) argue convincingly that pyramidal trigons are etch features, and explain how the curved salients so typical of diamond surfaces are formed by etching. They describe the structural reason for the stability of edges in $\langle 110 \rangle$ directions on diamond (during growth or etching), and demonstrate that the bounding vicinal surfaces of trigons, which are in negative orientation, are of the type $(1,1,1-\delta)$, i.e. $\{hhl\}$ or trisoctahedral, with $h > l$. The vicinal surfaces of etch pits in positive orientation are of the type $(1,1,1+\delta)$, i.e. $\{hll\}$ with $h > l$ or trapezohedral. During natural etching the latter may form steps on a retreating inclined surface, or truncate the corners of trigons. Frank and Puttick (1958) demonstrated that etch pits of negative orientation can be produced experimentally by etching diamond in fused kimberlite. Evans and Sauter (1961) were able to develop pits in negative orientation on $\{111\}$ faces by etching in wet oxygen above 1000°C and in positive and negative orientation by etching in air at different temperatures.

The classic work of Lang (1964), combining X-ray topography and phase-contrast microscopy, showed that pyramidal trigons are always associated with dislocation outcrops, but the latter do not occur in flat-bottomed trigons, giving very strong support to the view that trigons are formed by etching.

The growth stratigraphy of diamond has been clearly demonstrated by artificial etching in fused KNO_3 , e.g. on octahedral cleavages by Patel and Tolansky (1957), and on sawn and polished cube surfaces by Seal (1965) and by Harrison and Tolansky (1964). Seal obtained many and varied examples of stratigraphic etching in type I diamonds, but virtually uniform etching in type IIa and type IIb diamonds. Harrison and Tolansky attributed the differential etching of adjacent growth horizons to the presence of type I and type II diamond, as indicated by the characteristic absorption of ultraviolet light, the former being more readily etched. Lang (1967) does not believe that alternating zones of type I and type II diamond are generally present, but attributes growth stratigraphy to variations of lattice parameters between adjacent growth horizons, due to non-uniform concentrations of incorporated impurity. The essential chemical difference between type I and type II diamonds is in the higher nitrogen content of the former. This element is known to be present in

variable concentration, which can be correlated with the absorption of ultraviolet light.

The existence of extensive natural dissolution, forming the so-called 'dodecahedral' surfaces on diamond, has been demonstrated by Moore and Lang (1974), using X-ray topography, and by Seal (1965), etching cut and polished slices, both methods showing that the curved surfaces cut across the traces of $\{111\}$ growth horizons. Orlov (1977) has shown that curve-faced forms are secondary, and that the dodecahedroid is one of the most important dissolution forms of diamond.

A phenomenon variously described as lamination, glide, or slip has been described by numerous authors. It occurs on planes parallel to $\{111\}$, which freely intersect each other and also octahedral growth horizons. Sutton (1928) records that most brown diamonds are visibly laminated, and where lamination is not seen on the surface its presence may be indicated by close parallel tinted streaks inside a crystal. Lamination and colour are closely correlated, the darker the brown or mauve colour the greater is the likelihood that the crystal will be laminated in two or three octahedral directions. Williams (1932, p. 460) states that diamonds are often found that show striations parallel to one and sometimes two of the four possible cleavage planes, but specimens showing the presence of three planes in the case of the octahedron and four planes in the case of the rhombic dodecahedron are rare. Williams (1932, p. 426) also records that most brown stones and some white and other coloured stones show glide planes to a very marked degree. Lamination has been found in a type II diamond by Custers (1951) and in a type I diamond by Denning (1961). Naturally induced slip in diamond has been recorded by Tolansky *et al.* (1958). Experimental plastic bending of diamond plates at 1800°C , with the development of multiple slip, showed that type I specimens deformed at significantly higher stresses than specimens of type II diamond (Evans and Wild, 1965). Urussovskaya and Orlov (Orlov, 1977), by the use of Laue diagrams, showed that plastic deformation led to polygonization, and the formation of misorientated regions, believed to occur at high temperatures. Some colours, such as mauve and smoky-brown, are caused by deformation, coloured bands being contiguous with glide planes (Orlov, 1977). It has been suggested that the coloration is due to defect structures, or possibly to the presence of finely dispersed graphite.

