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Interferometric Studies on Very Pure Silicon Carbide Crystals

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Optical multiple-beam interferometric studies are reported for over one hundred extremely pure, clear, colourless, transparent silicon carbide crystals. With one exception they are thin, effectively parallel-sided plates, having surfaces of very high interferometric quality. Most surfaces show regular steps or curvature and these may be due to slip and to buckling after cooling.

The spiral growths so common on the less pure common quality commercial silicon carbide crystals appear here on one crystal only. A defect goes right through the crystal plate and a pair of spirals develops simultaneously, one on the back and one on the front of the plate, from this common defect. Spiral steps measured interferometrically exhibit unusual anomalies which are discussed.

It is well known that a large proportion of even the best-formed silicon carbide crystals hitherto examined exhibit notable growth spirals on the basal pinacoid. These have been studied by many investigators, notably Verma (1951), Amelinckx (1951) and others. Not only have spiral growths been established, but in addition a variety of step heights have been evaluated by using the precision techniques of multiple-beam interferometry. It is now well known that these spiral growths afford a most comprehensive confirmation of the dislocation theory of growth put forward by Burton, Cabrera & Frank (1949) and later more extensively developed especially by Frank (1951).

Most silicon carbide crystals exhibit some colour, varying from a light shade of green, through to a brilliant black. Through the courtesy of Dr Knippenberg of Phillips, Eindhoven, we have available a collection of extremely pure crystals of silicon carbide. These were received as an incrustation of transparent colourless thin crystal plates (some hundreds), growing out towards the centre from an annular ring of friable graphite. The crystals (many of them several millimetres across) were all beautifully formed and each was clearly a single crystal, all being notable for their transparency and complete absence of any colour. They were clearly of much higher purity than the usual commercial silicon carbide crystals.

Since spiral growth formation is certainly linked to the presence of severe dislocation, and as included impurities must clearly encourage development of such screw dislocations, it was suggested to us by S. Tolansky that there was a considerable likelihood that these very pure silicon carbide crystals might indeed not exhibit any spiral growths at all, in contradistinction to the usual run of silicon carbide crystals. We have therefore carried through a multiple-beam interferometric study of the surface microtopographies of over a hundred of these pure crystals and this report summarizes the findings. The crystals studied were mostly of similar thickness, transparent plates of the order of a quarter of a millimetre thick, all plane, and exhibiting well developed pinacoid (0001) faces. In accordance with standard practice in this laboratory (Tolansky, 1948) the crystals were silvered (reflectivity >95%) and examined, first by phase contrast, then with multiple-beam Fizeau fringes (green mercury source) and in selected cases with white-light fringes of equal chromatic order. In specific instances information was obtained without silvering, and at times by the use of polarized light.

Observations

With one single and obviously an unusual exception, our examination of over 100 crystals failed to reveal any spiral growths at all. Now evidence has long since accumulated that the familiar spirals on silicon carbide have step heights normally several integral multiples of the 15 Å crystal spacing. The refined multiple-beam techniques and phase contrast techniques available in this laboratory are such that one could not fail to detect a spiral of step height of even one lattice spacing, *i.e.* of 15 Å. Thus it can be asserted with confidence that no spirals have gone undetected through lack of resolution.

Following a conjecture put to us by Tolansky that these pure crystal plates might be uniform like mica cleavages, *i.e.* show regions of molecular uniformity in thickness over large regions, our first experiments consisted in silvering both sides of a crystal and examining it with Fizeau fringes. Fig. 1 shows such an interferogram for a part of a crystal, and this is indeed the familiar uniform tint type of interferogram shown by high quality mica cleavages. This establishes, in accordance with long known conclusions, that the uniform tint areas shown reveal regions which are of uniform thickness, true to within a single crystal lattice spacing.

A step separating two uniform regions appears and is irregular in outline, but constant in height. Local variations appear as small areas with different tint.

