

## Macroscopic theory for the flow behavior of smectic-C and smectic-C\* liquid crystals

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Equations governing the flow behavior of Sm-C and Sm-C\* liquid crystals are derived. It is shown that the general theory for the description of the macroscopic flow properties of the system demands 20 viscosity coefficients. Using thermodynamic arguments, one can show that these coefficients must fulfill certain restrictions in terms of inequalities and also, from symmetry arguments, that a specific tilt angle dependence can be assigned to them. With the Sm-C stress tensor as a starting point we discuss the concept of viscous torques. We define the rotational viscosity of the system and write down the proper equations governing the shear flow behavior for two different geometries. Using these equations we discuss the concept of flow alignment and define the effective viscosities of the system. We also show that in most cases backflow (flow induced by director rotations), as well as transverse flow effects (flow or permeation perpendicular to the shear plane induced by the shear), is associated with the macroscopic flow.

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### I. INTRODUCTION

The discovery of the possibility of developing electro-optical devices using ferroelectric smectic-C\* liquid crystals [1] has necessitated the need for a complete viscoelastic theory for this system. An elastic theory for Sm-C and Sm-C\* liquid crystalline systems has already been given by de Gennes [2]. This theory was later refined and reformulated by several authors [3–6]. However, apart from the paper by Martin, Parodi, and Pershan [7] providing a microscopic hydrodynamic theory of Sm-C liquid crystals, papers concerning macroscopic flow are very scarce in the literature [8,9]. Still, dynamical problems for Sm-C liquid crystals are most often treated by the use of some “nematiclike” theory and not by a proper smectic theory (for convenience we write Sm-C throughout this paper, although most of the results derived are also applicable to Sm-C\* liquid crystals). The aim of this paper is to provide such a theory. With the paper by Leslie, Stewart, and Nakagawa [8] as a starting point, we show how one can develop a coherent and physically sound picture of the macroscopic flow properties of Sm-C liquid crystalline systems.

The outline of the paper is as follows. In Sec. II we define the quantities necessary to describe the system studied and also introduce the notations adopted in this work. In Sec. III we then give a brief derivation and a summary of the equations governing the flow behavior of Sm-C liquid crystals, showing that the symmetry of the system demands in the stress tensor 20 viscosity coefficients as well as 9 elastic constants. Section IV presents a classification of these coefficients and here it is also proved that the form of the stress tensor implies that

a specific tilt-angle dependence can be assigned to each of these. In this work we restrict our study to the case for which the smectic planes are assumed to remain fixed. In order to fulfill this requirement, a torque of constraint must be transmitted to the director via the smectic layers. In Sec. V we discuss the way in which the symmetry of the system affects the form that this countertorque can adopt.

Sections VI and VII introduce the concept of a viscous torque and show how this can be divided into two parts: the rotational torque connected to the rotation of the  $c$  director and the shearing torque, which is due to the macroscopic flow of the system. In these two sections we define the rotational viscosity of the system and also discuss the possibility of flow alignment for two different flow geometries. Sections VIII and IX derive the expressions of the torques that are due to elasticity and the application of electric or magnetic fields, respectively. Section X examines the balance of linear momentum and introduces the effective viscosities for the two flow geometries discussed. In this section it is shown that the phenomenon of backflow is most likely to appear in the system studied. In Sec. XII we show that the form of the stress tensor implies that in most cases either a transverse flow or a permeation of molecules between the smectic layers is inevitably associated with the macroscopic flow of the system.

Finally, in Sec. XIII a comparison between the Sm-C and the nematic stress tensors is made. From this comparison we draw conclusions regarding some of the viscosity coefficients entering the Sm-C theory. This section also summarizes what can be stated concerning the Sm-C viscosity coefficients from a theoretical point of view.

## II. SMECTIC-C LIQUID CRYSTALS: DEFINITIONS AND NOTATIONS

Figure 1 introduces the basic quantities needed to describe a Sm-C liquid crystal and sets the corresponding notations unambiguously. We describe the normal to the smectic layers by a unit vector  $\mathbf{a}$ . Assuming that the system studied is free from dislocations and of constant layer thickness, the layer normal  $\mathbf{a}$  must fulfill [10] the constraint

$$\nabla \times \mathbf{a} = 0. \quad (2.1)$$

In this work we consider only systems for which the smectic layers are planar with an orientation constant in time. We choose the coordinate system in such a way that the smectic planes are parallel to the  $xy$  plane and the layer normal falls along the positive  $z$  axis, i.e.,  $\mathbf{a} = \hat{\mathbf{z}}$ , and thus the constraint (2.1) is always automatically fulfilled. The direction of the long molecular axes is defined by a unit vector  $\mathbf{n}$ , the director. The angle that the director makes with the layer normal, "the tilt angle," is denoted by  $\theta$ . Assuming that the system is not studied in the immediate vicinity of the Sm-C-Sm-A phase transition temperature  $T_c$ , one can assume  $\theta$  to be constant [11], depending solely on the temperature of the system. The projection of the director onto the smectic planes is described by a unit vector  $\mathbf{c}$ , commonly denoted the  $c$  director. In order to describe the orientation of the  $c$  director one introduces the phase angle  $\phi$ , which is the angle between the  $c$  director and the  $x$  axis, counting  $\phi$  positive for a rotation of the  $c$  director around the positive  $z$  axis. For mathematical convenience we also introduce a third unit vector

$$\mathbf{b} = \mathbf{a} \times \mathbf{c}. \quad (2.2)$$

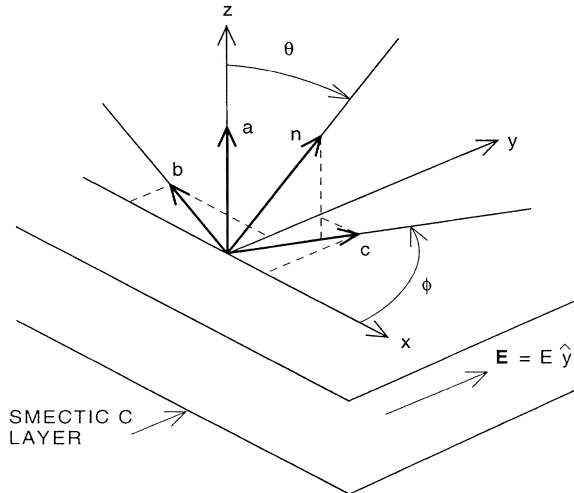


FIG. 1. Notations used in the present work. The average molecular direction, the director, is given by a unit vector  $\mathbf{n}$  making an angle  $\theta$  with the layer normal  $\mathbf{a}$ , which is taken to be parallel to the  $z$  axis. The  $c$  director, being a unit vector parallel to the projection of the director onto the smectic planes (the  $xy$  plane), is denoted by  $\mathbf{c}$  and is described by the phase angle  $\phi$ . The unit vector  $\mathbf{b}$ , which is also confined to lie within the smectic planes, is defined by the relation  $\mathbf{b} = \mathbf{a} \times \mathbf{c}$ .

For a chiral, ferroelectric Sm-C\* liquid crystal of the (+) type in the nomenclature of Clark and Lagerwall [12], the unit vector  $\mathbf{b}$  coincides with the polarization vector. We also occasionally study the influence of applying an electric field over the system. When doing so, the field is assumed to be applied parallel to the smectic layers, pointing in the  $y$  direction.

When studying the flow properties of the system, the liquid crystal is considered to be incompressible and the velocity field  $\mathbf{v}$  is subject to the constraint

$$\nabla \cdot \mathbf{v} = 0. \quad (2.3)$$

Furthermore, we neglect the possibility of transportation of material between the smectic layers, thus assuming the velocity field everywhere to be parallel to the smectic layers.

To solve the general viscoelastic problem one now has, for a given set of boundary conditions and external forces, to calculate the space and time dependence of the layer normal, the  $c$  director, and the velocity field. In the particular case studied in this work (an incompressible, isothermic system with constant tilt and fixed, planar smectic layers), we thus want to calculate a one-component  $c$ -director field  $\phi(\mathbf{r}, t)$  and a two-component velocity field  $\mathbf{v}(\mathbf{r}, t) = v_x(\mathbf{r}, t)\hat{\mathbf{x}} + v_y(\mathbf{r}, t)\hat{\mathbf{y}}$  for a given set of external conditions. Demanding the smectic layers to remain planar and fixed in space requires some external torque to be transmitted to the system. This counter-torque will be obtained as a by-product of the calculations.

## III. GOVERNING EQUATIONS: THE SMECTIC-C STRESS TENSOR

This section presents a brief summary of the continuum theory proposed recently by Leslie, Stewart, and Nakagawa [8] for certain equilibrium and dynamic problems in Sm-C liquid crystals. In addition to the normal assumption of incompressibility, their model is further constrained in that it assumes that the layer thickness remains constant and also that the tilt of the director with respect to the layer normal remains fixed. These assumptions appear to be reasonable for many problems, although clearly too restrictive for others.

Given the assumption of incompressibility, their continuum theory essentially rests on two balance laws for linear and angular momentum, namely,

$$\rho \dot{v}_i = F_i + t_{ij,j}, \quad (3.1)$$

$$\Gamma_i^{\text{ext}} + \varepsilon_{ijk} t_{kj} + l_{ij,j} = 0, \quad (3.2)$$

where  $\mathbf{F}$  and  $\mathbf{\Gamma}$  denote external body force and moment per unit volume and  $t_{ij}$  and  $l_{ij}$  the stress and the couple stress tensors, respectively. Given the constraints, the latter can be expressed as

$$t_{ij} = -p \delta_{ij} + \beta_p \varepsilon_{pjk} a_{k,i} - \frac{\partial w}{\partial a_{k,j}} a_{k,i} - \frac{\partial w}{\partial c_{k,j}} c_{k,i} + \tilde{t}_{ij}, \quad (3.3)$$

$$l_{ij} = \beta_p a_p \delta_{ij} - \beta_i a_j + \varepsilon_{ipq} \left[ a_p \frac{\partial w}{\partial a_{q,j}} + c_p \frac{\partial w}{\partial c_{q,j}} \right] + \tilde{l}_{ij}, \quad (3.4)$$

the pressure  $p$  arising from the assumed incompressibility and the vector  $\beta$  stemming from the layer constraint (3.1). Furthermore,  $\tilde{\tau}_{ij}$  and  $\tilde{l}_{ij}$  denote dynamic contributions, while  $w$  is the elastic energy of the system given by [5]

$$w = \frac{1}{2}A_{12}(\mathbf{b} \cdot \nabla \times \mathbf{c})^2 + \frac{1}{2}A_{21}(\mathbf{c} \cdot \nabla \times \mathbf{b})^2 + A_{11}(\mathbf{b} \cdot \nabla \times \mathbf{c})(\mathbf{c} \cdot \nabla \times \mathbf{b}) + \frac{1}{2}B_1(\nabla \cdot \mathbf{b})^2 + \frac{1}{2}B_2(\nabla \cdot \mathbf{c})^2 + \frac{1}{2}B_3[\frac{1}{2}(\mathbf{b} \cdot \nabla \times \mathbf{b} + \mathbf{c} \cdot \nabla \times \mathbf{c}) + q]^2 + B_{13}(\nabla \cdot \mathbf{b})[\frac{1}{2}(\mathbf{b} \cdot \nabla \times \mathbf{b} + \mathbf{c} \cdot \nabla \times \mathbf{c})] + C_1(\nabla \cdot \mathbf{c})(\mathbf{b} \cdot \nabla \times \mathbf{c}) + C_2(\nabla \cdot \mathbf{c})(\mathbf{c} \cdot \nabla \times \mathbf{b}), \quad (3.5)$$

$q$  being the wave vector of the pitch in the case we are studying a Sm-C\* system. It can be also shown [8] that

$$\tilde{l}_{ij} = 0, \quad (3.6)$$

analogous to the case for a nematic liquid crystal.

