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Nematic Liquid Crystal Optical Dispersion in the Visible-Near Infrared Range

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A commercial variable angle spectroscopic ellipsometer from J. A. Woollam Company is modified in order to make possible measurements in guiding structures. The half leaky guided mode technique is realised for liquid crystal optical dispersion measurements. Features and problems related to experimental data interpretation and error sources in the refractive index measurements are carefully analyzed, achieving a final accuracy of ± 0.0002 . We present the measured optical dispersion curves in the wavelength range from 0.5 to 1.7 μm for two widely studied liquid crystals, namely E7 and 5CB.

Keywords: ellipsometry; half leaky guided mode; liquid crystal; optical dispersion model; refractive index

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INTRODUCTION

The growing interest in precise measurements of the liquid crystals (LC) refractive indices in the visible and near IR range is stimulated both by the need of experimental verification of the LC dispersion models [1,2] (see also references therein), and by numerous applications of LC in flat panel displays [3] and integrated optics [4,5]. Most of the commonly used methods for the optical characterization of LCs present some disadvantages. Commercial Abbe refractometer [6] can be used only in the visible range. Techniques based on laser sources, like for instance half-leaky guided mode (HLGM) [7] and wedge-cell [8], operate at a small number of wavelengths, furthermore they may require source replacing, with a consequent increased uncertainty in the investigated sample area. Spectroscopic Ellipsometry (SE) suffers for a strong dependence of its results on the data inversion procedure, often a quite cumbersome mixing of different chosen physical models and initial guess for the fitting parameters. In a recent paper [9], we presented the results of the refractive index and birefringence dispersion measurements for 5CB obtained by the reflection ellipsometry technique. Although our results have proved to be quite accurate, in our conclusion we recommended to complement SE with different techniques, even less accurate, in order to get rid of systematic errors that may occur frequently in SE measurements on liquid crystal samples.¹ HLGM has proved to be a very reliable and accurate technique for single wavelength refractive index measurements of LCs [7,9], thus is one of the most suitable candidate for complementing SE in the measurement of LC dispersion curves, permitting to remove the ambiguity in the physical model choice and initial guess for the SE data analysis. We have actually done this in Ref. [9] using a multi-laser source set-up and now we propose to overcome the single lines limitation of HLGM integrating this technique in the SE hardware. In this way, using as a light source a high pressure Xe lamp we can perform wide range continuous spectroscopic measurements. Moreover, being the two techniques (HLGM and SE) integrated in the same hardware, all the measurements are performed exactly on the same sample area and exploiting the same 3/29 mechanical set-up, automatically controlled with a very high accuracy and repeatability. The results on LC refractive index dispersion measurements presented in this paper were obtained by means of this integrated set-up

¹The main reason for this is the need of confinement of the LC sample in a glass cell, so that the first layer of the multi-layer sample structure is always a glass window, irrelevant for the searched result but determining the absolute major fraction of the detected light signal.

realized by us on a Variable Angle Spectroscopic Ellipsometer (VASE[®], J. A. Woollam Company).

HLGM METHOD IMPLEMENTED ON A VARIABLE ANGLE SPECTROSCOPIC ELLIPSOMETER

The principle of LC optical characterization by HLGM method is as follows. LC is placed in a standard cell formed by two glasses. The upper glass has a refractive index higher than the highest one of the LC, and the bottom glass lower than the lowest one. The light beam impinges on the cell through a prism made of the same material as the upper glass. A matching fluid ensures optical contact between them. The reflection coefficient is measured in the angular range where the planar waveguide modes of the LC layer can be excited. The exciting condition of the transversal mode of index m is described by the expressions:

$$\sin \gamma_m = n_p \sin(\alpha - \theta_m) \quad (1)$$

$$\theta_m = \arcsin \frac{\beta_m}{kn_p} \quad (2)$$

where γ is the incidence angle of the light on the prism, n_p – the refractive index of the prism, α – the angle between the base and the input face of the prism, k – the beam wave number in vacuum, θ – the internal incidence angle, calculated at the upper LC layer interface, β – the propagation constant of the mode (it can be easily obtained from the planar waveguide theory).

