JOURNAL OF MATERIALS SCIENCE 7 (1972) 687-693

Development of the Surface Topography on Silica Glass due to Ion-Bombardment

I. S. T. TSONG, D. J. BARBER
Department of Physics, University of Essex, Colchester, Essex, UK

Mechanically polished fused silica surfaces have been bombarded by energetic Ar^+ ions and the development of surface topography examined by scanning electron microscopy and by transmission electron microscopy of direct carbon replicas in order to study the parameters affecting the surface finish such as charging, angle of ion incidence and rotation of the specimen. A theory based on a simple model of initial surface unevenness on a microscopic scale is proposed to explain the observed surface features due to ion-bombardment.

1. Introduction

The preparation of thin foils of non-metallic materials by ion-bombardment for transmission electron microscopy is now an established method and has been discussed previously [1, 2]. During the process of ion-thinning, the specimen is rotated in its plane about an axis through its centre and its two surfaces are simultaneously bombarded by Ar^+ ions from opposite directions at incident angles of about 70 to 75°. (We take 0° as normal incidence.) The purpose of rotation is to even out the non-uniformity of the ion beam and to minimise other effects such as surface relief and faceting on single crystals etc. When the ion-thinned specimen is examined under an optical reflection microscope the surface is often found to exhibit hummock-like features. This surface topography gives rise to changes in electron absorption in the thin foil and therefore is visible in transmission electron-micrographs as a mottled background, as shown in fig. 1. The chief objective of the present work is to investigate how this surface topography is developed during ion-bombardment. Silica glass was chosen as the target material because it is single-phase and non-crystalline, which make results easily interpretable.

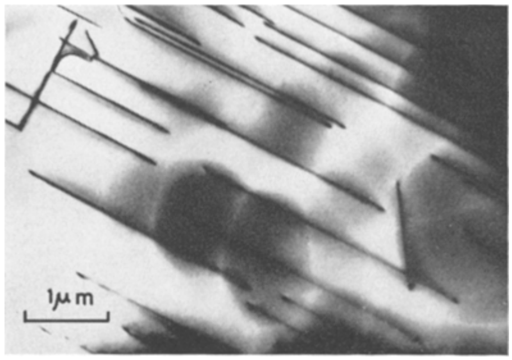

Figure 1 Ion-thinned specimen of hortonolite, an olivine mineral, showing dislocations and hummock-like surface topography visible as mottled background.

2. Experimental

The construction of the ion-source and its operating conditions are described elsewhere [1, 3, 4]. The experimental set-up is shown in fig. 2. The axes of the ion-source and the electron gun (for electrically neutralising the surface) are at right angles to each other. Surfaces of fused silica mechanically polished to plate-glass finish were bombarded by Ar^+ ions at an applied voltage of 6 kV at various incident angles from 0 to 85° and at an ion current density of about 200 $\mu A\ cm^{-2}$. The ion beam produced by the simple ion source is not monoenergetic and has a broad energy spectrum [5]. The silica disc specimen, 12 mm in diameter and 3 mm in thickness, was glued on to a specimen holder which was mounted on the spindle of a motor (10 rpm) so that the specimen could be rotated if desired during ion-bombardment. Typical bombardment times were 1 to 3 h. A charged-particle oscillator type source [6] was also employed as an alternative ion-source to ensure

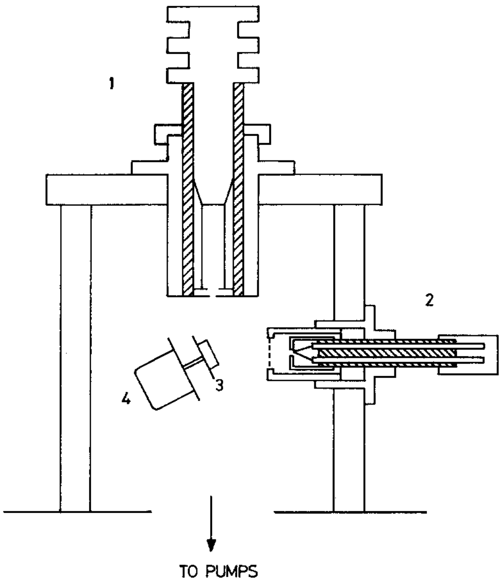

Figure 2 Schematic diagram of the ion-bombardment unit. 1. simple hollow anode ion-source, 2. electron gun, 3. specimen, 4. motor.

