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Optical properties of β -BBO and potential for THz applications

N. A. Nikolaev^{1, 2, a}, Yu. M. Andreev^{3, 4}, V. D. Antsygin², T. B. Bekker⁵, D. M. Ezhov¹, A. E. Kokh⁵, K. A. Kokh⁵, G. V. Lanskii^{3,4}, A. A. Mamrashev^{1, 2}, V. A. Svetlichnyi^{1, 4}

¹High Current Electronics Institute, SB RAS, 2/3 Akademicheskii Ave., 634055, Tomsk, Russia; ²Institute of Automation and Electrometry, SB RAS, 1 Academician Koptyug Ave., 630090, Novosibirsk, Russia; ³Institute of Monitoring of Climatic and Ecological Systems, SB RAS, 10/3 Akademicheskii Ave., 634055, Tomsk, Russia; ⁴Siberian Physical Technical Institute of Tomsk State University, 1 Novosobornaya Sq., 634050, Tomsk, Russia; ⁵Institute of Geology and Mineralogy, SB RAS, 3 Academician Koptyug Ave., 630090, Novosibirsk, Russia;

^aCorresponding author's e-mail: nazar@iae.nsk.su

Abstract. The anisotropy of optical properties of high quality beta barium borate crystal $(\beta$ -BaB₂O₄, β -BBO) was studied in the main transparency window by using classic spectroscopic methods and in the range of 0.2 - 2 THz by using THz time-domain spectroscopy. β -BBO crystals were grown by the top-seeded solution technique in a highly resistive furnace with a heat field of 3-fold axis symmetry. At room temperature (RT), absorption coefficient in the maximal transparency window in grown crystals did not exceed 0.05 cm⁻¹. Strong absorption anisotropy was observed in 3 – 5 μ m and the THz range. At 1 THz absorption coefficients for e and o wave were, respectively, 7 cm⁻¹ and 21 cm⁻¹ at RT; 2 cm^{-1} and 10 cm^{-1} at 81 K. At the most attractive for out-of-door applications range < 0.4 THzthe absorption coefficient is found to be very low: below 0.2 cm⁻¹ at RT and 1 cm⁻¹ at 81 K. Refractive indices dispersions measured by THz-TDS were approximated in the form of Sellmeier equations. Birefringence is found quite large for phase matched difference frequency generation (DFG) or down-conversion into the THz range (THz-DFG) under near IR pump at RT and 81 K. Type II (oe-o and eo-o), and type I (ee-e) three wave interactions can be realized at RT. THz-DFG of Nd:YAG laser and KTP OPO can be realized by type II (oe-o) three-wave interaction. For selected spectral ranges of femtosecond Ti:Sapphire laser efficient phase matched and group velocity matched optical rectification can be realized by another two types of three wave interactions. Accounting other well-known attractive physical properties of β-BBO crystal, wide application in THz technique can be forecasted.

1. Introduction

The interest to terahertz (THz) sources has been growing significantly because of promising applications in the fields of spectroscopy of biomolecules and rotation lines of gases, nondestructive imaging, communication [1] and environmental monitoring systems [2]. Progress in the THz



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technology developments has, however, been hampered both by a lack of intense THz sources and operational difficulties in using available nonlinear crystals for down-converters of IR lasers as THz wave generators [2, 3]. Disadvantages of widely used nonlinear crystals for THz applications stimulate a growing interest in searching for new or known, but not efficiently utilized, highly-reliable anisotropic crystals.

In this study, anisotropy of optical properties of high quality β -BBO crystal discovered in 1985 by Chen et al. [4] was studied in the main transparency window by using classic spectroscopic methods and, for the first time, in the range of 0.2 - 2 THz by the methods of THz time-domain spectroscopy to find out possibility of efficient down-conversion of near IR lasers into the THz range.

2. Experimental results

The High quality β -BBO crystals were grown by the modified top-seeded solution technique in a highly resistive furnace with a heat field of 3-fold axis symmetry [5] (Fig. 1).



Fig. 1. As-grown single crystal BBO boule (a) and test samples (b).

Crystals were grown in BaB₂O₄-NaBaBO₃-Ba₂Na₃[B₃O₆]₂F system. After determining the equilibrium temperature, the seed was allowed to grow with continuous rotation 1 rpm. The cooling and pulling rates varied, respectively, from 0.4 to 2 °C/day and 0.5 to 0.1 mm/day. Test samples of $10 \times 10 \times 2 \text{ mm}^3$ cut at $\theta = 0^\circ$ and 90° to study optical properties for *o*- and *e*-wave were manufactured (Fig. 2b). Test samples were cooled by mounting in the bath cryostat with liquid nitrogen.



Fig. 2. Absorption spectra (a) in the maximal transparency window and (b) in the THz range at RT (solid lines) and 81 K (dashed lines) in BBO crystal.

At room temperature (RT), absorption coefficient in the maximal transparency window in grown BBO

crystals did not exceed 0.05 cm⁻¹ (Fig. 2a). Strong absorption anisotropy was observed in $3 - 5 \mu m$ and the THz range (Fig. 2b). At 1 THz absorption coefficients for *e* and *o* wave were, respectively, 7 cm⁻¹ and 21 cm⁻¹ at RT; 2 cm⁻¹ and 10 cm⁻¹ at 81 K. At the most attractive for out-of-door applications range < 0.4 THz, absorption coefficient is found to be very low: below 1 cm⁻¹ at RT and 0.2 cm⁻¹ at 81 K that is favorable for long THz wave generation.



Fig. 3. (a) Dispersions of refractive indices in BBO crystal in the THz range and (b) and RT *oe-o* type phase matching curves.

Dispersions of refractive indices were approximated in the form of Sellmeier equations for RT (1, 2) and 81 K (3, 4) with validity range $140 - 3500 \mu m$.

$$n_o^2 = 2.040 + \frac{0.816\,\lambda^2}{\lambda^2 - 12815},\tag{1}$$

$$n_e^2 = 2.478 + \frac{0.160\,\lambda^2}{\lambda^2 - 15598} \tag{2}$$

$$n_o^2 = 4.612 + \frac{3.117\,\lambda^2}{\lambda^2 - 16175},\tag{3}$$

$$n_e^2 = 5.970 + \frac{0.800\,\lambda^2}{\lambda^2 - 16659} \tag{4}$$

Birefringence is found quite large for phase matched difference frequency generation (DFG) or downconversion into the THz range (THz-DFG) under near IR pump at RT and 81 K (Fig. 3b). It was established that type II (*oe-o* and *eo-o*), and type I (*ee-e*) of three wave interactions can be realized at room temperature. THz-DFG of Nd:YAG laser and KTP OPO can be realized by type II (*oe-o*) threewave interaction. For selected spectral ranges of femtosecond Ti:Sapphire laser efficient phase matched and group velocity matched optical rectification can be realized by another two types of three wave interactions.

3. Conclusion

Optical properties of high optical quality BBO crystals were studied in the THz range. It was found that at long wavelength range (> $650 \mu m$) BBO possesses low optical absorption coefficient down to

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below 1 cm⁻¹ and it is phase matchable for down conversion of near IR lasers into this range. Accounting other well-known attractive physical properties of β -BBO crystal, wide application in THz technique can be forecasted.

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