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Optical properties and potential of LB4 for THz wave generation

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ABSTRACT

Optical properties of a $Li_2B_4O_7$ (LB4) crystal are determined in the spectral range 0.2-1.6 THz. Dispersion of the refractive index components for o- and e-wave are approximated in the form of Sellmeier equations. They are subsequently used to determine the possible interaction types and to calculate the phase-matching angles to get THz waves by difference frequency generation. The damage threshold is determined as well as the coherence length for all possible types of three wave interactions under the pump by fs Ti: Sapphire laser pulses at 950 nm. The efficiency of the processes is estimated. Using trains of hundreds of pulses at 950 nm it was found to be 1.32 times of that for β-BBO crystal laser pump.

Keywords: lithium tetraborate, difference frequency generation, femtosecond pulses, phase matching

1. INTRODUCTION

The negative uniaxial ($n_o > n_e$) nonlinear lithium tetraborate ($Li_2B_4O_7$ or LB4) crystal belongs to 4*mm* point group symmetry. Its primitive cell consists of 104 (8*13) atoms [1]. The maximum transparency range at "0" transmission level extends from a short wavelength of 160 nm to 3500 nm [2]. The crystal is characterized by low level of optical absorption coefficient $\alpha < 10^{-4}$ cm^{-1} in the maximum transparency window [3] and high degree of uniformity of the refractive index $\Delta n/n = 10^{-6}$/mm [2]. The average number of dislocations in the as-grown high-quality crystals does not exceed 10/cm^2 with large defect-free regions [4]. The refraction coefficient in this spectral range are slightly changes with the temperature ($\partial n/\partial T \approx 10^{-6}$ K^{-1}) [5], and the damage threshold exceeds almost for 2 times of that for the LBO crystal and equal 1 GW/cm^2 under the 10 ns pulse pumping [6].

At present there exists an impressive technology of LB4 manufacturing of up to 70 mm in diameter and 200 mm in length [7]. Such situation together with the non-hygroscopicity renders LB4 as an extremely attractive crystal for application in laser frequency conversion. However, despite of 40-year application and a set of extraordinary properties, the LB4 has not yet found any significant application in practice for frequency conversion within the main transparency window and was not used for THz wave generation. The main reason for the lack of a noticeable interest to this crystal is its low nonlinear coefficients.

The values of the second-order nonlinear susceptibility coefficients of the LB4 responsible for the efficiency of frequency conversion into the THz range are twice lower than that of the popular LBO crystals. Namely, when the Kleinman symmetry conditions are valid $d_{15} = d_{24} = d_{31} = d_{32} = 0.12 \pm 0.03$ pm/V, $d_{33} = 0.47 \pm 0.03$ pm/V at a wavelength of 1064 nm [2]. It should be outlined, the meaning of nonlinear susceptibility coefficient outside the main transparency window, where the Kleinman conditions are not valid, the meaning of the coefficient d_{31} is not known yet. It means that the LB4 is 4 times inferior to the LBO crystal by the figure of merit (FOM) that is proportional to the potential efficiency of frequency conversion. On the other hand, the extraordinary high damage threshold of the LB4 to 10 ns pulses at 1064 nm is twice of that of the LBO and 8 times of fused quartz [2, 6].

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The extraordinary short wavelength boundary of the transmission spectrum also indicates this property. Dispersion equations for transparency window are available in [5]. This data demonstrates the possibility of effective generation of THz radiation given that the phase-matching conditions are met. Again, limited data on the damage threshold may be inconsistent and should be rechecked. So, the main factors that hindering investigation of the THz wave generation are few available pumping sources with extraordinary high output energy and lack of enough knowledge on the dispersive properties of the LB4 in the THz range.

In the present work, the optical properties of the LB4 crystal are investigated at wavelengths range of 0.2–1.6 THz (1500-180 μm) through THz time domain spectrometer, and the possibility of the THz wave generation is estimated.

2. METHODS AND MATERIALS

A LB4 single-crystal boule was grown by Bridgman technique, and specimens were fabricated by the Institute of Geology and Mineralogy of the SB RAS. The $\vec{Z}$ optical axis of the samples is in the plane of the input face that allows us to determine the optical properties of pumping radiation polarized in directions perpendicular to the X and Y axes. The appearance of the sample is shown in the insert of Figure 1.