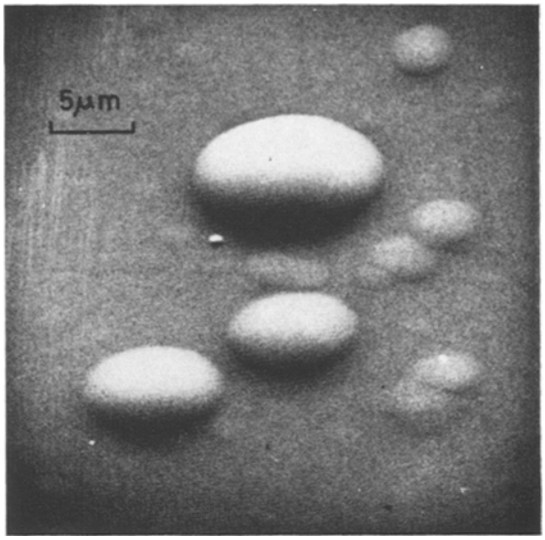

Figure 4 Scanning micrograph of the hummocks on an ion-bombarded silica surface. Incident angle = 75°.

that the observed surface features were not peculiar to one particular type of source.

3. Results

The development of surface topography in the form of hummocks was observed during ion-bombardment (figs. 3, 4 and 5). This kind of surface feature is consistent with the mottled background which appears in the micrographs of ion-thinned specimens. The results are summarised below:

1. The hummocks on the fused silica surface are created only when the specimen is rotated. They are not present if the specimen is stationary, whatever the angle of incidence.

2. The greatest number of these hummocks occur at angles of ion-incidence equal to the angle corresponding to maximum sputtering yield. In fused silica this angle is approximately 75° for Ar^+ bombardment [7].

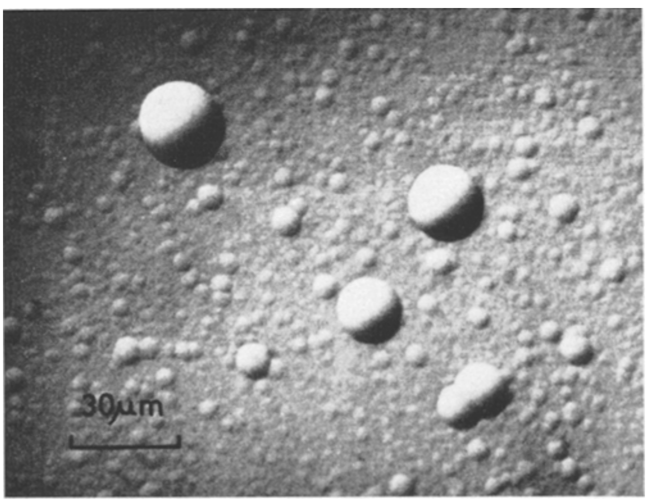

Figure 3 Optical micrograph of the surface topography of silica after 3 h bombardment at 75° incidence with rotation of the specimen.

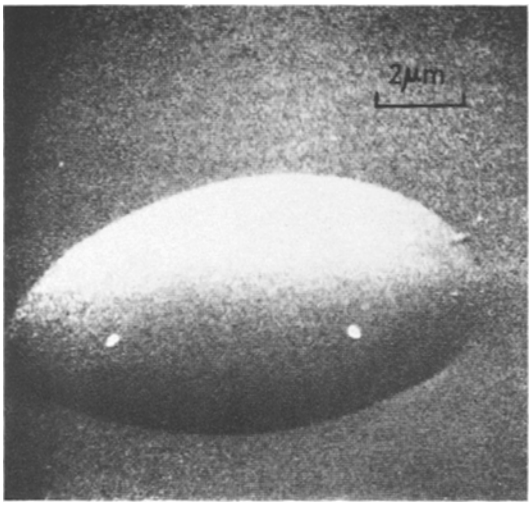

Figure 5 Scanning micrograph of a surface hummock.

