X-Ray Diffraction and Absorption Topography of Synthetic Diamonds*

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1. INTRODUCTION

NATURAL diamonds have been the subject of more intensive scientific study than probably any other crystal found in nature, not least because of their striking variety in habit and other properties. This fact alone would attach interest to a study of the synthetic product. But the long duration of the struggle to make diamonds in the laboratory, the numerous early unsubstantiated claims of success, the sudden reversal of this unfortunate record by the development of an efficient synthesis process in the GE Research Laboratory, and all strengthen the interest in a thorough examination of the man-made specimens. The general morphology of synthetic diamonds has been described by Bovenkerk, and numerous interferometric studies of their surfaces have been made by Tolansky. Some of the chief differences in appearance between the natural and man-made stones are as follows: In the GE process for growing diamonds a thin film of molten metal "catalyst" separates the growing diamond from the reaction mixture. Dissolved carbon diffuses rapidly through this film to feed the growing crystals. The product often bears upon its faces an imprint resulting from the solidification of the "catalyst" film. The diamond surfaces are then veined and wrinkled in a manner not seen on natural stones. Another difference from natural stones is the occurrence of hopped and skeletal growth, an understandable consequence of the faster growth rate. A more interesting and fundamental habit difference, however, is the frequent occurrence on synthetic diamonds of excellently smooth cube faces. Such faces are rare on natural stones, being found chiefly in specimens from the Congo, and are always extremely rough.

The x-ray diffraction patterns of synthetic diamonds have been thoroughly studied by Lonsdale, Milledge, and co-workers. Their most notable finding with GE diamonds was the presence of a "satellite" pattern due to a small proportion of a face-centered cubic nickel-rich phase in parallel orientation with the diamond matrix. This fcc phase has a lattice parameter of 3.539 Å, a value intermediate between that of diamond, 3.567 Å, and of pure nickel, 3.523 Å.

Natural diamonds may exhibit a wide range of lattice perfection. In better specimens x-ray topography can reveal individual dislocations and determine their Burgers vectors. Good long-range regularity of the crystal lattice can be maintained even with the presence of appreciable impurity concentrated in the form of the platelets that produce the anomalous "spike" x-ray reflections of type I diamonds. In other specimens a dispersion of precipitates may be present, causing displacements to be naturally "decorated." Experience of the variety of diffraction effects found in x-ray topographic studies of natural diamonds provided a background for the study of man-made diamonds by similar techniques.

Our investigations had three main aims: (1) to compare the lattice perfection attained in synthetic diamonds with that observed in natural stones, (2) to study topographically the distribution of impurity in synthetic diamonds, and (3) to look for topographic evidence bearing on the different surface and habit

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characteristics of synthetic compared with natural diamond. Although our studies were mainly of an exploratory type we have obtained definite information on all the above-mentioned topics. Our findings suggest that further diffraction and absorption topographic work on synthetic diamonds would be very profitable.

We did not study many stones; only four GE and two South African synthetic diamonds were examined topographically. An account of the best GE specimen and also of one much less perfect but more typical is given here. These two specimens adequately illustrate the principal topographic phenomena observed. For each stone we describe the x-ray diffraction and absorption experiments performed upon it. After discussion of these we elaborate some ideas suggested by the topographs concerning the origin of the cubic habit in synthetic diamonds.

2. EXPERIMENTAL METHOD

The lattice perfection of the synthetic diamonds was studied by the methods of “projection topographs,” and “section topographs.” Variation in blackness on the topograph image can be due either to “orientation contrast” or to “extinction contrast,” or to both together. Orientation contrast arises from the mutual misorientation of parts of the specimen preventing them from simultaneously satisfying the Bragg condition with equal efficiency. It can be detected, and the range of misorientations measured, by taking a series of section or projection topographs with the crystal inclined at different angles to a well-collimated incident beam. Extinction contrast is found only in crystals which are at least locally (i.e., in domains some microns in extent) sufficiently perfect for definite phase relationships to be maintained between all the rays participating in Bragg reflection. The diffraction contrast exhibited by the good specimen described in Sec. 3 was practically all extinction contrast, whereas the poorer specimen described in Sec. 4 showed both orientation and extinction contrast. Shorter x-ray wavelengths give stronger extinction contrast but, for various reasons, poorer topographic resolution. The expected and observed image widths of individual dislocations on diamond topographs, and other diffraction effects found in topographs of this crystal, have recently been discussed.

