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Perturbative Approach to the Relaxation of the Nematic Deformation: Surface Viscosity and Electric Field

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The effect of a localized surface viscosity on the relaxation of a pre-existing deformation in a nematic liquid crystal cell is perturbatively analyzed in the presence of an external field. When the surface viscosity of the system is small enough to use perturbation developments, approximated solutions can be obtained describing the space-time profile of the director angle. It is shown that if the surface viscosity increases, the relaxation becomes slower when compared to the case corresponding to the absence of viscosity. The temporal behavior of the optical path difference is analytically established by incorporating the contribution of the surface viscosity.

Keywords Electric field effects; reorientation; surface viscosity

1. Introduction

The concept of surface viscosity can find application in different experimental contexts including some interfaces in soap films, where its relation to the diffusion constants can be established [1], in adsorbed albumin films in which its strong variation with time aging can be determined [2], and in liquid-crystalline systems, where it plays a special role in the reorientation process [3]. The surface viscosity together with the anchoring energy completes the framework to understand surface properties connected with relaxation processes of nematic liquid crystals (NLC) samples under the action of external fields [3–12]. However, in comparison with the anchoring energy the liquid crystal surface viscosity is very little documented because it is a quantity difficult to measure [13]. On the theoretical side, the role of surface viscosity [14] on the dynamical behavior of nematic liquid crystal (NLC) cells has been object of attention in the last years also in connection with important mathematical problems it evokes [15–18]. In recent papers, particular attention was devoted to the problem of incompatibility which arises when the initial derivative of the director angle on the bounding surface is deduced from bulk or from surface dynamic equations [15–20]. Anyway, the role of the surface viscosity on the molecular orientation

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in NLC samples deserves further analysis if a more detailed comprehension of surface effects in liquid-crystalline systems is desired.

For this reason, in this article, we examine the mathematical problem of obtaining the director angle profile in an NLC sample when the surface viscosity can be considered small enough to allow for a perturbative treatment. In Sec. 2, the problem is formulated in general terms, introducing appropriate boundary conditions embodying the presences of a surface viscosity and of a finite anchoring energy. In Sec. 3, the perturbative treatment is presented and use is made of the Green's function approach to determine the space-time director angle profile in first order of perturbation in the surface viscosity coefficient. In Sec. 4, some theoretical results for the time derivative of the director angle and for the optical path difference are presented for a relevant set of parameters characterizing a typical NLC sample. In Sec. 5, some concluding remarks are presented.

2. Statement of the Problem

The sample is an NLC cell in the shape of a slab of thickness d in such a manner that the z -axis of a Cartesian reference frame is normal to the bounding walls placed at $z = \pm d/2$. The twist angle formed by the nematic director \mathbf{n} with the x -axis, characterizing the nematic deformation, is indicated by ϕ , and the one-elastic constant approximation is assumed. We also consider that the sample is subjected to an electric field $E(t) = E(t)\hat{z}$, where \hat{z} is the unit vector specifying the \hat{z} -direction. The dynamics of the director angle in the cell is governed by the equation arising from the balance condition between the elastic and viscous torque [21]:

$$\eta_b \frac{\partial}{\partial t} \phi(z, t) = K \frac{\partial^2}{\partial z^2} \phi(z, t) - \varepsilon_a E^2(t) \phi(z, t) \quad (1)$$

where K is the elastic constant, η_b is the (effective) bulk viscosity and ε_a is the dielectric anisotropy of the liquid crystal. The equilibrium profile of the director angle has to be searched satisfying the boundary conditions stating that the elastic torque transmitted by the liquid crystal to the limiting surface is balanced by the restoring torque due to the anisotropic interaction of the nematic with the surface and by the viscous torque, due to the surface dissipation, namely

$$\left(K \frac{\partial}{\partial z} \phi(z, t) + W(\phi(z, t) - \phi_s) + \eta_s \frac{\partial}{\partial t} \phi(z, t) \right) \Big|_{z=-d/2} = 0 \quad (2)$$

and

$$\frac{\partial}{\partial z} \phi(z, t) \Big|_{z=0} = 0 \quad (3)$$

connected with the finite anchoring energy, surface viscosity and to the symmetry of the problem, that implies $\phi(z, t) = \phi(-z, t)$. In Eqs. (2) and (3), we have assumed that the surface anchoring energy can be approximated by the Rapini-Papoular form, where W is the anchoring energy strength [4], and ϕ_s is the surface easy axis, as $f_s = W(\phi - \phi_s)^2/2$. We have indicated by η_s the surface viscosity due to the presence