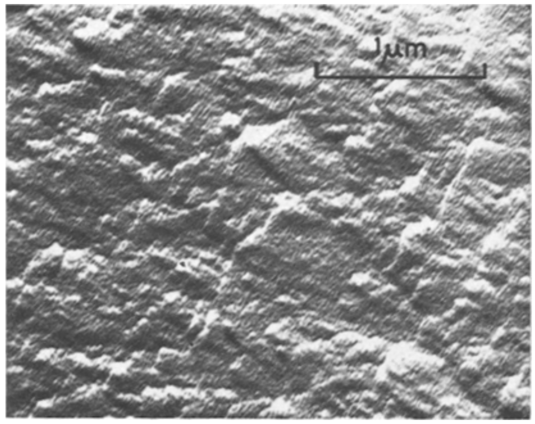

Figure 6 Transmission micrograph of a direct carbon replica of an ion-bombarded silica surface without rotation. Incident angle = 75°.

3. At angles of incidence away from 75° fewer hummocks are created, and at angles near normal incidence there are virtually none.

4. Charging of the surface does not appear to be the cause since this surface feature is present even if the surface is neutralised by electrons.

5. The hummocks become larger as sputtering continues. The increase in size is directly proportional to the sputtered depth.

6. It can be shown by partial masking of the target that the hummocks do not protrude above the original surface, indicating that they are not the result of a growth phenomenon but of some sort of sputter-protection.

7. The positions of the hummocks do not change during continued sputtering.

8. The hummocks often tend to congregate at surface scratches.

9. When the specimen is stationary during bombardment, a fully etched appearance somewhat like a sand-blasted surface is observed for angles of incidence near 75° (fig. 6). This surface feature is absent for angles near normal incidence.

10. Under the same bombardment conditions, identical development of surface topography is observed using the charged-particle oscillator source. This is perhaps not very significant because this source also produces a broad energy ion-beam and the ion-current density is also low ($\sim 100\ \mu A\ cm^{-2}$).

4. Discussion

The most notable aspects about the surface feature are (1) the fact that the hummocks only appear when the specimen is rotated, and (2) the fact that the number of these hummocks is much greater at grazing incidence than at normal incidence. These two facts seem to eliminate the idea that their formation may be due to small dirt particles, resting on the surface, that protect the underlayers from being sputtered [8]. It is also unlikely that this is the so called "blistering" effect due to intense radiation damage as described by Primak and Luthra [9] because such damage is highly improbable at the ion-energy range in the present work. Moreover, in contrast to Primak's report, exfoliation of the hummocks was never observed. Wehner and Hajicek [10] have shown that in certain cases of sputtering, singly distributed foreign atoms can surface-migrate together to form protecting nuclei which then lead to a formation of cones. But in the present case the dependence of hummock formation on specimen rotation and on angle of incidence appears to suggest that this is not the responsible mechanism. So we have to look for some other explanation.

We shall proceed to demonstrate how the observed surface feature can be explained in terms of a model based on the initial surface unevenness on a microscopic scale. Similar treatment concerning surface topography due to sputtering has been reported by various workers [8, 11, 12]. We assume that surface erosion is due to sputtering only. Other processes such as surface-diffusion and redeposition of sputtered material are ignored.

Let us consider two intersecting planes (fig. 7).

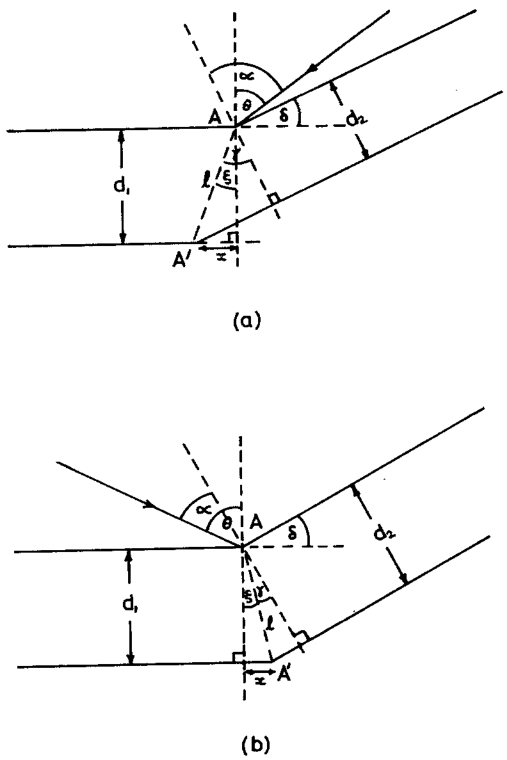

Figure 7 Movement of the intersection of two planes due to sputtering.

