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ACOUSTICAL ACTIVITY IN LIQUID CRYSTALS

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The existence of acoustical activity, analogous to optical activity, is postulated for liquid crystals. It is demonstrated that for cholesterics and the smectic-A phase, as a result of spatial dispersion effects, there exist two acoustically rotated transverse acoustic waves. It is predicted that in the gigahertz region cholesterics will display acoustical rotary power in the neigborhood of 1000 radians/cm.

Experimental investigations[1] of sound propagation in liquid crystals indicate that at certain frequencies (2-10 MHZ), liquid crystals behave like elastic solids. These investigations have demonstrated small anisotropies in the sound velocity, and dispersion of sound waves. It is also well known[1,2] that certain types of liquid crystals, cholesterics in particular, exhibit a dramatically high degree of optical activity. A comparison of the elastic aspects of liquid crystals with their optical properties leads to the conclusion and prediction that acoustical activity, where the plane of polarization of transverse acoustic waves is rotated, must be present in cholesteric liquid crystals, and possibly other liquid crystalline phases.

It is the purpose of this letter to establish the nature of acoustical activity in liquid crystals arising from the spatial dispersion effects of sound waves that are due to elastic effects. Later communications will describe the electrohydrodynamic effects, such as flexoelectricity and electric displacement gradients coupled with the elastic structure as sources of additional types of acoustical activity.

The importance of acoustical activity in liquid crystals is easily found in the possibility of acoustoptical/acoustic devices and other commercial applications. Reliable experimental results will shed light on understanding the molecular effects in mechanical evanescence[3] as well as determining whether rotation effects (optical) are due to helical array structure in cholesterics, as opposed to the structure of individual molecules[4]

The total stress tensor for liquid crystals can be expressed as

$$-\sigma_{ij}=p\delta_{ij}-\sigma^{0}_{ij}-\sigma^{1}_{ij}$$

where p is the hydrostatic pressure, σ^{0}_{ij} is the stress tensor due to elastic deformations while σ^{1}_{ij} is the stress tensor due to viscous effects.

For elastic crystals it is possible to relate the stress tensor σ to the strain tensor t through elastic stiffness constants C_{ijkl}[5] by the relation

$$\sigma_{ij}=C_{ijkl}t_{kl}$$

For liquid crystals, it is not at all simple to obtain a similar relationship. For cholesterics, Nehring and Saupe[6] demonstrate that terms of higher order than quadratics in director gradients may be significant in determining the elastic energy density while Moritz and Franklin[7] demonstrate how simple nematics may include terms of high order in strain rate tensor and director gradients in the viscous stress tensor. In order to facilitate the discussion, the approach of Martin, Parodi and Pershan[8] is adopted in this paper. Thus, the viscous stress tensor is written as

$$\sigma^{1}_{ij}=-\eta_{ijkl}\partial v_{i}/\partial x_{l}$$

while the expression for the elastic stress, in terms of the gradients of the director $\vec{n}$ ($n_{ij}=\partial n_{i}/\partial x_{j}$), for small deformations can be written as[6]

$$\sigma^{0}_{ij}=\xi_{ir}n_{rj}+\xi'_{irst}n_{rs}n_{tj}$$

For our development, we may ignore the hydrostatic pressure term $-p\delta_{ij}$ to remain with a total stress tensor

$$\sigma_{ij} = \xi_{ir}n_{rj} + \xi'_{irst}n_{rs}n_{tj} - \eta_{ijrs}\partial v_r/\partial x_s \qquad (1)$$

In general, there exist dispersion effects for sound propagation[1], and as such it is advantageous to write the frequency dependences as

$$\xi(\omega) = {}_0\int^{\infty} \xi(t)e^{i\omega t}$$

with similar expressions for ξ' and η.

It is now immediately apparent that the expressions $\xi(\omega), \xi'(\omega), \eta(\omega)$ are merely first order terms of expansions of the general quantities $\xi(\omega,\vec{q})$ $\xi'(\omega,\vec{q})$ and $\eta(\omega,\vec{q})$.

