
Baric Changes in the Optical Indicatrix of TGS Crystals with L-threonine Admixture

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Abstract

The influence of uniaxial mechanical stresses, applied along the principal crystal-physical axes, on the principal values of birefringence Δn_i is studied for TGS crystals with 5% L-threonine admixture. It is revealed that Δn_i are quite sensitive to the mechanical stresses. The baric coefficients $\partial T_c / \partial \sigma_m$ for the phase transition temperature appear to be less than those for the pure TGS. Temperature dependences and dispersion of the piezooptic coefficients are calculated. The jump-like changes in all of the piezooptic coefficients occurring at the Curie temperature in the doped crystals are explained with the contributions of a secondary electrooptic effect.

Key words: birefringence, uniaxial pressure, phase transitions, piezooptic coefficients

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Introduction

Triglycine sulphate $(\text{NH}_2\text{CH}_2\text{COOH})_3 \cdot \text{H}_2\text{SO}_4$ crystals (abbreviated as TGS) are among the most known organic ferroelectric materials which are used as pyroelectric infrared sensors [1]. Introduction of amino acids (L- and D-alanine, L- and DL- tryptophan, D-phenylalanine, as well as L-threonine (the LTTGS crystals)) into TGS leads to improvement of their infrared parameters [1], fixing the spontaneous polarization and appearance of the biasing field E_b [2-5]. On the other side, it is known [6] that the TGS crystals are characterized by large piezooptic (PO) coefficients and since can be used in the optical devices utilizing the PO effect.

The present paper is devoted to studies of the influence of L-threonine dopant on the PO properties of TGS crystals.

Experimental

The temperature dependences and dispersion of the birefringence have been measured with the

combined interferometric-polarimetric technique. The sample has been placed between the crossed polarizers in the “diagonal” position and the uniaxial mechanical pressure has been applied with the help of a special device, allowing application of stresses as high as ~ 200 bar. At each temperature or light wavelength, the birefringence change caused by the mechanical stress is defined by the relation

$$\Delta n_i(\sigma_m) = k(\sigma_m) \frac{\lambda}{d_i(\sigma_m)}, \quad (1)$$

where Δn_i is the birefringence value along the i axis, k the order of the interference maximum, λ the light wavelength in vacuum, and d_i the sample thickness along the light propagation direction.

Results and discussions

The baric dependences of birefringence in the TGS crystals doped with 5% L-treonine at the room temperature and $\lambda=500$ nm are shown in Figure 1. One can see that the Δn value changes

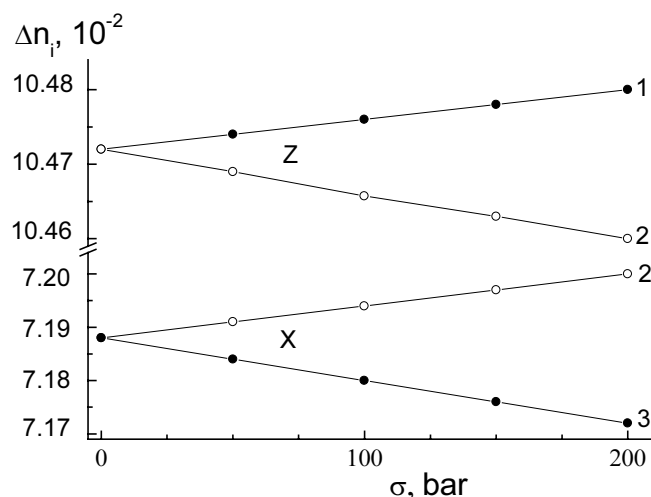


Fig. 1. Baric birefringence change in the TGS crystal doped with 5% L-treonine at the room temperature and $\lambda=500$ nm: 1 - σ_x , 2 - σ_y , and 3 - σ_z . Z and X mean the directions of light propagation along the principal axes of the optical indicatrix.

linearly with stress increasing. The σ_x and σ_y stresses lead to different values and opposite signs of the Δn_z changes.

The temperature changes in the principal values of the birefringence for LTTGS are depicted in Figure 2.

