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The Effect of Dissolved Side-Group Polymers on Pattern Dynamics in Nematic Liquid Crystals in a Rotating Magnetic Field

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Abstract. — Patterns formed by inversion walls in nematic layers exposed to a rotating magnetic field were studied. Dilute solutions of a mesogenic side group polymethacrylate in a low molecular weight liquid crystal (5CB) were used in comparison with the pure solvent. As found in a previous work, in this system the intensity of backflow (fluid flow induced by director rotation) can be controlled by the polymer concentration due to a specific increase of shear viscosity coefficients. In the synchronous regime of director rotation no significant effects of backflow on the dynamics of the walls are observed. Dynamic solitons known from the synchronous regime were also found at asynchronous rotation, when soliton lattices are formed by continuous nucleation. Here comparison with theory for given values of the lattice period shows soliton currents significantly reduced by backflow. Two of the three additional pattern forming states exclusively found at asynchronous rotation were completely suppressed in solutions with sufficient polymer concentration. The third of these states is affected by backflow in the growth rate of its patterns. Numerical calculations were performed to explain behavior of the patterns in the asynchronous state. For pure 5CB a quantitative comparison with the experiment was possible.

1. Introduction

Pattern formation in liquid crystalline systems has attracted considerable interest as a remarkable example of self-organization of systems far from equilibrium. Both transient [1–4] and stationary periodic states [5–11] were studied. Transient patterns occur when a magnetic field $\mathbf{B}$ of sufficient strength is suddenly applied perpendicular to the initial director $\mathbf{n}_0$ of a nematic liquid crystal. As a result, a periodically distorted director field is favorable rather than a uniformly distorted one despite the penalty in elastic energy of the system. The periodic response produces a faster rotation of the director in comparison with that for non-periodic reorientation. This transient state represents a particular example of pattern forming phenomena in

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systems which are driven far from equilibrium. The transient periodic state is unstable with respect to the final uniformly distorted non-periodic equilibrium state. However for systems steadily maintained far from equilibrium (using rotating magnetic or electric fields) periodic patterns may be observed for a long time.

Different patterns are stable at certain ranges of the external driving parameters. These ranges depend on material properties of the system. Material parameters of low molecular weight liquid crystals are controlled by their chemical structure and may be varied only in a limited range. In this article a study of pattern formation in polymer nematic solutions is presented, where viscous material parameters can be controlled in a wider range.

As was shown in a previous article on the bend Frederiks transition [12], backflow (i.e. convective flow induced by director rotation) can be strongly damped if polymer side-group mesogenic molecules are dissolved in a low molecular weight liquid crystal. This effect is especially strong for polymers with high values of the molecular weight. We also found a very strong influence of polymer molecules on formation of transient patterns at the twist Frederiks transition [4]. It was found that the onset of pattern formation is shifted to very high values of the field even for dilute polymer solutions. Applying the theory of Lonberg, Fraden, Hurd and Meyer [1], one can calculate the Miesowicz shear viscosity coefficient $\eta_a$ from these critical field values. In Figure 1, $\eta_a$ obtained in this way is shown in comparison with the twist viscosity $\gamma_1$ as a function of the polymer weight fraction in the same nematic system as studied in the present paper. A large contribution of macromolecules to the shear viscosity coefficient $\eta_a$ compared to that for the rotational viscosity $\gamma_1$ is observed. Gu et al. [13] performed dynamic light scattering studies using nematic solutions of a similar polymer. Again, with increasing polymer concentration a larger increase of the Miesowicz shear viscosity coefficient $\eta_a$ compared to that of $\gamma_1$ can be deduced from the effective viscosities for different modes of the scattering experiment.

Fig. 1. — Dependence of the Miesowicz shear viscosity $\eta_a$ and the twist viscosity $\gamma_1$ on the weight fraction $w$ of the polymer PMC-312 in a 5CB solution at $T = 23 \, ^\circ C$ (from Ref. [4]).
This behavior is predicted by a theory of Brochard [14] for the case of nearly spherical shape of the dissolved polymer coils. The additional contribution $\Delta \gamma_1$ of polymer molecules to the rotational viscosity in the regime of dilute solutions should be given by

$$\Delta \gamma_1 = C \frac{(R_{||}^2 - R_\perp)^2}{R_\perp^2 / \lambda_{||} + R_{||}^2 / \lambda_{\perp}},$$

(1)

where $R_{||}$ and $R_{\perp}$ are the coil dimensions measured parallel and perpendicular to the nematic director, $\lambda_{||}$ and $\lambda_{\perp}$ are the friction coefficients of the macromolecule for translational motions parallel and perpendicular to the director, and $C$ is the number density of dissolved chains. Clearly, the concentration dependence of $\gamma_1$ should be small in case of a small form anisotropy of the coil, $R_{\perp}^2 \approx R_{||}^2$. In comparison to that the contributions $\Delta \eta_a$, $\Delta \eta_b$ and $\Delta \eta_c$ to the Miesowicz coefficients should all scale with the square of the coil dimension, for example

$$\Delta \eta_a = C \lambda_{\perp} R_{\perp}^2,$$

(2)

because a coil extension parallel to the flow velocity gradient will increase the shear stress in any way, as known from solutions of ordinary polymers.

This selective influence of macromolecules on the viscosity coefficients of a nematic liquid crystal changes drastically its dynamics. Thus, uniform states become more favorable than periodic ones even for high values of the external field.

The mechanism of pattern formation far from equilibrium proposed by Meyer et al. for several pattern types in nematic systems [1,2,5,7] is based on the concept of viscosity reduction. Since the average rotation rate of the director is higher for the periodic response, the effective value of the rotational viscosity $\gamma_1^{\text{eff}}$ is smaller than the pure rotational viscosity $\gamma_1$ which controls non-periodic (uniform) reorientation.

The viscosity reduction mechanism is intrinsically related to the backflow effect. This effect was studied in detail for the bend Frederiks transition [15] and has also been found to be important for the formation of periodic structures in the splay and twist geometries.

