Optical microscopic, synchrotron X-ray topographic and reticulographic study of homoepitaxial CVD diamond

A.R. Lang*, A.P.W. Makepeace, W.B. Alexander, T. McCormick, P.E. Pehrsson, J.E. Butler

*H.H. Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol BS8 1TL, UK
MENC Electronic Materials Inc., St. Peters, MS 63376, USA
GeoCenters Inc., Fort Washington, MD 20744, USA
Naval Research Laboratory, Code 6174, Washington, DC 20375, USA

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Abstract

Surface topography and crystal-lattice perfection of homoepitaxial layers deposited by microwave plasma CVD on (0 0 1) and near-(0 0 1) facets polished on HPHT synthetic diamond are described. Optical microscopic techniques included birefringence, Nomarski and 2-beam interference. The synchrotron X-ray experiments comprised Laue topography plus a recently developed sensitive misorientation-measuring technique, reticulography. Two special circumstances enhanced information yield from the experiments. First, the substrate crystal was unusually strain-free and had a very low dislocation content. Second, epilayer growth had taken place in two stages, depositing thicknesses of 10 μm and 30–34 μm, respectively. This double deposition complicated the observations, but added features of scientific and practical interest. Epilayer cracking finally present had occurred almost entirely before the second growth stage. With assistance from quantitative data provided by reticulography, the X-ray diffraction properties of the substrate and epilayers are analysed. Lattice misorientations on the untreated lower surface of the substrate were only ~ 1 arcsec except close to growth-sector boundaries and dislocation outcrops. The final epilayer growth surface above areas where cracking in the first epilayer was absent or sparse exhibited near-perfect-crystal diffraction behaviour. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Understanding homoepitaxial chemical vapour deposition (CVD) growth of diamond on single-crystal diamond substrates is important.
scientifically and technologically. Scientifically, the growth of diamond exemplifies an extreme case in the sphere of crystal growth: a highly rigid lattice, high Debye temperature, and growth driven by direct reaction from the gas phase species [1]. Differences of lattice parameter between substrate and epilayer, consequent upon their differences in dopant and impurity content, cause stress and resultant extended defects. Technologically, single-crystal diamond growths can be useful in a wide variety of electrical and mechanical device structures, particularly when one can control the properties of the material by intentionally doping or introducing desired defects during the growth process.

For imaging surface morphology and assessing integrity of CVD diamond films the optical microscope and the SEM are well-established tools. The application of X-ray imaging methods to CVD diamond is a newer activity, which can be expected to expand. Different X-ray imaging methods are appropriate according to whether heteroepitaxially grown polycrystalline films or homoepitaxial films are the subject of study. When dealing with polycrystalline films, the techniques of optical microscopy, X-ray microradiography, microdiffraction and topography can be brought together to enable the diffraction properties of individual crystallites to be examined and correlated with morphological features of interest [2,3]. In the homoepitaxial case, the CVD films are essentially single crystals (of varying perfection) up to several mm² in area. Consequently, the experimental finesse that was needed in the earlier work for probing a particular area of film only some tens of micrometres in diameter was not essential in the work here reported. On the other hand, experimental complications arose in the present work resulting from the CVD film remaining attached to the substrate crystal. However, independent of whether single crystal or polycrystalline films are under investigation, the capabilities and speed of X-ray imaging experiments on CVD diamond are greatly enhanced when advantage can be taken of the high intensity and excellent collimation of X-rays produced by a synchrotron radiation source.

Two characteristics of homoepitaxial diamond films are noteworthy. First is a strong dependence of film morphology upon substrate orientation. In addition to differences between films grown on precisely oriented (1 1 1), (0 1 1) and (0 0 1) substrates, marked morphological differences appear between films grown on exact (1 1 1) or (0 0 1) surfaces and those grown on substrates polished some degrees off these planes [4,5]. The second characteristic is the prevalence of cracking in the epitaxial layer. Provided neither epilayer nor substrate is patently imperfect, this cracking is attributable to stress arising from difference between substrate and epilayer lattice parameters.

