Birefringence measurements of liquid crystals and an application: An achromatic waveplate

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Birefringence measurements of liquid crystals and an application: An achromatic waveplate

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We demonstrate a quarter wave phase retarder consisting of two layers of different liquid crystals (LCs). The phase retardation does not differ by more than $\pm 5\%$ from the target value $\pi/2$ within a spectral interval of 200 nm in the visible range. These values are comparable to the data for commercial two layer crystalline retarders which are much more expensive, especially if large apertures are required. In order to design the LC retarder we measured the birefringence of the LC within the entire visible spectrum using a published technique which does not consider interference effects due to reflections within indium–tin oxide coated LC cells. We show that these effects do not affect the results adversely provided that the birefringence data are taken over a broad spectral range.

1. Introduction

Liquid crystals (LCs) can be a low cost alternative to birefringent crystals commonly used in polarization optics, especially if large apertures are needed. One possible application is an achromatic phase retarder (e.g. $\lambda/4$-plate) to manipulate the polarization of a white light continuum, e.g. for astronomical purposes. Such devices can be assembled from at least two layers of different birefringent materials. This was shown by Clarke [1] using crystalline plates. Their main drawback (besides their cost) is that only a few suitable crystalline materials are available. Their spectral properties require that the individual layers perform as higher order waveplates in order to achieve an achromatic behaviour. LC materials may solve this problem because many different chemical classes with strongly different spectral properties can be synthesized. Thus the chance of finding better combinations is much higher compared with crystalline materials.

In order to apply LC materials precise knowledge of their spectral properties is needed. For instance, the construction of achromatic waveplates requires the accurate knowledge of the spectral behaviour of the birefringence of the LC over the entire visible spectrum.

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Therefore measuring techniques to determine refractive indices or the birefringence are very important in order to design optical devices. Refractive indices can be determined accurately using a refractometer, but its wavelength range is restricted. On the other hand direct measurement of the spectral behaviour of the birefringence is possible by the method proposed by Wu et al. [2]. One possible drawback of this method is the neglect of reflections within the LC cell which may lead to interference fringes and reduce the accuracy, especially if the LCs investigated are contained in commercial LC cells with a typical thickness in the µm-range.

The first part of this paper demonstrates that this method is still applicable to thin LC layers provided that the data are taken, not only at a few discrete wavelengths, but over a broad spectral range. In the second part, we present the combination of different LC layers in order to design a waveplate with an achromatic behaviour comparable to that of expensive crystalline waveplates.

2. Determination of the birefringence of the LC

Figure 1 shows the main features of the experimental set-up. Commercially available LC cells with indium–tin
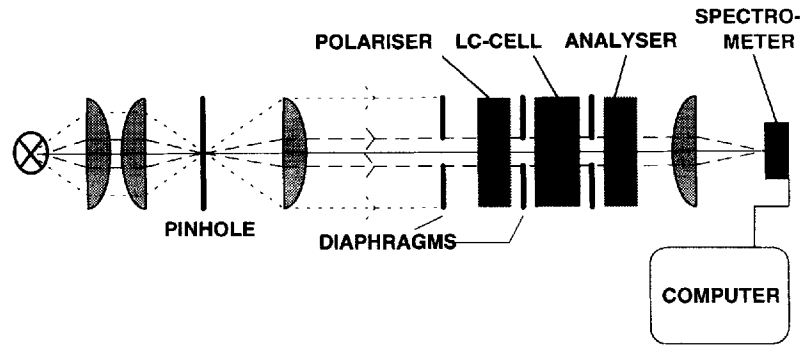
Figure 1. Schematic diagram of the experimental set-up.