In our article, we present the study of patterns for nematic side chain polymer solutions in a rotating magnetic field. A variety of different states which occur for these conditions were discovered and classified by Migler and Meyer [5]. A particular state occurs at certain values of the magnetic field $B$ and its angular frequency $\omega$. In the synchronous regime ($\omega \tau < 1$), where $\tau$ defines a characteristic time scale of the system, the formation of dynamic solitons (propagating inversion walls) was discovered. When nucleated repeatedly by dust particles, these solitons form systems of concentric rings (soliton lattices or target patterns). Similar target patterns were observed in freely suspended films of smectic C [9] and smectic C* [10,11] liquid crystals in rotating electric fields. Also the formation of spirals was observed [7-9].

The dependence of the velocity of individual solitons on the rotation frequency may be explained using the driven overdamped sine–Gordon equation studied by Büttiker and Landauer [16]. The discrepancy of this theory and the experiment observed in [5] was explained hypothetically by a possible influence of the backflow on the soliton motion. Thus, the utilization of polymer solutions, for which the backflow is known to be strongly damped, should provide a useful tool to check this hypothesis.

In the asynchronous regime ($\omega \tau > 1$), the behavior of the nematic director is rather complex. Different pattern forming states in the asynchronous regime may be observed for different values of the external control parameters ($B$ and $\omega$). With increasing magnetic field at constant frequency one observes a decrease in symmetry of the patterns. Starting from very regular structures at relatively low fields the system undergoes several successive transitions from regular states to a complex, chaotic state. The situation is similar to that for electrohydrodynamic
patterns which are explained in detail by Ribotta [17]. The transition from regular patterns to chaotic ones is one of the most important problems for pattern forming systems. Hydrodynamic flow or convection plays a crucial role in these transitions. More ordered structures show coherent convection correlated on a large scale whereas for chaotic states this scale is strongly reduced. Therefore polymer solutions may provide us with better understanding of all these phenomena because material parameters of the system responsible for the backflow may be controlled.

2. Experimental Section

2.1. Samples. — A polymethacrylate with mesogenic side groups, poly[4-(4-methoxyphenylazo)phenoxy]butyl]methacrylate (PMC-312) was used:

\[
\text{CH}_3 - \text{C} - \text{COO} - (\text{CH}_2)_4 - \text{N} = \text{N} - \text{OCH}_3
\]

Synthesis of this polymer was reported elsewhere [18]. We used a sample with \( M_n = 110\ 000 \) \((N_n = 312)\), \( M_w/M_n = 1.12 \). The glass transition temperature is \( T_g = 86 \) °C, and the nematic-isotropic transition occurs at \( T_{NI} = 144 \) °C. The polymer sample was dissolved in a low molecular weight liquid crystal, pentylcyanobiphenyl (5CB) with \( T_{NI} = 35.5 \) °C for 30 min at \( T = 120 \) °C. Careful determination of clearing temperatures of the nematic solutions was made with a hot stage equipped polarizing microscope.

The range of the polymer weight fraction \( w \) in the solutions was \( 0 \leq w \leq 2.12\% \). The values of \( T_{NI} \) for the solutions (measured as the mean value of the biphasic nematic–isotropic temperature range) were practically equal to that for pure 5CB.

2.2. Experiments. — In order to obtain homogeneous homeotropic orientation of the nematic director, we used glass plates with edge lengths 16 × 16 × 0.9 mm\(^3\) coated with lecithin (from 0.5% solution in ethyl alcohol). The thickness \( d \) of the cells was controlled by mylar spacers (75 \( \mu \text{m} \leq d \leq 100 \) \( \mu \text{m} \)). All measurements were performed at the temperature \( T = 23 \pm 0.1 \) °C.

The optical scheme to observe and to record to periodic patterns included a microscope objective, crossed polarizer and analyzer oriented at angles of ±45° to the magnetic field, a charge coupled device (CCD) video camera and a video recorder. A video digitizer (Screen Machine II, FAST Electronic) was used to interface the video signal to a personal computer.

Instead of rotation of the magnetic field we used rotation of the cell. From the theoretical point of view rotation of the cell is equivalent to rotation of the field. The rotation of the cell has to be very smooth to prevent undesirable vibrations. For this purpose we used a stepper motor combined with a gear reduction providing very smooth rotation. The angular frequency \( \omega \) was controlled by the step rate of the motor.

For determination of the bend and splay elastic constants of 5CB and the polymer solutions the effective birefringence of the layer was measured as a function of the magnetic field as described by Saupe [19]. The field was varied sufficiently slow, adapted automatically to the field dependent response time of the director. As soon as the field \( B \) exceeds the Frederiks threshold field \( B_F \), the intensity of transmitted He–Ne laser light (for crossed polarizers) changes as a result of the changing phase difference between ordinary and extraordinary beam. The number of oscillations \( N \) in intensity is related to the effective value of the refractive index of the
distorted nematic layer for light polarization parallel to $\mathbf{B}$. Experimental field dependencies of $N$ were fitted to the theoretical dependence [19] with given values of the extraordinary and ordinary refractive indices for 5CB, $n_e = 1.715$ and $n_o = 1.529$ [20]. The resulting fitting parameters $B_P$, $d$, and $\kappa = K_1/K_3 - 1$ ($K_1$ and $K_3$ being the splay and bend elastic constants) were obtained with high accuracy. Values of $K_3$ were calculated from

$$K_3 = \frac{\chi_a}{\mu_0} \left( \frac{B_P d}{\pi} \right)^2,$$

where $\chi_a$ is the anisotropy of the diamagnetic susceptibility. A value of $\chi_a = 1.357 \times 10^{-6}$ (in SI units) for 5CB at $T = 23^\circ C$ was obtained by torque equilibrium experiments in superposed electric and magnetic fields, described in reference [21].

