Combined piezo-electrooptical effect in $Pb_5Ge_3O_{11}$ crystals induced by spontaneous polarization

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Abstract

In this paper combined piezo-electrooptical effect in $Pb_5Ge_3O_{11}$ crystals was studied. The temperature dependencies of birefringence change $\delta(\Delta n)_{13}$ for the lead germanate crystals at proper ferroelectrical phase transition were investigated under the influence of mechanical strain σ_{33}. It was shown that the application of compressive strain σ_{33} leads to the increasing of phase transition temperature and $\partial T_c/\partial\sigma_{33}=0.47\times10^{-6}$ Km^2/N. The temperature change of birefringence can be described by a quadratic electrooptical effect induced by spontaneous polarization. The calculated value of the Kerr effect coefficient for the paraelectrical phase is $n_3^3R_{3333}-n_1^3R_{1133}=0.9$ m^4/C^2.The temperature dependence of piezooptical constants for the lead germanate crystals $n_3^3\pi_{3333}-n_1^3\pi_{1133}$ was experimentally studied. It was shown that these dependences are described by a combined piezo-electrooptical effect induced by spontaneous polarization and external mechanical strain. The temperature change of $n_3^3\pi_{3333}-n_1^3\pi_{1133}$ is proportional to the square of P_s. The calculated value of the coefficients of combined piezo-electrooptical effect is $n_3^3N_{333333}-n_1^3N_{113333}==0.73\times10^{-8}$ $m^2/N\cdot C^2$.

Key words: combined effects, piezo-electrooptical effect, ferroelectrical phase transition, lead germanate crystals

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Introduction

To the combined effects of parametrical crystalloptics belong the phenomena that appeared under the mutual influence of external fields of different nature. One of these effects consist in the change of refractive index of crystals under the simultaneously applied electrical field and mechanical strain and can be described by the relation [1]

$$\Delta B_{ij}=M_{ijklm}\sigma_{kl}E_m+N_{ijklmn}\sigma_{kl}E_mE_n, \qquad (1)$$

where ΔB_{ij} - is the change of the tensor of polarization constants, σ_{kl} - is the mechanical strain tensor, E_m, E_n - strength of electrical field, M_{ijklm} , N_{ijklmn} - fifth and sixth rank polar tensors, respectively. Eq.(1) described linear and the quadratic combined piezo-electrooptical effect and can be rewritten as

$$\pi_{ijkl}=M_{ijklm}E_m+N_{ijklmn}E_mE_n \qquad (2)$$

or

$$r_{ijm}=M_{ijklm}\sigma_{kl},\ R_{ijmn}=N_{ijklmn}\sigma_{kl}. \qquad (3)$$

Eq.(2), (3) describe the change of the piezooptical and electrooptical coefficients under the

influence of electrical field and mechanical strain, respectively. Obviously this effects are of high order and coefficients M_{ijklm}, N_{ijklmn} should be quite small. In [2] we were reported about the measuring of piezo-electrooptical effect in $LiTaO_3$ crystals. The change of induced birefringence by mutual influence of electrical field and mechanical strain is 10% in comparesson with the piezooptical one. One of the possibilities of the investigation of this effect is the study of the temperature behavior of piezooptical coefficients at proper ferroelectrical phase transitions. In such a case the change of piezooptical coefficients could be induced by the spontaneous polarization P^s_m, P^s_n

$$\pi_{ijkl}=M'_{ijklm}P^s_m+N'_{ijklmn}P^s_mP^s_n \qquad (4)$$

In our previous paper [3] we informed about the study of a quadratic combined piezo-electrooptical effect induced by spontaneous polarization in $CaPb(C_2H_5CO_2)_6$ crystals which undergo the second order ferroelectrical phase transition at T=332K with a change of symmetry 422F4 [4]. It was found that the change of piezooptical constant $n_3^3\pi_{3333}-n_1^3\pi_{1333}$ in the ferroelectrical phase is proportional to the square of spontaneous polarization. But the ferroelectrical phase transition in $CaPb(C_2H_5CO_2)_6$ crystals is diffused [5] and the change of piezooptical constants at phase transition is very small. More over $CaPb(C_2H_5CO_2)_6$ crystals undergo ferroelastical phase transition at 191K [6] and the existance of this phase transition can sufficiently change the temperature behavior of piezooptical coefficients. From these facts follow that for confirmation of relation (4) it is necessary to conduct experimental investigations using the proper ferroelectrical crystals which undergo the second order phase transition and do not undergo any more structural phase transitions. The lead germanate ($Pb_5Ge_3O_{11}$) belongs to such a group of crystals and possesses a ferroelectrical phase transition at 450K with change of point group of symmetry $\bar{6}$F3 [7,8].