To present the constitutive equations for the viscous stress it is helpful to introduce initially the rate of strain and vorticity tensors

$$D_{ij} = \frac{1}{2}(v_{i,j} + v_{j,i}), \quad (3.7)$$

$$W_{ij} = \frac{1}{2}(v_{i,j} - v_{j,i}) \quad (3.8)$$

and two vectors  $\mathbf{A}$  and  $\mathbf{C}$  related to the material time derivatives of the unit vectors  $\mathbf{a}$  and  $\mathbf{c}$  by

$$A_i = \dot{a}_i - W_{ik}a_k, \quad (3.9)$$

$$C_i = \dot{c}_i - W_{ik}c_k. \quad (3.10)$$

Also we find it convenient to employ the notations

$$D_i^a = D_{ij}a_j, \quad (3.11)$$

$$D_i^c = D_{ij}c_j. \quad (3.12)$$

With these definitions Leslie, Stewart, and Nakagawa [8] write

$$\tilde{\tau}_{ij} = \tilde{\tau}_{ij}^s + \tilde{\tau}_{ij}^a, \quad (3.13)$$

where

$$\begin{aligned} \tilde{\tau}_{ij}^s = & \mu_0 D_{ij} + \mu_1 a_p D_p^a a_i a_j + \mu_2 (D_i^a a_j + D_j^a a_i) + \mu_3 c_p D_p^c c_i c_j + \mu_4 (D_i^c c_j + D_j^c c_i) + \mu_5 c_p D_p^a (a_i c_j + a_j c_i) \\ & + \lambda_1 (A_i a_j + A_j a_i) + \lambda_2 (C_i c_j + C_j c_i) + \lambda_3 c_p A_p (a_i c_j + a_j c_i) + \kappa_1 (D_i^a c_j + D_j^a c_i + D_i^c a_j + D_j^c a_i) \\ & + \kappa_2 [a_p D_p^a (a_i c_j + a_j c_i) + 2a_p D_p^c a_i a_j] + \kappa_3 [c_p D_p^c (a_i c_j + a_j c_i) + 2a_p D_p^c c_i c_j] + \tau_1 (C_i a_j + C_j a_i) \\ & + \tau_2 (A_i c_j + A_j c_i) + 2\tau_3 c_p A_p a_i a_j + 2\tau_4 c_p A_p c_i c_j \end{aligned} \quad (3.14)$$

is the symmetric part of the stress tensor and

$$\begin{aligned} \tilde{\tau}_{ij}^a = & \lambda_1 (D_j^a a_i - D_i^a a_j) + \lambda_2 (D_j^c c_i - D_i^c c_j) + \lambda_3 c_p D_p^a (a_i c_j - a_j c_i) + \lambda_4 (A_j a_i - A_i a_j) \\ & + \lambda_5 (C_j c_i - C_i c_j) + \lambda_6 c_p A_p (a_i c_j - a_j c_i) + \tau_1 (D_j^a c_i - D_i^a c_j) + \tau_2 (D_j^c a_i - D_i^c a_j) \\ & + \tau_3 a_p D_p^a (a_i c_j - a_j c_i) + \tau_4 c_p D_p^c (a_i c_j - a_j c_i) + \tau_5 (A_j c_i - A_i c_j + C_j a_i - C_i a_j) \end{aligned} \quad (3.15)$$

is the antisymmetric part. Associated with the latter one can introduce two vectors  $\tilde{\mathbf{g}}^a$  and  $\tilde{\mathbf{g}}^c$  such that

$$\varepsilon_{ijk} \tilde{\tau}_{kj} = \varepsilon_{ijk} a_j \tilde{g}_k^a + \varepsilon_{ijk} c_j \tilde{g}_k^c \quad (3.16)$$

and therefore

$$\begin{aligned} \tilde{g}_i^a = & -2(\lambda_1 D_i^a + \lambda_3 c_i c_p D_p^a + \lambda_4 A_i + \lambda_6 c_i c_p A_p \\ & + \tau_2 D_i^c + \tau_3 c_i a_p D_p^a + \tau_4 c_i c_p D_p^c + \tau_5 C_i) \end{aligned} \quad (3.17)$$

and

$$\tilde{g}_i^c = -2(\lambda_2 D_i^c + \lambda_5 C_i + \tau_1 D_i^a + \tau_5 A_i). \quad (3.18)$$

The viscous dissipation takes the form

$$T\sigma = \tilde{\tau}_{ij}^s D_{ij} - \tilde{g}_i^a A_i - \tilde{g}_i^c C_i \geq 0, \quad (3.19)$$

which limits the choice of the viscous coefficients. The above relationships are the most general isotropic expressions that are linear in the rate of strain and the two vectors  $\mathbf{A}$  and  $\mathbf{C}$ , invariant to the simultaneous change of sign of the two quantities  $\mathbf{a}$  and  $\mathbf{c}$ , and finally satisfy Onsager relations. Given the symmetry assumed, the above

theory contains 20 viscous coefficients, which may seem excessive. An alternative is to assume a different symmetry with the theory invariant to independent changes of sign in  $\mathbf{a}$  and  $\mathbf{c}$ , this reducing the number of viscous terms to 12 as in a biaxial nematic liquid crystal, the terms with coefficients  $\tau_i$  and  $\kappa_i$  being eliminated. However, we reject this option because it leads to a theory in which there would be no flow alignment in the geometry depicted in Fig. 5; this is unacceptable because such a flow alignment does occur.

A comparison of the above theory with an earlier one by Martin, Parodi, and Pershan [7] is not easy given that they are rather different both in their range of applicability and in their formulation. The present theory is nonlinear in that the layers can be deformed in an arbitrary finite manner as long as the equal spacing is maintained, with the consequent changes in alignment not restricted to small perturbations. Also it allows for nonlinear interactions between alignment, layer orientation, and flow, aspects that are essential for successful device modeling. The earlier theory [7], however, is restricted to small perturbations of a uniformly aligned smectic in planar lay-

ers, mainly motivated by an analysis of light scattering experiments, and does not include any nonlinear effects. For example, it cannot model flow alignment. Indeed, its limitation to small disturbances of planar layers has rather restricted interest in it, to the extent that very little by way of analyses based on it have appeared in the literature over the past 20 years.

The formulations also differ in that the present theory is based on principles of continuum mechanics that generalize concepts of linear and angular momentum employed in classical Newtonian mechanics and is essentially an extension of the approach adopted by Leslie [13] and Ericksen [14] in the derivation of their very successful theory for nematic liquid crystals. In contrast, the earlier linear theory appeals to principles embodying microscopic concepts and is somewhat dismissive of angular momentum. However, where the comparison is straightforward, the two theories agree. The theory by Martin, Parodi, and Pershan employs a symmetric stress tensor dependent solely upon velocity gradients and contains nine terms. If the above stress is so restricted, it too reduces to nine terms. While some reduction of the above general stress is possible in special cases by using the equations for angular momentum to eliminate certain terms, further comparison between the two theories is not a simple matter given the differences in formulation. For nematic theory it did prove possible to show that the two approaches led to equivalent theories in the linear limit [2] and ultimately this may prove possible for smectic theory, but it is rather beyond the scope of the present paper to attempt this here.

With the assumption that the smectic layers are held fixed by some external constraint torque  $\Gamma^c$  and choosing the layer normal  $\mathbf{a}$  to point in the  $z$  direction, i.e.,  $\mathbf{a}=\hat{\mathbf{z}}$ , the torque equation (3.2) can, by adopting the notations

$$\Pi_i^a = \left[ \frac{\partial w}{\partial a_{i,j}} \right]_{,j} - \frac{\partial w}{\partial a_i}, \quad (3.20a)$$

$$\Pi_i^c = \left[ \frac{\partial w}{\partial c_{i,j}} \right]_{,j} - \frac{\partial w}{\partial c_i}, \quad (3.20b)$$

be written as [8]

$$\Gamma_x^{\text{ext}} + \tilde{\tau}_{zy}^a - \tilde{\tau}_{yz}^a + \beta_{z,x} - \beta_{x,z} - \Pi_y^a + c_y \Pi_z^c = 0, \quad (3.21a)$$

$$\Gamma_y^{\text{ext}} + \tilde{\tau}_{xz}^a - \tilde{\tau}_{zx}^a + \beta_{z,y} - \beta_{y,z} + \Pi_x^a - c_x \Pi_z^c = 0, \quad (3.21b)$$

$$\Gamma_z^{\text{ext}} + \tilde{\tau}_{yx}^a - \tilde{\tau}_{xy}^a + c_x \Pi_y^c - c_y \Pi_x^c = 0. \quad (3.21c)$$

Equation (3.21) is the basic torque equation governing the motion of the system to be employed in this work. The physical interpretation of the terms in the equation is as follows. The torque  $\Gamma^{\text{ext}}$  represents the external torque applied to the system. In this work  $\Gamma^{\text{ext}}$  will represent the torque associated with applying an electric (or magnetic) field over the system. The  $\tilde{\tau}_{ij}^a$  terms represent the viscous torque  $\Gamma^v$  exerted on the system, i.e.,

$$\Gamma_x^v = \tilde{\tau}_{zy}^a - \tilde{\tau}_{yz}^a, \quad (3.22a)$$

$$\Gamma_y^v = \tilde{\tau}_{xz}^a - \tilde{\tau}_{zx}^a, \quad (3.22b)$$

$$\Gamma_z^v = \tilde{\tau}_{yx}^a - \tilde{\tau}_{xy}^a. \quad (3.22c)$$

The elastic torque is given by

$$\Gamma_x^{\text{el}} = -\Pi_y^a + c_y \Pi_z^c, \quad (3.23a)$$

$$\Gamma_y^{\text{el}} = \Pi_x^a - c_x \Pi_z^c, \quad (3.23b)$$

$$\Gamma_z^{\text{el}} = c_x \Pi_y^c - c_y \Pi_x^c. \quad (3.23c)$$

The  $\Pi$  terms, appearing in Eqs. (3.23a) and (3.23b), are present because the system for a nonuniform  $c$  director might lower the total elastic energy by bending the smectic layers. Thus a system with planar layers and a nonuniform  $c$  director can be unstable against perturbations inducing a nonuniform layer normal  $\mathbf{a}$ . Experiments involving manipulations of  $\mathbf{c}$  suggest that the layer deformations accompanying change or nonuniformity of  $\mathbf{c}$  are unobservable, i.e., that the layers provide the required torques with very little deformation. Given this, one can expect  $\Gamma_x^{\text{el}}$  and  $\Gamma_y^{\text{el}}$  to be small.

The unknown counter-torque  $\Gamma^c$ , needed to keep the layers planar and fixed in space, is given by the Lagrangian multiplier  $\beta$ ,

$$\Gamma_x^c = \beta_{z,x} - \beta_{x,z}, \quad (3.24a)$$

$$\Gamma_y^c = \beta_{z,y} - \beta_{y,z}, \quad (3.24b)$$

$$\Gamma_z^c = 0. \quad (3.24c)$$

We notice that the mathematical structure of Eqs. (3.2)–(3.4) implies that  $\Gamma_z^c=0$ , which will be of importance when solving the torque equation (3.21) later.

#### IV. TILT ANGLE DEPENDENCE AND CLASSIFICATION OF THE VISCOSITY COEFFICIENTS AND ELASTIC CONSTANTS

Equations (3.14) and (3.15) introduce altogether 20 viscosity coefficients needed to give a complete description of the dynamical properties of a Sm-C liquid crystal. We notice that these coefficients can be classified to belong to one out of four groups. This classification scheme is shown in Table I. First of all there is the  $\mu_0$  term, which does not depend upon either  $\mathbf{a}$  or  $\mathbf{c}$  and thus is the isotropic part of the stress tensor. We then find the four terms connected to the constants  $\mu_1$ ,  $\mu_2$ ,  $\lambda_1$ , and  $\lambda_4$ . These depend only on  $\mathbf{a}$  and are independent of the  $c$  director. Thus these four terms are present also in the Sm-A phase where the  $c$  director is absent and we denote them as “smectic-A-like” coefficients. The four terms mentioned above, together with the isotropic  $\mu_0$  term, are those needed to describe the dynamical properties of the Sm-A phase. The number of terms agrees with the five terms given by Martin, Parodi, and Pershan [7] in their hydrodynamic theory of the Sm-A phase. Furthermore, there are four terms in the stress tensor, which depend only on the  $c$  director. These are the terms involving the coefficients  $\lambda_2$ ,  $\lambda_5$ ,  $\mu_3$ , and  $\mu_4$ . The form of these terms correspond to the way the four independent anisotropic terms can be written in the nematic stress tensor [15] and thus we denote these terms as “nematiclike.” However, we show later (cf. Sec. XIII) that some caution must be taken when comparing these four coefficients with the four independent anisotropic Leslie viscosities of nematic

TABLE I. Classification and tilt-angle dependence of the 20 Sm-C viscosity coefficients.

Isotropic	Smectic- <i>A</i> -like	Nematiclike	Coupling	Tilt dependence
$\mu_0$	$\mu_1, \mu_2, \lambda_1, \lambda_4$	$\mu_4, \lambda_2, \lambda_5$	$\mu_5, \lambda_3, \lambda_6$	independent
		$\mu_3$		$\theta^2$
				$\theta^4$
			$\kappa_1, \kappa_2, \tau_1, \tau_2, \tau_3, \tau_5$	$\theta$
			$\kappa_3, \tau_4$	$\theta^3$

liquid crystals. This is because the nematic stress tensor is expressed by employing  $\mathbf{n}$ , while the Sm-C stress tensor is expressed in terms of  $\mathbf{c}$ . Finally, there are the 11 terms associated with the coefficients  $\lambda_3, \lambda_6, \mu_5, \kappa_1, \kappa_2, \kappa_3, \tau_1, \tau_2, \tau_3, \tau_4$ , and  $\tau_5$ . These are the coupling terms involving  $\mathbf{a}$  as well as  $\mathbf{c}$  and do not have any counterpart in any other theory.

