

computing time is required for these calculations for non-periodic specimens if dynamical scattering and all electron optical parameters are included. The approximations outlined above make possible the simulation of these images and diffraction patterns within reasonable computing times. They therefore allow the possibility of least-squares refinement between computed images of trial structures and experimental many-beam images of defects.

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Use of Lattice Imaging in the Electron Microscope in the Structure Determination of the 126R Polytype of SiC

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Lattice fringes corresponding to (00.3) spacings of the 126R polytype of SiC have been obtained in the electron microscope. A number of 10.1 reflections were allowed to pass through the objective aperture to obtain the lattice fringes showing a periodicity of $\sim 105 \text{ \AA}$ (one third of the *c* parameter of the hexagonal cell) and each unit-cell block is subdivided into smaller blocks corresponding to the spacings of 6H, 15R, 21R etc. The stacking sequence of these blocks has been used to work out the detailed structure of the 126R polytype. It has been suggested that the lattice resolution technique in conjunction with X-ray diffraction is likely to prove a useful method for determining difficult polytypic structures.

Introduction

There are a number of substances like SiC, ZnS, CdI₂ etc. which are known to crystallize in many unique periodicities and crystal structures. SiC polytypes are characterized by the number of Si–C double layers and their stacking sequence in the unit cell. Out of the various polytypes, 6H is the most commonly occurring structure followed by 15R and 4H in order of frequency of occurrence. A unique feature of the high-period polytypes of SiC is that their unit cells are built

up by stackings of the unit cells of more common structures of SiC like 6H, 15R, 4H etc. For example the structure of the 39H polytype of SiC expressed in Zhdanov notation is (33)₂ 32(33)₂ (32)₂ which clearly shows that its unit cell consists of four unit cells of 6H and three unit cells of 15R stacked in a sequence represented by the sequence of 33 and 32 in this symbol (Azuma, Ohta & Tomita, 1963).

When a high-period unit cell is dominantly built up of units of one small-period structure, its structure determination becomes comparatively simple. However

a high-period structure which is not based on any common modification becomes exceedingly difficult to work out. The present communication describes the structure determination of such a polytype of SiC (126R) with the help of lattice-imaging technique in the electron microscope.

The 126R polytype of SiC was discovered by Verma (1957) and attempts to work out its structure since then have proved unsuccessful. This was primarily because its structure is not based on any single common modification and no clues are available for guessing trial structures.

Structure determination

Based on usual considerations employed in the structure determination of SiC polytypes (Verma & Krishna, 1966) nearly 150 structures were postulated out of which only one structure gave a rough match with the observed intensities (Singh, 1967).

The direct methods developed by Tokonami (1966) and Farkas-Jahnke (1966) were not attempted in view of the limitations of these methods (Gomes de Mesquita, 1968) which are likely to be as cumbersome as the trial-and-error method for an unusual structure like 126R.

Use of lattice imaging in structure determination

The lattice imaging of a number of extremely large period polytypes including that of the 126R of SiC has been reported recently (Dubey, Singh & Van Tendeloo, 1977) and it has been suggested that the details in the lattice images can possibly be correlated with the crystal structure. Since each reflection contributes a sinusoidal distribution of charge density whose wave fronts coincide with the family of lattice planes giving rise to that reflection, the lattice image is formed by superposition of all the sinusoidal distributions corresponding to the reflections used in the image formation. The sinusoidal distributions of charge density contributed by few $10.l$ reflections have been depicted in the form of traces of $(10.l)$ planes in Fig. 1. It may be seen that the traces of all the lattice planes superimpose at points 3.078 \AA apart along the a axis and such superpositions occur at intervals of one third of the c repeat period in the hexagonal cell. Normally this would give rise to pronounced dotted fringes parallel to the a axis separated at $\sim 105 \text{ \AA}$. However since a resolution of $\sim 3 \text{ \AA}$ was not possible in the present experimental set up, one observes uniform straight fringes. The smallest distance resolved in the present study was 10 \AA . Furthermore, there are other positions in the $\sim 105 \text{ \AA}$ interval where sinusoidal

