Appl. Phys. B 67, 211–215 (1998)

Applied Physics B
Lasers
and Optics

Polarisation control of light by light in a nonlinear polymer

B. Pura[1], **J. Petykiewicz**[1], **L. Adamowicz**[1], **W. Jęda**[1], **M. Wierzbicki**[1], **K. Brudzewski**[2]

[1] Institute of Physics, Warsaw University of Technology, Koszykowa 75, 00-662 Warszawa, Poland
(Fax: +48-22/660-5447, E-mail: adamo@if.pw.edu.pl)
[2] Department of Chemistry, Warsaw University of Technology, Noakowskiego 3, 00-664 Warszawa, Poland

Received: 11 December 1997/Revised version: 11 May 1998

Abstract. Ellipsometric measurements of polarisation of a light beam, called the signal beam, transmitted through a thin polymer layer affected by the influence of a stronger light beam, called the pump, with variable intensity for different polarisation and wavelength are reported. Polarisation parameters of the transmitted signal beam depend nonlinearly on the intensity of the pump beam. An appropriate theoretical approach is proposed.

PACS: 42.70; 42.65; 78.65

Polymeric materials with nonlinear optical properties were first studied nearly 20 years ago. Since that time these materials have been at the forefront of current or emerging technologies. They are synthesised, characterised, and tested to evaluate their merits to make a variety of all-optical devices. Third-order nonlinear optical polymer devices are still well beyond the technological horizon. The first observation of third-harmonic generation in a polymer film was reported by Matsumoto et al. [1]. Some applications of third-order nonlinear optical materials in optical switching and logic systems are based on the intensity-dependent refractive index. This nonlinear effect mediates the interaction between two beams so that one powerful beam, called the pump, can control the propagation of a weaker beam, often called a probe, data, or signal beam.

The modified polyarylates (guest-host and side-chain systems) are nonlinear optical materials which may be implemented in optoelectronic devices. Their thermal, mechanical, and electrical parameters have been extensively discussed earlier [2, 3].

In this paper we report observed nonlinear optical behaviour of light polarisation in a plane-parallel plate of thickness L formed of such a polymer that has shown high optical stability and reproducibility at the beam power range used for our experiment. Standard parameters describing light polarisation have been measured by an original experimental setup specially designed for this purpose. We also present a preliminary theoretical analysis of the observed phenomenon of nonlinear polarisation variation as a function of light intensity.

1 Experimental

The layout of a new fully computerised experimental setup is presented in Fig. 1. As a source of signal light beam we use the argon laser (LEXEL 3000) working at wavelength $\lambda_1 = 5145$ Å with constant intensity of 50 mW and linear polarisation. It passes through the polymer layer of thickness $L = 10\,\mu$m and is registered by the ellipsometer. We have measured four standard parameters describing the signal beam polarisation, with an accuracy of 0.1°.

The pump beam, with wavelength $\lambda_2 = 6500$ Å and linear polarisation different from that of the signal beam, illuminates the polymer surface at an angle of an 15° relative to the signal beam direction of propagation. At that angle the measured effects have appeared most strongly. At the constant polarisation the intensity of the pump beam has been varied in the interval of 100 mW to 1 W.

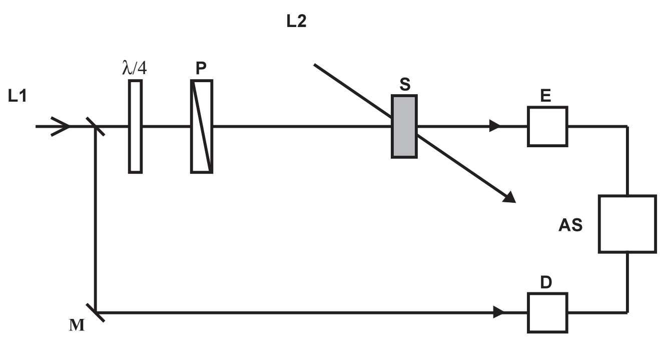

Fig. 1. Experimental setup. Argon laser L1 is a source of constant-intensity signal beam. Beam splitter and quarter-wave retarder are placed before polariser P. Strong and variable-intensity pump beam is emitted by laser L2 and passes through the sample S at a fixed angle relative to the signal beam. AS is the analysing system and D is the detector