The investigation here reported was concerned with the surface morphology and state of lattice strain in homoepitaxial diamond deposited on (0 0 1) and near-(0 0 1) facets polished on a high-quality synthetic diamond. Various experimental techniques were employed, including optical microphotography combining transmitted and reflected illumination, birefringence and 2-beam interference microscopy. The X-ray experiments comprised conventional Laue topography plus a new, misorientation-measuring technique, reticulography [6,7]. A couple of special circumstances enhanced the information yield of the experiments. First, the substrate crystal was untypically strain-free and had a very low dislocation content. This was of great benefit towards extracting information from birefringence and X-ray diffraction observations that was relevant to epilayer properties. Second, the specimen had undergone two stages of epitaxial growth. Although this complicated the features observed, it generated phenomena of both scientific and practical interest.

2. The specimen

The drawing, Fig. 1, and optical micrograph, Fig. 2, show the specimen dimensions, including the size, shape and angular rotation about axis [1 0 0] from exact (0 0 1) orientation for the four facets comprising strip a of original surface, nominally indexed (0 0 1), and strips b, c and d that were deliberately bevelled on that surface. The substrate was a commercially-available, (0 0 1)-orientation plate of HPHT-grown synthetic diamond, optical type Ib (Sumitomo Electric Industries Co., Hyogo,
Fig. 1. Side view of the specimen plate, height 3.9 mm, maximum thickness 1.04 mm. Crystallographic axes adopted are [1 0 0] towards observer, [0 1 0] vertically downwards and [0 0 1] horizontally to the right. The face with epilayer growth (faceting right) is divided into strips a, b, c, d, orientations (0 δ 1), which are rotated towards [0 1 0] with δ increasing from a to d. The rotation clockwise about [1 0 0] from exact (0 0 1) is stated (in degrees) below the arrows normal to each facet strip; the accuracy is estimated as ± 0.1° for facets a and b, and ± 0.2° for c and d. The strip widths parallel to [0 1 0] are a, 1.0 mm; b, 1.5 mm; c, 0.16 mm; d, 1.24 mm.

Japan). The crystallographic orientation of the untreated, well-polished base of the specimen (nominal index (0 0 1)) was accurately established by combined optical and X-ray goniometry, and the rotations about [1 0 0] relative to the base surface of facet strips a-d were found by two optical goniometric methods, with reasonable agreement.

In synthetic diamond substrates of the type used in this work internal strains arise principally from dislocations and from inhomogeneous distribution of the most important impurity, which is nitrogen in singly substitutional form. X-ray topography and birefringence detect such strains sensitively, and in the present specimen they revealed an unusually low level of strain except for regions affected by proximity to the epilayers. Uniformity of colouration also showed that substitutional nitrogen content was quite uniform within the central (0 0 1) growth sector of the substrate crystal that occupied most of its volume. Concentration of substitutional nitrogen is assessed from the strength of the diamond’s infrared absorption at 0.14 eV, 1130 cm⁻¹, applying the currently accepted conversion rate of 25 ± 2.0 atomic ppm N(subst.) per cm⁻¹ absorption coefficient at 1130 cm⁻¹ [8]. From the FTIR spectroscopic measurement of absorption through the central area of the specimen, i.e. through facet strip b, the substrate N(subst.) content was assessed as about 133 atomic ppm, a typical value. The relation between absorption at 1130 cm⁻¹ and lattice parameter excess over that of pure diamond is known from synchrotron X-ray double-crystal diffractometric measurements on dislocation-free volumes of HPHT synthetic diamond [9]; this relation indicated that the substrate crystal’s lattice parameter was greater than that of nitrogen-free diamond by the fraction Δa₀/a₀ = 1.6 × 10⁻².
The diamond epilayers were grown by microwave-plasma assisted CVD. The two stages of growth took place under generally similar conditions but differed in duration and pyrometer readings. The common conditions were: microwave power 1.1 kW, pressure 40 Torr (5.3 kPa), gas composition 1.2% CH₄ in hydrogen, and total flow rate 500 cm³ per minute at STP. For epilayer 1, the duration was 6 h and pyrometer reading 985°C. Then growth was interrupted for Raman spectroscopy and optical examination. For epilayer 2, the duration was 14.5 h (followed by slow shut-down) and the pyrometer reading during growth was 967°C. The thickness of epilayer 1 was about 10 μm, and that of epilayer 2 was in the range 30–34 μm as measured on strip b. By polishing away growth that had spread over on to the side surface (1 0 0), that surface was rendered optically smooth, permitting total epilayer thickness to be measured directly by microscopy, and it was found to be \( \approx 42 \mu \text{m} \) on strip b. A pronounced Schlieren effect was manifested at the substrate/epilayer 1 interface, but the epilayer 1/epilayer 2 interface was not visible optically. (In synthetic diamonds step-wise changes or strong gradients of refractive index can be distinctly observed by Schlieren effects [10].)