The general features are as follows: the uniaxial stresses change the absolute value of Δn_i , though remain a qualitative character of $\Delta n_i(T)$ unchanged, while the phase transition (PT) temperature is shifted under the stress

influence. The PT temperature decreases for the cases of σ_x and σ_y stresses ($T_c^x=321.7$ K and $T_c^y=321.4$ K) and increases when the σ_z stress is applied ($T_c^z=326.6$ K). The relevant temperature shift coefficients are equal to $\partial T_c/\partial \sigma_x = -0.0065$, $\partial T_c/\partial \sigma_y = -0.0081$ and $\partial T_c/\partial \sigma_z = 0.0179$ K/bar. The “total” (or hydrostatic) coefficient for the PT temperature shift is equal to

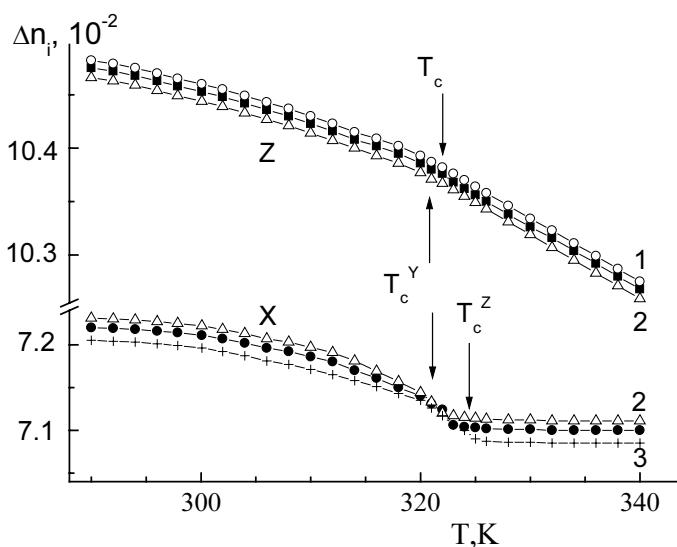
$$\frac{\partial T_c}{\partial p} = \sum_{i=1}^3 \frac{\partial T_c}{\partial \sigma_i} = +0.0033 \text{ K/bar}.$$


Fig. 2. Temperature dependences of birefringence in the TGS crystal doped with 5% L-treonine for differently directed uniaxial stresses and $\lambda=500$ nm: 1 - $\sigma_x=200$ bar, 2 - $\sigma_y=200$ bar, and 3 - $\sigma_z=200$ bar, respectively. Z and X mean the directions of light propagation along the principal axes of the optical indicatrix and the full circles correspond to $\sigma_m=0$.

The effective PO coefficients $\pi_{im}^0 = 0.5(n_m^3\pi_{mm} - n_k^3\pi_{km})$ have been calculated according to the formula

$$\pi_{im}^0 = \frac{2\delta(\Delta n)_i}{\sigma_m}, \quad (2)$$

where $\delta(\Delta n)_i$ represents the induced birefringence change for the light propagation direction along the i axis and the pressure applied along the m axis, π_{km} and π_{mm} the coefficients of the PO tensor, n_m and n_k the refractive indices, i , k and m the principal axes of the optical indicatrix ellipsoid. From Figure 3 it is obvious that the PO coefficients exhibit an anomalous behaviour and a jump-like change at T_c . The structure of the PO tensor in both the

paraelectric (point symmetry group $2/m$) and the ferroelectric (point group 2) phases is the same, while the PT is of a second order. Furthermore, one can come to conclusion that the anomalous temperature dependence of PO coefficient in the temperature range of ferroelectric phase is related to the secondary linear electrooptic effect, which is forbidden in centrosymmetric paraelectric phase. We have not really observed the temperature change in the PO coefficients within the paraelectric phase. Therefore, without the contribution of the secondary electrooptic effect, induced through a piezoelectric coupling, the temperature dependences of the PO coefficients around T_c should be continuous, due to the second order of the phase transition.