2 Polymer

The method of synthesis and the structure of our material is described elsewhere [4]. The temperature of the glass transition was determined by the TMA method of Perkin and Elmer to be 89.6 °C. The intrinsic viscosity parameter characterising the masses of the polymer molecules $\eta = 900\ \mathrm{cm}^3/\mathrm{g}$ was determined by means of the Ostwald viscosimeter. The tensile strength of the film before crosslinking the polymer structure was $\sigma = 350\ \mathrm{kG/cm}^2$. The tensile strength after exposing the film for 2 min to UV lamp radiation for crosslinking is $\sigma = 480\ \mathrm{kG/cm}^2$. The solvent used to produce the film was tetrachloroethane + methylene chloride 1:1. The beginning decomposition temperature is about 275 °C. The diazo dye Orange 1D (OR1D) was dissolved into the polymer matrix in 10 wt.%.

3 Measurement scheme

Ellipsometry is an experimental technique that examines the polarisation state of light, expressed by angles Ψ and Δ, due to reflection or transmission through a layered structure [5]. The angles are determined by varying the ellipticity of the incident beam and reducing its intensity to zero by a rotatable polariser. The alternative method [6] which is applicable not only to a continuous-working optical source but also to pulsed sources is based on the simultaneous measurement of all four Stokes parameters of light. In order to measure all Stokes parameters simultaneously for a nanosecond duration it is obvious that mechanical or rotating ellipsometric methods are not applicable. The simultaneous measurement can be accomplished by using the optical system presented in Fig. 2. In this system, the state of polarisation of the light beam is determined by dividing it into four beams using a beam splitter and two beam-splitting Wollaston prism polarisers. The light fluxes of the four component beams are linearly registered by four photodetectors to produce four proportional electrical output signals. For each spectral line the vector of light flux $\boldsymbol{J}$ is related to the input Stokes vector $\boldsymbol{S} = (S_0, S_1, S_2, S_3)$ by the linear transformation

$$\boldsymbol{J} = \mathcal{M}\boldsymbol{S}\,, \tag{1}$$

where $\mathcal{M}$ is the instrument matrix. The unknown Stokes vector $\boldsymbol{S}$ is given by

$$\boldsymbol{S} = \mathcal{M}^{-1}\boldsymbol{J}\,, \tag{2}$$

where matrix $\mathcal{M}^{-1}$ is the inverse of the matrix $\mathcal{M}$. The instrument matrix $\mathcal{M}$ has to be non-singular ($\det|\mathcal{M}| \neq 0$) so that all four Stokes parameters are measurable. In order to reach a maximum of the precision of measurements the instrument matrix $\mathcal{M}$ has to be "well conditioned".

Generally, the instrument matrix $\mathcal{M}$ has sixteen elements that can be determined from a set of sixteen independent linear equations. These equations are generated by (2) when a polarised light source is used. Recording of electrical output signals that correspond to four different input polarisation states, described by four linearly independent Stokes vectors, is required. Usually, the set of polarisation states consists of three linearly polarised ones 45° apart in azimuth and one right (or left) circular polarisation state. The calibration process is essential for the accuracy of the instrument. After the calibration process is reached the accuracy of testing measurements of the instrument is presented in Table 1.

In this paper we use a convention that specifies the sign of the ellipticity angle to be positive and the fourth Stokes parameter to be negative whenever the instantaneous electric field vector describes a left-handed helix in space. The ellipse of this type of polarisation is described in a counterclockwise sense when viewed by an observer facing the source. The adoption of sign convention affects the relations among the various representations of polarisation state: the magnitudes and phases of orthogonal electric field components, coherence matrix, Stokes parameters, polarisation ratio, ellipsometric parameters (Ψ, Δ), azimuth χ (that is the angle of inclination of the long axis of the polarisation ellipse), and ellipticity γ (tg γ is the ellipticity ratio b/a of the length of axes of the polarisation ellipse). The relationships between Stokes parameters Ψ and Δ as well as χ and γ are given below.

