Acoustooptic and acoustic properties of KLiB$_4$O$_7$ crystals

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Abstract

The acoustic and acoustooptic (AO) properties of KLiB$_4$O$_7$ crystals are experimentally studied and analysed. It is shown that the above crystals can be used as a material for AO applications. The AO figure of merit calculated for the slowest acoustic wave is equal to M$_2$=(332±120)×10^{-15}s^3/kg. Propagation of the acoustic energy flow in KLiB$_4$O$_7$ for the cases of transverse and longitudinal acoustic waves is considered. It is shown that the obliquity for the slowest transverse wave is relatively small and so it cannot affect essentially the efficiency of AO interaction.

Key words: KLiB$_4$O$_7$ crystals, acoustooptic figure of merit, acoustooptic interaction, acoustic waves, phase velocity, acoustic energy flow.

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Introduction

The borate family crystals such as α-BaB$_2$O$_4$, β-BaB$_2$O$_4$, Li$_2$B$_4$O$_7$, PbB$_4$O$_7$ and SrB$_4$O$_7$ are known as efficient materials for nonlinear optical, acoustooptic (AO) and acoustic applications (see [1–14]). A wide range of their optical transparency, particularly in the ultraviolet region [15–19] (up to 125 nm for SrB$_4$O$_7$ [18, 19]), and a high level of optical damage threshold [19, 20] make them attractive for optoelectronics.

KLiB$_4$O$_7$ crystals abbreviated hereafter as KLTB are grown by using isovalent substitution of half of Li atoms by K ones in the initial Li$_2$B$_4$O$_7$ compound [21, 22]. At the room temperature KLTB belongs to the acentric point group of symmetry 222 [21, 22], the lattice parameters being $a = 8.4915$A , $b = 11.1415$A and $c = 12.6558$A [22].

In our previous works [23, 24] some optical, acoustic and AO properties of the KLTB crystals have been studied. It has been shown that the AO figure of merit (AOFM) reaches the value of $M_2 = 32 \times 10^{-15}$s^3/kg for the case of propagation of transverse acoustic wave along the crystallographic direction [010], its polarization parallel to [001], the light wavelength of $\lambda = 632.8$ nm and the temperature $T = 293$ K [24]. It is necessary to note that, in principle, one can achieve a higher AOFM since the acoustic wave mentioned above could turn out to be not the slowest one, due to the acoustic anisotropy

effect. Therefore the aim of the present paper is to study anisotropy of the acoustic and AO properties of the KLTB. We address also the obliquity of the acoustic energy flow with respect to the wave normal which can give rise to changing conditions of AO interactions.

Results and discussion

Propagation of acoustic waves in crystals may be described in terms of the Christoffel equation [25, 26],

$$C_{ijkl}m_jm_kq_l = \rho\upsilon^2q_l,\qquad(1)$$

where C_{ijkl} denote the components of the elastic stiffness tensor, m_j the components of unit vector of the wave normal, q_l the components of the unit displacement vector, ρ the density of material, and υ the phase acoustic velocity. The true phase velocities are given by eigenvalues of Eq. (1). From Eq. (1) it follows that three acoustic waves can propagate along a given direction, with their displacement vectors being mutually orthogonal.

In a general case, the values of the phase and group velocities are different in acoustically anisotropic media. This can manifest itself in the obliquity of direction of the acoustic energy flow with respect to the wave normal. The components W_j of the group velocity vector may be determined from the transformed Christoffel equation [25, 26]:

$$W_j = \frac{1}{\rho\upsilon}C_{ijkl}q_iq_lm_k.\qquad(2)$$

The angle between the directions of the energy flow and the wave normal (i.e., the quantitative parameter of the obliquity) may be calculated using the relation [27]

$$\tan(\varphi-\psi) = \frac{1}{\upsilon(\varphi)}\frac{\partial\upsilon}{\partial\varphi},\qquad(3)$$

where φ is the angle between the crystallographic axis and the wave normal direction, ψ the angle between the crystallographic axis and the acoustic energy flow, and $\upsilon(\varphi)$ the phase acoustic wave velocity for the given direction of acoustic wave propagation. The relations describing changes in the acoustic wave velocity depending on the direction ($\upsilon(\varphi)$) may be obtained from the Christoffel equation.

Let us consider propagation of longitudinal and transverse acoustic waves in the KLTB crystals. The acoustic wave velocities and the stiffness tensor coefficients for the KLTB have already been determined in our previous works [24]. The corresponding surfaces of the acoustic wave velocities are shown in Fig. 1.

