

Optical properties of the langasite family crystals: $\text{La}_3\text{Ga}_5\text{SiO}_{14}$, $\text{La}_3\text{Ga}_{5.5}\text{Ta}_{0.5}\text{O}_{14}$, $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$

© E.V. Zabelina¹, N.S. Kozlova¹, O.A. Buzanov²

¹ National University of Science and Technology MISiS, Moscow, Russia

² JSC „FOMOS-MATERIALS“,
Moscow, Russia

e-mail: zabelina.ev@misis.ru

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Optical properties of lanthanum-gallium silicate ($\text{La}_3\text{Ga}_5\text{SiO}_{14}$), lanthanum-gallium tantalate ($\text{La}_3\text{Ga}_{5.5}\text{Ta}_{0.5}\text{O}_{14}$), calcium-tantalum-gallium silicate ($\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$) crystals in the near ultraviolet (UV) and visible wavelength ranges (200–800 nm) have been studied. For the first time the angles of deviation of the moments of the dipole transition spectral dependences $\theta(\lambda)$ were used to characterize the absorption processes in these crystals. These dependences make it possible to reveal absorption bands that are weakly noticeable on the transmission coefficients spectral dependences, and to estimate the degree of deviation of the dispersion dependence $\theta(\lambda)$ from the value $\theta = 45^\circ$ corresponding to an ideal ordered crystal. Analysis of $\theta(\lambda)$ dependencies showed that in the studied wavelength range, the lowest deviation from $\theta = 45^\circ$ is observed in $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$, and the highest in $\text{La}_3\text{Ga}_5\text{SiO}_{14}$. The coefficients of the specific rotation angle of the polarization plane ρ were obtained by the spectrophotometric interference method in polarized light. The measured values are best approximated by the Vyshin equation. For the first time, dependences of the modified Drude formula $1/\rho = f(\lambda^2)$ were calculated for these crystals. These dependences should be linear in case of an ideal crystal. The largest deviation from the linear dependence is observed for $\text{La}_3\text{Ga}_5\text{SiO}_{14}$ crystals, and the lowest for $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$. The discrete values of the refractive coefficients n_o and n_e were measured by the prism method. $\text{La}_3\text{Ga}_{5.5}\text{Ta}_{0.5}\text{O}_{14}$ crystals have the highest refractive coefficients, $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$ crystals have the lowest. The obtained values are approximated by the Sellmeier equation.

Keywords: lanthanum-gallium silicate, lanthanum-gallium tantalate, calcium-tantalum-gallium silicate, refractive index, transmission coefficients spectral dependences, polarization plane specific rotation, dipole moment of transition.

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Introduction

Crystals with the calcium-gallium germanate structure (CGG, $\text{Ca}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$) have been known since the 1980s, when this structural type was discovered in the USSR and the first crystals were synthesized [1–3]. The structural type of CGG can be represented by the chemical formula $\text{A}_3\text{BC}_3\text{D}_2\text{O}_{14}$, where A — site in the form of Thompson twisted cubes, B — octahedral site, and C and D — tetrahedral sites of different sizes. The complex three-sublattice [4,5] structure of this compound allows to include ions of different elements, which leads to a large number of synthesized modifications: by 2003 there were approximately 100 [6] such connections, and by 2018 — more than 200 [7]. Among crystals with the CGG structure, a family or group of langasite crystals can be singled out, which historically include primarily langasite (LGS, $\text{La}_3\text{Ga}_5\text{SiO}_{14}$) and langatate (LGT, $\text{La}_3\text{Ga}_{5.5}\text{Ta}_{0.5}\text{O}_{14}$) — two the most studied and currently used crystals, as well as catangasite (CTGS, $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$).

Occupation of sites by cations in the LGS, LGT, and CTGS structures is presented in Table 1.

