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## Liquid Crystals

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# Ultrafast, electric field-induced fingers in an anticlinic liquid crystal

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Propagating fingers of synclinic liquid crystalline phase were observed to invade the anticlinic phase for applied electric fields  $E$  larger than a characteristic threshold field  $E_{th}$ . The front velocity was found to be highly non-linear in  $E$ , with enormous velocities of at least  $10 \text{ cm s}^{-1}$ , and perhaps as high as  $400 \text{ cm s}^{-1}$  for the maximum applied field. These are by far the largest velocities ever observed for a liquid crystal. The results are discussed theoretically, including the possibilities of a field-dependent molecular interaction coefficient and shear thinning.

## 1. Introduction

The physics of anticlinic liquid crystals is rich and diverse [1, 2]. In both the anticlinic and synclinic phases the director  $\hat{n}$  tilts by polar angle  $\theta$  with respect to the smectic layer normal. In the synclinic phase the azimuthal angle  $\varphi$  is identical in every layer, aside from a spatially slow layer-to-layer rotation due to the helical nature of the chiral molecules. On the other hand, in the chiral anticlinic phase the azimuthal orientation changes by an angle of approximately  $\pi$  from one layer to the next. Materials having an anticlinic phase exhibit tristable behaviour [1], and can switch from anticlinic to synclinic for sufficiently large electric field. This switching, in fact, has been shown to occur via finger-like solitary waves of the synclinic phase invading the anticlinic region [3]. The fingers, of order  $10 \mu\text{m}$  in width, tend to nucleate at a defect, and propagate in a direction parallel to the smectic layers. In [3] we showed that the threshold field  $E_{th}$  for the onset of fingering is approximately independent of cell thickness and that the fingering is reversible by reducing the electric field below  $E_{th}$ . The data showed that the finger velocity  $v$ , which may depend on surface treatment, rises linearly with reduced field  $E_r$  ( $\equiv (E - E_{th})/E$ ) for very small  $E_r$ , but begins to exhibit upward curvature for  $E_r$  as small as 0.02. For a polyimide-treated surface, for example,  $v$  was found to be  $0.2 \text{ cm s}^{-1}$  at  $E_r = 0.03$ . A theory also was presented to explain the results. In [3], however, velocity

measurements were limited by the speed of the video equipment—approximately 30 frames per second—when the polarized microscope image was recorded and later viewed frame-by-frame. We have recently circumvented this problem by using a completely different scheme: the solitary wave is now imaged onto a small slit, and the total intensity of polarized light passing through the slit followed by an analyser is recorded as a function of time. With this new technique we obtained results similar to those in [3] at the same range of (very small) reduced fields reported in that paper. However, at higher fields we now observe that the velocity is extremely nonlinear in  $E_r$ . Moreover, the velocities are *much* larger than before, at least  $10 \text{ cm s}^{-1}$  and perhaps as high as  $400 \text{ cm s}^{-1}$ , which is several orders of magnitude faster than ever observed for a nematic or a ferroelectric liquid crystal.

## 2. Experimental and results

Cells were constructed on electrically conducting indium tin oxide coated glass plates, which were first spin coated with the polyimide CU-2012 (Merck), baked, and rubbed unidirectionally using a dedicated rubbing machine and cotton cloth. The slides were then placed together (with the rubbing directions parallel) separated by a pair of mylar spacers of nominal thickness  $d = 5 \mu\text{m}$ , and cemented. The cells were filled with the liquid crystal TFMHPOBC, 4-(1-trifluoromethylhexyloxy carbonyl)-phenyl 4'-octyloxybiphenyl-4-carboxylate [4], in the isotropic phase, placed into a temperature controlled oven, and allowed to cool through the Smectic A—

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Smectic  $C_A^*$  ( $SmC_A^*$ ) transition temperature  $115.3^\circ\text{C}$ , and into the anticlinic  $SmC_A^*$  phase. Microscope observations revealed a nearly texture-free sample with planar orientation.

