

## POLYMERS. LIQUID CRYSTALS

### Effect of a bounding surface on the rotational viscosity of liquid crystals

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The effect of a surface, bounding a nematic-liquid phase, on the rotational viscosity  $\gamma_1$  is investigated on the basis of a statistical approach employing direct correlation functions. Specific calculations are performed for a model system consisting of ellipsoidal molecules interacting via a Gay–Berne potential near the bounding surface. © 1998 American Institute of Physics. [S1063-7834(98)03610-7]

The theoretical description of dissipation processes in liquid crystals (LCs) is once again attracting investigators.<sup>1</sup> Despite the fact that certain qualitative advances have been made in the construction of a molecular theory of the rheological properties of nematic-liquid crystals (NLCs), it is still too early to talk about the development of a theory which would make it possible to describe the rheological processes from first principles, based only on the form of the Hamiltonian.

But, since the coefficient of rotational viscosity  $\gamma_1$  is one of the parameters determining the dynamics of LCs, and extensive experimental data on the viscosity of LCs have now been accumulated,<sup>2</sup> the question of a theoretical interpretation of these data inevitably arises.

For the most part, this problem was solved on the one hand on the basis of either a simplified molecular model of the free volume<sup>3</sup> or a theory based on the Fokker-Planck equation for a model of NLCs in a polymer solution<sup>4</sup> or by using the same Fokker–Planck kinetics to estimate the coefficient of rotational viscosity,<sup>5</sup> and on the other hand by the nonequilibrium statistical operator (NSO) method,<sup>6</sup> taking account of different correlations to describe the rheological processes in NLCs.<sup>7</sup>

These approaches to describing rotational viscosity in NLCs have the serious drawback that the approximations introduced into the theory do not fall into a definite hierarchy and they serve only to resolve particular difficulties. Ultimately, this even precludes making comparisons of the final results. Another weak point of all approaches, without exception, is the large number of adjustable parameters.

The objective of the present work is to investigate the effect of a bounding surface on the rotational viscosity of liquid crystals on the basis of a treatment combining the Nemtsov model<sup>7</sup> and the Brook–Levinson–Zakharov model.<sup>8</sup> The orientational distribution function required for the final calculations of  $\gamma_1$  can be obtained on the basis of an independent statistical theory<sup>9</sup> or by molecular-dynamics methods.<sup>10</sup> The theoretically established relation between  $\gamma_1$  and the order parameters of the LC system, in turn, makes it

possible to describe the temperature dependence of the coefficient of rotational viscosity.

#### 1. BASIC RESULTS OF THE STATISTICAL THEORY OF VISCOSITY

The dissipation energy of nematic-liquid crystals is determined by the viscous-stress tensor

$$\sigma = \alpha_1(\mathbf{nn}:A)nn + \alpha_2\mathbf{nN} + \alpha_3\mathbf{Nn} + \alpha_4A + \alpha_5\mathbf{nnA} + \alpha_6\mathbf{Ann}, \quad (1)$$

where  $A = 1/2(\partial_i v_j + \partial_j v_i)$  is the symmetric part of the tensor of the gradients of the flow velocity of the NLC,  $\mathbf{N} = d\mathbf{n}/dt - (\mathbf{\Omega} \times \mathbf{n})$ , where  $\mathbf{\Omega} = 1/2(\partial_i v_j - \partial_j v_i)$ ,  $\mathbf{n}$  is the unit vector of the director in the NLC, and the  $\alpha_i$  ( $i = 1, \dots, 6$ ) are the coefficients of viscosity, known as the Leslie coefficients.<sup>1</sup>

The Leslie coefficients satisfy the Parodi relation<sup>1</sup>

$$\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5, \quad (2)$$

so that only five of the six coefficients  $\alpha_i$  are independent. In the isotropic phase all  $\alpha_i$ , except  $\alpha_4$ , equal zero, and  $\alpha_4$  is the shear viscosity of an isotropic liquid.

In the case of a liquid-crystal phase the coefficient that plays the role of the coefficient of rotational viscosity  $\gamma_1 = \alpha_3 - \alpha_2$  is of greatest interest, since it determines in the absence of hydrodynamic flow the dissipation of energy on account of only the rotation of the director  $\mathbf{n}$  under the action of external forces.

