

## Thermal diffusivity and the conformal transformation on nematic liquid crystals

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(Received 23 November 2007; published 29 April 2008)

In this paper, we will study theoretically and experimentally the anisotropy of the thermal diffusivity in nematic liquid crystals. We will show that the Baalss-Hess conformal transformation [D. Baalss and S. Hess, Phys. Rev. Lett. **57**, 86 (1986)], can be used to obtain a free of adjustable parameters equation that describes the anisotropy of the heat diffusion in these materials. The results of this theory will be compared with experimental data. This study will partially confirm the widely known experimental evidences that indicate that the thermal diffusivity is larger in the director direction than in the one perpendicular to it. For calamitic nematic phases this is true; for discotic nematic ones the reverse situation would be found; the diffusivity would become larger in the direction perpendicular to the director. We will also present experimental data supporting this theoretical prediction. The data comprehend the calamitic and discotic nematic lyotropic phases and a nematic thermotropic phase.

DOI: 10.1103/PhysRevE.77.041709

PACS number(s): 42.70.Df, 61.30.-v

### I. INTRODUCTION

The anisotropy observed in the thermodynamical properties of the liquid crystals (LC) are among the most unexpected phenomena of the nature; liquid crystals are liquids that present anisotropic behavior. Their anisotropies cover a broad range of properties such as viscosity, elasticity, birefringence, electric permittivity, magnetic susceptibility, thermal diffusion, etc. As each of these properties is function of the degree of alignment of the anisotropic grains that constitute the nematic liquid crystals (NLC) (molecules for thermotropics and micelles for lyotropics) [1], a straightforward idea to understand the origin of the nematic anisotropies is to attribute to the microscopic anisotropy of their grains the source of the anisotropies observed on macroscopic scale [2]. Accordingly, Hess and Baalss (HB) [2–6] assumed that if one could imagine a way by which the molecules of these liquids 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 its vanishing. Inversely, if the idealized spherical molecules of an isotropic liquid could be deformed until they assume the ellipsoidal form of an idealized nematic molecule, the macroscopic physical properties would be transformed on those observed on the NLC. Essentially, these are the fundamentals of the ideas of the (HB) conformal transformation hypotheses, originally applied to the study of the viscosity of the NLC [2] but, later, also applied to the study of diffusion [7] and elasticity [8], all with excellent results.

In their work, HB assumed that the molecular deformations supposed by their reasoning could not be effectively done on the laboratory; these deformations were conjectured to justify the use of a mathematical transformation in which the physics of an anisotropic fluid could be written in terms of the physics of an isotropic one. Nevertheless, as it will be

shown here, it is possible to do what HB assumed as a conjecture. In order to make our study we have looked for a phenomenon for which the HB approach gives new and unquestionable results; the thermal diffusivity. For as long as we know, there is not yet a theory of thermal diffusivity for the nematic phase. Even being expected that nematic materials would present anisotropic heat propagation, a detailed quantitative explanation for this fact is yet missing. Nevertheless, without exception, every experimental data agree that the thermal diffusivity along the direction parallel to the director is greater than the one in the direction perpendicular to it [9–18]. As we will see ahead, the HB approach gives a very precise explanation to it and, furthermore, predicts the conditions in which this rule would be broken: calamitic nematic lyotropic phase being replaced by discotic nematic ones. This prediction is a consequence of the fact that the ratio between the thermal diffusivity along the two main molecular directions is proportional to the eccentricity of the nematic phase [see Eq. (13)]. Consequently, a change in the micellar shape would be immediately detected in the thermal diffusivity, which is exactly the HB conjecture. The aim of this work is to furnish the theoretical fundamentals of this model, present an experimental example in which the HB conjecture can be explicitly observed, and, furthermore, examine a situation in which is presented an experimental measurement in which the thermal diffusivity along the direction perpendicular to the director is greater than the one along the direction parallel to it.

