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Photoelastic properties of the beta barium borate crystals

Anatoliy S. Andrushchak[1], Yaroslav V. Bobitski[1,2], Mykola V. Kaidan[1], Volodymyr T. Adamiv[3], Yaroslav V. Burak[3], Bohdan G. Mytsyk[4]

[1]Lviv Polytechnic National University, 12 S. Bandery Str., 79013, Lviv, Ukraine.

[2]Institute of Technics, Rzeszów University, ul. Rejtana 16A, 35–959 Rzeszów, Poland.

[3]Institute of Physical Optics, 23 Dragomanov Str., 79005, Lviv, Ukraine.

[4]Lviv Centre of Space Research Institute, 5a Naukova Str., 79027, Lviv, Ukraine.

The single crystals of the beta barium borate were grown by the top-seeded solution method. For these crystals all the components of piezooptic effect tensor were determined by a two-fold measurement method. Based on this method some magnitudes of elastic compliance coefficients were fixed. The magnitudes and signs of all elastooptic coefficients were also calculated, acoustic quality parameter was analysed and the possibility of using the beta barium borate crystals as perspective acoustooptic material was discussed.

Keywords: beta barium borate crystals, piezo- and elastooptic effect, two-fold measurement method, acustooptic material.

1. Introduction

The beta barium borate crystals (β-BaB_2O_4 or BBO for short) were found out by the group of Chinese scientists headed by Prof. Chuantian Chen [1]. Now these crystals attract great attention of investigators because of their perfect non-linear optical properties [2]. So, BBO is an optically negative uniaxial crystal, which belongs to 3m point group of symmetry [3] with high transparency in the range from 189 to 3500 nm, great birefringence and small dispersion, providing the phase concordance for harmonic generation in the 200–1500 nm region [1], [4]. High optical homogeneity, wide temperature phase synchronism, chemical inertness and good mechanical properties [3], [5] complete the set of unique characteristics of this material. In spite of wide investigation of dielectric, pyroelectric and piezoelectric [6], elastic, non-linear-optical and thermal [3], electrooptical and electrostrictive [7], and also optical [8] and pyroelectroluminescence [9] properties of BBO crystals, their photoelastic properties still have not been investigated.

The aim of the present work is to determine all components of piezooptic effect tensor of grown BBO crystals by a two-fold measurement method put forward and to calculate all elastooptic coefficients for these crystals.

2. Experimental procedure

2.1. Crystal growth and sample preparation

The single crystals BBO were grown by the top-seeded solution method. The mixture for growth in a composition of 78 mol% $Ba_2B_2O_4$ and 22 mol% Na_2O was prepared by means of solid reaction between Na_2CO_3, $BaCO_3$ and H_3BO_3, which were taken in weight ratio of 1:6.8:4.3. For obtaining optically qualitative crystals the low pulling rate of 0.5–2 mm/24h, slow stable rotation and fluent smooth temperature control in the process of growing were provided. The decrease of temperature was 2 K/24h.

The parallelepiped direct-cut samples were of dimensions of ~5×7×9 mm^3, with their sides normal to the X_1, X_2 X_3 crystallophysical axes. The X/45°-cut samples were prepared from direct-cut samples. But before that, with the aim of resolving the ambiguity in determination of coefficients π_{14} and π_{41} [10], the positive directions (*i.e.*, signs) of crystallophysical axes were determined for these samples. To this end, advantage was taken of longitudinal piezoelectric effect (due to the presence of longitudinal piezoelectric coefficients d_{22} and d_{33} for 3m point group of symmetry) based on which the necessary signs of the two axes X_2 and X_3 were found. This was done in accordance with practical recommendations, which for these crystals are similar to those applied to piezoelectric [11] and piezooptic [12] investigations. According to [11], the direction, coinciding with "–" sign of electric polarization, which appeared on crystal face upon applying a compressive deformation (*i.e.*, negative mechanical stress), was called the positive direction of axis.

2.2. Experimental determination of all components of piezooptic effect tensor

The measurements of piezooptic effect were carried out using the experimental installation [10], the principal scheme of which was based on Mach–Zender interferometer. The experimental installation was modified for simultaneous measurement of the absolute piezooptic coefficients π_{im}, by interferometric method and piezooptic coefficients of induced birefringence, π^*_{km}, by polarized-optic method.