oxide (ITO) and polyimide coatings were used. The LC cells were filled under vacuum conditions with the LC in the isotropic phase after the cell thickness d had been measured interferometrically [3]. The cells delivered by Displaytech (Boulder, Colorado) and EKMO (Minsk, Belarus) were mounted in a temperature stabilized environment provided by a Peltier thermoelement and a temperature sensor. A controller circuit allowed temperature stabilization to within ± 0.02 K in the temperature range from 10 to 80°C. The LC cell could be rotated, the rotation axis being the light beam axis. A battery powered tungsten lamp was used as a stable broad band light source. A system of lenses and apertures produced a uniform white light beam with a divergence less than 1° that passed through a polarizer (Glan prism with extinction ratio 10^{-6}), the cell and an analyser (equivalent to the polarizer). The light was then focused on the fibre entrance of a fast miniature spectrometer manufactured by Zeiss (Oberkochen, Germany). The spectrometer, controlled by a personal computer, contained a grating and a photodiode array cemented on a UV-transparent body. One pixel covered a spectral range of 3·3 nm. The resolution due to the entrance slit width was 10 nm. For a spectrum from $\lambda = 300$ to 900 nm, several integrations over 10 to 100 ms measuring times with 12 bit a/d conversion were averaged.

The LC cell acts as an etalon with different optical thicknesses for a polarization parallel (suffix p) and perpendicular (suffix s) to the fast axis of the LC. We assume effective wavelength-dependent reflection coefficients R_p and R_s for reflections at the entire LC/polyimide/ITO/glass interface system.

With $\tau_{p,s}$ accounting for the wavelength-dependent intensity transmission, including the reflection losses at the outer glass surfaces and the absorption losses, the electrical field amplitude $E_{p,s}$ of the transmitted light wave can be calculated following the familiar etalon formula [4], which has to be modified to consider the phase shifts due to the layers coated onto the glass:

$$E_{p,s} = \tau_{p,s}^{1/2} E_0\, e^{-i\delta_{p,s}}\, e^{-i\delta_{glass}}\, e^{-i\delta_{ITO}}\, e^{-i\delta_{polyimide}}$$

$$\times\; \frac{1 - R_{p,s}}{1 - R_{p,s}e^{-2i\varphi_{p,s}}} \tag{1}$$

In equation (1) E_0 is the incoming electrical field, and $\delta_{p,s}$ are the one-pass phase retardations due to the LC for the extraordinary and ordinary beam, respectively. δ_{glass}, δ_{ITO} and $\delta_{polyimide}$ are the one-pass phase retardations due to one pair of glass substrates or the ITO or polyimide layers, respectively, which do not depend on the polarization of the incoming beam. $\varphi_{p,s}$ is the phase shift of the light between two subsequent reflections at the LC/polyimide/ITO/glass interface system. In this term, the shifts due to the reflections at the glass/ITO, the ITO/polyimide, and the polyimide/LC interface have to be considered. These phase shifts are different for the extraordinary and ordinary beam, because the reflection coefficient of the LC/polyimide interface depends on the polarization of the light beam.

If the angle between the orientation of the polarizer and the fast axis of the LC is β and the angle between the orientation of the polarizer and that of the analyser is α, the field transmitted by the analyser is given by:

$$E = \cos(\beta - \alpha)\cos(\beta)E_p + \sin(\beta - \alpha)\sin(\beta)E_s \tag{2}$$

By squaring equation (1) we find the transmitted intensities $I_{p,s}$, where I_p is measured with $\alpha = \beta = 0°$; I_s results from measurements with $\alpha = 0°$, $\beta = 90°$:

$$I_{p,s} = \tau_{p,s} I_0\, \frac{1}{1 + \dfrac{4R_{p,s}}{(1 - R_{p,s})^2}\sin^2\varphi_{p,s}} \tag{3}$$

where I_0 is the incoming intensity which is measured without the LC cell.

With $\beta = 45°$ and introducing $\delta = \delta_p - \delta_s$, a straightforward calculation for polarizer positions $\alpha = 0°$ and $\alpha = 90°$, squaring equation (2), results in:

$$I(\alpha = 0°) = \frac{1}{4}(I_p + I_s) + \frac{I_p I_s}{(\tau_p \tau_s)^{1/2} I_0}$$

$$\times \left[\cos^2\frac{\delta}{2} - \frac{1}{2} + \frac{(R_p + R_s)}{(1 - R_p)(1 - R_s)} \sin\varphi_p \sin\varphi_s \right]$$

(4 a)

$$I(\alpha = 90°) = \frac{1}{4}(I_p + I_s) + \frac{I_p I_s}{(\tau_p \tau_s)^{1/2} I_0}$$

$$\times \left[\sin^2\frac{\delta}{2} - \frac{1}{2} - \frac{(R_p + R_s)}{(1 - R_p)(1 - R_s)} \sin\varphi_p \sin\varphi_s \right]$$

(4 b)

where δ is the phase retardation between extraordinary and ordinary beam resulting from the birefringence of the LC layer.

