Vol. A53 (1978) *ACTA PHYSICA POLONICA* No 6

CALCULATION OF THE DISTRIBUTION OF STRESSES IN SEMI-TRANSPARENT SINGLE CRYSTAL PULLING BY THE CZOCHRALSKI TECHNIQUE

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(Received March 15, 1977, final version received December 1, 1977)

Numerical computations of the distribution of components of thermoelastic stresses in single crystals of lithium niobate, $LiNbO_3$ and yttrium-aluminum garnet, $Y_3Al_5O_{12}$, grown in appropriate technological regime by the Czochralski method were performed. The greatest stresses appeared close to the front of crystallization, i. e. in the area, where thermal gradients are relatively high. In the plane perpendicular to the axis of crystal growth the stresses focus close to the side surface of the solid state. The time relations for stresses were discussed. In the case of crystals grown under severe technological conditions, the so-called neutral zone appears; formation of this zone is explained by the example of YAG.

1. *Numerical computations*

In the former paper the analytical formulae for components of thermoelastic stresses in single crystals grown by the Czochralski method were developed [1]. Now, using these formulae, the values of stress components are evaluated numerically. This has been performed by calculating the case of the crystals of lithium niobate, $LiNbO_3$ and yttrium-aluminum garnet, $Y_3Al_5O_{12}$.

The machine used for numerical calculation was an electronic digital computer JEC-6 produced by the Japanese Company JEOL. The following crystal dimensions were assumed: length — 5 cm, diameter — 1 cm. The ambient temperature was 1070 K in case

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TABLE I

Values of stiffness coefficients of single crystals of lithium niobate and yttrium-aluminum garnet

c_{ij} [10^{11} dyn/cm^2]	$LiNbO_3$ [2]	$Y_3Al_5O_{12}$ [3]
c_{11}	20.30	33.40
c_{33}	24.24	
c_{44}	5.95	11.51
c_{12}	5.73	11.12
c_{13}	7.52	
c_{14}	0.85	
c_{66}	7.28	

of $LiNbO_3$ and 1270 K in case of YAG. The value of stiffness coefficients were assumed in accordance with Table I.

For each stress component a map of values has been made for r varying in steps of 0.1 cm, and z — in steps of 0.5 cm. The results are shown on Figures 1—4.

2. *Discussion of the form of stress components in a crystal*

From the diagrams of stress components one can draw the following conclusions.

a) Generally, under the specified technological conditions, the stresses in single crystals of yttrium-aluminum garnet are several times greater than those in single crystals of lithium niobate. The general features of diagrams for the two materials are however similar.

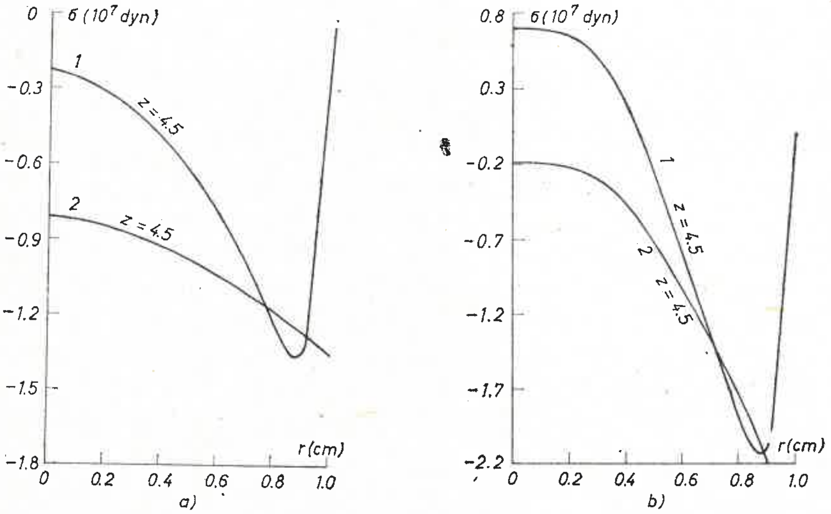

Fig. 1. Radial distribution of the stress components σ_{rr} (curve *1*) and $\sigma_{\varphi\varphi}$ (curve *2*) in single crystals of: a. lithium niobate and b. yttrium-aluminum garnet

b) The component σ_{rr} of stress decreases with the increase of distance from the front of crystallization (Figs. 1, 2). In the case of $LiNbO_3$ the dominating mechanism is compression; however in the case of YAG lengthening of the internal volume close to the front of crystallization appeares as well. This situation resembles the distribution of stresses in leucosapphire [4–6] and ferrites [7] single crystals grown by the flame fusion method. In this technique crystals are subjected especially strong thermal shocks in the moment when the hydrogen-oxygen burner is switched off. The external layers, more coded, execute the pressure against the internal volume subjected to a much higher temperature and featuring, correspondingly, smaller contraction ability.

