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INVESTIGATION OF LIGHT DIFFRACTION ON AN ULTRASONIC WAVE IN LIQUID NEMATIC CRYSTALS

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During the investigation of laser polarized light diffraction on a pulse ultrasonic wave a fine structure in diffraction orders was observed in a cell filled with a nematic liquid crystalline medium (N – 4 hexyloxybenzyliden, merck 4, merck 5 or a mixture of liquid W5 crystals). The fine structure is visible only after exceeding of the threshold intensities of the ultrasonic wave introduced into the nematic first ordered by the external magnetic field (B = 0.8 T) for polarized light in the polarization plane of the ordinary ray. This effect is connected with the reorientation of the location of the molecule axis caused by the ultrasonic wave. Basing on the interference images (obtained by orthoscopic method) average values of torsion angles of the optical molecule axis were calculated in the volume of the sample.

1. Introduction

The following works [1, 2, 3, 4] were devoted to the investigations of acoustic–optical effects in anisotropic liquid media. In this paper we present the investigation results of the changes of molecule orientation of a nematic liquid crystal with planar texture caused by an ultrasonic wave. The information concerning the order of the molecules of the liquid crystalline medium was based on the interpretation of the diffraction images of the light interacting with the acoustic field introduced into the liquid–crystalline sample. Light diffraction by an ultrasonic wave first investigated by DEBYE and SEARS [5] as well as by LUCAS and BIQUARD [6] has been described by many authors. Later on the problem of light diffraction on an ultrasonic wave was dealt with in a number of papers and monographs e.g. [9, 10, 11, 12] though the diffraction of Bragg's type and Raman–Nath's type in isotropic media was treated separately. In 1974 Tshirikov and Parigin presented a theoretical paper combining both cases of light diffraction by one mathematical description [13]. The

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interaction of light with an acoustic wave is particularly interesting in anisotropic media. The optical birefringes of these media can change the polarization state of both the light and acoustic wave passing through the medium and thus it can effect the final diffraction pattern. In liquid–crystalline media the acoustic wave can change the molecule arrangement. The orientating effect of the ultrasonic wave of liquid crystal molecules was first stated by KESSLER and SAWYER [14] who had observed a dynamic light scattering in an originally non-oriented PAA sample (para-azoksyanizol). The orientation effect of the ultrasonic wave in a monocrystalline layer of the nematics was also described by MAILER and LINKS [15] et al. [16, 17, 18]. The changes of orientation arrangement of the liquid–crystalline molecules in the volume of the sample caused by the acoustic wave changes the light diffraction image after passing through the sample. This effect was the subject of papers [19] and [3]. Liquid crystals are media in which molecules have anisotropic dielectric properties (that is also optical ones) [20, 21] and show a defined degree of a far range orientation order. The molecule axes are placed, parallely to each other in the volume of the sample. In such a mezophase molecules have translational degrees of freedom, unlike those of a solid. The acoustic wave introduced into the sample interrupts the state of mutual arrangement which can evoke progressive movements of the media of the masses of molecule groups v.s. the neighbor groups in the range of the acoustic field. Such movements cause the formation of local streamings of laminar or turbulent character: the dynamics of these processes is described by hydrodynamics equations completed by the law of energy and mass conservation [22, 23, 24, 25] taking into account the anisotropic visco-elastic constants. Below a general description on the light and ultrasonics interaction is presented as a guide to the experiments which following results qualitatively have confirmed the description.

2. Description of light and ultrasonics interaction in liquid crystals

The Fig. 1 presents the experimental geometry for perpendicular interaction between linearly polarized light beam and the ultrasonic one in the liquid crystal sample. In the case of nematic sample with a planar texture (Fig. 1) located in the range of the light beam passing along the optical axis z of a liquid crystal we do not observe the effect of double refraction because the angle between the optical axis (director) and the direction of the polarization plane $\phi(z) = 0$. On the introduction of the ultrasonic wave into the sample the orientations of the axes of the liquid crystal molecules change under the influence of the acoustic field and birefringence occur. The angle between the polarization plane and the direction of the directors $\phi(z) \neq 0$ and is dependent on the parameters of the acoustic field [19, 29]. Let us denote the refractive index of the ordinary beam as n_0 and the refractive index of the extraordinary as n_θ beam only the second one will be changed and for that variation n_θ one has:

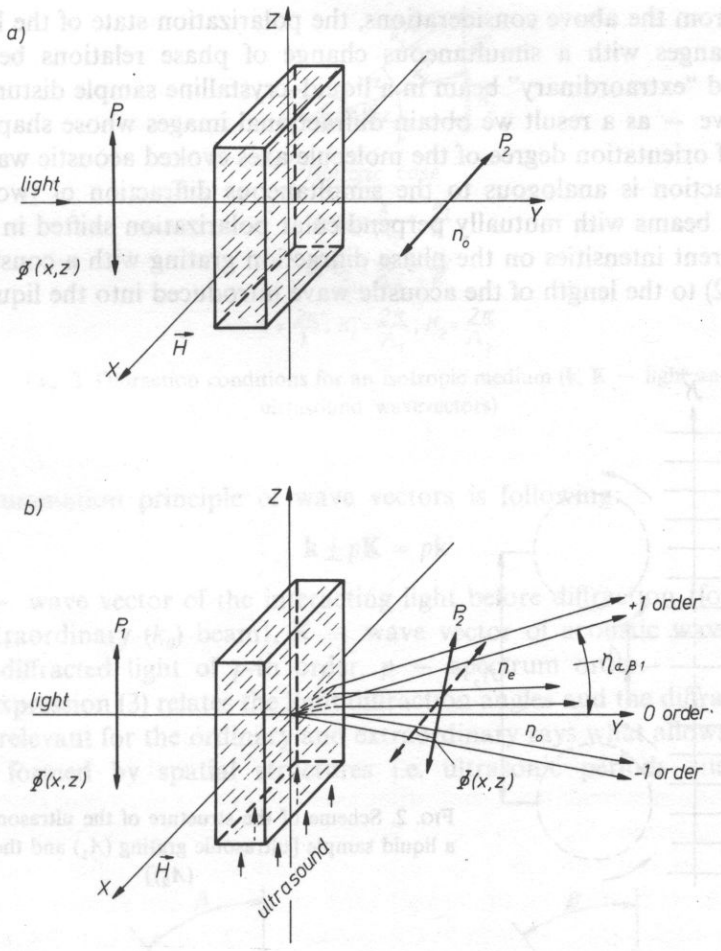


FIG. 1. Geometry and coordinate system for perpendicular iteraction lineary polarized light beam (a) and the ultrasonic one in the liquid crystal sample (b)

