

# Model for the electrorheological effect in flowing polymeric nematics

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An analytical model to study the response of a polymeric nematic confined in a rectangular cell, to a dc electric field is presented. The effect of a pressure-driven plane Poiseuille flow and its competition with the electric field is explicitly considered. For the final stationary state where the induced reorientation of the director has already occurred, an aligned structure with a greatly enhanced viscosity (electrorheological effect) is produced. For this same state the first normal stress difference is calculated as a function of position and of the applied field. For this quantity, regions of negative and positive values develop along the direction of the velocity gradient and an increase in the electric field or the flow causes an augmented effect in its profiles. The net force between the plates is also calculated. As the Reynolds number increases, it is found that it also changes sign from positive to negative, and the effect is more pronounced as the strength of the electric field increases. Finally, the paper is closed by discussing the scope and limitations of the model and methods employed. © 1999 American Institute of Physics. [S0021-9606(99)51416-5]

## I. INTRODUCTION

Recently there has been a great deal of interest in the use of rheological materials to manufacture a large variety of electrorheological (ER) devices. These materials are essentially fluids which are imbedded with particulate solids which react to an electrical field by producing dramatic and significant changes in their viscosity and other material properties. More precisely, an ER fluid undergoes a transition from a liquid state into a viscoelastic solidlike state upon the application of strong electric field (typically measured<sup>1</sup> in  $10^6$  V/m), which is characterized by a large reversible increase in their viscosity. The usual systems that exhibit this ER transition are composed of individual particles that become polarized by the applied field and align themselves into chains and filaments giving rise to a structure that is responsible for the gelationlike transition mentioned above. This occurs, for example, in concentrated suspensions, in solid particles in a dielectric medium or in some special polymeric fluids.<sup>2</sup>

Actually, some of these features are also present in nematic liquid crystals, which are fluids that exhibit long-range orientational order over distances many times larger than the dimensions of the molecules of which they are composed.<sup>3</sup> The intrinsic anisotropy of their molecules gives rise to macroscopic properties that are also anisotropic; this is the case, for instance, of the dielectric constant and the magnetic susceptibility. In fact, this intrinsic anisotropy is also responsible for considering liquid crystals as interesting fluids for

electrorheological applications. They offer obvious advantages over the more conventional ER fluids such as avoiding the problems associated with the settling of the dispersed phase; these complications are inexistent for liquid crystals since they are homogeneous phases. The feasibility of liquid crystal systems to produce a practical ER effect has been shown recently by Yang *et al.*,<sup>4</sup> who have observed an order of magnitude increase of the viscosity of a solution of a polymeric liquid crystal when acted upon by an external electric field in a rotational rheometer.

However, in spite of the large variety of ER devices that have been invented,<sup>1</sup> our understanding of the basic mechanisms responsible for the ER effect is, in general, rather poor. The basic purpose of this work is to study analytically a simple model for the reorientation of a thin film of the same polymeric nematic *poly(n-hexyl isocyanate)* (PHIC) used in by Yang *et al.*<sup>4</sup> under a constant electric field. Since its molecular weight is not too large ( $\sim 10^5$ ), to describe its dynamics we use, as a first approximation, hydrodynamic equations of motion for the director and the velocity fields which we have derived before for thermotropics.<sup>5</sup> In describing the realignment of the director we take explicitly into account the effects produced by an imposed plane Poiseuille flow. We show that the induced reorientation produces an increase of almost three orders of magnitude in the apparent viscosity of PHIC, which shows that the reorientation is the mechanism leading to the existence of the ER effect in this model.

To this end the paper is organized as follows. In the next section, Sec. II, we define the model and by assuming that the reorientation is a relaxation process, we derive a complete set of hydrodynamic equations for the director and the velocity field. Then in Sec. III, we specialize these equations

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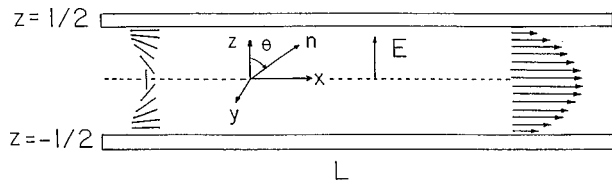


FIG. 1. Schematics of a planarly aligned liquid crystal film in the presence of a constant electric field. The velocity profile of a plane Poiseuille flow is also shown.

for a particular pressure-driven flow (plane Poiseuille) and derive an equation for the stationary orientational profile. We solve this nonlinear equation in an approximate but analytical way by using boundary layer methods. Using this solution, in Sec. IV we first calculate the apparent (orientational) viscosity as a function of position within the cell and then we average it over the whole cell. A plot of this averaged viscosity as a function of the dimensionless external field, shows a large increase (electrorheological effect) for values of the applied field which are about 10 times the critical field. We then calculate the first normal stress difference,  $N_1$ , as a function of position, the applied field and the Reynolds' number of the imposed flow. We find that  $N_1$  develops positive and negative values in different regions within the cell along the direction of the velocity gradient, and that an increase in the electric field or in the flow intensity produces an increase in  $N_1$ , but keeping the same qualitative behavior. It is worth emphasizing that, to our knowledge, this type of behavior of  $N_1$  has not been predicted at a hydrodynamic level of description before. Then the force on the plates as a function of the Reynolds' number for different values of the electric field is also calculated. For sufficiently slow flows, the force is always positive, but at high fluid velocities the force is always negative over all the electric field strengths considered. Finally, we close the paper by discussing the limitations and advantages of our approach.