### 3. Synchrotron X-ray topography and reticulography techniques

The X-ray experiments were conducted at the Synchrotron Radiation Source, Daresbury Laboratory, UK, using Station 7.6, which is located 80 m from the source. Since the source produces a continuous spectrum of radiation, the conditions of Laue-pattern recording apply, and Bragg reflections from lattice planes will occur at any angular setting of the specimen crystal with respect to the incident beam. In the present work back-reflection Laue images were recorded from either the epilayer-covered face (0 0 1) or from the untreated base surface (0 0 1), for both conventional surface-reflection topographic imaging and for reticulography. The specimen was oriented to back-reflect with \( 2θ = 135° \). In most experiments symmetrical back-reflections were recorded. Then, for the lowest-order reflection, 0 0 4, the wavelength is 0.165 nm. Contributions to topograph images from the harmonic reflection, 0 0 8, \( λ = 0.082 \text{ nm} \), were in most cases unimportant due to lower source intensity and lower photoplate X-ray absorption efficiency at this shorter wavelength.

Regarding reticulography, only brief accounts of this technique have been published [6,7], so an outline of the experimental arrangement is appropriate here. The technique provides a simple but sensitive method of measuring lattice misorientations point-by-point, applicable when a highly collimated polychromatic incident beam is available. A fine-scale, X-ray absorbing mesh is placed in the diffracted beam, close to the crystal. Precision electro-deposited Ag and Au meshes, periods 63.5 and 34 μm, respectively, were used in the present work. The mesh splits the diffracted beam into an array of individually trackable microbeam elements. Direction differences between microbeams are measured from their relative shifts within images of the array, the 'reticulograph', when the mesh-to-photoplate distance, \( M \), is changed. Fig. 3(a) shows the general arrangement and 3(b) indicates schematically how mesh image distortions are produced by variations in lattice tilt over the surface imaged. For reference, an undistorted image of the mesh can be obtained using the minimum value of \( M \) (which is about 1.5 mm in the apparatus specially constructed for reticulography) if the specimen is not more than moderately distorted, or from an optical micrograph of the mesh in the case of highly distorted crystals. In high-angle back reflections, direction differences between diffracted microbeams are twice the orientation differences between the crystal elements reflecting them, both in the plane of incidence and normal to it. Thus a single back-reflection reticulograph provides all needed data for a two-dimensional map of misorientation vectors over the Bragg plane concerned.

### 4. Observations

#### 4.1. Optical microscopy

Fig. 2 shows how strongly the surface texture and cracking density depend upon surface orientation.
Most of the epilayer surface on \( a \) is low relief, showing linear features parallel to \([1 1 0]\) and \([1 0 0]\) on a 10 \( \mu \)m length scale. However, towards the top edge of the specimen (as viewed in Fig. 2) the texture coarsens and comprises contacting conical growth hillocks up to 150 \( \mu \)m in diameter, slopes \( \approx 1^\circ \), and almost devoid of 4-fold symmetric features.

All of facet \( b \) has a low-relief surface. Low-elevation growth hillocks, highly elongated parallel to \([0 1 0]\) and tapering in that direction, spread down from the \( ab \) boundary. The whole of \( b \) is covered by fine steps trending roughly parallel to \([1 0 0]\), heights not more than a few tens of nm, and laterally persistent. A description of facet \( b \) introduces the topic of cracking, and the overall cracking pattern will now be considered.