$$\begin{aligned}
S_1/S_0 &= \cos 2\chi \cos 2\gamma\,,\\
S_2/S_0 &= \sin 2\chi \cos 2\gamma\,,\\
S_3/S_0 &= -\sin 2\gamma\,,\\
\cos 2\Psi &= -\cos 2\chi \cos 2\gamma\,,\\
\tan \Delta &= \tan 2\gamma / \sin 2\chi\,,\\
\tan 2\chi &= \tan 2\Psi \cos \Delta\,,\\
\sin 2\gamma &= \sin 2\Psi \sin \Delta\,.
\end{aligned} \tag{3}$$

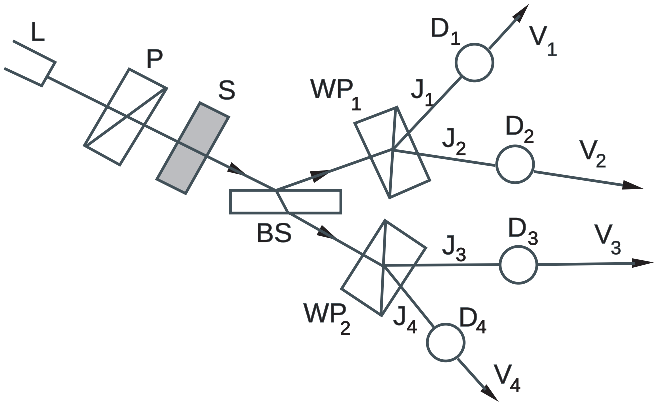

Fig. 2. The general scheme of the Stokes ellipsometer. BS is the beam splitter, WP1 and WP2 are Wollaston prisms, D1, D2, D3, and D4 are linear photodetectors that produce electrical output signals V1, V2, V3, and V4. L is the laser He−Ne light source, P is the polariser and S is the sample. The quarter-wave retarder was introduced during calibration of the instrument

Table 1. Various states of light polarisation represented in terms of measured Stokes vectors

Input state of polarisation	Measured Stokes vectors S_1/S_0	S_2/S_0	S_3/S_0
TE	−0.998	0.004	−0.006
TM	0.999	−0.003	−0.004
+45°	−0.002	0.991	0.008
−45°	−0.001	−0.995	0.006
near rc.	0.002	−0.004	0.999
near lc.	0.001	0.006	−0.997

The estimated accuracies of Ψ and Δ are $\delta\Psi = 0.05°$ and $\delta\Delta = 0.1°$, respectively.

4 Results

The absorption coefficient of the polymer film of thickness $L = 10\,\mu\text{m}$ as a function of the wavelength in the range of 350 nm to 800 nm is presented in Fig. 3. The maximum of absorption in the polymer layer corresponds to wavelengths less than 400 nm in the ultraviolet region. In the interval from 500 nm to 700 nm, where the two-wave interaction was registered, the absorption and the thermal effects are negligible.

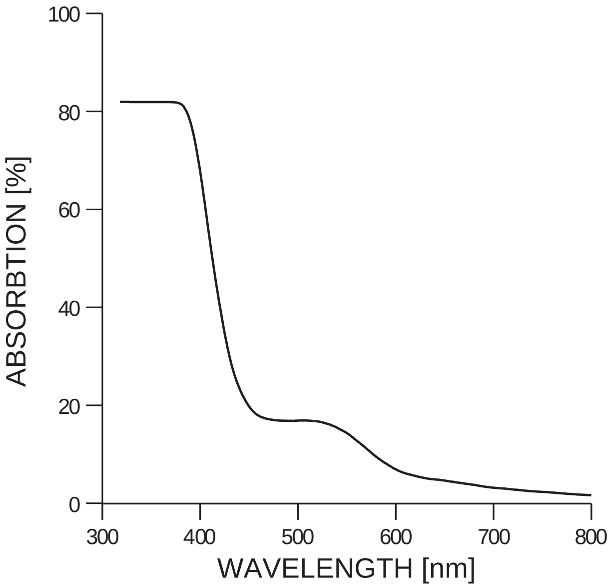

Fig. 3. The absorption coefficient of the 10 μm polymer film versus wavelength in the interval 350 nm to 800 nm