## II. MODEL AND GOVERNING EQUATIONS

Consider a quiescent nematic layer of thickness  $l$  contained between two parallel conducting plates, as depicted in Fig. 1. It is assumed that the transverse dimensions,  $L$ , along the  $x$  and  $y$  axes are large compared to  $l$ ,  $L \gg l$ , so that the cell has a large aspect ratio but a finite volume  $V = L^2 l$ . The initial orientation of the director is planar and, therefore, when an external dc electric field  $E$  is applied along the  $z$  direction, the director  $\vec{n}$  will reorient inside the cell for values of  $E > E_c$ , where  $E_c$  is the critical field that has to be exceeded to initiate the reorientation. Owing to the low aspect ratio of the cell, it is reasonable to assume that the reorientation occurs in the  $x-z$  plane. Moreover, spatial homogeneity in the  $x$  direction can be also assumed to simplify the description and as a result  $\vec{n} = [\sin \theta(z,t), 0, \cos \theta(z,t)]$ . We shall assume that the reorientation angle  $\theta$  satisfies strong anchoring conditions at the plates

$$\theta\left(z = \pm \frac{l}{2}\right) = \pm \frac{\pi}{2}. \quad (1)$$

In addition to the electric field we assume that a constant pressure gradient is maintained along the  $x$  axis. If the reorientation occurs in the  $x-z$  plane, it is also feasible to assume that the hydrodynamic flow will also take place in the same plane giving rise to the plane Poiseuille velocity profile shown in Fig. 1. Then the only relevant component of the velocity field is  $v_x$  which will be assumed to satisfy no-slip boundary conditions at the plates

$$v_x\left(z = \pm \frac{l}{2}\right) = 0. \quad (2)$$

Now, if as usual the reorientation of the director is considered to be an isothermal process, its equilibrium states may be described in terms of the associated Helmholtz free energy functional, which for the assumed geometry and in MKS units takes the form

$$F = \int_V dV \left\{ \frac{K}{2} \left( \frac{d\theta}{dz} \right)^2 - \frac{E^2}{2} \epsilon_0 (\epsilon_{\perp} + \epsilon_a \cos^2 \theta) + \frac{1}{2} \rho v_x^2(z) \right\}. \quad (3)$$

Here  $\epsilon_a \equiv \epsilon_{\parallel} - \epsilon_{\perp}$  stands for the dielectric anisotropy of the nematic,  $\epsilon_0$  denotes the permittivity of the vacuum and  $\rho(z,t)$  denotes its local mass density. The last term in this equation represents the contribution to  $F$  due to the external pressure gradient along the  $x$  direction. It should be stressed that in writing this expression, for simplicity, we have made the assumption of equal elastic constants for the splay, bend and twist elastic deformations,  $K \equiv K_1 = K_2 = K_3$ .

Following the usual procedure to derive nematodynamic equations,<sup>3,6</sup> from Eq. (3) we arrive at the following set of coupled dynamical equations<sup>7</sup> for  $\theta$  and  $v_x$

$$\frac{\partial \theta}{\partial t} = - \frac{2}{\gamma_1} \frac{\delta F}{\delta \theta} - (1 - \lambda) \cos \theta \frac{\partial v_x}{\partial z}, \quad (4)$$

$$\frac{\partial v_x}{\partial t} = \frac{\nu_3}{\rho} \frac{\partial^2 v_x}{\partial z^2} + \frac{\lambda - 1}{2\rho} \frac{\partial}{\partial z} \left[ \frac{1}{\cos \theta} \frac{\delta F}{\delta \theta} \right] - \frac{1}{\rho} \frac{\partial p}{\partial x}. \quad (5)$$

The variational derivative  $\delta F / \delta \theta$  is given explicitly by

$$\frac{\delta F}{\delta \theta} = K \frac{d^2 \theta}{dz^2} - \frac{\epsilon_0 \epsilon_a}{2} E^2 \sin 2\theta, \quad (6)$$

and  $\gamma_1$ ,  $\gamma_2$ ,  $\nu_3$ , with  $\lambda \equiv \gamma_1 / \gamma_2$ , denote the various viscosity coefficients of the nematic. Note that the last term in Eq. (5) represents the externally applied pressure gradient along the  $x$  direction.

At this point it should be emphasized that, strictly speaking, Eqs. (4)–(6) provide for a closed set of hydrodynamic equations for a low molecular weight nematic (thermotropic), since in this case the director is the only additional hydrodynamic variable, apart from the usual conserved variables of mass, specific entropy, and momentum densities.<sup>8</sup> However, the corresponding description for a polymeric nematic (lyotropic) is much more involved owing to the large number of degrees of freedom that may contribute to the dynamics in the hydrodynamic limit. But since a complete formulation of a hydrodynamic description for arbitrary lyotropics is still an open issue, as a first approximation we shall use the above formalism to describe the hydrodynamic behavior of a polymeric nematic solution of PHIC, as de-

scribed by Yang.<sup>4</sup> It is important to stress that this is, indeed, a strong approximation; however, it is expected to be a reasonable one owing to the fact that molecular weight of PHIC (~10<sup>5</sup>) is not too large. If in addition, the system is always away from a critical point, it is not necessary to account for the dynamics of an order parameter, which is not a hydrodynamic variable, and therefore, the hydrodynamic behavior can be described in terms of the director field only. This approximation has the great advantage of keeping the description simple enough so that an analytical treatment is possible and specific calculations can be carried out.

