Investigations of the optical activity of nonlinear crystals by means of dual-wavelength polarimeter

Mykola Shopa* and Nazar Ftomyn
*Gdansk University of Technology, Department of Atomic, Molecular and Optical Physics, Faculty of Applied Physics and Mathematics, Gdansk, Poland
Ivan Franko National University of Lviv, Department of General Physics, Faculty of Physics, Lviv, Ukraine

Abstract. A dual-wavelength method in high accuracy polarimetry has been successfully tested and applied to measure optical activity (OA) of nonlinear crystals. In proposed polarimetric scheme two neighboring semiconductor laser wavelengths (635 and 650 nm) are used, which increases number of parameters measured simultaneously and improves the data processing. By neglecting dispersion of eigen wave ellipticity in crystals, more efficient elimination of the systematic errors, in comparison with the known HAUP technique, is possible. We have tested our experimental setup on optically inactive lithium niobate crystal and obtained OA in the perpendicular to the optical axis direction for quartz and DKDP crystals. © 2018 Society of Photo-Optical Instrumentation Engineers (SPIE) [DOI: 10.1117/1.OE.57.3.034101]

Keywords: dual-wavelength; polarimetry; polarization; crystal optics; birefringence; optical activity.

1 Introduction

Dual-wavelength technique is widely used to expand the experimental methods, improve the measuring equipment, and reduce the measurement error. Examples of applications of dual-wavelength can be found in spectrophotometry, interferometry, holography, polarimetry, etc.

We have introduced an extended laser polarimeter, which implements two close wavelengths and uses different principles for data gathering and processing. The system is based on the optical scheme polarizer-sample-analyzer (PSA) and allows for full computer control over the experimental process. We have successfully tested it on an optically inactive LiNbO3 crystal and have measured the eigen waves ellipticities of SiO2 crystal. We have also applied it to acquire new results for deuterated potassium dihydrogen phosphate [K2HPO4, x = 0.94] or DKDP crystal.

Optical properties of crystals, such as linear birefringence, circular birefringence, linear, and circular dichroism, can be simultaneously and successfully studied by high-accuracy universal polarimeter (HAUP). HAUP has evolved over time and can be effectively applied to obtain information about main optical anisotropic parameters of crystals. Polarimeters allow measuring of optical activity (OA) for light propagation directions other than the optical axes, but results can significantly differ between themselves because of the systematic errors. Quantitatively one can take them into account, by considering small ellipticities of polarizer and analyzer azimuth.

Therefore, measurement errors can significantly impact the results.

2 Measuring Method and Experimental Details

The main principles of high-accuracy polarimetry are the following: as little number of elements in the polarization scheme as possible, use of monochromatic light sources (lasers), small input azimuths of light polarization before the sample, precise measuring of light intensities for different polarizer and analyzer positions, elimination of systematic errors (parasitic ellipticity and angular error), constant control of intermediate measuring results for verification of their correctness, full results processing only after completion of the measuring cycle, universality of methods, and possibility of the simultaneous measuring of crystal optical anisotropy parameters.

The relative intensity of transmitted light through the PSA system is a second-order function of the polarizer azimuth \( \theta \) and analyzer azimuth \( \chi \), which are measured from the principal axes and are relatively small (\( \theta, \chi \ll 1 \)). Then, the transmitted light intensity \( J(\theta, \chi) \) can be presented in a matrix form, similarly to Refs. 7 and 10

\[
J(\theta, \chi) = [1 \ (\chi - \theta) \ (\chi - \theta)^2]C \begin{pmatrix} \theta \\ \theta^2 \end{pmatrix}. \tag{1}
\]