The first x-ray topographs of synthetic diamonds were made in 1959 using Mo Ka radiation. The present diffraction experiments were all done with Cu Ka radiation which allows a topographic resolution of about 1 µ to be attained. Dislocation images in Cu Ka topographs of natural diamonds are generally about 3–5 µ wide. Ilford L4 nuclear emulsion was used as the recording medium so that the contributions of emulsion grain size were negligible; any apparent granularity seen in the topographs arises from statistical fluctuations in image density. Stereopairs of projection topographs are helpful in locating internal defects; but with Cu Ka radiation and the small interplanar spacings of diamond the high Bragg angles provide unnaturally large convergence angles for stereos.

Our absorption topographic experiments followed up the method used by Lonsdale, Milledge, and Nave to establish the chemical identity of the fcc impurity phase. They compared the relative intensities of diamond and fcc satellite reflections produced by Cu Ka and Cu KB radiations on single-crystal rotation photographs. With Cu KB radiation, which has a shorter wavelength than the nickel Ka absorption edge, they found a marked weakening of the satellite reflections. This identified nickel as a major constituent of the impurity phase, and the degree of absorption loss of Cu KB showed moreover that the impurity particles must be of order 5 µ in size. We have taken absorption topographs of our specimens using strictly monochromatic Cu Ka and Cu KB radiations. A parallel beam of uniform intensity was obtained through the 111 Bragg reflection of x rays from a highly perfect germanium crystal. During the exposure the germanium crystal was translated to and fro to provide a sufficiently broad and very uniform beam. The x-ray tube kilovoltage was kept below 24 kV so that the third harmonic wavelength would not be present in the reflected beam. The absorption topographs were, of course, on the same scale as the diffraction topographs, and the specimens were frequently set in approximately the same orientation as for Bragg reflection in order to facilitate correlation of features on diffraction and absorption topographs. The resolution on absorption topographs was at least equal to, and generally rather better than, that on diffraction topographs. Nickel thicknesses of 5, 10, and 15 µ reduce the intensity of Cu KB radiation to 0.27, 0.07, and 0.02 of its initial value, respectively. Thus nickel-rich inclusions, say 1 µ in diameter and above are effectively quite opaque to Cu KB radiation and stand out with extreme contrast. Nickel 15 µ thick and carbon 400 µ thick both reduce Cu Ka to half its initial intensity, so that the larger nickel-rich inclusions are easily recognizable also on the Cu Ka absorption topographs; and it follows too that their shadows can be seen on the Cu Ka diffraction topographs, especially when the inclusions lie close to the x-ray exit surface of the specimen. Stereopairs of absorption topographs can be taken without the restriction that the specimen must make the Bragg angle with the incident rays. Thus the convergence angle can be chosen to produce the clearest depth visualization—an angle of 10° was found suitable.

The satellite diffraction pattern was studied by taking oscillation photographs of small angular range with the photographic plate sufficiently far from the specimen to separate the satellite reflections from those of the diamond. A plate distance of 11.4 cm was used for the 111
reflection and 5.4 cm for the higher-order reflections. Satellite reflections occurring together with allowed diamond reflections are superimposed on the Laue streak from the strong diamond reflection. This interference can be avoided by studying the satellite reflections of index 222 and 200. We used the latter in some of our experiments, since the diamond 200 "forbidden" reflection is extremely weak.