Among all the possible nonequilibrium states the system might have, here we only consider the final stationary state characterized by the fact that the reorientation has already occurred, but flow effects are still present. For this case the calculation of some rheological properties such as the viscometric functions, can be carried out explicitly,<sup>9</sup> as we shall see below. The final stationary state is defined by setting the left hand sides of Eqs. (4) and (5) equal to zero. Thus, from Eq. (5) we arrive at

$$\frac{d^2 v_x(\zeta)}{d\zeta^2} = \frac{l}{\nu_3} (\nabla p)_{\text{ef}}, \quad (7)$$

where  $(\nabla p)_{\text{ef}}$  is an effective pressure gradient defined by

$$(\nabla p)_{\text{ef}} \equiv \frac{Q}{\rho l L} = \frac{\nu_3}{2} \frac{dp(\chi)}{d\chi} \left[ \nu_3 + \frac{(\lambda - 1)^2 \gamma_1}{4} \right]^{-1}, \quad (8)$$

$Q$  is the mass flow rate and we have introduced the dimensionless variables  $\zeta \equiv z/l$  and  $\chi \equiv x/l$ . If Eq. (7) is solved for the boundary conditions (2) and if this solution is inserted into Eq. (4) setting  $\partial\theta/\partial t = 0$ , this would yield a closed equation for the final stationary orientational configuration. We shall carry out this procedure explicitly for a particular pressure driven flow, namely, the plane Poiseuille flow.

### III. ORIENTATIONAL CONFIGURATION FOR PLANE POISEUILLE FLOW

Let us assume that the external pressure gradient is constant,  $dp(x)/dx \equiv \Delta p/L = \text{const}$ . Then the solution of Eq. (7) is given by

$$v_x(\zeta) = \frac{1}{\nu_3} (\nabla p)_{\text{ef}} (1/4 - \zeta^2). \quad (9)$$

This defines the well known parabolic velocity profile of the plane Poiseuille flow. If we now substitute this solution into Eq. (4) with  $\partial\theta/\partial t = 0$ , we arrive at the following closed, nonlinear and dimensionless equation for the final orientational state:

$$\frac{d^2 \theta}{d\zeta^2} + q\bar{E}^2 \sin 2\theta + 6N\zeta \cos \theta = 0. \quad (10)$$

Here  $\bar{E} \equiv E/E_c$ , where  $E_c = \pi/l\sqrt{2K/\epsilon_a \epsilon_0}$  is the critical field,<sup>10</sup> such that reorientation occurs if  $\bar{E} > 1$ . It is essential to point out that there are two important physical parameters in Eq. (10). On the one hand,  $q \equiv \epsilon_0 \epsilon_a \bar{E}^2 l^2 / 2K$ , is proportional to the ratio between the energy of the incident field and the nematic's elastic energy. Therefore, it is a measure of the strength of the coupling between the external field and

the induced orientational configuration. On the other hand, the parameter  $N \equiv 6R\nu_3\gamma_1(\lambda - 1)/K\rho$  contains the effects due to the hydrodynamic flow through the Reynolds number  $R \equiv \rho l^2 (\nabla p)_{\text{ef}} / \nu_3^2$ .

In a previous work,<sup>9</sup> Eq. (10) was solved exactly in an analytical way for the case of strong fields ( $q\bar{E}^2 > 1$ ) and in the absence of flow,  $N = 0$ . There it was found that  $\theta(s)$  varies abruptly in the cell's center vicinity, while it is almost two pieces of horizontal lines in the right and left parts of the cell. This singular behavior is the result of the competition between the strong external field and the hard-anchoring boundary condition. Since exact solutions of Eq. (10) when  $N \neq 0$  are, in general, difficult to obtain, the presence of the dimensionless parameters  $q$  and  $N$  in Eq. (10) allows for the possibility of carrying out systematic and analytic expansions in powers of them. However, instead of performing these expansions, in this work we shall obtain approximate analytical solutions of Eq. (10) based on the following ideas.

As mentioned for the case  $N = 0$ , it is to be expected that in the bulk of the cell the electric energy would be much larger than the nematic's elastic energy, so that  $q \gg 1$ . In contrast, near the solid boundaries, the opposite behavior is expected,  $q < 1$ . These different physical situations and behaviors of the solutions of Eq. (10) may be modelled by considering these two regions as boundary layers,  $\theta_{\text{out}}(z)$  and  $\theta_{\text{in}}(z)$ .<sup>11,12</sup> Then, these solutions have to be "matched" or joint asymptotically in order to obtain a valid solution,  $\theta_{\text{adj}}$ , in the whole domain. Thus, the adjusted solution  $\theta_{\text{adj}}$  is given by

$$\theta_{\text{adj}} = \theta_{\text{out}} + \theta_{\text{in}} - \theta_{\text{match}}, \quad (11)$$

where  $\theta_{\text{match}}$  must be determined so that  $\theta_{\text{out}}(z)$  and  $\theta_{\text{in}}(z)$  are joint asymptotically. To apply this method to the problem under consideration, note that if  $q\bar{E}^2 \gg 1$  and  $N \gg 1$ , with  $q\bar{E}^2 \sim N$ , Eq. (10) reduces to

$$2q\bar{E}^2 \sin \theta_{\text{out}} + 6N\zeta = 0. \quad (12)$$

This approximated equation is no longer a second-order differential equation and hence its solution

$$\theta_{\text{out}} = -\arcsin(3N\zeta/q\bar{E}^2), \quad (13)$$

is not able to satisfy both boundary conditions, Eq. (1). This means that the angle near the borders must vary within a faster spatial scale than in the center of the cell, in such a way that the corresponding boundary condition will be fulfilled. If we choose  $\mu \equiv \sqrt{q\bar{E}^2}\zeta$  as a fast variable and rewrite Eq. (10), we obtain

$$\frac{d^2 \theta}{d\mu^2} + \sin 2\theta + \frac{6N\mu}{(q\bar{E}^2)^{3/2}} \cos \theta = 0. \quad (14)$$