For a birefringent and optically active crystal, matrix \( C \) has a simple form: \( C_{11} = C_{33} = C_{31} = 0 \), \( C_{13} = 2(p + q) \sin \Gamma \), \( C_{21} = 2(1 - \cos \Gamma) \), \( C_{23} = -2(k - p) \sin \Gamma \), and \( C_{22} = 2(1 - \cos \Gamma) \), \( C_{31} = 1 \). Here \( \Gamma = 2\pi d/\lambda \), where \( d \) is a thickness of specimen, \( \Delta n \) is the linear birefringence.
\( \lambda \) is the wavelength, and \( k \) is the ellipticity of eigen waves in crystals, which is measured as a ratio of the minor to the major axes of the ellipse.\(^6,11\) The ellipticities of polarizer \( p \) and analyzer \( q \) are taken into account. After the angular error for analyzer position \( \delta \chi \) is introduced, as the small difference in coordinates for PA and PSA polarization systems due to mechanical inaccuracies, we get from Eq. (1)

\[
J(\theta, \chi) = (\chi - \theta)^2 + A\chi \theta + B\theta + C, \tag{2}
\]

where \( A = 4\sin^2(\Gamma/2) \), \( B = 2[(k + q) \sin \Gamma - \delta \chi \cos \Gamma] \), and \( C = -2(k - p - \delta \chi) \). In practice, the constant component is also present in Eq. (2), which is independent on the azimuths \( \chi, \theta \) and therefore does not affect the measurement results. We also do not take into account the small quantities that are proportional to \( k^2, kp, kq \), etc.

The main feature of our polarimetric method is the experimental search of the positions of three characteristic azimuth angles \( \theta_{1,2} \) of the incident light in the PSA system with the preceding coordination of the polarizers and analyzers scales in PA system,\(^12,13\) in particular:

- \( \theta_0 \) is the polarizer azimuth, which is equal to the azimuth of the major axis of the polarization ellipse after the crystal,
- \( \theta_1 \) is the polarizer azimuth that corresponds to a global minimum of the emergent light intensity, when the ellipticity of the light passed through the PSA system is equal to zero,
- \( \theta_2 \) corresponds to a local minimum light intensity between the crossed polarizers in PSA system.

In \((\theta, \chi)\) coordinates, the minimum intensity azimuths of analyzer in the PSA system [correspond to the constant \((\partial J/\partial \chi)_{\theta} = 0\)] form a straight line \( x_{\text{min}}(\theta) \). The tangent of its slope angle is equal to one of the directly measurable quantities \( \cos \Gamma \).

We have measured three characteristic azimuth angles simultaneously for two wavelengths \( \lambda_1 \) and \( \lambda_2 \) by scanning the analyzer \( (\chi) \) angle in the range of about \( \pm 0.5 \) deg from the minimum transmittance of PSA system for each of about 20 positions of the polarizer \((\theta)\) angle in the same angular range. Due to the thermo-optical effect, the temperature change of the crystal invariably affects birefringence \( \Delta n_{1,2} \) for \( \lambda_1 \) and \( \lambda_2 \), respectively. As a result, we are able to vary the phase differences \( \Gamma_{1,2} = 2\pi \Delta n_{1,2}d/\lambda_{1,2} \).

The difference between the lasers wavelengths \( \lambda_1 \) and \( \lambda_2 \) was chosen to be relatively small in such a way that one can neglect the dispersion of the eigen wave ellipticity \( k \) and also assume that the parasitic ellipticities of the polarizer \( p \) and the analyzer \( q \) and also angular error \( \delta \chi \) are the same for both close wavelengths. At the same time, we can separately measure the phase differences \( \Gamma_{1,2} \) for each wavelength and use them later during the measurement results processing. From our point of view, the difference \( \Delta \lambda = \lambda_2 - \lambda_1 \approx 10 - 15 \) nm is optimal for the dual-wavelength polarimeter.

In dual-wavelength polarimeter experiments, we have used two semiconductor laser sources with close wavelength \((\lambda_1 = 635 \text{ nm and } \lambda_2 = 650 \text{ nm})\), which were switched by a mirror on a flip mount (Fig. 1). Two Glan–Taylor prisms with extinction ratio greater than 1:10\(^5\) were used in polarimeter as polarizer and analyzer. High-quality ball screw mechanical linear actuators translate linear motion with little friction to PC-controlled rotational motion of the polarizer and analyzer through the two-phase hybrid stepper motors. The light transmitted through the analyzer was detected with a photodiode in photovoltaic mode which was followed by a high impedance amplifier and digital voltmeter connected to PC. The PC software for measuring control was realized in a free Lazarus visual integrated development environment. The histogram analysis of the light detection system assumes good signal-to-noise ratio that is equal to 87 dB.\(^13\)

According to our procedure,\(^12,13\) measurement has been conducted in a relatively small \((\Delta T < 50 \text{ K})\) temperature interval for the two specimen positions in the PSA system which were obtained with 90 deg rotation of the crystal around the beam propagation direction. This rotation of the specimen ensured a change of the sign of the \( \Gamma_{1,2} \) and \( k \) values, but it did not change the systematic errors.

