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Geometrical content of Leslie coefficients

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In this work, we will study how the effective geometry acquired by nematic molecules under thermal vibration contribute to the determination of the Leslie coefficients. To do this, we will divide this work in two sections. In the first section, we present the geometrical fundamentals of the so-called Hess-Baalss (HB) approach [D. Baalss and S. Hess, Phys. Rev. Lett. **57**, 86 (1986)] where we show that its basic assumptions can be understood as a geometrical interpretation of de Gennes' passage from the microscopic to the macroscopic order parameter. In the second section, we use an extended version of the HB approach [M. Simes, K. Yamaguti, and A. J. Palangana, Phys. Rev. E **80**, 061701 (2009)] to obtain the geometrical contribution to each Leslie coefficient. Our results will be compared with experimental data, and we will show that the Miesowicz's coefficients are connected as long as the ratio α_3/α_4 between these Leslie coefficients can be considered small.

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I. INTRODUCTION

The anisotropic viscosity of liquid crystals (LCs) is one of the most challenging properties of these materials [1]. It was discovered in 1935 by Miesowicz [2,3] when he showed that LCs exhibit a direction-dependent viscosity when submitted to an external field. A large amount of experimental and theoretical investigation has been devoted to this subject [4–30] but a satisfactory microscopic theory for it has not been found [15–17]. A widespread approach to the continuum mechanics of the properties of nematic liquid crystals (NLCs) is the Ericksen-Leslie-Parodi (ELP) approach [4–9]. The ELP approach establishes that the nematic viscosity is determined by six viscosity coefficients [Eq. (48)] that are connected by the Onsager-Parodi relationship that shows that five of them are effectively independent. There is also the hydrodynamic theory of Martin, Parodi, and Pershan [8,9] which, according de Gennes [1], is effectively equivalent to the ELP approach. The kinetic approach of Doi is the most accepted microscopic theory of nematic viscosity [18–23]. It produces expressions free of adjustable parameters that capture the essence of the phenomena, furnishing a semimicroscopic explanation to the origin of their anisotropy. Nevertheless, it also presents well-documented disagreements with experimental data. It is unable to describe, for example, the regularities observed in the viscosity data, mainly as the nematic crystalline regions are approached [15–17].

Recently, through the so-called Hess-Baalss (HB) approach, which is defined in Refs. [24–28], 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]. It must be clear that this approach does not intend to propose that nematic viscosity is a geometrical problem; it is definitively a dissipative many-body problem whose solution needs to be in accord with the fundamentals of dissipative statistical mechanics. Nevertheless, this circumstance does not forbid the study of the role played by the geometry of the nematic molecule in this dissipative problem. To capture the essence of this geometrical contribution is the purpose of the HB approach and the aim of this work. The HB approach assumes that if one could imagine a way by which nematic molecules could be continuously deformed up to the point at which they become spherical, it would be possible to observe a corresponding reduction of the macroscopic anisotropies until they vanish. Conversely, 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 similarly to those observed on the NLCs. Nevertheless, the mathematical formulation of the task proposed by this approach is not yet fully developed. A consistent determination of all five Leslie coefficients in terms of this geometrical transformation has not been done. The purpose of this work is to use the HB approach to determine the geometrical contribution to the viscosity coefficients of the nematic phase. We will also give an exposition of the fundamentals of such nematic geometry. In fact, a complete exposition of this approach is not found in a single work. Frequently, some important relations only are found when specific results of a given work are correlated with results contained in other works. In this paper we will give a detailed exposition of the geometrical content of the HB approach and use it to study the Onsager-Parodi relations [6], showing that under the context of the HB approach these relations acquire a geometrical interpretation that leads to a connection with Miesowicz's coefficients [31]. Without a clear exposition of the exact meaning of this temperature-dependent nematic geometry, the importance of this connection may not be fully understood. The aim of this work is also to give a detailed and quasi-self-contained exposition of the significance of such results.

II. THERMAL GEOMETRY AND THE NEMATIC GRAIN

A. The geometry of the nematic molecule

A simple way to give a geometrical basis to the anisotropies observed in nematic materials is to attribute to the microscopic anisotropies of their constituent molecules the source of the anisotropies observed on a macroscopic scale. This idea constitutes de Gennes' passage from a microscopic to a macroscopic order parameter [1] [see Eq. (3) below] and is

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also the heart of the HB approach. We will begin this study by establishing a connection between these two approaches, showing how the de Gennes formulation for the passage between the microscopic and macroscopic order parameter has a straightforward geometrical interpretation, and is the appropriate tool to provide a formal foundation for the HB approach to the nematic viscosity.

Let us consider some fundamentals of the order-parameter concept. These ideas are well know [1], but as they are essential to understand the geometrical approach that we will take to the nematic LC theory, we will review them here. So, consider that

$$\hat{Q}_{ij}(\overrightarrow{\boldsymbol{n}}) = -\tfrac{1}{3}\delta_{ij} + \hat{n}_i\hat{n}_j \tag{1}$$

describes the microscopic anisotropy of some physical quantity. It is the microscopic order parameter [1], where $\hat{\mathbf{n}}$ is the corresponding rigid axis of the molecular-micellar symmetry, while

$$Q_{ij}(\vec{n}) = -\tfrac{1}{3}\delta_{ij} + n_i n_j \tag{2}$$

is the corresponding macroscopic order parameter, which describes macroscopic anisotropies, where $\vec{n}$ is the associated director [1]. From now on, variables with a hat, such as $\overrightarrow{n}$, are microscopic variables. When a variable appears without a hat it is a macroscopic variable, such as $\vec{n}$, which is the director. The connection between the microscopic order parameter $\hat{Q}_{ij}(\overrightarrow{n})$ and the macroscopic order parameter $Q_{ij}(\vec{n})$ is made by assuming that the microscopic rigid axis of a nematic molecule $\overrightarrow{n}$ is a random variable that oscillates so fast that when $\hat{Q}_{ij}(\overrightarrow{n})$ is averaged, on the time and/or in the neighborhood of a point, that average determines the macroscopic order parameter $Q_{ij}(\vec{n})$. In mathematical terms, we would have [1]

