Acoustooptic interaction and photoelastic properties of $Li_2B_4O_7$ and α -BaB₂O₄ crystals at the wavelength of 442 nm

Martynyuk-Lototska I., Dudok T., Mys O., Romanyuk G. and Vlokh R.

Institute of Physical Optics, 23 Dragomanov St., 79005 Lviv, Ukraine

Received: 13.11.2009

Abstract

Acoustooptic and photoelastic properties of $\rm Li_2B_4O_7$ and $\alpha\text{-}BaB_2O_4$ crystals have been studied for the light wavelength of 442 nm . It has been shown that the efficiency of acoustooptic diffraction increases essentially at 442 nm, when compare with that typical for 632.8 nm.

Keywords: acoustooptic diffraction, borate crystals, dispersion

PACS: 78.20.Hp, 78.20.Ci **UDC**: 535.42

Introduction

Crystals of borate family are well known as efficient nonlinear optical materials. They are highly resistant to powerful laser radiation and transparent in deep ultraviolet spectral range. Recently we have shown that these materials could be efficiently used for acoustooptic (AO) operation of optical radiation [1-7]. In particular, the AO figure of merit (AOFM) for α -BaB₂O₄ crystals reaches the value $M_2 = (270 \pm 70) \times 10^{-15} \text{ s}^3/\text{kg}$ for the wavelength 632.8 nm of optical radiation, provided that the geometry of interaction is optimised [8]. As already mentioned, one of the advantages of borate crystals is their wide range of transparency. For example, SrB₄O₇ crystals are transparent in the deep ultraviolet region down to 130 nm [9, 10]. However, AO properties of the borates have so far been studied only at the wavelength of He-Ne laser (632.8 nm). It is impossible even to estimate the AO efficiency of those crystals for the shorter wavelengths because of a lack of data on both the photoelastic coefficients and the refractive indices. That is why the present work is devoted to studies of dispersion of the refractive indices, piezooptic and photoelastic coefficients, and AO diffraction at the operational wavelength of 442 nm. These studies are performed for the cases of Li₂B₄O₇ and α -BaB₂O₄ crystals.

Experimental

Dispersion of the refractive indices for Li₂B₄O₇ crystals in the visible spectral range had been earlier studied in our work [11]. Here we studied the refractive indices of α -BaB₂O₄ in the visible range, using index-matching liquids. The accuracy for the refractive indices was not worse than $\pm 10^{-3}$. Dispersion of the refractive indices was obtained with the aid of Sellmeier equation

$$n^{2} = A + B_{1} \frac{\lambda^{2}}{\lambda^{2} - \lambda_{0}^{2}} + B_{2} \frac{\lambda^{2}}{\lambda^{2} - \lambda_{1}^{2}},$$
(1)

where λ_0 and λ_1 are the characteristic wavelengths of oscillators, which correspond respectively to the short-wave and long-wave absorption, and *A*, *B*₁ and *B*₂ denote the fitting parameters. Then the λ_0 and λ_1 values were critically compared with the spectral absorption data [12, 13]).

The piezooptic coefficients were studied at the wavelength of He-Cd gas laser $(\lambda = 442 \text{ nm})$ with interferometric technique, using a Mach-Zender interferometer. The technique has been described in detail in the open literature (see, e.g., [8]). The photoelastic coefficients were calculated with the formula $p_{\lambda\mu} = \pi_{\lambda\nu}C_{\nu\mu}$. AO diffraction was studied at the RF signal frequency of 100MHz with the aid of AO cells (see [8]). The incident optical radiation of He-Cd laser propagated through a crystal, in which a piezoelectric transducer excited a longitudinal acoustic wave. The intensity of incident light I_i and that of zero-order diffraction maximum I_0 were measured with photomultiplier or semiconductor photodetector. The diffraction efficiency η was calculated according to the relation $\eta = (I_i - I_0)/I_i$, where I_0 was measured as a function of driving electric power applied to the transducer. Notice that the intensity of the diffracted beam is equal to $I_d = I_i - I_0$. We determined the AO efficiency on the basis of the above relation, since the intensities of the zero-order and incident beams were comparable, thus allowing us to perform measurements in the same linear range of photodetector response. The error for the AO diffraction efficiency was about 10%.

