

# Dual-wavelength polarimeter application in investigations of the optical activity of langasite crystal

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**A method of high accuracy polarimetry, which includes optical activity measurement's systematic errors, was realized with dual-wavelength polarimeter for two wavelengths of 635 and 650 nm. Simultaneous measurement with neighboring wavelengths significantly improved the data processing, by increasing the amount of obtained data to eliminate the systematic errors. For langasite crystal  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  we measured temperature dependence of the gyration tensor component  $g_{11}$ . Our acquired value doesn't exceed  $0.47 \times 10^{-5}$  and is much smaller than previous results obtained by different experimental methods. Results presented in this paper are consistent with the calculated optical rotatory power from crystal structure data and polarizabilities of the atoms.**

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## 1. INTRODUCTION

High-accuracy universal polarimeter (HAUP) [1], which undergone several modifications and improvements over time [2–6], can be effectively applied to obtain information about main optical anisotropic parameters of crystals such as linear birefringence, circular birefringence (also known as optical rotation or optical activity), linear and circular dichroism. Recently, also Mueller matrix polarimetry was successfully applied for simultaneous measurement of main optical anisotropic crystal parameters [7–9]. First experimental data with this method for quartz and achiral  $\text{AgGaS}_2$  crystals shows good results.

Polarimeters allow measuring of optical activity (OA) for light propagation directions distinct from the optical axes, but results can significantly differ between themselves. The main cause of such discrepancies are the systematic errors. Quantitatively one can take them into account by considering parasitic ellipticities of polarizer and analyzer –  $p$  and  $q$  respectively. These ellipticities (normally of  $10^{-4}$  order) are usually comparable with eigen waves ellipticities  $k$  in crystals or sometimes they can be even bigger than  $k$ . Ellipticities are measured as a ratio of the minor to the major axes of the ellipse, which define the polarization state of two waves [10,11]. Therefore, measurement errors are significant. Experiments [1–5] showed that systematic errors should be estimated for each experimental process, because they depend also on specimen quality, system alignment, even on the precise profile of the laser beam passing through the specimen.

Measurement with different crystals keeping all the initial parameters constant lead to varying systematic errors.

An extended laser polarimeter, with similar design to HAUP, but with implementation of two wavelengths and different principles for data gathering and processing, was designed and was applied to investigate the OA of langasite crystal  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  (point group 32). Optical properties of these crystals are well studied [12], but precise value of OA was acquired along the optical axis only, which corresponds to the gyration tensor component  $g_{33}$ . The full detection of OA in uniaxial crystals of this point group requires measurement in direction perpendicular to the optical axis and calculation of the  $g_{11}$  gyration component. There are other uniaxial point groups ( $\bar{4}$  and  $\bar{4}2m$ ) of nonenantiomorphous (achiral) uniaxial crystals, which have null optical activity along the optical axis [10]. For example, the well-known KDP group and  $\text{AgGaS}_2$  are of such type of crystals.

OA is a vital parameter of crystal optical anisotropy and is heavily related to the structural peculiarities of the crystal. Therefore, we believe that it is important to find the experimental value of OA, by means of dual wavelength polarimeter and compare it with the results based on the classical polarization model [13].

## 2. PRINCIPLES OF EXPERIMENT

The polarimetric scheme used for studying of OA in birefringent sections is a polarizer-sample-analyzer (PSA), in which polarizer azimuth  $\theta$  and analyzer azimuth  $\chi$  are measured from the principal

crystal axes and are small ( $\theta, \chi \ll 1$ ). Both the polarizer and the analyzer are Glan type calcite prisms. The measuring procedure is fully automated with independent rotations of both polarizer and analyzer, controlled by the stepper motors. In order to measure small intensity changes, the so-called delta-sigma analog-to-digital converter with high resolution is used.

During the experiment the intensity  $I$  of transmitted through the PSA system light is measured as a function of angles  $I(\theta, \chi)$ . Intensity readings are typically collected for 15-20 values of polarizer azimuths,  $\theta$ , and for the same amount of analyzer azimuth,  $\chi$ , close to the position of minimum intensity. This data  $I(\chi)$  is then fitted to a biquadratic function using the method of linear least squares, which allows one to find the analyzer azimuth  $\chi_{\min}$  that corresponds to minimum transmission of polarization system, i.e.  $(\partial I / \partial \chi)_{\theta} = 0$ . For PSA system the corresponding relation for  $\chi_{\min}^{\text{PSA}}(\theta)$  may be expressed as [6,14]:

$$\chi_{\min}^{\text{PSA}} = \theta \cos \Gamma + (k - p) \sin \Gamma - \delta \chi, \quad (1)$$

where  $\Gamma = 2\pi \Delta n d / \lambda$  is the phase difference,  $d$  is the thickness of the specimen,  $\Delta n$  - linear birefringence and  $\lambda$  - wavelength of monochromatic light. One should also define angular systematic error  $\delta \chi$ . It is considered that most likely  $\delta \chi$  error is associated with mechanical elements of the polarimeter [4,5].

