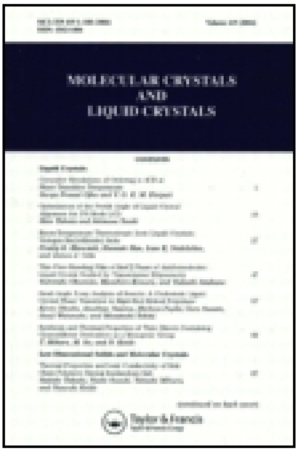

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Nematoacoustic Theory

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Nematoacoustic Theory

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*There is plenty of experimental evidence that the propagation of an ultrasonic wave in a nematic liquid crystal affects the director **n**, which represents the average molecular orientation, thus producing detectable optical effects. There have been several attempts to explain these observations on the basis of a coherent variational theory. We present here a general theory for nematoacoustics that incorporates flow effects. An explicit application of the proposed theory to a simple computable case is given in order to estimate phenomenological parameters involved in the theory and by using available experimental data.*

Keywords nematic liquid crystals; ultrasonic waves; acoustics; Korteweg fluids; capillarity

PACS numbers 61.30.-v, 62.60.+v

I. Introduction

It has long been recognized that sound waves interact with the orientational order of nematic liquid crystals, leading to a realignment of the liquid–crystal molecules [1–3].

Among the experimental observations, the most important ones have been the anisotropy and the frequency dependence of both attenuation and dispersion of ultrasonic waves propagating in nematic liquid crystals [4–8] and the re–orientating acoustic action exerted on a uniformly aligned nematic cell [9–11].

A fundamental task for the interpretation of the results has been the understanding of the interaction mechanism between the acoustic field (represented by a wave vector $\boldsymbol{k}$) and the mesophase director $\boldsymbol{n}$ (representing the average molecular orientation). It was already anticipated by Helfrich [12] that the acoustic field has an orienting action on the nematic director field. In Ref. [13] the first experimental results are reported which suggested to introduce an elastic interaction energy between the acoustic wave and the nematic director field to account for a direct nemato–acoustic coupling capable of inducing distortions on the texture. On the theoretical side, the nature of the interaction was later elucidated as reported in Refs. [14] and [15], where a theory was proposed in which the acoustic–nematic interaction results from the coupling between the density modulation induced by the acoustic wave and the nematic director. According to this theory, the actual realization of the nematoacoustic coupling occurs at time scales much larger than those of acoustic vibrations and, as a result, it is actually the time averaged interaction energy that enters the nematic elastic energy. This is because one presumes that the variations of the nematic texture take place at time and length scales much larger than the acoustic characteristic times

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and lengths, so that especially an ultrasonic wave propagates in an undistorted medium. The proposed averaged interaction energy $\langle V_{\rm avg}\rangle$ has the form

$$\langle V_{\rm avg}\rangle = \frac{1}{2}u^2\left(\boldsymbol{k}\cdot\boldsymbol{n}\right)^2, \tag{1}$$

where the brackets $\langle\rangle$ denote a time average over rapid oscillations and u is related to the coupling constant between the director field and the acoustically–induced mass density gradients, here defined as a nematoacoustic susceptibility.

Very recently, a slight variant of this assumption was posited in Ref. [16] where a variational theory for nematoacoustics was put forward. According to this new theory, liquid crystals are to be regarded as anisotropic *Korteweg fluids* [17] at the time and length scale at which the sound field produces density modulations. In general, a Korteweg fluid is a special fluid with the elastic stress tensor depending on both first and second gradients of the density field [18]. For anisotropic fluids, such as nematic liquid crystals, Korteweg theory was adapted in [16] to nematoacoustics by positing an elastic additional energy density which is quadratic in the fields ($\boldsymbol{n}$, $\nabla\varrho$), where ϱ is the mass density. The corresponding Korteweg stress turns out to be hyperelastic (see [19]). On the other hand, in [16], at the time scale of the acoustic vibrations, the director texture is still regarded as immobile in the same spirit of previous ideas and works and in line with experimental studies where the director is kept fixed by external magnetic fields. Here, within the theory proposed in [16], we shall relax such an assumption and the director is left free to vary slightly in time and distorted in space around a uniform and constant in time orientation, this latter kept fixed by an external magnetic field. Under these general conditions, we further assume that the allowed director motion is actually librational as a consequence of the sound wave that propagates through the nematic liquid crystal. We shall interpret by means of the theoretical outcome of our analysis, the experimental results long published in the literature and we shall estimate some phenomenological parameters involved in the theory.

The paper is organized as follows. In Sec. II we shall recapitulate the nematoacoustic variational theory built upon Korteweg theory and put forward in Ref. [16] and the general system of governing balance equations that stem from it. In Sec. III plane wave solutions of these equations are sought, in particular, including the librational motion of the director. In Sec. IV an estimate of phenomenological parameters is provided based on experimental data on anisotropic dispersion and wave attenuation theoretically predicted by our nematoacoustic theory. Finally, in Sec. V a conclusion is reported and an outline of future research directions.

II. Theory

In this section we shall review a variational theory for acoustic interaction in nematic liquid crystals [16].