3. OBSERVATIONS ON A WELL-FORMED SPECIMEN

3.1. Morphology

We will describe first our experiments on a synthetic diamond which ranked among the most perfect of our specimens. Scale drawings of this crystal are shown in Fig. 1, diffraction topographs of it are reproduced in Figs. 2–6 and absorption topographs in Figs. 7 and 8. Like many synthetic diamonds this specimen is a macle (i.e., twin). The 111 twin plane divides the crystal into a major component, thickness 190 μ, and a minor component, thickness 60 μ. Figure 1(a) is a projection on to the twin plane. Continuous lines represent visible edges, interrupted lines are hidden edges (belonging to the minor component) and the chain line outlines the edges of the composition plane. Figure 1(b) is a view from the side and Fig. 1(c) a top view showing the location of three inclusions, A, B, and C. This crystal exhibited only two small cube faces. All surfaces of the crystal were rough. They showed no features characteristic of solution and appeared to be the final growth surfaces, a conclusion confirmed by x-ray studies. Although the crystal was transparent (color light yellow) the wrinkled surfaces made optical examination of the interior very difficult. This crystal showed relatively little birefringence, and most of it was concentrated in the vicinity of inclusions B and C.

3.2. Diffraction Topography

Figure 2 is a projection topograph of the 111 reflection from the twin plane. Both components of the twin are reflecting and so the whole crystal contributes to the image. The shape of the image in Fig. 2 and of those in the other topographs shown of this crystal can be understood by reference to Fig. 1. Thus, Fig. 1(d) shows the outline of the crystal when viewed in a direction making the Bragg angle 22° with the twin plane, i.e., at an angle of view intermediate between those of Figs. 1(a) and (b). In fact the crystal was not mounted with an edge quite parallel to the goniometer axis, so that Fig. 2 is a little skewed with respect to Fig. 1(d). Much of the diffraction contrast in Fig. 2 comes from strained surface layers, particularly on the left-hand part of the image. However, one can discern within the crystal a general pattern of radiating lines of enhanced diffracting power and a broad column of imperfect crystal running upwards from the center of the specimen. Figure 3 is a projection topograph of the major component of the twin, obtained by Bragg reflection from planes parallel to the octahedral face marked o in Fig. 1(c). Contrast due to the strained surface is most evident in the marginal areas of this view of the crystal so that the radiating pattern of internal defects is less obscured. The rod-like inclusion A, the larger round inclusion B, and smaller round inclusion C produce faint shadows in Fig. 2 but are not seen in Fig. 3 since in the latter reflection most of the image-producing volume lies between them and the x-ray exit surface of the crystal. In Fig. 4, on the other hand, which is a projection topograph of the minor component of the twin taken by Bragg reflection from planes parallel to p, the shadows of the three inclusions are clear. This is because B and C lie in the twin-plane and A is imbedded in the major component, i.e., on the observer's side of the diffracting volume. Measurement of the relative positions of the shadows of A, B, and C in these and other topographs

![Fig. 1. Scale drawing of more perfect diamond twin, explained in text. Scale mark 100 μ.](image-url)

![Fig. 2. Projection topograph of synthetic diamond twin, twin plane reflection. Compare shape with Fig. 1(d). Scale mark 100 μ.](image-url)
enable their positions to be fixed within about \( \pm 5 \mu \). The reflection used in Fig. 4 has the advantage that the x-ray exit surface over most of the image area is the composition plane and not a strained outer crystal surface. Hence the internal defects show up well; most of them appear to radiate from the vicinity of the inclusions B and C. However, on the left-hand margin of this image extreme surface strain is evident, causing loss of reflection from one small region by misorientation.

The technique of “limited projection topographs,” in which the diffracted-beam slits are adjusted to prevent the unwanted images of surface damage reaching the photographic emulsion, cannot conveniently be applied to such small crystals as now concern us, but the superimposition of the direct images of internal and surface defects is avoided in section topographs. Moreover, section topographs show higher diffraction contrast from individual defects than do projection topographs. Figures 5 and 6 are section topographs in which the incident x-ray beam (about 15 \( \mu \) wide) cuts the specimen along the lines marked 5 and 6 in Fig. 1(c), using the twin-plane Bragg reflection. Section 5 cuts the crystal close to inclusions B and C; columns of defects fan out above and below these inclusions, individual lines fan out in all directions and surface strain is locally intense. Figure 6 is generally similar. This section cuts through several radiating columns but the inclusion A does not appear responsible for generating noticeable lattice defects; it lies to the right of the highly distorted central region cut by the section. Nor is there a concentration of defects in the twin plane which is intersected close to the right-hand margin of the image.