The first microscopic description of the rotational viscosity of NLCs (and all other Leslie coefficients) was given by Diogo and Martins<sup>3</sup> who advanced idea that the coefficient of rotational viscosity

$$\gamma_1 \sim S^2 \exp[S^2 E / (k(T - T_{NI}))] \quad (3)$$

is proportional to the characteristic relaxation time of the NLC, associated with the probability of overcoming a potential barrier in a molecular reorientation process. The magnitude of the barrier is determined both by the average orienting field acting on a molecule and by the parameters of the free volume of the NLC that is required for molecular reori-

entation. An essential point is that the activation energy associated with the average molecular field is proportional to the squared order parameter  $S$  and varies appreciably near the transition point  $T_{NI}$  into the isotropic phase. Here  $S = \langle P_2(\cos \theta_i) \rangle$  is the order parameter,  $\cos \theta_i = \mathbf{n} \cdot \mathbf{k}$ ,  $\mathbf{k}$  is a unit vector directed along the major axis of the  $i$ -th molecule,  $P_2$  is a second-order Legendre polynomial,<sup>1</sup> and the brackets denote a statistical average.

This approach has the considerable drawback that it is impossible to describe the difference between the Leslie coefficients, since they are all assumed to be determined by the same relaxation time. At the same time, it is known from experiments that Leslie coefficients differ considerably not only in absolute magnitude but also in sign.<sup>1</sup>

An interesting approach to the description of rotational viscosity of LCs of polymer solutions has been proposed by Doi and Kuzuu (DK).<sup>1</sup> However, this approach is limited primarily to polymer systems characterized by a high viscosity and small volume fraction of the molecules. Another drawback of this approach is the absence of an exponential temperature dependence, observed in experiments,<sup>2</sup> of the Leslie coefficients and the fact that the coefficient of rotational viscosity,

$$\gamma_1 \sim \frac{\rho k T S}{D_R \xi}, \quad (4)$$

can be calculated only once the rotational self-diffusion coefficient  $D_R$  and the parameter  $\xi = 1/\cos(2\varphi_0)$ , associated with the maximum angle  $\varphi_0$  of the orientations of the axes of the nematic order by the hydrodynamic flow, have been calculated. Here there arises an independent problem—the calculation of the rotational self-diffusion coefficient or “microscopic friction.”

Several years ago Brook–Levinson and Zakharov (BLZ)<sup>8</sup> proposed a simple molecular model for calculating the rotational self-diffusion coefficient of liquid-crystal systems. The model is based on the random-walk theory combined with some statistical-mechanical ideas. The result is an expression for the rotational self-diffusion coefficient

$$D_R = \pi^3 [kT/2\pi I]^{1/2} F\left(\frac{\pi}{2}\right) \quad (5)$$

presented in the form of a function of temperature  $T$ , the molecular moment of inertia  $I$ , and the value of the orientational distribution function  $F(\theta)$  at the point  $\theta = \pi/2$ , which corresponds to a molecular orientation perpendicular to the director. As is well known, in a liquid crystal  $F(\theta)$  possesses a fairly sharp maximum at  $\theta = 0$ , rapidly decreasing to the point  $\theta = \pi/2$ , where it has a small but finite value. The quantity  $F(\pi/2)$  determines the width of the “gates” through which molecules diffuse in orientational space. If  $F(\pi/2) = 0$ , then  $D_R = 0$ , i.e., there is no rotational diffusion, which corresponds to a completely ordered crystal. This *a priori* unknown factor  $F(\pi/2)$  can be taken, for example, from computational results obtained by molecular-dynamics<sup>10</sup> or from a statistical theory.<sup>9</sup> All this makes it possible to give the DK theory a logically complete form.

The next step in constructing a molecular theory of viscosity was taken by Osipov and Terent’ev (OT),<sup>5</sup> who estimated, on the basis of molecular-field theory using the Fokker–Planck equation, the coefficient of rotational viscosity as

$$\gamma_1 \sim \frac{\rho \lambda}{6} [J/kT]^{1/2} \exp[J/kT], \quad (6)$$

where  $\rho = N/V$  is the density of a system of  $N$  particles in a volume  $V$ , while  $J = J_0 S$ , where  $J_0$  is the height of the Maier–Saupe potential. But even in this theory the basic difficulty is to estimate the coefficient of “microscopic friction” of the molecules  $\lambda = kT/D_R$ . In the OT work an estimate of the coefficient  $\lambda$  was given in the form  $\lambda \sim \eta l^3$  on the basis of general dimensional considerations, where  $\eta$  is a certain initial coefficient of viscosity which is of the order of the isotropic viscosity  $\alpha_4$ , while  $l$  is the molecular length. Adding to this the fact that the values of the adjustable parameter  $J$  lie in the range  $4.0 < J/kT < 7.0$ , a reasonable choice of the other parameters in the OT theory gives order-of-magnitude agreement with experimental data for the coefficient of rotational viscosity of *p*-azoxyanisole.<sup>2</sup>

