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# Prism coupling technique investigation of elasto-optical properties of thin polymer films

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The use of thin polymer films in optical planar integrated optical circuits is rapidly increasing. Much interest, therefore, has been devoted to characterizing the optical and mechanical properties of thin polymer films. This study focuses on measuring the elasto-optical properties of three different polymers; polystyrene, polymethyl-methacrylate, and benzocyclobutane. The out-of-plane elastic modulus, refractive index, film thickness, and birefringence of thin polymer films were determined by means of the prism coupling technique. The effect of the applied stress on the refractive index and birefringence of the films was investigated. Three-dimensional finite element method analysis was used so as to obtain the principal stresses for each polymer system, and combining them with the stress dependent refractive index measurements, the elasto-optic coefficients of the polymer films were determined. It was found that the applied stress in the out-of-plane direction of the thin films investigated leads to negative elasto-optic coefficients, as observed for all the three thin polymer films. © 2004 American Institute of Physics. [DOI: 10.1063/1.1812823]

## I. INTRODUCTION

Over the last decade, thin polymer films have attracted an increasing interest for applications in microelectronic industry and in integrated optical components due to their ease of processing, low dielectric constant, and useful electro- and stress-optic properties.<sup>1-3</sup> The optical properties of thin polymer films are of great importance in optoelectronic and especially in integrated optical components and circuits. These properties are strongly influenced by the stresses inherent in the films. The stresses are generated due to the interaction of the polymer with the substrate, solvent evaporation induced shrinkage, etc.<sup>4</sup> For optical waveguide devices, deposited onto substrates, the mechanical properties of thin polymer films may differ by several orders of magnitude from those of bulk materials.<sup>4</sup> Furthermore, it is known that the stresses generated in the films cause the polymer chains to orient in the plane of the film resulting in anisotropic optical properties. Therefore, it is important to be able to measure mechanical and optical properties of thin polymer films simultaneously.

There are a number of recent studies that have analyzed polymeric films showing in-plane and out-of-plane optical anisotropy (birefringence).<sup>5,6</sup> In addition, there are also studies investigating the mechanical properties of thin polymer films, most of which are limited only to the in-plane direction. This is due to the difficulty of measuring small thickness changes of thin films in the out-of-plane direction.<sup>7</sup> The refractive index measurements of thin films ( $t < 10 \mu\text{m}$ ) are usually done by making use of ellipsometry or prism coupling methods; the latter is preferred due to the higher accuracy and capability to measure birefringence. For the out-of-

plane mechanical properties, on the other hand, there are several methods that have been reported recently. Kumer *et al.* measured the out-of-plane modulus by Brillouin scattering technique, but the method is limited to minimum film thickness of  $100 \mu\text{m}$ .<sup>8</sup> Another method is the parallel plate capacitor method, where metal layers are deposited onto polymer surfaces, which could modify the mechanical properties of the film.<sup>9</sup> Other approaches include use of nanoindenter,<sup>10</sup> atomic force microscopy,<sup>11</sup> and precision capacitance dilatometry.<sup>12</sup> While all of these methods measure the out-of-plane mechanical properties of the polymer films, they do not measure optical properties simultaneously. Therefore, the relation between the stress and the optical properties can only be established indirectly for thin polymer films. While much of the above mentioned shortcomings may be overcome with the use of prism coupling technique, the effect of applied stress on the measured quantity during the use of prisms to couple light into the film have not, so far, been taken into account.

In this study, an approach is proposed for elasto-optic characterization of thin polymer films, making use of the well known prism coupling technique.<sup>13</sup> This method allows us to determine the optical anisotropy and out-of-plane mechanical properties and to correlate both in order to obtain the elasto-optical properties of thin polymer films.

## II. EXPERIMENT

In this study poly (methyl methacrylate) (PMMA), polystyrene (PS), and benzocyclobutane (BCB) polymers were used. The BCB polymer was Dow Chemical Cyclotone 3022-46; PMMA and PS were obtained from Sigma-Aldrich with molecular weights of 15 000 and 150 000, respectively. The PMMA and PS were dissolved in chloroform with 15

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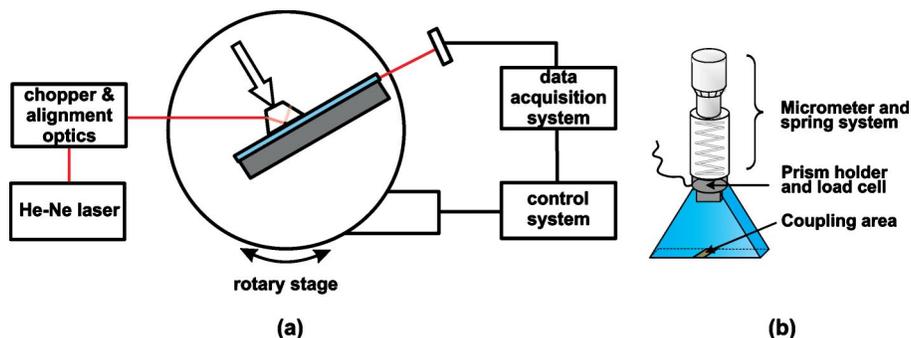