$$\langle\hat{Q}_{ij}(\overrightarrow{n})\rangle = S\,Q_{ij}(\vec{n}), \tag{3}$$

where $\langle x\rangle$ stands for the statistical average of the random variable x. Notice, as a consequence of the ergodic hypothesis [39], that the statistical average of this expression considers spatial and temporal mean over the aggregate of molecules-micelles around a given point. Consequently, when the microscopic order parameter is averaged to obtain a macroscopic one, a non-null value of S implies the existence of a macroscopic molecular ordering that defines a non-null $\vec{n}$ in the neighborhood of every point. Below we will give a geometrical interpretation for this local macroscopic ordering that we will refer to as nematic grain. To obtain an expression for S in terms of $\overrightarrow{n}$ and $\vec{n}$ it is enough to multiply the above expression by the macroscopic order parameter $Q_{ji}(n)$ and take the trace of the resulting expression

$$Q_{ji}(n)\langle\hat{Q}_{ij}(\hat{n})\rangle = S\,Q_{ji}(n)Q_{ij}(n), \tag{4}$$

to obtain

$$S = \tfrac{3}{2}\big[-\tfrac{1}{3} + \langle(\overrightarrow{n}\cdot\vec{n})^2\rangle\big]. \tag{5}$$

In Eq. (4) we have used the usual repeated indices sum rule. From now on, repeated indices will be subject to it.

To see how these basic ideas about the meaning of the order parameter can acquire a geometrical interpretation, let us consider two analytic expressions, the first for a sphere $\hat{S}$, and the second for an ellipsoid $\hat{E}$. The equation of a sphere $\hat{S}$ with radius $\overrightarrow{r}$ is given by

$$\hat{S}_{ij}x_ix_j = 1, \quad S_{ij} = \frac{\delta_{ij}}{\hat{r}^2}, \quad i = \{1,2,3\}. \tag{6}$$

Otherwise, if $\hat{r}_1$, $\hat{r}_2$, and $\hat{r}_3$ are the dimensions of an ellipsoid $\hat{E}$ along the orthonormal directions $\vec{p}$, $\vec{q}$, and $\vec{r}$ respectively, the equation of $\hat{E}$ is be given by

$$\hat{E}_{ij}x_ix_j = 1, \quad \hat{E}_{ij} = \frac{\hat{p}_i\hat{p}_j}{\hat{r}_1^2} + \frac{\hat{q}_i\hat{q}_j}{\hat{r}_2^2} + \frac{\hat{r}_i\hat{r}_j}{\hat{r}_3^2}, \quad i = \{1,2,3\}. \tag{7}$$

Furthermore, as $\vec{p}$, $\vec{q}$, and $\vec{r}$ form a local orthonormal basis, any vector $\vec{x}$ can be written in terms of them. In components, $\vec{x}$ can be written as $x_i = p_i(p_jx_j) + q_i(q_jx_j) + r_i(r_jx_j) = (\hat{p}_i\hat{p}_j + \hat{q}_i\hat{q}_j + \hat{r}_i\hat{r}_j)x_j$, from which follows the completeness relation follows:

$$\hat{p}_i\hat{p}_j + \hat{q}_i\hat{q}_j + \hat{r}_i\hat{r}_j = \delta_{ij}. \tag{8}$$

Using this equation in Eq. (7) we arrive at

$$\hat{E}_{ij} = \frac{\delta_{ij}}{\hat{r}_3^2} + \left(\frac{1}{\hat{r}_1^2} - \frac{1}{\hat{r}_3^2}\right)\hat{p}_i\hat{p}_j + \left(\frac{1}{\hat{r}_2^2} - \frac{1}{\hat{r}_3^2}\right)\hat{q}_i\hat{q}_j. \tag{9}$$

At this point we will use the idea of the order parameter to continue the development of our geometrical approach. To do that, observe that the ellipsoid described above has only two axes of symmetry, $\overrightarrow{p}$ and $\overrightarrow{q}$, and not three. Due to the completeness relation in Eq. (8) only two axes are necessary. Consequently, we will need two microscopic order parameters, one for each symmetrical axis,

$$\hat{Q}_{ij}(\overrightarrow{p}) = -\tfrac{1}{3}\delta_{ij} + \hat{p}_i\hat{p}_j, \quad \hat{Q}_{ij}(\overrightarrow{q}) = -\tfrac{1}{3}\delta_{ij} + \hat{q}_i\hat{q}_j. \tag{10}$$

Using the definitions of microscopic order parameter in Eq. (9) it is found that

$$\hat{E}_{ij} = \frac{1}{3}\left(\frac{1}{\hat{r}_1^2} + \frac{1}{\hat{r}_2^2} + \frac{1}{\hat{r}_3^2}\right)\delta_{ij} + \left(\frac{1}{\hat{r}_1^2} - \frac{1}{\hat{r}_3^2}\right)\hat{Q}_{ij}(\overrightarrow{p}) + \left(\frac{1}{\hat{r}_2^2} - \frac{1}{\hat{r}_3^2}\right)\hat{Q}_{ij}(\overrightarrow{q}), \tag{11}$$

which shows that the microscopic ellipsoidal matrix $\hat{E}_{ij}$ can be written as a sum of two kinds of terms. The first is proportional to δ_{ij}, which, according Eq. (6), represents a sphere with a radius $\hat{r}_s$ given by

$$\frac{1}{\hat{r}_S^2} = \frac{1}{3}\left(\frac{1}{\hat{r}_1^2} + \frac{1}{\hat{r}_2^2} + \frac{1}{\hat{r}_3^2}\right). \tag{12}$$

The second is proportional to two microscopic order parameters, $\hat{Q}_{ij}(\overrightarrow{p})$ and $\hat{Q}_{ij}(\overrightarrow{q})$. To understand our further developments, observe that in Eq. (11) the order parameter only expresses the direction of the microscopic deformation. The amount of the deformation is given by the expressions $(1/\hat{r}_1^2 - 1/\hat{r}_3^2)$ and $(1/\hat{r}_2^2 - 1/\hat{r}_3^2)$ in front of each order parameter. In the next section we will take the thermal average of Eq. (11) and the terms describing this amount of deformation will become temperature dependent.