In a study of birefringence in diamond, Lang (1967) has described and illustrated tatami patterns, which may resemble a rice-straw mat. They correspond to the laminated structure of many earlier authors. Lang noted that the lamellae of the tatami

pattern may be present parallel to one or more octahedral planes. When the lamellae are present in two or more orientations, they intersect each other and the growth stratification. The tatami pattern has been attributed to plastic deformation on sets of slip planes by Frank (1967). Moore and Lang (1972), using X-ray topography, have shown the presence of a strong tatami pattern in a diamond included within a relatively strain-free diamond. Lang (1967) has described a diamond with mosaic structure, which is believed to have been deformed plastically and subsequently annealed naturally, and polygonization has been recorded by Frank (1967) and Orlov (1977, p. 144).

The octahedral cleavage of diamond is well known, but Sutton (1928, pp. 16–17) also mentions a 'less perfect though quite good' $\{110\}$ cleavage, and that it occurs occasionally on $\{100\}$. Williams (1932) mentions the highly perfect $\{111\}$ cleavage, and, referring to Sutton's observations, states that he has not been able to prove that $\{110\}$ or $\{100\}$ cleavage has taken place (pp. 463–4). However, in a reference to broken diamonds he states, 'When a diamond is broken the fracture is usually along the crystallographic planes, although many stones break at right angles to these planes' (p. 427), which could be a reference to $\{110\}$ cleavage. Williams (p. 427) records a diamond said to have burst during cutting, of which 'the majority of the tiny fragments took up the shape of the regular tetrahedron'. Orlov (1977) mentions octahedral cleavage and lists many additional cleavages on surfaces of fractures, including $\{110\}$, but not $\{100\}$.

Diamonds obtained directly from kimberlite are often found to be broken, but very few fractures can be attributed to the processes of extraction. Such crystals are referred to by Sutton (1928, pp. 68–72), Williams (1932, pp. 425–30), and Orlov (1977, p. 205).

Orlov (1977) reviews the literature on the various structures that have been proposed for diamond, and the observations or assumptions on which they were based. Covalent and ionic structures have been postulated, and symmetries of classes $\bar{3}2/m$, $2/m\bar{3}$, $\bar{4}3m$, and $4/m\bar{3}2/m$ proposed. He quotes Raman's suggestion that diamond may possess four structures, two of which belong to class $\bar{4}3m$ and two to the holosymmetric class. However, Orlov does not mention the strong criticism by Lonsdale (1945) and Taylor (1947) of Raman's proposed structures, neither of whom accepted the evidence for tetrahedral symmetry. After a summation of the evidence, including the effects of nitrogen content upon properties, Orlov concludes that the structure for which there is most theoretical and experimental evidence is cubic holosymmetric, with space group $Fd\bar{3}m$.

Interpretation of the origin of the tetrahedral diamond

Three quadrants of the tetrahedral diamond, ABC, BCD, and ABD, are very similar in their general characteristics, having a central, approximately plane area of simple crystallographic orientation, surrounded by curved $\{hkl\}$ surfaces. The fourth quadrant, ACD, is markedly different, and may be regarded as the 'unique' face. The presence of trigons is accepted as evidence that the crystal suffered etching at some period prior to its separation from the kimberlite matrix (Frank and Puttick, 1958). The first three faces mentioned all show banding of trigons, which in each case is parallel to $(\bar{1}\bar{1}\bar{1})$, on which such banding is absent. Where some bands of trigons meet the $\{hkl\}$ surfaces there is a linear continuation of distinctive surface markings, which can be proved in a few cases to meet in a common edge, e.g. bands 1 and 2. These are thus coplanar, and band 3 almost certainly lies in the same plane. Measurement of the distance of bands from the common coign B has shown that bands 4, 5, and 6 are coplanar, and so are bands 10, 11, and 12. All are parallel to $(\bar{1}\bar{1}\bar{1})$. The bands of trigons on $\{111\}$ faces and their extensions as topographic features on $\{hkl\}$ surfaces are regarded as examples of stratigraphic etching (Patel and Tolansky, 1957) on a much more extensive scale than has been produced experimentally. The coarse to fine striations that are present in the three similar quadrants, on the curved $\{hkl\}$ surfaces adjacent to $(\bar{1}\bar{1}\bar{1})$, e.g. adjacent to the edges AC, AD, and CD, are also regarded as the expression of stratigraphic etching of these surfaces. The very small change of direction across the salient edge between two adjacent $\{hkl\}$ surfaces and the direction of the sharper curvature near the strongly rounded edges are all compatible with differential etching of the traces of planes parallel to $(\bar{1}\bar{1}\bar{1})$. Differential etching of impurity-laden growth horizons has been demonstrated by X-ray topography (Moore and Lang, 1974). The intersecting lamellae on $(\bar{1}\bar{1}\bar{1})$ are interpreted as examples of slip, which is known to be common in brown diamonds (Sutton, 1928; Williams, 1932; Orlov, 1977). In the quadrant BCD reflections have been observed from internal fractures near the edge CD, which seem to coincide with a set of lamellae parallel to $(\bar{1}\bar{1}\bar{1})$.