FIG. 1. (a) Schematic representation of the experimental arrangement of the prism coupling setup, (b) detailed representation of the system used for pressure application and readout.

wt % and 6 wt %. The PMMA, PS, and BCB polymer solutions were spin coated onto substrates for 40 sec at 3000 rpm, 2000 rpm, and 5000 rpm, respectively. The substrates were  $3\text{ cm} \times 1.5\text{ cm}$  thermally oxidized silicon samples with a  $\text{SiO}_2$  layer of  $7.2\text{ }\mu\text{m}$  thickness. Upon coating, the PMMA and PS films were cured in an oven in nitrogen atmosphere at  $110\text{ }^\circ\text{C}$  for 30 min and the BCB films were treated similarly at  $250\text{ }^\circ\text{C}$  for 60 min. All of the resulting polymers were about  $2.3\text{ }\mu\text{m}$  thick, as verified by stylus profilometer (Sloan Dektak 3030ST) thickness measurements.

The refractive index, birefringence, films thickness, and out-of-plane modulus of the polymer films were all determined at room temperature ( $21\text{ }^\circ\text{C}$ ) by a prism coupling setup which was built in our laboratory. A schematic representation of the setup is illustrated in Fig. 1(a). A linearly polarized He-Ne laser ( $\lambda=632.8\text{ nm}$ ) is used to excite the modes in the polymer slab waveguides in transverse electric (TE) and transverse magnetic (TM) polarizations. The waveguide and the coupling prism are mounted on a computer-controlled motorized rotary stage. As the stage rotates, the guided mode intensity is measured on the output of the waveguide for each polarization by a detector as a function of the incidence angle and is stored in a computer. The coupling angles are then used to find the index of refraction and thickness of the guiding polymer layer for TE and TM polarizations by a computer program.<sup>13–16</sup> All the measurements are performed by using symmetric SF-14 prisms with base angles of  $60^\circ$  and refractive index of 1.7561 ( $\lambda=632.8\text{ nm}$ ). The coupling pressure is adjusted by a micrometer in contact with a calibrated spring system and a load cell that allows us to monitor the applied force onto the prism. The typical error in the refractive index measurements is found to be less than  $\pm 0.0002$  and  $\pm 0.3\%$  for the thickness measurements.

Since the introduction of the prism coupling method by Tien and Ulrich, it has been argued that the coupling strength can cause shifting and broadening of the modes due to presence of the prism in the vicinity of the film.<sup>13,14</sup> This was attributed to the decrease of the air gap thickness with the applied pressure that originates from the dust particles and is located between the prism and the film. Recently, Monneret *et al.* made a theoretical analysis to estimate the coupling angle shifts observed in m-lines prism coupling method.<sup>17</sup> For  $\text{SrTiO}_3$  prism and film with refractive index of 2.27 they have found that the shifts result in an index change of

$5 \times 10^{-5}$ , which is well below our precision. In our case, the prism used is of lower refractive index and the index contrast between the film and the prism is low, which should result in even smaller changes in the refractive index.<sup>14</sup> Moreover, the decrease of the air layer thickness and thus of the coupling efficiency is a function of the mechanical properties of the film under investigation. In our case, the Young's moduli of the polymers are few orders of magnitude smaller than that of the ordinary glassy dielectric materials, which makes them softer.<sup>18</sup> Therefore, it is reasonable to assume that the reduction of the air gap thickness with applied pressure is small in the range of the thickness variations of the polymer films used in this study.

The out-of-plane elastic moduli of the thin polymer films were deduced by using the same prism coupling setup. The thickness change was used together with the applied stress information in order to obtain the corresponding stress-strain curves. Use of finite element calculations was made to establish connection between measured values and elastomechanic properties, such as the Young's modulus. Since the polymers under investigation are viscoelastic, both plastic and elastic deformations may be expected to occur during the loading process. Therefore, all measurements of out-of-plane modulus and refractive index were performed during the unloading process.<sup>12</sup>

### III. RESULTS AND DISCUSSIONS

In order to be able to mutually compare the stress effects on the guiding films, the polymer layer thicknesses were adjusted to be nearly identical. Film thicknesses had values of  $2.37\text{ }\mu\text{m}$  for PS,  $2.38\text{ }\mu\text{m}$  for BCB, and  $2.30\text{ }\mu\text{m}$  for PMMA layers, as verified by both stylus profilometer and prism coupling methods, being in good agreement. These thicknesses were chosen so that there are at least two guided modes in the waveguides. The mode calculations were done by solving the Maxwell's equations with the corresponding boundary conditions.<sup>19</sup> The number of modes calculated in this manner was further confirmed by calculating the mode spectrum of the same slab waveguides using beam propagation techniques employing finite difference methods. The calculations predicted four modes (both TE and TM) for the BCB, four modes for PS, and three modes for PMMA slab waveguides. The predictions were in agreement with the observed number of modes.

