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THERMO-OPTIC PROPERTIES OF ORTHORHOMBIC $BeAl_6O_{10}$ CRYSTAL

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Abstract. Thermo-optic coefficients dn/dT were determined for orthorhombic $BeAl_6O_{10}$ crystal for light polarizations $E \parallel N_p$, N_m and N_g in the wavelength range of 0.4–1.1 μm. Thermo-optic dispersion formulas for principal thermo-optic coefficients of $BeAl_6O_{10}$ are presented for this spectral range. The principal dn/dT are positive and show a polarization-anisotropy: $dn_p/dT = 6.4$, $dn_m/dT = 5.0$, $dn_g/dT = 5.6$ (10^{-6} K^{-1}) at wavelength of 0.83 μm. Thermo-optic dispersion data are modeled in accordance with theory taking into account the contributions from the change of the electronic band gap with temperature and from the volumetric thermal expansion coefficient.
Key words: orthorhombic crystal, thermo-optic coefficient, $BeAl_6O_{10}$ crystal, thermo-optic dispersion.

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ТЕРМООПТИЧЕСКИЕ СВОЙСТВА ОРТОМБИЧЕСКОГО КРИСТАЛЛА $BeAl_6O_{10}$

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Аннотация. Определены термооптические коэффициенты dn/dT для орторомбического кристалла $BeAl_6O_{10}$ для поляризаций света ***E*** || Np, Nm и Ng в диапазоне длин волн 0,4–1,1 мкм. Для этого спектрального диапазона получены формулы дисперсии главных термооптических коэффициентов $BeAl_6O_{10}$. Значения dn/dT положительны и характеризуются поляризационной анизотропией: $dn_p/dT = 6{,}4$, $dn_m/dT = 5{,}0$, $dn_g/dT = 5{,}6$ (10^{-6} K^{-1}) на длине волны 0,83 мкм. Данные по дисперсии термооптических коэффициентов моделируются в рамках модели, учитывающей вклады от изменения ширины электронной запрещенной зоны с температурой и от объемного коэффициента теплового расширения.
Ключевые слова: орторомбический кристалл, термооптический коэффициент, кристалл $BeAl_6O_{10}$, дисперсия термооптического коэффициента.

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Beryllium hexaaluminate $BeAl_6O_{10}$ crystal is a promising laser matrix for various transition metal ions such as Cr^{3+}, Ti^{3+}, Ni^{2+}. The $BeAl_6O_{10}$:Cr^{3+} (Ti^{3+}, Ni^{2+}) crystals are promising for the use as active media for tunable solid state lasers in the spectral regions of 0.7–1.0 μm (Cr^{3+}), 0.6–1.1 μm (Ti^{3+}), and 1.35–1.65 μm (Ni^{2+}) [1]. The first laser operation for $BeAl_6O_{10}$ crystal was achieved on Cr^{3+} ions [2]. $BeAl_6O_{10}$ crystals doped with Cr^{3+} and Ti^{3+} ions are promising media for the development of femtosecond lasers [1]. However, thermo-optic properties of $BeAl_6O_{10}$ (optically biaxial) crystal have not been studied in detail to date.

In the present work, we report on a measurements of the thermo-optic coefficients (TOCs, dn/dT) of hexaaluminate crystal $BeAl_6O_{10}$.

Beryllium hexaaluminate $BeAl_6O_{10}$ is grown by the Czochralski technique. The $BeAl_6O_{10}$ is an orthorhombic crystal with space group *Pcam*, and thus is optically biaxial one. Optical properties of the $BeAl_6O_{10}$ are described within the frame of the optical indicatrix with the orthogonal principal exes N_p, N_m, and N_g. For $BeAl_6O_{10}$, these three axes are related to the crystallographic axes as follows: N_p//***c***, N_m//***a*** и N_g//***b*** [3]. Like the three principal refractive indices, the $BeAl_6O_{10}$ crystal should be characterized by three principal TOCs, namely dn_p/dT, dn_m/dT and dn_g/dT.

To determine the TOCs, the sample from the $BeAl_6O_{10}$ is prepared to be rectangular parallelepiped with dimensions $10\times10\times10 mm^3$. The edges of the parallelepiped are parallel to the optical indicatrix (crystallographic) axes N_p(***c***), N_m(***a***), N_g(***b***). All six surfaces of the sample were of a lasergrade quality.

For the determinations of the TOCs, the laser beam deviation technique for a material with a linear thermal gradient is used. Experimental setup and measurement procedure can be found elsewhere [4]. The measurements are performed at seven wavelengths by means of laser diodes (405 nm, 450 nm, 650 nm, and 800 nm), He-Ne laser (633 nm), and microchip diode-pumped Nd:YAG laser with frequency doubling (532 nm and 1064 nm). The laser beam deviation method allows one to measure the so-called thermal coefficients of the optical path

(TCOP), $W=dn/dT+(n-1)\mu$, where μ is the linear thermal expansion coefficient in the direction of light propagation $\boldsymbol{k}$, n and dn/dT are the refractive index and TOC for corresponding light wavelength λ and polarization $\boldsymbol{E}$. The precision of the TCOP measurements is ~10%.

In order to extract the dn/dT value from the TCOP, literature data on the refractive index for $BeAl_6O_{10}$ is used [3]. The principal linear thermal expansion coefficients $\mu_{a(m)}$, $\mu_{b(g)}$, and $\mu_{c(p)}$ are measured by means of Horizontal pushrod dilatometer Netzsch DIL 402 PC to be $\mu_{a(m)} = 6.5$, $\mu_{b(g)} = 6.9$, and $\mu_{c(p)} = 7.1$ (10^{-6} K^{-1}). The value of $\mu_{a(m)}$ obtained in present paper are in good accordance with the value of $\mu_a = 6.8\cdot10^{-6}$ K^{-1} from [5].

