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Properties of Oxide Nonlinear Crystals in the Wavelength Range Over 600 μm

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Abstract. Absorption coefficients and refractive index components of polyatomic oxide nonlinear crystals LBO, β-BBO, and LB4 are determined in the THz range. The possibility of creating of highly efficient radiation sources operating in the wavelengths range over 600 μm is shown.

Keywords: nonlinear crystal, sub-THz range, THz range, optical properties, temperature dispersion.

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Introduction

Creation of new radiation sources operating in the sub-THz spectral range (0.05 – 0.5 THz) is of urgent need for application in long trace monitoring systems of the atmosphere [1]. Popular THz sources based on parametric frequency converters of IR lasers in semiconductor ($ZnGeP_2$, GaSe, etc.) and organic (DAST, OH1, etc.) nonlinear crystals cannot be fully used to solve applied problems due to low operational characteristics [2] including low damage threshold. A solution of the problem of creating the long-wavelength radiation sources is application of parametric frequency converters based on oxide nonlinear crystals (NC). Due to high mechanical properties suitable for cutting and polishing, high optical damage threshold and excellent thermal conductivity, several polyatomic oxide NC's (β-BaB_2O_4 (BBO), LiB_3O_5 (LBO), $Li_2B_4O_7$ (LB4), etc.) are widely used as optical parametric frequency converters within their main transparency windows [3–5]. These crystals have possess low or even very low nonlinear susceptibility coefficients (from 2.3 pm/V (BBO) down to 0.45 pm/V (LB4) in difference to above mentioned

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semiconductor and especially organic nonlinear crystals. Nevertheless, we assume the possibility of creation high-efficient frequency oscillators that are based these crystals. Due to their extremely high laser damage thresholds it is possible to use them under high-intensity femtosecond pulses pump. High intensity pump pulses can compensate or even overcompensate low nonlinear coefficients of these crystals.

However, their application for THz generation were strongly prevented by limited number of suitable fs lasers suitable for pumping, lack or straightly scattered data on damage thresholds of the considered crystals [6], and optical loss coefficients in the THz range. This work is devoted to study of the optical properties of oxide NC's and possibilities of THz wave generation in the spectral range over 600 μm that coincides with the maximum transparency range of the atmosphere that is the most suitable for trace sensing of gas pollutions.

1. Main results

Beta-barium borate (β-BaB_2O_4 or BBO) is negative uniaxial crystal of point group symmetry $3m$. Main transparency window is 185 – 3500 nm. The obtained data on optical properties in the THz range (Fig. 1) shows possibility of difference frequency generation (DFG) under phase matching (PM) conditions [7]. Low optical loss coefficients (< 0.5 cm^{-1}) at the long-wavelength

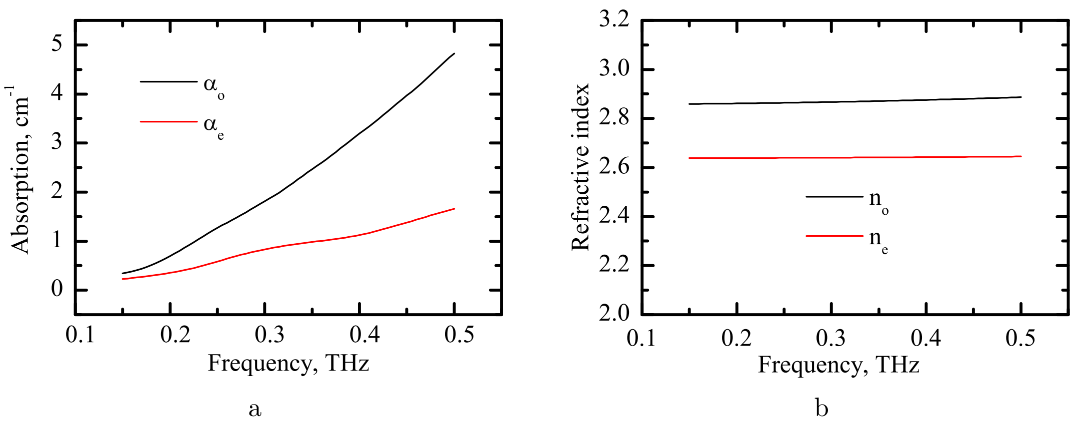

Fig. 1. Absorption coefficient (a) and refractive index component (b) spectra in the THz range of the BBO crystal

range of the absorption spectra and maximal nonlinear coefficient from all considered NC's will ensure high efficiency of frequency conversion.