If the light losses at the substrates and the interferences within the LC cell are neglected, R_p and R_s equal zero, τ_p and τ_s are equal to 1, I_p and I_s equal I_0, and equation (4) reduces to the intensities given by Wu *et al.* [2]. Figure 2 shows an example (LC: sample A, see below, LC cell thickness 4 μm) of the measured transmission ratios I_p/I_0, I_s/I_0, $I(\alpha = 0°)/I_0$ and $I(\alpha = 90°)/I_0$. For the latter two ratios, it is obvious that short scale interferences from reflections within the LC cell are superimposed on the large scale interference figure that results from the birefringent LC layer.

From these measured values we derive the birefringence in the following way. First we use the maxima of the interference of I_p/I_0 and I_s/I_0 in equation (3) (where $\sin\varphi_{p,s} = 0$) to derive the transmission coefficients $\tau_{p,s}$. For wavelengths between the maxima, we interpolate the calculated $\tau_{p,s}$ values linearly. Using equation (3) again, we get the reflection coefficients $R_{p,s}$ when measured values of the amplitude of the interferences are inserted. Again a linear interpolation is used to get the

values for wavelengths between the maxima. Finally we calculate the spectral dependence of $\sin\varphi_{p,s}$ using equation (3). Inserting the measured $I(\alpha = 0°)$ and $I(\alpha = 90°)$ values into equations (4 a) and (4 b), we get the spectral dependences of $\cos^2(\delta/2)$ and $\sin^2(\delta/2)$. From these values, we obtain the phase retardation δ as a function of wavelength, where the problem of ambiguity is considered following [2].

With the known LC thickness d, finally the birefringence $\Delta n(\lambda)$ is determined:

$$\Delta n(\lambda) = \frac{\lambda}{2\pi d}\delta(\lambda)$$

(5)

3. Results

Figure 3 shows the spectral dependence of the birefringence of sample A derived from figure 2. Comparison with the birefringence values obtained without interference corrections shows that the influence of reflections at the interfaces is rather important. On the other hand, our results show that a simple smoothing of the data obtained by the method described in [2], without interference corrections, can be used. The resulting accuracy will be sufficient for most practical applications. We note that although it seems obvious that the smoothing procedure gives correct results, this fact is not in itself compelling without the verification presented here. In every case, it is necessary that the birefringence data are measured at many wavelengths. It is not sufficient to measure the birefringence at only a few discrete lines when thin LC layers are used.

Errors in our measurements due to inaccurate knowledge of the cell thickness of the order of 1% are not wavelength dependent and were corrected by calibrating the results to refractometer measurements at $\lambda = 589$ nm.