The present paper is devoted to the study of a combined piezo-electrooptical effect in $Pb_5Ge_3O_{11}$ crystals induced by spontaneous polarization.

Experimental

In our study the radiation of He-Ne gas laser (λ=632.8nm) was propagated in a perpendicular direction to z axis and mechanical compressive stress was applied along z axis. In this case the equation of the optical indicatrix as for the ferroelectric phase as well as for the paraelectric one can be written as

$$(B_{11}+\pi_{1133}\sigma_{33})x^2+(B_{11}+\pi_{1133}\sigma_{33})y^2 +(B_{33}+\pi_{3333}\sigma_{33})z^2=1 \qquad (5)$$

with the solution

$$\delta(\Delta n)_{13}=(n_3^3\pi_{3333}-n_1^3\pi_{3333})\sigma_{33} \qquad (6)$$

where $\delta(\Delta n)_{13}$ - is the piezooptical change of birefringence.

Investigation of the temperature dependencies of the birefringence $\delta(\Delta n)_{13}$ for the $Pb_5Ge_3O_{11}$ crystals were carried out by Senarmont method. The precision of the determination of the change of birefringence was 10^{-6}. The temperature control in a specially designed optical furnace with the possibility of application of compressive strain was not worse than 0.1K. Applied mechanical strain was not larger than 4×10^6N/m^2.

Results and discussion

The temperature dependencies of birefringence change under the compressive strain are presented on Fig.1. As it is evident from these dependencies the increasing of the strain leads to insufficient increase of the phase transition temperature (insert on Fig.1). From these dependencies on the base of Eq.(6) one can calculate the temperature behavior of the piezooptical constant (Fig.2). This temperature dependence is quite unusual because the piezooptical constant possesses a minimum on 7K below phase transition temperature. But as one can see from Fig.1 due to the shifting of the phase transition temperature birefringence

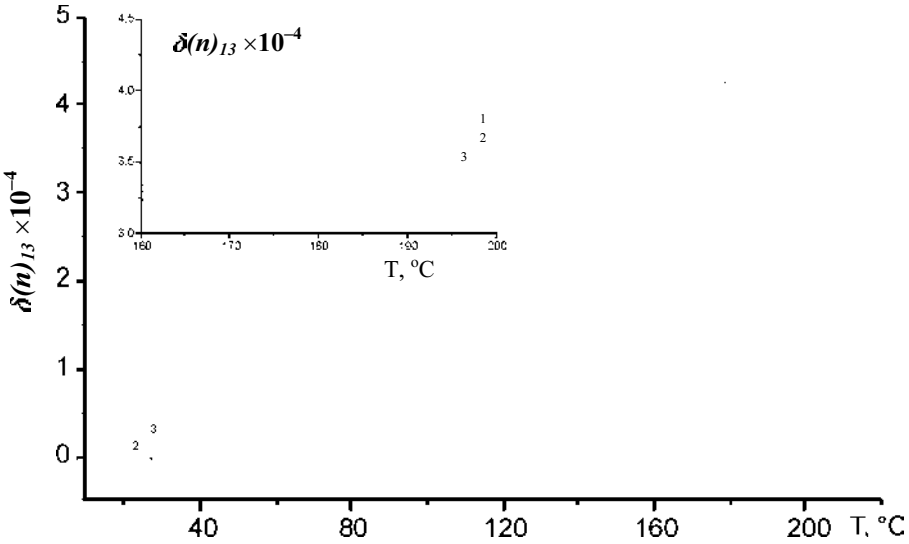

Fig. 1. The temperature dependences of the birefringence change $\delta(\Delta n)_{13}$ for the $Pb_5Ge_3O_{11}$ crystals 1-σ_{33}=0, 2 - σ_{33}=1.86×10^{6}N/m^2, 3-σ_{33}=3.62×10^{6}N/m^2 (λ=632.8nm). Insert: $\delta(n)_{13}$ in the vicinity of T_c.

curves at $\sigma_{33}\neq0$ become more close to the curve of the temperature birefringence change at σ_{33}=0 at $T=T_c$-7K. In order to eliminate the shifting of the phase transition temperature by shifting the curves of temperature dependencies of birefringence at non zero mechanical strain to the lower temperature region on the respective value of ΔT_c then the calculated temperature change of piezooptical constant (Fig.2) doesn't possess such deep minimum.

