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Acousto- and acousto-optic effects in liquid crystals: An overview

*Abraham George**

Department of Physics, Sultan Qaboos University, P.O. Box 36, Al Khodh 123, Muscat, Sultanate of Oman
*) Email: akgeorge@squ.edu.om

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An overview of the following three aspects of the acoustical and related optical behavior of liquid crystals, namely i) ultrasonic studies, ii) acousto-optical effects, and iii) surface wave induced optical effects have been discussed. The temperature dependent ultrasound velocity has been used for the evaluation of the adiabatic compressibility (κ_S) and the acoustic impedance (Z). A correlation between thermodynamic functions and thermo-elastic properties facilitated the evaluation of the specific heat at constant volume (C_V), the ratio of specific heats (γ) and the isothermal compressibility (κ_T) across liquid crystal-isotropic phase transition. Acousto-optical effect in liquid crystals has been described in terms of molecular reorientation related to flows and turbulences due to acoustic vibration and radiation pressure in the medium. One important aspect of the acousto-optical effects is the surface wave induced optical effects. The change in transparency of a normal layer of liquid crystal under crossed polarizers are discussed which is interpreted as a consequence of the rotation of the optic axis. In addition, the acoustic emission occurring from homeotropically-aligned liquid crystals irradiated by surface acoustic waves are also discussed.

Keywords: Acousto-Optic Effects, Adiabatic and Isothermal Compressibility, Specific Heat at Constant Volume, Surface Acoustic Waves, Acoustic Emission

1. INTRODUCTION

Liquid crystals (LCs) are used in numerous applications including displays, smart windows and data storage due to their excellent electro-optical properties [1-2]. Besides electrical and optical driving, LC re-alignment based on the acousto-optic effect has also been demonstrated where acoustic wave changes the optical axis, thus changing the transmitted light intensity [3-4]. Acoustic methods are one of the useful tools for characterization of some of the LCs properties. In spite of their relatively limited use, several successful acoustic experiments were realized.

Acoustic experiments are based on the utilization of three types of acoustic waves: longitudinal, shear and surface acoustic waves (SAW). Both longitudinal and shear waves were used mostly in the study of viscoelastic properties, rheological behavior, phase transitions, and also to study the reorientation of LC molecules [5]. SAW is used to determine the viscosity distribution in

LC layer depending on the applied electric field and to the study of molecular reorientation induced by acoustic streaming.

It is well known that the acoustic methods are very useful when the phase transition in the bulk samples of liquid crystals is studied [6-8]. The objectives of the ultrasound experiments performed in LCs can be broadly characterized as, to: (a) study the sound propagation and investigate the anisotropy in attenuation (α) and velocity (v) in each mesophase, (b) investigate the dispersion process through the study of frequency and temperature dependence of α and v, (c) study the transition between various mesophases and between the mesophases and the isotropic state, (d) analyze the acousto-optical effects, a consequence of the effect of sound on molecular ordering, (e) measure the viscosity coefficients and elastic constants, and (f) correlate the ultrasonic data to thermo-physical functions.

The objective of this paper is to provide an overview of the acoustic and related optical behavior of LCs. The paper is divided in to five sections. The first section introduces the general background. The second section describes how the study of ultrasonic velocity can be used to evaluate the thermodynamic and thermos-elastic properties of LCs. The acousto-optic effects in LCs is discussed in the third section. The fourth section is devoted to the SAW induced optical effects in LCs. This section also discusses the acoustic emission from SAW wave irradiated LCs. The last section is devoted to general conclusions.

2. ULTRASONIC STUDIES IN LIQUID CRYSTALS

2.1 Ultrasound velocity and associated parameters

Thermo-elastic characterization of LCs from the ultrasonic velocity and attenuation measurements is well established. It has been observed that the velocity decreases with increasing temperature, both in the isotropic and anisotropic regions, but shows an abrupt minimum in the vicinity of the LC– isotropic phase transition. This is behavior is true for various LC-LC phase transitions as well. Such a variation of the ultrasound velocity with temperature has been interpreted as characteristic of the LC phase transition [7, 8]

To depict these characteristics, we present here the results of a comprehensive study on a liquid crystal formed by mixing two non-mesogens, *viz*., cholesterol and cetyl alcohol [8]. Textural study to identify the liquid crystal phase and the determination of the LC – isotropic transition temperature using thermal microscopy has confirmed that the mixture exhibits smectic A (S_A) phase below 48.2^{o}C. The thermo-elastic characterization of the LC was carried out from the measurement of the ultrasound velocity and density in the anisotropic and isotropic phases of the LC mixture. The associated properties, *viz*., the adiabatic compressibility (K_S) and the acoustic impedance (Z) are also studied. The thermodynamic characterization of the LC phase has been carried out from the measurement of specific heat at constant pressure using differential scanning calorimetry (DSC). A correlation between thermodynamic and thermo-elastic properties was developed which enabled the estimation of the temperature dependent ratio of specific heats (γ) using experimentally determined quantities. This has facilitated the determination of the specific heat under constant pressure (C_P), and the isothermal compressibility (K_T).