B. The thermalized molecule: the nematic grain

Let us assume that we have an ensemble of ellipsoidal molecules forming a liquid crystal. Because, according Eq. (11), the microscopic deviation from the spherical geometry of each of these molecules is proportional to the microscopic order parameters, we can use the statistical media of each term of Eq. (10) that appears in Eq. (11) to find the mean value of $\hat{E}_{ij}$ on this ensemble. That is,

$$\begin{aligned}\langle \hat{E}_{ij}\rangle &= \frac{1}{3}\left(\frac{1}{\hat{r}_1^2}+\frac{1}{\hat{r}_2^2}+\frac{1}{\hat{r}_3^2}\right)\delta_{ij}+\left(\frac{1}{\hat{r}_1^2}-\frac{1}{\hat{r}_3^2}\right)\langle \hat{Q}_{ij}(\overrightarrow{p}\rangle\\ &\quad +\left(\frac{1}{\hat{r}_2^2}-\frac{1}{\hat{r}_3^2}\right)\langle \hat{Q}_{ij}(\overrightarrow{q})\rangle\\ &= \frac{1}{3}\left(\frac{1}{\hat{r}_1^2}+\frac{1}{\hat{r}_2^2}+\frac{1}{\hat{r}_3^2}\right)\delta_{ij}+\left(\frac{1}{\hat{r}_1^2}-\frac{1}{\hat{r}_3^2}\right)S^pQ_{ij}(p)\\ &\quad +\left(\frac{1}{\hat{r}_2^2}-\frac{1}{\hat{r}_3^2}\right)S^qQ_{ij}(q),\end{aligned} \tag{13}$$

where we have generalized Eq. (3) along the directions $\overrightarrow{p}$ and $\overrightarrow{q}$, assuming that $\langle \hat{Q}_{ij}(\overrightarrow{p})\rangle = S^p\ Q_{ij}(\vec{p})$ and $\langle \hat{Q}_{ij}(\overrightarrow{q})\rangle = S^q\ Q_{ij}(\vec{q})$, where S^p and S^q are the two scalar order parameters describing the thermal vibrations of the two independent director directions, $\overrightarrow{p}$ and $\overrightarrow{q}$ respectively. We have also assumed that the microscopic dimensions of the grain $\hat{r}_1$, $\hat{r}_2$, and $\hat{r}_3$ do not change with temperature. Now, for each of these order parameters we can use the macroscopic definition of order parameter,

$$Q_{ij}(\vec{p}) = \left(-\tfrac{1}{3}\delta_{ij}+p_ip_j\right),\quad Q_{ij}(\vec{q}) = \left(-\tfrac{1}{3}\delta_{ij}+q_iq_j\right), \tag{14}$$

in Eq. (13), to obtain

$$\begin{aligned}\langle \hat{E}_{ij}\rangle = &\left(\frac{1}{3\hat{r}_1^2}(2S^p+1)+\frac{1}{3\hat{r}_2^2}(1-S^q)\right.\\ &\left.+\frac{1}{3\hat{r}_3^2}(1-2S^p+S^q)\right)p_ip_j+\left(\frac{1}{3\hat{r}_1^2}(1-S^p)\right.\\ &\left.+\frac{1}{3\hat{r}_2^2}(2S^q+1)+\frac{1}{3\hat{r}_3^2}(1-2S^q+S^p)\right)q_iq_j\\ &+\left(\frac{1}{3\hat{r}_1^2}(1-S^p)+\frac{1}{3\hat{r}_2^2}(1-S^q)\right.\\ &\left.+\frac{1}{3\hat{r}_3^2}(1+S^p+S^q)\right)r_ir_j.\end{aligned} \tag{15}$$

We have also used the macroscopic version of Eq. (8). Finally, by comparing this expression with Eq. (7) we see that this equation describes a macroscopic ellipsoid

$$\langle \hat{E}_{ij}\rangle = \frac{1}{r_1^2}p_ip_j+\frac{1}{r_2^2}q_iq_j+\frac{1}{r_3^2}r_ir_j \tag{16}$$

with dimensions

$$\begin{aligned}\frac{1}{r_1^2} &= \frac{1}{3\hat{r}_1^2}(2S^p+1)+\frac{1}{3\hat{r}_2^2}(1-S^q)+\frac{1}{3\hat{r}_3^2}(1-2S^p+S^q),\\ \frac{1}{r_2^2} &= \frac{1}{3\hat{r}_1^2}(1-S^p)+\frac{1}{3\hat{r}_2^2}(2S^q+1)+\frac{1}{3\hat{r}_3^2}(1-2S^q+S^p),\\ \frac{1}{r_3^2} &= \frac{1}{3\hat{r}_1^2}(1-S^p)+\frac{1}{3\hat{r}_2^2}(1-S^q)+\frac{1}{3\hat{r}_3^2}(1+S^p+S^q).\end{aligned} \tag{17}$$

This result shows that after the thermal average defined at Eq. (3) an ensemble of ellipsoidal nematic molecules acquire a macroscopic thermalized geometry that is also ellipsoidal. Their dimensions depend on the order parameters and, of course, on the temperature: this is the nematic grain. For example, at the isotropic phase we have that $S^m = S^n = 0$ and

$$\frac{1}{r_1^2} = \frac{1}{r_2^2} = \frac{1}{r_3^2} = \frac{1}{3}\left(\frac{1}{\hat{r}_1^2}+\frac{1}{\hat{r}_2^2}+\frac{1}{\hat{r}_3^2}\right), \tag{18}$$

which shows that at isotropic phase the nematic grain assumes the effective form of a sphere. Otherwise, at low temperatures, where $S^m = S^n = 1$, we have that

$$\frac{1}{r_1^2} = \frac{1}{\hat{r}_1^2},\quad \frac{1}{r_2^2} = \frac{1}{\hat{r}_2^2},\quad \frac{1}{r_3^2} = \frac{1}{\hat{r}_3^2}, \tag{19}$$

showing that at very low temperatures it is ellipsoidal.

In synthesis, these results show that the definition of tensorial order parameters [Eqs. (1)–(5)] realizes the passage from the microscopic ellipsoid-shaped nematic molecule to a thermalized macroscopic object, the nematic grain that is also ellipsoidal. These facts allow us to name as thermal geometry the relation contained in Eq. (17). Please note that this is a direct consequence of the definition of microscopic and macroscopic order parameters, nothing more.

