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Measurement of refractive index of biaxial potassium titanyl phosphate crystal plate using reflection spectroscopic ellipsometry technique

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Abstract. The paper reports the measurement of refractive indices and anisotropic absorption coefficients of biaxial potassium titanyl phosphate (KTP) crystal in the form of thin plate using reflection ellipsometry technique. This experiment is designed in the Graduate Optics Laboratory of the Addis Ababa University and He–Ne laser ($\lambda = 632.8$ nm), diode laser ($\lambda = 670.0$ nm) and temperature-tuned diode laser ($\lambda = 804.4$ and 808.4 nm), respectively have been employed as source. The experimental data for n_x, n_y are fitted to the Marquardt–Levenberg theoretical model of curve fitting. The obtained experimental data of refractive indices are compared with different existing theoretical and experimental values of KTP crystals and found to be in good agreement with them.

Keywords. Ellipsometry; He–Ne laser; lock-in amplifier; potassium titanyl phosphate.

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1. Introduction

Generally, the refractive indices of nonlinear crystals are measured by minimum deviation technique, where the materials are cut in prism form and the beam of monochromatic light is allowed to incident perpendicular to the surface of the prism. The angle of minimum deviation of the refracted light from the second surface of the prism is measured accurately by goniometer. A uniaxial nonlinear crystal which has two refractive indices n_o and n_e, respectively requires two prisms with two defined orientations. One of these prisms has its crystallographic axis perpendicular to the surface of incidence of the prism whereas the other one has its crystallographic axis on the surface only [1]. However, this problem becomes more complicated in the case of a biaxial nonlinear crystal because it has three crystallographic axes parallel to x,y and z directions and therefore it has three refractive indices n_x, n_y and n_z respectively [2,3].

The above-mentioned method of minimum deviation using goniometer requires large-sized materials for making prisms of different orientations resulting in the wastage of expensive nonlinear materials. Moreover, the optical quality of the surface should also be very good. Shukla *et al* have reported interferometry-based techniques to measure the refractive indices of birefringent materials and thin films [4,5]. But this technique is restricted to visible range of wavelength and a crystal of suitable wedged angle with thickness of the order of 4.0 mm is one of the indispensable requirement.

There are some more reports on the measurement of optical constants of anisotropic nonlinear materials in thin film form [6–9]. However, Xiong *et al* have reported the characterization of KTP films fabricated by pulsed laser deposition on substrates of sapphire and silica [10]. They have also deduced the refractive index of KTP films from spectroscopic ellipsometry data either by direct de-convolution or by least square fitting technique and compared their results with Sellmeier equations given by Fan *et al* [11].

In this paper, we have also made an attempt to measure the refractive indices of a thin plate of biaxial KTP crystal grown by flux growth technique at four different wavelengths of lasers using reflection ellipsometry technique. This technique is developed in the Graduate Optics Laboratory of the university. The experiment is carried out between VIS and NIR region using different types of laser sources. By this technique the change in polarization of light that is incident on a material from an oblique angle is measured. The incident light beam interacts with each layer of the material. Some part of the light will be reflected at each interface, creating an interference pattern representative of the material's optical properties. The said interference pattern is collected at several polarizations and processed using suitable software.

The compound KTP was discovered and introduced in the field of optics in 1986 [12,13]. It is a biaxial crystal which belongs to the orthorhombic group with point group symmetry mm2. Its high transparency range (0.35–4.5 μm), higher damage threshold and non-hygroscopic nature shows its superiority over many nonlinear crystals and it is proved to be ideal for making nonlinear devices [14–16]. Since nonlinear devices made of KTP crystals in different optical regions exclusively depend on phase matching angle, very accurate knowledge of the refractive indices is necessary. The x–y–z cut KTP crystal used in the experiment is a flux-grown crystal having unit parameters a_0 = 12.87 Å, b_0 = 6.406 Å and c_0 = 10.64 Å, respectively.

