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The concept of rotational viscosities of smectic C and chiral smectic C* liquid crystals

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The concept of rotational viscosities of smectic C and ferroelectric chiral smectic C* liquid crystals is reviewed. We give the expressions for the elastic torque, the rotational torque and the entropy production in spherical polar coordinates for smectic C and ferroelectric chiral smectic C* liquid crystals for pure rotational motion. Comparing these with the corresponding expressions for nematic liquid crystals, we show the way in which the rotational viscosity coefficients must be defined for smectic C and ferroelectric smectic C* liquid crystals in order to obtain consistency between the description of rotational motion of these systems and nematic liquid crystals.

1. Introduction

Smectic C (S_C) and chiral smectic C* (S_C^*) liquid crystals (tilted smectics) are layered structures [1], where the molecules within each layer are tilted, on average, with respect to the normal of the layer. In order to describe the orientation of the molecules within each layer the coordinates θ and ϕ are normally introduced. Here θ describes the tilt of the molecules with respect to the normal of the smectic layer, and ϕ describes the phase of the molecules, i.e. the angle between the projection of the molecules into the smectic planes and some reference direction within this plane. In effect, we have therefore introduced a spherical polar coordinate system.

Here we study the dynamics of the S_C and S_C^* phases, restricted to pure rotations, i.e. the macroscopic flow \mathbf{v} of the molecules is assumed to be zero. Important material parameters connected to this motion are the rotational viscosities, which enable us to relate the rotational motion of the director to the corresponding generated viscous rotational torque Γ^r . Generally, this torque is related to the velocity $\dot{\alpha}$ of the director by a 3×3 rotational viscosity tensor γ

$$\Gamma^r = \gamma \dot{\alpha}, \quad (1)$$

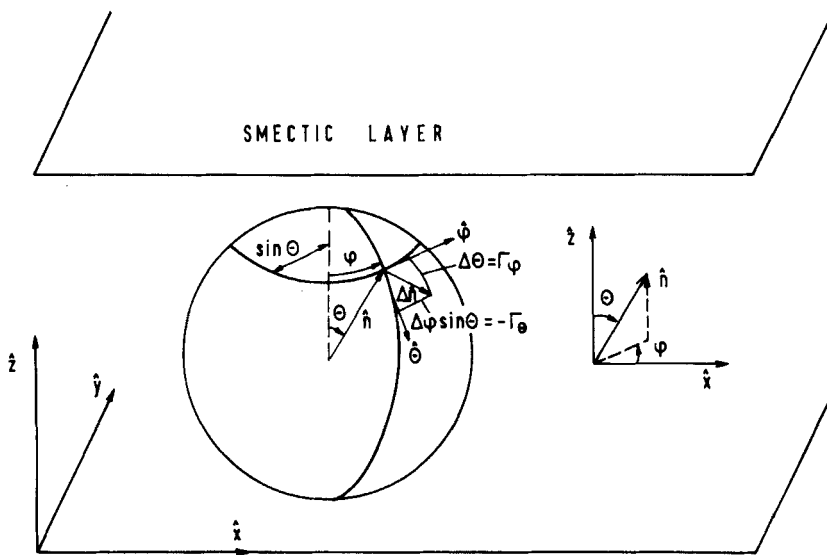
where α is a coordinate describing the director. By symmetry reasoning, Escher *et al.* have argued [2] that the principal axes of γ are those corresponding to the rotations where θ (the soft mode) and ϕ (the Goldstone mode) change, respectively, and to the rotation of the molecules around their long molecular axes. As we are not interested in this last type of rotation and, as the corresponding eigenvalue of γ should be expected to be much smaller than the other two, we are left with two rotational viscosities which we denote γ_s and γ_G .

