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Backflow effects in the Goldstone mode dielectric response of chiral smectic C* liquid crystals

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Employing the elastic-hydrodynamic theory of the ferroelectric smectic C* phase, it is easily seen that in most dynamic processes in the system, a coupling between director rotations and macroscopic mass flow exists. Taking this possibility of backflow into account, the equations governing the Goldstone mode dielectric susceptibility are rederived, and it is shown that the corresponding rotational viscosity, measured in a conventional dielectric experiment, is renormalized due to backflow effects. An expression for this renormalization is given and the crucial parameter, determining whether backflow effects are of importance or not in the dielectric experiment, is derived.

1. Introduction

The general dynamical behaviour of chiral smectic C* (SmC*) liquid crystals can be described by a set of equations governing the time evolution of the *c*-director, the macroscopic mass flow and the orientation of the smectic layer normal. The first dynamical studies of SmC* liquid crystals presented in the literature were studies concerning the rotation of the *c*-director, neglecting the possible onset of macroscopic flow and also assuming the smectic layers to remain fixed. Such studies mainly concerned the switching dynamics of ferroelectric surface stabilized liquid crystal cells [1, 2] and the dynamics of the dielectric response of ferroelectric SmC* liquid crystals [3, 4]. The equations used in these studies are heuristic equations based on a set of Landau–Khalatnikov type equations. Using the basic assumption that the smectic layers remain fixed, a general elastic-hydrodynamic theory of the SmC* phase, formally derived in the language of rational mechanics, has been put forward by Leslie *et al.* [5]. This theory demonstrates clearly how a reorientation of the *c*-director couples to a macroscopic mass flow and *vice versa*. By reformulating this theory, Carlsson *et al.* [6] showed how one, in a simple way, can achieve physical insight into this more general dynamical behaviour of the SmC* phase. All theoretical models of the dynamics of the SmC* phase referred to above, assume that the smectic layer normal is fixed, although it is easily seen that most motion in a SmC* system is associated with torques acting to rotate the smectic layers [6]. Recently, it has also been experimentally observed that in some circumstances the smectic layers start rotating as a consequence of a molecular reorientation in the system [7–11]. A theoretical model explaining the mechanism of layer rotations in the SmA* phase has been put forward by Carlsson and Osipov [12], while the generalization of the theory to the SmC* phase is currently in progress [13].

In this work we consider a SmC* system for which the smectic layers are assumed to be so strongly oriented by the substrates, that they remain fixed irrespective of which torques are applied to them. The behaviour of the system is analysed theoretically using the model devised by Carlsson *et al.* [6]. This model demands 20 viscosity coefficients and 9 elastic constants to be specified in the most general case. A relevant question in this context is how to design experiments in order to determine these constants experimentally. Various experiments have been proposed (and performed) for this purpose. Among these are the study of Fréedericksz transitions in different geometries [14, 15]. By incorporating the possibility of backflow, i.e. the fact that most reorientations of the *c*-director inevitably induce a macroscopic flow in the system, not only the elastic constants but also some relevant viscosity coefficients should be measurable. Such calculations have been published by Carlsson *et al.* [16, 17] who have demonstrated how backflow affects the switching behaviour of surface stabilized ferroelectric liquid crystal cells, not only in a quantitative way but also qualitatively.

Unfortunately, today there exists very little experimental information on the values of the viscosity coefficients of the SmC* phase. The only exception is the rotational viscosity γ_G associated with the rotational motion of the director around the smectic cone. This coefficient is related to one of the viscosity coefficients defined by Carlsson *et al.* as $\gamma_G = 2\lambda_5$ [6] and has been

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measured experimentally, essentially by two different methods: either by the measurement of the response time of a surface stabilized ferroelectric liquid crystal cell [18] or by dielectric spectroscopy [19, 20]. None of these experiments was analysed by a model incorporating the possibility of backflow. However, it has since been clearly demonstrated that essentially all rotational motion of the director around the smectic cone in the SmC* phase is expected to be associated with backflow [16, 17]. In the present work it is shown how the evaluation of the Goldstone mode dielectric constant is affected by taking backflow into account in the analysis of the experiment. It is shown that backflow effects renormalizes γ_G so that the simple relation $\gamma_G = 2\lambda_5$ is changed into a more complex one, where a number of other viscosity coefficients enter the expression for γ_G.

The outline of the paper is as follows. In § 2 we define the quantities necessary to describe the system studied, introducing coordinates and notations. We also specify the geometry of the particular dielectric experiment for which the influence of backflow on the dynamical behaviour is investigated. In § 3 the general elastic-hydrodynamic equations in the SmC* phase are summarized. The general dynamic equations governing the Goldstone mode dielectric response of the SmC* phase, taking backflow into account, are derived in § 4. By solving these equations, in § 5 it is shown how the switching equation, and accordingly the Goldstone mode rotational viscosity, is renormalized by the backflow. Finally, in § 6, some inequalities that the viscosity coefficients of the SmC* phase must fulfill are discussed, and from these it is shown that backflow effects generally accelerate the response of the system. We also derive the crucial parameter that determines how pronounced will be the backflow effects in the dielectric experiment described in the present paper.