Another approach to describing the rotational viscosity in NLCs was developed by Nemtsov<sup>7</sup> on the basis of the NSO method.<sup>6</sup> In contrast to the investigations mentioned above, this approach takes account of not only the autocorrelation of the microscopic stress tensor but also the correlations of this quantity with the fluxes of the tensor order parameter and director. A calculation of these correlations shows the importance of the contribution of the interaction between the flow of the medium and the molecular orientation to the coefficient of viscosity. The nonequilibrium averaging of the microscopic stress tensor and the equations of motion for the order parameter and director [see Eqs. (30)–(36) in Ref. 11], performed on the basis of the NSO method, taking account of the fact that the temporal correlation functions determining the viscosity coefficients are calculated approximately using the Fokker–Planck equation [see Eqs. (37)–(40) in Ref. 11], made it possible to obtain an expression, of interest to us, for the rotational viscosity coefficient  $\gamma_1$  in the form

$$\gamma_1 = \frac{6\rho}{D_R} f(S), \quad (7)$$

where

$$f(S) = \frac{(3.181 + 0.757S)S^2}{2.881 + S12.36S^2 + 4.69S^3 - 0.743S^4}.$$

But even in this approach the main difficulty is to calculate the rotational self-diffusion coefficient  $D_R$ .

Several years ago a comparative analysis was made of the computational results for  $\gamma_1$ , obtained on the basis of the DM and OT models, and the experimental data for a number of LC compounds.<sup>12</sup> The results appear to be quite encouraging at first glance, but the large number of adjustable parameters employed in the calculations decreases somewhat the value of these approaches and makes it impossible to

calculate the rotational viscosity from first principles, based only on the form of the Hamiltonian of the system.

**2. INFLUENCE OF THE BOUNDING SURFACE ON THE ROTATIONAL VISCOSITY**

The main problem studied in the present paper is to investigate the effect of an interacting surface on the rotational viscosity on the basis of the already-developed statistical approach<sup>7</sup> using the tools of direct correlation functions.<sup>7,11</sup>

Preference is given to the Nemtsov approach<sup>7</sup> for a variety of reasons: In the first place, the correlations of the microscopic stress tensor with the fluxes of the tensor order parameter and the director are taken into account more completely and, in the second place, the contributions due to the interaction between the flow of the medium and the molecular orientations are taken into account. Using the BLZ theory<sup>8</sup> to describe rotational self-diffusion processes makes it possible to express the coefficient  $\gamma_1$  in a form convenient for direct calculations

$$\gamma_1 = \frac{6\rho}{\pi^3} [2\pi I/kT]^{1/2} \frac{1}{F(\pi/2)} f(S). \tag{8}$$

Another possible representation of  $\gamma_1$  is provided by the well-known expansion of the orientational distribution function in a series in Legendre polynomials

$$F(\theta) = \sum_l \frac{2l+1}{2} \langle P_l(\cos \theta) \rangle P_l(\cos \theta), \tag{9}$$

where  $l$  is an even number. Setting here  $\theta = \pi/2$  we obtain

$$F(\pi/2) = \sum_l \frac{2l+1}{2} \langle P_l(\cos \theta) \rangle P_l(\cos \pi/2). \tag{10}$$

Since most experimental methods permit finding only the first few coefficients in Eq. (10), limiting the series to the first few terms up to  $l=3$  we obtain

$$F(\pi/2) = \frac{1}{2} \left[ 1 - \frac{5}{2} \langle P_2 \rangle + \frac{27}{8} \langle P_4 \rangle - \frac{65}{16} \langle P_6 \rangle \right]. \tag{11}$$

Substituting the expression (11) into Eq. (8) we find that the coefficient of rotational viscosity can be expressed in terms of the temperature  $T$ , the molecular moment of inertia  $I$ , the order parameters  $\langle P_l \rangle$ , and the density  $\rho$  of the system.