In the literature we can find two different ways [2-5] of defining the Goldstone mode rotational viscosity γ_G . We shall argue that one of these definitions [3-5] produces apparent singularities in the description of the system, which are not due to the physics but rather to the fact that the mathematical description is singular whenever θ approaches zero, i.e. close to the transition temperature T_c to the smectic A (S_A) phase. Instead we will show the way in which the rotational viscosities of the system must be introduced if we want to achieve consistency with the corresponding description [6-8] of nematic liquid crystals.

2. The spherical polar coordinate system: relationships between torques and rotations

The equations governing the statics and dynamics of the systems we study can be viewed [8, 9] as a balance of torque equation, where the torques acting on the director are generalized forces in a spherical polar coordinate system. The coordinate system we use is shown in the figure. The average direction in which the molecules point is described by a unit vector \hat{n} , commonly called the director. The intersection between the director and the unit sphere determines the average orientation of the molecules uniquely, and the rotation of the director can thus be described by the motion of this trace point on the unit sphere [8, 9]. We orient the coordinate system in such a way that the direction $\theta = 0$ corresponds to the normal to the smectic layers which we take to be the z direction. As pointed out in §1, we shall study only pure rotations of the director. We also restrict the study to systems at a uniform temperature.

The general dynamic equations can be written down as a balance of torque equation, $\Gamma = 0$, where Γ is the sum of all torques which act upon the director.



The coordinate system used in this work is a conventional spherical polar one, oriented in such a way that the pole coincides with the z axis, which is taken to be the normal to the smectic layers. The angle θ thus denotes the tilt of the director with respect to the normal of the smectic layers, whereas ϕ is the angle between the projection of the director into the smectic plane and the x axis. The effect of an applied torque $\Gamma = \Gamma_\theta \hat{\theta} + \Gamma_\phi \hat{\phi}$ is to rotate the director with the momentary axis of rotation given by Γ . The change of the director is then given by $\Delta \hat{n} \sim (\Gamma_\phi \hat{\theta} - \Gamma_\theta / \sin \theta \hat{\phi})$.

We study the case where the molecules relax under the influence of elastic forces only. The torque acting on the director can be divided into two parts [8, 9], i.e. we write

$$\Gamma = \Gamma^r + \Gamma^{\text{el}},$$

where Γ^r is the rotational torque which is related to the viscous dissipation and Γ^{el} is the elastic torque. As we are using a spherical polar coordinates system the torques have to be divided into their proper components,

$$\Gamma = \Gamma_\theta \hat{\theta} + \Gamma_\phi \hat{\phi},$$

where the direction of the basis vectors $\hat{\theta}$ and $\hat{\phi}$ are shown in the figure. If we consider only systems where the director is allowed to vary in the z direction, i.e. $\theta = \theta(z)$ and $\phi = \phi(z)$, we can express the free energy density of the system as $g = g(\theta, \theta', \phi, \phi')$, where $\theta' \equiv d\theta/dz$ and $\phi' \equiv d\phi/dz$, respectively. The discussion can, of course, be generalized to the case where electric or magnetic fields are present, just by adding the corresponding contributions to the free energy density. If the system is in a state such that g is not at a minimum, an elastic torque will be exerted on the director according to [8, 9]

$$\Gamma_\theta^{\text{el}} = \frac{1}{\sin \theta} \left(\frac{\partial g}{\partial \phi} - \frac{d}{dz} \frac{\partial g}{\partial \phi'} \right), \quad (2a)$$

$$\Gamma_\phi^{\text{el}} = - \left(\frac{\partial g}{\partial \theta} - \frac{d}{dz} \frac{\partial g}{\partial \theta'} \right). \quad (2b)$$