2. Geometry of the dielectric experiment: introduction to notation and definition of coordinates

In this work a liquid crystalline sample in the bookshelf geometry is studied. Thus the smectic layers, which are assumed to consist of uniform planes with fixed orientation, are standing perpendicular to the surrounding glass plates, which are taken to be parallel to the xz-plane. The basic quantities needed to describe the system are defined in figure 1. The layer normal, taken to be parallel to the z-axis, is denoted **a**. The average orientation of the molecular long axes is tilted with respect to the layer normal, the tilt being denoted θ. A unit vector, the director **n**, is introduced in this direction, while the projection of the director into the smectic planes is described by a unit vector **c**, commonly called the c-director. In order to describe the orientation of the c-director we introduce the phase angle ϕ, which is the angle between the c-director and the x-axis, counting ϕ positive for a rotation around the positive z-axis. The spontaneous polarization **P** of a SmC* liquid crystal is confined within the smectic planes and is at right angles [21] to the c-director. We introduce a unit vector **b** according to

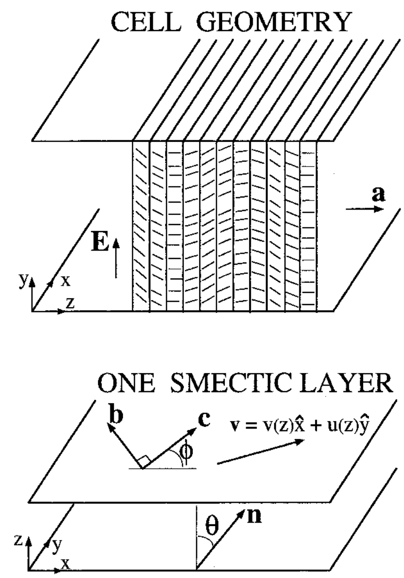

Figure 1. The geometry of the system.

$$\mathbf{b} = \mathbf{a} \times \mathbf{c} \tag{1}$$

which will coincide with the polarization vector provided that we are studying a (+) compound in the nomenclature of Clark and Lagerwall [22]. Assigning the polarization, P_o, to be positive for a (+) compound and negative for a (−) compound we can write

$$\mathbf{P} = \mathbf{P}_o \mathbf{b}. \tag{2}$$

If the sample studied is sufficiently thick, the direction of the molecular tilt precesses on going from one smectic layer to another, forming a helicoidal structure. The in-plane spontaneous polarization, being perpendicular to the tilt, also forms a helix and the macroscopic polarization of the system equals zero. Applying an electric field $\mathbf{E} = E\hat{\mathbf{z}}$ across the sample deforms the helix in two ways, changing the magnitude as well as the direction of the tilt. The dielectric response, connected to director fluctuations of the SmC* phase, therefore consists of two contributions [4, 23]. These are the soft mode part corresponding to the changes of the magnitude of the tilt, and the Goldstone mode part corresponding to changes in the tilt direction. Unless the system is very close to the SmC*–SmA* phase transition temperature, T_c, the tilt can be assumed to be constant [24] and the soft mode dielectric response will be quenched [23]. Only the case of constant tilt is studied in this work, thus assuming that the system is sufficiently far below T_c. For most compounds the condition $T_c - T \geqslant 1$ K is enough for this assumption to be valid.

Due to the deformation of the helix in the presence of the electric field a net macroscopic polarization $\langle P_i \rangle$ is induced and the corresponding dielectric response χ of the system is defined as

$$\chi = \lim_{E \to 0} \frac{\langle P_i \rangle}{E}. \tag{3}$$

Assuming θ to be constant, this quantity can be calculated [4, 19] from the switching equation [1]

$$B_3 \phi'' - P_o E \sin \phi = \gamma_G \dot{\phi}. \tag{4}$$

This approach has been used by several authors [3, 19, 20, 23, 25–28] to determine the Goldstone mode dielectric susceptibility experimentally. However, nobody has analysed how backflow effects, which are inevitably associated with director rotations in the system [16, 17], affect the analysis of the dielectric experiment. In order to do so, a velocity field $\mathbf{v}$, describing a macroscopic mass flow in the system, is introduced. Studying a sufficiently thick sample the influence from the bounding plates can be neglected and the system is assumed to be xy-invariant, i.e. the only spatial dependence all physical quantities can adopt is a z-dependence. Neglecting the possibility of transportation of matter between the smectic layers, the most general form of the velocity field can be written as $\mathbf{v} = v(z)\hat{\mathbf{x}} + u(z)\hat{\mathbf{y}}$. Thus we make the following ansatz for the quantities $\mathbf{a}$, $\mathbf{c}$, $\mathbf{b}$, $\mathbf{P}$, $\mathbf{E}$, $\mathbf{v}$ and the time derivative $\dot{\mathbf{c}}$:

$$a_x = 0, \quad a_y = 0, \quad a_z = 1 \tag{5a}$$

$$c_x = \cos \phi(z), \quad c_y = \sin \phi(z), \quad c_z = 0 \tag{5b}$$

$$b_x = -\sin \phi(z), \quad b_y = \cos \phi(z), \quad b_z = 0 \tag{5c}$$

$$P_x = -P_o \sin \phi(z), \quad P_y = P_o \cos \phi(z), \quad P_z = 0 \tag{5d}$$

$$E_x = 0, \quad E_y = E, \quad E_z = 0 \tag{5e}$$

$$v_x = v(z), \quad v_y = u(z), \quad v_z = 0 \tag{5f}$$

$$\dot{c}_x = -\dot{\phi} \sin \phi, \quad \dot{c}_y = \dot{\phi} \cos \phi, \quad \dot{c}_z = 0. \tag{5g}$$

It is the aim of the present work to show how the switching equation (4), and thus the Goldstone mode dielectric response of the system, is modified by including backflow effects through the velocity field (5f) into the analysis of the behaviour of the system.

3. Summary of the equations governing the elastic-hydrodynamic behaviour of the smectic C* phase

In this section the equations governing the elastic-hydrodynamic behaviour of the SmC* phase are summarized. The basic mathematical formulation of these equations was derived by Leslie *et al.* [5] and has been interpreted further and reformulated by Carlsson *et al.* [6, 15]. The governing equations consist of one equation for the balance of linear momentum,

$$\rho \dot{v}_i = F_i + \tilde{t}_{ij,j} \tag{6}$$

and one equation for the balance of angular momentum,

$$\Gamma_i + \varepsilon_{ijk} \tilde{t}_{kj} = 0. \tag{7}$$

In these equations, ρ is the density of the liquid crystal, F_i is the sum of all external forces and $\tilde{t}_{ij}$ is the viscous part of the stress tensor. Equation (7) can be interpreted as a balance of the torque equation in which the term $\varepsilon_{ijk}\tilde{t}_{kj}$ is the viscous torque Γ^v, and Γ_i represents the sum of all other torques acting on the system. The viscous part of the stress tensor $\tilde{t}_{ij}$ is most conveniently expressed as the sum of its symmetrical ($\tilde{t}^s_{ij}$) and antisymmetrical ($\tilde{t}^a_{ij}$) parts:

$$\tilde{t}_{ij} = \tilde{t}^s_{ij} + \tilde{t}^a_{ij}. \tag{8}$$

Adopting the usual summation convention and introducing the following quantities

$$D_{ij} = \frac{1}{2}(v_{i,j} + v_{j,i}), \quad W_{ij} = \frac{1}{2}(v_{i,j} - v_{j,i}) \tag{9}$$

$$D^a_i = D_{ij} a_j, \quad D^c_i = D_{ij} c_j \tag{10}$$

$$A_i = \dot{a}_i - W_{ik} a_k \quad C_i = \dot{c}_i - W_{ik} c_k \tag{11}$$

the viscous stress tensor can be written as

$$\begin{aligned}
\tilde{t}^s_{ij} = {} & \mu_0 D_{ij} + \mu_1 a_p D^a_p a_i a_j + \mu_2 (D^a_i a_j + D^a_j a_i) \\
& + \mu_3 c_p D^c_p c_i c_j + \mu_4 (D^c_i c_j + D^c_j c_i) \\
& + \mu_5 c_p D^a_p (a_i c_j + a_j c_i) + \lambda_1 (A_i a_j + A_j a_i) \\
& + \lambda_2 (C_i c_j + C_j c_i) + \lambda_3 c_p A_p (a_i c_j + a_j c_i) \\
& + \kappa_1 (D^a_i c_j + D^a_j c_i + D^c_i a_j + D^c_j a_i) \\
& + \kappa_2 [a_p D^a_p (a_i c_j + a_j c_i) + 2 a_p D^c_p a_i a_j] \\
& + \kappa_3 [c_p D^c_p (a_i c_j + a_j c_i) + 2 a_p D^c_p c_i c_j] \\
& + \tau_1 (C_i a_j + C_j a_i) + \tau_2 (A_i c_j + A_j c_i) \\
& + 2\tau_3 c_p A_p a_i a_j + 2\tau_4 c_p A_p c_i c_j
\end{aligned} \tag{12}$$

$$\begin{aligned}
\tilde{t}^a_{ij} = {} & \lambda_1 (D^a_j a_i - D^a_i a_j) + \lambda_2 (D^c_j c_i - D^c_i c_j) \\
& + \lambda_3 c_p D^a_p (a_i c_j - a_j c_i) + \lambda_4 (A_j a_i - A_i a_j) \\
& + \lambda_5 (C_j c_i - C_i c_j) + \lambda_6 c_p A_p (a_i c_j - a_j c_i) \\
& + \tau_1 (D^a_j c_i - D^a_i c_j) + \tau_2 (D^c_j a_i - D^c_i a_j) \\
& + \tau_3 a_p D^a_p (a_i c_j - a_j c_i) + \tau_4 c_p D^c_p (a_i c_j - a_j c_i) \\
& + \tau_5 (A_j c_i - A_i c_j + C_j a_i - C_i a_j).
\end{aligned} \tag{13}$$

These two equations define the viscosity coefficients of the system.

