Piezooptic Properties of $\alpha$-BaB$_2$O$_4$ and Li$_2$B$_4$O$_7$ Crystals

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Received: 18.12.2003

Abstract

Photoelastic coefficients of $\alpha$-BaB$_2$O$_4$ and Li$_2$B$_4$O$_7$ crystals are calculated on the basis of piezooptic measurements performed with interferometric technique and the elastic compliance and stiffness data. Using the experimental results, the acoustooptic (AO) figure of merit (FM) has been estimated for the possible geometries of AO interaction. It is shown that the AO FM for the ABO and LTB crystals reach respectively the values $M^2=243.4 \times 10^{-15}$ s$^3$/kg and $M^2=2.57 \times 10^{-15}$ s$^3$/kg, if the interaction with the slowest ultrasonic waves ($v=933.5$ m/s and $v=3173$ m/s) is concerned. The directions of propagation and polarization of those acoustic waves are obtained on the basis of construction of indicative surfaces of the acoustic wave velocities.

Key words: piezooptic effect, borate crystals, photoelastic effect

PACS 78.20.Hp

Introduction

$\alpha$-BaB$_2$O$_4$ and Li$_2$B$_4$O$_7$ crystals belong to borate crystal family and are described respectively with the point symmetry groups 3m and 4mm. Acentric borate crystals are widely used as nonlinear optical materials due to high values of their nonlinear susceptibilities [1,2] and a high level of optical damage threshold [3]. In our previous papers [4,5] we have reported that, for example, $\beta$-BaB$_2$O$_4$ is characterized with quite low transverse acoustic wave velocities and could be therefore used as a promising acoustooptic (AO) material. The value of AO figure of merit (abbreviated hereafter as AOFM) for $\beta$-BaB$_2$O$_4$ crystals [5] calculated on the basis of photoelastic coefficients, refractive indices and the ultrasonic wave velocities is comparable with those typical for the well-known AO materials such as lithium niobate or Pb$_2$MoO$_5$ [6]. However, the growing process for $\beta$-BaB$_2$O$_4$ crystals is time-consuming, when compare to $\alpha$-BaB$_2$O$_4$ and Li$_2$B$_4$O$_7$. On the other side, the photoelastic parameters that affect the AOFM of $\alpha$-BaB$_2$O$_4$ and Li$_2$B$_4$O$_7$ crystals are still unknown (to our knowledge, only $p_{66}$ coefficient has been determined for Li$_2$B$_4$O$_7$ crystals [7]). This is why we report below the results of studies of piezooptic effect in these crystals, together with the estimation of AOFM.

Experimental results

$\alpha$-BaB$_2$O$_4$ and Li$_2$B$_4$O$_7$ crystals were grown with the Czochralski method. Single crystals of a good optical quality with $3\times3\times3$ cm$^3$ dimensions were obtained after a one-week growing process. Their piezooptic coefficients were measured at room temperature with interferometric technique using the Mach-Zender interferometer ($\lambda=632.8$ nm). For avoiding ambiguity in the presentation of results, the elastic contribution was derived with the aid of the relationship
\(\delta(\Delta nd)_{ij} = \pi_{ijkl}\sigma_{kl} - (n_c - 1)S_{ijkl}\sigma_{ij}\). The ultrasonic wave velocities were measured at room temperature with the pulse-echo overlap method [8]. The accuracy for the absolute velocity values was about 0.5%. The acoustic waves in samples were excited with LiNbO\(_3\) transducers characterized with the resonance frequency of \(f = 10\) MHz, the bandwidth of \(\Delta f = 0.1\) MHz and the acoustic power from \(P_a = 1\) to \(2W\). The photoelastic coefficients were calculated on the basis of elastic stiffness data obtained earlier [9], using the known formula \(p_{\lambda\nu} = \pi_{\lambda\nu}C_{\nu\mu}\).