In nematoacoustics two important hydrodynamic variables are the director $\boldsymbol{n}$ and the mass density ϱ. In particular, the spatial gradients of the mass density $\nabla\varrho$ play a crucial role when acoustic phenomena are concerned. For isotropic fluids, Korteweg proposed constitutive equations for stresses arising in response to density gradients, thus positing a theory for the so–called *second–grade* fluids. For anisotropic fluids, such as a nematic liquid crystal, the local microstructure, here represented by the unit vector field $\boldsymbol{n}$, has to be taken into account. In terms of potential energy, the corresponding elastic energy density is a function of ϱ, $\nabla\varrho$, $\boldsymbol{n}$ and the spatial derivatives of $\boldsymbol{n}$. In [16] a generalization of Korteweg fluids to include complex fluids has been introduced and the balance equations have been derived along with stresses and traction laws as resulting from a principle of virtual power. At the same time, the theory presented in [16] generalizes the Ericksen–Leslie–Parodi theory for incompressible nematic liquid crystals by removing the incompressibility constraint and taking density gradients $\nabla\varrho$ into appropriate account.

Following Ref. [16], we consider a nematic liquid crystal occupying a region $\mathscr{B}$. Let the total free energy stored in $\mathscr{B}$ be given by

$$\mathscr{F} := \int_{\mathscr{B}} F dV, \tag{2}$$

with V the volume measure and F the energy density given by

$$F := \frac{1}{2}\varrho \boldsymbol{v}^2 + W_{\mathrm{e}}(\boldsymbol{n}, \nabla\boldsymbol{n}) + \varrho\sigma_{\mathrm{K}}(\varrho, \nabla\varrho, \boldsymbol{n}). \tag{3}$$

Here $\boldsymbol{v}$ is the velocity field of the liquid crystal and

$$W_{\mathrm{e}}(\boldsymbol{n}, \nabla\boldsymbol{n}) := \frac{1}{2}K_1(\operatorname{div}\boldsymbol{n})^2 + \frac{1}{2}K_2(\boldsymbol{n}\cdot\operatorname{curl}\boldsymbol{n})^2 + \frac{1}{2}K_3|\boldsymbol{n}\times\operatorname{curl}\boldsymbol{n}|^2, \tag{4}$$

$$\sigma_{\mathrm{K}}(\varrho, \nabla\varrho, \boldsymbol{n}) := \sigma_0(\varrho) + \frac{1}{2}[u_1|\nabla\varrho|^2 + u_2(\nabla\varrho\cdot\boldsymbol{n})^2], \tag{5}$$

are two different contributions to the elastic energy density, the former being the Frank distortion energy per unit volume and the latter the Korteweg elastic energy per unit mass of an acoustic origin. Notice that we are not considering here either the kinetic energy of the microstructure $\boldsymbol{n}$, usually taken to be a quadratic form of the material derivative $\dot{\boldsymbol{n}} = \frac{\partial\boldsymbol{n}}{\partial t} + (\nabla\boldsymbol{n})\boldsymbol{v}$, or the potential energy for external actions exerted on $\boldsymbol{n}$. The term σ_{K} in (3) is a contribution of free energy per unit mass describing the elastic energy of the nematic liquid crystal due to spatial variations of the mass density. It also takes into account the interaction between the variations of the mass density and the nematic texture, thus bearing the meaning of acoustic interaction. The acoustic susceptibilities u_1 and u_2 are assumed to be constitutive parameters independent of ϱ. Moreover, it is easily seen that for such an additional energy density to be positive semidefinite, it is necessary and sufficient that

$$u_1 \geq 0 \quad \text{and} \quad u_1 + u_2 \geq 0. \tag{6}$$

As for W_{e} in (3), it represents a general curvature elastic energy stored in the nematic texture described by the field $\boldsymbol{n}$. In the following we shall consider the one-constant approximation, $K_1 = K_2 = K_3 = K$ and W_{e} reduces to the simpler harmonic map energy

$$W_{\mathrm{e}}(\nabla\boldsymbol{n}) = \frac{1}{2}K|\nabla\boldsymbol{n}|^2. \tag{7}$$

Nematic liquid crystals are dissipative fluids. Accordingly, we suppose that there exists a positive-definite, frame–indifferent dissipation functional represented as

$$\mathscr{R} := \int_{\mathscr{B}} R_{\mathrm{a}} dV. \tag{8}$$

The corresponding acoustic dissipation function, Rayleigh function R_a, depends on the director $\boldsymbol{n}$, its corotational derivative

$$\mathring{\boldsymbol{n}} := \dot{\boldsymbol{n}} - \mathbf{W}\boldsymbol{n}, \tag{9}$$

where

$$\mathbf{W} := \frac{1}{2}[(\nabla\boldsymbol{v}) - (\nabla\boldsymbol{v})^{\mathsf{T}}]$$

is the *vorticity* tensor, and on the *stretching* tensor

$$\mathbf{D} := \frac{1}{2}[(\nabla\boldsymbol{v}) + (\nabla\boldsymbol{v})^{\mathsf{T}}].$$

R_a is a quadratic function in the pair $\left(\mathring{\boldsymbol{n}}, \mathbf{D}\right)$. In nematoacoustics, a nematic liquid crystal is regarded as being compressible, and so the velocity field $\boldsymbol{v}$ is no longer solenoidal. As a consequence, R_a also depends on $\operatorname{tr}\mathbf{D}$. The most general dissipation function turns out to be

$$\begin{aligned} R_a := {} & \frac{1}{2}\gamma_1\mathring{\boldsymbol{n}}\cdot\mathring{\boldsymbol{n}} + \gamma_2\mathring{\boldsymbol{n}}\cdot\mathbf{D}\boldsymbol{n} + \frac{1}{2}\gamma_3\mathbf{D}\boldsymbol{n}\cdot\mathbf{D}\boldsymbol{n} + \frac{1}{2}\gamma_4\mathbf{D}\cdot\mathbf{D} \\ & + \frac{1}{2}\gamma_5(\boldsymbol{n}\cdot\mathbf{D}\boldsymbol{n})^2 + \frac{1}{2}\gamma_6(\operatorname{tr}\mathbf{D})^2 + \gamma_7(\operatorname{tr}\mathbf{D})\boldsymbol{n}\cdot\mathbf{D}\boldsymbol{n}, \end{aligned} \tag{10}$$

where $\gamma_1, \ldots, \gamma_7$ are viscosities, considered as functions of the mass density ϱ.

