

Ceramics International 30 (2004) 1675-1677



www.elsevier.com/locate/ceramint

Elastic and elastooptic properties of LiB₃O₅

I.I. Zubrinov^a, V.K. Sapozhnikov^a, N.A. Pylneva^b, V.V. Atuchin^{a,*}

^a Laboratory of Optical Materials and Structures, Institute of Semiconductor Physics, Novosibirsk 630090, Russia
 ^b Laboratory of Crystal Growth, Institute of Mineralogy and Petrography, Novosibirsk 630090, Russia

Received 8 December 2003; received in revised form 16 December 2003; accepted 22 December 2003

Available online 6 May 2004

Abstract

 LiB_3O_5 (LBO) crystals are of great importance for nonlinear optical applications. In this study a set of elastic coefficients has been determined for LBO by the interference acoustooptical method and a set of elastooptic coefficients has been measured by Dixon's method. © 2004 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

Keywords: LiB₃O₅ (LBO) crystal; Elastic coefficients; Elastooptic coefficients

1. Introduction

Lithium triborate (LiB₃O₅ or LBO) is one from the basic crystal materials used for laser beam control in visible and near IR ranges [1-3]. LBO crystals, Pn2₁a point group [4] and a = 8.441, b = 7.379 and c = 5.138 Å, are characterized by such valuable properties as appropriate nonlinear optical properties in combination with high optical damage threshold [2,5], electrooptic and piezoelectric effects [6,7], and relative surface chemical stability [8,9]. Recently a method of LBO crystal growth with using molybdate flux has been proposed [10]. The optimization of the growth conditions yields large size, \sim 500 g, LBO crystals with very low defect content [11,12]. As a practical matter, it is desirable to have as more complete set of physical and chemical characteristics of low defect LBO as possible. In the present study the elastic and elastooptic properties of LBO grown by top seeded solution growth (TSSG) method from molybdate flux have been evaluated.

2. Experiment

The crystal studied was grown by TSSG method without pulling and rotating. Detailed description of charge preparation and growth conditions has been presented in [13]. Paral-

lelepiped sample with the dimensions $10 \text{ mm} \times 8 \text{ mm} \times 6 \text{ mm}$ was cut and polished perpendicular to the three principal crystallographic axes.

In accordance with C_{2v} symmetry nine independent nonzero elastic constants and 12 independent nonzero elastooptic constants characterize LBO. Elastic constants were calculated with using experimental data on phase velocities of bulk elastic waves in LBO sample. Ultrasonic wave velocities have been measured with original pulse interference acoustooptic method with very high-resolution [14]. The structure of acousto-optical cell is shown in Fig. 1. In this configuration elastic waves are excited by piezoelectric transducer positioned on buffer. The polarization of elastic wave is defined by the transducer used. When a single high frequency electric pulse with duration τ is fed on the transducer, a set of ultrasound pulses of the same duration and frequency is exited in the system transducer-buffer-sample due to consecutive reflections at the interfaces. So, in buffer the following pulses will be observed: U₁-directly from transducer, U2-reflected from the interface buffer-sample, U₃-doubly passed through the sample (passed into sample, then reflected from external face of the sample and returned to buffer), and so on. The high frequency filling of all three pulses in buffer is of the same frequency, wavelength and polarization. The pulse U₃ keeps the information on velocity of ultrasonic wave in the sample and U₂ pulse is a reference. With τ increasing, the pulses overlap and the interference is observed. In this case, high frequency variation will induce the variation of the magnitude (beating) of sum pulse in the range from minimum $A_{23} = A_3 - A_2$ to maximum

^{*} Corresponding author. Tel.: +7-3832-343889; fax: +7-3832-332771. *E-mail address:* atuchin@thermo.isp.nsc.ru (V.V. Atuchin).

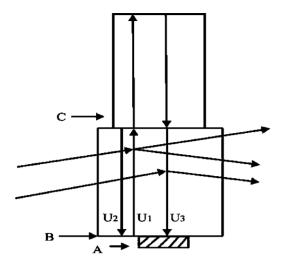


Fig. 1. Acoustooptic cell for elastic constants determination: (A) transducer; (B) buffer and (C) LBO sample.

 $A_{32} = A_3 + A_2$. Here A_i is a magnitude of ultrasonic pulse U_i . Respectively, the beating of intensities appeared for the laser beams I_2 , I_3 diffracted on the ultrasonic pulses. Let us suppose that at any frequency f_1 the maximum of intensity of diffracted laser beam is observed under the condition

$$2\pi f_1\left(\frac{2L}{V}\right) + a_1 + a_2 = 2\pi n,\tag{1}$$

where a_1 and a_2 are phase shifts by reflections, L-sample thickness in the direction of ultrasonic wave and V-ultrasonic wave velocity. Then, at frequency f_2 some maximum will be detected with the number m=n+N under the phase resonance condition

$$2\pi f_2\left(\frac{2L}{V}\right) + a_1 + a_2 = 2\pi m. \tag{2}$$

So, on frequency variation $\Delta f = f_2 - f_1$ we will see N = m - n maximums defined by the condition

$$N = \Delta f\left(\frac{2L}{V}\right). \tag{3}$$

Now, if to measure the value Δf and maximum number N, the ultrasonic wave velocity is defined by simple Eq. $V = 2L \Delta f/N$.