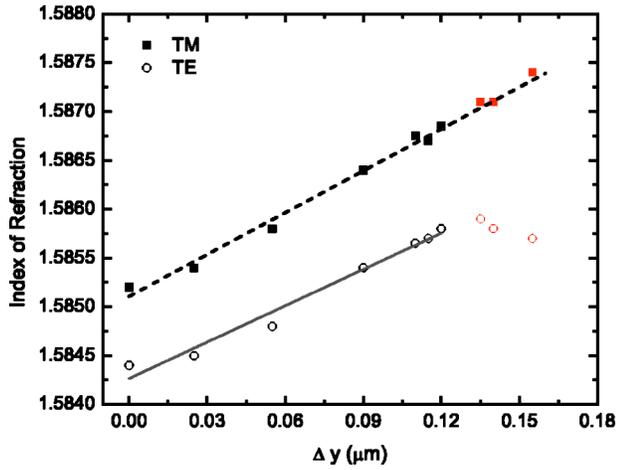


FIG. 2. Change of the TE and TM refractive index for PS films with change in the film thickness.

The results of refractive index variation versus the thickness change with the applied pressure for PS films are given in Fig. 2. A steady increase of the refractive index is clearly observed for both polarizations. The out-of-plane refractive index  $n_{TM}$  increased from a value of 1.5852 up to a value of 1.5874 as the film thickness decreased by 0.15  $\mu\text{m}$  with the applied pressure. Similarly, the in-plane refractive index  $n_{TE}$  increased from 1.5844 to 1.5856 in the same range. At higher applied stresses, there is a flat region, where the in-plane refractive index does not increase with the applied stress. This may be attributed to the saturation of the alignment of molecules contributing to refractive index as well as the saturation of the density variation of dipoles contributing to index of the polymer in the in-plane direction. On the other hand, the density of the polymer continues to increase with the applied pressure leading to an increase in the refractive index for the out-of-plane direction. The flat region was not taken into account in the calculations of the elasto-optical constants, to be discussed below. Similar trends for refractive index increase were observed for both BCB and PMMA layers. Figure 3 shows the results obtained for BCB films. An increase for both polarizations is observed with the decrease

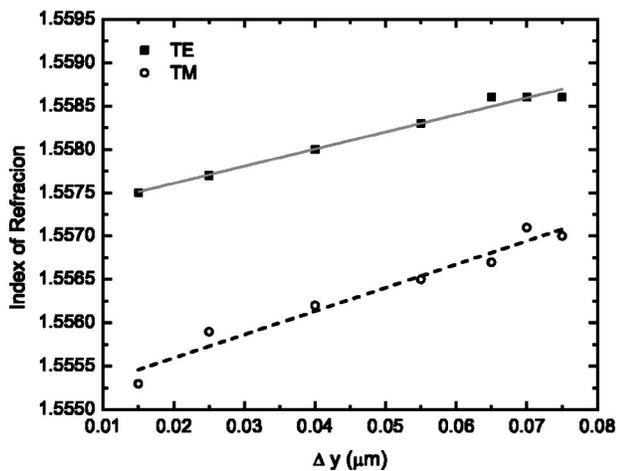


FIG. 3. Change of the TE and TM refractive index for BCB films with change in the film thickness.

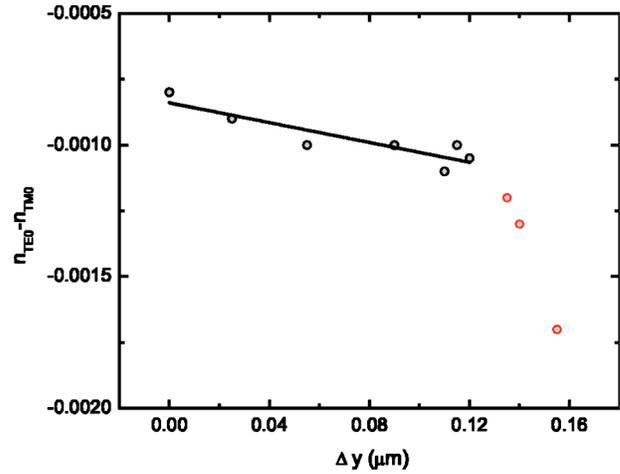


FIG. 4. Change of birefringence for PS films with change in the film thickness.

of the film thickness as pressure is applied on the polymer layer. The value of  $n_{TE}$  increased from 1.5575 up to a value of 1.5586 as the thickness of the guiding film was reduced by about 0.07  $\mu\text{m}$  with the applied pressure, while  $n_{TM}$  has increased from 1.5553 to 1.5566 in the same range. As for the PMMA layers, the refractive index was measured to increase from about 1.4869 to 1.4876 for both polarizations.