In a  $(\theta, \chi)$  coordinate system the intensity minima azimuths  $\chi_{\min}^{\text{PA}}(\theta)$  in the absence of a sample (PA system) form a straight line  $\chi_{\min}^{\text{PA}} = \theta$  with a slope angle of  $45^\circ$  (Fig. 1).

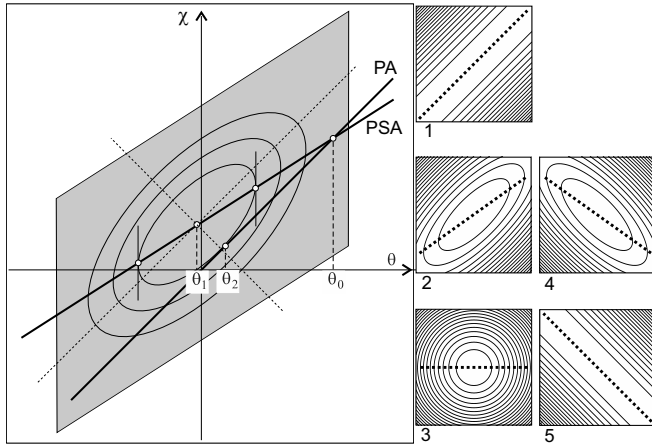


Fig. 1. Characteristic azimuths  $\theta_0, \theta_1, \theta_2$  for optically active birefringent crystal in  $(\theta, \chi)$  coordinate system. Scanning area of polarizer and analyzer in PSA system is shown as a grey parallelogram. Also the HAUP-maps schematic images for different phase differences are presented: 1 -  $\Gamma = 2\pi m$ , 2 -  $\Gamma = \pi/4 + 2\pi m$ , 3 -  $\Gamma = \pi/2 + 2\pi m$ , 4 -  $\Gamma = 3\pi/4 + 2\pi m$ , 5 -  $\Gamma = (2m + 1)\pi$ ;  $m$  - natural number.

In the PSA system, the intensity minima also form a straight line  $\chi_{\min}^{\text{PSA}}(\theta)$ , however according to the relation (1) the tangent of its slope angle is equal to  $\cos \Gamma$ . Thus, the optimum scanning area of the analyzer (when  $\chi_{\min}^{\text{PSA}}$  is in the middle) depends on the phase difference  $\Gamma$ . Fig. 1 shows the surface  $I(\theta, \chi)$  cross-sections with planes of constant intensities  $I(\theta, \chi) = \text{const}$ , which form the so-called HAUP maps in the shape of ellipses [2,15]. Major axes of these ellipses are

always tilted by  $45^\circ$  (see maps 1-5 on Fig. 1), but  $\chi_{\min}^{\text{PSA}}(\theta)$  tilt angle changes (dashed lines on maps 1-5).

On Fig. 1 the positions of three characteristic polarizer azimuths  $\theta_0$  (invariant azimuth, for which  $\chi_{\min}^{\text{PSA}}(\theta_0) = \chi_{\min}^{\text{PA}}(\theta_0)$ ),  $\theta_1$  (corresponds to the global minimum) and  $\theta_2$  (corresponds to the minimum light intensity with crossed polarizers) of the incident light in the PSA system are shown, and the principles of their definition are schematically represented. The relations for these azimuths can be expressed as [6,14]:

$$\theta_0 = (k - p) \cot(\Gamma/2) - \delta \chi / (1 - \cos \Gamma), \quad (2)$$

$$\theta_1 = (k - p) \cot \Gamma - (k + q) / \sin \Gamma, \quad (3)$$

$$\theta_2 = -(1/2)(p + q) \cot(\Gamma/2) - \delta \chi / 2. \quad (4)$$

The relation (4) is also used in the HAUP method [1-5].

