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Testing the dynamic theory of nematics using fully-leaky guided modes and a convergent beam system

J.R. Sambles*, L.Z. Ruan

Thin Film Photonics Group, School of Physics, University of Exeter, Stocker Road, Exeter EX4 4QL, UK

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Abstract

The dynamic response of a twisted nematic cell is explored using a convergent beam system and the fully-leaky guided-mode technique. Application of an ac field results in a realignment of the director on the time scale of a few milliseconds. Fitting the guided-mode reflectivity and transmissivity data taken at different times after removal of the ac field allows a complete test of the dynamic theory of the nematic director behaviour. Direct confirmation of the director back-flow is established during the first few milliseconds after removal of the voltage. The optical results when compared with predictions of model theory establishes fully the validity of the Ericksen–Leslie theory. Exploration of the time dependent guided-mode data taken as the director relaxes to the zero volt state shows how the various viscosity coefficients influence the director relaxation yielding values for these coefficients.

Keywords: Liquid crystals; Viscosities; Dynamics; Optics

1. Introduction

The remarkable success of liquid crystal display technology rests on three basic foundation stones. Firstly there is a great body of practical knowledge, much established as long ago as the 1970s. Secondly, and at about the same time, high stability cyanobiphenyl liquid crystals were developed by George Gray and co-workers. Thirdly there already existed the Frank–Oseen–Zocher [1–3] static elastic theory and, importantly for device switching, the Ericksen–Leslie theory [4–8], a robust theoretical framework for predicting the dynamic director response of such nematic liquid crystals. This foundational work, concerning flow and dynamic effects in nematic liquid crystal, was developed in the 1960s, before the explosion of use of liquid crystal displays in the early 1970s. It has been extensively used, being regarded by most practitioners as the definitive theory. A useful reduced version of the general theory was developed by Berreman [9] and van Doorn [10]. Surprisingly there are rather few published accounts of the experimental confirmation of the dynamic theory. Possibly this is because it is regarded as being so robust that it is not really necessary to test it. Many simple experiments on response times, often one of the vital factors in device performance, have of course been undertaken. In addition there are a range of more elaborate experiments seeking to determine nematic viscosity coefficients. However one of the most striking predictions of the theory, that of 'back-flow' in a twisted nematic cell has not previously been directly recorded. This 'back-flow' that is the director, due to viscous forces, tilting away from the relaxation direction expected as it recovers from its distorted state, is expected on removal of a field from a twisted nematic cell. It is caused by the interaction of the rapidly recovering boundary regions with the central, near-homeotropic director, forcing the director to tilt 'beyond' 90° in the cell centre before recovering and coming back through 90° again as the velocity gradients diminish. The signature of this effect has been recorded as an optical 'bounce' in the light transmission [11,12]. From optical modelling of the expected director response this was regarded as strong confirmation of the predicted 'back-flow' of the liquid crystal in the centre of the cell. However since the optical response recorded is an integral of the optical response of the director through the whole cell it could not confirm details of the process. Indeed all experiments reported previously [13] based on simple transmission or reflection observations during switching suffer

* Corresponding author.
E-mail address: J.R.Sambles@exeter.au.uk (J.R. Sambles).

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from this same problem, they are an integrated response. It is impossible to establish the details of the dynamic director profile in the centre of the cell. Thus, no one has been able to establish unequivocally the predictions and therefore the validity of the dynamic theory. Nevertheless it has to be stated strongly that all the dynamic results obtained on nematic liquid crystals (with the possible exclusion of surface dynamics) are fully consistent with the Ericksen–Leslie theory.

In order to fully establish the validity of the theory one needs an experiment that allows the detailed probing of the director profile in a nematic liquid crystal cell as it is switched. One of the most powerful techniques for exploring the director profile in a liquid crystal cell involves using the liquid crystal layer as an optical waveguide. By examining the optical response of the layer for different in-plane momenta of incident light it is possible to probe the director profile in detail. This is because each different optical waveguide mode excited has a different optical field profile and so is sensitive to different parts of the liquid crystal through the cell. For example if the cell is a true optical guide (fully-guiding) the highest in-plane momentum mode will have only one optical field maximum at the centre of the liquid crystal layer and this mode will thus be sensitive primarily to the director at the cell centre. The next mode would have a zero at the cell centre and would be sensitive to the director at points one quarter way through the cell and three quarters through the cell. Over the past fifteen years the use of this waveguide probing technique for determining the director profile for a range of liquid crystal cell configurations has been established [14–19]. However only in the last few years, following the implementation of the fully-leaky waveguide procedure [19] have standard commercial cells been amenable to study using these powerful optical techniques. In this procedure the guided optical modes are in fact rather weak Fabry–Perot type modes created through the weak reflections at the liquid crystal/glass substrate boundary. They are labelled fully-leaky simply because the light may readily refract out of the liquid crystal layer into the surrounding low index glass plates.

Very recently this fully-leaky technique has been developed one stage further through the use of a convergent beam to cover the requisite incident angle range and a charge coupled device (CCD) array detector to collect either the reflectivity or transmissivity data from a static sample [20,21]. This replaces the standard collimated input beam, rotating sample and silicon photodiode detectors. With the standard waveguide mode excitation system the sample is slowly rotated and reflectivities and transmissivities recorded with varying angle of incidence (in-plane momenta). It thereby takes some time (minutes) to record the optical response of the cell over the range of incidence angles required to excite several different order guided modes. Furthermore, with prism-coupling (not glass hemispheres), as the sample rotates the place which the incident beam strikes the liquid crystal layer may move. It is apparent that this conventional technique is not particularly well-suited to the study of dynamic processes or to non-uniform, possibly pixelated, cells. The convergent beam technique, using either hemispherical or prism coupling resolves these difficulties. Because the incident convergent beam has a wide angle-range, the sample does not need to be rotated. This guarantees that data is recorded from a part of the sample corresponding to just the focused beam spot size. More significantly, as far as dynamic studies of the director are concerned the CCD camera means that ‘snapshots’ of the optical response function of the cell may be recorded on time scales of order milliseconds or less. This then allows the direct study of the whole liquid crystal profile through the cell during switching.

In this research a convergent beam system is used to investigate the dynamics of a twisted nematic cell. This simple configuration admits to the exploration of ‘back-flow’ of the director at the centre of the cell and also yields values for the viscosity coefficients of the liquid crystal. Primarily of course it is the ideal experiment for rigorous testing of the Ericksen–Leslie dynamic theory of nematics.