It in instructive to compare this interferogram with a phase contrast picture (Fig. 2) taken for the face of this crystal plate. Phase contrast vividly reveals the diffracting step edge, but as we already know, gives no hint as to differences of height over the extended regions away from the edge. Comparison of Figs. 1 and 2 clearly establishes that the smaller tint features in Fig. 1 refer to elevations and depressions on the rear side of the crystal, *i.e.* that not shown in Fig. 2. (Note: The faint low-visibility narrow parallel fringes in Fig. 2 are spurious and arise from the internal reflector of the metallurgical microscope employed.)

A multiple-beam Fizeau fringe interferogram over the surface of which Fig. 2 is a part, is shown in Fig. 3. This is notable and curiously resembles the kind of topography shown by a cleavage of good quality mica. There are two broad regions separated by a growth edge, which is of course that shown so vividly in Figs. 1 and 2. The fringe definition is so good that it establishes that the surface quality is very smooth indeed, yet the surfaces are not flat. There is a marked cylindrical curvature over one half, and in various places small raised flat areas appear. The surface quality is exceptionally fine and it is abundantly clear that growth has taken place by plane sheet propagation. It is probable that the cylindrical curvature indicates some form of buckling which took place after completion of growth.

The step height measured both by Fizeau fringes and fringes of equal chromatic order is 770 ± 10 Å. Within limits of measurement it appears to be uniform in value along the whole length. It may be a growth step; alternatively it may be a slip-step occasioned by strain. Indeed the marked difference in curvature on either side favours the strain conjecture.

The close similarity between these growth surfaces and the cleavage faces of mica is surprising. Thus Fig. 4 shows the interferogram given by a complete face and without any doubt it very closely simulates many of the hundreds of mica cleavage interferograms frequently studied in this laboratory. As on mica the fringes are (*a*) very smooth, (*b*) show typical hills and dales, (*c*) reveal discontinuities which most closely simulate the cleavage steps on mica.

Although we clearly are dealing with a single crystal, the directions of the discontinuities are certainly not in crystallographic directions. It is not likely that they represent slip lines or cleavages. They are probably edges of growth fronts.

Fig. 5 is another example. Here a few small short steps are visible. In one case the step is inclined to the surface and gives the impression of emergence of a screw dislocation. However the notable overall feature is the fact that the surface is smooth yet 'wrinkled', exactly as is so often seen on very good mica cleavages.

Yet it is certain that these are growth faces, not cleavages, since the crystals cluster out in perfect formation from the graphite core. It is reasonable to conclude that these crystals grow into thin platelets by plane sheet propagation and then either mechanically buckle, or else show slip-step discontinuity There is not the slightest evidence on over a hundred crystals examined (excluding one) of any of the familiar spiral growth mechanism so characteristic of normal commercial silicon carbide crystals. Clearly this must be associated with the clarity and transparency, *i.e.* with the absence of occluded impurity which no doubt initiates the screw dislocations so common in commercial silicon carbide crystals.

Spiral growth

Out of the crystals examined (over a hundred) one only revealed spiral growth and this only over one small region (some 10% of the area, the remaining 90% of the crystal having clearly grown by plane sheet propagation as with all the others. The spiral feature is most exceptional and appears to differ from any previously reported, for spirals occur on both opposing faces of the plate and it is quite clear too that both originate at one and the same single defect region, which obviously goes right through the whole crystal plate and emerges on both sides. On the one side the spiral is circular, as shown in Fig. 6 and winds anti-clockwise. On the other side (Fig. 7) the spiral starts off circular but quickly tends towards a curvilinear hexagonal character and is also anti-clockwise.

By setting the microscope focus suitably it was possible to secure a photograph of both spirals simultaneously; this is shown in Fig. 8, which establishes unequivocally that both spirals initiate from one common defect which penetrates through the crystal. The region of origin is shown to advantage in the higher magnification picture (Fig. 9).