In the LGS compound, the tetrahedral sites D are statistically occupied with silicon and gallium ions [8,9]; in CTGS, element ions occupy their intended sites [14] in the structure. Regarding the occupancy of sites in LGT, the information presented in the literature differs: in the paper [12] it is shown that all ions occupy their sites in the structure, and in the papers [8,13] it is shown that gallium ions can occupy not only tetrahedral sites, but also octahedral sites B. Due to the fact that sites are occupied, CTGS compounds are classified as ordered crystals, while LGS and LGT — are classified as disordered.

It is known [15–17] that the growth atmosphere has a significant effect on the optical properties of these crystals. In an oxygen-free atmosphere, colorless crystals are obtained, and in an oxygen-containing atmosphere — colored ones. It has been shown [18] that colorless crystals are characterized by greater homogeneity and a higher refraction index. However, such crystals are difficult to obtain. Specifying the nature of the defect structure will allow to control the processes of defect formation and obtain crystals with desired properties, the required quality and high homogeneity. One approach to studying the nature

Table 1. Occupation of sites by cations in the LGS, LGT, and CTGS structures

Stoichiometric formula	Parameter of lattice, nm		A (dodecahedron)	B (octahedron)	C (tetrahedron)	D (tetrahedron)	Source
	<i>a</i>	<i>c</i>					
$La_3Ga_5SiO_{14}$	0.81652	0.50958	La^{3+}	Ga^{3+}	Ga^{3+}	Si^{4+} Ga^{3+}	[5,8–11]
$La_3Ga_{5.5}Ta_{0.5}O_{14}$	0.82260	0.51207	La^{3+}	Ta^{5+}	Ga^{3+}	Ga^{3+}	[5,10–12]
				$Ta^{5+}Ga^{3+}$			[5,8,10,11,13]
$Ca_3TaGa_3Si_2O_{14}$	0.81056	0.49800	Ca^{2+}	Ta^{5+}	Ga^{3+}	Si^{4+}	[5,10,13,14]

of the defect structure in crystals is to study their optical properties.

The structural type of CGG is morphologically described by the point symmetry group 32 (third-order axis and three second-order axes perpendicular to it). Thus, the crystals belong to the trigonal crystal system, are optically anisotropic, uniaxial, are characterized by two main refraction indices N_o and N_e and the presence of dichroism — difference in the absorption of electromagnetic waves depending on their polarization. Due to the fact that the symmetry group 32 is a subgroup of the limit group ∞m , such crystals are optically active (gyrotropic); in particular, there is rotation of the light polarization plane in them: if linearly polarized light falls on such a crystal in a certain direction, then at the output its polarization plane rotates through a certain angle α [19].

The study results of the langasite family LGS, LGT, CTGS optical crystals are presented in a number of papers [2,17,20–40]. However, for the most part (except for papers [20,22,18,39,40]), the results of studies are given without taking into account the optical anisotropy of these crystals, in particular, without taking into account dichroism, which can lead to incorrect interpretation of experimental data. As part of the rotation of the polarization plane, only the values of the specific rotation and their dispersion dependences are presented in the literature, while the study of optical activity can provide information on the structure of crystals.

The aim of this paper was to study the optical properties of crystals of the langasite family LGS, LGT, CTGS, grown in an oxygen-containing atmosphere, taking into account optical anisotropy and using underutilized used methods for analyzing the results obtained.

Samples and methods of study

LGS, LGT, and CTGS crystals were grown at JSC „FOMOS-MATERIALS“ by the Czochralski method in Ir crucibles in an argon atmosphere with the addition of oxygen. All samples were transparent, did not contain visible inclusions, and there was no scattering in the beams of the He-Ne laser. Samples were prepared from these crystals in the form of prisms with base faces perpendicular to the 3rd

order axis, in the form of oriented cubes with face sizes on the order of $15 \times 15 \times 15$ mm, and in the form of polished plates 1.5 mm thick, cut perpendicular to the 2nd order axis.

