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N. J. Mottram [a], T. J. Sluckin [b], S. J. Elston [a] & M. J. Towler [c]

[a] Department of Engineering Science, University of Oxford, Parks Road, Oxford, OX1 3PJ, U.K.

[b] Faculty of Mathematical Studies, University of Southampton, Highfield, Southampton, SO17 1BJ, UK

[c] Sharp Laboratories of Europe Ltd., Edmund Halley Road, Oxford Science Park, Oxford, OX4 4GA, UK

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N. J. MOTTRAM[a], T. J. SLUCKIN[b], S. J. ELSTON[a] and M. J. TOWLER[c]

[a]*Department of Engineering Science, University of Oxford, Parks Road, Oxford, OX1, 3PJ, U.K,* [b]*Faculty of Mathematical Studies, University of Southampton, Highfield, Southampton SO17 1BJ, UK and* [c]*Sharp Laboratories of Europe Ltd., Edmund Halley Road, Oxford Science Park, Oxford OX4 4GA, UK*

In this paper we present a theoretical analysis of shear induced melting in smectic A liquid crystals. We have performed a detailed numerical analysis and find that for a range of parameter values a first order transition, from a supersheared state to a more relaxed state, is periodically encountered as the total shear is increased. This first order transition involves the melting and reformation of the smectic layers. In this paper we present a summary of this analysis while a more detailed analysis, including analytic expressions for certain critical quantities, is presented elsewhere.

Keywords: theory; smectic A; smectic layer melting; shear deformation

INTRODUCTION

In the smectic A phase there exists one-dimensional positional ordering in the form of a periodic molecular density function, which forms a layered structure[1]. In equilibrium, the unconstrained liquid crystal layers are uniformly separated by a distance $2\pi/q$, where q is the preferred wave number of the density fluctuations. However, in the presence of distorting forces, the layer structure can be significantly different. In this paper we discuss one situation in which this layer structure may be distorted.

We consider a smectic A liquid crystal sample initially in the bookshelf geometry, in which the director is aligned in the same uniform direction on each surface of a cell (Fig. 1). Although this bookshelf configuration may be disturbed by external factors such as an electric field, applied stresses and alterations in the temperature, such perturbations typically do not alter the smectic layer configuration close to the surfaces. Cagnon and Durand[2] have experimentally investigated this interaction between the cell surfaces and the smectic layers. In a cell filled with a smectic A liquid crystal, one wall was moved laterally with respect to the other and they found

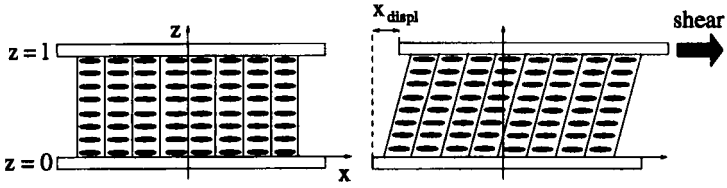

Figure 1: Cell configuration: Smectic A material is sandwiched between glass plates at $z = l$ and $z = 0$ where the director is aligned homogeneously. The shear force applied to the upper glass plate induces layer distortion.

periodic discontinuities superimposed on a monotonic, background stress, which were interpreted as layers slipping with respect to the walls. These results enabled Cagnon and Durand to estimate the energy associated with the surface-memory-induced layer anchoring at the walls.

This paper aims to examine the general question of the interplay between the energetically favorable bookshelf geometry and an applied shear stress which distorts it. Elston and Towler[3] have previously shown that the presence of a large inter-wall shear can lead to a significant reduction in the smectic order parameter in the middle of the cell, and sufficiently large shear can, under some circumstances, destabilize the deformed bookshelf geometry. In this paper we report on calculations which extend this analysis to find a complex structure of stable, metastable and unstable states and describe the associated transitions between them. A more complete account of this work is presented elsewhere[4].