2. Experimental

In Fig. 1 is a schematic of the optical convergent beam system used in this study. A He–Ne laser provides a collimated beam of wavelength 632.8 nm. This is converted, by

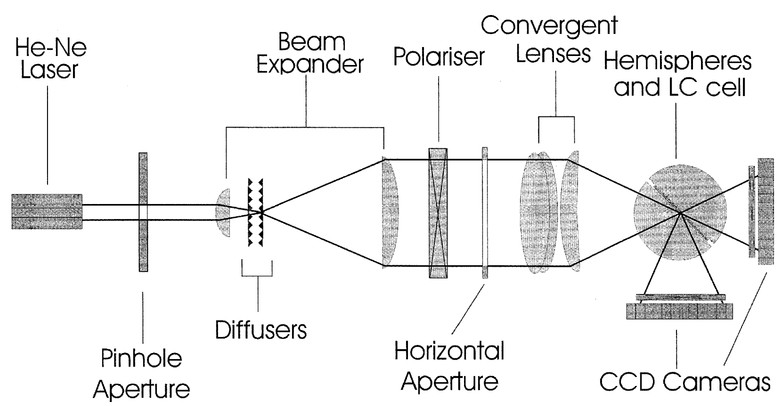

Fig. 1. A schematic of the experimental set-up for the convergent beam system.

the use of two diffusers (one rotating at high speed, one fixed) and beam expander into a less-coherent parallel beam of diameter about 5 cm [20]. This wide beam passes through a polarizer, a horizontal slit aperture and a pair of converging lenses being focused to a spot of diameter about 100 μm at the centre of the liquid crystal cell. Optical coupling, both in and out, is achieved by two hemispherical prisms which are optically coupled to the cell by matching fluid (in practice these are hemispherical surfaces with about 1 mm thickness removed from the flat faces to allow for the liquid crystal cell glass thickness). The diverging reflectivity and transmissivity signals pass through a detector polarizer (analyzer) before arriving at the CCD camera (manufacturer DALSA).

This configuration, with low index ($n \sim 1.52$) glass plates, matching fluid and coupling hemispheres comprises the fully-leaky geometry. Each of the low index glass plates ($n = 1.5170$), is coated with a thin (about 50 nm) layer of indium tin oxide (ITO), a transparent conductor. On top of these layers are surface-aligning layers of rubbed polyimide to provide low-tilt homogeneous alignment. These coated glass plates are then set parallel with 4.0 μm spacers between them. The rubbing directions of the upper and lower plates are set such that they are rather less than 90° apart, at about 87°, forming a twist cell. This cell is then heated and filled with ZLI-2293 (Merck) in the isotropic phase before being slowly cooled to rom temperature. Observations using a polarization microscope show that the director is uniformly aligned (twisted) with no visible defects.

This complete cell is then placed between the glass hemispheres with optical contact being established with matching fluid. The glass substrates of the liquid crystal cell, the hemispherical prisms and the matching fluid have the same refractive index. This complete assembly is then placed so that the centre of the liquid crystal sample is at the convergent beam focus. The rubbing direction at the incident surface is set at an angle of about 45° with the incident plane to give higher sensitivity, in the optical data, to the director profile in the cell. Further, theoretical modelling using a proposed model of the director profile in the cell shows that the angular region of the fully-leaky guided mode data which is most sensitive to the director alignment, is in the high in-plane wavevector area. Thus, to explore this angular region, the sample is set so that the angle between the central axis of the incident (convergent) beam and the cell normal is ∼70°. This angle setting, combined with the angle spread in the incident convergent beam provides detailed optical reflectivities and transmissivities associated with guided modes excited in the range from about 60 to 78° incident angle.

To establish the detail one requires in the director profile it is essential that the angular dependent data is as free from noise as possible. There are essentially two types of noise. The first appears as sharp, non-random, repeatable features in the angle dependent data which are present even with the liquid crystal in the isotropic phase. These features are due to interference in the various optical components in the system which is minimized by making the source spatially incoherent. This is achieved by the combination of a fixed diffuser and a very high speed rotating diffuser. An air-turbine dental drill (LARES APOLLO 557) with a rotation rate of up to 300,000 rpm is used for the rotating diffuser. This combination, used with a 70 mW He–Ne laser gives sufficient brightness at the sample to record signals with low noise due to the spatial coherence. However with very strong diffusers the signal may be so weakened that the second type of noise, due to low photon counts in each pixel of the CCD may arise. Then to obtain high quality dynamic data the exposure time of the CCD camera is set in accord with the required temporal resolution. This of course depends on the sensitivity and the line transfer rate of the CCD camera. In addition, since the rotating diffuser has inconsistent diffuseness, it is essential that the rotating diffuser completes an exact integer of revolutions in the exposure time. Using a

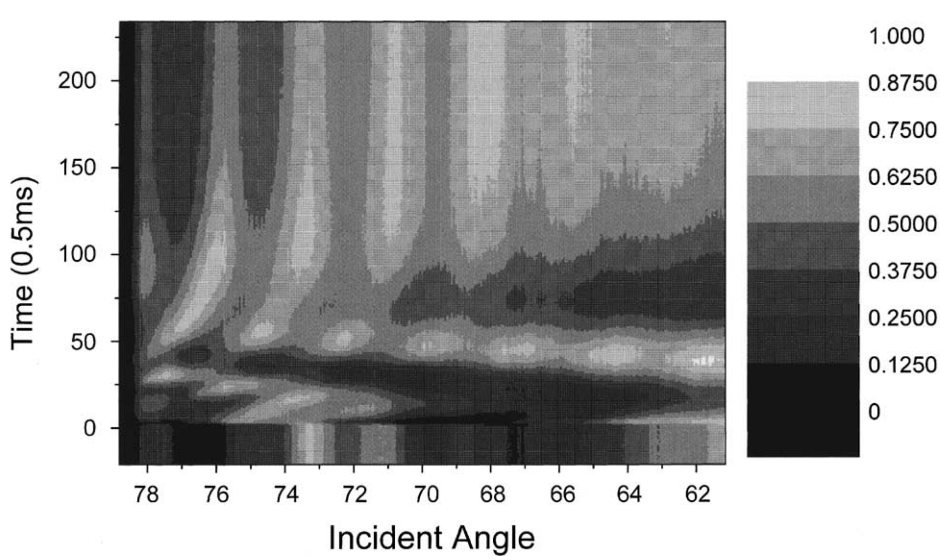

Fig. 2. The dynamic data of T_{ss} after switching off an applied electric field of 6.56 V, the grey scale to the right indicates transmitted intensities.

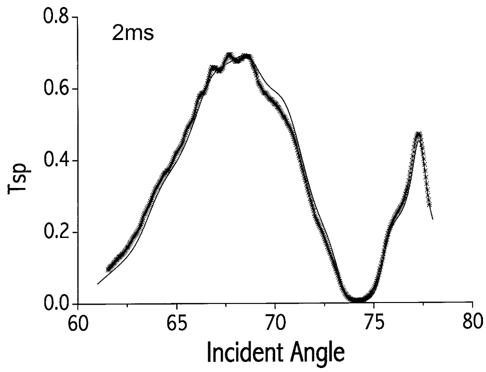

Fig. 3. Dynamic T_{sp} data (crosses) with the first theoretical fit (solid line) 2 ms after removal of the field.