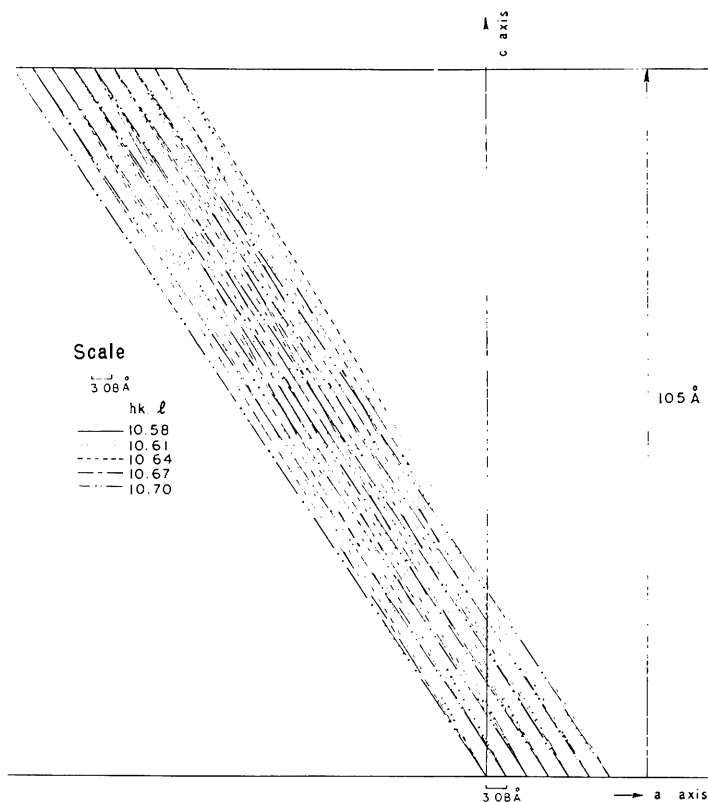


Fig. 1. The sinusoidal distribution of charge density contributed by few $10.l$ reflections of 126R polytype is depicted in the form of traces of $(10.l)$ planes.

distributions superimpose less efficiently along lines parallel to the a axis (Fig. 1). The superpositions of charge density with proper phases along these lines give rise to subsidiary fringes which divide the $\sim 105 \text{ \AA}$ interval into smaller blocks. Therefore the pronounced fringes separated at $\sim 105 \text{ \AA}$ (Fig. 2) essentially represent the lattice resolution of c planes of the 126R polytype. The lattice image contrast may bear a close correlation with the crystal structure provided that (Allpress & Sanders, 1973; McConnell, Hutchison & Anderson, 1974) (1) it is possible to select an objective aperture which excludes those beams which are severely affected by spherical aberration and still include many reflections to contribute to the image, (2)

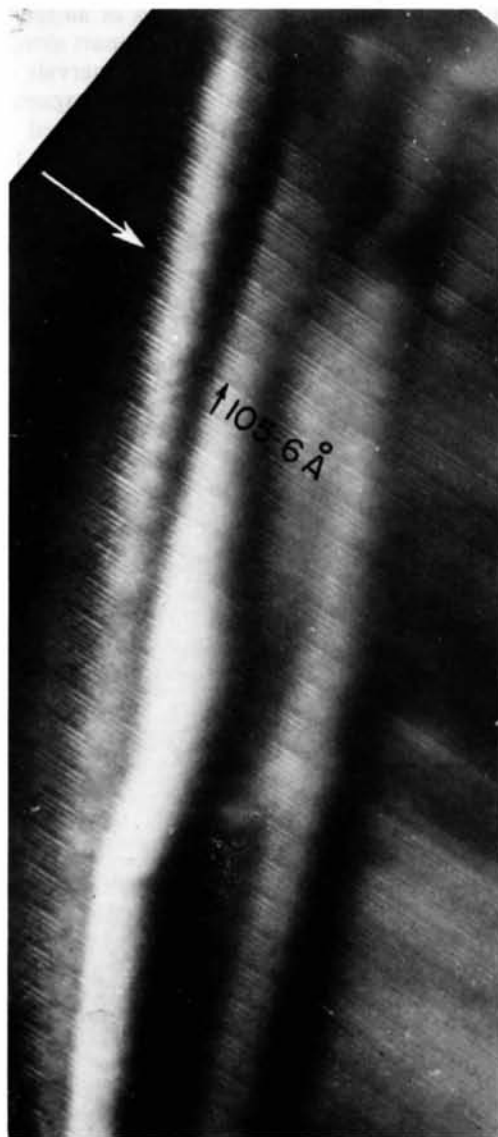


Fig. 2. Direct lattice imaging of 126R, SiC; a periodicity of 105.6 Å is observed. The arrow indicates the edge of the crystal foil.

lattice planes to be resolved are exactly oriented parallel to the beam and (3) the crystal foils are thin enough to approximate to phase gratings. The first two conditions are exactly satisfied in obtaining the lattice image shown in Fig. 2, while the third condition is presumably satisfied only at the edges (marked by the arrow in Fig. 2) of the crystal foil. It was therefore felt that subsidiary fringe separations in a unit-cell block of $\sim 105 \text{ \AA}$ may be correlated with the constituent structure blocks of commonly occurring polytypes.

