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Orienting Action of Sound on Nematic Liquid Crystals

W. Helfrich
Physics Department, F. Hoffmann-La Roche & Co., Basle, Switzerland
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A theory is outlined of the aligning action of sound on nematic liquid crystals. It is based on sound-induced stresses and forces and identifies four contributions to the transverse stress. Formulas for the threshold sound intensities of a few simple deformations are given.

Sound is known to generate a radiation pressure[1,2] which leads to a force density in the propagating medium if there is sound absorption. The radiation pressure and other time averages of the second-order stresses are proportional to the sound intensity, in contrast to the first-order stresses which vary as the wave amplitude and whose time averages are always zero. In an isotropic medium neither sound nor shear waves produce second-order forces perpendicular to the wave vector. Transverse second-order forces are possible in anisotropic solids, but are not easily detectable. The situation is different in nematic liquid crystals. They combine anisotropy and fluidity, thus permitting transverse motion.

In the present note we are concerned with the averaged second-order transverse stress $\langle\sigma_{xz}\rangle$. The z axis is parallel to the wave vector, and the x axis is chosen such that the x, z plane contains the nematic axis. We consider only oscillations in the x, z plane. Any motions along the y axis of our Cartesian coordinates are not coupled to motions along the x and z axes or to an incident longitudinal sound wave. A wave traveling through a uniformly oriented liquid crystal may then be expressed to first order by[3]

$$(u_x, u_z) = (u_{x0}, u_{z0}) \exp[i(qz - \omega t)], \tag{1}$$

where u_x and u_z are the transverse and longitudinal displacements, q the wave number which is complex because of absorption, ω the angular frequency, and t the time. The first-order equations of motion, after differentiation, may be written as

$$\rho\omega^2 u_{x0} - \Delta c_{xx} q^2 u_{x0} + i\eta_{xx}\omega q^2 u_{x0} - \Delta c_{xz} q^2 u_{z0} + i\eta_{xz}\omega q^2 u_{z0} = 0, \tag{2}$$

$$\rho\omega^2 u_{z0} - (c + \Delta c_{zz}) q^2 u_{z0} + i\eta_{zz}\omega q^2 u_{z0} - \Delta c_{xz} q^2 u_{x0} + i\eta_{xz}\omega q^2 u_{x0} = 0. \tag{3}$$

Here ρ is the density, c the isotropic hydrodynamic elasticity, the Δc's are frequency-dependent viscoelastic corrections, and the η's are the viscosities [their corrections are neglected as they would lead in (6) to powers of ω higher than the fourth]. The Δc's and η's depend on the alignment angle, all coupling terms being zero for alignment parallel or perpendicular to q.

For weak coupling the two possible waves are almost purely longitudinal and purely transverse. We disregard the latter, mainly on the assumption that it is absorbed within a much shorter distance than the former. The transverse admixture to the longitudinal wave is readily deduced from (2) to be

$$u_{x0} = u_{z0} \frac{\Delta c_{xz}/c - i\eta_{xz}\omega/c}{1 + (\Delta c_{zz} - \Delta c_{xx})/c - i(\eta_{zz} - \eta_{xx})\omega/c}, \tag{4}$$

if one inserts the approximation

$$\rho\omega^2/q^2 = (c + \Delta c_{zz}) - i\eta_{zz}\omega. \tag{5}$$

Assuming that all fractions in (4) are much smaller than unity, we obtain for the transverse second-order stress

$$\langle\sigma_{xz}\rangle = \left[\frac{\eta_{xz}\omega}{c}\frac{(\eta_{zz} - \eta_{xx})\omega}{c} + \frac{\Delta c_{xz}}{c} - \frac{\eta_{xz}\omega}{c}\frac{\rho_0(d\eta_{xx}/d\rho)\omega}{c} + \frac{\eta_{xz}{}^{(2)}\omega^2}{c}\right](-\Pi). \tag{6}$$

From the four terms of (6) we have separated a common factor $-\Pi$. The quantity $\Pi = \rho\langle(\mathrm{Re}v_z)^2\rangle = \frac{1}{2}u_{z0}u_{z0}{}^*\omega^2$ is the longitudinal momentum flux density due to oscillatory mass transport, i.e., the ordinary radiation pressure, with v denoting velocity. (We remark that $-\Pi$ need not be identical to the total second-order stress $\langle\sigma_{zz}\rangle$ in a condensed medium.) The first two terms of (6) represent $-\rho\langle(\mathrm{Re}v_x)(\mathrm{Re}v_z)\rangle$ which may be called negative transverse radiation pressure. Parts of expression (4) for u_{x0} have been dropped in forming $\langle(\mathrm{Re}v_x)(\mathrm{Re}v_z)\rangle$ for reasons to become clear below. The third term is $(d\eta_{xx}/d\rho)\langle(\mathrm{Re}\rho)(\mathrm{Re}dv_x/dz)\rangle$. Being proportional to the density increment and the component of transverse shear in phase with it, it takes account of the net transverse stress which may arise if the viscosity of transverse shear depends on density. The derivative $d\eta_{xx}/d\rho$ is adiabatic, thermal conduction being regarded as negligible. The fourth term is $-\eta_{xz}{}^{(2)}\langle(\mathrm{Re}dv_z/dz)^2\rangle$, the contribution of second-order viscosity $\eta^{(2)}$. To eliminate q in the last two terms we have used $q^2 = \rho\omega^2/c$ instead of the more complicated Eq. (5), which will be justified immediately.