### 3 Testing Procedure on Lithium Niobate and Quartz Crystals

#### 3.1 Optically Inactive Sample of Lithium Niobate

We have started our experiments with a well-known non-linear lithium niobate crystal LiNbO\(_3\). Since this material is optically inactive \((k = 0)\) the basic formulas of our measuring method are simplified. We have, therefore, a possibility to eliminate all systematic errors \( p, q, \) and \( \delta \chi \). In contrast, in this most simple case, systematic errors can be easily defined using a standard procedure,\(^14\) but we have a possibility to check the basic relations, which can be used in the case of dual-wavelength laser polarimetry.

Detailed selection of the thickness of specimen \((d = 0.4555 \text{ mm})\) ensured a change of the phases difference \( \Gamma_{1,2} \). For two close wavelengths, there is a clear shift in the temperature dependencies of \( \cos \Gamma_{1,2} \) values (Fig. 2). Using the relations for \( \theta_0 \) and \( \theta_1 \) from Ref. 13 and by introducing the azimuth angles differences \( \Delta \theta_{01} = \theta_{01}(\lambda_1) - \theta_{01}(\lambda_2) \), \( \Delta \theta_{12} = \theta_{12}(\lambda_1) - \theta_{12}(\lambda_2) \), and \( \Delta \theta_{21} = \theta_{21}(\lambda_1) - \theta_{21}(\lambda_2) \) for two wavelengths, we can write for an optically inactive LiNbO\(_3\) crystal...
\[ \Delta \theta_{1\theta} = -A_1 p - B_1 \delta \chi, \quad \Delta \theta_{1\lambda} = -A_2 p - B_2 q, \]
\[ \Delta \theta_{2\lambda} = -A_1 (p + q)/2, \] (3)

where \( A_1 = \cot(\Gamma_1/2) - \cot(\Gamma_2/2), \) \( B_1 = 1/(1 - \cos \Gamma_1) - 1/(1 - \cos \Gamma_2), \) \( A_2 = \cot \Gamma_1 - \cot G_2, \) \( B_2 = 1/\sin \Gamma_1 - 1/\sin \Gamma_2, \) and \( \Gamma_1 = \Gamma(\lambda_1), \) \( \Gamma_2 = \Gamma(\lambda_2). \) Since the relations, which are expressed by Eq. (3), are linear, values of the systematic errors \( p \) and \( \delta \chi \) can be defined using procedure of the straight line approximation. In particular, the dependence of the specific value \( \Delta \theta_{1\theta}/B_1 = f(A_1/B_1) \) for two specimen positions in the PSA system and the best fit of the experimental data are presented in Fig. 3.

In addition, the magnitude of systematic errors \( p \) and \( q \) can be successfully determined using the second Eq. (3) too, as can be seen in Fig. 3 [experimental dependence of the \( \Delta \theta_{1\lambda}/A_2 = f(B_2/A_2) \)]. In this case, the slope and intercept values of the fitted lines can be used for systematic errors calculation. Finally, the mean absolute values of systematic errors, which have been obtained from the Eq. (3), are the following: \( p = 6.5 \times 10^{-4}, q = 3.0 \times 10^{-4}, \) and \( \delta \chi = 1.8 \times 10^{-4}. \) Acquired value of the \( p \) is comparable with the results for systematic errors in the HAUP method,\(^9\,14,\,15\) where the reference LiNbO\(_3\) crystal has been used.