Whenever the crystal hardness allowed the registration of piezoinduced change for optical path of $\delta\Delta_{ikm} = \delta(n_i t_k)$ or optical retardation of $\delta\Delta^o_{km} = \delta(\Delta n_k t_k)$ was carried out by the well-known half-wave stress method or by a modified method of extreme intensities, in which several half-wave lengths are induced (here indexes m, k and i indicate the **m**, **k** and **i** directions for application of normal mechanical stress σ_m, propagation of light wave and its polarization, respectively, $\Delta n_k = n_i - n_j$ denotes the birefringence taken as a difference between refractive indexes of n_i and n_j for two light waves, which propagate in **k** direction). In the opposite case, the value of $\delta\Delta_{ikm}$ was measured by the method of interferometric fringe shift registration and the measurements of value $\delta\Delta_{km}^{o}$ were carried out by well-known Senarmont method.

The main coefficients π_{im} (for i, m = 1, 2, 3) were calculated on the direct-cut sample according to equation [10]

$$\pi_{im} = -\frac{2\delta\Delta_{ikm}}{\sigma_m t_k n_i^3} + \frac{2S_{km}(n_i - 1)}{n_i^3}. \tag{1}$$

The coefficients π_{km}^* (for k, m = 1, 2, 3) were determined on direct-cut sample according to the following equation [10]:

$$\pi_{km}^* = -\frac{2\delta\Delta_{km}^{o}}{\sigma_m t_k} + 2\Delta n_k S_{km} = \pi_{km}^{o} + 2\Delta n_k S_{km} \tag{2}$$

where: t_k – the sample length in **k** direction of light propagation; S_{km} – elastic compliance coefficient, π_{km}^{o} – piezooptic coefficient of induced retardance.

For the calculation of coefficients π_{im} and π_{km}^* the signs of $\delta\Delta_{ikm}$ and $\Delta n_k = n_i - n_j$ were determined on the basis of three criteria, given in Sec. 2.3. Then these coefficients co-ordinated one by one according to the well-known equation

$$\pi_{km}^* = \pi_{im} n_i^3 - \pi_{jm} n_j^3. \tag{3}$$

The non-main coefficients π_{14}, π_{41}, π_{44} were calculated by a two-fold measurement method [13] on X/45°-cut sample according to the equations:

$$\pi_{14} = -2n_1^{-3}\left(\frac{\delta\Delta_{1\bar{4}4}}{t_{\bar{4}}\sigma_4} - \frac{\delta\Delta_{14\bar{4}}}{t_4\sigma_{\bar{4}}}\right), \tag{4}$$

$$\pi_{41} = -n_4^{-3}\left(\frac{\delta\Delta_{4\bar{4}1}}{t_{\bar{4}}\sigma_1} - \frac{\delta\Delta_{\bar{4}41}}{t_4\sigma_1}\right) - n_4^{-3}(n_4 - 1)S_{14}, \tag{5}$$

$$\pi_{44} = -2n_4^{-3}\left(\frac{\delta\Delta_{4\bar{4}4}}{t_{\bar{4}}\sigma_4} - \frac{\delta\Delta_{\bar{4}4\bar{4}}}{t_4\sigma_{\bar{4}}}\right) + n_4^{-3}(n_4 - 1)(S_{22} + S_{33} + 2S_{23} - S_{44})$$
$$- \frac{\pi_{22} + \pi_{23} + \pi_{32} + \pi_{33}}{2}. \tag{6}$$

One can see that for determination of coefficient π_{14} it is necessary to carry out two measurements of $\delta\Delta_{1\bar{4}4}$ and $\delta\Delta_{14\bar{4}}$ for direct (i = 1, $k = \bar{4}$, m = 4) and symmetric

($i = 1$, $k = 4$, $m = \bar{4}$) experimental conditions ($\bar{4}$ corresponds to [01$\bar{1}$] direction and 4 corresponds to [011] direction, $n_4 = n_{\bar{4}} = ((n_o + n_e)/2)^{-1/2}$, n_o and n_e – ordinary and extraordinary refractive indexes, respectively). Similarly for determination of the coefficients π_{41} or π_{44} the two measurements of necessary values of $\delta\Delta_{ikm}$ for two experimental conditions (direct and symmetric) must be carried out on the X/45°-cut sample.

For comparison the coefficients π_{41}, π_{14}, π_{44} were also determined by the method of one-fold measurements according to Eqs. (4)–(6) from [10].

