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Effect of anisotropy on thermoelastic stresses in cylindrical gallium oxide crystals grown from a melt

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The study of thermal stresses in crystals grown from a melt is of great importance for optimizing growth regimes. The emergence of new promising materials, such as gallium oxide, requires stress calculations taking into account the anisotropy of the thermal and elastic properties of the material. A study was carried out of the influence of anisotropy on the distribution of thermoelastic stresses in thin crystalline rods of gallium oxide. Approximate formulas for the components of the stress tensor are given, obtained using the asymptotic integration of the thermoelasticity equations taking into account rectilinear anisotropy of a general form. A comparison of stress values for two growth directions was carried out. It is shown that choosing the orientation of the growth direction makes it possible to control the magnitude and distribution of thermoelastic stresses that arise in gallium oxide crystals when they are grown from a melt.

Keywords: thermoelastic stresses, asymptotic method, anisotropy of thermal and elastic properties.

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Introduction

Recently, transparent semiconductors based on gallium oxide crystals ($\beta - Ga_2O_3$) have been of great interest in the creation of new types of electronic devices (photodiodes, transparent thin-film transistors, energy-saving windows, etc.) [1,2]. One of the ways to grow bulk gallium oxide crystals is the Czochralski method, in which the crystal is drawn from the melt in an iridium crucible. Despite a number of technological difficulties, crystals in the form of cylinders with a diameter of up to 50 mm [3–6]. However, for the further development of the technology for obtaining perfect crystals, it is necessary to study thermal stresses and structural defects that arise during the growth process. For β gallium oxide phase crystals with a low-symmetrical monoclinic structure, a strong anisotropy of thermal and elastic properties must also be considered. In a number of papers, numerical simulation of heat transfer processes in the crystal and growth zone was carried out, and calculations of thermal stresses were carried out [7–9]. It is shown that the numerically calculated maximum stresses according to the Mises criterion strongly depend both on the crystallographic direction of growth [7] and on the configuration of the elements of the thermal zone [9]. Definitely, these calculations are a good tool for studying the processes of heat transfer in the growth zone and determining the fields of temperature and thermal stresses in the grown crystals, but they require complex packages of special programs and powerful computing resources.

In this paper, on the basis of analytical calculations, the effect of anisotropy of thermal and elastic properties on the distribution of thermoelastic stresses in thin cylindrical gallium oxide crystals grown from a melt by the Czochralski method is studied. The calculations were performed according to approximate formulas obtained by solving the stationary thermoelasticity problem by the method of singular perturbations, considering the rectilinear anisotropy of the general form [10,11].

1. Approximate Formulas for Thermoelastic Stresses

Here are the coefficients of thermal expansion and elastic stiffness for the standard coordinate system (axis z coincides with crystallographic direction [001] [7]. Coefficients of thermal expansion: $\overline{\alpha}_{11} = 4.7 \cdot 10^{-6}$, $\overline{\alpha}_{12} = \overline{\alpha}_{21} = 0$, $\overline{\alpha}_{13}=\overline{\alpha}_{31}= -0.17 \cdot 10^{-6}$, $\overline{\alpha}_{22}=8.3 \cdot 10^{-6}$, $\overline{\alpha}_{23}=\overline{\alpha}_{32} = 0$, $\overline{\alpha}_{33} = 8.5 \cdot 10^{-6}\,(\mathrm{K}^{-1})$. The elastic stiffness coefficient tensor (dimension $(10^{10}\,\mathrm{N/m^2})$) is written as

$$(\overline{c}_{ij}) = \begin{pmatrix} 23.8 & 13.0 & 15.2 & 0 & -0.4 & 0 \\ 13.0 & 35.9 & 7.8 & 0 & 0.2 & 0 \\ 15.2 & 7.8 & 34.6 & 0 & 1.9 & 0 \\ 0 & 0 & 0 & 4.9 & 0 & 0.6 \\ -0.4 & 0.2 & 1.9 & 0 & 9.1 & 0 \\ 0 & 0 & 0 & 0.6 & 0 & 10.7 \end{pmatrix}.$$

The dimensional values of the coefficients of thermal expansion and elastic stiffness are indicated by a line at the top. We invert this matrix and move on to the elastic compliance coefficients. Let us consider a crystal rod of length $L = 0.1\,\mathrm{m}$ and radius $R = 0.01\,\mathrm{m}$. Let us move on to the dimensionless coordinates as follows: $\overline{r} = R\,r$, $\overline{z} = L\,z$. Small parameter of the problem $\varepsilon = R/L$. Let us proceed to the dimensionless coefficients of thermal expansion and elastic compliance by normalizing to the corresponding invariants $\overline{\alpha}_{00} = 7.17 \cdot 10^{-6}\,(\mathrm{K}^{-1})$, $\overline{s}_{00} = 0.051 \cdot 10^{-10}\,(\mathrm{m}^2/\mathrm{N})$.

