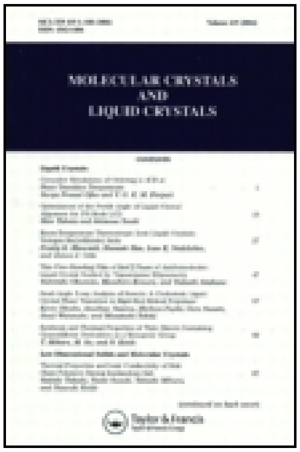

Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl17

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Version of record first published: 20 Apr 2011.

To cite this article: Kapustina Olga (1990): Analysis of Surfaces Properties by Acoustic Methods as Applied to Liquid Crystals, Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics, 179:1, 173-185

To link to this article: http://dx.doi.org/10.1080/00268949008055367

Mol. Cryst. Liq. Cryst., 1990, Vol. 179, pp. 173–185
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Printed in the United States of America

ANALYSIS OF SURFACES PROPERTIES BY ACOUSTIC METHODS AS APPLIED TO LIQUID CRYSTALS

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Abstract Acoustic visualization, acoustic microscopy, acoustic probing, method of acoustic emission and photoacoustic methods are discussed as applied to liquid crystals.

INTRODUCTION

Surface physics is an extremely diversified field of science as far as the range of its problems is concerned. Among present-day experimental techniques of studying free surfaces and phase interfaces, it is acoustic methods of nondestructive testing based on the penetrability of acoustic waves that are particularly attractive. Some of these methods may be recommended for investigating the interaction between liquid crystals (LC) and boundary phases (solids, gases, liquids). Those are: acoustic visualization, acoustic microscopy, acoustic probing, method of acoustic emission, photoacoustic methods. Among all known LCs, nematic LCs which have only one type of thelong rang order are the most susceptible to external actions, including surfaces forces. For this reason we shall refer mainly to the nematic liquid crystal (NLC).

ACOUSTIC VISUALIZATION

Nature has not gifted man the ability to "see" with the help of sound. However, during the last sixty years many various ideas have been sugges - ted to devise acoustic instruments which could enable man to overcome the limitation. A characteristic feature of LCs consists in their unique capability of transforming the information of an acoustic field into the visible image without using any axiliary devices. Figure 1 presents simplified scheme of producing the image of a wave field in NLC placed in a capillary whose walls orient somehow the mesophase molecules. Visualization becomes possible due to an ultrasonically induced change in polarization of the light passing through the NLC layer, or due to a change in the light scattering intensity.

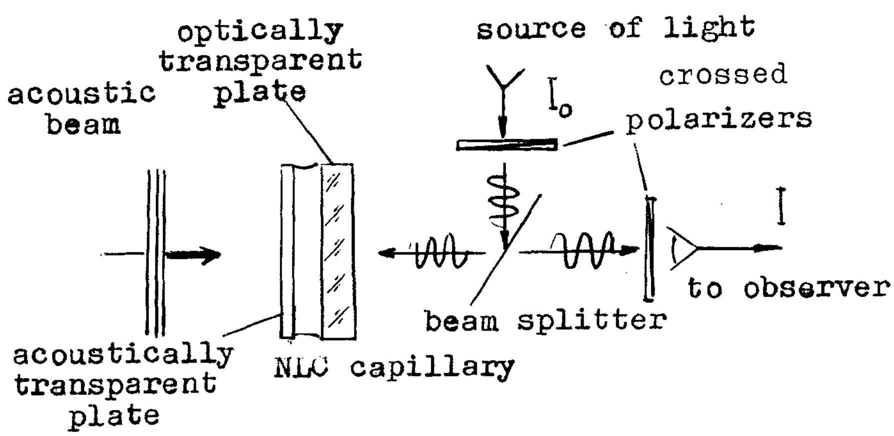

FIGURE 1. Scheme of acoustic visualization

A solid wall acts on the LC in two way: a)it imposes boundary conditions forthe whole volume of the specimen and thus determines the behaviour of the NLC under the action of disturbances; b)it changes the structure and properties of the NLC boundary layer. The above scheme makes it possible to identify the boundary conditions in the capillary and find the degree of the optical homogeneity of specimens (completely dependent on surface conditions) by means of a volumetric change in orientation caused by acoustic actions.We shall not touch upon various acousto-optical effects in NLCs reported in numerous papers (see reviews [1,2]), but we consider particular examples to find how the picture of the wave field in the NLC layer is connected with the type of boundary conditions in the capillary and with the optical homogeneity of the specimen.