As the modes are guided only by the substrate surface and are leaky on the border with the upper glass, the method has got its name. With a suitable combination of the polarization states of the input and the detected light, a whole set of HLGM data is obtained. The optical parameters and the thickness of the LC layer can be obtained by a data inversion and fitting procedure based on generated data using the multilayer Fresnel theory [7].

In order to modify the commercial ellipsometer VASE[®] from J. A. Woollam Company [10] for adapting it to the HLGM method, we have designed a sample holder that can accommodate an SF6 prism on the upper side of the sample and an additional converging lens between the sample and the detector arm, as it is shown in Figure 1.

The lens partially compensates for the beam displacement from the detector position, caused by the refraction on the prism. The small residual beam displacement affects systematically the reflection

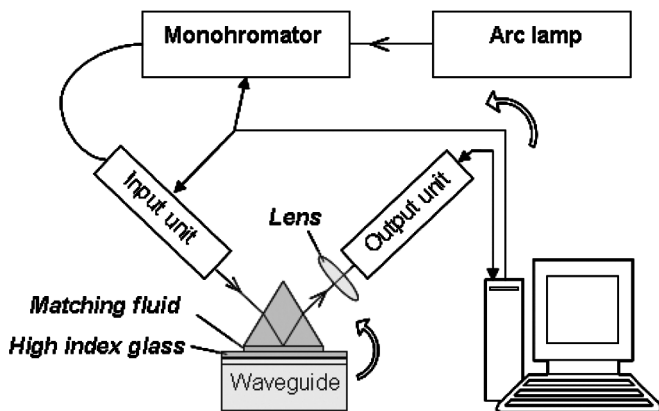


FIGURE 1 Modified ellipsometer.

coefficient curve but having negligible effects on the angular location of the excited modes.

The integration of the HLGGM technique with ellipsometry on the same device gives a clear advantage of a simultaneous and complementary use of both techniques to overcome most of the problems encountered in applying ellipsometry to LC. The fitting procedure for HLGGM data is easy and robust, so it gives reliable results for the refractive indices measurements. Furthermore, these results can be also used as a good initial guess for the fitting procedure in spectroscopic ellipsometry.²

The LC cells were made according to the demands of the HLGGM method. The upper and bottom glass are SF6 and fused silica, respectively. The inner side of the walls was coated by a 30 nm thick SiO_x layer to achieve planar alignment of LC [11]. The cell gap is fixed to about 9.5 μm by mylar spacers. The cells are filled with nematics E7 (sample N1) and 5CB (sample N2) by capillarity and are sealed by Norland optical adhesive. A matching fluid by Cargille ensured the optical continuity among cell and prism.

MEASUREMENTS AND DISCUSSION

HLGGM measurements have been performed at 8 wavelengths: $\lambda = 0.532, 0.6328, 0.7, 0.8, 1.064, 1.3, 1.55, 1.7 \mu\text{m}$. Some of the chosen wavelengths coincide with laser lines to have opportunity to compare

²Our custom modification of the VASE[®] ellipsometer allows also the implementation of the M-line technique for the optical characterisation of solid guiding structures [9].

these results with the ones obtained by proper laser sources. The monochromator slit width was set to $100\text{ }\mu\text{m}$ in order to increase the spectral resolution.

The LC cell was oriented in such a way to have the nematic director along the vertical direction. As the plane of incidence of VASE[®] ellipsometer lies horizontally, TE and TM modes of the LC layer were excited by *s*- and *p*-polarized beam, respectively. In the first case, the beam interacts with the extraordinary index n_e of the nematic, and in the second case with the ordinary one n_o .

The E7 reflectivity curves, measured and simulated by the software, are shown in the Figure 2a and b for wavelengths 0.532 and $1.55\text{ }\mu\text{m}$, respectively. The internal incidence angle θ is reported in the horizontal axes of the graphs. The angle θ is obtained by means of Eq. (1), from measured values of the incidence angle γ , the prism index n_p and prism angle α . We used the Sellmeier coefficients for