As seen from Fig. 1, the minimal acoustic wave velocity (QT_1) for the KLTB corresponds to the transverse acoustic wave propagating along the direction that lies in (011) plane (the angle 45° with the crystallographic axis c). Its displacement vector belongs to $(01\bar{1})$ plane and is oriented at the angle of 45° with respect to that
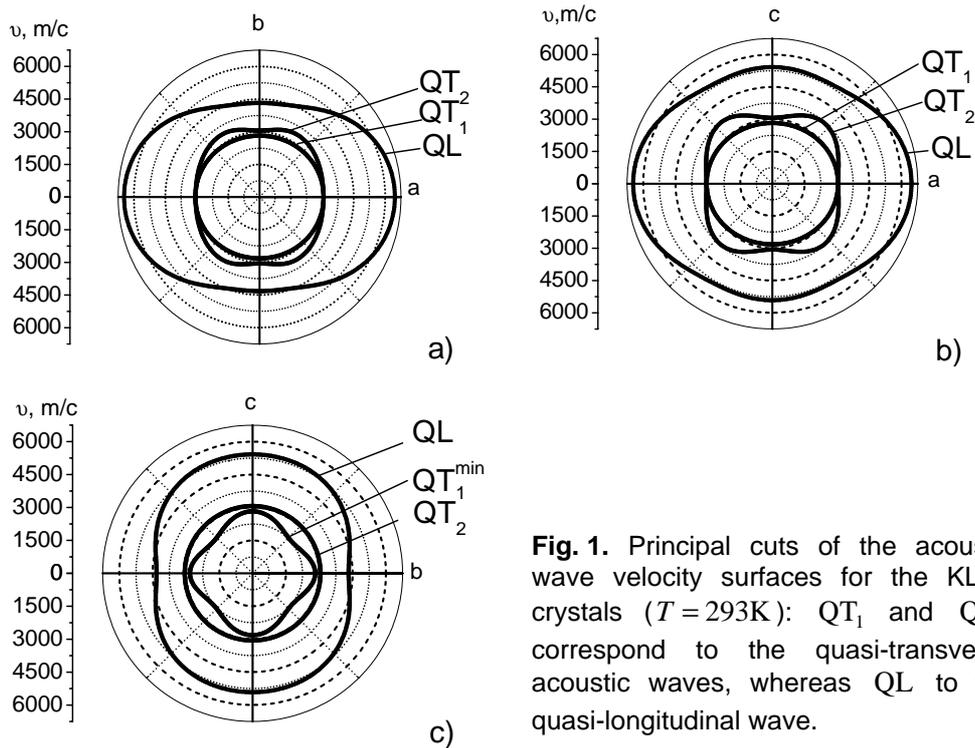
Fig. 1. Principal cuts of the acoustic wave velocity surfaces for the KLTB crystals ($T = 293\mathrm{K}$): QT_1 and QT_2 correspond to the quasi-transverse acoustic waves, whereas QL to the quasi-longitudinal wave.

crystallographic axis (see Fig. 1c). For this case one can evaluate the AOFM for different geometries of AO interactions. The conditions of AO interactions are presented in Fig. 2.

Given the mentioned polarization of the acoustic wave, the strain tensor may be written as

$$\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0.5e_{ef} & -0.5e_{ef} \\ 0 & -0.5e_{ef} & 0.5e_{ef} \end{bmatrix}, \tag{4}$$

where e_{eff} means the effective strain caused by displacement parallel to the polarization direction. The equation of the optical indicatrix perturbed by the strains given by Eq. (4) could be written in the following form:

$$\begin{aligned} &\left(B_1 + 0.5\left(p_{12} + p_{13}\right)e_{ef}\right)x^2 + \left(B_2 + 0.5\left(p_{22} + p_{23}\right)e_{ef}\right)y^2 \\ &+ \left(B_3 + 0.5\left(p_{32} + p_{33}\right)e_{ef}\right)z^2 - p_{44}e_{ef}\,yz = 1 \end{aligned}, \tag{5}$$

where B_i are the initial optical-frequency dielectric impermeability components. Now we will derive the relations describing the AO interaction presented in Fig 2a. Let the incident light propagate along the direction c and the electric displacement vector be directed along the crystallographic direction b (see Fig. 2a, case 1). Accounting for Eq. (5), we can write the relations for the AO interaction as