We take the plane to the left of the intersection A to be a flat surface so that the normal to this plane is the line of observation and is also parallel to the axis of rotation. The plane to the right of A we take as a deviation from flatness and it makes a small angle δ with the flat plane. After a certain time of bombardment, let d_1 be the sputtered depth on the left of intersection A, d_2 be the sputtered depth on the right of A, l be the distance advanced by A, and x be the sideways movement of A from the normal of the flat plane.

In fig. 7a, we can write

$$d_1 = l \cos \xi \qquad (1)$$

$$d_2 = l \cos \gamma = l \cos (\xi + \delta) \qquad (2)$$

$$x = l \sin \xi \qquad (3)$$

Also we have

$$d_1 = \frac{\Phi}{n} \cos \theta \, S(\theta) \, t \qquad (4)$$

$$d_2 = \frac{\Phi}{n} \cos \alpha \, S(\alpha) \, t \qquad (5)$$

where Φ is the number of ions per second striking unit area of surface normal to their direction, n is the number of atoms per unit volume of target material, $S(\theta)$ is the sputtering yield for incident angle θ expressed as the ratio atoms/ion, and t is the time of bombardment. Also we note $|\theta - \alpha| = \delta$.

From these five equations we find

$$\frac{x}{d_1} = \frac{\cos \delta \cos \theta \, S(\theta) - \cos \alpha \, S(\alpha)}{\sin \delta \cos \theta \, S(\theta)} \qquad (6)$$

In fig. 7b we proceed as above and we obtain

$$\frac{x}{d_1} = \frac{\cos \alpha \, S(\alpha) - \cos \delta \cos \theta \, S(\theta)}{\sin \delta \cos \theta \, S(\theta)} \qquad (7)$$

This is identical to equation 6 except for the change of sign. So the sign of equation 6 determines whether the intersection A moves to the left or right of the line of observation. In our convention we choose positive for moving left and negative for moving right for the intersecting planes in fig. 7.

Consider the numerator of the rhs of equation 6. Since we are dealing with well-polished surfaces, δ is usually very small. Hence $\cos \delta$ can be taken to be equal to 1.

If $x = 0$, i.e. there is no sideways movement of the intersection during sputtering, then

$$\cos \theta \, S(\theta) - \cos \alpha \, S(\alpha) = 0 \qquad (8)$$

One solution is $\theta = \alpha$, i.e. a perfectly flat surface. However, if the angle of incidence is near normal, then for any given ion-energy approximately equal to E_a*, the energy required for closest approach in a head-on collision [8] ($E_a \simeq 12$ keV for Ar^+ on silica), the sputtering yield can be written as

$$S(\theta) = K \sec \theta \qquad (9)$$

where K is a constant. For the sputtering curve of silica glass (fig. 8), this relation holds true for incident angles $\theta = 0°$ to $\sim 40°$. If the incident angle of the ions is in this range, we obtain once again equation 8 which means there is no movement of the intersection of planes. This theoretical prediction agrees with experimental observations showing that for incident angles from 0 to 40° the sputtered surface is as smooth as the original surface regardless whether the specimen is rotated or not.

For incident angles near θ_p, the angle corresponding to maximum sputtering yield, $\cos \theta \, S(\theta)$

* $E_a = 2 E_r (Z_1 Z_2)^{7/6} (M_1 + M_2)/M_2 e$, where E_r is the Rydberg energy, 13.6 eV, e is 2.718, Z_1 and Z_2 are atomic numbers of ion and atom respectively and M_1 and M_2 are the corresponding mass numbers.

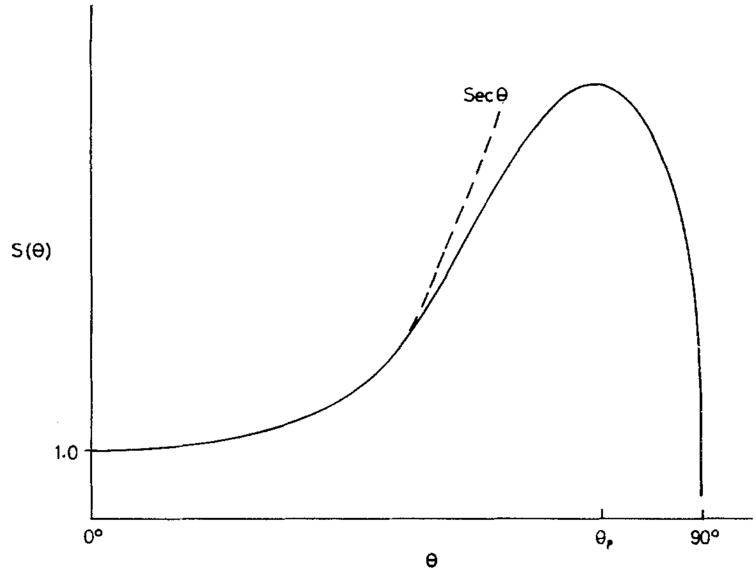

Figure 8 Sputtering yield of silica glass as a function of the angle of ion-incidence.