The torques which we will have reason to incorporate in the analysis in this work are, apart from the viscous torque Γ^{v} also the elastic torque Γ^{el}, the electric torque Γ^{ε} and the countertorque Γ^{c}, which is the torque acting on the system in order to keep the layers fixed [6, 12, 29], i.e.

$$\Gamma = \Gamma^{\mathrm{el}} + \Gamma^{\varepsilon} + \Gamma^{\mathrm{c}}. \tag{14}$$

It is possible to show that when studying the elastic-dynamic behaviour of the c-director in a system for which the smectic layers are assumed to be fixed, the z-component of the torque equation (7) is the relevant one to study [6], while the countertorque needed to stabilize the layers can be calculated by the aid of the x- and y-components of this equation. This is because the symmetry of the system implies that the most general form the countertorque can adopt is given by [6]

$$\Gamma^{\mathrm{c}} = \Gamma^{\mathrm{c}}_x \hat{\mathbf{x}} + \Gamma^{\mathrm{c}}_y \hat{\mathbf{y}} \tag{15}$$

where Γ^{c}_x and Γ^{c}_y are the x- and y-components of the countertorque, respectively.

The z-component of the elastic torque can be calculated as [6, 30]

$$\Gamma^{\mathrm{el}}_z = -\left(\frac{\partial g^{\mathrm{el}}}{\partial \phi} - \frac{\partial}{\partial x}\frac{\partial g^{\mathrm{el}}}{\partial \phi'_x} - \frac{\partial}{\partial y}\frac{\partial g^{\mathrm{el}}}{\partial \phi'_y} - \frac{\partial}{\partial z}\frac{\partial g^{\mathrm{el}}}{\partial \phi'_z}\right) \tag{16}$$

where g^{el} is the elastic free energy density which is given by [15, 31]

$$\begin{aligned} g^{\mathrm{el}} = {} & \frac{1}{2}A_{12}(\mathbf{b}\;\nabla\times\mathbf{c})^2 + \frac{1}{2}A_{21}(\mathbf{c}\;\nabla\times\mathbf{b})^2 \\ & - A_{11}\left[\frac{1}{2}(\mathbf{c}\;\nabla\times\mathbf{c} - \mathbf{b}\;\nabla\times\mathbf{b}) - \delta\right]^2 \\ & + \frac{1}{2}B_1(\nabla\;\mathbf{b})^2 + \frac{1}{2}B_2(\nabla\;\mathbf{c})^2 \\ & + \frac{1}{2}B_3\left[\frac{1}{2}(\mathbf{b}\;\nabla\times\mathbf{b} + \mathbf{c}\;\nabla\times\mathbf{c}) + q\right]^2 \\ & + B_{13}(\nabla\;\mathbf{b})\left[\frac{1}{2}(\mathbf{b}\;\nabla\times\mathbf{b} + \mathbf{c}\;\nabla\times\mathbf{c})\right] \\ & + C_1(\nabla\;\mathbf{c})(\mathbf{b}\;\nabla\times\mathbf{c}) + C_2(\nabla\;\mathbf{c})(\mathbf{c}\;\nabla\times\mathbf{b}). \end{aligned} \tag{17}$$

In this expression A_i, B_i and C_i are the elastic constants of the system, q is the wave vector of the pitch, and δ is a material constant related to an inherent tendency of the smectic layers to be non-planar [31]. The unit vectors $\mathbf{b}$ and $\mathbf{c}$ are those already defined in figure 1.

Similarly, the electric torque can be calculated as

$$\Gamma^{\varepsilon}_z = -\left(\frac{\partial g^{\varepsilon}}{\partial \phi} - \frac{\partial}{\partial x}\frac{\partial g^{\varepsilon}}{\partial \phi'_x} - \frac{\partial}{\partial y}\frac{\partial g^{\varepsilon}}{\partial \phi'_y} - \frac{\partial}{\partial z}\frac{\partial g^{\varepsilon}}{\partial \phi'_z}\right) \tag{18}$$

where g^{ε} is the electric free-energy density which, including both the ferroelectric and the dielectric coupling, can be written

$$g^{\varepsilon} = -\frac{1}{2}\varepsilon_{\mathrm{a}}\varepsilon_{\mathrm{o}}(\mathbf{n}\;\mathbf{E})^2 - \mathbf{P}\;\mathbf{E} \tag{19}$$

where ε_{a} is the dielectric anisotropy of the molecules and ε_{o} is the permittivity of free space. This expression neglects the dielectric biaxiality [32] which is of no concern here as dielectric terms will be discarded anyway in our treatment of the problem.