The consequence of this elastic torque acting on the director in a state given by a point (θ, ϕ) on the unit sphere is to rotate the director with the momentary axis of rotation given by Γ^{el} . As is shown in the figure, θ and ϕ will then change according to

$$\Delta\phi \sim -\Gamma_\theta^{\text{el}}/\sin \theta, \quad (3a)$$

$$\Delta\theta \sim \Gamma_\phi^{\text{el}}. \quad (3b)$$

The minus sign in equation (3a) comes from the fact that a rotation along the positive $\hat{\theta}$ axis corresponds to decreasing ϕ and the factor $\sin \theta$ is due to the fact that when changing ϕ , the director travels along a circle on the unit sphere, which has a radius of $\sin \theta$. The balance of torque equation is now written in component form as

$$\Gamma_\theta^{\text{el}} + \Gamma_\theta^r = 0, \quad (4a)$$

$$\Gamma_\phi^{\text{el}} + \Gamma_\phi^r = 0, \quad (4b)$$

where Γ_θ^r and Γ_ϕ^r are the spherical polar components of the rotational torque. Generally, this rotational torque is related to the rotations of the director by a viscosity tensor (see equation (1)). The diagonal form of equation (1) is obtained if we use the spherical polar coordinate system introduced by the figure and we can therefore write

$$\Gamma_\theta^r = \gamma_G \dot{\phi} \sin \theta, \quad (5a)$$

$$\Gamma_\phi^r = -\gamma_S \dot{\theta}. \quad (5b)$$

By equations (5) we have defined the rotational viscosities γ_G and γ_S . The signs in these definitions have been chosen in such a way that if both γ_G and γ_S are taken to be positive, then the corresponding rotational torque will counteract the corresponding rotation (cf. the figure). The factor $\sin \theta$ in equation (5a) comes from the fact that we

wish to introduce the rotational viscosities in such a way that they relate the rotational torque to the physical displacement of the director under a rotation. In such a way the definitions of equations (5) will be consistent with the choice of the generalized fluxes and forces in §3, which in turn are chosen to be consistent with the Leslie–Ericksen formulation [6–8] of the hydrodynamic theory of nematics.

3. The dissipation function: definition of the rotational viscosity of nematics

We now study the rotational motion of the director from the point of view of irreversible statistical mechanics [10]. We shall write down the entropy production σ of the system expressed in terms of a set of generalized fluxes J_i and forces X_i . The choice of J_i and X_i is to a certain extent arbitrary, but the product of any flux and its conjugate force must have the dimension of entropy production. Our choice of fluxes are

$$J_1 = -\dot{\phi} \sin \theta, \quad (6a)$$

$$J_2 = \dot{\theta}. \quad (6b)$$

The conjugate forces to these fluxes are chosen to be the torque components of equations (1)

$$X_1 = \Gamma_{\theta}^{\text{el}}, \quad (7a)$$

$$X_2 = \Gamma_{\phi}^{\text{el}}. \quad (7b)$$

The general phenomenological equations relating these forces and fluxes are written $\mathbf{J} = \mathbf{L}\mathbf{X}$. In the coordinate representation that we have chosen the tensor \mathbf{L} is diagonal, the elements of which are just the reciprocal of the rotational viscosities γ_G and γ_S . The entropy production of the system can now be written (T being the temperature of the system) as

$$T\sigma = \mathbf{J} \cdot \mathbf{X} = \gamma_G \dot{\phi}^2 \sin^2 \theta + \gamma_S \dot{\theta}^2, \quad (8)$$

where we have utilized equations (4)–(7).

We study briefly how this compares to the Leslie–Ericksen theory of nematodynamics [6–8]. The entropy production per unit volume is, if the system performs pure rotational motion, given by [11, 12]

$$T\sigma = \gamma_1 \dot{\mathbf{n}} \cdot \dot{\mathbf{n}}, \quad (9)$$

where γ_1 is the nematic rotational viscosity and \mathbf{n} is the nematic director. Expressing \mathbf{n} by the polar angles θ and ϕ , equation (9) gives

$$T\sigma = \gamma_1 (\dot{\phi}^2 \sin^2 \theta + \dot{\theta}^2). \quad (10)$$

Furthermore, the spherical polar components of the rotational torque acting on the director under a general rotation can be derived [8, 9] to be

