

Measurements of small optical absorption and scattering coefficients of high-power polarised laser radiation in lithium triborate crystals

I.V. Grishchenko, A.K. Vorob'ev, A.Yu. Ostapiv, Yu.S. Stirmanov, A.V. Konyashkin, O.A. Ryabushkin

Abstract. We present the results of measurements of low optical absorption and scattering coefficients in lithium triborate crystals using the method of piezoelectric resonance laser calorimetry under the action of polarised single-mode laser radiation at wavelengths of 1070 and 532 nm in the power ranges of 50–300 and 5–50 W, respectively. It is shown that when radiation propagates along the crystal axis a , these coefficients remain constant in the specified power ranges and do not depend on the direction of laser radiation polarisation. The measured optical scattering coefficients are compared with the results obtained by the method of integrating spheres.

Keywords: lithium triborate, laser calorimetry, piezoelectric resonance laser calorimetry, optical absorption, optical scattering, equivalent temperature, ionic conductivity, polarised IR radiation, visible radiation.

1. Introduction

To date, direct generation of laser radiation can be obtained in narrow wavelength ranges. Generation of laser radiation at new wavelengths is possible due to nonlinear processes of frequency conversion of laser radiation in nonlinear-optical crystals. For the first time, the generation of the second harmonic of laser radiation was experimentally demonstrated in 1961 in a quartz crystal [1]. The generation efficiency was extremely low due to nonfulfilment of the phase matching conditions [2] and small value of the coefficient of nonlinear susceptibility of quartz. Since then, a range of materials used for nonlinear optical conversion has increased significantly and continues to grow to this day. At the dawn of nonlinear optics, the absorption coefficients of the employed crystals were quite high. For example, potassium dihydrogen phosphate crystals (KH_2PO_4 , KDP) had absorption coefficients higher than 10^{-2} cm^{-1} at a wavelength of 1064 nm [3], which

was considered to be quite good quality at that time. The continuous development and improvement of methods for growing and preparing optical elements for applied problems of laser physics in subsequent decades made it possible to improve considerably the optical quality of nonlinear optical materials. For example, nonlinear optical crystals of lithium triborate (LiB_3O_5 , LBO), which are the object of research in this work, have optical absorption coefficients of the order of 10^{-5} – 10^{-4} cm^{-1} in the visible and near-IR ranges [4]. Along with a high optical damage threshold of these crystals, this is one of the reasons why they are currently widely used for generation of high-intensity radiation. Using LBO crystals as converters, output powers of 700, 160, and 3.3 W were obtained at the wavelengths of the second (532 nm), third (355 nm), and fourth (266 nm) harmonics of ytterbium laser radiation with a conversion efficiency of 70%, 30% and 14%, respectively [5, 6].

In the case of nonlinear conversion of high-intensity radiation, the smallness of the optical absorption coefficients is critical, since due to the nonuniform heating of the crystals conditioned by the absorption of laser radiation at the main and converted wavelengths, a violation of the phase-matching conditions is possible, which, as a consequence, entails a decrease in the conversion efficiency. Along with the laser radiation absorption, scattering is an additional mechanism of losses in nonlinear optical crystals [7]. It is important to note that at a fixed wavelength, the scattering coefficient value may exceed the absorption coefficient value in the material [8] due to the optical inhomogeneity of the sample.

To diagnose the quality of optical materials, in particular, to measure low optical absorption coefficients at a level of 10^{-6} – 10^{-4} cm^{-1} , various precision techniques have been developed to date [9]. The most conventional methods used for these purposes are laser calorimetry [10], photoacoustics [11], photothermal deflection [12], and photothermal common-path interferometry (PCI) [13].

The aim of this work was to measure for an LBO crystal the dependences of the coefficients of optical absorption and scattering of laser radiation at wavelengths of 1070 and 532 nm on its power and polarisation direction. For this purpose, the method of piezoelectric resonance laser calorimetry (PRLC) [14–16] was used, which is an improved modification of the laser calorimetry method.