2. Theory

We have employed reflection spectroscopic ellipsometry technique to measure the refractive indices and absorption coefficients of a biaxial KTP crystal plate having $10 \times 10 \times 3.0$ mm^3 size. This technique is non-invasive and non-destructive in nature and requires low-power light source along with low-cost detection system such as photodiode power meter/energy meter. Moreover, the angle of incidence is kept fixed during the entire measurement and therefore it can be chosen maximum so as to minimize the direct coupling between the source and the detector. It is based on measurement of the polarization of a beam of light reflected from the surface of the sample at a known angle of incidence. Therefore, no reference samples are required for calibration. Two parameters (ψ and Δ) are measured as a function of wavelength and the angle of incidence. These parameters are related to the complex reflectivity ratio ρ

$$\rho = r_p/r_s = \tan\psi \cdot \mathrm{e}^{i\Delta}, \tag{1}$$

where r_s and r_p are the complex reflection coefficients for light polarized perpendicular and parallel to the plane of incidence, respectively.

The intensity waveform obtained from the ellipsometric measurements are sinusoidal in nature and is given by

$$I_{\mathrm{D}} = \langle I_{\mathrm{o}} \rangle [1 + \alpha\cos 2A + \beta\sin 2A]. \tag{2}$$

I_{o} denotes the intensity of the light beam at the output of the optical system S, I_{D} represents the proportion of I_{o} passed by the analyzer A and detected by the photodetector D, whereas $\langle I_{\mathrm{o}} \rangle$ represents the average detected signal over one full rotation of the analyzer and α and β represent the normalized cosine and sine Fourier coefficients of the signal I_{D} (A). The values of C, α and β are given as follows:

$$C = \sin^2[P]/2 + \cos^2[P]\tan^2[\Psi]/2, \tag{3}$$

$$\alpha = \cos^2[P]\tan^2[\Psi]/2 - \sin^2[P]/2, \tag{4}$$

$$\beta = \cos[P]\sin[P]\tan[\Psi]\cos[\Delta]/C. \tag{5}$$

The relationship between α and β with the ellipsometer parameters Ψ and Δ are given by

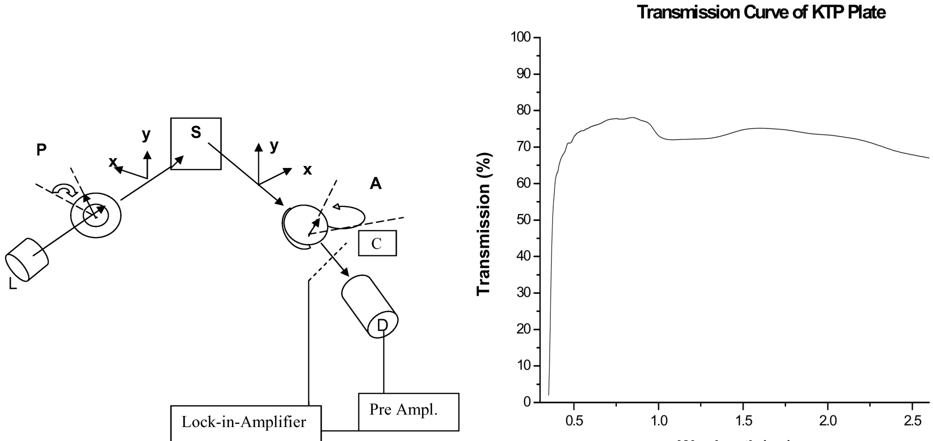
$$\tan[\Psi] = \sqrt{}[1+\alpha]/[1-\alpha]\tan P \tag{6}$$

$$\cos\Delta = \beta/\sqrt{}1-\alpha^2. \tag{7}$$

The values of α and β are determined by curve fitting methods from the graph of intensity vs. angle of analyzer whereas eqs (6) and (7) are the desired relationship between the ellipsometer parameters Ψ and Δ with the constants α and β for a given azimuthal angle P of the polarizer. Thus the values of α and β can easily be determined by the curve fitting of eq. (2). The value of ρ, the ratio of the complex Fresnel coefficients, is given by eq. (1).

