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Viscosity coefficients of partially aligned nematic and nematic discotic liquid crystals

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The affine transformation model—formulated for ellipsoidal particles with axis ratio Q and previously used for the calculation of the viscous properties of nematic liquid crystals with perfect orientation order—is generalized to realistic uniaxial phases with imperfect order (Maier-Saupe order parameter $S < 1$). For the nematic liquid crystals N-(4′-methoxybenzylidene)-4-(n-butyl) aniline (MBBA) and p-methoxy-p'-n-butylazoxybenzene (N4) and some cyanobiphenyls (kCB and kOCB with $k = 5, 8$) a comparison of the theoretically obtained Miesowicz viscosities with experimental data is made.

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INTRODUCTION

A good qualitative and reasonable quantitative understanding of the anisotropy of the viscosity and of the coefficients characterizing the flow alignment in nematic and nematic discotic liquid crystals is obtained by the affine transformation model [1–4]. By this approach, the transport coefficients of perfectly aligned particles with ellipsoidal equipotential surfaces are related to the transport coefficients of a reference fluid of spherical particles at the same density and one or two parameters which specify the shape of the uniaxial [1–4] or biaxial [5] ellipsoidal equipotential surfaces. Here, we restrict our attention to (effectively) uniaxial particles with the axis ratio $Q = a/b$ where a and $b = c$ are the semiaxes of the equipotential surfaces. The cases $Q > 1$ and $Q < 1$ pertain to prolate and oblate particles which form nematic and nematic discotic phases. The affine transformation model has been tested successfully in nonequilibrium molecular dynamics computer simulations [3,4,6,7] of fluids with perfectly oriented particles corresponding to a Maier-Saupe order parameter $S = 1$. All real nematic liquid crystals, however, are only partially aligned ($S < 1$). For a comparison of the theoretical results based on the affine transformation model for particles of axis ratio Q one expects that the behaviour of a nematic liquid crystal with $S < 1$ somehow corresponds to that of a model nematic liquid crystal with $S = 1$ and an effective axis ratio Q_{eff} which is smaller (larger) than Q for prolate (oblate) particles. Qualitatively, this conjecture made previously [3,4] is correct. A quantitative prescription for the relation between the anisotropy of the viscosity of perfectly and of partially aligned nematic liquid crystals is presented here in the spirit of a modified affine transformation model which has been tested for the anisotropy of the (self-)diffusion in fluids of partially oriented nematic liquid crystals [8]. This theory, which aims to extend a well established model from complete to partial alignment, is complementary to a mesoscopic approach to the calculation of the anisotropy of the viscosity based on an expansion with respect to powers of the alignment tensor as follows from an (approximate) solution of a Fokker-Planck equation [9–16].

I. PRESSURE TENSOR AND VISCOSITY COEFFICIENTS

A. Phenomenological ansatz

Before any properties of the viscosity of a nematic liquid are discussed, two equivalent (well known) versions of an ansatz for the friction pressure tensor are presented in order to specify the meaning of the various viscosity coefficients.

Let ρ and $\boldsymbol{v}$ be the mass density and the flow velocity of a fluid. With the help of a substantial time derivative $\frac{d}{dt} = \frac{\partial}{\partial t} + v_\lambda \nabla_\lambda$, the momentum balance can be written as

$$\rho \frac{d}{dt} v_\mu + \nabla_\nu P^{\text{eq}}_{\nu\mu} + \nabla_\nu p_{\nu\mu} = 0 \, , \tag{1}$$

where the total pressure tensor $P_{\nu\mu}$ already has been split into its equilibrium part $P^{\text{eq}}_{\nu\mu}$ and the friction pressure tensor $p_{\nu\mu}$. Here and in the following, greek subscripts refer to Cartesian components and the summation convention is used for them.

The viscosity coefficients occur in the (linear) relation between $p_{\nu\mu}$ and the gradient of the velocity $\nabla_\nu v_\mu$. This second rank tensor is decomposed into its isotropic, antisymmetric, and symmetric traceless parts:

$$\nabla_\nu v_\mu = \frac{1}{3} \nabla_\lambda v_\lambda \delta_{\mu\nu} + \varepsilon_{\nu\mu\lambda} \omega_\lambda + \gamma_{\nu\mu} \tag{2}$$

with

$$\omega_\lambda = \frac{1}{2} \varepsilon_{\lambda\alpha\beta} \nabla_\alpha v_\beta \, , \tag{3}$$

$$\gamma_{\mu\nu} = \overline{\nabla_\nu v_\mu} = \frac{1}{2} \left(\nabla_\nu v_\mu + \nabla_\mu v_\nu \right) - \frac{1}{3} \nabla_\lambda v_\lambda \delta_{\mu\nu} \, . \tag{4}$$

The symbol $\overline{\cdots}$ indicates the symmetric traceless part of a tensor. The quantities ω_λ and $\gamma_{\nu\mu}$ are referred to

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as the vorticity and the deformation rate (shear rate) tensor. For a nematic with the director field $\boldsymbol{n} = \boldsymbol{n}(t, \boldsymbol{r})$ and its corotational time derivative

$$N_\mu = \frac{d}{dt} n_\mu - \varepsilon_{\mu\nu\lambda}\omega_\nu n_\lambda \ , \tag{5}$$

the ansatz [17–19]

$$\begin{aligned} - p_{\nu\mu} = {} & \alpha_1 n_\nu n_\mu n_\lambda n_\kappa \gamma_{\lambda\kappa} + \alpha_2 n_\nu N_\mu + \alpha_3 n_\mu N_\nu \\ & + \alpha_4 \gamma_{\nu\mu} + \alpha_5 n_\nu n_\lambda \gamma_{\lambda\mu} + \alpha_6 n_\mu n_\lambda \gamma_{\lambda\nu} \\ & + \zeta_1 n_\lambda n_\kappa \gamma_{\lambda\kappa} \delta_{\mu\nu} + \zeta_2 n_\nu n_\mu \nabla_\lambda v_\lambda + \zeta_3 \nabla_\lambda v_\lambda \delta_{\mu\nu} \end{aligned} \tag{6}$$

is used. The Leslie coefficients $\alpha_1, \ldots, \alpha_6$ and the coefficients ζ_1, ζ_2, ζ_3 have the dimension of a viscosity. Decomposition of the pressure tensor in analogy to (2) yields [9]

$$\begin{aligned} \overline{p_{\nu\mu}} = {} & -2\eta\gamma_{\nu\mu} - 2\tilde{\eta}_1 \overline{\overline{n_\nu n_\lambda}\, \gamma_{\lambda\mu}} \\ & -2\tilde{\eta}_2 \overline{n_\nu N_\mu} - 2\tilde{\eta}_3 \overline{n_\nu n_\mu}\, n_\lambda n_\kappa \gamma_{\lambda\kappa} \\ & -\zeta_2 \overline{n_\nu n_\mu} \nabla_\lambda v_\lambda \ , \end{aligned} \tag{7}$$

$$(p_{\nu\mu})^a = \gamma_1 (n_\nu N_\mu)^a + \gamma_2 \left(\overline{n_\nu n_\lambda}\, \gamma_{\lambda\mu} \right)^a \ , \tag{8}$$

$$\tfrac{1}{3} p_{\lambda\lambda} = -\eta_V \nabla_\lambda v_\lambda - \kappa n_\lambda n_\kappa \gamma_{\lambda\kappa} \ . \tag{9}$$