It has been shown in [16] that the dissipation functional (density) is positive semidefinite provided that the following inequalities hold

$$\gamma_1 \geqslant 0, \tag{11a}$$

$$\gamma_3 + 2\gamma_4 \geqslant 0, \tag{11b}$$

$$\gamma_4 \geqslant 0, \tag{11c}$$

$$\gamma_4 + 2\gamma_6 \geqslant 0, \tag{11d}$$

$$\gamma_3 + \gamma_4 + \gamma_5 + \gamma_6 + 2\gamma_7 \geqslant 0, \tag{11e}$$

$$\gamma_1\gamma_3 + 2\gamma_1\gamma_4 - \gamma_2^2 \geqslant 0, \tag{11f}$$

$$\gamma_4(\gamma_3 + \gamma_4 + \gamma_5 + \gamma_6 + 2\gamma_7) + 2[\gamma_6(\gamma_3 + \gamma_4 + \gamma_5) - \gamma_7^2] \geqslant 0. \tag{11g}$$

All these inequalities play a crucial role in proving the positivity of the attenuation of sound waves propagating through the nematic liquid crystal.

A. *Balance Equations*

We introduce here the equations that govern the acoustic propagation in nematic liquid crystals, assuming that a nematic liquid crystal, as seen from an acoustic wave propagating through it, behaves like an anisotropic, compressible Korteweg fluid with free energy as in Eq. (2) and with Rayleigh dissipation function R_a as in Eq. (10). Following the general theory presented in [16] and in [20], in the absence of body force, the balance of linear momentum is expressed by the equation

$$\varrho \dot{\boldsymbol{v}} = \operatorname{div}(\mathbf{T}_{\mathrm{K}} + \mathbf{T}_{\mathrm{dis}} + \mathbf{T}_{\mathrm{E}} + \mathrm{W}_{\mathrm{e}}\mathrm{I}). \tag{12}$$

In this equation the three tensors $\mathbf{T}_{\mathrm{K}}, \mathbf{T}_{\mathrm{dis}}, \mathbf{T}_{\mathrm{E}}$ represent respectively the Korteweg elastic stress tensor, the dissipative stress tensor and the Ericksen elastic tensor and $\dot{\boldsymbol{v}} = \frac{\partial \boldsymbol{v}}{\partial t} + (\nabla \boldsymbol{v})\, \boldsymbol{v}$ is the material derivative of $\boldsymbol{v}$. $\mathbf{T}_{\mathrm{K}}$ is expressed as follows

$$\mathbf{T}_{\mathrm{K}} = -p_{\mathrm{K}}\mathbf{I} - \varrho[u_1 \nabla\varrho \otimes \nabla\varrho + u_2(\nabla\varrho \cdot \boldsymbol{n})\nabla\varrho \otimes \boldsymbol{n}], \tag{13}$$

where

$$p_{\mathrm{K}} = p_0(\varrho) - \varrho \operatorname{div}[\varrho\,(u_1 \nabla\varrho + u_2(\nabla\varrho \cdot \boldsymbol{n})\boldsymbol{n})] \tag{14}$$

with

$$p_0(\varrho) := \varrho^2 \frac{d\sigma_0}{d\varrho} \tag{15}$$

an *increasing* function of ϱ, i.e. the *first-grade limit* of the Korteweg pressure field p_{K}. The Ericksen elastic stress tensor is given by

$$\mathbf{T}_{\mathrm{E}} = -K\,(\nabla \boldsymbol{n})^{\mathsf{T}}\,(\nabla \boldsymbol{n}). \tag{16}$$

Finally, the viscous stress tensor is expressed by

$$\begin{aligned}\mathbf{T}_{\mathrm{dis}} =& \frac{1}{2}\gamma_1(\boldsymbol{n} \otimes \mathring{\boldsymbol{n}} - \mathring{\boldsymbol{n}} \otimes \boldsymbol{n}) + \frac{1}{2}\gamma_2(\boldsymbol{n} \otimes \mathbf{D}\boldsymbol{n} - \mathbf{D}\boldsymbol{n} \otimes \boldsymbol{n}) + \frac{1}{2}\gamma_2(\mathring{\boldsymbol{n}} \otimes \boldsymbol{n} + \boldsymbol{n} \otimes \mathring{\boldsymbol{n}}) \\ &+ \frac{1}{2}\gamma_3(\boldsymbol{n} \otimes \mathbf{D}\boldsymbol{n} + \mathbf{D}\boldsymbol{n} \otimes \boldsymbol{n}) + \gamma_4\mathbf{D} + (\gamma_5 \boldsymbol{n} \cdot \mathbf{D}\boldsymbol{n} + \gamma_7 \operatorname{tr}\mathbf{D})\, \boldsymbol{n} \otimes \boldsymbol{n} \\ &+ (\gamma_6 \operatorname{tr}\mathbf{D} + \gamma_7 \boldsymbol{n} \cdot \mathbf{D}\boldsymbol{n})\,\mathbf{I}. \end{aligned} \tag{17}$$