Lithium triborate (LiB_3O_5 or LBO) is a negative biaxial crystal of the point group symmetry $mm2$ [8]. The crystal is characterized by an extremely low absorption coefficient of down to or below 10^{-5} cm^{-1} in the main transparency window [9] and low absorption coefficients at the long-wavelength part of THz spectra (Fig. 2). For the values of its refractive indices, the relation $n_x < n_y < n_z$ is observed in the visible range. It changes in the THz range for $n_z < n_x < n_y$ [10]. This is the only one NC under consideration that can be used to manufacture large-aperture samples for parametric frequency converters [11]. The refractive index components nonlinear decrease in the visible range within $(1.5 - 3.5) \cdot 10^{-3}$ a.u., as the temperature rises from 20 °C to 200 °C [12]. The largest change in Δn is observed for the n_y component. Temperature dependences of the refractive indices clearly appear in the THz range. When LBO crystal is

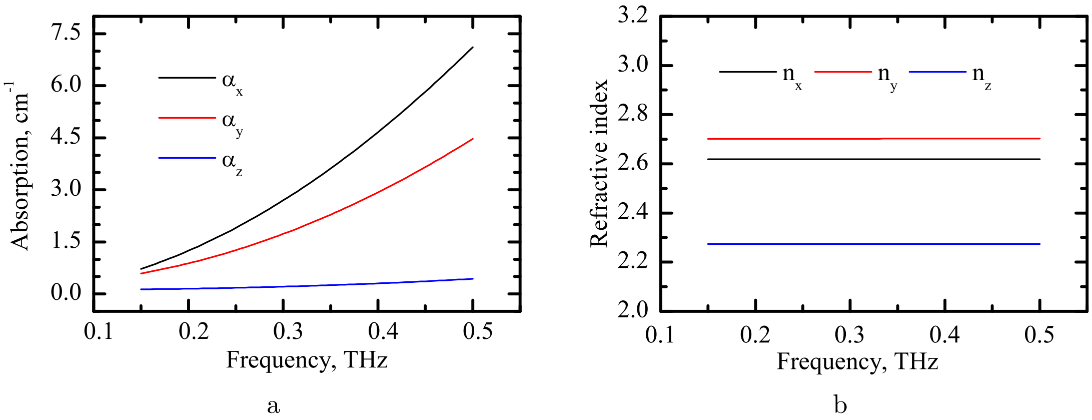

Fig. 2. Absorption coefficient (a) and refractive index component (b) spectra in the THz range for LBO crystal

heated from −190 ℃ to 20 ℃ [13], the values of the refractive index components n_x and n_y are increasing respectively, by about 0.15 and 0.5. Optical properties of crystals under the study heated up to 100 ℃ were investigated in the THz range with a commercial spectrometer Zomega Z3 (Zomega, USA). This measurements was also independently confirmed on a laboratory time-domain THz spectrometer assembled in accordance with modified scheme [14]. For this purposes we used femtosecond Ti:Sapphire laser Chameleon Vision 2 (Coherent, USA) was used as a pump source. Registration of THz signal was based on the electro-optical detection in the ZnTe crystal using the ABL-100 (Zomega, USA) auto-balance detector. Temperature dependences of the output signals was based on a laboratory setup with a thermostat supplied with a PID-controller which allows maintaining the parameters of the test sample by temperature stabilization within 0.1 ℃. The results of the measurements of temperature dependence of refractive indices are shown in Fig. 3.