### II. FUNDAMENTALS

In order to propose an expression to the heat conduction in an isotropic medium we start from the known equation

$$\frac{d}{dt}T - K\nabla^2T = q, \quad (1)$$

where  $T$  is the temperature,  $t$  is the time,  $q$  is the heat flux at the point  $(\vec{r}, t)$ , and  $K$  is the thermal conductivity and consid-

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ers the Laplacian operator in a generalized coordinate system [19]. Moreover, we consider the director of a nematic phase uniformly oriented. In this way, the scale factor  $h_i$ ,  $i = 1, 2, 3$ , become constant that can be absorbed in  $K$ , defining a different  $K$  for each direction,  $K \rightarrow K/h_i^2 = K_i$ . In this context, the above equation can be expressed as

$$\frac{d}{dt}T - K_i \frac{d^2}{dx_i^2}T = q. \quad (2)$$

Furthermore, as a rule, we will use Greek symbols to indicate particles indexes, and latin indexes to indicate its Cartesian components. In order to obtain an expression for the anisotropic nematic thermal conductivity  $K_i$ , we recall the standard expression for the coefficient of thermal conductivity along the direction  $\vec{e}_i$ , given as a time correlation function [20,21],

$$K_i = \frac{1}{Vk_B T^2} \int_0^\infty dt \langle E^i(0) E_i(t) \rangle, \quad (3)$$

where  $E_i(t) = \sum_{\alpha=1}^N W^\alpha p_i^\alpha$  is the transversal current,  $V$  is the volume of the sample,  $k_B$  is the Boltzmann constant,  $p_i^\alpha$  is the  $i$  Cartesian component of the momentum of the particle  $\alpha$ , and  $W^\alpha$  is given by

$$W^\alpha = \left[ \frac{(\vec{p}^\alpha)^2}{2m} - \langle E^\alpha \rangle \right] + \frac{1}{2} \sum_{\beta \neq \alpha} [U(\vec{r}^{\alpha\beta}) + \vec{r}^{\alpha\beta} \cdot \vec{F}^{\alpha\beta}], \quad (4)$$

where  $\vec{r}^{\alpha\beta}$  is the vector giving the distance between the particles  $\alpha$  and  $\beta$ ,  $\vec{F}^{\alpha\beta}$  is the force between the particles  $\alpha$  and  $\beta$ ,  $N$  is total number of particles in the sample, and  $\langle \chi \rangle$  is the statistical average of the random variable  $\chi$ .

Our procedure to obtain  $K_i$  will be to apply on it the Hess conformal transformation, as defined in Ref. [8], and write it in terms of the diffusivity of an hypothetic isotropic fluid. As was done by Hess, it is assumed that the interacting potential  $U(\vec{r}^{\alpha\beta})$  is invariant by a conformal transformation that changes a sphere to an ellipsoid having the shape of the nematic micelles (or molecules). As consequence, it follows that the vector components  $x^i$  and  $s^i$ , used to describe, respectively, elliptical and the spherical symmetric potentials, would transform according to the rule

$$dx^i = \sum_{j=1}^3 \frac{dx^i}{ds^j} ds^j, \quad \frac{d}{dx^i} = \sum_{j=1}^3 \frac{ds^j}{dx^i} \frac{d}{ds^j}, \quad (5)$$

which are constrained by the rule  $\sum_{m=1}^3 \frac{ds^i}{dx^m} \frac{dx^m}{ds^j} = \delta_j^i$ . According to these relations if  $E_i$  and  $S_i$  are the transversal currents for the liquids having ellipsoidal and spherical micelles, respectively, we would have,

$$E^i = \sum_{j=1}^3 \frac{dx^i}{ds^j} S^j, \quad E_i = \sum_{j=1}^3 \frac{ds^j}{dx^i} S_j, \quad (6)$$

which are the rules for contravariant and covariant vectors, respectively. Therefore, in terms of a liquid with spherical symmetric micelles, Eq. (3) becomes

$$K_i = \frac{1}{Vk_B T^2} \sum_{j=1}^3 \sum_{k=1}^3 \left\langle \frac{dx^i}{ds^j} \frac{ds^k}{dx^i} \right\rangle \int_0^\infty dt \langle S^j(0) S_k(t) \rangle, \quad (7)$$

where we have assumed that the connected part in the equation leading Eq. (3) to Eq. (7) can be disregarded. In Ref. [8] it has been shown that the conformal transformations can be written as