In (8), $(\,)^a$ indicates the antisymmetric part of a second rank tensor, e.g., $(p_{\nu\mu})^a = \frac{1}{2}(p_{\nu\mu} - p_{\mu\nu})$. The viscosity coefficients used in Eqs. (7)–(9) are related to those of Eq. (6) by

$$\eta = \frac{1}{2}\left(\alpha_4 + \frac{1}{3}(\alpha_5 + \alpha_6)\right) \ , \tag{10a}$$

$$\tilde{\eta}_1 = \frac{1}{2}(\alpha_5 + \alpha_6) , \quad \tilde{\eta}_2 = \frac{1}{2}(\alpha_2 + \alpha_3) , \quad \tilde{\eta}_3 = \frac{1}{2}\alpha_1 \ , \tag{10b}$$

$$\gamma_1 = \alpha_3 - \alpha_2 , \quad \gamma_2 = \alpha_6 - \alpha_5 \ , \tag{10c}$$

$$\eta_V = \frac{1}{3}\zeta_2 + \zeta_3 , \quad \kappa = \zeta_1 + \frac{1}{3}(\alpha_1 + \alpha_5 + \alpha_6) \ . \tag{10d}$$

Due to the Onsager-Parodi relation

$$\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5 \ , \tag{11}$$

or equivalently

$$2\tilde{\eta}_2 = \gamma_2 \ , \tag{12}$$

and the relation $\zeta_2 = \kappa$, only seven of the nine viscosity coefficients are linearly independent. In an isotropic fluid all coefficients vanish except for the shear viscosity $\eta = \frac{1}{2}\alpha_4$ and the bulk viscosity $\eta_V = \zeta_3$. Positive entropy production requires $\eta > 0$, $\eta_V > 0$, $\gamma_1 > 0$ but $\tilde{\eta}_1$, $\tilde{\eta}_2$, $\tilde{\eta}_3$, and γ_2 may have either sign. One standard set of viscosity coefficients is the $\alpha_1, \ldots, \alpha_6$ of Eq. (6). The coefficients occurring in the ansatz (7)–(9) can be preferable in connection with theoretical considerations. In experiments, linear combinations of the "basic" viscosity coefficients are measured. Some examples are mentioned next.

B. Miesowicz viscosities

In a plane Couette or a plane Poiseuille flow with the velocity in the x direction and its gradient in the y direction, one has

$$\nabla_\nu v_\mu = \Gamma e^y_\nu e^x_\mu \ , \quad \omega_\mu = -\frac{1}{2}\Gamma e^z_\mu \tag{13}$$

with the shear rate

$$\Gamma = \frac{\partial v_x}{\partial y} \ . \tag{14}$$

$\boldsymbol{e}^{x,y,z}$ are unit vectors parallel to the x, y, z coordinate axes.

The Miesowicz viscosities η_i, $i = 1, 2, 3$ are defined by

$$p_{yx} = -\eta_i \Gamma \tag{15}$$

with the director $\boldsymbol{n}$ parallel to the x, y, and z axes, respectively; cf. Fig. 1. The orienting (magnetic) field has to be strong enough to overcome the flow induced orientation. A fourth coefficient η_4 analogous to (15) with $\boldsymbol{n}$ parallel to the bisector between the x and y axes is needed to characterize the shear viscosity. Instead of η_4, the Helfrich viscosity coefficient

$$\eta_{12} = 4\eta_4 - 2(\eta_1 + \eta_2) \tag{16}$$

is used in addition to the Miesowicz coefficients. Those four effective viscosities measurable in a flow experiment are related to the viscosity coefficients of Eq. (6) and Eqs. (7)–(9) by

$$\begin{aligned} \eta_1 &= \frac{1}{2}(\alpha_4 + \alpha_6 + \alpha_3) \\ &= \eta + \frac{1}{6}\tilde{\eta}_1 + \frac{1}{2}\tilde{\eta}_2 + \frac{1}{4}(\gamma_1 + \gamma_2) , \end{aligned} \tag{17a}$$

$$\begin{aligned} \eta_2 &= \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2) \\ &= \eta + \frac{1}{6}\tilde{\eta}_1 - \frac{1}{2}\tilde{\eta}_2 + \frac{1}{4}(\gamma_1 - \gamma_2) , \end{aligned} \tag{17b}$$

$$\eta_3 = \frac{1}{2}\alpha_4 = \eta - \frac{1}{3}\tilde{\eta}_1 , \tag{17c}$$

$$\eta_{12} = \alpha_1 = 2\tilde{\eta}_3 \ . \tag{17d}$$

The antisymmetric part of the pressure tensor contributes to η_1 and η_2 but not to η_3 and η_{12}. Notice that

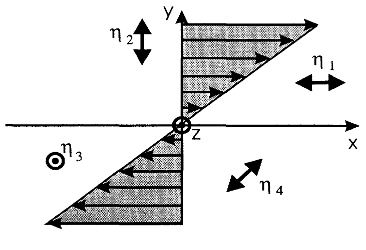


FIG. 1. Geometry of a plane Couette flow and definition of the Miesowicz viscosities.

$$\eta_1 + \eta_2 + \eta_3 = 3\eta, \tag{18a}$$

$$\eta_1 - \eta_2 = 2\tilde{\eta}_2 = \gamma_2, \tag{18b}$$

$$\eta_1 + \eta_2 - 2\eta_3 = \tilde{\eta}_1 + \tfrac{1}{2}\gamma_1 \ . \tag{18c}$$

The dependence of the normal pressure differences $p_{xx} - p_{yy}$ and $p_{zz} - \frac{1}{2}(p_{xx} + p_{yy})$ as well as of p_{yz} and p_{zy} on the various viscosity coefficients which follows from the ansatz (6) or from (7)–(9) is not needed here. However, some brief remarks on flow alignment are in order.

C. Flow alignment and viscosity for a free flow

If no external orienting field is applied, the director $\boldsymbol{n}$ in the flowing nematic liquid crystal orients such that the antisymmetric part of the pressure tensor associated with the torque acting on the fluid vanishes. This leads to [17–19]

$$n_\mu = \cos(\phi_0)e^x_\mu + \sin(\phi_0)e^y_\mu \ , \qquad \cos 2\phi_0 = -\gamma_1/\gamma_2 \ , \tag{19}$$

provided that $|\gamma_2| > \gamma_1$. The angle ϕ_0 is referred to as the flow alignment angle.

The viscosity of a freely flowing nematic liquid crystal with the director n_μ determined by the flow alignment is

$$\begin{aligned}\eta_S &= \frac{1}{2}(\eta_1 + \eta_2 - \gamma_1) + \frac{1}{4}\eta_{12}\left(1 - (\gamma_1/\gamma_2)^2\right) \\ &= \eta + \frac{1}{6}\tilde{\eta}_1 - \frac{1}{4}\gamma_1 + \frac{1}{2}\tilde{\eta}_3\left(1 - (\gamma_1/\gamma_2)^2\right) \ . \end{aligned} \tag{20}$$

It should be mentioned that the antisymmetric part of the pressure tensor also vanishes for $n_\mu = e^z_\mu$ where the effective viscosity equals η_3. This second type of flow orientation is not stable, in general.

II. FROM PERFECT TO PARTIAL ALIGNMENT

Let $\boldsymbol{u}$ be a unit vector parallel to the figure axis of a molecule. The second rank alignment tensor can be defined by $\langle \overline{u_\mu u_\nu} \rangle = \langle u_\mu u_\nu \rangle - \frac{1}{3}\delta_{\mu\nu}$. In a uniaxial state one has

$$\langle \overline{u_\mu u_\nu} \rangle = S_2 \overline{n_\mu n_\nu}, \quad S_2 = \langle P_2(\boldsymbol{u}\cdot\boldsymbol{n})\rangle \ , \tag{21}$$

where $S_2 = S$ is the usual Maier-Saupe order parameter and $P_\ell(\boldsymbol{u}\cdot\boldsymbol{n})$ is the ℓth Legendre polynomial.