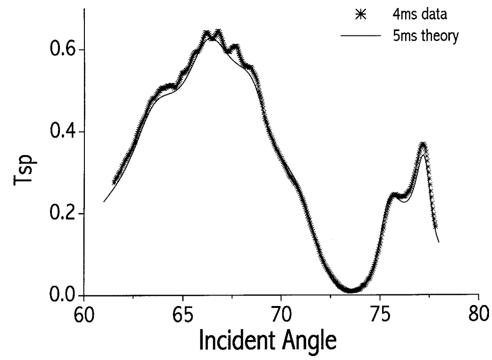

Fig. 4. Dynamic data T_{sp}, (crosses) at 4 ms compared with initial theory at 5 ms.

0.5 ms exposure time and the appropriate rotation speed of the diffuser gives data with a good signal to noise ratio. This time resolution is also quite adequate for the rather slow nematic liquid crystal switching.

There are two types of angle-dependent signals recorded both in transmission and reflection. These comprise polarisation-conserving signals and polarisation-converting signals. The choice is made by selecting the appropriate input and output (before the CCD) polariser angle (p, transverse magnetic or s, transverse electric). Polarisation-conserving reflectivities R_{pp} and R_{ss}, polarisation-conserving transmissivities T_{pp} and T_{ss}, as well as polarisation conversion transmissivity signals T_{ps} and T_{sp} may all be recorded. The first subscript indicates the input polarisation, the second that detected. To establish the initial state of the liquid crystal director within the cell data is first recorded with no voltage applied. Next an ac voltage of 6.56 V (10 kHz) is applied to the cell to give near-homeotropic alignment

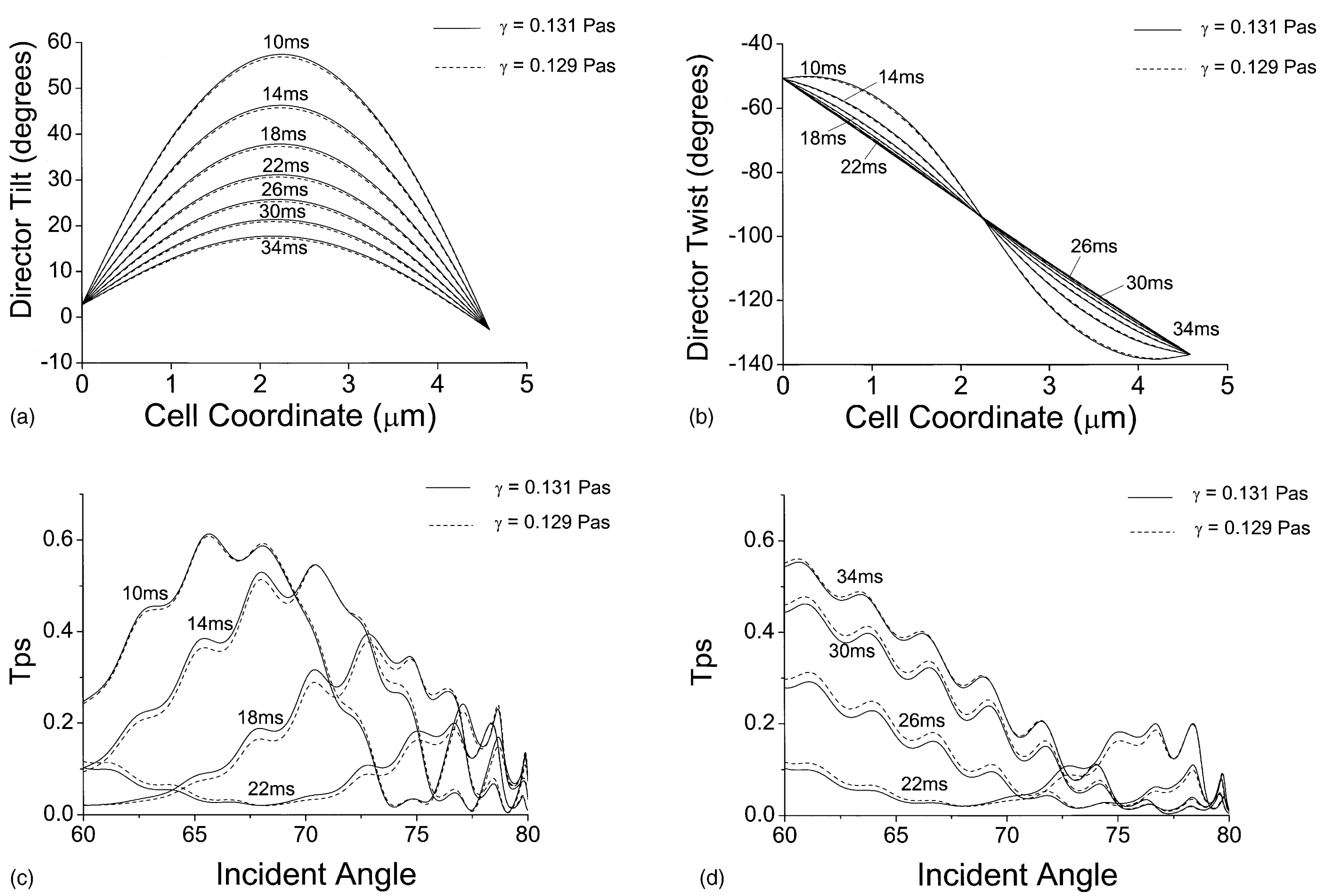

Fig. 5. The influence of the viscosity γ on the time dependence of: (a) the director tilt; (b) the director twist; (c and d) the optical T_{ps} response.

at the centre of the cell and data recorded in the stable voltage-on state of the cell. Fitting to this data will give the static voltage-on director profile. Finally on removal of the voltage from 6.56 V data is taken during the dynamic relaxation of the cell using the CCD with a time step of 0.5 ms to capture the data. One complete set of dynamic data comprises over 500 lines (angle dependent intensities), giving a total recording time for one switch-off of ∼250 ms. Fig. 2 shows how one set of data, T_{ss}, which has clear oscillations in intensity with angle, varies with time. From this figure the striking change in the guided mode structure during the switch-off is apparent, particularly at the early stages. It is also obvious that the cell has a total relaxation time of about 80 ms, well inside the total data recording time.