For future use, let us specialize the above expressions for the uniaxial case, which is the situation found more frequently. For this case we have, $\hat{r}_2^2 = \hat{r}_3^2$ and $S^q = 0$, which gives

$$\begin{aligned}\frac{1}{r_1^2} &= \frac{1}{3}\left(\frac{1}{\hat{r}_1^2}(2S+1)+\frac{2}{\hat{r}_2^2}(1-S)\right),\\ \frac{1}{r_2^2} &= \frac{1}{r_3^2} = \frac{1}{3}\left(\frac{1}{\hat{r}_1^2}(1-S)+\frac{1}{\hat{r}_2^2}(2+S)\right),\end{aligned} \tag{20}$$

where we have made $S \equiv S^p$, to be in accord with the usual notation in which the uniaxial scalar order parameter is named as S. This result shows that a thermalized microscopic uniaxial molecule produces a nematic grain that is also uniaxial, $r_2^2 = r_3^2$. Therefore, similarly to Eq. (9),

$$\begin{aligned}\langle E_{ij}\rangle &= \frac{1}{r_2^2}\delta_{ij}+\left(\frac{1}{r_1^2}-\frac{1}{r_2^2}\right)p_ip_j\\ &= \frac{1}{3}\left(\frac{1}{r_1^2}+\frac{2}{r_2^2}\right)\delta_{ij}+\left(\frac{1}{r_1^2}-\frac{1}{r_2^2}\right)Q_{ij}(\vec{p}).\end{aligned} \tag{21}$$

As we have described above, the thermal ellipsoidal shape depends not only on the nematic order parameter $Q_{ij}(\vec{p})$, but also on the thermalized geometry of the nematic grain $1/r_1^2 1 - 1/r_2^2$. If we write this expression in the form

$$\langle E_{ij}\rangle = \frac{1}{r_2^2}\left[\frac{1}{3}\left(2+\frac{r_2^2}{r_1^2}\right)\delta_{ij}-\left(1-\frac{r_2^2}{r_1^2}\right)Q_{ij}(\vec{p})\right], \tag{22}$$

we recognize the term

$$e = 1-\frac{r_2^2}{r_1^2} \tag{23}$$

as the eccentricity of a uniaxial ellipsoid. Nevertheless, as r_1 and r_2 are temperature dependent, this eccentricity is the thermalized eccentricity of the nematic grain, which would be distinguished form the microscopic eccentricity $\hat{e}$,

$$\hat{e} = 1 - \frac{\hat{r}_2^2}{\hat{r}_1^2}, \tag{24}$$

which, of course, does not depend on the temperature. A straightforward use of Eq. (20) shows that

$$e = \frac{3\, S\, \hat{e}}{3 - \hat{e}(S-1)}. \tag{25}$$

From this equation we see that as the isotropic phase is approached we have that $S \to 0$ giving $e \to 0$. Otherwise, as the temperature is reduced, $S \to 1$, we find that $e \to \hat{e}$, which reproduces the microscopic value of the eccentricity. In terms of e, Eq. (22) becomes

$$\langle E_{ij} \rangle = \frac{1}{[3 - \hat{e}(S-1)]r_2^2}\{[3 + \hat{e}(1-2S)]\delta_{ij} - 3\, S\, \hat{e} Q_{ij}(\vec{p})\}. \tag{26}$$

Let us finish this section by making an observation about the values that are hoped for $\hat{e}$, the microscopic eccentricity of the nematic molecule. Suppose, for example, a typical calamitic nematic molecule where its length is five times its width. For this geometry Eq. (24) gives that $\hat{e} = 24/25 \approx 0.96$, which is very close to 1. Since for any sound value of this ratio we would always find $\hat{e} \approx 1$, for practical applications of this theory we can assume that, for a calamitic nematic molecule, $\hat{e} = 1$. Under this approximation, the macroscopic eccentricity e would be completely determined by the scalar order parameter

$$e = \frac{3\, S}{4 - S}. \tag{27}$$

Likewise, under these conditions, we have

$$\langle E_{ij} \rangle = \frac{1}{(4-S)r_2^2}[2(2-S)\delta_{ij} - 3\, S\, Q_{ij}(\vec{p})]. \tag{28}$$

C. Conformal transformation

In the preceding, we have shown that the thermal geometry of a nematic grain can be seen as a result of the sum of two types of terms: a spherical term and the deformations from this spherical shape that are proportional to the order parameters. This separation of $\langle E_{ij} \rangle$ into two parts contains the essence of the HB hypothesis [24–28]. We can interpret this separation in terms of the HB hypothesis by stating that the spherical part of the decomposition of $\langle E_{ij} \rangle$ would correspond to an isotropic fluid, while the addition of the remaining terms deforms it, making what could be described as a LC. Accordingly, a NLC can be conceived as a normal liquid in which its spherical molecules have been deformed to attain an ellipsoidal form. In this section, we will look for the microscopic and macroscopic versions of these transformations. Later, we will show how these transformations work.

Consider that at a point of our sample we have a vector $\vec{s}$ that is submitted to an elongation $\tilde{r}_1$, $\tilde{r}_2$, and $\tilde{r}_3$ of all its components in the directions $\vec{p}$, $\vec{q}$, and $\vec{r}$, respectively. Analytically, this elongation will be

$$\vec{x} = \tilde{r}_1 \vec{p}(\vec{p} \cdot \vec{s}) + \tilde{r}_2 \vec{q}(\vec{q} \cdot \vec{s}) + \tilde{r}_3 \vec{r}(\vec{r} \cdot \vec{s}), \tag{29}$$

which can be put in the form

$$\begin{aligned} \hat{x}_i &= \tilde{r}_1 \hat{p}_i(\hat{p}_j \hat{s}_j) + \tilde{r}_2 \hat{q}_i(\hat{q}_j \hat{s}_j) + \tilde{r}_3 \hat{r}_i(\hat{r}_j \hat{s}_j) \\ &= (\tilde{r}_1 \hat{p}_i \hat{p}_j + \tilde{r}_2 \hat{q}_i \hat{q}_j + \tilde{r}_3 \hat{r}_i \hat{r}_j)\hat{s}_j. \end{aligned} \tag{30}$$

