Rheol Acta (2000) 39: 294–300

ORIGINAL CONTRIBUTION

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Dynamics of shear aligning of nematic liquid crystal monodomains

Received: 13 September 1999
Accepted: 24 January 2000

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Presented at EuroRheo 99-1, May 3–7, 1999, Sophia-Antipolis, France

Abstract The equations of linear and angular momentum for nematic liquid crystals have been described with Ericksen's transversely isotropic fluid [TIF] model and solved for start-up of shear flow at constant rate and varying initial alignment conditions. An analytical solution for the rotation provides predictions of the nematic director which closely agree with experimental results of Boudreau et al. (1999), supporting the validity of Ericksen's TIF model. The solution is limited to flows where the effects of director gradients are negligible.

Key words Linear and angular momentum · Nematic liquid crystals · TIF model

Introduction

Liquid crystals can have the spatial disorder and flow behavior of liquids while displaying the anisotropic properties of crystals. In the nematic phase, the *centers-of-gravity* of the liquid crystal molecules are randomly positioned in space, showing no long-range order, yet are aligned towards a common direction, the director **n**. The anisotropy of nematics leads to complex flow behavior that has been the focus of many studies over the past few decades (Larson, 1999).

The continuum theory of Leslie and Ericksen (Leslie, 1968) has proven very effective in describing the flow behavior of small nematic liquid crystals. When subject to shear flow, it is well known that nematic liquid crystals will either align in the direction of shear within a small angle θ_L (against vorticity) or will tumble with vorticity, finding no preferred orientation. This behavior was predicted by Leslie (1968) and experimentally observed (Gähwiller, 1972; Meiboom, 1973; Skarp; 1981). Theoretical investigation of tumbling phenomena in nematics has been specific to alignments in the vorticity plane (Carlsson, 1984; Burghardt and Fuller, 1990), ($\phi = 0$), or to numerical analysis of the Leslie-Ericksen equations (Han and Rey, 1993; Han and Rey, 1995). The flow aligning-tumbling behavior of small-molecule nematics has also been predicted using molecular theories (Archer and Larson, 1995; Kröger and Sellers, 1995).

Here we explore predictions of the continuum theory for nematics in start-up of shear flow at constant rate and compare the predictions to experimental observations of Boudreau et al. (1999) who studied the dynamic response of small, shear aligning nematic liquid crystals, 4-*n*-pentyl-4′-cyanobiphenyl [5CB] and *N*-(4-methoxybenzylidene)-4-butylaniline [MBBA]. Samples were sheared with various initial alignment conditions so a fair assessment of the continuum theory's ability to predict the response of nematics to shear should be possible using their experimental results.

Continuum Theory

The Leslie-Ericksen continuum theory of nematic liquid crystals (Ericksen, 1961; Leslie, 1968) has been described extensively (Stephen and Straley, 1974; Leslie, 1979; de Gennes and Prost, 1993; Larson, 1999) so only a few points will be discussed here. In continuum theory two vector fields, the velocity, υ, and the director, **n**, describe the dynamic phenomena of the nematic. The coupling between the director orientation and flow fields requires two balance laws, the balance of linear momentum and the balance of angular momentum.

If we assume the nematic to be incompressible then the director is constrained to unit length. The director is also assumed to be non-polar such that **n** and **−n** are identical. The relative importance of viscous and distortional elastic forces on the behavior of the nematic is described through the Ericksen number, Er, which is the ratio of viscous torques to elastic torques and is given by (Larson, 1999)

$$\mathrm{Er} = \frac{\eta \dot{\gamma} l^2}{K} \tag{1}$$

where η, $\dot{\gamma}$, l and K are the characteristic viscosity, shear rate, length, and elastic constant of the system, respectively.

