ELECTRO-ACOUSTIC COUPLINGS IN NEMATIC LIQUID CRYSTALS

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Summary Two intriguing effects—acoustic realignment and acoustic generation—due to electro-acoustic couplings in nematic liquid crystals are theoretically elucidated. As regards the first, an existing theory is criticized and a new simpler theory is proposed. As regards the latter, a theoretical explanation is provided for the first time.

INTRODUCTION

Liquid crystals (LCs) are mesophases that have properties between those of a conventional liquid and those of a solid crystal: LCs may flow like a liquid, but their molecules may be oriented in a crystal-like way. There exist different LC phases, characterized by the type of ordering. One of the most common of them is the nematic phase, where rod-shaped molecules with no positional order self-align to have long-range directional order. Nematics have fluidity similar to that of ordinary (isotropic) liquids, but they can be easily aligned by an external magnetic or electric field. For this reason, NLCs exhibit plenty of coupled phenomena, both scientifically rich and technologically significant. The best known and most studied of them is the electro-optical coupling, as a result of the large consumer demand generated by the display industry. Less widely known is the acousto-optical effect, by which a low-intensity acoustic field realigns the molecules, leading to a change in the optical transmission through the NLC cell. Even less studied—but not less interesting—is the phenomenon of acoustic generation, in which sound is emitted by the NLC cell during the orientational switching triggered by an applied electrical signal. In this short paper, I will focus on the two coupling effects involving acoustics, namely, acoustic realignment and acoustic generation.

ACOUSTIC REALIGNMENT

The acousto-optical effect was extensively studied by J.V. Selinger and coworkers [1, 2, 3, 4], both experimentally and theoretically. Their model hinges on the idea of a director–density coupling, mathematically represented (in my own notations) by the following assumption for the interaction energy-density per unit reference volume:

$$w_{\rm I} = \frac{1}{2}\alpha\big((\nabla(\operatorname{tr}\mathbf{E}))\cdot\mathbf{n}\big)^2, \tag{1}$$

where $\mathbf{E}$ is the (small) deformation from a reference configuration with constant density ρ_0 and uniform director field, $\mathbf{n}$ is the actual director field, and α is a scalar coefficient measuring the strength of the coupling between the director $\mathbf{n}$ and the density gradient $\nabla\rho$ (because of mass conservation, $\nabla\rho = -\rho_0\nabla(\operatorname{tr}\mathbf{E})$). Since $\mathbf{E}$ is the symmetrized gradient of the (small) displacement, eq. (1) patently establishes a second-grade theory, later fully developed by Virga and DeMatteis [5, 6].

Overall, predictions from this anisotropic Korteweg-like theory agree very well with available experimental data. However, the above authors implicitly dismiss the hypothesis that a satisfactory model of anisotropic acoustics can be obtained with a simpler, i.e., first-grade theory, such as the one based on the alternative assumption

$$w_{\rm I} = a\,\mathbf{n}\cdot\big((\operatorname{dev}\mathbf{E})\,\mathbf{n}\big)\operatorname{tr}\mathbf{E}\,, \tag{2}$$

where $\operatorname{dev}\mathbf{E}$ is the deviatoric strain and a is a scalar coefficient, analogous to α in eq. (1)—but be it noticed that their physical dimensions differ by a length squared.

To compare predictions stemming from eqs. (1) and (2), let us consider how the sound speed c depends on the wave vector $\mathbf{k}$ and the director $\mathbf{n}$. One obtains

$$c^2 = \text{const.} + (\alpha/\rho_0)(\mathbf{k}\cdot\mathbf{n})^2 \tag{3}$$

from eq. (1), and

$$c^2 = \text{const.} + (a/\rho_0)(\mathbf{k}\cdot\mathbf{n}/|\mathbf{k}|)^2 \tag{4}$$

from eq. (2). Since the effective moduli α and a are not susceptible of direct measurement, predictions (3) and (4) essentially coincide for a given wave number $|\mathbf{k}|$. Under the reasonable assumption that neither α nor a depend (strongly) on frequency, it should be easy—in principle—to disprove either eq. (1) or (2). However, all experiments by J.V. Selinger and coworkers have seemingly been done at one and the same frequency, namely, 3.3 MHz.

As a provisional conclusion, I would affirm that until experimental evidence discriminates hypothesis (1) from hypothesis (2), the lower-grade theory put forth here should be preferred to Selinger-Virga's theory on methodological grounds.

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ACOUSTIC GENERATION

In 1999 Y.J. Kim and J.S. Patel [7] experimentally demonstrated that a faint, but audible, acoustic signal is generated by the Fréedericksz transition—i.e., the sudden switch from an undistorted to a distorted director field—triggered in a nematic liquid-crystal (NLC) cell by an electrical pulse. On the explanatory side, these authors just "speculate that the mechanism of excitation is closely related to the reorientation of the molecules, which produces a transient thickness change of the liquid-crystal container." To the best of my knowledge, no further study—either theoretical or experimental—has been published in the intervening years.