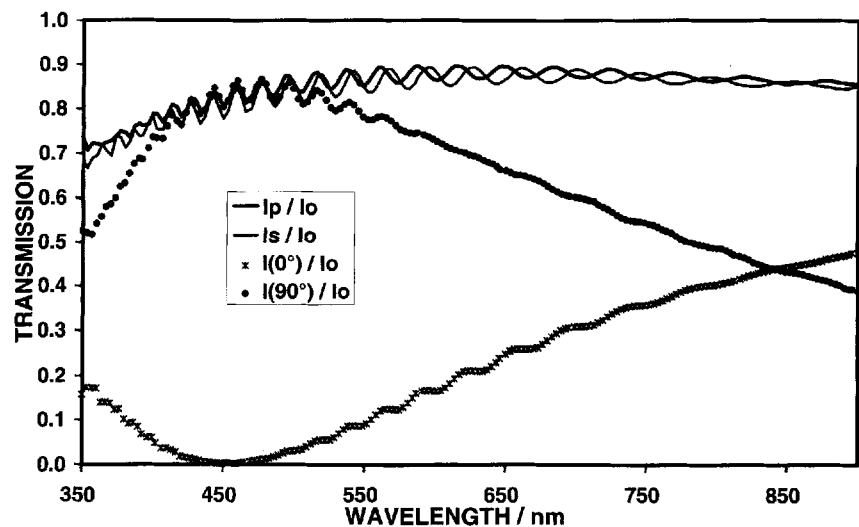Figure 2. The spectral dependence of the transmitted intensity ratios I_p/I_0, I_s/I_0, $I(\alpha = 0°)/I_0$, and $I(\alpha = 90°)/I_0$ of LC sample A at 20°C.
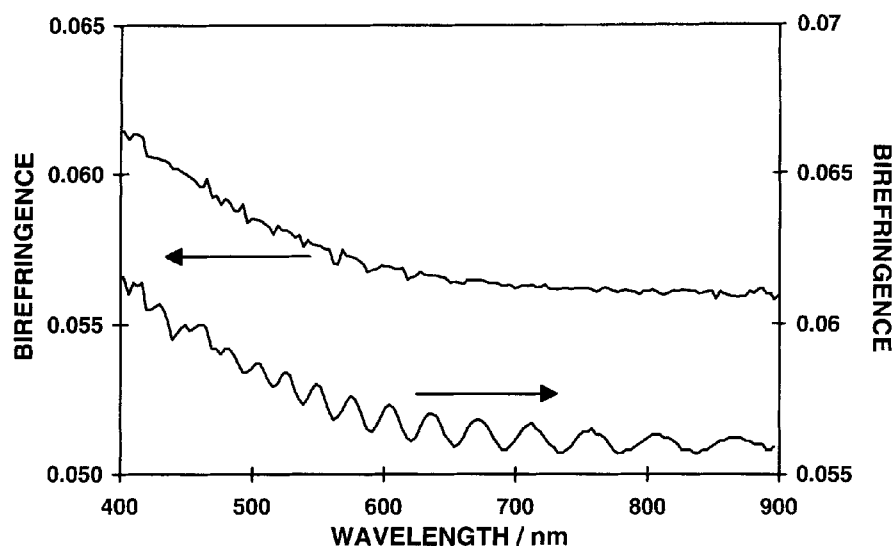
Figure 3. Birefringence derived from the measured intensity ratios given in figure 2. Upper curve: birefringence obtained with interference corrections (left scale). Lower curve: birefringence obtained without interference corrections (right scale).

From the scattering and the reproducibility of the values obtained, we estimate an accuracy of the birefringence in the order of 10^{-3} to 10^{-4}. This may be improved by using a longer data acquisition time or a spectrometer with a higher resolution.

The wavelength-dependent birefringence of several LC mixtures which are nematic at room temperature was measured [5]. For further calculations, the experimental data were fitted with the single band dispersion formula [6]:

$$\Delta n(\lambda, T) = G(T) \frac{\lambda^2 \lambda_0^2}{\lambda^2 - \lambda_0^2} \qquad (6)$$

Here λ_0 is an averaged resonance wavelength, and G incorporates both molecular parameters and the temperature dependence of the order parameter. This formula proved to fit the experimental data within the experimental uncertainty over the wavelength region 400 to 900 nm.

The results of our measurements are published elsewhere [5]. In this paper we present how the spectral data can be applied in order to design a waveplate with an achromatic behaviour over a given spectral range.

4. An application: the achromatic waveplate

An ideal achromatic waveplate requires a material for which the birefringence is proportional to the wavelength. A way to come close to this ideal is to combine materials with different dispersions of the birefringence. For a two-layer achromatic waveplate, one LC with a very low dispersion of the birefringence (usually equivalent to a very low birefringence) and another with a high dispersion of the birefringence, with their fast axes crossed, is a good choice. In order to design a quarter-waveplate, we chose two wavelengths λ_a and λ_b at which

the retardation should be $\pi/2$. The required thicknesses d_{LC1} and d_{LC2} of the two LC layers can be calculated by solving

$$\Delta n_{LC1}(\lambda_i)d_{LC1} - \Delta n_{LC2}(\lambda_i)d_{LC2} = \lambda_i/4 \qquad (i = a, b) \qquad (7)$$

The quality of the combination is determined by the maximum deviation of the retardation from the target value $\pi/2$ for wavelengths between λ_a and λ_b.