3. Results and discussion

The angle-dependent reflection and transmission data are fitted, through a multi-dimensional steepest-decent least squares fitting routine, to predictions from model theory using 4 × 4 scattering matrix modelling [22]. Firstly the data taken with no applied field is fitted yielding the optical parameters of the different layers in the cell at 632.8 nm. The primary parameters are those of the liquid crystal layer. These are a thickness of 4.62 μm, and optical permittivities (for ZLI-2293) of $\varepsilon_\perp = 2.2403 + i0.0005$ and $\varepsilon_{||} = 2.6482 + i0.004$. Both surface tilts are of magnitude 2.70°, but of opposite sign, leading to a linear tilt profile through the cell, with zero tilt at the centre. In addition there is a uniform director twist from the top to the bottom through an angle of 82.24°. Other layer parameters are for the layers of ITO thicknesses of 49 nm and optical permittivities of $\varepsilon = 3.158 + i0.08$; and for the polyimide layers anisotropic optical permittivities $\varepsilon_\perp = 2.0946 + i0.001$, $\varepsilon_{||} = 2.2000 + i0.001$ with a thickness of 24 nm. On application of a 10 kHz 6.56 V ac signal to the cell there is a fast director restructuring, which stabilises after a few ms. Data is then again recorded and from fits to this data the static director profile at 6.56 V_{ac} is established. Modelling of the director profile to fit this director profile at 6.56 V_{ac} is based on the Frank–Oseen theory. The fits give a director profile for the cell in which for most of the cell the director is almost homeotropically aligned. This near-homeotropic alignment at the centre of the cell decouples the two surface torques, allowing the surface directors to return to nearer the original 'easy' alignment axes at the two surfaces. Thus the director twist through the cell is found to have increased to 86.09°, within error the same as the original setting twist of 87°. In addition values of the elastic con-

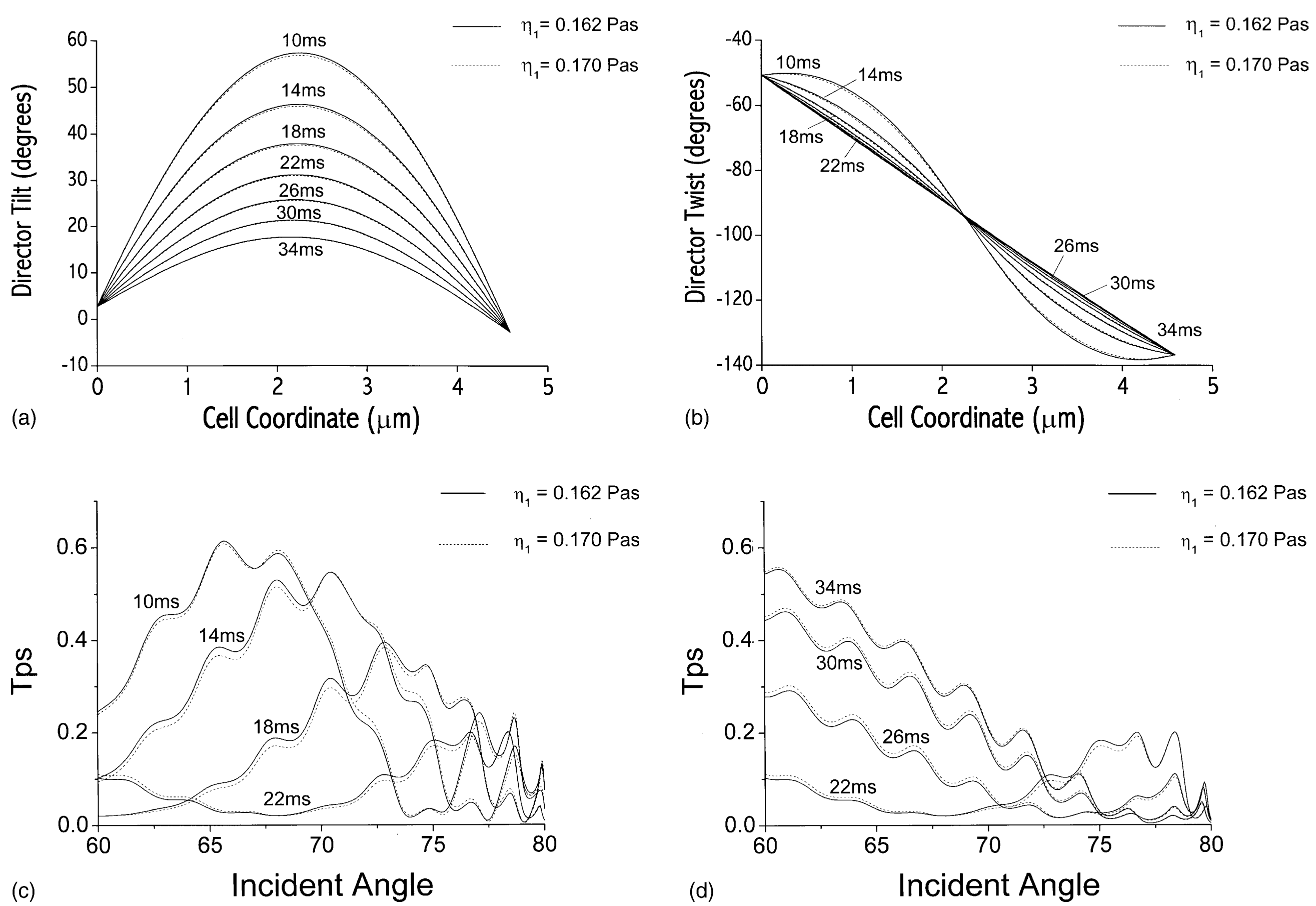

Fig. 6. The influence of the viscosity η_1 on the time dependence of: (a) the director tilt; (b) the director twist; (c and d) the optical T_{ps} response.

stants and the dielectric permittivities of the liquid crystal are determined from the static director profile fits to the optical data, and found to be in good agreement with values supplied by Merck.

Having established the director profile and all boundary layer parameters attention turns to the key part of this study, the dynamic data, taken after the voltage has been switched off from 6.56 V_{ac}. In order to analyse this data more rapidly a commercial modelling program (DIMOS), based on the Ericksen–Leslie hydrodynamic theory [4–8], was employed to model the likely evolution of the director profile with time. This program takes the Berreman–van Doorn [9,10] approximation which assumes that the fluid inertia may be ignored and the flow is restricted to the plane of the cell. Other physical parameters of the liquid crystal, including the elastic constants, the dielectric and optical permittivities, the director surface angles and the cell thickness, are taken from the data fitting at 6.56 V_{ac}. The model dynamic response then depends on the values for the various viscosity coefficients. At the start of the fitting process the viscosity coefficients were set at those given by other studies [23]. The two dominant viscosity coefficients, γ the rotational viscosity coefficient and η_1 the corresponding to the director being parallel to the velocity gradient but perpendicular to the flow were set at $\gamma = 0.149$ Pa s and $\eta_1 = 0.170$ Pa s. The other coefficients were given values of $\eta_2 = 0.017$ Pa s, $\eta_3 = 0.04$ Pa s and $\eta_{12} = 0.0$ Pa s (note the definitions of η_1 and η_2 are those introduced by Helfrich, see for example, Gahwiller [24], and correspond to the original Miesowicz coefficients η_2 and η_1, respectively). Using these values in the DIMOS modelling program together with the exact 4 × 4 multilayer optics scattering matrix routine [22] leads to a set of predicted angle dependent reflectivities and transmissivities as a function of time. For these values it is found that there appears to be quite good agreement between theory and experiment at times up to 2 ms after removal of the field. A fit to T_{sp} data is shown in Fig. 3. However, it soon becomes apparent that the theoretical results predicted after this time corresponded much more closely with the data taken somewhat earlier. Thus, data taken at 4, 5, 6 and 8 ms correspond closely with the model predictions for 5, 6, 7 and 9 ms, respectively. A typical erroneous fit of 5 ms theory to data at 4 ms is given in Fig. 4. This of course simply indicates that the viscosity coefficients initially used were somewhat in error. These are now adjusted to give the best fits to the data at all times. In order to illustrate which viscosity coefficients are important the director response through the cell is modelled for different values for each of the parameters. From these altered director profiles the predicted optical responses are produced and those which indicate the