The temperature dependence of the viscosity coefficients discussed above does generally have two contributions, one due directly to the tilt angle dependence and the other to the usual temperature dependence of condensed phase viscosities. If the system is in a not too large temperature interval below the Sm-C–Sm-A phase transition temperature  $T_c$ , we can probably neglect, as a fairly good approximation, the second of these two effects. However, as the tilt angle changes dramatically in the temperature interval of interest, we have to investigate the first of the two effects in detail. Keeping the layer normal  $\mathbf{a}$  unchanged and at the same time changing the tilt  $\theta$  to  $-\theta$  and  $\mathbf{c}$  to  $-\mathbf{c}$  provides a symmetry operation of the system. As a consequence, the stress tensor must be invariant under this operation. Thus we conclude that for all the terms in the stress tensor in which  $\mathbf{c}$  appears an odd number of times, the corresponding coefficient must be odd in  $\theta$ , while for the terms in which  $\mathbf{c}$  appears an even number of times the corresponding coefficient must be even in  $\theta$ . Furthermore, of the even coefficients,  $\mu_0, \mu_1, \mu_2, \lambda_1$ , and  $\lambda_4$  are independent of the  $\mathbf{c}$  director and should remain also in the Sm-A phase where the tilt is zero. Thus these coefficients should be  $\theta$  independent. Of the remaining seven terms that are connected to coefficients that are even in  $\theta$  ( $\lambda_2, \lambda_3, \lambda_5, \lambda_6, \mu_3, \mu_4, \mu_5$ ), the  $\mu_3$  term depends on the fourth power of  $\mathbf{c}$  while the other six depend only on the second power of  $\mathbf{c}$ . Thus we expect the  $\mu_3$  term to vanish faster than the other six terms when the system approaches  $T_c$  and we can write down the tilt angle dependence of the seven constants mentioned above as

$$\lambda_2 = \bar{\lambda}_2 \theta^2, \quad \lambda_3 = \bar{\lambda}_3 \theta^2, \quad \lambda_5 = \bar{\lambda}_5 \theta^2, \quad \lambda_6 = \bar{\lambda}_6 \theta^2, \quad (4.1a)$$

$$\mu_4 = \bar{\mu}_4 \theta^2, \quad \mu_5 = \bar{\mu}_5 \theta^2, \quad (4.1b)$$

$$\mu_3 = \bar{\mu}_3 \theta^4, \quad (4.1c)$$

where the constants  $\bar{\lambda}_i$  and  $\bar{\mu}_i$  can be assumed to be only weakly temperature dependent. Of the terms connected to the coefficients that are odd in  $\theta$  ( $\tau_1, \tau_2, \tau_3, \tau_4, \tau_5, \kappa_1, \kappa_2, \kappa_3$ ) some depend linearly on  $\mathbf{c}$  while some depend on the third power of  $\mathbf{c}$ . By the same reasoning as above we will assume that the tilt angle

dependence of these constants can be written as

$$\tau_1 = \bar{\tau}_1 \theta, \quad \tau_2 = \bar{\tau}_2 \theta, \quad \tau_3 = \bar{\tau}_3 \theta, \quad \tau_5 = \bar{\tau}_5 \theta, \quad (4.2a)$$

$$\kappa_1 = \bar{\kappa}_1 \theta, \quad \kappa_2 = \bar{\kappa}_2 \theta, \quad (4.2b)$$

$$\tau_4 = \bar{\tau}_4 \theta^3, \quad \kappa_3 = \bar{\kappa}_3 \theta^3, \quad (4.2c)$$

where again the temperature dependence of the constants  $\bar{\tau}_i$  and  $\bar{\kappa}_i$  is assumed to be weak. In Table I we have summarized the classification and the tilt angle dependence of the viscosity coefficients as introduced above.

The elastic constants introduced by Eq. (3.5) can be divided [5] into three groups. The  $A_i$  constants are related to deformations of the layer normal  $\mathbf{a}$  while the  $B_i$  constants are related to deformations of the  $\mathbf{c}$  director. The constants  $C_i$  represent cross-coupling terms appearing when  $\mathbf{a}$  and  $\mathbf{c}$  vary simultaneously. By employing the same symmetry arguments as above, Carlsson, Stewart, and Leslie [5] have shown that the tilt angle dependence of the elastic constants can be written as

$$A_{12} = K + \bar{A}_{12} \theta^2, \quad A_{21} = K + \bar{A}_{21} \theta^2, \quad (4.3a)$$

$$A_{11} = -K + \bar{A}_{11} \theta^2, \quad (4.3b)$$

$$B_1 = \bar{B}_1 \theta^2, \quad B_2 = \bar{B}_2 \theta^2, \quad B_3 = \bar{B}_3 \theta^2, \quad B_{13} = \bar{B}_{13} \theta^3, \quad (4.3c)$$

$$C_1 = \bar{C}_1 \theta, \quad C_2 = \bar{C}_2 \theta, \quad (4.3c)$$

where the constants  $K, \bar{A}_i, \bar{B}_i$ , and  $\bar{C}_i$  can be assumed to be only weakly temperature dependent.

## V. EQUATION OF MOTION FOR THE $\mathbf{c}$ DIRECTOR AND CALCULATION OF THE TORQUE OF CONSTRAINT

In this work we study the dynamics of a Sm-C system when being subject to some external driving force. The basic assumption is that the smectic layers are held fixed in space during the motion. In order to fulfill this constraint we show below that some external counter torque  $\Gamma^c$  has to be exerted on the director. To understand the nature of this counter torque, we study Fig. 2. A torque  $\Gamma = \Gamma \hat{\mathbf{z}}$  acting on the system causes a rotation  $\omega_z$  of the director around the  $\hat{\mathbf{z}}$  axis. Such a torque brings the director around the smectic cone, causing the  $\mathbf{c}$  director to rotate, but leaves the layer normal unchanged [Fig. 2(a)]. Figure 2(b) depicts what happens if an unbalanced torque  $\Gamma = \Gamma \mathbf{b}$  is acting on the director. In this case a rotation  $\omega_b$  of the director around the  $\mathbf{b}$  axis is the consequence. Such a rotation changes the tilt of the director with respect to the layer normal. Assuming the tilt to

remain constant, instead the entire smectic layer starts rotating around the  $\mathbf{b}$  axis in order to preserve the tilt. Having assumed the smectic layers to remain fixed, some external constraint countertorque  $\Gamma^c = \Gamma_b^c \mathbf{b}$  must thus be exerted on the system, balancing the torque  $\Gamma = \Gamma \mathbf{b}$ . By the same reasoning one concludes from Fig. 2(c) that any unbalanced torque  $\Gamma = \Gamma \mathbf{c}$  causes a rotation  $\omega_c$  of the layer normal around the  $\mathbf{c}$  axis in order to preserve the tilt. Thus a countertorque  $\Gamma^c = \Gamma_c^c \mathbf{c}$  is needed in order to balance any torque  $\Gamma = \Gamma \mathbf{c}$  acting on the system. In Fig. 1 the coordinates are introduced in such a way that

$$c_x = \cos\phi, \quad c_y = \sin\phi, \quad c_z = 0, \quad (5.1)$$

$$b_x = -\sin\phi, \quad b_y = \cos\phi, \quad b_z = 0 \quad (5.2)$$

and thus the most general countertorque acting on the system can be written

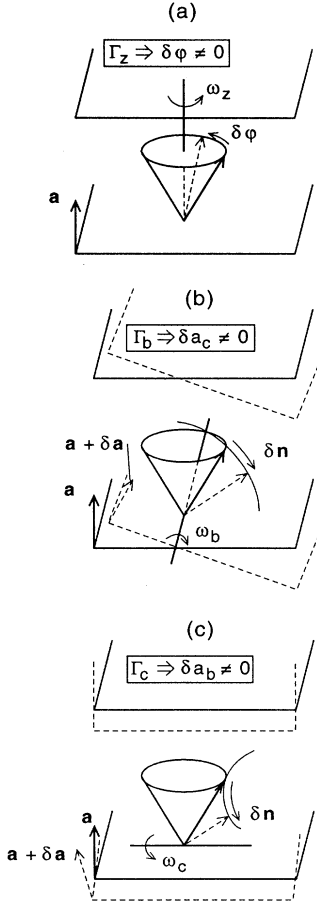


FIG. 2. Stabilizing countertorque needed to keep the smectic layers fixed in a Sm-C liquid crystal with constant tilt  $\theta$ . (a) A torque  $\Gamma_z$  acts to rotate the director around the smectic cone. (b) A torque  $\Gamma_b$ , on the other hand, will, for constant  $\theta$ , rotate the smectic layers around  $\mathbf{b}$ . (c) In the same manner a torque  $\Gamma_c$  will rotate the smectic layers around  $\mathbf{c}$ . Thus, if the smectic layers are assumed to be fixed, an external countertorque is needed to balance  $\Gamma_b$  and  $\Gamma_c$ .

$$\begin{aligned} \Gamma^c = \Gamma_b^c \mathbf{b} + \Gamma_c^c \mathbf{c} = & (\Gamma_c^c \cos\phi - \Gamma_b^c \sin\phi) \hat{\mathbf{x}} \\ & + (\Gamma_c^c \sin\phi + \Gamma_b^c \cos\phi) \hat{\mathbf{y}}. \end{aligned} \quad (5.3)$$

This form of the countertorque agrees with the result of Eq. (3.24), showing that the mathematical structure of the dynamic equation demands  $\Gamma_z^c = 0$ .

Finally, we write down the torque equation (3.21) as we use it throughout this work. The dynamics of the  $c$  director is given by Eq. (3.21c), which can be written

$$\Gamma_z^{\text{ext}} + \Gamma_z^v + \Gamma_z^{\text{el}} = 0, \quad (5.4)$$

where the three torques entering this equation are the  $z$  components of the external torque, the viscous torque [Eq. (3.22)], and the elastic torque [Eq. (3.23)], respectively. Once Eq. (5.4) is solved,  $\phi(\mathbf{r}, t)$  is known. Substituting  $\phi(\mathbf{r}, t)$  into Eqs. (3.21a) and (3.21b) allows the components of the countertorque  $\Gamma_x^c$  and  $\Gamma_y^c$ , needed to preserve the orientation of the smectic layers, to be calculated. As we prefer to express the countertorque in terms of its projections along  $\mathbf{b}$  and  $\mathbf{c}$ , we use Eqs. (5.1) and (5.2) to rewrite  $\Gamma^c$  as

$$\begin{aligned} \Gamma_b^c = & -\Gamma_x^c \sin\phi + \Gamma_y^c \cos\phi \\ = & (\Gamma_x^{\text{ext}} + \Gamma_x^v + \Gamma_x^{\text{el}}) \sin\phi - (\Gamma_y^{\text{ext}} + \Gamma_y^v + \Gamma_y^{\text{el}}) \cos\phi, \end{aligned} \quad (5.5a)$$

$$\begin{aligned} \Gamma_c^c = & \Gamma_x^c \cos\phi + \Gamma_y^c \sin\phi \\ = & -(\Gamma_x^{\text{ext}} + \Gamma_x^v + \Gamma_x^{\text{el}}) \cos\phi - (\Gamma_y^{\text{ext}} + \Gamma_y^v + \Gamma_y^{\text{el}}) \sin\phi, \end{aligned} \quad (5.5b)$$

where the last part of the equalities in the equations stems from the fact [cf. Eqs. (3.21)] that  $\Gamma_i^c = -\Gamma_i^{\text{ext}} - \Gamma_i^v - \Gamma_i^{\text{el}}$  for  $i = x, y$ . Apart from the torque equations (5.4) and (5.5), the equation for the balance of linear momentum (3.1) also needs to be solved for a complete treatment of the dynamical behavior of the system.

## VI. ROTATIONAL TORQUE AND ROTATIONAL VISCOSITIES

Neglecting the inertia of the system, the balance law for angular momentum is given by Eq. (3.2). As discussed in Secs. III and V, this equation can be interpreted as a balance of torque equation. The viscous torque  $\Gamma^v$  can be divided into two parts

$$\Gamma_i^v = \Gamma_i^s + \Gamma_i^r = \varepsilon_{ijk} \tilde{\tau}_{kj}^a. \quad (6.1)$$

In Eq. (6.1)  $\Gamma^s$  is the shearing torque, i.e., the torque acting on the director due to velocity gradients, while  $\Gamma^r$  is the rotational torque, i.e., the torque that appear whenever the director is rotating.

In this section we derive the expression for the Sm-C rotational torque and with this as a starting point define the rotational viscosity of the system. The rotational torque is given by Eq. (6.1) if in the expression of the stress tensor (3.15) one retains only the terms that remain in the absence of velocity gradients. We also reject any terms proportional to  $\dot{\mathbf{a}}$  as the system studied assumes that the smectic layers are fixed. The remaining part of

the stress tensor can in this case be written as

$$\bar{\tau}_{ij}^{ar} = \lambda_5(\dot{c}_j c_i - \dot{c}_i c_j) + \tau_5(\dot{c}_j a_i - \dot{c}_i a_j). \quad (6.2)$$

Recalling that the layer normal is assumed to point in the  $z$  direction, i.e.,  $a_z = 1$ , and employing Eq. (5.1) now allows the calculation of the rotational torque

$$\Gamma_x^r = 2\tau_5 \dot{\phi} \cos \phi, \quad (6.3a)$$

$$\Gamma_y^r = 2\tau_5 \dot{\phi} \sin \phi, \quad (6.3b)$$

$$\Gamma_z^r = -2\lambda_5 \dot{\phi}. \quad (6.3c)$$

From Eq. (6.3c) one can define the rotational viscosity  $\gamma$  associated with the motion of the  $c$  director as

$$\gamma = 2\lambda_5. \quad (6.4)$$

If the rotational torque shall be a dissipative one, the inequality

$$\lambda_5 > 0 \quad (6.5)$$

must hold, a result that also can be verified by demanding that the entropy production of the system, given by Eqs. (3.17)–(3.19),

$$T\sigma = 2\lambda_5 \dot{c}_i \dot{c}_i = 2\lambda_5 \dot{\phi}^2 \quad (6.6)$$

be positive.