3. Theoretical Background

3.1. Uniform Rotation. — The first experiment on the measurement of the twist viscosity coefficient using a rotating magnetic field was performed by Tsvetkov [22]. A tube filled with a nematic liquid crystal was suspended in a horizontal magnetic field rotating with constant angular velocity $\omega$ around the vertical axis $z$. Later Brochard, Léger and Meyer [23] studied the dynamics of the director for a homeotropic cell in a rotating magnetic field. The geometry of the experiment is shown in Figure 2. The orientation of the director $\mathbf{n}$ is given by two angles, $\varphi$ and $\theta$. The orientation of the magnetic field $\mathbf{B}$ rotating in the $x - y$ plane at the time $t$ is given by the angle $\omega t$. The phase lag angle $\alpha = \omega t - \varphi$ between the projection of $\mathbf{n}$ on the $x - y$ plane $\mathbf{n}_\perp$ and $\mathbf{B}$ provokes the magnetic torque per volume $\Gamma_m = \chi_a B^2/(2\mu_0) \sin(2\alpha)$ acting on the director. There are two other torques, viscous and elastic, which affect the dynamics of the director. The elastic torque per volume due to bend and splay distortion in the homeotropic cell is given by $\Gamma_{el} = K(\partial^2 \theta/\partial x^2)$, where the one constant approximation $K = K_1 = K_3$ is used. For large values of $B$, $\Gamma_{el}$ acts only in thin layers near the glass surfaces where the director orientation varies from $\theta = 0$ to $\theta = \pi/2$. The typical thickness of these boundary regions is given by the magnetic coherence length

$$\xi = \frac{1}{B} \left( \frac{\mu_0 K}{\chi_a} \right)^{1/2},$$
which is small compared to \( d \). Therefore for strong fields these thin regions have no effect on the director dynamics in the bulk. The torque balance for the director in the bulk is given by:

\[
\gamma_1 \left( \omega - \frac{\partial \alpha}{\partial t} \right) = \frac{\chi_a B^2}{2\mu_0} \sin(2\alpha). \tag{5}
\]

For high values of \( B \) the pure rotational viscosity \( \gamma_1 \) can be used in equation (5). For low values of \( B \), however, it has to be replaced by an effective value, reduced due to backflow. There are two regimes of the director rotation, synchronous and asynchronous. In the synchronous regime the director follows \( B \) and rotates with the same angular frequency \( \omega \) as \( B \). The phase lag angle \( \alpha \) is constant in this case. In the asynchronous regime the director cannot follow the field and \( \alpha \) is increasing continuously. Note that the maximum value of \( \Gamma_m \) at a given field \( B \) occurs when the phase lag angle reaches the value \( \alpha = \pi/4 \); if the magnetic field is not strong enough to fix the angle at \( \alpha \leq \pi/4 \) the transition from synchronous to asynchronous behavior takes place.

The synchronous steady state solution of equation (5) reads

\[ \omega \tau = \sin(2\alpha), \tag{6} \]

where \( \tau \) is a characteristic time of the system:

\[ \tau = \frac{2\gamma_1 \mu_0}{\chi_a B^2}. \tag{7} \]

The condition \( \omega \tau = 1 \) defines a characteristic field which corresponds to the transition to the asynchronous regime:

\[ B_a(\omega) = \left( \frac{2\gamma_1 \mu_0 \omega}{\chi_a} \right)^{1/2}. \tag{8} \]

In the asynchronous regime \( \alpha \) periodically increases with time by \( \pi \) in a period \( T_h \) given by

\[ T_h = \frac{\pi \tau}{[(\omega \tau)^2 - 1]^{1/2}}. \tag{9} \]

### 3.2. Dynamic Solitons.

Different pattern forming states were described by Migler and Meyer [5-7]. In the synchronous regime dynamic solitons can be observed. These solitons are inversion walls created from some inhomogeneities (for instance, dust particles) or by the side boundaries of the cell. The reason is a local increase of the torque counteracting the rotation of the director near the dust particle. As a result, the phase lag angle \( \alpha \) near the dust particle is higher than in the bulk. This leads to an elastic distortion near the inhomogeneity. Nucleation occurs when this distortion becomes too large; the director near the particle is retarded by an angle of \( \pi \) with respect to the bulk orientation, creating a soliton of the amplitude \( \pi \) [5]. This soliton propagates outward and the diameter of the ring increases. In the rotating magnetic field the director inside the inversion walls also rotates. These walls are splay-bend walls which move in the rotating field. The dynamics of solitons may be studied using the torque balance equation for torques acting around the \( z \)-axis in the approximation \( n_z = 0 \):

\[
K \frac{\partial^2 \alpha}{\partial x^2} - \gamma_1 \frac{\partial \alpha}{\partial t} + \gamma_1 \omega - \frac{\chi_a B^2}{2\mu_0} \sin(2\alpha) = 0. \tag{10}
\]

The first term in equation (10) is the elastic torque per volume due to the bend and splay distortion in the inversion wall. The other terms are the same as in equation (5). Using \( \xi \) and \( \tau \) for the characteristic length and time scales one obtains

\[
2\xi \frac{\partial^2 \alpha}{\partial x^2} - \tau \frac{\partial \alpha}{\partial t} - \sin(2\alpha) + \omega \tau = 0. \tag{11}
\]
This equation has the form of the driven overdamped sine–Gordon equation [16] which is known to have a soliton solution in the form \( \alpha(x, t) = \alpha(x - vt) \) where \( v \) is the soliton velocity. With increasing time the soliton of the amplitude \( \pi \) always moves in direction of decreasing phase lag. The shape of the solitons for low values of \( \omega \tau \) is in good approximation given by the static solution

\[
\alpha(x) = 2 \arctan[\exp(-x/\xi)]
\]  

of (11) plus a homogeneous offset given by equation (6). For values of \( \omega \tau \) close to unity the steady state solution results in a broader, asymmetric shape.

A similar equation of motion holds for smectic C* layers in a rotating electric field [11], where \( \alpha \) has to be replaced by \( \alpha/2 \) due to the different symmetry of director field and ferroelectric interaction, and solitons of the amplitude \( 2\pi \) are formed.

For a given value of \( \omega \tau < 1 \) one can find a soliton solution with a value of the reduced velocity \( v/v_0 \) which depends on \( \omega \tau \). Here \( v_0 \) is defined as \( v_0 = 2\xi/\tau \). These velocities were compared with experimental values by Migler and Meyer [5], however, they found a discrepancy with the experiment. A possible influence of fluid backflow was proposed as a reason for this discrepancy. In addition the one–constant approximation might cause some deviations.

At low values of \( \omega \tau \) a structural transition to non–propagating static solitons is observed, which are explained as inversion walls in which the director twists out of the plane [5,8], saving elastic energy due to the low twist elastic constant \( K_2 \).