### 3.3. Absorption Topographs

Figures 7 and 8 show a stereopair of absorption topographs taken with Cu \( K\beta \) radiation, the former with an angle of view making about 42° with the twin plane, and the latter 32°. The latter view of the crystal is thus roughly comparable with that of Figs. 1(d) and 2. The large internal inclusions A, B, and C are obvious. A number of small globular absorbing particles with diameters in the 2- to 5-\( \mu \) range are also evident; stereo examination shows that they are concentrated at or near the surface. But the most striking feature is presented by Fig. 7. As already mentioned, the crystal was mounted slightly skew with respect to the orientation drawn in Fig. 1. This tilt brings the two octahedral faces

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Fig. 7. Cu $K_\beta$ absorption topograph, angle of view about 42° with twin plane of specimen drawn in Fig. 1.

Fig. 8. Cu $K_\beta$ absorption topograph, angle of view about 32° with twin plane of specimen drawn in Fig. 2.

$q$ and $r$ parallel to the x-ray beam in Fig. 7. Strong absorption contrast is produced by the surface skin on these faces, now seen edge-on. The ratio of absorption contrast shown in Cu $K\alpha$ and Cu $K\beta$ absorption topographs of this crystal is quite consistent, with nickel being the major absorbing constituent in the individually resolvable particles, with the exception of inclusion B which appears to absorb Cu $K\alpha$ more strongly than would pure nickel.

3.4. Satellite Diffraction Pattern

It is a reasonable assumption that these individually resolvable nickel-rich particles seen on absorption topographs give rise to the satellite reflections. Oscillation photographs of small angular range were taken about the 111 and 220 diamond reflections. No satellite spots were detected. It is possible that the major particles may have been missed in these experiments since the incident beam did not bathe the whole crystal, or that the satellite structures may have been appreciably misorientated from the crystal matrix. However, considering that in this specimen the estimated aggregate weight of nickel in individually resolvable particles is only about 0.02 $\mu g$, that is about $2 \times 10^{-4}$ of the weight of diamond present, failure to observe the satellite reflections is understandable.

4. OBSERVATIONS ON A LESS PERFECT SPECIMEN

4.1. Morphology

Apart from its relatively large size, 0.5 mm, the specimen we will now describe was much more representative of synthetic diamonds in general. It was again a macle, consisting of two incomplete elongated octahedra joined together on the twin plane. Figure 9(a) is a view of the crystal corresponding roughly with those of the diffraction topograph Fig. 10 and the absorption topographs Figs. 11–13. The better developed faces are on the x-ray exit surface, nearer the observer. All other surfaces were quite irregular. The base was possibly a fracture surface. The surfaces far from the observer showed skeletal growth and were partly hallowed out. There was some evidence that solution had occurred. The crystal was opaque except for a few transparent patches close to some edges.

4.2. Diffraction Topographs

A number of projection topographs and section topographs taken of this specimen showed that the lattice perfection was greatly inferior to that of the specimen described in Sec. 3. Orientation contrast as well as extinction contrast contributed to the variation of density of the image; in parts of the specimen the orientation spread was of order 5°. The projection topograph Fig. 10 uses the twin plane reflection so that the whole crystal appears in the image. Intense concentrations of strain are seen in several regions, and lines of imperfection tend to run parallel with (110) directions. They appear related to the strings of inclusions in similar directions seen on the absorption topographs. We wish to draw attention to a sheet of imperfection running

Fig. 9. (a) Shape of imperfect synthetic diamond twin as it appears in topographs Figs. 10–12, (b) location of sheet of occluded impurity ABB'A' in top left corner of Fig. 9(a). Scale mark 100 $\mu$. 

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down from the left-hand top corner of the image; this is further discussed below.

