Highly efficient acoustooptic diffraction in α-BaB_2O_4 crystals: improving of geometry of acoustooptic interaction

Martynyuk-Lototska I.

Institute of Physical Optics, 23 Dragomanov St., 79005 Lviv, Ukraine,
iml@ifo.lviv.ua

Received: 25.01.2012

Abstract. In the present work we show experimentally on the example of α-BaB_2O_4 crystals that a choice of the slowest acoustic wave for acoustooptic interaction made on the basis of acoustic anisotropy of crystals can essentially increase the acoustooptic figure of merit. The latter parameter for α-BaB_2O_4 obtained by us is equal to M_2=(211±31)×$10^{-15}$$s^3$/kg. We demonstrate that the acoustooptic diffraction efficiency in α-BaB_2O_4 increases for shorter wavelengths of optical radiation and reaches almost ~ 75% at the wavelength 325 nm (at the acoustic signal power 0.04 W). This fact, along with transparency of those crystals in the ultraviolet spectral range and their high resistance against a powerful laser radiation, make them attractive materials for operating high-power ultraviolet laser radiation.

Keywords: acoustooptic diffraction, α-BaB_2O_4 crystals, acoustooptic figure of merit, acoustic anisotropy

PACS: 78.20.Hp
UDC: 535.42

1. Introduction

Crystals of borate family, such as α-BaB_2O_4, β-BaB_2O_4, $Li_2B_4O_7$, PbB_4O_7 and SrB_4O_7, are known as efficient materials for nonlinear optical applications (see [1–6]). A wide range of their optical transparency, particularly in the ultraviolet (UV) spectral region [7–11] (up to 125 nm for SrB_4O_7 [10, 11]), and a high level of optical damage threshold [11, 12] make them one of the best materials for operation of short-wavelength high-power laser radiation. As shown in the works [13–20], some of the borate crystals also manifest high enough acoustooptic figure of merit (abbreviated hereafter as AOFM). We have reported in our previous papers [14, 18] that, for example, β-BaB_2O_4 (the point group of symmetry 3m) is characterised with very low transverse acoustic wave velocities and could therefore be used as a promising acoustooptic (AO) material.

The AOFM for the β-BaB_2O_4 crystals [18] calculated on the basis of their photoelastic (PE) coefficients, refractive indices and the ultrasonic wave velocities is comparable with those typical for such well-known AO materials as lithium niobate or Pb_2MoO_5 [21]. Of course, the AOFM of such materials as, e.g., TeO_2 crystals, is much higher than that of the borates. However, the paratellurite is not transparent in the deep UV spectral range and the range of its transparency is limited by the wavelength of 350 nm [21]. On the other hand, KDP and SiO_2 crystals are transparent down to 250 nm and 150 nm, respectively, though their AOFM is rather low, $M_2 = (1-2.5)\times10^{-15}\,s^3/kg$ [21]. Contrary to the crystalline materials mentioned above, the borate crystals are transparent in the deep UV spectral range. For example, α-BaB_2O_4 is transparent down to 189 nm [22].

Thus, α-BaB_2O_4 could, in principle, turn out to be a promising material for the AO devices operating a short-wavelength optical radiation. Notice that the growing process for the β-BaB_2O_4 crystals is time-consuming, when compare to α-BaB_2O_4 (the point symmetry group $\bar{3}$m) [14]. Moreover, β- and α-phases of BaB_2O_4 actually represent respectively ferroelectric and ferroelastic modifications of a parent phase described by the point symmetry m3m. Let us remind that low acoustic wave velocities are peculiar for proper ferroelastics in the vicinity of their phase transition temperatures (see, e. g., [23]). On the other hand, the principal parameters determining the AOFM M_2 are the ultrasonic wave velocities v, the refractive indices n, the effective PE coefficients p_{eff}, and the crystal density ρ [21, 24-26]:

$$M_2 = \frac{n^6 p_{eff}^2}{\rho \mathrm{v}^3}. \qquad (1)$$

The authors of the works [27, 28] have attempted to search for geometries of AO interaction characterised by high values of AOFM. The method suggested there has been based upon revealing such OA diffraction geometries in which the effective PE coefficients reaches their highest values. For instance, this approach leads to the estimation 40×10^{-15} s^3/kg for the AOFM of β-BaB_2O_4 crystals at the light wavelength of $\lambda = 632.8$ nm [27], while we have even higher value (49.4×10^{-15} s^3/kg [14]) in the case of AO interaction with a slow, though not the slowest, acoustic wave propagated along the principal crystallographic directions. It is obvious (see Eq. (1)) that the AOFM is mainly influenced by the acoustic wave velocity which appears in the denominator raised to the third power. At the same time, M_2 is proportional only to the squared effective PE coefficient and so the latter affects the AOFM to a lesser degree. In other words, the anisotropy of the effective PE coefficient can determine the value of the AOFM only when the acoustic velocities do not reveal a prominent anisotropy, i.e. in case when it is impossible to find the acoustic wave with considerably low velocity for all the directions of acoustic propagation and polarisation in a crystal.