$$\begin{cases} \Delta E_2^{\omega+\Omega} = 0.5(p_{22} + p_{23} - 2p_{24})e_{ef}^{\Omega}D_2^{\omega} \\ \Delta E_3^{\omega+\Omega} = 0.5(p_{42} + p_{43} - 2p_{44})e_{ef}^{\Omega}D_2^{\omega} \end{cases}, \tag{6}$$
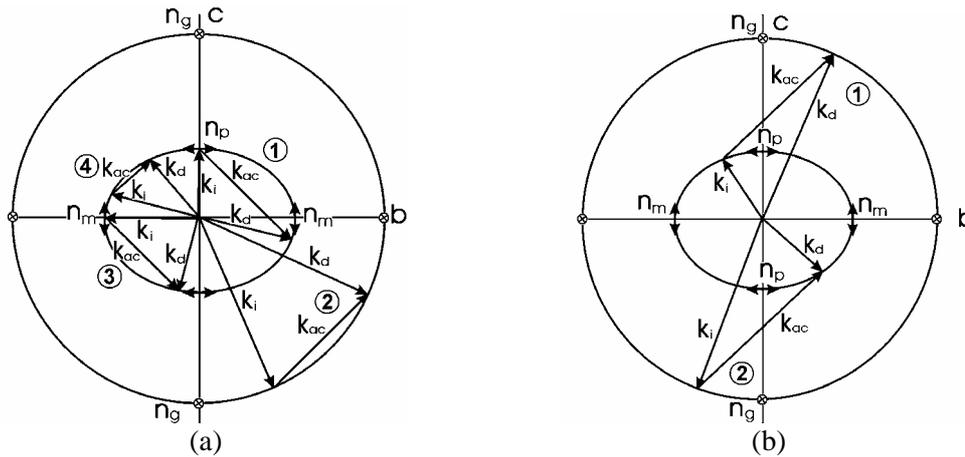
Fig. 2. Schematic diagrams of AO interaction with the slowest acoustic wave propagated along [011] direction in the KLTB (the velocity $v_j = 2280\,\text{m/s}$).

where $\Delta E_j^{\omega+\Omega}$ and D_i^{ω} are respectively the components of the electric field of the diffracted optical wave and the electric displacement for the incident wave.

The photoelastic coefficients p_{42}, p_{43} and p_{24} for the symmetry group 222 are equal to zero, so that Eqs. (6) become as follows:

$$\begin{cases} \Delta E_2^{\omega+\Omega} = 0.5(p_{22} + p_{23})e_{ef}^{\Omega}D_2^{\omega} \\ \Delta E_3^{\omega+\Omega} = -p_{44}e_{ef}^{\Omega}D_2^{\omega} \end{cases}. \tag{7}$$

It follows from Eqs. (7) that the case 1 in Fig. 2a corresponds to anisotropic type of AO interaction. Let us estimate the corresponding AOFM. The direction of polarization of the diffracted light may be obtained after calculating the angle between the acoustic wave and the diffracted light on the basis of Fig. 2 (case 1) and using some algebra. The polarization vector of the diffracted light in the KLTB crystals makes the angle 2° with the crystallographic axis c. Therefore Eqs. (7) read as

$$\begin{cases} \Delta E_2^{\omega+\Omega} = 0.49\left(p_{22}e_{ef}^{\Omega} + p_{23}e_{ef}^{\Omega}\right)D_2^{\omega} \\ \Delta E_3^{\omega+\Omega} = -0.04 p_{44}e_{ef}^{\Omega}D_2^{\omega} \end{cases}. \tag{8}$$

Notice that nearly isotropic AO interaction takes place in this case. The relation between the electric field $\Delta E_j^{\omega+\Omega}$ of the diffracted optical wave and the electric displacement of the incident light is as follows:

$$\Delta E_d^{\omega+\Omega} = \sqrt{0.24\left(p_{22} + p_{23}\right)^2 + \left(-0.04 p_{44}\right)^2}\, e_{ef}^{\Omega}D_2^{\omega}. \tag{9}$$

The AOFM is defined as (see, e.g., [25])

$$M_2 = \frac{n_2^6 p_{ef}^2}{\rho v^3}. \tag{10}$$

Using the values of piezooptic coefficients and the elastic stiffness tensor coefficients obtained in our recent study [24], one can calculate the photoelastic coefficients with the standard relation (see [26])

$$p_{ijmn} = \pi_{ijkl}C_{klmn}.$$ (11)