It is concluded from the planar character of the stratigraphic etching parallel to $(\bar{1}\bar{1}\bar{1})$ on the three similar faces that they were produced by cleavage from a considerably larger crystal, exposing only traces of growth horizons parallel to $(\bar{1}\bar{1}\bar{1})$, which, from its topography, is believed to be part of one of the original faces of the crystal. The octahedral-plane stratification indicates that the crystal is a

type I diamond (Orlov, 1977). Extensive etching or dissolution, after this natural cleavage, appears to account for many of the observed features of this tetrahedral diamond. Friedel (1926) gave a generalized account of the rounding of edges and coigns of crystals during dissolution. Frank and Puttick (1958) produced various rounded forms experimentally by immersing octahedra of diamond in fused kimberlite, and explained theoretically why such forms should be developed by dissolution. When their argument is applied to a tetrahedron, it would be expected to dissolve most rapidly on the coigns, less on the edges, and least rapidly on the faces, thus developing the characteristic double curvature of the $\{hkl\}$ surfaces that surround the tetrahedron faces. Such is the morphology of the tetrahedral diamond described in this paper.

Placing the photograph of each quadrant in a triangle, the edges of which lie in $\langle 110 \rangle$ directions and are tangential to the edges, gives an indication of the minimum dissolution of the coigns and their surrounding areas. Dissolution in the region of the lamellae on $(\bar{1}\bar{1}\bar{1})$ is obviously much more extensive than elsewhere. The retreat of coign C is considerably greater than that of the other three coigns. The relative dissolution is shown by the lengths (in mm) of the perpendiculars from each coign to the opposite face: A = 4.37, B = D = 4.12, C = 3.66. Each vertex was also projected on to the opposite face, and its displacement from the centre of the circumscribing triangle determined. For A, B, and D the displacement varied from 0.08 to 0.16 mm. For C the displacement on A'B'D' was 0.59 mm, precisely in the direction of B', i.e. away from $(\bar{1}\bar{1}\bar{1})$. The great extent of dissolution in the region of the lamellae is evident in figs. 2 and 3, which show the very strong curvature of AC from A'C' and of DC from D'C'. The recession of C from the plane A'C'D' $(\bar{1}\bar{1}\bar{1})$ is greater than the retreat of C from C'. Lang (1967) showed that diamonds that had suffered plastic deformation (laminated diamonds) exhibited very strong strain birefringence, and gave tatami patterns, both of which are attributable to slip. It is to be expected that unannealed strained regions would dissolve more readily than those that had not suffered deformation.

The quadrant ABD, including $(\bar{1}\bar{1}\bar{1})$, is opposite coign C and most closely approximates an equilateral triangular outline. The base of feature E is densely populated with trigons, therefore cleavage must have occurred in this area before etching ceased. Feature F may represent an earlier and deeper cleaved area than E: earlier because the edges are more rounded and leave no octahedral surfaces for trigons to develop; deeper because F forms a marked recess on the edge BD, as seen on

BCD. Features G and K are due to the high-relief topography of ACD. The quadrant ABC, including the face (111) , differs from ABD in the step that nearly bisects the face and broadens into the delta-like feature H. The step is interpreted as a relatively large cleavage step, which dissociates into tear lines (Zapffe *et al.*, 1951) or river systems (Wilks, 1958) in the area of the 'delta'. Along the sloping surface, and smaller steps formed by the cleavage, etching has developed ridges in a $\langle 110 \rangle$ direction, which have $\{hll\}$ surfaces facing AB (Frank *et al.*, 1958). Numerous trigons on this crystal also have $\{hhl\}$ surfaces truncating one or more of their corners. In the quadrant BCD, including face $(\bar{1}\bar{1}\bar{1})$, the origins of the incline and slope need explanation. The incline is presumed to represent a region of stepped octahedral cleavage, which develops similar characteristics to the step on (111) after etching. The slope could be due to a small etched $\{110\}$ cleavage step. This cleavage has been recorded by Sutton (1928) and Orlov (1977), and would not be inconsistent with Williams's statement (1932, p. 427) that many crystals break at right angles to the (octahedral) cleavage.

