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LINEAR BIREFRINGENCE OF CARPATHIAN QUARTZ THIN ANISOTROPIC LAYERS ESTIMATED BASED ON THE LIGHT POLARIZATION ELLIPSE

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Abstract: A simple method for determining the linear birefringence of the transparent uniaxial anisotropic layers is described in this manuscript. Linear birefringence is a dispersive, material-dependent parameter computed by the difference between the principal refractive indices of the anisotropic layers. The proposed method consists of estimating the polarization state of light using a device composed of two identical polarizers, one anisotropic layer, a light source and a light detector. The method is based on the existent relation between the material linear birefringence and the azimuth of incident linearly polarized light in anisotropic layer as well as the rotation angle of the polarization ellipse at the exit from it. The method can be applied to different thin uniaxial anisotropic layers, such as crystalline inorganic, liquid crystals, or polymeric films. Here, the validity of the new method is verified for quartz, a very used material in different technical applications.

Keywords**:** Linear birefringence; Azimuth angle; Polarization ellipse; Carpathian quartz

Introduction

The optical linear birefringence[1,2] of the transparent anisotropic materials is an important parameter which determines changes of the light

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polarization state in the propagation process. In the anisotropic layers, the velocity depends both on the propagation direction and on the polarization state of light. In the principal system of coordinates, *a*O*bc*, only diagonal elements differ from zero and the refractive indices of substance show only three values, n_a, n_b and n_c. In the case of uniaxial crystal, two values, $n_e = n_c$ and $n_o = n_a = n_b$ refractive indices are measured, while in the case of biaxial crystals three values of the refractive indices are characteristic.

In the uniaxial crystals there is a unique direction for which the propagation velocity does not depend on the polarization state of light while in biaxial crystals there are two such propagation directions. The principal directions O*a* and O*b* perpendicular on the optical axis, O*c*, are characterized by the fact that the linear polarized radiation does not change its polarization state in the propagation process. Thus, the easier mode for describing the polarization state of light[1] is to judge the phase difference between the components of light acting on the two principal directions.

The polarization state at the exit from anisotropic layer can be established only for the incident total polarized light. The total polarized light propagates only on distances smaller than the coherence distance.[1,2] For higher distances, light becomes non-polarized because it results from different emission acts.

The anisotropic layers have multiple applications: they can change the polarization state of the total polarized radiation, can change the light spectral composition, can produce linearly polarized radiations by dichroism, or can compensate the optical pathway etc.

The linear birefringence[1,2] of the uniaxial anisotropic layers is defined as the difference between the principal refractive indices, extraordinary (n_e) and ordinary (n_o) indices, measured with linearly polarized radiations with their electric field acting parallel, respectively perpendicular to optical axis.

$$\Delta n = n_e - n_o \quad (1)$$

The linear birefringence depends both on the nature of the anisotropic material and the spectral composition of light because the principal refractive indices are dependent on the light wavelength.

There are methods to determine the birefringence by direct measurements of the principal refractive indices of anisotropic layers.[1] The interferential methods[3,4] for the birefringence measurements are the most precise. The birefringence can be directly measured by the phase difference introduced in the anisotropic layer between the principal components of light in propagation process for inorganic crystals,[5,6] polymer films,[7-9] or liquid crystals.[10-12]

In this manuscript, one interferometric method, based on the dependence of the phase difference (introduced by the anisotropic layer between the principal components of light) and both the azimuth angle of the incident linearly polarized radiation and the angle of rotation of the polarization ellipse at the exit from the anisotropic layer, is described. This method, called determination of linear birefringence based on polarization ellipse, can be applied only for thin anisotropic layers, as it will be demonstrated.

Experimental set-up

The experimental set-up used for measuring the linear birefringence is schematically drawn in Figure 1. The light source S illuminates the lens L with collimating role. The parallel beam passes through two identical polarizing filters P and A, named polarizer and analyzer, having between them an anisotropic layer, AL. At the exit from the analyzer A, the light flux is measured by the detector D. The changes in the flux at rotation of the analyzer A are analyzed by the light detector D.