Photographs of the specimen taken between epilayer growth stages 1 and 2 show that almost all the specimen’s presently-visible cracks had already appeared before stage 2. On facet \( b \) the cracks are sparse except within a distance of about 0.3 mm from the \( bc \) boundary; and the pattern of cracking, and the tapering towards \([0 1 0]\) of individual cracks, indicate that cracks have spread into facet \( b \) from facet \( d \). Close examination by Nomarski differential interference contrast and by 2-beam interferometry confirms that both facet \( b \) and facet \( c \) are free from crack outcrops, with the insignificant exception of just three short crack segments lying somewhat off \([1 1 1]\), one segment crossing the \( bc \) boundary. The fine-scale surface topography of facet \( b \) is entirely unaffected by the cracks below in epilayer 1, which have all been completely overgrown. These cracks have well-defined upper edges at the epilayer 1/epilayer 2 interface. Long crack segments under facet \( b \) do not visibly extend into the substrate crystal. A representative field where some such extension has occurred is shown in the micrograph, Fig. 4. Focus is on the upper edges of cracks in the field centre, the focus level being 34 \( \mu \)m true depth below the present surface. The cracks lie on \([1 1 1]\) imperfectly, and crack surfaces are irregularly stepped. Lack of straightness in crack upper edge images arises in part from this cleavage unsmoothness but also because the crack upper edges reveal the undulations of epilayer 1 surface topography (and in fact provide the
Fig. 4. Transmitted-light micrograph of upper edges of cracks in a region of facet strip b 1 mm from the right-hand edge and 1.6 mm from the bottom edge of the specimen as viewed in Fig. 2. Field width 0.25 mm. Focus level is 34 μm below the present specimen surface. (Hence the finely stepped structure of the present surface is completely out-of-focus). The two parallel cracks running from upper right to lower left lie on (1 1 1). As for cracks running from upper left to lower right, the upper lies on (1 1 1) and the two lower lie on (1 1 1).

only evidence on this topography, in consequence of the optical homogeneity of epilayers 1 and 2). In the Fig. 4 field the greatest optically visible crack penetration below the epilayer 1/epilayer 2 interface is ~ 40 μm. (Here and elsewhere fracture depth may exceed that optically visible by an unknown amount.)

The surface topography on strip c was considerably rougher than on b and a. It suggested step flow ‘downhill’ relative to the (0 0 1) plane, towards [0 1 0], with arrays of dentate steps facing that direction.

More rough still was the surface of facet d, and its topography showed strong evidence of step flow towards [0 1 0], as evidenced in the interaction between crack outcrops and surface profile that was observed. In contrast to facets b and c, on facet d only the shallower cracks have been grown over, in general. Deeper cracks (which range up to 150 μm in visible depth) form obstacles that have been only locally bridged. Where not bridged, the crack outcrop is marked by an asymmetric groove. There, the side of the crack closer to strip c forms a scarp up to ~ 10 μm high (maximum measured height 13 μm), whereas on the opposite side of the crack there is a ~ 50 μm-wide strip of surface running parallel to the crack where the surface slope relative to (0 0 1) is about 10° less than the average on facet d (≈ 15°). This strip has been ‘starved’ of growth steps flowing towards [0 1 0] due to the barrier formed by the crack outcrop.

4.2. X-ray topographs and reticulographs

Fig. 5(a) and (b) are conventional synchrotron X-ray Laue-image surface reflection topographs of the face of nominal index (0 0 1). Diffraction contrast is produced dominantly by the 0 0 4 reflection, wavelength 0.165 nm. Contributions from the harmonic 0 0 8 reflection are not specifically identifiable, and can be taken as insignificant in Fig. 5. The diffracted beam is back-reflected 45° upwards from the horizontal specimen-to-source axis. Hence Bragg-reflecting (0 0 1) planes are viewed from a direction rotated 22.5° upwards from [0 0 1]. Some minor vertical compression of images of facets a–d results from viewing them with this moderate obliquity. However, these geometrical factors do not impede easy recognition in Fig. 5(a) and (b) of major facet-strip-dependent image differences, in particular those related to crack density and depth.

Areas densely and more deeply cracked, which include the lower part of facet b as well as all of c and d, generate very strong diffraction contrast due to the overlapping and interaction of crack-associated deformation fields. As the specimen-to-plate distance, P, is increased, the contrast pattern coarsens, deformations due to the major cracks becoming dominant. However, lattice orientation differences not greater than a few arcminutes can account for the pattern geometry observed.