We work in the cylindrical coordinate system (r, θ, z). Using the method of asymptotic integration of the thermoelastic equations [11] we obtain with an accuracy of terms of order ε^2 the formulas for the components of the thermoelastic stress tensor

$$\overline{\sigma}_r(r, z) = A_1\,(1 - r^2)\,\overline{\Delta}\,\frac{d^2T_0}{dz^2}\,\varepsilon^2,$$

$$\overline{\sigma}_\theta(r, z) = A_1\,(1 - 3r^2)\,\overline{\Delta}\,\frac{d^2T_0}{dz^2}\,\varepsilon^2, \quad \overline{\sigma}_{r\theta} = 0,$$

$$\overline{\sigma}_{rz}(r, \theta, z) = \big(A_2\sin(\theta) + A_3\cos(\theta)\big)\,(1 - r^2)\,\overline{\Delta}\,\frac{d^2T_0}{dz^2}\,\varepsilon^2,$$

$$\overline{\sigma}_{\theta z}(r, \theta, z) = \big(A_2\cos(\theta) - A_3\sin(\theta)\big)\,(1 - 3r^2)\,\overline{\Delta}\,\frac{d^2T_0}{dz^2}\,\varepsilon^2,$$

$$\overline{\sigma}_z(r, \theta, z) = B_0[1 + r^2[B_1\cos^2(\theta) + B_2\sin(\theta)\cos(\theta) - (4 + B_1)\sin^2(\theta)]\,\overline{\Delta}\,\frac{d^2T}{dz^2}\,\varepsilon^2,$$

where

$$B_0 = -\frac{1}{s_{33}}\left[(s_{13} + s_{23})A_1 + s_{34}A_2 + s_{35}A_3 - \frac{1}{8}\,(\alpha_1 + \alpha_2 - \alpha_3)\right],$$

$$B_1 = \frac{1}{B_0}\left[(s_{13} + 3s_{23})A_1 + 3s_{34}A_2 + s_{35}A_3 - \frac{1}{2}\left(\alpha_1 - \frac{1}{2}\,\alpha_3\right)\right],$$

$$B_2 = -\frac{1}{B_0}\left[2(s_{36}A_1 + s_{35}A_2 + s_{34}A_3) + \frac{1}{2}\,\alpha_6\right],$$

$$\alpha_i = \alpha_{ii},\ i = 1, 2, 3,\ \alpha_4 = \alpha_{23},\ \alpha_5 = \alpha_{13},\ \alpha_6 = \alpha_{12}.$$

In these formulas, $\overline{\Delta} = \overline{\alpha}_{00}\,\overline{T}_{00}/\overline{s}_{00}$ — is the normalizing factor that provides the transition from the dimensionless to the dimensional components of the thermoelastic stress tensor, and d^2T_0/dz^2 — the dimensionless second derivative of the temperature along the rod axis, $\overline{T}_{00}$ — melting point of gallium oxide (2080° K). The temperature is normalized to the melting point: $\overline{T}_0 = T_0\,\overline{T}_{00}$. We consider the second derivative of the temperature constant and choose the value of the second derivative of the temperature at distance 0.02 m from the crystallization front: $d^2\overline{T_0}/d\overline{z}^2 = -5 \cdot 10^4\,\mathrm{K/m}^2$, $d^2T_0/dz^2 = -5\cdot10^4 L^2/\overline{T}_{00} = -0.24$. The constants A_1, A_2, A_3 are defined from a system of three linear algebraic equations

$$a_{11}A_1 + a_{12}A_2 + a_{13}A_3 = b_1,$$

$$a_{12}A_1 + a_{22}A_2 + a_{23}A_3 = b_2,$$

$$a_{13}A_1 + a_{23}A_2 + a_{33}A_3 = b_3,$$

$$a_{11} = 3s'_{22} + 2s'_{12} + s'_{66} + 3s'_{11},$$

$$a_{12} = 3s'_{24} + s'_{14} + s'_{56},\ a_{13} = 3s'_{15} + s'_{25} + s'_{46},$$

$$a_{22} = 3s'_{44} + s'_{55}, \quad a_{23} = 2s'_{45},$$

$$a_{33} = 3s'_{55} + s'_{44},\ s'_{ij} = s_{ij} - \frac{s_{i3}s_{3j}}{s_{33}},\ i, j = 1, 2, 4, 5, 6,$$

$$b_1 = -\frac{1}{4s_{33}}\,[(2\alpha_2 - \alpha_3)s_{13} + (2\alpha_1 - \alpha_3)s_{23} - \alpha_6 s_{36} + (\alpha_1 + \alpha_2)s_{33}],$$

$$b_2 = -\frac{1}{4s_{33}}\,[(2\alpha_1 - \alpha_3)s_{34} - \alpha_6 s_{35} + \alpha_4 s_{33}],$$

$$b_3 = -\frac{1}{4s_{33}}\,[(2\alpha_2 - \alpha_3)s_{35} - \alpha_6 s_{34} + \alpha_5 s_{33}].$$