We shall estimate  $D_R$  and  $\gamma_1$  for the nematic *p*-azoxyanisole. According to Ref. 9,  $F(\pi/2) \approx 0.05$ ; for  $T=390$  K,  $I=20 \times 10^{-44}$  kg·m<sup>2</sup>, and  $k=1.38 \times 10^{-23}$  J/K Eq. (5) gives the estimate  $D_R \approx 1.1 \times 10^{10}$  s<sup>-1</sup>, which is quite close to both the molecular-dynamics estimate<sup>13</sup> ( $D_R=0.4 \times 10^{10}$  s<sup>-1</sup>) and the experimental estimates<sup>14</sup> ( $D_R \approx 10^{10} - 10^{11}$  s<sup>-1</sup>), while Eq. (8) gives  $\gamma_1 \approx 0.0536$  ps. This value agrees quite well with the experimental estimate  $\gamma_1^{exp} \approx 0.067$  ps.<sup>15</sup>

In the case of a nematic phase bounded by a flat wall, the order parameters  $\langle P_2 \rangle$ ,  $\langle P_4 \rangle$ , and  $\langle P_6 \rangle$  are functions of the distance from the surface. The computational results show that, irrespective of the nature of the interacting surface, the effect of the wall extends only to several molecular layers and is determined mainly by intermolecular correlations.<sup>9</sup>

Using the order parameters  $\langle P_2 \rangle$ ,  $\langle P_4 \rangle$ , and  $\langle P_6 \rangle$ , calculated using the single-particle molecular-orientational-distribution function determined in Ref. 9, we shall calculate the coefficient of rotational viscosity  $\gamma_1$  as a function of the distance from the bounding surface on the basis of the Nemtsov, Brook–Levinson, and Zakharov approach according to Eq. (8) and expression (11). As already mentioned, the orientational distribution function  $F(\theta)$  appearing in expression (8) for the coefficient of rotational viscosity can be calculated either on the basis of an independent statistical theory or using molecular-dynamics.

One such approach to calculating the function  $F(\theta)$  on the basis of the method of conditional distributions<sup>16–18</sup> is presented in detail in Ref. 9. In this approach, the interaction potential of ellipsoidal molecules forming a nematic phase was chosen in the Gay–Berne form<sup>16</sup>

$$\Phi(\mathbf{q}_{ij}, \mathbf{e}_i, \mathbf{e}_j) = 4\epsilon_0 \epsilon(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij}) \{ (\sigma_0/r_{ij})^{12} - (\sigma_0/r_{ij})^6 \}, \tag{12}$$

where  $r_{ij} = |\mathbf{q}_{ij}| - \sigma(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij} + \sigma_0)$ . Here  $\mathbf{e}_i$  and  $\mathbf{e}_j$  are, respectively, unit vectors directed along the major axes of the ellipsoidal *a*-th and *j*-th molecules,  $\mathbf{q}_{ij}$  is a vector connecting the centers of the molecules, and  $\mathbf{e}_{ij} = \mathbf{q}_{ij}/|\mathbf{q}_{ij}|$ . The parameters of the potential energy  $\epsilon(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij})$  and the size  $\sigma(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij})$  are functions of the relative orientation of the molecules *i* and *j*. They are determined by Eqs. (3), (4), and (8)–(10) from Ref. 16, respectively. These functions also depend on the molecular shape, determined by the molecular ellipticity parameter  $\sigma_{\parallel}/\sigma_{\perp}$ , where  $\sigma_{\parallel}$  is the length of the major semiaxis of an ellipsoidal molecule and  $\sigma_{\perp}$  is the length of the minor semiaxis, and on the energy parameter  $\epsilon_l/\epsilon_s$  (in the notations of Ref. 16).

The presence of a solid wall presupposes that the molecules forming the nematic phase occupy the half-space  $x \geq 0$  (the coordinate system is chosen in a manner such that the *x* axis is directed along the normal to the surface, while the *z* axis is oriented so that the director lies in the *xz* plane). The wall also interacts with the molecules of the nematic through the potential

$$\Phi(\mathbf{e}_i, x_i) = \frac{2\pi}{3} \frac{\epsilon_w}{\sigma_w^2} \left[ \frac{2}{15} (\sigma_0/\sigma_w x_i)^9 - (\sigma_0/\sigma_w x_i)^3 \right]. \tag{13}$$

The energy  $\epsilon_w$  and size  $\sigma_w$  parameters depend on the orientation of the *i*-th molecule and are determined by the expression (20) and (21) of Ref. 9, respectively. The energy parameter  $\epsilon_w = \epsilon\epsilon_0$  determines the strength of the interaction of the wall with the molecules in the system;  $x_i$  is the distance between a molecule and the wall; and,  $\mathbf{e}_i = (e_{i,x}, e_{i,y}, e_{i,z})$ .