Variation of ultrasonic velocity with temperature is given in Fig. 1. The velocity decreases with increasing temperature, in both the isotropic and anisotropic regions, but shows an abrupt minimum in the vicinity of the S_A - I transition. Such a variation of the ultrasound velocity with temperature is interpreted as the characteristic of liquid crystal phase transitions [8].

Two parameters of immediate interest are the adiabatic compressibility and acoustic impedance. The adiabatic compressibility (K_S) can be expressed as a function of temperature and can be determined using,

$$K_s(t) = -\frac{1}{V}\left(\frac{\partial V}{\partial P}\right) = \omega^{-2}\rho^{-1} \quad (1)$$

K_S increases sharply with increasing temperature in the LC phase as one approaches the transition region (Fig. 2). Beyond the transition temperate, it decreases with a sharp gradient. Away from the transition temperature, K_S increases with increasing temperature in the isotropic phase.

Acoustic impedance, *Z*, is the pressure difference created by a compression wave traveling in a liquid. The electrical equivalence of acoustic impedance is the resistance experienced by electrical currents with

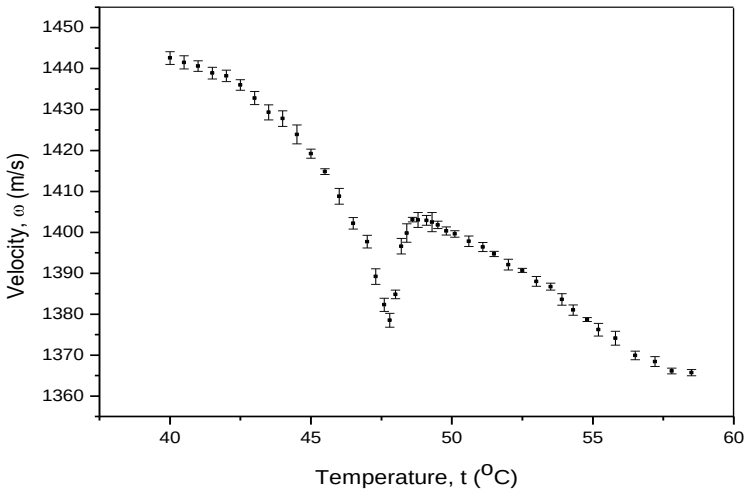

Figure 1 Variation of ultrasonic velocity with temperature. The velocity shows an abrupt minimum in the vicinity of the S_A – I transition.

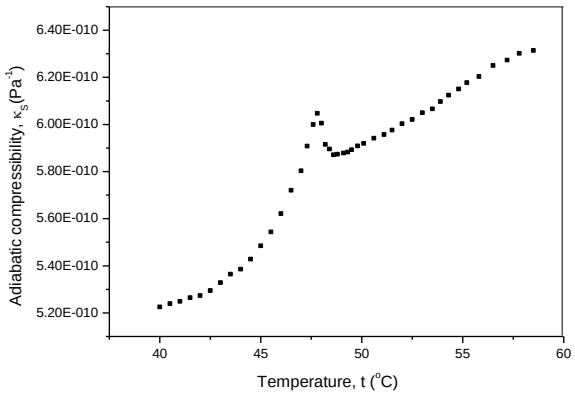

Figure 2 Adiabatic compressibility data plotted as a function temperature shows a sharp increase on reaching the transition temperature.

voltage being analogous to the pressure difference. The density (ρ) and ultrasound velocity (ω) measurements can be used to determine the temperature dependent acoustic impedance using,

$$Z(t) = \omega(t)\rho(t) \tag{2}$$

Z(t) also is found to show anomalies at the phase transition (Fig. 3). It decreases with increasing temperature in the smectic phase. A sharp decrease is seen in the vicinity of the transition followed by a sharp increase. In the isotropic phase *Z*(t) decreases with increasing temperature.