The results for the piezooptic coefficients of Li\(_2\)B\(_4\)O\(_7\) crystals are presented below in the form of matrix:

\[
\begin{pmatrix}
\pi_{11} & \pi_{12} & \pi_{13} & 0 & 0 & 0 \\
\pi_{12} & \pi_{11} & \pi_{13} & 0 & 0 & 0 \\
\pi_{31} & \pi_{31} & \pi_{33} & 0 & 0 & 0 \\
0 & 0 & 0 & \pi_{44} & 0 & 0 \\
0 & 0 & 0 & \pi_{44} & 0 & 0 \\
0 & 0 & 0 & \pi_{66} & 0 & 0 \\
\end{pmatrix}
= \begin{pmatrix}
-2.75 & -0.45 & 1.08 & 0 & 0 & 0 \\
-0.45 & -3.52 & 1.08 & 0 & 0 & 0 \\
-2.53 & -2.53 & 2.9 & 0 & 0 & 0 \\
0 & 0 & 0 & 1.1 & 0 & 0 \\
0 & 0 & 0 & 1.1 & 0 & 0 \\
0 & 0 & 0 & 0 & 0.15 & 0 \\
\end{pmatrix} \times 10^{-12} \text{ m}^2 / \text{N}
\]

while for \(\alpha\)-BaB\(_2\)O\(_4\) we have

\[
\begin{pmatrix}
\pi_{11} & \pi_{12} & \pi_{13} & \pi_{14} & 0 & 0 \\
\pi_{12} & \pi_{11} & \pi_{13} & -\pi_{14} & 0 & 0 \\
\pi_{31} & \pi_{31} & \pi_{33} & 0 & 0 & 0 \\
\pi_{41} & \pi_{41} & \pi_{44} & 0 & 0 & 0 \\
0 & 0 & 0 & \pi_{44} & 2\pi_{41} & 0 \\
0 & 0 & 0 & \pi_{66} & 0 & 0 \\
\end{pmatrix}
= \begin{pmatrix}
1.58 & 2.08 & -4.32 & -14.22 & 0 & 0 \\
2.08 & 1.58 & -4.32 & 14.22 & 0 & 0 \\
1.52 & 1.52 & -7.24 & 0 & 0 & 0 \\
2.85 & 2.85 & 0 & -24.58 & 0 & 0 \\
0 & 0 & 0 & 0 & -24.58 & 5.7 \\
0 & 0 & 0 & 0 & -14.22 & 0.5 \\
\end{pmatrix} \times 10^{-12} \text{ m}^2 / \text{N}
\]

The calculated values of photoelastic coefficients for Li\(_2\)B\(_4\)O\(_7\) crystals are as follows:

\[
\begin{pmatrix}
p_{11} & p_{12} & p_{13} & 0 & 0 & 0 \\
p_{12} & p_{11} & p_{13} & 0 & 0 & 0 \\
p_{31} & p_{31} & p_{33} & 0 & 0 & 0 \\
0 & 0 & 0 & p_{44} & 0 & 0 \\
0 & 0 & 0 & p_{44} & 0 & 0 \\
0 & 0 & 0 & p_{66} & 0 & 0 \\
\end{pmatrix}
= \begin{pmatrix}
-0.32 & -0.04 & -0.06 & 0 & 0 & 0 \\
-0.04 & -0.32 & -0.06 & 0 & 0 & 0 \\
-0.24 & -0.24 & -0.02 & 0 & 0 & 0 \\
0 & 0 & 0 & 0.05 & 0 & 0 \\
0 & 0 & 0 & 0.05 & 0 & 0 \\
0 & 0 & 0 & 0.06 & 0 & 0 \\
\end{pmatrix}
\]

The same values for \(\alpha\)-BaB\(_2\)O\(_4\) crystals may be written as

\[
\begin{pmatrix}
p_{11} & p_{12} & p_{13} & p_{14} & 0 & 0 \\
p_{12} & p_{11} & p_{13} & -p_{14} & 0 & 0 \\
p_{31} & p_{31} & p_{33} & 0 & 0 & 0 \\
-p_{41} & 0 & p_{44} & 0 & 0 & 0 \\
0 & 0 & 0 & p_{44} & p_{41} & 0 \\
0 & 0 & 0 & p_{44} & p_{41} & 0 \\
\end{pmatrix}
= \begin{pmatrix}
0.03 & 0.16 & 0.14 & 0.06 & 0 & 0 \\
0.16 & 0.03 & 0.14 & -0.06 & 0 & 0 \\
-0.15 & -0.15 & -0.15 & 0 & 0 & 0 \\
-0.05 & 0.05 & 0 & -0.1 & 0 & 0 \\
0 & 0 & 0 & -0.1 & -0.05 & 0 \\
0 & 0 & 0 & 0 & 0.06 & -0.07 \\
\end{pmatrix}
\]