Due to the unknown initial angles between crystallophysical axes and azimuth of the polarizer, exact measurement of angles  $\theta_0, \theta_1, \theta_2$  is not possible. Therefore, we analyze experimentally only their differences  $\Delta\theta_{01} = \theta_0 - \theta_1$ ,  $\Delta\theta_{02} = \theta_0 - \theta_2$  and  $\Delta\theta_{12} = \theta_1 - \theta_2$ . Relations between  $\theta_0$  and  $\theta_1$  can be derived from (2) - (4):

$$\Delta\theta_{01} \sin \Gamma = 2k - p + q - \delta \chi \cot(\Gamma/2). \quad (5)$$

It is easy to notice that

$$\Delta\theta_{01}(1 + \cos \Gamma) = 2\Delta\theta_{02}; \quad \Delta\theta_{01}(1 - \cos \Gamma) = -2\Delta\theta_{12}, \quad (6)$$

so determining  $\cos \Gamma$  and two of the characteristic azimuth  $\theta_0, \theta_1$  and  $\theta_2$ , or one of the differences  $\Delta\theta_{01}, \Delta\theta_{02}, \Delta\theta_{12}$  is sufficient to get complete data. However, permanent verification of equation (6) during the experiment gives us an additional evaluation criterion of correct measurement procedures. We should remember that precision of acquired  $\theta_0, \theta_1$  and  $\theta_2$  greatly depends on the phase difference  $\Gamma$ .

By using two sources of light with almost coinciding wavelengths  $\lambda_1$  and  $\lambda_2$ , we can neglect, in good approximation, the effects of  $k$  value dispersion and assume values  $p, q, \delta \chi$  to be constant. In this dual-wavelength polarimeter systematic errors can be differently eliminated. In particular, we can have a set of data for characteristic azimuths, which were measured in identical conditions but with alternating laser wavelengths. From this point of view the differences  $\Delta\theta_{01}, \Delta\theta_{02}, \Delta\theta_{12}$  for separately  $\lambda_1$  and  $\lambda_2$ , but also the differences  $\Delta\theta_{i\lambda} = \theta_i(\lambda_1) - \theta_i(\lambda_2)$ , ( $i = 0, 1, 2$ ) can be successfully analyzed. To increase the number of measured quantities we use two orientations of the crystal in the polarization system. It can be achieved by rotation of the sample  $90^\circ$  around the incident light, then the signs of  $\Gamma$  and  $k$  parameters are reversed [1-3]. Perpendicularity of the sample surface to the incident beam was ensured by adjusting the crystal position, so that the beam reflects back to the laser.

Using the relations (2)-(4) for characteristic azimuths, the differences  $\Delta\theta_{0\lambda}, \Delta\theta_{1\lambda}$  and  $\Delta\theta_{2\lambda}$  can be expressed as:

$$\Delta\theta_{0\lambda} = A_1(k - p) - B_1 \delta \chi, \quad (7)$$

$$\Delta\theta_{1\lambda} = A_2(k - p) - B_2(k + q), \quad (8)$$

$$\Delta\theta_{2\lambda} = -A_1(p + q) / 2. \quad (9)$$

Here we introduced the following notations:  $A_1 = \cot(\Gamma_1/2) - \cot(\Gamma_2/2)$ ,  $B_1 = (1 - \cos \Gamma_1)^{-1} - (1 - \cos \Gamma_2)^{-1}$ ,  $A_2 = \cot \Gamma_1 - \cot \Gamma_2$ ,  $B_2 = 1/\sin \Gamma_1 - 1/\sin \Gamma_2$ ,  $\Gamma_1 = \Gamma(\lambda_1)$  and  $\Gamma_2 = \Gamma(\lambda_2)$ . As a result, the number of equations which can be used to

calculate the eigen wave ellipticity  $k$  and eliminate the systematic errors is higher when compared to the previously used high-accuracy polarimetric methods.

### 3. RESULTS AND DISCUSSION

#### 3.1. Characteristic azimuths differences and systematic errors

The experiment was performed on (010)-plates of  $\text{La}_3\text{Ga}_5\text{SiO}_{15}$  crystal with 2.51 mm thickness in a temperature range from 290 to 370 K. In dual-wavelength laser polarimeter we used two semiconductor lasers with neighboring wavelengths of  $\lambda_1 = 635$  and  $\lambda_2 = 650$  nm. We consider that in equations (1)–(9) all quantities but  $\Gamma$  (Fig. 2), do not depend on the small wavelength change  $\delta\lambda = \lambda_2 - \lambda_1 = 15$  nm.

Because the ellipticity  $k$  of eigen waves does not depend on the specimen thickness, we chose it to be about 2.5 mm. With such thickness we achieved optimal change of phase difference  $\Delta\Gamma = 0.65$  in convenient temperature range of 290–370 K. The temperature of the sample was stabilized with precision of  $\pm 0.5$  K, which is enough, considering the slow change of birefringence  $\Delta n$  with temperature for langasite crystals.