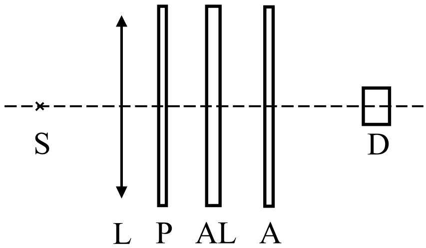

Figure 1. Experimental set-up: S – light source, L – collimating lens, P and A – identical polarizing filters, AL – anisotropic thin layer, D – detector.

The source is placed in the object focus of the lens L. The polarizing filter P is fixed in such a way that its transmission direction reaches 45° with the principal axes of the AL, cut parallel to its optical axis (see Figure 2). So, the projections of the amplitudes of light on the principal axes of LA are equal. Function of the phase difference introduced between the two light components, the exit radiation from AL can be linear, elliptic or circular polarized at the entrance in A.

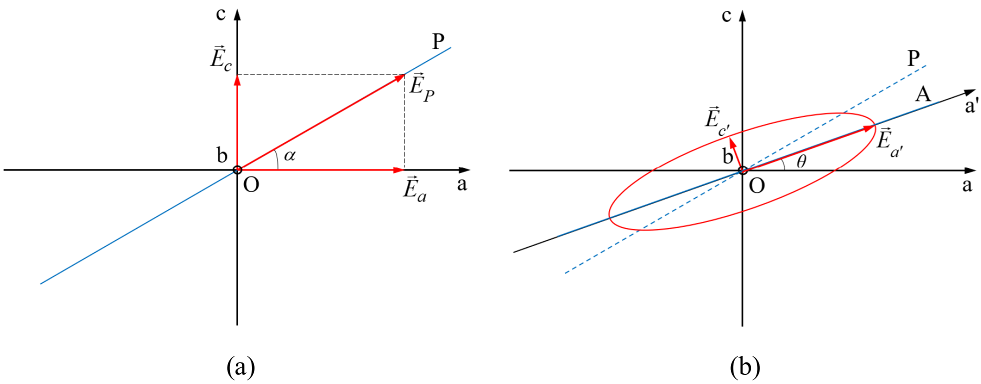

Figure 2. a) Reciprocal orientation of the transmission direction of polarizing filter P and the principal axes of AL; b) Polarizing ellipse at the exit from AL and its rotation angle relative to the ellipse with axes on the ellipse reported to the principal axes of AL.

When the transmission direction of P is perpendicular to transmission direction of A, light does not pass through the system. When AL is introduced between the polarizing filters, the light polarization state is changed as function of the phase difference introduced between the principal components of light by the anisotropic layer AL. Let us suppose that the transmission direction of P makes angle α with principal axis of AL. The analyzer A is used to estimate the polarization state of light at the exit from AL.

A) If at a complete rotation of A, one obtains two maxima and two null minima, the emerging from AL is linear polarized with the same azimuth α, or with an azimuth $\pi - \alpha$. The phase difference between the principal components introduced by AL is an integer number of π.

B) When after A, the detector D measures equal fluxes, indifferent on the rotation angle of A, the emergent light from AL is circularly polarized, if the phase difference is $(2k + 1)\pi/2$ and the azimuth angle is 45°. For azimouth angles different on 45°, one obtains an polarization ellipse with semiaxes parallel to principal axes of AL.

C) When the phase difference between the principal components of light is between $k\pi$ and $(2k + 1)\pi/2$, when analyzer A is rotated, one obtains two maxima and two non-null minima, because the polarization state becomes elliptical.

The polarization state of the incident light in analyzer A is estimated by rotating the analyzer with 360° and recording the maxima and minima of flux density.

Theoretical bases

The natural light emitted by source S is transformed into linearly polarized radiation by dichroism when it passes through the polarizer P (see Figure 2). To simplify the calculations, the linearly polarized radiation incident on AL is considered as being decomposed in two linearly polarized coherent components acting on the principal directions of the anisotropic layer[2]. Let be the principal axis O*b* the propagation direction (see Figure 2). The two light components acting on principal axes O*a* and O*c* have the phase difference $\Delta\psi$ at the exit from AL.

$$\Delta\psi = \frac{2\pi}{\lambda}\Delta n L \qquad (2)$$

In Equation (2), λ is the radiation wavelength, and L is the thickness of the AL.