$$\frac{dx^i}{ds^j} = a_1 \delta_{ij} + b_1 n_i n_j, \quad \frac{ds^i}{dx^j} = a_2 \delta_{ij} + b_2 n_i n_j, \quad (8)$$

where  $\vec{n}$  is the director at the point  $\vec{x}$  and

$$a_1 = \frac{1}{a_2}, \quad a_2 = \pm \sqrt{\frac{3 - e(1 - S)}{3 - e(1 + 2S)}}, \quad b_1 = \pm 1 - a_1, \quad (9)$$

$$b_2 = \pm 1 - a_2$$

$S$  is the scalar order parameter and  $e = 1 - (\frac{b}{a})^2$  is the micellar eccentricity, where  $b$  and  $a$  are the respective micellar dimensions along the directions perpendicular and parallel to the director. Using these relations it is obtained,

$$K_i = \frac{1}{\tau V k_B T^2} \{ \langle S^i(0) S_i(t) \rangle + b_1 b_2 n_i n_i \langle [\vec{n} \cdot \vec{S}(0)] [\vec{n} \cdot \vec{S}(t)] \rangle \}, \quad (10)$$

where it has been used that  $\langle [\vec{n} \cdot \vec{S}(0)] S_i(t) \rangle = \langle S^i(0) [\vec{n} \cdot \vec{S}(t)] \rangle = 0$ . So, for  $k_i$  perpendicular to  $\vec{n}$  ( $k_\perp$ ) and, for  $K_i$  parallel to  $\vec{n}$  ( $K_\parallel$ ) it is obtained that

$$K_\perp = K_i^S, \quad K_\parallel = (1 + b_1 b_2) k_i^S. \quad (11)$$

Consequently, the anisotropy  $A$  of the thermal conductivity, defined as  $A = K_\parallel / K_\perp - 1$ , is given by

$$A = \frac{3Se}{\sqrt{[3 + e(S - 1)][3 - e(2S + 1)]}}, \quad (12)$$

which is completely determined by the parameters  $S$  and  $e$ . In addition, the parallel (perpendicular) thermal diffusivity  $k_\parallel(k_\perp)$  is related with the parallel (perpendicular) thermal conductivity  $K_\parallel(K_\perp)$  by  $k_\parallel(k_\perp) = K_\parallel(K_\perp) / \rho C_p$ , where  $\rho$  is the density and  $C_p$  is the specific heat of the nematic sample. In this way, the ratio  $k_\parallel / k_\perp$  is given by

$$\frac{k_\parallel}{k_\perp} = 1 + \frac{3Se}{\sqrt{[3 + e(S - 1)][3 - e(2S + 1)]}}. \quad (13)$$

### III. RESULTS AND DISCUSSION

The aim of this section is to give experimental support to the above expression for the anisotropy on the thermal diffusivity and, in consequence, afford consistence to the theory leading to it. An important aspect of the above result is that it does not depend on any adjustable parameter; Eq. (13) is entirely determined by  $S$  and  $e$ , which can be eventually measured in a nematic phase. So, an experimental verification of this equation would afford a strong support to our approach to the nematic thermal diffusivity and, in particular,