The well-separated cracks in the upper part of facet b serve best towards understanding crack images quantitatively. Their basic characteristic is that each individual crack is imaged by a pair of parallel lines of strong diffracted intensity into which diffracted rays are focussed. The magnitude, W, of the line separation increases with increasing P, but at a rate less than proportional to P. This
Fig. 5. Back-reflection synchrotron X-ray topographs of the epilayer-covered face of the specimen. Reflection 004, 2θ = 135°. Specimen view corresponds to Fig. 2, with some vertical compression due to non-perpendicular viewing, the compression factors ranging from 0.92 for facet strip a to 0.79 for strip d. The plane of incidence, trace vertical, is (1 0 0); direction [0 0 1] lies in this plane, 22.5° below the diffracted beam direction. Note that in consequence of some epitaxial growth having spread over on to all side faces of the specimen, these sides are loci of strained layers whose lattice curvature produces a dispersion of diffracted-beam directions. This causes unsharp imaging of the periphery of the surface viewed in Fig. 5 where it meets the (1 0 0), (0 1 0) and (1 0 0) side faces. (All topographs are recorded on Ilford L4 nuclear plates, emulsion thickness 25 μm, and, printed as positives, i.e. greater blackness corresponds to greater X-ray intensity). Specimen-to-plate distances, P, are: (a) 35 mm; (b) 185 mm.

means that the strong lines are foci of rays whose mean divergence decreases as P increases. The corresponding mean mutual tilts of Bragg-plane elements reflecting into the lines are given by W/2P, and are found to be 0.1°, 0.06° and 0.03° when P is 35, 85 and 185 mm, respectively. Measurements on topographs taken with P < 35 mm have a similar trend. These observations are discussed further in Section 5.

The pattern of focussings and intensity concentrations in caustics seen in the images of facets a and b can be regarded as the reflection counterparts of optical transmission patterns through 'bathroom window' glass, being produced by reflection from lattice planes deformed in an undulatory or rippled fashion. The pattern coarsens with increasing P as reflections from area elements of lesser curvature come into focus. Over a substantial fraction of facets a and b the diffraction contrast pattern geometry is suggestive of growth hillocks elongated parallel to [0 1 0], but on the present surface of facet a such elongated hillocks do not appear. Furthermore, the large-diameter conical growth hillocks on the present surface near the top edge of facet a described in Section 4.1 do not have corresponding contrast features on the X-ray topographs.

Images of the (0 0 1) surface were also recorded using other Bragg reflections, 0 2 6, 1 1 3 and 1 1 5. For recording the last two, the specimen was rotated in its own plane by 45° to change the plane of incidence from (1 0 0) to (1 1 0). These additional topographs did not reveal any noteworthy new features not seen in the symmetrical 0 0 4 reflections.

Reflection topographs of the substrate base surface (0 0 1) were informative both on lattice defects in the substrate crystal bulk and on lattice deformations in proximity to the epilayers on the remote face. The 0 0 4 surface reflection topograph image, Fig. 6, is affected similarly to the images in Fig. 5 by lattice curvature in the strained layers on the side
Fig. 6. Symmetric reflection X-ray topograph of the untreated polished base surface of the substrate diamond. A 45° rotation of the specimen about [0 0 1] has changed the X-ray incidence plane from (1 0 0), its orientation in Fig. 5, to (1 1 0). The resulting slightly oblique diagonal view of the specimen has produced slight rhomboidal distortion of the cubic symmetry of the (0 0 1) face. In this figure the trace of (1 0 0) is vertical as in Fig. 5, but the topograph has been printed left-to-right for comparison with all other figures, which view the (0 0 1) face. Topograph recorded with specimen-to-plate distance $P = 467$ mm.

faces of the specimen. These give rise to the pattern of caustics surrounding the image in Fig. 6 that render the bounding edges of the face ill-defined. However, a short distance inwards from the face edges, well-defined diffraction-contrast images of growth-sector-boundary outcrops on (0 0 1) are visible. Reference to X-ray topographs, cathodoluminescence topographs and growth sector maps of polished (± 0 0 1) surfaces on other (0 0 1)-orientation plates of HPHT synthetic diamond identify the growth sectors concerned [10,13]. In the present instance it is sufficient to report that the greater part of the crystal volume is composed of growth sectors of form {1 0 0}, the large central (0 0 1) sector having an outcrop width of $\sim 3$ mm on the (0 0 1) face. Also, there is no diffraction-contrast evidence of any impurity zoning within this growth sector. Dislocations, probably not more than a few tens in number, outcrop close to the centre of the (0 0 1) face. They run roughly parallel to [0 0 1], threading the crystal from one polished cube face to the other. Of particular present interest is the form taken by images of the deformation fields due to cracks in epilayer 1 and substrate below. In Fig. 6 the double-line images that are seen in Fig. 5 are replaced by single lines, and these single-line images are visible in reflection topographs of the (0 0 1) face taken over a wide range of distances $P$. For forming images of deformations close to the remote face of the specimen, the harmonic reflection 0 0 8 is important. Absorption in the double passage through the substrate crystal to the (0 0 1) face and back attenuates the $\lambda/2$ harmonic wavelength 30 times less than the 0.165 nm wavelength.