For each light polarization $\boldsymbol{E}$, two values of dn/dT are determined corresponding to two different directions of light propagation $\boldsymbol{k}$ by the formula $dn_i/dT = W_{ij} - (n_i - 1)\,\mu_j$,, where i = p, m, g is the light polarization index, j = p, m, g is the index of the direction of light propagation, and $i \neq j$. Principal TOCs dn_p/dT, dn_m/dT, and dn_g/dT are defined as the average over the corresponding values for the two directions of light propagation. The results are presented in the Table 1. All three principal TOCs for $BeAl_6O_{10}$ are positive. For the whole studied spectral range, anisotropy of the TOCs characterized by $dn_p/dT > dn_g/dT > dn_m/dT$. The value of $dn_{m(a)}/dT$ corresponds fairly well to the value of $dn_a/dT = 11.6\times10^{-6}$ K^{-1} measured in [5] for visible spectral region with the minimum deviation method. There is no previous data on the dn_p/dT and dn_g/dT.

Table 1. Principal thermo-optic coefficients dn/dT (10^{-6} K^{-1}) for $BeAl_6O_{10}$

Wavelength, nm	dn_p/dT	dn_m/dT	dn_g/dT
405	15,6	11.9	13.6
450	13.0	9.5	10.8
532	9,7	7.9	8.9
633	7,9	6.25	6.85
650	7,7	6.0	6.55
800	6.55	5.0	5.5
1064	5.75	4.55	5.1

Experimental data on the wavelength dependence (dispersion) of the dn/dT are modeled by phenomenological model [6] according to which dn/dT is a sum of electronic and lattice contributions, $dn/dT = (dn/dT)_e + (dn/dT)_L$, where

$$(dn/dT)_e = (n^2_\infty - 1)/[2n(\lambda)]\{-\mu_{vol}\lambda^2/(\lambda^2 - \lambda^2_g) - (2/E_g)dE_g/dT[\lambda^2/(\lambda^2 - \lambda^2_g)]^2\}. \quad (1)$$

Here, $n(\lambda)$ is the dispersion equation for refractive index, λ is the wavelength, n_∞ is the low frequency refractive index, μ_{vol} is the volumetric thermal expansion coefficient, E_g is the electronic bandgap, λ_g is the wavelength corresponding to E_g. For $BeAl_6O_{10}$, the lattice contribution $(dn/dT)_L$ is small compared with the electronic part $(dn/dT)_e$, and can be neglected. Thus, the dn/dT values are considered to be $dn/dT = (dn/dT)_e$.

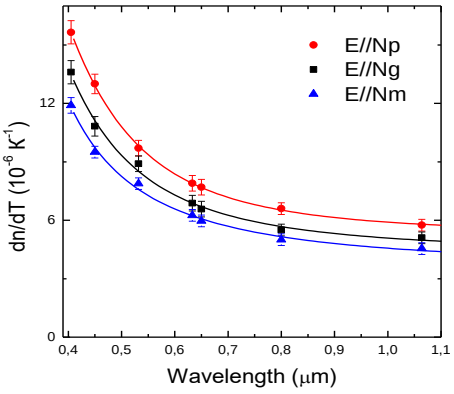

Figure 1 – Dispersion of principal thermo-optic coefficients dn/dT for $BeAl_6O_{10}$ crystal: symbols – experimental data, lines – modeling using equation Eq. (1)

It follows from the Eq. (1) that the dn/dT value is controlled by two factors. The first factor $(dn/dT)_\mu$ is the contribution from the volumetric thermal expansion coefficient and is negative because μ_{vol} is normally positive for optical materials [6]. The second factor $(dn/dT)_g$ is the contribution from the change of the electronic band gap with temperature and is normally positive because the value of dE_g/dT is normally negative for optical materials [6]. These two factors compete with one another giving positive or negative values of dn/dT.

The results of the TOCs dispersion simulation with Eq. (1) are presented in Fig. 1. The free fitting parameters are E_g and dE_g/dT. The best-fit parameters are 5.6 and –8.2 (for $\boldsymbol{E}$ // N_p), 6.0 and –8.2 ($\boldsymbol{E}$ // N_m), and 5.8 (eV) and –0.80 (10^{-5} eV/K) ($\boldsymbol{E}$ // N_g), respectively. For $BeAl_6O_{10}$, the electronic bandgap was estimated to be 5.5 eV for unpolarized light propagating along $N_p(\boldsymbol{c})$-axis [3] which is in agreement with the data obtained in this work. According to the Eq. (1), the positive dn/dT coefficients of $BeAl_6O_{10}$ are related to the weak thermal expansion, so that the contribution of the $(dn/dT)_g$ term is dominant over the $(dn/dT)_\mu$ one.

To calculate the dn/dT value at some arbitrary wavelength it is more convenient to use the analytical thermo-optic dispersion formulas which are obtained based on the principal thermo-optic coefficients given in Table 1. It is

$$dn_i/dT = A_i + B_i/\lambda^2 + C_i/\lambda^4 + D_i/\lambda^6 \ (10^{-6}\ \mathrm{K}^{-1}), \quad (2)$$

where $i = p, m, g$, λ is in μm, and A, B, C, D are constant parameters chosen to give the best agreement with experimental TOCs data (Table 2).