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Use of the Laser Beam Deflection Technique for Thermo-Optic Coefficients Study in Gadolinium-Yttrium Oxyorthosilicate Doped with Erbium Ions

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Abstract

Results of use of the laser beam deflection technique for determination of thermo-optic coefficients (TOCs) of the Er^{3+}-doped gadolinium-yttrium oxyorthosilicate crystal (Er^{3+}:$(GdY)_2SiO_5$ – Er:GYSO) are presented. A 0.1 at.% Er-doped gadolinium-yttrium oxyorthosilicate crystal was grown by the Czochralski method under nitrogen atmosphere. Raw materials such as Er_2O_3, Gd_2O_3, Y_2O_3, and SiO_2 were weighed according to the formula $(Er_{0.001}Gd_{0.8995}Y_{0.0995})_2SiO_5$. Optical properties of the biaxial Er:GYSO crystal are described within the frame of the optical indicatrix with orthogonal principal axes N_p, N_m, and N_g. To characterize the anisotropy of the TOCs a sample from the grown Er:GYSO crystal was prepared in a shape of a rectangular parallelepiped with dimensions of 7.0 (N_p)×8.0 (N_m)×8.5 (N_g) mm^3. Each face of the sample is perpendicular to one of the optical indicatrix axes N_p, N_m and N_g. For determination of the TOCs the laser beam deflection technique for a material with a linear temperature gradient is used. Measurements are performed at the wavelength of 632.8 nm. The thermal coefficient of the optical path (TCOP) for the Er:GYSO crystal measured at the wavelength of 632.8 nm at different light polarization $\boldsymbol{E}$ and propagation direction $\boldsymbol{k}$ were obtained. The TCOP values are positive for all directions of the light propagation $\boldsymbol{k}$ // N_p, N_m, N_g. This means that the sign of the thermal lens which is directly related to the TCOP value will also be positive, and the positive thermal lens is then expected for N_p- N_m-, and N_g-cut Er:GYSO. Applying an analysis of the thermal lensing the dn/dT value for Yb:GYSO is estimated to be 6.5×10^{-6} K^{-1}.

Keywords: beam deflection technique, thermo-optic coefficient, Er^{3+} ions, Gadolinium-Yttrium Oxyortosilicate crystal, thermal lens

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Применение метода отклонения лазерного пучка для определения термооптических коэффициентов в кристалле гадолиний-иттриевого ортосиликата, легированного ионами эрбия

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В статье представлены результаты использования метода отклонения лазерного пучка для определения величин термооптических коэффициентов (ТОК) в кристалле гадолиний-иттриевого ортосиликата, легированного ионами эрбия Er^{3+} (Er^{3+}:$(GdY)_2SiO_5$ – Er:GYSO). Кристалл Er:GYSO, легированный ионами эрбия в количестве 0,1 ат.%, выращен методом Чохральского в атмосфере азота. Шихта состояла из оксидов Er_2O_3, Gd_2O_3, Y_2O_3 и SiO_2 в пропорции, соответствующей формуле $(Er_{0.001}Gd_{0.8995}Y_{0.0995})_2SiO_5$. Оптические свойства кристалла Er:GYSO описываются на основе оптической индикатрисы с тремя ортогональными главными осями N_p, N_m и N_g. Для характеризации ТОК использовался образец кристалла Er:GYSO в форме прямоугольного параллелепипеда размером 7,0 (N_p)×8,0 (N_m)×8,5 (N_g) мм^3. Грани образца перпендикулярны осям оптической индикатрисы N_p, N_m и N_g. Метод отклонения лазерного пучка в результате распространения через исследуемый материал, в котором создан линейный градиент температуры, использован для определения ТОК. Измерения проведены на длине волны 632,8 нм. Установлены также термические коэффициенты оптического пути (ТКОП) для кристалла Er:GYSO на длине волны 632,8 нм для различных поляризаций света $\boldsymbol{E}$ и волнового вектора $\boldsymbol{k}$. Величины ТКОП являются положительными для всех направлений распространения света $\boldsymbol{k}//N_p$, N_m, N_g. Это означает, что знак термической линзы, которая непосредственно связана с величиной ТКОП, будет также положительным и, следовательно, положительная термическая линза будет наблюдаться в кристалле Er:GYSO, вырезанном вдоль направлений N_p, N_m и N_g. Из анализа значений термической линзы величина dn/dT в кристалле Yb:GYSO оценена как $6{,}5\times10^{-6}$ K^{-1}.