Reticulographs were recorded from the (0 0 1) surface using the 0 0 4 and 1 1 5 X-ray reflections, and from the (0 0 1) surface with the 0 0 4 reflection, in each case working with two or more mesh-to-plate distances, $M$. The reticulograph chosen for display here, Fig. 7 was taken under the same diffraction conditions as one of the conventional topographs reproduced, Fig. 5(b). This similarity facilitates relating contrast features appearing on the conventional topograph (together with the features seen on the corresponding optical micrograph, Fig. 2) with the distortions (if any) in the mesh image on the reticulograph. Reference to Fig. 3(b) shows that the reticulograph image of an area where the Bragg-reflecting lattice planes have convex curvature towards the observer will exhibit an enlarged mesh spacing, whereas concave curvature decreases the spacing. Thus, provided distortions of the mesh image are small (a condition usually achievable by choosing a sufficiently low value of $M$ when recording the reticulograph), simple mirror-reflection optics can be applied to map lattice curvature point-by-point on the specimen by measuring the deviations of mesh cell image dimensions from their true values. Excluding from consideration in Fig. 7 the regions close to the (1 0 0), (0 1 0) and (1 0 0) sides of the specimen, and facets $c$ and $d$ where some uncertainty in the delineation of mesh cell images arises locally, the overall regularity of mesh cell periodicity is noteworthy. Some regions where quantitative analysis of the reticulograph image is straightforward and informative are the finger-like areas enclosed between
Fig. 7. Reticulograph of the epilayer-covered specimen face. Diffraction conditions the same as for topograph Fig. 5(b). Mesh-to-plate distance $M = 150$ mm, mesh period 63.5 $\mu$m.

the pairs of lines of enhanced intensity that are associated with individual cracks in the upper part of facet $D$. In Fig. 7 it can be seen that along the median strips of the 'fingers' there is a slight rhombohedral distortion of mesh cell images, their diagonals being expanded perpendicular to the finger axis. This corresponds to cylindrical Bragg-plane curvature about the finger axis, convex towards the observer. Rough values of the bend radius are 2.5 m measured on Figs. 7 and 2.2 m on another reticulograph, taken at the same distance $M$, but using the 34 $\mu$m period mesh.

Reticulographs of the untreated (0 0 1) surface of the substrate crystal were highly informative. A reticulograph, 0 0 4 reflection, taken with the high $M$ value of 1050 mm still showed good definition of 63.5 $\mu$m period cell images (at least with regard to horizontal mesh bars, for which image sharpness benefited from the small X-ray source dimension in the vertical plane). It was demonstrated that within some areas up to 0.6 mm diameter on the (0 0 1) face parallelism of the Bragg-reflecting planes was maintained to 1 arcsec. Higher variations of lattice orientation, ranging up to 5–10 arcsec, only occurred locally, for example in proximity to the face periphery, to growth sector boundaries, and to dislocation outcrops.

5. Interpretation and discussion

The first topic for discussion is the noteworthy difference in character of images of individual cracks depending upon whether they are recorded in reflections from (0 0 1) or from (0 0 1). The diagram, Fig. 8, explains how this difference arises. To begin, a couple of remarks on the diffraction physics are relevant. On synchrotron radiation Laue topograph images of concavely-curved Bragg-reflecting planes, the increased diffracted intensity comes in part from focussing of diffracted rays, which occurs with all magnitudes of curvature, and in part from progression towards imperfect-crystal diffracting behaviour, which develops as lattice-plane curvature strengthens. Under the diffraction conditions applying in the present experiments, augmentation of diffracted intensity as a result of lattice imperfection will become increasingly prominent as the curvature radius decreases below about 1m, and hence is important in the crack images under consideration. The second remark is to make clear that the topographs offer no diffraction evidence for incoherence between substrate and epilayer 1. If incoherence existed on relatively large areas of interface, exceeding some tens of micrometres in diameter, then the appearance of X-ray moiré fringes on those areas would be expected. No such fringes appear, either in symmetrical or oblique reflections.