Once the value of ρ is determined, we can calculate the refractive index (RI) of the same using the equation

$$n = \sin\theta_0[1 + [1-\rho/1+\rho]^2\tan\theta_0]^{1/2}. \tag{8}$$



(a) (b)

Figure 1. (**a**) The experimental set-up for the measurements of RI of KTP crystal plate, where L is the laser source, P is the polarizer, S is the sample, A is the analyzer, C is the mechanical chopper, D is the photo pin detector, and Pre-ampl is the pre-amplifier. (**b**) Transmission curve of KTP plate between 350 and 2600 nm wavelength.

The values of anisotropic absorption coefficient (K) can be calculated using the following equation [17]:

$$K = 1/2n[\sin^2\theta_0 \tan^2\theta_0 \sin 2\Psi \sin\Delta/(1+\sin 2\Psi\cos\Delta)^2]. \tag{9}$$

Theoretical values of refractive indices n_x, n_y and n_z of KTP crystals along the x, y and z-axes can be calculated using Sellmeier's equations. Equations (10i)–(10iii) are the popular Sellmeier's equations for KTP crystals given by Kato [18].

$$n_x^2 = 3.0065 + 0.03901/(\lambda^2 - 0.04251) - 0.01327\lambda^2, \tag{10i}$$

$$n_y^2 = 3.0333 + 0.04154/(\lambda^2 - 0.04547) - 0.01408\lambda^2, \tag{10ii}$$

$$n_z^2 = 3.3134 + 0.05694/(\lambda^2 - 0.05658) - 0.01682\lambda^2, \tag{10iii}$$

where λ is in μm.

A model for RI is constructed after the data are acquired covering the desired wavelength and angle of incidence. The Fresnel equation with the assumed model is then used to predict the expected ψ and Δ data for the chosen wavelength and angle of incidence. Data fitting is then applied by adjusting the model parameters to find the best-fit values. The Marquardt–Levenberg algorithm is most commonly used to determine the smallest difference between the measured and calculated values quickly.

3. Experimental

The experimental set-up used in our experiment is shown in figure 1a, where He–Ne laser of wavelength 632.8 nm, a diode laser of wavelength 670 nm and temperature-tuned semiconductor–diode laser model LDC01, MEOS made tunable between 804.4 and 808.4 nm have been used as a source. A set of linear polarizers in visible optical range, available in the laboratory for different types of optical experiments is also used. The semiconductor laser is able to produce 804.4 nm at temperature 16.24°C and 808.4 nm at temperature 27.9°C, respectively. By knowing the incident polarization state of the laser light using polarizer P, the polarization state of the reflected light from the surface of KTP crystal plate S is ascertained with the help of analyzer A. The intensity of the reflected polarized light is chopped with the help of a chopper C at 50 Hz and detected by PIN photodetector D. The output of the PIN photodetector is further amplified using a pre-amplifier and fed to the lock-in amplifier, Model SR 830 DSP, Stanford Research System Inc., USA, to measure the intensity of the reflected polarization state at different angles of polarization. The use of lock-in amplifier helps to nullify the noises from different sources and plays a very significant role to measure some of the weak reflected signals very accurately. Since the ellipsometric measurement technique relies on the relative amplitudes between the reflected polarized components and not the absolute reflectivity of the sample materials, the variation in intensity of the incident laser radiation does not cause loss of accuracy. The beam diameter of the laser beam is 2.0 mm which can further be reduced with the help of an aperture. The entire measurements were carried out at an angle of incidence of $62.2\pm2°$. The values of Δ and ψ are measured with a typical precision of $\pm0.02°$ at room temperature. Apart from polarization, the RI's of the crystal also depend on the direction of the incident angle of the light waves. Therefore, in order to get consistent results, the angle of incidence of the laser light is set near the Brewester's angle for each set of the given wavelength.