Therefore,

$$\frac{\partial \hat{x}_i}{\partial \hat{s}_j} = \tilde{r}_1 \hat{p}_i \hat{p}_j + \tilde{r}_2 \hat{q}_i \hat{q}_j + \tilde{r}_3 \hat{r}_i \hat{r}_j. \tag{31}$$

Using the completeness relation in Eq. (8), this becomes

$$\frac{\partial \hat{x}_i}{\partial \hat{s}_j} = \tilde{r}_3 \delta_{ij} + (\tilde{r}_1 - \tilde{r}_3)\hat{p}_i \hat{p}_j + (\tilde{r}_2 - \tilde{r}_3)\hat{p}_i \hat{p}_j. \tag{32}$$

Furthermore, using the definition of the tensorial order parameter in Eq. (10), we obtain

$$\begin{aligned} \frac{\partial \hat{x}_i}{\partial \hat{s}_j} &= \frac{1}{3}(\tilde{r}_1 + \tilde{r}_2 + \tilde{r}_3)\delta_{ij} + (\tilde{r}_1 - \tilde{r}_3)\hat{Q}_{ij}(\vec{p}) \\ &\quad + (\tilde{r}_2 - \tilde{r}_3)\hat{Q}_{ij}(\vec{q}). \end{aligned} \tag{33}$$

A straightforward calculation shows that the inverse of this equation is given by

$$\begin{aligned} \frac{\partial \hat{s}_j}{\partial \hat{x}_i} &= \frac{1}{3}\left(\frac{1}{\tilde{r}_1} + \frac{1}{\tilde{r}_2} + \frac{1}{\tilde{r}_3}\right)\delta_{ij} + \left(\frac{1}{\tilde{r}_1} - \frac{1}{\tilde{r}_3}\right)\hat{Q}_{ij}(\vec{p}) \\ &\quad + \left(\frac{1}{\tilde{r}_2} - \frac{1}{\tilde{r}_3}\right)\hat{Q}_{ij}(\vec{q}). \end{aligned} \tag{34}$$

In order to understand how the HB approach acts, let us suppose that we have a molecule of an isotropic fluid that, hypothetically, has a spherical symmetry. Under this condition the equipotential surfaces generated by this molecule would also have a spherical form, as given by Eq. (6). Suppose we assume that this spherical surface has been obtained from the deformation,

$$\hat{s}_i = \frac{\partial \hat{s}_i}{\partial \hat{x}_j}\hat{x}_j, \tag{35}$$

of a ellipsoidal surface, as given by Eq. (7) where we have used the notation $\hat{s}_i$ to assign the coordinates of the spherical surface and $\hat{x}_i$ to assign coordinates of the ellipsoidal surface. Therefore, we can start from Eq. (6) and write the sequence of operations

$$\begin{aligned} \hat{S}_{ij}\hat{s}_i\hat{s}_j &= 1 \to \hat{S}_{ij}\left(\frac{\partial \hat{s}_i}{\partial \hat{x}_k}\hat{x}_k\right)\left(\frac{\partial \hat{s}_j}{\partial \hat{x}_l}\hat{x}_l\right) \\ &= 1 \to \hat{S}_{ij}\frac{\partial \hat{s}_i}{\partial \hat{x}_k}\frac{\partial \hat{s}_j}{\partial \hat{x}_l}\hat{x}_k\hat{x}_l = 1 \to O_{kl}\hat{x}_k\hat{x}_l = 1, \end{aligned} \tag{36}$$

where the operator

$$O_{kl} = \hat{S}_{ij}\frac{\partial \hat{s}_i}{\partial \hat{x}_k}\frac{\partial \hat{s}_j}{\partial \hat{x}_l} \tag{37}$$

expresses the result of the action of the HB transformation over the sphere. Using Eq. (34), a straightforward calculation shows that

$$O_{kl} = S_{ij}\frac{\partial \hat{s}_i}{\partial \hat{x}_k}\frac{\partial \hat{s}_j}{\partial \hat{x}_l} = \frac{1}{3}\left(\frac{1}{(\hat{r}\tilde{r}_1)^2}+\frac{1}{(\hat{r}\tilde{r}_2)^2}+\frac{1}{(\hat{r}\tilde{r}_3)^2}\right)\delta_{ij} + \left(\frac{1}{(\hat{r}\tilde{r}_1)^2}-\frac{1}{(\hat{r}\tilde{r}_3)^2}\right)\hat{Q}_{ij}(\overrightarrow{p}) + \left(\frac{1}{(\hat{r}\tilde{r}_2)^2}-\frac{1}{(\hat{r}\tilde{r}_3)^2}\right) \times \hat{Q}_{ij}(\overrightarrow{q}). \tag{38}$$

When we compare this equation with the equation describing an ellipsoid [Eq. (11)], we see that the quantities $\tilde{r}_1$, $\tilde{r}_2$, and $\tilde{r}_3$ actually act as parameters of deformation. The original sphere of radius $\hat{r}$ has been deformed to an ellipsoid, according to the rules $\hat{r} \rightarrow \hat{r}\tilde{r}_1$, along the direction $\vec{p}$; $\hat{r} \rightarrow \hat{r}\tilde{r}_2$, along the direction $\vec{q}$; and $\hat{r} \rightarrow \hat{r}\tilde{r}_3$, along the direction $\vec{r}$.

If we identify these transformations with the microscopic ellipsoidal parameters defined at Eq. (11), $\hat{r}\tilde{r}_1 \equiv \hat{r}_1$, $\hat{r}\tilde{r}_2 \equiv \hat{r}_2$, $\hat{r}\tilde{r}_3 \equiv \hat{r}_3$, where $\hat{r}_1$, $\hat{r}_2$, and $\hat{r}_3$ are the microscopic ellipsoidal dimensions defined in Eq. (7), we find that the deformations are given by

$$\tilde{r}_1 = \frac{\hat{r}_1}{\hat{r}}, \quad \tilde{r}_2 = \frac{\hat{r}_2}{\hat{r}}, \quad \tilde{r}_3 = \frac{\hat{r}_3}{\hat{r}}. \tag{39}$$

This is a satisfying result. The transformation defined in Eqs. (33) and (34) can be taken as the transformation that realizes the aim of the HB approach: to find an operator that transforms spheres with radius r into ellipses with dimensions $\hat{r}_1$, $\hat{r}_2$, and $\hat{r}_3$ along the perpendicular directions $\overrightarrow{p}$, $\overrightarrow{q}$, and $\overrightarrow{r}$, respectively.