PHYSICAL UNDERSTANDING

In this section we give a brief overview of the physics involved in the model we consider. For simplicity we suppose that the layer anchoring is infinite and thus in the presence of shear the layers are forced, initially, to follow the moving surfaces (Fig. 1).

We denote the smectic order parameter by the complex variable[5], $\Psi = \rho e^{i\Phi}$, where ρ is the degree of order and Φ is its phase, and measure the degree of stress by the relative lateral displacement, x_{displ} of the walls (Fig. 1). The quantity $\tau = 2\pi x_{displ}/m$, where m is the smectic layer thickness, is the change in phase between points which would lie opposite to each other in the bookshelf geometry. We shall find different behaviour depending on whether the ratio of the cell thickness to the smectic correlation length l/ξ is much greater than or much less than unity, with a sharp

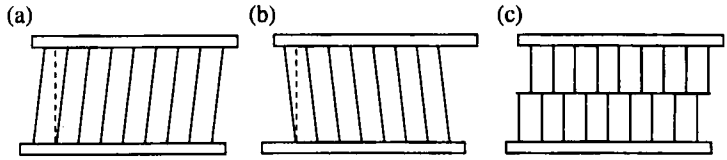

Figure 2: (a), (b) Energetically equivalent tilted layers for $\tau = \pm\pi$. (c) Mismatched layers where the phase difference of $+\pi$ and $-\pi$ are indistinguishable.

crossover between the two régimes.

We find that for extremely small shears, $-\pi \leq \tau \leq \pi$, the layers will tilt in order to connect between the surfaces, resulting in an increase in the curvature free energy. In order to minimize this curvature free energy the smectic liquid crystal may reduce its order parameter in the bulk of the system.

When the smectic layers at the cell surfaces are *completely mismatched* i.e. exactly out of phase, there are two energetically equal layer configurations. The layer at one surface is positioned exactly between two layers at the opposite surface and may join to either one. The crucial question is whether these states are physically distinct.

If they are different (Fig. 2(a) and (b)), then as the amount of shear is increased through π the sheared state becomes a metastable state, and the previously stable free energy branch is extended into a metastable region. We shall refer to this state as *supershear*. Transforming from Fig. 2(a) to (b) involves breaking up or melting the layer structure.

An alternative scenario is illustrated in Fig.2(c), in which the phase difference of $+\pi$ and $-\pi$ are identical. The system melts and the phase is undefined at the centre of the cell. At larger shear reverse tilt layers reform with a phase difference across the sample between $-\pi$ and 0 .

The first scenario occurs for thick and the second for thin cells and at a critical cell thickness $l = l_c$ the behaviour changes. For very thick films $l/\xi \gg 1$, there will be many metastable supersheared states for a given τ. The loss of stability of the highest energy supersheared state moves the system to the next state down in energy.

In the following section we shall develop a mathematical formalism to describe the physical ideas presented in this section.

THEORETICAL MODEL

The Landau-de Gennes free energy of the smectic liquid crystal may be written as[6]

$$F = \int d\mathbf{r} \left[a|\Psi|^2 + \frac{b}{2}|\Psi|^4 + \gamma_{\|}|(\hat{\mathbf{n}}\cdot\nabla - iq)\Psi|^2 + \gamma_{\perp}|(\hat{\mathbf{n}}\times\nabla)\Psi|^2 + \frac{1}{2}K_{11}(\nabla\cdot\hat{\mathbf{n}})^2 + \frac{1}{2}K_{33}(\hat{\mathbf{n}}\times(\nabla\times\hat{\mathbf{n}}))^2 \right]. \quad (1)$$

Minimization of the first two terms in (1) leads to the bulk smectic order parameter modulus, $\rho_c = (-a/b)^{1/2}$ and stability of the smectic phase requires that $a < 0$ and $b > 0$. In equation (1) the $\gamma_{\|}$ term is associated with layer dilations whilst the $\gamma_{\perp}$ term is associated with departures of the director from the smectic normal. The last two terms represent the elastic energy of distortions to the director $\hat{\mathbf{n}}$ within the smectic layer.