The cell was then placed in the optical apparatus shown in figure 1. Light from a 35 mW He-Ne laser passed through a polarizer and was then focused onto the sample using a 7.3 cm focal length lens  $L_1$ . The rubbing direction, corresponding to the smectic layer normal, was oriented parallel to the polarizer along the  $z$ -axis. A second lens  $L_2$  of focal length 6.5 cm was used to form a real image of the liquid crystal sample on a small slit, approximately  $w = 10\ \mu\text{m}$  wide and  $l = 150\ \mu\text{m}$  long. The long axis of the slit was oriented parallel to the smectic layers, and the narrow  $10\ \mu\text{m}$  width ensured that only one, or at most two, fingers would be observed. The slit was placed directly in front of an analyser oriented along the  $x$ -axis, followed by a photodiode detector having a response time  $< 1\ \mu\text{s}$ . The detector output was fed into a fast digitizing storage oscilloscope. In the anticlinic phase the molecules are tilted by a polar angle  $\theta$  with respect to the layer normal (see figure 2) and the azimuthal orientation changes by approximately  $\pi$  from one layer to the next. Thus, the director orientation, spatially-averaged over a pair of layers, is virtually parallel to the  $z$ -axis; the only component in the  $xy$ -plane occurs because of the helical pitch. However, since the pitch is also much shorter than optical wavelengths, incoming light sees an average director along only the  $z$ -axis, parallel to the polarizer. In consequence, in the absence of an electric field, the intensity at the detector was zero. For  $E > E_{th}$  a finger of synclinc phase invading the anticlinic region resulted in a spatially-averaged director making an angle  $\theta$  with the  $z$ -axis in the  $xz$ -plane; the intensity of light passing through the slit for that region was therefore non-zero. Thus, a finger travelling at constant velocity would result in principle

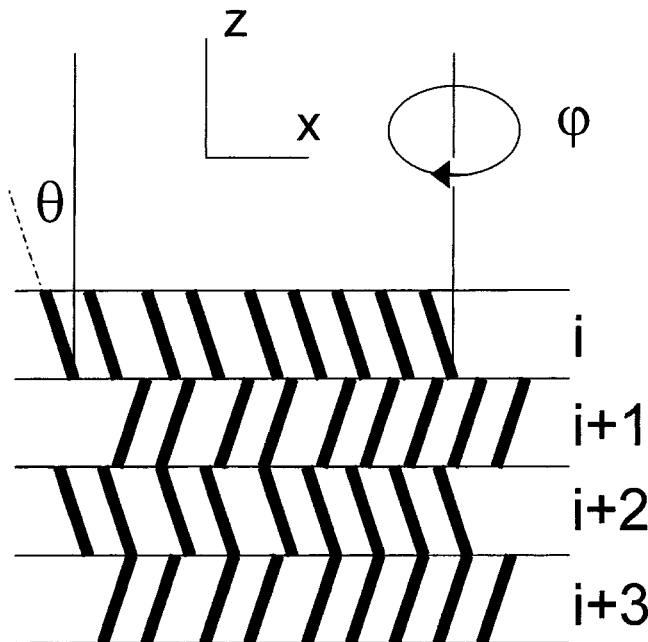


Figure 2. Schematic representation of unwound anticlinic phase.  $\theta$  corresponds to the polar tilt angle,  $\phi$  to the azimuthal orientation, and  $i$  to the layer index.

in an intensity profile rising linearly in time to some maximum value, at which point only the synclinc phase would be imaged onto the entire slit. The velocity  $v$  of the finger would therefore correspond to  $l/\tau$ , where  $\tau$  is the rise time of the detector intensity.

A d.c.-biased square wave voltage of period 0.1 s and having rise and fall times  $< 5\ \mu\text{s}$  was applied to the liquid crystal cell. The bias was adjusted so that  $|E| > E_{th}$  during only the positive half of the cycle; during the negative half  $|E|$  was nearly zero. During both the positive and negative halves of the cycle while  $|E|$  was less than the threshold field  $E_{th}$ , only a small optical response was observed. This was due partially to an electroclinic effect in the  $SmC_A^*$  phase [5] and partially to the interaction of the field with the Goldstone modes [6–9]. The electric field also tended to unwind any remnant helix. When the pulse height was set to be larger than  $E_{th}$ , however, there was a sharp rise to the intensity with time, as observed on the oscilloscope (figure 3). Because the single trace tended to be very noisy, it was necessary to average the response over many thousands of pulses. In order to determine the velocity, the averaged intensity vs. time trace was numerically differentiated (figure 3). For a single, infinitely sharp solitary wave the intensity is expected to be flat until the threshold field is achieved, after which the signal would increase linearly with time until the entire length of the  $150\ \mu\text{m}$  long slit is sampling only a synclinc image. Thus the time derivative of the signal would be a square wave whose amplitude would correspond to the finger velocity.