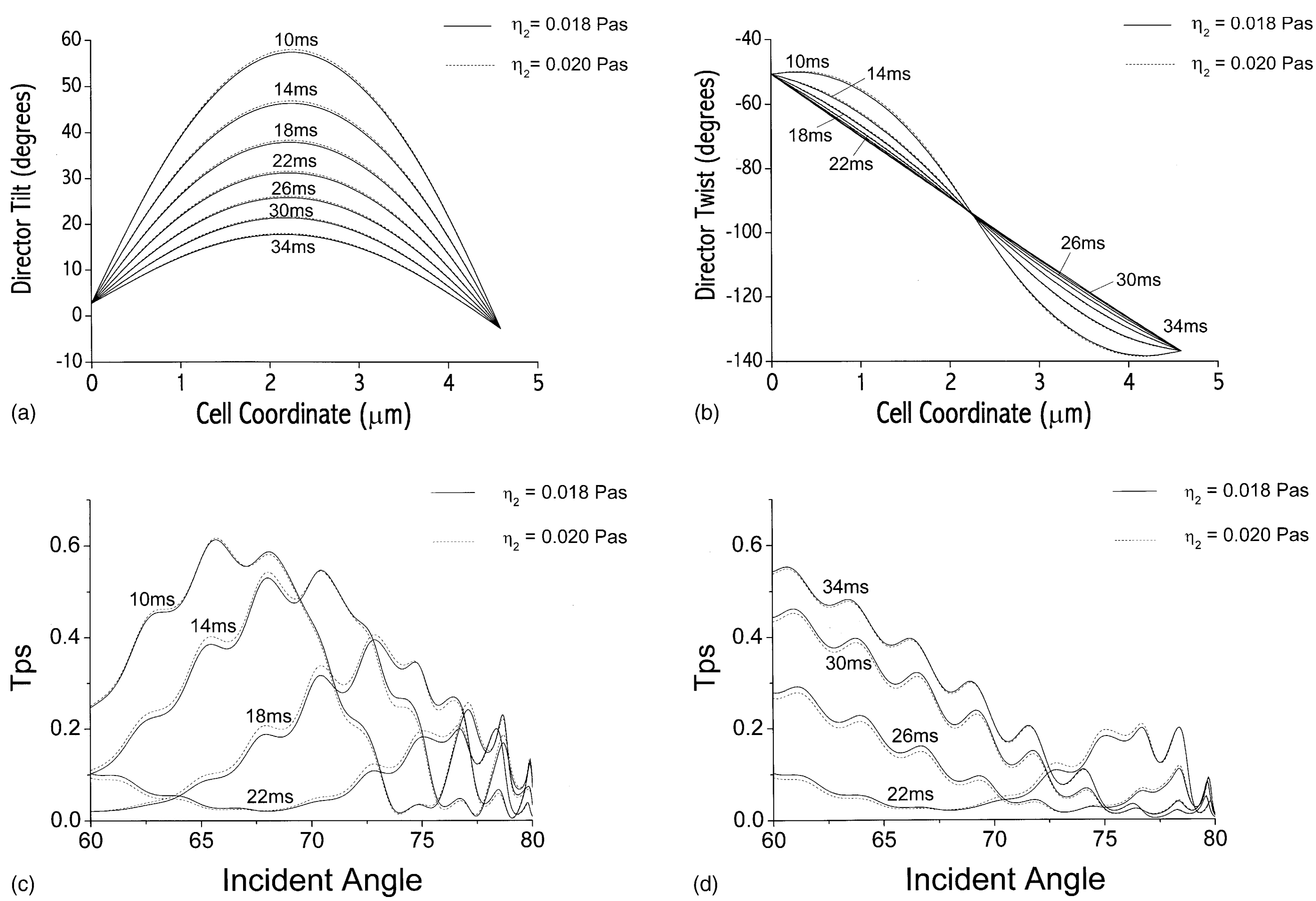

Fig. 7. The influence of the viscosity η_2 on the time dependence of: (a) the director tilt; (b) the director twist; (c and d) the optical T_{ps} response.

changes in the direction required to fit the data guide the next selection of viscosity values.

It is expected that, in the present geometry, the rotational viscosity γ would play the most important role. Fig. 5 shows the predicted director twist Fig. 5a and tilt Fig. 5b profiles for a small change in γ of -0.002 Pa s from the final fit value of 0.131 Pa s. This rather small change in profile leads to the measurable changes in optical response predicted in Fig. 5c and d. Note that here are shown the signals which give most response, that is the polarisation conversion transmission signal T_{ps}. Similarly the changes in the director profile and optical response due to small changes in the other key viscosity η_1 is shown in Fig. 6. Here, to create as significant effect as for γ, the final fitted value of 0.162 Pa s is changed by $+0.008$ to 0.170 Pa s. It is immediately apparent that increasing η_1 has much the same effect as decreasing γ. This is rather surprising and we shall return to this point later.

From this initial modelling it is clear that small changes in γ and η_1 cause very obvious differences in the transmitted signal T_{ps}. By comparison η_2, which corresponds to the director lying parallel to the flow and η_3 which corresponds to the director lying perpendicular to both the flow and velocity gradient are expected to have rather small effects. This is because it is only in the surface layer that these flow conditions prevail. The effect of a 10% increase in η_2 is seen in Fig. 7, while the effect of a 50% increase of η_3 is shown in Fig. 8. It is very obvious that broadly speaking the data will be rather insensitive to variations in these latter two parameters. Finally the affect of η_{12} is illustrated in Fig. 9. Here is shown the influence of changing its value from 0.0 to 0.1 Pa s. At this stage it is worthwhile examining the influence of the various parameters on the tilt and twist profiles. Comparison of Figs. 5a, 6a, 7a, 8a, and 9a show firstly that η_1 is indeed the odd one out in that increasing η_1 has the opposite effect to increasing all the others. It also appears that the influence of γ remains for a very long time, its effect on the tilt profile remaining much the same after even 34 ms. Comparison of the twist profiles reveals that η_{12} has no effect, while again η_1 effects a change which is opposite in character to the remaining viscosities. With this information available an iterative procedure was adopted to fitting all the data, changing first γ and η_1 until the overall response of the system was as close as possible to the data, finally adjusting η_2 and η_3, keeping η_{12} set at zero.

