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PACS 76.20.Hp

RELAXATION OF STATIC PHOTOELASTICITY IN LEAD GERMANATE CRYSTALS

The relaxation of static photoelasticity in lead germanate ($Pb_5Ge_3O_{11}$) crystals has been studied. The anisotropy of relaxation amplitudes and their anomalies near the phase transition point were revealed. The photoelastic relaxations and peculiarities in their temperature behavior are explained as manifestations of the piezocaloric mechanism.

Keywords: photoelasticity, photoelasticity relaxation, ferroelectrics.

1. Introduction

When studying the static photoelasticity in crystals, relaxation-induced changes of their optical parameters under the action of stationary mechanical stresses are not discussed, as a rule [1–3]. For many photoelastic and acousto-optic materials (paratellurite, lead molybdate, gallium phosphide, strontium borate, and others [4–7]), the transformation of mechanical stresses arising in a photoelastic element into a change of its optical parameters is assumed to take place without residual and relaxation phenomena up to the destroying mechanical stresses. It is the absence of photoelastic relaxations that determines a high Q-factor of photoelastic and acousto-optic light modulators.

The prospects for low-symmetry crystals to be used in acousto-optic modulators is associated with their high photoelastic coefficients [8,9]. However, the careful experiments with an error of 0.02–0.1% [10–12] demonstrate that the photoelastic relaxation (the afteraction) can be detected even at low mechanical stresses that amount to 5–10% of the destruction ones. In this work, the relaxation of static photoelasticity in ferroelectric lead germanate ($\mathrm{Pb_5Ge_3O_{11}}$) crystals is studied. The obtained results are compared with the photoelastic relaxation in model ferroelectric crystals belonging to the triglycine sulfate group.

2. Experiment Technique

Photoelastic (piezooptic) relaxation is a variation of the optical parameters of a specimen in time, provided that mechanical stresses are constant. We registered the dependences of the relaxation-induced changes in the stress-induced path difference $\delta\Delta_{km}^{\mathrm{rel}}$ on the time t, when the specimen was subjected to the action of constant mechanical stresses σ_m (the subscripts $k, m = 1, 2, 3$ correspond to the X_1, X_2, and X_3 axes of crystal's optical indicatrix and specify experimental conditions, namely, the directions of light propagation and uniaxial pressure action, respectively).

The relaxations are studied with the help of polarization-optical method [13] by determining the time-variations of the light intensity I in the linear section of the periodic dependence

$$I(\Delta_{km}) = (I_{\max} - I_{\min}) \sin^2 \frac{\pi}{\lambda} \Delta_{km} + I_{\min}. \qquad (1)$$

Here, $I_{\max}$ and $I_{\min}$ are the maximum and minimum, respectively, values of light intensity at the analyzer output, when the specimen undergoes the action of a mechanical stress σ_m; Δ_{km} the path difference (here, the subscripts k and m also mark experimental conditions), and $\lambda = 0.633$ μm is the light wavelength.

The relaxation amplitudes $\delta\Delta_{km}^{\mathrm{rel}}$ are determined on the basis of Eq. (1) as the difference $\Delta_{km}^{\sigma} - \Delta_{km}^{\tau}$, where Δ_{km}^{σ} is the path difference at the beginning of the action of a stress σ_m, and Δ_{km}^{τ} is the same parameter after the relaxation termination (after the time interval τ had passed).

The relative relaxation amplitude β_{km} is introduced by the relation [10]

$$\beta_{km} = \frac{\delta\Delta_{km}^{\mathrm{rel}}}{\delta\Delta_{km}} \times 100\% = -\frac{2\delta\Delta_{km}^{\mathrm{rel}}}{\pi_{km}^{\circ} d_k \sigma_m} \times 100\%, \qquad (2)$$

where $\delta\Delta_{km}$ is the path difference induced by the mechanical stress σ_m, π_{km}° are piezo-optic coefficients of the path difference, and d_k is the specimen thickness

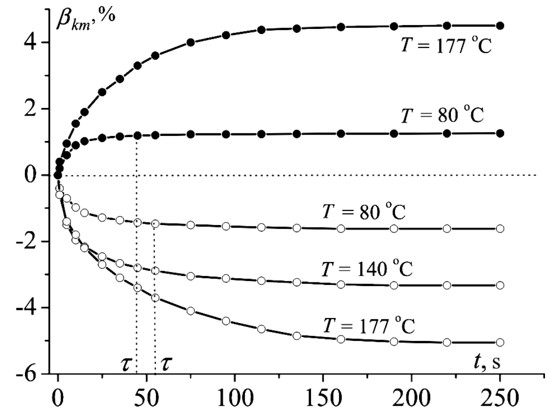

Fig. 1. Time dependences of the relative relaxation amplitudes β_{12} (solid circles) and β_{13} (hollow circles)

along the light propagation direction. The relaxation amplitude is assumed positive if the stress-induced, $\delta\Delta_{km}$, and relaxation-induced, $\delta\Delta_{km}^{\rm rel}$, path differences vary analogously, and negative if $\delta\Delta_{km}^{\rm rel}$ reduces the stress-induced path difference $\delta\Delta_{km}$.