Suppose that the viscosity coefficients occurring in (6) or in (7)–(9) are known for a fluid of perfectly aligned or "ordered" particles where $\boldsymbol{n} = \boldsymbol{u}$ and the coefficients are distinguished by the superscript "ord." Then Eq. (6) reads

$$\begin{aligned} -p^{\mathrm{ord}}_{\nu\mu} = {}& \alpha_1^{\mathrm{ord}} u_\nu u_\mu u_\lambda u_\kappa \gamma_{\lambda\kappa} + \alpha_2^{\mathrm{ord}} u_\nu U_\mu \\ & + \alpha_3^{\mathrm{ord}} u_\mu U_\nu + \alpha_4^{\mathrm{ord}} \gamma_{\nu\mu} + \alpha_5^{\mathrm{ord}} u_\nu u_\lambda \gamma_{\lambda\mu} \\ & + \alpha_6^{\mathrm{ord}} u_\mu u_\lambda \gamma_{\lambda\mu} + \zeta_1^{\mathrm{ord}} u_\lambda u_\kappa \gamma_{\lambda\kappa}\delta_{\mu\nu} \\ & + \zeta_2^{\mathrm{ord}} u_\nu u_\mu \nabla_\lambda v_\lambda + \zeta_3^{\mathrm{ord}} \nabla_\lambda v_\lambda \delta_{\mu\nu} \end{aligned} \tag{22}$$

with $U_\mu = \dot{u}_\mu - \varepsilon_{\mu\lambda\kappa}\omega_\lambda u_\kappa$.

Of course, equations analogous to (7)–(9) can be written down for the perfectly oriented fluid. In the spirit of the considerations of Ref. [8] which worked quite successfully in the case of the anisotropy of the diffusion the pressure tensor of a partially aligned fluid is now obtained from (22) by averaging $p^{\mathrm{ord}}_{\nu\mu}$ over orientations, viz., $p_{\nu\mu} = \langle p^{\mathrm{ord}}_{\nu\mu}\rangle$. This conjecture is based on the assumption that the molecular orientation is practically perfect in small volume elements and that $S_2 < 1$ is caused by orientational changes over distances which do not affect the viscous properties as expressed by the coefficients occurring in (22). The averaging is straightforward for terms like

$$\begin{aligned}\langle u_\mu u_\nu \rangle &= \langle \overline{u_\mu u_\nu} \rangle + \frac{1}{3}\delta_{\mu\nu} \\ &= S_2 \overline{n_\nu n_\mu} + \frac{1}{3}\delta_{\mu\nu} \\ &= S_2 n_\nu n_\mu + \frac{1}{3}\delta_{\mu\nu}(1 - S_2) \ . \end{aligned} \tag{23}$$

Here we have assumed uniaxial symmetry of the orientational distribution. The average of the first term of (22) is rewritten as $\frac{1}{3}\delta_{\mu\nu}\langle u_\lambda u_\kappa\rangle\gamma_{\lambda\kappa} + \langle \overline{u_\mu u_\nu}\ \overline{u_\lambda u_\kappa}\rangle\gamma_{\lambda\kappa}$. Use of

$$\begin{aligned}\overline{u_\mu u_\nu}\ \overline{u_\lambda u_\kappa}\gamma_{\lambda\kappa} = {}& \overline{u_\mu u_\nu u_\lambda u_\kappa}\gamma_{\lambda\kappa} \\ & + \frac{4}{7}\overline{u_\mu u_\lambda}\gamma_{\lambda\nu} + \frac{2}{15}\gamma_{\mu\nu} \ , \end{aligned} \tag{24}$$

and of

$$\langle \overline{u_\mu u_\nu u_\lambda u_\kappa} \rangle = S_4 \overline{n_\mu n_\nu n_\lambda n_\kappa} \ , \qquad S_4 = \langle P_4(\boldsymbol{n}\cdot\boldsymbol{u})\rangle \ , \tag{25}$$

where S_4 is an order parameter analogous to S_2, leads to

$$\begin{aligned}\langle u_\mu u_\nu u_\lambda u_\kappa\rangle\gamma_{\lambda\kappa} = {}& S_4 n_\mu n_\nu n_\lambda n_\kappa \gamma_{\lambda\kappa} \\ & + \frac{2}{7}(S_2 - S_4)(n_\mu n_\lambda \gamma_{\lambda\nu} + n_\nu n_\lambda \gamma_{\lambda\mu}) \\ & + \frac{2}{15}\left(1 - \frac{10}{7}S_2 + \frac{3}{7}S_4\right)\gamma_{\mu\nu} \\ & + \frac{1}{7}(S_2 - S_4)\delta_{\mu\nu} n_\lambda n_\kappa \gamma_{\lambda\kappa} \ . \end{aligned} \tag{26}$$

Notice that $S_2 = 1$ and $S_4 = 1$ for perfect alignment.

The remaining average $\langle u_\mu U_\nu\rangle$ contains the competing influences of the internal dynamics—represented by $\langle u_\mu \dot{u}_\nu\rangle$—and of the macroscopic vorticity ω_λ. In the uniaxial case the evaluation of these mean values leads to

$$\begin{aligned}\langle u_\mu U_\nu\rangle = {}& \langle u_\mu \dot{u}_\nu\rangle - S_2 n_\mu \varepsilon_{\nu\lambda\tau}\omega_\lambda n_\tau \\ & + \frac{1}{3}\varepsilon_{\nu\mu\lambda}\omega_\lambda(1 - S_2) \ . \end{aligned} \tag{27}$$

In many relevant cases the (macroscopic) director n_ν is stationary. Thus we will assume that $\dot{n}_\nu = 0$ is sufficiently well realized in the experimental setup—a situation which includes the measurement of Miesowicz viscosities as well as the phenomenon of flow alignment. Then the corotational derivative N_ν of n_ν reduces to $N_\nu = -\varepsilon_{\nu\lambda\tau}\omega_\lambda n_\tau$ and Eq. (27) becomes

$$\langle u_\mu U_\nu\rangle = \langle u_\mu \dot{u}_\nu\rangle + S_2 n_\mu N_\nu + \frac{1}{3}\varepsilon_{\nu\mu\lambda}\omega_\lambda (1 - S_2) \ . \qquad (28)$$

In general, the vorticity ω_ν has also a component parallel to n_ν which may not be neglected as was presupposed for a single needle-shaped molecule. Decomposition of ω_ν into contributions parallel and perpendicular to n_ν is expressed by

$$P_{\lambda\tau} = \frac{1}{3}\left[(S_2 + 2)(\delta_{\lambda\tau} - n_\lambda n_\tau) + (1 - S_2) n_\lambda n_\tau\right] \qquad (29)$$

and we finally obtain

$$\langle u_\mu U_\nu\rangle = \langle u_\mu \dot{u}_\nu\rangle + S_2\, \overline{n_\mu N_\nu} - \varepsilon_{\nu\mu\lambda} P_{\lambda\tau}\frac{1}{2}\omega_\tau \ . \qquad (30)$$