Another interesting outcome of the refractive index measurements is the evolution of birefringence ( $\Delta n = n_{TE} - n_{TM}$ ) with the applied pressure. The birefringence for the PS films was measured to be negative (see Fig. 4) with an initial value of about  $-0.0007$  which increased slightly in the negative direction up to a value of about  $-0.0011$ , as the film thickness decreased by 0.12  $\mu\text{m}$ , with applied pressure. BCB layers have also been measured to be birefringent, however, with a positive sign (see Fig. 5). For these layers, the value of the birefringence was initially about 0.0022, which decreased slightly down to 0.0017 with the film thickness decrease of 0.07  $\mu\text{m}$ , as a result of the applied pressure. On the other hand, the PMMA films have been found to be optically isotropic. Zero birefringence was observed in the unstressed condition and there was no change in the birefringence as the

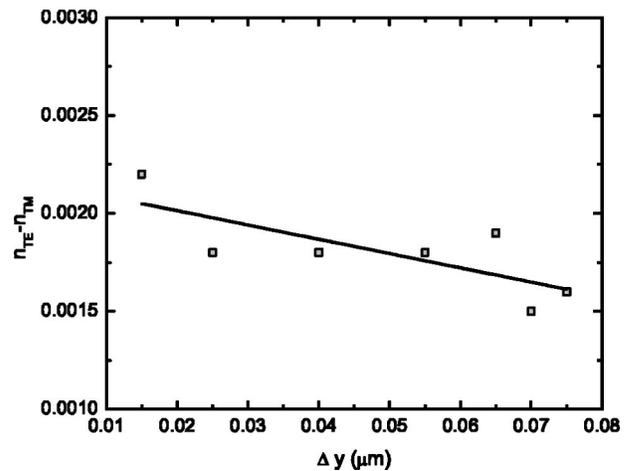


FIG. 5. Change of birefringence for BCB films with change in the film thickness.

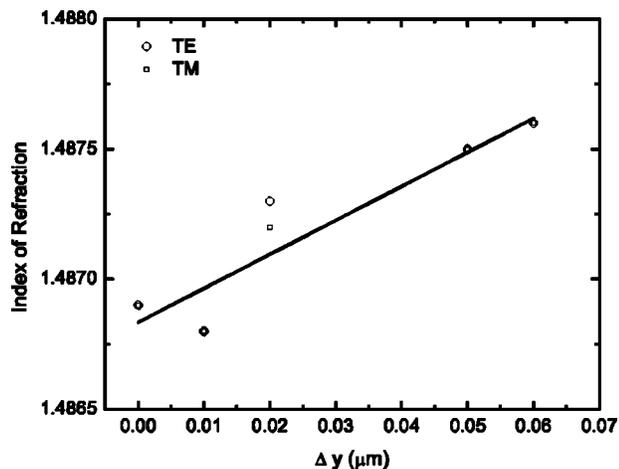


FIG. 6. Change of birefringence for BCB films with change in the film thickness.

pressure was increased (see Fig. 6), although both  $n_{TE}$  and  $n_{TM}$  showed small increases with applied pressure.

The refractive index values determined from these measurements are in agreement with those reported in the literature. The refractive indices of  $n_{TE}=1.587\pm 0.002$  and  $n_{TM}=1.582\pm 0.002$  were previously reported for the atactic PS films having negative birefringence that are in good agreement with our results.<sup>20</sup> For the BCB films, to the best of our knowledge, the number of polarization dependent refractive index values reported is very limited. For BCB type similar to ours the only measurement was reported by Tanikella *et al.*, where it was reported that  $n_{TE}=1.55$  with a positive birefringence of  $\Delta n=0.002$ .<sup>21</sup> These values are also in a very good agreement with ours. As for the PMMA layers, they are known to be optically isotropic, and White *et al.* reported a refractive index of 1.488, identical with our measurements.<sup>22</sup> However, in all of these reports no mention of stress induced changes in the quantities measured were made. Finally, the measurements that we report for PS, BCB, and PMMA have higher precision from those reported in literature.