Director Rotation in Shear Flow

We begin the study of nematic liquid crystals in shear flow by defining a Cartesian coordinate system according to Fig. 1. The x-component is in the shearing direction, the y-component in the gradient direction, and the z-component in the vorticity direction thus making the x-y plane the vorticity plane and the x-z plane the shear plane. The director is defined by its two polar angles, θ and ϕ, where θ is the tilt angle of the director out of the shear plane and ϕ is the rotation angle of the director out of the vorticity plane. In Cartesian component form, the director will then be written as

$$\begin{aligned} n_x &= \cos\theta\cos\phi, \\ n_y &= \sin\theta, \\ n_z &= \cos\theta\sin\phi \ . \end{aligned} \tag{2}$$

With this definition, the unit length constraint on the director is automatically satisfied. To avoid the confusion of multiple representations of the director in polar form, we limit the range of θ and ϕ to

$$-90° \le \theta \le 90°; \quad 0° \le \phi \le 180° \ . \tag{3}$$

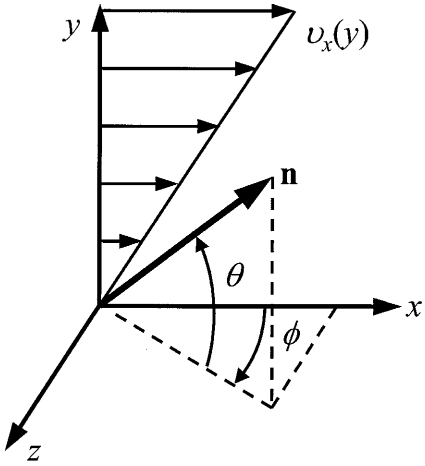

Fig. 1 Cartesian coordinate system with x the shear direction, y in the gradient direction and z in the vorticity direction. The director is defined with the polar angles θ, the rotation out the shear plane, and ϕ, the rotation out of the vorticity plane

To simplify the analysis we assume that $\mathrm{Er} \gg 1$ and that no other forces act on the nematic so that we can neglect all spatial gradients in the director field such that the liquid crystal will behave as a single monodomain. The velocity gradient for shear can then be assumed uniform across the sample and is defined as

$$\upsilon_x = \dot{\gamma} y, \quad \upsilon_y = 0, \quad \upsilon_z = 0 \ . \tag{4}$$

These simplifications yield a set of differential equations for the balance of angular momentum during transient shear flow (Leslie, 1987)

$$(\alpha_3 - \alpha_2)\frac{\mathrm{d}\theta}{\mathrm{d}t} + \dot{\gamma}\left(\alpha_3\cos^2\theta - \alpha_2\sin^2\theta\right)\cos\phi = 0 \tag{5}$$

$$(\alpha_3 - \alpha_2)\cos\theta\frac{\mathrm{d}\phi}{\mathrm{d}t} - \dot{\gamma}\alpha_2\sin\theta\sin\phi = 0 \ , \tag{6}$$

where α_2 and α_3 are the second and third Leslie viscosity coefficients. We also impose initial conditions on the director

$$\theta(t=0) = \theta_0, \quad \phi(t=0) = \phi_0 \ . \tag{7}$$

To solve these equations, we begin by following Leslie's (1987) procedure of eliminating time in these equations to yield

$$\frac{\mathrm{d}\theta}{\mathrm{d}\phi} = \frac{\left(\alpha_2\sin^2\theta - \alpha_3\cos^2\theta\right)}{\alpha_2\tan\theta\tan\phi} \ . \tag{8}$$

Integration gives

$$\frac{\sin^2\phi}{\sin^2\phi_0} = \frac{\tan^2\theta - \alpha_3/\alpha_2}{\tan^2\theta_0 - \alpha_3/\alpha_2} \ . \tag{9}$$

The stable solutions (Leslie, 1968) for the director angles,

$$\begin{aligned} \theta &= \theta_L, \quad \phi = 0° \\ \theta &= -\theta_L, \quad \phi = 180° \end{aligned} \tag{10}$$

are defined by the Leslie angle, θ_L, where

$$\tan^2\theta_L = \alpha_3/\alpha_2 \ . \tag{11}$$

The two solutions result from the constraint that **n** = −**n**; so only one solution, $\theta = \theta_L$ and $\phi = 0°$, needs to be considered. $(\alpha_3/\alpha_2) > 0$ predicts the well known shear aligning behavior which was found for many nematic liquid crystals where hydrodynamic torques align the liquid crystal in the direction of shear but tilted out of the shear plane against vorticity by an angle θ_L. When $(\alpha_3/\alpha_2) < 0$, there is no solution and the liquid crystal will find no preferred orientation, indicating tumbling director behavior.