Some fits of the guided mode data, T_{ps}, at different times are shown in Fig. 10. These figures show very clearly, during the very early stages of the director relaxation, the back-flow

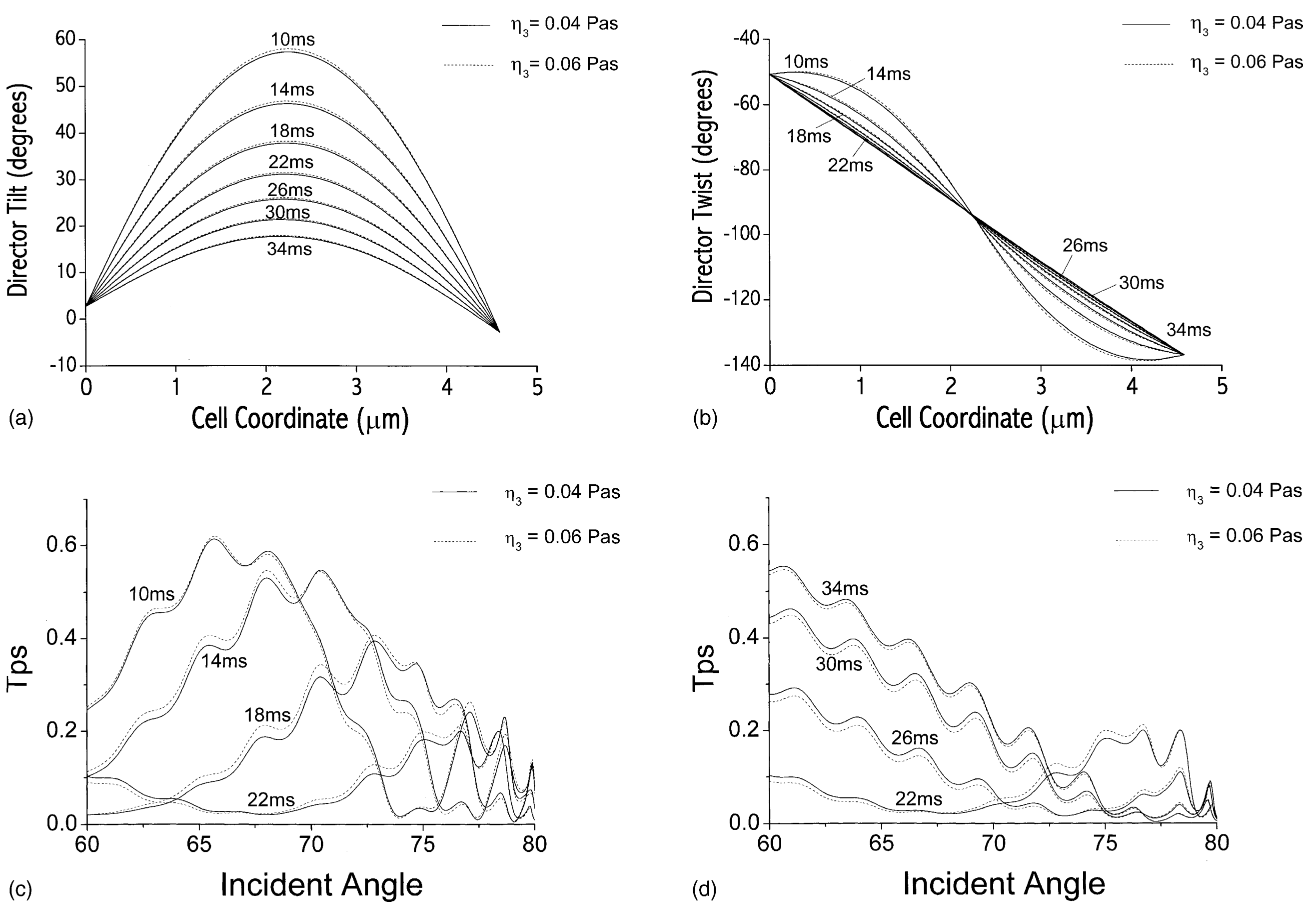

Fig. 8. The influence of the viscosity η_3 on the time dependence of: (a) the director tilt; (b) the director twist; (c and d) the optical T_{ps} response.