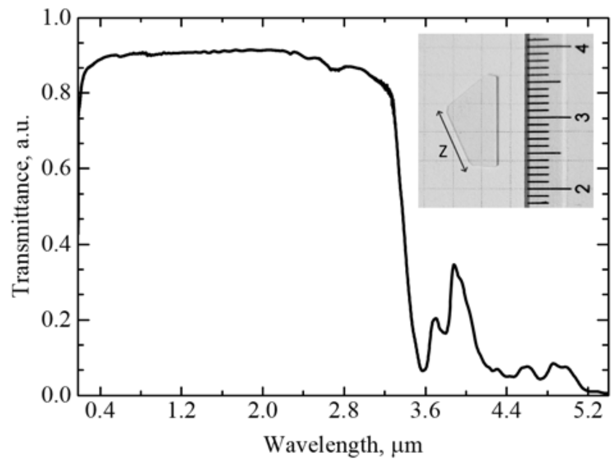

Figure1. Transmission spectrum in the main transparency window is shown. Photo of a sample under test is on the insert.

At the UV and VIS range, transmission spectrum was measured at room temperature by using an UV spectrophotometer UV 3100 (Shimadzu, Japan). FTIR spectrometer Nicolet 6700 (Thermo Scientific, USA) was used to determine mid-IR absorption spectrum. It was found the crystal possesses high optical quality corresponding to the best-known samples [3]. The arrow in the figure insert indicates the direction of the $\vec{Z}$ axis.

The dielectric properties of the sample were studied in the frequency range 0.2–1.6 THz (1500-180 μm) at room temperature through a commercial terahertz time domain spectrometer Teraplus 4000 (Teraview, UK). The aperture of the sample and TPX lens considerably exceeded the aperture of the interacting beams that exclude the influence of the edge effects on the measurement results. Absorption coefficient and calculated refractive index components spectra are shown in Figure 2. The obtained results on dispersion properties are approximated in the form of dispersive equations as:

$$n_o^2 = 8.085 + \frac{0.644\lambda^2}{\lambda^2 - 14277},$$
$$n_e^2 = 7.169 + \frac{0.345\lambda^2}{\lambda^2 - 18139}. \quad (1)$$

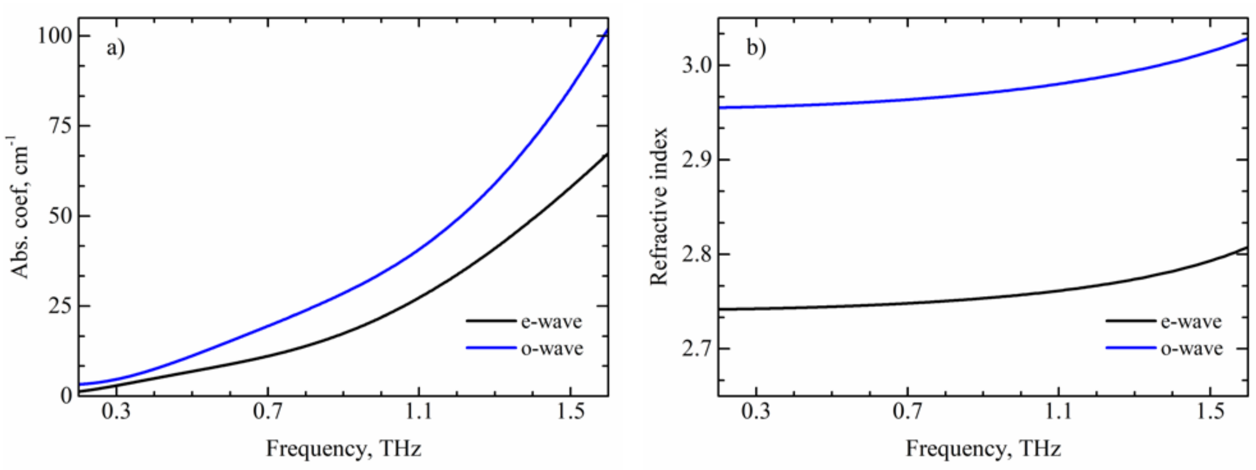

Figure 2. Absorption coefficient (a) and refractive index component (b) spectra in THz region.