The above deformations are microscopic. In exact analogy with the above procedures, we can also define the macroscopic deformations, $\bar{r}_1$, $\bar{r}_2$, and $\bar{r}_3$. As the aim of such transformations is to make symmetric spherical objects become ellipsoidal objects, and vice versa, and the equation that is the basis of these operations is Eq. (38), it is clear to see that if

$$\frac{\partial x_i}{\partial s_j} = \frac{1}{3}(\bar{r}_1+\bar{r}_2+\bar{r}_3)\delta_{ij} + (\bar{r}_1-\bar{r}_3)Q_{ij}(\vec{p}) + (\bar{r}_2-\bar{r}_3)Q_{ij}(\vec{q}), \tag{40}$$

$$\frac{\partial s_j}{\partial x_i} = \frac{1}{3}\left(\frac{1}{\bar{r}_1}+\frac{1}{\bar{r}_2}+\frac{1}{\bar{r}_3}\right)\delta_{ij} + \left(\frac{1}{\bar{r}_1}-\frac{1}{\bar{r}_3}\right)Q_{ij}(\vec{p}) + \left(\frac{1}{\bar{r}_2}-\frac{1}{\bar{r}_3}\right)Q_{ij}(\vec{q}), \tag{41}$$

are the assumed macroscopic counterparts of Eqs. (33) and (34), respectively, we will arrive at the macroscopic version of Eq. (38)

$$\bar{r}_1 = \frac{r_1}{r}, \quad \bar{r}_2 = \frac{r_2}{r}, \quad \bar{r}_3 = \frac{r_3}{r}, \tag{42}$$

where r_1, r_2, and r_3 are the macroscopic ellipsoidal axes defined in Eq. (17).

Finally, for future use, let us write the uniaxial versions of these transformations for the case where $\bar{r}_2 = \bar{r}_3$,

$$\frac{\partial x_i}{\partial s_j} = \frac{1}{3}(\bar{r}_1+2\bar{r}_2)\,\delta_{ij} + (\bar{r}_1-\bar{r}_2)\,Q_{ij}(\vec{p}) = \bar{r}_2\delta_{ij} + (\bar{r}_1-\bar{r}_2)\,p_i p_j, \tag{43}$$

$$\frac{\partial s_j}{\partial x_i} = \frac{1}{3}\left(\frac{1}{\bar{r}_1}+\frac{2}{\bar{r}_2}\right)\delta_{ij} + \left(\frac{1}{\bar{r}_1}-\frac{1}{\bar{r}_2}\right)Q_{ij}(\vec{p}) = \frac{1}{\bar{r}_2}\delta_{ij} + \left(\frac{1}{\bar{r}_1}-\frac{1}{\bar{r}_2}\right)p_i p_j. \tag{44}$$

In Sec. III, we will give an example of how these transformations act on physical quantities and we will study the nematic viscosity from the point of view of the HB approach.

III. A GEOMETRICAL APPROACH TO THE LESLIE COEFFICIENTS

A. The eccentricity and the Onsager relation

Previously, we have seen that the operators dx^i/ds^j and ds^i/dx^j [Eqs. (40) and (41)] transform spherically symmetric objects into ellipsoidal objects, and vice versa. According to the HB approach these transformations transcend the geometrical scenario in which they have been deduced. They transform all mathematical quantities describing spherical symmetric objects into the corresponding quantities in an ellipsoidal geometry. Consequently, when they act in a mathematical quantity of an isotropic liquid, they will transform it into the corresponding mathematical quantity of a nematic liquid. In this section we will apply this prescription to study the nematic viscosity of a uniaxial sample. It will be assumed that the stress tensor σ_{ij} of a LC composed of uniaxial particles should be obtained through the application of Eqs. (43) and (44) [17,32–35] on $\eta\partial_l v^k$, which is the stress tensor of an isotropic liquid, where η is the corresponding isotropic viscosity and $\partial_i v_j$ is the gradient of the isotropic fluid velocity. Namely, it is assumed that

$$\sigma_{ij} = \frac{ds^l}{dx^i}\frac{dx^j}{ds^k}(\eta\partial_l v^k). \tag{45}$$

According to this proposition, after the full development of this equation only one parameter would be free: the viscosity η of the virtual isotropic liquid. All other parameters would be determined by the geometry of the nematic cell, which are contained in the transformations in Eqs. (43) and (44). This is the reason why we have studied the geometry of the nematic grain that, save for η, would completely determine the nematic viscosity. Notwithstanding its strong physical appeal and conceptual simplicity, this approach never gives a complete explanation of the phenomenology observed in the LC's viscosity. We have explained this in a previous work [31]. In essence, Eq. (45) does not take into account all the degrees of freedom of an anisotropic particle. For an observer at the laboratory the velocity $\vec{v}$ of an extended anisotropic rigid body is composed of two terms,

$$\vec{v} = \vec{v}^c + \vec{w} \times \vec{\rho}, \tag{46}$$

one considering the motion of the its center of mass $\vec{v}^c$ and the other considering the rotation of its internal points (which are assigned by the parameter $\vec{\rho}$) around the center of mass, where $\vec{w}$ is the instantaneous angular velocity. Both of these terms produce dissipation, but the latter is not contained in Eq. (45), even after the application of the HB transformation. The absence of this rotational term makes the HB approach incomplete, hindering, for example, the correct use of the Onsager theorem [31], as we will see.

In order to resolve these problems and construct a model that embraces the content of Eq. (46), the form of the isotropic stress tensor has been modified with the introduction of two viscosity coefficients, one for the shearing flow and the other for the rotational flow of a liquid, i.e.,

$$\sigma_{ij} = 2\frac{ds^l}{dx^i}\frac{dx^j}{ds^k}(\eta_1 A_{ij} + \eta_2 N_i n_j), \tag{47}$$

where $A_{ij} = (\partial_i v_j + \partial_j v_i)/2$ and $\vec{N} = [\dot{\vec{n}} - (\vec{w} \times \vec{n})]$ represent the shearing flow and the rotational flow, respectively. η_1 is the extension of the viscosity term already present in the usual HB approach and η_2 has been introduced to attend to the rotational viscosity.

