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## A Geometrical Approach to the Miesowicz's Coefficients

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*Here we will show how the use of a geometrical approach leads to a reduction of the number of the viscosity coefficients of the nematic liquid crystals. Usually they are described by five free parameters; we will show that a geometrical approach can reduce this number to three. This result will follow from the modified version of the Hess-Baalss approach. As a consequence of the application of the Onsager relation, these three viscosity terms become related by the effective eccentricity of the nematic grain, leading to a connection between the Miesowicz's coefficients.*

**Keywords** Liquid crystals; rheology; nematic viscosity; nematic liquid crystals

### 1. Introduction

Since the discovery of liquid crystals (LCs), keen attention has been paid to their anisotropic properties. In particular, the understanding of the flow properties of these materials has been shown as one of the most challenging problems in the LCs physics [1]. Early in 1936 Miesowicz showed that, in the presence of an external magnetic field, LCs exhibiting a direction-dependent viscosity [2,3]. Since then, many efforts have been made in order to verify the theoretical predictions and completely characterize the flow properties of them [4–30], but a satisfactory microscopic theory for it has not been found yet [15–17]. Ericksen, Leslie and Parodi [4–9] derived a hydrodynamic theory of nematic liquid crystals (NLCs) according to which the rheological properties of incompressible nematics depend on five coefficients. Three of them can be chosen to be the Miesowicz viscosities. The kinetic approach of Doi was for some time the most accepted microscopic theory of the nematic viscosity [18–23], but even having the great merit of producing an expression free of adjustable parameters, which captures the essence of the phenomena and furnishes a semi-microscopic explanation to the origin of their anisotropy, it presents well-documented disagreements with the experimental data, shows to be unable to describe the essential aspects of the phenomenology observed in these systems, especially when the entire range of the nematic phase is considered [15–17]. Baalss, Hess and co-workers [24–28] proposed

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a different theory for the viscosities of NLCs - the HB approach. According to this approach, all mathematical quantities describing a spherical symmetric system can be transformed, by means of an affine transformation, into objects which describe the corresponding quantities of a system with ellipsoidal symmetry. Although this approach does not intend to propose that nematic viscosity is a geometrical problem; it has been proposed that many aspects of nematic viscosity can be better understood if a geometrical point of view is considered [15–17, 31–38].

Recently, Simões and co-workers [31–33] showed that the usual HB approach to the nematic viscosity is incomplete. Furthermore, they proposed a geometrical approach to express the Leslie's viscosity coefficients [33]. In this paper, we will show how the use of this geometrical approach leads to a reduction of the number of viscosity coefficients. Usually they are described by five free parameters; we will show that they can be reduced to three.

## 2. Fundamentals

In this section we shall express Leslie's viscosity coefficients in terms of the eccentricity of a uniaxial ellipsoid. The argument will proceed along the standard method of the thermodynamics of irreversible processes.

According to the Ericksen, Leslie and Parodi (ELP) approach [4–9], the general expression for the dissipation associated with the viscous flow in a nematic sample is given by

$$T\dot{S} = \int d^3r \left( \sigma_{ij} A_{ij} + \vec{h} \cdot \vec{N} \right). \quad (1)$$

The first, characterized by a fluid flow with the form  $A_{ij} = (\partial_i v_j + \partial_j v_i)/2$ , describes the shear flow dissipation. The second, characterized by the term  $\vec{N} = \dot{\vec{n}} - \vec{w} \times \vec{n}$ , describes the rotation of the director with respect to the background fluid, and gives to the rotational dissipation. In these equations,  $\vec{n}$  is the nematic director field, and the variables  $\vec{v}$  and  $\vec{w}$  are point dependent fluxes, where  $\vec{v}$  is velocity of the fluid and  $\vec{w}$  is the director's angular velocity. Associated with each of these fluxes are the thermodynamical forces  $\sigma_{ij}$  and  $h_i$ , which describe, respectively, the dissipative stress tensor and the molecular field [1]. The Onsager theorem states that, in the linear regime, the generalized forces  $\sigma_{ij}$  and  $h_i$  are proportional to the fluxes  $A_{ij}$  and  $\vec{N}$ . The dissipative stress tensor of the ELP approach,