Comparison of $\langle -p^{\rm ord}_{\nu\mu}\rangle$ with (6) yields

$$\alpha_1 = S_4 \alpha_1^{\rm ord} \ , \qquad (31a)$$

$$\alpha_4 = \alpha_4^{\rm ord} + \frac{2}{15}\left(1 - \frac{10}{7}S_2 + \frac{3}{7}S_4\right)\alpha_1^{\rm ord} + \frac{1}{3}(1 - S_2)\left(\alpha_5^{\rm ord} + \alpha_6^{\rm ord}\right) \ , \qquad (31b)$$

$$\alpha_5 = S_2\alpha_5^{\rm ord} + \frac{2}{7}(S_2 - S_4)\,\alpha_1^{\rm ord} \ , \qquad (31c)$$

$$\alpha_6 = S_2\alpha_6^{\rm ord} + \frac{2}{7}(S_2 - S_4)\,\alpha_1^{\rm ord} \ , \qquad (31d)$$

$$\zeta_1 = S_2\zeta_1^{\rm ord} + \frac{1}{7}(S_2 - S_4)\,\alpha_1^{\rm ord} \ , \qquad (31e)$$

$$\zeta_2 = S_2\zeta_2^{\rm ord}, \ \zeta_3 = \zeta_3^{\rm ord} + \frac{1}{3}(1 - S_2)\,\zeta_2^{\rm ord} \ . \qquad (31f)$$

Nevertheless, there is no exact correspondence of $\langle -p^{\rm ord}_{\nu\mu}\rangle$ and $-p^{\rm ord}_{\nu\mu}$ with respect to terms involving $\alpha_2^{\rm ord}$ and $\alpha_3^{\rm ord}$, as can easily be seen from (30). Even if $\langle u_\nu \dot{u}_\mu\rangle$ is canceled—which may be justified for the flow experiment determining the Miesowicz viscosities (the experiment is performed in a way to prevent the rotation of the director and thus it is a reasonable assumption that the motion $\dot{u}_\nu$ of a singular molecular axis is strongly hindered as well) but which is clearly not fulfilled for a free flow—there remains the contribution due to the part of ω_ν parallel to the director n_ν.

For the sake of simplicity we assume $\langle u_\nu \dot{u}_\mu\rangle \equiv 0$ and focus our attention on flow experiments with this special constraint. Then (30) is just the decomposition of $\langle u_\nu U_\mu\rangle$ into its symmetric and skew symmetric parts. The crucial term $\langle \alpha_2^{\rm ord} u_\nu U_\mu + \alpha_3^{\rm ord} u_\mu U_\nu\rangle$ can now be rewritten as

$$\langle \alpha_2^{\rm ord} u_\nu U_\mu + \alpha_3^{\rm ord} u_\mu U_\nu\rangle = S_2\left(\alpha_2^{\rm ord} + \alpha_3^{\rm ord}\right)\overline{n_\nu N_\mu} - \varepsilon_{\nu\mu\lambda}\gamma^{(1)}_{\lambda\tau}\frac{1}{2}\omega_\tau \ . \qquad (32)$$

We have introduced the new tensor of rotational viscosity $\gamma^{(1)}_{\lambda\tau}$,

$$\gamma^{(1)}_{\lambda\tau} = \frac{1}{3}\left(\alpha_3^{\rm ord} - \alpha_2^{\rm ord}\right)\left[(2 + S_2)\left(\delta_{\lambda\tau} - n_\lambda n_\tau\right) + 2\left(1 - S_2\right) n_\lambda n_\tau\right] \ . \qquad (33a)$$

This tensor replaces the rotational viscosity coefficient γ_1 introduced in (8). For perfect alignment $\gamma^{(1)}_{\lambda\tau}$ reduces to $(\alpha_3^{\rm ord} - \alpha_2^{\rm ord})(\delta_{\lambda\tau} - n_\lambda n_\tau)$ which represents $\gamma_1^{\rm ord} = \alpha_3^{\rm ord} - \alpha_2^{\rm ord}$ together with the symmetry of one molecule. The effective rotational viscosity γ_1 that shows up in a given flow geometry depends on the angle between director n_ν and vorticity ω_ν. The scalar coefficient γ_1 is calculated from the tensor in Eq. (33) according to

$$\gamma_1 = \gamma^{(1)}_{\lambda\tau}\omega_\lambda\omega_\tau/(\omega_\kappa\omega_\kappa) \ . \qquad (33b)$$

Similarly, for the other coefficients occurring in (7)–(9) one obtains by the analogous procedure

$$\eta = \eta^{\rm ord} + \frac{2}{15}(1 - S_4)\,\tilde{\eta}_3^{\rm ord} \ , \qquad (34a)$$

$$\tilde{\eta}_1 = S_2\tilde{\eta}_1^{\rm ord} + \frac{4}{7}(S_2 - S_4)\,\tilde{\eta}_3^{\rm ord} \ , \qquad (34b)$$

$$\tilde{\eta}_2 = S_2\tilde{\eta}_2^{\rm ord} \ , \qquad (34c)$$

$$\tilde{\eta}_3 = S_4\tilde{\eta}_3^{\rm ord} \ , \qquad (34d)$$

$$\gamma_2 = S_2\gamma_2^{\rm ord} \ , \qquad (34e)$$

$$\eta_V = \eta_V^{\rm ord} \ , \qquad (34f)$$

$$\kappa = S_2\kappa^{\rm ord} \ . \qquad (34g)$$

For the Miesowicz viscosities and for η_{12} one now infers from (33), (34), and the definition in Eq. (15) that

$$\eta_1 = \eta^{\rm ord} + \frac{1}{6}S_2\tilde{\eta}_1^{\rm ord} + \frac{1}{2}S_2\tilde{\eta}_2^{\rm ord} + \frac{2}{15}\left(1 + \frac{5}{7}S_2 - \frac{12}{7}S_4\right)\tilde{\eta}_3^{\rm ord} + \frac{1}{12}(2 + S_2)\,\gamma_1^{\rm ord} + \frac{1}{4}S_2\gamma_2^{\rm ord} \ , \qquad (35)$$

$$\eta_2 = \eta^{\rm ord} + \frac{1}{6}S_2\tilde{\eta}_1^{\rm ord} - \frac{1}{2}S_2\tilde{\eta}_2^{\rm ord} + \frac{2}{15}\left(1 + \frac{5}{7}S_2 - \frac{12}{7}S_4\right)\tilde{\eta}_3^{\rm ord} + \frac{1}{12}(2 + S_2)\,\gamma_1^{\rm ord} - \frac{1}{4}S_2\gamma_2^{\rm ord} \ , \qquad (36)$$

$$\eta_3 = \eta^{\rm ord} - \frac{1}{3}S_2\tilde{\eta}_1^{\rm ord} + \frac{2}{15}\left(1 - \frac{10}{7}S_2 + \frac{3}{7}S_4\right)\tilde{\eta}_3^{\rm ord} + \frac{1}{6}(1 - S_2)\,\gamma_1^{\rm ord} \ , \qquad (37)$$

$$\eta_{12} = S_4\eta_{12}^{\rm ord} \ . \qquad (38)$$

For perfect alignment with $S_2 = S_4 = 1$ the relations given in (35)–(38) reduce to Eqs. (17). However, as a consequence of the dependence of γ_1 on the director alignment with respect to vorticity the general expression for η_3 in (37) cannot simply be derived from Eq. (17c): The term involving $\gamma_1^{\rm ord}$ would have been missed, causing the paradox that in an isotropic state there would be a difference between $\eta_1 = \eta_2$ and η_3—which clearly is not possible.