In Fig. 8 the 004 back-reflected rays are drawn as originating from points such as $x_1$ and $x_2$ lying on strongly curved surfaces close to the crack, with $R_1$ and $R_2$ indicating the different tilts of the local Bragg-plane normals at $x_1$ and $x_2$, respectively. Similarly back-reflected rays are produced from points on the left of $C$, related by mirror symmetry in the vertical plane through $C$. From the diffraction point of view, it is immaterial whether points $x_1$, $x_2$ etc. are regarded as lying on level 2 or on level 3. The feature of importance is that in proximity to $C$ at the levels concerned the local cylindrical lattice curvature radii (e.g. $R_1$ and $R_2$) decrease, and their tilts increase, as $C$ is approached both from left and right. Denoting lattice displacement parallel to [0 0 1] by $y$, and magnitude of distance from $C$ by $x$, a simple physically plausible relation describing the Bragg-plane profile is $y = ax^n$. The
Fig. 8. Model of lattice deformations that accounts for the difference between the topograph images of cracks recorded in reflection from (0 0 1) and from (0 0 1). Lattice tilts exaggerated about a thousandfold. The crack C, which penetrates epilayer 1 and some distance into the substrate below, is for simplicity drawn lying in the vertical plane, whereas observed cracks lie in \{1 1 1\}, albeit imperfectly. To conform with the lattice tilt exaggeration, the crack C is sketched widely open. In fact, on micrographs such as Fig. 4 the crack opening is not resolved. Important levels are numbered, viz. 1, specimen top surface; 2, epilayer 2/epilayer 1 interface; 3, epilayer 1/substrate interface; 4 and 5, levels in the substrate at depths, of order 50 and 100 μm below level 3; and 6, the specimen bottom surface. Relative thicknesses of epilayers 1 and 2 are shown to scale, but the greater thicknesses below are compressed.

measurements of W and P described in Section 4.2 do not provide numerical values of \(a\) and \(n\), but are consistent with \(n\) being about unity. These mirror-related lattice-plane curvatures on either side of C produce the pair of caustics that are intersected by X-ray topograph plates placed at different distances \(P\). On the other hand, for the 0 0 4-reflected rays, diffraction contrast is generated when X-rays entering the substrate from below penetrate to levels of downward-facing concave curvature such as 4 and 5, and these levels reflect rays to a single line image over a wide range of distances \(P\).

Next to be discussed is interpretation of reticulographs such as that shown in Fig. 7. The high regularity of the mesh image over facets \(a\) and \(b\) has already been noted. Excluding the minor distortions associated with the ‘fingers’ on facet \(b\), analysed in Section 4.2, and distortions close to the face edges, the range of orientations of Bragg planes responsible for the mesh image is estimated as about 6 arcsec, taking a traverse across the face. This is probably no greater than in the underlying substrate crystal, judging from the reticulographic assessment of its (0 0 1) surface. The sharpness of individual mesh shadow edges shows that Bragg-plane tilts on a spatial scale of order 10 μm cannot be greater than a few arcseconds. These properties of the reticulographs indicate Bragg-reflection by a single sheet of coherent crystal whose diffraction behaviour does not substantially fall short of that of a perfect crystal. With such perfection the depth-extent of Bragg planes building up the reflection is limited to the X-ray extinction distance, or not much greater. Under the diffraction conditions applying the extinction distance for the 0 0 4 reflection is about 5 μm, much smaller than the X-ray absorption distance, 540 μm. It is therefore concluded that reticulographs of the (0 0 1) face record high lattice perfection in the surface layers of the specimen, at least in the uppermost fraction of epilayer 2. No plausible case can be made for any other interface or stratum below the present surface being responsible for the main reticulograph pattern. The reasoning is as follows. If epilayer 2 were so imperfect as to be unable to register a mesh image by surface reflection at any of the \(M\) values used for recording reticulographs then its integrated reflecting power would be high enough to blanket topographs taken of both (0 0 1) and (0 0 1) surfaces, and all the orientation contrast and diffraction contrast details actually observed would be masked. Certainly the near-independence of mesh-images of facet \(b\) from crack-related Bragg-plane curvatures below it demonstrates that interfaces at levels 2 and 3 do not generate this main pattern. The slight arching of present-surface Bragg-planes over the axes of ‘fingers’, quantified in Section 4.2, is an understandable consequence of the deformation.
below involving epilayer 1 and the substrate. Note, however, the smallness of the uplift that this sensitive reticulographic curvature measurement implies. Assuming an arch span as great as 0.2 mm, then the surface elevation at the arch apex is only \( \sim 4 \) nm, too small to be detectable in the optical interference micrographs of the areas concerned, which are dominated by the step structure described in Section 4.1.

