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The crossflow piezo-electrooptic effect in crystals. Example of lithium tantalate

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In the present paper the thermodynamic and phenomenological descriptions of the crossflow piezo-electrooptic effect in crystals have been made. For example the necessary experimental measurements of this effect have been carried out in the lithium tantalate crystals. For these crystals at first the $P_{33113} - P_{11113} = -4.6 \cdot 10^{-19}$ m^3/ N·V absolute coefficients difference of crossflow piezo-electrooptic effect were determined by two measurement methods.

Key words: *thermodynamic and phenomenological descriptions, crossflow piezo-electrooptic effect, lithium tantalate crystals, induced birefringence*

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1. Introduction

The crossflow piezo-electrooptic effect, resulting from mutual interaction of piezo- and electrooptic effects in crystal materials, is poorly investigated because of its small values [1] on the background of a direct action of the piezo- or electrooptic effects.

The aim of the present work is to carry out thermodynamic and phenomenological description of the crossflow piezo-electrooptic effect and make necessary experimental measurements for its verification.

2. Thermodynamic description

According to the general thermodynamic theory [2–4], a crystal appears as a thermodynamic system the monocrystal state of which is determined by certain numbers of variables. Mechanical stress σ_{ij}, electrical field strength E_i and temperature T are chosen as independent variables. Herein Gibbs free energy G is assumed as thermodynamic function for which [1,5]:

$$G = U - \boldsymbol{\varepsilon\sigma} - \boldsymbol{E}(\boldsymbol{D}\ /4\pi) - TS, \tag{1}$$

where U is the inner energy. Then, the components of the mechanical deformation ε_{ij}, electrical field induction D_i and entropy S will be functions of independent variables. Let's spread in the MacLaurin's series the ε_{ij} and D_i up to the third order of smallness (no thermal effects are taken into account) because their meanings of the second and the third order of smallness are insignificant [6]). Setting for convenience $\delta_m = D_m/4\pi$, we will have:

$$\begin{aligned}\varepsilon_{kl} = {} & \frac{\partial \varepsilon_{kl}}{\partial \sigma_{ij}}\sigma_{ij} + \frac{\partial \varepsilon_{kl}}{\partial E_n}E_n + \frac{1}{2}\left[\frac{\partial^2 \varepsilon_{kl}}{\partial \sigma_{ij}\partial \sigma_{qr}}\sigma_{ij}\sigma_{qr} + 2\frac{\partial^2 \varepsilon_{kl}}{\partial \sigma_{ij}\partial E_n}\sigma_{ij}E_n + \frac{\partial^2 \varepsilon_{kl}}{\partial E_n \partial E_o}E_nE_o\right] \\ & + \frac{1}{6}\left[\frac{\partial^3 \varepsilon_{kl}}{\partial \sigma_{ij}\partial \sigma_{qr}\partial \sigma_{st}}\sigma_{ij}\sigma_{qr}\sigma_{st} + 3\frac{\partial^3 \varepsilon_{kl}}{\partial \sigma_{ij}\partial \sigma_{qr}\partial E_n}\sigma_{ij}\sigma_{qr}E_n \right. \\ & \left. +3\frac{\partial^3 \varepsilon_{kl}}{\partial \sigma_{ij}\partial E_n\partial E_o}\sigma_{ij}E_nE_o + \frac{\partial^3 \varepsilon_{kl}}{\partial E_n\partial E_o\partial E_p}E_nE_oE_p\right],\end{aligned} \tag{2}$$

$$\begin{aligned}\delta_m = {} & \frac{\partial \delta_m}{\partial \sigma_{ij}}\sigma_{ij} + \frac{\partial \delta_m}{\partial E_n}E_n + \frac{1}{2}\left[\frac{\partial^2 \delta_m}{\partial \sigma_{ij}\partial \sigma_{qr}}\sigma_{ij}\sigma_{qr} + 2\frac{\partial^2 \delta_m}{\partial \sigma_{ij}\partial E_n}\sigma_{ij}E_n + \frac{\partial^2 \delta_m}{\partial E_n \partial E_o}E_nE_o\right] \\ & + \frac{1}{6}\left[\frac{\partial^3 \delta_m}{\partial \sigma_{ij}\partial \sigma_{qr}\partial \sigma_{st}}\sigma_{ij}\sigma_{qr}\sigma_{st} + 3\frac{\partial^3 \delta_m}{\partial \sigma_{ij}\partial \sigma_{qr}\partial E_n}\sigma_{ij}\sigma_{qr}E_n \right. \\ & \left. +3\frac{\partial^3 \delta_m}{\partial \sigma_{ij}\partial E_n\partial E_o}\sigma_{ij}E_nE_o + \frac{\partial^3 \delta_m}{\partial E_n\partial E_o\partial E_p}E_nE_oE_p\right].\end{aligned} \tag{3}$$