Elastooptic constants were determined by Dixon's method [15]. The structure of the acoustooptic cell used is shown in Fig. 2. In this cell LBO sample and reference sample (silica) are linked in tandem by special bonding layer. Piezoelectric transducer is positioned on external face of reference sample. In this configuration ultrasonic pulse generated by transducer travels through the reference sample and LBO sample, is reflected by free face of LBO sample and returns to reference sample. The ultrasonic wave is probed by laser beam under Bragg's angle. The intensity of diffracted beam is defined by the relation

$$I_1 = I_0 \sin^2\left(\frac{V}{2}\right),\tag{4}$$

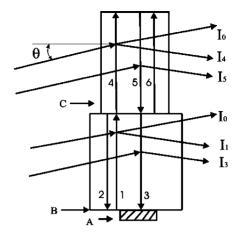


Fig. 2. The cell for elastooptic coefficients measurement: (A) transducer; (B) reference sample (silica) and (C) LBO sample.

where I_0 is an intensity of light transmitted through the sample and V is governed by the equation

$$V = \frac{\pi L (2M_2 I_a)^{1/2}}{\lambda \cos \theta}.$$
 (5)

where L is a length of acoustooptic interaction, λ the light wavelength in vacuum, θ an angle between light beam in crystal and wavefront of ultrasonic wave, I_a the ultrasonic wave intensity and

$$M_2 = \frac{n^6 p^2}{\rho V^3},\tag{6}$$

where M_2 is a coefficient of acoustooptical figure of merit, n the refractive index of the crystal, ρ the density, V the an ultrasonic wave velocity and p the elastooptic constant. If the sequence of transmittance of ultrasonic waves is designated by indices i, as it is illustrated in Fig. 2, and I_i is an intensity of the optical beam diffracted by ultrasonic pulse with number i, then M_2 for the sample may be resulted as

$$\frac{M_2}{M_2^*} = \left(\frac{I_4 I_5}{I_1 I_3}\right)^{1/2},\tag{7}$$

where M_2^* the coefficient of acoustooptical figure of merit of silica reference sample.

The density of our LBO crystal, $\rho = 2.4789 \text{ g/cm}^3$, necessary for calculations, was measured by hydrostatic method.

3. Results and discussion

The values of ultrasonic wave velocities measured in LBO are presented in Table 1. It is reasonable to be noted that all velocities were determined for the same LBO sample and possible error is not above 5 m/s. If to compare the absolute values of velocities of ultrasonic waves in LBO with those for some recently studied materials, the highest value detected in LBO exceed that in KTiOPO₄ (7680 m/c) [16], and is substantially below those found for another borate,

Table 1 Ultrasonic wave velocities in LBO

Propagation direction	Type	Polarization	Velocity (m/c)
[100]	L	[100]	6338
	S	[0 1 0]	4050
	S	[001]	5846
[0 1 0]	L	[010]	8612
	S	[001]	5319
	S	[100]	4024
[001]	L	[001]	6940
	S	[010]	5324
	S	[100]	5737

Table 2 Elastic constants of LBO (10¹⁰ N/m²)

$\overline{\mathrm{C}_{ij}}$	C ₁₁ E	C_{22}^{E}	C ₃₃ E	C ₃₃ ^D	C ₄₄ ^E	C ₅₅ ^E	C ₆₆ E
This work Brillouin [6] Ultrasonic [6]	10.92		10.32 9.04	11.94	7.03 7.10 6.46	8.16 8.66 8.22	3.60

Table 3 Elastooptic constants of LBO

p ₁₁	0.2
p ₂₂	0.2
p ₃₃	0.3
p ₁₂	0.08
p ₂₃	0.5
p ₃₁	0.05
p ₁₃	0.7
p_{21}	0.1
p ₃₂	0.26

 CuB_2O_4 (9917 m/c) [17], and $BeAl_6O_{10}$ (12,000 m/c) [18]. For the lowest velocity value the distribution of the materials is the same. The elastic constants of LBO in comparison with earlier reported results are given in Table 2. In all cases the constants obtained in present study is few beyond those measured in reference [6] at ultrasonic frequencies. Supposedly this shift to higher values is appeared due to progress in LBO crystal quality. Elastooptic constants of LBO are listed in Table 3. The possible error in determination of p_{ij} is defined by the intensity of the lowest I_i signal and is not above 20%. The coefficients p₄₄, p₅₅ and p₆₆ were not evaluated because the constants are manifested in light diffraction on shear ultrasonic waves with change of lightwave polarization. The observation of this diffraction in LBO is possible only at frequencies above \sim 1 GHz but strong attenuation of ultrasonic waves at the conditions greatly complicates the experiment. As a whole, acoustooptic figure of merit of LBO is in close relation to that of silica.