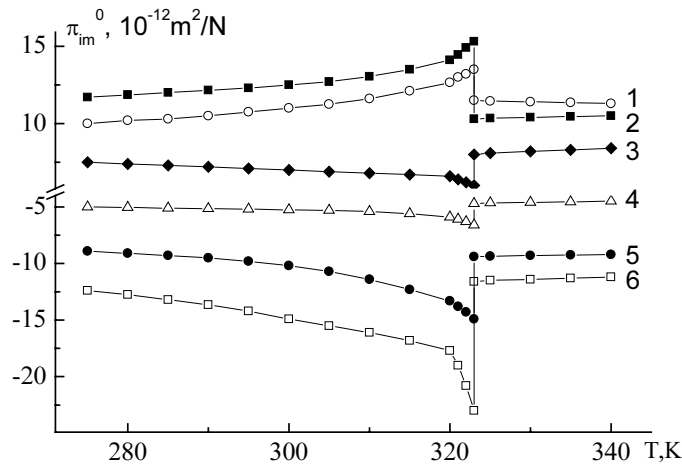


Fig. 3. Temperature dependences of PO coefficients for the TGS crystals doped with 5% L-threonine for $\lambda=500$ nm: 1 - π_{23}^0 , 2 - π_{12}^0 , 3 - π_{31}^0 , 4 - π_{21}^0 , 5 - π_{32}^0 , and 6 - π_{13}^0 .

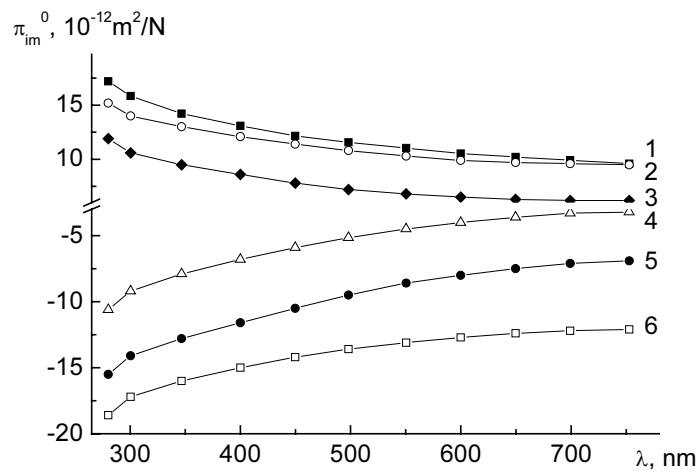


Fig. 4. Dispersion of PO coefficients of the TGS crystals doped with 5% L-threonine at the room temperature: 1 - π_{23}^0 , 2 - π_{12}^0 , 3 - π_{31}^0 , 4 - π_{21}^0 , 5 - π_{32}^0 , and 6 - π_{13}^0 .

The dispersion of PO constants in the LTTGS crystals at room temperature is shown in Figure 4. It is seen that the PO constants of the doped crystals manifest a weak dispersion in the visible spectral range. Let us notice an equality of the π_{23}^0 and π_{12}^0 coefficients: $\pi_{23}^0 = \pi_{12}^0 = 10.88 \times 10^{-12} \text{ m}^2/\text{N}$ at $\lambda = 628 \text{ nm}$. This means that the increment of the birefringences Δn_z and Δn_y under the action of σ_z and σ_y stresses would be equal.

Conclusion

Hence, in this work we have studied the influence of uniaxial mechanical stresses, applied along the principal axes of the optical indicatrix, on the principal birefringence values of the TGS crystals doped with 5% L-threonine admixture. It is revealed that the Δn_i parameters are sensitive to the action of uniaxial stresses. The baric coefficients of $\partial T_c / \partial \sigma_m$ of the PT shift are determined. They are slightly less than those for the pure TGS crystals. The temperature dependences and the dispersion of the effective PO coefficients π_{im}^0 are calculated. It is shown that the π_{im}^0 values in the doped crystals are somewhat larger than those in the pure crystals

at $293\text{K} < T < 320 \text{ K}$. The jump-like behaviour of PO coefficients at T_c is most probably associated with the contribution of a secondary electrooptic effect in the ferroelectric phase. The equality of the PO constants π_{23}^0 and π_{12}^0 is established.

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