If α is the azimuth angle of the linearly polarized light relative to O*a* axis, and the phase difference $\Delta\psi$ between the light principal components is attributed to O*c* component, one can write:

$$e_a = E_P \cos\alpha \cos\left(\omega t + \psi_0\right), \qquad (3)$$

$$e_c = E_P \sin\alpha \cos\left(\omega t + \psi_0 + \Delta\psi\right). \qquad (4)$$

In relations (3) and (4), e and E are the elongation and magnitude of the electric field of light transmitted by P, ω is the light angular speed, ψ_0 is the initial phase of light.

At the exit from AL, the two components of light interfere, and the result is the polarization ellipse.

$$\left(\frac{e_a}{E_a}\right)^2 + \left(\frac{e_c}{E_c}\right)^2 - 2\frac{e_a e_c}{E_a E_c}\cos\Delta\psi = \sin^2\Delta\psi \qquad (5)$$

At the exit from AL, the electric field of light having the origin on the propagation direction, describes, according relation (5), an ellipse with its semiaxes parallel to the principal directions of AL, when $\Delta\psi = (2k + 1)\pi/2$.

When $\Delta\psi = k\pi$, the ellipse (5) can degenerate in a line with non-changed azimuth α for k an even number, or in a line having azimuth $\pi - \alpha$, when k is an odd number.

If the azimuth is 45° ($E_a = E_c$), and $\Delta\psi = (2k + 1)\pi/2$, the polarization ellipse degenerates in a circle. In the case $k\pi < \Delta\psi < (2k + 1)\pi/2$, the polarization ellipse has its axes rotated relative to the principal axes of AL with angle θ. When the polarization ellipse is rotated with the angle θ, in order to have the semiaxes parallel to the principal axes of AL, one obtains the condition (6).

$$\cos\Delta\psi = \frac{\tan 2\theta}{\tan 2\alpha} \qquad (6)$$

To avoid the incertitude bond on the periodicity of the trigonometric function, one can use two similar layers of very small difference in thickness, introduced between the polarizing filters with crossed optical axes. In this way, the ordinary ray in the first layer becomes extraordinary in the second and the extraordinary one becomes ordinary, and the pathway depends only on the difference of the thicknesses of the layers.

Results and Discussion

Three layers of Carpathian quartz having different thicknesses are used to illustrate the applicability of the proposed method. The results of the measurements are given in Tables 1 - 3. The values of the linear birefringence of Carpathian quartz are determined with a light filter of λ = 500 nm. The disadvantage of this method is obtaining results for each component of white light in different experiments.

Four azimuthal angles were fixed for each experiment in order to obtain information about the possibility to estimate the angle θ with enough precision.

Table 1. Orientation α of polarizer P relative to Oa axis for quartz layer with L = 3 μm; relative orientation θ of polarizer A and principal axis of the quartz layer; phase difference, $\Delta\psi$, and linear birefringence of quartz, Δn.

No.	α (°)	tan 2α	Θ (°)	$\Delta\psi$ (°)	Δn
1	10	0.36397	9.46	19.6535	0.009099
2	20	0.83910	19.18	19.4008	0.008982
3	30	1.73205	29.20	20.2035	0.009535
4	40	5.67128	39.69	19.8848	0 009258

The values obtained for different azimuths, α, are alike and have the average value 0.009219. The measurements results do not depend on the azimuth angle. To verify the precision of the measurements another two layers with other thickness were measured in the same kind, using the method described previously. The results were similar.

Table 2. Orientation α of polarizer P relative to Oa axis for quartz layer with L = 5 μm; relative orientation θ of polarizer A and principal axis of the quartz layer; phase difference, $\Delta\psi$, and linear birefringence of quartz, Δn.

No.	α (°)	tan 2α	Θ (°)	$\Delta\psi$ (°)	Δn
1	10	0.36397	8.533	32.488	0.00904
2	20	0.83910	17.650	32.456	0.009016
3	30	1.73205	27.810	32.488	0.009024
4	40	5.67128	39.075	32.821	0 009117

Table 3. Orientation α of polarizer P relative to Oa axis for quartz layer with L = 10 μm; relative orientation θ of polarizer A and principal axis of the quartz layer; phase difference, $\Delta\psi$, and linear birefringence of quartz, Δn.

No.	α(°)	tan 2α	θ(°)	$\Delta\psi$(°)	Δn
1	10	0.36397	8.78	64.89	0.009013
2	20	0.83910	19.63	64.84	0.009006
3	30	1.73205	36.36	64.85	0.009007
4	40	5.67128	67.46	64.86	0 009008

The average value of the data from Table 2 was 0.009049, while for the data from Table 3 one obtains an average value of about 0.009009.