Similarly, the balance of torques is expressed by the dynamical equation (see Refs. [16] and [20]) for the evolution of the director $\boldsymbol{n}$

$$\frac{\partial W_{\mathrm{e}}}{\partial \boldsymbol{n}} - \operatorname{div}\left(\frac{\partial W_{\mathrm{e}}}{\partial \nabla \boldsymbol{n}}\right) + \frac{\partial R_{\mathrm{a}}}{\partial \mathring{\boldsymbol{n}}} + \varrho \frac{\partial \sigma_{\mathrm{K}}}{\partial \boldsymbol{n}} + \mu \boldsymbol{n} = \mathbf{0}, \tag{18}$$

where μ represents a Lagrange multiplier for the constraint $\boldsymbol{n} \cdot \boldsymbol{n} = 1$. To the above balance equations, the mass continuity equation has to be supplemented, as follows

$$\frac{\partial \varrho}{\partial t} + \operatorname{div}(\varrho \boldsymbol{v}) = 0. \tag{19}$$

In order to obtain a system of pure equations, with no Lagrange multiplier, we cross by $\boldsymbol{n}$ Eq. (18), thus obtaining

$$\left[\frac{\partial W_{\mathrm{e}}}{\partial \boldsymbol{n}} - \operatorname{div}\left(\frac{\partial W_{\mathrm{e}}}{\partial \nabla \boldsymbol{n}}\right)\right] \times \boldsymbol{n} + \frac{\partial R_{\mathrm{a}}}{\partial \mathring{\boldsymbol{n}}} \times \boldsymbol{n} + \varrho \frac{\partial \sigma_{\mathrm{K}}}{\partial \boldsymbol{n}} \times \boldsymbol{n} = \mathbf{0}. \tag{20}$$

Equation (20) expresses the balance of three torques: the elastic torque arising from the curvature density energy W_{e}, the *viscous torque* arising from the dissipation function R_{a} and the *acoustic torque* which determines the action exerted by acoustic fields on the nematic director by means of the interaction density energy σ_{K}.

By using the constitutive equations (5), (7) and (10) for σ_K, W_e and R_a we further simplify Eq. (20) and arrive at the following form

$$\gamma_1 \left[\frac{\partial \boldsymbol{n}}{\partial t} + (\nabla \boldsymbol{n})\, \boldsymbol{v} \right] \times \boldsymbol{n} + [\gamma_2 \mathbf{D}\boldsymbol{n} - \gamma_1 \mathbf{W}\boldsymbol{n}] \times \boldsymbol{n} - K\, \mathrm{div}\,(\nabla \boldsymbol{n}) \times \boldsymbol{n}$$
$$+ u_2 \varrho\, (\nabla \varrho \cdot \boldsymbol{n})\, \nabla \varrho \times \boldsymbol{n} = 0 \tag{21}$$

where we can now clearly recognize the three competing torques: the elastic one signified by the elastic constant K, the viscous one related to the viscosities γ_1 and γ_2 and the acoustic one proportional to the susceptibility u_2. Hence, we can summarize the complete system of governing balance equations as follows:

$$\frac{\partial \varrho}{\partial t} + \mathrm{div}\,(\varrho \boldsymbol{v}) = 0 \tag{22}$$

$$\varrho \dot{\boldsymbol{v}} = \mathrm{div}\,(\mathbf{T}_K + \mathbf{T}_{dis} + \mathbf{T}_E + \mathrm{W_e I}) \tag{23}$$

$$\gamma_1 \left[\frac{\partial \boldsymbol{n}}{\partial t} + (\nabla \boldsymbol{n})\, \boldsymbol{v} \right] \times \boldsymbol{n} + [\gamma_2 \mathbf{D}\boldsymbol{n} - \gamma_1 \mathbf{W}\boldsymbol{n}] \times \boldsymbol{n} - K\, \mathrm{div}\,(\nabla \boldsymbol{n}) \times \boldsymbol{n}$$
$$+ u_2 \varrho\, (\nabla \varrho \cdot \boldsymbol{n})\, \nabla \varrho \times \boldsymbol{n} = 0. \tag{24}$$

where $\mathbf{T}_K$, $\mathbf{T}_{dis}$, $\mathbf{T}_E$ are as above.

III. Plane Wave Solution

We need to solve the above balance equations and we seek for plane wave solutions, so we study the propagation of forced infinitesimal harmonic plane waves with wave vector $\boldsymbol{k}$ and angular frequency ω. The linearized balance laws resulting from (22), (23), and (24) and the constitutive relations in (13), (16), and (17) are solved and used to find the anisotropic dispersion of waves and to study the relationship between energy dissipation and wave attenuation. Solutions are sought in the plane wave form:

$$\varrho(\boldsymbol{x}, t) = \varrho_0\, (1 + s_0 \Re E)\,, \quad \boldsymbol{v}(\boldsymbol{x}, t) = s_0 \Re(E\boldsymbol{a}), \tag{25}$$

where $E := \mathrm{e}^{\mathrm{i}(\boldsymbol{k}\cdot\boldsymbol{x} - \omega t)}$, $\Re$ denotes the real part of a complex number, $\boldsymbol{x}$ is the position vector, ϱ_0 is the unperturbed mass density and s_0 a small dimensionless parameter measuring the scale of perturbation, $\boldsymbol{k}$ is the *complex* wave vector to be determined in terms of the angular frequency ω, and $\boldsymbol{a}$ is an unknown complex amplitude vector. We also allow for a director *libration* described by

$$\boldsymbol{n} = [\mathbf{I} + s_0 \Re(E\mathbf{A})]\, \boldsymbol{n}^*, \tag{26}$$

where $\mathbf{A}$ is a complex skew-symmetric tensor and $\boldsymbol{n}^*$ is a uniform unperturbed director field. By *libration* we mean here a motion in which the director keeps a nearly uniform orientation $\boldsymbol{n}^*$ and vibrates about it as a consequence of the flow perturbation. The basic governing equations are solved within the above class of flows, in the limit where s_0 is a small perturbation parameter.