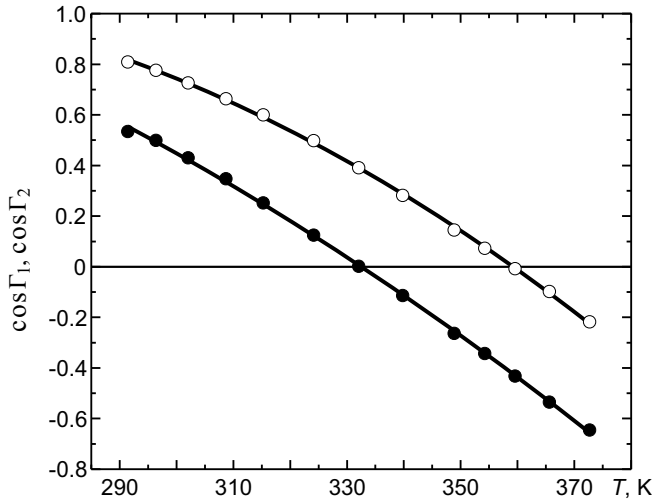


Fig. 2. Temperature dependences of  $\cos\Gamma_1$  for  $\lambda_1 = 635$  nm (○) and  $\cos\Gamma_2$  for  $\lambda_2 = 650$  nm (●) acquired from the tangent angle of linear dependences of  $\chi_{\min}^{\text{PSA}}(\theta)$  on (010)-plates of  $\text{La}_3\text{Ga}_5\text{SiO}_{15}$ .

Experimental temperature dependences of the characteristic azimuths differences  $\Delta\theta_{0\lambda}$ ,  $\Delta\theta_{1\lambda}$  and  $\Delta\theta_{2\lambda}$  for langasite crystal are shown on Fig. 3. As one can see, differences of the characteristic azimuths become too big compared to the typical value of  $k \approx 10^{-4}$ . With the considerable change of characteristic azimuths it is difficult to find the systematic errors  $p$ ,  $q$ ,  $\delta\chi$ .

We also did not observe contributions from multiple light reflections inside the crystal during our experiments, as, for example, did the authors of [16] during their experiments. If this effect would take place, then relations (6) would not execute correctly, and slope angles of the major axes of ellipses on the HAUP-maps (Fig. 1) would not be equal to  $45^\circ$ . This fact underlines once again the importance of experimental evaluation of the relations between azimuth differences  $\Delta\theta_{01}$ ,  $\Delta\theta_{02}$ ,  $\Delta\theta_{12}$ .

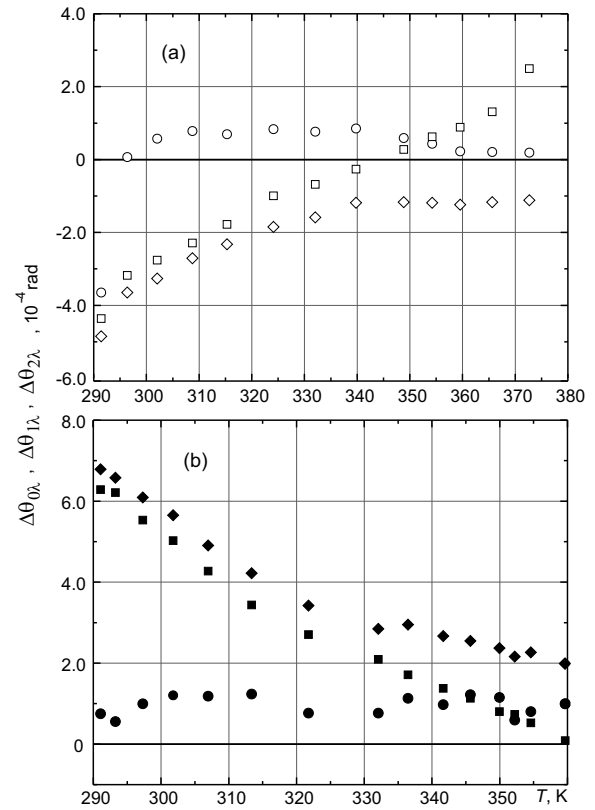


Fig. 3. Dependencies of characteristic azimuths differences  $\Delta\theta_{0\lambda}$  (○, ●),  $\Delta\theta_{1\lambda}$  (□, ■),  $\Delta\theta_{2\lambda}$  (◇, ◆) for LGS crystal on the temperature change for alternative crystal orientations as obtained before (a) and after (b)  $90^\circ$  rotation of the specimen around the light beam direction.