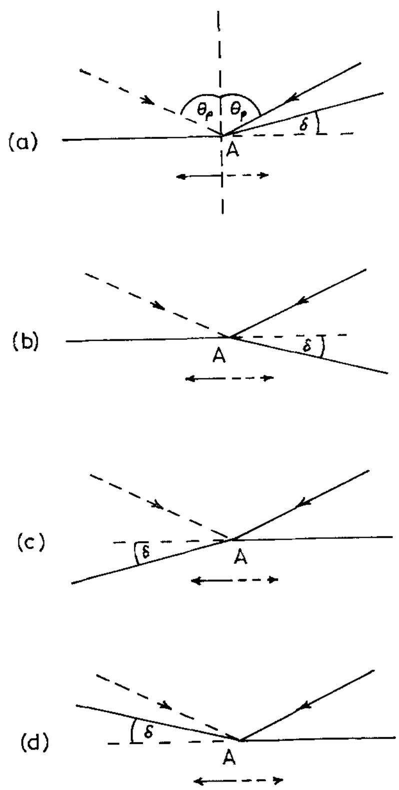

Figure 9 The direction of the sideways movement of the intersection from the normal of the flat plane. Full and dotted arrows indicate the directions of movement due to ion incidence shown by full line and dotted line respectively

$-\cos\alpha\, S(\alpha)$ can be either positive or negative depending on whether $\theta < \alpha$ or $\theta > \alpha$ as shown in fig. 7a and b respectively. This is because in this region of the sputtering curve (fig. 8), $S(\theta)$ increases rather more slowly than sec θ and actually decreases for angles greater than θ_p. So there is movement of the intersection of planes. In fact the direction of the movement of the intersection of a flat plane and an inclined plane due to ion-incidence at angles near θ_p can be summarised in fig. 9. Taking the surface as a whole, the movements of the undulations with respect to each other probably result in a sand-blasted appearance of the surface (fig. 6) when the specimen is stationary during bombardment at incident angles near θ_p.

If the specimen is rotated in its plane, the incident angle θ remains unchanged throughout the rotation while the other at the intersection varies continuously back and forth from α to β on a hummock-type irregularity on the surface (fig. 10). For simplicity, let us consider the case where $\theta = \theta_p$. At the point of the intersection A the sputtering yield to the right of A, i.e. the hummock side, as the specimen is rotated will appear as in fig. 11. We can therefore take an approximate effective sputtering yield $S(\psi)$ for the hummock side of the intersection A. There are two values of ψ (the effective mean angle of incidence averaged across the hummock) which will give $S(\psi)$ on the sputtering yield curve, either $\psi < \theta_p$ or $\psi > \theta_p$. If we consider the effect of rotation is equivalent to the ion beam precessing around the intersection A at an angle θ_p to the normal of the flat plane, it can easily be shown that $\psi > \theta_p$ is the solution. Therefore $\cos\theta\, S(\theta) - \cos\psi\, S(\psi)$ is always positive. By our convention this means that the size of hummock always increases at incident angles near θ_p in agreement with experimental observations. Since $\psi > \theta_p$ also implies reflection of ions from the hummock surface (which means there are probably more ions arriving on the flat surface), the hummock will appear to grow in height as well.

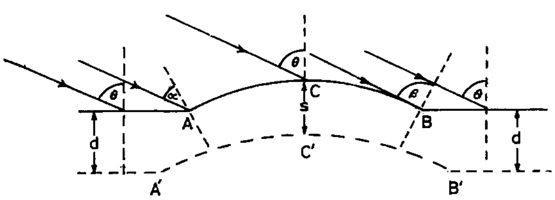

Figure 10 Growth of hummock due to ion-bombardment at incident angles near θ_p with rotation.