Building on the raw conjecture by Kim and Patel, in this short paper I put forth a precise, computable mathematical model of the phenomenon of acoustic generation in NLCs, whose numerical predictions are in excellent agreement with their experimental findings. The main underlying hypothesis of my model is that the long rod-like molecules comprising the nematic mesophase are more or less densely packed depending on their relative orientation. More precisely, I assume that the pressure at a point depends (strongly) on the density and (in a much weaker way) on the director gradient at that point. This assumption is embodied in an energy-density functional that (weakly) couples acoustics with nematic elasticity. In turn, nematic elasticity is coupled with electrostatics in the standard way [8, 9, 10]. No direct coupling between acoustics and electrostatics is assumed. Namely, I posit that the NLC free-energy per unit volume splits into the sum of three contributions: $\psi(\varrho,\mathbf{n},\nabla\mathbf{n},\nabla\varphi)=\psi_{\mathsf{A}}(\varrho,\mathbf{n},\nabla\mathbf{n})+\psi_{\mathsf{F}}(\mathbf{n},\nabla\mathbf{n})+\psi_{\mathsf{E}}(\mathbf{n},\nabla\varphi)$, where ϱ is the mass density, $\mathbf{n}$ the nematic director field, $\nabla\mathbf{n}$ its gradient and $\nabla\varphi$ the gradient of the electric potential φ (all of them evaluated at the place and time considered). The Frank-Oseen energy density

$$\psi_{\mathsf{F}}(\mathbf{n},\nabla\mathbf{n})=\frac{\kappa_1}{2}(\operatorname{div}\mathbf{n})^2+\frac{\kappa_2}{2}(\mathbf{n}\cdot\operatorname{rot}\mathbf{n})^2+\frac{\kappa_3}{2}(\mathbf{n}\times\operatorname{rot}\mathbf{n})^2+\frac{\kappa_2+\kappa_4}{2}\big((\nabla\mathbf{n})^{\top}\cdot\nabla\mathbf{n}-(\operatorname{div}\mathbf{n})^2\big)\tag{5}$$

and the electrostatic energy density

$$\psi_{\mathsf{E}}(\mathbf{n},\nabla\varphi)=-\frac{\varepsilon_{\perp}}{2}\nabla\varphi\cdot\nabla\varphi-\frac{\varepsilon_a}{2}(\nabla\varphi\cdot\mathbf{n})^2\tag{6}$$

are standard, while the acoustic energy density

$$\psi_{\mathsf{A}}(\varrho,\mathbf{n},\nabla\mathbf{n})=\frac{k}{2}\Big((\varrho-\varrho_0)/\varrho_0+\beta(\mathbf{n},\nabla\mathbf{n})\Big)^2\tag{7}$$

is original. Divergence and curl are distinguished projections of the gradient: $\operatorname{div}\mathbf{n}=\mathbf{I}\cdot\nabla\mathbf{n}$, $(\operatorname{rot}\mathbf{n})\times\mathbf{r}=(\operatorname{skw}\nabla\mathbf{n})\,\mathbf{r}$ for all vector $\mathbf{r}$; κ_1, κ_2, κ_3 and $\kappa_2+\kappa_4$ are the Frank elastic moduli (splay, twist, bend and saddle-splay, respectively); $\varepsilon_{\perp}$ and ε_a are the transverse and anisotropic permittivities; ϱ_0 is the unperturbed mass density and k the (large) bulk modulus; the effect of the nematic curvature on density is parameterized by the (small) dilational moduli δ_1, δ_2, δ_3 and $\delta_2+\delta_4$ (splay, twist, bend and saddle-splay, respectively):

$$\beta(\mathbf{n},\nabla\mathbf{n})=\delta_1(\operatorname{div}\mathbf{n})^2+\delta_2(\mathbf{n}\cdot\operatorname{rot}\mathbf{n})^2+\delta_3(\mathbf{n}\times\operatorname{rot}\mathbf{n})^2+(\delta_2+\delta_4)\big((\nabla\mathbf{n})^{\top}\cdot\nabla\mathbf{n}-(\operatorname{div}\mathbf{n})^2\big).\tag{8}$$

The formal identity between (5) and (8) is imposed by invariance requirements. In fact, they both represent the lowest-order nontrivial scalar-valued polynomial in $\nabla\mathbf{n}$ invariant under change of observer and director reversal. The kinetic energy density is assumed to equal $\frac{1}{2}\varrho\,\mathbf{v}\cdot\mathbf{v}$, with $\mathbf{v}$ the translational velocity, rotational inertia effects being disregarded. Viscosity effects are neglected altogether.

CONCLUSIONS

It is interesting to observe that the mechanisms underlying the phenomena of acoustic realignment and acoustic generation in NLCs are different and altogether independent from one another, despite being both intrinsic to the nematic structure.

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