In our recent work [16], we have constructed the acoustic slowness surfaces for the α-BaB_2O_4 crystals basing on measurements of the acoustic wave velocities and derivation of the complete matrices of elastic stiffness coefficients. We have shown that the lowest velocity (943 m/s) is peculiar for the transverse acoustic wave of which the wave vector lies in (011) plane and makes the angle 58° with *z* axis (the latter is parallel to the optic axis of crystal while the *y* axis belongs to the symmetry mirror plane), whereas the polarisation belongs to (011) plane. The AOFM for the case of interaction with this wave should reach its highest value, $M_2 = (270\pm70)\times10^{-15}$ s^3/kg for $\lambda = 632.8$ nm One can compare this value with the AOFM $M_2 = (54.5\pm7.4)\times10^{-15}$ s^3/kg [16] which also refers to the α-BaB_2O_4 crystals and corresponds to the case of interaction with the acoustic wave having its propagation and polarisation directions parallel to the principal axes ([100] and [001], respectively). Notice that this high AOFM has been obtained following only from the calculations using Eq. (1) and the known values of the effective PE coefficient, refractive index and the acoustic wave velocity.

As a matter of fact, still there has been no experimental confirmation of such a high AOFM for the α-BaB_2O_4 crystals. Besides, the statement [16] that both the diffraction efficiency and the AOFM should increase essentially in the UV spectral range due to dispersion, when compare to

the corresponding values in the visible range, needs its experimental verification, too. As a consequence, the present work is devoted to experimental studies of AO diffraction in the α-BaB_2O_4 crystals dealing with the improved interaction geometry, in order to examine a real possibility for high values of AO efficiency in a wide spectral range, including the UV one.

2. Experimental

α-BaB_2O_4 crystals were grown with a standard Czochralski technique. Single crystals of a good optical quality, with $3\times3\times3$ cm^3 dimensions, were obtained after one-week growing process.

Experimental investigations of AO diffraction in α-BaB_2O_4 were carried out for the case when a transverse acoustic wave has been excited in α-BaB_2O_4 sample with a piezoelectric $LiNbO_3$ transducer. The electric circuit had the resistance of 50 ohm at the resonant frequency of 10.42 MHz. Intermediate indium and silver films were deposited between the $LiNbO_3$ transducer and the samples. The amount of electric power transferred to the acoustic wave was controlled with the aid of a pulse-echo technique. It was found that the total transfer losses were equal to 13.98 dB. The AO cell was made using a cold vacuum welding technique when connecting the AO material with the piezoelectric transducer (see Fig. 1).

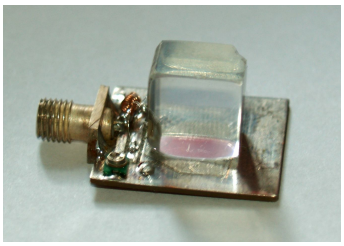

Fig. 1. An AO cell made on the basis of α-BaB_2O_4 crystals.

A transverse acoustic wave was excited in the sample of parallelepiped shape, with the wave vector lying in the (011) plane and making the angle $58°$ with the *z* axis and the polarisation belonging to the (011) plane (see above). After cutting and polishing of the sample we examined the velocity of this transverse wave and found that it was equal to 970 m/s, quite close to the calculated value (943 m/s [16]). The optical radiation propagated almost normal to the acoustic propagation direction so that the optical wave vector belonged to the (011) plane and the polarisation of the incident optical wave was parallel to the [100] axis.

In order to measure the AO diffraction efficiency in α-BaB_2O_4 depending on the electric power applied to transducer, we used He-Ne ($\lambda = 632.8$ nm) and He-Cd (λ=442 nm and 325nm) gas lasers, and second-harmonic radiation of the DPSS laser (the wavelength of 532 nm). The experimental setup is shown in Fig. 2.