The smectic A material is placed in a cell with flat walls at $z = 0$ and $z = l$ (Fig. 1). We choose boundary conditions that give a homogeneous alignment of the director: $\hat{\mathbf{n}}(z = 0) = \hat{\mathbf{n}}(z = l) = \hat{\mathbf{x}}$, and fix the smectic order parameter such that the phase, Φ, is strongly anchored at the surface and the modulus, ρ, remains at the bulk equilibrium value: $\Psi(z = 0) = \rho_c e^{iqx}$, $\Psi(z = l) = \rho_c e^{i(qx+\tau)}$.

It will be useful to parameterize the order parameter in the following way,

$$\Psi(x, z) = \rho(z)e^{i\Phi(x,z)} = \rho(z)e^{i(qx+\phi(z))} = \psi(z)e^{iqx}. \quad (2)$$

where $\psi(z) = \rho(z)e^{i\phi(z)}$. This parameterization separates out the explicit x-dependent layer behaviour, given by $\Phi_0(x) = qx$, from the shear-induced variation across the cell, given by $\phi(z)$. The Ψ boundary conditions can now be rewritten as: $\psi(z = 0) = \rho_c$, $\psi(z = l) = \rho_c e^{i\tau}$. Thus the phase $\phi(z)$ ranges from 0 to τ across the cell, though of course layer slippage may allow the phase to vary between 0 and $\tau \pm 2n\pi$ where n is an integer. In this way the free energy may now be written as a functional of $\rho(z)$ and $\phi(z)$ rather than Ψ.

We will assume that layer compression is energetically unfavourable compared to other forms of distortion within the liquid crystal (i.e. $\gamma_{\|}$ is very large compared to all other energy coefficients). Therefore the director is prevented from rotating with the layer since the layers would then have the wrong thickness along the x direction. This implies that $\hat{\mathbf{n}} = \hat{\mathbf{x}}$.

Using the non-dimensionalisations, $\tilde{\psi} = \psi/\rho_c$, $r = \rho/\rho_c$, $\tilde{z} = z/l$, $d = l/\xi$, $\tilde{F} = (b/(2la^2))F$, where $\xi = (\gamma_{\perp}/|a|)^{1/2}$ is the smectic correlation

length, the free energy becomes,

$$F = \int_0^1 dz \left[\frac{1}{4} \left(r^2 - 1 \right)^2 + \frac{1}{2d^2} \left(r'^2 + r^2 \phi'^2 \right) \right], \tag{3}$$

where the tildes have been dropped and derivatives with respect to z are denoted by primes. With the above non-dimensionalisation the boundary conditions are $r(0) = 1$, $\phi(0) = 0$ and $r(1) = 1$, $\phi(1) = \tau$.

Minimization of the free energy (3) leads to the Euler-Lagrange equations:

$$r'' + d^2 r(1 - r^2) + r\phi'^2 = 0; \tag{4}$$

$$\left(r^2 \phi' \right)' = 0. \tag{5}$$

We now have two control parameters, the nondimensional thickness d and the shear τ. When d is small, the $(1 - r^2)^2$ term in (3) can be ignored so that changes in the smectic amplitude r are not energetically expensive and smectic melting can occur in the center of the sample. For large d, r is constrained to be close to 1 and the system now prefers changes in ϕ, i.e. layer tilting.

Equations (4) and (5) are in fact the time-independent Ginzburg-Landau equations and can be used to describe a vast array of nonlinear phenomena such as Rayliegh-Benard convection, Taylor-Couette flow, reaction diffusion systems, nonlinear optics, superfluidity, combustion and other liquid crystal systems (see the references contained in [7]). The analogies between such systems and ours are therefore strong. Subsequently, these analogies will be extremely useful in suggesting the mechanism of the dynamics of layer slipping.

