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DEFECTS IN DETWINNED $LaGaO_3$ SUBSTRATES

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Single crystals of lanthanum gallate would be the suitable substrate for YBaCuO films except for the phase transition and the tendency to twinning existing in this material. However, by appropriate choice of growth conditions in the Czochralski method, it is possible to grow single crystal of $LaGaO_3$ with low density of twin boundaries. Special stress and temperature treatment can then be applied to such materials to remove majority of existing twins. The substrates were examined by X-ray topography before and after detwinning and the surface was scanned with a profilometer.

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1. Introduction

The general interest in obtaining good quality single crystals of various perovskite materials by Czochralski methods is directly related to the discovery of high-T_c superconductors (HTSC). Lanthanum, neodymium and praseodymium gallates [1, 2] have often been expected as substrate materials for HTSC epitaxy mainly because of their attractive dielectric properties (low ε and $\tan\delta$) and low lattice mismatches, both at deposition and application temperatures. The most crucial problem in application of these materials as substrates for the epitaxy is the existence of the structural phase transitions [3] leading to the twinning and surface roughening [4]. First-order phase transition, from orthorhombic to rhombohedral structure, was observed in $LaGaO_3$ and $SmAlO_3$ at 150°C and 785°C, respectively. In turn, second-order phase transition occurs at 1180°C for $LaGaO_3$, 950°C for $NdGaO_3$, 870°C for $PrGaO_3$ and around 500°C for $LaAlO_3$ [3].

2. Experiment and results

$LaGaO_3$ — one of materials, which is relatively easy to obtain by the Czochralski method, seems to be very useful for the epitaxy of HTSC, however, it shows a tendency to twinning, which is connected with the phase transition. For this material the lowest density of twins is observed in single crystals grown on ⟨101⟩ and ⟨100⟩ oriented seeds [5, 6]. In each of the crystal growth processes, regardless of the crystal orientation, the very important factors deciding on the low twins density are: (i) good quality of the seed, (ii) low temperature gradients above the melt surface and (iii) the good beginning of crystal growth [5], in which the growth process starts only at the original seed material (after melting of the outer parts of the seed, occurring just below the melt surface, and without crystallization on rapidly growing region at the seed center).

There are two main types of twins in this material. One of them is connected with variety of orientations of the orthorhombic *c*-axis. There are three possible spatial orientations of this axis, each of them can be transformed into another by mirror reflection in the appropriate (112) boundary plane.

The second type of twins is connected with spatial orientation of *a* and *b* orthorhombic axes. In this case the twin boundary is (110)-type plane. This second type of twins should vanish at temperature above 145°C associated with the first-order transition from orthorhombic to rhombohedral structure. The volume twins (large domains of different orientation) and the thin twin lamellae are very often observed in this material.

It is well known that, under a stress applied along specific crystallographic directions, at temperatures near the phase transition, the twin boundaries can be moved leading to a single-domain crystal. We tested this procedure for $LaGaO_3$ [5].

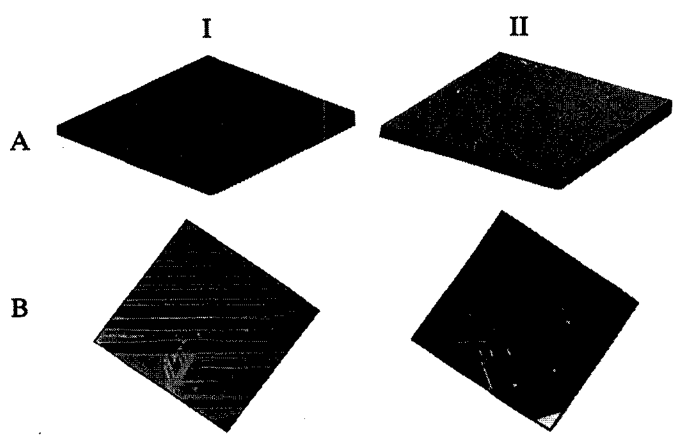

Fig. 1. The X-ray topography (A) and optical images (B) of $LaGaO_3$ (100) oriented substrate before (I) and after detwinning (II).

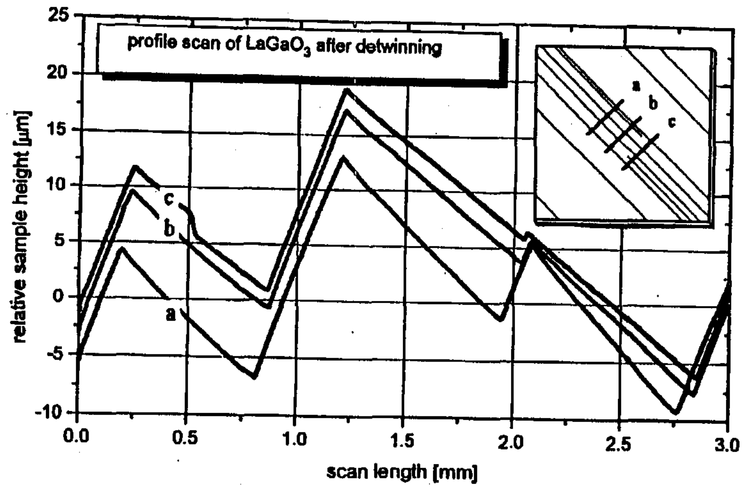

Fig. 2. The profile scan image of the $LaGaO_3$ substrate surface measured after detwinning. Measurements were performed along various positions (*a*), (*b*) and (*c*) as schematicaly marked in the inset.

An uniaxial stress, typically 6 to 12 MPa was applied in the ⟨001⟩ direction on the substrate cut from the crystal grown along the ⟨100⟩ orientation. Heating of thus compressed substrate above the phase transition temperature followed by slow cooling to room temperature decreased considerably the number of twins (both lamellae and volume domains). Detwinned samples were examined by X-ray topography and observed with polarizing microscope and no residual twins were found. The X-ray topography and optical images of $LaGaO_3$ (100) oriented substrate before and after detwinning are shown in Fig. 1. Unfortunately, after detwinning vanishing of twin lamellae involves forming of ramps on polished sample's surface. These ramps were scanned with a profilometer stylus (DEKTAR 8000, version 4.04). Typical results obtained along the direction perpendicular to removed twin boundaries are shown in Fig. 2. The height and slope of these ramps correspond to the dimensions of the previously existing twins.

Another method, which enables one to introduce controlled stresses during the crystal growth, is based on replacement of some ions with the other having ionic radius smaller than these in the original material. It has already been shown by Koren et al. [7] that in the case of $LaGaO_3$ the substitution of 5 at.% of lanthanum by gadolinium atoms led to the twin-free crystalline structure. Lanthanum doped $NdGaO_3$ [8] is equivalent to the formation of solid solutions between $NdGaO_3$ and $LaGaO_3$. In order to obtain the perfect twin-free substrate for YBaCuO epitaxy, material should contain of about 20 at.% of lanthanum.

3. Summary

Single crystals of lanthanum gallate would be the suitable substrate for YBaCuO films except for the tendency to twinning. However, by appropriate choice of growth condition in the Czochralski method it is possible to grow single crystals of $LaGaO_3$ with low density of twins. Next, we can obtain twin-free samples after detwinning by a special stress treatment. But unfortunately, such performed substrates are very sensitive to external stresses, and consequently both during polishing and film deposition new twins can be formed. Vanishing of twin lamellae or arising new ones involves forming defects on the surface in form of ramps on polished sample.

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