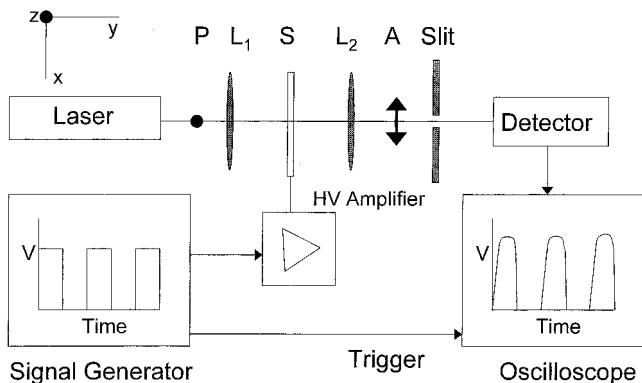


Figure 1. Schematic representation of the experimental arrangement. P and A correspond to polarizer and analyzer, respectively,  $L_1$  and  $L_2$  are focusing and collimation lenses, respectively, and S is the sample cell.

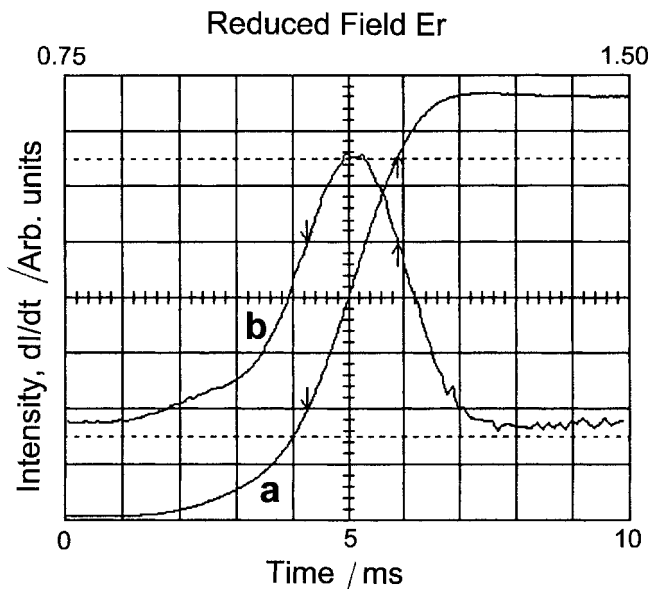


Figure 3. Oscilloscope traces as functions of time (bottom scale) and reduced field  $E_r$  (top scale). (a) Trace of intensity, averaged over 5000 shots; (b) time derivative  $dI/dt$  of trace (a).

In reality, however, there was rounding of the time derivative of the intensity. We believe that this was due to the shot-to-shot variation of the nucleation time of the finger relative to the reference signal from the square wave generator. Smaller contributions to the rounding came from the non-zero rise time of the voltage pulse, and the response time of the detector. In consequence, we associate the maximum slope of the intensity, corresponding to the maximum value of its time derivative, with the velocity. This actually corresponds to a lower limit for the velocity, as the effects discussed above would tend to reduce the apparent velocity.

Figure 4 shows the velocity  $v$  vs.  $E_r$  at three temperatures in the  $\text{SmC}_A^*$  phase. (For this experiment,  $E_{th}$  was found to be  $244 \text{ statv cm}^{-1}$  at  $T = 114^\circ\text{C}$ , with similar values at the two other temperatures.) For  $E_r < 0.05$  (figure 4, inset), which corresponds to the maximum fields used for the polyimide-treated substrate in [3], we find velocities to be in reasonable agreement with those previously obtained [3] using a videographic technique<sup>†</sup>. For larger values of  $E_r$ , figure 4 shows a strong and continued non-linearity of the apparent velocity with  $E_r$ , with  $v$  seemingly rising to values greater than  $400 \text{ cm s}^{-1}$ , or 1000 times larger than the pre-

<sup>†</sup> We note that the precision of this new technique, which was designed to measure large velocities, is not as good as that of the videographic technique. Thus, the uncertainty in velocity as a fraction of the actual velocity is larger than that reported in [3]. Additionally, due to the uncertainty in precisely locating  $E_{th}$ , there is a systematic uncertainty of  $\Delta E_r \sim -0.005/+0.025$  along the abscissa in figure 4.