**Discussion**

Basing upon the photoelastic coefficients determined above, together with the ultrasonic velocities, crystal density and the refractive indices, one can estimate the AO FM for different geometries of AO interaction, under the circumstances that the incident light and the acoustic wave propagate along the principal axes of optical indicatrix (see Table 1a,b).

It is interesting to note that the Bragg conditions can be satisfied only in a few cases of anisotropic AO interaction. The maximum value
of AOFM \( (M_2=32.971\times10^{-15}\text{s}^3/\text{kg}) \) refers to the velocities that the minimum value of

Table 1a. AO parameters of the LTB crystals \((\rho=2420\text{kg/m}^3, n_e=1.6084 \text{ and } n_o=1.5516)\).

| Acoustic wave V, m/s | Propagation direction, Polarization | p | \( |p_{\text{eff}}| \) | n | Light Direction | Polarization | \( M_2,10^{-15},\text{s}^3/\text{kg} \) or possibility for matching the Bragg conditions |
|----------------------|-------------------------------------|---|----------------|---|----------------|-------------|-------------------|
| 7358 [100], [001]   | p_{11} 0.32 n_e [010] [100]       |   |                |   |                |             | not               |
|                     | p_{31} 0.24 n_o [001]             |   |                |   |                |             | not               |
|                     | p_{11} 0.32 n_o [001]             |   |                |   |                |             | not               |
|                     | p_{12} 0.04 n_o [010]             |   |                |   |                |             | not               |
| 7460 [010], [010]   | p_{11} 0.32 n_e [100] [010]       |   |                |   |                |             | not               |
|                     | p_{31} 0.24 n_o [001]             |   |                |   |                |             | not               |
|                     | p_{12} 0.04 n_o [010]             |   |                |   |                |             | not               |
|                     | p_{11} 0.32 n_o [010]             |   |                |   |                |             | not               |
| 5036 [001], [001]   | p_{13} 0.06 n_e [100] [010]       |   |                |   |                |             | not               |
|                     | p_{33} 0.02 n_o [001]             |   |                |   |                |             | not               |
|                     | p_{13} 0.06 n_o [100] [010]       |   |                |   |                |             | not               |
|                     | p_{33} 0.02 n_o [100] [010]       |   |                |   |                |             | not               |
| 4448 [100], [010]   | p_{66} 0.06 n_e [001] [100], [010]|   |                |   |                |             | not               |
| 4769 [100], [001]   | p_{44} 0.05 n_e [100] [001]       |   |                |   |                |             | 0.134             |
| 4610 [010], [001]   | p_{44} 0.05 n_e [010] [001]       |   |                |   |                |             | 0.149             |

ABO crystals and corresponds to the case of interaction with the slowest acoustic wave having the direction of propagation [100] and that of polarization [001] (see Figure 1).

On the other hand, it follows from the calculated indicative surfaces of ultrasonic velocities that the minimum value of \( v=933.5\text{m/s} \) for the ABO crystals corresponds to the acoustic wave with the \( k \)-vector lying in (011) plane and making the angle 18° with respect to \( z \) axis and the projections of the unit displacement vector \( X_x=0.457, X_y=-0.899 \) and \( X_z=0 \) (see Figure 2a). The LTB crystals exhibit the two such directions of propagation for the slowest waves, with the same velocities \((v=3173\text{m/s})\), i.e. we have the cases:

1) \( k \)-vector lies in (011) plane at the angle of 39° with respect to \( z \) axis; the projections of the unit displacement vector are \( X_x=0.794, X_y=-0.606 \) and \( X_z=0 \) (Figure 2b);

2) \( k \)-vector lies in (101) plane at the angle of 54° with respect to \( z \) axis; the projections of the unit displacement vector are \( X_x=0.79, X_y=0 \) and \( X_z=-0.61 \) (Figure 2b).