Fig. 1

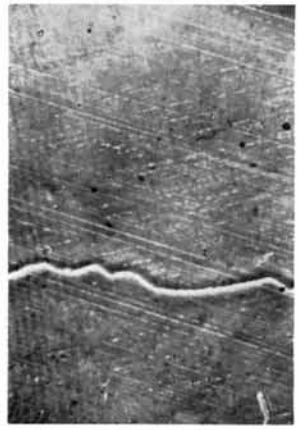
Fig. 2

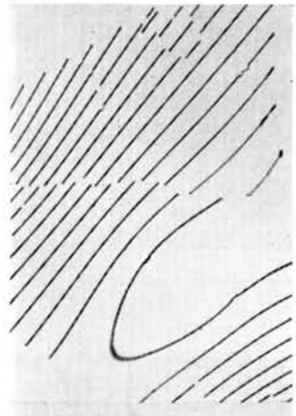
Fig. 3


Fig. 4


Fig. 5

Fig. 1. (×45) Multiple-beam fringes of equal tint through a very pure SiC crystal.
Fig. 2. (×50) Phase contrast photograph revealing a diffracting step edge.
Fig. 3. (×15) Multiple beam interferogram on the same crystal that appears in Figs. 1 and 2.
Fig. 4. (×48) Multiple beam interferogram on the surface of a very pure SiC crystal.
Fig. 5. (×48) Multiple beam interferogram on SiC, as in Fig. 4. This closely simulates a mica cleavage interferogram.

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Fig. 6

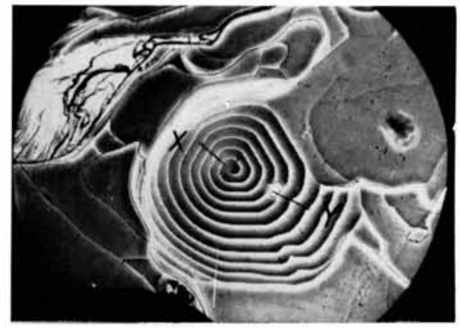

Fig. 7

Fig. 8

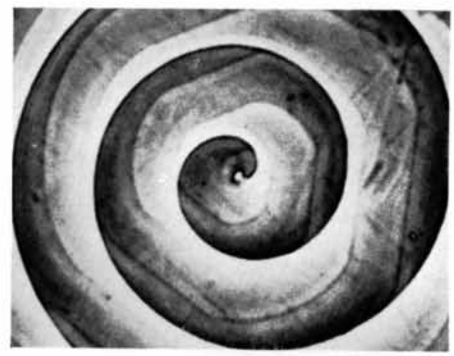

Fig. 9

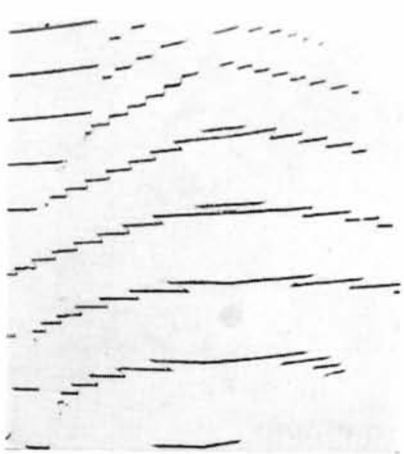

Fig. 10

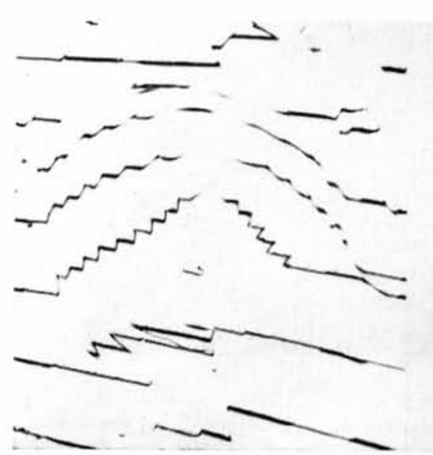

Fig. 11

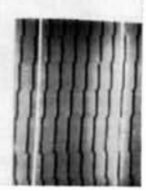

Fig. 12

Fig. 6. (× 50) Phase contrast photograph. One side of a spiral with viscular habit.
Fig. 7. (× 72) Phase contrast photograph of the other side of the same crystal region shown in Fig. 6.
Fig. 8. (× 60) Phase contrast photograph showing both spirals simultaneously.
Fig. 9. (× 108) Phase contrast photograph showing the region of the origin.
Fig. 10. (× 45) Multiple beam interferogram on the side of the crystal with circular spiral.
Fig. 11. (× 48) Multiple beam interferogram on the other side of the crystal of Fig. 10.
Fig. 12. Fringes of equal chromatic order across the peak of the spiral.