2.3. Sign criteria for unambiguous measurement of piezooptic coefficients

For unambiguous determination of sign and in some cases of the magnitude of piezooptic coefficients and also for correct account of elasticity in piezooptic experiment (according to Eqs. (1) or (2)), as well as for unambiguous mutual correlation between absolute piezooptic coefficients and piezooptic coefficients of induced birefringence (according to Eq. (3)) it is necessary to take into account the following sign criteria:

1. The determination of the sign (or positive direction) of crystallophysical axes without which it is impossible to determine unambiguously the non-main piezooptic coefficients [10]. Necessary practical recommendations related to selection of positive direction of crystallophysical axes for right coordinate system during piezooptic effect investigations for crystals of different symmetry classes are given in [12].
2. The determination of the sign of absolute piezooptic coefficients π_{im} during the interferometric investigations. To this end it is necessary to determine the sign for induced change of optical path $\delta\Delta_{ikm}$ of light beam. We take into account the fact that if applied mechanical stress leads to an increase of the optical path of light beam, then the value $\delta\Delta_{ikm}$ is positive, in the opposite case, the value $\delta\Delta_{ikm}$ is negative. Let us note that the mechanical compression in calculations is assumed to be negative.
3. The determination of the sign of induced change of optical retardation in polarized-optic measurements according to Eq. (2). This criterion was called generalized sign determination rule and was given in [10]. But it is written only for the case of light propagation along the X_1, X_2, X_3 crystallophysical axes. For these directions the sign of $\Delta n_k = n_i - n_j$ was determined by the well-known rule [14] for cyclic replacement of indexes $1\to2\to3$, $3\to1\to2$ and $2\to3\to1$, othergates $\Delta n_1 = n_2 - n_3$ and so on. But the needs of experimental investigations require the refinement of this rule for the case of light propagation in any direction of uni- or biaxial crystals, when the sign of birefringence cannot be obtained by index cyclic replacement rule.

Our proposition is the following. One can see that under the change of Δn_k sign the sign of $\delta\Delta^o_{km}$ will also change and this leads to a change of π^*_{km} sign. That is why for unambiguous sign determination of $\pi^{*\prime}_{km}$ (the stroke points an arbitrary direction of light propagation and stress action) we suggest always to take the "+" sign for $\Delta n'_k$ birefringence in an arbitrary direction of light propagation. Then the last criterion for $\delta\Delta_{km}^{o'}$ sign as usual reduces to: the sign for induced change of retardation $\delta\Delta_{km}^{o'}$ is positive if it leads to an increase of the magnitude $\delta\Delta_{km}^{o'}$ and is negative if it leads to a decrease of $\delta\Delta_{km}^{o'}$.

We suppose that it is reasonable to apply this rule to the propagation of light along crystallophysical axes. Only in cases where it is possible to operate with negative values of birefringence (*e.g.*, during construction of indicative surfaces of piezooptic effect, see [15], [16]), we propose to use the generalized sign rule [10] for determination of $\delta\Delta_{km}^{o}$ (or $\delta\Delta_{km}^{o'}$) sign.

3. Results and discussion

All results of experimental measurements on the direct and X/45°-cut samples for BBO crystals are given in Tabs. 1, 2 and 3. The magnitudes of $S_{11} = 25.63$, $S_{12} = -14.85$, $S_{13} = -9.97$, $S_{33} = -37.21$, $S_{14} = -63.97$, $S_{44} = 331.3$ (all values are in $\mathrm{TPa}^{-1} = 10^{-12}\ \mathrm{m}^2/\mathrm{N}$) and $n_1 = n_2 = n_o = 1.5573$, $n_3 = n_e = 1.5560$ from [3] were used in calculations. The piezooptic effects were investigated on unshort-circuit samples, that is at constant electric induction. The measurements were carried out at room temperature with the help of He-Ne laser ($\lambda = 632.8$ nm).

The values of the coefficients π_{im} and π_{km}^{*} are given in Tabs. 1 and 2 with values of their errors, which were calculated as mean-square value from applied load error δP (relative error of the measurement of applied load P_m was $\delta P/P_m = 1\%$), the error of interference fringe shift δk (registration accuracy was $\delta k = 0.02$, and in Senarmont method the accuracy of polarizer rotation angle was $\delta\varphi° = 0.1°$) and the error of elastic compliance coefficients δS (their measurement error was assumed as the value of the least significant digit of coefficients which are given in [3]). The measurement errors of the crystal length and refractive indexes, as the higher infinitesimal order magnitude, were neglected.