Equations (6.3) give the expression of the rotational torque acting on the director when this is rotating around the smectic cone at an angular velocity  $\dot{\phi}$ . The component  $\Gamma_z^r$  is the one that balances the external driving torque maintaining the motion. However, there are also nonzero components  $\Gamma_x^r$  and  $\Gamma_y^r$ . This means that provided the smectic layers remain planar and fixed in space, an external countertorque must be exerted on the system. The rotational part of this countertorque  $\Gamma^{cr}$  is calculated from Eqs. (5.5) and (6.3) as

$$\Gamma_b^{cr} = 0, \quad (6.7a)$$

$$\Gamma_c^{cr} = -2\tau_5 \dot{\phi}. \quad (6.7b)$$

From Eq. (6.7b) we can determine the sign of the viscosity coefficients  $\tau_5$ . Let a molecule rotate along the smectic cone with an angular velocity  $\dot{\phi} > 0$ . The drag force  $\mathbf{F}^d$  acting on the molecule in this case is along  $-\mathbf{b}$ , i.e.,  $\mathbf{F}^d = -F^d \mathbf{b}$ ,  $F^d$  being proportional to  $\dot{\phi}$ . The drag torque acting on the molecule is the rotational torque, which is proportional to  $\mathbf{r} \times \mathbf{F}^d \sim (\mathbf{c} \sin \theta + \hat{\mathbf{z}} \cos \theta) \times (-\dot{\phi} \mathbf{b}) = \dot{\phi} (\mathbf{c} \cos \theta - \hat{\mathbf{z}} \sin \theta)$ . This proves the  $z$  component of the rotational torque to be negative for positive  $\dot{\phi}$ , a result already recognized above. However, one also sees that  $\Gamma_c^r > 0$  for  $\dot{\phi} > 0$ . As  $\Gamma_c^r = -\Gamma_c^{cr}$ , one concludes from Eq. (6.7b) that the following inequality must be valid:

$$\tau_5 > 0. \quad (6.8)$$

## VII. SHEARING TORQUE AND FLOW ALIGNMENT ANGLES

We now calculate the shearing torque acting on the director if velocity gradients are present in the system.

The setup of the shear flow is shown in Fig. 3. Neglecting permeation of molecules between the layers, the velocity vector must be parallel to the smectic planes, which are chosen to be parallel to the  $xy$  plane. Also neglecting the possibility of transverse flow, which will be discussed in Sec. XII, one can without loss of generality assume the velocity field to be of the form

$$v_x = v(y, z), \quad v_y = 0, \quad v_z = 0. \quad (7.1)$$

From Fig. 3 one sees that the system permits two different types of shear, either between the planes ( $dv/dz \neq 0$ ) or within the planes ( $dv/dy \neq 0$ ). We introduce the shorthand notations  $v'_z$  and  $v'_y$  for the local shear rate in these two cases, respectively. In the case of steady shear flow (i.e.,  $\dot{c}_i = 0$ ,  $\dot{a}_i = 0$ ) the nonzero components of the quantities (3.7)–(3.12) entering the stress tensor are calculated by using Eqs. (6.3) and (5.1),

$$D_{xy} = \frac{1}{2} v'_y, \quad D_{xz} = \frac{1}{2} v'_z, \quad D_{yx} = \frac{1}{2} v'_y, \quad D_{zx} = \frac{1}{2} v'_z, \quad (7.2)$$

$$W_{xy} = \frac{1}{2} v'_y, \quad W_{xz} = \frac{1}{2} v'_z, \quad W_{yx} = -\frac{1}{2} v'_y, \quad W_{zx} = -\frac{1}{2} v'_z, \quad (7.3)$$

$$D_x^a = \frac{1}{2} v'_z, \quad (7.4)$$

$$D_x^c = \frac{1}{2} \sin \phi v'_y, \quad D_y^c = \frac{1}{2} \cos \phi v'_y, \quad D_z^c = \frac{1}{2} \cos \phi v'_z, \quad (7.5)$$

$$A_x = -\frac{1}{2} v'_z, \quad (7.6)$$

$$C_x = -\frac{1}{2} \sin \phi v'_y, \quad C_y = \frac{1}{2} \cos \phi v'_y, \quad C_z = \frac{1}{2} \cos \phi v'_z. \quad (7.7)$$

The Cartesian components of the shearing torque calculated from Eqs. (3.15), (3.22), and (7.2)–(7.7) take the form

$$\Gamma_x^s = \cos \phi (\tau_2 + \tau_5 + 2\tau_4 \sin^2 \phi) v'_y + \sin \phi \cos \phi (\lambda_3 - \lambda_2 - \lambda_5 - \lambda_6) v'_z, \quad (7.8a)$$

$$\Gamma_y^s = \sin \phi (\tau_5 - \tau_2 - 2\tau_4 \cos^2 \phi) v'_y + [\lambda_4 - \lambda_1 + (\lambda_2 - \lambda_3 + \lambda_5 + \lambda_6) \cos^2 \phi] v'_z, \quad (7.8b)$$

$$\Gamma_z^s = [\lambda_2 (\sin^2 \phi - \cos^2 \phi) - \lambda_5] v'_y + \sin \phi (\tau_1 - \tau_5) v'_z. \quad (7.8c)$$

From Eqs. (5.5), (7.8a), and (7.8b) the  $b$  and the  $c$  components of the shearing part of the countertorque are now

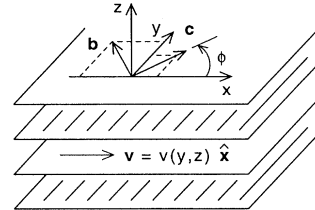


FIG. 3. Definition of coordinates for calculating the shearing torque. The smectic layers are parallel to the  $xy$  plane and the layer normal is pointing in the  $z$  direction. The velocity field is parallel to the smectic layers (here the  $x$  direction is chosen) and we assume that the velocity gradient can be both in the  $y$  direction (shear within the layers) as well as in the  $z$  direction (shear between the layers).

given by

$$\Gamma_b^{cs} = 2(\tau_2 + \tau_4)\sin\phi \cos\phi v'_y + (\lambda_1 + \lambda_3 - \lambda_2 - \lambda_4 - \lambda_5 - \lambda_6)\cos\phi v'_z, \quad (7.9a)$$

$$\Gamma_c^{cs} = [\tau_2(\sin^2\phi - \cos^2\phi) - \tau_5]v'_y + (\lambda_1 - \lambda_4)\sin\phi v'_z. \quad (7.9b)$$

From Eqs. (7.9) one concludes that generally, when a Sm-C liquid crystal is subject to shear, an external stabilizing torque has to be applied to the system in order to keep the smectic layers fixed.

With Eq. (7.8c) as a starting point, we discuss the possibility of flow alignment in the system. We treat the two cases of shear separately, starting with the case for which the shear is within the smectic planes. Such a shear is realized with a sample in the bookshelf geometry, allowing one of the glass plates to move with respect to the other along the smectic layers. This is the situation depicted in Fig. 4 provided the bounding plates are parallel to the  $xz$  plane. Setting  $v'_z = 0$  in Eq. (7.8c), the  $z$  com-

ponent of the shearing torque vanishes for an angle  $\phi_0$  given by

$$\cos 2\phi_0 = -\frac{\lambda_5}{\lambda_2}. \quad (7.10)$$

Thus flow alignment is possible whenever the relation

$$\lambda_5 < |\lambda_2| \quad (7.11)$$

is valid (we know [cf. Eq. (6.5)] that  $\lambda_5$  must be positive). In order to examine the stability of the flow alignment angles given by Eq. (7.10) one must write down the complete torque equation  $\Gamma'_z + \Gamma_z^s = 0$ . From Eqs. (6.3c) and (7.8c) one has

$$2\lambda_5\dot{\phi} + (\lambda_5 + \lambda_2 \cos 2\phi)v'_y = 0. \quad (7.12)$$

Allowing a small perturbation  $\beta$  of the  $c$  director with respect to the flow alignment angle  $\phi_0$ , i.e., by writing  $\phi = \phi_0 + \beta$ , one can expand Eq. (7.12) for small  $\beta$  and by using Eq. (7.10) write

$$\dot{\beta} = v'_y \sin 2\phi_0 \frac{\lambda_2}{\lambda_5} \beta. \quad (7.13)$$

As the quantity  $\lambda_5$  is a rotational viscosity always being positive, the stability of the flow alignment demands the quantity  $\lambda_2 \sin 2\phi_0$  to be negative. Thus there are two possibilities for the stability of the flow alignment (cf. Fig. 4).

(i)  $\lambda_2 < 0$ . In this case  $\cos 2\phi_0 > 0$  [cf. Eq. (7.10)] and the solutions for which  $\sin 2\phi_0 > 0$  are the stable ones. This means that the four solutions of  $\phi_0$  given by Eq. (7.10) occur in pairs, centered around the  $x$  axis, and the two solutions in the first and third quadrants are stable.

(ii)  $\lambda_2 > 0$ . In this case  $\cos 2\phi_0 < 0$  [cf. Eq. (7.10)] and the solutions for which  $\sin 2\phi_0 < 0$  are the stable ones. This means that the four solutions of  $\phi_0$  given by Eq. (7.10) occur in pairs, centered around the  $y$  axis, and the two solutions in the second and fourth quadrants will be stable.

Presently nothing is known about the values and signs of the Sm-C viscosity coefficients apart from some inequalities that thermodynamics require. The coefficient  $\lambda_2$  does not enter any of these inequalities, but we argue that case (i) above is the case to which the system studied most likely belongs. For nematics there is a similar situation where the stable flow alignment belongs to one of two cases depending [16] on whether the system consists of rodlike or disklike molecules. By similar reasoning we believe that the system studied, if flow alignment occurs, should belong to case (i), which is the one resembling the behavior of a rodlike nematic system exhibiting flow alignment. Thus we believe that the coefficient  $\lambda_2$  is likely to be negative. Further arguments for this opinion are presented in Sec. XIII.

We now turn attention to the case for which the shear is between the smectic planes. Such a shear is realized if the smectic layers are aligned parallel to the bounding glass plates, which thus are parallel to the  $xy$  plane using the geometry of Fig. 5. By setting  $v'_y = 0$  in Eq. (7.8c), the  $z$  component of the shearing torque vanishes if  $\phi_0 = 0$  or  $\phi_0 = \pi$ . Thus flow alignment is always possible in this

W: SHEAR WITHIN THE PLANES

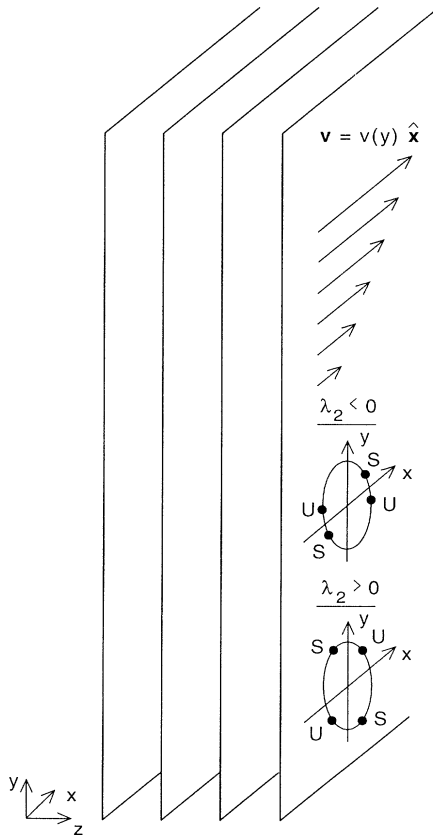


FIG. 4. Flow alignment when the shear is within the smectic planes. In this case we assume that the liquid crystal is confined between two glass plates parallel to the  $xz$  plane, one of which is moving in the  $x$  direction with respect to the other. The circles represent a top view of the smectic cone and the points denote the equilibria of the  $c$  director under the shear flow. We have denoted these points by  $S$  (stable equilibrium implies flow alignment) and  $U$  (unstable equilibrium), respectively.