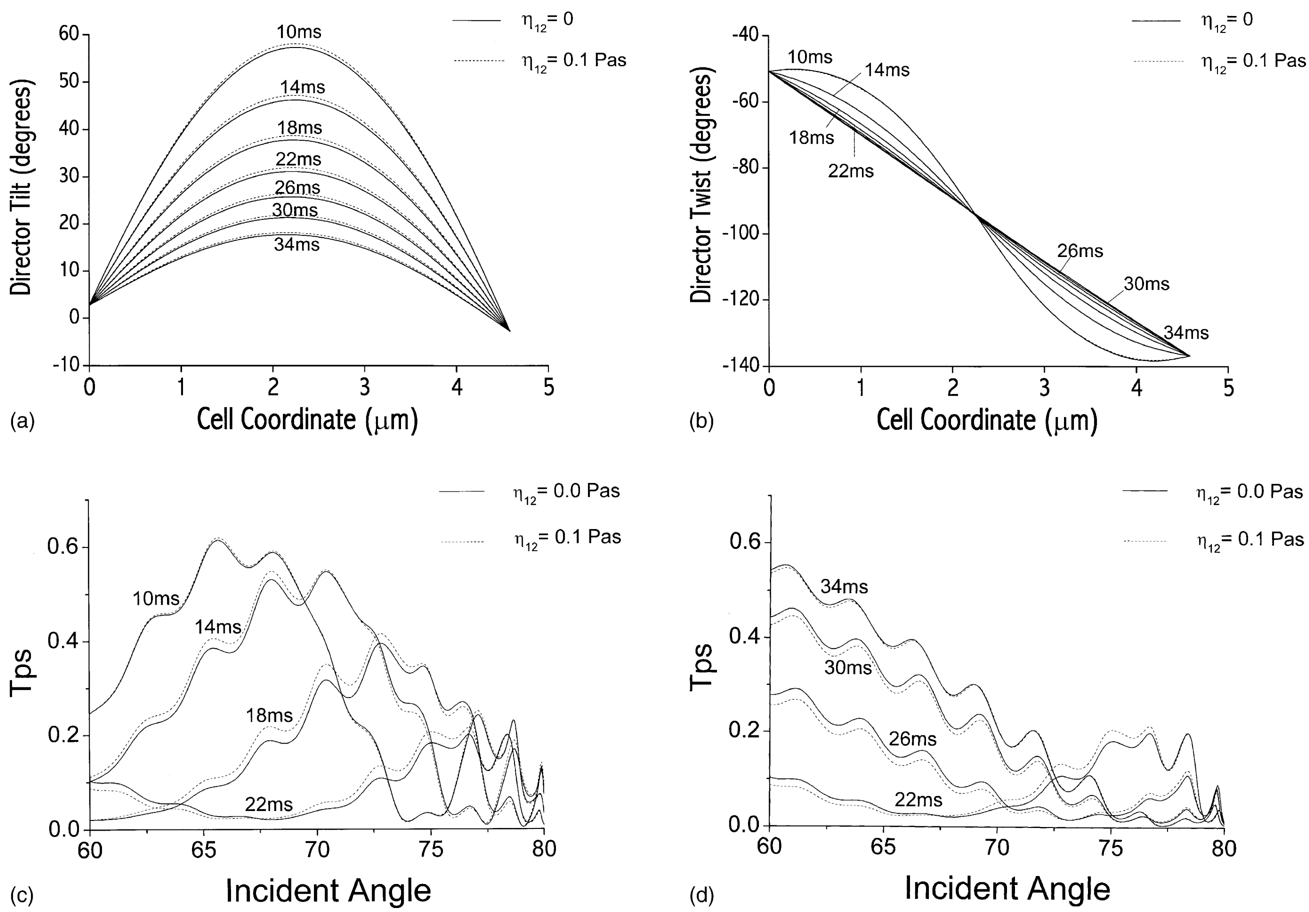

Fig. 9. The influence of the viscosity η_{12} on the time dependence of: (a) the director tilt; (b) the director twist; (c and d) the optical T_{ps} response.

predicted from the Ericksen–Leslie theory. Initially at the centre of the cell the director tilt exceeds 90°, and there is a reverse twist, which recover after a few ms.

This fitting procedure initially failed because the director twists at the surfaces were locked to their initial value with the field on. However, it became apparent that this was an unacceptable constraint and after about 4 ms after removal of the field the director twist on the surfaces had to be changed by about 1°. This is simply explained by the reappearance of the twist structure in the cell, which can once again elastically cause a change of the surface twist-off from the 'easy' axes.

At this stage it is plausible to believe there is substantial degeneracy in the fitting of the five viscosity coefficients to the data. That this is not so is perhaps best illustrated by reference, not to Figs. 4–9, which contain a wealth of detailed information, but simply to the maximum director tilt at the centre of the cell. Fig. 11a shows very clearly the influence of the dominant viscosity γ on this parameter. Notice how increasing γ slows the relaxation down for all times, increases the effect of back-flow on director tilt and delays the time at which maximum tilt is observed. By contrast Fig. 11b may suggest that increasing η_1 actually speeds-up the relaxation. This was also apparently present in the earlier modelling. In actuality what is happening is that increasing η_1 primarily reduces the back-flow, bringing the time for maximum tilt in the centre back towards the beginning, while also initially appearing to speed up the tilt relaxation. Clearly at a later stage an increase in η_1 correctly slows the relaxation down, as expected. Thus these two dominant viscosities have very different influences on the global director response.

11c shows the more subtle response due to changes in η_2. Increasing η_2 causes a decrease in maximum tilt during back-flow while also shifting the maximum tilt to a later time and slowing down the overall relaxation. η_3 has a much weaker effect, as shown in Fig. 11d. Increasing η_3 gives a small increase in the maximum tilt and also shifts the maximum tilt later in time. But these are second order effect by comparison with the dominant viscosities. Finally η_{12} has a strikingly different effect, although to see it the changes in η_{12} have to be substantial. These effects are seen in Fig. 11e.

Thus, by fitting all the data, after removal of the field, from 1 to 70 ms, with the theoretical model, values for all the viscosity coefficients of this liquid crystal at room temperature (in this case ∼20 °C) have been obtained. Several are smaller than the values given by Armitage and Larimer [23] and are shown in Table 1. From the influence of the viscosities on the maximum tilt, the details in the vicinity

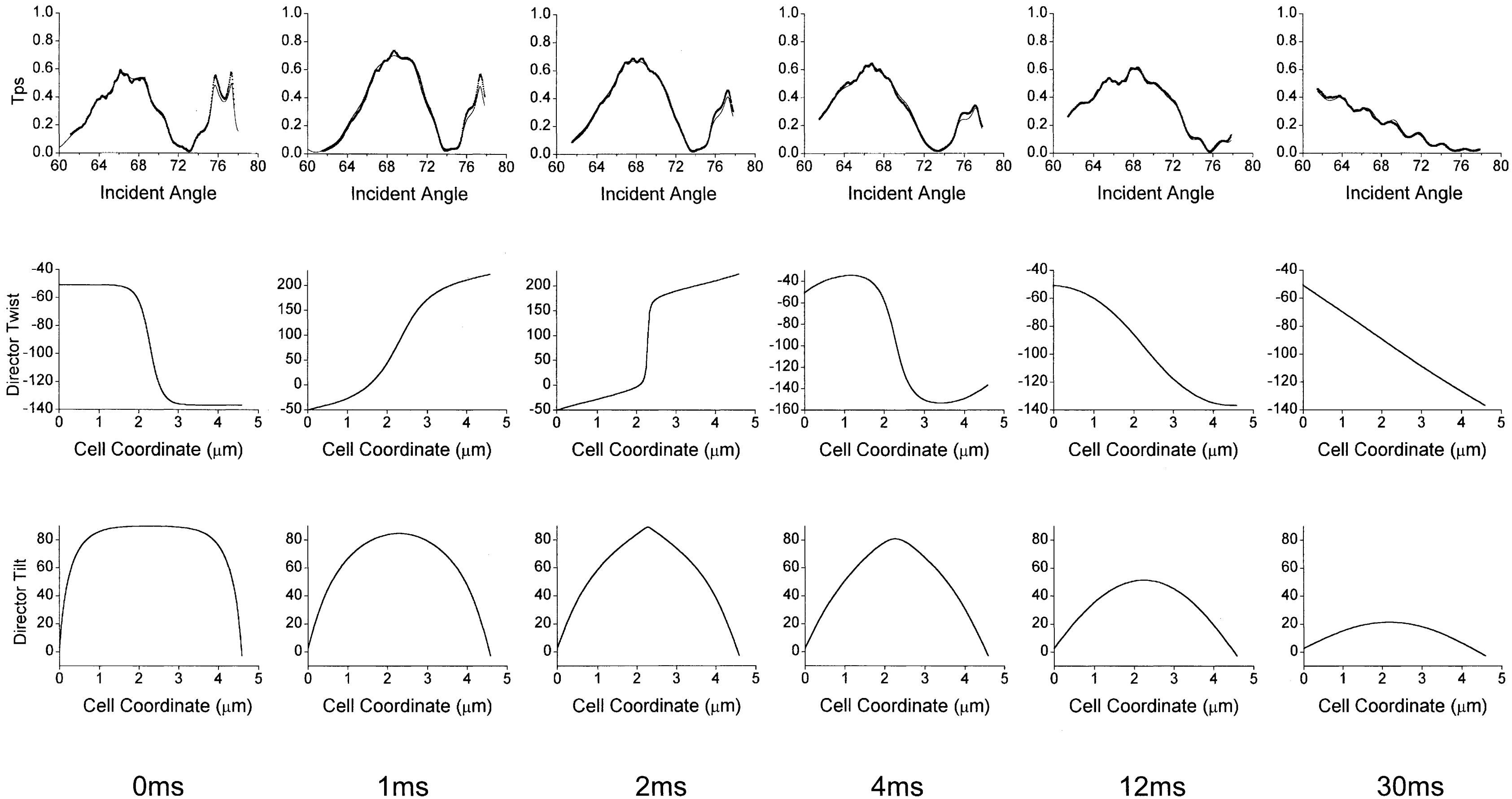

Fig. 10. The final fitted T_{ps} data, the director tilt and twist profiles for various times during the relaxation.