The out-of-plane elastic moduli for the polymer films were calculated by using the applied stress and film thickness measurements. In calculation of the applied stress the area considered was the coupling area having size of  $8\text{ mm} \times \sim 1\text{ mm}$  at the base of the coupling prism. The size of the coupling area was measured by inspecting the dark region formed at the base of the prism. The value of the force was measured by a load cell and ranged approximately between 0 and 150 N. The coupling pressure was adjusted by a micrometer in contact with a calibrated spring system [Fig. 1(b)]. As seen from the figure, a load cell was incorporated into the prism attachment setup. The load cell was Sensotec Model 13 (AL322BN) subminiature compression only load cell allowing measurement of loads of up to 50 lbs.

The values obtained for out-of-plane elastic moduli were  $0.3\pm 0.1 \times 10^9\text{ N/m}^2$ ,  $0.3\pm 0.1 \times 10^9\text{ N/m}^2$ , and  $0.9\pm 0.1 \times 10^9\text{ N/m}^2$  for PS, BCB, and PMMA thin films, respectively. The elastic moduli obtained for the investigated three polymer layers are approximately one order of magnitude smaller than those reported in the literature for their bulk counterparts. For PS films, the corresponding value is re-

ported to be about  $(2-3) \times 10^9\text{ N/m}^2$ .<sup>23,24</sup> Most of the films studied in the literature were bulk PS specimens with typical dimensions of the order of millimeters. The moduli were measured under tensile stress in stretching condition corresponding to an in-plane modulus. Thus, we can expect these values to differ from our thin film results in the out-of-plane direction. For PMMA layers, the reported Young's moduli are  $\approx 3.2 \times 10^9\text{ N/m}^2$ ,<sup>16,24</sup> again measured for bulk material under the in-plane stretching condition. As for the BCB films, there are several studies reporting on the out-of-plane modulus of thin BCB films ( $\sim 15\text{ }\mu\text{m}$ ) with value of  $3 \times 10^9\text{ N/m}^2$ .<sup>9,25</sup> In these reports the authors made use of parallel plate capacitor method, where metal layers are deposited onto polymer surfaces, which may result in modification of the mechanical properties of the film itself.

Although there are studies that report moduli of thin films close to their bulk counterparts, a number of studies were also done analyzing the thickness dependence of the out-of-plane mechanical properties of thin polymer films. They report clear differences between thin film and bulk polymers. For example, Liou *et al.* measured out-of-plane elastic properties for thin polyimide films as a function of thickness using precision capacitance dilatometer.<sup>12,26</sup> They have found that the out-of-plane elastic moduli of the thin polymer films decrease with decreasing film thickness. For polyimide films, Liou *et al.* obtained one order of magnitude decrease from about  $2.8 \times 10^9\text{ N/m}^2$  down to  $0.3 \times 10^9\text{ N/m}^2$  in the out-of-plane modulus as the film thickness decreased from  $\sim 15\text{ }\mu\text{m}$  to  $\sim 4\text{ }\mu\text{m}$  and verified that the in-plane modulus has a larger value. A similar behavior of modulus anisotropy and decrease of the out-of-plane modulus with decreasing thickness can be expected for the polymer films studied in this work. This possibility was tested by performing out-of-plane modulus measurements for PMMA films of various thicknesses. In this series PMMA was dissolved in chlorobenzol and was spincoated at 2000–5000 rpm in order to obtain varying thickness films, which were not obtainable by using chloroform as solvent. Five PMMA films with thicknesses of 3.10, 3.22, 4.30, 4.75, and  $6.10\text{ }\mu\text{m}$  were obtained. The values of out-of-plane moduli for these films were obtained as described above. Figure 7 shows the experimental data for variation of  $\Delta y/y_0$  with the applied force  $F$ . As is observed, the slopes of the graph increase with increasing film thickness. For all the polymer films a minimum force of about 15 N is required to obtain strain larger than zero, i.e., to obtain coupling between the prism and polymer film. Thus the variation of  $\Delta y/y_0$  with the applied force can only be observed for applied force of about 15 N or greater. As the value of strain increases the variation becomes linear due to the stabilization of the coupling area between the prism and the film and this is the region which was used to determine the out-of-plane elastic moduli of the layers. The slopes are directly proportional to the out-of-plane moduli of the films and therefore our expected trend has been verified. The calculated out-of-plane moduli variation with film thickness for PMMA layers are depicted in Fig. 8. The geometry and coordinate axis convention is illustrated in the inset of the figure. It is evident from the figure that the out-of-plane moduli of the layers show a