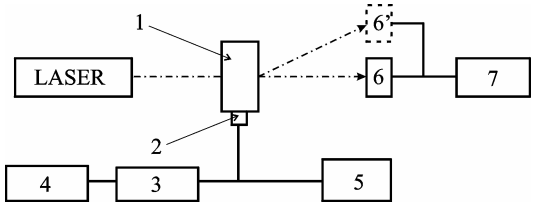

Fig. 2. Experimental set-up for the studies of AO diffraction: 1 – AO crystal; 2 – piezoelectric transducer; 3 – amplifier; 4 – high-frequency generator; 5 – oscilloscope; 6 and 6' – photodetectors; 7 – voltmeter.

A high-frequency electric signal from a generator 4 has been amplified with an amplifier 3 and then applied to piezoelectric transducer 2. The parameters of the electric signal at the transducer were detected with an oscilloscope 5. The incident optical radiation of a laser propagated through an AO crystal 1 in which the transducer excited transverse acoustic waves. The incident light intensity I_i and the intensity I_0 of zero-order diffraction maximum were measured with a semiconductor photodetector 6 and a voltmeter 7. The diffraction efficiency η was calculated using the relation

$$\eta = \frac{I_i - I_0}{I_i}, \tag{2}$$

where I_0 was measured as a function of driving electric power applied to the transducer. Notice that the diffracted beam intensity was equal to $I_d = I_i - I_0$. We determined the AO efficiency with the relation (2), because the intensities of the zero-order and the incident beams were comparable, thus allowing to measure these intensities in the same linear range of photodetector response. The error for the AO diffraction efficiency was about 2–13%.

3. Results and discussion

We have observed a diffraction of isotropic type in the geometry of AO interaction mentioned above. When the electric signal power applied to the transducer is less than 0.1 W, a single diffraction maximum appears, i.e. the diffraction in this case corresponds to the Bragg regime. The diffraction angle θ_d decreases with decreasing optical wavelength (see Fig. 3), thus agreeing well with the known relation that follows from the Bragg condition ($\theta_d = \arcsin(\lambda / 2\Lambda)$ or $\theta_d \approx \lambda / 2\Lambda$ if the angles are small, with λ / Λ being the ratio of the optical and acoustic wavelengths).

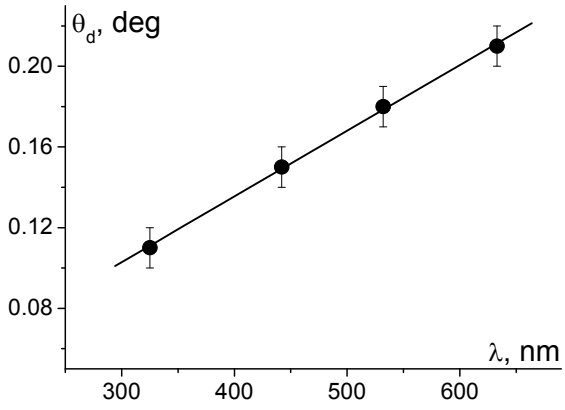

Fig. 3. Dependence of Bragg angle on the optical radiation wavelength for the α-BaB_2O_4 crystals: points correspond to experimental data and line to fitting by the linear dependence $\theta_d \approx \lambda / 2\Lambda$.

A nonlinear dependence of the diffraction efficiency on the electric signal power P_{el} which observed experimentally (see Fig. 4) follows the relation

$$\eta = \frac{I}{I_0} = \sin^2\left[\frac{\pi n L}{2\lambda \cos\theta_d}\frac{\Delta\varepsilon_i}{\varepsilon_i^0}\right] = \sin^2\left[\frac{\pi}{\lambda\cos\theta_d}\sqrt{\frac{P_a L M_2}{2H}}\right], \tag{3}$$

where $\Delta\varepsilon_i = \varepsilon_i^p - \varepsilon_i^0$ means the difference between the perturbed ($\varepsilon^p = 1/B_i^p$) and non-perturbed

($\varepsilon^0 = 1/B_i^0$) dielectric permittivities, B_i the optical-frequency dielectric impermeability, P_a the acoustic power, $L = 10$ mm and $H = 1$ mm are respectively the length and the width of the interaction region of acoustic and optical waves.

For the region of weak acoustooptic interaction, which corresponds to a linear range of the experimental dependence (see Fig. 4) the relation given by Eq. (3) can be simplified:

$$\eta \approx \frac{\pi^2 P_a L M_2}{2\lambda^2 H \cos^2 \theta_d} \tag{4}$$

The dispersion of the diffraction efficiency for the case of weak AO interaction (at the electric signal power equal to 0.1 W) determined from the data measured (Fig. 4) is presented in Fig. 5.