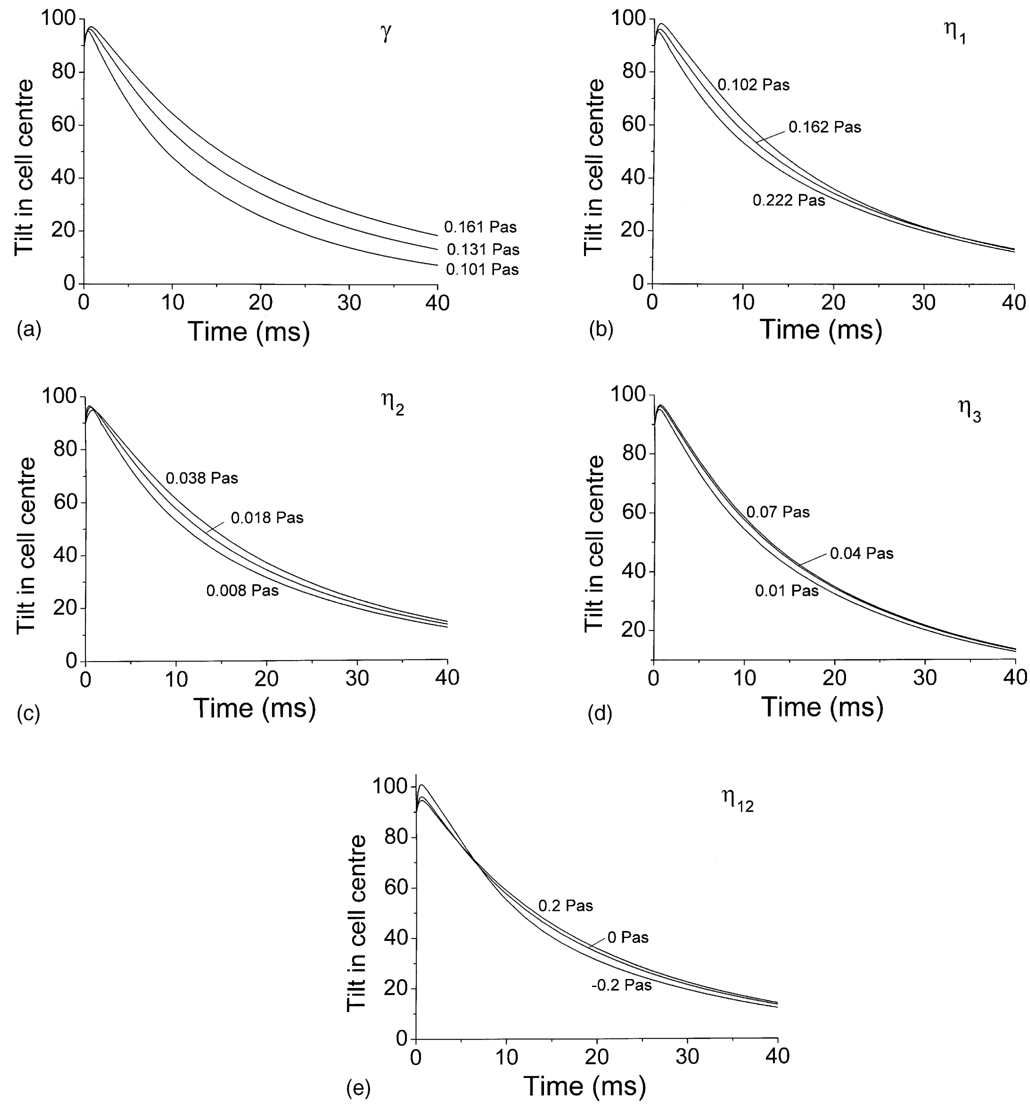

Fig. 11. The effect of the various viscosities on the mid-plane director tilt during relaxation. (a) γ, (b) η_1, (c) η_2, (d) η_3, (e) η_{12}.

of the maximum back-flow and the long time behaviour the influences of the various viscosities may be separated to a large extent. Given then the uncertainty of the director profile established from data fitting it is possible to put some levels of uncertainty to each viscosity coefficient. On average, it is possible to detect the director tilt angle in the cell centre to a precision of better than 0.5°, this leads to the uncertainties given in Table 1.

All this data fitting shows excellent agreement between these new experimental results and the theoretical calculations. This confirms unequivocally the validity of the Ericksen–Leslie theory and its Berreman and van Doorn approximation.

Table 1
Values of the room temperature viscosities for the liquid crystal ZLI-2293

$\gamma = 0.131 \pm 0.001$ Pa s
$\eta_1 = 0.162 \pm 0.004$ Pa s
$\eta_2 = 0.018 \pm 0.001$ Pa s
$\eta_3 = 0.04 \pm 0.01$ Pa s
$\eta_{12} = 0.00 \pm 0.04$ Pa s

4. Conclusions

The Ericksen–Leslie theory of the dynamic behaviour of nematics has been tested by optical guided wave probing of the relaxation of a twisted nematic cell. By use of a convergent beam system together with the fully-leaky optical guided mode technique it has been possible to quantify in detail the director relaxation of the cell on removal of an ac field. There is substantial back-flow in the first few ms after switching, as predicted by the theory. The director tilt at the centre of the cell increases to over 96°with reverse twist, before recovering at longer times.

Use of computer modelling based on the Ericksen–Leslie theory has allowed an examination of the influence of the various viscosity coefficients upon the director relaxation and, through a scattering matrix routine, upon the optical signals expected. Detailed fitting of all the optical data taken over the whole of the director relaxation gives the dominant viscosity coefficients γ and η_1 of the liquid crystal ZLI-2293 very accurately. In addition good estimates are also obtained for the other viscosity coefficients. These numbers, given in Table 1, are the full set of viscosity coefficients required for this nematic liquid crystal. The relaxation time constant was also obtained. All these values, obtained optically here agree with previously determined values.

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