of the bounding surface [14]. For $t \leq 0$, the director angle profile is $\phi(z, t) = \Phi(z)$, with $\Phi(z)$, obtained by solving the equation

$$K \frac{d^2}{dz^2} \Phi(z) - \varepsilon_a E_0^2(t) \Phi(z) = 0 \quad (4)$$

subjected to the conditions

$$\left(K \frac{d}{dz} \Phi(z) + W(\Phi(z) - \phi_s) \right) \Big|_{z=\pm d/2} = 0 \quad (5)$$

and

$$\frac{d}{dz} \Phi(z) \Big|_{z=\pm d/2} = 0 \quad (6)$$

In particular, the solution for Eq. (4) subjected to the condition given by Eqs. (5) and (6) is given by

$$\Phi(z) = \frac{u \phi_s \cos(z/\lambda)}{\sin(z/(2\lambda)) + u \cos(z/(2\lambda))} \quad (7)$$

with $u = Wd/K$ and $\lambda = 1/E_0 \sqrt{\varepsilon_a/K}$. From Eqs. (1), (2) and (3) it follows that

$$\left(\frac{\partial \phi}{\partial t} \right)_{\text{surface}, t=0} = \left(\frac{\partial \phi}{\partial t} \right)_{\text{bulk}, t=0} \quad (8)$$

This result shows that there is no incompatibility between the time derivative on the surface evaluated by means of the bulk equation, Eq. (1), and by means of the boundary conditions, Eqs. (2) and (3), at $t = 0$, in contrast to the case discussed in [2].

3. Perturbative Approach

In the following we write Eqs. (1)–(3) in the dimensionless form

$$\frac{\partial}{\partial t_r} \phi(z_r, t_r) = \frac{\partial^2}{\partial z_r^2} \phi(z_r, t_r) - \frac{1}{\lambda^2} f(t_r) \phi(z_r, t_r) \quad (9)$$

and the corresponding boundary conditions as:

$$\left(\frac{\partial}{\partial z_r} \phi(z_r, t_r) + u(\phi(z_r, t_r) - \phi_s) + v \frac{\partial}{\partial t} \phi(z_r, t) \right) \Big|_{z_r=-1/2} = 0 \quad (10)$$

and

$$\frac{\partial}{\partial z_r} \phi(z_r, t_r) \Big|_{z_r=0} = 0 \quad (11)$$

where $z_r = z/d$, $t_r = t/\tau_D$, $\nu = \eta_s/(\eta_b d)$, and $f(t)$ represents the time dependent part of the electric field. The intrinsic time $\tau_D = \eta_b d^2/K$ is the diffusion time. The above equation subjected to the conditions given by Eqs. (10) and (11) leads us to cumbersome calculations. For this reason, we consider the viscosity of the system as sufficiently small in order to make possible the use a perturbation development to obtain approximated solutions. In fact, by assuming that the thickness of the sample is $d = 10^{-6}$ m, and $\eta_s/\eta_b = 10^{-8}$ s, as reported in [6], we get $\nu = 0.01$ which is a small value and may be considered as a perturbation on the relaxation of the system. In this manner, we consider that the solution for the director angle profile may be written as [22]

$$\phi(z_r, t_r) = \phi^{(0)}(z_r, t_r) + \nu \phi^{(1)}(z_r, t_r) \quad (12)$$

where $\phi^{(0)}(z_r, t_r)$ is the unperturbed solution of the system and $\phi^{(1)}(z_r, t_r)$ corresponds to the first order correction for the unperturbed solution due to the viscosity ν . By substituting Eq. (12) in Eqs. (9), (10) and (11), we obtain for $\phi^{(0)}(z_r, t_r)$ the equation

$$\frac{\partial}{\partial t_r} \phi^{(0)}(z_r, t_r) = \frac{\partial^2}{\partial z_r^2} \phi^{(0)}(z_r, t_r) - \frac{1}{\lambda^2} f(t_r) \phi^{(0)}(z_r, t_r) \quad (13)$$