The laser complex Start-480M (HCEI SB RAS, Russia) operating in femtosecond regime [8] was used to estimate the laser damage threshold. External view on the experiment facility is shown in Figure 3. To obtain correct data, we tend to compare data on LB4 with that for well-known β-BBO crystal. The pump laser pulses were passed through a half-wave attenuator and then through a 400-mm focal length lens installed on a motorized stage. By moving the lens, the beam cross-section was controlled that makes possible to adjust smoothly the pump beam intensity to control the progress of the laser-induced damage process. The comparison was also made in ps regime of operation. It was estimates that the damage threshold for LB4 is ~1.3 times higher to that for β-BBO [9], in the experiment conditions under the study it is ≥ 100 TW/cm^2.

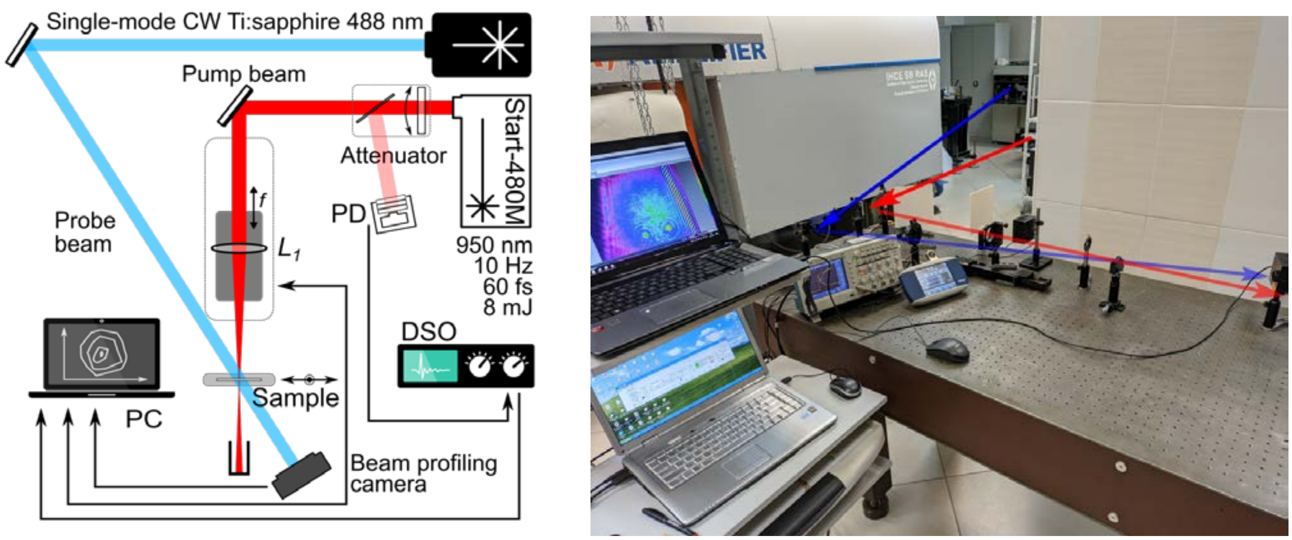

Figure 3. Optical set-up: PD is a photodiode, DSO is a digital oscilloscope, L_1 is a 400-mm focusing lens (a); external view on the facility used (b).

3. MODELING RESULTS

When the Kleinman symmetry conditions are valid, the effective second-order nonlinear susceptibility coefficients for o-o $\rightarrow$ e, o-e $\rightarrow$ o and e-o $\rightarrow$ o type of interactions are, $d_{\text{eff}}^{oo\rightarrow e} = d_{\text{eff}}^{eo\rightarrow o} = d_{\text{eff}}^{oe\rightarrow o} = d_{15}\sin\theta$, respectively. For e-e$\rightarrow$ e interaction type, the effective nonlinear susceptibility coefficient is $d_{\text{eff}}^{ee\rightarrow e} = 3d_{15}\cos^2\theta\sin\theta + d_{33}\sin^3\theta$ [10]. It should be outlined that the coefficients d_{15} is 8 times less than coefficient d_{33} = 0.42 pm/V. So, when θ = 90° the d_{15} contained part is vanished, and it is possible to measure d_{33} component [10]. While if the Kleinman symmetry conditions are not valid (in the case of THz wave generation), the effective second-order nonlinear susceptibility coefficients are $d_{\text{eff}}^{eo\to o} = d_{\text{eff}}^{oe\to o} = d_{15}\sin\theta$ for o-e→o and e-o→o type of interaction, and $d_{\text{eff}}^{oo\to e} = d_{31}\sin\theta$ for o-o→e. For e-e→e type of interaction $d_{\text{eff}}^{ee\to e} = (2d_{15} + d_{31})\cos^2\theta\sin\theta + d_{33}\sin^3\theta$. The value of the coefficient d_{33} for the THz range is still unknown.