Besides, the following equations are valid [5]:

$$\frac{\partial G}{\partial \sigma_{kl}} = -\varepsilon_{kl}, \qquad \frac{\partial G}{\partial E_m} = -\delta_m, \qquad \frac{\partial \delta_m}{\partial \sigma_{kl}} = \frac{\partial \varepsilon_{kl}}{\partial E_m}. \tag{4}$$

Considerations of the physical sense of the partial derivatives are given more in full in [1]. We will take into consideration only components describing crossflow effects. They are:

$$A_{klijn} = \frac{\partial^2 \varepsilon_{kl}}{\partial \sigma_{ij}\partial E_n} = -\frac{\partial^3 G}{\partial \sigma_{ij}\partial \sigma_{kl}\partial E_n} = \frac{\partial S_{klij}}{\partial E_n} \equiv \frac{\partial^2 \delta_n}{\partial \sigma_{ij}\partial \sigma_{kl}} = \frac{\partial d_{nij}}{\partial \sigma_{kl}}. \tag{5}$$

It is a correction part to the elastic compliance constant S_{klij}, which is connected with the electrical field E_n acting in the crystal, or the same, correction part to the reverse piezoelectric effect constant d_{nij} under the action of the mechanical stress σ_{ij}. It is fifth-rank tensor, which describes a crossflow piezo-electrostrictive effect caused in the crystal by mutual interaction of the electrical field and the mechanical stress. Furthermore:

$$\begin{aligned}B_{mnijo} & = \frac{\partial^3 \delta_m}{\partial \sigma_{ij}\partial E_n\partial E_o} = -\frac{\partial^4 G}{\partial \sigma_{ij}\partial E_m\partial E_n\partial E_o} = \frac{\partial \mu_{mnij}}{\partial E_o} \equiv \frac{\partial}{\partial E_o}\left(\frac{\partial k_{mn}}{\partial \sigma_{ij}}\right) \\ & = \frac{\partial}{\partial \sigma_{ij}}\left(\frac{\partial k_{mn}}{\partial E_o}\right) = \frac{\partial \rho_{mno}}{\partial \sigma_{ij}}.\end{aligned} \tag{6}$$

Here k_{mn} – permittivity tensor and B_{mnijo} – a tensor of crossflow piezo-electrooptic effect, which determine the change of the piezooptic module $\mu_{mnij} = \partial k_{mn}/\partial \sigma_{ij}$ under the action of the electrical field E_o, or the same, the change of the liner electrooptic effect tensor $\rho_{mno} = \partial k_{mn}/\partial E_o$ under the action of the mechanical stress σ_{ij}.

Then, as parts of the second order of smallness are neglected, the simplified correlations (2) and (3) are:

$$\varepsilon_{kl} = S_{klij}\sigma_{ij} + d_{nkl}E_n + A_{klijn}\sigma_{ij}E_n, \tag{7}$$

$$D_m = 4\pi d_{mij}\sigma_{ij} + E_n[k_{mn} + \mu_{mnij}\sigma_{ij} + \rho_{mno}E_o/2 + B_{mnijo}\sigma_{ij}E_o/2], \tag{8}$$

where d_{mij} is a tensor of piezoelectric effect.

3. Phenomenological description

In the given work, the phenomenological description of a crossflow effect needed for the explanation of our experimental measurements is carried out.

As is generally known, the external action (be it the mechanical stress σ_{mn} or the external electric field E_l) exerted on the crystal sample results in the change of birefringence $\delta(\Delta n_k) = \delta n_i - \delta n_j$ (or refractive indices $\delta n_i, \delta n_j$) of the sample as well as its length δt_k in the direction k of light propagation, which are registered by polarization-optic method through the change of an optical path difference $\delta\Delta_k$ of this sample:

$$\delta\Delta_k = \delta(\Delta n_k t_k) = t_k\delta(\Delta n_k) + \Delta n_k \delta t_k. \tag{9}$$