Ключевые слова: метод отклонения лазерного пучка, термооптический коэффициент, ионы Er^{3+}, кристалл гадолиний-иттриевого ортосиликата, термическая линза

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Introduction

Yttrium and gadolinium oxyorthosilicates [Y_2SiO_5 (YSO) and Gd_2SiO_5 (GSO)] are used as laser crystal hosts for doping with trivalent lasing rare-earth ions (RE^{3+}) such as Dy^{3+} [1], Nd^{3+} [2, 3], Yb^{3+} [4–11], Er^{3+} [12, 13], Tm^{3+} [14–16]. Three level dopants in the YSO and GSO crystals have a large ground state Stark splitting (the overall crystal field splitting of a ground multiplet is 922 cm^{-1} for Dy^{3+}:GSO [1], 472 cm^{-1} for Nd^{3+}:YSO [2], 985 cm^{-1} for Yb^{3+}:YSO [5], 1076 cm^{-1} for Yb^{3+}:GSO [5], 1000 cm^{-1} for Tm^{3+}:YSO [14]) which is higher than in other RE^{3+}-doped laser crystals. The high-energy components of the Stark level splitting cause the thermal population of the upper laser level to be low, and the laser system approaches the operating state of a four-level laser system. As a result, the lasing threshold is significantly reduced, and the emission cross section has a wide band, which is essential for producing a broad tuning wavelength range as well as ultrashort pulses of femtosecond duration. A continuous tunability from 1009 to 1112 nm and from 1025 to 1091 nm has been realized for continuous-wave Yb^{3+}:GSO [9] and Yb^{3+}:YSO [10] lasers, respectively. Passively mode-locked laser pulses as short as 122 fs and 343 fs have been demonstrated with Yb^{3+}:YSO [17] and Yb^{3+}:GSO [18] crystals, respectively, using a semiconductor saturable absorption mirror (SESAM).

The crystalline structure of the GSO and YSO is monoclinic being optically biaxial crystals, but the space group depends on their atomic constituents. The GSO having large Gd^{3+} ionic radius (0.94 Å) crystallizes in the monoclinic $P2_1/c$ space group [19]. This crystalline structure is characterized by perfect splitting of the crystal along the (100) plane, which makes it difficult for mechanical crystal processing. The YSO with a small Y^{3+} ionic radius (0.9 Å) crystallizes in the monoclinic C2/c space group [19]. This crystalline structure is less prone to cleavage, there is no clearly visible cleavage plane. Yb:GSO possesses a larger thermal conductivity of 4.9 $Wm^{-1} K^{-1}$ than that of Yb:YSO (4.4 $Wm^{-1} K^{-1}$ [20]) which reduces the thermal load during the laser operation. Furthermore, Yb:GSO has higher ground state Stark splitting 1076 cm^{-1} than that of Yb:YSO (985 cm^{-1}) [5], but suffers from problems with cleavage. Mixed $(Gd_{1-x}Y_x)_2SiO_5$ (GYSO) crystals have been introduced to eliminate cleavage of the GSO and to combine the benefits of the ground state splitting of the GSO with good mechanical properties of YSO (Nd^{3+}:GYSO [21–24], Yb^{3+}:GYSO [25–29]). A continuous tunability from 1004 to 1110 nm has been realized for a continuous-wave Yb^{3+}:GYSO laser [28]. A passively mode-locked Nd^{3+}:GYSO laser has been demonstrated using a SESAM with the pulse width of 5 ps [24]. Using Yb:GYSO crystals, mode-locked laser pulses as short as 55 fs have been demonstrated by Kerr-Lens mode-locking [28] and 210 fs pulses using a SESAM [27]. However, anisotropy of thermo-optic coefficients of the GYSO gadolinium-yttrium oxyorthosilicates (optically biaxial) crystals has not been studied in detail to date. These parameters are important for application of these crystals as laser crystal hosts.

In the present work, we report on the measurements of the thermal coefficients of the optical path (TCOP) and characterizations of anisotropy of thermo-optic coefficients (TOCs, dn/dT) of the gadolinium-yttrium oxyorthosilicate crystal doped with Er^{3+} ions.