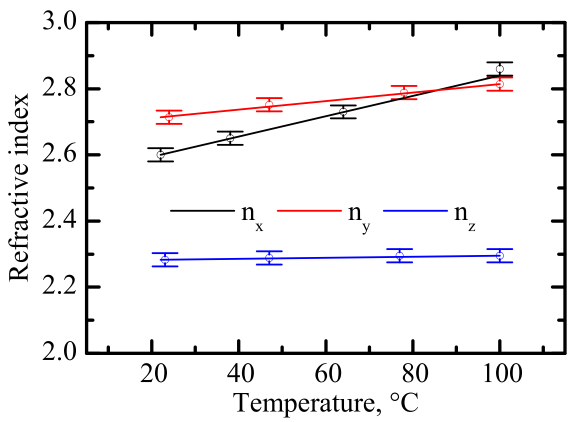

Fig. 3. Temperature dispersion of refractive index components of LBO at 200 GHz frequency

It should be noted presence of cross-point of the refractive index components n_x and n_y due to the difference of their temperature dispersions (that excludes related type of phase matched interactions) at 84 °C. Similar feature is observed for LGS and LGSE nonlinear crystals at room temperature at wavelength, respectively, of about 7 and 9 μm [15]. These results violate the existing classification of nonlinear crystals on positive and negative. Which requires indicating the boundaries of both frequency and temperature ranges for different types of nonlinear interactions.

DFG from visible to THz range under PM conditions is also possible for LBO and long-wavelength losses than is less than 0.8 cm^{-1} for x and y axes and less than 0.2 cm^{-1} for z.

Lithium tetraborate $Li_2B_4O_7$ (LB4) is uniaxial negative crystal that belongs to the $4mm$ point group symmetry [16]. Its absorption coefficient in long-wavelength range of the THz domain is

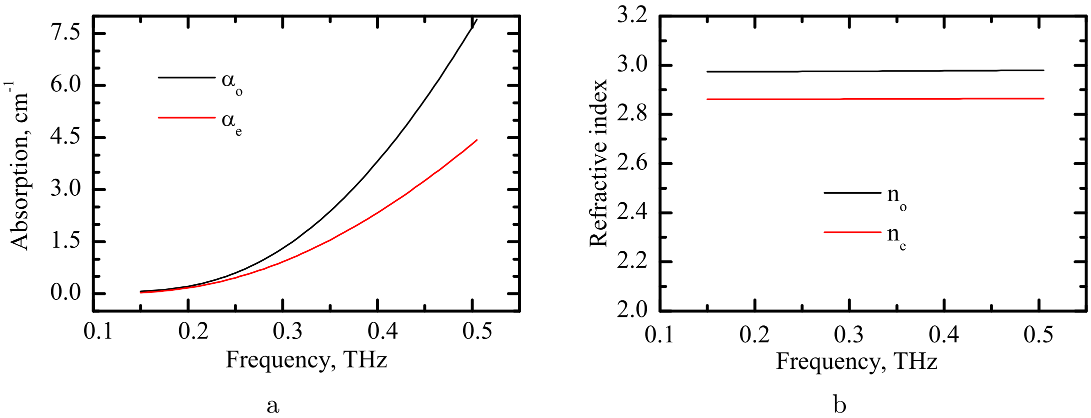

Fig. 4. Absorption (a) and refractive indices (b) spectra in the THz range for LB4 crystal

lower than that of NC BBO (Fig. 4). But DFG under PM conditions is possible only for low-efficiency interactions [7]. Thus, despite the suitable optical characteristics in the THz range using LB4 for creating THz sources is less preferable than LBO and BBO crystals.

Conclusion

The considered THz spectral properties of BBO, LBO and LB4 oxide nonlinear crystals show their applicability for creating sub-THz wave generators that could be pumped by high-power fs UV, visible, or near-IR lasers. Meanwhile optical parametric oscillators based on BBO and LBO crystals renders highest efficiency. We suppose to use LBO crystals, that could be grown with a largest optical-aperture among the known nonlinear optical crystal, to obtain sub-THz sources with highest output power.

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Свойства оксидных нелинейных кристаллов на длинах волн более 600 мкм

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Аннотация. Определены спектры коэффициентов поглощения и компонент показателей преломления многоатомных оксидных нелинейных кристаллов LBO, β-BBO, LBO для ТГц диапазона, показывающие возможность создании высокоэффективных генераторов излучения на длинах волн более 600 мкм.

Ключевые слова: нелинейные кристаллы, суб-ТГц диапазон, ТГц диапазон, оптические свойства, температурная дисперсия.