Let us consider the equation of the optical indicatrix for the paraelectric phase with point group of symmetry $\bar{6}$ on the appearance of spontaneous polarization P^s_3 and under the influence of mechanical strain σ_{33}

$$(B_{11}+\pi_{1133}\sigma_{33}+R_{1133}P^s_3P^s_3+N_{113333}\sigma_{33}P^s_3P^s_3)x^2+$$
$$(B_{11}+\pi_{1133}\sigma_{33}+R_{1133}P^s_3P^s_3+N_{113333}\sigma_{33}P^s_3P^s_3)y^2+ \quad (7)$$
$$(B_{33}+\pi_{3333}\sigma_{33}+R_{3333}P^s_3P^s_3+N_{333333}\sigma_{33}P^s_3P^s_3)z^2=1$$

where R_{ijmn} are the coefficients of the fourth rank polar tensor of quadratic electrooptical effect induced by spontaneous polarization and N_{ijklmn} are coefficients of the six rank polar tensors of combined piezo-electrooptical effect induced by spontaneous polarization and mechanical strain. In this case the temperature change of birefringence and piezooptical constant $n_3^3\pi_{3333}-n_1^3\pi_{1133}$ below T_c should be

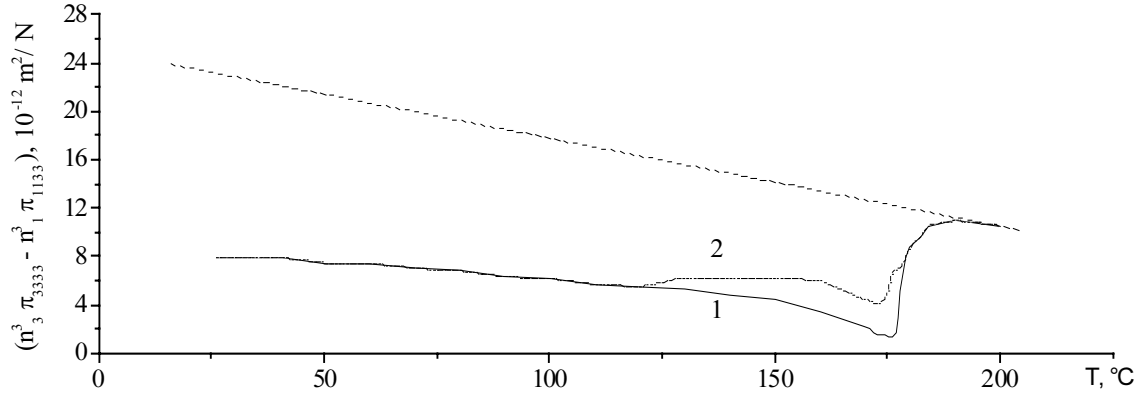

Fig. 2. Temperature dependence of the piezooptical coefficient $n_3^3\pi_{3333}-n_1^3\pi_{1133}$ (λ=632.8nm) (1- experimental results; 2 - experimental results after the elimination of the phase transition temperature shifting; dash line - approximation of the temperature dependence from paraelectric phase to ferroelectric one)

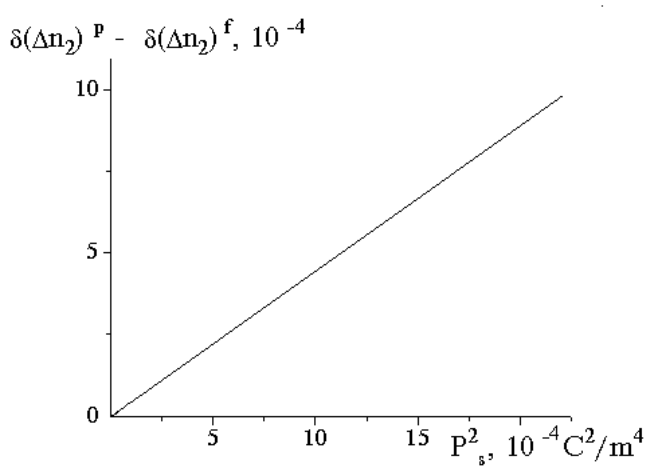

Fig. 3. Dependence of the increment of birefringence on square of spontaneous polarization.

described by relations

$$\delta(\Delta n)_{13}=(n_3^3R_{3333}-n_1^3R_{3333})P^s_3/2, \quad (8)$$

$$n_3^3N_{333333}-n_1^3N_{113333}=(n_3^3\pi_{3333}-n_1^3\pi_{3333})/P^s_3 \quad (9)$$

and both dependencies should be proportional to the square of spontaneous polarization (Fig.3,4) (temperature dependence of P_s is taken from [8]). Calculated values of electrooptical coefficients $n_3^3R_{3333}-n_1^3R_{1133} = 0.9$ m^4/C^2 as well as coefficients of combined piezo-electrooptical effect $n_3^3N_{333333}-n_1^3N_{113333}= 0.73\cdot10^{-8}$ m^2/N·C^2**.**

The value of the coefficient of combined piezo-electrooptical effect was calculated on the the base of the linear part of dependence presented on Fig. 4.

From the obtained results it is possible to construct the surface of the change of the birefringence under the mutual influence of mechanical strain and square of spontaneous polarization (Fig. 5).

