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# DIRECTOR FIELD PATTERNS OF NEMATIC POLYMER SOLUTIONS AT ASYNCHRONOUS ROTATION IN A MAGNETIC FIELD

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Abstract A variety of different patterns is observed when a magnetic field rotating with an angular frequency  $\omega$  is applied on a nematic cell with homeotropic boundary conditions. To study the role of backflow in the formation of these dissipative structures, we compared the behaviour of dilute solutions of a mesogenic side group polymethacrylate in a low molar mass nematic liquid crystal (5CB) with that of the pure solvent. The dissolved polymer molecules lead to a strong reduction of the existence range of the so-called VRL-TI (viscosity reduction lattice transverse instability) and complex states. For mass fractions w > 2% of the polymer these states were found to be suppressed completely due to the efficient reduction of backflow. The growth rate of the viscosity reduction lattices, which can still be observed, is in addition dependent on the polymer concentration. Numerical calculations were performed to explain the behaviour.

#### INTRODUCTION

The formation of director field patterns of a nematic layer in a rotating magnetic field **B** was studied in detail by Migler and Meyer. <sup>1,2</sup> Viscosity reduction mechanisms based on the presence of backflow were proposed to explain some of the structures. In this paper we investigate the role of backflow for the formation of these dissipative structures by comparing the behaviour of dilute solutions of a mesogenic side group polymethacry-late<sup>3</sup> (PMC-312, degree of polymerisation N = 312) in a low molar mass nematic liquid crystal (5CB) with that of the pure solvent.

PMC-312 
$$CH_3 - C - COO - (CH_2)_4 - N = N - OCH$$

5CB  $CH_3 - (CH_2)_4 - CN$ 

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In a previous study<sup>4</sup> we observed a strong reduction of backflow in these polymer solutions due to a large increase of shear viscosity coefficients compared to that of the rotational viscosity  $\gamma_1$ , as expected from a theory of Brochard<sup>5</sup> when the shape of the polymer coils is nearly spherical.

A characteristic time scale of the system is given by the field dependent time constant  $\tau = 2\gamma_1\mu_0/(B^2\Delta\chi)$ . Here  $\Delta\chi$  denotes the anisotropy of the diamagnetic susceptibility. Depending on B and its angular frequency  $\omega$  two different regimes of the director rotation are obtained. In the synchronous regime, where  $\omega\tau < 1$ , the phase lag angle  $\alpha$  between B and the nematic director n is constant. In this regime one can observe propagating inversion walls, which are nucleated either by dust particles or by outer side walls. When nucleated by dust particles, the inversion walls form growing rings. These propagating walls were denoted by Migler and Meyer as dynamic solitons. Soliton lattices of equidistant walls are formed when nucleation occurs repeatedly.

In the asynchronous regime at  $\omega \tau > 1$ , where  $\alpha$  increases continuously with time, the director rotation is even more complex. In addition to a state of spatial uniform rotation, different pattern forming states supposed to be caused by the coupling of director rotation to fluid flow are found. With decreasing  $\omega \tau$  a loss of symmetry is observed, in this order the structures are denoted as viscosity reduction lattice (VRL), viscosity reduction lattice transverse instability (VRL-TI), and complex state.

### **EXPERIMENTAL**

The range of the polymer mass fraction w in the solutions was  $0 \le w \le 2.12$  %. To obtain homeotropic surface orientation we used glass plates coated with lecithin (from 0.5 % solution in ethyl alcohol). Mylar spacers kept the layer thickness constant to  $d = 100 \, \mu \text{m}$ . The measurements were performed at a temperature  $T = 23.0 \pm 0.1 \,^{\circ}\text{C}$ .

The patterns were observed and recorded using an optical scheme including a microscope objective, crossed polarizers oriented at angles of  $\pm 45^{\circ}$  to the magnetic field, a charge coupled device (CCD) video camera, and a video recorder. The video signal was interfaced to a personal computer by a video digitizer (Screen Machine II, FAST Electronic).