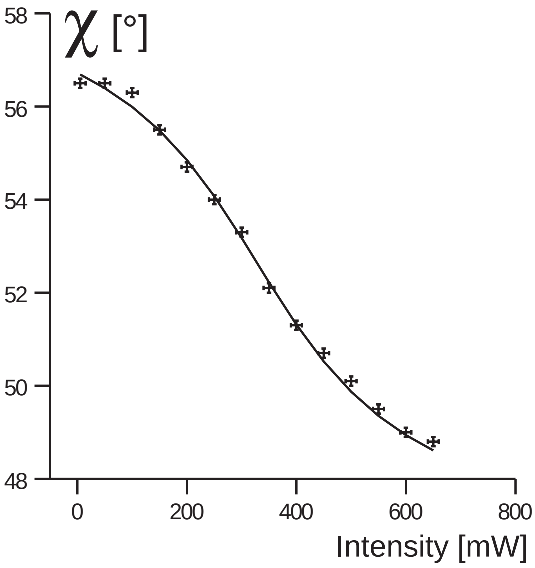

Fig. 4. Transmission characteristics of the parameter χ. The output polarisation state of the signal wave is controlled by the intensity of the pump wave. Experimental points are compared with theoretical *solid curve* described by (13)

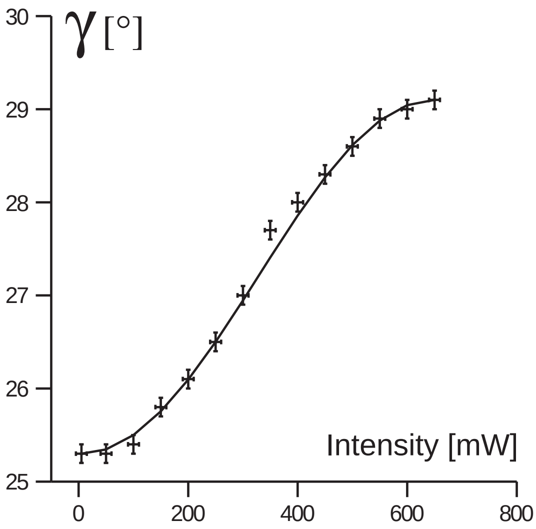

Fig. 5. Transmission characteristics of the parameter γ. The output polarisation state of the signal wave is controlled by the intensity of the pump wave. Experimental points are compared with theoretical *solid curve* described by (14)

The dependence of the polarisation variations due to the two-wave interaction in a nonlinear medium is shown in Figs. 4 and 5. The polarisation was determined by the measurement of the four different parameters. The variations of the angle χ for the signal beam L1 with wavelength λ_1 are presented in Fig. 4. It decreases abruptly by 5° during the 400 mW change of the L2 beam with wavelength λ_2. The smallest variation we have observed is that one for the parameter γ. The same increase of 400 mW in the intensity of the pump beam L2 with the wavelength λ_2 has only produced the γ increase of 2°, as presented in Fig. 5.

5 Theory

To explain the polarisation variation of light in the studied polymers we assume that the considered medium is characterised by isotropic optical nonlinearity of the third order. Nevertheless, despite its simplifications the model describes the main features of physical interaction mechanisms of light with the polymer. The influence of the "cross-modulation" effect on the intensities and polarisations of two monochromatic waves interacting within a nonlinear isotropic Fabry–Pérot cavity has been studied earlier [7, 8]. The problem of the cross-modulation effect with frequencies ω_1 and ω_2 is solved in the form of a nonlinear response on an external EM field

$$\boldsymbol{P}_j^{\mathrm{NL}} = \left(\boldsymbol{\chi}_j^{(3)} \vdots \boldsymbol{E}_j \boldsymbol{E}_j^* + \boldsymbol{\chi}_{3-j}^{(3)} \vdots \boldsymbol{E}_{3-j} \boldsymbol{E}_{3-j}^* \right) \boldsymbol{E}_j \,, \tag{4}$$