$$\Gamma_{\theta}^r = \gamma_1 \dot{\phi} \sin \theta, \quad (11a)$$

$$\Gamma_{\phi}^r = -\gamma_1 \dot{\theta}. \quad (11b)$$

Equations (10) and (11) are derived with the Leslie–Ericksen stress tensor [6] of nematic liquid crystals as a starting point. We conclude therefore that the symmetry of this stress tensor allows only one rotational viscosity for the nematic phase: only

the velocity of the trace point of the director on the unit sphere determines the dissipation, independently if the rotation is of a θ or a ϕ nature.

4. The smectic C and smectic C* rotational motion: torques and dissipation

Generally, a viscosity is a material parameter which is a measure of the response of a system when it is acted upon by a certain force, i.e. the viscosity can be defined as the ratio force/flux. This definition can be formalized in the language of irreversible statistical mechanics, discussed in §3. As there is a certain degree of freedom of choosing the fluxes and forces, there will be the possibility of introducing different definitions of the viscosities of the system. Generally the choice of fluxes and forces should be such, that the fluxes (see equations (6)) correspond to the time derivatives of some generalized coordinates, while the forces (see equations (7)) correspond to the derivatives (see equations (2)) of a potential function with respect to these generalized coordinates. We have, when studying the S_C and S_C^* phases, chosen these generalized coordinates to correspond to the physical displacements θ and $\phi \sin \theta$ of the director under a rotation. This has, as a consequence, that the rotational torque of the system is given by equations (5) while the entropy production is given by equation (8). These equations can be viewed as the definition of the rotational viscosities of the S_C and S_C^* phases and is, in our opinion, the most physically sound one as it relates the rotational viscosities to the true motion of the director. We also see when comparing equations (5) with equations (11) and equation (8) with equation (10) that this definition makes the description of rotational motion of the S_C and S_C^* phases consistent with the corresponding description of nematics.

Through equations (5) and (8) we have introduced two rotational viscosities for and the S_C and S_C^* phases: γ_G which is related to rotations along the smectic cone (the Goldstone mode) and γ_s which is related to rotations in which the tilt of the molecules changes (the soft mode). The molecules which constitute the smectic systems and the nematics are of the same kind. The fact that the dissipation for both θ and ϕ rotations in the nematic phase is regulated by the same rotational viscosity γ_1 makes us believe that even if we generally have to introduce two rotational viscosities for the tilted smectics, these should be approximately equal. We do not think that the nature of the dissipation changes drastically just because the molecules are confined in layers. We should also keep in mind that the layers are far from being perfect, the molecules frequently stick into the neighbouring layer, and so on. Thus we think that the approximation $\gamma_G \simeq \gamma_s$ should be valid to a reasonable accuracy. The validity of this approximation should also be better the closer to T_c the system is, because at T_c the θ and ϕ rotations are degenerate. In the end, however, experiments will have to confirm this approximation.

5. Rotational motion along the smectic cone: connection between proper and effective material parameters

In this section we write down the equation governing the electro-optic switching [13] of the ferroelectric S_C^* phase, and we show how the analysis of this equation is related to a proper way of defining as well as the rotational viscosity γ_G the twist elastic constant K_3 . We make the analysis as simple as possible, considering only the contributions to the free energy density from the twisting of the layers and from the interaction between the spontaneous polarization and the electric field. Furthermore we make the constant amplitude approximation and consider only switching for

which θ is constant. This is a good approximation, except [14] very close to T_c where the soft mode [15] deformations become important. The simplest form of the Landau free energy density [16, 17] appropriate for the analysis can, assuming that the electric field is applied along the y direction, be written [14] as

$$g = \frac{1}{2}K_3 \left(\frac{d\phi}{dz} - \frac{\Lambda}{K_3} \right)^2 \theta^2 - PE \cos \phi. \quad (12)$$