4. Conclusions

A set of elastic and elastooptic constants of LBO has been determined at room conditions. LBO possesses middle ultra-

sonic wave velocities for the longitudinal and shear modes and large electromechanical coupling coefficients 28–52% [6]. So, in combination with other physical properties, high optical damage threshold in particular, the crystals may be actively used in acoustoelectronics and acoustooptics.

References

- S. Zhao, C. Huang, H. Zhang, Crystal growth and properties of lithium triborate, J. Cryst. Growth 99 (1990) 805–810.
- [2] D.N. Nikogosyan, Lithium triborate (LBO), a review of its properties and applications, Appl. Phys. A58 (1994) 181–190.
- [3] J. Jiang, T. Hasama, High repetition-rate femtosecond optical parametric oscillator based on LiB₃O₅, Opt. Commun. 211 (2002) 295–302.
- [4] H. König, R. Hoppe, Zur Kenntnis von LiB₃O₅, Anorg. Allg. Chem. 439 (1978) 71–79.
- [5] Y. Furukawa, S.A. Markgraf, M. Sato, H. Yoshida, T. Sasaki, H. Fujita, T. Yamanaka, S. Nakai, Investigation of the laser damage of lithium triborate, LiB₃O₅, single crystals, Appl. Phys. Lett. 65 (12) (1994) 1480–1482.
- [6] Y. Wang, Y.J. Jiang, Y.L. Liu, F.Y. Cai, L.Z. Zeng, The elastic and piezoelectric properties of a lithium triborate single crystal, Appl. Phys. Lett. 67 (17) (1995) 2462–2464.
- [7] V.D. Antsygin, V.A. Gusev, A.M. Yurkin, Electrooptical properties of barium and lithium borates, Avtometriya 3 (1996) 16–21
- [8] V.V. Atuchin, V.G. Kesler, I.A. Lisova, L.D. Pokrovsky, N.A. Pylneva, A.M. Yurkin, Stabilization of LiB₃O₅ crystal surfaces, Proc. SPIE 4513 (2001) 107–113.
- [9] V.V. Atuchin, L.D. Pokrovsky, V.G. Kesler, L.I. Isaenko, L.I. Gubenko, Structure and chemistry of LiB₃O₅ (LBO) optical surfaces, J. Ceram. Proc. Res. 4 (2) (2003) 84–87.
- [10] V.I. Kosyakov, N.A. Pylneva, Z.G. Bazarova, A.M. Yurkin, Topology of liquidus surface in B₂O₃–Li₂O·B₂O₃–Li₂O·MoO₃–MoO₃ system: implications to the growth of lithium triborate single crystals, Mater. Res. Bull. 36 (2001) 573–584.
- [11] N. Pylneva, V. Kosyakov, A. Yurkin, G. Bazarova, V. Atuchin, A. Kolesnikov, E. Trukhanov, C. Ziling, Real structure of LiB₃O₅ (LBO) crystals grown in Li₂O-B₂O₃-MoO₃ system, Cryst. Res. Technol. 36 (12) (2001) 1377–1384.
- [12] A.P. Vasilenko, V.V. Kolesnikov, E.M. Trukhanov, N.A. Pylneva, A.M. Yurkin, V.V. Atuchin, X-ray topography study of LiB₃O₅ crystals grown from molybdate flux, J. Phys.: Condens. Matter 15 (40) (2003) 6801–6808.
- [13] N.A. Pylneva, N.G. Kononova, A.M. Yurkin, G.G. Bazarova, V.I. Danilov, Growth and nonlinear optical properties of lithium triborate crystals, J. Cryst. Growth 198/199 (1999) 546–550.
- [14] S.V. Bogdanov, I.I. Zubrinov, E.V. Pestryakov, V.K. Sapozhnikov, Interference acoustooptic technique for sound velocity measurements, Acoust. Phys. 46 (1) (2000) 27–32.
- [15] R.W. Dixon, M.G. Cohen, A new technique for measuring magnitudes of photoelastic tensors and its application to lithium niobate, Appl. Phys. Lett. 8 (8) (1966) 205–206.
- [16] I.I. Zubrinov, V.K. Sapozhnikov, E.V. Pestryakov, V.V. Atuchin, Elastic and elasto-optic properties of KTiOPO₄, Proc. SPIE 5129 (2003) 249–254.
- [17] K.S. Alexandrov, B.P. Sorokin, D.A. Glushkov, L.N. Bezmaternykh, S.I. Burkov, S.V. Belushenko, Electromechanical properties and anisotropy of acoustic wave propagation in CuB₂O₄ copper metaborate, Phys. Solid State 45 (1) (2003) 41–45.
- [18] I.I. Zubrinov, V.I. Semenov, E.V. Pestryakov, V.V. Petrov, A.I. Alimpiev, Elastic and elastooptical properties of beryllium hexaaluminate (BeAl₆O₁₀) crystal, Cryst. Rep. 43 (4) (1998) 657–660.