where $j = 1, 2$. We use the abbreviations for electric polarisation $\boldsymbol{P}_j^{\mathrm{NL}} = \boldsymbol{P}^{\mathrm{NL}}(\omega_j)$ and electric field vector $\boldsymbol{E}_j = \boldsymbol{E}(\omega_j)$. The dielectric susceptibility tensor (of 4th rank) $\boldsymbol{\chi}_j^{(3)} = \boldsymbol{\chi}^{(3)}(\omega_j)$ describes the 3rd-order optical nonlinearity. Its influence on the intensity and polarisation states of two monochromatic plane waves with different frequencies can be studied. In the paper [8] the following problem was considered: two light waves illuminate perpendicularly the $z < 0$ side of the plate made from a medium with the Kerr nonlinearity. The electric vectors,

$$\boldsymbol{E}_j^0 = \boldsymbol{e}_x\, E_{xj}^0 + \boldsymbol{e}_y\, E_{yj}^0, \quad j = 1, 2\,, \tag{5}$$

oscillate with different frequencies ω_j. These two waves mutually interact inside the nonlinear medium of the cavity. Their multiple reflections at the inside boundaries of the cavity result in transmitted waves with electric vectors

$$\boldsymbol{E}_j^{\mathrm{T}} = \boldsymbol{e}_x\, E_{xj}^{\mathrm{T}} + \boldsymbol{e}_y\, E_{yj}^{\mathrm{T}}, \quad j = 1, 2\,. \tag{6}$$

The Cartesian components of the transmitted waves are given by the following expressions

$$\begin{aligned} E_{xj}^{\mathrm{T}} &= \frac{\eta_j T}{G_j} E_{xj}^0 \exp \mathrm{i}[\phi_{xj}^0 - \phi_{xj}^{\mathrm{F}}(0)]\,, \\ E_{yj}^{\mathrm{T}} &= \frac{\eta_j T}{G_j} E_{yj}^0 \exp \mathrm{i}[\phi_{yj}^0 - \phi_{yj}^{\mathrm{F}}(0)]\,, \end{aligned} \tag{7}$$

where

$$G_j = 1 - \eta_j^2 R \exp(\mathrm{i}\, Q_j) \tag{8}$$

and

$$\begin{aligned} Q_j =& \zeta_j \left[9(E_{xj}^{\mathrm{T}})^2 + 4(E_{yj}^{\mathrm{T}})^2\right] \\ &+ \zeta_{3-j}\left[12(E_{x(3-j)}^{\mathrm{T}})^2 + 4(E_{y(3-j)}^{\mathrm{T}})^2\right] - 2k_j L\,. \end{aligned} \tag{9}$$

The parameter $\eta_j = \exp(-\alpha_j L)$ where α_j is a linear dumping coefficient. The quantities ζ_j and ζ_{3-j} are defined as an invariant product of the nonlinear susceptibility tensor and electric field unit vectors $\boldsymbol{e}_j$ and $\boldsymbol{e}_{3-j}$, respectively, of interacting waves and represent wave phase-change coefficients inside the nonlinear medium. The quantity ζ_{3-j} is responsible for "cross-modulation" of interacting waves and depends on relative orientation of unit vectors $\boldsymbol{e}_j$ and $\boldsymbol{e}_{3-j}$. In our case it was verified experimentally that ζ_{3-j} gets maximum value at the angle 15° between the pump and the signal beam of given polarisations. $\phi_{xj(yj)}^0$ are phases of the incident wave and $\phi_{xj(yj)}^{\mathrm{F}}(0)$ are phases of the forward-propagating waves for $z = 0$, R is the intensity reflection coefficient at the boundary planes of the nonlinear layer, $T = 1 - R$ is the intensity transmission coefficient and k_j are the magnitudes of the plane wave-propagation vectors in the cavity medium.

The polarisation state of the transmitted wave can be described by the coherence matrix $\mathcal{I}_j$

$$\begin{aligned} \mathcal{I}_j &= \begin{bmatrix} I_{(xx)j}\, I_{(xy)j} \\ I_{(yx)j}\, I_{(yy)j} \end{bmatrix} \\ &\equiv \begin{bmatrix} E_{xj}^{\mathrm{T}}\, E_{xj}^{\mathrm{T}*} & E_{xj}^{\mathrm{T}}\, E_{yj}^{\mathrm{T}*} \exp\left[\mathrm{i}\,\Delta_j\right] \\ E_{xj}^{\mathrm{T}*}\, E_{yj}^{\mathrm{T}} \exp\left[-\mathrm{i}\,\Delta_j\right] & E_{yj}^{\mathrm{T}}\, E_{yj}^{\mathrm{T}*} \end{bmatrix}, \end{aligned} \tag{10}$$