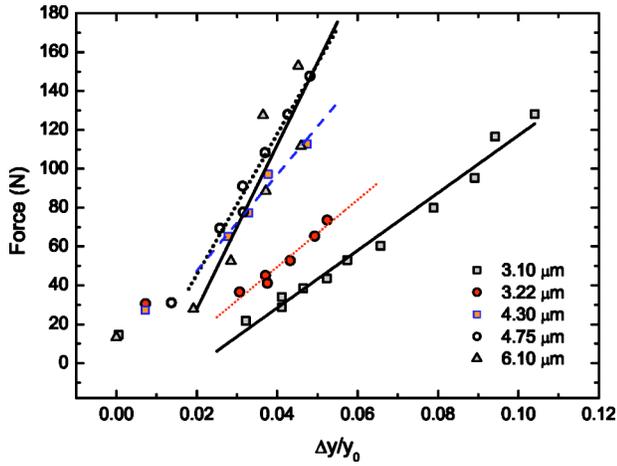


FIG. 7. Experimental data for variation of  $\Delta y/y_0$  with the applied force  $F$  for PMMA films with thicknesses of 3.10, 3.22, 4.30, 4.75, and 6.10  $\mu\text{m}$ . Note that the coupling is completed for  $F > 15$  N.

steady increase with increasing film thickness. Namely, the modulus showed an increase from  $0.18 \pm 0.03 \times 10^9 \text{ N/m}^2$  up to  $0.5 \pm 0.1 \times 10^9 \text{ N/m}^2$  with increase in thickness from 3.10  $\mu\text{m}$  to 6.10  $\mu\text{m}$ . Therefore, the analysis of the change of the out-of-plane elastic modulus with change in film thickness has two important results. First, as was expected, the moduli increase with increasing film thickness. If the increase is assumed to be linear, the moduli are expected to reach their bulk value at a thickness of about 27  $\mu\text{m}$ . The second finding that should be mentioned is the effect of the solvent used in preparation of the films. It was found that the PMMA films produced using chloroform as solvent resulted in larger out-of-plane elastic moduli compared to the ones prepared using chlorobenzol.

The procedure for determination of the elasto-optical properties of the polymer films includes finite element method (FEM) calculations of the unknown stress components together with the results of experimental measurements of the index and thickness change with the applied stress. The theoretical basis of the procedure is described below.

The stress dependence of the refractive index is expressed in terms of the Fresnel's index ellipsoid and the

Cauchy's stress ellipsoid. In the elastic range of the material and when the ellipsoids are coaxial, the principal refractive indices are related to the principle stresses by the Neumann-Maxwell stress equations

$$n_i = n_0 + C_1 \sigma_i + C_2 (\sigma_j + \sigma_k), \tag{1}$$

where  $n_i$  are the principal refractive indices,  $\sigma_i$  are principal stresses, and  $C_1$  and  $C_2$  are the elasto-optic coefficients.<sup>27,28</sup> The difference  $(C_1 - C_2) = C$  is the usually reported and widely used value for the stress-optic coefficient of a material when a measurement of the birefringence is made under uniaxial stress conditions.<sup>23,29</sup>

The bulk counterpart of the polymer films investigated in this study are known to be isotropic. In the thin film form, on the other hand, these polymers were found to be slightly anisotropic. The observed anisotropy is mainly due to effects such as in-plane orientation of the polymeric chains during the spinning process and solvent evaporation and may depend on the specific type of polymer itself.<sup>12,30</sup> Due to the spinning process, the expected anisotropy of the layers is of uniaxial type and one of the principal axis of symmetry is parallel to the incident polarization. The measured birefringence in this case is  $< 0.002$  and in this regime the isotropic assumption for mode equations remains as a reasonable approximation.<sup>31-33</sup> The variation of birefringence under stress, on the other hand (i.e., stress induced birefringence), is governed by Neumann-Maxwell stress equations. Therefore, in our case, we assume that the polymer films are isotropic in the in-plane direction and with the described coordinate system convention the following stress equations can be written as<sup>27,28</sup>

$$n_x = n_{0x} + C_1 \sigma_x + C_2 (\sigma_y + \sigma_z), \tag{2}$$

$$n_y = n_{0y} + C_1 \sigma_y + C_2 (\sigma_x + \sigma_z). \tag{3}$$

In order to be able to obtain the elasto-optic coefficients, the variation of the refractive index and film stress should be expressed in terms of identical quantities. For this reason 3D FEM analysis was used so as to obtain the principal stresses for each polymer system. The material properties were defined by their elastic moduli  $E$ , Poisson's ratio  $\nu$ , and initial thickness  $y_0$ . The out-of-plane elastic moduli and film thicknesses were determined experimentally as described above, while the values of Poisson's ratio  $\nu$  for each film were used as reported in the literature. In particular, the following values were employed: 0.33, 0.34, and 0.35 for PS, BCB, and PMMA, respectively.<sup>9,18,34</sup> The change of the film thickness  $\Delta y$  with applied stress was simulated and equations of the following form were obtained for each direction and polymer film:

$$\sigma_i = \text{const} \times \Delta y = K_i \Delta y. \tag{4}$$

Moreover, the refractive index variations with the applied pressure can also be expressed in terms of  $\Delta y$ . As seen from Figs. 2 and 3 the following linear relations can be written as