to the generality of the applicability of the Hess conformal transformation. In order to investigate the validity of the above equation we notice that, according to it, the product of the variables  $S$  and  $e$  in its numerator determines the sign of the anisotropy on the heat diffusion. It would be null at the isotropic phase,  $S=0$  (which is not an unexpected result, because, if something different happens, we have to say that such a formula is undoubtedly wrong). But, that numerator is also proportional to  $e$  and this proportionality could be easily tested. As we have said at the beginning of this paper, there are lyotropic compounds for which the molecular shape can be changed and, according to this formula, such a change of shape would be detected in the thermal diffusivity. This is one of the purposes of this section, to show that it is possible to detect the molecular change of shape through thermal diffusivity data and, consequently, to test the validity of the Hess argument. According to Eq. (13), for  $e > 0$  (calamitic nematic phase) the anisotropy  $A$  in the thermal conductivity would be positive and  $\kappa_{\parallel}/\kappa_{\perp} - 1 > 0$ . Nevertheless, for  $e < 0$  (discotic nematic phase) it would be negative and in this way is expected the ratio  $k_{\parallel}/k_{\perp} - 1 < 0$ . From an experimental point of view, this ratio was recently determined [22] in a discotic nematic phase at room temperature ( $T=25^{\circ}\text{C}$ ), through the use of the Z-scan technique [23], in a lyotropic mixture of potassium laurate, decanol, and  $D_2O$ . In this paper, using the same Z-scan technique, we report on new measurements of the thermal diffusivity ( $\kappa$ ) in a calamitic nematic phase [17] at room temperature ( $T=25^{\circ}\text{C}$ ) for a particular concentration in weight percentage of potassium laurate (KL:27.07), decanol (DeOH:6.17) and ( $D_2O$ :66.76) [24]. The phase sequence is the following: reentrant isotropic ( $12.5^{\circ}\text{C}$ ) calamitic nematic ( $54.5^{\circ}\text{C}$ ) isotropic. In this study, the two thermal diffusivities  $k_{\parallel}$  and  $k_{\perp}$  have been obtained in an experimental configuration as a laser beam travels in the nematic medium with polarization parallel or perpendicular, respectively, to the director of the nematic sample. The details of the experimental Z-scan technique setup utilized in this work are described in Refs. [22,23]. In the Z-scan experimental technique a Gaussian laser beam ( $\text{TEM}_{00}$ ) is focused to a narrow waist by a lens along the propagation direction of the beam defined as being the  $z$  axis. The sample is moved through the focal plane along the  $z$  direction and the far-field transmittance of an iris centered along the beam propagation direction is measured as a function of the position  $z$  of the sample. As the sample moves along the beam focus, further focusing or defocusing modifies the wave front phase, thereby modifying the detected intensity. A sketch of the Z-scan setup is shown in Fig. 1. Our experimental setup includes a diode laser Ventus MPC600 (from Quantum) with power output adjusted to 47 mW. The beam waist radius  $\omega_o$  is about  $21.5 \mu\text{m}$  and a mechanical chopper (Standford SR540) provides laser pulses (33 ms) incident on the sample. Data acquisition with temporal resolution is made by an oscilloscope model TDS3012 (from Tektronix) and a GPIB board.

The values of the thermal diffusivity for a thermal nonlinear optical response can be determined from fitting the spatial dependence on  $z$  and the temporal dependence via thermal lens model [25] as described by Palffy-Muhoray *et al.* [26] The normalized light transmittance ( $\Gamma$ ), as a function of  $z$  and time  $t$ , can be written as [26]

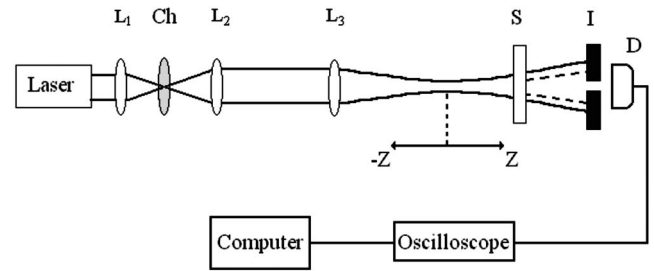


FIG. 1. Sketch of the Z-scan apparatus.  $L_1$ ,  $L_2$ , and  $L_3$  are lenses. Chopper (Ch); sample (S); iris (I), and detector (D).

$$\Gamma(z,t) = \left[ 1 + \left( \frac{\theta}{1 + (1+x^2)t_{co}/2t} \right) \frac{2x}{1+x^2} \right]^{-1}, \quad (14)$$

