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THERMOELASTIC STRESSES IN OXIDE SINGLE CRYSTALS PULLING BY THE CZOCHRALSKI TECHNIQUE

BY E. ZALEWSKI

Centre of Research and Production of Semiconductor Materials, Warsaw*

AND J. ŻMIJA

Military Technical Academy, Warsaw**

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Thermoelastic stresses belong to the most important factors determining the quality of a single crystal grown by the Czochralski method. In the paper, the distribution of the stress components σ_{rr}, $\sigma_{\varphi\varphi}$, σ_{zz} and σ_{rz} is determined from the potential of the thermoelastic strains for semi-transparent single crystals. The orthotropic case is considered. It has been assumed that the mechanical properties of single crystals do not depend on temperature. The form of the potential of thermoelastic stresses does not take into account convection of the protective atmosphere in the course of crystal growth by Czochralski's method.

1. Generation of stresses in the pulling processes and their contribution to the quality of a monocrystal

In the process of crystallization one has to create those thermodynamic conditions which prefer the formation of the solid phase (crystal) over the liquid one. In Czochralski's method this is reduced to determining a fixed temperature gradient at the front of crystallization and in the crystal. This means that the crystal cannot be in a uniform thermal field. So the different parts of the solid phase may be subjected to different states of stresses, the so-called thermoelastic stresses, caused by the nonuniform extension of the material due to the influence of temperature. Because of great differences in temperature this is the most serious factor deteriorating the structural quality of a crystal.

Thermoelastic stresses are the factors generating other defects in crystals, such as dislocations grain boundaries etc. Let us consider, for example, the formation of dislocations due to radial and axial temperature gradients (Fig. 1) [1]. Due to these differences

* Address: Ośrodek Naukowo-Produkcyjny Materiałów Półprzewodnikowych, Konstruktorska 6, 02-673 Warszawa, Poland.

** Address: Wojskowa Akademia Techniczna, Lazurowa 225, 01-980 Warszawa, Poland.

the distances between planes are distorted: "stretching" occurs in the crystal's cooler parts; thus there appears an cumulation of energy in these thermoelastic stresses which is diminished by the formation of dislocations.

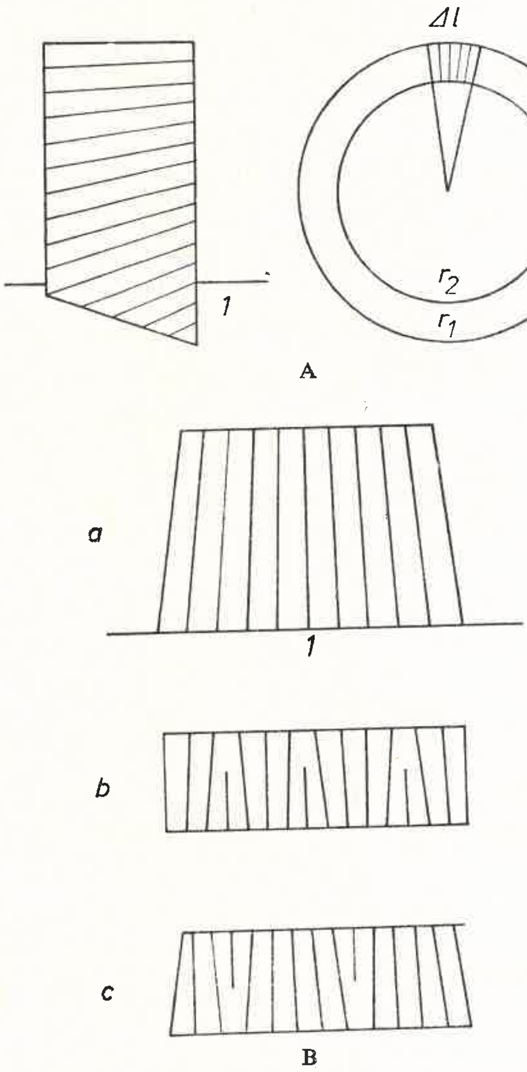

Fig. 1. Formation of dislocations in a crystal due to radial A and axial B temperature gradients, a — deformation of crystallographic planes due to a temperature gradient, b – unannealed crystal, c — partly annealed crystal, 1 – solid phase