$$n_x = n_{0x} + m_x \Delta y, \tag{5}$$

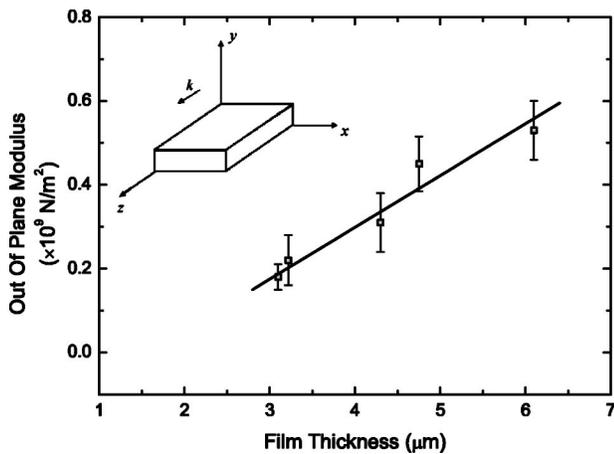


FIG. 8. Variation of the out-of-plane elastic modulus with film thickness for PMMA films.

TABLE I. Results of FEM analysis and elasto-optic coefficient calculations. The tabulated data include the initial thickness of the guiding polymer films  $y_0$ , maximum film thickness change with the applied pressure  $\Delta y_{\max}$ , calculated elastic moduli of the layers  $E$ , constants  $K_1, K_2$ , and  $K_3$  as determined by FEM simulations, the slope values  $m_x$  and  $m_y$ , and the calculated elasto-optic constants  $C_1, C_2$ , and  $C$ .

Polymer	$y_0(\mu\text{m})$	$\Delta y_{\max}(\mu\text{m})$	$E(10^9 \text{ N/m}^2)$	$(10^{14} \text{ N/m}^2)$			$(10^4 \text{ 1/m})$		$(10^{-12} \text{ m}^2/\text{N})$		
				$K_1$	$K_2$	$K_3$	$m_x$	$m_y$	$C_1$	$C_2$	$C$
PS	2.37	0.16	0.3	-0.9	-1.9	-0.9	1.241	1.428	-48	-29	-19
BCB	2.38	0.07	0.3	-1.0	-1.9	-1.0	1.969	2.698	-107	-30	-77
PMMA	2.30	0.06	0.9	-3.4	-6.3	-3.4	1.306	1.306	-10	-10	0

$$n_y = n_{0y} + m_y \Delta y, \quad (6)$$

where  $m_x$  and  $m_y$  are the slopes obtained from the linear fits to the refractive index measurements. Substituting Eqs. (4)–(6) into Eqs. (2) and (3) and taking their difference the equations for the elasto-optic coefficients  $C_1$  and  $C_2$  take on the form

$$C_1 = \frac{(m_x + m_y)(K_2 - K_1) - (m_x - m_y)(K_1 + K_2 + 2K_3)}{(K_1 + K_2)(K_2 - K_1) - (K_1 - K_2)(K_1 + K_2 + 2K_3)}, \quad (7)$$

$$C_2 = \frac{(m_x - m_y) - C_1(K_1 - K_2)}{K_2 - K_1}. \quad (8)$$

Here, the elasto-optic coefficients  $C_1$  and  $C_2$  are expressed in terms of quantities determined from refractive index measurements (i.e.,  $m_x$  and  $m_y$ ) and the parameters found from FEM calculations (i.e.,  $K_1, K_2$ , and  $K_3$ ).

The results of FEM analysis and elasto-optic coefficient calculations are given in Table I. The table contains the initial thickness of the guiding polymer films  $y_0$ , maximum film thickness change with the applied pressure  $y_{\max}$ , calculated elastic moduli of the layers  $E$ , constants  $K_1, K_2$ , and  $K_3$  determined by FEM simulations as explained above, the slope values  $m_x$  and  $m_y$  given in Eqs. (5) and (6), and the calculated elasto-optic constants  $C_1, C_2$ , and  $C$ .

For PS thin films, the elasto-optical coefficients  $C_1$  and  $C_2$  were calculated to be  $-48 \pm 3 Br$  and  $-29 \pm 3 Br$  ( $1 Br = 10^{-12} \text{ m}^2/\text{N}$ ), respectively. There are strain elasto-optical coefficients ( $p_{11}, p_{12}$ ) reported for PS films.<sup>35</sup> These coefficients can be translated into the stress-optical coefficients by using the following relations:<sup>36</sup>