3.1. Measurements on the direct-cut sample

The followings is an analysis of experimental investigations of the piezooptic effect on the direct-cut samples of BBO crystal (see Tab. 1):

The list of all possible geometries of experiment which is presented in Tab. 1 shows:

– pairwise equality in the range of errors for absolute piezooptic coefficients of $\pi_{11} \approx \pi_{22}$, $\pi_{12} \approx \pi_{21}$, $\pi_{13} \approx \pi_{23}$, $\pi_{31} \approx \pi_{32}$, which corresponds to the matrix of these coefficients for 3m point group of symmetry;

– equality of coefficients π_{11}, π_{22}, π_{33}, which were obtained for different geometries of experiment;

– equality of piezooptic coefficients for induced birefringence of $\pi_{12}^{*} \approx \pi_{21}^{*}$, $\pi_{13}^{*} \approx \pi_{23}^{*}$, $\pi_{31}^{*} \approx \pi_{32}^{*}$.

These results show satisfactory accuracy of the experimental measurements. Besides, the coefficients π_{km}^{*} calculated on the basis of absolute coefficients π_{im}, which were

T a b l e 1. Results of piezooptic effect investigation on the direct-cut sample of the BBO crystals.

Interferometric measurement							Calculated π^*_{km} from π_{im}, Br	Polarized-optic measurement			
Exper. conditions			$^{\dagger}\sigma^o_{km}$ 10^3 N/m	Absolute coefficients π_{im}, Br	Contribution π_{im}, %	Contribution S_{km}, %		σ^o_{km} 10^3 N/m	Measured coefficient		
k	m	i							Contribution π^o_{km}, Br	Contribution S_{km}, TPa^{-1}	π^*_{km}, Br
2	3	1	–30	$\pi_{13} = 1.8 \pm 0.24$	38	62	$\pi^*_{23} = \pi_{13} n_o^3 - \pi_{33} n_e^3$	–190	–3.3	–2.3	$\pi^*_{23} = -5.6 \pm 0.07$
2	3	3	–25	$\pi_{33} = 3.9 \pm 0.39$	57	43	$= -6.2$		(59%)	(41%)	
1	3	2	–30	$\pi_{23} = 1.7 \pm 0.22$	37	63	$\pi^*_{13} = \pi_{23} n_o^3 - \pi_{33} n_e^3$	–340	–1.9	–2.3	$\pi^*_{13} = -4.2 \pm 0.04$
1	3	3	–26	$\pi_{33} = 3.5 \pm 0.35$	55	45	$= -5.2$		(45%)	(55%)	
3	2	1	–93	$\pi_{12} = -1.4 \pm 0.08$	–97	197	$\pi^*_{32} = (\pi_{12} - \pi_{22}) n_o^3$	–740	0.86	0	$\pi^*_{32} = 0.86 \pm 0.03$
3	2	2	–109	$\pi_{22} = -1.6 \pm 0.14$	–130	230	$= 0.93$		(100%)	(0%)	
1	2	3	–58	$\pi_{32} = -1.5 \pm 0.15$	–50	150	$\pi^*_{12} = \pi_{22} n_o^3 - \pi_{32} n_e^3$	–1090	0.58	–3.5	$\pi^*_{12} = -2.92 \pm 0.02$
1	2	2	–56	$\pi_{22} = -1.9 \pm 0.16$	–76	176	$= -3.2$		(–20%)	(120%)	
3	1	2	–85	$\pi_{21} = -1.3 \pm 0.07$	–77	177	$\pi^*_{31} = (\pi_{11} - \pi_{21}) n_o^3$	–650	–0.97	0	$\pi^*_{31} = -0.97 \pm 0.03$
3	1	1	–101	$\pi_{11} = -1.5 \pm 0.14$	–110	210	$= -0.93$		(100%)	(0%)	
2	1	3	–63	$\pi_{31} = -1.7 \pm 0.14$	–61	161	$\pi^*_{21} = \pi_{11} n_o^3 - \pi_{31} n_e^3$	–2300	0.28	–3.5	$\pi^*_{21} = -3.22 \pm 0.02$
2	1	1	–56	$\pi_{11} = -1.8 \pm 0.16$	–76	176	$= -2.0$		(–9%)	(109%)	

† – operating mechanical stress $\sigma^o_{km} = \sigma_m^{\lambda/2} t_k$ (where $\sigma_m^{\lambda/2}$ is half-wave mechanical stress).

measured by interferometric method, and obtained on the basis of measured induced change of optical retardation by polarized-optic method, are equal by sign and by magnitude. These results show the reliability of experimental results obtained and at the same time demonstrate the concordance and correctness of solving the problem of piezooptic coefficient sign according to the sign criteria (see Sec. 2.3).