Experimental

A 0.1 at.% Er-doped gadolinium-yttrium oxyorthosilicate crystal (Er:GYSO) was grown by the Czochralski method using iridium crucibles under nitrogen atmosphere. Raw materials such as Er_2O_3, Gd_2O_3, Y_2O_3, and SiO_2 were weighed according to the formula $(Er_{0.001}Gd_{0.8995}Y_{0.0995})_2SiO_5$. Thus, in the crystal under study, approximately 10 % of Gd^{3+} ions are replaced by Y^{3+} ions. The pulling speed was 1.5–2 mm/h and the speed of rotation was 20 rpm. The growth direction was [100]. The Er:GYSO boule was up to 55 mm in length and 19–24 mm in diameter (Figure 1). The crystal structure was analyzed by X-ray diffraction (XRD). The results reveal the as-grown Er:GYSO crystal has a primitive monoclinic structure with a space group of $P2_1/c$. This agrees with the fact that the space group $P2_1/c$ in the GSO matrix retains at 20 % substitution of Gd^{3+} ions by Y^{3+} ions [19].

Optical properties of the biaxial Er:GYSO crystal are described within the frame of the optical indicatrix with orthogonal principal axes N_p, N_m, and N_g, which are located in such a way that between the values of the corresponding principal indices of refraction (for the polarizations $E//N_p$, $E//N_m$, and $E//N_g$, respectively) the relation $n_p<n_m<n_g$ is fulfilled [30]. Like the three principal refractive indices, the Er:GYSO crystal should be characterized by the

three principal TOCs, namely, dn_p/dT, dn_m/dT and dn_g/dT. It should be noted that there is no predetermined relationship for dn_p/dT, dn_m/dT and dn_g/dT (as opposed to n_p, n_m, and n_g). For crystals belonging to the space group $P21/c$, the principal axis N_p coincides with the crystallographic axis $\boldsymbol{b}$ (= N_p), the other two principal axes N_m and N_g lie in the perpendicular plane [6].

Figure 1 – The as-grown 0.1 at.% Er:GYSO crystal

To characterize the anisotropy of the TOCs, a sample from the grown Er:GYSO crystal is prepared in a shape of a rectangular parallelepiped with dimensions of 7.0 (N_p)×8.0 (N_m)×8.5 (N_g) mm^3. Each face of the sample was perpendicular to one of the optical indicatrix axes N_p, N_m, and N_g. All six surfaces of the Er:GYSO sample are polished to a laser grade quality. The orientation of the sample was accomplished firstly identifying the $\boldsymbol{b}$ axis of the crystal by X-raying the crystal sample in a backscattered Laue geometry, with a precision of 0.2°. Preparation of the sample with faces normal to the $\boldsymbol{b}$ direction then allowed the remaining two principal axes to be found by identifying extinction directions (with a precision of 0.4°) when the sample was viewed in the $\boldsymbol{b}$ direction between crossed polarizers [31].

For determination of the TOCs, the laser beam deflection technique for a material with a linear temperature gradient is used [32]. This method is based on the registration of a deflection angle θ of the laser beam passing through the sample in the shape of a rectangular parallelepiped with creation of a linear temperature gradient in it. The laser beam is linearly polarized and has a flat wave front, and the linear temperature gradient is orthogonal to the beam propagation direction. A thermally induced stress in the sample which can modify a refractive index is avoided under a linear thermal gradient. In this way, by measuring the angle θ and the temperature gradient, it is possible to obtain the thermal coefficient of the optical path (TCOP), $W = dn/dT + (n–1)\alpha$, where α is a linear thermal expansion coefficient in the direction of light propagation $\boldsymbol{k}$, n and dn/dT are a refractive index and a thermo optic coefficient for the corresponding light wavelength λ and polarization $\boldsymbol{E}$. Then, using the refractive index for the corresponding wavelength and light polarization, and the linear thermal expansion coefficient one can derive the thermo optic coefficient as $dn/dT = W – (n–1)\alpha$. Similarly, one can find the corresponding linear thermal expansion coefficient as $\alpha = (W – dn/dT)/(n–1)$ using the known values of n and dn/dT. The experimental setup and the measurement procedure can be found elsewhere [33].