To this end, we let $\boldsymbol{k}$ and $\boldsymbol{a}$ be represented as

$$\boldsymbol{k} = k\boldsymbol{e} \quad \text{and} \quad \boldsymbol{a} = a_e \boldsymbol{e} + a_n \boldsymbol{n}^*, \quad a_n,\ a_e,\ k \in \mathbb{C}, \tag{27}$$

where the unit vector $\boldsymbol{e}$ designates the propagation direction and k, a_e and a_n are all complex numbers to be determined. The imaginary part k_2 of k is associated with the attenuation of the wave: $1/k_2$ represents the attenuation length, that is, the length over which the wave amplitude is reduced by the factor $1/\mathrm{e}$. The linearized (in s_0) mass continuity equation (22), by (27), takes the form

$$ka_e + ka_n \cos\beta = \omega, \quad \text{with} \quad \cos\beta := \boldsymbol{e} \cdot \boldsymbol{n}^*. \tag{28}$$

It follows from (27) and (28) that, whenever $\sin\beta = 0$, a_e and a_n are not uniquely defined; we resolve this ambiguity by setting $a_n = 0$ for $\sin\beta = 0$.

Let us now consider the balance equation of torques (24) and the above ansatz (26) on the librational motion of the director. At the lowest order with respect to s_0 and under the assumption of uniform and constant texture $\boldsymbol{n}^*$, equation (24) becomes

$$[K(\boldsymbol{k}\cdot\boldsymbol{k}) - \mathrm{i}\gamma_1\omega]\left(\mathbf{A}\boldsymbol{n}^*\right) \times \boldsymbol{n}^* + \frac{1}{2}\mathrm{i}(\gamma_2 - \gamma_1)\left(\boldsymbol{k}\cdot\boldsymbol{n}^*\right)\boldsymbol{a} \times \boldsymbol{n}^* + \frac{1}{2}\mathrm{i}(\gamma_2 + \gamma_1)\left(\boldsymbol{a}\cdot\boldsymbol{n}^*\right)\boldsymbol{k} \times \boldsymbol{n}^* = \mathbf{0}. \tag{29}$$

Thus, as shown by (29), the viscous torque exerted by the acoustic flow entrains a director vibration. Notice that at the lowest order of approximation, corresponding to the acoustic dynamics, no contribution from the acoustic torque, i.e. the term accompanied by the susceptibility u_2 in (24), enters the libration equation (29). In other words, at the acoustic time scales the contribution from the interaction density energy σ_{K} is washed away as far as the balance of torques is concerned. The acoustic torque is actually of second–order with respect to s_0 and manifests itself at times longer than the acoustic period through a time-averaged action similar in character to an *acoustic streaming*.

Equation (29) can be easily solved for the libration motion as follows

$$\mathbf{A}\boldsymbol{n}^* = -\Sigma\left\{\gamma_2(\boldsymbol{k}\cdot\boldsymbol{n}^*)(\boldsymbol{a}\cdot\boldsymbol{n}^*)\boldsymbol{n}^* + \frac{1}{2}(\gamma_1 - \gamma_2)(\boldsymbol{k}\cdot\boldsymbol{n}^*)\boldsymbol{a} - \frac{1}{2}(\gamma_1 + \gamma_2)(\boldsymbol{a}\cdot\boldsymbol{n}^*)\boldsymbol{k}\right\}, \tag{30}$$

where

$$\Sigma := \frac{1}{\gamma_1\omega + \mathrm{i}Kk^2}, \quad k^2 = \boldsymbol{k}\cdot\boldsymbol{k}.$$

Accordingly, $\mathbf{A}\boldsymbol{n}^* \cdot \boldsymbol{n}^* = 0$, being the libration motion a slight perturbation around $\boldsymbol{n}^*$ that complies with the requirement of keeping $\boldsymbol{n}$ a unit vector within the linear approximation in s_0.

In the limit as both viscosities γ_1 and γ_2 vanish, so does $\mathbf{A}\boldsymbol{n}^*$, this implying by (26) that no director libration occurs in that limit and a wave can propagate while $\boldsymbol{n}$ remains immobile, even in the absence of any external restraining field. In brief, one could also say that the director libration is a motion fed by dissipation. Hereafter we shall assume that $\gamma_1 > 0$.

The explicit solution (30) for the librational motion of the director is to be used in order to form the equation for the wave propagation, i. e. the linearized (in s_0) version of the linear momentum balance equation (23), and solve it accordingly. The details of this computation are reported in [21]. Here we just focus on the results of this linear analysis.

Were $\Sigma = 0$, there would be no director libration and all the computations would reduce to those ones in [16] obtained in the absence of director libration. What makes the wave propagation problem more difficult to solve here is the way Σ depends on the unknown k^2. Here we assume that

$$\gamma_1 \omega \gg K|k^2|, \tag{31}$$

so that $\Sigma\omega$ can be approximated by $1/\gamma_1$. Physically, this approximation amounts to disregarding the elastic torque in the balance equation (24). Moreover, since $\boldsymbol{n}^*$ is assumed to be uniform in space, it follows from (16) that $\mathbf{T}_{\mathrm{E}}$ is $o(s_0)$, and so at the lowest approximation in s_0 the elastic stress does not contribute to the balance of linear momentum. Elastic effects thus disappear from the balances of both linear momentum and torque, though for different reasons. In Sec. A, we shall derive the *upper* bound to be imposed on ω to make (31) compatible with the solution to the propagation equation. As in [16], we also consider the limit of the propagation equation where all viscosities are small. More precisely, we assume that there is a small dimensionless parameter ε_0 such that

$$\gamma_i = \varrho_0 \frac{c_0^2}{\omega} O(\varepsilon_0), \qquad i = 1, \dots, 7 \tag{32}$$

and we further seek solutions of the linear momentum balance equation (23) such that

$$k_2 = \frac{\omega}{c_0} O(\varepsilon_0), \quad a_n = c_0\, O(\varepsilon_0), \tag{33}$$

where $c_0(\varrho_0) := \sqrt{p_0'(\varrho_0)}$. The validity of (32) requires that ω does not exceed an upper bound that will be discussed in Sec. A along with the one that makes (31) compatible.