This result of Leslie is important in understanding the steady-state flow phenomena of liquid crystals but does not address transient flow situations where the Leslie-Ericksen model has also been used. Predictions on flow induced instabilities have been made by examining small deviations from a director initially in the vorticity direction (Pieranski and Guyon, 1973; Leslie, 1976; Manneville and Dubois-Violette, 1976). The nematic response to oscillatory shear has been studied for flow alignment instabilities (Clark et al., 1981) and for determining the ratios (α_3/α_2) and ($|\alpha_2|/K_3$) (Mather et al., 1995). In the following analysis we will neglect elastic forces, using Ericksen's TIF model for the extra-stress tensor.

In this study, we are interested in the transient angle $\theta(t)$ during the start-up of shear in shear aligning nematics. A general time dependent solution to Eqs. (5) and (6) can be found by first eliminating the strain rate dependence when dividing Eq. (5) by the strain rate $\dot{\gamma}$, and then eliminating ϕ with Eq. (9). The resulting differential equation is

$$\frac{d\theta}{d\gamma} = \left(\frac{\tan^2\theta_L - \tan^2\theta}{1 - \tan^2\theta_L}\right) \times \cos^2\theta\sqrt{1 - \sin^2\phi_0\left(\frac{\tan^2\theta_L - \tan^2\theta}{\tan^2\theta_L - \tan^2\theta_0}\right)} \quad . \qquad (12)$$

Integration yields the strain, γ, as a function of θ

$$\gamma(\theta) = \left(\frac{\tan^2\theta_L - 1}{2\tan\theta_L}\right)\left[\ln\left(\frac{\tan\theta_L - \tan\theta}{\tan\theta_L + \tan\theta}T(\theta)\right) - \ln\left(\frac{\tan\theta_L - \tan\theta_0}{\tan\theta_L + \tan\theta_0}T(\theta_0)\right)\right] \quad , \qquad (13)$$

where $T(\theta)$ is a function of the initial angles θ_0 and ϕ_0:

$$T(\theta) = \frac{1 - \sin^2\phi_0\left(\frac{\tan^2\theta_L + \tan\theta_L\tan\theta}{\tan^2\theta_L - \tan^2\theta_0}\right) + \sqrt{1 - \sin^2\phi_0\left(\frac{\tan^2\theta_L - \tan^2\theta}{\tan^2\theta_L - \tan^2\theta_0}\right)}}{1 - \sin^2\phi_0\left(\frac{\tan^2\theta_L - \tan\theta_L\tan\theta}{\tan^2\theta_L - \tan^2\theta_0}\right) + \sqrt{1 - \sin^2\phi_0\left(\frac{\tan^2\theta_L - \tan^2\theta}{\tan^2\theta_L - \tan^2\theta_0}\right)}} \quad . \qquad (14)$$

The solution is explicit in $\gamma(\theta)$ but can not be inverted analytically into $\theta(t)$ or $\phi(t)$. However, evaluation of the solution to provide values for γ (Eq. 13) and ϕ (Eq. 9) is straightforward.

Figure 2 shows plots of the dimensionless tilt angle Θ, defined as

$$\Theta = \frac{\theta - \theta_0}{\theta_L - \theta_0} \quad , \qquad (15)$$

versus the strain for different values of the initial rotation angle ϕ_0 and with $\theta_0 = 0°$. The only material parameter is θ_L. In this example we use $\theta_L = 7°$ as reported for MBBA by Gähwiller (1972). For $\phi_0 < 45°$, Θ has little dependence on the initial rotation angle, ϕ_0, appearing to approach the stable solution of $\Theta = 1$ in a simple exponential manner. As ϕ_0 approaches 90°, however, a sigmoidal behavior of Θ becomes increasingly apparent, being characterized by an increasing incubation period before Θ's approach steady-state. With $\theta_0 = 0°$, as $\phi_0 \to 90°$ the incubation period approaches infinity since the condition ($\theta = 0°$, $\phi = 90°$) is an unstable steady-state. Figure 3 shows plots of (ϕ/ϕ_0) versus strain for the same conditions as in Fig. 2; since ϕ and θ are related through Eq. (9) the primary focus of further discussion will only be on the behavior of θ during shear.