$$n_\theta(z, f_{a,k}) = \frac{n_0 n_\theta}{(n_0^2 \cos\phi(z, f_{ak}) + n_e^2 \sin\phi(z, f_{ak}))^{1/2}} \tag{1}$$

where: f_{ak} – characterizes the influence of an acoustic field (intensity, frequency, field distribution etc.).

The phase shift between the ordinary and extraordinary beams will be:

$$\delta(z) = \frac{2\pi d(f_{ak}) < \Delta n(z, f_{ak})}{\lambda} \tag{2}$$

where: d – thickness of the traversed way, λ – wavelength of light, $\Delta n = n_0 - n_\theta$, $\langle \ldots \rangle$ – sign of averaging process along the thickness of the layer.

As seen from the above considerations, the polarization state of the light beam undergoes changes with a simultaneous change of phase relations between the "ordinary" and "extraordinary" beam in a liquid–crystalline sample disturbed by an ultrasonic wave – as a result we obtain diffractional images whose shapes depend on the state of orientation degree of the molecule axes evoked acoustic wave. In this case the diffraction is analogous to the simultaneous diffraction of two (Fig. 1b) coherent light beams with mutually perpendicular polarization shifted in the phase and with different intensities on the phase diffraction grating with a constant equal (see also Fig. 2) to the length of the acoustic wave introduced into the liquid crystal.

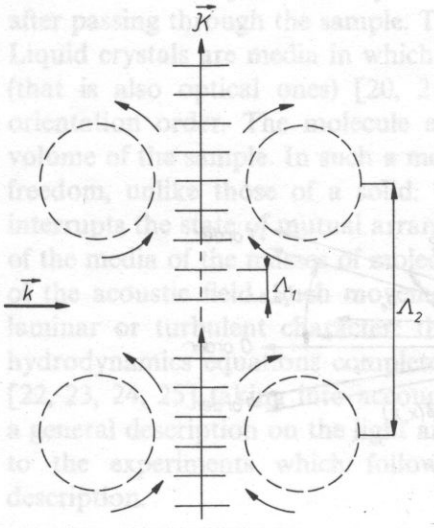
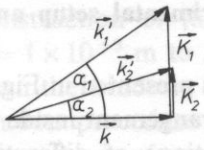


FIG. 2. Scheme of the structure of the ultrasonic field into a liquid sample [ultrasonic grating (Λ_1) and the vortex one (Λ_2)]

That picture is different, however, when the ultrasonic beam has great intensity i.e. when its radiation pressure causes a streaming of the liquid. After achieving a threshold of intensity the laminar streaming becomes a turbulent one and characteristic vortecess occur. According to MIYANA and SHEN [27] in the case of the turbulent reorientation rotation of liquid crystal molecules caused by the acoustic field formed vortecess [22, 23, 25, 26] which represent an additional spatially periodic deformation creating an additional diffractional grating with the constant (Λ_2) equal to the distance between the spatial structures (vortecess). The situation depends on the visco-elastic properties of the liquid crystal molecules their orientation and the parameters of the acoustic field (f_{ak}). Schematically such a model is presented in Fig. 2 [29]. The overlapping of these two diffraction gratings should manifest itself by the appearance of additional orders in the diffraction pattern (fine structure). As an example, Fig. 3 presents the expected diffraction conditions for an isotropic medium, and Fig. 4 – analogous diffraction conditions for a birefringent refraction medium (as liquid crystal).



isotropic case

$$tg\,\alpha_1 = \frac{\lambda}{\Lambda_1} = \frac{K_1}{k}$$

$$tg\,\alpha_2 = \frac{\lambda}{\Lambda_2} = \frac{K_2}{k}$$

$$k = \frac{2\pi}{\lambda},\ K_1 = \frac{2\pi}{\Lambda_1},\ K_2 = \frac{2\pi}{\Lambda_2}$$

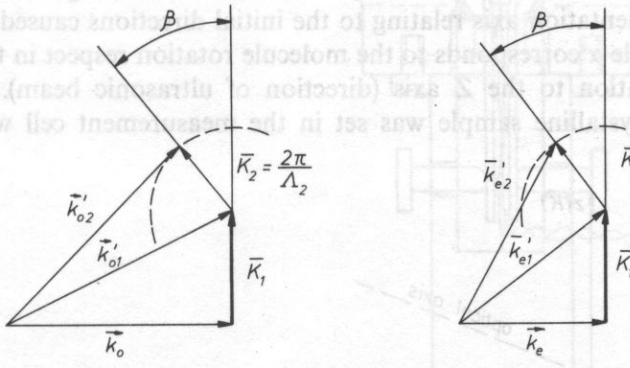
FIG. 3. Diffraction conditions for an isotropic medium ($\mathbf{k}$, $\mathbf{K}$ – light and ultrasound wavevectors)

The summation principle of wave vectors is following:

$$\mathbf{k} \pm p\mathbf{K} = p\mathbf{k}' \tag{3}$$

where: $\mathbf{k}$ – wave vector of the interacting light before diffraction {for an ordinary (k_0) or extraordinary (k_θ) beam}, $\mathbf{K}$ – wave vector of acoustic wave, $\mathbf{k}'$ – wave vector of diffracted light of p-th order, p – spectrum order.