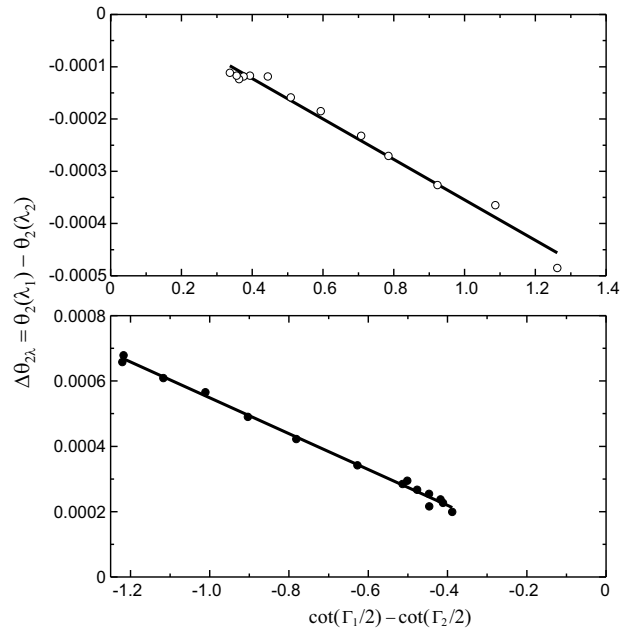


Fig. 4. Dependencies of characteristic azimuths  $\Delta\theta_{2\lambda}$  on  $\cot(\Gamma_1/2) - \cot(\Gamma_2/2)$  in LGS crystal for alternative crystal orientations as obtained before (○) and after (●)  $90^\circ$  rotation of the specimen around the light beam direction. Solid lines represent the best linear fit.

Consecutive analysis of the characteristic azimuths for two wavelengths in stable experimental conditions allows us to find the desired quantities. Fig. 4 shows that angles difference  $\Delta\theta_{2\lambda}$  does not depend on the crystal orientation. Sign change of the phase differences  $\Gamma_1, \Gamma_2$  and  $\Delta\theta_{2\lambda}$ , has also been taken into account because the sum of parasitic ellipticities of polarizer and analyzer  $p + q$  should remain unchanged. For alternative crystal orientations before ( $0^\circ$ ) and after  $90^\circ$  rotation of the specimen around the light beam direction, we could find (using equation (9)) that for  $0^\circ$  crystal setup  $p + q = (7.74 \pm 0.28) \times 10^{-4}$  (in radians) and for  $90^\circ$  crystal setup  $p + q = (11.0 \pm 0.2) \times 10^{-4}$ . The difference between these two values is insignificant, and x- and y-intercepts of the fitted lines are very close to their origin, as follows from the equation (9).

For further calculations we used the characteristic azimuths differences  $\Delta\theta_{1\lambda}$ . By adjusting the temperature of the crystal we acquired two experimental dependencies (8) for alternative crystal orientations ( $0^\circ$  and  $90^\circ$ ). Keeping in mind that after the rotation the eigen waves ellipticity  $k$  should change its sign, the sum of the respective characteristic azimuths differences becomes the following:

$$\Delta\theta_1^+ = \Delta\theta_{1\lambda}^0 + \Delta\theta_{1\lambda}^{90} = -2pA_2 - 2qB_2. \quad (10)$$

Afterwards, by plotting the temperature dependencies  $\Delta\theta_{1\lambda}^0/A_2$  and  $\Delta\theta_{1\lambda}^{90}/A_2$  (Fig. 5), we calculated the sum of these values  $\Delta\theta_1^+/A_2 = -2p - 2qB_2/A_2$ , as a result of addition of second degree polynomial fitted curves. Figure 5 also represents the temperature dependence of the  $B_2/A_2$  ratio values. This value doesn't change its sign after the rotation of the crystal and we get well matching experimental data for two crystal setups. Such fact confirms that  $90^\circ$  rotation procedure of the specimen around the light beam direction was very accurate.

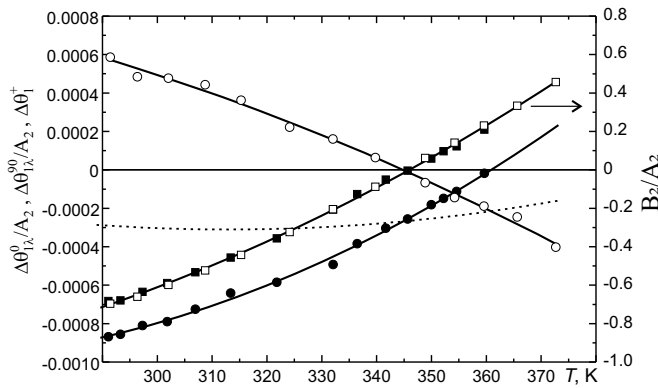


Fig. 5. The two plots of the parameters  $\Delta\theta_{1\lambda}^0/A_2$ ,  $\Delta\theta_{1\lambda}^{90}/A_2$  and their sum  $\Delta\theta_1^+/A_2$  (short dash line) versus temperature  $T$  of LGS crystal correspond to alternative crystal orientations before ( $\circ$ ) and after ( $\bullet$ )  $90^\circ$  rotation of the specimen. The points ( $\square$ ,  $\blacksquare$ ) for two crystal setups represent the temperature dependence of the  $B_2/A_2$  values, that changes sign at 346 K. Solid lines represent the best second degree polynomial fits.