## B: SHEAR BETWEEN THE PLANES

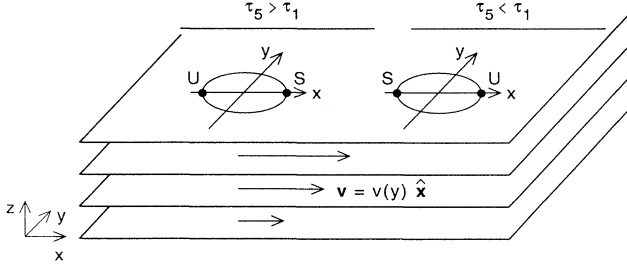


FIG. 5. Flow alignment when the shear is between the smectic planes. In this case we assume that the liquid crystal is confined between two glass plates parallel to the  $xy$  plane, one of which is moving in the  $x$  direction with respect to the other. The circles represent a top view of the smectic cone and the points denote the equilibria of the  $c$  director under the shear flow. We have denoted these points by  $S$  (stable equilibrium implies flow alignment) and  $U$  (unstable equilibrium), respectively.

case. To examine the stability of the two flow alignment angles one must write down the torque equation  $\Gamma_z^r + \Gamma_z^s = 0$ , which, using the expressions (6.3c) and (7.8c), can be reformulated as

$$\dot{\phi} = \frac{\tau_1 - \tau_5}{2\lambda_5} \sin\phi v_z' . \quad (7.14)$$

From this equation one concludes that the solution  $\phi = 0$  is stable if  $\tau_5 > \tau_1$ , while the solution  $\phi = \pi$  is stable if this inequality is reversed. Again, employing the knowledge of the behavior of rodlike nematic liquid crystals, we favor the solution  $\phi = 0$  to be the stable one and thus suggest that the inequality

$$\tau_5 > \tau_1 \quad (7.15)$$

holds.

One of the few shear flow experiments performed with a Sm-C\* liquid crystalline system reported in the literature is presented in a paper by Pieranski, Guyon, and Keller [17]. In this work it is demonstrated how shear between the layers unwinds the helix of the system due to flow alignment for which the  $c$  director becomes parallel to the flow direction. This corresponds to the solution  $\phi = 0$  in Fig. 5 and provides further support for the inequality (7.15).

## VIII. ELASTIC TORQUE

We now evaluate the expression for the elastic torque given by Eqs. (3.23). Assuming the smectic layers to be planar ( $\mathbf{a} = \hat{\mathbf{z}}$ ), only the  $B_i$  terms contribute to the elastic free-energy density (3.5) which, assuming we allow for gradients of  $\phi$  in the  $y$  and the  $z$  directions only, can be written as

$$w = \frac{1}{2} B_1 c_{x,y}^2 + \frac{1}{2} B_2 c_{y,y}^2 + \frac{1}{2} B_3 [c_y c_{x,z} - c_x c_{y,z} + q]^2 + B_{13} (c_y c_{x,z} c_{x,y} - c_x c_{y,z} c_{x,y}) . \quad (8.1)$$

Substituting the ansatz (5.1) for  $\mathbf{c}$  into this expression

gives, after some calculations, the  $z$  component of the elastic torque as

$$\Gamma_z^{\text{el}} = (B_1 \sin^2\phi + B_2 \cos^2\phi) \phi''_{yy} + \frac{1}{2} (B_1 - B_2) \sin 2\phi \phi_y'^2 + B_3 \phi''_{zz} + B_{13} (2 \sin\phi \phi''_{yz} + \cos\phi \phi_y' \phi_z') . \quad (8.2)$$

As our main objective is to study the behavior of the  $c$  director, we do not explicitly evaluate the expressions (3.23a) and (3.23b) for  $\Gamma_x^{\text{el}}$  and  $\Gamma_y^{\text{el}}$  [a calculation that must be performed with the complete expression (3.5) of the elastic free-energy density].

Wiring down the components of the elastic counter-torque along  $\mathbf{b}$  and  $\mathbf{c}$  we get, from Eqs. (3.23a), (3.23b), and (5.5),

$$\Gamma_b^{\text{cel}} = -(\Pi_x^a \cos\phi + \Pi_y^a \sin\phi) + \Pi_z^c , \quad (8.3a)$$

$$\Gamma_c^{\text{cel}} = -\Pi_x^a \sin\phi + \Pi_y^a \cos\phi . \quad (8.3b)$$

Although the elastic energy  $w$  has an implicit  $\theta$  dependence through the scaling properties (4.3b) of the elastic constants, the theory does not consider any gradients of  $\theta$ . Because of this the components  $\Gamma_b^{\text{cel}}$  and  $\Gamma_c^{\text{cel}}$  lack terms that one would expect to be present as a result of derivatives of the elastic energy with respect to  $\theta$ .

## IX. TORQUE FROM ELECTRIC AND MAGNETIC FIELDS

In this section the torque acting on the director when an electric (or magnetic) field is applied over the system is calculated. We study the case when the field is parallel to the smectic layers, assuming without loss of generality that the field is applied in the  $y$  direction, according to Fig. 1,

$$\mathbf{E} = E \hat{\mathbf{y}} . \quad (9.1)$$

If the system we study is chiral, the response to an electric field is twofold: the torque exerted on the director exhibits both a linear (ferroelectric) part and a quadratic (dielectric) part with respect to the field strength. We treat these two cases separately below.

## A. Ferroelectric torque

A Sm-C\* liquid crystal exhibits a spontaneous polarization  $\mathbf{P}$ , which for a (+) compound [12] points in the  $\mathbf{b}$  direction,  $\mathbf{b}$  being defined by Eq. (2.2),

$$\mathbf{P} = P_0 \mathbf{b} , \quad (9.2)$$

$P_0$  being the magnitude of the spontaneous polarization. The ferroelectric torque  $\Gamma^f$  due to the interaction between the field and the polarization can be written as

$$\Gamma^f = \mathbf{P} \times \mathbf{E} , \quad (9.3)$$

which by the use of Eqs. (5.2) and (9.1)–(9.3) can be written

$$\Gamma^f = -P_0 E \sin\phi \hat{\mathbf{z}} , \quad (9.4)$$

$\phi$  being the angle between the polarization vector and the field. As the polarization vector always is confined

within the smectic layers, it is obvious that an electric field, also being confined within the smectic layers, has no tendency of bringing the polarization out of these. Thus no counter torque is exerted on the system in this case, i.e., we expect  $\Gamma_x^f = \Gamma_y^f = 0$ , in accordance with what is observed from Eq. (9.4).

### B. Dielectric torque

Neglecting dielectric biaxiality, the dielectric part  $\Gamma^\epsilon$  of the torque due to the field is calculated as [2]

$$\Gamma^\epsilon = \delta \hat{\mathbf{n}} \cdot \hat{\mathbf{E}} (\hat{\mathbf{n}} \times \hat{\mathbf{E}}), \quad (9.5)$$

$\hat{\mathbf{E}}$  being a unit vector parallel to the electric field while the director  $\hat{\mathbf{n}}$  is given by

$$\hat{\mathbf{n}} = \hat{\mathbf{x}} \sin \theta \cos \phi + \hat{\mathbf{y}} \sin \theta \sin \phi + \hat{\mathbf{z}} \cos \theta, \quad (9.6)$$

and  $\delta$  is the coupling coefficient

$$\delta = \epsilon_a \epsilon_0 E^2. \quad (9.7)$$

In this expression  $\epsilon_a$  is the dielectric anisotropy of the system and  $\epsilon_0$  the dielectric permittivity of free space. From Eqs. (9.1), (9.5), and (9.6), the dielectric torque is calculated as

$$\Gamma_x^\epsilon = -\delta \sin \theta \cos \theta \sin \phi, \quad (9.8a)$$

$$\Gamma_y^\epsilon = 0, \quad (9.8b)$$

$$\Gamma_z^\epsilon = \delta \sin^2 \theta \sin \phi \cos \phi. \quad (9.8c)$$

It is obvious that in this case the direction of the director is not compatible with that of the electric field, because the equilibrium of the director is either parallel ( $\delta > 0$ ) or perpendicular ( $\delta < 0$ ) to the field. Thus a counter torque  $\Gamma^{\epsilon c}$  must be supported to the system in order to keep the smectic layers in place,

$$\Gamma_b^{\epsilon c} = -\delta \sin \theta \cos \theta \sin^2 \phi, \quad (9.9a)$$

$$\Gamma_c^{\epsilon c} = \delta \sin \theta \cos \theta \sin \phi \cos \phi. \quad (9.9b)$$

If instead a magnetic field is applied over the system,  $\delta$  has to be replaced by  $\delta^{\text{magnetic}}$ , given by

$$\delta^{\text{magnetic}} = \frac{\chi_a}{\mu_0} B^2, \quad (9.10)$$

$B$  being the magnetic field strength,  $\chi_a$  the magnetic anisotropy of the system, and  $\mu_0$  the permeability of free space. Thus the expressions derived in this section cover also the case when a magnetic field is applied to the system

### X. BALANCE OF LINEAR MOMENTUM: EFFECTIVE VISCOSITIES AND BACKFLOW

In this section we write down the equation for balance of linear momentum in shear flow. This is done for both the flow geometries discussed in the preceding section. Confining the system between two parallel glass plates, either in the bookshelf geometry or with the smectic layers parallel to the bounding plates, we now assume that one plate is moving with respect to the other under the influence of a force  $\tau$  (per unit area) acting on the moving plate. These two situations are depicted in Fig. 6. The equations obtained are used to discuss two items, the effective viscosities of the system, a discussion that provides some additional inequalities for the viscosity coefficients, and the concept of backflow. We derive the equations for the two geometries at the same time and label the equations by a **W** (shear within the planes [Fig. 6(a)],  $v'_y \neq 0$ ) and **B** (shear between the planes [Fig. 6(b)],  $v'_z \neq 0$ ), respectively. Neglecting the inertia of the system and assuming that no external body forces  $F_i$  are present, the equation for balance of linear momentum (3.1) reduces to

$$\mathbf{W}: \tilde{\tau}_{xy,y} = 0, \quad (10.1a)$$

$$\mathbf{B}: \tilde{\tau}_{xz,z} = 0. \quad (10.1b)$$

These equations can be integrated to read

$$\mathbf{W}: \tilde{\tau}_{xy} = \tau, \quad (10.2a)$$

$$\mathbf{B}: \tilde{\tau}_{xz} = \tau, \quad (10.2b)$$

where the integration constant  $\tau$  is the force per unit area applied to the moving plate. The stress tensor (3.13)–(3.15), together with Eqs. (5.1) and (7.2)–(7.7), now gives

$$\mathbf{W}: v'_y = \frac{\tau - \dot{\phi} [\lambda_5 + \lambda_2 (\cos^2 \phi - \sin^2 \phi)]}{\frac{1}{2} [\mu_0 + \mu_4 + \lambda_5 + 2\mu_3 \sin^2 \phi \cos^2 \phi + 2\lambda_2 (\cos^2 \phi - \sin^2 \phi)]}, \quad (10.3a)$$

$$\mathbf{B}: v'_z = \frac{\tau + (\tau_1 - \tau_5) \dot{\phi} \sin \phi}{\frac{1}{2} [\mu_0 + \mu_2 - 2\lambda_1 + \lambda_4 + (\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6) \cos^2 \phi]}. \quad (10.3b)$$

These equations can be written in a more condensed form

$$\mathbf{W}: v'_y = \frac{\tau - \dot{\phi} [\lambda_5 + \lambda_2 (\cos^2 \phi - \sin^2 \phi)]}{g_W(\theta, \phi)}, \quad (10.4a)$$

$$\mathbf{B}: v'_z = \frac{\tau + (\tau_1 - \tau_5) \dot{\phi} \sin \phi}{g_B(\theta, \phi)}, \quad (10.4b)$$

where  $g_W$  and  $g_B$  are implicitly  $\theta$  dependent through the  $\theta$  dependence (4.1) and (4.2) of the viscosity coefficients. If one performs a shear flow experiment, keeping the  $c$  director fixed, e.g., by applying a strong enough electric or magnetic field in a suitable direction, the quantity  $g_i(\theta, \phi)$  gives the ratio of the driving force per unit area and the corresponding shear rate. Thus this quantity

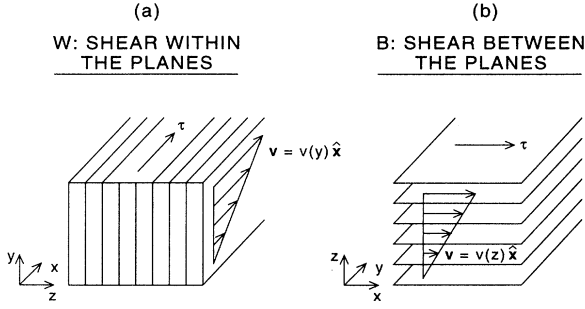


FIG. 6. Shear flow of a smectic liquid crystal (a) in the bookshelf geometry and (b) if the smectic layers are parallel to the bounding glass plates. The lower plate is at rest while the upper one is moving in the  $x$  direction under the influence of a force that has the magnitude  $\tau$  per unit area.

represents the effective viscosity of the system, which in the two cases can be written

$$\mathbf{W}: g_W(\theta, \phi) = \frac{1}{2}[\mu_0 + \mu_4 + \lambda_5 + 2\mu_3 \sin^2 \phi \cos^2 \phi + 2\lambda_2(\cos^2 \phi - \sin^2 \phi)], \quad (10.5a)$$

$$\mathbf{B}: g_B(\theta, \phi) = \frac{1}{2}[\mu_0 + \mu_2 - 2\lambda_1 + \lambda_4 + (\mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6)\cos^2 \phi]. \quad (10.5b)$$

By demanding the effective viscosity to be positive definite, it is possible to derive a number of inequalities that the viscosity coefficients must fulfill. The exact appearance of these inequalities depend on the signs of some coefficients of which we lack knowledge and for that reason the derivation of all possible inequalities is a rather involved task. Thus we are content to write down some of the more obvious inequalities that can be derived from Eqs. (10.5),

$$\mu_0 > 0, \quad (10.6a)$$

$$\mu_0 + \mu_4 + \lambda_5 - 2|\lambda_2| > 0, \quad (10.6b)$$

$$\mu_0 + \mu_4 + \lambda_5 + \frac{1}{2}\mu_3 > 0, \quad (10.6c)$$

$$\mu_0 + \mu_2 - 2\lambda_1 + \lambda_4 > 0, \quad (10.6d)$$

$$\mu_0 + \mu_2 - 2\lambda_1 + \lambda_4 + \mu_4 + \mu_5 + 2\lambda_2 - 2\lambda_3 + \lambda_5 + \lambda_6 > 0. \quad (10.6e)$$

Finally, we make a short remark concerning the phenomenon of backflow. From Eqs. (10.4) one concludes that whenever the  $c$  director is rotating, a nonzero velocity gradient develops as a consequence of this rotation. As can be seen from the structure of these equations, this coupling can never be neglected and thus a correct treatment of all dynamical problems for Sm-C liquid crystals must always take backflow into account to be complete. The influence of backflow on the switching dynamics of a ferroelectric Sm-C\* liquid crystal in the bookshelf geometry is presently being studied and the governing equations of the system has been solved analytically as well as numerically [18,19]. Of course, as long as the relevant viscosity coefficients are not known, one can-

not make any statements regarding how important the incorporation of backflow in the analysis is likely to prove to be in the end.