satisfying the boundary conditions

$$\left(-\frac{\partial}{\partial z_r} \phi^{(0)}(z_r, t_r) + u(\phi^{(0)}(z_r, t_r) - \phi_s) + \nu \frac{\partial}{\partial t} \phi^{(0)}(z_r, t_r) \right) \Big|_{z_r=-1/2} = 0 \quad (14)$$

and

$$\frac{\partial}{\partial z_r} \phi^{(0)}(z_r, t_r) \Big|_{z_r=0} = 0 \quad (15)$$

together with the initial condition $\phi^{(0)}(z_r, 0) = \Phi(z)$. For $\phi^{(1)}(z_r, t_r)$, it is possible to show that it obeys the differential equation

$$\frac{\partial}{\partial t_r} \phi^{(1)}(z_r, t_r) = \frac{\partial^2}{\partial z_r^2} \phi^{(1)}(z_r, t_r) - \frac{1}{\lambda^2} f(t_r) \phi^{(1)}(z_r, t_r) \quad (16)$$

and has to satisfy the boundary conditions

$$\left(\frac{\partial}{\partial z_r} \phi^{(1)}(z_r, t_r) + u(\phi^{(1)}(z_r, t_r) - \phi_s) \right) \Big|_{z_r=-1/2} = -\nu \frac{\partial}{\partial t} \phi^{(0)}(z_r, t_r) \Big|_{z_r=-1/2} \quad (17)$$

and

$$\frac{\partial}{\partial z_r} \phi^{(1)}(z_r, t_r) \Big|_{z_r=0} = 0 \quad (18)$$

with the initial condition $\phi^{(1)}(z_r, t_r)$. Note that the compatibility condition required by the time derivative of the director angle on the surface and the time derivative on the bulk is also verified in the functions obtained by means of the perturbative treatment. In fact, for $\phi^{(0)}(z_r, t_r) = \Phi(z_r)$ it is possible to show that

$$\left(\frac{\partial}{\partial t} \phi^{(0)}(z_r, t_r) \right)_{\text{surface}, t=0} = \left(\frac{\partial}{\partial t} \phi^{(0)}(z_r, t_r) \right)_{\text{bulk}, t=0} \quad (19)$$

and, consequently,

$$\left(\frac{\partial}{\partial t} \phi^{(1)}(z_r, t_r) \right)_{\text{surface}, t=0} = \left(\frac{\partial}{\partial t} \phi^{(1)}(z_r, t_r) \right)_{\text{bulk}, t=0} \quad (20)$$

Let us start the search for the solutions by considering first the unperturbed problem. In order to obtain the solutions for $\phi^{(0)}(z_r, t_r)$ we perform the following change:

$$\phi^{(0)}(z_r, t_r) = e^{-\frac{1}{\lambda^2} \int_0^{t_r} f(\tilde{t}_r) d\tilde{t}_r} \psi(z_r, t_r) \quad (21)$$

This equation, when substituted in Eqs. (14), (15) and (16), yields

$$\frac{\partial}{\partial t_r} \psi(z_r, t_r) = K \frac{\partial^2}{\partial z_r^2} \psi(z_r, t_r) \quad (22)$$

and

$$\left(\frac{\partial}{\partial z_r} \psi(z_r, t_r) + u \psi(z_r, t_r) \right) \Big|_{z_r=-1/2} = \phi_s e^{\frac{1}{\lambda^2} \int_0^{t_r} f(\tilde{t}_r) d\tilde{t}_r} \quad (23)$$

$$\frac{\partial}{\partial z_r} \psi(z_r, t_r) \Big|_{z_r=0} = 0 \quad (24)$$

Now, we use the Green's function approach in order to obtain the solution to Eq. (21) subjected to the boundary conditions stated in Eqs. (22) and (23). The Green's function used here satisfies the differential equation

$$\left(\frac{\partial^2}{\partial z_r^2} - \frac{\partial}{\partial t_r} \right) \mathcal{G}(z_r, z'_r; t_r, t'_r) = \delta(z_r - z'_r) \delta(t_r - t'_r) \quad (25)$$

is subjected to the conditions

$$\left(\frac{\partial}{\partial z_r} \mathcal{G}(z_r, z'_r; t_r, t'_r) + u \mathcal{G}(z_r, z'_r; t_r, t'_r) \right) \Big|_{z_r=-1/2} = 0 \quad (26)$$