In this equation K_3 is the twist elastic constant, Λ the Lifschitz coefficient responsible for the twisting of the layers, P the spontaneous polarization of the system and E the magnitude of the electric field. We also assume that the system is sufficiently close to T_c that we can replace $\sin \theta$ by θ , although not so close that the constant amplitude approximation is not valid. The sum of the elastic and electric torques acting on the director is now calculated from equations (2a) and (12):

$$\Gamma_\theta^{\text{el}} = \frac{PE}{\theta} \sin \phi - K_3 \theta \frac{d^2 \phi}{dz^2}, \quad (13)$$

where we again have replaced $\sin \theta$ by θ . The equation governing the rotation of the director is given by inserting the rotational (see equation (5a)) and the elastic (see equation (13)) torques into the balance of torque equation (see equation (4a)). In this way we find

$$\theta^2 K_3 \phi'' - PE \sin \phi = \theta^2 \gamma_G \dot{\phi}. \quad (14)$$

This equation is often found written in the slightly different form [3]

$$K_3^{\text{eff}} \phi'' - PE \sin \phi = \gamma_G^{\text{eff}} \dot{\phi}. \quad (15)$$

Equation (15) is obtained if we do not define the rotational and the elastic torques with respect to the true physical displacement $\phi \sin \theta$ but instead with respect to the angle ϕ , and if we also include in the free energy density of equation (12) the θ^2 dependence in the twist elastic constant K_3 . Thus, if equation (15) is used to analyse an electro-optic switching experiment in order to obtain the Goldstone mode rotational viscosity or the twist elastic constant, we see by comparing equations (14) and (15) that the quantities determined are what can be called an effective rotational viscosity $\gamma_G^{\text{eff}} = \gamma_G \sin^2 \theta$ and an effective twist elastic constant $K_3^{\text{eff}} = K_3 \sin^2 \theta$, where for completeness we have not made the approximation $\sin \theta \simeq \theta$ when defining the effective material parameters.

The benefit of using equation (15) when analysing an experiment is, of course, that only K_3^{eff} and γ_G^{eff} can be determined unless we also measure the tilt of the system. However, it is K_3 and γ_G which are the quantities connected to the true physical torques which act on the director. Thus, when studying the temperature dependence of the material parameters of the system we must be aware of the fact that in K_3^{eff} and γ_G^{eff} we have imposed an extra temperature dependence through the factor $\sin^2 \theta$. This leads to a rather unphysical description, because γ_G^{eff} approaches zero at T_c and a vanishing viscosity should indicate some kind of superfluid-like rotation of the molecules. This is, of course, not the case, but rather a specific rotation $\Delta\phi$ corresponds to a smaller physical displacement of the molecules the closer to T_c the system is, a displacement which degenerates to one point at T_c .

6. Conclusions

In this paper we have shown that if we wish to achieve consistency between the viscoelastic description of the S_C and S_C^* phases and the already existing and accepted description for nematic liquid crystals, the rotational torque acting on the director must be that given by equation (5) whereas the twist elastic torque must be derived from equation (2a) using the Landau free energy density given by equation (12). This is due to the fact that effectively we are using the spherical polar coordinate system of the figure in describing the system, the coordinate ϕ of which corresponds to a physical displacement $\phi \sin \theta$ of the director. If we do not adopt these definitions, instead hiding the θ terms in the definitions of γ_0^{eff} and K_3^{eff} instead of keeping them in the equations, we lose track of which part of the temperature dependence of the physical quantities that we study is related to the temperature dependences of the material parameters and which part is related to the temperature dependence of the tilt, θ . In conclusion we argue therefore that in order to obtain a coherent and physically sound description of the viscoelastic behaviour of the S_C and S_C^* phases, the rotational viscosities should be defined with equations (5) and (8) as a basis, whereas the twist elastic part of the Landau free energy density must be that first introduced by Pikin and Indenbom [16], the corresponding twist electric torque then being derived by the use of equation (2a).

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