The quadrant ACD, approximately represented by $(\bar{1}\bar{1}\bar{1})$, is believed to be the modified remnants of an original face, since it is parallel to the growth planes revealed by stratigraphic etching, $(\bar{1}\bar{1}\bar{1})$. If there had been, originally, a relatively plane octahedral face $(\bar{1}\bar{1}\bar{1})$, then ACD should be similar in its topography to the other three quadrants, with a plane central area surrounded by six $\{hkl\}$ surfaces, but lacking the banding of trigons. $\{hkl\}$ surfaces are present at each coign and extend along varying proportions of the edges. Where these surfaces are absent there is a much more rugged topography. These $\{hkl\}$ surfaces could have formed part of a hexoctahedron, but it seems much more satisfactory and consistent to account for the formation of all the $\{hkl\}$ surfaces on the crystal by one mechanism, instead of proposing different, and rather arbitrary, origins for the six surfaces of one quadrant. If one cleavage occurred parallel to each of the three similar faces, the outline of the original $(\bar{1}\bar{1}\bar{1})$ face, immediately after cleavage, would have been an equilateral triangle with edges parallel to A'C'D', but somewhat larger than it is now, because dissolution has occurred on the newly formed edges and coigns. On ACD $\{hkl\}$ surfaces are developed to varying extents around all three coigns, but the remaining area has much stronger relief, and most of it stands above these surfaces, culminating in I. A sudden change of slope occurs as the smooth $\{hkl\}$ surfaces are traced inwards from the coigns. One of the most obvious examples is in the area between the ridges DI and JI. A much more extensive change of slope extends almost across the face from just

below trigon O to the edge CD at J. The $\{hkl\}$ surface extending from D towards A suffers a sudden change where it meets the ridge IK. All the ridges extending from I have shield-shaped striations, characteristic of salient edges that have retreated through dissolution (Frank *et al.*, 1958). Less prominent striations of this type also occur nearer D and on P. On the other three quadrants the $\{hkl\}$ surfaces are very smoothly curved. If the process of dissolution had reached completion, only curved $\{hkl\}$ surfaces would be present in each quadrant. In the three similar quadrants lack of completion is shown by the remaining presence of areas of $\{111\}$ planes. Further, on BCD the incline terminates at the edges of the face $(\bar{1}\bar{1}\bar{1})$, and on ABC the step and delta terminate at the edge of (111) , because any further pre-existing extension would have been removed by the dissolution on the $\{hkl\}$ surfaces. On ACD the incomplete reduction of the original topography has left areas of strong relief. The series of ridges L, in a $\langle 110 \rangle$ direction, also indicates the retreat of a surface from AD through dissolution.

The slip planes in the area P have two orientations. The direction and curvature of the longer traces of the lamellae, approximately parallel to C'D', suggest that slip occurred on planes parallel to $(\bar{1}\bar{1}\bar{1})$, and the shorter traces, approximately parallel to A'C', represent slip on planes parallel to (111) . This area presents two problems, for which tentative solutions are suggested. The broader striations (fig. 9, top left), which are parallel to A'C' and to one set of lamellae, may represent slip bands that are present in addition to numerous discrete slip planes. These two structures may exhibit diverse sculptures (Orlov, 1977) or topographical features, after etching, if the slip bands and slip planes have different densities of dislocations. The presence of a small misorientated unit (fig. 9, bottom left) within the region of lamellar structure, mentioned above, may perhaps be due to polygonization.

