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# Molecular Crystals and Liquid Crystals

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# Friction Drag on a Sphere Moving in a Nematic Liquid Crystal

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# Friction Drag on a Sphere Moving in a Nematic Liquid Crystal

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The apparent viscosity  $\eta_{FB}$  of a nematic liquid crystal measured in a falling ball experiment is computed, assuming that the velocity distribution around the sphere is the same as in isotropic liquids for conditions of creeping flow. It is derived a general expression giving  $\eta_{FB}$  as a function of the director distribution which allows an easy computation of  $\eta_{FB}$  when an external field is present. For the case where there is no external field present, the apparent viscosity is also computed, and the result is (to a good approximation)

$$\eta_{FB} = \frac{1}{2}\alpha_4 + \frac{1}{4}(\gamma_3 - \gamma_1 + \frac{1}{2}\alpha_1)$$

This expression is valid when the hydrodynamic torque is zero and it is rather independent (less than 1%) of the particular distribution of the director provided that the hydrodynamic torque is zero. It is also shown that, for this last situation, director tumbling is expected to occur even when  $-\gamma_1/\gamma_2 < 1$ , and that although  $\eta_{FB}$  is numerically close to the Miesowicz viscosity  $\eta_b$  the nematic director should not be necessarily oriented along the flow direction. These results are compared to the experimental data about MBBA and the agreement found is good.

#### 1. INTRODUCTION

Nematic liquid crystals show five independent viscosity coefficients, and their apparent viscosity depends on the relative orientation among the director **n** (the optic axis), the velocity **v**, and the velocity gradient. The precise determination of all these viscosities requires the use of appropriate viscometers, 3-5 but conventional viscometry has also been performed since the early days up to now. Conventional viscometers are easier to work with but the interpretation of the experimental

results is difficulted by the complicated structure of the flows generated inside. As an example, the pressure dependence of the nematic viscosities has only been measured up to now by the falling ball method. The interpretation of these results, e.g. in terms of the relationship between the measured apparent viscosity  $\eta_{FB}$  and other molecular properties, cannot be satisfactorily done unless  $\eta_{FB}$  has been related to the standard nematic viscosities.

The aim of the present paper is to relate  $\eta_{FB}$  to the complete set of nematic viscosities  $\alpha_1$ ,  $\alpha_4$ ,  $\gamma_1$ ,  $\gamma_2$  and  $\gamma_3$  (defined below) assuming that creeping flow prevails. A general expression relating  $\eta_{FB}$  to the other nematic viscosities is presented in section 2. Next, in section 3,  $\eta_{FB}$  is explicitly computed for some director patterns. The results of the preceding sections are compared to the experimental data available on MBBA (section 4), and finally, in section 5, the main conclusions of this paper are presented together with some suggestions for further work.

# 2. GENERAL EXPRESSION FOR $\eta_{FR}$

It is well known that the friction force F acting on a ball moving at constant velocity  $v_{\infty}$  in a liquid can be related to the rate of dissipation per unit volume of the fluid,  $T\dot{\Sigma}$ , by the following expression<sup>9</sup>

$$F \cdot v_{\infty} = -\int d^3 \mathbf{r} \, T \dot{\Sigma} \tag{2.1}$$

where T is the temperature,  $\dot{\Sigma}$  is the rate of entropy production per unit volume, and the integral extends over the whole volume of the fluid. For a nematic liquid crystal  $T\dot{\Sigma}$  is given by  $^{10}$ 

$$T\dot{\Sigma} = \hat{\sigma}' : A + \mathbf{h} \cdot \mathbf{N} \tag{2.2}$$

where  $\sigma'$  is the viscous stress tensor,  $\hat{\sigma}'$  is the symmetric part of  $\sigma'$ , A is the symmetric velocity gradient tensor, h is the molecular field and N is the velocity of the director relative to the environning fluid

$$\mathbf{N} = \frac{d\mathbf{n}}{dt} - \frac{1}{2}(\operatorname{rot} \mathbf{v} \times \mathbf{n}) \tag{2.3}$$