The average value of the measured linear birefringence was 0.009092.

The standard deviation was about 0.000159. The result of the measurement of the linear birefringence of Carpathian Quartz was Δn = 0.009092 ± 0.000159. The obtained value confirms the applicability of the proposed method for determining the linear birefringence of the thin anisotropic layer.

This method can be used in laboratories which do not possess ellipsometer devoted to this kind of measurements.

Conclusions

The values of the linear birefringence contained in this article were determined for λ = 500 nm. The dispersive properties of the quartz samples can be estimated if one uses different components of white light. The method was applied for different azimuthal angles in order to search its precision for various inclinations of the electric field of the incident linearly polarized light relative to the optical axis of crystalline thin layer. The method is applicable only to thin layers due to imprecision in estimating the phase difference caused by the periodicity of trigonometric function. The disadvantage of the described method is that in order to estimate the birefringence dispersion, the method asks for different measurements using different components of light.

References

1. Pop, V. *Bazele Opticii* (in Romanian). "Alexandru Ioan Cuza" University Publishing House, Iasi, 1987; pp. 42 – 53.
2. Dorohoi, D. O. *Fundamental Optics*. Addleton Academic Publisher, New York, 2010; pp. 51 – 64.
3. Pop, V.; Dorohoi, D. O.; Cringeanu, E. A new method for determining birefringence dispersion. *J. Macromol. Sci. B: Phys.* **1994**, *33*, 373 – 385. https://doi.org/10.1080/00222349408248099
4. Dumitrascu, L.; Dumitrascu, I.; Dorohoi, D. O.; Toma, M. Interferometric method for birefringence determination with a polarizing microscope. *Opt. Express*. **2008**, *16*, 20884 – 20890. https://doi.org/10.1364/OE.16.020884
5. Androne, D. A. M.; Dorohoi, D. O.; Dimitriu D. G.; Kasper, H. U. Optical and chemical study of Quartz from granitic Pegmatites. *Rev. Chim*. **2018**, *69*, 1846 – 1850. https://doi.org/10.37358/RC.18.7.6429
6. Dumitrascu, L.; Dumitrascu, I; Dorohoi, D.O. Conoscopic method for determination of main refractive indices and thickness of a uniaxial crystal cut out parallel to its optical axis. *J. Appl. Cryst.* **2009**, *42*, 878 – 884. https://doi.org/10.1107/S0021889809025709
7. Postolache, M.; Dimitriu, D. G.; Nechifor, C. D.; Bota-Condurache, S.; Closca, V.; Dorohoi, D. O. Birefringence of thin uniaxial polymer films estimated using the light polarization ellipse. *Polymers* **2022**, *14*, 1063. https://doi.org/10.3390/polym14051063
8. Avadanei, M. I.; Dimitriu, D. G.; Dorohoi, D. O. Optical anisotropy of polyethylene terephthalate films characterized by spectral means. *Polymers* **2024**, *16*(6), 850. https://doi.org/10.3390/polym16060850

9. Dorohoi, D. O.; Postolache, M.; Nechifor, C. D.; Dimitriu, D. G.; Albu, R. M.; Stoica, I.; Barzic, A. I. Review on optical methods used to characterize the linear birefringence of polymer materials for various applications. *Molecules* **2023**, *28*, 2955. https://doi.org/10.3390/molecules28072955

10. Wu, S.T.; Efron, U.; Hess, L. D. Birefringence measurements of liquid crystals. *Appl. Optics.* **1984**, *23*(21), 3911 – 3915. https://doi.org/10.1364/AO.23.003911

11. Dorohoi, D.O.; Postolache M.; Postolache, M. Birefringence dispersion of poly (phenyl methacrylic) ester of cetyloxybenzoic acid in tetrachloromethane, determined from channeled spectra *J. Macromol. Sci., Part B: Phys*. **2001**, *40*(2), 239 – 243. https://doi.org/10.1081/MB-100001812

12. Picos, S.; Amarandei, G.; Diaconu, I.; Dorohoi, D. O. The birefringence of thin films of some nematic liquid crystals. *J. Optoelectron. Adv. Mater.* **2005**, *7*(2), 787 – 793.