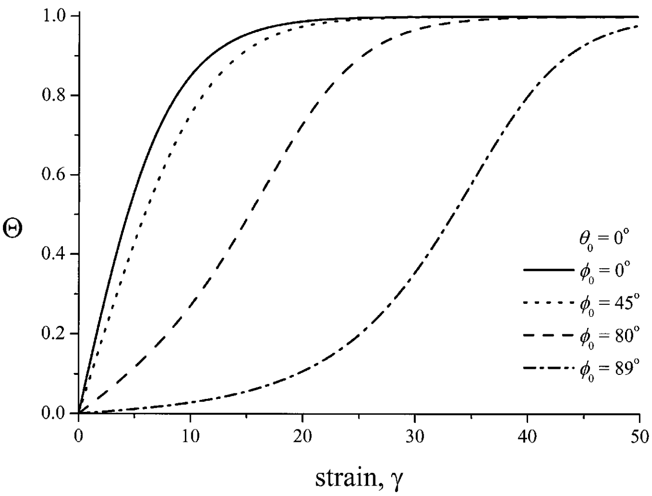

Fig. 2 Theoretical predictions for the start-up of shear flow for the dimensionless tilt Θ versus strain with $\theta_0 = 0°$, $\theta_L = 7°$ and $\phi_0 = 0°$ (—), 45° (···), 80° (- -) and 89° (-··-)

When the initial tilt angle of the director is within the range $-\theta_L < \theta_0 < \theta_L$, the director response is sigmoidal, as described above, and Eq. (13) provides a unique solution for $\gamma(\theta)$. However, when the initial director alignment lies in the range $\theta_L < \theta_0 < 90°$ and $90° < \phi_0 < 180°$, the director response is no longer sigmoidal and a unique solution for γ for a given θ no longer exists. Figure 4 shows a characteristic example of the director tilt angle response with an initial alignment in the non-sigmoidal range. The trajectory of the director from its initial state to steady-state is shown in Fig. 5. Hydrodynamic torques initially rotate the director with vorticity around the z-axis. This increases

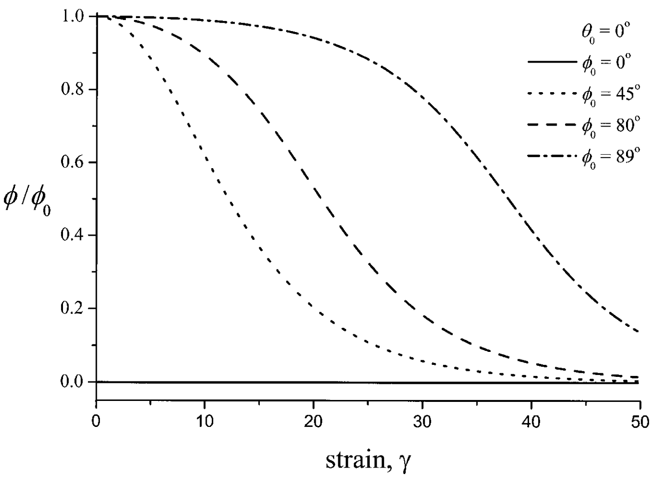

Fig. 3 Theoretical predictions for the start-up of shear flow for the rotation angle (ϕ/ϕ_0) versus strain with $\theta_0 = 0°$, $\theta_L = 7°$ and $\phi_0 = 0°$ (—), 45° (···), 80° (- -) and 89° (-··)

