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Synchrotron X-ray topography of bismuth silicon oxide crystals

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Abstract

The generation and distribution of growth defects in Czochralski-grown bismuth silicon oxide ($Bi_{12}SiO_{20}$) crystals have been studied by synchrotron X-ray topography. The relationship of the paths of grown-in dislocations with the shape of the crystal–melt interface is also reported.

1. Introduction

Bismuth silicon oxide ($Bi_{12}SiO_{20}$) possesses good piezoelectric properties and high optical activity. It is also photoconductive and exhibits a linear electro-optical effect. These properties allow these crystals to be used both in optics and in microwave acoustic devices [1,2].

These applications place severe requirements on the quality of the material, currently obtained by the Czochralski method. However, as-grown crystals usually contain strains due to different defects, for instance, facets, growth striations following the shape of the crystal–melt interface, inclusions of foreign material and dislocations.

The information obtained by X-ray topography concerns not only crystal perfection but also growth mechanism. In a preliminary investigation [3], X-ray topography allowed us to study the influence of the growth parameter variations with reference to the quality of the crystals obtained and to correlate the observed defects with the kind of crystal–melt interface.

The aim of this work is to characterize the arrangement of the dislocations in bismuth silicon oxide crystals and their mode of generation and propagation in order to control or eliminate these defects. For this purpose transmission X-ray topographs were made using synchrotron radiation, since conventional sources of X-ray radiation are extremely difficult to use because of the high absorption coefficient of the material.

2. Experimental procedure

The bismuth silicon oxide crystals used in our investigation were grown by the Czochralski method [4] with growth direction [001].

The crystals were sliced with two orientations: perpendicular and longitudinal with respect to the pulling axis. Perpendicular slices were studied in a previous work [3]. In order to study the interface in the earlier stages of growth, the longitudinal slices were cut from the neck and shoulder region, see Fig.

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1. Particular care was taken in the cutting process since this material is extremely brittle, so the thickness of slices obtained was about 500 μm. A conventional polishing process was carried out with SiC powder and alumina micropolish. Due to the high absorption of these crystals, the final thickness of the slices, t, must be reduced to about 150 μm.

X-ray topographs have been obtained in transmission Laue geometry using the white radiation delivered by the DCI synchrotron at Laboratoire pour l'Utilization du Rayonnement Électromagnétique, LURE (Orsay, France). High-resolution Laue pictures can be recorded in a simple way. There is no need to make an accurate setting of the crystal. The high absorption coefficient of this material makes it necessary also to work with the higher energy range of the spectrum; even under these conditions the exposure times are of the order of 60 min.

The film was adjusted parallel to the sample at a distance of about 20–25 cm, chosen in this way to provide in each case a good compromise between resolution and detachment of the diffraction spots.

Owing to the size of our slices, 18 × 20 mm, all these topographs are made up of two overlapping images.

3. Results and discussion

Figs. 2–4 show X-ray topographs taken on the three (100) slices (L1, L2, L3 in Fig. 1) of the same bismuth silicon oxide crystal. They show the characteristic strains generally present in the earlier stages of melt-grown crystals. The contrasts observed correspond mainly to grown-in dislocations (bundles and lines), growth striations and facets.

The distribution of the facet regions is consistent with our previous results [3]. Two types of facet regions can be observed. The first one, namely F_c on the topographs, is localized in the central part of the crystal and traverses longitudinally the crystal from beginning to end of the growth. On the other hand, the second ones, F, are concentrated in the outer regions of the crystal. They externally correspond to habit faces. The development of the growth facets in the context of a slightly convex interface, as in this crystal, is frequent and it occurs when the growth

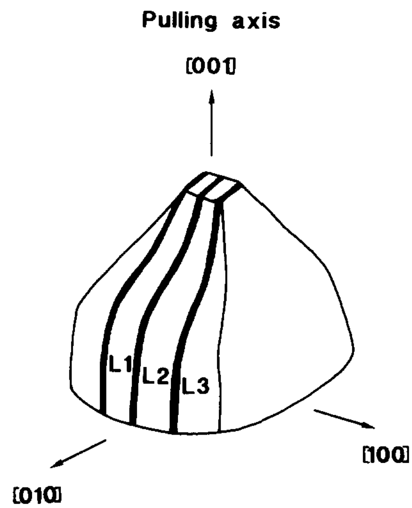

Fig. 1. Sketch of the location of three slices (L1, L2, L3) in the bismuth silicon oxide single crystal.