4. Results and discussions

KTP is a biaxial crystal which means it has two optic axes and three refractive indices n_x, n_y and n_z, respectively. Figure 1b shows the transmission curve of KTP crystal between 350 and 2600 nm wavelength. The I–A curves for n_x and n_y at He–Ne laser wavelength $\lambda = 632.8$ nm are given in figures 2a and 2b, respectively. These data are recorded at incident angles $\theta_0 = 60.4°$ and $\theta_0 = 60.6°$, respectively and shows excellent curve fittings of theoretical and experimental data. From figure 2a the calculated values of Fourier coefficients α and β are found to be 0.7506 ±0.02243 and -0.03148 ±0.01992, respectively which give corresponding values of $\tan\Psi = 1.0016$ and $\cos\Delta = -0.00006$. These values are substituted in eq. (1) to calculate the value of $\rho = -6.0096 \times 10^{-5}$. Using eqs (8) and (9) the values of refractive index (n) and anisotropic absorption coefficients (K) can easily be ascertained. The experimental values of n_x and K_x are estimated to be 1.7605 and 0.6654, respectively, whereas the theoretical value of n_x using Sellmeier eq. (10i) at 632.8 nm is 1.7635. The difference between the two results is $\Delta n_x = 0.003$. Similarly, from figure 2b the estimated value of $n_y = 1.7751$ and calculated value of

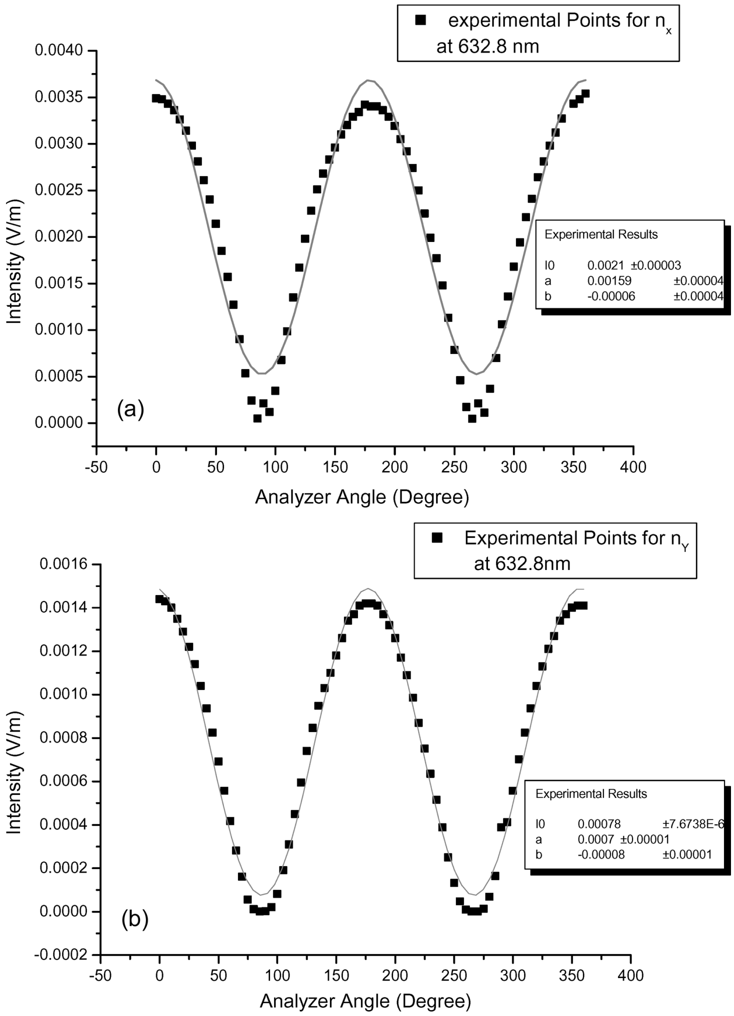


Figure 2 (**a**) and (**b**). The intensity vs. analyzer angle curves for KTP crystal plate at 632.8 nm wavelength. Here (■) shows the experimental data whereas (–) shows the theoretical curve.

RI using Sellmeier equation is $n_y = 1.7733$. The difference between the two results is $\Delta n_y = 0.0018$. The estimated value of the anisotropic absorption coefficient $K_y = 0.58037$.