and

$$\frac{\partial}{\partial z_r} \mathcal{G}(z_r, z'_r; t_r, t'_r) \Big|_{z_r=0} = 0 \quad (27)$$

and is such that $\varsigma(z_r, z_r; t_r, t'_r)$, for $t_r < t'_r$ is given by

$$\mathcal{G}(z_r, z'_r; t_r, t'_r) = - \sum_{n=1}^8 \frac{e^{-k_n^2(t_r-t'_r)}}{\tilde{\mathcal{F}}'(k_n)} \left\{ \cos\left(k_n\left(\frac{1}{2} + z_r\right)\right) + \frac{u}{k_n} \sin\left(k_n\left(\frac{1}{2} + z_r\right)\right) \right\} \times \cos(k_n z_r) \theta(t_r - t'_r) \quad (28)$$

for $1/2 \leq z_r < z_r$, and

$$\mathcal{G}(z_r, z'_r; t_r, t'_r) = - \sum_{n=1}^8 \frac{e^{-k_n^2(t_r-t'_r)}}{\tilde{\mathcal{F}}'(k_n)} \left\{ \cos\left(k_n\left(\frac{1}{2} + z_r\right)\right) + \frac{u}{k_n} \sin\left(k_n\left(\frac{1}{2} + z_r\right)\right) \right\} \times \cos(k_n z_r) \theta(t_r - t'_r) \quad (29)$$

for $z_r < z_r \leq 0$, where $\theta(x)$ is the theta function, and, finally,

$$\tilde{\mathcal{F}}'(k_n) = \frac{1}{2k_n} \left(1 + \frac{u}{2}\right) \sin\left(\frac{k_n}{2}\right) + \frac{1}{4} \cos\left(\frac{k_n}{2}\right) \quad (30)$$

The k_n are determined by means of the equation: $k_n \sin(k_n/2) - u \cos(k_n/2) = 0$. By using the previous Green's function, the solution may be written as

$$\psi(z'_r, t_r) = - \int_{-1/2}^0 dz_r \Phi(z_r) \mathcal{G}(z_r, z'_r; t_r, 0) - u \phi_s \int_0^{t_r} dt'_r e^{\frac{1}{2} \int_0^{t'_r} f(\tilde{t}_r) d\tilde{t}_r} \bar{\mathcal{G}}(z'_r; t_r, t'_r) \quad (31)$$

$\mathcal{G}(z'_r, t_r, t'_r) = \varsigma(z_r, z'_r; t_r, t'_r)|_{z_r=-1/2}$. The director angle, $\phi^{(0)}(z_r, t_r)$, can be obtained by substituting Eq. (31) in Eq. (21), which leads us to

$$\begin{aligned} \phi^{(0)}(z'_r, t_r) = & -e^{-\frac{1}{2} \int_0^{t_r} f(\tilde{t}_r) d\tilde{t}_r} \int_{-1/2}^0 dz_r \Phi(z_r) \mathcal{G}(z_r, z'_r; t_r, 0) \\ & - u \phi_s \int_0^{t_r} dt'_r e^{-\frac{1}{2} \int_0^{t'_r} f(\tilde{t}_r) d\tilde{t}_r} e^{\frac{1}{2} \int_0^{t'_r} f(\tilde{t}_r) d\tilde{t}_r} \bar{\mathcal{G}}(z'_r; t_r, t'_r) \end{aligned} \quad (32)$$

The first correction for $\phi^{(0)}(z_r, t_r)$ due to the surface viscosity, $\phi^{(1)}(z_r, t_r)$, can be obtained from Eq. (12) subjected to Eq. (13). In particular, it is given by

$$\phi^{(1)}(z'_r, t_r) = \int_0^{t_r} dt'_r e^{-\frac{1}{2} \int_0^{t'_r} f(\tilde{t}_r) d\tilde{t}_r} e^{\frac{1}{2} \int_0^{t'_r} f(\tilde{t}_r) d\tilde{t}_r} \frac{\partial}{\partial t'_r} \phi^{(0)}(z'_r, t'_r) \Big|_{z'_r=-1/2} \bar{\mathcal{G}}(z'_r; t_r, t'_r) \quad (33)$$

which represents the solution with a first correction in v . By substituting these results in Eq. (9), we obtain