Dependence of the phase matching angle versus the generating central THz frequency under the pump by fs-pulses at 950 nm, as well as corresponding FOM, are shown in Figure 4. From this figure, for a fixed pump frequency ω_3, a larger phase matching angle θ corresponds to a higher generating frequency. Coherence length for fabricated samples was calculated using obtained dispersion equations and data on refractive indices from [5] for 950 nm by using well known formula:

$$L_{coherence} = \frac{\pi}{\left|k(\omega_3) - k(\omega_3 - \omega_1 = \omega_2) - k(\omega_1)\right|}, \tag{2}$$

where $\omega_3 > \omega_2 > \omega_1$.

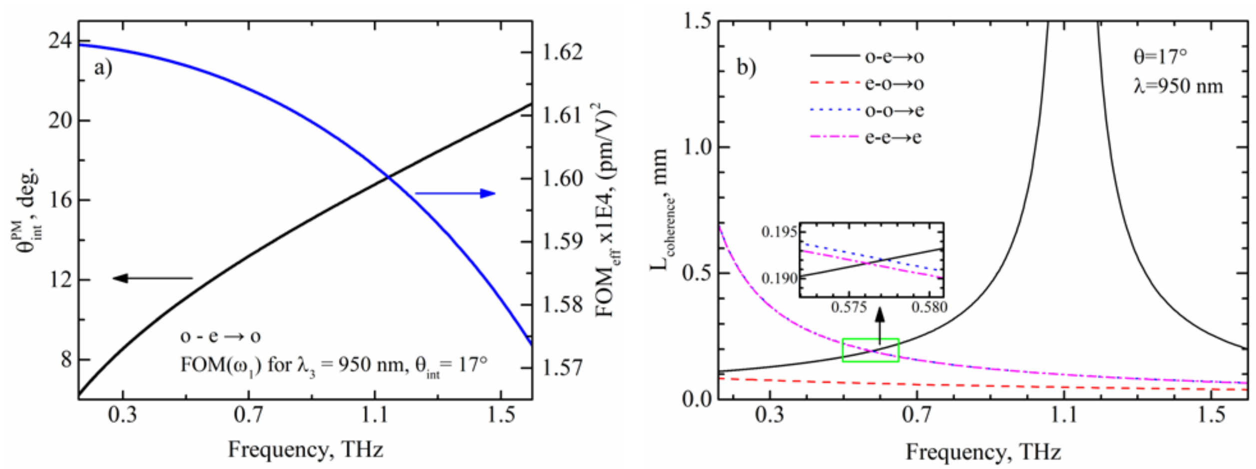

Figure 4. Figure of merit and the internal phase-matching angle for 950 nm pump (a) and coherence lengths for different types of iteration (b) at the THz frequencies.

So, for pure LB4 crystal, its potential long-wavelength THz conversion efficiency under the ultrashort pulses pump is not limited by optimal phase matching angle and the small coherence length. That means this crystal could be further enhance the THz wave conversion efficiency through periodically poled crystal form.

4. DISCUSSIONS AND SUMMARY

The optical properties of the nonlinear crystal $Li_2B_4O_7$ (LB4) have been studied in the terahertz range at room temperature. The small (~300 µm) thickness of the used crystal limits the accuracy of measurements of the optical loss coefficient by the value ≤ 1-2 cm^{-1} in the THz region. The dispersive equations were formulated as well as non-linear susceptibility coefficients determined excluding coefficient d_{33}. On basis of the measurement results, the possibilities of THz wave generation are estimated. The extra high damage threshold of ≥ 100 TW/cm^2 (30% higher to that for β-BBO) in line with the low absorption coefficient and extra-large size of the grown crystals, up to 200 mm in diameter, allows to realized record high efficient and output energy of the THz pulses even at estimated high optical absorption coefficient.

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