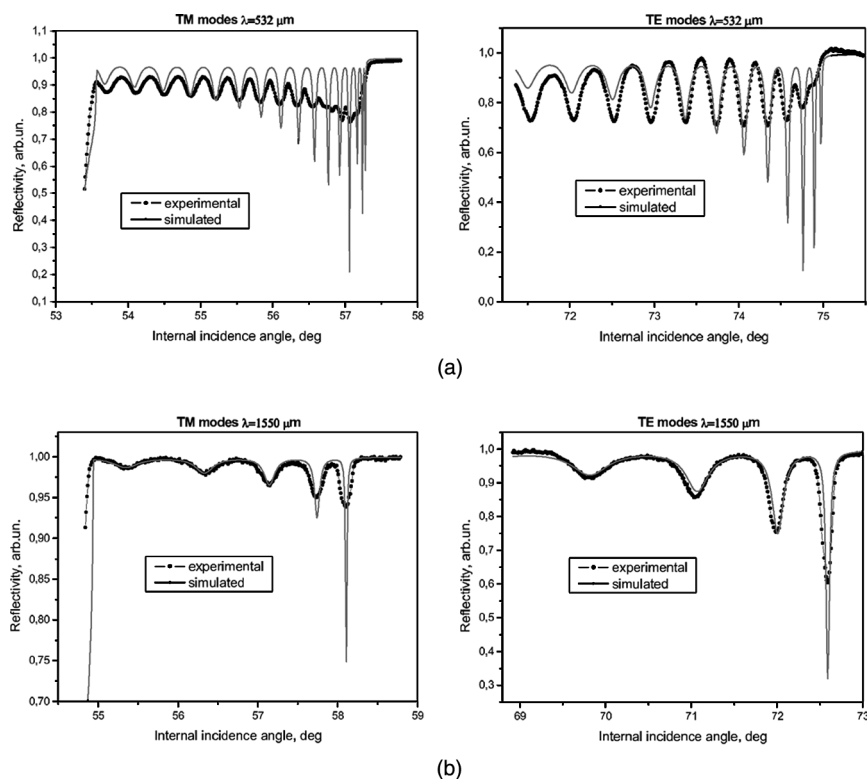


FIGURE 2 Reflectivity curves of E7 for $\lambda = 0.532$ (a) and $1.55\text{ }\mu\text{m}$ (b).

the SF6 optical dispersion from [12] and the measured value of $60.06 \pm 0.001^\circ$ for the prism angle.

There exists an angular window, going from the pseudo-critical angle between the high index prism and the effective index of the liquid crystal up to the critical angle between the high index prism and the low index substrate, over which sharp half-leaky resonant guided modes may propagate. The modes are excited within the interval between these cut-off angles and are characterized by local minima of the reflectivity. As the propagation constant of the mode decreases when the mode transversal index rises, the lowest mode is located near the first cut-off according to Eq. (2).

The zero of the angular scale was defined by the second cut-off point calculated according to the equation:

$$\theta_{\text{cut-off}} = \arcsin \frac{n_{\text{Silica}}}{n_p} \quad (3)$$

In this calculation the value for the SiO_2 refractive index, $n_{\text{Silica}} = 1.46071$ @ $\lambda = 532 \text{ nm}$, was used [13]. The precision in LC refractive index measurements is determined by the error in the second cut-off point measurement, $\Delta\Theta = \pm 0.01^\circ$, and can be evaluated by the formula:

$$\Delta n = n_p \cos \theta_{\text{cut-off}} \cdot \Delta\theta \quad (4)$$

In the wavelength range of interest, we get corresponding ranges for $\theta_{\text{cut-off}}$ of $53^\circ \div 55^\circ$ and for n_p of $1.81 \div 1.76$, thus, with the above mentioned angular measurement experimental error, our precision in the refractive index values is within $\Delta n = \pm 0.0002$. However, a different error source may significantly affect the accuracy of the results, namely the temperature control of the sample. In fact, the thermal coefficients of the refractive indices is very large in LC materials, especially approaching the clearing point. In our setup the temperature was held fixed within $\pm 1 \text{ K}$, and this was the main error source (with the exception of E7 ordinary index) in the final results. According to [6] for $T_c - T = 31 \pm 1 \text{ K}$, where T_c is the clearing point temperature, standard errors of the E7 ordinary and extraordinary index are $\pm 5 \cdot 10^{-6}$ and $\pm 1.5 \cdot 10^{-3}$, respectively, and according to [2] for $T_c - T = 8.5 \pm 1 \text{ K}$ standard errors for 5CB are $\pm 1.5 \cdot 10^{-3}$ and $\pm 5 \cdot 10^{-3}$, respectively.