The coefficients in (6) are composed of material constants multiplied by angle-dependent factors. A set of such constants has been formulated for first-order viscosity.[4] An analogous set applies to viscoelasticity, while a new set is needed for second-order viscosity. It is not attempted here to express $\langle\sigma_{xz}\rangle$ in terms of such constants as this would require a much longer paper. Symbolically putting $\eta = k\tau$, $\Delta c = k\omega^2\tau^2$, and $\eta^{(2)} = k\tau^2$, where the k's are elasticities and the τ's Maxwell relaxation times, we see that exactly those terms were retained in (6) that vary as ω^4. They all may be of similar magnitude, as each contains the same powers of coupling k's, noncoupling k's or c's, and τ's. It should be noted that (6) holds only if $\omega \ll 1/\tau$ with respect to all relaxation times.

Let us now discuss the hydrodynamic stability of an infinite nematic layer of constant thickness, taking both the initial alignment and the wave vector of sound to be normal to the bounding walls. Introducing the tilt angle φ, to be positive for tilt of the nematic axis toward $x > 0$, we may write for very small φ

$$\langle\sigma_{xz}\rangle = -\Pi\alpha\varphi \text{ with } |\alpha| = (\eta\omega/c)^2. \tag{7}$$

Here η is a compound quantity having the dimension and possibly the magnitude of a viscosity. If φ varies with z, the stress (7) leads to the transverse force density

$$f_x = d\langle\sigma_{xz}\rangle/dz = -\Pi\alpha\, d\varphi/dz, \tag{8}$$

which may produce a transverse flow. Assuming φ to be a function of z only, one has the following steady-state balances for the force and torque densities:

$$-\Pi\alpha\, d\varphi/dz + \eta_1 d^2\langle v_x\rangle/dz^2 = 0, \tag{9}$$

$$K_{33} d^2\varphi/dz^2 + \kappa_1 d\langle v_x\rangle/dz = 0, \tag{10}$$

where $\langle v\rangle$ is the second-order velocity, K_{33} the curvature-elastic modulus for bend, η_1 a viscosity coefficient, and κ_1 the ratio of torque density to shear rate. (Again the assumption is $|\varphi| \ll \pi/2$.) Combination of the equations yields

$$d^3\varphi/dz^3 = c\, d\varphi/dz, \text{ with } c = -\Pi\alpha\kappa_1/\eta_1 K_{33}. \tag{11}$$

Physically sound solutions are possible only for $c < 0$.

In our geometry there can be no force for $\varphi \equiv 0$,

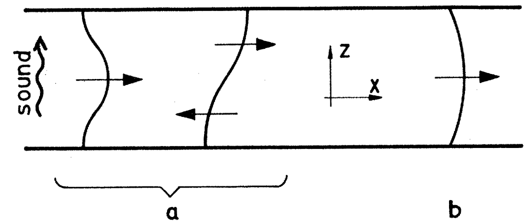

FIG. 1. Representative orientation lines in cases (a) and (b) of text. The arrows indicate the direction of flow.

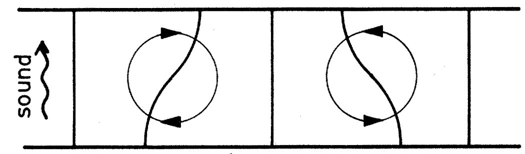

Fig. 2. Schematic orientation pattern associated with vortical flow. The circles indicate vortices.

which suggests that any deformation and the accompanying flow require a threshold sound intensity. For the sake of simplicity we assume the intensity to be practically equal all over the sample. We wish to set up formulas for the threshold, considering two well-defined cases: (a) rigid wall alignment $\varphi(L/2)=\varphi(-L/2)=0$, and (b) virtually loose normal wall alignment, the only restriction being $\varphi(L/2)=-\varphi(-L/2)$. Here L is the thickness of the sample extending from $z=-L/2$ to $z=L/2$. A generally valid boundary condition is, of course, $\langle v_x(L/2)\rangle = \langle v_x(-L/2)\rangle = 0$. Any infinitesimal deformation $\varphi(z)$ may be expanded into independent Fourier components. The uniform orientation pattern is at its instability threshold if the longest wavelength compatible with the boundary conditions is in neutral equilibrium. In case (a) the wavelength is L and the corresponding threshold sound pressure is

$$\Pi_c = (2\pi/L)^2 (\eta_1 K_{33})/(\alpha\kappa_1). \tag{12}$$

There are two deformations with this threshold, as indicated in Fig. 1, which may also occur in linear combinations. In case (b) the maximum possible wavelength is $2L$, leading to a threshold sound pressure

$$\Pi_c = (\pi/L)^2 (\eta_1 K_{33})/(\alpha\kappa_1). \tag{13}$$

There is only one type of solution, shown in Fig. 1, but as in case (a) the direction of flow is arbitrary within the x, y plane. Straight flow of the same direction throughout the sample is of course not possible in many practical cell geometries.