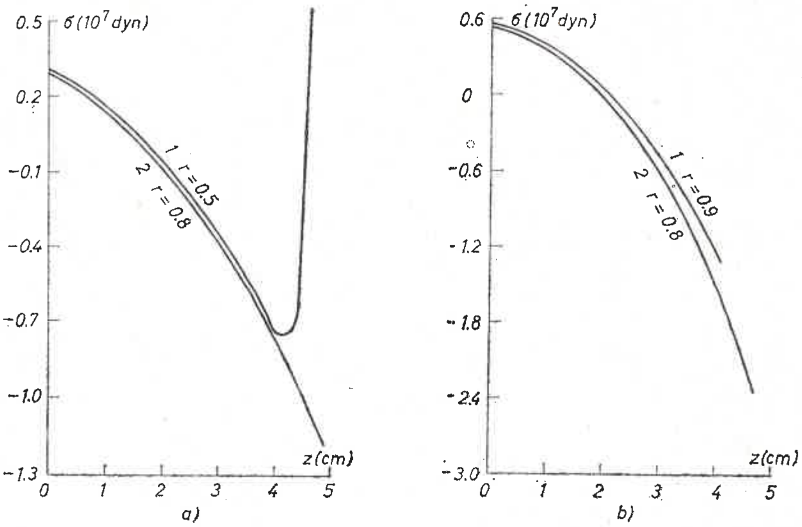

Fig. 2. Axial distribution of the stress components σ_{rr} (curve 1) and $\sigma_{\varphi\varphi}$ (curve 2) in single crystals of: a. lithium niobate and b. yttrium-aluminum garnet

After some time, at the stage of equalizing of the radial temperature distribution, the internal layers start to contract too. They tend to break off from the external layers, which is opposed by the forces of intermolecular cohesion. The compressing stresses are thus replaced by lengthening ones. Cooling the crystals grown by the flame fusion technique is performed very intensively and rapidly.

Due to this fact, the distribution of stresses as described herein becomes "frozen", which can be proved by the so-called neutral lines in the Verneuil crystals of leucosapphire [5, 6]. In the case of crystals pulling by Czochralski technique the above phenomenonl as a rule, does not occur. It results, first of all, from much more advantageous therma, conditions acting on solid phase. An effect similar to the mentioned one takes place in the case of YAG.

It does not occur in the case of lithium niobate where temperature difference at both ends of a crystal equals 450°C whereas in the case of yttrium-aluminum garnet it equals approximately 1000°C.

c) From the diagram of σ_{rr} as a function of r for two different values of z one can see that the gradient of temperature in the axial direction is the main factor determining the distribution $\sigma_{rr}(r)$. This effect of axial differences of temperature is especially important in the crystal regions near the interface. This observation is confirmed by the diagram of σ_{rr} as a function of z. With increasing distance from the interface, the lengthening

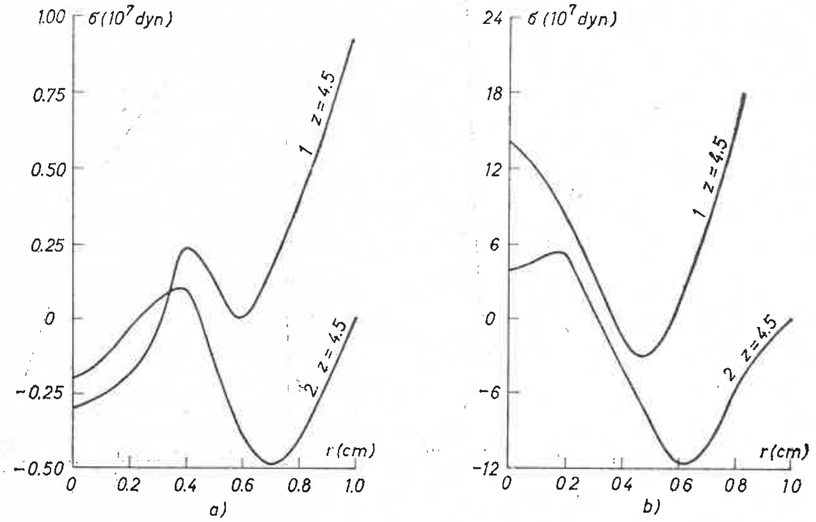

Fig. 3. Radial distribution of the stress components σ_{zz} (curve *1*) and σ_{rz} (curve *2*) in single crystals of: a. lithium niobate and b. yttrium-aluminum garnet