2. Stress Calculation Results

Fig. 1 shows the results of calculations of the components of the thermoelastic stress tensor for the standard coordinate system (axis z coincides with the crystallographic direction [001]). Analysis of the thermoelastic stress tensor components for the orientation of the crystal growth direction [001] shows that the shear stress $\sigma_{r\theta}$ is zero, the normal components σ_r and σ_θ (Fig. 1, a) are independent of the θ coordinate and are distributed in the direction along the radius by parabola. We note that this is the case for all orientations of the crystal rod, as follows from the approximate formulas for these tensor components given above. This is consistent with the data on stress calculations in the isotropic approximation. Strong anisotropy along the cross-section of the crystal is demonstrated by three components σ_{rz} (Fig. 1, b), $\sigma_{\theta z}$ (Fig. 1, c) and σ_z (Fig. 1, d). The σ_{rz} component is zero on the outer surface, grows toward the center, and has different signs in the $\theta = 0$ and π directions. The component $\sigma_{\theta z}$ has a more complex distribution over the cross section: the zero region is located around the circumference at $r = 0.5$ cm, the stress increases towards the center to 0.04 and to 0.08MPa towards the surface. Accordingly, the stress sign in the directions $\theta = \pi/2$ and $3\pi/2$ is different. It should also be noted that these components make a small contribution to the overall picture of the stress distribution. The most significant in magnitude is the normal component σ_z, the maximum values of 1.3 MPa on the surface at $\theta = \pi/2$ and $3\pi/2$. In the middle of the crystal there is a region of negative values with a maximum of -0.5 MPa, with all lines of equal stresses located practically along the direction $\theta = 0$.

Fig. 2 shows the results of calculations for the second orientation of the crystal: the axis z coincides with the crystallographic direction [010]. This orientation is most commonly used in the growth of gallium oxide crystals by the Czochralski method and is also discussed in [7]. To calculate the stresses in this orientation, the elastic