In this way the configurative energy of the solid phase increases but the total value of the free energy is reduced by the elastic energy released in this process. Billing [2] has calculated approximately the density of dislocations created in this way from considerations of a purely geometric character. Let e.g. the temperature in the external zone of a crystal be equal T_1 and in the zone directly under it T_2, with $T_2 > T_1$. Zone 2 forces zone 1 to have a perimeter of $2\pi r_2(r_2 = r_2(T_2))$ in length instead of the already existing $2\pi r_1(r_1 = r_1(T_1))$. This is equivalent to applying a force causing a fixed deformation. In order to diminish the elastic energy we have to introduce dislocations by:

$$n = 2\pi r \Delta T(\alpha/b). \tag{1}$$

This gives e.g. for some oxide crystals the following dislocation densities per unit of perimeter length:

$\alpha-Al_2O_3$	$0.5\times 10^2\Delta T$
$Y_3Al_5O_{12}$	$0.4\times 10^2\Delta T$
$CaWO_4$	$1.6\times 10^2\Delta T$
$LiNbO_3$	$1.9\times 10^2\Delta T$

(in the calculations it has been assumed that $r = 1$ cm).

The problem of distribution of stresses in a limited cylinder (this shape will be used to approximate a growing crystal) has not yet been solved exactly. The earlier works are limited to isotropic elastic media and to boundary cases, i.e. to the disc for which the length is assumed far smaller than the radius ($h \ll R$) [3, 4], or to the infinite cylinder ($h \gg R$) [4, 5]. Exact solutions for the case of an isotropic finite cylinder were given by Sundara Raja Iyengar and Chandrashekhara [6].

They assumed a symmetrical and an asymmetrical axial distribution of temperature; at the side surface of a cylinder $T(R, z) = 0$ was assumed. For the orthotropic case some simpler problems were elaborated: infinite space, semispace, elastic zone [7]. Sharma based his development [8] on the potential of thermoelastic deformations. Mossakowska and Nowicki [7] treated the problem more generally: besides the specific solution they gave the methodology for a general solution. They assumed that the thermal field is stationary and, moreover, that it does not influence the thermal and mechanical properties of a solid.

2. *Simplifying assumptions*

The complete and exact solution of the problem of thermoelastic stresses consists of a sum of two solutions: specific and general. The first is obtained from the potential of thermoelastic strains ψ. This potential should fulfil the following equation

$$\Delta\psi = T(r, z). \tag{2}$$

The stresses in an isotropic medium in cylindrical coordinates may be obtained from the formulae

$$\sigma'_r = -2G\left(\frac{1}{r}\frac{\partial\psi}{\partial r} + \frac{\partial^2\psi}{\partial z^2}\right); \quad \sigma'_\varphi = -2G\left(\frac{\partial^2\psi}{\partial r^2} + \frac{\partial^2\psi}{\partial z^2}\right)$$

$$\sigma'_z = -2G\left(\frac{\partial^2\psi}{\partial r^2} + \frac{1}{r}\frac{\partial\psi}{\partial r}\right); \quad \sigma'_{rz} = 2G\frac{\partial^2\psi}{\partial r\partial z}. \tag{3}$$

The general solutions for the stresses may be obtained by means of Love's functions for stresses with the help of formulae:

$$\sigma_r'' = \frac{\partial}{\partial z}\left(\nu\Delta\phi - \frac{\partial^2\phi}{\partial r^2}\right); \quad \sigma_\varphi'' = \frac{\partial}{\partial z}\left(\nu\Delta\phi - \frac{1}{r}\frac{\partial\phi}{\partial r}\right)$$

$$\sigma_z'' = \frac{\partial}{\partial z}\left[(2-\nu)\Delta\phi - \frac{\partial^2\phi}{\partial z^2}\right]; \quad \sigma_{rz}'' = \frac{\partial}{\partial r}\left((1-\nu)\Delta\phi - \frac{\partial^2\phi}{\partial z^2}\right). \tag{4}$$