The measurements are performed at the wavelength of 632.8 nm by means of a continuous-wave He-Ne laser. A telescopic system and a diaphragm were used to reduce the divergence of the laser beam. A diameter and a divergence of laser beams were ≈2 mm (FWHM) and ≈1 mrad, respectively. The linear polarization of the beam was provided by a Glan-Taylor prism. The linear temperature gradient in the sample was ≈4 °C/mm. The actual temperature of hot and cold surfaces of the crystal sample was measured with a precision of 0.1 K.

The error in determining the TCOP is mainly due to the error Δθ in determining the deflection angle θ of the laser beam. To reduce the error Δθ, a beam profiler with a high spatial resolution (≈20 μm) was used to record the laser beam displacement, and the distance from the sample to the beam profiler was increased to ≈5 m. As a result, the uncertainty in the TCOP determination is ≈(0.3–0.4)×10^{-6} K^{-1}. When determining the dn/dT (or α) coefficient, an additional error arises which is associated with the error in determining the α (or dn/dT) value, and the accuracy of the dn/dT (or α) measurements increases to ≈(0.6–0.9)×10^{-6} K^{-1}.

As usual, the refractive indices were measured by the minimum deviation technique. The sample under study is in the form of a triangular prism in which a homogeneous temperature distribution is maintained. The refraction indices can be calculated by measuring the minimum deviation angle (the angle between the emergent beam and an undeviated beam) and the prism angle. If the studied material is isotropic, thermal expansion under uniform heating does not cause the change of the prism shape (prism apex angle) but changes only the prism dimensions. However, for anisotropic materials, the values of expansion coefficients are strongly dependent on the direction. This causes the prism apex angle to be

now temperature-dependent ones. Thus, for correct refractive index measurements, the change of the prism apex angle caused by anisotropic thermal expansion effect under uniform heating should be taken into account. In [34], expressions describing the dependence of the prism apex angle on temperature for different orientations of the prisms with respect to the optical indicatrix axes were obtained. It is possible to obtain the change of the prism apex angle if the corresponding thermal expansion coefficients are previously known. This approach taking into account the change of the prism apex angle caused by the anisotropic thermal expansion effect under uniform heating (in contrast to the conventional minimum deviation method which does not take this change into account) was called the modified minimum deviation method [34].

To determine the TOCs the interferometric technique has been also used. According to this technique, the expression used to calculate dn/dT also includes the corresponding values of refractive index and the linear thermal expansions (together with the directly measured change in fringe number over the change in temperature).

To give you an example, previously results of dn/dT for $KGd(WO_4)_2$ obtained by the interferometric technique ($dn_p/dT = -15.0$, $dn_m/dT = -10.0$, $dn_g/dT = -16.0$ (10^{-6} K^{-1}) [35]) are perfectly correspond to data obtained by the deflection technique ($dn_p/dT = -14.4$, $dn_m/dT = -9.7$, $dn_g/dT = -15.7$ (10^{-6} K^{-1}) [36]), and show accordance with the results obtained by modified minimum deviation method ($dn_p/dT = -10.6$, $dn_m/dT = -8.4$, $dn_g/dT = -15.2$ (10^{-6} K^{-1}) [34]), and are significantly different from the data obtained using the minimum deviation method (TOCs are different-signed: $dn_p/dT = -1.10$, $dn_m/dT = +6.5$, $dn_g/dT = -18.2$ (10^{-6} K^{-1}) [37]).

Thus, in the case of anisotropic crystals, the use of the interferometric technique, minimum deviation method, and deflection technique does not allow to measure TOCs directly (in contrast to the conventional minimum deviation method for isotropic media). It is necessary to calculate TOCs by appropriate formulas using the previously known refractive index and the expansion coefficients.

The advantage of the modified minimum deviation method (compared with the interferometric and beam deflection methods) is the possibility to combine the measurements of the refractive indices with TOCs. The drawbacks of the modified minimum deviation method are the complicated sample geometry (a prism) and high requirements to the homogeneity of the sample heating. The modified minimum deviation method and deflection technique are experimentally simpler compared to the interferometric technique, since the interferometric approach requires a much more complex experimental setup, and the interferometer-based setups are very sensitive to external influences and sample heterogeneities. The advantage of the deflection technique (compared with the interferometric and modified minimum deviation method) is a simpler and more reliable experimental setup, simpler sample geometry.