The optical properties of the crystals were studied at the accredited laboratory of semiconductor materials and dielectrics „Single Crystals and Stock on their Base“ NUST „MISIS“ [41]. The spectral dependences of the $T(\lambda)$ transmittances were measured on an Agilent Technologies Cary-5000 spectrophotometer in polarized light, the polarizer is a Glan – Taylor prism. The specific rotation of the polarization plane ρ was measured during the propagation of light along the 3rd order axis by the direct method on the test complex IK EOE-1 at the wavelength of the He-Ne laser $\lambda = 630$ nm and by the spectrophotometric interference method [38,42–44] from the spectral dependences of the transmission coefficients in crossed polarizers.

For direct measurements ρ , the crystal is placed in an optical system, the main components of which are a light source with a known wavelength, two polarizers and a photodetector. When the polarizers are crossed in the case of a gyrotropic crystal, complete extinction is not observed. Next, it is essential to rotate the polarizer until there is a complete extinction of light α . The specific rotation of the polarization plane ρ is calculated by dividing the resulting extinction angle α by the sample thickness d [19]:

$$\rho = \alpha/d \quad (1)$$

When carrying out such measurements, samples of different thicknesses are required since the polarization plane can rotate by more than 180° .

When using the spectrophotometric method [38,42–44] the gyrotropic sample is placed between crossed or parallel polarizers. The intensity of light passing through such a system will be equal to

$$I_{\perp} = I_0 d \sin^2 \rho, \quad I_{\parallel} = I_0 d \cos^2 \rho, \quad (2)$$

where I_0 — intensity of incident light; I_{\parallel} — intensity of light transmitted through the system with parallel polarizers; I_{\perp} — intensity of light transmitted through the system with crossed polarizers. The spectral dependences of the transmission coefficients $T(\lambda)$ obtained under conditions of

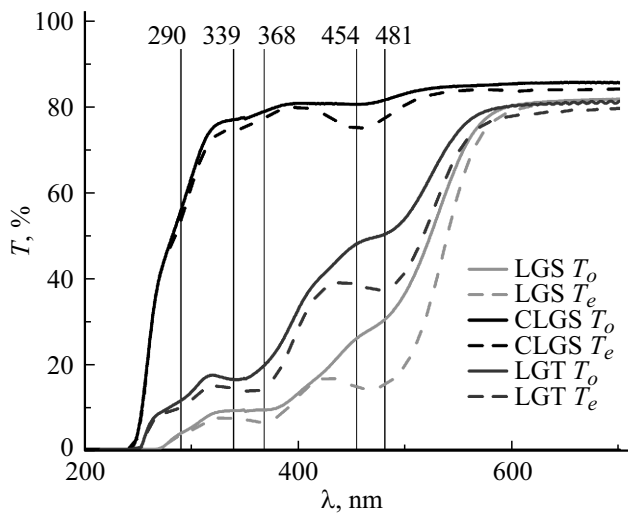


Figure 1. Spectral dependences of transmittances of LGS, LGT, and CTGS samples in polarized light.

crossed or parallel polarizers have a periodic character, on which the minima I_{\parallel} and the maxima I_{\perp} correspond to the value $\rho d = \pi/2 + \pi k$, and the maxima I_{\parallel} and minima I_{\perp} correspond to the value $\rho d = \pi + \pi k$, where k — an integer. The refraction indices were measured by the prism goniometric method on a GS-2 goniometer-spectrometer. The obtained experimental results were processed using the Origin software package.