3.3. Viscosity Reduction Lattice. — In the regime of asynchronous director rotation three different pattern forming states were observed, denoted by Migler and Meyer [5] as (in the order of increasing field) viscosity reduction lattice (VRL), viscosity reduction lattice transverse instability (VRL–TI), and complex state. When initiated by a point–like distortion of the director field, the VRL is formed as a system with an increasing number of shrinking rings. The rings again are identified as inversion walls. In contrast to the dynamic solitons, however, after initiation no source or sink of phase lag is necessary to maintain the state; the VRL itself acts as a phase lag sink. The VRL–TI structure is similar to the VRL structure, but with an additional undulation of the inversion walls. The complex structure, which is formed spontaneously in contrast to the VRL and VRL–TI, consists of irregular formed inversion walls.

In a concept of a viscosity reduction mechanism Migler and Meyer incorporated the coupling between director rotation and fluid flow in the equations of motion to explain the dynamics of the VRL [6]. They restricted the calculations to the case of planar inversion walls moving along \( x \). In this case the problem is invariant with respect to the \( y \) direction. It should be a good approximation also for walls forming rings, when their radius is large enough to neglect curvature effects. Further in a strong field limit the director was assumed to have no \( z \) component,

\[
u(x, t) = (\cos \varphi(x, t), \sin \varphi(x, t), 0),
\]  

with

\[
\varphi(x, t) = \omega t - \alpha(x, t).
\]

The fluid flow velocity \( \mathbf{v}_f \) was considered to be in \( y \) direction and, in order to incorporate an important finite–thickness effect, to have a parabolic \( z \) dependence with zero velocity at the boundaries \( z = \pm d/2 \):

\[
\mathbf{v}_f(x, z, t) = (0, v_f(x, z, t), 0),
\]  

\[
v_f(x, z, t) = v_0(x, t) \left[1 - \left(\frac{2z}{d}\right)^2\right].
\]
Table I. — Values of \( K_1, K_3, \kappa = K_1/K_3 - 1 \), and \( \gamma_1 \) for solutions with a weight fraction \( w \) of PMC-312 in 5CB. For the 2\% solution only \( \gamma_1 \) was determined.

<table>
<thead>
<tr>
<th>( w/% )</th>
<th>( K_1 \times 10^{12}/N )</th>
<th>( K_3 \times 10^{12}/N )</th>
<th>( \kappa )</th>
<th>( \gamma_1/(Pa\cdot s) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>6.22</td>
<td>8.42</td>
<td>-0.261</td>
<td>0.092</td>
</tr>
<tr>
<td>0.52</td>
<td>6.05</td>
<td>8.19</td>
<td>-0.261</td>
<td>0.105</td>
</tr>
<tr>
<td>1.05</td>
<td>6.01</td>
<td>8.26</td>
<td>-0.273</td>
<td>0.145</td>
</tr>
<tr>
<td>2.12</td>
<td></td>
<td></td>
<td></td>
<td>0.215</td>
</tr>
</tbody>
</table>

Application of the Ericksen–Leslie formalism [24] results in coupled equations of motion given by the balances of torques and forces (neglecting inertial terms):

\[
(K_3 \cos^2 \varphi + K_1 \sin^2 \varphi) \frac{\partial^2 \varphi}{\partial x^2} - \frac{1}{2} (K_3 - K_1) \sin(2\varphi) \left( \frac{\partial \varphi}{\partial x} \right)^2 + \frac{\Delta \chi B^2}{2\mu_0} \sin[2(\omega t - \varphi)]
\]

\[-\gamma_1 \frac{\partial \varphi}{\partial t} + \frac{1}{2} \left[ \gamma_1 + \gamma_2 (\sin^2 \varphi - \cos^2 \varphi) \right] \frac{\partial \psi}{\partial t} = 0,
\]

(17)

\[
[\eta_1 + (\alpha_1 \cos^2 \varphi + \gamma_2) \sin^2 \varphi] \frac{\partial^2 \psi}{\partial x^2} + 2[\alpha_1 (\cos^2 \varphi - \sin^2 \varphi) + \gamma_2] \cos \varphi \sin \varphi \frac{\partial \psi}{\partial x} \frac{\partial \varphi}{\partial x}
\]

\[-\frac{1}{2} [\gamma_1 - \gamma_2 (\cos^2 \varphi - \sin^2 \varphi)] \frac{\partial^2 \varphi}{\partial x^2} - 2\gamma_2 \cos \varphi \sin \varphi \frac{\partial \varphi}{\partial t} \frac{\partial \varphi}{\partial x}
\]

\[+(\eta_a \cos^2 \varphi + \eta_b \sin^2 \varphi) \frac{\partial^2 \varphi}{\partial z^2} = 0.
\]

(18)

Here the Miesowicz shear viscosities \( \eta_i \) as well as the viscosity coefficients \( \gamma_i \) are linear combinations of the Leslie coefficients \( \alpha_i \). Numerical simulations on the base of these equations resulted in qualitative agreement with the experimental findings of growing VRLs. Measurements on polymer solutions, however, where the amount of backflow may be controlled by the strongly concentration dependent shear viscosities, should be a way to prove this theory.

4. Results and Discussion

Values of the elastic constants of 5CB and the polymer solutions are collected in Table I. It is clearly seen that the polymer does not influence the elastic properties of the dilute solutions. The values of the material parameters are in a good accordance with literature [25]. The twist viscosities \( \gamma_1 \), given in the last column of Table I, were determined from the \( \omega \) dependence of the field \( B_a \) for the transition from the synchronous to the asynchronous regime using equation (8).