In the following section we solve the above governing equations for two sets of parameter values in order to demonstrate the two kinds of behaviour that this system exhibits, as discussed in the previous section. A detailed numerical analysis of the system for all parameter values has been carried out and is to be published elsewhere[4]. We are also able to obtain many analytical results for certain key quantities such as the critical thickness and the critical shear at which melting of the layers occurs, as a function of cell thickness. However, we will also leave this analysis for the more detailed paper.

NUMERICAL RESULTS

We solve equations (4) and (5) subject to the above boundary conditions (using the numerical continuation package AUTO97[8]) for two values of the

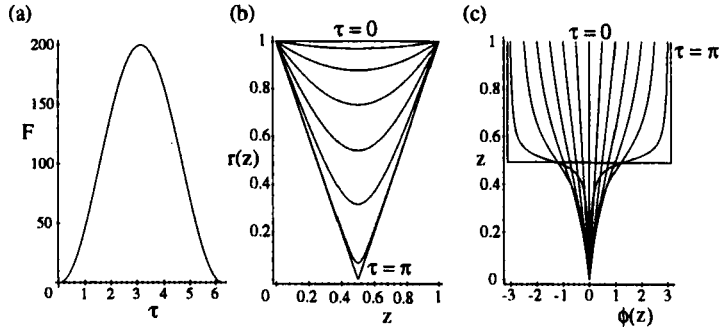

Figure 3: Numerical solutions for $d = 0.1$. (a) Energy, F, versus shear, τ. (b) The density modulation amplitude, $r(z)$. (c) The density modulation phase, $\phi(z)$ (in radians).

cell thickness, $d = 0.1$ and $d = 10.0$, and investigate the behaviour as the amount of shear τ is varied.

Fig. 3 shows the free energy versus shear and the $r(z)$ and $\phi(z)$ solutions for $d = 0.1$. As shear increases the phase $\phi(z)$ is almost linear across the cell whilst the order parameter r decreases in the centre of the cell. As the shear approaches π the phase change across the cell concentrates in the centre. The large value of $\mathrm{d}\phi/\mathrm{dz}$ favours *melting* of the smectic layers i.e. a reduction of the order parameter $r(z)$. At $\tau = \pi$ the phase and the derivative of $r(z)$ are discontinuous and $r(z = 0.5) = 0$. At this point the energy is a maximum. For $\tau > \pi$ this process is reversed as the system relaxes to the energy minimum at $\tau = 2\pi$.

For $d = 10.0$ the behaviour has changed significantly (Fig. 4). For shear values $0.458 < \tau < 5.825$ there are three solutions, two stable and one unstable. The stable solutions occur along branches 1, 2 whilst the unstable along branch 3. At two *limit points* the unstable solution and a stable solution meet and annihilate each other so that in the regions $0.0 < \tau < 0.458$ and $5.825 < \tau < 2\pi$ there exists only one solution. At the point $\tau = \pi$ the two stable branches have the same energy whereas for $0.458 < \tau < \pi$ branch 1 is the global minimum and for $\pi < \tau < 5.825$ branch 2 is the global minimum. As the cell is sheared past $\tau = \pi$ the system will remain on branch 1 (even though this state is not the global minimum state) until either defects or thermal fluctuations perturb the system or the limit point is reached at which point the system relaxes into the reverse tilt state on branch 2.

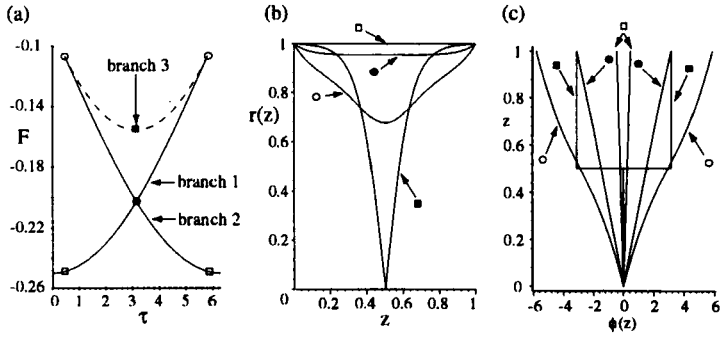

Figure 4: (a) Energy, F, versus shear, τ, for $d = 10.0$. (b) solutions for $r(z)$ and (c) $\phi(z)$ on all three branches for $d = 10.0$ and shear values, $\tau = 0.458, \pi, 5.825$. The symbols in (a) correspond to the symbols in (b) and (c).