Similarly, figures 3a and 3b show the I–A curves for n_x and n_y at semiconductor diode laser at $\lambda = 804.4$ nm at incident angles $\theta_0 = 60.2°$ and $\theta_0 = 60.4°$, respectively. The experimental and theoretical values of n_x, n_y and the differences Δn_x and Δn_y at different wavelengths along with their corresponding anisotropic absorption coefficients are summarized in table 1. The calculations in detail are discussed in the thesis of Molla [19].

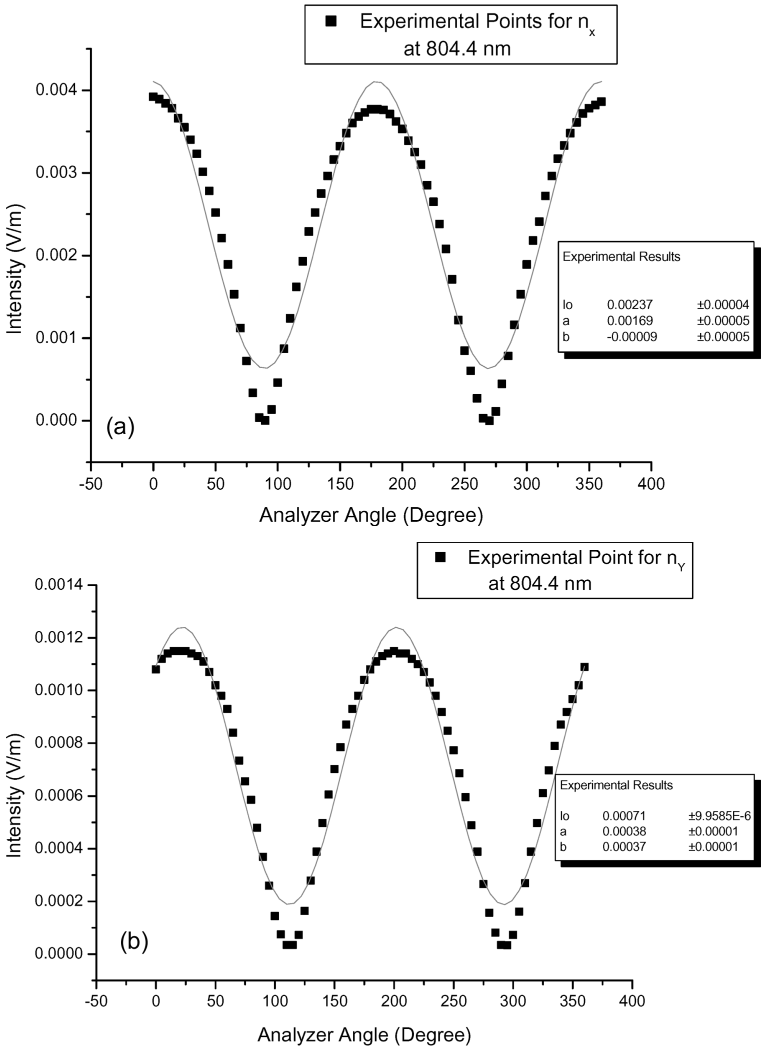


Figure 3 (**a**) and (**b**). The intensity vs. analyzer angle curves for KTP crystal plate at 804.4 nm wavelength. Here (■) shows the experimental data whereas (–) shows the theoretical curve.

Figures 4a and 4b show the dispersion curves of n_x and n_y for KTP crystals at different wavelengths. Here (■) are the experimental points whereas coloured lines show the theoretical values of eight existing Sellmeier equations given by different groups [10,18,20–26]. These equations are used to get best phase matching angles in different planes and different types of wavelength generation. Even though they are getting overlapped with each other, they also show large deviations at many wavelength regions too. Our experimental data for n_x agree with one theoretical

Table 1. A comparative chart of ellipsometrically measured optical parameters of KTP.