In Figure 3 experimentally determined state diagrams for solutions with different polymer concentration are shown. For better comparison, the abscissa is chosen to be \( \gamma_1 \omega \), resulting in synchronous–asynchronous transition lines equal for all solutions. Even the small amount of \( w = 0.52\% \) polymer leads to a reduced existence range of the VRL–TI state (Fig. 3a), caused by transitions between VRL and VRL–TI occurring at higher field values than for pure 5CB. At a concentration of \( w = 2.12\% \) (Fig. 3b) the VRL–TI state and the complex state were completely suppressed in the observable part of the state diagram, whereas the VRL state still was observed. We did not include transition lines between the VRL state and uniform rotation.
Fig. 3. — Experimental state diagrams for solutions containing different weight fractions \( w \) of PMC-312 at \( T = 23 \, ^\circ\text{C} \). a) open symbols: pure 5CB, filled symbols: \( w = 0.52\% \). b) \( w = 2.12\% \); for this concentration the transitions between static and dynamic solitons were not determined.

because a determination of the transitions in the sense of points of zero growth rate of the VRLs was not possible. As will be discussed further in Section 4.2, it is not clear whether the growth rate (reduced in the case of polymer solutions) reaches zero at finite values of \( \omega \tau \) or not.
4.1. Dynamic Solitons. — For polymer solutions backflow effects are found to be strongly reduced [4, 12, 13]. Thus, direct comparison of the pure nematic compound 5CB and polymer solutions may provide us with the understanding of role of the backflow in the soliton dynamics. A second source of discrepancy with theory may be caused by the one-constant approximation. In fact, the soliton accumulates two elastic deformations, splay and bend. From numerical simulations one can show that taking into account both deformations results in an effective value of $K$ given by the arithmetic average of the square roots of $K_1$ and $K_3$, $K^{1/2} = (K_1^{1/2} + K_3^{1/2})/2$, when the values of $K_1$ and $K_3$ are not too different. Values of $K$ averaged in this way were used in our calculations.

For soliton rings nucleated by dust particles, equation (11), which concerns solitons moving in the $x$ direction with a constant velocity $v$, may be used when the radius $R$ of the ring is much higher than the magnetic coherence length $\xi$. Therefore high values of the field were used in the experiment to neglect the curvature of the inversion walls. The results of numerical calculations and experimental measurements of soliton velocities are given in Figure 4. In fact, the observed solitons for $\omega \tau > 1$ always were elements of soliton lattices rather than single solitons. These lattices consist of concentric rings formed by inversion walls separated by the lattice period $\lambda$. The observed finite lattice periods are shown in Figure 5 for the corresponding measurements of Figure 4. In the regime of asynchronous rotation the solitons seem to prefer similar values of the reduced velocities and lattice periods for all samples at a given value of $\omega \tau$. In the regime of synchronous rotation the lattice period depends strongly on the "activity" of the nucleating dust particle and is difficult to control. Here the coexistence of ring systems with different lattice periods in the same sample is frequently observed. However, for values of $\omega \tau$ well below unity the observed finite lattice periods seem to have no significant effect on the soliton velocity. It is clearly seen that for low values of $\omega \tau$ (down to $\omega \tau \approx 0.6$, where the transition to static solitons occurs) experimental data for pure 5CB and the polymer solutions are in good agreement with the theory for isolated solitons. One can conclude that in this region backflow plays nearly no role in soliton dynamics. A discrepancy occurs near $\omega \tau = 1$ where a critical divergence of the slope of $v/v_0(\omega \tau)$ is expected from the theory. Instead of critical behavior the values of $v/v_0$ for the observed soliton lattices show a moderate increase with $\omega \tau$. 

Fig. 4. — Dependence of the reduced soliton velocity $v/v_0$ on $\omega \tau$ for solutions containing different weight fractions of PMC-312. Solid line: theory for single solitons.
Fig. 5. — Observed values of the reduced lattice period $\lambda/\xi$ in soliton lattices for solutions containing different weight fractions of PMC-312, corresponding to the velocity measurements shown in Figure 4. Points where $\lambda$ was very high or infinite (isolated solitons) are not shown here.

However, in the vicinity of $\omega \tau = 1$, where the transition from synchronous to asynchronous regime is observed, we found similar values of $v/v_0$ for the polymer solutions and for pure 5CB. Therefore the discrepancy in this region cannot be explained by backflow which is suppressed with increasing polymer concentration.

The most surprising fact is that the system of soliton rings is stable even for $\omega \tau > 1$. Figure 6 shows examples of soliton lattices observed in the regime of asynchronous rotation. For the case of pure 5CB in the complex state after some time the lattices nucleate and coexist with the complex pattern (Fig. 6a). The ring pattern propagates into the complex state and finally displaces the complex state completely. Similar observations were made by Migler and Meyer [6]. However, the stability of the ring pattern for $\omega \tau > 1$ cannot be an effect of backflow, because they also are observed in the polymer solutions. At a polymer concentration of $w = 2.06\%$ the complex and VRL-TI states are completely suppressed and the soliton lattices compete directly with the VRL state (Fig. 6b). Annihilation of inversion walls occurs when at the boundary of the soliton lattice a growing ring comes into contact with a shrinking ring of the VRL.

To explain the stability of a system of soliton rings in the asynchronous regime it is necessary to discuss the dynamics of non–isolated solitons. When the nucleation rate is high and the distance between the solitons is not too large, the solitons may be affected by each other. Though solitons are defined to be solitary waves, in case of small distances they are no longer “solitary waves” but may form “soliton lattices”.

This interaction between the solitons may be taken into account by using appropriate boundary conditions when solving the driven overdamped sine–Gordon equation. For instance, periodic boundary conditions may be used to illustrate what happens when the soliton lattice period $\lambda$ decreases from infinity to a finite value $\lambda = 20 \xi$. Figure 7 shows dependencies of $v/v_0$ on $\omega \tau$ calculated by numerical integration of equation (11) using a forward time centered space (FTCS) finite difference method [26] with periodic boundary conditions and different values of $\lambda/\xi$. The dependence for $\lambda/\xi = 100$ in the range $\omega \tau < 1$ is similar to that obtained for isolated solitons ($\lambda/\xi = \infty$). For soliton lattices with periods of $\lambda/\xi = 40$ and $\lambda/\xi = 20$ the velocity shows a rather continuous behavior at the transition to the asynchronous regime. Furthermore, a soliton lattice is stable at $\omega \tau > 1$ as it was observed in the experiment. The propagation in
Fig. 6. — Soliton lattices in the regime of asynchronous rotation. a) pure 5CB, sample thickness $d = 100 \ \mu m$, $T = 23^\circ C$, $B = 0.57 \ T$, and $\omega = 2.20 \ s^{-1}$. Soliton lattices grow into the complex structure. b) $\omega = 2.06 \%$, sample thickness $d = 100 \ \mu m$, $T = 23^\circ C$, $B = 0.60 \ T$, and $\omega = 1.34 \ s^{-1}$. Soliton lattices consisting of growing rings (short lattice period) grow into the VRL pattern consisting of shrinking rings.