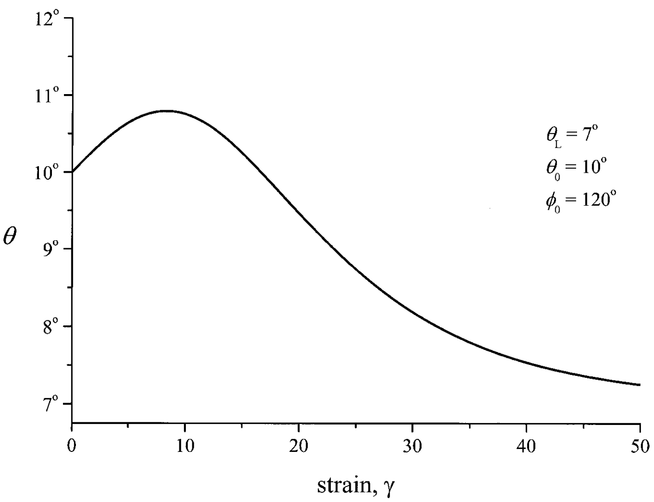

Fig. 4 Theoretical predictions for start-up of shear flow for tilt angle θ versus strain with $\theta_0 = 10°$, $\theta_L = 7°$ and $\phi_0 = 120°$

the tilt angle, θ, until $\phi < 90°$, after which rotation of the director around the z-axis decreases the tilt angle as it approaches the Leslie angle. When the director shows this type of behavior, Eq. (13) can still be used to predict the director response. Starting from the initial alignment, Eq. (13) is solved for the strain, γ, in a marching procedure to the maximum tilt angle, θ_{max}. Equation (13) is then reinitialized with $\theta_0 = \theta_{max}$ and the corresponding value for ϕ_0 according to Eq. (13), then solved in a marching procedure as $\theta \rightarrow \theta_L$. The two solutions are then combined to give the entire director response.

So far we have assumed the director response to be independent of Er. With extreme shear conditions, either very low or very high Er, this behavior may no longer exist. For example, when the director is normal or near normal to the vorticity plane, Pieranski and Guyon (1973) show that for shear rates less than a critical value (or Er less a critical value) elastic torques caused by wall anchoring will stabilize the director in its initial orientation. The director will thus remain normal to the vorticity plane instead of rotating towards it. Conversely, at higher shear rates (higher Er) the very long incubation times predicted when the director is initially normal to the vorticity plane may not be experimentally observed. Flow instabilities may cause the director to rotate out of its initial state and towards the vorticity plane earlier than predicted (Pieranski and Guyon, 1973; Leslie, 1976).

Approximate solution for $|\theta_0| < \theta_L$

To fit experimental data, Boudreau et al. (1999) employed a fit function of a much simpler form than Eq. (13) for cases where $|\theta_0| < \theta_L$. We would now like to develop that approximate expression using the analytic solution and evaluate its validity. We begin by considering the case where the director is initially aligned with velocity, $\phi_0 = 0°$ and $\theta_0 = 0°$. Under these initial conditions, Eq. (13) can be simplified to yield θ as a function of γ. The small angle approximation, tan $\theta \approx \theta$ and tan $\theta_L \approx \theta_L$, reduces Eq. (13) to

$$\Theta = \frac{\exp\left[\frac{\gamma}{\gamma_L}\right] - 1}{\exp\left[\frac{\gamma}{\gamma_L}\right] + 1}, \quad \gamma_L = \frac{1 - \tan^2\theta_L}{2\tan\theta_L} \quad . \tag{16}$$

The characteristic strain, γ_L, is fully defined by the Leslie angle, θ_L. Equation (16) suggests that an approximation of the solution given in Eq. (13) may be of the following form

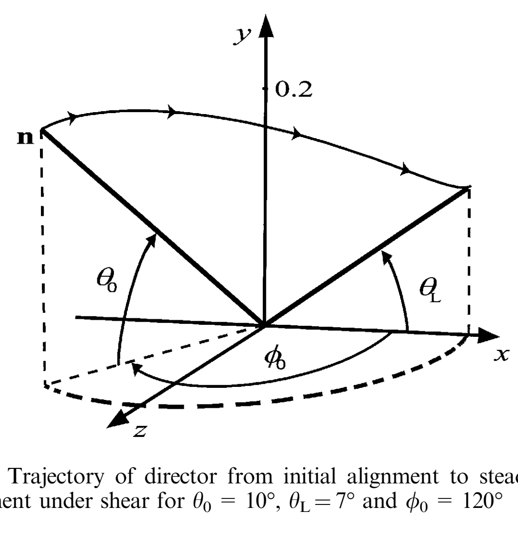

Fig. 5 Trajectory of director from initial alignment to steady-state alignment under shear for $\theta_0 = 10°$, $\theta_L = 7°$ and $\phi_0 = 120°$

