Quantum Electronics **48** (1) 19–21 (2018)

NONLINEAR OPTICAL PHENOMENA https://doi.org/10.1070/QEL16515

Terahertz optical properties of LBO crystal upon cooling to liquid nitrogen temperature

N.A. Nikolaev, Yu.M. Andreev, N.G. Kononova, A.A. Mamrashev, V.D. Antsygin, K.A. Kokh, A.E. Kokh, V.F. Losev, O.I. Potaturkin

Abstract. **The anisotropy of optical properties of nonlinear lithium borate (LBO) crystals in the range of 0.2–2 THz is investigated by time-domain THz spectroscopy at room temperature and at T = 81 K. It is shown that the birefringence dramatically decreases upon cooling, as a result of which phase-matching conditions cannot be implemented. At the same time, the absorption coefficients α_x and α_y are found to decrease significantly with decreasing temperature, due to which the LBO crystal becomes a promising material for generating THz radiation via optical rectification and fabricating periodic structures and optical elements.**

Keywords: *lithium borate crystal, LBO, THz radiation, absorption coefficient, refractive index.*

1. Introduction

Biaxial nonlinear lithium borate LiB_3O_5 (LBO) crystal has a unique set of physical and chemical properties [1]. Its maximum transparency window ranges from 155 to 3200 nm; in addition, the absorption coefficient of this crystal does not exceed 10^{-5} cm^{-1} in the spectral range of 500–1200 nm in view of its structural features, which exclude incorporation of impurities [2, 3]. Among the known nonlinear crystals, LBO has the highest radiation resistance: it is 11.2 GW cm^{-2} at a wavelength of 1.064 μm of a nanosecond (25 ns) Nd:YAG laser [4]. The short-wavelength edge of the transmission band, lying in the vicinity of 155 nm, makes it possible to avoid two-photon absorption of the main and second harmonics of visible- and near-IR-range lasers. Noncritical phase matching of types I and II can be implemented in wide spectral ranges of the maximum transparency window. The existing growth technologies allow one to obtain LBO crystals of decimeter sizes with a mass above 1.5 kg [5]. In total, despite the fact that the quadratic nonlinear susceptibility coefficients of nonlinear LBO crystals are rather small (d_{31} = 0.67 pm V^{-1}, d_{32} = 0.85 pm V^{-1}, and d_{33} = 0.04 pm V^{-1} [1]), their physical properties and the advance in growth technology determine their use for frequency conversion of high-power lasers [3].

The current state of things in this field gives grounds for application of the LBO crystal as an efficient converter of the radiation of visible- and near-IR-range lasers into the THz range. However, to date, there are no published data on the practical application of this converter. There are a limited number of studies of the optical properties in the THz range. IR reflection spectroscopy and Raman spectroscopy were used to investigate the spectral properties in the range of 40–2500 cm^{-1} [6–8]. A detailed study of the optical properties in the range of 0.2–2.4 THz (6.7–80 cm^{-1}) was performed by a team of Russian and British scientists [9]. The dispersion properties measured by time-domain THz spectroscopy were approximated in the form of Sellmeier equations; it was shown (with their use) that there is a possibility of phase-matched conversion of laser radiation of the visible and near-IR ranges into the THz range. According to the data of [9], despite the low (several tenths of cm^{-1}) absorption coefficient of crystals in the phase-matching direction in the range of up to 0.5 THz, available decimeter-long LBO crystals cannot be used to generate THz radiation. With allowance for the temperature dependence of the phonon absorption in LBO crystals, which determines the loss in this range, the problems of designing frequency converters of decimeter length and covering a higher THz-frequency range can be solved by cooling crystals. The temperature dependence of the optical properties in the THz range was investigated by Chinese scientists using a time-domain THz spectrometer [10]; however, they studied the optical properties for differently polarised waves, propagating in the directions of two from three optical axes. In addition, the absorption coefficients evidently became negative at frequencies below 0.5 THz, a fact suggesting incorrect normalisation of THz signals and throwing into question the reliability of determining the refractive indices. Correct knowledge of optical properties is a necessary condition for

N.A. Nikolaev, A.A. Mamrashev Institute of High-Current Electronics, Siberian Branch, Russian Academy of Sciences, prosp. Akademicheskii 2/3, 634055 Tomsk, Russia; Institute of Automation and Electrometry, Siberian Branch, Russian Academy of Sciences, prosp. Akad. Koptyuga 1, 630090 Novosibirsk, Russia; e-mail: nazar@iae.nsk.su, mamrash@iae.nsk.su;
Yu.M. Andreev Institute of Monitoring of Climatic and Ecological Systems, Siberian Branch, Russian Academy of Sciences, prosp. Akademicheskii 10/3, 634055 Tomsk, Russia; e-mail: yuandreev@yandex.ru;
N.G. Kononova, A.E. Kokh V.S. Sobolev Institute of Geology and Mineralogy, Siberian Branch, Russian Academy of Sciences, prosp. Akad. Koptyuga 3, 630090 Novosibirsk, Russia; e-mail: kononovang@igm.nsc.ru, kohae@igm.nsc.ru;
V.D. Antsygin, O.I. Potaturkin Institute of Automation and Electrometry, Siberian Branch, Russian Academy of Sciences, prosp. Akad. Koptyuga 1, 630090 Novosibirsk, Russia; e-mail: antsigin@iae.nsk.su, potaturkin@iae.nsk.su;
K.A. Kokh V.S. Sobolev Institute of Geology and Mineralogy, Siberian Branch, Russian Academy of Sciences, prosp. Akad. Koptyuga 3, 630090 Novosibirsk, Russia; Novosibirsk National Research State University, ul. Pirogova 2, 630090 Novosibirsk, Russia; e-mail: k.a.kokh@gmail.com;
V.F. Losev Institute of High-Current Electronics, Siberian Branch, Russian Academy of Sciences, prosp. Akademicheskii 2/3, 634055 Tomsk, Russia; e-mail: losev@ogl.hcei.tsc.ru