The piezooptic and elastic contributions to induced change of optical path $\delta\Delta_{ikm}$ are comparable by the magnitude and equal by the sign in the case of pressure application along X_1 axis. For other experimental geometries these contributions are opposite by sign, moreover piezooptic contribution is always smaller (2–3 times) than elastic one. So called cases of "imaginary" or "pure" piezooptic effects, when one contribution is much greater than another, are absent in BBO crystals, while they are present in $LiNbO_3$ [16] or $LiTaO_3$ [17] ones.

Unexpectedly high magnitudes of σ^o_{km} operating (or half-wave) mechanical stresses for π^*_{km} induced birefringence coefficients are proportional (for $m = 3$) or much greater than mechanical durability of BBO crystals (experimentally detected value of mechanical durability is $\sigma_{11} \approx \sigma_{22} \approx 8\times10^7$ N/m^2, $\sigma_{33} \approx 4.6\times10^7$ N/m^2). That is why the coefficients π^*_{km} on direct-cut sample were measured by Senarmont method.

3.2. Interferometric measurements on the X/45°-cut sample

Let us analyse the results of experimental investigations of the piezooptic effect on the X/45°-cut sample of BBO crystals, which are given in Tab. 2:

1. The values of piezooptic coefficients measured on the X/45°-cut sample by the one-fold measuring method [10] are given in column No. 3. The values of non-main piezooptic coefficients, which were calculated according to Eqs. (4)–(6) by suggested two-fold measurement method [13], and also possible values of coefficients π_{im}, S_{km} or their combinations, calculated from differences or sums of piezo-induced optical path change, are presented in column No. 4. The combinations of $\delta\Delta_{ikm}$ values, with the help of which the values of π_{im} or S_{km} were calculated, are also presented in the fourth column before symbol "⇒".

2. We obtained a good concordance in four cases between $S_{12} + S_{13} = -24.82$ TPa^{-1} taken from [3] and corresponding sum of these coefficients, which were calculated from our measured values of $\delta\Delta_{ikm}$ for direct and symmetric experimental conditions (see experiments Nos. 1, 4, 5, 6). On the contrary, there is an essential difference in sign and in magnitude between $S_{14} = -63.97$ TPa^{-1} from [3] and determined values of S_{14} from three independent geometries (see experiments Nos. 4, 5, 6). Besides, there is a divergence between the calculated combination of coefficients $S'_{4\bar{4}} = (S_{22} + S_{33} + 2S_{23} - S_{44})/2 = -144.2$ TPa^{-1} from [3] and experi-mentally measured value $S'_{4\bar{4}} = -180$ TPa^{-1} (see experiment No. 2). The last two cases of divergence testify to the necessity of making the correction for the aforementioned elastic compliance coefficients of BBO crystals. This necessity is also approved by such facts:

– good concordance between calculated piezooptic coefficients π_{14}, π_{41}, π_{44} by one-fold measuring method (for direct and symmetric experimental conditions, see the

T a b l e 2. Results of the interferometric investigation of piezooptic effect on the X/45°-cut sample.