For the data analysis it is important that at 346 K ratio  $B_2/A_2 = 0$  and we get just parasitic ellipticity of the polarizer  $p = (1.32 \pm 0.15) \times 10^{-4}$ , which, of course, does not depend on the temperature. Values of  $p$  and  $p + q$  are averaged for two wavelengths of 635 and 650 nm. So, we get two systematic errors, which are present

in equation (8). However, parasitic ellipticity  $q$  of the polarizer has two values for different orientations of the crystal  $q_0 = (6.42 \pm 0.28) \times 10^{-4}$  and  $q_{90} = (9.68 \pm 0.20) \times 10^{-4}$ . Difference in values can be explained by different conditions of light passing through the PSA system for two crystal positions. In further calculation we use mean value  $\bar{q} = (q_0 + q_{90})/2 = (8.05 \pm 0.28) \times 10^{-4}$ .

Using equation (7) we can find  $\Delta\theta_0^+ = \Delta\theta_{0\lambda}^0 + \Delta\theta_{0\lambda}^{90} = -2pA_1 - 2\delta\chi B_1$  and the angular error mean value  $\delta\chi = -(1.32 \pm 0.40) \times 10^{-4}$ . Let us note, that in dual-wavelength polarimeter determination of systematic error  $\delta\chi$  value is insignificant, while in standard HAUP methods, where this angular error is usually noted as  $\delta Y$ , its calculation is important for correct data processing [2-5, 14].

### 3.2. Optical activity of LGS crystal

Langasite family crystals belong to trigonal point group symmetry 32, space group P321, they are uniaxial and optically active. Along the optical axis  $g_{33}$  component of the gyration tensor is determined by measurement of the specific rotation  $\rho$ .

During the studies of the optical activity in a plate cut parallel to optical axis the gyration tensor component  $g_{11}$  is measured. For uniaxial crystals the relation between  $g_{11}$  and eigen wave ellipticity  $k$  is  $g_{11} = 2k\Delta n\bar{n}$  [10], where  $\bar{n}$  is the mean refractive index. For the mean wavelength of  $\lambda = 642.5$  nm  $\bar{n} = 1.905$  [17], and with channeled spectrum method [18] we acquired birefringence  $\Delta n = 0.0116$ . We also consider, that influence of the eigen waves ellipticity  $k$  dispersion is insignificant. For characteristic azimuths differences according to (8) we get:

$$\Delta\theta_1^- = \Delta\theta_{1\lambda}^0 - \Delta\theta_{1\lambda}^{90} = 2k(1 - B_2/A_2). \quad (11)$$

This allows finding  $k = (1.3 \pm 0.4) \times 10^{-4}$  for temperature 346 K. As one can see, the eigen waves ellipticity is significantly smaller than the parasitic ellipticities  $p$  and  $q$ , therefore calculation error of  $k$  is substantial and depends mostly on the experimental precision of characteristic azimuths and phase differences  $\Gamma$  for both wavelengths, which influence  $A_2$  and  $B_2$ .

Nevertheless, taking into account the systematic errors grants a chance of consecutive calculations of OA perpendicularly to the optical axis of the crystal. The temperature dependencies of  $g_{11}$  components of gyration tensor derived from two measurements of eigen waves ellipticity  $k$  are shown on Fig. 6. The presented results of calculations take into account equation (8), values  $p$  and  $q$ , and also temperature dependent mean refractive index and birefringence.

From the linear fitting of our data for temperature 295 K we get the value  $g_{11} = (0.47 \pm 0.17) \times 10^{-5}$ , which is much smaller than given in [17,19]. These values of OA were determined by the spectral measurement of the amplitude of oscillations of the rotation angle  $\chi$  of the major axis of the ellipse of passing light polarization. Such method assumes that polarizer is perfect (parasitic ellipticity  $p = 0$ ), is stationary during the experiment and its input azimuth should be  $\theta = 0$ . Then according to equation (1)  $\chi = k \sin \Gamma$  and  $\chi = \pm k$  for wavelengths for which  $\sin \Gamma = \pm 1$ . In [17] gyration tensor component  $g_{11} \approx 4.0 \times 10^{-5}$ , so the corresponding ellipticity  $k = 9.1 \times 10^{-4}$  for  $\lambda = 633$  nm. If this much larger ellipticity was real it would have prominently emerged on dependences shown in Fig. 3 and Fig. 5.