$$\Theta^* = \frac{\exp\left[\left(\frac{\gamma}{\gamma_{0.46}}\right)^m\right] - 1}{\exp\left[\left(\frac{\gamma}{\gamma_{0.46}}\right)^m\right] + 1} \quad , \tag{17}$$

where Θ^* is an approximation of Θ and $\gamma_{0.46}$ is the characteristic strain where $\Theta^* = (e-1)/(e+1) \approx 0.46$, as proposed by Boudreau et al. The parameters $\gamma_{0.46}$ and m can be found by equating the values and slopes of Θ^* and Θ at $\theta = \theta_{0.46}$. $\gamma_{0.46}$ is then given by Eq. (13) with $\theta = \theta_{0.46}$,

$$\theta_{0.46} = \frac{(e-1)(\theta_L - \theta_0)}{(e+1)} - \theta_0 \quad , \tag{18}$$

and has the following form

$$\gamma_{0.46} = x\gamma_L \tag{19}$$

where $x = fn\ (\phi_0, \theta_0, \theta_L)$ and is equal to the square-bracketed term of Eq. (13) for $\theta = \theta_{0.46}$. Once $\gamma_{0.46}$ is known, the exponent m can be found through differentiation of Eq. (17) with respect to γ and with Eq. (12):

$$m = x\frac{(e+1)^2}{4e(\theta_L - \theta_0)}\frac{\tan^2\theta_L - \tan^2\theta_{0.46}}{\tan\theta_L}\cos^2\theta_{0.46} \times \sqrt{1 - \sin^2\theta_0\left(\frac{\tan^2\theta_L - \tan^2\theta_{0.46}}{\tan^2\theta_L - \tan^2\theta_0}\right)} \tag{20}$$

In the special case of an initial alignment with $\theta_0 = 0°$ and for small Leslie angle ($<20°$), x and m can be approximated as

$$x \approx 1 + \ln\left[\frac{(e+1) - 2\sin^2\phi_0 + \sqrt{(e+1)^2 - 4e\sin^2\phi_0}}{(e+1) - 2e\sin^2\phi_0 + \sqrt{(e+1)^2 - 4e\sin^2\phi_0}}\right] \tag{21}$$

$$m \approx x\sqrt{1 - \frac{4e}{(e+1)^2}\sin^2\phi_0} \quad . \tag{22}$$

The dependence of x and m on θ_L drops out for $\theta_0 = 0°$. If, in addition, the director lies in the vorticity plane, $\phi_0 = 0°$, x and m adopt limiting values, $x = 1$ and $m = 1$, so that Eq. (17) reduces to Eq. (16).

Figures 6 and 7 show the dependence of x and m on ϕ_0 and θ_0 for $\theta_L = 7°$. For $-\theta_L < \theta_0 < \theta_L$, the director response to shear is simple enough to be captured by Eq. (17). When $\theta_L < \theta_0 < 90°$ and $90° < \phi_0 < 180°$, the director response to shear is too complicated to be captured by Eq. (17), which is unable to predict the relative maximum seen in Fig. 4. Within this region of initial alignments, marked as the dashed lines in Figs. 6 and 7, x and m can only be used to give $\gamma_{0.46}$ and the slope of Θ at $\gamma_{0.46}$.

A comparison of Θ and Θ^* is shown is Fig. 8 for the same conditions as in Fig. 2 ($\theta_0 = 0°$). We see good agreement between Θ^* and Θ for both high and low values of initial rotation angle ϕ_0. Further comparisons show similar agreement over the entire valid range of Eq. (17).