$$\sigma_{ij} = \alpha_1 n_i n_j n_k n_l A_{kl} + \alpha_2 n_i \Psi_{jk} n_k + \alpha_3 n_j \Psi_{ik} n_k + \alpha_4 A_{ij} + \alpha_5 n_i A_{jk} n_k + \alpha_6 n_j A_{ik} n_k, \quad (2)$$

where  $\alpha_1, \alpha_2, \dots, \alpha_6$  are the Leslie coefficients,  $\Psi_{ik} n_k = \dot{n}_i - W_{ik} n_k$  describes the rotation of the director  $\vec{n}$  relative to the background fluid, and  $W_{ij} = (\partial_i v_j - \partial_j v_i)/2$ . The molecular field is proportional to the antisymmetric part of the stress tensor,  $\sigma_{ij}^A = (h_i n_j - n_i h_j)/2$ , and it is given by,

$$h_i = (\alpha_2 + \alpha_3) n_k A_{ik} + (\alpha_3 - \alpha_2) N_i. \quad (3)$$

To derive these equations, Ericksen and Leslie took into account the spatial symmetry properties of the medium, and the equality of action and reaction. Parodi showed that the Onsager reciprocal relations [30], which reflect the time-reversal invariance of the equations of motion of the individual particles, applied in Eqs. (2) and (3) above, lead to the relation

$\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$  that connect the shearing flow and rotational flow terms of the two generalized forces,  $\sigma_{ij}$  and  $h_i$ . So, when the Ericksen-Leslie approach is considered, only five independent combinations of the viscosity coefficients need to be effectively measured. It is not the Leslie coefficients  $\alpha_i$  which are determined experimentally, but the coefficients of viscosity for a nematic liquid crystal with a constant velocity gradient (or constant shear rate) and the coefficient of viscosity describing director rotation.

Miesowicz defined three laminar flow geometries suitable for studying the shear viscosity coefficient anisotropy in the nematic phase, namely,  $\hat{n}||\vec{\nabla}v$ ,  $\hat{n}||\vec{v}$ , and  $\hat{n}\perp\vec{v}\perp\vec{\nabla}v$ . After Miesowicz, the shear viscosity coefficients measured under such circumstances are referred to as  $m_1$ ,  $m_2$  and  $m_3$ , respectively. Here, we have retained the notation of Helfrich [34] by which the definitions of  $m_1$  and  $m_2$  are interchanged. These viscosities are related to the  $\alpha_i$  by,

$$m_1 = (\alpha_4 + \alpha_5 - \alpha_2)/2, \quad m_2 = (\alpha_3 + \alpha_4 - \alpha_6)/2, \quad m_3 = \alpha_4/2. \quad (4)$$

The fourth independent experimental geometry for the viscosity coefficient measurement, the rotational viscosity  $\gamma_1$ , was proposed by Tsvetkov [7]. Finally, the fifth independent experimental geometry for the viscosity measurement, the viscosity  $m_{12}$ , was introduced by Helfrich [35]. The coefficients are linearly related to the theoretical ones, thus enabling rigorous verification of the ELP theory [36].

Hess and Balls and co-workers [24–28] have proposed a way by which the viscosity of an anisotropic liquid, the expression given at Eq. (2), can be written in terms of the viscosity of an isotropic liquid through an affine transformation. It is assumed that if one could imagine a way by which nematic molecules could be continuously deformed, up to the point in which they become spherical, it would be possible to observe a corresponding reduction of the macroscopic anisotropies, until they vanish. Inversely, if the idealized spherical molecules of an isotropic liquid could be deformed until they assumed the ellipsoidal form of an idealized nematic molecule; the macroscopic physical properties would be transformed into those observed in nematic liquid crystals. By definition of affine transformation, the HB approach is composed by two different mathematical entities: (a) the linear transformation between the tensor components  $ds^l/dx^i$ , and (b) the tensor components in which it acts,  $\partial_i v^k$ , where the letters  $x$  and  $s$  stand for ellipsoidal and spherical coordinates, respectively. So, if  $\sigma_{ij} = \eta \partial_i v_j$  gives the expression for the viscosity of an isotropic Newtonian fluid, where  $\eta$  is the corresponding isotropic viscosity and  $\partial_i v_j$  is the gradient of the isotropic fluid velocity, then