But since  $N \sim q\bar{E}^2$ , this equation for  $q\bar{E}^2 \gg 1$  reduces to

$$\frac{d^2 \theta_{\text{in}}}{d\mu^2} + \sin 2\theta_{\text{in}} = 0, \quad (15)$$

and its asymptotic solution, which satisfies the boundary condition  $\theta(\zeta = \frac{1}{2}) = \pi/2$ , is given by

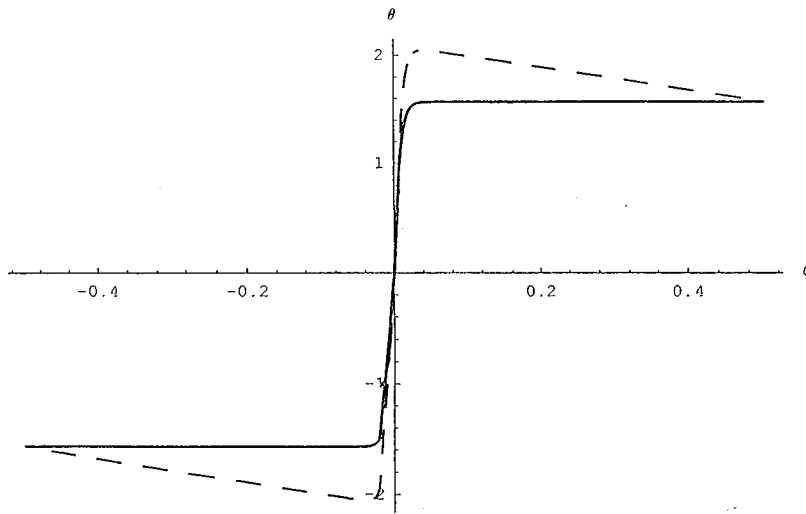


FIG. 2. Orientational configuration  $\theta$  vs  $\zeta$  of PHIC as calculated from Eq. (18). (—)  $N=0$ ; (---)  $N=106(q\bar{E}^2=160)$ .

$$\theta_{\text{in}} = \frac{\pi}{2} - 2 \arctan(Ae^{-\mu}), \quad (16)$$

where  $A$  is a constant to be determined. If the same procedure is followed for the other boundary condition, we find that  $\theta_{\text{match}}$  is given by

$$\theta_{\text{match}} = \pi/2 - 2 \arctan A = \mp \arcsin(3N/2q\bar{E}^2), \quad (17)$$

where the double sign  $\mp$  corresponds to applying the condition to the right and left plates, respectively.

Finally, substitution of  $\theta_{\text{out}}$ ,  $\theta_{\text{in}}$ , and  $\theta_{\text{match}}$  into Eq. (11) leads to

$$\begin{aligned} \theta(\zeta; N, q\bar{E}^2) &= \pi/2 + \text{sign}(\zeta) \arcsin\left(\frac{3N\zeta}{2q\bar{E}^2}\right) - \arcsin\left(\frac{3N}{q\bar{E}^2}\right) \\ &\quad - 2 \arctan\left(e^{-\zeta} \tan\left\{\pi/4 + \frac{1}{2} \text{sign}(\zeta)\right.\right. \\ &\quad \left.\left. \times \arcsin\left[\frac{3N\zeta}{2q\bar{E}^2}\right]\right\}\right), \end{aligned} \quad (18)$$

where  $\text{sign}(\zeta)$  is the sign function. We should mention that even though  $\theta$  is defined in terms of this discontinuous functions, it is easy to show that  $\theta$  and  $d\theta/d\zeta$  are continuous functions, while  $d^2\theta/d\zeta^2$  has a finite discontinuity at  $\theta=0$ . The plot of  $\theta$  versus  $\zeta$  as given by Eq. (18), is shown in Fig. 2 for the cases in which the flow is in the positive  $x$  direction and when the flow is such that  $N \sim 106$ , which corresponds to Reynolds numbers in the range  $10^{-1} - 10^{-2}$ . As mentioned earlier, this curve indeed shows that  $\theta(\zeta)$  has a sharp increase in a very narrow region around the central part of the cell for large values of  $q$ , that is,  $q\bar{E}^2 > 10$ , and it becomes sharper as  $q$  increases.

## IV. VISCOMETRIC FUNCTIONS

### A. Electrorheological effect

The viscosity function or apparent viscosity connects the shearing force per unit area and the magnitude of the local shear. It depends on the orientation of the director through<sup>13</sup> the expression

$$\eta(\theta(\zeta); \bar{E}) = \alpha_1 \cos^2 \theta \sin^2 \theta + \eta_c + (\alpha_2 + \alpha_3) \sin^2 \theta, \quad (19)$$

where  $\alpha_1$ ,  $\alpha_2$ ,  $\alpha_3$  are the Leslie coefficients<sup>14</sup> and  $\eta_c$  is the transverse Miesowicz viscosity.<sup>15</sup> Since the orientation angle  $\theta$  is given by Eq. (18), from the above equation it follows that the dependence of  $\eta$  on  $\theta$  indicates that the system is non-Newtonian in its behavior, in the sense that  $\eta$  is strongly dependent on the driving force.