The molecular field h is related to the antisymmetric part of the viscous stress tensor, ô' by the following expression

$$\epsilon_{ijk} \left( \sigma'_{jk} + n_j h_k \right) = 0 \tag{2.4}$$

In order to write explicitly eq. (2.1) it is convenient to consider the equivalent situation where the sphere of radius R is at rest and fixed to the origin of an orthogonal frame x, y, z, and the nematic flows along the positive z direction. Eq. (2.1) now reads

$$F = -uR^2 \cdot \int_0^{2\pi} d\phi \int_0^{\pi} \sin\theta \cdot d\theta \cdot \int_0^1 \frac{d\rho}{u^2 \rho^4} (\hat{\sigma}' : A + \mathbf{h} \cdot \mathbf{N}) \quad (2.5)$$

where  $\rho = R/r$ ,  $u = v_{\infty}/R$  and spherical coordinates  $(r, \theta, \phi)$  were used. By comparison of (2.5) to the well known Stokes formula for isotropic liquids<sup>11</sup>

$$F = -6\pi R^2 u \eta \tag{2.6}$$

we get

$$\eta_{\rm FB} = \frac{1}{6\pi} \int_0^{2\pi} d\phi \int_0^{\pi} \sin\theta \cdot d\theta \cdot \int_0^1 \frac{d\rho}{u^2 \rho^4} (\hat{\sigma}' : A + \mathbf{h} \cdot \mathbf{N}) \quad (2.7)$$

Now we assume that the velocity distribution around the sphere is the same as in isotropic liquids in conditions of creeping flow (Reynolds number  $Re \le 0.1$ ), that is<sup>11</sup>

$$\frac{v_r}{v_{\infty}} = \cos\theta \cdot \frac{1}{4} (4 - 6\rho + 2\rho^3) = \cos\psi$$
 (2.8)

$$\frac{v_{\theta}}{v_{\infty}} = -\sin\theta \cdot \frac{1}{4} (4 - 3\rho - \rho^3) = -\sin\psi \tag{2.9}$$

The validity of this hypothesis will be discussed below. From these expressions, the symmetric part  $A_{ij}$  and the antisymmetric part  $\mathbf{W} = (1/2)\text{rot }\mathbf{v}$  of the velocity gradient tensor are readily found. The only non-zero components are

$$A_{rr} = 2\lambda_1 \cdot u\rho^2$$

$$A_{\theta\theta} = A_{\phi\phi} = -\lambda_1 \cdot u\rho^2$$

$$A_{r\theta} = -\lambda_2 u\rho^2$$

$$W_{r\theta} = -W_{\theta r} = -u\lambda_2$$
(2.10)

where

$$\lambda_1 = \frac{3}{4} (1 - \rho^2) \cos \theta$$

$$\lambda_2 = \frac{3}{4} \rho^2 \sin \theta \tag{2.11}$$

On the other hand, when  $d\mathbf{n}/dt$  is zero, the only non-zero components of N are

$$N_r = W_{r\theta} n_{\theta}$$

$$N_{\theta} = -W_{r\theta} n_{r}$$
(2.12)

Now, using the constitutive equations of Leslie nematodynamics<sup>1,10</sup>

$$\hat{\sigma}'_{ij} = \frac{1}{2} \gamma_2 (n_i N_j + N_i n_j) + \frac{1}{2} \gamma_3 (n_i n_k A_{kj} + n_k A_{ki} n_j) + \alpha_4 A_{ij} + \alpha_1 n_k A_{kp} n_p n_i n_j$$
(2.13)

$$h_i = \gamma_1 N_i + \gamma_2 n_k A_{ki} \tag{2.14}$$

where  $\alpha_1$ ,  $\alpha_4$ ,  $\gamma_1 = \alpha_3 - \alpha_2$ ,  $\gamma_2 = \alpha_3 + \alpha_2 = \alpha_6 - \alpha_5$  and  $\gamma_3 = \alpha_6 + \alpha_5$  constitute a complete set of nematic viscosities, it is possible to write explicitly both terms of the rate of dissipation per unit volume. They are