Results and their discussion

The samples were yellow and orange, with the intensity of color being the brightest for LGS crystals and the least bright — for CTGS crystals. When observed in polarized light on all samples in the form of oriented cubes, the color was least pronounced when light propagated along the optical axis (3rd order axis). If polarized light propagated along the 2nd order axis, then the color intensity changed depending on the rotation of the sample around the direction of the light beam, which is a visible manifestation of dichroism. For LGS, LGT, CTGS crystals, the brightest color was observed when the oscillations of polarized light were parallel to the optical axis, and the brightest color was observed when the oscillations of polarized light were perpendicular to the optical axis. The spectral dependences of the transmittances of the samples were measured in polarized light when light passed through the samples in the form of plates, the working faces of which are perpendicular to the 2nd order axis. The resulting spectral dependences of the transmission of ordinary (T_o) and extraordinary (T_e) waves are shown in Fig. 1.

The intensity of the absorption of the extraordinary wave is stronger for all samples due to the phenomenon of dichroism and is most pronounced in the area of absorption bands in the visible range of the spectrum.

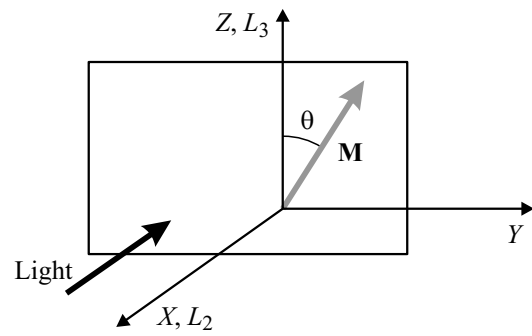


Figure 2. Coordinate system in a sample.

Therefore, for a correct interpretation of the experimental data, the measurement of the spectral dependences of the transmission and absorption of crystals of the langasite group should be carried out in the light of dichroism. There are pronounced absorption bands are observed at $\lambda_{\max} \sim 290$ nm, in the area of $\lambda_{\max} \sim 360$ – 370 nm and $\lambda_{\max} \sim 460$ – 480 nm in the near UV and visible areas of the spectrum for all crystals. The obtained results are in good agreement with the results presented in [22,25,32]. The nature of the first two bands has not been specified, the third band in the $\lambda_{\max} \sim 460$ – 480 nm area is associated with the presence of F -centers [18] in the crystals.

As part of the quantum mechanical approach, the absorption of electromagnetic radiation by a medium is determined by the mutual arrangement of the electric field strength vector (electric vector) \mathbf{E} of radiation and the vectors of dipole moments of transitions \mathbf{M} of structure elements in the medium: light is most effectively absorbed in when the direction \mathbf{E} and the direction \mathbf{M} coincide [45]. Physically, the transition dipole moment reflects the ability of dipoles to absorb light quantum [46]. The transition dipole moment is characterized by the magnitude and orientation relative to the chosen coordinate system, which is given by the angle θ . The orientation of the vectors of the dipole moment θ [45] is of practical importance.

In our case, the samples in the form of plates are cut perpendicular to the 2nd order axis, the 3rd order axis lies in the plane of the samples. Let us introduce an orthogonal coordinate system in which the Z axis is parallel to the 3rd order axis, and the X axis is parallel to the 2nd order axis (Fig. 2). Light propagates along the 2nd order axis (axis X) perpendicular to the working faces of the sample.

To determine the orientation of the transition dipole moment vectors, linear dichroism parameters are selected that depend only on the direction of the vector \mathbf{M} , but not on its magnitude. As such parameters, the dichroic ratio R and the degree of dichroism P [45] are used:

$$R = T_e/T_o, \quad P = \frac{T_e - T_o}{T_e + T_o}. \quad (3)$$

If the electric vector \mathbf{E} of the incident wave oscillates parallel to the 3rd order axis (along the Z axis), then for

each molecule the probability of light absorption will be proportional to $\cos^2\theta$, where θ — the angle between the 3rd order axis and transition dipole moment \mathbf{M} , and the relations for R and P can be written [45] as

$$R = \frac{T_e}{T_o} = \frac{\cos^2\theta}{\sin^2\theta} = \text{ctg}^2\theta, \tag{4}$$

$$P = \frac{\cos^2\theta - \sin^2\theta}{\cos^2\theta + \sin^2\theta} = \cos^2\theta. \tag{5}$$