Under assumptions (31), (32), and (33) we finally arrive at the following solution of the balance equations (22), (23) and (24) in the linearized setting (in s_0) and at the lowest order of approximation in ε_0:

$$k = \frac{\omega}{c} + \mathrm{i}k_2, \tag{34a}$$

$$k_2 = \frac{\omega^2}{2\varrho_0 c_0^3} \frac{1}{\frac{c}{c_0} + \frac{1}{2}\omega^2\tau^2\frac{c_0}{c}} \left[\gamma_4 + \gamma_6 + \left(\gamma_3 + 2\gamma_7 - \frac{\gamma_2^2}{\gamma_1}\right)\cos^2\beta + \left(\gamma_5 + \frac{\gamma_2^2}{\gamma_1}\right)\cos^4\beta\right], \tag{34b}$$

$$a_n = -\frac{\mathrm{i}}{2}\frac{\cos\beta}{\sin^2\beta}\frac{\omega}{c\varrho_0}\left[\gamma_2 + \gamma_3 + 2\gamma_7 - \left(\gamma_3 - 2\gamma_5 + 2\gamma_7 - 3\frac{\gamma_2^2}{\gamma_1}\right)\cos^2\beta - 2\left(\gamma_5 + \frac{\gamma_2^2}{\gamma_1}\right)\right.$$
$$\left.\cos^4\beta - \frac{\gamma_2(\gamma_1+\gamma_2)}{\gamma_1}\right] \quad \text{for } \sin\beta \neq 0, \tag{34c}$$

$$a_e = c - \mathrm{i}\frac{c^2}{\omega}k_2 - a_n\cos\beta. \tag{34d}$$

Here c is the velocity of sound along $\boldsymbol{e}$, which depends on both ω and β through the equation

$$\frac{c}{c_0} = \frac{\omega\tau}{\sqrt{2(\sqrt{1+\omega^2\tau^2}-1)}}, \tag{35}$$

where τ is the *anisotropic* characteristic time defined by

$$\tau := 2\frac{\varrho_0}{c_0^2}\sqrt{u_1 + u_2\cos^2\beta}. \tag{36}$$

The same expression (35) for c was obtained in [16] in the absence of director libration, while the expressions for k_2, a_n, and a_e above would formally reduce to the corresponding ones found in [16] in the limit as $\gamma_1/\gamma_2 \to \infty$, where the director libration would be hampered by an arbitrarily large rotational viscosity γ_1. We record here for future reference the limiting expression of k_2 in the absence of libration:

$$k_2^\infty = \frac{\omega^2}{2\varrho_0 c_0^3}\frac{1}{\frac{c}{c_0} + \frac{1}{2}\omega^2\tau^2\frac{c_0}{c}}[\gamma_4 + \gamma_6 + (\gamma_3 + 2\gamma_7)\cos^2\beta + \gamma_5\cos^4\beta]. \tag{37}$$

As already pointed out in [16] for k_2^∞, since c in (35) is a function of ω, k_2 in (34b) does not depend on ω in a purely quadratic fashion, as in earlier theoretical studies on wave propagation in nematic liquid crystals [22–25]. Such a non-quadratic dependence is a characteristic signature of our assumption on the Korteweg nature of the acoustic coupling; it will be quantitatively compared in Sec. IV with the available experimental data. It is also possible to show that both k_2 and k_2^∞ are not negative whenever the dissipation inequalities (11) are satisfied [21].

A. Admissible Frequency Ranges

Several simplifying assumptions have been made to arrive at (34); here we identify the ranges where to choose the angular frequency ω of the propagating wave to make these assumptions admissible.

First, we identify the values of ω that make ε_0 in (32) a small parameter. Estimating from [26] the velocity of sound $c_0 = 1.3 \times 10^3\,\mathrm{m\,s^{-1}}$, from [27] (p. 231) the average viscosity $\gamma = 10^{-1}\,\mathrm{Pa\,s}$, and from [28] the mass density $\varrho_0 = 10^3\,\mathrm{Kg\,m^{-3}}$, one easily sees from (32) that $\varepsilon_0 \ll 1$ whenever $\omega \ll \omega_\gamma$, with

$$\omega_\gamma := \frac{c_0^2\varrho_0}{\gamma} = 0.8 \times 10^{10}\,\mathrm{s^{-1}} \sim 10^4\,\mathrm{MHz}.$$

Second, we identify the angular frequencies that make the solution (34) of the propagation equations compatible with the assumption (31). To this end, we recall from [16] that $c \sim c_0$ for $\omega\tau < 10$, and so, since by (34a) $k \sim \omega/c$, estimating γ_1 as 2γ and taking $K \sim 10^{-11}\,\mathrm{N}$ from [27] (p. 103), we see that (31) is satisfied for the solution (34) whenever $\omega \ll \omega_K$, with

$$\omega_K := \frac{c_0^2\gamma_1}{K} = 3.38 \times 10^{16}\,\mathrm{s^{-1}} \sim 10^{10}\,\mathrm{MHz}.$$

It is clear from the estimates above that the largest upper bound on ω for the validity of our theory is ω_K. The most stringent bound on ω is thus ω_γ; it will follow from the estimate of τ in Sec. IV that this easily complies with the requirement that $\omega\tau < 10$.