Experiment number	Experimental conditions: direct and symmetric (a)			Measured π_{im} (in Br), S_{km} (in TPa^{-1}) coefficients or their combinations			
	k	m	i	By one-fold measurement method [10]		By two-fold measurement method [13]	
1	2			3		4	
1	4	1	$\bar{4}$	$\pi_{41} = -2.0 \pm 0.10$	(10.7‡)	$\pi_{41} = -2.03 \pm 0.07$	(10.6‡)
1a	$\bar{4}$	1	4	$\pi_{41} = -2.0 \pm 0.17$	(10.6‡)	$\delta\Delta'_{\bar{4}41} + \delta\Delta'_{4\bar{4}1} \Rightarrow S_{12} + S_{13} = -24.6$	(–24.82‡)
2	4	$\bar{4}$	1	$\pi_{14} = -2.0 \pm 1.3$	(–12.3‡)	$\pi_{14} = -2.0 \pm 0.8$	
2a	$\bar{4}$	4	1	$\pi_{14} = -2.0 \pm 1.2$	(8.2‡)	$\delta\Delta'_{14\bar{4}} + \delta\Delta'_{1\bar{4}4} \Rightarrow S'_{4\bar{4}} = -180$	(–144.2‡)
3	4	$\bar{4}$	$\bar{4}$	$\pi_{44} = -26 \pm 1.4$	(–15.3‡)	$\pi_{44} = -26.3 \pm 0.9$	(–15.8‡)
3a	$\bar{4}$	4	4	$\pi_{44} = -27 \pm 1.5$	(–16.3‡)	$\delta\Delta'_{\bar{4}4\bar{4}} - \delta\Delta'_{4\bar{4}4} \Rightarrow (\pi_{14}+2\pi_{41})/2 = -2.5$	
4	4	1	1	$\pi_{11} = -2.2$	(–14.6‡)	$\delta\Delta'_{141} + \delta\Delta'_{1\bar{4}1} \Rightarrow S_{12} + S_{13} = -21.6$	(–24.82‡)
4a	$\bar{4}$	1	1	$\pi_{11} = -2.2$	(10.2‡)	$\delta\Delta'_{141} - \delta\Delta'_{1\bar{4}1} \Rightarrow S_{14} = 22.6$	(–63.97‡)
5	1	$\bar{4}$	2	$\pi_{14} = -2.4$	(22.5‡)	$\delta\Delta'_{214} - \delta\Delta'_{21\bar{4}} \Rightarrow S_{14} = 22.6$	(–63.97‡)
5a	1	4	2	$\pi_{14} = -1.6$	(23.4‡)	$\delta\Delta'_{214} + \delta\Delta'_{21\bar{4}} \Rightarrow S_{12} + S_{13} = -24.0$	(–24.82‡)
6	1	$\bar{4}$	3	$\pi_{32} + \pi_{33} = 1.1$	(25.5‡)	$\delta\Delta'_{314} + \delta\Delta'_{31\bar{4}} \Rightarrow S_{12} + S_{13} = -23.3$	(–24.82‡)
6a	1	4	3	$\pi_{32} + \pi_{33} = 1.3$	(–23.4‡)	$\delta\Delta'_{314} - \delta\Delta'_{31\bar{4}} \Rightarrow S_{14} = 21.2$	(–63.97‡)

‡ – coefficients calculated with the use of the elastic compliance coefficients from [3].

third column) and by two-fold measurement method (see the fourth column) using S_{14} and $S'_{4\bar{4}}$ coefficients determined by us;

– great difference between π_{14} values obtained from two (direct and symmetric) conditions using literature data of $S'_{4\bar{4}}$ (see experiments Nos. 2, 2a, marked as (*));

– and also facts analogous to the previous two cases that refer to coefficients π_{11} (experiment No. 4), π_{14} (No. 5) and $\pi_{32} + \pi_{33}$ (No. 6), using for calculations the coefficients S_{14} and $S'_{4\bar{4}}$ which were measured by us or taken from literature (compare corresponding piezooptic coefficients in Tabs. 1 and 2).

The divergence between coefficients S_{14} and $S'_{4\bar{4}}$, determined from interferometric investigations and from dynamic measurements in [3], cannot be caused by the quality of grown BBO crystals (even owing to the concordance between other elastic coefficients which were measured by us and corresponding values in [3] – see experiments Nos. 1, 5, 6). This may be stipulated by physical inequality of elastic compliance coefficients, which were determined by means of static (in our investigations) or dynamic (in [3]) methods, or due to inexperience by taking into consideration in calculations by the dynamic method the ambiguity of the choice of right coordinate system. The last finally leads to another sign of elastic stiffness coefficient C_{14} and then of coefficient C_{13}, which calculates in dynamical measuring method [3], [18] through known value of C_{14}. This can essentially affect the values of recounted coefficients of elastic compliance S_{km}. That is why a more detailed and direct measurement of coefficients S_{km} for BBO crystals is necessary.

3. From the comparison of one-fold and two-fold measurement methods it is obvious that the last method has advantage because of its higher accuracy of determination of coefficients π_{14}, π_{41} and π_{44} and possibility of simultaneous determination of some elastic compliance coefficients or their combinations (see Tab. 2).

3.3. Polarized-optic measurements on X/45°-cut sample

We analyse the results of polarized-optic measurements on the *X*/45°-cut sample of BBO crystal, carried out by half-wave stress method (see Tab. 3).