Using relations (4), (5), the angle θ can be determined [45] by the formula

$$\theta = \text{arcctg} \sqrt{R} = \frac{1}{2} \arccos P. \tag{6}$$

If the vector \mathbf{M} is parallel to the 3rd order axis (axis Z), then $\theta = 0^\circ$, T_e will be the maximum, and $T_o = 0$. If vector \mathbf{M} is parallel to axis Y , then $\theta = 90^\circ$, T_o will be maximum and $T_e = 0$. If $T_e = T_o$, then $\cos^2\theta = \sin^2\theta$, i.e. at $\theta = 45^\circ$ the crystal is perfectly ordered, with no dichroism observed [45].

According to the measured dependences T_e and T_o , the angle θ was calculated; spectral dependences θ are shown in Fig. 3. The peaks in the spectral dependences of θ are identical to the maxima of the absorption bands in the dependences of $T(\lambda)$, however, they are better resolved and simultaneously take into account the absorption of both ordinary and extraordinary waves. For all crystals, a band with a maximum at $\lambda_{\text{max}} \sim 315$ nm appeared, and for catangasite, two additional bands appeared with maxima at $\lambda_{\text{max}} \sim 607$ and $\lambda_{\text{max}} \sim 651$ nm. An analysis of the data presented in Fig. 3 indicates that there is the smallest deviation of the $\theta(\lambda)$ dependence from $\theta = 45^\circ$ for CTGS crystals: the largest deviation corresponds to $\theta(\lambda) = 46^\circ 1' 48''$ at the maximum of the absorption band

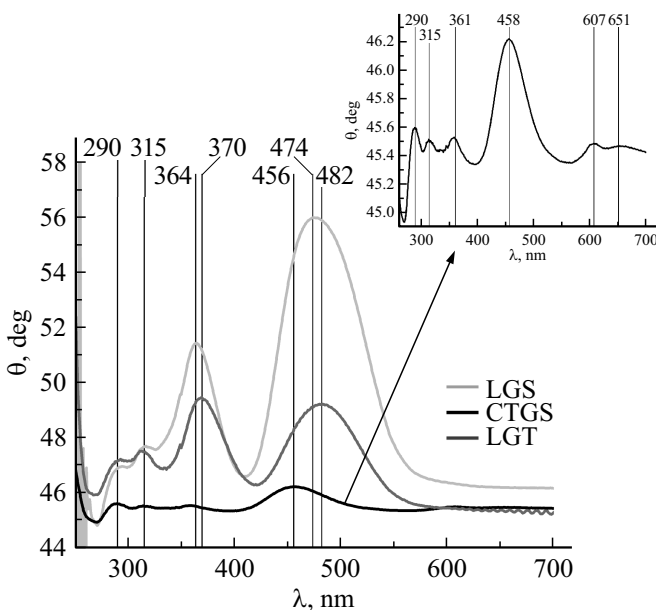


Figure 3. Spectral dependences of the transition dipole moment angles of LGS, LGT, CTGS crystals.

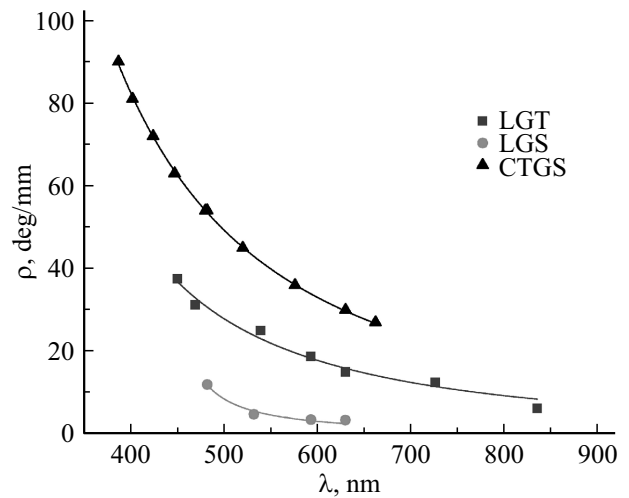


Figure 4. Dispersion dependences of the rotation angles of the polarization plane of LGS (circles), LGT (squares) and CTGS (triangles) crystals. The solid curves show the approximation dependences calculated by the Vyshin equation.