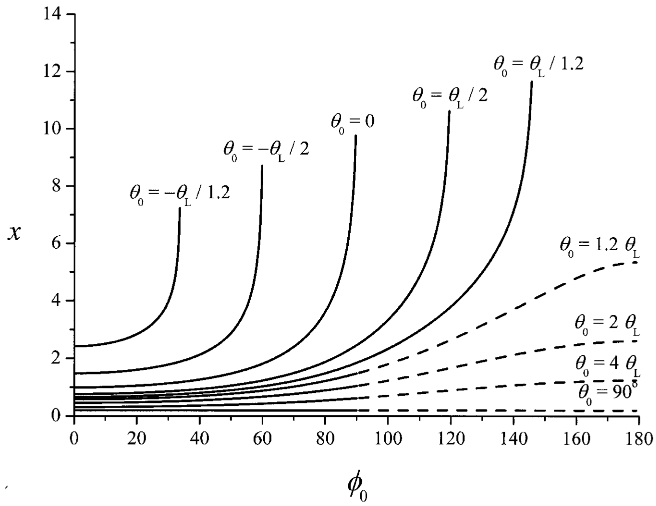

Fig. 6 x versus initial rotation ϕ_0 with the initial tilt angle θ_0 as a parameter

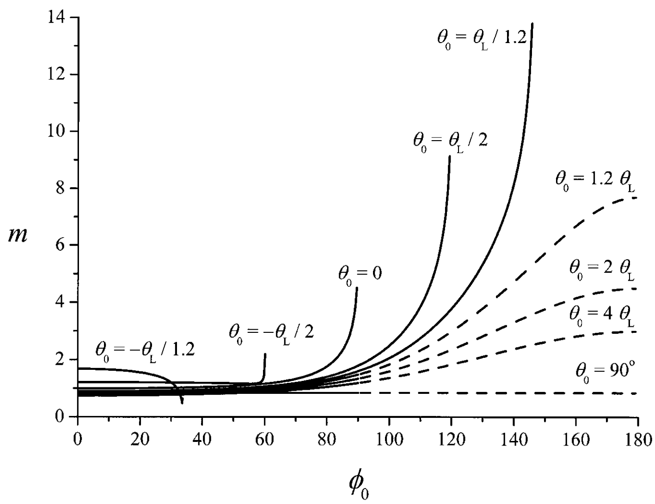

Fig. 7 m versus initial rotation ϕ_0 with the initial tilt angle θ_0 as a parameter

Comparison with Experiment

Recently, Boudreau et al. (1999) studied the orientational changes of small nematic liquid crystals, 4-n-pentyl-4′-cyanobiphenyl [5CB] and N-(4-methoxybenzylidene)-4-butylaniline [MBBA] under shear start-up conditions for various initial orientations. Orientation of the liquid crystals was observed through conoscopy

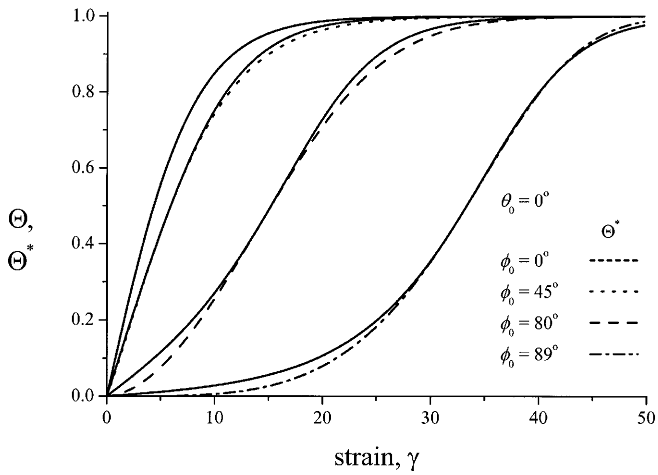

Fig. 8 Dimensionless tilt angle Θ versus strain for analytical solution (*solid lines*), Eq. (14), and approximation, Eq. (17), with $\theta_0 = 0°$, $\theta_L = 7°$ and $\phi_0 = 0°$, 45°, 80° and 90°