$$\begin{split} \hat{\mathbf{\sigma}}' : A &= \alpha_{1} \Big[ n_{r}^{2} A_{rr} + n_{\theta}^{2} A_{\theta\theta} + n_{\phi}^{2} A_{\phi\phi} + 2 n_{r} n_{\theta} A_{r\theta} \Big]^{2} \\ &+ \alpha_{4} \Big[ A_{rr}^{2} + 2 A_{r\theta}^{2} + A_{\theta\theta}^{2} + A_{\phi\phi}^{2} \Big] \\ &+ \gamma_{3} \Big[ n_{r}^{2} A_{rr}^{2} + n_{\theta}^{2} A_{\theta\theta}^{2} + n_{\phi}^{2} A_{\phi\phi}^{2} + \left( n_{r}^{2} + n_{\theta}^{2} \right) A_{r\theta}^{2} \\ &+ 2 n_{r} n_{\theta} A_{r\theta} (A_{rr} + A_{\theta\theta}) \Big] \\ &+ \gamma_{2} W_{r\theta} \Big[ \left( n_{\theta}^{2} - n_{r}^{2} \right) A_{r\theta} + n_{r} n_{\theta} (A_{rr} - A_{\theta\theta}) \Big] \\ &\mathbf{h} \cdot \mathbf{N} = \gamma_{1} \Big( n_{r}^{2} + n_{\theta}^{2} \Big) W_{r\theta}^{2} + \gamma_{2} W_{r\theta} \Big[ \Big( n_{\theta}^{2} - n_{r}^{2} \right) A_{r\theta} + n_{r} n_{\theta} (A_{rr} - A_{\theta\theta}) \Big] \end{split} \tag{2.15}$$

For a distribution of the director of the form

$$n_r = \sin \xi \cdot \cos \zeta$$

$$n_\theta = \sin \xi \cdot \sin \zeta$$

$$n_\phi = \cos \xi$$
(2.17)

Eq. (2.7) takes the form

$$\eta_{\rm FB} = \frac{1}{6\pi} \int d\Omega (a_1 + a_4 + g_1 + g_2 + g_3) \tag{2.18}$$

where

$$\int d\Omega = \int_0^{2\pi} d\phi \int_0^{\pi} \sin\theta \cdot d\theta \int_0^1 d\rho \tag{2.19}$$

$$a_1 = \alpha_1 \left\{ -\lambda_1 + \sin^2 \xi \left[ \frac{3}{2} \lambda_1 (1 + \cos(2\zeta)) + \lambda_2 \sin(2\zeta) \right] \right\}^2$$
 (2.20)

$$a_4 = \alpha_4 \left[ 6\lambda_1^2 + 2\lambda_2^2 \right] \tag{2.21}$$

$$g_1 = \gamma_1 \frac{\lambda_2^2}{\rho^4} \cdot \sin^2 \xi = \frac{9}{16} \gamma_1 \sin^2 \theta \cdot \sin^2 \xi \tag{2.22}$$

$$g_2 = -\frac{3}{2}\gamma_2 \sin\theta \cdot \sin^2\xi \left[\frac{3}{2}\lambda_1 \cdot \sin(2\zeta) + \lambda_2 \cos(2\zeta)\right]$$
 (2.23)

$$g_3 = \gamma_3 \left\{ \lambda_1^2 + \sin^2 \xi \left[ \lambda_2^2 + \frac{3}{2} \lambda_1^2 (1 + \cos(2\zeta)) - \lambda_1 \lambda_2 \sin(2\zeta) \right] \right\}$$
(2.24)

Expression (2.18) supplemented by expressions (2.20)–(2.24) allows the explicit computation of  $\eta_{FB}$  when the distribution of the director (2.17) is known, and it reduces to Stokes formula (Eq. (2.6)) when  $a_1$ ,  $g_2$ , and  $g_3$  are zero. But unless a particular director pattern has been imposed by an external field (e.g. a magnetic or an electric field) the precise computation of the director distribution involves the consideration of the relevant elastic terms of the stress tensor together with suitable boundary conditions so the problem is converted into an almost insoluble one. Therefore some simplifying assumptions must be done.

For the most common kinds of flows considered in practice, the viscous forces are largely predominant over the elastic ones, i.e. the Ericksen number<sup>12</sup> (Er = viscous forces/elastic forces) is large enough in order to have a negligible contribution from the elastic forces. This argument may also justify the hypothesis formulated above that the velocity profile is the same as in isotropic liquids, so the elastic terms have a negligible contribution to the total pressure. However the contribution of the disclinations to the distribution of the director field must not be neglected in general, and its contribution to the apparent viscosity should be considered. Since the velocity gradient decays quickly as the distance from the sphere increases (see Eq. (2.10)), and because the dissipation is proportional to the square of the velocity gradient, we may expect that the apparent viscosity will depend mainly on the director pattern near the surface of the ball so the contribution of the disclinations far from the sphere shall be neglected. As we shall see in the next section, the condition of a

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vanishing overall hydrodynamic torque leads to an expression for  $\eta_{FB}$  which does not depend too strongly on the particular director pattern; therefore  $\eta_{FB}$  may be computed to a good approximation even when director tumbling does not allow a precise computation of  $\eta_{FB}$ .