Results and discussion

The calculation results for the dispersion of refractive indices in Li₂B₄O₇ and α -BaB₂O₄ crystals are presented in Fig. 1. The corresponding fitting parameters are as follows: $\lambda_0 = 165 \text{ nm}$ and $\lambda_1 = 3550 \text{ nm}$; $A_e = 2.02922$, $B_{1e} = 0.36125$ and $B_{2e} = 0.04940$ for n_e ; $A_o = 2.26602$, $B_{1o} = 0.31050$ and $B_{2o} = 0.45813$ for n_o (Li₂B₄O₇ crystals), $\lambda_0 = 183 \text{ nm}$ and $\lambda_1 = 3500 \text{ nm}$; $A_e = 2.16808$, $B_{1e} = 0.17764$ and $B_{2e} = -0.02069$ for n_e ; $A_o = 2.53281$, $B_{1o} = 0.23664$ and $B_{2o} = 0.34332$ for n_o (α -BaB₂O₄ crystals).

The piezooptic and photoelastic coefficients of α -BaB₂O₄ and LiB₄O₇ measured at the wavelengths of 632.8 nm and 442 nm (see Table 1) do not differ much if one takes into account rather high error of their determination (at least ~ 30%).

From the relation $M_2 = n^6 p_{ef}^2 / \rho v^3$ (with *n* being the refractive index, p_{ef} the effective photoelastic coefficient, ρ the density of material and *v* the acoustic wave velocity) it follows that the dispersion of AOFM M_2 is determined by dispersions of both the refractive indices and the photoelastic coefficients. However, one can see from Table 1 that the photoelastic coefficients are determined with rather high error and, within this error, their dispersion cannot be clearly detected.



Fig. 1. Dispersion of refractive indices for $Li_2B_4O_7$ (a) and α -BaB₂O₄ (b) crystals: triangles correspond to n_o and circles to n_e . Full and open triangles and circles correspond respectively to the experimental data [10] and the results of approximation with the Sellmeier equation.

The experimental results for AO diffraction efficiency for the case of interaction with the longitudinal acoustic wave in α -BaB₂O₄ and Li₂B₄O₇ crystals are presented in Fig. 2. Notice that the longitudinal acoustic wave in the both AO cells propagates along *z* axis and the light beam is almost parallel to *y* axis. In all the cases mentioned above the isotropic diffraction is observed. The AO efficiencies for α -BaB₂O₄ crystals measured for the light polarizations parallel to *x* and *z* axes are almost the same, while the AO efficiency for Li₂B₄O₇ is almost four times greater in case of the polarization parallel to *z* axis than that for the *x* axis. The AO diffraction efficiency is as high as $\eta \approx 17\%$ (note that $\eta \approx 10\%$ for $\lambda = 632.8$ nm [8]) in the α -BaB₂O₄ crystals at *P* = 0.5 W, while for the





Fig. 2. Dependences of diffraction efficiency on the power of electrical signal for α -BaB₂O₄ (a) and Li₂B₄O₇ crystals ($\lambda = 442 \text{ nm}$), and AO diffraction pattern observed in α -BaB₂O₄ at $\lambda = 442 \text{ nm}$ and P = 0.5 W (c). Full and open circles correspond to the light polarizations parallel to *x* and *z* axes, respectively.

Ukr. J. Phys. Opt. 2009, V10, №4

	riezooplic coefficient at 442 nm, 10^{-12} m ² /N	Plezooptic coemcient at 632.8 nm, 10 ⁻¹² m ² /N [R]	Matrix component of photoelastic coefficients	Photoelastic coefficient at 442 nm	Photoelastic coefficient at 632.8 nm [8]
α-BaB₂O₄ crystals					
$\pi_{11} = \pi_{22}$	1.8 ± 0.6	1.6 ± 0.5	$p_{11} = p_{22}$	0.17 ± 0.10	0.13 ± 0.09
$\pi_{12} = \pi_{21}$	1.4 ± 0.4	2.1±0.6	$p_{12} = p_{21}$	•0.09	0.2 ± 0.1
$\pi_{13} = \pi_{23}$	-3.1±1.0	-4.3±1.3	$p_{13} = p_{23}$	0.07*	0.13*
$\pi_{31} = \pi_{32}$	1.0±0.3	1.5 ± 0.5	$p_{31} = p_{32}$	-0.1*	-0.15*
π_{33}	-4.3±1.3	-7.2±2.1	P_{33}	-0.06*	-0.11*
$\boldsymbol{\pi}_{66} = (\boldsymbol{\pi}_{11} - \boldsymbol{\pi}_{12})$	0.4 ± 0.1	1	$p_{66} = (p_{11} - p_{12})/2$	0.07*	0.06*
$\pi_{44} = \pi_{55}$	20.0±6.0	24.6±9.4	$p_{44} = p_{55}$	1	-0.17 ± 0.06
$\pi_{65} = \pi_{14} = -\pi_{24}$	-11.0 ± 3.0	-14.2 ± 3.6	11	-0.06	-0.08 ± 0.02
$\pi_{56} = 2\pi_{41} = -2\pi_{42}$	4.0±1.3	5.6*	$p_{56} = 2p_{41} = -2p_{42}$	0.6*	0.19*
Li ₂ B₄O ₇ crystals					
$\boldsymbol{\pi}_{11} = \boldsymbol{\pi}_{22}$	-1.5*	-2.8±0.8	$p_{11} = p_{22}$	I	-0.3 ± 0.1
$\pi_{12} = \pi_{21}$	-1.7*	-0.5 ± 0.2	$p_{12} = p_{21}$	I	-0.03*
$\pi_{13} = \pi_{23}$	3.8 ± 0.8	1.1 ± 0.3	$p_{13} = p_{23}$	1	-0.11 ± 0.05
$\pi_{31} = \pi_{32}$	-1.1*	-2.5±0.8	$p_{31} = p_{32}$	I	-0.2 ± 0.1
π_{33}	2.8 ± 0.5	2.9±0.8	P_{33}	1	-0.07 ± 0.03
π_{66}	2.1*	1.1*	P_{66}	0.1*	0.05 ± 0.02
$\pi_{_{44}}=\pi_{_{55}}$	1.3*	•	$p_{44} = p_{55}$	0.07*	0.16*