$$\sigma_{ij}^{HB} = \frac{ds^l}{dx^i} \frac{dx^j}{ds^k} (\eta \partial_l v^k), \quad (5)$$

would correspond to the Leslie expression to the viscosity of a nematic liquid. According to the reasoning followed at the previously quoted paper [17], a model of such deformations would be given by

$$\frac{dx^i}{ds^j} = r_b \delta_{ij} + (r_a - r_b) n_i n_j \quad \text{and} \quad \frac{ds_i}{dx_j} = \frac{1}{r_b} \delta_{ij} + \left( \frac{1}{r_a} - \frac{1}{r_b} \right) n_i n_j. \quad (6)$$

In these equations,  $r_a$  and  $r_b$  are dependent on the nematic order parameter  $S$  and characterize the thermalized shape of an ellipsoidal nematic grain [17,37–40],  $r_a$  describes the uniaxial axis of the thermalized grain and  $r_b$  describes the degenerated axis, in such a

way that the resulting effective eccentricity is given by

$$e = 1 - \frac{r_b^2}{r_a^2}. \quad (7)$$

The HB approach never explained completely the phenomenology observed in the LC's viscosity. Recently, Simões et al. [31–33] showed that Eq. (5) is incomplete because it does not take into account all degrees of freedom of an anisotropic particle; for an observer in the laboratory, the velocity  $\vec{v}$  of an extended anisotropic rigid body is composed of two terms,  $\vec{v} = \vec{v}^{cm} + \vec{w} \times \vec{\rho}$ , the first considering the motion of its centre of mass  $\vec{v}^{cm}$ , and the second considering the rotation of its internal points, at the position  $\vec{\rho}$  around the centre of mass. Both of these terms produce dissipation and, as the second term is not present in Eq. (5), the HB approach is incomplete.

In order to solve this problem and to construct a model that embraces all degrees of freedom, the form of the isotropic stress tensor has been modified with the introduction of three viscosity coefficients, one for the shearing flow and two for the rotational flow of a liquid, i.e.,

$$\sigma_{ij} = 2 \frac{ds^l}{dx^i} \frac{dx^j}{ds^k} (\eta_1 A_{lk} + \eta_2 N_l n_k + \eta_3 N_k n_l). \quad (8)$$

The parameters  $\eta_1$ ,  $\eta_2$  and  $\eta_3$  describe the dissipative channels. Specifically,  $\eta_1$  is the extension of the viscosity term already present in the usual HB approach,  $\eta_2$  and  $\eta_3$  have been introduced to account for the rotational viscosity. Here,  $N_k n_l$  is the transpose of  $N_l n_k$ . Moreover, by substituting Eq. (6) into Eq. (8) and comparing the resulting equation with the ELP form of the stress tensor [4–9], the Leslie coefficients are given by,

$$\begin{aligned} \alpha_1 &= -\frac{(r_a - r_b)^2}{r_a r_b} \eta_1, \quad \alpha_2 = \frac{r_a}{2r_b} \eta_2, \quad \alpha_3 = \frac{r_b}{2r_a} \eta_3, \\ \alpha_4 &= \eta_1, \quad \alpha_5 = \frac{r_a - r_b}{r_b} \eta_1, \quad \alpha_6 = \frac{r_b - r_a}{r_a} \eta_1. \end{aligned} \quad (9)$$

