Spreading of a smectic A liquid crystal on a rotating disc

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We describe the dynamics of spreading of a smectic A liquid crystal deposited on a rotating disc. We show the existence of several regimes according to the film thickness. If it is thick enough, the thinning is controlled by bulk permeation; at intermediate thicknesses the surface dissipation dominates, whereas viscous effects begin to be felt only at very small thickness.

1. Introduction

If a drop of an isotropic liquid is deposited on to a rotating disc, it spreads rapidly due to the centrifugal force. The experiment shows that the thickness, h, of the liquid film becomes quickly homogeneous in its central part and decreases asymptotically as $t^{-1/2}$ where t is time [1, 2]. This property is used by manufacturers in order to make homogeneous deposits of known thickness. The question is: does a smectic A liquid crystal behave in the same way?

We recall that a smectic A phase is a lamellar liquid crystal where the molecules are arranged in fluid layers which can glide viscously over each other. One of their essential hydrodynamic characteristics is that they behave like an ordinary viscous liquid when the barycentric velocity **v** of the molecules is parallel to the layers, and like a porous medium when **v** is perpendicular to the layers. In this case, the liquid crystal plays both the role of the fluid and of the porous medium: it is often said that there is permeation, a concept that was first introduced by Helfrich in 1969 [3]. Such a flow has been observed experimentally by studying the dynamics of the collapse of a smectic bubble [4]. This experiment is very delicate to perform and does not allow the permeation coefficient to be measured accurately. Here, we propose a new method for measuring this transport coefficient (both in the bulk and on the surface), which is much easier to perform than the previous method.

2. Summary of the hydrodynamic equations for a smectic A liquid crystal

The flow geometry is pictured in the figure. A smectic A film, of initial thickness h_0, is deposited on to a disc of radius R which is rotating around its axis of revolution with an angular velocity ω. The disc is treated in homeotropic anchoring, the layers remaining parallel to the substratum during spreading.

The stationary equations of motion in the rotating frame, can be obtained by adding to the balance of the forces, the centrifugal and the Coriolis forces. We shall neglect the Coriolis force, an assumption that is justified if ω is much smaller than $\mu/\rho h^2$, where h is the film thickness at time t, μ a typical viscosity coefficient and ρ is the density that we assume to be constant. In cylindrical coordinates (r, θ), the bulk

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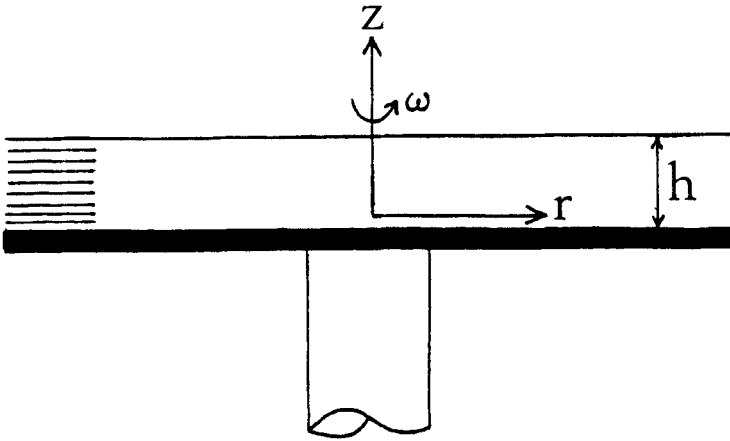
Smectic film on a rotating disc. The smectic layers are parallel to the surface of the disc (homeotropic anchoring), a condition easy to fulfil by coating the surface with a polymer (silane for instance).

equations simplify if it is assumed that the five viscosity coefficients are equal and that the convective terms are negligible (Reynolds number much smaller than 1) [5]

$$\frac{\partial P}{\partial r} = \mu\left(\frac{\partial^2 v_r}{\partial r^2} + \frac{1}{r}\frac{\partial v_r}{\partial r} - \frac{v_r}{r^2}\right) + \mu\frac{\partial^2 v_r}{\partial z^2} + \rho\omega^2 r, \tag{1}$$

$$\frac{\partial P}{\partial z} = \frac{v_z}{\lambda_p}, \tag{2}$$

$$\frac{\partial v_r}{\partial r} + \frac{\partial v_z}{\partial z} + \frac{v_r}{r} = 0. \tag{3}$$