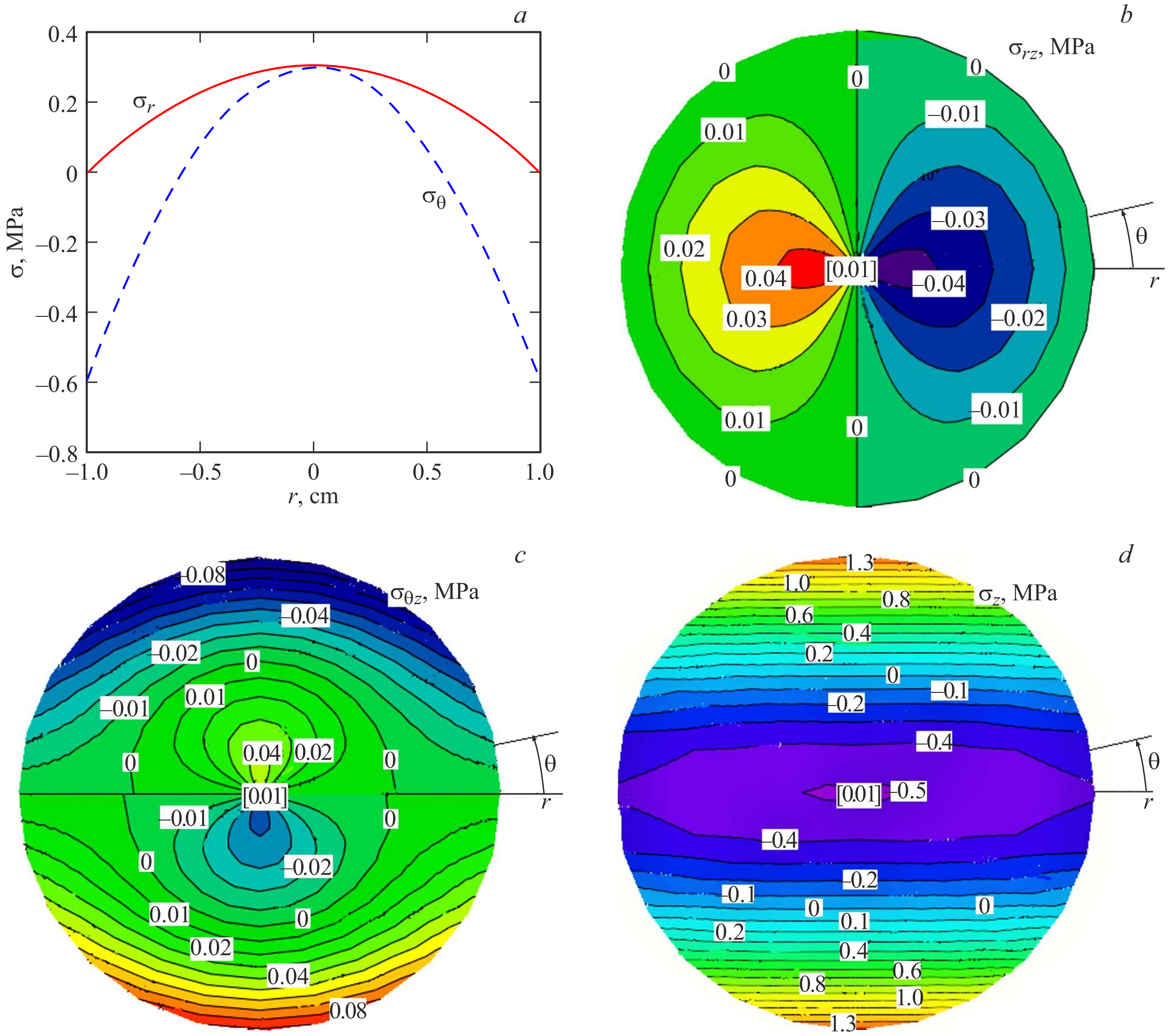

Figure 1. Stress distribution σ_r and σ_θ along the radius of the crystal cross-section (a); stress plots σ_{rz} (b), $\sigma_{\theta z}$ (c) and σ_z (d) in the cross-section of the crystal for the case of growth on axis z with direction [001].

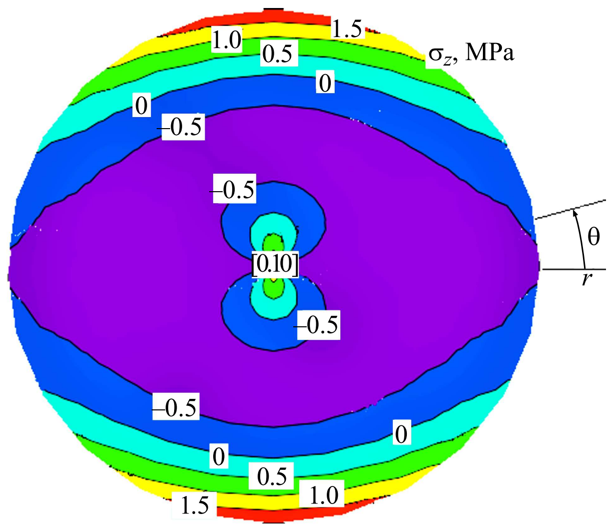

Figure 2. Normal stress plot σ_z for the case of crystal orientation in the direction [010].

constant tensors and thermal expansion coefficients were recalculated. Stress analysis for this case showed that the components σ_r and σ_θ did not change much, the tangent components of the stress tensor $\sigma_{rz}, \sigma_{\theta z}$ were zero. The most significant, as in the first case, was the component σ_z, and the cross-section distribution picture is close to the first case, but the value of the stresses at the surface is slightly higher (up to 1.5 MPa). In terms of magnitude of stresses, our calculations according to the formulas are close to the results of numerical calculations. The maximum average stresses calculated in [7] using the Mises criterion were approximately 6 MPa for a crystal twice as large in diameter. In our opinion, the calculations show a strong influence of the anisotropy of the properties of the gallium oxide crystal and the orientation of the growth direction on thermoelastic stresses. However, in order to select the optimal orientation of the growth direction and minimize the formation of structural defects (dislocations, block boundaries and twins), it is still necessary to solve the problem of recalculating the stress tensor components along the slip planes.

Conclusion

A comparison of stress values for two growing directions was carried out. It is shown that accounting the anisotropy of elastic properties and thermal expansion, as well as the choice of the orientation of the direction of growth of a gallium oxide crystal relative to the crystallographic axes, made it possible to control the magnitude and distribution of thermoelastic stresses arising in the process of growth, and therefore the degree of its structural perfection.

Conflict of interest

The authors declare that they have no conflict of interest.

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