where  $x=z/z_o$ ,  $z_o$  is the confocal parameter,  $\theta=2.303(-dn/dT)\alpha P/\lambda K$  is the phase shift [27] (phase difference between the center and the outer edge of the beam),  $t_{co} = \omega_o^2/4k$  is the characteristic thermal time,  $k=K/\rho C_P$  is the thermal diffusivity,  $P$  is the power of the laser beam,  $\lambda$  is the wavelength of the laser,  $\alpha$  is the linear optical absorption,  $dn/dT$  is the thermo-optical coefficient,  $K$  is the thermal conductivity,  $\rho$  is the density, and  $C_P$  is the specific heat. Equation (14) is valid under the condition  $\theta \ll 1$  [26]. In liquid crystal medium,  $\theta$ ,  $t_{co}$ , and  $k$  are anisotropic parameters. These parameters,  $\theta_{\parallel}(\theta_{\perp})$ ,  $t_{co\parallel}(t_{co\perp})$ , and  $k_{\parallel}(k_{\perp})$  are defined in a direction parallel (perpendicular) to the director of the nematic sample. The nematic sample was conditioned in sealed planar glass cells (1 mm of light path) from Hellma. Uniform orientation of the sample was performed using an electromagnet ( $H=10 \text{ kG}$ ) for 24 h and checked by crossed polarizers. The laboratory frame axes are defined with the boundary surfaces parallel to the  $x$ - $y$  plane and  $z$  is the axis normal to the biggest surface of the sample cell. The  $x$ - $y$  plane of the sample is defined with the  $x$  ( $y$ ) axis parallel to the length (width) of the cells. The planar alignment of the sample is produced by a magnetic field of 10 kG parallel to the  $x$  axis of the laboratory frame. A small quantity of ferrofluid ( $<0.04 \text{ wt}$ ) was added to the calamitic nematic sample in order to ensure a good quality alignment of the director in the  $N_C$  phase. It is important to mention that in

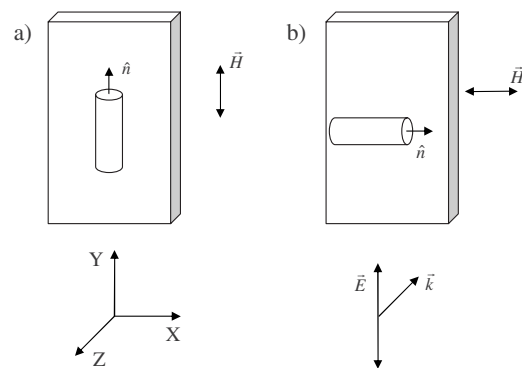


FIG. 2. Experimental configuration of the nematic director, magnetic field, and the laser beam polarization for thermal diffusivity measurements of (a)  $k_{\parallel}$  and (b)  $k_{\perp}$  on the  $N_C$  phase.