### 4.3. Absorption Topographs

Figures 11 and 12 are a stereopair of absorption topographs taken with Cu Kβ radiation. A great number of absorbing particles is seen compared with the few shown in Figs. 7 and 8. The smaller individual particles appear generally globular, with diameters in a rather narrow size range, 2–7 μ. The larger particles appear to have been formed by the coalescence of strings of globules and they often lie parallel to (110) directions. Figures 11 and 12 are members of a series of absorption topographs taken at roughly 10° intervals, starting with the x rays parallel to the twin plane. Figure 11 is a left-eye view, making 32° with the twin plane. Figure 12 is the right-eye view, making the Bragg angle with the twin plane and thus corresponding to the projection topograph Fig. 10. The parallax between near and far inclusions is easily noticed in Figs. 11 and 12, but prints such as these cannot adequately portray the striking appearance of the original plates when examined under twin microscopes. The opaque inclusions then appear to be suspended around and within the nebulous form of the weakly absorbing diamond. Relative positions of the inclusions can be fixed accurately, and it is clear that most of them are at or very close to the crystal surface. The specimen was subjected to a 50-h etch in aqua regia and the absorption topographs repeated, but none of the absorbing particles disappeared or detectably diminished in size through this treatment. The more irregular surfaces of this specimen compared with that described in Sec. 3 would render an absorbing skin less readily detectable, but we found definite evidence for the presence of such a skin on some parts of its surface.

One might expect occlusion of impurity to occur at the re-entrant angles where the two components of the twin pair meet, but the absorption topograph taken with the x rays passing parallel to the twin plane gave no evidence of any extra absorbing material in the twin plane. The absorption topographs do show, however, that considerable occlusion of absorbing material has occurred elsewhere. The diffuse region of extra absorp-
tion seen in the top left-hand part of Fig. 11 becomes sharp and strong in Fig. 12. It is due to a thin sheet of finely divided material lying in the dodecahedral plane which contains the short edge AB indicated in Fig. 9(b). The depth within the crystal at which the sheet commences cannot be clearly seen, but we believe its commencement is related to the development of the type of (110) edge of which AB is an example; and there is no doubt that it represents the trace of the outward motion of this edge as the crystal grew. The history is idealized in Fig. 9(b) which shows an early position A'B', and the final position AB, of this edge. The sheet ABB'A' also corresponds with the sheet of crystal imperfection seen on the diffraction topographs and pointed out in Sec. 4.2.

Figure 13 is a Cu Ka absorption topograph with the crystal orientation the same as in Fig. 11. The change in absorption contrast of the impurity particles is consistent with their being largely composed of nickel. The diamond itself shows up more strongly in this topograph. A "light–dark" alternation of intensity is associated with edges and steps of the crystal. It is believed to be an x-ray refraction effect. A similar though weaker effect is seen at the edge of the shadow of the nylon support upon which the crystal is mounted.

4.4. Satellite Diffraction Pattern

This crystal gave quite strong satellite reflections. Their diffraction angles were consistent with the satellite structure lattice parameter, 3.539 Å, found by Lonsdale, Milledge, and Nave,* though no attempt was made to measure it accurately. A notable feature of the satellite pattern was the division of its reflections into two groups. One group was produced by relatively large crystallites, misoriented up to several degrees from the matrix. The other group arose from quite small crystallites closely parallel to the matrix. This division is seen clearly on the oscillation photograph about the 200 reflection, Fig. 14. The large spots extend along about 10° of the Debye–Scherrer arc whereas the spread of the fine-crystallite spots amounts to some minutes of arc at most. The oscillation range in this photograph was not large enough to encompass the forbidden diamond 200 reflection. There is some indication that the interplanar spacing of the fine crystallites is about 1 part in 10° smaller than that of the large crystallites.