Fig. 7. — Dependence of the reduced velocity $v/v_0$ in soliton lattices on $\omega \tau$ in the vicinity of $\omega \tau = 1$ for different values of the reduced lattice period $\lambda/\xi$ and for a single soliton ($\lambda/\xi = \infty$), calculated numerically by solving equation (11). Broken lines: asymptotes given by equation (19).

Soliton lattices was studied previously by Burkov et al. [27]. The authors also found analytically an asymptote at large $\lambda/\xi$ and $\omega \tau > 1$, shown in Figure 7 by broken lines:

$$\frac{v}{v_0} = \frac{\lambda}{2\pi\xi}[(\omega \tau)^2 - 1]^{1/2}$$

(19)

For the case of ferroelectric smectic C* layers in a rotating electric field Kilian et al. [11] resulted in finite velocities even for single solitons at $\omega \tau > 1$. This seems due to their assumption, that the shape of the solitons can be approximated by the kink solution of the corresponding
sine–Gordon equation (which differs from the overdamped sine–Gordon equation by a second time derivative instead of the first time derivative). This solution, however, is equal to the corresponding solution of equation (12) only in the static limit \( v \to 0 \) and therefore leads to wrong results for \( \omega \tau \geq 1 \). We conclude that there exist soliton solutions of equation (11) for \( \omega \tau > 1 \) only when the solitons are not isolated.

The dependencies presented in Figure 7 were calculated by solving equation (11) for fixed lattice periods \( \lambda \) not depending on \( \omega \tau \). However, the experimentally observed values of \( \lambda \) (Fig. 5) are not constant. In order to compare the experimental values of \( v/v_0 \) with calculated values one has to use the resulting experimental values of \( \lambda \) in the calculations. Figure 8 shows values of \( v/v_0 \) calculated in this way together with experimental values. For pure 5CB there is a significant deviation at \( \omega \tau > 1 \). This can be explained by the backflow effect, which was not accounted for in the calculations. The reduction of the effective rotational viscosity caused by backflow leads to a faster director rotation, i.e. a slower increase of the phase lag. For a given lattice period therefore the backflow causes a lower soliton velocity. For solutions with higher polymer concentration the suppression of backflow therefore explains the better agreement between measured and calculated velocities. It should be mentioned that the effect of backflow on the soliton velocity is not seen directly in the experimental values (Fig. 4). The larger differences of the calculated velocities for different polymer concentrations (Fig. 8) originate from slightly different values of the (short) lattice periods (Fig. 5), as in the asynchronous regime the effect of \( \lambda \) on the velocity can be very strong (see Fig. 7). Therefore the effect of backflow is better discussed in terms of the phase lag increase.

A measure for the phase lag increase in a lattice is the soliton current \( J = v/\lambda \) given by the frequency of inversion walls passing a fixed point. In a steady state \( J \) has to be equal to the rate of solitons nucleated at the location of the phase lag source. Burkov et al. [27] resulted in two asymptotes for the soliton current: an upper limit \( J_{\text{max}} = \omega/\pi \) as an asymptote for short lattice periods \( \lambda \), corresponding to a situation where the director rotation is zero at the location of soliton nucleation, and a lower limit \( J_{\text{min}} = 1/T_h \) (corresponding to Eq. (19)) as an asymptote for \( \omega \tau > 1 \) and large values of \( \lambda \), equal to the inverse period of the phase lag increase in the state of homogeneous asynchronous rotation given by equation (9).
Fig. 9. — Dependence of the reduced soliton current $J_r = J \cdot \xi / \nu_0$ in soliton lattices on $\omega \tau$ for different values of the reduced lattice period $\lambda / \xi$, calculated numerically by solving equation (11). (- - -) $J_{r \ min}$; (--- - -) $J_{r \ max}$ (see text).

shows the dependence of the reduced soliton current $J_r = J \cdot \xi / \nu_0$ on $\omega \tau$, calculated on the base of equation (11), where backflow is not accounted for. Included are the two asymptotes. For any finite value of $\lambda$ the rate of soliton nucleation is higher than the rate of $\pi$-steps $J_{min}$ in the state of homogeneous asynchronous rotation. Therefore such lattices should grow when they compete with the state of uniform rotation. In the case of competition with the non-uniform asynchronous states (complex, VRL and VRL-TI) assumed to be connected with the viscosity reduction mechanism, one would have to compare the average phase lag increase rate with that of these states. In contrast to the lattices of nucleated dynamic solitons the soliton current of these states should always be below $J_{min}$, due to their reduced effective rotational viscosity. Of course, backflow, when not suppressed by a sufficient amount of polymer, also affects the soliton current of the soliton lattices. This can be seen from Figure 10, where the experimental values of $J_r$ (corresponding to Figs. 4 and 5) are compared with those calculated by solving equation (11). In contrast to the calculated values, especially for pure 5CB experimental values even below $J_{r \ min}$ are observed in the asynchronous regime. However, for growing soliton lattices the values of $J < J_{min}$ need to be still higher than that of the competing pattern forming state. For 5CB (in the range $1.0 < \omega \tau < 1.3$) the lattices competed with the complex state, which therefore seems to produce an even lower phase lag increase.

We can conclude this section as follows: Dynamic solitons are also observed in the asynchronous regime, where they always form soliton lattices. The simple, backflow neglecting theory (Eq. (11)) is utilized for comparison with the experimental soliton velocities $v$. Soliton lattice solutions of equation (11) do not predict the lattice period $\lambda$, however a relationship between $\lambda$ and $v$ is predicted. We experimentally find that this relationship holds in the case of finite polymer concentration, however it does not hold in the case of pure 5CB in the asynchronous regime. From this, we conclude that backflow is significantly reduced in the case of finite polymer concentration. Due to the simple relation between $v$, $\lambda$ and the soliton current $J$, the findings can also be discussed in terms of $J$. For pure 5CB the experimental values of $J$ in the asynchronous regime were significantly lower than those predicted by the simple theory, as expected for an effective rotational viscosity reduced by backflow.
4.2. Viscosity Reduction Lattice. — Two of the three pattern forming states exclusively found in the regime of asynchronous rotation, the VRL–TI and the complex state, were found to be completely suppressed in a solution containing only 2.12% of the polymer. Their existence therefore clearly requires a certain amount of backflow. We now concentrate on the dynamics of the third state assumed to base on a viscosity reduction mechanism.