From Eqs. (19) and (18) we obtain the spatial variation of  $\eta$  within the cell. Figure 3 shows a plot of  $|\eta(\zeta, \bar{E}) - \eta(\zeta = \pm \frac{1}{2}, \bar{E})|$  vs  $\zeta$  for PHIC for different values of  $q\bar{E}^2$  and  $N$  in units of Pa s. The required viscosity coefficients and other material parameters of PHIC<sup>4</sup> are taken as  $\alpha_1 = -1100$  Pa s,  $\alpha_2 = -3700$  Pa s,  $\alpha_3 = 320$  Pa s,  $\eta_c = 2100$  Pa s, and  $\epsilon_a \epsilon_0 = 6.19 \times 10^{-9} N/V^2$ . Here we have also taken  $K \sim 10^{-12} N$  for the elastic constant and  $l = 4 \times 10^{-6}$  m for the separation distance between the plates. In this case it turns out that  $E_c = 4490$  V/m.<sup>10</sup>

Now, in order to exhibit that an electrorheological effect does exist in the proposed model, we first substitute Eq. (18) into (19) and average the result over  $\zeta$ . This yields the averaged apparent viscosity  $\bar{\eta}(\bar{E}) \equiv \int_{\zeta=-1/2}^{\zeta=1/2} \eta(\theta; \bar{E}) d\zeta$ . If we now plot  $|\bar{\eta}(\bar{E}) - \bar{\eta}(\bar{E}=0)|$  vs  $\bar{E}$ , so that the curve starts at the origin, we arrive at the curves shown in Fig. 4. These curves show that the PHIC nematic solution exhibits a significant electrorheological effect, which is evidenced by the sharp increase of almost three orders of magnitude in its apparent viscosity. To estimate the corresponding value of the applied field, take a saturation value  $q\bar{E}^2 = 10$  in Fig. 4. If we take  $q = 10^{-1}$ , which corresponds to a strong interaction between the electric field and the nematic, this implies that

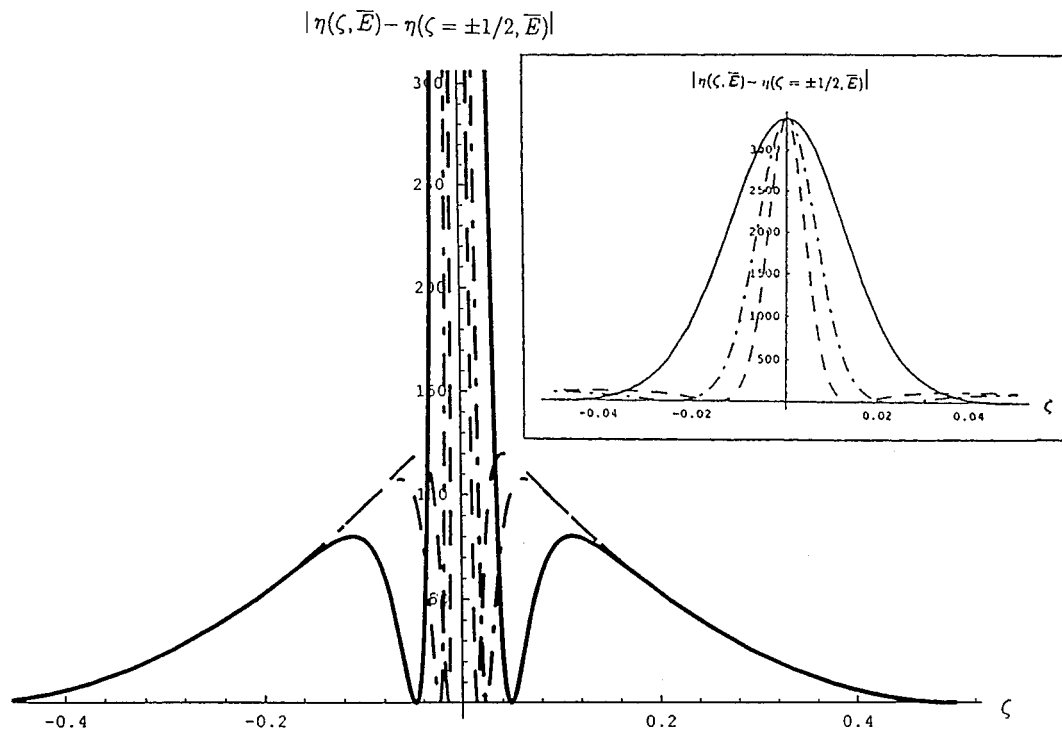


FIG. 3. Apparent viscosity  $|\eta(\zeta, \bar{E}) - \eta(\zeta = \pm 1/2, \bar{E})|$  in units of Pa s vs  $\zeta$  for different values of  $q\bar{E}^2$  and  $N$ , (—)  $N=50$ ; (- - -)  $N=100$ ; (- · -)  $N=160$ . The inset shows the maximum occurring at the center of the cell corresponding to values one order of magnitude larger.

$\bar{E} = 10$  or  $E = 10E_c$ . This shows that for rather small applied fields,  $\sim 45 \times 10^3$  V/m, a strong electrorheological effect may be produced in the cell.