As a chemical compound, lithium triborate was first synthesised in 1926 [17], and the possibility of its application in nonlinear optics was first demonstrated in the late 1980s [18]. LBO crystals are biaxial and, along with low optical absorption, exhibit anisotropy of the coefficients of thermal expansion, thermal conductivity and refractive index [19]. It should be noted that ionic conductivity is observed in LBO crystals,

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which is characterised by significant spatial anisotropy [20]. The conductivity value σ depends on the frequency of the applied external electric field and increases with crystal temperature. The σ value along the crystallographic axes a and b of the LBO crystal is $3 \times 10^{-7} (\Omega \text{ cm})^{-1}$, and along the c axis it is $10^{-6} (\Omega \text{ cm})^{-1}$ at a temperature of 373 K and an external electric field frequency of 50 kHz [21]. The influence of ionic conductivity on the Q -factor of piezoelectric resonances of LBO crystals was discovered earlier [22]. In samples with suppressed ionic conductivity, the Q -factor of the resonances virtually did not change upon heating. In samples where the ionic conductivity was not suppressed, an exponential broadening of the resonance line form was observed during heating until their complete disappearance. Presumably, this phenomenon was associated with an increase in σ with temperature. Additionally, the authors of [22] noted that under conditions of interaction with laser radiation the service life of LBO crystals with suppressed ionic conductivity was higher. Thus, when a crystal was exposed to pulsed laser radiation at a wavelength of 266 nm (pulse duration 5 ns, repetition rate 100 kHz, pulse energy 1.2 μJ), the time of degradation initiation for crystals with suppressed ionic conductivity was twice as long as the same characteristic time for crystals with unsuppressed ionic conductivity.

As mentioned above, the use of LBO crystals to generate harmonics of high-power laser radiation is due, in particular, to the high optical damage threshold, the value of which depends on the directions of propagation and polarisation of laser radiation with respect to the crystallographic axes. It has been shown that when radiation propagates along the crystal axis a at a wavelength of 1064 nm (pulse duration 6 ns, repetition rate 10 Hz, waist diameter 25 μm), being polarised along the c axis, the threshold energy density of optical damage in the case of single-pulse action (the 1-on-1 method [23]) is 3.2 times higher than in the case of polarisation along the b axis [24]. The authors of [24] suggested that such difference was due to the influence of the anisotropy of ionic conductivity on the optical destruction threshold.

In this regard, the study of the dependence of optical absorption and scattering coefficients for different directions of laser radiation polarisation with respect to the LBO crystallographic axes is an urgent task. Earlier in [25], the results of measuring the dependence of the optical absorption coefficient in an LBO crystal on the intensity of unpolarised laser radiation (wavelength 1070 nm, radiation intensity $I = 10\text{--}100 \text{ MW cm}^{-2}$) were presented. In this paper, we present the results of the PRLC measurements of the dependences of the optical absorption coefficient α and scattering coefficient μ in LBO crystals under the action of cw polarised single-mode laser radiation at $\lambda_1 = 1070 \text{ nm}$ with a power of 50–300 W (waist diameter 30 μm) and pulsed polarised single-mode laser radiation at $\lambda_2 = 532 \text{ nm}$ (pulse duration 1.3 ns, repetition rate 1 MHz, maximum pulse energy 50 μJ) with an average power of 5–50 W (waist diameter 6 μm) for the direction of laser radiation polarisation along the crystallographic axes b and c of the LBO crystal (radiation propagated along the a axis). The measured values of the scattering coefficients are compared with the results obtained by the method of integrating spheres.

2. Theoretical part

Laser calorimetry is an international standardised method for measuring low optical absorption coefficients of dielectrics

[10]. It is based on the search for a correspondence between the experimentally measured kinetics of the sample temperature when heated by laser radiation and the solution of the nonstationary heat conduction equation

$$\rho c_{\text{sp}} \frac{\partial T(x, y, z, t)}{\partial t} = \nabla(\kappa \nabla T(x, y, z, t)) + q, \quad (1)$$

where ρ is the density; c_{sp} is the specific heat capacity; κ is the thermal conductivity coefficient; $T(x, y, z, t)$ is the temperature distribution; and q is the volumetric density of power sources and heat losses in the sample.