## XI. THE COMPLETE TORQUE EQUATIONS GOVERNING SHEAR FLOW

We now summarize the results from the previous sections by writing down the complete torque equations governing the shear flow of the system for both the flow geometries depicted in Fig. 6, still neglecting the possibility of transverse flow, the effects of which are discussed in Sec. XII. The general torque equation governing the shear flow of the system is given by  $\Gamma^c + \Gamma^r + \Gamma^s + \Gamma^{\text{el}} + \Gamma^f + \Gamma^\epsilon = 0$ . From Eqs. (6.3), (6.7), (7.8), (7.9), (8.1), (8.2), (9.4), (9.8), and (9.9) one obtains, for the case of bookshelf geometry,

$$\mathbf{W}: \Gamma_b^c = 2(\tau_2 + \tau_4)\sin\phi \cos\phi v_y' - \delta \sin\theta \cos\theta \sin^2\phi + \Gamma_b^{\text{cel}}, \quad (11.1a)$$

$$\Gamma_c^c = -2\tau_5\dot{\phi} + [\tau_2(\sin^2\phi - \cos^2\phi) - \tau_5]v_y' + \delta \sin\theta \cos\theta \sin\phi \cos\phi + \Gamma_c^{\text{cel}}, \quad (11.1b)$$

$$\begin{aligned} & -2\lambda_5\dot{\phi} + [\lambda_2(\sin^2\phi - \cos^2\phi) - \lambda_5]v_y' \\ & - P_0 E \sin\phi + \delta \sin^2\theta \sin\phi \cos\phi \\ & + (B_1 \sin^2\phi + B_2 \cos^2\phi)\phi_{yy}'' + \frac{1}{2}(B_1 - B_2)\sin 2\phi \phi_y'^2 = 0, \end{aligned} \quad (11.1c)$$

while for the case when the layers are parallel to the bounding plates open obtains

$$\mathbf{B}: \Gamma_b^c = (\lambda_1 + \lambda_3 - \lambda_2 - \lambda_4 - \lambda_5 - \lambda_6)\cos\phi v_z' - \delta \sin\theta \cos\theta \sin^2\phi + \Gamma_b^{\text{cel}}, \quad (11.2a)$$

$$\Gamma_c^c = -2\tau_5\dot{\phi} + (\lambda_1 - \lambda_4)\sin\phi v_z' + \delta \sin\theta \cos\theta \sin\phi \cos\phi + \Gamma_c^{\text{cel}}, \quad (11.2b)$$

$$\begin{aligned} & -2\lambda_5\dot{\phi} + \sin\phi(\tau_1 - \tau_5)v_z' \\ & - P_0 E \sin\phi + \delta \sin^2\theta \sin\phi \cos\phi + B_3 \phi_{zz}'' = 0, \end{aligned} \quad (11.2c)$$

where the expressions for the elastic counter-torque are given by Eqs. (8.3). As one normally is interested in the tilt angle dependence of the behavior of the system, we also employ the scaling relations (4.1)–(4.3) to obtain the scaled versions of Eq. (11.1) and (11.2):

$$\mathbf{W}: \Gamma_b^c = [2(\bar{\tau}_2 + \bar{\tau}_4\theta^2)\sin\phi \cos\phi v_y' - \delta \sin^2\phi]\theta + \Gamma_b^{\text{cel}}, \quad (11.3a)$$

$$\Gamma_c^c = \{-2\bar{\tau}_5\dot{\phi} + [\bar{\tau}_2(\sin^2\phi - \cos^2\phi) - \bar{\tau}_5]v_y' + \delta \sin\phi \cos\phi\}\theta + \Gamma_c^{\text{cel}}, \quad (11.3b)$$

$$\begin{aligned} & -2\bar{\lambda}_5\dot{\phi} + [\bar{\lambda}_2(\sin^2\phi - \cos^2\phi) - \bar{\lambda}_5]v_y' \\ & - \frac{\bar{P}E}{\theta} \sin\phi + \delta \sin\phi \cos\phi \\ & + (\bar{B}_1 \sin^2\phi + \bar{B}_2 \cos^2\phi)\phi_{yy}'' + \frac{1}{2}(\bar{B}_1 - \bar{B}_2)\sin 2\phi \phi_y'^2 = 0, \end{aligned} \quad (11.3c)$$

$$\text{B: } \Gamma_b^c = [\lambda_1 - \lambda_4 + (\bar{\lambda}_3 - \bar{\lambda}_2 - \bar{\lambda}_5 - \bar{\lambda}_6)\theta^2] \cos\phi v'_z - \delta\theta \sin^2\phi + \Gamma_b^{cel}, \quad (11.4a)$$

$$\Gamma_c^c = -2\bar{\tau}_5\theta\dot{\phi} + (\lambda_1 - \lambda_4)\sin\phi v'_z + \delta\theta \sin\phi \cos\phi + \Gamma_c^{cel}, \quad (11.4b)$$

$$-2\bar{\lambda}_5\dot{\phi} + \frac{\bar{\tau}_1 - \bar{\tau}_5}{\theta} \sin\phi v'_z - \frac{\bar{P}E}{\theta} \sin\phi + \delta \sin\phi \cos\phi + \bar{B}_3\phi''_{zz} = 0, \quad (11.4c)$$

where the scaling relations (4.3) have to be applied to the

$$\text{W: } v'_y = \frac{\tau - \dot{\phi}[\bar{\lambda}_5 + \bar{\lambda}_2(\cos^2\phi - \sin^2\phi)]\theta^2}{\frac{1}{2}\{\mu_0 + [\bar{\mu}_4 + \bar{\lambda}_5 + 2\bar{\lambda}_2(\cos^2\phi - \sin^2\phi)]\theta^2 + 2\bar{\mu}_3 \sin^2\phi \cos^2\phi\}}, \quad (11.6a)$$

$$\text{B: } v'_z = \frac{\tau + (\bar{\tau}_1 - \bar{\tau}_5)\theta\dot{\phi} \sin\phi}{\frac{1}{2}\{\mu_0 + \mu_2 - 2\lambda_1 + \lambda_4 + (\bar{\mu}_4 + \bar{\mu}_5 + 2\bar{\lambda}_2 - 2\bar{\lambda}_3 + \bar{\lambda}_5 + \bar{\lambda}_6)\theta^2 \cos^2\phi\}}. \quad (11.6b)$$

The above equations are the equations governing the shear flow of Sm-C liquid crystals under the assumption that transverse flow is negligible and that the smectic layers remain planar and fixed. These provide a straightforward way of examining the flow properties of the system. Depending upon which of the geometries of the flow is under consideration, one of the two pairs of equations (11.3c) and (11.6a) or (11.4c) and (11.6b) gives the equations to use for calculating the time and space dependence of the  $c$  director and the velocity field. Once these equations have been solved, the remaining equations (11.3a) and (11.3b) or (11.4a) and (11.4b) are immediately used to calculate the countertorque required to keep the smectic layers fixed.

## XII. TRANSVERSE FLOW EFFECTS

It is well known from the study [21] of uniaxial nematic liquid crystals that in many cases a transverse flow can be induced in the system. This section investigates under which circumstances similar effects appear in the shear flow of Sm-C liquid crystals. Starting with the case when the shear is within the smectic planes [Fig. 6(a)] it is obvious that our model does not permit transverse flow, as it only allows for a velocity field that is parallel to the smectic layers. Nevertheless, we write down the  $z$  component of the balance of linear momentum equation (3.1) to obtain

$$\bar{\tau}_{zy,y} - \frac{\partial p}{\partial z} = 0. \quad (12.1)$$

Studying the steady state (i.e.,  $\dot{\phi} = 0$ ), Eqs. (7.2)–(7.7) with  $v'_z = 0$ , together with Eqs. (3.13)–(3.15), (4.1), and (4.2), now give

$$\frac{\partial p}{\partial z} = \frac{1}{2} \frac{d}{dy} \{v'_y [\bar{\kappa}_1 + \bar{\tau}_1 + \bar{\tau}_2 + \bar{\tau}_5 + 2(\bar{\kappa}_3 + \bar{\tau}_4)\theta^2 \sin^2\phi] \theta \cos\phi\}. \quad (12.2)$$

$\Gamma_i^{cel}$  terms once these have been evaluated. In Eqs. (11.3) and (11.4) the fact that the spontaneous polarization  $P_0$  is approximately proportional to the tilt [20] is also used, i.e., we introduce a weakly temperature-dependent quantity  $\bar{P}$  according to

$$P_0 = \bar{P}\theta. \quad (11.5)$$

Each of these sets of equations is now to be solved together with the relevant of the two balance of linear momentum equations (10.3), depending upon which flow geometry is being studied. For completeness we also write down the scaled versions these equations,

This equation shows that whenever the  $c$  director is pointing in a direction other than  $\phi = \pm\pi/2$ , a transverse pressure gradient, perpendicular to the smectic layers, develops as shown in Fig. 7(a). This transverse pressure gradient is probably the driving force of permeation of molecules between the layers. However, as our model does not permit such a motion, we do not take the discussion of this effect any further.

Studying the flow geometry of Fig. 6(b), i.e., when the shear is between the layers, one must, when allowing for transverse flow, consider a velocity field  $\mathbf{v}(z)$  of the form

$$v_x = v(z), \quad v_y = u(z), \quad v_z = 0. \quad (12.3)$$

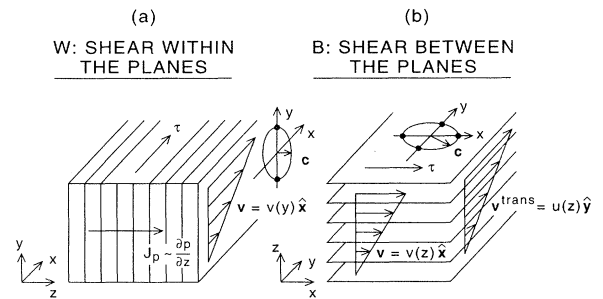


FIG. 7. (a) Transverse flow effects. When shearing the system in the bookshelf geometry a transverse pressure gradient  $\partial p/\partial z$  will generally develop. This transverse pressure gradient will force the molecules to hop between the layers and is the driving force of a permeation current  $J_p \sim \partial p/\partial z$ . The two filled circles on the smectic cone represents the positions of the  $c$  director for which permeation will not occur. In (b) the shear takes place between the layers. Here a nonzero shearing rate  $dv_x/dz$  will induce a transverse shear  $dv_y/dz$  unless the  $c$  director is either parallel or perpendicular to the primary velocity of the liquid. These positions of the  $c$  director are indicated by the filled circles on the smectic cone.