The expression (3) relates the light diffraction angles and the diffraction gratings constants relevant for the ordinary and extraordinary rays what allows to find those constants formed by spatial structures i.e. ultrasonic periods and distance of vortecess.



$$k_o = \frac{2\pi}{\Lambda_o} \qquad k_e = \frac{2\pi}{\Lambda_o} \qquad K_1 = \frac{2\pi}{\Lambda_1} \qquad K_2 = \frac{2\pi}{\Lambda_2}$$

$$k_{01}' = \frac{2\pi}{\lambda_{01}'} \qquad k_{e1}' = \frac{2\pi}{\lambda_{e1}'} \qquad k_{02}' = \frac{2\pi}{\lambda_{02}'} \qquad k_{e2}' = \frac{2\pi}{\lambda_{e2}}$$

FIG. 4. Diffraction conditions for anisotropic medium ($\mathbf{k}_0$, $\mathbf{k}_\theta$ – light wavevectors, $\mathbf{K}_1$ – ultrasonic wavevectors, $\mathbf{K}_2 = \frac{2\pi}{\Lambda_2}$, Λ_2 – distance between vortecess

3. Experimental setup and procedure.

The measurement system is presented in Fig. 5. The system can be used both for an investigation of molecule arrangement inside the sample by classical orthoscopic optical methods and investigations of diffraction images formed by the spatial structures of the investigated sample.

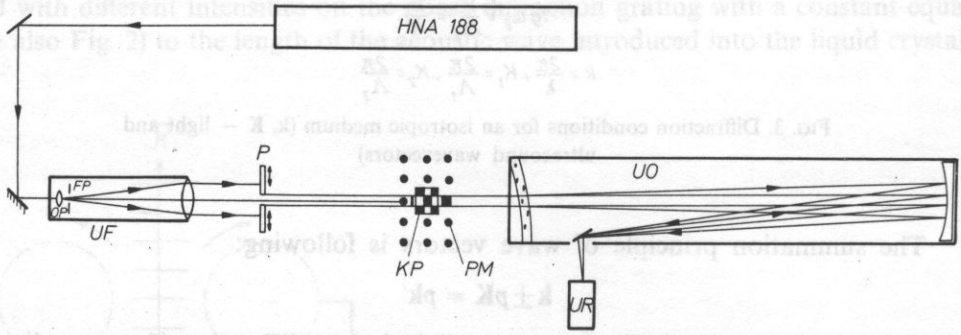


FIG. 5. Scheme of the experimental setup. HNA – He-Ne laser, UF–light beam formed system, OM–microscope objective, FP–spatial cell, PM–perpendicular magnetic field, UO–optical receiver, UR–record system

In the system the natural anisotropy of the medium can be modified using the external ordering magnetic field, and other external factors changing density, stress, streaming of molecule etc. by exposing the medium to an ultrasonic wave (see Fig. 1.b). The measure of the modification may be variation of the angles α and β (Fig. 6) of the molecule orientation axis relating to the initial directions caused by magnetic field only. The angle α corresponds to the molecule rotation respect in the plane XY and β to the rotation to the Z axis (direction of ultrasonic beam).

The liquid–crystalline sample was set in the measurement cell with precisely

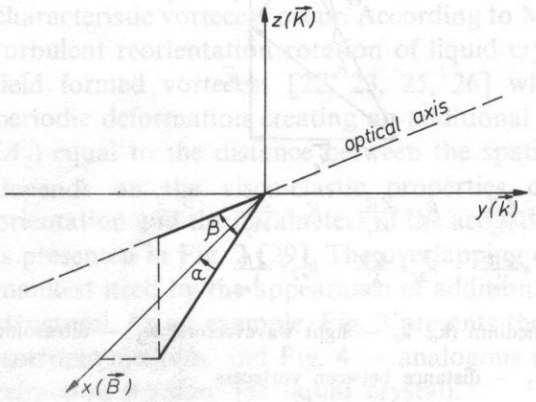


FIG. 6. Orientation liquid crystal molecule in the sample

parallel windows (Fig. 7). The construction of the cell enabled to adjust their distances within the limits from $d = 1 \times 10^{-3}$ m to $d = 10 \times 10^{-3}$ m with accuracy $\pm 0.1 \times 10^{-3}$ m. The distance between the windows – the sample thickness – was found in the interferometric calibration procedure filling the cell with additional isotropic liquid within the temperature range of nematic phase of the liquid crystal being examined. In the cell the temperature was kept within the limits of phase transitions with the accuracy to 0.01 deg by means of a thermoregulator.

The measurements were made on samples with thicknesses $d = 1 \times 10^{-3}$, 2×10^{-3} and 7×10^{-3} m for planar texture obtained by means of the external magnetic field induction $B = 0.8$T. Two transducers – quarz (9.70 MHz) or PZT ceramic (6.72 MHz) were used to form the longitudinal ultrasonic wave perpendicular to the initial axes of the liquid crystalline molecules (Fig. 1). In order to avoid thermal instabilities caused by a continuous ultrasonic wave, pulse supply was used with pulse duration 50 μs with repetition $10^4 s^{-1}$.