From the birefringence data obtained for about 30 LC materials we found that each combination of two LCs with different resonance wavelengths λ_0 results in an achromatic phase retardation within a limited spectral range. Achromatism is best for combinations where the LCs have nearly the same resonance wavelength. However, the required thicknesses calculated using equation (7) become very large in these cases and this limits the possible entrance angle to nearly 0. We therefore restricted our calculations to LC combinations where the order of each layer was below 2. The compositions of the LC samples which gave the best achromatism of retardation with $\lambda_a = 400$ nm and $\lambda_b = 550$ nm are given in figure 4. Each sample is composed of compounds differing only in the lengths of the terminal alkyl chains in order to achieve nematic properties at room temperature. The birefringence data for samples A and B are shown in figure 5, and the fit parameters for several temperatures are given in the table. The nematic phase was observed between 8°C and 36·5°C (sample A) and between 5·2°C and 44·5°C (sample B), allowing operation of the waveplate near room temperature. The viscosities of the LCs at 20°C are 70·77 mPa s (sample A) and 103·2 mPa s (sample B).

The cyanophenyl esters (sample B) [7] are widely used in commercial mixtures (e.g. as supplied by Merck Ltd
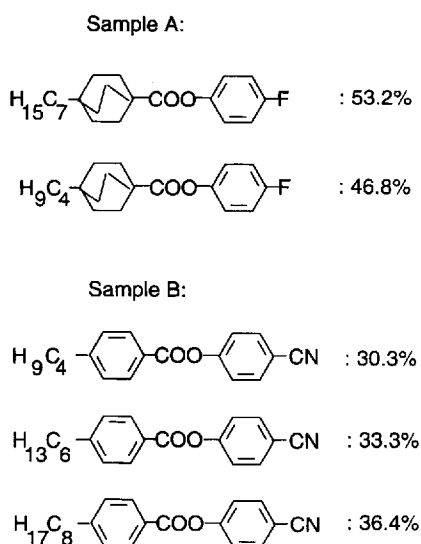
Sample A:

Sample B:

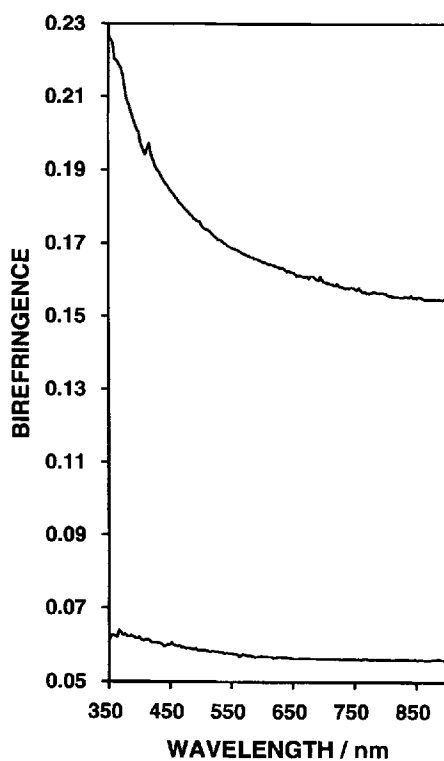Figure 4. Composition of LC samples A and B in weight %.

Figure 5. Birefringence of LC samples A (lower curve) and B at 20°C.

or Hoffman-La Roche). The fluorophenyl esters [8] of sample A can be supplied by the Military University of Technology, Warsaw, Poland. We note that the principal results of this work are not dependent on the use of these special LCs. We do not exclude the possibility that other combinations of materials not investigated by us will work even better. However, our results

Table. Fit parameters for equation (6) derived from birefringence measurements for LC samples A and B.