At $\lambda = 0.532$, and shorter wavelengths, the lower modes of experimental curves are partially overlapped due to the spectral bandwidth of the monochromator setup. In the near IR range the angular intervals between modes increase and the ellipsometer spectral resolution become high enough to completely resolve all the modes. However, this

proves not to be a critical issue. In fact, one of the main advantages of the HLGGM technique is that, even using a light source with a poor spectral resolution if compared with laser sources, it allows to fit quite well the experimental data because of the presence of a large number of excited modes (at least for thick cells). Furthermore only the peaks location, but not the whole curve shape, is relevant for index and thickness evaluation. This renders almost unessential the demand for a high spectral resolution, hence allows using the ellipsometer for HLGGM method with errors of the same order as obtained with laser sources.

Another advantage comes out from the possibility to fit the refractive index and the LC layer thickness independently, in contrast with other optical techniques which are sensitive only to the product of these layer parameters. In fact, in homogeneous cells, the refractive index determines the location of the first cut-off angle, while the LC film thickness closely correlates with the number of excited modes.

The obtained values of n_o and n_e for E7 and 5CB are shown in Figure 3.

Due to the absence of any resonances in the examined spectral range, we can describe the ordinary and extraordinary indices of the nematics by using the 3-parameter Cauchy formula:

$$n_{o,e} = A_{o,e} + B_{o,e}\lambda^{-2} + C_{o,e}\lambda^{-4} \quad (5)$$

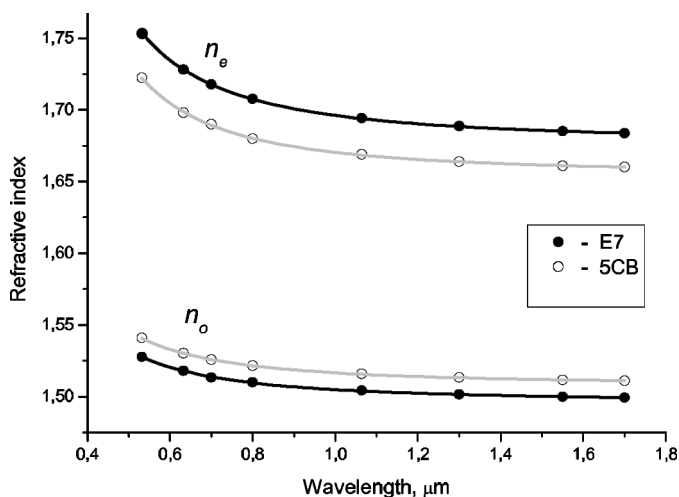


FIGURE 3 Dispersion curves of E7 and 5CB.

TABLE 1 Cauchy Parameters of E7 and 5CB Liquid Crystals

LC	A_o	$B_o, \mu\text{m}^2$	$C_o, \mu\text{m}^4$	A_e	$B_e, \mu\text{m}^2$	$C_e, \mu\text{m}^4$
E7	1.49669	0.00785	0.00026	1.67798	0.01696	0.00127
5CB	1.50849	0.00774	0.00040	1.65535	0.01355	0.00153

The Cauchy parameters obtained by the fit are presented in the Table 1 and the calculated dispersion curves are shown in the Figure 3 as solid lines.

CONCLUSION

We have modified our commercial ellipsometer, VASE[®] from J. A. Woollam Company, in order to make possible measurements in guiding structures. Thus, we supplied the HLGm method with the wide spectrum light from a Xe lamp, obtaining a full spectroscopic measurement opportunity. The measuring technique has demonstrated a very high accuracy for the refractive indices determination that can be achieved under good thermal control of the samples.

Results of the anisotropic refractive index measurements of two commonly used LC materials (E7 and 5CB by Merck) are presented in the visible and, that is more important due to the lack of experimental data, in NIR ranges.

Combined and simultaneous measurements performed with HLGm and Spectroscopic Ellipsometry on the same sample and with the same modifies apparatus are now in progress and will be presented shortly.

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