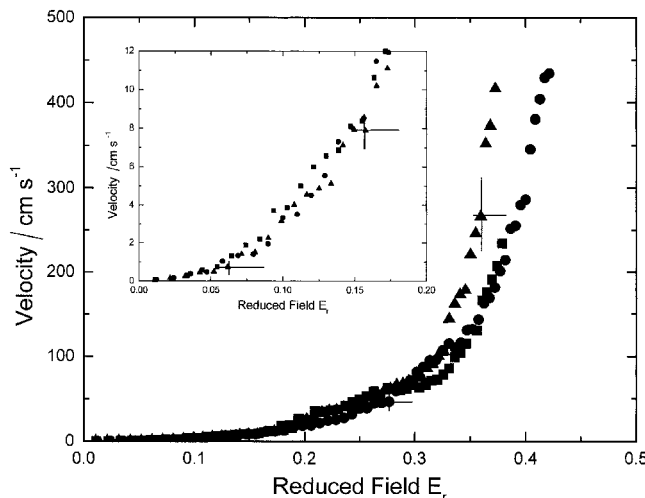


Figure 4. Measurements of velocity vs. reduced field  $E_r$ .  $\blacktriangle$  corresponds to  $T = 112^\circ\text{C}$ ,  $\blacksquare$  to  $113^\circ\text{C}$ , and  $\bullet$  to  $114.0^\circ\text{C}$ . Inset: the same data with expanded scales in the low  $E_r$  regime. Vertical error bars represent experimental uncertainty; horizontal bars represent systematic error ( $\Delta E_r \sim -0.005/+0.025$ ) due to difficulty in precisely locating  $E_{th}$ .

viously measured velocities. We fitted the data to several functional forms, including a single power law and an exponential law; none of these forms resulted in a satisfactory fit.

Before continuing, several caveats need to be mentioned. For the lower range of velocities ( $v < 5$  to  $10 \text{ cm s}^{-1}$ , figure 4 inset), we are clearly able to discern propagating fingers. These velocities are  $1\frac{1}{2}$  orders of magnitude larger than previous measurements [3] and, more importantly, are already highly non-linear in  $E_r$ . These data alone represent the gist of this paper. At higher velocities, however, neither our own visual response nor available imaging equipment is sufficiently fast to discern propagating fingers. Instead, one may imagine that at large  $E_r$  the transformation from the anticlinic to the synclinc phase may be driven by homogeneous nucleation and growth [10–12]. If this were the case, the reported velocities would correspond to the time associated with such a transition, rather than a real finger velocity. Although we cannot completely discount this possibility, we judge this to be improbable: The data do not seem to show any systematic critical field where either the velocity of its derivative with respect to  $E_r$  changes abruptly, a change that would be expected if the transition mechanism were to undergo a sudden modification. We are thus confident that the data shown in figure 4, and especially in the inset, represent true solitary wave behaviour.

A second concern is that we *occasionally* observed an apparent slowing of the solitary wave with time. To see this we employed a delay technique. Taking time  $t = 0$

as the turn-on time of the voltage pulse, we measured the delay time  $t$  required before a non-zero intensity was observed in the slit. If the slit were translated a distance  $x_0$  along the  $x$ -axis away from the nucleation point, the time at which a non-zero intensity at the detector would appear would be  $x_0/v$ , assuming that velocity  $v$  were constant. If, however,  $v$  were to increase or decrease with the position of the finger front, the delay time  $t$  would not be linear in  $x_0$ . An example of the delay time  $t$  vs. slip position from the approximate nucleation site is shown in figure 5. At later times the velocity is clearly slower than at earlier times. The existence of a delay time that is non-linear in  $x_0$  depends on the region of the cell being imaged, and may be due to interactions with defects in the sample. We should emphasize, however, that this behaviour was rare and that *linear* behaviour was obtained for most of our measurements. Thus, over many experimental runs at different sample positions, we obtained good reproducibility in velocity measurements using the intensity vs. time technique described earlier.