To determine the low coefficients of optical absorption from the solution to equation (1), boundary conditions of the third kind are used (the Robin problem or the Newton–Rikhman law). In practice, a simplification is most often used, which consists in assuming uniform heating of the sample by laser radiation [26]. In this case, the temperature change T of the sample appears as

$$\Delta T(t) = \begin{cases} 0, & t \leq t_1, \\ \frac{\alpha P}{\gamma m c_{\text{sp}}} (1 - e^{-\gamma(t-t_1)}), & t_1 < t < t_2, \\ \frac{\alpha P}{\gamma m c_{\text{sp}}} (e^{-\gamma(t-t_2)} - e^{-\gamma(t-t_1)}), & t \geq t_2. \end{cases} \quad (2)$$

Here α is the optical absorption coefficient at a given wavelength; P is the laser radiation power; m is the sample mass; l is the sample length; $\gamma = (h^T S)/(m c_{\text{sp}})$ is the temperature loss coefficient; h^T is the coefficient of heat exchange between the test sample and the surrounding air; S is the sample surface area; and t_1 and t_2 are the moments of the onset and termination of irradiation.

In laser calorimetry, the sample temperature is most often measured using external thermal sensors (thermocouple, thermistor, etc.). With this approach, it is necessary to use a relatively low power of laser radiation (from units to tens of milliwatt) to reduce the additional heating of thermal sensors caused by scattered radiation, which is inevitably present due to the optical inhomogeneity of the samples. However, this range does not correspond to the operating power range at which nonlinear optical crystals are used.

The PRLC method is based on impedance spectroscopy [27], piezoelectric effect [28, 29], and the concept of equivalent temperature [30]. All nonlinear optical crystals have piezoelectric properties. When an external radiofrequency (RF) electric field is applied, mechanical vibrations are excited in the crystal due to the inverse piezoelectric effect. Piezoelectric resonances that can be excited when the RF field frequency coincides with the frequencies of the crystal acoustic eigenmodes are characterised by a high Q -factor. That is why the frequencies can be measured with high accuracy.

The frequencies Rf_n of piezoelectric resonances (n is the number of the crystal eigenmode) are sensitive to the crystal temperature T_c . Under uniform heating, in the first approximation, the temperature dependences of the frequencies of piezoelectric resonances are linear: $Rf_n(T_c) = Rf_n(T_0) + K_n^{\text{prt}}(T_c - T_0)$, where K_n^{prt} is the piezoelectric resonance thermal coefficient of the n th mode. Then the change in the thermodynamic temperature T_c of the crystal heated by laser radiation with a given power P is identified with a change in its equivalent temperature $\Delta\theta_{\text{eq}}$:

$$\Delta T_c(P, t) \equiv \Delta \theta_{\text{eq}}(P, t) = \frac{\Delta R f_n(P, t)}{K_n^{\text{prt}}}. \quad (3)$$

Thus, the kinetics of the sample temperature is measured directly from the kinetics of the resonance frequency when the sample is exposed to laser radiation with power P . The heating of the crystal can be considered uniform, since the temperature distribution nonuniformity due to high thermal conductivity and small values of absorption coefficients is several orders of magnitude lower than the change in crystal temperature caused by the laser radiation absorption.

In practice, in the case of low optical absorption coefficients, their values can be determined from the initial time interval (5–10 s) of the crystal heating kinetics. The advantages of this approach are the short time of the experiment and the absence of significant heat exchange between the crystal and the air. In the initial time interval, given the smallness of the exponent index in expression (2), the exponential term can be expanded into a Taylor series [31]. In this case, the crystal temperature dependence on time will be linear with a slope directly proportional to the optical absorption coefficient:

$$\Delta \theta_{\text{eq}}(P, t) = \frac{\alpha P}{mc_{\text{sp}}} (t - t_1). \quad (4)$$

Then the optical absorption coefficient α is determined by the expression

$$\alpha = \frac{mc_{\text{sp}} k_n}{IP K_n^{\text{prt}}}, \quad (5)$$

where $k_n = dR f_n/dt$ corresponds to the tangent of the slope angle of the initial segment of the frequency kinetics $R f_n$.

To measure the scattering coefficients, a thin layer of an absorbing coating is applied to the side surfaces of the sample under study, thereby inducing additional heating of the sample due to the absorption of scattered radiation. After a coating is deposited, the kinetics of laser heating of the sample is measured, from which the optical loss coefficient β is deter-

mined, which is the sum of the optical absorption coefficient α and scattering coefficient μ .