Equations (12), (13) and (17) define 20 viscosity coefficients and 9 elastic constants. Using symmetry arguments, it has been shown by Carlsson *et al.* [6, 15] that these coefficients should be expected to exhibit the following scaling properties with respect to the tilt angle θ:

$$\begin{aligned} & \tau_1 = \bar{\tau}_1\theta, \quad \tau_2 = \bar{\tau}_2\theta, \quad \tau_3 = \bar{\tau}_3\theta, \quad \tau_5 = \bar{\tau}_5\theta, \\ & \kappa_1 = \bar{\kappa}_1\theta, \quad \kappa_2 = \bar{\kappa}_2\theta \end{aligned} \tag{20 a}$$

$$\begin{aligned} & \lambda_2 = \bar{\lambda}_2\theta^2, \quad \lambda_3 = \bar{\lambda}_3\theta^2, \quad \lambda_5 = \bar{\lambda}_5\theta^2, \quad \lambda_6 = \bar{\lambda}_6\theta^2, \\ & \mu_4 = \bar{\mu}_4\theta^2, \quad \mu_5 = \bar{\mu}_5\theta^2 \end{aligned} \tag{20 b}$$

$$\tau_4 = \bar{\tau}_4\theta^3, \quad \kappa_3 = \bar{\kappa}_3\theta^3 \tag{20 c}$$

$$\mu_3 = \bar{\mu}_3\theta^4 \tag{20 d}$$

$$\begin{aligned} & A_{12} = K + \bar{A}_{12}\theta^2, \quad A_{21} = K + \bar{A}_{21}\theta^2, \\ & A_{11} = -K + \bar{A}_{11}\theta^2 \end{aligned} \tag{21 a}$$

$$B_1 = \bar{B}_1\theta^2, \quad B_2 = \bar{B}_2\theta^2, \quad B_3 = \bar{B}_3\theta^2 \tag{21 b}$$

$$B_{13} = \bar{B}_{13}\theta^3, \quad C_1 = \bar{C}_1\theta, \quad C_2 = \bar{C}_2\theta \tag{21 c}$$

where the constants $\bar{\mu}_i$, $\bar{\lambda}_i$, $\bar{\kappa}_i$, $\bar{\tau}_i$, K, $\bar{A}_i$, $\bar{B}_i$ and $\bar{C}_i$ can be assumed to be only weakly temperature dependent. Furthermore, the coefficients μ_0, μ_1, μ_2, λ_1 and λ_4, which are those remaining in the SmA* phase, should be expected to be independent of the tilt, exhibiting only a weak temperature dependence. It should be noticed that essentially the same scaling of the viscosity coefficient given by equations (20 *a*–*d*) has also been obtained by Osipov *et al.* [33] in a calculation based on a statistical mechanical model.

The advantage in introducing this scaling of the material parameters is that one achieves a better understanding of how the governing equations, and thus the quantities calculated from these, scale with respect to θ. To make the scaling of the equations complete, one should also notice that the polarization is also tilt-dependent. As a reasonable approximation, we assume that the polarization is proportional to the tilt [34, 35], an approximation which is good except when the system is close to the transition to the SmA* phase. Thus we introduce a weakly temperature dependent quantity $\bar{P}$ according to

$$P_o = \bar{P}\theta. \tag{22}$$

4. The dynamic equations governing the Goldstone mode dielectric response with backflow

In this section, the dynamic equations governing the response of the system in the presence of an electric field, applied in the y-direction, are derived. The correct equations to study are the z-component of equation (7), governing the rotational dynamics of the c-director, and the x- and y-components of equation (6), governing the macroscopic flow of matter in the system. As was pointed out in the previous section, the x- and y-components of equation (7), for the balance of angular momentum, are not discussed in the present work because, assuming the smectic layers to be fixed, these two equations are merely of use to calculate the stabilizing countertorque needed to prevent the smectic layers from rotating [6, 12]. The z-component of the equation for the balance of linear momentum, equation (6), regulates the flow of matter in the direction perpendicular to the smectic layers. One can show using this equation [6] that, in most cases, a motion in the system induces a pressure gradient in this direction. Such a pressure gradient is the driving force of permeation of molecules between the layers. However, neglecting the possibility of the transport of material between the smectic layers, this equation is not needed here. We note also that the only spatial dependence allowed for in the present model is along the helicoidal axis, which coincides with the z-direction.