**B. First normal stress difference**

One of the distinctive phenomena observed in the flow of liquid crystal polymers in the nematic state is that of a negative steady-state first normal stress difference,  $N_1$ , in shear flow over a range of shear rates.  $N_1$  is zero or positive for isotropic fluids at rest over all rates of shear, which means that the force developed due to the normal stresses, tends to push apart the two surfaces between which the material is sheared. In liquid crystalline solutions, positive normal stress differences are found at low and high shear rates, with negative values occurring at intermediate shear rates.<sup>16</sup>

On the other hand, Marrucci *et al.*<sup>17,18</sup> have solved a two-dimensional version of the Doi model for nematics,<sup>19</sup> in which the molecules are assumed to lie in the plane perpendicular to the vorticity axis, that is, in the plane parallel to both, the direction of the velocity and the direction of the velocity gradient. Despite this simplification, the predicted range of shear rates over which  $N_1$  is negative, is in excellent agreement with observations. This result opens up the possibility that negative first normal stress differences may be predicted in a two dimensional flow. Indeed, in this section we shall show that over a range of Reynolds' numbers, negative values of  $N_1$  are predicted for the plane Poiseuille flow, taking into account the effect of an external electric field.

We shall now examine the effects produced by the stresses generated during the reorientation process by calcu-

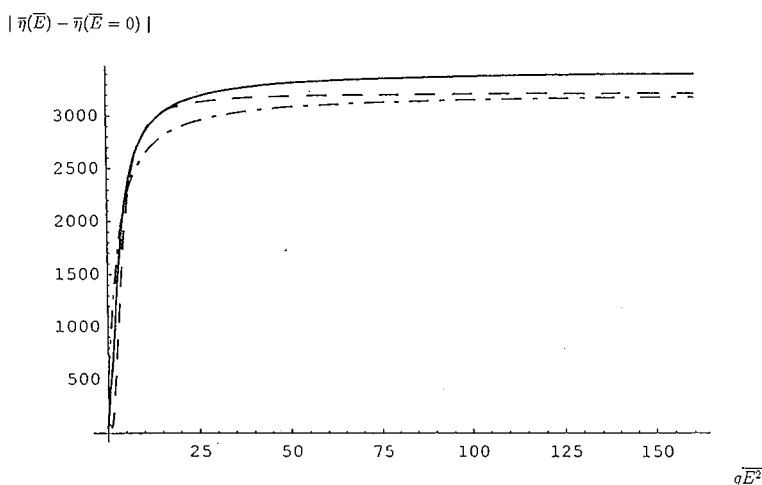


FIG. 4. The same apparent viscosity as in Fig. 3 vs  $q\bar{E}^2$  for PHIC.  $N$  takes the same values as in Fig. 3.

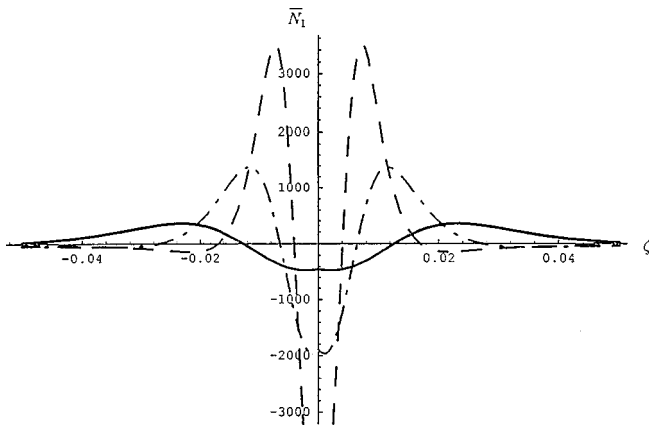


FIG. 5. Primary dimensionless normal stress differences  $\bar{N}_1 \equiv N_1/K$  vs  $\zeta$  as calculated from Eq. (22) for different values of  $q\bar{E}^2$  and  $N$  such that  $N/q\bar{E}^2 = \frac{1}{3}$ .  $N$  takes the same values as in Fig. 3.

lating the viscometric functions which relate the shear and normal stress differences. For the geometry under consideration and using the convention in Ref. 20, the first normal stress difference is defined by

$$N_1 \equiv \sigma_{xx} - \sigma_{zz}, \quad (20)$$

where  $\sigma_{ij}$  are the components of the stress tensor of the nematic used by De Gennes.<sup>3</sup>

$$\begin{aligned} \sigma_{ij} = & \alpha_1 n_i n_j n_\mu n_\rho A_{\mu\rho} + \alpha_2 n_i \Omega_j + \alpha_3 n_j \Omega_i, \\ & \alpha_4 A_{ij} + \alpha_5 n_i n_\mu A_{\mu j} + \alpha_6 n_i n_\mu A_{\mu j}. \end{aligned} \quad (21)$$

Here  $A_{ij} \equiv (\frac{1}{2})(\partial v_j / \partial x_i + \partial v_i / \partial x_j)$  is the symmetric part of the velocity gradient  $\partial v_i / \partial x_j$  and  $\vec{\Omega} \equiv d\hat{n}/dt - (\frac{1}{2})\nabla \times \vec{v} \times \hat{n}$  represents the rate of change of the director with respect to the background fluid. The  $\alpha_i$  for  $i = 1, \dots, 6$ , denote the Leslie coefficients of the nematic. Substitution of Eq. (18) into Eqs. (20) and (21) leads to

$$\begin{aligned} N_1(\theta(\zeta)) = & \nu_3 (\nabla p)_{e\zeta} (\nu_1 + \nu_2 - 2\nu_3) \sin 4\theta \\ & - \lambda \frac{K}{d^2} \left[ \sin 2\theta \frac{d^2 \theta}{d\zeta^2} + \cos 2\theta \left( \frac{d\theta}{d\zeta} \right)^2 - q \cos^2 \theta \right]. \end{aligned} \quad (22)$$