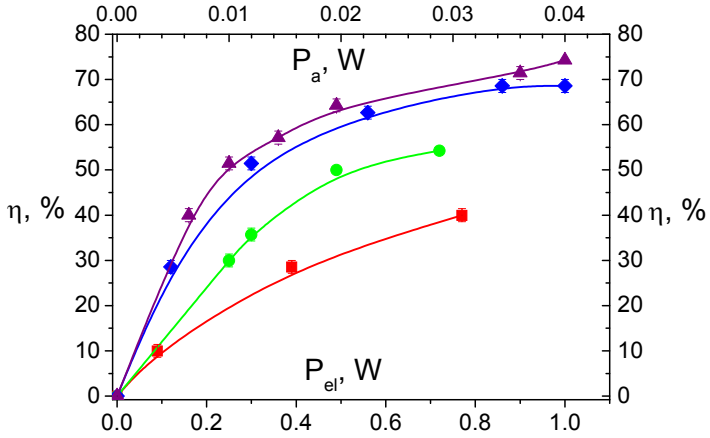

Fig. 4. Dependence of AO diffraction efficiency for the α-BaB_2O_4 crystals on electric signal power applied to piezoelectric transducer: squares correspond to $\lambda = 632.8$ nm, circles to $\lambda = 532$ nm, diamonds to $\lambda = 442$ nm, and triangles to $\lambda = 325$ nm.

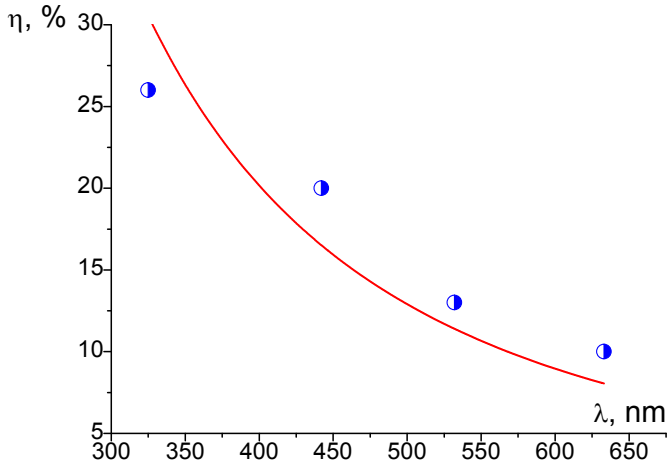

Fig. 5. Dispersion of AO diffraction efficiency at the electric signal power equal to 0.1 W: points correspond to experimental data and curve to fitting with Eq. (4).

As seen from Fig. 4 and Fig. 5, the AO diffraction efficiency for the α-BaB_2O_4 crystals increases for shorter wavelengths of optical radiation. At the wavelength of 325 nm it achieves

almost 75%, when the power of electric signal is 1 W (the corresponding acoustic power being equal to 0.04 W). This clearly demonstrates great prospects of α-BaB_2O_4 crystals prepared in the given orientation as a working element of the AO devices.

Let us now consider the diagram of AO interaction that corresponds to our experiment (see Fig. 6). It is seen that an isotropic AO interaction takes place here, with transferring of optical energy between the incident and diffracted beams with the wave vectors $k_{i,d}$ associated with the ordinary refractive index ($k_{i,d} = 2\pi n_o / \lambda$). Then one can calculate the AOFM of the α-BaB_2O_4 crystals for $\lambda = 632.8$ nm using the relation (3) and the experimental results shown in Fig. 4.

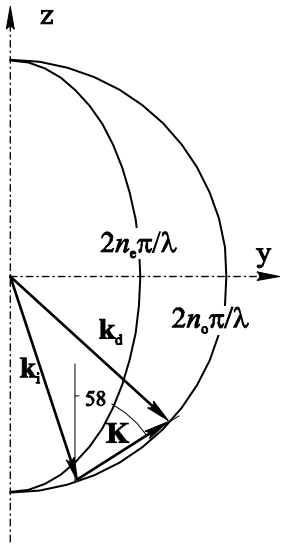

Fig. 6. Diagram of isotropic AO interaction with the slowest acoustic wave in α-BaB_2O_4 crystals.