The lattice periodicity of $\sim 105 \text{ \AA}$ is subdivided into different blocks of approximately 20 Å (8 layers), 17.6 Å (7 layers), 15 Å (6 layers) and 12.6 Å (5 layers) in the way represented in Fig. 3, which is slightly different from that reported earlier (Dubey, Singh & Van Tendeloo, 1977). On the basis of the majority of the structures worked out, blocks of 7-layer thickness are most likely to be 34 or 43, those of 6-layer thickness to be 33, and those of 5-layer thickness to be 32 or 23 in Zhdanov notation. For the lone 8-layer block 53 and 44 were taken as the possible stacking sequences. In this way 29 (twenty-nine) possible structures were postulated out of which (53 43 32 23 33 33 23)₃ in Zhdanov notation was found to give a good match with the observed intensities (Table 1). The calculated intensities were corrected for Lp factors. The structure (53 43 32 23 33 33 23)₃ in Zhdanov notation may be written in ABC sequence as follows:

*ABCABCACABCACBABCACBCACBABCACBAB
CACBABCACABCACBABCACBCACBABCBCB
ACABCACABCACABCACBABCACBABCACBABC
BCABCACABCACBABCACBABCACBACB| A....*

Atomic parameters derived from this are:

42 Si atoms at 0, 0, tz with $z = 1/159$ and
 $t = 0, 3, 7, 9, 12, 15, 18, 22, 25, 28, 31, 34, 37, 41,$
43, 46, 48, 52, 56, 58, 62, 66, 68, 72, 74, 78, 80,
82, 86, 89, 92, 95, 97, 101, 103, 105, 107, 111,
113, 117, 119, 123;

42 Si atoms at $\frac{2}{3}, \frac{1}{3}, \frac{1}{3}$ plus the above coordinates;
42 Si atoms at $\frac{1}{3}, \frac{2}{3}, \frac{2}{3}$ plus the above coordinates and
126 C atoms whose coordinates may be obtained by
simply adding p to the z coordinates of the 126 Si
atoms where

$$p = \frac{3}{4 \times 126} = \frac{1}{168}$$

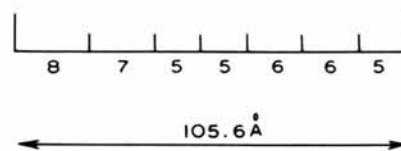


Fig. 3. Schematic representation of the stacking sequence of smaller blocks in the unit cell of the 126R polytype of SiC.

Table 1. *Calculated and observed relative intensities for the structure 126R*

<i>l</i> in 10. <i>l</i>	Calculated intensities	Observed intensities*	<i>l</i> in 10. <i>l</i>	Calculated intensities	Observed intensities*
1	0.0	<i>vvw</i>	2	0	<i>vvw</i>
4	0.1	<i>w</i>	5	0	<i>a</i>
7	0.0	<i>vvw</i>	8	0.1	<i>w</i>
10	0.2	<i>w</i> > 10.4	11	1.4	<i>ms</i> \approx 10.67
13	0.1	<i>w</i>	14	0	<i>a</i>
16	9.2	<i>s</i>	17	4.4	<i>ms</i> > 10. $\overline{11}$
19	0.1	<i>w</i>	20	2.6	<i>ms</i> < 10. $\overline{17}$
22	0.4	<i>w</i>	23	39.7	<i>vs</i>
25	53.3	<i>vs</i>	26	6.8	<i>ms</i>
28	0.1	<i>w</i>	29	0.1	<i>vw</i>
31	0.2	<i>w</i>	32	0.3	<i>w</i> > 10.10
34	23.0	<i>s</i> < 10.25	35	0.1	<i>w</i> < 10. $\overline{32}$
37	29.1	<i>s</i>	38	2.1	<i>ms</i> < 10. $\overline{20}$
40	68.9	<i>vs</i> > 10.25	41	484.0	<i>vs</i>
43	1.2	<i>ms</i>	44	32.5	<i>s</i> > 10.37
46	5.8	<i>ms</i>	47	1.2	<i>ms</i>
49	11.6	<i>ms</i> > 10.46	50	0.1	<i>w</i>
52	4.0	<i>ms</i>	53	0.2	<i>w</i> > 10. $\overline{50}$
55	3.2	<i>ms</i>	56	0.3	<i>w</i>
58	0.2	<i>w</i> \approx 10.10	59	3.0	<i>w</i> \approx 10. $\overline{32}$
61	24.0	<i>s</i> \approx 10.34	62	90.3	<i>vs</i> > 10.64
64	75.7	<i>vs</i> > 10.40	65	16.8	<i>s</i> < 10.34
67	1.4	<i>ms</i>	68	0.1	<i>w</i>
70	0.1	<i>w</i>	71	0.8	<i>w</i> > 10. $\overline{56}$
73	0	<i>vvw</i>	74	0.7	<i>w</i> < 10. $\overline{71}$
76	0	<i>vvw</i>	77	1.2	<i>ms</i>
79	0.1	<i>w</i>	80	0.4	<i>w</i> > 10. $\overline{56}$
82	2.1	<i>ms</i>	83	0.1	<i>w</i>
85	24.0	<i>s</i>	86	3.2	<i>ms</i>
88	0.1	<i>w</i>	89	1.2	<i>ms</i>
91	0	<i>vvw</i>	92	0.9	<i>w</i> 10. $\overline{71}$
94	0	<i>vvw</i>	95	0	<i>vvw</i>
97	0	<i>vvw</i>	98	0	<i>vvw</i>
100	0.3	<i>w</i>	101	2.6	<i>ms</i>
103	2.0	<i>ms</i>	104	0	<i>vvw</i>
106	0.2	<i>w</i>	107	0	<i>vvw</i>
109	0.3	<i>w</i>	110	0.5	<i>w</i>
112	0	<i>vvw</i>	113	0	<i>a</i>
115	0.1	<i>w</i>	116	0	<i>a</i>
118	0	<i>vvw</i>	119	0	<i>a</i>
121	0	<i>vvw</i>	122	0	<i>a</i>
124	0	<i>vvw</i>	125	0	<i>a</i>