Results and discussion

The TCOP for the Er:GYSO crystal measured at the wavelength of 632.8 nm at different light polarization ***E*** and propagation direction ***k*** are presented in Table and Figure 2. All TCOP are positive. The values of W show polarization anisotropy in their absolute values which is most evident for the N_m-cut and N_g-cut crystals.

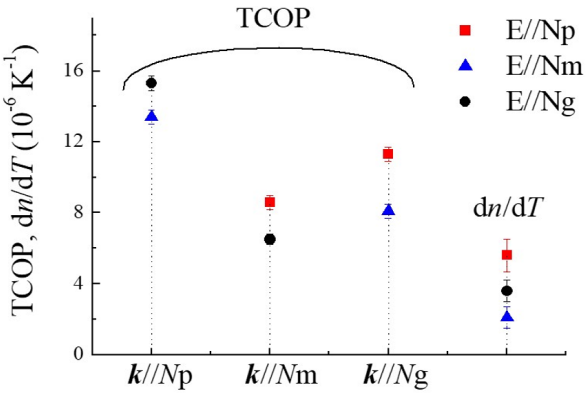

Figure 2 – Thermal coefficients of optical path (TOCP) and thermo optic coefficients (dn/dT) of Er:GYSO crystal for light polarizations $E//N_p$, N_m and N_g

To extract the dn/dT values from the TCOP, the formula $dn_i/dT = W_{ij} - (n_i - 1)\alpha_j$ should be used where $i = p, m, g$ is a light polarization index, $j = p, m, g$ is an index of the light propagation direction ***k*** ($i \neq j$). Therefore, literature data are needed on the refractive indices n_p, n_m and n_g and the linear thermal expansion coefficients α_p, α_m and α_g. At present, the values of n and α of Er:GYSO studied in this work are still unknown. We therefore used the refractive indices and the linear thermal expansion coefficient reported for GSO, namely $n_p = 1.871$, $n_m = 1.884$, $n_g = 1.910$ at the wavelength of 632.8 nm [38],

$\alpha_p = 12.8\times10^{-6}$ K^{-1} at the temperature of 100 °C [39]. It can be assumed that the *n*- and α-values do not change significantly when 10 % of Gd^{3+} ions in GSO are replaced by Y^{3+}ions, without changing the crystal structure. It may be also suggested that the refractive indices of the GYSO do not change significantly with Er doping because it is very small (only 0.001 part of the Gd and Y ions is replaced by the Er ones). For example, the refractive index contrast between double tungstates laser crystals doped with 1 at.% Er or Tm ions and the undoped ones is of the order of 10^{-3} [40, 41].

Table

Thermal coefficients of optical path (TCOP, 10^{-6} K^{-1}), thermo optic coefficients (dn/dT, 10^{-6} K^{-1}), and linear thermal expansion coefficients (α, 10^{-6} K^{-1}) of Er:GYSO for different light polarization *E* and propagation direction *k*

k //	TCOP			dn/dT			α_m	α_g
	E // N_p	***E*** // N_m	***E*** // N_g	***E*** // N_p	***E*** // N_m	***E*** // N_g		
N_p	–	13.4±0.4	15.3±0.4	–	2.1±0.6	3.6±0.6		
N_m	8.6±0.4	–	6.5±0.3	5.8±0.9	–		3.2±0.8	
N_g	11.3±0.4	8.1±0.4	–	5.4±0.9		–		6.8±0.8

Using the measured W_{mp} and W_{gp} the values of dn_m/dT and dn_g/dT are determined to be $(2.1\pm0.6)\times10^{-6}$ K^{-1} and $(3.6\pm0.6)\times10^{-6}$ K^{-1}, respectively. These values of dn_m/dT and dn_g/dT in combination with the measured W_{mg} and W_{gm} enable to find $\alpha_g = (6.8\pm0.8)\times10^{-6}$ K^{-1} and $\alpha_m = (3.2\pm0.8)\times10^{-6}$ K^{-1}. And finally, the obtained α_g- and α_m-values with the values of W_{pg} and W_{pm} make it possible to evaluate dn_p/d$T = (5.6\pm0.9)\times10^{-6}$ K^{-1} as an average over the values of $(5.4\pm0.9)\times10^{-6}$ K^{-1} (***k***//N_g) and $(5.8\pm0.9)\times10^{-6}$ K^{-1} (***k***//N_m) for two directions of the light propagation ***k***. The results on the obtained dn_p/dT, dn_m/dT, dn_g/dT, α_m and α_g are collected in Table 1 and Figure 2. Applying an analysis of the thermal lensing, the dn/dT value for Yb:GYSO was estimated as 6.5×10^{-6} K^{-1} [25]. However, the authors of [25] did not provide any information on the orientation, symmetry group, and stoichiometry of the Yb:GYSO crystal under study. There is no previous data on the dn/dT for GSO.