# 3. EXPLICIT COMPUTATION OF $\eta_{FB}$

Before going on let us recall some of the known results about simple shear flow in a nematic liquid crystal. For the three situations where:
a) n is oriented perpendicular to both the velocity and the velocity gradient, b) n is oriented along the velocity vector, and c) n is parallel to the velocity gradient, the measured apparent viscosities are the so-called Miesowicz viscosities defined as<sup>1,10</sup>

$$\eta_a = \frac{1}{2}\alpha_4 \tag{3.1}$$

$$\eta_b = \frac{1}{2}\alpha_4 + \frac{1}{4}(\gamma_1 + \gamma_3 + 2\gamma_2) \tag{3.2}$$

$$\eta_c = \frac{1}{2}\alpha_4 + \frac{1}{4}(\gamma_1 + \gamma_3 - 2\gamma_2) \tag{3.3}$$

Another interesting situation occurs where there is no external field present: the condition of zero hydrodynamic torque imposes that the director is on the plane defined by  $\mathbf{v}$  and  $\nabla \mathbf{v}$ , and that the angle  $\beta_0$  between  $\mathbf{n}$  and  $\mathbf{v}$  is such that

$$\cos(2\beta_0) = -\frac{\gamma_1}{\gamma_2} \tag{3.4}$$

The apparent viscosity is now given by

$$\eta_0 = \frac{1}{2}\alpha_4 + \frac{1}{4}(\gamma_3 - \gamma_1) + \frac{1}{8}\alpha_1[1 - \cos(4\beta_0)]$$
 (3.5)

Because  $\gamma_1$  is close to  $|\gamma_2|$  for most nematics,  $\eta_0$  is also close to  $\eta_b$ . When  $\gamma_1 > |\gamma_2|$  Eq. (3.4) has no sense, and the angle  $\beta$  between n and v is not constant: the director is said to "tumble".

For the case of the flow around a sphere, the comparison to the simple shear flow is always doubtful since the velocity depends now on the coordinates r and  $\theta$ .

Let us consider first the simpler case where  $\xi = 0$  everywhere  $(n_{\phi} = 1)$ . From the results of the preceding section it is straightforward to show that  $\eta_{FB}$  is given by

$$\eta_{\rm FB}(\xi=0) = \frac{1}{2}\alpha_4 + \frac{1}{15}(\gamma_3 + \alpha_1) \tag{3.6}$$

which is numerically close to  $\eta_a$ ; but here the hydrodynamic torque is zero in contrast to what happens in case a) of simple shear flow.

Another situation easy to solve is that when the director is oriented along the z axis everywhere. In order to get  $\eta_{FB}$ , the integrals involving  $a_1$ ,  $g_2$  and  $g_3$  (Eqs. (2.20), (2.23), (2.24)) were computed numerically; the final result is

$$\eta_{\rm FB}(\mathbf{n}||\mathbf{z}) = \frac{1}{2}\alpha_4 + \frac{1}{4}(\gamma_1 + \gamma_3) + 0.3 \times \gamma_2 + 0.1143 \times \alpha_1 \quad (3.7)$$

and it is very different from any of the Miesowicz viscosities.

A third situation which has some interest to consider is that when the director is constrained to lay on the  $(r, \theta)$  plane and to make a constant angle  $\beta$  with the velocity. Here  $\xi = \pi/2$ , and  $\zeta$  becomes a known function of  $\psi$  and  $\beta$  so the apparent viscosity can be computed again. The result is

$$\eta_{FB}(\beta) = \frac{1}{2}\alpha_4 + \frac{1}{4}\left(\gamma_1 + \frac{13}{15}\gamma_3 + \frac{7}{15}\alpha_1\right) \\
+ \cos(2\beta) \cdot \frac{1}{4}\left[1.2906 \times \gamma_2 + 6.76 \times 10^{-3}(\gamma_3 + \alpha_1)\right] \\
- \cos(4\beta) \times \frac{0.242}{4} \times \alpha_1 \tag{3.8}$$

and for  $\beta = 0$ ,  $\eta_{FR}(\beta)$  becomes

$$\eta_{FB}(\beta = 0) = \frac{1}{2}\alpha_4 + \frac{1}{4}(\gamma_1 + 0.8734 \times \gamma_3 + 1.2906 \times \gamma_2 + 0.2314 \times \alpha_1)$$
 (3.9)