With the help of Eqs. (31), the viscosity coefficients of a partially aligned nematic liquid crystal are expressed in terms of the viscosities of a perfectly aligned fluid and by the order parameters S_2 and S_4. The relations presented so far apply to both ordinary nematic and nematic discotic substances. Differences between these two classes of nematic liquid crystals appear in the specific expressions for coefficients of the perfectly oriented fluid to be discussed next.

III. MODIFIED AFFINE TRANSFORMATION MODEL

The pressure tensor and consequently also the viscosity coefficients of a fluid contain "kinetic" and "potential" contributions [3,4,6,7]. The latter dominate in the viscosity coefficients of liquids. If the small kinetic contributions are disregarded, the viscosity coefficients of a perfectly aligned fluid of ellipsoidal particles as given by the affine transformation model are

$$\alpha_1^{\mathrm{ord}} = -\frac{1}{2}\left(Q - Q^{-1}\right)^2 \alpha_4^{\mathrm{ref}} , \tag{39a}$$

$$\alpha_2^{\mathrm{ord}} = \frac{1}{2}\left(1 - Q^2\right) \alpha_4^{\mathrm{ref}} , \tag{39b}$$

$$\alpha_3^{\mathrm{ord}} = \frac{1}{2}\left(Q^{-2} - 1\right)^2 \alpha_4^{\mathrm{ref}} , \tag{39c}$$

$$\alpha_4^{\mathrm{ord}} = \alpha_4^{\mathrm{ref}} = 2\eta^{\mathrm{ref}}, \tag{39d}$$

$$\alpha_5^{\mathrm{ord}} = -\alpha_2^{\mathrm{ord}} , \quad \alpha_6^{\mathrm{ord}} = \alpha_3^{\mathrm{ord}} , \tag{39e}$$

where $Q = a/b$ is the axis ratio of the (effectively) uniaxial particles (the semiaxes of their equipotential surfaces are a and $b = c$) and $\eta^{\mathrm{ref}} = \frac{1}{2}\alpha_4^{\mathrm{ref}}$ is the shear viscosity of a reference fluid of spherical particles at the same density and temperature. Furthermore, one has $\eta_V^{\mathrm{ord}} = \eta_V^{\mathrm{ref}}$ for the bulk viscosity and $\kappa^{\mathrm{ord}} = \zeta_2^{\mathrm{ord}} = 0$. The viscosity coefficients of Eqs. (7)–(9) are given by

$$\eta^{\mathrm{ord}} = \left(1 + \frac{1}{6}\left(Q - Q^{-1}\right)^2\right) \eta^{\mathrm{ref}} , \tag{40a}$$

$$\tilde{\eta}_1^{\mathrm{ord}} = \frac{1}{2}\left(Q - Q^{-1}\right)^2 \eta^{\mathrm{ref}} , \tag{40b}$$

$$\tilde{\eta}_2^{\mathrm{ord}} = \frac{1}{2}\left(Q^{-2} - Q^2\right) \eta^{\mathrm{ref}} , \tag{40c}$$

$$\tilde{\eta}_3^{\mathrm{ord}} = -\tilde{\eta}_1^{\mathrm{ord}} , \tag{40d}$$

$$\gamma_1^{\mathrm{ord}} = \left(Q - Q^{-1}\right)^2 \eta^{\mathrm{ref}} = 2\tilde{\eta}_1^{\mathrm{ord}} , \tag{40e}$$

$$\gamma_2^{\mathrm{ord}} = \left(Q^{-2} - Q^2\right) \eta^{\mathrm{ref}} = 2\tilde{\eta}_2^{\mathrm{ord}} . \tag{40f}$$

Similarly, for the Miesowicz coefficients and for η_{12} and η_S one has

$$\eta_1^{\mathrm{ord}} = Q^{-2}\eta^{\mathrm{ref}} , \quad \eta_2^{\mathrm{ord}} = Q^2\eta^{\mathrm{ref}} , \quad \eta_3^{\mathrm{ord}} = \eta^{\mathrm{ref}} ,$$
$$\eta_{12}^{\mathrm{ord}} = -\gamma_1^{\mathrm{ord}} , \quad \eta_S^{\mathrm{ord}} = 4\left(Q + Q^{-1}\right)^{-2} \eta^{\mathrm{ref}} . \tag{41}$$

Due to (40) and (35)–(37), the results obtained by the modified affine transformation model for the Miesowicz viscosities are

$$\eta_1/\eta^{\mathrm{ref}} = 1 + S_2\left(Q^{-2} - 1\right) + \frac{4}{15}\left(1 - \frac{10}{7}S_2 + \frac{3}{7}S_4\right)\left(Q - Q^{-1}\right)^2 , \tag{42}$$

$$\eta_2/\eta^{\mathrm{ref}} = 1 + S_2\left(Q^2 - 1\right) + \frac{4}{15}\left(1 - \frac{10}{7}S_2 + \frac{3}{7}S_4\right)\left(Q - Q^{-1}\right)^2 , \tag{43}$$

$$\eta_3/\eta^{\mathrm{ref}} = 1 - \frac{1}{15}\left(\frac{3}{7}S_4 + \frac{25}{7}S_2 - 4\right)\left(Q - Q^{-1}\right)^2 . \tag{44}$$

Analogously, one finds

$$\eta_{12}/\eta^{\mathrm{ref}} = -S_4\left(Q - Q^{-1}\right)^2 \tag{45}$$

and

$$\gamma_2/\eta^{\mathrm{ref}} = S_2\left(Q^{-2} - Q^2\right) . \tag{46}$$

Notice that η_3 and η_{12} are invariant under the replacement of Q by Q^{-1}, γ_2 changes sign, and η_1, η_2 exchange their roles, i.e.,

$$\gamma_2(Q) = -\gamma_2(Q^{-1}) , \quad \eta_1(Q) = \eta_2(Q^{-1}) . \tag{47}$$

These symmetry properties relating viscosity coefficients of nematic and nematic discotic substances are a consequence of the affine transformation model.

For $S_2 = S_4 = 1$, the results of the original affine transformation model are recovered from (42)–(46). In the other limiting case of an isotropic state with $S_2 = S_4 = 0$, one finds

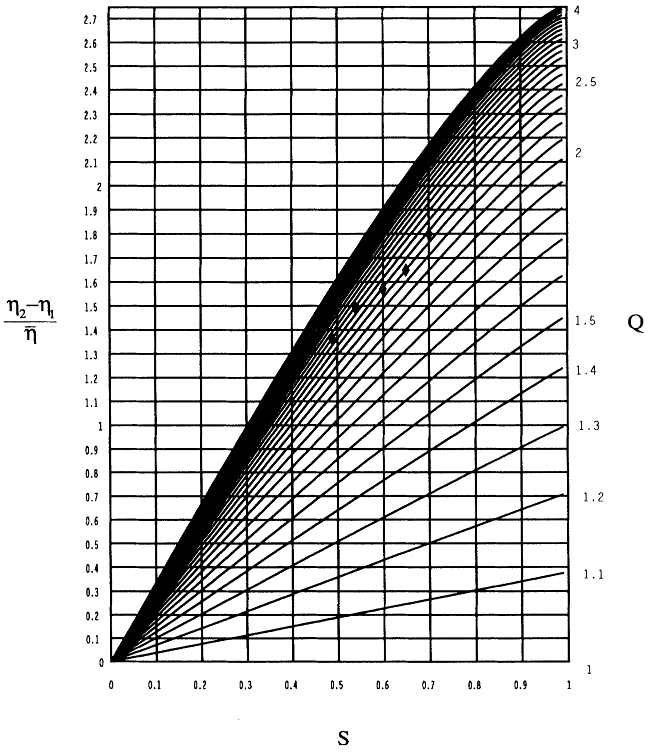


FIG. 2. Determination of the axis ratio Q of N4 (experimental values (taken from [28]) are marked with a filled ◊).