where

$$\Delta_j = Q_j + 2k_j L \tag{11}$$

is the phase difference between the x and y component of the jth forward propagating wave. Calculations performed in [8] show that linearly polarised input waves $E_{yj}^0/E_{xj}^0 = \mathrm{const}_j$, interacting mutually (cross-modulation) with the second wave [this is maintained by the form of (7)–(9)], change their initial polarisation state. The elements of the coherence matrix allow us to determine the parameters of the polarisation ellipse χ_j and γ_j for each of the two frequencies ω_j. The following relations hold [9].

$$\begin{aligned} \tan\, 2\chi_j &= (I_{(xy)j} + I_{(yx)j})(I_{(xx)j} - I_{(yy)j})^{-1}, \\ \sin\, 2\gamma_j &= \mathrm{i}\,(I_{(xy)j} - I_{(yx)j})(I_{(xx)j} + I_{(yy)j})^{-1}. \end{aligned} \tag{12}$$

As in [8] one can state that for large values of intensities of the pump beam the ellipse parameters exhibit a bistable character that, in a special case, appears as differential dependence [10], similar to the case discussed elsewhere [11].

Let us assume $\eta_j^2\, R \ll 1$, which means $G_j \approx 1$. From (7) and (10) one gets

$$\begin{aligned} I_{(\alpha\alpha)j} &= \eta_j^2 T^2 I_{(\alpha\alpha)j}^{(0)}\,, \\ E_{xj}^{\mathrm{T}}(E_{yj}^{\mathrm{T}})^* &= \eta_j^2 T^2 \sqrt{I_{(xx)j}^{(0)} I_{(yy)j}^{(0)}}\,, \end{aligned}$$

where $I_{(\alpha\alpha)j}^{(0)}$ are intensities connected with the amplitudes of incoming waves for $\alpha = x, y$, respectively.

According to (12), the dependencies of χ_j and γ_j are given by the following relations.

$$\chi_j = (1/2)\tan^{-1}[A_{(-)} \cos \Delta_j]\,, \tag{13}$$

$$\gamma_j = (1/2)\sin^{-1}[A_{(+)} \sin \Delta_j]\,, \tag{14}$$

where

$$A_{(\pm)} = 2\sqrt{I_{(xx)j}^{(0)} I_{(yy)j}^{(0)}}\, \left(I_{(xx)j}^{(0)} \pm I_{(yy)j}^{(0)}\right)^{-1}$$

and Δ_j depends on the incident wave intensity (cf. (11)). The experimental data representing the dependence of χ_1 and γ_1 parameters on the pump-wave intensity are compared in Figs. 4 and 5 with the theoretically predicted behaviour of $\tan^{-1}$ and $\sin^{-1}$ according to (13) and (14), respectively. More extensive comparison of experiment and theory would require detailed knowledge of material characteristics, which is not available at the moment.

6 Conclusion

Third-order optical nonlinearity in new polymer material has been investigated. The nonlinear polarisation effects observed here are produced by the optical dispersion of the medium and only to a very small degree by the temperature changes of the polymer layer. The intensity variation of polarisation and nonlinear coupling of TE and TM polarised waves may be useful in a suggested [12] new class of optical devices as optically activated polarisation switches or phase-sensitive discriminators as well as externally controlled TE – TM mode couplers for nonlinear planar waveguides. The observed effect relies on preferential transmission of one polarisation state of the input signal beam with simultaneous detuning for orthogonal component polarisation by suitable adjustment of the pump beam intensity. The change of the polarisation parameters χ and γ with the variation of intensity of the pump beam by 200 mW indicates possible application of the polyarylate polymer films to the manufacture of photonic converters. However, the observed effect is not strong enough for immediate applications. Optical stability of the studied polymer, its tailorable nonlinear susceptibility and ability to make thin films and fibres encourages further research. A quite realistic solution to enhance the observed nonlinearity might be to place the present polymer film between two Bragg reflectors, thus forming a resonant cavity or, in other words, a defect of periodicity in one-dimensional photonic structure [13].

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