Conclusions

1. The temperature dependence of birefringence $\delta(\Delta n)_{13}$ in lead germanate crystals at proper ferroelectrical phase transition is investigated under the influence of mechanical strain σ_{33}. It was shown that the application of compressive strain σ_{33} leads to the increase of phase transition temperature and $\partial T_c/\partial\sigma_{33}$=0.47.10-6 Km^2/N. It was shown that the temperature change of

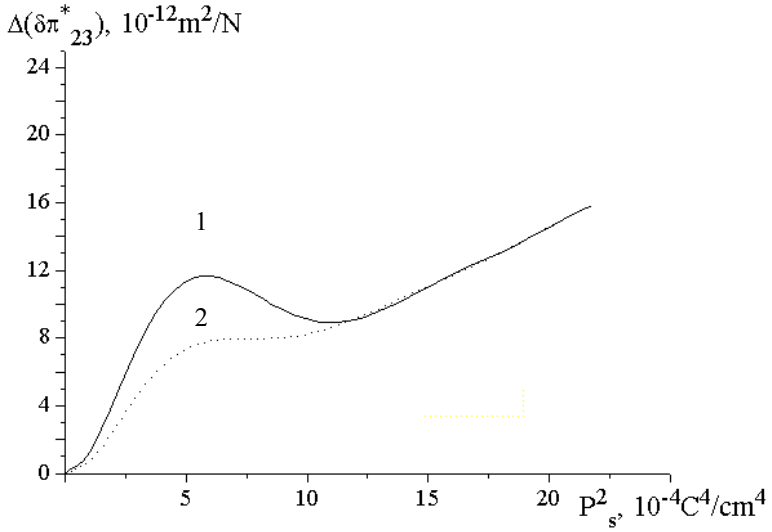

Fig. 4. Dependences of the increment of piezooptical coefficient on the square of spontaneous polarization (1 - plotted on the base of solid curve on Fig.2; 2 - plotted on the base of dash-and-dot curve on Fig.2)

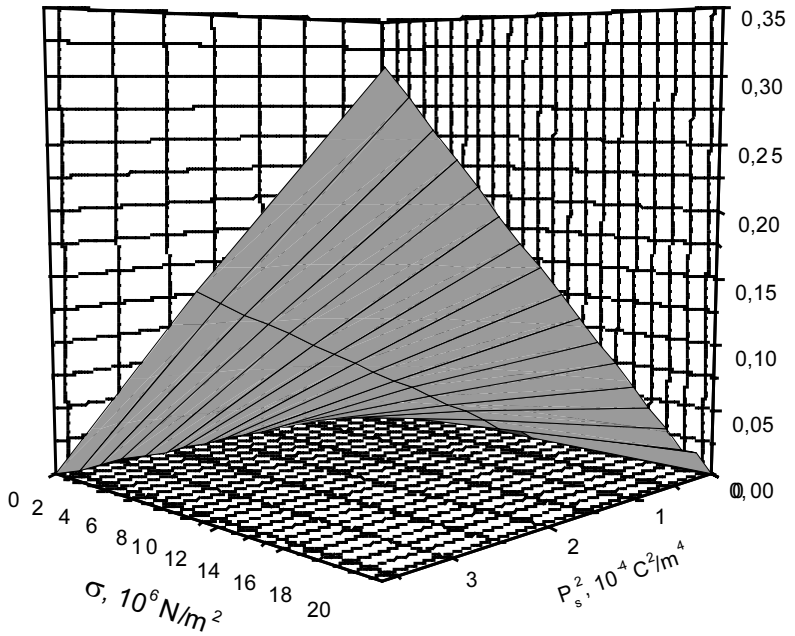

Fig. 5. Surface of the change of birefringence under the mutual influence of mechanical strain and square of spontaneous polarization in $Pb_5Ge_3O_{11}$ crystals (λ=632.8nm).

birefringence is described by quadratic electro-optical effect induced by spontaneous polarization. The calculated value of the Kerr effect coefficients for the paraelectrical phase is $n_3^3R_{3333}$-$n_1^3R_{1133}$ =0.9 m^4/C^2.

2. The temperature dependence of piezooptical constants of lead germanate crystals $n_3^3\pi_{3333} - n_1^3\pi_{1133}$ is experimentally studied. It was shown that these dependence is described by the combined piezo-electrooptical effect induced by spontaneous polarization and external mechanical strain. The temperature change of $n_3^3\pi_{3333}$ -$n_1^3\pi_{1133}$ is proportional to the square of P_s. The calculated value of the coefficients of combined piezo-electrooptical effect is $n_3^3N_{333333}$-$n_1^3N_{113333}$=0.73×10^{-8} m^2/N·C$^{2.}$

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