5. DISCUSSION

Let us consider first the interpretation to be put upon the diffraction contrast effects found in the more perfect specimen. We are reluctant to refer to the fine radiating lines we see on section topographs as individual dislocations unless by taking a set of topographs of different Bragg reflections we can demonstrate that they obey the usual visibility rules of dislocations. In this specimen the presence of a great amount of surface strain and of considerable regions of internal imperfec-

![Image](Fig. 13. Cu Ka absorption topograph, angle of view same as in Fig. 11.)

![Image](Fig. 14. Satellite 200 reflections from imperfect specimen.)

* The nickel-rich surface film and the severe strain it produces in the underlying diamond layers are notable features of this specimen, but we cannot say more about the fineness of division of the absorbing material on the surface other than that its particles are less than $\frac{1}{4}$ or $\frac{1}{2}$ μ in diameter.

It has already been remarked that the large inclusion A does not appear to be an important source of dislocations. Solid inclusions of this size in diamond would
generate many dislocations by lattice closure errors. We therefore think that A was included in the diamond as a molten droplet, and we believe that the globular inclusions seen in both specimens were in general incorporated in this state. It may be a coincidence that the apparent nucleating point of the crystal is close to inclusion B, but the indication this gives of the presence of an element more x-ray dense than nickel leads us to speculate that it is associated with a particle that was solid above the nickel-carbon eutectic solidification temperature and constituted the mote upon which the diamond nucleated.

The question of resolving individual dislocations does not arise in the case of the less perfect crystal described in Sec. 4; this specimen is much too imperfect. It is, however, far from an "ideally imperfect" mosaic crystal since it still shows considerable extinction contrast. It is noteworthy that more contrast, i.e., more lattice distortion, is produced by the finely divided occluded impurity than by the individually resolvable nickel-rich particles. This is in accord with our conclusion that the latter were incorporated as molten droplets. It is fortunate from the point of view of mechanical strength of synthetic diamonds that the many relatively large inclusions do not greatly strain the matrix, and that the presumably rounded internal surfaces of the diamond surrounding the globules do not give rise to stress concentrations when the crystal is under an external load. It may possibly be that these inclusions inhibit the propagation of cleavage through the crystal.

The reason for the two classes of crystallite size exhibited in the satellite diffraction pattern is not clear. It is tempting to associate the large crystallites with the large absorbing inclusions, and the small crystallites with the finely divided absorbing material, but we have no proof of this relationship. The finely divided material, whether on the surface or in the crystal interior, may not contain crystallites of satellite structure large enough to give a recognizable diffraction pattern at all.

The thin sheet of finely divided nickel-rich material occluded under the edge AB is of some morphological significance, so we believe, and is further discussed below.

6. THE CUBE FACES OF SYNTHETIC DIAMOND

A remarkable feature of well-formed synthetic diamonds is the strong development of the cubic in addition to the octahedral forms, with the cube faces possessing a flatness never exhibited by the occasionally observed cube faces on natural stones. Bovenkerk has described the transition from octahedral, through cubo-octahedral, to almost pure cubic habit; higher temperatures favor growth as an octahedron, a fall in temperature induces the cubic habit. We wish to put forward an explanation for the development of cube faces and to suggest a connection between the type of impurity occlusion observed at the edge AB of the specimen described in Sec. 4 and the initiation of such faces.

The natural mode of diamond growth is by the addition of layers to the octahedral faces. If the rates of addition on all faces keep pace with each other, and nothing impedes the lateral extension of growth layers, a regular octahedron is produced, with four {111} faces such as e, f, g, h shown in Fig. 15(a) meeting in a point. A fluctuation in growth rates may allow faces f and h to gain a little over e and g, causing a short length of (110) type edge to form between e and g, as shown in Fig. 15(b). Subsequent developments may best be understood by considering the diamond structure as drawn in Fig. 16. This projection on the (110) plane shows the zig-zag chains of carbon atoms parallel to [110] (in the plane of the diagram) and parallel to [110] (normal to the diagram) which extend throughout the crystal. Traces of the natural {111} faces in the [110] zone are shown. Atoms at these faces have, ideally, one unsatisfied bond, directed normal to the face. Growth layers on these faces advance by the addition at growth steps of C-C zig-zags, such as 1 and 2. The growth layer is complete when it reaches an edge between adjacent {111} faces. Atoms such as 3 and 4 at the stable type of [110] edge between the (111) and (111) faces each have only one unsatisfied bond. On the other hand, if a [110] edge between the acutely inclined (111) and (111) surfaces is present [corre-