We have also neglected the viscous term in the second equation, which supposes that the film thickness is larger than the permeation length $l_p = (\mu\lambda_p)^{1/2}$, which is of the order of a layer thickness [5]; λ_p is the bulk permeation coefficient. These two conditions are not very restrictive and almost always satisfied. The boundary conditions on the free surface read:

$$P = B\frac{\partial u}{\partial z} = \sigma, \tag{4}$$

$$\sigma_{zr}^v = \mu\left(\frac{\partial v_r}{\partial z} + \frac{\partial v_r}{\partial r}\right) = 0 \quad \text{at} \quad z = h, \tag{5}$$

$$v_z = \zeta\sigma, \tag{6}$$

while, on the surface of the glass, the layer displacement u and the velocity both vanish. The elastic stress σ is associated with the displacement u and ζ is a surface permeation coefficient that describes how the layers disappear at the free surface. Finally we must check that the pressure P vanishes on the side surface perpendicular to the layers, namely

$$P = 0 \quad \text{at} \quad r = R. \tag{7}$$

3. Search for an approximate solution

In this section, we shall content ourselves with describing the thinning of the film only in its central part ($r \ll R$). By analogy with the solution that is known for ordinary viscous fluids, we look for a velocity and a hydrostatic pressure field of the form

$$v_r = -Cz^2 r + Drz, \tag{8}$$

$$v_z = 2C\frac{z^2}{3} - Dz^2, \tag{9}$$

$$P = \frac{E}{2}(r^2 - R^2) + f(z), \tag{10}$$

where C, D and E are three constants to be determined as well as the function $f(z)$. It is easy to verify that this particular choice allows us to satisfy exactly the bulk equations (1)–(3) as well as the boundary conditions at the free surface $z = h$ (equations (4)–(6)) and on the plate at $z = 0$. By contrast, it is impossible to cancel exactly the pressure on the side surface with this type of solution. A reasonable assumption consists in fulfilling this condition on average on this boundary ($\langle P \rangle = 0$ at $r = R$), which imposes

$$\int_0^h f(z)\,dz = 0. \tag{11}$$

In order to calculate the constants C, D and E, it is sufficient to replace v_z and v_r by their expressions (8) and (9) in the equations of motion. Substituting into equation (1) for v_r yields

$$E = -2C\mu + \rho\omega^2, \tag{12}$$

while condition (5) on the vanishing of the shear stress at the free surface gives

$$D = 2Ch. \tag{13}$$

From equation (2) for v_z and condition (11) we obtain

$$P = \frac{E}{2}(r^2 - R^2) - \frac{2C}{\lambda_{\mathrm{p}}}\left(\frac{z^4}{12} - \frac{hz^3}{3}\right) - \frac{2}{15}\frac{C}{\lambda_{\mathrm{p}}}h^4. \tag{14}$$

One more relation is obtained by writing that $v_z = \zeta P$ on the horizontal free surface at $z = h$

$$\frac{4}{3}Ch^3 = \zeta\left(\frac{ER^2}{2} - \frac{11}{30}\frac{Ch^4}{\lambda_{\mathrm{p}}}\right). \tag{15}$$

Relations (12) and (15) allow us to calculate the constant C

$$C = \frac{\rho\omega^2 R^2 l_{\mathrm{p}}^2}{2\mu\left(\frac{4}{3}h^3\Lambda + \frac{11}{30}h^4 + R^2 l_{\mathrm{p}}^2\right)} \tag{16}$$

which is directly related to the evolution law of the film thickness

$$\frac{dh}{dt} = -\frac{4}{3}Ch^3. \tag{17}$$

This equation can be integrated immediately and gives

$$\frac{1}{R^2 l_p^2}\left(\frac{11}{30}h_2 + \frac{8}{3}\Lambda h\right) - \frac{1}{h^2} = \frac{4}{3}\frac{\rho\omega^2}{\mu}(t_0 - t). \tag{18}$$

At time $t = 0$, the film thickness is equal to h_0, which fixes the integration constant t_0. We see from this equation that there are several regimes of thinning according to the film thickness as well as three typical lengths

$$h_1 = \left(\frac{R^2 l_p^2}{\Lambda}\right)^{1/3}, \quad h_2 = (Rl_p)^{1/2} \quad \text{and} \quad \Lambda = \frac{\lambda_p}{\zeta}.$$

The first two lengths depend on the sample size. The third Λ is a length that depends only on the material chosen. This length is related to the structure of the smectic–air interface and depends only on the experimental conditions. For the time being, we shall consider it to be constant, an assumption that we shall discuss in the last section. According to the order of magnitude of each of these lengths, several spreading regimes can be considered.

4. Different spreading regimes

Two distinct situations must be considered according to whether Λ is greater or smaller than h_2.

