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Terahertz birefringence of potassium niobate crystals

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Introduction

Potassium niobate KNbO₃ (KN) is a nonlinear crystal used for frequency doubling [1], optical parametric amplification [2] and generation [3]. It is characterized by high nonlinearity, a wide transparency range that stretches from 0.4 to 4 µm with an absorption coefficient of <0.1 cm⁻¹ and a relatively high damage threshold [4]. KN is a negative biaxial crystal which refractive indices relate as follows $n_x > n_y > n_z$ in the visible range for waves with polarization parallel to the corresponding optical axes X, Y, and Z (b, a, and c crystallographic axes, respectively). It belongs to the *Amm2* space group of the orthorhombic crystal system at the room temperature. Linear and nonlinear optical properties of potassium niobate in the visible and the mid-infrared ranges were thoroughly studied [4]. In contrast, its terahertz properties require additional investigation. Early report of infrared reflectance noted the presence of strong phonon modes at 59 cm^{-1} (1.77 THz) and 197.5 cm^{-1} (5.92 THz) at the room temperature [5]. Raman spectroscopy results showed the presence of the modes as well [6]. Recent terahertz time-domain spectroscopy study reported optical properties of KN crystal only for two axes (X and Z) in the range of up to 1 THz [7]. In this paper we present refractive indices and absorption coefficients for all three optical axes of KN crystals in the terahertz range.

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ABSTRACT

We present terahertz optical properties (refractive indices and absorption coefficients) of potassium niobate crystals measured by time-domain spectroscopy in the range of 0.2–2.0 THz. We observe average refractive indices $n_x = 5.25$, $n_y = 4.8$, $n_z = 5.9$ for corresponding optical axes X, Y, Z with the large birefringence of $\Delta n = n_z - n_y = 1.1$. We report rising absorption coefficient at higher frequencies ($\alpha \sim 50$ cm⁻¹ at 1 THz for all three axes) while the dichroism is not pronounced. Somewhat higher absorption compared to the previous results could be attributed to some polydomain structure remaining in the crystal. © 2018 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND

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Methods

We studied potassium niobate monocrystals grown by a modified Czochralski method from solution-melt. The crystals were poled by applying electric field of 1.5 kV/cm while being cooled from ~200 °C to the room temperature in siloxane using the process analogous to [8]. Parallel plates of an approximate size of $10 \times 10 \times 0.25$ mm³ were cut perpendicular to different crystallographic axes from single-domain crystals. Terahertz properties were measured in the range of 0.2–2.0 THz by the custom-made terahertz time-domain spectrometer described elsewhere [9]. The spectrometer was tuned before the measurements using procedure allowing for a more precise investigation of anisotropic materials [10]. We performed three sets of measurements with terahertz radiation linearly polarized parallel to each of the three optical axes.

Results and conclusions

Measured refractive indices and absorption coefficients of the potassium niobate crystals are presented in the Figs. 1 and 2. Subscripts denote polarization of the terahertz radiation parallel to the corresponding optical axes *X*, *Y*, and *Z*. We find that KN exhibits low dispersion in the terahertz range with the average refractive indices $n_x = 5.25$, $n_y = 4.8$, $n_z = 5.9$ corresponding to rather large birefringence of $\Delta n = n_z - n_y = 1.1$ (see Fig. 1). Note that relation between the refractive indices is different compared to the visible range. The measured average refractive index $n_z = 5.9$ is somewhat









Fig. 1. Refractive indices of potassium niobate crystal for different optical axes in the terahertz range.



Fig. 2. Absorption coefficients of potassium niobate crystal for different optical axes in the terahertz range.

lower than the value of $n_z = 6.05$ in the work [7]. We also observe significantly lower average refractive index $n_x = 5.25$ compared to $n_x = 5.63$ reported in [7]. Absorption coefficients for all three axes behave similarly and rise at the higher frequencies and we do not observe significant dichroism in the terahertz range (see

Fig. 2). We attribute the main source of the absorption to the strong phonon modes at 1.77 THz and 5.92 THz. The measured absorption is somewhat higher compared to the previous results [7]. We think that this discrepancy could be attributed to some polydomain structure remaining in the crystal and observable in the visible range.

Relatively high nonlinearity (for example, $d_{32}(0.852 \,\mu\text{m}) = -11.0 \,\text{pm/V}$ [4]) of potassium niobate crystal indicates an opportunity for its usage for optical-to-terahertz and terahertz-to-terahertz conversion into long wavelength terahertz range. The knowledge of terahertz optical properties of the crystal is important for assessing such prospects.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.rinp.2017.12.078.

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