Wavelength (nm)	n_{x1} (Exp.)	n_{y1} (Exp.)	n_{x2} (Theo.)	n_{y2} (Theo.)	$\Delta n_x = n_{x1} - n_{x2}$	$\Delta n_y = n_{y1} - n_{y2}$	Absorption coefficient K_x	Absorption coefficient K_y
632.8	1.7605	1.77510	1.76350	1.77330	0.003	0.0018	0.6654	0.58307434
670.0	1.76369	1.76966	1.75969	1.75916	0.004	0.0005	0.6589	0.582174
804.4	1.7464	1.7594	1.7500	1.7588	0.0036	0.0006	0.6559	0.665994
808.8	1.7454	1.7590	1.7497	1.7585	0.0043	0.0005	0.6577	0.665918

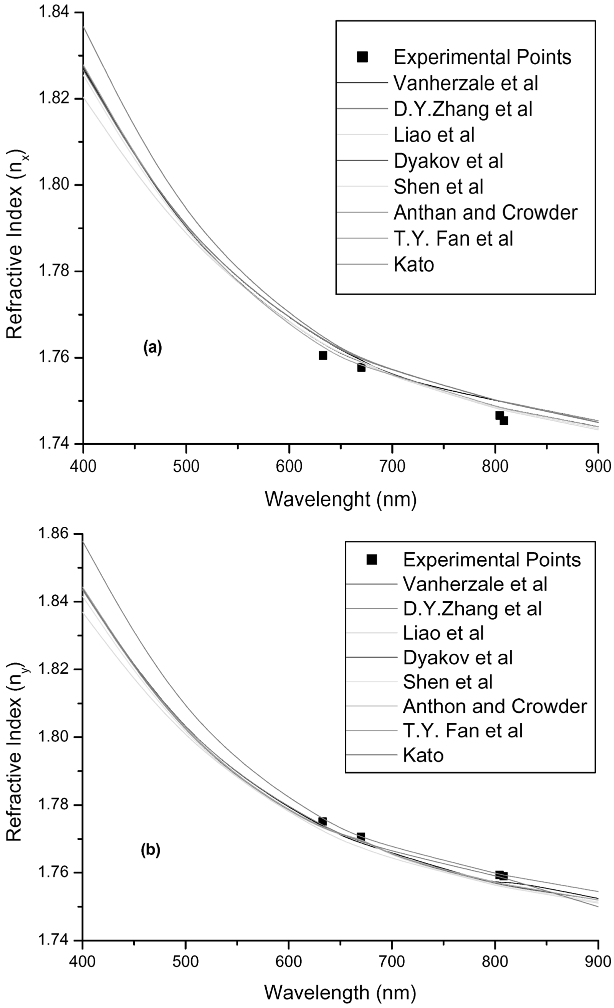


Figure 4 (**a**) and (**b**). The dispersion curves of n_x and n_y for KTP crystal plate at different wavelengths. Here (■) shows the experimental data whereas coloured lines (–) show the theoretical data based on the referred Sellmeier's equations.

value and very much close to the other theoretical values. But the experimental data of n_y are more accurate and overlapping the different curves too. Though we could not measure the values of n_z at different wavelengths due to poor surface quality of crystal, comparative dispersion curves of figures 4a and 4b confirm the reliability of our measurements. These results clearly show the superiority of reflection spectroscopic ellipsometry technique for the measurement of RI's of KTP crystal with great accuracy without using any specially designed prism. In addition, we have also shown the anisotropic absorption coefficients of KTP crystals for the first time.

5. Conclusions

We have successfully demonstrated the use of laboratory designed reflection ellipsometry technique to measure the refractive index of a thin plate of biaxial KTP crystal from visible to near infrared region using different wavelengths obtained from He–Ne and diode laser systems. Out of the three refractive indices, i.e. n_x, n_y and n_z, only n_z could not be measured due to poor quality of one of the surface of the crystal plate. We have also compared our measured values of refractive indices with the existing theoretical and experimental data of KTP crystal measured using conventional techniques. Our study shows that the precision in the measurement of refractive index n_x is found to be an order of 4.0062×10^{-5} while for n_y it is found around 1.7332×10^{-5}. In addition, we have also measured the anisotropic absorption coefficients of KTP at the aforesaid wavelengths which are found to be of the order of $\sim$0.6580.

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