What is important, the AOFM value obtained experimentally in the present work for $\lambda = 632.8$ nm ($M_2 = (211 \pm 31) \times 10^{-15}$ s^3/kg) is in good agreement with that theoretically estimated in our recent work [16] ($M_2 = (270 \pm 70) \times 10^{-15}$ s^3/kg). This AOFM is very high and, together with a transparency of α-BaB_2O_4 crystals in the UV spectral range and their high resistance against a laser radiation, this makes these crystals one of the most attractive materials for operating a powerful UV laser radiation.

4. Conclusions

In the present work we have proven experimentally a possibility for increasing essentially the efficiency of AO diffraction owing to a choice of the slowest acoustic wave for the AO interaction, basing on the acoustic anisotropy in crystals. It has been found on the example of α-BaB_2O_4 crystals that the AOFM can increase by an order of magnitude if one ensures the condition of AO interaction with the slowest acoustic wave. The AOFM experimentally obtained by us is equal to $M_2 = (211 \pm 31) \times 10^{-15}$ s^3/kg. It agrees well with that estimated theoretically ($M_2 = (270 \pm 70) \times 10^{-15}$ s^3/kg).

It has been revealed that the AO diffraction efficiency for the α-BaB_2O_4 crystals increases notably for shorter wavelengths of optical radiation. At the wavelength of 325 nm it reaches almost ~ 75% for the acoustic signal power equal to 0.04 W. This experimental fact, together with a

transparency in the UV spectral range and high resistance against the powerful laser radiation peculiar for the α-BaB_2O_4, prove that the above crystals should represent very attractive working elements of the AO devices designed for operating high-power ultraviolet laser radiation.

Acknowledgement

The author is grateful to Dr. E. Smirnov for fabricating AO cells, Mr. T. Dudok for his assistance during the experimental studies and Mr. I. Smaga for preparation of the sample.

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 : $Y_3Al_5O_{12}$ lasers. Appl. Phys. Lett. **70**: 3492–3494.
2. Ishida Y and Yajima T, 1987. Characteristics of a new-type SHG crystal β-BaB_2O_4 in the femtosecond region. Opt. Commun. **62**: 197–200.
3. Cheng L K, Bosenberg W and Tang C L, 1988. Broadly tunable optical parametric oscillation in β-BaB_2O_4. Appl. Phys. Lett. **53**: 175–177.
4. 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.
5. Petrov V, Rotermund F, Noack F, Komatsu R, Sugawara T and Uda S, 1998. Vacuum ultraviolet application of $Li_2B_4O_7$ crystals: generation of 100 fs pulses down to 170 nm. J. Appl. Phys. **84**: 5887–5892.
6. Petrov V, Noack F, Shen D, Pan F, Shen G, Wang X, Komatsu R and Alex V, 2004. Application of the nonlinear crystal SrB_4O_7 for ultra-fast diagnostics converting to wavelengths as short as 125 nm. Opt. Lett. **29**: 373–375.
7. Chen C, Wu B, Jiang A and You G, 1985. A new type ultraviolet SHG crystal – β-BaB_2O_4. Sci. Sinica, Ser. B. **28**: 235–243.
8. Bhar G, Das S and Chatterjee U, 1989. Evaluation of beta barium borate crystal for nonlinear devices. Appl. Opt. **28**: 202–204.
9. 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-BaB_2O_4 crystal. J. Cryst. Growth. **191**: 517–519.
10. Oseledchik Yu, Prosvirnin A, Pisarevskiy A, Starshenko V, Osadchuk V, Belokrys S, Svitanko N, Korol A, S. Krikunov A and Selevich A, 1995. New nonlinear optical crystals: strontium and lead tetraborates. Opt. Mater. **4**: 669–676.
11. Oseledchik Yu, Prosvirnin A, Svitanko N, Zhagrov A, Kudrjavtzev D and Shvets D, 2000. Electrooptical properties of strontium and lead tetraborate crystals. Visn. Zaporizh. Derzh. Univ. **1**: 189–193.
12. Vlokh R, Dyachok Ya, Krupych O, Burak Ya, Martynyuk-Lototska I, Andrushchak A and Adamiv V. 2003. Study of laser induced damage of borate crystals. Ukr. J. Phys. Opt. **4**: 101–104.
13. Martynyuk-Lototska I, Dudok T, Krupych O, Adamiv V, Smirnov Ye and Vlokh R, 2004. Acousto-optic diffraction in borate crystals. Ukr. J. Phys. Opt. **5**: 111–114.
14. Martynyuk-Lototska I, Mys O, Krupych O, Adamiv V, Burak Ya, Vlokh R and Schranz W, 2004. Elastic, piezooptic and acousto-optic properties of borate crystals (BaB_2O_4, $Li_2B_4O_7$ and $CsLiB_6O_{10}$). Integr. Ferroelectrics. **63**: 99–103.