Table 1. Piezooptic and photoelastic coefficients of $\alpha\text{-}BaB_2O_4$ and LiB_4O_7 crystals

Ukr. J. Phys. Opt. 2009, V10, №4

Li₂B₄O₇ crystals we have $\eta \approx 26\%$ at the same power of electric signal, if the polarization is parallel to the *x* axis ($\eta \approx 7.5\%$ for $\lambda = 632.8$ nm [8]). As one can see, the efficiency of acoustooptic diffraction for the both crystals increases several times at the wavelength of 442 nm, when compare with the same parameter at 632.8 nm.

The experimental results concerned with the AO diffraction in α -BaB₂O₄ and Li₂B₄O₇ correspond to the interaction of longitudinal acoustic wave ($v_{33} = 3239 \pm 51$ m/s for α -BaB₂O₄ and $v_{33} = 5040 \pm 100$ m/s for Li₂B₄O₇) propagating along *z* axis with the incident optical wave, of which propagation direction is slightly inclined to *y* axis. The Bragg angles for the α -BaB₂O₄ and Li₂B₄O₇ crystals are equal respectively to $\theta_B = 0.71^{\circ}$ (cf. with the value $\theta_B = 0.53^{\circ}$ for $\lambda = 632.8$ nm [8]) and $\theta_B = 0.46^{\circ}$ (cf. with the value $\theta_B = 0.37^{\circ}$ for $\lambda = 632.8$ nm [8]).

The relations that describe this kind of isotropic diffraction in our crystals may be written as

$$E_3^{\omega \pm \Omega} = p_{33} e_3^{\Omega} D_3^{\omega}$$

$$E_1^{\omega \pm \Omega} = p_{13} e_3^{\Omega} D_1^{\omega},$$
(2)

where E_i is the electric field of the diffracted wave (the frequency $\omega \pm \Omega$), D_j the electric displacement of the incident wave (the frequency ω), and e_3^{Ω} the mechanical strain caused by the acoustic wave (the frequency Ω). Notice that the value of acoustic wave frequency agrees well with that following from the Bragg diffraction conditions. According to [14], we present the relation for the diffraction efficiency as

$$\eta = \frac{I}{I_0} = \sin^2 \left[\frac{\pi nL}{2\lambda_0 \cos \theta_B} \frac{\Delta \varepsilon_i}{\varepsilon_i^0} \right] = \sin^2 \left[\frac{\pi}{\lambda_0 \cos \theta_B} \sqrt{\frac{P_a L M_2}{2H}} \right] \approx \frac{\pi^2 P_a L M_2}{2\lambda_0^2 H \cos^2 \theta_B}, \quad (3)$$

where $\Delta \varepsilon_i = \varepsilon_i^p - \varepsilon_i^0$ means the difference between the perturbed $(\varepsilon_i^p = \frac{1}{B_i^p})$ and nonperturbed $(\varepsilon_i^0 = \frac{1}{B_i^0})$ dielectric permittivities, B_i the optical-frequency dielectric impermeability, L the length of interaction of acoustic and optical waves, θ_B the Bragg angle, λ_0 the wavelength of optical radiation, n the refractive index and H the width of piezoelectric transducer.