In terms of these new parameters, the Parodi relation becomes

$$e\eta_1 + \eta_2 + (1 - e)\eta_3 = 0. \quad (10)$$

This equation shows that the terms  $\eta_1$ ,  $\eta_2$  and  $\eta_3$  are connected by the quenched geometry of the nematic phase. Furthermore, as the NI phase-transition is approached, the effective form of the nematic grains loses its ellipsoid shape, becoming more and more spherical, and gives rise to an increase in the ratio  $r_b/r_a$ , making  $e \rightarrow 0$ . In this situation,  $\eta_2 + \eta_3 = 0$ . Finally, using Eq. (10) we see that the Leslie coefficients are completely determined by two viscosity parameters,  $\eta_1$  and  $\eta_3$ , for example, and by the eccentricity  $e$ :

$$\begin{aligned} \alpha_1 &= -\frac{(1 - \sqrt{1 - e})^2}{\sqrt{1 - e}} \eta_1, \quad \alpha_2 = -\frac{e\eta_1 + (1 - e)\eta_3}{\sqrt{1 - e}}, \quad \alpha_3 = \sqrt{1 - e} \eta_3, \\ \alpha_4 &= \eta_1, \quad \alpha_5 = \left( \frac{1}{\sqrt{1 - e}} - 1 \right) \eta_1, \quad \alpha_6 = -(1 - \sqrt{1 - e}) \eta_1. \end{aligned} \quad (11)$$

The relevant aspect of these equations is that they lead to a reduction of number of the viscosity coefficients; this is the main result of paper. The LCs' stress tensor would be completely characterized by three free parameters and not by five as usually it has been described. Moreover, the use of the Eq. (11) in the definition of the Miesowicz's coefficients, Eq. (4), allows to construct an expression combining the Miesowicz's coefficients, the

dissipative parameters and the eccentricity of the nematic grain:

$$\begin{aligned} \frac{m_3 m_3}{m_1 m_2} &= \frac{1}{1+e} - \frac{2}{(1+e)^2} \frac{\eta_3}{\eta_1} + \frac{1+e^2}{(1+e)^3} \frac{\eta_3^2}{\eta_1^2} + \dots \\ &= \frac{1}{1+e} - \frac{2}{\sqrt{1-e(1+e)^2}} \frac{\alpha_3}{\alpha_4} + \frac{3+e^2}{(1-e)(1+e)^3} \frac{\alpha_3^2}{\alpha_4^2} + \dots \end{aligned} \quad (12)$$