The Love function should be chosen so as to satisfy the equation

$$\Delta^2\phi = 0. \tag{5}$$

In the present paper, in order to simplify calculations, the following was assumed:

a. the medium is orthotropic, i.e. its properties in the direction of pulling differ from the properties in the direction perpendicular to the one mentioned above (case of cross isotropy);

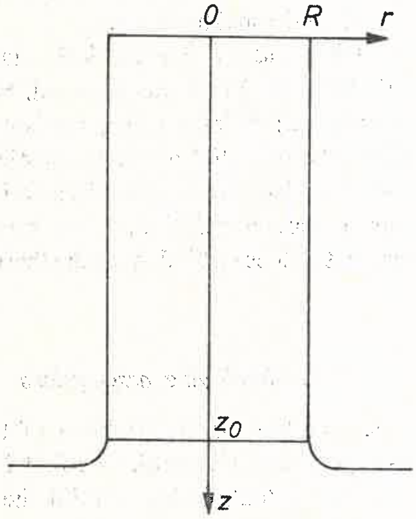

Fig. 2. Set of coordinates used for calculating the distribution of thermoelastic stresses in a single crystal

b. the calculations are carried out in cylindrical coordinates, as shown in Fig. 2. In Fig. 3 the definitions of individual stress components are given in both carthesian and cylindrical coordinates;

c. the crystal has a cylindrical shape with the generating line parallel to the direction of elongation. The axis of anisotropy of the crystal is in line with the generating line of the cylinder;

d. the mechanical properties of the material are independent of temperature;

e. the problem can be considered quasistationary, i.e. the process of redistribution of stresses does not depend significantly on the velocity of crystal growth;

f. the problem is characterized by an azimuthal symmetry, which means that the stress components do not depend on the angle φ.

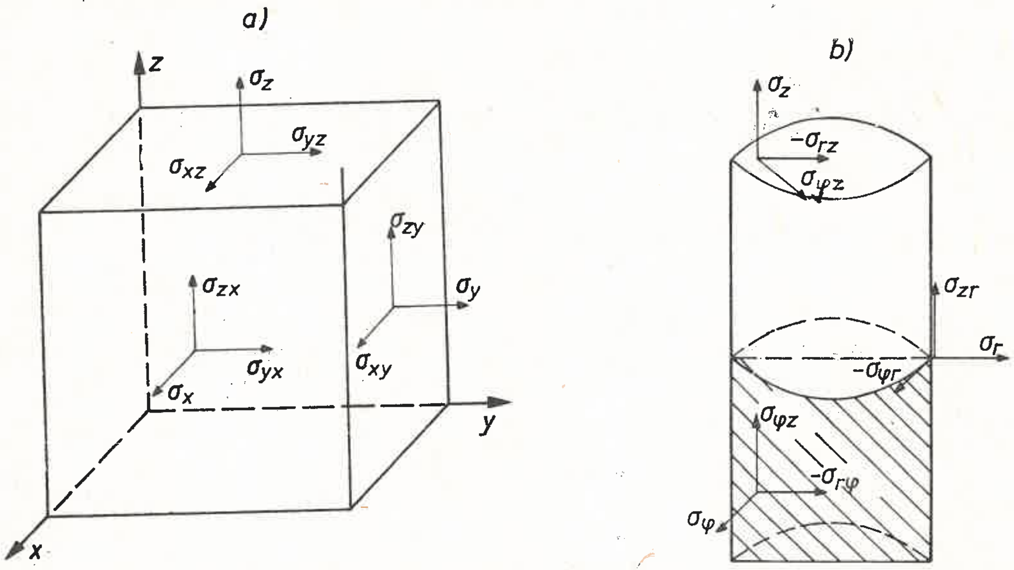

Fig. 3. Definitions of components of thermoelastic stresses: a — in carthesian coordinates, b — in cylindrical coordinates

3. Potential of thermoelastic deformations

The solution of the problem of thermoelastic stresses in single crystals is based only on the potential of thermoelastic stresse (specific solution) which is assumed to be of the form

$$\psi = \sum_{n=1}^{\infty} B_n J_0\left(\frac{\beta_n}{\sqrt{\kappa_r}}r\right) \operatorname{sh}\left(\frac{\beta_n}{\sqrt{\kappa_z}}z\right). \tag{6}$$