Sample	Temperature/°C	λ_0/nm	G/nm^{-2}
A	20	136·3	$2·92 \times 10^{-6}$
	30	136·5	$3·07 \times 10^{-6}$
B	20	207·6	$3·38 \times 10^{-6}$
	30	206·9	$3·10 \times 10^{-6}$
	40	206·9	$2·68 \times 10^{-6}$

demonstrate a performance that is already close to that obtained with expensive commercial crystalline retarders.

Using the birefringence values, the required thicknesses to achieve a quarterwave retardation at $\lambda_a = 400$ nm and $\lambda_b = 550$ nm are 9539 nm (sample A) and 2439 nm (sample B). The corresponding orders of the individual waveplates at $\lambda = 475$ nm are 1·2 and 0·9, respectively. A similar waveplate made from quartz and MgF_2 would have the orders 3·5 and 3·7 (thicknesses 179 μm and 151 μm), which means that the acceptance angle of the LC combination is considerably larger. The birefringence data for the crystalline materials were taken from [9].

To be able to adjust the retardation, we chose cells with thicknesses of 9500 ± 250 nm (sample A) and 3500 ± 100 nm (sample B) and used a 2 kHz sine wave voltage to tune the retardation of the latter. Computed and experimental results for two voltages applied to the thinner cell (RMS voltages 0·96 V and 0·93 V, corresponding to an effective thickness of sample B of 2339 nm and 2439 nm, respectively) are compared in figure 6.

No interference correction was applied to the experimental data, because in practice one is interested in the retardation of the entire device. It is obvious that a reduction of reflections within the LC cells, for example by antireflection coatings between substrate and ITO as well as between ITO and polyimide, is desirable for the construction of precise devices using thin LC layers.

The calculated retardation of the quartz-MgF_2 combination mentioned above is also shown and is similar to that of the LC combination. Crystalline achromatic retarders composed of a quartz-MgF_2 combination are available commercially, but they are very expensive, especially if large apertures are required. Typical prices are several thousand US$ for a two-layer retarder with a clear aperture of 25 mm. The LC retarder shown is an inexpensive alternative. As an additional advantage, the retardance of the LC device can be controlled by small voltages. Moreover, the orders of the LC layers are well below that of the commercial crystalline plates, thus allowing higher entrance angles. On the other hand, the crystalline plates can resist more powerful beams, but in
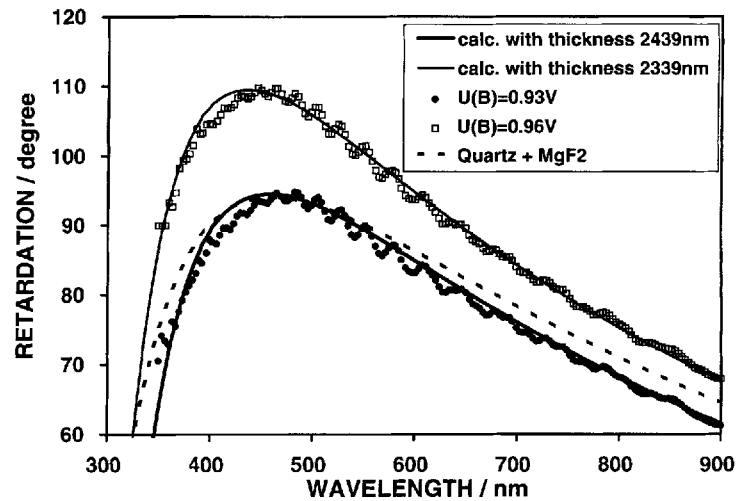
Figure 6. Calculated and measured phase retardation of the two-layer systems described in the text. An RMS voltage of 0·96 V (squares) and 0·93 V (circles) was applied to the thinner cell filled with sample B; for more details see text.

low-power applications, the LC device is an important alternative.

5. Conclusion

We have proved that the method described by Wu *et al.* [2] can be applied to birefringence determination even if reflections within thin LC cells are considered. As an application of birefringence measurements, a double layer waveplate with an achromatic behaviour comparable to that of expensive crystalline achromatic retarders was fabricated. Careful investigations of interference effects in the multilayer set-ups of LC cells are necessary for the construction of broadband devices.

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