The geometrical irregularities in the spirals are largely to be attributed to their location close to the crystal boundary which has thus led to deformation in growth. As a consequence a curious anomaly emerges with regard to the distances between successive turns of the spiral. For in the direction marked Y the spacing decreases, whilst in that marked X it increases.

Multiple-beam interferograms were secured for the two spirals and examples are shown respectively in Figs. 10 and 11.

From such interferograms step heights could be accurately measured, and as a precaution confirmed with fringes of equal chromatic order of the kind shown in Fig. 12.

Graphical plots of the distribution of step heights across the peaks of the spirals are shown for the two cases in Figs. 13 and 14. These height distributions taken along the line XY are completely anomalous in comparison with earlier reported spirals on silicon carbide, for the step heights, although quite uniform from centre to X and from centre to Y, are in fact quite different on the two sides. Thus the steps from centre to X are 431 ± 10 Å whilst those from centre to Y are 542 ± 10 Å. This difference, some 111 Å, is of the order of 7 to 8 lattice spacings. On the other face the steps also show a similar anomaly. From centre to X it is 414 ± 10 Å and from centre to Y it is 470 ± 10 Å. The difference, some 56 Å, is about 4 lattice spacings. These increases take place in the same direction on both spirals.

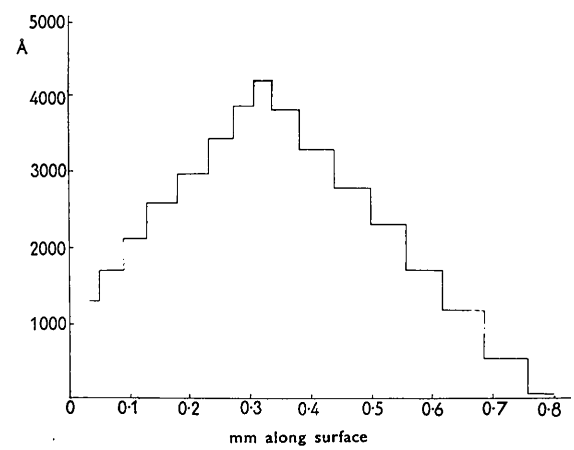

Fig. 13. Graphical plot of the distribution of step heights across the peak region of the circular spiral.

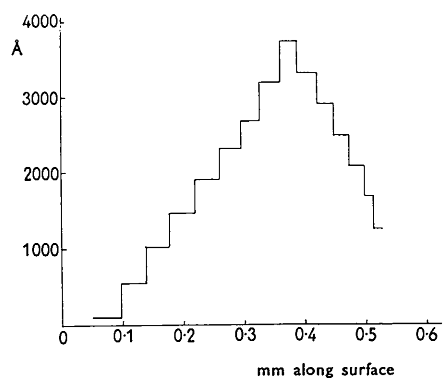

Fig. 14. Graphical plot of the distribution of step heights across the peak of the reverse side of the spiral.

Conclusions

It is clear that the hundred or so pure silicon carbide crystals studied all grow essentially through propagation of plane sheet growth. These were probably effectively parallel-sided, thin, stepped plates, which then subsequently buckled slightly on completion of cooling. The crystal surfaces show a high degree of perfection. Discontinuities appear and might be due to growth, or alternatively to slip.

In one instance only, a defect region appears which penetrates the whole thickness of the crystal and from either side spiral growth has taken place. This spiral region is not markedly thicker than the rest of the plate and clearly spiral growth has not been very much faster than plane sheet growth.

It is probable that the frequent occurrence of spirals on the more commonly available commercial silicon carbide crystals is largely due to inclusion of impurity defects which clearly lead simultaneously to a darkening colour and to frequency of screw dislocations which produce spiral growth.

The simultaneous appearance of two linked spirals, one on each face, appears to be recorded for the first time.

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