$$C_1 = -\frac{n^3}{2E}(p_{11} - 2\nu p_{12}), \quad (9)$$

$$C_2 = -\frac{n^3}{2E}(p_{12} - \nu p_{11} - \nu p_{12}). \quad (10)$$

Using the reported  $p_{11}, p_{12}, E$ , and  $\nu$  values for bulk PS films in the above equations, we obtain  $C_1 = -61.4 Br$  and  $C_2 = -69.7 Br$ . First, we see that the values for the bulk material and those we obtained for thin PS films are of the same order. The difference of  $(C_1 - C_2)$  can be used to compare our results with the widely reported stress-optic coefficient  $C$  for bulk polymers where measurement of birefringence is made under uniaxial stress conditions.<sup>23,29</sup> For bulk PS,  $C$  is positive with a value of  $8.3 Br$ , whereas the calcu-

lated  $C$  for PS thin films is negative with a value of  $-19 Br$ . The bulk material value is in agreement with the value of  $8-10 Br$  in other reports in the literature.<sup>23,24,27</sup> The process of determination of the stress-optic constant  $C$  is the key factor in the apparent sign mismatch of the mentioned parameter. Namely, the reported stress-optical coefficients  $C$  for bulk PS materials are calculated from the birefringence and stress measurements when the material is under tensile stress. The extent of motion of the polymer chains is limited upon such deformation. It is known that the birefringence of PS is mainly determined by the orientation of the phenyl groups. As discussed by Rudd and Gurnee, upon the application of tensile stress, the phenyl groups tend to align in the direction of the applied stress, i.e., in the in-plane direction.<sup>31</sup> This results in positive birefringence and positive stress-optic coefficient. However, in our case the stress is applied in the out-of-plane direction of the polymer film. We suggest that this should lead to at least partial alignment of the polymer chains in the out-of-plane direction and consequently to perpendicular alignment of the plane of the phenyl groups.<sup>37</sup> This, accordingly, should result in negative birefringence and negative stress-optic coefficient, as was observed for the PS films in this study.

The elasto-optic coefficients for the BCB layers were found to be as  $C_1 = -107 \pm 8 Br, C_2 = -30 \pm 8 Br$ , and  $C = -77 Br$ . As for the PMMA thin film layers, the obtained stress-optic coefficients are  $C_1 = -10 \pm 3 Br, C_2 = -10 \pm 3 Br$ , and  $C = 0 Br$ . The results for PMMA are reasonable since for optically isotropic films, equal elasto-optical coefficients and thus zero  $C$  are to be expected.

While the reported method for measuring the mechanical and optical properties simultaneously by making use of prism coupling technique is novel and interesting, we note that the measured values of the material properties such as out-of-plane elastic modulus and elasto-optical constants should be considered as preliminary. The technique still involves difficulties and assumptions that can possibly be improved. Namely, we have assumed that the applied stress is uniform across the coupling area and the Poisson's ratio  $\nu$  was assumed to be identical with the bulk value.

#### IV. SUMMARY AND CONCLUSIONS

An experimental setup based on the prism coupling method was built and utilized to measure the refractive indices, optical anisotropy, and out-of-plane elastic moduli of

PS, PMMA, and BCB thin polymer films. Moreover, the variation of the optical properties of these films with the applied pressure was characterized.

Although prism coupling technique is widely used in order to measure the refractive index and thickness of the polymer films, the effects of the stress that is applied on the film in this method is not taken into account. Inclusion of these effects allows one to measure the refractive index of the polymer films reliably and with higher accuracy. It was found that the refractive index values of the investigated three polymer films increased with the applied pressure for both TE and TM polarizations, in accordance with the stress-optic effect. A negative birefringence of  $-0.0007$  was observed for PS thin films, the birefringence for the PMMA layers was found to be zero, and that of the BCB films was found to be  $0.0022$  in the unstressed condition.

The out-of-plane elastic moduli of the three thin polymer films were found by making use of the prism coupling technique measurements of  $0.3 \pm 0.1 \times 10^9$  N/m<sup>2</sup>,  $0.3 \pm 0.1 \times 10^9$  N/m<sup>2</sup>, and  $0.9 \pm 0.1 \times 10^9$  N/m<sup>2</sup> for PS, BCB, and PMMA, respectively. Furthermore, the out-of-plane elastic modulus was found to increase with film thickness as verified for PMMA layers. For PMMA films dissolved using chlorobenzol the corresponding value increased from  $0.18 \pm 0.03 \times 10^9$  N/m<sup>2</sup> up to  $0.5 \pm 0.1 \times 10^9$  N/m<sup>2</sup> with increase in thickness from  $3.10 \mu\text{m}$  to  $6.10 \mu\text{m}$ .

Results of 3D FEM analysis together with the refractive index measurements were applied to the Neumann-Maxwell stress equations in order to obtain the elasto-optic coefficients for the PS, PMMA, and BCB thin films. In contrast with the measurements of elasto-optic coefficients made under tensile stress conditions for bulk polymers, it was found that the applied stress in the out-of-plane direction of the thin films investigated leads to negative elasto-optic coefficients, as observed for all of the three thin polymer films.

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