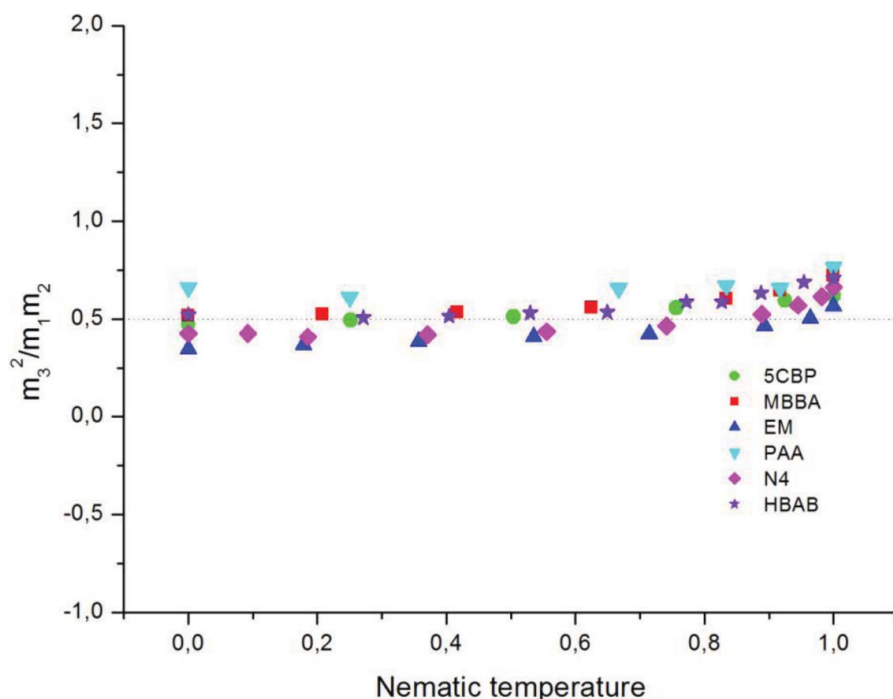
This result is a direct consequence of the use of the extended HB approach, where two rotational dissipative parameters has been added, and it shows that the Miesowicz's coefficients are connected not only by the eccentricity of the nematic grain but also by the parameters  $\eta_1$  and  $\eta_3$ . Furthermore, it describes an approximation for the case where  $\alpha_3$  can be disregarded. According to de Gennes [1],  $\alpha_3$  corresponds to 1.5% of the value of  $\alpha_4$ , the isotropic term, for all nematic calamitic compounds. Then, the ratio  $\alpha_3/\alpha_4$  or  $\eta_3/\eta_1$  can be neglected and  $m_3^2/m_1 m_2 \approx 1/(1+e)$ . This approximation can be easily tested experimentally. Recently, Simões and co-workers [32] showed that as the effective eccentricity of the nematic grains does not depend on their dimensions, but only on their ratios,  $m_3^2/m_1 m_2$  assume approximately the same values for all nematic compounds. Furthermore, they showed that save for the neighbourhoods of the NI phase transition where  $r_a \gg r_b$ ,  $e = 1 - (r_b/r_a)^2 \approx 1$ , implying that  $m_3^2/m_1 m_2 \approx 1/2$ . Otherwise, as the NI phase transition is approached the effective forms of the nematic grains lose their ellipsoidal shape, becoming more and more spherical, and giving rise to an increase in the ratio  $r_b/r_a$ , making  $e \rightarrow 0$ , and  $m_3^2/m_1 m_2 \rightarrow 1$ . Finally, Eq. (12) describes that the difference between isotropic and anisotropic liquids is not exclusively due to the geometry of their constituent grains.

### 3. Experimental Data

In order to verify whether the experimental results agree with Eq. (12), we have collected the experimental data from the literature [10–14]. The results are shown in Fig. 1, where a uniformised temperature scale was constructed [15–17, 41, 42] in such a way that for the nematic–crystalline transition the temperature  $T = 0$  was attributed, while for the NI transition the temperature  $T = 1$  was attributed. In the figure it can be observed that along the entire range of the nematic phase it is found that  $m_3^2/m_1 m_2 \approx 1/2$  and only at the neighbourhoods of the NI phase transition is an increase of the value observed.

### 4. Conclusions

In this paper, we have shown how the use of a modified version of the HB approach leads to a reduction of the number of the viscosity coefficients of the NLCs – only three free parameters are necessary: two viscosity parameters  $\eta_1$  and  $\eta_3$  and the quenched eccentricity  $e$  of the nematic grain. As a consequence of the application of the Onsager relation, the set of parameters  $\eta_1$ ,  $\eta_2$  and  $\eta_3$  that describes the dissipative channels are not independent, but connected by the eccentricity of the nematic grain, Eq. (10). Another astonishing result following from the Eq. (10) is that as the effective eccentricity becomes null when the NI phase-transition is approached, in this region the contribution each of the two rotational dissipative channels is non-null, but their sum is. Finally, we have found a meaning for the ratio  $m_3^2/(m_1 m_2)$  between the Miesowicz's coefficients, Eq. (12): it expresses the relative contribution of the two dissipative channels to the Miesowicz's coefficients.



**Figure 1.** The ratio  $m_3^2/m_1m_2$  versus the uniformized nematic temperature scale [15–17, 41, 42]. As the ratio  $\alpha_3/\alpha_4$  or  $\eta_3/\eta_1$  can be neglected, Eq. (12) assumes  $m_3^2/m_1m_2 \approx 1/2$ . The experimental data exhibited in this graph [10–14] completely confirm this prediction.

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