Let us evaluate the AOFM for the both cases mentioned above. For this aim one should derive the expression for the effective

**Fig. 1.** Diagram of AO interaction in the ABO crystals for the case of propagation of acoustic wave and the incident optical wave along one of the principal axis of optical indicatrix (in this particular case the value \( M_2=32.971\times10^{-15}\text{s}^3/\text{kg} \) can be achieved).
photoelastic coefficient $p_{\text{eff}}$. Let us, for instance, consider the ABO crystals and the incident optical wave propagated along the [010] direction with $E_3$ polarization. The acoustic wave (according to the Bragg condition, the acoustic frequency should be equal to $f_a=3\times10^9$ Hz) is propagated in the (011) plane at the angle of $18^\circ$ with respect to $z$ axis and the projections of the unit displacement vector are as follows: $X_z=0.457$, $X_y=-0.899$ and $X_x=0$. Then the optical indicatrix equation may be written as

$$
\begin{align*}
2 & (B_1 + p_{12}e_2 + p_{13}e_3 + p_{14}e_4)x^2 + \\
& + (B_2 + p_{31}e_2 + p_{32}e_3 - p_{34}e_4)y^2 + \\
& + 2(p_{44}e_4 - p_{41}e_2)yz = 1,
\end{align*}
$$

where $B_i$ are the optical impermeability constants and $e_j$ the strains induced by the

| Acoustic wave | p | $|p_{\text{eff}}|$ | n | Light Direction | Polarization | $M_2$, $10^{-15}$ s$^3$/kg or possibility for matching the Bragg conditions |
|---------------|---|-----------------|---|----------------|--------------|-------------------------|
| 5649          | [100], [100] | p_{11} 0.03 n_o [010] [100] not |
|               |               | p_{11} 0.15 n_o [010] [001] not |
|               |               | p_{21} 0.16 n_o [010] [010] not |
|               |               | p_{11} 0.03 n_o [010] [100] not |
| 5437          | [010], [010] | p_{11} 0.03 n_o [100] [010] not |
|               |               | p_{42}=-p_{41} 0.05 n_o [010] [010] 0.075 |
|               |               | p_{12} 0.16 n_o [001] [010] not |
|               |               | p_{22}=-p_{11} 0.03 n_o [010] [010] not |
| 3221          | [001], [001] | p_{33}=-p_{13} 0.14 n_o [100] [010] not |
|               |               | p_{13} 0.16 n_o [100] [001] not |
|               |               | p_{13} 0.16 n_o [010] [100] not |
|               |               | p_{13} 0.16 n_o [001] [001] not |
| 2959          | [100], [010] | p_{16}=-p_{41} 0.05 n_o [010] [010], [100] 0.468 |
|               |               | p_{11}=-p_{36} 0.05 n_o [100], [001] 0.277 |
|               |               | p_{66} 0.07 n_o [100], [010] not |
|               |               | p_{66} 0.07 n_o [001], [100] not |
| 1186          | [100], [001] | p_{55}=-p_{44} 0.1 n_o [010] [001], [100] 32.971 |
|               |               | p_{55}=-p_{44} 0.1 n_o [100], [001] 19.555 |
|               |               | p_{65}=-p_{14} 0.06 n_o [001], [100] not |
|               |               | p_{65}=-p_{14} 0.06 n_o [100], [010] not |
| 1230          | [010], [001] | p_{23}=-p_{14} 0.06 n_o [100], [010] not |
|               |               | p_{44} 0.1 n_o [010], [010] 29.557 |
|               |               | p_{44} 0.1 n_o [100], [001] 17.530 |
|               |               | p_{14} 0.06 n_o [001], [100] not |
|               |               | p_{14} 0.06 n_o [100], [010] not |
| 2942          | [010], [100] | p_{66} 0.07 n_o [001], [100] not |
|               |               | p_{66} 0.07 n_o [100], [010] not |
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acoustic wave. After rewriting Eq. (1) in the proper coordinate system of crystal, we obtain