4.1. *Balance of angular momentum: derivation of the switching equation*

Equation (7), which can be considered as an equation for the balance of torques, can be expressed as

$$\Gamma^{\mathrm{el}} + \Gamma^{\varepsilon} + \Gamma^{\mathrm{c}} + \Gamma^{\mathrm{v}} = 0. \tag{23}$$

The z-component of the elastic and electric torques, Γ^{el}_z and Γ^{ε}_z, are calculated from equations (16) and (18), respectively, while the corresponding component of the viscous torque Γ^{v}_z can be calculated [6] from the antisymmetric part of the stress tensor, equation (13), according to

$$\Gamma^{\mathrm{v}}_z = \varepsilon_{zjk}\tilde{t}_{kj} = \tilde{t}_{yx} - \tilde{t}_{xy} = 2\tilde{t}^{\mathrm{a}}_{yx}. \tag{24}$$

Substituting the ansatz given in equations (5 a–g) into the expressions of the elastic (17) and electric (19) energies, and only retaining linear terms in the electric field, then the sum of these two energies is given by

$$g^{\mathrm{el}} + g^{\varepsilon} = \frac{1}{2}B_3(\phi' - q)^2 - P_o E\cos\phi \tag{25}$$

where ϕ' denotes the spatial derivative $\mathrm{d}\phi/\mathrm{d}z$. From equations (16), (18) and (25) the sum of the elastic and electric torques is now calculated as

$$\Gamma^{\mathrm{el}}_z + \Gamma^{\varepsilon}_z = B_3\phi'' - P_o E\sin\phi. \tag{26}$$

The viscous torque can be divided into two parts. These are the shearing torque Γ^{s}, being the torque acting on the director due to velocity gradients, and the rotational torque Γ^{r} which is the torque that appears whenever the director is rotating. Thus in the case studied in this work, the shearing torque corresponds to the torque proportional to the velocity gradients,

$$v' = \frac{\mathrm{d}v}{\mathrm{d}z}, \quad u' = \frac{\mathrm{d}u}{\mathrm{d}z} \tag{27}$$

while the rotational torque is the torque proportional to the time derivative of the phase of the c-director, $\dot{\phi}$. Due to the linearity of the stress tensor, Γ^{s} and Γ^{r} can be calculated separately. Starting with the rotational torque, and only retaining terms that are proportional to $\dot{\phi}$, the rotational part of the antisymmetric stress tensor is given by

$$\tilde{t}^{\mathrm{ar}}_{ij} = \lambda_5(\dot{c}_j c_i - \dot{c}_i c_j) + \tau_5(\dot{c}_j a_i - \dot{c}_i a_j). \tag{28}$$

From equations (5), (24) and (28), the z-component of the rotational torque is calculated as

$$\Gamma^{\mathrm{r}}_z = -2\lambda_5\dot{\phi}. \tag{29}$$

Thus it is clear that the viscosity associated with director rotations, often denoted as the Goldstone mode rotational viscosity, γ_{G}, can be expressed as

$$\gamma_{\mathrm{G}} = 2\lambda_5. \tag{30}$$

To confirm the entropy production of the system to be positive, it is possible to show that the following inequality must hold [6]:

$$\lambda_5 > 0. \tag{31}$$

The shearing torque is calculated from equations (9)–(11), (13) and (24) by neglecting the time derivatives $\dot{a}_i$ and $\dot{c}_i$ in equation (11). Substituting the ansatz (5) into equations (9)–(11), the only non-zero components of the quantities are

$$D_{xz} = D_{zx} = \frac{1}{2}v', \quad D_{yz} = D_{zy} = \frac{1}{2}u' \tag{32}$$

$$W_{xz} = -W_{zx} = \frac{1}{2}v', \quad W_{yz} = -W_{zy} = \frac{1}{2}u' \tag{33}$$

$$D^{a}_{x} = \frac{1}{2}v', \quad D^{a}_{y} = \frac{1}{2}u', \quad D^{c}_{z} = \frac{1}{2}v' \cos\phi + \frac{1}{2}u' \sin\phi \tag{34}$$

$$A_x = -\frac{1}{2}v', \quad A_y = -\frac{1}{2}u', \quad C_z = \frac{1}{2}v' \cos\phi + \frac{1}{2}u' \sin\phi. \tag{35}$$

The z-component of the shearing torque now takes the form

$$\Gamma^{s}_{z} = (\tau_5 - \tau_1)(u' \cos\phi - v' \sin\phi). \tag{36}$$

The switching equation is now obtained by adding the torques given by equations (26), (29) and (36), putting the sum equal to zero,

$$B_3\phi'' - P_o E \sin\phi = 2\lambda_5\dot{\phi} + (\tau_5 - \tau_1)(v' \sin\phi - u' \cos\phi). \tag{37}$$

Comparing this equation with equation (4) one notices that backflow effects introduce the additional term proportional to $(\tau_5 - \tau_1)$ on the right hand side of the switching equation. To investigate how this term affects the solution of this equation, we have to study the balance law of linear momentum, equation (6), in order to obtain equations governing the quantities $v(z)$ and $u(z)$.