In what follows, we shall employ  $\epsilon_0$  as the unit of energy and  $\sigma_{\perp}$  as the unit of length. In this system of units the proposed theory contains several independent parameters: the dimensionless volume  $\nu^* = \nu/\sigma_{\perp}^3$  and the temperature  $\theta = kT/\epsilon_0$  as well as the parameters  $\epsilon_w$  and  $\sigma_{\parallel}/\sigma_{\perp}$ , reflecting the degree of interaction of the wall with the molecules in the system and the character of the ellipticity of the molecules themselves, respectively. The latter parameter was chosen as  $\sigma_{\parallel}/\sigma_{\perp} = 3$ .

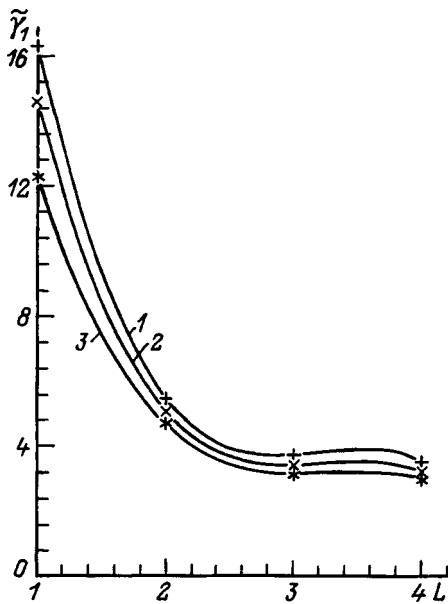


FIG. 1. Dimensionless coefficient of rotational viscosity  $\tilde{\gamma}_1$  versus the distance  $L$  from the wall with  $\nu^*=4.0$ ,  $\sigma_{\parallel}/\sigma_{\perp}=3.0$ ,  $\epsilon_w=5.0\epsilon_0$  and dimensionless temperatures  $\theta=0.75$  (1), 0.7885 (2), and 0.827 (3). The values of the parameters  $\mu$  and  $\nu$  (in the notations of Ref. 16) were chosen equal to 1 and 2, respectively.

The computational results obtained using the model potentials (12) and (13) showed that the dimensionless coefficient of rotational viscosity  $\tilde{\gamma}_1 = \gamma_1 \sigma_{\perp}^3 / (\epsilon_0 J)^{1/2}$  decreases as temperature  $\theta$  increases and depends strongly on the distance to the bounding surface  $L = x/\sigma_{\perp}$  (see Fig. 1). Ordinarily,  $\gamma_1$  decreases rapidly within the first two molecular volumes and rapidly reaches the bulk value  $\gamma_b$ . This behavior of the coefficient of rotational viscosity can be explained by the fact that molecules tend to be more strongly ordered near a strongly acting surface ( $\epsilon_w = 5.0\epsilon_0$ ) than in the interior of the nematic. This is confirmed by the results of calculations of the order parameter  $S(L) = \langle P_2^{(L)}(\cos \theta_i) \rangle$  as a function of the distance  $L$  to the bounding surface. The quantity  $S(L)$  also decreases rapidly within the first two molecular layers and rapidly reaches the bulk value (see Fig. 1 in Ref. 9). Alternative calculations by the molecular-dynamics methods, using the intermolecular potentials (12) and the interaction potentials (13) between the nematic molecules and a bounding surface, give good quantitative agreement (see Fig. 1 in Ref. 9). Since the order parameter  $S(L)$  has the highest absolute value among the order parameters, it determines the behavior of the orientational function  $F(\theta)$  and the coefficient of rotational viscosity. This explains the sharp decrease in  $\gamma_1$  as a function of distance from the bounding surface.

The main result of this work is that a new model description has been proposed for the rotational viscosity of a NLC

on the basis of a statistical-mechanics approach, combining the ideas of the nonequilibrium statistical-operator method<sup>6</sup> and the conditional-distributions method.<sup>18</sup> The coefficient of rotational viscosity  $\gamma_1$  is determined by the moment of inertia of the molecules comprising the nematic phase, the density and temperature of the phase, and also the order parameters, to calculate which it is necessary to know only the intermolecular interaction potentials. The theory contains no adjustable parameters and gives good quantitative agreement with both the experimental results for *p*-azoxyanisole and the results obtained by alternative methods, for example, molecular dynamics. The proposed model was used to investigate the effect of a bounding surface on the rotational viscosity of NLCs. The character of the influence of the surface is in many respects similar to the influence of a bounding surface on the shear viscosity of an isotropic liquid,<sup>19</sup> since this surface promotes the formation of oriented layers near the walls.

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