1. Let us note that in the sign determination of birefringence Δn_k and following sign measurement for induced change of optical retardance according to the approach suggested (see Sec. 2.3), the values of non-main coefficients of induced birefringence in all experimental geometries presented have good concordance with corresponding coefficients $\pi^{*\prime}_{km}$, calculated from interferometric investigations.

2. The reaching of half-wave mechanical stress upon exposure not along the main crystal directions, in contrast to impossibility of its reaching on direct cut samples (see above) points to high value of piezooptic effect anisotropy for birefringence in BBO crystals (this is obvious from the comparison of σ^{o}_{km} values in Tabs. 1 and 3, *e.g.*, 2.3×10^6 and 12.2×10^3 N/m, which differ by almost 200 times).

3. One emphasizes the fact of the presence of so called "pure" piezooptic effect for geometry $k = 4$, $m = 1$ (see experiment No. 1 in Tab. 3), where the value of piezooptical contribution is 30 times greater than the elastic contribution. This is another case of unique physical properties of BBO crystals, when simultaneously with "pure"

T a b l e 3. Results of polarized-optic investigation of piezooptic effect on the $X/45°$-cut sample.

No.	Experimental condition k	m	σ^o_{km}, 10^3 N/m	Sign $\Delta n'_k$	Sign $\delta\Delta^{o'}_{km}$	Working equation	Measured coefficients: Contribution π^o_{km}, TPa^{-1}	Contribution S_{km}, TPa^{-1}	$\pi^{*\prime}_{km}$, TPa^{-1}	$\pi^{*\prime}_{km}$ calculated from π'_{im} (see Tab. 2), TPa^{-1}
1	4	1	−58	$n_1 - n_{\bar{4}} > 0$	"−"	$\pi^{*\prime}_{41} = -\frac{-\lambda}{\sigma^o_{41}} + 2\Delta n_4 S'_{41}$	−10.9 (97%)	−0.15 (3%)	−11.1	$\pi^{*\prime}_{41} = \pi'_{11}n_1^3 - \pi'_{\bar{4}1}n_{\bar{4}}^3 = -12.1$
2	$\bar{4}$	1	−89	$n_1 - n_4 > 0$	"+"	$\pi^{*\prime}_{\bar{4}1} = -\frac{+\lambda}{\sigma^o_{\bar{4}1}} + 2\Delta n_{\bar{4}} S'_{\bar{4}1}$	8.5 (153%)	−2.9 (−53%)	5.6	$\pi^{*\prime}_{\bar{4}1} = \pi'_{11}n_1^3 - \pi'_{41}n_4^3 = 4.3$
3	$\bar{4}$	4	−12.2	$n_1 - n_4 > 0$	"+"	$\pi^{*\prime}_{\bar{4}4} = -\frac{+\lambda}{\sigma^o_{\bar{4}4}} + 2\Delta n_{\bar{4}} S'_{\bar{4}4}$	51 (128%)	−11 (−28%)	40	$\pi^{*\prime}_{\bar{4}4} = \pi'_{14}n_1^3 - \pi'_{44}n_4^3 = 43$

piezooptic effect the so-called "imaginary" piezooptic effect exists (see Tab. 1 – last experiment).

4. Calculation of all components of the elastooptic effect tensor

In our investigation, the piezooptic coefficients were measured on unshortened samples, otherwise all coefficients in Tabs. 1–3 are the coefficients measured at constant induction π_{im}^{D}. The elastooptic coefficients p_{in} were calculated according to the known equation

$$p_{in}^{E} = \pi_{im}^{E} C_{mn} = \pi_{im}^{E} S_{mn}^{-1} \tag{7}$$

where: C_{mn} – the matrix of elastic stiffness coefficients, which is inverse to the matrix of elastic compliance coefficients S_{mn}. For this it is necessary to know the real value of piezooptic coefficients, otherwise the coefficients π_{im}^{E} at constant electric field. The piezoelectric addition to elastic compliance coefficients for BBO crystals is less than 1%, that is why it can be neglected and one can consider that $S_{mn}^{D} = S_{mn}^{E}$.