Integrating spheres with an embedded photodetector are commonly used to measure optical scattering coefficients [32]. Radiation from a laser source is transmitted through small holes in the sphere. For calibration, one of the holes is closed with a thin, highly diffusing material that provides uniform illumination of the inner surface of the sphere. The material is then removed, and the sample is placed in one of three positions (inside the sphere, in front of the inlet hole, or behind the outlet hole) to measure integral scattering, forward, or backward scattering, respectively. The radiation of the laser source is modulated at a certain frequency, at which the signal from the photodetector is measured, proportional to the power of the scattered laser radiation. The value of the scattering coefficient is determined based on the preliminary calibration of the photodetector.

3. Experimental setup

A block diagram of the experimental setup for measuring small coefficients of optical absorption and scattering by the PRLC method is shown in Fig. 1a.

We examined an LBO crystal sample in the form of a rectangular parallelepiped (sizes $3.0 \times 3.3 \times 20.0$ mm corresponding to the axes b , c , and a ; Fig. 1b). The laser radiation polarisation vector e_b is parallel to the b axis (similarly, the vector e_c is parallel to the c axis). AC voltage was supplied from an RF generator to a circuit containing a capacitor in which the LBO crystal under study was placed, and a load resistor R . The amplitude and phase of the voltage on the load resistor were measured using a lock-in amplifier. The capacitor electrodes were thin copper plates (thickness 200 μm), which minimise their heating due to the absorption of scattered radiation.

The sources of radiation were:

1. Ytterbium-doped fibre laser with cw linearly polarised single-mode radiation at a wavelength $\lambda = 1070$ nm, an output power of 50–300 W, a beam quality factor $M^2 = 1.05$, and a waist diameter of 30 μm .

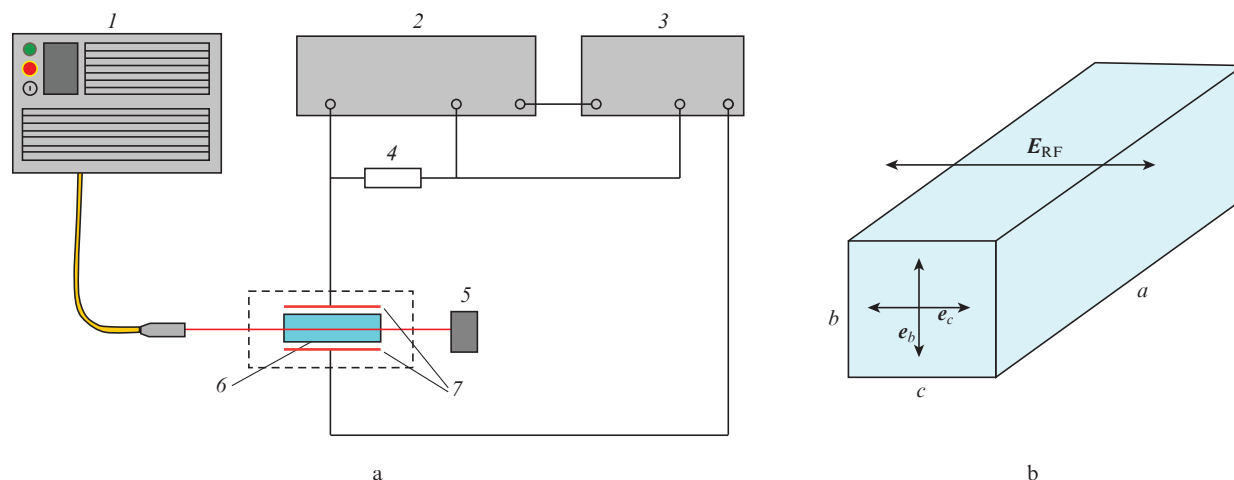


Figure 1. (Colour online) (a) Simplified block diagram of the experimental setup for measuring small optical absorption and scattering coefficients by the PRLC method [(1) laser radiation source at a wavelength of 1070 or 532 nm, (2) lock-in amplifier, (3) RF generator, (4) load resistor R , (5) optical power meter, (6) LBO sample under study placed in a thermostat, (7) capacitor plates]; (b) directions of the electric field vector E_{RF} of the probe RF field and polarisation vectors e_b , e_c of radiation propagating along the a axis.