### 3. Discussion

Let us now review the theory in [3]. The free energy  $F = \sum_i \int f_i dx$ , where  $f_i = f_i^{\text{elastic}} + f_i^{\text{electric}} + f_i^{\text{layer-layer}}$ , is given by:

$$f_i = \frac{1}{2} K \sin^2 \theta \left( \frac{\partial \varphi_i}{\partial x} \right)^2 - \mathbf{P}_0 \mathbf{E} \cos \varphi_i - \frac{\Delta \varepsilon \sin^2 \theta}{8\pi} \mathbf{E}^2 \sin^2 \varphi_i + \frac{U}{2} [\cos(\varphi_{i-1} - \varphi_i) + \cos(\varphi_i - \varphi_{i+1})]. \quad (1)$$

Here  $f_i$  is the free energy density of the  $i$ th smectic layer;  $K \sin^2 \theta$  is the effective elastic constant associated with a change in azimuthal orientation  $\varphi$  along the  $\hat{x}$ -axis (within a smectic layer, see figure 2);  $\varphi_i = 0$  is defined such that the molecules in layer  $i$  lie in the  $xz$ -plane;  $\mathbf{P}_0$  is the local polarization; and  $\Delta \varepsilon$  is the dielectric anisotropy. The coupling between layers is expressed in terms of the coupling coefficient  $U$ , which has dimensions of energy per volume and represents both entropy and a coarsening average of local interactions between layers involving molecular dipoles and steric effects.  $U$  plays the role of the spatially-uniform free energy of the system, where it is reasonable to assume that the polarization component of  $U$  scales as  $\mathbf{P}_0^2$  [13]. The resulting solitary wave velocity  $v$  is given by [14, 15]

$$v = \left( \frac{\mathbf{P}_0 - \frac{2U}{\mathbf{E}}}{\gamma} \right) \left( \frac{\pi K}{-\Delta \varepsilon} \right)^{1/2} \equiv v_0 \frac{\mathbf{E} - \mathbf{E}_{\text{th}}}{\mathbf{E}} \quad (2)$$

with  $\mathbf{E}_{\text{th}} = 2U/\mathbf{P}_0$  and  $v_0 = 2\mathbf{P}_0/\gamma(\pi K/ -\Delta \varepsilon)^{1/2}$ , where  $\gamma$  is a rotational viscosity. This solution is stable only for  $\mathbf{E}$  in the vicinity of  $\mathbf{E}_{\text{th}}$ , which is the so-called ‘Huxley regime’ [15] in which a synclinc phase invades a metastable anticlinc phase. For fields not close to  $\mathbf{E}_{\text{th}}$ , one enters the so-called ‘Fisher regime’ in which a synclinc phase invades an unstable anticlinc phase. Although no stable analytic solution exists in this regime, numerical analysis indicates that the velocity does not exhibit any upward curvature, contrary to the experimental observations shown in figure 4. Wang and Taylor included the additional effects of surface anchoring on the propagation velocity [16], but found that anchoring tends to reduce the velocity. In a more recent paper describing a synclinc to anticlinc phase transition via solitary waves on cooling [13], we introduced an additional symmetry-allowed term into the free energy, viz.  $1/2A \sin^2 \varphi$ , where  $A$  is a positive constant. This is the lowest order term that has the effect of favouring the odd-layer tilt plane normal being either parallel or antiparallel to the even-layer tilt plane normal, but disfavoring perpendicular tilt plane normals. Inclusion of this term, however, does not change the basic nature of the theoretical solution, and seems to leave the theory at odds with the experimental results.

Faced with an apparent deviation between experiment and extant theory, we now wish to conjecture about two other plausible mechanisms that could produce an upward curvature of  $v(\mathbf{E})$ . First, we reiterate that for large  $\mathbf{E}_r$ , equation (2) is not exactly valid, although numerical calculations show that the stable solution is not very different from that of equation (2). Thus, the shift to Fisher behaviour cannot be the cause of the observed large non-linear velocity vs. electric field.