Instead of straight flow, sound might produce vortices, as sketched in Fig. 2. It would seem that instabilities of this kind have higher thresholds than (12) or (13). However, only a detailed study involving a large number of material constants can show which type of deformation is favored at a given sound intensity. Account has to be taken not only of transverse forces, but also of the longitudinal force density $\partial\langle\sigma_{zx}\rangle/\partial x$. Furthermore, different thresholds are to be expected if the initial alignment of the nematic axis is parallel to the walls.

The hydrodynamic instabilities are linked with optical changes which are observable with light transmitted normally through the liquid-crystalline layer. A normally aligned layer appears at first isotropic and turns birefringent under straight flow. Vortical flow should result in domain patterns similar to those seen with electrohydrodynamic instabilities.[5] Birefringence[6] and domains[6,7] have, in fact, been observed under sound action in thin layers of nematic liquid crystals. In one of the experiments[6] the initial alignment was controlled, the nematic axis being normal to the bounding walls.

It is not yet possible to predict threshold intensities from our theory, mainly because of a lack of known material constants. In order to make at least a tentative estimate, we put $\eta = 10$ P in (7). This may be an adequate number to describe the first-order viscous contribution to $\langle\sigma_{xz}\rangle$ of the room-temperature nematic p'-methoxybenzylidene-p-n-butylaniline (MBBA). Its shear viscosities are known to be 1 P and less,[8] but it has been found with other nematics[9] that bulk viscosities exceed shear viscosities by more than a factor of 10. Using (13) and taking $L = 25$ μm, $\eta_1/\kappa_1 = 1$, $K_{33} = 1\times10^{-6}$ dyn, $\omega = 2\pi\times10^7$ sec^{-1}, and $c = 2.5\times10^{10}$ erg cm^{-3}, we obtain

$$\Pi_c = 2.5\times10^3 \text{ dyn cm}^{-2}.$$

With $v_s = 1\times10^5$ cm sec^{-1} for the sound velocity, this results in the threshold intensity

$$I_c = \Pi_c v_s \approx 25 \text{ W cm}^{-2}.$$

This very high value is possibly lowered by the viscoelastic contribution to $\langle\sigma_{xz}\rangle$ as there are indirect indications of a strong viscoelastic effect in MBBA.[10] The experimental thresholds[6,7] at the same thickness and frequency and also at room temperature seem to be of the order of 10^{-2} W cm^{-2}. We suspect that the materials used were more viscous than MBBA,[11] which might help substantially to explain the low intensities. It is of course also possible that the surprisingly strong action of ultrasound is due to the dependence of viscosity on density or to second-order viscosity, both of which are totally unknown for any liquid crystal. Another possibility to generate vortices, but no straight flow, is Bénard's instability in a thermal gradient.[12] The experimental reports[6,7] do not refer to this mechanism which would probably have been easy to recognize (for instance, by very long response times).

In summary, it seems that the proposed mechanism of sound action on nematics is plausible and may provide a means to explore novel material properties. Our high estimate of the threshold is based on fairly unfavorable assumptions. The use of larger viscosities and thicknesses than those inserted above would lower the preliminary theoretical value by orders of magnitude.[13]

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Multiple Soliton Production and the Korteweg–de Vries Equation*

Noah Hershkowitz, Thomas Romesser, and David Montgomery

Department of Physics and Astronomy, University of Iowa, Iowa City, Iowa 52240

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Compressive square-wave pulses are launched in a double-plasma device. Their evolution is interpreted according to the Korteweg–de Vries description of Washimi and Taniuti. Square-wave pulses are an excitation for which an explicit solution of the Schrödinger equation permits an analytical prediction of the number and amplitude of emergent solitons. Bursts of energetic particles (pseudowaves) appear above excitation voltages greater than an electron thermal energy, and may be mistaken for solitons.

A mathematical description of slightly nonlinear, one-dimensional, ion acoustic waves in a collisionless plasma with cold ions has been given by Washimi and Taniuti.[1] The resulting equation for the fractional perturbation of the ambient ion number density is the Korteweg–de Vries equation,[2,3]

$$\frac{\partial}{\partial t}\frac{\delta n}{n_0}+\frac{\delta n}{n_0}\frac{\partial}{\partial x}\frac{\delta n}{n_0}+\frac{1}{2}\frac{\partial^3}{\partial x^3}\frac{\delta n}{n_0}=0, \qquad (1)$$

where $\delta n = \delta n(x,t)$ is the perturbation on the ion number density, and n_0 is the steady-state constant density of the plasma. x is the spatial coordinate measured in units of the electron Debye length λ_D and t is the time measured in units of the inverse ion plasma frequency, ω_{pi}^{-1}. Equation (1) is written in a coordinate frame moving to the right with an ion acoustic speed $c_s = \lambda_D \omega_{pi}$.

Equation (1) has been intensively studied in the last decade.[4-10] In particular, an asymptotic long-time solution has been given[11] which applies to density perturbations $\delta n(x,0)/n_0$ which are pos-