Millimetre-wave range optical properties of BIBO

N A Nikolaev\textsuperscript{1,2}, A A Mamrashev\textsuperscript{2}, V D Antsygin\textsuperscript{2}, D M Ezhov\textsuperscript{3},
D M Lubenko\textsuperscript{4}, V A Svetlichny\textsuperscript{3}, Yu M Andreev\textsuperscript{3,4,5} and V F Losev\textsuperscript{4}

\textsuperscript{1}Institute of Laser Physics SB RAS, 15 B Academician Lavrentiev Ave., Novosibirsk, 630090, Russia
\textsuperscript{2}Institute of Automation and Electrometry SB RAS, 1 Academician Koptyug Ave., Novosibirsk, 630090, Russia
\textsuperscript{3}Tomsk State University, 36 Lenin Ave., Tomsk, 634050 Russia
\textsuperscript{4}Institute of High Current Electronics SB RAS, 2/3 Akademichesky Ave., Tomsk 634055, Russia
\textsuperscript{5}Institute of Monitoring of Climatic and Ecological Systems SB RAS, 10/3 Academichesky Ave., Tomsk 634055, Russia

Corresponding author: Nazar Nikolaev, email: Nazar@iae.nsk.su

Abstract. We present the thorough studies of optical properties of BiB\textsubscript{3}O\textsubscript{6} (BIBO) crystal in
the millimeter-wave (subterahertz) range. We observe a large birefringence $\Delta n = n_z - n_X = 1.5$
and the values of absorption coefficients of all three axes to be less than 0.5 cm\textsuperscript{-1} at the
frequency of 0.3 THz. The difference from visible range in angle $\phi$ between the dielectric axis $z$
and crystallographic axis $X$ is found to be more than 6°. The simulated phase-matching
curves in the $xz$ plane of the crystal show the optimal value of the angle $\theta$ to be around
25.5°±1° for an efficient millimeter-wave generation under the pump of 1064 nm laser
radiation.

1. Introduction
The negative biaxial crystal of bismuth triborate belongs to the non-centrosymmetric monoclinic space
group C2. BIBO was demonstrated as an efficient ultraviolet radiation source, a second harmonic
generator, and a parametric frequency converter of ultrashort pulses [1]. Its dielectric $x$-axis is parallel
to the crystallographic $Y$-axis (crystallographic $b$-axis) and the angle $\phi$ between the axes $z$ and $X$ is
wavelength-dependent. Its value is about 47° in the main transparency window [2]. Previously
reported that the crystal is transparent in the range below 2 THz and exhibits significant birefrinence
[3]. However, due to the small thickness of the samples, it was not possible to get a reliable measure of
the absorption coefficients. In this work, samples with a thickness of 5 mm were studied, which made
it possible to refine the absorption coefficient of the crystal and obtain some new data on the
properties of the BIBO at terahertz frequencies.

2. Methods and samples
Terahertz properties were measured using a conventional terahertz time-domain spectrometer (THz-
TDS). The description of the experimental setup can be found elsewhere [4]. In the current study, we
applied a newly developed measurement procedure described in [5]. It allowed us to measure the
absorption coefficient and the refractive index of the crystal for both axes during one cycle (without removing and rotating the sample) under nearly the same conditions. THz signals were acquired with a time step of 125 fs in the 60 ps time range which corresponded to a spectral resolution of about 20 GHz.

3. Experimental results

3.1. Optical properties

The absorption coefficient in the subterahertz range of spectra (below 0.3 THz) was found to be less than 0.5 cm\(^{-1}\) for all axes as it shown in figure 1. The absorption coefficients increase at higher frequencies showing typical behavior for the most of nonlinear crystals.

![Figure 1](image1)

**Figure 1.** Absorption coefficient components of BIBO crystal in the subterahertz range.

We have measured \(\phi\) angle using TeraScan frequency-domain spectrometer from Toptica Photonics (linewidth is about 10 MHz). The measurements were carried out with the crossed high-quality polarizers. It was shown that the dielectric frame \(xyz\) in the sub-THz is rotated more than 6 degrees from the visible and its dispersion was about 5 degrees (figure 2).

The dispersion of refractive indices decreases at sub-terahertz frequencies and almost disappears approaching millimeter waves (figure 2). In this area (~0.2 THz), the refractive index components are: \(n_x = 2.4, n_y = 2.6, n_z = 3.9\). The \(z\)-axis shows the greatest dispersion, which is due to the influence of the same strong absorption mode.

![Figure 2](image2)

**Figure 2.** The difference from visible range in angle \(\phi\) between the axes \(z\) and \(X\).
Taking this into account, the refractive indices were refined as it shown on a figure 3. We obtain the same order of refractive indices as earlier [3], but more precisely at the frequencies below 0.2 THz due to increased SNR.

Figure 3. Refractive index of BIBO crystal in the subterahertz range.

3.2. Phase-matching for a difference frequency generation
The fulfillment of phase-matching conditions for DFG is found to be possible in the main optical plane $xz$ ($\varphi = 0^\circ$). In total, two types of three-wave interactions (out of eight possible) were discovered - $sfs$ (figure 4) and $ffs$ (figure 5), where $f$ is fast wave, $s$ is slow wave, and the first letter corresponds to the longest wavelength (the ratio of the lengths of the interacting waves is determined by the agreement $\lambda_f \geq \lambda_s > \lambda_3$). The figures show the dependence of the phase-matching angle $\theta_{pm}$ on the generated wavelength. The curves for the case when the second wave is fixed (1064 nm, while the third wave is shorter) do not differ from the case when the third wave is fixed (while the second wave is longer).

Figure 4. The dependence of the phase-matching angle on the generated wavelength via $sfs$ type of interaction in the $xz$ principal plane.

Figure 5. The dependence of the phase-matching angle on the generated wavelength via $ffs$ type of interaction in the $xz$ principal plane.

4. Summary
In comparison with well-known $\beta$-BBO, or LBO crystals BIBO shows the highest nonlinear coefficients and the lowest absorption in the THz range, which in turn positions it as a promising downconverter of high-power laser radiation. A found dispersion of the $\phi$ angle should be considered
when generating broadband terahertz waves. The optimal value of the angle $\theta_{pm}$ found to be around $25.5^\circ\pm1^\circ$ for the efficient millimeter-wave generation under 1 $\mu$m laser pump.

**Acknowledgments**

This work was supported by the Russian Science Foundation (RSF), project № 19-19-00241.

**References**

[1] Petrov V et al., 2010 Femtosecond nonlinear frequency conversion based on BiB3O6 Laser & Photon. Rev., 4, 53-98
[2] Hellwig H, Liebertz J, Bohaty L 2000 Linear optical properties of the monoclinic bismuth borate BiB3O6 J. Appl. Phys., 88, 240–244
[3] Li Y et al. 2020 Optical properties of BiB3O6 in the terahertz range Results in Physics, 16, 102815.
[4] Rybak A et al. 2021 Terahertz Optical Properties of KTiOPO₄ Crystal in the Temperature Range of (− 192)–150°C, Crystals, 11(2), 125
[5] Mamrashev A et al. 2021 Terahertz Time-Domain Polarimetry for Principal Optical Axes of Anisotropic Crystals, Photonics 8(6), 213
[6] Antsygin V D et al. 2013 Optical properties of borate crystals in terahertz region Optics Communications 309, 333-337
[7] Nikolaev N A et al. 2018 Terahertz optical properties of LBO crystal upon cooling to liquid nitrogen temperature, Quantum Electronics 48(1), 19