Surface acoustic waves (SAW) seem to be the most attractive for the purpose of identifying boundary conditions, the corresponding scheme is shown in figure 2. In a general case, at the ar-

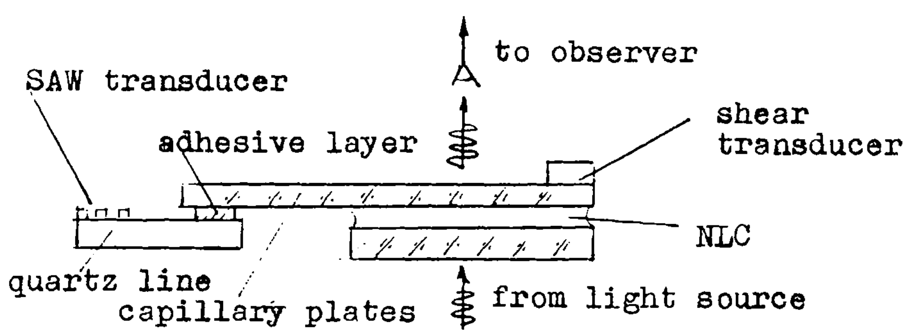

FIGURE 2. Identification of boundary conditions in NLC layer by SAW

bitrary relation of the layer thickness d and the surface wavelength, the wave field in the NLC layer is a complicated pattern of interference of modes with different wave numbers and attenuation constants. This interference pattern can be calculated only by numerical methods. However, we succeeded in showing[3] that the wave field in the layer is the superposition only of two modes in a frequency range where the thickness of the NLC layer and the characteristics of wave fields obey following inequalities

$$l_R \gg d, \quad l \gg d, \quad l_{vis} \ll d \qquad (1)$$

(l_R- surface wavelength in the plate material, l- wavelength in the NLC, l_{vis}- viscous wavelength in the NLC). In this case it becomes possible to obtain the analytical form of the relation between the orientation distortion, layer thickness and the acoustic parameters. The picture of the distortion is spatially modulated[4] but its periodicity depends on the type of boundary conditions. For homeotropic conditions the spatial period L of distortion caused by SAW is given by following relationship $L = 0{,}5\ l_R(\ C_R/C - 1\ )^{-1}$ and is independent of the layer thickness. Here C_R means the SAW velocity in the plate, and C is the velocity of the elastic wave in the NLC ($C_R = 3.2\times10^5$ cm/s; $C = 1.5\times10^5$ cm/s). For planar boundary conditions the period of distortion is $L = 0{,}61d$ and does not depend on the sound frequency.[5] (It is supposed that the SAW propagates along the director). At typical layer thicknesses of 10 to 40 μm inequality (1) is valid in a range of 5 to 40 MHz, this determining the limits of the applicability of the present method for NLC testing. The acoustic power P_R required for visualization of the image depends on the layer thickness, SAW frequency and physical constant of the NLC[3,4]. For a layer 40μm thick, at a SAW frequency of 28.38 MHz the value of P_R is about 10 mW/cm [3]. The method of acoustic visualization can be made more sensitive. For this purpose a source of shear vibrations (Y-cut quartz) is added into the above visualization scheme [6]. It produces a coherent viscous wave in a NLC near the phase boundary, this essentially changes the picture of stationary acoustic flows responsible for orientation distortion. The considered ways of combined actions on the NLC lead to an increase in the spatial homogeneity of the optical image and also result in that the value of P_R decreases by factor of 10 to 100.