The results obtained with the introduction of this new term are very stimulating, and are the basis of the developments presented in this paper. When we substitute Eqs. (43) and (44) in Eq. (47) and compare the ensuing equations with the ELP form of the stress tensor [4–9],

$$\begin{aligned}\sigma_{ij} &= \alpha_1 n_i n_j n_k n_l A_{kl} + \alpha_2 n_i N_k + \alpha_3 n_j N_k + \alpha_4 A_{ij} \\ &\quad + \alpha_5 n_i A_{jk} n_k + \alpha_6 n_j A_{ik} n_k,\end{aligned} \tag{48}$$

where α_1, α_2, α_3, α_4, α_5, and α_6 are the Leslie coefficients [5] that are subject to the Parodi relation [6], $\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$, we arrive at

$$\begin{aligned}\alpha_1 &= \frac{(r_1 - r_2)^2}{r_1 r_2}\eta_1, \quad \alpha_2 = \frac{r_1}{r_2}\eta_2, \quad \alpha_3 = 0, \\ \alpha_4 &= \eta_1, \quad \alpha_5 = \frac{r_1 - r_2}{r_2}\eta_1, \quad \alpha_6 = \frac{r_2 - r_1}{r_1}\eta_1.\end{aligned} \tag{49}$$

Before studying the consequences of the above relations, we must be aware that these equations cannot be the final answer to the nematic viscosity problem. From them we see that we would have $\alpha_3 = 0$, which clearly does not agree with the experimental facts. Nevertheless, we recall that the value of α_3 is always very small, according to de Gennes [1]. For N-(4-Methoxybenzylidene)-4-butylaniline (MBBA), its value corresponds to only 1.5% of the value of α_4, the isotropic term. So, although this is not the final solution of the viscosity problem, we will continue by considering the above relations as an approximation for the case where α_3 can be disregarded. We will show that this is indeed the case and that the above results dominate the experimental data when the ratio α_3/α_4 is small. For now, let us assume these equations and study their consequences.

Using the definition of Miesowicz's coefficients [1], $m_1 = (\alpha_4 + \alpha_5 - \alpha_2)/2$, $m_2 = (\alpha_3 + \alpha_4 + \alpha_6)/2$, $m_3 = \alpha_4/2$, in Eq. (49), we arrive at

$$m_1 = \frac{r_1}{2r_2}(\eta_1 - \eta_2), \quad m_2 = \frac{r_2}{2r_1}\eta_1, \quad m_3 = \frac{\eta_1}{2}. \tag{50}$$

Due to the form of these equations, it is possible to construct an expression combining the Miesowicz's coefficients,

$$\sigma = \frac{m_3}{m_1}\frac{m_3}{m_2} = \frac{\eta_1}{\eta_1 - \eta_2}, \tag{51}$$

which depends only on the coupling constants η_1 and η_2. Consequently, σ could be used to measure the relative values of η_1 and η_2. If, for example, the experimental data reveal that $\sigma = 1$, we would have $\eta_2 \approx 0$, and the introduction of η_2 in Eq. (47) would not have experimental support. In order to verify what the experimental results say about the values of η_1 and η_2, we have collected experimental data of nematic viscosity from the literature [10–14] and, in order to have a broad panorama of the range of the nematic phase in which η_2 exists, only those data encompassing the entire range of the nematic phase have been considered. The results are shown in Fig. 1, where a uniformized temperature scale was constructed [15–17,36,37] in such a way that, for the nematic-crystalline transition, the temperature $T = 0$ was attributed, while, for the nematic-isotropic (*N-I*) transition, the temperature $T = 1$ was attributed. The distribution of experimental data shows that along the entire range of the nematic phase one has $\sigma \approx 1/2$. This is a gratifying result because it clearly indicates that $\eta_2 \neq 0$, showing that the introduction of this new viscosity coefficient has strong experimental support. Furthermore, the quasiconstancy of σ along the entire nematic phase also indicates that the non-null values of η_2 are not accidental characteristics of a given region of the nematic phase, but that the term that we have added is always present. Finally, it must be noted that in the neighborhood of the *N-I* phase transition a slight increase of the value of σ is observed. This growth is a consequence of the approximation of the *N-I* phase transition region; at the isotropic phase we would have $\sigma = 1$. Of course, this growth in the value of σ does not increase continuously to $\sigma = 1$ because the *N-I* phase transition is discontinuous.

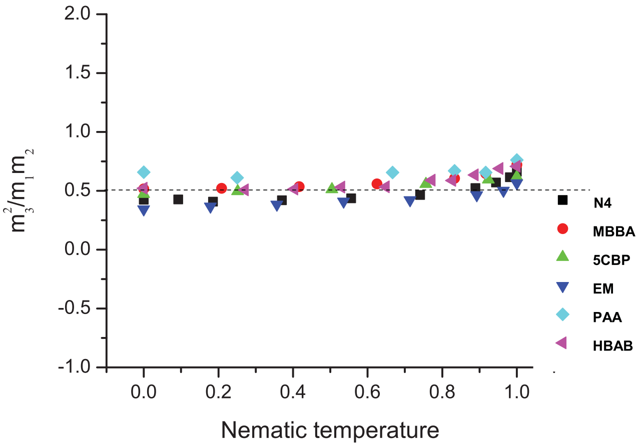

FIG. 1. (Color online) Parameter $\sigma = m_3^2/m_1 m_2$ vs the uniformized nematic temperature scale [15–17,36,37]. The use of the Onsager relation in the stress tensor obtained from the extended HB approach, proposed in Ref. [31], leads to the prediction that σ would assume values around $\sigma \approx 1/2$. Furthermore, as the *N-I* phase transition is approached the effective anisotropy of the nematic grain is diminished and the value of σ would increase, approaching the value $\sigma = 1$. The experimental data exhibited in this graph [10–14] completely confirm these predictions. Although the compounds can be easily recognized by their abbreviated names in the figure, their scientific names and the authors of the measurements can be found in the quoted references.