$$
(B_1 + p_{11}e_2 + p_{13}e_3 + p_{14}e_4) X^2 +
$$

$$
\left(\frac{p_{41}e_4 - p_{43}e_3}{(B_1 - B_3 + (p_{11} - p_{13})e_2 + (p_{11} - p_{13})e_3 - p_{14}e_4)}\right) Y^2 +
$$

$$
\left(\frac{p_{42}e_4 - p_{43}e_3}{(B_1 - B_3 + (p_{11} - p_{13})e_2 + (p_{11} - p_{13})e_3 - p_{14}e_4)}\right) Z^2 = 1
$$

(2)

The change in the refractive index $n_3$ is given by

$$
\Delta n_3 = \frac{1}{2} n_3^3 \left\{ p_{31}e_2 + p_{33}e_3 - \frac{(p_{41}e_4 - p_{43}e_3)^2}{\left(\frac{1}{n_1^2} - \frac{1}{n_3^2} + (p_{11} - p_{33})e_2 + (p_{11} - p_{33})e_3 - p_{14}e_4\right)}\right\},
$$

(3)

as well as by the relations

$$
p_{31}e_2 + p_{33}e_3 \gg \frac{(p_{41}e_4 - p_{43}e_3)^2}{2 \left(\frac{1}{n_1^2} - \frac{1}{n_3^2} + (p_{11} - p_{33})e_2 + (p_{11} - p_{33})e_3 - p_{14}e_4\right)}.
$$

(4)

Eq. (3) may be simplified to the form

$$
\Delta n_3 \approx 0.5 \times n_3^3 \left\{ p_{31}e_2 + p_{33}e_3 \right\} .
$$

(5)

After considering the orientation of the displacement vector of acoustic wave, Eq. (4) becomes

$$
\Delta n_3 \approx 0.5 \times n_3^3 \left\{ -0.899 p_{31} + 0.457 p_{33} \right\} e .
$$

(6)

Taking the values $p_{31}=-0.15$, $p_{13}=0.14$ and the relation $p_{ef} = \{-0.899 p_{31} + 0.457 p_{33}\}$ into account, one can arrive at $p_{ef}=0.19$ and $M_f=150.32 \times 10^{-15} \text{s}^3/\text{kg}$. It can be seen that in case of the ultrasonic velocity achieving its lowest value (933.5 m/s; to be compared with the value 1186 m/s), the AOFM would increase drastically up to $150.32 \times 10^{-15} \text{s}^3/\text{kg}$ (cf. with the previous value $32.971 \times 10^{-15} \text{s}^3/\text{kg}$). If we change the direction of the incident optical beam from the angle $\alpha=18^\circ$ to $\alpha=180^\circ$ with respect to the $z$ axis (see Figure 3a) and change additionally the frequency of the acoustic wave from $f_a=135 \times 10^6 \text{Hz}$ (the collinear diffraction) to $f_a=29 \times 10^6 \text{Hz}$, the AOFM would also change

from $M_f=240.7 \times 10^{-15} \text{s}^3/\text{kg}$ through the value $M_f=150.32 \times 10^{-15} \text{s}^3/\text{kg}$ (for $k_i$ parallel to the $y$ axis) up to $M_f=243.4 \times 10^{-15} \text{s}^3/\text{kg}$. This effect is only owing to anisotropy of the refractive index $n_e$.