$$\begin{aligned} \phi(z_r, t_r) = & -e^{-\frac{1}{2} \int_0^{t_r} f(\tilde{t}_r) d\tilde{t}_r} \int_{-1/2}^0 dz_r \Phi(z_r) \mathcal{G}(z_r, z'_r; t_r, 0) \\ & - u \phi_s \int_0^{t_r} dt'_r e^{-\frac{1}{2} \int_0^{t'_r} f(\tilde{t}_r) d\tilde{t}_r} e^{\frac{1}{2} \int_0^{t'_r} f(\tilde{t}_r) d\tilde{t}_r} \bar{\mathcal{G}}(z'_r; t_r, t'_r) \\ & + \int_0^{t_r} dt'_r e^{-\frac{1}{2} \int_0^{t'_r} f(\tilde{t}_r) d\tilde{t}_r} e^{\frac{1}{2} \int_0^{t'_r} f(\tilde{t}_r) d\tilde{t}_r} \frac{\partial}{\partial t'_r} \phi^{(0)}(z'_r, t'_r) \Big|_{z'_r=-1/2} \bar{\mathcal{G}}(z'_r; t_r, t'_r) \end{aligned} \quad (34)$$

We restrict our analysis to the approximated solution (34) since the values of surface viscosity considered here are small enough.

4. Relaxation and Surface Viscosity

The formalism presented in the preceding sections permits us to explore the dynamical behavior of the NLC under the action of the external field, taking into account the effect of the surface viscosity in a perturbative manner. In this regard, Figure 1 illustrates the director angle profile for different values of viscosity by considering the Eq. (34), whereas the time dependence of the time derivative of the director angle, at $z_r = -1/3$, is shown in Figure 2 for three different values of dimensionless surface viscosity $\nu = \eta_s/(\eta_b d)$.

This figure shows the effect of the surface viscosity on the relaxation of the director angle profile for a significant value of the reduced anchoring energy $u = Wd/K$ which represents a very weak anchoring situation for the surfaces placed at $z_r = \pm 1/2$. Note that if ν increases, the relaxation of the system is slower when compared to the case of $\nu = 0$, *i.e.*, in the absence of surface viscosity. This feature can also be verified in Figure 1 which shows that for small values of ν , *i.e.*, values closed to the case $\nu = 0$, the system goes faster to the stationary situation imposed by the boundary conditions.

From the above results, we may investigate measurable physical properties of the NLC sample. For instance, in the case in which a linear polarized beam impinges

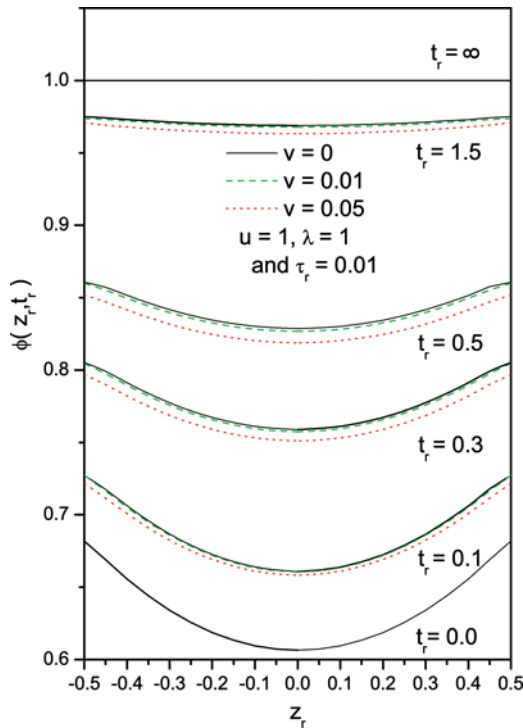


Figure 1. This figure illustrates the director angle profile $\phi(z_r, t_r)$ versus z_r for different values of t_r by considering some values of viscosity ν . In this figure, the initial and stationary (solid black lines) cases are also shown.