Let us write out equation for the optical indicatrix in the form

$$(B_1 + p_{13}e_3)x^2 + (B_1 + p_{13}e_3)y^2 + (B_3 + p_{33}e_3)z^2 = 1.$$
 (4)

Then we are able to derive the ratio of photoelastic coefficients as a function of AO efficiency determined for the light waves polarized along the $x(\eta_1)$ and $z(\eta_3)$ axes:

$$\frac{p_{13}}{p_{33}} = \frac{n_o^3}{n_e^3} \sqrt{\frac{\eta_1}{\eta_3}} \,. \tag{5}$$

From this relation and the experimental data available one finds $p_{13}/p_{33} = 1.3$ for α -BaB₂O₄ ($p_{13}/p_{33} = 1.2$ for $\lambda = 632.8$ nm [8]) and $p_{13}/p_{33} = 1.75$ for Li₂B₄O₇ crystals ($p_{13}/p_{33} = 1.66$ for $\lambda = 632.8$ nm [8]). It is evident the given ratios are almost the same for the both wavelengths. In other words, dispersion of the AOFM is determined mainly by dispersion of the refractive indices. Namely, we get the estimation

$$\left(\frac{M_2^{442\,\mathrm{nm}}}{M_2^{632.8\,\mathrm{nm}}}\right)_z = \left(\frac{n_e^{442\,\mathrm{nm}}}{n_e^{632.8\,\mathrm{nm}}}\right)^6 \simeq \left(\frac{M_2^{442\,\mathrm{nm}}}{M_2^{632.8\,\mathrm{nm}}}\right)_x = \left(\frac{n_o^{442\,\mathrm{nm}}}{n_o^{632.8\,\mathrm{nm}}}\right)^6 = 1.05 \quad \text{for} \quad \alpha-\mathrm{BaB}_2\mathrm{O}_4 \quad \text{and}$$

$$\left(\frac{M_2^{442\,\mathrm{nm}}}{M_2^{632.8\,\mathrm{nm}}}\right)_z = \left(\frac{n_e^{442\,\mathrm{nm}}}{n_e^{632.8\,\mathrm{nm}}}\right)^6 = 1.2, \\ \left(\frac{M_2^{442\,\mathrm{nm}}}{M_2^{632.8\,\mathrm{nm}}}\right)_x = \left(\frac{n_o^{442\,\mathrm{nm}}}{n_o^{632.8\,\mathrm{nm}}}\right)^6 = 1.3 \text{ for } \mathrm{Li}_2\mathrm{B}_4\mathrm{O}_7.$$

As a result, the AOFMs for the both crystals do not differ essentially for the wavelengths of 632.8 and 442 nm. However, it follows from Eq. (3) that the ratio of diffraction efficiencies also depends on the ratio of the corresponding wavelengths, under the conditions of equal other parameters:

$$\frac{\eta_{442\text{nm}}}{\eta_{632.8\text{nm}}} = \frac{M_2^{442\text{nm}}(632.8)^2}{M_2^{632.8\text{nm}}(442)^2} = 2.05 \frac{M_2^{442\text{nm}}}{M_2^{632.8\text{nm}}}.$$
(6)

Actually we have
$$\frac{\eta_{442\text{nm}}}{\eta_{632.8\text{nm}}} = 2.05 \frac{M_2^{442\text{nm}}}{M_2^{632.8\text{nm}}} = 2.15 \text{ for } \alpha\text{-BaB}_2\text{O}_4 \text{ (or } \frac{\eta_{442\text{nm}}}{\eta_{632.8\text{nm}}} = 1.7 \text{,}$$

according to the experimental data),
$$\left(\frac{\eta_{442\text{nm}}}{\eta_{632.8\text{nm}}}\right)_x = 2.05 \left(\frac{M_2^{442\text{nm}}}{M_2^{632.8\text{nm}}}\right)_x = 2.7$$
 (or

$$\left(\frac{\eta_{442\text{nm}}}{\eta_{632.8\text{nm}}}\right)_x = 3.5$$
, according to the experimental data) and

 $\left(\frac{\eta_{442\text{nm}}}{\eta_{632.8\text{nm}}}\right)_z = 2.05 \left(\frac{M_2^{442\text{nm}}}{M_2^{632.8\text{nm}}}\right)_z = 2.5. \text{ Thus, we obtain a quite satisfactory agreement}$

between the theoretical estimations and the experimental data for the diffraction efficiency ratio.