Let us now analyze the case of LTB crystals. When the incident optical wave is propagated along the [010] direction with $E_3$ polarization, while the acoustic wave (according to the Bragg condition, the acoustic frequency should be equal to $f_a=2.8 \times 10^9 \text{Hz}$) is propagated in the (011) plane at the angle of $39^\circ$ with respect to the $z$ axis (the relevant projections of the unit displacement vectors being $X_z=0.794$, $X_y=-0.606$ and $X_x=0$), the optical indicatrix equation may be written as

$$
(B_1 + p_{12}e_2 + p_{13}e_3) X^2 +
$$

$$+(B_1 + p_{14}e_2 + p_{13}e_3) Y^2 +

(B_3 + p_{31}e_2 + p_{33}e_3) Z^2 + 2 p_{44}e_4 y z = 1
$$

(6)

Rewriting Eq. (6) in the proper coordinate system of the crystal, we get
Fig. 2. Indicative surfaces of reciprocal acoustic wave velocities in the ABO (a) and LTB (b) crystals.

\[
\begin{align*}
[B_i + p_{i2}e_2 + p_{i3}e_3]X^2 + (p_{44}e_4)^2 + 
(B_i - B_3 + (p_{11} - p_{33})e_2 + (p_{13} - p_{33})e_3)]Y^2 + 
[B_3 + p_{31}e_2 + p_{33}e_3 - (p_{44}e_4)^2 + 
(B_i - B_3 + (p_{11} - p_{33})e_2 + (p_{13} - p_{33})e_3)]Z^2 = 1
\end{align*}
\]

(7)

Then the change in the refractive index \( n_3 \) of the acousto-optic effect reduces to

\[
\Delta n_3 \approx \frac{1}{2} n_3^3 \left[ p_{31}e_2 + p_{33}e_3 \right].
\]

(8)

With accounting for the orientation of the displacement vector of acoustic wave, Eq. (8) yields in

\[
\Delta n_3 \approx \frac{1}{2} n_3^3 \left[ -0.606 p_{31} + 0.794 p_{13} \right] e. \quad (9)
\]
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Since $p_{eff} = \{0.606 p_{31} + 0.794 p_{13}\}$ and $p_{31} = -0.24$, $p_{13} = 0.06$, we obtain $p_{eff} = 0.1$ and $M_2 = 2.07 \times 10^{-15}$ s$^3$/kg. If the ultrasonic velocity achieve its lowest value (3173 m/s, not 4610 m/s), the AO effect would increase by more than order of magnitude (2.07 $\times$ $10^{-15}$ s$^3$/kg; to be compared with the previous value 0.149 $\times$ $10^{-15}$ s$^3$/kg). Again, in case of changing direction of the incident optical beam from the angle $\alpha = 39^\circ$ to $\alpha = 180^\circ$ with respect to the z axis (see Figure 3b), as well as simultaneously changing the frequency of the acoustic wave from $f_a = 732 \times 10^6$ Hz (the collinear diffraction) to $f_a = 77.7 \times 10^9$ Hz, the AO effect would evolve from $M_2 = 2.35 \times 10^{-15}$ s$^3$/kg through $M_2 = 2.07 \times 10^{-15}$ s$^3$/kg (for $k_i$ parallel to the y axis) up to $M_2 = 2.57 \times 10^{-15}$ s$^3$/kg. Quite similar to the ABO crystals, this is owing to anisotropy of $n_e$.

The same value of the AO effect may be obtained when considering the second case of AO interaction in the LTB crystals. However, it is quite possible that the orientation of the acoustic wave vector intermediate between (011) and (101) planes could provide a less value of the ultrasonic wave velocity, when compare with the mentioned planes.

**Conclusion**

In conclusion, one can notice that the ABO and LTB borate crystals manifest a high AO effect. The value $M_2 = 243.4 \times 10^{-15}$ s$^3$/kg for the ABO crystals is comparable, in the order of magnitude, with those typical for good AO materials such as TeO$_2$, for example. It is evident from the presented results that the most important criterion for the choice of crystals with a high value of AO parameter $M_2$ is the velocity of the
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acoustic wave. Moreover, we have shown that the propagation direction of the slowest acoustic wave does not necessarily coincide with the principal axes of the optical indicatrix ellipsoid. While changing the propagation direction of the acoustic wave (e.g., in the $xy$-plane), we have to take changing orientation of the displacement vector into consideration, the latter leading to the corresponding changes in the $p_{eff}$ parameter.

Acknowledgement

We would like to acknowledge the financial support of this study from the Scientific and Technology Centre of Ukraine under the Project N1712.

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