### 3.2 Verification of the Optical Activity in Quartz by Dual-Wavelength Polarimeter

The SiO\(_2\) crystals belong to point group symmetry 32, the gyration tensor is diagonal with components \( g_{11} = g_{22}, \) and \( g_{33}.\)\(^11\) The various modifications of HAUP have been successfully applied for these materials.\(^5\,9,\,14\) Thanks to the high-quality crystals, quartz is a traditional test case for new techniques (see, for example, Ref. 16). Detailed investigations of the magnitude of \( g_{11} \) gyration tensor component of quartz have shown limited temperature dependence in the region of 200 to 360 K.\(^5\) We have also exploited this feature to test our method and hardware.

The SiO\(_2\) crystalline plates (\( d = 2.133 \text{ nm} \)) of high optical quality, cut parallel to optical axis, were used in our experiment. Measurement has been done in a temperature interval of 310 to 360 K. In this case, the phase difference could change on \( \Delta \Gamma \approx \pi/2. \) The parasitic ellipticities \( p \) and \( q \) are additive to \( k \) and, to successfully eliminate them, the experiment has been conducted for two specimen position in the PSA system. As mentioned earlier, the specimen 90 deg rotation along the beam propagation direction ensures that the sign of \( k \) changes.

It is obvious, that to exclude the systematic errors, it is enough to use one of the relations for characteristic azimuths \( \theta_0 \) or \( \theta_1. \) Using the notation \( \bar{k} = [k(\lambda_1) + k(\lambda_2)]/2, \) the difference between characteristic azimuths \( \theta_0 \) and \( \theta_1 \) may be represented as

\[ \Delta \theta_{1\theta} = A_1 (\bar{k} - p) - B_1 \delta \chi, \]
\[ \Delta \theta_{1\lambda} = A_2 (\bar{k} - p) - B_2 (\bar{k} + q). \] (4)

We should note, that Eq. (4) is valid at \( \Delta k = k(\lambda_1) - k(\lambda_2) \ll 2p \) and \( 2q. \) In other words, it is necessary to use sources of light with such wavelengths, that this requirement is satisfied for the investigated objects. Therefore, the small change (10 to 15 nm) in wavelength leads to changes of the \( k \) on \( \pm 1\% \) to \( 2\% \) for SiO\(_2\) crystals. Finally, we can neglect \( \Delta k \) value because it does not lead to essential errors in definition of magnitude. As a result, experimental dependencies \( \Delta \theta_{1\lambda}/A_2 \) versus \( B_2/A_2, \) measured for two specimen positions, are presented in Fig. 4. The straight line approximation procedure allows the determination of the systematic errors as magnitudes of slope and intercept values.
4 Eigen Wave Ellipticity of DKDP Crystal

Our next step was to apply the dual-wavelength polarimetric technique to observe the OA in a well-known material such as deuterated potassium dihydrogen phosphate or DKDP single crystal that is widely used in nonlinear optics and optoelectronics.

The processing of the results, measured on the dual-wavelength polarimeter, can be carried out in different ways. Here, in particular we use a different procedure from that used in Sec. 3.2 and in our previous work. Since in the general case, the value of eigen wave ellipticity $k$ also depends on the temperature, it is impossible to use similar to Fig. 4 straight line approximation.

Using expressions for the characteristic azimuth angles $\theta_1$ and $\theta_2$, we can obtain the following relation:

$$-2(\theta_1 - \theta_2) \cot(\Gamma/2) = -2\Delta\theta_{12} \cot(\Gamma/2)$$

$$= 2k - p + q - \delta\chi \cot(\Gamma/2). \quad (5)$$

To find ellipticity $k$ and systematic errors $p$, $q$, and $\delta\chi$, we calculated the quantities from the left side of Eq. (5) for each wavelength (635 and 650 nm). Using the rotation of the crystal for 90 deg around the light beam axis and considering the sign change of $k$ and $\Gamma_{1,2}$, we acquired four temperature dependencies for values from left side of Eq. (6). As a next step, we used the following notations for them:

$$\Delta^0_{1,2} = 2k - p + q - \delta\chi_0 \cot(\Gamma^0_{1,2}/2),$$

$$\Delta^90_{1,2} = -2k - p + q - \delta\chi_{90} \cot(\Gamma^{90}_{1,2}/2). \quad (6)$$

where lower index 1, 2 corresponds to the wavelength 635 and 650 nm, respectively, and upper index 0, 90 to the two setups of the crystal in PSA system (before and after its 90-deg rotation). It is obvious that $k_0 = k$, $k_{90} = -k$, $\Gamma^0_{1,2} = \Gamma_{1,2}$, and $\Gamma^{90}_{1,2} = -\Gamma_{1,2}$. We should also note, that the systematic angular error $\delta\chi$ in our case changes the value after the crystal is rotated; however, the parasitic ellipticities $p$ and $q$ remain unchanged.