Again the apparent viscosity is very different from that for the "corresponding" situation of simple shear flow  $(\eta_0)$ . Moreover if we try to find an angle  $\beta_0$  such that the hydrodynamic torque vanishes we get

$$\cos(2\beta_0) = -\frac{2\gamma_1}{1.2906 \times \gamma_2}$$

that is

$$\gamma_1 \leq 0.645 \times |\gamma_2|;$$

but there is no nematic material that fullfils this last inequality, to our knowledge.

In view of the preceding results, the question about how to compute  $\eta_{FB}$  when there is no external field acting on the director is now in

order. For this case the director field is not a given quantity so we must include some additional assumptions in order to get it. The simpler one is that of minimum dissipation: then the hydrodynamic torque must vanish as one easily sees by inspection of Eqs. (2.15) and (2.16). For instance, if  $\tilde{\sigma}_{\theta r} = 0$  then

$$\int d\Omega \left(g_1 + \frac{1}{2}g_2\right) = 0 \tag{3.10}$$

(see Eqs. (2.15), (2.16), (2.22) and (2.23); moreover, if  $n_{\phi} = 0$  then the contribution to  $\eta_{\rm FB}$  from the terms of (2.18) involving  $\gamma_1$  and  $\gamma_2$  becomes simply equal to  $-\gamma_1/4$ . Now, looking at Eqs. (2.15) to (2.24) it is apparent that the dissipation shall be minimum when  $\sin^2 \xi$  is maximum, i.e.,  $n_{\phi} = 0$ ; for this case, the apparent viscosity may be written as

$$\eta_{\rm FB}(\xi = \pi/2) = \frac{1}{2}\alpha_4 + \frac{1}{4}(\frac{13}{15}\gamma_3 - \gamma_1 + \frac{14}{30}\alpha_1) + \Delta\eta$$
 (3.11)

where the first two terms include the contributions from  $a_1$  and  $g_3$  which do not depend on  $\zeta$ , and  $\Delta \eta$  stands for the remaining terms. As  $\zeta$  depends now on the spatial coordinates, the precise computation of  $\Delta \eta$  is not so easy to perform; nevertheless  $\Delta \eta$  may be estimated from the preceding results. In fact,  $\zeta$  may be expressed as a function of the angle  $\beta$  between  $\eta$  and  $\eta$ , and of the angle  $\psi$  between  $\eta$  and the r-axis. We tried to estimate an upper limit to  $\Delta \eta$ , and by inspection of the corresponding formulae we arrived to the conclusion that the two following distributions of  $\beta$ :

a)  $\cos(2\beta) = 0$  for  $-\pi/4 < \theta < \pi/4$ , and  $\cos(2\beta) = 1$  otherwise b)  $\cos(2\beta) = 1$  everywhere

were good approximations to get  $\Delta \eta_{\text{max}}$ . By numeric computation we found respectively

a) 
$$\Delta \eta = 1.963 \times 10^{-2} \times \gamma_3 - 7.869 \times 10^{-3} \times \alpha_1$$

b) 
$$\Delta \eta = 1.69 \times 10^{-3} \times \gamma_3 - 5.88 \times 10^{-2} \times \alpha_1$$

Thus, expression (3.11) may be written as

$$\eta_{\rm FB}(\xi = \pi/2) = \frac{1}{2}\alpha_4 + \frac{1}{4}(\gamma_3 - \gamma_1) + \frac{1}{8}\alpha_1 \tag{3.12}$$

without appreciable error. As an example, for MBBA we found that expressions (3.11) and (3.12) differ by less than 1% whatever it was the chosen distribution for  $\beta$ : a) or b).

Therefore we could find a reasonable value for  $\eta_{FB}$  in the absence of an external field without a detailed knowledge of the director distribution and without too much numerical computation too. Let us remark that the value for  $\eta_{FB}$  provided by Eq. (3.12) is numerically very close to the viscosities  $\eta_b$  (or  $\eta_0$ ) in good agreement to what has been experimentally found<sup>8,13</sup> (see next section).