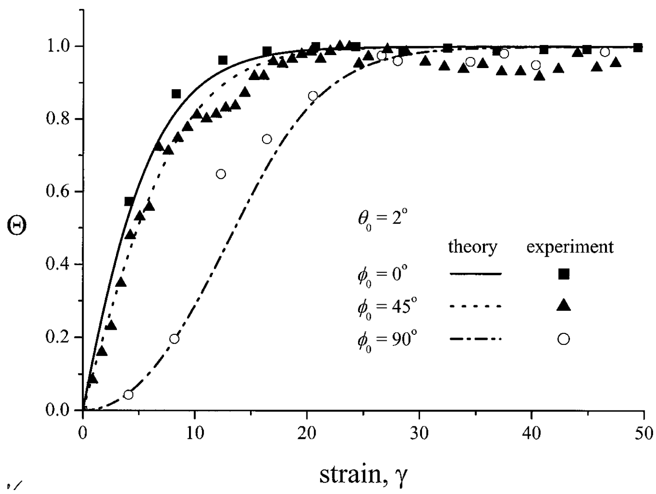

Fig. 9 Evolution of dimensionless tilt angle Θ for the start-up of shear flow for MBBA. Predictions made with $\theta_0 = 2°$, $\theta_L = 7°$, and $\phi_0 = 0°$ (—), 45° (···) and 90° (-··-). Experimental results with $\phi_0 = 0°$ (■) (Er = 2374), 45° (▲) (Er = 983) and 90° (○) (Er = 2374)

(Born, 1969; Wright, 1911) while initial alignment conditions were achieved through anchoring at the boundaries by rubbing of polyimide surface coatings on containing glass walls. This method will impose a small initial tilt angle θ_0 (Boudreau et al., 1999; Weiss et al., 1998). The initial rotation angle ϕ_0 can be chosen freely.

To get a sample with an average initial tilt of 0°, anchoring surfaces were buffed such that one surface would have a pre-tilt of around 2° and the other around −2°. This produces a splay of the director from one surface to the other such that the sample may slightly deviate from monodomain conditions. Since our solution to the director response does not consider spatial distributions of the director field, we need to assume that the observed response of splayed samples is that of monodomains. As a first approximation we assume that the observed director response of these splayed samples is dominated by the alignment with the fastest approach to steady-state. In all experiments studied, this alignment was that at the wall where the tilt angle was maximum, +2°. Therefore, all predictions to the experimental results of Boudreau et al. were made for monodomains with an initial tilt of $\theta_0 = 2°$.

For high Er, anchoring can safely be neglected except for providing the initial director alignment, θ_0 and ϕ_0. Boudreau et al. looked at both low and high Er flows but only those at high Er (>900) will be examined here so as to exclude elastic effects near walls as much as possible.

Figure 9 compares the reported experimental tilt angle to the θ-predictions for $\phi_0 = 0°$, 45° and 90° for MBBA. The predictions agree with the experimental results over the entire range of high Er conditions studied thereby indicating that Ericksen's TIF model accurately describes the director dynamics of small nematic liquid crystalline molecules.

Conclusions

The director response of nematic liquid crystals (with $(\alpha_3/\alpha_2) > 0$) to the start-up of shear has been predicted by solving the equations of linear and angular momentum for Ericksen's TIF model. The only material parameter is the Leslie angle, θ_L. For shear aligning nematics, the director always rotates to the alignment director of Eq. (10), but rotation is delayed with increasing ϕ_0. The analytical solution has been closely approximated by a simple sigmoidal function for $|\theta_0| < \theta_L$. Initial director angles, θ_0 and ϕ_0, can be freely chosen according to experimental conditions.

The theoretical predictions of director rotation quantitatively agree with experimental observations for high Er shear flow of MBBA with $\theta_0 = 2°$ and $\phi_0 = 0°$, 45° and 90°. Modeling calculations show that the small deviation of 2° from the perfect initial director alignment, $\theta_0 = 0°$, has a large influence on the start-up behavior. Further experiments are in progress with the objective of alternating the alignment conditions from (+2°,−2°) to (+2°,+2°).

Acknowledgements The research is supported by the National Science Foundation (grant DMR9422180). We thank David M. Boudreau for discussions of his experimental work. The anonymous review process was conducted by R. Larson as member of the Rheologica Acta editorial board.

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