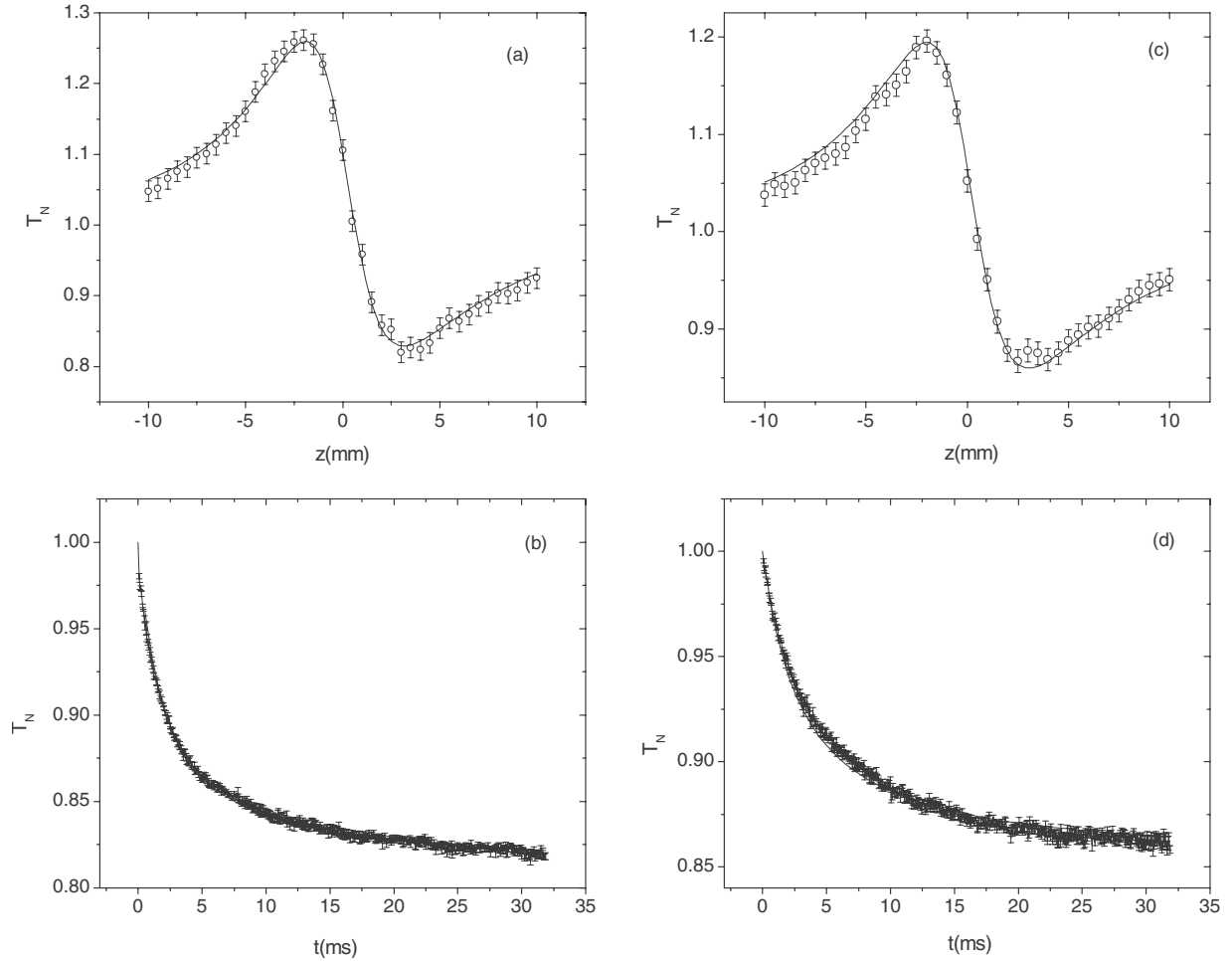


FIG. 3. Typical curves of Z-scan measurements on the lyotropic mixture on the  $N_C$  phase, where (a) and (b) concern the configuration  $\hat{n} \parallel \vec{E}$ , and (c) and (d) concern the configuration  $\hat{n} \perp \vec{E}$ . The solid lines (a) and (c) correspond to the fitting of Eq. (14) with  $t \sim 10t_{co}$  for  $\theta_{\parallel} = 2.261 \times 10^{-3}$  and  $\theta_{\perp} = 1.798 \times 10^{-3}$ , respectively [26]. Typical time dependence transmittance (b) and (d) at fixed ( $z=2.5$  mm) position [27] and  $\theta_{\parallel}(\theta_{\perp})$  phase shift. The solid lines correspond to the fitting of Eq. (14) with  $\theta_{\parallel}(\theta_{\perp}) = 2.261(1.798) \times 10^{-3}$  for  $t_{co\parallel}(t_{co\perp}) = 2.42(2.86)$  ms [26].

this case, the geometry of the planar glass cells favors the orientation of the director of this calamitic nematic phase parallel to the  $x$  axis also by surface effects. The measurements were made at  $T=25$  °C in a controlled temperature device stable at 10 mK. The anisotropic parameters  $\theta$ ,  $t_{co}$ , and  $k$  can be obtained by orientation of the director  $\hat{n}$  along two perpendicular directions, so the configurations between  $\hat{n}$  and the laser beam polarization  $\vec{E}$  are  $\hat{n} \parallel \vec{E}$  and  $\hat{n} \perp \vec{E}$ , respectively. Figure 2 shows a scheme of the relative configuration of the nematic director, magnetic field, and polarization of incident laser beam for measurements of these parameters in the  $N_C$  phase. Figures 3(a) and 3(c) present typical Z-scan curves obtained for the calamitic nematic phase at temperature  $T=25.0$  °C. The experimental data are determined according to Eq. (14) by means of a self-consistent fitting of  $\theta$  and  $t_{co}$  parameters. The fitting procedure employed here is the same utilized by Palfy-Muhoray *et al.* [26]. Figure 3(b) shows a typical time dependence transmittance at a fixed  $z$  position and the fit from Eq. (14). The experimental curves exhibited in Figs. 3(a) and 3(b) correspond to the  $N_C$  phase, for a laser beam traveling in the