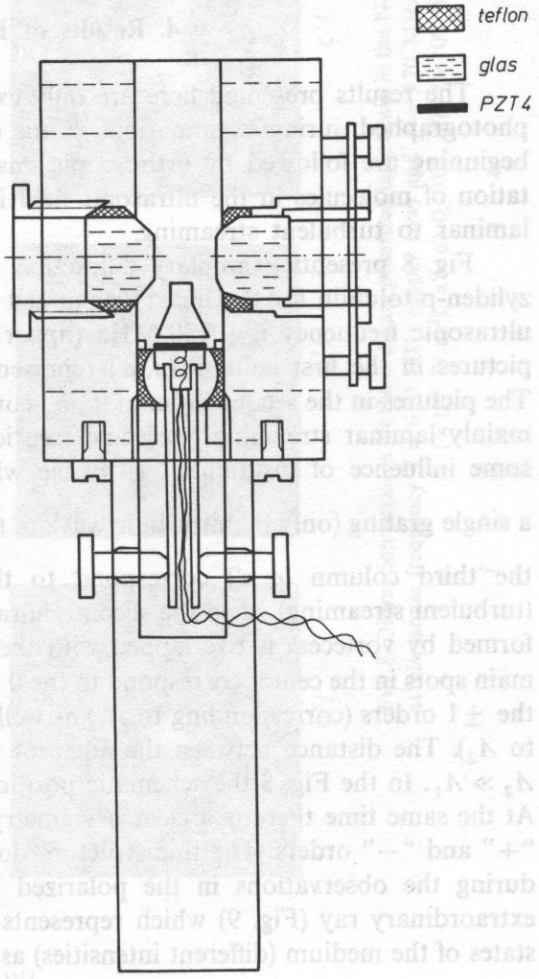


FIG. 7. Scheme of the experimental cell

The diffraction images were registered separately for the polarized light in the polarization plane of the "ordinary" and "extraordinary" ray. The observation of the diffraction images was carried out for liquid–crystalline samples: N–4hexyloxybenzyliden -p- toluidin, merck 4, merck 5 and W5 mixture for the ultrasonic wave with ultrasonic intensity from 0.0 to 1.0×10^4 W/m^2.

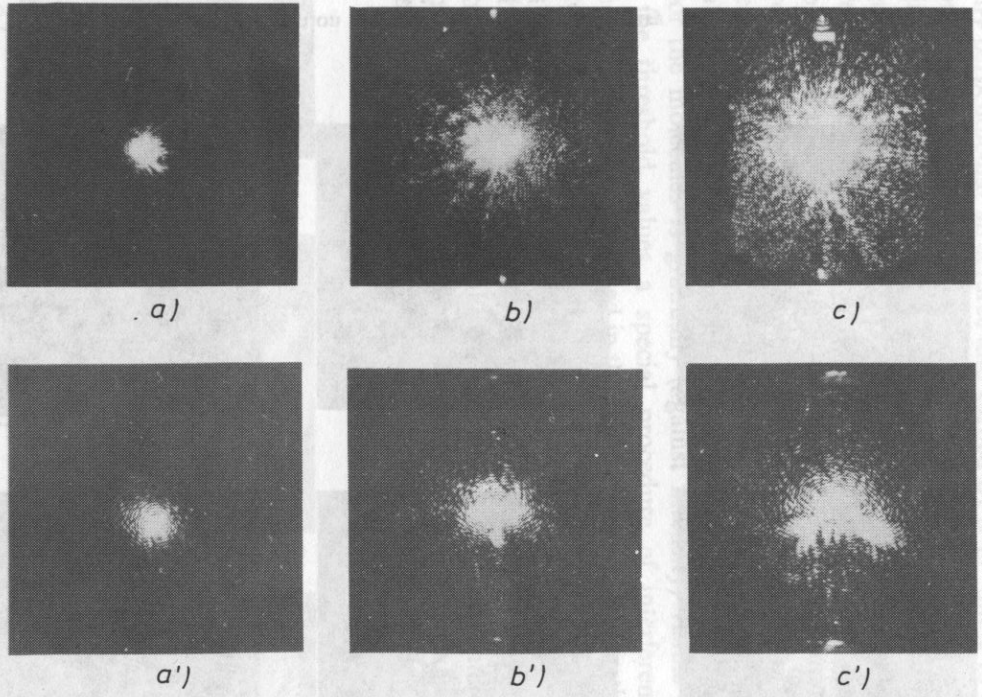
The intensity of the ultrasonic wave was estimated basing on the analysis of the equivalent circuit of the pistonlike radiating piezo-electric vibrator with regard to the shape of the applied energy concentrator and the sample thickness [28].

In order to find correlation between the observed diffraction effects and changes of the molecule arrangement of a nematics (which corresponds to the changes of the direction of the sample optical axis), orthoscopic investigations of the sample were made in parallel to diffraction investigations.

4. Results of Experiments

The results presented here are only examples of many pictures observed and photographed during examinations. Some characteristic diffraction patterns at the beginning are followed by orthoscopic images which enabled to determine orientation of molecules in the ultrasonic field including the threshold of passage from laminar to turbulent streaming.

Fig. 8 presents examplary diffraction patterns obtained for N-4hexyloxybenzyliden-p-toluidin for the lineary polarized light parallely to the ordinary beam at ultrasonic frequency $f = 6.70$ MHz (upper row) and 9.70 MHz (lower row). The pictures in the first column (a, a') represents the initial state without ultrasonics. The pictures in the second column (b, b') correspond to the state below the treshold; mainly laminar streaming, molecules reoriented of an angle ϕ occur; beside that some influence of instabilities is visible when diffraction of light takes place on a single grating (only by ultrasonic wave of the wave length $\Lambda_1 = \frac{c}{f}$). The pictures in the third column (c, c') correspond to the state above the intensity treshold (turbulent streaming) when the second diffraction grating (of the constant equal Λ_2) formed by vortecess is overlapped with the ultrasonic grating. In the patterns the main spots in the center correspond to the 0 − order of diffraction and the side spots the ± 1 orders (corresponding to Λ_1) as well as fine structure orders (corresponding to Λ_2). The distance between the adjacent fine structure fringes are small because $\Lambda_2 \gg \Lambda_1$. In the Fig. 8 the schematic positions of diffraction orders are drawn, too. At the same time there is a clear asymmetry (Fig. 8c) between the fine structure in "+" and "−" orders. The fine structure does not manifest itself in the ± 1 orders during the observations in the polarized light in the polarization plane of the extraordinary ray (Fig. 9) which represents patterns for the same frequencies and states of the medium (different intensities) as in Fig. 8 obtained for diffraction of the