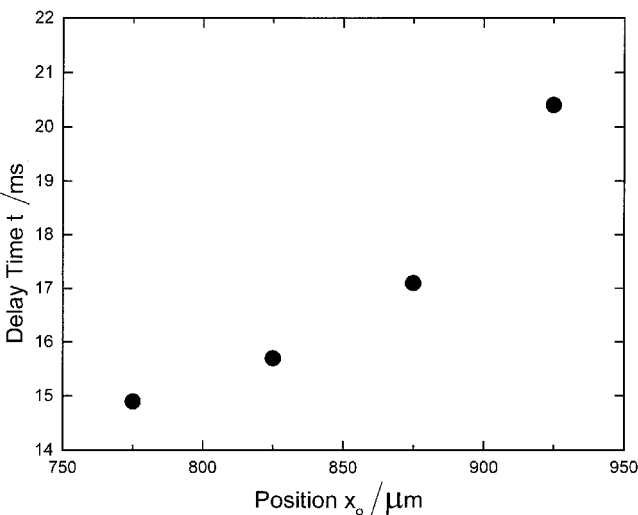


Figure 5. Delay time  $t$  between trigger pulse and the onset of a non-zero intensity at the detector vs. the slit position  $x_0$  (along the smectic layer) from the approximate nucleation site.

Clearly, then, the model needs to be modified. One possibility is that the spontaneous polarization of the smectic layers is field-dependent due to a biasing of the molecular rotations by  $\mathbf{E}$ . In fact, the quantity  $p\mathbf{E}$  can be of order  $0.1k_{\text{B}}T$  for the large fields used in our experiment, where  $p$  is a molecular dipole moment. This could have the effect of reducing the effective interlayer coupling parameter  $U$  as the field is increased, leading to some upward curvature in  $v(\mathbf{E})$ , but not to a sufficient extent to explain the experimental results.

A second possible mechanism for the upward curvature involves shear thinning. In [3] we assumed not only a constant  $U$ , but also a rotational viscosity  $\gamma$  that is independent of the rate of rotation of the molecule. It is well known that the phenomenon of shear thinning reduces the effective viscosity of polyethylene at shear rates  $\dot{\tau}$  greater than  $\sim 1 \text{ s}^{-1}$  [17]. In our experiments the alkane tails of the molecules move distances of the order of a molecular length  $a$  in times of the order  $0.1 \mu\text{s}$ , which is deduced by dividing the characteristic width of the solitary wave front (typically  $\sim 0.1 \mu\text{m}$ , cf.  $(K/U)^{1/2}$ ) by its velocity of the order  $100 \text{ cm s}^{-1}$ . Since the thickness of the layer in which the relative motion occurs was also of the order  $a$ , the shear rate at the boundary layer was  $\sim 10^7 \text{ s}^{-1}$ . It thus seems plausible that, despite the shortness of the alkane tails in comparison with those in polyethylene, some considerable shear thinning may occur, leading to an increased velocity. For example, in polymeric liquid crystals it has been found that in a certain range of  $\dot{\tau}$  the viscosity coefficient scales as  $\dot{\tau}^{-1/2}$ , and accordingly the viscous force scales as  $\dot{\tau}^{1/2}$  [18]. The tail–tail interactions in anticlinic liquid crystals have some characteristics of chain–chain interactions in polymers [19], and may have similar dynamic consequences. Our preliminary numerical calculations show that a shear rate-dependent viscosity coefficient  $\gamma \propto (\partial\phi/\partial t)^{-1/2}$  results in a field-dependent velocity with an increasing upward curvature. Work in this direction is underway.

#### 4. Summary

To summarize, we have extended our measurements of synclincic finger velocity invading an anticlinic liquid crystal phase to larger applied electric fields. We found that the velocity increases sharply with applied field, and is by far the largest ever observed for a liquid crystal. This behaviour at large fields is apparently inconsistent with a previously published model that adequately describes the fingering at low fields. We have suggested two possible modifications of the previous

model, viz., a field-dependent interaction parameter  $U$  and shear thinning, that may help explain the observed behaviour. Both experimental and theoretical work are now in progress to elucidate the mechanism responsible.

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