Thus, we obtained the temperature dependencies of $\Delta^0_{1,2}$, $\Delta^90_{1,2}$, $\Delta^90_{1,2}$, and $\Delta^90_{1,2}$, which are presented in Fig. 6. From this figure and on the basis of Eq. (6), we calculated the two angular error values $\delta\chi_{0/90} = -(\Delta^0_{1,2} - \Delta^90_{1,2})/\left[\cot(\Gamma^0_{1,2}/2) - \cot(\Gamma^{90}_{1,2}/2)\right]$, in particular $\delta\chi_{0} = -3.18 \times 10^{-4}$ and $\delta\chi_{90} = 5.60 \times 10^{-4}$ (in radians).

For two alternative crystal orientations (0 and 90 deg) in PSA system and two wavelengths, using four $\Delta_{1,2}$ quantities and angular systematic errors $\delta\chi$, we found for $T = 295$ K...
that $2k - p + q = 5.58 \times 10^{-4}$ and $-2k - p + q = -7.64 \times 10^{-2}$, respectively.

Taking into account the superposition of parasitic ellipticities $-p + q = -1.03 \times 10^{-4}$, the mean value of the eigen wave ellipticity $k = 3.31 \times 10^{-5}$. As a result, considering measurements errors, for DKDP crystal we could determine the magnitude of the gyration tensor component $g_{11} = (3.61 \pm 0.3) \times 10^{-5}$ and optical rotatory power perpendicular to the optical axis $\rho_1 = 6.91 \pm 0.57$ deg/mm.

Thus, we have obtained a quantitatively similar result to one in our other work13 however, here we have applied a more general data processing scheme in dual-wavelength polarimeter. The sequence of the dual-wavelength polarimeter measurement results processing, presented in this section, is better suited for the measurement of small ellipticity $k$ values, although it is more complicated when compared with the one used for quartz.

5 Conclusions

We used the dual-wavelength polarimetric method that is related to HAUP, for measuring of the eigen wave ellipticity and OA in birefringent direction of transparent nonlinear crystal. Introduction of two neighboring laser wavelengths into polarimeter scheme allowed us to more consistently eliminate systematic errors. We assumed an approximation that dispersion of the eigen wave ellipticity over small wavelength change is negligible and that the parasitic ellipticities of the polarizer and the analyzer and angular error are the same for both close wavelengths, but phase difference for each wavelength is different.

We tested our dual-wavelength polarimeter on optically inactive lithium niobate crystal and used new systematic errors elimination scheme for measuring the ellipticities of quartz crystal and acquired results for DKDP crystal. More precise measurements with the dual-wavelength scheme are possible when highly monochromatic and power stable laser sources are used.

References


Mykola Shopa is an assistant professor at the faculty of applied physics and mathematics at the Gdansk University of Technology, Poland. He received his BS and MS degrees in physics from Ivan Franko National University of Lviv, Ukraine, in 2006 and 2007, respectively, and his PhD in physics from the Institute of Physics PAS in Warsaw.
Poland, in 2013. His current research interests include polarimetry, optical anisotropy, optical metamaterials, plasmonics, and photonics.

Nazar Ftomyn received his PhD in optics and laser physics in 2013. Currently, he is an assistant professor at the Ivan Franko National University of Lviv, Ukraine. Prior to this, he was an assistant at the general physics department at the same university. From 2005 to 2007, he worked as an engineer at Ivan Franko National University of Lviv. His main spheres of scientific activity are high accuracy polarimetry and crystal optics. He is an author of over 13 refereed publications.