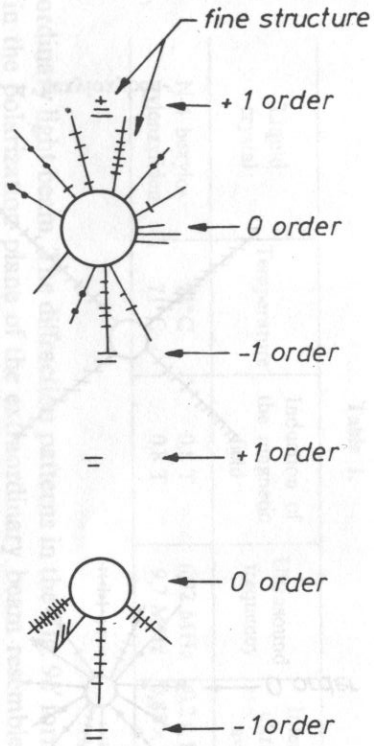


FIG. 8. Diffraction patterns for the ordinary polarized light beam in the N-4hexyloxybenzyliden-p-toluidin at ultrasonic frequency $f = 6.70$ MHz (upper row) and $f = 9.70$ MHz (lower row). a–c) 0, 0.02, 0.20 ($\times 10^4$ W/m^2); a′–c′) 0, 0.08, 0.74 ($\times 10^4$ W/m^2)

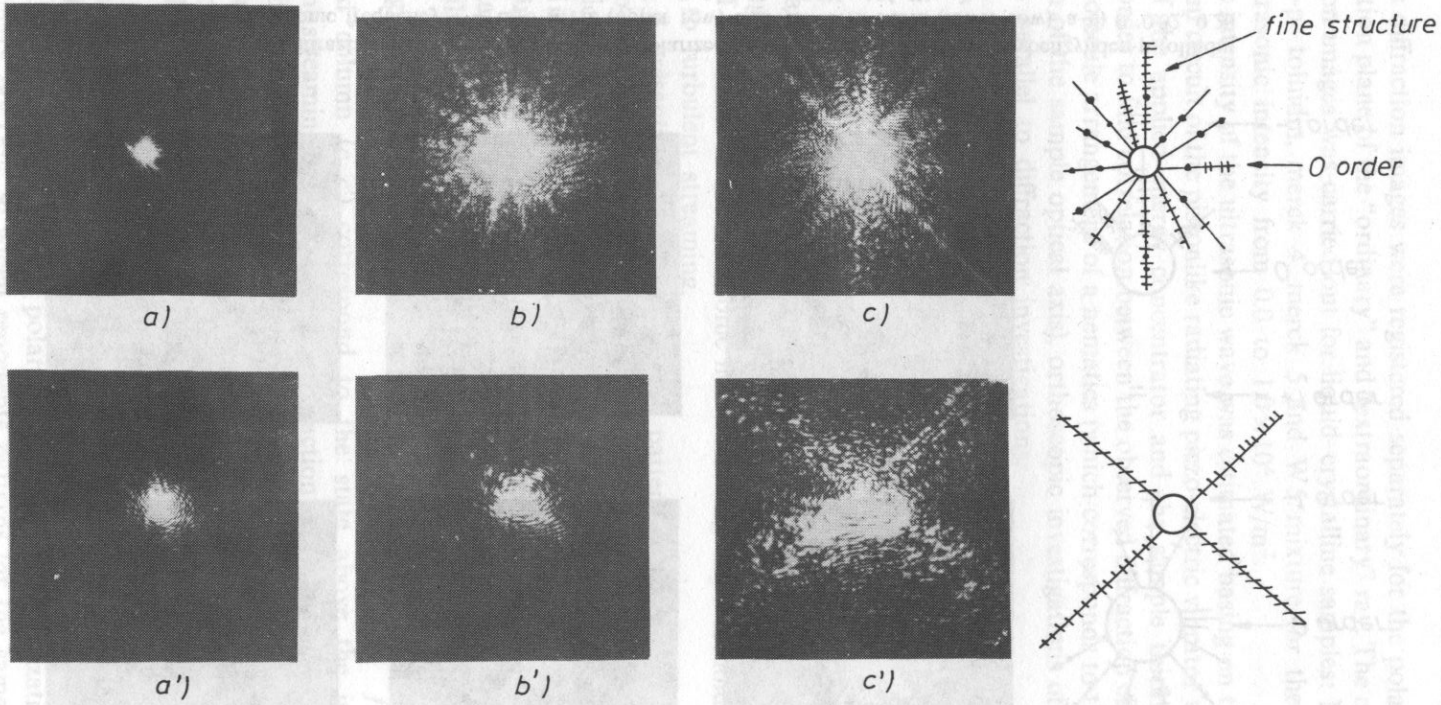


FIG. 9. Diffraction patterns for the extraordinary polarized light beam in the N−4 hexyloxybenzyliden-p-toluidin at ultrasonic frequency $f = 9.70$ MHz

Table 1.

Liquid crystal	Temperature	Induction of the magnetic field	Ultrasound frequency	Intensity of ultrasound average
N-4 hexylo--xylenzyliden	71°C 71°C	0,8 T 0,8 T	6,72 MHz 9,7 MHz	$0{,}2 \times 10^4$ W/m^2 $0{,}85 \times 10^4$ W/m^2

extraordinary light beam. The diffraction patterns in the Fig. 9c′ formed for polarized light in the polarization plane of the extraordinary beam resemble a diffraction on two mutually perpendicular structures and turned by a 45° angle to the direction of the ultrasonic wave [31].

The threshold values of the ultrasonic wave intensities at which one can observe a formation of the fine structure in the diffraction patterns of the investigated nematic are listed in the Table 1. The values where determined on the base of investigations in the orthoscopic system which enabled observing interference images resulting from the ordinary and extraordinary beams interaction. The examples of such interference fringes are shown in Fig. 10 (6.7 MHz) and Fig. 11 (9.7 MHz). The pictures were taken for different ultrasonic intensities. The influence of the turbulent streaming of the molecules is gradually (against intensity) visible (b. and c.). To determine the threshold values a special procedure of interference fringes was applied. The angles β and α presented in the Fig. 6 can be related to the inclination of the interference frings and the variation of the distance between them, respectively. The angles can be determined for different places of the interference image which correspond to local positions of the light beam cross-section in the sample. The coordinations of those positions taken for calculations are presented in the Fig. 12.