IV. Estimate of Phenomenological Parameters

Using data published in the literature for N-p-methoxybenzylidine-p'-n-butylaniline (MBBA), numerical calculations for the attenuation and dispersion are given in this section and compared to acoustic experiments.

In order to fit the experimental data available in literature (see Refs. [4], [13]), we introduce preliminarly and following Refs. [16] and [13] a measure of anisotropy in the speed of sound different from (35). We define the relative sound speed anisotropy Δc as

$$\Delta c := \frac{c - c|_{\beta=\frac{\pi}{2}}}{c|_{\beta=0}}. \tag{38}$$

Δc is a function of both ω and β, which vanishes for $\beta = \frac{\pi}{2}$. By assuming that

$$\varepsilon := \frac{u_2}{u_1}$$

is a small parameter, we can distinguish in Δc the dependence on ω from the dependence on β. Then, by (35), (38) yields

$$\Delta c = \varepsilon f(\omega\tau_1)\cos^2\beta + O(\varepsilon^2), \tag{39}$$

where

$$\tau_1 := 2\frac{\varrho_0}{c_0^2}\sqrt{u_1}, \qquad f(x) := \frac{1}{4}\frac{x^2 - 2(\sqrt{1+x^2}-1)}{\sqrt{1+x^2}(\sqrt{1+x^2}-1)}. \tag{40}$$

Notice that f is positive and strictly increasing function, so that the speed of propagation along the nematic director $\boldsymbol{n}^*$ is larger than the speed of propagation at right angles to it whenever $\varepsilon > 0$. We also introduce the following adimensional quantity as a measure of the wave attenuation (still depending on both ω and β)

$$\frac{\Delta k_2}{\Delta k_2|_{\beta=\frac{\pi}{2}}} = \frac{k_2 - k_2|_{\beta=0}}{k_2|_{\beta=\frac{\pi}{2}} - k_2|_{\beta=0}}. \tag{41}$$

In the limit of small ε Eq. (41) gives rise to

$$\frac{\Delta k_2}{\Delta k_2|_{\beta=\frac{\pi}{2}}} = -G(\beta) + O(\varepsilon), \tag{42}$$

where

$$G(\beta) := -\sin^2\beta\left(1 + \frac{\gamma_1\gamma_5 + \gamma_2^2}{\gamma_1(\gamma_3+\gamma_5+2\gamma_7)}\cos^2\beta\right), \tag{43}$$

which illuminates the dependence on the direction of propagation β. At the same time we take the dependence on ω by considering Eq. (41) for $\beta = \frac{\pi}{2}$ and taking a reference frequency ω_0

$$\frac{\Delta k_2|_{\beta=\frac{\pi}{2}}}{\left[\,\Delta k_2|_{\beta=\frac{\pi}{2}}\,\right]_{\omega=\omega_0}} = \frac{k_2|_{\beta=\frac{\pi}{2}} - k_2|_{\beta=0}}{\left[\,k_2|_{\beta=\frac{\pi}{2}} - k_2|_{\beta=0}\,\right]_{\omega=\omega_0}} =: S(\varepsilon, \omega; \omega_0). \tag{44}$$

In the limit of small ε, we get

$$S(\varepsilon, \omega; \omega_0) = Z(\omega; \omega_0) + O(\varepsilon) \tag{45}$$

where

$$Z(\omega; \omega_0) := \left(\frac{\omega}{\omega_0}\right) \sqrt{\frac{\sqrt{1+\tau_1^2\omega^2}-1}{\sqrt{1+\tau_1^2\omega_0^2}-1}} \sqrt{\frac{1+\tau_1^2\omega_0^2}{1+\tau_1^2\omega^2}}. \tag{46}$$

It is remarkable noticing that Z, unlike G, does not depend on the viscosities and just depends on a single phenomenological parameter, i.e. u_1, via τ_1. Now we shall be using formulae (38), (39), (42) and (46) in order to fit the data corresponding to the observations of [13] for *p*-*n*-butyl-aniline (MBBA) at 21°C and wave frequency 10 MHz under the action of an aligning magnetic field with strength 5 Oe, and to the observations of [4] in the range of frequencies 2–6 MHz.

We assume that the phenomenological parameters do not depend on the working frequency. Moreover we consider the limit of $\varepsilon = \frac{u_2}{u_1} \ll 1$. This latter is meant to be an assumption and it is found to be selfconsistent with the available data ([4] and [13]). We took the data from the Fig. 2 of [13] as far as the angular dependence of sound velocity is concerned and Figs. 1, 2 of [4] for the dependence of change in attenuation on angle and frequency, respectively.

First, we consider Fig. 2 of Ref. [4] which exhibits the dependence of the attenuation on the frequencies in the range 2–6 MHz. To fit these data we use formula (46), which is valid for small ε and we use as reference frequency $\frac{\omega_0}{2\pi} = 6$ MHz which turns out to be the highest value of frequencies employed in Ref. [4]. By using the built-in function `FindFit` in MATHEMATICA [29] for least-squares fit, we get the following value for the only parameter in the Z function

$$\tau_1 = 3.47 \times 10^{-8}\text{s}. \tag{47}$$

Using this value for the parameter τ_1, we compute the value of $\tau_1\omega$ at the frequency $\frac{\omega}{2\pi} = \frac{\omega_1}{2\pi} = 10$ MHz, the one used in Ref. [13]