Conclusions

On the basis of results presented above one can conclude that the efficiency of AO diffraction in the both α -BaB₂O₄ and Li₂B₄O₇ crystals increases essentially when passing from the 'red' spectral region (632.8 nm) to the 'violet' one (442 nm). The experimental data derived for the AO diffraction agree well with the diffraction efficiency values estimated theoretically. Moreover, we have obtained some information regarding spectral dependences of the refractive indices and the photoelastic coefficients. It has been shown that the increase in the diffraction efficiency occurring for the short wavelength is mainly

caused by dispersion of the refractive indices and a quadratic dependence of the diffraction efficiency on the wavelength of optical radiation.

References

- 1. Komatsu R, Sugawara T, Sassa K, Sarukura N, Liu Z, Izumida S, Segawa Y, Uda S, Fukuda T and Yamanouchi K, 1997. Growth and ultraviolet application of $Li_2B_4O_7$ crystals: Generation of the fourth and fifth harmonics of Nd:Y₃Al₅O₁₂ lasers. Appl. Phys. Lett. **70**: 3492–3494.
- 2. Kato K, 1986. Second-harmonic generation to 2048 Å in BaBa₂O₄. IEEE J. Quant. Electronics. **22:** 1013–1014.
- 3. Ishida Y and Yajima T, 1987. Characteristics of a new-type SHG crystal β -BaB₂O₄ in the femtosecond region. Opt. Comm. **62:** 197–200.
- 4. Cheng LK, Bosenberg W and Tang CL, 1988. Broadly tunable optical parametric oscillation in β -BaB₂O₄. Appl. Phys. Lett. **53:** 175–177.
- Chen CT, Wu BC, Jiang AD and You GM, 1985. A new ultraviolet SHG crystal β-BaB₂O₄. Sci. Sin. B. 28: 235–243.
- 6. Yoshida H, Fujita H, Nakatsuka M, Yoshimura M, Sasaki T, Kamimura T and Yoshida K, 2006. Dependences of laser-induced bulk damage threshold and crackp in several nonlinear crystals on irradiation direction. Japan. J. Appl. Phys. **45:** 766–769.
- Mori Y, Yap Y K, Kamimura T, Yoshimura M and Sasaki T, 2002. Recent development of nonlinear optical borate crystals for UV generation. Opt. Mater. 19: 1–5.
- 8. Martynyuk-Lototska I, Mys O, Dudok T, Adamiv V, Smirnov Y and Vlokh R, 2008. Acousto-optic interaction in α -BaB₂O₄ and Li₂B₄O₇ crystals. Appl. Opt. **47**: 3446–3454.
- 9. Oseledchik Yu S, Prosvirnin A L, Pisarevskiy A I, Starshenko V V, Osadchuk V V, Belokrys S P, Svitanko N V, Korol A S, Krikunov S A and Selevich A F, 1995. New nonlinear optical crystals: strontium and lead tetraborates. Opt. Mater. **4**: 669–674.
- 10. Pan F, Shen G, Wang R, Wang X and Shen D, 2002. Growth, characterization and nonlinear optical properties of SrB₄O₇ crystals. J. Cryst. Growth. **241**: 108–114.
- Martynyuk-Lototska I, Mys O, Adamiv V, Ya.Burak Ya and Vlokh R, 2002. Elastical, piezooptical and acoustooptical properties of lithium tetra borate crystals. Ukr. J. Phys. Opt. 3: 19–26.
- 12. Zhou Guoging, Xu Jun, Chen Xingda, Zhong Heyu, Wang Siting, Xu Ke, Deng Piezhen and Gan Fuxi, 1998. Growth and spectrum of a novel birefringent α -BaB₂O₄ crystal. J. Cryst. Growth. **191**: 517–519.
- Petrov V, Rotermund F, Noack F, Komatsu R, Sugawara T and Uda S, 1998. Vacuum ultraviolet application of Li₂B₄O₇ crystals: Generation of 100 fs pulses down to 170 nm. J. Appl. Phys. 84: 5887–5892.
- 14. Balakshii V I, Parygin V N and Chirkov L E, Physical fundamentals of acoustooptics. Moscow: Radio i Sviaz' (1985).

Martynyuk-Lototska I., Dudok T., Mys O., Romanyuk G. and Vlokh R., 2009. Acoustooptic interaction and photoelastic properties of $Li_2B_4O_7$ and α -BaB₂O₄ crystals at the wavelength of 442 nm Ukr.J.Phys.Opt. **10**: 218-225.

Анотація. В роботі досліджені акустооптичні і фото пружні властивості кристалів $Li_2B_4O_7$ і α -BaB_2O_4 на довжині хвилі оптичного випромінювання 442 нм. Показано, що ефективність акустооптичної дифракції суттєво зростає при 442 нм у порівнянні з 632.8 нм.