Acoustic visualization has been successfully applied in the characterization of the orientation homogeneity of NLC layer. The chematic drawing to obtain the image is shown in figure 1. The necessary condition for the succes of such tests is the homogeneity of the wave field in the NLC layer. In the case of remote visualization all observation must be carried out only in the Fraunhofer's zone. The contact method of visualization requires that the source of acoustic waves should be a Straubel-cut quartz. This enables a more uniform distribution of vibration amplitude over the transducer surface. The value of P_R at frequencies of about unities MHz for layer 10 to 100 μm thick is 40 and 600 mW/cm^2,respectively.[7]

ACOUSTIC MICROSCOPY

Acoustic microscopy is a modification of the method of acoustic visualization.There exist severel schemes of an acoustic microscope which differ one another mainly in the principle of transforming the acoustic image into the visible image. Specifications of modern acoustic microscopes are as follows: a frequency range - from 50 to 3000 MHz, resolution - 50 to 0,1 μm, magnification x50 to x5000, depth of penetration - 1μm to 1 mm. In the University of Stenford (USA) a family of cryogenic scanning acoustic microscopes has been devised for studing surfaces of materials. One of them[8] possesses the highest resolving power - 25 to 30 nm at 8 GHz, liquid helium at 0.1 K being used as the immersion liquid. The method of acoustic microscopy is rather sensitive to the presence of inhomogeneities in micro-objects,including optically opaque ones. Such a technique has the ability to detect disturbances of adhesion, flaking, microcracks, pores, foreign inclusions, deviations from the prescribed thickness of a layer in multilayer systems and coatings. Possible applications of this method are topographical and morphological investigations of smooth and textured surfaces. It may be helpful in quality control of multilayer structures used for NLC orientation and composite materials (NLC-polymer) whose components are very close to each other as regards their optical characteristics, but differ in their acoustic properties. Particularly, it may be recommended for studing the polymer-dispersed liquid crystal films (analysis the droplet size, spacing and distribution).

METHOD OF ACOUSTIC PROBING

In the last years investigations of the action of vibrations on the NLC have received a marked development[1,2] . In particular, experiments have shown that the sensitivity of NLC to shear vibration depends on the anchoring energy. In figure3 the solid and dashed lines show the amplitude dependence of the dc component of the optical signal for the NLC layer placed between "clean" polished glass plates and between plates with an amorphous film[9]; d = 100 μm, f = 500 Hz[10]. According to Naemura[11] and Kleman[9] for these specimens the anchoring energy W is $(3...5)10^{-3}$ and $(3...4)$ 10^{-4} erg/cm^2, respectively, i.e. the ratio is more than 10. These results give qualitative relation between the anchoring energy and the optical reaction of the NLC to the acoustic action.

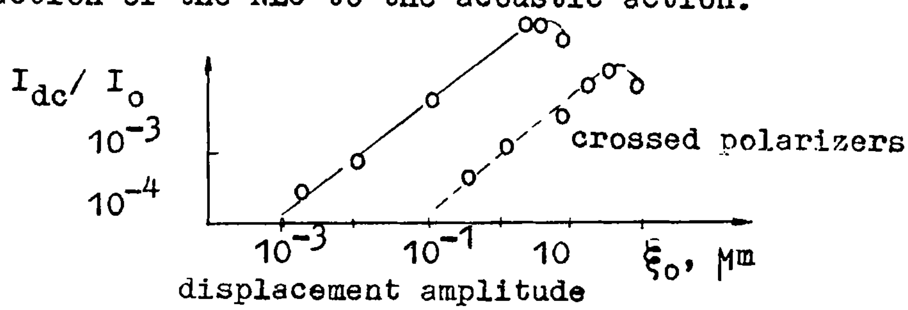

FIGURE 3. Behaviour of the dc component of the optical signal under shear vibrations

For a quantitative estimation of the anchoring energy we suggest the new method based on measuring the dc component of the optical signal for the homeotropic layer of the NLC under simultaneous action of two types of vibrations of frequencies ω shifted in phase by $\pi/2$. Suppose the motion of one of the plates is described by

$$V_x\big|_{z=d} = V_o \cos \omega t, \quad V_z\big|_{z=d} = a\, V_o \sin \omega t \qquad (2)$$

where a - ratio of the amplitudes of the components V_x and V_z. The character of the anchoring of the NLC molecules with the plate surfaces is determined by

$$W\theta - K_3 \partial\theta/\partial z\big|_{z=o} = 0, \quad W\theta - K_3 \partial\theta/\partial z\big|_{z=d} = 0 \quad (3)$$