2. A laser with pulsed linearly polarised single-mode radiation at a wavelength $\lambda = 532$ nm with an average output power of 5–50 W, a pulse duration of 1.3 ns, a repetition rate of 1 MHz, a maximum energy of 50 μ J per pulse, a beam quality factor $M^2 = 1.5$, and a waist diameter of 6 μ m.

4. Measurement of optical absorption and scattering coefficients by the PRLC method

In the experiment the spectral dependence of the response (in amplitude and phase of the voltage at the load resistor R) of the LBO crystal to the action of an external RF field was measured in the RF range from 0.5 to 1.5 MHz. To determine the optical absorption coefficient α by the PRLC method, a resonance at a frequency of $Rf_1 = 1068$ kHz (at a temperature of $T = 25^\circ\text{C}$) was selected.

Temperature calibration of the frequency Rf_1 of the selected piezoelectric resonance was carried out under conditions of uniform heating. A capacitor with the crystal was placed in a thermostat; the sample temperature varied in the range from 25 to 50°C . The piezoelectric resonance thermal coefficient of this mode was determined from the linear approximation of this dependence: $K_1^{\text{prt}} = -(0.39 \pm 0.02)$ kHz $^\circ\text{C}^{-1}$.

Figure 2 shows the dependences of the optical absorption coefficients at wavelengths λ_1 and λ_2 for the laser radiation polarisation along the crystallographic axes b and c on the laser radiation power. For a fixed wavelength, power, and polarisation of laser radiation, the optical absorption coefficient α was determined by formula (5). It follows from Fig. 2 that the optical absorption coefficients of the LBO crystal at the considered wavelengths remain constant for two directions of laser radiation polarisation; the average values are $(2.1 \pm 0.2) \times 10^{-4}$ cm^{-1} and $(11.9 \pm 1.2) \times 10^{-4}$ cm^{-1} for the wavelengths λ_1 and λ_2 , respectively.

Figure 3 shows the dependences of the optical scattering coefficients at wavelengths λ_1 and λ_2 for the laser radiation polarisation along the crystallographic axes b and c on the laser radiation power. At a fixed wavelength, power, and

polarisation of laser radiation, the optical scattering coefficient μ was determined as the difference between the optical loss coefficient β and optical absorption coefficient α . It can be seen that the scattering coefficients in the LBO crystal at the considered wavelengths remain constant for two directions of laser radiation polarisation. The average μ values are $(9 \pm 2) \times 10^{-4}$ cm^{-1} and $(240 \pm 20) \times 10^{-4}$ cm^{-1} for the wavelengths λ_1 and λ_2 , respectively.

According to the results of measurements by the method of integrating spheres at wavelengths λ_1 and λ_2 , the scattering coefficients were $(10 \pm 1) \times 10^{-4}$ cm^{-1} and $(200 \pm 10) \times 10^{-4}$ cm^{-1} , in good agreement with data obtained by the PRLC method.

5. Conclusions

The results obtained in this work demonstrate that the optical absorption and scattering coefficients of an LBO crystal remain constant for two orthogonal directions of radiation polarisations (along the b and c axes) when propagating along the crystallographic axis a of cw linearly polarised laser radiation with a wavelength $\lambda_1 = 1070$ nm (power range 50–300 W) and linearly polarised pulsed laser radiation with a wavelength $\lambda_2 = 532$ nm (power range 5–50 W, pulse duration 1.3 ns, repetition rate 1 MHz, maximum pulse energy 50 μ J). The values of the scattering coefficients are in good agreement with the values obtained using integrating spheres.

The difference between the scattering coefficients at the wavelengths λ_1 and λ_2 cannot be explained using conventional mechanisms, such as Rayleigh scattering, the intensity of which is proportional to λ^{-4} . This difference may be due to the optical inhomogeneity of the LBO crystal under study, scattering at the end facets of the crystal, as well as new mechanisms of laser radiation scattering in nonlinear optical crystals with pronounced ionic conductivity.