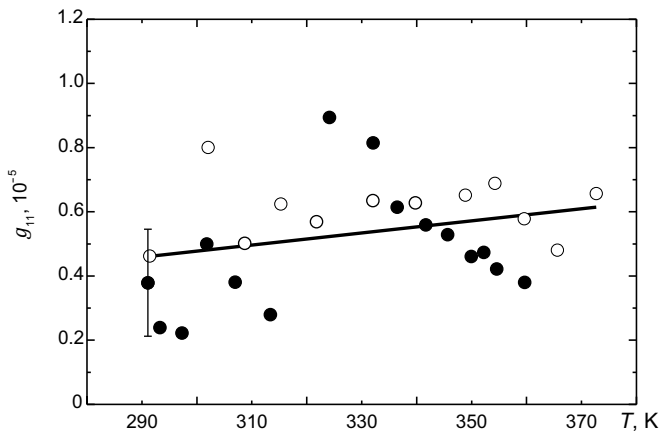


Fig. 6. The temperature dependencies of  $g_{11}$  components of the gyration tensor for the LGS crystal before ( $\circ$ ) and after ( $\bullet$ )  $90^\circ$  rotation of the sample. Error bar shows the goodness of linear fit.

Previous data [6] should also be treated critically, because the use of a reference crystal for measuring the parasitic ellipticity  $p$  often gives incorrect results. Our current method does not use the reference crystal, because earlier it was established that a measurement with different samples gives different values of parasitic errors [4,5]. Therefore, we used only rotation of the sample during the experiment and it allowed us to include systematic errors more accurately. Earlier results obtained for LGS crystal did not consider such errors and, in our opinion, this fact had negative impact on acquired in [6] results.

Using formally the relation  $\rho_{\perp} = \pi g_{11} / (\lambda n_e)$  for the specific rotation [10] perpendicular to the optic axis, we get  $\rho_{\perp} = 0.73$  deg/mm compared to  $\rho = -3.3$  deg/mm in direction of the optical axis [17].

The structure of langasite crystal does not contain screw axes. Its OA connected with helical formations of electron density, imitating the screw axis [20]. Therefore, in direction of the optical axis specific rotation in LGS is almost 6 times smaller than  $\alpha$  quartz  $\text{SiO}_2$ .

We would like to compare our results of polarimetric experimental data with calculated ones (the calculation method described in [13]). Using the same values of polarizability volumes [21] ( $\alpha_{\text{La}} = 1,886 \text{ \AA}^3$ ,  $\alpha_{\text{Ga}} = 0,375 \text{ \AA}^3$ ,  $\alpha_{\text{Si}} = 0,050 \text{ \AA}^3$ ,  $\alpha_{\text{O}} = 1,740 \text{ \AA}^3$ ) we obtained  $\rho_{\perp} = 1.1$  deg/mm. Unfortunately, this result is inaccurate, because  $\rho$  and  $\rho_{\perp}$  should be opposite in sign (according to gyrotropic properties of crystals with point group symmetry 32). Langasite crystals have the smallest OA among the langasite family [22]. With such crystal properties it is hard to expect theoretical calculations within the classical OA model to be accurate. Small changes of polarizability volumes influence the result significantly. One can only estimate the OA value, what we have accomplished. Nevertheless, we have attempted to verify theoretically whether our experimental results are correct.

In contrast, these calculating techniques were applied with success for pure  $\text{Ca}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$  crystals (langasite family) [23]. As a result, values of electronic polarizability volumes should be specified for studied LGS crystals. It is interesting to note, that some crystals with langasite structure show unusually large values of the specific rotation when compared to LGS [24].

Finally, it should be noted that almost all langasite family crystals are disordered, therefore, precise structure data and correct values of electronic polarizability volumes are necessary for consequent calculations.

#### 4. CONCLUSIONS

We used the extended HAUP polarimetric method by using two neighboring laser wavelengths and applied a new method for the

elimination of systematic errors in the measurement of optical activity in directions perpendicular to the optical axis of a crystal. Acquired value of eigen waves ellipticity  $k$  is significantly smaller than systematic errors, which are caused by polarizers imperfections. Nevertheless, we were able to find systematic errors and calculate temperature dependence of  $g_{11}$  component of the gyration tensor. The dual wavelength HAUP polarimetric method avoids the influence of angular errors, which play major perturbation role in the standard HAUP method. Optical activity of langasite crystals perpendicular to the optical axis turned out to be significantly lower than OA along the  $z$ -axis.