at ~ 458 nm. The largest deviation of $\theta(\lambda)$ from $\theta = 45^\circ$ is observed in the case of LGS crystals: the values of θ in the studied wavelength range exceed 46° , and at the maximum of the most pronounced absorption band in the area of 460–480 nm, the angle θ is $55^\circ 59' 11''$. Consequently, in the wavelength range under study, the highest \mathbf{M} vector disorder is observed in LGS crystals, and the smallest — in CTGS crystals.

The specific angle of rotation of the polarization plane was measured by two methods on samples in the form of cubes during the propagation of light along the threefold axis. The results are shown in Fig. 4. The obtained values of the specific rotation of the polarization plane of LGS, LGT, and CTGS crystals are in good agreement with those presented in the papers [38,42,43,47,48]. Optical effects in crystals are determined both by the symmetry or structure of the crystals and by the structure of the complexes forming the crystal [49,50]. Depending on what determines the rotational ability of a crystal (the structure of the complexes that form it or the structure of the crystal itself), the dispersion of optical activity is described [49] respectively by the Drude (7) or Chandrasekhar (8) formulas:

$$\rho = \frac{K_D}{(\lambda^2 - \lambda_D^2)}, \tag{7}$$

$$\rho = \frac{K_C \lambda^2}{(\lambda^2 - \lambda_C^2)^2}, \tag{8}$$

where K_D , K_C — constants; λ_D , λ_C — characteristic wavelengths of the bands.

In our case, to approximate the obtained values ρ , the Vyshin equation [49,51] is the best fit:

$$\rho = \frac{K_{V1} \lambda^2}{(\lambda^2 - \lambda_{V1}^2)^2} + \frac{K_{V2}}{(\lambda^2 - \lambda_{V2}^2)^2}, \tag{9}$$

Table 2. The constants of Vyshin approximation equation for the specific rotation of the polarization plane ρ

Crystal	K_{V1} , degree·nm	K_{V2} , degree·nm	λ_{V1} , nm	λ_{V2} , nm
LGS	1	533516.364	1	432.91261
LGT	1	$5.44021 \cdot 10^6$	1	234.56992
CTGS	1	$1.10263 \cdot 10^7$	1	162.57495

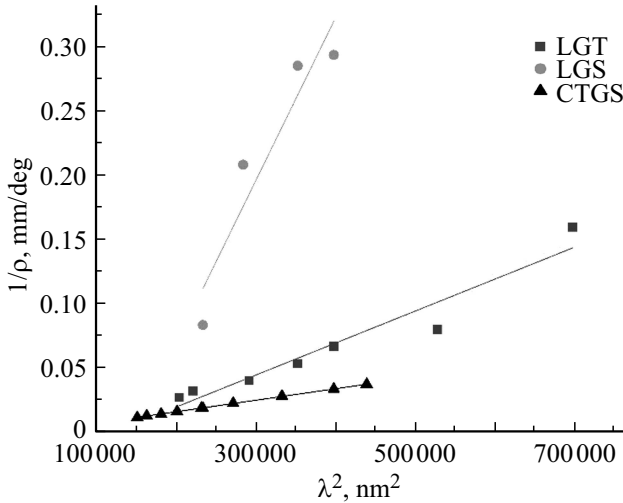


Figure 5. Dependences of the reverse specific rotation of the polarization plane on the squared wavelength of LGS (circles), LGT (squares) and CTGS (triangles) crystals. Solid curves show linear approximations.

where K_{V1} , K_{V2} — constants; λ_{V1} , λ_{V2} — characteristic wavelengths. The first term of equation (9) characterizes the optical activity arising due to the structure of the crystal, and the second term — due to the structure of the complexes that form the crystal. Consequently, in the case of crystals of the langasite family, both the structure of the crystal itself and the structure of the complexes contribute to the rotation of the polarization plane. The calculated values of the parameters according to equation (9) are presented in Table 2.