The  $x$  and the  $y$  components of Eq. (3.1) can, in the absence of external body forces, be written

$$t_{xz,z} = 0, \quad t_{yz,z} = 0. \quad (12.4)$$

By integrating these equations, assuming the driving force  $\tau$  to be applied in the  $x$  direction, one obtains

$$u'_z = v'_z \frac{2\bar{\lambda}_3 - \bar{\mu}_4 - \bar{\mu}_5 - 2\bar{\lambda}_2 - \bar{\lambda}_5 - \bar{\lambda}_6}{\mu_0 + \mu_2 - 2\lambda_1 + \lambda_4 + (\bar{\mu}_4 + \bar{\mu}_5 + 2\bar{\lambda}_2 - 2\bar{\lambda}_3 + \bar{\lambda}_5 + \bar{\lambda}_6)\theta^2 \sin^2\phi} \theta^2 \sin\phi \cos\phi. \quad (12.6)$$

We notice that the denominator of the expression on the right-hand side of this equation is an effective viscosity similar to the one defined by Eq. (10.5b). Thus this quantity is always positive definite. From Eq. (12.6) one thus concludes that whenever the  $c$  director does not point in a direction parallel or perpendicular to the moving plate, a transverse flow is inevitably induced [cf. Fig. 7(b)]. This transverse flow is associated with a torque that, if the switching dynamics of the  $c$  director is being studied, has to be added to the torque equation (11.2c). Finally, we note that for the flow geometry under consideration, the flow alignment angles are given by  $\phi=0$  and  $\pi$ . Thus these values are not affected by the presence of transverse flow. It might be possible that the stability of the flow alignment angles could be changed by taking the corresponding torque into account, but such an analysis is beyond the scope of this discussion.

### XIII. COMPARISON WITH RODLIKE UNIAXIAL NEMATICS

In Sec. III we examined the stress tensor and found that it is possible to divide the viscosity coefficients into four different categories (Table I). At the present stage, little can be stated regarding the values and even the signs of these coefficients because of the lack of experimental information. However, by thermodynamic reasoning, we were able, in Secs. VI and X, to derive

$$\begin{aligned} \bar{t}_{ij}^{\text{nematic}}(n_i) = & \alpha_4 D_{ij} + \alpha_1 n_p D_p n_i n_j + \frac{1}{2}(\alpha_5 + \alpha_6)(D_i n_j + D_j n_i) + \frac{1}{2}\gamma_1(N_j n_i - N_i n_j) \\ & + \frac{1}{2}\gamma_2(N_j n_i + N_i n_j) + \frac{1}{2}\gamma_2(D_j n_i - D_i n_j). \end{aligned} \quad (13.5)$$

Before comparing the two stress tensors (13.1) and (13.5) one must observe that while in the part of the Sm-C stress tensor studied the terms are expressed using the  $c$  director, the nematic stress tensor is expressed by using the director  $\mathbf{n}$ . Figure 8 depicts a situation in which it is assumed that the director rotates along the cone, the axis of which is taken to be  $z$  axis. The director can be either the nematic director or the director of a Sm-C liquid crystal with tilt  $\theta$ . The basic assumption is that the two stress tensors shall, as far as possible, have the same physical meaning in the two cases. We now see that if the nematic stress tensor is to be expressed in terms of  $\mathbf{c}$ , one must

$$t_{xz} = \tau, \quad t_{yz} = 0. \quad (12.5)$$

Assuming  $\dot{\phi}=0$ , the quantities given by Eqs. (7.2)–(7.7) will now include additional terms proportional to the transverse shear rate  $u'_z$ . Substituting these more general expressions into the stress tensor (3.13)–(3.15) and employing the scaling relations (4.1) and (4.2) of the viscosity coefficients, one derives from the second of Eqs. (12.5)

some inequalities that the viscosity coefficients of the Sm-C phase must fulfill. In this section we show that, by comparing the Sm-C and nematic stress tensors, one can make some additional statements regarding the nematic-like viscosity coefficients  $\bar{\mu}_3$ ,  $\bar{\mu}_4$ ,  $\bar{\lambda}_2$ , and  $\bar{\lambda}_5$ .

The nematiclike part of the Sm-C stress tensor (3.13)–(3.15), i.e., the part solely depending on the  $c$  director, is given by

$$\begin{aligned} \bar{t}_{ij}^{\text{smectic}}(c_i) = & \mu_0 D_{ij} + \mu_3 c_p D_p^c c_i c_j + \mu_4 (D_i^c c_j + D_j^c c_i) \\ & + \lambda_5 (C_j c_i - C_i c_j) + \lambda_2 (C_i c_j + C_j c_i) \\ & + \lambda_2 (D_j^c c_i - D_i^c c_j), \end{aligned} \quad (13.1)$$

where also, for completeness, the isotropic term  $\mu_0 D_{ij}$  has been retained. The expression for the nematic stress tensor [15] is

$$\begin{aligned} \bar{t}_{ij}^{\text{nematic}}(n_i) = & \alpha_1 n_k n_p D_{kp} n_i n_j + \alpha_2 N_i n_j + \alpha_3 N_j n_i + \alpha_4 D_{ij} \\ & + \alpha_5 D_{ik} n_k n_j + \alpha_6 D_{jk} n_k n_i. \end{aligned} \quad (13.2)$$

By introducing the notations

$$D_k = n_p D_{kp}, \quad \gamma_1 = \alpha_3 - \alpha_2, \quad \gamma_2 = \alpha_3 + \alpha_2 \quad (13.3)$$

and by employing the Parodi relation [22]

$$\alpha_6 - \alpha_5 = \alpha_3 + \alpha_2, \quad (13.4)$$

one can write the nematic stress tensor as

take into account that while  $\mathbf{c}$  is a unit vector, the components of  $\mathbf{n}$  when projected into the  $xy$  plane scale as  $\sin\theta$  and thus

$$n_x = c_x \sin\theta, \quad n_y = c_y \sin\theta. \quad (13.6)$$

Taking as an example the  $\gamma_1$  term of the nematic stress tensor, one can write

$$\frac{1}{2}\gamma_1(N_j n_i - N_i n_j) = \frac{1}{2}\gamma_1 \sin^2\theta (C_j c_i - C_i c_j). \quad (13.7)$$

This is compared with the corresponding term in the Sm-C stress tensor

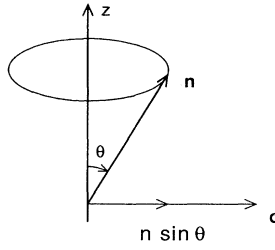


FIG. 8. Comparison between a nematic and a Sm-C liquid crystal for which the directors are moving along a cone with top angle  $2\theta$ . In the smectic case the stress tensor is expressed by  $\mathbf{c}$ , while in the nematic case  $\mathbf{n}$ , the projection of which onto the plane of the  $\mathbf{c}$  director has a magnitude  $n \sin\theta$ , is employed.

$$\lambda_5(C_j c_i - C_i c_j) = \bar{\lambda}_5 \theta^2 (C_j c_i - C_i c_j). \quad (13.8)$$

From these two expressions one thus concludes that the coefficients  $\bar{\lambda}_5$  of the Sm-C system is the one that corresponds to the coefficient  $\frac{1}{2}\gamma_1$  of nematic liquid crystals. By making the same comparison for the other coefficients, one finally arrives at the results shown in Table II.

We are now in a position to make some additional statements regarding the two nematiclike viscosity coefficients  $\bar{\lambda}_2$  and  $\bar{\lambda}_5$ . It has already been shown [Eqs. (6.5)] and  $\bar{\lambda}_5$  must be positive, a statement that corresponds to the well known nematic relation [15]  $\gamma_1 > 0$ . For a rodlike uniaxial nematic one knows [16] that  $\alpha_2$  must be negative and furthermore, in the case of flow alignment, which is by far the most common one, that also  $\alpha_3$  is negative. From Eq. (13.3) one notices that this implies  $\gamma_2 < 0$  and  $|\gamma_2| > \gamma_1$ . Thus we expect that  $\bar{\lambda}_2 < 0$  and  $|\bar{\lambda}_2| > \bar{\lambda}_5$  should be the most common, though not necessary, relations for a Sm-C system.

In Table III we summarize the inequalities for the Sm-C viscosity coefficients as derived throughout the present paper. The first two groups of inequalities in the table are based on thermodynamic arguments and must always be fulfilled. The last group, on the other hand, consists of inequalities that must be fulfilled under certain circumstances, e.g., depending on whether or not the system exhibits flow alignment. Some of the inequalities in the last group also depend on how far the comparison with uniaxial rodlike nematic liquid crystals can be taken. In

TABLE II. Comparison between the five Leslie viscosities of the nematic liquid crystals and the corresponding nematiclike viscosity coefficients of a Sm-C liquid crystal.

Smectic-C liquid crystal	Nematic liquid crystal
$\mu_0$	$\alpha_4$
$\bar{\mu}_3$	$\alpha_1$
$\bar{\mu}_4$	$\frac{1}{2}(\alpha_5 + \alpha_6)$
$\bar{\lambda}_2$	$\frac{1}{2}\gamma_2$
$\bar{\lambda}_5$	$\frac{1}{2}\gamma_1$

the end, however, experimental observations must be used to decide which of these inequalities is valid for a Sm-C liquid crystalline system.

#### XIV. DISCUSSION

In this paper we have discussed the macroscopic flow behavior of the Sm-C and the Sm-C\* phases with the stress tensor derived in Sec. III as a starting point. We have shown how the governing equations of the system consist of one torque equation—the  $b$  and the  $c$  components of which determine the external countertorque needed to keep the smectic layers fixed, while the  $z$  component governs the rotation of the  $c$  director—and one equation for the balance of linear momentum. Two different flow geometries have been studied and the final equations governing the flow behavior are summarized in Sec. XI.

Presently, almost no experimental information regarding the Sm-C viscosity coefficients exists. However, we have been able to give some general guidelines for their values, which are summarized in Tables I–III. Clearly most of the viscosity coefficients must approach zero as the system approaches the Sm-A phase, and from the structure of the stress tensor it is possible to assign to the coefficients the tilt angle dependence given by Eqs. (4.1) and (4.2). It is also possible, by thermodynamic reasoning, to derive a set of inequalities that must be fulfilled by the viscosity coefficients. Furthermore, depending on whether or not the system exhibits flow alignment when sheared in the bookshelf geometry, one can derive some further inequalities. In Sec. XIII we also showed by comparison with the nematic stress tensor how one can obtain some additional information concerning some of the viscosity coefficients.

As stated before, virtually no experimental information regarding the Sm-C viscosity coefficients exists in the literature. There is, however, one exception to this. In studying the switching behavior of a ferroelectric system, equations based on a “nematiclike” theory, similar to Eq. (9.6b), have been employed. In this way the rotational viscosity  $\gamma$  has been measured by different methods [23–25]; a reasonable value for  $\bar{\lambda}_5$  obtained from these experiments is  $\bar{\lambda}_5 = 0.1$  Pa s. If one wants to obtain numerical values for the flow behavior of a cell in the bookshelf geometry, apart from the coefficients  $\bar{\lambda}_5$ , one also needs to assign values to the viscosity coefficients  $\bar{\lambda}_2$ ,  $\mu_0$ ,  $\bar{\mu}_3$ , and  $\bar{\mu}_4$ , as well as to the elastic constants  $\bar{B}_1$  and  $\bar{B}_2$ . Several measurements of the elastic constant  $\bar{B}_3$  are available in the literature [23–25], although measurements of the constants  $\bar{B}_1$  and  $\bar{B}_2$  are scarce [26]. From the existing data we can do no better than suggest a one-constant approximation  $\bar{B}_1 = \bar{B}_2 = \bar{B}_3 \approx 10^{-11}$  N.

Using the guidelines given by Tables II and III, it is possible to suggest a reasonable set of model parameters for making numerical calculations. These are given in Table IV. When suggesting  $\bar{\mu}_3 = 0$ , we have utilized the approximation  $\alpha_1 = 0$ , often exploited for nematic liquid crystals. However, it must be emphasized that the parameters suggested in Table IV are not the consequences of any measurements and could in the end prove to be far

TABLE III. Summary of inequalities for the Sm-C viscosity coefficients. The first two groups of inequalities must always be fulfilled by thermodynamic reasons. The inequalities belonging to the last group are fulfilled under certain circumstances, which are stated in the table.

Inequality	Source and condition
$\bar{\lambda}_5 > 0, \bar{\tau}_5 > 0$	Rotational motion (inequalities are always fulfilled)
$\mu_0 > 0$ $\mu_0 + (\bar{\mu}_4 + \bar{\lambda}_5 - 2 \bar{\lambda}_2 )\theta^2 > 0$ $\mu_0 + (\bar{\mu}_4 + \bar{\lambda}_5 + \frac{1}{2}\bar{\mu}_3)\theta^2 > 0$ $\mu_0 + \mu_2 - 2\lambda_1 + \lambda_4 > 0$ $\mu_0 + \mu_2 - 2\lambda_1 + \lambda_4 + (\bar{\mu}_4 + \bar{\mu}_5 + 2\bar{\lambda}_2 - 2\bar{\lambda}_3 + \bar{\lambda}_5 + \bar{\lambda}_6)\theta^2 > 0$	Effective viscosities (inequalities are always fulfilled)
$\bar{\lambda}_5 <  \bar{\lambda}_2 $	Flow alignment (in <b>W</b> geometry)
$\bar{\lambda}_2 < 0$	Stable flow alignment angle corresponds to a rodlike nematic liquid crystal (in <b>W</b> geometry)
$\bar{\lambda}_5 >  \bar{\lambda}_2 $	No flow alignment (in <b>W</b> geometry)
$\bar{\tau}_5 > \bar{\tau}_1$	Flow alignment corresponds to a rodlike nematic liquid crystal (in <b>B</b> geometry)

from the typical values these parameters exhibit. In any event, they provide a consistent set of parameters, which can be used for model calculations of the flow behavior of the system in the bookshelf geometry.