#### References

1. J. Kobayashi and Y. Uesu, "A new optical method and apparatus HAUP for measuring simultaneously optical activity and birefringence of crystals. I. Principles and construction", *J. Appl. Crystallogr.* **16**, 204–211 (1983).
2. J. Moxon and A. Renshaw, "Improved techniques for the simultaneous measurement of optical activity and circular dichroism in birefringent crystal sections", *Zeitschrift für Kristallogr.* **185**, 636–655 (1988).
3. E. Dijkstra, H. Meekes and M. Kremers, "The high-accuracy universal polarimeter", *J. Phys. D.* **24**, 1861–1868 (1991).
4. C.L. Folcia, J. Ortega, and J. Etxebarria, "Study of the systematic errors in the HAUP measurements", *J. Phys. D: Appl. Phys.* **32**, 2266–2277 (1999).
5. C. Hernández-Rodríguez, P. Gomez-Garrido, and S. Veintemillas, "Systematic errors in the high-accuracy universal polarimeter: application to the determining temperature-dependent optical anisotropy of KDC and KDP crystals", *J. Appl. Cryst.* **33**, 938–946 (2000).
6. Y. Shopa, and M. Kravchuk, "Study of optical activity in  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  with high-accuracy polarimetric methods", *Phys. Stat. Sol. A.* **158**, 275–280 (1996).
7. O. Arteaga, A. Canillas, and G. E. Jellison, "Determination of the components of the gyration tensor of quartz by oblique incidence transmission two-modulator generalized ellipsometry," *Appl. Opt.* **48**, 5307–5317 (2009).
8. O. Arteaga, J. Freudenthal and B. Kahr, "Reckoning electromagnetic principles with polarimetric measurements of anisotropic optically active crystals," *J. Appl. Cryst.* **45**, 279–291 (2012).
9. O. Arteaga, "Spectroscopic sensing of reflection optical activity in achiral  $\text{AgGaS}_2$ ," *Optics Letters*, **40**, 4277–4280 (2015).
10. J. F. Nye, *Physical Properties of Crystals: Their Representation by Tensors and Matrices* (Oxford U. Press, 1985).
11. A. Yariv and P. Yeh, *Optical Waves in Crystals: Propagation and Control of Laser Radiation* (Wiley-Interscience, New York, 2002).
12. W. T. Arkin, *New Research on Lasers and Electro-Optics* (New York: Nova Science Publishers, 2007).
13. V. Devarajan and A. Glazer, "Theory and computation of optical rotatory power in inorganic crystals", *Acta. Cryst. A.* **42**, 560–569 (1986).
14. Y. Shopa, "High accuracy polarimetry and its application", *Ukr. J. Phys. Opt.* **2**, 58–74 (2001).
15. Y. Shopa and N. Ftomyn, "Polarimetric studies of linear dichroism in Cr-doped gallogermanate crystals", *Ukr. J. Phys. Opt.* **7**, 183–188 (2006).
16. C. Hernández-Rodríguez and P. Gomez-Garrido, "Optical anisotropy of quartz in the presence of temperature-dependent multiple reflections using a high-accuracy universal polarimeter," *J. Phys. D: Appl. Phys.* **33**, 2985–2994 (2000).
17. A. A. Kaminskii, B. V. Mill, G. G. Khodzhabagyan, A. F. Konstantinova, A. I. Okorochkov, and I. M. Silvestrova "Investigation of trigonal  $(\text{La}_{1-x}\text{Nd}_x)_3\text{Ga}_5\text{SiO}_{14}$  crystals", *Phys. Stat. Sol. (a)*, **80**, 387–398 (1983).
18. J. W. Ellis and L. Glatt, "Channeled infra-red spectra produced by birefringent crystals," *J. Opt. Soc. Am.* **40**, 141–142 (1950).

19. B. N. Grechushnikov and A. F. Konstantinova, "Crystal optics of absorbing and gyrotropic media," *Comput. Math. Applic.* **16**, 637–655 (1988).
20. A. P. Dudka and B. V. Mill', "Accurate crystal-structure refinement of  $\text{Ca}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$  at 295 and 100 K and analysis of the disorder in the atomic positions," *Crystallogr. Rep.*, **58**, 594–603 (2013).
21. Y. Shopa, N. Ftomyn, and I. Sokoliuk, "Crystal structure and optical activity of  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  crystals," *Ukr. J. Phys. Opt.* **15**, 155–161 (2014).
22. A. F. Konstantinova, T. G. Golovina, B. V. Nabatov, A. P. Dudka, and B. V. Mill', "Experimental and Theoretical Determination of the Optical Rotation in Langasite Family Crystals," *Crystallogr. Rep.* **60**, 907–914 (2015).
23. Y. Shopa, and N. Ftomyn, "Optical activity of  $\text{Ca}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$  crystals: experiment and calculus", *Optica Applicata.* **43**, 217–228 (2013).
24. Qi Haifeng, Wei Aijian and Yuan Duorong, "Investigation on giant optical activity of piezoelectric crystals with langasite structure," *Materials Science and Engineering: B*, **117**, 143–145 (2005).