![Fig. 15. View parallel to [001] of diamond octahedron: (a) regular octahedron, (b) acute [110] edge developed, (c) cube facets developed.](image)

![Fig. 16. Projection on (110) of diamond structure, explained in text.](image)
sponding to the short edge between faces e and g in Fig. 15(b), the atoms terminating the crystal at this edge each have two unsatisfied bonds. This occurs whether the terminating row is made up of atoms in position 5 or in position 6. Such an edge is a likely site for the attachment of more carbon atoms, such as 7 and 8, with the resulting nucleation of new growth layers on the (111) and (111) faces. Consequently, under natural conditions, growth will be accelerated on the faces e and g, Fig. 15(b), so as to restore the balance and revert to the initial situation of Fig. 15(a).

Now let us consider what can happen in the presence of nickel atoms. The distance between carbon atoms along a row such as 6 differs by only 1% from the interatomic distance in a close-packed row of nickel atoms. A very favorable site for the adsorption of a row of nickel atoms is thereby presented, and such a row is shown attached at 9 in Fig. 16. Adsorbed nickel along this edge prevents the nucleation there of diamond growth layers, and the edge is not eliminated. What follows probably depends rather critically upon the growth conditions. If they strongly favor further diamond growth then the row 9 can be enveloped by growth sheets nucleated elsewhere; but the edge will be reformed at a higher level and will adsorb fresh nickel atoms. Thus a sheet of nickel impurity, which might develop appreciable thickness, is occluded in the (110) plane above the edge. This corresponds to the situation revealed in Fig. II and drawn schematically in Fig. 9(b).

Now let the ambient conditions become less favorable for diamond growth. Layers nucleated on the octahedral planes are either unable to rise above the level of the nickel obstacle at position 9 or are halted there sufficiently long so that nickel atoms become attached in place of carbon in the [110] rows alongside row 9, as shown, for example, at row 10. Growth on the (111) and (111) faces thus becomes truncated by an adsorbed nickel layer on the (001) plane. The crystal habit changes to that of Fig. 15(c), and ultimately to a cube.

Finally let us consider what surface topography might be expected on a cube face produced in the way we have proposed. On cube faces which have not been obscured by a thick film of solidified catalyst Tolansky4–6 has found an exceptional smoothness, no steps being detectable within the limits of the most sensitive phase-contrast technique, say 3 Å. Some of these faces, however, are found to undulate gently with slopes of the order of 0.01. On the other hand, both Tolansky and Bovenkerk7 report the occasional occurrence on cube faces of coarse steps having heights between 100 and 1500 Å, arranged concentrically, and, in some instances, forming a recognizable spiral.

We explain these observations as follows: The layer of adsorbed nickel atoms forming the developing (001) face (Fig. 16) expands laterally on the (001) plane, occupying sites which lie in the cube atomic planes of the diamond structure. The spacing of these planes is 0.89 Å. The packing of nickel atoms on one such plane closely reproduces that of a cube plane in fcc nickel and is presumably a low-energy configuration. Occasional jumps in the level of attachment of nickel atoms, to cube planes above and below the previous level, may be expected however. Thus a slight regional slope can be produced, but with steps of only 0.89 Å, well below the phase contrast resolution limit.

A different type of step is produced by a dislocation whose Burgers vector makes 45° with [001]. A single such step has a height of 1.78 Å. When it occurs in a nickel monolayer it exposes a strip of (111) diamond surface with a row of unsatisfied carbon bonds. Under growth-promoting conditions more carbon can be attached here and a growth spiral will form. The difficulty of growing fresh diamond over a nickel monolayer will cause succeeding diamond growth sheets quickly to overtake the lowest sheet. A rapid pile-up of individual steps follows which produces an easily visible macrostep.

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