At a first sight, the viscosity reduction lattice seems to be a quite different phenomenon in comparison to the dynamic solitons: It consists of shrinking rings, and, after initiation, it does not need any sink or source of phase lag. However, a system of shrinking rings can also be obtained for some time from a soliton lattice of growing rings created by nucleation, after the field rotation is reversed suddenly [7]. The similarity also shows up in the propagation velocity or the current of the inversion walls respectively. In Figure 11 the reduced soliton current \( J_r \) in viscosity reduction lattices of pure 5CB and a polymer solution is shown in comparison with the reduced rate of \( \pi \)-steps \( J_{r, \text{min}} = 1/T_h \cdot \xi/v_0 \) in the state of homogeneous rotation. The reduced soliton currents in the VRLs are close to \( J_{r, \text{min}} \), but, in contrast to the soliton lattices formed by continuous nucleation, always below the limiting curve \( J_{r, \text{min}}(\omega \tau) \). It is this small difference in the rates which makes the VRL grow without a source of phase lag: The formation rate of new rings \( J_{\text{min}} \) at the boundary is higher than the rate \( J \) of their transport to the center, therefore an increasing number of inversion walls is accumulated in the lattice. The difference of the rates given as a relative growth rate

\[
\frac{J_{\text{min}} - J}{J_{\text{min}}} = 1 - \frac{J}{J_{\text{min}}} = 1 - T_h J,
\]

shown in Figure 12 for pure 5CB and a polymer solution. A common feature of both samples is a strong decrease of the growth rate with increasing values of \( \omega \tau \). Experimentally it is difficult to decide whether the growth rate reaches zero at a finite \( \omega \tau \) or not. By simulations Migler and Meyer resulted in such values [6]. Different for the two samples are the magnitudes of the growth rates: The amount of only 2.12% of polymer in the solution leads to a strong reduction of the growth rates especially at low values of \( \omega \tau \). As the (minor) effects of the polymer on the rotational viscosity \( \gamma_1 \) and the elastic constants cancel out in the reduced representation
Fig. 11. — Dependence of the reduced soliton current \( J_r \) in viscosity reduction lattices on \( \omega \tau \) for pure 5CB and a polymer solution with a weight fraction \( w = 2.12\% \) of PMC-312. Solid line: \( J_r \text{min} \).

Fig. 12. — Dependence of the relative growth rate \( 1 - T_h \cdot J \) of viscosity reduction lattices on \( \omega \tau \) for pure 5CB and a polymer solution with a weight fraction \( w = 2.12\% \) of PMC-312. Filled circles: values simulated with \( d/\xi = 50 \) and viscosity coefficients of 5CB (see text).

of the data, this should be an effect of the reduced backflow in the polymer solution due to a strong increase of the Miesowicz shear viscosities. Therefore this result strongly supports the model of a viscosity reduction mechanism.

Precise measurements of the lattice periods of the VRLs are difficult to perform. Frequently a slight gradient of the ring distance within the lattice is observed. However, the presence of dissolved polymer seem to have no significant effect on \( \lambda \). In Figure 13 the results are shown for VRLs in pure 5CB and in a solution containing 2.06\% of the polymer. In the reduced representation \( (\lambda/\xi) \) similar values were obtained for two different values of \( B \), as is expected for a characteristic length given by the field dependent magnetic coherence length \( \xi \). In the measurements also no significant effect of \( \omega \tau \) on the lattice period was found.

Again, numerical simulations can be used to illustrate the pattern forming process. For pure 5CB the complete set of viscosity coefficients are known from the literature. In this case
therefore a quantitative comparison with the experiment should be possible. For the polymer solutions, where we lack quantitative information about $\eta_h$ and $\eta_c$, we restrict ourselves to a qualitative comparison with the case of completely suppressed backflow.

In the equations of motion (17) and (18) the terms containing the flow velocity $v_f$ or its derivatives are depending on the $z$ coordinate. To avoid the difficulty of solving a problem depending on one time and two spatial coordinates, the backflow contribution to the torque in equation (17) proportional to the gradient $\partial_z v_f$ is approximated by its arithmetic average along the $z$ axis. The force balance equation (18) then is solved in the middle plane of the cell at $z = 0$ and the resulting velocity gradient in this plane $\partial_z v_0$ is used to calculate the average of the gradient:

$$\frac{\partial v_f}{\partial x} = \frac{2}{3} \frac{\partial v_0}{\partial x}. \quad (21)$$

Because of the coupling between director rotation and fluid flow the equations have to be solved simultaneously. Therefore at each time step of the FTCS scheme [26] the equations were iterated [6]. The boundary conditions $v_{\text{f}}(0, t) = v_{\text{f}}(x_{\text{max}}, t) = 0$ and $\partial_x \alpha(0, t) = \partial_x \alpha(x_{\text{max}}, t) = 0$ were applied, corresponding to a superstructure of patterns with a period of $2x_{\text{max}}$. For the elastic constants again a one constant approximation $K = K_1 = K_3$ was used. Results are shown in Figure 14 in comparison with the case of completely suppressed backflow. In the latter case the infinitely high Miesowicz coefficients $\eta$ cause a flow velocity of zero, and the torque balance therefore reduces to equation (10) or (11) respectively. An initially given lattice (in coexistence with the state of uniform rotation) then does not show any growth (Fig. 14a). In fact its growth rate is even negative because the soliton current then is larger than $J_{\text{min}}$ for any finite $\lambda$. When the equations are solved using the viscosity coefficients of 5CB [28], an initially given director field deformation leads to the development of a growing viscosity lattice (Fig. 14b). As already reported by Migler and Meyer, in this case motion and shape of the inversion walls are not very regular, in agreement with the experiment.