Similarly the above treatment can be applied to show that a hummock-like surface can be formed along a surface scratch (fig. 12a) for incident angles near θ_p plus rotation, but a polishing mark (fig. 12b) without raised sides will relax to a flat plane. Also when the above argument is applied to angles of incidence greater than θ_p, one would expect a polishing effect on the surface in agreement with experimental evidence but the sputtering rate at this angle of incidence is, of course, very slow due to reflection.

Figure 12 (*a*) Surface scratch, (*b*) polishing mark.

Preliminary experiments with glass-ceramics and alumina appear to suggest that if the initial surface of a material can be mechanically polished to a smooth finish, then bombardment at an incident angle of 40° will produce a virtually hummock-free surface. However, for materials which contain surface microcracks or other defects which cannot be easily removed by mechanical polishing, then it is preferable to bombard at incident angles greater than θ_p in order to minimise general surface relief.

5. Conclusion

For maximum sputtering efficiency and surface finish, polished silica glass surfaces should be bombarded at an incident angle of about 40°. This conclusion should be applicable to other non-crystalline materials with a sputtering yield curve similar to that of glass.

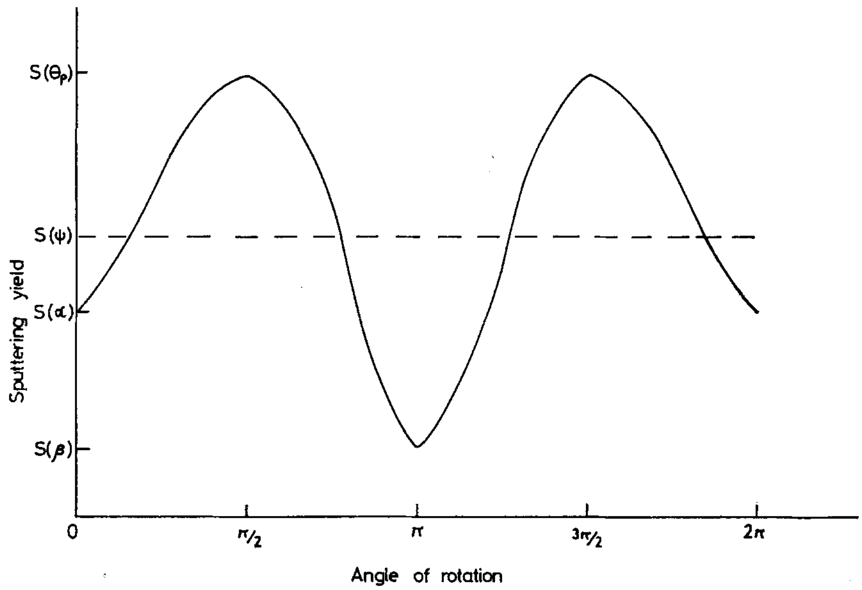

Figure 11 Variation of sputtering yield on the hummock side of intersection A due to rotation of the specimen.

Acknowledgement
This work is financially supported by the National Physical Laboratory. Thanks are due to Mr D. J. Clinton for assisting in the operation of the scanning electron microscope at the National Physical Laboratory.

References
1. D. J. BARBER, *J. Mater. Sci.* **5** (1970) 1.
2. M. PAULUS and F. REVERCHON, "Le Bombardment Ionique" (C.N.R.S., France, 1962) p. 223.
3. I. S. T. TSONG, *Phys. Stat. Sol.* (a) **7** (1971) 451.
4. L. HOLLAND, R. E. HURLEY, and L. LAURENSON, *J. Phys. E.* (*Sci. Instrum.*) **4** (1971) 198.
5. C. G. CROCKETT, to be published.
6. R. K. FITCH, T. MULVEY, W. J. THATCHER, and A. H. MCILRAITH, *J. Phys. D.* (*Appl. Phys.*) **3** (1970) 1399.
7. H. BACH, *J. Non-Crystalline Solids* **3** (1970) 1.
8. A. D. G. STEWART and M. W. THOMPSON, *J. Mater. Sci.* **4** (1969) 56.
9. W. PRIMAK and J. LUTHRA, *J. Appl. Phys.* **37** (1966) 2287.
10. G. K. WEHNER and D. J. HAJICEK, *ibid* **42** (1971) 1145.
11. M. J. NOBES, J. S. COLLIGON, and G. CARTER, *J. Mater. Sci.* **4** (1969) 730.
12. G. CARTER, J. S. COLLIGON, and M. J. NOBES, *ibid* **6** (1971) 115.

Received 23 November and accepted 13 December 1971.