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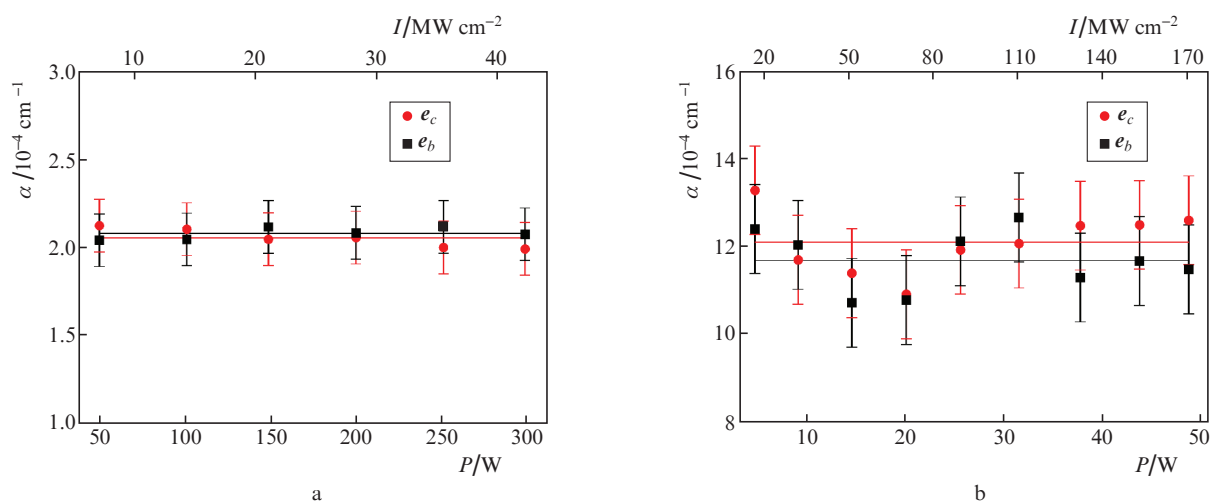


Figure 2. (Colour online) Dependences of the optical absorption coefficients of the LBO crystal at (a) $\lambda_1 = 1070$ nm and (b) $\lambda_2 = 532$ nm on the laser radiation power for two polarisation directions.

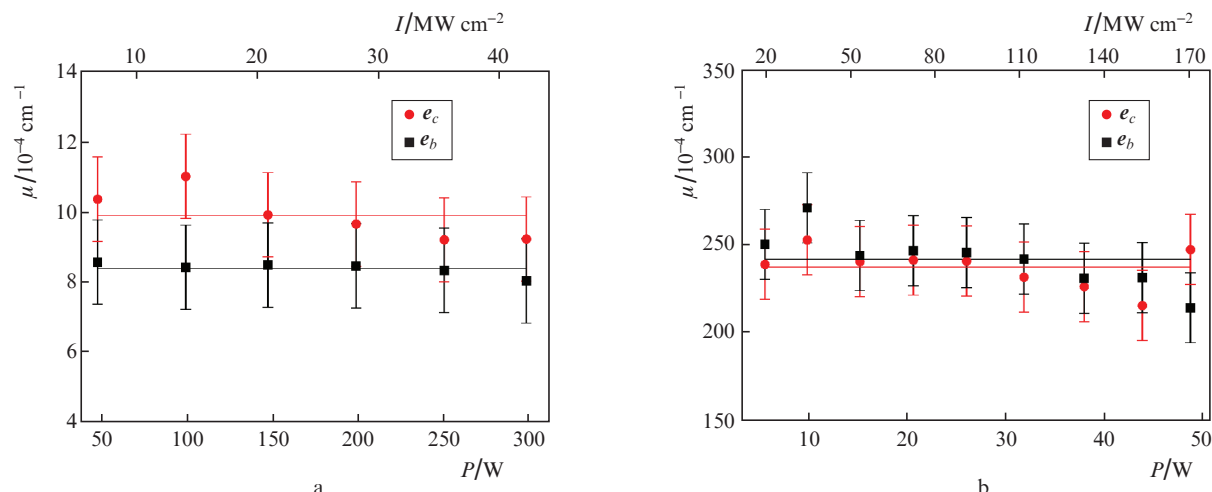


Figure 3. (Colour online) Dependences of the optical scattering coefficients in the LBO crystal at (a) $\lambda_1 = 1070$ nm and (b) $\lambda_2 = 532$ nm on the laser radiation power for two polarisation directions.

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