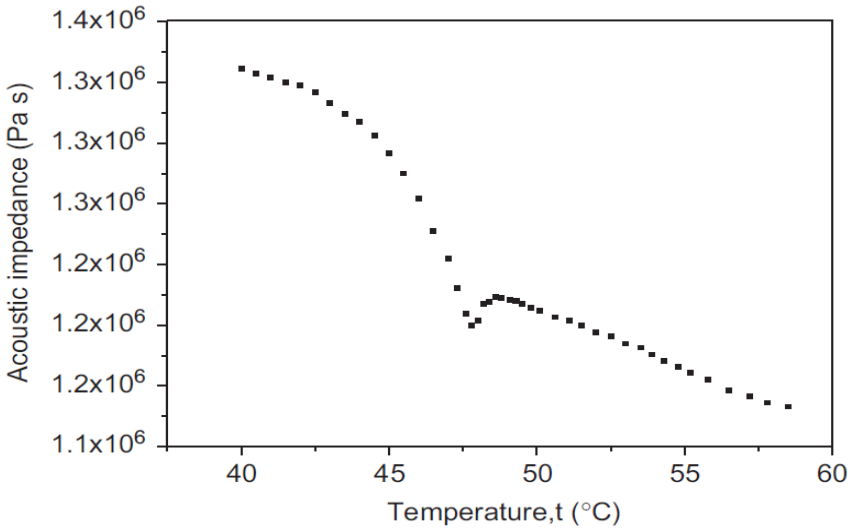

Figure 3 Variation of the acoustic impedance with temperature showing a minimum at the S_A – I transition.

It has been observed that the ultrasonic velocity and related constants such as adiabatic compressibility and acoustic impedance are temperature dependent. Besides their dependence on structure, they are related to intermolecular cohesion, molecular attraction and molecular order in LCs. Starting from the isotropic phase with a decrease in temperature, the molecular

interactions will increase and hence compressibility decreases while at the isotropic-liquid crystal phase transition, the low order molecular assembly changes to a high order one, thus leading to vigorous hetero-phase fluctuations. These fluctuations enhance the compressibility of the medium enormously and hence in turn lower the ultrasonic velocity and acoustic impedance.

2.2. Evaluation of isothermal compressibility and specific heat at constant temperature

A correlation between thermodynamic functions and thermo-elastic properties has been established through thermodynamic route [8]. The ratio of specific heats, γ can be determined from thermodynamic relation through the measured specific heat data C_P(t) , velocity ω(t) and density ρ(t) using equation,

$$\gamma = 1 + \frac{TV\alpha^2}{C_p K_s} = 1 + \frac{TV\alpha^2\rho}{C_p\omega^2} \tag{3}$$

Equation (3) can be used to determine the temperature-dependent γ because α, and K_S are known from the ultrasonic studies and C_P can easily be measured using a DSC. Thus the measured values of C_P and K_S to determine the isothermal compressibility (K_T) and the specific heat at constant volume (C_V) using,

$$K_T = \gamma K_S \tag{4a}$$

and

$$C_V = \frac{C_p}{\gamma} \tag{4b}$$

The variation of C_V and K_T are shown in Figs. 4 and 5. C_V values exhibit a large variation (Fig.4) in the region of the phase transformation. C_V for the S_A phase increases as one approaches the region of transition and then drops sharply and becoming almost flat in the isotropic region. K_T values are found to show a large increase in the S_A - I transition region with a kink at S_A – I transition (Fig. 5). The density fluctuations between S_A and isotropic phases will increase the isothermal compressibility in the vicinity of the phases transition, which is seen in Fig. 5.

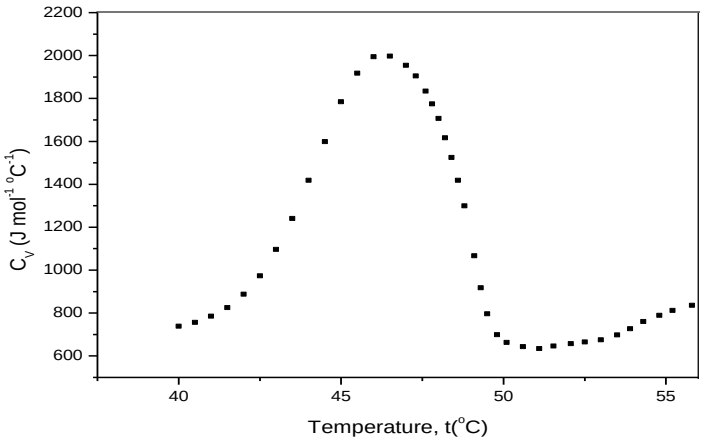

Figure 4 Variation of the specific heat at constant volume (C_V) with temperature