Here θ is the angle made by the director with the z -axis. We are interested in the orientational behaviour of the director in the audible frequency range satisfying the condition $l_{or} \leq d < l_{vis}$, $l < L_s < l_{vis}$, $L_s \gg d$ (here L_s - the sample length, l_{or} - the orientation wavelengths). For specimens 10 ... 100 μm thick this condition is valid up to frequencies of about 10^3 Hz. An analysis shows[12] that in the geometry considered the non-linear interactions between the director oscillations and the velocity field give rise to a stationary distortion of the structure of the layer so that the director oscillates near the new quasi-equilibrium state. Following our analysis[12] for the boundary condition (3) the angle θ_{st} describing a stationary effect is written as

$$\theta_{st}=1/2\gamma a\omega\xi_0^2 d^{-4}K_3^{-1}\left\{-\left[2z^4-3z^3d+zd^3-\delta zd^3(1+2\delta)^{-1} + \delta(1+\delta)d^4(1+2\delta)^{-1}\right]+3/2\gamma\eta_2^{-1}(z^4-2z^3d+z^2d^2)\right\} \quad (4),$$

where $\delta = K_3/Wd$ is the dimensionless parameter which characterizes the anchoring of the NLC molecules with the boundary. At amplitudes and frequencies satisfying the inequality $0{,}1\gamma a\omega\xi_0 d/K_3 \gg 1$ nonlinear effects are very considerable and the contributuion made by the ststionary distortion into change in the optical transmittivity of the layer is exhibited distinctly. In such a situation dc component of the optical signal is given by $m_0 = 0{,}5\left[1 - J_0(P_1)\cos P_0\right]I_0$ if the the angle between the polarizer and the direction of shear vibrations of the plate makes $\pi/4$. Here I_0 is the intensity of the incident linearly polarized light, J_0 is Bessel's function of the first kind. The parameters P_0 and P_1 depend on the values of ξ_0. It implies that the dc component m_0 is a function of ξ_0. At high value of P_0 (for $P_0 > P_1$) the first maximum of this function is determined by the following condition:

$$P_0 \approx \pi. \quad (5)$$

According to [12]

$$P_0 = 0.0075\, k_0 \Delta n d(a\gamma\omega\xi_0^2/K_3)^2\left[1 + 0{,}12(\gamma/\eta_2)^2 - 0{,}67\,\gamma/\eta_2 + f(\delta)\right] + \Delta n \xi_0^2\, k_0/2d, \quad (6)$$

where $k_0 = 2\pi/\lambda_0$, Δn is the birefringence of the NLC, η_2 and γ are viscous coefficients of the NLC, The relations between the anchoring energy W and the amplitude ξ_o^{max} corresponding to the first maximum of m_o derived from (5) and (6) are shown in figure 4 for a NLC sample, 15 μm thick, at 123 Hz for the following number of values of the parameter $a = V_z/V_x$: 0.16, 0.4 and 0.9 (curves 1-3). It can be seen that in the region of weak anchoring small changes in W lead to shift of ξ_o^{max} to amounts which can be measured. Experimental values of ξ_o^{max} are also shown in figure 4 (a mixture of MBBA and EBBA; the plates bounding the NLC layer were treated with lecithine, $W \simeq 10^{-3}$ erg/cm^2). The change in the values ξ_o^{max} as a function of W is in agreement with the dependence of $\Theta_{st}^{\times}$ on W (curve 4 for d = 50 μm, z = d/2, ξ_o= 1 μm, a = 0,1 and f = 100 Hz).