Received 6 October 2017; revision received 15 November 2017
Kvantovaya Elektronika **48** (1) 19–21 (2018)
Translated by Yu.P. Sin'kov

using the potential of LBO crystal to the full extent when mastering the THz range.

In this paper, we report the results of a detailed comparative study of the behaviour of the absorption coefficients and refractive indices of LBO crystals in the range of 0.2–2 THz upon their cooling from room temperature to the liquid nitrogen temperature.

2. Experimental

The LBO crystals under study were grown by the flux method at the Sobolev Institute of Geology and Mineralogy of the Siberian Branch of the Russian Academy of Sciences [5]. The samples were polished plane-parallel plates 10 × 10 mm in size with a thickness of 2068 ± 5 μm, cut orthogonally to the x, y, and z optical axes, which are related to the a, c, and b crystallographic axes, respectively [1]. Measurements were performed using a time-domain THz spectrometer; this instrument, as well as the technique for processing output signals, were described in [11]. The samples were cooled to liquid nitrogen temperature using a continuously filled cryostat. Before starting an experiment, the optical scheme of the spectrometer was aligned according to the technique described in [12].

3. Results

Cooling nonlinear LBO crystals to a temperature of 81 K leads to a dramatic decrease in the absorption coefficients α_x and α_y (Figs 1a, 1b) to an undetectable level; at the same time, the absorption coefficient α_z varies only slightly: by no more than 1.5 cm^{-1} at a frequency of 1 THz (Fig. 1c).

Cooling an LBO crystal also significantly reduces its refractive indices n_x and n_y (by ~0.37 and ~0.47, respectively) (Figs 2a, 2b). At the same time, the refractive index n_z (Fig. 2c) slightly (by ~0.07) increases, which can be explained by the short-wavelength shift of the neighbouring long-wavelength phonon absorption peak. An estimation yielded the following values of temperature-dispersion coefficients for the refractive indices in the THz range: $\mathrm{d}n_x/\mathrm{d}T \approx 1.7 \times 10^{-3}$ K^{-1}, $\mathrm{d}n_y/\mathrm{d}T \approx 2.2 \times 10^{-3}$ K^{-1}, and $\mathrm{d}n_z/\mathrm{d}T \approx -3.2 \times 10^{-4}$ K^{-1}; they are two to three orders of magnitude larger than the corresponding values for the visible spectral range. An inevitable consequence of the significant decrease in the birefringence values $n_x - n_z$ (from ~0.33 at room temperature to ~0.04 at 81 K) and $n_y - n_z$ (from ~0.43 to ~0.03) is that the phase-matching conditions for THz generation cannot be implemented by the methods of nonlinear crystal optics. On the other hand, the dramatic decrease in the absorption coefficients α_x and α_y upon cooling to liquid nitrogen temperature is favourable for increasing the potential efficiency of optical–THz conversion of femtosecond laser radiation by optical rectification. In addition, due to the original set of physical properties and large (above 0.3 mm) coherence length [13], cooled LBO crystals can be used to fabricate high-efficiency periodic structures for frequency converters into the low-frequency (less than 1 THz) THz range and highly stable optical elements with orthogonally oriented optical axes x and y for different purposes. LBO crystals oriented along the optical axis z can be used as a material for temperature-insensitive