An important finite size effect results from the finite layer thickness $d$, which leads to a non–vanishing term in equation (19) proportional to the second derivative $\partial_{zz} v_f$ of the flow
velocity. Assuming the parabolic \( z \) dependence of \( v_f \) given in equation (17) one obtains

\[
\frac{\partial^2 v_f}{\partial z^2} = -\frac{8}{d^2} v_{f0}(x,t).\]

The existence of this term is necessary for formation and growth of the VRL. Here the layer thickness \( d \) appears as a second characteristic length scale of the phenomenon, in addition to the magnetic coherence length \( \xi \). In a double logarithmic plot Figure 15 gives the dependence of the average lattice period \( \lambda \) on \( d \) in VRLs simulated with the viscosity parameters of 5CB. For both values of \( \omega T \) shown in the figure one results in nearly linear dependencies in the range \( 100 \leq d/\xi \leq 500 \). The slopes of the straight lines fitted to these regions are 0.83 and 0.90, i.e. close to one. At lower values of \( d/\xi \) a condition \( d \gg \xi \) does not hold and \( \xi \) as the other characteristic length scale becomes active as a limiting influence on the lattice period. At even lower values (\( d/\xi \approx 20 \)), in contrast to the experiment, instead of a complete lattice only a pair of inversion walls is formed. The values of \( \lambda/\xi \) for \( d/\xi = 20 \) in the figure are given by the average distance of the two walls.

The deviation from the straight lines at high values of \( d/\xi \) is probably caused by the finite size \( x_{\max} \) of the simulation grid, which is supposed to act as a third characteristic length scale when the size of the patterns is not longer small compared to it. In the simulations grid sizes up to \( x_{\max} = 2000 \xi \) were used.

A similarity to the linear layer thickness dependence of the periods in Rayleigh–Bénard patterns is obvious. However, in that case the flow velocity in the convection rolls essentially has no in–plane component, whereas in the viscosity reduction lattice the flow is assumed to be in the layer plane.

The experimentally observed lattice periods shown in Figure 13 are of the same order of magnitude as to be expected from Figure 15 (\( \log_{10}(d/\xi) \approx 1.3 \)). The values of \( d/\xi \) realized
Fig. 15. — Dependence of the average lattice period $\lambda$ on the layer thickness $d$ in simulations of viscosity reduction lattices for two different values of $\omega \tau$. The one constant approximation $K = K_1 = K_3$ and the viscosity coefficients of 5CB were used.

experimentally in the measurements of Figure 12 unfortunately are too low to result in complete lattices in the corresponding simulations. However, when complete lattices were formed in the simulations for larger values of $d/\xi$, their growth rate was affected only slightly by $d/\xi$. We therefore used simulations with $d/\xi = 50$ for a comparison with the experiment. The resulting growth rates, included in Figure 12, are in good accordance. Additional measurements at higher values of $d/\xi$ are planned, despite of the difficulty to achieve unperturbed homeotropic initial orientations in case of large cell thicknesses.

5. Conclusions

The dynamics of patterns in nematic liquid crystals exposed to a rotating magnetic field principally can be affected by backflow. The use of dilute solutions of a liquid crystalline side group polymer in a low molecular nematic solvent allows to control the suppression of backflow effects by the polymer concentration.

In the regime of synchronous rotation no significant backflow effect on the propagation of the inversion walls was found. Neglecting backflow, their equation of motion can be well-approximated by an overdamped sine–Gordon equation. The propagating inversion walls correspond to soliton solutions of this equation. These dynamic solitons can also be observed in the asynchronous regime, when they form soliton lattices consisting of inversion walls equally spaced by the lattice period $\lambda$. Whereas isolated soliton solutions of the overdamped sine–Gordon equation do not exist in this regime, soliton lattice solutions are found, resulting in finite propagation velocities $v$ for any finite value of $\lambda$. Especially for the case of solutions with relative high concentration of the polymer, the experimentally observed soliton currents agree well with calculated ones, obtained by numerical solutions of the overdamped sine–Gordon equation using the observed values of $\lambda$. With decreasing polymer concentration the increasing backflow leads to lower values of the soliton current. This can be understood by regarding the result of backflow as the reduction of an effective rotational viscosity, leading to a faster director rotation in the average. Faster rotation means lower increase of the phase lag $\alpha$ between director and magnetic field $B$ and therefore lower values of the soliton current $J = v/\lambda$, which is a measure of the local phase lag increase within a soliton lattice. In the experiment, the
reduction of the soliton current caused by backflow was found to be realized by a slight change of the lattices period $\lambda$ rather than by reduced propagation velocities.

The dynamic soliton lattices are patterns with increased phase lag with respect to the bulk regions of homogeneous director rotation. They need a continuous source of phase lag, experimentally given by the local hindrance of rotation near to dust particles or side walls. In contrast to this, in the regime of asynchronous rotation also three pattern forming states are observed, where the phase lag inside of the patterns is lower than that in the corresponding case of homogeneous rotation. The spontaneously formed complex state and the VRL–TI state are found to be completely suppressed in solutions with sufficient polymer concentration. This supports their explanation by the concept of viscosity reduction caused by backflow. The viscosity reduction lattice as the third state was found to be affected by the presence of polymer in terms of the growth rate of its patterns. Increasing polymer concentration leads to lower values of the growth rate. This again can be understood as a result of backflow suppression: For completely suppressed viscosity reduction the pattern itself does no longer act as a continuous sink of phase lag which otherwise leads to growing areas of accumulated phase lag difference with respect to the bulk.

Simulations on the base of the equations of motion with and without taking account of backflow can be used to confirm this picture of the viscosity reduction lattices: A growth of an initially given lattice is obtained only when backflow is accounted for. The values of growth rates and lattice periods obtained with the viscosity parameters of 5CB are in good accordance with the experiment. The simulations also show up the importance of the finite layer thickness $d$ for the formation of the viscosity reduction lattice. In an intermediate range the lattice period $\lambda$ obtained from simulated nucleation of lattices scales with the thickness $d$. In the limit $d \rightarrow \infty$ no nucleation was observed.

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