These coefficients are as follows: $p_{11} = (0.2 \pm 0.1)$, $p_{12} = (-0.36 \pm 0.13)$, $p_{21} = (-0.5 \pm 0.3)$, $p_{22} = (-0.8 \pm 0.2)$, $p_{23} = (-0.78 \pm 0.19)$, $p_{31} = (0.49 \pm 0.19)$, and $p_{44} = (0.4 \pm 0.1)$, while the error associated with determination of the coefficients p_{13}, p_{32} and p_{33} is about 100%. The p_{ef} value involved in Eq. (9) is equal to

$$p_{ef} = \sqrt{0.24\left(p_{22} + p_{23}\right)^2 + \left(-0.04 p_{44}\right)^2} = \left(0.8 \pm 0.3\right)$$ and the rest of the parameters are $\rho = 2190\,\text{kg/m}^3$, $\upsilon = 2280\,\text{m/s}$ and $n_2 = 1.54275$. Then one finally gets the AOFM: $M_2 = (332 \pm 120) \times 10^{-15}\,\text{s}^3/\text{kg}$.

For the incident light polarised along the crystallographic direction a (see Fig. 2a, case 2) the following relation describing the AO interaction holds true:

$$\Delta E_1^{\omega \pm \Omega} = 0.5(p_{12} + p_{13})e_{ef}^{\Omega}D_1^{\omega}.$$ (12)

In this case a purely isotropic type of AO interaction is realised. The p_{ef} value in Eq. (12) is $p_{ef} = 0.5\left(p_{12} + p_{13}\right) = \left(-0.5 \pm 0.2\right)$. Then on the basis of parameters $\rho = 2190\,\text{kg/m}^3$, $\upsilon = 2280\,\text{m/s}$ and $n_1 = 1.58236$ one gets the corresponding AOFM ($M_2 = (140 \pm 50) \times 10^{-15}\,\text{s}^3/\text{kg}$).

For the cases 3 and 4 (see Fig 2a) we have respectively the relations

$$\begin{cases} \Delta E_2^{\omega + \Omega} = -p_{44}e_{ef}^{\Omega}D_3^{\omega} \\ \Delta E_3^{\omega + \Omega} = 0.5\left(p_{32}e_{ef}^{\Omega} + p_{33}e_{ef}^{\Omega}\right)D_3^{\omega} \end{cases},$$ (13)

$$\begin{cases} \Delta E_2^{\omega + \Omega} = 0.5\left(p_{22}e_{ef}^{\Omega} + p_{23}e_{ef}^{\Omega}\right)D_2^{\omega} - p_{44}e_{ef}^{\Omega}D_3^{\omega} \\ \Delta E_3^{\omega + \Omega} = -p_{44}e_{ef}^{\Omega}D_2^{\omega} + 0.5\left(p_{32}e_{ef}^{\Omega} + p_{33}e_{ef}^{\Omega}\right)D_3^{\omega} \end{cases}.$$ (14)

Here Eqs. (14) describe a general case of AO interaction of the optical wave with the transverse acoustic wave propagating along (011) direction, with the displacement vector almost parallel to $(01\bar{1})$ (Fig. 2a). In order to estimate AOFM values following from Eqs. (14) it is necessary to know the direction of incidence of light and the acoustic wave frequency. Notice that the equations considered above are partial cases of Eqs. (14).

One can readily evaluate the AOFM for the case described by Eqs. (13) using the method adopted for the case described by Eqs. (7). For the optical wave propagating along the crystallographic direction b, the polarization vector of the diffracted light

propagating in the (011) plane makes the angle $2°$ with respect to the b axis. Then nearly isotropic AO interaction takes place. Eqs. (13) may be rewritten resulting in

$$\begin{cases} \Delta E_2^{\omega+\Omega} = -0.04\, p_{44} e_{ef}^{\Omega} D_3^{\omega} \\ \Delta E_3^{\omega+\Omega} = 0.49\left(p_{32} + p_{33}\right) e_{ef}^{\Omega} D_3^{\omega} \end{cases}. \tag{15}$$

Unfortunately, the p_{ef} value entering in Eqs. (13) could be determined with high enough error. We note here only that the AOFM for this case of AO interaction should be smaller than $\sim 50\times10^{-15}\,\mathrm{s}^3/\mathrm{kg}$.