Figure 4 also shows the $r(z)$ and $\phi(z)$ solutions at various points on branches 1-3. Solutions on branch 1 and 2 are characterised by almost linear shear across the cell with a small amount of melting in $r(z)$ whilst solutions on branch 3 are characterised by concentrated shear in the middle of the cell and a large amount of melting.

Since Fig. 4 is repeated periodically, for $\tau > \pi$, the shear stress in a linearly sheared cell would periodically increase and decrease as the system periodically followed branch 1 then fell to a relaxed state on branch 2.

The behaviour of the system for values of d other than 0.1 and 10.0 may be characterised by investigating how the two limit points vary as d is changed (Fig. 5). For $d < d_c \approx 3.5$ the behaviour is essentially the same as that of $d = 0.1$ and no limit points exist. For $d > d_c$ the behaviour is similar to that of $d = 10.0$, however as d increases the two limit points diverge (linearly) and eventually move out of the region $0 < \tau < 2\pi$. For large d, branches will overlap and there will exist increasingly more stable and unstable solutions for any fixed τ. Fig. 5(b) shows the crossing of the limit point loci. For all points in a diamond shaped region the number of stable and unstable solutions is fixed. Fig. 5(b) can therefore be thought of as a phase diagram. Whilst there will be only one global energy minimizer at each point (τ, d) there may be many metastable solutions which are locally stable.

In the smectic A phase, the smectic correlation length ξ is typically of the order of the size of a few molecules. For an experimental cell dimension

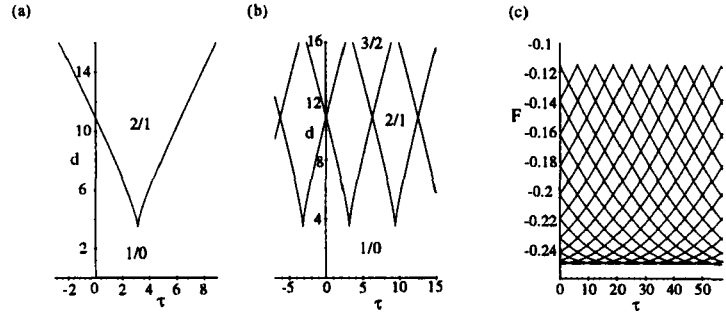

Figure 5: (a) The limit points formed at $d = 3.50$, $\tau = \pi$ diverge as d increases. (b) The system symmetry implies that limit points are formed at the points $d = 3.50$, $\tau = \pm\pi, \pm 3\pi, \pm 5\pi \ldots$ and as d increases the limit points diverge and eventually *cross over*. The label i_s/i_u denotes the number of stable and unstable solutions in each region. (c) Energy versus shear for $d = 100.0$, only the stable solutions are shown. There are now many stable solutions for each shear value.

1-10μm the nondimensional cell width is then $d \sim 100 - 1000$ and so we expect many metastable solutions.

For $d = 100$, the energy versus shear plot is shown in Fig. 5(c), where only the stable solutions are shown for simplicity. At this parameter value the limit point of branch 1 occurs at a shear of $\tau = 56.69$, and for each shear value τ there are indeed many stable solutions.

Although characteristic values of d are large, we note that ξ is expected to increase dramatically close to a continuous nematic-smectic A phase transition. In this region d may approach d_c and some of the interesting structure near to this critical point may be easier to observe.