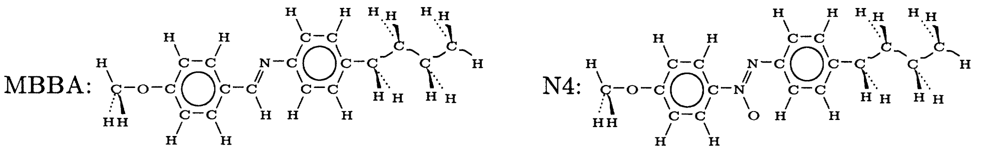


FIG. 3. Structural formulas of MBBA and N4 (isomeric mixture).

$$\eta_1 = \eta_2 = \eta_3 = \eta_S = \eta_0 \,,$$
$$\eta_0 = \left(1 + \frac{4}{15}\left(Q - Q^{-1}\right)^2\right)\eta^{\text{ref}} \,, \tag{48}$$
$$\eta_{12} = \gamma_2 = 0 \,.$$

In order to analyze the dependence of the viscosity coefficients of Eqs. (31) with (40) and (41) and of Eqs. (42)–(46) on the Maier-Saupe order parameter $S = S_2$, a specific form of the functional dependence of S_4 on S_2 is needed. As a reasonable guess which guarantees $S_4 = 0$ and $S_4 = 1$ for $S_2 = 0$ and $S_2 = 1$, respectively, the ansatz

$$S_4 = S_2(1 - (1 - S_2)^\nu) \,, \quad 0 < \nu \leq 1 \tag{49}$$

is made. The assumption that the orientational distribution is proportional to $\exp(\cdots P_2)$, where the Legendre polynomial depends on the angle between the figure axis of a molecule and the director, leads to $\nu \approx 0.6$. Such a distribution, e.g., underlies the Maier-Saupe theory, is motivated by entropy arguments discussed in the Appendix, and is supported by the results of computer simulations [20]. As will turn out in the following analysis, the viscosity coefficients, except for η_{12}, are not very sensitive to the choice of the parameter ν.

IV. COMPARISON WITH EXPERIMENTS

From (42)–(44) we have

$$\bar{\eta} := \frac{1}{3}\left(\eta_1 + \eta_2 + \eta_3\right)$$
$$= \left(1 + \frac{1}{15}\left(4 + S_4\right)\left(Q - Q^{-1}\right)^2\right)\eta^{\text{ref}} \tag{50}$$

and

$$\frac{\eta_2 - \eta_1}{\bar{\eta}} = \frac{S_2\left(Q^2 - Q^{-2}\right)}{1 + \frac{1}{15}\left(4 + S_4\right)\left(Q - Q^{-1}\right)^2} \,. \tag{51}$$

Thus we can derive the effective axis ratio Q of a liquid crystal from its Miesowicz viscosities. With the help of the nomogram in Fig. 2 the axis ratio of p-methoxy-p'-n-butylazoxybenzene (N4), a nematic mixture, is found to be $Q \approx 2.5$. Taking this Q value, the agreement of the theoretical predictions of the modified transformation model with the measured viscosities is quite remarkable. For the dependence of S_2 on temperature we used Haller's equation

$$S_2 = \left(1 - \frac{T}{T^*}\right)^\beta \tag{52a}$$

with the experimentally determined values [21]

$$T^*_{\text{MBBA}} = 319.2 \,, \quad \beta_{\text{MBBA}} = 0.188 \,, \tag{52b}$$
$$T^*_{\text{N4}} = 348.5 \,, \quad \beta_{\text{N4}} = 0.171 \,. \tag{52c}$$

Notice, that the axes ratio Q is—except for the choice of the parameter $\nu = 0.6$ which is not very sensitive—the only fit parameter of the model. Indeed, an ellipsoid with this particular axis ratio resembles the shape of a molecule of N-(4′-methoxybenzylidene)-4-(n butyl) aniline (MBBA) or N4 quite well (see Fig. 3, and compare the space filling model of MBBA in Fig. 4). The shapes of the molecules of MBBA and the main component of N4 (a nematic mixture) are very similar. The Miesowicz viscosities (see Fig. 5) are mostly influenced by the Maier-Saupe order parameter $S = S_2$; the dependence on the order parameter S_4 is comparatively weak. Thus it is instructive to look at a quantity like η_{12} which—according to Eq. (45)—depends on S_4 alone. The agreement of the experimental data with the theoretical prediction is not

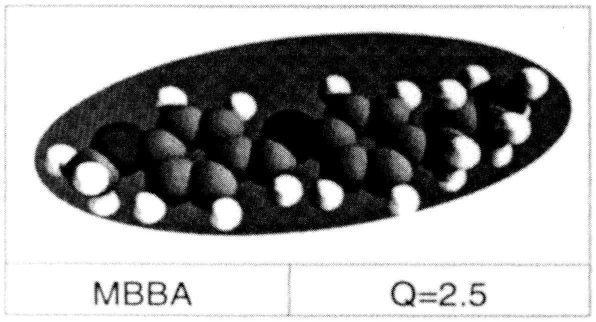


FIG. 4. Space filling model of MBBA and enclosing ellipsoid with axis ratio 2.5.

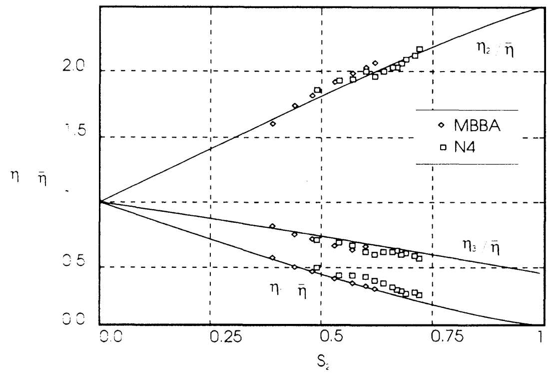


FIG. 5. Miesowicz viscosities of MBBA and N4 (data from [29,28] using Haller's equation [21]).

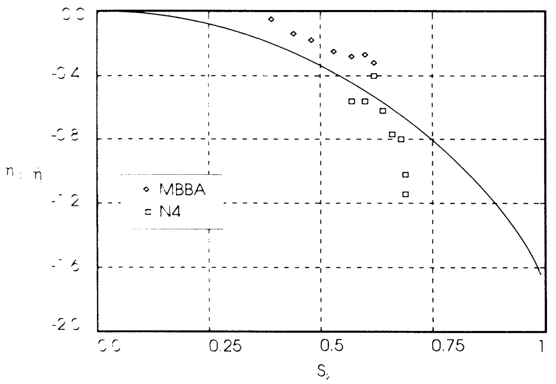


FIG. 6. Comparison of η_{12} with experimental values [29,28]. The order parameter S_2 is derived from temperature by the help of Haller's equation [21].

as good as it was for the Miesowicz viscosities but taking into account the variance of the data the agreement seems to be acceptable (see Fig. 6).