For exact measurements of the amplitudes of relaxation-induced changes in the path differences induced by mechanical stresses, the following conditions were provided: 1) the stiffness of the mechanical part of the installation was enhanced to ensure a stationary mechanical compressive stress σ_m, 2) the intensity instability of a monochromatic radiation source during the time t of relaxation amplitude measurements did not exceed 0.01%, and 3) the accuracy of specimen temperature stabilization amounted to 0.01 °C.

Specimens of cubic lead germanate crystals about $5 \times 5 \times 5$ mm^3 in size are studied. The crystal facets were oriented normally to the optical indicatrix axes.

3. Experimental Results and Their Discussion

Unlike biaxial crystals, e.g., the crystals belonging to the triglycine sulfate group [10–12], we did not succeed in detecting the relaxation of static photoelasticity in lead germanate crystals at room temperature. In Fig. 1, the dependences of the relative relaxation amplitudes β_{km} on the time t at the action of a constant mechanical stress σ_m obtained at various temperatures starting from $T = 80$ °C are depicted. One can see that the maximum relaxation amplitudes determined in the saturation section of the $\beta_{km}(t)$ dependences are anisotropic both in the sign and the magnitude. The relaxation times τ corresponding to the beginning of saturation in the dependences $\beta_{km}(t)$ vary together with the experimental geometry and equal about 50 s at $T = 80$ °C. They weakly depend on σ_m and considerably grow, as the temperature increases, achieving a value of 200 s in a vicinity of the phase transition point $T_c = 177$ °C (Fig. 1).

The anisotropy of the quantity β_{km} is associated with the piezocaloric effect [10–12, 14]. Namely, a rapid variation of σ_m gives rise to the adiabatic change of the temperature of the specimen by $\delta T^{\rm rel}$, whose magnitude is proportional to σ_m. In a certain time interval τ (the time of relaxation owing to the heat exchange with the environment), the temperature of the specimen becomes equal to that of the environment. This relaxation-induced temperature change is responsible for the thermooptical variation of the stress-induced path difference by the quantity $\delta\Delta_{km}^{\rm rel}$. This mechanism of photoelastic relaxation is described by the relation

$$\delta\Delta_{km}^{\rm rel} = \frac{d\Delta_k}{dT}\delta T_m^{\rm rel} = \frac{d\Delta_k}{dT}\frac{dT_m^{\rm rel}}{d\sigma_m}\sigma_m, \tag{3}$$

where $d\Delta_k/dT$ is the thermo-optic coefficient of path difference Δ_k, $\delta T_m^{\rm rel}$ the piezocaloric variation of the specimen temperature (the subscript m in the notation $T_m^{\rm rel}$ means the dependence of the piezocaloric effect magnitude on the uniaxial pressure direction), and $dT_m^{\rm rel}/d\sigma_m$ the piezocaloric coefficient.

Note that the piezocaloric change of the crystal temperature was determined in this work on the basis of the known relation [15]

$$\delta T_m^{\rm rel} = -\frac{T_\circ}{C_p'}\alpha_m\sigma_m, \tag{4}$$

where $T_\circ$ is the specimen (and environment) temperature, C_p' the specific heat of the crystal per unit volume ($C_p' = \rho C_p$, C_p the heat capacity at a constant pressure, ρ the material density), and α_m are the temperature expansion coefficients. On the basis of Eqs. (3) and (4), the following expression is obtained for the relaxation-induced variation in the stress-induced path difference $\delta\Delta_{km}^{\rm rel}$:

$$\delta\Delta_{km}^{\rm rel} = -\frac{d\Delta_k}{dT}\frac{T_\circ}{C_p'}\alpha_m\sigma_m. \tag{5}$$

Expressions (2) and (5) substantiate the anisotropy of the quantities β_{km} (Fig. 1): 1) the value of β_{13}

is equal to the value of relaxation amplitude β_{23}, because the thermo-optic coefficients $d\Delta_1/dT$ and $d\Delta_2/dT$ are identical [16]; 2) the inequality $\beta_{12} < \beta_{13}$ between the relaxation amplitudes corresponding to the same thermo-optic coefficient $d\Delta_1/dT$ is a result of the inequality $\delta T_2^{\rm rel} < \delta T_3^{\rm rel}$; the latter, in accordance with Eq. (4), corresponds to the inequality $\alpha_1 = \alpha_2 < \alpha_3$ for the temperature expansion coefficients in the ferroelectric phase [17]; 3) our failure in the detection of relaxation-induced changes in a crystal at room temperature can be explained by both the small value of thermo-optic coefficient $d\Delta_1/dT$ and small coefficients of temperature expansion α_1 and α_3 in the ferroelectric phase far from the phase transition point.