Now let us try to find a distribution of the director such that the hydrodynamic torque is zero everywhere, i.e. the integrand of Eq. (3.10) is zero. This condition may be written as

$$\sin^2 \xi \left\{ -\frac{\gamma_1}{\gamma_2} + \frac{3}{2} (1 - \rho^2) \cot \theta \cdot \sin(2\zeta) + \rho^2 \cos(2\zeta) \right\} = 0$$
(3.13)

and besides the trivial solution  $\xi = 0$  considered before (Eq. (3.6)), the other solution of Eq. (3.13) is

$$\cos(2\zeta) = \frac{1}{a^2 + \rho^4} \left\{ -g\rho^2 \pm |a| \cdot (a^2 + \rho^4 - g^2)^{1/2} \right\} \quad (3.14)$$

where

$$g = \frac{\gamma_1}{\gamma_2}$$
 and  $a = \frac{3}{2}(1 - \rho^2)\cot\theta$ 

Hence  $cos(2\zeta)$  is a real quantity if and only if

i) 
$$r \le R/g^{1/2}$$
 for every  $\theta$  (3.15)

ii) 
$$|\tan \theta| < \frac{3}{2} \frac{(1-\rho^2)}{(g^2-\rho^4)^{1/2}} \quad \text{for } r > R/g^{1/2}$$
 (3.16)

Outside the region defined by these two conditions one must have  $\xi = 0$  in order to satisfy Eq. (3.13); otherwise the director shall tumble in order that Eq. (3.10) will be satisfied. Thus we conclude that the hypothesis of zero hydrodynamic torque in every point of the nematic material leads to the rather artificial situation where  $\xi = \pm \pi/2$  and  $\zeta$  is given by Eq. (3.13) in the regions of space defined by expressions (3.15) and (3.16), and  $\xi = 0$  outside; the apparent viscosity for this situation depends on  $g = \gamma_1/\gamma_2$ , but, whatever the chosen solution of Eq. (3.13), we verified that  $\eta_{\rm FB}$  is much greater than that given by Eq. (3.12), so it must not be considered.

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Let us summarize some of the results presented above. First, even for  $\gamma_1/|\gamma_2| < 1$  there is a region around the sphere where the director shall tumble in order to vanish the overall hydrodynamic torque. As  $\gamma_1/|\gamma_2|$  increases and approaches one, this region (where inequality (3.16) does not hold) approaches the surface of the ball, i.e., the region where the velocity gradients are important. For  $\gamma_1/|\gamma_2| > 1$  one may expect that the flow in the wake of the ball shall be perturbed, but the apparent viscosity will not change too much in view of the estimate given by expression (3.12). This description is supported by the experimental observation of the flow of a nematic around a sphere as described in Ref. 13.

# 4. COMPARISON WITH THE EXPERIMENTAL DATA

Figure 1 displays the experimental values of  $\eta_{\rm FB}^{8,13}$  and  $\eta_b^{3,4,14}$  measured by different authors on MBBA. This material was chosen because it is one of the few nematic materials for which the order

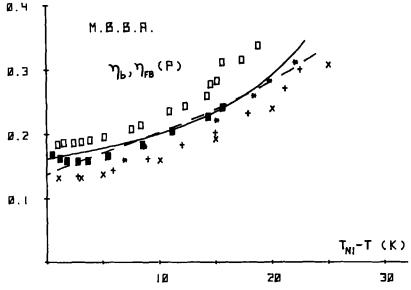


FIGURE 1 Temperature dependence of the falling ball apparent viscosity  $\eta_{FB}$  of M.B.B.A.. The full curve represents  $\eta_{FB}$  computed using Eq. (3.12) and the theoretical expressions for the nematic viscosities  $\gamma_1(T)$ ,  $\gamma_3(T)$ ,  $\alpha_4(T)$  and  $\alpha_1(T)$  given in the text without least squares fit. Experimental values of  $\eta_{FB}(T)$ :  $\blacksquare$  from Ref. 8;  $\square$  from Ref. 13. For the sake of completeness  $\eta_b(T)$  has also been computed from Eq. (3.2) and it is plotted (broken curve) together with the following experimental values of  $\eta_b(T)$ : + from Ref. 3;  $\times$  from Ref. 4; \* from Ref. 14.

parameter and temperature dependence of all its hydrodynamic viscosities is known. As it is apparent from Figure 1,  $\eta_{FB}$  is numerically very close to the Miesowicz viscosity  $\eta_b$  as pointed out by Refs. 8, 13, but, in view of the results of the preceding section, the viscosity  $\eta_0$  should be preferably chosen. The small discrepancy shown by the data taken from Ref. 13 is mainly due to the purity of the sample used  $(T_{NI} \approx 40 \, ^{\circ}\text{C})$  as compared to the other ones  $(T_{NI} \approx 46 \, ^{\circ}\text{C})$ .