Another class of nematic liquid crystals where measurements of all Miesowicz viscosities [22] are available and a relation between the order parameter S_2 and temperature T has been experimentally obtained by using NMR techniques [23] is the cyanobiphenyls [kCB and kOCB with k denoting the length of the alkyl (oxy) chain]. The functional relation used here is given by

$$S_2(T) = S_0\,(1 - T/T_C)^F \tag{53a}$$

with

$$S_0^{k\mathrm{CB}} = 1.2267\ , \quad F^{k\mathrm{CB}} = 0.2418\ , \tag{53b}$$

$$S_0^{k\mathrm{OCB}} = 0.9161\ , \quad F^{k\mathrm{OCB}} = 0.1392\ . \tag{53c}$$

The nomogram for obtaining axis ratios is shown in Fig. 7; the comparison of theoretical and experimental values of relative viscosities is made in Fig. 8. It can be seen that the effective axis ratio Q is increasing in the sequence 5OCB–8CB–8OCB–5CB. The difference in molecular shape is due to the alkyl (oxy) group (cf. Fig. 9) indicating that an even number of kinks (5OCB, 8CB) in that group leads to a more spherical molecule than a chain with an odd number of kinks (8OCB, 5CB). In addition, the greater flexibility of long chains tends to "smooth out" the difference between the kCB's and kOCB's whereas short chains have a more distinct influence on the form of the molecule.

Because an even number of kinks gives the aliphatic chain the tendency to protrude in a direction perpendicular to the plane of the biphenyl core the diameter of the molecule is increased at the cost of its length. Contrarily, an odd number of kinks bends the chain back to the core plane producing more elongated molecules. However, increasing mobility of the chain will flatten out this effect. Thus the observed odd-even effect as well as its

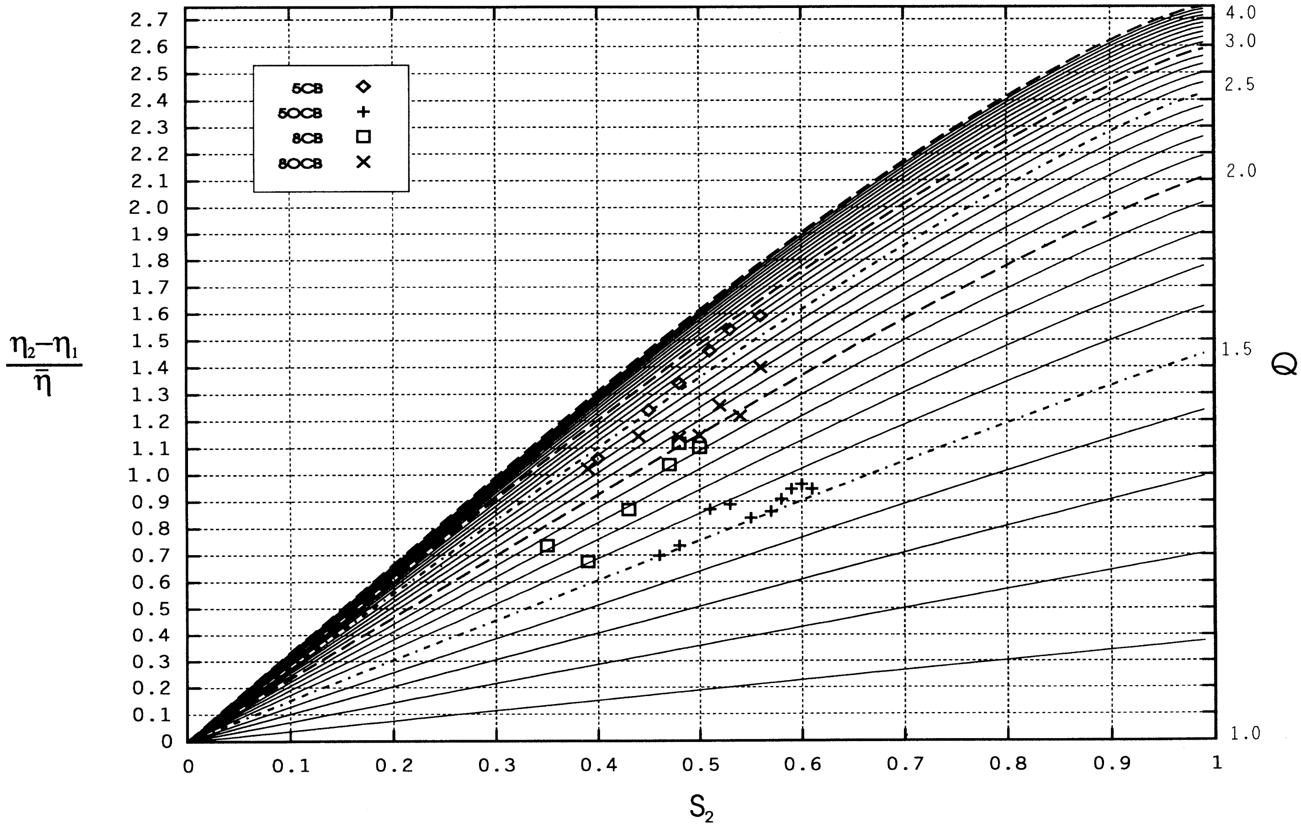


FIG. 7. Determination of axis ratios of kCB and kOCB ($k = 5, 8$).

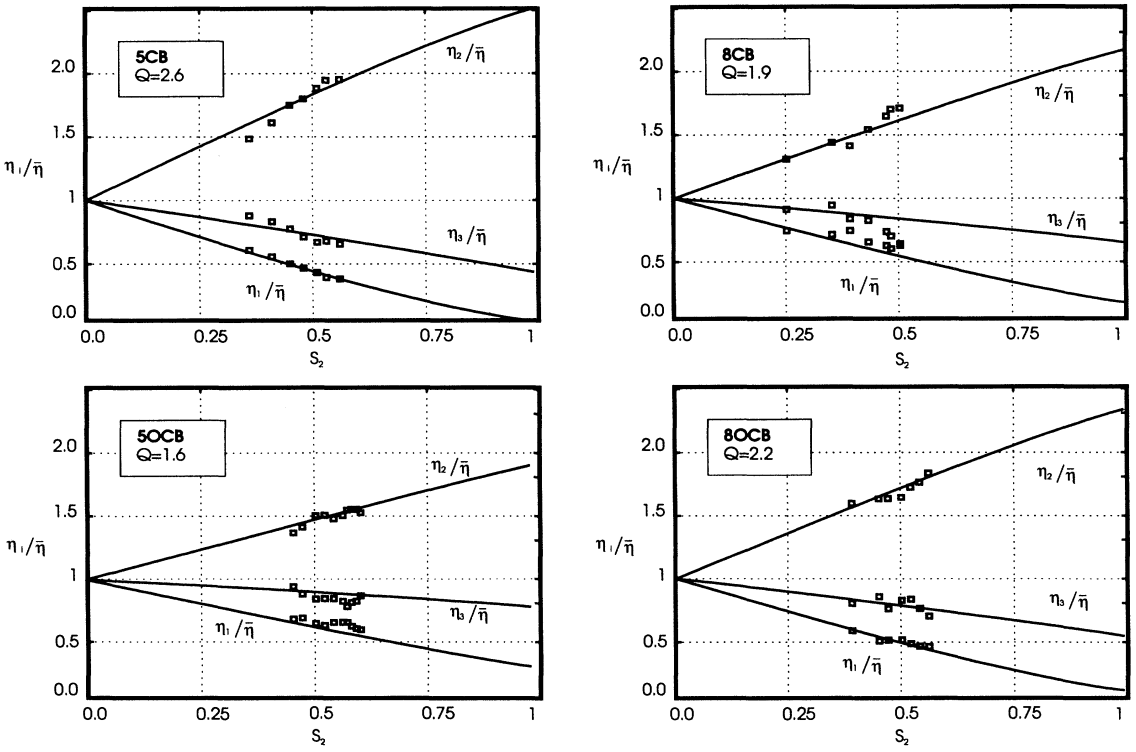


FIG. 8. Relative Miesowicz viscosities of *k*CB and *k*OCB ($k = 5, 8$); data taken from [22].

decreasing sensitivity with increasing chain length are in total accordance with the expectation arising from the molecular structure of those substances.