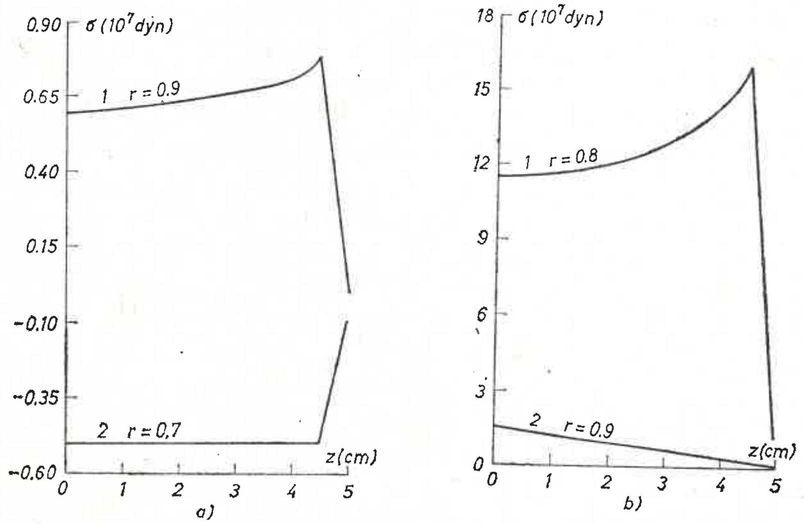

Fig. 4. Axial distribution of the stress components σ_{zz} (curve *1*) and σ_{rz} (curve *2*) in single crystals of: a. lithium niobate and b. yttrium-aluminum garnet

(tensile) stresses are replaced by the contracting ones and then, at the other end of crystal, they became again lengthening ones.

d) $\sigma_{\varphi\varphi}$ stresses form a part of the general image of phenomena involved in the cooling of crystals. Namely, sufficiently cooled internal layers counteract contraction of colder external layers of the crystal.

Due to this fact the external layers are lengthened in the direction perpendicular to crystal radius. The greatest, with respect to absolute values, stresses $\sigma_{\varphi\varphi}$ occur at the side surface of the solid phase, they decrease when approaching the geometric axis of a boule.

e) The axial course (form) of $\sigma_{\varphi\varphi}$ creates some interpretation confusion. Namely, it is difficult to explain the change of sign of this component in the part of crystal apart from the front of crystallization. The logical explanation of this relationship seems to be analogous to the one in the case of σ_{rr} stress diagram. Since in the vicinity of the geometric axis of the boule, the material is compressed in the radial direction and this must influence the sign of $\sigma_{\varphi\varphi}$ stress: extension in the direction perpendicular to the radius is replaced by contraction in the same direction.

f) The component σ_{zz} has greater values than other components of stress. Approximately in the middle of the radius, i.e. in the region where σ_{rr} changes its sign, the σ_{zz} component has its minimum value. (In soviet literature of this problem this region is named a "neutral zone"). Toward the surface of a crystal σ_{zz} increases.

Surprisingly there are some small changes in σ_{zz} occurring in the axial direction. Due to high thermal gradients along the z axis, the status of σ_{zz} stress becomes "frozen" here, similarly as in the case of σ_{rr}.

g) The component σ_{rz} also shows the existence of a neutral zone. With respect to absolute value, it increases when approaching the crystal surface.

Similar to the case of σ_{zz} the axial dependence of σ_{rz} does not show any greater changes.

The components σ_{rr} and $\sigma_{\varphi\varphi}$ determined here generally have diagrams similar to the ones in the case of isotropic materials [8]. The latter is understandable since the materials studied in these investigations are also isotropic in the directions in which these components act.

In the work by Tchebanova [9] a case of stress distribution in single crystal grown by the Kyropoulos method is discussed. The author assumed an increase of mass at the side surface of the solid phase. Contrary to the present paper, the author has not performed the computations in the quasistationary case.

The general image of stresses in a single crystal pulling by the Czochralski technique reads thus: in the crystal volume close to the interface (solid-liquid) there are very strong stresses with domination of compression ones; they cover e.g. in the case of σ_{rr} and $\sigma_{\varphi\varphi}$ almost the whole crystal. When the crystal grows and the distance of the given volume element from the interface increases, the stresses decrease and change sign due to the above described processes.

Evidently, the slower course of this phenomenon, the smaller is the rate of pulling. Together with a decrease in the absolute value of the stress components, their gradients decrease: stresses become uniform in the material. They are the so-called residual stresses, that can be partly removed in the process of thermal treatment of single crystals, i.e. annealing.

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