Measuring the inclinations of the interference fringes corresponding to the coordination net the local changes of the direction of the optical axis for the average

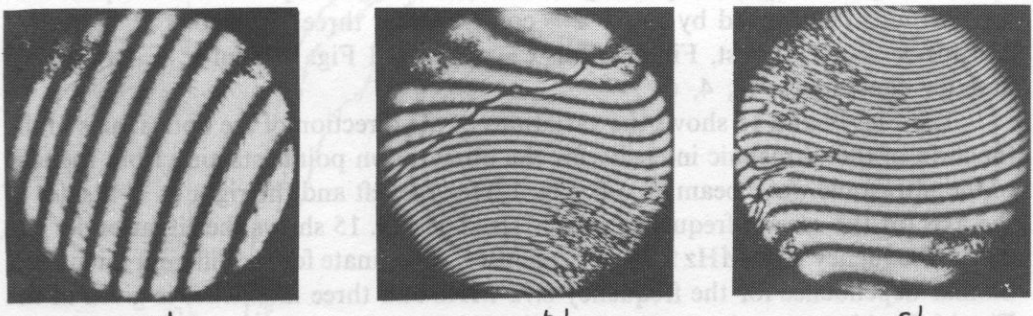


FIG. 10. Interference patterns for the N-4 hexyloxybenzyliden-p-toluidin at ultrasonic frequency $f = 6.70$ MHz, a) 0.18×10^4 W/m^2, b) 0.51×10^4 W/m^2, c) 0.74×10^4 W/m^2

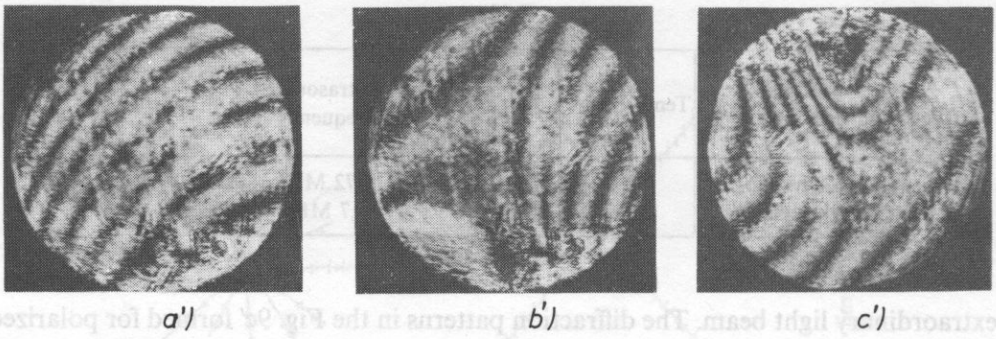


FIG. 11. Interference patterns for the N-4 hexyloxybenzyliden-p-toluidin at ultrasonic frequency $f = 9.70$ MHz. a') 0.16×10^4 W/m^2, b') 0.46×10^4 W/m^2, c') 0.72×10^4 W/m^2

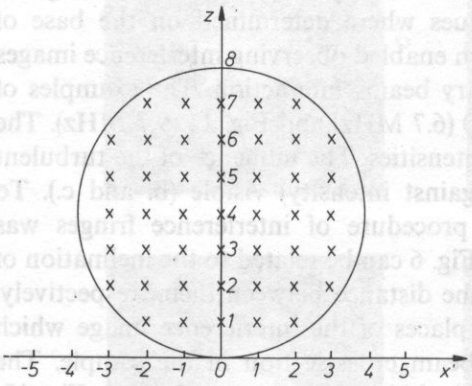
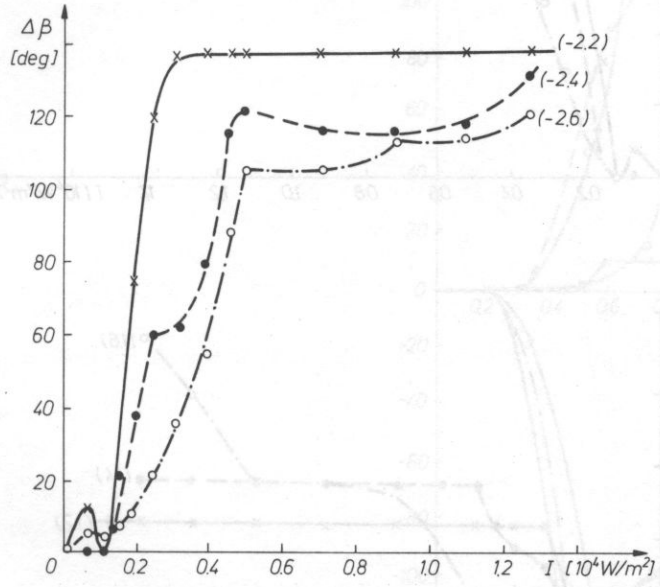


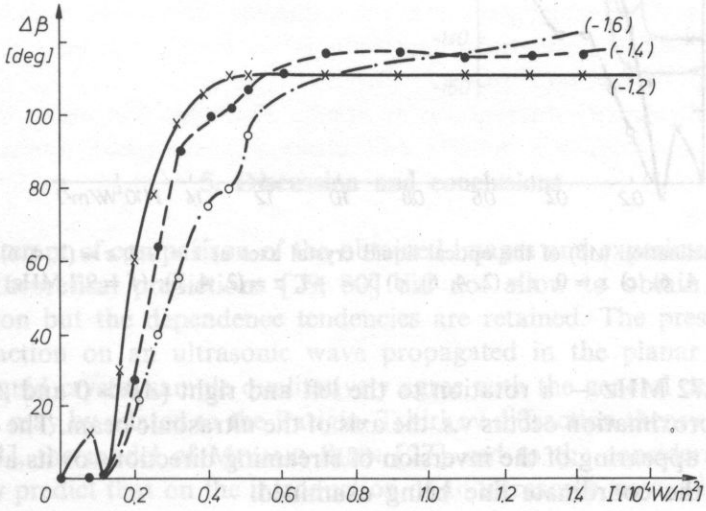
FIG. 12. Coordinat system for the liquid beam cross-section

thickness of the sample ($\Delta\beta$) were found in the function of ultrasonic wave intensity for chosen places of the sample (Fig. 13a-13d). The Fig. 13a presents the dependence for positions determined by $x = -2 = \text{const}$ and for three values of z (2, 4, 6), Fig. 13b for $x = -1 = \text{const}$, Fig. 13c for $x = \text{const}$ and Fig. 13d for $x = +1 = \text{const}$ and for the same z (2, 4, 6).

