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A geometrical relation between the Miesowicz's coefficients

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In this work, it will be shown that the Miesowicz's viscosity coefficients of the nematic liquid crystals are not independent, but connected by the effective geometry that the nematic molecules/micelles acquire under random oscillatory vibration. This result follows from a straightforward application of the Onsager reciprocal relation to a recent proposed generalisation of the conformal transformation approach of the nematic rheology [1], where it had been assumed that the nematic dissipative process is not driven by a unique initial isotropic viscosity term, but by two; a rotational term has been added. As a consequence of the application of the Onsager relation, these two viscosity terms become related by the effective eccentricity of the nematic grain, leading to a connection between the Miesowicz's coefficients. Known experimental data of nematic compounds are used to test this prediction, resulting in its confirmation, which gives to the conformal transformation approach a predictive vigour that is not found in any other nematic viscosity theory.

Keywords: liquid crystals; rheology; nematic viscosity; nematic liquid crystals

1. Introduction

In a recent work [1], it has been shown that the Hess and Balls (HB) approach [2–6] to the nematic viscosity is an incomplete approach; it does not consider the dissipation through rotation of the nematic material. To repair this problem, a new term which takes care of the internal rotation of the anisotropic liquid, has been added to the HB approach. The main result obtained with that improvement is that the HB approach describes a new relation between the Miesowicz's coefficients. In this paper we will apply the Parodi relation to this improved version of the HB approach, and show that this new relation also reveals a connection between these viscosity coefficients and the geometry of the nematic cell.

2. Fundamentals

The anisotropic viscosity of liquid crystals (LCs) is one of the most challenging properties of these materials [7]. This property was discovered in 1935 by Miesowicz [8, 9], when he showed that LCs are non-Newtonian fluids exhibiting direction-dependent viscosity when submitted to an external field. Since this time, enormous amounts of experimental and theoretical effort have been devoted to the subject [2–30], but a satisfactory microscopic theory for it has never been found [21–23]. The kinetic approach of Doi had been for some time the most accepted microscopic theory of the nematic viscosity [24–29], but even having the great merit of producing an expression free of

adjustable parameters, which captures the essence of the phenomena, furnishing a semi-microscopic explanation to the origin of their anisotropy, it presents well-documented disagreements with the experimental data, being unable to describe the essential aspects of the phenomenology observed in these systems, especially when the entire range of the nematic phase is considered [21–23].

A simple way to understand the origin of the nematic anisotropies attributes the source of the anisotropies observed on a macroscopic scale to the microscopic shape of their constituent molecules; this idea is the essence of the passage from a microscopic to a macroscopic order parameter [7], being also the heart of the HB conformal transformation [2–6]. In the HB approach 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 idealised spherical molecules of an isotropic liquid could be deformed until they assumed the ellipsoidal form of an idealised nematic molecule, the macroscopic physical properties would be transformed into those observed in nematic liquid crystals (NLCs).

Hess and Balls and co-workers have proposed a way by which the viscosity of an anisotropic liquid can be written in terms of the viscosity of an isotropic liquid. We will use this model below (see Equation (4)). In order to understand how it works, suppose

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we have a vector \vec{A} on which acts the first of the transformations,

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

where \vec{n} is the nematic director field defined at each point. After the application of dx^i/ds^j over \vec{A} , we find

$$A'_i \rightarrow A_j dx^j/ds^i = r_a n_i (\vec{A} \cdot \vec{n}) + r_b (\vec{A}_i - n_i (\vec{A} \cdot \vec{n})), \quad (2)$$

which expands the components of \vec{A} parallel to \vec{n} by r_a , while the components perpendicular to \vec{n} are expanded by r_b . When applied to the vectors covering the surface of a sphere, this transformation would transform it into an ellipsoid. Of course, ds_i/dx_j is the inverse of dx^i/ds^j . In these equations, $\{r_a, r_b\}$ are dependent on the nematic order parameter S and characterise the thermalised shape of an ellipsoidal nematic grain [23, 31–34], r_a gives the uniaxial axis of the thermalised grain and r_b gives the degenerated axis, in such a way that the resulting effective eccentricity is given by

$$e = 1 - \frac{r_b^2}{r_a^2}. \quad (3)$$

As the nematic–isotropic (NI) phase transition is approached, we have that $r_a \rightarrow r_b$ and, accordingly, $e \rightarrow 0$. A study of how these parameters are effectively dependent on temperature (or order parameter S) can be found in [23]. In terms of the transformations given in Equation (1), HB has assumed that, as the operator dx^i/ds^j transforms spherically symmetric objects into ellipsoidal ones, the stress tensor of a liquid with ellipsoidal particles, given in Equation (7), should be obtained through the application of Equation (1) [23, 31–34] on the stress tensor of an isotropic liquid, $\sigma_{ij}^I = \eta \partial_i v_j$, namely,

$$\sigma_{ij} = \frac{ds^l}{dx^i} \frac{dx^j}{ds^k} (\eta \partial_l v^k), \quad (4)$$

where $\partial_i v_j$ is the gradient of the fluid velocity.