It has been deduced from the observations given above that the tetrahedral morphology is due to natural cleavage on three surfaces. The presence of trigons, and other features on these surfaces indicative of etching, together with the marked rounding of all edges and coigns, shows that considerable dissolution occurred after cleavage. Thus the tetrahedral morphology of this crystal does not indicate that it necessarily belongs to class $\bar{4}3m$; in fact no evidence has been found for assigning this diamond to class $\bar{4}3m$ or $4/m\bar{3}2/m$. However, the existence of tetrahedral morphology has formerly been used to infer that diamond belongs to the lower class of symmetry. The study of this crystal shows that such an argument is

not necessarily valid, and it is possible that further examination of other tetrahedral crystals may add to this evidence. Since doubt has been cast upon tetrahedral morphology as evidence in favour of lower symmetry, the case is correspondingly strengthened for assigning this particular diamond to the holosymmetric class of the cubic system. Likewise, some authors (e.g. Williams, 1932; Orlov, 1977) do not believe that 'notched octahedra' are twinned crystals, thereby reducing the strength of another classical argument in favour of tetrahedral symmetry. Since the majority of workers believe that diamond has only one structural modification, either with the symmetry $\bar{4}3m$ or $4/m\bar{3}2/m$, the evidence presented in this paper weakens the case for the former, and may indicate that diamonds in general are holosymmetric.

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REFERENCES

- Casanova (R. D.), Simon (B.), and Turco (G.), 1972. *Am. Mineral.* **57**, 1871-3.
- Custers (J. F. H.), 1951. *Research*, **4**, 131-6.
- Denning (R. M.), 1961. *Am. Mineral.* **46**, 740-3.
- Evans (T.) and Sauter (D. H.), 1961. *Phil. Mag.* **6**, 429.
- and Wild (K. R.), 1965. *Ibid.* **12**, 479-89.
- Fersmann (A.) and Goldschmidt (V.), 1911. *Der Diamant*. Carl Winter, Heidelberg.
- Frank (F. C.), 1967. Proc. Intern. Industrial Diamond Conf., Oxford, 1966. (London: Industrial Diamond Information Bureau), 1967, 119-135.
- and Lang (A. R.), 1965. In Berman (R.) (ed.), *Physical Properties of Diamond*. Clarendon Press, Oxford.
- and Puttick (K. E.), 1958. *Phil. Mag.* **3**, 1273-9.
- and Wilks (E. M.), 1958. *Ibid.* 1262-72.
- Friedel (G.), 1926. *Leçons de cristallographie*. Berger-Levrault, Paris.
- Harrison (E. R.) and Tolansky (S.), 1964. *Proc. R. Soc. A*, **279**, 490-6.
- Hintze (C.), 1904. *Handbuch der Mineralogie* (Verlag von Veit & Comp., Leipzig), **1**, Pt. 1, 14.
- Lang (A. R.), 1964. *Proc. R. Soc. A*, **278**, 234-42.
- 1967. *Nature*, **213**, 248-51.
- Lonsdale (K.), 1945. *Ibid.* **155**, 144.
- Moore (M.) and Lang (A. R.), 1972. *Phil. Mag.* **25**, 219-27.
- 1974. *J. Crystal Growth*, **26**, 133-9.
- Orlov (Yu. L.), 1977. *The Mineralogy of the Diamond*. J. Wiley and Sons, New York, etc.
- Palache (C.), Berman (H.), and Frondel (C.), 1944. *Dana's System of Mineralogy*, **1**. J. Wiley and Sons, New York, etc.
- Patel (A. R.) and Tolansky (S.), 1957. *Proc. R. Soc. A*, **243**, 41-7.
- Polinard (E.), 1950. *Ann. (Bull.) Soc. géol. Belgique*, **74** (for 1950-1), B59-63.

- Seal (M.), 1965. *Am. Mineral.* **50**, 105-23.
- Shafranovskii (I. I.), Alyavdin (V. F.), and Botkunov (A. I.), 1966. *Zap. Vses. Min. Obshch.* **95**, 575-78.
- Sutton (J. R.), 1928. *Diamond*. Murby, London.
- Taylor (W. H.), 1947. *Nature*, **159**, 729-31.
- Tolansky (S.), Halperin (A.), and Emara (S. H.), 1958. *Phil. Mag.* **3**, 675-9.
- Varma (C. K. R.), 1967. *Ibid.* **16**, 959-74.
- Wilks (E. M.), 1958. *Ibid.* **3**, 1074-80.
- Williams (A. F.), 1932. *The Genesis of the Diamond*, 2. Benn, London.
- Zapffe (C. A.), Worden (C. O.), and Zapffe (C.), 1951. *Am. Mineral.* **36**, 202-32.

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