The value $\delta(\Delta n_k)$ or $\delta n_i, \delta n_j$ can be determined from the tensor for polarization constants a_{ij}. Beside the coefficients of piezo- π_{ijmn} and linear electrooptic r_{ijl} effects, this tensor in the first approximation for acentric crystals also contains the coefficients of their crossflow effect [1], analogous to (8):

$$\Delta a_{ij} = \pi_{ijmn}\sigma_{mn} + r_{ijl}E_l + P_{ijmnl}(\sigma_{mn}E_l), \tag{10}$$

where $P_{ijmnl} = \partial\pi_{ijmn}/\partial E_l = \partial r_{ijl}/\partial\sigma_{mn}$ is the tensor of crossflow piezo-electrooptic effect (tensor of absolute coefficients), $\sigma_{mn}E_l = g_{mnl}$ is the 3-rank tensor, which equals the product of the 2-rank tensor σ_{mn} and vector E_l. From a thermodynamic description of crossflow piezo-electrooptic effect P_{mnijl} (see formula (6)), one can describe symmetrical properties of this tensor while replacing the indexes $P_{ijmnl} = P_{jimnl} = P_{ijnml} = P_{ijlmn}$. The complete form of this tensor is given in [7].

Analogously, the value δt_k is determined from the deformation tensor ε_{kl} (see formula (7)), which beside the coefficients of elastic S_{klij} and piezoelectric d_{nkl} effects also contains the coefficients of their crossflow A_{klijn} effect. This crossflow A_{klijn} effect is named "false" effect in our case.

To derive the necessary operating correlations we use the matrix interpretation of the processed tensors. For a crystal with small initial birefringence the correlation (9) will be simplified to $\delta\Delta_k = t_k\delta(\Delta n_k)$, where the $\delta(\Delta n_k)$ value is equal to [5,8]:

$$\delta(\Delta n_k) = -\pi^*_{km}\sigma_m/2. \tag{11}$$

Here $\pi^*_{km} = \pi_{im}n_i^3 - \pi_{jm}n_j^3$ is a known piezooptic coefficient of the induced birefringence. For half-wave stresses σ^o_m, when $\delta\Delta_k = \lambda/2$ (λ is a length of light wave), one can obtain:

$$\lambda/2 = -\pi^*_{km}\sigma^o_m t_k/2. \tag{12}$$

After applying the electric field E_l, for repeatedly measured half-wave stresses $\sigma^{o'}_m$, we obtain:

$$\lambda/2 = -\pi^*_{km}\sigma^{o'}_m t_k/2 - P^*_{kml}\sigma^{o'}_m E_l t_k/2. \tag{13}$$

Analogously $P^*_{kml} = P_{iml}n_i^3 - P_{jml}n_j^3$ is the crossflow piezo-electrooptic coefficient of the induced birefringence. Mutual solution of (12) and (13) gives us the value of this coefficient to be searched for:

$$P^*_{kml} = \lambda[(\sigma^{o'}_m/\sigma^o_m) - 1]/(\sigma^{o'}_m E_l t_k). \tag{14}$$

A similar formula can be obtained at measuring the magnitudes of half-wave electric fields E^o_l and $E^{o'}_l$ for $\sigma_m = 0$ and $\sigma_m \neq 0$ accordingly.

4. Experimental results

The measurements of crossflow piezo-electrooptic effect were carried out using the polarisation-interferometrical technique by Senarmont method and half-wave stresses method [5].

To exclude a possible error, connected with the measurement of "false" crossflow effect, the $LiTaO_3$ crystals with a small initial birefringence ($\Delta n_k = 0,005$) were used. For these crystals, the elastic component in (9) and therefore "false" crossflow effect could be neglected.

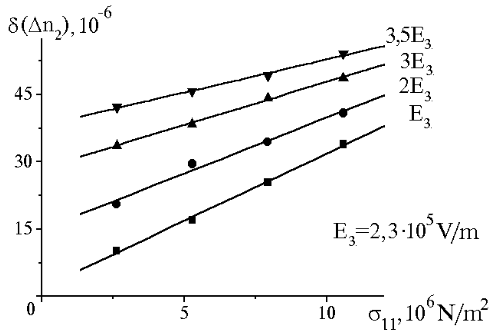

Figure 1. The dependences of birefringence change $\delta(\Delta n_2)$ for lithium tantalate crystals on the normal mechanical stress σ_{11} for different magnitudes of electrical fields E_3 (for light wave $\lambda = 0,6328$ μm and temperature T=293 K).

We have determined the crossflow piezo-electrooptic effect P^*_{22113} component on the sample of direct cut, when $k \parallel Y$, $\sigma_m \parallel X$ and $E_l \parallel Z$. In the figures the dependences for the $\delta(\Delta n_2)$ value under the normal mechanical stress σ_{11} for different magnitudes of E_3 (figure 1) are shown as well as the $\delta(\Delta n_2)$ value under the electrical field E_3 for different magnitudes of σ_{11} (figure 2). From the changes of angular coefficients for linear interpolation of these behaviours we have calculated the average magnitudes of $P^*_{22113} = -5.0\cdot10^{-18}$ m^3/N $\cdot$ V (from figure 1) and $P^*_{22113} = -4.4\cdot10^{-18}$ m^3/N $\cdot$ V (from figure 2).