In particular,

$$\xi_{ir}(\omega, \vec{q}) = \xi_{ir}(\omega) + i\xi^*_{irm}(\omega)q_m + \xi^{**}_{irmp}(\omega)q_m q_p + \ldots \qquad (2)$$

is the form of the expansion, and ξ' and η have expansions similar to equation (2). For simplicity, we retain only the next higher terms in the expansions, namely ξ^*, ξ'^*, and η^*. These form the crux of any formal development of a theory of acoustical activity in liquid crystals. These tensors are the analogs of the optical gyration tensor. Since the optical gyration tensor determines the rotation of the plane of polarization of of light incident on the symmetry axis of optically active material[9], it is postulated now that the starred quantities (which may be called the first and second elastoacoustic gyrotropic tensors and the viscoacoustic gyrotropic tensor) manifest themselves by acoustical activity in liquid crystals. As an example of such postulated activity, the cholesteric mesophase is predicted to display rotation of the plane of polarization of an incident transverse acoustic wave, for an appropriate frequency. The required frequencies will be discussed at the closing of this letter.

The full expansions of the viscoacoustic and elastoacoustic gyrotropic tensors, of which equation (2) is an example, express the complete description of the dependence of frequency on the molecular structure and the wavevector $\vec{q}$. The detailed algebraic development of the comlete set of equations will be

presented in a later paper describing all aspects of mechanical and electromagnetic aspects of evanescence; however an illustrative situation occurs when the dissipation-free limit is considered. In this case $\eta(\omega,\vec{q})$ is taken to be small compared to the acoustoelastic tensors, in a manner similar to Martin, Parodi, and Pershan[8], one can derive a fourth order dispersion relation for frequency and wavevector, for cholesterics and smectic-A phase, for wavector of arbitrary incidence angle ψ between the symmetry axis $\vec{z}$ and $\vec{q}$,

$$\omega^4-\{f_1(\Theta)\times(q_1^2+q_3^2)+f_2(\Theta)q_3^2+Kq_1^4\}\omega^2+f_3(\Theta)q_1^2q_3^2=0 \qquad (3)$$

where f_i are functions of a set of thermodynamic variables represented by Θ. The solution of equation (3) leads to propagating modes with velocities c_1 and c_2 satisfying

$$\begin{aligned} c_1^2 + c_2^2 &= f_1(\Theta) + f_2(\Theta)\cos^2\psi \equiv G_1(\Theta,\psi) \\ c_1^2c_2^2 &= \cos^2\psi\sin^2\psi f_3(\Theta) \equiv G_2(\Theta,\psi) \end{aligned} \qquad (4)$$

and thus

$$\begin{aligned} c_{1\pm} &= \{\tfrac{1}{2}(G_1(\Theta,\psi) \pm (G_1^2(\Theta,\psi)-4G_2(\Theta,\psi))^{\frac{1}{2}}\}^{\frac{1}{2}} \\ c_2 &= \pm G_2^{\frac{1}{2}}(\Theta,\psi)/c_1 \end{aligned}$$

are the solutions of equation (4).

It is now easy to see how acoustical activity is present. In complete anaology with the optical situation, the plane of polarization of an incident wave can be rotated by an angle $\Psi_{acoustic} = \frac{1}{2}\omega d(1/c_{i-} - 1/c_{i+})$ when going through a thickness d. It is obvious that two distinct possible acoustic rotations exist when $c_i \neq 0$ corresponding to the two different solutions of equation (4).

In the simple case where the wavector is incident parallel to the axis of symmetry of a cholesteric, $\psi=c_2=0$ and it is obvious that the acoustic activity is given by the expression $\Psi_{acoustic}=\omega d/\sqrt{G_1}$. The solution with $c_1=0$ leads to unphysical results, for instance infinite acoustical activity and no propagation of sound which is not generally true.

The next steps which are required to complete the theory of acoustical activity for liquid crystals are a) the accounting for dissipative effects which will influence the amplitude of transmitted waves but not their angle of rotation, and b)electrohydrodynamic influences which will introduce additional possible speeds of propagation, and therefor additional angles of rotation. These will be treated elsewhere.

From the definitions of acoustical and optical activities it becomes apparent that order of magnitude calculations are best performed by taking the ratios of activities, namely, $\Psi optic/\Psi acoustic \propto \lambda acoustic/\lambda optic$, where the λ's denote the appropriate wavelengths. Thus since acoustical velocities in liquid crystals are of the order of 10^5 cm/sec[1], acoustic waves in the Gigahertz region will display acoustic rotary powers comparable to optical rotary powers (10^3rad/cm) for cholesterics. These acoustical rotary powers are large enough to merit close investigation since they should be easily detactable and can be utilized in acoustical transducers.

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