The final interpretation topic considered here concerns the images of uncracked areas as they appear in the conventional topographs Fig. 5(a) and (b). X-ray topographic contrast from these areas diminishes when \( P \) is reduced to low values, such as 7.5 or 5 mm, supporting the geometrical-optical explanation of contrast in terms of diffracted-ray focussing that was advanced in Section 4.2. Also reported there was the general absence of correlation between the X-ray images and the present-surface contour detail on facet \( a \) and the uncracked area of \( b \). Furthermore, it is significant that where the double-line X-ray topographic images of cracks under facet \( b \) appear they completely supplant the characteristic pattern of adjacent areas. This shows that both patterns are generated at similar sub-surface levels. Stresses in the substrate/epilayer 1 interface rather than in the epilayer 2 interface are the likely sources of deformation, in view of the lattice parameter mismatch believed to occur at the former interface. Inhomogeneous strain at this interface would be produced in the following way. Epilayer 1 growth hillocks developing on the substrate will exert greatest tensile stress in the substrate surface where the hillock is thickest. Hence areas of upwards convex curvature of the interface will be centred on hillock summits. The lines along which developing growth hillocks meet are natural loci for incorporation of defects (such as molecular-scale non-diamond structures) in the growing epilayer, and the locally increased epilayer lattice spacing resulting therefrom will reduce the overall coherence stress and reverse the lattice curvature locally. In this way an undulatory or rippled contour of Bragg-reflecting planes lying in and close to the substrate/epilayer 1 interface can be produced.

All observations are consistent with growth stage 1 depositing homoepitaxial crystal with smaller lattice parameter than the nitrogen-containing substrate crystal, which induced tensile stress in the epilayer and the resulting stress-relieving cracking after growth stage 1. This was followed by growth stage 2, depositing material of similar lattice parameter to that of stage 1 and overgrowing the shallower cracks seen in strips \( b \) and \( c \). The interface between epilayers 1 and 2 is indistinguishable except where optically delineated by mouths of cracks in epilayer 1 that have been overgrown by epilayer 2.

Several studies of homoepitaxial CVD diamond films by X-ray diffraction methods have been reported. A very sensitive multicrystal diffractometer technique, capable of separating lattice orientation variations from lattice parameter variations, has been applied to specimens similar to that described here [14]; but since the measurements averaged over an area of 5 mm by 0.5 mm on the specimen they are not directly comparable with topographic imaging experiments. Conventional X-ray topography of homoepitaxial films separated from their substrates has been performed [15]. The films were relatively thick (70–80 \( \mu \)m), and their angular ranges of reflection (0.3°–0.75°) and imperfection content were high compared with the material illustrated here. Synchrotron X-ray section topographs [16] and CuK\( \alpha \) radiation double-crystal topographs [17] of homoepitaxial films on their substrates have been published, but the specimens were too densely cracked to allow observations of the phenomena that attracted attention in the present work. By combining a high-resolution topographic imaging capability with a wide range of sensitivity in quantitative misorientation measurement, the reticulographic technique is eminently suitable for studying homoepitaxial CVD-grown diamond. In particular, the evidence from reticulography concerning high perfection of the stage 2 growth that has successfully bridged cracks in the stage 1 growth on substrate strips \( b \) and \( c \) suggests adopting a 2-stage-growth route for achieving thick, crack-free epilayers.

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