15. Martynyuk-Lototska I, Dudok T, Mys O and Vlokh R, 2008. Elastic, piezooptic and acousto-optic properties of SrB_4O_7 and PbB_4O_7 crystals. Opt. Mater. **31**: 660–667.
16. Martynyuk-Lototska I, Mys O, Dudok T, Adamiv V, Smirnov Ye and Vlokh R, 2008. Acousto-optic interaction in α-BaB_2O_4 and $Li_2B_4O_7$ crystals. Appl. Opt. **47**: 3446–3454.
17. Martynyuk-Lototska I, Mys O, Adamiv V, Burak Ya and Vlokh R, 2002. Elastical, piezooptical and acousto-optic properties of lithium tetra borate crystals. Ukr. J. Phys. Opt. **3**: 264–266.
18. Andrushchak A, Adamiv V, Krupych O, Martynyuk-Lototska I, Burak Ya and Vlokh R, 2000. Anisotropy of piezo- and elastooptical effect in β-BaB_2O_4 crystals. Ferroelectrics. **238**: 299–305.
19. Mys O, 2008. Obliquity of the acoustic energy flow in the acousto-optic α-BaB_2O_4 and $Li_2B_4O_7$ crystals. Ukr. J. Phys. Opt. **9**: 256–260.
20. Aleksandrov K, Zamkov A, Zaitsev A, Turchin P, Sysoev A and Parfenov A, 2004. Acoustic and acousto-optic properties of lead tetraborate crystals. Fiz. Tverd. Tela. **46**: 1586–1587.
21. M.P. Shaskolskaya, Acoustic crystals (Moscow: Nauka, 1982).
22. http://www.casix.com
23. Vlokh R and Martynyuk-Lototska I 2009. Ferroelastic crystals as effective acoustooptic materials. Ukr. J. Phys. Opt. **10**: 89–99.
24. A. Yariv and P. Yeh, Optical waves in crystals: propagation and control of laser radiation (New York: Wiley, 1984).
25. J Xu and R Stroud, Acousto-optic devices: principles, design, and applications (New York: Wiley, 1992).
26. A Goutzoulis and D Pape, Design and fabrication of acousto-optic devices (New York: Marcel Dekker, 1994).
27. Andrushchak A S, Bobitski Ya V, Kaidan M V, Tybinka B V, Kityk A V and Schranz W, 2004. Spatial anisotropy of photoelastic and acoustooptic properties in β-BaB_2O_4 crystals, Opt. Mater. **27** 619–624.
28. Andrushchak A S, Bobitski Ya V, Kaidan M V, Mytsyk B G, Kityk A V and Schranz W, 2005. Two-fold interferometric measurements of piezo-optic constants: Application to β-BaB_2O_4 Crystals, Opt. & Laser Technol. **37:** 319–328.
29. Martynyuk-Lototska I, Dudok T, Mys O, Romanyuk G and Vlokh R, 2009. Acoustooptic interaction and photoelastic properties of $Li_2B_4O_7$ and alpha-BaB_2O_4 crystals at the wavelength of 442 nm. Ukr. J. Phys. Opt. **10**: 218–225.

Martynyuk-Lototska I., 2012. Highly efficient acoustooptic diffraction in α-BaB_2O_4 crystals: improving of geometry of acoustooptic interaction. Ukr.J.Phys.Opt. **13**: 28 – 35.

Анотація. *На прикладі кристалів α-BaB_2O_4 експериментально показано, що вибір повільної акустичної хвилі для акустооптичної взаємодії на основі акустичної анізотропії кристалів може значно збільшити коефіцієнт акустооптичної якості. Коефіцієнт акустооптичної якості для α-BaB_2O_4, отриманий експериментально, дорівнює $M_2=(211\pm31)\times10^{-15} c^3/кг$. Показано, що ефективність акустооптичної дифракції в α-BaB_2O_4 зростає зі зменшенням довжини хвилі оптичного випромінювання і досягає майже ~ 75% на довжині хвилі оптичного випромінювання 325 нм та при акустичній потужності сигналу 0,04 Вт. Цей факт, разом з прозорістю цих кристалів в ультрафіолетовій області спектру та їхньою високою стійкістю до потужного лазерного випромінювання, робить їх привабливим матеріалом для керування потужним ультрафіолетовим лазерним випромінюванням*