The Fig. 14 and 15 shows the variations of the direction of the optical axis in the function of the ultrasonic intensity for the observation points starting from the axis of the ultrasonic wave beam ($x = 0$, Fig. 12) for the left and the right ($x = 1$) sides of the axis for the case of frequency 6.72 MHz. The Fig. 15 shows the situation for the case of frequency 9.70 MHz for $z = 2$ against x coordinate for six different intensities. Similar dependence for the frequency 6.72 MHz and three intensities is given in the Fig. 16. In this case an interacting inversion of the optical axis at a distance from the ultrasonic beam axis is evidently manifested. It must be related to the change of the direction of the molecule streaming on the opposite one at the given place of the

cross–section of the sample. Such local change of the direction may confirm appearing of a vortex in the area and the inversion corresponds the passage through a izocline layer between two local streamings of opposite directions. A remarkable difference in the character of the local rotation of the optical axis of the nematic molecules with frequencies 9.70 and 6.72 MHz was observed. For frequency 9.70 MHz the rotation of the optical axis occurs only in one direction ($\Delta\beta > 0$) and





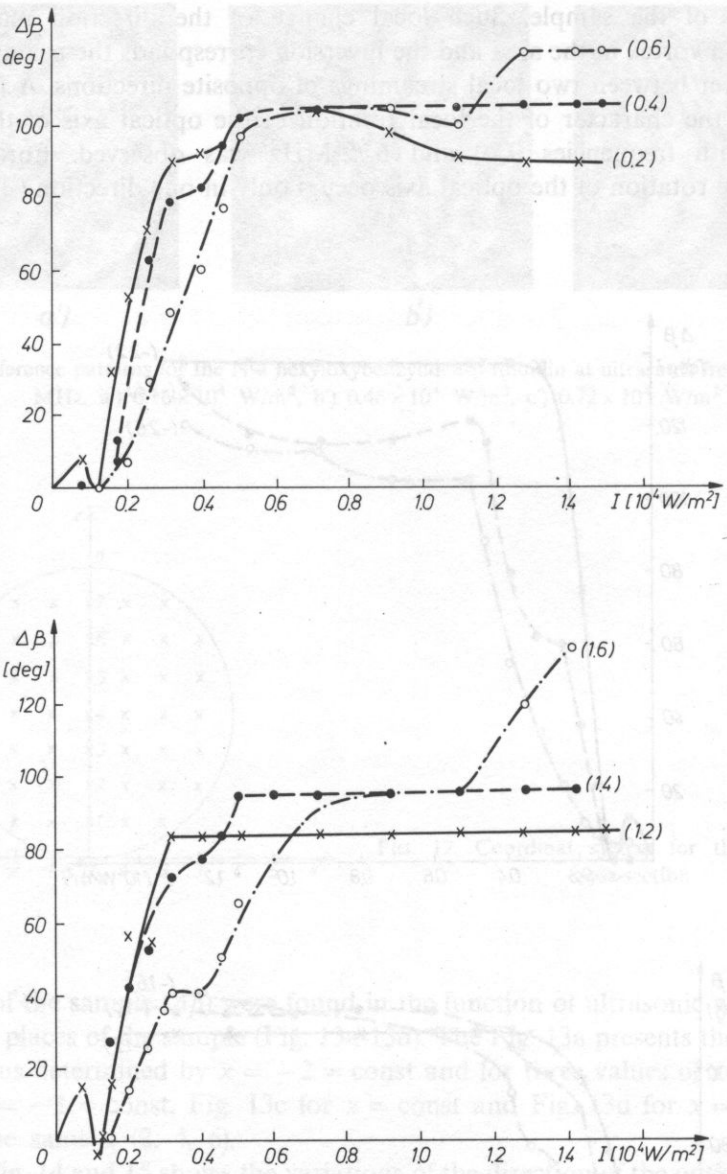


FIG. 13. Average inclination ($\Delta\beta$) of the optical liquid crystal axes a) $x = -2$, $z = (2, 4, 6)$ b) $x = -1$, $z = (2, 4, 6)$, c) $x = 0$, $z = (2, 4, 6)$, d) $x = +1$, $z = (2, 4, 6)$; ($f = 9.7$ MHz)

for frequency 6.72 MHz – a rotation to the left and right ($\Delta\beta > 0$ and $\Delta\beta < 0$) in symmetrical approximation occurs v.s. the axis of the ultrasonic beam. The difference is related to the appearing of the inversion of streaming directions or its absence on the way along the coordinate line being examined.