The step rate of a stepper motor combined with a gear reduction controlled the angular frequency of the cell rotating in the static field of an electromagnet. This is equivalent to the rotation of the field.

The same setup was used to determine the splay and bend constants by observing the field dependent response of the director in a bend Frederiks geometry.<sup>6</sup> For this purpose the continuous white light source was replaced by a He-Ne laser. The elastic

constants were found not to be affected by the presence of polymer in these dilute solutions.

#### RESULTS AND DISCUSSION

In Figure 1 the observed patterns are shown for pure 5CB as an example. With increasing concentration of the polymer the existence range of the VRL-TI and the complex state is strongly reduced. For mass fractions w > 2% of the polymer these states were found to be suppressed completely due to the efficient reduction of backflow. The VRL state, however, is still observed as a system with an increasing number of shrinking rings.

Dynamic solitons are not only found in the synchronous regime but also at asynchronous rotation, when repeated nucleation leads to the formation of soliton lattices instead of isolated solitons.<sup>6</sup> At synchronous rotation : equation of motion can be written in good approximation as an overdamped sine-Gordon equation:<sup>1,7</sup>

$$K\alpha_{xx} + \gamma_1(\omega - \alpha_x) = B^2 \Delta \chi / (2\mu_0) \sin(2\alpha)$$
 (1)

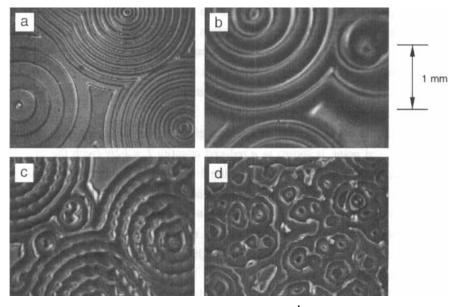
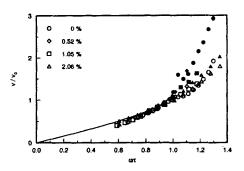


FIGURE 1 Structures of a 5CB sample at  $\omega = 2.2 \text{ s}^{-1}$ . (a) Dynamic solitons at B = 0.63 T, (b) VRL at B = 0.43 T, (c) VRL-TI at B = 0.52 T, (d) complex structure at B = 0.52 T.



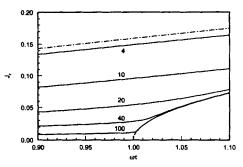


FIGURE 2 Reduced velocity  $v/v_0$  in soliton lattices  $(v_0 = (K\Delta\chi/\mu_0)^{0.5} B/\gamma_1)$  for several polymer solutions (open symbols) and values calculated by simulations (filled symbols). Solid line: theory for isolated solitons.

FIGURE 3 Dependence of the reduced soliton current  $J_r$  in soliton lattices on  $\omega \tau$  for different values of the reduced lattice period  $\lambda \xi$ .

Here  $\alpha_{xx}$  and  $\alpha_t$  denote the second spatial and first time derivative of the phase lag  $\alpha(x,t)$ , for the elastic constants a one-constant approximation  $K=K_1=K_3$  is used. This equation can also be used to explain the soliton lattices in the asynchronous regime, when it is solved with periodic boundary conditions given by the lattice period  $\lambda$  (the radial distance of the rings). In Figure 2 the propagation velocities  $\nu$  of the solitons obtained by numerical solutions of Equation (1) using the observed values of  $\lambda$  are compared with the experimental values. For pure 5CB there is a significant deviation at  $\alpha \tau > 1$  due to the backflow effect, which was not accounted for in the equation of motion. For solutions with higher polymer concentration the suppression of backflow leads to a better agreement.