The anisotropic types of AO interaction (see Fig 2b, cases 1 and 2) are described by the relations

$$\begin{aligned} \Delta E_1^{\omega+\Omega} &= 0.5\left(p_{62} e_{ef}^{\Omega} + p_{63} e_{ef}^{\Omega} - 2 p_{64} e_{ef}^{\Omega}\right) D_2^{\omega} + \\ &+ 0.5\left(p_{52} e_{ef}^{\Omega} + p_{53} e_{ef}^{\Omega} - 2 p_{54} e_{ef}^{\Omega}\right) D_3^{\omega} = 0 \end{aligned}, \tag{16}$$

$$\begin{cases} \Delta E_2^{\omega+\Omega} = 0.5\left(p_{62} e_{ef}^{\Omega} + p_{63} e_{ef}^{\Omega} - 2 p_{64} e_{ef}^{\Omega}\right) D_1^{\omega} = 0 \\ \Delta E_3^{\omega+\Omega} = 0.5\left(p_{52} e_{ef}^{\Omega} + p_{53} e_{ef}^{\Omega} - 2 p_{54} e_{ef}^{\Omega}\right) D_1^{\omega} = 0 \end{cases}. \tag{17}$$

These types of interactions cannot be implemented in practice since $p_{62} = p_{63} = p_{64} = p_{52} = p_{53} = p_{54} = 0$.

For the acoustic wave propagating along the direction that makes the angle $45°$ with respect to the crystallographic axis c in (011) plane, an obliquity of the acoustic energy flow can appear, leading to decreasing efficiency of the AO interaction. It is necessary to note that, in a general case, the displacement vector of the acoustic wave propagating oblique to the principal crystallographic directions cannot be orthogonal to the acoustic wave propagation direction. To calculate correctly the group velocity and the obliquity of acoustic energy flow, non-orthogonality of the displacement vector and the acoustic wave normal should be accounted for. It particular, this is of primary importance when deriving the relation that determines the effective photoelastic coefficients and, as a result, the conditions for the AO interaction.

Using the Christoffel equation, one can calculate the deviation angle Δ of acoustic polarization for the wave propagating in the KLTB crystals in (011) plane. It determines distinction of the acoustic polarization from its purely longitudinal type and may be found from the relation [25]

$$\Delta = \varphi - \frac{1}{2}\arctan\left[\frac{\sin\varphi\left(C_{23} + C_{44}\right)}{\cos\varphi\left(C_{22} - C_{44}\right) + \sin\varphi\left(C_{44} - C_{33}\right)}\right]. \tag{18}$$

Non-orthogonality of the quasi-transverse wave may also be calculated with Eq. (18), but the angle of $90°$ should then be added.

The dependence of deviation of the displacement vector from the propagation direction of the acoustic wave is shown in Fig. 3. As seen from Fig. 3, the deviation of
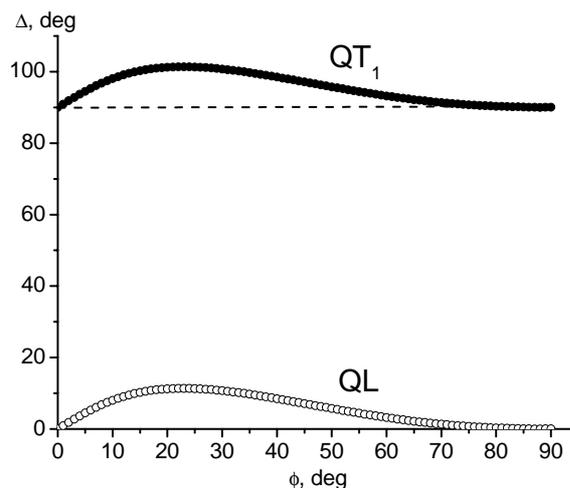
Fig. 3. Dependences of deviation of the displacement vector from the wave vector direction for the longitudinal and transverse acoustic waves propagating in (011) plane ($T = 293\,\text{K}$).

about $7°$ can appear for the acoustic wave propagating along the direction located in (011) plane and inclined by $45°$ with respect to the crystallographic axis c.

This deviation cannot notably affect the accuracy of determination of the effective photoelastic coefficients and, thus, the precision of the conditions of AO interaction.

Let us now consider propagation of the acoustic energy flow in the (011) plane of the KLTB crystals. We have calculated the obliquity for the acoustic energy flow from Eq. (3). The dependences of the obliquity value on the propagation direction of the acoustic wave for both the longitudinal and transverse waves are shown in Fig. 3.