Notwithstanding its strong physical appeal and conceptual simplicity, the HB approach never explained completely the phenomenology observed in the LC viscosity. In a previous work [1] we have shown that Equation (4) does not take into account all of the 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,

$$\vec{v} = \vec{v}^c + \vec{w} \times \vec{\rho}, \quad (5)$$

the first considering the motion of its centre of mass \vec{v}^c , and the second considering the rotation of its internal points, at the position $\vec{\rho}$, around the centre of mass, where \vec{w} is the instantaneous angular velocity. Both of these terms produce dissipation and, as the second term is not present in Equation (4), the HB approach is incomplete.

We will show now that the absence of this term provokes another serious problem in this approach: it hinders the correct use of the Onsager theorem. Let us see why. According to the Ericksen, Leslie and Parodi (ELP) approach [10–15], the general expression for the dissipation occurring in a nematic sample is given by

$$T\dot{S} = \int d^3r \left\{ \sigma_{ij} A_{ij} + \vec{h} \cdot \vec{N} \right\}, \quad (6)$$

where $A_{ij} = (\partial_i v_j + \partial_j v_i)/2$ and $\vec{N} = (\vec{n} - (\vec{w} \times \vec{n}))$ are the shearing flow and the rotational flow, respectively, and σ_{ij} and h_i are the corresponding generalised forces. The Onsager theorem states that, in the linear regime, when the generalised forces σ_{ij} and h_i are proportional to the fluxes A_{ij} and \vec{N} ,

$$\begin{aligned} \sigma_{ij} &= L_{ijkl} A_{kl} + M_{ijk} N_k \\ &= \alpha_1 n_i n_j n_k n_l A_{kl} + \alpha_2 n_i N_k + \alpha_3 n_j N_k + \alpha_4 A_{ij} \\ &\quad + \alpha_5 n_i A_{jk} n_k + \alpha_6 n_j A_{ik} n_k, \\ h_i &= M_{ijk} A'_{jk} + P_{ij} N_j \\ &= (\alpha_2 + \alpha_3) n_k A_{ik} + (\alpha_3 - \alpha_2) N_i, \end{aligned} \quad (7)$$

the relation [7] $M'_{ijk} = M_{ijk}$, which is the same as $\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$, would be obeyed. That is, there is a relation $M'_{ijk} = M_{ijk}$ connecting the shearing flow and rotational flow terms of the two generalised forces, σ_{ij} and h_i . As Equation (4) does not contain this rotational term, the Onsager relation cannot be correctly applied to it and, consequently, it cannot be in accordance with the fundamentals of the nematic dissipative theory.

We will see now that the main result of this paper arises from the solution of this problem; the application of the Onsager theorem to this improved version of the HB approach will lead to an up to now unsuspected geometrical connection between Miesowicz's coefficients. Namely, let us assume that the stress tensor is given by a conformal transformation with the form

$$\sigma_{ij} = 2 \frac{ds^l}{dx^i} \frac{dx^j}{ds^k} (\eta_1 A_{ij} + \eta_2 N_i n_j), \quad (8)$$

where the first term, $\eta_1 A_{ij}$, reproduces the symmetric part of the former form of the HB approach, and the

second term, $\eta_2 N_i n_j$, extends the initial HB hypotheses, considering the rotational dissipative process. η_1 and η_2 are the viscosities associated with each of these dissipative terms, respectively.

Using Equation (1), and equating the result with the stress tensor σ_{ij} given in Equation (7), we arrive at

$$\alpha_1 = \frac{(r_a - r_b)^2}{r_a r_b} \eta_1, \quad \alpha_2 = \frac{r_a}{r_b} \eta_2, \quad \alpha_3 = 0,$$

$$\alpha_4 = \eta_1, \quad \alpha_5 = \frac{(r_a - r_b)}{r_b} \eta_1, \quad \alpha_6 = \frac{(r_b - r_a)}{r_a} \eta_1. \quad (9)$$