The quantities  $\nu_1$  and  $\nu_2$  also denote several viscosity coefficients. A plot of  $N_1$  vs  $\zeta$  for several values of  $q\bar{E}^2$  and  $N$  is shown in Fig. 5. In nonhomogeneous plane Poiseuille flow between two parallel surfaces, the velocity gradient is not constant and varies between zero at the center of the cell and up to a maximum value at the walls. According to Marrucci's results,<sup>17</sup> it is likely that regions of negative and positive first normal stress differences will develop along the direction of the velocity gradient. In fact, in Fig. 5 negative values of  $N_1$  are shown in the central region of the cell, becoming positive as the velocity gradient increases and approaching zero near the plates region, where the nematic lies parallel to the plates in agreement with the chosen strong anchoring boundary conditions. An increase in the electric field pro-

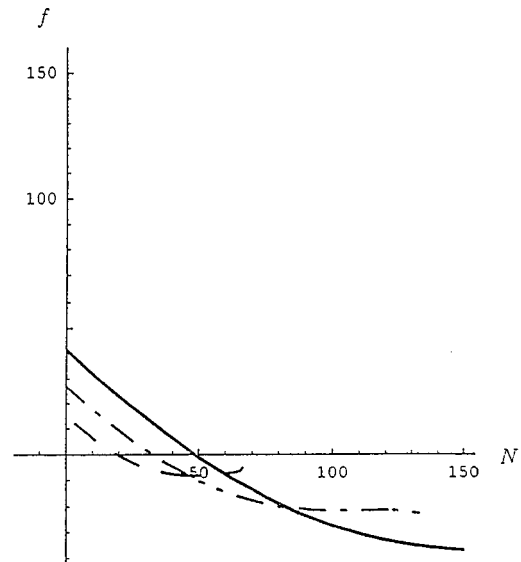


FIG. 6. The dimensionless force  $f \equiv (\int_{-1/2}^{1/2} N_1(\zeta) d\zeta) / K$  in units of the elastic constant  $K = 10^{-12} N$  as a function of  $N$  for different values of  $q\bar{E}^2$ . (—)  $q\bar{E}^2 = 50$ ; (- · -)  $q\bar{E}^2 = 100$ ; (- - -)  $q\bar{E}^2 = 160$ .

duces an increase in  $N_1$ , but keeps the same qualitative behavior, as observed in Fig. 5. It should be stressed that the chosen values of  $q\bar{E}^2$  and  $N$  must be of the same order, so that the boundary layer method discussed previously is applicable.

In Figs. 6 and 7, the integration of the first normal stress difference profile,  $\int_{-1/2}^{1/2} N_1(\theta(\zeta)) d\zeta$ , over the whole cell and along the velocity gradient direction renders the net force between the plates as a function of the Reynolds' number, which is proportional to  $N$ , and to the electric field. A positive force exerted by the fluid motion tends to push the plates apart, or otherwise, if the force is negative, the fluid tends to pull the plates close together. As observed in Fig. 6, as the Reynolds' number increases, the net force changes sign from positive to negative, and the effect is more pronounced as the electric field increases in magnitude. In Fig. 7, the force is plotted as a function of the electric field strength for several values of  $N$  (or  $R$ ). For sufficiently slow flows, the force is always positive, but at high fluid velocities the force is always negative over all the electric field strengths considered. The results shown in these figures do not exclude the possibility that a further increase in the fluid velocity will induce another change of sign in the force, becoming positive at very high velocity gradients. These prediction would be in accordance with the observed and predicted behavior of liquid crystal solutions in simple shear flow.<sup>17,21</sup> It should be remarked that in Figs. 6 and 7 we have plotted the dimensionless force  $f$  in units of the elastic constant  $K = 10^{-12}$  Newtons.

## V. DISCUSSION

We have shown that an electrorheological effect may be produced in a cell of a few microns thickness containing a

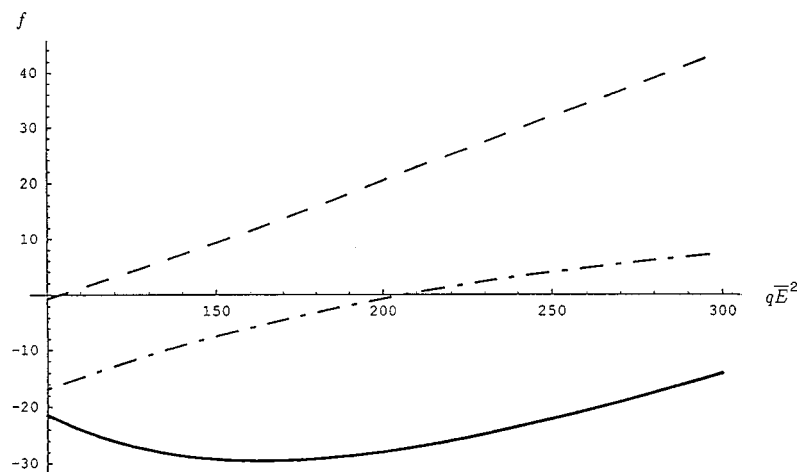


FIG. 7.  $f$  vs  $q\bar{E}^2$  for different values of  $N$ . (—)  $N = 50$ ; (- · -)  $N = 100$ ; (- - -)  $N = 160$ .