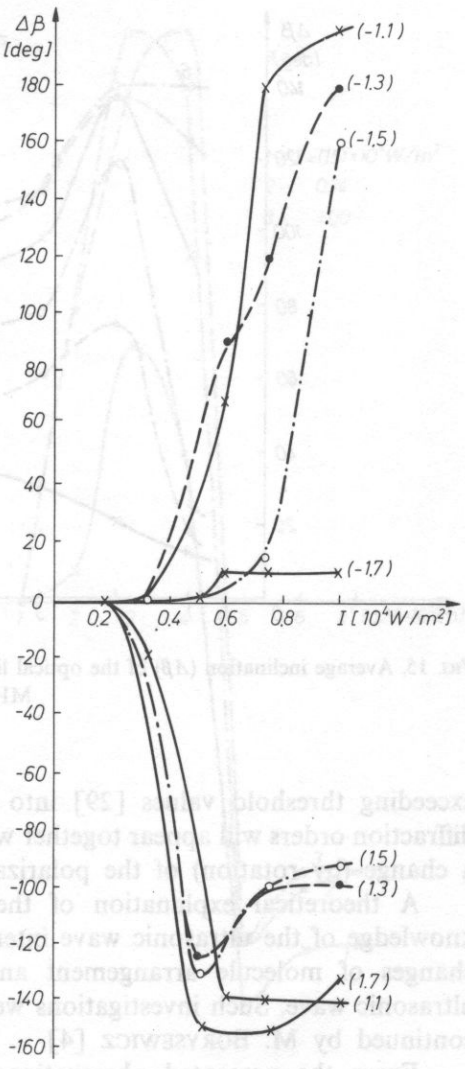


FIG. 14. Average inclination ($\Delta\beta$) of the optical liquid crystal axes for the fixed coordinats x, y ($f = 6.72$ MHz)

5. Discussion and conclusions

An attempt of comparison of the obtained images and experimental dependences with theoretical predictions [29, 30] did not allow to obtain a quantitative confirmation but the dependence tendencies are retained. The presented result of light diffraction on an ultrasonic wave propagated in the planar texture of the nematic liquid crystal sample qualitatively agree with the general description given above and may by related to the Parigin–Tshirkov diffraction theory for anisotropic bodies [13], the model of MIYANO–SHEN [27] and to the considerations given in [29]. They predict that on the introduction of an ultrasonic wave with the intensity

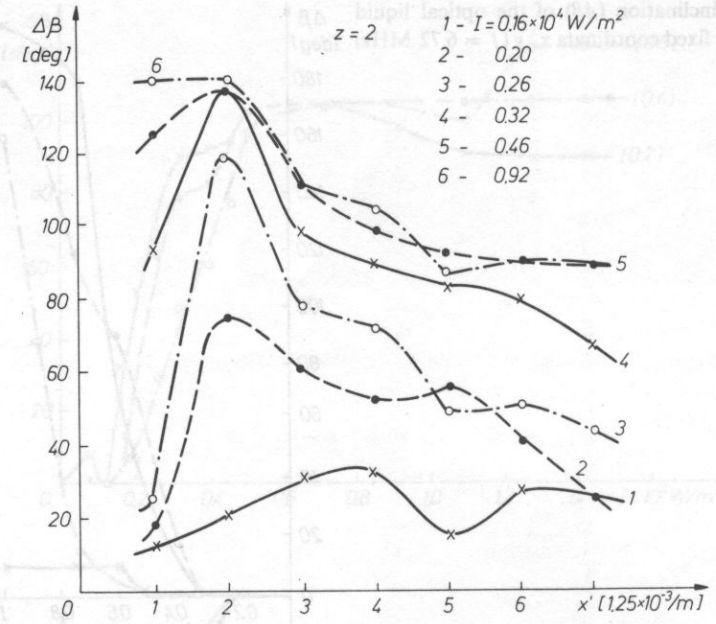


FIG. 15. Average inclination ($\Delta\beta$) of the optical liquid crystals axes in the x coordinat function (f = 9.7 MHz, z = 2)

exceeding threshold values [29] into the medium, changes of light intensity in diffraction orders will appear together with a change of ultrasonic wave intensity and a change (by rotation) of the polarization plane.

A theoretical explanation of the observed phenomena requires a precise knowledge of the ultrasonic wave interaction in a nematic sample, first of all, the changes of molecule arrangement and the molecule movement evoked by an ultrasonic wave. Such investigations were initiated by NAGAI and PETERS [17] and continued by M. BORYSEWICZ [4].

From the presented observations it results that the fine structure of the diffraction spectrum obtained in light and ultrasonic interaction appears at the moment of diversion from the laminar to turbulent streaming caused by acoustic radiation pressure.

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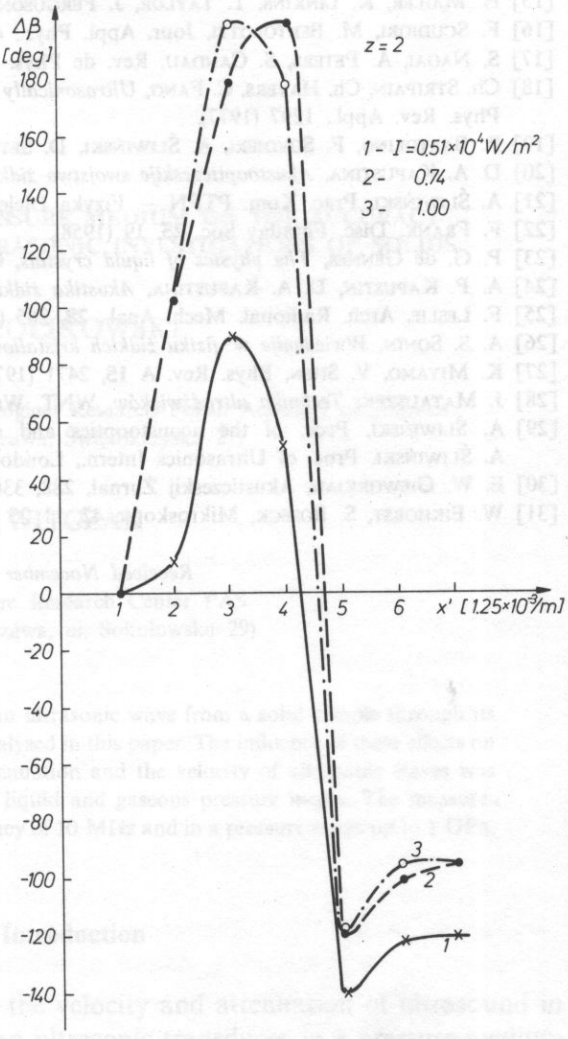

FIG. 16. Average inclination ($\Delta\beta$) of the optical liquid crystal axes in the x coordinat function (f = 6.72 MHz, z = 2)

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