$$\tau_1\omega_1 = 2.18. \tag{48}$$

We now plug this value in the exact formula (38) to validate our assumption about the smallness of ε and by fitting the data of Fig. 2 of Ref. [13] about the angular dependence of sound velocity we finally arrive at

$$\varepsilon = 7.74 \times 10^{-3}. \tag{49}$$

Thus, we find a small value of ε which makes selfconsistent the apriori assumption made for the fit of (46) within the two independent sources of data, i.e. Ref. [13] and Ref. [4]. Moreover a direct fit of data in Fig. 2 of Ref. [13] about the angular dependence of sound velocity, as also suggested by the authors of Ref. [13] with the most reasonable function (the one used by the authors of [13])

$$f_0(\beta) := A\cos^2\beta, \tag{50}$$

gives rise to the following value of fitted A

$$A = 11.25 \times 10^{-4}. \tag{51}$$

Notice that the value found in Ref. [13] is 12×10^{-4} by using direct data and it is consistent with our value. On the other hand, since the fitted value of $\varepsilon = 7.74 \times 10^{-3}$ justifies our assumption of smallness of ε, then we are allowed to use the approximate formula (39) for the anisotropy of sound velocity and accordingly we find a value of the amplitude of $\cos^2 \beta$ as follows

$$\begin{aligned} A \to \varepsilon f(\tau_1\omega_1) &= 7.74 \times 10^{-3} f(2.18) \\ &= 11.28 \times 10^{-4} \end{aligned} \tag{52}$$

which turns out to be in very good agreement with the direct fit with the function $f_0(\beta)$. Finally we can use formula (42) in order to find the value of the viscosity γ_7 using for the remaining ones appearing in the expression of G the standard values for MBBA (see [27] pag. 229–231). We arrive at

$$\gamma_7 = 1.58 \times 10^{-1}\,\mathrm{Pa\,s}. \tag{53}$$

It is worth noticing that only γ_7 can be estimated, while no value for γ_6 is available yet. Moreover, the numerical value for γ_7 turns out to be bigger than the other viscosities (see [27] pag. 229–231) and this is to be due to the smaller values of $\mathrm{tr}\,\mathbf{D}$, the invariant connected with the compressibility of the nematics and which accompanies the viscosities γ_6 and γ_7 in the dissipation function R_a (10).

V. Conclusions

In this paper we briefly introduced and analyzed a variational theory recently proposed in [16] on the interaction of acoustic fields and the nematic texture of a liquid crystal. This theory embodies the heart of the nematoacoustic interaction by an elastic additional energy of Korteweg type which is characterized by two main phenomenological constants: u_1 and u_2 in (5). In order to estimate these constitutive coefficients, we employed old experimental data available in literature against some consequences of the theory such as the anisotropy and dispersion in sound speed, and the non-quadratic frequency dependence of wave attenuation. We have also estimated one of the two viscosities (γ_7) usually absent when a constraint of incompressibility is present. The determination of such a viscosity using acoustic probe is not fortuitous as the very possibility of sound propagation in liquid crystals resides in their being compressible. The comparison of the available experimental data with our predictions points out the need of further experimental investigations for the determination of the still unknown viscosity γ_6 and the susceptibilities u_1 and u_2 for other liquid crystalline materials.

We treated the problem of solving the balance equations, stemming from the theory, in the linear approximation which is the most appropriate for acoustic propagation in the nematic medium. In particular, we sought for solutions to the balance dynamical equations in the form of planar waves and we extended this ansatz to the velocity field, the mass density and the director field, thus also allowing for librational motion of the nematic texture. The immobility and uniformity constraint of the director, assumed in [16], has been removed and our propagation equations in Sec. III were derived in the general case when the director is free to vary in time and be distorted in space as a consequence of a plane wave propagating through the nematic texture. The director can freely rotate slightly back and forth around a fixed direction. On the acoustic characteristic times and lengths this is the most reasonable and general motion of the director. Moreover in this fast (vibrational) regime elastic stresses do not affect the motion: actually, as for the linear momentum equation, the elastic tensor (Ericksen tensor) is second–order with respect to the perturbation parameter (see Eq. (16)). As for torque balance equation (see Eqs. (21) and (29)), we assumed that the elastic torque is negligible with respect to viscous torques. We argued that at the time scale of acoustic vibrations the elastic torque arising from the contribution W_e to the free energy does not affect the corresponding balance equation. We found an upper limit of frequencies at which this should happen. We computed accordingly the mass density and velocity modulations as well as libration of the director. At the acoustic scales, the acoustic torque does not enter the libration equation (29) at all, as it is of second-order with respect to the perturbation parameter (see Eqs. (21) and (29)). Elastic and acoustic torques as well as elastic stresses actually act at scales other than the acoustic ones along with elastic *streaming* sources taken as time average of second–order acoustic fields. Other variations of the director texture can then take place at time and length scales much larger than the acoustic characteristic times and lengths.

At the same time, though no macroscopic, hydrodynamic flow is percieved at the time scale of the acoustic vibrations, at longer times even initially stagnant nematic fluid may develop steady flows by *acoustic streaming* [30–34]. We actually imagine to distinguish a fast (acoustic time scales) and a slow dynamics, the former evolving as if the latter were not, this latter being influenced only by the time average of the other. The fast dynamics provides time–averaged sources that bear a mechanical meaning. The slow dynamics in turn may drive the background against which the fast dynamics is taking place. In particular, acoustic dynamics generates a time–averaged acoustic torque on the nematic director which is absent on the acoustic time scales (see Eq. (29)) but affects the slow director dynamics; this will eventually alter the wave propagation. All the second–order slow evolution and distortions of the velocity and director fields have to be taken into account and appropriate balance equations have to be formed: all this pertains to future work ([35]).

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