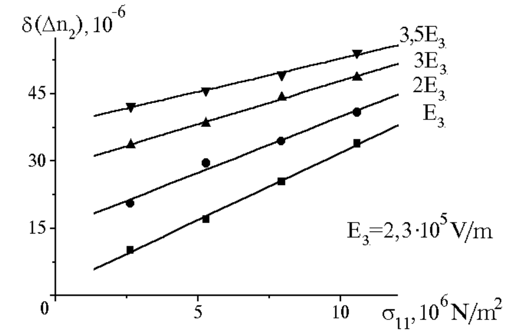

Figure 2. Analogous to figure 1 the dependences of $\delta(\Delta n_2)$ on the electrical fields E_3 for different magnitudes of mechanical stress σ_{11}.

The difference of these coefficients can be explained by the induced rotation of the optical indicatrix around the X axis under the action of the mechanical stress σ_{11}. It leads to the change of extraordinary index n_e of light wave according to the known equation:

$$\delta n_e = n_e(1 - \cos(\pi_{41}\sigma_{11}/(n_e^{-2} - n_o^{-2})). \quad (15)$$

Taking this into account, we have found the new magnitude of $P^*_{22113} = -4.7 \cdot 10^{-18}$ $\mathrm{m^3/N \cdot V}$ from figure 1.

This coefficient P^*_{22113} has also been determined from the method of half-wave stresses. Having determined the half-wave mechanical stress σ^o_m for $E_l = 0$ and $\sigma^{o'}_m$ upon the action of electrical field $E_l \neq 0$, we have calculated the magnitude P^*_{kkmml} according to formula (14), which was equal $P^*_{22113} = -5.1 \cdot 10^{-18}$ $\mathrm{m^3/N \cdot V}$.

It is also noted that from P^*_{22113} one can calculate the absolute coefficients combination of crossflow piezo-electrooptic effect (taking into account that $n_o \approx n_e = 2.175$): $P_{33113} - P_{11113} \approx P^*_{22113}/n_o^3 = -4.6 \cdot 10^{-19}$ $\mathrm{m^3/N \cdot V}$.

Besides, according to (6): $P^*_{22113} = \partial\pi^*_{2211}/\partial E_3 \approx \Delta\pi^*_{2211}/\Delta E_3$, and therefore at the electrical field change of $\Delta E_3 = 2.3 \cdot 10^5$ V/m the change of piezooptic coefficient is equal $\Delta\pi^*_{2211} = P^*_{22113}\Delta E_3 = 1.1 \cdot 10^{-12}$ $\mathrm{m^2/N}$. For comparison, the magnitude of coefficient π^*_{2211} from our measurements is equal to $10.4 \cdot 10^{-12}$ $\mathrm{m^2/N}$.

5. Conclusions

1. The thermodynamic descriptions of crossflow piezo-electrooptic effect in crystals and the necessary phenomenological descriptions for anisotropic materials with small initial birefringence have been made in the paper.

2. For lithium tantalate crystals, at first the $P_{33113} - P_{11113} = -4.6 \cdot 10^{-19} \mathrm{m^3/N \cdot V}$ absolute coefficients difference of crossflow piezo-electrooptic effect were determined. For these crystals, the change of $\Delta\pi^*_{2211}$ because of crossflow effect corresponds to $\sim 10\%$ of coefficient π^*_{2211} at electrical field change of $\Delta E_3 = 2.3 \cdot 10^5$ V/m.

3. The equality (within the limits of experimental error) of the P^*_{22113} coefficient, obtained from different measurement methods, demonstrated the correctness of our results and showed the possibility of measuring the crossflow piezo-electrooptic effect in anisotropic crystals with small initial birefringence using these methods.

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Перехресний п'єзо-електрооптичний ефект на прикладі кристалів танталату літію

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У даній роботі було здійснено термодинамічний і феноменологічний описи перехресного п'єзо-електрооптичного ефекту в кристалах. Для прикладу були проведені необхідні експериментальні вимірювання вказаного ефекту в кристалах танталату літію. Для цих кристалів на основі двох різних методів вимірювання вперше було визначено різницю абсолютних коефіцієнтів перехресного п'єзо-електрооптичного ефекту $P_{33113} - P_{11113} = -4,6 \cdot 10^{-19}$ м^3/Н·В.

Ключові слова: *термодинамічний та феноменологічний описи, перехресний п'єзо-електрооптичний ефект, кристал танталату літію, індуковане двозаломлення*

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