* *vs*, *s*, *ms*, *w*, *vw*, *vvw*, *a* stand for very strong, strong, medium strong, weak, very weak, very very weak and absent respectively.

Discussion

The intuitively suggested correlation (Dubey, Singh & Van Tendeloo, 1977) between the lattice image contrast and the crystal structure of the 126R polytype of SiC has been found to be true, as the inferred sequence of smaller blocks is proved to exist in the real structure. The lattice imaging technique has been rarely employed to work out a complete structure (Allpress, Iijima, Roth & Stephenson, 1973) as in the present case. Therefore lattice imaging seems to be a possible aid in conjunction with X-ray diffraction for working out crystal structures, particularly of polytypic crystals having hundreds of atoms in the unit cell. The lattice-imaging

technique has been found more suitable than X-ray diffraction methods (Dubey, Singh & Van Tendeloo, 1977) for determining extremely large periodicities, and under suitable conditions seems also to be useful in determining such structures, particularly for polytype structures which are not based on commonly occurring structures and are otherwise difficult to work out.

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A Phase Transition in a 3D Growth-Disorder Model

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It is shown that a model of growth disorder, describing the way in which substitutional disorder can be introduced into binary solid solutions at growth, exhibits a phase transition. In a particular case it is shown that the distribution in 2D sections of the crystal corresponds to the simple pair-interaction Ising model on a triangular lattice.

Introduction

In recent papers (Welberry & Galbraith 1973, 1975; Welberry 1977*a,b*) models of the way in which substitutional disorder can be introduced into crystals at growth have been described. So far, however, these models have merely enabled distributions of binary variables (representing two molecular species) to be produced in two dimensions (2D) from given sets of somewhat arbitrary 'growth probabilities'. Our present aim is to put this work on a more realistic footing by extending the models to three dimensions (3D) and in addition to relate the 'growth probabilities' more directly to the forces involved when the molecular species interact at the crystal surface.

It is important at the outset to emphasize the distinction between the disorder produced by the type of growth process described here and a more general type of disorder which we shall refer to as dynamic disorder. In the latter we imagine the crystal to consist of molecules of different species which can rearrange themselves at temperatures below the melting point to achieve a minimum free-energy configuration, while we imagine growth disorder to arise in situations where a molecule once embedded in the crystal surface is subject to energy barriers sufficiently high that the possibility of subsequent rearrangement may be neg-

lected. While the dynamic-disorder situation involves energy equilibrium over the whole 3D crystal, the growth-disorder process only involves equilibrium within the surface layer. A model that has been used extensively for describing systems involving energy equilibrium over the whole crystal is the nearest-neighbour Ising model for which the solution in 2D is known (Onsager, 1944) and for which a considerable amount of information in 3D is available from approximate methods (see Domb, 1974).

It has been shown (Enting, 1977*a*; Welberry, 1977*a*) that the 2D growth-disorder models previously described are equivalent to more general 2D Ising models on which restrictions have been imposed on the values of the parameters in the energy function (Hamiltonian). It has become apparent that these restrictions remove from the particular 2D Ising model just that character of the model essential for the occurrence of a phase transition at a finite temperature. In fact it appears that 2D growth-disorder models give rise to lattice distributions that are little more than compatible 1D distributions in different directions (see Welberry, 1977*a*; Enting, 1977*b*). In extending the growth-disorder models to three dimensions it is of prime interest whether the character of the disorder produced by these models is sufficiently removed from that of the 3D Ising model that a phase transition