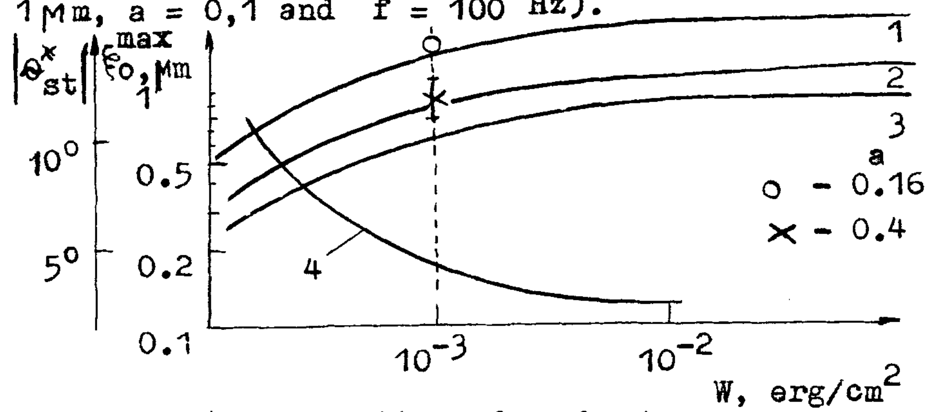

FIGURE 4. Evaluation of anchoring energy

METHOD OF ACOUSTIC EMISSION

It was considered for a long time that acoustic emission (AE) was inherent in solids only. However as it turned out AE is also characteristic of LCs in which there are a lot of inhomogeneities. Italian colleagues have shown that one of the simplest situations to observe AE is a change in the sample temperature, during this change the LC passes a number of phase transformations.[13] Such observations were carried out on thick (~1cm) nonoriented samples of MBBA, COOB, CBOOA. It was found that the acoustic activity was most clearly pronounced in the nematic-isotropic transition. A high sensitivity of the method to microstructurel inhomogeneities of materials allows one to view it as a mean of control of the alignment stability of LC sample. Unfortunately, little attention is given to this problem.

OPTOACOUSTIC SPECTROSCOPY

The successful development of powerful lasers has led to the progress of the existing methods of optical spectroscopy, for instance, of the optoacoustic method (OAM). It is based on the generation of a thermal or an acoustic waves by absorption of pulsed light. An important feature of the OAM is the dependence of the OA signal on thermal properties of the medium. It enables one to employ this method for studing the phase state of materials and for probing thermal properties of the samples. For these purposes the OAM with indirect detection of sounds in a gas surrounding the sample is most efficient in condensed media. Since the OA signal is proportional to the energy absorbed by the sample, the light scattering, which offers serious difficulties in standard spectroscopy does not play any essential role in the indirect OAM. This makes it possible to use the

OAM in the spectral analysis of strongly scattering objects. Rosencwaig was the first who demonstrated the possibility of applying the OAM to the stady of pretransitional effects in LCs [14]. More recently[15] within the framework of the general theory of OA spectroscopy it became possible to find a simple correlation between OA signal (phase, amplitude) and two dimensionless parameters of LCs (thermal-diffusion to optical-absorption length ratio, sample to gas effusivity ratio). Several methods are employed for the detection of the OA signal (gas microphones,piezoelectric transducers, probe beam refraction). Recently in the Institue of Acoustics of the Academy of Sciences (USSR) a family of optical microphones on LC has been devised. One of them[16] may be used in the detection of OA signal.

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APPENDIX

We collect here the set of symbols used in the paper but not defined in the text:

I_o - the intensity of the incident linearly polarised light;

I_{dc} - the dc component of the optical signal measured between crossed polarizers;

ξ_o - the displacement amplitude of the shear;

l_{vis} - the viscous wavelength;

K_3 - Frank elastic constant;

a - the ratio of the amplitudes of the sound particle velocity components V_z and V_x;

$$f(\delta) = \left[5.5(1+2\delta)^{-1} - 1.65\gamma\eta_2^{-1}\right]\delta + \frac{11\delta^2(1+3\delta+3\delta^2)}{(1+2\delta)^2}.$$

$$\theta_{st}^{x} = 1/4\, a\gamma\omega\xi_0^2 K_3^{-1}\left[3/16\,\gamma\eta_2^{-1} - 1/2 - \delta\right].$$