As seen from Fig. 4, the obliquity is equal about to $7°$ for the case of transverse acoustic wave QT_1 propagating in the (011) plane along the direction inclined by $45°$ with respect to the c axis. In principle, influence of the obliquity on the AO interaction efficiency should be taken into account under these experimental conditions.
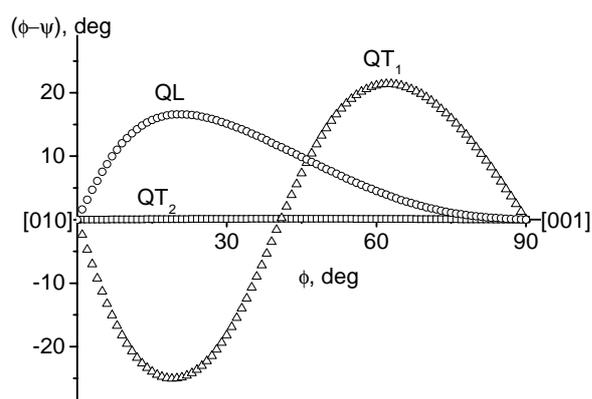
Fig. 4. Dependences of the obliquity angle for the longitudinal and transverse acoustic waves on the direction of acoustic propagation in the KLTB crystals ($T = 293\,\text{K}$).

Nevertheless, the obliquity is quite small and so does not impose a significant effect. The largest acoustic obliquity ($-26°$) have been obtained for the transverse acoustic wave QT_1 which is polarised along $[01\bar{1}]$ direction and propagates in the (011) plane under the angle of $\sim 20°$ with respect to the b axis ($70°$ with the c axis). It is seen from Fig. 1c that the latter direction corresponds to a maximal angular change in the acoustic wave velocity. In other terms, the results obtained by us agree well with the known theoretical predictions [26]: the highest value of the acoustic obliquity should take place for the direction characterised with the highest angular changes in the acoustic velocity.

Notice also that zero acoustic obliquity has to correspond to the condition $\dfrac{\partial \upsilon}{\partial \varphi} = 0$.

Hence, on the basis of the results for the KLTB crystals obtained above one can conclude the following. The obliquity of the acoustic energy flow, which has been determined for the transverse acoustic wave propagating in the (011) plane along the direction inclined by $45°$ with respect to the crystallographic axis c, should be accounted for when designing AO devices, though the effect cannot affect notably the efficiency of the AO interaction in any practical AO experiment.

Conclusions

1. The AO properties of the KLTB crystals are thoroughly analysed. It is shown that the KLTB may be used as a promising material for AO applications. The AOFM calculated for the slowest acoustic wave velocity is equal to $M_2 = (332 \pm 120) \times 10^{-15}\,\text{s}^3/\text{kg}$. This is comparable with the values peculiar for highly efficient AO materials.

2. The obliquity of the acoustic energy flow is analysed for the both longitudinal and transverse acoustic waves propagating in the KLTB crystals. It is shown that the deviation of the energy flow direction from the wave normal is equal only to $7°$ under the experimental conditions corresponding to the highest AOFM and arising from the interaction with the slowest acoustic wave. This allows for an efficient AO interaction.

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Ukr.J.Phys.Opt. **10**: 165-174.

Анотація. У даній роботі експериментально дослідженні і проаналізовані акустичні і акустооптичні (АО) властивості кристалів KLiB$_4$O$_7$. Показано, що ці кристали можуть використовуватись, як акустооптичні матеріали а розрахований коефіцієнт АО якості для найповільнішої акустичної хвилі становить M$_2$=(332±120)×10^{-15}с^3/кг. *Розглянуте поширення потоку акустичної енергії в кристалах KLiB$_4$O$_7$ для повздовжніх і поперечних акустичних хвиль і показано, що „знос" потоку енергії для найповільнішої акустичної хвилі є досить незначним і не може суттєво вплинути на ефективність АО взаємодії.*

Retraction

Behaviour of pulsed vortex beam with high topological charge in ionized dielectrics

Khasanov O.K., Fedotova O.M., Smirnova T.V., Petukh Yu.N., Volyar A.V. and Sukhorukov A.P. 2008. Ukrainian Journal of Physical Optics 9: 10-21.

It has come to the attention of Editorial Board of Ukr.J.Phys.Opt. that this article should not have been submitted for publication owing to its substantial replication of an earlier paper (R.A. Vlasov, O.K. Khasanov and T.V. Smirnova 2005. Evolution of tubular singular pulsed beams upon ionization. Quantum Electronics 35: 947-952).

Consequently this paper has been retracted by the Editorial Board of Ukr.J.Phys.Opt.