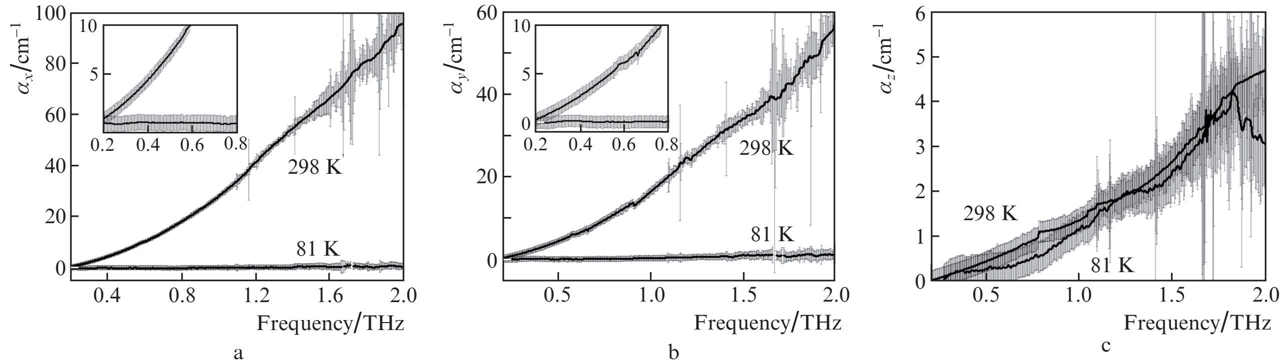

Figure 1. Absorption coefficients (a) α_x, (b) α_y, and (c) α_z of nonlinear LBO crystal at temperatures of 298 and 81 K. The insets in panels (a) and (b) show fragments of the same curves on an enlarged scale.

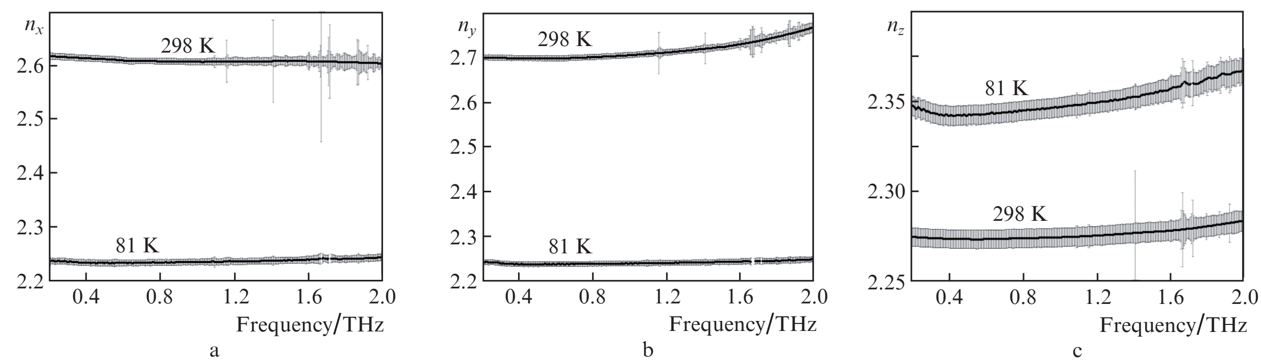

Figure 2. Refractive indices (a) n_x, (b) n_y, and (c) n_z of nonlinear LBO crystal at temperatures of 298 and 81 K.

elements of subcentimeter THz optics with high technical characteristics.

4. Conclusions

A complex study of the variations in the complete set of absorption coefficients and refractive indices of biaxial nonlinear LBO crystal in the range of 0.2–2 THz with a change in temperature from room temperature to 81 K was performed for the first time using a time-domain spectrometer. Cooling was found to reduce the absorption coefficients for waves polarised along the x and y axes to an undetectable level; at the same time, the birefringence drops by an order of magnitude, thus making the crystal structural properties close to isotropic. The optical properties of crystal for the waves polarised along the optical z axis change only slightly. Being cooled, the LBO crystal remains promising for use as an optical rectifier and for designing periodic structures and highly stable optical elements of the THz range, including those operating at room temperature.

Acknowledgements. This work was supported by the Russian Science Foundation (Project No. 1519-10021). The time-domain THz spectrometer was provided by the Shared Facilities Centre 'Spectroscopy and Optics' at the Institute of Automation and Electrometry of the Siberian Branch of the Russian Academy of Sciences.

References

1. Nikogosyan D.N. *Nonlinear Optical Crystals: A Complete Survey* (New York: Springer Science & Business Media, 2006).
2. Waasem N. et al. *Rev. Sci. Instrum.*, **84** (2), 023109 (2013).
3. Kokh A. et al. *J. Cryst. Growth*, **312** (10), 1774 (2010).
4. Furukawa Y. et al. *Appl. Phys. Lett.*, **65** (12), 1480 (1994).
5. Kokh A. et al. *J. Cryst. Growth*, **360**, 158 (2012).
6. Xia H.R. et al. *J. Mater. Res.*, **16** (12), 3464 (2001).
7. Xiong G. et al. *J. Raman Spectrosc.*, **24** (11), 785 (1993).
8. Jiang Y.J., Wang Y., Zeng L.Z. *J. Raman Spectrosc.*, **27** (8), 601 (1996).
9. Andreev Yu.M. et al. *Opt. Mater.*, **66**, 94 (2017).
10. Song K. et al. *Sci. Rep.*, **7**, 8122 (2017).
11. Antsygin V.D. et al. *Opt. Express*, **22** (21), 25436 (2014).
12. Antsygin V.D. et al. *Avtometriya*, **52** (4), 71 (2016).
13. Lubenko D.M., Losev V.F., Andreev Yu.M., Lanskii G.V. *Izv. AN SSSR, Ser. Fiz.*, **81** (10), 1375 (2017).