If the temperature distribution in a single crystal is [9]

$$T(r, z) = T_0 + (T_t - T_0) \sum_{n=1}^{\infty} A_n J_0\left(\frac{\beta_n}{\sqrt{\kappa_r}}r\right) \frac{\operatorname{sh}\left(\frac{\beta_n}{\sqrt{\kappa_z}}z\right)}{\operatorname{sh}\left(\frac{\beta_n}{\sqrt{\kappa_z}}z_0\right)} \tag{7}$$

then ψ fulfils condition (2) if

$$B_n = \frac{\kappa_r\kappa_z}{\kappa_r - \kappa_z} \frac{A_n}{\beta_n^2} \frac{T_t - T_0}{\operatorname{sh}\left(\frac{\beta_n}{\sqrt{\kappa_z}}z_0\right)}. \tag{8}$$

Now, according to Mossakowska and Nowacki [7], we determine the stresses according to the relations given by the authors mentioned above.

$$\sigma_{rr} = \gamma c_{33} c_{44} \left[\frac{\partial^2}{\partial z^2} \left(b\Delta_r + e \frac{\partial^2}{\partial z^2} \right) - \frac{q}{r} \frac{\partial}{\partial r} \left(\Delta_r + c \frac{\partial^2}{\partial z^2} \right) \right] \psi + \sum_{n=1}^{\infty} K_n \operatorname{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) \tag{9}$$

$$\sigma_{\varphi\varphi} = \gamma c_{33} c_{44} \left[\frac{\partial^2}{\partial z^2} \left(b\Delta_r + e \frac{\partial^2}{\partial z^2} \right) - q \frac{\partial^2}{\partial r^2} \left(\Delta_r + c \frac{\partial^2}{\partial z^2} \right) \right] \psi + \sum_{n=1}^{\infty} L_n \operatorname{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) \tag{10}$$

$$\sigma_{zz} = \gamma c_{33} c_{44} \left[\Delta_r \left(b\Delta_r + e \frac{\partial^2}{\partial z^2} \right) \right] \psi + \sum_{n=1}^{\infty} M_n J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \tag{11}$$

$$\sigma_{rz} = -\gamma c_{33} c_{44} \left[\frac{\partial^2}{\partial r \partial z} \left(b\Delta_r + e \frac{\partial^2}{\partial z^2} \right) \right] \psi + \sum_{n=1}^{\infty} N_n J_1 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \tag{12}$$

where

$$\gamma = \frac{(\alpha_r + \alpha_z \nu_z) E_r E_z}{(1-\nu_r) E_z - 2\nu_z^2 E_r} \tag{13}$$

$$e = \frac{2\nu_z E_r \alpha_r - (1-\nu_r) E_z \alpha_z}{E_r(\alpha_r - \alpha_z \nu_z)} \frac{c_{13}}{c_{33}} - 1 \tag{14}$$

$$c = \frac{c_{33}}{c_{44}} - \frac{2\nu_z E_r \alpha_r - (1-\nu_r) E_z \alpha_z}{E_r(\alpha_r - \alpha_z \nu_z)} \left(1 + \frac{c_{13}}{c_{44}} \right) \tag{15}$$

$$b = \frac{c_{13}}{c_{33}} - \frac{2\nu_z E_r \alpha_r - (1-\nu_r) E_z \alpha_z}{E_r(\alpha_r - \alpha_z \nu_z)} \tag{16}$$

$$\Delta_r = \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} \tag{17}$$

$$q = 2 \frac{c_{66}}{c_{33}}. \tag{18}$$

We thus obtain

$$\sigma_{rr} = \gamma c_{33} c_{44} \left\{ \sum_{n=1}^{\infty} \frac{B_n \beta_n^4}{\kappa_z} \left(\frac{e}{\kappa_z} - \frac{b}{\kappa_r} \right) J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \operatorname{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) \right.$$
$$\left. + \sum_{n=1}^{\infty} \frac{B_n \beta_n^4}{2\kappa_r} q \left(\frac{c}{\kappa_z} - \frac{1}{\kappa_r} \right) \left[J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) - J_2 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \right] \operatorname{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) \right\}$$
$$+ \sum_{n=1}^{\infty} K_n \operatorname{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) \tag{19}$$