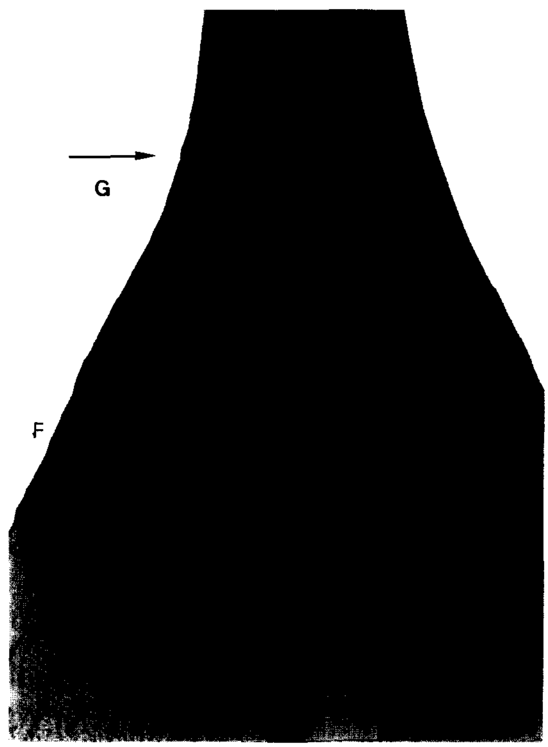

Fig. 2. Topograph of the L1 (100) slice. 060 reflection, $\lambda = 0.4$ Å, $\mu t = 3.53$.

front is parallel to morphologically important crystal faces, as shown by Brice [5].

Growth striations follow the crystal–melt interface. They appear in the outer facet region of this crystal showing generally a strong contrast, see S_1 in Figs. 2–4. The growth striations, namely S_2, are also visible on the topograph of Fig. 3 corresponding to the L2 central slice of the crystal. Their contrast is very slight and narrow corresponding to more stable growth conditions. As can be observed on the topographs of Figs. 2 and 4, the S_2 growth striations are not present in regions surrounding the central zone of the crystal. This fact is in agreement with our previous results showing that other facets in the ⟨100⟩ directions can develop, accompanying the central facet F_c.

Concerning the arrangement and propagation of the dislocations, two different kinds of grown-in dislocations can be observed in these topographs. The first ones, D_1, correspond to dislocations propagated from those already present in the seed. They follow slightly curved paths and appear as individuals and as bundle dislocations. Corresponding to a convex crystal–melt interface, this kind of dislocation spreads to the crystal edges and can be reduced or eliminated as we have reported in a previous work [3].

The second type of dislocations, D_2, is character-

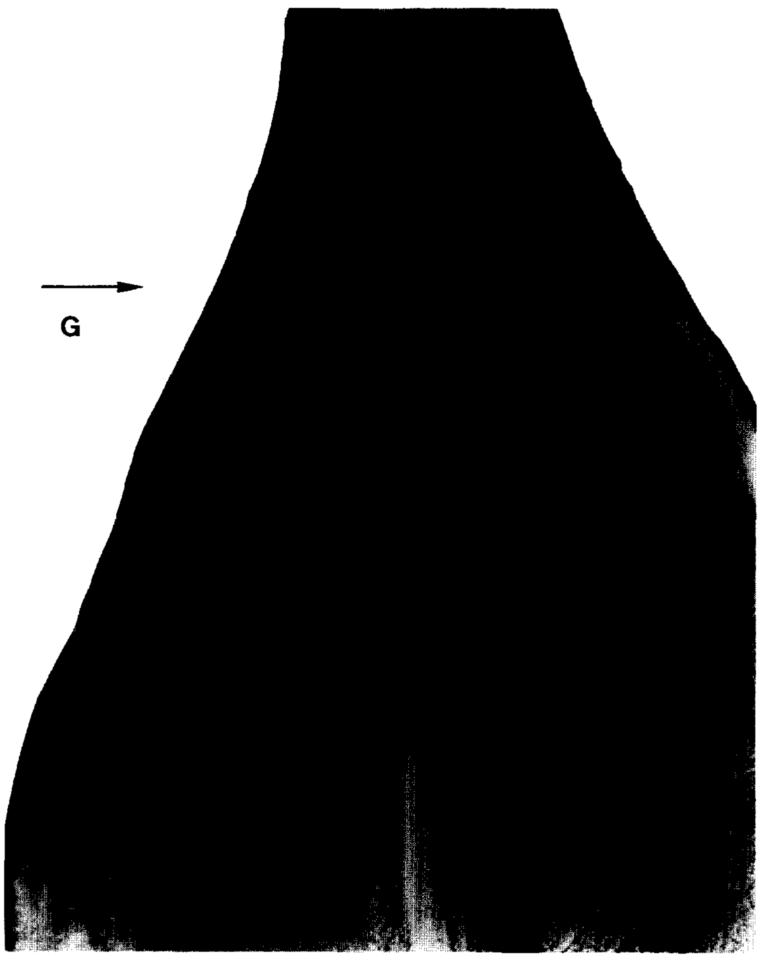

Fig. 3. Topograph of the L2 (100) slice. 060 reflection, $\lambda = 0.4$ Å, $\mu t = 3.53$.