We also computed the values of  $\eta_{FB}$  (from Eq. (3.12)) and  $\eta_b$  (from Eq. (3.2)) taking the theoretical expressions for  $\alpha_1(T)$ ,  $\alpha_4(T)$ ,  $\gamma_1(T)$ ,  $\gamma_2(T)$  and  $\gamma_3(T)$  presented in Refs. 15, 16. They are:

$$\alpha_1(T) = c_1 S^2 \exp\left(\frac{\theta_3 S^2}{T - T_0}\right) \tag{4.1}$$

$$\alpha_4(T) = \left(a - \frac{1}{3}bS\right) \exp\left(\frac{\theta_3 S^2}{T - T_0}\right) \tag{4.2}$$

$$\gamma_1(T) = g_1 \cdot S^2 \exp\left(\frac{\theta_1 S^2}{T - T_0}\right) \tag{4.3}$$

$$\gamma_2(T) = g_2 S \exp\left(\frac{\theta_2 S^2}{T - T_0}\right) \tag{4.4}$$

$$\gamma_3(T) = bS \exp(\theta_3 S^2 / T - T_0) \tag{4.5}$$

where S is the order parameter, T is the temperature and the other symbols stand for parameters which are (nearly) temperature independent. Their values for MBBA are:  $^{16}$   $c_1 = -0.147P$ , a = 0.296P, b = 0.188P,  $g_1 = 1.066P$ ,  $g_2 = -0.440P$ ,  $\theta_1 = 115.81K$ ,  $\theta_2 = 166.60K$ ,  $\theta_3 = 153.82K$  and  $T_0 = 255K$ . The curves  $\eta_{FB}(T)$  and  $\eta_b(T)$  computed by this way are plotted on Figure 1, and the agreement shown between these curves and the experimental results is good in view of the dispersion of the experimental points. In addition, we remark that the main source of error in the computation of  $\eta_{FB}(T)$  or  $\eta_b(T)$  comes from the dispersion (about 5 cp) of the experimental points used to compute  $\gamma_3(T)$  (see Ref. 16) which causes a slight overestimation of  $\eta_b$ .

#### 5. CONCLUSION

Starting from a general expression giving the apparent viscosity  $\eta_{FB}$  of a nematic measured in a falling ball experiment as a function of the distribution of the director field, we were able to relate  $\eta_{FB}$  to the

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standard nematic viscosities for some of the flows which may occur in this kind of viscometers. As a consequence, the range of application of falling ball viscometry to the measurement of the nematic viscosities has been increased, and, provided that a suitable orientation has been given to the director field (e.g. by an external field), other combinations of the standard nematic viscosities can also be measured. When an external field is absent, the structure of the velocity gradient tensor is such that director tumbling is present even for low Reynolds number and for  $-\gamma_1/\gamma_2 < 1$ . This is mainly due to the dependence of the velocity on the coordinates r,  $\theta$  so the analogy to simple shear flow is not meaningful, in general. But even in these conditions, the apparent viscosity  $\eta_{FB}$  may be estimated to a good approximation and the results were shown to be in good agreement to the experimental data.

As shown by Kuss,<sup>8</sup> falling ball viscometry can be used to investigate the pressure dependence of the nematic viscosities. The interpretation of such data can give some insight into the relationship between the molecular properties and the nematic viscosities provided that  $\eta_{\rm FB}$  has been previously related to the standard nematic viscosities. This work is currently under way, and it will be presented elsewhere.<sup>17</sup>

Most of the results contained in the present paper are easily generalized for the case of nematic polymers provided that they do not show a marked non-newtonian character. As a matter of fact the velocity gradient varies strongly in the vicinity of the falling ball so the interpretation of the results of an experiment performed with a strongly non-newtonian material needs further information on the rheological parameters of this material.

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