$$\sigma_{\varphi\varphi} = \gamma c_{33} c_{44} \left\{ \sum_{n=1}^{\infty} \frac{B_n \beta_n^4}{\kappa_z} \left(\frac{e}{\kappa_z} - \frac{b}{\kappa_r} \right) J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \mathrm{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) \right.$$

$$\left. + \sum_{n=1}^{\infty} \frac{B_n \beta_n^4}{2\kappa_r} q \left(\frac{c}{\kappa_z} - \frac{1}{\kappa_r} \right) \left[J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) + J_2 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \right] \mathrm{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) \right\}$$

$$+ \sum_{n=1}^{\infty} L_n \,\mathrm{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) \tag{20}$$

$$\sigma_{zz} = -\gamma \sum_{n=1}^{\infty} \frac{B_n \beta_n^4}{\kappa_r} \left(\frac{e}{\kappa_z} - \frac{b}{\kappa_r} \right) J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \mathrm{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) + \sum_{n=1}^{\infty} M_n J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \tag{21}$$

$$\sigma_{rz} = \gamma \sum_{n=1}^{\infty} \frac{B_n \beta_n^4}{\sqrt{\kappa_r \kappa_z}} \left(\frac{e}{\kappa_z} - \frac{b}{\kappa_r} \right) J_1 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \mathrm{ch} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z \right) + \sum_{n=1}^{\infty} N_n J_1 \left(\frac{\beta_n}{\sqrt{\kappa_r}} r \right) \tag{22}$$

since the crystal is not subjected to external tensions, the following boundary conditions should hold:

$$\sigma_{rr} = 0 \quad \text{and} \quad \sigma_{\varphi\varphi} = 0 \quad \text{for} \quad r = R$$

$$\sigma_{zz} = 0 \quad \text{and} \quad \sigma_{rz} = 0 \quad \text{for} \quad z = z_0. \tag{23}$$

From the above relations the constants K_n, L_n, M_n, N_n are determined.

$$K_n = -B_n \beta_n^4 \left\{ \frac{1}{\kappa_z} \left(\frac{e}{\kappa_z} - \frac{b}{\kappa_r} \right) J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} R \right) + \frac{q}{2\kappa_r} \left(\frac{c}{\kappa_z} - \frac{1}{\kappa_r} \right) \right.$$

$$\left. \times \left[J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} R \right) - J_2 \left(\frac{\beta_n}{\sqrt{\kappa_r}} R \right) \right] \right\} \tag{24}$$

$$L_n = -B_n \beta_n^4 \left\{ \frac{1}{\kappa_z} \left(\frac{e}{\kappa_z} - \frac{b}{\kappa_r} \right) J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} R \right) + \frac{q}{2\kappa_r} \left(\frac{c}{\kappa_z} - \frac{1}{\kappa_r} \right) \right.$$

$$\left. \times \left[J_0 \left(\frac{\beta_n}{\sqrt{\kappa_r}} R \right) + J_2 \left(\frac{\beta_n}{\sqrt{\kappa_r}} R \right) \right] \right\} \tag{25}$$

$$M_n = \frac{B_n \beta_n^4 \gamma}{\kappa_r} c_{33} c_{44} \left(\frac{e}{\kappa_z} - \frac{b}{\kappa_r} \right) \mathrm{sh} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z_0 \right) \tag{26}$$

$$N_n = -\frac{B_n \beta_n^4 \gamma}{\sqrt{\kappa_r \kappa_z}} c_{33} c_{44} \left(\frac{e}{\kappa_z} - \frac{b}{\kappa_r} \right) \mathrm{ch} \left(\frac{\beta_n}{\sqrt{\kappa_z}} z_0 \right). \tag{27}$$

Expressions (19)—(22) and the constants determined by (24)—(27) form the basis for calculation of thermoelastic stress distributions in single crystals of lithium niobate and yttrium-aluminium garnet with the aid of a computer [10].

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