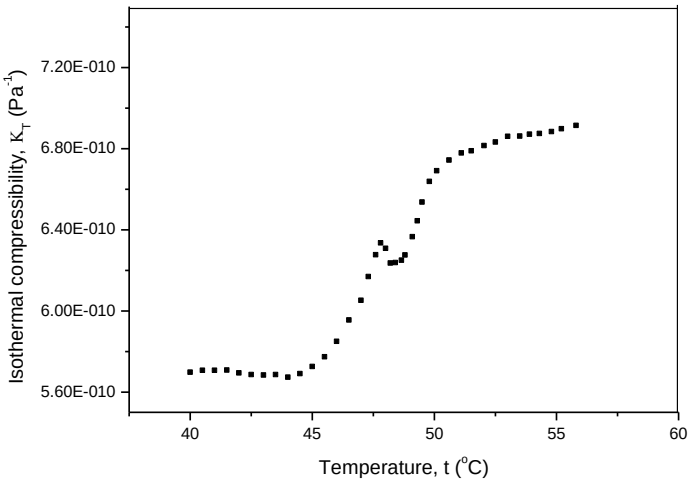

Figure 5 Variation of the isothermal compressibility K_T with temperature

3. ACOUSTO-OPTIC EFFECTS IN LIQUID CRYSTALS

The optic axis of the liquid crystal molecules can be rotated with the application of an acoustic field [9, 10]. The rotation of the molecules results in a change in the optical birefringence of the liquid crystal, which are observable by polarized light. The acousto-optic effect, analogous to electro-optic effect, have several interesting applications such as non-destructive evaluation and underwater sonar imaging [11,12].

Acoustically excited dynamic scattering was first demonstrated by Kapustin and Dmitriev [13] on un-oriented nematic LCs. Acousto-optical effect in LCs has been described in terms of molecular reorientation related to flows and turbulences due to acoustic vibration and radiation

pressure in the medium. Three parameters that control the acousto-optic effect in liquid crystals [14] are: (a) the incident angle of the acoustic wave, (b) the incident angle of the light wave and (c) the thickness of the liquid crystal layer. Several mechanisms have been proposed to explain the observed acousto-optical phenomenon which are: (a) Instability due to second order stresses, (b) static deformation resulting from a torque associated with the anisotropy of the acoustic attenuation, and (c) acoustic streaming

4. SURFACE ACOUSTIC WAVE (SAW) INDUCED OPTICAL EFFECTS

4.1 SAW excitation of oriented LCs

Though there have been several reports on the effect of ultrasonic bulk waves on the optical properties of LCs, there are only very few reports concerning the interaction of SAW waves with LCs [15, 16]. The study involves generation of SAW using interdigital transducer (IDT) fixed on one side of a Lithium Niobate ($LiNbO_3$) substrate and propagating on the highly polished surface of the crystal (Fig 6). The LC cell was prepared by sandwiching the sample between the $LiNbO_3$ and a Pyrex glass on the top. Homeotropically aligned LC samples were dark when viewed under crossed polarizers. On exciting the LC with SAW, a pattern of white stripes, which were bright at the feeding edge, started appearing. With increase of acoustic intensity, different colors started to appear at the center of the stripes, which moved out in turn to the boundaries of the stripes. Further increase of SAW wave intensity resulted in a streaming of the LC sample, leading to a condition similar to the *Dynamic Scattering* observed under electric field excitation. Increasing the SAW field to a much higher value led to the LC becoming completely randomly oriented as inferred from the appearance of overall darkness in the field of view.

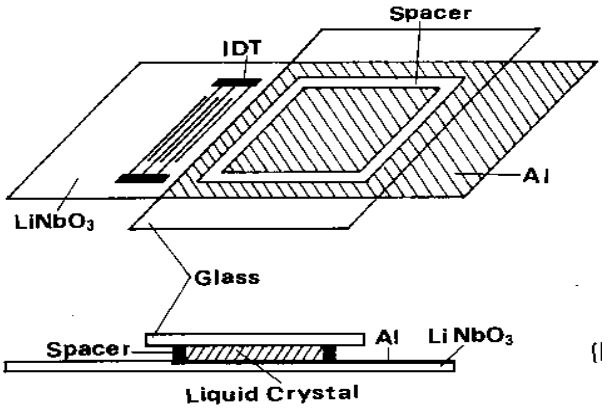

Figure 6 Structure of the liquid crystal cell: top and lateral view

The change is transparency of the LC cell was recorded as the transmitted light intensity through the cell. It has been observed that the transmitted light intensity (recorded as an output voltage *V*p) increases with the applied IDT voltage (*V*p), reaches a plateau, and then drops. Kapustina and Statnikv [15] proposed a theoretical model to explain the experimental results. The model considers the change in transparency of a normal layer of LC layer in polarized light, in consequence of the rotation of the optic axis. When crossed, the first polarizer transmits the component E_x of the electric vector, while the second polarizer the component E_y . When the direction of the incident beam coincides with the optic axis of the layer of molecules, the visual field under crossed polarizers will be dark. On rotation of the optic axis through an angle θ caused by the surface wave, double refraction occurs, and there appears a component of the