4.2. *Balance of linear momentum*

The equation for the balance of linear momentum is now derived. Neglecting the inertia of the system and assuming that no external body forces F_i are present, the x- and y-components of equation (6) reduce to

$$\tilde{t}_{xz,z} = 0, \quad \tilde{t}_{yz,z} = 0 \tag{38}$$

if the only spatial dependence allowed for in the model is a z-dependence. Equation (38) can be integrated to read

$$\tilde{t}_{xz} = \tau_x, \quad \tilde{t}_{yz} = \tau_y \tag{39}$$

where the integration constants τ_x and τ_y represent the force per unit area exerted on the glass plates surrounding the sample. Assuming this force to be negligible, equation (39) reduce to

$$\tilde{t}_{xz} = 0, \quad \tilde{t}_{yz} = 0. \tag{40}$$

In the same manner as described for the torque equation, the stress tensor can be divided into one rotational part and one shearing part. However, in the treatment of linear momentum not only the antisymmetric part, but also the full stress tensor needs to be considered. The rotational part, $\tilde{t}^{r}_{ij}$, can be expressed as

$$\begin{aligned}\tilde{t}^{r}_{ij} = {} & \lambda_2(\dot{c}_i c_j + \dot{c}_j c_i) + \tau_1(\dot{c}_i a_j + \dot{c}_j a_i) + \lambda_5(\dot{c}_j c_i - \dot{c}_i c_j) \\ & + \tau_5(\dot{c}_j a_i - \dot{c}_i a_j)\end{aligned} \tag{41}$$

where only terms that are proportional to $\dot{\phi}$ have been retained. Substituting the ansatz given in (5) into equation (41), the two components of the stress tensor of interest here, are calculated as

$$\tilde{t}^{r}_{xz} = (\tau_5 - \tau_1)\dot{\phi} \sin\phi \tag{42 a}$$

$$\tilde{t}^{r}_{yz} = -(\tau_5 - \tau_1)\dot{\phi} \cos\phi. \tag{42 b}$$

In order to calculate the shearing part of the stress tensor, equations (32)–(35) are substituted into equations (12) and (13):

$$\begin{aligned}\tilde{t}^{s}_{xz} = {} & \frac{1}{2}v'[\mu_o + \mu_2 - 2\lambda_1 + \lambda_4 \\ & + (\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6)\cos^2\phi] \\ & + \frac{1}{2}u'(\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6)\sin\phi\cos\phi\end{aligned} \tag{43 a}$$

$$\begin{aligned}\tilde{t}^{s}_{yz} = {} & \frac{1}{2}v'(\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6)\sin\phi\cos\phi \\ & + \frac{1}{2}u'[\mu_o + \mu_2 - 2\lambda_1 + \lambda_4 \\ & + (\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6)\sin^2\phi].\end{aligned} \tag{43 b}$$

From equations (40), (42) and (43), the final balance law for linear momentum is derived:

$$\begin{aligned} & v'[\mu_o + \mu_2 - 2\lambda_1 + \lambda_4 \\ & \quad + (\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6)\cos^2\phi] \\ & \quad + u'(\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6)\sin\phi\cos\phi \\ & \quad + 2(\tau_5 - \tau_1)\dot{\phi}\sin\phi = 0\end{aligned} \tag{44 a}$$

$$\begin{aligned} & v'(\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6)\sin\phi\cos\phi \\ & \quad + u'[\mu_o + \mu_2 - 2\lambda_1 + \lambda_4 \\ & \quad + (\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6)\sin^2\phi] \\ & \quad - 2(\tau_5 - \tau_1)\dot{\phi}\cos\phi = 0.\end{aligned} \tag{44 b}$$

The dynamic behaviour of the system studied is now given by equations (37) and (44).

5. Solution of the dynamic equations: renormalization of the switching equation

Equations (37) and (44) represent the set of equations governing the dynamic behaviour of the system when backflow effects have been taken into account. By comparing equations (37) and (4) one notices that backflow effects manifest themselves through the presence of the term $(\tau_5 - \tau_1)(v' \sin \phi - u' \cos \phi)$ on the right hand side of the switching equation. In order to perform a quantitative investigation of how backflow effects influence the rotational motion of the system, we rewrite this term by the aid of equations (44). Introducing the abbreviations

$$\tau_o = (\tau_5 - \tau_1) \tag{45 a}$$

$$\eta_A = \frac{1}{2}(\mu_o + \mu_2 - 2\lambda_1 + \lambda_4) \tag{45 b}$$

$$\eta_C = \frac{1}{2}(\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6) \tag{45 c}$$

equations (44) can then be written in a more condensed form:

$$v'(\eta_A + \eta_C \cos^2 \phi) + u'\eta_C \sin \phi \cos \phi + \tau\dot{\phi} \sin \phi = 0 \tag{46 a}$$

$$v'\eta_C \sin \phi \cos \phi + u'(\eta_A + \eta_C \sin^2 \phi) - \tau\dot{\phi} \cos \phi = 0. \tag{46 b}$$

From these two equations v' and u' can be expressed as

$$v' = -\frac{\tau}{\eta_A}\dot{\phi} \sin \phi \tag{47 a}$$

$$u' = \frac{\tau}{\eta_A}\dot{\phi} \cos \phi. \tag{47 b}$$

Substituting equation (47) into equation (37), the final form of the renormalized switching equation is obtained:

$$B_3\phi'' - P_o E \sin \phi = \left(2\lambda_5 - \frac{\tau_o^2}{\eta_A}\right)\dot{\phi}. \tag{48}$$