According to phenomenological model developed for frequencies between a fundamental lattice resonance and an electronic bandgap [42], the dn/dT value is controlled by two factors. The first factor $(dn/dT)_{tec}$ is the contribution from the volumetric thermal expansion coefficient and it is negative. The second factor $(dn/dT)_{bg}$ is the contribution from the change of the electronic bandgap with temperature and it is normally positive. These two factors compete with each other giving positive or negative values of dn/dT. Therefore, the positive dn/dT coefficients of the Er:GYSO crystal are due to the fact that the contribution of the $(dn/dT)_{bg}$ term is dominant over the $(dn/dT)_{tec}$ one.

The anisotropy of the TOCs is characterized by dn_p/d$T >$ dn_g/d$T >$ dn_m/dT. Thermal coefficients of the natural birefringence are $\Delta p = |dn_m/dT - dn_g/dT| = 1.5$ for ***k***//N_p (N_p-cut crystal), $\Delta m = |dn_p/dT - dn_g/dT| = 2.0$ for ***k***//N_m (N_m-cut crystal), and $\Delta g = |dn_p/dT - dn_m/dT| = 3.5$ (10^{-6} K^{-1}) for ***k***//N_g (N_g-cut crystal). The values of Δ are relatively small, the maximum variation occurs for the N_g-cut Er:GYSO. For comparison, the monoclinic YSO, $Ca_4YO(BO_3)_3$, $Ca_4GdO(BO_3)_3$ $KY(WO_4)_2$ have similar values of $\Delta p = 1.03$, 1.2, 1.1, 3.5; $\Delta m = 2.32$, 1.3, 0.1, 2.2 and $\Delta g = 3.35$, 2.5, 1.0, 5.7 (10^{-6} K^{-1}), respectively [36, 38, 43].

The TCOP values are positive for all directions of the light propagation ***k***//N_p, N_m, N_g. This means that the sign of the thermal lens, which is directly related to the TCOP value [44], will also be positive, and the positive thermal lens is then expected for N_p- N_m-, and N_g-cut Er:GYSO. The differences in the TCOP values for the same crystal cut and the orthogonal light polarizations (1.9, 2.1, 3.2 for p-, m-, and g-cut crystals, respectively) are close to the corresponding values of the thermal coefficients of the natural birefringence Δ.

The linear thermal expansion coefficients α_m and α_g for Er:GYSO obtained in this work (Table) are in agreement with the values of $\alpha_c = 5.6\times10^{-6}$ K^{-1} and $\alpha_{a^*} = 4.2\times10^{-6}$ K^{-1} reported for the GSO [39] (the *a** refers to direction which lies in the (***ac***) plane and is perpendicular to crystallographic axis ***c***).

Conclusion

Anisotropy of the thermal coefficients of the optical path TCOP and the thermo optic coefficients dn/dT of the 0.1 at.% Er:GYSO crystal (having space group of $P21/c$) has been studied at wavelength of 632.8 nm by means of the laser beam deflection technique. The principal thermo optic coefficients are obtained as $dn_p/dT = 5.6$, $dn_m/dT = 2.1$, $dn_g/dT = 3.6$ (10^{-6} K^{-1}). Using the measured values of dn/dT and TCOPs, the linear thermal expansion coefficients of Er:GYSO for directions along optical indicatrix axes N_m and N_g are estimated to be $\alpha_g = 6.8$ and $\alpha_m = 3.2$ (10^{-6} K^{-1}). Knowledge of the thermo-optical properties of the Er:GYSO crystal will be useful in designing laser cavities of high-power continuous wave and mode-locked oscillators based on this crystal as well as for GYSO doped with other lasing rare-earth ions.

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