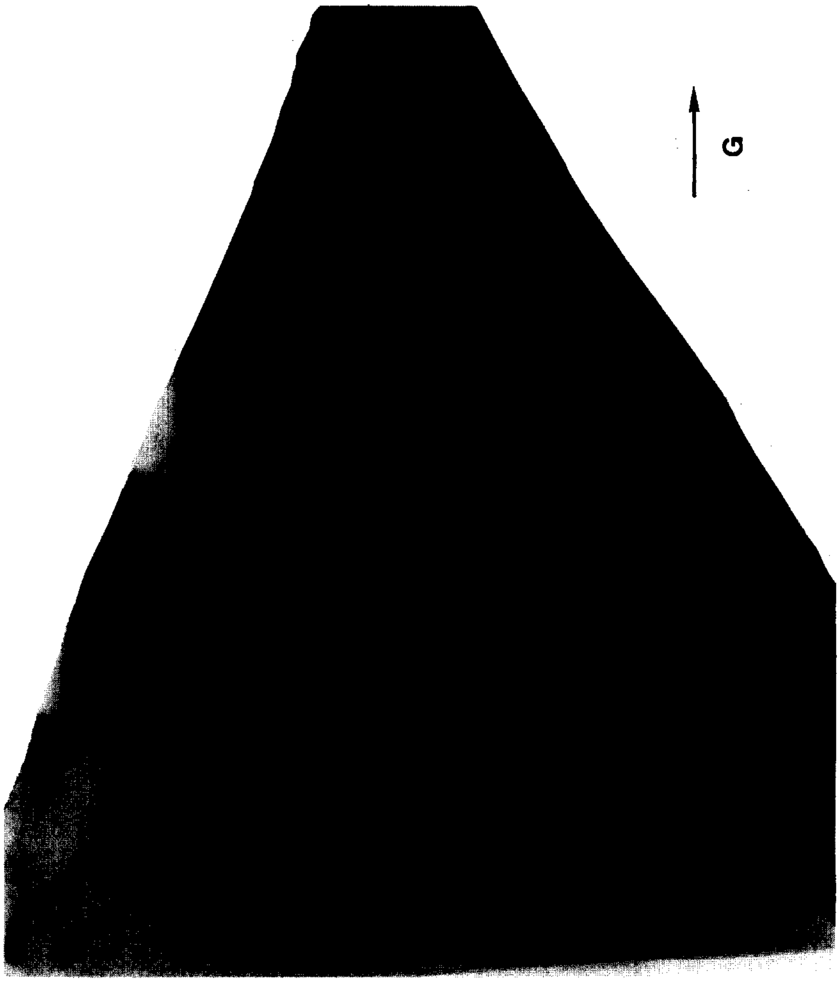

Fig. 4. Topograph of the L3 (100) slice. 006 reflection, $\lambda = 0.4$ Å, $\mu t = 3.53$.

ized by its appearance and localization. These dislocations are always related to the outer facet regions and developed as dislocation bundles showing a strong contrast. A sudden change in the crystal diameter can be observed in relation to the outer facet F, and D_2 dislocation development. These defects originate from local thermal fluctuations leading to a high supercooling and consequent rapid growth.

The propagation of the grown-in dislocations in this crystal is in agreement with that theoretically predicted [6]. In this way, their paths are nearly perpendicular to the crystal growth front and can be refracted, changing from one growth facet to another, see Fig. 3.

It can be concluded that the growth of bismuth silicon oxide crystals with a convex and slightly faceted interface promotes the divergence of dislocations and their spread to the crystal edges, so leading to crystals with a high-quality central zone.

References

[1] A.R. Tanguay, Jr., Doctoral Dissertation, Department of Physics, Yale University, 1977.
[2] B.C. Grabmaier and R. Oberschmid, Phys. Status Solidi (a) 96 (1986) 199.
[3] J. Martínez-López, M. González-Mañas, M.A. Caballero, E. Diéguez and B. Capelle, J. Physique IV, 4 (1994) 169.
[4] J. Martínez-López, M.A. Caballero, M.T. Santos, L. Arizmendi and E. Diéguez, J. Crystal Growth 128 (1993) 852.
[5] J.C. Brice, J. Crystal Growth 6 (1970) 205.
[6] H. Klapper and H. Küppers, Acta Cryst. A 29 (1973) 495.