Another striking result that follows from Eq. (47) is that η_1 and η_2 are not independent, but connected by the geometry of the nematic grain developed above. It is clear that the application of the Parodi relation, $\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$, to the set of equations given in Eq. (49) gives

$$\eta_2 = -e\eta_1, \tag{52}$$

where e has been defined in Eqs. (23) and (25). When we substitute this result in Eq. (51), we obtain

$$\sigma = \frac{1}{1+e}, \tag{53}$$

which completely explains the experimental data described above, as the effective eccentricity of a nematic grain does not depend on its dimensions (r_1 and r_2), but only on its ratios [see Eq. (23)]. We expect that σ would assume approximately the same values for all nematic compounds. Furthermore, save for the neighborhood of the *N-I* phase transition, we would always have $r_1 \gg r_2$. We see that $e = 1 - (r_2/r_1)^2 \approx 1$, implying that such a value would be found around $\sigma \approx 1/2$. Otherwise, as the *N-I* 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_2/r_1, making $e \to 0$ and $\sigma \to 1$. Using the value of e given in Eq. (27), we arrive at

$$\sigma = \frac{4-S}{4+2S}, \tag{54}$$

which shows that when $S \approx 1$, we would have $\sigma \approx 1/2$. Otherwise, as $S \to 0$, we would have $\sigma \to 1$.

These results have not been predicted by other nematic rheological models. They determine the value of the parameter defined in Eq. (51) along the entire range of the nematic phase without the need for any adjustable free variable. They are a direct consequence of the use of the extended HB approach, where a rotational term has been added, and show that the Miesowicz's coefficients are connected by the eccentricity of the nematic grain through the application of the Onsager theorem.

B. The Leslie coefficients

Notwithstanding the agreement between theory and experiment, the preceding results can be only an approximation of the viscosity problem. α_3 cannot be null as we assumed with the acceptance of Eq. (47) as the starting point of our development. Here, we will further develop the HB approach and obtain a consistent expression for the Leslie coefficients. To accomplish this task, we will further generalize the form of Eq. (47) by adding the transpose of $N_i n_j$ to it, giving,

$$\sigma_{ij} = 2\frac{ds^l}{dx^i}\frac{dx^j}{ds^k}(\eta_1 A_{ij} + \eta_2 N_i n_j + \eta_3 N_j n_i), \tag{55}$$

where η_3 is a new viscosity term coupling $N_j n_i$. Moreover, by repeating the procedure that we have followed to arrive at Eq. (48) we find that

$$\begin{gathered}\alpha_1 = -\frac{(r_1 - r_2)^2}{r_1 r_2}\eta_1, \quad \alpha_2 = \frac{r_1}{2r_2}\eta_2, \quad \alpha_3 = \frac{r_2}{2r_1}\eta_3,\\ \alpha_4 = \eta_1, \quad \alpha_5 = \frac{r_1 - r_2}{r_2}\eta_1, \quad \alpha_6 = \frac{r_2 - r_1}{r_1}\eta_1.\end{gathered} \tag{56}$$

With this result we understand why the previous versions of the HB approach were unable to describe the nematic viscosity problem: α_2 and α_3 are entirely determined by η_2 and η_3, and vice versa.

In terms of these new parameters, the Parodi relation becomes

$$e\eta_1 + \eta_2 + (1-e)\eta_3 = 0, \tag{57}$$

which contains the previous result, Eq. (52), as a special case and shows that the terms η_1, η_2, and η_3 are connected by the quenched geometry of the nematic phase. Finally, using Eq. (48) we see that the Leslie coefficients are completely determined by two viscosity parameters, η_1 and η_3, for example, and by the eccentricity e:

$$\begin{gathered}\alpha_1 = -\frac{(1-\sqrt{1-e})^2}{\sqrt{1-e}}\eta_1, \quad \alpha_2 = -\frac{e\eta_1 + (1-e)\eta_3}{\sqrt{1-e}}\eta_2,\\ \alpha_3 = \sqrt{1-e}\,\eta_3, \quad \alpha_4 = \eta_1, \quad \alpha_5 = \left(\frac{1}{\sqrt{1-e}} - 1\right)\eta_1,\\ \alpha_6 = -(1-\sqrt{1-e})\eta_1.\end{gathered} \tag{58}$$

In terms of these expressions for the Leslie coefficients, the parameter σ becomes

$$\begin{aligned}\sigma &= \frac{1}{\left(1+\frac{\eta_3}{\eta_1}\right)\left[(1+e) + (1-e)\frac{\eta_3}{\eta_1}\right]}\\ &= \frac{1-e^2}{(1-e)\left(\frac{\alpha_3}{\alpha_4}\right)^2 + 2\sqrt{1-e}\frac{\alpha_3}{\alpha_4} + (1-e^2)},\end{aligned} \tag{59}$$

where, for completeness, we have written σ in terms of η_3/η_1 and α_3/α_4. When η_3/η_1, or α_3/α_4 are small, we obtain

$$\begin{aligned}\sigma &= \frac{1}{1+e} - \frac{2}{(1+e)^2}\frac{\eta_3}{\eta_1} + \frac{3+e^2}{(1+e)^3}\frac{\eta_3}{\eta_1}^2 + \cdots\\ &= \frac{1}{1+e} - \frac{2}{\sqrt{1-e}(1+e)^2}\frac{\alpha_3}{\alpha_4}\\ &\quad + \frac{3+e^2}{(1-e)(1+e)^3}\frac{\alpha_3}{\alpha_4}^2 + \cdots,\end{aligned} \tag{60}$$

which confirms the results that followed from Eq. (40) when η_3/η_1or α_3/α_4 can be neglected, which are the conditions prevailing experimentally as long as we know that, for all nematic calamitic compounds, any of these ratios are always small. Under these conditions, the uniformity and diversity of data shown in Fig. 1 constitutes a striking confirmation that the model presented in this paper can correctly describe the nematic viscosity.

IV. CONCLUSION

In this paper, we have shown that the nematic viscosity can be understood from a geometric point of view. A modified version of the HB approach [31] has been used to express the Leslie coefficients [Eq. (58)] in terms of two viscosity parameters η_1 and η_3 and the quenched eccentricity e of the nematic grain. In order to clearly define the thermalized geometry and especially this parameter e, we have presented a study of the thermal behavior of a nematic molecule where we have shown that a straightforward understanding of this geometrization of the nematic phase is achieved using de Gennes' concepts of microscopic and macroscopic order parameters [1]. Our results have been compared with experimental results, where it has been shown that they agree with the experimental data.

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