(1) If $\Lambda > h_2$, three regimes occur successively:
   (a) at large thickness, $h > \Lambda$, the thinning kinetics are dominated by the bulk permeation. In this regime, h decreases as $R\omega l_p(t_0 - t)^{1/2}$;
   (b) at intermediate thicknesses, $h_1 < h < \Lambda$, the surface permeation dominates. The film thickness decreases linearly versus time as $\omega^2 R^2 \zeta(t_0 - t)$;
   (c) at small thickness, $h < h_1$, the viscosity dominates and the thickness decreases as $\omega^{-1} t^{-1/2}$. We find again the same behaviour as in an ordinary fluid.

(2) If $\Lambda < h_2$, the regime of surface permeation is screened by the other two. In this case, the bulk permeation dominates as long as $h > h_2$, whereas the viscosity becomes preponderant when $h < h_2$.

Let us emphasize that, at large thickness, the film dynamics are always dominated by the bulk permeation. We have, therefore, a convenient way to measure the bulk permeation coefficient λ_p.

5. Calculation of Λ

The length Λ and the associated surface permeation coefficient ζ depend on the structure of the smectic–air interface. If the interface is rough, Λ is of the order of a molecular length. In this case, $h_2 \gg \Lambda$, and we are in the second case that we have just described in the previous section. If the interface is faceted, which is the case experimentally for most of the materials [6], Λ is linked to the mean distance $\bar{\Lambda}$ between two steps on the free surface [4]. Indeed we have

$$v_z = \frac{b}{\bar{\Lambda}}v = \frac{b}{\bar{\Lambda}}m\sigma, \tag{19}$$

where v is the step velocity and m its mobility; we then find

$$\Lambda = \frac{\lambda_p}{bm}\bar{\Lambda}. \tag{20}$$

If the mobility is close to that of a bulk dislocation, namely $m \approx (\lambda_p/\mu)^{1/2}$ [7], then $\Lambda = \lambda_p/\zeta = (l_p/b)\bar{\Lambda} \approx \bar{\Lambda}$ and Λ can be identified with the mean distance between steps.

Let us now suppose that there exist a few screw dislocations emerging at the free surface. Experiments of microplasticity have proven that these dislocations exist [8]. From the emergence point of each dislocation starts a microscopic step of height equal to the layer thickness b. In rotation, the stress σ exerts a force σb on this step which consequently deforms. This force is analogous to the supersaturation force that acts on a step on crystal growth. It is known that, in a stationary regime, the step takes the form of a spiral whose geometrical characteristics have been calculated by Burton *et al.* [9]. In this model, $\bar{\Lambda}$ can be identified with the distance between two turns of the spiral. A complete calculation of the shape of the spiral shows that we must take in this case:

$$\bar{\Lambda} \approx \frac{4\pi}{0\cdot63}\frac{\beta}{\sigma b} \approx 20\frac{\beta}{\sigma b} \tag{21}$$

independently of the number of spirals per unit area. β is the line tension of a step. Thus, we see that the distance $\bar{\Lambda}$ is not independent of the velocity but depends on it via σ.

In order to conclude, let us make a few numerical estimates. We know that $\beta \approx 10^{-6}$ dyn [6], $b \approx 3 \times 10^{-7}$ cm. Furthermore, $|\sigma| \approx \rho R^2 \omega^2/2$, by taking $\rho = 1$ g cm^{-3}, $R = 1$ cm and $\omega = 100$ s$^{-1} \approx 1000$ rpm, $|\sigma| \approx 5000$ dyn cm^{-2}. We then calculate $\bar{\Lambda} \approx \Lambda \approx 100\,\mu$m. On the other hand, we calculate $h_1 \approx 3\,\mu$m and $h_2 \approx 1\,\mu$m by taking $l_p \approx 10^{-7}$ cm. We are then in case (1) discussed in §4. We conclude, therefore, that, if the thickness ranges between 3 and $100\,\mu$m $(h_1 < h < \Lambda)$, then the surface dissipation dominates and the thickness decreases linearly in time according to the following spreading law

$$h/\mu\text{m} = \frac{\rho\omega^2 R^2}{2}\zeta(t_0 - t) = \frac{m\rho^2 b^2 \omega^4 R^4}{80\beta}(t_0 - t) \approx 1000\,\text{m}\,(t_0 - t), \tag{22}$$

where time is expressed in seconds. If we assume that the step mobility is of the same order of magnitude as that of an edge dislocation, that is to say $m \approx 10^{-6}$ cm^3 dyn^{-1} s^{-1} [8], we find that the thinning velocity is close to $3\cdot6\,\mu$m/h for a velocity $\omega = 100$ s^{-1}. This velocity should be easily measurable experimentally. Its large dependence (in ω^4) on the rotation velocity should also constitute an excellent experimental test of the proposed mechanism.

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