PHIC nematic. To clarify and elaborate on this result we close the paper with the following comments.

First, in our model the mechanism that produces the increase in the apparent viscosity is the induced reorientation of the director by the applied electric field. This occurs because the permanent and induced dipole moments of PHIC align themselves with the field giving rise to an enhanced viscosity. As a matter of fact, this orientational distortion also manifests itself as a nonlinear optical process, in the sense that the external field gives rise to a refraction index gradient which produces self-focusing and waveguiding effects in the cell.<sup>5</sup> Although the existence of an electrorheological effect for PHIC have already shown to exist in a rotational rheometer by Yang and Shine,<sup>4</sup> here we have shown that the same effect might be induced in a small nematic planar cell with a few microns separation between its plates.

Secondly, it should be emphasized that our analysis was restricted to the final stationary state of the reorientation. A more general and physically richer situation would be to consider nonstationary states defined by the full equation [Eq. (10)] with  $N \neq 0$ . In this case, Eq. (10) cannot be solved exactly in an analytical form and one has to resort to the use of numerical methods. From this point of view, preliminary calculations based on the use of the well-known shooting method to solve Eq. (10), indicate that Eq. (10) can be solved only for values of  $q$  such that  $q\bar{E}^2 < 10$ . For larger values the numerical method becomes unstable and a much larger numerical precision is required. This reinforces the convenience of developing well defined analytic and asymptotic approximations to solve Eq. (10). However, even if this numerical analysis can be carried out explicitly, the simplicity and the analytical treatment of the model presented here would be lost. In spite of its simplicity, this model predicts an experimentally verified result for a different and new physical situation.

Thirdly, with regard to the reported behavior of the first normal stress differences  $N_1$ , it should be mentioned that Marrucci *et al.*<sup>17,18</sup> for the first time linked these normal stress differences to a tendency of the direction of average molecular orientation to tumble in shear flow about the vorticity axes, and to the arrest of the tumbling that eventually

occurs as the shear rate is increased. In this work, although we have not discussed the tumbling regime, we have obtained the same qualitative behavior but using a hydrodynamic description instead of a stochastic approach.

Finally, it should be pointed out once more that our theoretical description is approximated, since all the effects associated with possible additional hydrodynamic variables required to describe properly the hydrodynamics of PHIC have been entirely neglected. However, our description is simple, analytic, and predicts the existence of an interesting physical effect.

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<sup>1</sup> *Electrorheological Fluids*, edited by R. Tao (World Science, New York, 1992).

<sup>2</sup> H. Block and J. P. Kelly, *J. Phys. D* **21**, 1661 (1988).

<sup>3</sup> P. G. de Gennes, *The Physics of Liquid Crystals* (Clarendon, Oxford, 1964).

<sup>4</sup> I. K. Yang and A. D. Shine, *J. Rheol.* **36**, 1079 (1992).

<sup>5</sup> R. F. Rodríguez and J. A. Reyes, *J. Nonlinear Opt. Phys. Mater.* **4**, 943 (1995).

<sup>6</sup> M. San Miguel and F. Sagués, *Phys. Rev. A* **36**, 1883 (1987).

<sup>7</sup> R. F. Rodríguez, P. Ortega, and R. Díaz-Urbe, *Physica A* **230**, 118 (1996).

<sup>8</sup> R. F. Rodríguez and J. A. Reyes, *Mol. Cryst. Liq. Cryst.* **282**, 287 (1996).

<sup>9</sup> R. F. Rodríguez and J. E. Camacho, *Rev. Mex. Fis.* **44**, 1859 (1998).

<sup>10</sup> I. C. Khoo, *Nonlinear Optics of Liquid Crystals, Progress in Optics, Vol. 26*, edited by E. Wolf (North Holland, Amsterdam, 1988).

<sup>11</sup> C. M. Bender and M. C. Arszog, *Advanced Mathematical Methods for Scientists and Engineers* (McGraw-Hill, New York, 1978).

<sup>12</sup> H. Schlichting, *Boundary Layer Theory* (McGraw-Hill, New York, 1968).

<sup>13</sup> T. Carlsson, *Mol. Cryst. Liq. Cryst.* **104**, 307 (1984).

<sup>14</sup> O. Parodi, *J. Phys. (Paris)* **31**, 581 (1970).

<sup>15</sup> M. Miesowicz, *Nature (London)* **17**, 261 (1935).

<sup>16</sup> G. Kiss and R. S. Porter, *J. Polym. Sci., Part C: Polym. Symp.* **65**, 193 (1978).

<sup>17</sup>G. Marrucci and P. L. Maffettone, *Macromolecules* **22**, 4076 (1989).

<sup>18</sup>R. G. Larson, *Macromolecules* **23**, 3983 (1990).

<sup>19</sup>M. Doi and S. F. Edwards, *The Theory of Polymer Dynamics* (Oxford University Press, New York, 1986).

<sup>20</sup>R. B. Bird, R. C. Armstrong, and O. Hassager, *Dynamic of Polymer Liquids* (Wiley, New York, 1977), Vol. 1.

<sup>21</sup>S. Grisafi and P. O. Brunn, *J. Rheol.* **33**, 47 (1989); P. Biller and F. Petruccione, *J. Non-Newtonian Fluid Mech.* **25**, 347 (1987).