DYNAMICS OF LAYER SLIPPING

The theory in this paper is quasi-static and does not model the dynamics of the breakdown of supershear. Nevertheless our investigation of the quasi-static theory is able to give a qualitative picture of the dynamic process. At this point it is helpful to consider the system in terms of the real and imaginary components of the smectic order parameter $[R_1(z) = r(z)\cos(\phi(z))$ and $R_2(z) = r(z)\sin(\phi(z))]$ instead of the usual amplitude and phase, $\rho(z)$ and $\phi(z)$. The solutions to the equations may then be thought of as trajectories in the complex plane. The boundary conditions of the problem insist

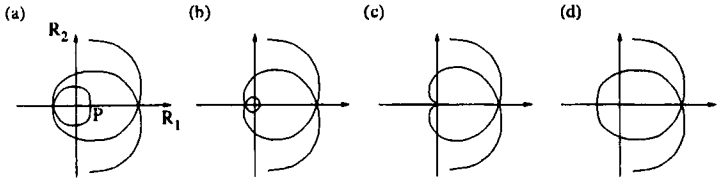

Figure 6: The dynamic process as the system relaxes from the supersheared state at the limit point. (a) The innermost loop of the original trajectory encircling the origin of the complex plane. (b) The loop starts to collapse. (c) The trajectory passes through the origin as the loop disappears at a cusp. (d) The new stable orbit now has one less loop.

that each solution starts at the point (1, 0), which corresponds to $r = 1$, $\phi = 0$, and finishes at the point $(\cos(\tau), \sin(\tau))$, which corresponds to $r = 1$, $\phi = \tau$. The larger τ the more times the trajectory winds around the origin. If the trajectory passes through the origin the amplitude r has gone to zero and at that point the smectic layers have melted. As these trajectories loop around the origin they spiral inwards, as ρ decreases, and then outwards in a symmetrical way.

As the limit point is reached the innermost loop of the trajectory collapses and the other loops readjust themselves (Fig. 6). The position closest to the origin, P, approaches the origin and passes through the origin as the loop disappears at a cusp. The original trajectory has now decayed to a new stable trajectory with one less loop around the origin. Thus each layer has *lost* a phase of 2π and the layers have *slipped* back to a more relaxed configuration.

The details of this description will depend on the dynamical structure of the equations governing smectic A layer motion. A simple time-dependent version of equations (4) and (5) which allows only for dissipative behaviour is the Ginzburg-Landau equation. The dynamics of the 2π reduction in phase seen above has previously been studied in detail[9-12]. However, the full equations are inevitably more complicated and include smectic A hydrodynamics in the presence of layer conservation. Whatever these details, they will not alter the stable qualitative dynamical features discussed here.

DISCUSSION

In this paper we have presented a summary of our analysis of shear-induced melting in smectic A liquid crystals. The calculations reveal a complex

phase diagram described by the two system control parameters, the nondimensionalised gap width d and the imposed shear τ.

We have found that there is a critical value of the cell thickness d_c at which the behaviour changes. For $d \leq d_c$ the layers continuously melt and reform as the shear increases through $n\pi$ for odd n. For $d > d_c$ the behaviour is significantly different. It is now possible to supershear the layers into a metastable state with $|\tau| > \pi$ until a critical value of the shear $\tau_c(d)$. The critical shear value is linear with respect to d (i.e. $\tau_c(d) \to \gamma_c d$) and for large values of d, there will be a large number of metastable states. When the system reaches the critical shear value the system relaxes into the next highest free energy metastable state reducing the phase by 2π and melting at the centre of the cell as it relaxes.

The experiments of Cagnon and Durand[2] showed that the response of a sheared smectic A in the bookshelf geometry had two components. The major component was a linear behaviour superposed on which was a smaller periodic response. Linear behaviour is just what is expected for $\tau < \tau_c$; the stored free energy is proportional to τ^2 just as in Hooke's law, as can be seen in Fig. 5(c). By contrast, periodic behaviour is what is expected for $\tau \sim \tau_c$, for now the system reaches a critical value, relaxing, increasing to its critical value, relaxing and so on.

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