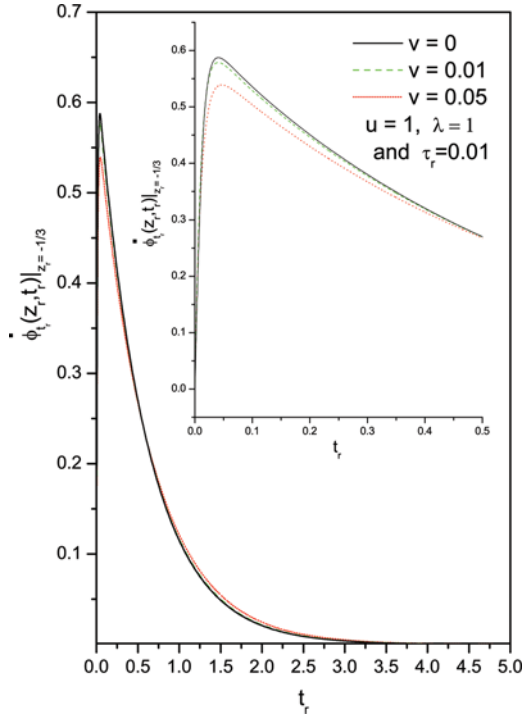


Figure 2. $\phi(z_r, t_r)$ versus t_r evaluated for $z_r = -1/3$ for some values of the surface viscosity v . This figure shows that for a small values of surface viscosity the director angle profile $\phi(z_r, t_r)$ relaxes faster than for large values of the surface viscosity.

normally on the nematic sample, the optical path difference Δl , between the ordinary and the extraordinary ray, is given by [23]

$$\Delta l = \int_{-\frac{d}{2}}^{\frac{d}{2}} \Delta n(\phi) dz \quad (35)$$

$$\Delta n(\phi) = n_{eff}(\phi) - n_0 = n_0 \left\{ \frac{1}{\sqrt{1 - r \sin^2(\phi)}} - 1 \right\} \quad (36)$$

with $r = 1 - (n_0/n_e)$, where n_0 and n_e are the ordinary and extraordinary refractive indices, respectively. Figure 3 illustrates Eq. (35) for different values the surface viscosity. Note that the values of Δl for $\nu = 0$ and $\nu = 0.01$ are essentially the same. For the case $\nu = 0.05$, the surface viscosity has a more significant effect on the system. This feature is also evidenced, for example, in Figures 1 and 2.

5. Discussions and Conclusions

We have analyzed the influence of the surface viscosity on the relaxation of the director angle profile by means of a perturbative method. We have shown that the

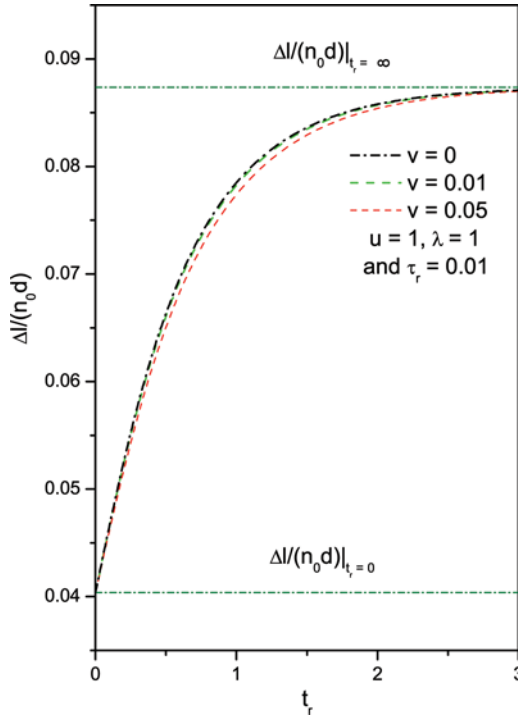


Figure 3. $\Delta l/(n_0 a)$ versus t_r to show the influence of the surface viscosity. We consider, for simplicity, $n_0 = 1.53$ and $n_e = 1.71$.

problem of the incompatibility between the time derivative on the surface and the bulk time derivative, at $t = 0$, for a given initial condition, may be avoided if the field is not removed in a discontinuous way. In addition, we have also shown that it is possible to use the perturbation theory in order to obtain approximated results for the director angle profile when the surface viscosity and the electric field are taken into account. The effects of the surface viscosity on molecular reorientation inside the NLC cell are evidenced by its influence on the space-time profile of the director angle and its derivatives, which show a slow relaxation of the profile when v is present. In addition, the measurable physical quantity represented by the optical path difference is also clearly influenced by the surface properties connected with the viscosity. It is then possible to conclude that the presence of the surface viscosity makes the system free from incompatibilities connected with the time derivative of the director angle in the surface and in the bulk. In addition, it furnishes a useful conceptual framework to investigate the dynamical behavior of the nematic director in the presence of time varying external fields.

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