Comparing this equation with equation (4), one concludes that backflow effects influence the rotational motion of the director in such a way that the rotational viscosity γ_G is renormalized into an effective rotational viscosity according to

$$\gamma_G^{\text{eff}} = 2\lambda_5 - \frac{\tau_o^2}{\eta_A}. \tag{49}$$

6. Discussion

In this work the role of backflow effects in the switching dynamics of a ferroelectric SmC* liquid crystalline cell has been theoretically investigated. The geometry of the system studied is depicted in figure 1. If the sample studied is sufficiently thin, and the director is strongly anchored to the substrates, the system represents a surface stabilized ferroelectric liquid crystal cell [36]. For such a system, the role of backflow has been discussed elsewhere [16, 17]. If, on the other hand, we study a sufficiently thick sample for which the influence of the substrates on the anchoring of the director can be neglected, the situation models the experiment determining the Goldstone mode dielectric susceptibility. Here the macroscopic flow is expected to be a two-dimensional velocity field according to the ansatz given by equation (5f). This is the case studied in this work.

The key result of the present work is the renormalization of the switching equation which, in its final form, can be expressed by equation (48). From this equation one notices that the qualitative behaviour of the dielectric response of the system is unaffected by the backflow. However, the Goldstone mode rotational viscosity, regulating the relaxation frequency of the system, is renormalized accordingly. Thus we can define an effective Goldstone mode rotational viscosity according to equation (49). It can be proven [6] by the study of entropy production that the viscosity coefficient λ_5 must be positive. Concerning the two viscosity coefficients τ_o and η_A, which enter the renormalization factor, it is possible to show [6] that these must also be strictly positive for a system composed of molecules of rod-like symmetry. The inequalities relevant to the present study can now be summarized as

$$\lambda_5 > 0, \quad \tau_o > 0, \quad \eta_A > 0, \quad |\eta_C| < \eta_A \tag{50}$$

and we can conclude that the renormalization of γ_G is such that γ_G^{eff} always decreases due to the backflow. Thus the corresponding relaxation frequency [4, 19]

$$f_G = \frac{B_3 q^2}{2\pi\gamma_G} \tag{51}$$

always increases when backflow is present in the system. The conclusion that backflow effects speed up the response of the system is consistent with the results from the calculations by Carlsson *et al.* [16, 17] where it is shown how backflow decreases the response time of the switching in a surface stabilized liquid crystal cell.

In equations (20) the scaling of the material parameters with respect to the tilt is given. Using this scaling, one can rewrite the expression for γ_G^{eff} as

$$\gamma_G^{\text{eff}} = \left[2\bar{\lambda}_5 - \frac{(\bar{\tau}_5 - \bar{\tau}_1)^2}{\eta_A}\right]\theta^2. \tag{52}$$

It is the quantity within the parenthesis that corresponds to the physical viscosity of the rotational motion of the system [37], while the factor θ^2 is merely a geometrical factor present because the motion along the smectic cone degenerates towards one point when the system approaches the SmC*–SmA* phase transition. In conclusion, in the present work we have shown how backflow effects influence the behaviour of a SmC* liquid crystalline system in a dielectric experiment. One should observe that, when the tilt dependence of the rotational viscosity has been taken into account, the viscosity which is measured in the experiment is not $2\bar{\lambda}_5$ but the renormalized viscosity $2\bar{\lambda}_5 - (\bar{\tau}_5 - \bar{\tau}_1)^2/\eta_A$. When comparing this result with results obtained by other experimental techniques, one must be aware that different geometrical arrangements generally permit different types of backflow and accordingly are associated with different renormalizations of the rotational viscosity.

The answer to the question of how pronounced backflow effects are in a real experiment is of course given by how large the renormalization of γ_G is, and can be quantified by defining the parameter κ according to

$$\kappa = \frac{(\bar{\tau}_5 - \bar{\tau}_1)^2}{2\bar{\lambda}_5 \eta_A} < 1 \tag{53}$$

where the limit of the magnitude of κ comes from the fact that we should never expect the system to become infinitely fast. Using equations (52) and (53) one can now write γ_G^{eff} as

$$\gamma_G^{\mathrm{eff}} = 2\bar{\lambda}_5(1 - \kappa)\theta^2 \tag{54}$$

and it is easy to see that whenever the quantity κ is small, backflow effects should be expected to be negligible. Unfortunately, to date rather little experimental information about the values of the viscosity coefficients of the SmC* phase exists, so we can make no statements regarding the experimental value of κ. However it is clear from other experiments [38] that the coupling between director reorientations and macroscopic flow in the SmC* liquid crystalline systems can be important. Thus one of the urgent tasks to be undertaken in current reasearch concerning the dynamical behaviour of the SmC* phase is to determine the viscosity coefficients of this system experimentally.

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