Close to the transition to a smectic phase the viscosities η_1 and η_3 for 8OCB [24] and 8CB [25] show an intersection when plotted against temperature. Such a pretransitional behavior, of course, is not described by the present approach.

CONCLUSIONS

For constrained flow experiments with strongly hindered rotation of the molecular axes and stationary director the complete set of viscosity coefficients could be calculated. The rotation viscosity coefficient γ_1 had to be replaced by the rotation viscosity tensor $\gamma^{(1)}_{\nu\mu}$ and therefore this quantity becomes direction dependent. Considering this particular dependence on the director alignment the Miesowicz viscosities are obtained. However, the constraints mentioned above are too restrictive to deal with the phenomenon of flow alignment or the measurement of the rotational viscosity. Those experiments are performed with a "free" flow. The director is stationary but "free:" The director alignment relaxes to a certain spatial direction but this direction is due to the flow field and it is not prescribed by external fields.

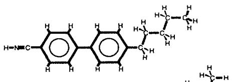
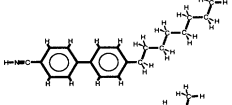
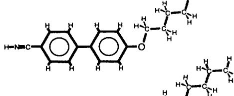
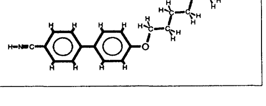
name	T_c[K]	structural formula
5CB 4-n-pentyl-4'-cyanobiphenyl	308.7	
8CB 4-n-octyl-4'-cyanobiphenyl	313.2	
5OCB 4-n-pentyloxy-4'-cyanobiphenyl	339.5	
8OCB 4-n-octyloxy-4'-cyanobiphenyl	353.0	

FIG. 9. Cyanobiphenyls; data taken from [22].

The affine transformation model for the viscosity coefficients of a totally aligned nematic or nematic discotic liquid crystal has successfully been applied to the presented approach. Thus the comparison with experimentally achieved coefficients is possible. The result shows a remarkable correspondence of theoretical and experimental data and confirms the affine transformation model, additional to nonequilibrium molecular dynamic (NEMD) numerical results obtained previously [3,4,6,7]. The fit parameter of the model is the axis ratio of the molecules which could be determined for MBBA and N4 in agreement with the shape of these particular molecules. For the cyanobiphenyls the axis ratios obtained can be easily understood when the molecular structure of these substances is taken into account. Thus they support the modified transformation model on a basis that should allow for the prediction of relative viscosities from the steric conformation of the molecules.

The model presented so far can be applied not only to nematic liquid crystals but also to nematic discotic ones. Due to a lack of measured viscosities of discotic liquid crystals the experimental verification of the theoretical predictions is still to be done. However, there are further theoretical results on disklike nematic liquid crystals [26,27] which support the results obtained in this paper.

ACKNOWLEDGMENTS

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APPENDIX: FUNCTIONAL RELATION OF S_4 AND S_2

Given a set of expectation values $g_i = \langle G_i \rangle$ $(i = 1, \ldots, n)$ it is a common problem of statistical physics to determine a reasonable distribution function f which reproduces the g_i. According to the principle of maximum entropy the distribution function which is the most unprejudiced guess is a function of exponential type:

$$f = \exp\left(\sum_{i=1}^{n} \lambda_i G_i\right) \Big/ Z \ .$$

The Lagrange parameters λ_i have to be determined from the n relations $g_i = \langle G_i \rangle_f$; Z denotes the partition function.

In the spirit of these general observations we want to establish a relation between S_4 and S_2. Starting with $S_2 = \langle P_2 \rangle$ the resulting uniaxial orientation distribution function f with macroscopic director $\boldsymbol{n}$ is given by

$$f(\boldsymbol{n} \cdot \boldsymbol{u}) = \exp[\lambda P_2(\boldsymbol{n} \cdot \boldsymbol{u})]/Z \ , \qquad \text{(A1)}$$

$$Z = \int \exp[\lambda P_2(\boldsymbol{n} \cdot \boldsymbol{u})]\, d^2u \ . \qquad \text{(A2)}$$

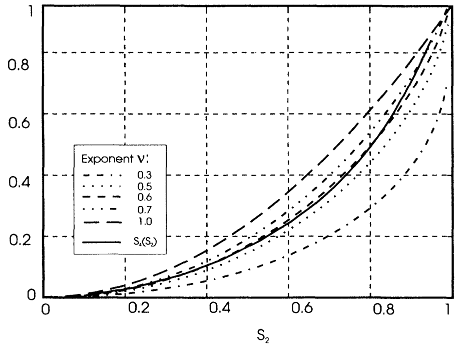

FIG. 10. Functional dependence of S_4 on S_2 according to the principle of maximum entropy together with various fit functions.

We obtain

$$S_2(\lambda) = \int P_2(\boldsymbol{n} \cdot \boldsymbol{u}) \exp[\lambda P_2(\boldsymbol{n} \cdot \boldsymbol{u})]\, d^2u \ , \qquad \text{(A3)}$$

$$S_4(\lambda) = \int P_4(\boldsymbol{n} \cdot \boldsymbol{u}) \exp[\lambda P_2(\boldsymbol{n} \cdot \boldsymbol{u})]\, d^2u \qquad \text{(A4)}$$

as the parametrized functional relation of S_4 and S_2. It should be noted that both S_2 and S_4 can be expressed with the help of derivatives of the partition function Z with respect to λ involving the error function. However, this specific form is not needed here. It is worth mentioning that—although based on totally different physical arguments—the Maier-Saupe orientational distribution function shows the same behavior as the function in Eq. (A1).

For practical purposes Eqs. (A3) and (A4) are too complicated. As shown in Fig. 10 the relation between S_4 and S_2 can be closely approximated in the experimentally relevant interval of S_2 [0.4, 0.8] by the fit function

$$S_4(S_2, \nu) = S_2\,[1 - (1 - S_2)^{\nu}] \qquad \text{(A5)}$$

with $\nu = 0.6$.

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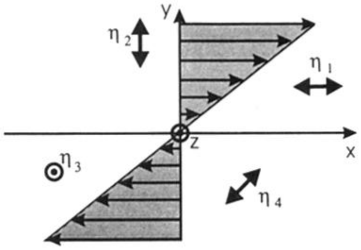

FIG. 1. Geometry of a plane Couette flow and definition of the Miesowicz viscosities.

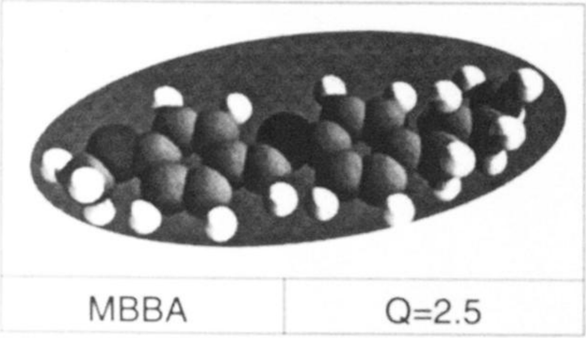

FIG. 4. Space filling model of MBBA and enclosing ellipsoid with axis ratio 2.5.