Furthermore, using the definition of Miesowicz's coefficients [7], $m_1 = (\alpha_4 + \alpha_5 - \alpha_2)/2$, $m_2 = (\alpha_3 + \alpha_4 + \alpha_6)/2$, $m_3 = \alpha_4/2$, in these equations, we arrive at

$$m_1 = \frac{r_a}{2r_b} (\eta_1 - \eta_2),$$

$$m_2 = \frac{r_b}{2r_a} \eta_1,$$

$$m_3 = \frac{\eta_1}{2}, \quad (10)$$

which leads to

$$\frac{m_3}{m_1} \frac{m_3}{m_2} = \frac{\eta_1}{\eta_1 - \eta_2}. \quad (11)$$

The relevant aspect of this equation is that it only depends on the coupling parameters appearing in Equation (8). Furthermore, it is straightforward to see that the set of parameters η_1 and η_2 are not independent; the application of the Parodi relation, $\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$, in the set of equations given in Equation (9) gives

$$\eta_2 = -e\eta_1. \quad (12)$$

where e has been defined in Equation (3). This leads to

$$\frac{m_3}{m_1} \frac{m_3}{m_2} = \frac{1}{1 + e}. \quad (13)$$

This result establishes a connection between the values of the nematic viscosity coefficients and the eccentricity of the nematic grain. It has not been predicted by any other nematic rheological model and appeared here because we have extended the HB approach to include a rotational term, which becomes connected with the eccentricity of the nematic grain through the application of the Onsager theorem. An amazing aspect of this equation is that it can be

easily tested experimentally; as the effective eccentricity of the nematic grains does not depend on their dimensions, but only on their ratios, we expect that $m_3^2/(m_1 m_2)$ would assume approximately the same values for all nematic compounds. Furthermore, save for the neighbourhoods of the NI phase transition, we would have $r_a \gg r_b$, so we can see that $e = 1 - (r_b/r_a)^2 \approx 1$, implying that such a value would be found around $m_3^2/(m_1 m_2) \approx 1/2$. 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 $m_3^2/(m_1 m_2) \rightarrow 1$.

3. Experimental data

In order to verify whether the experimental results agree with the relation deduced above, we have collected the experimental data from the literature [16–20] and, in order to have a broad scope of such relations, only those data encompassing the entire range of the nematic phase have been considered. For the compounds listed in Figure 1, a uniformised temperature scale was constructed [21–23, 35, 36] 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 the agreement between the predictions of the theory and the experimental results are excellent. 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 of r observed. Surely, in this region it does not arrive at $m_3^2/(m_1 m_2) \approx 1$ because the NI phase transition is discontinuous.

4. Conclusion

We have proposed in this paper the introduction of two 'isotropic' viscosity terms, η_1 and η_2 , in the HB stress tensor given by Equation (8): one corresponding to the dissipation occurring in the shearing term, and the other corresponding to the dissipation occurring in the rotational term. This procedure seems to be a contradiction; there is in fact only one isotropic viscosity term. Nevertheless, the application of the Onsager theorem restores the order; it links these dissipative channels revealing that they are not independent, but connected by the eccentricity of the nematic grain, Equation (12) – in the end only one viscosity term survives. Furthermore, as the effective eccentricity becomes null when the NI phase transition is approached, Equation (12) shows that in this region

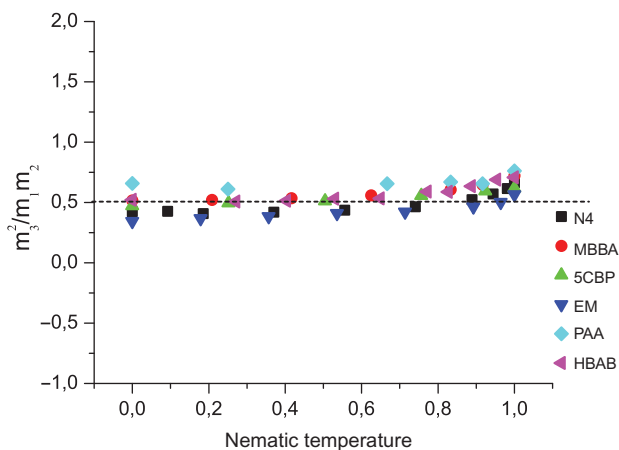


Figure 1. Parameter $r = m_3^2/(m_1m_2)$ versus the uniformised nematic temperature scale [21–23, 35, 36]. The use of the Onsager relation in the stress tensor obtained from the extended HB approach, proposed in [1], leads to the prediction that r would assume values around $r \approx 1/2$. Furthermore, as the NI phase transition is approached the effective anisotropy of the nematic grain is diminished and the value of r would increase, approaching the value $r = 1$. The experimental data exhibited in this graph [16–20] completely confirm these predictions. Although the compounds can be easily recognised by their abbreviated names in the figure, their scientific names and the authors of the measurements can be found in the quoted references (colour version online).

the dissipation through this channel disappears, attesting to the confidence of the approach that we have proposed. Finally, the experimental confirmation of the relation given in Equation (13) exposes the heart of the HB approach, which affirms that the difference between isotropic and anisotropic liquids remains in the geometry of their constituent grains. As the relation that we have found explicitly establishes a geometrical connection between the nematic viscosity coefficients, it gives to the HB approach a new perspective in the challenge to understand the physics behind the nematic viscosity.

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