The study of the specific rotation of the polarization plane allows to estimate the structural perfection of crystals [50,52]. According to the Kisel theory [50,52] the dependence described by the modified Drude formula

$$1/\rho = A\lambda^2 + \lambda_0, \quad (10)$$

where A — constants, λ_0 — characteristic wavelength, should be linear in case of an ideal crystal. Deviations from linearity indicate disorder and the presence of defects in the crystal structure. The dependencies constructed in accordance with formula (10) and their linear approximations are shown in Fig. 5.

The residual sum of square σ , mm/degree, which characterizes the discrepancy between the experimental

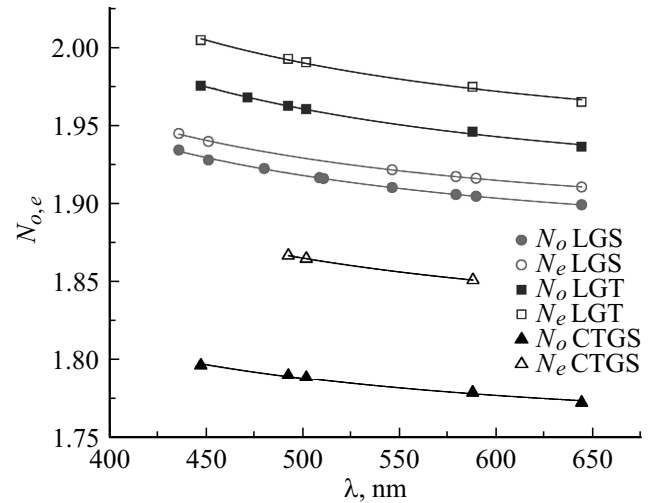


Figure 6. Dispersion dependences of the rotation angles of the polarization plane of LGS (circles), LGT (squares) and CTGS (triangles) crystals. The solid curves show the approximation dependences calculated using the Sellmeier equation.

data and the linear dependence, is $\sigma = 3.1 \cdot 10^{-3}$ for LGS, $\sigma = 8.4 \cdot 10^{-4}$ for LGT, and $\lambda = 1.5 \cdot 10^{-7}$ for CTGS. Therefore, the largest deviation from the linear dependence is observed for LGS crystals, and the smallest — for CTGS.

To fully describe the optical properties of the LGS, LGT, CTGS samples, we measured their refraction indices [53] by the prism method on a GS-2 goniometer-spectrometer. The results are shown in Fig. 6. The obtained refraction indices of LGS, LGT and CTGS crystals are in good agreement with those presented in the papers [2,54–56]. The highest refraction indices correspond to LGT crystals, the smallest to CTGS crystals.

Many equations are known for approximating the measured refraction indices [57]. However, as a rule [18,42], to approximate the refraction indices of crystals of the langasite family, the modified Sellmeier equation is used, which was used in this work,

$$N_{o,e}^2 - 1 = \frac{M_{o,e}\lambda^2}{\lambda^2 - \lambda_{o,e}^2}, \quad (11)$$

where $M_{o,e}$ — constant, $\lambda_{o,e}$ — characteristic wavelength. The constructed dispersion dependences are shown in Fig. 6, a the parameters of the Sellmeier equation — in Table 3.