When studying shear flow of the system we can generally do so in one of two different geometries (Figs. 4–7). In the bookshelf geometry the system will or will not exhibit flow alignment depending on whether or not the inequality (7.11) is fulfilled. In the case when the smectic layers are sliding on top of each other, on the other hand, flow alignment is always possible. By studying the effective viscosities of the system [Eqs. (10.5)] one can also derive some additional inequalities [Eqs. (10.6)] that must be fulfilled by the viscosity coefficients. Also shown in Sec. X is the existence of backflow. In most cases a rotation of the *c* director is coupled to the macroscopic motion of the molecules in such a way that a nonzero velocity gradient is developed for a nonzero  $\dot{\phi}$  [cf. Eq. (10.4)].

In Sec. XII the importance of transverse flow effects is demonstrated. In the bookshelf geometry such effects create a transverse pressure gradient, which probably causes permeation of molecules between the layers. If, on the other hand, the smectic layers are parallel to the bounding glass plates, a transverse flow is developed, which in turn develops additional torques in the system.

TABLE IV. A suggested set of parameters to be used for model calculations on the flow properties of the system in the bookshelf geometry.

Viscosity coefficients (Pa s)					Elastic constants (N)		
$\bar{\lambda}_2$	$\bar{\lambda}_5$	$\mu_0$	$\bar{\mu}_3$	$\bar{\mu}_4$	$\bar{B}_1$	$\bar{B}_2$	$\bar{B}_3$
−0.12	0.1	0.2	0	0.03	$10^{-11}$	$10^{-11}$	$10^{-11}$

It is of course obvious that if the sample is bounded, the transverse flow might be prohibited and instead a transverse pressure gradient can be developed.

Today, the study of the dynamic behavior of Sm-C liquid crystals is mainly limited to the study of switching phenomena for ferroelectric Sm-C\* liquid crystals in the bookshelf geometry. The theoretical description of these studies are normally based on some intuitive “nematic-like” theory. In this paper we have provided a complete “exact” smectic theory for the analysis of dynamic problems of the Sm-C phase. Although the nematiclike approach to the problem provides equations that in many cases qualitatively agree with the proper ones, this approach is too simple and will not give the correct scaling of the relevant expressions in terms of  $\theta$ . We have also demonstrated that the complete study of the switching behavior of the system must include backflow as well as transverse flow effects. How important these complications turn out to be depends on the values of some relevant viscosity coefficients for which we lack experimental information. Thus the experimental determination of these coefficients should be the next step towards a better understanding of the general macroscopic flow properties of Sm-C and Sm-C\* liquid crystalline systems.

## APPENDIX

In this appendix we use a mechanical analog to develop a more physical understanding of the external and constraint torques acting on **n** during rotation on the tilt cone in the smectic-C phase. We model the rotating molecule as a solid object having the mean smectic-C molecular shape rotating about a fixed point in a viscous fluid. The mean molecular shape in the smectic-C phase can be represented by the bent cylinder of Fig. 9(a). This shape

embodies the monoclinic  $C_{2h}$  symmetry of the smectic- $C$  molecular environment. For rotation at small Reynolds number in a viscous fluid, the torque  $\Gamma$  produced by a body in rotation with angular velocity  $\omega$  is given by  $\Gamma^r = -\underline{\gamma}\omega$ , where  $\underline{\gamma}$  is a tensor of drag coefficients. A proper choice of the principal axis coordinate system will diagonalize  $\underline{\gamma}$ , and for the object of Fig. 9(a) one principal axis is the twofold rotation axis  $\hat{\phi}$ . For the purposes of

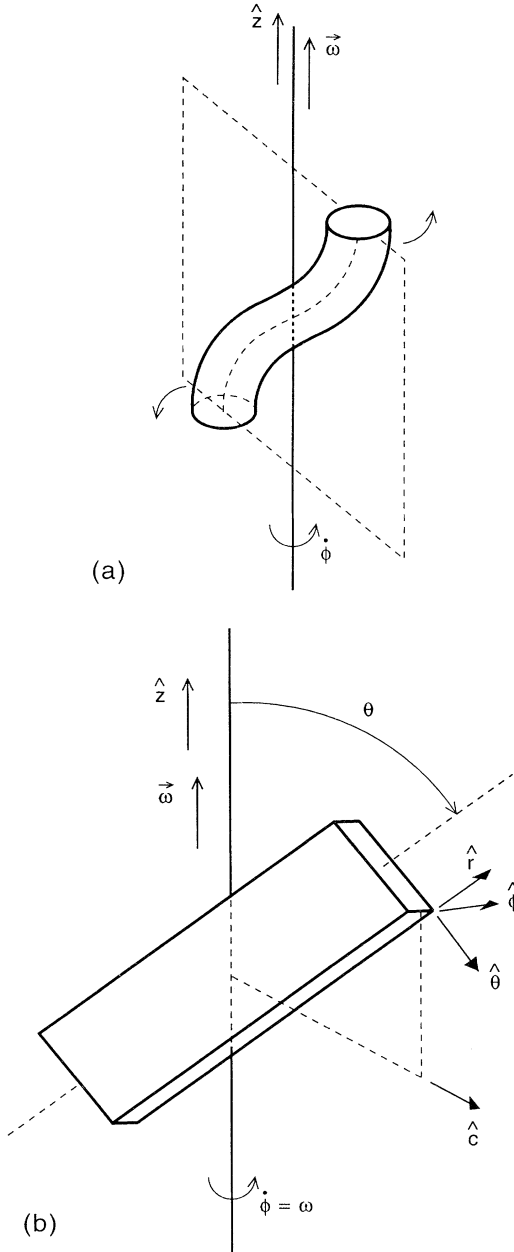


FIG. 9. Solid objects representing the mean molecular shape in the smectic- $C$  phase for modeling the coupling of director orientation and torque by rotation of a solid body in a viscous fluid: (a) Bent cylinder shape reflecting the monoclinic symmetry of the smectic- $C$  phase. (b) orthorhombic box equivalent for the present purposes. The box is constrained to rotate about the thin rod along  $\hat{z}$ .

this discussion nothing is lost by simplifying the object shape to the biaxial orthorhombic box of Fig. 9(b), for which  $\underline{\gamma}$  is diagonal in the coordinate frame having axes parallel to the three twofold symmetry axes of the box. We orient the box in the smectic- $C$  phase as shown in Fig. 9(b), with its longest edge parallel to  $\mathbf{n}$ , which is  $\hat{r}$ . The smectic- $C$  symmetry demands that one of the other twofold axes be along  $\hat{\phi}$  and we choose this to be the shortest edge. Nothing significant changes in the discussion to follow if the other choice were to be made. With this geometry we can define the three rotational viscosities  $\gamma_r$ ,  $\gamma_\theta$ , and  $\gamma_\phi$  for rotation of the box about its symmetry axes  $\hat{r}$ ,  $\hat{\theta}$ , and  $\hat{\phi}$ , respectively,

$$\Gamma_r^r = -\gamma_r \omega_r, \quad \Gamma_\theta^r = -\gamma_\theta \omega_\theta, \quad \Gamma_\phi^r = -\gamma_\phi \omega_\phi. \quad (\text{A1})$$

We now consider attaching the box to a thin rod constrained by bearings to remain parallel to  $\hat{z}$ , but free to rotate about  $\hat{z}$ , such that

$$\omega = \hat{z} \omega_z = \hat{z} \dot{\phi} = \dot{\phi} [-\hat{\theta} \sin\theta + \hat{r} \cos\theta]. \quad (\text{A2})$$

We apply a net torque  $\Gamma^{\text{net}}$  to the box, where  $\Gamma^{\text{net}}$  is a sum of external and constraint torques, in which case  $\Gamma^{\text{net}} + \Gamma^r = 0$ , i.e.,

$$\Gamma^{\text{net}} = \Gamma^{\text{ext}} + \Gamma^c = -\Gamma^r = \dot{\phi} [-\hat{\theta} \gamma_\theta \sin\theta + \hat{r} \gamma_r \cos\theta]. \quad (\text{A3})$$

The constraint torque  $\Gamma^c$  (due to the rod) can have no  $\hat{z}$  component and its  $\hat{\phi}$  component (note that  $\hat{\phi} = \mathbf{b}$ ) will simply balance the  $\hat{\phi}$  component of  $\Gamma^{\text{ext}}$  (due to external forces), leaving  $\Gamma_\phi^{\text{net}} = 0$  and  $\theta$  constant,

$$\Gamma_\phi^{\text{net}} = \Gamma_\phi^{\text{ext}} + \Gamma_\phi^c = 0. \quad (\text{A4})$$

Note that the orientation of  $\Gamma^{\text{net}}$  is independent of  $\omega$ , depending only on  $\gamma_\theta/\gamma_r$  and  $\theta$ . Thus, for a given external torque the remaining  $\mathbf{c}$  component of the constraint torque  $\Gamma^c$  must be applied by the bearings in order to obtain  $\Gamma_\theta^{\text{net}}/\Gamma_r^{\text{net}} = -(\gamma_\theta/\gamma_r)\tan\theta$ , which will maintain the rod orientation along  $\hat{z}$ .

In general, the external torque that affects the change of  $\phi$  will have both ferroelectric  $\hat{z}\Gamma^f$  and dielectric and magnetic  $\hat{z}\Gamma^\epsilon$  components

$$\Gamma^{\text{ext}} = \hat{z}\Gamma^f + \hat{\theta}\Gamma^\epsilon = \hat{r} \cos\theta \Gamma^f + \hat{\theta}(\Gamma^\epsilon - \sin\theta \Gamma^f), \quad (\text{A5})$$

while the most general form the countertorque can adopt is

$$\Gamma^c = \Gamma_c^c \mathbf{c} + \Gamma_\phi^c \hat{\phi} = \Gamma_c^c \sin\theta \hat{r} + \Gamma_c^c \cos\theta \hat{\theta} + \Gamma_\phi^c \hat{\phi}. \quad (\text{A6})$$

Writing down the  $\hat{r}$  and  $\hat{\theta}$  components, respectively, of the general torque equation  $\Gamma^{\text{ext}} + \Gamma^c + \Gamma^r = 0$  gives, by the use of Eqs. (A3), (A5), and (A6),

$$\dot{\phi} \gamma_r \cos\theta = \Gamma^f \cos\theta + \Gamma_c^c \sin\theta, \quad (\text{A7a})$$

$$\dot{\phi} \gamma_\theta \sin\theta = \Gamma^f \sin\theta - \Gamma_c^c \cos\theta - \Gamma^\epsilon, \quad (\text{A7b})$$

from which we obtain  $\dot{\phi}$  and  $\Gamma_c^c$  in terms of the external torque as

$$\dot{\phi} = \frac{\Gamma^f - \Gamma^\epsilon \sin\theta}{\gamma_r \cos^2\theta + \gamma_\theta \sin^2\theta}, \quad (\text{A8})$$



$$\Gamma_c^c = (\gamma_r - \gamma_\theta) \sin\theta \cos\theta \dot{\phi} - \Gamma^\epsilon \cos\theta . \quad (\text{A9})$$

Comparing the terms including  $\dot{\phi}$  in these expressions with Eqs. (6.3c) and (6.7b) we can relate the viscosities  $\lambda_5$  and  $\tau_5$  to the model viscosities of Eqs. (A1)

$$2\lambda_5 = \gamma_r \cos^2\theta + \gamma_\theta \sin^2\theta , \quad (\text{A10a})$$

$$2\tau_5 = (\gamma_r - \gamma_\theta) \sin\theta \cos\theta . \quad (\text{A10b})$$

The coefficient  $\gamma_\theta$  is the viscosity for the rotation of  $\mathbf{n}$  and  $\gamma_r$  is the viscosity for rotation about  $\mathbf{n}$ . In an uniaxial nematic liquid crystal the  $\gamma_r$  term is absent, but arises in

the smectic-*C* phase from the biaxiality of the molecular environment. As the temperature is raised towards the smectic-*C*–Sm-*A* transition,  $\theta$  decreases, apparently leaving a nonzero  $\lambda_5 = \gamma_r$  at  $\theta = 0$ . However, if  $\psi$  is the orientational order parameter for ordering the molecular short axis in the smectic-*C* mean field, then we expect  $\gamma_r \sim \psi^2$  and  $\psi \sim \theta$  so that the biaxiality of the smectic-*C* phase also decreases with  $\theta$ , giving  $\gamma_r \sim \theta^2$  and  $\lambda_5$  an overall  $\theta^2$  dependence. When  $\theta$  is large we expect  $\psi^2 \sim 1$  and  $\gamma_r/\gamma_\theta \sim |\psi|^2(D/L) \sim D/L \sim 0.17$  for typical smectic-*C* molecules ( $D \sim 5 \text{ \AA}$  and  $L \sim 30 \text{ \AA}$ ). The coefficient  $\tau_5$  determines the magnitude of the constraint torque, varying as  $\tau_5 \sim \theta$  for small  $\theta$ .

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