electric field along the *y*-axis that pass through the second polarizer. Based on the above discussion, Kapustina and Statnikov [15] obtained the following expression for the intensity of luminous flux over a period of oscillation:

$$\langle I_2 \rangle = A\left\{\frac{\pi}{4} - \frac{1}{\sqrt{1 + a^2\beta^2 v_{ox}^2}}\, arctg \frac{1}{\sqrt{1 + a^2\beta^2 v_{ox}^2}}\right\} \tag{5}$$

Here A is a constant. $a = \Lambda/\varepsilon_{//}$ where Λ is the Maxwell's constant related to the Maxwell's effect of dielectric permittivity tensor [17] and $\varepsilon_{//}$ is the electric permittivity parallel to the director, $\beta^2 = \omega/2\gamma$, where $\omega = 2\pi f$ and γ is the kinematic viscosity of the LC. v_{ox} is the amplitude of the oscillation velocity. Since the mean light flux falling on the light detector is proportional to the voltage V_p, and since the amplitude v_{ox} of the oscillation velocity in the wave is proportional to the voltage V_t of the IDT ($v_{ox} = k(f)Vt$), where $k(f)$ is a frequency-dependant coefficient), Eq. (5) can be reduced to the following form:

$$V_p = C_0\left\{\frac{\pi}{4} - \frac{1}{\sqrt{1 + C_1^2 V_t^2}}\, arctg \frac{1}{\sqrt{1 + C_1^2 V_t^2}}\right\} \tag{6}$$

where $C_1^2 = k(f)\alpha^2\beta^2$depends on the frequency, but C_o is a constant. Equation (6) is more convenient for practical use. For $C^2 V_t \ll 1$ and $C_1 V_t \gg 1$, we have respectively,

$$V_p = C_o C_1^2 V_t^2/2 \tag{7}$$

and,

$$V_p = C_o \pi/4. \tag{8}$$

George [16] using the experimental values, constructed the theoretical dependence of the IDT voltage with the voltage output of the light detector. Theoretical values are very close to the experimental values, which attests the correctness of the theoretical model.

4.2 Acoustic emission from SAW irradiated LC cell

Acoustic emission (AE) is the process of transient elastic wave generation due to rapid release of strain energy caused by structural alteration of solid material. As in the case of solids, when subjecting a LC to phase transition or to the interaction of external fields, structural changes and defect movements are possible. Under such conditions, due to the existence of elastic energy associated with the presence of defects, one should expect AE to take place inside the liquid crystal. However, a fraction of the released energy may be absorbed due to the viscous nature of the LCs.

George [18] studied the AE counts as a function of time and applied voltage for various cell thicknesses. The cell used was the same as in Fig. 6. The count rate dN/dt, which represents the AE activity increases with increasing transducer voltage, reaches a maximum and then drops. Further AE counts for various SAW voltage for a fixed time for various cell thicknesses. In all cases, the AE counts first increase with increasing SAW field and reach a maximum at some threshold voltage. These are found to decrease with further increase in SAW field. For

voltages much higher than that corresponding to the peak emission, the sample become very quiet.
AE is connected to the dynamics of defects when a sudden release of elastic energy associated to them is involved. Therefore, AE may constitute a new tool for the study of defects and their dynamics in LCs.

5. CONCLUSIONS

The acoustic properties of liquid crystals, specifically its interaction with ultrasonic waves and the related thermodynamic and thermos-elastic properties have been discussed. Two parameters of interest were the adiabatic compressibility and the acoustic impedance. Further, a method by which the thermodynamic parameter like, specific heat at constant volume, the ratio of specific heats and isothermal compressibility near the phase transition was demonstrated. The interaction of surface acoustic waves with a thin layer of liquid crystals and the related acousto-optical effects have been discussed in terms of the rotation of the optic axis of the liquid crystal. Further, the acoustic emission occurring from a thin film of liquid crystals when irradiated with surface acoustic waves was discussed.

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