nematic medium with polarization parallel to the optic axis of the nematic sample. In the same way, similar curves were obtained in a perpendicular direction to the optic axis of the nematic phase [Figs. 3(c) and 3(d)]. Taking the experimental values  $\omega_o = 21.5$   $\mu\text{m}$  and  $t_{co} = \omega_o^2/4k$ , with  $t_{co\parallel}(t_{co\perp})$  anisotropic parameters, into account, we obtain the parallel ( $k_{\parallel}$ ) and perpendicular ( $k_{\perp}$ ) thermal diffusivities defined, respectively, in a direction parallel or perpendicular to the director of the nematic sample, and the ratio  $\kappa_{\parallel}/\kappa_{\perp}$ . These important parameters are given in Table I.

From theoretical point of view, the ratio  $k_{\parallel}/k_{\perp}$  can be determined via Eq. (13). Taking the ratio between the micellar and the molecular dimensions [9,17], eccentricity and order parameter ( $S \cong 0.5$  for lyotropic nematic phases [28] and  $S \cong 0.59$  for a thermotropic nematic phase [9]) into account, we obtain from Eq. (13) the ratio  $k_{\parallel}/k_{\perp}$  as indicated in Table I. As we see in Table I, we have obtained a good accordance of the results predicted by our model concerning nematic phase studied in this work. Essentially, we have shown that the conformal transformation approach can be used to produce an adjustable parameters expression for the anisotropic part of the thermal diffusivity in NLC. The expression that

TABLE I. Values of the ratio of the micellar (molecular) dimensions, micellar (molecular) eccentricity, and ratio of the thermal diffusivities in the nematic lyotropic (thermotropic) phases at  $T = 25^\circ\text{C}$ .

Nematic phase	$b/a$	$e$	$k_{\parallel}/k_{\perp}$ (measured)	$k_{\parallel}/k_{\perp}$ (estimated)
$N_C$	0.59	0.65	1.18	1.46
$N_D$	1.39	-0.94	0.93	0.66
$N(5CB)$	0.36	0.87	1.73	1.90

we have found depends solely on the order parameter  $S$  and the eccentricity  $e$  of the nematic micelle or molecule and has predicted that the known experimental result (the thermal diffusivity is larger in the director direction than in the direction perpendicular to it) is only true for nematic phases ( $e > 0$ ). If a discotic nematic phase ( $e < 0$ ) was used the reverse result would be observed.

Some years ago Hess and co-workers studied the problem of (self-)diffusion in a liquid crystal and found a relation that resembles our Eq. (15). If  $D_{\parallel}$  and  $D_{\perp}$  are, respectively, the diffusion coefficients in the direction parallel and perpendicular to the director direction, in terms of our notation it can be shown that the equation obtained by those authors have the form  $D_{\parallel}/D_{\perp} = 3Se/[3 - e(S+2)]$ . The similarities found on the form of this equation and Eq. (13) above, re-

veals that these equations are closely related and would describe similar phenomena. Furthermore, to be in consonance with the experimental findings of our paper, we would like to notice that this equation also predicts a change in the direction of the diffusion as the geometry of the micelle changes from calamitic to discotic, i.e., the diffusion anisotropy changes sign with the eccentricity  $e$ .

To sum up, experimental data and theoretical studies of thermal diffusivity in NLC have been gathered to produce a new understanding of the thermal diffusivity in these materials and, furthermore, to illustrate the potentialities of use of the Baalss-Hess conformal transformation approach in the study of anisotropic phenomena in NLC. This work presents the results of such measurements, which comprehend the calamitic and discotic nematic lyotropic phases and a nematic thermotropic phase, and they confirm the theoretical prediction. If we consider the difficulties involved in the simultaneous determination of  $S$  and  $e$ , it must be concluded that the agreement is good.

#### ACKNOWLEDGMENTS

We are thankful to the Brazilian Agencies CAPES, CNPq, and Fundação Araucária (PR) for their financial support of this work.

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