The value of birefringence $\Delta = |N_o - N_e|$ was estimated at the standard wavelength of the yellow spectral line of sodium $\lambda_{Na} = 589.3$ nm and amounted to $\Delta_{LGS} = 0.0112$, $\Delta_{LGT} = 0.0295$, $\Delta_{CTGS} = 0.0772$. The results obtained indicate that CTGS crystals have the highest birefringence, and LGS — the lowest.

Table 3. Constants of the Sellmeier approximation equation for the refractive indices N_o and N_e

Parameters	LGS		LGT		CTGS	
	N_o	N_e	N_o	N_e	N_o	N_e
$M_{o,e}$	2.505	2.550	2.629	2.736	2.071	2.299
$\lambda_{o,e}$	127.2	125.6	137.5	137.5	119.6	133.9

Conclusion

Crystals $\text{La}_3\text{Ga}_5\text{SiO}_{14}$, $\text{La}_3\text{Ga}_{5.5}\text{Ta}_{0.5}\text{O}_{14}$, $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$ are grown in an argon-oxygen atmosphere. The optical properties of the crystals were studied in the near UV and visible areas of the spectrum 200–800 nm.

The spectral dependences of the transmittances in polarized light for ordinary and extraordinary waves — T_o and T_e , respectively, were obtained by spectrophotometry. There are pronounced absorption bands at $\lambda_{\max} \sim 290$ nm, in the area of $\lambda_{\max} \sim 360$ – 370 nm and $\lambda_{\max} \sim 460$ – 480 nm in the near UV and visible areas of the spectrum for all crystals. It is shown that at the maxima of the absorption bands, the absorption intensity of the extraordinary wave is greater than that of the ordinary wave.

Based on the measured spectral dependences T_o and T_e for the first time for these crystals, the dispersion dependences of the deviation angles of the dipole transition moments $\theta(\lambda)$ were calculated and plotted. It is shown that the construction of such dependences allows to reveal absorption bands that are weakly noticeable in the spectral dependences of the transmission and absorption coefficients and to estimate the degree of crystal ordering from the deviation of the value $\theta(\lambda)$ from $\theta = 45^\circ$ corresponding to an ideal ordered crystal. An analysis of the plotted dependences $\theta(\lambda)$ showed that in the studied range of the spectrum, the smallest deviation from $\theta = 45^\circ$ is characterized by $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$, and the largest — $\text{La}_3\text{Ga}_5\text{SiO}_{14}$.

Spectrophotometric interference method in polarized light obtained the coefficients of specific rotation of the plane of polarization ρ . The crystal $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$ is characterized by the largest value ρ , and the smallest — $\text{La}_3\text{Ga}_5\text{SiO}_{14}$. The obtained values are best approximated by the Vyshin equation. This indicates that both the symmetry of the crystal itself and the symmetry of the complexes that form them contribute to the rotational ability of these crystals. Dependences are constructed according to the modified Drude formula of the form $1/\rho = f(\lambda^2)$, which should be linear in the case of an ideal crystal. It is shown that the smallest deviation of this dependence from linearity is observed in the case of $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$, and the largest — if $\text{La}_3\text{Ga}_5\text{SiO}_{14}$.

The refraction index values N_o and N_e were measured by the prism method. All the studied crystals are optically positive. Crystals of $\text{La}_3\text{Ga}_{5.5}\text{Ta}_{0.5}\text{O}_{14}$ have the highest refraction indices, and the smallest — $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$

crystals. The obtained values are approximated by the Sellmeier equation. The value of birefringence was estimated at the standard wavelength of the yellow spectral line of sodium $\lambda_{\text{Na}} = 589.3$ nm and amounted to $\Delta_{\text{LGS}} = 0.0112$, $\Delta_{\text{LGT}} = 0.0295$, $\Delta_{\text{CTGS}} = 0.0772$. Crystals of $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$ are characterized by the highest birefringence, and crystals of $\text{La}_3\text{Ga}_5\text{SiO}_{14}$ are characterized by the smallest birefringence.

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Conflict of interest

The authors declare that they have no conflict of interest.

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