The piezocaloric mechanism of photoelastic relaxations can also satisfactorily describe their temperature anomalies in the phase transition interval (Fig. 2). From Fig. 2, it follows that the experimental and calculated dependences $\beta_{13}(T)$ are in good agreement. THis is also valid for the dependence $\beta_{12}(T)$. While calculating $\beta_{km}(T)$ on the basis of formulas (2) and (5), the temperature dependences for the crystal density, $\rho(T)$, and specific heat, $C_p(T)$, were taken from works [18, 19], and the coefficients of temperature expansion $\alpha_m(T)$ from work [17], whereas the temperature dependences for the thermo-optic coefficients of the path difference $d\Delta_k/dT$ were determined on the basis of the dependences $\delta\Delta_k(T)/d_k$ presented in work [16].

Figure 2 demonstrates that the temperature anomalies of relaxation amplitude β_{km} in a vicinity of the phase transition temperature T_c in lead germanate crystals are substantially smaller in comparison with the characteristics typical of the crystals belonging to the triglycine sulfate group [11,12,14]. This fact, in accordance with Eq. (4), is associated with smaller anomalies of the thermo-optic coefficients $d\Delta_k/dT$ in the phase transition interval [16] and the linear expansion coefficients α_1 and α_3 [17].

The maximum values of relaxation amplitudes in various experimental geometries were observed on different sides from T_c (Fig. 2). This result can be explained by the fact that the piezo-induced shifts of the phase transition temperature have opposite signs for the mechanical stresses σ_1 and σ_3 [16]. Moreover, the magnitude of relaxation amplitude in the paraelectric phase of lead germanate crystals does not differ substantially from that in a vicinity of T_c in the ferroelectric. This circumstance is connected with large values of temperature expansion coefficients in the paraelectric phase [17].

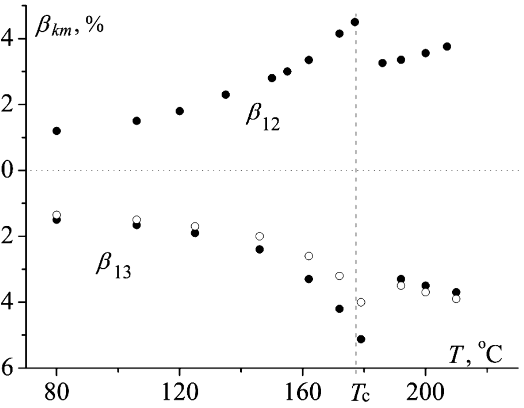

Fig. 2. Temperature anomalies of the relative relaxation amplitudes β_{km}: experimental (solid circles) and calculated values (hollow circles)

We would like to attract attention to the fact that no photoelasticity relaxation was detected in lead germanate crystals, as light propagates along the optical crystal axis. This fact originates from the zero values of thermo-optic coefficients in this direction.

4. Conclusions

In this work, we showed that the relaxations of static photoelasticity and their anisotropy in lead germanate crystals are well described on the basis of the piezocaloric mechanism. The temperature anomalies in the relative amplitudes of photoelastic relaxation near the phase transition point are connected with the anomalies in the thermo-optic coefficients and the coefficients of temperature expansion. At the same time, the absence of such relaxations far from the phase transition temperature (at $T < 80$ °C) is connected with too small values of thermo-optic coefficients for the crystal under study [16].

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Received 26.02.14.
Translated from Ukrainian by O.I. Voitenko

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РЕЛАКСАЦІЯ СТАТИЧНОЇ ФОТОПРУЖНОСТІ В КРИСТАЛАХ ГЕРМАНАТУ СВИНЦЮ

Резюме

Досліджено релаксацію статичної фотопружності в сегнетоелектричних кристалах германату свинцю. Виявлено анізотропію амплітуд релаксації, а також їх аномалії в околі фазового переходу. Фотопружні релаксації і особливості їх температурної поведінки пояснено в рамках п'єзокалоричного механізму.