The soliton current  $J = \nu/\lambda$  in a lattice, given by the frequency of inversion walls passing a fixed point, is shown as a reduced quantity  $J_r = J \cdot \xi/\nu$  (with the magnetic coherence length  $\xi = (\mu_0 K/\Delta \chi)^{0.5}/B$  in Figure 3 as resulting from numerical calculations. Included is an asymptotic  $J_{\text{max}}$  for short lattice periods. For  $\omega \tau > 1$  there is a second limiting asymptotic  $J_{\text{min}} = 1/T_{\text{h}}$ , equal to the inverse period of the phase lag increase in the state of uniform asynchronous rotation. A nucleation site producing a soliton lattice at  $\omega \tau > 1$  acts as a phase lag source with a higher rate than that of the phase lag increase of the competing state.

At a first sight, the viscosity reduction lattice seems to be a quite different phenomenon: 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 created by nucleation, when after development of the lattice

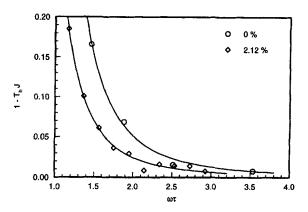
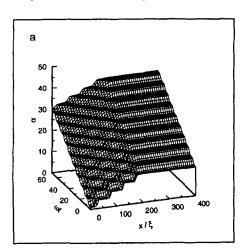


FIGURE 4 Dependence of the relative growth rate of viscosity reduction lattices  $1 - T_h J$  on  $\omega \tau$  for pure 5CB and a polymer solution containing 2.12 % PMC-312.

the rotation sense of the field is changed suddenly. The similarity also shows up in the propagation velocities of the inversion walls. In Figure 4 the soliton current of viscosity reduction lattices is shown as a relative difference to the phase lag increase rate in the uniform state, giving a relative growth rate of the lattice. At high values of  $\omega \tau$  this growth rate is small. At low values of  $\omega \tau$  the growth rate is reduced drastically in case of the polymer solution.

This verifies the theory of Migler and Meyer,<sup>2</sup> which explains the growth of viscosity reduction lattices by backflow, resulting in a reduced effective rotational viscosity



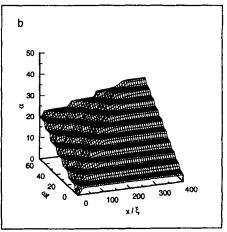


FIGURE 5 Simulation of the phase lag  $\alpha$  depending on  $\omega t$  and the reduced spatial coordinate  $x/\xi$  for  $\omega \tau = 1.1$ . (a) Behaviour of an initially given lattice without taking account of backflow, (b) development of a viscosity reduction lattice from an initial gaussian director field deformation (with reduced thickness  $d/\xi = 50$  and viscosity coefficients of 5CB).

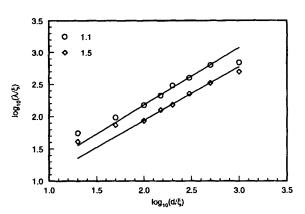


FIGURE 6 Dependence of the average lattice period  $\lambda$  on the layer thickness d in simulations of VRLs for two different values of  $\omega \tau$ . The one constant approximation  $K = K_1 = K_3$  and the viscosity coefficients of 5CB were used.

within the lattice. Numerical simulations on the base of this theory indeed show the same behaviour: Without backflow an initially given lattice does not grow (Figure 5a), because under these simulation conditions (infinitely high Miesowicz shear viscosities) the equations of motion reduce to the overdamped sine-Gordon equation. Then the soliton current is higher compared to the phase lag increase in the uniform state (see Figure 4). When (with Miesowicz coefficients for 5CB) backflow is accounted for (Figure 5b), the coupling between director rotation and fluid flow results in a higher rotation rate of the director in presence of director field gradients. The soliton current is lower than the creation rate of new inversion walls, the lattice itself acts as a phase lag sink.

When backflow is accounted for, the layer thickness d plays an important role for a finite size effect due to zero flow at the boundaries.<sup>2</sup> Simulations (see Figure 6) show that then in an intermediate range the lattice period  $\lambda$  of VRLs scales with d, when a parabolic flow velocity profile is assumed along the layer normal.<sup>6</sup>

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