The real values of coefficients π_{im}^{E} were calculated by known formula [14], [19] for non-centrosymmetric uniaxial crystals:

$$\pi_{im}^{E} \equiv \pi_{\lambda\mu\eta\nu}^{E} = \pi_{\lambda\mu\eta\nu}^{D} - \frac{r_{\lambda\mu\tau} d_{\tau\eta\nu}}{\varepsilon_o(\varepsilon_{\tau\tau} - 1)} = \pi_{\lambda\mu\eta\nu}^{D} - A_{\lambda\mu\eta\nu}. \tag{8}$$

In Eq. (8), the second term $A_{\lambda\mu\eta\nu}$ is the secondary electrooptical addition, in which $r_{\lambda\mu\tau}$ and $d_{\tau\eta\nu}$ are the coefficients of linear electrooptical effect and piezoelectric modules, respectively, $\varepsilon_o = 8.85\times10^{-12}$ F/m – electric constant, $\varepsilon_{\tau\tau}$ – permittivity of the crystal. In calculations the values of $r_{113} = 0.27$, $r_{222} = -2.41$, $r_{333} = 0.29$, $r_{131} = 1.7$ and $d_{311} = -1.17$, $d_{222} = 2.30$, $d_{333} = 3.4$, $d_{113} = -9.6$ (all values are in 10^{-12} m/V) from [7], and the permittivity values of $\varepsilon_{11} = \varepsilon_{22} = 6.7$ and $\varepsilon_{33} = 8.1$ from [3] were used.

Table 4. Average value of piezooptic coefficients π_{im}^{D} and π_{im}^{E} and calculated value of elastooptic coefficients p_{in}^{E}

Index *im* or *in*	π_{im}^{D} Br	π_{im}^{E} Br	p_{in}^{E}
11	-1.7	-1.6	-0.195
12	-1.35	-1.46	-0.197
13	1.75	1.73	-0.059
31	-1.6	-1.6	-0.112
33	3.7	3.7	0.039
14	-2.0	-1.54	-0.005
41	-2.03	-2.02	-0.007
44	-26.3	-26.3	-0.078

The values of elastooptic coefficients p_{im}^E and their signs, which were calculated for π_{im}^E values given in Tab. 4, are presented in the same table. The S_{mn} values from [3] were used in calculations, except coefficient S_{14}, which was taken from our experimental measurements and was equal to $S_{14} = 22.6$ TPa^{-1}. As can be seen from Tab. 4, the BBO crystals have great values of elastooptic coefficients, especially p_{11} and p_{12}. These crystals have the advantage over such acoustooptic materials as $LiNbO_3$ and $LiTaO_3$ and their coefficients are comparable with corresponding coefficients of fused quartz [20]. This and relatively small values of density ($\rho = 3.84\times10^3$ kg/m^3, see [3]) and acoustic wave velocities (*e.g.*, the measured velocity for wave with directions of [001] propagation and [100] polarization is about $v \approx 1000$ m/s) provide the high value of acoustooptic quality M_2 of this material, according to the well-known equation $M_2 = p_{\mathrm{eff}}^2 n_i^6 / \rho v^3$, which is the main criterion for practical choice of photoelastic material. Besides, the small light absorption in a wide wavelength range, the possibility of obtaining large samples of high optical quality, chemical inertness, mechanical stiffness, and temperature parameters stability make possible the use of the BBO crystals as perspective acoustooptic material.

5. Conclusions

This work can be sumarized as follows:

1. For the β-BaB_2O_4 crystals, which were grown by the top-seeded solution method, all absolute piezooptic coefficients by interferometric method and coefficients of induced birefringence by polarized-optic method were determined, and their mutual correlation was carried out. The advantages of two-fold measurement method, which allows us to determine with greater accuracy all the components of piezooptic effect tensor for various symmetry crystals [13], including triclinic symmetry, were experimentally analysed. This method has also given the possibility of adjusting some values of elastic compliance coefficients for BBO crystals.

2. The three sign criteria, which are necessary for measurements of all piezooptic coefficients, were generalized. The application of the sign criterion for induced change determination of optical retardation in the case of light propagation along any anisotropic direction of uniaxial or biaxial crystals was enlarged and its experimental confirmation on the X/45°-cut samples of BBO crystals was carried out.

3. Taking into account the secondary electrooptical addition in piezooptic coefficients at constant electrical induction, the values of all elastooptic coefficients for BBO crystals were calculated. Such a method for determination of elastooptic coefficients through piezooptic [21], [22] ones has advantage because it gives a possibility to calculate the sign and magnitude of each elastooptic coefficient in contrast to acoustooptic measurements, in which the sign of the same coefficients cannot be determined [19], [20]. On basis of these calculations the acoustooptic quality parameter was analysed and the conclusion about the possibility of using the β-BaB_2O_4 crystals as perspective acoustooptic material was made.

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